## **Response Functions as Quantifiers of Non-Markovianity**

Philipp Strasberg\* and Massimiliano Esposito

Physics and Materials Science Research Unit, University of Luxembourg, L-1511 Luxembourg, Luxembourg

(Received 15 December 2017; published 24 July 2018)

Quantum non-Markovianity is crucially related to the study of dynamical maps, which are usually derived for initially factorized system-bath states. We demonstrate that linear response theory also provides a way to derive dynamical maps but for initially correlated (and, in general, entangled) states. Importantly, these maps are always time-translational invariant and allow for a much simpler quantification of non-Markovianity compared to previous approaches. We apply our theory to the Caldeira-Leggett model, for which our quantifier is valid beyond linear response and can be expressed analytically. We find that a classical Brownian particle coupled to an Ohmic bath can already exhibit non-Markovian behavior, a phenomenon related to the initial state preparation procedure. Furthermore, for a peaked spectral density, we show that there is no monotonic relation between our quantifier and the system-bath coupling strength, the sharpness of the peak or the resonance frequency in the bath.

DOI: 10.1103/PhysRevLett.121.040601

Introduction.—A central problem of nonequilibrium statistical mechanics is to obtain a closed dynamical description for some "relevant" degrees of freedom without the need to explicitly model the remaining "irrelevant" degrees of freedom. Within the theory of open quantum systems, the complete system state  $\rho_S(t)$  is usually regarded as relevant while the bath is traced out [1,2]. Using the Nakajima-Zwanzig projection operator formalism, this can be done in a formally exact way, but unfortunately, initial system-bath correlations prevent the reduced dynamics from being closed due to the appearance of an inhomogeneous term.

We here show that within linear response theory it is possible (under certain conditions stated below) to obtain a reduced dynamical description for a set of system observables even in the presence of an initially entangled systembath state. Our findings allow us to define a rigorous, yet very simple, quantifier of non-Markovianity, which we can even express analytically for the Caldeira-Leggett model—a result which is very demanding to derive based on previous approaches [3,4].

*Linear response theory.*—We consider the standard system-bath setup and assume a global equilibrium state  $\rho_{SB}(t_0) \sim e^{-\beta(H_S+H_I+H_B)}$  (where  $H_{S/I/B}$  denotes the system/interaction/bath part of the Hamiltonian) prior to the "initial" time  $t_0$ . We then suddenly perturb the system part of the Hamiltonian such that

$$H_S(t) = H_S - \sum_i a_i \delta(t - t_0) A_i, \qquad (1)$$

where the  $A_i$  are system observables and the  $a_i \in \mathbb{R}$ (assumed to be sufficiently small) describe the respective strengths of the delta-kick  $\delta(t - t_0)$ . The purpose of the delta-kick is to generate a local unitary transformation  $U_0 = \exp[(i/\hbar) \sum_i a_i A_i] \otimes 1_B$ , which prepares the system in a nonequilibrium state at  $t_0$ . The initial state has then, to linear order, expectation values

$$\langle A_i(t_0) \rangle = \langle U_0^{\dagger} A_i U_0 \rangle_{\beta} = \langle A_i \rangle_{\beta} + \sum_j (\chi_+)_{ij} a_j. \quad (2)$$

Here,  $\langle ... \rangle_{\beta}$  denotes an expectation value with respect to the global equilibrium state. Furthermore, we introduced the skew-symmetric matrix  $(\chi_+)_{ij} = (i/\hbar) \langle [A_i, A_j] \rangle_{\beta}$ , where [A, B] denotes the commutator. We remark that the bath state does not change during this preparation procedure, and the system-bath correlations (as measured by the mutual information) also remain the same.

In the following, we consider only centered observables such that  $\langle A_i \rangle_{\beta} = 0$  without losing generality. The expectation value of  $A_i$  at a later time  $t \ge t_0$  is then connected to the response function  $\chi_{ij}(t) \equiv (i/\hbar)\Theta(t)\langle [A_i(t), A_j] \rangle_{\beta}$  via the Kubo formula [5]. In matrix notation, we have

$$\langle \mathbf{A}(t) \rangle = \chi(t - t_0) \mathbf{a} = G(t - t_0) \langle \mathbf{A}(t_0) \rangle,$$
 (3)

where we introduced the mean value propagator  $G(t) \equiv \chi(t)\chi_{+}^{-1}$ , which is the central object of interest in what follows. Note that  $\lim_{t \searrow 0} \chi(t) = \chi_{+}$ . Equation (3) amounts to our fundamental assumption in this paper as it is

Published by the American Physical Society under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

not guaranteed that the inverse of  $\chi_+$  exists (see the Supplemental Material [6], which includes Ref. [7]). If it exists, Eq. (3) describes a closed evolution equation for the mean values of the set of observables  $A_i$  for all times  $t \ge t_0$ . Two properties of G(t) are very important in the following. First, G(t) is independent of the initial state as it does not depend on any of the  $a_i$ . Second, the propagator G(t) depends only on the elapsed time, which follows from the fact that the response function is expressed in terms of time-translational invariant equilibrium correlation functions (CFs).

Therefore, if the system behaves Markovian, the mean value propagator must obey

$$G(t) = G(t-s)G(s) \quad \text{for all } s \in [0, t], \tag{4}$$

a condition which is also called divisibility. Equivalently, this implies for the response functions

$$\chi(t) = \chi(t-s)\chi_+^{-1}\chi(s).$$
(5)

Finally, for later use, we note that the response function also determines the temporal behavior of the equilibrium CFs due to the fluctuation dissipation theorem (FDT). Out of the many possible forms of the FDT, we only need [5]

$$\Im[\tilde{\chi}_{ii}(\omega)] = \frac{1}{2\hbar} (1 - e^{-\beta\hbar\omega}) \tilde{\mathcal{C}}_{ii}(\omega), \qquad (6)$$

where, in general,  $C_{ij}(t - t_0) \equiv \langle A_i(t)A_j(t_0) \rangle_{\beta}$ , which also depends only on the time difference. Furthermore, we introduced the Fourier transform  $\tilde{f}(\omega) \equiv \int_{-\infty}^{\infty} dt e^{i\omega\tau} f(\tau)$ . Note that the FDT also fixes the real part of the response function via the Kramers-Kronig relation.

Comparison with previous approaches.—Before proceeding, let us contrast our approach with the conventional one. Arguably the common considered scenario in the theory of open quantum systems starts with an initial product state  $\rho_S(t_0) \otimes \rho_B(t_0)$  [1–4] (for an exception, see [8]). This bears the advantage that the inhomogeneous term in the Nakajima-Zwanzig equation disappears, and the reduced dynamics of the system is described by a completely positive and trace preserving (CPTP) dynamical map

$$\Phi(t, t_0)\rho_S(t_0) \equiv \operatorname{tr}_B\{U\rho_S(t_0) \otimes \rho_B(t_0)U^{\dagger}\}, \quad (7)$$

where U is a unitary evolution operator acting on the joint system-bath state. Although it has been recently studied in greater generality whether it is possible to relax the initial product state assumption [9–12], the family of initially entangled states considered above will, in general, *not* give rise to a CPTP map. Therefore, there is no direct connection between our approach and previous results, although we can draw analogies.

Indeed, while  $\Phi(t, t_0)$  as G(t) is independent of the initial system state, the former does not propagate mean values but the complete system state  $\rho_S(t_0)$  to arbitrary later times  $t \ge t_0$ . It is interesting to ask whether G(t) can be extended to a dynamical map for the entire system density matrix by looking at a complete set of system observables  $\{A_i\}$ , whose expectation values are isomorphic to  $\rho_S(t)$ . In the Supplemental Material [6], we demonstrate that this is not possible because  $\chi_+$  in Eq. (3) becomes noninvertible.

Finally, to characterize non-Markovianity within the standard approach based on Eq. (7), the concept of completely positive (CP) divisibility is important. A CP divisible quantum stochastic process is characterized by a family  $\{\Phi(t_2, t_1) | t_2 \ge t_1 \ge t_0\}$  of CPTP maps, which obeys

$$\Phi(t_2, t_0) = \Phi(t_2, t_1) \Phi(t_1, t_0) \quad \text{for all } t_2 \ge t_1 \ge t_0, \qquad (8)$$

analogous to the classical Chapman-Kolmogorov equation. Consequently, if a process is CP divisible, then the evolution of the density operator is Markovian (although there seems to be less agreement about the converse statement [3,4]). Based on this concept or a related notion, various quantifiers of non-Markovianity have been recently put forward [13–18], and direct experimental evidence is also accumulating [19,20].

Unfortunately, evaluating non-Markovianity for time evolutions generated by Eq. (7) is demanding as it requires, e.g., optimization procedures, the inversion of dynamical maps, or the integration over complicated disconnected domains. In part, this problem is caused by the fact that the CPTP map  $\Phi(t, t_0)$  has a complicated time dependence: Even if the unitary U in Eq. (7) is generated by a timeindependent Hamiltonian, the dynamical map is not timetranslational invariant; i.e.,  $\Phi(t, t_0) \neq \Phi(t - t_0)$ , in general. This is in strong contrast to our result in the linear response regime, where G(t) always depends only on the elapsed time and which allows us to check the simpler condition (4) instead of Eq. (8).

Distance quantifier.—To introduce new quantifiers of non-Markovianity within our approach, we need to quantify the distance between two functions f(t) and g(t). We use the standard  $L_2$  scalar product  $\langle f, g \rangle = \int_{-\infty}^{\infty} dt f(t) g^*(t)$  and the induced norm  $||f|| = \sqrt{\langle f, f \rangle}$ , where it is tacitly assumed that the integrals are converging. We then define the distance

$$\mathcal{D}(f,g) \equiv \sqrt{1 - \frac{|\langle f, g \rangle|^2}{\|f\|^2 \|g\|^2}}.$$
(9)

By the Cauchy-Schwarz' inequality,  $0 \leq \mathcal{D}(f, g) \leq 1$  and  $\mathcal{D}(\lambda f, \lambda g) = \mathcal{D}(f, g)$  for any  $\lambda \in \mathbb{C}$ ; i.e., the difference has the favorable properties that it is positive, bounded, and independent of any global scaling. By analogy with the Euclidean scalar product,  $\mathcal{D}(f, g) = |\sin(\phi)|$  can be seen as quantifying the "angle"  $\phi$  between the two vectors f(t) and g(t). Most importantly for our applications, by Parseval's

theorem, we can deduce that  $\mathcal{D}(f,g) = \mathcal{D}(\tilde{f},\tilde{g})$ , where the right hand side is computed by using the  $L_2$  scalar product in Fourier space,  $\langle \tilde{f}, \tilde{g} \rangle = \int_{-\infty}^{\infty} [(d\omega)/(2\pi)]\tilde{f}(\omega)\tilde{g}^*(\omega)$ .

*New quantifiers of non-Markovianity.*—It is advantageous to work in Fourier space in the following. Integrating Eq. (5) over *s* from zero to *t* implies in Fourier space [6]

$$-i\frac{d}{d\omega}\tilde{\chi}(\omega) = \tilde{\chi}(\omega)\chi_{+}^{-1}\tilde{\chi}(\omega).$$
(10)

Then, to measure violations of Eq. (10) as a consequence of the (assumed) divisibility property, we propose the quantifier [denoting  $\tilde{\chi}'(\omega) = d\tilde{\chi}(\omega)/d\omega$ ]

$$\mathcal{N}_{ij}^{(1)} \equiv \mathcal{D}[-i\tilde{\chi}_{ij}', (\tilde{\chi}\chi_{+}^{-1}\tilde{\chi})_{ij}].$$
(11)

As a second quantifier of non-Markovianity, we also check the validity of the regression theorem (RT) [21,22], which allows us to relate the evolution of CFs to the evolution of mean values. Within our setting, the Markovian assumption enters here by using that Eq. (3) holds for all initial states and that there exists a dynamical map  $\Phi(t, t_0)$ , which is *independent* of  $\rho_S(t_0)$ . It is worth emphasizing that the validity of the RT does not *a priori* rely on an initial product state assumption or on the property of CP divisibility. It merely signifies that it is possible to find for any initial system state a map G(t) to propagate the mean values *and*—in addition to what is required to evaluate  $\mathcal{N}_{ij}^{(1)}$ —a map  $\Phi(t, t_0)$  to propagate  $\rho_S(t_0)$  (see the Supplemental Material [6] for more details). Thus, if the RT holds,

$$C^{\text{RT}}(t, t_0) = G(t - t_0)C(t_0, t_0).$$
(12)

Here, we have added the superscript "RT" to emphasize that this is the predicted CF assuming the validity of the RT. Note that  $C_{ij}^{\text{RT}}(t, t_0) \equiv \langle A_i(t)A_j(t_0) \rangle$  denotes, in general, an out-of-equilibrium CF, but we are only interested in equilibrium CFs which we denote with a calligraphic C. For them, we can deduce in Fourier space that [6]

$$\tilde{\mathcal{C}}^{\mathrm{RT}}(\omega) = \tilde{\chi}(\omega)\chi_{+}^{-1}\mathcal{C}(0) - \mathcal{C}(0)^{T}\chi_{+}^{-1}\tilde{\chi}(\omega)^{\dagger}.$$
 (13)

We add that the behavior of CFs (often in relation with the validity of the RT) has played an important role historically to define a quantum Markov process [21–25] and was also investigated in the recent debate about non-Markovianity [26–28]. However, its use in the linear response regime has not been noted before, although it is well known that all quantum systems violate the RT in that regime [29].

Then, based on the comparison of the exact equilibrium CFs [obtained from the FDT (6)] and their Markovian prediction [obtained from the RT (13)], we propose

$$\mathcal{N}_{ij}^{(2)} \equiv \mathcal{D}[\tilde{\mathcal{C}}_{ij}, \tilde{\mathcal{C}}_{ij}^{\mathrm{RT}}].$$
(14)

We here assume that the equilibrium covariance matrix C(0) is exactly known such that the prediction (13) uses the correct initial value.

To conclude, the magnitude of both  $\mathcal{N}_{ij}^{(1)}$  and  $\mathcal{N}_{ij}^{(2)}$  measures by how much we fail by naively assuming that the process is Markovian. They can be computed *without* the need to *a priori* derive any quantum master equation—only the knowledge of the linear response functions *or* the equilibrium CFs is required.

We now treat an important class of open system models with Gaussian dynamics *exactly*, i.e., without any approximation about the temperature, the coupling strength, or the spectral features of the bath. We also remark that for this class our results are valid beyond linear response. Related studies about non-Markovianity of Gaussian dynamics based on different approaches and various approximations can be found in Refs. [16,20,30–33].

*Quantum Brownian motion.*—We consider the standard Caldeira-Leggett model with Hamiltonian (in suitable mass-weighted coordinates)

$$H = \frac{p^2 + \omega_0^2 q^2}{2} + \frac{1}{2} \sum_{k} \left[ p_k^2 + \omega_k^2 \left( q_k - \frac{c_k}{\omega_k^2} q \right)^2 \right].$$
 (15)

Here, *q* and *p* refer to the position and momentum of the system with frequency  $\omega_0 > 0$ , whereas the bath oscillators with frequencies  $\omega_k > 0$  are specified with an additional index *k*. Furthermore,  $c_k$  denotes the coupling strength between the system and the *k*th oscillator. Of central importance is the spectral density (SD)  $J(\omega) \equiv (\pi/2) \sum_k (c_k^2/\omega_k) \delta(\omega - \omega_k)$ . It characterizes the coupling between system and bath, and it is assumed to be a continuous function of  $\omega$  in the limit of a large bath fulfilling  $J(0) = 0 = J(\omega \to \infty)$ . A great benefit of the Brownian motion model is that almost all quantities of interest are computable in closed form [34,35]; e.g., the matrix of response functions reads (see the Supplemental Material [6] for a derivation)

$$\begin{pmatrix} \tilde{\chi}_{qq}(\omega) & \tilde{\chi}_{qp}(\omega) \\ \tilde{\chi}_{pq}(\omega) & \tilde{\chi}_{pp}(\omega) \end{pmatrix} = \begin{pmatrix} 1 & i\omega \\ -i\omega & \omega^2 \end{pmatrix} \tilde{\chi}_{qq}(\omega) + \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix},$$
$$\tilde{\chi}_{qq}(\omega) = \frac{1}{\omega_0^2 - \omega^2 - i\omega\tilde{\gamma}(\omega)},$$
(16)

where  $\tilde{\gamma}(\omega)$  is the Fourier transform of the memory kernel  $\gamma(t) = \Theta(t)(2/\pi) \int_0^\infty d\omega \{[J(\omega)]/\omega\} \cos(\omega t)$ . In view of the general theory outlined above, our set of system observables will be the position and momentum of the system  $\{A_1, A_2\} = \{q, p\}$ , and one easily verifies that  $\chi_+$  is symplectic with  $(\chi_+)_{pq} = 1 = -(\chi_+)_{qp}$ . The delta-kick now creates the unitary  $U_0 = e^{(i/\hbar)(a_q q + a_p p)}$ . This shifts

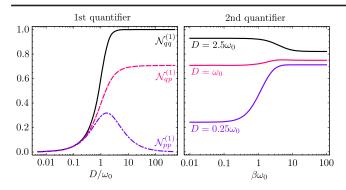


FIG. 1. Left: Plot of  $\mathcal{N}_{qq}^{(1)}$  (solid black),  $\mathcal{N}_{qp}^{(1)}$  (dashed pink), and  $\mathcal{N}_{pp}^{(1)}$  (dashed-dotted purple) for an Ohmic SD over the dimensionless coupling strength  $D/\omega_0$  in logarithmic scale. Note that  $\mathcal{N}_{qp}^{(1)} = \mathcal{N}_{pq}^{(1)}$ . Right: Plot of  $\mathcal{N}_{qq}^{(2)}$  over the dimensionless inverse temperature  $\beta\omega_0$  for various coupling strengths D in logarithmic scale. It demonstrates that at strong coupling higher temperatures can yield stronger non-Markovianity. We set  $\hbar \equiv 1$ .

the position and momentum operators  $U_0^{\dagger}qU_0 = q - a_p$ and  $U_0^{\dagger}pU_0 = p + a_q$ , thereby shifting the mean values but leaving the covariances unchanged. Furthermore, the equilibrium covariance matrix is diagonal with entries  $C_{qq}(0) = (\hbar/\pi) \int_0^{\infty} d\omega \coth(\beta \hbar \omega/2) \Im[\tilde{\chi}_{qq}(\omega)]$  and  $C_{pp}(0) =$  $(\hbar/\pi) \int_0^{\infty} d\omega \omega^2 \coth(\beta \hbar \omega/2) \Im[\tilde{\chi}_{qq}(\omega)]$ , and the equilibrium CFs are linked via

$$\begin{pmatrix} \tilde{\mathcal{C}}_{qq}(\omega) & \tilde{\mathcal{C}}_{qp}(\omega) \\ \tilde{\mathcal{C}}_{pq}(\omega) & \tilde{\mathcal{C}}_{pp}(\omega) \end{pmatrix} = \begin{pmatrix} 1 & i\omega \\ -i\omega & \omega^2 \end{pmatrix} \tilde{\mathcal{C}}_{qq}(\omega).$$
(17)

We now have all quantities at hand to compute our quantifiers. For the rest of the paper, we set  $t_0 = 0$ .

Classical Ohmic limit.—We consider the simplest case of a classical particle ( $\hbar = 0$ ) coupled to an Ohmic bath, which corresponds to a memory kernel of the form  $\gamma(t) = D\delta(t)$ . This follows from a linear SD  $J(\omega) =$  $D\omega$  in the limit of an infinitely high cutoff frequency. The resulting Langevin equation for the system reads (see Supplemental Material [6] for a detailed derivation)

$$\dot{q}(t) = p(t) - a_p \delta(t), \dot{p}(t) = -\omega_0^2 q(t) + a_q \delta(t) - D\dot{q}(t) + \xi(t).$$
(18)

Here, the noise obeys  $\langle\!\langle \xi(t) \rangle\!\rangle = 0$  and  $\langle\!\langle \xi(t)\xi(s) \rangle\!\rangle = \gamma(t-s)/\beta$  with the crucial requirement that  $\langle\!\langle ... \rangle\!\rangle$  refers to an average over an initial *conditional* equilibrium state of the bath [35–38]

$$\rho_B(0) \sim \exp\left\{-\frac{\beta}{2} \sum_k \left[p_k^2 + \omega_k^2 \left(q_k - \frac{c_k}{\omega_k^2} q(0^-)\right)^2\right]\right\}.$$
(19)

Here, the position  $q(0^-)$  of the Brownian particle *prior* to the delta-kick is a random variable distributed according to a Gaussian  $P[q(0^-)] \sim e^{-\beta \omega_0^2 q(0^-)^2/2}$  such that, shortly before the unitary kick, the global system-bath state is in equilibrium.

If we would not disturb the state,  $a_q = 0$  and  $a_p = 0$  and Eq. (18) reduces to the standard Langevin equation. However, the presence of the unitary kick results in an initial system state described by a shifted Gaussian  $P[q(0)] \sim e^{-\beta\omega_0^2[q(0)-a_q]^2/2}$  while the bath still resides in the state (19). The fact that the bath has no time to adapt to a new conditional equilibrium state causes non-Markovian

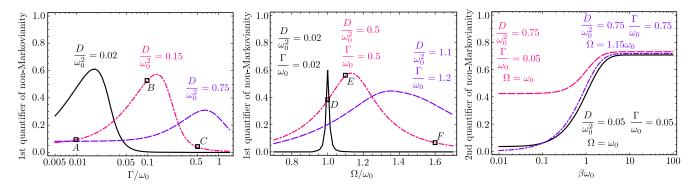


FIG. 2. We use the convention of Fig. 1 where a solid line refers to  $\mathcal{N}_{qq}^{(1/2)}$ , a dashed line to  $\mathcal{N}_{qp}^{(1/2)}$ , and a dashed-dotted line to  $\mathcal{N}_{pp}^{(1/2)}$ , but the color coding is different. Left: Plot (in logarithmic scale) over the dimensionless parameter  $\Gamma/\omega_0$  at resonance ( $\Omega = \omega_0$ ) for increasing coupling strengths  $D = 0.02\omega_0^2$  (black solid line),  $D = 0.15\omega_0^2$  (pink dashed-dotted line), and  $D = 0.75\omega_0^2$  (purple dashed line). Middle: Plot over the dimensionless resonance frequency  $\Omega/\omega_0$  of the bath for increasing  $(D, \Gamma)$ :  $D = 0.05\omega_0^2$ ,  $\Gamma = 0.05\omega_0$  (black solid line),  $D = 0.15\omega_0^2$ ,  $\Gamma = 1.2\omega_0$  (purple dashed line). Right: Plot over the dimensionless inverse temperature  $\beta\omega_0$  in logarithmic scale for  $D = 0.05\omega_0^2$ ,  $\Gamma = 0.05\omega_0$ ,  $\Omega = \omega_0$  (black solid line),  $D = 0.75\omega_0^2$ ,  $\Gamma = 0.05\omega_0$ ,  $\Omega = 0.05\omega_0^2$ ,  $\Gamma = 0.05\omega_0$ ,  $\Omega = \omega_0$  (pink dashed line) and  $D = 0.75\omega_0^2$ ,  $\Gamma = 0.75\omega_0$ ,  $\Omega = 1.15\omega_0$  (purple dashed-dotted line). We set  $\hbar \equiv 1$ . Furthermore, the black squares indicates the parameters for which we provide additional plots of the time evolution of  $\langle p(t) \rangle$  in the Supplemental Material [6].

behavior as we can rigorously show with our quantifier. For instance, in view of Eq. (10), we find that

$$-i\tilde{\chi}' - \tilde{\chi}\chi_{+}^{-1}\tilde{\chi} = \frac{D}{(\omega_{0}^{2} - \omega^{2} - iD\omega)^{2}} \begin{pmatrix} 1 & i\omega \\ -i\omega & \omega^{2} \end{pmatrix}, \quad (20)$$

which is clearly nonzero and only becomes negligible in the weak coupling regime, see Fig. 1. The subtle importance of the initial state preparation procedure for the validity of the Langevin equation was already noted in Refs. [36–38], but it had not been rigorously quantified.

We remark that it is a special property of the Caldeira-Leggett model that the first moments do not depend on  $\hbar$ . This changes for CFs, which depend on  $\hbar$  and the inverse bath temperature, see Fig. 1 again.

Peaked SD.—We now turn to the case described by the SD,

$$J(\omega) = \frac{D^2 \Gamma \omega}{(\omega^2 - \Omega^2)^2 + \Gamma^2 \omega^2}.$$
 (21)

This corresponds to the SD felt by a system, which is coupled with strength *D* to another harmonic oscillator of frequency  $\Omega$ , which is in turn coupled to an Ohmic bath with SD  $\Gamma\omega$  [39]. Note that the parameter  $\Gamma$  controls the structure of the SD: a small  $\Gamma$  corresponds to a sharp peak around the frequency  $\Omega$ , whereas a larger  $\Gamma$  smears out the peak resulting in an increasingly flat SD. Furthermore, the real part of the Fourier transformed memory kernel is  $\Re[\tilde{\gamma}(\omega)] = J(\omega)/\omega$ , and the imaginary part becomes  $\Im[\tilde{\gamma}(\omega)] = (\Gamma^2 + \omega^2 - \Omega^2)J(\omega)/\Gamma\Omega^2$  [6].

In practical considerations, non-Markovian behavior is often associated with a strong system-bath coupling and a structured SD [2]. Thus, one would intuitively expect that the degree of non-Markovianity increases for larger D and smaller  $\Gamma$  and that it reaches a maximum if the system is on resonance with the oscillator in the bath; i.e., if  $\omega_0 \approx \Omega$ . As Fig. 2 demonstrates, this intuition is not always correct. We observe that there is no simple (i.e., monotonic) relation between our quantifier of non-Markovianity and the parameters D,  $\Gamma$ , and  $|\omega_0 - \Omega|$ . In fact, one could ask whether this results from the particular definition (9) and (11) which we have used and which always entails a certain level of arbitrariness. Therefore, we have also plotted the time evolution of the observable  $\langle p(t) \rangle$  (see [6]), whose deviation from an exponentially damped oscillation seems to be roughly in agreement with our quantification scheme.

*Summary.*—This work shows that it is possible to quantify non-Markovianity in the linear response regime in a rigorous and straightforward manner. Since we can only treat initially correlated states, our approach is rather "orthogonal" to previous ones, but for many scenarios of experimental interest, this might be indeed a more realistic assumption. Furthermore, for the Caldeira-Leggett model, our quantifier is valid beyond linear response and can be expressed analytically in terms of an integral over known functions. We have then shown that even a classical particle coupled to an Ohmic bath can behave non-Markovian depending on the initial state preparation procedure and that one should not expect a simple relation between our quantifier of non-Markovianity and parameters in the SD or the temperature of the bath.

We are grateful to Victor Bastidas, John Bechhoefer, and Javier Cerrillo for discussions and comments. This research is funded by the European Research Council project NanoThermo (ERC-2015-CoG Agreement No. 681456).

<sup>\*</sup>philipp.strasberg@uni.lu

- [1] H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, Oxford, 2002).
- [2] I. de Vega and D. Alonso, Dynamics of non-Markovian open quantum systems, Rev. Mod. Phys. 89, 015001 (2017).
- [3] A. Rivas, S. F. Huelga, and M. B. Plenio, Quantum non-Markovianity: Characterization, quantification and detection, Rep. Prog. Phys. 77, 094001 (2014).
- [4] H.-P. Breuer, E.-M. Laine, J. Piilo, and B. Vacchini, Colloquium: Non-Markovian dynamics in open quantum systems, Rev. Mod. Phys. 88, 021002 (2016).
- [5] N. Pottier, Nonequilibrium Statistical Physics Linear Irreversible Processes (Oxford University Press, New York, 2010).
- [6] See Supplementary Material at http://link.aps.org/ supplemental/10.1103/PhysRevLett.121.040601 for additional discussions, detailed derivations and one additional figure supplementing Fig. 2.
- [7] R. Wu, A. Pechen, C. Brif, and H. Rabitz, Controllability of open quantum systems with Kraus-map dynamics, J. Phys. A 40, 5681 (2007).
- [8] F. A. Pollock, C. Rodríguez-Rosario, T. Frauenheim, M. Paternostro, and K. Modi, Operational Markov Condition for Quantum Processes, Phys. Rev. Lett. **120**, 040405 (2018).
- [9] C. A. Rodríguez-Rosario, K. Modi, A. Kuah, A. Shaji, and E. C. G. Sudarshan, Completely positive maps and classical correlations, J. Phys. A 41, 205301 (2008).
- [10] A. Brodutch, A. Datta, K. Modi, A. Rivas, and C. A. Rodríguez-Rosario, Vanishing quantum discord is not necessary for completely positive maps, Phys. Rev. A 87, 042301 (2013).
- [11] F. Buscemi, Complete Positivity, Markovianity, and the Quantum Data-Processing Inequality, in the Presence of Initial System-Environment Correlations, Phys. Rev. Lett. 113, 140502 (2014).
- [12] J. M. Dominy, A. Shabani, and D. A. Lidar, A general framework for complete positivity, Quant, Inf. Proc. 15, 465 (2016).
- [13] M. M. Wolf, J. Eisert, T. S. Cubitt, and J. I. Cirac, Assessing Non-Markovian Quantum Dynamics, Phys. Rev. Lett. 101, 150402 (2008).
- [14] H. P. Breuer, E.-M. Laine, and J. Piilo, Measure for the Degree of Non-Markovian Behavior of Quantum Processes in Open Systems, Phys. Rev. Lett. **103**, 210401 (2009).

- [15] A. Rivas, S. F. Huelga, and M. B. Plenio, Entanglement and Non-Markovianity of Quantum Evolutions, Phys. Rev. Lett. 105, 050403 (2010).
- [16] S. Lorenzo, F. Plastina, and M. Paternostro, Geometrical characterization of non-Markovianity, Phys. Rev. A 88, 020102 (2013).
- [17] M. J. W. Hall, J. D. Cresser, L. Li, and E. Andersson, Canonical form of master equations and characterization of non-Markovianity, Phys. Rev. A 89, 042120 (2014).
- [18] D. Chruściński and S. Maniscalco, Degree of Non-Markovianity of Quantum Evolution, Phys. Rev. Lett. 112, 120404 (2014).
- [19] B.-H. Liu, L. Li, Y.-F. Huang, C.-F. Li, G.-C. Guo, E.-M. Laine, H.-P. Breuer, and J. Piilo, Experimental control of the transition from Markovian to non-Markovian dynamics of open quantum systems, Nat. Phys. 7, 931 (2011).
- [20] S. Gröblacher, A. Trubarov, N. Prigge, G. D. Cole, M. Aspelmeyer, and J. Eisert, Observation of non-Markovian micromechanical Brownian motion, Nat. Commun. 6, 7606 (2015).
- [21] M. Lax, Quantum noise. IV. Quantum theory of noise sources, Phys. Rev. 145, 110 (1966).
- [22] M. Lax, Quantum noise. XI. Multitime correspondence between quantum and classical stochastic processes, Phys. Rev. 172, 350 (1968).
- [23] F. Haake, Statistical Treatment of Open Systems by Generalized Master Equations, Springer Tracts in Modern Physics Vol. 66 (Springer, New York, 1973).
- [24] G. Lindblad, Non-Markovian quantum stochastic processes and their entropy, Commun. Math. Phys. 65, 281 (1979).
- [25] P. Talkner, Gauss Markov process of a quantum oscillator, Z. Phys. B Condens. Matter 41, 365 (1981).
- [26] G. Guarnieri, A. Smirne, and B. Vacchini, Quantum regression theorem and non-Markovianity of quantum dynamics, Phys. Rev. A 90, 022110 (2014).
- [27] N. L. Gullo, I. Sinayskiy, T. Busch, and F. Petruccione, Non-Markovian criteria for open system dynamics, arXiv:1401.1126.

- [28] Md. M. Ali, P.-Y. Lo, M. Wei-Yuan Tu, and W.-M. Zhang, Non-Markovianity measure using two-time correlation functions, Phys. Rev. A 92, 062306 (2015).
- [29] G. W. Ford and R. F. O'Connell, There Is No Quantum Regression Theorem, Phys. Rev. Lett. 77, 798 (1996).
- [30] L. A. M. Souza, H. S. Dhar, M. N. Bera, P. Liuzzo-Scorpo, and G. Adesso, Gaussian interferometric power as a measure of continuous-variable non-Markovianity, Phys. Rev. A 92, 052122 (2015).
- [31] G. Torre, W. Roga, and F. Illuminati, Non-Markovianity of Gaussian Channels, Phys. Rev. Lett. 115, 070401 (2015).
- [32] J. Jin and C. s. Yu, Non-Markovianity in a collision model with environmental block, New J. Phys. 20, 053026 (2018).
- [33] G. Torre and F. Illuminati, Exact non-Markovian dynamics of Gaussian quantum channels: Finite-time and asymptotic regimes, arXiv:1804.03095 [Quantum Physics (to be published)].
- [34] H. Grabert, P. Schramm, and G.-L. Ingold, Quantum Brownian motion: The functional integral approach, Phys. Rep. 168, 115 (1988).
- [35] U. Weiss, *Quantum Dissipative Systems*, 3 ed. (World Scientific, Singapore, 2008).
- [36] W. Bez, Microscopic preparation and macroscopic motion of a Brownian particle, Z. Phys. B Condens. Matter 39, 319 (1980).
- [37] J. Sánchez Canizares and F. Sols, Translational symmetry and microscopic preparation in oscillator models of quantum dissipation, Physica A 212, 181 (1994).
- [38] P. Hänggi, Generalized Langevin Equations: A Useful Tool for the Perplexed Modeller of Nonequilibrium Fluctuations?, in *Stochastic Dynamics*, edited by L. S. Geier and T. Poschel, Lecture Notes in Physics Vol. 485 (Springer, Berlin, 1997), pp. 15–22.
- [39] A. Garg, J. N. Onuchic, and V. Ambegaokar, Effect of friction on electron transfer in biomolecules, J. Chem. Phys. 83, 4491 (1985).