Nonequilibrium fluctuation-dissipation inequality and nonequilibrium uncertainty principle

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Non-Equilibrium Fluctuation-Dissipation Inequality and Non-Equilibrium Uncertainty Principle

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The *fluctuation-dissipation relation* is usually formulated for a system interacting with a heat bath at finite temperature, and often in the context of linear response theory, where only small deviations from the mean are considered. We show that for an open quantum system interacting with a non-equilibrium environment, where temperature is no longer a valid notion, a *fluctuation-dissipation inequality* exists. Instead of being proportional, quantum fluctuations are bounded below by quantum dissipation, whereas classically the fluctuations vanish at zero temperature. The lower bound of this inequality is exactly satisfied by (zero-temperature) quantum noise and is in accord with the Heisenberg uncertainty principle, in both its microscopic origins and its influence upon systems. Moreover, it is shown that there is a coupling-dependent non-equilibrium fluctuation-dissipation relation that determines the non-equilibrium uncertainty relation of linear systems in the weak-damping limit.

I. INTRODUCTION

The *fluctuation-dissipation relation* (FDR) is a fundamental relation in non-equilibrium statistical mechanics, dating all the way back to Einstein's Nobel work on Brownian motion [1] and Nyquist's seminal work on electronic conductivity [2]. We distinguish the fluctuation-dissipation relation (FDR) from the more-common fluctuation-dissipation theorem (FDT), because, as we will demonstrate, non-equilibrium environments also permit a fluctuation-dissipation relation. In the non-equilibrium case, there is no universal relation, but we will show that there is a universal inequality that all such relations satisfy.

To give our inequality a physical interpretation, we will couch it within the tractable context of a general quantum system under the influence of quantum noise, from a linear environment or where the influence of the environment is weak. However, let us first briefly introduce the known results with the simple example of a one-dimensional Langevin equation. The equation of motion for a classical damped particle with time-local response coupled to a thermal bath with temperature T is given by

$$m\,\ddot{x}(t) + \underbrace{2\,\gamma_0\,\dot{x}(t)}_{\text{damping}} - F(x) = \underbrace{\xi(t)}_{\text{noise}}\,. \tag{1}$$

Deriving this equation from a microscopic model results in a relation between the damping and noise correlation

$$\nu(t,\tau) \equiv \langle \xi(t)\,\xi(\tau)\rangle_{\xi} = \gamma_0\,2k_B T\,\delta(t-\tau)\,, \tag{2}$$

or more simply in the frequency domain

$$\tilde{\nu}(\omega) = 2k_B T\,\gamma_0\,, \tag{3}$$

where $\nu(t,\tau) = \nu(t-\tau)$ and $\tilde{\nu}(\omega) = \int dt\, e^{-\imath\omega t}\,\nu(t)$. In short, the noise is proportional to both the temperature and the amount of damping or resistance. The same can also be said of the diffusion, which is the Einstein–Smoluchowski relation. This model can be easily generalized to a response that is not instantaneous, but where there is nonlocal damping:

$$m\,\ddot{x}(t) + 2\int_{-\infty}^{t} d\tau\,\gamma(t-\tau)\,\dot{x}(\tau) - F(x) = \xi(t)\,, \tag{4}$$

in which case the classical fluctuation-dissipation relation generalizes to

$$\tilde{\nu}(\omega) = 2k_B T\,\tilde{\gamma}(\omega)\,. \tag{5}$$

If the response has some lag, then the thermal noise is colored, and vice versa, whereas above for instantaneous response the noise was white. Finally, when we apply this same formalism to the phase-space characteristics of a quantum oscillator with the same type of Langevin equation [3], the quantum FDR can be seen to generalize to

$$\tilde{\nu}(\omega) = \hbar\omega\coth\!\left(\frac{\hbar\omega}{2k_B T}\right)\tilde{\gamma}(\omega)\,. \tag{6}$$

The qualitative difference between the classical and quantum FDR is that at zero temperature there are ground-state fluctuations proportional to $\hbar$. Only at sufficiently high temperature does the quantum FDR asymptote to the classical relation.

What we show in this work is that for any non-equilibrium environment, one has the fluctuation-dissipation inequality (FDI)

$$\tilde{\nu}(\omega) \geq \left|\hbar\omega\,\tilde{\gamma}(\omega)\right|\,, \tag{7}$$

or, in other words, for any amount of damping, the corresponding noise must be greater than the corresponding hypothetical ground-state fluctuations. Moreover, the FDI is fundamentally related to the Heisenberg uncertainty principle (HUP). As we will show, if one could have an environment that violated the FDI, then it would relax the system to a state that violates the HUP. Furthermore, if one allows the state of the environment to violate the HUP, then it may also violate the FDI.

Like the HUP, the FDI is mathematical in nature and is a precise inequality which must be obeyed by all quantum environments, in all regimes. Whereas, in its most general form the HUP relates two quantum operators of arbitrary systems, the FDI relates the two-time noise and dissipation (or equivalently, susceptibility) kernels of arbitrary environments. Though it is exact, we will mostly discuss its context and physical implications in one of two regimes,

both with stationary correlations: (1) the regime of weak coupling to a general environment and (2) the regime of nonperturbative coupling to a linear environment. Moreover, for linear systems we additionally show that the non-equilibrium FDR (which must satisfy the FDI) determines the non-equilibrium uncertainty relation (which must satisfy the HUP) for weak coupling.

In the following section we present the necessary background material of quantum open systems, wherein we formally categorize quantum noise in terms of its time dependence, dissipation and microscopic origin. Readers familiar with this may skip to our results in the later sections and refer back as needed. In Sec. III we derive the FDI from a microscopic model and contrast it to the usual thermal FDR. On the other hand, in Sec. IV we work from the other end and motivate the FDI phenomenologically, but less generally. This result also produces the (weak-coupling) non-equilibrium uncertainty relation for quantum Brownian motion, which can be contrasted to the finite-temperature uncertainty relation [4–7].

II. NOISE AND BACKREACTION

A. Open Systems and Noise

Consider the closed system + environment Hamiltonian

$$\mathbf{H}_{\mathrm{C}} = \underbrace{\mathbf{H}}_{\text{system}} + \overbrace{\mathbf{H}_{\mathrm{E}}}^{\text{environment}} + \underbrace{\mathbf{H}_{\mathrm{I}}}_{\text{interaction}} + \overbrace{\mathbf{H}_{\mathrm{R}}}^{\text{renormalization}} \,, \tag{8}$$

with the interaction Hamiltonian expanded as a sum of tensor products (in the Schrödinger picture):

$$\mathbf{H}_{\mathrm{I}}(t) = \sum_n \mathbf{L}_n(t) \otimes \mathbf{l}_n(t) \,, \tag{9}$$

where $\mathbf{L}_n(t)$ and $\mathbf{l}_n(t)$ are system and environment operators respectively, possibly with some intrinsic time dependence. The environment coupling operators $\mathbf{l}_n$ will typically be collective observables of the environment, with dependence upon very many modes. For linear environments or weak coupling to general environments, the central ingredient of any open-systems analysis is the second-order (multivariate) correlation function of the environment (E):

$$\alpha_{nm}(t,\tau) = \langle \underline{\mathbf{l}}_n(t)\, \underline{\mathbf{l}}_m(\tau) \rangle_{\mathrm{E}} \,, \tag{10}$$

where $\underline{\mathbf{l}}_n(t)$ represents the time-evolving $\mathbf{l}_n(t)$ in the interaction (Dirac) picture. The correlation function can always be written in terms of two real kernels, respectively symmetric and antisymmetric and traditionally referred to as the *noise kernel* and *dissipation kernel*:

$$\underbrace{\boldsymbol{\alpha}(t,\tau)}_{\text{complex noise}} = \underbrace{\boldsymbol{\nu}(t,\tau)}_{\text{noise}} + i\hbar\, \underbrace{\boldsymbol{\mu}(t,\tau)}_{\text{dissipation}} \,. \tag{11}$$

Note that despite this choice of name the dissipation kernel can also give rise to non-dissipative effective forces.

The correlation function $\boldsymbol{\alpha}(t,\tau)$ exists for any quantum system and environment, and its corresponding noise and dissipation kernels will always obey our inequality. However, the physical interpretation of these kernels is less obvious in the case of strong coupling to non-linear environments with significant higher-order cumulants for their processes, such as fermionic environments. For example, for a thermal environment consisting of two-level systems the noise kernel is temperature independent, whereas the dissipation kernel has explicit temperature dependence, as shown in Appendix A. This non-intuitive behavior is opposite to that of typical bosonic environments, such as photons and phonons. Leaving this consideration aside, let us briefly discuss next how the correlation function arises in the most common formalisms for open-system analysis.

In the *influence functional* formalism [8] for the quantum Brownian model with continuous system couplings $L_n(\mathbf{x};t)$ (in the Lagrangian) and a linear environment [9–11] the correlation function appears as the kernel in the exponent of a Gaussian influence functional, called the influence kernel ζ in Refs. [12, 13]. The reduced density matrix $\boldsymbol{\rho}$ for the open system evolves according to the double path integral

$$\langle \mathbf{x}|\, \boldsymbol{\rho}(t)\, |\mathbf{y}\rangle = \tag{12}$$
$$\left\langle \int_{\mathbf{x}_0}^{\mathbf{x}} \mathcal{D}\mathbf{x}(t) \int_{\mathbf{y}_0}^{\mathbf{y}} \mathcal{D}\mathbf{y}(t)\, e^{+\frac{i}{\hbar}S[\mathbf{x}(t)] - \frac{i}{\hbar}S[\mathbf{y}(t)] - \Phi[\mathbf{x}(t),\mathbf{y}(t)]} \right\rangle_{\mathbf{x}_0,\mathbf{y}_0} \,,$$

with the initial-value average given by

$$\langle \cdots \rangle_{\mathbf{x}_0,\mathbf{y}_0} \equiv \iint d\mathbf{x}_0\, d\mathbf{y}_0\ \langle \mathbf{x}_0|\, \boldsymbol{\rho}(0)\, |\mathbf{y}_0\rangle\ \cdots\,, \tag{13}$$

for factorized initial states $\boldsymbol{\rho}(0)\otimes\boldsymbol{\rho}_{\rm E}(0)$. The Gaussian *influence phase* is given by

$$\begin{aligned}\Phi[\mathbf{x}(t),\mathbf{y}(t)] &= \frac{1}{2\hbar^2}\sum_{n,m}\int_0^t d\tau\int_0^t d\tau'\,\Delta_n(\tau)\,\nu_{nm}(\tau,\tau')\,\Delta_m(\tau')\\ &+\frac{\imath}{\hbar}\sum_{n,m}\int_0^t d\tau\int_0^\tau d\tau'\,\Delta_n(\tau)\,\mu_{nm}(\tau,\tau')\,\Sigma_m(\tau')\,,\end{aligned} \tag{14}$$

in terms of the difference and sum coordinates

$$\Delta_n(t) = L_n(\mathbf{x};t) - L_n(\mathbf{y};t)\,, \tag{15}$$

$$\Sigma_n(t) = L_n(\mathbf{x};t) + L_n(\mathbf{y};t)\,. \tag{16}$$

The Gaussian influence is exact for a linear environment and perturbative for general environments. The conventional influence functional here only requires the system to be coupled with continuous variables, though the formalism can be extended to fermionic systems with Grassman numbers. The noise kernel $\boldsymbol{\nu}$ appears in the influence kernel as the correlation of an ordinary real stochastic source, whereas the dissipation kernel $\boldsymbol{\mu}$ alone would produce a purely homogeneous (though not positivity preserving in general) evolution.

These same roles can also be inferred from the Heisenberg equations of motion for the system operators after integrating the environment dynamics, producing the so-called *quantum Langevin equation* [14]. In Appendix B we generalize the quantum Langevin equation to nonlinear systems, although restricted to Gaussian influence functionals and, hence, perturbative for nonlinear environments. The Heisenberg equations of motion for an open system operator $\mathbf{S}(t)$, where the environment has been integrated out, can be expressed as

$$\hbar\,\dot{\mathbf{S}}(t) = +\imath[\mathbf{H},\mathbf{S}(t)] + \imath\sum_n \{\mathbf{l}_n(t),[\mathbf{L}_n(t),\mathbf{S}(t)]\}\,, \tag{17}$$

$$\mathbf{l}_n(t) \equiv \boldsymbol{\xi}_n(t) - 2\sum_m\int_0^t d\tau\,\mu_{nm}(t,\tau)\,\mathbf{L}_m(\tau)\,, \tag{18}$$

where $\mathbf{H}$ is the free system Hamiltonian and $\boldsymbol{\xi}_n(t)$ is an operator-valued stochastic process with two-time correlations

$$\langle \boldsymbol{\xi}_n(t)\,\boldsymbol{\xi}_m(\tau)\rangle_{\boldsymbol{\xi}} = \alpha_{nm}(t,\tau) = \nu_{nm}(t,\tau) + \imath\hbar\,\mu_{nm}(t,\tau)\,. \tag{19}$$

In the classical limit, the dissipation kernel vanishes in the commutator expectation value for the operator noise process, but not in the Langevin equation's memory kernel.

Finally, using the notation of Ref. [15], the second-order (in the interaction) master equation [16–18] of the reduced density matrix $\boldsymbol{\rho}$ can be represented in terms of the noise correlation as

$$\dot{\boldsymbol{\rho}} = -\frac{\imath}{\hbar}\,[\mathbf{H},\boldsymbol{\rho}] + \boldsymbol{\mathcal{L}}_2\{\boldsymbol{\rho}\}\,, \tag{20}$$

with the second-order contribution given by the operation

$$\boldsymbol{\mathcal{L}}_2\{\boldsymbol{\rho}\} \equiv \frac{1}{\hbar^2}\sum_{nm}\left[\mathbf{L}_n,\boldsymbol{\rho}\,(\mathbf{A}_{nm}\diamond\mathbf{L}_m)^\dagger - (\mathbf{A}_{nm}\diamond\mathbf{L}_m)\,\boldsymbol{\rho}\right]\,, \tag{21}$$

where the $\mathbf{A}$ operators and $\diamond$ product define the second-order operators

$$(\mathbf{A}_{nm}\diamond\mathbf{L}_m)(t) \equiv \int_0^t d\tau\,\alpha_{nm}(t,\tau)\ \{\boldsymbol{\mathcal{G}}_0(t,\tau)\,\mathbf{L}_m(\tau)\}\,, \tag{22}$$

given the free system propagator $\boldsymbol{\mathcal{G}}_0(t,\tau):\boldsymbol{\rho}(\tau)\to\boldsymbol{\rho}(t)$. E.g. for a time-independent system Hamiltonian $\mathbf{H}$ we have the free system propagator

$$\boldsymbol{\mathcal{G}}_0(t,\tau)\,\boldsymbol{\rho} = e^{+\frac{\imath}{\hbar}\mathbf{H}(t-\tau)}\,\boldsymbol{\rho}\,e^{-\frac{\imath}{\hbar}\mathbf{H}(t-\tau)}\,. \tag{23}$$

This formalism also makes no assumptions as to the structure of the system, e.g. continuous or discrete, however the master equation is strictly perturbative, even for linear environments. Therefore the system Hamiltonian must contain sufficiently large transition energies to justify a perturbative expansion in the interaction, and the environment correlations cannot be excessively long ranged, else the expansion can be secular in time.

One context in which the influence functional, Langevin equation and master equation all work together seamlessly is in the quantum Brownian motion of linear systems [14, 19, 20]. In addition to a a quantum Langevin equation for non-commuting operators, linearity makes it possible in that case to have a Langevin equation for real classical stochastic processes from which general quantum correlation functions and the master equation can be exactly derived [19, 20].

B. The Correlation Function

From its microscopic definition, Eq. (10), the environmental correlation function is Hermitian in the sense of

$$\boldsymbol{\alpha}(t,\tau) = \boldsymbol{\alpha}^{\dagger}(\tau,t)\,, \tag{24}$$

and also positive definite in the sense of

$$\int_0^t d\tau_1 \int_0^t d\tau_2\, \mathbf{f}^{\dagger}(\tau_1)\, \boldsymbol{\alpha}(\tau_1,\tau_2)\, \mathbf{f}(\tau_2) \geq 0\,, \tag{25}$$

for all vector functions $\mathbf{f}(t)$ indexed by the noise. Hermiticity is relatively straightforward to see, whereas the positivity in Eq. (25) can be proven by inserting Eq. (10) and manipulating this expression into the form

$$\int_0^t d\tau_1 \int_0^t d\tau_2\, \mathbf{f}^{\dagger}(\tau_1)\, \boldsymbol{\alpha}(\tau_1,\tau_2)\, \mathbf{f}(\tau_2) = \left\langle \mathbf{Q}^{\dagger}\, \mathbf{Q} \right\rangle_{\mathrm{E}}\,, \tag{26}$$

in terms of the operator

$$\mathbf{Q} = \sum_n \int_0^t d\tau_1\, f_n(\tau_1)\, \mathbb{l}_n(\tau_1)\,, \tag{27}$$

and so positivity must follow, as the environment's density matrix is positive definite. Positivity and the noise decomposition (11) are the key properties from which the FDI arises.

Stationary correlations are defined by their invariance under time translations,

$$\boldsymbol{\alpha}(t,\tau) = \boldsymbol{\alpha}(t-\tau)\,, \tag{28}$$

and can produce asymptotically stationary (time-independent) master equations. Such correlations are produced when the environment is in an initially stationary state and its coupling operators in the Schrödinger picture are constant in time:

$$\boldsymbol{\rho}_{\mathrm{E}}(0) = \sum_i p_{\mathrm{E}}(\varepsilon_i)\, |\varepsilon_i\rangle\langle\varepsilon_i|\,, \tag{29}$$

yielding the correlation function

$$\alpha_{nm}(t,\tau) = \sum_{ij} p_{\mathrm{E}}(\varepsilon_i)\, \langle\varepsilon_i|\, \mathbf{l}_n\, |\varepsilon_j\rangle\, \overline{\langle\varepsilon_i|\, \mathbf{l}_m\, |\varepsilon_j\rangle}\, e^{+\imath\varepsilon_{ij}(t-\tau)}\,, \tag{30}$$

where $\overline{z} = z^*$ denotes the complex conjugate, $\varepsilon_{ij} \equiv \varepsilon_i - \varepsilon_j$, $|\varepsilon_i\rangle$ denotes the energy basis of the environment, and $p_{\mathrm{E}}(\varepsilon_i)$ are its stationary probabilities at the initial time. The associated characteristic function can be obtained quite directly from the mode sum

$$\boldsymbol{\alpha}(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega\, e^{+\imath\omega t}\, \tilde{\boldsymbol{\alpha}}(\omega)\,, \tag{31}$$

yielding the Fourier transform

$$\tilde{\alpha}_{nm}(\omega) \underline{\propto} 2\pi \sum_i p_{\mathrm{E}}(\varepsilon_i)\, \langle\varepsilon_i|\, \mathbf{l}_n\, |\varepsilon_i - \omega\rangle\, \overline{\langle\varepsilon_i|\, \mathbf{l}_m\, |\varepsilon_i - \omega\rangle}\,, \tag{32}$$

where the underscored proportionality here is strictly in reference to the continuum limit of the reservoir which relates environmental mode sums to integrals given the infinitesimal strength of individual environmental mode couplings. This can be more rigorously defined through the use of a finite spectral density function in place of the infinitesimal environment couplings.

Also of note are *quasi-stationary* correlations of the form

$$\boldsymbol{\alpha}(t,\tau) = \boldsymbol{\alpha}_{\mathrm{S}}(t-\tau) + \boldsymbol{\delta\alpha}(t+\tau)\,, \tag{33}$$

where $\boldsymbol{\alpha}_{\mathrm{S}}(t-\tau)$ denotes a stationary correlation function, or, more specifically, the stationary projection of $\boldsymbol{\alpha}(t,\tau)$, while $\boldsymbol{\delta\alpha}(t+\tau)$ is an additional non-stationary contribution. Such correlations will result from (time-independent) linear coupling to an environment with non-stationary initial state, such as a squeezed thermal state. In these cases the stationary projection of the correlation function does correspond to the stationary projection of the initial state of the environment. As for the non-stationary contributions, due to their highly oscillatory behavior in the late-time limit they typically lose effect asymptotically. Therefore, quasi-stationary correlations can produce an asymptotically stationary master equation with equivalent asymptotics as generated by their corresponding stationary correlation.

C. The Damping Kernel

Here we will use the Langevin equation (B24) to determine the role of the dissipation kernel and to motivate an appropriate method of renormalization. The correlation function $\boldsymbol{\alpha}(t,\tau)$ is positive definite and therefore the noise kernel $\boldsymbol{\nu}(t,\tau)$ must also be positive definite, which follows from eq. (25) by taking a real $\mathbf{f}(t)$. However, the dissipation kernel $\boldsymbol{\mu}(t,\tau)$ is not positive definite, but it is related to the damping kernel $\boldsymbol{\gamma}(t,\tau)$, which is given by

$$\boldsymbol{\mu}(t,\tau) = -\frac{\partial}{\partial\tau}\boldsymbol{\gamma}(t,\tau)\,, \tag{34}$$

and can be positive definite, negative definite or indefinite, as will be discussed in Sec. II C 2. For non-stationary noise, Eq. (34) is an incomplete definition, and constructing a symmetric damping kernel will require additional considerations. Assuming that relation (34) is sufficient, which is the case for stationary correlations, the nonlocal term present in the Langevin equation can be represented

$$\begin{aligned}\underbrace{\int_0^t d\tau\,\mu_{nm}(t,\tau)\,\mathbf{L}_m(\tau)}_{\text{dissipation}} &= \underbrace{\int_0^t d\tau\,\gamma_{nm}(t,\tau)\,\dot{\mathbf{L}}_m(\tau)}_{\text{damping}} \\ &- \underbrace{\gamma_{nm}(t,t)\,\mathbf{L}_m(t)}_{\text{renormalization}} + \underbrace{\gamma_{nm}(t,0)\,\mathbf{L}_m(0)}_{\text{slip}}\,.\end{aligned} \tag{35}$$

In the damping-kernel representation we now have three terms: nonlocal damping, renormalization forces, and transient "slip" forces. The nonlocal damping and renormalization forces will be more thoroughly considered in the following subsections. The transient slip is a pathology associated with factorized initial conditions, and can be avoided with the consideration of a properly correlated initial state [21].

Given a symmetric damping kernel, which is the case for stationary correlations, our quantum Langevin equation can then be expressed as

$$\begin{aligned}\hbar\,\dot{\mathbf{S}}(t) &= +\imath\,[\mathbf{H}_{\mathrm{eff}}(t),\mathbf{S}(t)] + \frac{\imath}{2}\sum_n \{\boldsymbol{\xi}_n(t),[\mathbf{L}_n(t),\mathbf{S}(t)]\} \\ &+ \imath\sum_{nm}\int_0^t d\tau\,\gamma_{nm}(t,\tau)\left\{\dot{\mathbf{L}}_m(\tau),[\mathbf{L}_n(t),\mathbf{S}(t)]\right\}\,,\end{aligned} \tag{36}$$

when discarding the transient slip and where the effective Hamiltonian is given by

$$\mathbf{H}_{\mathrm{eff}}(t) = \mathbf{H}(t) - \sum_{nm}\mathbf{L}_n(t)\,\gamma_{nm}(t,t)\,\mathbf{L}_m(t)\,. \tag{37}$$

We denote this term as the "effective Hamiltonian" and not the "renormalized Hamiltonian", because the correction may contain both divergences which require renormalization and terms which describe completely new environmentally-induced forces. A canonical example of this is non-equilibrium electrodynamics, wherein these corrections contain both the mass renormalization of the electron as well as magnetostatic forces between electrons [22].

1. The Energy-Damping Property

To demonstrate that a symmetric damping kernel can be given a physical interpretation we will consider the time evolution of the effective system Hamiltonian. First we will assume the following dynamical evolution of the system-coupling operators:

$$\hbar\,\dot{\mathbf{L}}_m(t) = +\imath\,[\mathbf{H}_{\rm eff}(t), \mathbf{L}_m]\ . \tag{38}$$

This relation is exact if all of the system coupling operators $\mathbf{L}_n$ commute, otherwise it is only perturbative. Now we substitute Eq. (37) into Eq. (36), apply the above relation, and integrate to obtain the following open-system energy as a function of time when neglecting the transient slip:

$$\mathbf{H}_{\rm eff}(t) = \mathbf{H}_{\rm eff}(0) - \mathbf{H}_\gamma(t) + \mathbf{H}_\xi(t)\,, \tag{39}$$

$$\mathbf{H}_\gamma(t) \equiv +\sum_{nm}\int_0^t d\tau \int_0^t d\tau'\,\gamma_{nm}(\tau,\tau')\left\{\dot{\mathbf{L}}_n(\tau'), \dot{\mathbf{L}}_m(\tau)\right\}\,, \tag{40}$$

$$\mathbf{H}_\xi(t) \equiv -\sum_n \frac{1}{2}\left\{\boldsymbol{\xi}_n(t), \dot{\mathbf{L}}_n(t)\right\}\ . \tag{41}$$

This relation reveals that a positive-definite damping kernel will only decrease the system energy, whereas a negative-definite damping kernel will only increase the system energy. The criteria for each condition will be more thoroughly covered in Sec. II C 2. This expression for $\mathbf{H}_\gamma(t)$ also contrasts *nonlocal damping* to *local damping*. Evaluation with a delta correlated damping kernel yields damping which is strictly dissipative at every instant of time whereas nonlocal damping is only guaranteed to have an accumulated dissipative effect since the initial time.

2. Classification of Stationary Damping

Two-time correlation functions can always be decomposed into a real noise kernel and dissipation kernel as in Eq. (11). The Hermiticity stated in Eq. (24) leads to the relations

$$\boldsymbol{\nu}(t,\tau) = \frac{1}{2}\left[\boldsymbol{\alpha}(t,\tau) + \boldsymbol{\alpha}^{\rm T}(\tau,t)\right]\ , \tag{42}$$

$$\boldsymbol{\mu}(t,\tau) = \frac{1}{2\imath\hbar}\left[\boldsymbol{\alpha}(t,\tau) - \boldsymbol{\alpha}^{\rm T}(\tau,t)\right]\ . \tag{43}$$

For stationary correlations $\boldsymbol{\alpha}(t\text{–}\tau)$, which lead to a symmetric damping kernel, one can introduce the Fourier transform $\tilde{\boldsymbol{\alpha}}(\omega) = \int_{-\infty}^{+\infty} d\tau\, e^{-\imath\omega\tau}\boldsymbol{\alpha}(\tau)$. The noise and damping kernels are then Hermitian in both noise index and frequency argument:

$$\tilde{\boldsymbol{\gamma}}(\omega) = \tilde{\boldsymbol{\gamma}}^\dagger(\omega) = \tilde{\boldsymbol{\gamma}}^*(-\omega)\,, \tag{44}$$

$$\tilde{\boldsymbol{\nu}}(\omega) = \tilde{\boldsymbol{\nu}}^\dagger(\omega) = \tilde{\boldsymbol{\nu}}^*(-\omega)\,. \tag{45}$$

The first equality in Eqs. (44)-(45) follows from the Fourier transform of Eq. (24), whereas the second equality follows from the fact that $\boldsymbol{\nu}(\tau,t) = \boldsymbol{\nu}^{\rm T}(t,\tau)$ and $\boldsymbol{\gamma}(\tau,t) = \boldsymbol{\gamma}^{\rm T}(t,\tau)$. Moreover, by Bochner's theorem both $\tilde{\boldsymbol{\alpha}}(\omega)$ and $\tilde{\boldsymbol{\nu}}(\omega)$ are positive-definite for all frequencies; see Appendix C. For stationary correlations, the damping kernel is uniquely defined by relation (34). Again the damping kernel $\tilde{\boldsymbol{\gamma}}(\omega)$ may be positive definite, negative definite, or indefinite.

As proven in Sec. II C 1, environments with positive-definite damping kernels are *damping* or *resistive* environments, while those with negative-definite damping kernels are *amplifying*. If the system coupling operators $\mathbf{L}_n$ are position operators, the damping terms correspond to forces linear in momentum. Stationary correlations are the easiest to dissect and the most well behaved. Their dissipation and damping kernels are related by

$$\tilde{\boldsymbol{\mu}}(\varepsilon) = \imath\varepsilon\,\tilde{\boldsymbol{\gamma}}(\varepsilon)\,, \tag{46}$$

and from the definition of the dissipation kernel in Eq. (43) and the double Hermiticity in Eq. (44)-(45), the damping kernel will be exactly positive or negative definite, respectively, if we have a strict inequality between positive and negative energy argumented environmental correlations:

$$\tilde{\boldsymbol{\alpha}}(-|\omega|) > \tilde{\boldsymbol{\alpha}}^*(+|\omega|) \quad \text{(Damping)}\,, \tag{47}$$

$$\tilde{\boldsymbol{\alpha}}(-|\omega|) < \tilde{\boldsymbol{\alpha}}^*(+|\omega|) \quad \text{(Amplifying)}\,, \tag{48}$$

with inequality in the sense of Appendix C. From Eq. (32), one can show that damping environments result when the initial stationary probability of the environment $p_{\rm E}(\varepsilon)$ is a monotonically decreasing function of the environment energy. Amplifying environments result from monotonically increasing functions or *population inversion*. The most common example of each being positive and negative temperature reservoirs. Furthermore, in the perturbative master-equation formalism, the above equations imply that damping environments induce a higher probability of transitions to lower energy states, while amplifying environments induce a higher probability of transitions to higher energy states.

III. NON-EQUILIBRIUM RELATIONS

A. Non-Equilibrium Fluctuation-Dissipation Relation & Inequality

From the definitions of the multivariate noise kernel $\boldsymbol{\nu}$ (42), dissipation kernel $\boldsymbol{\mu}$ (43) and damping kernel $\boldsymbol{\gamma}$ (46), one can prove the *fluctuation-dissipation inequality*:

$$\boldsymbol{\nu} \geq \pm\imath\hbar\,\boldsymbol{\mu}\,, \tag{49}$$

with inequality in the sense of Appendix C. In the case of stationary noise it takes the more useful form

$$\tilde{\boldsymbol{\nu}}(\omega) \geq \pm\hbar\omega\,\tilde{\boldsymbol{\gamma}}(\omega)\,, \tag{50}$$

in the Fourier domain where the ω would denote energy level transitions of the system; When the noise is univariate, the inequality can also be written

$$\tilde{\nu}(\omega) \geq |\hbar\omega\,\tilde{\gamma}(\omega)|\,. \tag{51}$$

To prove the general inequality, one simply notes that the noise kernel is the sum of two positive-definite kernels whereas the dissipation kernel is given by their difference. The essential point is that if there is any damping, or amplification, there will be quantum noise and Eq. (50) determines its lower bound. This is quite a departure from classical physics where noise can be made to vanish in the zero-temperature limit, although the lower bound of this inequality is satisfied by zero-temperature quantum noise since $\tilde{\boldsymbol{\alpha}}(|\omega|) = 0$ in that case.

For the case of a single collective system coupling, coupled to one or more environments, it is sufficient to define a fluctuation-dissipation relation

$$\tilde{\nu}(\omega) = \tilde{\kappa}(\omega)\,\tilde{\gamma}(\omega)\,, \tag{52}$$

$$\tilde{\kappa}(\omega) \equiv \frac{\tilde{\nu}(\omega)}{\tilde{\gamma}(\omega)}\,, \tag{53}$$

with $\tilde{\kappa}(\omega)$ being the fluctuation-dissipation kernel [10, 11] which relates fluctuations to dissipation. For multivariate noise one might use the symmetrized product

$$\tilde{\boldsymbol{\nu}}(\omega) = \frac{1}{2}\left[\tilde{\boldsymbol{\kappa}}(\omega)\,\tilde{\boldsymbol{\gamma}}(\omega) + \tilde{\boldsymbol{\gamma}}(\omega)\,\tilde{\boldsymbol{\kappa}}(\omega)\right]\,, \tag{54}$$

which would ensure $\tilde{\boldsymbol{\kappa}}(\omega)$ to be positive definite if $\tilde{\boldsymbol{\gamma}}(\omega)$ is, in accord with this being a (continuous) Lyapunov equation [23]. We will use this particular definition for quantum Brownian motion in the next section. Inequality (50) can now be restated as

$$\tilde{\boldsymbol{\kappa}}(\omega) \geq |\hbar\omega|\,, \tag{55}$$

for damping environments. Typically $\tilde{\boldsymbol{\kappa}}(\omega)$ will contain dependence upon the precise nature of the environment couplings $\mathbf{l}_n$.

B. Equilibrium Fluctuation-Dissipation Relation

Let us consider a time-independent system-environment interaction and environment Hamiltonian as well as initial stationary probabilities of the environment given by $p_{\rm E}(\varepsilon)$. If the FDR is to be independent of precisely how the

system and environment are coupled, then one can work out from the microscopic theory (32) that the FDR kernel must be a scalar quantity, directly related to the initial state of the environment by way of

$$\frac{\tilde{\kappa}(\omega)}{\hbar\omega} = \frac{p_{\rm E}(\varepsilon-\omega)+p_{\rm E}(\varepsilon)}{p_{\rm E}(\varepsilon-\omega)-p_{\rm E}(\varepsilon)}\,, \tag{56}$$

for all ε. To prove this one first applies relation (32) to definitions (42)-(43), and notes that if the dissipation and noise are related in a manner independent of the coupling then the two kernels must be related term-by-term in a sum over couplings.

But such an equality between the FDR kernel and mode probabilities implies the functional relation

$$p_{\rm E}(\varepsilon-\omega) = \left[\frac{\frac{\tilde{\kappa}(\omega)}{\hbar\omega}+1}{\frac{\tilde{\kappa}(\omega)}{\hbar\omega}-1}\right] p_{\rm E}(\varepsilon)\,, \tag{57}$$

where the ω translations can factor out. This factorization property is unique to exponential functions; therefore, only the thermal distribution $p_{\rm E}(\varepsilon) \propto e^{-\beta\varepsilon}$ can produce a fluctuation-dissipation relation which is generally coupling independent. We then have that

$$\tilde{\kappa}_T(\omega) \equiv \hbar\omega\coth\left(\frac{\hbar\omega}{2k_BT}\right)\,, \tag{58}$$

for the thermal distribution. One should be careful to note that the thermal FDR is not special because it exists, nor because of its simple form, but because of its complete independence of the details of the system-environment interaction. "Observable dependence" for non-equilibrium correlations was also noticed in Ref. [24]. In a more general context, the thermal FDR is also special because it ensures a relaxation to detailed balance in a coupling-invariant manner. In fact, these properties can be shown to be equivalent [15].

As a concrete example of an elegant yet non-equilibrium FDR, the late-time dominating stationary correlations for linear coupling to a squeezed thermal reservoir [25, 26] will produce the FDR kernel

$$\tilde{\kappa}^r_T(\omega) = \cosh[2\,r(\omega)]\,\hbar\omega\coth\left(\frac{\hbar\omega}{2k_BT}\right)\,, \tag{59}$$

where $r(\omega)$ is the squeezing parameter, which may be allowed to vary with the energy scale. One can easily see that this FDR also obeys inequality (50) as it must.

C. Fluctuation-Dissipation Equality

For stationary environments, we can more easily determine when equality is achieved between the fluctuations and dissipation in the sense of

$$\tilde{\boldsymbol{\nu}}(\omega) = \pm\hbar\omega\,\tilde{\boldsymbol{\gamma}}(\omega)\,. \tag{60}$$

Working backwards through the FDI proof, this implies

$$(1\pm1)\,\tilde{\boldsymbol{\alpha}}(\omega) + (1\mp1)\,\tilde{\boldsymbol{\alpha}}(-\omega)^{\rm T} = \mathbf{0}\,, \tag{61}$$

which can only occur for all ω if either the environment is damping with $\tilde{\boldsymbol{\alpha}}(+|\omega|)=0$ or amplifying with $\tilde{\boldsymbol{\alpha}}(-|\omega|)=0$. In the perturbative master-equation formalism, these equalities correspond to there being no environmentally-induced transitions to higher energies and lower energies respectively. From relation (32), we then have the constraint upon the environment couplings

$$\langle\varepsilon|\mathbf{l}_n|\varepsilon-\omega\rangle = 0 \quad \text{(Damping)}\,, \tag{62}$$
$$\langle\varepsilon|\mathbf{l}_n|\varepsilon+\omega\rangle = 0 \quad \text{(Amplifying)}\,, \tag{63}$$

which must hold for all environment energies ε for which there is any population $p_{\rm E}(\varepsilon)$. Assuming that the environment coupling $\mathbf{l}$ is not sparse at particular energies, then the damping environment must have all of its population in the lowest-energy state, $T=+0$ or $\beta=+\infty$, while the amplifying environment must have all of its population in the highest-energy state, $T=-0$ or $\beta=-\infty$. If the environment coupling $\mathbf{l}$ is sparse, then it must only be the case that the population of states lies on a boundary of the energy spectrum accessible by $\mathbf{l}$.

IV. NON-EQUILIBRIUM UNCERTAINTY PRINCIPLE

In the context of second-order perturbation theory, quantum noise is effectively Gaussian in the influence functional, and Gaussian noise is equivalent to that arising from linear coupling to a bath of harmonic oscillators. Therefore, any violations of Eq. (50) must correspond to an environment of oscillators in a non-quantum state. In the phase-space or Wigner function representation [27], HUP violating states of the environment can be constructed which violate the quantum FDI. Such is the case for the classical vacuum, which has vanishing fluctuations yet finite damping. Now we shall show that FDI violating noise can also relax the system into a HUP violating state.

Let us consider weakly influencing a system of oscillators at resonance, all with mass m and frequency ω, via position-position coupling to some phenomenological set of noise processes with resistive correlation $\tilde{\boldsymbol{\alpha}}(\omega)$. We do not assume the system-environment couplings to be identical, nor do we neglect the presence of cross-correlations among the noise processes. Furthermore, the system may relax in general to stationary states which do not correspond to thermal equilibrium. A simple example would be a situation where there is a constant heat flow through the system between two thermal baths at different temperatures.

From the results of Ref. [20, 28] and the second-order master equation coefficients (21), the damping kernel $\tilde{\boldsymbol{\gamma}}(\omega)$ will play the role of the dissipation coefficients and the noise kernel $\tilde{\boldsymbol{\nu}}(\omega)$ will play the role of the normal diffusion coefficients in the Fokker-Planck or master equation. Integrals over the two kernels will then provide the system renormalization and anti-diffusion coefficients respectively. Given sufficient dissipation and bandwidth-limited correlations, the system will relax into a Gaussian state which satisfies the Lyapunov equation

$$\tilde{\boldsymbol{\nu}}(\omega) = \frac{1}{2}\left[\left(\frac{2}{m}\boldsymbol{\sigma}_{pp}\right)\tilde{\boldsymbol{\gamma}}(\omega) + \tilde{\boldsymbol{\gamma}}(\omega)\left(\frac{2}{m}\boldsymbol{\sigma}_{pp}\right)\right], \tag{64}$$

for the momentum covariance, which has elements $\langle p_i p_j \rangle$, and

$$\boldsymbol{\sigma}_{xx} = \frac{1}{(m\omega)^2}\boldsymbol{\sigma}_{pp}, \tag{65}$$

$$\boldsymbol{\sigma}_{xp} = \mathbf{0}, \tag{66}$$

for the remaining covariances in the phase-space (Wigner function) representation [27], and to lowest order in the system-environment interaction. Comparing Eq. (64) to Eq. (54), we can express our covariances as

$$\boldsymbol{\sigma}_{xx} = \frac{1}{2m\omega^2}\tilde{\boldsymbol{\kappa}}(\omega), \tag{67}$$

$$\boldsymbol{\sigma}_{pp} = \frac{m}{2}\tilde{\boldsymbol{\kappa}}(\omega), \tag{68}$$

in terms of the FDR kernel $\tilde{\boldsymbol{\kappa}}(\omega)$. So far our FDR kernel remains phenomenological and not microscopically derived. However, it must be positive definite for this to describe a physical state. Since $\tilde{\boldsymbol{\kappa}}(\omega)$ is positive definite, we can transform to the basis in which it diagonalizes. If $\tilde{\boldsymbol{\kappa}}(\omega)$ is a scalar quantity, then this is simply the normal basis of the free system. For each mode in this basis we have the phase-space covariance

$$\boldsymbol{\sigma}_n = \begin{bmatrix} \sigma_n^{xx} & \sigma_n^{xp} \\ \sigma_n^{px} & \sigma_n^{pp} \end{bmatrix} = \begin{bmatrix} \frac{1}{2m\omega^2}\tilde{\kappa}_n(\omega) & 0 \\ 0 & \frac{m}{2}\tilde{\kappa}_n(\omega) \end{bmatrix}, \tag{69}$$

which must then satisfy the generalized Heisenberg uncertainty relation due to Schrödinger [29, 30]:

$$\langle \Delta x^2 \rangle \langle \Delta p^2 \rangle - \frac{1}{2}\langle \{\Delta x, \Delta p\} \rangle \geq \frac{\hbar^2}{4}, \tag{70}$$

or in terms of the phase-space covariance

$$\det(\boldsymbol{\sigma}) \geq \frac{\hbar^2}{4} \tag{71}$$

and, therefore, it must be the case that

$$\tilde{\kappa}_n(\omega) \geq \hbar\omega, \tag{72}$$

for all $\omega > 0$. But this is equivalent to our previous statement

$$\tilde{\boldsymbol{\kappa}}(\omega) \geq \hbar\omega, \tag{73}$$

in terms of positive definiteness as ω is a scalar quantity. So not only do FDI violating correlations arise from HUP violating environment states, they can also produce HUP violating system states via dissipation and diffusion (and decoherence).

Furthermore we can say that in the weak-damping limit, the scalar FDR kernel $\tilde{\kappa}(\omega)$ precisely determines the (asymptotic) non-equilibrium uncertainty product

$$\det(\boldsymbol{\sigma}) = \left(\frac{1}{2}\frac{\tilde{\kappa}(\omega)}{\omega}\right)^2 , \tag{74}$$

for a single system mode of frequency ω. Larger FDR kernels naturally produce larger uncertainty and minimal FDR kernels (zero temperature) produce minimal uncertainty. Non-perturbative results require evaluation of the exact expressions found in Refs. [20, 28] for a single system oscillator and multiple system oscillators respectively.

V. DISCUSSION

In this paper we have derived a fluctuation-dissipation inequality (FDI) for an open quantum system interacting with a non-equilibrium environment from the microscopically-derived environment correlation function and recovered the well-known fluctuation-dissipation relation (FDR) for a thermal environment. The FDI is a very general statement contrasting quantum noise to classical noise, and is satisfied even for non-equilibrium fluctuations. Simply put, quantum fluctuations are bounded below by quantum dissipation, whereas classically the fluctuations can be made to vanish. The lower bound of this inequality is exactly satisfied by zero-temperature noise and is in accord with the Heisenberg uncertainty principle (HUP). FDI violating correlations arise from HUP violating states of the environment and can relax the open system into HUP violating states. Therefore, the FDI can be viewed as an open-system corollary to the HUP both from microscopic and phenomenological considerations. Analogously, the non-equilibrium FDR also determines the non-equilibrium uncertainty product, most directly in the limit of weak-damping [see Eq. (74)], and the corresponding FDI implies the HUP.

As an example of a practical application of the FDI, we note that there is a large class of situations where applying the FDI to the zero-temperature solution of an open system can produce general bounds on all related non-equilibrium solutions. For instance, consider a possibly-multipartite quantum optical and/or mechanical system with linear photonic and phononic environments, as would be the case with a system linearly coupled to the environment field operators and where the system moves very slowly relative to the speeds of sound and light. In the case we consider, the damping kernels are determined by the commutator of the field operators and not by the environment's state (see for instance Ref. [31]). Now let us say that we can solve the zero temperature case (or the result is already available), but we cannot solve the more-realistic non-equilibrium case, either because the realistic case is too difficult or because we lack a sufficient understanding of the non-equilibrium state of the combined environment. If we solve the zero-temperature case, and in our derivation we track all instances of the noise kernel and its resulting diffusion coefficients, then at the end of the calculation we can apply the FDI to obtain bounds on the true solution. As a trivial example, one can apply the FDI to our resulting Eq. (74). However, in a more complicated system, and particularly if the full-time evolution is considered, results of such an analysis can be very nontrivial.

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Appendix A: Kernels for Two-Level Environments

A model with an environment consisting of a set of two-level systems provides a useful illustration of how very non-linear environments can give rise to nonintuitive noise and dissipation kernels, which nevertheless respect fluctuation-dissipation relations. We consider the time-independent environment Hamiltonian

$$\mathbf{H}_{\mathrm{E}} = \sum_k \frac{\omega_k}{2}\boldsymbol{\sigma}_z^{(k)} , \tag{A1}$$

where $\boldsymbol{\sigma}^{(k)}$ denotes a Pauli spin matrix of the k^{th} particle. For the environment coupling operator, we take the most general time-independent non-commuting operator

$$\mathbf{l}_n = \sum_k \vec{g}_{kn} \cdot \vec{\boldsymbol{\sigma}}^{(k)} \,, \tag{A2}$$

with the vector of coefficients and spin matrices given by

$$\vec{g}_{kn} \equiv (g^x_{kn}, g^y_{kn}, 0) \,, \tag{A3}$$
$$\vec{\boldsymbol{\sigma}} \equiv (\boldsymbol{\sigma}_x, \boldsymbol{\sigma}_y, \boldsymbol{\sigma}_z) \,. \tag{A4}$$

Assuming a factorized initial state for the environment, with the state of each spin initially given by

$$\boldsymbol{\rho}^{(k)}_{\text{E}} = \begin{bmatrix} 1-\delta(\omega_k) & q(\omega_k) \\ q(\omega_k)^* & 1+\delta(\omega_k) \end{bmatrix} , \tag{A5}$$

it is then straightforward to calculate the noise and damping kernels to be

$$\nu_{nm}(t) = \sum_k (\vec{g}_{kn} \cdot \vec{g}_{km}) \, \cos(\omega_k t) \,, \tag{A6}$$

$$\gamma_{nm}(t) = \sum_k (\vec{g}_{kn} \cdot \vec{g}_{km}) \, \cos(\omega_k t) \, \frac{\delta(\omega_k)}{\hbar \omega_k} \,, \tag{A7}$$

which satisfies the non-equilibrium FDR

$$\tilde{\boldsymbol{\nu}}(\omega) = \frac{\hbar\omega}{\delta(\omega)} \, \tilde{\boldsymbol{\gamma}}(\omega) \,, \tag{A8}$$

and, since $-1 \leq \delta(\omega) \leq +1$ by the positivity of $\boldsymbol{\rho}^{(k)}_{\text{E}}$, this relation satisfies the FDI. For a thermal state we have

$$\delta(\omega) = \tanh\!\left(\frac{\hbar\omega}{2k_B T} \right) . \tag{A9}$$

which also satisfies the FDT. However, notice that the noise kernel has no relation to the temperature or even to the initial state of the environment, other than the fact that the environment spins are taken to be initially uncorrelated. Instead, the damping kernel decreases with temperature to satisfy the FDT.

Appendix B: The Quantum Langevin Equation

The quantum Langevin equation [14] is well understood in the context of bilinear position-position couplings between system and environment. Here we would like to extend the simpler theory by considering quantum Langevin equations that correspond to the same class of Gaussian influences which appear in the second-order master-equation and influence-functional formalisms. We begin with the same kind of Hamiltonian for our system and environment:

$$\mathbf{H}_{\text{C}} = \mathbf{H} + \mathbf{H}_{\text{I}} + \mathbf{H}_{\text{E}} \,, \tag{B1}$$
$$\mathbf{H}_{\text{I}}(t) = \sum_n \mathbf{L}_n(t) \otimes \mathbf{l}_n(t) \,. \tag{B2}$$

The Heisenberg equations of motion for any system operator $\mathbf{S}(t)$, as driven by the environment, are therefore given by

$$\hbar \, \dot{\mathbf{S}}(t) = +\imath \, [\mathbf{H}(t), \mathbf{S}(t)] + \imath \sum_n [\mathbf{l}_n(t) \otimes \mathbf{L}_n(t), \mathbf{S}(t)] \,. \tag{B3}$$

To generate a Gaussian influence, we specify the environment to be linear in its driven dynamics

$$\mathbf{H}_{\text{E}}(t) = \sum_k \frac{1}{2} \left(m_k^{-1}(t) \, \boldsymbol{\pi}_k(t)^2 + c_k(t) \, \mathbf{q}_k(t)^2 \right) , \tag{B4}$$

$$\mathbf{l}_n(t) = \sum_k \left(g^q_{kn}(t) \, \mathbf{q}_k(t) + g^{\pi}_{kn}(t) \, \boldsymbol{\pi}_k(t) \right) . \tag{B5}$$

To approximate nonlinear environments perturbatively, one would match the influence kernels at the end of the calculation.

Let us define the "phase-space" vectors

$$\mathbf{z}_k \equiv (\mathbf{q}_k, \boldsymbol{\pi}_k)\,, \tag{B6}$$

$$\mathbf{g}_{kn}(t) \equiv (g^q_{kn}(t), g^\pi_{kn}(t))\,. \tag{B7}$$

The Heisenberg equations of motion for the environment operators, as driven by the system, can then be expressed conveniently as

$$\dot{\mathbf{z}}_k(t) = \mathbf{F}_k(t)\,\mathbf{z}_k(t) + \sum_n \boldsymbol{\varepsilon}\,\mathbf{g}_{kn}(t)\,\mathbf{L}_n(t)\,, \tag{B8}$$

$$\mathbf{F}_k(t) \equiv \begin{bmatrix} 0 & m_k^{-1}(t) \\ -c_k(t) & 0 \end{bmatrix}, \tag{B9}$$

$$\boldsymbol{\varepsilon} \equiv \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix}, \tag{B10}$$

so that the driven solutions of the environment operators are given by

$$\mathbf{z}_k(t) = \boldsymbol{\Phi}_k(t)\,\mathbf{z}_k(0) + \int_0^t d\tau\,\boldsymbol{\Phi}_k(t,\tau)\sum_n \boldsymbol{\varepsilon}\,\mathbf{g}_{kn}(\tau)\,\mathbf{L}_n(\tau)\,, \tag{B11}$$

in terms of the free environment transition matrix $\boldsymbol{\Phi}_k$, which satisfies the relations

$$\dot{\boldsymbol{\Phi}}_k(t) = \mathbf{F}_k(t)\,\boldsymbol{\Phi}_k(t)\,, \tag{B12}$$

$$\boldsymbol{\Phi}_k(0) = \mathbf{1}\,, \tag{B13}$$

$$\boldsymbol{\Phi}_k(t,\tau) \equiv \boldsymbol{\Phi}_k(t)\,\boldsymbol{\Phi}_k(\tau)^{-1}\,. \tag{B14}$$

The driven solutions can then be written as

$$\mathbf{l}_n(t) = \boldsymbol{\xi}_n(t) + 2\sum_m \int_0^t d\tau\,\mu_{nm}(t,\tau)\,\mathbf{L}_m(\tau)\,, \tag{B15}$$

$$\boldsymbol{\xi}_n(t) = \sum_k \mathbf{g}_{kn}(t)^{\mathrm{T}}\,\boldsymbol{\Phi}_k(t)\,\mathbf{z}_k(0)\,, \tag{B16}$$

$$\mu_{nm}(t,\tau) = \frac{1}{2}\sum_k \mathbf{g}_{kn}(t)^{\mathrm{T}}\,\boldsymbol{\Phi}_k(t,\tau)\,\boldsymbol{\varepsilon}\,\mathbf{g}_{km}(\tau)\,, \tag{B17}$$

where μ is so far only proven to be a memory kernel in the driven solution. We further wish to prove that, when assuming the environment to be initially in a Gaussian state, $\boldsymbol{\xi}_n(t)$ is an operator-valued Gaussian stochastic process, with two-time correlation function

$$\langle \boldsymbol{\xi}_n(t)\,\boldsymbol{\xi}_m(\tau)\rangle_{\boldsymbol{\xi}} = \alpha_{nm}(t,\tau) = \nu_{nm}(t,\tau) + \imath\hbar\,\mu_{nm}(t,\tau)\,, \tag{B18}$$

in terms of the noise kernel ν and dissipation kernel μ, given by

$$\nu_{nm}(t,\tau) = \left\langle \frac{1}{2}\,\{\boldsymbol{\xi}_n(t), \boldsymbol{\xi}_m(\tau)\} \right\rangle_{\boldsymbol{\xi}}\,, \tag{B19}$$

$$\mu_{nm}(t,\tau) = \left\langle \frac{1}{2\imath\hbar}\,[\boldsymbol{\xi}_n(t), \boldsymbol{\xi}_m(\tau)] \right\rangle_{\boldsymbol{\xi}}\,. \tag{B20}$$

To prove this, we must relate the memory kernel to the commutator. From Eq. (B16), the commutator is given by

$$[\boldsymbol{\xi}_n(t), \boldsymbol{\xi}_m(\tau)] = \imath\hbar\sum_k \mathbf{g}_{kn}(t)^{\mathrm{T}}\,\boldsymbol{\Phi}_k(t)\,\boldsymbol{\varepsilon}\,\boldsymbol{\Phi}_k(\tau)\,\mathbf{g}_{km}(\tau)\,. \tag{B21}$$

Note that for any two-dimensional matrix $\mathbf{A}$

$$\mathbf{A}^{-1} = \frac{\boldsymbol{\varepsilon}\,\mathbf{A}\,\boldsymbol{\varepsilon}}{\det \mathbf{A}}\,, \tag{B22}$$

with $\boldsymbol{\varepsilon}^2 = \mathbf{1}$. It therefore suffices to prove that $\det \boldsymbol{\Phi}_k(t) = 1$. First note that $\det \boldsymbol{\Phi}_k(0) = \det \mathbf{1} = 1$. Then consider its dynamics:

$$\begin{aligned}
\frac{d}{dt} \det \boldsymbol{\Phi}_k(t) &= \frac{d}{dt} e^{\log \det \boldsymbol{\Phi}_k(t)} \\
&= \frac{d}{dt} e^{\mathrm{Tr} \log \boldsymbol{\Phi}_k(t)} \\
&= \left[\frac{d}{dt} \mathrm{Tr} \log \boldsymbol{\Phi}_k(t) \right] e^{\mathrm{Tr} \log \boldsymbol{\Phi}_k(t)} \\
&= \mathrm{Tr} \left[\dot{\boldsymbol{\Phi}}_k(t) \, \boldsymbol{\Phi}_k(t)^{-1} \right] \det \boldsymbol{\Phi}_k(t) \\
&= \mathrm{Tr}[\mathbf{F}_k(t)] \, \det \boldsymbol{\Phi}_k(t) \\
&= 0 \, .
\end{aligned} \tag{B23}$$

It was erroneously reported in Ref. [15] that the relation (B20) did not always hold for parametric bath oscillators.

Substituting (B15) back into (B3) results in a quantum Langevin equation for the open system. However, due to the fact that $\mathbf{l}(t)$ commutes with system operators at all times whereas the operator noise $\boldsymbol{\xi}(t)$ and dissipation separately do not, there is no unique method of expressing the quantum Langevin equation in a manifestly Hermitian manner. All representations generate equivalent solutions, but some are more robust with respect to approximations than others. In particular, non-Hermitian representations are rather pathological as they only preserve Hermiticity after noise averaging. The second-order adjoint master equation [15] would appear to suggest the following representation:

$$\hbar \dot{\mathbf{S}}(t) = +\imath \, [\mathbf{H}(t), \mathbf{S}(t)] + \frac{\imath}{2} \sum_n \{\mathbf{l}_n(t), [\mathbf{L}_n(t), \mathbf{S}(t)]\} \, , \tag{B24}$$

$$\mathbf{l}_n(t) = \boldsymbol{\xi}_n(t) + 2 \sum_m \int_0^t d\tau \, \mu_{nm}(t, \tau) \, \mathbf{L}_m(\tau) \, . \tag{B25}$$

Although we have derived this Langevin equation under the assumption of a linear environment, we may apply it self-consistently for any Gaussian influence. The quantum Langevin equation here is nonperturbative. However, it only approximates non-Gaussian environments in a perturbative manner.

Finally note that in the corresponding classical Langevin equation, $\boldsymbol{\xi}_n(t)$ would be real Gaussian noise with two-time correlation ν_{nm}, and the dissipation kernel would only appear as given in Eq. (B25). That the dissipation kernel appears in the operator-noise average is a quantum feature.

Appendix C: Inequality of Operators

The positivity of a matrix kernel $\boldsymbol{\kappa}(t, \tau)$ in the sense of

$$\int_0^t d\tau_1 \int_0^t d\tau_2 \, \mathbf{f}^\dagger(\tau_1) \, \boldsymbol{\kappa}(\tau_1, \tau_2) \, \mathbf{f}(\tau_2) \geq 0 \, , \tag{C1}$$

for all vector functions $\mathbf{f}(t)$, is the natural extension of the positivity of a matrix $\mathbf{K}$ in the sense of

$$\mathbf{F}^\dagger \, \mathbf{K} \, \mathbf{F} \geq 0 \, , \tag{C2}$$

for all vectors $\mathbf{F}$. The time arguments of a kernel can be viewed as the (continuous) indices of a matrix. The fact that we have two sets of indices, time-t and noise-n, is indicative of a convenient block-matrix structure being employed.

Given a notion of positivity for a matrix, there is a natural notion of inequality that can exist between matrices. If a matrix $\mathbf{K}$ is positive definite in the above sense, then we can express this more succinctly as

$$+\mathbf{K} > \mathbf{0} \, , \tag{C3}$$

$$-\mathbf{K} < \mathbf{0} \, , \tag{C4}$$

where $\mathbf{0}$ is the matrix with zero in every entry. Therefore if $\mathbf{K} - \mathbf{J}$ is a positive-definite matrix, then we can say

$$\mathbf{K} > \mathbf{J} \, , \tag{C5}$$

and if $\mathbf{K} \pm \mathbf{J}$ is a positive-definite matrix, then we can say

$$\mathbf{K} > \pm\mathbf{J}\,. \tag{C6}$$

Note that, unlike scalar quantities, kernels and matrices cannot always be ordered in this way. The relationship between matrices can be indetermined.

A famous theorem for stationary kernels, where $\kappa(t,\tau) = \kappa(t-\tau)$, is due to Bochner and states that if κ is positive definite, then it follows that the Fourier transform is nonnegative: $\tilde{\kappa}(\omega) \geq 0$ for all ω. For stationary matrix kernels $\boldsymbol{\kappa}(t-\tau)$, Bochner's theorem naturally implies that the Fourier transform is positive definite: $\tilde{\boldsymbol{\kappa}}(\omega) \geq \mathbf{0}$ for all ω. The basic idea behind the theorem in this case can be understood by realizing that for stationary kernels the time integration domain in Eq. (C1) can be extended to $(-\infty, \infty)$. Taking then into account that the Fourier transform of a convolution is simply the product of the Fourier transforms, together with Parseval's theorem for Fourier transforms, Eq. (C1) becomes equivalent to

$$\int_{-\infty}^{\infty} d\omega\, \tilde{\mathbf{f}}^{\dagger}(\omega)\, \tilde{\boldsymbol{\kappa}}(\omega)\, \tilde{\mathbf{f}}(\omega) \geq 0\,, \tag{C7}$$

for arbitrary $\tilde{\mathbf{f}}(\omega)$, which implies $\tilde{\boldsymbol{\kappa}}(\omega) \geq \mathbf{0}$ for all ω.

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