Stochastic unraveling of positive quantum dynamics

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Stochastic unravelings represent a useful tool to describe the dynamics of open quantum systems, and standard methods, such as quantum state diffusion (QSD), call for the complete positivity of the open-system dynamics. Here, we present a generalization of QSD, which also applies to positive, but not completely positive evolutions. The rate and the action of the diffusive processes involved in the unraveling are obtained by applying a proper transformation to the operators which define the master equation. The unraveling is first defined for semigroup dynamics and then extended to a definite class of time-dependent generators. We test our approach on a prototypical model for the description of exciton transfer, keeping track of relevant phenomena, which are instead disregarded within the standard, completely positive framework.

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I. INTRODUCTION

The investigation of open quantum systems coupled to complex and possibly structured environments has led to a renewed interest in the description of quantum dynamics beyond the paradigm of completely positive (CP) semigroups [1–3], as fixed by the well-known Gorini-Kossakowski-Sudarshan-Lindblad (GKSL) master equation (ME) [4,5]. The development of more general approaches has made it possible to take into account memory effects and others phenomena which are neglected within that framework; see, for example, the recent reviews [6–8].

Mostly, the assumption to have a semigroup dynamics is relaxed, while one holds firm that the evolution has to be given by CP maps. If there are no initial correlations between the system and the environment and the initial state of the latter is fixed, the exact reduced dynamics, mathematically obtained via the partial trace on the environmental degrees of freedom, is indeed CP [1–3]. On the other hand, the partial trace can hardly ever be performed explicitly, even with powerful numerical techniques. The restriction to CP maps, then, becomes questionable, not only when initial correlations have to be considered [9–12], but also when one uses an approximated description for specific open quantum systems at hand. The weaker condition that the dynamics is positive (P) may be enough to guarantee the consistency of the predictions one is interested in.

In addition, when a ME is derived from some underlying microscopic model, CP is usually obtained by introducing some specific approximations, which, needless to say, may overlook some relevant phenomena. As a paradigmatic example, in the weak-coupling regime, one imposes (on top of the Born-Markov approximation) the secular approximation. The latter is justified when the free dynamics of the system is much faster than its relaxation [1], which is not the case for several systems of interest. Nonsecular non-CP evolutions, possibly still in the semigroup regime, are extensively used, e.g., to model transport phenomena in nanoscale biomolecular networks [13–16].

Certainly, CP evolutions possess several advantages, mainly due to the general mathematical results which allow for their full characterization, such as the Kraus decomposition or the GKSL theorem itself [1]. Moreover, CP evolutions have been equivalently formulated in terms of unravelings in the form of stochastic trajectories, with jumps [17,18] or continuous [19–24]. These methods yield a very powerful tool to simulate numerically open-system dynamics, as well as a deeper understanding of the different effects induced on the system by the interaction with the environment, as in the theory of continuous measurement (see [25] and references therein). Also, unravelings of MEs play a role in the foundations of quantum mechanics, in connection with decoherent histories [26,27] and quantum state reduction theories [28–31].

Here, we prove that a proper unraveling can be generally formulated also for P, not necessarily CP, dynamics. We focus in particular on a continuous form of the unraveling, the so-called quantum state diffusion (QSD) [19–23], and we show how it can be directly extended to the more general case of P dynamics. The role of the rates and Lindblad operators in the CP unraveling is replaced by the eigenvalues and eigenvectors of a rate operator [19,21,32,33]. Our approach includes not only semigroup dynamics, but also a more general kind of evolution: namely, P-divisible dynamics [34–38], which has been recently taken into account within the context of the definition of quantum Markovianity. In this way, we provide a significant class of open-system dynamics with a useful tool to describe physical phenomena, which would be neglected within the usual CP framework. This is explicitly shown by taking into account a model, which is of interest for the description of energy transfer in biomolecular networks [15,16,39].

The rest of the paper is organized as follows. In Sec. II, we briefly recall the standard QSD unravelling of CP semigroups. In Sec. III, we introduce the QSD unraveling of P semigroups, which is then further extended to P-divisible maps in Sec. IV. In Sec. V, we present two examples of P non-CP dynamics,

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to which we apply our formalism; the first is a simple toy model for a qubit evolution, while the second is a significant model for the excitation transfer in dimeric systems. Finally, the conclusions and future perspectives are given in Sec. VI.

II. UNRAVELING OF CP SEMIGROUPS

Let us first briefly recall the standard results about (diffusive) unravelings of CP semigroups, as well as the relevant notation.

We consider a finite-dimensional quantum system, whose state ρ is an element of the set $\mathcal{S}(\mathbb{C}^n)$ of positive trace-one operators on $\mathbb{C}^n$. The dynamics is described by a one-parameter family of linear maps $\{\Lambda_t\}_{t\geqslant 0}$, where $\Lambda_t : \mathcal{S}(\mathbb{C}^n) \to \mathcal{S}(\mathbb{C}^n)$ evolves the state ρ at the initial time $t_0 = 0$ into the state $\rho_t = \Lambda_t[\rho]$ at time t. These maps satisfy the semigroup property whenever $\Lambda_t \Lambda_s = \Lambda_{t+s}, \forall t,s \geqslant 0$, and in this case they can be expressed as $\Lambda_t = e^{t\mathcal{G}}$ for some *generator* $\mathcal{G}$, so that ρ_t is fixed by the ME $d\rho_t/dt = \mathcal{G}[\rho_t]$. The maps Λ_t ensure the trace and Hermiticity preservation of the system's state ρ_t if and only if the generator $\mathcal{G}$ can be written as [4]

$$\mathcal{G}[\rho] := -i[H,\rho] + \sum_{j=1}^{n^2-1} c_j \left[L_j \rho L_j^\dagger - \frac{1}{2}\{L_j^\dagger L_j, \rho\}\right], \quad (1)$$

for some coefficients $c_j \in \mathbb{R}$, linear operators L_j, and a Hermitian operator $H = H^\dagger$. According to the GKSL theorem [4,5], the maps Λ_t generated by $\mathcal{G}$ are CP if and only if $c_j \geqslant 0\ \forall j$.

In addition to the GKSL theorem, another crucial feature of CP semigroups, further motivating their ubiquitous use to describe open-system's dynamics, is that they can be equivalently characterized via *unravelings*. An unraveling consists of a stochastic dynamics for the pure states $|\psi\rangle$ of the system, which reproduces the ME under stochastic average. Here, we focus on the case of a *diffusive* unraveling, associated to a stochastic differential equation (SDE) in the form [19–24]

$$|d\psi_t\rangle = A_{\psi_t}|\psi_t\rangle dt + \sum_{k=1}^{m} B_{\psi_t,k}|\psi_t\rangle d\xi_{k,t}, \quad (2)$$

where $A_{\psi_t}, B_{\psi_t,k}$ are (possibly nonlinear) operators and $\xi_{k,t}$ are independent complex-valued Wiener processes, with $\mathbb{E}[d\xi_{j,t} d\xi^*_{k,t}] = \delta_{jk} dt$, $\mathbb{E}[d\xi_{j,t} d\xi_{k,t}] = \mathbb{E}[d\xi_{j,t}] = 0$, where $\mathbb{E}$ denotes the statistical mean. The resulting trajectories in the Hilbert space are usually referred to as *quantum trajectories*. We always assume that the SDE preserves the norm of $|\psi_t\rangle$.

The connection with the statistical operator ρ_t is obtained via the stochastic average $\mathbb{E}$. Given the stochastic projector $P_t := |\psi_t\rangle\langle\psi_t|$ and its infinitesimal change dP_t fixed by the Itô formula, $dP_t = |d\psi_t\rangle\langle\psi_t| + |\psi_t\rangle\langle d\psi_t| + |d\psi_t\rangle\langle d\psi_t|$, one says that Eq. (2) is an unraveling of Eq. (1) when $\mathcal{G}[\rho_t] = \mathbb{E}[dP_t/dt]$. In general, there exist infinite unravelings for the same ME. In the case of CP semigroups, the QSD unraveling is given by [19–23] Eq. (2), with $m = n^2 - 1$ and

$$A_{\psi_t} = -iH - \frac{1}{2}\sum_{j=1}^{n^2-1} c_j (L_j^\dagger L_j - 2\ell^*_{\psi,j} L_j + |\ell_{\psi,j}|^2), \quad (3)$$

$$B_{\psi_t,j} = \sqrt{c_j}(L_j - \ell_{\psi,j}), \quad (4)$$

where $\ell_{\psi,j} := \langle\psi|L_j|\psi\rangle$.

III. UNRAVELING OF P SEMIGROUPS

A. Constructive proof of the unraveling

The previous approach can be extended to all the P, not necessarily CP, semigroups (for Hilbert spaces of arbitrary finite dimension). As long as we assume a semigroup evolution, P dynamics provide us with the largest class of dynamics that can have a norm-preserving unraveling for any initial condition: any state obtained via the statistical average is automatically positive, being the convex mixture of pure states. Later, we will see how the semigroup assumption can be replaced by a more general feature of the dynamics.

The unraveling of a P semigroup depends on the behavior of a nonlinear operator, whose relevance for the unraveling of semigroups was already noticed in [19,21,32,33]. Consider a generator as in Eq. (1); for any normalized vector $\psi \in \mathbb{C}^n$, we define the *generalized transition rate operator* (GTRO) as the linear combination [21,32,33]

$$W_\psi := \sum_{j=1}^{n^2-1} c_j (L_j - \ell_{\psi,j})|\psi\rangle\langle\psi|(L_j - \ell_{\psi,j})^\dagger. \quad (5)$$

The precise connection among the properties of this nonlinear operator and the unraveling of P semigroups traces back to the following result, which is a direct consequence of a theorem by Kossakowski [40,41].

Lemma 1. The dynamical map $\Lambda_t = e^{t\mathcal{G}}$ is P if and only if, for any normalized vector $\psi \in \mathbb{C}^n$, W_ψ is a positive semidefinite operator.

Proof. As noticed in [35], the aforementioned Kossakowski's theorem [40,41] can be rephrased as follows: given any orthonormal basis $\{|u_i\rangle\}_{i=1,\ldots,n}$,

$$(\rho \geqslant 0 \Rightarrow \Lambda_t[\rho] \geqslant 0) \Leftrightarrow \sum_{j=1}^{n^2-1} c_j |\langle u_i|L_j|u_{i'}\rangle|^2 \geqslant 0, \quad (6)$$

for any couple $i \neq i'$.

Let us consider two arbitrary states $|\psi\rangle, |\varphi\rangle$. Write

$$|\varphi\rangle = a|\psi\rangle + b|\psi_\perp\rangle, \quad (7)$$

where the two vectors on the right-hand side (rhs) are the components of $|\varphi\rangle$, which are parallel and perpendicular to $|\psi\rangle$, respectively. Notice the relations

$$\langle\psi|(L_j - \ell_{\psi,j})|\psi\rangle = 0, \quad (8)$$

$$\langle\psi|(L_j - \ell_{\psi,j})|\psi_\perp\rangle = \langle\psi|L_j|\psi_\perp\rangle. \quad (9)$$

Then, using Eq. (8) and Eq. (9), we obtain, for any ψ, the equivalence

$$\langle\varphi|W_\psi|\varphi\rangle = |b|^2 \sum_{j=1}^{n^2-1} c_j |\langle\psi|L_j|\psi_\perp\rangle|^2 \quad \forall |\psi\rangle, |\varphi\rangle. \quad (10)$$

Given the equation above, the proof of the Lemma is straightforward. On the one hand, the positivity of W_ψ for any $|\psi\rangle$ implies the positivity of the rhs of Eq. (6) for any couple of orthogonal elements of any given basis (just set $|\psi\rangle = |u_i\rangle$ and $|\psi_\perp\rangle = |u_{i'}\rangle$), from which the positivity of the semigroup follows. One the other hand, if Λ_t is P and hence the rhs of

Eq. (6) is positive, the non-negativity of the rhs of Eq. (10) for any $|\varphi\rangle$ and $|\psi\rangle$ and therefore the positive semidefiniteness of W_ψ for any $|\psi\rangle$, directly follows from setting $|u_i\rangle = |\psi\rangle$ and using the decomposition of $|\psi_\perp\rangle$ on the elements of the basis $|u_{i'}\rangle$ orthogonal to $|u_i\rangle$, i.e., with $i \neq i'$. ■

This result will be the building block of the construction of our unraveling of P semigroups. Some diffusive unravelings which can be applied beyond CP semigroups already appeared in the literature, i.e., for a qubit in [42] and for any finite-dimensional system in [33]. Nevertheless, let us stress how the definite connection between the possibility to formulate an unraveling and the positivity of the corresponding semigroup dynamics for any finite dimension was missing until now; see also the recent discussion in [43].

Proceeding further, Lemma 1 implies that when we have a semigroup of P maps and we consider the linear operator W_ψ for any fixed ψ, its eigenvalues $\lambda_{\psi,i}$ $(i = 0, \ldots, n-1)$ are non-negative, where $\lambda_{\psi,0} = 0$ corresponds to the eigenvector $|\psi\rangle$, so that we can write the spectral decomposition as

$$W_\psi = \sum_{i=1}^{n-1} \lambda_{\psi,i} |\phi_{\psi,i}\rangle\langle\phi_{\psi,i}| = \sum_{i=1}^{n-1} \lambda_{\psi,i} (V_{\psi,i}|\psi\rangle\langle\psi|V_{\psi,i}^\dagger), \tag{11}$$

with $\lambda_{\psi,i} \geqslant 0$ and $|\phi_{\psi,i}\rangle\langle\phi_{\psi,i}|$ the corresponding orthogonal projectors, satisfying $\langle\phi_{\psi,i}|\psi\rangle = 0$. The second equivalence in Eq. (11) is trivially justified by defining $V_{\psi,i} = |\phi_{\psi,i}\rangle\langle\psi|$, which will also provide us with a clear physical interpretation of the unraveling.

Now, by using Itô calculus, it is readily verified that Eq. (2) yields the following SDE for $P_t = |\psi_t\rangle\langle\psi_t|$:

$$dP_t = \left(A_{\psi_t} P_t + P_t A_{\psi_t}^\dagger + \sum_{k=1}^m B_{\psi_t,k} P_t B_{\psi_t,k}^\dagger \right) dt + \sum_{k=1}^m (B_{\psi_t,k} P_t d\xi_{k,t} + P_t B_{\psi_t,k}^\dagger d\xi_{k,t}^*). \tag{12}$$

In addition, since we want the SDE to be an unraveling of the ME fixed by $\mathcal{G}$ at any time t, we are assuming, in particular, that this is the case at time $t = 0$, i.e., $\mathbb{E}[dP_t/dt|_{t=0}] = \mathcal{G}[\rho_0]$. From this relation, along with Eq. (12), it follows that the noise term $\sum_{k=1}^m B_{\psi,k} P B_{\psi,k}^\dagger$ is given by the component of $\mathcal{G}[P]$ orthogonal to $|\psi\rangle$, i.e.,

$$\sum_{k=1}^m B_{\psi,k} P B_{\psi,k}^\dagger = (I - P)\mathcal{G}[P](I - P). \tag{13}$$

The last statement, which was first shown in [19], can be easily rederived consistently with our notation, as shown in the the following. First, let us take the expectation of Eq. (12) for a deterministic initial condition, $|\psi_0\rangle =: |\psi\rangle$, so that $\rho_0 = P_0 =: P$; since $\mathbb{E}[dP_t/dt|_{t=0}] = \mathcal{G}[\rho_0]$, we get

$$A_\psi P + P A_\psi^\dagger + \sum_{k=1}^m B_{\psi,k} P B_{\psi,k}^\dagger = \mathcal{G}[P]. \tag{14}$$

The SDE in Eq. (2) preserves the norm of the state vector only if

$$\langle\psi|B_{\psi,k}|\psi\rangle = 0 \quad \forall \psi, k. \tag{15}$$

Then, if we denote by $|\psi_\perp\rangle$ a vector orthogonal to $|\psi\rangle$, the norm constraint translates into $B_{\psi,k}|\psi\rangle = |\psi_\perp\rangle$. In other words, the noise operators must produce orthogonal changes to the state vector they act upon. For any fixed $|\psi\rangle$, this condition implies

$$P\left(\sum_{k=1}^m B_{\psi,k} P B_{\psi,k}^\dagger \right) P = 0; \tag{16}$$

on the other hand,

$$(\mathbb{I} - P)(A_\psi P + P A_\psi^\dagger)(\mathbb{I} - P) = 0, \tag{17}$$

so that by projecting Eq. (14) on the subspace orthogonal to $|\psi\rangle$, Eq. (16) together with Eq. (17) prove the validity of Eq. (13).

Now, with the help of simple algebra, Eq. (13) reduces to

$$\sum_{k=1}^m B_{\psi,k} P B_{\psi,k}^\dagger = W_\psi. \tag{18}$$

We can conclude that Eq. (18) has to be satisfied by *all* possible (norm-preserving) unravelings, as in Eq. (2), of the ME fixed by Eq. (1).

Moreover, Eq. (18), along with Eq. (14), imply that the action of the drift operator A_ψ on the state $|\psi\rangle$ is determined by W_ψ and the generator $\mathcal{G}$ via

$$A_\psi P + P A_\psi^\dagger = \mathcal{G}[P] - W_\psi. \tag{19}$$

This means that A_ψ can be set independently from the specific solution of Eq. (18) for the $B_{\psi,k}$ and, in particular, A_ψ is still fixed by Eq. (3). Let us emphasize that, indeed, this is not the only nonlinear operator satisfying Eq. (19), but any other solution $\tilde{A}_\psi$ would act on the state ψ exactly in the same way, such that

$$\tilde{A}_{\psi_t}|\psi_t\rangle = A_{\psi_t}|\psi_t\rangle; \tag{20}$$

in other words, it would lead exactly to the same unraveling [see Eq. (2)]: in this regard, the choice of A_ψ, for fixed $B_{\psi,k}$ and noise $\xi_{k,t}$, is *unique*. To prove the validity of Eq. (20), consider two different solutions, A_ψ and $\tilde{A}_\psi$, to Eq. (19). Then, $A_\psi P + P A_\psi^\dagger = \tilde{A}_\psi P + P \tilde{A}_\psi^\dagger$. Hence, for any state $|\psi_\perp\rangle$ orthogonal to $|\psi\rangle$, one has

$$\langle\psi_\perp|A_\psi|\psi\rangle = \langle\psi_\perp|\tilde{A}_\psi|\psi\rangle$$

and, analogously for the parallel component,

$$\mathrm{Re}[\langle\psi|A_\psi|\psi\rangle] = \mathrm{Re}[\langle\psi|\tilde{A}_\psi|\psi\rangle].$$

In principle, $A_{\psi_t}|\psi_t\rangle$ and $\tilde{A}_{\psi_t}|\psi_t\rangle$ could differ by a purely imaginary component parallel to $|\psi_t\rangle$; but it is then easy to see [31] that such a difference corresponds simply to an irrelevant global phase applied to $|\psi_t\rangle$.

All in all, to define a proper unraveling of a P semigroup, we are simply left with formulating a solution of Eq. (18). A natural choice is given by the spectral decomposition of W_ψ, which, by virtue of the positivity of the semigroup and then Lemma 1, is characterized by the non-negative eigenvalues $\lambda_{\psi,k}$. Hence, let us set $m = n - 1$ and

$$B_{\psi,k} = \sqrt{\lambda_{\psi,k}} V_{\psi,k}. \tag{21}$$

It is then easy to see that $B_{\psi,k}$ as in Eq. (21) satisfies Eq. (18) and, along with A_ψ as in Eq. (3), defines a SDE

as in Eq. (2), which provides us with a proper unraveling of the P semigroup generated by Eq. (1). We thus arrive at the wanted result: Eqs. (3) and (21) generalize the QSD unraveling of CP semigroups to the case of P, not necessarily CP, semigroups. Note that for $c_j \geqslant 0$, a solution to Eq. (18) is directly provided by $m = n^2 - 1$ and $B_{\psi,k} = \sqrt{c_k}(L_k - \ell_{\psi,k})$, so that one recovers Eq. (4). On the other hand, when some c_j takes on a negative value (as in the P, non-CP case), a solution of Eq. (18) as in Eq. (4) would give a set of SDEs as in Eq. (2), which are not consistent with the average dynamics: by deriving the stochastic MEs through $P_t := |\psi_t\rangle\langle\psi_t|$, one would get the positive coefficients $|c_j|$.

The crucial point for extending the unraveling to every P semigroup is the observation that the role of the rates c_j can be replaced by the eigenvalues $\lambda_{\psi,i}$ in the spectral decomposition (11) of W_ψ, whose positivity is ensured by Lemma 1. Accordingly, the operators $V_{\psi,i}$ replace the Lindblad operators L_j (of course, $\langle\psi|V_{\psi,i}|\psi\rangle = 0$). The physical meaning of the unraveling defined here is hence quite clear: the eigenvalues and eigenvectors of the GTRO set, respectively, the strength of the diffusive processes and how they act on the elements of the Hilbert space. In particular, $V_{\psi_t,i}$ maps the stochastic state at time t, $|\psi_t\rangle$, into the state $|\phi_{\psi_t,i}\rangle$, which appears in the spectral decomposition of W_{ψ_t} and is orthogonal to $|\psi_t\rangle$. To deal with P, but not CP semigroups, we exploit the diagonalization of the GTRO, while in the CP case the coefficients and operators in the Lindblad generator directly fix the quantum trajectories; the difference between the two cases will be illustrated later for a specific example.

B. CP and norm preservation

As a further remark, we note how the previous results imply that, indeed, the requirement of getting a closed ME from a diffusive norm-preserving SDE does not imply, by itself, CP. This was shown by direct counterexample in [44] (see also Sec. V A) and it can be easily related to the lack of norm preservation of the ME unraveling extended to an arbitrary ancilla.

To see this, let us recall that a linear map $\Lambda : \mathcal{S}(\mathbb{C}^n) \to \mathcal{S}(\mathbb{C}^n)$ is CP if and only if the map $\Lambda \otimes \mathbb{I} : \mathcal{S}(\mathbb{C}^n \otimes \mathbb{C}^n) \to \mathcal{S}(\mathbb{C}^n \otimes \mathbb{C}^n)$ is P. Let $\mathcal{G}$ and $\mathcal{G}'$ be the generators of Λ and $\Lambda \otimes \mathbb{I}$, respectively. Assume Λ (at least) P, and call $d\psi$ the norm-preserving unraveling of its generator. Moreover, we define $d\psi'$ to be a particular extension of the original SDE to an enlarged Hilbert space, such that it reproduces, on average, $\mathcal{G}'$. Then, we are led to the following diagram:

$$\begin{array}{ccc} d\psi & \longrightarrow & d\psi' \\ \uparrow\downarrow & & \uparrow\downarrow \\ \mathcal{G}[\rho] & \longrightarrow & \mathcal{G}'[\rho'] \end{array} \tag{22}$$

where the vertical arrows represent the operations of unraveling and taking the stochastic average, and the horizontal ones stand for tensoring with auxiliary operators, in such a way that the diagram commutes.

Now let us assume that Λ_t is not CP. Then, there exist $\rho' \in \mathcal{S}(\mathbb{C}^n \otimes \mathbb{C}^n)$ such that $\bar{\rho} = (\Lambda_t \otimes \mathbb{I})[\rho'] = \mathbb{E}[P'_t]$, where $P'_t = |\psi'_t\rangle\langle\psi'_t|$, is not a proper quantum state, i.e., $\bar{\rho}$ is either not positive, not trace one, or both. However, any operator obtained via stochastic average is positive, being the convex combination of the positive operators P'_t. Then, if $\Lambda \otimes \mathbb{I}$ is not P, it must be the case that $\mathrm{Tr}(\mathbb{E}[P'_t]) = \mathbb{E}[\mathrm{Tr}(P'_t)] \neq 1$, i.e., $|d\psi'\rangle$ does not preserve the norm of all state vectors. In summary, under the hypothesis that diagram (22) commutes, asking that the extended SDE be norm preserving is a sufficient condition for the CP of Λ.

IV. UNRAVELING OF P-DIVISIBLE DYNAMICS AND RELATION WITH MARKOVIANITY

Our approach can be straightforwardly generalized to a much wider class of dynamics, which goes beyond the class that can be treated via the usual unravelings for CP maps. We consider now evolutions where the coefficients, and possibly the operators, in the ME depend on time. This allows one to describe several situations of interest, where the semigroup approximation cannot be used, because time inhomogeneous and non-Markovian effects become relevant [6–8].

Consider a *time-dependent* generator $\mathcal{G}_t$. Once again, trace and Hermicity preservation constrain it to have the form as in Eq. (1), at any time t, i.e., one has

$$\mathcal{G}[\rho] := -i[H(t),\rho] + \sum_{j=1}^{n^2-1} c_j(t)\left[L_j(t)\rho L_j(t)^\dagger - \frac{1}{2}\{L_j(t)^\dagger L_j(t),\rho\}\right], \tag{23}$$

where now we have a time-dependent Hamiltonian as well as time-dependent rates and Lindblad operators. Note that the CP of the dynamics does not imply the positivity of the coefficients (since, indeed, the GKSL theorem does not apply). The most general conditions to guarantee CP, not to mention P, of the resulting dynamical maps $\Lambda_t = \mathcal{T}\exp(\int_0^t \mathcal{G}_s ds)$ (with $\mathcal{T}$ the time-ordering operator) are actually not known. Nevertheless, the positivity in time of the coefficients, $c_j(t) \geqslant 0$, guarantees that the dynamics is CP and can be decomposed into intermediate CP maps [3,45]: for any $t \geqslant s \geqslant 0$, there is a CP map $\Lambda_{t,s}$ such that

$$\Lambda_t = \Lambda_{t,s} \circ \Lambda_s. \tag{24}$$

In this case, the dynamics is said to be *CP divisible* and this property has been identified with the *Markovianity* of the quantum dynamics in [46]. Note that the positivity of the coefficients allows one to extend the QSD unraveling of CP semigroups to this case: one has simply to replace $c_j \to c_j(t)$, $L_j \to L_j(t)$, and $H \to H(t)$ in Eqs. (3) and (4).

The unraveling defined via Eqs. (3) and (21) can also be extended to ME with time-dependent coefficients, which need not be positive functions of time. Consider any ME leading to a dynamics which, instead of being CP divisible, is *P divisible*, which means that the decomposition in Eq. (24) still applies, but now we make the weaker requirement that the maps $\Lambda_{t,s}$ are P [34]; this property, in turn, has been identified with quantum Markovianity in [35]. The construction presented before can be immediately generalized to this situation since the equivalence in Lemma 1 still applies. Indeed, the extension of Lemma 1 to the case of P-divisible dynamics directly follows from the

analogous extension of the theorem by Kossakowski, pointed out in [35]. To see this, let us consider a ME as in Eq. (23). The resulting dynamical map Λ_t is P and can be decomposed via Eq. (24) with P $\Lambda_{t,s}$ if and only if

$$\sum_{j=1}^{n^2-1} c_j(t)|\langle u_i|L_j(t)|u_{i'}\rangle|^2 \geqslant 0, \tag{25}$$

for any couple $i \neq i'$ [35]. But then, similarly to the proof for the semigroup case, one can show that the latter condition is equivalent to the positivity of W_ψ, defined as in Eq. (5), with the replacements $c_j \to c_j(t)$ and $L_j \to L_j(t)$, so that Eqs. (3) and (21), with the proper introduction of time dependence, define a valid unraveling of a generic P-divisible ME.

Of course, there are several open-system dynamics which are not P divisible and, therefore, cannot be unraveled via our approach, but where other diffusive [47,48] or jump [49] techniques can be exploited. On the other hand, our approach yields a direct generalization of the construction for the semigroup case, without calling for hierarchical equations or for correlations between different trajectories, which are instead usually required by the above-mentioned techniques.

As a final remark, we note that Eqs. (2), (3), and (21) comprise the most general (Markovian) dynamics of collapse models [28–31]. Here, it suffices to say that collapse models consist of a modification of the Schrödinger equation with the addition of nonlinear stochastic terms, which ensure the localization of the wave function. The dynamics of collapse models is usually defined as a diffusion process in the Hilbert space [given by a SDE as in Eq. (2)], although piecewise evolutions involving jumps processes are also possible [28]. If we limit to a dynamics as in Eq. (2), the requirement of getting a closed linear average description, which is physically motivated by the request of no superluminal signaling [50,51], is not enough to guarantee the CP. As said, this traces back to the possible lack of norm preservation of the SDE trivially extended to an arbitrary ancilla. Such an extension, indeed, would be rather unmotivated for collapse models, since the collapsing field would act also on the ancilla, possibly in a nonlocal way: in this context, the CP of the ensemble dynamics is an extra assumption, not emerging from fundamental requirements.

V. EXAMPLES

A. Unraveling of a non-CP qubit ME

Here, we consider the unraveling of the non-CP semigroup which was first derived in [42,44]. Although the physical relevance of the model is not clear, it is the first example of an unraveling of a P, but non-CP semigroup and it was thus used in [44] to prove that the CP of the average dynamics is not guaranteed by the existence of a Markovian unraveling. We will show now how such a result can be straightforwardly rederived and further clarified using our method.

Hence, consider the non-CP semigroup acting on $\mathcal{S}(\mathbb{C}^2)$ and generated by

$$\frac{d\rho_t}{dt} = \sum_{j=1}^{3} c_j(\sigma_j\rho_t\sigma_j - \rho_t), \quad c_1 = c_2 = -c_3 = 1, \tag{26}$$

where σ_j are the usual Pauli matrices $\sigma_1 \equiv \sigma_x$, $\sigma_2 \equiv \sigma_y$, and $\sigma_3 \equiv \sigma_z$. The GTRO associated to Eq. (26) is

$$W_\psi = \sum_{j=1}^{3} c_j(\sigma_j - s_j)|\psi\rangle\langle\psi|(\sigma_j - s_j), \tag{27}$$

with $s_j := \langle\psi|\sigma_j|\psi\rangle$, and it has spectral decomposition

$$W_\psi = \lambda_1|\psi\rangle\langle\psi| + \lambda_2|\psi_\perp\rangle\langle\psi_\perp|,$$

where the eigenvalues are $\lambda_1 = 0$ and $\lambda_2 = 2s_3^2$, while the eigenvectors are $|\psi\rangle$ and $|\psi_\perp\rangle$ orthogonal to $|\psi\rangle$. The first eigenvalue and eigenvector can be easily found by noticing that $W_\psi|\psi\rangle = 0$, as Eq. (8) ensures [see also the discussion before Eq. (11)]. Then, we are left with verifying that

$$\begin{aligned}\big(W_\psi - 2s_3^2\big)|\psi_\perp\rangle &= \sum_{j=1}^{3} c_j\big(\sigma_j P\sigma_j - s_j P\sigma_j - 2s_3^2\big)|\psi_\perp\rangle \\ &= 0.\end{aligned} \tag{28}$$

To show that, we project Eq. (28) on the basis vectors $\langle\psi|$ and $\langle\psi_\perp|$, respectively. Since, for any j,

$$\begin{aligned}\langle\psi|\sigma_j P\sigma_j|\psi_\perp\rangle &= \langle\psi|\sigma_j|\psi\rangle\langle\psi|\sigma_j|\psi_\perp\rangle \\ &= \langle\psi|s_j|\psi\rangle\langle\psi|\sigma_j|\psi_\perp\rangle \\ &= \langle\psi|s_j P\sigma_j|\psi_\perp\rangle,\end{aligned}$$

we have

$$\begin{aligned}\langle\psi|\big(W_\psi - 2s_3^2\big)|\psi_\perp\rangle &= \sum_{j=1}^{3} c_j\langle\psi|(\sigma_j P\sigma_j - s_j P\sigma_j)|\psi_\perp\rangle \\ &= 0.\end{aligned} \tag{29}$$

On the other hand, since $\sum_{j=1}^{3} s_j^2 = 1$ and $r_j = |\langle\psi|\sigma_j|\psi_\perp\rangle|^2 = 1 - s_j^2$, we have

$$\begin{aligned}\langle\psi_\perp|\big(W_\psi - 2s_3^2\big)|\psi_\perp\rangle &= \sum_{j=1}^{3} c_j\langle\psi_\perp|\sigma_j P\sigma_j|\psi_\perp\rangle - 2s_3^2 \\ &= \sum_j c_j r_j - 2s_3^2 = 0.\end{aligned} \tag{30}$$

Equation (29) together with Eq. (30) prove Eq. (28), which implies $W_\psi = 2s_3^2|\psi_\perp\rangle\langle\psi_\perp|$.

Then, according to Eqs. (11) and (3), the noise and the drift terms which define an unraveling of Eq. (26) are given, for any $|\psi\rangle$, by

$$\begin{aligned}B_{\psi_t} &= \sqrt{2}s_3|\psi_{t\perp}\rangle\langle\psi_t|, \\ A_{\psi_t} &= -iH - \frac{1}{2}\sum_{j=1}^{3} c_j(\sigma_j - s_j)^2,\end{aligned}$$

so that

$$|d\psi_t\rangle = -iH - \frac{1}{2}\sum_{j=1}^{3}(\sigma_j - s_j)^2|\psi_t\rangle dt + \sqrt{2}\,s_3|\psi_{t\perp}\rangle d\xi_{j,t}. \tag{31}$$

Clearly, one can verify that Eq. (31) is norm preserving ($\langle\psi|d\psi\rangle + \langle d\psi|\psi\rangle + \langle d\psi|d\psi\rangle = 0$) and generates, on average, the ME in Eq. (26).

B. The Bloch-Redfield equation for a dimer system

As a specific, physically relevant application of our approach, we consider an example given by a simple description of a dimer system, which nevertheless represents a useful model to investigate exciton transfer, for example, in biomolecular complexes [15,16,39].

The state of the excitation is associated with a three-level system: two levels for the excitation being in one or the other site, and one level for the absence of excitation. The most relevant sources of noise are the pure dephasing and the recombination process. Using a perturbative approach (e.g., projection operator techniques) up to second order and the Born-Markov approximation, one gets the Bloch-Redfield equation [1]. This equation usually does not guarantee the positivity of the evolution and it is then further approximated by a Lindblad equation, which even ensures that the dynamics is CP. The Lindblad equation is obtained via the secular approximation (SA), which essentially neglects all the terms coupling population and coherences of the system. However, this approximation is not always justified from a physical point of view, as it calls for a large difference in the time scales of the free evolution and the dissipative relaxation of the system. To overcome this difficulty and retain all the relevant phenomena in the dimer evolution, yet in a semigroup description of the dynamics, a partial SA was introduced in [15]. The latter discards only some terms which couple population and coherences, while it preserves the most relevant ones. The resulting ME implies a P, but, in general, not CP evolution. Hence, it provides us with a natural benchmark to test our method.

The ME, both after the full and the partial SA, can be written as [15]

$$\dot{\rho}_{ij}(t) = \sum_{kl=1}^{3} \mathcal{R}^{\chi}_{ij;kl}\rho_{kl}(t), \tag{32}$$

where $\rho_{kl}(t) = \langle k|\rho(t)|l\rangle$. We will use the notation $\chi = CP$ for the full SA, while $\chi = P$ for the partial SA. In order to write the ME (32) into the Lindblad form as in Eq. (1), let us report the explicit expression of the coefficients $\mathcal{R}^{\chi}_{ij;kl}$. In the case of the partial SA, they are given by (see Eq. (11) in [15])

$$\begin{aligned}
&\mathcal{R}^{\mathrm{P}}_{11,11} = \mathcal{R}^{\mathrm{P}}_{22,22} = \mathcal{R}^{\mathrm{P}}_{33,33}/2 = -4,\\
&\mathcal{R}^{\mathrm{P}}_{11,33} = \mathcal{R}^{\mathrm{P}}_{33,11} = \mathcal{R}^{\mathrm{P}}_{22,33} = \mathcal{R}^{\mathrm{P}}_{33,22} = 4,\\
&\mathcal{R}^{\mathrm{P}}_{11,12} = \mathcal{R}^{\mathrm{P}}_{22,21} = \mathcal{R}^{\mathrm{P}}_{31,32} = \mathcal{R}^{\mathrm{P}}_{32,31} = -71i,\\
&\mathcal{R}^{\mathrm{P}}_{22,12} = \mathcal{R}^{\mathrm{P}}_{11,21} = \mathcal{R}^{\mathrm{P}}_{13,23} = \mathcal{R}^{\mathrm{P}}_{23,13} = 71i,\\
&\mathcal{R}^{\mathrm{P}}_{21,11} = \mathcal{R}^{\mathrm{P}*}_{12,11} = -\mathcal{R}^{\mathrm{P}*}_{12,22} = -\mathcal{R}^{\mathrm{P}}_{21,22} = -1 + 71i,\\
&\mathcal{R}^{\mathrm{P}}_{12,12} = \mathcal{R}^{\mathrm{P}*}_{21,21} = -8 - 46i,\\
&\mathcal{R}^{\mathrm{P}}_{13,13} = \mathcal{R}^{\mathrm{P}*}_{31,31} = -9 + 12210i,\\
&\mathcal{R}^{\mathrm{P}}_{23,23} = \mathcal{R}^{\mathrm{P}*}_{32,32} = -9 + 12256i,
\end{aligned} \tag{33}$$

and all the other coefficients are equal to 0; as one can directly check, this provides us with a P, but not CP evolution. On the other hand, as widely discussed in [15], a CP evolution is obtained with a full SA, which means that the terms coupling populations and coherences are set to 0:

$$\begin{aligned}
&\mathcal{R}^{\mathrm{CP}}_{11,12} = \mathcal{R}^{\mathrm{CP}}_{22,21} = \mathcal{R}^{\mathrm{CP}}_{31,32} = \mathcal{R}^{\mathrm{CP}}_{32,31} = 0,\\
&\mathcal{R}^{\mathrm{CP}}_{22,12} = \mathcal{R}^{\mathrm{CP}}_{11,21} = \mathcal{R}^{\mathrm{CP}}_{13,23} = \mathcal{R}^{\mathrm{CP}}_{23,13} = 0,\\
&\mathcal{R}^{\mathrm{CP}}_{21,11} = \mathcal{R}^{\mathrm{CP}}_{12,11} = \mathcal{R}^{\mathrm{CP}}_{12,22} = \mathcal{R}^{\mathrm{CP}}_{21,22} = 0,
\end{aligned} \tag{34}$$

while all the other coefficients in Eq. (33) are not changed. The parameters appearing in the two MEs express, in units of cm^{-1}, the effect of dephasing and recombination noise, which are modeled as a spatially uncorrelated noise with an ohmic spectrum [15], as well as the Hamiltonian part of the dynamics. In particular, note that the energy difference between the ground state and the two excitonic states is two or three orders of magnitude larger than any other relevant parameter in the free Hamiltonian [52], which explains the appearance of imaginary components in the ME parameters which are much bigger than the other values.

As described in Sec. III, to define our unraveling, we first need to write the Lindblad form of the ME, which can be readily obtained following Ref. [4]. First, note that the generator $\mathcal{G}$ can be directly reconstructed via the coefficients in Eq. (32) since

$$\mathcal{R}^{\chi}_{ij;kl} = \langle i|\mathcal{G}^{\chi}[|k\rangle\langle l|]|j\rangle. \tag{35}$$

Then, consider the basis of operators on $\mathbb{C}^3$ given by $\{\tau_i\}_{i=0,\ldots,8}$, with $\tau_0 = \mathbb{1}/\sqrt{3}$, while the τ_is with $i = 1,\ldots,8$ are the Gell-Mann matrices over $\sqrt{2}$ (to guarantee the normalization with respect to the Hilbert-Schmidt scalar product). Hence, the so-called nondiagonal form of the generator $\mathcal{G}$ is given by

$$\mathcal{G}[\rho] := -i[H,\rho] + \sum_{ij=1}^{8} d_{ij}\left[\tau_i\rho\tau_j^{\dagger} - \frac{1}{2}\{\tau_j^{\dagger}\tau_i,\rho\}\right], \tag{36}$$

with

$$\begin{aligned}
&H = \frac{1}{2i}(\tau^{\dagger} - \tau), \quad \tau = \frac{1}{3}\sum_{i=1,k=0}^{8} \mathrm{Tr}\{\tau_k\tau_i\mathcal{G}[\tau_k]\}\tau_i,\\
&d_{ij} = \sum_{k=0}^{8} \mathrm{Tr}\{\tau_j\tau_k\tau_i\mathcal{G}[\tau_k]\}, \quad i,j = 1,\ldots,8.
\end{aligned} \tag{37}$$

The matrix of coefficients d_{ij} is Hermitian, as the dynamics is Hermiticity preserving; so there is a unitary matrix U, with elements U_{ij}, which diagonalizes it. The resulting coefficients of the diagonal matrix are just the coefficients c_j appearing in the diagonal form of $\mathcal{G}$ in Eq. (1), and the matrix U also defines the corresponding Lindblad operators L_j: explicitly, one has

$$c_j = \sum_{kk'=1}^{8} U^*_{kj}d_{kk'}U_{k'j}, \quad L_j = \sum_{i=1}^{8} U_{ij}\tau_i. \tag{38}$$

For the generator $\mathcal{G}^P$ fixed by Eq. (33), we get the coefficients

$$\begin{aligned}
&c_1 = 2 + \sqrt{5}, \quad c_2 = c_3 = c_4 = c_5 = 4,\\
&c_6 = \tfrac{1}{3}(4 + \sqrt{19}), \quad c_7 = 2 - \sqrt{5}, \quad c_8 = \tfrac{1}{3}(4 - \sqrt{19}),
\end{aligned} \tag{39}$$

where, note, the last two are negative, thus witnessing the non-CP of the resulting semigroup dynamics. The corresponding (canonical) Lindblad operators are

$$L_1 = -f_{1,-}\tau_1 + f_{1,+}\tau_3, \quad L_7 = f_{1,+}\tau_1 + f_{1,-}\tau_3,$$
$$L_2 = \tau_4, \quad L_3 = \tau_5, \quad L_4 = \tau_6, \quad L_5 = \tau_7,$$
$$L_6 = if_{2,-}\tau_2 + f_{2,+}\tau_8, \quad L_8 = -if_{2,+}\tau_2 + f_{2,-}\tau_8,$$
$$f_{1,\pm} = \sqrt{\frac{1}{2} \pm \frac{1}{\sqrt{5}}}, \quad f_{2,\pm} = \sqrt{\frac{1}{2} \pm \frac{2}{\sqrt{19}}}; \tag{40}$$

finally, the Hamiltonian part of the dynamics is given by

$$H = -71\sqrt{2}\tau_1 - \frac{\sqrt{2}}{3}\tau_2 + 23\sqrt{2}\tau_3 - 12233\sqrt{\frac{2}{3}}\tau_8. \tag{41}$$

The unraveling operator A_ψ is hence directly defined by Eq. (3), while $B_{\psi,k}$ is obtained via the evaluation of the GTRO in Eq. (5) and its diagonalization; see Eqs. (11) and (21).

Repeating the same calculations for the generator $\mathcal{G}^{\rm CP}$ fixed by the full SA, i.e., Eq. (34), we directly get a diagonal form of the generator, with (positive) coefficients

$$c_1 = c_2 = c_3 = c_4 = c_5 = 4, \quad c_6 = \tfrac{8}{3}, \tag{42}$$

and Lindblad operators, as well as the Hamiltonian, given by

$$L_1 = \tau_3, \quad L_2 = \tau_4, \quad L_3 = \tau_5, \quad L_4 = \tau_6,$$
$$L_5 = \tau_7, \quad L_6 = \tau_8,$$
$$H = 23\sqrt{2}\tau_3 - 12233\sqrt{\frac{2}{3}}\tau_8. \tag{43}$$

Here, since the dynamics is CP, one could apply the usual formulation of the (diffusive) unraveling, which is directly fixed by Eqs. (3) and (4).

Now, our unraveling proceeds as the usual diffusive unraveling, with the addition that we have to diagonalize the GTRO in order to have the rate and noise operators providing the trajectories. In particular, the algorithm giving each trajectory goes as follows. First, $m = n - 1$ Wiener processes with derivatives $d\xi_{k,t}$, with $k = 0, \ldots, m$ and n the dimension of the Hilbert space, are generated over a computational time domain $[0, \delta t, 2\delta t, \ldots, T]$. Then, given the rates and the operators defining the ME (32) in the P case [see Eqs. (39)–(41)], the expectation values $\ell_{\psi_0,j}$, the drift operators A_{ψ_0}, and the GTRO W_{ψ_0} of Eqs. (3) and (5), respectively, are computed for a given initial state $|\psi_0\rangle$ at time t_0. Next, W_{ψ_0} is diagonalized, the positivity of its eigenvalues is checked (a negative eigenvalue would stop the algorithm), and the noise operators $B_{\psi_0,k}$ are constructed according to Eq. (21). Now, the state $|\psi_1\rangle$ after the first time step δt is computed through the iterative formula $|\psi_1\rangle = \exp[(-iH + A_{\psi_0})\delta t + \sum_{i=1}^{m} B_{\psi_0,i} d\xi_{i,0}]|\psi_0\rangle$ and then normalized. Finally, the state $|\psi_0\rangle$ is updated to $|\psi_1\rangle$ and the algorithm starts over with the evaluation of $\ell_{\psi_1,j}$, A_{ψ_1}, and W_{ψ_1} at time $t_1 = t_0 + \delta t$.

In Figs. 1(a) and 1(b), we report some trajectories for the evolution of the, respectively, first and second site populations, which are obtained by means of the unraveling of the P dynamics after the partial SA [see Eqs. (32) and (33)], thus demonstrating the effectiveness of our approach on a physically relevant model. Let us stress that the traditional unravelings for CP semigroups could not be applied to these

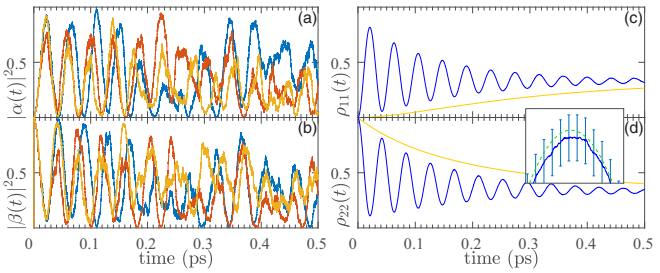

FIG. 1. Trajectories for the evolution of the population of (a) site one and (b) site two; each trajectory corresponds to a different realization of the solution of the SDE in Eq. (2), with A_{ψ_t} as in Eq. (3) and B_{ψ_t} as in Eq. (21), and derived by diagonalizing the GTRO at each point of the computational time domain. The deterministic initial state is $|\psi(0)\rangle = |2\rangle$, while the state at time t is $|\psi(t)\rangle = \alpha(t)|1\rangle + \beta(t)|2\rangle + \gamma(t)|3\rangle$. Evolution of the population of (c) site one and (d) site two given by the ensemble average of 1000 trajectories of our unraveling (blue/dark), and the solution of the Lindblad equation after full SA (yellow/light). In the inset, the ensemble average (blue/dark) and the solution of the P ME (green dotted) are shown to agree within the standard deviation of the mean (vertical bars) of the trajectories. The initial state is set as $\rho(0) = |2\rangle\langle 2|$.

dynamics since they require a Lindblad equation and, thus, in this context, a full SA. Crucially, the latter would cancel any coupling between population and coherences, therefore potentially disregarding some significant phenomena. This is explicitly shown in Figs. 1(c) and 1(d), where we compare the evolution of the populations obtained by solving the Lindblad equation after the full SA and the populations obtained by averaging 1000 trajectories of our unraveling. The former completely neglects significant oscillations [15], which are instead fully captured by the unraveling of the P dynamics.

As a final remark, we note that in the model at hand, the P of the dynamics was guaranteed by itself. On the other hand, if we want to apply our unraveling to a more complex system, starting from a generic ME as in Eq. (1) or in Eq. (23), in which we do not know whether or not it is P (divisible), we can still be sure that as long as the algorithm works, we are not dealing with ill-defined (i.e., nonpositive) states. In other words, any detected phenomenon cannot be traced back to a nonphysical description of the system's statistics. In fact, imagine that, on the contrary, the solution of a given ME maps the state of the system at a certain time t, $\rho(t)$, into a nonpositive state $\rho(t + \delta t)$. If we now look at the unraveling, this means that the GTRO will not be a positive operator, for at least one of the stochastic states at time t giving $\rho(t)$ on average. But then the algorithm will stop due to the appearance of nonpositive rates [see Eq. (11)], witnessing the nonpositivity of the map leading from time t to $t + \delta t$. This is fully analogous to what happens for, e.g., the non-Markovian quantum jumps approach [49], which can be safely applied to any ME, whose CP, or even P, may not be guaranteed.

VI. CONCLUSIONS

We have introduced a continuous unraveling for dynamics which are P, but not necessarily CP. Our approach directly generalizes the QSD method: the rates and operators extracted

from the ME have to be replaced by, respectively, the eigenvalues and eigenvectors of a proper rate operator. We have taken into account the case of semigroup dynamical maps and, additionally, we have extended our result to include a more general class of open-system evolutions, so that our unraveling can be applied to every P-divisible dynamics.

By virtue of the unraveling of P dynamics, one can avoid imposing approximations which could introduce significant errors in the system of interest, such as imposing the secular approximation on top of the weak-coupling approximation. This has been shown explicitly in a case study by investigating the population evolution in a dimer system.

Certainly, our approach can be improved in many regards. A crucial point will be to simplify the task of diagonalizing the GTRO at each time step, e.g., by looking for possible connections between its spectral decompositions at subsequent times. Also, it will be of interest to study how and to what extent the range of applicability of our method can be further extended, for example, combining it with other unraveling techniques [47–49], which apply to general non-Markovian dynamics. Finally, a central question, which is at the moment still open, is whether the unraveling we presented here can be formulated in terms of continuous measurements [24,25].

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