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Kraus map for non-Markovian quantum dynamics driven by a thermal reservoir

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Abstract – Starting from unitary dynamics we study the evolution in time of a non-relativistic quantum system that exchanges energy with a thermal reservoir of harmonic oscillators. System and reservoir are assumed to be initially decorrelated. Reservoir correlation functions are factorized by means of a Kraus compliant version of Wick's theorem. As a result, the non-Markovian Kraus map for the system density operator can be completely expressed in terms of system potentials and reservoir pair correlation functions. An infinite hierarchy for the evolution operators of the Kraus map is derived. The system density operator is obtained as a time-ordered exponential containing a non-Markovian counterpart of the standard Markovian generator for dissipative dynamics. We specify a condition on this non-Markovian generator for return to thermal equilibrium. We set up a non-Markovian perturbation theory that preserves both trace and positivity of the system density operator.

Introduction. – More than 40 years ago, Kraus [1] studied the evolution in time t of a quantum system that exchanges energy with a reservoir. Defining an infinite set of formal system operators $\{W_q(t)\}$, he pointed out that the system density operator evolves according to the map $\sum_q W_q(t)\rho_S W_q^\dagger(t)$. This is a manifestly positive form for any positive initial state ρ_S. In other words, the Kraus map is faithful to the basic physical principle that states with negative probability do not occur.

In view of the last observation the Kraus map may be considered as the natural representation of a quantum dissipative evolution. At the same time, this virtue is of limited practical value as long as we are stuck with the formal character of the Kraus operators $\{W_q(t)\}$. Therefore, to get the most out of the Kraus representation we should express its operators in terms of system potentials and reservoir correlation functions. In order to achieve that goal we must construct a road leading from any prescribed Hamiltonian to the exact Kraus map. Preliminary studies suggest the existence of such a road. In [2] an approximate Kraus description of quantum dissipative dynamics was obtained.

Our treatment relies on three assumptions. First, we work with a separable Hilbert space of the system. Second, at time $t = 0$ correlations between system and reservoir are absent. Here it should be noted that initial correlations have been the subject of some discussion in the recent literature [3]. Third, our reservoir consists of a set of harmonic oscillators that linearly interact with the system and that are in thermal equilibrium at $t = 0$.

Owing to the third assumption, reservoir correlation functions can be factorized by use of Wick's theorem. If we perform this in a Kraus compliant manner, the desired expressions for the Kraus operators materialize without much effort. It appears that the Kraus operators satisfy an infinite hierarchy of evolution equations. Moreover, the system density operator is found to depend on a single non-Markovian generator $L(t)$. With the help of the latter one can make a precise statement on the dissipative dynamics for large times.

Despite its decades-long history, documented in numerous fine books and reviews [4], the subject of quantum dissipation is still very much alive. Triggered by experimental advances, a large part of the current theoretical activity [5] is directed towards the important issue of embedding non-Markovian dynamics in the system Hilbert space. In the present letter we show how this demanding program can be carried out. Our theory is quite general and therefore applicable to a wide variety of physical problems.

Time-ordered representation. – The non-relativistic quantum system we study is called S. It

exchanges energy with a large reservoir R. As announced, we assume that at time $t = 0$ system and reservoir are uncorrelated. Hence, the initial condition for the composite density operator $\rho_{SR}(t)$ of system and reservoir reads $\rho_{SR}(t=0) = \rho_S \otimes \rho_R$. The system starts from state ρ_S and the reservoir from state ρ_R.

The energy of system and reservoir together is conserved, so that the composite density operator obeys the unitary evolution law,

$$\rho_{SR}(t) = \mathrm{e}^{iH_0 t}\mathrm{e}^{-iHt}\rho_S \otimes \rho_R \mathrm{e}^{iHt}\mathrm{e}^{-iH_0 t}. \tag{1}$$

We work in the interaction picture and scale the Hamiltonian $H = H_0 + H_1$ with Planck's constant. The Hamiltonian H_S of the system and the Hamiltonian H_R of the reservoir make up the free Hamiltonian $H_0 = H_S \otimes 1_R + 1_S \otimes H_R$. The interaction Hamiltonian H_1 is expanded as

$$H_1 = \sum_\alpha V_\alpha \otimes U_\alpha. \tag{2}$$

If the system Hilbert space is spanned by a countable number of ket vectors, then (2) is surely true. The system potential V_α and the reservoir potential U_α evolve as

$$V_\alpha(t) = \mathrm{e}^{iH_S t}V_\alpha \mathrm{e}^{-iH_S t}, \qquad U_\alpha(t) = \mathrm{e}^{iH_R t}U_\alpha \mathrm{e}^{-iH_R t}. \tag{3}$$

Note that these potentials need not be self-adjoint.

We expand the evolution operators figuring in (1) as

$$\begin{aligned}&\mathrm{e}^{iH_0 t}\mathrm{e}^{-iHt} = 1_S \otimes 1_R \\ &+ \sum_{m=1}^{\infty}\sum_{\alpha_1\cdots\alpha_m}(-i)^m \int_0^t \mathrm{d}t_1 \cdots \int_0^{t_{m-1}} \mathrm{d}t_m \\ &\times V_{\alpha_1}(t_1)\cdots V_{\alpha_m}(t_m) \otimes U_{\alpha_1}(t_1)\cdots U_{\alpha_m}(t_m).\end{aligned} \tag{4}$$

Subsequently, we obtain the dissipative dynamics of S by computing the system density operator $\rho_S(t) = \mathrm{Tr}_R[\rho_{SR}(t)]$. We encounter reservoir correlation functions of arbitrarily high order.

The reservoir is assumed: i) to consist of a collection of harmonic oscillators, ii) to linearly interact with the system, and iii) to start from the thermal state $\rho_R = \exp(-\beta H_R)/Z_R$, where Z_R equals $\mathrm{Tr}_R[\exp(-\beta H_R)]$ and β measures the inverse temperature. Consequently, the commutator $[\rho_R, H_R]$ is vanishing so that the average $\mathrm{Tr}_R[U_\alpha(t)\rho_R]$ no longer depends on time. Since H_S can be modified such that $\mathrm{Tr}_R[U_\alpha(0)\rho_R]$ equals zero, all correlation functions of odd order may be discarded in the following.

Correlation functions of even order can be factorized with the help of Wick's theorem, which is expressed below in a Kraus compliant way. If the conditions $t_1 > \cdots > t_m$ and $t'_1 > \cdots > t'_n$ hold true, one has for $m+n$ even

$$\begin{aligned}&\mathrm{Tr}_R\left[U_{\alpha_1}(t_1)\cdots U_{\alpha_m}(t_m)\rho_R U_{\alpha'_n}(t'_n)\cdots U_{\alpha'_1}(t'_1)\right] = \\ &\sum_{q=0}^{\min(m,n)} \frac{1}{q!} b\left(\frac{m-q}{2}\right) b\left(\frac{n-q}{2}\right) \sum_{PQ}\prod_{k=1}^{q}\{Q(k)' \quad P(k)\} \\ &\times \prod_{i=1}^{(m-q)/2}\{P(q+2i-1) \quad P(q+2i)\}^{(+)} \\ &\times \prod_{j=1}^{(n-q)/2}\{Q(q+2j-1)' \quad Q(q+2j)'\}^{(-)}.\end{aligned} \tag{5}$$

This identity shows that the thermal reservoir rules the system with the pair correlation function

$$c_{\alpha_1\alpha_2}(t_1,t_2) = \mathrm{Tr}_R\left[U_{\alpha_1}(t_1)U_{\alpha_2}(t_2)\rho_R\right] \equiv \{1 \quad 2\}. \tag{6}$$

The bracket $\{1' \quad 2\}$ denotes the left-hand side of (6) with t_1, α_1 replaced by t'_1, α'_1. The bracket $\{1' \quad 2'\}$ denotes the left-hand side of (6) with $t_1, \alpha_1, t_2, \alpha_2$ replaced by $t'_1, \alpha'_1, t'_2, \alpha'_2$. In (5) we sum over all permutations P of the integers $\{1,\ldots,m\}$ and all permutations Q of the integers $\{1,\ldots,n\}$. To avoid double counts of Wick factorizations, the combinatorial factors $q!$ and $b(k) = 1/(2^k k!)$ are included. For non-integer values of k one must set $b(k)$ equal to zero. The potentials of each pair correlation function must appear in the same order as on the left-hand side of (5). This requirement is met through the prescriptions

$$\begin{aligned}c^{(+)}_{\alpha_1\alpha_2}(t_1,t_2) &= \{12\}\theta(t_1-t_2) + \{21\}\theta(t_2-t_1), \\ c^{(-)}_{\alpha_1\alpha_2}(t_1,t_2) &= \{12\}\theta(t_2-t_1) + \{21\}\theta(t_1-t_2),\end{aligned} \tag{7}$$

where $\theta(t)$ denotes the Heaviside step function.

In all contributions to $\rho_S(t)$ the initial state ρ_S is multiplied from the left and from the right by potentials $V_\alpha(t)$. These are ordered via the integral boundaries of the expansion (4). This can be made explicit by means of the time-ordering prescriptions

$$\begin{aligned}&\mathcal{T}\left\{\prod_{i=1}^{m} V_{\alpha_i}(t_i)\rho_S \prod_{j=1}^{n} V_{\alpha'_j}(t'_j)\right\} = \\ &\mathcal{T}_+\left\{\prod_{i=1}^{m} V_{\alpha_i}(t_i)\right\}\rho_S \mathcal{T}_-\left\{\prod_{j=1}^{n} V_{\alpha'_j}(t'_j)\right\},\end{aligned} \tag{8}$$

where we define $\mathcal{T}_+\{\prod_{i=1}^m V_{\alpha_i}(t_i)\} = V_{\alpha_1}(t_1)\cdots V_{\alpha_m}(t_m)$ and $\mathcal{T}_-\{\prod_{j=1}^n V_{\alpha'_j}(t'_j)\} = V_{\alpha'_n}(t'_n)\cdots V_{\alpha'_1}(t'_1)$. The inequalities $t_1 > \cdots > t_m$ and $t'_1 > \cdots > t'_n$ are in force.

By definition (8), the system density operator as determined from (1), (4) and (5) possesses the property $\rho_S(t) = \mathcal{T}\rho_S(t)$, with $t > 0$. On the right-hand side of this identity we may freely permute system potentials. We thus can perform the sums over P and Q figuring in (5). Upon interchanging the sum over q with the sums over

m and n and shifting summation indices as $m \to m+q$, $n \to n+q$ we can perform the sums over m and n as well. Then the exponential operator $\exp[K^{(\eta)}(t,s)]$ comes into play, with $\eta = \pm 1$ and the generator given by

$$K^{(\eta)}(t,s) = -\frac{1}{2}\sum_{\alpha\beta}\int_s^t \mathrm{d}u \int_s^t \mathrm{d}v\; c^{(\eta)}_{\alpha\beta}(u,v)\mathcal{T}_\eta\left\{V_\alpha(u)V_\beta(v)\right\}. \quad (9)$$

On the right-hand side the symmetrized correlation functions (7) appear.

If we define the Kraus operators

$$W^{(\eta)}_q(t;t_1,\cdots,t_q;s)_{\alpha_1\cdots\alpha_q} = \mathcal{T}_\eta\left\{\exp[K^{(\eta)}(t,s)]\prod_{i=1}^q V_{\alpha_i}(t_i)\right\}, \quad (10)$$

we can represent the system density operator for $t>0$ as

$$\rho_S(t) = W^{(+)}_0(t;0)\rho_S W^{(-)}_0(t;0) + \sum_{q=1}^\infty \sum_{\alpha_1\cdots\alpha_q}\sum_{\alpha'_1\cdots\alpha'_q} \times \int_0^t \mathrm{d}t_1\cdots\int_0^t \mathrm{d}t_q\int_0^t \mathrm{d}t'_1\cdots\int_0^t \mathrm{d}t'_q \frac{1}{q!}\prod_{k=1}^q\{k'k\} \times W^{(+)}_q(t;t_1,\cdots,t_q;0)_{\alpha_1\cdots\alpha_q}\rho_S W^{(-)}_q(t;t'_1,\cdots,t'_q;0)_{\alpha'_1\cdots\alpha'_q}. \quad (11)$$

The system's evolution is completely determined by pair correlation functions and Kraus operators $\{W^{(\eta)}_q\}$. The latter act on the system Hilbert space. Note that by expanding the pair correlation functions $\{k'k\}_{k=1}^q$ with the help of reservoir eigenstates one can cast the dynamical map (11) into the diagonal Kraus form appearing in the introduction.

If we define the superoperator

$$L(t)\rho_S = K^{(+)}(t,0)\rho_S + \rho_S K^{(-)}(t,0) + \sum_{\alpha\beta}\int_0^t \mathrm{d}u\int_0^t \mathrm{d}v c_{\beta\alpha}(v,u)V_\alpha(u)\rho_S V_\beta(v) \quad (12)$$

we can cast (11) into the concise form

$$\rho_S(t) = \mathcal{T}\mathrm{e}^{L(t)}\rho_S, \quad (13)$$

with $t>0$. By making the choice $t=0$ we see that the relation $\mathcal{T}\rho_S = \rho_S$ must be true.

By carrying out the replacements $V_\alpha \to \lambda V_\alpha$ as well as $t \to t/\lambda^2$, and by making use of a Kato [6] identity for the form $\mathcal{T}\exp[L(t/\lambda^2)]\rho_S$, we obtain the familiar result $\lim_{\lambda\to 0}\rho_S(t/\lambda^2) = \exp(Lt)\rho_S$ [7]. The generator is given by $L = L_1 + L_2 + L_3$, with $L_1\rho_S = [K - K^\dagger, \rho_S]/2$ and $L_2\rho_S = \{K + K^\dagger, \rho_S\}/2$. The system operator K and the superoperator L_3 are defined as

$$K = -\lim_{T\to\infty}\sum_{\alpha\beta}\frac{1}{T}\int_0^T \mathrm{d}t\int_0^\infty \mathrm{d}s c_{\alpha\beta}(s,0)V_\alpha(t+s)V_\beta(t), \quad (14)$$

$$L_3\rho_S = \lim_{T\to\infty}\sum_{\alpha\beta}\frac{1}{T}\int_0^T \mathrm{d}t\int_{-\infty}^\infty \mathrm{d}s c_{\alpha\beta}(s,0)V_\beta(t)\rho_S V_\alpha(t+s). \quad (15)$$

The above limit $\lambda \to 0$ surely exists if the conditions

$$\sum_\alpha \|V_\alpha\| < \infty, \qquad \sup_{\alpha\beta}\int_{-\infty}^\infty \mathrm{d}s|s c_{\alpha\beta}(s,0)| < \infty \quad (16)$$

are satisfied.

From the weak-coupling limit we learn that (13) may be viewed as the non-Markovian extension of the standard Markovian map $\exp(Lt)\rho_S$. Unfortunately, computation of (13) constitutes a very difficult mathematical problem. In contrast to the Markovian case, differentiation of (13) does not produce a differential equation for $\rho_S(t)$. The reason is that the derivative $\mathrm{d}L(t)/\mathrm{d}t$ may not be taken outside the time-ordered form $\mathcal{T}\{\mathrm{d}L(t)/\mathrm{d}t\exp[L(t)]\rho_S\}$. Hence, in describing non-Markovian dynamics we should not focus on $\rho_S(t)$ itself but rather on the Kraus operators $\{W^{(\eta)}_q\}$. Their evolution will be investigated now.

Hierarchy for Kraus operators. – By way of the symmetrized Wick factorization (5) we were led to the conclusion that the Kraus operators (10) are the natural candidates for describing our dissipative quantum system. To find out how these operators evolve we free the potential $V_{\alpha_1}(t_1)$ from the time-ordered product figuring in (10). In doing so, we concentrate on the domain that is defined by the inequalities $t_1 > \cdots > t_q$. As will become clear below, there is no need to operate outside this domain.

We employ (7), (9) and (10) to arrive at the Kato identity [6],

$$W^{(+)}_q(t;t_1,\cdots,t_q;s)_{\alpha_1\cdots\alpha_q} = \mathcal{T}_+\left\{\exp[K^{(+)}(t_1,s)]\prod_{i=1}^q V_{\alpha_i}(t_i)\right\} - \sum_{\alpha\beta}\int_{t_1}^t \mathrm{d}u\int_s^u \mathrm{d}v \times c_{\alpha\beta}(u,v)\mathcal{T}_+\left\{\exp[K^{(+)}(u,s)]V_\alpha(u)V_\beta(v)\prod_{i=1}^q V_{\alpha_i}(t_i)\right\}. \quad (17)$$

Differentiation with respect to t shows that (17) is true.

In the first term on the right-hand side of (17) the potential $V_{\alpha_1}(t_1)$ may be taken outside the time-ordered product. Note that one has $t_1 > w$ for all potentials $V_\gamma(w)$ contributing to $\exp[K^{(+)}(t_1,s)]$. The same argument can be applied to the potential $V_\alpha(u)$ figuring in (17). One

then arrives at

$$W_q^{(+)}(t;t_1,\cdots,t_q;s)_{\alpha_1\cdots\alpha_q} = V_{\alpha_1}(t_1)W_{q-1}^{(+)}(t_1;t_2,\cdots,t_q;s)_{\alpha_2\cdots\alpha_q} - \sum_{j=1}^{q+1}\sum_{\alpha\beta}\int_{t_1}^{t}\mathrm{d}u\int_{t_j}^{t_{j-1}}\mathrm{d}v c_{\alpha\beta}(u,v)V_\alpha(u) \times W_{q+1}^{(+)}(u;t_1,\cdots,t_{j-1},v,t_j,\cdots,t_q;s)_{\alpha_1\cdots\alpha_{j-1}\beta\alpha_j\cdots\alpha_q}, \quad (18)$$

with $t > t_1 > \cdots > t_q > s$ and $q = 1, 2, \ldots$. In evaluating the boundaries of the integral over v, one has to choose $t_0 = u$ and $t_{q+1} = s$.

Separate treatment of the case $q = 0$ yields

$$W_0^{(+)}(t;s) = 1_S - \sum_{\alpha\beta}\int_s^t\mathrm{d}u\int_s^u\mathrm{d}v c_{\alpha\beta}(u,v)V_\alpha(u)W_1^{(+)}(u;v;s)_\beta, \quad (19)$$

with $t > s$. From the solution for $W_q^{(+)}$ and the symmetry relation

$$\mathcal{T}_-\left\{\exp[K^{(-)}(t,s)]\prod_{i=1}^{q}V_{\alpha_i}(t_i)\right\} = \left[\mathcal{T}_+\left\{\exp[K^{(+)}(t,s)]\prod_{i=1}^{q}V_{\alpha_i}(t_i)^\dagger\right\}\right]^\dagger, \quad (20)$$

the solution for $W_q^{(-)}$ can be found. In taking the adjoint of $K^{(+)}$ one should employ the self-adjointness of H_1.

The set (18)–(20) generates an infinite hierarchy for the Kraus operators $\{W_q^{(\eta)}\}$. Once this hierarchy has been solved one can compute the system density operator. For that purpose the above-employed inequalities $t_1 > \cdots > t_q$ must be incorporated into the representation (11). This can be achieved through modification of integral boundaries.

First, we divide the integration domain $t > t_j > 0$, with $j = 1,\ldots,q$, into the parts $t > t_{P(1)} > \cdots > t_{P(q)} > 0$, where P denotes any permutation of the integers $\{1,\ldots,q\}$. Next, we carry out the transformation $t_{P(j)} = \tilde{t}_j$ and note that summing over all permutations P^{-1} is equivalent to summing over all permutations P. Of course, for all primed times in (11) we should act likewise. Then the density operator is obtained as

$$\rho_S(t) = \sum_{q=0}^{\infty}\sum_{\alpha_1\cdots\alpha_q}\sum_{\alpha'_1\cdots\alpha'_q}\int_0^t\mathrm{d}t_1\cdots\int_0^{t_{q-1}}\mathrm{d}t_q \times\int_0^t\mathrm{d}t'_1\cdots\int_0^{t'_{q-1}}\mathrm{d}t'_q\frac{1}{q!}\sum_{PQ}\prod_{k=1}^{q}\{Q(k)' \quad P(k)\} \times W_q^{(+)}(t;t_1,\cdots,t_q;0)_{\alpha_1\cdots\alpha_q}\rho_S W_q^{(-)}(t;t'_1,\cdots,t'_q;0)_{\alpha'_1\cdots\alpha'_q}. \quad (21)$$

We sum over all permutations P and Q of the integers $\{1,\ldots,q\}$. Now the solutions for $\{W_q^{(\eta)}\}$ as obtained from (18)–(20) may be substituted into (21).

By construction, the hierarchy (18)–(20) and representation (21) provide us with an exact Kraus map for non-Markovian quantum dynamics. Only one major assumption underlies our derivation, namely the factorization $\rho_{SR}(t=0) = \rho_S\otimes\rho_R$. In other words, the thermal reservoir is supposed to be initially decorrelated from the system.

In contrast to dissipative maps emanating from Nakajima-Zwanzig projection techniques or time-local approaches, Kraus maps *manifestly* conserve the positivity of the density operator. This is a most important property, as quantum states with negative probability are unphysical. Hence, the evolution equations (18)–(21) make up the natural starting-point for the description of a quantum dissipative system within the Hilbert space of that system.

To further illustrate the advantages of our formalism we focus on the asymptotic behavior of the system density operator. The simple result (13) will allow us to formulate a condition that guarantees convergence of $\rho_S(t)$ to the thermal state as t becomes large.

Asymptotic behavior. – In deriving an asymptotic limit we assume the system Hilbert space to be of finite dimension. For arbitrary system operators A, B and superoperator $\mathcal{O}$ we define a thermal scalar product and an auxiliary superoperator $\mathcal{O}^{(a)}$as [4]

$$\langle A,B\rangle = \mathrm{Tr}_S(\rho_{th}A^\dagger B), \quad (22)$$

$$\mathrm{Tr}_S[(\mathcal{O}A)B] = \mathrm{Tr}_S[A(\mathcal{O}^{(a)}B)]. \quad (23)$$

The thermal state is given by $\rho_{th} = \exp(-\beta H_S)/Z_S$, with $Z_S = \mathrm{Tr}_S[\exp(-\beta H_S)]$. Without loss of generality we assume that the system potentials $\{V_\alpha\}$ make up an orthonormal basis for the linear space of system operators, with respect to the scalar product (22).

The Dunford-Taylor representation [6] is an effective tool to analyze the behavior of (13) for large times. If A denotes an arbitrary system operator one has

$$\mathrm{Tr}_S\left\{\left[\mathrm{e}^{L(t)}\rho_S\right]A\right\} = \int_C\frac{\mathrm{d}z}{2\pi i}\mathrm{e}^{zt}\mathrm{Tr}_S\left\{\rho_S\left[z1_S - L^{(a)}(t)/t\right]^{-1}A\right\}. \quad (24)$$

The closed contour C contains all eigenvalues of $L^{(a)}(t)$.

From (12) and (23) one infers that the unit operator is eigenvector of $L^{(a)}(t)$ with eigenvalue equal to zero. We are going to determine a sufficient condition such that i) this eigenvalue is non-degenerate and ii) the real part of all other eigenvalues is negative.

We start by observing that the non-Markovian generator $L^{(a)}(t)$ is continuous in t, and that (16) guarantees the existence of $L^{(a)}(t)/t$ for $t\to\infty$. Hence, in finite dimensions the asymptotic dynamics is determined by the time-independent operator $L_\infty^{(a)} = \lim_{t\to\infty}L^{(a)}(t)/t$.

Invoking (12) and (23) once more, we derive $L_\infty^{(a)} = L_1^{(a)} + L_2^{(a)} + L_3^{(a)}$. Here one must employ the definitions of L_1, L_2, and L_3 that were given earlier.

One verifies that the superoperators $iL_1^{(a)}$, $L_2^{(a)}$, and $L_3^{(a)}$ are self-adjoint with respect to the scalar product (22). The KMS property $c_{\alpha'\alpha}(t' - i\beta, t) = c_{\alpha\alpha'}(t, t')$ and the identity $[K, \rho_{th}] = 0$ are needed. As argued below, one may assume that the condition

$$\left[L_1^{(a)}, L_2^{(a)} + L_3^{(a)}\right] = 0 \tag{25}$$

is satisfied. Then all eigenvalues of $L_\infty^{(a)}$ can be expressed as $\lambda + i\mu$, with λ, μ real and λ eigenvalue of $L_2^{(a)} + L_3^{(a)}$. Moreover, the eigenvectors $\{F_\alpha\}$ of $L_\infty^{(a)}$ provide us with a basis for the linear space of system operators. After performing an orthonormalization, one thus may utilize the expansion $A = \sum_\alpha \langle F_\alpha, A\rangle F_\alpha$.

To locate λ we employ the KMS property once more so as to arrive at

$$\langle A, (L_2^{(a)} + L_3^{(a)})A\rangle = -\tfrac{1}{2}\sum_{\alpha\beta} d_{\alpha\beta}\langle [V_\alpha, A], [V_\beta, A]\rangle. \tag{26}$$

The damping matrix $d_{\alpha\beta}$ reads

$$d_{\alpha\beta} = \lim_{T\to\infty} T^{-1}\sum_{\gamma\delta}\int_0^T \mathrm{d}t \int_{-\infty}^{\infty} \mathrm{d}s\, c_{\gamma\delta}(s, 0) \times \langle V_\gamma^\dagger(t + s), V_\alpha\rangle\langle V_\beta, V_\delta(t)\rangle. \tag{27}$$

By expanding the pair correlation function with the help of reservoir eigenstates and employing Bochner's theorem [8] we demonstrate that the damping matrix is non-negative. Zero eigenvalues might occur, for instance, if some reservoir potentials U_α vanish. Then parts of the system do not interact with the reservoir. Excluding such ill-fated cases, we limit our attention to interaction Hamiltonians H_1 for which $d_{\alpha\beta}$ is strictly positive.

Identity (26) tells us that if an eigenvector F_β has a vanishing eigenvalue, the commutator $[V_\alpha, F_\beta]$ must be vanishing as well, with α arbitrary. Consequently, F_β must be equal to eigenvector 1_S. This proves statement i) mentioned above. In order to verify statement ii), we observe that (26) imposes the constraint $\lambda < 0$ on all eigenvalues belonging to eigenvectors $F_\alpha \neq 1_S$.

Now we can return to (24). On the right-hand side we expand A with the help of the orthonormal basis $\{F_\alpha\}$. On account of statements i) and ii), only the term $\langle 1_S, A\rangle 1_S$ contributes for t large. Next, the eigenvalue equation $L^{(a)}(t)1_S = 0$ can be exploited in the Dunford-Taylor integral. It then reduces to the residue of a simple pole at $z = 0$.

We now have shown that for large t the right-hand side of (24) converges to $\langle 1_S, A\rangle$. As A is arbitrary, we arrive at the asymptotic limit

$$\lim_{t\to\infty} \rho_S(t) = \rho_{th}. \tag{28}$$

We have acted with $\mathcal{T}$ on both sides of (28). On the right we have employed the identity $\mathcal{T}\rho_S = \rho_S$ appearing near (13), with $\rho_S = \rho_{th}$. On the left we have interchanged $\mathcal{T}$ and the limit $t \to \infty$. To justify this we observe that by taking t large we do not modify the ordering in (21).

The main condition accompanying the limit (28) appears in (25). One checks that (25) is true for a non-degenerate n-level atom. Diagonal reservoir potentials $U_{(kk)}$, with k labelling the atomic energy levels, must vanish.

Perturbation theory. – The Nakajima-Zwanzig projection method cannot be used for setting up a non-Markovian perturbation theory. The reason is that the perturbative Nakajima-Zwanzig expansion is constructed from repeated commutators. As a consequence, eigenvalues of the perturbed density operator may become negative in the course of time [9]. Such mishap does not occur within the framework of the Kraus representation.

In developing a physically sound perturbation theory we let us be guided by the behavior of the hierarchy (18)–(19) in the limit $t \to \infty$. Then all contributions of (18) with $q + 1 \geq j \geq 2$ no longer exist. To prove this, we interchange the integrals over u and v, perform the shift $u \to u + v$, and scale times as $t = \tau\tilde{t}$, $v = \tau\tilde{v}$, $s = \tau\tilde{s}$, $t_i = \tau\tilde{t}_i$, with $i = 1, \ldots, q$. In view of (21) the inequalities $\tilde{t} > \tilde{t}_1 > \cdots > \tilde{t}_q > \tilde{s}$ are valid. These ensure that for large τ all contributions under consideration behave as $\tau \sup_{\alpha\beta}\int_\tau^\infty \mathrm{d}u|c_{\alpha\beta}(u, 0)|$. In virtue of (16) the latter form tends to zero.

The above argument indicates that in the asymptotic regime all contributions of (18) with $q + 1 \geq j \geq 2$ may be discarded. For $t_0 > t_1 > \cdots > t_q > s$ and $q = 1, 2, \ldots$ the solution of the ensuing hierarchy reads

$$\tilde{W}_q^{(+)}(t_0; t_1, \cdots, t_q; s)_{\alpha_1\cdots\alpha_q} = \mathcal{T}_+\left[\prod_{j=1}^{q} \tilde{W}_0^{(+)}(t_{j-1}; t_j)V_{\alpha_j}(t_j)\right]\tilde{W}_0^{(+)}(t_q; s). \tag{29}$$

The identity

$$\tilde{W}_0^{(+)}(t; s) = 1_S - \sum_{\alpha\beta}\int_s^t \mathrm{d}u \int_s^u \mathrm{d}v \times c_{\alpha\beta}(u, v)V_\alpha(u)\tilde{W}_0^{(+)}(u; v)V_\beta(v)\tilde{W}_0^{(+)}(v; s) \tag{30}$$

determines the asymptotic zeroth-order Kraus operator.

In (21) only terms with $P(k) = Q(k), k = 1, \ldots, q$ survive for large times. Therefore, the asymptotic dynamics is described by $\tilde{\rho}_S(t) = \xi(t, t)$ and

$$\xi(t, t') = \tilde{W}_0^{(+)}(t; 0)\rho_S\tilde{W}_0^{(-)}(t'; 0) + \sum_{\alpha\alpha'}\int_0^t \mathrm{d}u \int_0^{t'} \mathrm{d}u' \times c_{\alpha'\alpha}(u', u)\tilde{W}_0^{(+)}(t; u)V_\alpha(u)\xi(u, u')V_{\alpha'}(u')\tilde{W}_0^{(-)}(t'; u'). \tag{31}$$

We now assess the merits of (30) and (31) for finite t.

Computation of the derivative $\mathrm{d}\mathrm{Tr}[\xi(t, t)]/\mathrm{d}t$ yields zero, so the trace of $\tilde{\rho}_S(t)$ is conserved in time. Furthermore, by inspection we verify that at all times t the perturbative

dynamics has the form of a Kraus map. Thus, positivity of $\tilde{\rho}_S(t)$ is conserved as well. Last, since the norm $\|\tilde{\rho}_S(t) - \rho_S(t)\|$ will vanish for $t \to \infty$, $\tilde{\rho}_S(t)$ inherits the asymptotic behavior of $\rho_S(t)$.

Altogether, the closed evolution equations (30) and (31) offer a perturbative description that is physically acceptable. Indeed, in previous work [2] it was found that the approximate density operator $\tilde{\rho}_S(t)$ furnishes an accurate description of both Rabi oscillations and Bloch relaxation of a two-level atom. As the system Hamiltonian may be freely chosen, our perturbative description is applicable not only to quantum optics, but also to quantum computing, spin dynamics, and chemical reactions.

Conclusion and outlook. – Any quantum system that exchanges energy with a thermal reservoir displays non-Markovian evolution in time. The evolution is governed by a Kraus map [1]. Employing a symmetrized form of Wick's theorem and benefiting from the use of time-ordering operators, we have succeeded in expressing the Kraus map in terms of system potentials and reservoir pair correlation functions. In the limit $t \to \infty$ the Kraus map takes on a factorized form that is well known from previous work [2]. Condition (25) guarantees return to thermal equilibrium.

As the Hamiltonian H_S has not been specified, our theory can be applied to all quantum systems with a separable Hilbert space. The only major assumption we have made is factorization of the initial state of system and reservoir. Obviously, for initial states given by $\sum_j \rho_{S,j} \otimes \rho_{R,j}$, with $\rho_{R,j}$ a thermal state of inverse temperature β_j, extension of our treatment is immediate. The same remark holds for a time-dependent system Hamiltonian. In (3) the series $\mathcal{T}_+ \exp[i \int_0^t \mathrm{d}u H_S(u)]$ and its Hermitian conjugate enter. Nonlinear interaction between system and reservoir renders the factorization scheme (5) invalid and falls outside the scope of this work therefore.

Our Kraus compliant theory has several advantages over traditional approaches that are rooted in master equations for the density operator. First, as testified by (21), states with negative probability are manifestly excluded at all times. In other words, the expansion (21) and the underlying Kraus hierarchy (18)–(19) constitute the natural representation of a quantum dissipative evolution. We dispense with the awkward projectors P and $Q = 1 - P$ of the old Nakajima-Zwanzig method. Also, we avoid the use of the inverse propagators figuring in time-local approaches. Such propagators may become divergent for large times [10].

Second, the concise result (13) provides us with the non-Markovian counterpart $L(t)$ of the standard Markovian generator L for quantum dissipative dynamics. Differentiation of (13) makes clear that the extension of the Markovian master equation $\mathrm{d}\rho_S(t)/\mathrm{d}t = L\rho_S(t)$ to the case of non-Markovian dynamics confronts us with a most complicated time-ordering problem. We believe therefore that in deriving quantum dissipative evolution equations one should focus on the Kraus operators instead of the system density operator itself.

Third, our findings enable us to set up a non-Markovian perturbation theory that conserves both positivity and trace of the density operator. This theory is based on the following truncation prescription for the Kraus hierarchy:

$$W_N^{(+)}(t; t_1, \cdots, t_N; s)_{\alpha_1 \ldots \alpha_N} = W_0^{(+)}(t; t_1) V_{\alpha_1}(t_1) \times W_{N-1}^{(+)}(t_1; t_2, \cdots, t_N; s)_{\alpha_2 \ldots \alpha_N}, \tag{32}$$

with $t > t_1 > \cdots > t_N > s$. The truncation parameter N is a fixed integer ranging from 1 to infinity. It determines the order of the perturbation theory. Since (29) satisfies (32), the exact asymptotic dynamics is reproduced for all values of N. The case of $N = 1$ corresponds to the dynamics (30), (31). The case of arbitrary N will be elaborated in a forthcoming paper.

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