Nonperturbative decay of an atomic system in a cavity

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Atoms can nowadays be placed in increasingly exotic environments such as microscopic cavities and materials with photonic band gaps. High-Q cavities can now easily result in a strong coupling between an atom and its environment where perturbation theory should no longer be appropriate. The purpose of this paper is to describe the dynamics of a multilevel V-type atomic system (including the case of a two-level system) which interacts with a reservoir modeled by a generalized density of states. A theoretical construct, the pseudomode, is utilized to develop general methods for solution. Without using perturbation theory the equivalent master equation is developed and the relationship between the master equation, the pseudomodes, and the generalized density of states function is explored with examples. Utilizing a straightforward definition of the pseudomode, it is found that many functions for the density of states lead to problematic non-Lindblad master equations. Several examples are given, and it is shown how to convert the non-Lindblad master equations into a Lindblad form in these cases. The examples include a non-Lorentzian resonance and a simple model of a photonic band gap. [S1050-2947(97)04102-4]

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I. INTRODUCTION

The decay of excited atomic systems has been of interest since the time of the pioneering work of Weisskopf and Wigner [1]. It has long been known that the decay of an atom is not an intrinsic property of that atom, but depends strongly on the nature of the environment of that atom [2]. Recently, this has been of much interest (see, for example, Refs. [3–16]) in part because of the development of microlasers (see, for example, [17–19]). The nonperturbative features of the interaction between an atom and its environment are especially apparent if there is such a strong coupling that energy leaving the atom can later return [20,21]. Changing the environment often means placing the atom in a cavity of some kind, but nowadays the atom may instead be an exciton, and the environment that of a quantum well. For brevity we will refer to a cavity throughout most of this paper.

The dynamics of the atom-environment interactions have long been described by master equations derived from perturbation theory [22,23], which is an approach that works well in two regimes: it works in the low-Q cavity, where the cavity field can be eliminated in favor of atomic dynamics, and it works in the high-Q regime where we consider the atomic system coupled to a damped cavity mode.

A central purpose of this paper is to provide a general description of the atom-environment interaction which does not rely on perturbation theory at all. Furthermore, one of the tasks is to compose the appropriate master equation to describe the decay of the atomic system for a given type of environment. In achieving this a particular difficulty is exposed—that a direct approach generates pathological master equations which do not have an acceptable physical interpretation. However, examples are given in this paper of a procedure to correct this problem and return to an acceptable form for the master equation. The acceptable form is the Lindblad form [24]

$$\frac{d}{dt}\hat{\rho}=-i[H_0,\hat{\rho}]-\sum_l\left(\frac{1}{2}\hat{L}_l^\dagger\hat{L}_l\hat{\rho}-\hat{L}_l\hat{\rho}\hat{L}_l^\dagger+\frac{1}{2}\hat{\rho}\hat{L}_l^\dagger\hat{L}_l\right), \qquad (1)$$

where the Lindblad operators are the $\hat{L}_l$, H_0 is the system Hamiltonian, and $\hat{\rho}$ is the density matrix. This form of master equation arises very naturally from time-dependent perturbation theory applied to the interaction of the system with a zero-temperature heat bath. The Lindblad terms then originate from the double commutator of the system-bath interaction evaluated to second order [22,23]. However, when the coupling of the system to the environment is very strong, perturbation theory cannot be expected to yield the correct result. In this paper we will see that for many decaying cavity-atom systems a form of the Lindblad master equation (1) is still appropriate.

In Sec. II we will formulate the problem. Then in Sec. III we will see how the time evolution can be solved in terms of a finite set of coupled differential equations, and how those equations can be formulated in terms of a nonperturbative master equation. Some straightforward examples are then given in Sec. IV. In Sec. V we identify some problems with some of the master equations that would be generated by the methods of Sec. III. A procedure for correcting the defect is shown and then the more general master equation is found in Sec. VI. Examples of this type of master equation are then given in Sec. VII. The Laplace transform method of solution is summarized in Sec. VIII. Some concluding remarks are then provided in Sec. IX.

II. MATHEMATICAL DESCRIPTION

In this paper we consider multilevel V-type atomic systems, including systems with only two levels, as illustrated in Fig. 1. Thus the atomic system comprises a single ground-state level, labeled 0, coupled to a number of excited states which have the labels i (or j) for i $(j)=1,2,3,\ldots$ and an energy difference (measured from the ground state) of ω_i. The labels i and j will be used for only the atomic system. The number of excited levels is not specified, but may be one or more. The multilevel system is coupled to a bath of oscillators and the creation and annihilation operators for each

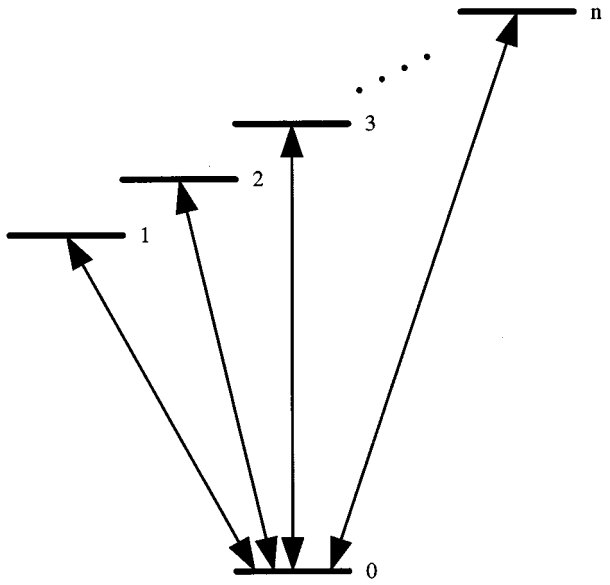

FIG. 1. The type of multilevel atomic system considered in this paper. The ground state 0 is coupled to the upper states 1,2,3, . . . by transitions with frequencies $\omega_1,\omega_2,\omega_3,\ldots$. The examples in the text are given for a two-level system, when only levels 0 and 1 are present.

oscillator are $a_\lambda^\dagger$ and a_λ, where the oscillator, which has frequency ω_λ, is labeled here by the index λ. Then, within the rotating wave approximation and with only dipole interactions, the Hamiltonian for the system can be written as

$$H=\sum_\lambda \omega_\lambda a_\lambda^\dagger a_\lambda+\sum_i \omega_i|i\rangle_{aa}\langle i| + \sum_{i,\lambda} g_\lambda^{(i)}(a_\lambda^\dagger|0\rangle_{aa}\langle i|+a_\lambda|i\rangle_{aa}\langle 0|), \tag{2}$$

where $g_\lambda^{(i)}$ is the frequency-dependent coupling between the transition $i-0$ and the mode labeled λ. For specific cavity-atom geometries, these couplings will be taken to include all the necessary spatial factors. For convenience in later parts of this paper, the basis for the states is chosen so that the couplings $g_\lambda^{(i)}$ are real.

The sum over modes λ is taken to include polarizations, and in the limit of a continuous distribution this sum can be converted into an integral by including the density of states ρ_λ,

$$\sum_\lambda \rightarrow \int d\omega_\lambda \rho_\lambda . \tag{3}$$

For the development in the next sections of this paper we will not have to assume any specific form for the density of states function, but ultimately the form plays an important role. To reflect this, we can extract the shape of the density of states function into a normalized density of states $D(\omega_\lambda)$ such that

$$\rho_\lambda (g_\lambda^{(i)})^2=\frac{\Omega_i^2}{2\pi}D(\omega_\lambda) \tag{4}$$

so each transition couples to the same density of states function, but with different strengths Ω_i. The function $D(\omega_\lambda)$ is then a generalized density of states which contains the frequency-dependent elements of both the density of states and the couplings. In the continuous limit, and with $\omega_\lambda \rightarrow \omega$, the normalization of $D(\omega)$ is given by

$$\int_{-\infty}^{\infty} d\omega\, D(\omega)=2\pi, \tag{5}$$

which is a normalization in terms of natural, rather than angular, frequency. This choice is simply for convenience. The extension of the integral to $-\infty$ is very useful to the physical interpretation, though not essential. The main requirement is that the integral over the density of states and the frequency-dependent couplings should be a good approximation in the region of interest. This is true for optical cavities, though we note that for other physical systems it may not be true and we can then obtain ''threshold'' effects [25]. The strengths Ω_i are defined from the weight of the density of states,

$$\Omega_i^2=\sum_\lambda (g_\lambda^{(i)})^2, \tag{6}$$

and we will define a total strength for all the transitions as

$$\Omega_0^2=\sum_i \Omega_i^2. \tag{7}$$

We may split the Hamiltonian Eq. (2) into two pieces comprising the interacting part and the noninteracting (bare) part so that

$$H=H_B+H_I,$$

$$H_B=\sum_\lambda \omega_\lambda a_\lambda^\dagger a_\lambda+\sum_i \omega_i|i\rangle_{aa}\langle i|,$$

$$H_I=\sum_{\lambda,i} g_\lambda^{(i)}(a_\lambda^\dagger|0\rangle_{aa}\langle i|+a_\lambda|i\rangle_{aa}\langle 0|). \tag{8}$$

The interaction Hamiltonian H_I will only connect certain combinations of atomic states and field states. For a single excitation of the total system these states are

$$\psi_i=|i\rangle_a\otimes|000\ldots\ldots000\rangle,$$

$$\psi_\lambda=|0\rangle_a\otimes|000\ldots\ldots010\ldots\ldots000\rangle, \tag{9}$$

where the ket $|000\ldots\ldots000\rangle$ indicates the field state where all the radiation modes are in a vacuum state, and the ket $|000\ldots\ldots010\ldots\ldots000\rangle$ in Eq. (9) indicates a state of the radiation field where all of the modes are in a vacuum state apart from mode λ which is in the first excited state. The unexcited state

$$\psi_0=|0\rangle_a\otimes|000\ldots\ldots000\rangle \tag{10}$$

is not coupled to any other state.

Now for the noninteracting part of the Hamiltonian we will trivially obtain

$$H_B\psi_i=\omega_i\psi_i\,,\quad H_B\psi_\lambda=\omega_\lambda\psi_\lambda\,, \tag{11}$$

while for the interacting part of the Hamiltonian

$$H_I\psi_i=\sum_\lambda\, g_\lambda^{(i)}\psi_\lambda\,,\quad H_I\psi_\lambda=\sum_i\, g_\lambda^{(i)}\psi_i. \tag{12}$$

It is clear from these equations that we have a closed system of equations for the time evolution. We will now expand a general state vector of the system as

$$\Psi(t)=c_0\psi_0+\sum_i\, c_i(t)\psi_i+\sum_\lambda\, c_\lambda(t)\psi_\lambda \tag{13}$$

in terms of the states (9) and insert this into the Schrödinger equation $i(d/dt)\Psi=H\Psi$ to obtain the following (infinite) set of coupled equations:

$$i\frac{d}{dt}c_i=\omega_i c_i+\sum_\lambda\, g_\lambda^{(i)}c_\lambda\,,\quad i\frac{d}{dt}c_\lambda=\omega_\lambda c_\lambda+\sum_i\, g_\lambda^{(i)}c_i\,. \tag{14}$$

The coefficient c_0 is constant in time. It is convenient to move to an interaction representation by means of the following time-dependent transformations:

$$\tilde{c}_i(t)=e^{i\omega_i t}c_i(t),\quad \tilde{c}_\lambda(t)=e^{i\omega_\lambda t}c_\lambda(t), \tag{15}$$

so that we obtain the following coupled equations:

$$i\frac{d}{dt}\tilde{c}_i=\sum_\lambda\, g_\lambda^{(i)}e^{-i\Delta_\lambda^i t}\tilde{c}_\lambda\,, \tag{16}$$

$$i\frac{d}{dt}\tilde{c}_\lambda=\sum_i\, g_\lambda^{(i)}e^{i\Delta_\lambda^i t}\tilde{c}_i\,, \tag{17}$$

with the detunings from the mode λ defined by

$$\Delta_\lambda^i=\omega_\lambda-\omega_i\,. \tag{18}$$

Now we can eliminate the coefficients $\tilde{c}_\lambda$ by integrating Eq. (17) (in time) and substituting the resulting expression for $\tilde{c}_\lambda$ into Eq. (16). The integration of Eq. (17) yields

$$\tilde{c}_\lambda(t)=-i\int_0^t dt'\sum_i\, g_\lambda^{(i)}e^{i\Delta_\lambda^i t'}\tilde{c}_i(t'), \tag{19}$$

where the initial condition assumed is

$$\tilde{c}_\lambda(0)=c_\lambda(0)=0, \tag{20}$$

which simply means that there are no photons in the external bath (or cavity). We thus obtain a finite set of coupled integro-differential equations

$$\frac{d}{dt}\tilde{c}_i(t)=-\int_0^t dt'\sum_j\, \tilde{G}_{ij}(t,t')\tilde{c}_j(t'), \tag{21}$$

where the functions $\tilde{G}_{ij}(t,t')$ are defined by

$$\tilde{G}_{ij}(t,t')=\sum_\lambda\, g_\lambda^{(i)}g_\lambda^{(j)}\exp[i(\Delta_\lambda^j t'-\Delta_\lambda^i t)] \tag{22}$$

for a pair of transitions i,j. In writing down Eq. (21) we have exchanged the order of summation over λ and integration over time. This then allows us to write down the expressions (22) which can be evaluated analytically for a specific expression of the coupling given in Eq. (4).

If we transfer the equations (21) back into the original basis we obtain

$$\frac{d}{dt}c_i(t)=-i\omega_i c_i(t)-\int_0^t dt'\sum_j\, G_{ij}(t-t')c_j(t'), \tag{23}$$

where the difference kernel $G_{ij}(t-t')$ is

$$\begin{aligned}G_{ij}(t-t')&=\exp(-i\omega_i t+i\omega_j t')\tilde{G}_{ij}(t,t')\\&=\sum_\lambda\, g_\lambda^{(i)}g_\lambda^{(j)}\exp[-i\omega_\lambda(t-t')].\end{aligned} \tag{24}$$

Then when the sum over λ becomes a continuous integral we obtain the kernels as integrals, which can be regarded as Fourier transforms of $D(\omega)$,

$$G_{ij}(t-t')=\frac{\Omega_i\Omega_j}{2\pi}\int_{-\infty}^{\infty}d\omega D(\omega)e^{-i\omega(t-t')} \tag{25}$$

$$=\frac{\Omega_i\Omega_j}{\sqrt{2\pi}}\overline{D(t-t')}. \tag{26}$$

In the following sections we will examine some of the ways we can solve the integrodifferential equations (23) for different forms of $D(\omega)$.

III. METHOD OF POLES

A. Pseudomodes

It proves very useful to be able to calculate the integral (25) from a contour in the complex ω plane. In this case the function $D(\omega)$ should be analytic and the poles of that function will correspond to resonances. The contour is closed in the lower half plane, where the exponential part of Eq. (25) causes the integrand to vanish [because $t\geqslant t'$ in Eq. (21)]. There are functions $D(\omega)$ which cannot have contours closed in the lower half plane, and then other methods have to be used. An example of an alternative approach is given in Sec. VIII.

So we take a contour in the lower half plane and we will assume that the contribution to the contour integral from the semicircle is negligible. Then we will have

$$G_{ij}(t-t')=-\frac{\Omega_i\Omega_j}{2\pi}\oint_C dz D(z)e^{-iz(t-t')}. \tag{27}$$

Now we will suppose that the function $D(z)$ has poles in the lower half plane at $z=z_1,z_2,\ldots,z_l,\ldots$. (The index l will be reserved for the positions of poles hereafter.) And we will denote the residues of $D(z)$ at these poles by r_l. Then by the theorem of residues,

$$G_{ij}(t-t')=-i\Omega_i\Omega_j\sum_l r_l e^{-iz_l(t-t')}. \tag{28}$$

We note that this is now the case of a separable kernel and we have excluded any possibility of interfering branch cuts in the lower half plane. However, we do not need to have only simple poles. With this result inserted into Eq. (23) we will find that

$$i\frac{d}{dt}c_i(t)=\omega_i c_i(t)-\sum_l \Omega_i r_l\sum_j \Omega_j e^{-iz_l t}\int_0^t dt' e^{iz_l t'}c_j(t'). \tag{29}$$

On the basis of Eq. (29) we can now define a fictional *pseudomode* amplitude as [16]

$$b_l(t)=-i\sum_i \Omega_i\sqrt{-ir_l}e^{-iz_l t}\int_0^t dt' e^{iz_l t'}c_i(t'). \tag{30}$$

This pseudomode amplitude is chosen to be associated with the pole at z_l so that Eqs. (23) become

$$i\frac{d}{dt}c_i(t)=\omega_i c_i(t)+\sum_l g_{il}b_l(t), \tag{31}$$

$$i\frac{d}{dt}b_l(t)=z_l b_l(t)+\sum_i g_{il}c_i(t), \tag{32}$$

where Eq. (32) follows from the differentiation of Eq. (30). Note that we now have a *finite* set of coupled differential equations instead of the infinite set found in Eq. (14). The coupling between the pseudomode l and the atomic level i is

$$g_{il}=\Omega_i\sqrt{-ir_l}, \tag{33}$$

which is, in general, a complex quantity. Note that g_{il} appears in both Eq. (31) and Eq. (32) rather than g_{il} and its complex conjugate. The ramifications of this will be discussed further in Sec. V. To proceed we will assume here that the couplings g_{il} are real. Indeed, we note that this is the case for Lorentzian resonances. In the general case the residues r_l all sum to i and have no *net* real part. This is because of the normalization (5) which means that $G_{ij}(0)=\Omega_i\Omega_j$, and thus from Eq. (28) we always have

$$\sum_l (-ir_l)=1. \tag{34}$$

The case of a two-level system interacting with a simple Lorentzian resonance is considered further in Sec. IV A.

If we now review the problem in hand we note that we have converted the original problem, which consisted of an infinite set of ordinary differential equations [Eq. (14)], into a finite set of ordinary differential equations, Eqs. (31) and (32). These equations can now be solved by well known analytic, or numerical methods. The restriction on $D(\omega)$ is that it is analytic in the lower half plane, and contains only poles there. The number of coupled differential equations is the number of poles added to the number of upper atomic levels. If there are an infinite number of poles, then we do again obtain an infinite number of coupled differential equations, though with discrete frequencies corresponding to the positions of the poles.

B. Equivalent master equation

We have seen that if we know the positions and residues of the poles of $D(z)$ we can compute the time evolution of the atomic system from the straightforward equations (31) and (32). However, it is possible to gain insight into the atom-field dynamics by examining the equations of motion of a specially constructed density matrix. This takes the form of a master equation. The equations are more complex, but they provide more convincing evidence that the amplitudes b_l are connected to the amplitude of a mode.

We start by constructing a state vector and a basis for a system comprising the pseudomode and the atom. The (unnormalized) state vector is

$$\begin{aligned}|\tilde{\psi}(t)\rangle=&c_0|0\rangle_a\prod_l|0\rangle_l+c_1(t)|1\rangle_a\prod_l|0\rangle_l\\&+c_2(t)|2\rangle_a\prod_l|0\rangle_l+\cdots+b_1(t)|0\rangle_a|1\rangle_1\prod_{l\neq1}|0\rangle_l\\&+b_2(t)|0\rangle_a|1\rangle_2\prod_{l\neq2}|0\rangle_l+b_3(t)|0\rangle_a|1\rangle_3\prod_{l\neq3}|0\rangle_l\\&+\cdots,\end{aligned} \tag{35}$$

where $|i\rangle_a$ are the basis states of the atomic system [as in Eq. (2)] and $|n_l\rangle_l$ (where $n_l=0,1$) are the newly introduced states for the pseudomode l. The fact that the state vector $|\tilde{\psi}(t)\rangle$ is not normalized is emphasized by the tilde. We will refer to the set of states

$$\begin{aligned}&|0\rangle_a|0\rangle_1|0\rangle_2|0\rangle_3\ldots,\\&|1\rangle_a|0\rangle_1|0\rangle_2|0\rangle_3\ldots,\\&|2\rangle_a|0\rangle_1|0\rangle_2|0\rangle_3\ldots,\\&\vdots\\&|0\rangle_a|1\rangle_1|0\rangle_2|0\rangle_3\ldots,\\&|0\rangle_a|0\rangle_1|1\rangle_2|0\rangle_3\ldots,\\&|0\rangle_a|0\rangle_1|0\rangle_2|1\rangle_3\ldots\\&.\\&.\end{aligned} \tag{36}$$

[used in Eq. (35) above] as the *pseudomode basis*. We should stress that this basis is a mathematical construction and, strictly, does not exist physically. The lowest energy, or vacuum, state will be denoted by the symbol $|\mathbf{0}\rangle$ so that

$$|\mathbf{0}\rangle=|0\rangle_a|0\rangle_1|0\rangle_2|0\rangle_3\ldots. \tag{37}$$

By using the basis (36) we can define an effective non-Hermitian evolution operator which replicates the equations (31) and (32) with the unnormalized state vector (35):

$$H_{\rm eff}=\sum_l z_l\hat{a}_l^\dagger\hat{a}_l+\sum_i \omega_i|i\rangle_{aa}\langle i|+\sum_{il} g_{il}(\hat{a}_l^\dagger\hat{\sigma}_-^{(i)}+\hat{a}_l\hat{\sigma}_+^{(i)}), \tag{38}$$

where the atomic raising and lowering operators between levels 0 and i are $\hat{\sigma}_+^{(i)}=|i\rangle_{a\,a}\langle 0|$ and $\hat{\sigma}_-^{(i)}=|0\rangle_{a\,a}\langle i|$. We will also have $(\hat{\sigma}_z^{(i)}+1)/2=|i\rangle_{a\,a}\langle i|$. The operators $\hat{a}_l$ and $\hat{a}_l^\dagger$ are the annihilation and creation operators for excitations of the fictional mode l. For example, $\hat{a}_l|1\rangle_l=|0\rangle_l$, and $[\hat{a}_l^\dagger,\hat{a}_m]=\delta_{lm}$. Then $|\tilde{\psi}(t)\rangle$ satisfies the equation

$$\frac{d}{dt}|\tilde{\psi}(t)\rangle=-iH_{\rm eff}|\tilde{\psi}(t)\rangle. \tag{39}$$

The effective Hamiltonian $H_{\rm eff}$ can be split into a Hermitian part and an anti-Hermitian part in the form

$$H_{\rm eff}=H_0-\frac{i}{2}\sum_l \hat{L}_l^\dagger\hat{L}_l, \tag{40}$$

where we have the Hermitian Hamiltonian

$$H_0=\sum_l \mathrm{Re}(z_l)\hat{a}_l^\dagger\hat{a}_l+\sum_i \omega_i|i\rangle_{aa}\langle i| + \sum_{il} g_{il}(\hat{a}_l^\dagger\hat{\sigma}_-^{(i)}+\hat{a}_l\hat{\sigma}_+^{(i)}), \tag{41}$$

and Lindblad operators involving the pseudomode,

$$\hat{L}_l=\sqrt{-2\,\mathrm{Im}(z_l)}\hat{a}_l. \tag{42}$$

The Lindblad (42) requires that $\mathrm{Im}(z_l)<0$, i.e., we closed the contour (27) in the lower half plane. We see now that because we consider the poles of $D(z)$ in the lower half complex plane the Lindblads $\hat{L}_l$ have real coefficients. The sign of the energy of the pseudomode l depends on whether the pole at z_l is to the right or left of the imaginary axis. The particular form of splitting in Eqs. (40)–(42) depends on the coefficients g_{il} being real. The complications of complex g_{il} are considered in Secs. V and VI.

Thus we may determine the dynamics of the upper atomic energy levels from Eqs. (31) and (32) or the equivalent equation (39). However, to obtain the dynamics of the atomic ground-state population we need more than just these differential equations because there are contributions from both the initial ground-state population and from each excited field state (which would have the atomic system in the ground state). Thus there cannot be a description in terms of a pure state. If we start from the fundamental equation (13) and denote the atomic ground-state population by Π_0, we have

$$\Pi_0=|c_0|^2+\sum_\lambda |c_\lambda|^2 \tag{43}$$

in terms of the original modes of the system. Now we obtain a differential equation for the population by differentiating and using Eq. (17) so that

$$\frac{d}{dt}\Pi_0=-\sum_i \frac{d}{dt}|c_i|^2, \tag{44}$$

which simply expresses conservation of probability. But now we use the pseudomode equations (31) to find that

$$\frac{d}{dt}\Pi_0=\sum_{il}(ic_i^*g_{il}b_l-ic_ig_{il}^*b_l^*) \tag{45}$$

and we compare this to the total population growth of the pseudomodes from Eq. (32),

$$\sum_l \frac{d}{dt}|b_l|^2=2\sum_l \mathrm{Im}(z_l)|b_l|^2+\sum_{il}(ic_i^*g_{il}^*b_l-ic_ig_{il}b_l^*) \tag{46}$$

so that (in the case that g_{il} is real)

$$\frac{d}{dt}\Pi_0=\sum_l \frac{d}{dt}|b_l|^2-2\sum_l \mathrm{Im}(z_l)|b_l|^2 =\sum_l \frac{d}{dt}|b_l|^2+\sum_l \langle\tilde{\psi}|\hat{L}_l^\dagger\hat{L}_l|\tilde{\psi}\rangle, \tag{47}$$

where the last line follows from Eqs. (35) and (42). Clearly, in the pseudomode system, the rise in Π_0 is not given by just the increase in the pseudomode population, because the pseudomodes are *lossy* and lose population.

We will now proceed to a density matrix description of the system by combining the results of both Eqs. (39) and (47) in the spirit of master equation unraveling [26] (though the process performed here is master equation composition). We know that all the atomic coherences and populations (with the exception of the ground-state population) are described by

$$\hat{\rho}_{nj}=|\tilde{\psi}\rangle\langle\tilde{\psi}| \tag{48}$$

from Eq. (35). And from Eq. (39) we know that $\hat{\rho}_{nj}$ obeys the differential equation

$$\frac{d}{dt}\hat{\rho}_{nj}=-i[H_{\rm eff},\hat{\rho}_{nj}]. \tag{49}$$

Then to complete the density matrix we must include an additional term to account for the behavior of the ground-state population which is described by Eq. (47). Thus the additional term belongs to the pseudomode vacuum state and is of the form

$$\hat{\rho}_j=\Pi_j(t)|\mathbf{0}\rangle\langle\mathbf{0}|. \tag{50}$$

Then the total population of the pseudomode vacuum comprises two pieces: the new component from $\hat{\rho}_j$, and a contribution from $\hat{\rho}_{nj}$, the time-independent $|c_0|^2$. Thus

$$\Pi_{\rm vac}(t)=\Pi_j(t)+|c_0|^2, \tag{51}$$

where $\Pi_{\rm vac}(t)$ is simply the population of the pseudomode vacuum. Now by analogy with Eq. (43) (for the original system), the atomic ground-state population will be given by the sum of population of the pseudomode vacuum and the populations of all of the pseudomodes so that

$$\Pi_0=\Pi_{\rm vac}+\sum_l |b_l|^2. \tag{52}$$

Then by considering Eq. (47) and Eq. (51) we have

$$\frac{d}{dt}\Pi_j=\sum_l \langle\widetilde{\psi}|\hat{L}_l^\dagger\hat{L}_l|\widetilde{\psi}\rangle=\sum_l \langle\mathbf{0}|\hat{L}_l|\widetilde{\psi}\rangle\langle\widetilde{\psi}|\hat{L}_l^\dagger|\mathbf{0}\rangle, \quad (53)$$

where the last line follows because the action of any of the Lindblads [Eq. (42)] is to project the pseudomode system [Eq. (35)] onto the vacuum. Then by utilizing this result in Eq. (50) we can write a differential equation for $\hat{\rho}_j$ in the form

$$\frac{d}{dt}\hat{\rho}_j=\sum_l \hat{L}_l\hat{\rho}_{nj}\hat{L}_l^\dagger. \quad (54)$$

Now, we note that

$$\hat{L}_l\hat{\rho}_j\hat{L}_l^\dagger=0 \quad (55)$$

and that

$$[H_{\rm eff},\hat{\rho}_j]=0 \quad (56)$$

so that we can easily combine our equations for $\hat{\rho}_j$ and $\hat{\rho}_{nj}$ into an equation for the complete density matrix,

$$\frac{d}{dt}\hat{\rho}=-i[H_{\rm eff},\hat{\rho}]+\sum_l \hat{L}_l\hat{\rho}\hat{L}_l^\dagger, \quad (57)$$

where

$$\hat{\rho}=\hat{\rho}_{nj}+\hat{\rho}_j \quad (58)$$

forms the complete composed density matrix. If we substitute Eq. (40) for $H_{\rm eff}$ we obtain a master equation which is in the exact form of Eq. (1) and thus the process of constructing a master equation for the system is complete.

The solution of the master equation is given by Eq. (58) and is clearly in the form of a statistical mixture of the vacuum state and the state vector $\widetilde{\psi}$,

$$\hat{\rho}(t)=\Pi_j(t)|\mathbf{0}\rangle\langle\mathbf{0}|+|\widetilde{\psi}(t)\rangle\langle\widetilde{\psi}(t)|. \quad (59)$$

We can then use Eqs. (47), (51), and (52), and the initial conditions for the b_l and Π_j (all zero) to determine $\Pi_j(t)$ from the integral,

$$\Pi_j(t)=-2\sum_l \mathrm{Im}(z_l)\int_0^t|b_l(t')|^2dt'. \quad (60)$$

IV. APPLICATIONS TO SIMPLE SYSTEMS

A. Master equation for a single pseudomode

As a simple example of a master equation generated by the nonperturbative behavior of the atom-field coupling we consider a two-level atom, and let it interact with a density of states function $D(\omega)$ which has a single pole in the lower half plane which is located at

$$z_1\equiv\omega_c-i\Gamma/2. \quad (61)$$

Because there is only one pole it follows from the normalization property (34) that the single pseudomode coupling is simply the real quantity

$$g_{11}=\Omega_0. \quad (62)$$

Now to proceed to a master equation we simply use Eqs. (41), (42), and (1) to find that

$$\frac{d}{dt}\hat{\rho}=-i[H_0,\hat{\rho}]-\frac{\Gamma}{2}(\hat{a}^\dagger\hat{a}\hat{\rho}-2\hat{a}\hat{\rho}\hat{a}^\dagger+\hat{\rho}\hat{a}^\dagger\hat{a}),$$

$$H_0=\omega_0(\hat{\sigma}_z+1)/2+\omega_c\hat{a}^\dagger\hat{a}+\Omega_0(\hat{a}^\dagger\hat{\sigma}_-+\hat{a}\hat{\sigma}_+). \quad (63)$$

This master equation is the well known equation for the damped Jaynes-Cummings model [27] describing the quantum coupling of a two-level atom to a damped mode. However, this master equation is usually derived using perturbation theory.

The master equation is *completely independent* of the function $D(\omega)$, for a single simple pole and within the restrictions described above (i.e., with no branch cuts and integrable through the lower half plane contour). This result is strongly suggestive of the wide applicability of the damped Jaynes-Cummings model, Eq. (63).

The most straightforward example of a single pseudomode is given by the Lorentzian resonance,

$$D(\omega)=\frac{\Gamma}{(\omega-\omega_c)^2+(\Gamma/2)^2}. \quad (64)$$

The coupling of this type of resonance to three- (and two-) level systems has been considered in detail in Ref. [16], but here we note first that for a very narrow cavity resonance, $\Gamma\to 0$ in Eq. (61), we will be able to neglect the dissipative part of Eq. (63) and we then simply obtain the master equation for a two-level system coupled to a single mode. Secondly, in the case that the cavity resonance is very broad, and the coupling strength Ω_0 weak, it is well known that for the master equation (63) we can perform an adiabatic elimination of the field variable (for simplicity we consider the case of cavity-atom resonance where $\hat{a}\approx -2i\Omega_0\sigma_-/\Gamma$). This is done in exactly the same way as the atomic variable is eliminated in the semiclassical theory of the laser (see, e.g., Ref. [23]). This then leads to the well known low-Q master equation for the atomic density matrix alone:

$$\frac{d}{dt}\hat{\rho}_A=-i[H,\hat{\rho}_A]-\frac{\gamma}{2}(\hat{\sigma}_+\hat{\sigma}_-\hat{\rho}_A-2\hat{\sigma}_-\hat{\rho}_A\hat{\sigma}_++\hat{\rho}_A\hat{\sigma}_+\hat{\sigma}_-), \quad (65)$$

where $H=\omega_0(\hat{\sigma}_z+1)/2$ and the spontaneous emission rate γ is

$$\gamma=4\Omega_0^2/\Gamma_0. \quad (66)$$

This is, of course, exactly the peturbative result we obtain by applying Fermi's golden rule to the system (2) with g_λ determined by Eqs. (64) and (4). The dependence of γ on $1/\Gamma$ is exactly as expected from the Purcell formula where $\gamma(Q)\propto Q$, with $Q=\omega_0/\Gamma_0$ [2]. We can also obtain the same limiting result from the pseudomode equations (31) and (32) [16]. In the case of the Lorentzian resonance (64), and a resonant two-level system, Eqs. (31) and (32) take the form

$$i\frac{d}{dt}c_1(t)=\omega_0 c_1(t)+\Omega_0 b_1(t), \tag{67}$$

$$i\frac{d}{dt}b_1(t)=(\omega_0-i\Gamma/2)b_1(t)+\Omega_0 c_1(t), \tag{68}$$

where the only time-dependent variables are c_1 and b_1. For the low-Q cavity, Eq. (68) yields $b_1(t)\approx -2i\Omega_0 c_1(t)/\Gamma$ which can be substituted back into Eq. (67) to yield the same low-Q, approximate, decay rate as in Eq. (66).

B. Example with two Lorentzian modes

We will now give a very simple example of an atomic system coupled to two pseudomodes. In this case we consider a density of states function which is simply a sum of two Lorentzian functions,

$$D(\omega)=W_1\frac{\Gamma_1}{(\omega-\omega_c^{(1)})^2+(\Gamma_1/2)^2}+W_2\frac{\Gamma_2}{(\omega-\omega_c^{(2)})^2+(\Gamma_2/2)^2}, \tag{69}$$

where the weights of the two Lorentzians are such that $W_1+W_2=1$. The two cavity resonances are located at the different frequencies $\omega_c^{(1)}$ and $\omega_c^{(2)}$. This time the poles are located at

$$z_1=\omega_c^{(1)}-i\Gamma_1/2, \quad z_2=\omega_c^{(2)}-i\Gamma_2/2, \tag{70}$$

and the squares of the couplings ($g_{il}^2=-ir_l\Omega_i^2$) are

$$g_{11}^2=W_1\Omega_0^2, \quad g_{12}^2=W_2\Omega_0^2.$$

Then if we follow the procedure of Eqs. (41), (42), and Eq. (1) we simply obtain the master equation

$$\frac{d}{dt}\hat{\rho}=-i[H_0,\hat{\rho}]-\frac{\Gamma_1}{2}(\hat{a}_1^\dagger\hat{a}_1\hat{\rho}-2\hat{a}_1\hat{\rho}\hat{a}_1^\dagger+\hat{\rho}\hat{a}_1^\dagger\hat{a}_1)-\frac{\Gamma_2}{2}(\hat{a}_2^\dagger\hat{a}_2\hat{\rho}-2\hat{a}_2\hat{\rho}\hat{a}_2^\dagger+\hat{\rho}\hat{a}_2^\dagger\hat{a}_2), \tag{71}$$

with the Hamiltonian

$$H_0=\omega_0(\hat{\sigma}_z+1)/2+\omega_c^{(1)}\hat{a}_1^\dagger\hat{a}_1+\omega_c^{(2)}\hat{a}_2^\dagger\hat{a}_2+\Omega_0\sqrt{W_1}(\hat{a}_1^\dagger\hat{\sigma}_-+\hat{a}_1\hat{\sigma}_+)+\Omega_0\sqrt{W_2}(\hat{a}_2^\dagger\hat{\sigma}_-+\hat{a}_2\hat{\sigma}_+). \tag{72}$$

We see that this time we find a two-mode damped Jaynes-Cummings model with the atomic system coupled to two decaying pseudomodes. This result generalizes straightforwardly to multiple Lorentzian resonances and multiple atomic levels of the form given in Fig. 1.

V. COUPLED PSEUDOMODES

A. The non-Lindblad problems associated with complex g_{il}

When we consider more complex functions than Lorentzians for the generalized density of states, we can quickly run into serious difficulties because the couplings g_{il} would no longer be real. (Some examples will be given in Sec. VII.) We can then no longer use the results given in Eqs. (41), (42), and (1). This is a general problem which arises only if we consider more than one pseudomode, and when we have density of states functions which are more complex than Lorentzian functions.

There are at least two types of difficulty. First, we can start with $H_{\rm eff}$ in the form given in Eq. (38) and splitting g_{il} into real and imaginary parts so that

$$g_{il}=g_{il}^{(r)}+ig_{il}^{(i)} \tag{73}$$

we can split $H_{\rm eff}$ into Hermitian and anti-Hermitian parts as we have done in Eq. (40). Then we find for the Hermitian piece

$$H_0=\sum_l \mathrm{Re}(z_l)\hat{a}_l^\dagger\hat{a}_l+\sum_i \omega_i|i\rangle_{aa}\langle i|+\sum_{il} g_{il}^{(r)}(\hat{a}_l^\dagger\hat{\sigma}_-^{(i)}+\hat{a}_l\hat{\sigma}_+^{(i)}), \tag{74}$$

and the anti-Hermitian piece leads to the requirement for the following sum over general Lindblad operators $\hat{L}_m$:

$$\sum_m \hat{L}_m^\dagger\hat{L}_m\equiv -2\sum_l \mathrm{Im}(z_l)\hat{a}_l^\dagger\hat{a}_l-2\sum_{il} g_{il}^{(i)}(\hat{a}_l^\dagger\hat{\sigma}_-^{(i)}+\hat{a}_l\hat{\sigma}_+^{(i)}), \tag{75}$$

which cannot be satisfied. The first sum on the right hand side is not a problem and can be treated using the Lindblads of Eq. (42). The second sum cannot, apparently, be factored into the $\hat{L}_m^\dagger\hat{L}_m$ form. For example, we can rewrite Eq. (75) in the form

$$\sum_m \hat{L}_m^\dagger\hat{L}_m\equiv -2\sum_l \mathrm{Im}(z_l)\hat{a}_l^\dagger\hat{a}_l-2\sum_{il} g_{il}^{(i)}(\alpha_f\hat{a}_l^\dagger+\alpha_a\hat{\sigma}_+^{(i)})\times(\alpha_f\hat{a}_l+\alpha_a\hat{\sigma}_-^{(i)})+2\sum_{il} g_{il}^{(i)}\alpha_f^2\hat{a}_l^\dagger\hat{a}_l+2\sum_{il} g_{il}^{(i)}\alpha_a^2\hat{\sigma}_+^{(i)}\hat{\sigma}_-^{(i)}, \tag{76}$$

where $\alpha_f\alpha_a=1$. The right hand side of this equation is nearly in the required form, but in fact the equivalence can never be satisfied. The reason is that whatever choice is made over the signs of $g_{il}^{(i)}$, some of the terms in the sums will have positive coefficients and some will have negative coefficients. The terms with negative coefficients cannot satisfy the equivalence in Eq. (76).

The second difficulty arises over the identification of the connection between the rate of change of the ground-state population, Eq. (45), with the rate of change of the pseudomode populations in Eq. (46). This is because the coupling g_{il} appears with the wrong phase factors. However, we can still calculate the rate of change of $\Pi_{\rm vac}$, which from Eq. (52) is given by the rate of change of the difference in the ground-state population and total population of the pseudomodes. For the rate of change of the ground-state population

we can use Eq. (45), and for the rate of change of the pseudomode populations we have Eq. (46). The difference is found to be

$$\frac{d}{dt}\Pi_{\rm vac}=-2\sum_l \mathrm{Im}(z_l)|b_l|^2-2\sum_{il} g_{il}^{(i)}(c_i^* b_l+c_i b_l^*). \tag{77}$$

The right hand side of this equation is identical to the expectation value of the right hand side of Eq. (75) evaluated with $|\tilde{\psi}(t)\rangle$ as found in Eq. (35). This means that although we cannot, in this general case write a master equation in the form of Eq. (1), we can write an equation based on the general form

$$\frac{d}{dt}\hat{\rho}=-i[H_0,\hat{\rho}]-\sum_m s_m\left(\frac{1}{2}\hat{L}_m^\dagger\hat{L}_m\hat{\rho}-\hat{L}_m\hat{\rho}\hat{L}_m^\dagger+\frac{1}{2}\hat{\rho}\hat{L}_m^\dagger\hat{L}_m\right), \tag{78}$$

where $s_m=\pm 1$ controls the signs of the Lindblad terms. Then by using Eq. (76) we can identify four types of ''Lindblad'' as

$$L_l^{(1)}=\sqrt{-2\,\mathrm{Im}(z_l)}\hat{a}_l,\quad L_{il}^{(2)}=\sqrt{2|g_{il}^{(i)}|}(\alpha_f\hat{a}_l+\alpha_a\hat{\sigma}_-^{(i)}),$$
$$L_{il}^{(3)}=\sqrt{2|g_{il}^{(i)}|}\alpha_f\hat{a}_l,\quad L_{il}^{(4)}=\sqrt{2|g_{il}^{(i)}|}\alpha_a\hat{\sigma}_-^{(i)}, \tag{79}$$

where

$$s_{il}^{(2)}=\sigma(-g_{il}^{(i)}),\quad s_{il}^{(3)}=\sigma(g_{il}^{(i)}),\quad s_{il}^{(4)}=\sigma(g_{il}^{(i)}), \tag{80}$$

and where the function $\sigma(x)$ is used to indicate the sign of x.

This approach is not, however, satisfactory from a physical point of view. In a different context [28,29] a master equation of the more general form (78) has been found to be defective because positive probabilities cannot be assigned to component processes. This fact is illustrated clearly by the process of master equation unraveling [26]. The master equation (1) can be unraveled into stochastic processes for a normalized state vector $|\Psi\rangle$ by the statement that over an infinitesimally short time Δt there is a probability $\langle\Psi|\hat{L}_l^\dagger\hat{L}_l|\Psi\rangle\Delta t$ for the transition

$$|\Psi\rangle\rightarrow\frac{\hat{L}_l|\Psi\rangle}{\sqrt{\langle\Psi|\hat{L}_l^\dagger\hat{L}_l|\Psi\rangle}} \tag{81}$$

and there is a probability $1-\Sigma_l\langle\Psi|\hat{L}_l^\dagger\hat{L}_l|\Psi\rangle\Delta t$ for the transition

$$|\Psi\rangle\rightarrow\frac{\left(1-iH_0\Delta t-(\Delta t/2)\sum_l \hat{L}_l^\dagger\hat{L}_l\right)|\Psi\rangle}{\sqrt{1-\sum_l \langle\Psi|\hat{L}_l^\dagger\hat{L}_l|\Psi\rangle\Delta t}}. \tag{82}$$

The statements (81) and (82) are exactly equivalent to the master equation (1) when used as part of a stochastic simulation [26]. These kinds of simulations correspond to real physical processes for detecting system excitations. However, the problem with the master equation (78) is that it cannot be simulated in the manner of Eqs. (81) and (82) because the probabilities of some of the processes will be negative according to the signs s_m given in Eq. (80).

Thus we have seen that although the equations (31) and (32) yield the correct results for the atomic system amplitudes $c_i(t)$, the behavior of the amplitudes b_l can be clearly unphysical in the case when the g_{il} are complex. Especially when the coupling is strong and the dissipation is weak we can find that the pseudomode amplitudes of Eq. (30) can exceed unity for complex g_{il}. We will correct this problem in the remaining part of this paper (by means of an adjustment to the pseudomodes), but here we would like to point out the close affinity between this problem, complex couplings g_{il}, and the pathological master equations (78).

B. Lindblad formulation for two pseudomodes

We will now resolve the problems discussed in the preceding section by means of a rotation of the pseudomode basis. We will consider the case of a single atomic transition coupled to two pseudomodes. The coupled equations (31) and (32) are then

$$i\frac{d}{dt}c(t)=\omega c(t)+g_1 b_1(t)+g_2 b_2(t), \tag{83}$$

$$i\frac{d}{dt}b_1(t)=z_1 b_1(t)+g_1 c(t),$$

$$i\frac{d}{dt}b_2(t)=z_2 b_2(t)+g_2 c(t), \tag{84}$$

where c ($\equiv c_1$) now stands for the amplitude of the single atomic transition and b_1 and b_2 are the amplitudes of the two pseudomodes as defined in Eq. (30). The couplings $g_l=\sqrt{-ir_l}\Omega_0$ may now be complex, but they are constrained by the addition rule of Eq. (34) so that $\Sigma_l g_l^2=\Omega_0^2$, where Ω_0^2 ($\equiv\Omega_1^2$) is the strength of the single atomic transition as in Eq. (6).

Equations (83) and (84) can be placed in a matrix form

$$i\frac{d}{dt}c=\omega_0 c+\tilde{\mathbf{g}}\mathbf{b},\quad i\frac{d}{dt}\mathbf{b}=\mathbf{Z}\mathbf{b}+\mathbf{g}c, \tag{85}$$

where

$$\mathbf{Z}=\begin{bmatrix} z_1 & 0\\ 0 & z_2\end{bmatrix},\quad \mathbf{g}=\begin{pmatrix} g_1\\ g_2\end{pmatrix},\quad \mathbf{b}=\begin{pmatrix} b_1\\ b_2\end{pmatrix} \tag{86}$$

are all complex quantities.

Now we apply to the pseudomodes $\mathbf{b}$ the complex rotation matrix

$$\mathbf{R}(\beta_0)=\begin{bmatrix} \cos\beta_0 & \sin\beta_0\\ -\sin\beta_0 & \cos\beta_0\end{bmatrix}, \tag{87}$$

where β_0 may be complex. Clearly, $\mathbf{R}(\beta_0)$ is not, in general, unitary, but is always orthogonal in the sense that $\mathbf{R}(\beta_0)\tilde{\mathbf{R}}(\beta_0)=1$. The transformation will not affect any of the atomic dynamics. The transformed modes will be denoted

$$\mathbf{b}'=\mathbf{R}(\beta_0)\mathbf{b}. \tag{88}$$

The effect on $\mathbf{Z}$ is in the form of a similarity transformation so that

$$\mathbf{Z}'=\mathbf{R}(\beta_0)\mathbf{Z}\widetilde{\mathbf{R}}(\beta_0)=\frac{z_1+z_2}{2}+\frac{\Delta z}{2}\begin{bmatrix}-\cos 2\beta_0 & \sin 2\beta_0\\ \sin 2\beta_0 & \cos 2\beta_0\end{bmatrix}, \tag{89}$$

where the difference in the complex locations of the two pseudomodes is simply

$$\Delta z=z_2-z_1. \tag{90}$$

We will parametrize the complex couplings g_l by a *complex* angle β_g such that

$$\mathbf{g}=\Omega_0\begin{pmatrix}\cos\beta_g\\ \sin\beta_g\end{pmatrix}, \tag{91}$$

which means that the two complex pseudomode couplings are characterized by a complex ratio μ where

$$\mu=\tan\beta_g=g_2/g_1. \tag{92}$$

The complex rotation of the pseudomode basis means that we obtain the new couplings

$$\mathbf{g}'=\mathbf{R}(\beta_0)\mathbf{g}=\Omega_0\mathbf{R}(\beta_0-\beta_g)\begin{pmatrix}1\\ 0\end{pmatrix}, \tag{93}$$

where we have used the addition formula to combine two rotations on the right hand side. As in Eq. (91) $\Sigma_l g_l^2=\widetilde{\mathbf{g}}\mathbf{g}=\widetilde{\mathbf{g}}'\mathbf{g}'=\Omega_0^2$ is preserved during the rotations.

We have now set up the basic definitions required, so that the rotated pseudomode equations become

$$i\frac{d}{dt}c=\omega c+\widetilde{\mathbf{g}}'\mathbf{b}, \tag{94}$$

$$i\frac{d}{dt}\mathbf{b}'=\mathbf{Z}'\mathbf{b}'+\mathbf{g}'c, \tag{95}$$

where c and the time evolution of c are unaffected by the transformation. Now the angle of rotation β_0 is determined by the two important constraints on the physical form of Eqs. (94) and (95). First, the off-diagonal parts of $\mathbf{Z}'$ must be Hermitian. If they are not Hermitian we still have the same kinds of problems as discussed in Sec. V A. Inspection of the transformed matrix, Eq. (89), shows that the two off-diagonal elements are equal and hence must be real. Denoting the off-diagonal element by the real parameter $V_{12}(=V_{21})$ we have the restriction

$$V_{12}=\frac{\Delta z\sin 2\beta_0}{2}. \tag{96}$$

The second condition arises because the transformed atom-mode couplings $\mathbf{g}'$ and $\widetilde{\mathbf{g}}'$ must, in general, be Hermitian conjugates. Because the rotation matrix we are using is not unitary, we will have

$$\mathbf{g}'=\mathbf{R}(\beta_0)\mathbf{g}=[\widetilde{\mathbf{g}}\widetilde{\mathbf{R}}(\beta_0)]^\dagger=[\mathbf{R}(\beta_0)\mathbf{g}]^*=[\mathbf{g}']^* \tag{97}$$

and we are constrained to have $\mathbf{g}'$ real. This means that $\mathbf{g}'$ can be described by a *real* rotation in the form

$$\mathbf{g}'=\Omega_0\mathbf{R}(\theta_g)\begin{pmatrix}1\\ 0\end{pmatrix}. \tag{98}$$

Now if we compare this definition of $\mathbf{g}'$ with Eq. (93) we obtain the second condition as

$$\mathbf{R}(\beta_0-\beta_g)=\mathbf{R}(\theta_g) \tag{99}$$

or on using the addition formula for rotations,

$$\theta_g=\beta_0-\beta_g. \tag{100}$$

The task now is to determine the transformed matrices $\mathbf{Z}'$ and $\mathbf{g}'$ with the imposed constraints (96) and (100). This becomes an algebraic problem. We will separate β_0 and β_g into their real and imaginary parts: $\beta_0^{(r)}$, $\beta_0^{(i)}$ and $\beta_g^{(r)}$, $\beta_g^{(i)}$. Then we have from the imaginary part of Eq. (100)

$$\beta_0^{(i)}=\beta_g^{(i)}, \tag{101}$$

and if we also separate the modulus and phase of Δz so that

$$\Delta z=|\Delta z|\exp(i\theta_z), \tag{102}$$

and utilize Eq. (101), we can rewrite Eq. (96) in the form

$$V_{12}=\frac{|\Delta z|e^{i\theta_z}}{2}(\sin 2\beta_0^{(r)}\cosh 2\beta_g^{(i)}+i\cos 2\beta_0^{(r)}\sinh 2\beta_g^{(i)}). \tag{103}$$

Then by considering the real and imaginary parts of Eq. (103), which must be real, we obtain

$$\tan 2\beta_0^{(r)}=-\frac{\tanh 2\beta_g^{(i)}}{\tan\theta_z} \tag{104}$$

and

$$V_{12}=\frac{|\Delta z|}{2}\frac{\sinh 2\beta_g^{(i)}}{\sqrt{\sin^2\theta_z+\cos^2\theta_z\tanh^2 2\beta_g^{(i)}}}, \tag{105}$$

where we have chosen the angle $\beta_0^{(r)}$ to lie in the quadrant after θ_z for $\beta_g^{(i)}>0$. This will ensure that V_{12} is positive unless $\beta_g^{(i)}<0$ in which case it is possible, if preferred, to ensure V_{12} is positive by exchanging the labels of the two pseudomodes. Thus we have determined, in principle, V_{12} [from Eq. (105)] and β_0 (from Eqs. (104) and (101)].

To obtain the diagonal elements of $\mathbf{Z}'$ (i.e., z_1', z_2') we need $\cos 2\beta_0$ in Eq. (89). We can expand $\cos 2\beta_0$ in terms of the real and imaginary parts of $2\beta_0$ and then use Eqs. (101) and (104) to obtain

$$\Delta z' = z_2' - z_1' = \Delta z \cos 2\beta_0 \tag{106}$$

$$= -\Delta z \cosh 2\beta_g^{(i)} \frac{\sin\theta_z + i\cos\theta_z \tanh 2\beta_g^{(i)}}{\sqrt{\sin^2\theta_z + \cos^2\theta_z \tanh^2 2\beta_g^{(i)}}}. \tag{107}$$

Then to obtain $\mathbf{g}'$ [Eq. (93)] i.e.,

$$g_1' = \Omega_0 \cos\theta_g, \quad g_2' = -\Omega_0 \sin\theta_g, \tag{108}$$

we can use a similar approach. Because θ_g is real, we have from Eq. (101) $\theta_g = \beta_0^{(r)} - \beta_g^{(r)}$, and so by expanding $\cos 2\theta_g$ in terms of $\beta_0^{(r)}$ and $\beta_g^{(r)}$ and using Eq. (104) we may obtain

$$\cos 2\theta_g = \frac{\sin 2\beta_g^{(r)} \cos\theta_z \tanh 2\beta_g^{(i)} - \cos 2\beta_g^{(r)} \sin\theta_z}{\sqrt{\sin^2\theta_z + \cos^2\theta_z \tanh^2 2\beta_g^{(i)}}} \tag{109}$$

from which the two couplings can be determined by the use of half-angle formulas.

In practice, it may be preferable to determine V_{12}, $\mathbf{Z}'$ and $\mathbf{g}'$ in terms of z_1, z_2 (or Δz and θ_z) and the coupling ratio $\mu = \mu^{(r)} + i\mu^{(i)}$ [Eq. (92)]. To this end we first consider the real and imaginary parts of β_g, and we find from Eq. (92)

$$\sin 2\beta_g^{(r)} = \frac{2\mu^{(r)}}{|1+\mu^2|}, \quad \cos 2\beta_g^{(r)} = \frac{1-|\mu|^2}{|1+\mu^2|},$$

$$\sinh 2\beta_g^{(i)} = \frac{2\mu^{(i)}}{|1+\mu^2|}, \quad \cosh 2\beta_g^{(i)} = \frac{1+|\mu|^2}{|1+\mu^2|}. \tag{110}$$

This also shows that $\mu^{(i)}$ is responsible for the sign changes in V_{12} in Eq. (105) for the given choice of $\beta_0^{(r)}$. By utilizing these equations in Eqs. (105), (107), and (109) we obtain the following results:

$$V_{12} = -\frac{|\Delta z|\mu^{(i)}}{|1+\mu^2|} \frac{1+|\mu|^2}{\sqrt{\sin^2\theta_z (1+|\mu|^2)^2 + (2\mu^{(i)}\cos\theta_z)^2}}, \tag{111}$$

$$\Delta z' = \frac{-\Delta z}{|1+\mu^2|} \frac{(1+|\mu|^2)^2 \sin\theta_z + i(2\mu^{(i)})^2 \cos\theta_z}{\sqrt{(1+|\mu|^2)^2 \sin^2\theta_z + (2\mu^{(i)}\cos\theta_z)^2}}, \tag{112}$$

and

$$(g_1')^2 = \frac{\Omega_0^2}{2}\left[1 - \frac{(1-|\mu|^4)\sin\theta_z - 4\mu^{(r)}\mu^{(i)}\cos\theta_z}{|1+\mu^2|\sqrt{(1+|\mu|^2)^2\sin^2\theta_z + (2\mu^{(i)}\cos\theta_z)^2}}\right],$$

$$(g_2')^2 = \frac{\Omega_0^2}{2}\left[1 + \frac{(1-|\mu|^4)\sin\theta_z - 4\mu^{(r)}\mu^{(i)}\cos\theta_z}{|1+\mu^2|\sqrt{(1+|\mu|^2)^2\sin^2\theta_z + (2\mu^{(i)}\cos\theta_z)^2}}\right]. \tag{113}$$

C. Parallel poles

1. Real Δz

In this section we consider some special cases of Eqs. (111)–(113) above, namely, for the situations that arise when the poles are parallel with either the real or imaginary axes in the complex ω plane. These results will be useful for application to some particular examples in Sec. VII and will be given in terms of both the coupling ratio μ and the couplings g_1, g_2. If the poles are parallel to the real axis, we will have Δz real ($\theta_z = 0, \pi$), and we find for the coupling

$$V_{12} = \left|\frac{\Delta z(1+|\mu|^2)}{2(1+\mu^2)}\right| \sigma(\mu^{(r)})$$

$$= \frac{|\Delta z|}{2}[(|g_1|^2 + |g_2|^2)/\Omega_0^2]\sigma(\mu^{(r)}), \tag{114}$$

where the function $\sigma(x)$ is again used to indicate the sign of x: $\sigma(x) = \pm 1$. Then for the new difference in diagonal elements,

$$\Delta z' = -2i\left|\frac{\Delta z \mu^{(i)}}{1+\mu^2}\right| = -2i|\Delta z \,\mathrm{Im}(g_1^* g_2)|/\Omega_0^2, \tag{115}$$

and for the squares of the new couplings,

$$(g_1')^2 = \frac{\Omega_0^2}{2}\left[1 + \frac{2\mu^{(r)}}{|1+\mu^2|}\sigma(\mu^{(i)})\sigma(\Delta z)\right]$$

$$= \frac{\Omega_0^2}{2}\left[1 + \frac{1}{2}(g_1^* g_2 + g_1 g_2^*)/\Omega_0^2 \sigma(\mu^{(i)})\sigma(\Delta z)\right],$$

$$(g_2')^2 = \frac{\Omega_0^2}{2}\left[1 - \frac{2\mu^{(r)}}{|1+\mu^2|}\sigma(\mu^{(i)})\sigma(\Delta z)\right]$$

$$= \frac{\Omega_0^2}{2}\left[1 - \frac{1}{2}(g_1^* g_2 + g_1 g_2^*)/\Omega_0^2 \sigma(\mu^{(i)})\sigma(\Delta z)\right]. \tag{116}$$

If the two couplings g_1 and g_2 are equal in magnitude then we may let

$$\mu = \exp(2i\phi) \tag{117}$$

and Eqs. (114)–(116) may be simplified to

$$V_{12}=\left|\frac{\Delta z}{2\cos 2\phi}\right|\sigma(\mu^{(r)})=|\Delta z||g_1|^2/\Omega_0^2\sigma(-\mathrm{Im}(g_1^2)),$$

$$\Delta z'=-i|\Delta z\tan 2\phi|=-2i|\Delta z(g_2^2-g_1^2)|/\Omega_0^2,$$

$$(g_1')^2=\frac{\Omega_0^2}{2}[1+\sigma(\mu^{(i)})\sigma(\Delta z)],$$

$$(g_2')^2=\frac{\Omega_0^2}{2}[1-\sigma(\mu^{(i)})\sigma(\Delta z)]. \tag{118}$$

2. Imaginary Δz

If the poles are parallel to the imaginary axis, we will have Δz imaginary ($\theta_z=\pm\pi/2$) and we find that

$$V_{12}=\left|\frac{\Delta z}{1+\mu^2}\right|\mu^{(i)}=|\Delta z|\mathrm{Im}(g_1^* g_2)/\Omega_0^2,$$

$$\Delta z'=-i\left|\frac{\Delta z(1+|\mu|^2)}{1+\mu^2}\right|=-i|\Delta z|(|g_1|^2+|g_2|^2)/\Omega_0^2,$$

$$(g_1')^2=\frac{\Omega_0^2}{2}\left[1-\frac{1+|\mu|^2}{|1+\mu^2|}\sigma(\mathrm{Im}(\Delta z))\right]$$
$$=\frac{\Omega_0^2}{2}\{1-(|g_1|^2-|g_2|^2)/\Omega_0^2\sigma(\mathrm{Im}(\Delta z))\},$$

$$(g_2')^2=\frac{\Omega_0^2}{2}\left[1+\frac{1-|\mu|^2}{|1+\mu^2|}\sigma(\mathrm{Im}(\Delta z))\right]$$
$$=\frac{\Omega_0^2}{2}\{1+(|g_1|^2-|g_2|^2)/\Omega_0^2\sigma(\mathrm{Im}(\Delta z))\}. \tag{119}$$

Again, if the two couplings g_1 and g_2 are equal in magnitude (complex conjugates) then

$$V_{12}=\frac{|\Delta z|\tan 2\phi}{2}\sigma(\cos 2\phi)=\frac{|\Delta z|}{2}(g_2^2-g_1^2)/\Omega_0^2,$$

$$\Delta z'=-i\left|\frac{\Delta z}{\cos 2\phi}\right|=-2i|\Delta z||g_1|^2/\Omega_0^2,$$

$$g_1'=\Omega_0/\sqrt{2}, g_2'=\Omega_0/\sqrt{2}. \tag{120}$$

VI. COUPLED PSEUDOMODE MASTER EQUATION

We will now generate the master equation corresponding to Eqs. (94) and (95), and we return to a general notation for multiple levels and pseudomodes as in Sec. III. The procedure given in the preceding section easily extends to multiple levels, but extends less readily to multiple pseudomodes. This time, then, Eqs. (94) and (95) can be represented by a state vector $|\tilde{\psi}'(t)\rangle$ [in the basis (36)] and an effective non-Hermitian Hamiltonian which takes the form

$$H_{\mathrm{eff}}=\sum_l z_l'\hat{a}_l^\dagger\hat{a}_l+\sum_i \omega_i|i\rangle_{a\,a}\langle i|+\sum_{il} g_{il}'(\hat{a}_l^\dagger\hat{\sigma}_-^{(i)}+\hat{a}_l\hat{\sigma}_+^{(i)})$$
$$+\sum_{ll'} V_{ll'}(\hat{a}_l^\dagger\hat{a}_{l'}+\hat{a}_{l'}^\dagger\hat{a}_l). \tag{121}$$

For the treatment of the preceding section $l,l'=1,2$. This is quite analogous to Eq. (38), but with the addition of the mode-mode coupling term. Then Eqs. (94) and (95) are reproduced by the equation

$$\frac{d}{dt}|\tilde{\psi}'(t)\rangle=-iH_{\mathrm{eff}}|\tilde{\psi}'(t)\rangle \tag{122}$$

as in Eq. (39). The effective Hamiltonian H_{eff} is again split into a Hermitian part and an anti-Hermitian part in the form

$$H_{\mathrm{eff}}=H_0-\frac{i}{2}\sum_l \hat{L}_l^\dagger\hat{L}_l \tag{123}$$

and now the Hamiltonian is

$$H_0=\sum_l \mathrm{Re}(z_l')\hat{a}_l^\dagger\hat{a}_l+\sum_i \omega_i|i\rangle_{a\,a}\langle i|+\sum_{il} g_{il}'(\hat{a}_l^\dagger\hat{\sigma}_-^{(i)}$$
$$+\hat{a}_l\hat{\sigma}_+^{(i)})+\sum_{ll'} V_{ll'}(\hat{a}_l^\dagger\hat{a}_{l'}+\hat{a}_{l'}^\dagger\hat{a}_l), \tag{124}$$

where the new term is the mode-mode coupling $V_{ll'}$. The Lindblad operators for the pseudo-modes are

$$\hat{L}_l=\sqrt{-2\,\mathrm{Im}(z_l')}\hat{a}_l. \tag{125}$$

To obtain the appropriate master equation we now simply insert the Hamiltonian and Lindblad operator into Eq. (1). The justification for this is that the introduction of the coupling $V_{ll'}$ does not change the differential equation for the ground-state population, Eq. (45), and thus Eq. (47) is unchanged as is the master equation (57) and the solution (59) with the substitution of $|\tilde{\psi}'(t)\rangle$ for $|\tilde{\psi}(t)\rangle$.

VII. EXAMPLES OF COUPLED PSEUDOMODES

A. Master equation for a non-Lorentzian density of states

We will now apply the theory given above to a density of states function $D(\omega)$ which is not Lorentzian and contains two poles in the lower half plane. The normalized function is

$$D(\omega)=\frac{4\Gamma^3/\sqrt{2}}{(\omega-\omega_c)^4+\Gamma^4}, \tag{126}$$

where ω_c is the center frequency of the resonance, and Γ is the width of the resonance. In this case we find four poles, of which two are located in the lower half plane at

$$z_1=\omega_c-\frac{\Gamma}{\sqrt{2}}(1+i),\quad z_2=\omega_c+\frac{\Gamma}{\sqrt{2}}(1-i). \tag{127}$$

We note that $\Delta z=z_2-z_1=\sqrt{2}\Gamma$ is real. We now evaluate the residues at z_1 and z_2 we obtain the squares of the couplings ($g_{il}^2=-ir_l\Omega_0^2$) which are

$$g_{11}^2=\frac{1}{2}(1-i)\Omega_0^2, \quad g_{12}^2=\frac{1}{2}(1+i)\Omega_0^2.$$

It follows that the pseudomode couplings to the atom are complex, and they are complex conjugates with equal magnitude. Because Δz is real we can use Eqs. (118) with $\phi=\pi/8$. Then if we follow the pseudomode coupling procedure we obtain

$$V_{12}=\Gamma, \quad z_1'=\omega_c-i\sqrt{2}\Gamma, \quad z_2'=\omega_c,$$
$$g_{11}'=0, \quad g_{12}'=\Omega_0. \tag{128}$$

Thus we see that in the proper pseudomode basis the atom does not couple to the first pseudomode at all. In fact it only couples to the second pseudomode which is in turn coupled to the first pseudomode. Also, only the first pseudomode will show any decay—the mode which is directly coupled to the atom does not decay in this model.

We now proceed to the master equation by following the procedure of Eqs. (124), (125), and Eq. (1) to obtain

$$\frac{d}{dt}\hat{\rho}=-i[H_0,\hat{\rho}]-\sqrt{2}\Gamma(\hat{a}_1^\dagger\hat{a}_1\hat{\rho}-2\hat{a}_1\hat{\rho}\hat{a}_1^\dagger+\hat{\rho}\hat{a}_1^\dagger\hat{a}_1), \tag{129}$$

with the Hamiltonian

$$H_0=\omega_0(\hat{\sigma}_z+1)/2+\omega_c\hat{a}_1^\dagger\hat{a}_1+\omega_c\hat{a}_2^\dagger\hat{a}_2+\Gamma(\hat{a}_1^\dagger\hat{a}_2+\hat{a}_1\hat{a}_2^\dagger)$$
$$+\Omega_0(\hat{a}_2^\dagger\hat{\sigma}_-+\hat{a}_2\hat{\sigma}_+). \tag{130}$$

B. Example of a band-gap model

We have already seen in Sec. IV B and Eqs. (71) and (72) how a combination of two Lorentzians results in a master equation involving two pseudomodes. In that case all the coupling coefficients were positive. However, in this section we consider an idealized model of a band gap (or photon density of states gap) in which both Lorentzians are centered at the same frequency, and one of them is given a negative weighting so that

$$D(\omega)=W_1\frac{\Gamma_1}{(\omega-\omega_c)^2+(\Gamma_1/2)^2}-W_2\frac{\Gamma_2}{(\omega-\omega_c)^2+(\Gamma_2/2)^2}, \tag{131}$$

where now the weights of the two Lorentzians are such that $W_1-W_2=1$. The effect of the Lorentzian with negative weight is to introduce a dip into the density of states function $D(\omega)$ where the coupling of the atom will be inhibited. We note that this type of density of states function is not easily expressed as a sum of positive Lorentzians, which makes the non-Markovian approach of Ref. [30] difficult to apply. The time evolution for a related problem (with $\Gamma_1\to\infty$) has been solved in Ref. [6] by using a Laplace technique similar to the simple version described in this paper in Sec. VIII. Laplace transforms have also been used in the study of band gaps in Refs. [9,10], but the pseudomodes were not identified.

The focus here is to identify the master equation associated with the density of states gap in Eq. (131). Because the density of states function must be positive we have $W_1\Gamma_1>W_2\Gamma_2$ [condition for $D(\omega)$ to be positive at large ω], and $W_1/\Gamma_1>W_2/\Gamma_2$ [condition for $D(\omega)$ to be positive at the center of the resonance]. Combining these two relations we also have the condition $\Gamma_1>\Gamma_2$, which is also the condition for a localized dip. We now determine the locations of the poles at

$$z_1=\omega_c-i\Gamma_1/2, \quad z_2=\omega_c-i\Gamma_2/2, \tag{132}$$

and the squares of the couplings ($g_{il}^2=-ir_l\Omega_0^2$) are

$$g_{11}^2=W_1\Omega_0^2, \quad g_{12}^2=-W_2\Omega_0^2. \tag{133}$$

In this case we have Δz imaginary and the coupling ratio is $\mu=i\sqrt{W_2/W_1}$. If we now apply Eqs. (119) with the understanding that $\Gamma_1>\Gamma_2$ we find

$$V_{12}=\frac{\sqrt{W_1W_2}(\Gamma_1-\Gamma_2)}{2},$$
$$\Delta z'=-i\frac{\Gamma_1-\Gamma_2}{2}(W_1+W_2),$$
$$g_1'=0,$$
$$g_2'=\Omega_0. \tag{134}$$

Then if we follow the procedure of Eqs. (124), (125), and Eq. (1) we obtain the master equation

$$\frac{d}{dt}\hat{\rho}=-i[H_0,\hat{\rho}]-\frac{\Gamma_1'}{2}(\hat{a}_1^\dagger\hat{a}_1\hat{\rho}-2\hat{a}_1\hat{\rho}\hat{a}_1^\dagger+\hat{\rho}\hat{a}_1^\dagger\hat{a}_1)$$
$$-\frac{\Gamma_2'}{2}(\hat{a}_2^\dagger\hat{a}_2\hat{\rho}-2\hat{a}_2\hat{\rho}\hat{a}_2^\dagger+\hat{\rho}\hat{a}_2^\dagger\hat{a}_2), \tag{135}$$

with the Hamiltonian

$$H_0=\omega_0(\hat{\sigma}_z+1)/2+\omega_c\hat{a}_1^\dagger\hat{a}_1+\omega_c\hat{a}_2^\dagger\hat{a}_2$$
$$+\frac{\sqrt{W_1W_2}(\Gamma_1-\Gamma_2)}{2}(\hat{a}_1^\dagger\hat{a}_2+\hat{a}_1\hat{a}_2^\dagger)$$
$$+\Omega_0(\hat{a}_2^\dagger\hat{\sigma}_-+\hat{a}_2\hat{\sigma}_+). \tag{136}$$

The new decay rates of the two pseudomodes are $\Gamma_l'=-2\,\mathrm{Im}(z_l')$,

$$\Gamma_1'=\Gamma_1\Gamma_2\left(\frac{W_1}{\Gamma_1}-\frac{W_2}{\Gamma_2}\right), \quad \Gamma_2'=W_1\Gamma_1-W_2\Gamma_2, \tag{137}$$

which are both positive given the constraints on Γ_1 and Γ_2 above. The decay rate Γ_1' is proportional to the center height of the density of states and thus the first pseudomode, which is not directly coupled to the atom, may be associated with a band-gap mode. The decay rate Γ_2' appears in the large ω behavior of $D(\omega)$ and may be associated with the decay of a background mode. If the density of states is reduced to zero at the center of the gap, the decay rate of the band-gap mode, which is only coupled to the background mode, will be zero. This leads to population trapping because there is then a superposition of the atomic state and band-gap mode which

does not decay (provided the atomic transition is resonant with the band-gap center). If the density of states does not quite reduce to zero at the band-gap center, there is some leakage from the band-gap mode which allows the steady removal of the initially trapped population.

Finally, the master equation (135) is entirely consistent (in the limit $\Gamma_1 \to \infty$) with the Laplace transform solution for the upper state amplitude found in the band-gap model of Nabiev *et al.* [6]. They considered a perfect gap in a uniform (background) density of states. The limit is found if we ensure that the mode density at the band-gap center is exactly zero, $W_1 = \Gamma_1/(\Gamma_1 - \Gamma_2), W_2 = \Gamma_2/(\Gamma_1 - \Gamma_2)$, and is easily confirmed by comparing their Laplace transform solution for the upper atomic state amplitude with the general Laplace transform solution (142) given below. The parameters z_l and g_{1l} are determined by Eqs. (132) and (133) and we will then find (for a ''resonant'' gap)

$$\overline{c(p - i\omega_c)} = \frac{p + \Gamma_2/2}{p(p + \Gamma_2/2) + \Omega_0^2 - (\Omega_0^2 \Gamma_1/2)/(p + \Gamma_1/2)}. \tag{138}$$

This corresponds to the solution of Nabiev *et al.* [6] when we consider the limit $\Gamma_1 \to \infty$, and insert the decay rate to the Γ_1 pseudomode $\gamma = 4\Omega_0^2/\Gamma_1$ [as in Eq. (66)]:

$$\overline{c(p - i\omega_c)} \to \frac{p + \Gamma_2/2}{p(p + \Gamma_2/2) + \gamma p/2} \tag{139}$$

(though to conform with the notation of Ref. [6] we should let $\Gamma_2/2 \to \Gamma$). This is now easily inverted to obtain [6]

$$c(t) = \{c_\infty + (1 - c_\infty)\exp[-(\Gamma_2 + \gamma)t/2]\} e^{-i\omega_c t}, \tag{140}$$

with $c_\infty = \Gamma_2/(\Gamma_2 + \gamma)$. This result provides a clear example of the population trapping described above.

VIII. THE LAPLACE METHOD OF SOLUTION

In this section we briefly summarize the Laplace approach to the solution of the analytic equations (31) and (32). In fact we can directly Laplace transform the integro-differential equation (29) to obtain the coupled linear equations

$$(ip - \omega_i)\overline{c_i}(p) = ic_i(0) - \Omega_i \sum_{jl} \frac{r_l}{p + iz_l} \Omega_j \overline{c_j}(p), \tag{141}$$

which can, in principle be solved for the transformed amplitudes $\overline{c_i}(p)$. The time dependence can then be found if the Laplace transform can be inverted. In the case of a single atomic transition the problem becomes that of Laplace inverting

$$\overline{c(p)} = \left[p + i\omega_1 + \sum_l \frac{g_{1l}^2}{p + iz_l} \right]^{-1}. \tag{142}$$

IX. CONCLUSION

In this paper we have seen the development of a very general, exact, and nonperturbative approach to the decay of an atomic system coupled to a nonuniform density of states describing the environment. The theoretical treatment could apply to many situations, such as atoms in cavities or excitons in quantum wells and is especially appropriate to strongly coupled systems. The first result is the derivation of the coupled, linear, first order equations (31) and (32). These allow a straightforward numerical solution of many problems. The second result is the derivation of an associated master equation defined by an effective Hamiltonian (41) and Lindblad operators (42). This development allows us to have many of the insights acquired in the study of master equations in quantum optics. It also allows a physical interpretation of the differential equations. That physical interpretation is in terms of the coupling of the atomic system to *pseudomodes*. These pseudomodes are strongly allied to poles in the density of states function which are located in the lower half complex plane. The decay of the pseudomodes leads to master equations which are relatives of the damped Jaynes-Cummings model with a single excitation. Further, we have seen that a universal feature of systems containing a single simple density of states pole, or pseudomode, is the realization of the Jaynes-Cummings model with appropriate parameters [Eq. (63)].

The third result is the removal of difficulties present in some systems with multiple pseudomodes. We find that if the pseudomode is defined in a straightforward way by Eq. (30) we can obtain a pathological master equation. Although the atomic dynamics is correctly described by the differential equations (31) and (32), the pseudomodes can develop populations that are greater than unity in the strong-coupling regime, and then the associated master equation is not in the form of Eq. (1), but in the form of Eq. (78). These difficulties have been removed by utilizing an appropriate (nonunitary) transformation in Sec. V which couples the pseudomodes in a basis where each pseudomode has its own decay channel.

The theoretical predictions of this paper have been tested using numerical models for two- and three-level systems coupled to various types of continuum. The fundamental Hamiltonian (2) can be modeled by a discrete bath with a large, but finite number of reservoir modes. Then the numerical solution of the problem amounts to integrating Eqs. (16) and (17) with a large but finite number of modes λ. This is then the same approach as that taken in the work of Swain [31], except that here we use a frequency-dependent coupling and a sufficiently dense set of modes to represent a continuum and prevent spurious revivals. The results for the atomic dynamics (populations, or coherences) from this bath model can then be compared to the integration of the much simpler pseudomode equations (31) and (32) or the appropriate master equation such as Eqs. (63), (129), or (135). With an appropriately dense set of reservoir modes the agreement between all three numerical methods is excellent.

It is appropriate to mention here some of the weaknesses of the approach taken in this paper. Certainly, the restriction to analytic functions for $D(\omega)$, where a contour can be taken in the lower half plane, is restrictive, though it also allows some considerable insight into the atomic decay. The exclusion of branch cuts in the lower half plane is a restriction that cannot be avoided. Against this, the method provided should be very appropriate for handling polynomial approximations to the density of states, of the form $P(\omega)/Q(\omega)$.

The master equations that have been derived are exact results, but of course, in the systems that have been studied,

there are no elements of driving fields or pumping. The effects of strong, classical driving, would almost certainly have to be treated by perturbation theory, but it seems plausible that master equations of the type derived in this paper should provide a better framework for the expressions of perturbation theory close to, or in, the strong-coupling regime.

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