Decay of an atom coupled strongly to a reservoir

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An approach to the decay of a two-state quantum system coupled to a resonant environment is developed in terms of *exact* (nonperturbative) master equations. Starting from a quantum optical model of a two-level system coupled to a heatbath, it is shown that the effect of the bath can be replaced by one or more *pseudomodes*. This description is valid for a wide class of analytic density-of-state functions and it leads to exact master equations which fully describe the non-Markovian decay of the quantum system without the use of perturbation theory, the Born approximation, or the Markov approximation. Two examples are given: a simple model of a density-of-states gap which has two poles, and a non-Lorentzian resonance. [S1050-2947(97)09906-X]

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The decay of quantum systems has long been of interest and encompasses the fundamental problem of a small quantum system coupling to a large world. The approaches used are many and include Fermi's golden rule, the Weisskopf-Wigner approach, the method of Heitler-Ma, Goldberger-Watson, and the Lehmberg and Agarwal master equations [1]. The master equation approaches, describing the time evolution of the density matrix, are usually used in conjunction with time-dependent perturbation theory. The purpose of this short paper, however, is to show that master equations can be used to describe *exactly* the atomic dynamics of the decay problem—even when the bath has a complicated structure. The key tool used to do this, is the concept of a *pseudomode*, a mode, or modes, which completely replaces the heatbath. The importance of exact approaches is of current interest because of the recent experimental realizations of quantum systems strongly interacting with resonators both in the context of cavity QED [2] and semiconductor microcavities [3].

We will consider a two-level system coupled to a bath of oscillators with a density of states described by a frequency dependent function $D(\omega)$. Then we know that if the function $D(\omega)$ has a pole close to the real ω axis, the short-time behavior of the two-level system is modified and leads to exponential decay. However, if there are two or more poles close to the real ω axis, it is not clear that a simple picture of exponential decay will apply because the poles can interfere with each other. In this paper we consider the class of functions D comprised of meromorphic functions, that is, smooth functions, without branch cuts, that can be expressed as ratios of polynomials of ω. The first example will show that if the residues of the meromorphic function D can change sign, we obtain a coupling between the decay channels which we would not find in the ordinary case of residues with the same sign. Of course, the chosen function $D(\omega)$ may well be an approximation to the real density-of-states function, but the important point is that it should be a good approximation in the region of interest. This also enables us to use the entire ω axis, an approximation which neglects threshold effects [4]. However, such threshold effects, due to the turn on of D at $\omega=0$ are negligible at optical frequencies and will only slightly modify the long-time behavior of the decay. Likewise, the realistic behavior of $D(\omega)$ as $\omega\to\infty$ [5] will deviate from the model function and lead to slight changes in the behavior as $t\to\infty$ and will be ignored here. Thus we focus on model, normalizable, meromorphic functions $D(\omega)$.

We consider first a simple model of a gap in the photon density of states where the function $D(\omega)$ is comprised of two Lorentzians, of which the second has a negative sign

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

(with $\Gamma_2<\Gamma_1$ to ensure positivity of D). The negative Lorentzian introduces a dip into the density of states which can inhibit spontaneous emission in the region of the dip [6,7] (at least in the weak-coupling limit where Fermi's golden rule is appropriate). The positive Lorentzian models a broad, resonant background structure. For consistency with previous works [8,9] we choose here to normalize $D(\omega)$ to 2π and this means that the two weights must satisfy $W_1-W_2=1$. For a perfect gap, where $D(\omega_c)=0$, we would also have $W_1/\Gamma_1=W_2/\Gamma_2$. The two poles in D are located at $\omega_c-i\Gamma_1/2$, $\omega_c-i\Gamma_2/2$, and we note that there is a change in sign of the residues of D between these poles. The analysis given in Eqs. (9)–(20) below shows the key result that the associated, *exact*, master equation is [9]

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

with the Hamiltonian

$$H_0=\omega_0(\hat{\sigma}_z+1)/2+\omega_c\hat{a}_1^\dagger\hat{a}_1+\omega_c\hat{a}_2^\dagger\hat{a}_2+V(\hat{a}_1^\dagger\hat{a}_2+\hat{a}_1\hat{a}_2^\dagger)$$
$$+\Omega_0(\hat{a}_2^\dagger\hat{\sigma}_-+\hat{a}_2\hat{\sigma}_+), \tag{3}$$

where the operators $\hat{\sigma}_\pm$ are the usual Pauli raising and lowering operators for the two-level system with the commutator $[\hat{\sigma}_+,\hat{\sigma}_-]=\sigma_z$. The operators $\hat{a}_1$, and $\hat{a}_2$ are the annihilation operators for two modes with decay rates

$$\Gamma_1'=W_1\Gamma_2-W_2\Gamma_1,$$

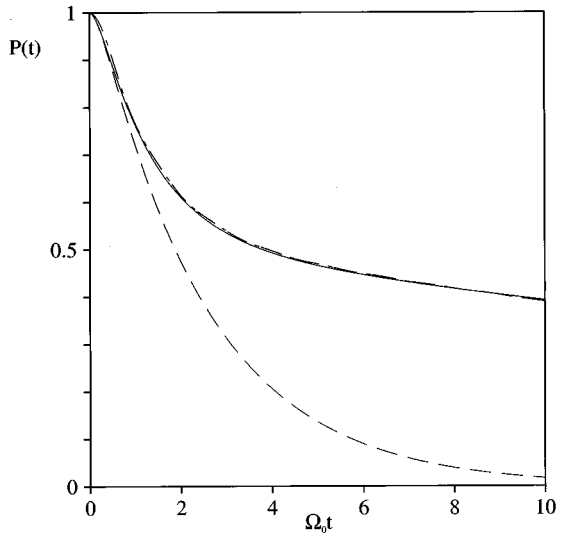

FIG. 1. The probability $P=|c_a(t)|^2$ of finding the excited atomic state in the the band-gap model (1). The solid curve shows the result from the density-matrix master equation (2). The nearly coincident chained curve shows the result from integrating the heat-bath equation (5) with just 30 modes equally spaced between $\pm 5\Omega_0$. The parameters used are: $\Gamma_1/\Omega_0=10$, $\Gamma_2/\Omega_0=1$, and $W_1=1.1$ ($W_2=0.1$). From these parameters we derive the fast decay rate $\Gamma_2'/\Omega_0=10.9$ and the slow rate $\Gamma_1'/\Omega_0=0.1$. The coupling between the two pseudomodes is then approximately 1.49. The dashed curve shows the decay in the absence of a gap ($W_2=0$).

$$\Gamma_2'=W_1\Gamma_1-W_2\Gamma_2. \tag{4}$$

The two modes are coupled by $V=\sqrt{W_1W_2}(\Gamma_1-\Gamma_2)/2$. The atom couples to the continuum with Ω_0, and the frequency of the two-level transition is ω_0. It is to be stressed that neither the Born nor Markov approximations have been made and that the only approximation is the chosen form of D in Eq. (1).

The master equation (2) is a two-mode version of the damped Jaynes-Cummings model [10] with the addition of a mode-mode coupling in the Hamiltonian. We will clearly see that if we remove the negative Lorentzian, $W_2\to 0$ ($W_1\to 1$), the master equation reduces to a single-mode damped Jaynes-Cummings model: the exact result for a Lorentzian resonance (without Born or Markov approximations). The two modes are clearly connected to the two Lorentzians in Eq. (1), but their decay rates are different from the widths of the Lorentzians. The mode-mode coupling is a direct result of the negative Lorentzian; it scales with $\sqrt{W_2}$ and disappears as the widths of the two Lorentzians become equal (resulting in a single positive Lorentzian). There would also be no mode-mode coupling if we considered two Lorentzians with *positive* weights instead of Eq. (1). In that case we would simply obtain a two-mode damped Jaynes-Cummings model with decay rates given *exactly* by the widths of the Lorentzians [9], in contradistinction with the decay rates in the master equation (2).

We can examine the decay dynamics of the band-gap system by a numerical integration of the master equation (2). An example is given in Fig. 1 where Γ_2 is much smaller than Γ_1. As a result the modes have very different decay rates. There is a fast decaying background mode (Γ_2'), and a slowly decaying band-gap mode (Γ_1'), and thus Fig. 1 shows a two-step decay process. The upper-state population first decays rapidly as a component of the initial population is lost to the background mode. This decay is not exponential because of the strong coupling. Thereafter, the state decays very slowly because of the slow band-gap mode decay rate. The dashed curve in the figure shows the fast decay in the absence of a density-of-states gap. Clearly the gap inhibits the decay of the system, even when the coupling strength Ω_0 is comparable to the width of the gap. If we had a zero in the density-of-states function $W_1/\Gamma_1=W_2/\Gamma_2$, the decay rate of one of the modes becomes zero ($\Gamma_2'=0$), and then we have some permanently trapped atomic population (if the two-level system is resonant with the gap).

We note that in the limit $\Gamma_1\to\infty$, the density of states $D(\omega)$ becomes very flat away from the gap. In this limit we will obtain results for the atomic dynamics that agree with the Laplace transform solutions for c_a given in Ref. [7]. Also, if the coupling Ω_0 becomes very weak, then the oscillatory exchange of energy with the resonance ceases and we obtain exponential decay consistent with the Wigner-Weisskopf approximation and Fermi's golden rule. In this regime the field modes can be adiabatically eliminated.

To test the master-equation interpretation we can also perform a numerical integration of the underlying heatbath model (in the rotating-wave approximation),

$$H=\sum_\lambda \omega_\lambda a_\lambda^\dagger a_\lambda+\omega_0(\hat\sigma_z+1)/2+\sum_\lambda g_\lambda(a_\lambda^\dagger\hat\sigma_-+a_\lambda\hat\sigma_+), \tag{5}$$

where g_λ is the frequency dependent coupling between the atomic transition and the reservoir modes with labels λ and frequency ω_λ. The relationship between the couplings g_λ and the normalized density-of-states function $D(\omega)$ is

$$\rho_\lambda(g_\lambda)^2=\Omega_0^2 D(\omega_\lambda)/(2\pi), \tag{6}$$

where ρ_λ is the density of reservoir modes. In this way the shape of the bath resonances are described by $D(\omega)$ and the strength of the coupling is described by Ω_0.

The time evolution of the underlying system can then be described by a set of amplitudes: c_a for the excited atomic state (and empty bath modes), c_λ for an excited bath mode (atom in ground state), c_0 for the atom in the ground state, with an empty bath. It is convenient to move to an interaction representation with the time-dependent transformations $\tilde c_a(t)=e^{i\omega_0 t}c_a(t)$, $\tilde c_\lambda(t)=e^{i\omega_\lambda t}c_\lambda(t)$, and then by utilizing the Schrödinger equation with the Hamiltonian Eq. (5) we obtain the following well-known coupled equations [1]:

$$i\frac{d}{dt}\tilde c_a=\sum_\lambda g_\lambda e^{-i\Delta_\lambda t}\tilde c_\lambda, \tag{7}$$

$$i\frac{d}{dt}\tilde c_\lambda=g_\lambda e^{i\Delta_\lambda t}\tilde c_a, \tag{8}$$

where the detuning of the atomic transition from the mode λ is $\Delta_\lambda=\omega_\lambda-\omega_0$. The amplitude c_0 is constant in time.

Then to simulate the heatbath with a large but finite number of discrete modes, we may numerically integrate Eqs. (7)

and (8) with the couplings g_λ determined from Eq. (6) and the chosen, normalized functon D. This type of approach has also been taken for explicitly discrete systems [11], but here the density of states should be sufficiently large to avoid recurrences. In Fig. 1 the result is shown for $D(\omega)$ given by Eq. (1) and with just 30 heatbath modes. There are slight deviations from the master-equation result, but as the number of heatbath modes is increased it is found that the differences become indiscernible.

We can formally establish the exact correspondence between the master equation (2) and a density-of-states function, such as Eq. (1), by examining the integro-differential equation associated with the continuum Eqs. (7) and (8). To obtain this we integrate Eq. (8) and substitute the result for $\tilde{c}_\lambda$ into Eq. (7), so that

$$\frac{d}{dt}\, c_a(t) = -i\omega_0 c_a(t) - \int_0^t dt' G(t-t') c_a(t'), \qquad (9)$$

where the difference kernel $G(t-t')$ can be placed in the form ($\tau = t-t'$)

$$G(\tau) = \frac{\Omega_0^2}{2\pi}\int_{-\infty}^{\infty} d\omega\, D(\omega) e^{-i\omega\tau} = -\frac{\Omega_0^2}{2\pi}\oint_C dz\, D(z) e^{-iz\tau}$$
$$= \Omega_1^2 e^{-iz_1\tau} + \Omega_2^2 e^{-iz_2\tau}, \qquad (10)$$

in the limit where the sum over λ becomes an integral. Crucial assumptions about the form of $D(\omega)$ have been made in Eq. (10). To convert the line integral to a contour integral it is assumed that the contribution from a large semicircle in the lower-half plane is negligible. The residue theorem has been used to evaluate the contour integral resulting in a sum of exponential terms typical of meromorphic functions with simple poles. Here two poles have been included (located at z_1 and z_2) in readiness for $D(\omega)$ given by Eq. (1), but the same result applies for other functions D and generalization to more poles is possible [9]. We return to the case of a double pole later in this paper. The values of Ω_1^2 and Ω_2^2 are determined by the two residues and thus the normalization of D will mean that $\Omega_1^2 + \Omega_2^2 = \Omega_0^2$.

Now we could try to solve Eq. (9) by Laplace transforms (exactly, or approximately) (see, e.g., [6,12,7]), but here we observe that the integro-differential equation (9) [with G given by Eq. (10)] also results from the elimination of b_1, b_2 from the differential equations

$$i\frac{d}{dt}\, c_a(t) = \omega_0 c_a(t) + \Omega_1 b_1(t) + \Omega_2 b_2(t), \qquad (11)$$

$$i\frac{d}{dt}\, b_1(t) = z_1 b_1(t) + \Omega_1 c_a(t), \qquad (12)$$

$$i\frac{d}{dt}\, b_2(t) = z_2 b_2(t) + \Omega_2 c_a(t). \qquad (13)$$

The advantage of these equations, as compared to Eqs. (7) and (8), is that there are now only *three* amplitudes (when D has two simple poles). The amplitudes b_1, b_2 behave similarly to amplitudes of real modes, and have thus been called the amplitudes of *pseudomodes* [8,9]. The concept is similar to the *faked continuum* introduced by Stenholm [13]. These amplitudes can be considered to be defined by Eqs. (12) and (13), i.e.,

$$b_l(t) = -i\Omega_l e^{-iz_l t}\int_0^t dt' e^{iz_l t'} c_a(t'). \qquad (14)$$

for $l = 1,2$. Now we will introduce parameters specific to the band-gap model (1). The poles are located at $z_1 = \omega_c - i\Gamma_1/2, z_2 = \omega_c - i\Gamma_2/2$ and from the residues of D we will find that $\Omega_1^2 = W_1\Omega_0^2$ and $\Omega_2^2 = -W_2\Omega_0^2$. We note that the second residue is negative, because of the negative weight of the Lorentzian, and as a result the coupling Ω_2 is *imaginary*. In itself, this does not affect the validity of Eqs. (11)–(13), but it does mean that we cannot construct a Lindblad master equation directly from these equations [9]. [In part, this is because Eqs. (11)–(13) do not contain the conjugates Ω_1^*, Ω_2^*.] To resolve this problem we employ an orthogonal transformation which forms the true pseudomode amplitudes a_1, a_2 from the amplitudes b_1, b_2

$$\begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \frac{1}{\sqrt{\cos 2\alpha}} \begin{bmatrix} \cos\alpha & i\,\sin\alpha \\ -i\,\sin\alpha & \cos\alpha \end{bmatrix} \begin{pmatrix} b_1 \\ b_2 \end{pmatrix}. \qquad (15)$$

This transformation applies to poles which lie on a line parallel to the imaginary ω axis, the more general case is treated in Ref. [9]. The angle α is complex, and when

$$\exp(4i\alpha) = \Omega_2^2/\Omega_1^2 = -W_2/W_1 \qquad (16)$$

we obtain the well behaved equations

$$i\frac{d}{dt}\, c_a = \omega_0 c_a + \Omega_0 a_2,$$

$$i\frac{d}{dt}\, a_1 = z_1' a_1 + V a_2,$$

$$i\frac{d}{dt}\, a_2 = z_2' a_2 + V a_1 + \Omega_0 c_a, \qquad (17)$$

where $z_l' = \omega_c - i\Gamma_l'/2$, $l = 1,2$, and Γ_l' and V have been defined above. These equations are well behaved because the parameter V and the coupling Ω_0 are real. To construct the master equation (2) we build an unnormalized state vector in the basis of the pseudomodes

$$|\tilde{\psi}(t)\rangle = c_0|0\rangle|0\rangle_1|0\rangle_2 + c_a(t)|1\rangle|0\rangle_1|0\rangle_2$$
$$+ a_1(t)|0\rangle|1\rangle_1|0\rangle_2 + a_2(t)|0\rangle|0\rangle_1|1\rangle_2. \qquad (18)$$

By using Eqs. (11)–(13) we can show that this state vector satisfies $d/dt\,|\tilde{\psi}(t)\rangle = -iH_{\rm eff}|\tilde{\psi}(t)\rangle$, where $H_{\rm eff} = H_0 - i(\Gamma_1'/2)\hat{a}_1^\dagger\hat{a}_1 - i(\Gamma_2'/2)\hat{a}_2^\dagger\hat{a}_2$ and $\hat{a}_1$ annihilates $|1\rangle_1$, etc. Then it is easily shown that the density matrix

$$\hat{\rho}(t) = \Pi_j(t)|\mathbf{0}\rangle\langle\mathbf{0}| + |\tilde{\psi}(t)\rangle\langle\tilde{\psi}(t)|, \qquad (19)$$

is in fact the solution to the master equation (2). Here $|\mathbf{0}\rangle$ is to be identified with the pseudomode system vacuum state $|0\rangle|0\rangle_1|0\rangle_2$, and the quantity $\Pi_j(t)$ satisfies

$$\frac{d}{dt}\,\Pi_j(t)=\Gamma_1'|a_1(t)|^2+\Gamma_2'|a_2(t)|^2. \tag{20}$$

Thus the term $\Pi_j(t)|\mathbf{0}\rangle\langle\mathbf{0}|$ in the density matrix (19) represents the effect of entanglement of the two-level system with the original bath modes. The ground-state population of the two-level system (after tracing out the field modes) is $\Pi_j(t)+|c_0|^2+|a_1(t)|^2+|a_2(t)|^2$ which naturally obeys the same time evolution as $|c_0|^2+\Sigma_\lambda|c_\lambda|^2$ in the original heat-bath model.

Now we return to the problem of a second-order pole, where we have a meromorphic function, but Eq. (10) is not valid. As an example, we suppose that instead of Eq. (1) we have the squared Lorentzian

$$D(\omega)=\frac{\Gamma^3/2}{[(\omega-\omega_c)^2+(\Gamma/2)^2]^2}, \tag{21}$$

for which we will find that

$$G(\tau)=\Omega_0^2(1+\Gamma\tau/2)e^{(i\omega_c-\Gamma/2)\tau}. \tag{22}$$

Then the first pseudomode may be defined as in Eq. (14), and for the second pseudomode, $b_2(t)=-i\Omega_0\int_0^t dt'(iz_l\tau)\exp(-iz_l\tau)c_a(t')$ with $\tau=t-t'$. If we now let $a_1=b_1+b_2$ and $a_2=-ib_2$ we will find that the two-level system and two pseudomode amplitudes obey the differential equations,

$$i\,\frac{d}{dt}\,c_a=\omega_0 c_a+\Omega_0 a_2,$$

$$i\,\frac{d}{dt}\,a_1=(\omega_c-i\Gamma)a_1+Va_2,$$

$$i\,\frac{d}{dt}\,a_2=\omega_c a_2+Va_1+\Omega_0 c_a, \tag{23}$$

where $V=\Gamma/2$. Note that the first pseudomode decays at twice the normal Lorentzian rate, whereas the second mode does not decay at all. The associated master equation is then found to be,

$$\frac{d}{dt}\,\hat\rho=-i[H_0,\hat\rho]-\Gamma(\hat a_1^\dagger\hat a_1\hat\rho-2\hat a_1\hat a_1^\dagger+\hat\rho\hat a_1^\dagger\hat a_1), \tag{24}$$

with the Hamiltonian (3) and V as given above. As before, this result follows without Born or Markov approximations.

In conclusion, we have seen that the dynamics of a two-state system coupled to a zero-temperature resonant bath can be represented by an appropriate form of the damped Jaynes-Cummings model in which the two-state system is coupled to a lossy pseudomode or modes. In the case where the bath resonance $D(\omega)$ has two nearby poles in the complex plane we have seen that it is possible for the pseudomodes to become coupled leading to phenomena such as population trapping. This is seen to be related to a change in sign between the residues of $D(\omega)$. Then by using the pseudomode basis we may derive master equations that are exact, for the given functions $D(\omega)$, i.e., determined without the use of the Born or Markov approximations. The formalism can be extended to cover many functions $D(\omega)$.

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