Two-level atom-field interaction: Exact master equations for non-Markovian dynamics, decoherence, and relaxation

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(Received 1 February 1999; published 18 August 2000)

We perform a first-principles derivation of the general master equation to study the non-Markovian dynamics of a two-level atom (2LA) interacting with an electromagnetic field (EMF). We use the influence functional method, which can incorporate the full back reaction of the field on the atom, while adopting Grassmannian variables for the 2LA and the coherent-state representation for the EMF. We find exact master equations for the cases of a free quantum field and a cavity field in the vacuum. In response to the search for mechanisms to preserve maximal coherence in quantum computations in ion trap prototypes, we apply these equations to analyze the decoherence of a 2LA in an EMF, and find that decoherence time is close to relaxation time. This is at variance with the claims by authors who studied the same system but used a different coupling model. We explain the source of difference and argue that, contrary to common belief, the EMF, when resonantly coupled to an atom, does not decohere it as efficiently as a bath does on a quantum Brownian particle. The master equations for non-Markovian dynamics derived here are expected to be useful for exploring new regimes of 2LAEMF interaction, which is becoming physically important experimentally.

PACS number(s): 42.50.Ct, 42.50.Lc, 03.65.Bz

I. INTRODUCTION

A two-level system (2LS) interacting with a quantum field —an electromagnetic field (EMF) in particular—has proven to be a very useful model for a wide range of problems from atomic-optical [1–7] and condensed matter [8,9] processes to quantum computation [10]. For the latter application stringent limits in maintaining the coherence of the the 2LS (called qubits) are required. This prompted us to revisit the theoretical structure of the 2LS model, paying special attention to its coherence properties. Treatment of spontaneous emission and relaxation are standard textbook topics, whereas decoherence and dissipation, especially in the context of quantum computation, are the focus of more recent investigations [11–18].

Because of the familiarity of the model (see Sec. II A) and its theoretical and practical values, we do not need to emphasize the general motivation, but can go right to the point about the aim and results of this paper. The description of this system generally comprises two parts: (i) spontaneous emission in the 2LS, and (ii) decoherence due to the interaction of 2LS with the EM field, treated as a bath. The first part allows little room for disagreement, as it can be obtained from elementary calculations. The second part on decoherence is more subtle.

Environment-induced decoherence [19] has been studied extensively in recent years, and it has been primarily based on models of quantum Brownian motion (QBM) [20–26] for the interaction of a simple harmonic oscillator (Brownian particle) with a harmonic-oscillator bath (HOB) at a finite temperature, leading to a reasonably good understanding of its characteristic features. Decoherence of a 2LS in an EM field has been studied by a number of authors, notably [11,12,16], and their dissipative and decoherent behavior are reported to be similar to that of a QBM in a harmonic-oscillator bath. The progression in three stages—quiescent, vacuum fluctuation-dominated, and thermal fluctuation-dominated, separated by the cutoff frequency and the thermal de Broglie frequency (wavelength)—are indeed characteristic of the QBM results [21,23–25,27,28].

Our findings, in contrast, are in stark disagreement with those reported in the literature. We work with the standard 2LSEMF model [1] and obtain an exact master equation for depicting non-Markovian dynamics. Solving this equation for the reduced density matrix of the 2LS shows that the decoherence rate is close to the relaxation rate. This is in first appearance rather couterintuitive, and different from all previous findings. Upon careful deliberation we realize that the ''intuition'' researchers (including us at the start) have acquired for dissipation and decoherence is based on the QBM model, which influenced the choice of model in the investigation of decoherence for a 2LS. However, we find that such a commonly invoked intuition for QBM in a HOB fails to apply to that of a two-level atom (2LA) interacting with an electromagnetic field with the commonly assumed type of resonance coupling in quantum optics.

A. Decoherence in QBM

Physically, when we say that decoherence of the system of a Brownian oscillator proceeds in a very short time as it is brought in contact with an environment, such as a HOB at some temperture, we are usually conjuring a model with bilinear [20] (or polynomial [24]) coupling of the oscillator-bath coordinates, and an Ohmic or sub-Ohmic spectral function [21] in the bath. Intuitively, the bath needs to have many degrees of freedom, preferably acting independently of each other so that the phase information in the system will be dispersed to the largest extent amongst the many bath degrees of freedom and afford little chance of taking an inordinately long time to be revived or reconstituted (recoher-

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ence [29]). The opposite picture (of very long decoherence time) is exemplified by two coupled subsystems where no coarse graining is introduced, or for system-environment couplings that maintain some high level of coherence, or for an environment whose degrees of freedom have long correlation times like in a zero-temperature, supra-Ohmic bath. The case of a (spin) particle or (plasma) wave interacting with an averaged (collective) variable from the environment, such as the mean field, showing Landau damping in Vlasov dynamics, is another example [30,31]. Just as in the spin-echo phenomena (e.g, Chap. 3 [4]), the basic physics in this case is not dissipation in the Boltzmann sense, but statistical mixing [32]. We will see that this example is of more physical relevance to our problem than the QBM.

B. Coherence in the 2LS

For the 2LAEMF system, one clear distinction between an EM field as an environment and a system of harmonic oscillators as bath is that the field (coupled to a detector) has an intrinsic spectral density function, not to be chosen arbitrarily. For example, it has been shown [26] that a conformal scalar field in two dimensions coupled to a monopole detector has an Ohmic character while in four dimensions it is supra-Ohmic. Barone and Caldeira [33] showed that the spectral density function for EM fields with momentum coupling to an oscillator detector is supra-Ohmic. These density functions would show very different decoherence behavior from the high-temperature Ohmic HOB used in many discussions of decoherence, the latter case is what the general folklore is based on. But the most important distinction from QBM is that the 2LA couples with the EMF in the discrete number basis for the field, unlike the continuous amplitude basis in the QBM. This fact (which is true in the rotating-wave and dipole approximation) implies that the 2LS plus the EMF system is a *resonant* one. Hence even though the EM field has just as many (large number of) modes as the HOB, only a very small fraction of them in a narrow range of the resonance frequency are efficiently coupled to the atom. This is the root cause for the very different qualitative behavior between the QBM and the 2LS as far as decoherence is concerned.

One extreme case is that of a single-mode field described by the Jaynes-Cummings model, where Rabi nutation takes place and the atom field remains largely a coherent system. (For a coherent field, the probability for the atom to be found in the excited state at time t regardless of the state of the field obeys a Poisson distribution. This distribution in the photon number induces a spread in the Rabi frequencies, and causes the collapse and revival of the Rabi nutation. These are distinct features of quantum coherence [1].) Adding all modes to the field we see spontaneous emission and the decay of the atom. The probability of an initially excited atom (remaining in the excited state) decays exponentially in the Wigner-Weisskopf form (characteristic of Markovian processes) with relaxation time constant Γ. For purely radiative decay the decay time T_1 of the inversion is half the decay time T_2 of the polarization. There are no large order-of-magnitude differences between dissipation and decoherence time (which in typical QBM high-temperature conditions could be as high as 40 [19]). In fact, it is perhaps inappropriate to talk about dissipation for a 2LAEMF system because the conditions for a bath to actuate such a process are lacking. The transition from the excited to the ground state is closer in nature to relaxation (in the spin-echo sense) than dissipation. In a cavity where excitation of the atom from the field (absorption) balances with emission, it is more appropriate to refer to the resonant state of the atom field as a coherent system. In these senarios, the distinction between QBM and 2LA cannot be clearer.

C. Difference between QBM and 2LS

So what led earlier authors to make the qualitative claim that 2LS decoheres easily? We think the confusion arises when the picture of QBM dissipation and decoherence is grafted on the 2LAEMF system indiscriminantly. If the field that acts as the environment is a phonon field (from ion vibrations, see, e.g., [17]) and if the coupling is of the nonresonant type, then there is no disagreement. Decoherence should follow the QBM pattern as reported by many authors.[1] Such sources (including atomic collisions in a cavity [7]) can be important for some setups. However, when one claims that the EM field can decohere a 2LS (with which it is coupled in a resonant way, as in the standard model)—that is where we disagree.

Quantitatively, the model for the 2LS used by most authors for the discussion of decoherence inspired by QBM type of behavior has the atom in a σ_z state (the diagonal Pauli matrix) coupled to the field mode operators $\hat{b}^\dagger, \hat{b}$. This type of coupling term (call it σ_z type for convenience) commutes with the Hamiltonian of the system, and admits a diagonalization in the eigenbasis of the Hamiltonian. The field is coupled to the atom as a whole and thus is insensitive to the two-level transition activity. In particular, it does not probe the resonance or coherent properties of the two-level atom, which is the most important feature, for quantum computation. In contrast, the standard model for 2LAEMF, which we studied, has a $\sigma_\pm$ coupling (call it standard coupling) to the field modes that highlights the two-level activity of the atom and the field. This coupling considered in the standard model is indispensible, i.e., it *cannot* be removed from the two-level atom as it *defines* it and will be present in any realistic situation. What then is the origin of the QBM type of contribution to the 2LS ?

If one accepts an environment other than the EM field, the question comes down to the characteristics of the experimental apparatus. For well-prepared ion traps we would expect it to be rather unimportant. If the EM field is the only field

[1]Even in such cases, one also needs to pay closer attention to the QBM behavior than what has been accorded for this model. Subtle points unnoticed before include, e.g., the imposition of a high-frequency cutoff and Ohmic spectral function that restricts to a Markovian behavior [21] can lead to a violation of the positivity of the reduced density matrix [24], the violation of the fluctuation-dissipation relation [23], and the prolongation of coherence in a low-temperature supra-Ohmic bath [24]. They deserve more attention in the theoretical design of cavity qubit computers.

present, we can still ask if a QBM type of coupling term with the EM field would appear, and if yes, how strong would its effect be? This would be a useful way to accommodate the two different types of coupling terms.

Recall that the standard model is derived under the dipole and rotating-wave approximation. In Sec. II we will show that the σ_z type of coupling appears only in the next-order expansion after the dipole approximation. Since these are good approximations for a large class of atomic states when the atom is nonrelativistic, the contribution from the QBM type of coupling used in [11,12,16] should be negligible and its ensuing decoherent effect insignificant. In this sense the EM field does not in leading order of approximation act like a bath in the QBM way, and coherence in a 2LAEMF system is quite well preserved (excepting other processes, e.g., [15,14]).

Our puzzle over the result on decoherence in the 2LS reported in the literature was what prompted us to begin this study. Without letting any familiar and convenient analogy influence our judgement, and without any preconceived notion, we choose to perform a first-principles calculation of the two-level atom electromagnetic field (2LA-EMF) system making as few assumptions and covering as wide a range of conditions as possible. We use the influence functional method [20] to take into account the full back reaction of the field on the atom, while adopting Grassmannian variables for the 2LA and the coherent-state representation for the EMF. We find exact master equations for the full (non-Markovian) dynamics in the cases of a free quantum field and a cavity field at zero temperature.

In Sec. II we present the model and the formalism. A detailed derivation of our model is contained in Appendix A. In Sec. III we derive the master equations. In Sec. IV we study different mode composition of the field, including that of an atom in a cavity. We end in Sec. V with a discussion of our findings and their implications.

This is the first in a series of papers on the 2LA and quantum decoherence. The subsequent papers will treat 2LAEMF interaction at finite temperatures, for EM fields in a coherent and squeezed state, and for multipolar models (where coupling other than the minimal is assumed). We will also tend to collective qubit systems and moving atoms interacting with an EM field. These results will have corresponding applications in atom optics and quantum computation problems.

II. THE INFLUENCE FUNCTIONAL

A. The model

Our model for atom-field interaction is the standard one (see Appendix A for details) [1,2,4].[2] The total Hamiltonian for a (stationary) atom interacting with a quantum electromagnetic field (EMF) under the dipole, rotating-wave (RW), and two-level (2L) approximation is given by

$$\hat{H}=\hbar\omega_0\hat{S}_z+\hbar\sum_{\mathbf{k}}\left[\omega_{\mathbf{k}}\hat{b}_{\mathbf{k}}^{\dagger}\hat{b}_{\mathbf{k}}+(g_{\mathbf{k}}S_{+}\hat{b}_{\mathbf{k}}+\overline{g_{\mathbf{k}}}S_{-}\hat{b}_{\mathbf{k}}^{\dagger})\right], \tag{2.1}$$

where $\hat{b}_{\mathbf{k}}^{\dagger},\hat{b}_{\mathbf{k}}$ are the creation and annihilation operators for the kth normal mode with frequency $\omega_{\mathbf{k}}$ of the electromagnetic field (thus for the field vacuum $\hat{b}_{\mathbf{k}}|0\rangle=0,[\hat{b}_{\mathbf{k}},\hat{b}_{\mathbf{k}'}^{\dagger}]=\delta_{\mathbf{k},\mathbf{k}'}$, for all $\mathbf{k}$), and $\omega_0=\omega_{21}$ is the frequency between the two levels. Here

$$\hat{S}_z=\frac{1}{2}\hat{\sigma}_z,\quad \hat{S}_{\pm}=\hat{\sigma}_{\pm}\equiv\frac{1}{2}(\hat{\sigma}_x\pm i\hat{\sigma}_y),$$

where $\sigma_{x,y,z}$ are the standard 2×2 Pauli matrices with $\sigma_z=\mathrm{diag}\,(1,-1)$, etc. The coupling constant $g_{\mathbf{k}}\equiv d_{21\mathbf{k}}f_{\mathbf{k}}(\mathbf{X})$ where

$$d_{ij\mathbf{k}}\equiv-\frac{i\omega_{ij}}{\sqrt{2\hbar\omega_{\mathbf{k}}\epsilon_0 V}}\mathbf{d}_{ij}\cdot\hat{\mathbf{e}}_{\mathbf{k}\sigma} \tag{2.2}$$

and $\mathbf{d}_{ij}\equiv e\int\overline{\phi_i}\mathbf{x}\phi_j d^3x$ is the dipole matrix element between the eigenfunctions ϕ_i of the electron-field system, $\hat{\mathbf{e}}_{\mathbf{k}\sigma}$ is the unit polarization vector ($\sigma=1,2$ are the two polarizations), and $f_{\mathbf{k}}(\mathbf{x})$ are the spatial mode functions of the vector potential of the electromagnetic field (in free space, $f_{\mathbf{k}}(\mathbf{x})=e^{-i\mathbf{k}\cdot\mathbf{x}}$, V is the volume of space). Under the dipole approximation, $f_{\mathbf{k}}$ is evaluated at the position of the atom $\mathbf{X}$. Since $\mathbf{d}_{ij}=\overline{\mathbf{d}}_{ji}$, $\overline{d}_{ij\mathbf{k}}=d_{ji\mathbf{k}}$, we will choose a mode function representation such that $g_{\mathbf{k}}$ is real.

To see how this could possibly be related to the σ_z type of coupling with Hamiltonian (used by, e.g., [12,16] for the study of decoherence in 2LS)

$$\hat{H}=\hbar\omega_0\hat{S}_z+\hbar\sum_{\mathbf{k}}\left[\omega_{\mathbf{k}}\hat{b}_{\mathbf{k}}^{\dagger}\hat{b}_{\mathbf{k}}+\hbar\sigma_z(\overline{g_{\mathbf{k}}}\hat{b}_{\mathbf{k}}+g_{\mathbf{k}}\hat{b}_{\mathbf{k}}^{\dagger})\right], \tag{2.3}$$

we examine the next term after the dipole approximation in Eq. (A15). This has a contribution to $g_{ij\mathbf{k}}$ even when $i=j$, which is equal to

$$g_{ii\mathbf{k}}=c_{\mathbf{k}}\mathbf{k}\cdot\mathbf{q}_i,$$

where

$$\mathbf{q}_i=\sum_{\sigma}\int\overline{\phi_i}\delta\mathbf{x}(\mathbf{p}\cdot\hat{\mathbf{e}}_{\mathbf{k}\sigma})\phi_i dx^3$$

and c_k is a constant given by

$$c_{\mathbf{k}}=-\frac{e}{m}(2\hbar\omega_{\mathbf{k}}\epsilon_0 V)^{-1/2}.$$

This generates an additional coupling term

[2]Our Hamiltonian is given in the so-called minimal coupling (MC) as different from the multipolar coupling (MP) [6], which may be more relevant to atoms in a cavity because the explicit Coulomb interaction between the atom and its image charge is removed.

$$\sum_{\mathbf{k}} \sigma_z(g_{1\mathbf{k}}b_{\mathbf{k}}+\overline{g}_{1\mathbf{k}}b_{\mathbf{k}}^{\dagger})+1(g_{2\mathbf{k}}b_{\mathbf{k}}+\overline{g}_{2\mathbf{k}}b_{\mathbf{k}}^{\dagger}),$$

where

$$g_{1\mathbf{k}}=g_{11\mathbf{k}}-g_{22\mathbf{k}},\quad g_{2\mathbf{k}}=g_{11\mathbf{k}}+g_{22\mathbf{k}}.$$

This gives the lowest order σ_z type of coupling in a 2LAEMF system. The ratio of the coupling $g_{1\mathbf{k}}$ of the σ_z type in Eq. (2.3) to the dipole coupling $g_{\mathbf{k}}$ in Eq. (2.1) is

$$|g_{1\mathbf{k}}/g_{\mathbf{k}}|=\left|\frac{\mathbf{k}(\mathbf{q}_1-\mathbf{q}_2)}{m\omega_{\mathbf{k}}d_{12}}\right|\leqslant\frac{\omega_{\mathbf{k}}|\mathbf{q}_1-\mathbf{q}_2|}{m\omega_{\mathbf{k}}d_{12}}. \tag{2.4}$$

Thus the σ_z type of coupling generated from the 2LAEMF interaction will be significant only for very high frequencies ω_k of the EM field, a point intuitively clear from the meaning of the dipole approximation.

B. Grassmannian variables and coherent-state integrals

Since Feynman and Vernon [20] invented the influence functional method, this formalism has been applied to treat the Brownian motion of a harmonic-oscillator interacting with a harmonic oscillator bath by many authors [21,9,22,24]. The two-level system in tunneling has been discussed in detail by Leggett *et al.* [8], but the derivation of a master equation by this method that can traverse the non-Markovian regimes has not yet been carried out. We shall perform such a calculation for a two-level system, with the aid of Grassmannian variables convenient for treating fermions and the coherent-state representation in a path integral form. We construct the coherent state of the combined atom-field system as

$$|\{z\},\eta\rangle=|\{z\}\rangle\times|\eta\rangle, \tag{2.5}$$

where $|z\rangle$, z a complex number, denotes the EM field coherent states and $|\eta,\ \eta$ a Grassmannian or anticommuting number, denotes the electron coherent state. The transition amplitude between the initial state (i) at $t=0$ and the final state (f) at $t=t_f$ is expressed formally as [34] (here we suppress the index k)

$$\langle\overline{\eta}_f,\overline{z}_f;t|\eta_i,z_i;0\rangle=\int DzD\overline{z}D\eta D\overline{\eta}e^{(i/\hbar)S[z,\overline{z},\eta,\overline{\eta}]}, \tag{2.6}$$

where the action is

$$\frac{i}{\hbar}S[z,\overline{z},\eta,\overline{\eta}]=\overline{z}z(t)+\overline{\eta}\eta(t)-\int_0^t ds\left[\dot{\overline{z}}z+\dot{\overline{\eta}}\eta+\frac{i}{\hbar}H(\overline{\eta},\eta,\overline{z},z)\right]. \tag{2.7}$$

Here H is the Q symbol of the Hamiltonian [35] and there is an implied summation over field modes:

$$H(\overline{\eta},\eta,\overline{z},z)=\hbar\left(\sum_k \omega_k\overline{z}_kz_k-g_k(\overline{z}_k\eta+\overline{\eta}z_k)+\omega_0\overline{\eta}\eta\right). \tag{2.8}$$

In Eq. (2.8) we have subtracted a constant term $\frac{1}{2}\omega_0 1$ to Eq. (2.1) so that the ground state now has zero energy. Henceforth we set $\hbar=1$.

The Hamiltonian in Eq. (2.8) is not a c-number function; it has terms that are odd. One might then question the validity of Eq. (2.6) for the path integral; it clearly exists as a formal expression, but its evaluation with a saddle-point method that is based on the Hamiltonian of Eq. (2.8) might be problematic. We dispel this doubt for the vacuum case with an operator method proof of the master equation in Appendix B. It shows that at least for the vacuum case that saddle-point evaluation yields the correct result. The general cases need separate considerations. For many qubits coupled to the EM field vacuum, we believe that the path-integral method yields a simpler treatment than the operator method.

The integration is over all paths satisfying

$$z(0)=z_i,\quad \overline{z}(t)=\overline{z}_f, \tag{2.9}$$

$$\eta(0)=\eta_i,\quad \overline{\eta}(t)=\overline{\eta}_f. \tag{2.10}$$

We assume initially that the density matrix of the total system plus the environment is factorizable $\hat{\rho}(0)=\hat{\rho}_e(0)\otimes\hat{\rho}_b(0)$. Only at that time would z and η be pure complex and Grassmannian numbers, respectively. As the system evolves, both η and z contain Grassmann and c-number parts. The mixing of even and odd parts (note g_k is odd) comes about as the initially factorized atom state becomes ''dressed.''

In the open system philosophy, as we are interested in the averaged effect of the field on the atom, the atom is considered the ''system'' while the field is the ''environment.'' The path integral is performed over the variables z, while $\eta,\overline{\eta}$ are treated as external sources. When only one field mode is considered, we have

$$\langle\overline{z}_f;t|z_i;0\rangle_{\eta,\overline{\eta}}=\int DzD\overline{z}\exp\left\{\overline{z}z(t)-\int_0^t ds[\dot{\overline{z}}z+i\omega\overline{z}z-g(\overline{z}\eta+\overline{\eta}z)(s)]\right\} \tag{2.11}$$

with summation over paths satisfying the boundary condition (29) for z. We use the saddle-point method. Minimizing the action yields the following equations:

$$\dot{z}+i\omega z=-ig\eta, \tag{2.12}$$

$$\dot{\overline{z}}-i\omega\overline{z}=ig\overline{\eta}, \tag{2.13}$$

with solutions

$$z(s)=z_ie^{-i\omega s}-ig\int_0^s ds'e^{-i\omega|s-s'|}\eta(s'), \tag{2.14}$$

$$\bar{z}(s)=\bar{z}_f e^{-i\omega(t-s)}+ig\int_s^t ds' e^{-i\omega|s-s'|}\bar{\eta}(s'). \quad (2.15)$$

Using these for the transition amplitude (2.11) with the minimum value for the action, we obtain

$$\langle \bar{z}_f;t|z_i;0\rangle_{\eta,\bar{\eta}}=\exp\left\{\bar{z}_f z_i e^{-i\omega t}-ig\left[\bar{z}_f\int_0^t ds e^{-i\omega(t-s)}\eta(s)+\int_0^s ds e^{-i\omega s}\bar{\eta}(s)z_i\right]-g^2\int_0^t ds\int_0^s ds' e^{-i\omega|s-s'|}\bar{\eta}(s)\,\eta(s')\right\}. \quad (2.16)$$

A prefactor in the coherent-state path integral is equal to one. Now the influence functional due to this single mode reads

$$\mathcal{F}[\eta,\bar{\eta};\eta',\bar{\eta}']=\int\frac{d\bar{z}_i dz_i}{\pi}\frac{d\overline{z'_i}dz'_i}{\pi}\frac{d\bar{z}_f dz_f}{\pi}\times e^{-\bar{z}_i z_i-\bar{z}'_i z'_i-\bar{z}_f z_f}\langle\bar{z}_f;t|z_i;0\rangle_{\eta,\bar{\eta}}\langle\bar{z}_i|\rho_0|z'_i\rangle\times\langle\bar{z}'_i;0|z_f;t\rangle_{\eta,\bar{\eta}}, \quad (2.17)$$

where the completeness relation for (unnormalized) coherent states has been used:

$$\int\frac{d\bar{z}_i dz_i}{\pi}e^{-\bar{z}z}|z\rangle\langle\bar{z}|=1. \quad (2.18)$$

Writing with an obvious identification

$$\langle\bar{z}_f;t|z_i;0\rangle_{\eta,\bar{\eta}}=\exp(A\bar{z}_f z_i+i\bar{z}_f\beta+i\bar{\gamma}z_i+D), \quad (2.19)$$

we can use the identity

$$\int\frac{d\bar{z}_i dz_i}{\pi}e^{-\bar{z}z+\bar{f}z+\bar{z}f}=e^{\bar{f}f} \quad (2.20)$$

to obtain

$$\mathcal{F}[\bar{\eta},\eta,\bar{\eta}',\eta']=e^{\bar{\beta}'\beta-(D+D')} \quad (2.21)$$

for an initial vacuum state $\hat{\rho}_0=|0\rangle\langle 0|$. Substituting, we get the contribution to the influence functional from one mode

$$\mathcal{F}_k[\bar{\eta},\eta,\bar{\eta}',\eta']=\exp\left\{g_k^2\int_0^t ds\int_0^s ds'[\bar{\eta}'(s)\eta(s')e^{-i\omega_k(s-s')}+\bar{\eta}'(s')\eta(s)e^{i\omega_k(s-s')}-\bar{\eta}(s)\eta(s')e^{-i\omega_k(s-s')}-\bar{\eta}'(s')\eta(s)e^{i\omega_k(s-s')}]\right\}. \quad (2.22)$$

The influence functional for all modes $\mathcal{F}=\Pi_k\mathcal{F}_k$ is finally given by

$$\mathcal{F}[\bar{\eta},\eta,\bar{\eta}',\eta']=\exp\left\{\int_0^t ds\int_0^s ds'(\mu(s-s')\times[\bar{\eta}'(s)+\bar{\eta}(s)]\eta(s')+\mu^*(s-s')\times\bar{\eta}'(s')[\eta(s)+\eta'(s)])\right\} \quad (2.23)$$

in terms of the kernel

$$\mu(s)=\sum_k g_k^2 e^{-i\omega_k s}. \quad (2.24)$$

III. THE MASTER EQUATION

A. The reduced density-matrix propagator

Having computed the influence functional we have an expression for the reduced density-matrix propagator

$$J(\bar{\eta}_f\eta'_f;t|\bar{\eta}'_i\eta_i;0)=\int D\bar{\eta}D\eta D\overline{\eta'D}\,\eta'\exp\left\{\bar{\eta}\eta(t)+\bar{\eta}'\eta'(t)-\int_0^t ds[\bar{\eta}\dot{\eta}+\bar{\eta}'\dot{\eta}'+i\omega\bar{\eta}\eta-i\omega\bar{\eta}'\eta'(s)]+\int_0^t ds\int_0^s ds'(\mu(s-s')[\bar{\eta}'(s)+\bar{\eta}(s)]\eta(s')+\mu^*(s-s')\bar{\eta}'(s')[\eta(s)+\eta'(s)])\right\} \quad (3.1)$$

where the summation over all paths obey the boundary conditions (2.10) and

$$\bar{\eta}'(0)=\bar{\eta}'_i,\quad \eta'(t)=\eta'_f. \quad (3.2)$$

We can compute the path integral with saddle-point evaluation and get

$$\dot{\eta}+i\omega\eta+\int_0^s ds'\mu(s-s')\eta(s')=0, \quad (3.3)$$

$$\dot{\bar{\eta}}'-i\omega\bar{\eta}'+\int_0^s ds'\mu^*(s-s')\bar{\eta}'(s')=0, \quad (3.4)$$

$$\dot{\eta}'+i\omega\eta'+\int_0^s ds'\mu(s-s')\eta(s')-\int_s^t ds'\mu^*(s-s')[\eta(s')+\eta'(s')]=0, \quad (3.5)$$

$$\dot{\bar{\eta}} - i\omega\bar{\eta} + \int_0^s ds'\,\mu^*(s-s')\,\bar{\eta}'(s') - \int_s^t ds'\,\mu(s-s')[\bar{\eta}'(s') + \bar{\eta}(s')] = 0. \tag{3.6}$$

It will turn out that only the solution of the first two of these equations will contribute to the path integral. We will therefore write

$$\eta(s) = \eta_i u(s), \quad \bar{\eta}'(s) = \bar{\eta}'_f \bar{u}(s), \tag{3.7}$$

where $u,\bar{u}$ are the solutions to Eqs. (3.3) and (3.4) under the condition

$$u(0) = \bar{u}(t) = 1. \tag{3.8}$$

Now Eq. (3.3) is a linear integrodifferential equation of first order and as such can be solved with the use of the Laplace transform and the convolution theorem. It is easy to show that

$$u(s) = \mathcal{L}^{-1}\left(\frac{1}{z + i\omega + \tilde{\mu}(z)}\right) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} \frac{dz e^{zs}}{z + i\omega + \tilde{\mu}(z)}, \tag{3.9}$$

where $\tilde{\mu}(z)$ is the Laplace transform of the kernel (2.24) and c is a real constant larger than the real part of the poles of the integrand. It turns out that this function $u(s)$ contains all necessary information for the computation of the density matrix propagator. Substituting our expressions (3.8) and (3.9) in Eq. (3.1), we can obtain the following expression for the propagator:

$$J(\bar{\eta}_f \eta'_f; t|\bar{\eta}'_i \eta_i; 0) = \exp(\bar{\eta}_f \eta_i u(t) + \bar{\eta}'_i \eta'_f \bar{u}(t) - [1 - \bar{u}(t)u(t)]\bar{\eta}'_i \eta_i). \tag{3.10}$$

Since we are using coherent-state path integrals, we have departed in our evaluation from the standard saddle-point approximation used on configuration space path integrals. In these cases, the standard procedure is to distinguish the imaginary part of the kernel as corresponding to dissipation and consider only its contribution when performing the saddle-point evaluation. The resulting equations are then the classical dissipative equations of motion. But in the case of the coherent-state path integral, there is no correspondence between extrema of the action and actual classical paths. Hence there is no sense in splitting the kernel η into real and imaginary parts, and the saddle-point evaluation should be carried out for the whole of the exponential.

B. Master equation for a field in a vacuum state

It is a standard procedure now to find the master equation [21,24]. We compute the time derivative of the propagator:

$$\dot{J} = \left(\dot{u}\,\bar{\eta}_f \eta_i + \dot{\bar{u}}\,\bar{\eta}'_i \eta_f + \frac{d(\bar{u}u)}{dt}\bar{\eta}'_i \eta_i\right) J. \tag{3.11}$$

The next step is to remove from the above equation the dependence on the initial values. This is done with the use of the following identities:

$$\eta_i J = \frac{1}{u}\frac{\delta J}{\delta \bar{\eta}_f}, \quad \bar{\eta}'_i J = \frac{1}{\bar{u}}\frac{\delta J}{\delta \eta'_f}. \tag{3.12}$$

Note that we are suppressing (for ease of notation) symbols denoting left or right Grassmann differentiation. In all our expressions we implicitly assume that differentiation with respect to η is always right and with respect to $\bar{\eta}$ always left.

For the density matrix at time t,

$$\rho_t(\bar{\eta}_f, \eta'_f) = \int d\bar{\eta}_i d\eta_i e^{-\bar{\eta}_i \eta_i} d\bar{\eta}'_i d\eta'_i \times e^{-\bar{\eta}'_i \eta'_i} J(\bar{\eta}_f \eta'_f; t|\bar{\eta}'_i \eta_i; 0)\rho_0(\bar{\eta}_i, \eta'_i), \tag{3.13}$$

we obtain the evolution equation

$$\frac{\partial}{\partial t}\rho = \frac{\dot{u}}{u}\bar{\eta}\frac{\delta\rho}{\delta\bar{\eta}} + \frac{\dot{\bar{u}}}{\bar{u}}\frac{\delta\rho}{\delta\eta}\eta + \frac{(d/dt)(\bar{u}u)}{\bar{u}u}\frac{\delta^2\rho}{\delta\eta\delta\bar{\eta}}. \tag{3.14}$$

This is one of our main results: the master equation for the two-level atom interacting with an environment of an electromagnetic field at its vacuum state. The effect of the field is contained within the function u, which can be determined by the solution of Eq. (3.3) or equivalently by the computation of the contour integral (3.9). In Sec. IV we are going to find explicit expressions for u for particular choices of the field configuration.

Let us return for the moment to Eq. (3.14) and write this in an operator language. It is easy to verify that

$$\bar{\eta}\frac{\delta\rho}{\delta\bar{\eta}} = S_+ S_- \rho, \quad \frac{\delta\rho}{\delta\eta}\eta = \rho S_+ S_-, \quad \frac{\delta^2\rho}{\delta\eta\delta\bar{\eta}} = S_+ \rho S_-, \tag{3.15}$$

If we write

$$\frac{\dot{u}(t)}{u(t)} = \Gamma(t) + i\Omega(t), \tag{3.16}$$

the master equation reads

$$\frac{\partial}{\partial t}\rho = -i[H(t), \rho] + \Gamma(t)\{S_+ S_-, \rho\} - 2\Gamma(t) S_- \rho S_+, \tag{3.17}$$

where

$$H(t) = \Omega(t) S_+ S_-. \tag{3.18}$$

The first term corresponds to the unitary Hamiltonian evolution, only now the effect of the environment has induced a

time-dependent shift in the value of the frequency, the second term is time-dependent dissipation, and the third corresponds to noise.

C. Spontaneous emission

To show how the standard results are regained, and to understand the meaning of the new function in the master equation, let us consider the physical process of spontaneous emission. Start with a generic initial density matrix

$$\rho=\begin{pmatrix} 1-x & y \\ y^* & x \end{pmatrix}. \tag{3.19}$$

Its corresponding Q symbol is

$$\rho(\overline{\eta},\eta)=x+y^*\eta+y\overline{\eta}+(1-x)\overline{\eta}\eta. \tag{3.20}$$

If we evolve it with the density-matrix propagator (3.10) we obtain for the state at time t

$$\rho_t(\overline{\eta},\eta)=1-\overline{u}u(1-x)+(\overline{u}y^*\eta)+(u\overline{\eta}y)+(\overline{u}u(1-x))\overline{\eta}\eta \tag{3.21}$$

corresponding to

$$\rho_t=\begin{pmatrix} \overline{u}u(1-x) & uy \\ \overline{u}y^* & 1-\overline{u}u(1-x) \end{pmatrix}. \tag{3.22}$$

Considering the case $x=y=0$ we get for the probability of spontaneous emission

$$P(1\to 0,t)=1-\overline{u}u. \tag{3.23}$$

Also, we should remark that the rate of decoherence in the energy eigenstates is governed by the absolute value of the function u (the off-diagonal terms). But on the other hand u itself determines the rate of energy flow from the atom to the environment. Hence for our particular choice of initial state (vacuum) we find that decoherence and relaxation time are essentially identical. We shall use this equation to study decoherence in an ion trap in a later paper.

IV. FIELD MODES AND ANALYTIC $u(t)$

Our master equation (3.17) depends solely on the function $u(t)$, which in its turn is determined by the kernel $\mu(s)$. In this section we will try to give some analytic expressions for this function in various different cases.

A. A single mode

To connect with known results [1], let us start with the case where the field contains only a single mode with frequency $\omega_k=k$. Then $\mu(s)$ will read

$$\mu(s)=g^2e^{-iks} \tag{4.1}$$

and

$$\tilde{\mu}(z)=-g\int_0^\infty e^{-sz}e^{-iks}=\frac{g^2}{z+ik}. \tag{4.2}$$

The integrand has two poles at the solutions of the equation

$$z^2+i(\omega+k)z-\omega k+g^2=0 \tag{4.3}$$

given by

$$z=-i\frac{\omega+k\pm[(\omega-k)^2+g^2]^{1/2}}{2}=-i\omega_{1,2}. \tag{4.4}$$

Hence

$$u(s)=\frac{k-\omega_1}{\omega_2-\omega_1}e^{-i\omega_1 s}-\frac{k-\omega_2}{\omega_2-\omega_1}e^{-i\omega_2 s}. \tag{4.5}$$

This result is in agreement with standard ones [1].

B. Infinite number of modes

Now we consider the case of the vacuum electromagnetic field in free space, i.e., not constrained by a cavity. The kernel will then read [using Eq. (2.24)]

$$\mu(s)=2\lambda^2\int\frac{d^3k}{(2\pi)^3}k^{-1}e^{-iks}=\frac{\lambda^2}{\pi^2}\int_0^\infty kdke^{-iks}=\frac{d}{ds}\nu(s), \tag{4.6}$$

where

$$\nu(s)=\frac{i\lambda^2}{\pi^2}\int_0^\infty dke^{-iks}. \tag{4.7}$$

Note the factor of 2 in Eq. (4.6) coming from the two-photon polarizations and that in view of Eq. (2.3) we have written $g_{\mathbf{k}}=\lambda\omega_{\mathbf{k}}^{-1/2}$.

Since the integral (4.7) is not convergent, we will introduce an exponential cutoff in the higher frequency modes. The presence of the cutoff is of physical significance since we do not expect high electromagnetic modes to couple with our two-level atom.

Hence the kernel ν will read

$$\nu(s)=\frac{i\lambda^2}{\pi^2}\int_0^\infty dke^{-iks-k\epsilon}=\frac{\lambda^2}{\pi^2}\frac{1}{s-i\epsilon}. \tag{4.8}$$

The Laplace transform of ν is then

$$\tilde{\nu}(z)=\frac{\lambda^2}{\pi^2}\int_0^\infty ds\frac{e^{-sz}}{s-i\epsilon}=-\frac{\lambda^2}{\pi^2}e^{-i\epsilon z}\mathrm{Ei}(-i\epsilon z) \tag{4.9}$$

where Ei denotes the exponential integral function analytically continued to the complex domain. At the limit $\epsilon\to 0$ this is essentially

$$\mathrm{Ei}(-i\epsilon z)=\gamma+\ln(-i\epsilon z)+O(\epsilon), \tag{4.10}$$

where γ is the Euler-Macheronni constant and the logarithm is taking values in the primary branch. Thus $\tilde{\mu}(z)$ reads [$\nu(0)$ is here $\nu(s=0)$ obtained by the integration by parts of the Laplace transform]

$$\tilde{\mu}(z) = -\nu(0) + z\tilde{\nu}(z) = -\frac{i\lambda^2}{\pi^2\epsilon} - \frac{\lambda^2}{\pi^2} z e^{-i\epsilon z}\mathrm{Ei}(-i\epsilon z). \tag{4.11}$$

Note that the cutoff ϵ significantly effects $\mu(z)$ only at large values of z, which essentially correspond to the very short time limit, i.e., the time where the two-level atom starts ''getting acquainted'' with the photon reservoir. At larger times ($t \gg \epsilon$) we do not expect the cutoff to contribute significantly in the evolution. This is a rather typical behavior in quantum Brownian motion models, provided that the ultraviolet cutoff of the environment is much larger than the natural frequencies of the system.

To evaluate the integral we first have to find the poles of the denominator. We can do that in a perturbation expansion. First, let us absorb the divergent $\nu(0)$ factor in a frequency renormalization

$$\tilde{\omega} = \omega - \frac{\lambda^2}{\pi^2\epsilon}, \tag{4.12}$$

so that we need find the zeros of the function $z + i\omega z - (\lambda^2/\pi^2)e^{-i\epsilon z}\mathrm{Ei}(-i\epsilon z)$. Looking for a solution in the vicinity of $z = -i\tilde{\omega}$, we find

$$\begin{aligned} z &= -i\tilde{\omega} - \tilde{\mu}(-i\tilde{\omega}), \\ &-i\left[\tilde{\omega} - \frac{\lambda^2\tilde{\omega}}{\pi^2}\ln(e^{\gamma}\epsilon\tilde{\omega})\right] - \frac{\lambda^2\tilde{\omega}}{\pi} + O(\lambda^4) := -i\Omega - \Gamma. \end{aligned} \tag{4.13}$$

We have a pole with a negative real part and we can verify numerically (also physically, as expected) that there is no pole with a greater real part. This means that when evaluating the inverse Laplace transform we can ignore the contribution of the branch cut at $z=0$ (being on the right of the pole) and hence after some time, where all possible other poles with absolutely larger values of their real parts will have stopped contributing, the solution will be

$$u(s) = e^{-i\Omega s - \Gamma s}. \tag{4.14}$$

This implies a Markovian time evolution and the identification of decoherence with relaxation time with $\Gamma^{-1} = (\pi/\lambda^2\omega)$. This is a general feature of the presence of a continuum of modes as can be seen from the case of an atom transparent to all modes but the ones in a strip, say, $[\omega_1, \omega_2]$, containing the resonance frequency.

It is easy to verify that in this case we have again

$$\tilde{\mu}(z) = -i\frac{\lambda^2}{\pi^2}(\omega_2 - \omega_1) + \frac{\lambda^2}{\pi^2}\ln\left(\frac{\omega_2 - iz}{\omega_1 - iz}\right). \tag{4.15}$$

Hence defining again

$$\tilde{\omega} = \omega - \frac{\lambda^2}{\pi^2}(\omega_2 - \omega_1), \tag{4.16}$$

we can find the pole at

$$z = -i\left[\tilde{\omega} - \frac{\lambda^2\omega}{\pi^2}\ln\left(\frac{\omega_2 - \tilde{\omega}}{\tilde{\omega} - \omega - 1}\right)\right] - \frac{\lambda^2\tilde{\omega}}{\pi} + O(\lambda^4). \tag{4.17}$$

Note that the real part of the pole comes from the presence of a minus sign in a logarithm of some real valued object. Hence in the case where the atom's frequency is outside the strip of interacting modes there will be no dissipation. This feature separates us from the QBM case, characterizing the atom-field system as primarily a resonant one.

C. Atom in a cavity

Let us now consider the case of the atom lying within a cavity consisting of two parallel plates at distance L. The field satisfies Dirichlet boundary conditions on the surface of the plates, hence the modes in the normal direction to the plates are multiples of π/L. The kernel then reads

$$\begin{aligned} \mu(s) &= \frac{\lambda^2}{2\pi L}\sum_n \int_0^\infty \frac{k\,dk}{[k^2 + (n\pi/L)^2]^{1/2}} e^{-i[k^2 + (n\pi/L)^2]^{1/2}(s - i\epsilon)} \\ &= \frac{1}{2\pi L}\sum_n \int_{|n\pi/L|}^\infty dk\, e^{-ik(s - i\epsilon)} \\ &= \frac{1}{2\pi L}\frac{1}{\epsilon + is}\sum_n e^{-i|n\pi/L|(s - i\epsilon)} \qquad (4.18) \\ &= \frac{1}{2\pi L}\frac{1}{\epsilon + is}\frac{1 + e^{-i\pi/L(s - i\epsilon)}}{1 - e^{-i\pi/L(s - i\epsilon)}}. \qquad (4.19) \end{aligned}$$

Hence we can compute its Laplace transform [36]

$$\begin{aligned} \tilde{\mu}(z) &= \frac{-i\lambda^2}{2\pi L}\int_0^\infty ds e^{-sz}\frac{1 + e^{-i\pi/L(s - i\epsilon)}}{s - i\epsilon}\frac{1 + e^{-i\pi/L(s - i\epsilon)}}{1 - e^{-i\pi/L(s - i\epsilon)}} \\ &= \frac{-i\lambda^2}{2\pi L} J\left(-i\epsilon z, \frac{i\pi}{2Lz}\right), \end{aligned} \tag{4.20}$$

where $J(x,a)$ is defined by

$$J(x,a) = \int_x^\infty \frac{dy e^{-y}}{y}\coth(ay) \tag{4.21}$$

and appears in Eq. (4.20) through analytical continuation in the complex plane. This integral can actually be computed at the limit of vanishing x ($\epsilon \to 0$)—see Refs. [37], Eq. 3.427.4

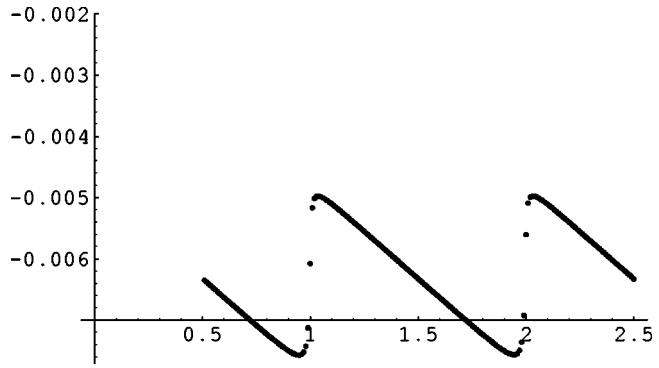

FIG. 1. Real part of a pole (negative dissipation constant Γ) as a function of $\omega/(\pi/L)$.

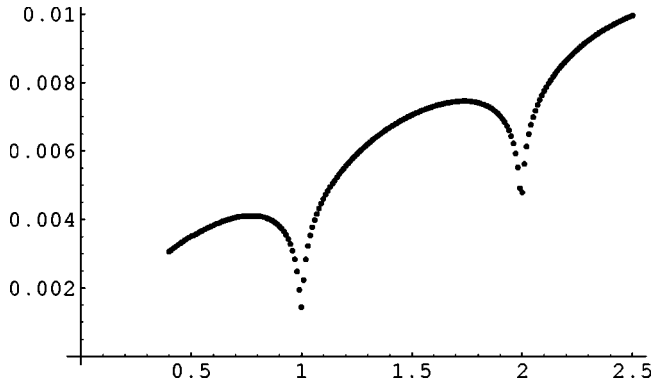

FIG. 2. Frequency shift $\Delta\Omega$ as a function of $\omega/(\pi/L)$.

$$\tilde{\mu}(z)=\frac{-i\lambda^2}{\pi^2\epsilon}+\frac{\lambda^2}{\pi^2}z\ln(ie^{\gamma}\epsilon z)-\frac{i\lambda^2}{\pi L}\left[\ln\Gamma\left(\frac{Lz}{i\pi}\right)-\frac{Lz}{i\pi}\ln\left(\frac{Lz}{i\pi}\right)+\frac{Lz}{i\pi}+\frac{1}{2}\ln\frac{Lz}{2i\pi^2}\right]+O(\epsilon). \tag{4.22}$$

Note that $\tilde{\mu}(z)$ is a sum of the term of case 2 ($L\to\infty$) and a finite one (no dependence on ϵ). The logarithm of the Γ function gives a countable number of branch cuts at $z=-i(n/\pi L)$, where n is the positive integer, the resonance modes of the cavity. Again the important pole has a negative real part. We can again compute the pole perturbatively. It lies at

$$z=-i\left[\tilde{\omega}+\frac{\lambda^2}{\pi^2}\tilde{\omega}\ln(e^{\gamma}\epsilon\tilde{\omega})-\frac{\lambda^2}{\pi L}\ln\Gamma(-L\tilde{\omega}/\pi)-\frac{L\tilde{\omega}}{\pi}\ln(-L\tilde{\omega}/\pi)-\frac{L\tilde{\omega}}{\pi}-\frac{1}{2}\ln\left(-\frac{L\tilde{\omega}}{2\pi^2}\right)\right]. \tag{4.23}$$

Clearly the logarithm of the Γ functions is the term out of which the real part of the pole appears. Since the real part of the pole is negative the branch cut is excluded from the integration contour and hence

$$u(s)=e^{-i\Omega s-\Gamma s}. \tag{4.24}$$

Here Γ gives a dissipation constant. In Fig. 1 we give a plot of the real part of the pole $(-\Gamma)$ as a function of the frequency ω. Note that it has sharp maxima on the resonance points, implying persistence of coherence.

Already from the approximation (4.23) we observe that the difference in the renormalized frequency from the case $L\to\infty$ $(\Delta\omega)$ is finite. Unfortunately, perturbation expansion is not reliable when $\tilde{\omega}$ is close to the resonance frequencies (this corresponds to negative integers arguments in the Γ function where it diverges) and for this regime we have not been able to get any analytic results. In Fig. 2 we have plotted the dependence of $\Delta\omega=\Omega[L]-\Omega[\infty]$ as the frequency changes. This effect of the frequency shift for an atom within the cavity is well known, as well as its relation to the Casimir effect [38].

V. DISCUSSION

Let us now integrate what we have found and look at the overall picture. The physics of a 2LAEMF system at zero temperature is characterized by a number of time constants: (i) the inverse natural frequency ω_0^{-1}, (ii) the inverse coupling constant $g_k^{-1}=\sqrt{\omega_k}/\lambda$, (iii) the relaxation time constant Γ^{-1}, and (iv) The cavity size L (divided by c).

First consider a zero-temperature field in free space, thus ignoring factors (iv). Start with only one mode in the field in resonance with the atom, then the system undergoes Rabi nutation with frequency $\Omega\approx g\sqrt{n+1}$, where n is the photon number in the field. The collapse time (assuming a large mean photon number $\bar{n}$) is g^{-1}, and the revival time is $2\pi\sqrt{\bar{n}}/g$ [1]. Atom excitation becomes significant in a time much greater than ω_0^{-1} but shorter than g^{-1}. (This is the condition for a first-order perturbation theory to give reasonable results.) For a large number of modes, spontaneous emission occurs at the relaxation time scale $\Gamma^{-1}=\pi/g\gg\omega_0^{-1}$, which we found to be the same as the decoherence time – the time for the off-diagonal elements of the reduced density matrix to decay (Sec. IV). When the mean number of photons in the field is large ($\bar{n}\gg 1$), they become comparable to the collapse time. This is a measure of the coherence in the atom-field system, and is controlled mainly by their coupling and the photon number in the field. We see that with the resonance condition, the nature of decoherence in 2LS is very different from the QBM situation, where phase information in the Brownian particle is efficiently dispersed in the many modes in the bath coupled almost equally to the system. As we remarked in the Introduction, the identification of the phase information and energy flow from the 2LS to its environment is similar to the spin-echo phenomena (Landau ''damping'') which is based on statistical mixing rather than dissipation. The mathematical distinction lies between considering the system coupled to the discrete number basis (our model) and the continuous amplitude basis (QBM) of the environment. The latter case essentially produces noise that drives the system in a way insensitive to its own intrinsic

dynamics. While in the former case, the coupling respects the internal dynamical structure of the 2LS and allows it to keep its coherence.

To see how the distribution of modes in a field changes the picture, the cavity-field calculation in Sec. IV is useful. As shown in Fig. 1, the relaxation constant develops peaks and minima. The resonance effect is enhanced by a cavity size commensurate with the natural frequency of the 2LA and dissipation weakens. Narrow-band resonance fluorescence as well as inhibition of spontaneous decay by frequent measurements – the quantum Zeno effect – are interesting phenomena for which our equations can provide finer details.

Non-Markovian processes involve memory effects (nonlocal in time). For the QBM problem, except for the case of a high-temperature Ohmic bath that gives Markovian dynamics, other types of spectral density (supra-Ohmic) or low temperatures, the dynamics of the system is non-Markovian [24]. When the reaction time of the bath is comparable to or faster than the natural time scale of the system (ω_0), one also expects to see non-Markovian behavior. In contrast, the 2LA is quite different: At zero temperature there is only one time scale, $\Gamma^{-1}=\lambda^{-2}\omega^{-1}\gg\omega^{-1}$, that determines both decoherence and relaxation. There is no memory effect and hence the process is Markovian. We expect that in finite temperatures the dynamics of the 2LA will be non-Markovian [39]. This is because there are more ways for the atom and the field to get entangled, and the memory effects of their interaction would presumably persist. In conclusion, we find that the 2LS interacting with an EM field is far more coherent than what is commonly believed, the misconception probably arising from the mistaken identification of this system with the Brownian model of an oscillator interacting with a harmonic-oscillator bath.

ACKNOWLEDGMENTS

We wish to thank Phillip Johnson and Adrian Dragulescu for a close reading of our paper and for making useful suggestions, and Sanjiv Shresta for checking the formulas and solutions to some nonlocal differential equations numerically and providing us with the figures. Sanjiv Shresta also first noticed a sign disrepancy in a sample calculation, which alerted us to possible ambiguities in the Grassmannian variable approach for finite temperature and coherent-state fields. We also thank Dr. Juan Pablo Paz for pointing out a mistake in an earlier version of this paper. This work is supported in part by NSF Grant No. PHY98-00967.

APPENDIX A: ATOM-FIELD INTERACTION: TWO-LEVEL SYSTEM

In this Appendix we give a rather detailed derivation of the Hamiltonian for a nonrelativistic atom interacting with a second-quantized electromagnetic field under the dipole, rotating-wave, and two-level approximations. This is to facilitate the comparison of our model (2.1) with $\sigma_\pm$ coupling with that used by others [Eq. (2.3)] with σ_z coupling. (See the Introduction.) To also make this useful for later papers in this series, we have included atomic motion. Note that the convention here is closer to [1] than that used in the text, which is closer to [5]. The conversion is explained in footnote 2.

The dynamics of a moving atom (mass M, momentum $\mathbf{P}$) whose electrons (charge e, mass m) interact with an electromagnetic field (vector potential $\mathbf{A}$, Coulomb potential V) is described by the (classical) Hamiltonian

$$H=\frac{\mathbf{P}^2}{2M}+\frac{1}{2m}(\mathbf{p}-e\mathbf{A})^2+eV(\mathbf{x})+H_b\,, \tag{A1}$$

where H_b is the Hamiltonian for the electromagnetic field. Expanding this equation, one can write

$$H=H_a+H_e+H_b+H_c\,, \tag{A2}$$

where

$$H_a=\frac{\mathbf{P}^2}{2M} \tag{A3}$$

describes nonrelativistic atom motion,

$$H_e=\frac{p^2}{2m}+eV(\mathbf{x}) \tag{A4}$$

decribes the dynamics of the (one) electron, while

$$H_{c1}=-\frac{e}{m}\mathbf{A}\cdot\mathbf{p} \tag{A5}$$

and

$$H_{c2}=\frac{e^2}{2m}\mathbf{A}^2 \tag{A6}$$

describe the coupling between the electron and the field. The second term makes no contribution to one-photon processes and will be ignored. We will refer to $H_0=H_e+H_b$ as the unperturbed Hamiltonian, and $H_I=H_{c1}$ as the interaction Hamiltonian.

In a second-quantized form, the Hamiltonian for the radiation field is given by

$$\hat{H}_b=\sum_{\mathbf{k}}\hbar\omega_{\mathbf{k}}\hat{b}^\dagger_{\mathbf{k}}\hat{b}_{\mathbf{k}}\,, \tag{A7}$$

where $\hat{b}^\dagger_{\mathbf{k}}$, $\hat{b}_{\mathbf{k}}$ are the creation and annihilation operators for the kth normal mode of a free massless vector field. Thus for the field vacuum $b_{\mathbf{k}}|0\rangle=0$, $\hat{b}_{\mathbf{k}}|0\rangle=0$, $[\hat{b}_{\mathbf{k}},\hat{b}^\dagger_{\mathbf{k}'}]=\delta_{\mathbf{k},\mathbf{k}'}$, for all $\mathbf{k}$. We can perform a harmonic decomposition of the vector potential of the electromagnetic field

$$\mathbf{A}(\mathbf{x},t)=\sum_{\mathbf{k}}\left(\frac{\hbar}{2\omega_{\mathbf{k}}\epsilon_0}\right)^{1/2}[b_{\mathbf{k}\sigma}\mathbf{u}_{\mathbf{k}\sigma}(\mathbf{x})e^{i\omega_{\mathbf{k}}t}+b^\dagger_{\mathbf{k}\sigma}\overline{\mathbf{u}}_{\mathbf{k}\sigma}(\mathbf{x})e^{-i\omega_{\mathbf{k}}t}] \tag{A8}$$

where, assuming the field is contained in a box of size L, the spatial mode functions $\mathbf{u}_{\mathbf{k}\sigma}$ are given by

$$\mathbf{u}_{\mathbf{k}\sigma}(\mathbf{x}) = L^{-3/2}\hat{\mathbf{e}}_{\mathbf{k}\sigma} f_{\mathbf{k}}(\mathbf{x}). \tag{A9}$$

Here $\hat{\mathbf{e}}_{\mathbf{k}\sigma}$ is the unit polarization vector and $\sigma = 1,2$ are the two (transverse) polarizations. In free space, $f_{\mathbf{k}}(\mathbf{x}) = e^{-i\mathbf{k}\cdot\mathbf{x}}$.

We assume that electron motion is much faster than the motion of the atom, thus it sees a stationary central Coulomb potential around the center of mass of the atom. Denoting the (time-independent, nonrelativistic) electronic wave-function eigenstate belonging to the eigenvalue E_i by ϕ_i, i.e., $H_e\phi_i = E_i\phi_i$, we can write the Hamiltonian for the electron in the second-quantized form as

$$\hat{H}_e = \sum_i E_i \hat{a}_i^\dagger \hat{a}_i, \tag{A10}$$

where i labels the bound states of the eletron (we assume vanishing probability for the atom to ionize) and $\hat{a}_i^\dagger$ and $\hat{a}_i$ are the creation and annihilation operators. As fermions they obey the anticommutation relations $\hat{a}_i^\dagger \hat{a}_j + \hat{a}_j \hat{a}_i^\dagger = \delta_{ij}$.

To perform perturbation theory, the electronic wave function of the interacting system is expanded in terms of the eigenfunctions ϕ_i of the unperturbed Hamiltonian, with basis formed by the direct product of the electron and the field states. Thus the electron field operator $\hat{\psi}(\mathbf{x})$ can be expanded as

$$\hat{\psi}(\mathbf{x}) = \sum_i \hat{a}_i \phi_i(\mathbf{x}). \tag{A11}$$

With this, the interaction Hamiltonians is given by

$$\hat{H}_I = -\frac{e}{m}\int \hat{\psi}^\dagger(\mathbf{x})(\mathbf{p}\cdot\mathbf{A})\hat{\psi}(\mathbf{x}) d^3x \tag{A12}$$

or in terms of $\hat{a}_i, \hat{b}_{\mathbf{k}}$ operators

$$\hat{H}_I = \hbar \sum_{i,j,\mathbf{k}} \hat{a}_i^\dagger \hat{a}_j (g_{ij\mathbf{k}} \hat{b}_{\mathbf{k}} + \overline{g}_{ij\mathbf{k}} \hat{b}_{\mathbf{k}}^\dagger) \tag{A13}$$

where

$$g_{ij\mathbf{k}} = -\frac{e}{m}\frac{1}{\sqrt{2\hbar\omega_{\mathbf{k}}\epsilon_0}}\int \overline{\phi}_i(\mathbf{x})\mathbf{u}_{\mathbf{k}\sigma}(\mathbf{x})\cdot\mathbf{p}\phi_j(\mathbf{x}) d^3x. \tag{A14}$$

1. Dipole approximation

Now consider conditions when the spatial variation of the vector potential $\mathbf{A}$ of the electromagnetic field is small compared to the electronic wave function ψ; one can expand $f_{\mathbf{k}}(\mathbf{x})$ in $\mathbf{u}_{\mathbf{k}\sigma}(\mathbf{x})$ around the position of the atom $\mathbf{x} = \mathbf{X} + \delta\mathbf{x}$:

$$e^{i\mathbf{k}\cdot\mathbf{x}} = e^{i\mathbf{k}\cdot\mathbf{X}}\left[1 + \mathbf{k}\cdot\delta\mathbf{x} - \frac{1}{2}(\mathbf{k}\cdot\delta\mathbf{x})^2 + \cdots\right]. \tag{A15}$$

The dipole approximation amounts to keeping just the leading term. Doing so, we can take the field mode function $f_{\mathbf{k}}(\mathbf{x})$ out of the integration above and evaluate it at the atomic position. To evaluate

$$\frac{e}{m}\int \overline{\phi}_i \mathbf{p} \phi_j d^3x,$$

we make use of

$$\frac{d\hat{x}_i}{dt} = \frac{\hat{p}_i}{m} = \frac{1}{i\hbar}[\hat{x}_i, \hat{H}_e], \tag{A16}$$

yielding,

$$\frac{e}{m}\int \overline{\phi}_i \mathbf{p} \phi_j d^3x = i\omega_{ij}\mathbf{p}d_{ij}, \tag{A17}$$

where $\hbar\omega_{ij} = E_i - E_j$, and $\mathbf{d}_{ij} \equiv e\int \overline{\phi}_i \mathbf{x} \phi_j d^3x$ is the dipole matrix element $\mathbf{d}_{ij} = \overline{\mathbf{d}}_{ji}$. Define

$$d_{ij\mathbf{k}} \equiv -\frac{i\omega_{ij}}{\sqrt{2\hbar\omega_{\mathbf{k}}\epsilon_0 V}}\mathbf{d}_{ij}\cdot\hat{\mathbf{e}}_{\mathbf{k}\sigma}. \tag{A18}$$

Note that $\overline{d}_{ij\mathbf{k}} = d_{ji\mathbf{k}}$. With this under the dipole approximation,

$$g_{ij\mathbf{k}} = d_{ij\mathbf{k}} f_{\mathbf{k}}(\mathbf{X}). \tag{A19}$$

2. Rotating-wave approximation

In the interaction picture, recalling that the time evolution of the ladder operators are given by

$$\hat{a}_i^\dagger(t) = \hat{a}_i^\dagger e^{i\omega_i t}, \quad \hat{a}_j(t) = \hat{a}_j e^{-i\omega_j t}, \quad \hat{b}_{\mathbf{k}}(t) = \hat{b}_{\mathbf{k}} e^{-i\omega_{\mathbf{k}} t}, \tag{A20}$$

the interaction Hamiltonian H_I in the interaction picture becomes

$$\tilde{H}_I = \hbar \sum_{i,j,\mathbf{k}} g_{ij\mathbf{k}} \hat{a}_i^\dagger \hat{a}_j \hat{b}_{\mathbf{k}} e^{i(\omega_{ij} - \omega_{\mathbf{k}})} + \hbar \sum_{ij\mathbf{k}} \overline{g}_{ij\mathbf{k}} \hat{a}_i^\dagger \hat{a}_j \hat{b}_{\mathbf{k}}^\dagger e^{i(\omega_{ij} + \omega_{\mathbf{k}})}, \tag{A21}$$

where $\omega_{ij} \equiv \omega_i - \omega_j$. We see there are two types of oscillatory terms present: $e^{-i(\omega_{ij} \pm \omega_{\mathbf{k}})}$. Processes most effective in the absorption or emission of a photon by the atom correspond to those with near resonance frequency $\omega_{ij} \approx \omega_{\mathbf{k}}$. Assuming $\omega_{ij} > 0$ $(E_i > E_j)$, the first type with $e^{-i(\omega_{ij} + \omega_{\mathbf{k}})}$ has a rapidly oscillating phase and its contribution is small compared with the second type with $e^{-i(\omega_{ij} - \omega_{\mathbf{k}})}$, whose stationary phase at near resonance gives a large contribution. Physically, the first type corresponds to either the excitation of the atom *along with* the emission of a photon or the relaxation of the atom *along with* the absorption of a photon, which is less probable than the second type corresponding to the excitation of an atom upon the absorption of a photon or the relaxation of an atom with the emission of a photon. We shall therefore ignore the first type of terms, which amounts to

working under the rotating-wave approximation (RWA). This is the second major approximation in this standard model.

3. Two-level atom

Let us now consider the idealized case when the atom has only two electronic states, $|+\rangle,|-\rangle$ corresponding to $i=2,1$, with energies equal to $E_{\pm}=\pm\frac{1}{2}\hbar\omega_0$. (The two states can interchangeably be labeled as $|1\rangle,|0\rangle$, or $|e\rangle,|g\rangle$ or $|\uparrow\rangle,|\downarrow\rangle$.) Thus $\omega_{ij=21}=\omega_0$. Thus

$$\hat{H}_a=\frac{\hbar\omega_0}{2}(a_2^\dagger a_2-a_1^\dagger a_1)\equiv\frac{\hbar\omega_0}{2}\sigma_z\equiv\hbar\omega_0 S_z, \quad \text{(A22)}$$

where we have introduced a Pauli matrix (2×2) representation $\sigma_z=\mathrm{diag}(1,-1)$ For the interaction Hamiltonian above, under the RWA, in the $i=2,j=1$ contribution to the summation, the first line containing $e^{i(\omega_0-\omega_{\mathbf{k}})}$ is kept, while the second line is dropped. The reverse is true for the $i=1,j=2$ term. The interaction Hamiltonian (in the interaction picture) now becomes

$$\tilde{H}_I=\hbar\sum_{\mathbf{k}}\ [g_{21\mathbf{k}}\hat{a}_2^\dagger\hat{a}_1\hat{b}_{\mathbf{k}}e^{-i(\omega_{\mathbf{k}}-\omega_{21})t} + \overline{g}_{12\mathbf{k}}\hat{a}_1^\dagger\hat{a}_2\hat{b}_{\mathbf{k}}^\dagger e^{-i(\omega_{\mathbf{k}}+\omega_{12})t}]. \quad \text{(A23)}$$

Introducing the Pauli matrix representation for the fermion operators $a_2^\dagger a_1\to\sigma_+\equiv S_+$ and $a_1^\dagger a_2\to\sigma_-\equiv S_-$, and defining $g_{\mathbf{k}}\equiv d_{21\mathbf{k}}=\overline{d}_{12\mathbf{k}}$ [recall $g_{ij\mathbf{k}}\equiv d_{ijk}f_{\mathbf{k}}(\mathbf{X})$] we can write the interaction Hamiltonian (in the Heisenberg picture) in a simple form:

$$\hat{H}_I=\hbar\sum_{\mathbf{k}}\ g_{\mathbf{k}}[S_+b_{\mathbf{k}}f_{\mathbf{k}}(\mathbf{X})+S_-b_{\mathbf{k}}^\dagger\overline{f}_{\mathbf{k}}(\mathbf{X})]. \quad \text{(A24)}$$

Therefore the total Hamiltonian for our model of a moving atom interacting with a quantum electromagnetic field under the dipole, rotating-wave, and two-level approximation is given by

$$\hat{H}=\frac{\hat{\mathbf{P}}^2}{2M}+\hbar\omega_0\hat{S}_z+\hbar\sum_{\mathbf{k}}\ [\omega_{\mathbf{k}}\hat{b}_{\mathbf{k}}^\dagger\hat{b}_{\mathbf{k}}+g_{\mathbf{k}}(f_{\mathbf{k}}(\mathbf{X})S_+b_{\mathbf{k}} + \overline{f}_{\mathbf{k}}(\mathbf{X})S_-b_{\mathbf{k}}^\dagger)]. \quad \text{(A25)}$$

APPENDIX B: AN OPERATOR PROOF OF THE MASTER EQUATION

We use the resolvent decomposition of the propagator

$$e^{-iHt}=\int\frac{dEe^{-iEt}}{E-H}. \quad \text{(B1)}$$

Then by writing $H=H_0+H_I$ we can expand the resolvent and get

$$(E-H)^{-1}=(E-H_0)^{-1}(1-(E-H_0)^{-1}H_I)^{-1} =(E-H_0)^{-1}\Big(1+(E-H_0)^{-1}H_I+\frac{1}{2}(E-H_0)^{-1}H_I(E-H_0)^{-1}H_I+\ \ldots\Big). \quad \text{(B2)}$$

When we act, the expanded resolvent in any vector $|0,i\rangle=|0\rangle\otimes|i\rangle$, ($|i\rangle$ denotes an eigenstate of the Hamiltonian of the two-level system) we see that only expanded terms that contain alternating sequences of σ_+ and σ_- survive. This makes the summation much easier. It is also easier if we note that

$$(E-H_0)^{-1}\sum_k\ g_kb_k(E-H_0)^{-1}g_{k'}\sum_{k'}\ b_{k'}^\dagger|0\rangle =(E-\omega_0)^{-1}\sum_k\ \frac{g_k^2}{E-\omega_k}|0\rangle:=(E-\omega_0)^{-1}F(E). \quad \text{(B3)}$$

Hence we can compute the matrix elements by resumming the expansion

$$\langle z,0|(E-H)^{-1}|0,0\rangle=E^{-1}, \quad \text{(B4)}$$

$$\langle z,1|(E-H)^{-1}|0,0\rangle=0, \quad \text{(B5)}$$

$$\langle z,1|(E-H)^{-1}|0,1\rangle=(E-\omega_0-F(E))^{-1}, \quad \text{(B6)}$$

$$\langle z,0|(E-H)^{-1}|0,0\rangle=\frac{E-\omega_0}{E-\omega_0-F(E)}\sum_k\ \frac{g_k}{(E-\omega)^2}\overline{z}_k. \quad \text{(B7)}$$

The reduced density-matrix propagator in the energy basis of the two-level atom and for the vacuum initial state is

$$J(i,j;t|m,n;0)=\int dEdE'e^{-i(E-E')t} \times\int DzD\overline{z}e^{-\sum_k\ \overline{z}_kz_k}[\langle z,i|(E-H)^{-1}|0,m\rangle \times\langle 0,n|(E-H)^{-1}|z,j\rangle]. \quad \text{(B8)}$$

We can then verify that the only nonzero elements $mn\to ij$ are the following and their conjugates

$$J(0,0;t|0,0;0)\to E^{-1}E'^{-1}, \quad \text{(B9)}$$

$$J(0,1;t|0,1;0)\to E^{-1}(E'-\omega_0-F(E'))^{-1}, \quad \text{(B10)}$$

$$J(0,0;t|0,0;0)\to\frac{E-\omega_0}{E-\omega_0-F(E)}\frac{E'-\omega_0}{E'-\omega-F(E')} \times\sum_k\ \frac{g_k^2}{(E-\omega_k)(E'-\omega_k)}, \quad \text{(B11)}$$

$$J(11;t|1,1;0)\rightarrow(E-\omega_0-F(E))^{-1}(E-\omega_0-F(E'))^{-1}. \tag{B12}$$

Then it is easy to check that this reproduces the propagation as given by Eq. (3.21) that was obtained through the influence functional method. Indeed

$$u(t)=\int \frac{dEe^{-iEt}}{E-\omega_0-F(E)} \tag{B13}$$

is exactly the same as the one defined by Eq. (3.9).

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