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Time-Convolutionless Master Equation for Multi-Level Open Quantum Systems with Initial System-Environment Correlations

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Abstract: Formally exact time-convolutionless master equation was derived by means of the projection operator method for the reduced statistical operator of a multi-level quantum system interacting with arbitrary external deterministic fields and dissipative environment simultaneously. While being closed and homogeneous in the reduced statistical operator, this equation accounts for thermodynamically equilibrium correlations between the multi-level system and the environment at the initial moment of time. On the basis of the exact master equation, an approximate time-convolutionless master equation for the reduced statistical operator was derived in the second order of the system-environment interaction strength. It was shown that the analysis of this equation can be simplified if the free Hamiltonian dynamics of an arbitrary quantum multi-level system driven by the external fields is described in terms of the $SU(N)$ algebra representation, so the master equation in question can be reduced to a set of ordinary differential equations for a finite number of time-dependent coefficients. As a consequence, efficient numerical methods can be employed to solve this master equation for various physically realistic quantum models of theoretical and practical importance.

Keywords: Initial correlations, Multi-level quantum system, Open system, Operator algebra, Projection operator, Reduced statistical operator, Time-convolutionless master equation

1 Introduction

Nowadays, theoretical research activities in every domain of applied physics imply, in effect, studies of various models of open quantum systems interacting with their respective environments and driven by external fields simultaneously. Under general circumstances, quantum objects can be rarely considered as closed systems isolated from their environment. As a rule, the environment affects the dynamics of a quantum system significantly by leaking the energy from the system by means of irreversible dissipative processes and by inducing decoherence processes within it. As a conventional starting point in most studies of open quantum systems, the quantum system of interest and its environment are considered a closed system. This compound system is described by some model Hamiltonian, which governs its combined unitary and reversible dynamics. In most instances of practical importance, such a total system possesses an enormous and infinite number of degrees of freedom. Most of these degrees, which belong to the environment, cannot be observed or controlled individually by any available experimental technique. Hence, the detailed knowledge about their behavior is of little interest for any practical purpose. Equally, from the purely theoretical point of view, there is no way to explicitly deduce from the original model Hamiltonian a total statistical operator describing the evolving non-equilibrium entangled state of the system and the environment altogether for the majority of the physically relevant realistic models. However, it is possible, at least formally, to “trace out” the environmental degrees of freedom from the total statistical operator. The ensuing reduced statistical operator describes the dynamics of the quantum system of interest only. Hence, the first step in any theoretical study in the field of open quantum systems is to derive the so-called master equation, i.e. the equation of motion for the reduced statistical operator. This equation provides

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one with complete knowledge about the evolution of the quantum state of the system itself and allows to study the processes of energy dissipation and decoherence. In fact, any master equation describes a complicated process of the system self-interaction. First, the system of interest interacts with the environment while changing its own state and the state of the environment in the process. Thereafter, the changed state of the environment affects the state of the system and changes its state. Thus, it seems that the system interacts with itself, then the state of the system at any given time is influenced by its states at all previous moments of time, i.e. the dynamics of the system is stipulated by its history. An essential and very important feature of master equations is that they are formally exact equations, which are closed in their respective reduced statistical operators. In most cases, these master equations are not tractable in their original form. Nevertheless, they are invaluable for the subsequent derivation of various approximate master equations which, in their turn, can be treated analytically and/or numerically. Numerous approaches to derivation of master equations have been proposed so far and are prevalently used at the present. The most rigorous are the microscopic ones, which start with explicitly formulated model Hamiltonian representing the generator of unitary and reversible dynamics of the total system. This dynamics is defined by the Liouville-von Neumann equation for the total statistical operator describing the entangled state of the quantum system and its environment. Among microscopic approaches, broadly applicable projection operator formalism, proposed in [1], proved that it is a flexible and universal mathematical tool. This approach allows to derive formally exact closed equation in the reduced statistical operator integra-differential time-local (time-convolution) or purely differential time-nonlocal (time-convolutionless) equations [2,3,4,5], albeit with one essential caveat: in general case, these equations contain non-homogeneous in the reduced statistical operator additive terms for arbitrary initial conditions. These terms represent formidable obstacles for subsequent attempts at numerical or analytical solution of exact and approximate master equations by conventional methods. The non-homogeneous terms disappear if the total statistical operator can be factorized at the initial moment of time, e.g. if the system and its environment are initially independent and uncorrelated. This assumption seems plausible for a great number of realistic experimental setups, but it is unjustified in general unless the initial uncorrelated state can be prepared artificially. In this study, we are preoccupied with a broad class of open quantum systems of practical importance in various fields of physics. All of them allow description by essentially one and the same model of an open quantum system possessing finite number of energy eigenstates and driven by multiple external deterministic fields of various nature, while interacting simultaneously with its environment. The said environment comprises of a multitude of heat bathes of various nature and structure represented by their own respective, either quantum or stochastic classical models. For example, in the domain of quantum optics, this model may represent an N-level one-electron “atom” excited by external electromagnetic semiclassical (laser) fields and interacting with an environment made of plurality of electromagnetic modes in free space or a cavity. Alternatively, the same model may describe an open quantum system of reduced dimensionality, like a quantum dot, interacting with external fields and its environment is made of not only electromagnetic modes but also quantized collective solid state quasi-particle excitations, like phonons. Generally, in either of these occurrences, the external fields are applied to a quantum system that is already correlated with its environment to some degree by the initial moment of time. Typically, the system and its environment are in the state of thermal equilibrium initially and their correlations can not be always neglected. Therefore, conventional homogeneous master equations are not useful anymore in this case. At the same time, it is highly desirable to derive master equations, which account for initial correlations while preserving homogeneity in reduced statistical operator. In this work, we employ well-established projection operator techniques supplemented with representation of an N-level open quantum system in terms of the SU(N) operator algebra formalism to derive exact and approximate time-convolutionless homogeneous master equations for the reduced statistical operator of the N-level quantum system while taking into account, along with the approach recently outlined in [6], the correlations between the system and its environment, which are initially in the state of thermal equilibrium.

The paper is organized, as follows: In Section 2, general features pertinent to the driven dynamics of a multi-level quantum system interacting with arbitrary external deterministic fields are discussed. In Section 3, we derive formally exact, as well as approximate, time-convolutionless, or time-local, differential equations for the reduced statistical operator of the quantum system in question under the assumption that the system and its environment are uncorrelated at the initial moment of time. In Section 4, this assumption is avoided in favor of much more general and plausible assumption that the system and the environment are in the state of thermal equilibrium initially. Under this assumption, formally exact time-convolutionless differential equation for the reduced statistical operator of the quantum system is derived. This equation is closed and homogeneous in the reduced statistical operator. An approximate time-convolutionless differential equation for the reduced statistical operator is derived in the second order of the system-environment interaction strength, as well. This approximate equation is thoroughly analyzed in Section 5 by means of its proper reformulation in terms of the SU(N) algebra representation of the quantum system dynamics previously outlined in Section 2. Section 6 involves the results.

2 Dynamics of a multi-level quantum system driven by deterministic external fields

Typical Hamiltonian of an open multi-level quantum system driven by deterministic external fields can be written as

$$\hat{H}(t) = \hat{H}_S(t) + \hat{H}_B + \hat{H}_{SB}, \tag{1}$$

where $\hat{H}_B$ stands for the Hamiltonian of the environment B and $\hat{H}_{SB}$ describes the interaction between the system S and the environment. The driven dynamics of the system S, induced solely by external fields of arbitrary nature and time dependence, is governed by the following Hamiltonian

$$\hat{H}_S(t) = \sum_m^N E_m \hat{\sigma}_{mm} + \sum_{m,n}^N V_{mn}(t)\hat{\sigma}_{mn}, \tag{2}$$

where N is the number of the system energy eigenstates with their corresponding eigenvalues E_m, and the projection and transition operators $\hat{\sigma}_{mn} = |m\rangle\langle n|$ are given in the standard bra- and ket- notation, so conventional commutation relations hold

$$[\hat{\sigma}_{ij}, \hat{\sigma}_{kl}] = \hat{\sigma}_{il}\delta_{jk} - \hat{\sigma}_{kj}\delta_{il}. \tag{3}$$

The factors $V_{mn}(t)$ account for the intensity and time dependence of the external fields. It is assumed that these fields are switched on at the time moment t_0, so

$$V_{mn}(t) = 0 \quad \text{for} \quad t \le t_0 \quad \forall m,n. \tag{4}$$

The driven dynamics of the system S is governed by the Liouville-von Neumann equation for its statistical operator

$$\frac{\partial}{\partial t}\hat{\rho}_S(t) = -\frac{i}{\hbar}[\hat{H}_S(t), \hat{\rho}_S(t)]. \tag{5}$$

Three groups of Hermitian operators u, v and w were introduced in[6]:

$$\hat{u}_{jk} = \hat{\sigma}_{jk} + \hat{\sigma}_{kj}, \hat{v}_{jk} = -i(\hat{\sigma}_{jk} - \hat{\sigma}_{kj}),\ 1 \le j < k \le N, \tag{6}$$

$$\hat{w}_l = -\left(\frac{2}{l(l+1)}\right)^{1/2} (\hat{\sigma}_{11} + \ldots + \hat{\sigma}_{ll} - l\hat{\sigma}_{l+1,l+1}),$$

$$1 \le l \le N-1, \tag{7}$$

so there exist $N^2 - 1$ operator variables totally. These operators are the generators of the SU(N) algebra, and if a vector $\hat{\mathbf{s}}$ is defined as an ordered sequence of these operators as

$$\hat{\mathbf{s}} = (\hat{u}_{12}, \ldots, \hat{u}_{N-1,N}, \hat{v}_{12}, \ldots, \hat{v}_{N-1,N}, \hat{w}_1, \ldots, \hat{w}_{N-1}), \tag{8}$$

then its components $\hat{s}_i$ satisfy commutation relations

$$[\hat{s}_j, \hat{s}_k] = 2i \sum_{l=1}^{N^2-1} f_{jkl}\hat{s}_l, \tag{9}$$

where f_{jkl} is a completely antisymmetric structure tensor of the SU(N) algebra. In terms of the vector $\hat{\mathbf{s}}$ components, the statistical operator and Hamiltonian (2) take their respective forms

$$\hat{\rho}_S(t) = N^{-1}\hat{I} + \frac{1}{2}\sum_{j=1}^{N^2-1} S_j(t)\hat{s}_j, \tag{10}$$

$$\hat{H}_S(t) = \frac{\hbar}{2}\sum_{j-1}^{N^2-1} \Gamma_j(t)\hat{s}_j + C(t)\hat{I}, \tag{11}$$

where the coefficients $S_j(t)$ and $\Gamma_j(t)$ are defined as

$$S_j(t) = Sp_S\{\hat{\rho}_S(t)\hat{s}_j\}, \tag{12}$$

$$\Gamma_j(t) = \hbar^{-1}Sp_S\{\hat{H}_S(t)\hat{s}_j\}, \quad C(t) = N^{-1}Sp_S\{\hat{H}_S(t)\}. \tag{13}$$

Expressions (10-13) were derived by means of the identity

$$Sp_S\{\hat{s}_j\hat{s}_k\} = 2\delta_{jk}, \tag{14}$$

which can also be employed for the calculation of the structure tensor f_{jkl} by multiplying relation (9) with arbitrary vector component $\hat{s}_p$ and tracing its both sides as

$$Sp_S\{\hat{s}_j\hat{s}_k\hat{s}_p\} - Sp_S\{\hat{s}_k\hat{s}_j\hat{s}_p\} = 4if_{jkp}, \tag{15}$$

wherefrom

$$f_{jkp} = \frac{i}{4}(Sp_S\{\hat{s}_k\hat{s}_j\hat{s}_p\} - Sp_S\{\hat{s}_j\hat{s}_k\hat{s}_p\}). \tag{16}$$

The last term in Eq.(11) is proportional to the identity operator $\hat{I}$, it does not affect the system dynamics and will be omitted in what follows. Then, a system of ordinary differential equations for the vector made of coefficients $\mathbf{S}$

$$\frac{d}{dt}S_i(t) = \sum_{j,k=1}^{N^2-1} f_{ijk}\Gamma_j(t)S_k(t) \tag{17}$$

follows from Eqs.(5), (10) and (14). All components of the vector $\mathbf{S}$ are real numbers and, due to complete antisymmetry of the tensor f_{ijk}, the length of the vector $\mathbf{S}$ is conserved. Thus, Eqs.(17) describe the rotation of this vector in the space of N^2-1 dimensions. Let us notice that all the operators $\hat{s}_j$ are traceless, i.e. $Sp\{\hat{s}_j\}=0$, so the probability conservation condition for the statistical operator $\hat{\rho}_S(t)$ presented in the form (10) is satisfied automatically for arbitrary values of the coefficients $S_j(t)$, whose feature may be of convenience upon usage of approximative numerical methods to calculate them. Let us consider a system of the Heizenberg equations

$$\frac{\partial}{\partial t}\hat{s}_l(t)=\frac{i}{\hbar}[\hat{H}_S(t),\hat{s}_l(t)],\ l=1,...,N^2-1 \tag{18}$$

for the operator components $\hat{s}_l(t)$. Its solutions can be always written as

$$\hat{s}_l(t,t_0)=\sum_{p=1}^{N^2-1}C_{lp}(t,t_0)\hat{s}_p,$$

$$C_{lp}(t_0,t_0)=\delta_{lp},\ l=1,...,N^2-1. \tag{19}$$

From this ansatz as well as relations (9) and (11), a system of equations

$$\frac{d}{dt}C_{lp}(t,t_0)=-\sum_{j,k=1}^{N^2-1}f_{pjk}\Gamma_j(t)C_{lk}(t,t_0),$$

$$C_{lp}(t_0,t_0)=\delta_{lp},\ l,p=1,...,N^2-1 \tag{20}$$

for the coefficients $C_{lp}(t,t_0)$ follows. For any fixed index l, this system is similar to that of Eqs.(17) for the statistical operator coefficients $S_i(t)$ and describes the rotation of the vector with components $C_{lp}(t,t_0)$ in the space of N^2-1 dimensions, too.

3 Time-convolutionless master equation without initial system-environment correlations

In the interaction picture

$$\tilde{\hat{\rho}}(t)=U_0^+(t,t_0)\hat{\rho}(t)U_0(t,t_0), \tag{21}$$

$$\tilde{\hat{H}}_{SB}(t)=U_0^+(t,t_0)\hat{H}_{SB}(t)U_0(t,t_0), \tag{22}$$

we find, as usual, the Liouville-von Neumann equation for the total statistical operator of the system (1)

$$\frac{\partial}{\partial t}\tilde{\hat{\rho}}(t)=-\frac{i}{\hbar}[\tilde{\hat{H}}_{SB}(t),\tilde{\hat{\rho}}(t)]=\tilde{L}(t)\tilde{\hat{\rho}}(t), \tag{23}$$

where the superoperator $\tilde{L}(t)$ is given by

$$\tilde{L}(t)...=-\frac{i}{\hbar}[\tilde{\hat{H}}_{SB}(t),...]. \tag{24}$$

Here the evolution operator $U_0(t,t_0)$ is defined as

$$U_0(t_2,t_1)=U_{0B}(t_2,t_1)U_{0S}(t_2,t_1), \tag{25}$$

where $U_{0B}(t_2,t_1)$ and $U_{0S}(t_2,t_1)$ are a time-ordered exponential operator function

$$U_{0S}(t_2,t_1)=\overleftarrow{T}\exp\{-\frac{i}{\hbar}\int_{t_1}^{t_2}dt'\hat{H}_S(t')\}, \tag{26}$$

$$U_{0S}^+(t_2,t_1)=\overrightarrow{T}\exp\{\frac{i}{\hbar}\int_{t_1}^{t_2}dt'\hat{H}_S(t')\}, \tag{27}$$

with $\overleftarrow{T}$ and $\overrightarrow{T}$ that represents the chronological and anti-chronological time ordering operator, respectively, and

$$U_{0B}(t_2,t_1)=\exp\{-\frac{i}{\hbar}\hat{H}_0(t_2-t_1)\}, \tag{28}$$

$$U_{0B}^+(t_2,t_1)=\exp\{\frac{i}{\hbar}\hat{H}_B(t_2-t_1)\}. \tag{29}$$

This evolution operator possesses the conventional properties

$$U_0(t_2,t_1)U_0^+(t_2,t_1)=1, \tag{30}$$

$$U_0(t_3,t_2)U_0(t_2,t_1)=U_0(t_3,t_1), \tag{31}$$

$$U_0(t_2,t_1)U_0(t_1,t_2)=1,\ U_0(t_1,t_2)=U_0^+(t_2,t_1). \tag{32}$$

Employing the projection operator formalism introduced originally in[1], Eq.(23) can be transformed into a couple of equations for the relevant $\tilde{\hat{\rho}}_1(t)$ and irrelevant $\tilde{\hat{\rho}}_2(t)$ parts of the total statistical operator as

$$\frac{\partial}{\partial t}\mathscr{P}\tilde{\hat{\rho}}(t)=\frac{\partial}{\partial t}\tilde{\hat{\rho}}_1(t)=\mathscr{P}\tilde{L}(t)(\tilde{\hat{\rho}}_1(t)+\tilde{\hat{\rho}}_2(t)), \tag{33}$$

$$\frac{\partial}{\partial t}\mathscr{Q}\tilde{\hat{\rho}}(t)=\frac{\partial}{\partial t}\tilde{\hat{\rho}}_2(t)=\mathscr{Q}\tilde{L}(t)(\tilde{\hat{\rho}}_1(t)+\tilde{\hat{\rho}}_2(t)), \tag{34}$$

where the operators $\mathscr{P}$ and $\mathscr{Q}$ are some projection operators such that

$$\mathscr{P}^2=\mathscr{P},\ \mathscr{Q}=1-\mathscr{P},\ \mathscr{P}\mathscr{Q}=0, \tag{35}$$

and

$$\tilde{\rho}(t) = \mathscr{P}\tilde{\rho}(t) + (1-\mathscr{P})\tilde{\rho}(t) = \tilde{\rho}_1(t) + \tilde{\rho}_2(t). \tag{36}$$

The second of these equations for the irrelevant part can be formally integrated to give

$$\tilde{\rho}_2(t) = \overleftarrow{\mathscr{G}}(t,t_0)\mathscr{Q}\hat{\rho}(t_0)$$

$$+\int_{t_0}^{t} d\tau \overleftarrow{\mathscr{G}}(t,\tau)\mathscr{Q}\tilde{L}(\tau)\mathscr{P}(\tilde{\rho}_1(\tau)+\tilde{\rho}_2(\tau)), \tag{37}$$

where the propagator

$$\overleftarrow{\mathscr{G}}(t,\tau) = \overleftarrow{T}\exp\left[\int_{\tau}^{t} dt'\mathscr{Q}\tilde{L}(t')\right] \tag{38}$$

is a solution to the equation

$$\frac{\partial \overleftarrow{\mathscr{G}}(t,\tau)}{\partial t} = \mathscr{Q}\tilde{L}(t)\overleftarrow{\mathscr{G}}(t,\tau), \quad \overleftarrow{\mathscr{G}}(\tau,\tau) = 1. \tag{39}$$

Let us notice that Eq.(37) can be also presented in the form

$$\tilde{\rho}_2(t) = \overleftarrow{\mathscr{G}}(t,t_0)\mathscr{Q}\hat{\rho}(t_0)$$

$$+\int_{t_0}^{t} d\tau \overleftarrow{\mathscr{G}}(t,\tau)\mathscr{Q}\tilde{L}(\tau)\mathscr{P}\overrightarrow{\mathscr{G}}(t,\tau)(\tilde{\rho}_1(t)+\tilde{\rho}_2(t)), \tag{40}$$

where the propagator

$$\overrightarrow{\mathscr{G}}(t,\tau) = \overrightarrow{T}\exp\left[-\int_{\tau}^{t} dt'\mathscr{Q}\tilde{L}(t')\right] \tag{41}$$

allows to express complete statistical operator $\tilde{\rho}(\tau)$ through $\tilde{\rho}(t)$ as

$$\tilde{\rho}(\tau) = \overrightarrow{\mathscr{G}}(t,\tau)\tilde{\rho}(t) \tag{42}$$

to get rid of the temporal non-locality in its integrand. Let us introduce an operator

$$\Sigma(t) = \int_{t_0}^{t} d\tau \overleftarrow{\mathscr{G}}(t,\tau)\mathscr{Q}\tilde{L}(\tau)\mathscr{P}\overrightarrow{\mathscr{G}}(t,\tau), \tag{43}$$

which helps transform Eq.(40) into

$$[1-\Sigma(t)]\tilde{\rho}_2(t) = \overleftarrow{\mathscr{G}}(t,t_0)\mathscr{Q}\hat{\rho}(t_0) + \Sigma(t)\tilde{\rho}_1(t). \tag{44}$$

Assuming the existence of the inverse operator $[1-\Sigma(t)]^{-1}$, the irrelevant part of the statistical operator can be written as

$$\tilde{\rho}_2(t) = [1-\Sigma(t)]^{-1}\overleftarrow{\mathscr{G}}(t,t_0)\mathscr{Q}\hat{\rho}(t_0)+$$

$$+[1-\Sigma(t)]^{-1}\Sigma(t)\tilde{\rho}_1(t). \tag{45}$$

Inserting this formal solution (37) into Eq.(33), we obtain an exact closed equation of motion for the relevant part of the statistical operator

$$\frac{\partial}{\partial t}\tilde{\rho}_1(t) = \mathscr{P}\tilde{L}(t)\mathscr{P}\tilde{\rho}_1(t)$$

$$+\mathscr{K}(t)\tilde{\rho}_1(t) + \mathscr{I}(t)\mathscr{Q}\hat{\rho}(t_0), \tag{46}$$

where

$$\mathscr{K}(t) = \mathscr{P}\tilde{L}(t)[1-\Sigma(t)]^{-1}\mathscr{P}, \tag{47}$$

$$\mathscr{I}(t) = \mathscr{P}\tilde{L}(t)[1-\Sigma(t)]^{-1}\overleftarrow{\mathscr{G}}(t_0,t)\mathscr{Q}. \tag{48}$$

with initial condition

$$\tilde{\rho}(t_0) = \hat{\rho}(t_0). \tag{49}$$

An assumption about the existence of the inverse operator $[1-\Sigma(t)]^{-1}$ constitutes major problem in justification of the applicability of the time-convolutionless master equation. Nevertheless, for some models systems, it can be rigorously proved[4] in the case of weak enough interaction $\hat{H}_{SB}$ and/or short enough time interval $t-t_0$. In numerous practical cases, it is possible to ensure that

$$\mathscr{P}\tilde{L}(t)\mathscr{P} = 0, \tag{50}$$

$$\mathscr{Q}\hat{\rho}(t_0) = 0, \tag{51}$$

so, Eq.(46) becomes more simple:

$$\frac{\partial}{\partial t}\tilde{\rho}_1(t) = \mathscr{K}(t)\tilde{\rho}_1(t), \tag{52}$$

Since one is interested, as a rule, in the weak-coupling limit of the system-environment interaction, one can find by assuming $\overleftarrow{\mathscr{G}}(t,\tau)\approx 1$, $\overrightarrow{\mathscr{G}}(t,\tau)\approx 1$, that

$$\frac{\partial}{\partial t}\tilde{\rho}_1(t) = \int_{t_0}^{t} d\tau \mathscr{P}\tilde{L}(t)\tilde{L}(\tau)\mathscr{P}\tilde{\rho}_1(t) \tag{53}$$

in the second order in the interaction Hamiltonian $\hat{H}_{SB}$.

4 Accounting for initial system-environment correlations

Now, let us account for the initial correlations in the same way as it was proposed in [7], assuming that the system S and the environment B were in thermal equilibrium at the initial moment of time t_0. Thus,

$$\hat{\rho}(t_0) = e^{-(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}/Sp\{e^{-(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}\}. \tag{54}$$

Then, by means of the identity

$$e^{-\beta\hat{H}} = e^{-\beta\hat{H}_0} -$$

$$-\int_0^\beta d\lambda e^{-\lambda\hat{H}_0}\hat{H}_1 e^{\lambda\hat{H}}e^{-\beta\hat{H}},\ \hat{H} = \hat{H}_0 + \hat{H}_1, \tag{55}$$

and noting that for the choice of the projection operator $\mathscr{P}$ in the form (72)

$$(1-\mathscr{P})e^{-\beta(\hat{H}_S(t_0)+\hat{H}_B)} = \mathscr{Q}e^{-\beta(\hat{H}_S(t_0)+\hat{H}_B)} = 0, \tag{56}$$

the irrelevant part of the total statistical operator at the initial moment of time $\tilde{\rho}_2(t_0) = \mathscr{Q}\hat{\rho}(t_0)$, which is responsible for the initial correlations, can be written as

$$\tilde{\rho}_2(t_0) = -\frac{\mathscr{Q}\int_0^\beta d\lambda e^{-\lambda(\hat{H}_S(t_0)+\hat{H}_B)}\hat{H}_{SB}e^{\lambda(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}}{Sp\{e^{-(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}\}}$$

$$\times e^{-\beta(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}$$

$$= -\mathscr{Q}\int_0^\beta d\lambda e^{-\lambda(\hat{H}_S(t_0)+\hat{H}_B)}\hat{H}_{SB}e^{\lambda(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}$$

$$\times[\overleftarrow{\mathscr{G}}(t,t_0)]^{-1}(\tilde{\rho}_1(t)+\tilde{\rho}_2(t))$$

$$= -I_Q(t,t_0,\beta)(\tilde{\rho}_1(t)+\tilde{\rho}_2(t)), \tag{57}$$

where

$$I_Q(t,t_0,\beta) = \mathscr{Q}\int_0^\beta d\lambda e^{-\lambda(\hat{H}_S(t_0)+\hat{H}_B)}\hat{H}_{SB}e^{\lambda(\hat{H}_S(t_0)+\hat{H}_B+\hat{H}_{SB})}$$

$$\times[\overleftarrow{\mathscr{G}}(t,t_0)]^{-1}. \tag{58}$$

As follows from Eq.(37),

$$\tilde{\rho}_2(t) = -\overleftarrow{\mathscr{G}}(t,t_0)I_Q(t,t_0,\beta)(\tilde{\rho}_1(t)+\tilde{\rho}_2(t))$$

$$+\int_{t_0}^t d\tau\overleftarrow{\mathscr{G}}(t,\tau)\mathscr{Q}\tilde{L}(\tau)\mathscr{P}\tilde{\rho}_1(\tau). \tag{59}$$

However, by definition,

$$\tilde{\rho}(\tau) = [\overleftarrow{\mathscr{G}}(t,\tau)]^{-1}\tilde{\rho}(t), \tag{60}$$

so

$$\tilde{\rho}_1(\tau) = \mathscr{P}[\overleftarrow{\mathscr{G}}(t,\tau)]^{-1}(\tilde{\rho}_1(t)+\tilde{\rho}_2(t)). \tag{61}$$

As a result, Eq.(37) can be transformed into

$$\tilde{\rho}_2(t) = \overleftarrow{\mathscr{G}}(t,t_0)\mathscr{Q}\hat{\rho}(t_0)$$

$$+\int_{t_0}^t d\tau\overleftarrow{\mathscr{G}}(t,\tau)\mathscr{Q}\tilde{L}(\tau)\mathscr{P}[\overleftarrow{\mathscr{G}}(t,\tau)]^{-1}(\tilde{\rho}_1(t)+\tilde{\rho}_2(t)), \tag{62}$$

wherefrom

$$\tilde{\rho}_2(t) = [1-\alpha(t,t_0)]^{-1}[\alpha(t,t_0)\tilde{\rho}_1(t)+\overleftarrow{\mathscr{G}}(t,t_0)\tilde{\rho}_2(t_0)], \tag{63}$$

where

$$\alpha(t,t_0) = \int_{t_0}^t d\tau\overleftarrow{\mathscr{G}}(t,\tau)\mathscr{Q}\tilde{L}(\tau)\mathscr{P}[\overleftarrow{\mathscr{G}}(t,\tau)]^{-1}. \tag{64}$$

Inserting this expression for $\tilde{\rho}_2(t)$ into Eq.(33), we obtain the inhomogeneous equation for the relevant part $\tilde{\rho}_1(t)$

$$\frac{\partial}{\partial t}\tilde{\rho}_1(t) = \mathscr{P}\tilde{L}(t)[1-\alpha(t,t_0)]^{-1}\times$$

$$\times[\tilde{\rho}_1(t)+\overleftarrow{\mathscr{G}}(t,t_0)\tilde{\rho}_2(t_0)]. \tag{65}$$

By means of Eqs.(57,63), it is now possible to express the initial irrelevant part $\tilde{\rho}_2(t_0)$ via the relevant part $\tilde{\rho}_1(t)$, and inserting this formal expression for the initial irrelevant part into Eq.(65), we obtain the closed homogeneous equation for the relevant part of the statistical operator

$$\frac{\partial}{\partial t}\tilde{\rho}_1(t) = \mathscr{P}\tilde{L}(t)[1-\alpha(t,t_0)]^{-1} \tag{66}$$

$$\times\{1-\overleftarrow{\mathscr{G}}(t,t_0)[1+\gamma(t,t_0,\beta)\overleftarrow{\mathscr{G}}(t,t_0)]^{-1}\gamma(t,t_0,\beta)\}\tilde{\rho}_1(t),$$

where

$$\gamma(t,t_0,\beta) = I_Q(t,t_0,\beta)[1-\alpha(t,t_0)]^{-1}. \tag{67}$$

Being restricted to the second order in the system-environment interaction strength approximation, this equation can be transformed into the following time-local equation

$$\frac{\partial}{\partial t}\tilde{\hat{\rho}}_1(t)=\mathscr{P}\tilde{L}(t)\mathscr{P}\tilde{\hat{\rho}}_1(t)$$

$$-\mathscr{P}\tilde{L}(t)\int_{t_0}^{t}d\tau\mathscr{Q}\tilde{L}(\tau)\mathscr{P}\tilde{\hat{\rho}}_1(t)$$

$$-\frac{1}{2}\mathscr{P}\tilde{L}(t)\mathscr{Q}\left[I_{(2)}(t,t_0,\beta)\tilde{\hat{\rho}}_1(t)+\tilde{\hat{\rho}}_1(t)I^{+}_{(2)}(t,t_0,\beta)\right], \quad (68)$$

or, taking into account technical assumption (50),

$$\frac{\partial}{\partial t}\tilde{\hat{\rho}}_1(t)=-\mathscr{P}\tilde{L}(t)\int_{t_0}^{t}d\tau\mathscr{Q}\tilde{L}(\tau)\mathscr{P}\tilde{\hat{\rho}}_1(t)$$

$$-\frac{1}{2}\mathscr{P}\tilde{L}(t)\mathscr{Q}\left[I_{(2)}(t,t_0,\beta)\tilde{\hat{\rho}}_1(t)+\tilde{\hat{\rho}}_1(t)I^{+}_{(2)}(t,t_0,\beta)\right], \quad (69)$$

where

$$I_{(2)}(t,t_0,\beta)=\int_0^{\beta}d\lambda e^{-\lambda(\hat{H}_S(t_0)+\hat{H}_B)}\hat{H}_{SB}e^{\lambda(\hat{H}_S(t_0)+\hat{H}_B)}, \quad (70)$$

$$I^{+}_{(2)}(t,t_0,\beta)=\int_0^{\beta}d\lambda e^{\lambda(\hat{H}_S(t_0)+\hat{H}_B)}\hat{H}_{SB}e^{-\lambda(\hat{H}_S(t_0)+\hat{H}_B)}. \quad (71)$$

5 Approximate time-convolutionless master equation in terms of the SU(N) algebra representation

Let us choose the projection operator $\mathscr{P}$ as

$$\mathscr{P}...=\hat{\rho}_B Sp_B\{...\}, \quad \hat{\rho}_B=e^{-\beta\hat{H}_B}/Sp_B\{e^{-\beta\hat{H}_B}\}, \quad (72)$$

where the reference state of the environment $\hat{\rho}_B$ is a thermal equilibrium state of the environment, so the reduced statistical operator of the system S is given by

$$\hat{\rho}_S(t)=Sp_B\{\hat{\rho}_1(t)\}. \quad (73)$$

In what follows, we also assume

$$Sp_B\{\hat{H}_{SB}\hat{\rho}_B\}=0, \quad (74)$$

without loss of generality, because we can always redefine the Hamiltonians $H_S(t)$ and H_{SB} as

$$\hat{H}_S(t)\to\hat{H}_S(t)+Sp_B\{\hat{H}_{SB}\hat{\rho}_B\}, \quad (75)$$

$$\hat{H}_{SB}\to\hat{H}_{SB}-Sp_B\{\hat{H}_{SB}\hat{\rho}_B\}, \quad (76)$$

without making any alteration to the original Hamiltonian (1). The system-environment interaction Hamiltonian H_{SB} can be always written in the form

$$\hat{H}_{SB}=\sum_{k=1}^{N^2-1}\hat{E}_k\hat{s}_k+\hat{E}_0\hat{I}, \quad (77)$$

where the environment related operators $\hat{E}_k$ are defined as

$$\hat{E}_k=\frac{1}{2}Sp_S\{\hat{H}_{SB}\hat{s}_k\},\ \hat{E}_0=N^{-1}Sp_S\{\hat{H}_{SB}\} \quad (78)$$

in full analogy with expressions (13). The second term does not contain any of the operators $\hat{s}_k$ and is often equal to zero for practically useful models. Otherwise, it can be included into the Hamiltonian H_B. Summing up all these assumptions, we derive from Eq.(53) an approximate equation for the reduced statistical operator $\hat{\rho}_S(t)$ in the interaction picture

$$\frac{\partial}{\partial t}\tilde{\hat{\rho}}_S(t)=-\frac{i}{2\hbar}\int_0^{\beta}d\lambda\sum_{n,m}^{N^2-1}\Bigg\{\left[\tilde{\hat{s}}_n(t,t_0),\hat{s}_m(\lambda)\tilde{\hat{\rho}}_S(t)\right]$$

$$\times Sp_B\left\{\tilde{\hat{E}}_n(t,t_0)\hat{E}_m(\lambda)\hat{\rho}_B\right\}-h.c.\Bigg\}$$

$$-\frac{1}{\hbar^2}\sum_{n,m=1}^{N^2-1}\int_{t_0}^{t}dt'\left[\tilde{\hat{s}}_n(t,t_0),\tilde{\hat{s}}_m(t',t_0)\tilde{\hat{\rho}}_S(t)\right]$$

$$\times Sp_B\{\tilde{\hat{E}}_n(t,t_0)\tilde{\hat{E}}_m(t',t_0)\hat{\rho}_B\}$$

$$-\frac{1}{\hbar^2}\sum_{n,m=1}^{N^2-1}\int_{t_0}^{t}dt'\left[\tilde{\hat{s}}_n(t,t_0),\tilde{\hat{\rho}}_S(t)\tilde{\hat{s}}_m(t',t_0)\right]$$

$$\times Sp_B\{\tilde{\hat{E}}_m(t',t_0)\tilde{\hat{E}}_n(t,t_0)\hat{\rho}_B\}, \quad (79)$$

where

$$\tilde{\hat{s}}_n(t,t_0)=U^{\dagger}_{0S}(t,t_0)\hat{s}_nU_{0S}(t,t_0)=\sum_{l=1}^{N^2-1}\tilde{C}_{nl}(t,t_0)\hat{s}_l, \quad (80)$$

$$\hat{s}_n(\lambda)=e^{-\lambda\hat{H}_S(t_0)}\hat{s}_ne^{\lambda\hat{H}_S(t_0)}=\sum_{l=1}^{N^2-1}C_{nl}(\lambda)\hat{s}_l, \quad (81)$$

$$\tilde{\hat{E}}_n(t,t_0)=U^{\dagger}_{0B}(t,t_0)\hat{E}_nU_{0B}(t,t_0), \quad (82)$$

$$\hat{E}_n(\lambda)=e^{-\lambda\hat{H}_B}\hat{E}_ne^{\lambda\hat{H}_B}, \quad (83)$$

and the coefficients $\tilde{C}_{nk}(t)$ are calculated by integrating Eqs.(20). In accordance with expansion (10) for the statistical operator $\tilde{\tilde{\rho}}_S(t)$, we will search for a solution to Eq.(79) in the form

$$\tilde{\tilde{\rho}}_S(t) = N^{-1}\hat{I} + \frac{1}{2}\sum_{j=1}^{N^2-1}\tilde{S}_i(t)\hat{s}_i. \tag{84}$$

Inserting expansions (80) and (84) into Eq.(79) and collecting coefficients at the operators $\hat{s}_k$, we obtain, after some lengthy algebra, a system of differential equations for the coefficients $\tilde{S}_k(t)$:

$$\frac{\partial}{\partial t}\tilde{S}_i(t) = \sum_{k=1}^{N^2-1}\left[\tilde{A}_{ik}(t,t_0) + \tilde{A}'_{ik}(t,t_0)\right]\tilde{S}_k(t)$$

$$+\tilde{I}_i(t,t_0) + \tilde{I}_i{}'(t,t_0), \tag{85}$$

where

$$\tilde{A}_{ik}(t,t_0) =$$

$$= -\frac{1}{4\hbar^2}\sum_{n,m}^{N^2-1}\int_{t_0}^{t}dt'\tilde{C}^{(1)i}_{nmk}(t,t',t_0)Sp_B\{\tilde{\hat{E}}_n(t,t_0)\tilde{\hat{E}}_m(t',t_0)\hat{\rho}_B\}$$

$$-\frac{1}{4\hbar^2}\sum_{n,m}^{N^2-1}\int_{t_0}^{t}dt'\tilde{C}^{(2)i}_{nmk}(t,t',t_0)Sp_B\{\tilde{\hat{E}}_m(t',t_0)\tilde{\hat{E}}_n(t,t_0)\hat{\rho}_B\}, \tag{86}$$

$$\tilde{I}_i(t,t_0) = \int_{t_0}^{t}dt'\tilde{I}_i(t,t',t_0), \tag{87}$$

$$\tilde{C}^{(1)i}_{nmk}(t,t',t_0) =$$

$$= \sum_{l,p=1}^{N^2-1}\tilde{C}_{nl}(t,t_0)\tilde{C}_{mp}(t',t_0)Sp_S\{[\hat{s}_l,\hat{s}_p\hat{s}_k]\hat{s}_i\}, \tag{88}$$

$$\tilde{C}^{(2)i}_{nmk}(t,t',t_0) =$$

$$= \sum_{l,p=1}^{N^2-1}\tilde{C}_{nl}(t,t_0)\tilde{C}_{mp}(t',t_0)Sp_S\{[\hat{s}_l,\hat{s}_k\hat{s}_p]\hat{s}_i\}, \tag{89}$$

$$\tilde{I}_i(t,t',t_0) =$$

$$= -\frac{1}{2N\hbar^2}\sum_{n,m}^{N^2-1}\tilde{C}^i_{nm}(t,t',t_0)Sp_B\{\tilde{\hat{E}}_n(t,t_0)\tilde{\hat{E}}_m(t',t_0)\hat{\rho}_B\}$$

$$-\frac{1}{2N\hbar^2}\sum_{n,m}^{N^2-1}\tilde{C}^i_{nm}(t,t',t_0)Sp_B\{\tilde{\hat{E}}_m(t',t_0)\tilde{\hat{E}}_n(t,t_0)\hat{\rho}_B\}, \tag{90}$$

$$\tilde{C}^i_{nm}(t,t',t_0) = 4i\sum_{l,p=1}^{N^2-1}\tilde{C}_{nl}(t,t_0)\tilde{C}_{mp}(t,t',t_0)f_{ilp}. \tag{91}$$

$$\tilde{A}'_{ik}(t,t_0) =$$

$$= -\frac{i}{4\hbar}\int_0^{\beta}d\lambda\sum_{n,m}^{N^2-1}\tilde{B}^i_{nmk}(\lambda,t,t_0)$$

$$\times Sp_B\left\{\tilde{\hat{E}}_n(t,t_0)\hat{E}_m(\lambda)\hat{\rho}_B\right\}, \tag{92}$$

$$\tilde{I}_i{}'(t,t_0) =$$

$$= -\frac{i}{2N\hbar}\int_0^{\beta}d\lambda\sum_{n,m}^{N^2-1}\tilde{B}^i_{nm}(\lambda,t,t_0)$$

$$\times Sp_B\left\{\tilde{\hat{E}}_n(t,t_0)\hat{E}_m(\lambda)\hat{\rho}_B\right\}, \tag{93}$$

$$\tilde{B}^i_{nmk}(\lambda,t,t_0) = \sum_{l,p}^{N^2-1}\tilde{C}_{nl}(t,t_0)C_{mp}(\lambda)Sp_S\left\{[\hat{s}_l,\hat{s}_p\hat{s}_k]\hat{s}_i\right\}, \tag{94}$$

$$\tilde{B}^i_{nm}(\lambda,t,t_0) = 4i\sum_{l,p}^{N^2-1}\tilde{C}_{nl}(t,t_0)C_{mp}(\lambda)f_{ilp}. \tag{95}$$

6 Conclusion

We derived formally exact time-convolutionless master equation for the reduced statistical operator of an open quantum N-level system driven by external deterministic fields and interacting with its environment. This equation is homogeneous and closed in the reduced statistical operator, though it is assumed that the system and its environment are in the state of thermal equilibrium at the initial moment of time. It was also shown that the formally exact master equation may serve as a source for derivation of approximate master equations. One exemplary approximate master equation was obtained under the assumption of weak system-environment interaction strength. No assumptions were made regarding the system-field interaction strength. Approximate master equations can be derived in a regular way within the frame of a perturbative scheme whose essence is an expansion of all super-operators in exact Eq.(66) in powers of the system-environment interaction strength assumed to be weak. It is important to notice that the terms, taking into account initial correlations in the approximate Eq.(69), are formally exact in the second order in the system-environment interaction strength, so they match the other terms in this equation. Representation of the driven dynamics of the system, resulting from its interaction with external fields, in terms of the SU(N) algebra formalism is deemed to be instrumental in facilitating numerical solution of the approximate equation, which is applied to realistic physical models of open multi-level quantum systems. The main difference between the conventional Nakajima-Zwanzig time-convolution, or time-nonlocal, master equation and the time-convolutionless equation of the type (66) is in the time-local form of the latter, resulting in formally exact and approximate differential equations. However, the Nakajima-Zwanzig equation requires an integration over the history of the quantum system, leading to correspondent integra-differential equations. Nevertheless, the procedure of derivation for both types of equations, whether exact or approximate, relies on the same set of assumptions and approximations. The absence of time-convolution in Eqs.(66,69,79,85) does not mean at all that the history of the system is totally neglected. It is retained, either in full through the time dependence of super-operators acting on the statistical operator $\tilde{\rho}_1(t)$ in the formally exact Eq.(66), or partially through the time-dependent super-operators or C-number coefficients of the approximate differential equations (69,79,85). Therefore, it is generally assumed that both types of master equations - time-convolution and time-convolutionless - are able to describe the approximate non-Markovian dynamics of open quantum systems with the same accuracy at least in the case of weak system-environment interaction strength. At the same time, some arguments in [8] stated that in the limit of weak system-environment interaction, a time-convolutionless approximate equations may provide more precise description of the system evolution than the time-convolution ones. As a rule, it is easier to solve purely differential equations than the integra-differential ones. Another attractive advantage of dealing with purely differential approximate equations is that in some cases they may be analyzed, interpreted and solved by methods of an unraveling for non-Markovian time evolution by means of a stochastic process in the extended state space [9].

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Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this article

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