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Improved Heisenberg equations-of-motion approach for nonequilibrium decay phenomena

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It is pointed out that the usual Heisenberg equations-of-motion or Liouvillian superoperator perturbation-theory approaches to systems with arbitrary statistical ensembles can give spurious, nonconvergent results when attempts are made to calculate the expectation value for certain operators which commute with the zeroth-order Hamiltonian. The non-Markovian, long-time tail of the Weisskopf-Wigner spontaneous-emission decay of a two-level atom is presented as an example of this failure. The general reason for such failures is elucidated and a new method applicable to arbitrary equilibrium or nonequilibrium statistical ensembles is formulated. The new method is applied to the same example of spontaneous emission to derive the correct short- and long-time behavior. The physical model of a two-level atom is discussed and some mathematical techniques of general usefulness are presented. The present results are also briefly discussed in the context of previous work on nonexponential decay. A fully quantum-mechanical definition of time-dependent spectral line shape is given in terms of the response of a two-level—atom detector with variable frequency interacting with electromagnetic radiation. The non-Lorentzian line shape recorded by such a detector for the spontaneous emission of a source two-level atom is calculated as another example of a problem for which the usual methods give incorrect results. The extension of the present method to Liouvillian or master-equation approaches is discussed.

I. INTRODUCTION

The calculation of time-dependent properties of nonequilibrium many-body systems is of considerable current interest. Lasers, plasmas, and chemical reactions are some obvious examples of systems which can be far from equilibrium for which one would like a reliable method for calculating physical properties such as lifetimes of various excitations, energy spectra, and response to external perturbations. Standard temperature-time Green's-function perturbation techniques,[1,2] being dependent on specific properties of thermal equilibrium, are not applicable to systems far from equilibrium, and different methods must be employed. The most common lines of approach are based upon the systematic application of the Heisenberg-operator equations of motion[3] or variations of this,[4,5] such as master equations, using the Liouvillian and Liouville space. Laplace transform techniques figure prominently in these methods since one calculates the dynamics of propagation, to arbitrary times $t > 0$, of correlations which are presumed known at $t = 0$ and not restricted to equilibrium.

A problem can arise when using these Heisenberg equations-of-motion or Liouvillian approaches to calculate higher-order perturbation corrections—the resulting perturbation expansion may not be convergent and can give spurious, unphysical answers. This problem occurs, for example, when calculating the non-Markovian, long-time tail of a decaying two-level atom[6] (Weisskopf-Wigner spontaneous emission), in that the expectation value for the number operator will become negative for certain times. Since this expectation value must be between zero and one, it is clear that the straightforward application of these methods can give erroneous results. It is the purpose of this paper to discuss this problem, to show why it occurs, and to formulate an improved Heisenberg equations-of-motion procedure applicable to pure state, equilibrium, and, most importantly, nonequilibrium systems.

Since the author finds the familiar model of a two-level atom interacting with electromagnetic radiation to be of intrinsic interest, while also providing a fairly simple, yet nontrivial, example of a many-body system which exhibits the aforementioned problems, this paper presents calculations using the two-level-atom example. The usual Heisenberg equations-of-motion approach is applied to the two-level model in Sec. II and shown to give spurious results for the number operator at second order in a perturbation expansion. The reason for this is elucidated and in Sec. III an alternative, improved Heisenberg equations-of-motion approach is formulated and then applied to the same example. The nonexponential decay at both short and long times for realistic quantum systems is a well-known consequence of the Paley-Wiener theorem[7] and we briefly discuss how our results fit into the general context of previous work in this area. In Sec. IV we discuss the question of how to give an operational definition of spectral line shape. We argue that a correct definition can be given in terms of the response of a fully quantum-mechanical two-level atom interacting with the radiation emitted from a fully quantum-mechanical source. We calculate the time-dependent spectral response of a two-level atom with adjustable frequency response, to the spontaneous emission of a source two-level atom as an example of the improved method of Sec. III. Section V is a summary of the paper and also discusses the relationship of this work to that of others. Two appendices present some mathematical formulas and machinery needed in the text.

II. HEISENBERG EQUATIONS-OF-MOTION APPROACH FOR TWO-LEVEL ATOM

In this section we derive the standard results for a two-level atom interacting with electromagnetic radiation and discuss the problem which arises in higher orders in a perturbation expansion. The Hamiltonian $\hat{H}$ of the model is taken to be (dipole approximation and dropping the $\hat{A}^2$ term)

$$\begin{aligned}\hat{H}&=\hat{H}_0+\hat{H}' ,\\ \hat{H}_0&=\kappa\hat{a}^\dagger\hat{a}+\sum_\lambda \lambda\hat{b}_\lambda^\dagger\hat{b}_\lambda ,\\ \hat{H}'&=i\sum_\lambda M_\lambda(\hat{b}_\lambda^\dagger+\hat{b}_\lambda)(\hat{a}-\hat{a}^\dagger) .\end{aligned} \tag{1}$$

Here κ is the energy difference between the unperturbed atomic levels, in units with $\hbar=1$, $\hat{a}$ is a Fermi annihilation operator lowering the atom from its upper to its lower state, and $\hat{a}^\dagger$ is the corresponding creation (raising) operator. The eigenvalue 0 for the Fermi occupation-number operator $\hat{N}=\hat{a}^\dagger\hat{a}$ then implies that the atom is in its lower state; whereas, the eigenvalue 1 implies that it is in its upper state. The $\hat{b}_\lambda$ and $\hat{b}_\lambda^\dagger$ are Bose annihilation and creation operators for photons of wave vector $\vec{k}$, polarization index 1 or 2, and energy $\lambda=\omega_k=|\vec{k}|$ (in units with $c=1$). We use a condensed notation where λ, γ, η, etc., represent both wave vectors and polarization indices, while summations are understood to include sums (or integrals) over wave vectors and polarization sums. The commutation and anticommutation relations are

$$\begin{aligned}&\hat{a}^2=(\hat{a}^\dagger)^2=0,\quad [\hat{a},\hat{a}^\dagger]_+=\hat{1} ,\\ &[\hat{b}_\lambda,\hat{b}_\gamma]_-=[\hat{b}_\lambda^\dagger,\hat{b}_\gamma^\dagger]_-=0 ,\\ &[\hat{b}_\lambda,\hat{b}_\gamma^\dagger]_-=\delta_{\lambda\gamma}\equiv\delta_{\lambda\gamma}\delta_{k_\lambda k_\gamma} ,\\ &[\hat{a},\hat{b}_\lambda]_-=[\hat{a},\hat{b}_\lambda^\dagger]_-=0 .\end{aligned} \tag{2}$$

The photon wave vectors $\vec{k}$ are quantized in the usual way, i.e., a discrete $\vec{k}$-space lattice with lattice constant $2\pi/\Omega^{1/3}$ where Ω is the macroscopic system volume which will be taken to be infinite (this will replace

$$\sum_\lambda \rightarrow (2\pi)^{-3}\Omega\sum_{\text{polarization}}\int d^3k$$

(see Ref. 8). The interaction matrix element is

$$M_\lambda\equiv\left[\frac{2\pi}{\lambda\Omega}\right]^{1/2}\kappa\,\vec{d}\cdot\vec{e}_\lambda , \tag{3}$$

where $\vec{d}$ is the transition dipole matrix element (assumed real for simplicity) and $\vec{e}_\lambda$ are the unit transverse polarization vectors which satisfy

$$\vec{k}\cdot\vec{e}_{\vec{k}\lambda}=0,\quad \vec{e}_{\vec{k}\lambda}\cdot\vec{e}_{\vec{k}\lambda'}=\delta_{\lambda\lambda'} . \tag{4}$$

The Hamiltonian (1) differs from the usual two-level model[9,10] only in that Fermi operators, instead of Pauli spin matrices, are used to describe the two-level atom—only a trivial change of notation ($\sigma_+=\hat{a}^\dagger$, $\sigma_-=\hat{a}$, $\sigma_3=2\hat{a}^\dagger\hat{a}-\hat{1}$) and shift of $\kappa/2$ in the energy origin. The two-level model is also closely related to the well-known Lee[11] and Friedrichs[12] models.

We work in the Heisenberg picture where the operators are time dependent while the state vectors remain fixed. The Heisenberg-operator equations of motion are given by

$$i\frac{\partial}{\partial t}\hat{O}(t)=[\hat{O}(t),\hat{H}]_-\equiv L\hat{O}(t) , \tag{5}$$

which also defines L, the Liouvillian superoperator. We wish to compute $\langle\hat{N}(t)\rangle=\langle\hat{a}^\dagger(t)\hat{a}(t)\rangle$ and

$$\langle\hat{O}(t)\rangle\equiv\mathrm{Tr}[\hat{\rho}\hat{O}(t)] , \tag{6}$$

where $\hat{\rho}$ is the initial ($t=0$) statistical ensemble density operator. Standard perturbation-theory methods for solving for $\hat{O}(t)$ are basically similar in spirit to what follows.[3–6] The Hamiltonian (1) and Eq. (5) give

$$i\frac{\partial}{\partial t}\hat{N}(t)=-i\sum_\lambda M_\lambda(\hat{b}_\lambda+\hat{b}_\lambda^\dagger)(\hat{a}+\hat{a}^\dagger) \tag{7}$$

to which we apply the Laplace transform[13]

$$\begin{aligned}&\mathscr{L}\{\hat{O}(t)\}\equiv[O]_p=\int_0^\infty dt\,e^{-pt}\hat{O}(t) ,\\ &\mathscr{L}^{-1}\{[O]_p\}=\hat{O}(t)=\frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}dp\,e^{pt}[O]_p\end{aligned} \tag{8}$$

obtaining

$$\begin{aligned}p[N]_p=\hat{N}(0)-\sum_\lambda M_\lambda([b_\lambda a]_p+[b_\lambda^\dagger a]_p\\ +[b_\lambda a^\dagger]_p+[b_\lambda^\dagger a^\dagger]_p) .\end{aligned} \tag{9}$$

We next solve for the various $[b_\lambda a]_p$ in (9), again applying the equations of motion (5). This gives

$$[b_\lambda a]_p=\frac{\hat{b}_\lambda(0)\hat{a}(0)}{p+i(\lambda+\kappa)}+\frac{M_\lambda}{p+i(\lambda+\kappa)}([N]_p-[1]_p)+\sum_\gamma\frac{M_\gamma}{p+i(\lambda+\kappa)}(2[b_\gamma b_\lambda N]_p-[b_\gamma b_\lambda]_p+2[b_\gamma^\dagger b_\lambda N]_p-[b_\gamma^\dagger b_\lambda]_p) , \tag{10}$$

$$[b_\lambda a^\dagger]_p=\frac{\hat{b}_\lambda(0)\hat{a}^\dagger(0)}{p+i(\lambda-\kappa)}+\frac{M_\lambda}{p+i(\lambda-\kappa)}[N]_p+\sum_\gamma\frac{M_\gamma}{p+i(\lambda-\kappa)}(2[b_\gamma^\dagger b_\lambda N]_p-[b_\gamma^\dagger b_\lambda]_p+2[b_\gamma b_\lambda N]_p-[b_\gamma b_\lambda]_p) ,$$

where $[1]_p$ is the Laplace transform of the unit operator $\hat{1}$. The corresponding results for $[b_\lambda^\dagger a^\dagger]_p$ and $[b_\lambda^\dagger a]_p$ can be obtained via Hermitian conjugation of (10) with p^* replaced by p. We next insert the transforms (10) into (9) and take the expectation value (6) which gives

$$p\langle[N]_p\rangle=\langle\hat{N}(0)\rangle-\sum_\lambda M_\lambda\left[\frac{\langle\hat{b}_\lambda(0)\hat{a}(0)\rangle}{p+i(\lambda+\kappa)}+\frac{\langle\hat{b}^\dagger_\lambda(0)\hat{a}(0)\rangle}{p-i(\lambda-\kappa)}+\frac{\langle\hat{b}_\lambda(0)\hat{a}^\dagger(0)\rangle}{p+i(\lambda-\kappa)}+\frac{\langle\hat{b}^\dagger_\lambda(0)\hat{a}^\dagger(0)\rangle}{p-i(\lambda+\kappa)}\right]$$
$$-\sum_\lambda M_\lambda^2\left[\langle[N]_p\rangle\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p-i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}+\frac{1}{p-i(\lambda-\kappa)}\right]-\langle[1]_p\rangle\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p-i(\lambda+\kappa)}\right]\right]$$
$$-\sum_{\lambda,\gamma}M_\lambda M_\gamma\left[\left[2\langle[b_\gamma b_\lambda N]_p\rangle-\langle[b_\gamma b_\lambda]_p\rangle+2\langle[b^\dagger_\gamma b_\lambda N]_p\rangle-\langle[b^\dagger_\gamma b_\lambda]_p\rangle\right]\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}\right]\right.$$
$$\left.+\left[2\langle[b^\dagger_\lambda b^\dagger_\gamma N]_p\rangle-\langle[b^\dagger_\lambda b^\dagger_\gamma]_p\rangle+2\langle[b^\dagger_\lambda b_\gamma N]_p\rangle-\langle[b^\dagger_\gamma b_\gamma]_p\rangle\right]\left[\frac{1}{p-i(\lambda+\kappa)}+\frac{1}{p-i(\lambda-\kappa)}\right]\right]. \tag{11}$$

It is clear that the term on the right-hand side of (11) which has $\langle[N]_p\rangle$ in it can be moved to the left-hand side. Our strategy in calculating any operator expectation value is to derive a self-energy representation using the operator equations of motion and Laplace transform. In other words, we want to have[14]

$$\langle[O]_p\rangle=\frac{\langle\hat{O}(0)\rangle}{p+iE_{\hat{O}}+\Sigma_{\hat{O}}(p)}+\cdots \tag{12}$$

where the ellipsis stands for higher-order terms in powers of $|\vec{d}|$ and

$$[\hat{O}(t),\hat{H}_0]_-\equiv L_0\hat{O}(t)\equiv E_{\hat{O}}\hat{O}(t) \tag{13}$$

and $\Sigma_{\hat{O}}(p)$ is the generalized self-energy which will give the line shift and decay of the operator expectation value $\langle\hat{O}(t)\rangle$. If $p+iE_{\hat{O}}+\Sigma_{\hat{O}}(p)$ were to have one simple zero, this would be a pole for $\langle[O]_p\rangle$ and by the inverse Laplace transform (8)

$$\langle\hat{O}(t)\rangle\approx\langle\hat{O}(0)\rangle e^{-E'_{\hat{O}}t-\Gamma_{\hat{O}}t}, \tag{14}$$

where $E'_{\hat{O}}$ and $\Gamma_{\hat{O}}$ are the imaginary and real parts of the simple zero. All four terms in (11) of the form $\langle[bbN]_p\rangle$ can be considered as environmental contributions to $\langle[N]_p\rangle$ which will contribute to both the decay and line shift (in Ref. 4—these are L_0 degenerate with $\hat{N}$); however, they cannot be moved from the right-hand side to the left-hand side of (11) since they are not in the form of a product of Laplace transforms. In order to properly take into account these environmental effects we add to both sides of (11)

$$2\sum_{\lambda,\gamma}M_\lambda M_\gamma\langle\hat{b}_\gamma(0)\hat{b}_\lambda(0)\rangle\langle[N]_p\rangle\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}\right]$$

plus the three other similar terms to obtain

$$\langle[N]_p\rangle=[p+\Sigma_{\hat{N}}(p)]^{-1}\left\{\langle\hat{N}(0)\rangle-\sum_\lambda M_\lambda\left[\frac{\langle\hat{b}_\lambda(0)\hat{a}(0)\rangle}{p+i(\lambda+\kappa)}+\frac{\langle\hat{b}^\dagger_\lambda(0)\hat{a}(0)\rangle}{p-i(\lambda-\kappa)}+\frac{\langle\hat{b}_\lambda(0)\hat{a}^\dagger(0)\rangle}{p+i(\lambda-\kappa)}+\frac{\langle\hat{b}^\dagger_\lambda(0)\hat{a}^\dagger(0)\rangle}{p-i(\lambda-\kappa)}\right]\right.$$
$$+\sum_\lambda M_\lambda^2\langle[1]_p\rangle\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p-i(\lambda+\kappa)}\right]$$
$$-\sum_{\lambda,\gamma}M_\lambda M_\gamma\left[\{2\langle[b_\gamma b_\lambda N]_p\rangle-2\langle\hat{b}_\gamma(0)\hat{b}_\lambda(0)\rangle\langle[N]_p\rangle-\langle[b_\gamma b_\lambda]_p\rangle+2\langle[b^\dagger_\gamma b_\lambda N]_p\rangle\right.$$
$$\left.-2\langle b^\dagger_\gamma(0)\hat{b}_\lambda(0)\rangle\langle[N]_p\rangle-\langle[b^\dagger_\gamma b_\lambda]_p\rangle\}\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}\right]\right.$$
$$\left.+[2\langle[b^\dagger_\gamma b^\dagger_\lambda N]_p\rangle-2\langle\hat{b}^\dagger_\gamma(0)\hat{b}^\dagger_\lambda(0)\rangle\langle[N]_p\rangle-\langle[b^\dagger_\lambda b^\dagger_\gamma]_p\rangle\right.$$

$$+2\langle[b_\lambda^\dagger b_\gamma N]_p\rangle-2\langle\hat{b}_\lambda^\dagger(0)\hat{b}_\gamma(0)\rangle\langle[N]_p\rangle-\langle[b_\lambda^\dagger b_\gamma]_p\rangle]\times\left[\frac{1}{p-i(\lambda+\kappa)}+\frac{1}{p-i(\lambda-\kappa)}\right]\Bigg]\Bigg\}, \tag{15}$$

where the generalized self-energy $\Sigma_{\hat{N}}(p)$ is given by

$$\Sigma_{\hat{N}}(p)\equiv\sum_\lambda M_\lambda^2\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}+\frac{1}{p-i(\lambda+\kappa)}+\frac{1}{p-i(\lambda-\kappa)}\right]$$
$$+2\sum_{\lambda,\gamma}M_\lambda M_\gamma\left[[\langle\hat{b}_\gamma(0)\hat{b}_\lambda(0)\rangle+\langle\hat{b}_\gamma^\dagger(0)\hat{b}_\lambda(0)\rangle]\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}\right]\right.$$
$$\left.+[\langle\hat{b}_\lambda^\dagger(0)\hat{b}_\gamma^\dagger(0)\rangle+\langle\hat{b}_\lambda^\dagger(0)\hat{b}_\gamma(0)\rangle]\left[\frac{1}{p-i(\lambda+\kappa)}+\frac{1}{p-i(\lambda-\kappa)}\right]\right]. \tag{16}$$

In order to take the inverse Laplace transform of (15) we need to have explicit expressions for the Laplace transforms $\langle[b_\gamma b_\lambda N]_p\rangle$, $\langle[b_\gamma b_\lambda]_p\rangle$, etc. It is here that perturbation theory is used. In the general case the Hamiltonian for a given problem will consist of a zeroth-order term $\hat{H}_0$, plus a term $\hat{H}'$, which can be considered small in that it is proportional to some small number.[15] In the case of the two-level atom this small number is the norm of the dipole-matrix element $|\vec{d}|$ which occurs in the M_λ. $\Sigma_{\hat{N}}(p)$ has been given to $O(|\vec{d}|^2)$. To have the numerator of the right-hand side of (15) correct to $O(|\vec{d}|^2)$ we need only the zeroth-order expressions for the Laplace transform terms therein. These are readily computed via the same procedure as was used to arrive at (15), and will give expressions of the form (12). It is clear that the higher-order terms in the perturbation expansion can then be derived iteratively and we will only pursue the calculations to $O(|\vec{d}|^2)$. Equation (15) reduces to

$$\langle[N]_p\rangle^{[2]}=[p+\Sigma_{\hat{N}}(p)]^{-1}\left\{\langle\hat{N}(0)\rangle-\sum_\lambda M_\lambda\left[\left[\frac{\langle\hat{b}_\lambda(0)\hat{a}(0)\rangle}{p+i(\lambda+\kappa)}+\frac{\langle\hat{b}_\lambda^\dagger(0)\hat{a}(0)\rangle}{p-i(\lambda-\kappa)}\right]+\text{c.c. }(p^*\to p)\right]\right.$$
$$+\sum_\lambda M_\lambda^2\frac{\langle[1]_p\rangle}{p}\left[+\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p-i(\lambda+\kappa)}\right]$$
$$-\sum_{\lambda,\gamma}M_\lambda M_\gamma\left[\left[\frac{2\langle\hat{b}_\gamma(0)\hat{b}_\lambda(0)\hat{N}(0)\rangle}{p+i(\lambda+\gamma)+\Sigma_{\hat{N}}(p)}-\frac{2\langle\hat{b}_\gamma(0)\hat{b}_\lambda(0)\rangle\langle\hat{N}(0)\rangle}{p+\Sigma_{\hat{N}}(p)}\right.\right.$$
$$-\frac{\langle\hat{b}_\gamma(0)\hat{b}_\lambda(0)\rangle}{p+i(\lambda+\gamma)}+\frac{2\langle\hat{b}_\gamma^\dagger(0)\hat{b}_\lambda(0)\hat{N}(0)\rangle}{p+i(\lambda-\gamma)+\Sigma_{\hat{N}}(p)}$$
$$\left.\left.-\frac{2\langle\hat{b}_\gamma^\dagger(0)\hat{b}_\lambda(0)\rangle\langle\hat{N}(0)\rangle}{p+\Sigma_{\hat{N}}(p)}-\frac{\langle\hat{b}_\gamma^\dagger(0)\hat{b}_\lambda(0)\rangle}{p+i(\lambda-\gamma)}\right]\right.$$
$$\left.\times\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}\right]+\text{c.c.}(p^*\to p)\right]\Bigg\}, \tag{17}$$

where c.c. means that the complex conjugate of the preceding terms in parentheses is to be taken and then p^* replaced by p. Note that we have replaced $\langle[N]_p\rangle$ on the right-hand side of (15) with its lowest-order value $[p+\Sigma_{\hat{N}}(p)]^{-1}\langle\hat{N}(0)\rangle$.

Since it is the purpose of this paper to discuss the failure of the above procedure for the calculation of the higher-order perturbation corrections, we need only consider the pure state initial condition characteristic of spontaneous emission. The initial-state vector is assumed to be of the Weisskopf-Wigner form

$$|\Psi^{\text{WW}}\rangle\equiv|1\rangle\otimes|\text{vac}\rangle\equiv|1;\text{vac}\rangle\ , \tag{18}$$

which gives

$$\langle\hat{O}(t)\rangle=\langle\Psi^{\text{WW}}|\hat{O}(t)|\Psi^{\text{WW}}\rangle\ .$$

Here $|1\rangle$ indicates that the atom is in its unperturbed upper state while $|\text{vac}\rangle$ is the free-photon vacuum. In this case all of the $\langle[bbN]_p\rangle$ and $\langle[bb]_p\rangle$ terms are zero to lowest order, as are the $\langle\hat{b}(0)\hat{a}(0)\rangle$ terms, and (17) reduces to

$$\langle[N]_p\rangle^{\mathrm{WW}}=[p+\Sigma_{\hat{N}}^{\mathrm{WW}}(p)]^{-1}\left[1+\sum_{\lambda}M_{\lambda}^2\frac{1}{p}\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p-i(\lambda+\kappa)}\right]\right], \tag{19}$$

where WW refers to the Weisskopf-Wigner initial condition (18) and the explicit expression for $\Sigma_{\hat{N}}^{\mathrm{WW}}(p)$ is found upon taking the infinite-volume limit to be

$$\begin{aligned}\Sigma_{\hat{N}}^{\mathrm{WW}}(p)&=\beta\int_0^{\Lambda}d\lambda\,\lambda\left[\frac{1}{p+i(\lambda+\kappa)}+\frac{1}{p-i(\lambda+\kappa)}+\frac{1}{p+i(\lambda-\kappa)}+\frac{1}{p-i(\lambda-\kappa)}\right]\\&\approx i\beta\left[2\kappa\ln\frac{p-i\kappa}{p+i\kappa}+2ip\ln\frac{p^2+\kappa^2}{p^2+\Lambda^2}\right].\end{aligned} \tag{20}$$

We have carried out the angular integrations and defined

$$\beta\equiv\frac{2|\vec{d}|^2\kappa^2}{3\pi}. \tag{21}$$

The high-energy cutoff $\Lambda(\approx m_ec^2)$ is included to avoid divergences due to the two-level and dipole approximations (see discussion of this point in the next section). It is readily ascertained, by writing $p=p_R+ip_I$ and separating $p+\Sigma_{\hat{N}}^{\mathrm{WW}}(p)$ into its real and imaginary parts, that $p+\Sigma_{\hat{N}}^{\mathrm{WW}}(p)$ has simple zeros for

$$p\approx-2\Gamma\equiv-2\kappa\beta\pi \tag{22}$$

and for $p\approx\pm i(\Lambda+\delta)$, $\delta\ll\Lambda$. We ignore these latter two poles in what follows (see discussion in the next section). There is, in addition, a branch cut from $p\approx-i\Lambda$ to $p\approx i\Lambda$ along the imaginary axis. We must therefore perform the inverse Laplace transform according to the contour of Fig. 1. We will not give the details of the calculation here, as an analogous calculation is performed in the next section, but we will just state the result,[3] valid for times $t\gg\kappa^{-1}$:

$$\langle\hat{N}(t)\rangle^{\mathrm{WW}}\approx e^{-2\Gamma t}-\frac{2\Gamma\cos\kappa t}{\pi\kappa^3t^2}+O(t^{-3}). \tag{23}$$

The first term is the standard Weisskopf-Winger natural-lifetime exponential decay, while the second term is the non-Markovian, long-time tail which comes from the branch cut in Fig. 1 and which is expected on general principles as discussed below. We also note that the very-short-time, $t\ll\kappa^{-1}$, behavior is highly nonexponential. As alluded to in the Introduction, it is clear that (23) cannot be a correct description of the decay for times $t\gg(2\Gamma)^{-1}$, since the dominant long-time behavior $-\cos\kappa t/t^2$, will, for certain values of t, give a negative number for $\langle\hat{N}(t)\rangle$. This is impossible because $\langle\hat{N}(t)\rangle$ must be a positive, real number between zero and one. This problem has been pointed out previously in a master equation approach to the same model.[6] It is important that the result (23) also occurs when the "rotating-wave approximation" is made and therefore has nothing to do with "persistent perturbation effects."[16] It is also not an artifact of the high-frequency cutoff since the troublesome term comes from the branch cut and branch cuts of this form will be present in any dissipative system.

The origin of this difficulty can be understood by noting that the procedure used to derive Eq. (19) is not straightforward textbook, perturbation theory—in fact, it is obvious that a naive perturbation calculation will never give exponential decay. To $O(|\vec{d}|^2)$ we would just get $1-2\Gamma t$, i.e., the first terms in the expansion of $e^{-2\Gamma t}$. The primary reason for using the Laplace transform technique was to derive a generalized self-energy $\Sigma_{\hat{O}}(p)$ in order that the inversion would give the exponential. Perturbation theory is then applied as a correction to the exponentially dominant result. This all seems rather obvious, yet it has important implications for calculating the correct answers. Since the long-time behavior of an inverse Laplace transform is dominated by the contributions near the

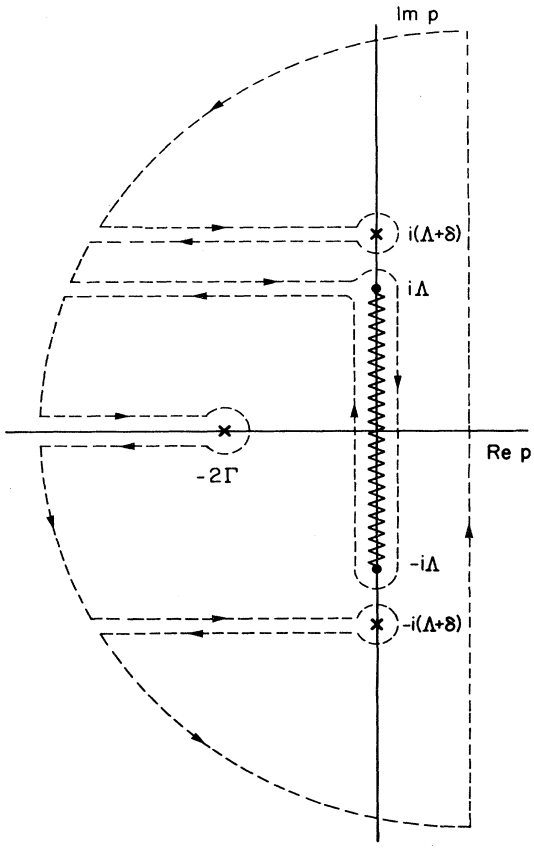

FIG. 1. Contour for inverse Laplace transform for calculation of $\langle\hat{N}(t)\rangle^{\mathrm{WW}}$ using Eq. (19).

imaginary axis (i.e., the branch cut—any terms in the left-hand plane give exponential decay) we see from (19) that for $p \approx 0$ the denominator $[p+\Sigma_n^{WW}(p)]$ is of $O(|\vec{d}|^{-2})$. Therefore, the attempted perturbation expansion is certain not to converge near the origin. This is not too surprising—we know from the theory of Laplace transforms that the inversion of convergent series is often not convergent.[17] This problem of convergence does not occur for operators such as $\hat{a}$ for which $[\hat{O}(t),\hat{H}_0]_- \neq 0$. Near $p=0$ the denominator will look like $[iE_{\hat{O}}+\Sigma_{\hat{O}}(0)]$, which is $O(|\vec{d}|^{0})$, and therefore the perturbation expansion will be reliable. Another way of gaining some insight into this problem is that for $E_{\hat{O}}=0$ there is no natural energy scale with which to compare p. Such a situation often signals the breakdown of a perturbation expansion.

We can conclude that the standard method described in this section will fail to give correct higher-order terms whenever the operator, of which one is calculating the expectation value, is such that

$$L_0\hat{O}(t)=[\hat{O}(t),\hat{H}_0]_-=0\ . \tag{24}$$

This criteria applies to Liouvillian Green's-function approaches and for master equations it implies that the diagonal elements of $\hat{\rho}$ for decaying states will be incorrect. Since this problem only occurs for long times, very short times, and/or higher-order terms, previous calculations restricted to intermediate times and lowest order are correct.

The nonexponential decay at both short and long times found above is a well-known consequence of the Paley-Wiener theorem[7] for Fourier transforms and is not just an artifact of our two-level model. Nonexponential decay is, in fact, a general feature of any quantum system.[18,19] To prove this we need only require a complete orthonormal basis set and that the Hamiltonian have a spectrum which is bounded from below. Nonexponential decay is of obvious relevance to the theory of nuclear decay and the asymptotic time development for partial-wave scattering as well as resonance decay in simplified models has been studied.[20] These calculations use a pure-state Schrödinger approach which, as discussed in the next section, does not have the problem of nonconvergence of the perturbation expansion. Nonexponential decay is also known to be a feature of nonequilibrium statistical mechanics.[21]

There are, at present, many differing views on how to formulate the theory of decaying systems and, in particular, the meaning of nonexponential decay. It is known that the asymptotic behavior of a decaying system explicitly depends upon the initial conditions whereas the exponential term is a characteristic of the particular decay[22] which, however, can depend upon environmental contributions when one has a mixed-state density matrix. For example, the lifetime and level shift of the two-level atom in a radiation bath at nonzero temperature depends explicitly on the temperature.[23] This has led several investigators to try and formulate a theory of decaying states in which a physical state decays only exponentially. In order to do this, the consequences of the Paley-Wigner theorem must be circumvented. Two interesting methods are the rigged Hilbert-space approach to quantum mechanics[22] and the physical particle picture of Prigogine and the Brussels School.[24] The rigged Hilbert-space formulation avoids nonexponential decay by using a different Hilbert space than the usual one (but the nonexponential terms will, in general, show up as background contributions which depend upon the state preparation). The physical particle picture involves a nonunitary ("star unitary") transformation which explicitly breaks time-reversal invariance to relate the usual observables to the new "physical" observables. The reader is referred to the literature for more information on these and related ideas.[22,24,25] Here we shall only point out that the aforementioned problem of having a nonconvergent perturbation expansion is a general feature of any method which uses a Heisenberg equations-of-motion approach or any variation of this and the solution described in the next section is of general applicability.

III. IMPROVED HEISENBERG EQUATIONS-OF-MOTION APPROACH

The solution to the problem we found in the previous section is contained in the observation made there that the Heisenberg equations-of-motion method (and variations) will give a convergent result for operators which do not satisfy (24). In this section we formulate a new method, applicable to arbitrary ensembles, which is based upon this observation and apply the new method to the second-order calculation of $\langle\hat{N}(t)\rangle^{WW}$. The validity of the mathematical model of the two-level atom which we have been using and some interesting calculation techniques are also discussed.

Instead of calculating

$$\langle\hat{N}(t)\rangle^{WW}=\langle\Psi^{WW}|\hat{N}(t)|\Psi^{WW}\rangle$$

as in Sec. II, what we propose to do is first calculate $\hat{a}(t)|\Psi^{WW}\rangle$ using the Heisenberg equations-of-motion approach and then multiply this by its Hermitian conjugate $\langle\Psi^{WW}|\hat{a}^{\dagger}(t)$ in order to get a convergent answer. Since we are multiplying a state vector of modulus less than or equal to one by its Hermitian conjugate, it is clear that the resulting answer will be a real, positive number between zero and one, thereby circumventing the problem of Sec. II.[26] To extend this procedure to arbitrary operators and arbitrary density operators we proceed as follows. In general, for an operator $\hat{O}$ which fulfills Eq. (24), we will be able to write it as a product of two operators $\hat{A}$ and $\hat{B}$, $\hat{O}=\hat{A}\hat{B}$, such that neither $\hat{A}$ nor $\hat{B}$ fulfills (24). If this is not possible, an alternative, but more difficult, method would be to repartition the original Hamiltonian $\hat{H}$ into $\hat{\bar{H}}_0+\hat{\bar{H}}'$ where $[\hat{O}(t),\hat{\bar{H}}_0]_-\neq 0$. However, perturbation theory would then probably not be easily applicable. Assuming that such a product can be found, we wish to compute

$$\langle\hat{O}(t)\rangle=\mathrm{Tr}[\hat{\rho}\hat{O}(t)]=\mathrm{Tr}[\hat{\rho}\hat{A}(t)\hat{B}(t)]\ . \tag{25}$$

From the defining properties of density operators (assuming the usual mathematical apparatus of complete sets, etc.) and in particular since $\hat{\rho}$ is non-negative, we have

$$\hat{\rho}=\sum_n |\Psi_n\rangle\beta_n\langle\Psi_n|$$
$$=\left[\sum_n |\Psi_n\rangle\beta_n^{1/2}\langle\Psi_n|\right]\left[\sum_m |\Psi_m\rangle\beta_m^{1/2}\langle\Psi_m|\right]$$
$$=\hat{\rho}^{1/2}\hat{\rho}^{1/2},\quad (\hat{\rho}^{1/2})^\dagger=\hat{\rho}^{1/2}. \tag{26}$$

Equation (25) can now be rewritten as

$$\mathrm{Tr}[\hat{\rho}\hat{A}(t)\hat{B}(t)]=\sum_i\langle\Psi_i|\hat{\rho}\hat{A}(t)\hat{B}(t)|\Psi_i\rangle$$
$$=\sum_{j,k}\langle\Psi_j|\hat{\rho}^{1/2}\hat{A}(t)|\Psi_k\rangle$$
$$\times\langle\Psi_k|\hat{B}(t)\hat{\rho}^{1/2}|\Psi_j\rangle\,, \tag{27}$$

which is applicable to any initial-condition density operator $\hat{\rho}$. A simple example is that of equilibrium at temperature T where

$$\hat{\rho}^{1/2}=e^{-\hat{H}/2k_BT}/(\mathrm{Tr}e^{-\hat{H}/k_BT})^{1/2}\,.$$

The matrix elements in (27) can be computed using an obvious variation of the Heisenberg equations-of-motion approach of Sec. II, and then the resulting sums (integrals) over the intermediate states computed. The method embodied in Eq. (27) is new, as far as the author is aware, and should be of general applicability to many-body systems.

For $\hat{O}(t)=\hat{N}(t)$ we have

$$\langle\hat{N}(t)\rangle=\sum_{j,k}|\langle\Psi_j|\hat{a}(t)\hat{\rho}^{1/2}|\Psi_k|^2$$

and for the Weisskopf-Wigner state (18) this reduces to

$$\langle\hat{N}(t)\rangle^{\mathrm{WW}}=\sum_j|\langle\Psi_j|\hat{a}(t)|\Psi^{\mathrm{WW}}\rangle|^2$$

which is obviously the same as calculating $\langle\Psi^{\mathrm{WW}}|\hat{a}^\dagger(t)\hat{a}(t)|\Psi^{\mathrm{WW}}\rangle$ as proposed earlier. Using the equations-of-motion twice and taking the Laplace transform gives

$$[a]_p|\Psi^{\mathrm{WW}}\rangle=[p+i\kappa+\Sigma_{\hat{a}}^{\mathrm{WW}}(p)]^{-1}$$
$$\times\left[|0;\mathrm{vac}\rangle+\sum_\lambda\frac{2M_\lambda}{p-i\lambda}|1;\lambda\rangle+O(|\vec{d}|^2)\right], \tag{28}$$

where $|1;\lambda\rangle=\hat{a}^\dagger|0\rangle\otimes\hat{b}_\lambda^\dagger|\mathrm{vac}\rangle$ and

$$\Sigma_{\hat{a}}^{\mathrm{WW}}(p)=2\beta p\int_0^\Lambda d\lambda\frac{\lambda}{\lambda^2+p^2}$$
$$=p\beta[\ln(p+i\Lambda)+\ln(p-i\Lambda)-2\ln p]\,. \tag{29}$$

The $O(|\vec{d}|^2)$ term is orthogonal to $\langle 0;\mathrm{vac}|$ and does not contribute at second order. It is easily verified that $p+i\kappa+\Sigma_{\hat{a}}^{\mathrm{WW}}(p)$ has three branch cuts, $p=\pm i\Lambda-x$ and $p=-x$ for $x=0\to\infty$, and three poles: one which gives decay

$$p_D\approx-i(\kappa-\Delta)-\Gamma,\quad \Delta\equiv2\kappa\beta\ln\frac{\Lambda}{\kappa},\ \Gamma\equiv\kappa\beta\pi \tag{30}$$

and two on the imaginary axis

$$P_\pm\approx\pm i\Lambda(1+\tfrac{1}{2}e^{-1/\beta})\,. \tag{31}$$

The inversion integral is given by the contour of Fig. 2. Δ, as has been previously noted, is an improved version of the Lamb shift which is only logarithmically divergent.[3,27] If we had not included the "counter-rotating-wave" terms $\hat{b}\hat{a}$ and $\hat{b}^\dagger\hat{a}^\dagger$ in the Hamiltonian (1), Δ would have been linearly divergent and a second decaying pole proportional to Λ could have arisen for certain values of the parameters.[28] The two imaginary poles $p_\pm$ give rise to a nondecaying, nonergodic contribution to the decay[28,29] which is an artifact of the dipole approximation and use of a high-frequency cutoff Λ. In a more realistic model of a decaying system (the hydrogen atom, for example), the interaction matrix element M_λ in (3) is replaced by a Yukawa-type form factor which provides the necessary high-frequency convergence and there are no poles on the imaginary axis.[29] The two-level model is sometimes criticized as being unphysical due to the occurrence of the $p_\pm$ poles. A simple computation of the residue at these poles gives $R_\pm\approx e^{i\Lambda t}e^{-1/\beta}/2\beta$. Using reasonable values for lifetimes and frequencies appropriate to excited atomic states gives $\beta\approx10^{-8}$ and $R_\pm\approx e^{-10^8}=0$ which shows that these pole contributions are completely negligible. The branch cuts, on the other hand, arise in any model of a decaying system and are not an artifact of our approximations.[30] They influence not only the long-time behavior, but also, as will be shown below, the very-short-time behavior.

Instead of actually computing the inverse Laplace transform via the contour of Fig. 2, it is simpler to rewrite (28) using

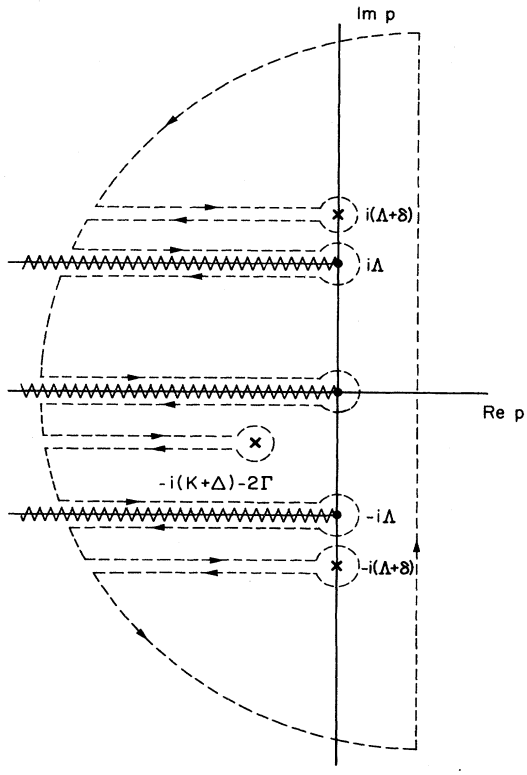

FIG. 2. Contour for inverse Laplace transform for calculation of $[a]_p|\psi^{\mathrm{WW}}\rangle$, Eq. (28).

$$[p+i\kappa+\Sigma_{\hat{a}}^{\mathrm{WW}}(p)]^{-1}=\{p+i(\kappa-\Delta)+\Gamma+[\Sigma_{\hat{a}}^{\mathrm{WW}}(p)+i\Delta-\Gamma]\}^{-1}$$
$$\approx\frac{1}{p+i\kappa'+\Gamma}-\left[\frac{\Sigma_{\hat{a}}^{\mathrm{WW}}(p)+i\Delta-\Gamma}{(p+i\kappa'+\Gamma)^2}\right]+\cdots , \tag{32}$$

where $\kappa'\equiv\kappa-\Delta$. This is justified since we have shown that the poles $p_\pm$ are ignorable—the expansion (32) only picks out the pole p_D and the three branch cuts. The necessary inverse Laplace transforms can now be computed exactly to the order desired.

For the term in (28) with $|0;\mathrm{vac}\rangle$ we need

$$\mathscr{L}^{-1}\left[\frac{1}{p-p_D}+\frac{\Gamma}{(p-p_D)^2}-\frac{i\Delta}{(p-p_D)^2}-\frac{\Sigma_{\hat{a}}^{\mathrm{WW}}(p)}{(p-p_D)^2}\right] . \tag{33}$$

It is easily verified that this gives

$$e^{p_Dt}+(\Gamma-i\Delta)te^{p_Dt}-(te^{p_Dt})*\mathscr{L}^{-1}[\Sigma_{\hat{a}}^{\mathrm{WW}}(p)] , \tag{34}$$

where

$$\mathscr{L}^{-1}[\Sigma_{\hat{a}}^{\mathrm{WW}}(p)]=\frac{2\beta}{t^2}(\cos\Lambda t+\Lambda t\sin\Lambda t-1) \tag{35}$$

and $*$ in (34) means to take the Laplace convolution. Evaluating the necessary integrals as in Appendix A and some tedious algebra gives for the convolution term in (34)

$$-2\beta\left[1-tp_De^{p_Dt}E_1(p_Dt)-\cos\Lambda t+\frac{t}{2}e^{p_Dt}[(p_D+i\Lambda)E_1((p_D+i\Lambda)t)+(p_D-i\Lambda)E_1((p_D-i\Lambda)t)]\right.$$
$$-\frac{i\Lambda}{2}te^{p_Dt}[E_1((p_D+i\Lambda)t)-E_1((p_D-i\Lambda)t)]+\frac{p_Dt}{2}e^{p_Dt}\ln\frac{P_D^2+\Lambda^2}{p_D^2}-e^{p_Dt}E_1(p_Dt)$$
$$\left.+\frac{e^{p_Dt}}{2}[E_1((p_D+i\Lambda)t)+E_1((p_D-i\Lambda)t)]+\frac{\Lambda}{\Lambda^2+p_D^2}(\Lambda\cos\Lambda t+p_D\sin\Lambda t)-\frac{\Lambda^2e^{p_Dt}}{\Lambda^2+p_D^2}+e^{p_Dt}\ln\frac{P_D^2+\Lambda^2}{P_D^2}\right] , \tag{36}$$

where $E_1(z)$ is the exponential integral given in Appendix A. Remember that (36) will be added to the other terms in (34) and then multiplied by the complex conjugate. For $t=0$, (36) is zero, while for times $t\ll\Lambda^{-1}$, (36) is highly oscillatory with every term being important. For $\Lambda^{-1}\ll t\ll\kappa^{-1}$ the terms with Λ in them either cancel or go as powers of $(\Lambda t)^{-1}\ll 1$ and can be dropped. Once $t\gg\kappa^{-1}$ we can use the asymptotic expansion for $E_1(z)$, (A3), to express (36) as

$$-\frac{2\beta}{\kappa^2t^2}-\Gamma te^{p_Dt}+i\Delta te^{p_Dt}-\left[\frac{i\Gamma}{\kappa}+\frac{\Delta}{\kappa}\right]e^{p_Dt} . \tag{37}$$

Returning to the second term in (28) we have

$$\mathscr{L}^{-1}\left[2\sum_\lambda\frac{M_\lambda}{(p-i\lambda)(p-p_D)}\,|\,1;\lambda\rangle\right]=2\sum_\lambda M_\lambda\left[\frac{e^{i\lambda t}}{i\lambda-p_D}+\frac{e^{p_Dt}}{p_D-i\lambda}\right]|\,1;\lambda\rangle . \tag{38}$$

The second-order contribution to $\langle\hat{N}(t)\rangle^{\mathrm{WW}}$ resulting from (38) is

$$\beta\int_0^\Lambda d\lambda\,\lambda\left[\frac{-ie^{i\lambda t}}{\lambda+ip_D}+\frac{ie^{p_Dt}}{\lambda+ip_D}\right]\left[\frac{ie^{-i\lambda t}}{\lambda-ip_D^*}-\frac{ie^{p_D^*t}}{\lambda-ip_D^*}\right]$$
$$=(1+e^{-2\Gamma t})\beta\ln\frac{\Lambda}{\kappa}+\frac{\beta}{2\Gamma}\{p_D^*e^{-2\Gamma t}[E_1(p_D^*t)-E_1(it(-ip_D^*+\Lambda))]+p_De^{-2\Gamma t}[E_1(p_Dt)-E_1(-it(ip_D+\Lambda))]$$
$$+p_D^*[E_1(-p_D^*t)-E_1(-it(-ip_D^*+\Lambda))]+p_D[E_1(-p_Dt)-E_1(it(ip_D+\Lambda))]\} , \tag{39}$$

which is zero for $t=0$, while for $t\gg\kappa^{-1}$ gives

$$(1+e^{-2\Gamma t})\beta\ln\frac{\Lambda}{\kappa}+\frac{2\beta e^{-\Gamma t}\cos\kappa' t}{t^2(\kappa'^2+\Gamma^2)} . \tag{40}$$

Combining (34), (37), and (40) we have for $t\gg\kappa^{-1}$

$$\langle\hat{N}(t)\rangle^{\mathrm{WW}}\approx\frac{\Delta}{2\kappa}+e^{-2\Gamma t}\left[1-\frac{3\Delta}{2\kappa}\right]$$
$$-\frac{2\Gamma e^{-\Gamma t}\cos\kappa' t}{t^2\kappa^3\pi}+\frac{4\Gamma^2}{\kappa^6t^4} . \tag{41}$$

The last term in (41) is $O(|\vec{d}|^4)$, and since we have not included other contributions to this order, it does not represent the true $O(|\vec{d}|^4)$ contribution.

It is interesting that the improved answer (41) has the same $\cos(\kappa t)/t^2$ dependence as did (23) except modified by $e^{-\Gamma t}$. The result (23) also did not include the line shift Δ in $\cos\kappa t$; whereas (41) has $\cos(\kappa-\Delta)t$. We can conjecture that the method in Sec. II picked out just the first term in the expansions of $e^{-\Gamma t}$ and $\cos\kappa' t$, and that higher-order terms in the perturbation theory of Sec. II would give the higher-order terms in the expansions of $e^{-\Gamma t}$ and $\cos\kappa' t$. It is important to notice that as $t\to\infty$, $\langle\hat{N}(t)\rangle^{WW}$ does not go to zero, but rather it approaches $\Delta/2\kappa$. The reason for this (not usually mentioned in the literature because the rotating-wave approximation is made) is easily ascertained. $\hat{N}(t)$ does not have expectation value zero because the interacting ground state for the Hamiltonian (1) is not the product of the two free-field ground states: $|0\rangle\otimes|\text{vac}\rangle$. The excited state (18) will decay away to the true ground state which, using standard perturbation techniques,[31] is

$$|\text{ground state}\rangle = |0;\text{vac}\rangle + i\sum_{\lambda}\frac{M_\lambda}{\lambda+\kappa}|1;\lambda\rangle + O(|\vec{d}|^2) \tag{42}$$

and $\langle\hat{N}\rangle=\Delta/2\kappa$ in this state. It is doubtful that the correction terms to the purely exponential decay of (41) are measurable in that they do not become important for excited atomic states until times on the order of 50 lifetimes.[29] It is possible that such correction terms are observable for other many-body systems and it is important that the present method gives convergent results. Also, the calculation in the next section is an example of a problem where the incorrect result using the method of Sec. II is apparent in the lowest relevant order.

IV. SPECTRAL LINE SHAPE DEFINED BY TWO-LEVEL–ATOM DETECTOR

Standard definitions[1,32–34] of spectral lineshape are usually given in terms of current-current correlation functions $\langle\vec{J}(\vec{r},t)\vec{J}(\vec{r}',t')\rangle$ or dipole autocorrelation functions $\langle\vec{d}(t)\vec{d}(t')\rangle$ and derived using assumptions which are, in general, of questionable validity. Perturbation theory in the form of "Fermi's Golden Rule" is used and the coupling between radiation sources and the electromagnetic field ignored.[35,36] These definitions are, at best, semiclassical, and it is important to investigate their applicability.[37] Any correct derivation of a general definition for line shape must certainly include a fully quantum-mechanical and qualitatively reasonable model for the process of detection.

A particularly useful and fairly simple model for a detector is that of a two-level atom which interacts with radiation as in the Hamiltonian (1).[38,39] We measure the degree of resonant excitation of the detector by allowing it to have an adjustable frequency ν. For a completely quantum-mechanical calculation, the source of radiation must also be explicitly included in the total Hamiltonian. In this section we calculate the line shape defined by the time-dependent spectral response of a variable-frequency, two-level atom detector to the spontaneous emission of a two-level source atom. The sort of experimental situation we have in mind is that of photon-counting or microwave detection by a variable-Q cavity. One could easily generalize the following to include a reservoir of two-level atom detectors with the corresponding increase in difficulty of calculation.

The Hamiltonian for our model is

$$\begin{aligned}\hat{H}&=\hat{H}_0+\hat{H}',\\ \hat{H}_0&=\nu\hat{d}^\dagger\hat{d}+\kappa\hat{a}^\dagger\hat{a}+\sum_\gamma\gamma\hat{b}_\gamma^\dagger\hat{b}_\gamma,\\ \hat{H}'&=i\sum_\gamma M_\gamma(\hat{b}_\gamma+\hat{b}_\gamma^\dagger)(\hat{a}-\hat{a}^\dagger)\\ &\quad+i\sum_\gamma D_\gamma(\hat{b}_\gamma e^{-ik_\gamma r_0}+\hat{b}_\gamma^\dagger e^{ik_\gamma r_0})(\hat{d}-\hat{d}^\dagger),\end{aligned} \tag{43}$$

where $\hat{d}$ and $\hat{d}^\dagger$ are the Fermi lowering and raising operators for the detector which satisfy

$$\begin{aligned}&[\hat{d},\hat{d}^\dagger]_+=\hat{1},\quad \hat{d}^2=(\hat{d}^\dagger)^2=0,\\ &[\hat{d},\hat{a}]_-=[\hat{d}^\dagger,\hat{a}]_-=[\hat{d},\hat{b}_\gamma]_-\\ &\qquad=[\hat{d},\hat{b}_\gamma^\dagger]_-=0,\end{aligned} \tag{44}$$

ν is the variable frequency of the detector which is assumed located at $\vec{r}_0$ with the source atom at the origin, and

$$D_\gamma\equiv\left[\frac{2\pi}{\gamma\Omega}\right]^{1/2}\nu\hat{d}'\cdot\vec{e}_{\vec{k}\gamma}, \tag{45}$$

where $\vec{d}'$ is the detector transition dipole-matrix element (assumed real). In order to simplify the calculation we shall assume, as described in Appendix B, that the detector is along the z axis, in the radiation zone, and that the dipole-matrix elements of both the source and detector atoms have components only in the x direction. We define the spectral line shape to be $\langle\hat{N}_D(t)\rangle=\langle\hat{d}^\dagger(t)\hat{d}(t)\rangle$ which is the probability that the detector atom is in its excited state at time t. We shall choose the initial condition such that the detector is in its ground state at $t=0$.

Since $[\hat{N}_D(t),\hat{H}_0]_-=0$ we have the case discussed in the previous two sections and if one calculates $\langle\hat{N}_D(t)\rangle$ by the incorrect method of Sec. II, the resulting answer has the same problem discussed there of becoming negative for certain large times. We therefore proceed as in Sec. III to calculate $\hat{d}(t)|\psi^{WW}\rangle$ where $|\psi^{WW}\rangle$ is now defined to be

$$|\psi^{WW}\rangle\equiv|0_D;1_S;\text{vac}\rangle\equiv|0_D\rangle\otimes|1_S\rangle\otimes|\text{vac}\rangle, \tag{46}$$

where the subscripts D and S refer to the detector and source atoms, respectively. It is clear that since we are describing the detector fully quantum mechanically, the detector atom will have a natural lifetime and will therefore decay. This decay must be assumed to be much slower than that of the source atom in order to properly describe the detection process. We therefore assume that $|\vec{d}'|\ll|\vec{d}|$ and keep only the lowest relevant order in powers of $|\vec{d}'|$ in the subsequent calculation.

Applying the Heisenberg operator equations of motion (5) twice, using the Hamiltonian (43), and taking the Laplace transform (8) gives

$$\left[p+i\nu+\sum_\gamma D_\gamma^2\left[\frac{1}{p+i\gamma}+\frac{1}{p-i\gamma}\right]\right][d]_p$$
$$=\hat{d}(0)+\sum_\gamma D_\gamma\left[\frac{e^{-ik_\gamma r_0}}{p+i\gamma}[2\hat{b}_\gamma(0)\hat{N}_D(0)-\hat{b}_\gamma(0)]+\text{H.c.}(p^*\to p)\right]$$
$$+\sum_\gamma D_\gamma M_\gamma\left[\frac{e^{-ik_\gamma r_0}}{p+i\gamma}(2[aN_D]_p-2[a^\dagger N_D]_p+[a^\dagger]_p-[a]_p)+\text{H.c.}(p^*\to p)\right]. \tag{47}$$

It is clear from (47) that we need self-energy representations (12) for $[aN_D]_p$, $[a^\dagger N_D]_p$, $[a^\dagger]_p$, and $[a]_p$; however, $[aN_D]_p\,|\,\psi^{\text{WW}}\rangle$ and $[a^\dagger N_D]_p\,|\,\psi^{\text{WW}}\rangle$ are $O(|\vec{d}|)$ and can be dropped. $[a^\dagger]_p\,|\,\psi^{\text{WW}}\rangle$ and $[a]_p\,|\,\psi^{\text{WW}}\rangle$ are given by [see (28)]

$$[a]_p\,|\,\psi^{\text{WW}}\rangle\approx\frac{|\,0_D;0_S;\text{vac}\rangle}{p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)}+\sum_\lambda M_\lambda\frac{|\,0_D;1_S;\lambda\rangle}{(p-i\lambda)[p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)]},$$
$$[a^\dagger]_p\,|\,\psi^{\text{WW}}\rangle\approx\sum_\lambda M_\lambda\frac{|\,0_D;1_S;\lambda\rangle}{(p-i\lambda)[p+i\kappa+\Sigma_{\hat{a}^\dagger}^{\text{WW}}(p)]}, \tag{48}$$

where

$$\Sigma_{\hat{a}^\dagger}^{\text{WW}}(p)=[\Sigma_{\hat{a}}^{\text{WW}}(p)]^*\,|_{p^*\to p}=\Sigma_{\hat{a}}^{\text{WW}}(p)$$

by (29). Equations (47) and (48) give

$$[p+i\nu+\Sigma_{\hat{d}}^{\text{WW}}(p)][d]_p\,|\,\psi^{\text{WW}}\rangle$$
$$\approx-\sum_\gamma D_\gamma\frac{e^{-ik_\gamma r_0}}{p-i\gamma}|\,0_D;1_S;\gamma\rangle+\sum_\gamma D_\gamma M_\gamma\left[\frac{e^{ik_\gamma r_0}|\,0_D;0_S;\text{vac}\rangle}{(p-i\gamma)[p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)]}-\frac{e^{-ik_\gamma r_0}|\,0_D;0_S;\text{vac}\rangle}{(p+i\gamma)[p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)]}\right]$$
$$+\sum_{\lambda,\gamma}D_\lambda M_\lambda M_\gamma\left[\frac{e^{-ik_\lambda r_0}}{(p+i\lambda)(p-i\gamma)}\left[\frac{1}{p-i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)}-\frac{1}{p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)}\right]|\,0_D;1_S;\gamma\rangle\right.$$
$$\left.+\frac{e^{ik_\lambda r_0}}{(p-i\lambda)(p-i\gamma)}\left[\frac{1}{p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)}-\frac{1}{p-i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)}\right]|\,0_D;1_S;\gamma\rangle\right]. \tag{49}$$

We shall make a further simplification in that we will only calculate the lowest-order contributions to $\langle\hat{N}_D(t)\rangle^{\text{WW}}$ and basically ignore the highly oscillatory, short-time behavior. This allows us to replace $[p+i\nu+\Sigma_{\hat{d}}^{\text{WW}}(p)]^{-1}$ and $[p+i\kappa+\Sigma_{\hat{a}}^{\text{WW}}(p)]^{-1}$ by their first term in the expansion (32), and (49) reduces to

$$[d]_p\,|\,\psi^{\text{WW}}\rangle=-\sum_\lambda\frac{D_\lambda e^{ik_\lambda r_0}|\,0_D;1_S;\lambda\rangle}{(p-i\lambda)(p+i\nu+\Gamma_D)}+\sum_\lambda D_\lambda M_\lambda\left[\frac{e^{-ik_\lambda r_0}}{p-i\lambda}-\frac{e^{ik_\lambda r_0}}{p+i\lambda}\right]\frac{|\,0_D;0_S;\text{vac}\rangle}{(p+i\kappa+\Gamma_S)(p+i\nu+\Gamma_D)}$$
$$+\sum_{\lambda,\gamma}\frac{D_\lambda M_\lambda M_\gamma}{p+i\nu+\Gamma_D}\left[\frac{e^{-ik_\lambda r_0}}{(p+i\lambda)(p-i\gamma)}\left[\frac{1}{p-i\kappa+\Gamma_S}-\frac{1}{p+i\kappa+\Gamma_S}\right]\right.$$
$$\left.+\frac{e^{ik_\lambda r_0}}{(p-i\lambda)(p-i\gamma)}\left[\frac{1}{p+i\kappa+\Gamma_S}-\frac{1}{p-i\kappa+\Gamma_S}\right]\right]|\,0_D;1_S;\gamma\rangle, \tag{50}$$

where κ and ν now represent the line-shifted frequencies $\kappa-\Delta_S$ and $\nu-\Delta_D$, respectively. The first term on the right-hand side of (50) represents the self-energy of the detector. This term times its Hermitian conjugate gives a contribution

$$\langle\hat{N}_D(t)\rangle^{\text{WW}}\approx\beta_D\left[(1+e^{-2\Gamma_D t})\ln\frac{\Lambda}{\nu}-2e^{-\Gamma_D t}[\text{ci}(\Lambda t)-\text{ci}(\nu t)-\cos\nu t+\nu t\,\text{si}(\Lambda t)-\nu t\,\text{si}(\nu t)]\right], \tag{51}$$

where the sine and cosine integrals, si and ci, are given in Appendix A and where

$$\beta_D\equiv\frac{2|\,\vec{d}\,'\,|^2\nu^2}{3\pi}. \tag{52}$$

For $t=0$, (51) is zero; however, for $\Gamma_D^{-1} \gg t \gg \nu^{-1} \gg \Lambda^{-1}$ its value is $\langle \hat{N}_D(t) \rangle_{\text{self}}^{\text{WW}} = \Delta_D/\nu$ or twice its interacting ground-state value. For $t \gg \Gamma_D^{-1}$, the detector returns to its ground-state value of $\Delta_D/2\nu$. Since we are only concerned with times $t \ll \Gamma_D^{-1}$, Δ_D/ν sets a lower limit on the observability of the spontaneous emission.

The important contribution to $\langle \hat{N}_D(t) \rangle^{\text{WW}}$ comes from the second term in (50), which, using the results of Appendix B, can be rewritten as

$$\frac{\bar{\beta}}{r_0} \int_0^\infty d\eta \sin \eta r_0 \frac{2i\eta \,|\, 0_D;0_S;\text{vac}\rangle}{(p^2+\eta^2)(p+i\kappa+\Gamma_S)(p+i\nu+\Gamma_D)} = \frac{i\bar{\beta}\pi e^{-pr_0}}{r_0} \frac{|\, 0_D;0_S;\text{vac}\rangle}{(p+i\kappa+\Gamma_S)(p+i\nu+\Gamma_D)} , \tag{53}$$

where $\bar{\beta} \equiv (\beta_D \beta_S)^{1/2}$ and where we have extended the integration from Λ to ∞, which has the effect of suppressing transients which occur for times on the order of Λ^{-1}. Taking the inverse Laplace transform of (53) and multiplying by the Hermitian conjugate gives

$$\langle \hat{N}_D(t) \rangle^{\text{WW}} \approx \frac{\beta_S \beta_D \Theta(\bar{t})}{r_0^2[(\kappa-\nu)^2+(\Gamma_S-\Gamma_D)^2]} [e^{-2\Gamma_S \bar{t}} + e^{-2\Gamma_D \bar{t}} - 2e^{-(\Gamma_S+\Gamma_D)\bar{t}} \cos(\kappa-\nu)\bar{t}] , \tag{54}$$

where $\bar{t} \equiv t - r_0$ and $\Theta(\bar{t})$ is the (theta) step function: $\Theta(\bar{t})=1$ for $\bar{t}>0$ and $\Theta(\bar{t})=0$ for $\bar{t}<0$. Before discussing (54) let us turn to the other terms in (50) which could contribute to $\langle \hat{N}_D(t) \rangle^{\text{WW}}$ to $O(|\vec{d}|^2 |\vec{d}'|^2)$. The third set of terms in (50), when multiplied by the Hermitian conjugate of the first term in (50), looks like

$$O(|\vec{d}|^2 |\vec{d}'|^2) \cdot \langle 0_D;1_S;\gamma \,|\, 0_D;1_S;\lambda \rangle$$

and will contribute when $\gamma=\lambda$. It can easily be shown after doing the necessary (tedious) integrations that the resulting contributions are unimportant. They are either proportional to $[(\kappa+\nu)^2+\Gamma_S^2]$—a nonresonant denominator—or are proportional to higher powers of r_0^{-n} for $n>2$, which we ignore since we are in radiation zone, or have terms which can be shown to be very small in the limit $\nu \to \kappa$ (and also very small otherwise).

Equation (54) therefore gives the time-dependent spectral line shape for the spontaneous emission of a two-level atom. Since we are interested in the case $\Gamma_D \ll \Gamma_S$, we can rewrite this, valid for $t\Gamma_D \ll 1$, as

$$\langle \hat{N}_D(t) \rangle^{\text{WW}} = \frac{\Theta(\bar{t})\beta_S \beta_D}{r_0^2[(\kappa-\nu)^2+\Gamma_S^2]} \times [1+e^{-2\Gamma_S \bar{t}} - 2e^{-\Gamma_S \bar{t}} \cos(\kappa-\nu)\bar{t}] . \tag{55}$$

We have the characteristic inverse square law intensity and the $\Theta(\bar{t})$ maintains causality—the detector does not turn on until the radiation has reached it at $t=r_0$. It is important to note that the line shape is not the usual Lorentzian which is given in the standard semiclassical derivations.[38] $\langle \hat{N}_D(t) \rangle^{\text{WW}}$ does give the usual Lorentzian for times such that $\Gamma_S^{-1} \ll t \ll \Gamma_D^{-1}$; however, its behavior for shorter times is much more interesting. There are oscillations in the wings which come from the $\cos[(\kappa-\nu)\bar{t}]$ term, where κ and ν are the line-shifted frequencies. Accurate time-dependent photon-counting experiments should verify this phenomena, assuming, of course, that ours is a reasonable model for photon detection. It is also clear that for $\nu \sim \kappa$, (55) is greater than (51) for the times of experimental interest.

V. DISCUSSION AND SUMMARY

We have shown in this paper that the standard Heisenberg equations-of-motion, Liouvillian Green's-function, and master equation methods for computing expectation values of physical observables can give incorrect results at higher orders in the usual perturbation expansion for operators which commute with the free or zeroth-order Hamiltonian. The improved Heisenberg equations-of-motion method, formulated in Sec. III, whereby the original operator $\hat{O}$ is first rewritten as a product of operators $\hat{O}=\hat{A}\hat{B}$, and then the matrix elements of $\hat{\rho}^{1/2}\hat{A}$ and $\hat{B}\hat{\rho}^{1/2}$ are calculated, has been shown to give convergent answers and to be applicable to pure-state, equilibrium, and more importantly, nonequilibrium systems. Calculations using the model of a two-level atom interacting with electromagnetic radiation were presented to give an explicit example of the utility of the present approach.

The definition of time-dependent, spectral line shape in terms of a two-level—atom detector was discussed in Sec. IV. Some important variations of this calculation would be to take the initial condition to include a coherent state for the photon field, to add a classical source term to discuss the time-dependent resonant fluorescence and the limits of using classical sources, and to investigate more complicated ensembles such as finite temperature. In some interesting papers, Eberly *et al.*[39,40] have also investigated the question of giving a more physical definition of spectral line shape. They employ a standard definition of photon-counting rate, but they also argue that any meaningful definition of spectrum must take into account the physical process of filtering the incoming signal to the photodetector. They include the filter phenomenologically by convolving the filter response with the incoming radiation field—this is analogous to the tunability and natural lifetime of our two-level atom. Although the emphasis of their work is quite different than that of this paper, it is interesting to note that their semiclassical calculation of the "physical spectrum" of spontaneous emission gives a spectrum equivalent to (54), where instead of Γ_D representing the natural lifetime of the detector, they have the full width of a Fabry-Perot interferometer. One can conclude from this that in a real experiment, the actual detector response will be a complicated convolution of both the filter response and the detector response. This is in many ways similar to analogous problems in electrical engineering.

Finally, it is important to emphasize that even though we did not apply our formalism to any nonequilibrium examples, the improved Heisenberg equations-of-motion method has been formulated so as to apply to any arbi-

trary statistical operator $\hat{\rho}$. This is the major advantage of our method in comparison to the simpler temperature-time Green's-function techniques. The more elegant Liouvillian Green's function and self-energy approach,[23] which is expressly designed to apply to nonequilibrium systems, can now be suitably modified along the lines of Sec. III so as to give correct results. This is particularly important in that the environmental contributions which we treated in a somewhat *ad hoc* manner in Sec. II are dealt with in a very natural and consistent manner in this approach.

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APPENDIX A: FORMULAS NEEDED FOR CALCULATIONS

The following formulas are used in the text. They are listed for convenience and taken from Ref. 41. The exponential integral is defined by

$$E_1(z)=\int_z^\infty dt\frac{e^{-t}}{t},\quad |\arg z|<\pi \tag{A1}$$

and has the series expansion

$$E_1(z)=-\gamma-\ln z-\sum_{n=1}^{\infty}\frac{(-1)^n z^n}{n\,n!} \tag{A2}$$

and the asymptotic expansion

$$E_1(z)\sim\frac{e^{-z}}{z}\left[1-\frac{1}{z}+\frac{1\times 2}{z^2}-\frac{1\times 2\times 3}{z^3}+\cdots\right], \tag{A3}$$

where γ is Euler's constant, $\gamma=0.577\,21\ldots$.

The sine and cosine integrals are given by

$$\int_z^\infty dt\frac{\sin t}{t}=-\mathrm{si}(z)=-\mathrm{Si}(z)+\frac{\pi}{2}, \tag{A4}$$

$$\int_z^\infty dt\frac{\cos t}{t}=-\mathrm{Ci}(z)=\mathrm{ci}(z),\quad \arg|z|<\pi \tag{A5}$$

with series expansion

$$\mathrm{Si}(z)=\sum_{n=0}^{\infty}\frac{(-1)^n z^{2n+1}}{(2n+1)(2n+1)!}, \tag{A6}$$

$$\mathrm{Ci}(z)=\gamma+\ln z+\sum_{n=1}^{\infty}\frac{(-1)^n z^{2n}}{2n\,(2n)!}. \tag{A7}$$

The auxiliary functions $f(z)$ and $g(z)$ are given by

$$f(z)=\mathrm{Ci}(z)\sin z-\mathrm{si}(z)\cos z, \tag{A8}$$

$$g(z)=-\mathrm{Ci}(z)\cos z-\mathrm{si}(z)\sin z \tag{A9}$$

and have the asymptotic expansion

$$f(z)\sim\frac{1}{z}\left[1-\frac{2!}{z^2}+\frac{4!}{z^4}-\frac{6!}{z^6}+\cdots\right], \tag{A10}$$

$$g(z)\sim\frac{1}{z^2}\left[1-\frac{3!}{z^2}+\frac{5!}{z^4}-\frac{7!}{z^6}+\cdots\right]. \tag{A11}$$

The method used for calculating the convolution integral in (34) is to rewrite sine and cosine in terms of exponentials. This gives integrals of the form (A1), where, for integrals with t^{-n} for $n\geq 2$, integration by parts is performed. The integrals needed in Sec. IV either give logarithms or can be written as the sine and cosine transforms:

$$\int_0^\infty d\lambda\frac{\sin\lambda a}{\lambda-\nu\pm i\gamma}=f[(-\nu\pm i\gamma)a]=f[(\nu\mp i\gamma)a]+\pi e^{\mp i(\nu\mp i\gamma)a}, \tag{A12}$$

$$\int_0^\infty d\lambda\frac{\cos\lambda a}{\lambda-\nu\pm i\gamma}=g[(-\nu\pm i\gamma)a]=g[(\nu\mp i\gamma)a]\mp i\pi e^{\mp i(\nu\mp i\gamma)a}, \tag{A13}$$

$$\mathscr{P}\int_0^\infty d\lambda\frac{\sin\lambda a}{\lambda-\nu}=-f(\nu a)+\pi\cos\nu a, \tag{A14}$$

$$\mathscr{P}\int_0^\infty d\lambda\frac{\cos\lambda a}{\lambda-\nu}=g(\nu a)-\pi\sin\nu a, \tag{A15}$$

where ν is real and $\mathrm{Re}\gamma>0$.

APPENDIX B: GEOMETRY FOR TWO-LEVEL DETECTOR (REF. 38)

For ease of computation we choose

$$\vec{d}=|\vec{d}|\hat{x}\ \text{and}\ \vec{d}\,'=|\vec{d}\,'|\hat{x},$$

where $\vec{d}$ and $\vec{d}\,'$ are the transition dipole-matrix elements (assumed real) for the source and detector atom, respectively. The polarization sum which arises in (50) in the interaction terms simplifies to

$$\sum_\lambda \vec{e}_\lambda\cdot\vec{d}\,\vec{e}_\lambda\cdot\vec{d}\,'=|\vec{d}\,||\vec{d}\,'|(1-\sin^2\theta\cos^2\phi), \tag{B1}$$

where r, θ, ϕ are the usual spherical coordinates. We choose the detector atom to be located at $\vec{r}_0=r_0\hat{z}$ where the source atom is at the origin. The resulting angular integrals are

$$\int_0^\pi d\theta\int_0^{2\pi}d\phi\sin\theta(1-\sin^2\theta\cos^2\phi)e^{\pm ir_0\gamma\cos\theta}=\int_0^\pi d\theta(2\pi\sin\theta-\pi\sin^3\theta)e^{\pm ir_0\gamma\cos\theta}. \tag{B2}$$

If we let $u=\cos\theta$, (B2) reduces to

$$2\pi\int_0^1 du\,(1+u^2)\cos r_0\gamma u = 4\pi\left[\frac{\sin r_0\gamma}{r_0\gamma}+\frac{\cos r_0\gamma}{(r_0\gamma)^2}-\frac{\sin r_0\gamma}{(r_0\gamma)^3}\right], \quad \text{(B3)}$$

where γ is the relevant photon energy. The latter two terms in (B3) contribute only in the near zone. Since our calculation is for the radiation zone these terms are dropped.

[1]A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, New Jersey, 1963).

[2]A. L. Fetter and J. D. Walecka, *Quantum Theory of Many-Particle Systems* (McGraw-Hill, New York, 1971).

[3]K. Wodkiewicz and J. H. Eberly, Ann. Phys. (N.Y.) 101, 574 (1976).

[4]M. D. Girardeau, Phys. Rev. A (in press).

[5]See, e.g., R. Balescu, *Equilibrium and Nonequilibrium Statistical Mechanics* (Wiley, New York, 1975).

[6]R. Davidson and J. J. Kozak, J. Math. Phys. 11, 189 (1970).

[7]R. Paley and N. Wiener, *Fourier Transforms in the Complex Domain* (American Mathematical Society, Providence, Rhode Island, 1934).

[8]It is important to take the limit $\Omega\to\infty$ before letting $t\to\infty$ in order to exhibit dissipative behavior.

[9]J. R. Ackerhalt, P. L. Knight, and J. H. Eberly, Phys. Rev. Lett 30, 456 (1973); P. W. Milonni, J. R. Ackerhalt, and W. A. Smith, *ibid.* 31, 958 (1973).

[10]P. W. Milonni, in *Foundations of Radiation Theory and Quantum Electrodynamics,* edited by A. O. Barut (Plenum, New York, 1980).

[11]See S. S. Schweber, *An Introduction to Relativistic Quantum Field Theory* (Harper and Row, New York, 1961).

[12]K. O. Friedrichs, Commun. Pure Appl. Math. 1, 361 (1948); *Perturbation of Spectra in Hilbert Space* (American Mathematical Society, Providence, Rhode Island, 1965).

[13]We use the definitions in *Table of Integral Transforms,* edited by A. Erdelyi *et al.* (McGraw-Hill, New York, 1954), Vol. I. This differs by $p\to iz$ from usual physics notation.

[14]Note that operators without any $\hat{a}$ or $\hat{a}^\dagger$'s in them will not have a self-energy: $\Sigma_{\hat{O}}(p)=0$. The Laplace transform for these, Eq. (12), will look like $\langle[O]_p\rangle\approx\langle\hat{O}(0)\rangle/(p+iE_{\hat{O}})+O(|\vec{d}|)$. Also, if in Eq. (12), $\langle\hat{O}(0)\rangle=0$, the ansatz (12) fails and we have to go to a higher order in the perturbation expansion—see Ref. 23.

[15]It is important to emphasize that we may also be able to perform a second perturbation expansion because of the statistical averaging operation $\langle\hat{O}\rangle$. The obvious example is thermal equilibrium where one can perform a second perturbation expansion of

$$\exp(\hat{H}/k_BT)=\exp(\hat{H}_0/k_BT+\hat{H}'/K_BT)$$

in powers of $\hat{H}'$.

[16]The "rotating-wave approximation" drops the nonenergy-conserving terms $-\hat{b}_\lambda^\dagger\hat{a}^\dagger$ and $\hat{b}_\lambda\hat{a}$ in (1). "Persistent perturbation effects" refer to the fact that if the full Hamiltonian (1) is used, the interacting states are dressed and the free-field states are not an adequate basis. See. Ref. 29.

[17]B. Van der Pol and H. Bremmer, *Operational Calculus* (Cambridge Press, Great Britain, 1955), second edition, Chap. VII.

[18]L. Fonda, G. C. Ghirardi, and A. Rimini, Rep. Prog. Phys. 41, 587 (1978).

[19]L. A. Khalfin, Zh. Eksp. Teor. Fiz. 33, 1371 (1957) [Sov. Phys.—JETP 6, 1053 (1958)]; M. Levy, Nuovo Cimento 14, 612 (1959).

[20]M. L. Goldberger and K. M. Watson, *Collision Theory* (Wiley, New York, 1964); see also derivation in Ref. 18 and additional references cited therein.

[21]R. W. Zwanzig, in *Lectures in Theoretical Physics (Boulder, 1960),* edited by W. E. Brittin, B. W. Downs, and J. Downs (Interscience, New York, 1961), Vol. III.

[22]A. Bohm, J. Math. Phys. 21, 1040 (1980); 22, 2813 (1981).

[23]M. D. Girardeau and C. F. Hart, Phys. Rev. A (in press).

[24]C. I. George, F. Henin, F. Mayné, and I. Prigogine, Had. J. 1, 520 (1978) and references cited therein to earlier work of Brussels School.

[25]M. D. Girardeau, Physica (Utrecht) 98A, 139 (1979).

[26]The idea of calculating $\langle\hat{N}(t)\rangle$ in this way is not new (see Ref. 28) and is, in fact, equivalent to a Schrödinger-picture approach. The method of Eq. (27) is new and is not equivalent to the Schrödinger-picture approach.

[27]H. A. Bethe, Phys. Rev. 72, 339 (1947).

[28]R. L. Davidson and J. J. Kozak, J. Math. Phys. 12, 903 (1971); 14, 423 (1973); J. Jyy-wen Yang, R. Davidson, and J. J. Kozak, *ibid.* 15, 491 (1974).

[29]L. Davidovich and H. M. Nussenzveig, in *Foundations of Radiation Theory and Quantum Electrodynamics,* edited by A. O. Barut (Plenum, New York, 1980).

[30]The Yukawa-type form factors $k^n/(k^2+\Lambda^2)^m$ will give logarithmic contributions which will have branch cuts. See Ref. 17.

[31]See, e.g., K. Gottfried, *Quantum Mechanics* (W. A. Benjamin, Reading, Mass., 1966), Chap. VII.

[32]M. Baranger, Phys. Rev. 111, 481 (1958); 111, 494 (1958), and references cited therein.

[33]F. Wooten, *Optical Properties of Solids* (Academic, New York, 1972).

[34]R. G. Breene, Jr., *Theories of Spectral Line Shape* (Wiley, New York, 1981) and references cited therein.

[35]J. R. Klauder and E. C. G. Sudarshan, *Fundamentals of Quantum Optics* (W. A. Benjamin, New York, 1968).

[36]R. J. Glauber, Phys. Rev. Lett. 10, 84 (1963); Phys. Rev. 130, 2529 (1963); 131, 2766 (1963).

[37]P. C. Kelley and W. H. Kleiner; Phys. Rev. A 136, 316 (1964).

[38]E. Fermi, Rev. Mod. Phys. 4, 87 (1932); W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).

[39]I. C. Khoo and J. H. Eberly, Phys. Rev. A 18, 2184 (1978).

[40]J. H. Eberly and K. Wodkiewicz, J. Opt. Soc. Am. 67, 1252 (1977); J. H. Eberly, C. V. Kunasz, and K. Wodkiewicz, J. Phys. B 13, 217 (1980); X. Y. Huang, R. Tanas, and J. H. Eberly, Phys. Rev. A 26, 892 (1982).

[41]*Handbook of Mathematical Functions,* edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1972), Chap. 5.