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***Ab initio* construction of an analytically tractable Kraus map for non-Markovian quantum dissipation**

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Abstract

Starting from a prescribed Hamiltonian, we construct a non-Markovian evolution equation for a non-relativistic quantum system that exchanges energy with a large reservoir. In order to create sufficient mathematical freedom, the density operator is replaced by a more flexible entity that depends on two times. If these times are chosen equal, the density operator is recovered. In deriving a non-Markovian integral equation for our bitemporal operator, it is assumed that initially system and reservoir are completely uncorrelated. Furthermore, in employing Wick's theorem for factorization of reservoir correlation functions, only those Wick contractions between reservoir potentials are retained that belong to a generalized nearest-neighbour class. The latter is established by subjecting the set of plain nearest-neighbour contractions to any cyclic permutation of reservoir potentials. Through generalizing the notion of nearest-neighbour contraction, it is ensured that the trace of the density operator is conserved. By construction, our bitemporal evolution equation agrees with the Kraus map for quantum dissipation. Moreover, a sound Markovian limit exists that reproduces the complete van Hove–Davies theory. By making use of a rotating-wave approximation and Laplace transformation, the density operator of a damped N -level atom can be computed. For large times and moderate coupling to the reservoir, the atom ends up near the state of thermal equilibrium. At zero temperature, our non-Markovian integral equation gives an exact solution for the atomic density operator.

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1. Introduction

Dissipation of energy is omnipresent, not only in our everyday life, but also in the world of quantum physics. This rather loose observation acquired a somewhat urgent status at the beginning of the 1960s. Due to the arrival of the laser, a sudden need arose for accurate and comprehensive descriptions of lossy quantum systems. As a result, the theory of open quantum systems blossomed out into a major research area of statistical mechanics and quantum optics. During the 1970s, our knowledge of Markovian quantum processes almost reached a state of perfection. To date, the field is very much alive. Recent years saw publication of some excellent textbooks on the theory of open quantum systems and related topics [1–5].

A great number of workers tried to transfer the Markovian successes of the 1970s to the case of non-Markovian quantum dynamics. Unfortunately, this proved to be a difficult enterprise. Therefore, one has to admit that, in spite of a large demand [6–11], our understanding of dissipative quantum processes is still not mature. One of the main open questions is how to derive a non-Markovian evolution equation of reasonable complexity from a prescribed Hamiltonian. Of course, the corresponding density operator should respect all basic constraints. These are conservation of trace, agreement with the Kraus map for quantum dissipation [12], and existence of a sound Markovian limit reproducing the complete van Hove–Davies theory [13, 14]. The goal of the present treatment is to come up with an analytically solvable evolution equation obeying all of the afore-mentioned requirements.

Over the years, an impressive amount of expertise was collected on virtues and vices of non-Markovian master equations for density operators. This judgement did not encourage us to undertake any further research on master equations containing one single time variable. Instead, we gradually came to the conviction that a density operator does not constitute the natural quantity for capturing non-Markovian processes. In order to increase mathematical freedom, we decided to work with a novel and more flexible entity. It depends on two times t and t' , and delivers the density operator if the choice $t = t'$ is made. This so-called bitemporal operator permits one to set up a perturbation theory that is free of any commutators. In doing so, one makes sure from the very outset that the non-Markovian evolution of the ensuing density operator is governed by a Kraus map. In a recent letter, the above ideas were applied to the case of a damped two-level atom [15].

Our paper is organized as follows. In section 2 an evolution equation for the bitemporal operator is derived. It is assumed that initially the damped system is not entangled with the surroundings. Furthermore, in factorizing correlation functions with the help of Wick's theorem, only Wick contractions of a generalized nearest-neighbour type are taken into account. The bitemporal operator gives rise to a physically acceptable density operator, because all necessary conditions are fulfilled. This is shown in section 3. Explicit solutions for the density operator of a two-level and three-level atom are derived on the basis of Laplace transformation. In section 4 the temperature is kept at absolute zero, but in section 5 the atom is immersed in a thermal bath. The rotating-wave approximation underlies all of our solutions. As for exactly solvable models [16], in performing Laplace backtransformation one has to deal with branch cuts. Section 6 contains a discussion of all approximations that are needed in order to arrive at our main result, the non-Markovian evolution equation (12).

2. Bitemporal approach

Starting from the level of unitary dynamics, we wish to evaluate the density operator $\rho_S(t)$ describing the evolution in time t of a non-relativistic quantum system S that can exchange energy with a large reservoir R . Throughout this treatment, we assume that at time $t = 0$

system and reservoir are completely uncorrelated, S being in the state ρ_S and R being in the state ρ_R . As we shall exclusively work in the interaction picture, we write the Hamiltonian H of the composite SR as a sum $H_0 + \lambda H_1$ of a free part H_0 and an interaction part H_1 . The dimensionless coupling parameter λ is real. The free Hamiltonian can be expressed as a sum $H_S \otimes 1_R + 1_S \otimes H_R$ of a part H_S pertaining to the system and a part H_R pertaining to the reservoir. In the absence of classical fields all of our Hamiltonians do not depend on time. For notational convenience, we scale each Hamiltonian with Planck's constant.

Given the foregoing assumptions and conventions, the evolution law for the density operator can be cast into the form

$$\rho_S(t) = \xi(t, t). \quad (1)$$

On the right-hand side the definition

$$\xi(t, t') = \text{Tr}_R[e^{iH_0 t} e^{-iH t} \rho_S \otimes \rho_R e^{iH t'} e^{-iH_0 t'}] \quad (2)$$

is employed. We aim at constructing a linear integral equation for the bitemporal operator $\xi(t, t')$.

If the Hilbert space of S is separable, the interaction Hamiltonian can be factorized. The representation

$$H_1 = \sum_{\alpha} V_{\alpha} \otimes U_{\alpha} = \sum_{\alpha} V_{\alpha}^{\dagger} \otimes U_{\alpha}^{\dagger} \quad (3)$$

is found. In the interaction picture the system potential V_{α} and the reservoir potential U_{α} evolve as

$$V_{\alpha}(t) = e^{iH_S t} V_{\alpha} e^{-iH_S t} \quad U_{\alpha}(t) = e^{iH_R t} U_{\alpha} e^{-iH_R t}. \quad (4)$$

The perturbative expansion for the unitary operators governing the evolution of $\xi(t, t')$ reads then [17]

$$\begin{aligned} e^{iH_0 t} e^{-iH t} &= 1 + \sum_{n=1}^{\infty} \sum_{\alpha_1 \alpha_2 \dots \alpha_n} (-i\lambda)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{n-1}} dt_n \\ &\times V_{\alpha_1}(t_1) V_{\alpha_2}(t_2) \dots V_{\alpha_n}(t_n) \otimes U_{\alpha_1}(t_1) U_{\alpha_2}(t_2) \dots U_{\alpha_n}(t_n). \end{aligned} \quad (5)$$

Upon substituting (5) into (2), we meet the m -point reservoir correlation functions

$$\text{Tr}_R [U_{\alpha_1}(t_1) U_{\alpha_2}(t_2) \dots U_{\alpha_m}(t_m) \rho_R] \equiv \{12 \dots m\}. \quad (6)$$

They determine if, and in what manner, the system relaxes towards a state of equilibrium.

We assume that (6) vanishes for m odd. Moreover, we assume that for m even (6) can be factorized into products of pair-correlation functions. The latter are defined as

$$c_{\alpha_1 \alpha_2}(t_1, t_2) = \text{Tr}_R [U_{\alpha_1}(t_1) U_{\alpha_2}(t_2) \rho_R] \equiv \{12\}. \quad (7)$$

In this work, we shall rely on the following factorization scheme:

$$\begin{aligned} \{1234\} &= \{12\}\{34\} + \{14\}\{23\} \\ \{123456\} &= \{12\}\{34\}\{56\} + \{12\}\{36\}\{45\} + \{14\}\{23\}\{56\} + \{16\}\{23\}\{45\} + \{16\}\{25\}\{34\} \\ &\dots \end{aligned} \quad (8)$$

It is straightforward to construct a graphical representation of (8). Consider the points with $x = 1, 2, 3, \dots, m$ and $y = 0$ in the xy plane; form pairs by drawing lines in the lower halfplane; only retain those graphs for which lines never intersect. Now if we link up 1 and 3 as well as 2 and 4, then one intersection of lines is needed. Indeed, the factorization $\{13\}\{24\}$ does not show up in (8). We emphasize that (8) includes all factorizations that make a nonzero

contribution in the limit of weak damping. Hence, we shall be able to reproduce the complete van Hove–Davies theory of Markovian quantum dissipation.

For a thermal reservoir consisting of harmonic oscillators, factorization of correlation functions can be carried out with the help of Wick’s theorem [17]. Obviously, a Wick contraction between reservoir potentials corresponds to a pair $\{kl\}$ of the graphs just introduced. In (8) we only retain those Wick contractions that are of a generalized nearest-neighbour type. To collect these, one must subject the set of plain nearest-neighbour contractions to any cyclic permutation of reservoir potentials, in each order of λ . We can graphically perform this exercise. Under the permutation $1234 \rightarrow 2341$ the graph $\{12\}\{34\}$ is mapped to $\{14\}\{23\}$. The other cyclic permutations do not produce the graph $\{13\}\{24\}$, which is indeed lacking in (8). To find all generalized nearest-neighbour contractions of order λ^6 , we first extend all graphs of order λ^4 with the plain nearest-neighbour contraction $\{56\}$. The resulting graphs $\{12\}\{34\}\{56\}$ and $\{14\}\{23\}\{56\}$ are then subjected to cyclic permutations of reservoir potentials. The additional graphs $\{16\}\{23\}\{45\}$, $\{12\}\{36\}\{45\}$ and $\{16\}\{25\}\{34\}$ appear. We thus reproduce all Wick contractions figuring in (8).

We implement the prescription (8) in the perturbative expansion for the bitemporal operator $\xi(t, t')$ that follows from (2) and (5). Let us first make the choice $t' = 0$, so that all system potentials stand to the left of ρ_S . It is quite clear that after employment of (8), $\xi(t, 0)$ depends on the initial reservoir state ρ_R in a nonlinear fashion. Consequently, if we wish to sum up all contributions to $\xi(t, 0)$, we should really search for a nonlinear equation. This happens by scrutinizing how (8) modifies the first few terms of the perturbative expansion for $\xi(t, 0)$. We are led to the statement that $\xi(t, 0)$ becomes equal to $Q(t, 0)\rho_S$. The new operator satisfies the nonlinear integral equation

$$Q(t, s) = 1_S - \lambda^2 \sum_{\alpha\beta} \int_s^t du \int_s^u dv V_\alpha(u) Q(u, v) V_\beta(v) Q(v, s) c_{\alpha\beta}(u, v). \tag{9}$$

The choice $t \geq s \geq 0$ should be made.

Iteration of (9) gives

$$\begin{aligned} Q(t, s) = & 1_S - \lambda^2 \sum_{\alpha_1\alpha_2} \int_s^t dt_1 \int_s^{t_1} dt_2 V_{\alpha_1}(t_1) V_{\alpha_2}(t_2) c_{\alpha_1\alpha_2}(t_1, t_2) \\ & + \lambda^4 \sum_{\alpha_1\alpha_2\alpha_3\alpha_4} \int_s^t dt_1 \int_s^{t_1} dt_2 \int_s^{t_2} dt_3 \int_s^{t_3} dt_4 V_{\alpha_1}(t_1) V_{\alpha_2}(t_2) V_{\alpha_3}(t_3) V_{\alpha_4}(t_4) \\ & \times [c_{\alpha_1\alpha_2}(t_1, t_2) c_{\alpha_3\alpha_4}(t_3, t_4) + c_{\alpha_1\alpha_4}(t_1, t_4) c_{\alpha_2\alpha_3}(t_2, t_3)] \\ & - \lambda^6 \sum_{\alpha_1\alpha_2\alpha_3\alpha_4\alpha_5\alpha_6} \int_s^t dt_1 \int_s^{t_1} dt_2 \int_s^{t_2} dt_3 \int_s^{t_3} dt_4 \int_s^{t_4} dt_5 \int_s^{t_5} dt_6 V_{\alpha_1}(t_1) V_{\alpha_2}(t_2) \\ & \times V_{\alpha_3}(t_3) V_{\alpha_4}(t_4) V_{\alpha_5}(t_5) V_{\alpha_6}(t_6) [c_{\alpha_1\alpha_2}(t_1, t_2) c_{\alpha_3\alpha_4}(t_3, t_4) c_{\alpha_5\alpha_6}(t_5, t_6) \\ & + c_{\alpha_1\alpha_2}(t_1, t_2) c_{\alpha_3\alpha_6}(t_3, t_6) c_{\alpha_4\alpha_5}(t_4, t_5) + c_{\alpha_1\alpha_4}(t_1, t_4) c_{\alpha_2\alpha_3}(t_2, t_3) c_{\alpha_5\alpha_6}(t_5, t_6) \\ & + c_{\alpha_1\alpha_6}(t_1, t_6) c_{\alpha_2\alpha_3}(t_2, t_3) c_{\alpha_4\alpha_5}(t_4, t_5) + c_{\alpha_1\alpha_6}(t_1, t_6) c_{\alpha_2\alpha_5}(t_2, t_5) c_{\alpha_3\alpha_4}(t_3, t_4)] \\ & + \dots, \tag{10} \end{aligned}$$

where contributions of order λ^8 and higher have been omitted. One checks that the iterative series for $Q(t, 0)\rho_S$ is indeed identical to the perturbative expansion for $\xi(t, 0)$ as obtained from (2), (5) and (8). The orders of λ^8 and higher can be covered by setting up an induction proof.

We are now in a position to perform a summation of all terms contributing to the perturbative expansion for the full bitemporal operator $\xi(t, t')$. Upon inserting (5) into (2), we

see that potentials $V_\alpha \otimes U_\alpha$ appear on both sides of $\rho_S \otimes \rho_R$. Due to the cyclic property of the trace over R , the order of the reservoir potentials is not fixed. However, as we implement the prescription (8), we refrain from modifying the order of the potentials. Moreover, we count the number p of pairs $U_{\alpha_1}(t_1), U_{\alpha_2}(t_2)$ that make up a pair-correlation function and enclose ρ_R at the same time. That is to say, before factorization of correlation functions is carried out, the potential $U_{\alpha_1}(t_1)$ should stand to the left of ρ_R , and the potential $U_{\alpha_2}(t_2)$ to the right of ρ_R (or the other way round).

The sum of all contributions to $\xi(t, t')$ for which p equals 0, amounts to $Q(t, 0)\rho_S Q^\dagger(t', 0)$. This follows directly from our discussion of the case $t' = 0$. The sum of all contributions to $\xi(t, t')$ for which p equals 1, amounts to

$$\lambda^2 \sum_{\alpha\alpha'} \int_0^t ds \int_0^{t'} ds' Q(t, s) V_\alpha(s) Q(s, 0) \rho_S Q^\dagger(s', 0) V_{\alpha'}(s') Q^\dagger(t', s') c_{\alpha'\alpha}(s', s). \quad (11)$$

We substitute the iterative solution of (9) into (11) and interchange integrals in a suitable manner. Then all contributions to $\xi(t, t')$ with $p = 1$ indeed emerge.

The integrand of (11) contains the operator $Q(s, 0)\rho_S Q^\dagger(s', 0)$, which is the solution for $\xi(s, s')$ if p equals 0. The foregoing recognition suggests that for a proper handling of the cases $p = 0, 1, 2, 3, \dots$, one should perform an iteration of

$$\begin{aligned} \xi(t, t') = & Q(t, 0)\rho_S Q^\dagger(t', 0) \\ & + \lambda^2 \sum_{\alpha\alpha'} \int_0^t ds \int_0^{t'} ds' Q(t, s) V_\alpha(s) \xi(s, s') V_{\alpha'}(s') Q^\dagger(t', s') c_{\alpha'\alpha}(s', s). \end{aligned} \quad (12)$$

This is the desired integral equation for the bitemporal operator. It is important to point out that (9) and (12) constitute a closed set of equations that is embedded in the Hilbert space of the system. The reservoir manifests itself exclusively through the pair-correlation function (7). We shall not investigate convergence of the iterative solution generated by (9) and (12). On the other hand, via a simple but rather lengthy induction proof one formally demonstrates that this iterative solution coincides with the perturbative expansion for $\xi(t, t')$ that is obtained from (2), (5) and (8).

One might wonder whether it is possible to avoid the use of a bitemporal operator. Then (12) should be replaced by an equation for the density operator itself. Upon choosing $t = t'$, the left-hand side of (12) indeed reduces to $\rho_S(t)$. However, on the right-hand side the integrand still depends on the variables s and s' . We therefore conclude that summation of our expansion for the density operator requires employment of a bitemporal quantity.

Adopting a perturbative approach and systematically retaining generalized nearest-neighbour Wick contractions, we have devised dissipative dynamics of non-Markovian nature. Next, we have to find out whether this dynamics complies with the conditions that safeguard physical relevance.

3. Physical content

Postponing a continuum limit for the reservoir, we may expand the initial reservoir state as

$$\rho_R = \sum_j \mu_j |r_j\rangle \langle r_j|. \quad (13)$$

The c -numbers $\{\mu_j\}$ are positive and add up to unity. The kets $\{|r_j\rangle\}$ make up an orthonormal basis for the reservoir Hilbert space. Use of (13) in (2) shows that the evolution of the density operator is dictated by the Kraus map [12]

$$\rho_S(t) = \sum_m W_m(t) \rho_S W_m^\dagger(t). \quad (14)$$

The operators $\{W_m(t)\}$ act within the Hilbert space of the system. They must obey the requirement $\sum_m W_m^\dagger(t)W_m(t) = 1_S$, otherwise the trace of the density operator is not conserved. The precise definition of $W_m(t)$ can be found in the literature [18]. From (14) one learns that $\rho_S(t)$ is self-adjoint and positive if the same is true for ρ_S .

The evolution equations (9) and (12) furnish a density operator that evolves in accordance with the Kraus map. This can be demonstrated by factorizing the pair-correlation function (7) with the help of the completeness relation for the basis $\{|r_j\rangle\}$. The resulting expansion for $c_{\alpha'\alpha}(s', s)$, as well as the self-adjointness (3) of the interaction Hamiltonian, should be utilized in (12). Upon iterating (12) and choosing t equal to t' , one gets a density operator that indeed attains the form (14).

The next important issue is conservation of trace. We already explained that the collection of graphs drawn under (8) is closed under any cyclic permutation of reservoir potentials. Accordingly, one may hope that the trace of $\rho_S(t)$ is conserved. Our expectations are confirmed by evaluating the derivative of $\xi(t, t)$ with respect to time. Differentiation of (12) leads to

$$\begin{aligned} \frac{d\xi(t, t)}{dt} &= \frac{dQ(t, 0)}{dt} \rho_S Q^\dagger(t, 0) + \lambda^2 \sum_{\alpha\alpha'} \int_0^t ds' V_\alpha(t) \xi(t, s') V_{\alpha'}(s') Q^\dagger(t, s') c_{\alpha'\alpha}(s', t) \\ &\quad + \lambda^2 \sum_{\alpha\alpha'} \int_0^t ds \int_0^t ds' \frac{\partial Q(t, s)}{\partial t} V_\alpha(s) \xi(s, s') V_{\alpha'}(s') Q^\dagger(t, s') c_{\alpha'\alpha}(s', s) + \text{h.c.} \end{aligned} \quad (15)$$

On the right-hand side three Hermitian conjugate terms must be added. The derivatives of $Q(t, s)$ are eliminated with the help of (9). Furthermore, the operator $\xi(t, s')$ is eliminated by merely making use of (12) again. In total, four contributions of order λ^2 are obtained, in which the initial state ρ_S figures. Furthermore, four contributions of order λ^4 are obtained, in which the bitemporal operator ξ figures. Next, we take the trace over S and exploit its cyclic property. Upon interchanging some integrals and making use of the self-adjointness relation (3), we are left with a vanishing result. The conclusion must be that the equality

$$\text{Tr}_S[\rho_S(t)] = 1 \quad (16)$$

holds true for all t , at least, as long as the trace of the initial state ρ_S equals unity.

In the weak-coupling or van Hove [13] limit, given by $t \rightarrow \infty$, $\lambda \rightarrow 0$ and $\lambda^2 t$ constant, only plain nearest-neighbour Wick contractions survive if it comes to application of Wick's theorem. Hence, in the van Hove limit solution of (9) and (12) surely reproduces the celebrated weak-coupling density operator that is highlighted in any textbook on quantum dissipation [17, 18]. Conditions under which the van Hove limit exists and converges were formulated by Davies [14]. In the following, we do not address these matters. We are merely going to calculate the density operator that is generated by (9) and (12) if the van Hove limit is taken.

As a first step, we derive from (9) the auxiliary result

$$\lim_{\lambda \rightarrow 0} Q(t/\lambda^2, s/\lambda^2) = e^{K(t-s)}, \quad (17)$$

where K denotes a time-averaged operator

$$K = - \lim_{T \rightarrow \infty} \sum_{\alpha\beta} T^{-1} \int_0^T dt \int_0^\infty ds V_\alpha(t+s) V_\beta(t) c_{\alpha\beta}(t+s, t). \quad (18)$$

It is assumed that the limit on the left-hand side of (17) exists. As a consequence, for small λ the operator $Q(v/\lambda^2 + u, v/\lambda^2)$ converges to $Q(v/\lambda^2, v/\lambda^2)$, which is just equal to unity.

Thus (9) becomes a linear equation in the van Hove limit. The fact that a time-averaged operator comes into existence is due to the identity

$$\lim_{\lambda \rightarrow 0} \int_0^t ds f(s/\lambda^2)g(s) = \lim_{T \rightarrow \infty} T^{-1} \int_0^T du f(u) \int_0^t ds g(s) \quad (19)$$

which is valid for well-behaved functions f and g . Verification of (19) goes by means of partial integration.

As a second step, we derive from (12) and (17) the integral equation

$$\lim_{\lambda \rightarrow 0} \rho_S(t/\lambda^2) = e^{L_1 t} \rho_S + \int_0^t ds e^{L_1(t-s)} L_2 \lim_{\lambda \rightarrow 0} \rho_S(s/\lambda^2) \quad (20)$$

where the superoperators

$$\begin{aligned} L_1 \rho_S &= K \rho_S + \rho_S K^\dagger \\ L_2 \rho_S &= \lim_{T \rightarrow \infty} \sum_{\alpha\beta} T^{-1} \int_0^T dt \int_0^\infty ds [V_\alpha(t) \rho_S V_\beta(t+s) c_{\beta\alpha}(t+s, t) \\ &\quad + V_\alpha(t+s) \rho_S V_\beta(t) c_{\beta\alpha}(t, t+s)] \end{aligned} \quad (21)$$

come into play. Before invoking (19), one has to separate the double integral of (12) into two parts $\int_0^t ds \int_s^t ds'$ and $\int_0^t ds' \int_s^t ds$.

Recognizing that (20) is nothing more than a Kato identity [19], we arrive at

$$\lim_{\lambda \rightarrow 0} \rho_S(t/\lambda^2) = e^{L t} \rho_S \quad L = L_1 + L_2. \quad (22)$$

This is the well-known Markovian result for the density operator of a weakly damped quantum system. Note that in [15] we did not succeed in reproducing the van Hove–Davies theory of Markovian damping.

In conclusion, the integral equations (9) and (12) provide us with a non-Markovian density operator that displays a physically acceptable behaviour. Apart from being self-adjoint and positive, the density operator also obeys the constraint of trace conservation. Furthermore, it possesses a sound Markovian limit. On top of that, it can be analytically computed by means of Laplace transformation. This job will be taken care of in the next two sections.

4. N -level atom at zero temperature

For many applications the spectrum of the system Hamiltonian may be taken as discrete and nondegenerate. We put the eigenvalues of H_S into the order $\omega_1 < \omega_2 < \omega_3 < \dots < \omega_N$, where N may become infinitely large. Denoting the eigenstates of H_S as $\{|k\rangle\}$, we can make the following choice for the potentials:

$$\alpha \rightarrow (kl) \quad V_\alpha \rightarrow |k\rangle\langle l| \quad U_\alpha \rightarrow U_{(kl)} = U_{(lk)}^\dagger. \quad (23)$$

The orthonormality of the set $\{|k\rangle\}$ will be of frequent help.

At zero temperature, the initial state ρ_R may be replaced by the ground state $|0\rangle_{RR}\langle 0|$ of the reservoir. Without loss of generality, the property $H_R|0\rangle_R = 0$ may be assumed. The fact that ρ_R and H_R commute enables us to write

$$c_{(kl)(mn)}(t, s) = c_{(kl)(mn)}(t - s, 0) \quad (24)$$

for arbitrary t and s . This identity is useful, but does not bring much mathematical relief. Hence, we are in need of further assumptions.

First, we shall make use of the property $U_{(kl)}|0\rangle_R = 0$, whenever the inequality $k \geq l$ is true. This means that we restrict our description to pure decay. In reality, excitations do

take place in S . The reason is that even at zero temperature the system experiences quantum fluctuations [17].

A second important simplification is brought about by the rotating-wave approximation, which can be stated as

$$c_{(kl)(mn)}(t, s) = 0 \quad \text{if} \quad \omega_k - \omega_l + \omega_m - \omega_n \neq 0. \quad (25)$$

To interpret the above restriction, we observe that the system potentials generate oscillations in (12), which have a period of order $1/\omega_1$. The latter time-scale is much smaller than a typical reservoir decorrelation time. The occurrence of fast oscillations indicates that H_1 can induce excitations for which energy is not conserved. As long as the coupling between system and reservoir is moderate, such transitions can be safely ignored. This happens by assuming (25).

On the basis of (23)–(25) one proves by induction in k that the solution of (9) has the form

$$\langle k|Q(t, s)|l\rangle = \delta_{kl}Q_k(t - s) = \delta_{kl}Q_k^*(s - t). \quad (26)$$

The function on the right-hand side is obtained by iterating the Laplace relation

$$\hat{Q}_k(\omega + i\epsilon) = \left[\omega + \lambda^2 \sum_{l=1}^{k-1} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi i} \hat{Q}_l(\omega' + i\epsilon) \hat{c}_{(kl)(lk)}(\omega - \omega' + \omega_k - \omega_l + i\epsilon) \right]^{-1} \quad (27)$$

and utilizing the Laplace identities

$$\hat{f}(z) = -i \int_0^{\infty} dt e^{izt} f(t) \quad f(t) = \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \hat{f}(\omega + i\epsilon). \quad (28)$$

As usual, ϵ is infinitesimally positive. The Laplace transform $\hat{f}(z)$ must be analytic in the upper halfplane $\text{Im } z > 0$.

It is essential to check that the last-mentioned condition is respected by all Laplace transforms figuring in (27). As a continuum limit has not yet been taken, the eigenvalues of the reservoir Hamiltonian make up a discrete set $\{\epsilon_j\}$. From (7) one then infers that $\hat{c}_{(kl)(lk)}(z)$, which is the Laplace transform of $c_{(kl)(lk)}(t, 0)$, is a meromorphic function. Its poles $\{z = \epsilon_j\}$ are located on the real axis.

Elaborating $\hat{c}_{(kl)(lk)}(z)$ with the help of the eigenstates of H_R , one verifies the inequalities

$$\text{Im } z \text{Im } \hat{c}_{(kl)(lk)}(z) < 0 \quad \frac{d\hat{c}_{(kl)(lk)}(x)}{dx} < 0, \quad (29)$$

where $\text{Im } z$ differs from zero and the real variable x differs from all eigenvalues $\{\epsilon_j\}$. Via induction in k , we are going to show that (i) $\hat{Q}_k(z)$ is a meromorphic function, the poles $\{z = \eta_j^{(k)}\}$ of which lie on the real axis, and (ii), the derivative $d\hat{Q}_k^{-1}(x)/dx$ is positive, with x real.

For $k = 1$, the foregoing results are trivial, because $Q_1(t)$ is equal to 1, as follows from (27). For $k \geq 2$, the induction assumption and the residue theorem enable us to derive from (27)

$$\hat{Q}_k^{-1}(z) = z - \lambda^2 \sum_{l=1}^{k-1} \sum_j \hat{c}_{(kl)(lk)}(z - \eta_j^{(l)} + \omega_k - \omega_l) \left[\frac{d\hat{Q}_l^{-1}(x)}{dx} \right]_{x=\eta_j^{(l)}}^{-1}. \quad (30)$$

The above representation and the fact that $\hat{c}_{(kl)(lk)}(z)$ is meromorphic imply that $\hat{Q}_k(z)$ is meromorphic as well. In view of (29) and (30), both $\text{Im } \hat{Q}_k^{-1}(z)/\text{Im } z$ and $d\hat{Q}_k^{-1}(x)/dx$ must be positive. This statement completes our proof, as we may now conclude that the poles of $\hat{Q}_k(z)$ lie on the real axis.

For the cases of a two-level and three-level atom iteration of (27) yields

$$\begin{aligned}
Q_2(t) &= \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} [\omega - \lambda^2 \hat{c}_{(21)(12)}(\omega + \omega_2 - \omega_1 + i\epsilon)]^{-1} \\
Q_3(t) &= \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \left[\omega - \lambda^2 \hat{c}_{(31)(13)}(\omega + \omega_3 - \omega_1 + i\epsilon) \right. \\
&\quad \left. + \lambda^2 \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi i} \frac{\hat{c}_{(32)(23)}(\omega - \omega' + \omega_3 - \omega_2 + i\epsilon)}{\omega' - \lambda^2 \hat{c}_{(21)(12)}(\omega' + \omega_2 - \omega_1 + i\epsilon)} \right]^{-1}
\end{aligned} \tag{31}$$

with $\epsilon = 0^+$ and $t \geq 0$. If the reservoir is subjected to a continuum limit, the poles of $\hat{c}_{(kl)(lk)}(z)$ unite so as to generate a branch cut on the real axis. This signals the onset of irreversible behaviour. Through a Riemann–Lebesgue argument one makes sure that $Q_k(t)$ converges to zero for t tending to ∞ and $k \geq 2$. Consequently, $\rho_S(t)$ ends up in the ground state $|1\rangle\langle 1|$. This is in keeping with our expectations, because we did not take into account any zero-point fluctuations.

By carrying out a double Laplace transformation one can solve (12) in the same manner as (9) was solved. At zero temperature, direct iteration of (12) offers a shorter path, because only a finite number of iterations is required. The density operator of a damped two-level atom comes out as

$$\langle 2|\rho_S(t)|2\rangle = |Q_2(t)|^2 \langle 2|\rho_S|2\rangle \quad \langle 2|\rho_S(t)|1\rangle = Q_2(t) \langle 2|\rho_S|1\rangle. \tag{32}$$

The remaining two matrix elements follow from self-adjointness and conservation of trace.

In our preliminary letter [15], the density operator (32) was elaborated for the case of spontaneous emission of a photon at zero temperature. The standard rotating-wave Hamiltonian was employed. It was found that (32) coincides with the density operator resulting from an exact diagonalization of the Hamiltonian [17]. Next, we tested whether (32) could also predict Rabi oscillations of a two-level atom at zero temperature. To that end, use was made of the Jaynes–Cummings Hamiltonian. Surprisingly, (32) once more reproduced the exact result for $\rho_S(t)$. The latter can be found in [20]. All in all, (32) is capable of accurately describing dynamics of both irreversible and reversible nature. These findings convey the message that the evolution equation (12) has a truly non-Markovian character.

For the case of a damped three-level atom at zero temperature, solution of (9) and (12) brings us to the following density operator:

$$\begin{aligned}
\langle 2|\rho_S(t)|2\rangle &= |Q_2(t)|^2 \langle 2|\rho_S|2\rangle + \lambda^2 \int_0^t ds \int_0^t ds' Q_2(t-s) Q_2^*(t-s') \\
&\quad \times Q_3(s) Q_3^*(s') c_{(32)(23)}(s', s) e^{i(\omega_3 - \omega_2)(s' - s)} \langle 3|\rho_S|3\rangle \\
\langle 3|\rho_S(t)|3\rangle &= |Q_3(t)|^2 \langle 3|\rho_S|3\rangle \\
\langle k|\rho_S(t)|l\rangle &= Q_k(t) Q_l^*(t) \langle k|\rho_S|l\rangle.
\end{aligned} \tag{33}$$

One must choose $k \neq l$. As in (32), only independent matrix elements are given. Note that for some configurations of the energy levels $\omega_1 < \omega_2 < \omega_3$ other correlation functions than $c_{(kl)(lk)}$ might contribute to the off-diagonals of the density operator. For instance, if ω_2 coincides with $(\omega_1 + \omega_3)/2$, then (25) allows the correlation function $c_{(32)(12)}$ to differ from zero.

It is instructive to systematically evaluate all diagonals of the density operator by employing Laplace transformation in (12). This opens up the possibility of verifying that the sum of all diagonals indeed amounts to unity. The proof relies on the method of partial fractions and Cauchy's theorem. One should start with the case $N = 2$ and utilize the corresponding expertise in treating the case $N = 3$.

Once the interaction Hamiltonian H_1 is specified, the full evaluation of the solutions (31)–(33) can be commenced. For the case of spontaneous emission interesting results were reported, indicating that in a strongly non-Markovian regime an excited atomic level might exhibit power-law decay instead of exponential decay [21, 22]. Incidentally, we cannot recommend to replace correlation functions $c_{(kl)(lk)}(t, 0)$ by phenomenological expressions that do not stem from a particular choice for H_1 . Such an uncontrolled move surely endangers the positivity of the density operator.

Finally, we argue that our non-Markovian formalism is capable of delivering exact results for atoms with an arbitrary number N of quantum levels. To see this in detail, we couple each atomic transition to a separate zero-temperature reservoir. Then (25) may be replaced by the stronger restriction

$$c_{(kl)(mn)}(t, s) = c_{(kl)(lk)}(t, s)\delta_{kn}\delta_{lm}. \quad (34)$$

For $l \geq k$ the right-hand side is vanishing because of the earlier made assumption $U_{(mn)}|0\rangle_R = 0$, with $m \geq n$. Upon employing (34) the iterative solution of (12) becomes identical to the exact perturbative series for the bitemporal operator. The latter is obtained by inserting (5) into (2) and making use of Wick's theorem. A proof of the foregoing surprising statement is outlined in the appendix. Altogether, (32) and (33), as well as the corresponding solutions for $N = 4, 5, 6, \dots$, are exact if (34) holds true and the reservoir is at absolute zero.

5. Finite temperature

From here onwards, we keep the reservoir at temperature β^{-1} and assume that ρ_R is in the thermal state $\exp(-\beta H_R)/\text{Tr}_R[\exp(-\beta H_R)]$. The spectrum of H_R can be rendered continuous in due course. The reservoir potentials no longer act on the ground state $|0\rangle_R$, so all reservoir correlation functions of type $c_{(kl)(lk)}$ differ from zero. The cyclic property of the trace permits us to establish the KMS identity

$$c_{(kl)(mn)}(t, 0) = c_{(mn)(kl)}(-t - i\beta, 0). \quad (35)$$

For the case of weak damping, (35) guarantees that the density operator $\rho_S(t)$ converges to a final state as t becomes large [23].

The foregoing statement motivates us to examine the asymptotic dynamics that is induced by (9) and (12) at finite temperature. First of all, we want to know whether the density operator still converges to a final state. To answer this question, we shall look for a particular solution $\xi_p(t, t')$ of (12) that becomes independent of time for the choice $t = t'$. In short, we shall investigate whether (12) allows for a fixed point. To avoid any misunderstandings, it should be stressed that the mere existence of a fixed point does not guarantee that the density operator really converges to a final state. The situation is the same as for a linear differential equation of type $df(t)/dt = L[f](t)$, where solution of $L[f](t) = 0$ does not provide a mathematically rigorous statement on the behaviour of $f(t)$ for large times.

In preparation of our search for a fixed point, we make a few crucial remarks on the solution of (9) at finite temperature. It still takes on the form (26). This becomes clear upon iterating (9) under use of (25). In (27) the sum over l now runs from 1 to N . Therefore, the asymptotic result $\lim_{t \rightarrow \infty} Q_k(t) = 0$ is valid for all k , because $\hat{Q}_1(z)$ no longer has a pole for $z = 0$. Since $Q_k(t)$ is a smooth function, the analytic continuation $Q_k(y)$, with y complex, exists in a certain strip around the axis $\text{Im } y = 0$. From (9) and the inequality $c_{(kl)(lk)}(ix, 0) \geq 0$, with x real, one deduces that $Q_k(-i\beta)$ is positive.

In the asymptotic regime, (12) possesses a fixed point. It is given by

$$\langle k|\xi_p(t, t')|l\rangle = \delta_{kl}Q_k(t - t' - i\beta)e^{-\beta\omega_k}/Z(\beta). \quad (36)$$

The choice $Z(\beta) = \sum_{k=1}^N Q_k(-i\beta) \exp(-\beta\omega_k)$ properly normalizes the trace of $\xi_p(t, t)$. To show that (36) is indeed a solution of (12), one needs the relation

$$Q_k(t - t') = Q_k(t) Q_k^*(t') + \lambda^2 \sum_{l=1}^N \int_0^t ds \int_0^{t'} ds' Q_k(s) Q_k^*(s') Q_l(t - t' - s + s') \times c_{(kl)(lk)}(t - t' - s + s', 0) e^{i(\omega_k - \omega_l)(t - t' - s + s')}. \quad (37)$$

To prove this, the substitutions $t \rightarrow t + u$ and $t' \rightarrow t' + u$ should be carried out, followed by differentiation with respect to u . The derivatives of Q_k are eliminated with the help of (9).

Let us now insert (36) into (12) and take profit of (25) as well as the KMS identity. In the ensuing relation we perform the integral transformations $s \rightarrow t - s$ and $s' \rightarrow t' - s'$. Subsequently, we perform the overall shift $t \rightarrow t + i\beta$. We divide the contour from $s = 0$ to $s = t + i\beta$ into a part with $0 \leq \text{Re } s \leq t$, $\text{Im } s = 0$, and a part with $\text{Re } s = t$, $0 \leq \text{Im } s \leq \beta$. After employment of (37), a few terms still remain. These vanish by setting t equal to t' and taking t to infinity. If β equals zero the last manipulations need not be carried out. In that case (36) is a solution of (12) for arbitrary t and t' .

The last observation tells us that in the classical limit, i.e., the limit of $\beta \rightarrow 0$, the state of maximum entropy, given by $\sum_{k=1}^N |k\rangle\langle k|/N$, acts as fixed point for the density operator. The inverse limit of $\beta \rightarrow \infty$ leads to a fixed point that is familiar as well. In evaluating the last-mentioned limit, one should be aware of the fact that the magnitude of $Q_k(-i\beta)$ is set by the dimensionless product $\lambda\beta\gamma$, where $\gamma^2 = \sum_{k,l=1}^N c_{(kl)(lk)}(0, 0)$ is an upper bound on all pair-correlation functions. The foregoing estimate can be derived by scaling (9) in a suitable manner. Altogether, for $\lambda\gamma < \omega_1$ and $\beta \rightarrow \infty$, the exponentials $\exp(-\beta\omega_k)$ become dominant in (36), so that $\langle k|\xi_p(t, t)|k\rangle$ converges to δ_{k1} for large t , as desired.

Finally, for λ tending to zero at fixed temperature, all weights $Q_k(-i\beta)$ converge to unity, by the same remark as made under (18). Then (36) reduces to the thermal state for $t = t'$. As a consequence, on the basis of a continuity argument in the norm $\|\lambda H_1\|$ one may claim that the fixed point (36) remains stable up to a certain, probably modest, value of $\|\lambda H_1\|$. In case the foregoing norm does not have a finite value, one should truncate the Hilbert space of system and reservoir by introducing provisional cut-offs.

Exactly solvable models, such as the Ullersma model [16] for the damped harmonic oscillator, teach us that in the strong-coupling regime quantum fluctuations seriously affect the dynamics, also for large times. Due to the rotating-wave approximation (25), we largely ignore quantum fluctuations. Hence, if the norms $\|H_0\|$ and $\|\lambda H_1\|$ become of comparable magnitude, in other words, if we approach the strong-coupling regime, (25) breaks down. Consequently, the fixed point (36) loses its physical relevance. Then the question of its stability becomes redundant. As we do not specify H_1 in this work, it is hard to estimate under which circumstances employment of (25) is still meaningful. If the bookkeeping parameter λ is set equal to unity, the inequality $\|H_1\| \ll \|H_0\|$ seems to offer a crude but safe criterion.

Both (9) and (12) simplify a lot under the use of (25). Unfortunately, at finite temperature the nonlinear character of (9) persists. As a consequence, analytic solution of (9) becomes a heavy task. There is only one escape out of this impasse. A way of linearizing (9) has to be developed that does not affect the basic physical properties of the density operator. Naturally, in making (9) analytically accessible one must be prepared to accept a major concession at some point.

Placing ourselves in the regime of moderate coupling, we shall sacrifice the Kraus criterion (14). Then positivity of the density operator is no longer guaranteed, but must be proved separately. We are fully aware of the fact that, strictly speaking, derivations violating the Kraus criterion fall outside the scope of this paper. On the other hand, it seems

to us that any physically meaningful result on non-Markovian damping at finite temperature is worth publishing, in view of applications to qubit dynamics, nuclear magnetic resonance, and other quantum processes. Indeed, the thermalized two-level atom has attracted considerable attention lately [11, 24].

Owing to (25), the diagonals of the density operator do not couple with any off-diagonals. Thus, it is meaningful to define an evolution matrix $\Lambda(t, t')$ as

$$\langle k | \xi(t, t') | k \rangle = \sum_{l=1}^N \Lambda(t, t')_{kl} \langle l | \rho_S | l \rangle. \quad (38)$$

We are going to subject Λ to a temperature-dependent transformation. Next, we shall compute the transformed matrix $\tilde{\Lambda}$ and, subsequently, return to the original frame.

Since the trace of $\xi(t, t)$ is conserved, the transformed matrix

$$\tilde{\Lambda}(t, t') = B \Lambda(t, t')^T B^{-1} \quad B_{kl} = \delta_{kl} - \delta_{k, l+1} \quad (39)$$

possesses the property

$$\tilde{\Lambda}(t, t) \cdot \mathbf{g} = \mathbf{g} \quad \mathbf{g}_k = \delta_{k1}. \quad (40)$$

We see that the density operator generated by $\tilde{\Lambda}(t, t)$ has the ground state $|1\rangle\langle 1|$ as fixed point. Our hope is now that in calculating $\tilde{\Lambda}(t, t')$ one can pick up some advantages of the zero-temperature case. We propose to calculate $\tilde{\Lambda}(t, t')$ from the following set:

$$\tilde{Q}_k(t) = 1 + \lambda^2 \sum_{l=1}^N \int_0^t ds \int_0^s du \tilde{G}_{kl}^{(1)}(s-u) \tilde{Q}_l(u) \quad (41)$$

$$\begin{aligned} \tilde{\Lambda}(t, t')_{kl} &= \tilde{Q}_k(t) \tilde{Q}_k^*(t') \delta_{kl} \\ &+ \lambda^2 \sum_{m=1}^N \int_0^t ds \int_0^{t'} ds' \tilde{Q}_k(t-s) \tilde{Q}_k^*(t'-s') \tilde{G}_{km}^{(2)}(s'-s) \tilde{\Lambda}(s, s')_{ml}. \end{aligned} \quad (42)$$

The generators $G^{(j)}$ and their transformation law will be given below.

Upon removing all tildes, the above set becomes identical to (9) and (12), at least, if the right-hand side of (9) is linearized. The operator $Q(u, v)$ must be replaced by unity. In the weak-coupling limit this linearization automatically takes place. Hence, the untransformed version of (42), that is to say, (42) without tildes, is capable of reproducing the Markovian density operator (22). Via the method explained in section 3 one proves that

$$\lim_{\lambda \rightarrow 0} \Lambda(t/\lambda^2, t/\lambda^2) = \exp \left\{ t \int_{-\infty}^{\infty} ds [G^{(1)}(s) + G^{(2)}(s)] \right\}. \quad (43)$$

By making a comparison with (22), we can identify the untransformed generators. The expressions

$$\begin{aligned} G^{(1)}(t)_{kl} &= -\delta_{kl} \sum_{m=1}^N e^{i(\omega_k - \omega_m)t} c_{(km)(mk)}(t, 0) \\ G^{(2)}(t)_{kl} &= e^{i(\omega_l - \omega_k)t} c_{(lk)(kl)}(t, 0) \end{aligned} \quad (44)$$

are found.

We now come to the central question of how to fix the transformed generators. There is only one rigorous possibility. We transform (9) and (12) in the way as prescribed by rule (39), and compare the ensuing equations with (41) and (42). It appears that in doing so, we do not benefit from any zero-temperature advantages. In particular, (42) cannot be solved by merely performing a finite number of iterations.

The situation drastically improves if the transformed generators are determined from

$$\tilde{G}^{(1)}(t) + \tilde{G}^{(2)}(t) = B[G^{(1)}(t)^T + G^{(2)}(t)^T]B^{-1}. \quad (45)$$

As in (44), we allot all terms containing a Kronecker delta to the first generator, and all remaining terms to the second one. We are led to

$$\begin{aligned} \tilde{G}^{(1)}(t)_{kl} &= -\delta_{kl} \sum_{m,n=1}^N e^{i(\omega_n - \omega_m)t} c_{(nm)(mn)}(t, 0) (\theta_{k,m+1} \delta_{kn} + \theta_{mk} \delta_{k,n+1}) \\ \tilde{G}^{(2)}(t)_{kl} &= \sum_{m,n=1}^N e^{i(\omega_n - \omega_m)t} c_{(nm)(mn)}(t, 0) (\theta_{k,l+1} \theta_{l,m+1} - \theta_{l,k+1} \theta_{ml}) (\delta_{k,n+1} - \delta_{k,n}). \end{aligned} \quad (46)$$

The symbol θ_{kl} is equal to 1 for $k \geq l$, and equal to 0 otherwise.

As testified by (43), in the van Hove limit the transformation (45) agrees with (39). Thus, (45) constitutes a reasonable approximation in the moderate-coupling regime, although the Kraus criterion (14) is abandoned. The last observation derives from the fact that a subtraction is carried out in (39).

The set (41)–(42), with (46) inserted, can be solved along the same lines as discussed in section 4. Once $\tilde{\Lambda}(t, t')$ is known, the inverse transformation (39) can be employed to compute $\Lambda(t, t')$, as well as the diagonals of the density operator. The identities $\tilde{G}^{(1)}(t)_{1k} = \tilde{G}^{(2)}(t)_{k1} = 0$ guarantee that (40) is true, so the trace of $\rho_S(t)$ is always conserved. Outside the van Hove limit we no longer obey (14). Therefore, one has to wait and see whether all diagonals of the density operator stay nonnegative if λ is increased.

For $N = 2$ execution of the above program is straightforward. As in the zero-temperature case, the function $\tilde{Q}_1(t)$ equals unity. A comparison with the zero-temperature result (31) directly yields

$$\begin{aligned} \tilde{Q}_2(t) &= \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \\ &\times [\omega - \lambda^2 \hat{c}_{(21)(12)}(\omega + \omega_2 - \omega_1 + i\epsilon) - \lambda^2 \hat{c}_{(12)(21)}(\omega + \omega_1 - \omega_2 + i\epsilon)]^{-1}. \end{aligned} \quad (47)$$

To get $\tilde{\Lambda}(t, t')$, only a single iteration of (42) is required. Use of the inverse transformation (39) provides us then with the following extension of (32):

$$\langle 2|\rho_S(t)|2\rangle = P_{12}(t)\langle 1|\rho_S|1\rangle + [1 - P_{21}(t)]\langle 2|\rho_S|2\rangle. \quad (48)$$

For notational convenience, the abbreviation

$$P_{kl}(t) = \lambda^2 \int_0^t ds \int_0^t ds' \tilde{Q}_2(s) \tilde{Q}_2^*(s') e^{i(\omega_k - \omega_l)(s' - s)} c_{(kl)(lk)}(s', s) \quad (49)$$

has been introduced. Note that the evolution of the ground state is described by $\langle 1|\rho_S(t)|1\rangle = 1 - \langle 2|\rho_S(t)|2\rangle$. In view of the identity

$$P_{12}(t) + P_{21}(t) + |\tilde{Q}_2(t)|^2 = 1 \quad (50)$$

and the inequality $P_{kl}(t) \geq 0$, both diagonals of $\rho_S(t)$ cannot become negative. Although the Kraus criterion is not obeyed, the density operator (48) meets the requirements of positivity and trace conservation. The task of checking (50) is fulfilled via differentiation with respect to t .

A useful application of (48) is a non-Markovian description of finite-temperature atomic decay. At equilibrium, the relative occupation number of the excited level is given by

$$\lim_{t \rightarrow \infty} \frac{\langle 2|\rho_S(t)|2\rangle}{\langle 1|\rho_S(t)|1\rangle} = \frac{P_{12}(\infty)}{P_{21}(\infty)}. \quad (51)$$

This is a non-Markovian counterpart of the Boltzmann factor $\exp[-\beta(\omega_2 - \omega_1)]$. For weak coupling (48) is equivalent to the well-known optical Bloch equations [17]. In checking this assertion, one has to evaluate the van Hove limit of (49). The double integral must be treated in the same way as explained under (21).

Finally, we mention that in order to solve (42) for $N = 3$, an infinite number of iterations is needed. In contrast, at zero temperature only two iterations are sufficient. Therefore, one can very well argue that the full iterative solution of (42) may be truncated as long as the temperature is kept sufficiently low.

6. Conclusion

Adopting a bitemporal approach, we succeeded in summing up a subseries of the standard perturbative expansion for the density operator of a quantum system that exchanges energy with a reservoir. The dissipative dynamics we found complies with the Kraus map (14) and conserves probability. For the case of weak coupling between system and reservoir, we recovered the complete van Hove–Davies theory of quantum damping. The closed set (9) and (12) constitutes our main result. The approximations and assumptions underlying this set are listed below.

(i) System and reservoir were assumed to be completely uncorrelated at time zero. In the literature [25], it was pointed out that failure to account for initial entanglement might lead to a poor description, especially if the coupling between system and reservoir becomes strong. Unfortunately, our attempts to incorporate entanglement in the present treatment were not successful. A major obstacle is that entanglement causes the dependence of the density operator on its initial state to become nonlinear [26, 27].

(ii) Reservoir correlation functions were factorized into products of pair-correlation functions. For a thermal reservoir this is indeed allowed, in virtue of Wick’s theorem [17]. For other cases, such as a squeezed reservoir, one may question the decision of dealing with the reservoir through pair-correlation functions only. On the other hand, we stress that if one leaves all correlation functions intact, the possibility of summing up a perturbative series gets lost. Higher order terms can no longer be expressed as products of lower order terms. At this point, we should mention that correlation functions can always be factorized by invoking the completeness relation for the reservoir Hilbert space. However, this possibility is impractical, because it does not match with our basic intention of deriving a description that is completely embedded in the Hilbert space of the system.

(iii) In appealing to Wick’s theorem we systematically discarded contractions that are not of generalized nearest-neighbour type. In the van Hove limit this automatically happens, so for moderate values of λ the error we made might be modest still. We recall that no error is made at all, if (34) is assumed and the temperature equals zero. Of course, the performance of (12) is also influenced by the choice of initial conditions. For a damped harmonic oscillator that starts from a coherent state of high energy, all Wick contractions are needed, even if the reservoir is kept at absolute zero. We can try to improve on the generalized nearest-neighbour approximation by performing all three Wick contractions for the correlation function {1234}. It appears that upon including the irreducible graph {13}{24} into equations (9) and (12), the Kraus criterion (14) is no longer fulfilled. A complete discussion is presented in the appendix.

(iv) The Hilbert space of the system had to be chosen as separable. If not, the decomposition (3) of the interaction Hamiltonian ceases to exist. Separability is not a severe limitation, because in practice one encounters many damped systems the energy spectrum of which is discrete. Examples are the N -level atom and the harmonic oscillator.

(v) We worked with a system Hamiltonian that does not depend on time. This is not a serious deficit either. The presence of a classical field does not alter our equations, except for (4). There the unitary operator $\exp(iH_S t)$ makes way for the time-ordered form $T \left\{ \exp \left[i \int_0^t dt' H_S(t') \right] \right\}$, where T denotes a time-ordering operator.

In section 4 we outlined how the set (9) and (12) can be solved for the case of an N -level atom or harmonic oscillator at zero temperature. The rotating-wave approximation (25) turned out to be indispensable. Making use of the solutions (31), we wrote down clear-cut expressions for the density operator of a damped two-level and three-level atom. In further evaluating the integrals showing up in (31), one has to search for analytic continuation of the integrands below the real axis. The circumstance that one has to struggle with a branch cut is not incidental, but very much typical for a non-Markovian treatment of quantum dissipation. If one desires to steer clear of branch cuts, one either has to resort to Markovian theory, or undertake a direct numerical computation of the density operator. In general, the second option is useful for testing concrete conjectures on a given system. It is not particularly helpful in deepening our understanding of the physics behind quantum dissipative processes.

In section 5 the temperature of the reservoir was taken to be finite. The resulting nonlinear character of (9) kept us back from deriving analytical solutions of (12). In spite of that, we managed to demonstrate that for moderate coupling the density operator is likely to end up in the vicinity of the familiar thermal state. In order to clear the way to analytical work, we linearized (9) and gave up our intention of meeting constraint (14). Therefore, our non-Markovian description (48) of a two-level atom immersed in a thermal bath should, strictly speaking, not form part of this paper. Nevertheless, we feel that the derivation (38)–(51) is worth publishing, because analytical results on finite-temperature damping are rather scarce.

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Appendix. Going beyond the generalized nearest-neighbour approximation

In section 2 we factorized higher order reservoir correlation functions by invoking the generalized nearest-neighbour approximation. This led to the factorization scheme (8) and, eventually, to the evolution equations (9) and (12). By construction, this set generates a density operator that is exact up to order λ^2 . The first error already appears in order λ^4 , because in the Wick identity $\{1234\} = \{12\}\{34\} + \{13\}\{24\} + \{14\}\{23\}$ the irreducible graph $\{13\}\{24\}$ is discarded. Hence, the contribution $\{13\}\{24\}$ represents the lowest order correction to the generalized nearest-neighbour approximation.

Inspection of (9) shows that the irreducible part $\{12\}$ must be accompanied by the integrand $V_{\alpha_1}(t_1)Q(t_1, t_2)V_{\alpha_2}(t_2)Q(t_2, s)c_{\alpha_1\alpha_2}(t_1, t_2)$. Hence, the integrand going with $\{13\}\{24\}$ should read

$$V_{\alpha_1}(t_1)Q(t_1, t_2)V_{\alpha_2}(t_2)Q(t_2, t_3)V_{\alpha_3}(t_3)Q(t_3, t_4)V_{\alpha_4}(t_4)Q(t_4, s)c_{\alpha_1\alpha_3}(t_1, t_3)c_{\alpha_2\alpha_4}(t_2, t_4). \quad (\text{A.1})$$

Extension of (9) with the irreducible part {13}{24} thus leads to

$$\begin{aligned}
Q(t, s) = & 1_S - \lambda^2 \sum_{\alpha_1 \alpha_2} \int_s^t dt_1 \int_s^{t_1} dt_2 V_{\alpha_1}(t_1) Q(t_1, t_2) V_{\alpha_2}(t_2) Q(t_2, s) c_{\alpha_1 \alpha_2}(t_1, t_2) \\
& + \lambda^4 \sum_{\alpha_1 \alpha_2 \alpha_3 \alpha_4} \int_s^t dt_1 \int_s^{t_1} dt_2 \int_s^{t_2} dt_3 \int_s^{t_3} dt_4 V_{\alpha_1}(t_1) Q(t_1, t_2) V_{\alpha_2}(t_2) Q(t_2, t_3) \\
& \times V_{\alpha_3}(t_3) Q(t_3, t_4) V_{\alpha_4}(t_4) Q(t_4, s) c_{\alpha_1 \alpha_3}(t_1, t_3) c_{\alpha_2 \alpha_4}(t_2, t_4). \tag{A.2}
\end{aligned}$$

Note that the sign of the fourth-order contribution is determined by the identity $i^4 = 1$.

Incorporation of the irreducible part {13}{24} into (12) is more involved, because the bitemporal operator ξ must be placed inside the product $VVVV$ in all possible ways. This yields the forms $V\xi VVV$, $VV\xi VV$ and $VVV\xi V$. As in (12), operators Q must be inserted into these three forms such that ξ and V remain neighbours. Then one collects all Wick contractions that can be assembled from the irreducible parts {12} and {13}{24}.

The foregoing considerations bring us to the following extension of (12):

$$\begin{aligned}
\xi(t, t') = & Q(t, 0) \rho_S Q^\dagger(t', 0) \\
& + \lambda^2 \sum_{\alpha_1 \alpha_1'} \int_0^t dt_1 \int_0^{t'} dt_1' Q(t, t_1) V_{\alpha_1}(t_1) \xi(t_1, t_1') V_{\alpha_1'}(t_1') Q^\dagger(t', t_1') c_{\alpha_1' \alpha_1}(t_1', t_1) \\
& - \lambda^4 \sum_{\alpha_1 \alpha_1' \alpha_2' \alpha_3'} \int_0^t dt_1 \int_0^{t'} dt_1' \int_0^{t_1'} dt_2' \int_0^{t_2'} dt_3' Q(t, t_1) V_{\alpha_1}(t_1) \xi(t_1, t_3') V_{\alpha_3'}(t_3') \\
& \times Q^\dagger(t_2', t_3') V_{\alpha_2'}(t_2') Q^\dagger(t_1', t_2') V_{\alpha_1'}(t_1') Q^\dagger(t', t_1') c_{\alpha_2' \alpha_1}(t_2', t_1) c_{\alpha_3' \alpha_1'}(t_3', t_1') \\
& + \lambda^4 \sum_{\alpha_1 \alpha_2 \alpha_1' \alpha_2'} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_1'} dt_1' \int_0^{t_1'} dt_2' Q(t, t_1) V_{\alpha_1}(t_1) Q(t_1, t_2) V_{\alpha_2}(t_2) \\
& \times \xi(t_2, t_2') V_{\alpha_2'}(t_2') Q^\dagger(t_1', t_2') V_{\alpha_1'}(t_1') Q^\dagger(t', t_1') c_{\alpha_2' \alpha_1}(t_2', t_1) c_{\alpha_1' \alpha_2}(t_1', t_2) \\
& - \lambda^4 \sum_{\alpha_1 \alpha_2 \alpha_3 \alpha_1'} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_1'} dt_1' Q(t, t_1) V_{\alpha_1}(t_1) Q(t_1, t_2) V_{\alpha_2}(t_2) \\
& \times Q(t_2, t_3) V_{\alpha_3}(t_3) \xi(t_3, t_1') V_{\alpha_1'}(t_1') Q^\dagger(t', t_1') c_{\alpha_1 \alpha_3}(t_1, t_3) c_{\alpha_1' \alpha_2}(t_1', t_2). \tag{A.3}
\end{aligned}$$

If the irreducible part {13}{24} makes no contribution, as is for instance the case in the van Hove limit, then (A.2) and (A.3) reduce to the old evolution equations (9) and (12).

Following the same procedure as in section 3, we compute the derivative of $\text{Tr}_S[\xi(t, t)]$ with respect to t . A lengthy calculation shows that this derivative is equal to zero. Consequently, (A.2) and (A.3) conserve the trace of the density operator. For the choice $t' = t$ the solution of the adjoint equation (A.3) is identical to the solution of (A.3) itself. Hence, (A.3) gives rise to a self-adjoint density operator. Furthermore, one verifies by induction that an iteration of (A.2) and (A.3) yields the perturbative series that is obtained from (2), (5) and (8), provided that the right-hand side of (8) be systematically extended with the irreducible graph {13}{24}. Unfortunately, (A.3) brings us some bad news as well. The two terms of order λ^4 , the sign of which is negative, do not comply with the Kraus criterion (14).

The exact perturbative series for $\xi(t, t')$ contains the term $(V + VVV)\rho_S(V + VVV)$, which does comply with the Kraus criterion. This tells us that terms of types $V\xi VVV$ and $VVV\xi V$ may be taken into account, only if they are accompanied by the sixth-order term

$VVV\xi VVV$. Hence, (A.3) can be reconciled with the Kraus criterion if one is prepared to consider terms containing irreducible graphs of order λ^6 . Four different species exist, namely

$$\{13\}\{25\}\{46\} \quad \{14\}\{26\}\{35\} \quad \{15\}\{24\}\{36\} \quad \{14\}\{25\}\{36\}. \quad (\text{A.4})$$

As before, the constraint of trace conservation prescribes that the graphs (A.4) be combined with all possible products of ξ and system potentials V . Hence, apart from the Kraus consistent form $VVV\xi VVV$, we also have to allow the forms $V\xi VVVVV$ and $VV\xi VVVV$, as well as their adjoints. Once again the Kraus criterion is compromised.

The material presented above permits us to put forward the following tentative conclusion: collect all irreducible graphs up to a fixed order in λ ; construct from these graphs evolution equations for Q and ξ in the same way as for (9), (12), (A.2) and (A.3), i.e., by interleaving system potentials with operators Q and ξ in a suitable manner. The ensuing evolution equations conserve the trace and self-adjointness of the density operator. They do not comply with the Kraus criterion, unless we work in lowest, that is to say, quadratic order of λ . Of course, the more irreducible graphs we take into account, the closer we approach the exact perturbative series for the density operator.

The foregoing statements imply that for a given Hamiltonian the accuracy of (9) and (12) can be assessed by estimating the relative magnitude of higher order irreducible graphs such as $\{13\}\{24\}$ and the graphs (A.4). As we shall argue below, under assumption (34) all irreducible graphs vanish at zero temperature, except for $\{12\}$. Consequently, (9) and (12) give rise to the exact density operator, i.e., the density operator that is obtained by applying Wick's theorem to (2) and (5). Note that use of Wick's theorem is surely permitted as long as all reservoirs are made up by harmonic oscillators.

Our proof is based on the representation (23). We first focus on (A.2). A key role is played by the identity

$$V_{\alpha_1} V_{\alpha_2} V_{\alpha_3} V_{\alpha_4} c_{\alpha_1 \alpha_3} c_{\alpha_2 \alpha_4} = |k_1\rangle \langle l_4 | \delta_{k_2 l_1} \delta_{k_3 l_2} \delta_{k_4 l_3} c_{(k_1 l_1)(l_2 l_3)} c_{(l_1 l_2)(l_3 l_4)}, \quad (\text{A.5})$$

where time arguments are irrelevant. From (34) it follows that the right-hand side of (A.5) is vanishing at zero temperature. By induction one proves then that the iterative solution of (A.2) possesses the following two properties: (i) $\langle p | Q(t, s) | q \rangle$ equals zero for $p \neq q$, and, (ii) the fourth-order term figuring in (A.2) does not contribute at all.

Let us now extend (A.2) with irreducible graphs of arbitrary order $2n$. In that case one has to implement (34) in the following expression:

$$V_{\alpha_1} V_{\alpha_2} V_{\alpha_3} V_{\alpha_4} \cdots V_{\alpha_{2n}} c_{\alpha_{j_1} \alpha_{j_2}} c_{\alpha_{j_3} \alpha_{j_4}} \cdots c_{\alpha_{j_{2n-1}} \alpha_{j_{2n}}}. \quad (\text{A.6})$$

The product of correlation functions corresponds to an irreducible graph consisting of n pairs. Elementary topological analysis shows that this graph always contains a segment $\{m, m+n\} \prod_{j=1}^{n-1} \{m+j, p_j\}$, with $n \geq 2$ and $p_j > m+n$ for all j . To the irreducible graph $\{13\}\{25\}\{46\}$ the choice $m=1, n=2$, and $p_1=5$ applies. With the help of (23) and the choice $\alpha_j \rightarrow (k_j l_j)$ we find that the afore-mentioned segment generates a product

$$c_{(k_m l_m)(l_{m+n-1} l_{m+n})} c_{(l_m l_{m+1})(\cdots)} c_{(l_{m+1} l_{m+2})(\cdots)} \cdots c_{(l_{m+n-2} l_{m+n-1})(\cdots)}. \quad (\text{A.7})$$

As in (A.5), the orthogonality of the states $\{|k\rangle\}$ has been exploited. Since the temperature is at absolute zero, the inequalities $l_m > l_{m+1} > l_{m+2} \cdots > l_{m+n-2} > l_{m+n-1}$ must hold true. On the other hand, (34) prescribes that one must choose $l_{m+n-1} = l_m$ in the first correlation function. This leaves us only one option: we have to conclude that the form (A.6) equals zero for $n \geq 2$.

Making use of induction, one proves that the iterative solution for $Q(t, s)$ is diagonal and independent of the graph (A.6). In other words, (9) is exact if (34) is valid and the temperature equals zero. The same statement can be proved for (12) by means of similar techniques as

outlined above. The diagonals $\langle p|\xi(t, t')|p\rangle$ and off-diagonals $\langle p|\xi(t, t')|q\rangle$, with $p \neq q$, must be treated separately. In order to gain some expertise in structuring the proofs, it is recommendable to first prove that (A.3) is equivalent to (12) under the assumptions stated above.

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