Population equations for quantum systems in contact with dissipation mechanisms

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We discuss the construction of population equations for driven quantum systems in contact with dissipation mechanisms in the limit where the strength of the driving force is sufficiently weak that a suitable Born expansion can be carried out in powers of the coupling constant of the coherent interaction. The Zwanzig projector technique and the application of an appropriate eigenfunction-expansion method due to Weidlich lead to an elegant derivation of population equations. If the decay rates of the irreversible processes allow the application of the Markoff approximation, ordinary first-order differential equations for the level populations can be derived. The transition rates are constructed explicitly in terms of the coherent Liouville operator and the Weidlich eigenfunctions.

I. INTRODUCTION

Many quantum-mechanical systems of statistical interest often allow a clear distinction to be made between a limited set of relevant dynamical variables (the system) and a much larger collection of degrees of freedom (the reservoir) whose statistical properties remain essentially unchanged during the evolution. The reduced density operator W of the system of interest often obeys a master equation of the form[1]

$$\frac{dW}{dt} \equiv \left(\frac{dW}{dt}\right)_{\text{coh}} + \left(\frac{dW}{dt}\right)_{\text{incoh}} = -iL_c W + \Lambda W ,$$
$$L_c W = \frac{1}{\hbar}[H,W] , \tag{1.1}$$

where the total rate of change is simply given by the sum of a coherent and a dissipative part. In Eq. (1.1) the so-called Liouville operator L_c controls the coherent part of the evolution; H is the system's total Hamiltonian, and ΛW describes the dissipative action of the reservoir(s).

Both open and closed systems have been described by master equations of this form.[2] Well-known examples include the laser,[1] closed systems interacting with a thermal bath,[1,2(c)] and open bistable systems driven by external sources.[2(d)] Obviously, a much more extensive list could be compiled.

The reduced density operator contains complete statistical information about the observables of interest; this can be extracted using various techniques. Thus, for example, in the case of the resonance fluorescence problem[2(b),3] an atom is driven by an external field and forced to undergo Rabi oscillations between two resonant levels, while the vacuum of radiation causes a decay of the atomic polarization and excited population. The off-diagonal matrix elements of the atomic density operator provide information that is needed, for example, to calculate the spectrum of resonance fluorescence, the diagonal elements, instead, describe the energy relaxation process and are connected to the correlation properties of the radiation field intensity.

It is clear that, in general, an accurate description of the system's dynamics requires knowledge of a large number of matrix elements of W. If the decay process of the off-diagonal part of the density operator is sufficiently fast, one can introduce a drastic simplification by assuming that the evolution of the relevant variables can be described adequately by the set of occupation probabilities $W_n(t)$.

In the so-called rate equation limit, the population dynamics is assumed to obey a set of equations whose typical structure is[4]

$$\frac{dW_i}{dt} = \sum_j (P_{ji}W_j - P_{ij}W_i) , \tag{1.2}$$

where $W_i(t)$ denotes the occupation probability of the ith level, and P_{ij} denotes the time-independent transition rate from level i to level j. As appealing as Eq. (1.2) may appear on intuitive grounds, it is not obvious in general under what conditions it can be applied with confidence or how one should construct the transition rates P_{ij}.[5]

An elegant procedure developed by Zwanig[6] (projector technique) has been used to generate rate equations of the form

$$\frac{dW_i}{dt} = \sum_j \int_0^t d\tau [\tilde{P}_{ji}(\tau)W_j(t-\tau) - \tilde{P}_{ij}(\tau)W_i(t-\tau)] , \tag{1.3}$$

where the generalized transition coefficients $\tilde{P}_{ij}(\tau)$ [*note*: $\tilde{P}_{ij}(\tau)$ are no longer rates] contain information about memory effects. Equations of type (1.3), in general, provide an improved description relative to the rate equations (1.2). On occasions, however, warning flags have been

raised with regard to the application of this result. Thus, for example, the formal development of an early single-mode model of superfluorescence[7] led to a set of equations of type (1.3) for the occupation probabilities of the atomic levels. A careful analysis of the time scales of the problem, however, has shown that the decay time of the kernel functions $\tilde{P}_{ij}(\tau)$ is the shortest resolvable time and that the non-Markoffian master equation ought to be replaced, for consistency's sake, by its Markoffian counterpart (1.2) according to the usual rules.

It is clear from a recent paper by Swain[5(b)] that the exact construction of rate equations is not a trivial task. This is not surprising, of course, because such development requires the exact elimination of the off-diagonal part of the density operator, unless one is willing to relinquish coherence information.

If one is mainly concerned with the population dynamics, and if the decay rate of the off-diagonal matrix elements is much larger than that of the diagonal ones, population equations can be derived after adiabatic elimination of the faster evolving variables. This procedure is straightforward from a conceptual point of view, but it is rarely easy to carry out, in practice, except when the number of variables to be eliminated is manageably small.

In this paper we want to discuss a class of problems of physical interest when the explicit derivation of population equations can be carried out with relative ease and in such a way that the nature of the approximations can be assessed with some confidence. We are especially interested in the case when the characteristic rate of the coherent exchange induced by the system Hamiltonian is sufficiently smaller than the largest decay rate of the dissipative process (formally $L_c \ll \Lambda$). A Born expansion scheme can then be carried out, and a set of rate equations of type (1.3) can be derived. If, in addition, the decay rate of the off-diagonal matrix elements is also much larger than that of the diagonal part of W, ordinary rate equations of type (1.2) can be derived as a limiting case. This is, in fact, the usual scenario when the rate equation approximation is expected to be valid.

In this paper we discuss these ideas with the help of the Zwanig projector technique. In the special case when the dynamical variables of interest are harmonic oscillator degrees of freedom, the application of a clever scheme of calculation proposed by Weidlich[8] provides a transparent illustration of our results.

In Sec. II we describe the formal derivation of the population equations. In Sec. III we illustrate the main results of our formal development with the help of the well-known model of driven and damped harmonic oscillator. Under conditions such that the Markoff approximation can be carried out, the population equations are reduced to the familiar rate equation form (Sec. IV).

II. MASTER EQUATION FOR THE DIAGONAL PART OF THE DENSITY OPERATOR

In anticipation of the needed distinction between phase and energy relaxation processes, whose end and effect is to cause a decay of the diagonal part of the reduced density operator, or of both diagonal and off-diagonal matrix elements, we consider a system governed by the master equation

$$\frac{dW}{dt} = -iL_c W + \Lambda_1 W + \Lambda_2 W \equiv -i\mathcal{L}W \tag{2.1}$$

for the density operator W of the relevant observables. The reversible term $-iL_cW \equiv -(i/\hbar)[H,W]$ describes the coherent interaction of the system's internal degrees of freedom with one another or with external driving forces. The irreversible decay terms $\Lambda_1 W$ and $\Lambda_2 W$ result from the action of one or more thermal reservoirs. We shall assume $\Lambda_2 W$ to be responsible for the decay of the off-diagonal matrix elements.

Following Zwanig,[6] we introduce a projector operator $P(P=P^2)$ and formally define

$$W_r = PW, \quad W_i = (1-P)W \equiv \bar{P}W, \tag{2.2}$$

as the relevant and irrelevant parts of the density operator, respectively. The coupled equations of motion for W_r and W_i are

$$\dot{W}_r = -iP\mathcal{L}(W_r + W_i), \tag{2.3a}$$

$$\dot{W}_i = -i\bar{P}\mathcal{L}(W_r + W_i). \tag{2.3b}$$

If the Liouville operator $\mathcal{L}$ is explicitly time independent, we can solve Eq. (2.3b) formally with the result

$$W_i(t) = e^{-i\bar{P}\mathcal{L}t}W_i(0) + \int_0^t d\tau\, e^{-i\bar{P}\mathcal{L}(t-\tau)}[-i\bar{P}\mathcal{L}W_r(\tau)]. \tag{2.4}$$

The case in which $\mathcal{L}$ depends explicitly on time can be handled in a similar way with the added complications introduced by the need for considering the time-ordering prescription. If we now assume, as is often the case, that $W_i(0)=0$, and if we substitute the formal solution (2.4) into Eq. (2.3a), the relevant part of the density operator is found to satisfy the following integro-differential equation of motion:

$$\dot{W}_r(t) = -iP\mathcal{L}W_r(t) - iP\mathcal{L}\int_0^t dt\, e^{-i\bar{P}\mathcal{L}(t-\tau)} \times [-iP\mathcal{L}W_r(\tau)]. \tag{2.5}$$

This result is exact. If $W_r(t)$ is the diagonal part of the reduced density operator, Eq. (2.5) provides, in principle, an exact dynamical description of the population evolution. Unfortunately, an exact handling of Eq. (2.5) is an exceedingly complicated (and probably hopeless) task.

If $\{|n\rangle\}$ is a complete orthonormal collection of states of the system, a convenient representation for the projector operator P, such that $W_r = PW$ is the diagonal part of the density operator, is given by

$$P \equiv \sum_n |n\rangle\langle n|\langle n|\ldots|n\rangle . \tag{2.6}$$

The symbol $\langle n|\ldots|n\rangle$ denotes the diagonal matrix element of any operator to which P is applied.

We shall now assume the validity of a few formal properties of the chosen projector. They appear to be satisfied rather generally and can be readily tested in the specific case discussed in Secs. III and IV. Specifically, we assume the validity of the equations[9]

$$P\Lambda_1 = \Lambda_1 P , \tag{2.7a}$$

$$P\Lambda_2 = \Lambda_2 P = 0 , \tag{2.7b}$$

$$PL_c P = 0 . \tag{2.7c}$$

It is now a simple matter to show that Eq. (2.5) with the help of Eqs. (2.7) can be cast into the new and still exact form

$$\dot{W}_r = \Lambda_1 W_r - \int_0^t d\tau PL_c e^{-i\bar{P}\mathcal{L}(t-\tau)} L_c W_r(\tau). \tag{2.8}$$

The integral term on the right-hand side of Eq. (2.8) contains the coupling constant of the coherent interaction term to all orders. If the system is such that the decay rate of the irreversible process is much larger than the characteristic frequency associated with the coherent energy exchanges induced by the Hamiltonian H (formally, if Λ_1 or $\Lambda_2 \gg L_c$), one may neglect all terms in the Born expansion of the exponential operator except for the largest one. In this case we set

$$e^{-i\bar{P}\mathcal{L}(t-\tau)} \simeq e^{\bar{P}(\Lambda_1+\Lambda_2)(t-\tau)} . \tag{2.9}$$

Furthermore, Eqs. (2.7) can be used again to yield the identities

$$e^{\bar{P}(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r = e^{(\Lambda_1+\Lambda_2)(t-\tau)} PL_c W_r = e^{(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r . \tag{2.10}$$

Thus, in the Born approximation, the diagonal part of the density operator satisfies the generalized master equation

$$\frac{dW_r}{dt} = \Lambda_1 W_r - \int_0^t d\tau PL_c e^{(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r(\tau) . \tag{2.11}$$

This development, of course, is not new. It has been carried out here in some detail to emphasize the nature of approximation (2.9) and to set the stage for the next phase of our discussion. The structure of Eq. (2.11) guarantees the conservation of the trace of W_r because

$$\text{(i)}\quad \mathrm{Tr}\Lambda_1 W_r = 0 , \tag{2.12}$$

$$\text{(ii)}\quad \mathrm{Tr}PL_c e^{(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r(\tau) = \mathrm{Tr}L_c X(t,\tau) = 0 \tag{2.13}$$

$$X(t,\tau) \equiv \exp[(\Lambda_1+\Lambda_2)(t-\tau)] L_c W_r(\tau) .$$

The last step in Eq. (2.13) follows from the definition of the projector P and from the well-known property of the trace, $\mathrm{Tr}[A,B]=0$, where A and B are arbitrary operators in the Hilbert space of the system. From Eq. (2.11) it is obvious that the diagonal matrix elements of W satisfy the coupled equations

$$\frac{d(W_r)_n}{dt} = (\Lambda_1 W_r)_n - \int_0^t d\tau \langle n| L_c e^{(\Lambda_1+\Lambda_2)(t-\tau)} \times L_c W_r(\tau)|n\rangle , \tag{2.14}$$

where $(W_r)_n \equiv \langle n|W_r|n\rangle$. The problem is now reduced to the explicit evaluation of the matrix elements that appear inside the integral in Eq. (2.14). This may still be a difficult problem, in general. Its solution can be simplified considerably in a number of problems of interest with the help of a clever procedure developed by Weidlich.[8]

It is convenient to regard the Liouville space of the $\mathcal{L}$ operators as a Hilbert space in which the scalar product between any pair of "state vectors" $\|A)$ and $\|B)$ is defined as

$$(A\|B) \equiv \mathrm{Tr}(A^\dagger B) . \tag{2.15}$$

In the Hilbert space of the system's observables, A is an ordinary operator, as it appears on the right-hand side of the above equation; on the left-hand side, instead, it plays the role of a state vector which obeys the mapping rule $\mathcal{L}\|A) = \|A')$ under the action of an arbitrary Liouville operator. Weidlich has shown[8] that if the "state vectors" $\|A_p)$ satisfy the eigenvalue equation

$$\mathcal{L}\|A_p) = \lambda_p \|A_p) , \tag{2.16}$$

and $\|B_p)$ satisfy the adjoint equation

$$\mathcal{L}^\dagger \|B_p) = \lambda_p^* \|B_p) , \tag{2.17}$$

the following completeness relation holds:

$$\sum_{\rho} \| A_\rho)(B_\rho \| = \underline{1}\,, \tag{2.18}$$

where $\underline{1}$ is the identity operator. The eigenstates $\| A_\rho)$ and $\| B_\rho)$ form a biorthogonal set in the sense that $(B_\rho \| A_{\rho'}) = \delta_{\rho\rho'}$. Explicit expressions for $\| A_\rho)$ and $\| B_\rho)$ have been derived in Ref. 8 for two rather common models of damping Liouvillians. They will be discussed further in the next section of this paper. Here we observe that the matrix elements in Eq. (2.14) can be put in a much more convenient form with the help of the completeness relation (2.18) and the eigenvalue equation (2.16). This can be accomplished through the following steps:

$$\begin{aligned}\langle n | L_c e^{(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r(\tau)|n\rangle &= \langle n | L_c e^{(\Lambda_1+\Lambda_2)(t-\tau)} \sum_\rho \| A_\rho)(B_\rho \| L_c W_r(\tau)|n\rangle \\ &= \sum_\rho e^{\lambda_\rho(t-\tau)} \langle n | L_c A_\rho | n\rangle \operatorname{Tr}[B_\rho^\dagger L_c W_r(\tau)]\,.\end{aligned} \tag{2.19}$$

In Eq. (2.19), λ_ρ denotes the eigenvalue of the total damping Liouvillian $(\Lambda_1+\Lambda_2)$. The relation between Eq. (2.14) and the more familiar looking population equations becomes clear at this point if one observes that

$$\operatorname{Tr}[B_\rho^\dagger L_c W_r(\tau)] = -\operatorname{Tr}[(L_c B_\rho^\dagger) W_r(\tau)]$$

and that

$$\operatorname{Tr}[(L_c B_\rho^\dagger) W_r(\tau)] = \sum_{n'} (L_c B_\rho^\dagger)_{n'} (W_r(\tau))_{n'}\,, \tag{2.20}$$

where the symbol $(\ldots)_{n'}$ denotes the diagonal matrix element of the operator in parentheses. After minor manipulations, Eq. (2.14) can be cast into the final form

$$\frac{d(W_r)_n}{dt} = (\Lambda_1 W_r)_n + \int_0^t d\tau \sum_{n'} \tilde{P}_{nn'}(t-\tau)(W_r(\tau))_{n'}\,. \tag{2.21}$$

The generalized transition coefficients $\tilde{P}_{nn'}$ have been defined by

$$\tilde{P}_{nn'}(t-\tau) = \sum_\rho e^{\lambda_\rho(t-\tau)} (L_c A_\rho)_n (L_c B_\rho^\dagger)_{n'}\,. \tag{2.22}$$

The range of validity of the integro-differential equation (2.21) is determined by the accuracy of the Born approximation $(\Lambda_1+\Lambda_2 \gg L_c)$. The implicit dependence of the matrix elements $(W_r)_n$ on the details of the previous history through the memory kernel functions $\tilde{P}_{nn'}$ is a reflection of the role played by the off-diagonal matrix elements of the density operator on the evolution of the system.

If the physical situation of interest is such that the relaxation rate of the off-diagonal elements of W is much larger than that of the diagonal part (formally, if $\Lambda_2 \gg \Lambda_1$), the Markoff approximation can be carried out in the usual way with the result

$$\frac{d(W_r)_n}{dt} = (\Lambda_1 W_r)_n - \sum_{n'} P_{nn'}(W_r)_{n'}\,, \tag{2.23}$$

where

$$\begin{aligned}P_{nn'} &\equiv \int_0^\infty d\tau \tilde{P}_{nn'}(\tau) \\ &= \sum_\rho \frac{1}{|\lambda_\rho|} (L_c A_\rho)_n (L_c B_\rho^\dagger)_{n'}\end{aligned}$$

can be identified with the time-independent transition rates of the ordinary population rate equations.

III. DRIVEN DAMPED HARMONIC OSCILLATOR—AN EXAMPLE OF POPULATION EQUATION IN THE BORN APPROXIMATION

As an application of the formalism developed in the previous section, we consider the well-known model of a driven damped harmonic oscillator. For the sake of simplicity, we consider the case of a resonant harmonic driving force. In the interaction representation, the coherent Liouvillian takes the form

$$L_c = \Omega_R[a^\dagger + a, \ldots]\,, \tag{3.1}$$

where Ω_R plays the role of a Rabi frequency. The dissipative terms are given by[10]

$$\begin{aligned}\Lambda_1 W &= \gamma_\downarrow([a, Wa^\dagger] + [aW, a^\dagger]) \\ &\quad + \gamma_\uparrow([a^\dagger, Wa] + [a^\dagger W, a])\,,\end{aligned} \tag{3.2}$$

$$\Lambda_2 W = \eta([a^\dagger a, Wa^\dagger a] + [a^\dagger aW, a^\dagger a])\,, \tag{3.3}$$

where the damping rates $\gamma_\uparrow$, $\gamma_\downarrow$, and η contain implicit information about the reservoir(s). Their physical significance is well known. Thus, the expectation values of the amplitude and number operators a, $a^\dagger a$ under the action of the damping mechanism alone are governed by equations of motion,

$$\frac{d}{dt}\langle a\rangle_{\text{irrev}} = -(\gamma_\downarrow - \gamma_\uparrow)\langle a\rangle - \eta\langle a\rangle\,, \tag{3.4}$$

$$\frac{d}{dt}\langle a^\dagger a\rangle_{\text{irrev}} = -2(\gamma_\downarrow - \gamma_\uparrow)\langle a^\dagger a\rangle + 2\gamma_\uparrow\,, \tag{3.5}$$

with Λ_2 affecting only off-diagonal elements of the density operator.

The first step, and very nearly also the end of

the calculation, consists of deriving an explicit expression for the generalized transition coefficients $\tilde{P}_{nn'}(t-\tau)$ defined by Eq. (2.22). The method adopted here is not unique, but it is probably the most elegant and direct procedure. In Appendix A, we illustrate a different scheme in connection with the problem of the driven-damped two-level atom.

For our purposes, we need the "eigenstates" $\|A_p), \|B_p)$ and the eigenvalues λ_p [see Eqs. (2.16) and (2.17)]. These quantities appear explicitly in Weidlich's paper.[8] Here we only need to recall the main results of relevance for our development. The spectrum of eigenvectors and eigenvalues of the (non-Hermitian) Liouville operator $\Lambda_1+\Lambda_2$ [Eqs. (3.2) and (3.3)] is labelled by two indices $p=0,1,2,\ldots$, and $q=0,\pm1,\pm2,\ldots$. The $\|A)$ eigenstates have the form

$$\|A_{pq}) = \begin{cases} \left[e^{-a^\dagger a/\bar{n}} a^q L_p^q\left(\frac{a^\dagger a}{\bar{n}}\right)\right]_{\text{antinor}}, & q \geq 0 \quad (3.6) \\ \left[e^{-a^\dagger a/\bar{n}} a^{\dagger|q|} L_p^{|q|}\left(\frac{a^\dagger a}{\bar{n}}\right)\right]_{\text{antinor}}, & q \leq 0 \quad (3.7) \end{cases}$$

where

$$\bar{n} = \gamma_\dagger/\kappa, \quad \kappa = \gamma_\downarrow - \gamma_\dagger \quad (3.8)$$

and the label "antinor" (antinormal) implies as usual[2(c)] that a and $a^\dagger$ must be arranged with all the creation operators to the right of the annihilation operators; the functions $L_p^q(x)$ are Laguerre polynomials. The eigenvectors of the Hermitian adjoint Liouvillian are given by

$$\|B_{pq}) = \begin{cases} \gamma_{pq}\left[a^q L_p^q\left(\frac{a^\dagger a}{\bar{n}}\right)\right]_{\text{nor}}, & q \geq 0 \quad (3.9) \\ \gamma_{p|q|}\left[a^{\dagger|q|} L_p^{|q|}\left(\frac{a^\dagger a}{\bar{n}}\right)\right]_{\text{nor}}, & q \leq 0 \quad (3.10) \end{cases}$$

where "nor" (normal) prescribes normal ordering of the creation and annihilation operators. The choice of the constants γ_{pq},

$$\gamma_{pq} = \frac{p!}{(p+|q|)!\bar{n}^{|q|+1}}, \quad (3.11)$$

ensures the orthogonality relation

$$(B_{pq}\|A_{pq}) = \mathrm{Tr}(B_{pq}^\dagger A_{p'q'}) = \delta_{pp'}\delta_{qq'}. \quad (3.12)$$

Finally, the eigenvalues are given by

$$\lambda_{pq} = -\kappa(2p+|q|) - \eta q^2, \quad \begin{matrix} p=0,1,2,\ldots \\ q=0,\pm1,\pm2,\ldots \end{matrix} \quad (3.13)$$

The equation of motion for the diagonal matrix elements in the Born approximation [Eq. (2.21)] is reproduced here for convenience

$$\frac{d}{dt}(W_r(t))_n = (\Lambda_1 W_r(t))_n + \int_0^t d\tau \sum_{m=0}^{\infty} (W_r(\tau))_m \left(\sum_{p,q} e^{\lambda_{pq}(t-\tau)} (L_c A_{pq})_n (L_c B_{pq}^\dagger)_m\right). \quad (3.14)$$

The evaluation of the matrix elements $(L_c A_{pq})_n$ and $(L_c B_{pq}^\dagger)_m$ is algebraically cumbersome but straightforward; the details are summarized in Appendix B. Here we mention that the only non-zero diagonal matrix elements of both operators $L_c A_{pq}$ and $L_c B_{pq}$ correspond to arbitrary values of the index p and to $q=\pm1$. Furthermore, the following symmetry relations hold:

$$\begin{aligned} (L_c A_{p,-1})_n &= -(L_c A_{p1})_n, \\ (L_c B_{p,-1}^\dagger)_n &= -(L_c B_{p1}^\dagger)_n. \end{aligned} \quad (3.15)$$

The results derived in Appendix B [Eqs. (B.4), (B.9), and (B.10)] can be used to express the integrand of Eq. (3.14) in the form

$$\begin{aligned} &\sum_{m=0}^{\infty} (W_r(\tau))_m \tilde{P}_{mn}(t-\tau) \\ &= \sum_{m=0}^{\infty} (W_r(\tau))_m 2\Omega_R^2 \Bigg\{ \sum_{p=0}^{m} e^{\lambda_{p1}(t-\tau)} (-1)^p (p+1) \frac{(1+\bar{n})^p}{\bar{n}^{p+2}} P_p^{m-p,0}\left(\frac{1-\bar{n}}{1+\bar{n}}\right) \\ &\quad \times \left[\left(\frac{\bar{n}}{1+\bar{n}}\right)^{n+1} nF\left(-p,n+1;2;\frac{1}{1+\bar{n}}\right) - \left(\frac{\bar{n}}{1+\bar{n}}\right)^{n+2}(n+1)F\left(-p,n+2;2;\frac{1}{1+\bar{n}}\right)\right] \\ &\quad + \sum_{p=m+1}^{\infty} e^{\lambda_{p1}(t-\tau)} (-1)^m (p+1) \frac{(1+\bar{n})^m}{\bar{n}^{m+2}} P_p^{p-m,0}\left(\frac{1-\bar{n}}{1+\bar{n}}\right) \\ &\quad \times \left[\left(\frac{\bar{n}}{1+\bar{n}}\right)^{n+1} nF\left(-p,n+1;2;\frac{1}{1+\bar{n}}\right) - \left(\frac{\bar{n}}{1+\bar{n}}\right)^{n+2}(n+1)F\left(-p,n+2;2;\frac{1}{1+\bar{n}}\right)\right] \Bigg\}, \end{aligned} \quad (3.16)$$

where $p_l^{\alpha,\beta}(x)$ are Jacobi polynomials of the indicated argument and $F(a,b;c;x)$ is the hypergeometric

function. The complicated structure of the generalized transition coefficients is, in part, a consequence of the role played by the off-diagonal matrix elements on the evolution of the system and, in part, of the finite temperature of the reservoir. In general, aside from the possible simplification induced by the Markoff approximation (if applicable), it appears that the structure of the generalized transition coefficients cannot be clarified any further.

Here, for the sake of algebraic simplicity, we confine our attention to a zero temperature bath ($\bar{n} \to 0$). A careful analysis of the second sum ($m+1 \leq p < \infty$) in Eq. (3.16) reveals that every term vanishes in the limit $\bar{n} \to 0$. The first sum, instead, reduces to the much simpler looking result

$$\sum_{m=0}^{\infty} (W_r(\tau))_m \tilde{P}_{mn}(t-\tau) \xrightarrow[\bar{n}\to 0]{} -2\Omega_R^2 \sum_{m=0}^{\infty} (W_r(\tau))_m \sum_{p=0}^{m} e^{\lambda_{p1}(t-\tau)} (-1)^{n+p} \frac{(p+1)^2 m!}{n!\,(m-p)!\,(p-n+1)!}. \tag{3.17}$$

The main steps leading to Eq. (3.17) are summarized in Appendix C.

Not all terms should be kept in the double sum on the right-hand side of Eq. (3.17) because of the appearance of negative factorials in the denominator. It is easy to see that, depending on the value of n, the appropriate way to write Eq. (3.17) is

$$\sum_{m=0}^{\infty} (W_r(\tau))_m \tilde{P}_{mn}(t-\tau) = -2\Omega_R^2 \sum_{\substack{m=n-1\,(n\geq 1)\\ m=0\,(n=0)}}^{\infty} (W_r(\tau))_m \sum_{\substack{p=n-1\,(n\geq 1)\\ p=0\,(n=0)}}^{m} e^{\lambda_{p1}(t-\tau)} (-1)^{n+p} \frac{(p+1)^2 m!}{n!\,(m-p)!\,(p-n+1)!}, \tag{3.18}$$

where the lower limit of the sums are $m(p)=0$ or $m(p)=n-1$, depending on whether $n=0$ or $n \geq 1$, respectively. After substituting Eq. (3.18) into (3.14), the population equations in the Born approximation take the form

$$\frac{d(W_r)_n}{dt} = (\Lambda_1 W_r)_n - 2\Omega_R^2 \int_0^t d\tau \sum_{\substack{m=n-1\\ (m=0)}} (W_r(\tau))_m \sum_{\substack{p=n-1\\ (p=0)}}^{m} e^{\lambda_{p1}(t-\tau)} (-1)^{n+p} \frac{(p+1)^2 m!}{n!\,(m-p)!\,(p-n+1)!}. \tag{3.19}$$

The validity of this approach is predicated on the strength of the driving force being such that the associated Rabi frequency Ω_R is much smaller than the incoherent decay rates κ or η (or both). It is clear that, as long as κ and η are of comparable magnitude, the population equations form a set of coupled integro-differential equations whose solution is a cumbersome task, even by numerical methods. This is the price one has to pay for the reduction of the original set of N^2 coupled equations for the matrix elements of W (actually, N^2-1, if one takes the trace condition explicitly into account) to the much smaller set of N population equations.[11]

A curious feature of this problem is that the structure of Eq. (3.19) allows the derivation of a simple differential equation for the average excitation $\langle n(t)\rangle \equiv \mathrm{Tr}[a^\dagger a W_r(t)]$, as one can check with a few algebraic manipulations. The result is

$$\frac{d}{dt}\langle n\rangle = \sum_{n=0}^{\infty} n(\Lambda_1 W_r)_n + 2\Omega_R^2 \int_0^t d\tau\, e^{-(\kappa+\eta)\tau}$$

$$= \sum_{n=0}^{\infty} n(\Lambda_1 W_r)_n + \frac{2\Omega_R^2}{\kappa+\eta}\left(1 - e^{-(\kappa+\eta)t}\right), \tag{3.20}$$

where the first term on the right-hand side is identical to the right-hand side of Eq. (3.5) in the zero-temperature limit.

IV. POPULATION EQUATIONS IN THE MARKOFF APPROXIMATIONS

A significant simplification can be obtained if the relaxation rate of the off-diagonal matrix elements is much faster than that of the diagonal ones ($\eta \gg \kappa$). In this case, the integrand of the population equations (3.14) consists of the product of a rapidly varying exponential function and a slowly varying one. Application of the Markoff approximation in the usual way leads to the much simpler set of coupled differential equations

$$(\dot{W}_r)_n = (\Lambda_1 W_r)_n + \sum_{\substack{m=n-1\\ (m=0)}}^{\infty} (W_r)_m P_{mn}, \tag{4.1}$$

where the transition rates P_{mn}, to lowest order in the small parameter $\epsilon = \kappa/\eta$, are given by

$$P_{mn} = -\frac{2\Omega_R^2}{\eta} \sum_{\substack{p=n-1\\ (p=0)}}^{m} (-1)^{n+p} \frac{(p+1)^2}{n!\,(p-n+1)!} \frac{m!}{(m-p)!}$$

$$= \frac{2\Omega_R^2}{\eta} \begin{cases} n, & m = n-1 \\ -(2n+1), & m = n \\ n+1, & m = n+1 \\ 0, & \text{otherwise} \end{cases}. \tag{4.2}$$

Hence, only nearest-neighbor levels are coupled together, and the rate equations become

$$(\dot{W}_r)_n = (\Lambda_1 W_r)_n + \frac{2\Omega_R^2}{\eta}[nW_{n-1} - (2n+1)W_n + (n+1)W_{n+1}]. \quad (4.3)$$

Interestingly, the action of the driving field on the oscillator is equivalent, in this approximation, to raising the temperature of the reservoir from $T=0$ (as chosen in this example) to a nonzero value that depends on the magnitude of the Rabi frequency.

A comparison between the time dependence of the exact occupation probabilities and that of the solutions of Eq. (4.3) is provided in Figs. 1 and 2. The agreement is rather satisfactory, as one can see, when the conditions for the Born and Markoff approximation are well satisfied.

V. CONCLUSIONS

We have explored the possibility of deriving, more or less from first principles [if the master equation (1.1) can be viewed as a microscopic equation of motion for the system of interest], coupled equations for the occupation probabilities. We have shown that under well-defined sufficient conditions involving the strength of the off-diagonal coupling mechanism and the rate of irreversible relaxation, population equations can be easily derived (at least formally) using an elegant procedure due to Weidlich. Our method displays the structure of the generalized transition coefficients that govern the population evolution and that contains information on memory effects. The simple example of a driven-damped harmonic oscillator has been discussed in detail to clarify the relations among the different time scales.

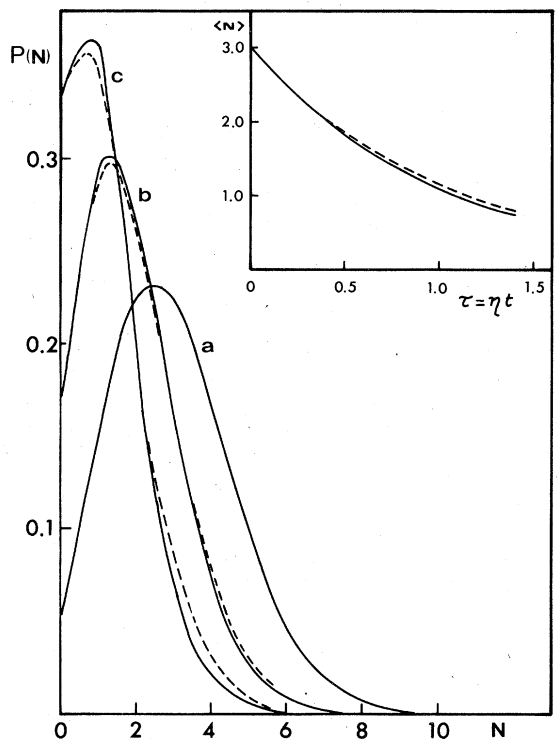

FIG. 1. Population evolution for a driven-damped harmonic oscillator and time dependence of the average occupation number $\langle a^\dagger a(t)\rangle$ (inset). The solid lines connect the discrete values $(W_r)_n$ $(n=0,1,\ldots)$ obtained from the exact numerical solution of the master equation (1.1) with the coherent Liouvillian (3.1) and the dissipative terms [(3.2) and (3.3)]. The dashed lines connect the solution of Eq. (4.3). The relevant parameters are $\Omega_R/\eta=0.2$, $\kappa/\eta=0.5$. Curve (a) represents an initial Poisson distribution with average value $\bar{n}=3.0$. Curves (b) and (c) corresponds to the times $\tau=0.5$ and 0.1, respectively.

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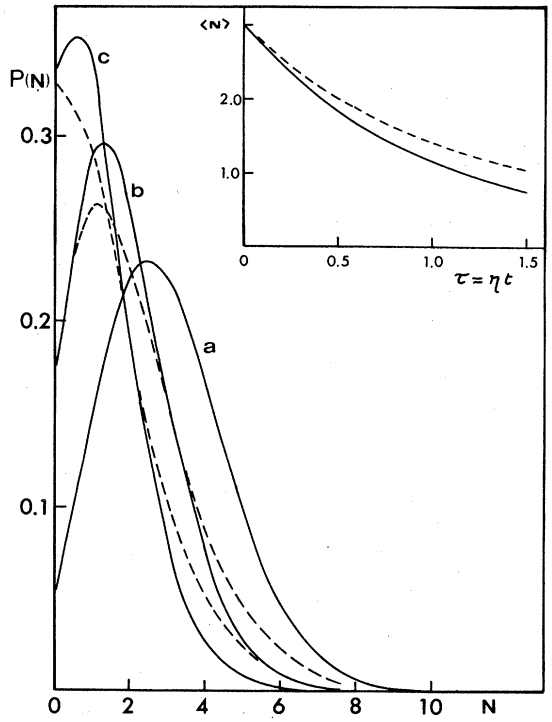

FIG. 2. Same as Fig. 1, except that $\Omega_R/\eta=0.5$. The quantitative agreement between exact and approximate solution suffers from the fact that the Born approximation is no longer well satisfied.

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APPENDIX A: DRIVEN-DAMPED TWO-LEVEL SYSTEM

As an example of a different method for calculating the diagonal matrix elements in Eq. (2.14), we consider the simple case of a driven-damped two-level system.

The master equation for the reduced atomic density operator has the same structure as Eq. (2.1), with

$$\begin{aligned} L_c W &= \Omega_R[S^+ + S^-, W], \\ \Lambda_1 W &= \kappa([S^-, WS^+] + [S^- W, S^+]), \\ \Lambda_2 W &= \eta([S_z, WS_z] + [S_z W, S_z]), \end{aligned} \tag{A1}$$

where Ω_R is the usual Rabi frequency (essentially the amplitude of the driving electric field) and the damping rates are related to the longitudinal and transverse relaxation rates $\gamma_\parallel, \gamma_\perp$ by

$$\kappa = \frac{\gamma_\parallel}{2}, \quad \eta = \gamma_\perp - \frac{\gamma_\parallel}{2}. \tag{A2}$$

The operators $S^\pm, S_z$ are angular momentum operators corresponding to $S = 1/2$.

Once again, the required rate equation can be derived from the general result

$$(\dot{W}_r)_n = (\Lambda_1 W_r)_n - \int_0^t d\tau \langle n | L_c e^{(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r(\tau) | n \rangle, \tag{A3}$$

provided $\Omega_R \ll \kappa, \eta$. The index n labels the states $|\pm\rangle$ of the two-level system. The explicit evaluation of the diagonal matrix elements in Eq. (A3) can be carried out as follows. We define

$$\begin{aligned} \chi(t,\tau) &\equiv e^{(\Lambda_1+\Lambda_2)(t-\tau)} L_c W_r(\tau), \\ \chi(\tau,\tau) &\equiv L_c W_r(\tau). \end{aligned} \tag{A4}$$

From the definition of $L_c W$ we have

$$\begin{aligned} &\langle + | L_c \chi(t,\tau) | + \rangle \\ &\quad = \Omega_R[\langle - | \chi(t,\tau) | + \rangle - \langle + | \chi(t,\tau) | - \rangle] \\ &\quad = -\langle - | L_c \chi(t,\tau) | - \rangle. \end{aligned} \tag{A5}$$

The matrix elements $\langle + | \chi(t,\tau) | - \rangle$ and $\langle - | \chi(t,\tau) | + \rangle$ can be evaluated with the help of their equations of motion. Thus, we have for example,

$$\begin{aligned} \frac{d}{dt}\langle + | \chi(t,\tau) | - \rangle &= \langle + | (\Lambda_1 + \Lambda_2)\chi(t,\tau) | - \rangle \\ &= -(\kappa + \eta)\langle + | \chi(t,\tau) | - \rangle, \end{aligned} \tag{A6}$$

where the last step follows from the definition of Λ_1 and Λ_2. In this case, it is especially simple to solve the equations of motion. For the harmonic oscillator model discussed in Sec. III, the procedure is more cumbersome but still usable. The integration of Eq. (A6) leads to

$$\begin{aligned} \langle + | \chi(t,\tau) | - \rangle &= e^{-(\kappa+\eta)(t-\tau)} \langle + | L_c W_r | - \rangle \\ &= e^{-(\kappa+\eta)(t-\tau)} \Omega_R (\langle - | W_R | - \rangle - \langle + | W_r | + \rangle). \end{aligned} \tag{A7}$$

The population equations follow at once:

$$\begin{aligned} \frac{d}{dt}\langle + | W_r | + \rangle &= (\Lambda_1 W_r)_+ \\ &\quad + \int_0^t d\tau\, 2\Omega_R^2 e^{-(\kappa+\eta)(t-\tau)}[(W_r)_- - (W_r)_+], \\ \frac{d}{dt}\langle - | W_r | - \rangle &= -\frac{d}{dt}\langle + | W_r | + \rangle. \end{aligned} \tag{A8}$$

With a little more effort, one can apply this procedure to a multilevel atom model with equal level spacing.

APPENDIX B: EXPLICIT EVALUATION OF THE DIAGONAL MATRIX ELEMENTS $(L_c A_{pq})_n$ AND $(L_c B_{pq}^\dagger)_n$

In this appendix we outline the derivation of explicit formulas for the diagonal matrix elements of $L_c A_{pq}$ and $L_c B_{pq}^\dagger$. We consider first the operator $L_c A_{pq}$.

The antinormal ordered form of the operators A_{pq} makes it possible to use the Glauber coherent state representation according to the prescription

$$\begin{aligned} (L_c A_{pq})_n &= \Omega_R \left\langle n \left| \frac{\partial A_{pq}}{\partial \alpha^\dagger} - \frac{\partial A_{pq}}{\partial \alpha} \right| n \right\rangle \\ &= \Omega_R \int \frac{d^2\alpha}{\pi} e^{-|\alpha|^2} \frac{|\alpha|^{2n}}{n!} \left(\frac{\partial \mathcal{A}_{pq}}{\partial \alpha^*} - \frac{\partial \mathcal{A}_{pq}}{\partial \alpha} \right), \end{aligned} \tag{B1}$$

where $\mathcal{A}_{pq}(\alpha_1\alpha)$ is the c-number representative of the operator A_{pq} and $d^2\alpha \equiv d(\mathrm{Re}\alpha)d(\mathrm{Im}\alpha)$. We must consider the two cases $q \geq 0$ and $q \leq 0$ separately. For positive integer (or zero) values of the index q, Eq. (B1) can be cast into the form

$$\begin{aligned} (L_c A_{pq})_n = \Omega_R \int \frac{d^2\alpha}{\pi} e^{-|\alpha|^2/\bar{n}} \alpha^q L_p^q\left(\frac{|\alpha|^2}{\bar{n}}\right) \\ \times \left(\frac{\partial}{\partial \alpha} - \frac{\partial}{\partial \alpha^*} \right) e^{-|\alpha|^2} \frac{|\alpha|^{2n}}{n!}. \end{aligned} \tag{B2}$$

After transforming to polar coordinates, the integral becomes

$$(L_c A_{pq})_n = \Omega_R \int \frac{d\rho\, d\theta}{\pi} e^{-[(1+\bar{n})/\bar{n}]\rho^2} L_p^q\left(\frac{\rho^2}{\bar{n}}\right) \rho^{q+2} \left(\frac{\rho^{2(n-1)}}{(n-1)!} - \frac{\rho^{2n}}{n!} \right) e^{i(1-q)\theta}. \tag{B3}$$

It is now a simple matter to show that

$$(L_c A_{pq})_n = \Omega_R \left[\left(\frac{\bar{n}}{1+\bar{n}} \right)^{n+1} n(p+1) F\left(-p, n+1; 2; \frac{1}{1+\bar{n}} \right) - \left(\frac{\bar{n}}{1+\bar{n}} \right)^{n+2} (n+1)(p+1) F\left(-p, n+2; 2; \frac{1}{1+\bar{n}} \right) \right] \delta_{q,1}, \tag{B4}$$

where $F(a,b;c;x)$ is the hypergeometric function.

It is also simple to show that for negative values of the index q, the only nonzero matrix elements correspond to arbitrary values of p and $q=-1$. Furthermore, we have

$$(L_c A_{p,-1})_n = -(L_c A_{p1})_n . \tag{B5}$$

The required elements of the operators $L_c B^\dagger_{pq}$ can be evaluated as follows. From the identity

$$L_c B^\dagger_{pq} = \Omega_R [a^\dagger + a, B^\dagger_{pq}] = \Omega_R \left(\frac{\partial B^\dagger_{pq}}{\partial a^\dagger} - \frac{\partial B^\dagger_{pq}}{\partial a} \right) \tag{B6}$$

and the definition of $B^\dagger_{pq}$ [Eqs. (3.9) and (3.10)], it follows at once that $(\partial B^\dagger_{pq}/\partial a)_n = 0$ for every positive value of q. Furthermore, one can show that

$$\left(\frac{\partial B^\dagger_{p1}}{\partial a^\dagger} \right)_n = \frac{1}{(p+1)\bar{n}} \sum_{j=0}^{p} \frac{(-1)^j}{j!} \binom{p+1}{p-j} \frac{1}{\bar{n}^j} (j+1)(a^{\dagger j} a^j)_n , \tag{B7}$$

and that $(\partial^\dagger_{pq}/\partial a^\dagger)_n$ for $q>0$, $q \neq 1$ is identically equal to zero.

From Eqs. (B6) and (B7), one can readily arrive at the required result

$$(L_c B^\dagger_{p1})_n = \frac{1}{\bar{n}^2} \Omega_R \sum_{j=0}^{p} \left(-\frac{1}{\bar{n}} \right)^j \frac{p!}{j!(p-j)!} \frac{n!}{j!(n-j)!} . \tag{B8}$$

It is possible to express the finite sum on the right-hand side of Eq. (B8) in terms of Jacobi polynomials, provided we distinguish two cases:

(a) For $n \geq p$ we find

$$(L_c B^\dagger_{p1})_n = \frac{\Omega_R}{\bar{n}^2} (-1)^p \left(\frac{1+\bar{n}}{\bar{n}} \right)^p P_p^{n-p,0} \left(\frac{1-\bar{n}}{1+\bar{n}} \right), \tag{B9}$$

(b) For $p \geq n$, the matrix elements become

$$(L_c B^\dagger_{p1})_n = \frac{\Omega_R}{\bar{n}^2} (-1)^n \left(\frac{1+\bar{n}}{\bar{n}} \right)^n P_n^{p-n,0} \left(\frac{1-\bar{n}}{1+\bar{n}} \right). \tag{B10}$$

The matrix elements corresponding to negative values of q are all identically zero, except for $q=-1$. In this case, the following symmetry relation holds:

$$(L_c B^\dagger_{p,-1})_n = -(L_c B^\dagger_{p1})_n . \tag{B11}$$

APPENDIX C: DERIVATION OF EQ. (3.17)

In carrying out the low-temperature limit in Eq. (3.16), some care must be exercised in identifying the leading terms of the various $\bar{n}$-dependent contributions. The leading term of each hypergeometric function can be calculated as follows. Starting from the identity

$$F(-p,2;2;z) = (1-z)^p \tag{C1}$$

and the well known function relation

$$F(-p,n+2;2;z) = \frac{1}{(2)_n z} \frac{d^n}{dz^n} [z^{n+1} F(-p,2;2;z)], \tag{C2}$$

it is simple to prove the equality

$$F(-p,n+2;2;z) = \frac{1}{(2)_n z} \sum_{k=0}^{n} \binom{n}{k} \frac{(n+1)!}{(n-k+1)!} \frac{p!}{(p-n+k)!} (-1)^{n-k} z^{n-k+1} (1-z)^{p-n+k} . \tag{C3}$$

The limits of interest have the general form

$$\lim_{\bar{n}\to 0} \bar{n}^{l-m} F\left(-p, l+2; 2; \frac{1}{1+\bar{n}} \right) = \lim_{\bar{n}\to 0} \frac{1}{(2)_l} \sum_{k=0}^{l} \binom{l}{k} \frac{(l+1)!}{(l-k+1)!} \frac{p!}{(p-l+k)!} (-1)^{l-k} \bar{n}^{p+k-m}, \quad l = 0,1,2,\ldots \tag{C4}$$

Clearly, if $p = m+1, m+2\ldots$, the result of the limit is identically zero. When $p \leq m$, the dominant terms can be readily identified with the help of Eq. (C4), with the result

$$\lim_{\bar{n}\to 0} \tilde{P}_{m,n}(t-\tau) = 2\Omega_R^2 \sum_{p=0}^{m} e^{\lambda_{p1}(t-\tau)} (p+1)(-1)^p \binom{m}{p} \left(\frac{n}{(2)_{n-1}} \frac{p!}{p-n+1} (-1)^{n-1} - \frac{n+1}{(2)_n} \frac{p!}{(p-n)!} (-1)^n \right). \tag{C5}$$

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