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Virtues and Limitations of Markovian Master Equations with a Time-Dependent Generator

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A Markovian master equation with time-dependent generator is constructed that respects basic constraints of quantum mechanics, in particular the von Neumann conditions. For the case of a two-level system, Bloch equations with time-dependent parameters are obtained. Necessary conditions on the latter are formulated. By employing a time-local counterpart of the Nakajima–Zwanzig equation, we establish a relation with unitary dynamics. We also discuss the relation with the weak-coupling limit. On the basis of a uniqueness theorem, a standard form for the generator of time-local master equations is proposed. The Jaynes–Cummings model with atomic damping is solved. The solution explicitly demonstrates that reduced dynamics can be described by time-local master equations only on a finite time interval. This limitation is caused by divergencies in the generator. A limit of maximum entropy is presented that corroborates the foregoing statements. A second limiting case demonstrates that divergencies may even occur for small perturbations of the weak-coupling regime.

KEY WORDS: Quantum dissipation; Markovian master equations; Gorini–Kossakowski–Sudarshan–Lindblad generators; Jaynes–Cummings model.

1. INTRODUCTION

Almost 25 years ago, Gorini, Kossakowski, and Sudarshan,[(1)] as well as Lindblad,[(2)] made a contribution of axiomatic character to the quantum theory of dissipative systems. Specializing to the case of constant generators, these authors managed to construct the most general Markovian master equation, accordant with one single principle: validity of the representation $\Lambda(t)\,\rho = \sum_j W_j(t)\,\rho\,W_j^\dagger(t)$ for the quantum dynamical map $\Lambda(t)$ governing the evolution in time of a reduced density operator ρ.[(3–5)]

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An axiomatic approach offers the possibility to find out at one blow, which types of dissipative dynamics are tolerated by the basic principles of quantum mechanics. For instance, from the above-mentioned master equation one learns that exponential decay of a two-level system is driven by at most three different damping constants, which must satisfy certain inequalities.[6] As can be seen from the standard Bloch equations,[7–10] violation of these may impair the positivity of the density matrix for large times.[11]

If one plans to apply Markovian master equations to concrete problems in, e.g., quantum optics or nuclear magnetism, then one cannot exclusively work in an axiomatic fashion. One has to make a connection to unitary dynamics, by deriving generators on the basis of the Schrödinger equation. As regards the case of constant generators, this task can be accomplished with the help of either the weak-coupling[12] or the singular-coupling[13] limit.

Constant generators, which originate from mathematically less rigorous procedures, frequently disobey the axiomatic constraints. Some derivations, however, do not merit to be set aside, in view of their relevance to experiment or innovative power for theory. Examples are adiabatic elimination of fast variables,[14, 15] avoidance of the rotating-wave approximation,[16, 17] and nonperturbative study of atomic decay.[18]

To avoid losing those constant generators that are physically useful, but occasionally violate the positivity of the density operator, a mending trick was proposed in the literature.[15, 19, 20] By adapting the initial conditions, one can make sure that states with negative probability do not show up at physically relevant times. The initial density operator is sacrificed under the argument that a Markovian master equation with constant generator is anyhow incapable of providing a faithful description for small times, when the dynamics is still reversible.

The unsatisfactory state of affairs as outlined above, and in particular the last observation, motivates us to enlarge the family of sound Markovian master equations by dropping the concept of constant generators. We shall pursue the axiomatic approach, working with time-dependent generators.

In our opinion, the restriction to constant generators is too stringent sometimes. We cannot expect that each derivation of a Markovian master equation will allow us to render the generator independent of time. Indeed, time-local master equations were already employed in studying a variety of problems, including description of dissipative dynamics in the presence of external forces,[21] quantization of an electromagnetic field inside a lossy optical cavity,[22] application of the stochastic wave-function method to dynamics of quasi-irreversible character,[23] exact solution of a model for quantum mechanical dissipation,[24] and quantum mechanical description

of a damped oscillator.[25] As appears from refs. 26–28, the demand for time-dependent generators also exists among experimentalists.

Our paper is organized as follows. In the next section, we derive sufficient conditions for extending the axiomatic work on constant generators to the time-dependent case. For the Bloch equations with time-dependent parameters, the issue of positivity is studied in detail. A relation between Markovian master equations and unitary dynamics is established in Section 3. With the help of a uniqueness theorem, proved in Appendix A, a standard form for generators is derived. An important complication regarding practical employment of Markovian master equations, is discussed in Section 4. Use is made of the solution of the Jaynes–Cummings model with atomic damping, which is obtained in Appendix B. A summary of our findings is presented in Section 5.

Before closing, a few words must be said about terminology. Following original definitions, we call in this work the master equation $d\rho(t)/dt = L(t)\,\rho(t)$ either nonstationary Markovian or time-local. In the physics literature, the term Markovian usually indicates that generator L is constant. The case of a time-dependent generator may be termed then non-Markovian. As appears from the footnote on p. 274, this convention was adopted in ref. 11.

2. CONSTRUCTION OF A TIME-LOCAL MASTER EQUATION

We consider a non-relativistic open quantum system $\mathscr{S}$ that is composed of N non-degenerate levels. The dynamical behavior is described by a density matrix $\rho(t)$ that belongs to the Banach space $\mathscr{M}(N)$ of complex $(N \times N)$ matrices, and acts on the Hilbert space C^N of complex N dimensional vectors. An arbitrary set of linearly independent matrices spanning the space $\mathscr{M}(N)$, will be denoted as $\{B_k\}_{k=1}^{N^2}$. Sometimes, use will be made of the more specific basis set $\{F_k\}_{k=0}^{N^2-1}$. The new matrices possess the convenient property $\mathrm{Tr}(F_m F_n^\dagger) = \delta_{mn}$ for $0 \leqslant m,\, n \leqslant N^2 - 1$. Matrix F_0 is equal to $N^{-1/2}\mathbf{1}_N$, so all other matrices of the set are traceless.

We interpret the set $\{\langle \phi_j \mid \rho(t) \mid \phi_j \rangle\}_{j=1}^{N}$ as a normalized probability distribution $\{p_j \mid p_j \geqslant 0,\ \sum_{k=1}^{N} p_k = 1\}_{j=1}^{N}$; the vectors $\{|\phi_j\rangle\}_{j=1}^{N}$ make up an arbitrary orthonormal basis of the Hilbert space. Therefore, on the density matrix the von Neumann conditions

$$\mathrm{Tr}\,\rho(t) = 1, \qquad \rho(t) \geqslant 0 \tag{1}$$

must be imposed. It follows that the matrix $\rho(t)$ is Hermitian.

We set out to construct a Markovian master equation with time-dependent generator that respects the constraints (1). Guided by the structure of the quantum dynamical map $\Lambda(t)$ as given in the Introduction, we start from the inequality

$$W_m(t)\,\rho(t)\,W_m^\dagger(t) \geqslant 0 \tag{2}$$

For each integer m, element $W_m(t)$ of $\mathscr{M}(N)$ is assumed to be continuous in time. Then the Volterra-type equation

$$\tilde{\rho}(t) = \rho(t_0) + \lambda \sum_{m=1}^{J} \int_{t_0}^{t} ds\; W_m(s)\,\tilde{\rho}(s)\,W_m^\dagger(s) \tag{3}$$

is well defined. Parameter λ is real and positive. The iterative solution of Eq. (3) converges uniformly on each time interval $t_0 \leqslant t \leqslant T$. With the help of induction in the order of iteration, one proves that it can be represented as

$$\tilde{\rho}(t) = \rho(t_0) + \sum_{n=1}^{\infty} \lambda^n \sum_{k,l=1}^{N^2} \tilde{\Theta}_{kl}^{(n)}(t, t_0)\,B_k \rho(t_0)\,B_l^\dagger \tag{4}$$

where the matrix $[\tilde{\Theta}_{kl}^{(n)}(t, t_0)]_{k,l=1}^{N^2} \in \mathscr{M}(N^2)$ is positive for $t > t_0$, and vanishing for $t = t_0$. In Eqs. (3) and (4) tildes have been added so as to stress that the equality $\mathrm{Tr}\,\tilde{\rho}(t) = \mathrm{Tr}\,\rho(t_0)$ is false in general.

The condition of trace conservation can be fulfilled upon transforming the operator $\tilde{\rho}(t)$ in the following vein:

$$\rho(t) = Q_+(t)\,\tilde{\rho}(t)\,Q_+^\dagger(t) \tag{5}$$

$$Q_\eta(t) = \mathsf{T}_\eta \exp\left[\eta \int_{t_0}^{t} ds\, K(s)\right] \tag{6}$$

with $\eta = +, -$. Operator $\mathsf{T}_{+(-)}$ orders a product of matrices such that time arguments increase (decrease) when going from the right to the left. Owing to (2), the transformed matrix $\rho(t)$ is still positive. There is no restriction on the choice of the matrix $K(t) \in \mathscr{M}(N)$; just for convenience, continuity in time will be assumed.

By employing the identities (5) and (6), as well as

$$Q_+(t)\,Q_-(t) = \mathbf{1}_N \tag{7}$$

we derive from (3)

$$\frac{d}{dt}\rho(t) = K(t)\,\rho(t) + \rho(t)\,K^\dagger(t) + \sum_{m=1}^{J} \hat{W}_m(t)\,\rho(t)\,\hat{W}_m^\dagger(t) \tag{8}$$

Parameter λ has been set equal to unity, and the transformed matrix

$$\hat{W}_m(t) = Q_+(t)\, W_m(t)\, Q_-(t) \tag{9}$$

has been introduced. We ensure trace conservation by letting the trace of the right-hand side of (8) vanish. This surely happens if

$$K(t) + K^\dagger(t) + \sum_{m=1}^{J} \hat{W}_m^\dagger(t)\, \hat{W}_m(t) = 0 \tag{10}$$

Hence, the Hermitian matrix

$$H(t) = \frac{i}{2}\,[K(t) - K^\dagger(t)] \tag{11}$$

can be freely chosen. The choice of the set $\{\hat{W}_m(t)\}_{m=1}^{J}$ is also free, because matrices $Q_{+,-}(t)$ are invertible.

The structure of Eq. (8) can be simplified through use of a fixed basis for the space $\mathscr{M}(N)$. By writing

$$\hat{W}_m(t) = \sum_{k=1}^{N^2} c_{mk}(t)\, B_k \tag{12}$$

we maintain full generality. The complex coefficients $\{c_{mk}(t)\}$ are not subject to any constraints, so the sum

$$d_{kl}(t) = \sum_{m=1}^{N^2} c_{mk}(t)\, c_{ml}^*(t) \tag{13}$$

represents the (kl) element of any positive $(N^2 \times N^2)$ matrix. Integer J has been set equal to N^2.

Substitution of (12) into (8), and use of (10), provides us with the time-local master equation in final shape. It is given by

$$\frac{d}{dt}\,\rho(t) = L(t)\,\rho(t) \tag{14}$$

with the time-dependent generator

$$L(t) = L_H(t) + L_D(t) \tag{15}$$

$$L_H(t)\, M = -i[H(t), M] \tag{16}$$

$$L_D(t)\, M = \tfrac{1}{2} \sum_{k,l=1}^{N^2} d_{kl}(t)\{[B_k M, B_l^\dagger] + [B_k, M B_l^\dagger]\} \tag{17}$$

and $M \in \mathscr{M}(N)$ arbitrary. Any matrix commutes with the unit matrix, so the property

$$\mathrm{Tr}\, H(t) = 0 \tag{18}$$

can be employed at will. By means of the choice $\{B_k = F_{k-1}\}_{k=1}^{N^2}$ one can reduce the dimension of the damping matrix $[d_{kl}(t)]$ to $[(N^2-1)\times(N^2-1)]$; all contributions with $k=0$ or $l=0$ can be moved to Hamiltonian $H(t)$. For the sake of clarity, we emphasize that our construction of master equation (14) puts into existence two conditions, namely, $H^\dagger(t) = H(t)$ and $[d_{kl}(t)] \geqslant 0$.

Defining a quantum dynamical map $\Lambda(t, t_0)$ according to

$$\rho(t) = \Lambda(t, t_0)\, \rho(t_0) \tag{19}$$

we deduce from solution (4) and transformation (5) that the evolution equations (14)–(17) generate a map of the following standard[3–5] form:

$$\Lambda(t, t_0)\, M = \sum_{k,l=0}^{N^2-1} \Theta_{kl}(t, t_0)\, F_k M F_l^\dagger \tag{20}$$

with $M \in \mathscr{M}(N)$ arbitrary. The initial condition on map Λ is translated as

$$\Theta_{kl}(t_0, t_0) = N\delta_{k,0}\delta_{l,0} \tag{21}$$

By construction, the matrix $[\Theta_{kl}(t, t_0)]$ is differentiable for all $t \geqslant t_0$ and positive. Result (20) thus demonstrates that the positivity of the initial density matrix $\rho(t_0)$ is preserved by the evolution equations (14)–(17). On the other hand, since conservation of trace is manifest from (14)–(17), the map (20) surely satisfies the constraint

$$\sum_{k,l=0}^{N^2-1} \Theta_{kl}(t, t_0)\, F_l^\dagger F_k = \mathbf{1}_N \tag{22}$$

Note that the vanishing of $\mathrm{Tr}(M\rho)$ for arbitrary density matrix ρ implies the vanishing of matrix M.

In case the Hamiltonian and damping matrix do not depend on time, prescription (15)–(17) generates a well-known class of Markovian master equations. It was created during the mid seventies by Gorini, Kossakowski, and Sudarshan,[1] as well as by Lindblad.[2] Focusing on Eq. (14) with generator L independent of time, these authors proved that L has structure (15)–(17), with $H^\dagger = H$, $[d_{kl}] \in \mathscr{M}(N^2-1)$, $[d_{kl}] \geqslant 0$, and $\{B_k = F_k\}_{k=1}^{N^2-1}$, if and only if the corresponding quantum dynamical map has structure (20)–(22), with $[\Theta_{kl}(t, t_0)] \geqslant 0$.

Sometimes,[29] the construction of a constant generator (15)–(17) is carried out on the basis of the following argument: for safeguarding the positivity of the density matrix, it is sufficient to prove that the derivative $d\lambda(t)/dt$ of any eigenvalue $\lambda(t)$ of the density matrix is nonnegative whenever $\lambda(t)$ becomes zero. Unfortunately, this argument ignores the possibility of inflection points. For any physically acceptable dynamics the derivative of $\lambda(t)$ must become zero whenever one has $\lambda(t) = 0$. It is not the first, but rather the second derivative of $\lambda(t)$ that rules over the positivity of the density matrix.

Returning to the case of a time-dependent generator, we diagonalize in (17) $[d_{kl}(t)]$, which is positive by construction. The ensuing master equation has the so-called Lindblad form, and appeared in the recent literature.[23] We should also mention that time-local master equations were in use for describing a quantized electromagnetic field inside a lossy optical resonator.[22] However, systematic classifications of nonstationary Markovian master equations, and more specifically, general theorems on the relation between such equations and the quantum dynamical map (20), have not been published as yet.

By deriving the evolution equations (14)–(17), we have made a modest start in tackling the complicated problem of extending current knowledge on Markovian master equations. We have formulated conditions on Eqs. (14)–(17), which guarantee that their solution evolves in line with a map (20)–(22), for which the matrix $[\Theta_{kl}(t, t_0)]$ is positive. As pointed out earlier, for the case of a constant generator these conditions are both necessary and sufficient. It is important to recognize that the situation becomes much different if the generator depends on time. An example will be presented below.

Before treating this example, we shall develop some feeling for the difficulties provoked by time-dependent generators. Let us walk the route from Eqs. (19)–(22) to Eqs. (14)–(17) that demonstrates for the case of a constant generator the necessity of the earlier-mentioned conditions on the Hamiltonian and damping matrix. We differentiate (19) with respect to t, employ (20)–(22), and choose time t equal to t_0. Comparison of the resulting identity to (14) reveals that generator $L(t_0)$ takes on structure (15)–(17), the basis set of matrices being $\{F_k\}_{k=0}^{N^2-1}$. The Hamiltonian and damping matrix come out as

$$H(t_0) = \frac{i}{2} N^{-1/2} \sum_{k=1}^{N^2-1} \lim_{\varepsilon \downarrow 0} \Theta_{k0}(t_0 + \varepsilon, t_0)\, \varepsilon^{-1} F_k + \text{h.c.} \tag{23}$$

$$d_{kl}(t_0) = \lim_{\varepsilon \downarrow 0} \Theta_{kl}(t_0 + \varepsilon, t_0)\, \varepsilon^{-1} \geqslant 0 \tag{24}$$

with $1 \leqslant k,\ l \leqslant N^2-1$. Matrix (23) is Hermitian, and matrix (24) positive. However, as long as the Hamiltonian and damping matrix depend on time, this conclusion does not offer us any information about the properties of the generator at times later than t_0.

It is instructive to examine the quantum dynamical map that belongs to the Bloch equations with time-dependent coefficients. These read

$$\begin{aligned} \frac{d}{dt}\, p(t) &= -\left[\gamma_\perp(t) + i\omega(t)\right] p(t) \\ \frac{d}{dt}\, d(t) &= -\gamma_{||}(t)\left[d(t) - d_\infty(t)\right] \end{aligned} \tag{25}$$

where all coefficients $\gamma_\perp(t)$, $\gamma_{||}(t)$, $d_\infty(t)$, and $\omega(t)$ are real valued. The polarization and inversion are defined as $p(t) = \langle 1|\ \rho(t)\ |2\rangle$ and $2d(t) = \langle 2|\ \rho(t)\ |2\rangle - \langle 1|\ \rho(t)\ |1\rangle$, respectively. In Eqs. (14)–(17) we have made the choices $N=2$, $B_1 = F_0 = \mathbf{1}_2/\sqrt{2}$, $\{B_{k+1} = F_k = \sigma_k/\sqrt{2}\}_{k=1}^{3}$, $H(t) = \omega(t)\ \sigma_3/2$, $2d_{22}(t) = 2d_{33}(t) = \gamma_{||}(t)$, $d_{44}(t) = \gamma_\perp(t) - \gamma_{||}(t)/2$, and $d_{23}(t) = d_{32}^*(t) = i\gamma_{||}(t)\ d_\infty(t)$. The remaining elements of the damping matrix equal zero. The set $\{\sigma_k\}_{k=1}^{3}$ contains the Pauli matrices.

Upon comparing the solution of (25) to the prescription (20) for $\Lambda(t, t_0 = 0)$, we arrive at the following results for nonzero matrix elements:

$$\begin{aligned} \Theta_{00} = \tfrac{1}{2} + \tfrac{1}{2}\alpha_2 + \mathrm{Re}\ \alpha_3, \qquad \Theta_{33} = \tfrac{1}{2} + \tfrac{1}{2}\alpha_2 - \mathrm{Re}\ \alpha_3, \qquad \Theta_{11} = \Theta_{22} = \tfrac{1}{2} - \tfrac{1}{2}\alpha_2 \\ \Theta_{03} = \Theta_{30}^* = -\alpha_1\alpha_2 - i\ \mathrm{Im}\ \alpha_3, \qquad \Theta_{12} = \Theta_{21}^* = i\alpha_1\alpha_2 \end{aligned} \tag{26}$$

Time arguments have been omitted. The new functions are given by

$$\begin{aligned} \alpha_1(t) &= \int_0^t ds\ \gamma_{||}(s)\ d_\infty(s) \exp\left[\int_0^s du\ \gamma_{||}(u)\right] \\ \alpha_2(t) &= \exp\left[-\int_0^t ds\ \gamma_{||}(s)\right] \\ \alpha_3(t) &= \exp\left[-\int_0^t ds\ \gamma_\perp(s) - i\int_0^t ds\ \omega(s)\right] \end{aligned} \tag{27}$$

Matrix $[\Theta_{kl}(t, t_0 = 0)]$ is positive iff two inequalities hold true

$$1 + 2\ |\alpha_1(t)| \leqslant \alpha_2^{-1}(t), \qquad 4\alpha_1^2(t)\ \alpha_2^2(t) + 4\ |\alpha_3(t)|^2 \leqslant [1 + \alpha_2(t)]^2 \tag{28}$$

These are surely satisfied if for all positive times t one has

$$2\int_0^t ds\ \gamma_\perp(s) \geqslant \int_0^t ds\ \gamma_{||}(s) \geqslant 0, \qquad |d_\infty| \leqslant 1/2 \tag{29}$$

where parameter d_∞ no longer depends on time. The above conditions relate to the global properties of damping coefficients $\gamma_\perp(t)$ and $\gamma_{||}(t)$. They are much weaker than the conditions accompanying evolution equation (14); for the present case the latter read $2\gamma_\perp(t) \geqslant \gamma_{||}(t) \geqslant 0$, $|d_\infty| \leqslant 1/2$.[30]

From the above example one learns that it is certainly rewarding to try to minimize for Eqs. (14)–(17) the package of conditions guaranteeing a physically acceptable structure of the quantum dynamical map. We emphasize that the earlier-discussed constraints on $\Lambda(t, t_0)$ are well-grounded. Any quantum dynamical map, which is derived from the Schrödinger equation by factorizing the initial state and taking a partial trace, can be represented as (20), with $[\Theta_{kl}(t, t_0)]$ positive.[3–5]

The foregoing remarks give rise to the question whether it is possible to make a direct contact between the time-local master equation itself and the so-called reduced dynamics. This point will be addressed in the next section.

3. RELATION TO REDUCED DYNAMICS

As long as we do not construct a solid bridge between the Schrödinger equation and the evolution equations (14)–(17), the status of the latter remains purely phenomenological. Adopting the standard method for deriving reduced dynamics,[7, 31] we model the sinks of energy in system $\mathscr{S}$ through a coupling to a reservoir $\mathscr{R}$. In general, description of the reservoir requires a Hilbert space $\mathscr{H}_\mathscr{R}$ of infinite dimension. The evolution in time of system and reservoir is governed by the density operator

$$\rho_{\mathscr{S}\mathscr{R}}(t) = \exp[-iH_{\mathscr{S}\mathscr{R}}(t-t_0)/\hbar]\, \rho_{\mathscr{S}\mathscr{R}}(t_0) \exp[iH_{\mathscr{S}\mathscr{R}}(t-t_0)/\hbar] \tag{30}$$

where the Hamiltonian

$$H_{\mathscr{S}\mathscr{R}}/\hbar = H_\mathscr{S} \otimes \mathbf{1}_\mathscr{R} + \mathbf{1}_\mathscr{S} \otimes H_\mathscr{R} + \lambda V_{\mathscr{S}\mathscr{R}} \tag{31}$$

is a sum of self-adjoint operators.

The Hilbert space $\mathbb{C}^N$ of $\mathscr{S}$ is spanned by the orthonormal set $\{|k\rangle\}_{k=1}^N$ of eigenvectors of $H_\mathscr{S}$; the eigenvalues $\{\varepsilon_k\}_{k=1}^N$ of the latter are assumed to be non-degenerate. The interaction potential can be factorized now as follows[32]:

$$V_{\mathscr{S}\mathscr{R}} = \sum_{\alpha=1}^{N^2} V_\alpha \otimes U_\alpha \tag{32}$$

If the double index α equals (kl), then by definition the potential V_α is equal to the matrix $|k\rangle\langle l|$, with $1 \leqslant k,\ l \leqslant N$. In the sequel the same notation will be used. We assume that the reservoir potentials $\{U_\alpha\}$ are bounded in sup-norm.

As initial condition for the full density operator we choose a direct product of density operators

$$\rho_{\mathscr{S}\mathscr{R}}(t_0) = \rho_{\mathscr{S}} \otimes \rho_{\mathscr{R}} \tag{33}$$

Then the reduced density matrix $\rho_{\mathscr{S}}(t)$ governing the dynamical behavior of $\mathscr{S}$, is found as

$$\begin{aligned}\rho_{\mathscr{S}}(t) &= \mathrm{Tr}_{\mathscr{R}}\{\exp[-iH_{\mathscr{S}\mathscr{R}}(t-t_0)/h]\ \rho_{\mathscr{S}} \otimes \rho_{\mathscr{R}} \exp[iH_{\mathscr{S}\mathscr{R}}(t-t_0)/h]\} \\ &= \Lambda(t, t_0)\ \rho_{\mathscr{S}}\end{aligned} \tag{34}$$

Upon inserting for density operator $\rho_{\mathscr{R}}$ its diagonal representation, one sees that $\Lambda(t, t_0)$ possesses the structure (20). Matrix $[\Theta_{kl}(t, t_0)]$ turns out to be positive.

On the basis of result (34), an integral equation for the reduced density matrix can be derived. In the interaction picture this Nakajima–Zwanzig equation possesses a time-local counterpart,[33–36, 11] which reads

$$\frac{d}{dt}\,\rho_{\mathscr{S}}^{(I)}(t) = L(t, t_0)\,\rho_{\mathscr{S}}^{(I)}(t) \tag{35}$$

The generator $L(t, t_0)$ acts on $\mathscr{M}(N)$, and is linear. In explicit terms the generator is given by a series expansion in the coupling constant λ, which can be found in ref. 11 for the case that $\rho_{\mathscr{R}}$ and $H_{\mathscr{R}}$ commute.

By utilizing the boundedness of $V_{\mathscr{S}\mathscr{R}}$, as well as the continuity in t of $\exp(iH_{\mathscr{S}}t)$ and $\exp(iH_{\mathscr{R}}t)$, one can prove that for any $M \in \mathscr{M}(N)$ the series $L(t, t_0)\,M$ is convergent and continuous on an interval $t_0 \leqslant t \leqslant t_0 + a$, where the bound a lies between zero and infinity.[11] Construction of superoperator $L(t, t_0)$ entails inversion of $\Lambda(t, t_0)$ in the interaction picture. If t exceeds the value of $t_0 + a$, the series expansion for the inverse starts to diverge. Consequently, evolution equation (35) no longer exists. This temporal restriction cannot be circumvented by commencing integration at a time t_1 later than $t_0 + a$. Density matrix $\rho_{\mathscr{S}}^{(I)}(t_1)$ cannot be freely chosen, as it is completely determined by map (34). Incidentally, the particularity of instant $t = t_0$ is also illustrated by the property $L(t = t_0, t_0) = 0$,[11] which causes the derivative in (35) to vanish at time t_0.

We shall bring the time-local Nakajima–Zwanzig equation (35) onto the structure (15)–(17). To that end, we merely need the following characteristics of the generator[11]:

$$\begin{aligned} [L(t, t_0)\, M]^\dagger &= L(t, t_0)\, M^\dagger \\ \mathrm{Tr}_{\mathscr{S}}[L(t, t_0)\, M] &= 0 \end{aligned} \tag{36}$$

with matrix $M \in \mathscr{M}(N)$ arbitrary. They safeguard normalization and Hermiticity of the reduced density matrix.

By exploiting the linearity of $L(t, t_0)$ and the completeness of set $\{|k\rangle\}_{k=1}^{N}$, we obtain the identity

$$L(t, t_0)\, M = \sum_{\alpha,\beta=1}^{N^2} L_{\alpha\beta}(t, t_0)\, V_\alpha M V_\beta^\dagger \tag{37}$$

For $1 \leqslant k, l, m, n \leqslant N$ we have defined

$$L_{(kl)(mn)}(t, t_0) = \langle k|\, [L(t, t_0)\, |l\rangle\langle n|]\, |m\rangle \tag{38}$$

Between the square brackets $L(t, t_0)$ acts on the matrix $|l\rangle\langle n|$. The new matrix on the left-hand side of (38) inherits from the superoperator its continuity in time, its convergence on interval $t_0 \leqslant t \leqslant t_0 + a$, and properties (36). These become

$$L^*_{\beta\alpha}(t, t_0) = L_{\alpha\beta}(t, t_0) \tag{39}$$

$$\sum_{k=1}^{N} L_{(kl)(kn)}(t, t_0) = 0 \tag{40}$$

The last property implies that the right-hand side of (37) is of structure (15)–(17). The Hamiltonian $H(t, t_0)$ equals zero for the moment.

To make the division (15) explicit, we transform the matrices $\{V_\alpha\}$ of decomposition (37) to a basis set containing the unit matrix. Opting for the set $\{F_k\}_{k=0}^{N^2-1}$ and employing the representation

$$M = \sum_{k=0}^{N^2-1} \mathrm{Tr}(M F_k^\dagger)\, F_k \tag{41}$$

we find that $L(t, t_0)$ has the structure (15)–(17) again, with $\{B_k = F_k\}_{k=1}^{N^2-1}$ of course. Now the Hamiltonian differs from zero

$$H(t, t_0) = \frac{i}{2N} \sum_{\alpha,\beta=1}^{N^2} \mathrm{Tr}(V_\beta)\, L_{\alpha\beta}(t, t_0)[V_\alpha - \mathrm{Tr}(V_\alpha)\, \mathbf{1}_N/N] + \text{h.c.} \tag{42}$$

Property (18) is manifestly true. For the damping matrix of dimension $[(N^2-1)\times(N^2-1)]$ the following form appears:

$$d_{kl}(t, t_0) = \sum_{\alpha,\beta=1}^{N^2} \mathrm{Tr}(V_\alpha F_k^\dagger)\,\mathrm{Tr}(V_\beta F_l^\dagger)^*\, L_{\alpha\beta}(t, t_0) \tag{43}$$

with $1 \leqslant k, l \leqslant N^2-1$. Derivation of the last two results requires that property (40) be expressed with the help of the basis set $\{F_k\}_{k=0}^{N^2-1}$.

All of the above steps can also be carried out for the case that the right-hand side of (35) is truncated at some order in the coupling constant. Accordingly, the right-hand side of (43) can be regarded as a series expansion in λ, which converges uniformly on the interval $t_0 \leqslant t \leqslant t_0 + a$. Each term is continuous in t, and corresponds to a Hermitian matrix. Analogous observations can be made for (42). Moreover, one should realize that the expressions (42) and (43) are uniquely determined. This claim relies on the following statement, the proof of which is deferred to Appendix A: the equality

$$\sum_{k,l=0}^{N^2-1} c_{kl} F_k \rho_{\mathscr{S}} F_l^\dagger = 0 \tag{44}$$

is true for arbitrary density matrix $\rho_{\mathscr{S}}$ if and only if all constants $\{c_{kl}\}$ equal zero. Therefore, expression (42) is maximal in the sense that no further contributions to $H(t, t_0)$ can be extracted from the dissipative generator $L_D(t, t_0)$.

For quite a number of applications it is useful to represent the dissipative generator on the basis of the eigenstates of the free Hamiltonian $H_{\mathscr{S}}$. Making the choice $\{B_k = F_k\}_{k=1}^{N^2-1}$, we substitute (43) into expression (17). Employment of the representation $F_k = \sum_{\alpha=1}^{N^2} \mathrm{Tr}(F_k V_\alpha^\dagger)\, V_\alpha$ allows us to bring the dissipative generator onto the structure (17) again, with the replacement $\{B_k\} \to \{V_\alpha\}$ carried out. The new damping matrix $[d_{\alpha\beta}(t, t_0)]$ is of dimension $(N^2 \times N^2)$, and attains the form

$$\begin{aligned} d_{(kl)(mn)}(t, t_0) = {} & L_{(kl)(mn)}(t, t_0) - \delta_{kl} \frac{1}{N} \sum_{p=1}^{N} L_{(pp)(mn)}(t, t_0) \\ & - \delta_{mn} \frac{1}{N} \sum_{p=1}^{N} L_{(kl)(pp)}(t, t_0) + \delta_{kl}\delta_{mn} \frac{1}{N^2} \sum_{p,q=1}^{N} L_{(pp)(qq)}(t, t_0) \end{aligned} \tag{45}$$

As a check, one can evaluate the right-hand side of (15) with the help of results (40), (42), and (45). One recovers decomposition (37), which is identical to the left-hand side of (15).

By invoking the intermediate identity (35), we have made contact between Eqs. (14)–(17) and the old Nakajima–Zwanzig equation describing

reduced dynamics. In short, each density matrix of class (34) has been shown to obey a nonstationary Markovian master equation of type (14)–(17). Referring to the argument put forward near Eqs. (23) and (24), we emphasize that the damping matrices (43) and (45) are Hermitian, but not positive in general. Since the weak-coupling limit constitutes an exception to this statement, we should clarify its relation with (45).

The weak-coupling theorem of the literature[12] specifies conditions under which the limit $\lambda \to 0$ causes density matrix $\rho_{\mathscr{S}}^{(I)}(t/\lambda^2)$ to converge to the solution $\sigma(t)$ of a Markovian master equation with a constant generator L_0. The latter is fixed by the relations

$$\begin{aligned}
&\langle k|\,(L_0 M)\,|l\rangle \\
&= \sum_{m,n=1}^{N} \delta(\omega_{kl}+\omega_{nm},0) \int_{-\infty}^{+\infty} dt\, \exp(i\omega_{mk}t)\, c_{(nl)(mk)}(t) \langle m|\, M\, |n\rangle \\
&\quad - \sum_{m=1}^{N} \int_0^{\infty} dt\, [\exp(i\omega_{ml}t)\, c^*_{(lm)(lm)}(t) + \exp(i\omega_{km}t)\, c_{(km)(km)}(t)] \langle k|\, M\, |l\rangle
\end{aligned} \tag{46}$$

with $M \in \mathscr{M}(N)$ arbitrary. The frequencies ω_{kl} are defined as $\varepsilon_k - \varepsilon_l$. The correlation function must be taken as[32]

$$c_{\alpha\beta}(t) = \mathrm{Tr}_{\mathscr{R}}[\exp(iH_{\mathscr{R}}t)\, U_\alpha \exp(-iH_{\mathscr{R}}t)\, U_\beta^\dagger \rho_{\mathscr{R}}] \tag{47}$$

Owing to the symmetry relation $c^*_{\beta\alpha}(-t) = c_{\alpha\beta}(t)$, the first integral on the right-hand side of (46) covers the complete time axis.

In rederiving the standard result (46), we discard all contributions which are not of lowest, that is to say, quadratic order in λ. This truncation, the justification of which will not be addressed, is indicated by adding a superscript (2) to $L(t, t_0)$; the matrix $\rho_{\mathscr{S}}^{(I)(2)}(t)$ denotes the solution of the truncated master equation (35). From (38) and the series for generator $L(t, t_0)$ (ref. 11) one obtains

$$\begin{aligned}
L^{(2)}_{(kl)(mn)}(t, t_0 = 0) = &-\lambda^2 \delta_{kl} \exp(i\omega_{nm}t) \sum_{p=1}^{N} \int_0^t ds \exp(i\omega_{pn}s)\, c^*_{(mp)(np)}(s) \\
&+ \lambda^2 \exp[\,i(\omega_{kl}+\omega_{nm})\,t\,] \int_0^t ds \\
&\times [\exp(i\omega_{lk}s)\, c_{(nm)(lk)}(s) + \exp(i\omega_{mn}s)\, c^*_{(lk)(nm)}(s)] \\
&- \lambda^2 \delta_{mn} \exp(i\omega_{kl}t) \sum_{p=1}^{N} \int_0^t ds \exp(i\omega_{lp}s)\, c_{(kp)(lp)}(s)
\end{aligned} \tag{48}$$

By mimicking the proof presented in Appendix A of ref. 11, we can demonstrate that the weak-coupling theorem is also valid for the truncated version of (35). The mathematical statement reads

$$\lim_{\lambda \to 0} \sup_{0 \leqslant t \leqslant \tilde{a}} \| \rho_{\mathscr{S}}^{(I)(2)}(t/\lambda^2) - \sigma(t) \| = 0 \tag{49}$$

Constant $\tilde{a}$ can be chosen such that the time $\tilde{a}/\lambda^2$ lies within the domain of existence $0 \leqslant t \leqslant a$ for (35).[11]

Last, it must be verified that, as announced above, the damping matrix (45) becomes positive in the weak-coupling limit. Combination of (45) and (46) yields

$$\begin{aligned} d_{(kl)(mn)} = & \sum_{\{\omega_j\}} \int_{-\infty}^{+\infty} dt \exp(-i\omega_j t)\, \delta(\omega_{kl}, \omega_j)\, \delta(\omega_{mn}, \omega_j)\, c_{(nm)(lk)}(t) \\ & + \delta_{kl}\delta_{mn} \int_{-\infty}^{+\infty} dt \sum_{p,q=1}^{N} (\delta_{kq} - 1/N)(\delta_{mp} - 1/N)\, c_{(pp)(qq)}(t) \end{aligned} \tag{50}$$

The set $\{\omega_j\}$ contains all nonzero values of differences $\{\varepsilon_k - \varepsilon_l\}_{k,l=1}^{N}$. One proves that for each set $\{v_\alpha\} \subset \mathsf{C}$ the sum $\sum_{\alpha,\beta=1}^{N^2} d_{\alpha\beta} v_\alpha v_\beta^*$ is positive indeed; Bochner's theorem[37] should be applied to the function $\sum_{\alpha,\beta=1}^{N^2} c_{\alpha\beta}(t)\, w_\alpha w_\beta^*$, with set $\{w_\alpha\} \subset \mathsf{C}$ suitably chosen.

In the preceding, we have tacitly assumed that system $\mathscr{S}$ does not experience any external forces. For that reason, $H_{\mathscr{S}}$ has been taken independent of time. If the reverse is true, then one can still set up a weak-coupling limit for the reduced density matrix.[21] It creates a nonstationary Markovian master equation. Hence, in deriving Eqs. (14)–(17) from first principles, one is not obliged to pursue a route that passes through Eq. (35). An advantage of the alternative route is that generators exist at all times.

4. DAMPED JAYNES–CUMMINGS MODEL

The significance of studying properties and solutions of time-local master equations will stand or fall with the question whether the domains of existence of the involved generators extend to physically relevant times. If an observer does not record any appreciable changes in system $\mathscr{S}$ during an interval $t_0 \leqslant t \leqslant t_0 + a$, then one may criticize any attempt to explore the dynamical behavior of $\mathscr{S}$ within the framework of (35). In the following, several restrictions on domains of existence will be discussed for the case of a two-level atom.

To achieve a fair degree of flexibility, we assume a coupling to a big reservoir that brings about irreversibility, and on top of that, a coupling

to a small reservoir that is incapable of depleting $\mathscr{S}$ permanently. The coupling constants are called γ and λ, respectively. The small reservoir is made up by a single mode of the quantized electromagnetic field. The corresponding Hilbert space $\mathscr{H}_{\mathscr{F}}$ is spanned by the orthonormal photon-number states $\{(p!)^{-1/2}(a^\dagger)^p |0\rangle\}_{p=0}^{\infty}$, and has state $a|0\rangle$ as zero element. The ladder operators a and $a^\dagger$ satisfy the standard commutation relation $[a, a^\dagger] = 1$.

In the interaction picture the evolution equation for the density operator $\rho(t)$ describing the composite of atom and field mode, is given by

$$\begin{aligned}\frac{d}{dt}\rho^{(I)}(t) = &-i\lambda[\sigma_+ \otimes a + \sigma_- \otimes a^\dagger, \rho^{(I)}(t)] \\ &+ \gamma\{2(\sigma_- \otimes \mathbf{1}_{\mathscr{F}})\rho^{(I)}(t)(\sigma_+ \otimes \mathbf{1}_{\mathscr{F}}) \\ &- (i_+ \otimes \mathbf{1}_{\mathscr{F}})\rho^{(I)}(t) - \rho^{(I)}(t)(i_+ \otimes \mathbf{1}_{\mathscr{F}})\}\end{aligned} \tag{51}$$

The abbreviations $|2\rangle\langle 2| = i_+$, $|1\rangle\langle 1| = i_-$, $|2\rangle\langle 1| = \sigma_+$, and $|1\rangle\langle 2| = \sigma_-$ have been used. Equation (51) can be solved by means of a recipe developed in ref. 38. As initial condition we choose

$$\rho(t=0) = \rho_{\mathscr{S}} \otimes |p\rangle\langle p| \tag{52}$$

where $\rho_{\mathscr{S}}$ denotes an arbitrary density matrix of dimension (2×2), and $|p\rangle$ a photon-number state. Then the solution for the atomic density matrix $\rho_{\mathscr{S}}^{(I)}(t) = \mathrm{Tr}_{\mathscr{F}}[\rho^{(I)}(t)]$ can be expressed as

$$\langle 2|\, \rho_{\mathscr{S}}^{(I)}(t)\, |2\rangle = \langle 1|\, \rho_{\mathscr{S}}\, |1\rangle\, \beta_1(t) + \langle 2|\, \rho_{\mathscr{S}}\, |2\rangle\, \beta_2(t) \tag{53}$$

$$\langle 2|\, \rho_{\mathscr{S}}^{(I)}(t)\, |1\rangle = \langle 2|\, \rho_{\mathscr{S}}\, |1\rangle\, \beta_3(t) \tag{54}$$

The functions $\{\beta_j(t)\}$ are computed in Appendix B.

For $\gamma = 0$ Eq. (51) corresponds to a Jaynes–Cummings model with zero detuning.[39] The composite of atom and field mode undergoes a unitary evolution, so for matrix $\rho_{\mathscr{S}}^{(I)}(t)$ the complete theory of the previous section is available. The complication that the potentials $U_{(21)} = a$ and $U_{(12)} = a^\dagger$ are unbounded, can be taken care of. One starts to work with a basis set $\{|p\rangle\}_{p=0}^{J}$ for $\mathscr{H}_{\mathscr{F}}$, and eventually takes integer J to infinity.

The choice $\lambda = 0$ allows us to write $\rho^{(I)}(t) = \rho_{\mathscr{S}}^{(I)}(t) \otimes |p\rangle\langle p|$ for the solution of (51). The atomic density matrix satisfies a Markovian master equation with constant generator, which describes spontaneous emission of photons into electromagnetic modes other than the privileged one. The generator belongs to class (46); obviously, its domain of existence covers the complete positive time axis.

For the case $\lambda > 0$, $\gamma > 0$ the route from the level of unitary dynamics down to the dynamics of density matrix $\rho_{\mathscr{S}}(t)$ does not count one, but two instants at which a partial trace is evaluated. In between those, a weak-coupling limit is taken. Despite this difficulty, one can still construct a time-local master equation (35) for $\rho_{\mathscr{S}}^{(I)}(t)$. In the formalism presented in Section 3 of ref. 11, the superoperator A need not be specified. One has the freedom of matching operator $A\rho$ to the right-hand side $\lambda A_1 \rho + \gamma A_2 \rho$ of (51). Subsequently, the series expansion for generator $L(t, t_0)$ can be derived along the same lines as for the case $\gamma = 0$. Altogether, it is permitted to talk about a time-local master equation for the atomic density matrix within the setting of the damped Jaynes–Cummings model.

In calculating $L(t, t_0)$, we shall not resort to the series expansion. A much faster method adopts Eqs. (53)–(54) as a starting point. Substitution into (35), and employment of the linearity of $L(t, t_0)$ as well as the arbitrariness of the initial density matrix $\rho_{\mathscr{S}}$, leads to

$$
\begin{aligned}
L(t, t_0 = 0)[i_-] &= (i_+ - i_-)\left[\beta_2 \frac{d}{dt}\beta_1 - \beta_1 \frac{d}{dt}\beta_2\right] \Big/ (\beta_2 - \beta_1) \\
L(t, t_0 = 0)[i_+] &= (i_+ - i_-)\left[(\beta_2 - 1)\frac{d}{dt}\beta_1 - (\beta_1 - 1)\frac{d}{dt}\beta_2\right] \Big/ (\beta_2 - \beta_1) \\
L(t, t_0 = 0)[\sigma_+] &= \sigma_+ \left[\frac{d}{dt}\beta_3\right] \Big/ \beta_3
\end{aligned}
\tag{55}
$$

The result for matrix σ_- is fixed by the symmetry relation (36).

Identities (55) make the singular behavior of $L(t, t_0)$ tangible. Divergencies occur if one of the following equations is true:

$$
\beta_1(t) = \beta_2(t), \qquad \beta_3(t) = 0 \tag{56}
$$

The functions $\{\beta_j(t)\}$ have the initial values $\beta_1(t=0) = 0$, $\beta_2(t=0) = \beta_3(t=0) = 1$, and are continuous in time. Therefore, (56) confirms our earlier-made claim that on an interval $t_0 \leqslant t \leqslant t_0 + a$, with $0 < a < \infty$, generator $L(t, t_0)$ surely exists.

To make the case final, one should check that the singularities (56) really originate from the process of constructing a time-local counterpart of the Nakajima–Zwanzig equation. For the dynamics (51)–(52) the latter can be written as

$$
\rho_{\mathscr{S}}^{(I)}(t) = \rho_{\mathscr{S}} + \int_0^t ds\, K(t-s)\, \rho_{\mathscr{S}}^{(I)}(s) \tag{57}
$$

The integral kernel $K(t)$ is a superoperator, characterized by

$$\begin{aligned} K(t)[i_-] &= (i_+ - i_-)\, k_1(t), \\ K(t)[i_+] &= (i_+ - i_-)\, k_2(t), \\ K(t)[\sigma_+] &= \sigma_+ k_3(t) \end{aligned} \tag{58}$$

Laplace transformation of (57) according to

$$\hat{f}(z) = -i \int_0^\infty dt \exp(izt)\, f(t), \qquad \mathrm{Im} z > 0 \tag{59}$$

enables us to express the c-number functions as

$$\hat{k}_1(z) = \frac{-i\hat{\beta}_1(z)}{\hat{\beta}_2(z) - \hat{\beta}_1(z)}, \quad \hat{k}_2(z) = \frac{i[1 - z\hat{\beta}_2(z)]}{z[\hat{\beta}_2(z) - \hat{\beta}_1(z)]}, \quad \hat{k}_3(z) = \frac{i}{z\hat{\beta}_3(z)} \tag{60}$$

In Appendix B it becomes clear that for the initial condition (52) all transforms $\{\hat{k}_j(z)\}$ are meromorphic, the number of poles being finite. Hence, the kernel $K(t)$ exists on the complete positive time axis.

As a side remark, we point out that for the case of undamped Rabi oscillations, i.e., damping constant γ taken as zero and initial conditions (52) in force, all inverse transforms of (60) can be computed analytically. The integral kernel is stripped of its formal[40] character. One gets

$$k_j(t) = \mu_j \sin(\nu_j t) + \xi_j t \tag{61}$$

with $j = 1, 2, 3$. Into the linear term the equalities $\xi_1 = -\xi_2 = 4p(p+1)\,\lambda^2/(2p+1)$, $\xi_3 = -\lambda^2/(2p+1)$ must be inserted. The constants $\{\mu_j, \nu_j\}$ depend on p as well. In contrast to the density matrix $\rho_{\mathscr{S}}^{(I)}(t)$, $K(t)$ is not uniformly bounded on the positive time axis.

We are in a position now to investigate on which time intervals the integral equation (57) has a time-local counterpart. The choice $\gamma = 0$ makes analytic solution of the conditions (56) feasible. Generator $L(t, t_0 = 0)$ exists on the interval $[0, a]$, where the endpoint a may not exceed the value of $T_d \sqrt{1+p}/(\sqrt{1+p} + \sqrt{p})$. After a decay time of $T_d = \pi/(2\lambda\sqrt{1+p})$, an excited atom reaches the ground state through emission of a photon, so the interval $[0, a]$ definitely contains some relevant physics. This conclusion need not be modified if the ratio γ/λ is increased to a small nonzero value.

Any evolution exhibiting a series of almost undamped Rabi oscillations, may be rightly typified as quasi-reversible for times much smaller

than $1/\gamma$. Hence, for the case $\gamma/\lambda \ll 1$ the result $a < \infty$ was to be expected; one is far away from the truly irreversible case $\lambda = 0$, for which constant a surely equals infinity. Less obvious is the fact that for some nearly irreversible evolutions, a may become finite already. An example is contained in the atomic dynamics governed by master equation (51). Below it is argued that a slight perturbation of the weak-coupling regime causes a to decrease from infinity to a finite value; the initial photon number p is assumed to be high. Under such circumstances an intermediate coupling regime[23] does not exist. Meant is a choice of coupling constants, for which on the one hand a is infinitely large, and on the other hand, significant departures from the weak-coupling dynamics can be observed, for instance, oscillatory[38] or non-exponential decay.[41]

We shall work in the limit $p \to \infty$, $\lambda/\gamma \to 0$, with γt fixed, and the parameter $\varepsilon = \lambda^2 p/\gamma^2$ small. The material of Appendix B guides us to the approximate solutions

$$\begin{aligned} \beta_1(t) &= \varepsilon w^{-2} e^{-\gamma t}[e^{-w\gamma t} + e^{w\gamma t} - 2] + \mathcal{O}(\varepsilon^2) \\ \beta_2(t) &= w^{-2} e^{-\gamma t}\left[\left(\frac{1}{2} + \frac{w}{2} - \varepsilon\right) e^{-w\gamma t} + \left(\frac{1}{2} - \frac{w}{2} - \varepsilon\right) e^{w\gamma t} - 2\varepsilon\right] + \mathcal{O}(\varepsilon^2) \end{aligned} \tag{62}$$

where the root $w = (1 - 4\varepsilon)^{1/2}$ has been defined. Discarding the quadratic remainders $\mathcal{O}(\varepsilon^2)$, we infer from the first condition (56)

$$a = (2w\gamma)^{-1} \log\left(\frac{1+w}{1-w}\right) \tag{63}$$

Elaboration of the second condition (56) does not tighten this boundary.

We should estimate the maximum magnitude of ε for which bound (63) is still reliable. For that purpose, we also compute the second diagonal of the atomic density matrix in leading order. As for (62), the ensuing expression is exact at $t = 0$, and converges to zero for t tending to infinity. Consequently, for large times our approximation breaks down due to violation of trace conservation. At instant $t = a$, both diagonals do not depend on the initial density matrix $\rho_{\mathscr{S}}$; they behave as

$$\begin{aligned} \langle 1|\, \rho_{\mathscr{S}}^{(I)}(t=a)\, |1\rangle &= (\varepsilon^{-1/2} - 3\varepsilon^{1/2} - 2\varepsilon)\, w^{-2} \left(\frac{1-w}{1+w}\right)^{1/(2w)} \\ \langle 2|\, \rho_{\mathscr{S}}^{(I)}(t=a)\, |2\rangle &= (\varepsilon^{1/2} - 2\varepsilon)\, w^{-2} \left(\frac{1-w}{1+w}\right)^{1/(2w)} \end{aligned} \tag{64}$$

For $\varepsilon \downarrow 0$, $\varepsilon = 0.001$, and $\varepsilon = 0.01$ the sum of these expressions equals $1 + 0$, $0.995 + 0.935 \cdot 10^{-3}$, and $0.972 + 0.803 \cdot 10^{-2}$, respectively. All figures lie inside interval $[0, 1]$, and the relative errors in the trace amount to 0, 0.4, and 2%, respectively.

With ε taken as above, prediction (63) can be trusted. One finds that generator $L(t, t_0 = 0)$ does not exist beyond times $a = \infty$, $3.46/\gamma$, and $2.34/\gamma$, respectively. The last two bounds are surprisingly low. To see this in more detail, we choose $\lambda/\gamma = 0.01$ and $p = 100$, so that $\varepsilon = 0.01$ again. The process of spontaneous emission is weakly perturbed indeed. Nevertheless, there is a probability of $\exp(-\gamma a) = 0.1$ that up to time $t = a = 2.34/\gamma$ an excited atom does not decay. Then no phenomena of interest take place inside interval $[0, a]$.

A further restriction on domains of existence has its origin in a limit of maximum entropy. Departing from the solutions of Appendix B, one proves that the atomic density matrix as determined by (51) and (52), obeys the limit

$$\lim' \rho_{\mathscr{S}}^{(I)}(t) = \tfrac{1}{2}\,\mathbf{1}_2 \tag{65}$$

The prime stands for the prescription that variables λ/γ, λt, and p be taken to infinity such that products $(\gamma/\lambda)^{4/3}\,\lambda t$ and $(\gamma/\lambda)^{2/3}\,p$ remain constant. An analogous limit was established for the Jaynes–Cummings model with cavity damping.[38] A proof of (65) will appear in a forthcoming publication.

Since the initial density matrix $\rho_{\mathscr{S}}$ is arbitrary, (65) supplies us with the limits $\lim' \beta_1(t) = \lim' \beta_2(t) = 1/2$ and $\lim' \beta_3(t) = 0$; these imply that the conditions (56) are satisfied. Whenever limit (65) is realized during the evolution of the density matrix $\rho_{\mathscr{S}}^{(I)}(t)$, generator $L(t, t_0)$ becomes singular. For the Jaynes–Cummings model with cavity damping an example of such a realization can be found in ref. 38; there an evolution is scrutinized that is quasi-reversible for small times, and purely irreversible for long times. The transition between these stages is not immediate. Before commencing its exponential decay to the ground state, the atomic density matrix maximizes its entropy during a long time interval. The central state $\mathbf{1}_2/2$ thus plays the role of a temporal attractor.

All in all, the limit of maximum entropy (65) is of fundamental importance. It immediately tells us that, regardless of the details of the dynamics, time-local master equations are unsuited for furnishing a comprehensive description of the evolution of the two-level atom. In view of the results on maximum entropy that were obtained in ref. 38, we are confident that the limit (65) remains valid if in (52) the photon-number state is exchanged for a coherent state.

5. DISCUSSION

Before listing our main conclusions, we briefly review the setting in which we worked. Each state $\rho_{\mathscr{S}}$ of a closed quantum system $\mathscr{S}$ evolves as $U(t-t_0)\,\rho_{\mathscr{S}}\,U^{\dagger}(t-t_0)$ from $t=t_0$ onwards. Time t links unitary operator $U(t)$ to unit operator $U(t=0)=\mathbf{1}_{\mathscr{S}}$ in a continuous fashion. In case $\mathscr{S}$ suffers from energy losses to a reservoir $\mathscr{R}$, it is called open. Then the foregoing simple rotation must be replaced by a mapping $\Lambda(t, t_0)\,\rho_{\mathscr{S}} = \sum_j W_j(t-t_0)\,\rho_{\mathscr{S}}\,W_j^{\dagger}(t-t_0)$, where time t continuously connects the operators $W_j(t)$ and $W_j(t=0)=c_j\mathbf{1}_{\mathscr{S}}$ to each other. The constants $\{c_j\}$ are real. The property $\sum_j W_j^{\dagger}(t)\,W_j(t)=\mathbf{1}_{\mathscr{S}}$ comes in the place of unitarity. It safeguards conservation of trace.

In finite dimensions, the quantum dynamical map $\Lambda(t, t_0)$ can be cast into the structure defined by (20)–(22). A matrix $\Theta(t, t_0)$ emerges that is continuous in t and positive. In this work we regarded obedience to map (20), and all of its afore-mentioned properties, as a fundamental constraint on the evolution of a density matrix. One should realize that positivity of matrix $\Theta(t, t_0)$ is a stronger constraint than positivity of the density matrix. Furthermore, one should be well aware of the fact that a factorization of the full density operator at $t=t_0$ underlies the structure of map (20). The use of such an initial condition can be questioned.[42–45]

In this paper, we tried to disclose the main pros and contras regarding employment of time-local master equations for the study of open quantum systems. Throughout our treatment, we assumed the number of quantum levels of system $\mathscr{S}$ to be finite. Three positive arguments were presented.

First, any time-local master equation can be reshaped such that the structure (15)–(17) appears. Owing to the uniqueness theorem of Appendix A, one can derive unique relations between the Hamiltonian $H(t)$ and the damping matrix $[d_{kl}(t)]$ on the one hand, and the original generator on the other hand. Consequently, for the latter there exists a unique decomposition (15) into a conservative part and a dissipative part. In short, time-local master equations can be standardized.

Second, the old Nakajima–Zwanzig equation describing reduced dynamics of an open quantum system $\mathscr{S}$, possesses a nonstationary Markovian counterpart. Therefore, the status of time-local master equations is not phenomenological. A well-paved road from unitary dynamics to Eqs. (14)–(17) exists. Evaluation of the generator produces an infinite series that depends on the energy eigenvalues of $\mathscr{S}$, as well as on the correlation functions of $\mathscr{R}$. The relation with the constant generator resulting from the weak-coupling limit, is lucid.

Third, Hermiticity of $H(t)$ and positivity of $[d_{kl}(t)]$ imply that the evolution equations (14)–(17) meet all basic constraints on the quantum

dynamical map. This statement may be welcomed by any experimentalist seeking a simple, and at the same time conceptually correct, modelization of observations on dissipative quantum systems. Often, Markovian master equations with a constant generator fail to catch the observed phenomena.[26, 27] We stress that the afore-mentioned conditions are not necessary. For the Bloch equations with time-dependent coefficients, the condition $[d_{kl}(t)] \geqslant 0$ can be much weakened. Inequalities can be proposed that have a global rather than a local character.

The use of time-local master equations has two obvious disadvantages. First, due to time-ordering problems, analytic solution of these equations will be a cumbersome undertaking in general. As illustrated in Section 4, for convolutional master equations the situation is much better. The tool of Laplace transformation can be utilized in an effective manner.

Second, if one decides to analyze a given quantum dynamical map $\Lambda(t, t_0)$ on the basis of a time-local master equation, then one encounters a serious obstacle. The construction of generator $L(t, t_0)$ necessitates the inversion of $\Lambda(t, t_0)$; usually, this is not possible on the complete time axis $t \geqslant t_0$, but merely on a closed time interval $t_0 \leqslant t \leqslant t_0 + a$.

As becomes clear in Section 4, we need not be afraid that the map $\Lambda(t, t_0)$ does not start any detectable phenomena during the interval $[t_0, t_0 + a]$. On the other hand, unpleasant cases are likely to occur. For instance, a minor perturbation of a constant generator may cause bound a to become finite, such that a considerable part of the evolution takes place on the interval $t \geqslant t_0 + a$.

In conclusion, we advocate the use of time-local master equations in studying dissipative quantum systems, provided that one keeps a sharp eye on the existence of generators.

APPENDIX A. PROOF OF A UNIQUENESS THEOREM

As noted in the main text, one should prove that the statement (44) leads to the result $c_{kl} = 0$ for all (kl), at least, if the density matrix $\rho_{\mathscr{S}}$ may be arbitrarily chosen.

It is possible to set $\rho_{\mathscr{S}}$ equal to $\mathbf{1}_N/N$, $\mathbf{1}_N/N + \varepsilon(F_k + F_k^\dagger)$, and $\mathbf{1}_N/N + i\varepsilon(F_k - F_k^\dagger)$, $1 \leqslant k \leqslant N^2 - 1$, provided that ε is taken real and small. Hence, in (44) density matrix $\rho_{\mathscr{S}}$ can be replaced by any element of $\mathscr{M}(N)$, for instance, matrix $|p\rangle\langle q|$. We thus arrive at the following consequence of (44):

$$\sum_{k,\,l=0}^{N^2-1} c_{kl} \langle m| F_k |p\rangle \langle q| F_l^\dagger |n\rangle = 0 \tag{A1}$$

The choice of integers m, n, p, q is free.

For the basis set of matrices we take[5]

$$\begin{aligned}
\langle m|\, F_0\, |n\rangle &= N^{-1/2}\delta_{mn} \\
\langle m|\, F_1^{(r)}\, |n\rangle &= [r(r+1)]^{-1/2}\left(\sum_{h=1}^{r}\delta_{hm} - r\delta_{r+1,m}\right)\delta_{mn} \\
\langle m|\, F_2^{(s,t)}\, |n\rangle &= (\delta_{sm}\delta_{tn} + \delta_{tm}\delta_{sn})/\sqrt{2} \\
\langle m|\, F_3^{(s,t)}\, |n\rangle &= -i(\delta_{sm}\delta_{tn} - \delta_{tm}\delta_{sn})/\sqrt{2}
\end{aligned} \tag{A2}$$

with $1 \leqslant r \leqslant N-1$ and $1 \leqslant s < t \leqslant N$. These equations define the four subsets of matrices, to which we shall refer below. One verifies that the above basis set complies with all properties, proposed at the beginning of Section 2. If the proof of uniqueness is ready for set (A2), then we are done with each set relating to (A2) via a unitary transformation.

Each summation in (A1) falls apart into a summation over the four subsets, and a summation over the elements of a set. We thus have to denote the constants in (A1) as $c_{(j_1)(j_2)}^{(\beta_1)(\beta_2)}$, where the indices j_1, j_2 label sets, and the indices β_1, β_2 label elements of sets. For $j_i = 0$ the index β_i is not needed, whereas for $j_i = 1, 2, 3$ one must take $(\beta_i) = (r)$, (s, t), (s, t), respectively $(i = 1, 2)$.

In the adapted version of (A1) we utilize (A2). Subsequently, we make the choices: $q < n$, $m < p$; $q > n$, $m < p$; $q < n$, $m > p$; $q > n$, $m > p$. Upon performing the interchanges $q \leftrightarrow n$ and/or $p \leftrightarrow m$, we find that the constants $c_{(j_1)(j_2)}^{(m,\,p)(q,\,n)}$ vanish for $j_1, j_2 = 2, 2; 2, 3; 3, 2; 3, 3$.

The choices $m = p$, $q < n$ and $m = p$, $q > n$ reduce the adapted version of (A1) to

$$N^{-1/2}c_{(0)(3)}^{(q,\,n)} + \sum_{r=1}^{N-1}[r(r+1)]^{-1/2}\left(\sum_{h=1}^{r}\delta_{hm} - r\delta_{r+1,m}\right)c_{(1)(3)}^{(r)(q,\,n)} = 0 \tag{A3}$$

with $1 \leqslant m \leqslant N$. By setting $m = N$, constant $c_{(0)(3)}^{(q,\,n)}$ can be eliminated. One is left with a homogeneous linear set of $(N-1)$ equations for the constants $\{c_{(1)(3)}^{(m)(q,\,n)}\}_{m=1}^{N-1}$, the determinant of which appears to be nonzero. Consequently, all constants featuring in (A3) vanish. The choices $m = p$, $q > n$; $m < p$, $q = n$; $m > p$, $q = n$ lead us to a zero result for all remaining constants with either j_1 or j_2 greater than unity.

Last, for the four cases $m = n = N$; $m = N$, $1 \leqslant n \leqslant N-1$; $1 \leqslant m \leqslant N-1$, $n = N$; $1 \leqslant m, n \leqslant N-1$, with $m = p$ and $n = q$ valid throughout, the adapted version of (A1) gives rise to four identities for the constants $c_{(0)(0)}$,

$c^{(r)}_{(0)(1)}$, $c^{(r)}_{(1)(0)}$, and $c^{(r)(r')}_{(1)(1)}$. Elimination of the first three constants produces the system

$$\sum_{r,\,r'=1}^{N-1} \xi_{mr}\xi_{nr'} c^{(r)(r')}_{(1)(1)} = 0 \tag{A4}$$

with $1 \leqslant m,\ n \leqslant N-1$ and the abbreviation

$$\xi_{mr} = [r(r+1)]^{-1/2} [\theta_{mr} - r\delta_{r+1,\,m} + r\delta_{r+1,\,N}] \tag{A5}$$

We have defined a discrete step function $\theta_{mn} =: 1$ for $m \leqslant n$; 0 for $m > n$.

The matrix appearing in (A4) is a direct product of two matrices $[\xi_{mr}]$, so its determinant equals $\{\det[\xi_{mr}]\}^{2N-2}$. In processing (A3), the determinant between curly brackets has been found to differ from zero. Therefore, the constants $c^{(r)(r')}_{(1)(1)}$ must equal zero. Returning to the original four identities, we see that there are no nonzero constants left anymore.

APPENDIX B. SOLUTION OF THE JAYNES–CUMMINGS MODEL WITH ATOMIC DAMPING

Use is made of a method that was developed in ref. 38. We decompose the density operator satisfying master equation (51) as

$$\rho^{(I)}(t) = i_+ \otimes \rho_1(t) + \sigma_- \otimes \rho_2(t) + \sigma_+ \otimes \rho_3(t) + i_- \otimes \rho_4(t) \tag{B1}$$

The atomic matrices were specified in Section 4. The field operators $\{\rho_j(t)\}_{j=1}^4$ act on the Hilbert space $\mathscr{H}_{\mathscr{F}}$. These operators can be represented with the help of photon-number states. The corresponding matrix elements can be stored up in the following fashion:

$$\mathbf{v}(t; m, n) = [\rho_1(t)_{m,n}, \rho_2(t)_{m+1,n}, \rho_3(t)_{m,n+1}, \rho_4(t)_{m+1,n+1}]^T \tag{B2}$$

The photon numbers are taken as $m, n \geqslant -1$, with the convention that the matrix element $\rho_j(t)_{m,n} = \langle m|\ \rho_j(t)\ |n\rangle$ equals zero in case either m or n is negative.

In Laplace language, the master equation (51) is equivalent to an algebraic recursion for the vector (B2), which can be solved iteratively. Backtransformation brings us to

$$\mathbf{v}(t; m, n) = \sum_{k=0}^{\infty} \oint \frac{dz}{2\pi i} e^{-izt} \prod_{l=0}^{k-1} \{[z\mathbf{1}_4 - iA(m+l, n+l)]^{-1} S(m+l, n+l)\}$$
$$\times [z\mathbf{1}_4 - iA(m+k, n+k)]^{-1} \mathbf{v}(t=0; m+k, n+k) \tag{B3}$$

for $m, n \geqslant -1$. The contour encircles all poles counterclockwise. In elaborating the matrix product, one must preserve the order of matrices inside the curly brackets, and furthermore, put matrices with lower index l to the left of those with higher index l.

The matrices in (B3) are given by

$$A(m,n) = \begin{pmatrix} -2\gamma & -i\lambda(m+1)^{1/2} & i\lambda(n+1)^{1/2} & 0 \\ -i\lambda(m+1)^{1/2} & -\gamma & 0 & i\lambda(n+1)^{1/2} \\ i\lambda(n+1)^{1/2} & 0 & -\gamma & -i\lambda(m+1)^{1/2} \\ 0 & i\lambda(n+1)^{1/2} & -i\lambda(m+1)^{1/2} & 0 \end{pmatrix} \tag{B4}$$

$$\begin{aligned} A(-1,n)_{kl} &= A(0,n)_{kl}\,(\delta_{k2}+\delta_{k4})(\delta_{l2}+\delta_{l4}) \\ A(n,-1)_{kl} &= A(n,0)_{kl}\,(\delta_{k3}+\delta_{k4})(\delta_{l3}+\delta_{l4}) \end{aligned} \tag{B5}$$

with $m, n \geqslant 0$. Furthermore, one has $A(-1,-1) = 0$ and

$$S(m,n)_{kl} = 2i\gamma\delta_{k4}\delta_{l1} \tag{B6}$$

for $m, n \geqslant -1$.

Evaluation of the atomic density matrix $\rho^{(I)}_{\mathscr{S}}(t)$ requires that the vector elements $\mathbf{v}(t;n,n)_1$, $\mathbf{v}(t;n,n)_4$, and $\mathbf{v}(t;n+1,n)_3$ be summed over all integers $n \geqslant -1$. Use of (B3) leads to a double summation over k and n, which reduces to a single finite summation after insertion of the initial condition (52). Therefore, we do not run into problems of convergence. From (B3) it is manifest that the functions $\{\hat{\beta}_j(z)\}_{j=1}^{3}$, defined by (53), (54), and (59), are meromorphic. Since all summations and products are finite, the number of poles is finite as well.

Computation of the inverse matrices of (B3), followed by factorization of the associated determinants, furnishes explicit expressions for the functions $\{\beta_j(t)\}_{j=1}^{3}$; choosing $j=1, 2$, we end up with

$$\begin{aligned} \beta_1(t) &= -2\lambda^2 \sum_{n=0}^{p-1} \frac{p!}{(p-n-1)!}\,(-4i\gamma\lambda^2)^n \\ &\quad \times \oint \frac{dz}{2\pi i}\,\frac{e^{-izt}}{(z+i\gamma)^{n+1}}\,\frac{1}{\prod_{\eta=\pm1}\prod_{s=0}^{n}(z+i\gamma+2\eta u_{p-s-1})} \\ \beta_2(t) &= \sum_{n=0}^{p} \frac{p!}{(p-n)!}\,(-4i\gamma\lambda^2)^n \\ &\quad \times \oint \frac{dz}{2\pi i}\,\frac{e^{-izt}}{(z+i\gamma)^{n+1}}\,\frac{[z(z+i\gamma)-2\lambda^2(p+1)]}{\prod_{\eta=\pm1}\prod_{s=0}^{n}(z+i\gamma+2\eta u_{p-s})} \end{aligned} \tag{B7}$$

The definition $u_n = [\lambda^2(n+1) - \gamma^2/4]^{1/2}$ has been employed. As the function $\beta_3(t)$ is not urgently needed in Section 4, its explicit representation is not given.

For the undamped case $\gamma = 0$ one obtains $\beta_1(t) = \sin^2[p^{1/2}\lambda t]$, $\beta_2(t) = \cos^2[(1+p)^{1/2}\lambda t]$, and $\beta_3(t) = \cos[(1+p)^{1/2}\lambda t]\cos[p^{1/2}\lambda t]$. These solutions of the standard Jaynes–Cummings model underlie the findings (61), and the result for bound a given below (61).

In deriving Eqs. (62), we have retained in (B7) only terms with $n = 0$. To arrive at Eq. (64), the diagonals of the atomic density matrix must be calculated independently of each other, that is to say, on the basis of the solutions for the vector elements $\mathbf{v}(t; n, n)_1$ and $\mathbf{v}(t; n, n)_4$. The summation that equals the diagonal $\langle 1| \rho_{\mathscr{S}}^{(I)}(t) |1\rangle$, must be treated carefully. In contrast to the summations (B7), some terms with $n = 1$ are of zeroth order in ε. They make a nonzero contribution to (64).

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