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# Two level dipolar system in a heat bath

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## Abstract

Using a theory developed by Fulinsky and Kramarcyk, combined with weak coupling limit asymptotic perturbative expansion we obtain rate equations for the reduced density matrix of a two level system immersed in a heat bath. **PACS**: 31.70.Dk; 33.15.Kr

**Key words:** memoryless master equations; weak-coupling limit; two-level systems.

# Sistema dipolar de dos niveles en un baño térmico

## Resumen

Usando una teoría desarrollada por Fulinski y Kramarcyk, combinada con un desarrollo perturbativo en el límite de acoplamiento débil, obtenemos la ecuación de evolución para la matriz densidad reducida de un sistema de dos niveles inmerso en un baño térmico. **PACS**: 31.70.Dk; 33.15.Kr

**Palabras clave:** Ecuaciones maestras sin memoria; límite de acoplamiento débil; sistemas de dos niveles.

## Introduction

The study of stochastic effects on the interaction of light with a two-level system has attracted considerable interest. Many experimental as well as theoretical studies have done in an attempt to quantify the ubiquitous quantum fluctuations (1-4). Thus, Mollow (1) carried out fluorescence experiments and found that the emitted spectrum was very sensitive to the stochastic fluctuations of the pumping field. Various theoretical models have been developed to account for this effect (2, 3). These include the so-called phase diffusion model (2), in which only the field phase fluctuates, the chaotic field model (3), where the fields amplitude was modeled as a two-dimensional complex Gaussian process, and jump models (4), where the amplitude phase or frequency of the field is taken as a discontinuous Markov process. In other words, the dynamic study of many systems, have been treated and/or characterized by an intermediate mesoscopic description, where the important quantities are represented by random variables. The rate of change of the joint probability distribution of these variables is given in terms of the so-called master equation (ME).

The master equation (ME) technique is a time honored technique to obtain reduced and/or decoupled dynamic equations for complex or large systems. In (5) and (6) the reader can find considerable description of

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the original work and ideas. One interesting line of research was initiated in (7)-(8) and applied to the simple Friedrichs model in quantum field theory, which consists of a procedure to obtain memoryless ME, that is, reduced equations in which the rate terms do not depend on the previous history of the system. In (9) this line of work was suplemented with perturbative expansion techiques and asymptotic limits of the weak coupling type, and it was shown that some of the typical ME models can be so obtained. The theory of limits of the weak coupling type was well developed by then, see (10)-(11) for example.

These reduced density matrix equations, are very important in quantum optics, when is necessary the optics response of a molecular system, interacting with a thermal bath and radiation field. Cases of ME as the Nakajima-Swanzig equations, have been employed in the literature for the dynamic study of the reduce density matrix. In this case, is very important the memory kernel associated to the system-bath correlation, and the information term. We should mention that a Hamiltonian similar to [2] appear as reduced Hamiltonian to describe tunneling in a double-well potential, when the system is coupled to dissipative environment, and when transitions to and from higher energy states can be neglected. A comprehensive review of this problem appears in chapter 18 of reference (12), but the techniques described there are quite different from ours.

## The asymptotic limit of the reduced equations

Consider to begin with a two-level system with Hamiltonian operator

$$H = \overline{E}_1 |1)(1| + \overline{E}_2 |2)(2| + \mathbf{m}\cdot\mathbf{H} \qquad [1]$$

describing a simple system interacting with a dipole. If a usual, we orient the z-axis along the polarization field **H** and denote the components of $m_z$ by $\mu_{ij}$ the Hamiltonian becomes

$$\tilde{H} = \overline{E}_1 |1)(1| + \overline{E}_2 |2)(2| + \delta\{|1)(2| + |2)(1|\} \qquad [2]$$

where $\delta = \mu_{12}H = \mu_{21}H$ represents the strength of the dipole coupling, say. Also, $E_1 = \overline{E}_{11} + \mu_{11}H$ and $E_2 = \overline{E}_{12} + \mu_{22}H$ represent the shifted energies of the two levels of interest. It is an elementary exercise in matrix diagonalization, to see that the eigenvalues of $\tilde{H}$ are given by

$$\varepsilon_0 = \frac{E_1 + E_2}{2} - \left(\Delta^2 + \delta^2\right)^{1/2},$$
$$\varepsilon_1 = \frac{E_1 + E_2}{2} + \left(\Delta^2 + \delta^2\right)^{1/2} \qquad [3]$$

where $\Delta = \dfrac{E_1 - E_2}{2}$, and the new basis vectors are

$$|0\rangle = -\frac{\delta}{\left(\delta^2 + (\Delta - \Theta)^2\right)^{1/2}}|1\rangle + \frac{\Delta - \Theta}{\left(\delta^2 + (\Delta - \Theta)^2\right)^{1/2}}|2\rangle \qquad [4]$$

and

$$|1\rangle = -\frac{\delta}{\left(\delta^2 + (\Delta + \Theta)^2\right)^{1/2}}|1\rangle + \frac{\Delta + \Theta}{\left(\delta^2 + (\Delta + \Theta)^2\right)^{1/2}}|2\rangle \qquad [5]$$

where $\Theta = \left(\Delta^2 + \delta^2\right)^{1/2}$.

Now we want to study the effect of a heat bath on this system. In the basis in which the Hamiltonian of the system is diagonal, the full Hamiltonian of the system plus heat bath will be

$$H = H_0 + H_1 = \varepsilon_0 |0\rangle\langle 0| + \varepsilon_1 |1\rangle\langle 1| +$$
$$\sum_N |\omega_N\rangle\langle\omega_N| + \sum_{i,n} V_{i,n}\left(|(i+1)_2, \omega_N\rangle\langle\omega_{n,i}| + |i, \omega_n\rangle\langle\omega_n, (i+1)_2|\right) \qquad [6]$$

where $(i+1)_2$ denotes sum modulo 2, and of course, since our system is composite system, to be proper we should have written

$$\varepsilon_0 |0\rangle\langle 0| + \varepsilon_1 |1\rangle\langle 1| = \left(\varepsilon_0 |0\rangle\langle 0| + \varepsilon_1 |1\rangle\langle 1|\right) \otimes \sum\nolimits_n |\omega_n\rangle\langle\omega_n|$$
$$= \sum\nolimits_N \left(\varepsilon_0 |0, \omega_n\rangle\langle 0, \omega_n| + \varepsilon_1 |1, \omega_n\rangle\langle 1, \omega_n|\right) \qquad [7]$$

and similarly for the other term in $H_0$. The evolution equation for the density matrix $\rho(t)$ of this system is the standard

$$i\frac{d\rho}{dt} = L\rho \equiv [\mathrm{H}, \rho] \qquad [8]$$

Mathematically speaking, our problem consists of finding an evolution equation for $\rho_d(t) = P_\rho(t)$, where $PA$ is the projection operator on the "relevant" or "diagonal" part of $A$. That is if then $A = \sum_{i,n} A_{i,j;n,m} |i, \omega_n\rangle\langle\omega_m, j|$

$$P(A) = \sum_{i,j} A_{ij} |i\rangle\langle j| \equiv \sum_{i,j} tr\left(|i\rangle\langle j|A|\right)|i\rangle\langle j| \qquad [9]$$

or perhaps a bit more explicitly $\hat{A}_{i.j} = \sum_N A_{i,j;n,n}$. If we denote by $U(t) = e^{tL}$ the propagator or time evolution operator associated with [8], it was proved in (2) or (6) that $\rho_d(t)$ satisfies the equation

$$\frac{d\rho_d(t)}{dt} = R(t)\rho_n(0) + R(t)\rho_d(t) \qquad [10]$$

where $R(t) = \dot{N}(t)N(t)^{-1}$ and where $N(t) \equiv Q + P(U(t) - \mathrm{I})$ which is invertible, and $Q = \mathrm{I} - P$. If we write $H = H_0 + \lambda H_I$, we can obtain a perturbative expansion for $G(t) \equiv PR(t)P = R(t)P$ of the form

$$G(t) = PL_0P + \lambda G_1(t) + \lambda G_2(t) + \ldots \qquad [11]$$

where $L_0 = [H_0, \cdot]$ and $L_1 = [H_1, \cdot]$ with $G_1(t) \equiv PL_1P$, and

$$G_2(t) \equiv PL_1Q\int_0^1 U_o(s)QL_1PU_0(-s)ds,$$

and more elaborate expressions for the following terms of the expansion. There $U_0(t) = e^{tL_0}$ is the evolution operator induced by $L_0$. The theory of weak coupling limits (see (10)-(11)) asserts that small and appropriately stretched out time scale, the asymptotic behavior of $\rho_d(t)$ happens to be determined by the averaged Liouville (or Hamiltonian) operator

$$\bar{L} = PL_0P + \lambda PL_1P \qquad [12]$$

and now we can stop referring to the perturbative constant $\lambda$. After some simple, but somewhat tedious computations we obtain

$$PL_0P\rho = P[H_{0.}, P\rho] = \sum_{i,j=0}^{1}\left(\varepsilon_i - \varepsilon_j\right)\hat{\rho}_{i.j}|i\rangle\langle j| \qquad [13]$$

with the understanding that as operator on the product space, the right hand side must be multiplied by the identity operator $\sum_n |\omega_n\rangle\langle\omega_n|$. Also

$$PL_1P\rho = P[H_I, P\rho] = \sum_{i,j}\hat{V}_i\left(\hat{\rho}_{(i+1)2,j}|i\rangle\langle j| - \hat{\rho}_{i.j}|(i+1)_2\rangle\langle j|\right)$$
$$-\sum_{i,j}\hat{V}_i\left(\hat{\rho}_{j,i}|j\rangle\langle(i+1)_2| - \hat{\rho}_{j,(i+1)_2}|j\rangle\langle i|\right) \qquad [14]$$

After equating coefficients of the same basis vectors, we obtain the following system of equations for the density matrix $\hat{\rho}$

$$i\frac{d\hat{\rho}_d}{dt} = i\frac{d}{dt}\begin{pmatrix}\hat{\rho}_{00}\\ \hat{\rho}_{01}\\ \hat{\rho}_{10}\\ \hat{\rho}_{11}\end{pmatrix} = \begin{pmatrix}0 & -W & W & 0\\ -W & -\Delta & 0 & W\\ W & 0 & -\Delta & -W\\ 0 & W & -W & 0\end{pmatrix}\begin{pmatrix}\hat{\rho}_{00}\\ \hat{\rho}_{01}\\ \hat{\rho}_{10}\\ \hat{\rho}_{11}\end{pmatrix} = \hat{L}\hat{\rho}_d \qquad [15]$$

where $\Delta \equiv \varepsilon_1 - \varepsilon_0$ and $W \equiv \hat{V}_0 + \hat{V}_1$, and of course $V_i = \sum_n V_{i,n}$. To integrate this system it is easier to begin by finding its eigenvalues and eigenvectors. The eigenvalues are

$$\lambda_1 = -\Delta;\ \gamma_2 = a_+;\ \lambda_3 = a_-;\ \lambda_4 = 0$$

where $a_\pm = -\frac{\Delta}{2} \mp \frac{1}{2}\left(\Delta^2 + 16W^2\right)^{1/2}$ and the corresponding un-normalized (transposed) eigenvectors are

$$v_1 = (0,1,1,0);\ v_2 = \left(-1, \frac{a_+}{2W}, -\frac{a_+}{2W}, 1\right).$$

$$v_3 = \left(-1, \frac{a_-}{2W}, -\frac{a_+}{2W}, 1\right);\ v_4(1,0,0,1).$$

which can easily seen to be orthogonal. After normalization, the $\hat{\rho}_{ij}(t)$ can be explicitly computed once initial data is supplied.

With this information relative to the reduced density matrix, it is possible the determination of important fundamental properties. For this, we have considered a macroscopic system, consisting of N nonintereacting dipoles, immersed in the heat bath, and under the action of the same polarization field, where the average moment is given by $M(t) = Ntr(m_z\hat{\rho}(t))$. Notice that the trace being basis independent can be computed in any basis. Thus in order to compute $M(t)$, once [15] is integrated, we can either express $\sum_{i,j=0}^{1}\mu_{ij}|i)(j|$ in terms of the (normalized) basis $v_i$ displayed above, or write that basis, and the matrix $\hat{\rho}_d$ in terms of the $|i)(j|$, and carry out the computation. In the present model, the intensity of any nonlinear signal in the local approximation is related to the quantity $M$, given by $I \propto M(t)^2$.

## Final comments

The model developed in the present study represents an alternative of the conventional local models proposed to date in the literature. The explicit inclusion of nonzero molecular dipole moments in the present model is a necessary and sufficient condition for the generation of nonlinear optical signals. Details concerning the modification of the response by including propagation of the field in a medium material, and inclusion of the fields with temporal dependence, are to be published elsewhere.

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