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Conditions for Bounded Solutions of Non-Markovian Quantum Master Equations

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For the most general quantum master equations, also called Nakajima–Zwanzig or non-Markovian equations, we define suitable boundedness conditions on integral kernels and inhomogeneity terms in order to derive with mathematical rigor an upper bound on solutions, as required by the von Neumann conditions. Such equations are of importance for quantum dynamics of open systems with arbitrary couplings to environment and arbitrary entangled initial states. The derivation is based on an equivalent coherence-vector representation in finite dimension n leading to coupled Volterra integro-differential equations of second kind and convolution type in an (n^2-1)-dimensional real vector space. As examples, analytical and numerical model solutions are worked out for 2-level systems in order to test suitable trial functions for input quantities. All this is motivated by the fact that exact solutions can hardly be found but appropriate trial functions may provide a reasonable semiphenomenological description of complicated quantum dynamics.

KEY WORDS: Non-Markovian quantum master equations; irreversible quantum dynamics; entanglement.

Quantum Markovian master equations are successfully applied whenever a system is weakly coupled to its surroundings and an average time-evolution starting from a disentangled initial state can be considered on a relatively large time-scale. Mathematically, the underlying dynamical equations are coupled linear first-order differential equations with constant coefficients satisfying well-defined quantum dynamical semigroup properties.[(1–4)] The developments in laser physics and mainly the possibility of monitoring time-evolution on very short scales down to femtoseconds made it clear that the

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assumptions underlying the weak-coupling and van Hove limit for Markovian master equations should be abandoned for a reliable description. For many related physical conditions neither a weak-coupling assumption without van Hove limit nor a disentangled initial state provides a realistic approximation. As a striking example where such assumptions must fail one may mention the particular quantum collapse and revival dynamics of the cavity damped Jaynes–Cummings model in quantum optics.[5–9] It has been shown[5, 6] that for relatively long times there is either strongly varying oscillatory dynamics or, lateron, a chaotically fluctuating behavior in a quasi-steady state of maximum entropy and, only on extremely long time-scale, cavity damping induces a Markovian type exponential decay.

If one wants to describe such complicated dynamical details by a master equation one has to go back to the most general version known as Nakajima–Zwanzig equation[10] for which detailed derivations are well documented in the literature[3, 4, 10, 11] but it will be necessary to collect a few prerequisits. The desired equation of "non-Markovian" type for the density operator $\rho(t)$ of an open system $\mathscr{S}_1$ reads

$$\dot{\rho}(t) = -i[\tilde{H}_1\,, \rho(t)] + \int_0^t \mathbf{K}(t-s; \rho(s))\, ds + \mathbf{J}(t), \tag{1}$$

where kernel and inhomogeneity are given by

$$\mathbf{K}(t-s; \rho(s)) = \mathrm{Tr}_2[\mathscr{L}\, e^{Q\mathscr{L}(t-s)} Q\, \mathscr{L}(\rho(s) \otimes \omega)], \tag{2}$$

$$\mathbf{J}(t) = \mathrm{Tr}_2[\mathscr{L}\, e^{Q\mathscr{L}t}(W(0) - \rho(0) \otimes \omega)]. \tag{3}$$

The setting is as follows. $\mathscr{S}_1$ is coupled to a system $\mathscr{S}_2$ via a bounded interaction V_{12} appearing in the total Hamiltonian $H = H_1 \otimes \mathbb{1}_2 + \mathbb{1}_1 \otimes H_2 + V_{12}$, and $\tilde{H}_1$ includes a modification of H_1 due to V_{12}. $\mathscr{L}$ abbreviates the commutator $\mathscr{L}X = -i[H, X]$, $X \in T(\mathscr{H})$, the set of trace-class operators in the tensor space $\mathscr{H} = \mathscr{H}_1 \otimes \mathscr{H}_2$ associated with the total closed system $\mathscr{S} = \mathscr{S}_1 \cup \mathscr{S}_2$. Furthermore, Tr_2 denotes the trace in $\mathscr{H}_2$, and Q projects any operator $Y \in T(\mathscr{H})$ into $QY = Y - \mathrm{Tr}_2[Y] \otimes \omega$, ω being an arbitrary density operator of $\mathscr{S}_2$. Finally, $W(0)$ is a possibly nonfactorizable (entangled) initial state of $\mathscr{S}$.

Serious difficulties in a further treatment of (1) arise from the nasty exponentials in (2) and (3) which make the general equation untractable, even in simple model cases. It should be emphasized that any approximation procedures are likely to violate the von Neumann conditions of positivity and trace-normalisation of $\rho(t)$. Therefore, one is led to take as much advantage as possible of the exact structure (1–3).

It is the aim of this note to present a first step in defining conditions to be imposed on $\mathbf{K}$ and $\mathbf{J}$ in order to guarantee at least bounded solutions. These conditions are expected to set useful guidelines for the choice of parametrized model functions which may provide a first semiphenomenological description of experiments. This idea bears some analogy to the well-known Bloch equations of optical or magnetic resonance where a parametrization of quantum mechanically consistent equations in terms of relaxation times $2\,T_1 \geqslant T_2$ has proven to be useful even if no ab initio derivation of the latter is known.[4, 12, 13]

First of all, we restrict derivations to a finite-dimensional Hilbert space $\mathscr{H}_1$ with $\dim(\mathscr{H}_1) = n < \infty$. Since $\rho(t)$ is hermitian with $\mathrm{Tr}_1[\rho(t)] = 1$ its matrixelements $\rho_{ik}(t) = (\varphi_i, \rho(t)\,\varphi_k)$ in any orthonormal basis $\{\varphi_i\}_1^n$ in $\mathscr{H}_1$ are defined in terms of $N = n^2 - 1$ real-valued functions of time. It is, therefore, most convenient to reformulate (1) in a higher-dimensional real vector space $\mathbf{V}^N$ by going over to a coherence-vector representation[4, 14] in terms of infinitesimal generators $\{F_i\}_1^N$ of $SU(n)$. A vector $\mathbf{v}(t) = \{v_1(t), v_2(t),..., v_N(t)\}^T$ with real-valued functions of time as components is then defined by

$$\rho(t) = \frac{1}{n}\,\mathbb{1}_n + \sum_{i=1}^{N} v_i(t)\,F_i, \tag{4}$$

$$F_i = F_i^*, \qquad \mathrm{Tr}[F_i] = 0, \qquad \mathrm{Tr}[F_i\,F_k] = \delta_{ik}, \qquad 1 \leqslant i, k \leqslant N. \tag{5}$$

Most important, the Euclidian norm of $\mathbf{v}(t)$ is bounded by

$$\|\mathbf{v}(t)\|^2 \leqslant 1 - \frac{1}{n}, \tag{6}$$

which is trivially required of any solution of (1), unavoidably also of approximate ones. It follows from (2) that matrixelements of $\mathbf{K}$ must be written as

$$(\varphi_i, \mathbf{K}(t-s; \rho(s))\,\varphi_j) = \sum_{\mu, \nu=1}^{n} K_{ij}^{\mu\nu}(t-s)\,\rho_{\mu\nu}(s), \qquad \mathrm{Tr}_1[\mathbf{K}(t-s; \rho(s)] = 0, \tag{7}$$

in terms of complex-valued functions $\{K_{ij}^{\mu\nu}(\tau), \tau \geqslant 0\}$. Since also $\mathrm{Tr}_1[\mathbf{J}(t)] = 0$, and only the traceless part of the Hamiltonian matters, we write

$$\mathbf{J}(t) = \sum_{k=1}^{N} \varepsilon_k(t)\,F_k, \qquad H = \sum_{k=1}^{N} h_k\,F_k. \tag{8}$$

The systematic transformation of (1) yields the desired final, real vector form

$$\dot{\mathbf{v}}(t) = A\,\mathbf{v}(t) + \int_0^t C(t-s)\,\mathbf{v}(s)\,ds + \mathbf{f}(t), \qquad 0 \leqslant t < \infty, \tag{9}$$

where $A = -A^T$ with entries given by $\{h_i\}_1^N$, and the entries of C are real, linear combinations of $\{K_{ij}^{\mu\nu}\}_1^n$-functions, whereas the components of $\mathbf{f}(t)$ are related to $\{\varepsilon_i(t)\}_1^N$, but also to certain integrals over $\{K_{ij}^{\mu\nu}\}$-functions. We concentrate on the relaxing part of (9) and set A to zero since the skew-symmetry induces orthogonal transformations which due not affect bound estimates. This can also be seen from a variation of parameter formula applied to (9).

Note first that all functions in (9) are continuous. More precisely, the components $f_i(t)$ are defined for $[0, \infty) \to \mathbf{R}$, and the elements $C_{ik}(t-s)$ for $[0 \leqslant s \leqslant t < \infty) \to \mathbf{R}$. Integration yields

$$\mathbf{v}(t) = \mathbf{v}(0) + \int_0^t \mathbf{f}(u)\,du + \int_0^t \int_0^u C(u-s)\,\mathbf{v}(s)\,ds\,du, \tag{10}$$

and, after interchanging order of integrations, one obtaines a Volterra convolution integral equation of second kind,[15]

$$\mathbf{v}(t) = \mathbf{F}(t) + \int_0^t R(t-s)\,\mathbf{v}(s)\,ds, \qquad 0 \leqslant t < \infty, \tag{11}$$

where

$$\mathbf{F}(t) = \mathbf{v}(0) + \int_0^t \mathbf{f}(u)\,du, \tag{12}$$

$$R(t-s) = \int_0^{t-s} C(u)\,du. \tag{13}$$

Again, $\mathbf{F}$ and R contain only continuous functions on the respective intervals and, consequently, the integral equation (11) admits a unique solution[16] $\mathbf{v}(t)$. Furthermore, we introduce

$$G(t-s) = -\int_{t-s}^{\infty} C(u)\,du, \tag{14}$$

and the constant matrix

$$Q = \int_0^{\infty} C(u)\,du = -G(0). \tag{15}$$

Suppose now the existence of positive constants $\{\alpha, M, \gamma, L, \kappa\}$ such that the following semi-inequalities for upper bounds hold,

$$\|e^{Q(t-u)}\| \leqslant M e^{-\alpha(t-u)}, \qquad 0 \leqslant u \leqslant t < \infty, \tag{16}$$

$$\|G(t-s)\| \leqslant L e^{-\gamma(t-s)}, \qquad 0 \leqslant s \leqslant t < \infty, \tag{17}$$

$$\int_0^t \|\mathbf{f}(u)\| \, du \leqslant \kappa, \qquad t \geqslant 0. \tag{18}$$

Under the above assumptions it will be possible to prove that the solutions $\mathbf{v}(t)$ of (11) are restricted by an upper bound that is analytically given by the introduced constants and the initial condition. To this end, consider first the identity

$$\frac{d}{dt}\int_0^t G(t-s)\,\mathbf{v}(s)\,ds = -\left(\int_0^\infty C(u)\,du\right)\mathbf{v}(t) + \int_0^t C(t-s)\,\mathbf{v}(s)\,ds, \tag{19}$$

and insert (15) and (19) in (9) to obtain

$$\dot{\mathbf{v}} - Q\,\mathbf{v} = \mathbf{f}(t) + \frac{d}{dt}\int_0^t G(t-s)\,\mathbf{v}(s)\,ds. \tag{20}$$

Multiplying this equation by e^{-Qt} yields

$$\frac{d}{dt}\left(e^{-Qt}\mathbf{v}(t)\right) = e^{-Qt}\left\{\mathbf{f}(t) + \frac{d}{dt}\int_0^t G(t-s)\,\mathbf{v}(s)\,ds\right\}, \tag{21}$$

and subsequent integration from 0 to t gives

$$e^{-Qt}\,\mathbf{v}(t) = \mathbf{v}(0) + \int_0^t e^{-Qu}\,\mathbf{f}(u)\,du + \int_0^t e^{-Qu}\left\{\frac{d}{du}\int_0^u G(u-s)\,\mathbf{v}(s)\,ds\right\}du. \tag{22}$$

Partial integration of the last term above yields

$$\begin{aligned} e^{-Qt}\,\mathbf{v}(t) = \mathbf{v}(0) &+ \int_0^t e^{-Qu}\,\mathbf{f}(u)\,du + e^{-Qt}\int_0^t G(t-s)\,\mathbf{v}(s)\,ds \\ &+ \int_0^t\int_0^u Q\,e^{-Qu}\,G(u-s)\,\mathbf{v}(s)\,ds\,du, \end{aligned} \tag{23}$$

and final multiplication by e^{Qt} provides a representation suitable for norm estimates,

$$\mathbf{v}(t) = e^{Qt}\,\mathbf{v}(0) + \int_0^t e^{Q(t-u)}\mathbf{f}(u)\,du + \int_0^t G(t-s)\,\mathbf{v}(s)\,ds$$

$$+ \int_0^t \int_0^u Q\,e^{Q(t-u)}G(u-s)\,\mathbf{v}(s)\,ds\,du. \tag{24}$$

Upon use of (16) a first norm estimate of this equation is given by

$$\|\mathbf{v}(t)\| \leqslant M\,\|\mathbf{v}(0)\|\,e^{-\alpha t} + M\int_0^t e^{-\alpha(t-u)}\,\|\mathbf{f}(u)\|\,du + \int_0^t \|G(t-s)\|\,\|\mathbf{v}(s)\|\,ds$$

$$+ M\,\|Q\|\int_0^t \int_0^u e^{-\alpha(t-u)}\,\|G(u-s)\|\,\|\mathbf{v}(s)\|\,ds\,du. \tag{25}$$

In the last term above the order of integration is interchanged,

$$\int_0^t \int_0^u e^{-\alpha(t-u)}\,\|G(u-s)\|\,\|\mathbf{v}(s)\|\,ds\,du = e^{-\alpha t}\int_0^t \left\{\int_s^t e^{\alpha u}\,\|G(u-s)\|\,du\right\}\|\mathbf{v}(s)\|\,ds, \tag{26}$$

and, taking into account (24), one obtaines

$$\int_0^t \int_0^u e^{-\alpha(t-u)}\,\|G(u-s)\|\,\|\mathbf{v}(s)\|\,ds\,du \leqslant Le^{-\alpha t}\int_0^t e^{\gamma s}\left\{\int_s^t e^{(\alpha-\gamma)\,u}\,du\right\}\|\mathbf{v}(s)\|\,ds,$$

$$= \frac{L}{\alpha-\gamma}\int_0^t (e^{-\gamma(t-s)} - e^{-\alpha(t-s)})\,\|\mathbf{v}(s)\|\,ds. \tag{27}$$

Inserting (27) in (25) and replacing $e^{-\alpha\,t}$ and $e^{-\alpha\,(t-u)}$, $u \leqslant t$, by 1 in the first two terms leads to the inequality

$$\|\mathbf{v}(t)\| \leqslant M(\|\mathbf{v}(0)\| + \kappa) + \int_0^t \phi(t-s)\,\|\mathbf{v}(s)\|\,ds, \tag{28}$$

where

$$\phi(\tau) = L\,e^{-\gamma\,\tau} + \frac{M\,\|Q\|\,L}{\alpha-\gamma}\,(e^{-\gamma\,\tau} - e^{-\alpha\,\tau}). \tag{29}$$

Due to Gronwall's inequality[16] one can rewrite (28) as

$$\|\mathbf{v}(t)\| \leqslant M(\|\mathbf{v}(0)\| + \kappa)\,\exp\left[\int_0^t \phi(\tau)\,d\tau\right]. \tag{30}$$

Note that $\phi(\tau) \geqslant 0$ for any values of $\{\alpha, \gamma\}$, with positive integral

$$\psi(t) = \int_0^t \phi(\tau)\, d\tau = \frac{L}{\gamma}\left(1+\frac{M\,\|Q\|}{\alpha-\gamma}\right)(1-e^{-\gamma t}) - \frac{M\,\|Q\|\,L}{\alpha(\alpha-\gamma)}(1-e^{-\alpha t}) \geqslant 0. \quad (31)$$

Therefore, the right-hand side of (28) is majorized by taking the limit $t \to \infty$,

$$\psi(t) \leqslant \lim_{\tau\to\infty} \psi(\tau) = \frac{L}{\alpha\gamma}(\alpha + M\,\|Q\|). \quad (32)$$

Finally, we have the following desired

Theorem. Subject to conditions (16–18) the solutions $\mathbf{v}(t)$ of the vector representation (9), equivalent to the non-Markovian quantum master equation (1), are bounded by

$$\|\mathbf{v}(t)\| \leqslant M(\|\mathbf{v}(0)\| + \kappa)\exp\left[\frac{L}{\alpha\gamma}(\alpha + M\,\|Q\|)\right]. \quad (33)$$

Two interpretations of this result must now be discussed. First of all, for sufficiently small initial norm $\|\mathbf{v}(0)\|$ and correspondingly modest values of constants (16–18) the required exact bound (6) will not be violated. Physically, this is a realistic scenario since neigther in Markovian[4] nor in non-Markovian[9] cases is it generally true that the dynamics is exclusively contractive in the sense $\|\mathbf{v}(t)\| \leqslant \|\mathbf{v}(0)\|$ for all initial conditions. This property is only strictly required for initial states very close to pure ones, all of them with maximum $\|\mathbf{v}(0)\| = (1-1/N)$. Second, even for the latter cases our present experience from numerous numerical tests has shown that result (33) is very useful. As a matter of fact, for a given choice of $\{\mathbf{K}, \mathbf{J}\}$ with corresponding parameters (16–18) such that, roughly, $\|\mathbf{v}(t)\| \leqslant 2\,(1-1/N)$ is fulfilled, we find from exact numerical calculations strict contractivity for maximum initial norm. The obviously somewhat too large bound in (33) is certainly due to the techniques applied in the derivation since norm estimates may considerably overestimate values as obtained from more refined methods. In summary, it is for all the above mentioned reasons that (33) turns out to provide a useful guideline in conveiving and testing trial functions.

As an illustration we consider 2 examples for a 2-level system ($n = 2$; $N = 3$). For a reliable numerical solution of Eq. (9) we have developed a very accurate procedure based on the Adams–Moulton method[17–19] of order 3.

(I) A simple choice of a diagonal kernel and an inhomogeneous term is given by

$$R(t) = -xe^{-\lambda t}\mathbb{1}, \qquad \mathbf{f}(t) = \kappa\sqrt{\frac{2}{3\pi}}\,e^{-t^2}(1\,,2\,,-1)^T. \tag{34}$$

The spectral norms are exactly found as

$$\|G(t)\| = \frac{x}{\lambda}\,e^{-\lambda t}, \qquad \|Q\| = \frac{x}{\lambda}, \qquad \|e^{Q\,t}\| = e^{-\frac{x}{\lambda}t}, \tag{35}$$

yielding a bound $\|\mathbf{v}(t)\| \leqslant (\|\mathbf{v}(0)\| + \kappa)\exp(2\,x/\lambda^2)$. In order to favor the central state for long times the value of κ must be small, similar to the Markovian case.[4] The choice $\{\kappa = 0.01, x = 1, \lambda = 5\}$ then gives us $\|\mathbf{v}(t)\| \leqslant 0.777$. Figure 1 shows that the numerical solution for these parameters even respects $\|\mathbf{v}(t)\| \leqslant \|\mathbf{v}(0)\| = 1/\sqrt{2} = 0.707$ for the maximally admitted value of initial vector length. Furthermore, adding a Hamiltonian contribution as, e.g.,

$$A = \frac{1}{2}\begin{pmatrix} 0 & -2 & 1 \\ 2 & 0 & -1 \\ -1 & 1 & 0 \end{pmatrix}, \tag{36}$$

introduces oscillations which, however, do not affect the norm decay.

(II) Consider a more general kernel like, for instance,

$$C(t) = \frac{1}{20}\begin{pmatrix} -\operatorname{sech}(t) & e^{-t}\cos(5\,t) & \frac{1}{2}\,[1+100\,t^2]^{-1} \\ e^{-t}\cos(5\,t) & -2\,[1+t^2]^{-2} & \frac{1}{4}\,e^{-t}\sin^2(5\,t) \\ \frac{1}{2}\,[1+100\,t^2]^{-1} & \frac{1}{4}\,e^{-t}\sin^2(5\,t) & -2\,e^{-t} \end{pmatrix}, \tag{37}$$

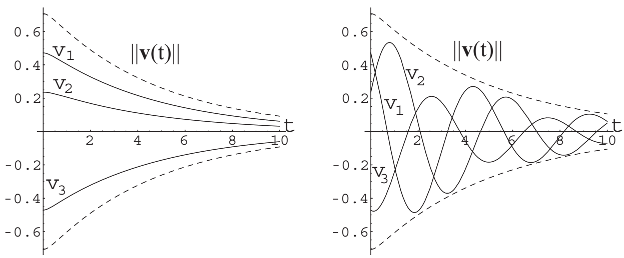

Fig. 1. Time-dependence of the 3 coherence-vector components as solutions of (9) for input quantities as defined in (34) and (36) with initial conditions $v_1(0) = -v_3(0) = \sqrt{2}/3$, $v_2(0) = 1/3\,\sqrt{2}$. The norm obviously satisfies $\|\mathbf{v}(t)\| \leqslant \|\mathbf{v}(0)\|$.

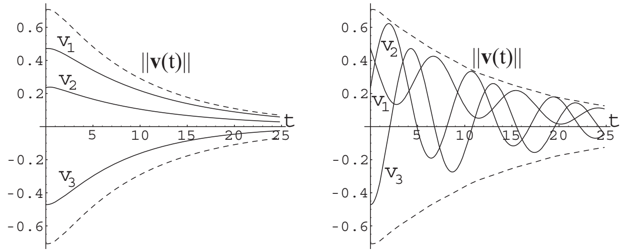

Fig. 2. Time-dependence of the 3 coherence-vector components as solutions of (9) for input quantities as defined in (37) and (38) with initial conditions $v_1(0) = -v_3(0) = \sqrt{2}/3$, $v_2(0) = 1/3\sqrt{2}$. The norm obviously satisfies $\|\mathbf{v}(t)\| \leqslant \|\mathbf{v}(0)\|$.

together with the same inhomogeneity as in (I). Here, the time-dependent norms have been calculated numerically, and from exponential fits one deduces the parameter values $\{L = \|Q\| = 0.102, \gamma = 0.9, M = 1, \alpha = 0.075\}$ with resulting bound $\|\mathbf{v}(t)\| \leqslant 0.937$. Again, if the dynamics includes an additional Hamiltonian contribution

$$A = \frac{1}{10}\begin{pmatrix} 0 & -1 & 3 \\ 1 & 0 & -6 \\ -3 & 6 & 0 \end{pmatrix}, \tag{38}$$

and even an initial state vector of maximum length is chosen, the time-dependent vector norm exhibits essentially the same decreasing behavior as for $A = 0$, as is shown in Fig. 2.

In conclusion, conditions (16–18) are very helpful for testing suitable trial functions for kernel and inhomogeneity in Eq. (9). One should notice that solutions bounded by (6) provide a density matrix which satisfies all von Neumann conditions only in case of $n = 2$. For $n \geqslant 3$ this bound is necessary but not yet completely sufficient to guarantee positivity. Even if the latter is likely to hold an extra test should be made. In brief, for $n = 2$ this is due to a 1-1-correspondence between the length of the coherence-vector and the only free eigenvalue of the densiy matrix, whereas for $n \geqslant 3$ there are two or more free eigenvalues, and uniqueness is lost. Of course, state space is convex but for $n \geqslant 3$ it is no longer a full Bloch sphere as is the case for $n = 2$. It is for this reason that a more general formulation of inequalities on input quantities is urgent in order to simultaneously guarantee positivity of the density matrix. Some successful progress in this direction seems possible and is currently under investigation.

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