Partial-trace-free time-convolutionless equation of motion for the reduced density matrix

Irena Knezevic* and David K. Ferry†
Department of Electrical Engineering and Center for Solid State Electronics Research, Arizona State University, P.O. Box 876206, Tempe, Arizona 85287-6206

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Evolution of a system, coupled to its environment and influenced by external driving fields, is an old problem that remains of interest. In this paper, we derive an equation of motion for the reduced system density matrix, which is time convolutionless and free of the partial trace with respect to the environment states. This new approach uses an extension of the projection-operator technique, which incorporates an isomorphism between the system's Liouville space and the unit eigenspace of the projection operator induced by the uniform environment density matrix. Numerical application of the present approach is particularly useful in large externally driven systems, as the partial-trace-free equation is given in terms of submatrices significantly smaller than the matrices in the conventional time-convolutionless approaches, which alleviates the computational burden. We also show that all time-convolutionless approaches, conventional or partial-trace-free, are based upon a hidden underlying assumption of time reversibility of the system's evolution. This feature puts significant constraints on applicability of time-convolutionless approaches when employing approximations that yield time irreversibility. Also, we investigate the application of the approach in the description of far-from-equilibrium systems.

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I. INTRODUCTION

Describing the evolution of a quantum-mechanical system, coupled to its environment and influenced by external driving fields, is one of the oldest and most important problems in quantum mechanics [1]. The problem actually lies in attempting to describe the system's time development, without necessarily having to collect too much information about the environment. A widely used approach to obtaining the equation of motion for the reduced system density matrix, which contains full information about the system, is by using a *projection-operator technique*, introduced by Nakajima [2], Zwanzig [3], and Mori [4]. Variants of the projection-operator technique have successfully been used in many fields. For example, Argyres and Kelley [5] presented a theory of linear response in spin systems, Barker and Ferry [6] treated quantum transport in very small semiconductor devices, Kassner [7] analyzed relaxation in systems with initial system-bath coupling, Sparpaglione and Mukamel [8] presented a theory for electron transfer in polar media, followed by analyses of condensed-phase electron transfer by Hu and Mukamel [9], and Romero-Rochin and Oppenheim [10] addressed the relaxation of a two-level system weakly coupled to a bath. Essentially, all variations of the technique rely upon the use of two complementary projection operators to generate the equations of motion for two mutually orthogonal projections of the total ''system + environment'' density matrix. Equations of motion for the two projections are coupled, and the equation for the relevant projection, i.e., the one yielding the reduced density matrix after a partial trace is taken over the environment states, can be obtained in a closed form. This requires incorporating a formal solution of the equation for the orthogonal projection. The resulting equation of motion for the reduced density matrix typically exhibits non-Markovian (or time-convolution) behavior. The non-Markovian nature and the need for full knowledge of the system+environment (in order to take the partial trace over environmental states) are significant constraints on this approach. An equation of motion containing a term with a memory kernel is particularly difficult to solve self-consistently, and one must often be satisfied with the fast-modulation (Markov) limit.

In response to the difficulties arising from memory kernels in nonequilibrium statistical mechanics, Tokuyama and Mori [11] first proposed a time-convolutionless (also known as ''memoryless'') equation of motion in the Heisenberg picture. Soon afterwards, derivations of Shibata and co-workers [12,13] in the Schrödinger picture appeared, and this approach is the basis for much work that followed. This includes Saeki's analysis of linear response of an externally driven system coupled to a heat bath [14] and work on systems coupled to a stochastic reservoir [15,16]. The latter was extended by Ahn to formulate the quantum kinetic equations for semiconductors [17,18] and arrive at a theory of optical gain in quantum-well lasers [19]. Chang and Skinner [20] applied the time-convolutionless approach to analyze the relaxation of a two-level system strongly coupled to a harmonic bath. More recently, Ahn *et al.* treated noisy quantum channels [21] and quantum information processing [22], and Golosov and Reichmann [23] analyzed condensed-phase charge-transfer processes. Both the time-convolutionless equation of motion [12–23] and the non-Markovian equations described in the previous paragraph [2–10] are based on projection-operator techniques. The difference is that, in the time-convolutionless approach, the memory effects are taken into account by evaluating particular evolution operators, which couple states of the system with the environment states, rather than through a term with a memory kernel.

*Email address: irenak@asu.edu
†Email address: ferry@asu.edu

However, the entire approach is based on the assumption of invertibility of one of the evolution operators, and this is an important issue that has not been addressed adequately in the literature so far. Once a time-convolutionless equation for the relevant projection is obtained, a partial trace with respect to the environment states is performed, as before, to uncover the evolution of the reduced density matrix.

The resulting time-convolutionless equation of motion for the reduced density matrix still has two major shortcomings. First, it has an explicit dependence on the choice of the projection operator (or, on the environment density matrix that induces the projection operator). This is unphysical, as the projection operator is just a tool, an external assumption, and in the end the equation of motion for the reduced density matrix should not depend on such assumptions. This does not mean that the time-convolutionless approach is incorrect, it just means that all the equations for different projection operators eventually must reduce to one and the same equation. The second shortcoming is that the partial trace used to obtain the evolution of the reduced density matrix (from the equation of motion for the relevant projection) requires one to evaluate large and unpleasant evolution matrices, only to extract significantly less information after the partial trace. In other words, one must obtain much more information than actually necessary. With increasing size of the system + environment, and inclusion of a time dependence due to external forces, this obstacle becomes more serious.

In this paper, two main results are presented. First, we introduce a very useful and simple isomorphism between the space of operators acting on the system's Hilbert space and the unit eigenspace of the projection operator induced by the uniform density matrix of the environment. This allows us to derive a time-convolutionless equation of motion for the reduced density matrix, which is free of the partial trace with respect to environment states. The isomorphism enables us to *effectively perform a partial trace with respect to the environment states without actually performing it: what we perform instead is a well-defined basis transformation*. Being partial-trace-free, this equation successfully addresses one of the shortcomings mentioned in the previous paragraph. It depends on submatrices considerably smaller than those in the conventional time-convolutionless approach [12–23], which makes the present approach especially interesting for numerical application in those cases where evaluation of large evolution matrices is particularly undesirable. Second, we point out that a time-convolutionless approach, conventional or partial-trace-free, is based on the assumption of invertibility of a particular ''entanglement operator,'' and show that this assumption is equivalent to the assumption of reversibility of the system's evolution. This puts constraints on the applicability of time-convolutionless approaches, in general. In particular, we investigate the application of the approach to the description of a steady state in far-from-equilibrium situations.

In Sec. II, we first introduce some basic definitions and notation (Sec. II A), and then review the projection-operator technique employed to obtain the conventional time-convolutionless equation of motion (Sec. II B). In Sec. III, we first outline major steps in solving the eigenproblem of the projection operator itself, and construct the desired isomorphism, which effectively performs the partial trace (Sec. III A). This isomorphism enables us to derive our main result, the partial-trace-free equation of motion (34) for the reduced density matrix (Sec. III B). Numerical applicability of the present approach is analyzed in detail in Sec. III C. Section IV A addresses the second important issue of this work, the assumption of reversibility, which we prove is inherent to any time-convolutionless approach. We illustrate how a time-convolutionless approach behaves when describing far-from-equilibrium situations. We also show how the partial-trace-free equation is used for the case of no initial coupling between the system and the environment (Sec. IV B). Finally, we conclude with a brief summary in Sec. V.

II. PROJECTION-OPERATOR TECHNIQUE

A. Basic definitions

Consider a system S, interacting with its environment E, so that the system + environment ($S+E$) is either closed, or influenced by external driving fields that are assumed known and unaffected by the feedback from $S+E$. The Hilbert spaces of both the environment and the system, $\mathcal{H}_E$ and $\mathcal{H}_S$, respectively, are assumed to be finite dimensional, $d_E = \dim \mathcal{H}_E$, $d_S = \dim \mathcal{H}_S$. These two spaces form a tensor-product Hilbert space of the system+environment, $\mathcal{H} = \mathcal{H}_E \otimes \mathcal{H}_S$, with dimensionality $d = \dim \mathcal{H} = d_E d_S$. The spaces of operators acting on $\mathcal{H}_E$, $\mathcal{H}_S$, and $\mathcal{H}$ will be denoted by $\mathcal{H}_E^2$, $\mathcal{H}_S^2$, and $\mathcal{H}^2$ [24], respectively, whereas the elements of $\mathcal{H}_E^2$, $\mathcal{H}_S^2$, and $\mathcal{H}^2$ (i.e., operators on $\mathcal{H}_E$, $\mathcal{H}_S$, and $\mathcal{H}$) will be denoted by lowercase letters, Greek or Roman. Moreover, operators acting on $\mathcal{H}_E^2$, $\mathcal{H}_S^2$, and $\mathcal{H}^2$, sometimes called *superoperators*, will be denoted by Roman capital letters. When there is no risk of confusion, we will simply refer to them as operators.

Let us choose a basis $\{|i\rangle \; | i=1,\ldots,d_E\}$ in $\mathcal{H}_E$ and a basis $\{|\alpha\rangle \; | \alpha=1,\ldots,d_S\}$ in $\mathcal{H}_S$, which induce a tensor-product basis $\{|i\alpha\rangle \equiv |i\rangle \otimes |\alpha\rangle \; | i=1,\ldots,d_E; \; \alpha=1,\ldots,d_S\}$ in $\mathcal{H}$. These naturally give rise to the following expanded bases:

$$\{\|ij\rangle\rangle \equiv |i\rangle \otimes \langle j| \, | i,j=1,\ldots,d_E\} \quad \text{in } \mathcal{H}_E^2,$$

$$\{\|\alpha\beta\rangle\rangle \equiv |\alpha\rangle \otimes \langle\beta| \, | \alpha,\beta=1,\ldots,d_s\} \quad \text{in } \mathcal{H}_s^2, \qquad (1)$$

and

$$\{\|i\alpha, j\beta\rangle\rangle \equiv |i\alpha\rangle \otimes \langle j\beta| \, | i,j=1,\ldots,d_E; \; \alpha,\beta=1,\ldots,d_s\} \quad \text{in } \mathcal{H}^2,$$

[we use the double ket $\|\ldots\rangle\rangle$ for the Dirac notation of vectors in $\mathcal{H}_E^2$, $\mathcal{H}_S^2$, and $\mathcal{H}^2$ (double bra $\langle\langle\ldots\|$ for their adjoints) to distinguish from single ket and bra, reserved for vectors in $\mathcal{H}_E$, $\mathcal{H}_S$, and $\mathcal{H}$]. For an operator x on $\mathcal{H}$ (i.e., an element of $\mathcal{H}^2$), one can write

$$x=\|x\rangle\rangle=\underbrace{\sum_{i,j=1}^{d_E}\sum_{\alpha,\beta=1}^{d_S}x_{j\beta}^{i\alpha}|i\alpha\rangle\langle j\beta|}_{\text{as operator on }\mathcal{H}}=\underbrace{\sum_{i,j=1}^{d_E}\sum_{\alpha,\beta=1}^{d_S}x^{i\alpha,j\beta}\|i\alpha,j\beta\rangle\rangle}_{\text{as vector in }\mathcal{H}^2},\quad x_{j\beta}^{i\alpha}=x^{i\alpha,j\beta}, \tag{2}$$

whereas for an operator A on $\mathcal{H}^2$, the following form is valid

$$A=\sum_{i,j,p,q=1}^{d_E}\sum_{\alpha,\beta,\sigma,\gamma=1}^{d_S}A_{p\sigma,q\gamma}^{i\alpha,j\beta}\|i\alpha,j\beta\rangle\rangle\langle\langle p\sigma,q\gamma\|. \tag{3}$$

Analogous expressions are easily obtained for elements of $\mathcal{H}_E^2,\mathcal{H}_S^2$ [25], and operators on $\mathcal{H}_E^2,\mathcal{H}_S^2$.

The total Hamiltonian h, acting on $\mathcal{H}$, can be written as

$$h(t)=h_s(t)+h_E(t)+h_{\text{int}}(t), \tag{4}$$

where h_S is the system Hamiltonian, h_E is the environment Hamiltonian, and h_{int} describes the system-environment interaction. It is important to note that driving fields, provided by applied potentials, are included as part of the system. These are assumed to arise for $t>0$, and provide an explicit time dependence for h. Since $S+E$ is isolated, the total density matrix ρ evolves according to the Liouville equation ($\hbar=1$),

$$\frac{d\rho(t)}{dt}=-i[h(t),\rho(t)]\equiv -iL(t)\rho(t), \tag{5}$$

with $L=L_S+L_E+L_{\text{int}}$ being the Liouville superoperator, in one-to-one correspondence with the Hamiltonian (4). Equation (5) actually represents a system of $d_E^2d_S^2$ linear first-order differential equations on the time interval $t\in[0,\infty)$. Since L is continuous throughout this entire interval, we are guaranteed to have a unique solution of Eq. (5) on $t\in[0,\infty)$ for a given initial condition $\rho(0)$ [26].

The quantity that describes the evolution of the system S is *the reduced density matrix* ρ_S, defined by

$$\rho_s=\text{Tr}_E(\rho), \tag{6}$$

where $\text{Tr}_E(\ldots)$ denotes the partial trace over the environment states. The goal is to deduce how ρ_S evolves, without having to gather too much information about the environment.

B. Projection-operator technique

Let us now choose *an arbitrary density matrix* ρ_D (subscript D stands for dummy, as ρ_D is a test object for the time being), which acts on $\mathcal{H}_E$ and is constant in time. We introduce time-independent projection operators P_D and Q_D, which are associated with ρ_D and act on $\mathcal{H}^2$, as

$$P_Dx=\rho_D\otimes\text{Tr}_E\,x,\quad Q_D=1-P_D\quad(x\in\mathcal{H}^2). \tag{7}$$

In particular, since $\text{Tr}_E(\rho_D)=1$ (ρ_D is a proper density matrix), it follows that

$$\text{Tr}_E(P_D\rho)=\text{Tr}_E(\rho_D)\text{Tr}_E\,\rho=\text{Tr}_E\,\rho=\rho_s. \tag{8}$$

Therefore, the evolution of $\text{Tr}_E(P_D\rho)$ should be described by an equation that is independent of ρ_D (or, equivalently, P_D).

Since P_D and Q_D are time independent, they commute with the time derivative. Therefore, for the equations of motion of $P_D\rho$ and $Q_D\rho$, we obtain from Eq. (5)

$$\frac{dP_D\rho(t)}{dt}=-iP_DL(t)\rho(t)$$
$$=-iP_DL(t)P_D\rho(t)-iP_DL(t)Q_D\rho(t), \tag{9a}$$

$$\frac{dQ_D\rho(t)}{dt}=-iQ_DL(t)\rho(t)$$
$$=-iQ_DL(t)Q_D\rho(t)-iQ_DL(t)P_D\rho(t). \tag{9b}$$

A formal solution of Eq. (9b) is of the form

$$Q_D\rho(t)=-i\int_0^t dt'\,H(t,t')Q_DL(t')P_DG(t',t)\rho(t)$$
$$+H(t,0)Q_D\rho(0), \tag{10}$$

where for $t>t'$,

$$H(t,t')=T^c\exp\left(-i\int_{t'}^t ds\,Q_DL(s)Q_D\right),$$

$$G(t',t)=T^a\exp\left(i\int_{t'}^t ds\,L(s)\right), \tag{11}$$

and T^c and T^a denote the time-ordering and the anti-time-ordering operators, respectively. Upon introducing

$$K(t)=1+i\int_0^t dt'\,H(t,t')Q_DL(t')P_DG(t',t), \tag{12}$$

Eq. (10) can be rearranged to give

$$K(t)Q_D\rho(t)=[1-K(t)]P_D\rho(t)+H(t,0)Q_D\rho(0). \tag{13}$$

If $K(t)$ is invertible, which is an important question to which we will return, Eq. (13) becomes

$$Q_D\rho(t)=[K(t)^{-1}-1]P_D\rho(t)+K(t)^{-1}H(t,0)Q_D\rho(0), \tag{14}$$

and Eq. (9a) can be rewritten as

$$\frac{dP_D\rho(t)}{dt} = iP_DL(t)K(t)^{-1}P_D\rho(t) - iP_DL(t)K(t)^{-1}H(t,0)Q_D\rho(0). \quad (15)$$

After taking a partial trace with respect to the environment states, Eq. (15) will give us the equation of motion for ρ_S as

$$\begin{aligned}\frac{d\rho_s(t)}{dt} &= -i\,\mathrm{Tr}_E[P_DL(t)K(t)^{-1}P_D\rho(t)] \\ &\quad -i\,\mathrm{Tr}_E[P_DL(t)K(t)^{-1}H(t,0)Q_D\rho(0)] \\ &= -i\,\mathrm{Tr}_E[L(t)K(t)^{-1}\rho_D\otimes\rho_s(t)] \\ &\quad -i\,\mathrm{Tr}_E[L(t)K(t)^{-1}H(t,0)Q_D\rho(0)] \\ &= -i\langle L(t)K(t)^{-1}\rangle_D\rho_s(t) \\ &\quad -i\,\mathrm{Tr}_E[L(t)K(t)^{-1}H(t,0)Q_D\rho(0)]. \end{aligned} \quad (16)$$

The last line was obtained by introducing $\langle\cdots\rangle_D$, which maps operators acting on $\mathcal{H}^2$ onto operators acting on $\mathcal{H}_S^2$: for any given A, $\langle A\rangle_D$ is defined as $(\langle A\rangle_D)^{\alpha\beta}_{\sigma\gamma} = \sum_{i,p,q=1}^{d_E} A^{i\alpha,i\beta}_{p\sigma,q\gamma}(\rho_D)^{pq}$ [see Eq. (3)]. Equation (16) is what we have, so far, referred to as *the conventional time-convolutionless equation of motion for* $\rho_S(t)$ [12–23].

As already mentioned in the Introduction, there are two features of Eq. (16) that need attention. First, within the partial trace in both terms on the right-hand side of Eq. (16), there is an explicit dependence on the choice of the projection operator P_D (or, equivalently, on the environment density matrix ρ_D that induces the projection operator), so one must make a choice of ρ_D to actually be able to use Eq. (16). As ρ_D is just an external arbitrary tool, in the end the equation of motion for $\rho_S(t)$ should not depend on it. This does not mean that the time-convolutionless approach is incorrect, it just means that all the equations for different projection operators ought to, eventually, reduce to one and the same equation. What that equation should be, or how to rigorously prove mutual equivalence of all the equations with different P_D's, is not presently known.

The second important issue is that, due to the partial trace, one must evaluate generally large and unpleasant evolution matrices H, G, and K, only to extract significantly less information after the partial trace. Much more information than actually necessary has to be obtained. With increasing size of the system + environment and inclusion of a time dependence due to external driving, this obstacle becomes increasingly serious when numerical implementation of Eq. (16) is considered.

III. PARTIAL-TRACE-FREE DIFFERENTIAL EQUATION OF MOTION FOR THE REDUCED DENSITY MATRIX

In the preceding section, we realized that a serious constraint on the application of the conventional time-convolutionless approach (16) in large systems is evaluation of large matrices, when not all the information contained in them is needed. In this section, we derive a partial-trace-free equation of motion for ρ_S, which successfully addresses this issue.

A. Choice of $\overline{P}_D$

Some features of the eigenvalue problem of P_D are virtually obvious: P_D is Hermitian, and it is easily verified that P_D is idempotent, i.e., $P_D^2 = P_D$. Hence, the eigenvalues are 0 and 1. On the other hand, by construction (7), the image space of P_D corresponds with $\mathcal{H}_S^2$, so one sees *that the eigenspace of* P_D*, corresponding to the eigenvalue 1, ought to be isomorphic to* $\mathcal{H}_S^2$. A formal solution of the eigenproblem of P_D is given in the Appendix. An important and useful point that we need to recognize is the decomposition of $\mathcal{H}^2$ into a direct sum of eigenspaces of P_D,

$$\mathcal{H}^2 = (\mathcal{H}^2)_{P_D=1} \oplus (\mathcal{H}^2)_{P_D=0}, \quad (17)$$

where $(\mathcal{H}^2)_{P_D=1}$ is the d_S^2-dimensional unit eigenspace and $(\mathcal{H}^2)_{P_D=0}$ is the $d_S^2(d_E^2-1)$-dimensional zero eigenspace.

If we choose an orthonormal eigenbasis of P_D $\{\|n_D\rangle\rangle \mid n_D = 1,\ldots,d_E^2d_S^2\}$ so that, according to decomposition (17), the first d_S^2 basis vectors span $(\mathcal{H}^2)_{P_D=1}$, we will be able to write

$$P_D = \sum_{n_D=1}^{d_S^2} \|n_D\rangle\rangle\langle\langle n_D\|. \quad (18)$$

The crucial question that we ask is: *is it possible not to mix original basis vectors* $\|i\alpha,j\beta\rangle\rangle$ *with different* α, β *to obtain a given* $\|n_D\rangle\rangle$? The answer is: *yes, only if the density matrix inducing the projection operator is the uniform density matrix* [27], i.e.,

$$\overline{\rho}_D \equiv d_E^{-1}1_{d_E\times d_E}, \quad (19)$$

with the associated projection operator denoted by $\overline{P}_D$ (see the Appendix for a formal proof of this statement). One indeed finds that the vectors defined as

$$\|\overline{\alpha\beta}\rangle\rangle \equiv \frac{1}{\sqrt{d_E}}\sum_{i=1}^{d_E}\|i\alpha,i\beta\rangle\rangle \quad (20)$$

constitute an orthonormal basis within the unit eigenspace of $\overline{P}_D$ [28], namely,

$$\overline{P}_D\|\overline{\alpha\beta}\rangle\rangle = \|\overline{\alpha\beta}\rangle\rangle, \quad \langle\langle\overline{\alpha\beta}\|\overline{\sigma\gamma}\rangle\rangle = \delta_{\alpha\sigma}\delta_{\beta\gamma} \quad (\forall\ \alpha,\beta,\sigma,\gamma). \quad (21)$$

$\overline{P}_D$ can therefore be written as

$$\overline{P}_D = \sum_{\alpha,\beta=1}^{d_S}\|\overline{\alpha\beta}\rangle\rangle\langle\langle\overline{\alpha\beta}\|, \quad (22)$$

and it follows that

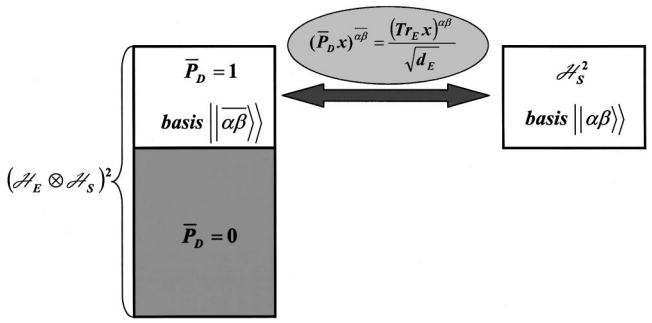

FIG. 1. The isomorphism (26) between the unit eigenspace of $\bar{P}_D$ and the system's Liouville space $\mathcal{H}_S^2$. This isomorphism enables us to effectively perform a partial trace over the environmental states by remaining in $\mathcal{H}^2$ and working in the eigenbasis of $\bar{P}_D$.

$$\bar{P}_D x = \sum_{\alpha,\beta=1}^{d_S} (\bar{P}_D x)^{\overline{\alpha\beta}} \|\overline{\alpha\beta}\rangle\rangle, \tag{23}$$

where

$$(\bar{P}_D x)^{\overline{\alpha\beta}} = \langle\langle \overline{\alpha\beta} \| x \rangle\rangle = \frac{1}{\sqrt{d_E}} \sum_{i=1}^{d_E} \langle\langle i\alpha, i\beta \| x \rangle\rangle = \frac{1}{\sqrt{d_E}} \sum_{i=1}^{d_E} x^{i\alpha,i\beta} \tag{24}$$

(we identified $x = \|x\rangle\rangle$ to take advantage of the compactness of the Dirac notation). Using the fact that $\mathrm{Tr}_E x$, being a vector on $\mathcal{H}_S^2$, is written in terms of the basis $\{\|\alpha\beta\rangle\rangle\}$ as

$$\mathrm{Tr}_E x = \sum_{\alpha,\beta=1}^{d_S} (\mathrm{Tr}_E x)^{\alpha\beta} \|\alpha\beta\rangle\rangle = \sum_{\alpha,\beta=1}^{d_S} \left(\sum_{i=1}^{d_E} x^{i\alpha,i\beta} \right) \|\alpha\beta\rangle\rangle, \tag{25}$$

from Eq. (24) we obtain the crucial equation for the rest of this work,

$$(\bar{P}_D x)^{\overline{\alpha\beta}} = \frac{(\mathrm{Tr}_E x)^{\alpha\beta}}{\sqrt{d_E}}. \tag{26}$$

Equation (26) represents a very useful isomorphism between $(\mathcal{H}^2)_{P_D=1}$ (the unit eigenspace of $\bar{P}_D$) and $\mathcal{H}_S^2$, which is depicted in Fig. 1. We have managed to ''preserve'' the identity of basis vectors $\{\|\alpha\beta\rangle\rangle\}$ from $\mathcal{H}_S^2$ when mapping to $(\mathcal{H}^2)_{P_D=1}$. This isomorphism is basis induced, and enables us to *effectively perform a partial trace with respect to the environment states without actually performing it: instead, what we do perform is a basis transformation in* $\mathcal{H}^2$. This is the basis for the derivation of a partial-trace-free equation of motion for the reduced density matrix.

B. Partial-trace-free differential equation of motion for ρ_S

According to Eq. (26), there is a very simple correspondence between how $\bar{P}_D\rho$ looks in terms of the eigenbasis $\{\|\overline{\alpha\beta}\rangle\rangle\}$, Eq. (20), and how ρ_S looks in terms of the basis $\{\|\alpha\beta\rangle\rangle\}$ in $\mathcal{H}_S^2$, namely,

$$(\rho_S)^{\alpha\beta} = \sqrt{d_E} (\bar{P}_D \rho)^{\overline{\alpha\beta}}. \tag{27}$$

Using this result, we conclude that the evolution of the representation matrix of ρ_S can be tracked in $(\mathcal{H}^2)_{P_D=1}$ directly, by following the evolution $\bar{P}_D\rho$ without taking the partial trace with respect to the environment states (which would mean going back to $\mathcal{H}_S^2$). The price is, however, that everything must be written in the eigenbasis of $\bar{P}_D$, whose first d_S^2 vectors are $\{\|\overline{\alpha\beta}\rangle\rangle\}$, Eq. (20), and the rest can be straightforwardly constructed (for details, see Sec. III C and the Appendix). In this basis, the projection operators are represented according to (boldface characters denote the representation matrices)

$$\bar{\boldsymbol{P}}_D = \begin{bmatrix} \boldsymbol{1}_{d_S^2 \times d_S^2} & \boldsymbol{0}_{d_S^2 \times d_S^2 (d_E^2-1)} \\ \boldsymbol{0}_{d_S^2 (d_E^2-1) \times d_S^2} & \boldsymbol{0}_{d_S^2 (d_E^2-1) \times d_S^2 (d_E^2-1)} \end{bmatrix},$$

$$\bar{\boldsymbol{Q}}_D = \begin{bmatrix} \boldsymbol{0}_{d_S^2 \times d_S^2} & \boldsymbol{0}_{d_S^2 \times d_S^2 (d_E^2-1)} \\ \boldsymbol{0}_{d_S^2 (d_E^2-1) \times d_S^2} & \boldsymbol{1}_{d_S^2 (d_E^2-1) \times d_N^2 (d_E^2-1)} \end{bmatrix}, \tag{28}$$

where we have explicitly written out the dimensions of each submatrix for clarity. Also, the density matrix is represented by

$$\boldsymbol{\rho} = \begin{bmatrix} \boldsymbol{\rho}_1 \\ \boldsymbol{\rho}_2 \end{bmatrix}, \quad \bar{\boldsymbol{P}}_D \boldsymbol{\rho} = \begin{bmatrix} \boldsymbol{\rho}_1 \\ 0 \end{bmatrix}, \tag{29}$$

where $\boldsymbol{\rho}_1$ is a d_S^2 column and $\boldsymbol{\rho}_2$ is a $d_S^2(d_E^2-1)$ column. According to Eq. (27),

$$\boldsymbol{\rho}_S = \sqrt{d_E} \boldsymbol{\rho}_1, \tag{30}$$

with $\boldsymbol{\rho}_S$ being the column that represents ρ_S in the basis $\{\|\alpha\beta\rangle\rangle\}$ of $\mathcal{H}_S^2$. The block forms of other operators involved in Eqs. (5)–(16) are readily written as

$$\boldsymbol{L}(t) = \begin{bmatrix} \boldsymbol{L}_{11}(t) & \boldsymbol{L}_{12}(t) \\ \boldsymbol{L}_{21}(t) & \boldsymbol{L}_{22}(t) \end{bmatrix}, \tag{31a}$$

$$\boldsymbol{G}(t',t) = T^a \exp\left(i \int_{t'}^{t} ds\, \boldsymbol{L}(s) \right) = \begin{bmatrix} \boldsymbol{G}_{11}(t',t) & \boldsymbol{G}_{12}(t',t) \\ \boldsymbol{G}_{21}(t',t) & \boldsymbol{G}_{22}(t',t) \end{bmatrix}, \quad t' < t. \tag{31b}$$

$$\boldsymbol{H}(t,t') = T^c \exp\left(-i \int_{t'}^{t} ds\, \overline{\boldsymbol{Q}}_D \boldsymbol{L}(s) \overline{\boldsymbol{Q}}_D \right) = \begin{bmatrix} \boldsymbol{1} & \boldsymbol{0} \\ \boldsymbol{0} & T^c \exp\left(-i \int_{t'}^{t} ds\, \boldsymbol{L}_{22}(s) \right) \end{bmatrix} = \begin{bmatrix} \boldsymbol{1} & \boldsymbol{0} \\ \boldsymbol{0} & \boldsymbol{H}_{22}(t,t') \end{bmatrix}, \tag{31c}$$

$$\begin{aligned} \boldsymbol{K}(t) &= \boldsymbol{1} + i \int_0^t dt' \begin{bmatrix} \boldsymbol{1} & \boldsymbol{0} \\ \boldsymbol{0} & \boldsymbol{H}_{22}(t,t') \end{bmatrix} \begin{bmatrix} \boldsymbol{0} & \boldsymbol{0} \\ \boldsymbol{L}_{21} & \boldsymbol{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{G}_{11}(t',t) & \boldsymbol{G}_{12}(t',t) \\ \boldsymbol{G}_{21}(t',t) & \boldsymbol{G}_{22}(t',t) \end{bmatrix} \\ &= \begin{bmatrix} \boldsymbol{1} & \boldsymbol{0} \\ i \int_0^t dt'\, \boldsymbol{H}_{22}(t,t') \boldsymbol{L}_{21}(t') \boldsymbol{G}_{11}(t',t) & 1 + i \int_0^t dt'\, \boldsymbol{H}_{22}(t,t') \boldsymbol{L}_{21}(t') \boldsymbol{G}_{12}(t',t) \end{bmatrix} \\ &= \begin{bmatrix} \boldsymbol{1} & \boldsymbol{0} \\ \boldsymbol{K}_{21}(t) & \boldsymbol{K}_{22}(t) \end{bmatrix}. \end{aligned} \tag{31d}$$

We see that $K(t)$ is invertible if

$$\begin{aligned} \det \boldsymbol{K}(t) &= \det \boldsymbol{K}_{22}(t) \\ &= \det\left[\boldsymbol{1} + i \int_0^t dt'\, \boldsymbol{H}_{22}(t,t') \boldsymbol{L}_{21}(t') \boldsymbol{G}_{12}(t',t) \right] \\ &\neq 0. \end{aligned} \tag{32}$$

If Eq. (32) is fulfilled, from Eq. (31d) we obtain

$$\boldsymbol{K}^{-1}(t) = \begin{bmatrix} \boldsymbol{1} & \boldsymbol{0} \\ -\boldsymbol{K}_{22}^{-1}(t) \boldsymbol{K}_{21}(t) & \boldsymbol{K}_{22}^{-1}(t) \end{bmatrix}. \tag{33}$$

Using the block forms (29)–(33), the equation of motion for $\overline{P}_D \rho$, Eq. (15), and the isomorphism (27), we obtain

$$\begin{aligned} \frac{d\boldsymbol{\rho}_S(t)}{dt} &= -i[\boldsymbol{L}_{11}(t) - \boldsymbol{L}_{12}(t) \boldsymbol{K}_{22}^{-1}(t) \boldsymbol{K}_{21}(t)] \boldsymbol{\rho}_S(t) \\ &\quad + i \sqrt{d_E} \boldsymbol{L}_{12}(t) \boldsymbol{K}_{22}^{-1}(t) \boldsymbol{H}_{22}(t,0) \boldsymbol{\rho}_2(0). \end{aligned} \tag{34}$$

Equation (34) is the *partial-trace-free time-convolutionless equation of motion for* ρ_S. It actually describes the evolution of the *representation matrix of* ρ_S *in the basis* $\{\|\alpha\beta\rangle\rangle\}$. Dealing with representation matrices in a given basis, rather than a representation-independent form, is not a downside in itself, particularly if one has numerical implementation in mind. In this partial-trace-free form, multiplication of generally nonsquare submatrices (e.g., $\boldsymbol{L}_{12}\boldsymbol{K}_{22}^{-1}$) is what mimics the partial trace. Moreover, since no explicit dependence on the projection operator used is present, the partial-trace-free equation (34) offers a clearer picture as to what elements of the Hamiltonian h, through the evolution of submatrices, actually come into play. This increased transparency should be useful when choosing and employing different approximations.

C. Numerical applicability of the partial-trace-free equation (34)

The present approach, which culminates in Eq. (34), has interesting features as far as numerical applicability is concerned. As already mentioned, in order to solve a conventional time-convolutionless equation of motion (16), which contains a partial trace and, within it, an assumption on the environment density matrix used, one needs to first evaluate the full evolution matrices (H,G,K), only to extract the required information by the partial trace. With increasing size of the systems analyzed, the task of evaluating the entire evolution matrices becomes increasingly cumbersome. If the Liouville operator L, Eq. (5), is time independent, this problem is less difficult, as, after initially having solved the eigenproblems of L and $Q_D L Q_D$, the evolution matrices at any point in time are readily evaluated by exponentiation of these operators in their respective eigenbases, and then going back to the initial basis. However, in cases of externally driven systems, for which L is generally time dependent, the problem of evaluation of evolution matrices becomes more computationally demanding: namely, $L(t)$'s at different points in time do not commute with each other [the same holds for $Q_D L(t) Q_D$'s], and the time-ordered products in the evolution operators (11),(12) need to be evaluated for every order in the exponential series, and for each point in time.

Having this in mind, the biggest numerical advantage of Eq. (34) over the conventional time-convolutionless equation (16) is its dependence on submatrices that are appreciably smaller than the full evolution matrices used in the conventional approach. In small time-independent systems, this advantage is less important, but with increasing size and/or time dependence, any reduction in the dimension of the matrices is very welcome. Therefore, the present approach is especially adequate for large externally driven systems. (However, there is the issue of irreversibility, which is an important one when it comes to implementation, and is addressed in Sec. IV A.)

The actual implementation should follow the sequence given below.

(1) Usually, start from the matrix elements of the Liouville operator (5) in a tensor-product basis $\|i\alpha, j\beta\rangle\rangle$.

(2) According to Eq. (20), first the vectors $\|\overline{\alpha\beta}\rangle\rangle$ and then the rest of the eigenvectors of $\overline{P}_D$ are constructed (see Appendix for details). We will just note that all the vectors of the initial basis $\|i\alpha,j\beta\rangle\rangle$, such that $i\neq j$, are already both mutually orthogonal, and orthogonal to all $\|\overline{\alpha\beta}\rangle\rangle$, and that leaves, for every α,β, only d_E-1 vectors to construct, following the standard Gram-Schmidt orthogonalization procedure. Of course, any other basis in $(\mathcal{H}^2)_{P_D=0}$ may be chosen, Eq. (34) will remain the same; however, the basis in $(\mathcal{H}^2)_{P_D=1}$ must be $\|\overline{\alpha\beta}\rangle\rangle$.

(3) The matrix elements of the Liouville operator are rewritten in the new total basis of $\mathcal{H}^2$, and then the evolution operators, i.e., their submatrices (31a)–(31d), are found.

(4) Equation (34) is set up and solved.

IV. DISCUSSION

A. Irreversibility and breakdown of time-convolutionless approaches. Far-from-equilibrium situations

It was already mentioned that the existence of a time-convolutionless approach relies upon the assumption that K is invertible. However, this cannot be taken for granted. Namely, if we were capable of using the exact form of the Hamiltonians (4) and the exact initial state $\rho(0)$, and the system+environment were indeed closed except for the external driving force, then, as already mentioned, since Eq. (5) is a linear equation, there always exists a unique solution $\rho(t)$ fixed by $\rho(0)$. Consequently, there exists a unique solution for $\rho_S(t)$. Unfortunately, we are most often not quite so lucky. We are forced to make approximations, for instance, about the initial state (e.g., assume that the environment is in thermal equilibrium, or perform some type of averaging) or about the dynamics (e.g., Markov approximation, retardation effects, local approximations). Also, very often the system +environment that we consider is not actually closed, but coupled to some outer environment, and we might want to include some information about that coupling. The bottom line is that introducing approximations may lead to irreversible behavior. This has been well known for non-Markovian approaches, which, due to the robustness of the memory kernel's integral form, seem to tolerate such approximations fairly well. However, the same does not hold for time-convolutionless approaches.

The assumption of invertibility of K is actually *the assumption of time reversibility of ρ_S's evolution*. Namely, let us introduce shorthand notation

$$\boldsymbol{Z}(t)=\boldsymbol{L}_{11}(t)-\boldsymbol{L}_{12}(t)\boldsymbol{K}_{22}^{-1}(t)\boldsymbol{K}_{21}(t),$$

$$\boldsymbol{Y}(t,0)=i\sqrt{d_E}\boldsymbol{L}_{12}(t)\boldsymbol{K}_{22}^{-1}(t)\boldsymbol{H}_{22}(t,0)\boldsymbol{\rho}_2(0). \tag{35}$$

Then, Eq. (34) can then be rewritten as

$$\frac{d\rho_S(t)}{dt}=-i\boldsymbol{Z}(t)\boldsymbol{\rho}_S(t)+\boldsymbol{Y}(t,0). \tag{36}$$

In order for Eq. (36) to have a unique solution, fixed by the initial $\rho(0)$, for all $t\in[0,\infty)$, $\boldsymbol{Z}$ and $\boldsymbol{Y}$ must be continuous in this time interval [26]. This requirement is fulfilled if $\boldsymbol{K}_{22}$ is invertible for all $t\in[0,\infty)$, in which case the evolution of the reduced density matrix ρ_S is described by

$$\boldsymbol{\rho}_S(t)=\boldsymbol{U}(t,0)\left\{\boldsymbol{\rho}_S(0)+\int_0^t d\tau[\boldsymbol{U}(\tau,0)]^{-1}\boldsymbol{Y}(\tau,0)\right\},$$

$$\boldsymbol{U}(t,0)=T^c\exp\left(-i\int_0^t d\tau\,\boldsymbol{Z}(\tau)\right). \tag{37}$$

Apparently, the assumption of invertibility of K, which led us to the differential equation (34) in the first place, also guarantees the existence and uniqueness of the solution (37). Therefore, *time-irreversible processes cannot be described by a time-convolutionless approach*, as that implies that the inverse of K vanishes. This statement holds for both the conventional and the partial-trace-free approach, although the above equations are given for only the latter.

There is another way of grasping the true meaning behind the vanishing inverse of K. Namely, by using the block forms (28)–(33), Eq. (13) can be written as

$$\begin{bmatrix}\boldsymbol{1} & \boldsymbol{0}\\ \boldsymbol{K}_{21}(t) & \boldsymbol{K}_{22}(t)\end{bmatrix}\begin{bmatrix}\boldsymbol{\rho}_1(t)\\ \boldsymbol{\rho}_2(t)\end{bmatrix}=\begin{bmatrix}\boldsymbol{\rho}_1(t)\\ \boldsymbol{0}\end{bmatrix}+\begin{bmatrix}\boldsymbol{1} & \boldsymbol{0}\\ \boldsymbol{0} & \boldsymbol{H}_{22}(t)\end{bmatrix}\begin{bmatrix}\boldsymbol{0}\\ \boldsymbol{\rho}_2(0)\end{bmatrix}, \tag{38a}$$

which yields

$$\boldsymbol{K}_{22}(t)\boldsymbol{\rho}_2(t)=-\boldsymbol{K}_{21}(t)\boldsymbol{\rho}_1(t)+\boldsymbol{H}_{22}(t,0)\boldsymbol{\rho}_2(0). \tag{38b}$$

If $\det\boldsymbol{K}_{22}(\tilde{t})=0$ at some point in time $\tilde{t}$, there actually is a constraint on the right-hand side of Eq. (38b), since we require that the solution $\boldsymbol{\rho}_2(\tilde{t})$ exists. Namely, the right-hand side of Eq. (38b) must be orthogonal to all solutions $\boldsymbol{y}$ of the equation $\boldsymbol{K}_{22}^{+}(\tilde{t})\boldsymbol{y}=\boldsymbol{0}$ [29], i.e.,

$$\langle-\boldsymbol{K}_{21}(t)\boldsymbol{\rho}_1(t)+\boldsymbol{H}_{22}(t,0)\boldsymbol{\rho}_2(0)|\boldsymbol{y}\rangle=0$$

$$(\forall\ \boldsymbol{y},\quad \boldsymbol{K}_{22}^{+}(\tilde{t})\boldsymbol{y}=\boldsymbol{0}), \tag{39}$$

where $\langle\cdot\cdot|\cdot\cdot\rangle$ is the standard scalar product of two columns. Therefore, any approximation made for the dynamics (i.e., to $\boldsymbol{L},\boldsymbol{K},\boldsymbol{H}$) and the initial state $\boldsymbol{\rho}_2(0)$ must be such that, if the evolution becomes irreversible, i.e., $\det\boldsymbol{K}_{22}(\tilde{t})=0$, then Eq. (39) must also hold. If this requirement is fulfilled [for instance, if the right-hand side of Eq. (38b) is zero], the vanishing determinant simply means that the number of mutually independent equations in this system is less than $d_S^2(d_E^2-1)$. There will be "free elements" in the solution column $\rho_2(\tilde{t})$, in terms of which all other elements will be given. This appears as a degree of arbitrariness, or insufficiency of the information contained in the evolution submatrices and the initial condition. Simply, this is another way of stating that irreversibility occurs. In order to reset the time-

convolutionless approach after breakdown, a new initial condition $\rho(\tilde{t}^+)$ needs to be obtained from an external assumption, and that is the crucial obstacle for the application of time-convolutionless approaches. Again, although the formal steps were given for the partial-trace-free approach, they can easily be generalized for the conventional approach.

A typical situation for which one encounters time irreversibility is far from equilibrium, in externally driven systems. In this case, one actually requires that a steady-state value for ρ_S be independent of the initial state $\rho(0)$. The independence of the final state from the initial state is another way of stating that there is no unique connection between them, or that the evolution is *not reversible* [26,30]. Rather, the steady-state value is achieved by a balance between driving forces and decay forces (from the system to the environment), namely, by

$$[\boldsymbol{L}_{11}(t)-\boldsymbol{L}_{12}(t)\boldsymbol{K}_{22}^{-1}(t,\tau)\boldsymbol{K}_{21}(t,\tau)]\boldsymbol{\rho}_S(t)=\boldsymbol{0},$$
$$\tau>t_{\text{last}}, \quad \boldsymbol{\rho}_S(t)=\text{const}, \quad (40)$$

where $K(t,\tau)$ is a natural extension of the definition of $K(t)\equiv K(t,0)$, Eqs. (12) and (31d), with the lower limit of integration equaling τ; t_{last} is the last moment in time, preceding t, for which the determinant of $K(t)$ vanished [as all functions involved in K are continuous, and the determinant is a continuous function, there may be either isolated points in time or entire time intervals for which $\det K(t)$ vanishes].

In order for the steady state to be achieved, the following must hold simultaneously with Eq. (40):

$$\boldsymbol{Y}(t,\tau)=\boldsymbol{0}, \quad \tau>t_{\text{last}}. \quad (41)$$

This condition represents the vanishing of the so-called ''random force,'' and is presently not fully understood [31], but should hold for an arbitrary $\boldsymbol{\rho}_2(\tau)$. We believe that further investigation of the properties of the operator $\boldsymbol{L}_{12}(t)\boldsymbol{K}_{22}^{-1}(t,\tau)\boldsymbol{H}_{22}(t,\tau)$, Eq. (35), will reveal that its approximately null subspace increases in dimension, as time increases, which enables Eq. (41) to hold for an arbitrary $\boldsymbol{\rho}_2(\tau)$.

B. Application of Eq. (34) to the case of initially decoupled system and environment

The case of no initial coupling is important for several reasons. First, it serves as a reality check, to test the correctness of the present approach. Furthermore, it is a good starting point in many essentially nonequilibrium situations, in which the coupling builds up in time. And third, if one is interested in cases with initial coupling, the no-initial-coupling limit marks just the opposite end of the spectrum.

Let us assume that at $t=0$ the system and the environment are uncoupled, so $\rho_u(0)=\rho_E(0)\otimes\rho_S(0)$ (the extra subscript u stands for *uncoupled*). A very common approach in this case is to choose $\rho_{D,u}=\rho_E(0)$ for the projection-operator technique, so that the free term in Eq. (16) would vanish. Then, Eq. (16) can be rewritten as

$$\frac{d\rho_S(t)}{dt}=-i\langle L(t)K_u(t)^{-1}\rangle_{D,u}\rho_S(t)-i\,\mathrm{Tr}_E\left[L(t)K_u(t)^{-1}H_u(t,0)\underbrace{Q_{D,u}\rho_u(0)}_{=0}\right]$$
$$=-i\langle L(t)K_u(t)^{-1}\rangle_{D,u}\rho_S(t). \quad (42)$$

The solution of Eq. (42) is given by

$$\rho_S(t)=T^c\exp\left(-i\int_0^t d\tau\langle \boldsymbol{L}(\tau)\boldsymbol{K}_u(\tau)^{-1}\rangle_{D,u}\right)\rho_S(0). \quad (43)$$

However, in the partial-trace-free equation (34) the rightmost term survives, as $\overline{Q}_D\rho_u(0)\neq 0$, and it has no counterpart in Eq. (42). This leads us to question whether from Eq. (34) we will also obtain the solution of the form

$$\rho_S(t)=\mathcal{E}(t,0)\rho_S(0), \quad (44)$$

as we did from Eq. (42).

To answer this problem we should remember that, as seen in Sec. III A, every projection operator has a unit-eigenspace isomorphic to $\mathcal{H}_S^2$. In the eigenbasis of $P_{D,u}$, $\rho_u(0)$ can be written as

$$[\boldsymbol{\rho}_u(0)]_u=\begin{bmatrix}\boldsymbol{\rho}_{1,u}(0)\\ \boldsymbol{0}\end{bmatrix}, \quad (45)$$

where $[\cdots]_u$ denotes the representation column in the eigenbasis of $P_{D,u}$. In the eigenbasis of $\overline{P}_D$, on the other hand, $\rho_u(0)$ is written as

$$\boldsymbol{\rho}_u(0)=\begin{bmatrix}\boldsymbol{\rho}_1(0)\\ \boldsymbol{\rho}_2(0)\end{bmatrix}. \quad (46)$$

Since both eigenbases are orthonormal, there exists a unitary matrix

$$\boldsymbol{M}=\begin{bmatrix}\boldsymbol{M}_{11} & \boldsymbol{M}_{12}\\ \boldsymbol{M}_{21} & \boldsymbol{M}_{22}\end{bmatrix}, \quad (47)$$

which relates the representation columns in the two bases,

$$\begin{bmatrix} \boldsymbol{\rho}_{1,u}(0) \\ 0 \end{bmatrix} = \begin{bmatrix} \boldsymbol{M}_{11} & \boldsymbol{M}_{12} \\ \boldsymbol{M}_{21} & \boldsymbol{M}_{22} \end{bmatrix} \begin{bmatrix} \boldsymbol{\rho}_1(0) \\ \boldsymbol{\rho}_2(0) \end{bmatrix}. \tag{48}$$

As $\det \boldsymbol{M} \neq 0$ ($\boldsymbol{M}$ is unitary), one can solve uniquely for $\boldsymbol{\rho}_1(0), \boldsymbol{\rho}_2(0)$, if $\boldsymbol{\rho}_{1,u}(0)$ is given. From the second row in Eq. (48), we obtain

$$\boldsymbol{\rho}_2(0) = -\boldsymbol{M}_{22}^{-1}\boldsymbol{M}_{21}\boldsymbol{\rho}_1(0) = -d_E^{-1/2}\boldsymbol{M}_{22}^{-1}\boldsymbol{M}_{21}\boldsymbol{\rho}_S(0), \tag{49}$$

and consequently, $\boldsymbol{Y}$, Eq. (35), becomes

$$\boldsymbol{Y}(t,0) = -i\boldsymbol{L}_{12}(t)\boldsymbol{K}_{22}^{-1}(t)\boldsymbol{H}_{22}(t,0)\boldsymbol{M}_{22}^{-1}\boldsymbol{M}_{21}\boldsymbol{\rho}_S(0), \tag{50}$$

yielding

$$\boldsymbol{\rho}_S(t) = \left\{ \boldsymbol{U}(t,0) - i\int_0^t d\tau\, \boldsymbol{U}(t,\tau)\boldsymbol{L}_{12}(\tau)\boldsymbol{K}_{22}^{-1}(\tau) \times \boldsymbol{H}_{22}(\tau,0)\boldsymbol{M}_{22}^{-1}\boldsymbol{M}_{21} \right\} \boldsymbol{\rho}_S(0) \tag{51}$$

(note that the $\sqrt{d_E}$ factors have canceled out). Obviously, Eq. (51) is of the form (44), as ought to be expected for the case of no initial coupling.

V. SUMMARY

In this paper, evolution of an externally driven system, coupled to its environment, was investigated. First, a partial-trace-free time-convolutionless equation of motion for the reduced density matrix of the system was derived, Eq. (34). This result was obtained by implementing an extension of the projection-operator technique, which is based on an isomorphism, introduced in this paper, between the system Liouville space (space of operators acting on the system's Hilbert space) and the unit eigenspace of the projection operator induced by the uniform density matrix of the environment. Being partial-trace-free, this equation depends on submatrices considerably smaller than those in the conventional time-convolutionless approach. As a result, numerical application of our partial-trace-free equation (34), rather than of the conventional time-convolutionless equation (16), becomes more advantageous with increasing size and in time-dependent systems, as in these cases any reduction in the dimension of the matrices is desirable.

Second, we showed that any time-convolutionless approach, either a conventional or a partial-trace-free one, is actually based on an inherent assumption of reversibility of the system evolution. Namely, we have demonstrated that the vanishing inverse of a particular operator (whose invertibility is not *a priori* guaranteed when approximations are made, but is the underlying assumption of time-convolutionless approaches) actually means the onset of time irreversibility; i.e., the available information is insufficient to uniquely solve the system's equation of motion. Furthermore, we investigated the description of far-from-equilibrium situations, when a steady state that is independent of initial conditions is achieved. Lastly, we illustrated the application of the partial-trace-free equation to the case of no initial coupling between the system and the environment.

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APPENDIX: EIGENPROBLEM OF THE PROJECTION OPERATOR AND THE CHOICE OF $\overline{P_D}$

In this appendix, we first solve the eigenvalue problem of a projection operator P_D, which is induced by a given environment density matrix ρ_D. This solution enables us to justify the choice of $\overline{P_D}$, made in Sec. III A. We conclude with a detailed construction of an eigenbasis of $\overline{P_D}$.

If we choose $\{|i\rangle\}$ to be an orthonormal eigenbasis of ρ_D (ρ_D being considered as an operator on $\mathcal{H}_E$), it follows that

$$\rho_D = \sum_{i=1}^{d_E} \rho_{Di} \| i,i \rangle\rangle, \quad \text{where} \quad \rho_{Di} \geqslant 0, \quad \sum_{i=1}^{d_E} \rho_{Di} = 1. \tag{A1}$$

The action of P_D on x is then given by

$$P_D x = \rho_D \otimes \mathrm{Tr}_E\, x = \sum_i \rho_{Di} \| i,i \rangle\rangle \otimes \sum_{k,\alpha,\beta} x^{k\alpha,k\beta} \| \alpha,\beta \rangle\rangle = \sum_{i,k,\alpha,\beta} \rho_{Di} x^{k\alpha,k\beta} \| i\alpha, i\beta \rangle\rangle. \tag{A2}$$

From the above expression, one can see that the action of P_D on linear combinations of basis vectors $\| i\alpha, j\beta \rangle\rangle$, such that $i \neq j$, gives zero (since the environment partial trace over such vectors is zero). Consequently, the zero-eigenvalue subspace of P_D is at least $d_S^2 d_E(d_E - 1)$ dimensional. The remaining $(d_S^2 d_E)$-dimensional subspace contains vectors of the form $\Sigma_{i,\alpha,\beta} x^{i\alpha,i\beta} \| i\alpha, i\beta \rangle\rangle$. In this subspace, according to Eq. (A2), solving the eigenvalue problem of P_D, namely, the equation $P_D x = \lambda x$, can be written as

$$\rho_{Di} \sum_k x^{k\alpha,k\beta} - \lambda x^{i\alpha,i\beta} = 0 \quad (\forall \ \alpha,\beta,i), \tag{A3}$$

which, for an arbitrary pair α,β, becomes a matrix equation

$$\begin{bmatrix} \rho_{D1}-\lambda & \rho_{D1} & \rho_{D1} & \cdots \\ \rho_{D2} & \rho_{D2}-\lambda & \rho_{D2} & \cdots \\ \rho_{D3} & \rho_{D3} & \rho_{D3}-\lambda & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix} \begin{bmatrix} x^{1\alpha,1\beta} \\ x^{2\alpha,2\beta} \\ x^{3\alpha,3\beta} \\ \cdots \end{bmatrix} = 0. \tag{A4}$$

There is a nontrivial solution to Eq. (A4) only if the determinant of the system matrix vanishes, namely, if

$$\det(\boldsymbol{R}-\lambda\boldsymbol{I}) = \det \begin{bmatrix} \rho_{D1}-\lambda & \rho_{D1} & \rho_{D1} & \cdots \\ \rho_{D2} & \rho_{D2}-\lambda & \rho_{D2} & \cdots \\ \rho_{D3} & \rho_{D3} & \rho_{D3}-\lambda & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix} = 0, \tag{A5}$$

where $\boldsymbol{R}$ is defined as

$$\boldsymbol{R} = \begin{bmatrix} \rho_{D1} & \rho_{D1} & \rho_{D1} & \cdots \\ \rho_{D2} & \rho_{D2} & \rho_{D2} & \cdots \\ \rho_{D3} & \rho_{D3} & \rho_{D3} & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix}. \tag{A6}$$

We will now prove that Eq. (A5) has exactly one nonzero solution, $\lambda=1$, whereas the solution $\lambda=0$ is (d_E-1)-fold degenerate.

Say there are n nonzero ρ_i. We may always perform a simple permutation of basis vectors, or effectively transpositions of rows and columns, such that from Eq. (A5) we indeed obtain

$$\det(\boldsymbol{R}-\lambda\boldsymbol{I}) = \det \begin{bmatrix} \rho_{D1}-\lambda & \rho_{D1} & \cdots & \rho_{D1} & \rho_{D1} & \cdots & \cdots & \rho_{D1} \\ \rho_{D2} & \rho_{D2}-\lambda & \cdots & \rho_{D2} & \rho_{D2} & \cdots & \cdots & \rho_{D2} \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \underbrace{\rho_{Dn} \quad \rho_{Dn} \quad \cdots \quad \rho_{Dn}-\lambda}_{n\times n} & & & & \underbrace{\rho_{Dn} \quad \cdots \quad \cdots \quad \rho_{Dn}}_{n\times(d_E-n)} & & & \\ 0 & 0 & \cdots & 0 & -\lambda & 0 & \cdots & 0 \\ 0 & 0 & \cdots & 0 & 0 & -\lambda & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \underbrace{0 \quad 0 \quad \cdots \quad 0}_{(d_E-n)\times n} & & & & \underbrace{0 \quad 0 \quad \cdots \quad -\lambda}_{(d_E-n)\times(d_E-n)} & & & \end{bmatrix}$$

$$= (-\lambda)^{d_E-n} \det \begin{bmatrix} \rho_{D1}-\lambda & \rho_{D1} & \cdots & \rho_{D1} \\ \rho_{D2} & \rho_{D2}-\lambda & \cdots & \rho_{D2} \\ \cdots & \cdots & \cdots & \cdots \\ \rho_{Dn} & \rho_{Dn} & \cdots & \rho_{Dn}-\lambda \end{bmatrix}, \tag{A7}$$

where $\rho_{D1},\ldots,\rho_{Dn}$ are now the n nonzero ρ_{D1}. Let us now define

$$\boldsymbol{A}_n(x_1,\ldots,x_n) \equiv \begin{bmatrix} x_1-\lambda & x_1 & \cdots & x_1 \\ x_2 & x_2-\lambda & \cdots & x_2 \\ \cdots & \cdots & \cdots & \cdots \\ x_n & x_n & \cdots & x_n-\lambda \end{bmatrix}, \tag{A8}$$

where all x_i are nonzero. Then we may write

$$\det \boldsymbol{A}_n(x_1,\ldots,x_n)$$

$$= \det \begin{bmatrix} x_1-\lambda & x_1 & \cdots & x_1 \\ x_2 & x_2-\lambda & \cdots & x_2 \\ \cdots & \cdots & \cdots & \cdots \\ x_n & x_n & \cdots & x_n-\lambda \end{bmatrix}$$

$$= x_1 x_2 \cdots x_n \det \begin{bmatrix} 1-\dfrac{\lambda}{x_1} & 1 & \cdots & 1 \\ 1 & 1-\dfrac{\lambda}{x_2} & \cdots & 1 \\ \cdots & \cdots & \cdots & \cdots \\ 1 & 1 & \cdots & 1-\dfrac{\lambda}{x_n} \end{bmatrix}. \tag{A9}$$

Since a determinant does not change if we subtract one row from another, we first subtract the second row from the first, then the third from the second, and so on, finishing with the subtraction of the nth row from the $(n-1)$th row. The result can be displayed as

$$\det \boldsymbol{A}_n(x_1,\ldots,x_n)=x_1x_2\cdots x_n \det \tilde{\boldsymbol{A}}_n(x_1,\ldots,x_n), \tag{A10}$$

where we have introduced

$$\tilde{\boldsymbol{A}}_n(x_1,\ldots,x_n) \equiv \begin{bmatrix} -\frac{\lambda}{x_1} & \frac{\lambda}{x_2} & 0 & \cdots & 0 & 0 \\ 0 & -\frac{\lambda}{x_2} & \frac{\lambda}{x_3} & \cdots & 0 & 0 \\ 0 & 0 & -\frac{\lambda}{x_3} & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \cdots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & -\frac{\lambda}{x_{n-1}} & \frac{\lambda}{x_n} \\ 1 & 1 & 1 & \cdots & 1 & 1-\frac{\lambda}{x_n} \end{bmatrix}. \tag{A11}$$

If we expand the determinant of the above matrix in terms of the first column, we obtain the equation

$$\det \tilde{\boldsymbol{A}}_n(x_1,\ldots,x_n)=-\frac{\lambda}{x_1}\det \tilde{\boldsymbol{A}}_{n-1}(x_2,\ldots,x_n) +(-1)^{n+1}\prod_{i=2}^{n}\frac{\lambda}{x_i}. \tag{A12}$$

For $n=1$ and $n=2$,

$$\det \tilde{\boldsymbol{A}}_1(x_1)=1-\frac{\lambda}{x_1}, \quad \det \tilde{\boldsymbol{A}}_2(x_1,x_2)=\frac{\lambda}{x_1x_2}[\lambda-(x_1+x_2)]. \tag{A13}$$

Let us assume that

$$\det \tilde{\boldsymbol{A}}_n(x_1,\ldots,x_n)=\frac{(-1)^n\lambda^{n-1}}{x_1\cdots x_n}\left[\lambda-\sum_{i=1}^{n}x_i\right]. \tag{A14}$$

We will use mathematical induction to justify our guess (A14), based on the forms of Eq. (A13). Obviously, Eq. (A14) is true for $n=1$ and $n=2$. Suppose it is true for some n. According to Eq. (A14), for $n+1$ we obtain

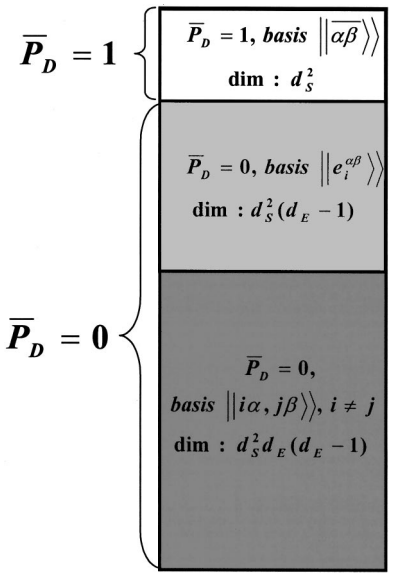

FIG. 2. An eigenbasis of $\overline{P}_D$ that enables use of the partial-trace-free equation (34).

$$\begin{aligned}\det \tilde{\boldsymbol{A}}_{n+1}(x_1,\ldots,x_{n+1}) &=-\frac{\lambda}{x_1}\det \tilde{\boldsymbol{A}}_n(x_2,\ldots,x_{n+1})+(-1)^{n+2}\prod_{i=2}^{n+1}\frac{\lambda}{x_i} \\ &=-\frac{\lambda}{x_1}\frac{(-1)^n\lambda^{n-1}}{x_2\cdots x_{n+1}}\left[\lambda-\sum_{i=2}^{n+1}x_i\right]+(-1)^{n+2}\prod_{i=2}^{n+1}\frac{\lambda}{x_i} \\ &=\frac{(-1)^{n+1}\lambda^n}{x_1x_2\cdots x_{n+1}}\left[\lambda-\sum_{i=2}^{n+1}x_i\right]+(-1)^{n+2}\prod_{i=2}^{n+1}\frac{\lambda}{x_i} \\ &=\frac{(-1)^{n+1}\lambda^n}{x_1x_2\cdots x_{n+1}}\left[\lambda-\sum_{i=2}^{n+1}x_i-x_1\right] \\ &=\frac{(-1)^{n+1}\lambda^n}{x_1x_2\cdots x_{n+1}}\left[\lambda-\sum_{i=1}^{n+1}x_i\right], \end{aligned} \tag{A15}$$

which proves our assumption (A14). Consequently, after Eq. (A10), we obtain

$$\begin{aligned}\det \boldsymbol{A}_n(x_1,\ldots,x_n)&=x_1\cdots x_n \det \tilde{\boldsymbol{A}}_n(x_1,\ldots,x_n) \\ &=(-1)^n\lambda^{n-1}\left[\lambda-\sum_{i=1}^{n}x_i\right]. \end{aligned} \tag{A16}$$

Finally, according to Eqs. (A7)–(A10) and (A16)

$$\begin{aligned}\det(\boldsymbol{R}-\lambda\boldsymbol{I})&=(-\lambda)^{d_E-n}\det \boldsymbol{A}_n(\rho_{D1},\ldots,\rho_{Dn}) \\ &=(-\lambda)^{d_E-n}(-1)^n\lambda^{n-1}\left(\lambda-\sum_{i=1}^{n}\rho_{D1}\right) \\ &=(-1)^{d_E}\lambda^{d_E-1}(\lambda-1). \end{aligned} \tag{A17}$$

The above determinant vanishes for $\lambda=0$ [(d_E-1)-fold degenerate solution] and $\lambda=1$ (nondegenerate). As Eqs. (A3)

and (A4) are defined for any given pair α, β, we conclude that the total eigenspace of P_D, corresponding to the unit eigenvalue, is d_S^2 dimensional.

So, in principle, we are now able to find eigenvectors of P_D, by solving Eq. (A3) for $\lambda=0$ and $\lambda=1$. We will not find the eigenvectors, in general, but will rather try to answer the following question, first asked in Sec. III A: *is it possible not to mix original basis vectors* $\|i\alpha,j\beta\rangle\rangle$ *with different* α,β *to obtain any given eigenvector?* The answer is: yes, *if the matrix* $\boldsymbol{R}$, *Eq.* (A6), *is normal*, so that it can be diagonalized. It can be easily checked that this requirement is fulfilled only if the density matrix inducing the projection operator is the *uniform environment density matrix* (19), i.e.,

$$\overline{\rho}_D \equiv d_E^{-1} 1_{d_E \times d_E}. \tag{A18}$$

When matrix elements of $\boldsymbol{R}$ correspond to $\overline{\rho}_D$, the eigenvector of $\boldsymbol{R}$ associated with the unit eigenvalue is of the form const$\times(1,1,\ldots,1)^T$, meaning that all $x^{i\alpha,i\beta}$ are equal for the given α,β. This implies that the vectors of the form (20), repeated here,

$$\|\overline{\alpha\beta}\rangle\rangle = \frac{1}{\sqrt{d_E}} \sum_{i=1}^{d_E} \|i\alpha,i\beta\rangle\rangle \quad (\forall\ \alpha,\beta), \tag{A19}$$

constitute an orthonormal eigenbasis in the unit eigenspace of $\overline{P}_D$ [Eq. (21) in Sec. III A), i.e.,

$$\overline{P}_D \|\overline{\alpha\beta}\rangle\rangle = \|\overline{\alpha\beta}\rangle\rangle, \quad \langle\langle\overline{\alpha\beta}\|\overline{\sigma\gamma}\rangle\rangle = \delta_{\alpha\sigma}\delta_{\beta\gamma} \quad (\forall\ \alpha,\beta,\sigma,\gamma). \tag{A20}$$

When constructing the rest of the eigenbasis of $\overline{P}_D$, it is noteworthy that all the vectors of the initial basis $\|i\alpha,j\beta\rangle\rangle$, such that $i\neq j$, are already both mutually orthogonal and orthogonal to all $\|\overline{\alpha\beta}\rangle\rangle$. This leaves, for every α,β, only d_E-1 vectors to construct (we will call them $\|e_1^{\alpha\beta}\rangle\rangle,\ldots,\|e_{d_E-1}^{\alpha\beta}\rangle\rangle$), and once they have been constructed for one pair α,β, the same construction procedure holds for every other pair (and the expansion coefficients are also the same), and follows the standard Gram-Schmidt orthogonalization procedure outlined below,

$$\|e_1^{\alpha\beta}\rangle\rangle = \frac{\|a_1^{\alpha\beta}\rangle\rangle}{\|a_1^{\alpha\beta}\|}, \quad \|a_1^{\alpha\beta}\rangle\rangle = \|1\alpha,1\beta\rangle\rangle - \underbrace{\langle\langle\overline{\alpha\beta}\|1\alpha,1\beta\rangle\rangle}_{1/\sqrt{d_E}}\|\overline{\alpha\beta}\rangle\rangle,$$

$$\|e_2^{\alpha\beta}\rangle\rangle = \frac{\|a_2^{\alpha\beta}\rangle\rangle}{\|a_2^{\alpha\beta}\|}, \quad \|a_2^{\alpha\beta}\rangle\rangle = \|2\alpha,2\beta\rangle\rangle - \underbrace{\langle\langle\overline{\alpha\beta}\|2\alpha,2\beta\rangle\rangle}_{1/\sqrt{d_E}}\|\overline{\alpha\beta}\rangle\rangle - \langle\langle e_1^{\alpha\beta}\|2\alpha,2\beta\rangle\rangle\|e_1^{\alpha\beta}\rangle\rangle,$$

$$\vdots$$

$$\|e_{d_E-1}^{\alpha\beta}\rangle\rangle = \frac{\|a_{d_E-1}^{\alpha\beta}\rangle\rangle}{\|a_{d_E-1}^{\alpha\beta}\|}, \quad \|a_{d_E-1}^{\alpha\beta}\rangle\rangle = \|d_E-1,\alpha;d_E-1,\beta\rangle\rangle - \underbrace{\langle\langle\overline{\alpha\beta}\|d_E-1,\alpha;d_E-1,\beta\rangle\rangle}_{1/\sqrt{d_E}}\|\overline{\alpha\beta}\rangle\rangle$$
$$- \sum_{k=1}^{d_E-2} \langle\langle e_k^{\alpha\beta}\|d_E-1,\alpha;d_E-1,\beta\rangle\rangle\|e_k^{\alpha\beta}\rangle\rangle. \tag{A21}$$

The decomposition of the subspaces $(\mathcal{H}^2)_{P_D=1}$ and $(\mathcal{H}^2)_{P_D=0}$, induced by the outlined procedure, looks as shown in Fig. 2.

[1] See, e.g., D. N. Zubarev, *Nonequilibrium Statistical Thermodynamics* (Consultants Bureau, New York, 1974), and references therein.

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[24] For instance, the space of linear operators acting on $\mathcal{H}$ is $\mathcal{H}\otimes\mathcal{H}^{+}$, where $\mathcal{H}^{+}$ is the dual space of $\mathcal{H}$; however, $\mathcal{H}\otimes\mathcal{H}^{+}$ is isomorphic to $\mathcal{H}^{2}=\mathcal{H}\otimes\mathcal{H}$ in a finite-dimensional case, and we choose $\mathcal{H}^{2}$ as the shortest label.
[25] Though our primary interest will lie among the elements of $\mathcal{H}_E^2$, $\mathcal{H}_S^2$, and $\mathcal{H}^2$, we did start with bases in $\mathcal{H}_E$, $\mathcal{H}_S$, and $\mathcal{H}$, as the partial traces, which we will be using later on, are over states in $\mathcal{H}_E$, $\mathcal{H}_S$, and $\mathcal{H}$.
[26] W. E. Boyce and R. C. DiPrima, *Elementary Differential Equations and Boundary Value Problems*, 4th ed. (Wiley, New York, 1986), p. 327.
[27] This choice of a density matrix has interesting properties itself; namely, it corresponds to the most chaotic ensemble, in the sense that the probabilistic entropy $S=-kT\Sigma_i\,\rho_l\ln\rho_l$ is maximal in the case where all weights are equal.
[28] Orthonormal with respect to the standard scalar product inherited from $\mathcal{H}$: namely, for $x,y\epsilon\mathcal{H}^2$, $\langle\langle x\|y\rangle\rangle=\mathrm{Tr}(x^{+}y)$.
[29] W. E. Boyce and R. C. DiPrima, *Elementary Differential Equations and Boundary Value Problems* (Ref. [26]), p. 340.
[30] Interested readers are referred to the popular text: I. Prigogine, *From Being to Becoming* (Freeman, San Francisco, 1980).
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