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Time-dependent projection-operator approach to master equations for coupled systems

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In this paper we derive master equations for two or more systems coupled to each other, perhaps strongly, by using a generalization of the usual projection-operator technique to include time-dependent projection operators. The coupled systems may be either similar or dissimilar and classical or quantum mechanical. Whereas the customary approaches to coupled systems are best able to treat situations in which some of the systems are "baths" with a specified density operator or phase-space probability density, our approach allows us to treat situations where it is necessary or convenient to treat the coupled systems on an equal footing. In our scheme the "relevant" part of the full density operator is considered to be the uncorrelated part of the full density operator and is a symmetric functional of the reduced density operators of each of the coupled subsystems. The "irrelevant" part of the density operator is then the part describing correlations between the coupled systems. Our formalism is particularly useful where systems are coupled to one another predominantly in a self-consistent fashion. First, we develop exact master equations for two coupled systems, taking as our prototype the dynamical problem of quantum optics, where a spatially extended collection of two-level atoms interact with a multimode optical field. We then generalize our results to N coupled systems, taking as our prototype the kinetics of a classical nonideal gas interacting through two-body forces, and derive exact master equations for the system. We then consider as examples several approximate theories resulting from our exact equations. In the case of the imperfect gas we investigate the low-density limit and show how Bogoliubov's form of the Boltzmann equation emerges from our formalism, as well as corrections due to Klimontovich. We consider as special cases of our exact quantum-optical equations the equations in the first Born approximation, with and without memory, and show how several existing quantum-optical master equations are contained in our general results. As a second example in quantum optics, we consider the case where the predominant behavior of the system is described by the self-consistent-field or coupled Bloch and Maxwell equations and derive a first-order perturbation description for deviations from self-consistent-field behavior.

I. INTRODUCTION

The use of projection-operator techniques[1-3] to derive master equations has become increasingly more common in recent years. Although projection-operator techniques are extremely general, they have been especially useful in problems where systems are in contact with reservoirs or where one is interested only in the diagonal matrix elements of the density operator $F(t)$ in some representation. In these problems the degrees of freedom of the reservoirs or the off-diagonal part of the density operator are considered to be irrelevant, and the "irrelevant" part $F_i(t)$ of the density operator is formally eliminated. One is left with an exact equation for the remaining "relevant" part of the density operator $F_r(t)$, which is related to $F(t)$ through a projection operator P.

In a recent article Haake[4] has studied a wide variety of problems involving coupled systems using standard projection-operator techniques; these problems have in common that one of the coupled systems can be treated as a bath and its degrees of freedom treated as irrelevant. However, in many problems involving coupled dynamical systems, it is very convenient to have a description where each of two or more coupled systems is considered to be on an equal footing, neither of them being an irrelevant part of the system. In this paper we will derive master equations for coupled dynamical systems where the irrelevant degrees of freedom are not the dynamical degrees of freedom of one of the coupled systems, but rather the correlations between the systems, produced by the interaction. Moreover, we will allow for the possibility that the reduced density operators of the subsystems possess off-diagonal matrix elements. Such a view is particularly profitable in situations where a self-consistent treatment of the interacting systems is important or where the subsystems are strongly interacting and neither of them can be considered large or reservoir-like. For example, such situations arise (i) in quantum optics, where optical fields interact with a polarizable medium and either optical feedback or propagation is an important effect; (ii) in the kinetic theory of imperfect gases; and (iii) in the theory of inhomogeneous plasmas, where a description of self-consistent effects is desired.

One advantage of this approach will become apparent. Although the rigorous master equations are equivalent to master equations using other projection operators (and to the Liouville equation), a much better description of the system is possible in a suitable low order of approximation using the coupled-systems approach. A disadvantage of the approach is that our projection operators will become time dependent; however, this disadvantage is not serious.

First, we will consider two coupled systems to demonstrate the general approach and derive the exact master equations. To be specific, we will usually speak in quantum-optical terms, where an optical field is coupled to a matter system consisting of polarizable two-level atoms. However, the approach applies equally well to many other systems. Then we will consider a case of N coupled systems, namely a classical imperfect gas of N molecules interacting with one another through an arbitrary force law. We will show that, in the low-density limit when the force range is short, one obtains the Boltzmann equation of Bogoliubov,[5] and we will consider corrections to it. Finally, we will return to our general quantum-optical master equations and consider two different approximations to them.

Hence, turning to the problem of two coupled systems, or the quantum-optical case, we divide the system into relevant and irrelevant parts such that the relevant part is an uncorrelated state of the two coupled systems. Thus we may write for the complete density operator $F(t)$,

$$F(t)=F_r(t)+F_i(t), \tag{1.1}$$

where

$$F_r(t)=R(t)\rho(t), \quad F_i(t)=F(t)-R(t)\rho(t), \tag{1.2}$$

and R and ρ are the reduced density operators of the field and matter, respectively, defined by

$$R(t)=\mathrm{Tr}_m F(t), \quad \rho(t)=\mathrm{Tr}_f F(t). \tag{1.3}$$

The symbol Tr_m denotes a trace over all degrees of freedom of the matter and Tr_f a trace over all those of the field.

Uncorrelated states of matter and field are familiar from the well-known and important self-consistent-field approximation (SCFA) for interacting radiation-matter systems, where the correlations are identically zero for all time; that is, $F_i(t)=0$, so that

$$F_{\mathrm{SCF}}(t)=R(t)\rho(t). \tag{1.4}$$

One does not normally use the projection-operator formalism to obtain the SCFA. One need only substitute Eq. (1.4) into the quantum-Liouville equation for $F(t)$, which can be written in tetradic or Liouvillian form[2]

$$\begin{aligned}\dot{F}(t)&=-iLF(t)\\ &\equiv -i\hbar^{-1}[H,F(t)],\end{aligned} \tag{1.5}$$

and take traces of the equation over matter and over field variables to obtain equations for $\dot{R}(t)$ and $\dot{\rho}(t)$, respectively. In this equation the superior dot stands for $\partial/\partial t$, and H is the total system Hamiltonian. The resultant coupled equations for R and ρ constitute the SCFA. If the matter is composed of two-level atoms, the SCFA leads to the Bloch-Maxwell (BM) equations of quantum optics for the first moments of R and ρ.

In the general case, even though the system is initially in an uncorrelated state $F(0)=R(0)\rho(0)$ and the part of the density operator describing correlations $F_i(t)=F-R\rho$ vanishes initially, correlations will develop in time through the interaction so that $F_i(t)\neq 0$ for $t>0$. Consequently, one needs to develop coupled master equations which include coupling between R and ρ, not only through the self-consistent field (SCF) but also through dynamically induced correlations. The projection-operator formalism provides a convenient means for doing this by allowing us to express the dynamical correlations in terms of the uncorrelated part of the density operator, leading to exact closed coupled equations for $R(t)$ and $\rho(t)$.

An interesting feature of our approach to coupled systems is that it requires a time-dependent projection operator. Time-dependent projection operators have been used by several authors.[6-9] One of the subtleties in deriving master equations with time-dependent projection operators $P(t)$ is that the projection operator does not commute with the time-derivative operator $\partial/\partial t$ on the left-hand side of the Liouville equation, Eq. (1.5), but rather

$$[P(t),\partial/\partial t]\equiv -\dot{P}(t). \tag{1.6}$$

In our case, however, we will see that the projection operator we shall choose behaves more like a time-independent projector in this regard, since

$$\dot{P}(t)F(t)=0, \qquad (1.7)$$

so that $P(t)$ effectively commutes with $\partial/\partial t$ when operating on the full density operator.

In Sec. II we introduce the projection operator and derive the exact coupled equations for R and ρ. In particular, we mention the quantum-optical case where R and ρ are the field and matter density operators, respectively. Then in Sec. III we generalize the formalism to the case of N coupled systems and apply the resulting master equations to the kinetics of a nonideal classical gas. In the low-density limit we obtain the generalized Boltzmann equation,[5] as well as corrections to it due to Klimontovich.[10] In the following two sections (Secs. IV and V) we return to the quantum-optical case and discuss two different approximations to the exact master equations. In Sec. IV we treat the field-matter interaction in the first Born approximation (FBA) and show the important role played in the treatment of optical interactions by non-Markoffian effects. We also discuss how coupling atoms or field strongly to a reservoir leads to Markoffian behavior. In Sec. V we present a perturbative treatment of fluctuations about the SCF and show how the coupling between field fluctuations and polarization fluctuations is modified by the presence of a strong average field. We conclude in Sec. VI by comparing our work to that of others.

II. DERIVATION OF MASTER EQUATIONS

The full Hamiltonian H of our system is composed of a free part H^0 and an interaction part H'. In turn, H^0 is the sum of the free Hamiltonians of each of the coupled systems, that is

$$H=H^0+H'=H_m+H_f+H', \qquad (2.1)$$

where H_m is the matter and H_f the field Hamiltonian. As we mentioned earlier, the derivation holds equally well for any two coupled systems. Substituting Eq. (2.1) into the right-hand equality in Eq. (1.5), we see that the tetradic operator L is also a sum of matter, field, and interaction terms,

$$L=L^0+L'=L_m+L_f+L'. \qquad (2.2)$$

The first task in applying the projection-operator formalism to our coupled system is to define the projection operator P which will perform the projection onto $F_r(t)$, the relevant part of $F(t)$, indicated in Eqs. (1.1) and (1.2). Although there is more than one operator that will accomplish this projection, it is advantageous to have one which is symmetric in matter and field variables. Thus we choose

$$P(t)=R(t)\,\mathrm{Tr}_f+\rho(t)\,\mathrm{Tr}_m-R(t)\rho(t)\,\mathrm{Tr}, \qquad (2.3)$$

where $\mathrm{Tr}\equiv\mathrm{Tr}_f\,\mathrm{Tr}_m$, in terms of which

$$F_r(t)=P(t)F(t), \quad F_i(t)=[1-P(t)]F(t). \qquad (2.4)$$

From Eq. (2.3), one can derive the simple properties[11]

$$P(t_1)P(t_2)\equiv P(t_1), \qquad (2.5a)$$

$$[1-P(t_1)][1-P(t_2)]\equiv 1-P(t_2), \qquad (2.5b)$$

$$P(t_1)[1-P(t_2)]\equiv 0, \qquad (2.5c)$$

for arbitrary t_1 and t_2. Letting $t_1=t_2$ in Eq. (2.5a), we find that $P^2(t)\equiv P(t)$, so that $P(t)$ is indeed a projection operator.

We are now ready to derive the equation of motion of $F_r(t)$ by operating on the Liouville equation, Eq. (1.5), with $P(t)$. Since

$$\dot{P}(t)=\dot{R}(t)\,\mathrm{Tr}_f+\dot{\rho}(t)\,\mathrm{Tr}_m-[\dot{R}(t)\rho(t)+R(t)\dot{\rho}(t)]\,\mathrm{Tr}, \qquad (2.6)$$

Eq. (1.7) is satisfied. Therefore, using Eq. (1.6), we have

$$\left[P(t),\frac{\partial}{\partial t}\right]F(t)=0. \qquad (2.7)$$

From Eqs. (2.7), (1.2), and (1.1), it follows that

$$\dot{F}_r(t)=-iP(t)LF_r(t)-iP(t)LF_i(t). \qquad (2.8)$$

To eliminate $F_i(t)$ from Eq. (2.8), we require its equation of motion, obtained by operating on Eq. (1.5) with the projection operator $1-P(t)$, namely

$$\dot{F}_i(t)=-i[1-P(t)]LF_i(t)-i[1-P(t)]LF_r(t). \qquad (2.9)$$

By integrating Eq. (2.9) and substituting the result into Eq. (2.8), we obtain the master equation for $F_r(t)$,

$$\dot{F}_r(t)=-iP(t)L\mathcal{G}(t,0)F_i(0)-iP(t)LF_r(t)-\int_0^t dt'P(t)L\mathcal{G}(t,t')[1-P(t')]LF_r(t'), \qquad (2.10)$$

where

$$\mathcal{G}(t,t')\equiv T\exp\left(-i\int_{t'}^{t}dt''[1-P(t'')]L\right), \qquad (2.11)$$

T being the Dyson time-ordering operator.

We will ignore the first term of Eq. (2.10), since this term vanishes for the usual case where the system is in an initial state without matter-field correlations, in which case $F(0)=R(0)\rho(0)$, or

$$F_i(0)=0. \qquad (2.12)$$

The vanishing of the initial correlations is one aspect of the asymptotic condition in Bogoliubov's method of deriving kinetic equations.[5] Important examples of such uncorrelated initial states for systems of many two-level atoms coupled to resonant fields include (i) the state where every atom in the sample is in the excited state and the field is in the vacuum state and (ii) the state where all atoms are in the ground state and a specified field impinges on the sample.

Next we show that both operators L in the integral of Eq. (2.10) can be replaced by L'. We replace the first L by L' with the help of the relation

$$P(t)L_m = L_m P(t) - L_m \rho(t)\,\mathrm{Tr}_m[1 - R(t)\,\mathrm{Tr}_f] \tag{2.13}$$

and an analogous expression for $P(t)L_f$, where we used $\mathrm{Tr}_m L_m \equiv \mathrm{Tr}_f L_f \equiv 0$. With the use of Eq. (2.13) and its analog, noting that $\mathcal{G}(t,t')[1-P(t')]$ is a sum of projection operators of the form $1-P(\bar{t})$ according to Eqs. (2.11) and (2.6b), and employing

$$\mathrm{Tr}_f[1-P(\bar{t})] \equiv \mathrm{Tr}_m[1-P(\bar{t})] \equiv 0, \tag{2.14}$$

we find that

$$P(t)L^0\mathcal{G}(t,t')[1-P(t')] \equiv 0. \tag{2.15}$$

Moreover, the second operator L in Eq. (2.10) can be replaced by L'. With the use of Eq. (2.13) and its analog coupled with

$$\begin{aligned}[1-P(t)]F_r(t) &= [1-\rho(t)\,\mathrm{Tr}_m]F_r(t)\\ &= [1-R(t)\,\mathrm{Tr}_f]F_r(t) = 0,\end{aligned}$$

we obtain

$$[1-P(t')]L_0 F_r(t') = 0. \tag{2.16}$$

When we substitute Eqs. (2.12), (2.15), and (2.16) into the master equation, Eq. (2.10), we obtain

$$\begin{aligned}\dot{F}_r(t) = -iP(t)LF_r(t) - \int_0^t dt'\,P(t)L'\\ \times\mathcal{G}(t,t')[1-P(t')]L'F_r(t').\end{aligned} \tag{2.17}$$

When we express $F_r(t)$ and $P(t)$ in terms of $R(t)$ and $\rho(t)$, use Eqs. (1.2) and (2.3) and the relation $\mathrm{Tr}L' \equiv 0$, we can express Eq. (2.17) as the sum of two parts,

$$R(t)\dot{\rho}(t) + \rho(t)\dot{R}(t) = R(t)\mathcal{O}_m(t) + \rho(t)\mathcal{O}_f(t),$$

where $\mathcal{O}_m$ and $\mathcal{O}_f$ are operators in the subspaces of matter and field density operators, respectively. Operating on this equation with Tr_f yields an equation for $\dot{\rho}(t)$, while operating with Tr_m yields an equation for $\dot{R}(t)$. Thus Eq. (2.17) is equivalent to two coupled master equations for ρ and R,

$$\begin{aligned}\dot{\rho}(t) = -iL_m\rho(t) - i\langle L'\rangle_{f,t}\,\rho(t) - \int_0^t dt'\,\mathrm{Tr}_f L'\\ \times\mathcal{G}(t,t')\Delta_{t'}L'R(t')\rho(t'),\end{aligned} \tag{2.18a}$$

$$\begin{aligned}\dot{R}(t) = -iL_f R(t) - i\langle L'\rangle_{m,t}R(t) - \int_0^t dt'\,\mathrm{Tr}_m L'\\ \times\mathcal{G}(t,t')\Delta_{t'}L'\rho(t')R(t'),\end{aligned} \tag{2.18b}$$

where we have defined

$$\langle L'\rangle_{f,t} \equiv \mathrm{Tr}_f[L'R(t)],\quad \langle L'\rangle_{m,t} \equiv \mathrm{Tr}_m[L'\rho(t)], \tag{2.19}$$

$$\Delta_t L' \equiv L' - \langle L'\rangle_{f,t} - \langle L'\rangle_{m,t}, \tag{2.20}$$

and used

$$[1-P(t')]L'R(t')\rho(t') = \Delta_{t'}L'R(t')\rho(t'). \tag{2.21}$$

Equations (2.18) may be written in a slightly more symmetrical form by replacing the first L' in the integral by $\Delta_t L'$. This replacement[12] is possible by the relations

$$\mathrm{Tr}_f\langle L'\rangle_{m,t} \equiv \mathrm{Tr}_m\langle L'\rangle_{f,t} \equiv 0,$$

$$[\mathrm{Tr}_f, \langle L'\rangle_{f,t}] \equiv [\mathrm{Tr}_m, \langle L'\rangle_{m,t}] = 0,$$

along with Eqs. (2.21) and (2.14). The final form of the general coupled master equations is then

$$\begin{aligned}\dot{\rho}(t) = -iL_m\rho(t) - i\langle L'\rangle_{f,t}\,\rho(t) - \int_0^t dt'\,\mathrm{Tr}_f\Delta_t L'\\ \times\mathcal{G}(t,t')\Delta_{t'}L'R(t')\rho(t'),\end{aligned} \tag{2.22a}$$

$$\begin{aligned}\dot{R}(t) = -iL_f R(t) - i\langle L'\rangle_{m,t}R(t) - \int_0^t dt'\,\mathrm{Tr}_m\Delta_t L'\\ \times\mathcal{G}(t,t')\Delta_{t'}L'R(t')\rho(t').\end{aligned} \tag{2.22b}$$

If Eqs. (2.22) are truncated by neglecting the last term on the right-hand side of each, the equations of the SCFA result. Thus the last term in each equation contains corrections of all orders to the SCF equations brought about through dynamical field-matter correlations.

So far Eqs. (2.22) are formally exact equations for the time evolution of the coupled system and as such are no simpler than the Liouville equation. To make use of Eqs. (2.22), we must approximate the final term of each equation in some suitable fashion. Typical approximations introduced at this stage in the development of master equations are the Born approximation, the Markoff approximation (MA), and the Boltzmann (or low-density) approximation. We note that although the two coupled systems have been treated quite symmetrically so far, it is by no means necessary to continue treating them so, and the approximation introduced need not change both of Eqs. (2.22) in a like manner.

Later in this paper (Secs. IV and V) we will apply Eqs. (2.22) to the treatment of two different quantum-optical problems, where the interacting systems are an optical field and a collection of polarizable atoms. Some of these problems require that we couple either field or atoms to a reservoir in addition to their mutual coupling. Because of this complication, we first show in Sec. III how our formalism may be generalized in a straightforward fashion to the case of N interacting systems and apply this generalization to the treatment of the kinetics of an isolated system, namely an imperfect classical gas. In the low-density limit we obtain the Boltzmann equation for the gas in the form first obtained by Bogoliubov[5] and examine higher-order corrections to it.

III. GENERALIZATION TO N COUPLED SYSTEMS: KINETIC THEORY OF A NONIDEAL GAS

The approach in Sec. II, dealing with two coupled systems by projecting onto uncorrelated states, can be generalized to the case of N coupled systems. We may think of the N coupled systems as being N atoms in an imperfect gas interacting through a two-body force, although the development is much more generally applicable.

In this section we will proceed classically, although it would still be possible to proceed quantum mechanically, as in Secs. I and II. We do this for a twofold purpose: (i) to show that the classical and quantum problems are completely analogous and that techniques developed for the one apply equally to the other and (ii) to make contact with Bogoliubov's derivation[5] of the Boltzmann equation and Klimontovich's corrections to it,[10] in the low-density limit of our general equation.

The Hamiltonian of our system is

$$H_{(N)}=\sum_{\alpha}^{N} H_{\alpha}^{0}+\sum_{\alpha<\beta}^{N} H'_{\alpha\beta}, \tag{3.1}$$

where $H_\alpha^0 \equiv p_\alpha^2/2m$ is the free Hamiltonian of the αth molecule and $H'_{\alpha\beta}\equiv H'(|\vec{q}_\alpha-\vec{q}_\beta|)$ is the two-body interaction energy between molecules α and β. The parameters $\vec{q}_\alpha$ and $\vec{p}_\alpha$ are the center-of-mass position and momentum for the αth molecule, and m is the molecular mass. We assume that the force has a range $r_0 \ll V^{1/3}$, where V is the volume of the system. Corresponding to this Hamiltonian we have a Liouvillian

$$\begin{aligned} L_{(N)} &= L_{(N)}^0 + L'_{(N)}, \\ L_{(N)}^0 &= \sum_{\alpha}^{N} L_\alpha^0, \quad L'_{(N)}=\sum_{\alpha<\beta}^{N} L'_{\alpha\beta}, \end{aligned} \tag{3.2}$$

where

$$\begin{aligned} L_{(N)} &\equiv \{H_{(N)}, \cdots\} \\ &\equiv \sum_{\alpha}^{N}\left(\frac{\partial H_{(N)}}{\partial \vec{q}_\alpha}\cdot\frac{\partial}{\partial\vec{p}_\alpha}-\frac{\partial H_{(N)}}{\partial\vec{p}_\alpha}\cdot\frac{\partial}{\partial\vec{q}_\alpha}\right), \end{aligned} \tag{3.3}$$

and so forth, the braces denoting the Poisson bracket.

In terms of Eqs. (3.2) and (3.3), the Liouville equation is

$$\frac{\partial F_N}{\partial t}=L_{(N)}F_N, \tag{3.4}$$

where F_N is the N-atom probability density function. Since we wish ultimately to make a density expansion, F_N is normalized to V^N rather than to unity,

$$\int d1 \cdots \int dN\, F_N(1,\ldots,N,t)=V^N,$$

or

$$V^{-1}\int d1 \cdots V^{-1}\int dN\, F_N=1, \tag{3.5}$$

where $d1$ represents the sixfold integral $d\vec{q}_1\, d\vec{p}_1$, and so forth. We define reduced probability density functions

$$F_s \equiv V^{-1}\int d(s+1)\cdots V^{-1}\int dN\, F_N, \tag{3.6}$$

normalized to V^s. In particular, the single-molecule probability function is then

$$F_1 \equiv V^{-1}\int d2\cdots V^{-1}\int dN\, F_N \tag{3.7a}$$

and is normalized to V,

$$V^{-1}\int d1\, F_1(1,t)=1. \tag{3.7b}$$

We define a projection operator which projects out of F_N a relevant part corresponding to the uncorrelated SCF probability density function, and, once again, we see that the projection operator is time-dependent,

$$F_N=F_{N,r}(t)+F_{N,i}(t), \tag{3.8}$$

where

$$\begin{aligned} F_{N,r}(t) &= P(t)\,F_N(t) \\ &= \prod_{\alpha}^{N} F_1(\alpha,t), \end{aligned} \tag{3.9}$$

and

$$\begin{aligned} F_{N,i}(t) &= [1-P(t)]F_N(t) \\ &= F_N(t)-\prod_{\alpha}^{N} F_1(\alpha,t). \end{aligned} \tag{3.10}$$

By analogy to Eq. (2.3), the projection operator required is

$$P(t)\equiv\sum_{\alpha}^{N}\prod_{\beta}^{N}{}'\left(F_1(\beta,t)\int\frac{d\beta}{V}\right)-(N-1)\prod_{\alpha}^{N}\left(F_1(\alpha,t)\int\frac{d\alpha}{V}\right),\tag{3.11}$$

the prime on the product meaning that the factor $\beta=\alpha$ is to be omitted. It is easily verified that this expression for $P(t)$ satisfies the properties of time-dependent projectors [Eqs. (2.5)] as well as Eq. (2.7). Moreover, except for normalization, it is strictly analogous to the projector of Eq. (2.3) when $N=2$. As in Sec. II, we find the equation for the relevant part of F_N,[13]

$$\dot{F}_{N,r}(t)=P(t)L_{(N)}\mathcal{G}^{(N)}(t,-\infty)F_{N,i}(-\infty)+P(t)L_{(N)}F_{N,r}(t)+\int_{-\infty}^{t}dt'P(t)L_{(N)}\times\mathcal{G}^{(N)}(t,t')[1-P(t')]L_{(N)}F_{N,r}(t'),\tag{3.12}$$

where

$$\mathcal{G}^{(N)}(t,t')\equiv T\exp\left(\int_{t'}^{t}dt''[1-P(t'')]L_{(N)}\right).\tag{3.13}$$

By integrating over $2,\ldots,N$, using Eqs. (3.7) and (3.9), and neglecting initial correlations, $F_{N,i}(-\infty)=0$, we obtain an equation of motion for F_1,

$$\dot{F}_1(1,t)=\left(L_1^0+(N-1)V^{-1}\int d2\,L'_{12}F_1(2,t)\right)F_1(1,t)+\int_{-\infty}^{t}dt'V^{-1}\int d2\cdots V^{-1}\int dN\,P(t)L_{(N)}\times\mathcal{G}^{(N)}(t,t')[1-P(t')]L_{(N)}\prod_{\alpha}^{N}F_1(\alpha,t').\tag{3.14}$$

In deriving Eq. (3.14) we have used the relations

$$\int d1\,L_1^0\equiv0,\quad\int d1\int d2\,L'_{12}\equiv0,\tag{3.15}$$

the volume integral in phase space reducing to a phase-space surface integral, which vanishes because of the short range of H' and Eq. (3.5), the normalization condition.

At this stage, we may proceed in a parallel fashion to the development in Eqs. (2.13)–(2.22) to obtain a nonlinear equation for $F_1(\alpha,t)$ similar to Eq. (3.14) with $L_{(N)}$ replaced by $\Delta_tL'_{(N)}$ and $\Delta_{t'}L'_{(N)}$ in its first and second occurrences; the fluctuation in $L'_{(N)}$ is defined by

$$\Delta_tL'_{(N)}\equiv\sum_{\alpha<\beta}^{N}\Delta_tL'_{\alpha\beta},\tag{3.16a}$$

$$\Delta_tL'_{\alpha\beta}\equiv L'_{\alpha\beta}-\int\frac{d\alpha'}{V}L'_{\alpha'\beta}F_1(\alpha',t)-\int\frac{d\beta'}{V}L'_{\alpha\beta'}F_1(\beta',t).\tag{3.16b}$$

However, since we wish ultimately to compare our equations with those of Bogoliubov and Klimontovich, we will find it advantageous to proceed somewhat differently. We use the analog of Eq. (2.13),

$$P(t)L_\alpha^0\equiv L_\alpha^0P(t)-L_\alpha^0\sum_{\beta}^{N}{}'\prod_{\gamma}^{N}{}'\left(F_1(\gamma,t)\int\frac{d\gamma}{V}\right)\times\left(1-F_1(\beta,t)\int\frac{d\beta}{V}\right),\tag{3.17}$$

along with Eq. (2.5c) and the relation

$$\prod_{\gamma}^{N}{}'\left(\int\frac{d\gamma}{V}\right)(1-P(\bar{t}))\equiv0,\tag{3.18}$$

to replace L_N by L'_N only in its first appearance in Eq. (3.14). In Eq. (3.17) the prime on the β sum means that the term $\beta=\alpha$ is omitted, and the prime on the γ product means that the factor $\gamma=\beta$ is omitted. Moreover, we may use the symmetry of $L_{(N)}$ under permutation of the molecules to write our general master equation for $\partial F_1/\partial t$,

$$\dot{F}(1,t)=\left(L_1^0+(N-1)V^{-1}\int d2L'_{12}F_1(2,t)\right)F_1(1,t)+(N-1)\int_{-\infty}^{t}dt'V^{-1}\int d2\,L'_{12}V^{-1}\int d3\cdots V^{-1}\times\int dN\,\mathcal{G}^{(N)}(t,t')[1-P(t')]L_{(N)}\prod_{\alpha}^{N}F_1(\alpha,t').\tag{3.19}$$

Before performing a density expansion, to compare with the equations of Bogoliubov and Klimontovich, we perform one additional transformation on Eq. (3.19). Differentiating Eq. (3.13) leads to the equation for $\mathcal{G}^{(N)}$,

$$\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'}=-\mathcal{G}^{(N)}(t,t')[1-P(t')]L_{(N)};\tag{3.20}$$

then Eq. (3.19) can be written

$$\dot{F}_1(1,t)=\left(L_1^0+(N-1)V^{-1}\int d2\,L'_{12}F_1(2,t)\right)F_1(1,t)-(N-1)\int_{-\infty}^{t}dt'\int\frac{d2}{V}L'_{12}\int\frac{d3}{V}\cdots\times\int\frac{dN}{V}\frac{\partial\mathcal{G}^{(N)}(t,t')}{\partial t'}\prod_{\alpha}^{N}F_1(\alpha,t').\tag{3.21}$$

When we integrate the last term of this equation by parts, the integrated part evaluated at $t'=t$ cancels the preceding term in Eq. (3.21)—that is, the SCF term—and the master equation reads

$$\dot{F}_1(1,t)=L_1^0F_1(1,t)+(N-1)\lim_{T\to\infty}\int\frac{d2}{V}L'_{12}\int\frac{d3}{V}\cdots\int\frac{dN}{V}\mathcal{G}^{(N)}(t,t-\tau)\prod_{\alpha}^{N}F_1(\alpha,t-\tau)+(N-1)\int_0^\infty d\tau\int\frac{d2}{V}L'_{12}$$
$$\times\int\frac{d3}{V}\cdots\int\frac{dN}{V}\mathcal{G}^{(N)}(t,t-\tau)\frac{\partial}{\partial t}\prod_{\alpha}^{N}F_1(\alpha,t-\tau) \tag{3.22}$$

after changing the integration variable from t' to $\tau\equiv t-t'$.

To simplify Eq. (3.22) further, we assume we are in the kinetic regime, describing phenomena whose time scale τ satisfies

$$\tau_{\text{int}}\ll\tau\ll\tau_{\text{rel}}\,. \tag{3.23a}$$

Here τ_{int} is the duration of a collision, and τ_{rel} is the time between collisions. Thus $\tau_{\text{int}}=r_0/v$ and $\tau_{\text{rel}}=l/v$, where v is the average molecular speed and $l\cong(4\pi r_0^2N/V)^{-1}$ is the mean free path. We introduce an expansion in powers of the density, or more precisely, powers of $Nr_0^3/V\cong\tau_{\text{int}}/\tau_{\text{rel}}$. Keeping only the lowest order term in the expansion in the limit

$$Nr_0^3/V\ll1 \tag{3.23b}$$

leads to the Boltzmann approximation. The last two terms of Eq. (3.22) are first order in this parameter already since $V^{-1}\int d2\,L'_{12}\sim r_0^3/V$. Now we look at the expression for $\mathcal{G}^{(N)}$, Eq. (3.13). First, any term containing $P(t)L'_{(N)}$ is negligible because it involves expressions like $V^{-1}\sum_\alpha\int d\alpha\,L'_{\alpha\beta}$ and hence is down by an extra factor of Nr_0^3/V. Second, many of the terms in $P(t)L_{(N)}^0$ can also be neglected, since by Eqs. (3.11) and (3.15)

$$P(t)L_\gamma^0=L_\gamma^0\prod_{\beta}^{N}{}'F_1(\beta,t)\int\frac{d\beta}{V}, \tag{3.24}$$

the prime on the product indicating $\beta\neq\gamma$. When we consider that $V^{-(N-2)}\int d3\cdots\int dN$ is at the left of the function $\mathcal{G}^{(N)}$, we may ignore all terms containing L_α^0 unless $\alpha=1$ or 2, and all terms containing $L'_{\alpha\beta}$ unless $(\alpha,\beta)=(1,2)$, according to Eqs. (3.15) and (3.23b). Performing the integral over the coordinates $3,\ldots,N$, we obtain, instead of Eq. (3.22),

$$\dot{F}_1(1,t)=L_1^0F_1(1,t)+\left(\frac{N-1}{V}\right)\lim_{T\to\infty}\int d2\,L'_{12}\mathcal{G}^{(2)}(t,t-\tau)F_1(1,t-\tau)F_1(2,t-\tau)+\left(\frac{N-1}{V}\right)\int_0^\infty d\tau\int d2\,L'_{12}$$
$$\times\mathcal{G}^{(2)}(t,t-\tau)\frac{\partial}{\partial t}F_1(1,t-\tau)F_1(2,t-\tau), \tag{3.25}$$

where[14]

$$\mathcal{G}^{(2)}(t,t-\tau)\equiv T\exp\int_{t-\tau}^{t}dt''\left(L_1^0+L_2^0+L'_{12}-L_1^0F_1(2,t'')\int\frac{d2}{V}-L_2^0F_1(1,t'')\int\frac{d1}{V}\right). \tag{3.26}$$

At this stage, we note that $\mathcal{G}^{(2)}$ is not the usual two-particle time-development operator, which we call $G^{(2)}(\tau)$. From Eq. (3.26) the former satisfies

$$\frac{\partial\mathcal{G}^{(2)}(t,t-\tau)}{\partial\tau}=\mathcal{G}^{(2)}(t,t-\tau)[L_{(2)}-\mathcal{L}_{(2)}(t-\tau)], \tag{3.27}$$

where $L_{(2)}$ is given by Eq. (3.2) with $N=2$ and

$$\mathcal{L}_{(2)}(t-\tau)\equiv L_1^0F_1(2,t-\tau)V^{-1}\int d1$$
$$+L_2^0F_1(1,t-\tau)V^{-1}\int d2; \tag{3.28}$$

on the other hand, the latter satisfies Eq. (3.27) with $\mathcal{L}_{(2)}=0$,

$$\frac{\partial G^{(2)}(\tau)}{\partial\tau}=G^{(2)}(\tau)L_{(2)}, \tag{3.29}$$

whose solution is

$$G^{(2)}(\tau)=e^{(L_1^0+L_2^0+L'_{12})\tau}. \tag{3.30}$$

Equation (3.27) may be transformed into an integral equation which shows clearly the relation between $\mathcal{G}^{(2)}$ and $G^{(2)}$, namely

$$\mathcal{G}^{(2)}(t,t-\tau)=G^{(2)}(\tau)$$
$$-\int_0^T d\tau'\,\mathcal{G}^{(2)}(t,t-\tau')L_{(2)}(t-\tau'). \tag{3.31}$$

Iterating this equation once, one obtains

$$\mathcal{G}^{(2)}(t,t-\tau)=G^{(2)}(\tau)-\int_0^\tau d\tau'\,G^{(2)}(\tau')\times\left(L_1^0F_1(2,t-\tau')V^{-1}\int d1+(1\leftrightarrow 2)\right), \tag{3.32}$$

where $(1\leftrightarrow 2)$ indicates a similar term with 1 and 2 interchanged. Higher-order iterates will not be important, as we will see soon.

We now note that Eq. (3.25), with $\mathcal{G}^{(2)}$ given by Eq. (3.32), still contains a power series in τ/τ_{rel} and $\tau_{\text{int}}/\tau_{\text{rel}}$. Both $\partial F_1(1,t)/\partial t$ and $L_1^0F_1(1,t)$ are $\sim F_1(1,t)/\tau_{\text{rel}}$. Moreover, every $\int d\tau$ leads to an additional power of τ or τ_{int}. Thus Eq. (3.32) may be looked upon as a power series in τ/τ_{rel}. The first term neglected in the equation is $\sim(\tau/\tau_{\text{rel}})^2$. Similarly, the successive terms in the expansion of $F_1(1,t-\tau)$ about $F_1(1,t)$ constitute a power series in τ/τ_{rel}, which we will write later. To implement our series expansion we write Eq. (3.25) for $N>>1$ as

$$\dot{F}_1(1,t)=L_1^0F_1(1,t)+\frac{N}{V}\int d2\,L_{12}'\Psi_{12}(t), \tag{3.33}$$

where

$$\Psi_{12}(t)\equiv\lim_{\tau\to\infty}\mathcal{G}^{(2)}(t,t-\tau)F_1(1,t-\tau)F_1(2,t-\tau)+\int_0^\infty d\tau\,\mathcal{G}^{(2)}(t,t-\tau)\frac{\partial}{\partial t}F_1(1,t-\tau)F_1(2,t-\tau), \tag{3.34}$$

and we note that the first term of Ψ_{12} starts with a zero-order contribution in τ/τ_{rel}, whereas the lowest-order contribution of the second term is already first-order.

We will now show that $\Psi_{12}^{(0)}$, the zero-order term in the expansion of Ψ_{12}, leads to Bogoliubov's form of the Boltzmann equation.[5] Later we will show that the first-order terms $\Psi_{12}^{(1)}$ lead to Klimontovich's corrections.[10] Using only the first term of Eq. (3.32), we have

$$\Psi_{12}(t)\cong\lim_{\tau\to\infty}G^{(2)}(\tau)F_1(1,t-\tau)F_1(2,t-\tau). \tag{3.35}$$

Before expanding F_1 in a Taylor series in Eq. (3.35), we note the effect of $G^{(2)}$ on the coordinates and momenta. Following Bogoliubov, we define the constants[15]

$$\vec{\mathrm{P}}_\alpha^{(2)}(1,2)\equiv\lim_{\tau\to\infty}G^{(2)}(\tau)\vec{\mathrm{p}}_\alpha, \tag{3.36a}$$

$$\vec{\mathrm{Q}}_\alpha^{(2)}(1,2)\equiv\lim_{\tau\to\infty}G^{(2)}(\tau)[\vec{\mathrm{q}}_\alpha+\vec{\mathrm{p}}_\alpha\tau/m], \tag{3.36b}$$

where $\alpha=1,2$. These definitions assume implicitly that the force is sufficiently short range and the density sufficiently low that the colliding particles may be considered to be asymptotically free, an assumption we have already made in Eq. (3.23) above. Thus we can write

$$\lim_{\tau\to\infty}G^{(2)}(\tau)F_1(1,t-\tau)F_1(2,t-\tau)=\lim_{\tau\to\infty}F_1(\vec{\mathrm{Q}}_1^{(2)}-\vec{\mathrm{P}}_1^{(2)}\tau/m,\vec{\mathrm{P}}_1^{(2)},t-\tau)\times F_1(\vec{\mathrm{Q}}_2^{(2)}-\vec{\mathrm{P}}_2^{(2)}\tau/m,\vec{\mathrm{P}}_2^{(2)},t-\tau), \tag{3.37}$$

where m is the molecular mass. Setting $\tau=0$ in the right-hand side of Eq. (3.37) and substituting into Eq. (3.35), we obtain the zero-order part of $\Psi_{12}(t)$,[16]

$$\Psi_{12}^{(0)}(t)=F_1(\vec{\mathrm{Q}}_1^{(2)},\vec{\mathrm{P}}_1^{(2)},t)F_1(\vec{\mathrm{Q}}_2^{(2)},\vec{\mathrm{P}}_2^{(2)},t). \tag{3.38}$$

Substituting Eq. (3.38) into Eq. (3.33) yields exactly Bogoliubov's form of the Boltzmann equation [Eq. (9.17) of Ref. 5]. It differs only in form from the usual spatially inhomogeneous Boltzmann equation, as shown in Bogoliubov's subsequent development.

The next step is to examine corrections to Eq. (3.38) which are first-order in τ/τ_{rel}, that is $\Psi_{12}^{(1)}(t)$. While these terms do not follow from a strict density expansion of the master equation,[17] Klimontovich pointed out that they play an important role in the description of the kinetics of a nonideal gas. In fact, in the case of a spatially homogeneous system, they restore the conservation law for the total energy of the molecular system, in contrast to the usual Boltzmann equation, which conserves only the kinetic energy. To obtain $\Psi_{12}^{(1)}$, we must substitute both terms of Eq. (3.32) into the first term of Eq. (3.34), but only the first term of Eq. (3.32) into the second term of Eq. (3.34). The result is

$$\begin{aligned}\Psi_{12}(t)=&\lim_{\tau\to\infty}F_1(\vec{\mathrm{Q}}_1^{(2)}-\vec{\mathrm{P}}_1^{(2)}\tau/m,\vec{\mathrm{P}}_1^{(2)},t-\tau)\,F_1(\vec{\mathrm{Q}}_2^{(2)}-\vec{\mathrm{P}}_2^{(2)}\tau/m,\vec{\mathrm{P}}_2^{(2)},t-\tau)\\&-\lim_{\tau\to\infty}\int_0^\tau d\tau'\,G^{(2)}(\tau')[L_1^0F_1(1,t-\tau)F_1(2,t-\tau')+(1\leftrightarrow 2)]\\&+\int_0^\infty d\tau\,G^{(2)}(\tau)\frac{\partial}{\partial t}F_1(1,t-\tau)F_1(2,t-\tau),\end{aligned} \tag{3.39}$$

where we have made use of Eq. (3.37). We also wish to expand F_1 in powers of τ/τ_{rel} in Eq. (3.39). In the last two terms we may write $F_1(1,t-\tau)\cong F_1(1,t)$, and so forth, while the first term requires the expansion of F_1 to first order in $\tau/\tau_{\rm rel}$,

$$F_1(\vec{Q}_1^{(2)} - \vec{P}_1^{(2)}\tau/m, \vec{P}_1^{(2)}, t - \tau) \cong \left[1 - \tau\left(\frac{\partial}{\partial t} + \frac{\vec{P}_1^{(2)}}{m}\frac{\partial}{\partial \vec{Q}_1^{(2)}}\right)\right] F_1(\vec{Q}_1^{(2)}, \vec{P}_1^{(2)}, t). \tag{3.40}$$

Performing these expansions and integrating the final two terms of Eqs. (3.39) by parts, the result is

$$\begin{aligned}\Psi_{12}^{(1)}(t) = &-\lim_{\tau\to\infty}\tau\left(\frac{\partial}{\partial t} + \frac{\vec{P}_1^{(2)}}{m}\frac{\partial}{\partial \vec{Q}_1^{(2)}} + \frac{\vec{P}_2^{(2)}}{m}\frac{\partial}{\partial \vec{Q}_2^{(2)}}\right) F_1(\vec{Q}_1^{(2)}, \vec{P}_1^{(2)}, t) F_1(\vec{Q}_2^{(2)}, \vec{P}_2^{(2)}, t) \\ &- \tau G^{(2)}(\tau)(L_1^0 + L_2^0) F_1(1,t) F_1(2,t)\big|_0^\infty + \int_0^\infty d\tau\, \tau \frac{\partial}{\partial \tau} G^{(2)}(\tau)(L_1^0 + L_2^0) F_1(1,t) F_1(2,t) \\ &+ \tau G^{(2)}(\tau) \frac{\partial}{\partial t} F_1(1,t) F_1(2,t)\big|_0^\infty - \int_0^\infty d\tau\, \tau \frac{\partial}{\partial \tau} G^{(2)}(\tau) \frac{\partial}{\partial t} F_1(1,t) F_1(2,t).\end{aligned} \tag{3.41}$$

Since $L_1^0 = -(\vec{p}_1/m)\partial/\partial\vec{q}_1$, we find that the first term of Eq. (3.41) is canceled by the second and fourth terms, when we use Eqs. (3.36) and keep only zero-order terms in $\tau/\tau_{\rm rel}$. The surviving terms of Eq. (3.41) may then be combined to yield

$$\begin{aligned}\Psi_{12}^{(1)}(t) = &-\int_0^\infty d\tau\, \tau \frac{\partial}{\partial \tau} G^{(2)}(\tau)\left(\frac{\partial}{\partial t} - L_1^0 - L_2^0\right) \\ &\times F_1(1,t) F_1(2,t).\end{aligned} \tag{3.42}$$

Substitution of this result into Eq. (3.33) yields a term which can be reduced to Klimontovich's correction term I_2 [Eq. (39) of Ref. 10], the final result being

$$\dot{F}_1(1,t) = L_1^0 F_1(1,t) + \mathcal{I}_1 + \mathcal{I}_2, \tag{3.43a}$$

where

$$\mathcal{I}_1 \equiv \frac{N}{V}\int d2\, L'_{12} F_1(\vec{Q}_1^{(2)}, \vec{P}_1^{(2)}, t) F_1(\vec{Q}_2^{(2)}, \vec{P}_2^{(2)}, t) \tag{3.43b}$$

and

$$\begin{aligned}\mathcal{I}_2 \equiv &-\frac{N}{V}\int d2 \int_0^\infty d\tau\, L'_{12}\, \tau \frac{\partial}{\partial \tau} G^{(2)}(\tau) \\ &\times\left(\frac{\partial}{\partial t} - L_1^0 - L_2^0\right) F_1(1,t) F_1(2,t)\end{aligned} \tag{3.43c}$$

are obtained from $\Psi_{12}^{(0)}(t)$ and $\Psi_{12}^{(1)}(t)$, respectively.[18] The discussion of the way in which the term $\mathcal{I}_2$ affects the conservation laws will not be repeated here, since it is adequately treated in Ref. 10.

In summary, we believe that the derivation of the Boltzmann equation using Bogoliubov's method carried out here provides important new insight into the connection of Bogoliubov's functional techniques with projection-operator methods. Our choice of the relevant part of $F(t)$ to be a product of single-molecule distribution functions in Eq. (3.9), along with our assumption that $F_{N,i}(-\infty) = 0$, is the counterpart of Bogoliubov's asymptotic condition. Moreover, we have shown how our derivation leads naturally to the correction terms of Klimontovich. Here we have proceeded in an entirely different manner than that in which he derived them, which involved truncating the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy through a binary-collision approximation, whereby $F_3(1,2,3,t)$ could be expressed in terms of $F_2(1,2,t)$ and $F_1(1,t)$.

IV. FIRST BORN APPROXIMATION IN QUANTUM OPTICS

One of the ways of simplifying the rigorous Eqs. (2.22) to a more manageable form is to retain only terms linear and quadratic in the interaction L', enabling us to neglect the interaction completely in the propagator $\mathcal{G}(t,t')$. By Eq. (2.11) the unperturbed propagator is

$$\mathcal{G}_0(t,t') = T\exp\left(-i\int_{t'}^t dt''[1 - P(t'')]L^0\right). \tag{4.1}$$

An additional simplification follows from the relation

$$\mathcal{G}_0(t,t')[1 - P(t')] = e^{-iL^0(t-t')}[1 - P(t')]. \tag{4.2}$$

Equation (4.2) follows from Eq. (4.1) by expanding the exponent in a series, using Eq. (2.13) and its analog for $P(t)L_f$ to obtain either $P(t'')$, Tr_m, or Tr_f on the extreme right of any term containing $P(t'')$, and then taking note of Eqs. (2.5c) and (2.14). The resulting approximation is referred to as the first Born Approximation (FBA) with memory.[19,20] In this paper we will not attempt to provide a justification of the FBA with memory,[21] but we will examine the equations of the FBA with memory for the important special case of the quantum-optical Hamiltonian.

Our system consists of a continuum of plane-wave radiation modes $e^{i(\vec{k}\cdot\vec{r}-\Omega_k t)}$ interacting through a dipole interaction with N two-level atoms. The αth atom is at position $\vec{X}_\alpha$ with upper and lower states $|\pm\rangle_\alpha$ separated in energy by $\hbar\omega_0$. In this case we have

$$H_m=\frac{\hbar\omega_0}{2}\sum_{\alpha=1}^{N}s_\alpha\equiv\frac{\hbar\omega_0}{2}S_0,$$

$$H_f=\sum_{k,s}\hbar\Omega_k(a_{ks}^{+}a_{ks}^{-}+\tfrac{1}{2}),\tag{4.3a}$$

$$H'=-\int d\vec{r}\,\vec{P}(\vec{r})\cdot\vec{E}(\vec{r})$$

$$=\hbar\sum_{k,s}(\mu_{ks}P_k^{+}a_{ks}^{-}+\mu_{ks}^{*}P_k^{-}a_{ks}^{+}),\tag{4.3b}$$

where $\vec{P}(\vec{r})$ is the electric-polarization density, $\vec{E}(\vec{r})$ is the electric-field amplitude,

$$s_\alpha\equiv|+\rangle_{\alpha\,\alpha}\langle+|-|-\rangle_{\alpha\,\alpha}\langle-|=[\sigma_\alpha^{+},\sigma_\alpha^{-}],$$

$$\sigma_\alpha^{\pm}\equiv|\pm\rangle_{\alpha\,\alpha}\langle\mp|,\tag{4.4}$$

$$P_k^{\pm}\equiv\sum_\alpha e^{\pm i\vec{k}\cdot\vec{x}_\alpha}\sigma_\alpha^{\pm},\quad S_\kappa\equiv\sum_\alpha e^{i\vec{\kappa}\cdot\vec{x}_\alpha}s_\alpha.\tag{4.5}$$

The operators in Eq. (4.5) satisfy the commutation relations

$$[P_k^{+},P_l^{-}]=S_{k-l},\quad [P_k^{-},S_\kappa]=2P_{k-\kappa}^{-},$$

$$[S_\kappa,P_k^{+}]=2P_{k+\kappa}^{+}.\tag{4.6}$$

In the expression for H_f, $a_{ks}^{\pm}$ is the usual creation (annihilation) operator for the mode with wave vector $\vec{k}$ and polarization index s. In writing the second form of H' we have used the rotating-wave approximation. The coupling constant μ_{ks} is defined by

$$\hbar\mu_{ks}\equiv\vec{\wp}^{*}\cdot\vec{\epsilon}_{ks}(2\pi\hbar\Omega_k/V)^{1/2}.\tag{4.7}$$

where $\vec{\wp}\equiv\langle+|e\vec{r}|-\rangle$ is the transition dipole moment and ϵ_{ks} is the polarization vector of the $(\vec{k},s)$ mode.

When we substitute Eq. (4.3) into Eqs. (2.22), use Eqs. (4.2) and (2.21), apply the commutation relations, Eqs. (4.6), and perform the traces, we obtain

$$\dot{R}+i\sum_l\Omega_l[a_l^{+}a_l^{-},R]+i\sum_l(\mu_l\langle P_l^{+}\rangle[a_l^{-},R]-\text{H.c.})$$

$$=-\sum_{l,m}\mu_l\int_0^t d\tau\,e^{-i(\omega_0-\Omega_m)\tau}\{\mu_m^{*}\langle\Delta P_l^{+}\Delta P_m^{-}\rangle_{t-\tau}[a_l^{-},[a_m^{+},\tilde{R}(t-\tau)]]+\mu_m\langle\Delta P_l^{+}\Delta P_m^{+}\rangle_{t-\tau}[a_l^{-},[a_m^{-},\tilde{R}(t-\tau)]]$$

$$-\mu_m^{*}\langle S_{l-m}\rangle_{t-\tau}[a_m^{+},\Delta a_l^{-}\tilde{R}(t-\tau)]\}+\text{H.c.}\tag{4.8a}$$

and

$$\dot{\rho}+\tfrac{1}{2}i\omega_0[S_0,\rho]+i\sum_l(\mu_l\langle a_l^{-}\rangle[P_l^{+},\rho]-\text{H.c.})$$

$$=\sum_l\int_0^t d\tau\,e^{-i(\omega_0-\Omega_l)\tau}|\mu_l|^2[P_l^{-},\tilde{\rho}(t-\tau)\Delta P_l^{+}]-\sum_{l,m}\int_0^t d\tau\,e^{-i(\omega_0-\Omega_m)\tau}\mu_m$$

$$\times\{\mu_l^{*}\langle\Delta a_l^{+}\Delta a_m^{-}\rangle_{t-\tau}[P_l^{-},[P_m^{+},\tilde{\rho}(t-\tau)]]+\mu_l\langle\Delta a_l^{-}\Delta a_m^{-}\rangle_{t-\tau}[P_l^{+},[P_m^{+},\tilde{\rho}(t-\tau)]]\}+\text{H.c.},\tag{4.8b}$$

where $\langle(\cdots\cdot)\rangle\equiv\text{Tr}_{f,m}(\cdots\cdot)R\rho$, tilde denotes an interaction-picture operator, and $\Delta\Theta\equiv\Theta-\langle\Theta\rangle$.

The left-hand sides of Eqs. (4.8) contain zeroth-order and linear terms in μ and constitute the SCFA, or equivalently the BM equations. On the other hand, the right-hand sides of Eqs. (4.8) are quadratic in μ and contain stimulated absorption and emission terms, as well as spontaneous emission terms. For example, the first two terms of Eq. (4.8a) describe coherent and incoherent spontaneous processes, while the last term describes induced processes. The nonlocal time dependence in Eq. (4.8), which constitutes the memory part of the FBA with memory, is the mathematical representation of the retardation effects present in radiation-matter interactions. In a future publication we will show that Eqs. (4.8) are capable of explaining the experiments in which Skribanowitz *et al.*[22] observed strong coherent spontaneous emission or superradiance from an inverted sample of HF gas. The retardation introduced into the coherent spontaneous emission terms, as well as the presence of stimulated absorption and emission terms, serves to explain the ringing which they observed.

In most quantum-optical problems, in addition to the atom-field coupling, there are usually interactions with matter and or radiation reservoirs which lead to dissipation. The only changes that occur if we add interaction with reservoirs to the system described by the Hamiltonian of Eqs. (4.3) are to add operators $(\delta R/\delta t)_{\text{res}}$ and $(\delta\rho/\partial t)_{\text{res}}$ describing the reservoir damping to the right-hand sides of Eqs. (4.8) and to change the time dependence $e^{i(\omega_0-\Omega_k)\tau}$ in the kernels of the integrals to $e^{[i(\omega_0-\Omega_k)-\nu]\tau}$, where ν^{-1} is the fastest dissipative relaxation time introduced by the reservoirs. However, if the relaxation time due to the reservoir is faster than the relaxation time due to the atom-field coupling treated in the FBA, we obtain the FBA without memory, that is, the FBA in the Markoff approximation. In the Markoff limit Eqs. (4.8) become local in time; that is, all arguments $t-\tau$ in density operators are replaced by the argument t. Furthermore, for $t>\nu^{-1}$ the upper limit in the integral $\int_0^t d\tau\, e^{[-i(\omega_0-\Omega_k)-\nu]\tau}$ is replaced by infinity, and the value of the integral is $[i(\omega_0-\Omega_k)+\nu]^{-1}$. We refer the reader to Ref. 23 for an analysis of the theory in the Markoff limit and a more complete derivation of the master equations.

The only comment we make here is that we obtain the usual superradiance master equation[24] from Eqs. (4.8) in the Markoff limit when the radiation field is required to be in the vacuum state. Then the $\dot{R}$ equation becomes unnecessary, and the SCF terms vanish, as do the induced emission and absorption terms. The resultant superradiance master equation is

$$\dot{\rho}+\tfrac{1}{2}i\omega_0[S_0,\rho]=\sum_l|\mu_l|^2C_l[P_l^-,\tilde{\rho}P_l^+]+\text{H.c.}, \tag{4.9}$$

where

$$C_l=\pi\delta_-(\omega_0-\Omega_l)\equiv\pi\delta(\omega_0-\Omega_l)-i\mathcal{P}(\omega_0-\Omega_l)^{-1}.$$

Bonifacio *et al.*[25] have derived an equation for a collection of atoms coupled to a lossy single-mode field. They required that the field loss rate dominate the atom-field coupling and obtained a theory whose Markoffian limit can be derived from Eq. (4.9) by setting all C_l equal to zero except for the one exactly resonant mode ($\Omega_l=\omega_0$) for which C_l is replaced by ν^{-1}, ν being the loss rate of radiation from the single mode.

Recently, Sasada[26] has shown that the equations of Ref. 25 have interesting non-Markoffian behavior for times short compared with ν^{-1}. This non-Markoffian behavior for times short compared with ν^{-1} should not be confused with the more general long-time, non-Markoffian behavior of the FBA with memory, Eqs. (4.8a) and (4.8b), which persists for times $t>\nu^{-1}$. Since Sasada (as well as Bonifacio *et al.*) uses a single-mode model, he cannot treat the spatially dependent retardation effects that are contained in the multimode Eqs. (4.8a) and (4.8b).

V. PERTURBATION ABOUT THE SELF-CONSISTENT FIELD

In this section we discuss a different approximation to the general master equations for a system consisting of an optical field coupled to polarizable atoms—that is, another special case of Eqs. (2.22) with a Hamiltonian given by Eqs. (2.1) and (4.3). In this approximation we assume that a strong coherent field is present initially, while the atomic population may or may not be inverted. (The situation where the atoms are strongly polarized with no field present initially is also allowable.) In other words, we consider the case where the predominant behavior of the system is described by the SCF or BM equations, while deviations from the SCF equations represent small effects, which we will treat in the FBA.

We can express this approximation quantitatively if we rewrite the Hamiltonian and Liouvillian of Eqs. (2.1) and (2.2) as

$$H=H_m^0(t)+H_f^0(t)+\Delta_t H', \tag{5.1}$$

$$L=L_m^0(t)+L_f^0(t)+\Delta_t L', \tag{5.2}$$

where

$$H_m^0(t)\equiv H_m+\langle H'\rangle_{f,t}, \tag{5.3a}$$

$$H_f^0(t)\equiv H_f+\langle H'\rangle_{m,t}, \tag{5.3b}$$

$$\Delta_t H'\equiv H'-\langle H'\rangle_{f,t}-\langle H'\rangle_{m,t}. \tag{5.3c}$$

The Liouvillians are defined by analogy with Eq. (1.5), and the subscripted angular brackets are defined in Eq. (2.19); the definition of $\Delta_t L'$ following from Eq. (5.3c) agrees with that given in Eq. (2.20). The Hamiltonian $H_m^0+H_f^0$ by itself generates the SCF equations, composed of all but the time-integral terms of Eqs. (2.22). Thus we use the abbreviations

$$\begin{aligned}H_{\text{SCF}}(t)&\equiv H_m^0(t)+H_f^0(t),\\ L_{\text{SCF}}(t)&\equiv L_m^0(t)+L_f^0(t).\end{aligned} \tag{5.4}$$

The approximation we wish to consider is that in which the deviations from behavior described by the SCF are small, that is $\|\Delta L'\|\ll\|L_{\text{SCF}}\|$, in some sense.

We now proceed to implement this approximation in Eqs. (2.22) by expanding them in a Born series and keeping only the first term (FBA). Since the time-integral terms of Eqs. (2.22) are already second-order in $\Delta_t L'$, we may replace $\mathcal{G}$ by

$$\mathcal{G}_{\text{SCF}}(t,t')=T\exp\left(-i\int_{t'}^{t}dt''[1-P(t'')]L_{\text{SCF}}(t'')\right). \tag{5.5}$$

This result can be simplified further by noting that any term with $P(t'')$ in $\mathcal{G}_{\text{SCF}}$ does not contribute. The proof uses Eq. (2.21) and is completely parallel to the proof of Eq. (4.2). Thus we can write

$$\mathcal{G}_{\text{SCF}}(t,t')\Delta_{t'}L'\rho(t')R(t')=G_{\text{SCF}}(t,t')\Delta_{t'}L'\rho(t')R(t'), \tag{5.6}$$

where

$$G_{\rm SCF}(t,t')\equiv T\exp\left(-i\int_{t'}^{t}dt''\,L_{\rm SCF}(t'')\right), \qquad (5.7)$$

and Eqs. (2.22) become in this approximation

$$\dot{\rho}(t)=-iL_m^0(t)\rho(t)-\int_0^t dt'\,{\rm Tr}_f\Delta_t L'G_{\rm SCF}(t,t')$$
$$\times\Delta_{t'}L'R(t')\rho(t'), \qquad (5.8a)$$

$$\dot{R}(t)=-iL_f^0(t)R(t)-\int_0^t dt'\,{\rm Tr}_m\Delta_t L'G_{\rm SCF}(t,t')$$
$$\times\Delta_{t'}L'R(t')\rho(t'). \qquad (5.8b)$$

Note that $G_{\rm SCF}$ operates on matter and field variables separately; that is

$$G_{\rm SCF}(t,t')=G_m^0(t,t')\cdot G_f^0(t,t'), \qquad (5.9a)$$

where

$$G_m^0(t,t')=T\exp\left(-i\int_{t'}^{t}dt''\,L_m^0(t'')\right), \qquad (5.9b)$$

$$G_f^0(t,t')=T\exp\left(-i\int_{t'}^{t}dt''\,L_f^0(t'')\right). \qquad (5.9c)$$

We now consider specifically the model of N two-level atoms coupled to an optical field with Hamiltonian given by Eqs. (4.3). We will further specialize to the case where the atoms interact with a single field mode.[27] In this case the effect of the time-development Liouvillian $G_f^0(t,t')$ on the field operators $a^\pm$ in L' is to cause them to develop from time t' to time t like radiation oscillators driven by a c-number polarization current $\omega_0\langle P^\pm\rangle_t$. However, the driving current in this case is not an external current, but the current determined self-consistently from the solution of the coupled master equations. In like manner, the time-development Liouvillian $G_m^0(t,t')$ acts on the atomic-polarization operators in L' causing them to develop like two-level atoms subject to driving field $\langle a^\pm\rangle_t$, which is, once again, determined self-consistently.

In order to show explicitly the effects of the mean field and mean polarization on the time development, we rewrite the master equations [Eqs. (5.8)] in Hamiltonian form. They take the form

$$\dot{\rho}(t)=-i\hbar^{-1}[H_m^0(t),\rho(t)]+(\delta\rho/\delta t)_{\rm res}-\hbar^{-2}\int_0^t d\tau\,e^{-\nu\tau}$$
$$\times{\rm Tr}_f[\Delta_t H',[\Delta_{t-\tau}H'(t,t-\tau),\tilde{R}(t-\tau)\tilde{\rho}(t-\tau)]], \qquad (5.10)$$

with a similar equation for $\dot{R}(t)$. In Eq. (5.10) we have changed the integration variable to $\tau\equiv t-t'$; we have also included the term $(\delta\rho/\delta t)_{\rm res}$ and the factor $e^{-\nu\tau}$ in the kernel of the integral to allow for the possibility that atoms or field are coupled to a dissipative reservoir, as discussed in Sec. IV. The operators $\tilde{R}(t-\tau)$ are the interaction-picture density operators at time $t-\tau$ corresponding to the Schrödinger-picture $R(t-\tau)$ and $\rho(t-\tau)$, where the two pictures coincide at time t and the unperturbed time-development is due to $H_{\rm SCF}(t)$. Thus we have

$$\tilde{R}(t-\tau)\equiv G_f^0(t,t-\tau)R(t-\tau)$$
$$=U_f^0(t,t-\tau)R(t-\tau)U^{0\dagger}(t,t-\tau), \qquad (5.11a)$$

$$\tilde{\rho}(t-\tau)\equiv G_m^0(t,t-\tau)\rho(t-\tau)$$
$$=U_m^0(t,t-\tau)\rho(t-\tau)U_m^{0\dagger}(t,t-\tau), \qquad (5.11b)$$

where U_f^0 and U_m^0 are the unperturbed time-development operators, satisfying

$$\frac{i\hbar\partial U_f^0(t,t-\tau)}{\partial\tau}=U_f^0(t,t-\tau)H_f^0(t-\tau), \qquad (5.12a)$$

$$\frac{i\hbar\partial U_m^0(t,t-\tau)}{\partial\tau}=U_m^0(t,t-\tau)H_m^0(t-\tau), \qquad (5.12b)$$

$$U_m^0(t,t)=U_f^0(t,t)=1. \qquad (5.12c)$$

Finally, we have defined the effect of $U_{\rm SCF}(t,t')\equiv U_f^0(t,t')U_m^0(t,t')$ on observables in Eq. (5.10) by

$$\Theta(t,t-\tau)\equiv G_{\rm SCF}(t,t-\tau)\Theta$$
$$=U_{\rm SCF}(t,t-\tau)\Theta U_{\rm SCF}^\dagger(t,t-\tau). \qquad (5.13)$$

According to Eqs. (4.3b), (4.5), (2.20), and (1.5), for our single-mode case,

$$\Delta_{t-\tau}H'(t,t-\tau)=\hbar\mu e^{i\vec{k}\cdot\vec{X}_\alpha}\Delta_{t-\tau}\sigma_\alpha^+(t,t-\tau)\Delta a^-(t,t-\tau)$$
$$+{\rm H.c.}+c\text{ number}, \qquad (5.14)$$

the c number being of no consequence since its commutator vanishes. Thus to simplify Eq. (5.10) further we must (i) solve Eq. (5.12a) and use the solution to determine $\Delta_{t-\tau}a^-(t,t-\tau)$ and (ii) solve Eq. (5.12b) and use the solution to determine $\Delta_{t-\tau}\sigma_\alpha^+(t,t-\tau)$. Procedure (i) is simple to carry out, since the interaction with the average polarization only shifts the operator a^- by a c number and this c number cancels out when we subtract the average value of a^-, as required to obtain

$$\Delta_{t-\tau}a^-(t,t-\tau)\equiv a^-(t,t-\tau)-\langle a^-(t,t-\tau)\rangle_{f,t-\tau}.$$

Thus $\Delta_{t-\tau}a^-$ develops in time only due to $H_f=\hbar\Omega(a^+a^-+\frac{1}{2})$, and we have

$$\Delta_{t-\tau}a^-(t,t-\tau)=\Delta_{t-\tau}a^-e^{i\Omega\tau}. \qquad (5.15)$$

The calculation called for in procedure (ii) is slightly more complex. It is most easily carried out in the "rotating frame,"[28] defined by the transformation

$$U_m^0(t,t-\tau)=U_m^R(t,t-\tau)e^{-i\Omega\tau S_0/2}. \tag{5.16}$$

It follows from Eq. (5.12b) and the algebra of the matter operators in Eqs. (4.4)–(4.6) that U_m^R satisfies

$$\frac{\partial U_m^R(t,t-\tau)}{\partial\tau}=-\tfrac{1}{2}iU_m^R(t,t-\tau)\sum_\alpha\{(\omega_0-\Omega)\tau s_\alpha+2[\mu^-\langle a^-\rangle_{t-\tau}e^{-i\Omega\tau}e^{i\vec{k}\cdot\vec{X}_\alpha}\sigma_\alpha^+ +\text{H.c.}]\},$$

whose solution is

$$U_m^R(t,t-\tau)=\prod_\alpha T\exp\{-\tfrac{1}{2}i[(\omega_0-\Omega)\tau s_\alpha-(\Delta\varphi(t,t-\tau)\eta_\alpha(t,\tau)\sigma_\alpha^+ +\text{H.c.})]\}. \tag{5.17}$$

In Eq. (5.17) we have defined

$$\Delta\varphi(t,t-\tau)e^{\pm i\chi(t,t-\tau)}\equiv 2\mu\int_{t-\tau}^{t}dt'\langle\tilde{a}^\pm\rangle_{t'}, \tag{5.18}$$

$\Delta\varphi$ and χ are real, $\langle\tilde{a}^\pm\rangle_t$ is the slowly-varying part of $\langle a^\pm\rangle_t$, where the optical-frequency time dependence has been removed,

$$\langle\tilde{a}^\pm\rangle_t\equiv\langle a^\pm\rangle_t e^{\mp i\Omega t}, \tag{5.19a}$$

and

$$\eta_\alpha(t,\tau)\equiv\exp\{i[\vec{k}\cdot\vec{X}_\alpha-\Omega t-\chi(t,t-\tau)]\} \tag{5.19b}$$

is a phase factor. Carrying along the analogy[28] with paramagnetic resonance, we see that $|\mu\langle\tilde{a}^-\rangle_t|$ is the instantaneous Rabi frequency and $\Delta\varphi(t,t')$ is the turning angle of the two-level atoms, or electric pseudodipoles, from time t' to time t due to the nutation at the Rabi frequency. Making use of the algebra of the matter operators, we may use Eqs. (5.16) and (5.17) to write

$$U_m^0(t,t-\tau)=\prod_\alpha\{\cos\tfrac{1}{2}\psi(t,\tau)-i\psi^{-1}(t,\tau)\sin\tfrac{1}{2}\psi(t,\tau)[\Delta\omega s_\alpha-\Delta\varphi(t,t-\tau)(\eta_\alpha(t,\tau)\sigma_\alpha^+ +\text{H.c.})]\}e^{-(1/2)i\Omega\tau s_\alpha}, \tag{5.20}$$

where

$$\psi(t,\tau)\equiv[\Delta\omega^2\tau^2+\Delta\varphi^2(t,t-\tau)]^{1/2}, \tag{5.21a}$$

$$\Delta\omega\equiv\omega_0-\Omega. \tag{5.21b}$$

From Eqs. (5.13) and (5.20) we find

$$\sigma_\alpha^+(t,t-\tau)=e^{-i\Omega\tau}[G_{++}(t,t-\tau)\sigma_\alpha^+ +G_{+z}(t,t-\tau)\eta_\alpha^*(t,\tau)s_\alpha+G_{+-}(t,t-\tau)\eta_\alpha^{*2}(t,\tau)\sigma_\alpha^-], \tag{5.22}$$

where

$$G_{++}(t,t-\tau)\equiv\cos^2\tfrac{1}{2}\psi(t,\tau)-\psi^{-2}(t,\tau)\Delta\omega^2\tau^2\sin^2\tfrac{1}{2}\psi(t,\tau)-i\psi^{-1}(t,\tau)\Delta\omega\tau\sin\psi(t,\tau), \tag{5.23a}$$

$$G_{+z}(t,t-\tau)\equiv-i\psi^{-1}(t,\tau)\Delta\varphi(t,t-\tau)\sin\tfrac{1}{2}\psi(t,\tau)[\cos\tfrac{1}{2}\psi(t,\tau)-i\Delta\omega\tau\psi^{-1}(t,\tau)\sin\tfrac{1}{2}\psi(t,\tau)], \tag{5.23b}$$

$$G_{+-}(t,t-\tau)\equiv\psi^{-2}(t,\tau)\Delta\varphi^2(t,t-\tau)\sin^2\tfrac{1}{2}\psi(t,\tau). \tag{5.23c}$$

Combining Eqs. (5.10), (5.15), and (5.22), we may write the master equation

$$\begin{aligned}\dot{\rho}(t)=&-i\hbar^{-1}[H_m^0(t),\rho(t)]+(\delta\rho/\delta t)_{\text{res}}\\&-\sum_\alpha\int_0^t d\tau\, e^{-\nu\tau}\,\text{Tr}_f\,[\mu e^{i\vec{k}\cdot\vec{X}_\alpha}\Delta_t\sigma_\alpha^+\Delta_t a^- +\text{H.c.},\\&\qquad\times[\mu e^{i\vec{k}\cdot\vec{X}_\alpha}\{G_{++}(t,t-\tau)\Delta_{t-\tau}\sigma_\alpha^+ +\eta_\alpha^*(t,\tau)G_{+z}(t,t-\tau)\Delta_{t-\tau}s_\alpha\\&\qquad\qquad+\eta_\alpha^{*2}(t,\tau)G_{+-}(t,t-\tau)\Delta_{t-\tau}\sigma_\alpha^-\}\Delta_{t-\tau}a^- +\text{H.c.},\tilde{R}(t-\tau)\tilde{\rho}(t-\tau)]].\end{aligned} \tag{5.24}$$

If we were to perform the indicated trace over field degrees of freedom and compare with Eq. (4.8b), we would find that we have all the terms of Eqs. (4.8) suitably generalized to include reservoir coupling, with $e^{-i(\omega_0-\Omega)\tau}$ replaced by $G_{++}(t,t-\tau)$. In addition, we would have two new sets of terms appearing—namely those arising from the terms containing G_{+z} and G_{+-} in Eqs.

(5.24). We will not carry the solution of the non-Markoffian equation (5.24) and the corresponding equation for $\dot{R}(t)$ any further, our purpose having been only to indicate how our formalism is useful in treating perturbatively fluctuations about a strong SCF. We will treat this problem in more detail elsewhere.

The auxiliary problem in our formalism of determining the time-development of a two-level atom due to the interaction with the SCF is related to a problem which has been studied by Newstein[29] and by Mollow.[30] These authors studied the behavior of a two-level atom subject to a near-resonant *external* driving field. If the envelope of our field were of constant amplitude α and phase ϕ, then we would have

$$\langle \tilde{a}^{\pm} \rangle_t = \alpha e^{\pm i\phi}, \quad \chi(t, t-\tau) = \phi, \tag{5.25}$$

$$\Delta\varphi(t, t-\tau) = 2\mu\alpha\tau, \quad \psi(t, \tau) = Q\tau, \tag{5.26}$$

where $Q \equiv [\Delta\omega^2 + (2\mu\alpha)^2]^{1/2}$. This would enable us to rewrite G_{++}, and so forth, as functions of τ alone; for example,

$$G_{++}(t, t-\tau) \equiv \cos^2 \tfrac{1}{2}Q\tau - Q^{-2}\Delta\omega^2 \sin^2 \tfrac{1}{2}Q\tau - iQ^{-1}\Delta\omega \sin Q\tau. \tag{5.27}$$

Our time-development operator $U_m^0(t, t')$ would then be the same as that of Refs. 29 and 30. They showed that this operator has three poles when Laplace transformed on t; when $\Delta\omega = 0$, one of these poles is at zero frequency and the other two are displaced by $\pm 2\mu\alpha$, the Rabi frequency. These correspond to coherent swinging of the atoms between the upper and lower states at the Rabi frequency during the dissipation time ν^{-1}. Our general problem is more complex in that the field is self-consistently determined and can include, for example, the reaction of the atoms back on the driving field. In addition, our formalism describes, not a single atom, but a collection of N atoms, and can describe spontaneous emission effects $\sim N^2$ (superradiant emission) from a sample driven by an externally applied or self-consistently determined driving field.

As a final point in this section, we note the form assumed by our coupled master equations in the Markoffian limit, where memory effects are strongly suppressed. In this case, where ν is large enough that $\tilde{R}$ and $\tilde{\rho}$ do not change during a time ν^{-1}, we may make the replacements in Eq. (5.24), $\tilde{R}(t-\tau) \to R(t)$, $\tilde{\rho}(t-\tau) \to \rho(t)$, $\Delta_{t-\tau} \to \Delta_t$, and let the limit of the τ integration go to infinity. We may also make the substitutions indicated in Eq. (5.25) where $\alpha = \alpha(t)$ and $\phi = \phi(t)$, since $\langle \tilde{a}^{\pm} \rangle_t$ does not change much during the range of τ which is important. With this interpretation of α, Eq. (5.27) and the corresponding Eqs. for G_{+z} and G_{+-} hold. We may then perform the τ integration in Eq. (5.24) with the result

$$\dot{\rho}(t) = i\hbar^{-1}[H_m^0(t), \rho(t)] + (\delta\rho/\delta t)_{\text{res}} - \sum_\alpha \operatorname{Tr}_f [\mu e^{i\vec{k}\cdot\vec{x}_\alpha} \Delta_t \sigma_\alpha^+ \Delta_t a^- + \text{H.c.}, [\mu e^{i\vec{k}\cdot\vec{x}_\alpha}\{C_{++}(t)\Delta_t \sigma_\alpha^+ + \eta_\alpha^*(t) C_{+z}(t)\Delta_t s_\alpha + \eta_\alpha^{*2}(t) C_{+-}(t)\Delta_t \sigma_\alpha^-\}\Delta_t a^- + \text{H.c.}, R(t)\rho(t)]], \tag{5.28}$$

where

$$\eta_\alpha(t) \equiv e^{i[\vec{k}\cdot\vec{x}_\alpha - \Omega t - \phi(t)]}. \tag{5.29}$$

and

$$C_{++}(t) \equiv \int_0^\infty d\tau\, e^{-\nu\tau} G_{++}(t, t-\tau). \tag{5.30}$$

The quantity $C_{++}(t)$ is the analog of C_l of the FBA without memory of Sec. IV; see, for example, Eq. (4.9) or Eq. (2.34) of Ref. 23. On the other hand, $C_{+z}(t)$ and $C_{+-}(t)$ are quantities without analog in the usual first-Born–Markoff theory. The explicit form of $C_{++}(t)$ may be obtained by substituting Eq. (5.27) into Eq. (5.30), namely

$$C_{++}(t) = \{\Delta\omega^2 + \nu^2 + [2\mu\alpha(t)]^2\}^{-1} \times \{\nu[1 + \tfrac{1}{2}(2\mu\alpha(t)/\nu)^2] + \tfrac{1}{2} i\Delta\}. \tag{5.31}$$

The effect of the strong driving field $\alpha(t)$ may be seen in both the absorptive and dispersive parts of Eq. (5.31).

VI. COMPARISON WITH OTHER WORK

The number of articles using projection-operator techniques to derive kinetic equations is numerous and diverse. However, we will comment on a few of the more recent approaches that serve to put ours in perspective. In the general approach to the treatment of open systems by Haake,[4] the kinetic equation for the system density matrix is obtained by formally eliminating the "bath" degrees of freedom. The time-independent projection operator used is

$$P = B_{\text{ref}} \operatorname{Tr}_{\text{bath}},$$

where B_{ref} is a time-independent operator which is a function of the "bath" coordinates and whose only restriction is that its trace is unity, that is, $\operatorname{Tr}_{\text{bath}} B_{\text{ref}} = 1$. Haake stresses that the formalism is correct for any B_{ref} and the selection of B_{ref} should be based on the physics of the given system and "bath." However, he points out that in cases where the system and "bath" influence each other strongly B_{ref} should be determined self-consistently. If we consider the $R(t)$ of Sec. II as the "bath" density operator and $\rho(t)$ as the system density operator, then the stationary solution of Eq. (2.22b) for R is just the self-consistent determination of B_{ref} that Haake requires in Ref. 4.

In Ref. 7 Robertson derived an exact kinetic equation for the density matrix of an arbitrary system. Recently, Kawasaki[8] generalized and used the results of Ref. 7. Robertson assumes that the density operator is a generalized canonical density operator depending on a finite number of operators whose average values are thermodynamic variables, or that local equilibrium prevails. Our derivation is similar to Robertson's in that both of our projectors are time-dependent operators satisfying Eqs. (2.5) and the form of his equation for $\dot{F}_r$ is the same as ours. Our equations are fundamentally different from each other because our different choices for the breakup of F_N into relevant and irrelevant parts lead to very different projectors $P(t)$. Robertson assumes a generalized canonical form in order to have a description in terms of hydrodynamic and nonequilibrium thermodynamic variables. In Sec. III we assume a product ansatz for the relevant part $F_{N,r}$ of the N-particle density operator F_N in order to have a description of the kinetic stage in terms of the single-particle density operator F_1.

Another recent paper by Ochiai[9] attempts to achieve a description of the kinetic stage. He uses an exact analog of Robertson's projector $P(t)$ as a basis for a general approach to the classical N-body problem by (i) replacing the n macroscopic operators of Robertson by the microscopic operators $\delta(\vec{q}-\vec{q}_\alpha)\delta(\vec{p}-\vec{p}_\alpha)$, one for each particle, and (ii) requiring that the relevant part of F_N should depend on time only through its functional dependence on F_1 instead of specifying the form of the N-particle distribution function as the generalized canonical distribution. Thus Ochiai hoped not only to obtain a kinetic equation for F_1 but also to determine the actual form of $F_{N,r}[1,2,\ldots,N|F_1(t)]$ itself. However Robertson's $P(t)$ was a projector and satisfied Eq. (2.5a) only because he assumed the N-particle distribution function was a generalized canonical distribution. There is no analogous requirement in Ochiai's work and in fact Ochiai's $P(t)$ does not satisfy Eq. (2.5a) and hence is not a projection operator.

In our notation we can write the $P(t)$ of Ochiai as

$$P'(t)=\frac{V}{N}\sum_{\alpha}^{N}\int\frac{d\alpha'}{V}\,\frac{\delta F_{N,r}[1,\ldots,N|F_1(\alpha',t)]}{\delta F_1(\alpha',t)}\prod_{\beta}^{N}{}'\times\int\frac{d\beta'}{V}\,. \tag{6.1}$$

To compare this operator with our projector we assume a product ansatz for $F_{N,r}$, Eq. (3.9), so that

$$\frac{\delta F_{N,r}[1,\ldots,N|F_1(\alpha',t)]}{\delta F_1(\alpha',t)}=\sum_{\alpha}\delta(\alpha'-\alpha)\prod_{\beta}{}'F_1(\beta,t). \tag{6.2}$$

Substituting Eq. (6.2) into Eq. (6.1) yields

$$P'(t)=\sum_{\alpha}\prod_{\beta}{}'\left(F_1(\beta,t)V^{-1}\int d\beta\right). \tag{6.3}$$

Comparing Eq. (6.3) with Eq. (3.11) we see that Ochiai's "projection operator" is only the first term of our projector. Thus even in the case where a product ansatz is assumed for the functional dependence of $F_{N,r}$ on F_1, the operator $P'(t)$ of Ref. 9 is not a projection operator.

In Secs. IV and V we have shown that our approach lends itself naturally to the problem of quantum optics, where the self-consistency of the radiation-matter interaction is important. We were able to obtain previous results for kinetic equations as the limits of our equations, and more importantly we were able to derive new equations which are useful for the investigation of some of the unsolved problems in quantum optics.

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[11] However, it is important to realize that $[1-P(t_1)]P(t_2)\neq 0$, unless $t_1=t_2$.

[12] Of course, the time argument t appearing in $\Delta_t L'$

could be replaced by any other time $\bar{t}$.

[13] It is convenient to choose the initial time to be $t_0 = -\infty$, rather than $t_0 = 0$, as we did in Sec. II.

[14] Strictly, $\mathcal{G}^{(2)}(t, t-\tau)$ should also have L_1^0 replaced by $L_1^0 + L'_{12}$ and L_2^0 by $L_2^0 + L'_{12}$ in the last two terms of the exponent. However, the extra terms are negligible because of the low-density limit.

[15] Reference 5, Eqs. (8.7) and (8.9).

[16] It seems incorrect upon superficial examination to set $\tau = 0$ in Eq. (3.37), since we are taking the limit as $\tau \to \infty$. Upon careful examination, however, we see that setting $\tau = 0$ is allowed because of $\tau \ll \tau_{\text{rel}}$, whereas $\tau \to \infty$ means $\tau \gg \tau_{\text{int}}$. Hence, there is no contradiction here.

[17] Indeed, we have already ignored first-order terms in τ/τ_{rel} in Eqs. (3.23) to (3.26) above.

[18] Klimontovich performs an additional operation by expanding $\vec{q}_2$ about $\vec{q}_1$, the rationale being that because of the presence of L'_{12} acting on Ψ_{12}, a value of $\vec{q}_2$ different from $\vec{q}_1$ would bring in an additional power of $\tau_{\text{int}}/\tau_{\text{rel}}$. His procedure might be described as assuming that the system is only weakly spatially inhomogenous. This procedure produces only a minor change in $\mathcal{I}_2$, consisting of replacing $\vec{q}_2$ by its zero-order value $\vec{q}_1$, since $\mathcal{I}_2$ is already second-order; however, both zero- and first-order terms in $\vec{q}_2 - \vec{q}_1$ are kept in $\mathcal{I}_1$, since $\mathcal{I}_1$ is only first-order. The zero-order term I_1 reduces to the Bogoliubov term for a homogeneous system while the first-order term, which Klimontovich calls I_3, vanishes for a homogenous system. This further operation on the master equation is not necessary for our purposes and the reader is referred to Ref. 10 for details.

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