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PART I: CLASSICAL LASER

PART II. THE EFFECT OF VELOCITY CHANGING.
COLLISIONS ON THE OUTPUT OF A GAS LASER

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N O T I C E

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Summary

Part I. Classical Laser

In this paper a completely classical model for laser action is discussed. An active medium consisting of classical anharmonic oscillators interacts with a classical electromagnetic field in a resonant cavity. Comparison with the case of a medium consisting of harmonic oscillators shows the significance of nonlinearities for producing self-sustained oscillations in the radiation field. The results for the classical model are found to be similar to those for a semiclassical model of the ammonia beam maser. The conclusion is that laser action is not intrinsically a quantum mechanical effect. The classical laser theory as given in this paper can also be applied to the case of the electron-cyclotron maser.

Part II. The Effect of Velocity-Changing Collisions on the Output of a Gas Laser.

A theoretical model for the pressure dependence of the intensity of a gas laser is presented in which only velocity-changing collisions with foreign gas atoms are included. This is a special case where the phase shifts are the same for the two atomic laser levels or are so small that deflections are the dominant effect of collisions. A collision model for hard sphere repulsive interactions is derived and the collision parameters - persistence of velocity and collision frequency - are assumed to be independent of velocity. The collision theory is applied to a third order expansion of the polarization in powers of the cavity electric field (weak signal theory). The resulting expression for the intensity shows strong pressure dependence. The collisions reduce the amount of saturation and the laser intensity increases with pressure in a characteristic fashion. It is recommended that the best way to look for this effect is to make the measurements under conditions of constant relative excitation. The velocity-changing collision theory is also applied to a high intensity laser theory. The results for the velocity dependence of the population inversion are evaluated in the rate equation approximation. The equations contain terms not considered by Smith and Hänsch in their work on the cross-relaxation effects in the saturation of the 6328 Å⁰ neon laser line.

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...all the knowledge on earth will give me nothing to assure me that this world is mine. You describe it to me and you teach me to classify it. You enumerate its laws and in my thirst for knowledge I admit that they are true. You take apart its mechanism and my hope increases. At the final stage you teach me that this wondrous and multi-colored universe can be reduced to the atom and that the atom itself can be reduced to the electron. All this is good and I wait for you to continue. But you tell me of an invisible planetary system in which electrons gravitate around a nucleus. You explain this world to me with an image. I realize then that you have been reduced to poetry: I shall never know.

Albert Camus, The Myth of Sisyphus

PART I: CLASSICAL LASER

I. INTRODUCTION

In recent years, there have been many advances in the theory of the laser. For a gas laser the active medium was treated as a quantum mechanical ensemble of two level atoms and the radiation as a classical electromagnetic field.¹ Scully and Lamb² have generalized this theory by treating both atoms and fields quantum mechanically. Other authors have given alternate formulations of this theory.³ Results of these calculations have been in good agreement with experiments, and except for possible refinements, the understanding of laser theory appears to be satisfactory.

There is, however, a fundamental question still to be considered. Is the operation of the laser a result of quantum effects (an avalanche of photons caused by stimulated emission⁴) or can the laser be described completely in classical terms? (Maxwell's equations for the field and Newton's equations of motion for the medium)

The laser is an example of a self-sustained oscillator. Such devices are well known in electronics. The first of these devices for which a theory was developed was the triode oscillator.⁵ In that case, the energy required for sustaining oscillations was provided by a battery. The nonlinear characteristics of the triode-battery system served to provide a negative nonlinear resistance which could drive an L-C circuit into a state of sustained oscillations.

In this paper, a totally classical model of a laser is investigated. The possibility of such a system was first discussed by Gapanov.⁶ The model here was independently suggested by one of the authors in a later publication.⁷ It is shown that the essential features of laser action arise from nonlinearities in the active medium and not from quantum effects. The calculation closely parallels the semiclassical theory of Ref. 1.

II. MODEL FOR CALCULATION

The model to be used is similar to the one used by Helmer⁸ and Lamb⁹ to describe the ammonia beam maser. An unpolarized beam of classical molecules passes through a resonant radiation cavity, and interacts with the radiation field. The induced polarization of the beam of molecules is calculated from the dynamics of the interaction. It is required that this polarization be the source for the radiation field. The equations for this self-consistency requirement will be introduced in the next section.

The following simplifying assumptions will also be used:

- (I) The mechanical oscillators move with a single, constant velocity through the cavity in a uniform one-dimensional beam perpendicular to the electric field.
- (II) Only one cavity mode is considered and the spatial variation of its electric field along the beam will be neglected. Loss in the cavity is described by a phenomenological Q-factor.
- (III) The mechanical oscillators are represented by a particle of mass m and charge e vibrating in one dimension parallel to the electric field in the cavity.
- (IV) Internal damping of the mechanical oscillator is neglected.
- (V) The mechanical oscillators enter the cavity with a fixed internal energy but with random phase with respect to the electric field.

The geometry of the model is shown in Figure 1.

III. SELF-CONSISTENCY CONDITIONS

The following discussion, based on Maxwell's equations, can be found in Ref. 1 in greater detail. Only one mode of a high Q electromagnetic resonator is considered. Let its frequency be Ω in the absence of an active medium. The electric field is taken in the form

$$(1) \quad E(z,t) = A(t) U(z)$$

where $U(z)$ satisfies the cavity mode eigenvalue problem. Maxwell's equations can be combined to give

$$(2) \quad \ddot{A} + \left(\frac{\sigma}{\epsilon_0}\right)\dot{A} + \Omega^2 A = -\left(\frac{1}{\epsilon_0}\right)\ddot{P}$$

P is the polarization of the medium and σ is a fictional conductivity adjusted to give the required damping of the radiation field in the cavity, i.e.,

$$(3) \quad \sigma = \frac{\epsilon_0 \nu}{Q}$$

Further assume that the electric field and polarization can be taken in the slowly varying amplitude and phase approximation

$$(4a) \quad A(t) = E(t) \cos(\nu t + \varphi(t))$$

$$(4b) \quad P(t) = C(t)[\cos[\nu t + \varphi(t)]] + S(t) \sin[\nu t + \varphi(t)]$$

where ν is a constant frequency yet to be determined. Inserting (4a) and (4b) into equation (2) and neglecting small terms in \ddot{E} , etc., the following self-consistency equations are obtained

$$(5a) \quad \dot{E} = -\frac{1}{2} \frac{\nu}{Q} E - \frac{1}{2} \left(\frac{\nu}{\epsilon_0}\right) S$$

$$(5b) \quad (\nu + \dot{\varphi} - \Omega)E = -\frac{1}{2} \left(\frac{\nu}{\epsilon_0}\right) C$$

IV. POLARIZATION OF THE MEDIUM

Let the internal motion of the mechanical oscillator in the presence of the cavity electric field be $x(t_0, \theta_0; t)$. The oscillator entered the cavity at $z = 0$ at time t_0 with phase θ_0 with respect to the electric field $A(t)$. The oscillators move with a constant single velocity so that they are at $z = v(t - t_0)$ at time t . The dipole moment p of each oscillator is

$$(6) \quad p(t_0, \theta_0; t) = ex(t_0, \theta_0; t)$$

The macroscopic polarization of the medium is obtained by summing up contributions of individual oscillators. For a collection of oscillators that entered the cavity with phases between θ_0 and $\theta_0 + d\theta_0$ around time t_0 , the contribution to the macroscopic polarization $dP(\theta_0; z, t)$ (dipole moment/unit length) is

$$(7) \quad dP(\theta_0; z, t) = N p(t_0, \theta_0; t) d\theta_0 / 2\pi$$

where N = the number of molecules/unit length in the cavity and the entry time t_0 replaced by

$$(8) \quad t_0 = t - (z/v)$$

Summing the contributions from all initial phases θ_0 gives

$$(9) \quad P(z,t) = (N/2\pi) \int_0^{2\pi} d\theta_0 p(t_0, \theta_0; t).$$

The component of the polarization which is the source of the cavity radiation is found by projecting P on the uniform cavity mode. Thus,

$$(10) \quad P(t) = (1/L) \int_0^L dz P(z,t)$$

where L is the length of the cavity.

V. LINEAR OSCILLATOR

The equation of motion of a linear oscillator of frequency ω in the presence of the assumed cavity field is

$$(11) \quad \ddot{x} + \omega^2 x = [eE/m] \cos(\nu t)$$

The phase of the electric field ϕ has been set = 0. The phase of the oscillator is then measured relative to that of the cavity field. The solution of (11) subject to the initial conditions $x(t_0) = \dot{x}(t_0) = 0$ is

$$(12) \quad x(t) = A_0 \cos(\omega(t-t_0) + \theta_0) + [eE/m] (\omega^2 - \nu^2)^{-1} \\ \times \left[\cos(\nu t) - \frac{(\omega + \nu)}{2\omega} \cos(\omega(t-t_0) + \nu t_0) \right. \\ \left. - \frac{(\omega - \nu)}{2\omega} \cos(\omega(t-t_0) - \nu t_0) \right]$$

From equation (8) the polarization of a collection of oscillators with initial phase θ_0 is

$$(13) \quad dP(\theta_0; z, t) = (N/2\pi)e [A_0 \cos((\omega - \nu)t - \omega t_0 + \theta_0) - [eE/m] \\ \times (\omega^2 - \nu^2)^{-1} (1 - \cos((\omega - \nu)z/\nu))] \cos(\nu t) \\ + (N/2\pi)e [-A_0 \sin((\omega - \nu)t - \omega t_0 + \theta_0) + [eE/m] \\ \times (\omega^2 - \nu^2)^{-1} \sin((\omega - \nu)z/\nu)] \sin(\nu t)$$

where non resonant terms have been neglected.

Equations (9) and (10) give

$$(14) \quad P(t) = [Ne^2 E/m] (\omega^2 - \nu^2)^{-1} \left[1 - \frac{\sin((\omega - \nu)T)}{T(\omega - \nu)} \right] \cos(\nu t) \\ + [2Ne^2 E/m] (\omega^2 - \nu^2)^{-1} \left[\frac{\sin^2((\omega - \nu)T/2)}{T(\omega - \nu)} \right] \sin(\nu t)$$

where

$$(15) \quad T = L/\nu$$

is the time spent by a molecule in the cavity. Comparing (14) with (4b), and letting ν be close to resonance, the coefficients C and S can be determined.

$$(16a) \quad C = [Ne^2 E/2m\nu] (\omega - \nu)^{-2} T^{-1} [T(\omega - \nu) - \sin((\omega - \nu)T)]$$

$$(16b) \quad S = [Ne^2 E/m\nu] \frac{(1/T) \sin^2((\omega - \nu)T/2)}{(\omega - \nu)^2}$$

The amplitude equation (5a) gives the following result for the cavity field

$$(17) \quad \dot{E}(t) = -\left[\frac{\nu}{2Q} + (Ne^2/\epsilon_0 m) \frac{\sin^2((\omega - \nu)T/2)}{T(\omega - \nu)^2} \right] E(t)$$

Equation (17) shows that an injected stream of randomly phased harmonic oscillators will always increase the damping of the field in the cavity. Steady state oscillations cannot be achieved with such a medium. The familiar result ¹⁰ that a randomly phased linear oscillator can only absorb energy from an electric field has been rederived.

If the phases of the oscillators before entering the cavity had been properly correlated to the electric field, S as calculated from (13) with a constant θ_0 could have been negative for suitable transit times T . That is equivalent to coupling a signal generator to the resonant cavity. The problem under consideration, however, is to construct a model for a generator.

In order to see more clearly why the harmonic oscillator will not sustain oscillations in the radiation field, evaluate (12) for $\dot{x}(t)$ at resonance ($\omega = \nu$)

$$(18) \quad \dot{x}(t) = A_0 \cos(\omega(t - t_0) + \theta_0) + [eE/2m\omega] (t - t_0) \sin(\omega t)$$

The power absorbed by the oscillator is

$$(19) \quad \frac{dW(t)}{dt} = F(t) \dot{x}(t) \quad \text{where } F(t) \text{ is the force on the oscillator}$$

Using (11), (19) becomes

$$(20) \quad \frac{dW(t)}{dt} = [\omega eEA_0/2] \sin(-\omega t_0 + \theta_0) + [(eE)^2/4m] (t-t_0)$$

where high frequency 2ω terms have been neglected. The oscillators that are initially phased to gain energy from the

electric field will do so for all times. The others initially lose energy, but eventually gain. The average of (20) over the injection phase θ_0 is positive definite which corresponds to the result derived earlier for the entire ensemble.

VI. NONLINEAR OSCILLATOR

The frequency of oscillation in the case of a nonlinear or anharmonic oscillator is amplitude dependent. Consider the situation where such an oscillator is introduced into the resonant cavity at an amplitude corresponding to a frequency slightly lower than the cavity frequency (See Figure 2). As in the case of the harmonic oscillator, upon entering the cavity some oscillators will gain energy from the field and some will lose, depending on the phase. As any oscillator gains energy it gradually goes out of resonance with the electric field in the cavity since the frequency is amplitude dependent. The energy absorption is thus severely limited in comparison with the linear oscillator.

Those oscillators that initially lose energy come closer to resonance with the driving field (and may even pass through resonance) and could lose a substantial amount of energy before rephasing or being removed from the cavity. Under certain conditions a net loss of energy to the cavity field is therefore possible.

This rough description gives some motivation for investigating a nonlinear oscillator as a medium for laser action.

The model for a classical, nonlinear oscillator will be the familiar but nontrivial case of a simple pendulum of mass m , length a , and charge e . The equation of motion is

$$(21) \quad \ddot{x} + a\omega^2 \sin(x/a) = [eE/m] \cos(\nu t)$$

where ω denotes the small amplitude resonant frequency.

Using the series expansion of $\sin(x/a)$ to third order, (21) becomes

$$(22) \quad \ddot{x} + \omega^2(x - \frac{1}{6}(x^3/a^2)) = [eE/m] \cos(\nu t)$$

which is known as Duffing's equation. There is extensive literature on the problem. There are subharmonic solutions, stable and unstable oscillations, and jump phenomena.¹¹ The following treatment corresponds most closely to that of Bogoliubov and Mitropolsky. Assume a solution with slowly varying amplitude and phase which can be expressed as a Fourier series in odd harmonics of the driving frequency ν . Let

$$(23) \quad x(t) = \sum_{n=0}^{\infty} B_{2n+1}(t) \cos[(2n+1)\nu t + \theta_{2n+1}(t)]$$

where the amplitudes B_{2n+1} and phases θ_{2n+1} are slowly varying in comparison with $\cos(\nu t)$. Only the component of the polarization varying at the fundamental frequency ν is of interest in this problem. Since numerical analysis has shown that the most significant elements of the motion vary at this frequency, (23) is replaced by

$$(24) \quad x(t) = B(t) \cos(\omega t + \theta(t))$$

Inserting (24) into (22), equating coefficients of $\cos(\omega t + \theta)$ and $\sin(\omega t + \theta)$ and neglecting terms in $\ddot{B}, \dot{B}^2, \ddot{\theta}, \dot{\theta}^2, \dot{B}\dot{\theta}$ (slowly varying amplitude and phase approximation) yields two coupled, first order differential equations for $B(t)$ and $\theta(t)$

$$(25a) \quad \dot{B} = -[eE/2m\omega] \sin \theta$$

$$(25b) \quad \dot{\theta} = \frac{(\omega^2 - v^2)}{2v} - \frac{\omega^2 B^2}{16va^2} - [eE/2m\omega] B^{-1} \cos \theta$$

Equations (25) can be rewritten in terms of a dimensionless force parameter

$$(26a) \quad G = [eE/2m\omega v]$$

and dimensionless variables

$$(26b) \quad A = B/a$$

for amplitude and

$$(26c) \quad \tau = \omega t$$

for time as

$$(27a) \quad dA/d\tau = -G \sin \theta$$

$$(27b) \quad d\theta/d\tau = \Delta - (\omega A^2/16\nu) - [G/A] \cos \theta$$

where

$$(28) \quad \Delta = (\omega^2 - \nu^2)/2\nu\omega.$$

When $G = 0$ the solutions of (27) are

$$(28a) \quad A(\tau) = A_0 = B_0/a$$

$$(28b) \quad \theta(\tau) = -\left(\frac{1}{16} A_0^2 \tau + \theta_0\right)$$

so that the motion of the oscillator is

$$(29) \quad x(t) = B_0 \cos\left[\omega\left(1 - \left(\frac{1}{16}\right)A_0^2\right)t + \theta_0\right]$$

The familiar $\left(\frac{1}{16}\right)A_0^2$ correction to the frequency of a simple pendulum is confirmed by this analysis.

Some of the properties of the solutions of equations (27) can be found by investigating the stationary points. These occur when $dA/d\tau = 0$ and $d\theta/d\tau = 0$ giving stationary solutions

$$(30a) \quad \theta = n\pi \quad \text{for } n = \pm 1, \pm 2, \pm 3, \dots$$

and A determined as a root of the cubic equation

$$(30b) \quad A[\Delta - \omega A^2/16\nu] - (-1)^n G = 0 \quad \text{for } n = \pm 1, \pm 2, \dots$$

Without loss of generality, consider only the solutions with $A > 0$. Figures 3a and 3b show the solutions of (31b) for $\Delta > 0$ and $\Delta < 0$ respectively. For $\Delta > 0$, Fig. 3a, there are three possible stationary points: a and b with $\theta = 0, 2\pi, \dots$, and c with $\theta = \pi, 3\pi, \dots$. By linearizing equations (27) about these points, a and c are found to give stable solutions and b to give an unstable solution. When $G > [4\Delta/3]^{3/2}$ (G_2 in Fig. 3a), a and b disappear leaving c as the only possible stationary solution. When $\Delta < 0$, Fig. 3b, only one stationary is found with the same stability as point c in Fig. 3a.

Equations (27) have been solved numerically on an IBM 7094 computer using a predictor-corrector method.¹² Figures 4a-4c exhibit the solutions in a phase diagram where $\theta(t)$ is plotted as a function of $A(t)$. The relationship between the amplitude and phase of the oscillator can be used to determine some important qualitative aspects of the motion under the influence of a driving field.

In Fig. (4a), $\Delta > 0$ and $G < [4\Delta/3]^{3/2}$. The stability properties of the stationary points a, b, c are easily seen. Figure (4b) corresponds to $\Delta > 0$ and $G > [4\Delta/3]^{3/2}$ while in Fig. (4c) $\Delta < 0$. In each of the latter two cases only the one stable point c is found.

Figures 5 show the time evolution of a collection of Duffing oscillators which enter the radiation cavity at a fixed amplitude, $A_0 = 0.32$, but with random phase. The amplitude A is an

indicator of the energy of the oscillator (i.e., energy = $\omega^2 a^2 [(\frac{1}{2})A^2 - (\frac{1}{24})A^4]$). The rough description of the nonlinear oscillator given at the beginning of this section can be made more explicit by examining Figs. 5. In Figure 5a, the oscillators have been in the cavity for a time $\omega t = 150$. The oscillators with initial phase greater than π are increasing in amplitude while those with initial phase less than π are decreasing in amplitude. By $\omega t = 900$, Fig. 5d, most of the oscillators have lost energy. Those oscillators that initially gained energy have "rephased" so as to lose energy. Those oscillators that initially lost energy have not yet returned to their original amplitude. If the oscillators are removed from the cavity at such a time, a net transfer of energy to the cavity radiation field can be expected. Therefore, a beam of nonlinear molecules injected with a suitable energy and removed from the cavity at the proper time could produce laser action.

The next section treats equations (25) to first order in the driving field. That analysis will find a threshold condition for the onset of laser oscillations and frequency pulling effects.

VII. WEAK SIGNAL THEORY

To first order in the force parameter G , the amplitude and phase of the Duffing oscillator are

$$(31a) \quad A = A^{(0)} + G A^{(1)}$$

$$(31b) \quad \theta = \theta^{(0)} + G \theta^{(1)}$$

Using (31) in the differential equations (27) gives

$$(32a) \quad dA^{(0)}/d\tau = 0$$

$$(32b) \quad dA^{(1)}/d\tau = -\sin \theta^{(0)}$$

$$(32c) \quad d\theta^{(0)}/d\tau = \Delta - \omega A^{(0)2}/16\nu$$

$$(32d) \quad d\theta^{(1)}/d\tau = \omega A^{(0)} A^{(1)}/8\nu - (1/A^{(0)}) \cos \theta^{(0)}$$

with solutions for $\omega \approx \nu$

$$(33a) \quad A^{(0)} = A_0 = \text{constant}$$

$$(33b) \quad \theta^{(0)} = (\mu - \nu)(t - t_0) + \theta_0$$

$$(33c) \quad A^{(1)} = [\omega/(\mu - \nu)] [\cos((\mu - \nu)(t - t_0) + \theta_0) - \cos \theta_0]$$

$$(33d) \quad \theta^{(1)} = \frac{\omega^3 A_0 \cos \theta_0}{8v(\mu-v)} (t-t_0) - \frac{\omega}{A_0(\mu-v)} \left[1 + \frac{\omega^2 A_0^2}{8v(\mu-v)} \right] \\ \times [\sin (\mu-v)(t-t_0) + \theta_0] - \sin \theta_0$$

where

$$(34) \quad \mu = \omega(1 - A_0^2/16)$$

is the free oscillation frequency of the injected oscillator.

Using equation (9) the polarization of a collection of oscillators with initial phase θ_0 is

$$(35) \quad dP(\theta_0; z, t) = [N/2\pi] \exp\left(t - \frac{z}{v}, \theta_0; t\right) d\theta_0 \\ = [Ne/2\pi] A [\cos \theta \cos vt - \sin \theta \sin vt] d\theta_0$$

Identifying the coefficients of $\sin vt$ and $\cos vt$ gives

$$(36a) \quad dC(\theta_0; z, t) = [Ne/2\pi] A \cos \theta d\theta_0$$

$$(36b) \quad dS(\theta_0; z, t) = -[Ne/2\pi] A \sin \theta d\theta_0$$

The first order contribution to C and S can now be found by using the solutions (33)

$$(37a) \quad dC^{(1)}(\theta_0; z, t) = [Ne/2\pi] \left[A_0 \cos \theta^{(0)} + G(A^{(1)} \cos \theta^{(0)} - A_0 \theta^{(1)} \sin \theta^{(0)}) \right] d\theta_0$$

$$(37b) \quad ds^{(1)}(\theta_0; z, t) = - [Ne/2\pi] [A_0 \sin \theta^{(0)} + G(A^{(1)} \sin \theta^{(0)} + A_0 \theta^{(1)} \cos \theta^{(0)})] d\theta_0$$

Averaging equations (37) according to the prescription of equations (9) and (10) gives

$$(38a) \quad c^{(1)} = [\omega Ne a G / (\mu - \nu)^2] [(\omega - \nu) - \frac{[(\omega - \mu) + (\omega - \nu)]}{T(\mu - \nu)} \sin((\mu - \nu)T) + (\omega - \mu) \cos((\mu - \nu)T)]$$

$$(38b) \quad s^{(1)} = [\omega Ne a G / (\mu - \nu)^2] [(\omega - \mu) \sin((\nu - \mu)T) - \frac{2[(\omega - \mu) + (\omega - \nu)]}{T(\nu - \mu)} \times \sin^2((\nu - \mu)T/2)]$$

In the limit of a linear oscillator, $\omega = \mu$, and equations (38) are identical to (16).

Using (38b) in the electric field amplitude equation (5a) the conditions necessary for the onset of laser oscillations can be determined. At steady state, $\dot{E} = 0$ and

$$(39) \quad 1/Q = \frac{Ne^2}{2m \nu \epsilon_0 T(\nu - \mu)^2} \left\{ \frac{4d}{(\nu - \mu)} \sin^2((\nu - \mu)T/2) - 2 \sin^2((\nu - \mu)T/2) - Td \sin((\nu - \mu)T) \right\}$$

where

$$(40) \quad d = (\omega - \mu) = A_0^2/16$$

is a measure of the initial excitation of the oscillator.

For a given cavity transit time $T = L/v$, the self-consistency condition (39) can be satisfied within finite frequency bands. If the R.H.S. of equation (39) is positive, then N and Q can be adjusted to give threshold. Figure 6 shows a plot of the bracketed expression on the R.H.S. of (39) as a function of

$$(41) \quad \psi = (\nu - \mu)T/2$$

for various values of the parameter Td . The domains where the R.H.S. of (39) is positive occur when

$$(42) \quad n\pi \geq \psi \geq \psi_n^+$$

$$-n\pi \geq \psi \geq \psi_n^- \quad \text{where } n = 1, 2, 3, \dots$$

The angles ψ_n^+ and ψ_n^- are solutions of the transcendental equation

$$(43) \quad \tan(\psi) = \frac{(Td)\psi}{(Td - \psi)}$$

For the remainder of this paper, only the region $\pi \geq \psi \geq \psi_1^+$

will be considered. This corresponds to the widest frequency band which gives a self-consistent solution and is closest to the linear resonance $\nu = \omega$ (frequency for small amplitude oscillations). The frequency band to be considered is then

$$(44) \quad \nu_{\min} \leq \nu \leq (2\pi/T) + \mu$$

$$\text{where } \nu_{\min} = \mu + 2\psi_1^+/T$$

Figure 7 shows a plot of the width of the above region as a function of transit time in the cavity. There is a linear variation for short transit times and $1/T$ dependence for long exposures. Figure 8 is a plot of ν_{\min} as a function of transit time. For this frequency band, ν_{\min} is always greater than μ . Shorter transit times require that the Duffing oscillators be sent through a cavity tuned to a higher frequency. As the transit time increases, the driving frequency must be proportionally decreased so that the oscillators do not begin to absorb energy.

Inserting (39a) into the "frequency determining" equation (5b) at steady state gives

$$(45) \quad (\nu - \Omega) = -(\nu/2\epsilon_0) [Ne^2/2m(\nu - \mu)^2] \left[(\omega - \nu) - \frac{(d + (\omega - \nu)) \sin((\nu - \mu)T)}{T(\nu - \mu)} + d \cos((\nu - \mu)T) \right]$$

Using N at threshold in (41) gives

$$(46) \quad (v - \Omega) = \frac{vT}{2Q} \frac{[(\omega - v) - \frac{(d + (\omega - v))}{T(v - \mu)} \sin((v - \mu)T) + d \cos((v - \mu)T)]}{[-2 \frac{(d + (\omega - v))}{T(v - \mu)} \sin^2((v - \mu)T/2) + Td \sin((v - \mu)T)]}$$

Figure 9a is a plot of the R.H.S. of (46) as a function of $(\mu - v)$ for $\omega T = 800$ while Figure (9b) has $\omega T = 200$. For short transit times (e.g. Figure 9b) the frequency is double valued. Thus it is possible to have two different types of oscillation under the single cavity mode. However, the analysis here is only of a single frequency. The equations of motion would have to be solved with a two frequency driving force in order to determine whether they could coexist. Therefore, the analysis will be restricted to longer transit times such at $\omega T = 800$ (Figure 9a). The frequency is then single valued and the pulling has a well defined linear region. To examine linear pulling, expand equation (42) about the zero point, $v = v_0$, giving

$$(47) \quad (v - \Omega) = -g(v_0 T)(v - v_0)$$

where

$$(48) \quad g = \frac{v_0 T}{Q} F(v_0 T) \quad \text{where } F \text{ is a complicated dimensionless function.}$$

S is known as the stabilization factor which apart from F is the ratio of the cavity bandwidth (ν/Q) to the transit time band width ($1/T$) of the molecules

VIII. COMPARISONS WITH AMMONIA BEAM MASER WEAK SIGNAL THEORY

It is instructive to compare the classical theory with one in which a simple, quantum mechanical, nonlinear oscillator is used. The results of the Helmer⁽⁷⁾-Lamb⁽⁸⁾ small signal theory of an ammonia beam maser will be used. The mechanical systems are two level atoms with energy difference $h\omega$ injected into the resonant cavity in their upper state. With the simplifying assumptions of section (II), equation (5a) at threshold gives

$$(49) \quad 1/Q = \frac{N P^2}{\hbar \epsilon_0} \frac{2}{(\nu - \omega)^2 T} \sin^2((\nu - \omega)T/2)$$

and the frequency equation (5b) becomes

$$(50) \quad (\nu - \Omega) = - \frac{N P^2}{2\hbar \epsilon_0} \frac{\nu}{(\nu - \omega)^2} \left[(\nu - \omega) - \frac{\sin((\nu - \omega)T)}{T} \right]$$

where P is the dipole matrix element for the radiative transition. The similarity between equation (39) and (49) and (45) and (50) should be noted.¹³ The second \sin^2 term in the expression for the Duffing oscillator (39) is always negative. That term is exactly the same as the total expression for the linear oscillator, Eq. (16). The other two terms in (39) combine to make the expression positive under certain conditions. They are both proportional to $d^2(\omega - \mu)$ which is a measure of the non-linearity of the oscillator.

IX. STRONG SIGNAL THEORY

It has been seen that, at least for small signals, a completely classical system provides reasonable model for laser action. An unpolarized beam of anharmonic oscillators of fairly high amplitude is injected into a radiation cavity and the conditions for the buildup of laser oscillations are not very different from those of a simple quantum mechanical model.

The nonlinearities of molecular medium play an essential role in that they provide for a coupling between the amplitude and phase of the mechanical oscillator in the presence of an electric field. This coupling, not present in the linear oscillator, allows the phases to readjust giving the medium a net active polarization.

The next problem is to determine the intensity and frequency of the classical laser. Ideally, the perturbation expansion in the dimensionless parameter G could be continued to high orders. Unfortunately, the amount of algebra involved is enormous. Using a computer, however, it was relatively easy to use numerical methods to calculate the polarization of an ensemble of Duffing oscillators.

The technique employed was to solve equations (27) simultaneously with the same initial amplitude A_0 for a set of equally spaced initial phases between 0 and 2π . The phase averages, equation (9), of S and C were found using Simpson's rule at each time. Since the molecules move at uniform velocity, the

mode projection of equation (10) is just the time average. In terms of the numerical procedure this time average is just the cumulative sum for previous times divided by the total elapsed time. For small amplitudes, these coarse phase and time averages are in excellent agreement with the first order theory.

Figure (10) is a plot of $-S$ as a function of G for various values of $\Delta = (\omega - \nu)/\omega$ with $\omega T = 800$. Figure (11) shows a plot of $-C$ as a function of G . The amplitude equation at steady state ($\dot{E} = 0$) gives

$$(51) \quad E/Q = [G/Q][2m\omega\nu a/e] = -(1/\epsilon_0)S(G)$$

The intersection of a straight line through the origin in Fig. 10 with slope $(1/Q)[2m\omega\nu a/e]$ with any one of the $-S$ curves will give an operating point of the laser. Figure 12 is a plot of a set of such intersection points showing E^2 as a function of $\nu - \mu$ for several values of Q . Thus, the theory has given the intensity as a function of $\nu - \mu$.

Figure 10 shows the characteristic behavior of saturation phenomena: $-S$ increases linearly for small amplitudes and then curves back downward for larger values of the electric field. The gain ($-S$) becomes negative at very high amplitudes.

From the values of the electric field obtained for the operating points and the numerical values of $C(E)$, the frequency of the laser can be determined. Figure (13) is a plot of $C(E)/E$

as a function of $(\nu - \mu)$ for different values of Q . The form $C^{(1)}(E)/E$ of Fig. 9a is also included to show the pulling is apparently linear.

The next section will show that the classical laser theory can be applied to a physical problem of the electron-cyclotron maser.

X. ELECTRON-CYCLOTRON MASER

The electron-cyclotron maser¹⁴ is an example of a real physical system for which the classical laser theory is applicable. This oscillator uses a system of energetic free electrons in a dc magnetic field (H_z) field which undergo radiative transitions in a microwave cavity. In quantum mechanical terms, the transitions are induced between adjacent Landau levels w_n where

$$(52) \quad w_n = mc^2 \left[(1 + 2(n+1/2)h\omega/mc^2)^{1/2} - 1 \right]$$

with

$$(53) \quad \omega = eH_z/mc \quad (\text{cyclotron frequency})$$

For slightly relativistic electrons ($\sim 5\text{Kev}$) and for typical laboratory magnetic fields ($H_z \sim 2000$ gauss) the relevant quantum numbers are of the order of 10^8 (i.e., $10^8 h\omega = 5\text{Kev}$). A classical treatment of this problem should be used for such high quantum numbers.

Consider electrons moving in a uniform magnetic field H_z in a rectangular microwave cavity. Assume a TE mode in the cavity with the dc magnetic field H_z perpendicular to the electric field. Neglect the transverse spatial variations of the cavity mode and the rf magnetic fields. Also, assume that

most of the electronic energy is in its transverse motion (i.e., $\dot{x}, \dot{y} \gg \dot{z}$)

The equations of motion of an electron with charge e and mass m injected into a cavity according to the above scheme are

$$(54) \quad \frac{d}{dt} [\gamma m \dot{x}] - \frac{eH_z}{c} \dot{y} = eE_x$$

$$(55) \quad \frac{d}{dt} [\gamma m \dot{y}] + \frac{eH_z}{c} \dot{x} = eE_y$$

where

$$(56) \quad \gamma = [1 - (\dot{x}^2 + \dot{y}^2)/c^2]^{-1/2}$$

As in the case of the Duffing oscillator, let $E_x = E \cos vt$ and let $E_y = 0$. Integrating (55) gives

$$(57) \quad \dot{y} = \frac{-eH_z x}{\gamma mc}$$

Substituting (57) into (54) gives

$$(58) \quad \frac{d}{dt} [\gamma m \dot{x}] + \frac{e^2 H_z^2 x}{\gamma mc^2} = eE \cos(vt)$$

Assume the following solutions for $x(t)$ and $y(t)$ for single mode operation

$$(59) \quad x(t) = r(t) \cos(vt + \theta(t))$$

$$y(t) = -r(t) \sin(vt + \theta(t))$$

where $r(t)$ is the radius of the orbit of the electron and $r(t)$ and $\theta(t)$ are taken in the slowly varying amplitude and phase approximation. Neglecting terms in \dot{r}^2 , \ddot{r} , $\ddot{r}\theta$, $\ddot{\theta}$, and for slightly relativistic electrons.

$$(60) \quad \gamma \cong 1 + \frac{r^2 v^2}{2c^2}$$

$$(61) \quad \dot{\gamma} \cong \frac{\dot{r} r v^2}{c^2}$$

Using (59), (60), and (61) in (58) the following first order differential equations for $r(t)$ and $\theta(t)$ are obtained

$$(62) \quad \dot{r} = -G \left(1 - \frac{r^2 v^2}{c^2}\right) \sin \theta$$

$$(63) \quad \dot{\theta} = \frac{\omega^2 - v^2}{2v} - \frac{r^2 v (\omega^2 + v^2)}{4c^2} - Gr^{-1} \cos \theta$$

where $G = [eE/2mv]$

Since $r^2 v^2 / c^2$ is small compared to unity in (57), (62) and (63)

are identical to the equations (25) for A and θ in the Duffing problem.

$$(64) \quad \dot{r} = -G \sin \theta$$

$$(65) \quad \dot{\theta} = \frac{\omega^2 - v^2}{2v} - \frac{r^2 v (\omega^2 + v^2)}{4c^2} - Gr^{-1} \cos \theta$$

The electron-cyclotron maser can therefore be treated using the theory given in the last section.

XI. SUMMARY

A totally classical model of a laser has been treated, in which no mention has been made of photons or stimulated emission. A beam of randomly phased classical anharmonic oscillators passes through a resonant cavity and gives up energy to the radiation field. Nonlinearities in the medium are essential for producing self-sustained oscillations. A medium consisting of randomly phased harmonic oscillators (linear medium) can only absorb energy from the radiation field.

This model has been used to calculate the intensity and frequency of the resulting laser. The theory can be applied to Hirschfield's electron cyclotron maser since extremely high quantum numbers are involved.

Footnotes

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12. Equations (27a) and (27b) were originally solved numerically by M. Sargent III and some of his results are shown in Ref. 7.

We gladly acknowledge his assistance on the numerical part of this problem and his sharing of some of the difficulties.

13. The coefficient on the r.h.s. of Eq. (49) $\frac{Np^2}{\hbar}$

has the dimensions of $\frac{[\text{number density}] \times [\text{dipole moment}]^2}{[\text{angular momentum}]}$

In Eq. (39) for the classical model, the coefficient on the r.h.s. can be written in the form $\frac{N(ea)^2}{2m(va)a}$ where "a" is the length of

the pendulum, "ea" is the dipole moment, and $m(va)a$ is the angular momentum.

14. The possibility of a "cyclotron maser" was first suggested by J. Schneider, Phys. Rev. Letters, 2, 504 (1959) and first realized by J.L. Hirshfield and J.M. Wachtel, Phys. Rev. Letters, 12, 533 (1964). For a more complete discussion see J.L. Hirshfield, I.B. Bernstein, and J.M. Wachtel, J.Quant. Elect., QE-1, 237, (1965).

Figure Captions

1. Geometry of the classical laser. A one dimensional beam of molecules moves through a resonant radiation cavity with velocity v . The direction of internal oscillation of each oscillator (x axis) is parallel to the electric field.
2. Anharmonic oscillator potential. The frequency of oscillation is amplitude dependent. The oscillators are injected into the cavity with amplitude A_0 and corresponding frequency $\mu = \mu(A_0)$. This gives an energy slightly higher than if they were oscillating at the cavity electric field frequency $\nu > \mu$. Depending on the initial phase, some oscillators gain energy from the field and move away from resonance while others lose energy and move toward resonance with the electric field.
- 3a. Plot of $A(\Delta - \frac{\omega A^2}{16\nu})$ for $\Delta > 0$. The intersection of this curve with horizontal straight lines of ordinates $|G_1| < (4\Delta/3)^{3/2}$ give stationary points a, b, c. The intersection with $-G_2$, $|G_2| > (4\Delta/3)^{3/2}$ gives only stationary point c.
- 3b. Plot of $A(\Delta - \frac{\omega A^2}{16\nu})$ for $\Delta < 0$. The intersection with horizontal line of ordinate $-G_1$ gives only one stationary point c.
- 4a. Duffing phase plot with $G = 10^{-4}$ and $\Delta = 4.625 \times 10^{-3}$. Solutions of Eqs. (27) where θ is plotted as a function of A with $\Delta > 0$ and $G < (4\Delta/3)^{3/2}$. The stationary points a and c are stable while point b is unstable.

4b. Duffing phase plot with $G = 6.0 \times 10^{-4}$ and $\Delta = 4.625 \times 10^{-3}$. Solution of Eqs. (27) where θ is plotted as a function of A for $\Delta > 0$ and $G > (4\Delta/3)^{3/2}$. The only stationary point is c.

4c. Duffing phase plot for $G = 10^{-4}$ and $\Delta = -10^{-3}$. Solution of Eqs. (27) where θ is plotted as a function of A for $\Delta < 0$. The only stationary point is c.

5. Time evolution of Duffing oscillators. Solutions of Eqs. (27). θ (modulo 2π) is plotted as functions of A stopped at times $\omega\tau = 0, 300, 600, 900, 1200, 1500$. Fifteen oscillators start at equally spaced initial phases between 0 and 2π with amplitude $A_0 = 0.32$ and $G = 4.0 \times 10^{-4}$ and $\Delta = 3 \times 10^{-3}$.

6. Plots of $y = 2 \frac{(Td - \psi) \sin^2 \psi}{\psi} - (Td) \sin 2\psi$

where $\psi = (\nu - \mu)T/2$ for $Td = 0, 1, 2$. The parameter $d = \omega - \mu$ of Eq. (40) is a measure of the injection energy of the oscillators. The ranges where y is positive give self-consistent solutions of the threshold condition equation (41).

7. The dimensionless width of the first frequency band of laser oscillations $(\nu_{\max} - \nu_{\min})/\omega$ is plotted as a function of the dimensionless transit time ωT in the cavity.

8. The minimum frequency of laser oscillations ν_{\min} is plotted as a function of the dimensionless transit time for the first band. For large T , $\nu_{\min} \sim \mu$.

9a. Frequency pulling. A plot of Eq. (46) as a function of ν for $\omega T = 800$. Intersection with the straight line $(\nu - \Omega)$ gives the operating laser frequency. The quantity $(\nu/2Q)(C^{(1)}/S^{(1)})$ is an abbreviated form of the r.h.s. of Eq. (46).

- 9b. Frequency pulling. A plot of Eq. (46) as a function of ν for $\omega T = 200$. In this case the laser frequency is double valued. The stability properties of these oscillations are yet to be determined.
10. $-S$ plotted as a function of G for strong signals for various values of $\Delta' = 10^3 \Delta$ with $\omega T = 800$. Intersection of the curves with the straight line $-S = (2m\omega\nu a/e)(G/Q)$ will give the laser operating points as a function of Δ .
11. $-C$ plotted as a function of G for strong signals for various values of $\Delta' = 10^3 \Delta$ for $\omega T = 800$.
12. Laser intensity I as a function of $(\nu - \mu)$ for various values of Q .
13. Plots of $C(E)/E$ as a function of ν for strong signals and various values of Q . Intersection with the straight line $(\nu - \Omega)$ gives the laser frequency. The dashed curve shows $C(E)/E$ of Eq. (46) in the first order theory for comparison.

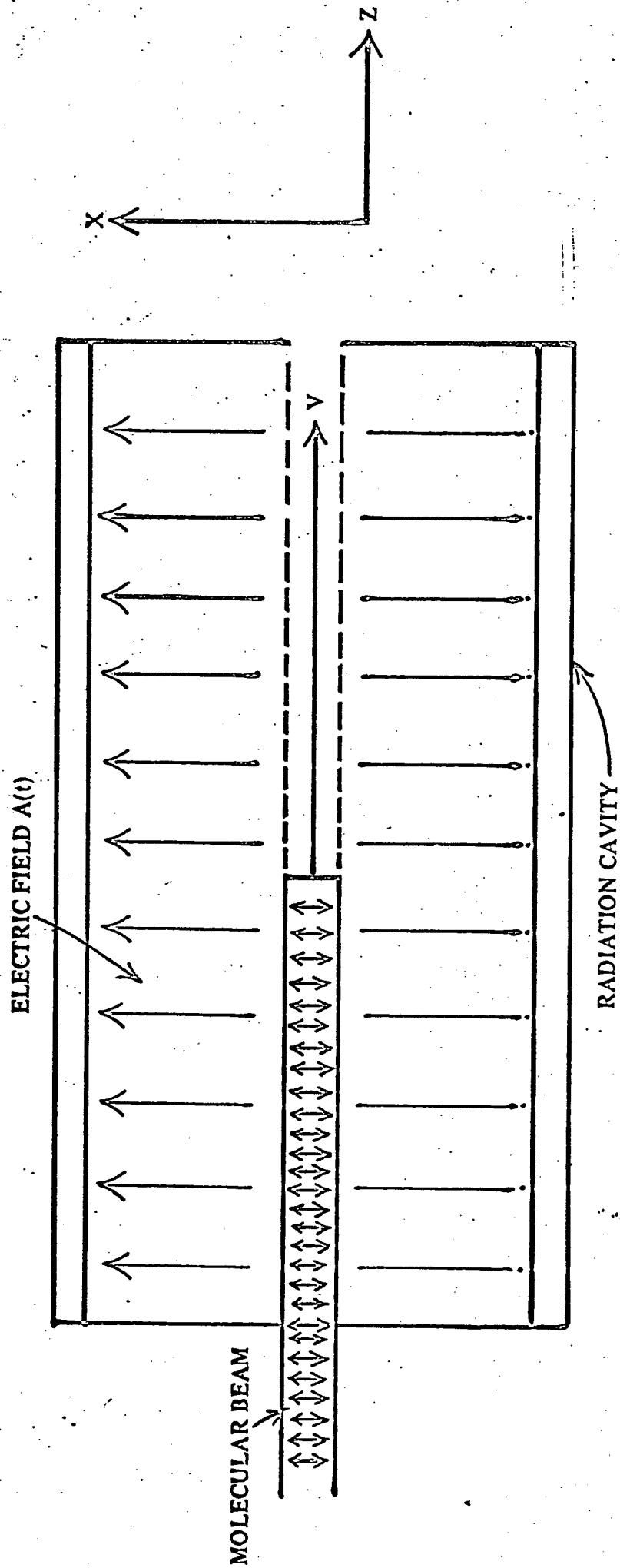


Figure 1

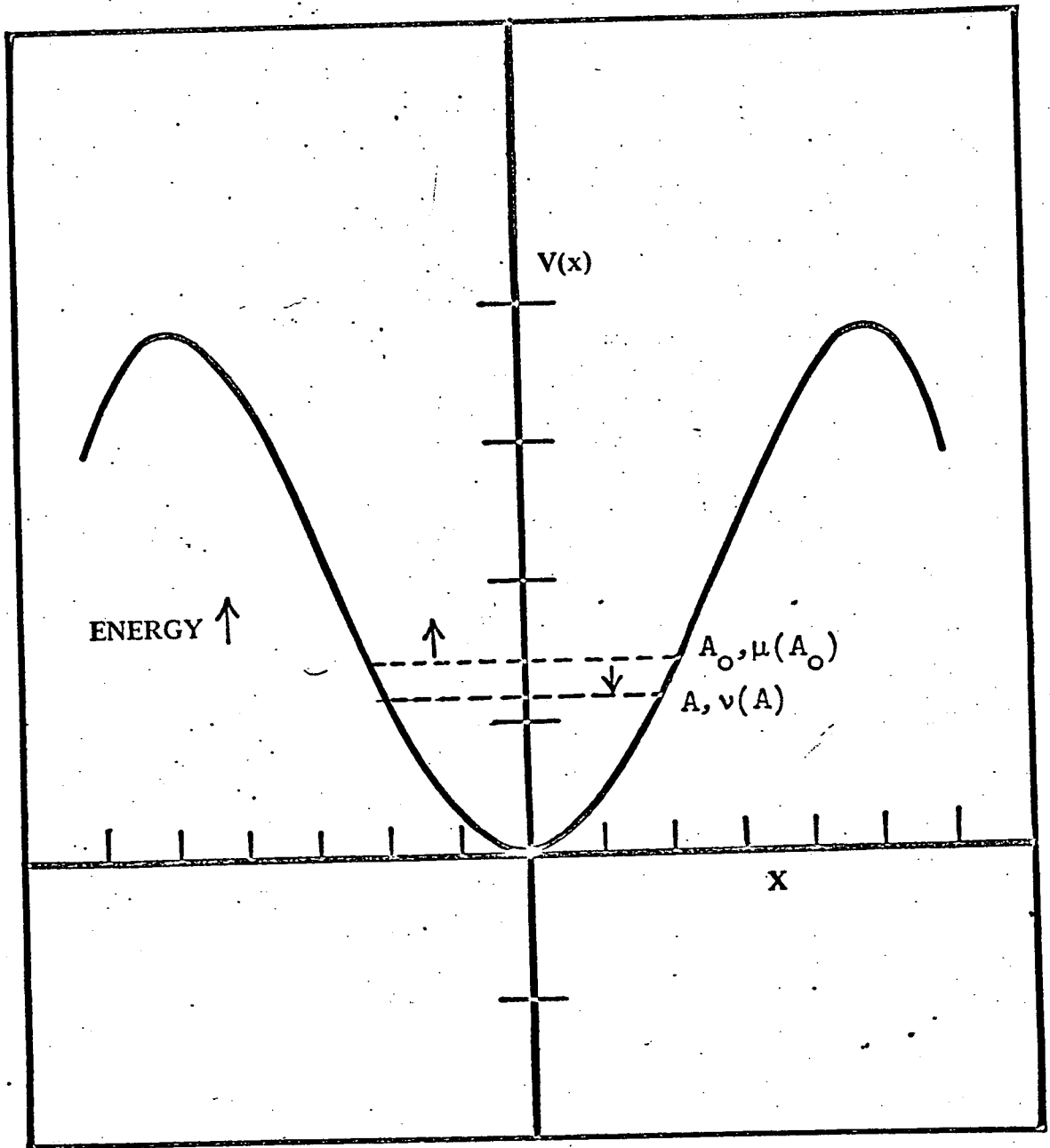
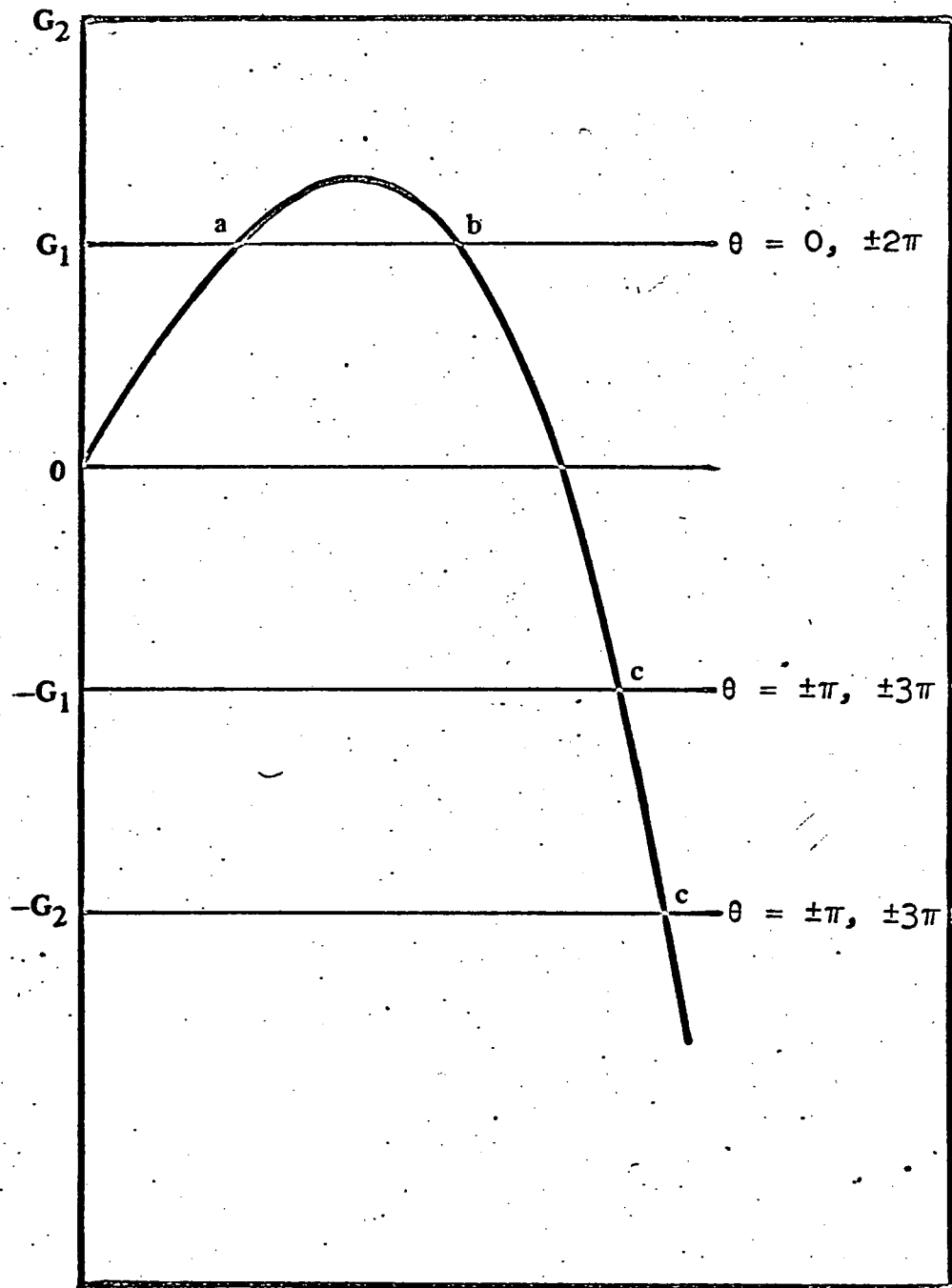


Figure 2



A

Figure 3a

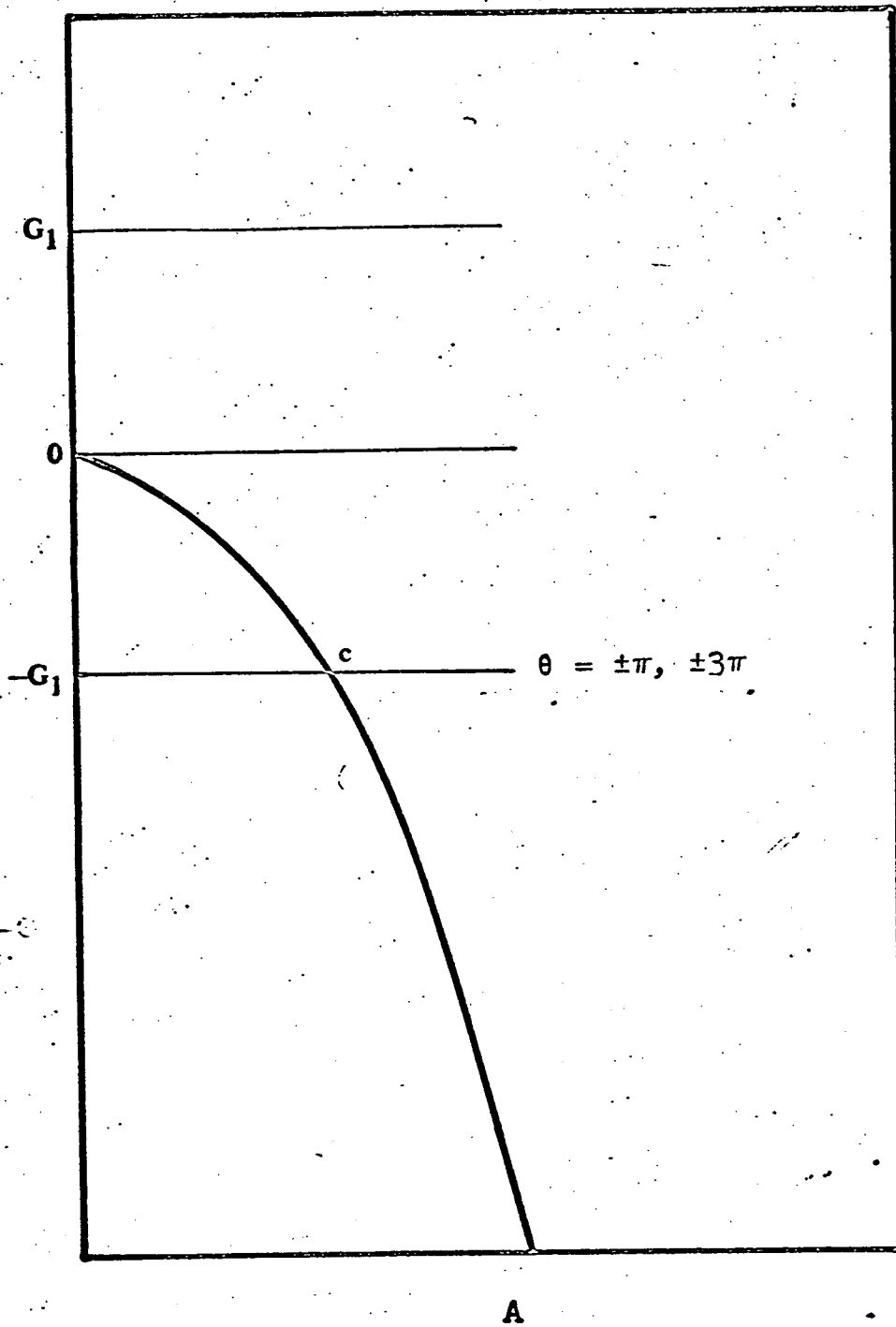


Figure 3b

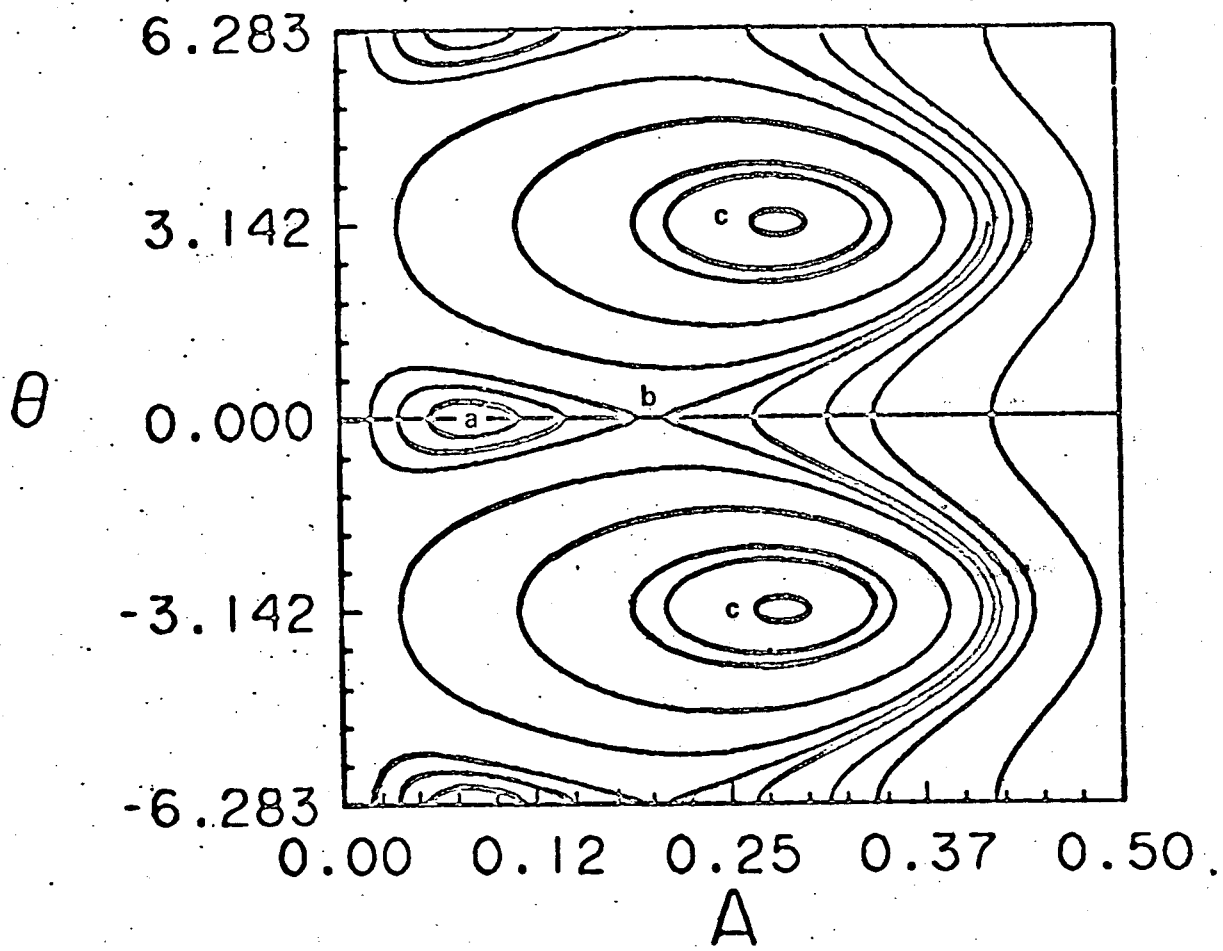


Figure 4a

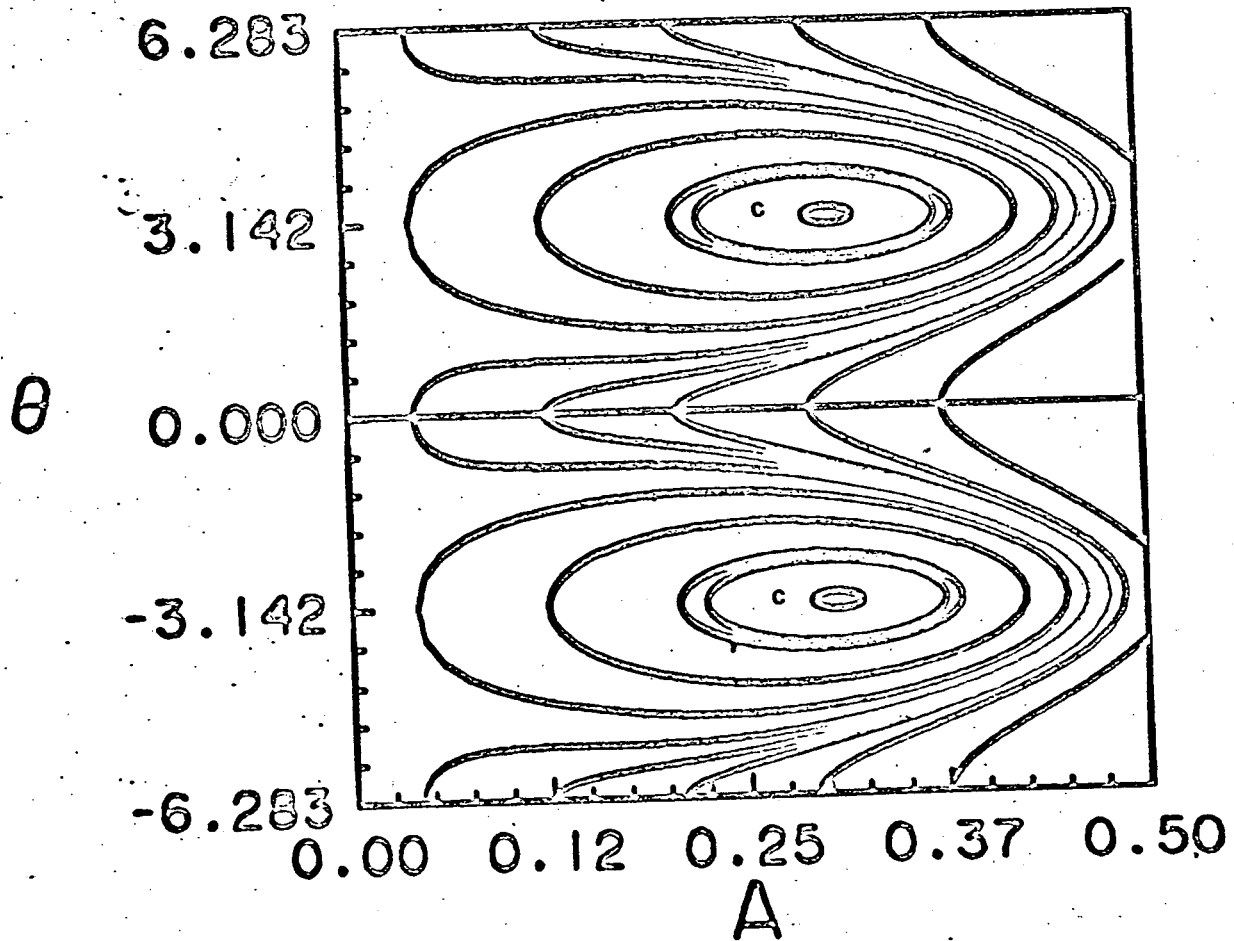


Figure 4b

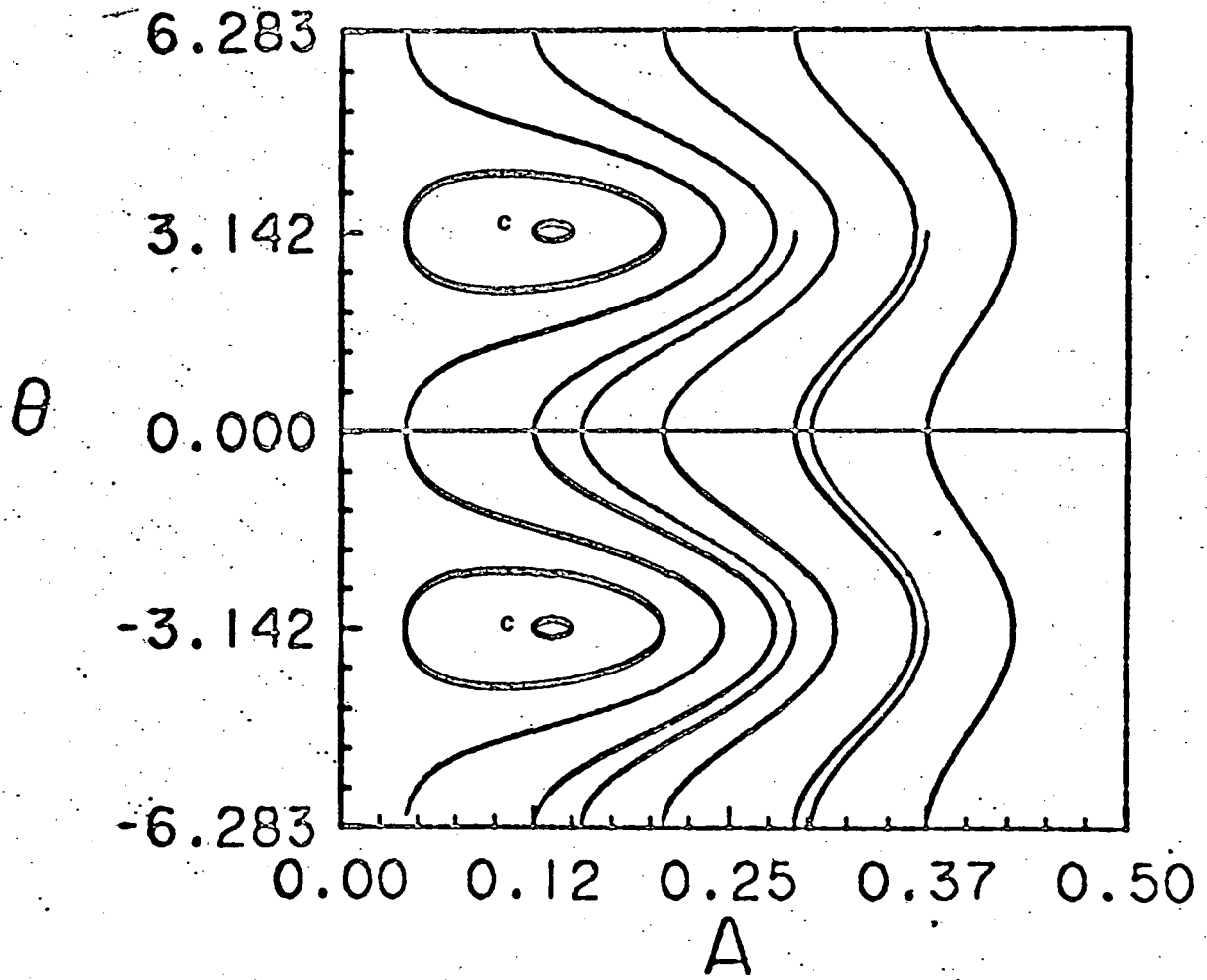


Figure 4c

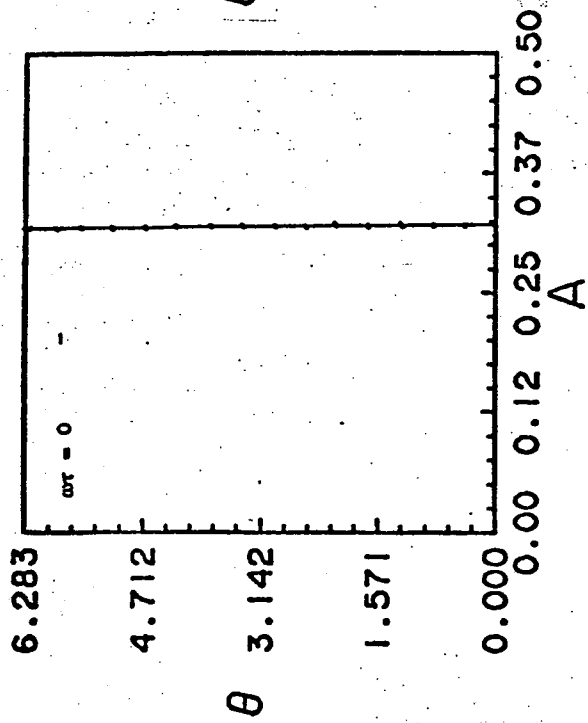


Figure 5

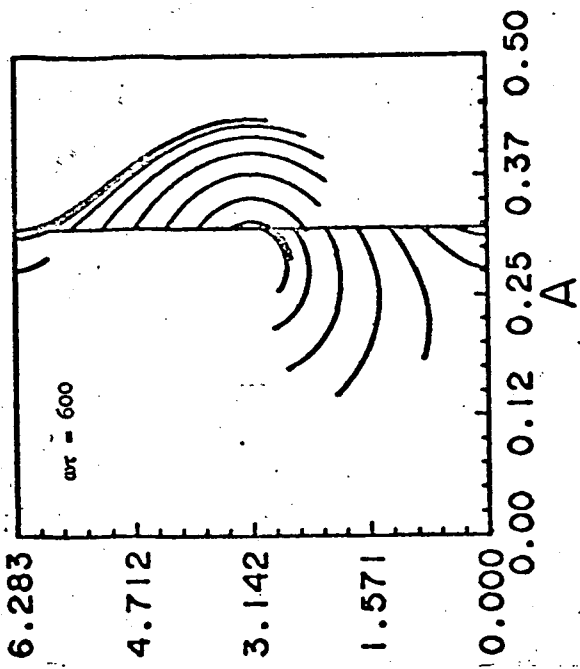
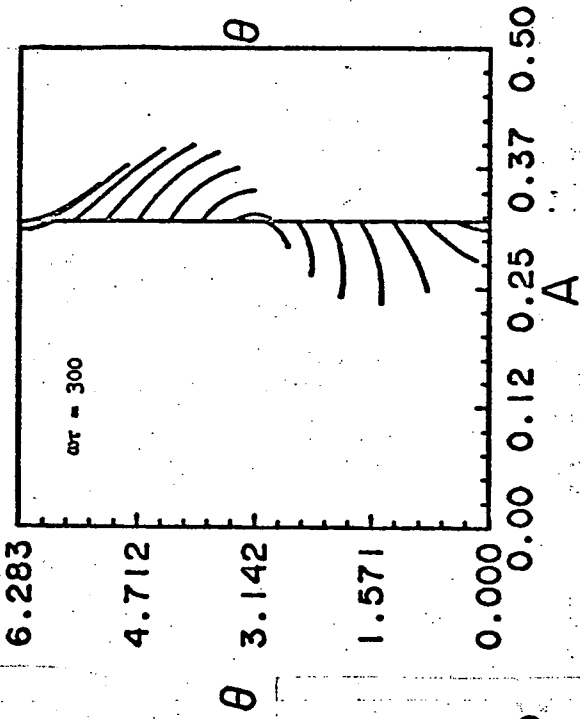
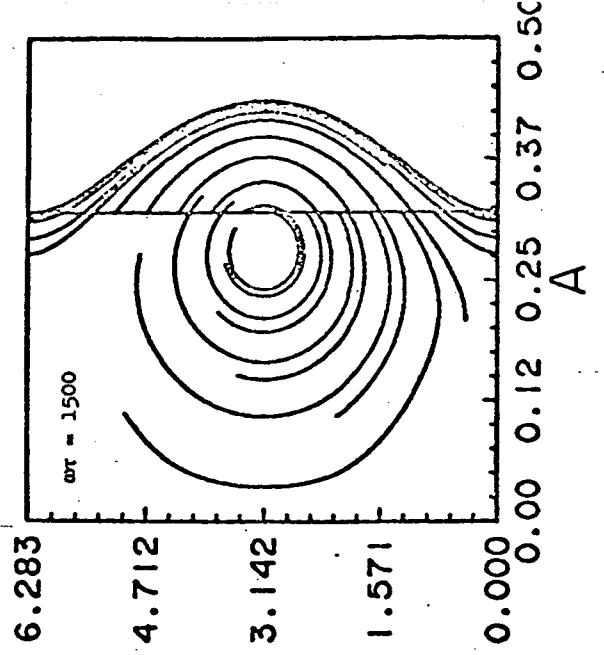
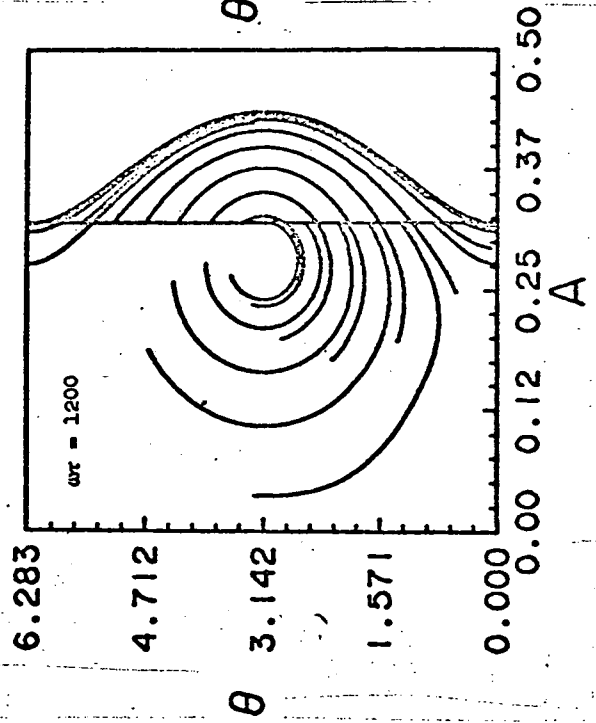
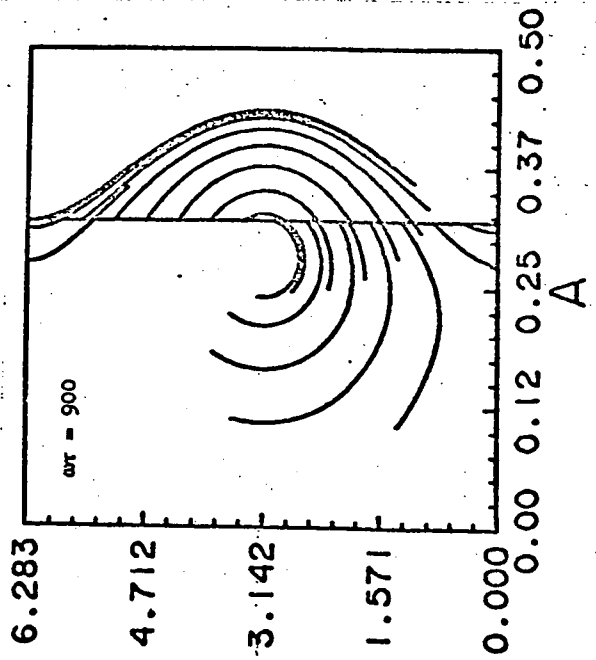


Figure 5



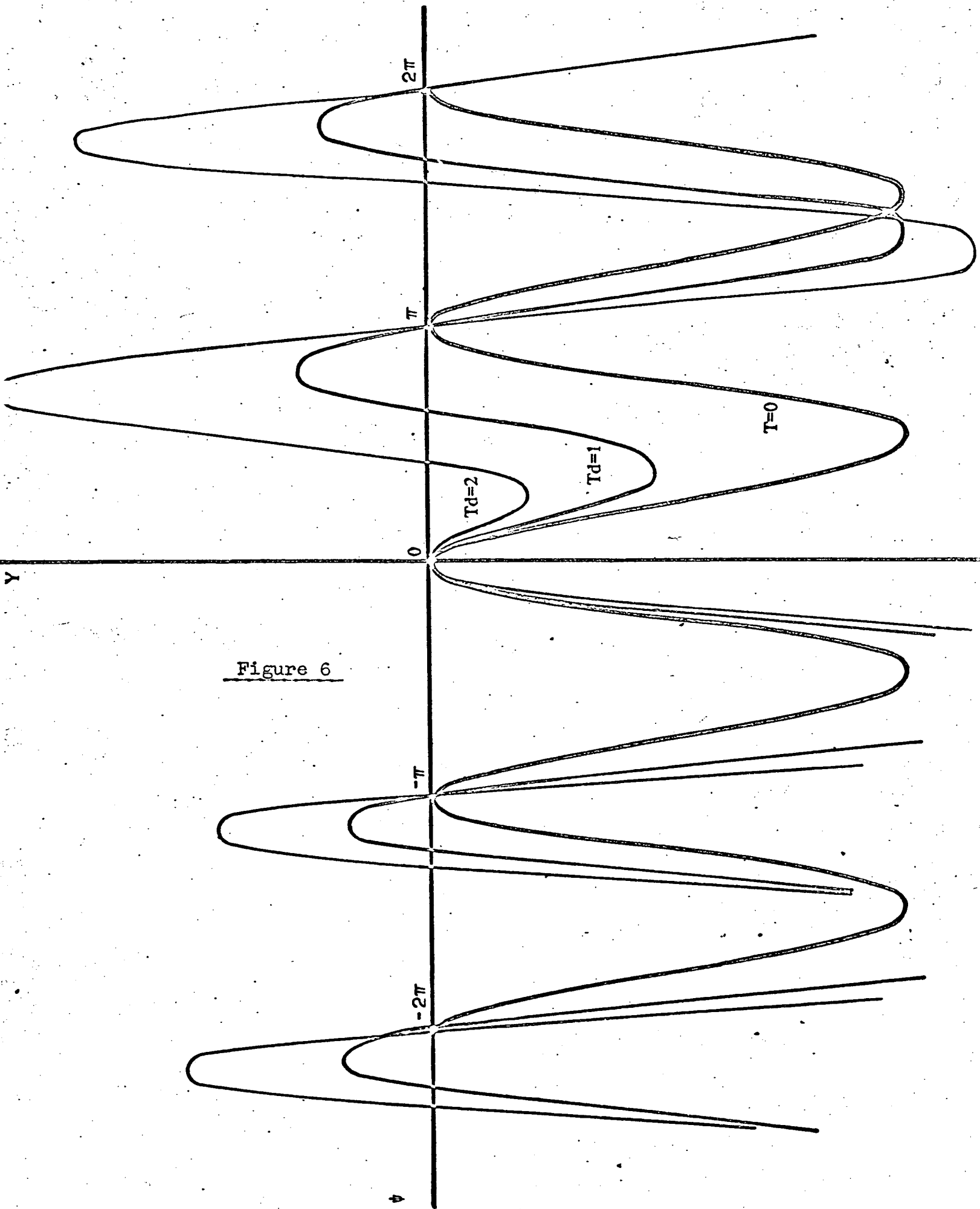


Figure 6

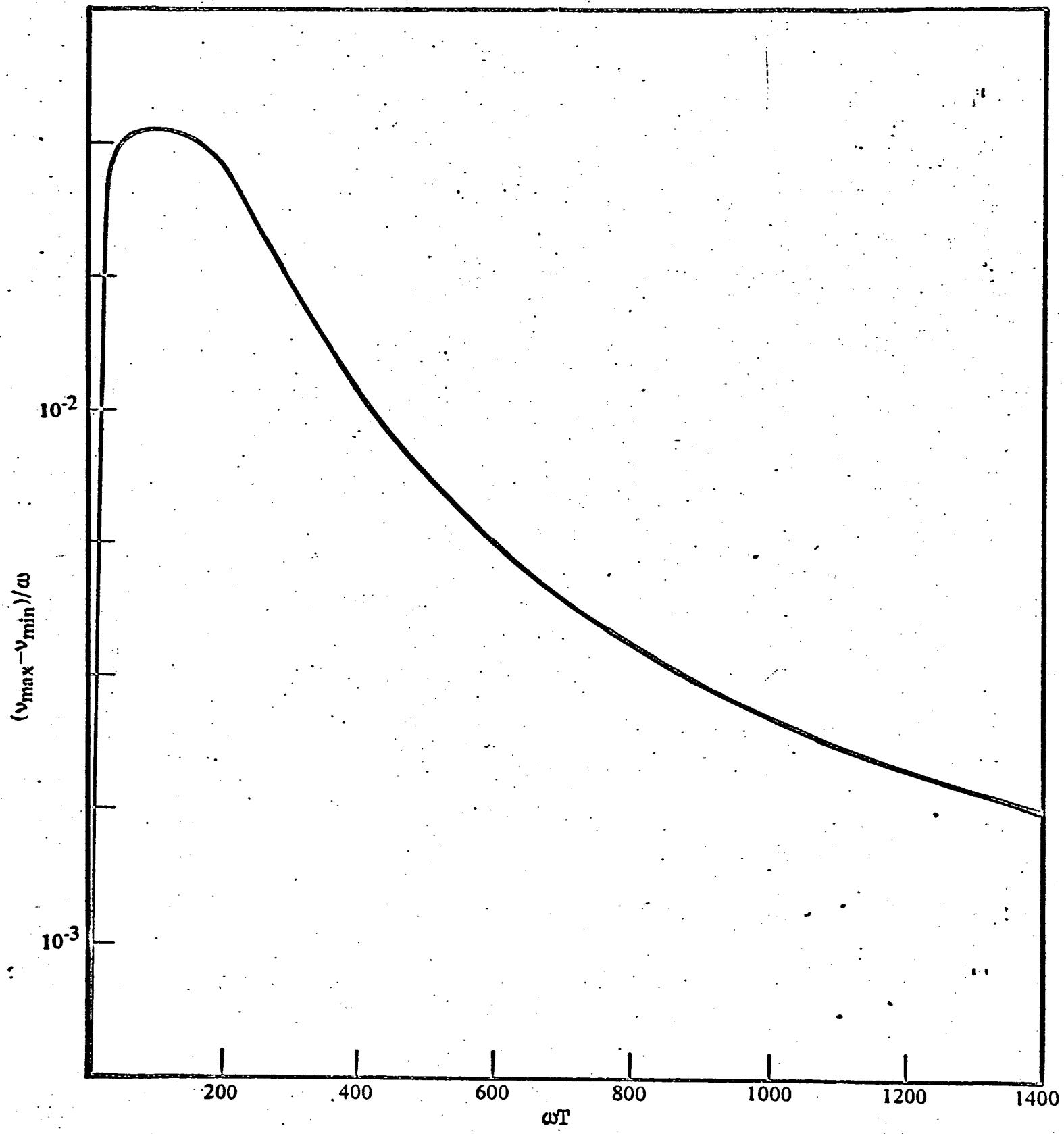
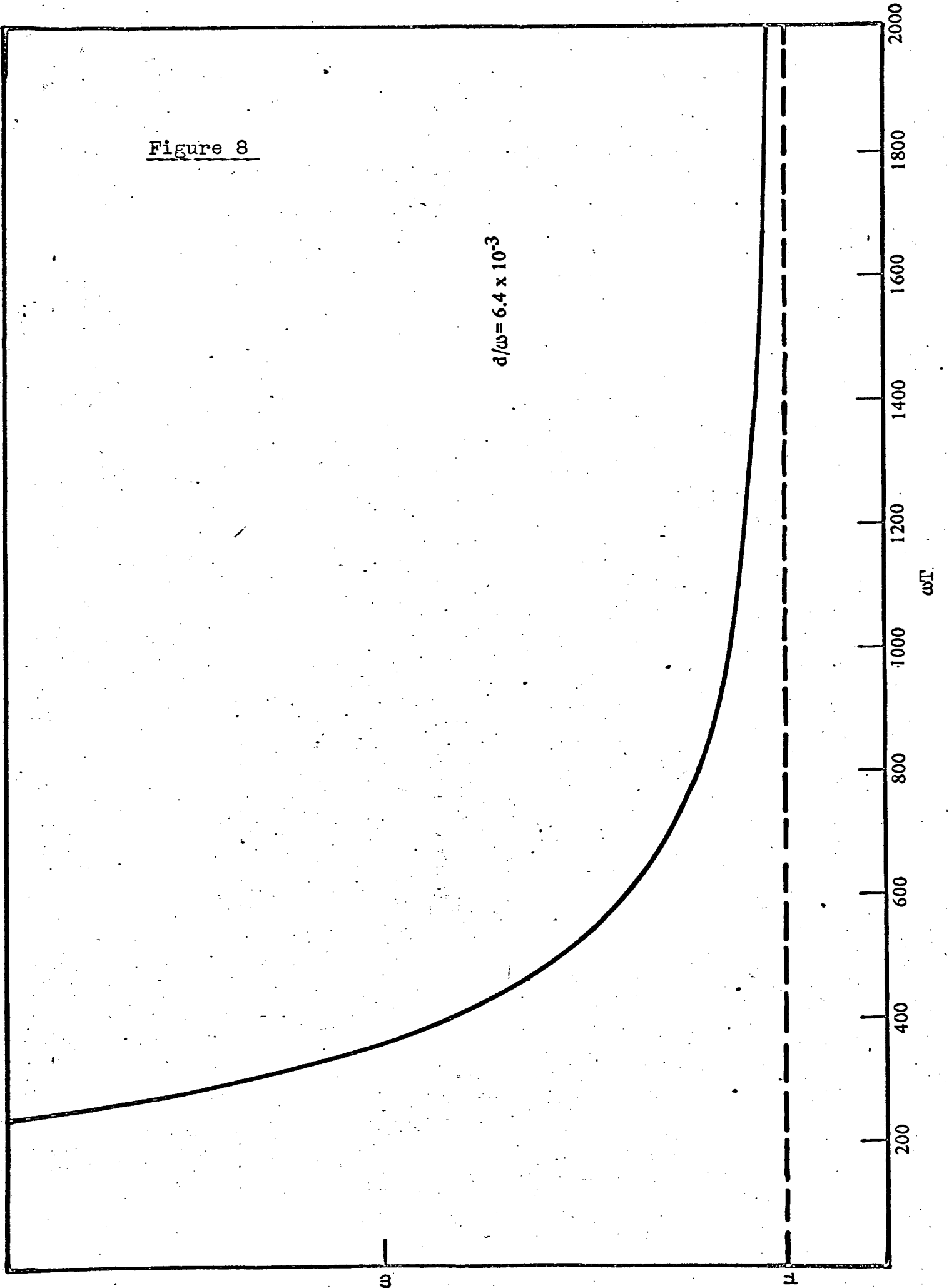


Figure 7

Figure 8



$\omega T = 800$
 $d = 6.4 \times 10^{-3}$

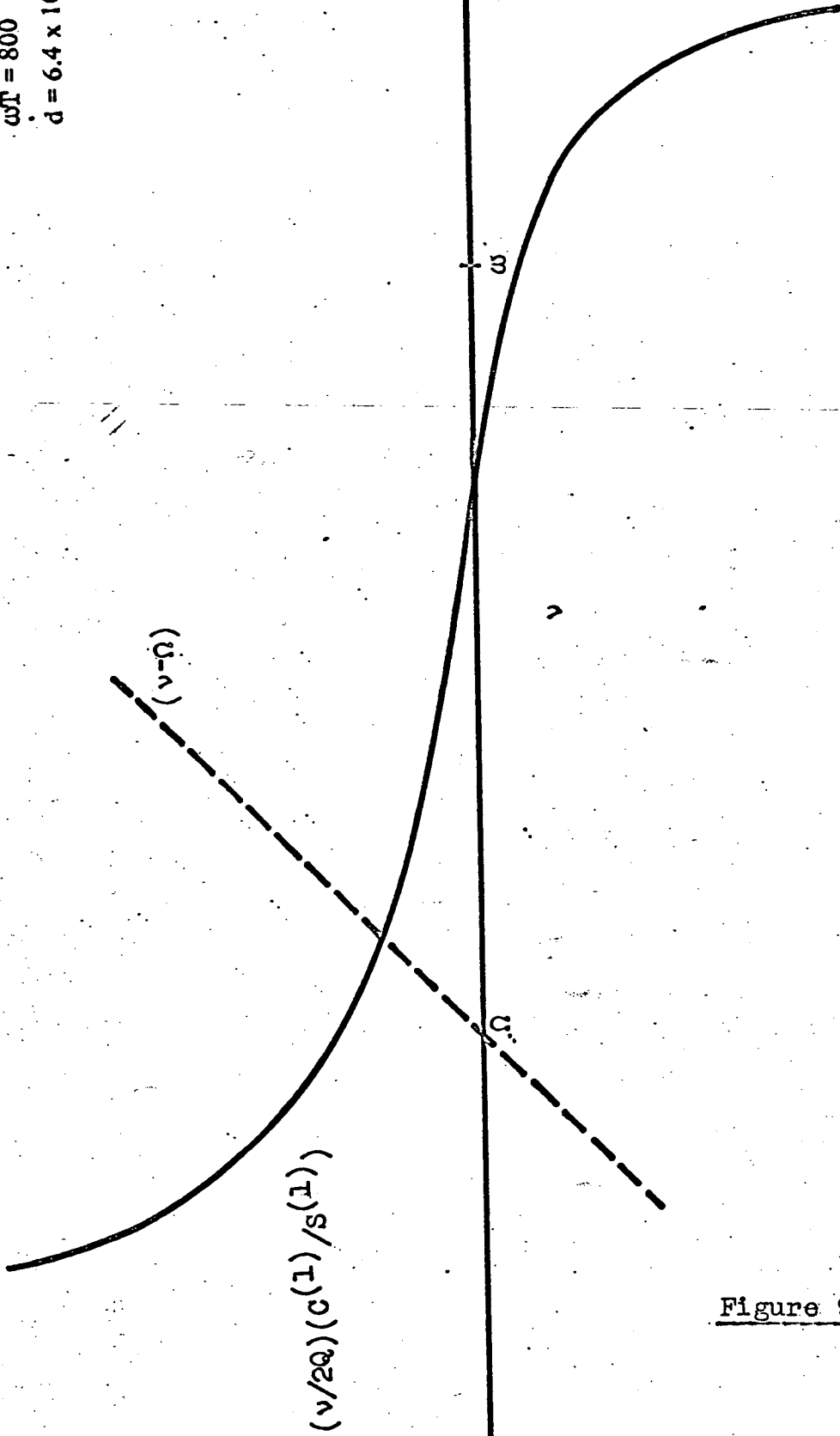


Figure 9a

$\omega T = 200$
 $d = 6.4 \times 10^{-3}$

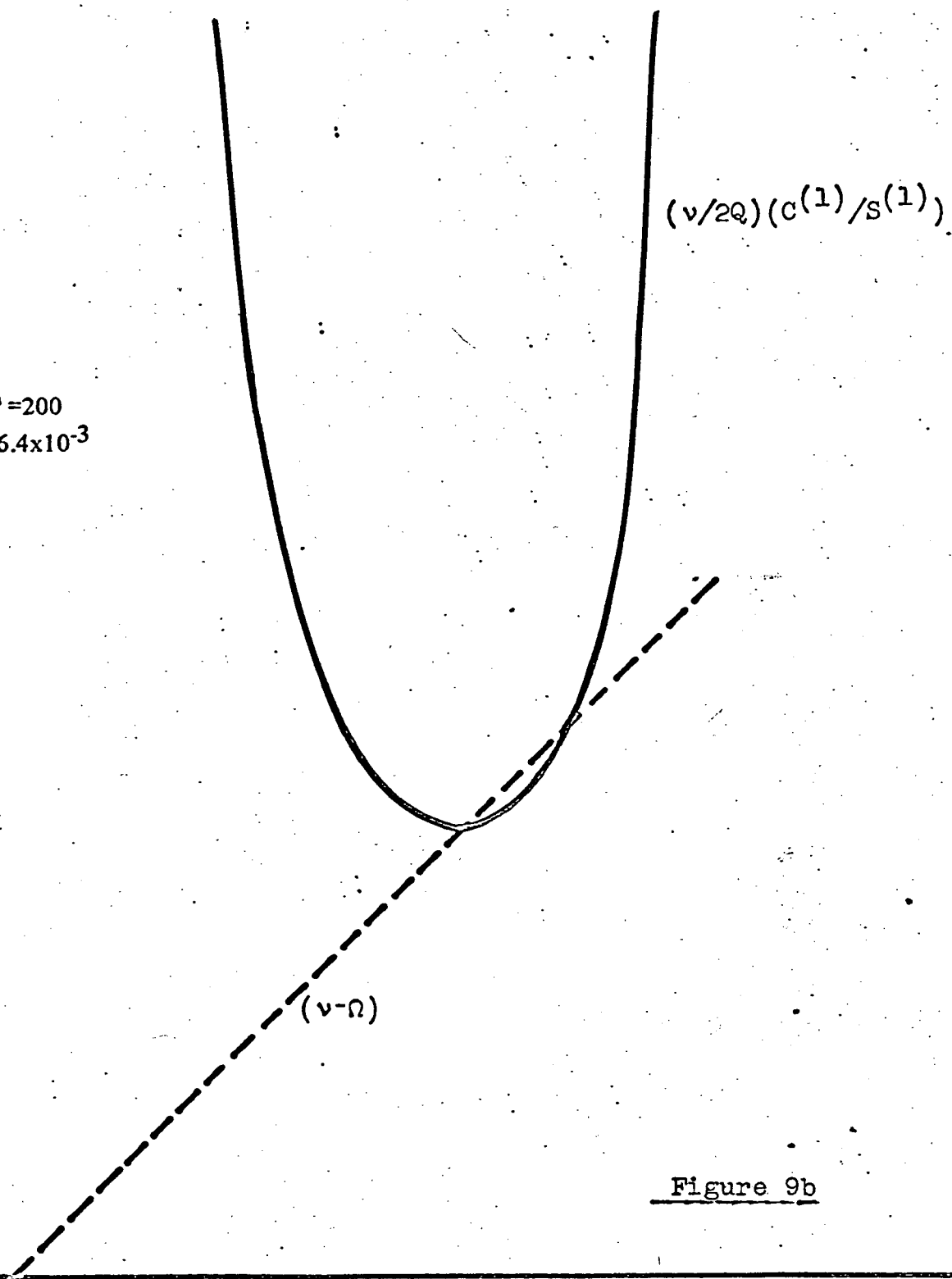


Figure 9b

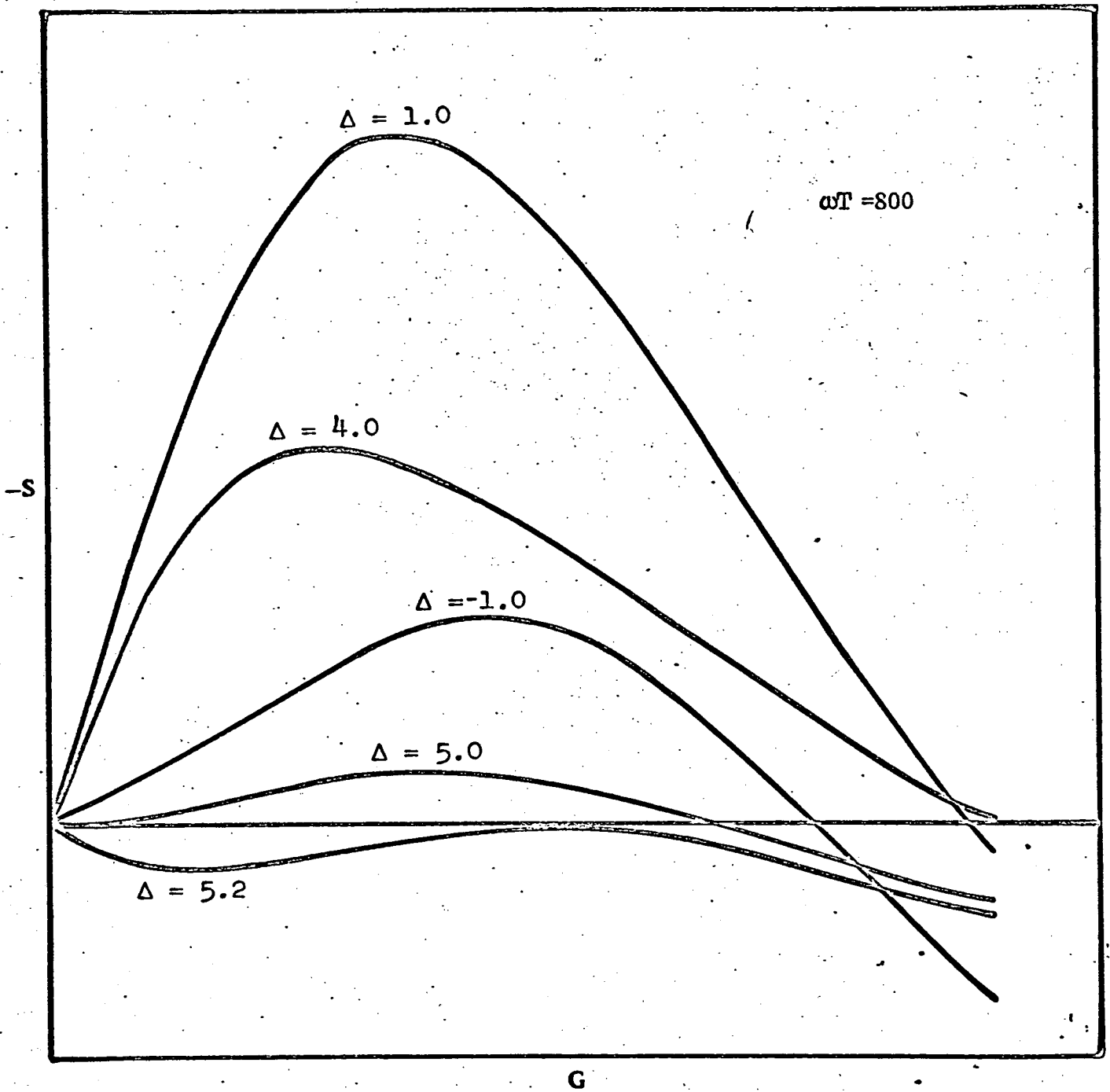


Figure 10

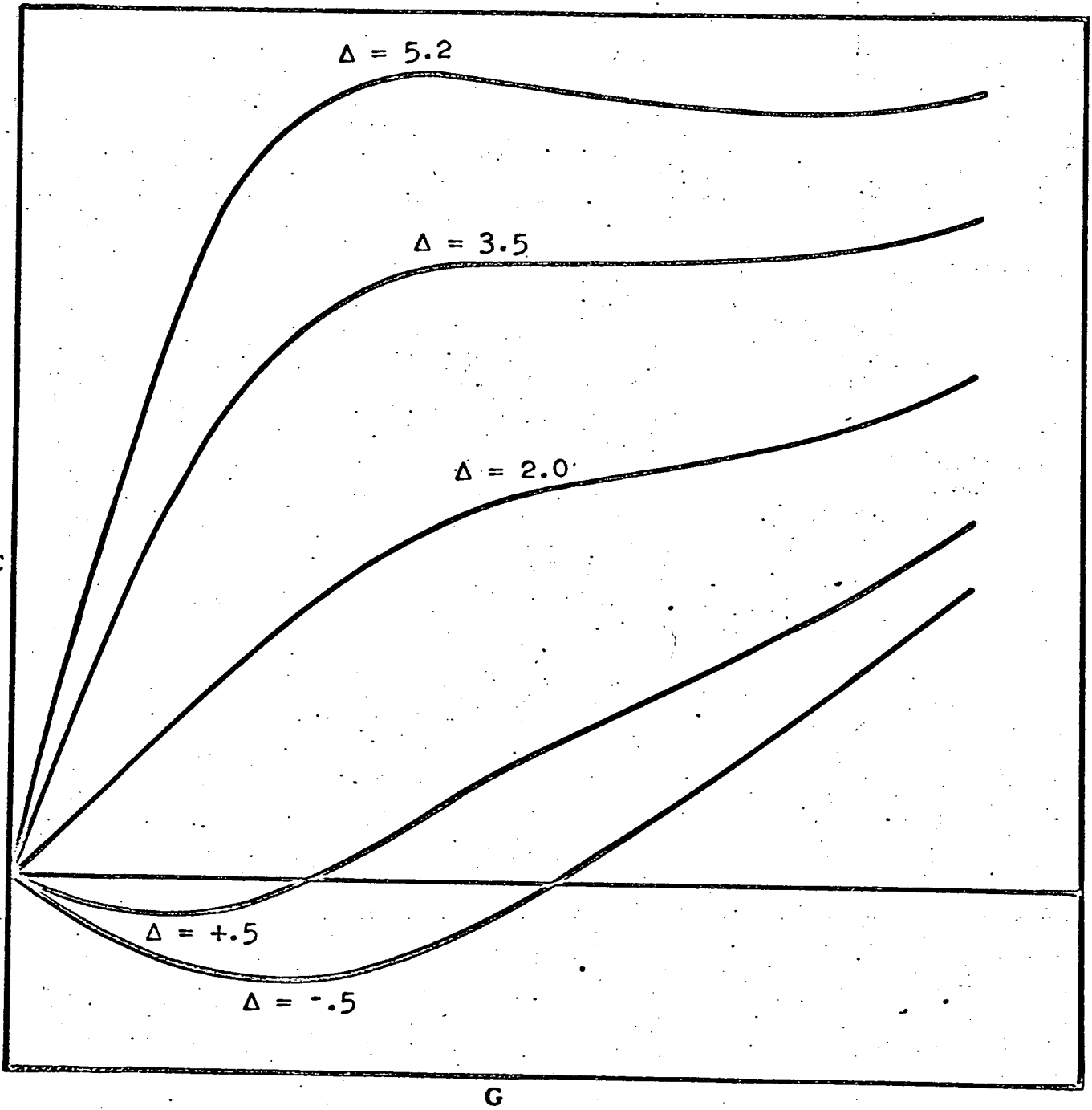


Figure 11

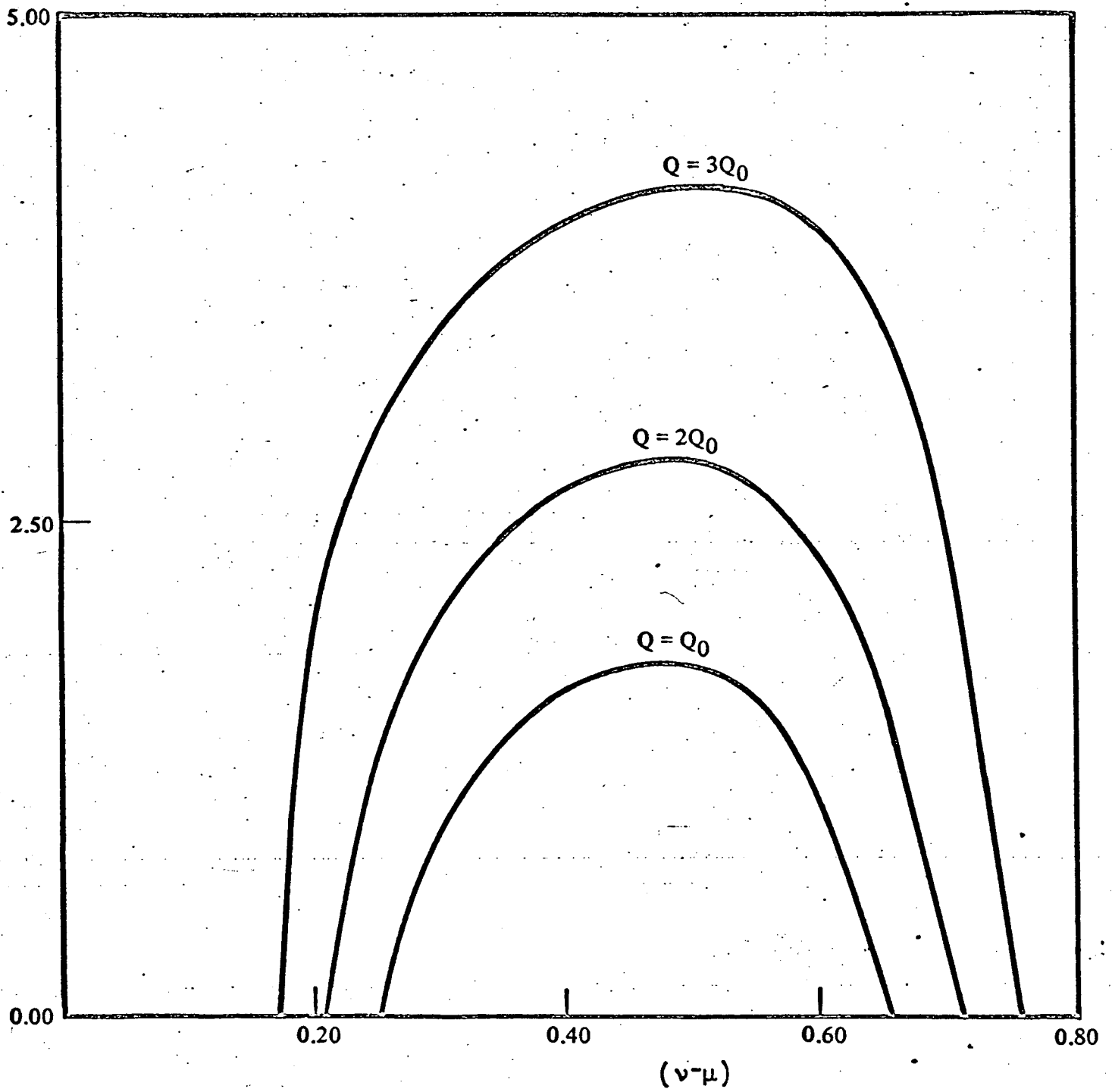
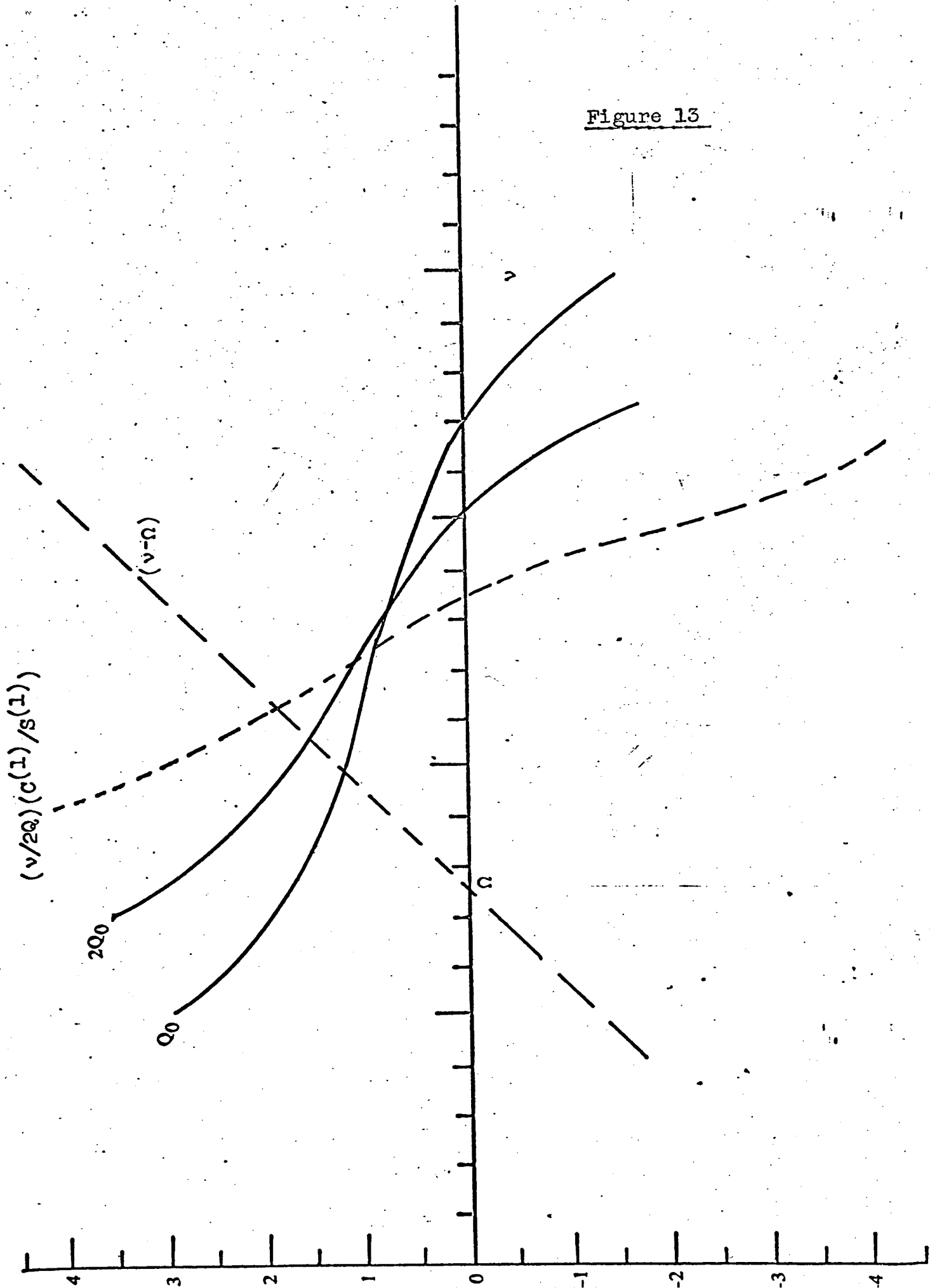


Figure 12

C2

Figure 13



PART II: THE EFFECT OF VELOCITY-CHANGING COLLISIONS

ON THE OUTPUT OF A GAS LASER

I. Introduction

The radiation emitted by an atomic system can be significantly affected by collisions with neighboring atoms. The parameters which determine the shape of a spectral line (atomic energy level separation, decay rate, velocity) fluctuate due to random collisions during the radiative lifetime of the atomic system. There is an extensive literature on the effects of collisions on the shape of spectral lines covering about 70 years. A recent paper¹ gives a comprehensive list of references on this subject.

In a previous publication², a model for a laser oscillator was presented in which the active atoms undergo collisions during their lifetimes. The result was a theoretical expression for the pressure dependence of the intensity of the laser in satisfactory agreement with the experimental studies of Szöke and Javan³ and Cordover⁴. Other authors have derived similar theoretical expressions.⁵

In I, two types of dynamic collisions were considered. The first effect of a foreign perturbing atom on a radiating atom was regarded as a van der Waals interaction which caused the atomic transition frequency to change adiabatically with time (phase changing collisions). In the second effect, considered as independent of the first, the forces on the active atoms caused them to follow some complex zig-zag path. A model in which the atoms return to equilibrium after each collision was used to describe the velocity changes.

The calculations in this paper are similar in form to those of I. The main difference is that a more reasonable model for deflecting collisions is used. It has recently been found⁶ that the simultaneous consideration of deflecting and phase changing collisions requires a complete quantum mechanical treatment of the collision process. A radiating

atom is in a mixture of two atomic states and the center of mass motion of this system, after a collision, cannot in general be described classically.

However, the special case where the van der Waals interaction is the same for both atomic states, can be treated classically. In that situation, phase effects are absent and collisions only produce velocity changes. This paper will only deal with velocity changing collisions. The resulting theoretical expression for the laser intensity may be helpful in isolating the effects of deflecting collisions.

II. Nature of Collisions

The collisions in this paper will be described by the binary interaction of a foreign gas (perturbing) atom with the radiating (emitter) atom. The collision time can be approximated by the quantity $t_c = b_o/v_{rel}$ where b_o is the impact parameter and v_{rel} is the relative velocity of emitter and perturber. The time between collisions for a typical impact parameter b_o is approximately $T = [n\pi b_o^2 v_{rel}]^{-1}$ where n is the number density of perturbers. For pressures of about one Torr, $T \sim 10^{-7}$ seconds, while for most significant collisions t_c is less than 10^{-11} seconds. The case where $T \gg t_c$ is called the impact limit for collisions.

The assumption of impact collisions permits a greatly simplified mathematical treatment of the collision problem. The properties of the system after the collision only depend on the properties before the collision. This situation is characteristic of a Markoff process and facilitates the computation of complicated statistical averages. In the case of binary impact collisions, the Boltzmann equation may be used to obtain a fairly simple mathematical description of the collision history of the atoms.

The next section gives a formal presentation of the laser problem which includes the effects of deflecting collisions.

III. Laser Model

The following model for a gas laser is taken from an earlier paper.⁷ Suitable modifications are made to allow for collision processes.

The laser operates in a one-dimensional, high-Q resonant cavity of length L . The cavity contains a medium of active atoms which acquire nonlinear dipole moments through interaction with a single mode electromagnetic field of the cavity. The requirement for self-sustained oscillations is that the macroscopic polarization of the medium acts as the source for the assumed electromagnetic field (self-consistent field). The electric field in the cavity mode is

$$E(z,t) = E(t) \cos[\nu t + \varphi(t)] \sin Kz \quad (1)$$

and the macroscopic polarization projected on that mode is

$$P(z,t) = [C(t)\cos(\nu t + \varphi(t)) + S(t)\sin(\nu t + \varphi(t))]\sin Kz \quad (2)$$

Using the assumption of slowly varying amplitudes and phases the self-consistency requirement is

$$\dot{E} + \frac{1}{2}(\nu/Q)E = -\frac{1}{2}(\nu/\epsilon_0)S \quad (3a)$$

$$(\nu + \dot{\varphi} - \Omega)E = -\frac{1}{2}(\nu/\epsilon_0)C \quad (3b)$$

where Ω is the cavity frequency with no active medium present.

The active medium consists of an ensemble of atoms with levels a , b and with natural decay rates γ_a , γ_b . The active atoms are introduced into the cavity at rates Λ_a , Λ_b . If the atoms move through the cavity, the position z at time t of a

an atom is given by

$$z = z_0 + \int_{t_0}^t v(\hat{t}) d\hat{t} \quad (4)$$

The integral on the r.h.s. of Eq. (4) allows for the possibility that the atoms undergo deflecting collisions which cause the z-component of velocity to change. If the atomic energy levels are shifted by collisions with neighboring atoms, the transition frequency will be a function $\omega(t)$ of time. As explained in the introduction, these changes will be neglected.

An atom is introduced into the cavity at the position z_0 at the time t_0 in state a or b. The atomic transitions $a \leftrightarrow b$ are caused by the perturbation

$$hV(z,t) = -\rho E(z,t) = -\rho E(t) \sin[K(z_0 + \int_{t_0}^t v(\hat{t}) d\hat{t})] \cos(\nu t + \varphi) \quad (5)$$

where ρ is the electric dipole matrix element

$$\rho = e \langle a | x | b \rangle \quad (6)$$

The equations for the time development of the density matrix ρ for one atom are

$$\begin{aligned} \dot{\rho}_{aa} &= -\gamma_a \rho_{aa} + iV(z,t)(\rho_{ab} - \rho_{ba}) \\ \dot{\rho}_{bb} &= -\gamma_b \rho_{bb} - iV(z,t)(\rho_{ab} - \rho_{ba}) \\ \dot{\rho}_{ab} &= -\gamma_{ab} \rho_{ab} - i\omega \rho_{ab} + iV(z,t)(\rho_{aa} - \rho_{bb}) \\ \rho_{ba} &= \rho_{ab}^* \end{aligned} \quad (7)$$

where $\gamma_{ab} = \frac{1}{2}(\gamma_a + \gamma_b)$ and ω is the transition frequency between levels a and b. Removing the optical frequency ν from the off-diagonal elements of the density matrix by writing

$$\rho_{ab} = \rho_1 e^{-i\nu t} \quad (8)$$

and neglecting terms with time dependence $e^{\pm 2i\nu t}$, Eq. (7) can be rewritten as

$$\begin{aligned} \dot{\rho}_{aa} &= -\gamma_a \rho_{aa} - \frac{1}{2}i(\rho E/\hbar) \sin Kz (\rho_1 - \rho_1^*) \\ \dot{\rho}_{bb} &= -\gamma_b \rho_{bb} + \frac{1}{2}i(\rho E/\hbar) \sin Kz (\rho_1 - \rho_1^*) \\ \dot{\rho}_1 &= -(\gamma_{ab} + i(\omega - \nu)) \rho_1 - \frac{1}{2}i(\rho E/\hbar) \sin Kz (\rho_{aa} - \rho_{bb}) \\ \dot{\rho}_1^* &= -(\gamma_{ab} - i(\omega - \nu)) \rho_1^* + \frac{1}{2}i(\rho E/\hbar) \sin Kz (\rho_{aa} - \rho_{bb}) \end{aligned} \quad (9)$$

where z is given by Eq. (4).

The initial conditions for equations (9) are

$$\begin{aligned} \rho_{\alpha\beta}(a, z_0, t_0, t_0) &= \delta_{\alpha\beta} \delta_{a\alpha} \\ \rho_{\alpha\beta}(b, z_0, t_0, t_0) &= \delta_{\alpha\beta} \delta_{b\alpha} \end{aligned} \quad (10)$$

depending on whether the atom has been introduced into the cavity in state a or b. Formally solving Eqs.(9) gives

$$\begin{aligned} \rho_{aa}(\alpha, z_0, t, t_0) &= e^{-\gamma_a(t-t_0)} \delta_{a\alpha} \\ &\quad - \frac{1}{2}i(\rho E/\hbar) \int_{t_0}^t dt' e^{-\gamma_a(t-t')} \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})] \\ &\quad \times [\rho_1(\alpha, z_0, t', t_0) - \rho_1^*(\alpha, z_0, t', t_0)] \end{aligned}$$

$$\begin{aligned}
\rho_{bb}(\alpha, z_0, t, t_0) &= e^{-\gamma_b(t-t_0)} \delta_{ab} \\
&+ \frac{1}{2} i (\rho E / \hbar) \int_{t_0}^t dt' e^{-\gamma_b(t-t')} \sin\left[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})\right] \\
&\quad \times [\rho_1(\alpha, z_0, t', t_0) - \rho_1^*(\alpha, z_0, t', t_0)] \\
\rho_1(\alpha, z_0, t, t_0) &= -\frac{1}{2} i (\rho E / \hbar) \int_{t_0}^t dt' e^{-[\gamma_{ab} + i(\omega - \nu)](t-t')} \\
&\quad \times \sin\left[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})\right] \\
&\quad \times [\rho_{aa}(\alpha, z_0, t', t_0) - \rho_{bb}(\alpha, z_0, t', t_0)]
\end{aligned} \tag{11}$$

The macroscopic polarization $P(z, t)$ is obtained by summing the dipole moments of all active atoms that arrive at z at time t -- no matter where or when they were excited or how they got to (z, t) , i.e.,

$$\begin{aligned}
P(z, t) &= \rho \int_{-\infty}^t dt_0 \langle \int dz_0 \sum_{\alpha=a, b} \Lambda_{\alpha}(z_0, t_0) \delta(z - z_0 - \int_{t_0}^t v(\hat{t}) d\hat{t}) \\
&\quad \times [\rho_{ab}(\alpha, z_0, t, t_0) + \rho_{ba}(\alpha, z_0, t, t_0)] \rangle_{\text{path}}
\end{aligned} \tag{12}$$

The symbol $\langle \rangle_{\text{path}}$ in Eq. (12) denotes a statistical average over all collision histories of atoms which start at (z_0, t_0) and end at (z, t) . This average will be considered in detail in subsequent sections. In order to find the appropriate path averages, the history of each atom must be traced using the microscopic equations (11). It is convenient at this time to define microscopic versions of

macroscopic variables to be used later. Let

$$n(\alpha, z_0, t, t_0) = [\rho_{aa}(\alpha, z_0, t, t_0) - \rho_{bb}(\alpha, z_0, t, t_0)] \quad (13a)$$

$$s(\alpha, z_0, t, t_0) = -i\rho[\rho_1(\alpha, z_0, t, t_0) - \rho_1^*(\alpha, z_0, t, t_0)] \quad (13b)$$

where $n(\alpha, z_0, t, t_0)$ is the microscopic version of the population inversion density of the atomic ensemble and $s(\alpha, z_0, t, t_0)$ is the microscopic version of the out-of-phase part of the polarization S defined in Eq. (2).

Using Eqs. (11) two coupled integral equations for n and s can be obtained.

$$\begin{aligned} n(\alpha, z_0, t, t_0) = & [e^{-\gamma_a(t-t_0)}\delta_{aa} - e^{-\gamma_b(t-t_0)}\delta_{ab}] \\ & + \frac{1}{2}(E/\hbar) \int_{t_0}^t dt' [e^{-\gamma_a(t-t')} - e^{-\gamma_b(t-t')}] \\ & \times \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t})d\hat{t})] s(\alpha, z_0, t', t_0) \quad (14a) \end{aligned}$$

$$\begin{aligned} s(\alpha, z_0, t, t_0) = & -\frac{1}{2}(\rho^2 E/\hbar) \int_{t_0}^t dt' [e^{-[\gamma_{ab} - i(\omega - \nu)](t-t')} + \text{c.c.}] \\ & \times \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t})d\hat{t})] n(\alpha, z_0, t', t_0) \quad (14b) \end{aligned}$$

For the perturbation treatment in this section, Eqs. (14) will be reduced to a single integral equation. First define

(9)

$$s(z_0, t, t_0) = \sum_{\alpha=a, b} \Lambda_{\alpha}(z_0, t_0) s(\alpha, z_0, t, t_0) \quad (15)$$

Substituting (14a) into (14b) and using (15) gives a single integral equation for $s(z_0, t, t_0)$

$$\begin{aligned} s(z_0, t, t_0) = & -\frac{1}{2}(\rho^2 E/\hbar) \int_{t_0}^t dt' \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})] \\ & \times [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] \\ & \times [\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)}] \\ & - \frac{1}{4}(\rho^2 E^2/\hbar^2) \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})] \\ & \times \sin[K(z_0 + \int_{t_0}^{t''} v(\hat{t}) d\hat{t})] [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] \\ & \times [e^{-\gamma_a(t'-t'')} + e^{-\gamma_b(t'-t'')}] s(z_0, t'', t_0) \end{aligned} \quad (16)$$

where $\mu = \gamma_{ab} - i(\omega - v)$.

Equation (16) is still a microscopic equation. The solution of Eq. (16) to first order in the electric field E is

$$\begin{aligned} s^{(1)}(z_0, t, t_0) = & -\frac{1}{2}(\rho^2 E/\hbar) \int_{t_0}^t dt' \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})] \\ & \times [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] \\ & \times [\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)}] \end{aligned} \quad (17)$$

and the third order solution is

$$\begin{aligned}
s^{(3)}(z_0, t, t_0) &= \frac{1}{8}(\rho^4 E^3 / \hbar^3) \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' \\
&\quad \sin[K(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t})] \sin[K(z_0 + \int_{t_0}^{t''} v(\hat{t}) d\hat{t})] \\
&\quad \times \sin[K(z_0 + \int_{t_0}^{t'''} v(\hat{t}) d\hat{t})] [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] \\
&\quad \times [e^{-\gamma_a(t'-t'')} + e^{-\gamma_b(t'-t'')}] [e^{-\mu(t''-t''')} + e^{-\mu^*(t''-t''')}] \\
&\quad \times [\Lambda_a e^{-\gamma_a(t'''-t_0)} - \Lambda_b e^{-\gamma_b(t'''-t_0)}]
\end{aligned} \tag{18}$$

The atoms under consideration arrive at the point z at time t . If $s^{(n)}(z_0, t, t_0)$ is the n th iteration of Eq. (16), then define

$$s^{(n)}(z, t, t_0) \equiv \int dz_0 s^{(n)}(z_0, t, t_0) \delta(z - z_0 - \int_{t_0}^t v(t) dt) \tag{19}$$

The n th order contribution to the out-of-phase macroscopic polarization is

$$S^{(n)}(z, t) = \int_{-\infty}^t dt_0 \langle s^{(n)}(z, t, t_0) \rangle_{\text{path}} \tag{20}$$

The path average is taken before summing over all initial excitation times, t_0 . The first order contribution then becomes

$$\begin{aligned}
S^{(1)}(z, t) &= -\frac{1}{2}(\rho^2 E / \hbar) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \langle \sin[K(z - \int_{t_0}^{t'} v(\hat{t}) d\hat{t})] \\
&\quad \times [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] [\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)}] \rangle_{\text{path}}
\end{aligned} \tag{21}$$

and the third order contribution is

$$\begin{aligned}
 s^{(3)}(z, t) = & \frac{1}{8} (e^4 E^3 / \hbar^3) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' \\
 & \langle \sin[K(z - \int_{t'}^t v(\hat{t}) d\hat{t})] \sin[K(z - \int_{t''}^t v(\hat{t}) d\hat{t})] \sin[K(z - \int_{t'''}^t v(\hat{t}) d\hat{t})] \rangle \\
 & \times [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] [e^{-\gamma_a(t'-t'')} + e^{-\gamma_b(t'-t'')}] \\
 & \times [e^{-\mu(t''-t''')} + e^{-\mu^*(t''-t''')}] \\
 & \times [\Lambda_a e^{-\gamma_a(t'''-t_0)} - \Lambda_b e^{-\gamma_b(t'''-t_0)}] \rangle_{\text{path}}
 \end{aligned}$$

(22)

When the product of the three sine functions is written in exponential form eight terms will result.

The next section will deal with path averages.

IV. Path Averages

The path averages may be calculated using a classical density function $f(z_0, v_0, t_0 | z, v, t)$ for the motion of the active atoms. The mathematical formulation of the problem is given by the Boltzmann equation for the z-component motion of the atoms

$$\partial f / \partial t + v_z \partial f / \partial z = J(f) \quad (23)$$

where $J(f)$ is an integral operator describing the collisions. The initial condition for Eq. (23) is

$$f(z_0, v_0, t_0 | z, v, t_0) = \delta(v - v_0) \delta(z - z_0) \quad (24)$$

If the process under consideration is stationary in time and the medium spatially homogeneous then f may be rewritten in the form

$$f(z_0, v_0, t_0 | z, v, t) = f(v_0 | z - z_0, t - t_0) \quad (25)$$

The path average of a function $R(v_0 | v, z - z_0, t - t_0)$ is then given by⁸

$$\langle R(v_0 | v, z - z_0, t - t_0) \rangle_{\text{path}} = \int_{-\infty}^{+\infty} dv_0 W(v_0) \int dv \int d(\Delta z) R(v_0 | v, \Delta z, t - t_0) f(v_0 | v, \Delta z, t - t_0) \quad (26)$$

where $W(v_0)$ is the initial velocity distribution and

$$\Delta z = z - z_0 .$$

The path averages to be evaluated must take into consideration the complete history of each atom. Therefore, the quantity to be averaged in Eq. (21) is

$$\begin{aligned} \mathcal{J}^{(1)} = & \sin\left[K\left(z - \int_{t'}^t v(\hat{t})d\hat{t}\right)\right] \left[e^{-\mu(t-t')} + e^{-\mu^*(t-t')}\right] \\ & \times \left[\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)}\right] \end{aligned} \quad (27)$$

In order to compute the path average of $\mathcal{J}^{(1)}$ two density functions $f(v_0 | v', z'-z_0, t'-t_0)$ and $f(v' | v, z-z', t-t')$ must be used since $\mathcal{J}^{(1)}$ is a product $\Lambda(t'-t_0)R(z-z', t-t')$ where

$$\Lambda(t'-t_0) = \left[\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)}\right] \quad (28a)$$

and

$$R(z-z', t-t') = \sin [Kz - K(z-z')] \left[e^{-\mu(t-t')} + e^{-\mu^*(t-t')}\right] \quad (28b)$$

where $z-z' = \int_{t'}^t v(t)dt$. The path average of $\mathcal{J}^{(1)}$ can then be defined as

$$\begin{aligned} \langle \mathcal{J}^{(1)} \rangle_{\text{path}} = & \int dv_0 W(v_0) \int dv' \int d(\Delta z_0) f(v_0 | v', \Delta z_0, t'-t_0) \\ & \times \Lambda(t'-t_0) \int dv \int d(\Delta z') f(v' | v, \Delta z, t-t') R(\Delta z', t-t') \end{aligned} \quad (29)$$

where $\Delta z_0 = z'-z_0$ and $\Delta z' = z-z'$.

It is useful to define the quantities

$$G_{\kappa}(v' | v, t-t') = \int d(\Delta z') f(v' | v, \Delta z', t-t') e^{i\kappa K \Delta z'} \quad (30)$$

so that Eq. (29) becomes

$$\begin{aligned}
 \langle \mathcal{J}^{(1)} \rangle_{\text{path}} &= \int dv_0 W(v_0) \int dv' \int dv G_0(v_0 | v', t' - t_0) \\
 &\quad \times [e^{iKz} G_{+1}(v' | v, t - t') - e^{-iKz} G_{-1}(v' | v, t - t')] \\
 &\quad \times [\Lambda_a e^{-\gamma_a(t' - t_0)} - \Lambda_b e^{-\gamma_b(t' - t_0)}] [e^{-\mu(t - t')} + e^{-\mu^*(t - t')}]
 \end{aligned}
 \tag{31}$$

Then from Eq. (21), the first order contribution to $S(z, t)$ is

$$S^{(1)}(z, t) = -\frac{1}{2} (\rho^2 E / \hbar) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \langle \mathcal{J}^{(1)} \rangle_{\text{path}}
 \tag{32}$$

Using the same procedure for the third order term, Eq. (22) becomes

$$S^{(3)}(z, t) = (1/32) (\rho^4 E^3 / \hbar^3) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' \langle \mathcal{J}^{(3)} \rangle_{\text{path}}
 \tag{33}$$

The function $\langle \mathcal{J}^{(3)} \rangle_{\text{path}}$ is the integrand of Eq. (22) and can be written in the following form

$$\begin{aligned}
 \langle \mathcal{J}^{(3)} \rangle_{\text{path}} &= \frac{1}{2i} \int dv_0 \int dv'''' \int dv''' \int dv'' \int dv' \int dv G_0(v_0 | v''''', t'''' - t_0) \\
 &\quad \times \{ e^{iKz} [G_{-1}(v'''' | v''', t'' - t''') G_0(v'' | v', t' - t'') G_{-1}(v' | v, t - t')] \\
 &\quad + G_{-1}(v'''' | v''', t'' - t''') G_{-2}(v'' | v', t' - t'') G_{-1}(v' | v, t - t')] \\
 &\quad + G_{+1}(v'''' | v''', t'' - t''') G_0(v'' | v', t' - t'') G_{-1}(v' | v, t - t')] \\
 &\quad - e^{-iKz} [G_{+1}(v'''' | v''', t'' - t''') G_0(v'' | v', t' - t'') G_{+1}(v' | v, t - t')] \\
 &\quad - e^{-iKz} [G_{-1}(v'''' | v''', t'' - t''') G_0(v'' | v', t' - t'') G_{+1}(v' | v, t - t')]
 \end{aligned}$$

(continued on next page)

$$\begin{aligned}
& \times [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] [e^{-\gamma_a(t'-t'')} + e^{-\gamma_b(t'-t'')}] \\
& \times [e^{-\mu(t''-t''')} + e^{-\mu^*(t''-t''')}] \Lambda(t'''-t_0)
\end{aligned}
\tag{34}$$

where $\Lambda(t'''-t_0)$ is given in Eq.(28) and the two terms in $e^{\pm 3iKz}$ have been neglected.

By changing orders of time integration, $S^{(1)}(z,t)$ and $S^{(3)}(z,t)$ can be written in terms of Laplace transforms G_κ of the G_κ 's where

$$G_\kappa(v'|v,\alpha) = \int_0^\infty d\tau e^{-\alpha\tau} G_\kappa(v'|v,\tau)
\tag{35}$$

Then

$$\begin{aligned}
S^{(1)}(z,t) = & - (1/4i) (\rho^2 E/\hbar) \int dv_0 W(v_0) \int dv' \int dv \\
& [\Lambda_a G_0(v_0|v',\gamma_a) - \Lambda_b G_0(v_0|v',\gamma_b)] \\
& \times \{e^{iKz} [G_{-1}(v'|v,\mu) + G_{-1}(v'|v,\mu^*)] \\
& - e^{-iKz} [G_{+1}(v'|v,\mu) + G_{+1}(v'|v,\mu^*)]\}
\end{aligned}
\tag{36}$$

and

$$\begin{aligned}
S^{(3)}(z, t) = & (1/64i) (p^4 E^3 / \hbar^3) \int dv_0 W(v_0) \int dv'''' \\
& [\Lambda_a G_0(v_0 | v'''' , \gamma_a) - \Lambda_b G_0(v_0 | v'''' , \gamma_b)] \\
\times & \int dv'' \int dv' \int dv \{ e^{iKz} [G_{+1}(v'''' | v'' , \mu) + G_{+1}(v'''' | v'' , \mu^*) \\
& + G_{-1}(v'''' | v'' , \mu) + G_{-1}(v'''' | v'' , \mu^*)] \\
& \times (G_0(v'' | v' , \gamma_a) + G_0(v'' | v' , \gamma_b)) \\
& \times (G_{-1}(v' | v , \mu) + G_{-1}(v' | v , \mu^*))] \\
& - e^{-iKz} [G_{+1} - G_{-1}] \}
\end{aligned}$$

(37)

In the third order contribution only the terms corresponding to the high Doppler limit have been included. The next section will be devoted to finding a reasonable collision operator $J(f)$ used in Eq.(23) so that expressions for G_{κ} in Eqs. (36) and (37) can be calculated.

V. Binary Collisions and
the Boltzmann Equation

For the following calculations, assume that the active atoms have mass m and undergo binary collisions with inactive atoms of mass M . The gas of inactive atoms is considered to be in thermal equilibrium. At any time let the velocity of the active atom be denoted by \vec{v} and the velocity of the perturbing atom by \vec{V} both measured in the laboratory frame.

Viewed in the center of mass system (cm) the scattering process changes the velocity of the active atom from \vec{v}_{cm} to \vec{v}'_{cm} by rotating it through an angle θ . (See Fig. 1). The velocity of the active atom in the cm system before the collision is given by

$$\vec{v}_{cm} = [M/(m+M)] (\vec{v} - \vec{V}) \quad (38)$$

where $(\vec{v} - \vec{V})$ is the relative velocity of the emitter and perturbing atoms. The velocity of the center of mass in the laboratory system is

$$\vec{U} = (m\vec{v} + M\vec{V})/(m+M) \quad (39)$$

Since the magnitude of the relative velocity does not change after the collision \vec{v}'_{cm} is given by

$$\vec{v}'_{cm} = [M/(m+M)] |\vec{v} - \vec{V}| \hat{\theta} \quad (40)$$

where the unit vector $\hat{\theta}$ makes an angle θ with the relative velocity $(\vec{v} - \vec{V})$ (see Fig. 1). The velocity of the active atom in the laboratory system after a collision is

$$\vec{v}' = \vec{v}'_{cm} + \vec{U} \quad (41)$$

Adding and subtracting \vec{v} from the r.h.s. of Eq. (41) and using (39) and (40) Eq. (41) becomes

$$\vec{v}' = \vec{v} + [M/(m+M)] \{ \vec{v} - \vec{V} | \hat{\theta} - (\vec{v} - \vec{V}) \} \quad (42)$$

Let \hat{p} and \hat{q} be unit vectors parallel and perpendicular respectively to $(\vec{v} - \vec{V})$ (see Fig. 1). Projecting the vectors in curly brackets in Eq. (42) onto \hat{p} and \hat{q} gives the result

$$\vec{v}' = \vec{v} + [M/(m+M)] |\vec{v} - \vec{V}| \{ \sin\theta \hat{q} - (1 - \cos\theta) \hat{p} \} \quad (43)$$

or

$$\vec{v}' = \vec{v} - [2M/(m+M)] \{ \sin(\theta/2) \hat{p} - \cos(\theta/2) \hat{q} \} \left[\sin(\theta/2) |\vec{v} - \vec{V}| \right] \quad (44)$$

The quantity in curly brackets in Eq. (44) is a unit vector \hat{s} making an angle $[(\theta - \pi)/2]$ with the relative velocity $(\vec{v} - \vec{V})$ and the quantity in square brackets is the inner product $\hat{s} \cdot (\vec{v} - \vec{V})$. Equation (44) then becomes

$$\vec{v}' = \vec{v} - [2M/(m+M)] \hat{s} [\hat{s} \cdot (\vec{v} - \vec{V})] \quad (45)$$

If the potential between the two atoms is $U(r)$ then θ is given by⁹

$$\theta = |\pi - 2\Phi_0|$$

where $\Phi_0 = \int_{r_{\min}}^{\infty} dr [(2\eta E/J^2) - (2\eta U(r)/J^2) - (1/r^2)]^{-\frac{1}{2}} r^{-2}$

and $\eta = [mM/(m+M)]$ (reduced mass)

$E = (\eta/2) |\vec{v} - \vec{V}|^2$ (kinetic energy in cm system)

$J^2 = 2\eta b^2 E$ (square of angular momentum)

$b =$ impact parameter.

(46)

The collision operator in Eq. (23) in general is¹⁰

$$J(f) = \int d\vec{V} \int d\Omega |\vec{v} - \vec{V}| \sigma(|\vec{v} - \vec{V}|, \theta) [f(\vec{v}') F(\vec{V}') - f(\vec{v}) F(\vec{V})]$$

(47)

where $F(\vec{V}) = N(\beta_M/\pi)^{3/2} \exp[-\beta_M V^2]$

(with $\beta_M = [M/2K_B\Theta]$ and $\Theta =$ temperature)

is the velocity distribution of the perturber atoms multiplied by the number density of perturber atoms. The differential cross section for the collision $(\vec{v}, \vec{V}) \rightarrow (\vec{v}', \vec{V}')$ which turns the relative velocity through the angle θ is $\sigma(|\vec{v} - \vec{V}|, \theta)$.

In practice, $J(f)$ is difficult to express in closed form for specific laws of force. In only one case, that of Maxwell molecules ($U(r) = B/r^5$), can a usable form of $J(f)$ be obtained. In that situation the product $|\vec{v} - \vec{V}| \sigma$ depends only on θ .

For most applications it is sufficient to choose a phenomenological collision kernels $W(\vec{v}|\vec{v}')$ (probability per unit time for going from velocity \vec{v} to velocity \vec{v}'). In that case the Boltzmann equation may be written in the following three-dimensional form

$$\partial f / \partial t + \vec{v} \cdot \vec{\nabla} f = \int d\vec{v}' [W(\vec{v}'|\vec{v})f(\vec{v}', \vec{r}, t) - W(\vec{v}|\vec{v}')f(\vec{v}, \vec{r}, t)] \quad (48)$$

Comparing Eq.(48) and Eq.(47) for $J(f)$ gives an equation for $W(\vec{v}|\vec{v}')$

$$W(\vec{v}|\vec{v}') = \int d\vec{V} \int d\Omega |\vec{v}-\vec{V}| \sigma(|\vec{v}-\vec{V}|, \theta) F(\vec{V}) \delta(\vec{v}' - \vec{v}'(\vec{v}, \vec{V}, \theta)) \quad (49)$$

where $\vec{v}'(\vec{v}, \vec{V}, \theta)$ is given by Eq. (45).

For the laser problem, only velocity changes along the cavity axis (z-axis) will affect the polarization. Therefore, a collision kernel $W(v_z|v'_z)$ can be used which only describes the z-velocity changes. Averaging Eq.(49) over all possible initial v_x and v_y with a Maxwell distribution and integrating over all final v_x' and v_y' gives

$$\begin{aligned} W(v_z|v'_z) &= \int dv_x W_m(v_x) \int dv_y W_m(v_y) \int dv_x' \int dv_y' W(\vec{v}|\vec{v}') \\ &= \int dv_x W_m(v_x) \int dv_y W_m(v_y) \int d\vec{V} \int d\Omega |\vec{v}-\vec{V}| \sigma(|\vec{v}-\vec{V}|, \theta) F(\vec{V}) \\ &\quad \times \int dv_x' \int dv_y' \delta(v_x' - v_x'(\vec{v}, \vec{V}, \theta)) \delta(v_y' - v_y'(\vec{v}, \vec{V}, \theta)) \\ &\quad \times \delta(v_z' - v_z'(\vec{v}, \vec{V}, \theta)) \end{aligned} \quad (50)$$

Doing the v_x' and v_y' integrations gives the expected one-dimensional counterpart of equation (49)

$$W(v_z' | v_z') = \int dv_x W_m(v_x) \int dv_y W_m(v_y) \int dV \int d\Omega |\vec{v}-\vec{V}| \sigma(|\vec{v}-\vec{V}|, \theta) \\ \times F(\vec{V}) \delta(v_z' - v_z'(\vec{v}, \vec{V}, \theta)) \quad (51)$$

The quantity $v_z'(\vec{v}, \vec{V}, \theta)$ is given by Eq. (45) as

$$v_z'(\vec{v}, \vec{V}, \theta) = v_z - [2M/(m+M)] s_z [\hat{s} \cdot (\vec{v} - \vec{V})] \quad (52)$$

Figure 2 shows a typical intermolecular potential. The potential usually varies as $1/r^6$ for large values of r . The repulsive part of the potential is not very well determined and fits of $1/r^{12}$ and higher inverse powers of r have been used. In order to simplify the calculation, the repulsive part of the intermolecular potential will be represented by a hard core. The potential $U(r)$ to be used then becomes (see Fig. 2)

$$U(r) = \begin{cases} -B/r^6 & \text{for } r > r_0 \text{ (van der Waals potential)} \\ \infty & \text{for } r = r_0 \text{ (hard sphere potential)} \end{cases} \quad (53)$$

where B is the dipole-dipole interaction coefficient and r_0 is the hard core radius.

It is very difficult to calculate a closed form for $W(v_z | v_z')$ from equation (51) using the potential (53). An approximate form can be deduced using a computer to simulate the integrals in Eq. (51). The following procedure was used to determine $W(v_z | v_z')$ for the potential in Eq. (53):

Choose and fix v_z . The following steps are repeated many times:

- (i) Choose v_x and v_y from a Maxwell velocity distribution.
- (ii) Choose V_x, V_y, V_z from a Maxwell velocity distribution.
- (iii) Choose impact parameter b at random in the range $0 \rightarrow 10^{-6}$ cm.
- (iv) Calculate v_z' from equations (46) and (52) for the potential (53) by integrating the equations of motion.
- (v) Assign weight $Nb\Delta b |\vec{v}-\vec{V}|$ (probability of collision per unit time associated with impact parameter in the range $[b, b+\Delta b]$ and with relative velocity $(\vec{v}-\vec{V})$).
- (vi) Construct frequency table, i.e. sum up all the weights $Nb\Delta b |\vec{v}-\vec{V}|$ of final z -velocities in bins of size Δv_z .

For the computer calculation $B = 4.22 \times 10^{-56}$ erg-cm⁶ and $r_0 = 5.0 \times 10^{-8}$ cm.

Considering only collisions which miss the hard core, the final z -velocity distribution is sharply peaked around the initial velocity v_z with over 95% of the v_z' within 1% of v_z . Collisions reaching the hard core, however, lead to more significant velocity changes. For these collisions, the resulting $W(v_z | v_z')$ has the form of a displaced Gaussian (see Fig. 3).

The following discussion on one-dimensional, hard sphere, elastic collisions may give some insight into the above result for hard core collisions. Assume all the particles are constrained to move only in one dimension and make elastic collisions. The same notation will be used as in the three-dimensional case.

Using conservation of energy and momentum

$$v' = [M/(m+M)]\{ V + (m/M)v \pm |v-V| \} \quad (54)$$

In the case of a collision ($v' \neq v$)

$$v' = [2M/(m+M)]V + [(m-M)/(m+M)]v \quad (55)$$

The probability of going from v to v' is analogous to Eq. (49)

$$W(v|v') = (1/T) (\beta_M/\pi)^{\frac{1}{2}} \int dV e^{-\beta_M V^2} \times \delta(v' - [2M/(m+M)]V - [(m-M)/(m+M)]v) \quad (56)$$

where $(1/T)$ = the frequency of collisions.

Let $U = [2M/(m+M)]V$. Then Eq. (56) becomes

$$W(v|v') = (1/T) (\beta/\pi)^{\frac{1}{2}} \exp[-\beta(v' - \Gamma v)^2] \quad (57)$$

where $\Gamma = [(m-M)/(m+M)]$ and $\beta = \beta_M [(m+M)/2M]^2$ (58)

The results of the above one-dimensional calculation suggest fitting the numerical results for the three-dimensional hard core collisions to a kernel of the form

$$W(v_z | v_z') = (1/T)(\beta/\pi)^{\frac{1}{2}} \exp[-\beta(v_z' - \Gamma v_z)^2] \quad (59)$$

As in the one dimensional problem, β and Γ are functions of the mass ration (m/M) (see Fig. 4). In addition β and Γ and $(1/T)$ are functions of v_z . However, the v_z dependence of those parameters will be neglected in order to simplify subsequent calculations.

Note: Henceforth v_z will be denoted by v since only one velocity component is under consideration.

The conditions of equilibrium impose certain restraints on the values of β and Γ . At equilibrium, the collision operator in Eq. (23) must vanish. Writing $J(f)$ in terms of the collision kernel (59) in the form of Eq. (48) at equilibrium gives

$$\int dv' [W(v|v')W_m(v) - W(v'|v)W_m(v')] = 0 \quad (60)$$

where $W(v|v')$ is given by Eq. (59) and

$$W_m(v) = (\beta_m/\pi)^{\frac{1}{2}} \exp[-\beta_m v^2] \quad (61)$$

Doing the v' integral in Eq.(60) gives

$$(1/T)\exp[-\beta_m v^2] - (1/T)[\beta/(\beta\Gamma^2 + \beta_m)]^{\frac{1}{2}} \exp[-\beta\beta_m v^2/(\beta\Gamma^2 + \beta_m)] = 0$$

(62)

Simplifying Eq. (62) gives

$$[\beta/(\beta\Gamma^2 + \beta_m)]^{\frac{1}{2}} \exp\{-\beta_m v^2([\beta(1-\Gamma^2) - \beta_m]/(\beta\Gamma^2 + \beta_m))\} = 1$$

(63)

Equation (63) is satisfied for every v only if

$$\beta(1-\Gamma^2) = \beta_m$$

(64)

For the collision kernel (59) as obtained by numerical methods, it was found that the quantity $[\beta(1-\Gamma^2)/\beta_m]$ ranged from .982 to .962 for mass ratios $(m/M) = 1.0, 2.9, 4.0, 5.3$ when 5000 collisions were used in each case.

In the one dimensional model of Eqs. (54) - (58) β and Γ fulfill the same equilibrium condition. Taking β and Γ from Eq. (58) gives the required relationship

$$\beta(1-\Gamma^2) = (m/M)\beta_M = (m/M) (M/2K_B^\ominus) = [m/2K_B^\ominus] = \beta_m$$

(65)

The significance of the parameter Γ can be determined by finding the average velocity, $\langle v' \rangle$, after a collision in a time T

$$\langle v' \rangle = T \int dv' v' W(v|v') = \Gamma v$$

(66)

Therefore, Γ is the ratio of the mean v after a collision to the velocity before a collision. It can also be considered as the fraction of the original velocity that is "remembered" or the "persistence of velocity."

In the case of the one-dimensional model it was found that after a collision

$$v' = [2M/(m+M)]V + [(m-M)/(m+M)]v \quad (67)$$

The mean value of v' is

$$\langle v' \rangle = \int dV W_M(V) v' = [(m-M)/(m+M)]v \quad (68)$$

This direct calculation gives a Γ of $[(m-M)/(m+M)]$ which was obtained in deriving $W(v|v')$ of Eq. (57).

For the three-dimensional case Γ can be calculated exactly for hard sphere collisions from first principles.¹¹ The result given by Chapman and Cowling is

$$\Gamma = [m/(m+M)] + \frac{1}{2}[M/(m+M)] \left\{ x^{-3}(1-2x^2)\text{Erf}(x) - x^{-2}e^{-x^2} \right\} \\ \times \left\{ e^{-x^2} + (2x + x^{-1})\text{Erf}(x) \right\}^{-1} \quad (69)$$

where $x = v/\bar{v}$ with $\bar{v} = [2K_B\theta/M]^{1/2}$.

If $M \gg m$, $x \gg 1$ for most of the range of v and

$$\Gamma \approx m/M \ll 1 \quad (70)$$

This situation corresponds to what is usually called a "strong collision model." For $\Gamma = 0$ in Eq. (59)

$$W(v|v') = (1/T)(\beta/\pi)^{1/2} \exp[-\beta v'^2] \equiv A(v') \quad (71)$$

which is an equilibrium distribution.

When $m \gg M$, $x \ll 1$ for most of the range of v and

$$\Gamma \approx 1 - (3/2)(M/m) \quad (72)$$

or

$$(1 - \Gamma) \ll 1 \quad (73)$$

This case is called the "weak collision model." If the collision operator $J(f)$ is expressed in terms of the kernel of Eq. (59) and expanded to first order in $(1 - \Gamma)$, the Boltzmann equation reduces to a Fokker-Planck diffusion equation.¹²

In intermediate cases Γ depends on v in contrast to the assumption of constant β and Γ following equation (59). For the remainder of the paper it will be assumed that β , Γ , and $(1/T)$ are independent of velocity and that the kernel of Eq. (59) is a reasonable good model for elastic, hard sphere collisions.

VI. Strong Collision Model

When the velocity after a collision is totally independent of the velocity before a collision

$$W(v|v') = A(v')$$

(74)

It is required that the gas approach equilibrium with the passage of time. Therefore, the collision operator in Eq. (23) must vanish at $t = \infty$, giving

$$f(v, \infty)A(v') = f(v', \infty)A(v)$$

(75)

This gives

$$A(v') = cf(v', \infty)$$

(76)

or $A(v')$ is an equilibrium distribution. This was the result obtained in Eq. (71).

VII. Weak Collision Model

If the active atoms are scattered by light perturbing particles, the velocity undergoes significant changes only after many collisions. Section V, Eq. (73) gave $(1-\Gamma) \ll 1$. Expanding the collision integral $J(f)$ of Eq. (23) in a Taylor series in $(1-\Gamma)$ gives¹²

$$\partial f / \partial t + v \partial f / \partial z = \sum_{n=1}^{\infty} (1/n!) [\partial^n / \partial v^n] \{A_n(v) f(v, t)\} \quad (77)$$

where

$$A_n(v) = \int dv' (v-v')^n W(v|v') \quad (78)$$

Using $W(v|v') = (1/T) (\beta/\pi)^{1/2} \exp[-\beta(v'-\Gamma v)^2]$ and keeping only first order terms in $(1-\Gamma)$ gives

$$\partial f / \partial t + v \partial f / \partial z = [(1-\Gamma)/T] \partial / \partial v [v f] + [1/2\beta T] \partial^2 f / \partial v^2 \quad (79)$$

Equation (79) is a Fokker-Planck diffusion equation for Brownian motion.

The kernel $W(v' - \Gamma v)$ of Eq. (59) can be used for a wide range of collisions problems with Γ between 0 and 1.

VIII. Solution of Boltzmann Equation
with Persistence of Velocity

With $W(v|v') = (1/T)(\beta/\pi)^{1/2} \exp[-\beta(v'-\Gamma v)^2]$ the Boltzmann equation, (48), can be written in terms of G_κ (defined in Eq. (30) as the Fourier transform of f) as follows,

$$\begin{aligned} \partial G_\kappa / \partial \tau = & -[(1/T) - i\kappa K v] G_\kappa(v_0|v, \tau) \\ & + (1/T)(\beta/\pi)^{1/2} \int dv' \exp[-\beta(v-\Gamma v')^2] G_\kappa(v_0|v', \tau) \end{aligned} \quad (80)$$

The formal solution of Eq. (80) with the initial condition $G_\kappa(v_0|v, 0) = \delta(v_0 - v)$ is

$$\begin{aligned} G_\kappa(v_0|v, \tau) = & \delta(v_0 - v) \exp[-((1/T) - i\kappa K v)\tau] \\ & + (1/T)(\beta/\pi)^{1/2} \int_0^\tau d\tau' \exp[-((1/T) - i\kappa K v)(\tau - \tau')] \\ & \times \int dv' \exp[-\beta(v - \Gamma v')^2] G_\kappa(v_0|v', \tau') \end{aligned} \quad (81)$$

The expressions for $S^{(1)}(z, t)$ and $S^{(3)}(z, t)$, Eqs. (36) and (37), involve G_κ , the Laplace transform of G_κ (see Eq. (35)). Taking the Laplace transform of both sides of Eq. (81) gives the integral equation

$$\begin{aligned} G_\kappa(v_0|v, \alpha) = & \delta(v_0 - v) [\alpha' - i\kappa K v]^{-1} \\ & + (1/T)(\beta/\pi)^{1/2} [\alpha' - i\kappa K v]^{-1} \\ & \times \int dv' \exp[-\beta(v - \Gamma v')^2] G_\kappa(v_0|v', \alpha) \end{aligned} \quad (82)$$

where $\alpha' = \alpha + (1/T)$

A solution of Eq. (82) can be found by iteration with the following sequence of equations

$$G_{\kappa}^{(0)}(v_0 | v, \alpha) = \delta(v_0 - v) [\alpha' - i\kappa Kv]^{-1} \quad (83a)$$

$$G_{\kappa}^{(N)}(v_0 | v, \alpha) = (1/T)(\beta/\pi)^{1/2} [\alpha' - i\kappa Kv]^{-1} \\ \times \int dv' \exp[-\beta(v - \Gamma v')^2] G_{\kappa}^{(N-1)}(v_0 | v', \alpha) \quad (83b)$$

It can be verified by induction that the solution of (83b) is

$$G_{\kappa}^{(N)}(v_0 | v, \alpha) = [(1/T)(\beta/\pi)^{1/2}]^N [\alpha' - i\kappa Kv_0]^{-1} [\alpha' - i\kappa Kv]^{-1} \\ \times \exp[-(\beta/\Delta_N)(v - \Gamma^N v_0)^2] \\ \times \int \dots \int dv_N \dots dv_2 \prod_{n=2}^N \exp\{-(\beta\Delta_n/\Delta_{n-1})[v_n - (\Gamma v_{n+1} \\ + \Gamma^{n-1} v_0/\Delta_{n-1})(\Delta_{n-1}/\Delta_n)]^2\} [\alpha' - i\kappa Kv_n]^{-1} \quad (84)$$

where $\Delta_n = (1 - \Gamma^{2n})/(1 - \Gamma^2)$; $v_{N+1} = v$ and

$$G_{\kappa}(v_0 | v, \alpha) = \sum_{N=0}^{\infty} G_{\kappa}^{(N)}(v_0 | v, \alpha).$$

For $\kappa = 0$, Eq.(84) simplifies to

$$G_0^{(N)}(v_0 | v, \alpha) = (1/\alpha') [1/(\alpha' T)]^N \exp[-(\beta/\Delta_N)(v - \Gamma^N v_0)^2] \\ \times [\beta/(\pi\Delta_N)]^{1/2} \quad (85)$$

so that

$$\begin{aligned}
 G_0(v_0 | v, \alpha) &= \delta(v_0 - v) / \alpha' \\
 &+ \alpha'^{-1} \sum_{N=1}^{\infty} (\Gamma \alpha')^{-N} [\beta / (\pi \Delta_N)]^{1/2} \exp[-(\beta / \Delta_N) (v - \Gamma^N v_0)^2]
 \end{aligned}
 \tag{86}$$

Equation (86) is identical to the result of Keilson and Störmer¹².

IX. Calculation of Intensity Profile

For both $S^{(1)}(z, t)$ and $S^{(3)}(z, t)$ (Eqs. (36) and (37)), the following integral $R(v')$ is required:

$$R(v') = \int dv_0 W_m(v_0) [\Lambda_a G_0(v_0 | v', \gamma_a') - \Lambda_b G_0(v_0 | v', \gamma_b')] \quad (87)$$

where $W_m(v_0) = [1/(u_m^2 \pi)]^{1/2} \exp[-v_0^2/u_m^2]$ and $u_m^{-2} = \beta_m = \beta(1-\Gamma^2) = u^{-2}(1-\Gamma^2)$.

Using (86), (87) becomes

$$R(v') = \bar{N} W_m(v') \quad (88)$$

where

$$\bar{N} = [(\Lambda_a/\gamma_a) - (\Lambda_b/\gamma_b)] \quad (89)$$

is the unsaturated population inversion of the active medium. The significance of the result given in Eq. (88) is that a gas starting in equilibrium will remain in equilibrium.

Using the solution for G_n (Eq. (84)) to first order in $(1/T)$ for low pressures, in Eq. (36) gives

$$\begin{aligned} S^{(1)}(z, t) = & -\frac{1}{2} \bar{N} (P^2 E/\hbar) \sin Kz \int dv' W_m(v') \int dv \{ \delta(v'-v) [\mu' - iKv]^{-1} \\ & + (1/T) [1/(u^2 \pi)]^{1/2} [\mu' - iKv]^{-1} [\mu' - ikv']^{-1} \\ & \times \exp[-(1/u^2)(v-\Gamma v')^2] \} + c.c. \end{aligned} \quad (90)$$

where $\mu' = \gamma_{ab} - i(\omega - \nu) + (1/T)$

Recognizing the plasma dispersion function⁷

$$Z(\mu', u) = iKu [1/(u^2\pi)]^{1/2} \int dv \exp[-v^2/u^2] [\mu' \pm iKv]^{-1} \quad (91)$$

Eq. (90) reduces to

$$\begin{aligned} S^{(1)}(z, t) = & \frac{-1\bar{N}(\rho^2 E/\hbar) \sin Kz}{2} \{ (iKu_m)^{-1} Z(\mu', u_m) \\ & + (1/T) [1/(u^2\pi)]^{1/2} (iKu)^{-1} \\ & \times \int dv W_m(v) [\mu' - iKv]^{-1} Z(\mu' - i\Gamma Kv, u) \} + c.c. \end{aligned} \quad (92)$$

In the Doppler limit where $[\gamma_{ab}'/(Ku_m)] \ll 1$ the plasma dispersion function is approximately

$$Z(\mu', u) \approx i\pi^{1/2} \exp[-\mu_i'^2/(Ku)^2] - 2i\mu_r'/(Ku) \quad (93)$$

where $\mu_i' = \text{Im}(\mu') = -(\omega - \nu)$

$\mu_r' = \text{Re}(\mu') = \gamma_{ab} + (1/T)$

The expression for $S^{(1)}(z, t)$ in the Doppler limit is then

$$\begin{aligned} S^{(1)}(z, t) = & -\pi^{1/2} \bar{N}(\rho^2 E/\hbar) (Ku_m)^{-1} \sin Kz \{ \exp[-(\omega - \nu)^2/(Ku_m)^2] \\ & \times [1 + \epsilon\pi^{1/2} \exp[-(\omega - \nu)^2(1 - \Gamma)^2/(Ku)^2] \\ & - 2\gamma_{ab}'/(Ku_m) \} \end{aligned} \quad (94)$$

where $\epsilon = (KuT)^{-1}$

The expression for $S^{(3)}(z,t)$, Eq. (37), can be evaluated using similar techniques. To first order in $(1/T)$

$$\begin{aligned}
S^{(3)}(z,t) &= (1/32)(\rho^4 E^3 / \hbar^3) \bar{N} \sin Kz \sum_{\alpha=a,b} (1/\gamma_{\alpha}') (iKu_m)^{-1} \\
&\quad \times [\mu'^{-1} Z(\mu', u_m) + (2\gamma_{ab}')^{-1} (Z(\mu', u_m) + Z(\mu'^*, u_m))] \\
&+ (1/T) (iKu_m)^{-1} (1/\gamma_{\alpha}') \{ (2\mu')^{-1} \int dv W_m(v) [Z(\mu' + i\Gamma Kv, u) \\
&\quad + Z(\mu' - i\Gamma Kv, u)] [(\mu' - ikv)^{-1} + (\mu' + ikv)^{-1}] \\
&\quad + (2\gamma_{ab}')^{-1} \int dv W_m(v) [Z(\mu'^* + i\Gamma Kv, u) + Z(\mu' - i\Gamma Kv, u)] \\
&\quad \quad \quad \times [(\mu' - ikv)^{-1} + (\mu'^* + ikv)^{-1}] \\
&+ 2\gamma_{\alpha}'^{-2} (1/T) (iKu_m)^{-1} \int dv W_m(v) Z(\mu' - i\Gamma Kv, u) (\mu' + ikv)^{-1} \\
&+ 2\gamma_{\alpha}'^{-2} (1/T) (iKu_m)^{-1} \int dv W_m(v) Z(\mu' - i\Gamma Kv, u) (\mu'^* + ikv)^{-1}
\end{aligned}
\tag{95}$$

In the Doppler limit, Eq. (95) reduces to

$$\begin{aligned}
S^{(3)}(z,t) &\approx (1/8) (\rho^4 E^3 / \hbar^3) \bar{N} \pi^{1/2} (\gamma_a \gamma_b)^{-1} \exp[-(\omega - \nu)^2 / (Ku_m)^2] \\
&\quad \times \{ 1 + \mathcal{L}'(\omega - \nu) + \epsilon \pi^{1/2} \mathcal{L}'(\omega - \nu) \left(\exp[-(\omega - \nu)^2 (1 - \Gamma)^2 / (Ku)^2] \right. \\
&\quad \quad \quad \left. + \exp[-(\omega - \nu)^2 (1 + \Gamma)^2 / (Ku)^2] \right) + 2\epsilon \pi^{1/2} \exp[-(\omega - \nu)^2 (1 - \Gamma)^2 / (Ku)^2] \\
&\quad \quad \quad \left. + \epsilon \pi^{1/2} \gamma_a' \gamma_b' (\gamma_a'^{-2} + \gamma_b'^{-2}) \left(\exp[-(\omega - \nu)^2 (1 - \Gamma)^2 / (Ku)^2] \right. \right. \\
&\quad \quad \quad \quad \left. \left. + \exp[-(\omega - \nu)^2 (1 + \Gamma)^2 / (Ku)^2] \right) \right\}
\end{aligned}
\tag{96}$$

where $\mathcal{L}'(\omega-\nu) = \gamma_{ab}'^2 [\gamma_{ab}'^2 + (\omega-\nu)^2]^{-1}$

Taking the projections of $S^{(1)}(z,t)$ and $S^{(3)}(z,t)$ on the cavity mode (this merely eliminates the factor $\sin Kz$ in Eqs. (94) and (96)) and substituting the result into the amplitude equation (3) at steady state ($\dot{E} = 0$) the following equation results:

$$0 = \epsilon_0 E/Q + S^{(1)}(t) + S^{(3)}(t) \quad (97)$$

Define the dimensionless intensity as

$$I(\omega-\nu) \equiv (\rho^2 E^2 / \hbar^2) (\gamma_a \gamma_b)^{-1} \quad (98)$$

and the threshold population inversion density \bar{N}_T as \bar{N} when $I = 0$ and $\omega = \nu$, i.e.,

$$\bar{N}_T = (\epsilon_0/Q) [(\hbar K u_m) / (\rho^2 \pi^{1/2})] [1 + \epsilon \pi^{1/2} - (2\gamma_{ab}') / (\pi^{1/2} K u_m)]^{-1} \quad (99)$$

Let

$$n = \bar{N} / \bar{N}_T \quad (100)$$

To first order in $\epsilon = [KuT]^{-1}$ the intensity of the laser is

$$\begin{aligned}
 I(\omega-\nu) = & 8[(\gamma_a' \gamma_b') / (\gamma_a \gamma_b)] \\
 & \times \left\{ 1 + \epsilon \pi^{1/2} l_- - \exp[(\omega-\nu)^2 / (Ku_m)^2] [(2\gamma_{ab}') / (\pi^{1/2} Ku_m) \right. \\
 & \qquad \qquad \qquad \left. + n^{-1} (1 + \epsilon \pi^{1/2} - \frac{2\gamma_{ab}'}{Ku_m \pi^{1/2}})] \right\} \\
 & \times \left\{ \mathcal{L}'(\omega-\nu) [1 + \epsilon \pi^{1/2} (l_+ + l_-)] + 1 + 2\epsilon \pi^{1/2} l_- \right. \\
 & \qquad \qquad \qquad \left. + \epsilon \pi^{1/2} (\gamma_a' \gamma_b')^{-1} (\gamma_a'^2 + \gamma_b'^2) (l_+ + l_-) \right\}^{-1}
 \end{aligned}$$

$$\text{where } l_{\pm} = \exp[-(\omega-\nu)^2 (1 \pm \Gamma)^2 / (Ku)^2] \quad (101)$$

The frequency of collisions ($1/T$) is directly proportional to the number density of atoms in the laser cavity and is therefore directly proportional to the pressure p . Thus,

$$\epsilon = [KuT]^{-1} = [KuT_1]^{-1} p \quad (102)$$

where ($1/T_1$) is the collision frequency per Torr.

Figure 5 shows a plot of Eq. (101) as a function of $(\omega-\nu)$ for various values of the pressure p . At each pressure the relative excitation n is kept constant.

Figure 6 is a plot of the maximum intensity I_{\max} and the intensity at the central tuning dip I_{dip} for each tuning curve in figure 5 as a function of pressure. The non-linear variation of I_{\max} and I_{dip} with pressure comes

mainly from the coefficient $A = (\gamma_a' \gamma_b') / (\gamma_a \gamma_b)$ in Eq. (101).
 Recalling that $(1/T) = (p/T_1)$,

$$A = (\gamma_a \gamma_b)^{-1} [\gamma_a \gamma_b + 2\gamma_{ab}(p/T_1) + (p/T_1)^2] \quad (103)$$

This increase of laser intensity with pressure comes basically from a reduction of the third order (or saturation) term. An atom gives up energy to the radiation field and then makes a deflecting collision before it can re-absorb any radiation at the same frequency.

If there were no deflecting collisions and only phase changing collisions (See I, Eq. (126)), $(1/T) = 0$ and A becomes

$$A = \gamma_{ab}^{-1} [\gamma_{ab} + \delta_1 p] \quad (104)$$

where δ_1 is the broadening factor per Torr from phase changing collisions (See I, Eq. (144) for definition of $\delta = \delta_1 p$). In that case the maximum intensity would have a linear variation with pressure

Thus, if the tuning curves are measured as in Figure 5 with n constant, the existence and magnitude of the effect of deflecting collisions can easily be determined. It is not expected that the coefficient A (Eq. (103)) will be as simple as the pure velocity changing case, but the major effects of deflecting collisions can nevertheless be discerned.

The detailed features of the tuning dip will not be discussed here. In general, the dip includes the effects of phase changing collisions. The fine structure determined from Eq. (101) will be useful when there are only velocity-changing collisions present. This might be the case in some molecular lasers.

X. High Intensity Theory

Stenholm and Lamb¹³ have developed an extension of the perturbation theory for applications to a high intensity laser. They expressed the polarization as a Fourier series in harmonics of the spatial dependence of a single cavity mode.

Using equations (14a), (14b), and (15) a similar theory can be developed which includes the effects of velocity-changing collisions. The equations of motion of the microscopic polarization and population inversion as defined by Eq. (15) are

$$s(z_0, t, t_0) = -\frac{1}{2}(\rho^2 E/\hbar) \int_{t_0}^t dt' [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] \\ \times \sin[K(z_0 + \int_{t_0}^{t'} v(t) dt)] n(z_0, t', t_0) \quad (105a)$$

$$n(z_0, t, t_0) = [\Lambda_a e^{-\gamma_a(t-t_0)} - \Lambda_b e^{-\gamma_b(t-t_0)}] \\ + \frac{1}{2}(E/\hbar) \int_{t_0}^t dt' [e^{-\gamma_a(t-t')} + e^{-\gamma_b(t-t')}] \\ \times \sin[K(z_0 + \int_{t_0}^{t'} v(t) dt)] s(z_0, t', t_0) \quad (105b)$$

Now express $s(z_0, t, t_0)$ and $n(z_0, t, t_0)$ in a Fourier series in Kz_0

(41)

$$s(z_0, t, t_0) = -i\rho\bar{N} \sum_{n=-\infty}^{n=\infty} s_n(t, t_0) \exp(inKz_0) \quad (106a)$$

$$n(z_0, t, t_0) = \bar{N} \sum_{n=-\infty}^{n=\infty} d_n(t, t_0) \exp(inKz_0) \quad (106b)$$

The microscopic polarization and population inversion at point z in the laser are (see Eq. (12) or (19))

$$s(z, t, t_0) = \int dz_0 \delta(z - z_0 - \int_{t_0}^t v(t) dt) s(z_0, t, t_0) \quad (107a)$$

$$n(z, t, t_0) = \int dz_0 \delta(z - z_0 - \int_{t_0}^t v(t) dt) n(z_0, t, t_0) \quad (107b)$$

Performing the z_0 integration of Eqs. (107) on Eqs. (106) and (105) and substituting the resulting Fourier expansions for $s(z, t, t_0)$ and $n(z, t, t_0)$ into the integral equations gives¹⁴

$$\begin{aligned} & -i \sum s_n(t, t_0) \exp[inK(z - \int_{t_0}^t v(t) dt)] \\ &= -\frac{1}{2}(\rho E/\hbar) \int_{t_0}^t dt' [e^{-\mu(t-t')} + e^{-\mu^*(t-t')}] \sin[K(z - \int_{t'}^t v(t) dt)] \\ & \quad \times \sum d_n(t', t_0) \exp[inK(z - \int_{t_0}^t v(t) dt)] \end{aligned} \quad (108a)$$

$$\begin{aligned}
& \bar{N} \sum d_n(t, t_0) \exp\left[inK\left(z - \int_{t_0}^t v(t) dt\right)\right] \\
& = \left[\Lambda_a e^{-\gamma_a(t-t_0)} - \Lambda_b e^{-\gamma_b(t-t_0)}\right] \\
& \quad - \frac{1}{2}i(\bar{E}N/\hbar) \int_{t_0}^t dt' \left[e^{-\gamma_a(t-t')} + e^{-\gamma_b(t-t')}\right] \sin\left[K\left(z - \int_{t'}^t v(t) dt\right)\right] \\
& \quad \times \sum s_n(t', t_0) \exp\left[inK\left(z - \int_{t_0}^t v(t) dt\right)\right]
\end{aligned} \tag{108b}$$

Take the spatial averages of the left and right hand sides of Eqs. (108) (as per the description of Eqs. (26) and (29) respectively) with respect to the variables

$$\begin{aligned}
\Delta z &= \int_{t_0}^t v(t) dt \\
\Delta z_0 &= \int_{t_0}^{t'} v(t) dt \\
\Delta z' &= \int_{t'}^t v(t) dt
\end{aligned}$$

and integrate over intermediate velocities $v' = v(t')$. Writing the sine function in terms of exponentials,

Eqs. (108) reduce to

$$\begin{aligned}
& \sum s_n(t, t_0) e^{inKz} \int d(\Delta z) f(v_0 | v, \Delta z, t-t_0) e^{-inK\Delta z} \\
& = -\frac{1}{4}(\rho E/\hbar) \int_{t_0}^t dt' \int dv' \left[e^{-\mu(t-t')} + e^{-\mu^*(t-t')}\right] \\
& \quad \times \sum d_n(t', t_0) \int d(\Delta z_0) f(v_0 | v', \Delta z_0, t'-t_0) \int d(\Delta z') f(v' | v, \Delta z', t-t') \\
& \quad \times \left\{ \exp[i(n+1)Kz - inK\Delta z_0 - i(n+1)K\Delta z'] - \exp[i(n-1)Kz - inK\Delta z_0 \right. \\
& \quad \left. - i(n-1)K\Delta z'] \right\}
\end{aligned} \tag{109a}$$

(44)

$$\begin{aligned}
& \sum d_n(t, t_0) G_{-n}(v_0 | v, t-t_0) e^{inKz} \\
& = (1/\bar{N}) [\Lambda_a e^{-\gamma_a(t-t_0)} - \Lambda_b e^{-\gamma_b(t-t_0)}] G_0(v_0 | v, t-t_0) \\
& - \frac{1}{4} (PE/\hbar) \int_{t_0}^t dt' [e^{-\gamma_a(t-t')} + e^{-\gamma_b(t-t')}] \int dv' \sum s_n(t', t_0) \\
& \times G_{-n}(v_0 | v', t'-t_0) \{ e^{iK(n+1)z} G_{-(n+1)}(v' | v, t-t') \\
& \quad - e^{iK(n-1)z} G_{-(n-1)}(v' | v, t-t') \}
\end{aligned} \tag{110b}$$

As in the calculation in the weak signal theory, the macroscopic variables are found by averaging the microscopic variables over all displacements and by integrating over all initial excitation times. Thus,

$$\begin{aligned}
S(v_0 | v, z, t) & = \int_{-\infty}^t dt_0 \int d(\Delta z) f(v_0 | v, \Delta z, t-t_0) \\
& \quad \times \int dz_0 \delta(z-z_0-\Delta z(t, t_0)) s(z_0, t, t_0)
\end{aligned} \tag{111}$$

Applying Eq. (111) to the Fourier series on the left hand sides of Eqs. (110) gives

$$S(v_0 | v, z, t) = \int_{-\infty}^t dt_0 \sum s_n(t, t_0) G_{-n}(v_0 | v, t-t_0) e^{inKz} \tag{112a}$$

Define the averages

$$s_n(v) = \int dv_0 W_m(v_0) s_n(v_0 | v) \quad (116a)$$

$$d_n(v) = \int dv_0 W_m(v_0) d_n(v_0 | v) \quad (116b)$$

over initial velocities v_0 to give

$$s_n(v) = \frac{1}{4}(\rho E/\hbar) \int dv' [d_{n+1}(v') - d_{n-1}(v')] \times [G_{-n}(v' | v, \mu) + G_{-n}(v' | v, \mu^*)] \quad (117a)$$

$$d_n(v) = W_m(v) \delta_{n0} + \frac{1}{4}(\rho E/\hbar) \int dv' [s_{n+1}(v') - s_{n-1}(v')] \times [G_{-n}(v' | v, \gamma_a) + G_{-n}(v' | v, \gamma_b)] \quad (117b)$$

In the absence of collisions, Eq. (83) gives

$$G_{-n}(v' | v, \alpha) = \delta(v' - v) [\alpha + inKv]^{-1} \quad (118)$$

and Eqs. (117) become

$$s_n(v) = \frac{1}{4}(\rho E/\hbar) [d_{n+1}(v) - d_{n-1}(v)] [(\mu + inKv)^{-1} + (\mu^* + inKv)^{-1}]$$

$$d_n(v) = W_m(v) \delta_{n0} + \frac{1}{4}(\rho E/\hbar) [s_{n+1}(v) - s_{n-1}(v)] \times [(\gamma_a + inKv)^{-1} + (\gamma_b + inKv)^{-1}] \quad (119)$$

Equations (119) are essentially equivalent to Eqs (61) of Ref. 13.

From Eq. (117b), with collisions, the normalized population inversion density as a function of velocity is

$$d_0(v) = W_m(v) + \frac{1}{4}(\rho E/\hbar) \int dv' [s_1(v') - s_{-1}(v')] \\ \times [G_0(v'|v, \gamma_a) + G_0(v'|v, \gamma_b)] \quad (120)$$

Expressing $s_1(v)$ and $s_{-1}(v)$ in terms of $d_0(v)$ in Eq. (117) neglecting $d_{\pm 2}(v)$, and substituting back in Eq. (120) gives the "rate equation approximation" for the population inversion

$$d_0(v) = W_m(v) - [(\rho E)/(4\hbar)]^2 \int dv'' d_0(v'') \\ \times [G_{+1}(v''|v', \mu) + G_{+1}(v''|v', \mu^*) + (+1 \rightarrow -1)] \\ \times [G_0(v'|v, \gamma_a) + G_0(v'|v, \gamma_b)] \quad (121)$$

The strong collision model ($\Gamma = 0$) is used in subsequent calculations to most easily illustrate the possibility of using Eq. (121). From Eq. (84) with $\Gamma = 0$

$$G_{\pm 1}(v''|v, \mu) = \delta(v'' - v') [\mu' \mp iKv']^{-1} \\ + (1/T) W_m(v') [\mu' \mp iKv']^{-1} [\mu' \mp iKv'']^{-1} \\ \times [1 + i\epsilon Z(\mu', u_m)]^{-1}$$

$$G_0(v'|v, \gamma_\alpha) = \delta(v' - v) \gamma_\alpha^{-1} + W_m(v) [\gamma_\alpha^{-1} - \gamma_\alpha'^{-1}] \quad (122)$$

Substituting (122) into Eq. (121) gives an integral equation for $d_0(v)$

$$\begin{aligned}
 d_0(v) = & W_m(v) - [(\rho E)/(4\hbar)]^2 d_0(v) \left[\frac{1}{\gamma_a} + \frac{1}{\gamma_b} \right] R(v) \\
 & - [(\rho E)/(4\hbar)]^2 W_m(v) \left[\frac{1}{\gamma_a} + \frac{1}{\gamma_b} - \frac{1}{\gamma_a'} - \frac{1}{\gamma_b'} \right] \\
 & \quad \times \int dv' R(v') d_0(v') \\
 & - [(\rho E)/(4\hbar)]^2 (1/T) W_m(v) \left[\frac{1}{\gamma_a} + \frac{1}{\gamma_b} \right] \int dv' B(v'|v) d_0(v') \\
 & - [(\rho E)/(4\hbar)]^2 (1/T) W_m(v) \left[\frac{1}{\gamma_a} + \frac{1}{\gamma_b} - \frac{1}{\gamma_a'} - \frac{1}{\gamma_b'} \right] \\
 & \quad \times \int dv'' \int dv' W_m(v') B(v'|v'') d_0(v'')
 \end{aligned}
 \tag{123}$$

where

$$R(v) = (2/\gamma_{ab'}) [\mathcal{E}'(\omega - v + Kv) + \mathcal{E}'(\omega - v - Kv)]$$

$$\begin{aligned}
 B(v'|v) = & [1 + ieZ(\mu', u_m)]^{-1} [(\mu' - iKv)^{-1} (\mu' - iKv')^{-1} \\
 & + (\mu' + iKv)^{-1} (\mu' + iKv')^{-1}] + c.c.
 \end{aligned}
 \tag{124}$$

If there are no collisions, the solution of Eq. (123) gives the familiar inhomogeneous saturation of the population inversion density ("hole burning"),

$$d_0(v) = W_m(v) \left[1 + [(\rho E)/(4\hbar)]^2 \left(\frac{1}{\gamma_a} + \frac{1}{\gamma_b} \right) R(v) \right]^{-1} \tag{125}$$

where $(1/T)$ is to be set equal to zero in $R(v)$.

Equation (123) can give some insight into how the velocity profile of the population inversion is modified by collisions. The second term on the r.h.s. of Eq. (123) is like the usual saturation term but smaller by the factor $(\gamma_a'^{-1} + \gamma_b'^{-1})/(\gamma_a^{-1} + \gamma_b^{-1})$. The effect can be attributed to atoms "knocked" out of the velocity v by collisions. The third term then represents the number of atoms "knocked" into velocity v . Smith and Hänsch¹⁵ called this process "cross-relaxation" and obtained an equation containing the above two terms using the rate equation for the atomic populations as the starting point.

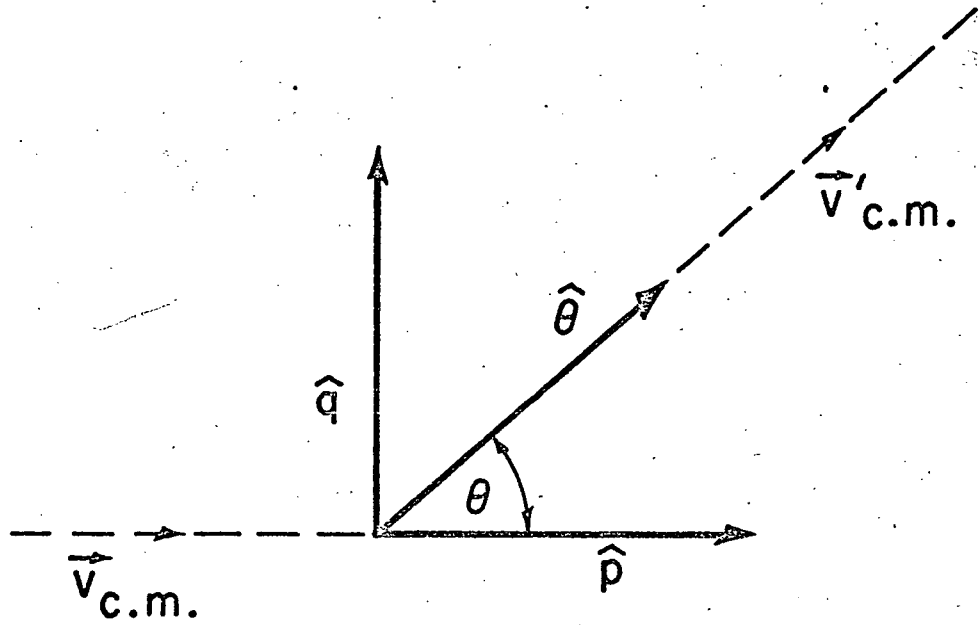
The last two terms on the r.h.s. of Eq. (123), not included in the analysis of Smith and Hänsch, represent removal and addition of atoms with dipole moments. A low intensity solution of Eq. (123) can be obtained by substituting $W_m(v)$ for $d_0(v)$ in the integrals on the r.h.s. Thus, to first order in ϵ and in the Doppler limit,

$$\begin{aligned}
 d_0(v) = & W_m(v) [1 + [(\rho E)/(4h)]^2 (\frac{1}{\gamma_a'} + \frac{1}{\gamma_b'}) R(v)]^{-1} \\
 & \times \{ 1 - 4\epsilon [(\rho E)/(4h)]^2 [(\gamma_a \gamma_a')^{-1} + (\gamma_b \gamma_b')^{-1}] \pi^{1/2} \\
 & \times \exp[-(\omega-v)^2 / (Ku_m)^2] - \epsilon [(\rho E)/(4h)]^2 \pi^{1/2} \\
 & \times R(v) [\frac{1}{\gamma_a'} + \frac{1}{\gamma_b'}] \exp[-(\omega-v)^2 / (Ku_m)^2] \}
 \end{aligned}
 \tag{126}$$

Figure 7 is a plot of $d_0(\nu)$ with $Ku_m = 900$ MHz and $\omega - \nu = Ku_m$. The dominant effect of the collisions is a reduction of the saturation terms. Figure 8 is a plot of $d_0(\nu)$ with $\omega - \nu = 0$. The dashed curve shows $d_0(\nu)$ using the equations of Smith and Hänsch which is obtained by omitting the last term on the r.h.s. of Eq. (126).

Figure Captions

1. Scattering in center of mass (c.m.) frame. Velocity \vec{v}_{cm} is scattered through angle θ and becomes \vec{v}'_{cm} . The unit vector $\hat{\theta}$ is in the direction of \vec{v}'_{cm} ; the unit vector \hat{p} is parallel to \vec{v}_{cm} (and the relative velocity); the unit vector \hat{q} is perpendicular to \vec{v}_{cm} .
2. Intermolecular potentials. The solid curve is a typical intermolecular potential and the dashed curve is a simplified version used to calculate $W(v|v')$. Note that the coefficient B used in the text is Ar_0^6 in the Figure.
- 3a. Numerical result for $W(v_0|v)$ for hard sphere collisions with $(m/M) = 1.0$. Five thousand (5000) numerical collisions were used to obtain this result.
- 3b. Numerical result for $W(v_0|v)$ for hard sphere collisions with $(m/M) = 4.0$. Five thousand (5000) numerical collisions were used to obtain this result.
4. (β_m/β) and Γ as a function of (m/M) for hard sphere collisions based on 5000 encounters.
5. Intensity (I) as a function of detuning $[(\omega-v)/(Ku_m)]$ for various values of the pressure. For this plot, $Ku_m = 5000$ M Hz, $(1/T_1) = 58$ M Hz, $\gamma_a = 17.7$ M Hz, $\gamma_b = 8.3$ M Hz.
6. I_{max} and I_{dip} for tuning curves as a function of pressure. The parameters are the same as in Fig. 5.



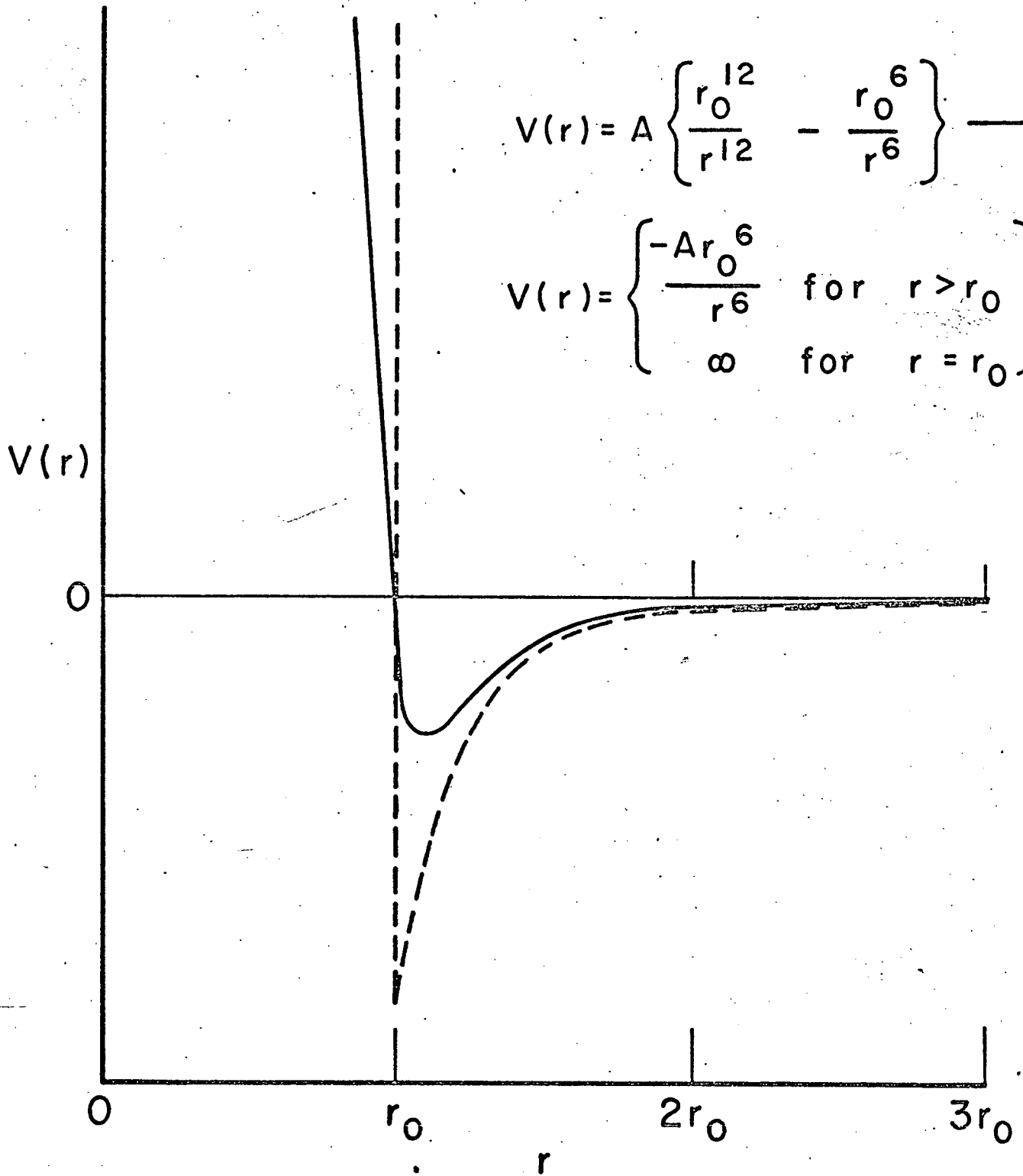
Footnotes

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Figure Captions ...

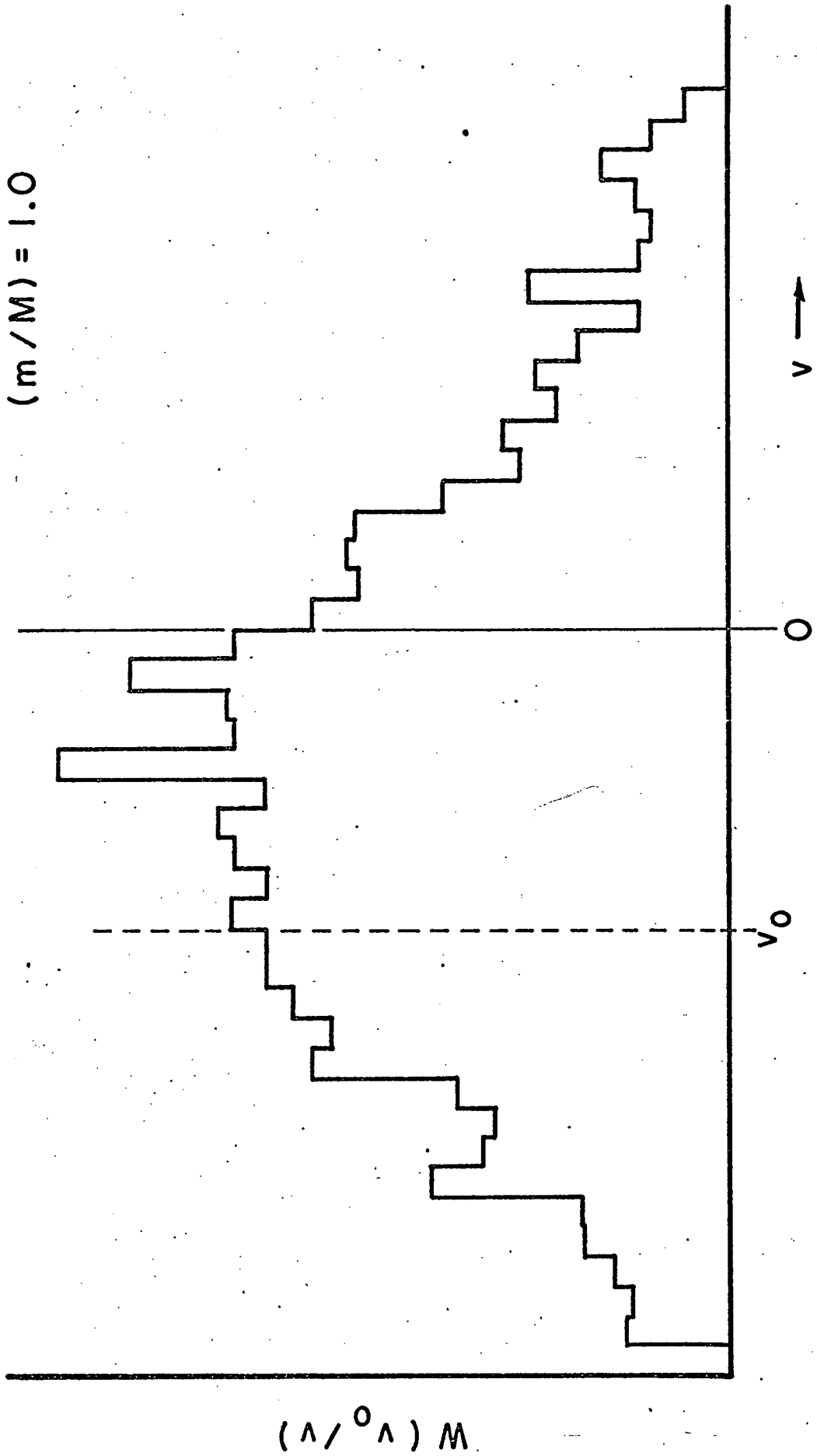
7. d_0 plotted as a function of $[v/(Ku_m)]$ from Eq. (126). The solid curve is the case of no collisions and the dashed curve has $p=.25$ Torr with $\omega-v = Ku_m$. The Doppler width, $Ku_m = 900$ M Hz.

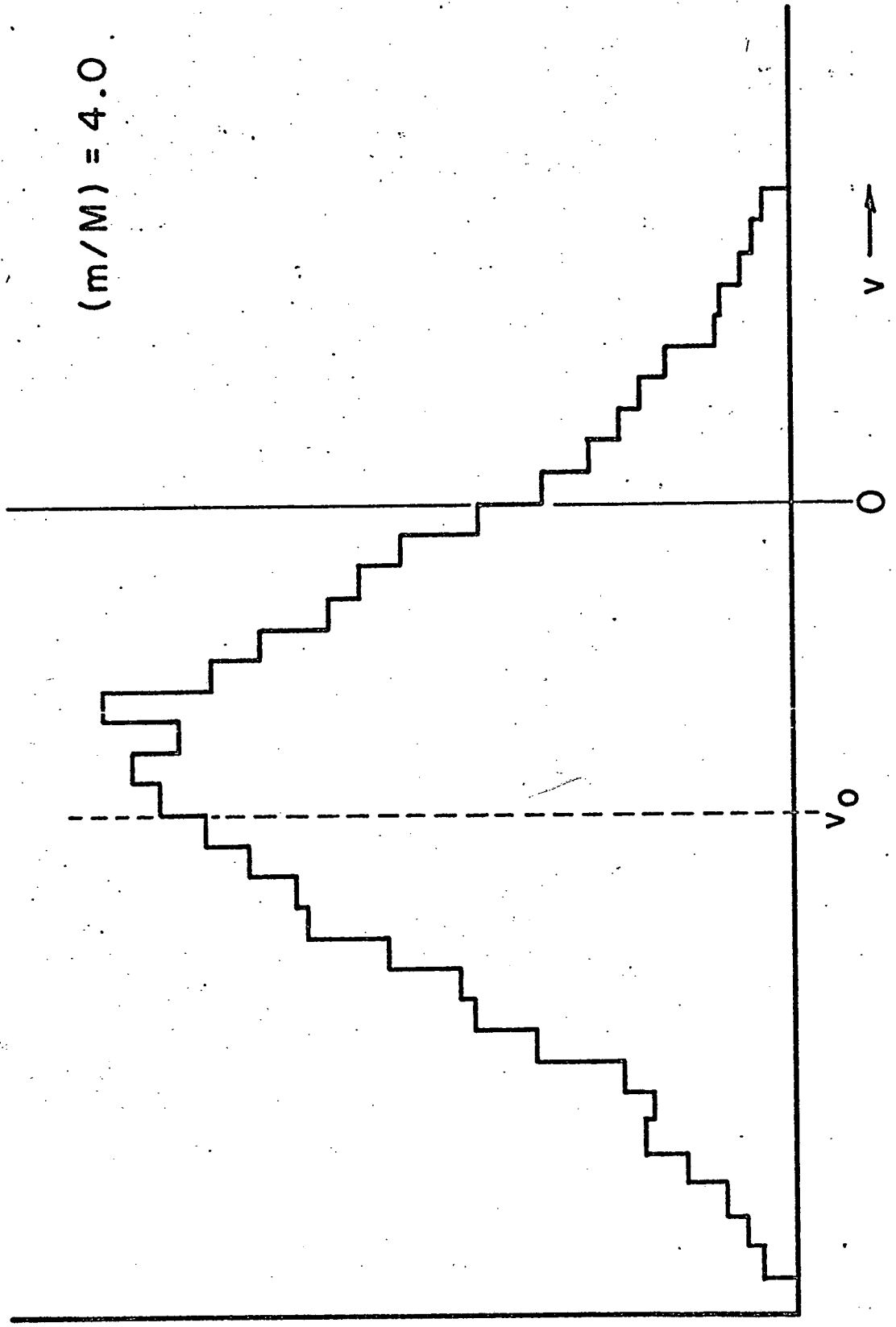
8. d_0 plotted as a function of $[v/(Ku_m)]$ from Eq. (126) with $\omega-v = 0.0$. The top curve is the case of no collisions and the bottom curves have $p = .25$ Torr. The dashed curve is the Smith and Hänsch result. The Doppler width, $Ku_m = 900$ M Hz.



$$V(r) = A \left\{ \frac{r_0^{12}}{r^{12}} - \frac{r_0^6}{r^6} \right\} \text{ ---}$$

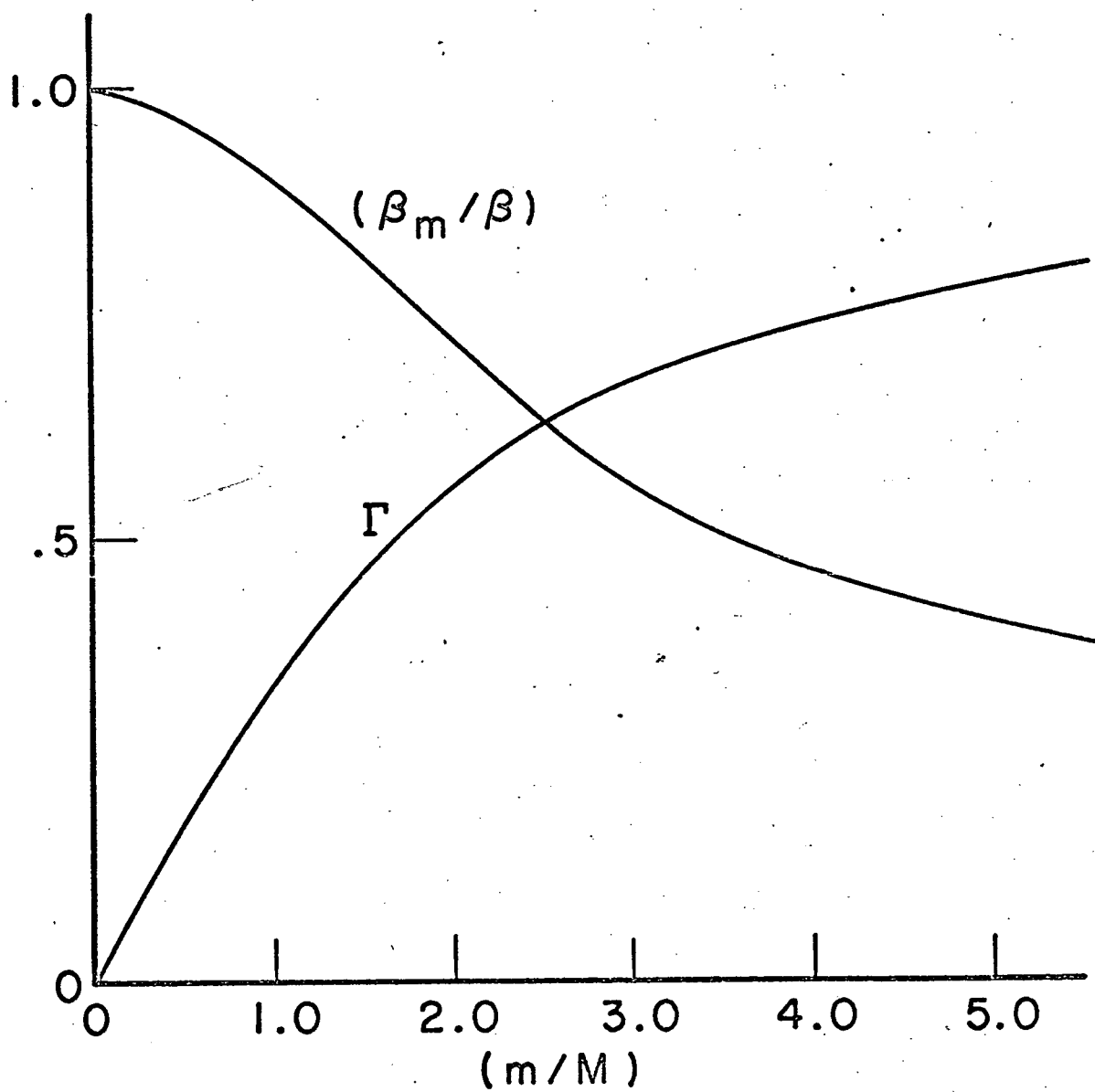
$$V(r) = \left\{ \begin{array}{ll} \frac{-Ar_0^6}{r^6} & \text{for } r > r_0 \\ \infty & \text{for } r = r_0 \end{array} \right\} \text{ ---}$$





$(m/M) = 4.0$

$(v^0 v) M$



C3

