# Liouvillian Green's-function theory of spectral line shape

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The general method of the preceding paper is applied to the theory of spectral line shape. Illustrative calculations of the transition-energy shift and width are carried out for a two-level atom interacting with the quantized electromagnetic field. In the thermal-equilibrium case, explicit results are given in terms of the digamma function and Bose-Einstein distribution, generalizing the standard Lamb-shift and Weisskopf-Wigner natural width results to nonzero temperature. In the nonequilibrium case expressions are obtained which exhibit the influence of nonequilibrium photon distributions on the shift and width. A general theory of the time-dependent power radiated in the decay of an excitation is developed for systems not necessarily at or near equilibrium, in terms of appropriate generalizations of the Liouvillian Green's functions of the preceding paper. This approach is illustrated by calculation of the time-dependent radiated power for the same two-level model.

## I. INTRODUCTION

In the preceding paper,[1] a general Liouville-space method has been developed for evaluation of Liouvillian Green's functions and transition self-energies of dissipative quantum-mechanical systems not necessarily in or near equilibrium. The objectives of the present paper are firstly to illustrate that method by explicit transition-energy shift and width calculations for a two-level atom in a radiation bath, secondly to formulate a general method of calculation of radiated power in terms of suitably generalized Liouvillian Green's functions, and thirdly to illustrate that method by explicit calculations for the same two-level model.

The transition-energy shift and width calculations for the two-level atom in an equilibrium or nonequilibrium radiation bath are carried out in Sec. II. In Sec. III formulas for the time-dependent power radiated in the decay of an excitation are derived for a system not necessarily at or near equilibrium, and a Liouvillian Green's-function hierarchy for evaluation of such quantities is derived. This general formalism is illustrated in Sec. IV by calculation of the radiated power for the same model considered in Sec. II. Analysis of the spectral line-shape function will be left for a future paper.

## II. NATURAL VERSUS ENVIRONMENTAL RADIATIVE WIDTHS AND SHIFTS FOR A TWO-LEVEL ATOM

In this section the general approach of the preceding paper[1] will be illustrated by calculation of transition-energy shifts and widths for the familiar two-level atomic model interacting with electromagnetic radiation. We shall first consider the case where the system of atom plus electromagnetic radiation differs from thermal equilibrium at some nonzero temperature $T$ only through the specification that at time $t=0$ the atom is excited into its upper state, with probability one. The radiative interaction and thermal radiation bath act to perturb the transition (excitation) energy, and the shift and width of this perturbed transition energy are given by the quantity $\Delta - i\gamma$ previously defined. This frequency shift and width will be shown to approach the standard results of natural—line-shape theory (Lamb-shift and Wigner-Weisskopf width) in the limit $T\rightarrow 0$. The same model system will then be considered in the case where the initial conditions are those describing the atom interacting with a nonequilibrium coherent state of the photons. This case is investigated as a simple example of a calculation of the self-energy of a system far from equilibrium.

The Hamiltonian $\hat{H}$ of the model is taken to be

$$\begin{aligned}\hat{H} &= \hat{H}_0 + \hat{H}' ,\\ \hat{H}_0 &= \omega_0 \hat{a}^\dagger \hat{a} + \sum_{\vec{k},\lambda} \omega_k \hat{b}^\dagger_{\vec{k}\lambda} \hat{b}_{\vec{k}\lambda} , \\ \hat{H}' &= i \sum_{\vec{k},\lambda} M_{\vec{k}\lambda} (\hat{b}_{\vec{k}\lambda} + \hat{b}^\dagger_{\vec{k}\lambda})(\hat{a} - \hat{a}^\dagger) .\end{aligned} \quad (1)$$

Here $\omega_0$ 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 zero for the Fermi occupation-number operator $\hat{N}=\hat{a}^\dagger\hat{a}$ then implies that the atom is in its lower state, whereas $\hat{N}=1$ implies that it is in its upper state. The $\hat{b}_{\vec{k}\lambda}$ and $\hat{b}^\dagger_{\vec{k}\lambda}$ are Bose annihilation and creation operators for photons of wave vector $\vec{k}$, polarization index $\lambda=1$ or 2, and energy $\omega_k=ck$. The commutation and anticommutation relations are

$$\begin{aligned}&\hat{a}^2=(\hat{a}^\dagger)^2=0,\ [\hat{a},\hat{a}^\dagger]_+=1\ ,\\&[\hat{b}_{\vec{k}\lambda},\hat{b}_{\vec{k}'\lambda'}]_-=[\hat{b}^\dagger_{\vec{k}\lambda},\hat{b}^\dagger_{\vec{k}'\lambda'}]_-=0\ ,\\&[\hat{b}_{\vec{k}\lambda},\hat{b}^\dagger_{\vec{k}'\lambda'}]_-=\delta_{\vec{k}\vec{k}'}\delta_{\lambda\lambda'}\ ,\\&[\hat{a},\hat{b}_{\vec{k}\lambda}]_-=[\hat{a},\hat{b}^\dagger_{\vec{k}\lambda}]_-=0\ .\end{aligned}\tag{2}$$

The photon wave vectors $\vec{k}$ are quantized in the usual way, i.e., a discrete cubic $\vec{k}$-space lattice with lattice constant $2\pi/\Omega^{1/3}$, where $\Omega$ is the macroscopic system volume which will eventually become infinite. The interaction matrix element is

$$M_{\vec{k}\lambda}=(2\pi/\omega_k\Omega)^{1/2}\omega_0\vec{d}\cdot\vec{e}_{\vec{k}\lambda}\ ,\tag{3}$$

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

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

The Hamiltonian (1) differs from the usual two-level model[2,3] only in that Fermi operators, instead of Pauli spin matrices, are used to describe the two-level atom—only a trivial change of notation and shift of $\frac{1}{2}\omega_0$ in the energy origin.

The first step in the evaluation of (I.58) consists of determination of the nonzero $c(1\mid n)$, the coefficients in the operator basis expansion[1] (I.5):

$$[\hat{a},\hat{H}]=\sum_n c(1\mid n)\hat{B}_n\ .\tag{5}$$

The notation here implies $\alpha=1$, $\hat{A}^\dagger_\alpha=\hat{a}^\dagger$, where the 1 is a reminder that the atom is in its upper state (denoted by 1) after excitation. By (1) and (2)

$$[\hat{a},\hat{H}]=\omega_0\hat{a}-i\sum_{\vec{k},\lambda}M_{\vec{k}\lambda}(\hat{b}_{\vec{k}\lambda}+\hat{b}^\dagger_{\vec{k}\lambda})(1-2\hat{a}^\dagger\hat{a})\ .\tag{6}$$

We thus identify the unperturbed transition energy $\epsilon_1=c(1\mid 1)$ as $\epsilon_1=\omega_0$ (as expected) and the nonzero $c(1\mid n)$ with $n\neq 1$ as

$$\begin{aligned}&c(1\mid\ ;\vec{k}\lambda)=c(1\mid\vec{k}\lambda;\ )=-iM_{\vec{k}\lambda}\\&c(1\mid 1;1,\vec{k}\lambda)=c(1\mid 1,\vec{k}\lambda;1)=2iM_{\vec{k}\lambda}\ .\end{aligned}\tag{7}$$

Here $c(1\mid\ ;\vec{k}\lambda)$ and $c(1\mid\vec{k}\lambda;\ )$ are the coefficients of $\hat{b}_{\vec{k}\lambda}$ and $\hat{b}^\dagger_{\vec{k}\lambda}$, respectively, whereas $c(1\mid 1;1,\vec{k}\lambda)$ and $c(1\mid 1,\vec{k}\lambda;1)$ are those of $\hat{a}^\dagger\hat{b}_{\vec{k}\lambda}\hat{a}$ and $\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda}\hat{a}$, respectively. In general, $c(1\mid\mathscr{S}_b;\mathscr{S}_f)$ is the coefficient of the normal-ordered basis element which is a product of creation operators labeled by the set $\mathscr{S}_b$ and annihilation operators labeled by the set $\mathscr{S}_f$; a blank before or after the semicolon denotes the absence of creation or annihilation operators, respectively.

It follows from (6) or (7) and (1) that the set of basis elements $\mathscr{L}_0$ degenerate[1] with $\hat{a}$ for which $c(1\mid n)\neq 0$, consists solely of those terms $\hat{b}_{\vec{k}\lambda}$ and $\hat{a}^\dagger\hat{b}_{\vec{k}\lambda}\hat{a}$ for which $ck=\omega_0$. This is a set of measure zero (a surface in the three-dimensional $\vec{k}$ space), from which it follows that the first-order contribution [first term in (I.58), with $n\in\mathscr{S}_1$] vanishes, as do the second-order contributions with $n\in\mathscr{S}_1$ [second line of (I.58)]. Thus (I.58) reduces, with (I.60), to[4]

$$\begin{aligned}\Delta-i\gamma&=\sum_{n\notin\mathscr{S}_1}\sum_{m\in\mathscr{S}_1}\frac{c(1\mid n)c(n\mid m)g(m,0\mid 1,0)}{(\omega_0-\epsilon_n+i\eta)g(1,0\mid 1,0)}\\&=\mathscr{P}\sum_{n\notin\mathscr{S}_1}\sum_{m\in\mathscr{S}_1}\frac{c(1\mid n)c(n\mid m)g(m,0\mid 1,0)}{(\omega_0-\epsilon_n)g(1,0\mid 1,0)}\\&\quad-i\pi\sum_n\sum_{m\in\mathscr{S}_1}c(1\mid n)c(n\mid m)\delta(\omega_0-\epsilon_n)\frac{g(m,0\mid 1,0)}{g(1,0\mid 1,0)}\end{aligned}\tag{8}$$

to second order in the interaction. To evaluate these expressions we need to evaluate the relevant $c(n\mid m)$ where the $n$ label the basis elements $\hat{B}_n$ defined by (7) and the subsequent discussion, and where $m\in\mathscr{S}_1$, i.e., $\hat{B}_m$ that are $\mathscr{L}_0$ degenerate with $\hat{a}$. Evaluating the relevant commutators $[\hat{B}_n,\hat{H}']$ with (1), one finds that the only $c(n\mid m)$ satisfying these criteria are[5]

$$\begin{aligned}&c(\ ;\vec{k}\lambda\mid 1)=iM_{\vec{k}\lambda}\leftrightarrow\hat{B}_m=\hat{a}\ ,\\&c(\vec{k}\lambda;\ \mid 1)=-iM_{\vec{k}\lambda}\leftrightarrow\hat{B}_m=\hat{a}\ ,\\&c(1,\vec{k}\lambda;1\mid 1)=-iM_{\vec{k}\lambda}\leftrightarrow\hat{B}_m=\hat{a}\ ,\end{aligned}\tag{9}$$

and

$$c(1;1,\vec{k}\lambda \mid \vec{k}\lambda;1,\vec{k}\lambda) = -iM_{\vec{k}\lambda} \leftrightarrow \hat{B}_m = \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a} ,$$
$$c(1,\vec{k}\lambda;1 \mid \vec{k}\lambda;1,\vec{k}\lambda) = -iM_{\vec{k}\lambda} \leftrightarrow \hat{B}_m = \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a} . \tag{10}$$

The contributions to (8) from $m=1$ ($\hat{B}_m=\hat{a}$) give a Weisskopf-Wigner (WW) transition self-energy $\Delta^{\mathrm{WW}}-i\gamma^{\mathrm{WW}}$ [see (I.61)] which is found from (7) and (9), upon writing $\sum_{\vec{k}} \to (2\pi)^{-3}\Omega \int d^3k$ (valid asymptotically for $\Omega \to \infty$), to be

$$\Delta^{\mathrm{WW}}-i\gamma^{\mathrm{WW}} = \mathscr{P} \sum_\lambda (2\pi)^{-3}\Omega \int d^3k M^2_{\vec{k}\lambda} \left[ \frac{1}{\omega_0-\omega_{\vec{k}}} + \frac{1}{\omega_0+\omega_k} \right]$$
$$-i\pi \sum_\lambda (2\pi)^{-3}\Omega \int d^3k M^2_{\vec{k}\lambda} [\delta(\omega_0-\omega_k)+\delta(\omega_0+\omega_k)] , \tag{11}$$

where the relevant $\epsilon_n$ (unperturbed) transition energies), which follow from

$$\mathscr{L}_0\hat{B}_n = [\hat{B}_n,\hat{H}_0] = \epsilon_n\hat{B}_n , \tag{12}$$

are

$$\epsilon( ;\vec{k}\lambda) = \omega_k, \; \epsilon(\vec{k}\lambda; ) = -\omega_k ,$$
$$\epsilon(1,\vec{k}\lambda;1 \mid 1) = -\omega_k . \tag{13}$$

Inserting $\omega_k = ck$ and the expression (3) for $M_{\vec{k}\lambda}$, noting that the angular integrations and polarization summation give a factor $8\pi/3$, and introducing a new integration variable $\omega = ck$, one finds

$$\Delta^{\mathrm{WW}} = \frac{8\pi}{3c^3} \left[ \frac{\omega_0 d}{2\pi} \right]^2 \times \mathscr{P} \int_0^\infty \left[ \frac{1}{\omega_0-\omega} + \frac{1}{\omega_0+\omega} \right] \omega \, d\omega ,$$
$$\gamma^{\mathrm{WW}} = \frac{2(\omega_0 d)^2}{3c^3} \int_0^\infty [\delta(\omega_0-\omega) + \delta(\omega_0+\omega)]\omega \, d\omega = \frac{2\omega_0^3 d^2}{3c^3} . \tag{14}$$

These expressions agree with those previously obtained[2,3] for the transition frequency shift and width by a Heisenberg equation-of-motion approach. $\gamma^{\mathrm{WW}}$ is half the Einstein $A$ coefficient for spontaneous emission. $\Delta^{\mathrm{WW}}$ is an improved version of the Weisskopf-Wigner line shift. As previously noted by Ackerhalt *et al.*,[2] it is only logarithmically divergent, in contrast with the linear divergence of the standard Weisskopf-Wigner line shift. In fact, the contribution to the expression (14) for $\Delta^{\mathrm{WW}}$ coming from $\omega$ in the range $\omega \gtrsim \omega_0$ but $\omega \lesssim mc^2$, found by expanding the integrand in powers of $\omega_0/\omega$, retaining the nonvanishing term, and integrating from $\omega_0$ to $mc^2$, is

$$\Delta_{\mathrm{Lamb}} \approx -\frac{4\omega_0^3 d^2}{3\pi c^3} \ln \left[ \frac{mc^2}{\omega_0} \right] . \tag{15}$$

This is the two-level version of Bethe's nonrelativistic expression[6] for the Lamb shift.

The environmental contributions to $\Delta$ and $\gamma$ come from the terms in (8) with $m \neq 1$ ($\hat{B}_m \neq \hat{a}$) but $m \in \mathscr{S}_1$ ($\hat{B}_m$ that are $\mathscr{L}_0$ degenerate with $\hat{a}$). These are found to be

$$\Delta^{\mathrm{env}}-i\gamma^{\mathrm{env}} = \mathscr{P} \sum_\lambda (2\pi)^{-3}\Omega \int d^3k \, 2M^2_{\vec{k}\lambda} \left[ \frac{1}{\omega_0-\omega_k} + \frac{1}{\omega_0+\omega_k} \right] \frac{\langle \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a}\hat{a}^\dagger \rangle}{\langle \hat{a}\hat{a}^\dagger \rangle}$$
$$-i\pi \sum_\lambda (2\pi)^{-3}\Omega \int d^3k \, 2M^2_{\vec{k}\lambda} [\delta(\omega_0-\omega_k)+\delta(\omega_0+\omega_k)] \frac{\langle \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a}\hat{a}^\dagger \rangle}{\langle \hat{a}\hat{a}^\dagger \rangle} . \tag{16}$$

These expressions are valid for either equilibrium or nonequilibrium ensemble averages (indicated by the angular brackets).

Consider first the case of thermal equilibrium. Then the averages are taken in the canonical ensemble

$$\hat{\rho} = Z^{-1}e^{-\beta\hat{H}}, \; Z = \mathrm{Tr}\, e^{-\beta\hat{H}} , \tag{17}$$

with $\beta = 1/k_BT$. Since the integrand of (16) is already of second order in the interaction, we need evaluate the averages (indicated by angular brackets) only to zeroth order, i.e., we may replace $\hat{H}$ by $\hat{H}_0$, Eq. (1), in (17). Then there is no statistical correlation between the atom and photon operators, i.e.,

$$\langle \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a}\hat{a}^\dagger \rangle_0 = \langle \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda} \rangle_0 \langle \hat{a}\hat{a}^\dagger \rangle_0 , \tag{18}$$

and one finds

$$\frac{\langle \hat{b}^{\dagger}_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a}\hat{a}^{\dagger}\rangle_0}{\langle \hat{a}\hat{a}^{\dagger}\rangle_0}=\langle \hat{b}^{\dagger}_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\rangle_0=f_{\vec{k}\lambda}$$
$$=(e^{\beta\omega_k}-1)^{-1}\,, \qquad (19)$$

the usual Bose-Einstein photon distribution function. Transforming to integrals over a dimensionless energy variable $x=\omega/\omega_0$ in analogy with the reduction of (11) to (14), one finds

$$\Delta^{\rm env}=\frac{4\omega_0^3 d^2}{3\pi c^3}I(\beta\omega_0)\,,$$

$$\gamma^{\rm env}=\frac{4\omega_0^3 d^2}{3c^3}\int_0^\infty [\delta(1-x)+\delta(1+x)]xf(\beta\omega_0 x)dx$$

$$=\frac{4\omega_0^3 d^2}{3c^3}f(\beta\omega_0)\,, \qquad (20)$$

where $f(y)$ is the Bose-Einstein distribution function

$$f(y)=(e^y-1)^{-1} \qquad (21)$$

and $I(y)$ is the integral

$$I(y)=\mathscr{P}\int_0^\infty \left[\frac{1}{1-x}+\frac{1}{1+x}\right]xf(yx)dx\,. \qquad (22)$$

This integral is expressed in Appendix A in terms of the digamma ($\psi$) function. It is shown there that its behavior for low and high temperature is

$$I(\omega_0/k_BT)=\begin{cases}\frac{1}{12}(2\pi k_BT/\omega_0)^2+O((k_BT/\omega_0)^4), & k_BT\ll\omega_0\\ \ln(2\pi k_BT/\omega_0)+O(1), & k_BT\gg\omega_0\end{cases} \qquad (23)$$

i.e., the environmental shift $\Delta^{\rm env}$ vanishes quadratically with $k_BT/\omega_0$ at low temperatures and increases logarithmically with the same quantity at high temperatures. The behavior of the environmental width $\gamma^{\rm env}$ is quite different. It follows from (21) that

$$f(\omega_0/kT)=\begin{cases}e^{-\omega_0/k_BT}+O(e^{-2\omega_0/k_BT}), & k_BT\ll\omega_0\\ k_BT/\omega_0+O(1), & k_BT\gg\omega_0\end{cases} \qquad (24)$$

i.e., $\gamma^{\rm env}$ vanishes exponentially at low temperatures and increases linearly at high temperatures. Thus the environmental width is negligible compared to the natural width $\gamma^{\rm WW}$ [Eq. (14)] at low temperatures, but dominates it at high temperatures $k_BT\gg\omega_0$. In contrast, the environmental shift is negligible compared to the Lamb shift $\Delta_{\rm Lamb}$ [Eq. (15)] at low temperatures, but it never exceeds the Lamb shift at physically reasonable temperatures. However, because both $\Delta_{\rm Lamb}$ and $\Delta^{\rm env}$ vary very slowly (logarithmically) at high $T$, it follows that $\Delta^{\rm env}$ is of the same order of magnitude as $\Delta_{\rm Lamb}$ already when $k_BT\sim\omega_0$. In Fig. 1 the expressions (20) for $\Delta^{\rm env}$ and $\gamma^{\rm env}$ are plotted as functions of $k_BT/\omega_0$, using the exact expressions for $I(\omega_0/k_BT)$ (Appendix A) and $f(\omega_0/k_BT)$ [Eq. (21)]. As a check of the derivation of the expressions (20) by our Liouvillian self-energy formalism, it is shown in Appendix B that the same formulas follow from the standard (but less general) thermodynamic Green's-function formalism.

Shifts and widths of highly excited (Rydberg) energy levels of a number of atoms due to blackbody-radiation environment have been calculated by Farley and Wing.[7] Their calculation treats the radiation classically, and expresses the widths in terms of an integral closely related to our integral $I(y)$, Eq. (22). The connection is discussed at the end of Appendix A; we show that their integral is expressible in terms of the digamma function, as is the integral of Eq. (22).

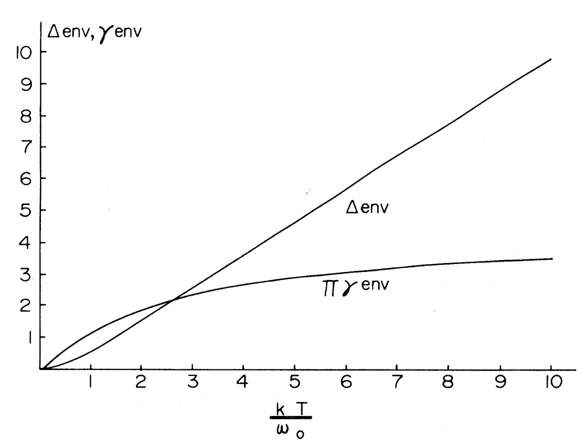


FIG. 1. Environmental shift $\Delta^{\rm env}$ and width $\gamma^{\rm env}$ as functions of $k_BT/\omega_0$.

Let us next consider a case where the radiation is not in thermal equilibrium, namely, that where the statistical average (I.3) is an expectation value[8]

$$\langle \hat{O} \rangle = \langle \psi | \hat{O} | \psi \rangle \tag{25}$$

in a normalized coherent state

$$|\psi\rangle = \text{const}\left[\exp\left[\sum_{\vec{k},\lambda} g_{\vec{k}\lambda}\hat{b}^\dagger_{\vec{k}\lambda}\right]\right]|0\rangle\,, \tag{26}$$

where $g_{\vec{k}\lambda}$ is a $c$-number function and $|0\rangle$ the vacuum of the $\hat{b}_{\vec{k}\lambda}$ and $\hat{a}$:

$$\hat{b}_{\vec{k}\lambda}|0\rangle = \hat{a}|0\rangle = 0\,. \tag{27}$$

Then

$$\hat{b}_{\vec{k}\lambda}|\psi\rangle = g_{\vec{k}\lambda}|\psi\rangle \tag{28}$$

so that the mean values in (16) are trivially evaluated:

$$\frac{\langle \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\hat{a}\hat{a}^\dagger\rangle}{\langle \hat{a}\hat{a}^\dagger\rangle} = \langle \hat{b}^\dagger_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\rangle = |g_{\vec{k}\lambda}|^2 \equiv f_{\vec{k}\lambda}\,. \tag{29}$$

Then

$$\Delta^{\text{env}} = \mathscr{P}\sum_\lambda (2\pi)^{-3}\Omega \times \int d^3k\, 2M^2_{\vec{k}\lambda}\left[\frac{1}{\omega_0-\omega_k} + \frac{1}{\omega_0+\omega_k}\right] f_{\vec{k}\lambda}\,,$$

$$\gamma^{\text{env}} = \pi\sum_\lambda (2\pi)^{-3}\Omega \times \int d^3k\, 2M^2_{\vec{k}\lambda}[\delta(\omega_0-\omega_k) + \delta(\omega_0+\omega_k)] f_{\vec{k}\lambda}\,. \tag{30}$$

Note that the coherent-state assumption is not essential for the derivation of (30). In fact, the only property of the coherent state necessary for the validity of (30) is the factorization (29) of the statistical average, and (30) remains valid for any ensemble satisfying this factorization property.[9] This is as far as we can go without assuming an explicit form for $f_{\vec{k}\lambda}$, the photon distribution function. If $f_{\vec{k}\lambda}$ is taken to be the thermal-equilibrium (Bose-Einstein) distribution (19), then the expressions (30) reduce to the thermal-equilibrium expression (20)–(22). More generally, if $f_{\vec{k}\lambda}$ is assumed to be *any* spherically symmetric function of $\vec{k}$ independent of polarization $\lambda$, so that it can be written as a function of $\omega_k$,

$$f_{\vec{k}\lambda} = f(\omega_k/\epsilon) = f(yx)\,, \tag{31}$$

where $x = \omega/\omega_0$ as before, $\epsilon$ is some natural energy scale (any parameter of the dimensions of energy[10]), and $y = \omega_0/\epsilon$, then Eqs. (30) reduce to

$$\Delta^{\text{env}} = \frac{4\omega_0^3 d^2}{3\pi c^3} I(y)\,,\qquad \gamma^{\text{env}} = \frac{4\omega_0^3 d^2}{3c^3} f(y)\,, \tag{32}$$

with $I(y)$ defined by (22). This generalizes (20)–(22) to a spherically symmetric nonequilibrium photon distribution function $f(y)$. Another interesting choice for $f_{\vec{k}\lambda}$ is that typical of a single-mode laser,

$$f_{\vec{k}\lambda} = f\delta(\vec{k}-\vec{k}_0)\delta_{\lambda 1}\,, \tag{33}$$

where $f$ is a real, positive constant. Then (30) reduces with (3), to

$$\Delta^{\text{env}} = \frac{f^2\omega_0^2(\vec{d}\cdot\vec{e}_{\vec{k}_0})^2}{2\pi^2 ck_0}\mathscr{P}\left[\frac{1}{\omega_0-ck_0} + \frac{1}{\omega_0+ck_0}\right],\qquad \gamma^{\text{env}} = \frac{f^2\omega_0^2(\vec{d}\cdot\vec{e}_{\vec{k}_0})^2}{2\pi ck_0}\delta(\omega_0-ck_0)\,. \tag{34}$$

The shift exhibits a resonance behavior as the laser frequency $ck_0$ approaches the atomic transition frequency, whereas the width is zero if $ck_0 \neq \omega_0$ but infinite when $ck_0 = \omega_0$. Since dissipation is a result of coupling to a continuum, the vanishing of $\gamma^{\text{env}}$ when $ck_0 \neq \omega_0$ is to be expected, a laser lasing in a single mode being a discrete state rather than a continuum. The singularity at $ck_0 = \omega_0$ is an artifact of the simplified ansatz (33). A real laser has nonzero intensity in a narrow but nonzero range $\epsilon$ of wave vectors, so that $\gamma^{\text{env}}$ will be small for $|k_0-\omega_0/c| \gtrsim \epsilon$ and large for $|k_0-\omega_0/c| \lesssim \epsilon$.

We conclude this section by establishing the connection of the expressions (8), (11), and (16) with a previously described[11] diagrammatic approach to the evaluation of Liouvillian self-energies. The "$\alpha$-irreducible self-energy diagrams" defined there are, for the Hamiltonian (1) and to second order in the interaction $\hat{H}'$, those shown in Figs. 2 and 3. Lines directed toward the right stand for the atomic lowering operator $\hat{a}$ (solid line) or photon annihilation operator $\hat{b}_{\vec{k}\lambda}$ (wavy line); lines directed toward the left stand for the atomic raising operator $\hat{a}^\dagger$ (solid line) or photon creation operator $\hat{b}^\dagger_{\vec{k}\lambda}$ (wavy line). Each vertex stands for one operation of the interaction Liouvillian $\mathscr{L}'$ (commutation with $\hat{H}'$), which gives a matrix-element factor from the expression (1). The ordering of lines and vertices from

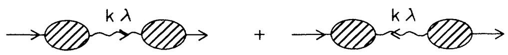


FIG. 2. Irreducible self-energy diagrams contributing to the natural shift and width $\Delta^{\rm WW}$ and $\gamma^{\rm WW}$ to second order.

left to right corresponds to the ordering of factors from right to left in the product

$$\mathscr{L}'\mathscr{G}_0(z)\mathscr{L}'\hat{a}\ , \tag{35}$$

where $\mathscr{G}_0(z)=(z-\mathscr{L}_0)^{-1}$. The diagrams of Fig. 2 yield a transition self-energy contribution

$$\Sigma^{\rm WW}(z)=\sum_{\lambda}(2\pi)^{-3}\Omega \times \int d^3k\, M^2_{\vec{k}\lambda}\left[\frac{1}{z-\omega_k}+\frac{1}{z+\omega_k}\right] \tag{36}$$

which agrees with (11) if we put $z=\omega_0+i\eta$, a replacement which is justified through second order in the interaction matrix element $M_{\vec{k}\lambda}$. Similarly, the diagrams of Fig. 3 yield[11,12]

$$\Sigma^{\rm env}(z)=\sum_{\lambda}(2\pi)^{-3}\Omega\int d^3k\, 2M^2_{\vec{k}\lambda}\left[\frac{1}{z-\omega_k}+\frac{1}{z+\omega_k}\right]\langle \hat{b}^{\dagger}_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}\rangle\ . \tag{37}$$

This agrees with (16) if we replace $z$ by $\omega_0+i\eta$ and note (19), replacements which are again correct to second order in $M_{\vec{k}\lambda}$. Note that the natural line shift and width expression (36) comes from the "completely diagonal" diagrams of Fig. 2, while the environmental contribution (37) comes from the "quasidiagonal" diagrams of Fig. 3.

## III. POWER RADIATED IN THE DECAY OF AN EXCITATION

Standard expressions[13–16] for spectral line shape in terms of current-current correlation functions $\langle \vec{J}(\vec{r},t)\vec{J}(\vec{r}',t')\rangle$ or dipole autocorrelation functions $\langle \vec{d}(t)\vec{d}(t')\rangle$ involve a number of assumptions that we wish to avoid here, in order to obtain a more general nonequilibrium theory. These include the assumptions that (a) the average indicated by angular brackets is taken in an *equilibrium* ensemble; (b) the line shape sought is that for *absorption* of *external* radiation which acts as a perturbing probe; (c) this perturbation (hence the incident radiation intensity) is small; and (d) the semiclassical theory of radiation may be used, according to which absorption, induced emission, and spontaneous emission are treated separately. We wish to give up all of these assumptions here. We are interested, in the first place, in the electromagnetic radiation produced by the decay of some specific kind of excitation, created by some excitation operator $\hat{A}^{\dagger}_{\alpha}$ as in the preceding paper[1] and in the example considered in Sec. II of this paper. On the other hand, the standard formulas involve total absorption summed over all types of excitation that might be created by a weak external electromagnetic field which is treated classically. The excitation created by the operator $\hat{A}^{\dagger}_{\alpha}$ is, in fact, quantized and hence not necessarily "small". Furthermore, we wish to consider the case where this excitation is relative to some initial state of the system which may itself be far from equilibrium, described by some nonequilibrium statistical ensemble. One can then hope to use the calculated line shape as a diagnostic of the nonequilibrium state of the medium, which influences the dynamics of the decay and hence the line shape. Finally, we wish to treat the radiation field by quantum electrodynamics so as to avoid an artificial separation of spontaneous and induced emission, which are both contributions from the same quantum-dynamical process.

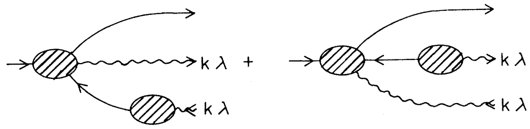


FIG. 3. Irreducible self-energy diagrams contributing to the environmental shift and width $\Delta^{\rm env}$ and $\gamma^{\rm env}$ to second order.

In order to obtain a sufficiently general theory, it is best to return to first principles. Determination of the time-dependent radiated power $P(t)$ is more straightforward in principle than that of the power spectrum, so we shall consider the former here. Let $\hat{H}_{\rm rad}$ be the portion of the total Hamiltonian representing the energy of the quantized radiation field:

$$\hat{H}_{\rm rad}=\sum_{\vec{k},\lambda}\omega_k\hat{N}_{\vec{k}\lambda}\ . \tag{38}$$

Here $\hat{N}_{\vec{k}\lambda}$ is the occupation-number operator $\hat{b}^{\dagger}_{\vec{k}\lambda}\hat{b}_{\vec{k}\lambda}$ for photons of wave vector $\vec{k}$ and polarization $\lambda$. The rest of the notation for $\hat{H}_{\rm rad}$ is as in Eq. (1), but the Hamiltonian of matter and matter-radiation interaction is general, not restricted to the special case of the two-level model of Eq. (1) and Sec. II. Suppose that the system is initially in some pure quantum state $|\psi_0\rangle$ and is then excited, at time $t=0$, into some nonstationary, decaying state $\hat{A}_\alpha^\dagger|\psi_0\rangle$, where $\hat{A}_\alpha^\dagger$ is an appropriate excitation creation operator, as in the preceding paper.[1] Assuming that this excitation couples to the electromagnetic radiation field, decay of the state $\hat{A}_\alpha^\dagger|\psi_0\rangle$ will result in a time-dependent increase in the energy of the quantized radiation field. The instantaneous power $P_\alpha(t)$ being radiated at time $t$ due to decay of the excitation $\alpha$ is

$$P_\alpha(t)=\partial_t\langle\psi_0|\hat{A}_\alpha\hat{H}_{\rm rad}(t)\hat{A}_\alpha^\dagger|\psi_0\rangle$$
$$=\sum_{\vec{k},\lambda}\omega_k\langle\psi_0|\hat{A}_\alpha\partial_t\hat{N}_{\vec{k},\lambda}(t)\hat{A}_\alpha^\dagger|\psi_0\rangle\ ({\rm ps})\ , \tag{39}$$

(where ps represents pure state) where the Heisenberg operators $\hat{H}_{\rm rad}(t)$ and $\hat{N}_{\vec{k}\lambda}(t)$ are propagated with the full Hamiltonian including the matter-radiation interaction and any other interactions contributing to the decay of the excitation. This pure-state expression is generalized to quantum statistical mechanics by replacing the expectation value $\langle\psi_0|\hat{O}|\psi_0\rangle$ by an ensemble average

$$\langle\hat{O}\rangle={\rm Tr}(\hat{O}\hat{\rho}) \tag{40}$$

over an equilibrium or nonequilibrium ensemble. Then

$$P_\alpha(t)=\sum_{\vec{k},\lambda}\omega_k\langle\hat{A}_\alpha\partial_t\hat{N}_{\vec{k}\lambda}(t)\hat{A}_\alpha^\dagger\rangle\ . \tag{41}$$

This expression is quite general, not being restricted to dipole approximation, far zone, equilibrium, etc. It includes, in principle, all spontaneous, induced-emission, and induced-absorption contributions. If $\hat{A}_\alpha^\dagger$ is truly an excitation operator, then $P_\alpha(t)$ is expected to be positive, at least for most values of $t$. However, there is no reason, in principle, to exclude the case that $\hat{A}_\alpha^\dagger$ is a *de*excitation operator, in which case $P_\alpha(t)$ is expected to be negative and to represent induced absorption. More generally, $\hat{A}_\alpha^\dagger$ could be taken to be a projection operator describing any well-defined state preparation. We are, however, assuming that all sources of radiation and any other mechanisms affecting the radiation process are included dynamically in some total, conserved, time-independent Hamiltonian $\hat{H}$ used to propagate the Heisenberg operators, rather than being described in the external-field approximation by parametric time-dependent fields.

Recalling that the Heisenberg operators required in (41) can be written as

$$\hat{N}_{\vec{k}\lambda}(t)=e^{-i\mathscr{L}t}\hat{N}_{\vec{k}\lambda} \tag{42}$$

in terms of the Liouvillian $\mathscr{L}$ [see[1] Eq. (I.64)], one can write (41) formally as

$$P_\alpha(t)=-i\sum_{\vec{k},\lambda}\omega_k\langle\hat{A}_\alpha(e^{-i\mathscr{L}t}\mathscr{L}\hat{N}_{\vec{k}\lambda})\hat{A}_\alpha^\dagger\rangle\ . \tag{43}$$

$\mathscr{L}\hat{N}_{\vec{k}\lambda}$ can be expanded in terms of an operator basis $\{\hat{B}_n\}$, as in[1] (I.8):

$$\mathscr{L}\hat{N}_{\vec{k}\lambda}=[\hat{N}_{\vec{k}\lambda},\hat{H}]=\sum_n c(\vec{k}\lambda|n)\hat{B}_n\ . \tag{44}$$

Then

$$P_\alpha(t)=-i\sum_{\vec{k},\lambda}\sum_n\omega_k c(\vec{k}\lambda|n)\langle\hat{A}_\alpha(e^{-i\mathscr{L}t}\hat{B}_n)\hat{A}_\alpha^\dagger\rangle\ . \tag{45}$$

One has the contour integral representation

$$e^{-i\mathscr{L}t}=(2\pi i)^{-1}\oint dz\, e^{-izt}(z-\mathscr{L})^{-1}\ , \tag{46}$$

where the contour encircles all eigenvalues of $\mathscr{L}$ in the counterclockwise sense. Since these all lie on the real axis, the contour is as shown in Fig. 4. Then

$$P_\alpha(t)=-(2\pi)^{-1}\sum_{\vec{k},\lambda}\sum_n\omega_k c(\vec{k}\lambda|n)\times\oint\tilde{g}_{\alpha n}(z)e^{-izt}dz\ , \tag{47}$$

where the Green's functions $\tilde{g}_{\alpha n}$ are

$$\tilde{g}_{\alpha n}(z)=\langle\hat{A}_\alpha[(z-\mathscr{L})^{-1}\hat{B}_n]\hat{A}_\alpha^\dagger\rangle\ . \tag{48}$$

For a macroscopic dissipative system, these are expected to have cuts along the real axis, as do the simpler Green's functions of the preceding paper.[1] Then the integrals in (47) with the contour of Fig. 4 can be written in terms of the discontinuities across the cut:

$$P_\alpha(t)=(2\pi i)^{-1}\sum_{\vec{k},\lambda}\sum_n\omega_k c(\vec{k}\lambda|n)\times\int_{-\infty}^{\infty}\psi_{\alpha n}(\omega')e^{-i\omega' t}d\omega'\ . \tag{49}$$

Here $\psi_{\alpha n}$ is the cut discontinuity function

$$\psi_{\alpha n}(\omega')=i[\tilde{g}_{\alpha n}(\omega'+i\eta)-\tilde{g}_{\alpha n}(\omega'-i\eta)]\ , \tag{50}$$

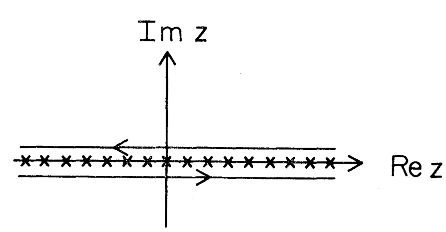


FIG. 4. Contour for the integral (46).

with $\eta=0+$.

A hierarchy of coupled Liouvillian Green's-function equations similar to Eqs. (I.17) can be derived from the identity

$$\begin{aligned}\mathscr{G}(z)&=\mathscr{G}_0(z)+\mathscr{G}_0(z)\mathscr{L}'\mathscr{G}_0(z)\\&\quad+\mathscr{G}_0(z)\mathscr{L}'\mathscr{G}_0(z)\mathscr{L}'\mathscr{G}_0(z)+\cdots\\&=\mathscr{G}_0(z)+\mathscr{G}(z)\mathscr{L}'\mathscr{G}_0(z)\,,\end{aligned} \tag{51}$$

with $\mathscr{G}(z)=(z-\mathscr{L})^{-1}$ and $\mathscr{G}_0(z)=(z-\mathscr{L}_0)^{-1}$. The decomposition of $\mathscr{L}$ into unperturbed and perturbation parts, $\mathscr{L}_0$ and $\mathscr{L}'$, is defined by the decomposition into diagonal and off-diagonal parts with respect to the operator basis $\{\hat{B}_n\}$:

$$\mathscr{L}\hat{B}_n=\epsilon_n\hat{B}_n+\sum_m{}' c(n\mid m)\hat{B}_m\equiv\mathscr{L}_0\hat{B}_n+\mathscr{L}'\hat{B}_n \tag{52}$$

[see Eqs. (I.8) and (I.14)]. Here $\epsilon_n=c(n\mid n)$. The $\hat{B}_n$ are eigenoperators of $\mathscr{L}_0$ with real eigenvalues $\epsilon_n$:

$$\mathscr{L}_0\hat{B}_n=\epsilon_n\hat{B}_n\,. \tag{53}$$

It is convenient to choose the basis such that

$$\begin{aligned}&\mathscr{L}\hat{B}_n=[\hat{B}_n,\hat{H}]=\mathscr{L}_0\hat{B}_n+\mathscr{L}'\hat{B}_n\,,\\&\mathscr{L}_0\hat{B}_n=[\hat{B}_n,\hat{H}_0],\quad \mathscr{L}'\hat{B}_n=[\hat{B}_n,\hat{H}']\,,\end{aligned} \tag{54}$$

where the interaction Hamiltonian $\hat{H}'$ includes the matter-radiation interaction and any other interactions contributing to the decay of the excitation $\alpha$. Since $\hat{H}_0$ is diagonal with respect to (commutes with) the photon occupation-number operators $\hat{N}_{\vec{k}\lambda}$, one has

$$\mathscr{L}_0\hat{N}_{\vec{k}\lambda}=0\,, \tag{55}$$

i.e., $\hat{N}_{\vec{k}\lambda}$ is an eigenoperator of $\mathscr{L}_0$ with eigenvalue zero.

Substitution of (53) into (51) yields

$$\begin{aligned}\mathscr{G}(z)\hat{B}_n&=(z-\epsilon_n)^{-1}\hat{B}_n\\&\quad+(z-\epsilon_n)^{-1}\sum_m{}' c(n\mid m)\mathscr{G}(z)\hat{B}_m\end{aligned} \tag{56}$$

and substitution into (48) then yields the hierarchy

$$\begin{aligned}(z-\epsilon_n)\tilde{g}_{\alpha n}(z)&=\langle\hat{A}_\alpha\hat{B}_n\hat{A}_\alpha^\dagger\rangle\\&\quad+\sum_m{}' c(n\mid m)\tilde{g}_{\alpha m}(z)\,.\end{aligned} \tag{57}$$

Note the similarity with Eqs. (I.17).

The unperturbed Green's functions are

$$\tilde{g}^{(0)}_{\alpha n}(z)=\frac{\langle\hat{A}_\alpha\hat{B}_n\hat{A}_\alpha^\dagger\rangle}{z-\epsilon_n}\,. \tag{58}$$

This suggests introduction of a self-energy function $\Sigma_{\alpha n}(z)$ by an ansatz similar to that of Eq. (I.19). However, we shall see in Sec. IV that for the $\tilde{g}_{\alpha n}$ occurring in (47) in the case of the two-level model, the initial values $\langle\hat{A}_\alpha\hat{B}_n\hat{A}_\alpha^\dagger\rangle$ vanish identically, invalidating the ansatz. More generally, they may be very small in some cases, in which case an ansatz of the form (I.19) leads to a poorly convergent expansion for $\Sigma_{\alpha n}(z)$. This problem is easily circumvented by using an iterated expression, writing each $\tilde{g}_{\alpha n}$ in (47) via (57) as

$$\tilde{g}_{\alpha n}(z)=(z-\epsilon_n)^{-1}\left[\langle\hat{A}_\alpha\hat{B}_n\hat{A}_\alpha^\dagger\rangle+\sum_m{}' c(n\mid m)\tilde{g}_{\alpha m}(z)\right]. \tag{59}$$

Even if $\langle\hat{A}_\alpha\hat{B}_n\hat{A}_\alpha^\dagger\rangle$ vanishes or is very small, this will not, in general, be true of the $\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle$ for the relevant $\tilde{g}_{\alpha m}$, and one may then use a self-energy ansatz similar to (I.19) for them:

$$\tilde{g}_{\alpha m}(z)=\frac{\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle}{z-\epsilon_m-\Sigma_{\alpha m}(z)}\,. \tag{60}$$

For example, for the two-level model to be studied in Sec. IV, the leading contributions to the sum over $m$ in (59) will be found to be those involving only two $m$ values, namely, $\hat{B}_m=\hat{N}=\hat{a}^\dagger\hat{a}$ (occupation-number operator for the atomic excitation) and $\hat{B}_m=\hat{1}$ (unit operator). The Green's function for the case $\hat{B}_m=\hat{1}$ can be trivially evaluated in closed form, and that for $\hat{B}_m=\hat{N}$ has a well-defined self-energy representation (60). In the general case (not restricted to two-level model) the corresponding $\tilde{g}_{\alpha m}$ in (57) are those coupling $\tilde{g}_{\alpha n}$ to those $\hat{B}_m$ which are bilinear in the electric current.[17] This is, again, physically satisfying in view of the prominence of current-current correlation functions in standard theories[13–16] of spectral line shape, which we are generalizing here.

The derivation of the explicit expressions for the terms in the expansion

$$\Sigma_{\alpha m}(z)=\Sigma^{(1)}_{\alpha m}(z)+\Sigma^{(2)}_{\alpha m}(z)+\cdots \tag{61}$$

of the self-energy proceeds exactly as in the derivation of the corresponding expressions in the preceding paper[1] for the different but closely related Green's functions defined therein. In fact, the expressions for the various terms in (61) are obtainable from the previously given ones by merely changing the notation appropriately. For the calculations we have in mind, sufficiently accurate results are obtained by terminating the expansion (61) with the second-order term. The first-order term, obtained by adaptation of Eq. (I.42), is

$$\Sigma_{\alpha m}^{(1)}(z)=\sum_{l\in\mathscr{S}_m}{}' \frac{c(m\mid l)\langle\hat{A}_\alpha\hat{B}_l\hat{A}_\alpha^\dagger\rangle}{\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle} +(z-\epsilon_m)\sum_{l\notin\mathscr{S}_m}\frac{c(m\mid l)\langle\hat{A}_\alpha\hat{B}_l\hat{A}_\alpha^\dagger\rangle}{(z-\epsilon_l)\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle}\ . \tag{62}$$

Here $\mathscr{S}_m$ is the set of $l$ values such that $\hat{B}_l$ is $\mathscr{L}_0$ degenerate with $\hat{B}_m$, i.e., has the same $\mathscr{L}_0$ eigenvalue as does $\hat{B}_m$; this corresponds to the previously discussed[1] separation into "resonant" and "nonresonant" contributions. Note that this is not the same as the usual distinction between "rotating-wave" and "counterrotating" contributions; we found in Sec. II and shall find in Sec. IV that resonant contributions to the self-energy in our sense contain *both* of the latter contributions. The expression for $\Sigma_{\alpha m}^{(2)}$ is the analog of (I.55). Omitting the pole term [analogous to first line of (I.55)] which vanishes for a macroscopic system by the same argument as given previously,[1] one has

$$\begin{aligned}\Sigma_{\alpha m}^{(2)}(z)=&\sum_{l\in\mathscr{S}_m}{}'\sum_{p\notin\mathscr{S}_m}\left[\frac{c(m\mid l)c(l\mid p)\langle\hat{A}_\alpha\hat{B}_p\hat{A}_\alpha^\dagger\rangle}{(z-\epsilon_p)\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle}-\frac{2c(m\mid l)c(m\mid p)\langle\hat{A}_\alpha\hat{B}_l\hat{A}_\alpha^\dagger\rangle\langle\hat{A}_\alpha\hat{B}_p\hat{A}_\alpha^\dagger\rangle}{(z-\epsilon_p)\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle^2}\right]\\ &+\sum_{l\notin\mathscr{S}_m}\sum_{p\in\mathscr{S}_m}\frac{c(m\mid l)c(l\mid p)\langle\hat{A}_\alpha\hat{B}_p\hat{A}_\alpha^\dagger\rangle}{(z-\epsilon_l)\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle}\\ &+(z-\epsilon_m)\sum_{l\notin\mathscr{S}_m}\sum_{p\notin\mathscr{S}_m}{}'\left[\frac{c(m\mid l)c(l\mid p)\langle\hat{A}_\alpha\hat{B}_p\hat{A}_\alpha^\dagger\rangle}{(z-\epsilon_l)(z-\epsilon_p)\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle}-\frac{c(m\mid l)c(m\mid p)\langle\hat{A}_\alpha\hat{B}_l\hat{A}_\alpha^\dagger\rangle\langle\hat{A}_\alpha\hat{B}_p\hat{A}_\alpha^\dagger\rangle}{(z-\epsilon_l)(z-\epsilon_p)\langle\hat{A}_\alpha\hat{B}_m\hat{A}_\alpha^\dagger\rangle^2}\right]\ .\end{aligned} \tag{63}$$

Let us conclude this section by considering the problem of the power spectrum of the radiation, i.e., the spectral line shape. This is not expressible directly in terms of the time-dependent radiated power $P_\alpha(t)$. It is, of course, well known that the Michelson-Lorentz[18] line shape corresponds with the approximation

$$P_\alpha(t)\approx\text{const}\times\sin^2(\omega_0 t)e^{-2\gamma t}$$

characteristic of a damped classical radiating dipole; however, there is no simple, general relation between $P_\alpha(t)$ and the line shape. Note, for example, that the line shape is peaked about $\omega\approx\omega_0$, whereas the Fourier transform of the above classical approximation to $P_\alpha(t)$ is peaked about $\omega\approx 2\omega_0$, the power being quadratic in the field. It seems to us that any correct derivation of a general expression for line shape cannot avoid the measurement problem, i.e., it is necessary to have some qualitatively correct model of how the line shape is to be measured. Actual detectors capable of spectral analysis do so by measuring the variation of the degree of resonant excitation of the detector as its resonance frequency $\omega$ is varied.[19] A simple and rather natural model of such a detector is a two-level atom coupled to the electric field in the same way as that of Sec. II, but with an adjustable transition frequency $\omega$. We plan to carry out a detailed analysis of such a model in a future publication. Here, however, we merely wish to point out that one could formally define a line-shape function $F_\alpha(\omega)$ by

$$F_\alpha(\omega)=\text{Re}\,\tilde{g}(\alpha,\omega+i\eta\mid\alpha)\ , \tag{64}$$

where $\tilde{g}(\alpha,z\mid\alpha)$ is the Green's function defined in the preceding paper,[1] on which the calculation of line shift and broadening is based in Sec. II of this paper:

$$\tilde{g}(\alpha z\mid\alpha)=\langle[(z-\mathscr{L})^{-1}\hat{A}_\alpha]\hat{A}_\alpha^\dagger\rangle =\frac{\langle\hat{A}_\alpha\hat{A}_\alpha^\dagger\rangle}{z-\Sigma_\alpha(z)}\ . \tag{65}$$

The self-energy $\Sigma_\alpha(z)$, is likewise, that previously defined,[1] which forms the basis of the evaluation of the line shift and broadening in Sec. II of this paper.

## IV. RADIATED POWER FOR THE TWO-LEVEL MODEL

In the case of the two-level model of Sec. II, $\hat{A}_\alpha^\dagger$ is the atomic raising operator $\hat{a}^\dagger$, which is a Fermi creation operator in the representation used in Eq. (1). Putting $\alpha=1$ to denote the atomic excited state (upper level) as in Sec. II, one has for the instantaneous radiated power at time $t$ [see Eqs. (41), (49), and (50)]

$$P_1(t)=\sum_{\vec{k},\lambda}\omega_k\langle\hat{a}\partial_t\hat{N}_{\vec{k}\lambda}(t)\hat{a}^\dagger\rangle$$
$$=(2\pi i)^{-1}\sum_{\vec{k},\lambda}\sum_n\omega_k c(\vec{k}\lambda\,|\,n)\int_{-\infty}^{\infty}\psi_{1n}(\omega')e^{-i\omega' t}d\omega' , \qquad (66)$$

where $\psi_{1n}$ is the cut discontinuity function

$$\psi_{1n}(\omega')=i[\tilde{g}_{1n}(\omega'+i\eta)-\tilde{g}_{1n}(\omega'-i\eta)] . \qquad (67)$$

The $c(\vec{k}\lambda\,|\,n)$ and $\hat{B}_n$ are determined from the operator-basis expansion

$$\mathscr{L}\hat{N}_{\vec{k}\lambda}=[\hat{N}_{\vec{k}\lambda},\hat{H}]=[\hat{N}_{\vec{k}\lambda},\hat{H}']$$
$$=iM_{\vec{k}\lambda}(\hat{b}^\dagger_{\vec{k}\lambda}-\hat{b}_{\vec{k}\lambda})(\hat{a}-\hat{a}^\dagger) \qquad (68)$$

which implies, with (1), the following operator-basis elements, coefficients, and transition energies:

$$\hat{B}_n=\hat{b}^\dagger_{\vec{k}\lambda}\hat{a}:\ c(\vec{k}\lambda\,|\,n)=iM_{\vec{k}\lambda},\ \ \epsilon_n=\omega_0-\omega_k ,$$
$$\hat{B}_n=\hat{a}^\dagger\hat{b}_{\vec{k}\lambda}:\ c(\vec{k}\lambda\,|\,n)=iM_{\vec{k}\lambda},\ \ \epsilon_n=\omega_k-\omega_0 ,$$
$$\hat{B}_n=\hat{b}_{\vec{k}\lambda}\hat{a}:\ c(\vec{k}\lambda\,|\,n)=-iM_{\vec{k}\lambda},\ \ \epsilon_n=\omega_0+\omega_k ,\qquad (69)$$
$$\hat{B}_n=\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda}:\ c(\vec{k}\lambda\,|\,n)=-iM_{\vec{k}\lambda},\ \ \epsilon_n=-\omega_0-\omega_k .$$

Since

$$\hat{a}(\hat{a}-\hat{a}^\dagger)\hat{a}^\dagger=0 , \qquad (70)$$

it follows that $\langle\hat{a}\hat{B}_n\hat{a}^\dagger\rangle$ vanishes identically (independently of which ensemble is used to evaluate the average) for all the $\hat{B}_n$ occurring in (66). This is the situation discussed in Sec. III, where the $\tilde{g}_{an}$ must first be expressed in terms of $\tilde{g}_{am}$ via Eq. (59) before applying the self-energy representation (60). In the present case Eq. (59) reads

$$\tilde{g}_{1n}(z)=(z-\epsilon_n)^{-1}\sum_m{}'c(n\,|\,m)\tilde{g}_{1m}(z) . \qquad (71)$$

The $c(n\,|\,m)$ and corresponding $\hat{B}_m$ are to be evaluated by taking the commutator of each of the $\hat{B}_n$ of (69) with $\hat{H}'$, as in the similar calculations in Sec. II. For example,

$$[\hat{b}^\dagger_{\vec{k}\lambda}\hat{a},\hat{H}']$$
$$=\sum_m{}'c(\hat{b}^\dagger_{\vec{k}\lambda}\hat{a}\,|\,m)\hat{B}_m$$
$$=-i\sum_{\vec{k}',\lambda'}M_{\vec{k}'\lambda'}\hat{b}^\dagger_{\vec{k}\lambda}(\hat{b}_{\vec{k}'\lambda'}+\hat{b}^\dagger_{\vec{k}'\lambda'})(1-2\hat{N})$$
$$+iM_{\vec{k}\lambda}\hat{N} , \qquad (72)$$

with $\hat{N}=\hat{a}^\dagger\hat{a}$. The terms in the summation over $(\vec{k}',\lambda')$ all have vanishing expectation values in the state $\hat{a}^\dagger|0\rangle$ (unperturbed atomic excited state), whereas $\hat{N}$ has expectation value unity. One therefore expects the terms in the summation $\sum_{\vec{k}',\lambda'}$ to give contributions to $\tilde{g}_{1n}$ which are of higher order in the interaction than the contribution from the term proportional to $N$, so we shall only retain the latter.[20] The same argument applies to the case $\hat{B}_n=\hat{a}^\dagger\hat{b}_{\vec{k}\lambda}$ and also to $\hat{b}_{\vec{k}\lambda}\hat{a}$ and $\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda}$, except that in the latter two cases there are contributions proportional to the unit operator $\hat{1}$ which must be retained as well as those from $\hat{N}$. One thus obtains the following leading-order expressions for the $\tilde{g}_{1n}$ of Eqs. (66), (67), and (71):

$$\hat{B}_n=\hat{b}^\dagger_{\vec{k}\lambda}\hat{a}:\ \tilde{g}_{1n}(z)\approx(z-\omega_0+\omega_k)^{-1}iM_{\vec{k}\lambda}\tilde{g}_{1a}(z) ,$$
$$\hat{B}_n=\hat{a}^\dagger\hat{b}_{\vec{k}\lambda}:\ \tilde{g}_{1n}(z)\approx(z-\omega_k+\omega_0)^{-1}iM_{\vec{k}\lambda}\tilde{g}_{1a}(z) ,$$
$$\hat{B}_n=\hat{b}_{\vec{k}\lambda}\hat{a}:\ \tilde{g}_{1n}(z)\approx(z-\omega_0-\omega_k)^{-1}iM_{\vec{k}\lambda}\tilde{g}_{1a}(z)-(z-\omega_0-\omega_k)^{-1}iM_{\vec{k}\lambda}z^{-1}(1-\langle\hat{N}\rangle) , \qquad (73)$$
$$\hat{B}_n=\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda}:\ \tilde{g}_{1n}(z)\approx(z+\omega_0+\omega_k)^{-1}iM_{\vec{k}\lambda}\tilde{g}_{1a}(z)-(z+\omega_0+\omega_k)^{-1}iM_{\vec{k}\lambda}z^{-1}(1-\langle\hat{N}\rangle) .$$

Here $\tilde{g}_{1a}$ is the Green's function for the atomic excitation occupation-number operator $\hat{N}$,

$$\tilde{g}_{1a}(z)=\langle\hat{a}[(z-\mathscr{L})^{-1}\hat{N}]\hat{a}^\dagger\rangle , \qquad (74)$$

and the corresponding Green's function for the unit operator is

$$\langle\hat{a}[(z-\mathscr{L})^{-1}\hat{1}]\hat{a}^\dagger\rangle=z^{-1}\langle\hat{a}\hat{a}^\dagger\rangle$$
$$=z^{-1}(1-\langle\hat{N}\rangle) \qquad (75)$$

in which $\langle\hat{N}\rangle$ may be interpreted as the excitation probability of the atom in the given ensemble, which vanishes in the unperturbed vacuum and hence arises only from the radiative interaction (vacuum fluctuations) and environmental effects. Equations (66), (67), (69), and (73) imply

$$P_1(t)\approx(2\pi)^{-1}\sum_{\vec{k},\lambda}\omega_k M^2_{\vec{k}\lambda}\int_{-\infty}^{\infty}d\omega' e^{-i\omega' t}$$
$$\times\Bigg[\tilde{g}_{1a}(\omega'+i\eta)\left[-\frac{1}{\omega'-\omega_0+\omega_k+i\eta}-\frac{1}{\omega'-\omega_k+\omega_0+i\eta}+\frac{1}{\omega'-\omega_0-\omega_k+i\eta}+\frac{1}{\omega'+\omega_0+\omega_k+i\eta}\right]$$
$$+\tilde{g}_{1a}(\omega'-i\eta)\left[\frac{1}{\omega'-\omega_0+\omega_k-i\eta}+\frac{1}{\omega'-\omega_k+\omega_0-i\eta}-\frac{1}{\omega'-\omega_0-\omega_k-i\eta}-\frac{1}{\omega'+\omega_0+\omega_k-i\eta}\right]$$
$$-\left[\frac{1-\langle\hat{N}\rangle}{\omega'+i\eta}\right]\left[\frac{1}{\omega'-\omega_0-\omega_k+i\eta}+\frac{1}{\omega'+\omega_0+\omega_k+i\eta}\right]$$
$$+\left[\frac{1-\langle\hat{N}\rangle}{\omega'-i\eta}\right]\left[\frac{1}{\omega'-\omega_0-\omega_k-i\eta}+\frac{1}{\omega'+\omega_0+\omega_k-i\eta}\right]\Bigg]\ . \tag{76}$$

We assume (and shall presently verify) that the discontinuity of $\tilde{g}_{1a}(z)$ across its cut on the real axis consists of a change of sign of its imaginary part, the real part remaining continuous. Then

$$\tilde{g}_{1a}(\omega'\pm i\eta)=\tilde{g}'_{1a}(\omega')\mp i\tilde{g}''_{1a}(\omega')\ , \tag{77}$$

where $\tilde{g}_{1a'}$ and $\tilde{g}''_{1a}$ are real. Inserting this into (76) along with the standard relation

$$\frac{1}{x\pm i\eta}=\mathscr{P}\frac{1}{x}\mp i\pi\delta(x)\ , \tag{78}$$

one finds that the expression reduces to

$$P_1(t)\approx i\sum_{\vec{k},\lambda}\omega_k M^2_{\vec{k}\lambda}\int_{-\infty}^{\infty}d\omega' e^{-i\omega' t}$$
$$\times\Bigg[\tilde{g}'_{1a}(\omega')[\delta(\omega'-\omega_0+\omega_k)+\delta(\omega'-\omega_k+\omega_0)-\delta(\omega'-\omega_0-\omega_k)-\delta(\omega'+\omega_0+\omega_k)]$$
$$+\pi^{-1}\tilde{g}''_{1a}(\omega')\left[\mathscr{P}\frac{1}{\omega'-\omega_0+\omega_k}+\mathscr{P}\frac{1}{\omega'-\omega_k+\omega_0}+\mathscr{P}\frac{1}{\omega'-\omega_0-\omega_k}-\mathscr{P}\frac{1}{\omega'+\omega_0+\omega_k}\right]$$
$$+(1-\langle\hat{N}\rangle)\mathscr{P}\frac{1}{\omega'}[\delta(\omega'-\omega_0-\omega_k)+\delta(\omega'+\omega_0+\omega_k)]\Bigg]\ . \tag{79}$$

The integrations over $\omega'$ can be easily carried out explicitly. Those involving the $\delta$ functions are trivial:

$$\int_{-\infty}^{\infty}d\omega' e^{-i\omega' t}\tilde{g}'_{1a}(\omega')[\delta(\omega'-\omega_0+\omega_k)+\delta(\omega'-\omega_k-\omega_0)-\delta(\omega'-\omega_0-\omega_k)-\delta(\omega'+\omega_0+\omega_k)]$$
$$=-2i\tilde{g}'_{1a}(\omega_0-\omega_k)\sin[(\omega_0-\omega_k)t]+2i\tilde{g}'_{1a}(\omega_0+\omega_k)\sin[(\omega_0+\omega_k)t] \tag{80}$$

and

$$\int_{-\infty}^{\infty} d\omega' e^{-i\omega' t}(1-\langle \hat{N} \rangle)\mathscr{P}\frac{1}{\omega'}[\delta(\omega'-\omega_0-\omega_k)+\delta(\omega'+\omega_k+\omega_k)] = -2i(1-\langle \hat{N} \rangle)\mathscr{P}\frac{1}{\omega_0+\omega_k}\sin[(\omega_0+\omega_k)t] \ . \tag{81}$$

Here we have assumed that $\tilde{g}'_{1a}$ is an odd function. We shall show presently that this is the case, and that $\tilde{g}''_{1a}$ is even:

$$\begin{aligned} \tilde{g}'_{1a}(-\omega) &= -\tilde{g}'_{1a}(\omega) \ , \\ \tilde{g}''_{1a}(-\omega) &= \tilde{g}''_{1a}(\omega) \ . \end{aligned} \tag{82}$$

The integrals involving the principal-part functions can be evaluated by contour integration. Recalling that $t \geq 0$, one can close the contour in the lower half-plane, as shown in Fig. 5. The indentations avoid the singularities at $\omega' = \omega_0 \pm \omega_k$ and $\omega' = -\omega_0 \pm \omega_k$. We shall find later that $\tilde{g}''_{1a}(\omega')$ has poles at $\omega' = \pm i\Gamma$, where $\Gamma$ is a real quantity which will be evaluated later to leading order in the radiative interaction. The contour encircles (in the negative sense) the pole at $\omega' = -i\Gamma$, but there are no other singularities within the contour, and $|\tilde{g}''_{1a}|$ is of order $|\omega'|^{-1}$ for $|\omega'| \to \infty$. Then the semicircle at infinity contributes nothing, whereas the integral along the indented real axis is the sum of the desired principal-value integral and half of the pole contributions from $\omega' = \omega_0 \pm \omega_k$ and $\omega' = -\omega_0 \pm \omega_k$.

In this way one finds

$$\begin{aligned} \frac{1}{\pi}\mathscr{P}\int_{-\infty}^{\infty} d\omega' e^{-i\omega' t}\tilde{g}''_{1a}(\omega') &\left[\frac{1}{\omega'-\omega_0+\omega_k}+\frac{1}{\omega'-\omega_k+\omega_0}-\frac{1}{\omega'-\omega_0-\omega_k}-\frac{1}{\omega'+\omega_0+\omega_k}\right] \\ &= -2ie^{-\Gamma t}\mathscr{R}''_{1a}(-i\Gamma)2i\Gamma\left[\frac{1}{(\omega_0-\omega_k)^2+\Gamma^2}-\frac{1}{(\omega_0+\omega_k)^2+\Gamma^2}\right] \\ &\quad -2i\tilde{g}''_{1a}(\omega_0-\omega_k)\cos[(\omega_0-\omega_k)t]+2i\tilde{g}''_{1a}(\omega_0+\omega_k)\cos[(\omega_0+\omega_k)t] \ , \end{aligned} \tag{83}$$

where $\mathscr{R}''_{1a}(-i\Gamma)$ is the residue of $\tilde{g}''_{1a}(\omega')$ at its pole $\omega' = -i\Gamma$. Use has been made of the symmetry property (82) of $\tilde{g}''_{1a}$.

The sum over $(\vec{k},\lambda)$ in (79) can be reduced, in the infinite-system limit, to an integral over $\omega = \omega_k$ according to

$$\sum_{\vec{k},\lambda} \omega_k M^2_{\vec{k}\lambda} f(\omega_k) = \frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2 \int_0^{mc^2} \omega^2 f(\omega)d\omega \ , \tag{84}$$

as in the derivation of Eq. (14). The high-frequency cutoff at $\omega = mc^2$ has been introduced as in the derivation of the Lamb-shift expression (15). Insertion of the results (80), (81), and (83) for the $\omega'$ integrals yields

$$\begin{aligned} P_1(t) \approx \frac{16\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2 \int_0^{mc^2} \omega^2 \Bigg[ &\tilde{g}'_{1a}(\omega_0-\omega)\sin[(\omega_0-\omega)t]-\tilde{g}'_{1a}(\omega_0+\omega)\sin[(\omega_0+\omega)t] \\ &+2i\Gamma e^{-i\Gamma t}\mathscr{R}''_{1a}(-i\Gamma)\left[\frac{1}{(\omega_0-\omega)^2+\Gamma^2}-\frac{1}{(\omega_0+\omega)^2+\Gamma^2}\right] \\ &+\tilde{g}''_{1a}(\omega_0-\omega)\cos[(\omega_0-\omega)t]-\tilde{g}''_{1a}(\omega_0+\omega)\cos[(\omega_0+\omega)t] \\ &+(1-\langle \hat{N} \rangle)\mathscr{P}\frac{1}{\omega_0+\omega}\sin[(\omega_0+\omega)t]\Bigg] d\omega \ . \end{aligned} \tag{85}$$

The term proportional to $\mathscr{R}''_{1a}(-i\Gamma)$ can be evaluated to leading order by contour integration, assuming $\omega_0 \ll mc^2$ and $\gamma^{WW} \ll \omega_0$, where $\gamma^{WW}$ is the natural line width (14). Then, as shown in Appendix C, the dominant contribution comes from the pole at $\omega = \omega_0 + i\Gamma$, yielding

$$\int_0^{mc^2} d\omega\, \omega^2 \left[2i\Gamma e^{-\Gamma t}\mathscr{R}''_{1a}(-i\Gamma)\left[\frac{1}{(\omega_0-\omega)^2+\Gamma^2}-\frac{1}{(\omega_0+\omega)^2+\Gamma^2}\right]\right] \approx 2\pi i\omega_0^2 e^{-i\Gamma t}\mathscr{R}''_{1a}(-i\Gamma) \ . \tag{86}$$

The last integral in (85) is expressible in terms of the sine integral[21] and trigonometric functions:

$$\int_0^{mc^2} \omega^2 \frac{\sin[(\omega_0+\omega)t]}{\omega_0+\omega} d\omega = \int_{\omega_0}^{mc^2+\omega_0} (\omega'-\omega_0)^2 \frac{\sin(\omega' t)}{\omega'} d\omega'$$
$$= \omega_0^2[\mathrm{Si}((mc^2+\omega_0)t)-\mathrm{Si}(\omega_0 t)]+2\omega_0 t^{-1}[\cos((mc^2+\omega_0)t)-\cos(\omega_0 t)]$$
$$+t^{-2}[\sin((mc^2+\omega_0)t)-(mc^2+\omega_0)t\cos((mc^2+\omega_0)t)$$
$$-\sin(\omega_0 t)+\omega_0 t\cos(\omega_0 t)] \ . \tag{87}$$

In order to analyze the contributions of the other terms in (85) it is necessary to evaluate the leading contributions to the functions $\tilde{g}'_{1a}(\omega')$ and $\tilde{g}''_{1a}(\omega')$. First write the Green's function $\tilde{g}_{1a}(z)$ of Eq. (74) in the form (60) exhibiting the self-energy $\Sigma_{1a}(z)$:

$$\tilde{g}_{1a}(z)=\frac{1-\langle \hat{N}\rangle}{z-\Sigma_{1a}(z)} \ , \tag{88}$$

use having been made of the identities

$$\langle \hat{a}\hat{N}\hat{a}^\dagger\rangle = \langle (\hat{a}\hat{a}^\dagger)^2\rangle$$
$$= \langle (1-\hat{N})^2\rangle = \langle 1-\hat{N}\rangle = 1-\langle \hat{N}\rangle \ . \tag{89}$$

Decomposing the self-energy of the real axis in analogy with (77),

$$\Sigma_{1a}(\omega\pm i\eta)=\Sigma'_{1a}(\omega)\mp i\Sigma''_{1a}(\omega) \ , \tag{90}$$

with $\Sigma'_{1a}$ and $\Sigma''_{1a}$ real, one finds with (88) and (77)

$$\tilde{g}'_{1a}(\omega)=\frac{(1-\langle\hat{N}\rangle)[\omega-\Sigma'_{1a}(\omega)]}{[\omega-\Sigma'_{1a}(\omega)]^2+[\Sigma''_{1a}(\omega)]^2} \ ,$$
$$\tilde{g}''_{1a}(\omega)=\frac{(1-\langle\hat{N}\rangle)\Sigma''_{1a}(\omega)}{[\omega-\Sigma'_{1a}(\omega)]^2+[\Sigma''_{1a}(\omega)]^2} \ . \tag{91}$$

$\Sigma_{1a}$, and hence $\Sigma'_{1a}$ and $\Sigma''_{1a}$, can be evaluated up to second order in the radiative interaction from the expressions (62) and (63) for the case $\hat{B}_m=\hat{N},\hat{A}_\alpha=\hat{a}$. The expression (62) for the first-order, self-energy specializes in the present case to

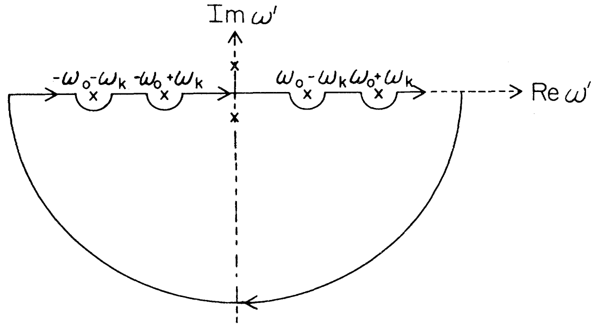


FIG. 5. Contour for the integral (83).

$$\Sigma^{(1)}_{1a}(z)=\sum_{n\in\mathscr{S}_0}{}' \frac{c(a\mid n)\langle \hat{a}\hat{B}_n\hat{a}^\dagger\rangle}{\langle \hat{a}\hat{N}\hat{a}^\dagger\rangle}$$
$$+z\sum_{n\notin\mathscr{S}_0}\frac{c(a\mid n)\langle \hat{a}\hat{B}_n\hat{a}^\dagger\rangle}{\langle \hat{a}\hat{N}\hat{a}^\dagger\rangle} \ , \tag{92}$$

where $\mathscr{S}_0$ is the set of $n$ such that $\hat{B}_n$ are $\mathscr{L}_0$ degenerate with $\hat{N}$, i.e., such $\hat{B}_n$ that have $\mathscr{L}_0$ eigenvalues zero.[22] The coefficients ($c$-matrix elements) $c(\alpha\mid n)$ and corresponding operator-basis elements $\hat{B}_n$ are read off from the identity

$$[\hat{N},\hat{H}]=[\hat{N},\hat{H}']=\sum_n c(a\mid n)\hat{B}_n$$
$$=-i\sum_{\vec{k},\lambda} M_{\vec{k}\lambda}(\hat{b}_{\vec{k}\lambda}+\hat{b}^\dagger_{\vec{k}\lambda})(\hat{a}+\hat{a}^\dagger) \tag{93}$$

analogous to (68), use having been made of the explicit expression (1) for the Hamiltonian. One has in comparison with (69)

$$\hat{B}_n=\hat{b}^\dagger_{\vec{k}\lambda}\hat{a}:\ c(a\mid n)=-iM_{\vec{k}\lambda},\ \epsilon_n=\omega_0-\omega_k \ ,$$
$$\hat{B}_n=\hat{a}^\dagger\hat{b}_{\vec{k}\lambda}:\ c(a\mid n)=-iM_{\vec{k}\lambda},\ \epsilon_n=\omega_k-\omega_0 \ ,$$
$$\hat{B}_n=\hat{b}_{\vec{k}\lambda}\hat{a}:\ c(a\mid n)=-iM_{\vec{k}\lambda},\ \epsilon_n=\omega_0+\omega_k \ ,$$
$$\hat{B}_n=\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda}:\ c(a\mid n)=-iM_{\vec{k}\lambda},\ \epsilon_n=-\omega_0-\omega_k \ . \tag{94}$$

As in the case considered in Sec. II, $\mathscr{S}_0$ is a "set of measure zero" with the consequence that the first line of the expression for $\Sigma^{(1)}_{1a}$ vanishes in the macroscopic limit $\Omega\to\infty$ [see the discussion before Eq. (8)]. The second line of (92) vanishes indentically in view of an identity differing from (70) only in the sign between $\hat{a}$ and $\hat{a}^\dagger$.

Next consider the second-order self-energy, Eq. (63) with $\hat{B}_m=\hat{N}$ and $\hat{A}_\alpha=\hat{a}$. The terms from the first double summation are negligible $[O(\Omega^{-1})]$ as were those in the first line of (108) for the same reason. Those from the last double summation in (63) are negligible for a different reason, namely, their prefactor[23] $z$. Since the two $c$-matrix elements imply that these terms are of second order in the interaction (second order in the $M_{\vec{k}\lambda}$), it follows that these terms will be smaller than the term $z$ in the denominator of (88) by a small factor of order $\alpha=e^2/\hbar c$. This argument does not apply to the terms from the middle double summation in (63), which contain no prefactor $z$. It follows that these terms will dominate when $|z|\le\alpha$. But it is precisely such small values of $z$ that are important for the long-time behavior of the radiated power, since the large-$t$ behavior of a Fourier transform is determined by the low-frequency behavior of its integrand.

We conclude, then, that the self-energy $\Sigma_{1a}$ reduces, through second order, to

$$\Sigma_{1a}(z)=\sum_{n\notin\mathscr{S}_0}\sum_{m\in\mathscr{S}_0}\frac{c(a\mid n)c(n\mid m)\langle\hat{a}\hat{B}_m\hat{a}^\dagger\rangle}{(z-\epsilon_n)\langle\hat{a}\hat{N}\hat{a}^\dagger\rangle}\ . \tag{95}$$

The $c(a\mid n)$ and $\hat{B}_n$ are given in (94) and the $c(n\mid m)$ and $\hat{B}_m$ are determined by taking the commutator of each of these $\hat{B}_n$ with $\hat{H}'$, Eq. (1), as in the derivation of Eqs. (11) and (16). The resultant $\hat{B}_m$ with $m\in\mathscr{S}_0$ are $\hat{N}$, the unit operator $\hat{1}$, the $\hat{N}_{\vec{k}\lambda}$, and the products $\hat{N}_{\vec{k}\lambda}\hat{N}$. The derivation parallels that in Sec. II, so we shall only give the result

$$\Sigma_{1a}(z)=\sum_{\vec{k},\lambda}M^2_{\vec{k}\lambda}\left[\frac{1}{z-\omega_0+\omega_k}+\frac{1}{z-\omega_k+\omega_0}\right]$$
$$+\sum_{\vec{k},\lambda}M^2_{\vec{k}\lambda}\left[\frac{1}{z-\omega_0+\omega_k}+\frac{1}{z-\omega_k+\omega_0}+\frac{1}{z-\omega_0-\omega_k}+\frac{1}{z+\omega_0+\omega_k}\right]\frac{\langle\hat{N}_{\vec{k}\lambda}(1-\hat{N})\rangle}{\langle 1-\hat{N}\rangle}\ . \tag{96}$$

In obtaining this result identities like (89), which follow from the fact that $\hat{a}$ and $\hat{a}^\dagger$ are Fermi operators, have been used.

The first summation is independent of the environment (independent of the ensemble[24]) as was the generalized Weisskopf-Wigner line shift and width expression of Eq. (11). This contribution is directly related to the spontaneous emission, and will therefore be denoted herein by $\Sigma^{\rm spon}_{1a}$. The second summation in (96) is an environmental[25] contribution similar to the environmental line shift and width expression (1b), and will therefore be denoted by $\Sigma^{\rm env}_{1a}$. Thus

$$\Sigma_{1a}(z)=\Sigma^{\rm spon}_{1a}(z)+\Sigma^{\rm env}_{1a}(z)\ . \tag{97}$$

Changing $\Sigma^{\rm spon}_{1a}$ to an integral in accordance with (84) and making use of (78) as in Eqs. (11) and (14), one finds[26]

$$\Sigma^{\rm spon}_{1a}(\omega\pm i\eta)=\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2\left\{(\omega_0-\omega)\left[\vartheta(\omega_0-\omega<0)\ln\left[\frac{mc^2}{\omega-\omega_0}+1\right]\right.\right.$$
$$\left.+\vartheta(0<\omega_0-\omega<mc^2)\ln\left[\frac{mc^2}{\omega_0-\omega}-1\right]+\vartheta(\omega_0-\omega>mc^2)\ln\left[1-\frac{mc^2}{\omega_0-\omega}\right]\right]$$
$$-(\omega_0+\omega)\left[\vartheta(\omega_0+\omega<0)\ln\left[\frac{mc^2}{-\omega_0-\omega}+1\right]\right.$$
$$+\vartheta(0<\omega_0+\omega<mc^2)\ln\left[\frac{mc^2}{\omega_0+\omega}-1\right]$$
$$\left.\left.+\vartheta(\omega_0+\omega>mc^2)\ln\left[1-\frac{mc^2}{\omega_0+\omega}\right]\right]\right\}$$
$$\mp i\pi\left[\frac{8\pi}{3c^3}\right]\left[\frac{\omega_0 d}{2\pi}\right]^2[(\omega_0-\omega)\vartheta(0<\omega_0-\omega<mc^2)+(\omega_0+\omega)\vartheta(0<\omega_0+\omega<mc^2)]\ , \tag{98}$$

where the $\vartheta$ function of an inequality is defined to be unity if the inequality is satisfied, and otherwise zero.

Let us consider first the case in which the environmental contribution $\Sigma^{\rm env}_{1a}$ in (97) is negligible compared to the spontaneous decay contribution $\Sigma^{\rm spon}_{1a}$. This is the case, for example, in thermal equilibrium at zero temperature. When $\Sigma^{\rm env}_{1a}$ is evaluated, we shall find that at zero temperature it reduces to a vacuum field fluctuation contribution which is very small compared to $\Sigma^{\rm spon}_{1a}$, due to the smallness of the virtual vacuum photon distribution function.[25] Then putting $\Sigma_{1a}=\Sigma_{1a}^{\rm spon}$, one can determine $\tilde{g}'_{1a}$ and $\tilde{g}''_{1a}$ from (90), (91), and (94). The integrals over $\tilde{g}'_{1a}$ and $\tilde{g}''_{1a}$ in (85) have to be subdivided into integrals over various ranges of $\omega$, due to the $\theta$ functions in (98). In the range $0<\omega<\omega_0$ one has

$$\Sigma'_{1a}(\omega)=\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2\left[(\omega_0-\omega)\ln\left[\frac{mc^2}{\omega_0-\omega}-1\right]-(\omega_0+\omega)\ln\left[\frac{mc^2}{\omega_0+\omega}-1\right]\right],$$
$$\Sigma''_{1a}(\omega)=\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2 2\pi\omega_0=\Sigma''_{1a}(0),\quad 0<\omega<\omega_0\,. \tag{99}$$

One sees that $\Sigma'_a(\omega)$ vanishes as $\omega\to 0$, and for $\omega\gg\omega_0^3d^2/c^3$ it is negligible compared to $\omega$. One may therefore drop $\Sigma'_{1a}$ in Eq. (91) in this range and write

$$\tilde{g}'_{1a}(\omega)\approx\frac{(1-\langle\hat{N}\rangle)\omega}{\omega^2+[\Sigma''_{1a}(0)]^2},$$
$$\tilde{g}''_{1a}(\omega)\approx\frac{(1-\langle\hat{N}\rangle)\Sigma''_{1a}(0)}{\omega^2+[\Sigma''_{1a}(0)]^2},\quad 0<\omega<\omega_0\,. \tag{100}$$

Note that $\tilde{g}''_{1a}$ becomes large of order $[\Sigma''_{1a}(0)]^{-1}$ for $\omega\ll\Sigma''_{1a}(0)$, and $\tilde{g}'_{1a}$ is large of the same order for $\omega\sim\Sigma''_{1a}(0)$, although it vanishes as $\omega\to 0$. Note also, from (99), that $\Sigma''_{1a}(0)=2\gamma^{\rm WW}$, so the width of the frequency region in which $\tilde{g}'_1$ and $\tilde{g}''_1$ become large is of the order of the natural line width. Then the corresponding range of photon energies for which $\tilde{g}'_{1a}(\omega_0-\omega)$ and $\tilde{g}''_{1a}(\omega_0-\omega)$ become large is of width $\sim\gamma^{\rm WW}$ centered on $\omega\approx\omega_0$, as expected physically. This suggests that the dominant contributions to (85) comes from these terms, in the range $|\omega-\omega_0|\lesssim\gamma^{\rm WW}$, and we shall presently verify this expectation. Note also that it follows from (100) that the quantity $\Gamma$ in (83), (85), and (86) is just $\Sigma''_{1a}(0)=2\gamma^{\rm WW}$ to leading order, and that the corresponding residue $\mathscr{R}''_{1a}(-i\Gamma)$ is $(1/2)i(1-\langle\hat{N}\rangle)$.

Arguments similar to those leading to (100) can be applied in the other subintervals of $\omega$, leading to the following expressions for $\tilde{g}'_{1a}$ and $\tilde{g}''_{1a}$ in these various subintervals:

$$\tilde{g}'_{1a}(\omega)\approx\frac{(1-\langle\hat{N}\rangle)}{\omega},$$
$$\tilde{g}''_{1a}(\omega)\approx\frac{(1-\langle\hat{N}\rangle)\Sigma''_{1a}(\omega)}{\omega^2},$$
$$\Sigma''_{1a}(\omega)=\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2\pi(\omega_0+\omega),$$
$$\omega_0<\omega<mc^2-\omega_0\,, \tag{101}$$

and

$$\tilde{g}''_{1a}(\omega)\approx\frac{1-\langle\hat{N}\rangle}{\omega},\quad \tilde{g}''_{1a}(\omega)=0\,,$$
$$\omega>mc^2-\omega_0\,. \tag{102}$$

In determining the boundaries of the subdivisions, it has been assumed that $\omega_0<(1/2)mc^2$. One can then write the integrals in (85) involving $\tilde{g}'_{1a}$ as

$$\int_0^{mc^2}\omega^2\{\tilde{g}'_{1a}(\omega_0-\omega)\sin[(\omega_0-\omega)t]-\tilde{g}'_{1a}(\omega_0+\omega)\sin[(\omega_0+\omega)t]\}d\omega$$
$$\approx(1-\langle\hat{N}\rangle)\int_0^{2\omega_0}\frac{\omega^2(\omega_0-\omega)}{(\omega_0-\omega)^2+[\Sigma''_{1a}(0)]^2}\sin[(\omega_0-\omega)t]d\omega$$
$$+(1-\langle\hat{N}\rangle)\int_{2\omega_0}^{mc^2}\frac{\omega^2\sin[(\omega_0-\omega)t]}{\omega_0-\omega}d\omega-(1-\langle\hat{N}\rangle)\int_0^{mc^2}\frac{\omega^2\sin[(\omega_0+\omega)t]}{\omega_0+\omega}d\omega\,, \tag{103}$$

where use has been made of the symmetry properties (82), which follow from (17), (91), and (98). One finds similarly that the integrals involing $\tilde{g}''_{1a}$ are

$$\int_0^{mc^2}\omega^2\{\tilde{g}''_{1a}(\omega_0-\omega)\cos[(\omega_0-\omega)t]-\tilde{g}''_{1a}(\omega_0+\omega)\cos[(\omega_0+\omega)t]\}d\omega$$
$$\approx(1-\langle\hat{N}\rangle)\Sigma''_{1a}(0)\int_0^{2\omega_0}\frac{\omega^2\cos[(\omega_0-\omega)t]}{(\omega_0-\omega)^2+[\Sigma''_{1a}(0)]^2}d\omega$$
$$+(1-\langle\hat{N}\rangle)\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2\pi\int_{2\omega_0}^{mc^2}\frac{\omega^3\cos[(\omega_0-\omega)t]}{(\omega_0-\omega)^2}d\omega$$
$$-(1-\langle\hat{N}\rangle)\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2\pi\int_0^{mc^2-2\omega_0}\frac{\omega^2(2\omega_0+\omega)\cos[(\omega_0+\omega)t]}{(\omega_0+\omega)^2}\,. \tag{104}$$

The last integral in (103) cancels the last one in (85) so the contribution (87) is not actually present in the expression for $P_1(t)$. The second integral in (103) differs from (87) only in the upper limit of integration:

$$\int_{2\omega_0}^{mc^2} \frac{\omega^2 \sin[(\omega_0-\omega)t]}{\omega_0-\omega}d\omega = \int_{\omega_0}^{mc^2-\omega_0} (\omega'-\omega_0)\frac{\sin(\omega' t)}{\omega'}d\omega'$$

$$= \omega_0^2[\mathrm{Si}(tmc^2-\omega_0)t)-\mathrm{Si}(\omega_0 t)] + 2\omega_0 t^{-1}[\cos((mc^2-\omega_0)t)-\cos(\omega_0 t)]$$
$$+t^{-2}[\sin((mc^2-\omega_0)t)-(mc^2-\omega_0)t\cos((mc^2-\omega_0)t)$$
$$-\sin(\omega_0 t)+\omega_0 t\cos(\omega_0 t)] \ . \tag{105}$$

The last two integrals in (104) can be combined and expressed in terms of the cosine integral[21] and the sine. However, the result will not be exhibited since these contributions to (85) are smaller than the contributions (105) by factors of order $(\gamma^{\mathrm{WW}} \mid \omega_0) \ll 1$. Here use has been made of the connection between $\Sigma''_{1a}(0)$ and the spontaneous linewidth $\gamma^{\mathrm{WW}}$, namely, $\Sigma''_{1a}(0)=2\gamma^{\mathrm{WW}}$; see Eqs. (99) and (14). The first integrals in (103) and (104) can be expressed in terms of explicitly evaluable contour integrals and integrals like (105) apart from terms negligible compared to those retained. This is done in Appendix C; the resultant expressions are

$$\int_0^{2\omega_0} \frac{\omega^2(\omega_0-\omega)\sin[(\omega_0-\omega)t]}{(\omega_0-\omega)^2+[\Sigma''_{1a}(0)]^2}d\omega$$

$$\approx \pi\omega_0^2\{e^{-2\gamma^{\mathrm{WW}}t}+2\pi^{-1}[\mathrm{Si}(\omega_0 t)-\pi/2]+2\pi^{-1}(\omega_0 t)^{-2}[\sin(\omega_0 t)-\omega_0 t\cos(\omega_0 t)]\} \tag{106}$$

and

$$\int_0^{2\omega_0} \frac{\omega^2\cos[(\omega_0-\omega)t]}{(\omega_0-\omega)^2+[\Sigma''_{1a}(0)]^2}d\omega \approx \tfrac{1}{2}\pi\omega_0^2(\gamma^{\mathrm{WW}})^{-1}e^{-2\gamma^{\mathrm{WW}}t} \ . \tag{107}$$

The only approximations made in evaluation of the integrals (106) and (107) consist of neglect of terms of order $(\gamma^{\mathrm{WW}} \mid \omega_0) \ll 1$ relative to those retained.

Combining the various results (100), (103)—(107), (86) and (87) and substituting into (85), one finds the following expression for the radiated power:

$$P_1^{\mathrm{spon}}(t) \approx 2\gamma^{\mathrm{WW}}\omega_0(1-\langle\hat{N}\rangle)\{e^{-2\gamma^{\mathrm{WW}}t}+\pi^{-1}[\mathrm{Si}((mc^2-\omega_0)t)+\mathrm{Si}(\omega_0 t)-\pi]$$
$$+2(\pi\omega_0 t)^{-1}[\cos((mc^2-\omega_0)t)-\cos(\omega_0 t)]$$
$$+\pi^{-1}(\omega_0 t)^{-2}[\sin((mc^2-\omega_0)t)-(mc^2-\omega_0)t\cos((mc^2-\omega_0)t)$$
$$+\sin(\omega_0 t)-\omega_0 t\cos(\omega_0 t)]\} \ . \tag{108}$$

The superscript "spon" is a reminder that we are considering here the case in which the environmental contribution to the self-energy is negligible compared with the spontaneous decay contribution [see Eqs. (96) and (97)]. The expression (108) has rather different behaviors in various different regimes of time. Consider first its initial ($t=0$) value. Using the limiting behavior of the sine integral[21] one finds that

$$P_1^{\mathrm{spon}}(0)=0 \ . \tag{109}$$

In fact, this can be shown to be a rigorous consequence of (43) in the case of the two-level model, which implies with (68)

$$P_1(0)=i\sum_{\vec{k},\lambda}\omega_k\langle\hat{a}[\hat{N}_{\vec{k}\lambda},\hat{H}]\hat{a}^\dagger\rangle$$
$$=\sum_{\vec{k},\lambda}\omega_k M_{\vec{k}\lambda}\langle\hat{a}(\hat{b}^\dagger_{\vec{k}\lambda}\hat{a}-\hat{a}^\dagger\hat{b}_{\vec{k}\lambda})\hat{a}^\dagger\rangle=0 \tag{110}$$

independently of the above statistical ensemble average indicated by angular brackets, since $\hat{a}^2$ and $(\hat{a}^\dagger)^2$ vanish identically; see also Eq. (70) and the following discussion. The result (109) can be regarded as a check on the derivation of (108). Next consider the behavior for small but nonzero time. The terms with argument $(mc^2-\omega_0)t$ cause $P_1^{\rm spon}(t)$ to increase extremely rapidly initially [rise time $\sim\omega_0^2/(mc^2)^3$], the behavior for $t \ll (mc^2)^{-1}$ being[27]

$$P_1^{\rm spon}(t) \approx (2\gamma^{\rm WW}/3\pi\omega_0)(mc^2)^3(1-\langle\hat{N}\rangle)t\,, \qquad t \ll (mc^2)^{-1}\,. \tag{111}$$

For $t\sim(mc^2)^{-1}$ the expression (109) oscillates extremely rapidly [frequency $(mc^2-\omega_0)\approx mc^2$] with very large amplitude of order

$$\gamma^{\rm WW}\omega_0(mc^2/\omega_0)^2\,,$$

taking on both positive and negative instantaneous values. These oscillations decay like $t^{-1}$, the behavior for $t \gg (mc^2)^{-1}$ being

$$P_1^{\rm spon}(t) \approx 2\gamma^{\rm WW}\omega_0(1-\langle\hat{N}\rangle)\left[\frac{1}{2}-\left[\frac{mc^2}{\omega_0}\right]^2\frac{\cos((mc^2-\omega_0)t)}{\pi mc^2 t}\right],\quad (mc^2)^{-1} \ll t \ll \omega_0^{-1}\,. \tag{112}$$

The dominant oscillatory term[28] in this regime of time is the one involving $\cos((mc^2-\omega_0)t)$ in the last line of (108); those from $\mathrm{Si}((mc^2-\omega_0)t)$ and the other sine and cosine terms with the same argument are smaller by factors of order $\omega_0/mc^2$. Note that the oscillation is still very violent [amplitude $\gg(mc^2/\omega_0)\gg 1$], of very large amplitude compared to the term $\frac{1}{2}$ in (112). Note also that $P_1^{\rm spon}$ still does not remain instantaneously positive in this time interval in view of this large amplitude of oscillation. However, the average of (112) over a time large compared with $(mc^2)^{-1}$ is positive, and $(mc^2)^{-1} \ll \omega_0^{-1}$ is an extremely short time interval. For $t\sim\omega_0^{-1}$ the oscillatory terms with argument $\omega_0 t$ also become important. In fact, all terms in (108) are important in this range of time; the function $\mathrm{Si}(\omega_0 t)$ is only given numerically in this range. The oscillations take place about a mean value $e^{-2\gamma^{\rm WW}t}\approx 1$. The amplitude of oscillation of the cosine term in (112) is still large compared to the terms with frequency $\omega_0$ for $t\sim\omega_0^{-1}$. For $t\gg\omega_0^{-1}$ the terms will frequency $\omega_0$ become negligible and

$$P_1(t) \approx 2\gamma^{\rm WW}\omega_0(1-\langle\hat{N}\rangle)\left[e^{-2\gamma^{\rm WW}t}-\left[\frac{mc^2}{\omega_0}\right]^2\frac{\cos((mc^2-\omega_0)t)}{\pi mc^2 t}\right],\quad t\gg\omega_0^{-1}\,. \tag{113}$$

The exponential decays away for $t\gg(\gamma^{\rm WW})^{-1}$ (natural lifetime), leaving the oscillatory nonexponential tail.

We turn now to the question of the physical interpretation of this very complicated behavior of the instantaneous radiated power. Note first that the nonexponential tail at large times and nonexponential behavior at very small times are in agreement with the well-known fact[29] that pure exponential decay is impossible in Hamiltonian quantum mechanics for both very small and very large values of the time. The nonexponential oscillatory terms average to zero over time intervals large compared to $\omega_0^{-1} \ll (\gamma^{\rm WW})^{-1}$. The time integral of the exponential term is

$$\int_0^\infty dt\, 2\gamma^{\rm WW}\omega_0(1-\langle\hat{N}\rangle)e^{-2\gamma^{\rm WW}t} = \omega_0(1-\langle\hat{N}\rangle) \tag{114}$$

which is, in leading order,[30] just the transition energy of the two-level atom. This is in agreement with the fact that the total radiated energy must equal the transition energy in order that energy conservation hold. The oscillatory terms with frequency $\omega_0$ are very short-lived transients which are expected on general grounds.[29] The terms with frequency $mc^2-\omega_0$ are more difficult to interpret. Two possibilities suggest themselves: (a) These terms are artifacts of the sharp frequency cutoff at $mc^2$, or (b) they are related to a real physical effect connected with the relativistic *Zitterbewegung*. Only a relativistic calculation can decide between these alternatives; we hope to investigate this in the future. If such very high-frequency terms are real, their detailed forms would almost certainly be changed by a relativistic calculation. Finally, note that there is no sign in (108) of a $\sin^2(\omega_0 t)$ modulation of the exponential decay, in contrast with the situation for a classical radiating dipole.[31] The behavior of the quantum system over very short-time intervals is very different from that of the classical system. However, if one averages the radiated power of both over a time interval $\gg\omega_0^{-1}$ but $\ll(\gamma^{\rm WW})^{-1}$ (the radiative lifetime), then one obtains the same exponential decay law for both. We are not aware of any other published results for the time-dependent radiated power for this model, valid over the whole time range $0\le t<\infty$, with which we can compare our result (108). There are, however, several published treatments of this model based on solution of Heisenberg equations of motion,[32,33] which are close in spirit to our Liouville-space treatment. (See note added in proof.) The results of these other treatments agree with ours insofar as the exponentially decaying term is concerned; they also find no $\sin^2(\omega_0 t)$ modulation.

We have thus far limited ourselves to the case where the environmental contribution to the self-energy (97) is negligible. Suppose now that it is not. Then the general form of the environmental[25] contribution to the self-energy [second term in (96)] is not difficult to find, and we shall evaluate it here. However, it follows from (91) that the environmental and spontaneous contributions to $\tilde{g}'_{1a}$ and $\tilde{g}''_{1a}$, hence to the radiated power, are not additive. We shall therefore defer the detailed analysis of such cases to later publications. The situation simplifies again when the spontaneous contributions become negligible compared with the environmental ones, i.e., when the environmental effects dominate the decay. Here we shall, however, limit ourselves to the determination of the general form of the expression for $\Sigma_{1a}^{\rm env}$. We assume that the ensemble is such that the mean value in (96) factorizes,[34]

$$\langle \hat{N}_{\vec{k}\lambda}(1-\hat{N})\rangle = \langle \hat{N}_{\vec{k}\lambda}\rangle\langle 1-\hat{N}\rangle = f_{\vec{k}\lambda}\langle 1-\hat{N}\rangle \tag{115}$$

as in (18), (19), and (29). Then

$$\Sigma_{1a}^{\rm env}(\omega\pm i\eta)=\sum_{\vec{k},\lambda} M_{\vec{k}\lambda}^2\left[\mathscr{P}\frac{1}{\omega-\omega_0-\omega_k}+\mathscr{P}\frac{1}{\omega-\omega_0+\omega_k}+\mathscr{P}\frac{1}{\omega+\omega_0-\omega_k}+\mathscr{P}\frac{1}{\omega+\omega_0+\omega_k}\right]f_{\vec{k}\lambda}$$
$$\mp i\pi\sum_{\vec{k},\lambda} M_{\vec{k}\lambda}^2[\delta(\omega-\omega_0-\omega_k)+\delta(\omega-\omega_0+\omega_k)+\delta(\omega+\omega_0-\omega_k)+\delta(\omega+\omega_0+\omega_k)]f_{\vec{k}\lambda}\,, \tag{116}$$

where $f_{\vec{k}\lambda}$ is the photon distribution function $\langle \hat{N}_{\vec{k}\lambda}\rangle$. In the thermal-equilibrium case (19), it is straightforward to reduce (116) to integrals of the same type occurring in Eqs. (20). We shall omit the derivation, which proceeds as in Sec. II, and only state the results:

$$\Sigma'_{1a,\rm env}(\omega)=\frac{8\pi}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2[(\omega-\omega_0)I(\beta\,|\,\omega-\omega_0\,|\,)+(\omega+\omega_0)I(\beta\,|\,\omega+\omega_0\,|\,)] \tag{117}$$

and

$$\Sigma''_{1a,\rm env}(\omega)=\frac{4\pi^2}{3c^3}\left[\frac{\omega_0 d}{2\pi}\right]^2\left[\frac{(\omega-\omega_0)\sinh[\beta(\omega-\omega_0)]}{\sinh^2[\frac{1}{2}\beta(\omega-\omega_0)]}+\frac{(\omega+\omega_0)\sinh[\beta(\omega+\omega_0)]}{\sinh^2[\frac{1}{2}\beta(\omega+\omega_0)]}\right], \tag{118}$$

where $\Sigma'_{1a,\rm env}$ and $\Sigma''_{1a,\rm env}$ are defined in terms of the real and imaginary parts of $\Sigma_{1a}^{\rm env}$ as in (90). The integral $I(y)$ is defined in Eq. (22) and expressed in terms of the digamma function in Appendix A. Note that $\Sigma'_{1a,\rm env}$ and $\Sigma''_{1a,\rm env}$ are, respectively, odd and even functions of $\omega$.

The expressions (117) and (118) are rather similar to some that were obtained in a study of thermal effects on the absorption spectrum of a two-level system coupled to a crystal lattice, by Huber and Van Vleck.[35] The similarity is not surprising, in view of the similarity of the models. Their treatment employed standard equilibrium thermal Green's functions, whereas our Liouville-space approach is equally applicable to nonequilbrium ensembles, although the specific expressions (117) and (118) are for the special case of equilibrium.

The asymptotic behavior of the functions $I$ and $f$, given in Eqs. (23) and (24), implies that the environmental contributions (117) and (118) to the self-energy are negligible compared to the spontaneous ones for $k_BT \ll \omega_0$, but become important for $k_BT \geq \omega_0$. The latter situation can occur, e.g., for Rydberg states of atoms.[7] We shall not carry the analysis further herein, but hope to come back to it in subsequent publications. Nonequilibrium situations of the sort considered in Sec. II can be analyzed by choosing appropriate nonequilibrium photon distributions $f_{\vec{k}\lambda}$ in (116).

*Note added in proof.* Since submission of this paper we have become aware of a Liouville-space treatment of the three-level model: R. Kornblith and J. H. Eberly, J. Phys. B 11, 1545 (1978). We thank Professor Eberly for bringing this work to our attention.

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## APPENDIX A: EVALUATION OF THE INTEGRAL $I(y)$

The integral $I(y)$ defined by Eq. (22), which occurs in the expression for the environmental line shift of the two-level atom interacting with thermal radiation, is expressible in terms of the digamma ($\psi$) function

$$\psi(z)=d\ln\Gamma(z)/dz\ , \tag{A1}$$

where $\Gamma(z)$ is the gamma function. Using the formula (I.59) one has

$$\begin{aligned} I(y)&=\mathrm{Re}\int_0^\infty\left[\frac{1}{1+i\eta-x}+\frac{1}{1+i\eta+x}\right]\frac{x}{e^{yx}-1}dx \\ &=-2\,\mathrm{Re}\left[(1+i\eta)\int_0^\infty\frac{x\,dx}{[x^2-(1+i\eta)^2](e^{yx}-1)}\right]\ , \end{aligned}\tag{A2}$$

where $\eta=0+$. Let

$$yx=2\pi t,\quad z=-i(1+i\eta)(y/2\pi)=(-i+\eta)(y/2\pi)\ . \tag{A3}$$

Then

$$\int_0^\infty\frac{x\,dx}{[x^2-(1+i\eta)^2](e^{yx}-1)}=\int_0^\infty\frac{t\,dt}{(t^2+z^2)(e^{2\pi t}-1)}\ . \tag{A4}$$

Then by a standard formula[36]

$$I(y)=\mathrm{Re}\{(1+i\eta)[\psi(z)-\ln z+\tfrac{1}{2}z^{-1}]\}\ . \tag{A5}$$

One has with (A3)

$$\begin{aligned} \ln z&=\ln|z|+i\arg z \\ &\underset{\eta\to0+}{\longrightarrow}\ln(y/2\pi)-i\pi/2\ , \\ z&\underset{\eta\to0+}{\longrightarrow}-iy/2\pi,\quad z^{-1}\underset{\eta\to0+}{\longrightarrow}i2\pi/y\ . \end{aligned}\tag{A6}$$

It follows from[36] NBS Eqs. (6.3.10), (6.3.11), (6.3.17), and (6.3.19) that $\psi(z)$ remains finite as $\eta\to0+$, so that the prefactor $i\eta$ in (A5) may be dropped. Then

$$I(y)=\mathrm{Re}\psi(-iy/2\pi)-\ln(y/2\pi)\ . \tag{A7}$$

Then by NBS Eqs. (6.3.10), (6.3.17), and (6.3.19) one has for small $y$

$$\begin{aligned} I(y)=&-\ln(y/2\pi)+1-\gamma-[1+(y/2\pi)^2]^{-1} \\ &+\sum_{n=1}^{\infty}(-1)^{n+1}[\zeta(2n+1)-1](y/2\pi)^{2n}\ , \end{aligned}\qquad y<4\pi \tag{A8}$$

for all $y$

$$\begin{aligned} I(y)=&-\ln(y/2\pi)-\gamma \\ &+(y/2\pi)^2\sum_{n=1}^{\infty}n^{-1}[n^2+(y/2\pi)^2]^{-1}\ , \end{aligned}\tag{A9}$$

and for large $y$

$$\begin{aligned} I(y)&\sim\sum_{n=1}^{\infty}\frac{(-1)^{n-1}B_{2n}}{2n}(2\pi/y)^{2n} \\ &=\tfrac{1}{12}(2\pi/y)^2+\tfrac{1}{120}(2\pi/y)^4 \\ &\quad+\tfrac{1}{252}(2\pi/y)^6+\cdots\ . \end{aligned}\tag{A10}$$

Here $\gamma$ is Euler's constant 0.577 21..., the $B_{2n}$ are Bernoulli numbers [NBS (Ref. 36) Chap. 23], and the $\zeta(2n+1)$ are Riemann zeta functions (NBS edition,[36] Chap. 23). The small-$y$ expansion (A8) still converges rather rapidly at $y=2\pi$; see NBS[36] p. 811 for the necessary $\zeta(2n+1)$. The series (A9) converges fairly rapidly for all $y$, the rate of convergence increasing slowly with increasing $y$. The asymptotic series (A10) is useful only for $y\gg2\pi$. A numerical table for Re $\psi(1+iy)$ is given at the bottom of NBS[36] p. 288. Note the function needed in Eq. (A7) is related to $\mathrm{Re}\psi(1+iy)$ by [NBS[36] Eq. (6.3.10)]

$$\mathrm{Re}\psi(-iy)=\mathrm{Re}\psi(1+iy)\ . \tag{A11}$$

In Eq. (20), the shift $\Delta^{\mathrm{env}}$ comes from the real part of the expression in curly brackets in (A5), whereas the width $\gamma^{\mathrm{env}}$ comes from the imaginary part:

$$\gamma^{\mathrm{env}}=-\frac{4\omega_0^3d^2}{3\pi c^3}\mathrm{Im}[\psi(z)-\ln z+\tfrac{1}{2}z^{-1}]\ . \tag{A12}$$

By (A6), (A7), and NBS (Refs. 36) Eq. (6.3.11) one has

$$\mathrm{Im}[\psi(z)-\ln z+\tfrac{1}{2}z^{-1}]=-\frac{\pi}{e^y-1}\ . \tag{A13}$$

Substitution into (A12) yields an expression in agreement with the second Eq. (20) when one notes $y=\beta\omega_0$ and Eq. (21). This is a check of the derivation of (A5).

As mentioned in the text, in the paper by Farley and Wing,[7] an integral similar to Eq. (22) is found to describe the dependence of the dynamic Stark shift on the blackbody spectral distribution. They compute the integral

$$F(y)=\int_0^\infty dx\frac{x^3}{e^x-1}\left[\frac{1}{y+x}+\frac{1}{y-x}\right] \tag{A14}$$

numerically; however, it is possible to derive an analytic expression for $F(y)$, analagous to the derivation (A7). First note that $F(-y)=-F(y)$. We shall assume that for $y$ positive in (A14), $y$ is really $\lim_{\eta\to 0+}(y-i\eta)$, the $i\eta$ coming from boundary conditions implicit in the problem. We then make the substitution $u=x^2$ to reexpress

$$F(y)=-y\int_0^\infty du\frac{u}{(e^{\sqrt{u}}-1)[u+(iy)^2]}\ . \tag{A15}$$

We next use the property of Stieltjes transforms[37] to rewrite this as

$$F(y)=-y\int_0^\infty\frac{du}{e^{\sqrt{u}}-1} -y^3\int_0^\infty\frac{du}{(e^{\sqrt{u}}-1)(u-y^2)}\ . \tag{A16}$$

We are interested in the real part of (A16); note that the second integral is essentially Eq. (22) when reexpressed in terms of $x=\sqrt{u}$. The first integral gives $\pi^2/3$ and we have

$$F(y)=-\frac{\pi^2 y}{3}-y^3\ln(y/2\pi) +y^3\mathrm{Re}\psi(-iy/2\pi)\ . \tag{A17}$$

Using the same expansions as before we have for $y<4\pi$,

$$F(y)\sim-\frac{\pi^2 y}{3}-y^3\ln(y/2\pi) +y^3(1-\gamma)-\frac{y^3}{1+y^2} +\sum_{n=1}^{\infty}(-1)^{n+1}y^{2n+3}[\zeta(2n+1)-1]\ ; \tag{A18}$$

for all $y$,

$$F(y)=-\frac{\pi^2 y}{3}-y^3\ln(y/2\pi) -\gamma y^3+y^5\sum_{n=1}^{\infty}n^{-1}(n^2+y^2)^{-1}\ ; \tag{A19}$$

and for large $y$,

$$F(y)\sim-\frac{\pi^2 y}{3}+\sum_{n=1}^{\infty}\frac{(-1)^{n-1}B_{2n}}{2ny^{2n-3}} =\frac{2\pi^4}{15y}+\frac{16\pi^6}{63y^3}+\cdots\ . \tag{A20}$$

Equations (A18) and (A20) agree with the expressions given by Farley and Wing for the small- and large-$y$ behavior of $F(y)$, while (A19) corresponds to the results given in their Fig. 1.

## APPENDIX B: STANDARD THERMODYNAMIC GREEN'S-FUNCTION FORMALISM APPLIED TO TWO-LEVEL MODEL

In the case of thermal equilibrium we may, with one important modification, apply standard temperature-time Green's function techniques[15,38] to verify the results derived in the text. The interaction Hamiltonian $\hat{H}'$ in Eq. (1) is linear in the Fermi operators $\hat{a}$ and $\hat{a}^\dagger$. Such a linear Fermi term in a Hamiltonian is usually considered unphysical; however, here it occurs as a result of the two-level approximation where we use the transition operators $\hat{a}$, $\hat{a}^\dagger$. Standard Green's-functions methods are not applicable to Hamiltonians linear in Fermi operators since the definition of time ordering cannot be applied consistently.[39] As shown below, this problem can be circumvented by applying a canonical transformation such that, to each order in a perturbation expansion, the resulting canonically transformed Hamiltonian is bilinear in Fermi operators. Let

$$\begin{gathered}\hat{H}=\hat{H}_0+\lambda\hat{V},\quad \hat{U}=e^{i\hat{W}},\quad \hat{W}=\hat{W}^\dagger\ ,\\ \hat{H}'=e^{i\hat{W}}\hat{H}e^{-i\hat{W}}=\hat{H}'_0+\hat{V}'\ ,\end{gathered} \tag{B1}$$

where $\hat{H}_0$ is as in Eq. (1) while $\lambda\hat{V}$ is $\hat{H}'$ in Eq. (1) and $\lambda$ indicates $M_{\vec{k}\lambda}$. Expand $\hat{W}$ and $\hat{V}'$ perturbatively as

$$\hat{W}=\sum_{n=1}^{\infty}\lambda^n\hat{W}_n,\quad \hat{V}'=\sum_{n=1}^{\infty}\lambda^n\hat{V}_n\ . \tag{B2}$$

We have

$$\hat{H}'=\hat{H}_0+i[\hat{W},\hat{H}_0]+\frac{i^2}{2!}[\hat{W},[\hat{W},\hat{H}_0]]+\cdots +\lambda\left[\hat{V}+i[\hat{W},\hat{V}]+\frac{i^2}{2!}[\hat{W},[\hat{W},\hat{V}]]+\cdots\right] \tag{B3}$$

which give

$$\hat{V}'_1 = i[\hat{W}_1,\hat{H}_0]+\hat{V}\ ,$$

$$\hat{V}'_2 = i[\hat{W}_2,\hat{H}_0]+\frac{i^2}{2!}[\hat{W}_1,[\hat{W}_1,\hat{H}_0]] + i[\hat{W}_1,\hat{V}]\ . \tag{B4}$$

Since $\hat{V}$ is odd in Fermi and $\hat{H}_0$ even, choose $\hat{W}_1$ odd such that $\hat{V}'_1=0$ and $[\hat{W}_1,\hat{H}_0]=i\hat{V}$. We then have to second order (which is all we need)

$$\hat{V}'_2 = i[\hat{W}_2,\hat{H}_0]+\tfrac{1}{2}[\hat{W}_1,[\hat{W}_1,\hat{H}_0]] = \tfrac{1}{2}i[\hat{W}_1,\hat{V}],\ \text{for}\ \hat{W}_2=0\ , \tag{B5}$$

and $\hat{H}'$ is now strictly bilinear in Fermi operators.

In order to calculate equilibrium expectation values using this transformation we must transform both the density matrix and the operator which we are taking the expectation value of:

$$\begin{aligned}\langle\hat{O}\rangle = \mathrm{Tr}(\hat{\rho}\hat{O}) &= \mathrm{Tr}[(e^{i\hat{W}}\hat{\rho}e^{-i\hat{W}})(e^{i\hat{W}}\hat{O}e^{-i\hat{W}})]\\ &= \mathrm{Tr}(\hat{\rho}'\hat{O}')\\ &= \mathrm{Tr}(\hat{\rho}'\hat{O})+i\lambda\,\mathrm{Tr}\{\hat{\rho}'[\hat{W},\hat{O}]\}\\ &\quad -\frac{i}{2}\lambda^2\,\mathrm{Tr}\{\hat{\rho}'[\hat{W}_1,[\hat{W}_1,\hat{O}]]\}\\ &\quad +O(\lambda^3)\ ,\end{aligned}$$

where

$$\hat{\rho}=\frac{e^{-\beta\hat{H}}}{\mathrm{Tr}e^{-\beta\hat{H}}}\ ,\quad \hat{\rho}'=\frac{e^{-\beta\hat{H}'}}{\mathrm{Tr}e^{-\beta\hat{H}'}}\ . \tag{B6}$$

For the two-level model, some tedious algebra gives the following:

$$\lambda\hat{W}_1 = -\sum_{\vec{k},\lambda}\left[\frac{M_{\vec{k}\lambda}}{\omega_0+\omega_k}(\hat{a}\hat{b}_{\vec{k}\lambda}+\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda})+\frac{M_{\vec{k}\lambda}}{\omega_0-\omega_k}(\hat{a}\hat{b}_{\vec{k}\lambda}+\hat{a}^\dagger\hat{b}^\dagger_{\vec{k}\lambda})\right]\ , \tag{B7}$$

$$\begin{aligned}[\lambda\hat{W}_1,\lambda\hat{V}] = -i\sum\sum{}' MM'[&-(\Omega_++\Omega_-)(\hat{b}\hat{b}'+\hat{b}^\dagger\hat{b}^{\dagger\prime})+(\Omega_+-\Omega_-)(\hat{b}^\dagger\hat{b}'+\hat{b}^{\dagger\prime}\hat{b})-2\delta\Omega_+\\ &+2(\Omega_++\Omega_-)\hat{a}^\dagger\hat{a}(\hat{b}\hat{b}'+\hat{b}^\dagger\hat{b}^{\dagger\prime}+\hat{b}^\dagger\hat{b}'+\hat{b}^{\dagger\prime}\hat{b}+\delta)]\ ,\end{aligned} \tag{B8}$$

where a condensed notation is used:

$$\Omega_+=(\omega_0+\omega)^{-1},\quad \Omega_-=(\omega_0-\omega)^{-1},\quad \delta=\delta_{\vec{k}\vec{k}'}\delta_{\lambda\lambda'}\ ,$$

and the unprimed $\hat{b}^{(\dagger)}$'s and $M$ have $(\vec{k},\lambda)$ indices while the primed have $(\vec{k}',\lambda')$ as indices. In order to calculate the self-energy for the two-level atom we need to compute $\langle T_\tau(\hat{a}(\tau)\hat{a}^\dagger(\tau'))\rangle$, where $T_\tau$ is the usual time-ordering operator. From (B6) we need the following:

$$[\hat{W}_1,\hat{a}] = -\sum M[\Omega_+\hat{b}^\dagger(2\hat{a}^\dagger\hat{a}-1)+\Omega_-\hat{b}(2\hat{a}^\dagger\hat{a}-1)]\ ,$$

$$\begin{aligned}[\hat{W}_1,[\hat{W}_1,\hat{a}]] = \sum\sum{}' MM'[&\Omega_+\Omega'_+(2\hat{a}\hat{b}^\dagger\hat{b}'+\hat{a}\delta-2\hat{a}^\dagger\hat{b}^\dagger\hat{b}^{\dagger\prime})+\Omega'_+\Omega_-(2\hat{a}\hat{b}'\hat{b}-2\hat{a}^\dagger\hat{b}\hat{b}^{\dagger\prime})\\ &+\Omega'_-\Omega_+(2\hat{a}\hat{b}^{\dagger\prime}\hat{b}^\dagger-2\hat{a}^\dagger\hat{b}^\dagger\hat{b}')+\Omega_-\Omega'_-(2\hat{a}\hat{b}^{\dagger\prime}\hat{b}+\hat{a}\delta-2\hat{a}^\dagger\hat{b}\hat{b}')]\ .\end{aligned} \tag{B9}$$

The corresponding expressions for $\hat{a}^\dagger$ are easily related to the above by conjugation. Using these expressions we have correct to $O(M^2)$:

$$\begin{aligned}\mathrm{Tr}\{\hat{\rho}'T_\tau[\hat{a}(\tau)\hat{a}^\dagger(\tau)]\} = \sum\{&-M^2(\Omega_+^2+\Omega_-^2)\coth(\omega\beta/2)\mathscr{D}(\tau-\tau')\\ &+M^2[\tanh(\omega_0\beta/2)]^2[\Omega_+^2\mathscr{S}(\tau-\tau')+\Omega_-^2\mathscr{S}(\tau'-\tau)]\\ &-4M^2[\Omega_+^2\mathscr{S}(\tau'-\tau)\mathscr{D}(\tau-\tau')\mathscr{D}(\tau'-\tau)\\ &\quad+\Omega_-^2\mathscr{S}(\tau-\tau')\mathscr{D}(\tau-\tau')\mathscr{D}(\tau'-\tau)]\}\\ &+\sum M^2\coth(\omega\beta/2)(\Omega_++\Omega_-)\int_0^\beta d\tau_1\mathscr{D}(\tau-\tau_1)\mathscr{D}(\tau_1-\tau')\ .\end{aligned} \tag{B10}$$

The first term (enclosed in curly brackets) comes from the terms (B9) while the second term is the $O(\lambda^2)$ term in the expansion of $(\mathrm{Tr}e^{-\beta\hat{H}'})^{-1}$:

$$[\mathscr{S}(\tau-\tau')]_{\lambda\lambda'}(\vec{k},\vec{k}')=(\mathrm{Tr}e^{-\beta\hat{H}_0})^{-1}\mathrm{Tr}\{e^{-\beta\hat{H}_0}T_\tau[\hat{b}_{0\vec{k}\lambda}(\tau)\hat{b}_{0\vec{k}'\lambda'}(\tau')]\}\ ,$$

$$\mathscr{D}(\tau-\tau')=(\mathrm{Tr}e^{-\beta\hat{H}_0})^{-1}\mathrm{Tr}\{e^{-\beta\hat{H}_0}T_\tau[\hat{a}_0(\tau)\hat{a}_0^\dagger(\tau')]\}\ ,$$

where $\hat{a}_0(\tau)=e^{-\beta\hat{H}_0}\hat{a}(0)e^{\beta\hat{H}_0}$ and the same for $\hat{b}_0$ ($\mathscr{D}$ and $\mathscr{S}$ are just the usual noninteracting temperature Green's functions). The terms in (B10) are all connected; the disconnected terms in the numerator are cancelled as usual by similar terms in $(\mathrm{Tr}e^{-\beta\hat{H}_0})^{-1}$. The expression (B10) is easily evaluated using standard finite-temperature Fourier expansions[38] and we shall just list the results:

$$\begin{aligned}
&\mathscr{D}\mathscr{D}:\ \frac{M^2\coth(\omega\beta/2)}{(i\omega_a-\omega_0)^2(\omega_0^2-\omega^2)}\ ,\\
&\mathscr{S}:\ M^2[\tanh(\omega_0\beta/2)]^2\coth(\omega\beta/2)\left[\frac{\Omega_+^2}{i\omega_a+\omega}+\frac{\Omega_-^2}{i\omega_a-\omega}\right],\\
&\mathscr{S}\mathscr{D}\mathscr{D}:\ M^2\coth(\omega\beta/2)[1-\tanh^2(\omega_0\beta/2)]\left[\frac{\Omega_+^2}{i\omega_a+\omega}+\frac{\Omega_-^2}{i\omega_a-\omega}\right],\\
&\mathscr{D}:\ -M^2\coth(\omega\beta/2)(\Omega_+^2+\Omega_-^2)\ .
\end{aligned}\tag{B11}$$

The total equals

$$\sum_{\vec{k},\lambda}M_{\vec{k}\lambda}^2\frac{\coth(\omega\beta/2)}{(i\omega_a-\omega_0)^2}\left[\frac{1}{i\omega_a+\omega}+\frac{1}{i\omega_a-\omega}\right].\tag{B12}$$

We therefore have for the self-energy

$$\Sigma^{(2)}(\omega_a)=\sum_{\vec{k},\lambda}M_{\vec{k}\lambda}^2\coth[\omega(k)\beta/2]\left[\frac{1}{i\omega_a+\omega(k)}+\frac{1}{i\omega_a-\omega(k)}\right],\tag{B13}$$

where

$$\mathscr{D}(\omega_a)=\mathscr{D}^0(\omega_a)+\mathscr{D}^0(\omega_a)\Sigma(\omega_a)\mathscr{D}(\omega_a)\ .$$

Let[40]

$$\Sigma_R(x)+i\Sigma_I(x)\equiv\Sigma(\omega_a)\,|_{\,i\omega_a=x-i\eta}\ ,\tag{B14}$$

Then

$$\mathscr{D}(\omega_a)=\int_{-\infty}^{\infty}\frac{dx}{2\pi}\frac{\rho(x)}{i\omega_a-x}\ ,$$

where

$$\rho(x)=\frac{2\Sigma_I(x)}{[x-\omega_0-\Sigma_R(x)]^2+\Sigma_I^2(x)}\tag{B15}$$

and $\mathscr{D}(\omega_a)$ is the full finite-temperature Green's function for the two-level model. In (B15) expand $x$ about $x=\nu$ where $\nu-\omega_0-\Sigma_R(\nu)=0$. Assume that $(\partial\Sigma_I(\partial x)_{x=\nu}$ is small so that $\rho(x)$ can be approximated as a Lorentzian

$$\rho(x)\sim\frac{2\Sigma_I(\nu)}{(x-\nu)^2\left[1-\left.\frac{\partial\Sigma_R}{\partial x}\right|_\nu\right]^2+[\Sigma_I(\nu)]^2}\ .\tag{B16}$$

For such a Lorentzian, the excitation energy is

$$\nu=\omega_0+\Sigma_R(\nu)\approx\omega_0+\Sigma_R^{(2)}(\omega_0)$$

and the damping is

$$\left[1-\left.\frac{\partial\Sigma_R}{\partial x}\right|_\nu\right]^{-1}\Sigma_I(\nu)\sim\Sigma_I^{(2)}(\omega_0)\ .$$

The result (B13) therefore gives exactly the same level shift and width as derived in the text [Eqs. (14) and (20)].

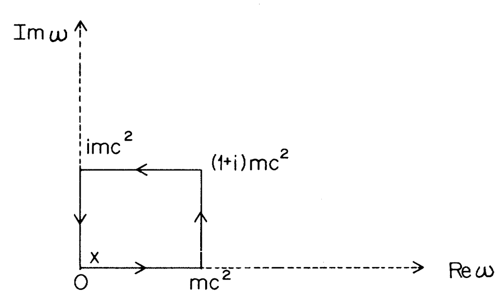


FIG. 6. Contour for the integral (86).

## APPENDIX C: EVALUATION OF INTEGRALS FOR THE RADIATED POWER

Consider first the integral of Eq. (86). This can be related to the integral around the square contour shown in Fig. 6. The $x$ indicates the pole at $\omega=\omega_0+i\Gamma$. Application of the residue theorem yields

$$\int_0^{mc^2} d\omega\,\omega^2\left[2i\Gamma e^{-\Gamma t}\mathscr{R}''_{1a}(-i\Gamma)\left[\frac{1}{(\omega_0-\omega)^2+\Gamma^2}-\frac{1}{(\omega_0+\omega)^2+\Gamma^2}\right]\right]$$

$$=2\pi i e^{-\Gamma t}\mathscr{R}''_{1a}(-i\Gamma)(\omega_0+i\Gamma)^2$$

$$-2i\Gamma e^{-\Gamma t}\mathscr{R}''_{1a}(-i\Gamma)\left[\int_{mc^2}^{0}(iy)^2\left[\frac{1}{(\omega_0-iy)^2+\Gamma^2}-\frac{1}{(\omega_0+iy)^2+\Gamma^2}\right]i\,dy\right.$$

$$+\int_0^{mc^2}(mc^2+iy)^2\left[\frac{1}{(mc^2-\omega_0+iy)^2+\Gamma^2}-\frac{1}{(mc^2+\omega_0+iy)^2+\Gamma^2}\right]i\,dy$$

$$\left.+\int_{mc^2}^{0}(x+imc^2)^2\left[\frac{1}{(x-\omega_0+imc^2)^2+\Gamma^2}-\frac{1}{(x+\omega_0+imc^2)^2+\Gamma^2}\right]dx\right]. \tag{C1}$$

We shall find later that $\Gamma$ is of order $\gamma^{WW}\ll\omega_0$. Then $\Gamma\ll|\omega_0\pm iy|$ and one may, in leading order, drop $\Gamma^2$ in all of the denominators on the right-hand side of (C1), yielding for the expression in large brackets

$$\int_0^{mc^2} dy\left[-\frac{4\omega_0 y^3}{(y^2+\omega_0^2)^2}+\frac{4i\omega_0(mc^2+iy)^3}{[(mc^2+iy)^2-\omega_0^2]^2}-\frac{4\omega_0(y+imc^2)^3}{[(y+imc^2)^2-\omega_0^2]^2}\right]. \tag{C2}$$

This integral is of order $\omega_0\ln(mc^2/\omega_0)$. Then the ratio of its contribution to (C2) to the leading (pole) contribution is of order $(\gamma^{WW}/\omega_0)\ln(mc^2/\omega_0)\ll 1$, assuming that $\gamma^{WW}\ll\omega_0$ and that the logarithm is not $\gg 1$. Here we have used the fact that $\Gamma$ is of order $\gamma^{WW}$. Under the same assumption one has $(\omega_0+i\Gamma)^2\approx\omega_0^2$. Then the integral (C1) reduces, in leading order, to the pole contribution (86).

Let us next evaluate the integrals in Eqs. (103) and (104). Consider first the one in Eq. (103):

$$\int_0^{2\omega_0}\frac{\omega^2(\omega_0-\omega)\sin[(\omega_0-\omega)t]d\omega}{(\omega_0-\omega)^2+[\Sigma''_{1a}(0)]^2}=\int_{-\omega_0}^{\omega_0}\frac{(\omega'+\omega_0)^2\omega'\sin(\omega't)}{(\omega')^2+(2\gamma^{WW})^2}d\omega'$$

$$=\frac{1}{2i}\int_{-\omega_0}^{\omega_0}\frac{(\omega'+\omega_0)^2\omega'(e^{i\omega't}-e^{-i\omega't})}{(\omega')^2+(2\gamma^{WW})^2}d\omega', \tag{C3}$$

use having been made of the relationship $\Sigma''_{1a}(0)=2\gamma^{WW}$, since we wish to eventually relate the decay rate of the radiated power to $\gamma^{WW}$, the spontaneous decay linewidth [see Eqs. (14) and (115)]. The integral (C3) will be evaluated as the difference of the two integrals $I_\pm$:

$$I_\pm=\frac{1}{2i}\int_{-\omega_0}^{\omega_0}\frac{(\omega'+\omega_0)^2\omega'e^{\pm i\omega't}}{(\omega')^2+(2\gamma^{WW})^2}d\omega'. \tag{C4}$$

In the case of $I_+$, close the contour in the upper half-plane, as shown in Fig. 7. The $x$'s indicate the poles at $\omega'=\pm 2i\gamma^{WW}$. The contour is closed at the top by the horizontal line at $\omega'=x+i\infty$ ($x$ real), which contributes nothing to the integral. By the residue theorem

$$2iI_+ + ie^{i\omega_0 t}\int_0^\infty\frac{(2\omega_0+iy)^2(\omega_0+iy)e^{-ty}}{(\omega_0+iy)^2+(2\gamma^{WW})^2}dy - ie^{-i\omega_0 t}\int_0^\infty\frac{(iy)^2(-\omega_0+iy)e^{-ty}}{(-\omega_0+iy)^2+(2\gamma^{WW})^2}dy=2\pi i\mathscr{R}(2i\gamma^{WW}), \tag{C5}$$

where the residue at $\omega'=2i\gamma^{WW}$ is

$$\mathscr{R}(2i\gamma^{WW})=\tfrac{1}{2}(\omega_0+2i\gamma^{WW})^2e^{-2\gamma^{WW}t}$$

$$\approx\tfrac{1}{2}\omega_0^2e^{-2\gamma^{WW}t} \tag{C6}$$

and we have assumed $\gamma^{\rm WW} \ll \omega_0$. For the same reason

$$2\gamma^{\rm WW} \ll |\omega_0 \pm iy| \ , \tag{C7}$$

so one may write for the sum of the two integrals along the vertical legs, apart from a negligible correction,

$$ie^{i\omega_0 t}\int_0^\infty \frac{(2\omega_0+iy)^2 e^{-ty}}{\omega_0+iy}dy - ie^{-i\omega_0 t}\int_0^\infty \frac{(iy)^2 e^{-ty}}{-\omega_0+iy}dy$$

$$= i\int_0^\infty \frac{[4\omega_0^3 e^{i\omega_0 t}+\omega_0(3e^{i\omega_0 t}-e^{-i\omega_0 t})y^2-2y^3\sin(\omega_0 t)]e^{-ty}}{y^2+\omega_0^2}dy$$

$$= i\omega_0^2[4e^{i\omega_0 t}f(\omega_0 t)-(3e^{i\omega_0 t}-e^{-i\omega_0 t})g'(\omega_0 t)-2\sin(\omega_0 t)g''(\omega_0 t)] \ . \tag{C8}$$

Here[21]

$$f(x)=\int_0^\infty \frac{e^{-xy}}{y^2+1}dy = \mathrm{Ci}(x)\sin x - [\mathrm{Si}(x)-\pi/2]\cos x \ ,$$
$$g(x)=\int_0^\infty \frac{ye^{-xy}}{y^2+1}dy = -\mathrm{Ci}(x)\cos x - [\mathrm{Si}(x)-\pi/2]\sin x \ , \tag{C9}$$

$g'$ and $g''$ are the first and second derivatives of $g$, and $x>0$. Then with (C5) and (C6)

$$I_+ \approx \tfrac{1}{2}\pi\omega_0^2 e^{-2\gamma^{\rm WW}t} - \tfrac{1}{2}\omega_0^2[4e^{i\omega_0 t}f(\omega_0 t)-(3e^{i\omega_0 t}-e^{-i\omega_0 t})g'(\omega_0 t)-2\sin(\omega_0 t)g''(\omega_0 t)] \ , \tag{C10}$$

the only approximation being those of Eqs. (C6) and (C7). A similar derivation, but with the contour closed in the lower half-plane, yields

$$I_- \approx -\tfrac{1}{2}\pi\omega_0^2 e^{-2\gamma^{\rm WW}t} + \tfrac{1}{2}\omega_0^2[4e^{-i\omega_0 t}f(\omega_0 t)-(3e^{-i\omega_0 t}-e^{i\omega_0 t})g'(\omega_0 t)-2\sin(\omega_0 t)g''(\omega_0 t)] \ . \tag{C11}$$

Then the integral (C3) is

$$\int_{-\omega_0}^{\omega_0} \frac{(\omega'+\omega_0)^2\omega'\sin(\omega' t)}{(\omega')^2+(2\gamma^{\rm WW})^2}d\omega' \approx \pi\omega_0^2 e^{-2\gamma^{\rm WW}t} - 4\omega_0^2\cos(\omega_0 t)f(\omega_0 t)$$
$$+2\omega_0^2\cos(\omega_0 t)g'(\omega_0 t)+2\omega_0^2\sin(\omega_0 t)g''(\omega_0 t)$$
$$=\pi\omega_0^2\{e^{-2\gamma^{\rm WW}t}+2\pi^{-1}[\mathrm{Si}(\omega_0 t)-\pi/2]$$
$$+2\pi^{-1}(\omega_0 t)^{-2}[\sin(\omega_0 t)-\omega_0 t\cos(\omega_0 t)]\} \ , \tag{C12}$$

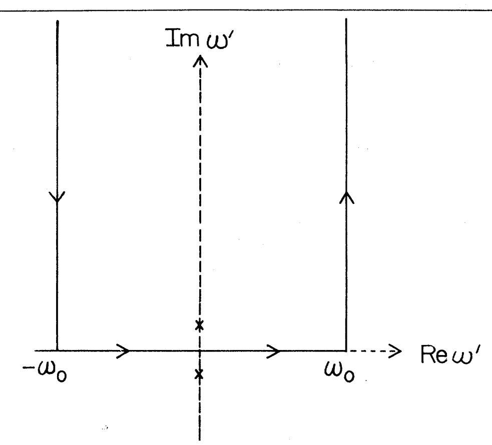


FIG. 7. Contour for the integral $I_+$.

the final expression being obtained with the aid of (C9) and the definitions[21] of the sine and cosine integrals.

The first integral in Eq. (104) can be evaluated in the same way. One has

$$\int_0^{2\omega_0} \frac{\omega^2\cos[(\omega_0-\omega)t]}{(\omega_0-\omega)^2+[\Sigma''_{1a}(0)]^2}d\omega = J_+ + J_- \ ,$$
$$J_\pm = \tfrac{1}{2}\int_{-\omega_0}^{\omega_0}\frac{(\omega'+\omega_0)^2 e^{\pm i\omega' t}}{(\omega')^2+(2\gamma^{\rm WW})^2}d\omega' \ . \tag{C13}$$

Proceeding as in the evaluation of $I_+$, one has

$$J_+ \approx \tfrac{1}{4}\pi\omega_0^2(\gamma^{\rm WW})^{-1}e^{-2\gamma^{\rm WW}t} - \tfrac{1}{2}ie^{i\omega_0 t}\int_0^\infty \frac{(2\omega_0+iy)^2e^{-ty}}{(\omega_0+iy)^2}dy + \tfrac{1}{2}ie^{-i\omega_0 t}\int_0^\infty \frac{(iy)^2e^{-ty}}{(\omega_0-iy)^2}dy \; . \qquad \text{(C14)}$$

The integrals in (C14) need not be evaluated, since they are of order $(\gamma^{\rm WW}/\omega_0) \ll 1$ relative to the pole contribution. The integral $J_-$ is the complex conjugate of $J_+$. Hence

$$J_+ + J_- \approx \tfrac{1}{2}\pi\omega_0^2(\gamma^{\rm WW})^{-1}e^{-2\gamma^{\rm WW}t} \; . \qquad \text{(C15)}$$


[1]M. D. Girardeau, Phys. Rev. A xx, xxx (19xx). Equation $(n)$ of that paper will be denoted herein by (I.$n$).

[2]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).

[3]P. R. Milonni, in *Foundations of Radiation Theory and Quantum Electrodynamics,* edited by A. O. Barut (Plenum, New York, 1980), p. 5, Eq. (7).

[4]The subscripts 1 have been dropped ($\Delta - i\gamma$ stands for $\Delta_1 - i\gamma_1$).

[5]Note that, in general, $c(n\mid 1)$ is not the complex conjugate of the corresponding $c(1\mid n)$ [compare (7) and (9)], i.e., the $c$ matrix is not Hermitian. This is because the operator basis $\{\hat{B}_m\}$ is not, in general, orthonormal. In fact, we have not required any metric in Liouville space, although one could, if desired, use the standard trace metric.

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

[7]J. W. Farley and W. H. Wing, Phys. Rev. A 23, 2397 (1981).

[8]In this case $\hat{\rho}$ is a pure-state projector $|\psi\rangle\langle\psi|$.

[9]In fact, the factorization need only be valid to zero order in the interaction, as is the case in (19).

[10]This generalizes the thermal-equilibrium case, where $\epsilon = k_B T$.

[11]M. D. Girardeau, in *Lecture Notes in Physics, Vol. 142, Recent Progress in Many-Body Theories: Proceedings, Oaxtepec, Mexico, 1981,* edited by J. G. Zabolitzky, M. de Llano, M. Fortes, and J. W. Clark (Springer, Berlin, Heidelberg, New York, 1981), pp. 355ff., particularly pp. 358–360.

[12]The factor 2 is the prefactor of $\hat{a}^\dagger\hat{a}$ in Eq. (6).

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

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

[15]A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, N. J., 1963), Chap. 6.

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

[17]The matter-field interaction is proportional to $\hat{\vec{J}}\cdot\hat{\vec{A}}$. The $\hat{B}_n$ occurring in (44) arise from the commutator of $\hat{N}_{\vec{k}\lambda}$ with this interaction. The commutator of $\hat{N}_{\vec{k}\lambda}$ with $\hat{\vec{A}}$ gives terms proportional to $\hat{b}_{\vec{k}\lambda}$ and $\hat{b}^\dagger_{\vec{k}\lambda}$, and hence $\hat{B}_n$ involving $\hat{\vec{J}}\,\hat{b}_{\vec{k}\lambda}$ and $\hat{b}^\dagger_{\vec{k}\lambda}\hat{\vec{J}}$. The $c(n\mid m)$ in (59) are then the coefficients occurring in the various nonzero terms in the commutator of the $\hat{\vec{J}}\,\hat{b}_{\vec{k}\lambda}$ and $\hat{b}^\dagger_{\vec{k}\lambda}\hat{\vec{J}}$ with $\hat{\vec{J}}\cdot\hat{\vec{A}}$. Retaining only the commutators of $\hat{b}_{\vec{k}\lambda}$ and $\hat{b}^\dagger_{\vec{k}\lambda}$ with $\hat{\vec{A}}$ (which are $c$ numbers), one obtains terms bilinear in the current, as implied in the text.

[18]Usually called the "Lorentzian" line shape; see A. A. Michelson, Astrophys. J. 11, 251 (1895) and H. A. Lorentz, Proc. R. Acad (Amsterdam) 8, 591 (1906).

[19]At least, this is how spectra are analyzed in the microwave domain. In the optical domain, filters can be thought of as operating in the same way, since they involve dye molecules with various resonant absorption peaks. Optical spectrographs do not operate in this way, but it is sufficient for our argument here that the spectra *could,* in principle, be analyzed by filters.

[20]This approximation could break down in the case that the ensemble is a highly nonequilibrium one (e.g., one representing the radiation field of a laser) or the temperature is very high. In such cases the contributions of the other terms in (72) should be investigated. However, we shall drop them here for simplicity.

[21]For properties of the sine integral Si($x$) and cosine integral Ci($x$) see, e.g., *Handbook of Mathematical Functions,* Natl. Bur. Stand. Appl. Math. Ser. No. 55, edited by M. Abramowitz and I. A. Stegun (U.S. GPO, Washington, D.C., 1964), pp. 231–233.

[22]Note that $\mathscr{L}_0\hat{N} = [\hat{N}, \hat{H}_0] = 0$ since $\hat{H}_0$ of Eq. (1) is diagonal in the occupation-number representation.

[23]Here $\epsilon_m$ is the $\mathscr{L}_0$ eigenvalue of $\hat{N}$, which implies $\epsilon_m = 0$.

[24]This contribution arises from the terms $\hat{B}_m = \hat{N}$ (111), which give a factor $\langle\hat{a}\,\hat{B}_m\hat{a}^\dagger\rangle/\langle\hat{a}\,\hat{N}\,\hat{a}^\dagger\rangle = 1$.

[25]This also includes the vacuum field fluctuation contributions, which cause $f_{\vec{k}\lambda} = \langle\hat{N}_{\vec{k}\lambda}\rangle$ to be nonzero even at zero temperature (interacting ground state of $\hat{H}$). These contributions to $f_{\vec{k}\lambda}$ are of second order (and higher) in the radiative interaction.

[26]Each $\delta$ function from (78) contributes only if the zero of its argument falls in the interval $[0, mc^2]$ of integration. Also, each principal-part term has a singularity only under the same condition. This is the origin of the $\vartheta$ functions in (98).

[27]The dominant terms for very small $t$ are the trigonometric terms with argument $(mc^2 - \omega_0)t \approx mc^2 t$ in (108), noting that $\sin x - x\cos x = x^3/3 + \cdots$.

[28]Although $\omega_0$ has been neglected compared with $mc^2$ in the denominator of (112), it is retained in the argument of the cosine since it gives a non-negligible phase shift.

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

[30]Recall that $\langle \hat{N} \rangle$ arises here only from vacuum field fluctuations, and is of order $\alpha \ll 1$ (fine-structure constant).

[31]The classical dipole moment, and hence its second derivative $\ddot{\vec{d}}$, vary like $\sin(\omega_0 t)e^{-\gamma^{WW}t}$. The instantaneous *classical* radiated power is proportional to $\ddot{\vec{d}}^2$, but this is not true quantum mechanically.

[32]J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D 10, 3350 (1974).

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

[34]This is required only in the macroscopic limit $\Omega \to \infty$. as in Sec. II.

[35]D. L. Huber and J. H. Van Vleck, Rev. Mod. Phys. 38, 187 (1966), particularly Eqs. (II.24)—(II.29) and (II.43)—(II.45).

[36]*Handbook of Mathematical Functions,* Natl. Bur. Stand. Appl. Math. Series No. 55, edited by M. Abramowitz and I. A. Stegun (U.S. GPO, Washington, D.C., 1964), pp. 258 and 259, Eq. (6.3.21).

[37]*Tables of Integral Transforms,* edited by A. Erdelyi *et al.* (McGraw-Hill, New York, 1954), Vol. 2.

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

[39]See p. 65 of Ref. 38.

[40]Reference 38, problem (9.3), p. 309.