Theory of Collision Effects on Line Shapes Using a Quantum-Mechanical Description of the Atomic Center-of-Mass Motion–Application to Lasers. II. The Pseudoclassical Collision Model and Steady-State Laser Intensities*

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Pressure effects in gas lasers are studied within the framework of a theory that treats the center-of-mass motion of the active and perturber atoms quantum mechanically. Such a theory considers the perturber-induced energy-level variations and the velocity changes of the active atom caused by collisions on an equal basis. The calculation is carried out assuming that the active atom undergoes many binary collisions before it decays. As such, this paper represents a generalization of our previous work, which was valid only if the active atom underwent, on the average, at most one collision in its lifetime. The transition from the "one-collision" to the "many-collision" domain is achieved by use of a pseudoclassical collision model. This model enables one to incoporate the results of a rigorous quantum-mechanical treatment of the one-collision problem into a standard classical procedure for handling many (binary) collisions. The result is a quantum-mechanical theory of collision effects valid in the many-(binary-)collision pressure region. Using this theory, expressions for the laser intensity profile are derived employing a collision model which, although quite elementary, should prove to be reasonably accurate for lasers operating slightly above threshold at pressures where both the collision-broadened linewidth and cavity detuning are much smaller than the Doppler width associated with the laser transition. A comparison of theory and experiment is made, and the significance of their relatively good agreement is discussed.

I. INTRODUCTION

In a previous paper[1] (hereafter referred to as QMI) we discussed the problem of an atom interacting with a radiation field and undergoing collisions with perturber atoms. Collisions will, in general, produce variations in the energy levels of the active (emitting or absorbing) atom and may also alter the atom's velocity. It was pointed out in QMI that if one hopes to consistently incorporate these collision effects into the radiation problem, it becomes necessary to use a quantum-mechanical description for the center-of-mass motion of the active atom.

Employing such a method in QMI, we solved a laser problem to first order in the field and obtained the gain and frequency pulling parameters of the laser. The calculation was limited to very low pressures since it was assumed that the active atoms underwent at most one collision in their lifetime. With this restriction, however, the results were applicable to absorption as well as stimulated emission experiments. The major purpose of this work is to extend the calculation of QMI to third order in the laser field and increase its applicability to higher-pressure regions. Working to third order in the laser field will enable us to obtain an expression for the steady-state laser intensity as a function of cavity detuning which, as pointed out in Sec. I of QMI, may provide a sensitive probe of the collision effects.

To perform the present calculation, we shall use the pseudoclassical collision model (Sec. III) which was first introduced in Sec. V of QMI. The model is based upon an interpretation of the quantum-mechanical results in terms of independently occurring Doppler-modifying and phase-shifting collisions and offers a simple alternative to the laborious use of time-dependent perturbation theory in QMI. (Doppler-modifying and phase-shifting collisions will be defined in Sec. III. The former are related to velocity changes caused by collisions and the latter to the perturber-induced energy-level variations of the active atom produced in collisions.) The pseudoclassical model is believed to be valid in the binary-collision pressure region but only for the case in which one of the two radiative states involved in the laser transition experiences a much stronger collision interaction than the other. Thus, this paper will be directed towards situations where only one of the radiative levels experiences a collision interaction and, unless noted to the contrary, the text refers specifically to this case. We do feel that the above condition is closely approximated in many physical systems.[2] To treat the case where the collision interaction in the two radiative levels is comparable, we offer a calculational model in Appendix A.

In Secs. II and III we review QMI and outline the philosophy of the present calculation, including a detailed exposition of the pseudoclassical model in Sec. III. The mathematical development of the problem is presented in Secs. IV–VI, with expressions for the gain, linear pulling, saturation, and nonlinear pushing parameters of the laser, as well

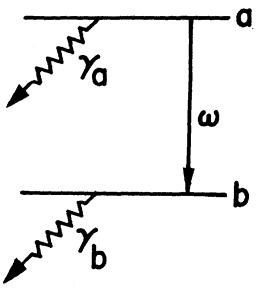

FIG. 1. The two states a and b involved in the laser transition are assumed to decay to some lower states not shown in the figure with rates γ_a and γ_b, respectively. The frequency difference between the two levels is denoted by ω.

as the steady-state field intensity, given in Sec. VI. In Sec. VII, we discuss the general properties of our solutions and compare our findings with experimental results. An evaluation of the theory is given in Sec. VIII.

It will prove useful (although not absolutely necessary) to be familiar with Secs. I–IV and IX and the beginning of Sec. V of QMI before proceeding. Equations referred to from QMI will be prefixed by a "I."

II. REVIEW OF QMI AND PHILOSOPHY OF CALCULATION

It will prove helpful to briefly review the calculation of QMI. The basic problem was to follow the time development of a moving atom which at $\hat{t} = 0$ was excited to the upper laser state a shown in Fig. 1 and then permitted to interact with both the external laser electric field and several stationary perturbers. The initial condition chosen for the active atom had to be consistent with the assumed macroscopic properties of the gas—namely, an isotropic spatial distribution of excited atoms characterized by some velocity distribution $W_a(\vec{v}_0)$. We found the most convenient choice of initial condition for the active atom, leading to the above properties, to be a plane-wave state of definite momentum $\vec{p}_0$ normalized to the laser volume. The velocity $\vec{v}_0 = \vec{p}_0/m$ that one associates with the plane-wave state must be weighted according to the distribution $W_a(\vec{v}_0)$.

Having decided on an initial condition, we proceeded to solve the Schrödinger equation for the problem by time-dependent perturbation theory to first order in the laser field and to lowest order in the collision interaction at each individual perturber site. We then generalized this result to all orders in the collision interaction at each perturber site by use of an assumption discussed in Sec. V of QMI. The result we obtained was valid only if the active atom made at most one collision in its lifetime, since our treatment did not allow for successive interactions at different perturber sites. Additional approximations of a less restrictive nature given in Sec. II of QMI are retained in this work. For convenience, the negligible photon recoil terms, included in QMI, will be neglected here.

The purpose of this paper is to generalize the results of QMI to (a) allow for moving perturbers, (b) work to third order in the laser field, and (c) allow for the active atom to undergo many binary collisions in its lifetime. A schematic form of the chain of reasoning used to accomplish this generalization is pictured in Fig. 2 and will be discussed below.

Although noted only briefly in QMI, it is possible to extend the quantum-mechanical calculation given there to the case of moving perturbers by taking each perturber atom to be initially in a plane-wave state [with its velocity determined by some distribution $W_p(\vec{v}_p)$] rather than fixed in the medium. The only difference from the stationary perturber results is that the line-shape parameters (i.e., widths, shifts, etc.) are modified—the form of the results of QMI [see especially Eqs. (I89) and (I90)] remains the same. In this paper, we shall give, without derivation, the line-shape parameters for the case of moving perturbers. They will appear as a somewhat obvious generalization of the stationary perturber results.

We have also extended the results of QMI by carrying out the time-dependent perturbation calcula-

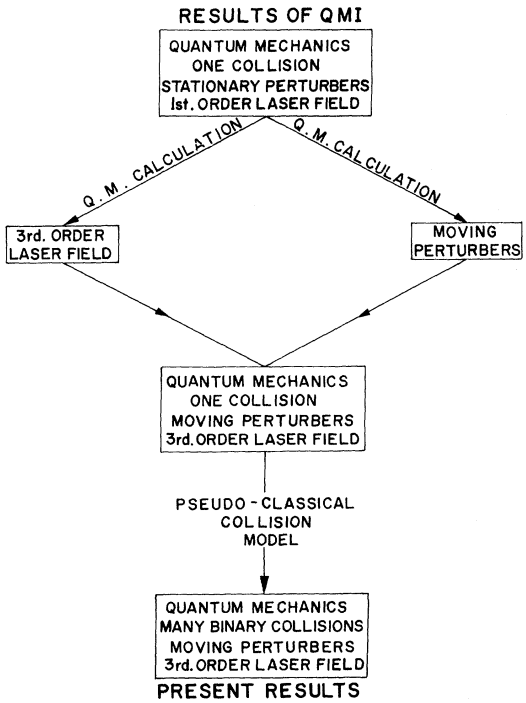

FIG. 2. Schematic representation of the chain of calculations leading from the results of QMI to the present results.

tion to third order in the laser field, thus providing a rigorous quantum-mechanical expression in this one-collision stationary perturber limit. Since the generalization to allow for moving perturbers is not directly dependent on the laser field, it is reasonable to assume that we may combine the moving perturber generalization of first-order laser theory with the stationary perturber third-order laser field calculation to arrive at a result valid to third order in the laser field for an active atom which undergoes at most one collision in its lifetime with a moving perturber atom.

Finally, we wish to remove the one-collision restriction and allow the active atoms to undergo many (binary) collisions in their lifetimes. In general, this will increase the region of perturber pressures for which the theory is applicable from about 0.2 Torr to a few hundred Torr, and it will certainly encompass normal laser operating pressures ($\approx 1-10$ Torr). To include the many-collision effects, one could attempt to continue with the time-dependent perturbation theory of QMI, but this approach would prove tedious and difficult.[3] Instead, we propose to extend the results of QMI by use of the pseudoclassical collision model (PCM) which was first introduced in Sec. V of QMI and will be described more fully in Sec. III. The PCM will enable us to easily incorporate binary-collision effects into the no-collision quantum-mechanical results. Based on our confidence in the PCM, we shall obtain a quantum-mechanical solution of the laser problem to third order in the laser field for the case of moving perturbers that is valid in the binary-collision pressure region.

We cannot offer direct proof that the PCM is correct. The only guarantee which may be given is that it will reduce to the above-mentioned quantum-mechanical result in the one-collision stationary perturber limit, and that it will also reduce to well-known impact theory limits[4,5] if one neglects velocity-changing collisions. In addition, we hope to develop the PCM in a manner that will attach some type of diagrammatic basis to the model and, hopefully, make it easier to accept. This diagrammatic basis is no longer obvious if there is a comparable collision interaction for both radiative levels. For this reason, we shall discuss only single-state (a or b) scattering in this paper. However, to include the possibility of collision interaction in both radiative states, we present a calculational model in Appendix A extending the two-level scattering result of QMI to the binary-collision pressure region. Since this calculational model lacks the diagrammatic basis which the PCM possesses, our confidence in it is somewhat reduced, although it does seem to provide reasonable results.[6]

Before discussing the PCM, we should like to add one additional note. The final results that will be obtained will depend critically on the quantum-mechanical (or corresponding classical) line-shape parameters. The evaluation of these parameters is far from trivial and, in fact, represents a significant problem in its own right. It is not the purpose of this paper to provide a sophisticated determination of these parameters, and we shall be content with examining the general properties of our solutions.

III. PSEUDOCLASSICAL COLLISION MODEL

The pseudoclassical collision model (PCM) permits one to use classical collision concepts within the confines of a quantum-mechanical theory. In order to provide the framework for the model, we shall proceed as follows.

(A) First, the quantum-mechanical solution of the laser problem with no collisions as developed in QMI is reviewed.

(B) Second, the effect of a classical collision on the active atom is discussed.

(C) Third, the one-collision quantum-mechanical solution of QMI is recalled and its relation to the classical picture of a collision is noted.

(D) Finally, the discussions of (A)–(C) are combined to produce the PCM.

A. Quantum-Mechanical Solution with No Collisions

In QMI, we found a quantity of interest in laser problems to be the polarization function density $A(\vec{R}, t, \vec{v}_0)$ which was related to the off-diagonal density matrix element of the atom by [see Eq. (I10)]

$$A(\vec{R}, t, \vec{v}_0) = \rho_{ab}(\vec{R}, t, \vec{v}_0)\, e^{i[\Omega t + \varphi(t)]}\,, \tag{1}$$

where a and b are the laser states depicted in Fig. 1, $\vec{v}_0$ is the velocity of the atom at its time of excitation, Ω is the laser cavity frequency, $\vec{R}$ is the center-of-mass coordinate of the atom, and $\varphi(t)$ is a slowly varying phase. One may recall that $\rho_{ab}(\vec{R}, t, \vec{v}_0)$ is the quantum-mechanical generalization of the atomic density matrix element $\rho_{ab}(t, \vec{v}_0)$ and that $\rho_{ab}(\vec{R}, t, \vec{v}_0)$ is, in effect, the amount of $\rho_{ab}(t, \vec{v}_0)$ associated with the volume d^3R [that is, $\int d^3R\, \rho_{ab}(\vec{R}, t, \vec{v}_0) = \rho_{ab}(t, \vec{v}_0)$]. In practice, one will have to solve for $\rho_{ab}(\vec{R}, t, \vec{v}_0)$, use Eq. (1) to obtain $A(\vec{R}, t, \vec{v}_0)$, and then project it onto the laser cavity mode [see Eq. (I13)] to find the polarization function

$$A(t, \vec{v}_0) = (2/V) \int d^3R\, A(\vec{R}, t, \vec{v}_0) \sin(\vec{k} \cdot \vec{R})\,, \tag{2}$$

where V is the laser volume and $\vec{k}$ is the propagation vector associated with the laser field. The polarization function is then used in the amplitude and phase equations of laser theory [Eqs. (I19) and (I21)] to obtain characteristic laser parameters.

To solve for $\rho_{ab}(\vec{R}, t, \vec{v}_0)$ in QMI, we worked in the

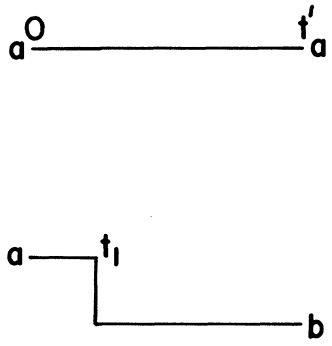

FIG. 3. The only type of diagram leading to a contribution for $\rho_{ab}(\vec{R}, t', \vec{v}_0)$ to first order in the laser field. The two lines can be thought of as representing two possible Feynman world lines or histories for the atomic state amplitudes. Density matrix elements are obtained at any time $\hat{t}$ by forming the product of the amplitude represented by the upper line with the complex conjugate of the amplitude represented by the lower one at time $\hat{t}$. Both amplitude histories begin with the atom excited to state a with velocity $\vec{v}_0$ at $\hat{t}=0$. While the upper line represents a history with no interaction with the laser field, the lower one indicates an interaction with the field at time t_1 taking the state amplitude from a to b. Note that one may interpret this diagram to imply that only $\rho_{aa}(\vec{R}, \hat{t}, \vec{v}_0) \neq 0$ for times between $\hat{t}=0$ and $\hat{t}=t_1$, while only $\rho_{bb}(\vec{R}, \hat{t}, \vec{v}_0) \neq 0$ for times between $\hat{t}=t_1$ and $\hat{t}=t'$. The value of $\rho_{ab}(\vec{R}, t', \vec{v}_0)$ to first order in the field is obtained by summing all such diagrams (i.e., by integrating over all t_1 from 0 to t').

momentum representation and sought the probability amplitudes $a(\vec{p}, t)$ and $b(\vec{p}', t)$ for the atom to be in states a and b with center-of-mass momenta $\vec{p}$ and $\vec{p}'$, respectively. A knowledge of these quantities enabled us to calculate the desired quantity [see Eq. (I34)]

$$\rho_{ab}(\vec{R}, t, \vec{v}_0) = (2\pi/\hbar)^{-3} \int d^3p \int d^3p' \exp(i\hbar^{-1}\{(\vec{p}-\vec{p}')\cdot\vec{R} - [p^2-(p')^2]t/2m\}) e^{-i\omega t} a(\vec{p}, t) b(\vec{p}', t)^* , \quad (3)$$

where ω is the frequency difference of the laser levels a and b, and m is the mass of the active atom. Perturbation theory was used to determine $a(\vec{p}, t)$ and $b(\vec{p}', t)$, and we found it was convenient to introduce diagrams to represent each term in the perturbation solution. For example, the only diagram leading to a contribution for $\rho_{ab}(\vec{R}, t', \vec{v}_0)$ to first order in the laser field is shown in Fig. 3. The upper line indicates the a-state amplitude and the lower one indicates what finally yields the b-state amplitude at time t'. In the Feynman sense, the field is assumed to act instantaneously at t_1 and it is necessary to carry out an integration over all possible t_1 from 0 to t'.

At any time $\hat{t}$ between 0 and t', the product $a(\vec{p}, \hat{t})\, b(\vec{p}', \hat{t})^*$ is well defined. It is clear from Fig. 3 that an interpretation of the diagram leads one to conclude that only $\rho_{aa}(\vec{R}, \hat{t}, \vec{v}_0)$ is nonzero for times between 0 and t_1, while only $\rho_{ab}(\vec{R}, \hat{t}, \vec{v}_0)$ is nonzero for times between t_1 and t'. Thus, the diagrammatic solution allows an interpretation in which, for a given diagram, only one density matrix element is nonzero at any time. This feature will be critical in formulating the PCM.

B. Effects of Classical Collision

Let us temporarily put aside our quantum-mechanical calculation and consider the effect of a classically described collision on the active atom. The active atom will also be thought of as classical in that one may assign to it a given position and velocity. Hence, its density matrix is specified by $\rho(\vec{R}, t, \vec{v}, \vec{v}_0)$, where $\vec{R}$ and $\vec{v}$ are the active atom's position and velocity, respectively, at time t.

Although not conventionally done, it is also possible to solve the classical problem using a diagrammatic approach. With this approach, it is relatively easy to determine how classical collisions affect the various density matrix elements. In time regions where a diagonal density matrix element is nonzero (in the diagrammatic sense), the only effect of a collision is to alter the velocity associated with that density matrix element (collisions are assumed to be adiabatic in the sense that they may not result in population transfer between levels a and b). On the other hand, in regions where an off-diagonal density matrix element is nonzero, collisions may alter the velocity associated with this element *and* produce a phase shift. If the collision parameters are represented symbolically by Θ, then the change in ρ_{ab} produced by a collision is given by

$$\rho_{ab}(\vec{R}, t, \vec{v}, \vec{v}_0) \Rightarrow \rho_{ab}(\vec{R}, t, \vec{v}'(\Theta, \vec{v}), \vec{v}_0)\, e^{i\chi(\Theta, \vec{v})} , \quad (4)$$

where $\chi = \chi_a - \chi_b$ is the relative collision-induced phase shift. (That is, the collision produces phase shifts χ_a and χ_b, respectively, in the a and b atomic state probability amplitudes.)

The changes in velocity which result from collisions may modify the Doppler effect if the additional Doppler phase shift resulting from the collision reaches a value of unity in the lifetime τ of the atom, i.e., if

$$|\vec{k}\cdot\Delta\vec{v}|\,\tau \gtrsim 1 , \quad \Delta\vec{v} = \vec{v}' - \vec{v} . \quad (5)$$

Collisions that produce a $\Delta\vec{v}$ satisfying Eq. (5) and, in addition, do not add any phase-shift factors to the specific density matrix element under consideration are termed *Doppler-modifying collisions* (DMC). On the other hand, collisions which do give rise to an additional phase-shift factor in the density matrix element under consideration but which produce negligible velocity changes, $|\vec{k}\cdot\Delta\vec{v}|\,\tau \ll 1$, will be termed *phase-shifting collisions*. If

a collision gives rise to an additional phase factor in the density matrix element under consideration and produces a $\Delta\vec{v}$ satisfying Eq. (5), it will be called a DMC-*phase collision*.

With this terminology and our diagrammatic interpretation of the problem, one may conclude that only DMC can affect the diagonal density matrix elements, while both phase-shifting and DMC-phase collisions may affect the off-diagonal matrix elements. The change in velocity and phase shift of DMC-phase collisions are correlated, both being functions of the active-atom velocity $\vec{v}$ and the collision parameters Θ. In general, the larger the phase shift, the larger the change in velocity.

One should note that the above "classical" description of a collision need not be purely classical in nature. That is, the diagrammatic approach allows us to view each collision as affecting only one density matrix element, and, as such, one could take different collision interactions depending on which density matrix element (ρ_{aa}, ρ_{bb}, ρ_{ab}, or ρ_{ba}) is nonzero (in the diagrammatic sense) at the time of the collision. (In a purely classical theory the interaction would be state independent.) Thus, the diagrammatic approach allows one to broaden his view of "classical" collisions by permitting state-dependent collision interactions. This "generalized classical picture" would be adequate for our purposes if it contained a prescription for determining the change in velocity associated with DMC-phase collisions which act on off-diagonal density matrix elements. We shall see that the quantum-mechanical result of QMI will provide the necessary prescription for treating such collisions.

C. Review of Quantum-Mechanical One-Collision Result

In QMI we found that the quantum-mechanical results could be interpreted in terms of DMC and phase-shifting collisions, but in a manner different from the classical one discussed above. Equation (I68) for the collisional factor shows its separation into DMC and phase-shifting collision terms, and for convenience it is rewritten here as (this was for a-state scattering only)

$$\text{collision factor} = \tfrac{1}{2}\overline{\Gamma}^{a}_{\mathrm{QM}}(v_0)\,(t'-t_1) + t_1\{\Gamma^{a}_{\mathrm{DM}}(v_0) - \int' d\Omega_v W_a(\vec{v}\,|\,\vec{v}_0) \times \exp[i\vec{k}\cdot(\vec{v}-\vec{v}_0)(t'-t_1)]\}, \quad (6)$$

where the prime restricts the angular integration to the DMC region. The complex quantity $\overline{\Gamma}^{a}_{\mathrm{QM}}(v_0)$ is a phase-shifting line-shape parameter and $\Gamma^{a}_{\mathrm{DM}}(v_0)$ and $W_a(\vec{v}\,|\,\vec{v}_0)$ are DMC line-shape parameters to be discussed later. The key point is that the DMC are associated with the time interval t_1, while the phase-shifting collisions are associated with the time interval $t'-t_1$. Reference to Fig. 3 will indicate that the intervals t_1 and $t'-t_1$ correspond to regions where only $\rho_{aa}\neq 0$ and only $\rho_{ab}\neq 0$, respectively. Hence, our quantum-mechanical result can be interpreted to imply that only DMC may occur if diagonal density matrix elements are nonzero and only phase-shifting collisions may occur if off-diagonal density matrix elements are nonzero. Recall that only one density matrix element may be nonzero in any time region according to our Feynman-type analysis, so that DMC and phase-shifting collisions must occur at mutually exclusive times.[7] The above interpretation in terms of separately occurring DMC and phase collisions is actually part of the PCM.

One may seek a comparison between the classical and quantum-mechanical descriptions of a collision. To make this comparison, one must first achieve some common ground. Hence, let us imagine that either the quantum-mechanical or classical center-of-mass calculation has been performed using the diagrammatic technique. In that case, each time interval between field interactions can be associated with a single density matrix element. If a diagonal density matrix element is nonzero, then only DMC are allowed in either the classical or quantum-mechanical models. On the other hand, if an off-diagonal element is nonzero, only phase-shifting collisions (recall that a phase-shifting collision implies no velocity change) are allowed in the quantum-mechanical model, while phase-shifting *and* DMC-phase collisions are allowed in the classical case. We see that a simple method for correcting the classical model is to permit only phase-shifting classical collisions in time regions where off-diagonal density matrix elements are nonzero (i.e., DMC-phase collisions are forbidden). With that restriction the generalized classical and quantum-mechanical collision models become equivalent in principle. Again we must stress that the diagrammatic interpretation is of the essence here. If one has a very strong collision at a time when an off-diagonal density matrix element is nonzero (in the diagrammatic sense), the collision cannot alter the velocity associated with this element.[7]

[The reason that DMC-phase collisions do not occur in the quantum-mechanical calculation is related to the fact that, after a collision, $\rho_{ab}(\vec{R}, t, \vec{v})$ is composed of the spatial overlap of a plane wave and spherically scattered wave. Such an overlap leads to a rapidly varying phase factor in all but the forward direction. Thus, $\rho_{ab}(\vec{R}, t, \vec{v})$ is effectively zero in all but the forward direction and this corresponds to phase shifting rather than DMC-phase collisions. If there were collision interaction in both radiative levels, DMC-phase collisions would appear in the quantum-mechanical calculations, but without classical analogs (see Appendix A).]

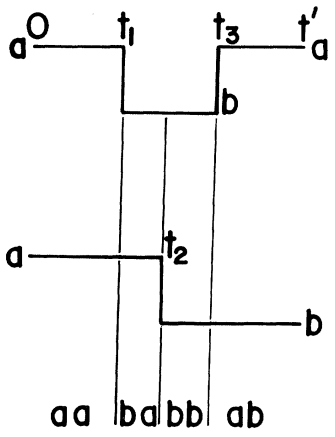

FIG. 4. Diagram leading to a contribution for $\rho_{ab}(\vec{R}, t', \vec{v}_0)$ to third order in the laser field. In the Feynman sense, the field is assumed to interact with the laser atom at times t_1, t_2, and t_3. The diagram is drawn with the convention that $t_3 > t_2 > t_1$, so that one may interpret it to imply that the only nonzero density matrix element which may exist in a given time interval is the one corresponding to the label for the interval. That is, only $\rho_{aa} \neq 0$ for time between 0 and t_1, only $\rho_{ba} \neq 0$ for times between t_1 and t_2, etc.

D. Pseudoclassical Collision Model (PCM)

We are now ready to combine Secs. III A–III C and establish rules for using the PCM.

(a) First, one must obtain the appropriate time-ordered diagrams for the sought after quantity [in laser theory this quantity is $\rho_{ab}(\vec{R}, t, \vec{v}_0)$] to the desired order in the external field with the neglect of all collisions. An example of such a diagram representing one contribution to $\rho_{ab}(\vec{R}, t', \vec{v}_0)$ in third-order laser theory is shown in Fig. 4, where $t_3 > t_2 > t_1$.

(b) Next, the diagrams must be partitioned into time regions representing the intervals between field interactions. This is also done in Fig. 4, where the various time intervals are labeled by the nonzero density matrix element associated with that interval. For the diagram shown, the time regions and their labels are $t_1 : aa$; $t_2 - t_1 : ba$; $t_3 - t_2 : bb$; and $t' - t_3 : ab$.

(c) Now one may put classical collisions into the no-collision result by allowing for the appropriate types of collisions in the various regions. For example, in regions *aa* and *bb*, classical DMC are permissible (lest one forget, the DMC will be allowed in region *aa* or *bb*, but not both, since we are dealing with collision interaction in one state only). On the other hand, in regions *ba* and *ab*, only classical phase-shifting collisions may occur. It should be remembered that these classical phase-shifting collisions do *not* change the velocity associated with the off-diagonal density matrix element, as discussed in Sec. III C. To obtain the final result with collisions, an average over all possible collision histories is performed.

(d) The final result will then depend on classical line-shape parameters. If desired, the corresponding quantum-mechanical line-shape parameters may be substituted for the classical ones, based on the results of QMI.

Thus, we see that by a slight revision of our classical concept of a collision (allowing *only* phase-shifting collisions to affect off-diagonal density matrix elements), used in conjunction with the diagrammatic interpretation of the time evolution of the density matrix for the system, the PCM for incorporating collisions into "no-collision" results has been developed. The revision was necessitated by our knowledge of the quantum-mechanical collision calculation and hence the term "pseudoclassical" is applied to the entire model. The reader may wish to return to the above outline of the PCM when reading Secs. IV–VI.

It is of interest to point out that the quantum-mechanical calculation has led to simplified results. That is, in previous classical theories,[8] it was the DMC-phase collisions that proved to be most difficult to handle. The net result of the quantum-mechanical calculation is that such collisions need not be considered when there is scattering interaction in one state only.

IV. LASER SOLUTION WITH NO COLLISIONS TO THIRD ORDER IN FIELD

The laser is assumed to be operating in a single mode of the Fabry-Perot cavity with the field given by

$$\vec{E}(t) = \hat{n} E(t) \cos[\Omega t + \varphi(t)] \sin(\vec{k} \cdot \vec{R}) , \tag{7}$$

where $k = \Omega/c$, $\hat{k}$ is the direction of the laser axis, and $\hat{n}$ is a direction perpendicular to $\hat{k}$. The net result of Sec. III of QMI was to obtain the self-consistent amplitude and phase equations for the slowly varying amplitude $E(t)$ and phase $\varphi(t)$ of the laser field. These equations, written in a slightly different form from that in (I19) and (I21), are

$$\dot{E} + \Omega/2Q E = -(\Omega \wp \lambda_a / \epsilon_0) \operatorname{Im} \mathcal{A}(t) , \tag{8}$$

$$\dot{\varphi} E = -(\Omega \wp \lambda_a / \epsilon_0) \operatorname{Re} \mathcal{A}(t) , \tag{9}$$

where the driving functions $\mathcal{A}(t)$ and $\mathcal{A}(t, \vec{v}_0)$ are given by

$$\mathcal{A}(t) = \int d^3 v_0 \, W_a(\vec{v}_0) \mathcal{A}(t, \vec{v}_0) , \tag{10}$$

$$\mathcal{A}(t, \vec{v}_0) = \int_0^\infty dt' \, A(t', \vec{v}_0) , \tag{11}$$

λ_a is the average rate for excitation of laser atoms to state a, $\wp$ is the matrix element for the $\hat{n}$ compo-

nent of the electric dipole moment of the atom between states a and b, Q is the quality factor of the cavity, and $W_a(\vec{v}_0)$ is the velocity distribution for initially excited laser atoms.[9] Both the amplitude and phase equations will be specified when one finds $A(t', \vec{v}_0)$, the polarization function of an atom excited with velocity $\vec{v}_0$.

To obtain $A(t', \vec{v}_0)$, one solves Eqs. (I30) for the probability amplitudes in momentum space using an iterative approach and then uses Eqs. (3), (1), and (2) to get $A(t', \vec{v}_0)$. Since we are not interested in collisions at present, all collision terms should be omitted in Eqs. (I30). To first order in the laser field, the contribution to $A(t', \vec{v}_0)$ can be represented by the one diagram shown in Fig. 3. We have already calculated this term in QMI [where it was designated $A^{00;01}(t', \vec{v}_0)$] and, transcribing the result from Eq. (I42), we have

$$A_{NC}^{(1)}(t', \vec{v}_0) = -\tfrac{1}{4} i \wp E(t') \hbar^{-1} V^{-1} \int_0^{t'} dt_1 \times \exp[(-\gamma_{ab} - i\Delta\omega + i\vec{k}\cdot\vec{v}_0)(t' - t_1) - \gamma_a t_1] + [(\text{term})\vec{k} \to -\vec{k}] , \quad (12)$$

where

$$\gamma_{ab} = \tfrac{1}{2}(\gamma_a + \gamma_b) , \quad (13a)$$

$$\Delta\omega = \omega - \Omega - \dot{\varphi}(t') , \quad (13b)$$

NC stands for "no collision," and the superscript refers to the order of the laser field. The driving function $\mathcal{A}(t, \vec{v}_0)$ is given by Eq. (11) as

$$\mathcal{A}_{NC}^{(1)}(t, \vec{v}_0) = \int_0^\infty dt' \, A_{NC}^{(1)}(t', \vec{v}_0) ,$$

and, making the obvious change of variable $\tau' = t' - t_1$, and using Eq. (12), one finds

$$\mathcal{A}_{NC}^{(1)}(t, \vec{v}_0) = -\tfrac{1}{4} i \wp E(t) \hbar^{-1} V^{-1} \times \int_0^\infty d\tau' \int_0^\infty dt_1 \mathcal{K}(t_1, \tau') \, e^{i\vec{k}\cdot\vec{v}_0\tau'} + [(\text{term})\vec{k} \to -\vec{k}] , \quad (14)$$

where

$$\mathcal{K}(t_1, \tau') = e^{-\gamma_a t_1} e^{(-\gamma_{ab} - i\Delta\omega)\tau'} . \quad (15)$$

One can continue the iterative solution of Eqs. (I30) to obtain all terms to third order in the laser field. The third-order contributions can be represented by the four diagrams shown in Fig. 5 and labeled (A)–(D). In all cases $t_3 > t_2 > t_1$, so that diagrams (B)–(D) differ only in their time ordering. It is a straightforward calculation, similar to that for $\mathcal{A}_{NC}^{(1)}(t, \vec{v}_0)$, to find the contribution of each of these terms to $\mathcal{A}_{NC}^{(3)}(t, \vec{v}_0)$, and we shall merely state the results. In the Doppler limit (Doppler width $\gg$ natural width and cavity detuning[10]) we find

$$\mathcal{A}_{NC}^{(3A)}(t, \vec{v}_0) = C[E(t)]^3 \int_0^\infty d\tau' \int_0^\infty d\tau'' \int_0^\infty d\tau''' \int_0^\infty dt_1 \, F_a(t_1, \tau''', \tau'', \tau') \, e^{i\vec{k}\cdot\vec{v}_0\tau'} e^{-i\vec{k}\cdot\vec{v}_0\tau'''} + [(\text{term})\vec{k} \to -\vec{k}] , \quad (16a)$$

$$\mathcal{A}_{NC}^{(3B)}(t, \vec{v}_0) = C[E(t)]^3 \int_0^\infty d\tau' \int_0^\infty d\tau'' \int_0^\infty d\tau''' \int_0^\infty dt_1 \, M_a(t_1, \tau''', \tau'', \tau') \, e^{i\vec{k}\cdot\vec{v}_0\tau'} e^{-i\vec{k}\cdot\vec{v}_0\tau'''} + [(\text{term})\vec{k} \to -\vec{k}] , \quad (16b)$$

$$\mathcal{A}_{NC}^{(3C)}(t, \vec{v}_0) = C[E(t)]^3 \int_0^\infty d\tau' \int_0^\infty d\tau'' \int_0^\infty d\tau''' \int_0^\infty dt_1 \, M_b(t_1, \tau''', \tau'', \tau') \, e^{i\vec{k}\cdot\vec{v}_0\tau'} e^{-i\vec{k}\cdot\vec{v}_0\tau'''} + [(\text{term})\vec{k} \to -\vec{k}] , \quad (16c)$$

$$\mathcal{A}_{NC}^{(3D)}(t, \vec{v}_0) = C[E(t)]^3 \int_0^\infty d\tau' \int_0^\infty d\tau'' \int_0^\infty d\tau''' \int_0^\infty dt_1 \, F_b(t_1, \tau''', \tau', \tau') \, e^{i\vec{k}\cdot\vec{v}_0\tau'} e^{-i\vec{k}\cdot\vec{v}_0\tau'''} + [(\text{term})\vec{k} \to -\vec{k}] , \quad (16d)$$

where

$$C = \tfrac{1}{64} i \wp^3 \hbar^{-3} V^{-1} , \quad (17)$$

$$F_\alpha(t_1, \tau''', \tau'', \tau') = e^{-\gamma_a t_1} e^{(-\gamma_{ab} - i\Delta\omega)\tau'''} \times e^{-\gamma_\alpha \tau''} e^{(-\gamma_{ab} - i\Delta\omega)\tau'} , \quad (18a)$$

$$M_\alpha(t_1, \tau''', \tau'', \tau') = e^{-\gamma_a t_1} e^{(-\gamma_{ab} + i\Delta\omega)\tau'''} \times e^{-\gamma_\alpha \tau''} e^{(-\gamma_{ab} - i\Delta\omega)\tau'} , \quad \alpha = a \text{ or } b \quad (18b)$$

$$\tau''' = t_2 - t_1 , \quad (19a)$$

$$\tau'' = t_3 - t_2 , \quad (19b)$$

$$\tau' = t' - t_3 . \quad (19c)$$

Note that the integrands may be factored into terms depending only on the time intervals t_1, τ''', τ'', and τ' between field interactions. This feature will prove helpful in Sec. V.

Thus, to third order in the field,

$$\mathcal{A}_{NC}(t, \vec{v}_0) = \mathcal{A}_{NC}^{(1)}(t, \vec{v}_0) + \mathcal{A}_{NC}^{(3A)}(t, \vec{v}_0) + \mathcal{A}_{NC}^{(3B)}(t, \vec{v}_0) + \mathcal{A}_{NC}^{(3C)}(t, \vec{v}_0) + \mathcal{A}_{NC}^{(3D)}(t, \vec{v}_0) . \quad (20)$$

It remains to put collisions into this equation by use of the PCM. The result with collisions will take the form of the no-collision result propagated through the various time regions t_1, τ''', τ'', and τ' with the appropriate types of collisions introduced in each region.

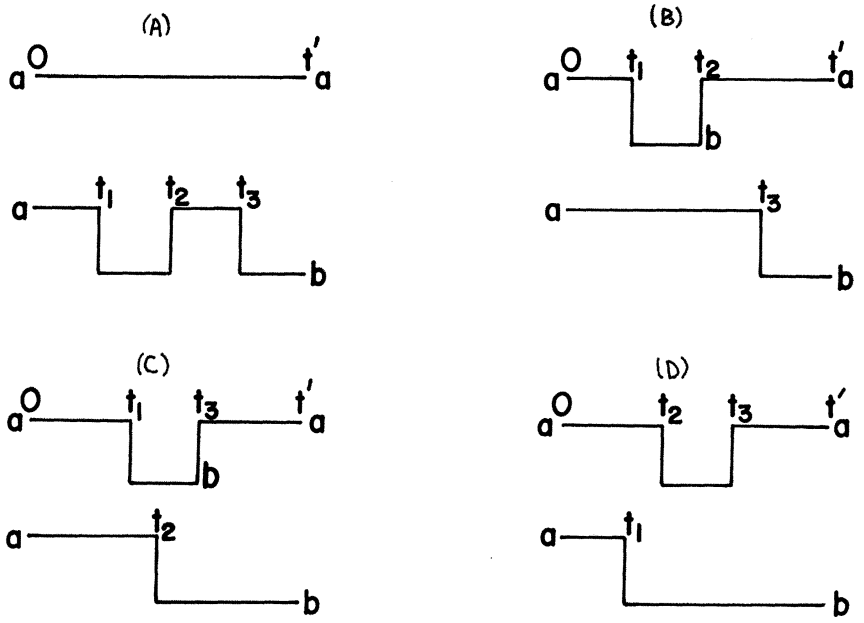

FIG. 5. The four diagrams leading to contributions for $\rho_{ab}(\vec{R}, t', \vec{v}_0)$ [and consequently $\mathcal{A}(t', \vec{v}_0)$] to third order in the laser field. With the adopted convention $t_3 > t_2 > t_1$, one can see that diagrams (B)–(D) differ only in their time ordering.

V. INCORPORATION OF COLLISIONS INTO NO-COLLISION RESULTS

Following the procedure outlined in Sec. III D, we shall use the PCM to incorporate collisions into the no-collision result. There are only four distinct types of time regions which need be considered, and these are shown in Fig. 6. All contributions to $\mathcal{A}(t, \vec{v}_0)$ can be constructed from a chain of such regions. For example, $\mathcal{A}^{(1)}(t, \vec{v}_0)$ (see Fig. 3) is composed of segments $aa(t_i = 0,\ t_{i+1} = t_1)$ and $ab(t_i = t_1,\ t_{i+1} = t')$, while $\mathcal{A}^{(3C)}(t, \vec{v}_0)$ (see Fig. 5) is composed of the four segments $aa(t_i = 0,\ t_{i+1} = t_1)$, $ba(t_i = t_1, t_{i+1} = t_2)$, $bb(t_i = t_2, t_{i+1} = t_3)$, and $ab(t_i = t_3, t_{i+1} = t')$. As a specific example, we shall compute the effects of collisions on the $\mathcal{A}^{(3C)}(t, \vec{v}_0)$ term. This term is is quite general, since it contains all types of time regions. After having calculated $\mathcal{A}^{(3C)}(t, \vec{v}_0)$, it will be an easy matter to write expressions for all the other terms.

For convenience, we rewrite the integrand of the expression for $\mathcal{A}_{NC}^{(3C)}(t, \vec{v}_0)$ [see Eqs. (16c) and (18b)], factoring it into functions of each time region, as

$$\mathcal{A}_{NC}^{(3C)}(t_1, \tau''', \tau'', \tau'; t, \vec{v}, \vec{v}_0) = C[E(t)]^3 \mathcal{A}_{aa}^{(3C)}(t_1, \vec{v})$$

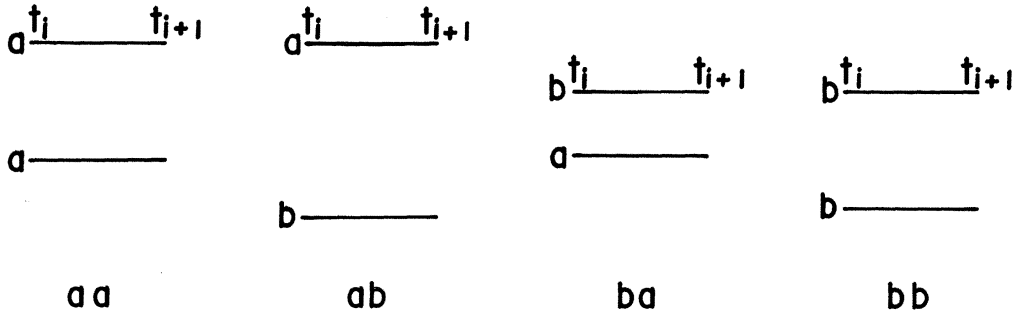

FIG. 6. The four types of "time regions" that form the substructure of the perturbation diagrams presented in this work. The times t_i and t_{i+1} represent the times at the beginning and end of the intervals, respectively, and the labels represent the nonzero density matrix elements associated with that region. By choosing appropriate values for t_i and t_{i+1}, one can construct any of the perturbation diagrams by using a chain of these time regions. For example, the diagram of Fig. 3 can be constructed from $aa(t_i = 0,\ t_{i+1} = t_1)$ and $ab(t_i = t_1,\ t_{i+1} = t')$.

$$\times \mathcal{A}_{ba}^{(3C)}(\tau''', \vec{v})\, \mathcal{A}_{bb}^{(3C)}(\tau'', \vec{v}) \mathcal{A}_{ab}^{(3C)}(\tau', \vec{v}) , \quad (21)$$

where

$$\mathcal{A}_{aa}^{(3C)}(t_1, \vec{v}) = e^{-\gamma_a t_1} , \quad (22a)$$

$$\mathcal{A}_{ba}^{(3C)}(\tau''', \vec{v}) = e^{(-\gamma_{ab} + i\Delta\omega)\tau'''} e^{-i\vec{k}\cdot\vec{v}\tau'''} , \quad (22b)$$

$$\mathcal{A}_{bb}^{(3C)}(\tau'', \vec{v}) = e^{-\gamma_b \tau''} , \quad (22c)$$

$$\mathcal{A}_{ab}^{(3C)}(\tau', \vec{v}) = e^{(-\gamma_{ab} - i\Delta\omega)\tau'} e^{i\vec{k}\cdot\vec{v}\tau'} , \quad (22d)$$

and $\vec{v}$ is the velocity associated with the given time region (for no collisions $\vec{v} = \vec{v}_0$ in all regions). To include collisions in this result, we must sum over all possible number of appropriate (i.e., either phase-shifting or DMC) collisions in each region and average over all collision parameters (i.e., impact parameter, perturber velocity, etc.).

The quantity $\mathcal{A}^{(3C)}(t_1, \tau''', \tau'', \tau'; t, \vec{v}_0)$ will denote the collision-modified value of $\mathcal{A}_{NC}^{(3C)}(t_1, \tau''', \tau'', \tau'; t, \vec{v}, \vec{v}_0)$ averaged over all possible collision histories. We may obtain a useful expression for $\mathcal{A}^{(3C)}(t_1, \tau''', \tau'', \tau'; t, \vec{v}_0)$ if we explicitly indicate the integration over a few of the collision variables. Hence, we write the collision-modified and -averaged value of Eq. (21) as

$$\mathcal{A}^{(3C)}(t_1, \tau''', \tau'', \tau'; t, \vec{v}_0) = C[E(t)]^3 \int d^3v \int d^3v_3 \int d^3v_2 \int d^3v_1 \mathcal{A}_{aa}^{(3C)}(t_1; \vec{v}_0, \vec{v}_1)$$
$$\times \mathcal{A}_{ba}^{(3C)}(\tau'''; \vec{v}_1, \vec{v}_2) \mathcal{A}_{bb}^{(3C)}(\tau''; \vec{v}_2, \vec{v}_3) \mathcal{A}_{ab}^{(3C)}(\tau'; \vec{v}_3, \vec{v}) , \quad (23)$$

where the density (in velocity space) $\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau; \vec{v}_i, \vec{v}_{i+1})$ is the collision-modified value of $\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau, \vec{v})$ averaged over all possible collision histories in the time region associated with the $\alpha\alpha'$-state configuration ($\alpha, \alpha' = a$ or b) which begin with the active atom having velocity $\vec{v}_i$ and end with it having velocity $\vec{v}_{i+1}$. One must perform the integrations over the atom's velocity at the end of each time region to allow for all possible atomic velocities consistent with a velocity $\vec{v}_0$ at the time of excitation.

Thus, Eq. (23) gives a collision-averaged contribution to the driving function from active atoms which had velocity $\vec{v}_0$ at their time of excitation. In Eq. (23) we have used the fact that collisions in different time regions are uncorrelated to write the contribution $\mathcal{A}^{(3C)}(t, \vec{v}_0)$ as a product of averages.

It remains to calculate the $\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau; \vec{v}_i, \vec{v}_{i+1})$. Since we are dealing with classical collisions, it is a simple matter to average over all possible collision histories in the $\alpha\alpha'$ region and obtain[11]

$$\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau; \vec{v}_i, \vec{v}_{i+1}) = P_{\alpha\alpha'}^0(\tau, \vec{v}_i) \mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau, \vec{v}_i) \delta(\vec{v}_{i+1} - \vec{v}_i)$$
$$+ \int d\Theta \int_0^\tau d\tau_1 P_{\alpha\alpha'}^0(\tau_1, \vec{v}_i) P_{\alpha\alpha'}(\Theta(\vec{v}_i \to \vec{v}_{i+1}, \tau_1)) P_{\alpha\alpha'}^0(\tau - \tau_1, \vec{v}_{i+1}) \mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau; \Theta(\vec{v}_i \to \vec{v}_{i+1}, \tau_1))$$
$$+ \int d^3v' \int d\Theta_2 \int d\Theta_1 \int_0^\tau d\tau_2 \int_0^{\tau_2} d\tau_1 P_{\alpha\alpha'}^0(\tau_1, \vec{v}_i) P_{\alpha\alpha'}(\Theta_1(\vec{v}_i \to \vec{v}', \tau_1)) P_{\alpha\alpha'}^0(\tau_2 - \tau_1, \vec{v}')$$
$$\times P_{\alpha\alpha'}(\Theta_2(\vec{v}' \to \vec{v}_{i+1}, \tau_2)) P_{\alpha\alpha'}^0(\tau - \tau_2, \vec{v}_{i+1}) A_{\alpha\alpha'}^{(3C)}(\tau; \Theta_1(\vec{v}_i \to \vec{v}', \tau_1); \Theta_2(\vec{v}' \to \vec{v}_{i+1}, \tau_2)) + \cdots , \quad (24)$$

where $P_{\alpha\alpha'}^0(\tau, \vec{v})$ is the probability that an atom moving with velocity $\vec{v}$ in a region $\alpha\alpha'$ undergoes no collisions in a time τ; $P_{\alpha\alpha'}(\Theta_1(\vec{v} \to \vec{v}', \tau_1))$ is the probability density per unit time that an active atom moving with velocity $\vec{v}$ undergoes a collision at time τ_1 in the region $\alpha\alpha'$ specified by collision parameters Θ_1 that changes its velocity to $\vec{v}'$ ($P_{\alpha\alpha'}$ is actually independent of τ_1); and

$$\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau; \Theta_1(\vec{v} \to \vec{v}', \tau_1); \Theta_2(\vec{v}' \to \vec{v}'', \tau_2); \text{etc.})$$

is the collision-modified value of $\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau, \vec{v})$ if a collision specified by Θ_1 occurs at time τ_1, a collision specified by Θ_2 occurs at time τ_2, etc. In all cases, "collision" means the type of collision appropriate to the given region $\alpha\alpha'$.

Inspection of Eqs. (22) will show that $\mathcal{A}_{\alpha\alpha'}^{(3C)}(\tau, \vec{v})$ has essentially two forms depending on whether $\alpha = \alpha'$ or $\alpha \neq \alpha'$.

$\alpha = \alpha'$. If $\alpha = \alpha'$, we find [see Eqs. (22)] that

$$\mathcal{A}_{\alpha\alpha}^{(3C)}(\tau, \vec{v}) = \mathcal{A}_{\alpha\alpha}(\tau) \quad (25)$$

with no velocity dependence.[12] Only DMC are allowed in these "diagonal" regions and these do not affect $\mathcal{A}_{\alpha\alpha}$, i.e.,

$$\mathcal{A}_{\alpha\alpha}^{(3C)}(\tau; \Theta(\vec{v}_i \to \vec{v}_{i+1}, \tau_1)) = \mathcal{A}_{\alpha\alpha}(\tau) . \quad (26)$$

Using this result, we may write Eq. (24) as

$$\mathcal{A}_{\alpha\alpha}^{(3C)}(\tau; \vec{v}_i, \vec{v}_{i+1}) = G_{\alpha\alpha}(\tau; \vec{v}_i, \vec{v}_{i+1}) \mathcal{A}_{\alpha\alpha}^{(3C)}(\tau) , \quad (27)$$

where the propagator

$$G_{\alpha\alpha}(\tau; \vec{v}_i, \vec{v}_{i+1}) = P_{\alpha\alpha}^0(\tau, \vec{v}_i, \text{DM}) \delta(\vec{v}_{i+1} - \vec{v}_i)$$

$$+\int d\Theta\int_0^\tau d\tau_1 P^0_{\alpha\alpha}(\tau_1, \vec{v}_i, \mathrm{DM}) \times P_{\alpha\alpha}(\Theta(\vec{v}_i \to \vec{v}_{i+1}, \tau_1, \mathrm{DM})) \times P^0_{\alpha\alpha}(\tau - \tau_1, \vec{v}_{i+1}, \mathrm{DM}) + \cdots \quad (28)$$

and the DM arguments in the P's imply that these probabilities apply to Doppler-modifying collisions only. It is easy to compute[11] the probability for no DMC to occur in a time τ as

$$P^0_{\alpha\alpha}(\tau, \vec{v}, \mathrm{DM}) = \exp[-\Gamma^{\mathrm{DM}}_\alpha(\vec{v})\tau] , \quad (29)$$

where $\Gamma^{\mathrm{DM}}_\alpha(\vec{v})$ is the rate at which DMC occur for active atoms in state α moving with velocity $\vec{v}$. Furthermore, from the definition of $P_{\alpha\alpha}(\Theta(\vec{v} \to \vec{v}', \tau, \mathrm{DM}))$, it follows that the quantity $W_\alpha(\vec{v} \to \vec{v}')$ defined by

$$W_\alpha(\vec{v} \to \vec{v}') = \int d\Theta P_{\alpha\alpha}(\Theta(\vec{v} \to \vec{v}', \tau, \mathrm{DM})) \quad (30a)$$

is just the probability density per unit time that a DMC changes the velocity of an atom in state α from $\vec{v}$ to $\vec{v}'$. Hence it is easy to see that $\Gamma^{\mathrm{DM}}_\alpha(\vec{v})$ and $W_\alpha(\vec{v} \to \vec{v}')$ are related by

$$\Gamma^{\mathrm{DM}}_\alpha(\vec{v}) = \int' d^3v' \, W_\alpha(\vec{v} \to \vec{v}') , \quad (30b)$$

where the prime on the integral indicates that we must restrict the integration to only those values of $\vec{v}'$ which represent DMC, i.e., such that $|\vec{k} \cdot (\vec{v}' - \vec{v})| \gamma_{ab}^{-1} \gtrsim 1$ [see Eq. (5)]. Using Eqs. (29) and (30), we may rewrite Eq. (28) as

$$G_{\alpha\alpha}(\tau; \vec{v}_i, \vec{v}_{i+1}) = \exp[-\Gamma^{\mathrm{DM}}_\alpha(\vec{v}_i)\tau]\,\delta(\vec{v}_{i+1} - \vec{v}_i) + \int_0^\tau d\tau_1 \exp[-\Gamma^{\mathrm{DM}}_\alpha(\vec{v}_i)\tau_1] W_\alpha(\vec{v}_i \to \vec{v}_{i+1}) \times \exp[-\Gamma^{\mathrm{DM}}_\alpha(\vec{v}_{i+1})(\tau - \tau_1)] + \cdots \quad (31)$$

$\alpha \neq \alpha'$. On the other hand, when $\alpha \neq \alpha'$, the "nondiagonal" terms have the form [see Eqs. (22)]

$$\mathcal{A}^{(3C)\pm}_{\alpha\alpha'}(\tau, \vec{v}) = \mathcal{A}^{(3C)}_{\alpha\alpha'}(\tau)\, e^{\pm i\vec{k}\cdot\vec{v}\tau} \quad (\alpha \neq \alpha') , \quad (32)$$

and only phase-shifting collisions (which imply no velocity changes) are allowed in these regions. These phase-shifting collisions merely alter the $\mathcal{A}_{\alpha\alpha'}(\tau)$ by introducing the appropriate phase factor for the collisions. For example, if two collisions occur in time τ, we find that

$$\mathcal{A}^{(3C)}_{\alpha\alpha'}(\tau; \Theta_1(\vec{v}_i \to \vec{v}_i, \tau_1); \Theta_2(\vec{v}_i \to \vec{v}_i, \tau_2)) = \mathcal{A}^{(3C)}_{\alpha\alpha'}(\tau) \exp[i\chi_{\alpha\alpha'}(\Theta_1, \vec{v}_i)] \exp[i\chi_{\alpha\alpha'}(\Theta_2, \vec{v}_i)] , \quad (33a)$$

where

$$\chi_{\alpha\alpha'}(\Theta_1, \vec{v}_i) = \chi_\alpha(\Theta_1, \vec{v}_i) - \chi_{\alpha'}(\Theta_1, \vec{v}_i) \quad (33b)$$

is the relative phase shift induced by the "1" collision when the active atom has velocity $\vec{v}_i$. Using the above result in Eq. (24), we obtain

$$\mathcal{A}^{(3C)\pm}_{\alpha\alpha'}(\tau; \vec{v}_i, \vec{v}_{i+1}) = G^\pm_{\alpha\alpha'}(\tau; \vec{v}_i, \vec{v}_{i+1}; \vec{k})\mathcal{A}^{(3C)}_{\alpha\alpha'}(\tau) \quad (\alpha \neq \alpha') , \quad (34)$$

where

$$G^\pm_{\alpha\alpha'}(\tau; \vec{v}_i, \vec{v}_{i+1}; \vec{k}) = e^{\pm i\vec{k}\cdot\vec{v}_i\tau}\,\delta(\vec{v}_{i+1} - \vec{v}_i)\{P^0_{\alpha\alpha'}(\tau, \vec{v}_i, \mathrm{ph}) + \int d\Theta\int_0^\tau d\tau_1 P^0_{\alpha\alpha'}(\tau_1, \vec{v}_i, \mathrm{ph}) P_{\alpha\alpha'}(\Theta(\tau_1, \vec{v}_i, \mathrm{ph})) \times P^0_{\alpha\alpha'}(\tau - \tau_1, \vec{v}_i, \mathrm{ph}) \exp[i\chi_{\alpha\alpha'}(\Theta, \vec{v}_i)] + \cdots\} \quad (\alpha \neq \alpha') , \quad (35)$$

where the ph argument implies that these probabilities apply to phase-shifting collisions only. The δ function assures that the velocity remains unchanged in these phase-shifting collisions as required by the PCM. Equation (35) assumes a simplified form if one recognizes that the term in curly brackets in Eq. (35) may be evaluated by standard "impact theory" pressure broadening techniques[11] to yield $\exp[-\bar{\gamma}_{\alpha\alpha'}(\vec{v}_i)\tau]$, where $\bar{\gamma}_{\alpha\alpha'}(\vec{v}_i)$ is a complex line-shape parameter associated with the $\alpha\alpha'$ time region and will be specified shortly. Thus Eq. (35) becomes

$$G^\pm_{\alpha\alpha'}(\tau; \vec{v}_i, \vec{v}_{i+1}; \vec{k}) = e^{\pm i\vec{k}\cdot\vec{v}_i\tau} e^{-\bar{\gamma}_{\alpha\alpha'}(\vec{v}_i)\tau} \times \delta(\vec{v}_{i+1} - \vec{v}_i) . \quad (36)$$

We now have expressions for all the collision "propagators." Using Fig. 5 or Eq. (23) [and Eqs. (22b) and (22d) to fix the sign in the Doppler phase factor], we may combine the propagators in the correct order to evaluate Eq. (23) as

$$\mathcal{A}^{(3C)}(t_1, \tau''', \tau'', \tau'; t, \vec{v}_0) = C[E(t)]^3 \int d^3v \int d^3v_3 \int d^3v_2 \int d^3v_1 \times G_{aa}(t_1; \vec{v}_0, \vec{v}_1)\, G^-_{ba}(\tau'''; \vec{v}_1, \vec{v}_2; \vec{k})\, G_{bb}(\tau''; \vec{v}_2, \vec{v}_3)\, G^+_{ab}(\tau'; \vec{v}_3, \vec{v}; \vec{k}) \times \mathcal{A}^{(3C)}_{aa}(t_1)\, \mathcal{A}^{(3C)}_{ba}(\tau''')\, \mathcal{A}^{(3C)}_{bb}(\tau'')\, \mathcal{A}^{(3C)}_{ab}(\tau') . \quad (37)$$

Performing the necessary time integrations indicated in Eq. (16c) and making use of Eqs. (25), (32), (22), and (18), we obtain the (3C) contribution to the collision-averaged driving function,

$$\mathcal{A}^{(3C)}(t,\vec{v}_0)=C[E(t)]^3\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\int_0^\infty dt_1\int d^3v\int d^3v_3\int d^3v_2\int d^3v_1$$
$$\times G_{aa}(t_1;\vec{v}_0,\vec{v}_1)\,G^-_{ba}(\tau''';\vec{v}_1,\vec{v}_2;\vec{k})\,G_{bb}(\tau'';\vec{v}_2,\vec{v}_3)\,G^+_{ab}(\tau';\vec{v}_3,\vec{v};\vec{k})\,M_b(t_1,\tau''',\tau'',\tau')+[(\text{term})\vec{k}\to-\vec{k}]\ . \quad (38)$$

One sees that our procedure has separated out all the velocity dependence of the integrand and included it in the propagators.

Proceeding in a similar manner, one can find all the other needed terms. In fact, a quick look at Figs. 5 and 3 and Eqs. (16), (18), and (12) will enable one to write, by inspection, the remaining third-order terms

$$\mathcal{A}^{(3A)}(t,\vec{v}_0)=C[E(t)]^3\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\int_0^\infty dt_1\int d^3v\int d^3v_3\int d^3v_2\int d^3v_1$$
$$\times G_{aa}(t_1;\vec{v}_0,\vec{v}_1)\,G^-_{ab}(\tau''';\vec{v}_1,\vec{v}_2;\vec{k})\,G_{aa}(\tau'';\vec{v}_2,\vec{v}_3)\,G^+_{ab}(\tau';\vec{v}_3,\vec{v};\vec{k})\,F_a(t_1,\tau''',\tau'',\tau')+[(\text{term})\vec{k}\to-\vec{k}]\ , \quad (39a)$$

$$\mathcal{A}^{(3B)}(t,\vec{v}_0)=C[E(t)]^3\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\int_0^\infty dt_1\int d^3v\int d^3v_3\int d^3v_2\int d^3v_1$$
$$\times G_{aa}(t_1;\vec{v}_0,\vec{v}_1)\,G^-_{ba}(\tau''';\vec{v}_1,\vec{v}_2;\vec{k})\,G_{aa}(\tau'';\vec{v}_2,\vec{v}_3)\,G^+_{ab}(\tau';\vec{v}_3,\vec{v};\vec{k})\,M_a(t_1,\tau''',\tau'',\tau')+[(\text{term})\vec{k}\to-\vec{k}]\ , \quad (39b)$$

$$\mathcal{A}^{(3D)}(t,\vec{v}_0)=C[E(t)]^3\int_0^\infty d\tau'\int_0^\infty d\tau''\int_0^\infty d\tau'''\int_0^\infty dt_1\int d^3v\int d^3v_3\int d^3v_2\int d^3v_1$$
$$\times G_{aa}(t_1;\vec{v}_0,\vec{v}_1)\,G^-_{ab}(\tau''';\vec{v}_1,\vec{v}_2;\vec{k})\,G_{bb}(\tau'';\vec{v}_2,\vec{v}_3)\,G^+_{ab}(\tau';\vec{v}_3,\vec{v};\vec{k})\,F_b(t_1,\tau''',\tau'',\tau')+[(\text{term})\vec{k}\to-\vec{k}]\ , \quad (39c)$$

and the first-order term

$$\mathcal{A}^{(1)}(t,\vec{v}_0)=-\tfrac{1}{4}i\wp E(t)\hbar^{-1}V^{-1}\int_0^\infty d\tau'\int_0^\infty dt_1\int d^3v\int d^3v_1\,G_{aa}(t_1;\vec{v}_0,\vec{v}_1)\,G^+_{ab}(\tau';\vec{v}_1,\vec{v};\vec{k})\mathcal{K}(t_1,\tau')+[(\text{term})\vec{k}\to-\vec{k}]\ , \quad (40)$$

where C, M, F, and $\mathcal{K}$ are given by Eqs. (17), (18a), (18b), and (15), respectively.

At this point one is ready to find the driving function $\mathcal{A}(t,\vec{v}_0)$ to third order in the laser field by using Eq. (20), and then substitute it into the amplitude and phase equations. Before this procedure is carried out, we should like to indicate values for the line-shape parameters $\bar{\gamma}_{\alpha\alpha'}$, $W_\alpha(\vec{v}\to\vec{v}')$, and Γ^{DM}_α which appear in Eqs. (31) and (36).

Values of line-shape parameters. The line-shape parameters may be evaluated classically by using the results of standard pressure broadening theory[13] and classical Boltzmann-equation theory. For this case

$$\bar{\gamma}_{ab}(v_i)=\bar{\gamma}_{ba}(v_i)^*$$
$$=\mathfrak{N}\int 2\pi b\,db\int d^3v_p\,|\vec{v}_i-\vec{v}_p|\,W_p(v_p)$$
$$\times\{\exp[i\chi(b,|\vec{v}_i-\vec{v}_p|)]-1\}\ , \quad (41a)$$

$W_\alpha(\vec{v}_i\to\vec{v}_{i+1})$ = Boltzmann collision kernel (defined for DMC only) for atoms in state α, (41b)

and

$$\Gamma^{DM}_\alpha(v_i)=\int' d^3v_{i+1}\,W_\alpha(\vec{v}_i\to\vec{v}_{i+1}) \quad (41c)$$

is the rate at which DMC collisions occur for atoms in state α moving with speed v_i; where $\mathfrak{N}$ is the density of perturbers, b is the impact parameter of the classical collision, χ is the relative collision-induced phase shift discussed in Sec III B (recall that $\chi=\chi_a-\chi_b$ and for single-state scattering either $\chi_a=0$ or $\chi_b=0$), $\vec{v}_p$ is the perturber velocity *before* the collision, $W_p(v_p)$ is the perturber velocity distribution, and the prime on the integral indicates the DMC region only, i.e., $|\vec{k}\cdot(\vec{v}_{i+1}-\vec{v}_i|\gtrsim\gamma_{ab}$.

On the other hand, the line-shape parameters may also be evaluated quantum mechanically if the proper correspondence is made between the classical parameters above and the quantum-mechanical parameters found in QMI [see, for example, Eqs. (I62) and (I66)]. Although the results in QMI were given for stationary perturbers, it

is not difficult to generalize to the case of moving perturbers, and one can obtain the line-shape parameters as

$$\bar{\gamma}_{ab}(v_i) = \bar{\gamma}_{ba}(v_i)^* = \tfrac{1}{2}\int d^3v_p W_p(v_p)\,\bar{\Gamma}_a^{\rm ph}(v_i^r)\,, \qquad a \text{ scattering}$$

$$= \tfrac{1}{2}\int d^3v_p W_p(v_p)[\bar{\Gamma}_b^{\rm ph}(v_i^r)]^*\,, \qquad b \text{ scattering} \tag{42a}$$

with

$$\bar{\Gamma}_\alpha^{\rm ph}(v_i^r) = \mathfrak{N}(4\pi/i\mu) f_\alpha(\vec{\mathrm{v}}_i^r \to \vec{\mathrm{v}}_i^r)\,, \tag{42b}$$

$$W_\alpha(\vec{\mathrm{v}}_i \to \vec{\mathrm{v}}_{i+1}) = \mathfrak{N}(m/\mu)^3 \int d^3v_p W_p(v_p) \int d^3v_{i+1}^r \times \delta(\vec{\mathrm{v}}_{i+1}^r + (m/m_p)\vec{\mathrm{v}}_i + \vec{\mathrm{v}}_p - (m/\mu)\vec{\mathrm{v}}_{i+1}) \times \delta(v_i^r - v_{i+1}^r)(v_i^r)^{-1} |f_\alpha(\vec{\mathrm{v}}_i^r \to \vec{\mathrm{v}}_{i+1}^r)|^2\,, \tag{42c}$$

and

$$\Gamma_\alpha^{\rm DM}(v_i) = \int' d^3v_{i+1} W_\alpha(\vec{\mathrm{v}}_i \to \vec{\mathrm{v}}_{i+1}) = \mathfrak{N} \int' d^3v_p W_p(v_p) v_i^r \sigma_\alpha^{\rm DM}(v_i^r)\,,$$

with

$$\sigma_\alpha^{\rm DM}(v_i^r) = \int' d\Omega_{v_{i+1}^r} |f_\alpha(\vec{\mathrm{v}}_i^r \to \vec{\mathrm{v}}_{i+1}^r)|^2\,, \tag{42d}$$

where $\vec{\mathrm{v}}_i^r = \vec{\mathrm{v}}_i - \vec{\mathrm{v}}_p$ and μ are the active atom-perturber relative velocity (before the collision) and reduced mass, respectively, m and m_p are the active atom and perturber mass, respectively, $f_\alpha(\vec{\mathrm{v}}_i^r \to \vec{\mathrm{v}}_i^r)$ is the forward-scattering amplitude, and $|f_\alpha(\vec{\mathrm{v}}_i^r \to \vec{\mathrm{v}}_{i+1}^r)|^2$ is the differential scattering cross section for atoms in state α. Note that in Eq. (42c), the δ functions restrict the integration over the relative velocity after the collision, $\vec{\mathrm{v}}_{i+1}^r$, to those values consistent with conservation of momentum and energy.

In writing $\bar{\gamma}_{ab}$ and $\Gamma_\alpha^{\rm DM}$ as a function of active-atom speed (and not velocity) in Eqs. (41) and (42) we have implicitly assumed an even perturber velocity distribution. This assumption also enables one to show that $W_\alpha(\vec{\mathrm{v}}_i \to \vec{\mathrm{v}}_{i+1}) = W_\alpha(-\vec{\mathrm{v}}_i \to -\vec{\mathrm{v}}_{i+1})$, which, together with Eq. (31), leads to the relation

$$G_{\alpha\alpha}(\tau; \vec{\mathrm{v}}, \vec{\mathrm{v}}') = G_{\alpha\alpha}(\tau; -\vec{\mathrm{v}}, -\vec{\mathrm{v}}')\,.$$

Equations (42), in some sense, indicate the way in which one may calculate line-shape parameters from first principles. Given the emitter-perturber interaction, one calculates the quantum-mechanical scattering amplitudes which, in turn, determine all the line-shape parameters. Of course, such calculations are very difficult and, in addition, require information about the emitter-perturber interaction which is not readily available. As an alternative to first-principles calculations, one could hope to use data from scattering experiments to obtain the scattering amplitudes; however, present experimental techniques are not capable of measuring differential cross sections when one of the colliding atoms is in a short-lived excited state. Hence, it appears that, at best, one must be content with an empirical determination of the line-shape parameters. Such determinations may be achieved if it is possible to get theoretical expressions for either spectral profiles or laser intensity versus detuning line shapes in a form where they can be easily compared with corresponding experimental results. In general, the theoretical line-shape expressions are too complicated to uniquely determine the parameters. However, as we shall see in Secs. VI and VII, the use of some limiting approximations will enable us to extract $\bar{\gamma}_{ab}$ and $\Gamma_\alpha^{\rm DM}$ from certain laser experiments.

VI. LASER RESULTS WITH COLLISIONS

Equations (10), (20), and (38)–(40), together with Eqs. (31) and (36), determine the driving function $\mathcal{A}(t)$ to third order in the laser field. Using these equations and performing the necessary time integrals, we obtain

$$\mathcal{A}(t) = \mathcal{A}^{(1)}(t) + \mathcal{A}^{(3)}(t)\,, \tag{43}$$

where

$$\mathcal{A}^{(1)}(t) = -\tfrac{1}{4} i \wp \hbar^{-1} V^{-1} E(t) \mathfrak{D}^{(1)}\,, \tag{44}$$

$$\mathcal{A}^{(3)}(t) = \tfrac{1}{64} i \wp^3 \hbar^{-3} V^{-1} [E(t)]^3 \mathfrak{D}^{(3)}\,, \tag{45}$$

and the $\mathfrak{D}$ functions are defined as

$$\mathfrak{D}^{(1)} = \int d^3v_0 \int d^3v\, W_a(\vec{\mathrm{v}}_0) S_a(\vec{\mathrm{v}}_0 \to \vec{\mathrm{v}}) \times [\tilde{\gamma}_{ab}(v) + i(\Delta\tilde{\omega}(v) - \vec{\mathrm{k}} \cdot \vec{\mathrm{v}})]^{-1} + [(\text{term})\,\vec{\mathrm{k}} \to -\vec{\mathrm{k}}] \tag{46}$$

and

$$\mathfrak{D}^{(3)} = \int d^3v_0 \int d^3v_2 \int d^3v\, W_a(\vec{\mathrm{v}}_0) S_a(\vec{\mathrm{v}}_0 \to \vec{\mathrm{v}}_2)[S_a(\vec{\mathrm{v}}_2 \to \vec{\mathrm{v}}) + S_b(\vec{\mathrm{v}}_2 \to \vec{\mathrm{v}})]\{[\tilde{\gamma}_{ab}(v_2) + i(\Delta\tilde{\omega}(v_2) + \vec{\mathrm{k}} \cdot \vec{\mathrm{v}}_2)]^{-1} + [\tilde{\gamma}_{ab}(v_2) - i(\Delta\tilde{\omega}(v_2) - \vec{\mathrm{k}} \cdot \vec{\mathrm{v}}_2)]^{-1}\}[\tilde{\gamma}_{ab}(v) + i(\Delta\tilde{\omega}(v) - \vec{\mathrm{k}} \cdot \vec{\mathrm{v}})]^{-1} + [(\text{term})\,\vec{\mathrm{k}} \to -\vec{\mathrm{k}}]\,, \tag{47}$$

with the line-shape parameters given by

$$S_\alpha(\vec{v}_i \to \vec{v}_j) = S_\alpha(-\vec{v}_i \to -\vec{v}_j) \equiv \int_0^\infty d\tau\, e^{-\gamma_\alpha \tau} G_{\alpha\alpha}(\tau;\, \vec{v}_i, \vec{v}_j) \,, \tag{48}$$

$$\tilde{\gamma}_{ab}(v) = \gamma_{ab} + \mathrm{Re}\bar{\gamma}_{ab}(v) \,, \tag{49a}$$

$$\Delta\tilde{\omega}(v) = \Delta\omega + \mathrm{Im}\bar{\gamma}_{ab}(v) \,, \tag{49b}$$

where $\bar{\gamma}_{ab}(v)$ is given by Eq. (42a), $G_{\alpha\alpha}(\tau;\vec{v}_i,\vec{v}_j)$ is given by Eq. (31), and $\Delta\omega$ is given by Eq. (13b). The driving function $\mathcal{A}(t)$ specified by Eqs. (43)–(49) may now be substituted into the amplitude and phase equations [Eqs. (8) and (9), respectively] to yield

$$\dot{E} = \alpha E - \beta E^3 \,, \tag{50a}$$

$$\dot{\varphi} = \sigma + \rho E^2 \,, \tag{50b}$$

where α, β, σ, and ρ are real and given by

$$\alpha = -\tfrac{1}{2}\Omega/Q + \tfrac{1}{4}(\Omega\wp^2\hbar^{-1}\Lambda_a/\epsilon_0)\,\mathrm{Re}\mathfrak{D}^{(1)} \,, \tag{51}$$

$$\beta = \tfrac{1}{64}(\Omega\wp^4\hbar^{-3}\Lambda_a/\epsilon_0)\,\mathrm{Re}\mathfrak{D}^{(3)} \,, \tag{52}$$

$$\sigma = -\tfrac{1}{4}(\Omega\wp^2\hbar^{-1}\Lambda_a/\epsilon_0)\,\mathrm{Im}\mathfrak{D}^{(1)} \,, \tag{53}$$

$$\rho = \tfrac{1}{64}(\Omega\wp^4\hbar^{-3}\Lambda_a/\epsilon_0)\,\mathrm{Im}\mathfrak{D}^{(3)} \,, \tag{54}$$

and $\Lambda_a = \lambda_a/V$ is the excitation rate per unit volume of atoms to state a. The quantity α is the linear gain, β is the saturation parameter, σ is the linear pulling term, and ρ the nonlinear pushing term. The steady-state amplitude is found to be

$$I \equiv E^2 = \frac{\alpha}{\beta} = \frac{-\tfrac{1}{2}\Omega/Q + \tfrac{1}{4}(\Omega\wp^2\hbar^{-1}\Lambda_a/\epsilon_0)\,\mathrm{Re}\mathfrak{D}^{(1)}}{\tfrac{1}{64}(\Omega\wp^4\hbar^{-3}\Lambda_a/\epsilon_0)\,\mathrm{Re}\mathfrak{D}^{(3)}} \,. \tag{55}$$

The driving terms $\mathfrak{D}^{(1)}$ and $\mathfrak{D}^{(3)}$ [see Eqs. (46) and (47)] implicitly contain the features of the pseudoclassical collision model (PCM). That is, in first-order theory the atom may change its velocity in one time region, while in third-order theory it is allowed to change its velocity in two regions. These changes are represented, respectively, by $S_a(\vec{v}_0 \to \vec{v})$ in the first-order terms and $S_a(\vec{v}_0 \to \vec{v}_2) \times [S_a(\vec{v}_2 \to \vec{v}) + S_b(\vec{v}_2 \to \vec{v})]$ in the third-order terms. From phase-shifting time regions, where there is no change of velocity, one invariably obtains resonance-type denominators with γ_{ab} and $\Delta\omega$ of the no-collision theory replaced by $\tilde{\gamma}_{ab} = \gamma_{ab} + \mathrm{Re}\bar{\gamma}_{ab}$ and $\Delta\tilde{\omega} = \Delta\omega + \mathrm{Im}\bar{\gamma}_{ab}$, respectively.

Before attempting an analysis of Eq. (55), it may prove useful to review the approximations that were used in its derivation. For the present, we shall be concerned with only those approximations pertinent to the laser problem and defer, until Sec. VIII, a discussion of the more general assumptions of our collision model. First, we note that Eqs. (51)–(54) were valid for the case of excitation of laser atoms to state a only. The simple extension of the calculation to also allow for excitation of laser atoms to state b is given in Appendix B. A more serious approximation we have made is to work in the Doppler limit. At negligible foreign gas pressures, use of the Doppler limit implies the neglect of terms on the order of γ_{ab}/ku or $\Delta\omega/ku$, where u is the most probable speed of the active atoms. Since, typically, $\gamma_{ab}/ku \approx 10^{-2}$, the errors are not excessive as long as one does not deal with large detunings $\Delta\omega$. However, with the presence of foreign gas, one must replace γ_{ab} by $\tilde{\gamma}_{ab} = \gamma_{ab} + \mathrm{Re}\bar{\gamma}_{ab}$, and at normal laser operating pressures $\tilde{\gamma}_{ab}$ may be five to ten times as large as γ_{ab}. In particular, at 1.3 Torr, a typical laser had $\gamma_{ab}/ku \approx 0.02$ while $\tilde{\gamma}_{ab}/ku \approx 0.1$, so that in taking the Doppler limit, errors of 10% may be introduced. As such, our Doppler-limit expressions are valid only at pressure where $\tilde{\gamma}_{ab}/ku \ll 1$ and $\Delta\tilde{\omega}/ku \ll 1$. However, it is possible to calculate the third-order polarization without restrictions on the ratios of $\tilde{\gamma}_{ab}$, $\Delta\tilde{\omega}$, and ku. The additional terms which will appear in such a calculation are given in Appendix B. Unfortunately, it is very difficult to evaluate these terms once collisions are present. A third assumption we have made is the neglect of spontaneous transitions between laser levels a and b, and this assumption is based on the premise that the partial decay rate for such transitions is negligible. Finally, we should point out that our theory will not be valid for high-intensity lasers. The problem associated with obtaining high-intensity solutions will be discussed in Sec. VIII.

VII. ANALYSIS OF INTENSITY PROFILE: COMPARISON WITH EXPERIMENT

Unless noted otherwise, our discussion will be restricted to the Doppler limit ($\tilde{\gamma}_{ab}/ku \ll 1$ and $\Delta\tilde{\omega}/ku \ll 1$). In order to analyze Eqs. (46) and (47), we shall return to the diagrams of the perturbation solution (see Figs. 3 and 5). In discussing these diagrams, we shall label the time intervals or "time regions" between field interactions by the time difference associated with the intervals. For example, the time region between $t = 0$ and $t = t_1$ appearing in Figs. 3 and 5 will be called "time region t_1," and the time region between $t = t_2$ and $t = t_3$ appearing in Figs. 5 will be called time region τ'' [recall, from Eq. (19b), that $\tau'' = t_3 - t_2$]. It is clear from Figs. 3 and 5 that DMC occur in the time region t_1 for both first- and third-order contributions and in the time region τ'' for third-order contributions. To understand the effects of the DMC, we shall consider these two time regions separately.

Time region t_1. During the time region t_1, which represents the interval between the active atom's excitation and first interaction with the field, the

only effect of collisions is to alter the velocity distribution of the atoms before they interact with the field. This may be seen formally by rewriting a factor common to all contributions to $\mathcal{A}(t)$ [e.g., see Eqs. (44)–(47)] as

$$\int d^3v_0\, W_a(\vec{v}_0)S_a(\vec{v}_0\to\vec{v}) = \int d^3v_0\, W_a(\vec{v}_0)\int_0^\infty d\tau\, e^{-\gamma_a\tau}G_{aa}(\tau;\vec{v}_0,\vec{v}) \equiv \gamma_a^{-1}W_a'(\vec{v})\,, \tag{56}$$

which is an equation defining $W_a'(\vec{v})$, the new effective velocity distribution for the atoms originally excited to state a. There is one case in which $W_a'(\vec{v}) = W_a(\vec{v})$ [recall that $W_a(\vec{v})$ is the velocity distribution characterizing atoms at their time of excitation to state a], and that is when $W_a(\vec{v})$ is taken to be an equilibrium distribution.[14] The equality of $W_a'(\vec{v})$ and $W_a(\vec{v})$ for this situation follows from the fact that, by definition of an equilibrium distribution, collisions can not alter the velocity *distribution* $W_a(\vec{v})$ even though they may change the velocities of *individual* atoms. For simplicity, we shall assume that $W_a(\vec{v})$ [and consequently $W_a'(\vec{v})$] is an equilibrium distribution given by

$$W_a(v) = W_a'(v) = (\pi u^2)^{-3/2}e^{-v^2/u^2}\,, \tag{57}$$

where u is the most probable speed of the distribution. With this assumption, Eq. (56) may be substituted into Eqs. (51), (52), and (55) (for the gain, saturation parameter, and intensity, respectively) to yield

$$\alpha = -\tfrac{1}{2}\Omega/Q + g^{(1)}\,\mathrm{Re}\mathcal{G}^{(1)}\,, \tag{58}$$

$$\beta = g^{(3)}\,\mathrm{Re}\mathcal{G}^{(3)}\,, \tag{59}$$

and

$$I = \frac{-\tfrac{1}{2}\Omega/Q + g^{(1)}\,\mathrm{Re}\mathcal{G}^{(1)}}{g^{(3)}\,\mathrm{Re}\mathcal{G}^{(3)}}\,, \tag{60}$$

where the factors $g^{(1)}$ and $g^{(3)}$ are defined by

$$g^{(1)} = \tfrac{1}{2}(\Lambda_a/\gamma_a)\Omega\wp^{-2}\hbar^{-1}(\epsilon_0 ku)^{-1} \tag{61}$$

and

$$g^{(3)} = \tfrac{1}{32}(\Lambda_a/\gamma_a)\Omega\wp^{-4}\hbar^{-3}(\epsilon_0 ku)^{-1}(\gamma_a\gamma_b)^{-1} \tag{62}$$

and the dimensionless quantities $\mathcal{G}^{(1)}$ and $\mathcal{G}^{(3)}$ are given by

$$\mathcal{G}^{(1)} = ku(\pi u^2)^{-3/2}\int d^3v\, e^{-v^2/u^2} \times\{\tilde{\gamma}_{ab}(v) + i[\Delta\tilde{\omega}(v) - \vec{k}\cdot\vec{v}]\}^{-1} \tag{63}$$

and

$$\mathcal{G}^{(3)} = \gamma_a\gamma_b ku(\pi u^2)^{-3/2}\int d^3v_2\int d^3v\, e^{-(v_2)^2/u^2}[S_a(\vec{v}_2\to\vec{v}) + S_b(\vec{v}_2\to\vec{v})] \times\{[\tilde{\gamma}_{ab}(v_2) + i(\Delta\tilde{\omega}(v_2) + \vec{k}\cdot\vec{v}_2)]^{-1} + [\tilde{\gamma}_{ab}(v_2) - i(\Delta\tilde{\omega}(v_2) - \vec{k}\cdot\vec{v}_2)]^{-1}\}\{\tilde{\gamma}_{ab}(v) + i[\Delta\tilde{\omega}(v) - \vec{k}\cdot\vec{v}]\}^{-1}\,. \tag{64}$$

[It follows from our choice of even velocity distributions for both the active atoms and perturbers that the $\vec{k}\to-\vec{k}$ terms which appear in Eqs. (46) and (47) are equal to the first terms in those expressions. This fact has been used in writing Eqs. (61)–(64).]

We should point out that, in actual experiments, $W_a(\vec{v})$ may not be an equilibrium distribution and collisions would effect changes in the distribution. In that case, the actual form of $W_a'(\vec{v})$ may be quite complicated, but, since it appears in *both* first- and third-order terms, the details of the intensity profile will probably not depend critically on its form. Thus, unless one is specifically interested in the effect of DMC in the t_1 region, he need not be overly concerned with collisions in this region.

Time region τ''. Unfortunately, the situation for the τ'' time region is not so simple (recall that this interval represents the time between the field interactions at times t_2 and t_3; see Figs. 5). To understand the effects of collisions in this region, one must return to the no-collision Doppler-limit expressions (16a)–(16d) in which all integrands contain the factor $e^{-\gamma_{ab}(\tau'+\tau''')}e^{i\vec{k}\cdot\vec{v}\tau'}e^{-i\vec{k}\cdot\vec{v}\tau'''}$. The reason why these terms give a relatively large contribution is that for all $\tau' = \tau'''$ the Doppler phase factor vanishes. When collisions are incorporated into Eqs. (16), one finds that collisions in the τ'' region (see Fig. 5) will cause the velocity in the τ' region to be different from that in the τ''' region and, in doing so, may somewhat destroy this Doppler phase cancellation. Explicitly, the factor which will appear in the integrands of expressions for $\mathcal{A}^{(3)}(t)$ will be of the form [see Eq. (64)]

$$[S_a(\vec{v}_2\to\vec{v}) + S_b(\vec{v}_2\to\vec{v})]\,e^{-\tilde{\gamma}_{ab}(\tau'+\tau''')} \times e^{i\vec{k}\cdot\vec{v}(\tau'-\tau''')}e^{i\vec{k}\cdot\Delta\vec{v}\tau'''}\,,$$

where $\Delta\vec{v} = \vec{v} - \vec{v}_2$, and $\vec{v}_2$ and $\vec{v}$ are the velocities associated with the τ''' and τ' time regions, respectively. The distribution of the $\Delta\vec{v}$'s produced in the τ'' region is determined by the S_α functions defined by Eq. (48). One easily observes that any

integrals over τ''' containing the above factor will receive significant contributions only for $\tau''' \lesssim [\tilde{\gamma}_{ab}]^{-1}$ and this feature enables one to separate the DMC into three categories. If $|\vec{k}\cdot\Delta\vec{v}| \ll \tilde{\gamma}_{ab}$, the effects of collisions are still negligible. However, if $|\vec{k}\cdot\Delta\vec{v}| \approx \tilde{\gamma}_{ab}$, the additional phase factor $e^{i\vec{k}\cdot\Delta\vec{v}\tau'''}$ must be reckoned with. The modification of the line shape produced by such collisions will depend on the details of collision process. Finally, any collision in which $|\vec{k}\cdot\Delta\vec{v}| \gg \tilde{\gamma}_{ab}$ virtually eliminates all chance for Doppler phase cancellation. Collisions of this type should reduce the contribution of these "Doppler-limit" terms to a magnitude comparable with non-Doppler-limit terms. Thus, if one were interested in the Doppler limit, he would need only make a collision model for the collisions with $|\vec{k}\cdot\Delta\vec{v}| \approx \tilde{\gamma}_{ab}$, since other types of collisions either destroy or do not affect the third-order contributions. On the other hand, if one seriously considers the non-Doppler-limit terms, he must also have a model for the "strong" collisions, $|\vec{k}\cdot\Delta\vec{v}| \gg \tilde{\gamma}_{ab}$.

It is difficult to get a detailed description of the line shape without employing some type of collision model. We choose to discuss one simple collision model. Although we do not claim that this model will provide an extremely accurate description of the actual physical processes involved in the collisions, it is hoped that the line shape calculated by using this model will enable us to draw some general conclusions as to the effects of collisions on the laser intensity profile.

A. Simple Collision Model

Before describing the collision model, we should point out that it will be most effective when applied to systems which conform to the Doppler limit. To this end, we shall adhere to the Doppler limit and evaluate $\mathcal{G}^{(1)}$ [see Eq. (63)] to lowest order in $\tilde{\gamma}_{ab}/ku$ (but for arbitrary detuning $\Delta\tilde{\omega}$) and $\mathcal{G}^{(3)}$ to lowest order in $\tilde{\gamma}_{ab}/ku$ and $\Delta\tilde{\omega}/ku$. The collision model is characterized by the following assumptions. (1) It is assumed that the complex line-shape parameter $\bar{\gamma}_{ab}(v)$ given by Eq. (42a) does not vary appreciably when v changes by an amount $\tilde{\gamma}_{ab}(u)/k$. The consequences of this assumption will be evident shortly. (2) A kind of "hard-sphere" model will be employed for collisions occurring in the τ'' time region. A cutoff parameter $\gamma'_{ab}(u)$ [not necessarily exactly equal to $\tilde{\gamma}_{ab}(u)$] is chosen to divide the collisions into two categories. Collisions with $|\vec{k}\cdot\Delta\vec{v}| < \gamma'_{ab}(u)$ are assumed to produce a negligible effect on $\mathcal{G}^{(3)}$, while collisions with $|\vec{k}\cdot\Delta\vec{v}| > \gamma'_{ab}(u)$ are assumed to be truly "Doppler modifying" and consequently reduce the contribution of $\mathcal{G}^{(3)}$ by order $\tilde{\gamma}_{ab}/ku$. Since terms of relative order $\tilde{\gamma}_{ab}/ku$ are neglected in the Doppler limit, we need only retain the contribution to $\mathcal{G}^{(3)}$ which results when *no* DMC occur in the τ'' region. Analytically, this corresponds to taking only the leading term in Eq. (31) for the propagator $G_{\alpha\alpha}$, i.e.,

$$G_{\alpha\alpha}(\tau'';\vec{v}_2,\vec{v}) = \exp[-\Gamma_\alpha^{\mathrm{DM}}(v)\tau'']\,\delta(\vec{v}-\vec{v}_2)\ .$$

Using Eq. (48), one then may obtain

$$S_\alpha(\vec{v}_2\to\vec{v}) = [\gamma_\alpha + \Gamma_\alpha^{\mathrm{DM}}(v)]^{-1}\delta(\vec{v}-\vec{v}_2)\ . \tag{65}$$

In the Doppler limit and with a proper choice of the cutoff parameter $\gamma'_{ab}(u)$, it seems plausible that this "hard-sphere" model and Eq. (65) will adequately describe the DMC in the τ'' time region.

With the above assumptions we are ready to proceed to calculate $\mathrm{Re}\,\mathcal{G}^{(1)}$, $\mathrm{Re}\,\mathcal{G}^{(3)}$, and the intensity I. Taking a cylindrical coordinate system $(v_\rho,\ v_\theta,\ v_z)$ with the v_z axis in the $\hat{k}$ direction, it follows directly from Eq. (63) that

$$\begin{aligned}\mathrm{Re}\,\mathcal{G}^{(1)} = {} & ku(\pi u^2)^{-3/2}\int_0^\infty 2\pi v_\rho dv_\rho \int_{-\infty}^{\infty} dv_z\, e^{-v_\rho^2/u^2} \\ & \times e^{-v_z^2/u^2}\tilde{\gamma}_{ab}((v_\rho^2+v_z^2)^{1/2}) \\ & \times\{[\gamma_{ab}((v_\rho^2+v_z^2)^{1/2})]^2 \\ & + [\Delta\tilde{\omega}((v_\rho^2+v_z^2)^{1/2}) - kv_z]^2\}^{-1}\ . \end{aligned}\tag{66}$$

The integral over v_z may be easily evaluated in the Doppler limit if assumption (1) of our model is employed. The major contribution to the v_z integral comes from a velocity region of width $\tilde{\gamma}_{ab}/k$ centered about the solution of the equation

$$v_z = k^{-1}\Delta\tilde{\omega}((v_\rho^2+v_z^2)^{1/2})\ . \tag{67}$$

The value of v_z satisfying Eq. (67) will be designated $v_z(\Delta\tilde{\omega})$. Since $\tilde{\gamma}_{ab}(v)$, $\Delta\tilde{\omega}(v)$, and $e^{-v_z^2/u^2}$ are assumed to be slowly varying functions of v_z over the range $\tilde{\gamma}_{ab}(u)/k$, we may evaluate v_z appearing in these functions at $v_z(\Delta\tilde{\omega})$. When this is done, the integral over v_z in Eq. (66) is readily performed to yield

$$\begin{aligned}\mathrm{Re}\,\mathcal{G}^{(1)} = {} & 2\pi^{1/2}u^{-2}\int_0^\infty v_\rho dv_\rho\, e^{-v_\rho^2/u^2} \\ & \times\exp[-\Delta\tilde{\omega}(\{v_\rho^2+[v_z(\Delta\tilde{\omega})]^2\}^{1/2})/ku]^2\ . \end{aligned}\tag{68}$$

Similar techniques may be used to calculate the function $\mathcal{G}^{(3)}$ given in Eq. (64), when S_α is determined by Eq. (65). To zeroth order in $\tilde{\gamma}_{ab}/ku$ and $\Delta\tilde{\omega}/ku$ one obtains

$$\begin{aligned}\mathrm{Re}\,\mathcal{G}^{(3)} = {} & 2\pi^{1/2}u^{-2}\gamma_a\gamma_b\int_0^\infty v_\rho dv_\rho\, e^{-v_\rho^2/u^2} \\ & \times\{[\gamma_a+\Gamma_a^{\mathrm{DM}}(v_\rho)]^{-1}+[\gamma_b+\Gamma_b^{\mathrm{DM}}(v_\rho)]^{-1}\} \\ & \times\{[\tilde{\gamma}_{ab}(v_\rho)]^{-1}+\tilde{\gamma}_{ab}(v_\rho)([\tilde{\gamma}_{ab}(v_\rho)]^2+[\Delta\tilde{\omega}(v_\rho)]^2)^{-1}\}\ . \end{aligned}\tag{69}$$

The intensity is found from Eqs. (60)–(62) to be

$$I=\frac{-\frac{1}{2}\Omega/Q+g^{(1)}\,\mathrm{Re}\mathcal{G}^{(1)}}{g^{(3)}\,\mathrm{Re}\mathcal{G}^{(3)}}\,, \tag{70}$$

where $\mathrm{Re}\mathcal{G}^{(1)}$ and $\mathrm{Re}\mathcal{G}^{(3)}$ are now given by Eqs. (68) and (69), respectively. An examination of Eqs. (68)–(70) reveals that the intensity profile will not, in general, be a symmetric function of cavity detuning due mainly to the dependence of $\Delta\tilde{\omega}$ on v_p. To actually compute the intensity profile indicated by Eq. (70), one must use some functional form for $\tilde{\gamma}_{ab}(v)$ and $\Gamma_\alpha^{\mathrm{DM}}(v)$ and then do the velocity integrals in Eqs. (68) and (69). Such a specific evaluation is not included in this work.

There is one case in which Eq. (70) becomes greatly simplified, and that is when the line-shape parameters do not depend on speed (or vary only slightly as v varies from $v=0$ to $v=u$). This situation is favored when perturber speeds are much greater than active-atom speeds or, equivalently, when the emitter-perturber relative velocity is approximately equal to the perturber velocity. In that case, the integral over v_p in Eq. (42a) will yield a $\bar{\gamma}_{ab}$ which is almost speed independent. If the line-shape parameters do not depend on speed, the intensity [no distinction will be made between the "intensity" I given by Eq. (60) and the intensity measured in experiments—in practice, these quantities are proportional to one another] will be given by

$$I=A(P)\frac{e^{-(\Delta\tilde{\omega}/ku)^2}-[\mathfrak{N}(P)]^{-1}}{1+\tilde{\gamma}_{ab}^2[\tilde{\gamma}_{ab}^2+(\Delta\tilde{\omega})^2]^{-1}}\,, \tag{71}$$

where the relative excitation $\mathfrak{N}$ (which may depend on pressure P) is equal to

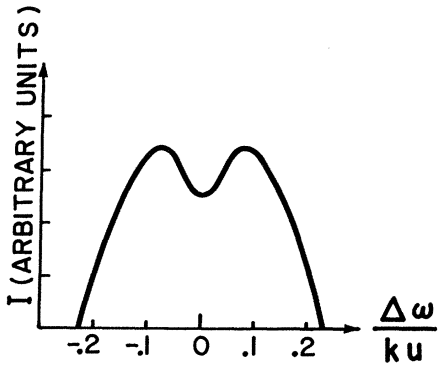

FIG. 7. Graph of laser intensity I versus dimensionless cavity detuning $\Delta\tilde{\omega}/ku$ (that is, cavity detuning divided by Doppler width) for a laser with relative excitation $\mathfrak{N}=1.05$ and dimensionless width parameter $\tilde{\gamma}_{ab}/ku=0.05$. The intensity goes to zero for values of $|\Delta\tilde{\omega}_c/ku|\geq 0.225$, a value which may be obtained using Eq. (75) of the text.

$$\mathfrak{N}(P)=\pi^{1/2}(Q/\Omega)[\Lambda_a(P)/\gamma_a]\Omega\wp^2\hbar^{-1}(\epsilon_0 ku)^{-1} \tag{72}$$

and $A(P)$ is given by

$$A(P)=16(\hbar/\wp)^2\tilde{\gamma}_{ab}(\gamma_a+\Gamma_a^{\mathrm{DM}})(\gamma_b+\Gamma_b^{\mathrm{DM}})\times(\gamma_a+\Gamma_a^{\mathrm{DM}}+\gamma_b+\Gamma_b^{\mathrm{DM}})^{-1}\,. \tag{73}$$

The line shape is a symmetric function of cavity detuning about $\Delta\tilde{\omega}=0$. The intensity at central tuning, $\Delta\tilde{\omega}=0$, denoted by $I_0(P)$, is equal to

$$I_0(P)=\tfrac{1}{2}A(P)\{1-[\mathfrak{N}(P)]^{-1}\}\,, \tag{74}$$

and the intensity goes to zero for a detuning larger than or equal to a cutoff value $|\Delta\tilde{\omega}_c|$ obtained from

$$e^{-(\Delta\tilde{\omega}_c/ku)^2}=[\mathfrak{N}(P)]^{-1}\,. \tag{75}$$

A typical curve for I [Eq. (71)] is shown in Fig. 7, in which the central tuning dip is evident.

Some general features of the line shape given by Eq. (71) or Eqs. (68)–(70) may be noted. The line shape effectively contains three collision parameters ($\mathrm{Re}\bar{\gamma}_{ab}$, $\mathrm{Im}\bar{\gamma}_{ab}$, and $\Gamma_\alpha^{\mathrm{DM}}$) that vary linearly with pressure.[15] Of these, it is experimentally easiest to observe $\mathrm{Re}\bar{\gamma}_{ab}$ and $\mathrm{Im}\bar{\gamma}_{ab}$, since these parameters are, more or less, directly related to the width and shift of the tuning dip shown in Fig. 7 [by "shift" we mean the collision-induced change in the *absolute* frequency (obtained, for example, by beating the laser against a reference laser) at which the center of the tuning dip occurs]. On the other hand, the DMC collision parameter $\Gamma_\alpha^{\mathrm{DM}}$ is more difficult to observe and, as will be discussed below, there is not yet any definitive experimental evidence of its existence. The dependence of $I_0(P)$ on pressure may provide a means of identifying $\Gamma_\alpha^{\mathrm{DM}}$.

We should like to discuss possible improvements and extensions of our model, but defer this discussion until the present experimental situation has been described.

B. Comparison with Experiment

We shall discuss three experiments[16–18] involving He-Ne lasers. In all cases, a quantity corresponding to $\tilde{\gamma}_{ab}$ was found to exist and to exhibit a linear pressure dependence. In none of these experiments was the absolute frequency of the laser determined.

Smith[16] measured intensity profiles for the 6328-Å ($3s_2\rightarrow 2p_4$) neon transition in a He-Ne laser (7 : 1 ratio of helium to neon) and was able to fit his data to an equation of the form

$$I=KF(P)\{e^{-(\Delta\tilde{\omega}/ku)^2}-[\mathfrak{N}(P)]^{-1}\}\times[1+\tilde{\gamma}_{ab}^2/(\tilde{\gamma}_{ab}^2+\Delta\tilde{\omega})^2]^{-1}\,, \tag{76}$$

in which K is a constant, P is the pressure in Torr, $\tilde{\gamma}_{ab}/2\pi=(9+69P)$ MHz, and $F(P)$, as empirically determined by Smith, is shown in Fig. 8.

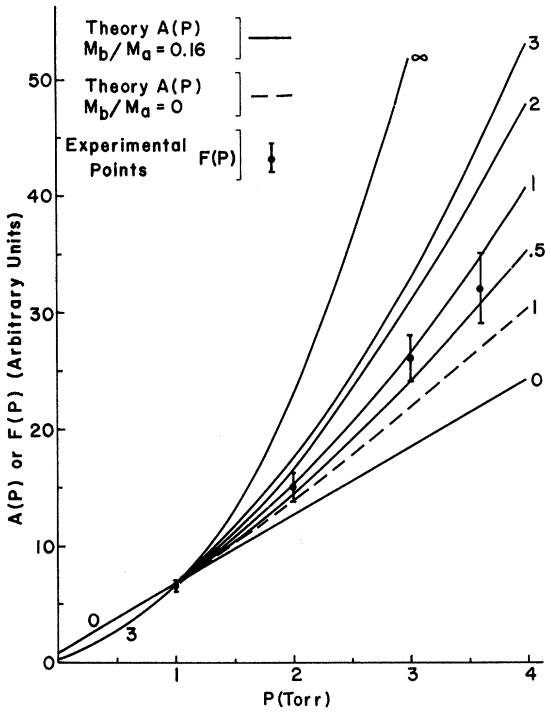

FIG. 8. Curves of $A(P)$ and points for $F(P)$ versus perturber atom pressure. The quantity $A(P)$ is defined by Eq. (73) and is proportional to the laser intensity, while $F(P)$ is a corresponding quantity determined experimentally by Smith (see Ref. 16). The shape of the curves depends on the rates Γ_a^{DM} and Γ_b^{DM} at which Doppler-modifying collisions occur for atoms in states a and b, respectively. These rates may be written as $\Gamma_\alpha^{DM} = M_\alpha \gamma_\alpha P$ ($\alpha = a, b$), where M_α is in Torr^{-1}, γ_α is the natural decay rate of state α, and P is the pressure in Torr. The dashed curve is drawn taking $M_b = 0.0$ Torr^{-1} and $M_a = 1$ Torr^{-1}, while the solid curves are drawn taking $M_b/M_a = 0.16$, with the numbers next to the curves indicating the value of M_a in Torr^{-1}. In the pressure range 0–1 Torr, only the $M_a = 0.0$ Torr^{-1} and the $M_a = 3$ Torr^{-1} curves have been shown. All the other curves will lie between these two, with the exception of $M_a = \infty$ Torr^{-1}, which lies below the $M_a = 3$ Torr^{-1} curve and passes through the origin. The $A(P)$ curves have been normalized to agree with $F(P)$ at $P = 1$ Torr.

(Since no absolute frequency measurements were made, it was impossible to ascertain the pressure dependence of $\Delta\tilde{\omega}$.) In practice, $F(P)$ was experimentally obtained as

$$F(P) = I_0[1 - \mathfrak{N}^{-1}(P)]^{-1}K^{-1} , \tag{77}$$

where I_0 is the measured intensity at zero detuning and $\mathfrak{N}(P)$ is the relative excitation at pressure P determined by the cutoff detuning $\Delta\tilde{\omega}_c$ at that pressure [see Eq. (75)].

A comparison of Eqs. (71) and (76) will show that for theory and experiment to be in agreement the pressure dependence of $F(P)$ and $A(P)$ must be of the same form. In Fig. 8 we have plotted a dashed theoretical curve for $A(P)$ [see Eq. (73)] assuming[19] $\gamma_a/2\pi = 8.3$ MHz, and $\gamma_b/2\pi = 9.75$ MHz and assuming a scattering only,[20] with Γ_a^{DM} given by

$$\Gamma_a^{DM} = M_a \gamma_a P . \tag{78}$$

The quantity M_a (in units of Torr^{-1}) is a parameter determining the relative importance of the DMC. It turns out that for a scattering only, $A(P)$ is not very sensitive to the value of M_a, and in plotting the dashed theoretical curve of Fig. 8 we have taken a value $M_a = 1$ Torr^{-1}, for reasons that will be evident shortly. It should also be noted that we have normalized our value of $A(P)$ to the $P = 1$ point of Smith's data.

In Fig. 8, we have also plotted solid theoretical values for $A(P)$ assuming *both* Γ_a^{DM} and Γ_b^{DM} are nonzero. Immediately, one may argue that this assignment contradicts the basic premise of the PCM: collision interaction in one state only. However, as long as $\Gamma_a^{DM} \gg \Gamma_b^{DM}$ or $\Gamma_b^{DM} \gg \Gamma_a^{DM}$, there is considerable theoretical basis (to be discussed in Sec. VIII) for taking both Γ_a^{DM} and Γ_b^{DM} nonzero but still allowing only phase-shifting collisions (and no DMC-phase collisions) to affect off-diagonal density matrix elements. Hence we assume that

$$\Gamma_b^{DM} = (M_b/M_a) M_a \gamma_b P , \tag{79}$$

where the ratio M_b/M_a indicates the relative strength of the DMC for atoms in state b to that for atoms in state a. If Eqs. (78) and (79) are substituted into Eq. (73), one obtains

$$A(P) = \frac{(\text{const})(1 + M_{ab}P)(1 + M_a P)[1 + (M_b/M_a)M_a P]}{1 + \{\gamma_a(1 + M_a P) + \gamma_b[1 + (M_b/M_a)M_a P]\}/2\gamma_{ab}} , \tag{80}$$

where M_{ab} is defined by

$$\tilde{\gamma}_{ab} = \gamma_{ab}(1 + M_{ab}P) \tag{81}$$

and is equal to 7.7 Torr^{-1} in Smith's experiment. Using the values of γ_a and γ_b given above and taking[21] $M_b/M_a = 0.16$, we graph $A(P)$ vs P in Fig. 8 for several values of M_a. In this case, one can see that $A(P)$ *is* sensitive to M_a with the best values of M_a obtained in the range $M_a = 0.5$ Torr^{-1} to $M_a = 1.5$ Torr^{-1}. [These curves of $A(P)$ have been normalized to fit the experimental point at $P = 1$.]

As can be seen from Fig. 8, there is qualitative agreement between theory and experiment. The curvature of $F(P)$ offers some evidence for the existence of Γ_α^{DM}, although this evidence is certainly not conclusive.[22]

A similar experiment was performed by Holt,[17] who studied the 1.15-μ ($2s_2 \rightarrow 2p_4$) Ne transition in pure neon and He-Ne lasers. Part of this study was directed towards establishing the regions of

validity of the third-order perturbative approach to the laser problem. It was found that, for relative excitations $\mathfrak{N} \lesssim 1.10$ and for $\tilde{\gamma}_{ab}/ku \lesssim 0.14$ (or for $\mathfrak{N} \lesssim 1.17$ and $\tilde{\gamma}_{ab}/ku \lesssim 0.08$), the *shape* of the theoretical curves of I_0 vs P for the high-intensity and third-order theories was the same, justifying the use of Eq. (74) in searching for DMC effects.[23] However, the comparison of the third-order and high-intensity theories also indicates that the dependence of intensity on detuning at a fixed pressure given by Eq. (71) may be in error by as much as 20% for relative excitations $\mathfrak{N} \gtrsim 1.05$. Thus, any detailed agreement between Eq. (71) and experiment at all but very low relative excitations is not particularly meaningful. Experimentally, values of I_0 vs P for fixed $\mathfrak{N}^{-1}$ were measured. While these measurements support the fact that $A(P)$ [see Eqs. (74) and (73)] depends linearly on $\tilde{\gamma}_{ab}$, they are not precise enough to determine whether or not a $\Gamma_\alpha^{\mathrm{DM}} \neq 0$ is also in evidence.[24]

Finally, there is the experiment of Cordover and Bonczyk,[18] in which intensity-versus-detuning curves were measured at several pressures. It was found that the experimental data could be fitted (remarkably well) to a five-parameter expression for the intensity. This expression was based, in part, on a theory of collision effects in lasers proposed by Szöke and Javan.[25] We do not find the excellent agreement between the theory of Szöke and Javan and the experimental results of Cordover and Bonczyk to be particularly significant, because (a) the collision theory of Szöke and Javan is highly phenomenological at best and (b) the experimental data were taken for relative excitations $\mathfrak{N} > 1.2$, where, as discussed above, one would not expect a third-order theory (as is Szöke and Javan's) to be highly accurate.

In the same experiment, a slight asymmetry was observed in the dependence of intensity on detuning. Such asymmetries will develop in our theory when it is extended to include the possibilities that (1) the line-shape parameter $\bar{\gamma}_{ab}$ depends on v [see the discussion following Eq. (70)], (2) there is a comparable collision interaction for both radiative states allowing for DMC-phase-type collisions which, it turn, can lead to asymmetries (see Appendix A), and (3) many-body collisions provide a correction to the binary-collision theory.[26] Which, if any, of these effects leads to the observed asymmetry is difficult to ascertain. Since the asymmetries are very slight, one must also be careful to remove all spurious experimental sources for asymmetry (e.g., isotopic impurities of the active atom or a frequency dependence of the laser window transmitivity could lead to asymmetries in the intensity-versus-detuning curves).

To summarize, the experiments have demonstrated that Eq. (71) provides a fairly good description of observed laser phenomena. The existence of a line-shape parameter $\tilde{\gamma}_{ab}$ which varies linearly with pressure has been established, but experimental confirmation of $\Gamma_\alpha^{\mathrm{DM}}$ is not yet conclusive.

C. What Next?

There are several obvious extensions of the theory which may be attempted to widen its regions of validity. First, one could try to develop an accurate collision kernel $W_\alpha(\vec{v} \to \vec{v}')$ by using any theoretical or experimental information about the scattering process that is available. Even if an accurate kernel could be obtained, the remaining chain of calculations would still be formidable. That is, from $W_\alpha(\vec{v} \to \vec{v}')$ one must find, in turn, $G_{\alpha\alpha}(\tau; \vec{v}, \vec{v}')$ using Eq. (31), $S_\alpha(\vec{v} \to \vec{v}')$ using Eq. (48), and finally $\mathcal{G}^{(3)}$ using Eq. (64). We do not believe that use of a more sophisticated collision model will substantially alter the form of the results that have been obtained using the "hard-sphere" model of Sec. VII B as long as one is considering the case of collision interaction in one state only in the Doppler limit. Second, one may wish to abandon the Doppler limit and calculate the third-order polarization to higher order in $\tilde{\gamma}_{ab}/ku$ and $\Delta\tilde{\omega}/ku$. In that case, he would have to evaluate all the non-Doppler-limit terms in Appendix B, which is a difficult task even for the simplest collision model. In addition, all terms or relative order $\tilde{\gamma}_{ab}/ku$ will depend strongly on the type of collision kernel used in the calculation, so that for this situation it would be advantageous to use an accurate collision model in formulating the theory. Since this, in turn, implies the difficult calculation mentioned above, one can see that leaving the simplicity of the Doppler limit will lead to severe additional problems. Third, one may wish to account for the possibility of a comparable collision interaction for both the atomic states involved in the laser transition. We shall defer discussion of this case to Sec. VIII. Fourth, a strong signal theory[17,27,28] may be sought to provide an adequate description of the line shape at higher relative excitations. Although such a theory might be developed for highly simplified collision kernels, it is not likely the calculation will be at all practical for more realistic kernels. Finally, and perhaps most feasible, one may try to perform an explicit evaluation of Eqs. (68) and (69) by doing the specified integrations over v_p which appear in these equations. To this end, the line-shape parameter $\bar{\gamma}_{ab}(v) = \tilde{\gamma}_{ab}(v) - \gamma_{ab} + i[\Delta\tilde{\omega}(v) - \Delta\omega]$ may be determined using Eq. (41a) with a classical phenomenological collision interaction (e.g., Lennard-Jones). The resulting line shape given by Eqs. (68)–(70) will then display the asymmetry to be expected from the speed dependence of the line-shape parameters.

It can be inferred from the above discussion that

attempts to extend the theory will be characterized by very complicated calculations. In light of this fact, we hope that future laser experiments intending to probe collision effects will be carried out under conditions when the present theory is valid. By measuring intensity-versus-detuning and I_0-vs-P curves at low relative excitations and at the lowest possible pressures in systems where γ_{ab}/ku is minimal, one may be able to obtain conclusive checks on the theory in general and the existence of Γ_α^{DM} in particular. To help reduce any modifications in the line shape arising from resonant collision broadening or discharge current effects,[29] these experiments should be carried out with a large ratio of buffer to active-atom gas pressures.[30,31] Since there is so little known about the details of collision processes in atomic systems, any information derived from such experiments would prove extremely useful and, in addition, may point out any failures of the theory. If and when the results of these idealized experiments can be understood, we shall be in a better position to extend the theory to cover more complicated experimental situations.

VIII. EVALUATION OF THEORY

In this paper we have developed the pseudoclassical collision model (PCM), which is designed to enable one to easily incorporate pressure effects into problems involving the interaction of radiation and matter. In particular, the problem of a laser has been solved to third order in the laser field and an expression for the intensity profile obtained. Including the calculations in the appendices, our result is quite general for a laser slightly above threshold. That is, an allowance has been made for incoherent excitation of laser atoms to state a or b, collisional scattering for both laser radiative states, and arbitrary ratios of decay parameters and detuning to some effective Doppler width (inclusion of non-Doppler-limit terms). No attempt to explicitly evaluate this general line shape was made, since we were content to concentrate our work on the case of collision interaction in one state only for systems in which the Doppler limit was applicable. Within these limitations, a very simple collision model was developed and found to be in rough agreement with experiment. There must certainly be additional theoretical and experimental research if pressure effects are to be fully understood. Several suggestions along these lines were presented in Sec. VII.

The basis for the PCM, as described in Sec. III, is an interpretation of the perturbation diagrams in light of the calculated one-collision quantum-mechanical result of QMI. The expressions which have been obtained reduce to the one-collision result of QMI in the limit of very low pressure and give the "impact" theory line-shape parameters of Lindholm[4] and Foley[4] or of Baranger[5] if we neglect velocity-changing collisions. An alternative to using the PCM is to extend the quantum-mechanical calculation of QMI to allow for many collisions of the active atom in its lifetime—a difficult undertaking at best.

There are certain limitations of the PCM which may be noted.

(1) Strictly speaking, we cannot apply the PCM to the situation where there is scattering interaction in both levels. In Appendix A, we present a calculational model intended to cover this case, but the only rationale for this model is that it seems to work and is "reasonable." That is, at low pressures it reduces to the one-collision result of QMI, and it also reduces to the proper limits for the three cases of (a) single-state scattering, (b) neglect of velocity-changing collisions, and (c) equal scattering interactions in both levels.

Determining whether or not one need consider collision interaction in both laser states is a problem requiring further study. That is, even though the collision interaction for one state may be much stronger than that for the other (as is assumed in this work), the cross section for DMC in the weakly interacting state may be large enough to cause such collisions to have a measurable effect on the intensity profile. The problem can be divided into two parts: the effects of collision interaction in both states on $G_{\alpha\alpha}(\tau;\vec{v},\vec{v}')$ (that is, on time regions where, in the diagrammatic sense, only a diagonal density matrix element is nonzero) or on $G_{ab}^{\pm}(\tau';\vec{v},\vec{v}';\vec{k})$ (that is, on time regions where only an off-diagonal density matrix element is nonzero). Calculations of $G_{\alpha\alpha}(\tau;\vec{v},\vec{v}')$ will not be altered if there is collision interaction in both states, although both Γ_α^{DM} and Γ_b^{DM} would be nonzero in this case. Thus the calculation of this paper is quite general as far as the "diagonal" propagators are concerned. It is for this reason that we were able to take both Γ_a^{DM} and Γ_b^{DM} nonzero in drawing the theoretical curve in Fig. 8.

On the other hand, $G_{ab}^{\pm}(\tau;\vec{v},\vec{v}';\vec{k})$ can be significantly altered if there is scattering interaction in both states, as discussed in Appendix A. It is our contention, however, that *changes in* $G_{ab}^{\pm}(\tau;\vec{v},\vec{v}';\vec{k})$ *will not be appreciable if the collision interaction for one state is somewhat greater than that for the other,* despite the fact that the DMC cross section for the more weakly interacting state is not negligible. While this contention should be proved rigorously, we feel that a reasonable argument may be given for its validity. Let $\Delta\vec{v}_w$ and $\Delta\vec{v}_s$ be the velocity changes caused by a collision for an atom in the weakly and strongly interacting states w and s, respectively. If a collision is to be Doppler modifying for an off-diagonal density matrix element

ρ_{ws}, then both $\Delta\vec{v}_w$ and $\Delta\vec{v}_s$ must correspond to a DMC. To ensure this, it is sufficient to have

$$|\vec{k}\cdot\Delta\vec{v}_w| \gtrsim \tilde{\gamma}_{ws} . \tag{82}$$

Since the rate for phase-shifting collisions[32] is $\gtrsim \Gamma_s^{DM}$ and the rate for DMC-phase collisions is $\approx \Gamma_w^{DM}$ [because of the requirement of Eq. (82)], the relative importance of DMC-phase to phase-shifting collisions is $\lesssim \Gamma_w^{DM}/\Gamma_s^{DM}$. If the ratio is small, DMC-phase collisions can be treated as pure phase-shifting collisions to a first approximation. Note that the DMC-phase collisions would enter the line-shape expression as a correction of order $\Gamma_w^{DM}/\Gamma_s^{DM}$ at most, but that the DMC collisions for the weakly interacting state will alter the intensity formula [see Eqs. (71) and (73)] by a factor $1+\Gamma_w^{DM}/\gamma_w$, where γ_w is the natural width of the weakly interacting state. Whereas $\Gamma_w^{DM}/\Gamma_s^{DM}$ will, in general, be small and provide only a slight modification to the line shape, Γ_w^{DM}/γ_w may be comparable to unity at pressures as low as a few Torr and, as such, may lead to significant changes in the pressure dependence of the intensity. This feature is easily seen in Fig. 8.

Thus, we have advocated a generalized PCM in which DMC are allowed in all regions where diagonal density matrix elements are nonzero (in the diagrammatic sense), but only phase-shifting collisions are allowed in regions where off-diagonal density matrix elements are nonzero. This generalized PCM will be valid if the collision interaction for one of the radiative states is somewhat stronger than that for the other. If the collision interactions for the two states are comparable, one must use the calculation of Appendix A.

As an interesting sidelight to this discussion, we should mention that it may be possible to determine the relative importance of collision interaction in both states by an independent experimental check. If linear absorption or emission experiments are performed on the laser transition under investigation, one can obtain a linewidth (including Doppler width) which will be pressure dependent. If the collision interaction in the two radiative states were nearly equal, the linewidth would *decrease* with increasing pressure, a phenomenon caused by the DMC and known as "Dicke narrowing."[33] On the other hand, if one state experiences a relatively weak collision interaction, there are very few DMC-phase collisions, Dicke narrowing may occur only at relatively high pressures, and there is an increase in linewidth with pressure due to the phase-shifting collisions given by $w \approx w_0 + \mathrm{Re}\bar{\gamma}_{ab}$, where w_0 is the width at zero pressure and $\bar{\gamma}_{ab}$ is proportional to pressure. Thus, by observing the change in width with pressure, one may be able to predict the relative importance of collision interaction in both states. It is interesting to note that the phenomenon of Dicke narrowing, which is used so widely in microwave resonance experiments to reduce the Doppler width (in such experiments, the atomic levels involved in the transition belong to the same fine-structure level and *do* experience about the same collisional interaction), may play only a minor role in determining the widths of optical spectral lines (which result from transitions between electronic levels).

(An alternative approach to an experimental determination of the relative importance of the collision interaction experienced by the two laser states is afforded by measuring the pressure dependence of the absolute frequency at the center of the tuning dip. If collision interaction for the two states were equal, there would be no shift on this frequency with pressure, while a shift comparable with $\mathrm{Re}\bar{\gamma}_{ab}$ would indicate that one state is experiencing a much stronger collision interaction than the other.)

(2) Another inherent difficulty of the PCM is its connection with perturbation theory. Thus, if one desired to go to higher order in the laser field, he would have to draw the fifth-, seventh-, etc., order diagrams and put collisions into these results. This type of perturbation solution of the laser problem is not the most convenient for examining a high-intensity laser and it appears that the PCM is impractical for such a case. However, it may be possible to directly incorporate the PCM into the differential equations of laser theory and then solve the resulting integrodifferential equations by techniques other than the iterative approach used in this paper.[34]

(3) A third drawback of the PCM is its limitation to binary-collision pressure regions. Only in the binary-collision approximation can one discuss separate collisions, a feature vital to the PCM. If one wished to study the higher pressures where many particle collisions become important, he would probably do better to try a different approach to the problem. Lasers and many other systems do operate in the binary-collision pressure region, although many-body collisions might produce slight corrections to the theory.

(4) A less serious criticism of the PCM may be leveled against the use of cutoffs on the integrals to separate out Doppler-modifying velocity collisions from other negligible velocity-changing collisions. This cutoff was taken when the change in velocity $\Delta\vec{v}$ was such that $|\vec{k}\cdot\Delta\vec{v}| = \tilde{\gamma}_{ab}$, but the cutoff is really quite arbitrary.[35] Thus, if one is worried about many small $\Delta\vec{v}$ giving a significant effect, he could take a cutoff at $|\vec{k}\cdot\Delta\vec{v}| = 0.01\tilde{\gamma}_{ab}$ (provided that the binary-collision approximation is still valid). The effect of changing the cutoff as described above would be to increase the rate of

"Doppler-modifying" collisions (since one would be including many more collisions in the "Doppler-modifying" category) but to decrease the effect of an average collision (since many of these "Doppler-modifying" collisions are now very weak). The combination of these two factors will probably not make the result very dependent on the cutoff as long as it is taken at $|\vec{k}\cdot\Delta\vec{v}| \approx \bar{\gamma}_{ab}$.

In summary, the pseudoclassical collision model should provide a simple means for including the effects of velocity-changing and phase-shifting collisions in many atomic physics problems. For example, the problem of determining spontaneous emission profiles in the presence of collisions is very much simplified by the use of the PCM. It may also be possible to extend the PCM to inelastic collision processes such as those which must be considered in Hanle effect (with collisions) and resonant broadening experiments.

APPENDIX A

In this appendix, we present a calculational model for treating the collision problem when there is scattering interaction for the two radiative states.

In time regions where diagonal density matrix elements are nonzero (see Fig. 3 or Fig. 5), nothing is changed from the PCM. That is, the propagators $G_{aa}(\tau;\vec{v},\vec{v}')$ and $G_{bb}(\tau;\vec{v},\vec{v}')$ are still given by Eq. (31) [in the PCM, either $G_{aa}(\tau;\vec{v},\vec{v}')$ or $G_{bb}(\tau;\vec{v},\vec{v}')$ was proportional to $\delta(\vec{v}-\vec{v}')$, depending on which state experienced no scattering interaction—in the present discussion, as in the generalized PCM of Sec. VIII, neither $G_{aa}(\tau;\vec{v},\vec{v}')$ nor $G_{bb}(\tau;\vec{v},\vec{v}')$ is proportional to $\delta(\vec{v}-\vec{v}')$, since there is scattering interaction for both the radiative states]. The only difficulty lies in evaluation of the propagators $G^{\pm}_{\alpha\alpha'}(\tau;\vec{v},\vec{v}';\vec{k})$ when $\alpha\neq\alpha'$. We need deal only with $G^{+}_{ab}(\tau;\vec{v},\vec{v}';\vec{k})$, since

$$G^{-}_{ab}(\tau;\vec{v},\vec{v}';\vec{k}) = G^{+}_{ab}(\tau;\vec{v},\vec{v}';-\vec{k}) \tag{A1}$$

and

$$G^{\pm}_{ba}(\tau;\vec{v},\vec{v}';\vec{k}) = [G^{\mp}_{ab}(\tau;\vec{v},\vec{v}';\vec{k})]^{*} . \tag{A2}$$

For scattering interaction in one state only $G^{+}_{ab}(\tau;\vec{v},\vec{v}';\vec{k}) \propto \delta(\vec{v}-\vec{v}')$, but this feature is lost when there is scattering interaction in both states.

In order to treat this case, it is necessary to distinguish between DMC-phase and phase-shifting collisions (see Sec. III for definitions of these terms). Whereas in the PCM there was a real collision kernel $W_{\alpha}(\vec{v}\rightarrow\vec{v}') \propto |f_{\alpha}(\vec{v}\rightarrow\vec{v}')|^2$ associated with the DMC, it will turn out that the quantity which plays the role of a "collision kernel" for the DMC-phase collisions will be complex and given by $\bar{W}_{\alpha\alpha'}(\vec{v}\rightarrow\vec{v}') \propto f_{\alpha}(\vec{v}\rightarrow\vec{v}')f_{\alpha'}(\vec{v}\rightarrow\vec{v}')^{*}$. Except for the presence of this complex kernel and permitting *both* DMC-phase and phase-shifting collisions to occur in regions where $\rho_{ab}(\vec{R},t)\neq 0$, the calculation proceeds as in the PCM.

To obtain $G^{+}_{ab}(\tau';\vec{v}_i,\vec{v}_{i+1};\vec{k})$, one must compute $\mathcal{A}^{+}_{ab}(\tau';\vec{v}_i,\vec{v}_{i+1})$, which is the density (in velocity space) representing the collision-modified value of $\mathcal{A}^{+}_{ab}(\tau',\vec{v})$ averaged over all possible collision histories in the τ' region which begin with the active atom having velocity $\vec{v}_i$ and end with it having velocity $\vec{v}_{i+1}$. From the results of QMI, it is already possible to recognize the structural form of $\mathcal{A}^{+}_{ab}(\tau';\vec{v}_i,\vec{v}_{i+1})$ and to conclude that the average over collision histories for the collision-modified value of $\mathcal{A}^{+}_{ab}(\tau',\vec{v})$ may be performed in a manner analogous to that given in Eq. (24). The only difference is that the "collision kernel" as well as the "probability" for no occurrence of these DMC-phase collisions are complex quantities. Using this observation and realizing that if no DMC-phase collisions occur in a time interval $\tau_2-\tau_1$, then the average over all possible *phase-shifting* collisions for this interval leads to a factor $\exp[-\bar{\gamma}_{ab}(v)\times(\tau_2-\tau_1)]$ [where $\vec{v}$ is the velocity at the beginning (and end) of the interval], one may compute the collision-averaged value of [see Eq. (32)]

$$\mathcal{A}^{+}_{ab}(\tau',\vec{v}) = \mathcal{A}_{ab}(\tau')\, e^{i\vec{k}\cdot\vec{v}\tau'}$$

to be

$$\mathcal{A}^{+}_{ab}(\tau',\vec{v}_i,\vec{v}_{i+1}) = G^{+}_{ab}(\tau';\vec{v}_i,\vec{v}_{i+1};\vec{k})\mathcal{A}_{ab}(\tau') ,$$

where

$$\begin{aligned}
G^{+}_{ab}(\tau';\vec{v}_i,\vec{v}_{i+1};\vec{k}) &= \bar{P}^{0}_{ab}(\tau',v_i)\exp[-\bar{\gamma}_{ab}(v_i)\tau']\, e^{i\vec{k}\cdot\vec{v}_i\tau'}\,\delta(\vec{v}_i-\vec{v}_{i+1}) \\
&+ \int_0^{\tau'} d\tau_1 \bar{P}^{0}_{ab}(\tau_1,v_i)\exp[-\bar{\gamma}_{ab}(v_i)\tau_1]\bar{W}_{ab}(\vec{v}_i\rightarrow\vec{v}_{i+1})\bar{P}^{0}_{ab}(\tau'-\tau_1,v_{i+1}) \\
&\times\exp[-\bar{\gamma}_{ab}(v_{i+1})(\tau'-\tau_1)]\exp\{i\vec{k}\cdot[\vec{v}_i\tau_1+\vec{v}_{i+1}(\tau'-\tau_1)]\} \\
&+ \int d^3v \int_0^{\tau'} d\tau_2 \int_0^{\tau_2} d\tau_1\, \bar{P}^{0}_{ab}(\tau_1,v_i)\exp[-\gamma_{ab}(v_i)\tau_1]\bar{W}_{ab}(\vec{v}_i\rightarrow\vec{v})\bar{P}^{0}_{ab}(\tau_2-\tau_1,v)
\end{aligned}$$

$$\times \exp[-\bar{\gamma}_{ab}(v)(\tau_2-\tau_1)]\bar{W}_{ab}(\vec{v}\to\vec{v}_{i+1})\bar{P}^0_{ab}(\tau'-\tau_2, v_{i+1})\exp[-\bar{\gamma}_{ab}(v_{i+1})(\tau'-\tau_2)]$$

$$\times \exp\{i\vec{k}\cdot[\vec{v}_i\tau_1+\vec{v}(\tau_2-\tau_1)+\vec{v}_{i+1}(\tau'-\tau_2)]\}+\cdots , \quad \text{(A3)}$$

where $\bar{P}^0_{ab}(\tau, v_i)$ is the complex "probability" for no DMC-phase collisions to occur in a time τ for an atom moving with velocity $\vec{v}_i$, and $\bar{W}_{ab}(\vec{v}_i\to\vec{v}_{i+1})$ is the complex collision kernel for DMC-phase collisions [the average over all possible types of collisions, similar to that in Eq. (30), leading to the change $\vec{v}_i\to\vec{v}_{i+1}$ is already included in $\bar{W}_{ab}(\vec{v}_i\to\vec{v}_{i+1})$]. In this case, we must also keep track of the Doppler factor $e^{i\vec{k}\cdot\vec{v}\tau'}$ since the velocity of the atom is changing in the interval. Equation (A3) for $G^+_{ab}(\tau;\vec{v},\vec{v}';\vec{k})$, used in conjunction with Eqs. (A1) and (A2), may be substituted in Eqs. (38)–(40), transforming them into results which we assert to be valid for collision interaction in both states.

Equation (41a) is still the appropriate classical expression for $\bar{\gamma}_{ab}(v)$, but no classical formulas for $\bar{P}^0_{ab}(\tau, v)$ and $\bar{W}_{ab}(\vec{v}\to\vec{v}')$ may be given. To achieve agreement with the one-collision results of QMI, we must choose the quantum-mechanical line-shape parameters as [compare with Eqs. (42)]

$$\bar{\gamma}_{ab}(v_i)=\bar{\gamma}_{ba}(v_i)^*=\int d^3v_p W_p(v_p)[\tfrac{1}{2}\bar{\Gamma}^{\rm ph}_a(v_i^r)+\tfrac{1}{2}\bar{\Gamma}^{\rm ph}_b(v_i^r)^*-\mathfrak{N}v_i^r\int d\Omega_{v^r_{i+1}} f_a(\vec{v}_i^r\to\vec{v}^r_{i+1})f_b(\vec{v}_i^r\to\vec{v}^r_{i+1})^*] , \quad \text{(A4a)}$$

$$\bar{W}_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})=\mathfrak{N}(m/\mu)^3\int d^3v_p W_p(v_p)\int d^3v^r_{i+1}\,\delta(\vec{v}^r_{i+1}+(m/m_p)\vec{v}_i+\vec{v}_p-(m/\mu)\vec{v}_{i+1})$$

$$\times \delta(v_i^r-v^r_{i+1})(v_i^r)^{-1}f_\alpha(\vec{v}_i^r\to\vec{v}^r_{i+1})f_{\alpha'}(\vec{v}_i^r\to\vec{v}^r_{i+1})^* , \quad \text{(A4b)}$$

and

$$P^0_{\alpha\alpha'}(\tau, v_i)=\exp[-\bar{\Gamma}^{\rm DM}_{\alpha\alpha'}(v_i)\tau] , \quad \text{(A5a)}$$

with

$$\bar{\Gamma}^{\rm DM}_{\alpha\alpha'}(v_i)=\int' d^3v_{i+1}\bar{W}_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1}) \quad \text{(A5b)}$$

$$=\mathfrak{N}\int' d^3v_p W(v_p)v_i^r\int' d\Omega_{v^r_{i+1}}$$

$$\times f_\alpha(\vec{v}_i^r\to\vec{v}^r_{i+1})f_{\alpha'}(\vec{v}_i^r\to\vec{v}^r_{i+1})^* . \quad \text{(A5c)}$$

The expression for $\bar{\gamma}_{ab}(v_i)$ may be shown to agree with the quantum-mechanical calculation of Baranger [see Eq. (82) and Ref. 21 of QMI]. Of course, the previous expressions for $W_\alpha(\vec{v}\to\vec{v}')$ and $\Gamma^{\rm DM}_\alpha(v)$ given by Eqs. (41b) and (41c) or Eqs. (42c) and (42d) must still be retained. [Notice that, by setting $\alpha=\alpha'$ in Eqs. (A4b) and (A5), one obtains $\bar{W}_{\alpha\alpha}(\vec{v}\to\vec{v}')=W_\alpha(\vec{v}\to\vec{v}')$ and $\bar{\Gamma}^{\rm DM}_{\alpha\alpha}(v)=\Gamma^{\rm DM}_\alpha(v)$, so that these equations may describe both DMC and DMC-phase collisions.]

Formally we may separate the velocity and phase changes occurring in collisions by writing

$$\bar{W}_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})=W_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})\times\exp[i\eta_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})] , \quad \text{(A6)}$$

where $W_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})$ is a real collision kernel and $\eta_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})$ is the effective average phase shift of a DMC-phase collision which changes the velocity associated with $\rho_{\alpha\alpha'}$ from $\vec{v}_i$ to $\vec{v}_{i+1}$. The quantity $W_{\alpha\alpha'}(\vec{v}_i\to\vec{v}_{i+1})$ has no classical analog except when there is equal collision interaction for the two states. In that case $f_a(\vec{v}\to\vec{v}')=f_b(\vec{v}\to\vec{v}')$, $\eta_{\alpha\alpha'}(\vec{v}\to\vec{v}')=0$, and a classical trajectory can be associated with collisions since the collision interaction is state independent. Equation (A6) shows that the probability for a given velocity change is correlated with the effective average phase shift produced by that change. Hence the velocity changes and phase shifts are "statistically dependent" and may lead to considerable asymmetry in the line shapes.[36] The absence of marked asymmetry in laser intensity profiles lends support to the theory that DMC-phase collisions are relatively unimportant.

To summarize, we see that, in general, three types of collisions are possible: normal DMC for diagonal density matrix elements, phase-shifting collisions (no velocity change) for off-diagonal density matrix elements, and DMC-phase collisions for off-diagonal density matrix elements. In a sense, these are the three types of possible *classical* collisions discussed in Sec. III. However, the connection of the collision parameters $\bar{W}_{\alpha\alpha'}(\vec{v}\to\vec{v}')$ and $\bar{\Gamma}^{\rm DM}_{\alpha\alpha'}(v_i)$ with corresponding classical quantities seems remote. It would certainly be advantageous to have a clearer picture of the collision process when there is scattering interaction in both states.

One satisfying (and necessary) feature of the results is that they give correct limiting forms for the following cases: (a) collision interaction in one state only, (b) "impact theory" limit (neglect of all velocity-changing collisions), (c) Boltzmann-equa-

tion approach limit (that is, when $f_a = f_b$, so that $\bar{\gamma}_{ab} = 0$ and the scattering process is state independent), and (d) very low-pressure limit (agrees with one-collision result of QMI). The confidence we have in this calculational model is based, to a great extent, on its success for these limiting cases.

APPENDIX B

In this appendix, we generalize our PCM results to allow for inclusion of non-Doppler-limit terms and excitation of laser atoms to state b (see Fig. 1).

Non-Doppler-Limit Terms

In writing down the third-order contributions to the no-collision results, two types of non-Doppler-limit terms were excluded. The first of these leads to expressions identical with Eqs. (16), but with $\exp[i\vec{k}\cdot\vec{v}_0(\tau'+\tau''')]$ replacing $\exp[i\vec{k}\cdot\vec{v}_0(\tau'-\tau''')]$ in the integrands of these equations. Following the method used in Sec. V, it is a straightforward matter to calculate the contribution to this non-Doppler-limit term when collisions are present. We find

$$\mathcal{A}^{(3)}_{\tau'+\tau'''}(t) = \tfrac{1}{64} i\wp^3\hbar^{-3}V^{-1}[E(t)]^3 \int d^3v_0 \int d^3v_2 \int d^3v W_a(v_0)\, S_a(\vec{v}_0\to\vec{v}_2)[S_a(\vec{v}_2\to\vec{v}) + S_b(\vec{v}_2\to\vec{v})]$$

$$\times\{[\tilde{\gamma}_{ab}(v_2) + i(\Delta\tilde{\omega}(v_2) - \vec{k}\cdot\vec{v}_2)]^{-1} + [\tilde{\gamma}_{ab}(v_2) - i(\Delta\tilde{\omega}(v_2) + \vec{k}\cdot\vec{v}_2)]^{-1}\}[\tilde{\gamma}_{ab}(v) + i(\Delta\tilde{\omega}(v) - \vec{k}\cdot\vec{v})]^{-1} + [(\text{term})\vec{k}\to -\vec{k}] \quad \text{(B1)}$$

The other non-Doppler-limit term that enters the calculation leads to expressions identical with Eqs. (16), but with $\exp[i\vec{k}\cdot\vec{v}_0(\tau'+2\tau''+\tau''')]$ replacing $\exp[i\vec{k}\cdot\vec{v}_0(\tau'-\tau''')]$ in the integrands of these equations. The calculation with collisions proceeds as in Sec. V, except that there is now a velocity-dependent term in the τ'' region. It is easy to obtain

$$\mathcal{A}^{(3)}_{\tau'+2\tau''+\tau'''}(t) = \tfrac{1}{64} i\wp^3\hbar^{-3}V^{-1}[E(t)]^3 \int d^3v_0 \int d^3v_2 \int d^3v\, W_a(v_0)\, S_a(\vec{v}_0\to\vec{v}_2)[S^+_a(\vec{v}_2\to\vec{v},\vec{k}) + S^+_b(\vec{v}_2\to\vec{v},\vec{k})]$$

$$\times\{[\tilde{\gamma}_{ab}(v_2) + i(\Delta\tilde{\omega}(v_2) - \vec{k}\cdot\vec{v}_2)]^{-1} + [\tilde{\gamma}_{ab}(v_2) - i(\Delta\tilde{\omega}(v_2) + \vec{k}\cdot\vec{v}_2)]^{-1}\}$$

$$\times[\tilde{\gamma}_{ab}(v) + i(\Delta\tilde{\omega}(v) - \vec{k}\cdot\vec{v})]^{-1} + [(\text{term})\vec{k}\to -\vec{k}], \quad \text{(B2)}$$

where

$$S^+_\alpha(\vec{v}_2\to\vec{v},\vec{k}) = \int_0^\infty d\tau''\, e^{-\gamma_\alpha\tau''}\, G^+_{\alpha\alpha}(\tau'';\vec{v}_2,\vec{v};2\vec{k}) \quad \text{(B3)}$$

and

$$G^+_{\alpha\alpha}(\tau'';\vec{v}_2,\vec{v};2\vec{k}) = P^0_{\alpha\alpha}(\tau'', v_2, \text{DM})\exp[2i\vec{k}\cdot\vec{v}\,\tau'']\delta(\vec{v}-\vec{v}_2)$$

$$+\int_0^{\tau''} d\tau_1 P^0_{\alpha\alpha}(\tau_1, v_2, \text{DM}) W_{\alpha\alpha}(\vec{v}_2\to\vec{v})\; P^0_{\alpha\alpha}(\tau''-\tau_1, v, \text{DM})\exp\{2i\vec{k}\cdot[\vec{v}_2\tau_1 + \vec{v}(\tau''-\tau_1)]\} + \cdots \quad \text{(B4)}$$

are obtained by techniques similar to those used in Sec. V and Appendix A [see Eq. (A3)]. An examination of Eqs. (B1)–(B4) will show that, if $\tilde{\gamma}_{ab}$ and $\Delta\tilde{\omega}$ do not depend on speed, the contributions from these terms will be symmetric functions of cavity detuning about $\Delta\tilde{\omega} = 0$. Thus, asymmetries in the intensity line shape cannot arise from non-Doppler-limit terms any more than they can from the Doppler-limit term. It is also easy to obtain formal expressions for the non-Doppler-limit terms when there is scattering interaction in both radiative states, but we shall not give those expressions here.

Both the above non-Doppler-limit terms become comparable with the Doppler-limit contribution for large decay parameters $\tilde{\gamma}_{ab}$ or detunings $\Delta\tilde{\omega}$. If these quantities are small, the correction to the Doppler-limit terms from Eq. (B1) is $\approx\tilde{\gamma}_{ab}/ku$, while that from (B2) is $\approx(\tilde{\gamma}_{ab}/ku)^2$.

Excitation of Laser Atoms to State b

It is easy to generalize our results to allow for excitation of laser atoms to the lower laser state b if we leave both the a- and b-scattering interactions in the differential equations (I30) (even though one interaction may be negligible). In that case, a careful examination of Eqs. (I30) along with the knowledge of how $\mathcal{A}(t)$ is formed [i.e., $\sim a(\vec{p},t)b^*(\vec{p},t)$] will reveal that

$$\mathcal{A}(t,b) = [\mathcal{A}(t,a)]^* \quad \text{with } a\longleftrightarrow b,\ \Delta\omega\longleftrightarrow -\Delta\omega, \quad \text{(B5)}$$

where $\mathcal{A}(t,\alpha)$ is the contribution to $\mathcal{A}(t)$ for excitation of laser atoms to state α. One should note

that $\bar{\gamma}_{ab}$ is unchanged by this transformation since $(\bar{\gamma}_{ba})^* = \bar{\gamma}_{ab}$ [see Eq. (42a) or (A4a)], which implies that both $\tilde{\gamma}_{ab}$ and $\Delta\tilde{\omega}$ are unchanged by the transformation. If desired, the $\mathcal{A}(t, b)$ terms may be added onto the $\mathcal{A}(t, a)$ terms already calculated in this paper. If the atoms start out with an equilibrium velocity distribution, the only change in the results of this work is to replace Λ_a/γ_a by $\Lambda_a/\gamma_a - \Lambda_b/\gamma_b$ wherever it appears.

Note added in proof. This work does not take into account the effects of radiation trapping on laser profiles {see, e.g., M. D'Yakonov and V. Perel, Zh. Eksperim. i Teor. Fiz. 58, 1090 (1970) [Sov. Phys. JETP 31, 585 (1970)]}. Such effects are somewhat greater than or of the order of non-Doppler-limit terms and should be included in non-Doppler-limit calculations.

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[1]P. R. Berman and W. E. Lamb, Jr., Phys. Rev. A 2, 2435 (1970).

[2]Methods for checking and modifying this hypothesis will be discussed in Sec. VIII. In general, the two radiative states will have different orbital radii and polarizabilities, leading to the result that one state experiences a stronger collision interaction than the other. However, if the two radiative states belong to the same electronic level (i.e., laser transitions between two molecular vibrational states of a given electronic level), the collisional interaction for these states would be equal in first approximation, and our hypothesis fails. Such cases may be treated by alternative means (see Ref. 23 of QMI).

[3]We have explicitly carried out a calculation of a typical term in the perturbation solution allowing for two collisions in the active atom's lifetime. The results of this calculation are in agreement with those achieved using the pseudoclassical collision model.

[4]E. Lindholm, Arkiv Mat. Astron. Fysik 32A, 17 (1945); H. M. Foley, Phys. Rev. 76, 616 (1946); P. W. Anderson, *ibid.* 76, 647 (1949). These papers deal with emission or absorption line profiles, but the ideas employed in the theory could be easily extended to cover laser profiles. The theory of these works neglects velocity changes due to collisions.

[5]M. Baranger, Phys. Rev. 111, 481 (1958).

[6]An alternative approach to use of a quantum-mechanical description for the atomic center-of-mass motion has been recently proposed by E. W. Smith, J. Cooper, W. R. Chappell, and T. Dillon (private communication). It is believed that the general results of their formalism and ours are in agreement, lending additional verification to both the PCM and the calculational model of Appendix A. (The authors are thankful to Dr. Smith for providing reports of their work.)

[7]The idea of the DMC and phase-shifting collisions occurring at different times is intimately connected with our diagrammatic approach to the problem. Had we looked at the change in the density matrix of the atom resulting from a collision, we would find *all* the density matrix elements to be altered by the collision. In that sense, a single collision produces both phase-shifting and DMC effects. Of course, the two methods are compatible, since the sum of all diagrams can be interpreted to imply that both DMC and phase-shifting effects occur in a single collision, even though a single diagram can be interpreted to imply that a collision is either a DMC or a phase-shifting collision.

[8]L. Gallatry, Phys. Rev. 122, 1218 (1961); S. G. Rautian and I. I. Sobelman, Usp. Fiz. Nauk 90, 209 (1966) [Sov. Phys. Usp. 9, 701 (1967)]; J. I. Gersten and H. M. Foley, J. Opt. Soc. Am. 58, 933 (1968); B. L. Gyorffy, M. Borenstein, and W. E. Lamb, Jr., Phys. Rev. 169, 340 (1968).

[9]In Eq. (11), the integration over t' goes from 0 to ∞, while that appearing Eq. (I19) goes from t to $t+\delta t$. The transformation $\int_t^{t+\delta t} \to \int_0^\infty$ was ultimately performed and justified in Sec. VII of QMI.

[10]In reality, the Doppler limit is formally defined for a Maxwellian velocity distribution only [see W. E. Lamb, Jr., Phys. Rev. 134, A1429 (1964)]. However, we shall use the "Doppler width" to mean ku, where u is the most probable speed of the active atoms at their time of excitation.

[11]B. L. Gyorffy, M. Borenstein, and W. E. Lamb, Jr., Phys. Rev. 169, 340 (1968). See Sec. Vb for an example of this type of approach to averaging over binary-collision histories. The actual calculations will differ slightly, since we have allowed for $\vec{v}$ dependence in our P_0 terms.

[12]Some non-Doppler-limit terms may possess $\vec{v}$ dependence (see Appendix B for the method of treating such terms).

[13]The manner in which the classical phase shifts appearing in Eq. (41a) may be calculated is under consideration. Intuitively, we feel that the phase shift χ_a or χ_b should be evaluated along an undeviated straight-line relative emitter-perturber path (rather than along the actual classical path associated with a or b scattering, respectively) since, according to our model, the velocity associated with off-diagonal density matrix elements is unchanged in a collision. As yet, we have not made a detailed study of the problem to see if this conjecture can be proven.

[14]In Sec. VII of QMI, we asserted that, if $W_a(v_0) = W_b(v_0)$ = equilibrium distribution, then the emission and absorption profiles would be identical. The statement was based on our ability to evaluate the integral in Eq. (56) for this case, in which $W'_\alpha(v_0) = W_\alpha(v_0)$. If $W'_\alpha(\vec{v}_0) \neq W_\alpha(\vec{v}_0)$ or if $W_a(v_0) \neq W_b(v_0)$, the emission and absorption will, in general, be different.

[15]The line-shape parameter Γ_α^{DM} may not depend linearly on pressure. Using some rough approximations, it is possible to show that the rate for DMC (i.e., those collisions that produce a $\Delta\vec{v}$ satisfying $|\vec{k}\cdot\Delta\vec{v}| > \tilde{\gamma}_{ab}$) goes as $(\text{pressure})^{1-2/n}$ for an emitter-perturber interaction of the form B/R^n, where R is the emitter-perturber internuclear separation. (The deviation from a linear pressure dependence is caused by the fact that $\tilde{\gamma}_{ab}$ depends on pressure.) Since we are not concerned with a detailed quantitative calculations of line shapes in this paper, we shall take $\Gamma_\alpha^{DM} \propto$ pressure in the present calculations.

[16]P. W. Smith, J. Appl. Phys. 37, 2089 (1966).

[17]H. K. Holt, Phys. Rev. A 2, 233 (1970).

[18]R. H. Cordover and P. A. Bonczyk, Phys. Rev. 188, 696 (1969).

[19]These values are taken from the resonance experiments of B. Decomps and M. Dumont, IEEE J. Quantum Electron. QE-4, 916 (1968). We should note that their value of $\gamma_b/2\pi = 9.75 \pm 0.15$ MHz disagrees with the value $\gamma_b/2\pi = 8.33 \pm 0.13$ MHz obtained by W. R. Bennett, Jr. and P. J. Kindlmann [Phys. Rev. 149, 38 (1966)], who measured decay radiation from the $2p_4$ state of Ne and the value $\gamma_b/2\pi = (8.3 \pm 10)\%$ MHz obtained by J. M. Bridges and W. L. Wiese [Phys. Rev. A 2, 285 (1970)], who made optical absorption measurements. It would be helpful to resolve these differences since $A(P)$ is dependent upon either γ_a/γ_{ab} or γ_b/γ_{ab}.

[20]The fact that the center of the tuning dip shifts to the blue with increasing pressure of He perturbers [T. P. Sosnowski and W. B. Johnson, IEEE J. Quantum Electron. QE-5, 151 (1969)] and to the red for increasing pressure of Ne perturbers [A. D. White, Appl. Phys. Letters 10, 24 (1967)] is consistent with a repulsive He-Ne interaction and an attractive Ne-Ne interaction if state a experiences a stronger collision interaction than does state b.

[21]To obtain $M_b/M_a = 0.16$, we assumed that the ratio $\Gamma_b^{DM}/\Gamma_a^{DM}$ is proportional to ratio of the repulsive-interaction cross sections for ground-state He atoms interacting with Ne atoms in states b and a, respectively. An estimate of these cross sections may be obtained from the theoretical work of G. A. Tsongas and S. P. Koutsoyannis [J. Phys. B 2, 437 (1969)] and leads to the ratio given above.

[22]Figure 8 has been drawn to illustrate the general *qualitative* agreement between theory and experiment. However, for a more detailed analysis, these following points must be noted. (1) Smith's results (see Ref. 16) reveal that the Doppler limit was not well satisfied at all his data points ($\tilde{\gamma}_{ab}/ku = 0.09$ at 1 Torr, 0.16 at 2 Torr, and 0.23 at 3 Torr). (2) Consequently, one might try to include non-Doppler-limit terms in the theory. Using some general arguments, one can show that, with the inclusion of non-Doppler-limit terms of relative order $\tilde{\gamma}_{ab}/ku$, Eq. (74) is replaced by

$$I_0 = \tfrac{1}{2}A(P)\, Z_i(-\Delta\tilde{\omega}/ku,\ \tilde{\gamma}_{ab}/ku) \times [1 - \mathfrak{N}^{-1}]\, [1 + \mathcal{S}(P)\tilde{\gamma}_{ab}/ku]^{-1},$$

with

$$\mathfrak{N}^{-1} = \frac{Z_i(-\Delta\tilde{\omega}_c/ku,\ \tilde{\gamma}_{ab}/ku)}{Z_i(0,\ \tilde{\gamma}_{ab}/ku)},$$

where Z_i is the imaginary part of the plasma dispersion function and $\mathcal{S}(P)$ is a factor of order unity that will depend on the type of collision model. Thus, to get a measure of $A(P)$ one must graph $I_0\{Z_i(-\Delta\tilde{\omega}/ku,\ \tilde{\gamma}_{ab}/ku)[1 - \mathfrak{N}^{-1}]\}^{-1}[1 + \mathcal{S}(P)\tilde{\gamma}_{ab}/ku]$ rather than I_0. (3) Smith's values do, in fact, represent $I_0\{Z_i(-\Delta\tilde{\omega}/ku,\ \tilde{\gamma}_{ab}/ku)[1 - \mathfrak{N}^{-1}]\}^{-1}$. Had he included the $(1 + \mathcal{S}(P)\,\tilde{\gamma}_{ab}/ku$ term, for which some theoretical evaluation of $\mathcal{S}(P)$ becomes necessary, his experimental points would have shown increased curvature, possibly indicating a higher best value for M_a. (4) In Ref. 15, we noted that Γ_α^{DM} will probably not depend quite linearly on pressure. This would tend to reduce the curvature of the theoretical curves, again possibly indicating a higher best value for M_a. The above comments reinforce the premise that Smith's data support a value $\Gamma_\alpha^{DM} \neq 0$.

[23]In Holt's work, the third-order and high-intensity theories were compared for the case $\Gamma_\alpha^{DM} = 0$ only. We assume that the general features of the comparison would not be altered for $\Gamma_\alpha^{DM} \neq 0$.

[24]Holt's data were taken in a region where the Doppler limit was fairly well satisfied ($\tilde{\gamma}_{ab}/ku \lesssim 0.1$). Still, had she plotted $A(P)$ vs P rather than I_0 vs $\tilde{\gamma}_{ab}$ (see Ref. 22 of this work), the graph would have shown increased curvature (although not enough to make a quantitative determination of Γ_α^{DM}).

[25]A. Szöke and A. Javan, Phys. Rev. 145, 137 (1966).

[26]See Secs. V c and VI of Ref. 11.

[27]S. Stenholm and W. E. Lamb, Jr., Phys. Rev. 181, 618 (1969).

[28]B. J. Feldman and M. S. Feld, Phys. Rev. A 1, 1375 (1970).

[29]Our theory has been oriented towards situations where one has neutral atom foreign gas perturbers, but it could also account for electron or ion perturbers if the appropriate active-atom–perturber interaction were used.

[30]A. D. White, Appl. Phys. Letters 10, 24 (1967).

[31]T. P. Sosnowski and W. B. Johnson, IEEE J. Quantum Electron. QE-5, 151 (1969).

[32]For example, in fitting the experimental data in Sec. VII, we found a value $\Gamma_s^{DM} \approx 0.1\,\tilde{\gamma}_{ab}$. This result has some significance in its own right. That is, using rough calculations, one might have guessed that the rate Γ_s^{DM} for producing collisions with $|\vec{k}\cdot\Delta\vec{v}_s| \geq \tilde{\gamma}_{ab}$ would be approximately equal to $\tilde{\gamma}_{ab}$. The fact that $\Gamma_s^{DM} \approx 0.1\,\tilde{\gamma}_{ab}$ indicates that the effective DMC cross section is smaller than that obtained by considering those collisions with $|\vec{k}\cdot\Delta\vec{v}_s| \geq \tilde{\gamma}_{ab}$.

[33]R. H. Dicke, Phys. Rev. 89, 472 (1953).

[34]An attempt along these lines has been made by S. Stenholm [Phys. Rev. Letters 26, 159 (1971)], although he ultimately performed his calculation in the low-intensity limit. We should point out that Stenholm did not correctly incorporate our theory into his work since, for collisional interaction in one state only, he still allowed a collision to both alter the velocity associated with and produce a phase shift in the off-diagonal density matrix elements of the emitter atom, in contrast to our model. In a future paper, we shall present a theory in which the collision phenomena are directly incorporated into the differential equations for the macroscopic density matrix.

[35]To recall how the cutoff originally entered the theory, one must refer to the discussion following Eq. (I65) of QMI, in which the replacement

$$\mathfrak{N}v\int d\Omega_{v'}\, |f_\alpha(\vec{v}\to\vec{v}')|^2\{1 - \exp[i\vec{k}\cdot(\vec{v} - \vec{v}')\tau']\}$$

$$\Rightarrow \Gamma_\alpha^{DM} - \int' d\Omega_{v'}\, W(\vec{v}\to\vec{v}')\exp[i\vec{k}\cdot(\vec{v} - \vec{v}')\tau']$$

was made. The prime on the integral restricted the integration to the "Doppler-modifying" region $|\vec{k}\cdot(\vec{v} - \vec{v}')| > \tilde{\gamma}_{ab}$, but it should be clear from the discussion following Eq. (I65) that any cutoff is permissible provided

$$[\int - \int']d\Omega_{v'}\exp[i\vec{k}\cdot(\vec{v} - \vec{v}')\tau']\}f_\alpha(\vec{v}\to\vec{v}')|^2 \approx [\int - \int']d\Omega_{v'}\, |f_\alpha(\vec{v}\to\vec{v}')|^2$$

for the time region τ' of interest (usually of order $\tilde{\gamma}_{ab}^{-1}$). Note that this is a much weaker definition of the cutoff than that given in the text [$|\vec{k}\cdot(\vec{v} - \vec{v}')| < \tilde{\gamma}_{ab}$ for a non-DMC] and provides a great deal of freedom in choosing the cutoff.

[36]S. G. Rautian, Zh. Eksperim. i Teor. Fiz. 51, 1176 (1966) [Sov. Phys. JETP 24, 788 (1967)].