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Generic Construction of Kraus Operators: *d*-level Systems in a Thermal Bosonic Bath

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Abstract: The dynamical evolution of open systems can be conveniently expressed in the operator sum representation in terms of Kraus operators. Here we introduce an expression for the canonical Kraus representation in terms of the eigenstates $|\gamma\rangle$ and eigenvalues E_γ of the total Hamiltonian, thus making a stronger connection to the tools in chemistry. The method is exact, and hence valid for non-Markovian evolution of the system to all orders of the coupling to the bath. Significantly, it provides a starting point for developing approximate Kraus operators based upon perturbative approximations to $|\gamma\rangle$ and E_γ. As an example, the explicit form of these operators is derived for a *d*-level system interacting with a single-mode thermal bath.

Keywords: decoherence · energy transfer · molecular dynamics · quantum chemistry · spin–boson systems

1. Introduction

Control and evolution in the dynamics of open systems is a challenge at the forefront of modern chemical physics.[1–3] In quantum mechanics, the evolution of an isolated system is described by the effect of a unitary evolution operator on the system density matrix. If, however the system of interest interacts with the surrounding environment (that is, it is "open") its time evolution cannot be described by a unitary transformation of the initial state. Such non-unitary system evolution is often treated approximately, for example via the Lindblad equation[4] in the limit of weak coupling with the environment and within the Markovian approximation. However, the evolution can be treated exactly up to all orders of coupling by the following map in the operator sum representation:[5,6]

$$\rho_S(t) = \sum_i E_i \rho_S(0) E_i^\dagger. \tag{1}$$

Here $\rho_S(t)$ is the system density matrix at time t, and the operator elements E_i are called Kraus operators[5,6] and satisfy the completeness relation $\sum_i E_i^\dagger E_i = \hat{\mathbf{1}}$. For the non-trace-preserving quantum operations (viz., measurements), the Kraus operators follow the more general completeness relation $\sum_i E_i^\dagger E_i \leq \hat{\mathbf{1}}$. Thus, the Kraus operators appear to be useful in representing a general class of evolution in an elegant form. In particular, it has been shown that for a d-dimensional system the non-unitary system evolution can be described by at most d^2 Kraus operators,[7] giving the so-called canonical Kraus representation.

The Kraus operators have been calculated for a variety of cases, viz., a single-mode harmonic oscillator interacting with a single-mode bosonic bath[6,8] and a multi-mode bosonic bath.[9] The bit-flip and the phase-flip operations for a single spin have been described in terms of Kraus operators in the context of quantum error correction.[6] In ref. [10], the relationship of the Lindblad equation and the Kraus representation has been identified. The solution of the Lindblad equation has also been written in terms of Kraus operators for a two-level system[6,11] and a three-level system.[11] Further, the Kraus operators for any arbitrary time-dependent density matrix can be expressed as a function of instantaneous eigenvalues of the density matrices.[12] However, there is surprisingly little available on a general method to construct the canonical Kraus operators when given the Hamiltonian for the system interacting with the environment. This is of particular interest in chemical physics, where the Hamiltonian provides the connection to physical intuition.

In this paper we introduce a general way of formulating the canonical Kraus operators for any system in terms of the eigenstates and eigenvalues of the underlying Hamiltonian. The resultant Kraus representation is exact and hence includes non-Markovian dynamics to all orders of the coupling constants. This construction serves a number of useful purposes. First, it is completely general, that is,

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applicable to any system-bath case. Second, it is designed to serve as a launching point for approximation methods based, for example, on perturbation expansions. Third, we expect that the resultant final form will prove useful as a starting expression to allow the introduction of external fields, an important challenge for the study of the control of open quantum systems.[3,13]

The approach below is completely general. However, for simplicity we introduce it via an example, the interaction of a single spin with an environment comprising a single-mode harmonic oscillator. This interaction includes both energy exchange with the environment and pure dephasing. Note that only approximate Kraus operators for this type of system have been previously obtained. For example, non-Markovian dissipation of a spin in a bosonic bath has previously been found up to second order in the coupling.[14] Similarly, the interaction of two spins with a single-mode thermal bath has been described in terms of Kraus operators for the Hamiltonian under rotating wave approximation.[15] By contrast, we focus on the most general Hamiltonian of the spin–boson interaction. We also extend our analysis to a d-dimensional spin system.

2. The Spin–Boson Hamiltonian

2.1. General Case

Consider a single spin interacting with a single mode thermal bosonic bath. The corresponding Hamiltonian is given by

$$\begin{aligned} H/\hbar &= \omega_0 S_z + \omega a^\dagger a + g_{ph} S_z\left(a + a^\dagger\right) \\ &+ g_r\left(S_+ a + S_- a^\dagger\right) + g_{nr}\left(S_+ a^\dagger + S_- a\right), \end{aligned} \tag{2}$$

where ω_0 and ω are the fundamental frequencies of the spin and the bath, respectively. The spin operators are $S_\pm = |\pm\rangle\langle\mp|$ and $S_z = (|+\rangle\langle+| - |-\rangle\langle-|)$, where $|\pm\rangle$ are the spin states. The g_r term in the Hamiltonian represents interaction between the spin and the bosonic bath that would be retained under the rotating wave approximation (RWA), whereas the g_{nr} term represents the interaction that would otherwise have been neglected under RWA. The g_{ph} term describes the dephasing of the spin. Eigenstates of the Hamiltonian in the absence of any couplings are termed "bare states" and are a product of the spin $|\pm\rangle$ and bath $|n\rangle$ eigenstates, which we denote as $|\pm,n\rangle$.

We assume that the bath at the initial time is in thermal equilibrium at temperature T with bath density matrix given by $\rho_B(0) = \Sigma_n p_n |n\rangle\langle n|$, where $p_n = \exp(-n\hbar\omega/k_B T)/Z$ and $Z = \Sigma_n \exp(-n\hbar\omega/k_B T)$. The evolution of the spin + bath system is described by

$$\rho_{SB}(t) = V_{SB}\left[\rho_S(0) \otimes \rho_B(0) V_{SB}^\dagger\right], \tag{3}$$

where $V_{SB} = e^{-iHt/\hbar}$. The reduced density matrix of the spin can then be written as

$$\rho_S(t) = \sum E_{mn} \rho_S(0) E_{mn}^\dagger, \tag{4}$$

where $E_{mn} = \sqrt{p_n}\langle m|V_{SB}|n\rangle$. We next expand the operator $V_{SB} = \exp(-iHt/\hbar)$ in terms of the eigenstates $|\gamma\rangle$ and eigenvalues E_γ of the Hamiltonian H. The operators E_{mn} can then be expressed as

$$E_{mn} = \sum_\gamma e^{-iE_\gamma t/\hbar} \langle m|\gamma\rangle \langle\gamma|n\rangle \sqrt{p_n}. \tag{5}$$

where E_γ are the eigenvalues of H, corresponding to the eigenstate $|\gamma\rangle$. Note that in Equation (5), the term $\langle m|\gamma\rangle\langle\gamma|n\rangle$ represents an operator in spin Hilbert space. We emphasize that the expression (4) is not the desired canonical Kraus representation for the evolution of the spin. Here there are infinite numbers of the operators E_{mn}, whereas the maximum number of Kraus operators in this case should be $2^2 = 4$.

The eigenstates $|\gamma\rangle$ can be written in terms of the bare states $|\pm,n\rangle$ of the combined Hilbert space of the spin and the bath. The overlap between these bare states and the eigenstates $|\gamma\rangle$ is denoted $C_{\pm,n}{}^\gamma = \langle\pm,n|\gamma\rangle$. Using these overlaps, the operators E_{mn} can be rewritten as

$$E_{mn} = I_{mn}\hat{\mathbf{1}} + Z_{mn} S_z + X_{mn} S_x + Y_{mn} S_y, \tag{6}$$

where

$$\begin{aligned} I_{mn} &= \frac{1}{2}\sum_\gamma e^{-iE_\gamma t/\hbar}\sqrt{p_n}\left(C_{+m}^\gamma C_{+n}^{\gamma *} + C_{-m}^\gamma C_{-n}^{\gamma *}\right) \\ Z_{mn} &= \frac{1}{2}\sum_\gamma e^{-iE_\gamma t/\hbar}\sqrt{p_n}\left(C_{+m}^\gamma C_{+n}^{\gamma *} - C_{-m}^\gamma C_{-n}^{\gamma *}\right) \\ X_{mn} &= \frac{1}{2}\sum_\gamma e^{-iE_\gamma t/\hbar}\sqrt{p_n}\left(C_{+m}^\gamma C_{-n}^{\gamma *} + C_{-m}^\gamma C_{+n}^{\gamma *}\right) \\ Y_{mn} &= \frac{i}{2}\sum_\gamma e^{-iE_\gamma t/\hbar}\sqrt{p_n}\left(C_{+m}^\gamma C_{-n}^{\gamma *} - C_{-m}^\gamma C_{+n}^{\gamma *}\right). \end{aligned} \tag{7}$$

Using Equation (6) in Equation (4), the time-dependence of the density matrix ρ_S can be expressed as

$$\rho_S(t) = \sum_{k,l=x,y,z,1} \theta_{kl} S_k \rho_S(0) S_l^\dagger, \tag{8}$$

where $S_1 = \hat{\mathbf{1}}$. Here the coefficients are

$$\theta_{kl} = \sum_{m,n} K_{mn} L^*_{mn}, \quad K, L = X, Y, Z, I. \tag{9}$$

It is easy to see that $\theta_{kl} = \theta^*_{lk}$, and therefore that $\boldsymbol{\theta}$ is an Hermitian matrix.

Equation (8) is given as a sum over a finite number of terms, unlike Equation (4). However, it still does not represent the canonical Kraus representation of the spin-boson problem because of the contribution of $k \neq l$ cross terms in Equation (8).

To find the canonical Kraus representation, we rewrite Equation (8) in terms of a row matrix $\mathbf{A} = [S_x, S_y, S_z, \hat{\mathbf{1}}]$ as

$$\rho_S(t) = \mathbf{A}\rho_S(0)\theta\mathbf{A}^\dagger = \mathbf{A}\rho_S(0)\mathbf{U}\mathbf{U}^\dagger\theta\mathbf{U}\mathbf{U}^\dagger\mathbf{A}^\dagger \tag{10}$$

where the unitary matrix $\mathbf{U}$ diagonalizes the matrix $\boldsymbol{\theta}$ as follows:

$$\mathbf{U}^\dagger\theta\mathbf{U} = \mathrm{diag}(d_{11}\, d_{22}\, d_{33}\, d_{44}), \tag{11}$$

where the d_{ii} are real since $\boldsymbol{\theta}$ is a Hermitian matrix. The ith column $(U_{ix}, U_{iy}, U_{iz}, U_{i1})$ of the matrix U is the ith eigenvector of the matrix $\boldsymbol{\theta}$, corresponding to the eigenvalue d_{ii}. Then $\rho_S(t)$ can be rewritten in the following canonical Kraus form:

$$\rho_S(t) = \sum_{i=1}^{4} O_i \rho_S(0) O_i^\dagger \tag{12}$$

where

$$O_i = \sqrt{d_{ii}}\left(U_{ix}S_x + U_{iy}S_y + U_{iz}S_z + U_{i1}\hat{\mathbf{1}}\right). \tag{13}$$

Further, we show below that the O_i and $O_i^\dagger$ satisfy, as required, the requisite sum rule. Hence, the $O_i, O_i^\dagger$ are the desired Kraus operators, and are defined via the eigenstates of $\boldsymbol{\theta}$.

We could, alternatively, find the Kraus operators without knowing its eigenspace by using the Cholesky decomposition of the Hermitian positive definite matrix $\boldsymbol{\theta} = \mathbf{M}\mathbf{M}^\dagger$, where the matrix $\mathbf{M}$ is strictly a lower triangular matrix with positive diagonal elements. This suggests a different form of the Kraus operators, other than that in Equation (13). Clearly, the Kraus operators are not unique.

Note that the O_i in (13) are not necessarily Hermitian (i.e., $O_i \neq O_i^\dagger$), because the elements U_{ix}, U_{iy} etc. are, in general, complex. Further, it is easy to see that $O_i^\dagger O_i \neq O_i O_i^\dagger$. The equality $O_i^\dagger O_i = O_i O_i^\dagger$ would hold if O_i would be Hermitian (or strictly diagonal, as a special case), which is not the case in a non-unitary evolution.

We next confirm that the operators O_i in (13) indeed satisfy the completeness relation $\Sigma_i O_i^\dagger O_i = \hat{\mathbf{1}}$. To do this, first take the trace on both sides of Equation (10) such that

$$Tr[\rho_S(t)] = Tr[\mathbf{A}\rho_S(0)\theta\mathbf{A}^\dagger] = Tr[\theta\mathbf{A}^\dagger\mathbf{A}\rho_S(0)]. \tag{14}$$

Since the evolution of a density matrix is a trace-preserving map $[Tr(\rho_S(t)) = 1\ \forall t]$, we have from Equation (14)

$$\begin{aligned} &\mathrm{Tr}[\boldsymbol{\theta}] + \left[2\mathrm{Re}(\theta_{x1}) + 2\mathrm{Im}(\theta_{yz})\right]\langle S_x\rangle_{t=0} \\ &+\left[2\mathrm{Re}(\theta_{y1}) + 2\mathrm{Im}(\theta_{zx})\right]\langle S_y\rangle_{t=0} \\ &+\left[2\mathrm{Re}(\theta_{z1}) + 2\mathrm{Im}(\theta_{xy})\right]\langle S_z\rangle_{t=0} = 1, \end{aligned} \tag{15}$$

where $\langle S_i\rangle_{t=0} = Tr[S_i\rho_S(0)]$. Because $Tr[\rho_S(t)] = 1$ must be satisfied irrespective of the initial conditions, the coefficients of the expectation values in the above expression must vanish, leading to $Tr[\boldsymbol{\theta}] = 1$. Further, from Equation (11), expanding $\mathbf{U}$ and $\boldsymbol{\theta}$, we find the following:

$$\theta_{\alpha\beta} = \sum_{i=1}^{4} d_{ii} U_{i\alpha} U^*_{i\beta}, \quad \alpha, \beta = x, y, z, 1. \tag{16}$$

Using (13), we can write

$$\begin{aligned} \sum_i O_i^\dagger O_i = \sum_i d_{ii}\Big[&\left\{|U_{ix}|^2 + |U_{iy}|^2 + |U_{iz}|^2 + |U_{i1}|^2\right\}\hat{\mathbf{1}} \\ &+2\left\{\mathrm{Re}\left(U_{ix}U^*_{i1}\right) + \mathrm{Im}\left(U_{iy}U^*_{iz}\right)\right\}S_x \\ &+2\left\{\mathrm{Re}\left(U_{iy}U^*_{i1}\right) + \mathrm{Im}\left(U_{iz}U^*_{ix}\right)\right\}S_y \\ &+2\left\{\mathrm{Re}\left(U_{iz}U^*_{i1}\right) + \mathrm{Im}\left(U_{ix}U^*_{iy}\right)\right\}S_z\Big]. \end{aligned} \tag{17}$$

Given Equation (16), we find that the coefficients of S_x, S_y, and S_z in (17) are the same as those of the initial expectation values $\langle S_i\rangle_{t=0}$ in Equation (15), which must vanish. Thus,

$$\sum_i O_i^\dagger O_i = \sum_i d_{ii}\Big[\left\{|U_{ix}|^2 + |U_{iy}|^2 + |U_{iz}|^2 + |U_{i1}|^2\right\}\hat{\mathbf{1}}. \tag{18}$$

Because, the columns of the matrix U represent normalized eigenvectors of the matrix θ and $Tr[\boldsymbol{\theta}] = \Sigma_i d_{ii} = 1$ (as the unitary transformation does not change the trace of a matrix), we have the completeness relation $\sum_i O_i^\dagger O_i = \hat{\mathbf{1}}$.

The above approach provides a direct route between the Hamiltonian and the Kraus operator representation. For completeness, we note that, as shown in ref. [16] if Equation (4) could be written as $\rho_S(t) = \mathbf{B}\rho_S(0)$, where $\mathbf{B}$ is a Hermitian matrix, then an operator sum representation of this evolution could be found in terms of the eigenvalues and eigenvectors of $\mathbf{B}$. Their work is complementary to that introduced here. For example, their ap-

proach is not Hamiltonian-based but is applicable to initially entangled open systems whereas our approach takes advantage of the Hamiltonian expressions and can also be applied to Measurements.

2.2. Special Cases

We can now obtain the Kraus operators for some special cases of the Hamiltonian (2).

Consider first the case $g_{ph}=0$ and $g_r=g_{nr}=g$. This represents a spin–boson interaction with no rotating wave approximation. A similar Hamiltonian can be found in systems like a Cooper pair box interacting with a nanomechanical oscillator.[17] It is easy to see that this Hamiltonian does not couple the states $|\pm,n\rangle$ for a given n. Thus we have from Equation (7), $X_{nn}=Y_{nn}=0$ and the matrix $\boldsymbol{\theta}$ is of the form

$$\boldsymbol{\theta}=\begin{pmatrix}\theta_{xx} & \theta_{xy} & 0 & 0\\ \theta_{yx} & \theta_{yy} & 0 & 0\\ 0 & 0 & \theta_{zz} & \theta_{z1}\\ 0 & 0 & \theta_{1z} & \theta_{11}\end{pmatrix}. \tag{19}$$

Note that θ_{zz} is obtained from the terms Z_{mn}, which represent the dephasing due to energy exchange between the spin and the bath. The spin does not undergo any pure dephasing since $g_{ph}=0$. The above matrix is in block form with two 2×2 submatrices. It is straightforward to find the eigenstates of each block of $\boldsymbol{\theta}$, elements of which are given by

$$\begin{aligned}&U_{ix}=\theta_{yy}/N_i\,,\quad U_{iy}=-(\theta_{xx}-\lambda_i)/N_i\,,\quad i=1,2\,;\\ &N_i=\sqrt{\left[(\theta_{xx}-\lambda_i)^2+\theta_{yy}\right]^2}\,,\\ &\lambda_{1,2}=\frac{1}{2}\left[\left(\theta_{xx}+\theta_{yy}\right)\pm\sqrt{\left(\theta_{xx}-\theta_{yy}\right)^2+4\left|\theta_{xy}\right|^2}\right].\end{aligned} \tag{20}$$

and

$$\begin{aligned}&U_{iz}=\theta_{11}/N_i\,,\quad U_{i1}=-(\theta_{zz}-\lambda_i)/N_i\,,\quad i=3,4\,;\\ &N_i=\sqrt{\left[(\theta_{zz}-\lambda_i)^2+\theta_{11}\right]^2}\,,\\ &\lambda_{3,4}=\frac{1}{2}\left[(\theta_{zz}+\theta_{11})\pm\sqrt{(\theta_{zz}-\theta_{11})^2+4|\theta_{z1}|^2}\right].\end{aligned} \tag{21}$$

The Kraus operators thus become $O_i=U_{ix}S_x+U_{iy}S_y$ $(i=1,2)$ and $O_i=U_{iz}S_z+U_{i1}\hat{\mathbf{1}}$ $(i=3,4)$, respectively. Similar conclusions can be drawn for the Hamiltonian within the rotating wave approximation, in other words, when only $g_r\neq0$ in the interaction part of the Hamiltonian.

As a second example, consider a case when $g_r=g_{nr}=0$, $g_{ph}\neq0$, a case of pure spin dephasing. The states $|+,m\rangle$ and $|-,n\rangle$ are not coupled, leading to $X_{mn}=Y_{mn}=0,\forall m,n$ (note that pure dephasing does not lead to any energy exchange between the spin and the bath). The matrix $\boldsymbol{\theta}$ is reduced to a 2×2 matrix in the (S_z,S_1) basis. The Kraus operators become $O_i=U_{iz}S_z+U_{i1}\hat{\mathbf{1}}$ $(i=3,4)$.

3. *d*-level Systems in a Thermal Bath

Thus far we have considered a two-level system (a qubit), which would be relevant in, for example, quantum computing. However, most physical systems of interest contain more than two levels. In this context, Kraus operators for destruction of states in a qudit has been derived.[18] Further, Markovian dynamics of a qudit in a thermal bath has been described in terms of Kraus operators.[19] Our technique of deriving Kraus operators, however, does not assume any such limitations or approximations.

Consider then a d-level system interacting with a single-mode thermal bath. The time-evolved density matrix of the system can be expressed in the same form (4), where $E_{mn}=\sum_\gamma e^{-iE_\gamma t/\hbar}\sqrt{p_n}\langle m|\gamma\rangle\,\langle\gamma|n\rangle$. Rewriting in terms of overlaps $C^\gamma_{im}=\langle i,m|\gamma\rangle$, we have

$$E_{mn}=\sum_\gamma\sum_{j,k=1}^{d}e^{-iE_\gamma t/\hbar}\sqrt{p_n}C^\gamma_{jm}C^{\gamma *}_{kn}|j\rangle\,\langle k|. \tag{22}$$

The d-level system can be represented in many convenient bases, e.g., the generalized Gell-Mann basis,[20] the polarization basis,[21] or the Weyl operator basis.[22] (A brief review of these representations can be found in ref. [23]). We choose to use the generalized Gell–Mann basis, which is an extension of the usual Pauli spin matrices. This is defined by a set of d^2-1 Hermitian and traceless operators as follows: $d(d-1)/2$ symmetric matrices $\Lambda^{jk}_s=|j\rangle\langle k|+h.c.$, $d(d-1)/2$ antisymmetric matrices $\Lambda^{jk}_a=-i|j\rangle\langle k|+h.c.$, $(1\le j<k\le d)$ and $(d-1)$ diagonal matrices

$$\wedge^{jj}_{dd}=\sqrt{\frac{2}{j(j+1)}}\left(\sum_{l=1}^{j}|l\rangle\,\langle l|-j|j+1\rangle\langle j+1|\right),\quad 1\le j\le d-1\,. \tag{23}$$

Note that the above operators are the $SU(d)$ generators and form an orthogonal basis of the operators. We can rewrite E_{mn} in terms of these operators as

$$\begin{aligned}E_{mn}=&\sum_{k,j<k}\left[\phi^{jk}_{mn,s}\Lambda^{jk}_s+\phi^{jk}_{mn,\alpha}\Lambda^{jk}_\alpha\right]\\ &+\sum\phi^{jj}_{mn,d}\Lambda^{jj}_d+\phi_{mn,0}\hat{\mathbf{1}},\end{aligned} \tag{24}$$

where $\phi^{jk}_{mn,\alpha}$ are the coefficients of the Gell-Mann operator Λ^{jk}_α $(\alpha\in s,a,d)$ and $\phi_{mn,0}$ are the coefficients of the identity operator $\hat{\mathbf{1}}$, respectively. These coefficients con-

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tain the sum over all the eigenstates $|\gamma\rangle$, the time-dependent and the temperature-dependent terms. That is,

$$\phi^{jk}_{mn,s} = \frac{1}{2}\sum_{\gamma} e^{-iE_\gamma t/\hbar}\sqrt{p_n}\left(C^{\gamma}_{jm}C^{\gamma *}_{kn} + C^{\gamma}_{km}C^{\gamma *}_{jn}\right)$$

$$\phi^{jk}_{mn,\alpha} = \frac{i}{2}\sum_{\gamma} e^{-iE_\gamma t/\hbar}\sqrt{p_n}\left(C^{\gamma}_{jm}C^{\gamma *}_{kn} - C^{\gamma}_{km}C^{\gamma *}_{jn}\right)$$

$$\phi^{jj}_{mn,d} = \sum_{\gamma} e^{-iE_\gamma t/\hbar}\left[-\sqrt{\frac{j}{2(j+1)}}C^{\gamma}_{(j+1)m}C^{\gamma *}_{(j+1)n} + \frac{1}{\sqrt{2j(j+1)}}\sum_{r=1}^{j} C^{\gamma}_{rm}C^{\gamma *}_{rn}\right], \quad j = 1, \cdots, d-1 \tag{25}$$

$$\phi_{mn,0} = \sum_{\gamma}\sum_{j=1}^{d} e^{-iE_\gamma t/\hbar}\sqrt{p_n}C^{\gamma}_{jm}C^{\gamma *}_{jn} \quad .$$

Note that there are d^2 independent operators in Equation (24), including the identity operator $\hat{\mathbf{1}}$. Using Equation (24), we can now write the time-dependent matrix in the following form:

$$\rho_S(t) = \sum_{\alpha,\beta\in s,a,d,0}\sum_{\{j,k\},\{j',k'\}} \theta^{jk,j'k'}_{\alpha,\beta}\Lambda^{jk}_{\alpha}\rho_S(0)\Lambda^{j'k'}_{\beta}, \tag{26}$$

where $\alpha,\beta = 0$ refers to the identity operator $\hat{\mathbf{1}}$ for all (j,k), whereas for $\alpha,\beta = d$, i.e., for diagonal matrices, $j = k$. Here $\boldsymbol{\theta}$ is a $d^2\times d^2$-dimensional Hermitian matrix and its elements have the form

$$\theta^{jk,j'k'}_{\alpha\beta} = \sum_{m,n}\phi^{jk}_{mn,\alpha}\left(\phi^{j'k'}_{mn,\beta}\right)^{*}. \tag{27}$$

As in the case of a qubit, the Kraus operators can now be derived in terms of the eigenstates of the matrix $\boldsymbol{\theta}$. For example, the ith eigenstate is given by the ith column of the unitary matrix $\mathbf{U}$ that diagonalizes $\boldsymbol{\theta}$ and can thus be written as $[U_{i,s,\{jk\}},U_{i,a,\{jk\}},U_{i,d,\{jj\}},U_{i,0}]$ (with all the possible combinations of k and $j<k$). Thus the ith Kraus operator becomes $O_i = \sqrt{d_{ii}}N_i$, where d_{ii} is the ith eigenvalue of $\boldsymbol{\theta}$ and

$$N_i = \sum_{k,j<k}\left(U_{i,s,jk}\Lambda^{jk}_{s} + U_{i,a,jk}\Lambda^{jk}_{a}\right) + \sum U_{i,d,jj}\Lambda^{jj}_{d} + U_{i,0}\hat{\mathbf{1}}. \tag{28}$$

The d-dimensional system would undergo an evolution through the operator N_i with a probability that is given by the ith eigenvalue of the matrix $\boldsymbol{\theta}$.

4. Summary

In summary, we have presented a general method of expressing, from first principles, the canonical Kraus operators for the evolution of any time-independent d-dimensional system coupled to an environment. As required for the Kraus representation, the initial state is a separable product of the system and bath density matrices. Specifically: (i) the reduced density matrix of the system is determined by tracing over the bath [Equation (4)]; (ii) the time evolution of the operators E_{mn} so generated is written in terms of the eigenstates and eigenvalues of the total Hamiltonian; (iii) the E_{mn}, which operate on the system space, are expanded in a complete set of d^2 operators; and (iv) the resultant Hermitian coefficient matrix $\boldsymbol{\theta}$ [for example, Equation (9)] is diagonalized to yield the canonical Kraus form. Being exact, the method gives the non-Markovian dynamics of the system to all orders in the coupling constants. The resultant equations provide a starting point for approximations to the Kraus operators based upon approximations to the exact eigenstates $|\gamma\rangle$, to the propagator $\exp(-iHt/\hbar)$, or to the matrices in Equation (7). The results also provide a link, via the $C^{\gamma}_{\pm,n} = \langle\pm,n|\gamma\rangle$, to overlapping resonances that have been shown[24] to quantify the resistance of system superposition states to decoherence.

Above, as an example, we have considered the evolution of a d-level system in the presence of a single-mode thermal environment and have obtained the corresponding canonical Kraus operators. For $d = 2$, the exact Kraus operators for the most general interaction between the spin and the environment has been calculated, motivating studies of larger bath cases.

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References

[1] S. A. Rice, *Procedia Chem.* **2011**, *3*, 17–32.
[2] S. A. Rice, M. Zhao, *Optical Control of Molecular Dynamics*, Wiley, New York, **2000**.
[3] M. Shapiro, P. Brumer, *Principles of the Quantum Control of Molecular Processes*, Wiley, New York, **2003**; M. Shapiro, P. Brumer, *Quantum Control of Molecular Processes*, Wiley-VCH, Weinheim, **2011**.
[4] H. Carmichael, *An Open System Approach to Quantum Optics*, Springer, Berlin, **1993**.
[5] K. Kraus, *Ann. Phys.* **1971**, *64*, 311–335.

[6] M. A. Nielsen, I. L. Chuang, *Quantum Computation and Quantum Information*, Cambridge University Press, Cambridge, **2000**.
[7] D. W. Leung, *J. Math. Phys.* **2003**, *44*, 528–533.
[8] I. L. Chuang, D. W. Leung, Y. Yamamoto, *Phys. Rev. A* **1997**, *56*, 1114–1125.
[9] Y.-X. Liu, S. K. Ozdemir, A. Miranowicz, N. Imoto, *Phys. Rev. A* **2004**, *70*, 042308.
[10] D. Bacon, D. A. Lidar, K. B. Whaley, *Phys. Rev. A* **1999**, *60*, 1944–1955.
[11] H. Nakazato, Y. Hida, K. Yuasa, B. Militello, A. Napoli, A. Messina, *Phys. Rev. A* **2006**, *74*, 062113.
[12] D. M. Tong, L. C. Kwek, C. H. Oh, J.-L. Chen, L. Ma, *Phys. Rev. A* **2004**, *69*, 054102.
[13] L.-A. Wu, G. Kurizki, P. Brumer, *Phys. Rev. Lett.* **2009**, *102*, 080405; L.-A. Wu, A. Bhairoke, P. Brumer, *J. Chem. Phys.* **2008**, *129*, 041105.
[14] A. J. van Wonderen, K. Lendi, *Europhys. Lett.* **2005**, *71*, 737–743.
[15] M. S. Kim, J. Lee, D. Ahn, P. L. Knight, *Phys. Rev. A* **2002**, *65*, 040101.
[16] T. F. Jordan, A. Shaji, E. C. G. Sudarshan, *Phys. Rev. A* **2004**, *70*, 052110.
[17] E. K. Irish, J. Gea-Banaclocke, I. Martin, K. C. Schwab, *Phys. Rev. B* **2005**, *72*, 195410.
[18] P. Caban, K. A. Smolinski, Z. Walczak, *Phys. Rev. A* **2003**, *68*, 034308.
[19] M. S. Tame, M. Paternostro, C. Hadley, S. Bose, M. S. Kim, *Phys. Rev. A* **2006**, *74*, 042330.
[20] G. Kimura, *Phys. Lett. A* **2003**, *314*, 339–349.
[21] S. Kryszewski, M. Zachcial, arXiv:quant-ph/0602065v1.
[22] H. Narnhofer, *J. Phys. A* **2006**, *39*, 7051–7064.
[23] R. A. Bertlmann, P. Krammer, *J. Phys. A* **2008**, *41*, 235303.
[24] A. Biswas, M. Shapiro, P. Brumer, *J. Chem. Phys.* **2010**, *133*, 014103.

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