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Time-dependent projection-operator approach to master equations for coupled systems. II. Systems with correlations*

R. H. Picard
Rome Air Development Center, Deputy for Electronic Technology, Hanscom AFB, Massachusetts 01731

C. R. Willis
Boston University, Boston, Massachusetts 02215

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In a previous paper we showed how time-dependent projection operators may be employed to enable the Nakajima-Zwanzig projection-operator technique for deriving exact master equations to deal efficiently with two or more coupled classical or quantum systems, neither of which is reservoir like. We considered in detail the case where the relevant part of the classical-system probability-density function (PDF) or quantum-system density operator (DO) is a product of the PDF's or DO's for the separate subsystems, and we applied the techniques developed to problems in quantum optics and in the kinetic theory of dilute nonideal gases. In this paper we make the time-dependent projection-operator approach useful for a greater variety of systems by allowing the relevant part of the PDF or DO to include correlations between two of the interacting subsystems. This extension allows us to describe well the dynamics of strongly interacting systems in a low degree of approximation while avoiding the use of infinite resummations. We derive exact generalized master equations in this manner for the same two cases as in our earlier work, namely a classical gas of N molecules interacting via a two-body potential and a quantum-optical system of N two-level atoms interacting with an electromagnetic field. In the former case, the relevant part of the PDF contains two-body correlations, and we obtain two exact coupled master equations for the singlet and doublet PDF's $F_1(t)$ and $F_2(t)$. In the latter case, the relevant part of the DO contains atom-field correlations, and the result is three exact coupled equations for the single-atom DO $\rho_1(t)$, the field DO $R(t)$, and the DO for one atom plus the field $\chi(t)$. From the exact equations we derive approximate equations by making simplifying assumptions. In the case of the gas we carry out a straightforward density expansion of the $\dot{F}_2$ master equation and obtain a set of kinetic equations for dense gases, which have been derived previously by Frieman, Goldman, and Dorfman and which describe well effects due to the finite mean free path. In the quantum-optical case we derive kinetic equations for a single-mode laser by treating the $\dot{\chi}$ equation in the Born-Markoff approximation. The equations describe gain saturation and other effects on the field dynamics which are of infinite order in the atom-field coupling constant. It is shown that these equations reduce to previously derived laser master equations if atom-atom correlations can be neglected. Finally, we mention possible generalizations of the time-dependent projection-operator approach used in this paper and briefly discuss its application to other problems, including the kinetics of liquids, collisional line broadening, superradiance, and amplified spontaneous emission.

I. INTRODUCTION

It has been known for some time[1,2] that, by the method of projection operators (PO's), one may obtain from the Liouville equation of a system an exact closed generalized master equation (ME) for the probability density function describing the "relevant" part of that system. The procedure involves partitioning the probability density $F(t)$, which satisfies the Liouville equation

$$\dot{F}(t) = LF(t)\ , \tag{1.1}$$

into a relevant part $F_r(t)$ and in irrelevant part $F_i(t)$,

$$F(t) = F_r(t) + F_i(t)\ . \tag{1.2}$$

Here L is the Liouvillian superoperator, such that LO is the Poisson bracket of the Hamiltonian H and O, where O is any ordinary operator. For a system of N structureless molecules with coordinates and momenta $(\vec{q}_\alpha, \vec{p}_\alpha)$, $\alpha = 1, \ldots, N$, L may be written

$$L \equiv \{H, \ldots\} = \sum_{\alpha}^{N} \left(\frac{\partial H}{\partial \vec{q}_\alpha} \cdot \frac{\partial}{\partial \vec{p}_\alpha} - \frac{\partial H}{\partial \vec{p}_\alpha} \cdot \frac{\partial}{\partial \vec{q}_\alpha} \right)\ . \tag{1.3}$$

One then operates on Eq. (1.1) with PO's P and $Q \equiv 1 - P$, satisfying

$$P^2 = P, \quad Q^2 = Q, \quad PQ = QP = 0\ , \tag{1.4}$$

such that

$$F_r(t) = PF(t), \quad F_i(t) = QF(t)\ , \tag{1.5}$$

and eliminates $F_i(t)$ to obtain the exact ME

$$\dot{F}_r(t) - PLF_r(t) = PL\mathcal{G}(t - t_0)F_i(t_0) + \int_{t_0}^{t} dt'\, PL\mathcal{G}(t - t')(1 - P)LF_r(t')\ , \tag{1.6}$$

where

$$\mathcal{G}(\tau) \equiv e^{(1-P)L\tau} \quad . \tag{1.7}$$

The irrelevant part occurs only through its initial value $F_i(t_0)$, which vanishes for some important physically realizable initial conditions. This procedure works equally as well for quantum systems, in which case F, F_r, and F_i are density operators and the Poisson bracket is replaced by $(-i/\hbar)$ times the commutator, so that

$$L = -(i/\hbar)[H, \ldots] \quad . \tag{1.8}$$

Most often, $F_r(t)$ is chosen to be (a) the diagonal part of the system density operator[2] or (b) for a system interacting with a reservoir, the product of an equilibrium distribution function for the reservoir and the distribution function for the system alone.[3,4] In a previous paper,[5,6] hereafter referred to as I, we have shown how one can enlarge the class of possible F_r's by allowing our PO's to be time dependent. Time-dependent projection operators (TDPO's) have been introduced by other authors[7,8] in connection with another class of problems. These operators $P(t)$ and $Q(t) \equiv 1 - P(t)$ satisfy,[9] instead of Eqs. (1.4),

$$P(t)P(t') = P(t) \ , \tag{1.9a}$$

$$Q(t)Q(t') = Q(t') \ , \tag{1.9b}$$

$$P(t)Q(t') = 0 \tag{1.9c}$$

for arbitrary t and t'. The partition of F, Eqs. (1.5), and the ME, Eqs. (1.6) and (1.7), are replaced by

$$F_r(t) = P(t)F(t) \ , \tag{1.10a}$$

$$F_i(t) = Q(t)F(t) \ , \tag{1.10b}$$

and

$$\dot{F}_r(t) - P(t)LF_r(t) = P(t)L\mathcal{G}(t, t_0)F_i(t_0) + \int_{t_0}^{t} dt' P(t)L\mathcal{G}(t, t') \times [1 - P(t')]\, LF_r(t') \ , \tag{1.11}$$

where

$$\mathcal{G}(t, t') \equiv T\exp\left(\int_{t'}^{t} dt''[1 - P(t'')]L\right) \tag{1.12}$$

and T is the Dyson time-ordering operator. In order to derive Eq. (1.11), one needs a condition on $P(t)$ in addition to Eqs. (1.9) and (1.10), namely, $P(t)$ must commute with the time derivative when operating on $F(t)$,

$$\left[P(t), \frac{d}{dt}\right] F(t) = 0 \ , \tag{1.13a}$$

from which it follows that

$$\dot{F}_r(t) = P(t)\dot{F}(t) \ . \tag{1.13b}$$

The purpose of introducing the apparent complication of TDPO's is not obvious, since both Eqs. (1.6) and (1.11) are *exact* equations. The advantage of considering TDPO's becomes clear, however, as soon as one attempts to *approximate* the ME in some way, a necessary procedure in any nontrivial problem. It will then become apparent that the use of TDPO's enables one to obtain very powerful descriptions of coupled systems in a low level of approximation. Typically, similar descriptions using ordinary time-dependent PO's require one to perform infinite resummations.

In I we showed that TDPO's were particularly well suited to describing a dilute gas of interacting molecules or self-consistently interacting systems, such as the system of two-level atoms interacting with a resonant em radiation field commonly considered in quantum optics. In the former case we chose for the relevant part of the N-particle distribution function F_N a product of one-particle distribution functions F_1,

$$F_{N,r}(1, \ldots, N, t) = \prod_{\alpha}^{N} F_1(\alpha, t) \ , \tag{1.14}$$

where

$$F_1(\alpha, t) \equiv \left(\prod_{\substack{\beta \\ (\beta \neq \alpha)}}^{N}{}' \int \frac{d\beta}{V} \right) F_N(1, \ldots, N, t) \ . \tag{1.15}$$

The properties of F_N, F_1, and $F_{N,r}$ and the notation are described more completely in[10] Eqs. (I3.4)–(I3.9). The integral $\int d\beta$ is an integral over the phase space of particle β. In the latter case, we chose for the relevant part of the system density operator F a product of the density operator ρ of the atoms and the density operator R of the field[11]

$$F_r(t) = R(t)\rho(t) \ . \tag{1.16}$$

That is, in each case we chose PO's which projected a general state of the system onto *uncorrelated* states of the interacting subsystems.

In this paper we would like to extend the method by introducing operators which project general system states onto *correlated* states of the interacting subsystems.[6] In the case of a system of interacting molecules, we will allow $F_{N,r}(t)$ to contain correlations between two molecules at a time, and we will obtain exact coupled ME's for the singlet and doublet distribution functions $F_1(1, t)$ and $F_2(1, 2, t)$. The function F_2 is defined by Eq. (I3.6), that is,

$$F_2(1, 2, t) \equiv \int \frac{d3}{V} \cdots \int \frac{dN}{V} F_N(t) \ . \tag{1.17}$$

We will find that inclusion of the doublet distribution function in the theory allows us to describe the kinetics of *dense* gases with ease. A density expansion of the ME for $\dot{F}_2$ will result in a theory which is free of divergence problems relative to the transport coefficients,[12] since it includes mean-free-path effects through the inclusion of irreducible three-particle correlations. To the lowest order in the density N/V, we will obtain in the binary-collision approximation[13] the renormalized kinetic equations for dense gases cited by Dorfman,[14] whose derivation was carried out by Frieman and Goldman[15] using multiple-time-scale perturbation theory. We will see that the TDPO approach is straightforward and can easily be generalized to higher order in the density. By contrast, in I when we expanded the master equation for $\dot{F}_1$ in powers of N/V, the first-order term gave rise to the Boltzmann equation.

In the quantum-optical case of atoms interacting with a resonant field, the generalization proposed in this paper will also allow us to obtain a significantly more powerful description. We will allow $F_{N,r}$ to include correlations between the field and a single atom at a time, and we will derive exact coupled ME's for the density operator $R(t)$ of the field, the density operator $\rho_1(\alpha, t)$ of the αth atom, and the joint density operator $\chi(\alpha, t)$ of the field and the αth atom.[6] The density operator χ is defined as a trace of the system operator F_N by

$$\chi(\alpha, t) \equiv \left(\prod_{\substack{\beta \\ (\beta \neq \alpha)}}^{N}{}' \mathrm{Tr}_\beta \right) F_N(1, \ldots, N, t) , \tag{1.18}$$

while R is defined by Eq. (I1.3), or as a trace of χ over the degrees of freedom of the αth atom

$$R(t) \equiv \mathrm{Tr}_\alpha \chi(\alpha, t) , \tag{1.19a}$$

and ρ_1 is a trace of χ over the field

$$\rho_1(\alpha, t) \equiv \mathrm{Tr}_f \chi(\alpha, t) . \tag{1.19b}$$

In the case of laser theory, where the atoms and field are coupled to reservoirs simulating relaxation mechanisms, we will show the connection of our equations with previous theories.[16-18] The standard theories are first order in the atom-field coupling constant μ and do not contain *any* correlations between two atoms and the field. We will show that *reducible* two-atom-field correlations lead to small, but not negligible, corrections of the same magnitude as ordinary spontaneous emission. We will also evaluate the relative magnitude of the correction terms $\sim\mu^2$, containing the effect of *irreducible* two-atom–plus–field correlations, and show that the corrections due to irreducible atom-atom correlations are small. The advantage of the TDPO approach is also apparent here if one compares the derivation of the terms $\sim\mu^2$ to a recent derivation by Mandel[19] using a time-independent PO; the latter derivation requires an elaborate resummation of an infinite set of diagrams of all orders in μ to obtain the atom-atom correlation terms.

In Sec. II we determine the PO for the case of a gas of structureless molecules interacting via two-body forces corresponding to an $F_{N,r}$ which includes two-body correlations. We then derive the exact coupled ME's for $\dot{F}_1$ and $\dot{F}_2$. We continue in Sec. III by deriving the PO for the quantum-optical case which projects out a state containing correlations between a single atom and the field. In that section we also derive exact coupled ME's for $\dot{\chi}$, $\dot{R}$, and $\dot{\rho}_1$. In Sec. IV the density expansion of the ME for the doublet distribution of the gaseous system is carried out to lowest order in the density, and the result is cast into the form of Dorfman's kinetic equation for a dense gas. As a preliminary exercise to illustrate the method, we rederive the Boltzmann equation, derived in I, by expanding the singlet distribution master equation, Eq. (I3.19) or (I3.22), in powers of N/V and focusing our attention on the first-order terms only. We continue in Sec. V by applying the exact quantum-optical ME's of Sec. III to the case of a laser. We introduce the first Born approximation (FBA) and the Markoff approximation into the $\dot{\chi}$ equation, identify the usual laser theories obtained from the terms $\sim\mu$, and show that the correction terms $\sim\mu^2$ depending on irreducible atom-atom correlations are small for typical values of relaxation times. Sections II–V will be written so that Secs. II and IV on kinetic theory can be read independently of Secs. III and V on quantum optics, insofar as possible. Finally, in Sec. VI we discuss the benefits of using the TDPO approach, especially with regard to projecting onto correlated states, and the connection with other approaches. We also discuss applications of the ME's obtained to other problems and a variety of extensions of the TDPO techniques described in the paper.

II. EXACT MASTER EQUATIONS FOR GAS KINETICS

Assume a gaseous system composed of N molecules interacting via a two-body potential $H'(|\vec{q}_\alpha - \vec{q}_\beta|)$. In I (Sec. III), we derived an exact ME for the time development of the singlet distribution function $F_1(1, t)$, assuming that the relevant part $F_{N,r}$ of the system distribution function was a product of the singlet distribution functions of the individual atoms [Eq. (1.14)]. In this section we would like to generalize this technique to obtain exact coupled ME's for the singlet and doublet distribution functions $F_1(1, t)$ and $F_2(1, 2, t)$ by choosing an $F_{N,r}$ which depends on both F_1 and F_2, that is, an F_r which contains irreducible two-body correlations between molecules. We will use the notation of I, Sec. III. The system Hamiltonian and Liouvillian are defined in Eqs. (I3.1)–(I3.3). First, we will write down $F_{N,r}$ in terms of F_1 and F_2. Then we will derive the TDPO $P_N(t)$ which projects F_N onto $F_{N,r}$ and $\dot{F}_N$ onto $\dot{F}_{N,r}$ [see Eqs. (1.10a) and (1.13b)]. Finally, we will substitute $F_{N,r}(t)$ and $P_N(t)$ into the ME for $\dot{F}_{N,r}(t)$, Eq. (1.11), and by integrating over degrees of freedom appropriately we will derive the exact coupled equations for $\dot{F}_1$ and $\dot{F}_2$.

In choosing $F_{N,r}$ it will be completely adequate, for our purposes, to assume that $F_{N,r}$ is a linear function of F_2. More precisely, we will take $F_{N,r}$ to be the sum of an uncorrelated term which is a product of F_1's and a term which is linear in the correlation distribution ΔF_2, whose definition is

$$\Delta F_2(1,2,t) \equiv F_2(1,2,t) - F_1(1,t)F_1(2,t)\ . \tag{2.1}$$

The latter term must be symmetrized with respect to all molecule labels. Hence, we write

$$F_{N,r}(1,\ldots,N,t) = \prod_{\alpha}^{N} F_1(\alpha,t) + \frac{1}{2}\sum_{\alpha}^{N}\sum_{\beta}^{N}{}' \Delta F_2(\alpha,\beta,t) \prod_{\gamma}^{N}{}'' F_1(\gamma,t)\ . \tag{2.2}$$

We use one or more primes on summation and product signs to indicate that the index does not assume the values of all previously occurring indices. Thus in Eq. (2.2) the prime on the β sum indicates $\beta \neq \alpha$, and the double prime on the γ product indicates $\gamma \neq \alpha, \beta$. Unless there is ambiguity the index values omitted from the sum (product) will not be specified. Equation (2.2) includes the first two terms in a Taylor series expansion of $F_{N,r}$, considered to be a function of F_1 and ΔF_2, about $\Delta F_2 = 0$. More general forms of $F_{N,r}$ depending nonlinearly on ΔF_2 will be considered briefly in Sec. VI. Usually it will be more convenient to consider $F_{N,r}$ to be a function of F_1 and F_2 than of F_1 and ΔF_2. Using Eq. (2.1) to eliminate ΔF_2 from Eq. (2.2), we find[20,6]

$$F_{N,r}(t) = \frac{1}{2}\sum_{\alpha,\beta}{}' F_2(\alpha,\beta,t)\prod_{\gamma}{}'' F_1(\gamma,t) - \left[\binom{N}{2} - 1\right]\prod_{\alpha} F_1(\alpha,t)\ , \tag{2.2$'$}$$

where $\binom{n}{m} \equiv n!/[m!(n-m)!]$ is the binomial coefficient, so that $\binom{N}{2}$ is the number of molecular pairs. One may easily verify that $F_{N,r}$ is normalized, that its integral over the phase space of $N-1$ molecules yields F_1, and that the integral over $N-2$ molecular phase-space coordinates yields F_2; that is, Eqs. (I3.5), (1.15), and (1.17) are satisfied by Eq. (2.2′). Moreover, we have

$$\int \frac{dN}{V} F_{N,r}(1,\ldots,N,t) = F_{N-1,r}(1,\ldots,N-1,t)\ . \tag{2.3}$$

There are no rules to find the PO which yields the relevant part of F_N given by Eq. (2.3), anymore than there are rules to find any PO. One is forced to proceed inductively at best or by trial and error at worst. We will attempt to find an optimum middle ground between these extremes. The PO must satisfy three conditions, Eqs. (1.9a), (1.10a), and (1.13b). The solution to these equations is not unique, since if $P_N(t)$ is a solution it follows that $P_N^2(t)$ is also.

In order to find the general projection operator, we begin by considering first the special case $N=3$, which we will use as a guide for the general case. In the case $N=3$, $F_{N,r}$ becomes

$$F_{3,r}(1,2,3,t) = F_2(1,2,t)F_1(3,t) + \mathrm{cycl}(1,2,3) - 2F_{3,nc}(1,2,3,t)\ , \tag{2.4}$$

where

$$F_{3,nc}(1,2,3,t) = F_1(1,t)F_1(2,t)F_1(3,t) \tag{2.5}$$

is an uncorrelated product state and cycl$(1,\ldots,s)$ indicates all cyclic permutations of the molecular labels $(1,\ldots,s)$. Consider the operator

$$P_3(t) \equiv F_1(3,t)\int\frac{d3}{V} + F_2(1,2,t)\int\frac{d1}{V}\int\frac{d2}{V} - F_2(1,2,t)F_1(3,t)\int d\Gamma_3 + \mathrm{cycl}(1,2,3) - 2P_{3,nc}(t)\ . \tag{2.6}$$

Here $\int d\Gamma_s \equiv V^{-s}\int d1\cdots\int ds$ represents a complete integral over the space of all the molecules, and $P_{3,nc}(t)$ is the TDPO satisfying

$$P_{3,nc}(t)F_3(t) = F_{3,nc}(t)\ , \tag{2.7a}$$

$$P_{3,nc}(t)\dot{F}_3(t) = \dot{F}_{3,nc}(t)\ . \tag{2.7b}$$

According to Eq. (I3.11) with $N=3$, $P_{3,nc}(t)$ has the form

$$P_{3,nc}(t) = F_1(1,t)F_1(2,t)\int\frac{d1}{V}\int\frac{d2}{V} + \mathrm{cycl}(1,2,3) - 2F_1(1,t) \times F_1(2,t)F_1(3,t)\int d\Gamma_3\ . \tag{2.8}$$

We would like to show that Eq. (2.6) satisfies the three requirements for our PO with F_r given by Eq. (2.4).

In the first place, we have immediately from Eqs. (2.6) and (2.7a),

$$\begin{aligned} P_3(t)F_3(t) &= (1+1-1)F_2(1,2,t)\,F_1(3,t) + \mathrm{cycl}(1,2,3) - 2F_{3,nc}(t) \\ &= F_{3,r}(t)\ . \end{aligned}$$

Secondly, from the condition $\int d\Gamma_3 \dot{F}_3(t) = 0$ and Eq. (2.7b) it follows that

$$\begin{aligned} P_3(t)\dot{F}_3(t) &= F_1(3,t)\dot{F}_2(1,2,t) + F_2(1,2,t)\dot{F}_3(t) \\ &\quad + \mathrm{cycl}(1,2,3) - \dot{F}_{3,nc}(t) \\ &= \dot{F}_{3,r}(t) \ . \end{aligned}$$

The last step follows by comparison with the derivatives of Eq. (2.4). Finally, we must show that Eq. (2.6) is a TDPO, or that Eq. (1.13b) is satisfied. Since Eq. (1.13b) must be true for every t, it follows generally that if $P(t)$ is a sum of l terms

$$P(t) = \sum_{i=1}^{l} p^{(i)}(t) \ , \tag{2.9}$$

we must have

$$p^{(i)}(t)P(t') = p^{(i)}(t) \ . \tag{2.10}$$

In the present case, we have the relations

$$\begin{aligned} p^{(1)}(3,t)P_3(t') &\equiv F_1(3,t)\int \frac{d3}{V} P_3(t') \\ &= F_1(3,t)\Big[\int \frac{d3}{V} + (1-1)F_2(1,2,t')\int d\Gamma_3 \\ &\qquad + \Big((1+1-2)F_1(1,t')\int \frac{d1}{V} \int \frac{d2}{V} + (1 \leftrightarrow 2)\Big) - (1+1+2-4)F_1(1,t')F_1(2,t')\int d\Gamma_3 \Big] \\ &= F_1(3,t)\int \frac{d3}{V} \equiv p^{(1)}(3,t) \ , \end{aligned} \tag{2.11a}$$

$$\begin{aligned} p^{(2)}(1,2,t)P_3(t') &\equiv F_2(1,2,t)\int \frac{d1}{V}\int \frac{d2}{V} P_3(t') \\ &= F_2(1,2,t)\Big((1+1+1-2)\int \frac{d1}{V} \int \frac{d2}{V} + (1-1+1-1+1-1-2-2+4)F_1(3,t')\, d\Gamma_3 \Big) \\ &= F_2(1,2,t)\int \frac{d1}{V} \int \frac{d2}{V} \\ &\equiv p^{(2)}(1,2,t) \ , \end{aligned} \tag{2.11b}$$

$$\begin{aligned} p^{(3)}(1,2,3,t)P_3(t') &\equiv -F_2(1,2,t)F_1(3,t)\int d\Gamma_3 P_3(t') \\ &= -F_2(1,2,t)F_1(3,t)[3(1+1-1) - 2(3-2)] \int d\Gamma_3 \\ &= -F_2(1,2,t)F_1(3,t)\int d\Gamma_3 \equiv p^{(3)}(1,2,3,t) \ , \end{aligned} \tag{2.11c}$$

$$p^{(4)}(1,2,t)P_3(t') \equiv -2F_1(1,t)F_1(2,t)\int \frac{d1}{V} \int \frac{d2}{V} P_3(t') = p^{(4)}(1,2,t) \ , \tag{2.11d}$$

and

$$p^{(5)}(1,2,3,t)P_3(t') \equiv 4F_{3,nc}(t) \int d\Gamma_3 P_3(t') = p^{(5)}(1,2,3,t) \ . \tag{2.11e}$$

The validity of Eqs. (2.11d) and (2.11e) follows from Eqs. (2.11b) and (2.11c), respectively, since the former are the same as the latter with different functions in front of the integrals. By adding Eqs. (2.11) and the necessary cyclic permutations it follows that $P_3(t)P_3(t') = P_3(t)$, completing the proof that $P_3(t)$ is a TDPO.

Of course, we are generally interested in systems of N particles and would like the appropriate generalization of Eq. (2.6). By examining Eqs. (2.2′), (2.4), and (2.6), we can guess the following properties of the general PO $P_N(t)$. (i) The cyclic permutation terms in Eq. (2.6) will be replaced by sums over all pairs of particles. (ii) The generalizations of the first, second, and third terms of Eq. (2.6) will have $N-2$, $N-1$, and N integrals, respectively. (iii) No doublet distribution F_2 may occur in the first term of the generalization and only one in the second and third terms. (iv) The coefficient of the third term will be chosen so that the first three terms yield the proper coefficient for the lead term of Eq. (2.2′). (v) The PO $P_{3,nc}(t)$ which projects out $F_{3,nc}(t)$ is replaced by $P_{N,nc}(t)$, given by Eq. (I3.11), which projects out

$$F_{N,nc}(t) \equiv \prod_{\alpha}^{N} F_1(\alpha, t) \ . \tag{2.12}$$

With these guidelines one arrives at the TDPO,[6]

$$P_N(t) = \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta}^{N} \sideset{}{''}\prod_{\gamma}^{N} F_1(\gamma, t) \int \frac{d\gamma}{V} + \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \int \frac{d\alpha}{V} \int \frac{d\beta}{V} \sideset{}{''}\sum_{\gamma} \sideset{}{'''}\prod_{\delta} F_1(\delta, t) \int \frac{d\delta}{V}$$
$$- \tfrac{1}{2}(N-2) \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \sideset{}{''}\prod_{\gamma} F_1(\gamma, t) \int d\Gamma_N - [\tbinom{N}{2} - 1] P_{N,nc}(t) \ . \tag{2.13}$$

The proof that this is the desired generalization of Eq. (2.6) is lengthy and tedious but straightforward. The spirit of the proof is indicated in Appendix A. The proof that $P_N(t)$ is a PO is facilitated if one uses the decomposition of Eq. (1.10a) stated in Eqs. (2.9) and (2.10). The principle difficulty in the proof then involves enumerating properly the cases corresponding to the various possible relations between the summation indices in $P_N(t)$ and those in $P_N(t')$ and assigning the proper weights to each.

The next step is to obtain the master equation. The ME for $\dot{F}_{N,r}$ follows from substitution of Eqs. (2.2′) and (2.13) into Eq. (1.11). We choose $t_0 = -\infty$ and assume the condition $F_{N,i}(-\infty) \equiv F_N(-\infty) - F_{N,r}(-\infty) = 0$, which is equivalent to assuming no irreducible three-body and higher-order correlations asymptotically in the infinite past. In the derivation we assume that the initial distribution is symmetric under permutation of particles. This coupled with the symmetry of the Hamiltonian, Eq. (I3.1), will allow us to permute particle labels at will.

In order to derive the explicit form of the ME for $\dot{F}_{N,r}$ we will need to write out some of the expressions entering Eq. (1.11). From Eqs. (2.13) and (I3.2) we find that

$$P_N(t) L^0_{(N)} = \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} (L^0_\alpha + L^0_\beta) \sideset{}{''}\prod_{\gamma} F_1(\gamma, t) \int \frac{d\gamma}{V} + \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \int \frac{d\alpha}{V} \int \frac{d\beta}{V} \sideset{}{''}\sum_{\gamma} L^0_\gamma \sideset{}{'''}\prod_{\delta} F_1(\delta, t) \int \frac{d\delta}{V}$$
$$- [\tbinom{N}{2} - 1] \sum_{\alpha} L^0_\alpha \sideset{}{'}\prod_{\beta} F_1(\beta, t) \int \frac{d\beta}{V} \tag{2.14}$$

and

$$P_N(t) L'_{(N)} = \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} L'_{\alpha\beta} \sideset{}{''}\prod_{\gamma} F_1(\gamma, t) \int \frac{d\gamma}{V} + \sideset{}{'}\sum_{\alpha,\beta} \sideset{}{''}\sum_{\gamma} F_1(\gamma, t) \int \frac{d\gamma}{V} L'_{\alpha\gamma} \sideset{}{'''}\prod_{\delta} F_1(\delta, t) \int \frac{d\delta}{V}$$
$$+ \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \int \frac{d\beta}{V} \sideset{}{''}\sum_{\gamma} \int \frac{d\alpha}{V} L'_{\alpha\gamma} \sideset{}{'''}\prod_{\delta} F_1(\delta, t) \int \frac{d\delta}{V}$$
$$+ \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \int \frac{d\alpha}{V} \int \frac{d\beta}{V} \sideset{}{''}\sum_{\gamma} \sideset{}{'''}\sum_{\delta} F_1(\delta, t) \int \frac{d\delta}{V} L'_{\gamma\delta} \sideset{}{''''}\prod_{\epsilon} F_1(\epsilon, t) \int \frac{d\epsilon}{V}$$
$$- [\tbinom{N}{2} - 1] \sideset{}{'}\sum_{\alpha,\beta} F_1(\beta, t) \int \frac{d\beta}{V} L'_{\alpha\beta} \sideset{}{''}\prod_{\gamma} F_1(\gamma, t) \int \frac{d\gamma}{V} \ , \tag{2.15}$$

where we have used

$$\int \frac{d\alpha}{V} L^0_\alpha \equiv 0, \quad \int \frac{d\alpha}{V} \int \frac{d\beta}{V} L'_{\alpha\beta} \equiv 0 \ . \tag{2.16}$$

Letting Eqs. (2.14) and (2.15) act on Eq. (2.2′) gives rise to two of the terms on the left-hand side of Eq. (1.11),

$$P_N(t) L^0_{(N)} F_{N,r}(t) = \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} (L^0_\alpha + L^0_\beta) F_2(\alpha, \beta, t) \sideset{}{''}\prod_{\gamma} F_1(\gamma, t) + \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \sideset{}{''}\sum_{\gamma} L^0_\gamma F_1(\gamma, t) \sideset{}{'''}\prod_{\delta} F_1(\delta, t)$$
$$- [\tbinom{N}{2} - 1] \sum_{\alpha} L^0_\alpha F_1(\alpha, t) \sideset{}{'}\prod_{\beta} F_1(\beta, t) \ , \tag{2.17}$$

$$P_N(t) L'_{(N)} F_{N,r}(t) = \frac{1}{2} \sideset{}{'}\sum_{\alpha,\beta} L'_{\alpha\beta} F_2(\alpha, \beta, t) \sideset{}{''}\prod_{\gamma} F_1(\gamma, t) + (N-2) \sideset{}{'}\sum_{\alpha,\beta} \int \frac{d\sigma}{V} L'_{\alpha\sigma} F_{3,r}(\alpha, \beta, \sigma, t) \sideset{}{''}\prod_{\gamma} F_1(\gamma, t)$$
$$+ \tfrac{1}{2}(N-1) \sideset{}{'}\sum_{\alpha,\beta} F_2(\alpha, \beta, t) \sideset{}{''}\sum_{\gamma} \int \frac{d\sigma}{V} L'_{\gamma\sigma} F_2(\gamma, \sigma, t) \sideset{}{'''}\prod_{\delta} F_1(\delta, t)$$
$$- [\tbinom{N}{2} - 1] (N-1) \sum_{\alpha} \int \frac{d\sigma}{V} L'_{\alpha\sigma} F_2(\alpha, \sigma, t) \sideset{}{'}\prod_{\beta} F_1(\beta, t) \ , \tag{2.18}$$

through the use of the properties of $F_{N,r}$, including Eq. (2.3); here σ denotes a dummy particle label.

Turning to the right member of the ME, Eq. (1.11), we first make use of the differential equation of the time-development operator $\mathcal{G}^{(N)}$ in the irrelevant subspace[5]

$$\frac{\partial \mathcal{G}^{(N)}(t,t')}{\partial t'} = -\mathcal{G}^{(N)}(t,t')[1-P_N(t')]L_{(N)} \ , \tag{2.19a}$$

with

$$\mathcal{G}^{(N)}(t',t') = 1 \ , \tag{2.19b}$$

which is easily derived by differentiating Eq. (1.12), to introduce $\partial\mathcal{G}^{(N)}/\partial t'$ into the ME and eliminate the second Liouvillian operator. It is also easy to show that the unperturbed part $L^0_{(N)}$ of the first Liouvillian does not contribute. This follows immediately from the very useful property[21] possessed by the $P_N(t)$ of Eq. (2.13),

$$\prod_{\alpha}{}'' \int \frac{d\alpha}{V}\,[1-P_N(t)] \equiv 0 \ , \tag{2.20}$$

whose proof is sketched in Appendix A. Equation (2.14) shows that every term of $P_N(t)L^0_{(N)}$ contains at least $N-2$ integrals on the extreme right-hand side, and that the integrals always operate directly on a factor $[1-P(\bar{t})]$ obtained from $\partial\mathcal{G}^{(N)}(t,t')/dt'$, so that Eq. (2.20) applies. Moreover, we can use Eq. (2.20) to simplify the interaction part of the factor $P(t)L_{(N)}$ in the right-hand member of the ME. Only the second term of Eq. (2.15) contributes, since all the other terms are terminated by $N-2$ integrals and vanish, by Eq. (2.20), that is

$$P_N(t)L'_{(N)}\,\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'} = \sum_{\alpha,\beta}{}'\sum_{\gamma}{}'' F_1(\gamma,t)\int\frac{d\gamma}{V}\,L'_{\alpha\gamma}\prod_{\delta}{}''' F_1(\delta,t)\int\frac{d\delta}{V}\,\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'} \ . \tag{2.21}$$

At this stage, the ME is given by

$$\dot{F}_{N,r}(t) - P_N(t)(L^0_{(N)} + L'_{(N)})F_{N,r}(t) = -\int_{-\infty}^{t} dt'\,P_N(t)L'_{(N)} \times \frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'}F_{N,r}(t') \ . \tag{2.22}$$

Moreover, the left-hand side is given explicitly by Eqs. (2.17) and (2.18), and Eq. (2.21) is available to simplify the right-hand side. It is most natural at this point to integrate over the phase space of all particles but 1 and 2 and obtain the ME for the doublet distribution function $F_2(1,2,t)$. From Eqs. (2.17), (2.18), and (2.21), it follows that

$$\prod_{\substack{\alpha \\ (\alpha\neq 1,2)}}{}'' \int\frac{d\alpha}{V}P_N(t)L^0_{(N)}F_{N,r}(t) = (L^0_1 + L^0_2)F_2(1,2,t) \ , \tag{2.23a}$$

$$\prod_{\substack{\alpha \\ (\alpha\neq 1,2)}}{}'' \int\frac{d\alpha}{V}P_N(t)L'_{(N)}F_{N,r}(t) = L'_{12}F_2(1,2,t) + (N-2)\int\frac{d3}{V}\,(L'_{13}+L'_{23}) \times F_{3,r}(1,2,3,t) \ , \tag{2.23b}$$

and

$$\prod_{\substack{\alpha \\ (\alpha\neq 1,2)}}{}'' \int\frac{d\alpha}{V}P_N(t)L'_{(N)}\,\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'} = (N-2)\int\frac{d3}{V}(L'_{13}+L'_{23}) \times \prod_{\substack{\alpha \\ (\alpha\neq 1,2,3)}}{}''' \int\frac{d\alpha}{V}\,\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'} \ . \tag{2.23c}$$

In deriving Eq. (2.23b), note that the only surviving contributions are from those parts of the first and second terms of Eq. (2.18) for which $(\alpha,\beta) = (1,2)$; the other terms all cancel one another ultimately. The ME for the doublet distribution follows from Eqs. (2.23),

$$\dot{F}_2(1,2,t) - L_{(2)}F_2(1,2,t) - (N-2)\int\frac{d3}{V}\,(L'_{13}+L'_{23})F_{3,r}(1,2,3,t) = -(N-2)\int_{-\infty}^{t} dt'\int\frac{d3}{V}\,(L'_{13}+L'_{23})\prod_{\alpha}{}'''\int\frac{d\alpha}{V}\;\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'}F_{N,r}(t') \ , \tag{2.24}$$

where

$$L_{(2)} \equiv L_1^0 + L_2^0 + L'_{12} \equiv L_{(2)}(1,2) \;. \tag{2.25}$$

The last term on the left-hand side of the ME includes the effects of reducible three-body correlations while the irreducible three-body and higher correlations are on the right-hand side. It will be more convenient to combine all three-body terms on the right-hand side through integration by parts with respect to t', in which case the integrated part evaluated at $t'=t$ cancels the last term on the left, and one is left with

$$\dot{F}_2(1,2,t) - L_{(2)}F_2(1,2,t) = (N-2)\int \frac{d3}{V}\,(L'_{13}+L'_{23}) \prod_{\alpha}{}''' \int \frac{d\alpha}{V}\left(\mathcal{G}^{(N)}(t,-\infty)F_{N,r}(-\infty) + \int_0^\infty d\tau\, \mathcal{G}^{(N)}(t,t-\tau)\frac{\partial F_{N,r}(t-\tau)}{\partial t}\right) . \tag{2.26}$$

An obvious change of variables has been made in the time integral.

Although Eq. (2.26) is the required exact ME for $\dot{F}_2$, the ME is not closed since it depends on the singlet distribution F_1. Hence, a ME for $\dot{F}_1$ is required. It is obtained by integrating Eq. (2.25) over the phase space of particle 2. Noting that the right-hand side does not contribute because of Eqs. (2.20) and (2.16), the result is

$$\dot{F}_1(1,t) - L_1^0 F_1(1,t) = (N-1)\int \frac{d2}{V}\, L'_{12}F_2(1,2,t) \;. \tag{2.27}$$

This equation is just the first member of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy,[22] relating $\dot{F}_1$ to F_2. The result of our analysis is contained in the set of two exact coupled ME's for $\dot{F}_1$ and $\dot{F}_2$, Eqs. (2.26) and (2.27). In Sec. IV we will consider density expansions of these exact equations in order to obtain kinetic equations valid for moderately dense gases. Meanwhile, in Sec. III we turn our attention to the quantum-optical problem.

III. EXACT MASTER EQUATIONS FOR QUANTUM OPTICS

In this section we treat the usual quantum-optical case involving N two-level atoms coupled to a near-resonant optical radiation field through the electric-dipole interaction. The system Hamiltonian H is a sum of matter, field, and interaction terms H_m, H_f, and H', according to Eq. (I2.1). The corresponding partition of the Liouvillian is stated in Eq. (I2.2). The explicit forms for H_m, H_f, and H' are listed in Eqs. (I4.3)–(I4.7); however, in the present section we will only need the fact that H_m and H', as well as the corresponding Liouvillians, are sums of operators involving the dynamical variables of a single atom[23]:

$$H_m = \sum_{\alpha}^{N} H_\alpha^0 \,, \quad H' = \sum_{\alpha}^{N} H'_\alpha \,,$$
$$L_m = \sum_{\alpha}^{N} L_\alpha^0 \,, \quad L' = \sum_{\alpha}^{N} L'_\alpha \,. \tag{3.1}$$

The system density operator is $F_N(1,\ldots,N,t)$; it is related to the density operator for the αth atom $\rho_1(\alpha,t)$, the density operator for the field $R(t)$, and the joint density operator of the field and the αth atom[23] $\chi(\alpha,t)$ by Eqs. (1.18) and (1.19). We will not assume the Hamiltonian and initial value of the density operator to be symmetric under particle interchange for the following reason: The positions $\vec{X}_\alpha$ of the atoms enter H', and usually it is appropriate to consider the $\vec{X}_\alpha$ as parameters rather than dynamical variables, since there is negligible reaction of the internal state of an atom on its translation. In addition to the dynamical evolution under the above Hamiltonian, both the atoms and field in quantum optics often relax. This relaxation may be simulated by treating the system as an open system and coupling it to reservoirs. We will ignore the reservoir coupling for now, since it is easy to add to the master equations later, as discussed in Sec. IV of I. We note only that the remarks made there apply here also provided we make the plausible assumption that the reservoirs for the field and for *each* atom are independent.

Rather than assuming the matter and field to be two coupled systems as in I, we consider them to be $N+1$ coupled systems (N atoms and field) and choose the relevant part of the density operator to be a product of the $N+1$ reduced density operators of the coupled subsystems, plus terms linear[24] in the correlation between one atom and the field

$$\Delta\chi(\alpha,t) \equiv \chi(\alpha,t) - R(t)\rho_1(\alpha,t) \;. \tag{3.2}$$

Hence, $F_{N,r}$ is given by[6]

$$F_{N,r}(t) = F_{N,nc}(t) + \sum_{\alpha}^{N} \Delta\chi(\alpha,t) \prod_{\beta}^{N}{}' \rho_1(\beta,t)$$
$$= \sum_{\alpha}^{N} \chi(\alpha,t) \prod_{\beta}^{N}{}' \rho_1(\beta,t) - (N-1)F_{N,nc}(t) \,, \tag{3.3a}$$

where $F_{N,nc}(t)$ is the uncorrelated density operator

$$F_{N,nc}(t) \equiv R(t) \prod_{\alpha}^{N} \rho_1(\alpha, t) \tag{3.3b}$$

and primes in products or sums have the same meaning as in Sec. II. The reader can easily check that $F_{N,r}$ is normalized

$$\mathrm{Tr} F_{N,r} = 1 , \tag{3.4}$$

where

$$\mathrm{Tr} \equiv \mathrm{Tr}_f \prod_{\alpha}^{N} \mathrm{Tr}_\alpha , \tag{3.5}$$

that the appropriate number of traces of $F_{N,r}$ yields χ, R, and ρ_1, as indicated in Eqs. (1.18) and (1.19), and that

$$\mathrm{Tr}_N F_{N,r}(1, \ldots, N, t) = F_{N-1,r}(1, \ldots, N-1, t) . \tag{3.6}$$

To find the TDPO which gives rise to the ME for $\dot{F}_{N,r}$, one could begin as in Sec. II by considering a system composed of only two atoms and the field. However, it is simpler to guess the form of the PO from the PO of Eq. (2.13) for a system of N gas molecules by considering the difference between the interactions involved in the gas-kinetic and quantum-optical cases. In the gas-kinetic case, one has a system where interactions occur between every pair of the N particles, or $\binom{N}{2}$ interactions. On the other hand, in the quantum-optical case, interactions occur only between each of the N atoms and the field, for a total of N interactions. The latter situation is analogous to a system of $N+1$ molecules, where one molecule is tagged and interactions occur between the tagged molecule and the N untagged molecules, while untagged molecules are not allowed to interact with one another. Thus double sums in Eq. (2.13) will be replaced by single sums, F_1 by R or ρ, F_2 by χ, and $V^{-1}\int d\alpha$ by Tr_α. In addition, the expression $\binom{N}{2}$ in the last term representing the number of interactions should be replaced by N, and the symbol N in the third term representing the number of interacting subsystems should be replaced by $N+1$. With these rules we may write the PO[20,6]

$$\begin{aligned} P(t) = {} & \sum_{\alpha} {\prod_{\beta}}' \rho_1(\beta, t) \mathrm{Tr}_\beta \\ & + \sum_{\alpha} \chi(\alpha, t) \mathrm{Tr}_\alpha \mathrm{Tr}_f {\sum_{\beta}}' {\prod_{\gamma}}'' \rho_1(\gamma, t) \mathrm{Tr}_\gamma \\ & - (N-1) \sum_{\alpha} \chi(\alpha, t) {\prod_{\beta}}' \rho_1(\beta, t) \mathrm{Tr} - (N-1) P_{nc}(t) , \end{aligned} \tag{3.7}$$

where $P_{nc}(t)$ is a TDPO which satisfies

$$\begin{aligned} & P_{nc}(t) F_N(t) = F_{N,nc}(t) , \\ & P_{nc}(t) \dot{F}_N(t) = \dot{F}_{N,nc}(t) , \end{aligned} \tag{3.8}$$

and is given explicitly by analogy with Eq. (I3.11),

$$\begin{aligned} P_{nc}(t) = {} & R(t) \mathrm{Tr}_f \sum_{\alpha} {\prod_{\beta}}' \rho_1(\beta, t) \mathrm{Tr}_\beta \\ & + \prod_{\alpha} \rho_1(\alpha, t) \mathrm{Tr}_m \\ & - N R(t) \prod_{\alpha} \rho_1(\alpha, t) \mathrm{Tr} . \end{aligned} \tag{3.9}$$

The symbol Tr_m denotes a trace over all atomic degrees of freedom.

In order for Eq. (3.7) to represent the desired TDPO, it must satisfy Eqs. (1.10a), (1.13b), and (1.9a), with $F_r(t)$ given by Eq. (3.3a). First, we have immediately

$$\begin{aligned} P(t) F_N(t) = {} & [1 + (N-1) - (N-1)] \\ & \times \sum_{\alpha} \chi(\alpha, t) {\prod_{\beta}}' \rho_1(\beta, t) - (N-1) F_{N,nc}(t) \\ = {} & F_{N,r}(t) , \end{aligned} \tag{3.10a}$$

$$\begin{aligned} P(t) \dot{F}_N(t) = {} & \sum_{\alpha} \dot{\chi}(\alpha, t) {\prod_{\beta}}' \rho_1(\beta, t) \\ & + \sum_{\alpha} \chi(\alpha, t) {\sum_{\beta}}' \dot{\rho}_1(\beta, t) {\prod_{\gamma}}'' \rho_1(\gamma, t) \\ & - (N-1) \dot{F}_{N,nc}(t) \\ = {} & \dot{F}_{N,r}(t) . \end{aligned} \tag{3.10b}$$

Next, we must show that $P(t)$ is a PO, or that $P(t)P(t') = P(t)$. This proof is not as lengthy as the corresponding proof of Sec. II, but is still somewhat tedious; it will be carried out in Appendix B. Since $P(t)$ is a sum of terms, as in Eq. (2.9), the proof will demonstrate the set of simpler relations in Eq. (2.10), instead of the TDPO property.

Now that we have obtained the correct PO for the quantum-optical problem, we are ready to derive the quantum-optical ME for $\dot{F}_{N,r}$. In this case we interpret Eq. (1.1) to be the quantum-mechanical Liouville equation with L given by Eq. (1.8), and the ME of Eq. (1.11) still follows.[25] Its explicit form is obtained by substituting Eqs. (3.7) and (3.3a) and simplifying. We choose the initial instant to be $t_0 = 0$ and assume that there are no irreducible two-atom–plus–field correlations initially, that is $F_{N,i}(0) = 0$. (Usually there are no atom-field correlations at all initially.)

We will need the effect of the PO on both the unperturbed and perturbation Liouvillians. From Eqs. (3.7), (I2.2), and (3.1) and the relations $\mathrm{Tr}_f L_f = \mathrm{Tr}_\alpha L_\alpha^0 = \mathrm{Tr}_f \mathrm{Tr}_\alpha L'_\alpha \equiv 0$, one has

$$P(t)L^0 = \sum_\alpha (L_f + L_\alpha^0){\prod_\beta}' \rho_1(\beta,t)\mathrm{Tr}_\beta + {\sum_{\alpha,\beta}}' L_\alpha^0 \chi(\beta,t)\mathrm{Tr}_\beta \mathrm{Tr}_f {\prod_\gamma}'' \rho_1(\gamma,t)\mathrm{Tr}_\gamma$$
$$-(N-1)\left(L_f \prod_\alpha \rho_1(\alpha,t)\mathrm{Tr}_m + R(t)\mathrm{Tr}_f \sum_\alpha L_\alpha^0 {\prod_\beta}' \rho_1(\beta,t)\mathrm{Tr}_\beta\right) , \tag{3.11}$$

$$P(t)L' = \sum_\alpha L'_\alpha {\prod_\beta}' \rho_1(\beta,t)\mathrm{Tr}_\beta + {\sum_{\alpha,\beta}}' [\rho_1(\alpha,t)\mathrm{Tr}_\alpha L'_\alpha + \chi(\beta,t)\mathrm{Tr}_\beta \mathrm{Tr}_f L'_\alpha] {\prod_\gamma}'' \rho_1(\gamma,t)\mathrm{Tr}_\gamma$$
$$-(N-1)\left(R(t)\mathrm{Tr}_f \sum_\alpha L'_\alpha + \sum_\alpha \rho_1(\alpha,t)\mathrm{Tr}_\alpha L'_\alpha\right){\prod_\beta}' \rho_1(\beta,t)\mathrm{Tr}_\beta . \tag{3.12}$$

Operating with these equations on $F_{N,r}(t)$ yields two of the terms on the left-hand side of the ME:

$$P(t)L^0 F_{N,r}(t) = \sum_\alpha (L_f + L_\alpha^0)\chi(\alpha,t) {\prod_\beta}' \rho_1(\beta,t) + {\sum_{\alpha,\beta}}' \chi(\alpha,t) L_\beta^0 \rho_1(\beta,t) {\prod_\gamma}'' \rho_1(\gamma,t)$$
$$-(N-1)\left(L_f R(t) \prod_\alpha \rho_1(\alpha,t) + R(t) \sum_\alpha L_\alpha^0 \rho_1(\alpha,t) {\prod_\beta}' \rho_1(\beta,t)\right) , \tag{3.13}$$

$$P(t)L' F_{N,r}(t) = \sum_\alpha L'_\alpha \chi(\alpha,t) {\prod_\beta}' \rho_1(\beta,t) + {\sum_{\alpha,\beta}}' [\rho_1(\alpha,t)\mathrm{Tr}_\alpha L'_\alpha F_{2,r}(\alpha,\beta,t) + \chi(\alpha,t)\mathrm{Tr}_f L'_\beta \chi(\beta,t)] {\prod_\gamma}'' \rho_1(\gamma,t)$$
$$-(N-1)\left(R(t)\sum_\alpha \mathrm{Tr}_f L'_\alpha \chi(\alpha,t) + \sum_\alpha \rho_1(\alpha,t)\mathrm{Tr}_\alpha L'_\alpha \chi(\alpha,t)\right) {\prod_\beta}' \rho_1(\beta,t) , \tag{3.14}$$

where Eqs. (3.6), (1.18), and (1.19) have been used.

Equations (3.11)–(3.14) will also be useful in representing the right-hand side of the ME. In addition, to reduce the right-hand side, we will need the very useful result that the projection operator $Q(t)$ on the irrelevant subspace vanishes identically when traced over any $N-1$ atomic variables, that is

$${\prod_\alpha}' \mathrm{Tr}_\alpha [1 - P(t)] \equiv 0 . \tag{3.15}$$

The proof of this relation is given in Appendix B. The property is analogous to that expressed by Eq. (2.20) for the correlated-gas PO, by Eq. (I2.14) for the uncorrelated-atom-field PO, and by Eq. (I3.18) for the uncorrelated-gas PO. Using Eq. (3.15), we can show that the first L factor on the right-hand side of the ME, Eq. (1.11), can be replaced by L'. The product $\mathcal{G}(t,t')[1-P(t')]$ always has a PO of the form $[1-P(\bar{t})]$ as its leftmost operator. Since, according to Eq. (3.11), each term of $P(t)L^0$ is terminated by at least $N-1$ traces over atoms, the result

$$P(t)L^0 \mathcal{G}(t,t')[1-P(t')] \equiv 0 \tag{3.16}$$

follows immediately. In addition, we can prove that L^0 in the second factor L of the right-hand side does not contribute. We do this by showing that

$$[1-P(t)]\, L^0 F_{N,r}(t) = 0 , \tag{3.17}$$

or, equivalently,

$$P(t)L^0 F_{N,r}(t) = L^0 F_{N,r}(t) . \tag{3.17$'$}$$

The left-hand side of Eq. (3.17′) is given in Eq. (3.13); we have only to compute the right-hand side. This is done in a completely straightforward manner using Eqs. (I2.2) and (3.1) for L^0, with the result being identical to Eq. (3.17′).

Having eliminated L^0 from both factors L on the right-hand side of the ME, we now examine the remaining factors L'. By Eq. (3.15), it is clear that only the second term of Eq. (3.12) contributes to the first factor L', since it alone contains only $N-2$ traces over atoms. Hence, it follows that

$$P(t)L' \mathcal{G}(t,t')[1-P(t')]$$
$$= {\sum_{\alpha,\beta}}' \rho_1(\alpha,t)\mathrm{Tr}_\alpha L'_\alpha {\prod_\gamma}'' \rho_1(\gamma,t)\mathrm{Tr}_\gamma \times \mathcal{G}(t,t')[1-P(t')] . \tag{3.18}$$

Finally, the last factor L' can be simplified by evaluating

$$[1-P(t)]L'F_{N,r}(t) \equiv L'F_{N,r}(t) - P(t)L'F_{N,r}(t) . \tag{3.19}$$

The second term on the right-hand side is given by Eq. (3.14), and the first term is

$$L'F_{N,r}(t) = \sum_\alpha L'_\alpha \chi(\alpha,t) {\prod_\beta}' \rho_1(\beta,t)$$
$$+ {\sum_{\alpha,\beta}}' L'_\beta \chi(\alpha,t)\rho_1(\beta,t) {\prod_\gamma}'' \rho_1(\gamma,t)$$
$$-(N-1)\sum_\alpha L'_\alpha R(t)\rho_1(\alpha,t) {\prod_\beta}' \rho_1(\beta,t) . \tag{3.20}$$

Combining Eqs. (3.19), (3.20), and (3.14) and simplifying, one obtains

$$[1-P(t)]\,L'F_{N,r}(t) = \sum_{\alpha}\Delta_t L'_\alpha\Big(\sum_{\beta}{}'\chi(\beta,t)\prod_{\substack{\gamma\\(\gamma\neq\beta)}}{}'\rho_1(\gamma,t) - (N-1)R(t)\prod_{\beta}\rho_1(\beta,t)\Big)\;. \tag{3.21}$$

We have defined $\Delta_t L'_\alpha$ analogously to Eq. (I2.20) as

$$\Delta_t\,L'_\alpha \equiv L'_\alpha - \langle L'_\alpha\rangle_{\alpha,t} - \langle L'_\alpha\rangle_{f|\alpha,t}\;, \tag{3.22}$$

where $\langle\cdots\rangle_{\alpha,t}\equiv \mathrm{Tr}_\alpha[\cdots\rho_1(\alpha,t)]$ denotes a partial average over particle α. In addition, we have introduced conditional averages,

$$\langle O(f,\alpha)\rangle_{f|\alpha,t}\equiv \mathrm{Tr}_f O(f,\alpha)\chi(f|\alpha,t)\;, \tag{3.23}$$

where the field dependence of the operators has been indicated explicitly by the symbol f and $\chi(f|\alpha,t)$ is the conditional density operator defined by

$$\chi(\alpha,t)\equiv\chi(f|\alpha,t)\rho_1(\alpha,t)\;. \tag{3.24}$$

The ME for $\dot F_{N,r}$ may now be written explicitly by combining Eqs. (1.10), (3.13), (3.14), (3.16)–(3.18), and (3.21); the ME is

$$\begin{aligned}\dot F_{N,r}(t) &- \sum_{\alpha}(L_f + L^0_\alpha + L'_\alpha)\chi(\alpha,t)\prod_{\beta}{}'\rho_1(\beta,t) - \sum_{\alpha}(L^0_\alpha + \langle L'_\alpha\rangle_{\alpha,t} + \langle L'_\alpha\rangle_{f|\alpha,t})\Big(\sum_{\beta}{}'\chi(\beta,t)\prod_{\substack{\gamma\\(\gamma\neq\beta)}}{}'\rho_1(\gamma,t)\\ &- (N-1)R(t)\sum_{\beta}{}'\rho_1(\beta,t)\Big)\\ &= \sum_{\alpha,\beta}{}'\rho_1(\alpha,t)\mathrm{Tr}_\alpha L'_\alpha\prod_{\gamma}{}''\rho_1(\gamma,t)\mathrm{Tr}_\gamma\int_0^t dt'\,g(t,t')\sum_{\delta}\Delta_{t'}L'_\delta\Big(\sum_{\substack{\epsilon\\(\epsilon\neq\delta)}}{}'\chi(\epsilon,t')\prod_{\substack{\zeta\\(\zeta\neq\epsilon)}}{}'\rho_1(\zeta,t')\\ &\qquad - (N-1)R(t')\prod_{\epsilon}\rho_1(\epsilon,t')\Big)\;. \end{aligned}\tag{3.25}$$

Since we are really interested in the density operator χ, we can operate on Eq. (3.25) with $N-1$ traces over all atoms but the αth one to obtain the ME for $\dot\chi(\alpha,t)$. After some cancellation and rearrangement, one obtains[6]

$$\begin{aligned}\dot\chi(\alpha,t) &- (L_f + L^0_\alpha + L'_\alpha)\chi(\alpha,t) - \sum_{\substack{\beta\\(\beta\neq\alpha)}}{}'\mathrm{Tr}_\beta L'_\beta F_{2,r}(\alpha,\beta,t)\\ &= \sum_{\substack{\beta\\(\beta\neq\alpha)}}{}'\mathrm{Tr}_\beta L'_\beta\prod_{\gamma}{}''\mathrm{Tr}_\gamma\int_0^t dt'\,g(t,t')\sum_{\delta}\Delta_{t'}L'_\delta\Big(\sum_{\substack{\epsilon\\(\epsilon\neq\delta)}}{}'\chi(\epsilon,t')\prod_{\substack{\zeta\\(\zeta\neq\epsilon)}}{}'\rho_1(\zeta,t') - (N-1)R(t')\prod_{\epsilon}\rho_1(\epsilon,t')\Big)\;.\end{aligned}\tag{3.26}$$

This *exact* expression cannot be simplified further without introducing approximations. The last term on the left-hand side incorporates the effect of reducible correlations between two atoms and the field, while the right-hand side incorporates irreducible two-atom-plus-field (as well as higher-order) correlations.

Since Eq. (3.26) depends on both R and ρ_1, we must obtain dynamical equations for the latter two density operators also. These are derived most easily by tracing Eq. (3.26) over the αth atom and over the field, respectively. Performing first the trace over matter, we note that neither the right-hand side nor the last term of the left-hand side of Eq. (3.26) contributes, for one has a complete trace of a commutator in each case. The result is

$$\dot\rho_1(\alpha,t) - L^0_\alpha\rho_1(\alpha,t) = \mathrm{Tr}_f L'_\alpha\chi(\alpha,t)\;. \tag{3.27}$$

Similarly, performing a trace over the field, the right-hand side of Eq. (3.26) still does not contribute, because one has $N-1$ traces of $[1-P(t)]$ over atomic degrees of freedom, but every term of the left-hand side makes a contribution. In this case, one obtains

$$\dot R(t) - L_f\,R(t) = \sum_{\beta}\mathrm{Tr}_\beta L'_\beta\chi(\beta,t)\;. \tag{3.28}$$

Equations (3.27) and (3.28) are the first members of the BBGKY hierarchy[22] for the quantum-optical Hamiltonian, relating $\dot R$ and $\dot\rho_1$ to χ. Together Eqs. (3.26)–(3.28) form an *exact* closed set of coupled ME's for χ, R, and ρ_1. In Sec. V we will apply these equations to the single-mode laser, making the first Born approximation in the $\dot\chi$ equation. However, before doing this, we will return in Sec. IV to the gas-kinetic ME of Sec. II and consider approximate solutions to the exact ME for the gas.

IV. APPROXIMATE MASTER EQUATIONS IN GAS KINETICS

The starting point in this section is the set of two coupled exact ME's for the singlet and doublet distributions $F_1(1,t)$ and $F_2(1,2,t)$ derived in Sec. II, Eqs. (2.26) and (2.27), obtained by assuming that the relevant part of the distribution function contains two-body correlations. We will carry out a density expansion of the $\dot{F}_2$ equation and, in addition, make the binary-collision approximation[13] (BCA). It will be seen that even the first-order term in the density N/V gives rise to a transcendental dependence on density in the $\dot{F}_1$ equation and provides a correction to the equation for the singlet distribution leading to a good description of mean-free-path effects in dense gases.

This behavior is in contrast to that found in I, where the equation for the singlet distribution, obtained under the assumption that $F_{N,r}(t)$ was uncorrelated, yielded the dilute-gas Boltzmann equation to lowest order in N/V. Before considering the density expansion of the present equations, we will illustrate the method by reconsidering briefly the derivation of the Boltzmann equation through expansion to order N/V of the equation for the singlet distribution of I, Eq. (I3.22). This is desirable because the derivation of the first-order terms carried out in I is not a good guide in the current case. The simplicity of the result was obscured somewhat by the desire to illustrate the origin there of certain correction terms of order $(N/V)^2$ derived by Klimontovich[26] which make the total energy, kinetic plus potential, a conserved quantity and by the desire to make contact with the expressions of Klimontovich.

We recall from I that, in the kinetic regime, we are interested in phenomena occurring on a time scale τ, $\tau_{\text{int}} \ll \tau \ll \tau_{\text{rel}}$, where τ_{int} is the duration of a collision and τ_{rel} is the time between collisions. Moreover, this allows us to introduce an expansion in powers of the density since

$$\tau_{\text{int}}/\tau_{\text{rel}} \cong N r_0^3/V \equiv \lambda \ll 1 \,, \tag{4.1}$$

where r_0 is the hard-core radius. Starting from the exact ME for F_1 in the case where $F_{N,r}$ is uncorrelated, Eq. (I3.22), we show that to first order in λ only the first two terms contribute and even then only in simplified form. Using the operator relation

$$\int \frac{d2}{V}\, L'_{12} < N \int \frac{d2}{V}\, L'_{12} = O(\lambda) \,. \tag{4.2}$$

along with the identities, Eqs. (2.16), the integral over $3,\ldots,N$ can be performed in Eq. (I3.22) with the result for $\dot{F}_1$ given in Eq. (I3.25). Note that $\mathscr{G}^{(2)}$ appearing in this equation and defined in Eq. (I3.26) is not exactly the same as $\mathscr{G}^{(N)}$ with $N=2$, but agrees with the latter to order λ, as discussed in footnote 14 of I.

To proceed further we note that $\mathscr{G}^{(2)}$ satisfies the integral equation[27]

$$\begin{aligned}\mathscr{G}^{(2)}&(1,2,t,t-\tau)\\ &= G^{(2)}(1,2,\tau)\\ &\quad -\int_0^\tau d\tau'\, \mathscr{G}^{(2)}(1,2,t,t-\tau')\\ &\qquad \times \mathfrak{L}_{(2)}(1,2,t-\tau')G^{(2)}(1,2,\tau-\tau') \,,\end{aligned} \tag{4.3}$$

where[27]

$$\mathfrak{L}_{(2)}(1,2,t-\tau) \equiv L_1{}^0 F_1(2,t-\tau)\int\frac{d2}{V} + L_2{}^0 F_1(1,t-\tau)\int\frac{d1}{V} \tag{4.4}$$

and

$$G^{(2)}(1,2,\tau) \equiv \exp[L_{(2)}(1,2)\tau] \tag{4.5}$$

is the solution to Eq. (I3.29). By iteration of Eq. (I3.25), we find that

$$F_1(1,t-\tau) = G^{(1)}(1,-\tau)F_1(1,t) + O(\lambda) \,, \tag{4.6}$$

where

$$G^{(1)}(1,\tau) \equiv \exp(L_1^0\tau) \,. \tag{4.7}$$

In addition, we iterate Eq. (4.3) once to obtain[27]

$$\begin{aligned}\mathscr{G}^{(2)}&(1,2,t,t-\tau)\\ &\cong G^{(2)}(1,2,\tau)\\ &\quad -\int_0^\tau d\tau'\, G^{(2)}(1,2,\tau')\\ &\qquad \times \mathfrak{L}_{(2)}(1,2,t-\tau')G^{(2)}(1,2,\tau-\tau') \,.\end{aligned} \tag{4.8}$$

It is not so clear that the iteration in Eq. (4.8) is related to an expansion in powers of λ. However, we will show in Appendix C that the term $\sim\lambda$ in the ME for $\dot{F}_1$ is obtained by substituting the first term of the $\mathscr{G}^{(2)}$ equation, Eq. (4.8), into the second of the $\dot{F}_1$ equation, Eq. (I3.25). On the other hand, the second term of the $\mathscr{G}^{(2)}$ equation substituted into the second of the $\dot{F}_1$ equation combines with the result of substituting the first term of $\mathscr{G}^{(2)}$ into the third term of $\dot{F}_1$ to yield a contribution $\sim\lambda^2$. The result is

$$\begin{aligned}\dot{F}_1&(1,t) - L_1^0 F_1(1,t)\\ &= N\int\frac{d2}{V}\, L'_{12}\lim_{\tau\to\infty}\mathcal{S}^{(2)}(1,2,\tau)F_1(1,t)F_1(2,t)\\ &\quad + N\int_0^\infty d\tau\int\frac{d2}{V}\, L'_{12}\mathcal{S}^{(2)}(1,2,\tau)\\ &\qquad \times\left(\frac{\partial}{\partial t} - L^0{}_1 - L_2{}^0\right)F_1(1,t)F_1(2,t) \,,\end{aligned} \tag{4.9}$$

where

$$\mathcal{G}^{(2)}(1,2,\tau)\equiv G^{(2)}(1,2,\tau)G^{(1)}(1,-\tau)G^{(1)}(2,-\tau)\ . \tag{4.10}$$

Clearly, by iteration, one finds

$$\left(\frac{\partial}{\partial t}-L^0{}_1-L^0{}_2\right)F_1(1,t)F_2(2,t)=O(\lambda)\ , \tag{4.11}$$

so that the term containing it in Eq. (4.9) is $\sim\lambda^2$ and can be neglected. The remaining terms of Eq. (4.9) are exactly the Boltzmann equation for the singlet distribution F_1, as derived in I.[28]

We have illustrated the technique for expanding to first order in N/V (or λ) by considering the general ME for $\dot{F}_1$ derived in I, where $F_{N,r}$ was taken to be uncorrelated. We wish now to apply the same technique to the more difficult density expansion of the doublet ME, Eq. (2.26), derived in Sec. II based on a relevant part of the probability density containing two-body correlations. When one solves the resulting equation for F_2 and substitutes the result into Eq. (2.27) for $\dot{F}_1$, one obtains an equation for $\dot{F}_1$ which is a transcendental function of the density, even though the $\dot{F}_2$ ME contained only terms up to first order. One hopes by this technique to bring into the theory a collisional damping factor depending upon the mean free path and, hence, obtain results valid at higher densities than would be possible by a naive density expansion of the $\dot{F}_1$ equation.

Using Eq. (4.2) we can perform the integrals over $4,\ldots,N$ in Eq. (2.26) to lowest order in λ with the result

$$\begin{aligned}\dot{F}_2(1,2,t)-L_{(2)}(1,2)F_2(1,2,t)=(N-2)\int\frac{d3}{V}\,(L'_{13}+L'_{23})\Big(&\lim_{\tau\to\infty}\mathcal{G}^{(3)}(1,2,3,t,t-\tau)F_{3,r}(1,2,3,t-\tau)\\&+\int_0^\infty d\tau\,\mathcal{G}^{(3)}(1,2,3,t,t-\tau)\,\frac{\partial}{\partial t}\,F_{3,r}(1,2,3,t-\tau)\Big)\ ,\end{aligned} \tag{4.12}$$

where

$$\mathcal{G}^{(3)}(1,2,3,t,t-\tau)\equiv T\exp\int_{t-\tau}^{t}dt'[L_{(3)}(1,2,3)-\mathfrak{L}_{(3)}(1,2,3,t')]\ , \tag{4.13a}$$

$$\begin{aligned}\mathfrak{L}_{(3)}(1,2,3,t)\equiv L_1^0\Big(&F_1(2,t)\int\frac{d2}{V}+(2\to3)+[F_2(2,3,t)-2F_1(2,t)F_1(3,t)]\int\frac{d2}{V}\int\frac{d3}{V}\Big)\\&+L'_{12}F_1(3,t)\int\frac{d3}{V}+\text{cycl}(1,2,3)\ ,\end{aligned} \tag{4.13b}$$

and we have written the phase-space arguments explicitly. Once again, $\mathcal{G}^{(3)}$ is not identical with the operator obtained by setting $N=3$ in Eqs. (2.19), but differs from it only in terms $\sim\lambda$. In deriving Eqs. (4.13) we have used an alternate form of Eq. (2.14),

$$\begin{aligned}P_N(t)L_\alpha^0=L_\alpha^0\Big\{&\sum_\beta{}'\prod_\gamma{}''F_1(\gamma,t)\int\frac{d\gamma}{V}+\frac{1}{2}\sum_\beta{}'\sum_\gamma{}''F_2(\beta,\gamma,t)\int\frac{d\beta}{V}\int\frac{d\gamma}{V}\prod_\delta{}'''F_1(\delta,t)\int\frac{d\delta}{V}\\&-\left[\tbinom{N}{2}-1\right]\prod_\beta{}'F_1(\beta,t)\int\frac{d\beta}{V}\Big\}\ ,\end{aligned} \tag{4.14a}$$

along with the expansion to lowest order in λ of Eq. (2.15),

$$P_N(t)L'_{\alpha\beta}=L'_{\alpha\beta}\prod_\gamma{}''F_1(\gamma,t)\int\frac{d\gamma}{V}+O(\lambda)\ . \tag{4.14b}$$

By iteration of Eq. (4.12), it is clear that

$$F_2(1,2,t-\tau)=G^{(2)}(1,2,-\tau)F_2(1,2,t)+O(\lambda)\ . \tag{4.15}$$

Moreover, the time-development operator $\mathcal{G}^{(3)}$ is related to $L_{(3)}$ and

$$G^{(3)}(1,2,3,\tau)\equiv\exp[L_{(3)}(1,2,3)\tau] \tag{4.16}$$

in the same way as $\mathcal{G}^{(2)}$ is related to $\mathfrak{L}_{(2)}$ and $G^{(2)}$, that is by an analogue of Eq. (4.3). Iterating this equation once results in

$$\mathcal{G}^{(3)}(1,2,3,t,t-\tau)\cong G^{(3)}(1,2,3,\tau)-\int_0^\tau d\tau'\,G^{(3)}(1,2,3,\tau')\mathfrak{L}_{(3)}(1,2,3,t-\tau')G^{(3)}(1,2,3,\tau-\tau')\ . \tag{4.17}$$

Proceeding as in the density expansion of the $\dot{F}_1$ ME of I, we now substitute Eqs. (4.15) for F_2, (4.6) for F_1, and (4.17) for $\mathcal{G}^{(3)}$ into the ME for $\dot{F}_2$, Eq. (4.12). The desired term ~λ in the ME is obtained from the first term of the $\mathcal{G}^{(3)}$ equation in the first term of the right member of the $\dot{F}_2$ equation, whereas the second term of the $\mathcal{G}^{(3)}$ equation in the first term of the $\dot{F}_2$ equation combines with the first term of $\mathcal{G}^{(3)}$ in the second term of $\dot{F}_2$ to yield a negligible contribution ~λ^2. The result is

$$\begin{aligned}
&\dot{F}_2(1,2,t)-L_{(2)}(1,2)F_2(1,2,t)\\
&\quad=N\int\frac{d3}{V}(L'_{13}+L'_{23})\Big\{\lim_{\tau\to\infty}G^{(3)}(1,2,3,\tau)F_{3,r}(1,2,3,t-\tau)\\
&\qquad+\int_0^\infty d\tau\,G^{(3)}(1,2,3,\tau)\Big[G^{(2)}(1,2,-\tau)G^{(1)}(3,-\tau)\left(\frac{\partial}{\partial t}-L_{(2)}(1,2)-L_3^0\right)F_2(1,2,t)F_1(3,t)\\
&\qquad\qquad+\mathrm{cycl}(1,2,3)-2\prod_{\alpha}^{3}G^{(1)}(\alpha,-\tau)\left(\frac{\partial}{\partial t}-\sum_{\beta}^{3}L_\beta^0\right)\prod_{\gamma}^{3}F_1(\gamma,t)\Big]\Big\}.
\end{aligned}\tag{4.18}$$

The details are carried out in Appendix C. By iteration of Eqs. (4.18) and (2.27), one finds that

$$\left(\frac{\partial}{\partial t}-L_{(2)}(1,2)-L_3^0\right)F_2(1,2,t)F_1(3,t)=O(\lambda)\tag{4.19a}$$

and

$$\left(\frac{\partial}{\partial t}-\sum_{\beta}^{3}L_\beta^0\right)\prod_{\gamma}^{3}F_1(\gamma,t)=O(\lambda)\ ,\tag{4.19b}$$

so that the integral over τ in Eq. (4.18) is ~λ^2 and, hence, is negligible. Writing the remaining term explicitly, still keeping only terms ~λ, one obtains

$$\begin{aligned}
&\dot{F}_2(1,2,t)-L_{(2)}(1,2)F_2(1,2,t)\\
&\quad=N\int\frac{d3}{V}L'_{13}\lim_{\tau\to\infty}G^{(3)}(1,2,3,\tau)\\
&\qquad\times\left(G^{(2)}(1,2,-\tau)G^{(1)}(3,-\tau)F_2(1,2,t)F_1(3,t)+\mathrm{cycl}(1,2,3)-2\prod_{\alpha}^{3}G^{(1)}(\alpha,-\tau)F_1(\alpha,t)\right)+(1\leftrightarrow 2)\ .
\end{aligned}\tag{4.20}$$

Solution of Eq. (4.20) requires knowledge of the three-body collision operator $G^{(3)}$. Although not strictly a consequence of the density expansion, we can introduce the BCA at this point to eliminate $G^{(3)}$ in favor of expressions depending only on the binary-collision operator $G^{(2)}$ and the free-particle evolution operator $G^{(1)}$. In the term shown explicitly in Eq. (4.20) involving L'_{13}, the BCA allows us to neglect all interaction Liouvillians in $G^{(3)}$ and $G^{(2)}$ except for L'_{13}. Hence, one has

$$\begin{aligned}G^{(3)}(1,2,3,\tau)&\cong\exp[(L_1^0+L_2^0+L_3^0+L'_{13})\tau]\\&=G^{(2)}(1,3,\tau)G^{(1)}(2,\tau)\ ,\end{aligned}\tag{4.21a}$$

$$\begin{aligned}G^{(2)}(1,2,-\tau)&\cong\exp[-(L_1^0+L_2^0)\tau]\\&=G^{(1)}(1,-\tau)G^{(1)}(2,-\tau)\ ,\end{aligned}\tag{4.21b}$$

and so forth. In the BCA, Eq. (4.20) assumes the form

$$\begin{aligned}
&\dot{F}_2(1,2,t)-L_{(2)}(1,2)F_2(1,2,t)\\
&\quad=N\int\frac{d3}{V}L'_{13}\{F_2(1,3,t)F_1(2,t)+\lim_{\tau\to\infty}\mathcal{G}^{(2)}(1,3,\tau)\\
&\qquad\times[\Delta F_2(1,2,t)F_1(3,t)+(1\leftrightarrow 3)]\}\\
&\quad+(1\leftrightarrow 2)\ ,
\end{aligned}\tag{4.22}$$

where we have introduced the correlation distribution ΔF_2 defined in Eq. (2.1).

Although Eq. (4.22) is the desired result of our expansion of $\dot{F}_2$ to first order in λ, the virtues of the equation are more apparent if it is rewritten as an equation for the time derivative of ΔF_2. The first term on the right-hand side can then be eliminated by using Eq. (2.27), with the result

$$\begin{aligned}
&\left(\frac{\partial}{\partial t}-L_{(2)}(1,2)\right)\Delta F_2(1,2,t)\\
&\quad=\frac{N}{V}\int d3\,\Lambda(1,2,3,t)\Delta F_2(1,2,t)\\
&\qquad+L'_{12}F_1(1,t)F_1(2,t)\ ,
\end{aligned}\tag{4.22$'$}$$

where

$$\Lambda(1,2,3,t) \equiv [L'_{13}\mathcal{S}^{(2)}(1,3,\infty)(1+\mathcal{P}_{13}) + (1 \leftrightarrow 2)]F_1(3,t) . \tag{4.23}$$

Here $\mathcal{P}_{\alpha\beta}$ is the permutation operator on particles α and β. Equations (4.22′) and (2.27) constitute closed, Markoffian equations for F_1 and F_2 which are equivalent to Dorfman's renormalized kinetic equation.[14] Since Eq. (4.22′) is a linear inhomogeneous integrodifferential equation for ΔF_2, we can solve it for ΔF_2 in terms of F_1. The result is

$$\Delta F_2(1,2,t) = \Delta F_2(1,2,-\infty) + \int_{-\infty}^{t} dt' \exp\left[\int_{t'}^{t} dt'' \left(L_{(2)}(1,2) + \frac{N}{V}\int d3\Lambda(1,2,3,t'')\right)\right] L'_{12}F_1(1,t')F_1(2,t') . \tag{4.24}$$

When we substitute this solution into Eq. (2.27), we obtain a self-contained nonlinear non-Markoffian integrodifferential equation for $\dot{F}_1$ which is infinite order in the density N/V, since N/V appears in the exponent on the right-hand side of Eq. (4.24). The non-Markoffian behavior and the transcendental density dependence arise from an effective resummation of an infinite class of diagrams representing a time integral over the past history of particles 1 and 2 interacting, one at a time, through repeated Boltzmann collisions with the remaining particles. As a consequence of the interaction of particles 1 and 2 with the remaining particles, the time integral in the exponential of Eq. (4.24) is effectively of the form $\exp(-\tau/\tau^*)$, where τ^* is the mean free time between collisions.

In passing, note that if one were to repeat the procedure of this section to calculate $\Delta\dot{F}_2$ to second order in the density before solving for ΔF_2 and substituting into Eq. (2.27) for $\dot{F}_1$, one would find the cutoff $\exp(-\tau/\tau^*)$ responsible for removing the divergence of the diffusion coefficient.[12] We feel the joint solution of our equations for $\dot{F}_1$ and $\dot{F}_2$ represents an appreciable simplification over most methods of obtaining mean-free-path effects in the kinetic equation for $\dot{F}_1$. Because of this simplification, we feel the present example demonstrates clearly the merits of considering an $F_{N,r}$ which contains two-body correlations, even for systems of low or moderate density.

The current method can be extended to the theory of liquids by applying the Rice-Allnatt[29] idea of separating the potential into a strong short-range repulsive part and a weak long-range attractive part. The resultant equations will be different due to the fact that our approach couples F_1 and F_2 strongly while the Rice-Allnatt theory results in completely separate equations for the singlet and doublet distribution functions.

We leave the problem of ME's in kinetic theory now and return, in Sec. V, to the exact quantum-optical ME's derived in Sec. III in order to apply them to the consideration of atom-atom correlations in lasers.

V. MASTER EQUATION FOR THE SINGLE-MODE LASER

In this section we apply the exact ME's for quantum optics, Eqs. (3.26)–(3.28), to the single-mode laser by restricting the number of modes coupled to the atoms to one and introducing reservoirs for the radiation field and for each atom. The field reservoir accounts for radiation losses from the optical cavity containing the atoms, while the atomic reservoirs destroy the phase of the atomic polarization and cause the population inversion to relax to a level determined by the pump. We show that in the first Born approximation (FBA), that is to order μ^2, the $\dot{\chi}$ equation gives rise to the conventional laser theory,[16–18] plus some additional two-body terms involving two-atom-plus-field correlations. We do not assume *a priori* that the two-body terms vanish, as is usually done. However, we are able to show explicitly that some of these terms make small but important corrections, within the usual range of laser parameters.

In order to introduce reservoirs, we proceed as in our previous treatment of superradiance ME's[30] and replace the field by an extended subsystem containing the field plus its reservoir; similarly, we replace the αth atom by an extended subsystem containing the atom plus its reservoir. The Hamiltonians of the extended subsystems are

$$\mathcal{H}_f = H_f + H^0_{\text{res},f} + H'_{\text{res},f} \tag{5.1a}$$

and

$$\mathcal{H}^0_\alpha = H^0_\alpha + H^0_{\text{res},\alpha} + H'_{\text{res},\alpha} , \tag{5.1b}$$

such that the total Hamiltonian of the extended system is[31]

$$\mathcal{H} = \mathcal{H}^0 + H' , \tag{5.2a}$$

$$\mathcal{H}^0 = \mathcal{H}_f + \sum_{\alpha}^{N} \mathcal{H}^0_\alpha . \tag{5.2b}$$

Here $H^0_{\text{res},f}$ and $H^0_{\text{res},\alpha}$ are the Hamiltonians of the field reservoir and reservoir for the αth atom, respectively, while $H'_{\text{res},f}$ and $H'_{\text{res},\alpha}$ are the corresponding system-reservoir interaction Hamiltonians. Of course, we may also introduce Liouvillians corresponding to each of the newly defined

Hamiltonians; in particular we will need $\mathcal{L}_f$, $\mathcal{L}^0_\alpha$, $\mathcal{L}^0$, and $\mathcal{L}$ corresponding to $\mathcal{H}_f$, $\mathcal{H}^0_\alpha$, $\mathcal{H}^0$, and $\mathcal{H}$. The derivation of Eqs. (1.11) and (1.12) and each step of Sec. III may now be carried out as before, with extended-system operators replacing system operators. The density operators in the resulting ME's are replaced by extended-system density operators. For example, L in Eq. (1.12) is replaced by $\mathcal{L}$, and $\chi(\alpha, t)$ in Eq. (3.26) is replaced by the reduced density operator of the field and the αth atom plus their respective reservoirs. By taking traces of these ME's over reservoir degrees of freedom, one obtains ME's for the system or subsystem alone.

The reservoirs play a critical role in ensuring the validity of the FBA. For the FBA to be valid in the $\dot{\chi}$ equation, we require an interaction time τ_{int} to be much shorter than the relaxation time τ_{rel} for the density operator χ. For the usual situation in a homogeneously broadened laser, the shortest damping time is T_2, the polarization decay time, and T_2 plays the role of τ_{int}. On the other hand, τ_{rel} is given by[17] $\tau_{\text{rel}}^{-1} = |\mu|^2 N T_2$, where μ is the atom-field coupling constant having dimensions of frequency, defined in Eq. (I4.7). (We have dropped the mode labels k and s since we are dealing with a single mode, having wave vector $\vec{k}$ and polarization index s, say.) Consequently, we find that the dimensionless ratio $\epsilon \equiv \tau_{\text{int}}/\tau_{\text{rel}} = |\mu|^2 T_2^2 N \ll 1$ for typical laser parameters, and we are justified in treating the right-hand side of Eq. (3.26) in the FBA.

To introduce the FBA into the extended-system version of Eq. (3.26), we replace[5] $\mathcal{L}$ by $\mathcal{L}^0$ in $\mathcal{G}(t, t')$, obtaining $\mathcal{G}_0(t, t')$ as in Eq. (I4.1), and use an analog of Eq. (I4.2) with $L^0 \to \mathcal{L}^0$. Then we obtain an equation for $\dot{\chi}$ by taking traces over the reservoirs. In addition, we choose to set average fields and average polarizations equal to zero by letting R and ρ_1 be diagonal operators in the H^0 representation. We obtain the following equation of motion for χ:

$$\begin{aligned}\dot{\chi}(\alpha, t) - (L_f + L^0_\alpha + L'_\alpha)\chi(\alpha, t) \\ - \sideset{}{'}\sum_{\substack{\beta \\ (\beta \neq \alpha)}} \langle L'_\beta \rangle_{\beta | f, t} R(t) \rho_1(\alpha, t) - \left(\frac{\delta\chi(\alpha, t)}{\delta t}\right)_{\text{res}} \\ = \sideset{}{'}\sum_{\substack{\beta \\ (\beta \neq \alpha)}} \int_0^t d\tau \, \text{Tr}_\beta L'_\beta \{\langle \tilde{\Delta}_{t-\tau} \mathcal{L}'_\beta(-\tau) \tilde{\rho}_1(\beta, t-\tau) \\ \times \Delta\tilde{\chi}(\alpha, t-\tau)\rangle_{\text{res}} + (\alpha \leftrightarrow \beta)\} . \end{aligned} \tag{5.3}$$

The Liouvillians occurring in Eq. (5.3) are related to Hamiltonians by Eq. (1.8), and the Hamiltonians are defined in Eqs. (I4.3)–(I4.7) and Eqs. (3.1), such that for the homogeneously broadened single-mode case,

$$H_f = \hbar\Omega(a^+ a^- + \tfrac{1}{2}) , \tag{5.4a}$$

$$H^0_\alpha = \tfrac{1}{2}\hbar\omega_0 s_\alpha , \tag{5.4b}$$

$$H'_\alpha = \hbar\mu e^{i\vec{k}\cdot\vec{X}_\alpha} \sigma^+_\alpha a^- + \mu^* e^{-i\vec{k}\cdot\vec{X}_\alpha} \sigma^-_\alpha a^+ . \tag{5.4c}$$

The conditional average $\langle L' \rangle_{\alpha | f, t}$ on the left-hand side of Eq. (5.3) is strictly analogous to the conditional average $\langle L' \rangle_{f | \alpha, t}$ of Eqs. (3.22)–(3.24), that is

$$\langle L'_\alpha \rangle_{\alpha | f, t} \equiv \text{Tr}_\alpha L'_\alpha \chi(\alpha | f, t) , \tag{5.5}$$

where $\chi(\alpha | f, t)$ is the conditional density operator satisfying

$$\chi(\alpha, t) \equiv \chi(\alpha | f, t) R(t) . \tag{5.6}$$

We have represented the time development of χ due to reservoirs alone symbolically by the term $(\delta\chi/\delta t)_{\text{res}}$.

In deriving the right-hand side of Eq. (5.3), we have assumed that the final density operator for the extended system appearing in the equation is a product of a system density operator and equilibrium density operators for the reservoirs. This assumption is consistent with the FBA and the fact that the reservoirs are large systems. The angular brackets with subscript "res" represent partial averages with respect to the reservoir density operators.[32] Density operators with tildes are interaction-picture operators,

$$\begin{aligned} \tilde{\chi}(\alpha, t-\tau) &= \exp[(\mathcal{L}_f + \mathcal{L}^0_\alpha)\tau] \chi(\alpha, t-\tau) , \\ \tilde{\rho}_1(\alpha, t-\tau) &= \exp(\mathcal{L}^0_\alpha \tau) \rho_1(\alpha, t-\tau) , \\ \tilde{R}(t-\tau) &= \exp(\mathcal{L}_f \tau) R(t-\tau) , \end{aligned} \tag{5.7}$$

and so forth. Moreover, we have defined

$$\mathcal{L}'_\alpha(-\tau) \equiv \exp[(\mathcal{L}_f + \mathcal{L}^0_\alpha)\tau] L'_\alpha \exp[-(\mathcal{L}_f + \mathcal{L}^0_\alpha)\tau] , \tag{5.8}$$

while $\tilde{\Delta}_t L'_\alpha$ is defined by Eq. (3.22) with the density operators in the partial averages replaced by their interaction-picture counterparts ($\chi \to \tilde{\chi}$ and $\rho_1 \to \tilde{\rho}_1$).

In addition to justifying the FBA, the condition $\epsilon \equiv \tau_{\text{int}}/\tau_{\text{rel}} \ll 1$ also permits us to introduce the Markoff approximation[33] on the right-hand side of Eq. (5.3). In the Markoff approximation we let $t \to \infty$ in the upper limit of the integral, and we replace the density operators $\tilde{\chi}(\alpha, t-\tau)$, $\tilde{R}(t-\tau)$, and $\tilde{\rho}_1(\alpha, t-\tau)$ by $\chi(\alpha, t)$, $R(t)$, and $\rho_1(\alpha, t)$, respectively. Since the shortest reservoir time constant is T_2, we have $\tau_{\text{int}} \cong T_2$, and we write phenomenologically

$$\langle \mathcal{L}'_\beta(-\tau) \rangle_{\text{res}} = e^{-\tau/T_2} L'_\beta(-\tau) , \tag{5.9}$$

where the τ dependence of L'_β is now due only to the system operator L^0. Consequently, with the Hamiltonian of Eq. (5.4), the τ integrations take the form of two definite integrals C and C^*, where[34]

$$C \equiv \int_0^\infty d\tau\, e^{-i(\omega_0-\Omega)\tau-\tau/T_2} = [T_2^{-1} + i(\omega_0 - \Omega)]^{-1} . \tag{5.10}$$

Purely for convenience in exposition, we will consider the field frequency Ω to be exactly resonant with the atomic frequency ω_0, in which case $C = C^* = T_2$. Hence, we obtain from Eq. (5.3) the Born-Markoff ME for a single-mode laser tuned to resonance,

$$\begin{aligned} &\dot{\chi}(\alpha, t) - (L_f + L_\alpha^0 + L'_\alpha)\chi(\alpha, t) \\ &\qquad - \sum_{\substack{\beta \\ (\beta\neq\alpha)}}{}' \langle L'_\beta\rangle_{\beta|f,t} R(t)\rho_1(\alpha, t) - \left(\frac{\delta\chi}{\delta t}\right)_{\text{res}} \\ &\quad = T_2 \sum_{\substack{\beta \\ (\beta\neq\alpha)}}{}' \mathrm{Tr}_\beta L'_\beta[\Delta_t L'_\beta \rho_1(\beta, t)\Delta\chi(\alpha, t) + (\alpha \rightarrow \beta)] . \end{aligned} \tag{5.11}$$

Correlations between two atoms and the field enter Eq. (5.11) in two ways: (a) through the next-to-last term on the left-hand side and (b) through the right-hand side. The former are *reducible* two-atom–plus–field correlations in the sense that they arise from the relevant part of the two-atom–plus–field density operator, that is from $F_{2,r}(\alpha, \beta, t)$, while the latter are *irreducible* correlations since they result from the irrelevant part $F_{2,i}(\alpha, \beta, t)$ of the density operator treated in the FBA. All such correlations are neglected from the beginning in the usual laser theories. In this section we wish to estimate the magnitude of these terms and determine the conditions under which they are negligible. We will assume throughout the current section that $\chi(\alpha)$ is diagonal in the unperturbed-energy $(H_f + H_\alpha^0)$ representation, so that

$$\langle\sigma_\alpha^\pm\rangle_t = \langle a^\pm\rangle_t = 0 ,$$

for example.

We begin by examining the irreducible correlations on the right-hand side of Eq. (5.11). Even though the right-hand side of the equation is rather complex, one may obtain a fairly accurate picture of its behavior with relative ease by examining moments. The simplest moments that measure atom-field correlations are $\langle a^+\sigma_\alpha^-\rangle$ and its conjugate $\langle\sigma_\alpha^+ a^-\rangle$. A better choice is to replace $\sigma_\alpha^\pm$ by the corresponding collective variables defined in Eq. (I4.5),

$$P_k^\pm \equiv \sum_\alpha e^{\pm i\vec{k}\cdot\vec{x}_\alpha}\sigma_\alpha^\pm , \tag{5.12}$$

representing the spatial Fourier component of the atomic polarization at the wave vector $\vec{k}$ corresponding to the field mode, and calculate the equations of motion of $\langle P_k^\pm a^\mp\rangle$. Even better is to write equations for the real and imaginary parts of $\mu\langle P_k^+ a^-\rangle$. The reason is that the real part is proportional to $\langle\mu P_k^+ a^- + \mu^* a^+ P_k^-\rangle = \hbar^{-1}\langle H'\rangle$, by Eqs. (3.1), (5.4c), and (5.12). Consequently, the contribution to the time derivative of the real part by the irreducible two-atom terms on the right-hand side of Eq. (5.11) vanishes, that is

$$\frac{d}{dt}\langle\mu P_k^+ a^- + \mu^* P_k^- a^+\rangle|_{2\text{-atom}} = 0 . \tag{5.13}$$

In Appendix D we calculate the time rate of change of the imaginary part of $\mu\langle P_k^+ a^-\rangle$ due to two-atom correlations. The result is

$$\begin{aligned} &\frac{d}{dt}\langle\mu P_k^+ a^- - \mu^* a^+ P_k^-\rangle|_{2\text{-atom}} \\ &\qquad = 2|\mu|^2(N-1)T_2\langle s\rangle\langle\mu P_k^+ a^- - \mu^* a^+ P_k^-\rangle , \end{aligned} \tag{5.14}$$

where

$$s \equiv N^{-1}\sum_\alpha s_\alpha \tag{5.15}$$

is the operator corresponding to the mean inversion per atom. Since the contribution to $\mathrm{Im}(\mu\langle P_k^+ a^-\rangle)$ from the single-atom terms on the left-hand side of the ME is dominated by the matter-reservoir term, we have

$$\begin{aligned} &\frac{d}{dt}\langle\mu P_k^+ a^- - \mu^* a^+ P_k^-\rangle|_{1\text{-atom}} \\ &\qquad \cong -T_2^{-1}\langle\mu P_k^+ a^- - \mu^* a^+ P_k^-\rangle . \end{aligned} \tag{5.16}$$

Hence, the effect of the irreducible two-atom correlation terms is to replace the dephasing rate T_2^{-1} by an effective reduced rate

$$T_{2,\text{eff}}^{-1} \equiv T_2^{-1}(1 - 2r) , \tag{5.17a}$$

$$r \equiv |\mu|^2 T_2^2(N-1)\langle s\rangle . \tag{5.17b}$$

We show in Appendix D that r is a small correction; in fact, upon substitution of the steady-state value of $\langle s\rangle$ into Eq. (5.17b), one obtains

$$r \cong T_2/2T_f , \tag{5.18}$$

where T_f is the decay time of the field *energy* due to the field reservoir. In typical gas lasers one has $T_2/T_f \sim 10^{-2}$ to 10^{-6}. The condition $T_2/T_f \ll 1$ has previously been noted by Kazantsev and Surdutovich[35] as the requirement for neglecting multiparticle effects in the laser.

In the following we will neglect the right side of the ME for $\dot{\chi}$, Eq. (5.11), and show that the left-hand side contains conventional single-mode laser theory, plus some corrections due to reducible two-atom–plus–field correlations. Thus we have

$$\begin{aligned} &\dot{\chi}(\alpha, t) - (L_f + L_\alpha^0 - K_m - K_f)\chi(\alpha, t) \\ &\qquad = L'_\alpha\chi(\alpha, t) + \sum_{\substack{\beta \\ (\beta\neq\alpha)}}{}' \langle L'_\beta\rangle_{\beta|f,t}\rho_1(\alpha, t)R(t) , \end{aligned} \tag{5.19}$$

where we have split $(\delta\chi/\delta t)_{\text{res}}$ into two parts, one $(-K_m)$ denoting the relaxation due to the matter reservoir and the other $(-K_f)$ relaxation due to the field reservoir:

$$(\delta\chi/\delta t)_{\text{res}} \equiv -K_m\chi - K_f\chi \ . \tag{5.20}$$

Taking the trace of Eq. (5.19) over the matter, we obtain the equation for the field density operator

$$\begin{aligned}\dot{R}(t) - (L_f - K_f)R(t) &= \sum_{\alpha} \langle L'_{\alpha}\rangle_{\alpha|f,t}R(t) \\ &= -i\mu \sum_{\alpha} e^{i\vec{k}\cdot\vec{x}_{\alpha}}[a^-, \mathcal{P}_{\alpha}^{+}(t)] + \text{H.c.} \ ,\end{aligned} \tag{5.21}$$

where in the last step we have used Eqs. (5.5) and (5.4c) and defined the two Hermitian-conjugate operators

$$\mathcal{P}_{\alpha}^{\pm}(t) \equiv \operatorname{Tr}_{\alpha}\sigma_{\alpha}^{\pm}\chi(\alpha, t) \equiv \langle\sigma_{\alpha}^{\pm}\rangle_{\alpha|f,t}R(t) \ , \tag{5.22}$$

In passing, we note that the same equation for $\dot{R}$ would have been obtained from Eq. (5.11) without neglecting the right-hand side and that Eq. (5.21) agrees with the result previously found in Eq. (3.28) with the exception of the added field-reservoir term.

Hence, to find the behavior of R, we need the dynamical equation of $\mathcal{P}_{\alpha}^{\pm}$. The equation for $\dot{\mathcal{P}}_{\alpha}^{+}$ is obtained easily by multiplying Eq. (5.19) by σ_{α}^{+} and tracing over α,

$$\begin{aligned}\dot{\mathcal{P}}_{\alpha}^{+}(t) - (i\omega_0 + L_f - T_2^{-1})\mathcal{P}_{\alpha}^{+}(t) \\ = -i\mu^* e^{-i\vec{k}\cdot\vec{x}_{\alpha}}[a^+\mathfrak{N}_{\alpha}^{+}(t) - \mathfrak{N}_{\alpha}^{-}(t)a^+] \ .\end{aligned} \tag{5.23}$$

We have neglected the term $\operatorname{Tr}_{\alpha}\sigma_{\alpha}^{+}K_f\chi(\alpha, t)$ because its magnitude is $T_f^{-1}\mathcal{P}_{\alpha}^{+}$ and $T_f^{-1} \ll T_2^{-1}$; in addition, we have defined

$$\mathfrak{N}_{\alpha}^{\pm}(t) \equiv \operatorname{Tr}_{\alpha}n_{\alpha}^{\pm}\chi(\alpha, t) \equiv \langle n_{\alpha}^{\pm}\rangle_{\alpha|f,t}R(t) \ , \tag{5.24a}$$

analogously to Eq. (5.22). In Eq. (5.24a) the operators for the upper and lower state occupation probabilities,

$$n_{\alpha}^{\pm} \equiv \sigma_{\alpha}^{\pm}\sigma_{\alpha}^{\mp} \ , \tag{5.24b}$$

have been introduced. We note for future use that

$$n_{\alpha}^{+} + n_{\alpha}^{-} = 1, \quad n_{\alpha}^{+} - n_{\alpha}^{-} = s_{\alpha} \tag{5.25a}$$

and

$$\mathfrak{N}_{\alpha}^{+}(t) + \mathfrak{N}_{\alpha}^{-}(t) = R(t) \ . \tag{5.25b}$$

We assume that T_2 is the shortest relaxation time in the laser,[36] $T_2 \ll T_1$, T_f, where T_1 is the relaxation time of the level populations and T_f is the relaxation time of the field energy due to leakage of photons out of the laser cavity. Then, since we already assumed $T_2 \ll \tau_{\text{rel}} \equiv (|\mu|^2NT_2)^{-1}$, Eq. (5.23) can be solved for $\mathcal{P}_{\alpha}^{+}(t)$ in terms of $\mathfrak{N}_{\alpha}^{\pm}$ at the same time t, and one can adiabatically eliminate the polarization from Eq. (5.21), that is

$$\mathcal{P}_{\alpha}^{+}(t) = -i\mu^* T_2 e^{i\vec{k}\cdot\vec{x}_{\alpha}}[a^+\mathfrak{N}_{\alpha}^{+}(t) - \mathfrak{N}_{\alpha}^{-}(t)a^+] \ , \tag{5.26}$$

with a similar equation for $\mathcal{P}_{\alpha}^{-}$, and

$$\begin{aligned}\dot{R}(t) - (L_f - K_f)R(t) \\ = -|\mu|^2T_2 \sum_{\alpha}[a^-, a^+\mathfrak{N}_{\alpha}^{+}(t) - \mathfrak{N}_{\alpha}^{-}(t)a^+] + \text{H.c.}\end{aligned} \tag{5.27}$$

To proceed with the determination of the equation of motion of R, we need the equation for $\dot{\mathfrak{N}}_{\alpha}^{+}(t)$, which is obtained by multiplying Eq. (5.19) by n_{α}^{+} and tracing over α, with the result

$$\begin{aligned}\dot{\mathfrak{N}}_{\alpha}^{+}(t) - (L_f - K_f)\mathfrak{N}_{\alpha}^{+}(t) + T_1^{-1}[\mathfrak{N}_{\alpha}^{+}(t) - R(t)] \\ = \operatorname{Tr}_{\alpha}n_{\alpha}^{+}L'_{\alpha}\chi(\alpha, t) + \langle n_{\alpha}^{+}\rangle_t {\sum_{\beta}}' \operatorname{Tr}_{\beta}L'_{\beta}\chi(\beta, t) \ .\end{aligned} \tag{5.28}$$

We have written

$$\operatorname{Tr}_{\alpha}n_{\alpha}^{+}K_m\chi(\alpha, t) = T_1^{-1}[\mathfrak{N}_{\alpha}^{+}(t) - R(t)] \ , \tag{5.29}$$

implying that the matter reservoir includes a pump term which tends to drive the upper-state occupation probability $\langle n^+\rangle$ to unity,[37] in the absence of coupling to the field, at a rate T_1^{-1}. The last term on the right-hand side of Eq. (5.28) represents the effect of reducible atom-field correlations on $\dot{\mathfrak{N}}_{\alpha}^{+}$. This term assures us of obtaining the correct $\dot{R}$ equation, Eq. (5.27), when the equation for $\dot{\mathfrak{N}}_{\alpha}^{-}$ is added to Eq. (5.28) for $\dot{\mathfrak{N}}_{\alpha}^{+}$ and Eq. (5.25b) is used. Previous laser theories[16-18] are obtained by neglecting this two-atom term, plus the field-reservoir term. It is convenient to rewrite Eq. (5.28) using the conditional-average notation of Eqs. (5.24a) and (5.5), yielding

$$\begin{aligned}\left(\frac{d}{dt} - L_f + K_f\right)\langle n_{\alpha}^{+}\rangle_{\alpha|f,t}R(t) + T_1^{-1}(\langle n_{\alpha}^{+}\rangle_{\alpha|f,t} - 1)R(t) \\ = \langle n_{\alpha}^{+}L'_{\alpha}\rangle_{\alpha|f,t}R(t) + \langle n_{\alpha}^{+}\rangle_t {\sum_{\beta}}' \langle L'_{\beta}\rangle_{\beta|f,t}R(t) \ .\end{aligned} \tag{5.30}$$

At this point, there are two possibilities: (a) The atomic populations can be adiabatically eliminated, like the polarization. This is possible if $T_1 \ll \tau_{\text{rel}}$, T_f. (b) The atomic populations cannot be adiabatically eliminated since $T_1 \gtrsim \tau_{\text{rel}}$. In the latter case, we must solve the coupled dynamical equations for $\dot{R}$ and $\dot{\mathfrak{N}}_{\alpha}^{\pm}$, Eqs. (5.27) and (5.28) or (5.30), and one will find population pulsations. We will not consider this case further. On the other hand, in case (a) we will see that $\mathfrak{N}_{\alpha}^{\pm}$ can be eliminated from Eq. (5.27) leaving a single ME for $\dot{R}(t)$.

Before obtaining the adiabatic solution for the population, we will eliminate the sum over β from Eq. (5.30) by subtracting from it the equation of motion of $\langle n_{\alpha}^{+}\rangle R$. To obtain the latter we need the equation for $\dot{R}$ [Eq. (5.27)] and the equation of motion for $\langle n_\alpha^+\rangle$, obtained by tracing Eq. (5.30) or (5.28) over the field

$$\frac{d}{dt}\langle n_\alpha^+\rangle_t + T_1^{-1}(\langle n_\alpha^+\rangle_t - 1) = \langle n_\alpha^+ L'_\alpha\rangle_t \ . \tag{5.31}$$

Combining Eqs. (5.30), (5.31), and (5.21), we obtain

$$\left(\frac{d}{dt} - L_f + K_f + T_1^{-1}\right)[(\langle n_\alpha^+\rangle_{\alpha|f} - \langle n_\alpha^+\rangle)R] = (\langle \Delta n_\alpha^+ L'_\alpha\rangle_{\alpha|f} - \langle n_\alpha^+ L'_\alpha\rangle)R \ . \tag{5.32}$$

We have dropped the time argument where there is no possible ambiguity. Equations (5.31) and (5.32) are still quite general. In the adiabatic case, $T_1 \ll \tau_{\text{rel}}, T_f$, we can set

$$d\langle n_\alpha^+\rangle/dt \cong 0$$

in Eq. (5.31) and

$$\left(\frac{d}{dt} - L_f\right)[(\langle n_\alpha^+\rangle_{\alpha|f} - \langle n_\alpha^+\rangle)R] \cong 0$$

in Eq. (5.32). The first condition enables us to write

$$T_1^{-1}(\langle n_\alpha^+\rangle - 1) \cong \langle n_\alpha^+ L'_\alpha\rangle \ ,$$

which combines with the second condition to yield the result

$$T_1^{-1}(\langle n_\alpha^+\rangle_{\alpha|f} - 1)R + K_f[(\langle n_\alpha^+\rangle_{\alpha|f} - \langle n_\alpha^+\rangle)R] = \langle \Delta n_\alpha^+ L'_\alpha\rangle_{\alpha|f} R. \tag{5.33}$$

Since $T_1 \ll T_f$, we may ignore the field-reservoir term in Eq. (5.33) in comparison with the matter-reservoir term, and the equation becomes especially simple. We may write

$$\mathfrak{N}_\alpha^+ - R = -2|\mu|^2 T_1 T_2[a^- a^+ \mathfrak{N}_\alpha^+ - a^- \mathfrak{N}_\alpha^- a^+ + \langle n_\alpha^+\rangle(\mathfrak{N}_\alpha^- + a^+\mathfrak{N}_\alpha^+ a^- + a^-\mathfrak{N}_\alpha^- a^+ - a^- a^+ R)], \tag{5.34}$$

where we have eliminated the polarization by Eq. (5.26) and used the commutation rule for a^- and a^+ and Eq. (5.25b). We have taken the diagonality of χ into consideration in the form

$$[a^- a^+, \chi] = 0.$$

Through the use of the definition of the commutator, in conjunction with Eq. (5.25b), we can easily rewrite Eq. (5.34) in the form

$$\mathfrak{N}_\alpha^+ - R = -\frac{1}{2n_{\text{sat}}}\{a^- a^+(\mathfrak{N}_\alpha^+ - \mathfrak{N}_\alpha^-) + a^-[a^+, \mathfrak{N}_\alpha^-] - \langle n_\alpha^+\rangle \times (\mathfrak{N}_\alpha^+ - \mathfrak{N}_\alpha^- + a^+[a^-, \mathfrak{N}_\alpha^+] + a^-[a^+, \mathfrak{N}_\alpha^-])\}, \tag{5.35}$$

where

$$n_{\text{sat}} \equiv [4|\mu|^2 T_1 T_2]^{-1} \tag{5.36}$$

is the photon number at which the gain, or population inversion, saturates.[38]

It is clear from this expression that $\mathfrak{N}_\alpha^+$ and $\langle n_\alpha^+\rangle \equiv \text{Tr}_f \mathfrak{N}_\alpha^+$ are independent of α. This is true because the atoms are interacting with a single-mode traveling-wave field in the rotating-wave approximation [see Eq. (5.4c)]. The lack of dependence of the inversion of the atom on its position could also have been predicted earlier; it is a direct consequence of the cancellation of the mode factors $\exp(\pm i\vec{k}\cdot\vec{X}_\alpha)$ when the polarization is eliminated from Eqs. (5.21) and (5.33) using Eq. (5.26) for $\mathcal{P}_\alpha^+$. In the future we will drop the label α and write $\mathfrak{N}^+$, $\langle n^+\rangle$, $\mathfrak{N}^- \equiv R - \mathfrak{N}^+$, and $\langle n^-\rangle \equiv 1 - \langle n^+\rangle$. One may now also perform the summation over α in the kinetic equation for R, Eq. (5.27), and write

$$\mathring{R}(t) - (L_f - K_f)R = -|\mu|^2 N T_2[a^-, a^+\mathfrak{N}^+ - \mathfrak{N}^- a^+] + \text{H.c.} \tag{5.37}$$

At this point one solves Eq. (5.35) for $\mathfrak{N}^+$ in terms of R, obtains $\mathfrak{N}^-$ from Eq. (5.25b), and substitutes the results into Eq. (5.37) to obtain the equation of motion of the radiation density matrix. However, before proceeding it is useful to make a few remarks about the nonlinear terms involving $\langle n^+\rangle$ in Eq. (5.35), which we will henceforth call simply the "nonlinear terms." The nonlinear terms in Eq. (5.35) arose from the reducible two-atom correlation term in the ME, that is they appeared when the last term in Eq. (5.28) was eliminated in passing to Eq. (5.32). The usual practice[16,17] is to neglect irreducible two-body terms of the form $\langle\sigma_\alpha^+\sigma_\beta^-\rangle$, since we have shown that they are of order T_2/T_f and thus are frequently negligible. The nonlinear two-body terms due to reducible two-atom correlations are also usually neglected. We will show that although small, they are not strictly negligible. In Appendix E we show the terms multiplying $\langle n^+\rangle$ in Eq. (5.35) are of order unity. Hence, they make a contribution of the same order as the incoherent spontaneous emission term in Eq. (5.35), which arises from the one in $a^-a^+ = a^+a^- + 1$.

In order to retain the main effect of the nonlinearity without the extra complication of a nonlinear term, we may simply set $\langle n^+\rangle = 1$ in Eq. (5.35) for the following reason: The magnitude of the spontaneous-emission term, and hence of the nonlinear term, relative to stimulated emission is n^{-1}, where n is the number of photons in the field. At threshold we have[39] $n \sim n_{\text{sat}}^{1/2} \cong 10^3$ (for a typical laser $n_{\text{sat}} \cong 10^6$), so that spontaneous emission is a correction of 0.1%. Above threshold, spontaneous emission becomes even less important and eventually becomes negligible. However, at threshold $\langle n^+\rangle \equiv 1 - \langle n^-\rangle$ can be approximated by one, since it follows from Eqs. (5.25a), (D10), and (5.36) that $\langle n^-\rangle \sim n/n_{\text{sat}}$, which is approximately 10^{-3} at threshold. Hence, the nonlinear term can be well represented by replacing $\langle n^+\rangle$ by one in the threshold region, which is the only region where it is appreciable.

When we set $\langle n^+\rangle$ equal to one in Eq. (5.35) we obtain

$$\mathcal{N}^+ - R = -(2n_{\text{sat}})^{-1}\{a^+a^-(\mathcal{N}^+ - \mathcal{N}^-) + a^+[a^-, \mathcal{N}^+]\}. \quad (5.38)$$

Using Eq. (5.25b) and the operator identity

$$a^+[a^-, \mathcal{N}^+] \equiv a^+ \frac{\partial}{\partial a^+} \mathcal{N}^+, \quad (5.39)$$

we can formally solve Eq. (5.38) for $\mathcal{N}^+$,

$$\mathcal{N}^+ = \left(1 + \frac{a^+a^-}{n_{\text{sat}}} - \frac{a^+}{2n_{\text{sat}}}\frac{\partial}{\partial a^+}\right)^{-1}\left(1 + \frac{a^+a^-}{2n_{\text{sat}}}\right)R. \quad (5.40)$$

We show below that, if one neglects the commutators and nonlinear terms in Eq. (5.35), the result is the conventional laser theory.[16-18] However, if one requires a theory that treats spontaneous emission rigorously near threshold, then it is necessary to retain Eq. (5.40) without approximation. Furthermore, if a rigorous theory of spontaneous emission for $n \sim n_{\text{sat}}$ is desired, then it is necessary to retain the full Eq. (5.35) including the nonlinear terms.

To show that neglecting commutators and nonlinear terms in Eq. (5.35) leads to a differential equation for $\mathring{R}$ which is identical to the ME of the usual laser theory, we take matrix elements in the photon-number representation and define[17]

$$R_n \equiv \langle n|R|n\rangle, \quad F_\pm(n) \equiv \langle n|\mathcal{N}^\pm|n\rangle. \quad (5.41)$$

The result is

$$\begin{aligned}\mathring{R}_n(t) + \langle n|K_f R(t)|n\rangle \\ = 2|\mu|^2NT_2\{(n+1)[F_-(n+1,t) - F_+(n,t)] \\ + n[F_+(n-1,t) - F_-(n,t)]\}, \quad (5.42\text{a})\end{aligned}$$

which agrees with Eq. (3.10c) and Eq. (4.1b) of Ref. 17 for the case of zero detuning currently under consideration.[40] Similarly, neglecting $\langle n^+\rangle$ and the commutator in Eq. (5.35) and taking matrix elements one obtains

$$F_+(n,t) = \frac{1 + (2n_{\text{sat}})^{-1}(n+1)}{1 + n_{\text{sat}}{}^{-1}(n+1)} R_n(t), \quad (5.42\text{b})$$

which agrees with Eq. (4.3), Ref. 17, if $n_0^+ \equiv \rho^0(+) = 1$.

Risken[16] has shown there are corrections to the usual laser theory at threshold of order $n/n_{\text{sat}} \sim 10^{-3}$, represented by the term $a^-[a^+, \mathcal{N}^-]$ in Eq. (5.35). On the other hand, we have shown that, due to the nonlinear terms in Eq. (5.35), there is an even larger contribution, which is of order one near threshold where $\langle n^+\rangle \approx 1$ and the commutator is of order one. This latter correction is of the same magnitude as ordinary spontaneous emission.

We now summarize the conditions required for the validity of Eqs. (5.35) and (5.37). First, we require $\tau_{\text{int}} \equiv T_2 \ll \tau_{\text{rel}} \equiv (|\mu|^2NT_2)^{-1}$ for the validity of the FBA and the Markoff approximation. Secondly, we require $T_2 \ll T_f$ in order to neglect the irreducible two-atom-plus-field correlations represented by the right-hand side of Eq. (5.11). Finally, we require $T_2, T_1 \ll T_f, \tau_{\text{rel}}$ in order to eliminate the matter adiabatically from the ME for the field density operator, Eq. (5.21), and to neglect the time development of $\mathcal{N}^\pm$ due to the field reservoir compared with that due to the matter reservoir.

The power of the TDPO technique can be appreciated by the relative ease with which the results of this section were obtained. Using the TDPO of Eq. (3.7), which projects the system density operator onto states having correlations between one atom and the field, we have shown that the usual laser theory is obtained by ignoring all but single-atom-plus-field correlations in the ME for $\chi(\alpha, t)$. In addition, we have determined the magnitude of corrections to the theory due to two-atom-plus-field correlations. The present treatment should be contrasted with that in a recent paper[19] using a time-independent PO without atom-field correlations, namely,[41]

$$P = A\,\mathrm{Tr}_m, \quad (5.43\text{a})$$

$$A \equiv \prod_\alpha^N [\tfrac{1}{2}(1+\sigma_0)n_\alpha^+ + \tfrac{1}{2}(1-\sigma_0)n_\alpha^-], \quad (5.43\text{b})$$

where σ_0 is a constant satisfying $0 \le \sigma_0 \le 1$. With this PO, it was necessary to perform an infinite summation of terms of all orders in μ instead of just the term of order $|\mu|^2$. This led to the difficulty that the general solution diverged; more precisely, the term containing $|\mu|^{2p}$ diverged as N^p. The proper procedure in this case, according to Ref. 19, would have been to reorder the summations in such a way as to lead to a convergent series but the labor involved was too great. Therefore, the author showed that summation of the most divergent terms at each order in μ gave rise to a convergent result. By contrast, with the proper choice of PO, namely, a TDPO containing atom-field correlations, we have made it unnecessary to deal with infinite-order terms and have avoided the need to perform infinite resummations.

VI. CONCLUSION

We will conclude by discussing the results obtained in Secs. I–V using TDPO techniques for systems with correlations, then mention some possible generalization of the methods of this paper, and finally discuss extensions of the TDPO technique to other problems in kinetic theory and quantum optics.

A. Discussion

In this paper we have extended the domain of applicability of the TDPO method to include systems involving correlations between two particles and between one particle and field variables. Our equations represent an exact transformation of the Liouville equation, expressed entirely in terms of the single- and two-body distribution functions in the particle-particle interaction case and in terms of the single-atom, field, and atom-plus-field density operators in the atom-field interaction case.

The equations of our model include nonlinearities on two levels. We will illustrate this in terms of the molecular-kinetics problem, but our remarks apply equally well to the quantum-optical case. First, our propagators $\mathcal{G}(t,t')$ depend on the TDPO $P(t)$, which in turn depends on the one- and two-body distribution functions. Consequently, we have a transcendental nonlinear dependence on the one- and two-body distribution functions. Second, since our equations for $\mathring{F}_1$ and $\mathring{F}_2$ are coupled, the elimination of either one in terms of the other leads to a nonlinear dependence.

The way nonlinearity enters the TDPO approach should be contrasted with the way it enters Mori's exact Langevin equations[42] using time-independent PO's. In the original Mori treatment averages over an equilibrium ensemble demand that the equations be explicitly linear in the Langevin variable. The Mori approach has been generalized to include nonlinearities in three recent publications.[43-45] However, the nonlinearity of the time-independent PO's in the latter publications is different from the nonlinearity of our TDPO with correlations, which arises from projecting the full distribution function onto suitable products of one- and two-body distribution functions. Since the full time-development operator causes the distribution function to change in time, the application of our PO at two different times constitutes a different projection. Our introduction of nonlinearity thus becomes time dependent because we are representing the linear time development of the Liouville operator in the N-molecule phase space or N-atom–plus–field Hilbert space exactly in terms of the time development of one- and two-particle distribution functions.

Of course, since all generalized ME's are exact, the advantage of any new generalized ME, whether obtained from time-independent or time-dependent PO's is to provide alternate starting points for making approximations. In I we showed that TDPO's projecting onto uncorrelated states are especially useful for problems in which self-consistent-field behavior is important and for Boltzmann-like problems where there are strong interactions between dilute systems. In the present paper we have shown that TDPO's are also useful in other situations, where the TDPO's of I do not result in a good low-order description of the physical phenomena. These situations include those where correlations are important due to the fact that one is dealing with strongly interacting systems such as a laser or a dense gas. The use of the TDPO with correlations in describing such systems effectively incorporates infinite resummations already in lowest order.

One can conceive of various generalizations of the TDPO techniques developed in this paper. However, one must pay the price of increased complexity and they offer little advantage in the current problems. For the sake of completeness, we mention two of these possible generalizations. First, the generalization to include three-body and higher-order correlations, or two-atom–plus–field and higher-order correlations in the quantum-optical case, directly in $F_{N,r}$ is completely straightforward. However, this process is probably too laborious to justify the effort. Secondly, the forms of $F_{N,r}$ used in Eqs. (2.2) and (3.3) can be generalized to include terms higher than first order in ΔF_2 and $\Delta\chi$, respectively. We refer to such forms of $F_{N,r}$ as nonlinear F_r's and discuss them briefly.

In the quantum-optical case, the most general nonlinear F_r containing correlations between the field and only a single atom at a time is[46,6]

$$F_{N,r}(1,\ldots,N,t)=R(t)\prod_{\alpha}\chi(\alpha\,|f,t). \tag{6.1}$$

One can check that this expression satisfies Eqs. (3.4) and (3.6); that is, it is normalized and tracing over the coordinates of an atom yields the relevant part of the density operator for $N-1$ atoms and the field. In addition, it is clear that the density operator of Eq. (3.3) results when one linearizes Eq. (6.1) in $\Delta\chi(\alpha,t)$. The TDPO associated with the F_r of Eq. (6.1) is[6]

$$P(t)=\sum_{\alpha}\prod_{\beta}{}'\,\chi(\beta\,|f,t)\,\mathrm{Tr}_{\beta}$$

$$-(N-1)\prod_{\alpha}\chi(\alpha\,|f,t)\,\mathrm{Tr}_{m}. \tag{6.2}$$

It can be verified directly that this is the desired PO by showing that Eqs. (6.1) and (6.2) satisfy the conditions in Eqs. (1.9a), (1.10a), and (1.13b). Moreover, the TDPO of Eq. (3.7) for the linearized F_r can be obtained by carefully linearizing Eq. (6.2). One can proceed with the derivation of the ME's as in Sec. III, but the results are quite complicated.

In the gas-kinetic case, the generalization to a nonlinear F_r is not straightforward.[6] One can attempt to obtain a nonlinear generalization of Eq. (2.2) or (2.2′) by using a form of the Kirkwood-superposition approximation[47] for $F_{N,r}$. For example, one could choose the form of Percus[48] where every molecule is linked with every other molecule in an F_2,

$$F_{N,r}(1,\ldots,N,t)=\prod_{\alpha} F_1(\alpha,t) \prod_{\substack{\beta \\ (\beta<\alpha)}} \frac{F_2(\alpha,\beta,t)}{F_1(\alpha,t)F_1(\beta,t)}. \tag{6.3}$$

However, there are well-known problems[49] associated with this class of expressions, such as the inability to prove that they are normalized (in some cases) or to prove that $V^{-1}\int dNF_{N,r}=F_{N-1,r}$ [Eq. (2.3)]. In our opinion, these difficulties preclude, or at least make unattractive, the use of any conceivable nonlinear F_r as a basis for deriving generalized ME's in the gas-kinetic case.

B. Impurity-atom line-shape problem

Many of the new methods developed in many-body physics and nonequilibrium statistical mechanics have been successfully applied to the problem of predicting the line-shape function for the radiation from an impurity atom in a fluid.[50] In particular, time-independent PO methods are frequently employed[51] to obtain the time-correlation functions that appear in the expression for the line shape. The new methods in question are usually based on linear response theory, that is they require only small deviations from equilibrium. The methods involving TDPO's developed in this paper can be used to study the line-broadening problem for situations where the deviations from equilibrium are *not* small.

It is beyond the scope of the present paper to derive expressions for the line shape of a radiating atom in a fluid using the TDPO formalism. However, we will give an indication of how such a theory might be developed. We wish to calculate the spectral density of dipole radiation from an atom in a surrounding fluid whose molecules act as perturbers. The spectral density is proportional to the line-shape function[52]

$$I(\omega)=\pi^{-1}\,\mathrm{Re}\int_0^\infty dt\, e^{i\omega t}\langle\vec{\mathrm{d}}\cdot\vec{\mathrm{d}}(t)\rangle, \tag{6.4}$$

where $\langle\vec{\mathrm{d}}\cdot\vec{\mathrm{d}}(t)\rangle\equiv\mathrm{Tr}\,\vec{\mathrm{d}}\cdot\vec{\mathrm{d}}(t)F_N$, $\vec{\mathrm{d}}$ is the dipole moment of the atom, $F_N\equiv F_N(a,1,\ldots,N,t)$ is the density operator of the radiating atom (degrees of freedom represented by a) and the N bath atoms (degrees of freedom $\alpha=1,\ldots,N$), and the trace is over all degrees of freedom of the radiating atom and the perturber atoms. The time dependence of $\vec{\mathrm{d}}(t)$ is due to the full system Hamiltonian, and we have arbitrarily treated the entire system as quantum mechanical. The relevant part of the density operator is chosen to be

$$\begin{aligned}F_{N,r}(t)=\sum_{\alpha}^{N} F_1(a,\alpha,t)\prod_{\beta}^{N}{}'\rho_1(\beta,t)\\ +f(a,t)\left\{\frac{1}{2}\sum_{\alpha,\beta}^{N}{}'\rho_2(\alpha,\beta,t)\prod_{\gamma}^{N}{}''\rho_1(\gamma,t)\right.\\ \left.-\left[\binom{N}{2}+N-1\right]\prod_{\alpha}^{N}\rho_1(\alpha,t)\right\},\end{aligned} \tag{6.5}$$

where $F_j(a,1,\ldots,j,t)$ is the density operator for the radiating atom and j perturber atoms, $\rho_j(1,\ldots,j,t)\equiv\mathrm{Tr}_aF_j$ is the density operator for j perturber atoms, and $f(a,t)\equiv\mathrm{Tr}_\alpha F_1(a,\alpha,t)$ is the density operator of the radiating atom. The form of $F_{N,r}$ in Eq. (6.5) contains both impurity-perturber and perturber-perturber correlations. As such, it represents a combination of the two problems we have treated in this paper, the dense gas of Sec. II, where $F_{N,r}$ is given by Eq. (2.2′), and the quantum-optical system of Sec. III, where $F_{N,r}$ is given by Eq. (3.3a). The impurity atom in the current problem plays the same role as the electromagnetic field does for the quantum-optical problem.

When we apply the theory of the present paper to the line-shape problem, we find an exact generalized ME, which is equivalent to a set of four closed coupled equations of motion for the quantities $\rho_1(\alpha,t)$, $f(a,t)$, $\rho_2(\alpha,\beta,t)$, and $F_1(a,\alpha,t)$. The advantages to be gained by the present formulation of the line-broadening problem are (i) the ability to include nonlinear effects, (ii) formally exact equations of motion for the four important distribution functions of the problem ρ_1, f, ρ_2, and F_1, and (iii) the relative ease of obtaining approximate solutions which are not perturbative in a small parameter. One may obtain approximations which include resummations of infinite subclasses of diagrams by expanding the coupled equations in a parameter such as the density, expressing ρ_1, ρ_2, and $\mathring{F}_1$ in terms of $f(a)$, and eliminating them from the $\mathring{f}(a)$ equation. The resultant equation for $f(a)$ will be transcendental in the density, just as the equation for $\mathring{F}_1(\alpha,t)$ was in the gas-kinetic case of Sec. IV when mean-free-path effects were included by eliminating $F_2(\alpha,\beta,t)$.

C. Quantum-optical problems: Amplified spontaneous emission and superradiance

There are several additional applications of TDPO methods involving projection onto correlated states in quantum optics. In this section we will mention applications to two processes which occur in high-gain laser media, namely, amplified spontaneous emission (ASE) and superradiance.

In the ASE problem[53-57] we study the output radiation from an inverted medium driven by spontaneous emission. The medium is continuously pumped, and the inversion density is sufficiently small that many-atom effects are negligible. The situation in ASE differs from the usual laser situation in that the absence of feedback arising from placing the active atoms in a resonator allows the mode structure to be continuous instead of discrete. Consequently, there is no sharp threshold as in a laser where the character of the emission changes from chaotic to ordered as the pump rate is increased. Nevertheless, nonlinear electromagnetic effects are still important. The atom-field correlations created by the interaction cause the radiation statistics to change from Gaussian to non-Gaussian,[53-55] and situations are possible where the radiation becomes quite coherent. In a future publication we will derive ME's for the ASE problem using the methods of Secs. III and V. The only fundamental change in the derivation is that it is necessary to include a continuum of modes instead of the discrete modes or single mode of laser theory. The solution of the resultant equation for χ will enable us to express the correlations between atoms and radiation that change the character of the radiation statistics as the length of the inverted medium is increased.

The problem of superradiance differs from that of ASE in that the initial inversion is much larger, in fact virtually complete, and the density of excited atoms is so high that collective effects arising from the $\vec{j}\cdot\vec{A}$ interaction lead to radiation rates proportional to N^2. There are two distinct regimes of interest in the superradiance problem. In weak superradiance[58] the shortest interaction time in the problem, the radiation dissipation time T_f, is sufficiently short to serve as the interaction time to justify the FBA and the Markoff approximation. In this parameter regime cooperative effects $\sim N^2$ are small compared to dissipative effects, and one obtains the usual superradiance ME[58] for $\mathring{F}_1$ in which the interaction induces atom-atom correlations starting from uncorrelated atom-field states. In the regime of strong superradiance,[59] on the other hand, the $\vec{j}\circ\vec{A}$ interaction is stronger than dissipative effects, and neither naive perturbation theory nor the Markoff approximation remain valid. Propagation effects become important, and the atoms and the field enter the problem on a more equal footing.[30] Neither matter variables, as in the laser, nor field variables, as in weak superradiance, may be eliminated adiabatically. Elsewhere we will show that the methods of Sec. III lead to a natural description of strong superradiance in terms of the joint atom-field density operator $\chi(\alpha,t)$. In this description atom-field correlations brought about by the $\vec{j}\circ\vec{A}$ interaction are fundamental entities while atom-atom correlations are derived ones. The atom-atom correlations responsible for superradiant emission arise indirectly from the fact that many atoms are correlated to the same field.

APPENDIX A: PROOF THAT EQ. (2.13) IS A PROJECTION OPERATOR; PROOF OF EQ. (2.20)

The purpose here is to show that the operator of Eq. (2.13) is the desired TDPO for the molecular system with two-body correlations by showing how one proves that it satisfies Eq. (1.13b). Making use of Eqs. (2.9) and (2.10) for $l=5$, it is sufficient to show that

$$p_N^{(i)}(t)P_N(t')\equiv p_N^{(i)}(t),\quad i=1,\ldots,5, \tag{A1}$$

where from Eqs. (2.13) and (I3.11) we have defined

$$p_N^{(1)}(t)\equiv\frac{1}{2}\sum_{\alpha,\beta}{}'\prod_{\kappa}{}''F_1(\kappa,t)\int\frac{d\kappa}{V}, \tag{A2a}$$

$$p_N^{(2)}(t)\equiv\frac{1}{2}\sum_{\alpha,\beta}{}'F_2(\alpha,\beta,t)\times\int\frac{d\alpha}{V}\int\frac{d\beta}{V}\sum_{\gamma}{}''\prod_{\kappa}{}'''F_1(\kappa,t)\int\frac{d\kappa}{V}, \tag{A2b}$$

$$p_N^{(3)}(t)\equiv-\tfrac{1}{2}(N-2)\sum_{\alpha,\beta}{}'F_2(\alpha,\beta,t)\prod_{\kappa}{}''F_1(\kappa,t)\int d\Gamma_N, \tag{A2c}$$

$$p_N^{(4)}(t)\equiv-\left[\binom{N}{2}-1\right]\sum_{\alpha}\prod_{\kappa}{}'F_1(\kappa,t)\int\frac{d\kappa}{V}, \tag{A2d}$$

$$p_N^{(5)}(t)\equiv(N-1)\left[\binom{N}{2}-1\right]\prod_{\kappa}F_1(\kappa,t)\int d\Gamma_N. \tag{A2e}$$

The main difficulty in proving Eq. (A1) is in listing and counting the various types of terms arising on the left-hand side and their contribution to the right-hand side. Different types of terms arise depending on the relation between the summation indices in the first and second factors of the left-hand side of Eq. (A1). We standardize the notation for the dummy indices involved and denote the dummy indices in the second factor of the product, $P_N(t')$

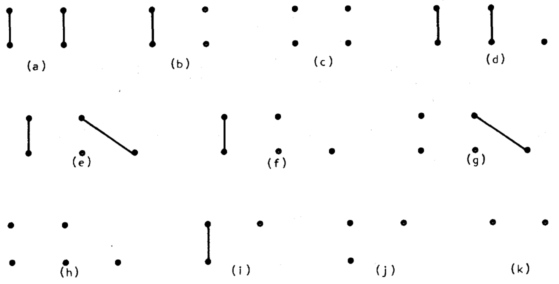

FIG. 1. Graphs associated with Table I arising in the evaluation of Eq. (A1) for $i = 1$.

$=\sum_j p_N^{(j)}(t')$, by making the replacements $(t;\alpha,\beta,\gamma,\kappa) \to (t';\delta,\epsilon,\zeta,\lambda)$ in Eqs. (A2). A graphical representation is extremely helpful; so we will denote the summation indices α,β,γ in the first factor by a superior row of dots and those in the second factor δ,ϵ,ζ by an inferior row of dots. Equality of two indices will be denoted by a line connecting the corresponding dots (see Fig. 1). The symmetry of the $p_N^{(i)}$ under interchange of α and β and under interchange of δ and ϵ enables us to reduce the number of distinct graphs by assigning weights to them. For example, a graph with $\alpha = \delta$ and no other indices equal [Fig. 1(b), for example] has a weight 4, since it contributes the same amount as graphs with $\alpha = \epsilon$, $\beta = \delta$, or $\beta = \epsilon$.

The various contributing terms for $i = 1$ and 2 in Eq. (A1) are listed in Tables I and II, respectively, along with their weights. The product $p_N^{(i)}(t)p_N^{(j)}(t')$ is represented by the couple $(i-j)$. The products are listed in column 1 of the tables along with the specific configuration of indices being considered. Data on the various terms associated with each product $(i-j)$ are listed in successive columns. The graphs corresponding to Table I are drawn in Figs. 1(a)-1(k) and those associated with Table II in Figs. 2(a) - 2(u). The letter in column 2 refers to the figure listing the graph for that term. Only four types of terms result from the products in Table 1, and they are abbreviated

$$A \equiv \frac{1}{4} \sum_{\alpha,\beta}{}' \prod_{\kappa}{}'' F_1(\kappa,t) \int \frac{d\kappa}{V} , \tag{A3a}$$

TABLE I. Terms contributing to $p_N^{(1)}(t)P_N(t')$, along with their representative graphs and their weights.

Class [a]	Graphs [b]	Weights	Terms [c]
$(1-1)$ $\alpha \longleftrightarrow \beta$	(a)	2	A
$\bullet \quad \bullet$	(b)	4	$(N-2)B$
$\bullet \quad \bullet$	(c)	1	$(N-2)(N-3)C$
$\delta \longleftrightarrow \epsilon$			
$(1-2)$ $\alpha \longleftrightarrow \beta$	(d)	2	$(N-2)D$
$\bullet \quad \bullet$	(e)	4	$(N-2)B$
$\bullet \quad \bullet \quad \bullet$	(f)	4	$(N-2)(N-3)C$
$\delta \longleftrightarrow \epsilon \quad \zeta$	(g)	2	$(N-2)(N-3)B$
	(h)	1	$(N-2)(N-3)(N-4)C$
$(1-3)$ $\alpha \longleftrightarrow \beta$	(a)	2	$-(N-2)D$
$\bullet \quad \bullet$	(b)	4	$-(N-2)^2C$
$\bullet \quad \bullet$	(c)	1	$-(N-2)^2(N-3)C$
$\delta \longleftrightarrow \epsilon$			
$(1-4)$ $\alpha \longleftrightarrow \beta$	(i)	2	$-2f_NB$
$\bullet \quad \bullet$	(j)	1	$-2(N-2)f_NC$
$\bullet$			
δ			
$(1-5)$ $\alpha \longleftrightarrow \beta$	(k)	1	$2(N-1)f_NC$
$\bullet \quad \bullet$			

[a] We define $(i-j) \equiv p_N^{(i)}(t)p_N^{(j)}(t')$. The first row of each pair below denotes particle summation indices in $p_N^{(i)}(t)$ and the second row indices in $p_N^{(j)}(t')$. Symmetric pairs of indices are connected by an arrow.

[b] The letters refer to labels in Fig. 1.

[c] The expressions A, B, C, and D are defined in Eqs. (A3) and $f_N \equiv \binom{N}{2} - 1$.

TABLE II. Terms contributing to $p_N^{(2)}(t)P_N(t')$, along with their representative graphs and their weights.

Class [a]	Graphs [b]	Weights	Terms [c]
$(2-1)$ $\alpha \longleftrightarrow \beta \quad \gamma$	(a)	2	F
$\bullet \quad \bullet \quad \bullet$	(b)	4	E
$\bullet \quad \bullet$	(c)	4	$(N-3)F$
$\delta \longleftrightarrow \epsilon$	(d)	2	$(N-3)E$
	(e)	1	$(N-3)(N-4)F$
$(2-2)$ $\alpha \longleftrightarrow \beta \quad \gamma$	(f)	2	E
$\bullet \quad \bullet \quad \bullet$	(g)	2	$(N-3)F$
$\bullet \quad \bullet \quad \bullet$	(h)	4	F
$\delta \longleftrightarrow \epsilon \quad \gamma$	(i)	4	$(N-3)F$
	(j)	4	$(N-3)F$
	(k)	4	$(N-3)E$
	(l)	4	$(N-3)(N-4)F$
	(m)	4	$(N-3)F$
	(n)	2	$(N-3)(N-4)F$
	(o)	2	$(N-3)(N-4)F$
	(p)	1	$(N-3)(N-4)E$
	(q)	1	$(N-3)(N-4)$ $\times(N-5)F$
$(2-3)$ $\alpha \longleftrightarrow \beta \quad \gamma$	(a)	2	$-(N-2)F$
$\bullet \quad \bullet \quad \bullet$	(b)	4	$-(N-2)F$
$\bullet$	(c)	4	$-(N-2)(N-3)F$
δ	(d)	2	$-(N-2)(N-3)F$
	(e)	1	$-(N-2)(N-3)$ $\times(N-4)F$
$(2-4)$ $\alpha \longleftrightarrow \beta \quad \gamma$	(r)	2	$-2f_NF$
$\bullet \quad \bullet \quad \bullet$	(s)	1	$-2f_NE$
$\bullet$	(t)	1	$-2(N-3)f_NF$
δ			
$(2-5)$ $\alpha \longleftrightarrow \beta \quad \gamma$	(u)	1	$2(N-1)f_NF$
$\bullet \quad \bullet \quad \bullet$			

[a] See footnote a, Table I.

[b] The letters refer to labels in Fig. 2.

[c] The expressions E and F are defined in Eqs. (A4) and $f_N \equiv \binom{N}{2} - 1$.

$$B \equiv \frac{1}{4} \sum_{\alpha,\beta}{}' F_1(\beta,t') \int \frac{d\beta}{V} \prod_{\kappa}{}'' F_1(\kappa,t) \int \frac{d\kappa}{V}, \tag{A3b}$$

$$C \equiv \frac{1}{4} \sum_{\alpha,\beta}{}' F_1(\alpha,t')F_1(\beta,t') \prod_{\kappa}{}'' F_1(\kappa,t) \int d\Gamma_N, \tag{A3c}$$

and

$$D \equiv \frac{1}{4} \sum_{\alpha,\beta}{}' F_2(\alpha,\beta,t') \prod_{\kappa}{}'' F_1(\kappa,t) \int d\Gamma_N. \tag{A3d}$$

Likewise, only two types of terms occur in Table II, namely,

$$E \equiv \frac{1}{4} \sum_{\alpha,\beta}{}' F_2(\alpha,\beta,t) \int \frac{d\alpha}{V} \int \frac{d\beta}{V} \sum_{\gamma}{}'' \prod_{\kappa}{}''' F_1(\kappa,t) \int \frac{d\kappa}{V} \tag{A4a}$$

and

$$F \equiv \frac{1}{4} \sum_{\alpha,\beta}{}' F_2(\alpha,\beta,t) \int \frac{d\alpha}{V} \int \frac{d\beta}{V} \sum_{\gamma}{}'' F_1(\gamma,t') \prod_{\kappa}{}''' F_1(\kappa,t) \int d\Gamma_N. \tag{A4b}$$

The final result is obtained by multiplying each term in column 4 by its weight in column 3 and summing. For $i=1$, the result obtained from Table I is

$$\begin{aligned} p_N^{(1)}(t)P_N(t') &= 2A + \{8(N-2) + 2(N-2)(N-3) - 4[\tbinom{N}{2} - 1]\}B \\ &\quad + \{5(N-2)(N-3) + (N-2)(N-3)(N-4) \\ &\qquad - 4(N-2)^2 - (N-2)^2(N-3) + 2[\tbinom{N}{2} - 1]\}C \\ &\quad + [2(N-2) - 2(N-2)]D \\ &= 2A \equiv p_N^{(1)}(t), \end{aligned} \tag{A5}$$

where the last step follows from Eq. (A2a). Similarly, we find from Table II,

$$p_N^{(2)}(t)P_N(t') = 2E \equiv p_N^{(2)}(t). \tag{A6}$$

The proof of Eq. (A1) for $i = 3, 4, 5$ is quite simple and should offer the reader no difficulty. The result is the desired proof that $P_N(t)$ is a TDPO.

The prove the identity, Eq. (2.20), we rewrite it in the form

$$\prod_{\substack{\kappa \\ (\kappa\neq\alpha,\beta)}}{}'' \int \frac{d\kappa}{V} P_N(t) \equiv \prod_{\substack{\kappa \\ (\kappa\neq\alpha,\beta)}}{}'' \int \frac{d\kappa}{V}. \tag{A7}$$

We let the summation indices in $P_N(t)$ be δ, ϵ, ζ and prove Eq. (A7) by simply noting the formal equivalence of the problem to that of proving Eq. (A1) for $i=1$. That is, the graphs representing the relationship of α, β to δ, ϵ, ζ and their weights are identical to those in Table I and Fig. 1. We can use Table I directly if we define $(1-j) \equiv \prod_{\kappa}'' \mathrm{Tr}_\kappa p_N^{(j)}(t)$ and replace $A, B, C,$ and D by

$$A' \equiv \frac{1}{2} \prod_{\substack{\kappa \\ (\kappa\neq\alpha,\beta)}}{}'' \int \frac{d\kappa}{V}, \tag{A8a}$$

$$B' \equiv \frac{1}{4} F_1(\alpha,t) \prod_{\substack{\kappa \\ (\kappa\neq\beta)}}{}' \int \frac{d\kappa}{V} + (\alpha \leftrightarrow \beta), \tag{A8b}$$

$$C' \equiv \tfrac{1}{2} F_1(\alpha,t)F_1(\beta,t) \int d\Gamma_N, \tag{A8c}$$

and

$$D' \equiv \tfrac{1}{2} F_2(\alpha,\beta,t) \int d\Gamma_N. \tag{A8d}$$

The sum of terms is equivalent to Eq. (A5), so that

$$\prod_{\substack{\kappa \\ (\kappa\neq\alpha,\beta)}}{}'' \mathrm{Tr}_\kappa P_N(t) = 2A' \equiv \prod_{\substack{\kappa \\ (\kappa\neq\alpha,\beta)}}{}'' \mathrm{Tr}_\kappa,$$

thus proving Eq. (A7).

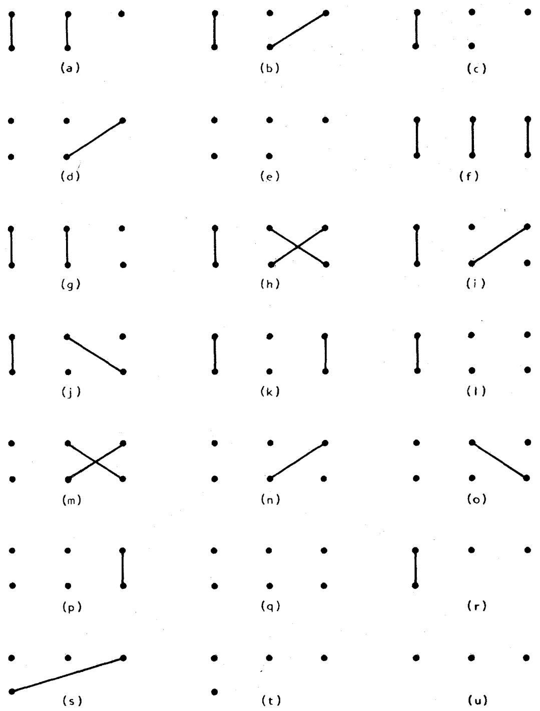

FIG. 2. Graphs associated with Table II and arising in the evaluation of Eq. (A1) for $i=2$.

APPENDIX B: PROOF THAT EQ. (3.7) IS A PROJECTION OPERATOR; PROOF OF EQ. (3.15)

Here we indicate how one proves that the operator $P(t)$ in Eq. (3.7) is the TDPO which projects out of the general density operator for N atoms plus electromagnetic field that part having correlations between a single atom at a time and the field. We decompose $P(t)$ in Eq. (3.7) into $p^{(i)}(t)$, $i=1,\ldots,6$, according to Eq. (2.9) with $l=6$ and prove Eq. (2.10) for $i=1,\ldots,6$, where

$$p^{(1)}(t)\equiv\sum_{\alpha}\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}_{\kappa}\,, \tag{B1a}$$

$$p^{(2)}(t)\equiv\sum_{\alpha}\chi(\alpha,t)\mathrm{Tr}_{\alpha}\mathrm{Tr}_{f}\sum_{\beta}{}^{'}\prod_{\kappa}{}^{''}\rho_1(\kappa,t)\mathrm{Tr}_{\kappa}\,, \tag{B1b}$$

$$p^{(3)}(t)\equiv-(N-1)\sum_{\alpha}\chi(\alpha,t)\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}\,, \tag{B1c}$$

$$p^{(4)}(t)\equiv-(N-1)R(t)\mathrm{Tr}_{f}\sum_{\alpha}\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}_{\kappa}\,, \tag{B1d}$$

$$p^{(5)}(t)\equiv-(N-1)\prod_{\kappa}\rho_1(\kappa,t)\mathrm{Tr}_{m}\,, \tag{B1e}$$

$$p^{(6)}(t)\equiv N(N-1)R(t)\prod_{\kappa}\rho_1(\kappa,t)\mathrm{Tr}\,. \tag{B1f}$$

TABLE III. Terms contributing to $p^{(1)}(t)P(t')$ for the quantum-optical case.

Class[a]	Graphs[b]	Terms[c]
$(1-1)$ α • • δ	(a) (b)	V $(N-1)W$
$(1-2)$ α • • • δ ϵ	(c) (d) (e)	$(N-1)X$ $(N-1)Y$ $(N-1)(N-2)Z$
$(1-3)$ α • • δ	(a) (b)	$-(N-1)X$ $-(N-1)^2Z$
$(1-4)$ α • • δ	(a) (b)	$-(N-1)Y$ $-(N-1)^2Z$
$(1-5)$ α •	(f)	$-(N-1)W$
$(1-6)$ α •	(f)	$N(N-1)Z$

[a] See footnote a, Table I.
[b] The letters refer to labels in Fig. 3.
[c] The expressions V, W, X, Y, and Z are defined in Eqs. (B3).

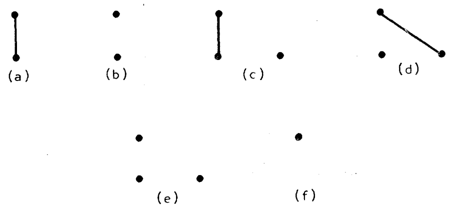

FIG. 3. Graphs associated with Table III and arising in the evaluation of Eq. (2.10) for $i=1$ with $P(t)$ given by Eq. (3.7).

A graphical representation will help us to classify the types of terms arising in the evaluation of the left-hand side of Eq. (2.10) for this case, as in Appendix A; however, the classification is simpler because there are fewer summation indices in this case. We obtain the terms contributing to $P(t')$ by making the replacements $(t;\alpha,\beta,\kappa)\rightarrow(t';\delta,\epsilon,\lambda)$ in Eqs. (B1). The only term containing more than one summation $p^{(2)}$ is not symmetric under interchange of the summation indices α,β (or δ,ϵ). Consequently, one cannot use symmetry to reduce the number of distinct graphs by assignment of weights, as in Appendix A.

The various terms contributing to Eq. (2.10) for $i=1$ are listed in Table III. Once again, we have defined

$$(i-j)\equiv p^{(i)}(t)p^{(j)}(t'). \tag{B2}$$

The graphs associated with Table III are shown in Figs. 3(a)-3(f). Only five types of terms occur in the evaluation $(1-j)$, namely,

$$V\equiv\sum_{\alpha}\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}_{\kappa}\,, \tag{B3a}$$

$$W\equiv\sum_{\alpha}\rho_1(\alpha,t')\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}_{m}\,, \tag{B3b}$$

$$X\equiv\sum_{\alpha}\chi(\alpha,t')\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}\,, \tag{B3c}$$

$$Y\equiv R(t')\mathrm{Tr}_{f}\sum_{\alpha}\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}_{\kappa}\,, \tag{B3d}$$

$$Z\equiv R(t')\sum_{\alpha}\rho_1(\alpha,t')\prod_{\kappa}{}^{'}\rho_1(\kappa,t)\mathrm{Tr}\,. \tag{B3e}$$

The final result is obtained by adding the terms in the last column, with the result

$$\begin{aligned}p^{(1)}(t)P(t')&=V+[(N-1)-(N-1)](W+X+Y)\\&\quad+[(N-1)(N-2)-2(N-1)^2+N(N-1)]Z\\&=V\equiv p^{(1)}(t).\end{aligned} \tag{B4}$$

The last step follows from Eq. (B3a) and verifies Eq. (2.10) for $i=1$.

The next step in the proof that Eq. (3.7) is a TDPO is to verify Eq. (2.10) for $i=2$. This can also be done by drawing graphs as for the case $i=1$. However, it is simpler to note that only two distinct terms arise from all possible graphs. If $\delta=\beta$ in products $(2-1)$ and $(2-4)$ or $\epsilon=\beta$ in products $(2-2)$, one obtains a term with $N-1$ traces over atomic degrees of freedom, namely,

$$T\equiv\sum_{\alpha,\beta}{}^{\prime}\chi(\alpha,t)\mathrm{Tr}_f\mathrm{Tr}_\alpha\prod_{\kappa}{}^{\prime\prime}\rho_1(\kappa,t)\mathrm{Tr}_\kappa\,. \tag{B5a}$$

In all other cases, one obtains a term with N traces over atoms, namely,

$$U\equiv\sum_{\alpha,\beta}{}^{\prime}\chi(\alpha,t)\rho_1(\beta,t')\prod_{\kappa}{}^{\prime\prime}\rho_1(\kappa,t)\mathrm{Tr}\,. \tag{B5b}$$

Hence, for each of the six products $(2-j)$ one must consider no more than two cases separately. When the products are added, one finds that the coefficient of $T\equiv p^{(1)}(t)$ is unity and the coefficient of U is zero, which proves the desired result.

The final steps in the proof that Eq. (3.7) is a PO are to prove Eq. (2.10) for $i=3,4,5,6$. These proofs are either very simple or trivial.

To prove the property of $P(t)$ in Eq. (3.15), we rewrite this identity as

$$\prod_{\substack{\kappa\\(\kappa\neq\alpha)}}{}^{\prime}\mathrm{Tr}_\kappa P(t)\equiv\prod_{\substack{\kappa\\(\kappa\neq\alpha)}}{}^{\prime}\mathrm{Tr}_\kappa\,. \tag{B6}$$

We let δ and ϵ be the summation indices in $P(t)$ and note that the graphs for this case corresponding to the possible relations between α,δ, and ϵ are the same as those for the proof of Eq. (2.10) with $i=1$. Table III and Fig. 3 can be used, provided we redefine $(1-j)\equiv\prod_\kappa'\mathrm{Tr}_\kappa p^{(j)}(t)$ and replace V,W,X,Y, and Z by

$$V'\equiv\prod_{\substack{\kappa\\(\kappa\neq\alpha)}}{}^{\prime}\mathrm{Tr}_\kappa\,, \tag{B7a}$$

$$W'\equiv\rho_1(\alpha,t)\mathrm{Tr}_m, \tag{B7b}$$

$$X'\equiv\chi(\alpha,t)\mathrm{Tr}, \tag{B7c}$$

$$Y'\equiv R(t)\mathrm{Tr}_f\prod_{\substack{\kappa\\(\kappa\neq\alpha)}}{}^{\prime}\mathrm{Tr}_\kappa\,, \tag{B7d}$$

and

$$Z'\equiv R(t)\rho_1(\alpha,t)\mathrm{Tr}. \tag{B7e}$$

According to Eqs. (B4) and (B7a), we then have

$$\prod_{\substack{\kappa\\(\kappa\neq\alpha)}}{}^{\prime}\mathrm{Tr}_\kappa P(t)=V'\equiv\prod_{\substack{\kappa\\(\kappa\neq\alpha)}}{}^{\prime}\mathrm{Tr}_\kappa\,,$$

which is the desired result, Eq. (B6).

APPENDIX C: DERIVATION OF EQS. (4.9) AND (4.18)

In order to derive the approximate ME for the singlet distribution F_1 using the *uncorrelated* form of $F_{N,\tau}$ from I, we substitute Eqs. (4.6) and (4.8) into Eq. (I3.25) and assume $N\gg1$. The result is

$$\dot{F}_1(1,t)-L_1^0F_1(1,t)=I_1+I_2\,, \tag{C1}$$

where

$$I_1\equiv N\int\frac{d2}{V}\,L_{12}'\lim_{\tau\to\infty}\,G^{(2)}(1,2,\tau)G^{(1)}(1,-\tau)G^{(1)}(2,-\tau)F_1(1,t)F_1(2,t)\,, \tag{C2}$$

$$\begin{aligned}I_2\equiv{}&-N\int_0^\infty d\tau'\int\frac{d2}{V}L_{12}'G^{(2)}(1,2,\tau')\left(G^{(1)}(2,\tau')L_1^0F_1(2,t)\int\frac{d2}{V}+(1\leftrightarrow2)\right)\lim_{\tau\to\infty}G^{(2)}(1,2,\tau-\tau')\\&\times G^{(1)}(1,-\tau)G^{(1)}(2,-\tau)F_1(1,t)F_1(2,t)\\&+N\int_0^\infty\int\frac{d2}{V}\,L_{12}'G^{(2)}(1,2,\tau)G^{(1)}(1,-\tau)\,G^{(1)}(2,-\tau)\frac{\partial}{\partial t}\,F_1(1,t)F_1(2,t)\,.\end{aligned} \tag{C3}$$

The term I_1 is obtained when the first term of Eq. (4.8) is substituted into the second of Eq. (I3.25) and is the desired Boltzmann collision term. The first (second) term of I_2 is the result of substituting the second (first) term of Eq. (4.8) into the second (third) term of Eq. (I3.25).

We wish to simplify the first term of I_2. This can be done by noting that we are performing an expansion in powers of λ and using the relations

$$\int\frac{d2}{V}\,G^{(2)}(1,2,\tau-\tau')=G^{(1)}(1,\tau-\tau')\int\frac{d2}{V}\,+O(\lambda) \tag{C4a}$$

and

$$\int\frac{d2}{V}\,G^{(1)}(2,-\tau)F_1(2,t)=\int\frac{d2}{V}\,F_1(2,t)=1\,, \tag{C4b}$$

along with the group property of the time-development operator $G^{(1)}$,

$$G^{(1)}(1,\tau_1)G^{(1)}(1,\tau_2)=G^{(1)}(1,\tau_1+\tau_2)\,. \tag{C4c}$$

Equations (C4) can be proven using Eqs. (4.5), (4.7), (2.16), and (I3.7b). Hence, the first term of I_2 can be written

$$-N\int_0^{\infty}d\tau'\int\frac{d2}{V}\,L_{12}'G^{(2)}(1,2,\tau')G^{(1)}(2,-\tau')L_1^0F_1(2,t)G^{(1)}(1,-\tau')F_1(1,t)+(1\leftrightarrow 2)\,. \tag{C5}$$

Using Eq. (C5) and noting that

$$[L_1^0,G^{(1)}(1,\tau)]=0\,, \tag{C6}$$

one may write Eq. (C3) as

$$I_2=N\int_0^{\infty}d\tau\int\frac{d2}{V}\,L_{12}'\mathcal{S}^{(2)}(1,2,\tau)\left(\frac{\partial}{\partial t}-L_1^0-L_2^0\right)F_1(1,t)F_1(2,t)\,, \tag{C7}$$

where $\mathcal{S}^{(2)}$ is defined in Eq. (4.10). The desired ME, Eq. (4.9), is obtained by substituting Eqs. (C2) and (C7) into Eq. (C1).

Next, we wish to derive an approximate ME for the doublet distribution function F_2 using the *correlated* form of $F_{N,r}$ discussed in Sec. II. Substituting Eq. (4.17) for $\mathcal{G}^{(3)}$ into Eq. (4.12), assuming $N\gg 1$, and using Eq. (4.13b), we obtain

$$\dot{F}_2(1,2,t)-L_{(2)}(1,2)F_2(1,2,t)=I_3+I_4\,, \tag{C8}$$

where

$$I_3\equiv N\int\frac{d3}{V}\,(L_{13}'+L_{23}')\lim_{\tau\to\infty}G^{(3)}(1,2,3,\tau)F_{3,r}(1,2,3,t-\tau) \tag{C9}$$

and

$$\begin{aligned}I_4\equiv -N\int_0^{\infty}d\tau'\int\frac{d3}{V}(L_{13}'+L_{23}')G^{(3)}(1,2,3,\tau')\Big[L_1^0\Big(F_1(2,t-\tau')\int\frac{d2}{V}+(2\to 3)\\ +[F_2(2,3,t-\tau')-2F_1(2,t-\tau')F_1(3,t-\tau')]\int\frac{d2}{V}\int\frac{d3}{V}\Big)\\ +L_{12}'F_1(3,t-\tau')\int\frac{d3}{V}+\text{cycl}(1,2,3)\Big]\\ \times\lim_{\tau\to\infty}G^{(3)}(1,2,3,\tau-\tau')F_{3,r}(1,2,3,t-\tau)\\ +N\int_0^{\infty}d\tau\int\frac{d3}{V}\,(L_{13}'+L_{23}')G^{(3)}(1,2,3,\tau)\,\frac{\partial}{\partial t}\,F_{3,r}(1,2,3,t-\tau)\,.\end{aligned} \tag{C10}$$

Here I_3 is obtained when the first term of Eq. (4.17) is substituted into the first term of the right-hand side of Eq. (4.12) and is the desired term of order λ. On the other hand, the first (second) term of I_4 results from substituting the second (first) term of Eq. (4.17) into the first (second) term on the right-hand side of Eq. (4.12).

In order to simplify Eq. (C10), we first note the following relations:

$$\int\frac{d2}{V}\,G^{(3)}(1,2,3,\tau-\tau')=G^{(2)}(1,3,\tau-\tau')\int\frac{d2}{V}+O(\lambda)\,, \tag{C11a}$$

$$\int\frac{d2}{V}\int\frac{d3}{V}\,G^{(3)}(1,2,3,\tau-\tau')=G^{(1)}(1,\tau-\tau')\int\frac{d2}{V}\int\frac{d3}{V}+O(\lambda)\,, \tag{C11b}$$

which can be proven from the definitions of $G^{(3)}$, $G^{(2)}$, and $G^{(1)}$ [Eqs. (4.16), (4.5), and (4.7)]. From Eqs. (C11) and (2.4), Eq. (C10) for I_4 may be written

$$\begin{aligned} I_4 = -N\int_0^\infty d\tau' \int \frac{d3}{V}\,(L'_{13}+L'_{23})G^{(3)}(1,2,3,\tau')\Big(& L_1^0\{F_1(2,t-\tau')\lim_{\tau\to\infty} G^{(2)}(1,3,\tau-\tau')F_2(1,3,t-\tau)+(2\leftrightarrow 3) \\ & +[F_2(2,3,t-\tau')-2F_1(2,t-\tau')F_1(3,t-\tau')] \\ & \times\lim_{\tau\to\infty} G^{(1)}(1,\tau-\tau')F_1(1,t-\tau)\} \\ & +L'_{12}F_1(3,t-\tau')\lim_{\tau\to\infty} G^{(2)}(1,2,\tau-\tau')F_2(1,2,t-\tau) \\ & +\text{cycl}(1,2,3)-\frac{\partial}{\partial t}F_{3,r}(1,2,3,t-\tau')\Big)\,. \end{aligned} \tag{C12}$$

Now one may use Eqs. (4.6) and (4.15), along with the group property of $G^{(1)}$, Eq. (C4c), and the analogous relation for $G^{(2)}$, to make the time argument t in every distribution function F_1 and F_2 of Eq. (C12). If we also replace some terms explicitly written in Eq. (C12) by their cyclic permutations and use Eq. (C6) and the relation

$$[L_1^0+L_2^0+L'_{12},G^{(2)}(1,2,\tau)]=0\,, \tag{C13}$$

the result is

$$\begin{aligned} I_4 = N\int_0^\infty d\tau \int \frac{d3}{V}\,(L'_{13}+L'_{23})G^{(3)}(1,2,3,\tau)\Big[& G^{(2)}(1,2,-\tau)G^{(1)}(3,-\tau)F_1(3,t)\Big(\frac{\partial}{\partial t}-L_1^0-L_2^0-L'_{12}\Big)F_2(1,2,t) \\ & +G^{(1)}(3,-\tau)[G^{(2)}(1,2,-\tau)F_2(1,2,t)-2G^{(1)}(1,-\tau) \\ & \times G^{(1)}(2,-\tau)F_1(1,t)F_1(2,t)]\Big(\frac{\partial}{\partial t}-L_3^0\Big)F_1(3,t)+\text{cycl}(1,2,3)\Big]\,, \end{aligned} \tag{C14}$$

where we have replaced the dummy variable τ' by τ. Rearranging terms in Eq. (C14) and combining it with Eqs. (C8) and (C9) yields the desired doublet ME, Eq. (4.18).

APPENDIX D: EFFECT OF IRREDUCIBLE TWO-ATOM CORRELATIONS IN A LASER

Here we evaluate the contribution of irreducible two-atom-plus-field correlations to the ME for χ, Eq. (5.11), and show explicitly that the contribution is small for any reasonable laser parameters. In order to do this, it is necessary to first write out explicitly the right side of Eq. (5.11) and then take moments to determine the relative contribution of the right-hand side to the dynamical equations for the moments.

Using Eqs. (3.2) and (3.22) the right-hand side of Eq. (5.11) may be written

$$\begin{aligned} \frac{\partial\chi(\alpha,t)}{\partial t}\bigg|_{2\text{-atom}} = T_2 \sum_{\substack{\beta \\ (\beta\neq\alpha)}}{}' \, \mathrm{Tr}_\beta L'_\beta\{ & L'_\beta\rho_1(\beta,t)\Delta\chi(\alpha,t)+L'_\alpha\rho_1(\alpha,t)\chi(\beta,t) \\ & -\mathrm{Tr}_f[L'_\beta\chi(\beta,t)]\,\Delta\chi(\alpha,t)-\mathrm{Tr}_f[L'_\alpha\chi(\alpha,t)]\,\chi(\beta,t)\}\,, \end{aligned} \tag{D1}$$

where we have used the vanishing of the average field and average polarization due to the fact that R and $\rho_1(\alpha)$ are diagonal. Of the four terms in Eq. (D1), not all contribute to the dynamical equation of any given moment. For example, only the fourth term contributes to the time development of diagonal field moments such as $\langle a^+a^-\rangle$ and only the first and fourth contribute to the dynamical equation of diagonal matter moments such as $\langle\sigma_\alpha^+\sigma_\alpha^-\rangle$. However, we will evaluate the time development of $\langle a^+\sigma_\alpha^-\rangle$, for which we only need the first and second terms of Eq. (D1). We call the ith term of Eq. (D1) $\psi_i(\alpha,t)$,

$$\frac{\partial\chi(\alpha,t)}{\partial t}\bigg|_{2\text{-atom}} \equiv \sum_{i=1}^{4}\psi_i(\alpha,t)\,, \tag{D2}$$

and evaluate ψ_1 and ψ_2 by using Eqs. (1.8) and (5.4c) to express L'_α. After some manipulation we obtain

$$\psi_1(\alpha,t)=-|\mu|^2T_2\sum_{\substack{\beta\\(\beta\neq\alpha)}}{}'\{\langle\sigma_\beta^+\sigma_\beta^-\rangle_t([a^-,[a^+,\Delta\chi(\alpha,t)]]+[a^-,\Delta\chi(\alpha,t)a^+]-[a^+,\Delta\chi(\alpha,t)a^-])$$
$$-\langle\sigma_\beta^-\sigma_\beta^+\rangle_t([a^-,\Delta\chi(\alpha,t)a^+]-[a^+,\Delta\chi(\alpha,t)a^-]-[a^+,[a^-,\Delta\chi(\alpha,t)]])\}, \tag{D3a}$$

$$\psi_2(\alpha,t)=-|\mu|^2T_2\sum_{\substack{\beta\\(\beta\neq\alpha)}}{}'\,e^{i\vec{k}\cdot(\vec{x}_\beta-\vec{x}_\alpha)}(\sigma_\alpha^-\rho_1(\alpha,t)[a^-,a^+\mathcal{P}_\beta^+(t)]-\rho_1(\alpha,t)\sigma_\alpha^-[a^-,\mathcal{P}_\beta^+(t)a^+])$$
$$-\mu^2T_2\sum_{\substack{\beta\\(\beta\neq\alpha)}}{}'\,e^{i\vec{k}\cdot(\vec{x}_\beta+\vec{x}_\alpha)}(\sigma_\alpha^+\rho_1(\alpha,t)[a^-,a^-\mathcal{P}_\beta^+(t)]-\rho_1(\alpha,t)\sigma_\alpha^+[a^-,\mathcal{P}_\beta^+(t)a^-])+\text{H.c.}, \tag{D3b}$$

where $\mathcal{P}_\beta^\pm$ is defined in Eq. (5.22).

Multiplying Eq. (D2) by $P_k^+a^-$, using Eqs. (D.3), and taking the trace, one obtains

$$\left.\frac{d\langle P_k^+a^-\rangle}{dt}\right|_{2\text{-atom}}=|\mu|^2T_2\sum_\beta\langle s_\beta\rangle\sum_\alpha{}'e^{i\vec{k}\cdot\vec{x}_\alpha}\langle\sigma_\alpha^+a^-\rangle$$
$$-\mu^{*2}T_2\sum_\alpha\langle s_\alpha\rangle\sum_\beta{}'\,e^{-i\vec{k}\cdot\vec{x}_\beta}\langle a^+\sigma_\beta^-\rangle\,. \tag{D4}$$

By a similar procedure, one finds

$$\left.\frac{d\langle a^+P_k^-\rangle}{dt}\right|_{2\text{-atom}}=|\mu|^2T_2\sum_\beta\langle s_\beta\rangle\sum_\alpha{}'e^{-i\vec{k}\cdot\vec{x}_\alpha}\langle a^+\sigma_\alpha^-\rangle$$
$$-\mu^2T_2\sum_\alpha\langle s_\alpha\rangle\sum_\beta{}'e^{i\vec{k}\cdot\vec{x}_\beta}\langle\sigma_\beta^+a^-\rangle\,. \tag{D5}$$

Multiplying Eq. (D4) by μ and Eq. (D5) by μ^* and adding, one obtains

$$\frac{1}{\hbar}\left.\frac{d\langle H'\rangle}{dt}\right|_{2\text{-atom}}=0\,, \tag{D6}$$

verifying Eq. (5.13). If we subtract instead of adding, we obtain

$$\frac{d}{dt}\langle\mu P_k^+a^- - \mu^*a^+P_k^-\rangle|_{2\text{-atom}}$$
$$=2|\mu|^2T_2$$
$$\times\sum_\beta\langle s_\beta\rangle\sum_\alpha{}'(\mu e^{i\vec{k}\cdot\vec{x}_\alpha}\langle\sigma_\alpha^+a^-\rangle-\mu^*e^{-i\vec{k}\cdot\vec{x}_\alpha}\langle a^+\sigma_\alpha^-\rangle)\,. \tag{D7}$$

Now the sums over α in Eq. (D7) would be $P_k^\pm$, were it not for the prime on the sum. Hence, we may eliminate them by using

$$\sum_{\substack{\alpha\\(\alpha\neq\beta)}}{}'e^{\pm i\vec{k}\cdot\vec{x}_\alpha}\sigma_\alpha^\pm\cong\frac{N-1}{N}P_k^\pm\,, \tag{D8}$$

with negligible error, and we have reduced Eq. (D7) to Eq. (5.14).

Now we show that the correction to the relaxation rate of $\text{Im}(\mu\langle P_k^+a^-\rangle)$ due to two-atom-plus-field correlations, that is r in Eqs. (5.17), is small. First, since $\langle s\rangle\leq1$, r is bounded by $|\mu|^2T_2^2N\equiv\epsilon$, where ϵ is the Born-approximation expansion parameter. To obtain a more accurate expression for the magnitude of r, we replace $\langle s\rangle$ by its steady-state value $\langle s\rangle_{ss}$. To calculate $\langle s\rangle_{ss}$, we assume that $T_2\ll T_1, T_f$, where T_1 is the lifetime of the two-level atom. We further assume that there is pumping only into the upper laser level and make the breakup

$$\langle a^+a^-s\rangle\cong\langle a^+a^-\rangle\langle s\rangle\,, \tag{D9}$$

corresponding to neglecting certain atom-field correlations. Then the equations for $\langle s\rangle$ and $\langle a^+a^-\rangle$ give rise to the steady-state results[17, 60]

$$\langle s\rangle_{ss}=\langle[1+4|\mu|^2T_1T_2(a^+a^-+1)]^{-1}\rangle$$
$$\cong[1+4|\mu|^2T_1T_2(\langle a^+a^-\rangle_{ss}+1)]^{-1} \tag{D10}$$

and

$$4|\mu|^2T_1T_2\langle a^+a^-\rangle_{ss}=N/N_{\text{th}}-1\,,\quad N\geq N_{\text{th}}\,, \tag{D11}$$

where $N_{\text{th}}\equiv(2|\mu|^2T_2T_f)^{-1}$ is the threshold number of two-level atoms. From Eqs. (D10) and (D11) we find

$$\langle s\rangle_{ss}=N_{\text{th}}/N\,,$$

from which Eq. (5.17b) yields

$$r=|\mu|^2T_2^2N_{\text{th}}=T_2/2T_f\,, \tag{D12}$$

in agreement with Eq. (5.18).

APPENDIX E: MAGNITUDE OF COMMUTATOR TERMS IN EQ. (5.35)

Here we wish to show that the commutator term appearing in Eq. (5.35), $a^+[a^-,\mathcal{N}^+]$, is of order unity near threshold, while the commutator terms $a^-[a^+,\mathcal{N}^-]$ are of order n/n_{sat}, or typically 10^{-3}. The operators $\mathcal{N}^\pm$ and R are functions of the photon-number operator a^+a^-, since they are diagonal in the number representation. According to the equations of the usual laser theory,[16-18] which are adequate for our estimates, the inversion exhibits a saturation-type behavior, so that[61]

$$\mathcal{N}^+-\mathcal{N}^-\cong(1+a^-a^+/n_{\text{sat}})^{-1}R(a^-a^+)\,, \tag{E1a}$$

from which

$$\mathcal{N}^+ = (1+\tfrac{1}{2}a^-a^+/n_{\text{sat}})(1+a^-a^+/n_{\text{sat}})^{-1}R(a^-a^+)$$
$$\equiv f(a^-a^+)R(a^-a^+) \tag{E1b}$$

and

$$\mathcal{N}^- = \tfrac{1}{2}(a^-a^+/n_{\text{sat}})(1+a^-a^+/n_{\text{sat}})^{-1}R(a^-a^+)\,. \tag{E1c}$$

Near threshold, we may write[39] $\langle a^+a^-\rangle \sim n_{\text{sat}}^{1/2} \ll n_{\text{sat}} \cong 10^6$. Hence, it is clear from Eqs. (E1b) and (E1c) that $\mathcal{N}^- \sim (n/n_{\text{sat}})\mathcal{N}^+ \cong 10^{-3}\mathcal{N}^+$ for typical lasers. This is the ultimate source of the disparity between the two commutators in question.

To demonstrate the result explicitly, we write the first of the expressions in whose magnitude we are interested as

$$a^+[a^-,\mathcal{N}^+] = a^+\left(\frac{\partial f(a^-a^+)}{\partial a^+}R(a^-a^+) + f(a^-a^+)\frac{\partial R(a^-a^+)}{\partial a^+}\right), \tag{E2}$$

using Eqs. (5.39) and (E1b). From Eq. (E1b) we estimate that $a^+\partial f/\partial a^+ \sim a^+a^-/n_{\text{sat}}$ near threshold, so that the magnitude of the first term of Eq. (E2) is

$$a^+\frac{\partial f}{\partial a^+}R \sim \frac{n}{n_{\text{sat}}}R\,. \tag{E3}$$

To estimate the magnitude of the second term in Eq. (E2), we can write

$$a^+\partial R(a^-a^+)/\partial a^+ \sim nR'(n)\,, \tag{E4}$$

where the prime indicates a derivative with respect to the argument n. Using the standard laser theory,[16-18] we can evaluate $R'(n)$ near threshold. By the method of Fleck,[62,63] we write

$$R(n) \cong A(\sigma, n_0/\sigma)\exp[-\tfrac{1}{2}(n-n_0)^2/\sigma^2]\,, \tag{E5a}$$

where A is a normalization factor involving the error function, so that

$$nR'(n) \cong -n(n-n_0)\sigma^{-2}R(n)\,. \tag{E5b}$$

Now at threshold we have $n_0 = 0$, and it follows that

$$nR'(n) = -(n/\sigma)^2R(n)$$
$$\sim -(\langle n^2\rangle/\sigma^2)R(n) = -R(n)\,. \tag{E6}$$

In the next-to-last step we have replaced n^2 by its average value. Since $f\sim 1$ near threshold, we conclude from Eq. (E4) that the second term of Eq. (E2) $\sim -R$. Moreover, it dominates the first term, from a comparison with Eq. (E3). Hence, we may estimate the magnitude of the commutator near threshold by

$$a^+[a^-,\mathcal{N}^+] \sim -R(a^-a^+)\,. \tag{E7}$$

By similar methods we can show that

$$a^-[a^+,\mathcal{N}^-] \sim \tfrac{1}{2}(a^-a^+/n_{\text{sat}})R(a^-a^+) \tag{E8}$$

near threshold and, hence, is negligible compared with the commutator of Eq. (E7).

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[21] Compare Eq. (2.20) to Eq. (I3.18) for the case of the uncorrelated $F_{N,r}(t)$ given by Eq. (1.14).

[22] See, for example, T.-Y. Wu, *Kinetic Equations of Gases and Plasmas* (Addison-Wesley, Reading, Mass., 1966), p. 62.

[23] Although the results of this section transcend any particular form for H_f, H_α^0, and H_α', the interested reader may refer to the explicit form of the single-mode Hamiltonian given in Eq. (5.4). The notation χ is actu-

ally redundant since $\chi(\alpha,t) \equiv F_1(\alpha,t)$. Nevertheless, we will use χ to distinguish the quantum-optical case from the gas-kinetic case of Sec. II.

[24] More general forms of $F_{N,r}(t)$ depending nonlinearly on $\Delta\chi$ will be considered in Sec. VI. However, the linear form indicated in Eq. (3.3) is usually sufficiently rich to provide a convenient description.

[25] Note that we have defined the quantum Liouvillian in a slightly different manner than in I and, consequently, we have a different form for the quantum-Liouville equation. Compare Eq. (I 1.5) to Eqs. (1.1) and (1.8). The definitions in the current paper allow the dynamical equations to assume the same form classically and quantum mechanically.

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[27] Equations (4.3), (4.4), and (4.8) correct misprints in Eqs. (I3.31), (I3.28), and (I3.32). In addition, they explicitly indicate the dependence of $\mathcal{G}^{(2)}$, $G^{(2)}$, $\mathfrak{L}_{(2)}$, and so forth, on phase-space arguments. Equation (4.5) agrees with Eq. (I3.30).

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[31] Compare Eqs. (2.20) and (2.21) of Ref. 30 and the associated discussion.

[32] Compare the discussion leading to Eqs. (2.27) in Ref. 30.

[33] Strictly speaking, the condition $\epsilon \ll 1$ justifies the Born-Markoff approximation and there is no small parameter to justify the first Born *non-Markoffian* equation for $\dot{\chi}$, Eq. (5.3). Nevertheless, non-Markoffian FBA equations often describe qualitatively significant effects, as discussed in Sec. IV of I for the superradiance problem. Non-Markoffian effects in the laser have been discussed by F. Haake, Z. Phys. 227, 179 (1969).

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[36] The condition $T_2 \ll T_1$ is not strictly necessary. In the gas laser case where T_1 and T_2 are comparable, it is necessary to eliminate the population and the polarization adiabatically at the same time. Since we will later eliminate the population also, our results apply to the gas laser.

[37] This follows from tracing Eq. (5.28) over the field [see Eq. (5.31)]. If a fraction n_0^+ of the atoms is pumped into the upper laser state and $n_0^- \equiv 1 - n_0^+$ into the lower state, $R(t)$ is replaced by $n_0^+ R(t)$ in Eq. (5.28). The matter-reservoir term in the $\mathcal{N}_\alpha^-$ equation will be the negative of the one in the $\mathcal{N}_\alpha^+$ equation. Using Eq. (5.25b), we can write it as $\mathrm{Tr}_\alpha n_\alpha^- K_m \chi(\alpha,t) = +T_1^{-1} \mathcal{N}_\alpha^-(t)$ in the case $n_0^+ = 1$, which when traced over the field clearly shows the pump drives the lower-state population to zero.

[38] See Ref. 17, Eq. (4.3), for example, or Eq. (D10) of the current paper.

[39] See Ref. 17, Eqs. (4.10b) and (4.13), for example, and note that $\langle n\rangle \cong \langle n^2\rangle^{1/2} \equiv \sigma$ at threshold.

[40] In general one can write $\langle n|K_f R|n\rangle = \sum_{n'} K_f^{nn;n'n'} R_{n'}$. In Eq. (3.10c) of Ref. 17, a zero-temperature reservoir has been chosen for K_f, in which case $\langle n|K_f R|n\rangle = T_f^{-1}[nR_n - (n+1)R_{n+1}]$.

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