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)Work based on a thesis submitted by M. Borenstein to Yale University in partial fulfillment of the requirements for the Ph. D. degree.

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 $^{13}$ The coefficient on the right-hand side of Eq. (49)  $N\mathcal{S}^2/\hbar$  has the dimensions of number density  $\times$  (dipole moment)<sup>2</sup>/angular momentum. In Eq. (39) for the classical model, the coefficient on the right-hand side can be written in the form  $N(ea)^2/2m(\omega a)a$  where "a" is the length of the pendulum, "ea" is the dipole moment, and  $m(\omega a)a$ is the angular momentum.

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# Effect of Velocity-Changing Collisions on the Output of a Gas Laser\*

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<sup>A</sup> 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. <sup>A</sup> 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.

### 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 paper' gives a comprehensive list of references on this subject.

In a previous publication<sup>2</sup> (I) 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 Javan<sup>3</sup> and Cordover.<sup>4</sup> Other authors have derived similar theoretical expressions. <sup>5</sup>

In Ref. 2, 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 (phasechanging 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 found<sup>6</sup> 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 c.m. 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  $t_c = b_0/v_{\text{rel}}$ , where  $b_0$  is the impact parameter, and  $v_{\tt rel}$  is the relative velocity of emitter and perturber The time between collisions for a typical impact parameter  $b_0$  is approximately  $T = \left( n \pi b_0^2 v_{\rm rel} \right)^{-1}$ , where  $n$  is the number density of perturbers. For pressures of about 1 Torr,  $T \sim 10^{-7}$  sec, while for pressures of about 1 Torr,  $T \sim 10^{-7}$  sec, while for most significant collisions  $t_c$  is less than 10<sup>-11</sup> sec. 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 immediately before the collisions. 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.

Section III 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.<sup>7</sup> 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 singlemode 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 (selfconsistent field). The electric field in the cavity mode 1s

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

and the macroscopic polarization projected on that mode is

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

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

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

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

where  $\Omega$  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  $\gamma_a$ ,  $\gamma_b$ . The active atoms are introduced into the cavity at rates  $\Lambda_a$ ,  $\Lambda_b$ . If the atoms move through the cavity, the position  $z$  at time  $t$  of an atom is given by

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

The integral on the right-hand side 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

$$
\hbar V(z, t) = -\mathcal{C}E(z, t)
$$
  
=  $-\mathcal{C}E(t) \sin\{K[z_0 + \int_{t_0}^t v(\hat{t}) d\hat{t}]\} \cos(\nu t + \varphi)$ , (5)  
where  $\mathcal{C}$  is the electric dipole matrix element

$$
\varphi = e \langle a | x | b \rangle \tag{6}
$$

The equations for the time development of the density matrix  $\rho$  for one atom are

$$
\dot{\rho}_{aa} = -\gamma_a \rho_{aa} + iV(z, t)(\rho_{ab} - \rho_{ba}),
$$
\n
$$
\dot{\rho}_{bb} = -\gamma_b \rho_{bb} - iV(z, t)(\rho_{ab} - \rho_{ba}),
$$
\n
$$
\dot{\rho}_{ab} = -\gamma_{ab}\rho_{ab} - i\omega \rho_{ab} + iV(z, t)(\rho_{aa} - \rho_{bb}),
$$
\n(7)

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

$$
\rho_{ab} = \rho_1 \, e^{-i\nu t} \tag{8}
$$

and neglecting terms with time dependence  $e^{\pm 2i\nu t}$ ,

Eq. (7) can be rewritten as

$$
\dot{\rho}_{aa} = -\gamma_a \rho_{aa} - \frac{1}{2} i (\varphi E/\hbar) \sin Kz (\rho_1 - \rho_1^*),
$$
  
\n
$$
\dot{\rho}_{bb} = -\gamma_b \rho_{bb} + \frac{1}{2} i (\varphi E/\hbar) \sin Kz (\rho_1 - \rho_1^*),
$$

$$
\dot{\rho}_1 = -\left[\gamma_{ab} + i(\omega - \nu)\right]\rho_1 - \frac{1}{2}i(\varphi E/\hbar)\sin\chi_Z(\rho_{aa} - \rho_{bb}),
$$

$$
\dot{\rho}_1^* = -\left[\gamma_{ab} - i(\omega - \nu)\right]\rho_1^* + \frac{1}{2}i\left(\frac{\varphi E}{\hbar}\right)\sinKz\left(\rho_{aa} - \rho_{bb}\right),\,
$$

where z is given by Eq.  $(4)$ .

The initial conditions for Eqs. (9) are  $\rho_{\alpha\beta}(a, z_0, t_0, t_0)=\delta_{\alpha\beta}\delta_{\alpha\alpha}$ .

$$
\rho_{\alpha\beta}(b, z_0, t_0, t_0) = \delta_{\alpha\beta}\delta_{b\alpha} \tag{10}
$$

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

$$
\rho_{aa}(\alpha, z_0, t, t_0) = e^{-\gamma_a (t - t_0)} \delta_{\alpha a} - \frac{1}{2} i (\mathscr{G} E/\hbar) \int_{t_0}^t dt' e^{-\gamma_a (t - t')} \sin{\left[K[\,z_0 + \int_{t_0}^{t'} v(\hat{t}) \, d\hat{t}\,]\right]} [\rho_1(\alpha, z_0, t', t_0) - \rho_1^*(\alpha, z_0, t', t_0)],
$$

$$
\rho_{bb}(\alpha, z_0, t, t_0) = e^{-\gamma_b (t-t_0)} \delta_{\alpha b} + \frac{1}{2} i (\mathscr{C} E/\hbar) \int_{t_0}^t dt' e^{-\gamma_b (t-t')} \sin \{K[z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t}]\} [\rho_1(\alpha, z_0, t', t_0) - \rho_1^*(\alpha, z_0, t', t_0)],
$$

$$
\rho_1(\alpha, z_0, t, t_0) = -\frac{1}{2}i(\varphi E/\hbar)\int_{t_0}^t dt' e^{-i\tau_{ab} + i(\omega - \nu) \cdot \int_0^t t - t'} \sin\{K[z_0 + \int_{t_0}^{t'} v(\hat{t})d\hat{t}]\} [\rho_{aa}(\alpha, z_0, t', t_0) - \rho_{bb}(\alpha, z_0, t', t_0)]
$$
\n(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  $(z, t)$ ; viz.,

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

The symbol  $\langle \ \rangle_{\text{path}}$  in Eq. (12) denotes a statistical average over all collision histories of atoms which start at  $(z_0, t_0)$  with a Maxwell velocity distribution in initial velocity  $v_0$  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

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

$$
s(\alpha, z_0, t, t_0) = -i \vartheta [\rho_1(\alpha, z_0, t, t_0) - \rho_1^*(\alpha, z_0, t, t_0)],
$$
  
(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 S, the out-of-phase part of the polarization of Eq. (2).

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

$$
n(\alpha, z_0, t, t_0) = \left[e^{-\gamma_a(t - t_0)} \delta_{\alpha a} - e^{-\gamma_b(t - t_0)} \delta_{\alpha b}\right] + \frac{1}{2} \left(E/\hbar\right) \int_{t_0}^t dt' \left[e^{-\gamma_a(t - t')} - e^{-\gamma_b(t - t')} \right] \sin\left\{K\left(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t}\right)\right\} s(\alpha, z_0, t', t_0),\tag{14a}
$$

$$
s(\alpha, z_0, t, t_0) = -\frac{1}{2} (\varphi^2 E/\hbar) \int_{t_0}^t dt' \big[ e^{-\Gamma \gamma_{ab} - i(\omega - \nu) \, 1(t - t')} + c.c. \big] \sin \{K[z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t}]\} n(\alpha, z_0, t', t_0).
$$
 (14b)

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

]

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

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

$$
s(z_0, t, t_0) = -\frac{1}{2} \left( \varphi^2 E/\hbar \right) \int_{t_0}^t dt' \sin\{K[z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t} ] \} (e^{-\mu (t-t')}) + e^{-\mu^*(t-t')}) \left( \Lambda_a e^{-r_a(t'-t_0)} - \Lambda_b e^{-r_b(t'-t_0)} \right)
$$

 $(n)$ 

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$$
- \frac{1}{4} \left( \varphi^2 E^2 / \hbar^2 \right) \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \sin\{K[z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t}]\} \sin\{K[z_0 + \int_{t_0}^{t''} v(\hat{t}) d\hat{t}]\} \left( e^{-\mu (t-t')} + e^{-\mu^*(t-t')} \right) \times \left( e^{-\gamma_a (t'-t'')} + e^{-\gamma_b (t'-t'')} \right) s(z_0, t', t_0), \quad (16)
$$

where  $\mu = \gamma_{ab} - i(\omega - \nu)$  and the arguments of  $\Lambda_a$  and  $\Lambda_b$  have been omitted.

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

$$
s^{(1)}(z_0, t, t_0) = -\frac{1}{2} \left(\varphi^2 E/\hbar\right) \int_{t_0}^t dt' \sin\left\{K\left(z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t}\right)\right\} \left(e^{-\mu(t - t')} + e^{-\mu^*(t - t')}\right) \left(\Lambda_a e^{-\gamma_a(t' - t_0)} - \Lambda_b e^{-\gamma_b(t' - t_0)}\right), \tag{17}
$$

and the third-order solution is

$$
s^{(3)}(z_0, t, t_0) = \frac{1}{8} \left( \varphi^4 E^3 / \hbar^3 \right) \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t'} dt''' \sin\{K[z_0 + \int_{t_0}^{t'} v(\hat{t}) d\hat{t}]\} \sin\{K[z_0 + \int_{t_0}^{t''} v(\hat{t}) d\hat{t}]\}
$$
  
×sin  $\{K[z_0 + \int_{t_0}^{t''t'} v(\hat{t}) d\hat{t}]\} \{e^{-\mu(t - t')} + e^{-\mu * (t - t')}\} \{e^{-\gamma} e^{(t' - t'')} + e^{-\gamma} e^{(t' - t'')} \} \{e^{-\mu (t' - t''')} + e^{-\mu * (t' - t''')}\}$ 

$$
\times (\Lambda_a e^{-\gamma_a (t^{\prime\prime\prime}-t_0)} - \Lambda_b e^{-\gamma_b (t^{\prime\prime\prime}-t_0)}).
$$
 (18)

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

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

The  $nth$ -order contribution to the out-of-phase macroscopic polarization is

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

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

$$
S^{(1)}(z,t) = -\frac{1}{2} (\varphi^2 E/\hbar) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \langle \sin\{K[z - \int_{t'}^t v(\hat{t}) d\hat{t}]\} \langle e^{-\mu(t-t')} + e^{-\mu^*(t-t')}\rangle \left(\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)}\right)_{\text{path}},
$$
\n(21)

and the third-order contribution is

$$
S^{(3)}(z,t) = \frac{1}{8} (\vartheta^4 E^3 / \hbar^3) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t'} dt''' \langle \sin \{K [z - \int_{t'}^t v(\hat{t}) d\hat{t} \}] \} \sin \{K [z - \int_{t'}^t v(\hat{t}) d\hat{t} \}] \sin \{K [z - \int_{t'}^t v(\hat{t}) d\hat{t} \}] \times
$$
  
 
$$
\times (e^{-\mu (t-t')} + e^{-\mu * (t-t')}) (e^{-\gamma_a (t'-t'')} + e^{-\gamma_b (t'-t'')} ) (e^{-\mu (t''-t''')} + e^{-\mu * (t''-t''')} ) \times (\Lambda_a e^{-\gamma_a (t'''-t_0)} - \Lambda_b e^{-\gamma_b (t'''-t_0)} ) \rangle_{\text{path}}.
$$
 (22)

I

In Eqs. (21) and (22)  $\Lambda_a$  and  $\Lambda_b$  have been assumed to be constant. When the product of the three sine functions is written in exponential form eight terms will result.

Section IV will deal with path averages.

#### IV. PATH AVERAGES

The calculation of the path average for the firstorder contribution to the polarization must consider the following history for a single active atom.

An atom is excited to state a or b at a time  $t_0$ , at  $z_0$  in the cavity, with random velocity  $v_0$  given by a Maxwell-Boltzmann distribution  $W(v_0)$ . The selection of  $z_0$ 's is restricted to those atoms which arrive at point z at time  $t$  [see Eqs. (12) and (19)]. At a later time  $t'$  at point  $z'$  in the cavity, the atom, which now has velocity  $v'$ , interacts with the cavity electric field and acquires a dipole moment. The

dipole moment of the atom is wanted at a later time t at a point z. The velocity at  $(z, t)$  is v. During the time intervals  $t' - t_0$  and  $t-t'$  the active atom undergoes collisions with foreign-gas atoms that change its z velocity.

To calculate the path average, the conditional probability that an atom with velocity  $v_0$  at point  $z_0$ at a time  $t_0$  goes to z' with velocity v' at t' and to z at time  $t$  with velocity  $v$  is needed. The conditional probability can be written as (see Appendix I of Ref. 2).

$$
f(v_0, z_0, t_0 | v', z', t'; v, z, t)
$$
  
=  $f(v_0, z_0, t_0 | v', z_0 + \Delta z_0, t'; v, z_0 + \Delta z_0 + \Delta z')$ ,  
where  $\Delta z_0 = z' - z_0$  and  $\Delta z' = z - z'$ . (23)

The path average of the first-order term in Eq. (21)

$$
(1) = \sin\{K[z - \int_{t}^{t} v(\hat{t}) d\hat{t}]\} \{e^{-\mu (t - t^*)} + e^{-\mu^* (t - t^*)}\}\
$$

$$
\times (\Lambda_a e^{-\gamma_a (t^* - t_0)} - \Lambda_b e^{-\gamma_b (t^* - t_0)}) \tag{24}
$$

 $is<sup>8</sup>$ 

 $\boldsymbol{\tau}$ 

$$
\langle \mathcal{T}^{(1)} \rangle_{\text{path}} = \int dv_0 W(v_0) \int d\Delta z_0 \int dv' \int d\Delta z' \int dv
$$
  
 
$$
\times f(v_0, z_0, t_0 \mid v', z_0 + \Delta z_0, t'; v, z_0 + \Delta z_0 + \Delta z', t') T^{(1)}.
$$
  
(25)

It has been assumed in Eq. (25) that the medium is spatially homogeneous. Therefore, except for the constraint on  $z_0$  that the atoms arrive at z at time t, the path average only considers the displacements  $\Delta z_0$  and  $\Delta z'$  and should be independent of  $z_0$ .

Using elementary probability theory, the conditional probability in Eq. (23) can be rewritten as the product of two conditional probabilities, i. e. ,

$$
f(v_0, z_0, t_0 | v', z', t'; v, z, t)
$$
  
=  $f(v_0, z_0, t_0 | v', z', t') f(v_0, z_0, t_0; v', z', t' | v, z, t).$   
(26)

Because of the Markoffian character of the statistical process, the second conditional probability on the right-hand side of Eq. (26) reduces to  $f(v', z', t' | v,$  $z, t$ , and Eq. (26) becomes

$$
f(v_0, z_0, t_0 | v', z', t'; v, z, t)
$$
  
=  $f(v_0, z_0, t_0 | v', z', t') f(v', z', t' | v, z, t).$  (27)

If the medium is spatially homogeneous and the statistical process stationary in time, f may be rewritten in the following form:

$$
f(v_0, z_0, t_0 \mid v', z', t') = f(v_0 \mid v, \Delta z_0, t' - t_0),
$$
  
where  $\Delta z_0 = z' - z_0$ . (28)

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

\nUsing Eqs. (24), (27), and (28), Eq. (25) becomes

\n
$$
\langle \tau^{(1)} \rangle_{\text{path}} = \int dv_0 \, W(v_0) \int dv' \int d(\Delta z_0) f(v_0 | v', \Delta z_0, t' - t_0)
$$
\n
$$
\times \int dv \int d(\Delta z') f(v' | v, \Delta z', t - t') \sin(Kz - K\Delta z')
$$
\n
$$
\times (e^{-\mu(t-t')} + e^{-\mu * (t-t')}) (\Lambda_a e^{-\gamma_a(t'-t_0)} - \Lambda_b e^{-\gamma_b(t'-t_0)},
$$

where  $\Delta z' = \int_{t}^{t} v(\hat{t}) d\hat{t}$ .

It is useful to define the quantities

It is useful to define the quantities  

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

so that Eq. (29) becomes

$$
\langle \tau^{(1)} \rangle_{\text{path}} = \int dv_0 \, W(v_0) \int dv' \int dv \, G_0(v_0) \, v', t' - t_0)
$$
  
×[ $e^{iKz} G_{+1}(v'|v, t - t') - e^{-ikz} G_{-1}(v'|v, t - t')]$   
×( $\Lambda_a e^{-r_a(t'-t_0)} - \Lambda_b e^{-r_b(t'-t_0)} (e^{-\mu(t-t')} + e^{-\mu^*(t-t')}).$ \n(31)

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

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

Although the quantities  $v_0$  and  $z_0$  do not appear explicitly in  $T^{(1)}$ , and will later drop out of the problem, it is instructive to include them at this time. The product  $G_0(v_0 | v', t' - t_0) G_{+1}(v' | v, t - t')$  in Eq. (31) clearly shows the history of an atom from  $t_0$ to t.

The same procedure may be used to evaluate the path average for the third-order term. The conditional probability

$$
f(v_0, z_0, t_0 | v''', z''', t'''; v'', z'', t''', v', z', t'; v, z, t)
$$

can be reduced to a product of four f functions of the form  $f(v_1, z_1, t_1 | v_2, z_2, t_2)$ . Equation (22) then becomes

$$
S^{(3)}(z,t) = \left(\frac{1}{32}\right) \left(\frac{\varphi^4 E^3}{\hbar^3}\right) \int_{-\infty}^t dt_0 \int_{t_0}^t dt' \int_{t_0}^{t'} dt' \int_{t_0}^{t'} dt'' \int_{t_0}^{t''} dt''' \langle \mathcal{T}^{(3)} \rangle_{\text{path}}. \tag{33}
$$

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

$$
\langle \mathcal{T}^{(3)} \rangle_{\text{path}} = (1/2i) \int dv_0 \int dv' \int dv' \int dv' \int dv G_0(v_0 | v''' , t''' - t_0)
$$
  
\$\times \{e^{iKz}[G\_{-1} (v''' | v'', t'' - t''') G\_0 (v' | v', t' - t'') G\_{-1} (v' | v, t - t')\$  
\$+ G\_{-1} (v''' | v'', t'' - t''') G\_{-2} (v' | v', t' - t'') G\_{-1} (v' | v, t - t')\$  
\$+ G\_{+1} (v''' | v'', t'' - t''') G\_0 (v' | v', t' - t'') G\_{-1} (v' | v, t - t')] - e^{-iKz}(G\_k + G\_{-k})\$  
\$\times (e^{-\mu(t-t')} + e^{-\mu \* (t-t')})(e^{-\gamma\_a(t'-t'')} + e^{-\gamma\_b(t'-t'')})\$

$$
\times (e^{-\mu(t''-t''')} + e^{-\mu^*(t''-t''')})(\Lambda_a e^{-\gamma_a(t''-t_0)} - \Lambda_b e^{-\gamma_b(t''-t_0)}),
$$
 (34)

where the two terms in  $e^{\pm 3\,i\,Kx}$  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  $G_{\kappa}$  of the  $G_{\kappa}$ 's where

 $(29)$ 

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$$
\mathcal{F}_{\mathbf{k}}(v' \mid v, \alpha) = \int_0^\infty d\tau \ e^{-\alpha \tau} G_{\mathbf{k}}(v' \mid v, \tau).
$$
 (35)

Then

$$
S^{(1)}(z,t) = - (1/4i)(\varphi^2 E/\hbar) \int dv_0 W(v_0) \int dv' \int dv \left[ \Lambda_a S_0(v_0 | v', \gamma_a) - \Lambda_b S_0(v_0 | v', \gamma_b) \right]
$$

$$
\times \left\{ e^{i K z} [S_{-1}(v' | v, \mu) + S_{-1}(v' | v, \mu^*)] - e^{i K z} [S_{+1}(v' | v, \mu) + S_{+1}(v' | v, \mu^*)] \right\} \tag{36a}
$$

and

$$
S^{(3)}(z, t) = (1/64i)(\varphi^4 E^3/\hbar^3) \int dv_0 W(v_0) \int dv' \, ' \left[ \Lambda_a \mathcal{G}_0(v_0) v' \, '', \gamma_a \right] - \Lambda_b \mathcal{G}_0(v_0) v' \, '', \gamma_b) ]
$$
  
 
$$
\times \int dv' \int dv' \int dv' \left[ \int dv' \left[ \mathcal{G}_{+1}(v''' \mid v'', \mu) + \mathcal{G}_{+1}(v''' \mid v'', \mu^*) + \mathcal{G}_{-1}(v'' \mid v'', \mu) + \mathcal{G}_{-1}(v'' \mid v'', \mu^*) \right] \right]
$$
  
 
$$
\times \left[ \mathcal{G}_0(v'' \mid v', \gamma_a) + \mathcal{G}_0(v'' \mid v', \gamma_b) \right] \left[ \mathcal{G}_{-1}(v' \mid v, \mu) + \mathcal{G}_{-1}(v' \mid v, \mu^*) \right] \right] - e^{-iKz} (\mathcal{G}_k + \mathcal{G}_{-k}) . \tag{36b}
$$

I

In the third-order contribution only the terms corresponding to the Doppler limit<sup>7</sup> have been included.

The function  $f(v_0, z_0, t_0, v, z, t)$  can be interpreted as the phase-space probability that an atom at  $(v_0,$  $z_0, t_0$  will go to  $(v, z, t)$  in the time  $t - t_0$ .  $f(v_0, z_0, t)$  $t_0, v, z, t$  may be found by using the Boltzmann equation for the z-component motion of the active atoms

$$
\frac{\partial f}{\partial t} + v_z \frac{\partial f}{\partial z} = J(f) \tag{37a}
$$

where  $J(f)$  is an integral operator describing the collisions. The initial condition for Eq. (37a) is

$$
f(v_0, z_0, t_0 | v, z, t_0) = \delta(v - v_0) \delta(z - z_0).
$$
 (37b)

Section V will be devoted to finding a reasonable collision operator  $J(f)$  so that the expression for  $\mathcal{G}_{\kappa}$ in Eqs. (36a) and (36b) 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  $\bar{v}$  and the velocity of the perturbing atom by  $\overline{V}$  both measured in the laboratory frame.

Viewed in the c.m. system the scattering process changes the velocity of the active atom from  $\vec{v}_{c.m.}$ to  $\bar{v}'_{c,m}$  by rotating it through an angle  $\theta$  (see Fig. 1). The velocity of the active atom in the c.m. system before the collision is given by

$$
\vec{\mathbf{v}}_{\mathbf{c.m.}} = [M/(m+M)] (\vec{\mathbf{v}} - \vec{\mathbf{V}}), \tag{38}
$$

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

$$
\overline{\mathbf{U}} = (m\overline{\mathbf{v}} + M\overline{\mathbf{V}})/(m+M). \tag{39}
$$

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

$$
\vec{\nabla}'_{\mathbf{c.m.}} = [M/(m+M)] |\vec{\nabla} - \vec{\nabla}| \hat{\theta}, \qquad (40)
$$

where the unit vector  $\hat{\theta}$  makes an angle  $\theta$  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

$$
\vec{\nabla}' = \vec{\nabla}'_{\text{c.m.}} + \vec{\vec{U}} \tag{41}
$$

Adding and subtracting  $\bar{v}$  from the right-hand side of Eq.  $(41)$  and using  $(39)$  and  $(40)$ , Eq.  $(41)$  becomes

$$
\vec{\mathbf{v}}' = \vec{\mathbf{v}} + [M/(m+M)][\vec{\mathbf{v}} - \vec{\mathbf{v}} \hat{\boldsymbol{\theta}} - (\vec{\mathbf{v}} - \vec{\mathbf{v}})]. \tag{42}
$$

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 square brackets inEq. (42) onto  $\hat{p}$  and  $\hat{q}$  gives the result

$$
\overline{\mathbf{v}}' = \overline{\mathbf{v}} + [M/(m+M)] \left| \overline{\mathbf{v}} - \overline{\mathbf{v}} \right| \left[ \sin \theta \hat{q} - (1 - \cos \theta) \hat{p} \right] \tag{43}
$$

or

or  
\n
$$
\vec{v}' = \vec{v} - [2M/(m+M)][\sin \frac{1}{2}\theta \hat{p} - \cos \frac{1}{2}\theta \hat{q}]
$$

 $\times \left[\sin^1_{\overline{2}}\theta\left|\vec{v}-\vec{V}\right|\right]$  . (44)

The quantity in the second square bracket in Eq. (44) is a unit vector  $\hat{s}$  making an angle  $\left[\frac{1}{2}(\theta-\pi)\right]$ with the relative velocity  $(\vec{v} - \vec{V})$ , and the quantity in the third square brackets is the inner product  $\hat{s} \cdot (\vec{v} - \vec{v})$ . Equation (44) then becomes



FIG. 1. Scattering in c.m. frame. Velocity  $\bar{\tilde{v}}_{\rm c.m.}$  is scattered through angle  $\theta$  and becomes  $\vec{v}_{c.m.}$ . The uni vector  $\hat{\theta}$  is in the direction of  $\tilde{v}_{\text{c.m.}}$ ; the unit vector  $\hat{p}$  is parallel to  $\overline{v}_{c.m.}$  (and the relative velocity); the unit vector  $\hat{q}$  is perpendicular to  $\bar{v}_{\alpha,m}$ .

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

If the potential between the two atoms is  $U(r)$  then  $\theta$  is given by<sup>9</sup>

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

where

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

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

- $E=\frac{1}{2}\eta|\vec{v}-\vec{V}|^2$  (kinetic energy in c.m. system). (46)
- $J^2 = 2\eta b^2 E$  (square of angular momentum),
- $b =$ impact parameter.

The collision operator in Eq. (37a) in general is<sup>10</sup>

$$
J(f) = \int d\vec{V} \int d\Omega \left| \vec{v} - \vec{V} \right| \sigma \left( \left| \vec{v} - \vec{V} \right|, \theta \right) \times \left[ f(\vec{v}') F(\vec{V}) - f(\vec{v}) F(\vec{V}) \right], \quad (47)
$$

where

 $F(\vec{V}) = N(\beta_{M}/\pi)^{3/2} e^{-\beta_{M}V^2}$ .

[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  $(\vec{v}, \vec{V})$  +  $(\vec{v}', \vec{V}')$  which turns the relative velocity

$$
W(v_z | v'_z) = \int dv_x W_m(v_x) \int dv_y W_m(v_y) \int dv'_x \int dv'_y W(\vec{v} | \vec{v}')
$$
  
=  $\int dv_y W(x) \int dv_y W(x) \int d\vec{v} \int d\vec{v} \cdot d\vec{v} = \vec{v} \cdot d\vec{v} \cdot d\vec{v}$ 

through the angle  $\theta$  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 useable form of  $J(f)$  be obtained. In that situation the product  $|\vec{v} - \vec{V}|$   $\sigma$  depends only on  $\theta$ .

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

$$
\frac{\partial f}{\partial t} + \vec{v} \cdot \vec{\nabla} f = \int d\vec{v}' \left[ W(\vec{v}' | \vec{v}) f(\vec{v}', \vec{r}, t) - W(\vec{v} | \vec{v}') f(\vec{v}, \vec{r}, t) \right]. \tag{48}
$$

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

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

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

For the laser problem, only velocity changes along the cavity axis  $(z \text{ axis})$  will affect the polarization. Therefore, a collision kernel  $W(v, |v')$  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

$$
= \int dv_x W_m(v_x) \int dv_y W_m(v_y) \int d\vec{V} \int d\Omega \left| \vec{v} - \vec{V} \right| \sigma(\left| \vec{v} - \vec{V} \right], \theta) F(\vec{V})
$$
  
 
$$
\times \int dv'_x \int dv'_y \delta(v'_x - v'_x(\vec{v}, \vec{V}, \theta)) \delta(v'_y - v'_y(\vec{v}, \vec{V}, \theta)) \delta(v'_x - v'_x(\vec{v}, \vec{V}, \theta)).
$$
 (50)

Doing the  $v'_x$  and  $v'_y$  integrations gives the expected one-dimensional counterpart of Eq. (49):

$$
W(v_{\boldsymbol{\star}}|\,v_{\boldsymbol{\star}}') = \int dv_{\boldsymbol{\star}}\,W_m(v_{\boldsymbol{\star}})\,\int dv_{\boldsymbol{\star}}\,W_m(v_{\boldsymbol{\star}})\,\int d\vec{\nabla}\int d\Omega\,\left|\,\vec{\nabla} - \vec{V}\,\right|\,\sigma(\left|\,\vec{\nabla} - \vec{V}\,\right|\,,\,\theta)\,F(\vec{V})\,\delta(v_{\boldsymbol{\star}}' - v_{\boldsymbol{\star}}'(\vec{\nabla},\,\vec{V},\,\theta)).\tag{51}
$$

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

$$
v'_{\mathbf{z}}(\vec{\mathbf{v}}, \vec{\mathbf{V}}, \theta) = v_{\mathbf{z}} - \left[2M/(m+M)\right] s_{\mathbf{z}}\left[\hat{s} \cdot (\vec{\mathbf{v}} - \vec{\mathbf{V}})\right]. \tag{52}
$$

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)

$$
U(r) =\begin{cases} -\frac{B}{r^6} & \text{for } r > r_0 \text{ (van der Waals potential)} \\ \infty & \text{for } r = r_0 \text{ (hard-sphere potential)}, \end{cases}
$$
(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  $W(v_{\mathbf{z}}|v_{\mathbf{z}}')$  from Eq. (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  $W(v_*)v'_*)$  for the potential in Eq. (53). Choose and fix  $v_{\ast}$ . The following steps are repeated many times: (i) Choose  $v_x$  and  $v_y$  from a Maxwell velocity distribution. (ii) Choose  $V_x$ ,  $V_y$ ,  $V_x$  from a Maxwell velocity distribution. (iii) Choose impact parameter  $b$  at random in the range  $0 \rightarrow 10^{-6}$  cm. (iv) Calculat  $v'_\text{z}$  from Eqs. (46) and (52) for the potential (53) by integrating the equations of motion. (b) Assign weight  $Nb\Delta b |\vec{v} - \vec{V}|$  {probability of collision per

 $\sqrt{2}$ 



FIG. 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  $Ar_0^3$  in the figure.

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  $\Delta v_{s}$ . For the computor calculation bilis of size  $\Delta v_x$ . For the computer cardial  $B = 4.22 \times 10^{-56}$  erg cm<sup>6</sup> and  $r_0 = 5.0 \times 10^{-8}$  cm.

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

The following discussion of 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 threedimensional case.

Using conservation of energy and momentum we find

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

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

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

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

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

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

After integration Eq. (56) becomes  $(1 + \lambda)$   $(4/(m)(e/\lambda)^{1/2} - 8(n^2 - Fn)^2)$ 

$$
W(v | v') = (1/T)(\beta/\pi)^{-1/2} e^{-\beta \sqrt{v^2 - 2V}}, \qquad (57)
$$

where

$$
\Gamma = [(m-M)/(m+M)]
$$

and

$$
\beta = \beta_M \left[ (m+M)/2M \right]^2. \tag{58}
$$

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

$$
W(v_z | v'_z) = (1/T)(\beta/\pi)^{1/2} e^{-\beta (v'_z - \Gamma v_z)^2} \quad . \tag{59}
$$

As in the one-dimensional problem,  $\beta$  and  $\Gamma$  are functions of the mass ratio  $(m/M)$  (see Fig. 4). In addition  $\beta$  and  $\Gamma$  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  $v<sub>z</sub>$  will be denoted by  $v$  since only one velocity component is under consideration. )

The conditions of equilibrium impose certain restraints on the values of  $\beta$  and  $\Gamma$ . At equilibrium,



FIG. 3. (a) Numerical resultfor  $W\! v_0 \rvert v)$  for hard-spher collisions with  $\langle m/M \rangle = 1.0$ . Five thousand (5000) numerical collisions were used to obtain this result. (b) 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.



FIG. 4.  $(\beta_m/\beta)$  and  $\Gamma$  as a function of  $(m/M)$  for hardsphere collisions based on 5000 encounters.

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

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

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

$$
W_m(v) = (\beta_m/\pi)^{1/2} e^{-\beta_m v^2} \qquad . \tag{61}
$$

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

$$
(1/T) e^{-\beta_m v^2} - (1/T) [\beta/(\beta \Gamma^2 + \beta_m)]^{1/2}
$$
  
× exp[- $\beta \beta_m v^2/(\beta \Gamma^2 + \beta_m)$ ] = 0. (62)

Simplifying Eq. (62) gives

$$
[\beta/(\beta \Gamma^2 + \beta_m)]^{1/2} \exp(-\beta_m v^2 \{ [\beta(1 - \Gamma^2) - \beta_m] / (\beta \Gamma^2 + \beta_m) \} ) = 1 . \quad (63)
$$

Equation (63) is satisfied for every  $v$  only if

$$
\beta(1-\Gamma^2)=\beta_m\quad .\tag{64}
$$

For the collision kernel (59) as obtained by numerical methods, it was found that the quantity  $\left[\beta(1-\Gamma^2)/\beta_m \right]$  ranged from 0.982 to 0.962 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.  $(54)$ – $(58)$  $\beta$  and  $\Gamma$  fulfill the same equilibrium condition. Taking  $\beta$  and  $\Gamma$  from Eq. (58) gives the required relationship

$$
\beta(1 - \Gamma^2) = (m/M)\beta_M = (m/M) (M/2K_B\Theta)
$$

$$
= [m/2K_B\Theta] = \beta_m . \qquad (65)
$$

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

$$
\langle v' \rangle = T \int dv' v' W(v | v') = \Gamma v . \qquad (66) \qquad W(v | v')
$$

Therefore,  $\Gamma$  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 we have

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

The mean value of  $v'$  is

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

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

For the three-dimensional case  $\Gamma$  can be calculated exactly for hard-sphere collisions from first principles.<sup>11</sup> The result given by Chapman and Cowling is

$$
\Gamma = [m/(m+M)] + \frac{1}{2}[M/(m+M)][x^{-3}(1-2x^2)
$$
  
×erf(x) - x<sup>-2</sup>e<sup>-x<sup>2</sup></sup>][e<sup>-x<sup>2</sup></sup>+(2x+x<sup>-1</sup>)erf(x)]<sup>-1</sup>, (69)

where  $x = v/\overline{V}$  with  $\overline{V} = [2K_B \Theta/M]^{1/2}$ .

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

$$
\Gamma \approx m/M \ll 1 \quad . \tag{70}
$$

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

$$
W(v | v') = (1/T)(\beta/\pi)^{1/2} e^{-\beta v' 2} \equiv A(v') \quad , \tag{71}
$$

which is an equilibrium distribution.

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

$$
\Gamma \approx 1 - \frac{3}{2}(M/m) \tag{72}
$$

 $(1 - \Gamma) \ll 1$  . (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.<sup>12</sup>

In intermediate cases  $\Gamma$  depends on  $v$  in contrast to the assumption of constant  $\beta$  and  $\Gamma$  following Eq. (59). For the remainder of the paper it will be assumed that  $\beta$ ,  $\Gamma$ , 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 we find

$$
W(v | v') = A(v') \quad . \tag{74}
$$

or

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

$$
f(v, \infty)A(v') = f(v', \infty)A(v) . \tag{75}
$$

This gives

$$
A(v') = cf(v', \infty) ; \qquad (76)
$$

that is,  $A(v')$  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. (37a) in a Taylor series in  $(1 - \Gamma)$  gives<sup>12</sup>

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

where

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

Using

so that

 $W(v\,|\,v')$  =  $(1/T)(\beta/\pi)^{1\,/\,2}\,e^{-\beta(v'\text{-}\Gamma\,v\,)^{2}}$ 

and keeping only first-order terms in  $(1 - \Gamma)$  gives

$$
\frac{\partial f}{\partial t} + v \frac{\partial f}{\partial z} = \frac{(1 - \Gamma)}{T} \frac{\partial}{\partial v} (vf) + \frac{1}{2\beta T} \frac{\partial^2 f}{\partial v^2} . (79)
$$

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 collision problems with  $\Gamma$  between 0 and 1.

#### VIII. SOLUTION OF BOLTZMANN EQUATION WITH PERSISTENCE OF VELOCITY

With 
$$
W(v | v') = (1/T)(\beta/\pi)^{1/2} e^{-\beta(v' - \Gamma v)^2}
$$
, the Boltz-

\n
$$
G_k
$$
 (definition (48) can be written in terms of  $G_k$  (defined in Eq. (30) as the Fourier transform of  $f$ ] as follows:\n

$$
\frac{\partial G_{\kappa}}{\partial \tau} = -\left(\frac{1}{T} - i\kappa K v\right) G_{\kappa}(v_0 | v, \tau)
$$

$$
+ \frac{1}{T} \left(\frac{\beta}{\pi}\right)^{1/2} \int dv' e^{-\beta (v - \Gamma v')^2} G_{\kappa}(v_0 | v', \tau) . \tag{80}
$$

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

$$
G_{\kappa}(v_0|v,\tau) = \delta(v_0-v) e^{-\frac{(1/T - i\kappa Kv)\tau}{T}} + (1/T)(\beta/\pi)^{1/2} \int_0^{\tau} d\tau' e^{-\frac{(1/T - i\kappa Kv)(\tau - \tau')}{T}} \times \int dv' e^{-\beta(v-\Gamma v')^2} G_{\kappa}(v_0|v',\tau') . \quad (81)
$$

The expressions for  $S^{(1)}(z, t)$  and  $S^{(3)}(z, t)$  [Eqs. (36a) and (36b)] involve  $G_{\kappa}$ , the Laplace transform of  $G_{\kappa}$  [see Eq. (35)]. Taking the Laplace transform of both sides of Eq. (81) gives the integral equation

$$
G_{\kappa}(v_0|v,\alpha) = \delta(v_0 - v)[\alpha' - i\kappa Kv]^{-1}
$$
  
+  $(1/T)(\beta/\pi)^{1/2} [\alpha' - i\kappa Kv]^{-1}$   
 $\times \int dv' e^{-\beta (v-\Gamma v')^2} S_{\kappa}(v_0|v',\alpha)$ , (82)

where  $\alpha' = \alpha + 1/T$ .

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

$$
G_{\kappa}^{(0)}(v_0|v,\alpha) = \delta(v_0 - v)[\alpha' - i\kappa Kv]^{-1},
$$
\n(83a)  
\n
$$
G_{\kappa}^{(N)}(v_0|v,\alpha) = (1/T)(\beta/\pi)^{1/2}[\alpha' - i\kappa Kv]^{-1}
$$
\n
$$
\times \int dv' e^{-\beta(v-\Gamma v')^2} G_{\kappa}^{(N-1)}(v_0|v',\alpha).
$$
\n(83b)

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

 ${}^{C}_{\kappa}(W)[v_{0}]\,v,\,\alpha) = \left[(1/T)(\beta/\pi)^{1/2}\right]^{N}(\alpha'-i\kappa K v_{0})^{-1}(\alpha'-i\kappa K v)^{-1}\,\exp[-\,(\beta/\Delta_{N})(v\,-\,\Gamma^{N}v_{0})^{2}]$ 

$$
\times \int \cdots \int dv_N \cdots dv_2 \prod_{n=2}^N \exp\left\{-\left(\beta \Delta_n/\Delta_{n-1}\right)\left[v_n - (\Gamma v_{n+1} + \Gamma^{n-1}v_0/\Delta_{n-1})(\Delta_{n-1}/\Delta_n)\right]^2\right\} [\alpha'-i\kappa K v_n]^{-1},
$$
 (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[-  $(\beta/\Delta_N)(v - \Gamma^N v_0)^2]$  [ $\beta/(\pi\Delta_N)$ ]<sup>1/2</sup> , (85)

 $\overbrace{\mathbb{G}_0(v_0|v,\alpha) = \delta(v_0-v)/\alpha' + \alpha' \cdot^1 \sum_{N=1}^{\infty} (T\alpha')^{-N}}^{\infty}$  $\times [\beta/(\pi \Delta_N)]^{1/2} \exp[-(\beta/\Delta_N)(v - \Gamma^N v_0)^2]$  . (86)

Equation (86) is identical to the result of Keilson and Störer. $^{12}$ 

# IX. CALCULATION OF INTENSITY PROFILE

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

where

and

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$$
R(v') = \int dv_0 W_m(v_0) [\Lambda_a G_0(v_0) v', \gamma_a')
$$

 ${W}_{m}(v_{0}) = [1/(u_{m}^{2} \pi)]^{1/2} e^{-v_{0}^{2}/u_{m}^{2}}$ 

Using (86), (87) becomes

 $u_m^{-2} = \beta_m = \beta(1 - \Gamma^2) = u^{-2}(1 - \Gamma^2)$ .

$$
R(v') = \int dv_0 W_m(v_0) [\Lambda_a G_0(v_0 | v', \gamma_a'])
$$
 (88)

 $-\Lambda_b S_0(v_0|v',\gamma'_b)$ , (87) where

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

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  $G_k$  [Eq. (84)], to first order in  $(1/T)$  for low pressures, in Eq. (36) gives

$$
S^{(1)}(z,t) = -\frac{1}{2}\overline{N}(\varphi^2 E/\hbar)\sinKz \int dv' W_m(v') \int dv \left\{ \delta(v'-v)[\mu'-iKv]^{-1} + (1/T)[1/(u^2\pi)]^{1/2}[\mu'-iKv]^{-1}[\mu'-iKv']^{-1} \exp[-(1/u^2)(v-\Gamma v')^2] \right\} + c.c. \quad , \quad (90)
$$

where

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

Recognizing the plasma dispersion function

<sup>'sion function'</sup>  
\n
$$
Z(\mu', u) = iKu \left[ 1/(u^2 \pi) \right]^{1/2} \int dv \, e^{-v^2/u^2} [\mu' \pm iKv]^{-1} , \qquad (91)
$$

we find that Eq. (90) reduces to

 $S^{(1)}(z, t) = -\frac{1}{2} \overline{N} (\vartheta^2 E/\hbar) \sin Kz \left\{ (i K u_m)^{-1} Z(\mu', u_m) + (1/T) [1/(u^2 \pi)]^{1/2} (i K u)^{-1} \right\}$  $\times \int dv W_{m}(v)[\mu' - iKv]^{-1} Z(\mu' - i\Gamma Kv, u) + c.c.$  (92)

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

$$
Z(\mu', u) \approx i\pi^{1/2} e^{-\mu'_i{}^2/(Ku)^2} - 2i\mu'_r/(Ku) \quad , \tag{93}
$$

where

 $\mu'_i = \text{Im}(\mu') = -(\omega - \nu)$ ,

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

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

$$
S^{(1)}(z,t) = -\pi^{1/2} \overline{N} (\vartheta^2 E/\hbar) (K u_m)^{-1} \sin K z \left\{ e^{-(\omega - \nu)^2 / (K u_m)^2} \left[ 1 + \epsilon \pi^{1/2} e^{-(\omega - \nu)^2 (1-\Gamma)^2 / (K u)^2} \right] - 2 \gamma_{ab}^{\prime} / (K u_m) \right\} , \qquad (94)
$$
  
where  $\epsilon = (K u T)^{-1}$ .

The expression for  $S^{(3)}(z, t)$  [Eq. (36b)] can be evaluated using similar techniques. To first order in  $(1/T)$  we find

$$
S^{(3)}(z,t) = \frac{1}{32} \left( \varphi^4 E^3 / \hbar^3 \right) \overline{N} \sinKz \sum_{\alpha = a,b} (1/\gamma_{\alpha}')(iKu_m)^{-1} \left\{ \mu'^{-1} Z(\mu', u_m) + (2\gamma_{ab}')^{-1} [Z(\mu', u_m) + Z(\mu'^*, u_m)] \right\}
$$
  
+  $(1/T)(iKu_m)^{-1} (1/\gamma_{\alpha}')(2\mu')^{-1} \int dv W_m(v) [Z(\mu' + i\Gamma Kv, u) + Z(\mu' - i\Gamma Kv, u)][(\mu' - ikv)^{-1} + (\mu' + iKv)^{-1}]$   
+  $(2\gamma_{ab}')^{-1} \int dv W_m(v) [Z(\mu'^* + i\Gamma Kv, u) + Z(\mu' - i\Gamma Kv, u)][(\mu' - iKv)^{-1} + (\mu'^* + iKv)^{-1}]$   
+  $2\gamma_{\alpha'}'^{-2} (1/T)(iKu_m)^{-1} \int dv W_m(v) Z(\mu' - i\Gamma Kv, u)(\mu' + iKv)^{-1}$   
+  $2\gamma_{\alpha'}'^{-2} (1/T)(iKu_m)^{-1} \int dv W_m(v) Z(\mu' - i\Gamma Kv, u)(\mu'^* + iKv)^{-1}$  (95)

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

$$
S^{(3)}(z,t) \approx (1/8)(\mathcal{P}^{4}E^{3}/\hbar^{3})\overline{N}\pi^{1/2}(\gamma_{a}\gamma_{b})^{-1}e^{-(\omega-\nu)^{2}/(K u_{m})^{2}}[1+\mathcal{L}'(\omega-\nu)+\epsilon\pi^{1/2}\mathcal{L}'(\omega-\nu)\n\n\times (e^{-(\omega-\nu)^{2}(1-\Gamma)^{2}/(K u)^{2}}+e^{-(\omega-\nu)^{2}(1+\Gamma)^{2}/(K u)^{2}})+2\epsilon\pi^{1/2}e^{-(\omega-\nu)^{2}(1-\Gamma)^{2}/(K u)^{2}}\n\n+\epsilon\pi^{1/2}\gamma_{a}'\gamma_{b}'(\gamma_{a}'^{-2}+\gamma_{b}'^{-2})(e^{-(\omega-\nu)^{2}(1-\Gamma)^{2}/(K u)^{2}}+e^{-(\omega-\nu)^{2}(1+\Gamma)^{2}/(K u)^{2}})],
$$
\n(96)

$$
\mathcal{L}'(\omega-\nu)=\gamma'^{2}_{ab}\left[\gamma'^{2}_{ab}+(\omega-\nu)^{2}\right]^{-1}.
$$

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 (8) at steady state  $(\dot{E}= 0)$ , we find that the following equation results:  $\mathfrak{N} = \overline{N}/\overline{N}_T$ . (100)

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

Define the dimensionless intensity as

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

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

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

Let

$$
\mathfrak{N} = \overline{N}/\overline{N}_T \ . \tag{100}
$$

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

$$
I(\omega - \nu) = 8[(\gamma_a' \gamma_b')/(\gamma_a \gamma_b)] \{1 + \epsilon \pi^{1/2} l_- - e^{(\omega - \nu)^2 / (K u_m)^2} [(2\gamma_{ab}')/(\pi^{1/2} K u_m) + \mathfrak{N}^{-1} (1 + \epsilon \pi^{1/2} - 2\gamma_{ab}' / K u_m \pi^{1/2})]\}
$$
  
 
$$
\times \{\mathfrak{L}'(\omega - \nu)[1 + \epsilon \pi^{1/2} (l_+ + l_-)] + 1 + 2\epsilon \pi^{1/2} l_- + \epsilon \pi^{1/2} (\gamma_a' \gamma_b')^{-1} (\gamma_a'^2 + \gamma_b'^2)(l_+ + l_-)\}^{-1}, \quad (101)
$$

where

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

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, we have

$$
\epsilon = (KuT)^{-1} = (KuT_1)^{-1}p, \qquad (102)
$$



FIG. 5. Intensity  $(I)$  as a function of detuning  $[(\omega - \nu)/$  $(Ku_m)$  for various values of the pressure. For this plot,  $Ku_m = 5000 \text{ MHz}, (1/T_1) = 58 \text{ MHz}, \gamma_a = 17.7 \text{ MHz}, \text{ and } \gamma_b$  $= 8.3$  MHz.

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  $\dot{p}$ . At each pressure the relative excitation  $\mathfrak K$  is kept constant.

Figure 6 is a plot of the maximum intensity  $I_{\text{max}}$ and the intensity at the central tuning dip  $I_{div}$  for each tuning curve in Fig. 5 as a function of pressure. The nonlinear variation of  $I_{\text{max}}$  and  $I_{\text{dip}}$  with pressures comes mainly from the coefficient  $A = (\gamma_a' \gamma_b')/(\gamma_a \gamma_b)$  in Eq. (101). Recalling that  $(1/T)$  $=(p/T_1)$ , we obtain



FIG. 6.  $I_{\text{max}}$  and  $I_{\text{dip}}$  for tuning curves as a function of pressure. The parameters are the same as in Fig. 5.

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

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 reabsorb any radiation at the same frequency.

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

$$
A = \gamma_{ab}^{-1} \left[ \gamma_{ab} + \delta_1 \, \rho \right] \,, \tag{104}
$$

where  $\delta_1$  is the broadening factor per torr from phase-changing collisions [see paper I, Eq.  $(144)$ , for definition of  $\delta = \delta_1 p$ . In that case the maximum

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intensity would have a linear variation with pressure. Thus, if the tuning curves are measured as in

Fig. 5 with  $\mathfrak X$  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 velocitychanging 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 shows 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.

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Pleochroic Halos and the Constancy of Nature: A Reexamination

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A thorough reanalysis of the early measurements of pleochroic halos in light of today's standards and knowledge indicates that, contrary to widespread belief, these halos do not provide proof that the laws of radioactive decay are constant in time.

One of the great bedrocks of physics and astronomy is the belief that physical processes are invariant over cosmologically long periods of time. Despite the fundamental importance of this belief, there is extraordinarily little direct evidence of its validity.

Qne very familiar piece of evidence comes from the examination of the identifiable spectral lines of distant galaxies. The frequencies of these lines appear to have been the same at the time of their

creation (billions of years ago) as the frequencies of equivalent lines created today in terrestrial laboratories. Qbservable differences between these two sets are attributed (almost certainly correctly) to the Doppler red shift caused by galactic recession.

There is another piece of evidence often cited, considerably less familiar however, which has been around for over  $60 \text{ yr}$ .<sup>1</sup> This is the information obtained from geological phenomena known as