

# Influence of Non-Markovian Dynamics in Equilibrium Uncertainty-Relations

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Contrary to the conventional wisdom that deviations from standard thermodynamics originate from the strong coupling to the bath, it is shown that in quantum mechanics, these deviations originate from the uncertainty principle and are supported by the non-Markovian character of the dynamics. Specifically, it is shown that the lower bound of the dispersion of the total energy of the system, imposed by the uncertainty principle, is dominated by the bath power spectrum and therefore, quantum mechanics inhibits the system thermal-equilibrium-state from being described by the canonical Boltzmann's distribution. We show that for a wide class of systems, systems interacting via central forces with pairwise-self-interacting environments, this general observation is in sharp contrast to the classical case, for which the thermal equilibrium distribution, irrespective of the interaction strength, is *exactly* characterized by the canonical Boltzmann distribution and therefore, no dependence on the bath power spectrum is present. We define an *effective coupling* to the environment that depends on all energy scales in the system and reservoir interaction. Sample computations in regimes predicted by this effective coupling are demonstrated. For example, for the case of strong effective coupling, deviations from standard thermodynamics are present and, for the case of weak effective coupling, quantum features such as stationary entanglement are possible at high temperatures.

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## I. INTRODUCTION

Thermodynamics was developed before the modern atomistic description of Nature was formulated. Statistical mechanics then led to an understanding of the laws of thermodynamics in terms of a microscopic description, thus closing the gap between macroscopic and microscopic description. Due to the interest in quantum technologies, there is a major ongoing effort to develop a consistent and well defined extension of thermodynamics to the quantum regime<sup>1-3</sup>. However, the majority of these theories are primarily based on Boltzmann's original ideas and are therefore plagued by issues concerning irreversibility, the origin of the second law, the relation between physics and information, the meaning of ergodicity, etc. (see, e.g., Refs.<sup>4,5</sup>).

Despite these issues, progress has been made. It is now well known that, e.g., Onsager's regression hypothesis fails in the quantum realm<sup>6,7</sup> and that non-Markovian dynamics are relevant in a variety of fields and applications, from foundations<sup>2,8</sup>, to nuclear physics<sup>9</sup>, quantum metrology<sup>10-12</sup> and biological systems (see, e.g.,<sup>13</sup> and references therein). It is also known that the thermal equilibrium state of a quantum system *strongly* coupled to a thermal bath deviates from the canonical Boltzmann distribution<sup>1,14-17</sup>. This is also expected to occur in the classical case<sup>18</sup>. Since both are incoherent thermal stationary situations, one would anticipate that the quantum

system is devoid of any coherence and hence, based on the decoherence program<sup>19</sup>, that both distributions should coincide. However, one might also suggest that the entanglement between the system and the bath, which has no classical counterpart, could introduce quantum-classical deviations<sup>1</sup>. Furthermore, the fact that extra deviations could be present even if the entanglement between the system and the bath is zero<sup>20</sup> or even in the weak field regime<sup>21,22</sup> makes the situation even more intriguing.

To explore this issue in greater detail, it is advantageous to find a situation where the classical and the quantum contributions to the deviation from the Boltzmann distribution can be clearly isolated and examined. Here we show that *irrespective of the interaction strength*, there are no deviations from Boltzmann's distribution for the generic case of a classical system interacting via central forces with a pairwise-self-interacting environment. Thus, if after quantum-mechanically treating the same case, deviations from the canonical Boltzmann's distribution are present, then they are purely quantum in nature. As shown below, deviations do appear not only for this systems but in general and, based on completely universal arguments, are shown to rely on the *uncertainty principle* characteristic of quantum mechanics and the power spectrum of the environment.

Specifically, the uncertainty principle not only inhibits the system's thermal-equilibrium-state from being described by the canonical Boltzmann distribution, but for

each system-bath interaction it also selects which thermal equilibrium states are physically accessible. This latter result, formulated here for the first time within the framework of quantum thermodynamics, constitutes the cornerstone of the theory of pointer states (the states which are robust against the presence of the environment)<sup>19</sup> and could have deep consequences for an understanding of the thermalization of quantum systems.

We note that that Romero & Oppenheim<sup>21</sup> as well as Geva *et al.*<sup>22</sup> previously addressed the issue, using a detailed master equation approach, of deviations from the quantum canonical state, even in the weak field regime. Here we display the quantum origin of those deviations for the large family of physical systems presented above, and clearly display intriguing aspects of the strong and weak coupling regimes.

### A. Classical Thermal-Equilibrium-State for a System Interacting via Central Forces with a Pairwise-Self-Interacting Environment.

The classical and quantum thermal state will turn out to be significantly different for the following general class of systems. We consider a classical particle of mass  $m$  with potential energy  $U_S(q)$  and Hamiltonian  $H_S(p, q) = \frac{1}{2m}p^2 + U_S(q)$ , and a bath of  $\mathfrak{N}$  classical particles interacting via the central force potential  $U_B^{i,j}(\mathbf{q}_i - \mathbf{q}_j)$  and Hamiltonian  $H_B(\mathbf{p}, \mathbf{q}) = \sum_j^{\mathfrak{N}} [\frac{1}{2m_j} \mathbf{p}_j^2 + \sum_{i,j} U_B^{i,j}(\mathbf{q}_i - \mathbf{q}_j)]$ . The classical system interacts with this bath via the central force potential energy  $\mathcal{V}_j(\mathbf{q}_j - q)$ , so that the total Hamiltonian is given by

$$H = H_S(p, q) + H_B(\mathbf{p}, \mathbf{q}) + \sum_j^{\mathfrak{N}} \mathcal{V}_j(\mathbf{q}_j - q). \quad (1)$$

In classical statistical mechanics, the thermal equilibrium distribution of the system S is defined by

$$\rho_S(p, q) = \frac{1}{Z} \int \prod_j^{\mathfrak{N}} d\mathbf{p}_j d\mathbf{q}_j \exp[-H(p, q, \mathbf{p}_j, \mathbf{q}_j)\beta], \quad (2)$$

where  $Z = \int \prod_j^{\mathfrak{N}} d\mathbf{p}_j d\mathbf{q}_j \int dp dq \exp(-H\beta)$  denotes the partition function of the total system, with  $\beta = 1/k_B T$  and  $T$  being the temperature of the environment. In reaching the equilibrium thermal state in Eq. (2) [also in Eq. (4) below], it is assumed that the central system S gets coupled at  $t = -\infty$  to the initially thermalized bath B. Therefore, the equilibrium state  $\rho_S(p, q)$  turns out to be an eigenmode of the full dissipative dynamics<sup>23–25</sup>.

The integral over  $\mathbf{p}_j$  in Eq. (2) trivially cancels out with the corresponding contribution in  $Z$  (see Appendix A for details). Due to the form of the dependence of  $\mathcal{V}_j$  and  $U_B^{i,j}$  on  $\mathbf{q}_i$ ,  $\mathbf{q}_j$  and  $q$ , the integrals over  $\{\mathbf{q}_j\}$  can be appropriately manipulated, with the net result that they cancel out with

the corresponding contribution in  $Z$ . Thus,

$$\rho_S(p, q) = Z_S^{-1} \exp[-H_S(p, q)\beta], \quad (3)$$

where  $Z_S = \int \prod_j^{\mathfrak{N}} dp_j dq_j \exp(-H_S\beta)$ . Hence, the thermal equilibrium distribution of a bounded particle in contact with a pairwise-self-interacting thermal bath via central forces, *irrespective of the coupling strength*, is exactly given by the canonical Boltzmann distribution. If the system S is composite, this result remains valid if each constituent of S is coupled to its own independent bath.

This result is enlightening because, in the strong coupling regime, there is no apparent physical reason why the equilibrium thermodynamic properties of a system are independent, for a wide class of systems, of both the nature of the bath to which it is coupled and of the functional form of the observables that mediate the interaction. For this particular type of systems, the physical picture that emerges from this result is that *in the long time regime*, any dissipative mechanism is equally effective in taking the system to thermal equilibrium. In other words, dissipative dynamics can contract the classical phase-space volume with no fundamental restriction and therefore, the resultant equilibrium state is independent of the dissipative coupling and the rate at which equilibrium [Eq. (3)] is reached. This suggests that the concept of intrinsic and extensive thermodynamic variables<sup>4</sup> can be extended, in some cases, to the strong coupling regime. By contrast, and as shown below, in the quantum realm dissipative mechanisms are accompanied by decoherence effects and are bath-nature and coupling-particularities sensitive<sup>19,26</sup> and are then capable of inducing a variety of different thermal states.

## II. QUANTUM THERMAL-EQUILIBRIUM-STATE

The thermal-equilibrium-state of a classical system interacting via central forces with a pairwise-self-interacting environment was discussed above. The quantum treatment discussed below extends beyond this particular type of system because it is performed on more general grounds. In doing so, the total Hamiltonian in Eq. (1) is replaced here by the general expression  $\hat{H} = \hat{H}_S + \hat{H}_B + \hat{V}$ . Based on the general description given in<sup>3,15,16</sup>, one can easily extend the classical definition in Eq. (2) to the quantum regime, namely,

$$\hat{\rho}_S = \frac{1}{Z} \text{tr}_B \exp \left\{ - \left[ \hat{H}(\hat{p}, \hat{q}, \hat{\mathbf{p}}_j, \hat{\mathbf{q}}_j) \right] \beta \right\}. \quad (4)$$

The operator character of the various terms in this expression and their commutativity relations prevent us from proceeding as we did in the classical case. However, these very same commutativity relations allow the immediate

formulation of the following set of inequalities:

$$[\hat{H}_S, \hat{V}] \neq 0 \Rightarrow \Delta \hat{H}_S \Delta \hat{V} \geq \frac{1}{2} |[\hat{H}_S, \hat{V}]|, \quad (5a)$$

$$[\hat{V}, \hat{H}_B] \neq 0 \Rightarrow \Delta \hat{V} \Delta \hat{H}_B \geq \frac{1}{2} |[\hat{V}, \hat{H}_B]|, \quad (5b)$$

where  $\Delta \hat{O} = \sqrt{\langle \hat{O}^2 \rangle - \langle \hat{O} \rangle^2}$  denotes the standard deviation of  $\hat{O}$ , with  $\langle \hat{O} \rangle = \text{tr}(\hat{O}\hat{\rho})$ ,  $\hat{\rho}$  being the thermal equilibrium state of the system S and the bath B. Because of the thermal character of the density operator discussed here, Eqs. (5) incorporate classical as well as quantum uncertainty, however, the lower bound in both cases differs. Equation (5) applies to *any* system, bath, and system-bath coupling Hamiltonians, but will prove particularly noteworthy in the case of Eq. (1) where the classical  $\rho_S$  is Boltzmann.

For example, consider  $[\hat{H}_S, \hat{V}] = 0$ . This implies a pure decohering interaction, which can be treated here in the framework of fluctuations without dissipation<sup>27</sup>. The equilibrium state is an incoherent mixture of system's eigenstates and is expected to be well characterized by the canonical Boltzmann distribution<sup>28</sup>. In this case  $[\hat{H}_S, \hat{V}] = 0$ , so that  $\Delta \hat{H}_S \Delta \hat{V} \geq 0$ , meaning that the commutativity relation here may result in the minimum lower bound on  $\Delta \hat{H}_S \Delta \hat{V}$ . Note that the same lowest limit may be obtained if, as in the classical case, the thermal equilibrium state of the system  $\hat{\rho}_S$  is formally the canonical Boltzmann distribution  $\hat{\rho}_S^{\text{can}}$ . Specifically, if  $\hat{\rho} = \hat{\rho}_S^{\text{can}} \otimes \text{tr}_S \hat{\rho}$ , then  $|[\hat{H}_S, \hat{V}]| = \text{tr}([\hat{H}_S, \hat{V}]\hat{\rho}) = \text{tr}([\hat{\rho}, \hat{H}_S]\hat{V}) = 0$  since  $[\hat{\rho}_S^{\text{can}}, \hat{H}_S] = 0$ , giving  $\Delta \hat{H}_S \Delta \hat{V} \geq 0$ . This is a consequence of the fact that the Boltzmann distribution is the zero-order-in-the-coupling thermal equilibrium state.

The quantity  $|[\hat{H}_S, \hat{V}]|$  is a measure of the quantum correlations between the system and the bath and Eqs. (5) dictate the lower bound of  $\Delta \hat{H}_S \Delta \hat{V}$ . This lower bound is different for each interaction since each particular form of  $\hat{V}$  imposes a different commutation relation. This last statement is precisely what allows, for example, for a connection between the theory of pointer states and quantum thermodynamics. If  $\hat{B}$  and  $\hat{S}$  denote observables of the bath and the system, respectively, the dependence on the type of interaction can be obtained by calculation. For example, if the interaction term is chosen to be  $\hat{V} = \hat{B} \otimes \hat{S}$  and the commutator  $[\hat{H}_S, \hat{V}]$  is calculated to second order in  $\hat{V}\beta$ , this leads to (see Appendix B)

$$|[\hat{H}_S, \hat{V}]| \propto \text{tr}_S \left\{ [\hat{H}_S, \hat{S}] e^{-\hat{H}_S \beta} \int_0^{\hbar\beta} d\sigma \hat{S}(-i\sigma) K(\sigma) \right\}, \quad (6)$$

where  $\hbar K(\sigma) = \langle \hat{B}(-i\sigma) \hat{B}(0) \rangle_B$  denotes the two-time correlation of the bath operators.

Note that as long as second order perturbation theory is valid, Eq. (6) holds for any  $\hat{S}$  and  $\hat{B}$  and can be straightforwardly generalized to the case of  $\hat{V} = \sum_\alpha \hat{B}_\alpha \otimes \hat{S}_\alpha$ .

A similar series-expansion analysis gives  $\hat{\rho}_S \propto e^{-\hat{H}_S \beta} [1 + \hbar^{-1} \int_0^{\hbar\beta} d\sigma \int_0^\sigma d\sigma' \hat{S}(-i\sigma) \hat{S}(-i\sigma') K(\sigma - \sigma')]$ . Thus, for non-singular  $K(\sigma)$ , the thermal equilibrium state  $\hat{\rho}_S$  formally approaches the canonical Boltzmann distribution only when  $\hbar\beta \rightarrow 0$ . Since each interaction is characterized by a given observable  $\hat{S}$  and a given two-point correlation function  $K(\sigma)$ , the general bounds in Eqs. (5, 6) predict different thermal equilibrium states for each type of interaction. Below, it is shown that the bath spectrum is also related to the lower bound and thus, Eq. (5) also allows for a clear connection to other fundamental features such as the failure of the Onsager's hypothesis in the quantum regime<sup>6,7</sup>.

### III. INFLUENCE OF THE SPECTRUM OF THE BATH: NON-MARKOVIAN CHARACTER AT THERMAL EQUILIBRIUM

Although the set of inequalities (5, 6) are general, it is not possible to infer the role that, e.g., the spectrum of the bath plays in establishing the thermodynamic bounds above. To provide a concrete expression for the lower bound in Eq. (5a), we focus on the case of  $\hat{V} = \sum_j^{\mathfrak{N}} \hat{V}_j(\hat{q}_j - \hat{q})$ , set the interaction between the bath particles to zero, i.e.,  $U_B^{i,j} = 0$  and consider the second order picture of the system-bath central force interaction, i.e.,  $\hat{V} \approx \sum_j \frac{1}{2} \mathfrak{m}_j \omega_j^2 (\hat{q}_j - \hat{q})^2$ , which yields to the well-known Ullersma-Caldeira-Leggett model<sup>29,30</sup>. After expanding  $\hat{V}$ , it is possible to redefine  $\hat{H}$  in Eq. (1) as  $\hat{H} = \hat{H}'_S + \hat{H}'_B + \hat{V}_{\text{SB}}$  with  $\hat{V}_{\text{SB}} = \hat{B} \otimes \hat{S}$ . Here  $\hat{B} = \sum_j^{\mathfrak{N}} \mathfrak{m}_j \omega_j^2 \hat{q}_j$  and  $\hat{S} = \hat{q}$ , which act in the Hilbert space of the bath and the system, respectively.  $\hbar K(\sigma) = \langle \hat{B}(-i\sigma) \hat{B}(0) \rangle_B$  given by<sup>31</sup>

$$K(\sigma) = \int \frac{d\omega}{\pi} J(\omega) \cosh\left(\frac{1}{2} \hbar\beta\omega - i\sigma\right) / \sinh\left(\frac{1}{2} \hbar\beta\omega\right), \quad (7)$$

with  $J(\omega) = \pi \sum_j \frac{1}{2} \mathfrak{m}_j \omega_j^3 \delta(\omega - \omega_j)$ , the spectral density of the bath.

The main feature of the quantum thermodynamic bound in Eq. (6) is the presence of the power spectrum of the bath  $I(\omega, T) = \hbar J(\omega) \coth(\frac{1}{2} \hbar\beta\omega)$ , which for bare Ohmic dissipation,  $J(\omega) = m\gamma\omega$ , at high temperatures,  $\hbar\beta \rightarrow 0$ , is flat ( $\omega$ -independent)  $I(\omega, T) \approx 2m\gamma k_B T$ . This is also true for higher orders in the serie expansion (see Appendix B). In this high temperature limit, and for non-singular  $K(\sigma)$ 's, the upper limit of the integral in Eq. (6) vanishes, leading to the vanishing of the commutator, even if  $[\hat{H}_S, \hat{S}] \neq 0$ . In other words, in the high temperature limit quantum correlations between the bath and the system disappear and the thermal equilibrium state is described by the canonical distribution, irrespective of the coupling strength or the functional form of the spectral density  $J(\omega)$ .

For out-of-equilibrium quantum dynamics, the low temperature condition, finite  $\hbar\beta$ , is associated with non-

Markovian dynamics<sup>31,32</sup>. Since at fixed  $T$ , this non-Markovian character can be modified by the functional form of the spectral density<sup>33</sup>, Eq. (6) makes clear that the equilibrium system properties depend on the non-Markovian character. This means that the quantum equilibrium statistical properties of a system experiencing Markovian dynamics (flat spectrum) are expected to differ from those of the same system experiencing non-Markovian dynamics (non-flat spectrum), which is in sharp contrast to the classical case (see Eq. 3). This can be clearly understood in terms of the different thermodynamic lower bounds resulting from either Markovian or non-Markovian interactions [see Eq. (6)].

We make a connection with the failure of Onsager's regression hypothesis in quantum mechanics<sup>6,7</sup>, an hypothesis which is valid under Markovian dynamics<sup>6,7</sup> and when correlations between the bath and the system are negligible at equilibrium (in general, at any time)<sup>34</sup>. Since formal Markovian dynamics can only be achieved for flat spectra<sup>33</sup> and for well behaved  $J(\omega)$  with sufficient effective weak coupling (high temperature regime, see below)<sup>35</sup>, these two conditions can be seen as a single one when formulated in terms of Eq. (6). Specifically, Markovian dynamics imply  $|\langle[\hat{H}_S, \hat{V}]\rangle| \rightarrow 0$  and, hence, the system-bath correlations vanish. This implies that Onsager's regression hypothesis, as well as the Boltzmann distribution, pertains exclusively to the classical realm. Note that this statement is general and valid as long as second order perturbation theory is valid.

To provide some insight into the magnitude and consequences of the fundamental limit derived above, an effective coupling to the bath is introduced below and two complementary regimes are analyzed. Specifically, (i) an effective strong coupling characterized by deviations from standard thermodynamics, and (ii) an effective weak coupling that is shown below to allow for the survival of entanglement even between two oscillators in thermal equilibrium at high temperatures.

### A. Effective Coupling to the Bath.

For the Ullersma-Caldeira-Leggett model, a standard calculation<sup>31</sup>, after removing a local contribution in the correlation function, yields  $K(\tau) = \frac{2m}{\hbar\beta} \frac{d}{d\tau} \sum_{l=1}^{\infty} \tilde{\gamma}(|\nu_l|) \sin(|\nu_l|\tau)$ , where  $\nu_l = l\Omega$ , with  $\Omega = 2\pi/\hbar\beta$  and  $\tilde{\gamma}(z)$  defines an effective coupling to the bath. Note that  $\tilde{\gamma}(|\nu_l|)$  contains all the information about the correlations of the bath operators and therefore defines the influence of the bath on the system at thermal equilibrium.

For the subsequent discussion we adopt the most commonly used spectral density: the regularized Drude model with a high frequency cutoff  $\omega_D$ ,  $J(\omega) = m_0\gamma\omega\omega_D^2/(\omega^2 + \omega_D^2)$ , where  $\gamma$  is the standard coupling strength constant to the thermal bath and  $\omega_D$  dictates the degree of non-Markovian dynamics. For a discussion on the experimental reconstruction of the spectral density

of open quantum systems see Ref.<sup>36</sup>. For the spectral density at hand,

$$\tilde{\gamma}(|\nu_l|) = \frac{\gamma}{1 + |l|\Omega/\omega_D}. \quad (8)$$

Below we analyze the effective strong coupling,  $\Omega/\omega_D \ll 1$ , and the effective weak coupling,  $\Omega/\omega_D \gg 1$ , regimes.

*Strong Effective Coupling Regime*—To quantify the consequences of non-flat spectra in this regime, consider as the system a harmonic oscillator of mass  $m_0$  and frequency  $\omega_0$  coupled to a thermal bath<sup>14,15</sup>. See Ref.<sup>37</sup> for a phase-space description of the dynamics and equilibrium characteristics of this model. In particular, we are interested in quantifying: (i) the generation of squeezing in the thermal equilibrium state, (ii) the deviation from the canonical partition function  $Z_{\text{can}} = \text{tr}_S e^{\hat{H}_S\beta}$  and (iii) the deviation from the canonical von-Neumann entropy  $S_{\text{can}} = \text{tr}_S [\hat{\rho}_{\text{can}} \ln(\hat{\rho}_{\text{can}})]$ .

For this case the momentum and position variances are given by<sup>14,15</sup>  $\langle p^2 \rangle = m_0^2 \omega_0^2 \langle q^2 \rangle + \Delta$ , where  $\langle q^2 \rangle = \langle q_{\text{cl}}^2 \rangle + \frac{2}{m_0\beta} \sum_{n=1}^{\infty} [\omega_0^2 + \nu_n^2 + \tilde{\gamma}(|\nu_n|)|\nu_n|]^{-1}$  and the squeezing parameter  $\Delta = -2m_0\gamma\beta^{-1} \partial \ln Z' / \partial \gamma$  with  $Z' = \frac{1}{\hbar\beta\omega} \prod_{n=1}^{\infty} |\nu_n| [\omega_0^2 + \nu_n^2 + \tilde{\gamma}(|\nu_n|)|\nu_n|]^{-1}$ . We recall that for this model, the classical theory predicts  $\langle p_{\text{cl}}^2 \rangle = m_0^2 \omega_0^2 \langle q_{\text{cl}}^2 \rangle$  and  $\langle q_{\text{cl}}^2 \rangle = k_B T / m_0 \omega_0^2$ , so that  $\Delta_{\text{cl}} = 0$ . For the effective weak coupling regime  $\Omega/\omega_D \gg 1$ , disregarding terms of the order  $\omega_0/\omega_D$  and  $\gamma/\omega_D$  gives  $\Delta \approx \pi \hbar \gamma m \omega_D / 6\Omega$ <sup>14</sup>. Thus  $\Delta$  vanishes at high temperatures, and the classical unsqueezed state is recovered. However, for the strong coupling regime  $\Omega/\omega_D \ll 1$ ,  $\Delta \approx \hbar \gamma m \ln(2\pi\omega_D/\Omega)$ <sup>14</sup>, meaning that the deviation from the canonical state translates into squeezing of the equilibrium state. This feature may be of relevance toward the generation of non-classical states, e.g., in nano-mechanical resonators.

Deviations from the canonical result are also evident in the partition function  $Z$ . Figure 1 shows the logarithm of the ratio of  $Z$  to the canonical partition function  $Z_{\text{can}}$  as a function of the dimensionless parameters  $k_B T / \hbar \omega_0$  and  $\omega_D / \omega_0$  for (from left to right)  $\gamma = 0.1\omega_0$ ,  $\gamma = 0.05\omega_0$ ,  $\gamma = 0.01\omega_0$  and  $\gamma = 0.005\omega_0$ . Deviations are observed at low temperatures and for high cutoff frequencies (i.e., in the effective strong coupling regime). In the opposite limit, regardless of the coupling parameter  $\gamma$ , both calculated partition functions show the same behavior, as expected from the discussion above. For the von Neumann entropy  $S = \text{tr}_S [\hat{\rho}_S \ln(\hat{\rho}_S)]$ , the behaviour of the ratio  $\log(S/S_{\text{can}})$  is essentially the same as the one described for the partition function ratio in Fig. 1, and is not shown here.

Since  $\tilde{\gamma}(z) = \frac{1}{m_0} \int_0^{\infty} \frac{d\omega}{\pi} \frac{J(\omega)}{\omega} \frac{2z}{\omega^2 + z^2}$ , each spectral density defines a particular functional form of the effective coupling and therefore, of the thermal equilibrium properties. Hence, as long as  $\hbar\beta$  remains finite, different spectral densities lead to different thermal equilibrium states. For the case of pair-wise central forces interactions, this can be considered as a quantum effect.

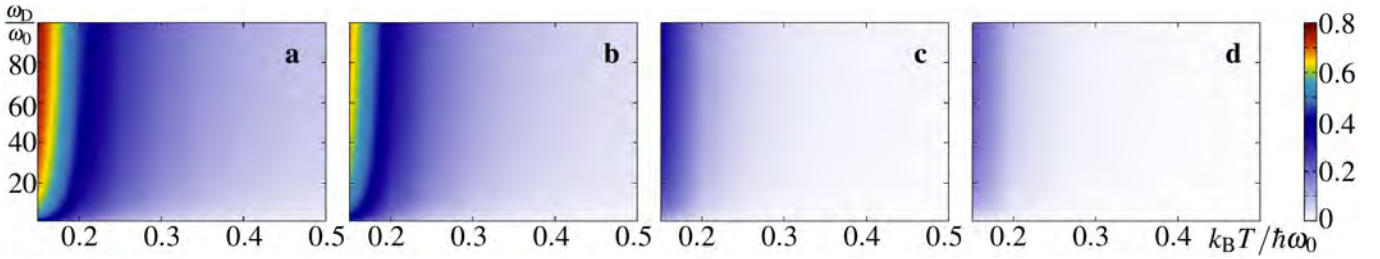


FIG. 1.  $\log(Z/Z_{\text{can}})$  for a harmonic oscillator as a function of the ratios  $k_B T / \hbar \omega_0$  and  $\omega_D / \omega_0$ . We compare the partition function for  $\gamma = 0.1\omega_0$  (a),  $\gamma = 0.05\omega_0$  (b),  $\gamma = 0.01\omega_0$  (c) and  $\gamma = 0.005\omega_0$  (d).

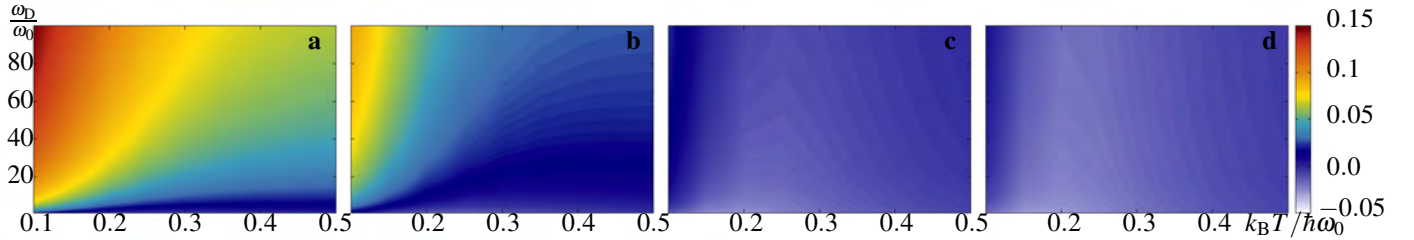


FIG. 2. Difference between the bounds quantum and classical in Eqs. (5) for a harmonic oscillator as a function of the ratio  $\omega_D / \omega_0$ . We show cuts for different  $k_B T / \hbar \omega_0$  for  $\gamma = 0.1\omega_0$  (a),  $\gamma = 0.05\omega_0$  (b),  $\gamma = 0.01\omega_0$  (c) and  $\gamma = 0.005\omega_0$  (d).

For this model, the bound in Eq. (5a) is given by  $\Delta \hat{H}_S \Delta \hat{q} = \sqrt{\frac{1}{2m_0^2} \langle \hat{p}^2 \rangle^2 + \frac{1}{2} m_0^2 \omega_0^4 \langle \hat{q}^2 \rangle^2 - \frac{1}{4} \hbar^2 \omega_0^2 \sqrt{\langle \hat{q}^2 \rangle}}$ . Contrary to the classical case where the bound is only determined by the temperature,  $(\Delta \hat{H}_S \Delta \hat{q})_{\text{cl}} = k_B T \sqrt{k_B T / m \omega^2}$ , the influence of the bath spectrum is clear in the quantum calculation of  $\Delta \hat{H}_S \Delta \hat{q}$ . Figure 2 depicts the difference  $\Delta \hat{H}_S \Delta \hat{q} - (\Delta \hat{H}_S \Delta \hat{q})_{\text{cl}}$  between the quantum and the classical bound for the  $\gamma$  kernel in Eq. (8) as a function of the ratio  $\omega_D / \omega_0$  for different values of the ratio  $k_B T / \hbar \omega_0$ . The influence of the non-Markovian character is clear.

*Weak Effective Coupling Regime*—As an example, consider the survival of entanglement at thermal equilibrium between two identical harmonic oscillators with masses  $m_0$  and frequencies  $\omega_0$  linearly coupled with coupling constant  $c_0$ . The Hamiltonian is given by

$$\hat{H} = \hat{H}_S + \sum_{j,\alpha}^{\mathfrak{R},2} \left[ \frac{\hat{p}_{j,\alpha}^2}{2m_j} + \frac{m_j \omega_j^2}{2} (\hat{q}_{j,\alpha} - \hat{q}_\alpha)^2 \right], \quad (9)$$

with  $\alpha = \{1, 2\}$  and  $\hat{H}_S = \frac{1}{2m_0} (\hat{p}_1^2 + \hat{p}_2^2) + \frac{1}{2} m_0 \omega_0^2 (\hat{q}_1^2 + \hat{q}_2^2) - c_0 q_1 q_2$ <sup>38</sup>. The introduction of independent baths for each oscillator ensures that no deviations from Boltzmann's distribution are present in the classical case. This can be verified directly from the multi-particle-system generalization of Eq. (1).

At equilibrium, the entanglement between the two harmonic oscillators can survive only when  $k_B T / \hbar \omega_0 \ll 1$ <sup>39</sup>. However, this limit only applies in the Markovian regime and  $\gamma \rightarrow 0$ . Thus, based on the discussion above, and supported by the recent observation that non-Markovian dynamics assists entanglement in the longtime limit<sup>11</sup>,

this limit needs to be refined in order to account for the non-Markovian character of the interaction and the finite value of  $\gamma$ <sup>40</sup>.

Due to the continuous-variable and Gaussian character of the systems, entanglement is quantified below by means of the logarithmic negativity  $E_N$ . This measure makes use of the positivity properties of the covariance matrix  $\sigma$  of the Gaussian state that defines the full state of the system<sup>39–41</sup> whose matrix elements are defined  $\sigma_{ij} = \frac{1}{2} \langle \hat{r}_i \hat{r}_j + \hat{r}_j \hat{r}_i \rangle - \langle \hat{r}_i \rangle \langle \hat{r}_j \rangle$ , where  $\hat{\mathbf{r}} = (\hat{q}_1, \hat{q}_2, \hat{p}_1, \hat{p}_2)$  and  $\hat{q}_1, \hat{q}_2, \hat{p}_1, \hat{p}_2$  are the position and momentum operators of the oscillators in the system of interest  $\hat{H}_S$  in Eq. (9).

$E_N$  is defined in terms of the eigenvalues  $l_i$ 's of  $-i\Sigma\sigma$ , with  $\Sigma = \begin{pmatrix} 0 & \mathbb{I}_2 \\ -\mathbb{I}_2 & 0 \end{pmatrix}$  the symplectic matrix and  $\mathbb{I}_2$  the identity matrix of dimension 2. Specifically<sup>42</sup>,

$$E_N = -\frac{1}{2} \sum_{i=1}^4 \log_2[\min(1, 2|l_i|)]. \quad (10)$$

The logarithmic negativity of the system is zero for separable states,  $\hat{\rho}_S = \sum_i p_i \hat{\rho}_1^{(i)} \otimes \hat{\rho}_2^{(i)}$ , but is unbounded from above since for continuous variable systems, the maximally entangled EPR wave-function has  $E_N \rightarrow \infty$ .

For different values of the coupling constant  $c_0$ , Fig. 3 shows the logarithmic negativity for a variety of values of the damping constant  $\gamma$  as a function of the dimensionless ratios  $k_B T / \hbar \omega_0$  and  $\omega_D / \omega_0$ . As expected, (i) the more coupled the oscillators are, the higher the temperature and the damping rate at which entanglement can survive at equilibrium, and (ii) the smaller the damping rate (the more isolated the system) is, the higher the temperature at which entanglement can be maintained. The new feature

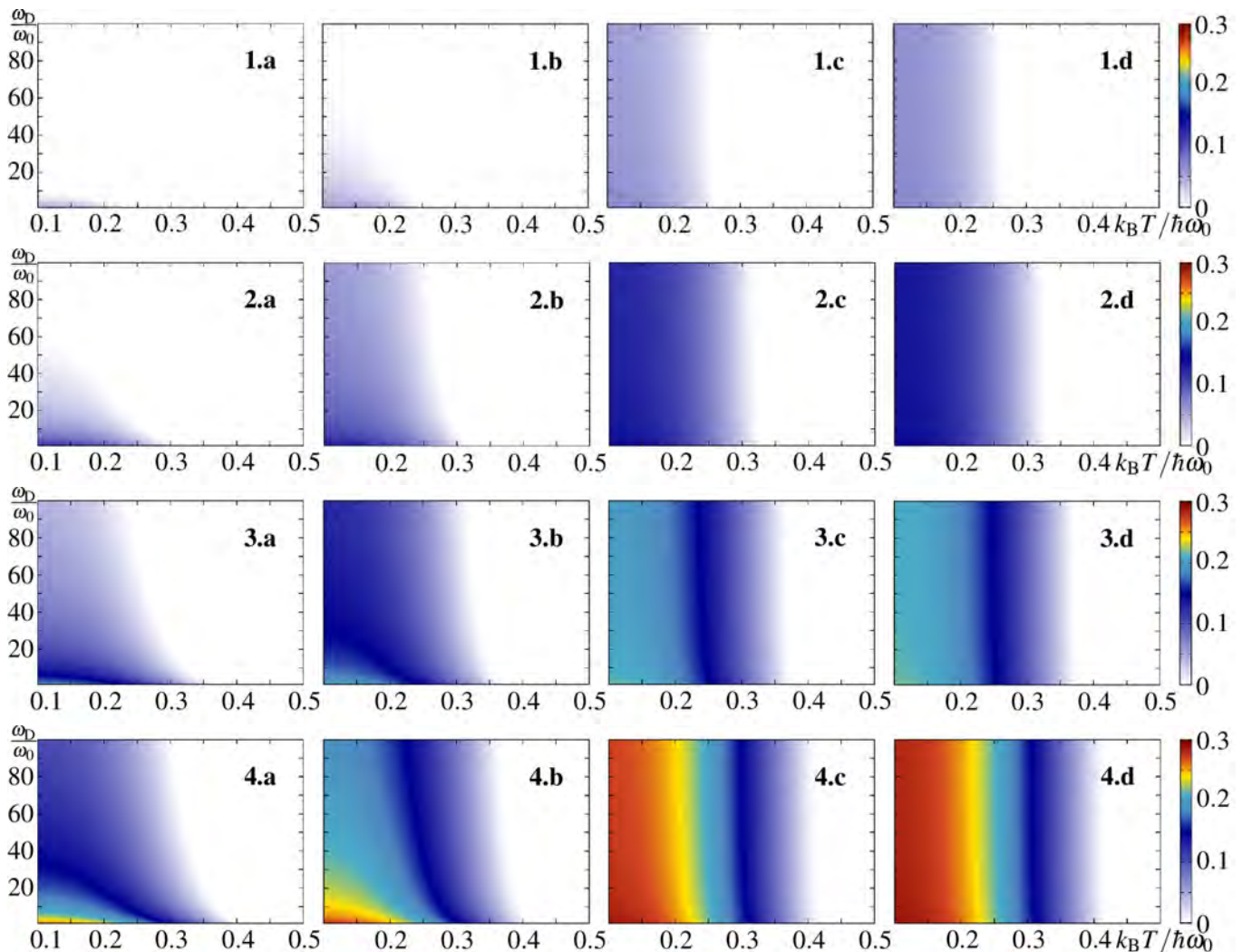


FIG. 3. Logarithmic Negativity in the presence of non-Markovian interactions for  $c_0 = 0.05m_0\omega_0^2$  first row,  $c_0 = 0.10m_0\omega_0^2$  second row,  $c_0 = 0.15m_0\omega_0^2$  third row and  $c_0 = 0.20m_0\omega_0^2$  fourth row. Parameters are  $\gamma = 0.1\omega_0$  (a),  $\gamma = 0.05\omega_0$  (b),  $\gamma = 0.01\omega_0$  (c) and  $\gamma = 0.005\omega_0$  (d) as a function of the dimensionless parameters  $k_B T / \hbar\omega_0$  and  $\omega_D / \omega_0$ .

here is that the more non-Markovian the interaction is, the higher the temperature and the damping rate at which entanglement can be maintained *at equilibrium*. This behaviour is also present at out-of-equilibrium<sup>40</sup> and can be easily interpreted in terms of the effective coupling introduced above.

#### IV. DISCUSSION.

We have shown the role of the Heisenberg uncertainty principle [Eq. (6)] in preventing quantum systems from relaxing to the Gibbs state that is dictated by the system Hamiltonian only. The Gibbs state is only recovered in the classical-high  $T$  limit ( $\hbar\beta \rightarrow 0$ ). The implications at low- $T$  for quantum thermodynamics are crucial, such as the failure for the Onsager hypothesis, or the difficulty in defining the specific heat<sup>43</sup>, and the temperature

definition<sup>44</sup>. Specifically, the high temperature regime defined by  $\hbar\beta \rightarrow 0$ , modified by an appropriate effective coupling, emerges as the main condition for the vanishing of these deviations.

For the wide class of classical systems discussed above (for which the thermal equilibrium state exactly corresponds to the Boltzmann distribution), the dependence of thermal equilibrium state on the spectral density is clearly a pure quantum effect. This feature can be explored as a quantum resources, e.g., in the one-photon phase control of biochemical and biophysical systems<sup>13,24,25</sup> or in understanding the coherent extent of excitation with incoherent light in biological systems<sup>25,45</sup>.

Although the examples presented are specific to the second order approximation of the interaction potential, the general picture provided here remains valid, albeit more involved, in non-linear cases and with non-Gaussian statistics. The results presented here clarify the role of

non-Markovian dynamics and its relevance at thermal equilibrium, and provide physical insights into how non-Markovian interactions protect quantum features such as entanglement. They may shed light on the role of non-Markovian dynamics in the derivation of fundamental limits in areas such as quantum metrology<sup>12</sup>, quantum speed limits<sup>8</sup> and cooling of nanomechanical resonators<sup>41</sup>.

Finally, if  $\mathcal{D}_S$  denotes the dimension of the Hilbert space of the system and  $\mathcal{D}_B^{\text{eff}}$  denotes an abstract effective dimension of the Hilbert space of the bath, it is interesting to note that in the context of canonical typicality<sup>46,47</sup>, the trace distance  $\langle D(\hat{\rho}_S, \hat{\rho}^{\text{can}}) \rangle = (1/2)\text{tr}\sqrt{(\hat{\rho}_S - \hat{\rho}^{\text{can}})^\dagger(\hat{\rho}_S - \hat{\rho}^{\text{can}})}$  between the general equilibrium state  $\hat{\rho}_S$  and the canonical distribution  $\hat{\rho}^{\text{can}}$  is bounded from above by the ratio  $\frac{1}{2}\sqrt{\mathcal{D}_S/\mathcal{D}_B^{\text{eff}}}$ . Formally, when the spec-

tral density is introduced, the limit  $\mathcal{D}_B \rightarrow \infty$  is implicit. Thus, if  $\mathcal{D}_B^{\text{eff}} = \mathcal{D}_B$ , contrary to the results presented here, no deviations from the canonical state are expected. However, based on the results above, the trace distance is given here by  $D(\hat{\rho}_S, \hat{\rho}^{\text{can}}) \propto \hbar^{-1}\text{tr}_S \int_0^{\hbar\beta} d\sigma \int_0^\sigma d\sigma' \hat{S}(-i\sigma)\hat{S}(-i\sigma')K(\sigma - \sigma')$ . This situation suggests that the abstract effective dimension  $\mathcal{D}_B^{\text{eff}}$  must be a function of the ratio  $\hbar/k_B T$ , the power noise of the environment  $I(\omega)$  and of the observable that couples the system to the environment. Deriving the specific functional relationship  $\mathcal{D}_B^{\text{eff}}[\hbar\beta, I(\omega)]$  would allow for bridging a gap between information theoretical and pure thermodynamic results. Work along this line will be reported soon.

### Appendix A: Reduced Classical Thermal State (Eq. 3)

Consider the general expression for the physical situation described in Sec. IA

$$\rho(\mathbf{p}, \mathbf{q}) = \frac{1}{Z} \exp \left\{ - \left[ H_S(\mathbf{p}, \mathbf{q}) + \sum_j^{\mathfrak{N}} \left[ \frac{1}{2m_j} \mathbf{p}_j \cdot \mathbf{p}_j + \sum_i^{\mathfrak{N}} U_{i,j}(\mathbf{q}_i - \mathbf{q}_j) + \mathcal{V}_j(\mathbf{q}_j - \mathbf{q}) \right] \right] \beta \right\}. \quad (\text{A1})$$

The reduced thermal state in Eq. (2) reads

$$\rho_S(\mathbf{p}, \mathbf{q}) = \frac{1}{Z} \exp[-H_S(\mathbf{p}, \mathbf{q})\beta] \int_{-\infty}^{\infty} \prod_j^{\mathfrak{N}} d\mathbf{p}_j d\mathbf{q}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \frac{1}{2m_j} \mathbf{p}_j \cdot \mathbf{p}_j + \sum_i^{\mathfrak{N}} U_{i,j}(\mathbf{q}_i - \mathbf{q}_j) + \mathcal{V}_j(\mathbf{q}_j - \mathbf{q}) \right] \beta \right\}, \quad (\text{A2})$$

with

$$Z = \int_{-\infty}^{\infty} d\mathbf{p} d\mathbf{q} \exp[-H_S(\mathbf{p}, \mathbf{q})\beta] \int_{-\infty}^{\infty} \prod_j^{\mathfrak{N}} d\mathbf{p}_j d\mathbf{q}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \frac{1}{2m_j} \mathbf{p}_j \cdot \mathbf{p}_j + \sum_i^{\mathfrak{N}} U_{i,j}(\mathbf{q}_i - \mathbf{q}_j) + \mathcal{V}_j(\mathbf{q}_j - \mathbf{q}) \right] \beta \right\}. \quad (\text{A3})$$

After defining  $\mathbf{q}_i - \mathbf{q} = \mathbf{z}_i$ , the integral over  $\prod_j^{\mathfrak{N}} d\mathbf{p}_j d\mathbf{q}_j$  becomes

$$\int_{-\infty}^{\infty} \prod_j^{\mathfrak{N}} d\mathbf{p}_j d\mathbf{z}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \frac{1}{2m_j} \mathbf{p}_j \cdot \mathbf{p}_j + \sum_i^{\mathfrak{N}} U_{i,j}(\mathbf{z}_i - \mathbf{z}_j) + \mathcal{V}_j(\mathbf{z}_j) \right] \beta \right\}. \quad (\text{A4})$$

So that,

$$\rho_S(\mathbf{p}, \mathbf{q}) = \frac{\exp[-H_S(\mathbf{p}, \mathbf{q})\beta] \int_{-\infty}^{\infty} \prod_j^{\mathfrak{N}} d\mathbf{p}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \frac{1}{2m_j} \mathbf{p}_j \cdot \mathbf{p}_j \right] \beta \right\} \prod_j^{\mathfrak{N}} d\mathbf{z}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \sum_i^{\mathfrak{N}} U_{i,j}(\mathbf{z}_i - \mathbf{z}_j) + \mathcal{V}_j(\mathbf{z}_j) \right] \beta \right\}}{\int_{-\infty}^{\infty} d\mathbf{p} d\mathbf{q} \exp[-H_S(\mathbf{p}, \mathbf{q})\beta] \int_{-\infty}^{\infty} \prod_j^{\mathfrak{N}} d\mathbf{p}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \frac{1}{2m_j} \mathbf{p}_j \cdot \mathbf{p}_j \right] \beta \right\} \prod_j^{\mathfrak{N}} d\mathbf{z}_j \exp \left\{ - \sum_j^{\mathfrak{N}} \left[ \sum_i^{\mathfrak{N}} U_{i,j}(\mathbf{z}_i - \mathbf{z}_j) + \mathcal{V}_j(\mathbf{z}_j) \right] \beta \right\}}.$$

Thus,

$$\rho_S(\mathbf{p}, \mathbf{q}) = \frac{\exp[-H_S(\mathbf{p}, \mathbf{q})\beta]}{\int_{-\infty}^{\infty} d\mathbf{p} d\mathbf{q} \exp[-H_S(\mathbf{p}, \mathbf{q})\beta]} = Z_S^{-1} \exp[-H_S(\mathbf{p}, \mathbf{q})\beta], \quad (\text{A5})$$

which corresponds to the canonical classical thermal state.

### Appendix B: Commutator $[\hat{H}_S, \hat{V}]$ at fourth order

The equilibrium state at third order is given by

$$\hat{\rho} = e^{-\beta \hat{H}_S} \left[ 1 - \int_0^{\hbar\beta} d\sigma V_{\text{SB}}(-i\hbar\sigma) + \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \hat{V}_{\text{SB}}(-i\hbar\sigma) \hat{V}_{\text{SB}}(-i\hbar\theta) - \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \int_0^{\theta} d\eta \hat{V}_{\text{SB}}(-i\hbar\sigma) \hat{V}_{\text{SB}}(-i\hbar\theta) \hat{V}_{\text{SB}}(-i\hbar\eta) \right], \quad (\text{B1})$$

therefore

$$\left| \langle [\hat{H}_S, \hat{V}_{\text{SB}}] \rangle \right| = \left| \text{tr} \left( [\hat{H}_S, \hat{S}] \otimes \hat{B} e^{-\beta \hat{H}_S} \left[ 1 - \int_0^{\hbar\beta} d\sigma \hat{S} \otimes \hat{B}(-i\hbar\sigma) + \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \hat{S} \otimes \hat{B}(-i\hbar\sigma) \hat{S} \otimes \hat{B}(-i\hbar\theta) - \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \int_0^{\theta} d\eta \hat{S} \otimes \hat{B}(-i\hbar\sigma) \hat{S} \otimes \hat{B}(-i\hbar\theta) \hat{S} \otimes \hat{B}(-i\hbar\eta) \right] \right) \right|, \quad (\text{B2})$$

the first term that corresponds to  $\text{tr}([\hat{H}_S, \hat{S}] \otimes \hat{B} e^{-\beta \hat{H}_S}) = \text{tr}([e^{-\beta \hat{H}_S}, \hat{H}_S] \hat{S} \otimes \hat{B}) = 0$ . Therefore, after tracing out over the bath, we get that the lower bound in Eq. (6) is given by

$$\left| \langle [\hat{H}_S, \hat{V}] \rangle \right| \propto_{\text{trs}} \left\{ [\hat{H}_S, \hat{S}] e^{-\hat{H}_S \beta} \left( - \int_0^{\hbar\beta} d\sigma \hat{S}(-i\hbar\sigma) K(\sigma) + \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \hat{S}(-i\hbar\sigma) \hat{S}(-i\hbar\theta) L(\sigma, \theta) - \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \int_0^{\theta} d\eta \hat{S}(-i\hbar\sigma) \hat{S}(-i\hbar\theta) \hat{S}(-i\hbar\eta) M(\sigma, \theta, \eta) \right) \right\} \quad (\text{B3})$$

$$\left| \langle [\hat{H}_S, \hat{V}] \rangle \right| \propto_{\text{trs}} \left\{ [\hat{H}_S, \hat{S}] e^{-\hat{H}_S \beta} \left( \int_0^{\hbar\beta} d\sigma \hat{S}(-i\hbar\sigma) K(\sigma) + \int_0^{\hbar\beta} d\sigma \int_0^{\sigma} d\theta \int_0^{\theta} d\eta \hat{S}(-i\hbar\sigma) \hat{S}(-i\hbar\theta) \hat{S}(-i\hbar\eta) M(\sigma, \theta, \eta) \right) \right\} \quad (\text{B4})$$

where  $\hbar K(\sigma) = \langle \hat{B}(-i\sigma) \hat{B}(0) \rangle_{\text{B}}$  denotes the two-time correlation of the bath operators given by<sup>31</sup> and  $\hbar L(\sigma, \theta) = \langle \hat{B}(-i\sigma) \hat{B}(-i\theta) \hat{B}(0) \rangle_{\text{B}} = 0$  and  $\hbar M(\sigma, \theta, \eta) = \langle \hat{B}(-i\sigma) \hat{B}(-i\theta) \hat{B}(-i\eta) \hat{B}(0) \rangle_{\text{B}}$  denotes the three and four-time correlation of the bath operators, respectively.

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