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Relationship between the adiabatic-following solutions of Bloch equations and the solutions of non-Markovian Pauli-type master equations

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A general derivation of non-Markovian Pauli-type master equations is given for systems interacting with pump and relaxation mechanisms. It is shown that the adiabatic-following solutions of Bloch equations, due to Grischkowski and co-workers, are obtained as a special case from Pauli equations of non-Markovian type. Finally, it is proved that for a two-level system the non-Markovian master equation obtained in the Born approximation is also exact.

The coupled Maxwell-Bloch equations are known to describe a wide variety of optical resonance phenomena[1,2] in two-level atoms, depending on the nature of relaxation parameters, pulse shapes, pulse widths, pulse coherence time, detunings, inhomogeneous broadening, etc. For example, for the case in which the pulse width is very long compared to the transverse relaxation time T_2 or even when the coherence time is much shorter compared to T_2, then the Bloch equations can be reduced to a single equation for the inversion, and this equation (which is now in the form of the rate equation[3,4]) involves the intensity of the field. In such a case, the equations describe the incoherent interaction of the pulse with a medium of two-level atoms. The rate equations have been used extensively in quantum optics to study a variety of incoherent interactions.[5,6] When the applied fields are close to resonance, the rate equation approximation breaks down. Improved solutions, which correspond to adiabatic following, have been obtained by Grischkowski and others.[7-9]

The purpose of this paper is to analyze the adiabatic-following solutions from the point of view of master equations.[10] It will be shown that the non-Markovian Pauli-type of master equations admit, in special cases, both rate-equation and adiabatic-following solutions. It is also shown that the master equations obtained in the Born approximation, for diagonal elements of a two-level systems, are *exact*. The formulation is applicable to multilevel systems, interacting with relaxation and pump mechanisms, and provides a systematic as well as rigorous way to arrive at the adiabatic-following solutions.

It is known from the work of Zwanzig[11,12] that the diagonal matrix elements of ρ of a system characterized by the Hamiltonian $H=H_0+H_1$, in the representation in which H_0 is diagonal, satisfy a Pauli-type master equation:

$$\frac{\partial \rho_{nn}}{\partial t}+\int_0^t d\tau \sum_m [R_{mn}(t-\tau)\rho_{nn}(\tau) - R_{nm}(t-\tau)\rho_{mm}(\tau)]=0, \quad (1)$$

where $R_{mn}(t)$ is the total transition probability that the system makes a transition from the level $|n\rangle$ to the level $|m\rangle$. Because of the non-Markovian character, (1) differs from the usual Pauli master equation or the rate equation. The Pauli master equation is obtained from (1) in the limit that the behavior of the system can be regarded as Markovian, whence one can replace $\rho(t-\tau)$ by $\rho(t)$ and extend the limit of integration to infinity.

In optical resonance phenomena, the relaxation parameters play an important role and therefore we will *generalize* (1) to make it *applicable to open systems*. The density matrix ρ of a multilevel system interacting with coherent fields satisfies

$$\dot{\rho}=L_0\rho+L_1(t)\rho, \quad (2)$$

where we assume that the relaxation mechanisms are such that L_0 couples diagonal elements to diagonal elements only and that $L_1(t)$ has no diagonal elements. We also assume that at $t=t_0$, the density matrix ρ is diagonal in the $|n\rangle$ representation. On introducing the projection operator $\mathcal{P}$ defined by[10]

$$\mathcal{P}\ldots=\sum_m \mathcal{P}_m \mathrm{Tr}\{\mathcal{P}_m\ldots\}, \quad \mathcal{P}_m=|m\rangle\langle m|,$$
$$\mathcal{P}G=\sum_m G_{mm}|m\rangle\langle m|, \quad (3)$$

and on using the standard projection operator techniques,[10,11] one can show that diagonal and off-diagonal elements of ρ are given by

$$(1-\mathcal{P})\rho(t)=\int_{t_0}^t d\tau\, e^{L_0 t}\mathfrak{U}(t,\tau) \times e^{-L_0\tau}L_1(\tau)\mathcal{P}\rho(\tau), \quad (4)$$

$$\mathcal{P}\dot{\rho}(t)=L_0\mathcal{P}\rho(t)+\int_{t_0}^{t}d\tau\,\mathcal{P}L_1(t)e^{L_0t}\mathcal{U}(t,\tau)e^{-L_0\tau}\times L_1(\tau)\mathcal{P}\rho(\tau),\tag{5}$$

where

$$\mathcal{U}(t,\tau)=T\exp\left(\int_\tau^t dt'(1-\mathcal{P})e^{-L_0t'}L_1(t')\times e^{L_0t'}(1-\mathcal{P})\right).\tag{6}$$

Equation (4) relates the off-diagonal elements to diagonal elements and Eq. (5) gives the *time evolution of diagonal elements in terms of diagonal elements only*. Because of the assumed properties of L_0, one will have

$$(e^{L_0t}G)_{\alpha\beta}=\sum_{mn}X_{\alpha\beta mn}(t)G_{mn},\tag{7}$$

with

$$X_{\alpha\beta mn}(t)=C_{\alpha m}(t)\delta_{\alpha\beta}\delta_{mn}+(1-\delta_{\alpha\beta})(1-\delta_{mn})D_{\alpha\beta mn}(t).\tag{8}$$

The master equations of the type in Eq. (5) are difficult to handle except in special cases. For a two-level system we will show that $\mathcal{U}(t,\tau)$ is exactly equal to the identity operator. For a multilevel system we will make the Born approximation. Thus the following results will be exact for a two-level system whereas for a multilevel system will hold only approximately:

$$\mathcal{P}\dot{\rho}(t)=L_0\mathcal{P}\rho(t)+\int_0^{t-t_0}d\tau\,\mathcal{P}L_1(t)e^{L_0\tau}\times L_1(t-\tau)\mathcal{P}\rho(t-\tau),\tag{9}$$

$$(1-\mathcal{P})\rho(t)=\int_0^{t-t_0}d\tau e^{L_0\tau}L_1(t-\tau)\mathcal{P}\rho(t-\tau).\tag{10}$$

In order to prove that $\mathcal{U}(t,\tau)=1$ for a two-level system, we consider the Liouville operator $\mathfrak{L}$ acting on any operator A:

$$\mathfrak{L}A=(1-\mathcal{P})L(1-\mathcal{P})A,\tag{11}$$

we write

$$(LB)_{mn}=\sum L_{mnpq}B_{pq},\tag{12}$$

and hence

$$[L(1-\mathcal{P})A]_{\alpha\beta}=\sum L_{\alpha\beta pq}[(1-\mathcal{P})A]_{pq}=\sum L_{\alpha\beta pq}(1-\delta_{pq})A_{pq}.\tag{13}$$

Using (13) we get for the operator $\mathfrak{L}$

$$[(1-\mathcal{P})L(1-\mathcal{P})A]=\sum_{\substack{m\neq n\\ \alpha\neq\beta}}L_{mn\alpha\beta}A_{\alpha\beta}|m\rangle\langle n|.\tag{14}$$

Note that the $\mathcal{U}$ operator involves the Liouville operator of the form (11) with

$$(LA)_{mn}=-i(e^{-L_0t}[H_1(t),(e^{L_0t}A)])_{mn},\tag{15}$$

and hence on comparing (12) and (15), we get

$$L_{mn\alpha\beta}=-i\sum_{pqr}X_{mnpq}(-t)\{[H_1(t)]_{pr}X_{rq\alpha\beta}(t)-[H_1(t)]_{rq}X_{pr\alpha\beta}(t)\}.\tag{16}$$

It is clear from (14) that only elements of L corresponding to $m\neq n$, $\alpha\neq\beta$ are needed and such elements are obtained by substituting (8) in (16):

$$L_{mn\alpha\beta}=-i(1-\delta_{mn})(1-\delta_{\alpha\beta})\sum_{pqr}(1-\delta_{pq})(1-\delta_{pr})(1-\delta_{qr})D_{mnpq}(-t)\times\{[H_1(t)]_{pr}D_{rq\alpha\beta}(t)-[H_1(t)]_{rq}D_{pr\alpha\beta}(t)\},\quad m\neq n,\ \alpha\neq\beta\tag{17}$$

where we have also used the fact that $H_1(t)$ has no diagonal elements. It is quite obvious from (17) and (14) that the operator $\mathfrak{L}$ as defined by (11) is identically zero for a two-level system and hence the evolution operator, as given by (6), is equal to unit operator for a two-level system. The above discussion has been included here as we have shown in the framework of master equations that $\mathcal{U}=1$ without resorting to any indirect methods. This is again very interesting from the viewpoint of master equations for here we have found a class of master equations which are exact in the Born approximation.[13]

On simplification Eq. (9) gives the following non-Markovian master equation of the Pauli type:

$$\dot{\rho}_{\alpha\alpha}=\sum_\beta\langle\alpha|(L_0\mathcal{P}_\beta)|\alpha\rangle\rho_{\beta\beta}-\sum\int_0^{t-t_0}d\tau\{[H_1(t)]_{\alpha m}[H_1(t-\tau)]_{p\beta}X_{m\alpha p\beta}(\tau)\rho_{\beta\beta}(t-\tau)-[H_1(t)]_{m\alpha}[H_1(t-\tau)]_{p\beta}X_{\alpha mp\beta}(\tau)\rho_{\beta\beta}(t-\tau)+\text{H.c.}\}.\tag{18}$$

The usual rate equations are obtained from (18) by making the Markovian approximation. The spirit of this calculation is similar to that of Wilcox and Lamb[4] and Haken,[5] who calculated the off-diagonal elements of the density matrix in terms of diagonal elements by assuming that the off-diagonal elements follow adiabatically the diagonal elements. Here we have done an exact calculation to eliminate the off-diagonal elements [cf. Eqs. (4) and 10)].

If we consider that the relaxation mechanisms are such that off-diagonal elements show a simple behavior (which is generally the case[14])

$$\rho_{ij}(t) = [e^{L_0 t}\rho(0)]_{ij} = e^{-t(\Gamma_{ij}+i\Delta_{ij})}\rho_{ij}(0), \quad i \neq j \tag{19}$$

$$X_{m\alpha p\beta}(t) = \delta_{mp}\delta_{\alpha\beta}Y_{m\alpha}(t), \quad m \neq \alpha,\ p \neq \beta \tag{20}$$

then on substituting (20) in (18) we get the final form of the non-Markovian master equation of the Pauli type:

$$\begin{aligned}\dot{\rho}_{\alpha\alpha}(t) &= \sum_\beta \langle\alpha|L_0 P_\beta|\alpha\rangle \rho_{\beta\beta} - \int_{t_0}^{t} d\tau \sum [R_{m\alpha}(t,\tau)\rho_{\alpha\alpha}(\tau) - R_{\alpha m}(t,\tau)\rho_{mm}(\tau)],\\ R_{m\alpha}(t,\tau) &= [H_1(t)]_{\alpha m}[H_1(\tau)]_{m\alpha}Y_{m\alpha}(t-\tau) + \text{H.c.}\end{aligned} \tag{21}$$

Note that $R_{m\alpha}(t,\tau)$ represents the total transition probability that the system makes a transition from the level $|\alpha\rangle$ to $|m\rangle$. It should also be noted that R, in our case, is a function of the relaxation parameters [cf. Eqs. (19) and (20)].

For a two-level atom (21) leads to (which is an exact result),

$$\begin{aligned}\dot{\rho}_{11} &= \sum_\beta \langle 1|L_0 P_\beta|1\rangle\rho_{\beta\beta} - 2g^2\int_0^{t-t_0} d\tau \mathcal{E}(t)\mathcal{E}(t-\tau)\\ &\quad\times e^{-\Gamma_{12}\tau}\cos(\Delta_{12}\tau)[\rho_{11}(t-\tau) - \rho_{22}(t-\tau)],\end{aligned} \tag{22}$$

and hence the inversion Z satisfies the equation[15] ($\Gamma_{12}=\Gamma$, $\Delta_{12}=\Delta$)

$$\begin{aligned}\dot{Z} &= -\frac{1}{T_1}(Z - Z^{(0)}) - 4g^2\int_0^{t-t_0} d\tau\mathcal{E}(t)\mathcal{E}(t-\tau)\\ &\quad\times e^{-\Gamma\tau}\cos\Delta\tau Z(t-\tau).\end{aligned} \tag{23}$$

The non-Markovian equation (23) has several interesting features, for example, if $\mathcal{E}$ is constant, then it predicts (a) coherent Rabi oscillations in the limit of very large T_1, T_2, (b) the Wigner-Weisskopf type of decay for short T_2. Thus, the non-Markovian equation (23) enables one to study the continuous transition from coherence to incoherence as T_2 is changed. In order to see how the adiabatic solution of Grischkowsky and others follow, we let $t_0 \to -\infty$ so as to avoid the transients and write

$$\mathcal{E}(t-\tau)Z(t-\tau) = \mathcal{E}(t)Z(t) - \tau\frac{\partial}{\partial t}[\mathcal{E}(t)Z(t)] \tag{24}$$

and retain only the first two terms in (24). Then (23) reduces to

$$\begin{aligned}\dot{Z} &= -\frac{1}{T_1}(Z - Z^{(0)}) - \frac{4g^2\Gamma}{\Gamma^2+\Delta^2}\mathcal{E}^2(t)Z(t)\\ &\quad+\frac{4g^2(\Gamma^2-\Delta^2)}{(\Gamma^2+\Delta^2)^2}\mathcal{E}(t)\frac{\partial}{\partial t}[Z(t)\mathcal{E}(t)].\end{aligned} \tag{25}$$

We again note here that the second term constitutes the rate-equation approximation.[2] Equation (23) on simplification leads to

$$\frac{\partial}{\partial t}[\ln(ZX^{1/2})] + \frac{4g^2\mathcal{E}^2(t)\Gamma}{(\Gamma^2+\Delta^2)X} = -\frac{1}{T_1 ZX}(Z - Z^{(0)}), \tag{26a}$$

where

$$X = 1 + \frac{4g^2\mathcal{E}^2(t)(\Delta^2-\Gamma^2)}{(\Delta^2+\Gamma^2)^2}. \tag{26b}$$

Equation (26a) in the limit $T_1 \to \infty$ leads[16] to

$$Z(t) = -[X(t)]^{-1/2}\exp\left(-\frac{4\Gamma g^2}{(\Gamma^2+\Delta^2)}\int_{-\infty}^{t} d\tau\,\mathcal{E}^2(\tau)[X(\tau)]^{-1}\right). \tag{27}$$

The off-diagonal elements are obtained from (10), i.e.,

$$\begin{aligned}\rho_{\alpha\beta}(t) &= -i\int_0^{t-t_0} d\tau\, Y_{\alpha\beta}(\tau)[H_1(t-\tau)]_{\alpha\beta}\\ &\quad\times[\rho_{\beta\beta}(t-\tau) - \rho_{\alpha\alpha}(t-\tau)]\\ &= -i\int_0^{t-t_0} d\tau\, Y_{\alpha\beta}(\tau)\sum_0^\infty \frac{(-\tau)^n}{n!}\frac{\partial^n}{\partial t^n}[H_1(t)]_{\alpha\beta}\\ &\quad\times[\rho_{\beta\beta}(t) - \rho_{\alpha\alpha}(t)].\end{aligned} \tag{28}$$

For the case of two-level atom (27) and (28), in the limit $\Delta \gg \Gamma$, lead to the standard results.[7,9]

For the case of multilevel atoms if we use

$$[H_1(t-\tau)]_{\alpha\beta}\rho_{\beta\beta}(t-\tau) = [H_1(t)]_{\alpha\beta}\rho_{\beta\beta}(t) - \tau\frac{\partial}{\partial t}[H_1(t)]_{\alpha\beta}\rho_{\beta\beta}(t),$$

then we get equations of the type (25) and (28). However, it is difficult to obtain analytically the solutions of such equations.

Finally it should be noted that Grischkowsky *et al.*[17] have also derived the vector model, for the two-photon process taking place between the levels $|1\rangle$ and $|2\rangle$. They have shown that the elements of the transformed density matrix in the 2×2 subspace satisfy Bloch-type equations. The coupling constant in the effective Hamiltonian now involves the square of the field. It is clear that the adiabatic-following solutions can again be discussed in terms of solutions of non-Markovian Pauli-type master equations like (22) with the difference that $\mathcal{E}(t)$ is now replaced by $\mathcal{E}^2(t)$.

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[10]For a review of master equations see, for example, G. S. Agarwal, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1973), Vol. XI, p. 1.

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[12]Note that in Ref. 10 Zwanzig's master equation has been obtained without directly using dyadics.

[13]Another class of master equations, which are exact in Born approximation, has been found recently [G. S. Agarwal, Phys. Rev. A (to be published)]. This class corresponds to quantum-mechanical systems interacting with stochastic fields, where stochastic fields are taken to be Gaussian δ correlated [cf. R. Kubo, J. Math. Phys. 4, 174 (1963); J. Fox, J. Math. Phys. 13, 1196 (1972)].

[14]Cf. Ref. 10, Secs. 4 and 8.

[15]It may be worthwhile to note here that in the conventional approach one formally integrates Bloch equations for dipole moments and substitutes these in the equation for inversion to get exact equations like (23). However, if one works with master equations in the Born approximation, then one still discovers Eq. (23). Thus the proof that $\mathfrak{U}(t,\tau)=1$ for a two-level system fills the existing gap regarding the exactness of master equations for a two-level system.

[16]It may be worthwhile to note that the usual derivation of $Z(t)$ is via the relation $U^2+V^2+Z^2(t)=1$ rather than by the integration of the equation of motion (25) for $Z(t)$.

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