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

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I. TIME-INDEPENDENT HAMILTONIAN ENSEMBLE

In the following, we elaborate in detail the proof that classical bipartite correlations allow for a time-independent Hamiltonian ensemble decomposition of the reduced system dynamics. In this proof, we do not assume a specific form of the total Hamiltonian $\widehat{H}_{\mathrm{T}}$. To be precise, we now show that, if the total Hamiltonian $\widehat{H}_{\mathrm{T}}$ is time-independent, and if, 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 $\{p_j\}$ being any time-independent probability distribution), the time-evolved total state $\rho_{\mathrm{T}}(t) = \widehat{U}(t)\rho_{\mathrm{T},0}\widehat{U}^\dagger(t)$ is always classically correlated between system and environment, displaying neither quantum discord nor entanglement at any time, then the reduced system dynamics admits a time-independent Hamiltonian ensemble decomposition.

Proof. Due to the zero-discord assumption, there exists an environmental basis $\{|k\rangle\}$ (in general time-dependent and different from $\{|j\rangle\}$), such that

$$\rho_{\mathrm{T}}(t) = \sum_{k,j} p_j \widehat{E}_{k,j}\rho_{\mathrm{S},0}\widehat{E}^\dagger_{k,j} \otimes |k\rangle\langle k|, \tag{1}$$

where $\widehat{E}_{k,j} = \langle k|\widehat{U}(t)|j\rangle$ are operators acting on the system Hilbert space satisfying $\sum_k \widehat{E}^\dagger_{k,j}\widehat{E}_{k,j} = \widehat{I}$ for each j. At this point, we are not yet clear about the time-dependence of $\widehat{E}_{k,j}$ and $|k\rangle$ nor the unitarity of $\widehat{E}_{k,j}$.

Crucially, the condition

$$\sum_j p_j \widehat{E}_{k,j}\rho_{\mathrm{S},0}\widehat{E}^\dagger_{k',j} = 0 \tag{2}$$

should hold for any $k \neq k'$, due to the zero-discord assumption. Therefore each term $\widehat{E}_{k,j}\rho_{\mathrm{S},0}\widehat{E}^\dagger_{k',j}$ in the above equation vanishes individually. The only possibility to reconcile Eqs. (1) and (2) is the existence of a specific bijection between $\{|j\rangle\}$ and $\{|k\rangle\}$, such that $\widehat{E}_{k,j} = \widehat{E}_{k_{j'},j}\delta_{j,j'}$ for each j, i.e., $\widehat{E}_{k,j}$ is non-zero only when its two indices match the bijection. Then the unitarity of $\widehat{E}_{k_j,j}$ can then be confirmed according to

$$\sum_k \widehat{E}^\dagger_{k,j}\widehat{E}_{k,j} = \widehat{E}^\dagger_{k_j,j}\widehat{E}_{k_j,j} = \widehat{I},\ \forall\ j. \tag{3}$$

The bijection between $\{|j\rangle\}$ and $\{|k\rangle\}$ can be expressed in terms of a unitary operator $\widehat{\mathcal{U}}(t)$, such that $\langle k_{j'}|\widehat{\mathcal{U}}(t)|j\rangle = \delta_{j,j'}$. The unitary evolution operator can then be recast in a separable form,

$$\widehat{U}(t) = \sum_j \widehat{E}_j(t) \otimes \widehat{\mathcal{U}}(t)|j\rangle\langle j|. \tag{4}$$

In the following discussion, we can, in order to keep the notation simple, safely neglect the index k.

Since $\{\widehat{U}(t) = \exp[-i\widehat{H}_{\mathrm{T}}t/\hbar] | t \in \mathbb{R}\}$ forms a group isomorphism on $\mathbb{R}$, we have the one-parameter group property

$$\widehat{U}(t+\delta t) = \widehat{U}(t)\widehat{U}(\delta t) \tag{5}$$

for $t \in \mathbb{R}$ and infinitesimal δt. Due to the unitarity of $\widehat{\mathcal{U}}(t)$, it can be expressed in terms of an Hermitian generator $\widehat{L}(t)$ in the $\mathfrak{u}(\dim\mathcal{H}_{\mathrm{E}})$ Lie algebra on the environmental Hilbert space $\mathcal{H}_{\mathrm{E}}$ such that $\widehat{\mathcal{U}}(t) = \exp[-i\widehat{L}(t)/\hbar]$. Together with Eq. (4), the left hand side of Eq. (5) can be written as

$$\widehat{U}(t+\delta t) = \sum_j \widehat{E}_j(t+\delta t) \otimes \left[\widehat{\mathcal{U}}(t) + \frac{\partial\widehat{\mathcal{U}}(t)}{\partial t}\delta t + \mathcal{O}(\delta t^2)\right]|j\rangle\langle j|. \tag{6}$$

On the right hand side of Eq. (6), we expand $\widehat{\mathcal{U}}(t+\delta t)$ around t to first order in δt. Notably, since we do not know the time-dependence and commutativity of $\widehat{L}(t)$ at this point, we can only achieve a formal expansion in Eq. (6).

Meanwhile, the right hand side of Eq. (5) reads

$$\widehat{U}(t)\widehat{U}(\delta t) = \sum_{j',j} \widehat{E}_{j'}(t)\widehat{E}_j(\delta t) \otimes \widehat{\mathcal{U}}(t)\Big[|j'\rangle\langle j|\delta_{j',j} - \frac{i}{\hbar}|j'\rangle\langle j'|\frac{\partial\widehat{L}(0)}{\partial t}|j\rangle\langle j|\delta t + \mathcal{O}(\delta t^2)\Big]. \tag{7}$$

We again expand $\widehat{\mathcal{U}}(\delta t)$ around $t=0$. However, unlike the formal expansion in Eq. (6), we now obtain an explicit expansion in Eq. (7), since $\widehat{\mathcal{U}}(0) = \widehat{I}$ commutes with any operator.

Comparing Eqs. (6) and (7), we conclude from their first terms that the group property $\widehat{E}_j(t+\delta t) =$

$\widehat{E}_j(t)\widehat{E}_j(\delta t)$ holds and, combined with the unitarity inferred in Eq. (3), that time-independent Hermitian operators $\widehat{H}_j$ exist, such that $\widehat{E}_j(t) = \exp[-i\widehat{H}_j t/\hbar]$, as well.

To reconcile the second terms of Eqs. (6) and (7), $\partial\widehat{L}(0)/\partial t$ should be diagonalized in the basis $\{|j\rangle\}$, such that $\partial\widehat{L}(0)/\partial t = \sum_j(\partial\theta_j(0)/\partial t)|j\rangle\langle j|$, with real parameters $\theta_j(t)$. Moreover, $\widehat{\mathcal{U}}(t)$ should satisfy

$$\frac{\partial\widehat{\mathcal{U}}(t)}{\partial t} = \widehat{\mathcal{U}}(t)\left[-\frac{i}{\hbar}\frac{\partial\widehat{L}(0)}{\partial t}\right]. \tag{8}$$

To guarantee its validity, $\partial\widehat{L}(0)/\partial t$ should commute with $\widehat{L}(t)$, since the latter is the generator of $\widehat{\mathcal{U}}(t)$. Consequently, the time-dependence of each $\theta_j(t)$ can be of first order, such that $\widehat{\mathcal{U}}(t) = \sum_j \exp[-i(\theta_j t/\hbar)]|j\rangle\langle j|$, with real constants θ_j. ■

Consequently, the total state in Eq. (1) can be rewritten as

$$\rho_{\mathrm{T}}(t) = \sum_j p_j \widehat{U}_j \rho_{\mathrm{S},0} \widehat{U}_j^\dagger \otimes |j\rangle\langle j|, \tag{9}$$

with $\widehat{U}_j = \exp[-i\widehat{H}_j t/\hbar]$, which corresponds to a time-independent Hamiltonian ensemble $\{(p_j, \widehat{H}_j)\}$ when tracing over the environment.

Finally, let us remark that, while we restrict ourselves to a time-independent total Hamiltonian, some of our conclusions can be easily generalized to the time-dependent case. This is because Eqs. (1-4) are consequences of the zero-discord assumption alone, regardless of the time-dependence of the total Hamiltonian. Therefore, we can also achieve the ensemble form with time-varying member Hamiltonians for a time-dependent total Hamiltonian. However, as discussed in the main article, in the case of autonomous system-environment arrangements, i.e., in the absence of external control, such generalization appears unjustified.

Additionally, we note that, for the case of time-independent total Hamiltonians, the separable form (4) not only guarantees a persistently classically correlated total state, but also keeps the environmental basis intact without rotation, up to a phase angle $\theta_j t$.

II. POSITIVE DEFINITENESS

Here, we present the proof of the positive definiteness of the dephasing factor $\phi(t) = \exp[i\omega_0 t - \Phi(t)]$. For completeness, we recall the definition of positive definiteness.

Positive definiteness: A function f defined on $\mathbb{R}$ is called positive definite if it satisfies

$$\sum_{j,k} f(t_j - t_k) z_j z_k^* \geq 0 \tag{10}$$

for any finite number of pairs $\{(t_j, z_j)|t_j \in \mathbb{R}, z_j \in \mathbb{C}\}$.

We now show that, if $\phi(t) = \exp[i\omega_0 t - \Phi(t)]$ defines a CPTP pure dephasing dynamics, and if $\Phi(t)$ is even and $\phi(-t) = \phi(t)^*$, then $\phi(t)$ defined on $\mathbb{R}$ is positive definite.

Note that $\phi(t)$ describing a CPTP pure dephasing dynamics implies that $\phi(0) = 1$, $\Phi(0) = 0$, and $|\phi(t)| \leq \phi(0)$ for any $t > 0$. This means that the coherence of the system can never exceed its initial value. These properties will be frequently used in the following proof.

Proof. To simplify the problem, we first observe that the positive definiteness of $\phi(t)$ is equivalent to that of $\exp[-\Phi(t)]$, since

$$\sum_{j,k}\phi(t_j - t_k) z_j z_k^* = \sum_{j,k}\exp[-\Phi(t_j - t_k)]\left(e^{i\omega_0 t_j} z_j\right)\left(e^{i\omega_0 t_k} z_k\right)^*. \tag{11}$$

Correspondingly, we can assume that $\omega_0 = 0$ without loss of generality.

Since Eq. (10) must be valid for any number of pairs, we give the proof in an inductive manner.

In the case of only one pair (t_1, z_1), Eq. (10) is trivially satisfied. We therefore start with the case of two pairs. As stated in the main article, Eq. (10) is equivalent to the positive semidefiniteness of the Hermitian matrix:

$$\mathcal{M}^{(2)} = \begin{bmatrix} 1 & \exp[-\Phi(t_2 - t_1)] \\ \exp[-\Phi(t_1 - t_2)] & 1 \end{bmatrix}. \tag{12}$$

It is automatically satisfied according to the CPTP dynamics defined by $\phi(t)$.

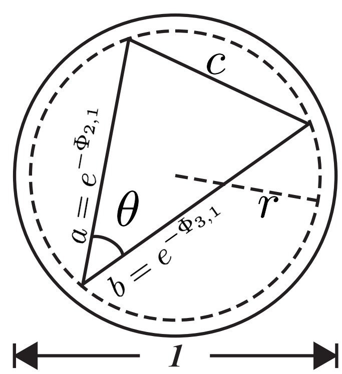

FIG. 1. A geometric visualization of Eq. (14). a and b can be considered as two sides of a triangle with angle θ and circumcircle (dashed circle) of diameter $2r$ less than 1. They are all enclosed in the circle (solid circle) of diameter 1.

We proceed to show the positive semidefiniteness of the Hermitian matrix

$$\mathcal{M}^{(3)} = \begin{bmatrix} 1 & e^{-\Phi_{2,1}} & e^{-\Phi_{3,1}} \\ e^{-\Phi_{1,2}} & 1 & e^{-\Phi_{3,2}} \\ e^{-\Phi_{1,3}} & e^{-\Phi_{2,3}} & 1 \end{bmatrix}, \tag{13}$$

for the case of three pairs. In the above matrix, and hereafter, the abbreviation $\Phi_{j,k} = \Phi(t_j - t_k)$ has been

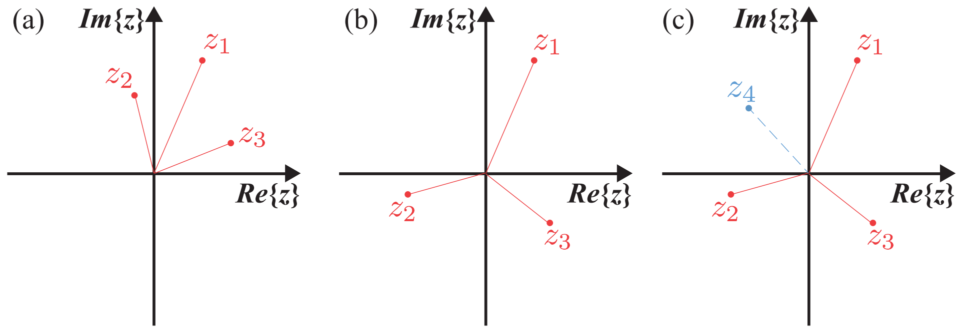

FIG. 2. (a) If the angles between any two z_j is less than $\pi/2$, the summation of all off-diagonal elements in array (15) is positive. (b) To maximize the negative contributions of off-diagonal elements, we must choose appropriate z_j, such that all the relative arguments strictly exceed $\pi/2$. (c) In the case of four pairs, it is impossible to insert the fourth z_4 such that all relative arguments are strictly larger than $\pi/2$.

adopted. Since $\mathcal{M}^{(3)}$ is three-dimensional, it is generically hard to write down an analytic expression for its three eigenvalues λ_μ. Nevertheless, analyzing its characteristic polynomial gives us substantial knowledge on the eigenvalues:

(i) $\lambda_1 + \lambda_2 + \lambda_3 = 3 \geq 0$ follows from the invariance of the trace.

(ii) $\lambda_1\lambda_2 + \lambda_2\lambda_3 + \lambda_3\lambda_1$ equals to the sum of all principal minors of $\mathcal{M}^{(3)}$ of order 2 and is consequently non-negative, since each principal minor is non-negative, following the positive semidefiniteness of $\mathcal{M}^{(2)}$.

(iii) $\lambda_1\lambda_2\lambda_3 = \det\left(\mathcal{M}^{(3)}\right)$. The positivity of the product of eigenvalues is verified with the help of a simple geometric visualization shown in Fig. 1. Explicitly expanding the determinant leads to

$$\begin{aligned}\det\left(\mathcal{M}^{(3)}\right) &= \left(1-\cos^2\theta\right) - \left(a^2+b^2-2ab\cos\theta\right)\\ &= \sin^2\theta - c^2, \end{aligned} \tag{14}$$

with the notation $\cos\theta = \exp[-\Phi_{3,2}]$, $a = \exp[-\Phi_{2,1}]$, and $b = \exp[-\Phi_{3,1}]$. This can be interpreted in terms of a triangle with circumcircle (dashed circle) of diameter $2r$ less than 1. With the help of $c/\sin\theta = 2r$, the positivity of Eq. (14) and, consequently, of the product of eigenvalues is then inferred.

Combining (i)-(iii), we can conclude that the three eigenvalues are non-negative each and, therefore, that $\mathcal{M}^{(3)}$ is positive semidefinite.

Before proceeding to the case of four pairs, it is worthwhile to discuss how the minimum of Eq. (10) is achieved. For the case of three pairs, the LHS of Eq. (10) is equivalent to the summation over entries in the following array:

$$\begin{array}{ccc} |z_1|^2 & e^{-\Phi_{2,1}} z_2 z_1^* & e^{-\Phi_{3,1}} z_3 z_1^* \\ e^{-\Phi_{1,2}} z_1 z_2^* & |z_2|^2 & e^{-\Phi_{3,2}} z_3 z_2^* \\ e^{-\Phi_{1,3}} z_1 z_3^* & e^{-\Phi_{2,3}} z_2 z_3^* & |z_3|^2 \end{array} . \tag{15}$$

If we first determine the amplitudes $|z_j|$ and adjust their arguments and t_j, it is clear that the diagonal elements in the array (15) are all positive and, to reduce the resulting summation, the possible negative contributions are given by the off-diagonal elements. If we choose three pairs such that the angles between any two z_j within them is less than $\pi/2$, as show in Fig. 2(a), the summation of all off-diagonal elements is positive. Therefore, we must choose appropriate pairs such that all their relative arguments strictly exceed $\pi/2$, as show in Fig. 2(b). To maximize the negative contributions, we assume $t_1 = t_2 = t_3$ and $\exp[-\Phi_{j,k}] = 1$. We therefore draw the conclusion that $\sum_{j,k} f(t_j - t_k) z_j z_k^* \geq |z_1 + z_2 + z_3|^2$ for the case of maximized relative arguments between three z_j.

However, in the case of four pairs, it is impossible to insert the fourth z_4 such that all relative arguments are strictly larger than $\pi/2$, as shown in Fig. 2(c). According to the above discussion, to deal with the array of four pairs,

$$\begin{array}{cccc} |z_1|^2 & e^{-\Phi_{2,1}} z_2 z_1^* & e^{-\Phi_{3,1}} z_3 z_1^* & e^{-\Phi_{4,1}} z_4 z_1^* \\ e^{-\Phi_{1,2}} z_1 z_2^* & |z_2|^2 & e^{-\Phi_{3,2}} z_3 z_2^* & e^{-\Phi_{4,2}} z_4 z_2^* \\ e^{-\Phi_{1,3}} z_1 z_3^* & e^{-\Phi_{2,3}} z_2 z_3^* & |z_3|^2 & e^{-\Phi_{4,3}} z_4 z_3^* \\ e^{-\Phi_{1,4}} z_1 z_4^* & e^{-\Phi_{2,4}} z_2 z_4^* & e^{-\Phi_{3,4}} z_3 z_4^* & |z_4|^2 \end{array} , \tag{16}$$

we can at most group three z_j with all three relative arguments strictly larger than $\pi/2$ by setting their corresponding t_j equal. Then the array (16) reduces to a simpler one:

$$\begin{array}{cc} |z_1 + z_2 + z_3|^2 & e^{-\Phi_{4,1}} z_4 (z_1 + z_2 + z_3)^* \\ e^{-\Phi_{1,4}} (z_1 + z_2 + z_3) z_4^* & |z_4|^2 \end{array} . \tag{17}$$

Again, in accordance with the positive semidefiniteness of $\mathcal{M}^{(2)}$, we can guarantee the validity of Eq. (10) for the case of four pairs.

For the case of five or more pairs, a similar procedure can be applied to continuously reduce the problem to an equivalent $\mathcal{M}^{(2)}$ or $\mathcal{M}^{(3)}$ case. This implies the validity of Eq. (10) for the general case. ■

Let us remark that the above proof already indicates the general impossibility of a Hamiltonian ensemble description for arbitrary pure dephasing dynamics. Many conclusions in the above proof hold since the phase angle of $\phi(t)$ is directly proportional to time t. This is particularly manifest in Eq. (11). However, this is in general not the case, e.g., in the extended spin-boson model below. We consequently may obtain invalid (or quasi-) distributions in the extended spin-boson model.

III. EXTENDED SPIN-BOSON MODEL

We proceed with the details of the extended spin-boson model, which consists of two qubits coupled to a common boson environment. The system and the interaction Hamiltonian of the conventional spin-boson model are thus replaced by

$$\begin{aligned}\widehat{H}_{\mathrm{S}} &= \sum_{j=1,2}\frac{\hbar\omega_j}{2}\hat{\sigma}_{z,j},\\ \widehat{H}_{\mathrm{I}} &= \sum_{j,\vec{k}}\hat{\sigma}_{z,j}\otimes\hbar(g_{j,\vec{k}}\hat{b}^{\dagger}_{\vec{k}}+g^{*}_{j,\vec{k}}\hat{b}_{\vec{k}}).\end{aligned} \tag{18}$$

Note that the two qubits do not interact with each other directly. Let us remark that, while we consider two qubits here, our treatment can straightforwardly be generalized to more than two qubits.

Transforming to the interaction picture with respect to $\widehat{H}_{\mathrm{S}}+\widehat{H}_{\mathrm{E}}$, the total system evolves according to the unitary evolution operator

$$\widehat{U}^{\mathrm{I}}(t) = \mathcal{T}\left\{\exp\left[-i\int_0^t\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\hat{b}^{\dagger}_{\vec{k}}(\tau)+\widehat{Z}^{\dagger}_{\vec{k}}\hat{b}_{\vec{k}}(\tau)d\tau\right]\right\}, \tag{19}$$

where $\mathcal{T}$ is the time-ordering operator, $\widehat{Z}_{\vec{k}} = \sum_{j=1,2}g_{j,\vec{k}}\hat{\sigma}_{z,j}$, and $\hat{b}_{\vec{k}}(t) = e^{-i\omega_{\vec{k}}t}\hat{b}_{\vec{k}}$, respectively. In the conventional spin-boson model with a single qubit, time-ordering $\mathcal{T}$ plays no significant role, since it merely introduces a global phase to the unitary evolution operator. However, this is not the case for extended models with more than one qubit, where one must carefully deal with the effect of time-ordering $\mathcal{T}$. We therefore have

$$\widehat{U}^{\mathrm{I}}(t) = \exp\left[-i\int_0^t\sum_{\vec{k}}\widehat{Z}_{\vec{k}}e^{i\omega_{\vec{k}}\tau}\hat{b}^{\dagger}_{\vec{k}}d\tau\right]\times\widehat{\mathcal{A}}(t), \tag{20}$$

with

$$\begin{aligned}\widehat{\mathcal{A}}(t) = \mathcal{T}\Bigg\{\exp\Bigg[-i\int_0^t d\tau\left(e^{i\int_0^\tau\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\hat{b}^{\dagger}_{\vec{k}}(s)ds}\right) \\ \times\sum_{\vec{k}}\widehat{Z}^{\dagger}_{\vec{k}}e^{-i\omega_{\vec{k}}\tau}\hat{b}_{\vec{k}}\left(e^{-i\int_0^\tau\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\hat{b}^{\dagger}_{\vec{k}}(s)ds}\right)\Bigg]\Bigg\}.\end{aligned} \tag{21}$$

By using the prescription $e^{\beta\hat{b}^{\dagger}}\hat{b}e^{-\beta\hat{b}^{\dagger}} = \hat{b}-\beta$, the operator $\widehat{\mathcal{A}}(t)$ can be recast into

$$\begin{aligned}\widehat{\mathcal{A}}(t) &= \exp\Bigg[-i\int_0^t d\tau\sum_{\vec{k}}\widehat{Z}^{\dagger}_{\vec{k}}e^{-i\omega_{\vec{k}}\tau} \\ &\qquad\times\left(\hat{b}_{\vec{k}}-i\int_0^\tau\widehat{Z}_{\vec{k}}e^{i\omega_{\vec{k}}s}ds\right)\Bigg] \\ &= \exp\left[-i\int_0^t\sum_{\vec{k}}\widehat{Z}^{\dagger}_{\vec{k}}e^{-i\omega_{\vec{k}}\tau}\hat{b}_{\vec{k}}d\tau\right]\times\widehat{\mathcal{B}}(t).\end{aligned} \tag{22}$$

with

$$\widehat{\mathcal{B}}(t) = \exp\left[-\int_0^t\int_0^\tau\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\widehat{Z}^{\dagger}_{\vec{k}}e^{-i\omega_{\vec{k}}(\tau-s)}dsd\tau\right]. \tag{23}$$

Given that both $\widehat{A}$ and $\widehat{B}$ commute with $\left[\widehat{A},\widehat{B}\right]$, they satisfy $e^{\widehat{A}}e^{\widehat{B}} = e^{[\widehat{A},\widehat{B}]/2}e^{\widehat{A}+\widehat{B}}$. Then $\widehat{U}^{\mathrm{I}}(t)$ can easily be calculated:

$$\begin{aligned}\widehat{U}^{\mathrm{I}}(t) &= \exp\left[\frac{1}{2}\int_0^t\int_0^t\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\widehat{Z}^{\dagger}_{\vec{k}}e^{i\omega_{\vec{k}}(\tau-s)}dsd\tau\right] \\ &\quad\times\exp\left[-i\int_0^t\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\hat{b}^{\dagger}_{\vec{k}}(\tau)+\widehat{Z}^{\dagger}_{\vec{k}}\hat{b}_{\vec{k}}(\tau)d\tau\right]\times\widehat{\mathcal{B}}(t) \\ &= \exp\left[i\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\widehat{Z}^{\dagger}_{\vec{k}}\left(\frac{\omega_{\vec{k}}t-\sin\omega_{\vec{k}}t}{\omega^2_{\vec{k}}}\right)\right] \\ &\quad\times\exp\left[\sum_{\vec{k}}\widehat{Z}_{\vec{k}}\alpha_{\vec{k}}(t)\hat{b}^{\dagger}_{\vec{k}}-\widehat{Z}^{\dagger}_{\vec{k}}\alpha^{*}_{\vec{k}}(t)\hat{b}_{\vec{k}}\right],\end{aligned} \tag{24}$$

where $\alpha_{\vec{k}}(t) = -i\int_0^t e^{i\omega_{\vec{k}}\tau}d\tau = \left(1-e^{i\omega_{\vec{k}}t}\right)/\omega_{\vec{k}}$.

Assuming the direct-product initial state

$$\rho_{\mathrm{T}}(0) = \rho_1(0)\otimes\rho_2(0)\otimes\rho_{\mathrm{E}}(0), \tag{25}$$

the reduced dynamics of qubit-1, which we now consider to be our system, can be obtained by

$$\rho^{\mathrm{I}}_1(t) = \mathrm{Tr}_{2,\mathrm{E}}\left[\widehat{U}^{\mathrm{I}}(t)\rho_{\mathrm{T}}(0)\widehat{U}^{\mathrm{I}\dagger}(t)\right]. \tag{26}$$

The superscript I reminds that the dynamics is formulated in the interaction picture. One can easily show that the reduced dynamics of each qubit describes pure dephasing. We can thus apply the same method for constructing a Hamiltonian ensemble as for the conventional spin-boson model. We therefore focus on the time evolution of the off-diagonal element of qubit-1, which is written as

$$\rho^{\mathrm{I}}_{1,\downarrow\uparrow}(t) = \rho_{1,\downarrow\uparrow}(0)\left(\rho_{2,\uparrow\uparrow}(0)\phi^{(\mathrm{X})}(t)+\rho_{2,\downarrow\downarrow}(0)\phi^{(\mathrm{X})*}(t)\right), \tag{27}$$

where $\rho_{1,\downarrow\uparrow}(0)$, $\rho_{2,\uparrow\uparrow}(0)$, and $\rho_{2,\downarrow\downarrow}(0)$ are the initial conditions for the two qubits and the dephasing factor $\phi^{(\mathrm{X})}(t)$ is written as

$$\phi^{(\mathrm{X})}(t) = e^{-i2(\theta_{1,2}(t)+\theta_{2,1}(t))}\langle\prod_{\vec{k}} \widehat{D}^{\dagger}_{\vec{k},+}(t)\widehat{D}^{\dagger}_{\vec{k},-}(t)\rangle, \quad (28)$$

where

$$\begin{aligned}\theta_{j,j'}(t) &= \sum_{\vec{k}} g_{j,\vec{k}}g^{*}_{j',\vec{k}}\left(\frac{\omega_{\vec{k}}t-\sin\omega_{\vec{k}}t}{\omega^{2}_{\vec{k}}}\right)\\ &= \int_{0}^{\infty}\frac{\mathcal{J}_{j,j'}(\omega)}{\omega^{2}}\left(\omega t-\sin\omega t\right)d\omega, \qquad (29)\end{aligned}$$

$\mathcal{J}_{j,j'}(\omega)=\sum_{\vec{k}} g_{j,\vec{k}}g^{*}_{j',\vec{k}}\delta(\omega-\omega_{\vec{k}})$ are the spectral density functions, and

$$\begin{aligned}\widehat{D}_{\vec{k},+}(t) &= \exp\Big[\left(g_{1,\vec{k}}+g_{2,\vec{k}}\right)\alpha_{\vec{k}}(t)\hat{b}^{\dagger}_{\vec{k}}\\ &\quad -\left(g_{1,\vec{k}}+g_{2,\vec{k}}\right)^{*}\alpha^{*}_{\vec{k}}(t)\hat{b}_{\vec{k}}\Big]\\ \widehat{D}_{\vec{k},-}(t) &= \exp\Big[\left(-g_{1,\vec{k}}+g_{2,\vec{k}}\right)\alpha_{\vec{k}}(t)\hat{b}^{\dagger}_{\vec{k}}\\ &\quad -\left(-g_{1,\vec{k}}+g_{2,\vec{k}}\right)^{*}\alpha^{*}_{\vec{k}}(t)\hat{b}_{\vec{k}}\Big] \qquad (30)\end{aligned}$$

represent the displacement operators, respectively.

The coupling constants $g_{j,\vec{k}}$ of the two qubits to the boson environment 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, but with a phase difference:

$$g_{2,\vec{k}} = g_{1,\vec{k}}\,\exp[i\varphi]. \qquad (31)$$

For a thermalized environment at temperature T, the two prescriptions $\exp\left(\alpha\hat{b}^{\dagger}-\alpha^{*}\hat{b}\right)\exp\left(\beta\hat{b}^{\dagger}-\beta^{*}\hat{b}\right) = \exp\left[(\alpha\beta^{*}-\alpha^{*}\beta)/2\right]\exp\left[(\alpha+\beta)\hat{b}^{\dagger}-(\alpha+\beta)^{*}\hat{b}\right]$ and $\langle\exp\left(\alpha\hat{b}^{\dagger}-\alpha^{*}\hat{b}\right)\rangle = \exp\left[-\coth(\hbar\omega/2k_{\mathrm{B}}T)|\alpha|^{2}/2\right]$, are helpful for calculating the desired result

$$\phi^{(\mathrm{X})}(t) = \exp\left[-i\vartheta_{\varphi}(t)-\Phi(t)\right], \qquad (32)$$

where

$$\begin{aligned}\vartheta_{\varphi}(t) &= \cos\varphi\int_{0}^{\infty}\frac{4\mathcal{J}(\omega)}{\omega^{2}}(\omega t-\sin\omega t)d\omega \qquad (33)\\ &\quad +\mathrm{sign}(t)\sin\varphi\int_{0}^{\infty}\frac{4\mathcal{J}(\omega)}{\omega^{2}}(1-\cos\omega t)d\omega,\end{aligned}$$

$\mathcal{J}(\omega)=\sum_{\vec{k}}|g_{j,\vec{k}}|^{2}\delta(\omega-\omega_{\vec{k}})$ is the spectral density function, and

$$\Phi(t) = \int_{0}^{\infty}\frac{4\mathcal{J}(\omega)}{\omega^{2}}\coth\left(\frac{\hbar\omega}{2k_{\mathrm{B}}T}\right)(1-\cos\omega t)d\omega \qquad (34)$$

is the same as the one in the conventional spin-boson model. In the second line of Eq. (33), we have manually inserted $\mathrm{sign}(t)$. While this does not affect the pure dephasing dynamics for $t\geq 0$, it ensures that the condition $\phi^{(\mathrm{X})}(-t)=\phi^{(\mathrm{X})*}(t)$ is satisfied and one always obtains a real distribution $\wp^{(\mathrm{X})}(\omega)$.

The presence of $\vartheta_{\varphi}(t)$ in Eq. (32) will in general result in the violation of positivity. Note that, similar to the conventional spin-boson model, individual member Hamiltonians in the Hamiltonian ensemble must be of the form $\omega\hat{\sigma}_{z}/2$, which allows us to follow the same line of argument.

IV. OHMIC SPECTRAL DENSITY

To demonstrate the violation of positivity explicitly, we consider the Ohmic spectral density function

$$\mathcal{J}_{\mathrm{o1}}(\omega) = \omega\exp(-\omega/\omega_{\mathrm{c}}), \qquad (35)$$

and the zero temperature limit where $T\to 0$. For simplicity, we also assume a degenerate system Hamiltonian.

In the case of conventional spin-boson model, the dephasing factor is

$$\phi_{\mathrm{o1}}(t) = \frac{1}{\left(1+\omega_{\mathrm{c}}^{2}t^{2}\right)^{2}}, \qquad (36)$$

and the corresponding distribution is

$$\wp_{\mathrm{o1}}(\omega) = \frac{1}{4\omega_{\mathrm{c}}^{2}}(\omega_{\mathrm{c}}+|\omega|)\exp[-\frac{|\omega|}{\omega_{\mathrm{c}}}]. \qquad (37)$$

This is obviously a legitimate probability distribution without negative values. The results are shown in Fig. 2(a) of the main article. Consequently, the Hamiltonian ensemble $\{(\omega\hat{\sigma}_{z}/2,\wp_{\mathrm{o1}}(\omega))\}$ resembles the same pure dephasing dynamics of the conventional spin-boson model characterized by $\phi_{\mathrm{o1}}(t)$. As expected, $\wp_{\mathrm{o1}}(\omega)$ is, due to the degeneracy of the system Hamiltonian, centered at $\omega=0$, and broadens with increasing ω_{c}.

Whereas, for the extended model, the dephasing factor reads

$$\phi^{(\mathrm{X})}_{\mathrm{o1}}(t) = \frac{\exp\left[-i4\cos\varphi\left(\omega_{\mathrm{c}}t-\arctan(\omega_{\mathrm{c}}t)\right)\right]}{\left(1+\omega_{\mathrm{c}}^{2}t^{2}\right)^{2(1+i\mathrm{sign}(t)\sin\varphi)}}. \qquad (38)$$

Since the condition $\phi^{(\mathrm{X})}_{\mathrm{o1}}(-t)=\phi^{(\mathrm{X})*}_{\mathrm{o1}}(t)$ is fulfilled, the corresponding distribution $\wp^{(\mathrm{X})}_{\mathrm{o1}}(t)$ is real. However, the positivity of $\wp^{(\mathrm{X})}_{\mathrm{o1}}(t)$ is in general lost due to the presence of the nontrivial phase angle $\vartheta_{\varphi}(t)$. The results are show in Fig. 2(b) and (c) of the main article.