

When can quantum decoherence be mimicked by classical noise?

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Bing Gu^{1,a)}  and Ignacio Franco^{1,2,b)} 

AFFILIATIONS

¹Department of Chemistry, University of Rochester, Rochester, New York 14627, USA

²Department of Physics, University of Rochester, Rochester, New York 14627, USA

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^{a)}**Present address:** Department of Chemistry, University of California, Irvine, California 92697, USA.

^{b)}ignacio.franco@rochester.edu

ABSTRACT

Quantum decoherence arises due to uncontrollable entanglement between a system and its environment. However, the effects of decoherence are often thought of and modeled through a simpler picture in which the role of the environment is to introduce classical noise in the system's degrees of freedom. Here, we establish necessary conditions that the classical noise models need to satisfy to quantitatively model the decoherence. Specifically, for pure-dephasing processes, we identify well-defined statistical properties for the noise that are determined by the quantum many-point time correlation function of the environmental operators that enter into the system-bath interaction. In particular, for the exemplifying spin-boson problem with a Lorentz-Drude spectral density, we show that the high-temperature quantum decoherence is quantitatively mimicked by colored Gaussian noise. In turn, for dissipative environments, we show that classical noise models cannot describe decoherence effects due to relaxation through spontaneous emission of photons/phonons. These developments provide a rigorous platform to assess the validity of classical noise models of decoherence.

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I. INTRODUCTION

The inevitable interaction between a quantum system and its surrounding environment leads to decoherence.^{1–6} The decoherence occurs because such interaction leads to system-bath entanglement that turns a pure system state into a statistical mixture of states. Understanding quantum decoherence is important for a wide range of fields such as quantum computation and quantum information processing,⁷ quantum control,⁸ measurement theory, spectroscopy, and molecular structure and dynamics.⁹

There are several theoretical frameworks to understand quantum decoherence and the effective dynamics of open quantum systems.¹ The most rigorous one of them consists of *explicitly* solving the time-dependent Schrödinger equation for the system and its environment and then tracing out the environmental degrees of freedom to obtain the system's reduced density matrix. However, this approach, while desirable,^{6,10,11} is often intractable due to the exponentially increasing computational cost of solving the

time-dependent Schrödinger equation with system/environment size. This limitation has led to significant advances developing methods in which the effect of the bath is considered *implicitly*,^{1,12} such as perturbative quantum master equations,¹³ path integral techniques,¹⁴ and hierarchical equations of motion.^{15,16} Despite this important progress, following the reduced dynamics of a primary system of interest interacting with a general quantum environment remains an outstanding challenge.

Due to the conceptual and technical complexities in dealing with the system plus environment fully quantum mechanically, an alternative approach is to simply consider that the effect of the environment is to introduce classical noise in the system's degrees of freedom.^{17–25} In this picture, quantum dissipation is mimicked by stochastic terms in the equation of motion that introduce random transitions between system energy eigenstates. In turn, pure-dephasing processes are modeled by introducing dynamic disorder (or, equivalently, spectral diffusion) in which classical noise perturbs the energy of the system eigenstates,

leading to an accumulated random phase. Decoherence is simulated by averaging over an ensemble of these stochastic but unitary quantum dynamics.

The conceptual difference between decoherence and classical noise is known.^{2,26,27} In “true” decoherence, a single quantum system becomes entangled with environmental degrees of freedom. The unitary deterministic evolution of the system plus environment leads to a nonunitary evolution of the reduced density matrix of the system. By contrast, in the classical noise model, the decay of coherence can manifest itself only in an ensemble. This is because in any individual realization of the noise process the dynamics of the system is completely unitary since the system is not coupled to any external environment, and thus, no coherence can be lost from the system. The decay of coherence emerges by averaging over an ensemble of realizations of such a noise process. Nevertheless, unless this difference is probed explicitly, the noise model can mimic well the effects of decoherence since they both effectively lead to a damping of coherences. In fact, this stochastic picture with classical noise has been widely used in chemistry and physics to capture the loss of interference,^{19,25} optical line shapes,^{20,21} noise-assisted energy transport,²² non-Markovian dynamics,²³ Landau-Zener,^{28,29} and central-spin problems¹⁸ and in the quantum simulation of open many-body systems.²⁴

The fundamental question that arises in this context is what is the regime of validity and the limitations of the classical noise picture. An initial discussion of this problem was provided by Stern *et al.*¹⁹ where it is argued that the loss of quantum interference can be mimicked by the phase uncertainty introduced by the classical noise. However, no formal criteria for the validity of classical noise were provided. Here, we identify necessary conditions under which the decoherence effects induced by a quantum environment in a quantum system can be understood and modeled through classical noise. Such conditions are obtained by comparing the reduced dynamics of an open quantum system to the ensemble average of a series of unitary quantum trajectories generated by a stochastic Hamiltonian. We consider the effects of dissipation and pure dephasing independently and do not take into account their possible interference which was recently demonstrated in Ref. 4.

This paper is organized as follows. Section II introduces decoherence functions that arise due to system-bath entanglement and due to classical noise in the pure dephasing limit. Through a term-by-term comparison of their cumulant expansion, we isolate conditions on the classical noise that need to be satisfied to mimic the quantum dynamics. These conditions are determined by the many-point time correlation functions of the environment operators that enter into the system-bath interaction. The application of these conditions to the spin-boson model shows that the decoherence effects can be captured through colored Gaussian noise provided that the environment time-correlation function can be described by a set of exponentially decaying functions. In turn, Sec. III focuses on decoherence through quantum relaxation. We show that classical noise cannot describe decoherence induced by spontaneous emission and thus these models are of limited applicability when spontaneous fluctuations play a critical role. The analysis pertains to any spontaneous emission process that leads to dissipation such as the emission of phonons in vibrational environments or photons in electromagnetic environments.

II. PURE DEPHASING DYNAMICS

We first focus on pure dephasing dynamics and establish general criteria that need to be satisfied to employ classical noise to mimic quantum decoherence. Pure dephasing refers to a process in which the decoherence arises without energy transfer between the system and the environment. For a general composite system with Hamiltonian,

$$H = H_S + H_B + H_{SB}, \quad (1)$$

where H_S is the Hamiltonian of the quantum system, H_B is the Hamiltonian of the environment, and H_{SB} is the interaction between the system and the bath; the pure-dephasing condition arises when $[H_S, H_{SB}] = 0$. Even when this condition is not strictly satisfied, the pure-dephasing effects may still be the dominant effect when the environment dynamics is nonresonant with the transition frequencies of the system such that the dissipation is much slower compared to pure-dephasing effects. For this reason, the pure-dephasing limit has been useful in describing electronic decoherence in molecules,^{4,6} elastic electron-phonon interaction in solid state systems, loss of quantum interference,¹⁹ line shape in spectroscopic measurements,²⁰ vibrational dephasing in solvents,³⁰ and the central spin problem.¹⁸

Below, we define decoherence functions that arise from system-bath entanglement and from noise-induced pure dephasing. By contrasting them, we isolate conditions that the classical noise needs to satisfy to mimic the quantum decoherence.

A. Quantum decoherence function

For pure-dephasing dynamics, the system-bath interaction can be written as

$$H_{SB} = \sum_{\alpha} |\alpha\rangle\langle\alpha| \otimes B_{\alpha}, \quad (2)$$

where $\{|\alpha\rangle\}$ are the eigenstates of H_S and B_{α} is a bath operator. Here, we assume that the system and bath are uncorrelated at initial time such that the density matrix can be written as

$$\rho(0) = \rho_S(0) \otimes \rho_B(0), \quad (3)$$

where ρ_S is the reduced density matrix for the system and ρ_B is the reduced density matrix for the bath. The Liouville-von Neumann (LvN) equation in the interaction picture of $H_0 = H_S + H_B$ reads

$$i \frac{d}{dt} \tilde{\rho}(t) = [\tilde{H}_{SB}(t), \tilde{\rho}(t)], \quad (4)$$

where $\tilde{A}(t) = U_0^{\dagger}(t) A U_0(t)$ is the operator A in this interaction picture and $U_0(t) = e^{-iH_0 t}$. For notational convenience, for system operators, $\tilde{A}_S(t) \equiv U_S^{\dagger} A_S U_S$, where $U_S = e^{-iH_S t}$. Similarly, for bath operators, $\tilde{A}_B(t) \equiv U_B^{\dagger} A_B U_B$, where $U_B = e^{-iH_B t}$. Here and throughout, we employ atomic units where $\hbar = 1$. The solution to the LvN equation can be written as

$$\tilde{\rho}(t) = \tilde{U}(t) \rho(0) \tilde{U}^{\dagger}(t), \quad (5)$$

where $\tilde{U}(t) = \mathcal{T} e^{-i \int_0^t \tilde{H}_{SB}(t') dt'}$ is the propagator in the interaction picture and \mathcal{T} is the time-ordering operator. Using Eq. (2), it follows

that $\tilde{H}_{\text{SB}}(t) = \sum_{\alpha} |\alpha\rangle\langle\alpha| \otimes \tilde{B}_{\alpha}(t)$ and

$$\begin{aligned} \tilde{U}(t) &= \mathcal{T} \sum_{n=0}^{\infty} \frac{(-i)^n}{n!} \left(\int_0^t dt' \sum_{\alpha} |\alpha\rangle\langle\alpha| \otimes \tilde{B}_{\alpha}(t') \right)^n \\ &= \sum_{\alpha} |\alpha\rangle\langle\alpha| \otimes \mathcal{T} \sum_{n=0}^{\infty} \frac{(-i)^n}{n!} \left(\int_0^t dt' \tilde{B}_{\alpha}(t') \right)^n \\ &= \sum_{\alpha} |\alpha\rangle\langle\alpha| \otimes V_{\alpha}(t), \end{aligned} \quad (6)$$

where $V_{\alpha}(t) \equiv \mathcal{T} \exp(-i \int_0^t \tilde{B}_{\alpha}(t') dt')$. Inserting Eq. (6) into Eq. (5), taking into account the uncorrelated initial system-bath state in Eq. (3), and tracing over the bath degrees of freedom (which is denoted by $\text{Tr}_{\text{B}}[\dots]$) yields the reduced density matrix for the system

$$\tilde{\rho}_{\alpha\beta}^{\text{S}}(t) = \langle \alpha | \text{Tr}_{\text{B}}[\tilde{\rho}(t)] | \beta \rangle = \rho_{\alpha\beta}^{\text{S}}(0) \Phi_{\alpha\beta}(t). \quad (7)$$

Here,

$$\Phi_{\alpha\beta}(t) \equiv \text{Tr}_{\text{B}}[\rho_{\text{B}}(0) V_{\beta}^{\dagger}(t) V_{\alpha}(t)] = \langle V_{\beta}^{\dagger}(t) V_{\alpha}(t) \rangle \quad (8)$$

is the quantum decoherence function (QDF), which characterizes the decoherence effects for pure-dephasing dynamics. In this pure-dephasing dynamics, the diagonal matrix elements of the reduced density matrix representing populations in the energy eigenstates are not influenced by the environment as $\langle V_{\alpha}^{\dagger}(t) V_{\alpha}(t) \rangle = 1$. However, the off-diagonal elements of the density matrix decay with a rate determined by $\Phi_{\alpha\beta}(t)$.

If the initial state of the environment is pure, i.e., $\rho_{\text{B}}(0) = |\chi\rangle\langle\chi|$, the QDF becomes

$$\Phi_{\alpha\beta}(t) = \langle \chi | V_{\beta}^{\dagger}(t) V_{\alpha}(t) | \chi \rangle. \quad (9)$$

In this case, the absolute square of decoherence function $|\Phi_{\alpha\beta}|^2$ is known as the Loschmidt echo $L(t)$.³¹ The Loschmidt echo measures the stiffness of the environment to the perturbation by the system and is deeply connected to quantum decoherence.³² A particular interesting case is that for a two-level system with an initial state $|\psi_0\rangle = c_0|0\rangle + c_1|1\rangle$, the Loschmidt echo connects directly to the purity of the system, defined as $\mathcal{P}(t) = \text{Tr}_{\text{S}}[\rho_{\text{S}}^2(t)]$, with the following relationship:

$$\mathcal{P}(t) = 1 + 2|c_0|^2|c_1|^2(L_{01}(t) - 1). \quad (10)$$

B. Noise-induced decoherence function

Consider now a quantum system that is subject to classical noise. The noise is supposed to cause spectral diffusion, i.e., to introduce stochastic dynamics to the energy eigenvalues of the system. The effective Hamiltonian of the system for a particular realization of the noise is

$$H(t) = H_{\text{S}} + \sum_{\alpha} \eta_{\alpha}(t) |\alpha\rangle\langle\alpha|, \quad (11)$$

where $\{\eta_{\alpha}(t)\}$ are real stochastic processes. For the Hamiltonian to be Hermitian, $\eta_{\alpha}(t)$ must be real. The density matrix for a single realization of the noise can be obtained from the LvN equation in the interaction picture of H_{S} to yield

$$i \frac{d}{dt} \tilde{\rho}_{\alpha\beta}(t) = (\eta_{\alpha}(t) - \eta_{\beta}(t)) \tilde{\rho}_{\alpha\beta}(t). \quad (12)$$

Taking a statistical average of the solution of Eq. (12) yields

$$\overline{\tilde{\rho}_{\alpha\beta}(t)} = \Phi_{\alpha\beta}^{\text{noise}}(t) \rho_{\alpha\beta}(0), \quad (13)$$

where we have introduced the noise-induced decoherence function (NIDF)

$$\Phi_{\alpha\beta}^{\text{noise}}(t) = \overline{e^{-i \int_0^t \Delta_{\alpha\beta}(s) ds}}, \quad (14)$$

$\Delta_{\alpha\beta}(s) \equiv \eta_{\alpha}(s) - \eta_{\beta}(s)$, and the overline denotes statistical averaging.

C. Contrasting quantum and noise-induced decoherence functions

Comparing Eqs. (7) and (13), it is clear that if the classical decoherence function coincides with the quantum decoherence function, i.e.,

$$\Phi_{\alpha\beta}(t) = \Phi_{\alpha\beta}^{\text{noise}}(t) \quad \forall \alpha, \beta, \quad (15)$$

the noise picture of decoherence accurately mimics the entanglement process that leads to the decoherence. This formal relation offers a general structure to understand how classical noise models can be related to physical pure dephasing processes. However, it does not offer a practical prescription to relate the decoherence dynamics with the statistical properties of the noise as the quantum decoherence function involves two time-ordered exponentials of the bath operators which are generally not available.

To make further progress, below we introduce a useful operatorial identity for products of time-ordered exponentials and use it to develop a cumulant expansion of the quantum decoherence function.

1. A useful operatorial identity

We now show that given two general Hermitian operators $A(t)$ and $B(t)$,

$$\tilde{\mathcal{T}} e^{i \int_0^t B(\tau) d\tau} \mathcal{T} e^{-i \int_0^t A(\tau) d\tau} = \mathcal{T}_C e^{-i \int_0^t (A(\tau_+) - B(\tau_-)) d\tau}, \quad (16)$$

where $\tilde{\mathcal{T}}$ is the antichronological time-ordering operator and \mathcal{T}_C is the contour-ordering operator defined in a complex time contour \mathcal{C} as specified in Fig. 1. The antichronological time ordering operator rearranges earlier-time terms to the left of the later-time ones, and the contour-ordering operator rearranges earlier-in-contour terms to the right of the later-in-contour ones. This contour consists of two time branches: the upper branch going forward in time from

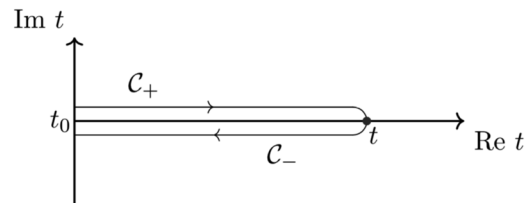


FIG. 1. The complex time contour that is used in Eq. (16).

$t_0 + i\epsilon \rightarrow t + i\epsilon$ and the lower one going backward in time from $t - i\epsilon \rightarrow t_0 - i\epsilon$, where $\epsilon = 0^+$ is an infinitesimal positive number.

Equation (16) can be understood as a direction extension of the semigroup property of the evolution operator [$U(t, t') = U(t, t'')U(t'', t')$] from real time to a complex time contour. A formal proof is provided as follows. We first note that

$$\bar{\mathcal{T}}_C e^{i \int_0^t B(\tau) d\tau} \mathcal{T}_C e^{-i \int_0^t A(\tau) d\tau} = \bar{\mathcal{T}}_C e^{i \int_0^t B(\tau_-) d\tau} e^{-i \int_0^t A(\tau_+) d\tau} \quad (17)$$

due to the fact that the effects of the two time-ordering operators in the left-hand side are being taken care of by the contour-ordering operator. Here, the subindex \pm indicates the upper/lower time branch of the contour. Using the Baker-Campbell-Hausdorff formula³³ $e^X e^Y = e^{X+Y+\frac{1}{2}[X,Y]+\dots}$ yields

$$\begin{aligned} \bar{\mathcal{T}}_C e^{i \int_0^t B(\tau_-) d\tau} e^{-i \int_0^t A(\tau_+) d\tau} &= \bar{\mathcal{T}}_C \exp \left\{ i \int_0^t (B(\tau_-) - A(\tau_+)) d\tau \right. \\ &\quad \left. - \frac{i^2}{2} \iint_0^t [B(\tau_-), A(\tau'_+)] d\tau d\tau' + \dots \right\}. \end{aligned} \quad (18)$$

Now, commutators vanish under the contour-ordering operator

$$\bar{\mathcal{T}}_C \{ [A(\tau), B(\tau')] \} = \bar{\mathcal{T}}_C \{ A(\tau)B(\tau') - B(\tau')A(\tau) \} = 0 \quad (19)$$

as the two terms will be ordered in the same way by the contour-ordering operator. Then, all commutators and nested commutators in Eq. (18) vanish, yielding the identity in Eq. (16).

The utility of Eq. (16) is that it enables us to express the two time-ordered exponentials in $\Phi_{\alpha\beta}(t)$ in terms of a single contour-ordered exponential. As shown below, such an exponential admits a simple cumulant expansion that will enable us to connect the desirable statistical properties of the noise with quantum time-correlation functions.

2. Decoherence function in the contour

Using Eq. (16), it follows that

$$V_\beta^\dagger(t) V_\alpha(t) = \bar{\mathcal{T}}_C \exp \left(i \int_0^t (\bar{B}_\beta(\tau_-) - \bar{B}_\alpha(\tau_+)) d\tau \right). \quad (20)$$

This equation can be simplified further if we define a function in the contour as

$$\mathcal{B}_{\alpha\beta}(\tau) = \theta_C(t - \tau) \bar{B}_\alpha(\tau) + \theta_C(\tau - t) \bar{B}_\beta(\tau), \quad (21)$$

where $\theta_C(\tau - \tau')$ is the Heaviside step function defined in the contour, $\theta_C(\tau - \tau') = 1$ if τ is later than τ' in the contour, and $\theta_C(\tau - \tau') = 0$ otherwise. Using this definition, Eqs. (20) and (8), the QDF can be written as a single contour-ordered exponential,

$$\Phi_{\alpha\beta}(t) = \left\langle \bar{\mathcal{T}}_C \left\{ e^{-i \int_C \mathcal{B}_{\alpha\beta}(\tau) d\tau} \right\} \right\rangle, \quad (22)$$

where the contour integral is defined as $\int_C = \int_{0+i\eta}^{t+i\eta} - \int_{0-i\eta}^{t-i\eta}$.

3. Cumulant expansion

With Eqs. (22) and (13), the condition Eq. (15) becomes

$$\overline{e^{-i \int_0^t \Delta_{\alpha\beta}(s) ds}} = \left\langle \bar{\mathcal{T}}_C e^{-i \int_C \mathcal{B}_{\alpha\beta}(\tau) d\tau} \right\rangle. \quad (23)$$

While formally exact, it is still nontrivial to directly infer from Eq. (23) whether it is possible to find random processes $\{\eta_\alpha(t)\}$ that satisfy it. Further progress can be made by performing a cumulant expansion for both sides of Eq. (23),

$$\begin{aligned} \ln \Phi_{\alpha\beta}(t) &\equiv K_{\alpha\beta}^q(t) = \sum_{n=1}^{\infty} \frac{(-i)^n}{n!} \kappa_{\alpha\beta}^{q,(n)}(t), \\ \ln \Phi_{\alpha\beta}^{\text{noise}}(t) &\equiv K_{\alpha\beta}^c(t) = \sum_n \frac{(-i)^n}{n!} \kappa_{\alpha\beta}^{c,(n)}(t). \end{aligned} \quad (24)$$

The cumulant expansion is the Taylor expansion of the logarithm of the decoherence function with respect to the system-bath coupling strength. This can be readily seen by parameterizing the system-bath interaction as $H_{SB} \rightarrow \lambda H_{SB}$.

For the classical and quantum decoherence functions to be equivalent irrespective of the system-bath interaction strength, the cumulants of $\Phi_{\alpha\beta}(t)$ and $\Phi_{\alpha\beta}^{\text{noise}}(t)$ need to match order by order. This condition is, in fact, stricter than Eq. (23). For the NIDF, the cumulant expansion can be obtained through the following recursive formula:³⁴

$$\kappa_{\alpha\beta}^{c,(n)} = \mu_{\alpha\beta}^{c,(n)} - \sum_{m=1}^{n-1} \binom{n-1}{m-1} \kappa_{\alpha\beta}^{c,(m)} \mu_{\alpha\beta}^{c,(n-m)}, \quad (25)$$

where

$$\mu_{\alpha\beta}^{c,(n)} = \int \dots \int_0^t \overline{\Delta_{\alpha\beta}(s_1) \dots \Delta_{\alpha\beta}(s_n)} ds_1 \dots ds_n \quad (26)$$

are the moments of the stochastic variable $\Delta_{\alpha\beta}$ and $\binom{n}{m}$ denote the binomial coefficients. One of the advantages of recasting the quantum decoherence function into a single exponential is that it becomes simpler to perform a cumulant expansion. A straightforward extension of the cumulant expansion for time-ordered exponentials by Kubo³⁵ leads to the conclusion that the quantum cumulants satisfy the same recursive formula Eq. (25), that is,

$$\kappa_{\alpha\beta}^{q,(n)} = \mu_{\alpha\beta}^{q,(n)} - \sum_{m=1}^{n-1} \binom{n-1}{m-1} \kappa_{\alpha\beta}^{q,(m)} \mu_{\alpha\beta}^{q,(n-m)}, \quad (27)$$

with the generalized quantum moments of operator $\mathcal{B}_{\alpha\beta}$ defined as

$$\mu_{\alpha\beta}^{q,(n)} = \int \dots \int_C \left\langle \bar{\mathcal{T}}_C \prod_{i=1}^n \mathcal{B}_{\alpha\beta}(\tau_i) \right\rangle \prod_{i=1}^n d\tau_i. \quad (28)$$

With the cumulant expansion for both sides of Eq. (23), the problem of whether classical noise can mimic quantum pure-dephasing dynamics can now be mapped to the much more manageable task of whether one can find a classical noise having correlation functions equivalent to the quantum time-correlation functions.

The first-order cumulant of the quantum and noise-induced decoherence function reads

$$\kappa_{\alpha\beta}^{q,(1)} = \int_C d\tau \langle \mathcal{B}_{\alpha\beta}(\tau) \rangle = \int_0^t \langle \bar{B}_\alpha(s) - \bar{B}_\beta(s) \rangle ds, \quad (29)$$

$$\kappa_{\alpha\beta}^{c,(1)} = \int_0^t \overline{\Delta_{\alpha\beta}(s)} ds = \int_0^t \overline{\eta_\alpha(s) - \eta_\beta(s)} ds. \quad (30)$$

At a quantum level, this cumulant is determined by the expectation value of the environment operators entering H_{SB} . At a noise level, it is determined by the expectation value of the noise. Since the expectation value of the environment operator is merely a real number, it is always possible to find noise with its average $\overline{\eta_\alpha(t)} = \langle \tilde{B}_\alpha(t) \rangle$ such that $\kappa_{\alpha\beta}^{q,(1)} = \kappa^{c,(1)}$.

A more stringent requirement comes from the second cumulant. As it is always possible to redefine the system Hamiltonian to make the expectation value of the environment operator vanish, we assume that the first cumulant vanishes in the following. From Eqs. (25)–(28), it is straightforward to obtain the second cumulant for the QDF and NIDF,

$$\kappa_{\alpha\beta}^{c,(2)} = \iint_0^t ds ds' \overline{\Delta_{\alpha\beta}(s) \Delta_{\alpha\beta}(s')} \quad (31)$$

and

$$\begin{aligned} \kappa_{\alpha\beta}^{q,(2)}(t) &= \iint_C d\tau d\tau' \langle \mathcal{T}_C \mathcal{B}_{\alpha\beta}(\tau) \mathcal{B}_{\alpha\beta}(\tau') \rangle \\ &= 2 \int_0^t ds \int_0^s ds' (D_{\alpha\alpha}(s, s') + D_{\beta\beta}(s', s)) \\ &\quad - 2 \iint_0^t ds ds' D_{\beta\alpha}(s, s'), \end{aligned} \quad (32)$$

where $D_{\alpha\beta}(s, s') = \langle \tilde{B}_\alpha(s) \tilde{B}_\beta(s') \rangle$ is the quantum time-correlation function of the environment. Because the classical noise is real, if the second cumulant for the QDF is complex, the classical noise cannot fully capture the effects of a quantum environment. Thus, a necessary condition to mimic the quantum decoherence with classical noise is that the cumulants are real.

Higher-order cumulants can be important for anharmonic and many-body environments. Using Eq. (25), it is now straightforward to obtain higher-order cumulants for QDF. For example, the third cumulant is given by

$$\kappa_{\alpha\beta}^{q,(3)} = \iiint_C \langle \mathcal{T}_C \mathcal{B}_{\alpha\beta}(\tau_1) \mathcal{B}_{\alpha\beta}(\tau_2) \mathcal{B}_{\alpha\beta}(\tau_3) \rangle d\tau_1 d\tau_2 d\tau_3. \quad (33)$$

If the higher-order quantum cumulants make significant contributions to decoherence, it requires the classical noise to have the corresponding higher-order correlations. This implies that, for such environments, the commonly used Gaussian noise model can be inadequate.^{18,36} We expect that such environments can arise in electronic decoherence in molecules where the environment consists of molecular vibrations which can be far from harmonic and also in central spin model where the environment consists of interacting spins.

Surprisingly, the cumulants, often considered as a convenient computational tool, carry direct physical meaning. To see this, we take the time-derivative of Eq. (7) and use the definition of the cumulants to obtain

$$\frac{d}{dt} \tilde{\rho}_{\alpha\beta}^S(t) = \dot{K}_{\alpha\beta}^q(t) \tilde{\rho}_{\alpha\beta}^S(t). \quad (34)$$

Equation (34) is the equation of motion for the coherences in the interaction picture. Clearly, the time-derivative of the cumulants is the generator of decoherence and each cumulant corresponds to a

particular order on the system-bath interaction. Explicitly, expressing the coherence in the polar form $\tilde{\rho}_{\alpha\beta}^S(t) = A_{\alpha\beta}(t) e^{i\phi_{\alpha\beta}(t)}$, it follows from Eq. (34) that

$$\dot{A}_{\alpha\beta}(t) = \text{Re} \dot{K}_{\alpha\beta}^q(t) A_{\alpha\beta}(t), \quad \dot{\phi}_{\alpha\beta}(t) = \text{Im} \dot{K}_{\alpha\beta}^q(t). \quad (35)$$

Equation (35) indicates that the real parts of the time-derivative of cumulants is responsible for decoherence, and the imaginary parts account for the environment-induced energy shifts.

D. Spin-boson model

We now illustrate how the above criteria can be applied using a concrete example: the quintessential spin-boson problem. The Hamiltonian for the pure-dephasing spin-boson model is

$$H = -\frac{\omega_0}{2} \sigma_z + \sigma_z \sum_k g_k (a_k^\dagger + a_k) + H_B, \quad (36)$$

where σ_z is the Pauli z matrix and ω_0 is the transition frequency for the two-level system. Here, $H_B = \sum_k \omega_k a_k^\dagger a_k$ describes a bosonic environment consisting of a distribution of harmonic oscillators of frequency ω_k with a_k, a_k^\dagger being the creation and annihilation operators for the k th mode, respectively. The coupling of the system with the environment leads to shifts in the system's energy levels, where g_k is the coupling constant to the k th harmonic mode.

The environment is assumed to be initially in thermal equilibrium at inverse temperature $\beta = 1/(k_B T)$ with density matrix $\rho_B = e^{-\beta H_B} / Z$, where $Z = \text{Tr}_B [e^{-\beta H_B}]$ is the partition function. For time-independent Hamiltonian, this leads to time-translational invariant time-correlation function

$$D_{\alpha\beta}(t, t') = D_{\alpha\beta}(t - t'). \quad (37)$$

For a two-level system, only one decoherence function has to be considered corresponding to $\alpha = 0, \beta = 1$. Since $\sigma_z = |0\rangle\langle 0| - |1\rangle\langle 1|$, one can identify $B_0 = -B_1 = \sum_k g_k (a_k + a_k^\dagger) \equiv B$ and $D_{00} = D_{11} = -D_{01} \equiv D$.

Using $\tilde{a}_k(t) = e^{-i\omega_k t} a_k$ and $\tilde{a}_k^\dagger(t) = e^{i\omega_k t} a_k^\dagger$, the time-correlation function $D(t)$ can be calculated as

$$\begin{aligned} D(t) &= \sum_k |g_k|^2 \left(\langle \tilde{a}_k(t) a_k^\dagger \rangle + \langle \tilde{a}_k^\dagger(t) a_k \rangle \right) \\ &= \sum_k |g_k|^2 [(1 - \tilde{n}_k) e^{-i\omega_k t} + \tilde{n}_k e^{i\omega_k t}], \end{aligned} \quad (38)$$

where $\tilde{n}_k = \langle a_k^\dagger a_k \rangle$ is the distribution function. At thermal equilibrium, $\tilde{n}_k = 1/(e^{\beta\omega_k} - 1)$ corresponding to the Bose-Einstein distribution and Eq. (38) yields

$$\begin{aligned} D(t) &= \int_0^\infty \frac{d\omega}{\pi} J(\omega) [\coth(\beta\omega/2) \cos(\omega t) - i \sin(\omega t)] \\ &= \int_{-\infty}^\infty \frac{d\omega}{2\pi} J(\omega) [\coth(\beta\omega/2) \cos(\omega t) - i \sin(\omega t)], \end{aligned} \quad (39)$$

where the spectral density is defined as $J(\omega) \equiv \pi \sum_k |g_k|^2 \delta(\omega - \omega_k)$ for $\omega > 0$ and extended to negative frequencies by $J(-\omega) = -J(\omega)$. This extension makes the integrand in Eq. (39) symmetric under

$\omega \rightarrow -\omega$, hence the second equality. Since the environment is Gaussian, the QDF is determined by the first two cumulants.^{35,37} The first cumulant vanishes, and the second cumulant can be calculated by inserting Eq. (39) into Eq. (32),

$$\kappa_{\alpha\beta}^{q,(2)}(t) = 8 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} J(\omega) \coth(\beta\omega/2) \frac{1 - \cos(\omega t)}{\omega^2}. \quad (40)$$

Interestingly, the cumulant is real even though the time-correlation function is complex. This is due to the property of the quantum time-correlation function

$$D(-\tau) = D^*(\tau). \quad (41)$$

Because the cumulant is real, as described below, its effects on the dynamics can be mimicked by classical noise.

Consider now the noise model intended to mimic the above decoherence dynamics with Hamiltonian

$$H(t) = -\frac{\omega_0}{2} \sigma_z + \eta(t) \sigma_x, \quad (42)$$

where the stochastic process $\eta(t)$ replaces the system-bath interaction in Eq. (36). Denoting the noise correlation function as $C(s, s') = \overline{\eta(s)\eta(s')}$, we show that if the noise satisfies the following three conditions: (i) $C(s, s') = C(s - s')$, (ii) $\overline{\eta(t)} = 0$, and (iii) $C(t) = S(t)$, where $S(t) \equiv \frac{1}{2} \langle \{B(t), B\} \rangle = \frac{1}{2} \langle B(t)B + BB(t) \rangle$, then the NIDF coincides with the QDF. The first condition implies that the noise is stationary corresponding to the equilibrium state of the environment. The second condition reflects the vanishing of the first cumulant of the QDF. The third one is required to make the second cumulants for QDF and NIDF equal. To see this, realizing that $\Delta_{01}(t) = 2\eta(t)$ and inserting the Fourier transform of the noise correlation function

$$C(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} C(\omega) e^{-i\omega t} \quad (43)$$

into Eq. (31) yields

$$\kappa^{c,(2)}(t) = 8 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{1 - \cos(\omega t)}{\omega^2} C(\omega). \quad (44)$$

Comparing Eqs. (40) and (44), it is clear that the condition $\kappa^{q,(2)}(t) = \kappa^{c,(2)}(t)$ is equivalent to

$$C(\omega) = J(\omega) \coth(\beta\omega/2). \quad (45)$$

According to Eq. (39), the right-hand side of Eq. (45) is the Fourier transform of the real part of the quantum time-correlation function [Eq. (39)]. Using Eq. (41), it follows that $S(t) = \text{Re } D(t)$ and thus to the third condition $S(t) = (1/2)(D(t) + D(-t)) = (1/2)\langle \{B(t), B\} \rangle$.

Equation (45) suggests that for each spectral density there is a corresponding classical noise leading to the same pure-dephasing dynamics provided that an adequate algorithm to generate the stochastic process is identified. Here, we exemplify the analysis with the widely used Ohmic environments with a Lorentz-Drude cutoff. The spectral density for such environments is

$$J(\omega) = 2\lambda \frac{\omega_c \omega}{\omega^2 + \omega_c^2}, \quad (46)$$

where ω_c is the cutoff frequency of the environment and λ characterizes the system-bath interaction strength. In the high-temperature limit $\beta\omega_c \ll 1$, $\coth(\beta\omega/2) \approx 2(\beta\omega)^{-1}$ and

$$J(\omega) \coth(\beta\omega/2) \approx 4\lambda k_B T \frac{\omega_c}{\omega^2 + \omega_c^2}. \quad (47)$$

Now, let $\eta(t)$ be a colored Gaussian noise with correlation function $C(\tau) = 2\lambda k_B T e^{-\omega_c \tau}$. This choice ensures that Eq. (45) is satisfied in the high temperature limit which can be seen by taking the Fourier transform of the noise correlation function and comparing with Eq. (47). Therefore, the quantum pure-dephasing effects of a high-temperature Ohmic bath can be fully captured by colored exponentially correlated Gaussian noise.

This conclusion is demonstrated in Fig. 2, which contrasts the exact quantum results with stochastic simulations. The exact results are obtained by first inserting Eq. (47) into Eq. (40) to obtain the second-order cumulant and thus the decoherence function $\Phi_{01}(t) = e^{-\frac{1}{2}\kappa^{q,(2)}(t)}$. This decoherence function is exact (compare with Ref. 38) as the contributions of higher order cumulants vanish

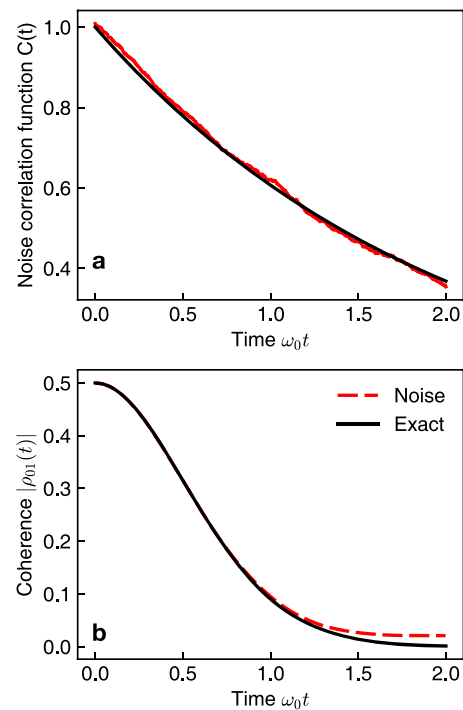


FIG. 2. (a) Correlation function of the generated noise (red) in comparison with the target (black). (b) Quantum and noise-induced decoherence dynamics in a spin-boson model starting from a superposition with equal coefficients of ground and excited states. Model parameters are $\lambda/\omega_0 = 0.5$, $\beta\omega_0 = 1$, $\omega_c/\omega_0 = 1$. The exact results are obtained through $\Phi_{01}(t) = e^{-\frac{1}{2}\kappa^{q,(2)}(t)}$ with the second cumulant computed using Eq. (40). The stochastic simulations are obtained with 2000 realizations of the colored noise and with a time step $\omega_0 dt = 0.002$. No revivals of the coherence are observed in this model.

in this case. The stochastic simulation is averaged over 2000 realizations of the exponentially correlated colored Gaussian noise generated using the algorithm in Ref. 39. The correlation function of generated noise is shown in Fig. 2(a). For each realization, the stochastic time dependent Schrödinger equation $i\frac{d}{dt}|\psi(t)\rangle = H(t)|\psi(t)\rangle$ with the initial condition $|\psi(0)\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ is integrated. As shown, the decoherence dynamics obtained with stochastic noise is in quantitative agreement with the exact quantum decoherence dynamics, consistent with our conclusion above.

For a low-temperature regime and other types of spectral densities, if $S(t)$ can be well-described by a set of exponential functions,

$$S(t-t') = \sum_n |c_n|^2 e^{-|t-t'|/\tau_n}, \quad (48)$$

one can choose a sum of exponentially colored Gaussian noises,

$$\eta(t) = \sum_n c_n \eta_n(t), \quad (49)$$

where $\{\eta_n(t)\}$ are Gaussian stochastic processes with statistical properties,

$$\overline{\eta_n(t)\eta_m^*(t')} = \delta_{nm} e^{-|t-t'|/\tau_n}. \quad (50)$$

In this case, the noise correlation function

$$C(t-t') = \sum_{n,m} c_n c_m^* \overline{\eta_n(t)\eta_m^*(t')} = \sum_n |c_n|^2 e^{-|t-t'|/\tau_n} = S(t-t'). \quad (51)$$

Thus, the quantum decoherence dynamics can still be captured by classical noise.

III. QUANTUM DISSIPATION

Another major source of decoherence is quantum dissipation due to transitions between system eigenstates induced by the environment. The role of the dissipative environment is to drive the system from an initially out-of-equilibrium state to thermal equilibrium.

The question we seek to address here is when can we understand quantum decoherence induced by dissipation in terms of classical noise. This problem has been studied previously by Tanimura and Kubo¹⁶ with the hierarchical equation of motion. The conclusion of such a formal study is that the classical noise can only be made to be equivalent to a full quantum treatment at infinite temperature, i.e., as $\beta \rightarrow 0$. Below, we provide a simpler analysis of this problem for Markovian environments and show that the physical reason behind this conclusion is that the classical noise cannot describe the decoherence effects due to spontaneous emission induced by a dissipative environment. Here, spontaneous emission is not restricted to electromagnetic environments but refers to a damping effect induced by the spontaneous fluctuations of any dissipative environment.

The simplest model that allows isolating this basic physics is a two-level system $|g\rangle, |e\rangle$ interacting with a thermal environment. A standard full quantum treatment of this model within the dipole approximation leads to the equation of motion for the reduced density matrix,⁴⁰

$$\begin{aligned} \frac{d}{dt}\rho_S(t) = & -i[H_S, \rho_S] + \Gamma_e \left(\sigma_- \rho_S(t) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho_S(t) \} \right) \\ & + \Gamma_a \left(\sigma_+ \rho_S(t) \sigma_- - \frac{1}{2} \{ \sigma_- \sigma_+, \rho_S(t) \} \right), \end{aligned} \quad (52)$$

where $H_S = -\omega_0 \sigma_z/2$ is the system Hamiltonian, σ_{\pm} is the raising/lowering operator, and $[A, B] = AB - BA$ and $\{A, B\} = AB + BA$ denote the commutator and anticommutator, respectively. The first term in the right-hand side of Eq. (52) accounts for the unitary dynamics of H_S , which does not contribute to decoherence. The meaning of the remaining dissipative terms is best revealed by decomposing Eq. (52) in terms of the matrix elements,

$$\frac{d}{dt}\rho_{gg}^S(t) = \Gamma_e \rho_{ee}^S(t) - \Gamma_a \rho_{gg}^S(t), \quad (53)$$

$$\frac{d}{dt}\rho_{eg}^S(t) = -i\omega_0 \rho_{eg}^S(t) - \Gamma_d \rho_{eg}^S(t), \quad (54)$$

where $\Gamma_d = (\Gamma_e + \Gamma_a)/2$. Clearly, the second term in Eq. (52) accounts for the emission of energy to the environment and the third one to absorption. Here, the emission rate Γ_e is a sum of the stimulated emission rate (which is equivalent to the absorption rate Γ_a) and spontaneous emission rate Γ_0 , i.e., $\Gamma_e = \Gamma_a + \Gamma_0$. The off-diagonal matrix elements (or coherence) represented in the eigenstates of H_S admits an exponential decay with the decoherence rate Γ_d .

Note that as a consequence of the Markovian approximation involved in the derivation of Eq. (52), the model does not capture the universal initial Gaussian purity decay for uncorrelated initial states which gives rise to quantum Zeno effects.^{41,42}

Now, consider the classical noise picture where the system is subject to a random term that induces transitions between system eigenstates, i.e.,

$$H = H_S + \eta(t)\sigma_- + \eta^*(t)\sigma_+. \quad (55)$$

Here, the stochastic variable η is allowed to be complex but still keeping the dynamics for each noise realization unitary. For Markovian environments without memory effects, it is appropriate to choose $\langle \eta(t)\eta^*(t') \rangle = \gamma \delta(t-t')$. In the interaction picture of H_S , the Liouville-von Neumann equation reads

$$i\frac{d}{dt}\tilde{\rho}_S(t) = [\eta(t)\tilde{\sigma}_-(t) + \eta^*(t)\tilde{\sigma}_+(t), \tilde{\rho}_S(t)]. \quad (56)$$

A quantum master equation can be obtained as follows. Integrating Eq. (56) yields

$$\tilde{\rho}_S(t) = \rho_S(0) - i \int_0^t dt' [\eta(t')\tilde{\sigma}_-(t') + \eta^*(t')\tilde{\sigma}_+(t'), \tilde{\rho}_S(t')]. \quad (57)$$

Inserting Eq. (57) back into the right-hand side of Eq. (56) and taking statistical average of the stochastic processes yields

$$\frac{d}{dt}\tilde{\rho}_S(t) = \gamma[\tilde{\sigma}_-(t), [\tilde{\sigma}_+(t), \tilde{\rho}_S(t)]] + \gamma[\tilde{\sigma}_+(t), [\tilde{\sigma}_-(t), \tilde{\rho}_S(t)]]. \quad (58)$$

Transforming into the Schrödinger picture gives the quantum master equation

$$\begin{aligned} \dot{\rho}_S(t) = & -i[H_S, \rho_S(t)] + \gamma \left(\sigma_- \rho_S(t) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho_S(t) \} \right) \\ & + \gamma \left(\sigma_+ \rho_S(t) \sigma_- - \frac{1}{2} \{ \sigma_- \sigma_+, \rho_S(t) \} \right). \end{aligned} \quad (59)$$

Comparing Eqs. (59) and (52), it becomes clear that the noise can mimic many of the effects of the quantum relaxation provided that one identifies γ with Γ_a . What becomes missing in this picture are the contributions due to spontaneous emission. In this case, one obtains a decoherence rate $\gamma_d = \Gamma_a$ from Eq. (59). Thus, the decoherence rate in the classical noise picture does not contain the contribution from spontaneous emission.

For photonic environments at thermal equilibrium, a criterion for the importance of spontaneous emission to decoherence can be identified. In this case, the ratio between the spontaneous and the stimulated emission rate is given by³⁸

$$\eta \equiv \frac{\Gamma_0}{\Gamma_a} = \frac{1}{N(\omega_{eg})} = e^{\beta\omega_{eg}} - 1, \quad (60)$$

where $N(\omega_{eg}) = 1/(e^{\beta\omega_{eg}} - 1)$ is the Bose-Einstein distribution function. In the high-temperature limit $\beta\omega_{eg} \ll 1$, $\eta \rightarrow 0$ such that the spontaneous emission plays a negligible role in decoherence. On the other hand, in the low-temperature regime $\beta\omega_{eg} \gg 1$, $\eta \gg 1$ and the spontaneous emission dominates.

The missing of spontaneous emission has a direct consequence in relaxation. Since the absorption and emission rates are equal, the stationary state at long times is the nonphysical infinite-temperature state. The above analysis demonstrates that any Hamiltonian in the form of Eq. (55) will necessarily lead to the master equation Eq. (59). To fix this problem and introduce spontaneous emission, one has to go beyond the classical noise model with Hamiltonian dynamics, for example, by promoting the classical noise to quantum noise,⁴³ by relaxing the constraint of unitary dynamics for each noise realization⁴⁴ as in the stochastic Liouville equation,⁴⁵ or by including phenomenological corrections to the equations of motion that force thermalization as in Refs. 46–48. These results agree with the analysis of Burgarth *et al.*⁴⁴ which shows that dissipation can only be ascribed to a classical noise process if probability is not conserved in individual realizations.

IV. CONCLUSIONS

To summarize, we have contrasted quantum decoherence that arises as a single quantum system becomes entangled with environmental degrees of freedom with the apparent decoherence that results by averaging over an ensemble of unitary evolutions generated by a Hamiltonian subject to classical noise. For dissipative environments, we showed that the classical noise cannot describe the decoherence induced by spontaneous emission and, thus, that the classical noise picture can only become quantitative in the infinite temperature limit. For pure-dephasing dynamics, we identified general conditions that determine whether the decoherence dynamics due to a quantum environment can be quantitatively mimicked

through classical noise. Specifically, we showed that for the two dynamics to agree, the cumulants of the quantum and noise-induced decoherence functions must coincide. These requirements impose restrictions on the statistical properties of the noise that are determined by the quantum many-point time correlation functions of the environmental operators that enter into the system-bath interaction. These conditions are valid for any pure dephasing problem including anharmonic environments and nonlinear system-bath couplings.

In particular, through the spin-boson model, we demonstrated numerically and analytically that the decoherence effects due to a harmonic Ohmic environment (in the high-temperature pure-dephasing limit) can be mimicked by exponentially correlated colored Gaussian noise. This observation is consistent with a recent study⁴⁹ of the quantum transport properties of a molecular junction subject to vibrational dephasing that finds agreement between a fully quantum model (harmonic, Ohmic, pure-dephasing environment in the high temperature limit) and a model in which the thermal environment manifests itself in (exponentially correlated Gaussian) fluctuating site energies. A challenge in employing classical noise models for environments with more complicated spectral densities is to generate noise with the correct statistical properties. A possible strategy to this end is to perform classical molecular dynamics to model with chemical detail the influence of the environment on the system.^{50,51}

Our results offer well-defined criteria to develop and to understand the validity of classical noise models of decoherence that are employed in chemistry, physics, and quantum information.⁵² These models are useful in the design of dynamic decoupling schemes to preserve coherence.¹⁸ In particular, in the context of optimal control computations, an effective stochastic model that captures the effects of a quantum environment is highly desirable⁵³ as those computations are challenging for a full quantum model.

In addition, the developed criterion in Eq. (23) can also be used to test the validity of mixed quantum-classical methods in decoherence research. Equation (23) also applies to hybrid quantum-classical schemes where the bath is treated classically and the effect of the bath on the system appears as a time-dependent term in the Hamiltonian, such as Ehrenfest dynamics^{6,47,54} and approaches based on (thermal and nonthermal) classical molecular dynamics for the bath.^{50,51} As is the case for noise-based strategies, this class of methods do not capture spontaneous emission and thus cannot lead to thermal equilibrium unless phenomenological corrections^{46–48} to the equations of motion are incorporated.

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