Simulating Open Quantum Systems with Hamiltonian Ensembles and the Nonclassicality of the Dynamics

Hong-Bin Chen, Clemens Gneiting, Ping-Yuan Lo, Yueh-Nan Chen, and Franco Nori

Phys. Rev. Lett. **120**, 030403 — Published 19 January 2018

DOI: 10.1103/PhysRevLett.120.030403

Simulating open quantum systems with Hamiltonian ensembles and the nonclassicality of the dynamics

Hong-Bin Chen,[1,*] Clemens Gneiting,[2] Ping-Yuan Lo,[3] Yueh-Nan Chen,[1,4,†] and Franco Nori[2,5]

[1] *Department of Physics, National Cheng Kung University, Tainan 70101, Taiwan*
[2] *Quantum Condensed Matter Research Group, RIKEN, Wako-shi, Saitama 351-0198, Japan*
[3] *Department of Electrophysics, National Chiao Tung University, Hsinchu 30010, Taiwan*
[4] *Physics Division, National Center for Theoretical Sciences, Hsinchu 30013, Taiwan*
[5] *Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1040, USA*

(Dated: November 21, 2017)

The incoherent dynamical properties of open quantum systems are generically attributed to an ongoing correlation between the system and its environment. Here, we propose a novel way to assess the nature of these system-environment correlations by examining the system dynamics alone. Our approach is based on the possibility or impossibility to simulate open system dynamics with Hamiltonian ensembles. As we show, such (im)possibility to simulate is closely linked to the system-environment correlations. We thus define the nonclassicality of open system dynamics in terms of the nonexistence of a Hamiltonian-ensemble simulation. This classifies any nonunital open system dynamics as nonclassical. We give examples for open system dynamics that are unital and classical, as well as unital and nonclassical.

Introduction.—When a quantum system interacts with its environment, its dynamical behavior will in general deviate from the dynamics of a strictly isolated one [1–6]. As a result of an ongoing bipartite correlation arising from the system-environment interaction, the system dynamics may display incoherent characteristics, such as dephasing or damping processes. Formally, such processes are captured by quantum master equations, replacing the von Neumann equation for isolated systems.

However, incoherent dynamics can also arise as a consequence of a purely classical averaging procedure over distinct autonomous evolutions. For example, the double slit experiment can, when exposed to a disordered potential and after averaging, encounter similar decoherence as if which-slit information had leaked into an environment [7]. In this sense, disordered quantum systems described by Hamiltonian ensembles can behave in an analogous manner as open quantum systems, even if individual realizations are strictly isolated (Fig. 1) [7–10].

Here, we exploit this dynamical correspondence to assess the nature of the system-environment correlations in terms of the system properties alone. As we show, the impossibility to simulate is necessarily linked to nonclassical system-environment correlations. On the other hand, if such simulation is possible, then there always exists a system-environment model, which reproduces the system dynamics by relying only on classical correlations. This leads us to defining the nonclassicality of open system dynamics in terms of the nonexistence of a Hamiltonian-ensemble simulation. Alternative definitions for the nonclassicality of system dynamics have been proposed [11, 12]. In these definitions, the dynamics is considered classical if the state preserves classicality during the temporal evolution. Typically, the classicality of states in these approaches is formulated in terms of the Wigner function or the Glauber-Sudarshan P-representation [13–18]. While these definitions also rely on system properties alone, their applicability is limited to systems amenable for such a phase space description, excluding other cases of interest. We, instead, propose to discuss the nonclassicality of *open* system dynamics separately, based on the system-environment correlations and independent of the nature of the system.

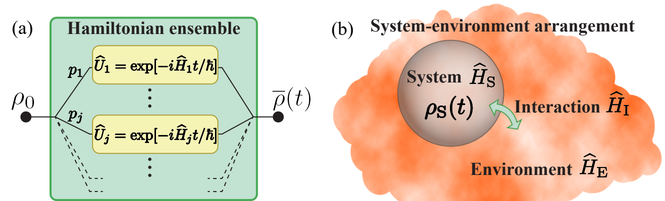

FIG. 1. (a) Schematic illustration showing the averaged state $\overline{\rho}(t)$ resulting from the Hamiltonian ensemble $\{(p_j, \widehat{H}_j)\}$. The averaged state in general follows incoherent dynamics, i.e., similar to open quantum systems, its time evolution cannot be accurately captured by the von Neumann equation alone. (b) If a quantum system interacts with a bath, their interaction will in general correlate them. As we show, the nature of these correlations, i.e., classical or quantum, is intimately connected to the (im)possibility to simulate the dynamics of the open system with a Hamiltonian ensemble.

As an immediate consequence of our definition, all nonunital dynamics, e.g., dissipative processes, are nonclassical. The spin-boson model, in contrast, which displays unital dynamics and—on the level of the model—quantum correlations, can, as we find, be simulated and thus exhibits classical open-system dynamics according to our definition, since these correlations cannot be certified by considering the system dynamics alone. In the

* hongbinchen@phys.ncku.edu.tw
† yuehnan@mail.ncku.edu.tw

case of an extended spin-boson model, however, where the environment is complemented by a second qubit and the system dynamics remains unital, we prove the nonexistence of a simulating Hamiltonian ensemble for certain spectral densities, i.e., the system's evolution is in these cases witnessed to be manifestly nonclassical.

Dynamics of Hamiltonian ensembles.—An isolated quantum system is described by a Hamiltonian ensemble (HE) $\{(p_j, \widehat{H}_j)\}$, if the autonomous Hamiltonian $\widehat{H}_j$ of the system is drawn from a probability distribution p_j [see Fig. 1(a)]. Such HEs are applicable to describing disordered quantum systems. Here, we relate HEs to open quantum systems.

The dynamics of the ensemble averaged state $\overline{\rho}(t)$ exhibits features distinct from the dynamics of any single realization. The latter is governed by the unitary evolution $\rho_j(t) = \widehat{U}_j \rho_0 \widehat{U}_j^\dagger$, with the initial state ρ_0 and $\widehat{U}_j = \exp[-i\widehat{H}_j t/\hbar]$, whereas the dynamics of the averaged state $\overline{\rho}(t)$ is given by the unital (i.e., identity invariant) map

$$\overline{\rho}(t) = \sum_j p_j \exp[-i\widehat{H}_j t/\hbar]\rho_0 \exp[i\widehat{H}_j t/\hbar]. \qquad (1)$$

Note that, an evolution equation for $\overline{\rho}(t)$ cannot be reduced to some effective Hamiltonian alone, but must in general take the form of a quantum master equation [7, 8] (see [19] for an experimental implementation of this).

A seminal and instructive example considers a single qubit subject to spectral disorder, i.e. the Hamiltonians in the ensemble only differ in their eigenvalues, while they share a common basis of eigenstates [8]. The HE may be given by $\{(p(\omega), \hbar\omega\hat{\sigma}_z/2)\}$, with the probability distribution $p(\omega)$ kept general. The resulting master equation reads

$$\frac{\partial}{\partial t}\overline{\rho}(t) = -\frac{i}{\hbar}[\varepsilon(t)\hat{\sigma}_z, \overline{\rho}(t)] + \gamma(t)[\hat{\sigma}_z\overline{\rho}(t)\hat{\sigma}_z - \overline{\rho}(t)], \quad (2)$$

where the effective energy $\varepsilon(t) = \hbar\mathrm{Im}[\partial_t \ln\phi(t)]/2$ and the decoherence rate $\gamma(t) = -\mathrm{Re}[\partial_t \ln\phi(t)]/2$ follow from the dephasing factor

$$\phi(t) = \int_{-\infty}^{\infty} p(\omega) e^{i\omega t} d\omega. \qquad (3)$$

Depending on the underlying probability distribution $p(\omega)$, the master equation (2) can range from time-constant dephasing to a strongly oscillating incoherent behavior, the latter even giving rise to purity revivals [8].

It is worthwhile to recall that the occurrence of incoherent dynamics in the case of HEs is a consequence of the averaging procedure. Nevertheless, it is reminiscent of open quantum systems, where, in contrast, an ongoing correlation between system and environment gives rise to the incoherent dynamics. This article explores the possibility to simulate open quantum systems with HEs and vice versa, and the implications on the system-environment correlations.

Simulating open quantum systems with Hamiltonian ensembles.—We now show that nonclassical system-environment correlations are necessarily linked to the impossibility to simulate the open system dynamics with a HE. To this end, we show—conversely—that, if system and environment are persistently classically correlated, then the reduced system state is described by a HE.

A system-environment arrangement is characterized by an autonomous total Hamiltonian $\widehat{H}_\mathrm{T} = \widehat{H}_\mathrm{S} + \widehat{H}_\mathrm{E} + \widehat{H}_\mathrm{I}$, with the system, $\widehat{H}_\mathrm{S}$, the environment, $\widehat{H}_\mathrm{E}$, and the interaction Hamiltonian $\widehat{H}_\mathrm{I}$ [see Fig. 1(b)]. The total system evolves unitarily as $\rho_\mathrm{T}(t) = \widehat{U}\rho_{\mathrm{T},0}\widehat{U}^\dagger$, with $\widehat{U} = \exp[-i\widehat{H}_\mathrm{T} t/\hbar]$. We say that an open system is described by a HE, if the reduced system state $\rho_\mathrm{S}(t) = \mathrm{Tr}_\mathrm{E}[\rho_\mathrm{T}(t)]$ allows a decomposition of the form (1), where the probabilities p_j and the Hamiltonians $\widehat{H}_j$ of the ensemble are determined by $\widehat{H}_\mathrm{T}$ and the initial state $\rho_{\mathrm{T},0}$.

Instead of further specifying the total Hamiltonian $\widehat{H}_\mathrm{T}$, we now assume that the total state $\rho_\mathrm{T}(t)$ remains at all times classically correlated, displaying neither quantum discord [20, 21] nor entanglement. Under this condition, we argue that, for every initial state of the form $\rho_{\mathrm{T},0} = \rho_{\mathrm{S},0} \otimes \sum_j p_j |j\rangle\langle j|$ (with $\{p_j\}$ a time-independent probability distribution and $\{|j\rangle\}$ a basis of the environment), the reduced system state can be described by a HE.

The detailed proof is presented in the Supplementary Material [22]. Here, we outline the central steps. First, as a direct consequence of the classical correlations, there exists an environmental basis $\{|k\rangle\}$ (in general different from $\{|j\rangle\}$), such that

$$\rho_\mathrm{T}(t) = \sum_{k,j} p_j \widehat{E}_{k,j}\rho_{\mathrm{S},0}\widehat{E}_{k,j}^\dagger \otimes |k\rangle\langle k|, \qquad (4)$$

where the operators $\widehat{E}_{k,j} = \langle k|\widehat{U}(t)|j\rangle$ act on the system and satisfy $\sum_k \widehat{E}_{k,j}^\dagger \widehat{E}_{k,j} = \widehat{I}$ for each j.

To demonstrate that the $\widehat{E}_{k,j}$ are unitary, we again use the zero-discord assumption, which implies that each environmental off-diagonal term vanishes, i.e., $\widehat{E}_{k,j}\rho_{\mathrm{S},0}\widehat{E}_{k',j}^\dagger = 0$ for $k \neq k'$. This, in turn, implies that there exists a bijection between $\{|j\rangle\}$ and $\{|k\rangle\}$ such that $\widehat{E}_{k,j}$ is non-zero only when its two indices match the bijection, i.e., $\widehat{E}_{k,j} = \widehat{E}_{k_{j'},j}\delta_{j,j'}$. Unitarity of the $\widehat{E}_{k,j}$ then follows directly.

Finally, we address the time-dependence of the $\widehat{E}_{k,j}$. Expressing the bijection as a unitary operator $\widehat{\mathcal{U}}(t)$ and safely neglecting the index k, we can recast $\widehat{U}(t)$ in separable form,

$$\widehat{U}(t) = \sum_j \widehat{E}_j(t) \otimes \widehat{\mathcal{U}}(t)|j\rangle\langle j|. \qquad (5)$$

The group properties of $\{\widehat{U}(t)|t \in \mathbb{R}\}$ are thus inherited by the operators $\widehat{E}_j(t)$ and $\widehat{\mathcal{U}}(t)$, i.e., due to the time-independence of the total Hamiltonian, we can write $\widehat{E}_j(t) = \exp[-i\widehat{H}_j t/\hbar]$ and $\widehat{\mathcal{U}}(t) = \sum_j \exp[-i(\theta_j t/\hbar)]|j\rangle\langle j|$, with $\widehat{H}_j$ time-independent Hermitian operators and θ_j real-valued constants. Consequently, Eq. (4) corresponds to a time-independent HE $\{(p_j, \widehat{H}_j)\}$ when tracing over the environment.

Simulating Hamiltonian ensembles with open quantum systems.—The impossibility to simulate an open system with a HE certifies the quantum nature of the system-environment correlations. Notably, this is achieved by considering system properties alone, i.e., without explicit reference to the environment. We now show that, on the other hand, the existence of a simulating HE always admits the possibility of classical system-environment correlations, i.e., the latter cannot be excluded by considering only the system.

We explicitly construct a system-environment arrangement, which reproduces an arbitrary HE $\{(p_j, \widehat{H}_j)\}$ relying only on classical correlations. To this end, we write $\widehat{H}_j = \widehat{\overline{H}} + \widehat{V}_j$ (with the average $\widehat{\overline{H}} = \sum_j p_j \widehat{H}_j$) and choose the interaction to be of the form $\widehat{H}_{\mathrm{I}} = \sum_j \widehat{V}_j \otimes |j\rangle\langle j|$, i.e., it associates with each $\widehat{H}_j$ of the ensemble a distinct state $|j\rangle$ of an (arbitrary) basis of the environment. Note that the index j is generic and may be continuous and/or a multi-index. The environment must then be chosen appropriately to accommodate the complexity of the HE. Moreover, we take the system Hamiltonian to be the average, $\widehat{H}_{\mathrm{S}} = \widehat{\overline{H}}$, and the bath Hamiltonian to be diagonal in the same basis as $\widehat{H}_{\mathrm{I}}$, i.e., $[\widehat{H}_{\mathrm{E}}, \widehat{H}_{\mathrm{E,I}}] = 0$.

With a separable initial state, $\rho_{\mathrm{T}}(0) = \rho_{\mathrm{S},0} \otimes \rho_{\mathrm{E}}$, and $\rho_{\mathrm{E}} = \sum_j p_j |j\rangle\langle j|$ (i.e., $[\rho_{\mathrm{E}}, \widehat{H}_{\mathrm{E}}] = 0$, and the probabilities of the Hamiltonian ensemble are assigned to the environmental populations), the time-evolved total state reads $\rho_{\mathrm{T}}(t) = \widehat{U}_{\mathrm{S+I}}(\rho_{\mathrm{S},0} \otimes \rho_{\mathrm{E}})\widehat{U}^\dagger_{\mathrm{S+I}}$, with $\widehat{U}_{\mathrm{S+I}} = \exp[-i(\widehat{H}_{\mathrm{S}} + \widehat{H}_{\mathrm{I}})t/\hbar]$. Rewriting $\widehat{U}_{\mathrm{S+I}} = \sum_j \widehat{U}_j \otimes |j\rangle\langle j|$, with $\widehat{U}_j = \exp[-i\widehat{H}_j t/\hbar]$, we obtain

$$\rho_{\mathrm{T}}(t) = \sum_j p_j e^{-\frac{i}{\hbar}\widehat{H}_j t} \rho_{\mathrm{S},0} e^{\frac{i}{\hbar}\widehat{H}_j t} \otimes |j\rangle\langle j|. \tag{6}$$

If we now trace over the environment, $\rho_{\mathrm{S}}(t) = \sum_j \langle j|\rho_{\mathrm{T}}(t)|j\rangle$, we recover the desired decomposition (1) in terms of the HE $\{(p_j, \widehat{H}_j)\}$. Moreover, it is easy to see that the total state (6) is exclusively classically correlated, as desired.

As an instructive example, we consider a pair of qubits coupled to each other via a controlled-NOT gate, where a control (C) qubit determines the operation on a target (T) qubit. If the state of the C qubit resides in the classical mixture $\rho_{\mathrm{C}} = a|1\rangle\langle 1| + (1-a)|0\rangle\langle 0|$ [23], the reduced dynamics of the T qubit will be described by the mixture of evolutions $\rho_{\mathrm{T}}(t) = a\widehat{U}_x \rho_{\mathrm{T},0} \widehat{U}_x^\dagger + (1-a)\rho_{\mathrm{T},0}$, with $\widehat{U}_x = \exp[-iJ\hat{\sigma}_x t/2\hbar]$ and J the coupling strength. We thus recover the HE $\{(a, J\hat{\sigma}_x/2), (1-a, \widehat{I})\}$. In this example, the C qubit plays the role of environment, and the qubit pair is at most classically correlated.

Nonclassicality of the dynamics.—It appears natural to classify open system dynamics according to their correlation with the environment, i.e., if system and environment are merely classically correlated, the dynamics may be considered classical; if they are quantum correlated, one may call the dynamics nonclassical. In most cases, however, one does not have (full) access to the environment, rendering such immediate definition problematic.

We now suggest to classify open system dynamics by the (im)possibility to describe the system dynamics by a HE. On the one hand, this definition relies only on system properties, as desired from a practical point of view. On the other hand, as we have shown, it directly links to the system-environment correlations, as desirable from a conceptual perspective. Whenever such simulation exists, it is impossible to exclude classical system-environment correlations by knowledge of the system dynamics alone, and we call the latter classical. If the simulation does not exist, quantum correlations must be involved, hence the dynamics is nonclassical.

As a direct consequence of our definition, any nonunital dynamics is classified nonclassical—a simulating HE is manifestly excluded, certifying the presence of quantum correlations. This includes, e.g., dissipative processes such as the spontaneous decay of an atom. On the other hand, according to our operational definition, we may even call an open system dynamics classical if the actual system-environment correlations are quantum. This is because our approach is deliberately ignorant of the actual environment and only relies on the *possibility* to explain the system dynamics with classical correlations. Next, we give an example for this.

Simulating the spin-boson model.—We now show that the system dynamics of the spin-boson model can be simulated by a HE, even though the actual model displays quantum correlations [24, 25]. The spin-boson model

$$\widehat{H}_{\mathrm{S}} = \frac{\hbar\omega_0}{2}\hat{\sigma}_z \qquad , \qquad \widehat{H}_{\mathrm{E}} = \sum_{\vec{k}} \hbar\omega_{\vec{k}} \hat{b}^\dagger_{\vec{k}} \hat{b}_{\vec{k}},$$
$$\widehat{H}_{\mathrm{I}} = \hat{\sigma}_z \otimes \sum_{\vec{k}} \hbar(g_{\vec{k}} \hat{b}^\dagger_{\vec{k}} + g^*_{\vec{k}} \hat{b}_{\vec{k}}), \tag{7}$$

has been extensively studied and is analytically solvable [2]. Tracing over the environment, the qubit system exhibits pure dephasing dynamics characterized by the dephasing factor

$$\phi(t) = \exp\left[i\omega_0 t - \Phi(t)\right]. \tag{8}$$

In contrast to Eq. (3), which results from averaging over a HE, the dephasing factor (8) incorporates the information of the interaction and the environment into $\Phi(t) = 4\int_0^\infty \omega^{-2}\mathcal{J}(\omega)\coth(\hbar\omega/2k_{\mathrm{B}}T)(1-\cos\omega t)\,d\omega$, where $\mathcal{J}(\omega)$ is the environmental spectral density. The above solution assumes that the initial state is a direct product, and that the environment is initially thermalized at temperature T.

We now construct a simulating HE. In view of Eq. (2), we deduce that individual member Hamiltonians in the ensemble must be of the form $\omega\hat{\sigma}_z/2$, which leaves us with determining the corresponding probabilities. Given a probability distribution $p(\omega)$, the averaged dynamics can be determined by Eq. (3). Conversely, the underlying distribution function leading to a specific dephasing factor (8) is obtained via the inverse Fourier transform

$$\wp(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp[i\omega_0 t - \Phi(t)]e^{-i\omega t}dt. \tag{9}$$

It is clear that the effect of ω_0 is merely to shift $\wp(\omega)$.

To be a legitimate probability distribution function, the resulting $\wp(\omega)$ in Eq. (9) must be normalized, $\int_{-\infty}^{\infty}\wp(\omega)d\omega = 1$, real, $\wp(\omega) \in \mathbb{R}$, and positive, $\wp(\omega) \geq 0$. Normalization is easily seen, since $\phi(0) = 1$ follows from the fact that the pure dephasing dynamics, characterized by Eq. (8), should be completely positive and trace-preserving (CPTP). We therefore have $\int_{-\infty}^{\infty}\wp(\omega)d\omega = (2\pi)^{-1}\int_{-\infty}^{\infty}\exp[i\omega_0 t - \Phi(t)]\,2\pi\delta(t-0)dt = 1$. Moreover, since one is generically interested in the dynamical properties only for $t \geq 0$, we can deliberately extend the time domain to the full real axis such that $\Phi(t)$ is even and $\phi(-t) = \phi(t)^*$. This guarantees that $\wp(\omega)$ is real: $\wp(\omega) = (\pi)^{-1}\int_0^{\infty}\exp[-\Phi(t)]\cos(\omega-\omega_0)tdt \in \mathbb{R}$.

Positivity of $\wp(\omega)$ is less obvious, due to the sinusoidal factors of the integrand in Eq. (9). In the following, we invoke *Bochner's theory* [26] to prove the general positivity of $\wp(\omega)$. To this end, we first introduce the notion of positive definiteness. A function $f:\mathbb{R}\to\mathbb{C}$ is called to be positive definite if it satisfies $\sum_{j,k} f(t_j - t_k)z_j z_k^* \geq 0$ for any finite number of pairs $\{(t_j, z_j)|t_j \in \mathbb{R}, z_j \in \mathbb{C}\}$. Note that positive definiteness of a function is different from a positive function, since the latter may not necessarily be positive definite and vice versa. Rather, it corresponds to the positive semidefiniteness of a Hermitian matrix $[f(t_j - t_k)]_{j,k\in\mathcal{S}}$, formed by the function values $f(t_j - t_k)$ in accordance with a certain set of indices $\mathcal{S}$. As one can show, $\phi(t)$ in Eq. (8) is indeed positive definite. The proof is given in [22].

Bochner's theorem states that a function f, defined on $\mathbb{R}$, is the Fourier transform of unique positive measure with density function $\wp$, if and only if f is continuous and positive definite [27, 28]. We can thus conclude that $\phi(t)$ in Eq. (8) is the Fourier transform of certain valid probability distribution $\wp(\omega)$, Eq. (9), i.e. an analog to Eq. (3).

In summary, we have proven that there exists a unique HE, $\{(\omega\hat{\sigma}_z/2, \wp(\omega))\}$, which simulates the system dynamics exactly, irrespective of the spectral density $\mathcal{J}(\omega)$ and the associated, possibly intricate system-environment entanglement. We thus call this dynamics classical.

Extended spin-boson model.—Unless the system dynamics is nonunital, proving the nonexistence of a simulating HE is in general a nontrivial task. We now accomplish this for an extended spin-boson model, at the same time deducing the presence of quantum correlations from system properties alone.

Our model consists of two qubits coupled to a common boson environment. The system and the interaction Hamiltonian are replaced by $\widehat{H}_{\mathrm{S}} = \sum_j \hbar\omega_j\hat{\sigma}_{z,j}/2$ ($j = 1, 2$) and $\widehat{H}_{\mathrm{I}} = \sum_{j,\vec{k}}\hat{\sigma}_{z,j}\otimes\hbar(g_{j,\vec{k}}\hat{b}^\dagger_{\vec{k}} + g^*_{j,\vec{k}}\hat{b}_{\vec{k}})$, respectively, while the environment Hamiltonian $\widehat{H}_{\mathrm{E}}$ is kept as in Eq. (7). Note that the two qubits do not interact directly. The coupling constants $g_{j,\vec{k}}$ are in general complex numbers. In order to reveal the nonclassical effects caused by their relative phase, we assume, for simplicity, that they have the same amplitude, i.e., $g_{2,\vec{k}} = g_{1,\vec{k}}e^{i\varphi}$.

In the interaction picture, the total system evolves according to $\widehat{U}^{\mathrm{I}}(t) = \mathcal{T}\left\{\exp\left[-i\int_0^t\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\hat{b}^\dagger_{\vec{k}}(\tau) + \widehat{Z}^\dagger_{\vec{k}}\hat{b}_{\vec{k}}(\tau)d\tau\right]\right\}$, with $\mathcal{T}$ the time-ordering operator, $\widehat{Z}_{\vec{k}} = \sum_j g_{j,\vec{k}}\hat{\sigma}_{z,j}$, and $\hat{b}_{\vec{k}}(t) = e^{-i\omega_{\vec{k}}t}\hat{b}_{\vec{k}}$, respectively. In contrast to the conventional spin-boson model, time ordering plays a nontrivial role here [29]. (For details, see Supplementary Material [22].)

In the following we regard one qubit as the system and the other as part of the environment. The reduced dynamics of the system qubit is then pure dephasing with the dephasing factor [cf. Eq. (8)]

$$\phi^{(\mathrm{X})}(t) = \exp\left[-i\vartheta_\varphi(t) - \Phi(t)\right], \tag{10}$$

where

$$\vartheta_\varphi(t) = \cos\varphi\int_0^\infty \frac{4\mathcal{J}(\omega)}{\omega^2}(\omega t - \sin\omega t)d\omega + \mathrm{sign}(t)\sin\varphi\int_0^\infty\frac{4\mathcal{J}(\omega)}{\omega^2}(1-\cos\omega t)d\omega. \tag{11}$$

In the second line, we have manually inserted sign(t). This ensures that $\phi^{(\mathrm{X})}(-t) = \phi^{(\mathrm{X})*}(t)$ and $\wp^{(\mathrm{X})}(\omega) \in \mathbb{R}$. The presence of $\vartheta_\varphi(t)$, however, will in general result in the violation of positivity. Note that, similar to the conventional spin-boson model, individual member Hamiltonians in the HE must be of the form $\omega\hat{\sigma}_z/2$, which allows us to follow the same line of argument.

To demonstrate this violation, we consider the Ohmic spectral density $\mathcal{J}_{\mathrm{o1}}(\omega) = \omega\exp(-\omega/\omega_{\mathrm{c}})$ in the zero-temperature limit and a degenerate system Hamiltonian, i.e., $\omega_j = 0$. In Fig. 2(a), we depict the legitimate probability distribution $\wp_{\mathrm{o1}}(\omega)$ for the conventional spin-boson model at $\omega_{\mathrm{c}} = 1$ (blue curve) and $\omega_{\mathrm{c}} = 3$ (red curve), while in Fig. 2(b), we show $\wp^{(\mathrm{X})}_{\mathrm{o1}}(\omega)$ for our extended model with $\varphi = \pi/4$ (solid curves) and $\varphi = 5\pi/4$ (dashed curves). The latter display a manifest violation of positivity. In Fig. 2(c), we show the landscape of negative contributions to $\wp^{(\mathrm{X})}_{\mathrm{o1}}(\omega)$ against ω and φ for $\omega_{\mathrm{c}} = 1$. The gray dashed lines highlight $\varphi = \pi/4$ and $5\pi/4$, chosen in Fig. 2(b).

Conclusions.—We propose a way to classify open system dynamics according to their system-environment correlations, i.e., if the latter are classical or quantum. As we showed, this can be tested by knowlewdge of the system evolution alone, based on the (im)possibility to

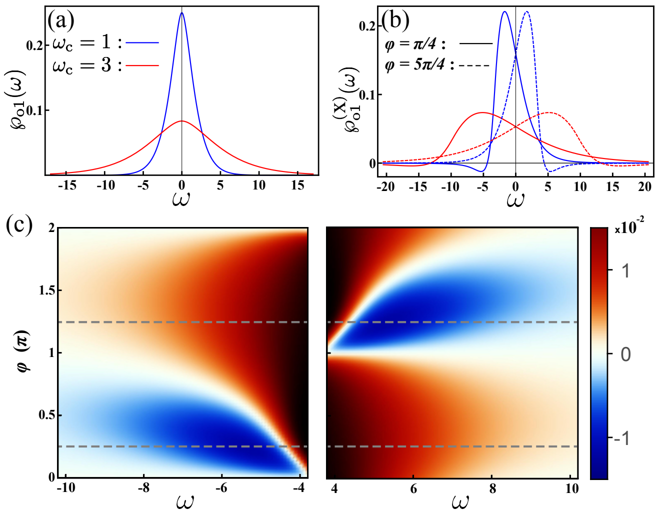

FIG. 2. (a) Legitimate probability distributions $\wp_{o1}(\omega)$ for the conventional spin-boson model. (b) Distributions $\wp^{(X)}_{o1}(\omega)$ for the extended model, violating positivity. (c) The landscape of negative contributions to $\wp^{(X)}_{o1}(\omega)$ against ω and φ for $\omega_c = 1$. The gray dashed lines denote the positions in Fig. 2(b).

simulate the open system dynamics with a Hamiltonian ensemble. According to our definition, any nonunital dynamics is nonclassical. Some unital system evolutions, however, such as in the spin-boson model, are classified classical, even though the model displays quantum correlations. This highlights the operational nature of our definition.

With the extended spin-boson model, we provide an example for unital dynamics which is nonclassical according to our definition. Let us note that one may be able to simulate a larger class of unital dynamics with time-dependent Hamiltonian ensembles. It is for example known that, in the case of qubits, any unital dynamics can be simulated with an ensemble of time-dependent Hamiltonians, albeit only if also the probabilities are allowed to be time-dependent [30, 31]. However, in the case of autonomous system-environment arrangements, which we consider here, such generalization appears unjustified. Finally, let us remark that demonstrating the nonexistence of a Hamiltonian-ensemble simulation is, in the case of unital evolutions, in general a nontrivial task. An equivalent, but simpler test appears desirable.

Acknowledgements.—This work is supported partially by the National Center for Theoretical Sciences and Ministry of Science and Technology, Taiwan, Grants No. MOST 103-2112-M-006-017-MY4 and No. MOST 105-2811-M-006-059, the MURI Center for Dynamic Magneto-Optics via the AFOSR Award No. FA9550-14-1-0040, the Japan Society for the Promotion of Science (KAKENHI), the IMPACT program of JST, JSPS-RFBR Grant No. 17-52-50023, CREST Grant No. JPMJCR1676, RIKEN-AIST Challenge Research Fund, and the Sir John Templeton Foundation.

[1] A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, "Dynamics of the dissipative two-state system," Rev. Mod. Phys. **59**, 1 (1987).
[2] H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, New York, 2002).
[3] U. Weiss, *Quantum Dissipative Systems*, 4th ed. (World Scientific, Singapore, 2012).
[4] W.-M. Zhang, P.-Y. Lo, H.-N. Xiong, M. W.-Y. Tu, and F. Nori, "General non-Markovian dynamics of open quantum systems," Phys. Rev. Lett. **109**, 170402 (2012).
[5] H.-N. Xiong, P.-Y. Lo, W.-M. Zhang, D. H. Feng, and F. Nori, "Non-Markovian complexity in the quantum-to-classical transition," Sci. Rep. **5**, 13353 (2015).
[6] H.-B. Chen, N. Lambert, Y.-C. Cheng, Y.-N. Chen, and F. Nori, "Using non-Markovian measures to evaluate quantum master equations for photosynthesis," Sci. Rep. **5**, 12753 (2015).
[7] C. Gneiting, F. R. Anger, and A. Buchleitner, "Incoherent ensemble dynamics in disordered systems," Phys. Rev. A **93**, 032139 (2016).
[8] C. M. Kropf, C. Gneiting, and A. Buchleitner, "Effective dynamics of disordered quantum systems," Phys. Rev. X **6**, 031023 (2016).
[9] C. Gneiting and F. Nori, "Quantum evolution in disordered transport," Phys. Rev. A **96**, 022135 (2017).
[10] C. Gneiting and F. Nori, "Disorder-induced dephasing in backscattering-free quantum transport," Phys. Rev. Lett. **119**, 176802 (2017).
[11] S. Rahimi-Keshari, T. Kiesel, W. Vogel, S. Grandi, A. Zavatta, and M. Bellini, "Quantum process nonclassicality," Phys. Rev. Lett. **110**, 160401 (2013).
[12] K. K. Sabapathy, "Process output nonclassicality and nonclassicality depth of quantum-optical channels," Phys. Rev. A **93**, 042103 (2016).
[13] E. P. Wigner, "On the quantum correction for thermodynamic equilibrium," Phys. Rev. **40**, 749 (1932).
[14] R. J. Glauber, "Coherent and incoherent states of the radiation field," Phys. Rev. **131**, 2766 (1963).
[15] E. C. G. Sudarshan, "Equivalence of semiclassical and quantum mechanical descriptions of statistical light beams," Phys. Rev. Lett. **10**, 277 (1963).
[16] A. Miranowicz, M. Bartkowiak, X. Wang, Y.-x. Liu, and F. Nori, "Testing nonclassicality in multimode fields: A unified derivation of classical inequalities," Phys. Rev. A **82**, 013824 (2010).
[17] M. Bartkowiak, A. Miranowicz, X. Wang, Y.-x. Liu, W. Leoński, and F. Nori, "Sudden vanishing and reappearance of nonclassical effects: General occurrence of finite-time decays and periodic vanishings of nonclassicality and entanglement witnesses," Phys. Rev. A **83**, 053814 (2011).
[18] A. Miranowicz, K. Bartkiewicz, A. Pathak, J. Peřina, Y.-N. Chen, and F. Nori, "Statistical mixtures of states can be more quantum than their superpositions: Comparison of nonclassicality measures for single-qubit states," Phys.

Rev. A **91**, 042309 (2015).

[19] J.-S. Xu, K. Sun, C.-F. Li, X.-Y. Xu, G.-C. Guo, E. Andersson, R. Lo Franco, and G. Compagno, "Experimental recovery of quantum correlations in absence of system-environment back-action," Nat. Commun. **4**, 2851 (2013).

[20] H. Ollivier and W. H. Zurek, "Quantum discord: A measure of the quantumness of correlations," Phys. Rev. Lett. **88**, 017901 (2001).

[21] B. Dakić, V. Vedral, and C. Brukner, "Necessary and sufficient condition for nonzero quantum discord," Phys. Rev. Lett. **105**, 190502 (2010).

[22] See the Supplementary Material at [URL will be inserted by publisher] for the proof.

[23] H.-B. Chen, J.-Y. Lien, G.-Y. Chen, and Y.-N. Chen, "Hierarchy of non-Markovianity and k-divisibility phase diagram of quantum processes in open systems," Phys. Rev. A **92**, 042105 (2015).

[24] S. Bera, S. Florens, H. U. Baranger, N. Roch, A. Nazir, and A. W. Chin, "Stabilizing spin coherence through environmental entanglement in strongly dissipative quantum systems," Phys. Rev. B **89**, 121108 (2014).

[25] J. Iles-Smith, N. Lambert, and A. Nazir, "Environmental dynamics, correlations, and the emergence of noncanonical equilibrium states in open quantum systems," Phys. Rev. A **90**, 032114 (2014).

[26] S. Bochner, "Monotone Funktionen, Stieltjessche Integrale und harmonische Analyse," Math. Ann. **108**, 378 (1933).

[27] L. H. Loomis, *An Introduction to Abstract Harmonic Analysis* (Van Nostrand, New York, 1953).

[28] W. Rudin, *Fourier Analysis on Groups* (John Wiley, New York, 1990).

[29] John H. Reina, Luis Quiroga, and Neil F. Johnson, "Decoherence of quantum registers," Phys. Rev. A **65**, 032326 (2002).

[30] L. J. Landau and R. F. Streater, "On Birkhoff's theorem for doubly stochastic completely positive maps of matrix algebras," Linear Alg. Appl. **193**, 107 (1993).

[31] K. M. R. Audenaert and S. Scheel, "On random unitary channels," New J. Phys. **10**, 023011 (2008).