Preparations and weak-field phase control can witness initial correlations

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(Received 7 April 2018; published 19 December 2019)

The dynamics of a system that is correlated with an environment is almost always non-Markovian. Hence, it is important to characterize such correlations experimentally and witness them in physically realistic settings. One such setting is weak-field phase control where control is sought by the shaping of the phase of weak laser pulses. In this paper, we show how weak-field phase controllability can be combined with quantum preparations to witness initial correlations between the system and the environment. Furthermore, we show how this protocol can be applied to almost always predict violations of the quantum regression formula.

DOI: 10.1103/PhysRevA.100.062120

I. INTRODUCTION

Almost all open quantum system evolution is non-Markovian (NM). Our ability to control and manipulate small quantum systems coupled to mesoscopic baths brings into focus the need to understand NM and detect it using simple experimental protocols. In this paper, we demonstrate how quantum control with weak (perturbative) Hamiltonian controls can be used to witness both initial and intermediate correlations. As an example of such weak control, we consider weak-field phase control (WFPC), which is a spectroscopic technique where the phase of weak-shaped laser pulses is used to control the dynamics of quantum systems. Although we will consider WFPC as the prototypical example, we note that any perturbative control Hamiltonian satisfying the stated assumptions can be used to witness non-Markovianity, making this paper relevant to diverse physical systems, such as vacancy centers [1], nano-, and optomechanical systems [2]. Examples of weak-field phase control of open quantum systems include interesting quantum biological systems, such as bacteriorhodopsin [3], and is the relevant regime of control for the dynamics of protein environments in normal functional conditions. Furthermore, WFPC has been shown to be directly influenced by the environment with strong solvent dependence of the stimulated emission [4]. Computational demonstrations of phase control were presented in Refs. [5,6].

We understand the evolution of a quantum system as non-Markovian when the quantum system exhibits memory of past dynamics. Examples of such practical NM can be seen in systems with structured environments [7], quantum biology [8], and nuclear magnetic resonance [9], making it ever more relevant to be able to detect NM in an arbitrary physical system. Several formal definitions of non-Markovianity have been proposed with conceptual similarities and key differences. Definitions of NM have been formulated based on the nondivisibility of maps [10–13] and the backflow of information from the environment [14,15]. These measures are part of a hierarchy of NM [16] and follow from the presence of correlations at intermediate times [17]. Other related definitions of NM involve the presence of initial correlations [18–24]. Such initial correlations result in the backflow of information [25]. Additionally, they violate the assumption of initially factorized states and, hence, lead to the breakdown of several well-known approaches to the dynamics of the reduced state, such as Lindblad dynamics and completely positive trace preserving (CPTP) maps [26]. Hence, it is imperative that we understand how to characterize and control NM in physical systems for future quantum technology applications [27]. Performing tomography on the system-environment state or the marginal postmeasurement system state [28] is the standard way to detect NM, which is tedious for large systems. Furthermore, since tomography of non-Markovian systems is cumbersome [29] and witnessing it through mapping quantum correlations [30] involves performing complicated measurements, it is important to find experimentally scalable methods to witness NM in arbitrary open quantum systems with minimal assumptions about the system.

We begin by discussing the necessary conditions for observing WFPC before relating it to the correlations between the system and the environment. We then present a method to witness initial and intermediate correlations. Finally, we comment on how the witnessing of correlations can be used as a method to detect physical systems that violate the quantum regression formula. Our results, hence, connect an important spectroscopic tool that is of importance to a variety of experimental systems to the problem of detecting and characterizing the nature of NM.

II. MODEL

Consider an open system evolution where a quantum system S is in contact with an environment E. The system is composed of two manifolds, a ground-state manifold and

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an excited-state manifold. The control task is to transfer population from the ground-state manifold to the excited-state manifold. To this end, we define several quantities of interest. First, let P be the projector onto the excited-state manifold. Furthermore, let the initial joint state of the system environment be $R(0)$. The bare Hamiltonian that governs the subsequent evolution is given by H_0 and a control field that is applied only on the system is given by $V(t) \otimes \mathbb{I}$. Without loss of generality, we take the form of the control Hamiltonian ($\hbar = 1$) to be composed entirely of off-diagonal blocks in the energy eigenbasis, namely,

$$V(t) = \begin{pmatrix} 0 & \mu\epsilon(t) \\ \mu\epsilon^*(t) & 0 \end{pmatrix}. \tag{1}$$

Here, μ is a Hermitian operator (such as the dipole moment operator) and $\epsilon(t) = \mathcal{F}[\tilde{\epsilon}(\omega)]$ is a time-dependant field that mediates the external control via the amplitude and phase of $\tilde{\epsilon}(\omega) = \tilde{A}(\omega)e^{i\varphi(\omega)}$. By phase control, we mean that the expectation value of P is controllable by $\varphi(\omega)$. The state of the system environment at a later time is given by a unitary rotation, namely,

$$R(t) = U(t)R(0)U^\dagger(t). \tag{2}$$

Here, $U(t) = \mathcal{T}\exp\left(-i\int_0^t du\{H_0 + V(t)\otimes\mathbb{I}\}\right)$. If the initial system-environment state is factorizable as $R(0) = \rho^{(S)}(0) \otimes \tau^{(E)}(0)$, the subsequent evolution of the marginal states is described as a completely positive trace preserving map acting on the initial marginal state alone.

The central quantity of interest is the rate of population change in the excited-state manifold at time t. Consequently, we can define the population rate in the excited-state manifold as

$$\dot{p}(t) := \mathrm{tr}[P \otimes \mathbb{I}\dot{R}(t)]. \tag{3}$$

If the field is sufficiently weak, the dynamics of the system is well understood by second-order perturbation theory. Following standard literature [31], we can write the evolution equation for the joint state at time t in this perturbative regime as

$$\dot{R}_I(t) = -i[V_I(t), R_I(0)] - \int_0^t du\{V_I(t), [V_I(u), R_I(0)]\}. \tag{4}$$

Here, R_I refers to the system environment state in the interaction picture given by $U_0 R U_0^\dagger = R_I$ with $U_0 = \exp(iH_0 t)$. The calculation of $\dot{p}(t)$ follows by substituting Eq. (4) into Eq. (3) in the appropriate picture.

We focus on WFPC with an eye to inspect the relationship of the projector P, the bare Hamiltonian, and their relation to the initial system-environment state. We begin with a brief summary of the results relating to WFPC before we prove a theorem relating phase control to correlations. Hence, in the next section, we will discuss the different conditions for the presence of weak phase control.

III. CONDITIONS FOR OBSERVING WFPC

We begin with the point of view that we have just observed phase control in a physical system. We want to find out what exactly caused this phase control. In Ref. [32], Am-Shallem and Kosloff produced a no-go theorem that asserted that no weak phase control is observed if a set of conditions are met. Since phase control has been observed, one or more of these conditions must have been violated. A generalized version of these conditions are statable in the following no-go theorem:

Theorem 1. Consider an open quantum system consisting of a ground- and an excited-state manifold. Let such a system be subject to a joint system-environmental unitary operator that depends on phase of a control field $\varphi(t) = \mathcal{F}[\tilde{\varphi}(\omega)]$ through the Hamiltonian given in Eq. (1) with the control objective being the population of the excited-state manifold. Such a quantum system is not phase controllable if: (1) The field is weak enough for second-order perturbation theory to be a good approximation. (2) The free evolution does not excite the system, i.e., $[P \otimes \mathbb{I}, H_0] = 0$. (3) The initial state is invariant under free evolution, i.e., $[H_0, R(0)] = 0$, and (4) stationarity of the bare evolution, defined as the evolution of the system-environment state $R(t_1)$ to $R(t_2)$ only depending on the difference $t_2 - t_1$. Condition (1) ensures that the physics excites only the low-lying energy sectors of the system, a condition important for several experimental scenarios [3] where (say) the molecule under consideration can photodisassociate under strong fields. Condition (2) simply is the statement that the bare evolution should not excite the quantum system to make transitions from the ground-state manifold $|g_i\rangle$ into the excited-state manifold $|e_i\rangle$. Condition (3) asserts that the initial state should commute with the bare Hamiltonian, but we note that, although the off-diagonal terms in the system energy eigenbasis of the type $|g_n\rangle\langle e_m|$ are important for phase control, not all off-diagonal terms in the energy eigenbasis produce phase control. Consider the bare Hamiltonian $H_0 = \omega^{(S)}(n^{(S)} + 1/2) + \sum_k \omega_k^{(E)}(n_k^{(E)} + 1/2) + gn^{(S)}\sum_k x_k^{(E)}$ where the excited state is defined by the projection operator $P = \mathbb{I} - |0\rangle\langle 0|$. Here, $\omega^{(S)}$ is the frequency of the system, and $\omega_k^{(E)}$ represents the frequencies of the environment made of harmonic-oscillator modes. If the initial state of the system environment is a Gibbs state given by $R(0) = \exp(-\beta H_0)/Z_0$, where $Z_0 = \mathrm{tr}\exp(-\beta H_0)$, then all conditions are satisfied, and there is, indeed, no phase control. Note that the absence of phase control survives the initial correlations between the system and the environment, but only because the off-diagonal terms (in the energy eigenbasis of the individual system and environment spaces) of $R(0)$ are all in the environment.

Condition (4) deals with stationarity, which can also be viewed as an extension of condition (2) where we have constrained the bare evolution of the system-environment state. Throughout the paper, we have assumed that the bare evolution of the system environment does not depend on time. This assumption can always be satisfied by sufficiently dilating the environment and is consistent with our results as we have made no assumptions about the dimension or structure of the environmental states in our paper. However, if for practical or theoretical reasons, it is more convenient to consider an environment with fewer degrees of freedom at the cost of introducing some time dependence into the bare Hamiltonian, then our framework would allow for this as long as the free evolution does not excite the system for all time t, i.e., $[P \otimes \mathbb{I}, H_0(t)] = 0$. None of our methods or results would change in this time-dependent regime if we are able to constrain our system to follow the above assumption,

and so without loss of generality, we assume that the bare Hamiltonian H_0 is independent of time.

In our notation, the projector is given by $P = \sum_i |e_i\rangle\langle e_i|$, and the control Hamiltonian can be written as

$$V(t) = \sum_{jk} \mu_{kj}\epsilon(t)|e_j\rangle\langle g_k| + \mu_{jk}\epsilon^*(t)|g_k\rangle\langle e_j|. \quad (5)$$

IV. VIOLATION OF CONDITIONS

To summarize the discussion so far, WFPC can be observed in a chemical reaction for several reasons. The first reason is that second-order perturbation theory is not valid. This is discounted since control fields are accessible to experimental tuning and, hence, can always be made weak.

The second reason is that free evolution operator does not commute with the target operator. For example, consider the bare Hamiltonian $H_0 = \omega^{(S)}(n^{(S)} + 1/2) + \sum_k \omega_k^{(E)}(n_k^{(E)} + 1/2) + gx^{(S)}\sum_k x_k^{(E)}$. Let the system and environment in a Gibbs state at temperature β^{-1}, i.e., $R(0) = \exp(-\beta H_0)/Z_0$. Since such a state commutes with H_0, it satisfies condition (3). Clearly, $[H_0, P] \neq 0$, and such an objective (the population in the excited-state manifold) is phase controllable following Ref. [32]. Here, WFPC is due to the system-environment Hamiltonian being off-diagonal in the energy eigenbasis of the system. This was pointed out in Refs. [33,34] where it was argued that the environment can assist in WFPC by having a generic system-environment Hamiltonian that does not commute with the target operator. Note that, although we have defined P as the projector onto the excited-state manifold, the choice of P can be determined by the experimentalists based on what they can measure. Thus, we can always choose P such that this condition is satisfied.

Given the condition that the bare evolution does not transfer population from the ground- to the excited-state manifold, the control Hamiltonian in the interaction picture becomes

$$V_I(t) = U(t)^\dagger(V(t)\otimes\mathbb{I})U(t),$$
$$V_I(t) = \sum_{jklm} \tilde{\mu}_{mj}\epsilon(t)|e_j\rangle\langle g_m| \otimes |\alpha_k\rangle\langle\alpha_k| + \text{H.c.} \quad (6)$$

Here, $\tilde{\mu}_{jk}$ is the matrix element of the operator μ in the interaction picture. We can show that the phase control arises from the off-diagonal blocks in the energy eigenbasis. To this end, we consider the initial state to be $R(0) = \sum_{lmnk} c_{lmnk}|g_l\rangle\langle e_m| \otimes |\alpha_n\rangle\langle\alpha_k| + \text{H.c.}$, where c_{lmnk} is the coefficient of the different operators such that the matrix $R(0)$ is a good density matrix and $\{|\alpha_k\rangle\}$ is a basis for the environmental states. Taking the first-order term from Eq. (4) and putting it in Eq. (3), we obtain $\dot{p} = -it_\alpha \sum_{lmn}[\tilde{\mu}_{ml}c_{lmnn}\epsilon(t) - \tilde{\mu}^*_{ml}c^*_{lmnn}\epsilon^*(t)]$ through

$$[V_I(t), R_I(0)] = \sum_{ilmnk} \tilde{\mu}_{il}c_{lmnk}\epsilon(t)|e_i\rangle\langle e_m| \otimes |\alpha_n\rangle\langle\alpha_k| - \tilde{\mu}^*_{im}c_{lmnk}\epsilon^*(t)|g_l\rangle\langle g_i| \otimes |\alpha_n\rangle\langle\alpha_k| - \text{H.c.}, \quad (7)$$

$$P\otimes\mathbb{I}[V_I(t), R_I(0)] = \sum_{ilmnk} \tilde{\mu}_{il}c_{lmnk}\epsilon(t)|e_i\rangle\langle e_m| \otimes |\alpha_n\rangle \times \langle\alpha_k| - \text{H.c.}, \quad (8)$$

$$\text{tr}[P\otimes\mathbb{I}\dot{R}_I(t)] = -i\,\text{tr}\{P\otimes\mathbb{I}[V_I(t), R_I(0)]\}, \quad (9)$$

$$p = t_\alpha \int_0^t -i\sum_{lmn}[\tilde{\mu}_{ml}c_{lmnn}\epsilon(t) - \tilde{\mu}^*_{ml}c^*_{lmnn}\epsilon^*(t)]. \quad (10)$$

This clearly depends on the phase. Here, t_α is obtained after tracing over the environmental state. For completeness, we show in Appendix A that, if we start with an initial state that is diagonal in the energy eigenbasis, there is no phase control.

Condition (3) deals entirely with the initial state of the system and is at the heart of the phase control that will help us witness initial correlations. Since we established that phase control arises from off-diagonal terms, such as $|g_m\rangle\langle e_n|$, we seek to place such terms either in the initial system marginal ρ or the initial correlations χ. Distinguishing these two scenarios would directly enable us to witness correlations. To this end, we will consider quantum preparations.

V. WITNESS OF CORRELATIONS

If a quantum system is initially correlated with the environment, then mathematical operations on the state alone are ill defined unless the effect of such operations on the environment are accounted for. In this context, we discuss preparations, defined as a map from an unknown quantum state to a known quantum state [22,23,35]. For example, the "throw and replace" preparation is given by the action $\mathcal{A}_1[\rho] = \rho_0\ \forall\ \rho \in \mathcal{B}(\mathcal{H})$ and simply maps any initial marginal state of the system to a fixed state ρ_0. Another example of a preparation is the disentanglement channel or the "marginal preserving" preparation. The action of such a preparation is to disentangle a given joint state, namely, $\mathcal{A}[R(0)] = \rho\otimes\tau$ which decorrelates the system and the environment [36,37]. This preparation cannot be carried out universally with just one density matrix [36] but can be performed easily with two copies [38] of the system-environment density matrices as shown in Appendix B.

Phase control can arise from off-diagonal terms of the type $|g_m\rangle\langle e_n|$ arising either in the marginal state of the system or the correlation matrix or both. To detect where phase controllability arises from, we consider marginal preserving preparations $\mathcal{A}$ defined above. We note that before $\mathcal{A}$, the joint system-environment state is given by $R(0) = \rho\otimes\tau + \chi$, whereas after $\mathcal{A}$, the joint system-environment state is given by $R(0) = \rho\otimes\tau$. If all of the off-diagonal terms are present in χ, the marginal preserving preparation $\mathcal{A}$ erases the correlation matrix, thus, removing weak-field phase controllability from the system. On the other hand, if the off-diagonals are all present in ρ, then the weak-field phase controllability is not disrupted by $\mathcal{A}$. Note that, although the reaction yield (whose rate is given by $\dot{p}$) changes because $\dot{p}$ depends on χ, in general, through t_α from Eq. (10), the phase dependence of $\dot{p}$ does not change because all of the control is attributed to the marginal state.

We propose an experiment performed on two copies [39] of the system-environment state that can witness correlations. The first copy is simply checked to see if the system enjoys WFPC (see Fig. 1). On the second copy, we perform the marginal preserving preparation on the system, and once

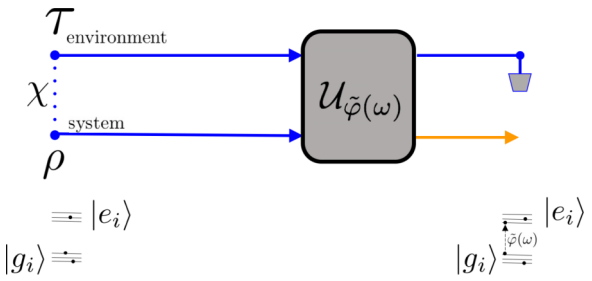

FIG. 1. Weak-field phase control: The system is depicted with a ground-state manifold and an excited-state manifold. An initially correlated system-environment state $R(0) = \rho \otimes \tau + \chi$ is subject to a joint unitary operator that depends on phase of a control field $\varphi(t) = \mathcal{F}[\tilde{\varphi}(\omega)]$. The system is said to be weak-field phase controllable if the population in the excited-state manifold can be controlled by the phase of sufficiently weak control pulses.

again check for WFPC. If we detect WFPC before $\mathcal{A}$ and no WFPC after $\mathcal{A}$, then we have detected the correlation matrix whose off-diagonal terms induced WFPC. This witnessing of the correlations is illustrated in Fig. 2. On the other hand, if we detect no WFPC both before and after the marginal preserving preparation $\mathcal{A}$, then we cannot say that there were no correlations between the system and the environment. We show below that the set of all system-environment density matrices which have a nonzero correlation matrix that cannot be detected by the dual WFPC test outlined above is measure zero. We express a general correlation matrix in the energy eigenbasis of the system as

$$\chi = \sum_{lmnk} a_{klmn}|g_k\rangle\langle g_l| \otimes |\alpha_m\rangle\langle\alpha_n| + b_{klmn}|g_k\rangle\langle e_l| \otimes |\alpha_m\rangle\langle\alpha_n| + c_{klmn}|e_k\rangle\langle e_l| \otimes |\alpha_m\rangle\langle\alpha_n| + \text{H.c.} \tag{11}$$

For there to be no $|g\rangle\langle e|$ terms,

$$b_{klmn} = 0 \quad \forall\, k, l, m, n. \tag{12}$$

With just one of these constraints, we have the remaining subset of χ confined to a lower-dimensional subspace of all χ. Thus, the set of χ without any $|g_m\rangle\langle e_n|$ terms has Lebesgue measure zero.

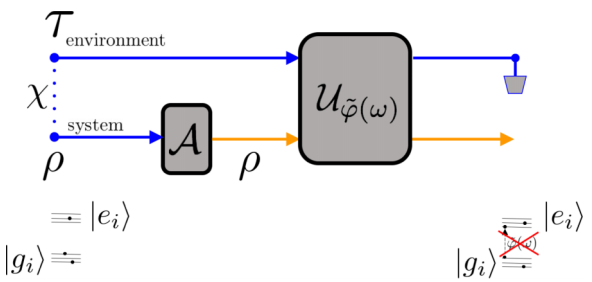

FIG. 2. Witness of correlations: A marginal preserving preparation $\mathcal{A}$ can be used to witness initial correlations between the system and the environment, denoted by χ. The preparation $\mathcal{A}$ witnesses correlations since the output of a marginal preserving preparation is an uncorrelated state, and, hence, any existing correlations can be seen in the phase controllability of the reaction products.

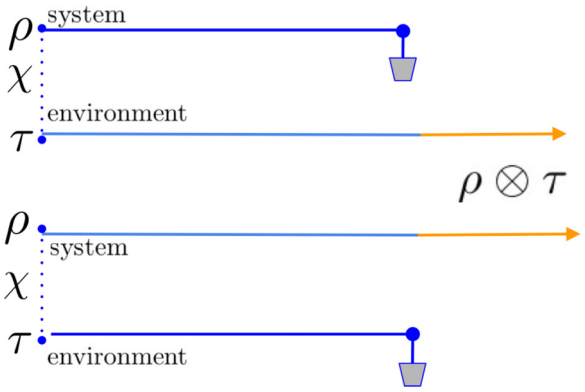

FIG. 3. Marginal preserving preparation illustrated with two copies of the system-environment state. By swapping the state of the system from one copy to another, the system state is no longer correlated with its new environment whereas preserving the marginal states.

We also consider the scenario where the joint state $R(0)$ has the relevant off-diagonal terms in both the marginal system state and the correlation matrix. In this case, the reaction yield $p(t)$ is phase controllable, but this phase controllability arises due to both aforementioned terms. This means that the marginal preserving preparation, which removes the correlation matrix χ will change the quantitative details of the phase controllability. This quantitative change can witness the presence of off-diagonal elements in both terms as there would be fewer terms in the summation in Eq. (10) after $\mathcal{A}$. For completeness, we also consider the case when the off-diagonal terms are in the marginal system state but not in the correlation matrix. In this case, the quantitative details of the phase controllability will not change after the marginal preserving preparation. We summarize this in Table I.

Another approach to detecting the correlation matrix when both the marginal system state and the correlation matrix have $|g_m\rangle\langle e_n|$ terms is as follows. We first make two separate preparations of the system into different states $|\psi_m\rangle\langle\psi_m| \otimes \tau^{E|\psi_m}$ where $\psi_m \in g, e$ and $\tau^{E|\psi_m}$ is the marginal environmental state given that the system is in ψ_m. Note that if there are no correlations, the marginal environmental state is the same for all such preparations of the system state. We then rotate this prepared state by a unitary L such that the resulting system state has off-diagonal terms of the form $|g_m\rangle\langle e_n|$. We can then check the amount of phase control from both our prepared and our rotated states. If both of these states have different environmental marginals, then the amount of phase control would be different as the trace over the environment t_α would be different. Thus, this would be a witness of initial correlations.

Finally, we consider the correlation matrix at intermediate times and apply this formalism to the quantum re-

TABLE I. Table summarizing the witnessing of correlations.

Off-diagonals	Not in χ	In χ
Not in ρ	No WFPC	WFPC → no WFPC
In ρ	$\frac{dp(t)}{d\varphi}$ unchanged	$\frac{dp(t)}{d\varphi}$ changes

gression formula (QRF). Consider again $R(0) = \rho(0) \otimes \tau(0)$ evolving as $R(t_1) = U_0(t_1)R(0)U_0^\dagger(t_1)$. Here, $U_0(t)$ is the free evolution before the laser field $\varepsilon(t)$ is switched on. Now, the two-time correlation function is given by $\langle B(t_2)A(t_1)\rangle = \mathrm{tr}[U_0^\dagger(t_2)BU_0(t_2)U_0^\dagger(t_1)AU_0(t_1)R(0)]$ [40]. This can be written as $\mathrm{tr}_S(B\mathbb{Z})$, where A, B are the system operators in the Schrödinger picture and $\mathbb{Z}$ is given by

$$\mathbb{Z} = \mathrm{tr}_E[U_0(t_2 - t_1)AR(t_1)U_0^\dagger(t_2 - t_1)]. \tag{13}$$

If $R(t_1) = \rho(t_1) \otimes \tau(t_1)$, then $\mathbb{Z} = \Phi_{t_1\to t_2}[A\rho(t_1)]$ where the CPTP map $\Phi_{t_1\to t_2}$ is the evolution operator for the system dynamics. Hence, the evolution of the two-time correlation function is governed by the same evolution equation as the density matrix, following the spirit of Onsager's regression theorem. If $R(t_1) = \rho(t_1) \otimes \tau(t_1) + \chi(t_1)$, then the regression formula almost always breaks down (see Appendix D). Thus, QRF relies on the fact that, for all $t_1 < t_2$, the total state at the intermediate time is assumed to be well approximated by a product state $\rho(t_1) \otimes \tau(t_1)$. The validity of this stronger "factorization" approximation [16] needs to be ascertained before QRF can be applied to a given physical system. This relationship between QRF and intermediate correlations can also be written in terms of a Markovian nondivisible master equation where the nondivisibility is a signature of the correlations shared between the system and the environment [40]. If we partition the physical system under consideration to have a similar structure (a ground- and an excited-state manifold) and, furthermore, if the system is not spontaneously excited by the free evolution $U_0(t)$, then, following our discussion, WFPC can almost always predict the violation of the QRF. Clearly, if we switch on a weak laser field at the intermediate time t_1, following our earlier discussion, we can witness the correlation matrix at intermediate times. Thus, we can almost always predict the violation of the QRF.

VI. CONCLUSIONS

Our ability to witness and characterize NM in physical systems is crucial to understand complex quantum systems. In this paper, we show how standard experimental techniques can be adapted to witness correlations which are closely related to NM. This is important since often the dynamical modeling of a physical system follows assumptions, such as the Born-Markov approximation. Such assumptions make strong claims about the nature of the initial and subsequent density matrix of the system environment. Furthermore, the applicability of important theorems, such as the generalized quantum regression formula almost always rely on the absence of intermediate correlations. The presence of (almost all) such correlations can be witnessed by making a small set of experimentally verifiable assumptions.

Finally, we note that WFPC is used here as an alternative to quantum process tomography on initially correlated systems [41,42]. The full reconstruction of the dynamical map for correlated dynamics typically scales very unfavorably with the size of the universe. Such proposals will herald new experimental progress in the detection, characterization, and control of non-Markovian systems.

ACKNOWLEDGMENTS

We thank R. Kosloff, K. Modi, J. Pillo, and A. Shaji for detailed discussions about the paper. S.V. acknowledges support from an IITB-IRCC Grant No. 16IRCCSG019 and by the National Research Foundation, Prime Minister's Office, Singapore under its Competitive Research Programme (CRP Award No. NRF-CRP14-2014-02).

APPENDIX A: PROOF THAT EXCITED STATES DO NOT PRODUCE WFPC

Consider an initial-state $R(0) = \sum_{jk} |e_j\rangle\langle e_j| \otimes |\alpha_k\rangle\langle\alpha_k|$ which has diagonal states in the excited-state manifold without loss of generality. The first-order term in the perturbative series expansion will clearly be 0 as

$$[V_I(t), R_I(0)] = \sum_{jkl} \tilde{\mu}_{lj}^* \epsilon^*(t)|g_l\rangle\langle e_j| \otimes |\alpha_k\rangle\langle\alpha_k| - \mathrm{H.c.} \tag{A1}$$

This has no diagonal terms and, hence, its trace is zero. To check for phase control in the second order, we evaluate it to be

$$\begin{aligned}[V_I(t), [V_I(u), R_I(0)]] &= \sum_{jklm} \tilde{\mu}_{lj}^* [\tilde{\mu}_{lm}\epsilon(t)\epsilon^*(u)|e_m\rangle\langle e_j| \otimes |\alpha_k\rangle\langle\alpha_k| \\ &\quad - \tilde{\mu}_{mj}\epsilon(t)\epsilon^*(u)|g_l\rangle\langle g_m| \otimes |\alpha_k\rangle\langle\alpha_k|] + \mathrm{H.c.}\end{aligned} \tag{A2}$$

Acting on this by the projection matrix and performing a trace, we get

$$t_\alpha \sum_{lj} |\tilde{\mu}_{lj}|^2 \epsilon(t)\epsilon^*(u) + \mathrm{c.c.}, \tag{A3}$$

where t_α is the trace of the environment marginal state. We can summarize the result as

$$\dot{p} = t_\alpha \int_0^t du \sum_{lj} |\tilde{\mu}_{lj}|^2 \epsilon(t)\epsilon^*(u) + |\tilde{\mu}_{lj}|^2 \epsilon^*(t)\epsilon(u). \tag{A4}$$

This is clearly dependent on the autocorrelation function and has been proved to be independent of the phase by Am-Shallem and Kosloff [32]. Thus, phase control is not possible if the initial density matrix is diagonal.

APPENDIX B: MARGINAL PRESERVING PREPARATION

Marginal preserving preparations (MPPs) refer to a general decorrelating map that takes a bipartite quantum system as input and outputs the marginal states (see Fig. 3). As shown in Refs. [36,37], this MPP cannot be universal for a single copy of the system. In the figure, we illustrate the marginal preserving preparation given two copies of the system with the environment. By construction, we hence show that a universal MPP is possible if two copies are available.

APPENDIX C: THE SET OF ALL χ's WITH NO $|g\rangle\langle e|$ TERMS IS A SET OF MEASURE ZERO

We express a general correlation matrix in the energy eigenbasis of the system as

$$\chi = \sum_{lmnk} a_{klmn}|g_k\rangle\langle g_l| \otimes |\alpha_m\rangle\langle\alpha_n| + b_{klmn}|g_k\rangle\langle e_l| \otimes |\alpha_m\rangle\langle\alpha_n| + c_{klmn}|e_k\rangle\langle e_l| \otimes |\alpha_m\rangle\langle\alpha_n| + \text{H.c.} \tag{C1}$$

For there to be no $|g\rangle\langle e|$ terms,

$$b_{klmn} = 0 \quad \forall\, k, l, m, n. \tag{C2}$$

With just one of these constraints, we have the remaining subset of χ confined to a lower-dimensional subspace of all χ. Thus, the set of χ without any $|g_m\rangle\langle e_n|$ terms has Lebesgue measure zero.

APPENDIX D: PROOF THAT QRF ALMOST ALWAYS BREAKS DOWN WHEN χ IS NONZERO

Here, we show that QRF is generally violated when χ is nonzero and show explicitly when QRF can hold even with nonzero χ. We can write the two-point correlation function as

$$\langle B(t_2)A(t_1)\rangle = \text{tr}[U_0^\dagger(t_2)B \otimes \mathbb{I}U_0(t_2)U_0^\dagger(t_1)A \otimes \mathbb{I}U_0(t_1)R(0)] \tag{D1}$$

$$= \text{tr}[U_0(t_1)U_0^\dagger(t_2)B \otimes \mathbb{I}U_0(t_2)U_0^\dagger(t_1)A \otimes \mathbb{I}R(t_1)] \tag{D2}$$

$$= \text{tr}[U_0^\dagger(t_2 - t_1)B \otimes \mathbb{I}U_0(t_2 - t_1)A \otimes \mathbb{I}R(t_1)] \tag{D3}$$

$$= \text{tr}[CR(t_1)], \tag{D4}$$

where $C(t_2 - t_1) = U_0^\dagger(t_2 - t_1)B \otimes \mathbb{I}U_0(t_2 - t_1)A \otimes \mathbb{I}$. We can perform our experiment to detect the correlations of the system at time t_1. If the experiment detects correlations, we can conclude that $R(t_1) = \rho \otimes \tau + \chi$ for some ρ, τ, and χ. Now, the quantum regression formula would not be violated if and only if $\text{tr}(C\chi) = 0$ [43]. Let the dimension of the system and the environment be N and K, respectively. Thus, we can expand C in the Fano representation to write it as $C = d\mathbb{I} \otimes \mathbb{I} + \sum_i a_i\sigma_i \otimes \mathbb{I} + \sum_j b_j\mathbb{I} \otimes \sigma_j + \sum_{i,j} c_{ij}\sigma_i \otimes \sigma_j$ where σ_i and σ_j are the traceless generators of $SU(N)$ and $SU(K)$, respectively. Since both the partial traces of χ are 0, the quantity of interest simplifies to $\text{tr}(C\chi) = \sum_{i,j} c_{ij}\text{tr}(\sigma_i \otimes \sigma_j\chi) = 0$. In this space of operators with both partial traces vanishing, the trace acts as an inner product. Thus, another way to restate this equation is to say that C is orthogonal to χ. This immediately makes the set of all χ's that satisfy this equation to be of Lebesgue measure 0 if and only if $c_{ij} \neq 0$ for some i and j. Thus, the problem is equivalent to showing that not all c_{ij}'s are 0.

If the observables are stationary and do not change with time, the quantum regression formula would, of course, hold. However, the QRF is most useful when the observables are not stationary. Thus, we first assume that our interaction Hamiltonian does not commute with the second measurement B. We then evaluate $D(t) = U_0^\dagger(t)B \otimes \mathbb{I}U_0(t)$ for small times dt ignoring terms of order dt^2. Here, $U_0(t) = e^{-iH_0t}$, and H_0 can be expanded as $H_0 = H_S \otimes \mathbb{I} + \mathbb{I} \otimes H_E + \sum_{i,j} H_i \otimes H_j$. Thus, up to order dt,

$$D(dt) \approx B \otimes \mathbb{I} - i[H_0, B \otimes \mathbb{I}]dt, \tag{D5}$$

$$= B \otimes \mathbb{I} - i\,dt[H_S, B] \otimes \mathbb{I} - i\,dt \sum_{i,j}[H_i, B] \otimes H_j, \tag{D6}$$

$$\text{tr}_S[D(dt)] = t_B\mathbb{I}, \tag{D7}$$

$$\text{tr}_E[D(dt)] = K * (B - i\,dt[H_S, B]) - i\,dt \sum_{i,j} t_j[H_i, B], \tag{D8}$$

where t_j is the trace of H_j and t_B is the trace of B. The correlations of D can be calculated as follows:

$$D_I = D(dt) - \text{tr}_E[D(dt)] \otimes \text{tr}_S[D(dt)], \tag{D9}$$

$$= -i\,dt \sum_{i,j}[H_i, B] \otimes H_j + i\,dt \sum_{i,j} t_j[H_i, B] \otimes \mathbb{I}, \tag{D10}$$

$$= -i\,dt \sum_{i,j}[H_i, B] \otimes (H_j - t_j\mathbb{I}). \tag{D11}$$

Thus, D can be written as $D_S \otimes D_E + D_I$ where $D_I = 0$ if and only if $[H_i, B] = 0$ for all i. Intuitively, this can be viewed as follows. If the system and the environment are interacting, and you measure the system in a way that disturbs it, this measurement would also disturb the environment. Finally, we can calculate $C = D_SA \otimes D_E + D_I(A \otimes \mathbb{I})$. Intuitively, we would expect this to be nonzero, in general, as a nonfactorizable system-environment state cannot, in general, become factorizable by operating on the system. Writing D_I and A in the Fano representation, we get

$$D_I(A \otimes \mathbb{I}) = \sum_{i,j,m} a_m d_{ij}\sigma_i\sigma_m \otimes \sigma_j + \sum_{i,j} t_A d_{ij}\sigma_i \otimes \sigma_j, \tag{D12}$$

$$= \sum_{i,j,m,k} a_m d_{ij}T_{imk}\sigma_k \otimes \sigma_j + \frac{1}{2}\sum_{i,j} a_j d_{ij}\mathbb{I} \otimes \sigma_j + \sum_{i,j} t_A d_{ij}\sigma_i \otimes \sigma_j, \tag{D13}$$

where we have used the property of generators of $SU(N)$ that $\sigma_i\sigma_m = \sum_k T_{imk}\sigma_k + \frac{1}{2}\delta_{im}$. Thus, we would get $c_{ij} = 0$ if and only if $\sigma_{i,m}a_m d_{ij}T_{imk} + t_A d_{kj} = 0\ \forall\, j, k$. This is certainly a set of measure 0. The set of all χ's such that $\text{tr}(C\chi) = 0$ is also of measure 0 if c_{ij} is nonzero. Thus, the set of all such χ's is of measure 0. Additionally, note that d_{ij} is a function of time as D_I changes with time. Thus, even if this condition is met at some instant of time, it will not hold true at some later time. As any experiment would involve a sampling at different instants of time, this would not effect any experimental data.

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