Open Systems with Error Bounds: Spin-Boson Model with Spectral Density Variations

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(Received 10 November 2016; published 7 March 2017)

In the study of open quantum systems, one of the most common ways to describe environmental effects on the reduced dynamics is through the spectral density. However, in many models this object cannot be computed from first principles and needs to be inferred on phenomenological grounds or fitted to experimental data. Consequently, some uncertainty regarding its form and parameters is unavoidable; this in turn calls into question the accuracy of any theoretical predictions based on a given spectral density. Here, we focus on the spin-boson model as a prototypical open quantum system, find two error bounds on predicted expectation values in terms of the spectral density variation considered, and state a sufficient condition for the strongest one to apply. We further demonstrate an application of our result, by bounding the error brought about by the approximations involved in the hierarchical equations of motion resolution method for spin-boson dynamics.

DOI: 10.1103/PhysRevLett.118.100401

Introduction.—One of the most fundamental models of open quantum systems is the spin-boson model, which comprises a two-level system, such as a spin-1/2 particle, and a large number of quantum harmonic oscillators linearly coupled to it and acting as the environment [1–4]. The influence of these degrees of freedom on the dynamics of the spin can be computed from the strength of the couplings between the spin and each oscillating mode and the frequency of the modes; these quantities can be combined to determine the spectral density of the environment, a function of frequency closely related to its internal correlations and their effect on the reduced dynamics of the spin. Depending on the number of harmonic oscillators present in the model and their dispersion, the spectral density may be a continuous function (for an uncountably infinite set of oscillators) or a linear combination of Dirac delta functions centered at some particular frequencies (for a finite or countably infinite set); the former type is convenient for analytical treatments, while the latter is necessary when performing numerical studies. Generally speaking, the spectral density is not a fundamental object, but rather a phenomenological quantity obtained by making assumptions on the kind of system under study or by fitting experimental data; hence, it is wise to keep in mind that there may always be some error in the functional form considered. This raises the question of how accurate any predictions for a given model can be, given the uncertainty in the spectral density of its environment.

To the best of our knowledge, it appears that no general, rigorous error bound to theoretical predictions for the spin dynamics with respect to changes in the spectral density exists in the literature. The purpose of this work is to address this issue and derive an upper bound to the deviation of the time-dependent expectation value of some spin observable $\hat{O}$ when the spectral density of the oscillator bath changes by a known amount. Aside from the mathematical motivation, such a result would be desirable from a physical point of view for two main reasons. First, when using spectral densities obtained from experiment, it would give a quantitatively certified range for theoretical results to be compatible with them, which can be helpful in order to determine the physical soundness of the theoretical models used. Second, it would make it possible to bound the error associated with numerical solutions for the spin-boson model, in analogy with, e.g., error bounds on the time-evolved density using orthogonal polynomials algorithm (TEDOPA) [5,6], whenever the method used entails some degree of approximation to the original spectral density of the problem at hand.

We derive our error bounds in the coherent-state path-integral formalism [7,8] using the Feynman-Vernon influence functional [9,10]: the idea behind this approach is to treat variations of the spectral density analytically with functional methods, without relying on approximations or numerical techniques. The final results are expressed in terms of canonical quantities such as the interaction Hamiltonian and the bath correlation function, with no need to refer to the path-integral expressions used in the derivation. We will state two forms of the bound, one stronger than the other at long times, and give a sufficient condition for the strong bound to apply, as well as a few examples of spectral density variations complying with it.

To provide a quantitative example of how our results can be used to certify the accuracy of numerical schemes, we will apply our error bound to the well-known hierarchical equations of motion (HEOM) resolution method [11,12] for spin-boson dynamics.

Model.—Consider the spin-boson Hamiltonian [2]

$$\begin{aligned}\hat{H} &= \hat{H}_S \otimes \mathbb{I}_B + \mathbb{I}_S \otimes \hat{H}_B + \hat{H}_I \\ &= \left(\frac{\epsilon}{2}\sigma_z + \frac{\Delta}{2}\sigma_x\right) \otimes \mathbb{I}_B + \mathbb{I}_S \otimes \int_0^\infty dk \omega_k \hat{a}_k^\dagger \hat{a}_k \\ &\quad + \frac{\lambda}{2}\sigma_z \otimes \int_0^\infty dk h(k)(\hat{a}_k^\dagger + \hat{a}_k), \end{aligned} \tag{1}$$

where $\hat{a}_k$ and $\hat{a}_k^\dagger$ are bosonic creation and annihilation operators satisfying the commutation relation $[\hat{a}_k, \hat{a}_l^\dagger] = \delta_{kl}$, the continuous label k identifying each boson may be thought of as a momentum variable, and ω_k as a dispersion relation, which we take to be linear: $\omega_k = gk$, with unit g and in natural units with $\hbar = 1$; the function $h(k)$ expresses the coupling strength between each mode and the qubit. Depending on the choice of h (which may well include delta functions), the model may have a finite, countably infinite or uncountably infinite number of bosons, with k bounded or unbounded; we will always assume the domain of h to be the whole positive k axis. With $\omega_k = k$, there is a one-to-one correspondence between the choice of $h(k)$ and the spectral density $J(\omega)$ of the bosonic environment: $J(\omega_k) = \pi h^2(\omega_k)$ [13–15].

To complete our ansatz, we take the initial state to be of the form $\hat{\rho}_0 = \hat{\rho}_{S0} \otimes [(e^{-\beta \hat{H}_B}/\mathrm{Tr}_B(e^{-\beta \hat{H}_B})]$, where $\hat{\rho}_{S0}$ is arbitrary and the bosons are in thermal equilibrium at temperature $T = (1/k_B\beta)$. In principle, this assumption could be relaxed to include more general Gaussian initial states for the bath [2], such as a thermal state perturbed by a laser pulse before the interaction with the spin begins, but for the sake of simplicity, we will keep our treatment within the standard framework of thermal environments for the time being and leave extensions to this first model to our future work.

Under these assumptions, the expectation value of some spin observable $\hat{O}$ at time t,

$$\langle \hat{O}(t) \rangle = \mathrm{Tr}(\hat{O} e^{-i\hat{H}t} \hat{\rho}_0 e^{i\hat{H}t}), \tag{2}$$

depends on the environment only via $J(\omega)$ [10]. We will use path-integral tools in order to quantify and bound the dependence of $\langle \hat{O}(t) \rangle$ on J: in order to better emphasize this concept, in what follows we shall refer to expectation values specifying the relevant spectral density as a subscript, effectively regarding $\langle \hat{O}(t) \rangle_J$ as a functional on the space of spectral densities as well as a function of time.

Spectral density variations and error bounds.—We want to compare the expectation values of $\hat{O}$ for arbitrary spectral densities $J_0(\omega)$ and $J(\omega) \coloneqq J_0(\omega) + \Delta J(\omega)$: in other words, we are interested in bounding the absolute value of the difference

$$\Delta \langle \hat{O}(t) \rangle \coloneqq \langle \hat{O}(t) \rangle_J - \langle \hat{O}(t) \rangle_{J_0}. \tag{3}$$

The path-integral formalism [10,16–22] makes it easy to eliminate the bosonic degrees of freedom from the expression for the expectation value of $\hat{O}(t)$ by performing the relevant Gaussian integral analytically [23]. The result is a path integral for the spin variables alone, with the time evolutions of the left and right part of the initial state no longer independent. The Feynman-Vernon influence functional encodes this mixing, which is a result of the partial trace over the bath: it has the form of a Gaussian functional of the spin variables, with the bath correlation function

$$\xi_J(t) \coloneqq \int_0^\infty \frac{d\omega}{\pi} J(\omega) \left[\coth\left(\frac{\beta\omega}{2}\right) \cos(\omega t) + i \sin(\omega t) \right]$$

coupling them. Note that since we have assumed a thermal initial state for the bosons, which is stationary with respect to their free dynamics, $\xi_J(t)$ is not a function of two time variables, but merely of their difference.

To write out $\langle \hat{O}(t) \rangle_J$ explicitly in terms of $J_0(\omega)$ and $\Delta J(\omega)$, we define the Heisenberg-picture operator $\hat{h}_I(t) \coloneqq e^{i\hat{H}t}[(\lambda/2)\sigma_z \otimes \mathbb{I}_B] e^{-i\hat{H}t}$ and the superoperator

$$\hat{\Phi}[\hat{h}_I, \hat{h}'_I, J] \coloneqq \mathcal{T} \int_0^t dt' \int_0^{t'} dt'' [\hat{h}_I(t') - \hat{h}'_I(t')] \times [\xi_J(t' - t'')\hat{h}_I(t'') - \xi_J^*(t' - t'')\hat{h}'_I(t'')], \tag{4}$$

which acts on a spin state $\hat{\rho}_0$ with all $\hat{h}_I(t)$ operators multiplying it from the left and all $\hat{h}'_I(t)$ from the right in the appropriate time order. This is, up to an overall minus sign, the operator version of the logarithm of the influence functional [23]. Using the exponential form of the Feynman-Vernon functional and the fact that $\hat{\Phi}[\hat{h}_I, \hat{h}'_I, J]$ is linear in the spectral density, it can be shown [23] that

$$\Delta \langle \hat{O}(t) \rangle = \sum_{n=1}^{\infty} \frac{\langle \hat{O}(t) \mathcal{T}(-\hat{\Phi}[\hat{h}_I, \hat{h}'_I, \Delta J])^n \rangle_{J_0}}{n!}. \tag{5}$$

Note that extending the same series by including the term with $n = 0$ just adds $\langle \hat{O}(t) \rangle_{J_0}$, giving $\langle \hat{O}(t) \rangle_J$.

The series in Eq. (5) may be bounded in magnitude term by term, using the singular-value decomposition of the spin operators to remove the complicated time dependence of the time-ordered correlation functions, and then summed: the result is the general formula

$$|\Delta \langle \hat{O}(t) \rangle| \leq ||\hat{O}|| \left(e^{\lambda^2 \int_0^t dt' \int_0^{t'} dt'' |\Delta\xi(t' - t'')|} - 1 \right), \tag{6}$$

where $\Delta\xi(t) \coloneqq \xi_J(t) - \xi_{J_0}(t) = \xi_{\Delta J}(t)$, and we have used the operator norm $\|\hat{O}\| \coloneqq \|\hat{O}\|_\infty = \sigma_1(\hat{O})$, $\sigma_1(\hat{O})$ being the highest singular value of $\hat{O}$.

Depending on $\Delta J(\omega)$, there are two options for bounding the double time integral in Eq. (6): the worst-case scenario is a $\Delta\xi(t)$, which never decays, as would be the case for singular contributions such as $\Delta J(\omega) = \kappa\delta(\omega - \omega_0)$. Then one would be forced to bound $|\Delta\xi(t)|$ by some constant $C > 0$, obtaining the error bound

$$|\Delta \langle \hat{O}(t) \rangle| \leq \|\hat{O}\| (e^{\lambda^2 C t^2/2} - 1). \tag{7}$$

However, if $\Delta J(\omega)$ is such that the resulting $\Delta\xi(t)$ decays fast enough to be absolutely integrable, i.e.,

$$\int_0^\infty dt|\Delta\xi(t)| = c < \infty, \tag{8}$$

then one can tighten the bound considerably. This is the case for many physically relevant situations, e.g., for Ohmic, superohmic, or antisymmetrized Lorentzian spectral density variations [23]. In practice, it is often easier to apply the triangle inequality to $|\Delta\xi(t)|$ first and then bound its real and imaginary parts separately, even though this may weaken the bound slightly: the result is

$$|\Delta\langle\hat{O}(t)\rangle| \leq \|\hat{O}\|(e^{\lambda^2[\gamma(\beta)+\eta]t} - 1), \tag{9}$$

where

$$\gamma(\beta) := \int_0^\infty dt\left|\int_0^\infty \frac{d\omega}{\pi}\Delta J(\omega)\coth\left(\frac{\beta\omega}{2}\right)\cos(\omega t)\right|,$$
$$\eta := \int_0^\infty dt\left|\int_0^\infty \frac{d\omega}{\pi}\Delta J(\omega)\sin(\omega t)\right|;$$

this is the central result of this Letter.

The error bound Eq. (9) manifestly satisfies all properties we expect from it: it is proportional to the norm of the spin observable being evaluated, vanishes at $t = 0$, and grows exponentially in time, which makes it scale linearly at short times, at a rate proportional to the square of the coupling in accordance with the relation $J(\omega) = \pi h^2(\omega)$. Note that the norm of the observable itself only enters the result as a prefactor: this is expected because the error is a consequence of an incomplete knowledge of the dynamics of the system, regardless of what observable is being estimated; the relative error bound is thus the same for all observables and only needs to be computed once.

Both bounds are very weak at long times because by construction they keep no account of the free dynamics of the spin. It is worth mentioning, however, that the singular-value decomposition used in our derivation does not affect the bounds in the case of pure dephasing, in which $[\hat{h}_I, \hat{H}_S] = 0$ and no interference effects due to time evolution take place inside $\hat{\Phi}[\hat{h}_I, \hat{h}_I', J]$: pure dephasing is the worst-case scenario with respect to this inequality.

Application to hierarchical equations of motion.—The HEOM method for solving open-system problems beyond standard perturbation theory was first proposed and tested around 1990 by Kubo, Tanimura, and others [11,24,25] for antisymmetrized Lorentzian spectral densities $J_L(\omega;\Omega,\Gamma) = (\pi/2)\omega/\{[(\omega+\Omega)^2+\Gamma^2][(\omega-\Omega)^2+\Gamma^2]\}$; their scheme replaces the possibly non-Markovian generalized quantum master equation for the state of some open system with a system of time-local differential equations for both the reduced density matrix and a set of so-called auxiliary density matrices, which encode information about the bath. In principle, this hierarchy of equations is infinite, but in computations it is necessary to truncate it at some order, which may be much higher than conventional perturbative approaches can usually attain [26].

The form of the bath spectral density is an important part of the scheme, because it is necessary for the bath correlation function to have the form of a sum of exponentials, as is the case with antisymmetrized Lorentzians; however, Meier and Tannor have shown [12] that many other spectral densities may be approximated very accurately by a suitable linear combination of Lorentzians, greatly extending the applicability of the method. Later studies such as Ref. [26] also explored the possibility of fitting arbitrary bath spectral densities using other functions yielding exponentially damped correlations.

We will now apply our findings to the results presented in Ref. [12] on the spin-boson application of HEOM; for the details of how the problem is formulated, the interested reader is referred to the original paper. For our purposes, it is sufficient to say that for an antisymmetrized Lorentzian spectral density, which yields a correlation function

$$\xi_L(t;\Omega,\Gamma) = \frac{e^{-\Gamma t}}{8\Omega\Gamma}\left[\coth\left(\frac{\beta}{2}(\Omega+i\Gamma)\right)e^{i\Omega t} + \coth\left(\frac{\beta}{2}(\Omega-i\Gamma)\right)e^{-i\Omega t} + 2i\sin(\Omega t)\right] - \frac{2}{\beta}\sum_{k=1}^{\infty}\frac{\nu_k e^{-\nu_k t}}{(\Omega^2+\Gamma^2-\nu_k^2)^2+4\Omega^2\nu_k^2}, \tag{10}$$

where $\nu_k := (2\pi k/\beta)$ are the Matsubara frequencies, the scheme computes dynamics and operator expectation values corresponding to a truncation of the series in Eq. (10) at order N. The accuracy of this approximation is unknown, and one usually performs numerical simulations with increasing N until the results stop changing appreciably. Convergence is thus declared heuristically, assuming that if the distance between the results obtained and those given by $N-1$ is negligible, then so is the difference between them and the true physics given by $\xi(t)$. With our result Eq. (9), the maximum distance between the predictions for some value of N and the physically correct result at $N \to \infty$ may be determined with a few lines of simple algebra instead of running an unpredictable number of costly simulations: we will now demonstrate this by giving the results of our bound Eq. (9) applied to the simulations in the paper by Meier and Tannor [12].

In their model, the spin Hamiltonian is $\hat{H}_S := (\epsilon/2)\times(\sigma_z+\sigma_x)$, the coupling is given by $\xi := (\lambda^2/4) = 0.1$, and the spectral density considered is Ohmic and defined as $J(\omega) := (\pi/2)\omega e^{-\omega/\Omega}$, with $\Omega = \frac{15}{4}\epsilon$ and fitted with three Lorentzians whose parameters are listed in Table I.

For a general linear combination of antisymmetrized Lorentzians $J(\omega) = \sum_{i=1}^n p_i J_L(\omega;\Omega_i,\Gamma_i)$, absorbing the overall coupling strength λ^2 in the coefficients p_i for the sake of simplicity, the truncation of $\xi(t)$ at order N gives

TABLE I. Parameters of the reconstructed spectral density $J(\omega) = \sum_{i=1}^{3} p_i J_L(\omega; \Omega_i, \Gamma_i)$ from Ref. [12].

$(p_i/\xi\Omega^4)$	(Ω_i/Ω)	(Γ_i/Ω)
12.0677	0.2378	2.2593
−19.9762	0.0888	5.4377
0.1834	0.0482	0.8099

$$\Delta\xi(t) = -\frac{\pi}{\beta}\sum_{i=1}^{n}\sum_{k=N+1}^{\infty}\frac{p_i\nu_k e^{-\nu_k t}}{(\Omega_i^2+\Gamma_i^2-\nu_k^2)^2+4\Omega_i^2\nu_k^2},$$

which is real and satisfies condition Eq. (8) [23], and, hence,

$$\gamma(\beta) = \int_0^\infty dt|\Delta\xi(t)| \le \frac{\pi}{\beta}\sum_{i=1}^{n}\sum_{k=N+1}^{\infty}\frac{|p_i|}{(\Omega_i^2+\Gamma_i^2-\nu_k^2)^2+4\Omega_i^2\nu_k^2} \quad (11)$$

and $\eta = 0$. The series $\sum_{k=1}^{\infty}[1/(\Omega^2+\Gamma^2-\nu_k^2)^2+4\Omega^2\nu_k^2]$ can be summed exactly, so we obtain the result as a difference:

$$\gamma_N(\beta) := \frac{\pi}{2\beta}\sum_{i=1}^{n}|p_i|\left(-\frac{1}{(\Omega_i^2+\Gamma_i^2)^2} + \frac{\beta\Omega_i\sin(\beta\Gamma_i)+\beta\Gamma_i\sinh(\beta\Omega_i)}{4\Omega_i\Gamma_i(\Omega_i^2+\Gamma_i^2)[\cosh(\beta\Omega_i)-\cos(\beta\Gamma_i)]} - \sum_{k=1}^{N}\frac{2}{(\Omega_i^2+\Gamma_i^2-\nu_k^2)^2+4\Omega_i^2\nu_k^2}\right), \quad (12)$$

using the triangle inequality on the $|p_i|$ as in Eq. (11).

In Ref. [12], the authors computed the time evolution of the expectation value $\langle\sigma_z\rangle$ at temperatures $\epsilon\beta = 0.4, 1.4$, and 10.0 for times until $\epsilon t_{\max} = 30$, at which point the system has thermalized almost completely. The number N of Matsubara frequencies needed for convergence for these three temperatures was 2, 7, and 48, respectively, due to the better performance of the HEOM method at high temperatures.

We calculated the error bound for all three cases, both with Eq. (12) and by performing the integral in Eq. (11) numerically instead of using the triangle inequality; in order to better assess the quality of our bound, we have also determined the truncation order necessary for the maximum error given by either bound to drop below 20% at each temperature. Table II shows our results.

The numerical integral gives remarkably strong bounds at the time scale of interest, given the exponential time dependence of our result Eq. (9): the maximum difference between the predicted and the actual value of $\langle\sigma_z\rangle$ at time $t_{\max}$ is guaranteed to lie between $0.09||\sigma_z|| = 0.09$ and $0.46||\sigma_z|| = 0.46$ in all three cases, and $\gamma(\beta)$ is small enough for the time scaling to be well within the linear regime at time $t_{\max}$, which is of the order of the equilibration time of the system [12]. It should also be noted that in many relevant applications (e.g., transient spectroscopy) the time scales of interest are much shorter.

Because the coefficients p_i of the components of the fitted spectral density and correlation function are both positive and negative while the analytical formula Eq. (12) only uses their absolute values, it overestimates $|\Delta\xi(t)|$ and $\gamma(\beta)$ considerably, explaining the suboptimal results given by the fully analytical bound for the case at hand.

Conclusions.—We have investigated the sensitivity of spin operator expectation values in the spin-boson model to changes in the spectral density, and derived two rigorous time-dependent error bounds under the only assumptions of factorizing initial conditions and a linearly coupled thermal bath of quantum harmonic oscillators. The results depend on the system-bath coupling strength and the spectral density variation considered, and can be expressed in a simple and elegant form in terms of these quantities. We also found the encouraging result that most of the commonly used bath models obey the strongest of the two bounds, the exceptions being baths with slowly decaying or nondecaying correlation functions.

These error bounds may be applied in many physically relevant contexts, such as comparing theoretical predictions with experimental results based on spectral densities known up to some error, determining whether a given environmental spectrum constitutes a reasonable ansatz for a physical system for which experimental or numerical data are available, or certifying the accuracy of theoretical or numerical results obtained by changing the bath correlation function in order to solve for the dynamics.

As an example application, we have demonstrated the latter use of the error bound by applying it to existing numerical results obtained with the HEOM scheme: we have shown that our results can quantitatively certify the robustness of the method, providing useful bounds on the

TABLE II. Results for the analytical and numerical bounds on the relative error at time $t_{\max}$, for the three cases considered in the original paper Ref. [12]. The last two columns indicate at what N the maximum relative error from both calculations would be under 20%.

$\epsilon\beta$	N	$[\lvert\Delta\langle\sigma_z\rangle(t_{\max})\rvert^{\mathrm{an}}/\lVert\sigma_z\rVert](N)$	$[\lvert\Delta\langle\sigma_z\rangle(t_{\max})\rvert^{\mathrm{num}}/\lVert\sigma_z\rVert](N)$	$N_{20\%}^{\mathrm{an}}$	$N_{20\%}^{\mathrm{num}}$
0.4	2	27.94%	9.43%	3	2
1.4	7	62.39%	23.77%	10	8
10.0	48	111.69%	45.34%	70	56

maximum physically possible difference between the predicted and the exact results, and that it can therefore be used to ascertain the achieved precision without testing it against more costly numerical computations.

In addition to backing up theoretical predictions with rigorous error bounds and finding practical applications in computational contexts such as HEOM simulations, this work also provides a route for the derivation of analogous bounds on many-time correlation functions or open quantum systems more complex than the spin-boson model, such as n-level systems, spin chains, or the like, as long as the environment and initial conditions satisfy the same assumptions and bounded observables are considered.

The authors thank Jaemin Lim for useful discussions about HEOM. This work was supported by an Alexander von Humboldt Professorship, the ERC Synergy grant BioQ, the CRC/TR21, the H2020- FETPROACT-2014 Grant QUCHIP (Quantum Simulation on a Photonic Chip; GA 641039), and the FP7 project PAPETS (Phonon-Assisted Processes for Energy Transfer and Sensing), GA 323901.

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