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# Non-Markovian quantum heat statistics with the reaction coordinate mapping

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# Quantum Science and Technology



## PAPER

# Non-Markovian quantum heat statistics with the reaction coordinate mapping

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## Abstract

The definition of heat in quantum mechanics is ambiguous. Complications arise in particular when the coupling between a quantum system and a thermal environment is non-negligible, as the boundary between the two becomes blurred, making the distinction between system and environment difficult to draw. The reaction coordinate (RC) mapping can be used in such regimes to redraw the boundary between the system and environment. In this paper we combine the RC technique with a two-point measurement protocol to compare two different definitions of heat: energetic changes with respect to the full environment Hamiltonian (prior to the mapping), and energetic changes with respect to the residual environment Hamiltonian (after the mapping). We find that the latter definition displays behaviour more expected of a heat bath in the highly non-Markovian regime considered.

## 1. Introduction

In classical thermodynamics a distinction is drawn between two classes of observable quantities: A *state function* is a physical quantity, such as temperature or internal energy, that is well defined for each point in the system's phase space. In contrast, a *path function* is a physical quantity that depends on the specific path taken between two points in this phase space, for example heat and work. In a quantum mechanical setting, state functions can generally be represented as the trace of some Hermitian operator with the state of the system, or deduced from the state itself. However, path functions, such as work, do not have a clear analogue [1]. Notably, this has generated significant debate within the quantum thermodynamics community regarding the appropriate definition of path functions like heat and work in quantum systems [2–10].

In regimes where system–environment interactions are weak, and are therefore accurately captured by a Born–Markov master equation, heat can quite naturally be identified as the energy irreversibly emitted into (or absorbed from) the environment [3]. However, when the interaction energy becomes comparable to the internal energy of the system, there is no longer a clear partition between system and environment degrees of freedom [11, 12] as the two become strongly correlated, potentially exchanging energy and information in a non-Markovian (or reversible) fashion. This leads to further ambiguity as to how one should appropriately apportion the changes in internal and interaction energies into heat and work [3, 7, 9, 13, 14].

In this paper, we investigate the role that different system–environment partitions play on quantum heat statistics in the non-Markovian regime. To do so, we employ the reaction coordinate (RC) mapping of the spin-boson model [15]. Here, a collective coordinate of the full environment (FE) is incorporated into an enlarged effective system Hamiltonian (the extended system (ES)), with the remaining environmental degrees of freedom included as a weakly-coupled residual environment (RE), which may be treated perturbatively using the RC master equation (RCME). The resulting description has proven useful in studying the dynamics [16–18] and thermodynamics [13, 19–21] of quantum systems in regimes of strong

and non-Markovian system-environment interactions. We extend the RC formalism to the two-point measurement protocol (TPMP) [2, 22, 23] to derive a heat-counting RCME (HC-RCME). This generalised master equation allows us to calculate the characteristic function (CF) that generates the stochastic heat probability distribution for strong system-environment interactions, which we also successfully benchmark in some exactly solvable cases.

Central to this work are the two possible definitions of heat provided by the RC formalism. Heat may naturally be defined as changes in measurements of the FE Hamiltonian prior to the mapping, or changes in measurements of the RE Hamiltonian after the mapping. We find that these two possible definitions of heat demonstrate qualitative and quantitative differences in the first two moments of their probability distributions. We also find corresponding differences in the change in ergotropy [24, 25] and von Neumann entropy of the original system and the ES over the process considered. Together, these results suggest that for non-Markovian systems, defining heat as changes in the energy of the RE is more in line with the classical definition of heat in thermodynamics.

The paper is arranged as follows. In section 2.1 we cover the TPMP, outlining how the CF of heat transfer can be written as the trace of a generalised density operator. In section 2.2 we show how the RC mapping (RCM) can be used in conjunction with the TPMP to probe quantum heat statistics in strong coupling and non-Markovian regimes. We do this by deriving the HC-RCME which describes the evolution of the generalised density operator. We also introduce the two possible definitions of heat stated above. In section 3.1 we analyse the CFs associated with the two definitions of heat, before looking at the first two moments of the corresponding probability distributions in section 3.2. Differences in the first moment of these two definitions of heat motivate studying the transfer of ordered energy between the original system and FE, and between the ES and RE, which we cover in section 3.3. We summarise and conclude in section 4.

## 2. Counting statistics in the strong coupling regime

Path functions do not have a clear and unambiguous definition in quantum systems; notably there are no unique Hermitian operators, and thus observables, associated with them. In order to define the statistics of path functions we consider performing projective measurements. The TPMP [2, 3, 5] is a framework which can be used to calculate the full counting statistics of the path function we want to define. However, within the TPMP there is ambiguity in choosing what basis we perform these projective measurements onto. In the strong coupling and non-Markovian regime that we consider, choosing a particular basis to perform the projective measurements onto is not always straightforward.

### 2.1. The two-point measurement protocol

We define changes in heat through the TPMP, a brief overview of which can be seen schematically in figure 1 and is as follows: An open quantum system  $S$  is coupled to an environment  $E$ , which together are governed by the Hamiltonian  $\hat{H}$ , evolving unitarily according to  $\hat{U}(t) = \exp[-i\hat{H}t]$ . We first prepare a product state  $\hat{\rho}(0) = \hat{\rho}_S(0) \otimes \hat{\rho}_{FE}$ , where  $\hat{\rho}_{FE}$  is a Gibbs state of the full environment and  $\hat{\rho}_S(0)$  is an arbitrary initial state of the open system. Next, a projective measurement onto the eigenbasis of an observable  $\hat{M}$  is applied to the composite system, which is then allowed to evolve unitarily for time  $t$ , before performing a second projective measurement onto the eigenbasis of  $\hat{M}$ . We define the difference of these two measurement outcomes as  $M$ , a stochastic quantity. By repeating the protocol many times we build up a probability distribution  $P(M, t)$ . Our choice of operator  $\hat{M}$  leads to the definition of heat that we use.

While the probability distribution  $P(M, t)$  contains the statistics that we are interested in, it is more convenient to work with its Fourier transform, the characteristic function (CF),

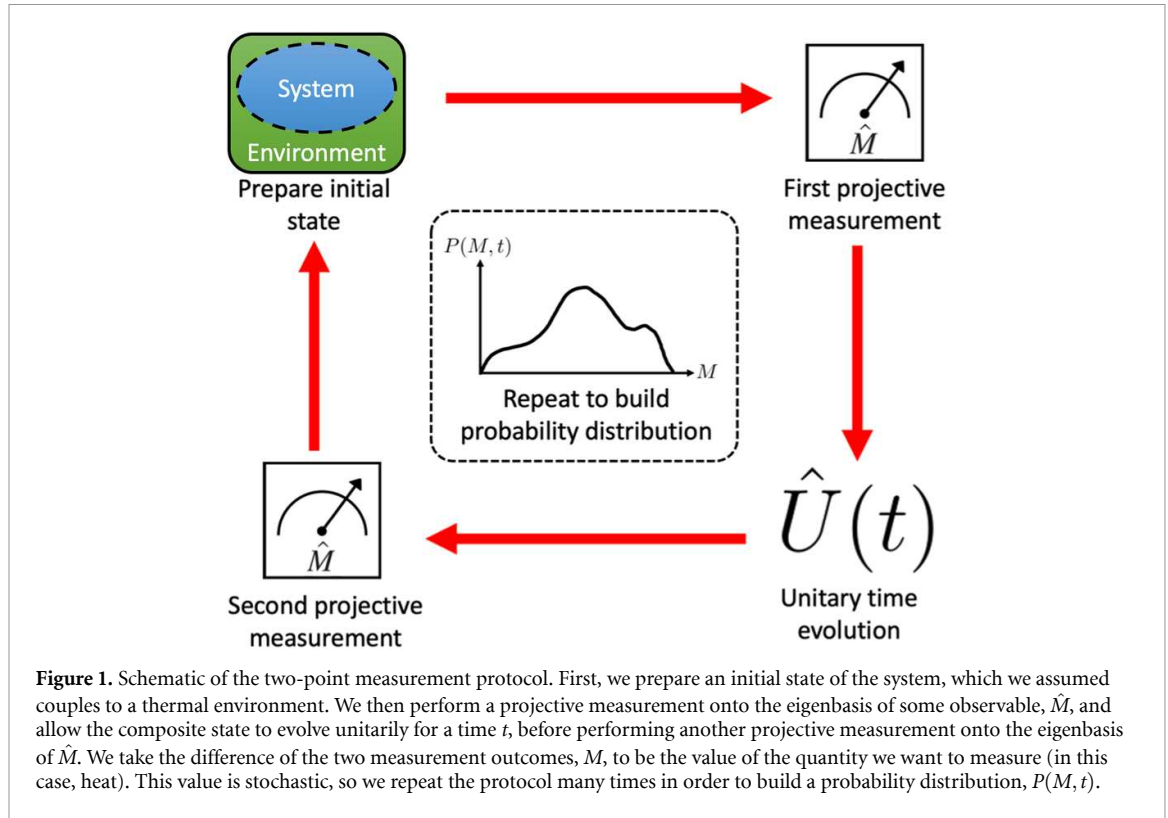
$$\Phi(\chi, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dM P(M, t) e^{iM\chi}, \quad (1)$$

a complex valued function of the counting parameter  $\chi$ , which is the conjugate parameter to the stochastic quantity  $M$ . Following Esposito *et al* [2], we can write the CF as

$$\Phi(\chi, t) = \text{Tr}[\hat{\rho}(\chi, t)], \quad (2)$$

where we have defined the generalised density operator

$$\hat{\rho}(\chi, t) = \hat{U}_{\frac{\chi}{2}}(t) e^{-i\frac{\chi}{2}\hat{M}} \hat{\rho}_0 e^{i\frac{\chi}{2}\hat{M}} \hat{U}_{-\frac{\chi}{2}}^\dagger(t), \quad (3)$$



and have made use of a generalised time evolution operator

$$\hat{U}_\chi(t) = e^{i\chi\hat{M}}\hat{U}(t)e^{-i\chi\hat{M}}. \quad (4)$$

In equation (3),  $\bar{\rho}_0 = \sum_{m_0} |m_0\rangle\langle m_0| \hat{\rho}(0) |m_0\rangle\langle m_0|$  is the initial state of the composite system after being averaged over the initial measurement outcomes of  $\hat{M} = \sum_{m_0} m_0 |m_0\rangle\langle m_0|$ . Applying a partial trace over the FE degrees of freedom in equation (2) gives

$$\Phi(\chi, t) = \text{Tr}_S[\hat{\rho}_S(\chi, t)], \quad (5)$$

where we have defined the reduced generalised density operator for the system as  $\hat{\rho}_S(\chi, t) = \text{Tr}_{\text{FE}}[\hat{\rho}(\chi, t)]$ . By deriving an equation of motion for  $\hat{\rho}_S(\chi, t)$  and taking the trace, we can generate the full probability distribution for the stochastic quantity  $M$  by inverting the Fourier transform.

As shown in [5], in the limit of weak system–environment interactions, we are able to derive a Born–Markov master equation describing the evolution of the reduced generalised density operator,

$$\frac{d}{dt}\hat{\rho}_S(\chi, t) = \mathcal{L}_S(\chi)\hat{\rho}_S(\chi, t), \quad (6)$$

where  $\mathcal{L}_S(\chi)$  is the generalised ( $\chi$ -dressed) Liouville superoperator [26] that describes both the coherent dynamics and the effect of the environment on  $\hat{\rho}_S(\chi, t)$  up to second order in the coupling strength. Solving this master equation with an appropriate initial condition,  $\hat{\rho}_S(\chi, 0)$ , yields the CF.

## 2.2. Reaction coordinate mapping

In this paper we are interested in studying heat exchange beyond the Born–Markov limit, and therefore must move past the generalised Born–Markov equation given in equation (6). We consider the spin-boson model,

$$\hat{H} = \frac{\epsilon}{2}\hat{\sigma}_z + \frac{\Delta}{2}\hat{\sigma}_x + \hat{\sigma}_z \otimes \sum_k f_k (\hat{c}_k^\dagger + \hat{c}_k) + \sum_k \nu_k \hat{c}_k^\dagger \hat{c}_k, \quad (7)$$

where the first two terms make up the system Hamiltonian  $\hat{H}_S$ , the third term is the interaction  $\hat{V}$ , and the fourth is the self Hamiltonian of the FE,  $\hat{H}_{\text{FE}}$ . In equation (7),  $\epsilon$  is the energy gap between basis states  $|e\rangle$  and  $|g\rangle$  of a two-level system (TLS), and  $\Delta$  is the tunneling between these states. The  $k^{\text{th}}$ -mode of the bosonic environment has creation (annihilation) operators  $\hat{c}_k^\dagger$  ( $\hat{c}_k$ ), with energy  $\nu_k$ , and couples to the TLS with

strength  $f_k$ . The TLS-FE coupling can be described by the spectral density function,

$$J_{\text{FE}}(\nu) = \sum_k |f_k|^2 \delta(\nu - \nu_k).$$

There has been significant effort in developing numerical methods to describe both the dynamics and thermodynamics of the spin-boson model. Notable examples include those based on discrete-time path integrals [27, 28] and their expression as tensor networks [29, 30], hierarchical equations of motion [31–33], and chain mapping methods [34, 35]. While such techniques can be used to obtain numerically exact results in many regimes, they are computationally demanding and can be challenging to interpret. In contrast, nonperturbative master equation techniques, such as the polaron theory [36] and its variational extensions [37, 38] are intuitive and computationally cheap, though often at the expense of restricted applicability. The nonperturbative method we use in this work is the RCM [13, 15, 19, 20]. To perform this mapping, a collective coordinate (the RC) is first extracted from the FE. The TLS, RC and their interaction are then incorporated into an ES, and the remaining environment degrees of freedom are collected into the RE, which couples to the ES through the RC. For a more detailed discussion of the RCM we refer the reader to references [15, 20]. Upon applying the RCM to the Hamiltonian in equation (7) with the unitary operator  $\hat{\mathcal{R}}$ , we obtain the Hamiltonian,

$$\hat{H}_{\mathcal{R}} = \hat{\mathcal{R}}\hat{H}\hat{\mathcal{R}}^\dagger = \hat{H}_{\text{ES}} + \hat{H}_{\text{I}} + \hat{H}_{\text{RE}}, \quad (8)$$

$$\hat{H}_{\text{ES}} = \frac{\epsilon}{2}\hat{\sigma}_z + \frac{\Delta}{2}\hat{\sigma}_x + \Omega\hat{a}^\dagger\hat{a} + \lambda\hat{\sigma}_z(\hat{a}^\dagger + \hat{a}), \quad (9)$$

$$\hat{H}_{\text{I}} = (\hat{a}^\dagger + \hat{a}) \sum_k g_k (\hat{b}_k^\dagger + \hat{b}_k) + (\hat{a}^\dagger + \hat{a})^2 \sum_k \frac{g_k^2}{\omega_k}, \quad (10)$$

$$\hat{H}_{\text{RE}} = \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k. \quad (11)$$

Here, equation (9) is the ES Hamiltonian, where the RC has frequency  $\Omega$ , and creation (annihilation) operators  $\hat{a}^\dagger$  ( $\hat{a}$ ), and is coupled to the TLS with strength  $\lambda$ . The self Hamiltonian of the RC is given by  $\hat{H}_{\text{RC}} = \Omega\hat{a}^\dagger\hat{a}$ . Equation (10) defines the interaction between the RC and the RE with a coupling strength  $g_k$  between the RC and  $k^{\text{th}}$  mode of the RE. The interaction also includes a counter term which ensures that the Hamiltonian is bounded from below. Equation (11) gives the RE Hamiltonian with modes of frequency  $\omega_k$ , and creation (annihilation) operators  $\hat{b}_k^\dagger$  ( $\hat{b}_k$ ).

We choose an underdamped Drude–Lorentz spectral density for the FE, describing a peak of width  $\Gamma$ , centered at  $\omega_0$ , with coupling strength  $\alpha$ ,

$$J_{\text{FE}}(\omega) = \frac{\alpha\Gamma\omega_0^2\omega}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2}. \quad (12)$$

Upon applying the RCM, the spectral density describing the ES-RE interaction becomes Ohmic,

$$J_{\text{RE}}(\omega) = \sum_k |g_k|^2 \delta(\omega - \omega_k) = \gamma\omega, \quad (13)$$

with the coupling strength between the ES and RE being  $\gamma = \Gamma/\omega_0$ . A hard cutoff of  $\omega_{\text{cut}} = 10\omega_0$ , is added to both spectral densities in later numerical simulations. This choice of cutoff ensures convergence of all the results that we show.

For sufficiently weak coupling to the RE (small  $\gamma$ ) we can derive a Born–Markov master equation for the ES state  $\hat{\rho}_{\text{ES}}(t)$ . After solving this RCME, we can trace out the RC degrees of freedom to obtain the reduced state of the TLS:  $\hat{\rho}_{\text{S}}(t) = \text{Tr}_{\text{RC}}[\hat{\rho}_{\text{ES}}(t)]$ . The RCME accurately captures both non-Markovian and strong coupling effects between the system and FE, since  $\gamma$  is independent of the TLS-FE coupling strength  $\alpha$ . To confirm this, we present a comparison of an exactly solvable limit of the spin-boson model (the independent boson model (IBM) for which  $\Delta = 0$  [26]) with the RCME in appendix A, demonstrating excellent agreement across all timescales tested within the non-Markovian regime.

### 2.3. Heat counting in the RC formalism

We now apply the RCM to heat counting statistics. Let us begin with the CF to count energetic changes in the FE, given by

$$\Phi_{\text{F}}^{\text{FC}}(\chi, t) = \text{Tr} \left[ e^{i\chi\hat{H}_{\text{FE}}}\hat{U}(t) e^{-i\chi\hat{H}_{\text{FE}}}\bar{\rho}_0\hat{U}^\dagger(t) \right], \quad (14)$$

where we assume projective measurements onto the FE Hamiltonian  $\hat{M} = \hat{H}_{\text{FE}}$  [39]. We make the assumption that the initial state is uncorrelated,  $\hat{\rho}(0) = \hat{\rho}_{\text{S}}(0) \otimes \hat{\rho}_{\text{FE}}$ , with  $\hat{\rho}_{\text{FE}}$  the Gibbs state

$\hat{\rho}_{\text{FE}} = \exp[-\beta\hat{H}_{\text{FE}}]/\mathcal{Z}_{\text{FE}}$ , where  $\mathcal{Z}_{\text{FE}} = \text{Tr}[\exp[-\beta\hat{H}_{\text{FE}}]]$  is the partition function and  $\beta$  is the inverse temperature of the FE. By resolving the identity as  $\hat{\mathcal{I}} = \hat{\mathcal{R}}^\dagger \hat{\mathcal{R}}$  we can rewrite the CF in equation (14) as

$$\Phi_{\text{F}}^{\text{rc}}(\chi, t) = \text{Tr} \left[ e^{i\chi \hat{\mathcal{R}}^\dagger \hat{H}_{\text{FE}} \hat{\mathcal{R}}} \hat{U}_{\hat{\mathcal{R}}}(t) e^{-i\chi \hat{\mathcal{R}}^\dagger \hat{H}_{\text{FE}} \hat{\mathcal{R}}} \hat{\mathcal{R}}^\dagger \hat{\rho}(0) \hat{\mathcal{R}} \hat{U}_{\hat{\mathcal{R}}}^\dagger(t) \right]. \quad (15)$$

We have also defined the time evolution operator in the RC frame as

$$\hat{U}_{\mathcal{R}}(t) = \hat{\mathcal{R}} \exp[-i\hat{H}t] \hat{\mathcal{R}}^\dagger = \exp[-i\hat{H}_{\mathcal{R}}t], \quad (16)$$

where  $\hat{H}_{\mathcal{R}}$  is the mapped Hamiltonian given in equation (8). We assume that the RCM is performed such that the interaction between the RC and RE is weak. Performing the RCM and making the weak coupling approximation gives us

$$\Phi_{\text{F}}^{\text{rc}}(\chi, t) \approx \text{Tr} \left[ e^{i\chi(\hat{H}_{\text{RC}} + \hat{H}_{\text{RE}})} \hat{U}_{\hat{\mathcal{R}}}(t) e^{-i\chi(\hat{H}_{\text{RC}} + \hat{H}_{\text{RE}})} \hat{\rho}_{\text{S}}(0) \otimes \hat{\rho}_{\text{RC}} \otimes \hat{\rho}_{\text{RE}} \hat{U}_{\hat{\mathcal{R}}}^\dagger(t) \right], \quad (17)$$

valid provided that the product  $\gamma\chi$  does not become too large. Here we have approximated the action of the RCM on the FE thermal state as

$$\hat{\mathcal{R}}^\dagger \hat{\rho}_{\text{FE}} \hat{\mathcal{R}} \approx \hat{\rho}_{\text{RC}} \otimes \hat{\rho}_{\text{RE}}, \quad (18)$$

where  $\hat{\rho}_{\text{RC}}$  and  $\hat{\rho}_{\text{RE}}$  are Gibbs states of the RC and RE, respectively. Using the cyclic property of the trace we rewrite the CF as

$$\Phi_{\text{F}}^{\text{rc}} = \text{Tr} \left[ e^{i\chi \hat{H}_{\text{RC}}} \hat{\rho}(\chi, t) \right], \quad (19)$$

where we have defined the generalised density operator as

$$\hat{\rho}(\chi, t) = e^{i\frac{\chi}{2} \hat{H}_{\text{RE}}} \hat{U}_{\hat{\mathcal{R}}}(t) e^{-i\chi(\hat{H}_{\text{RC}} + \hat{H}_{\text{RE}})} \hat{\rho}_{\text{S}}(0) \otimes \hat{\rho}_{\text{RC}} \otimes \hat{\rho}_{\text{RE}} \hat{U}_{\hat{\mathcal{R}}}^\dagger(t) e^{i\frac{\chi}{2} \hat{H}_{\text{RE}}}. \quad (20)$$

By taking the time derivative of this generalised density operator and moving into the interaction picture with respect to  $\hat{H}_{\text{ES}} + \hat{H}_{\text{RE}}$  we find an equation of motion which resembles the Liouville-von Neumann equation [26]

$$\frac{d}{dt} \tilde{\rho}(\chi, t) = -i(\tilde{H}_{\text{I}}(\chi, t) \tilde{\rho}(\chi, t) - \tilde{\rho}(\chi, t) \tilde{H}_{\text{I}}(-\chi, t)), \quad (21)$$

where

$$\tilde{H}_{\text{I}}(\chi, t) = e^{i\frac{\chi}{2} \hat{H}_{\text{RE}}} e^{i(\hat{H}_{\text{ES}} + \hat{H}_{\text{RE}})t} \hat{H}_{\text{I}} e^{-i(\hat{H}_{\text{ES}} + \hat{H}_{\text{RE}})t} e^{-i\frac{\chi}{2} \hat{H}_{\text{RE}}}. \quad (22)$$

We use this equation as a basis to derive a master equation which treats the Hamiltonian of the ES exactly and the effect of the RE on the ES up to second order in  $\hat{H}_{\text{I}}$ , leading us to the HC-RCME

$$\frac{d}{dt} \hat{\rho}_{\text{ES}}(\chi, t) = \mathcal{L}_{\text{ES}}(\chi) [\hat{\rho}_{\text{ES}}(\chi, t)]. \quad (23)$$

Details of the form of  $\mathcal{L}_{\text{ES}}(\chi)$  can be found in appendix B. By taking the partial trace over the RE degrees of freedom, the CF to count energetic changes in the FE Hamiltonian is given by

$$\Phi_{\text{F}}^{\text{rc}} = \text{Tr}_{\text{ES}} \left[ e^{i\chi \hat{H}_{\text{RC}}} \hat{\rho}_{\text{ES}}(\chi, t) \right], \quad (24)$$

where  $\hat{\rho}_{\text{ES}}(\chi, t)$  is found by solving the HC-RCME.

Notice, however, that the RCM gives us two possible environment Hamiltonians to perform projective measurements onto within the TPMP. There is the environment prior to the mapping, which includes all bosonic degrees of freedom (the FE), and there is the environment after the mapping (the RE). Performing the projective measurements onto these two Hamiltonians results in two different CFs and thus definitions of heat, which we can compare. When counting only on the RE we take  $\hat{M} = \hat{\mathcal{R}}^\dagger \hat{H}_{\text{RE}} \hat{\mathcal{R}}$ , the RE Hamiltonian in the unmapped frame, which leads to

$$\Phi_{\text{R}}^{\text{rc}}(\chi, t) = \text{Tr}_{\text{ES}} [\hat{\sigma}_{\text{ES}}(\chi, t)], \quad (25)$$

where the generalised density operator is given by

$$\hat{\sigma}(\chi, t) = e^{i\frac{\chi}{2} \hat{H}_{\text{RE}}} \hat{U}_{\hat{\mathcal{R}}}(t) e^{-i\chi \hat{H}_{\text{RE}}} \hat{\rho}_{\text{S}}(0) \otimes \hat{\rho}_{\text{RC}} \otimes \hat{\rho}_{\text{RE}} \hat{U}_{\hat{\mathcal{R}}}^\dagger(t) e^{i\frac{\chi}{2} \hat{H}_{\text{RE}}}. \quad (26)$$

The two generalised density operators  $\hat{\rho}_{\text{ES}}(\chi, t)$  and  $\hat{\sigma}_{\text{ES}}(\chi, t)$  both obey the HC-RCME but have different initial conditions, being

$$\hat{\rho}_{\text{ES}}(\chi, 0) = \hat{\rho}_{\text{S}}(0) \otimes \frac{e^{-(\beta+i\chi)\hat{H}_{\text{RC}}}}{\text{Tr}\left[e^{-\beta\hat{H}_{\text{RC}}}\right]}, \quad (27)$$

and

$$\hat{\sigma}_{\text{ES}}(\chi, 0) = \hat{\rho}_{\text{S}}(0) \otimes \hat{\rho}_{\text{RC}}. \quad (28)$$

Here,  $\hat{\rho}_{\text{S}}(0)$  is the initial state of the TLS, which we take to be  $\hat{\rho}_{\text{S}}(0) = |+\rangle\langle+|$  throughout, where  $|+\rangle = \frac{1}{\sqrt{2}}(|e\rangle + |g\rangle)$ .

While the HC-RCME can in principle be used to investigate the full spin-boson model, in order to compare the difference in system-environment partitions on the resulting heat statistics it is instructive to consider the ( $\Delta = 0$ ) IBM, which is exactly solvable since the system and interaction Hamiltonians now commute [40]. Following Popovic *et al* [41], we are able to find an exact analytic expression for the CF describing energetic changes in the FE, given by

$$\begin{aligned} \Phi_{\text{F}}^{\text{ex}}(\chi, t) = \exp \left[ -2 \int_0^{\infty} d\omega \frac{J_{\text{FE}}(\omega)}{\omega^2} (1 - \cos(\omega t)) \right. \\ \left. \times \left( \coth\left(\frac{\beta\omega}{2}\right) (1 - \cos(\omega\chi)) - i \sin(\omega\chi) \right) \right]. \end{aligned} \quad (29)$$

A similar analytic expression for changes in the RE is not available since this definition relies on performing the RCM, with the result that the mapped system and interaction Hamiltonians no longer commute. Despite being a dissipation-free model, the IBM is valuable for studying heat statistics as it enables benchmarking of the HC-RCME, while highlighting non-Markovian and strong-coupling effects. There has also been recent interest in the thermodynamics of pure decoherence processes, see e.g. [42].

### 3. Results

The results presented here are based on three distinct CFs. Two of these CFs quantify energetic changes in the FE Hamiltonian, using either the approximate (equation (24)) or exact (equation (29)) methods of calculating the dynamics of the generalised density operator (i.e. by solving the HC-RCME or the IBM dynamics, respectively). The third CF (equation (25)) quantifies energetic changes in the RE Hamiltonian and is obtained by solving the HC-RCME with the appropriate initial condition. We use subscripts to denote the definition of heat associated with the CF: ‘F’ for energetic changes in the FE and ‘R’ for energetic changes in the RE. The superscript denotes the method used for calculating the generalized density operator dynamics: ‘rc’ for the HC-RCME and ‘ex’ for the analytical dynamics derived from the IBM.

#### 3.1. Characteristic function

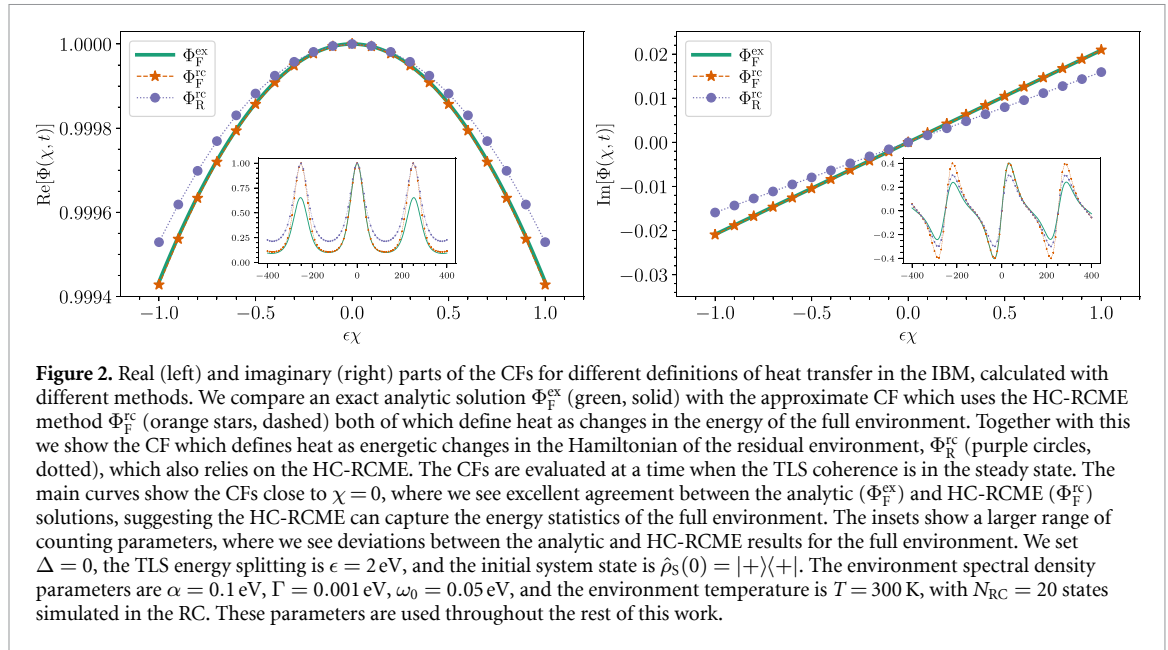
In figure 2 we plot the real (left) and imaginary (right) parts of  $\Phi_{\text{F}}^{\text{ex}}$  (green, solid) and  $\Phi_{\text{F}}^{\text{rc}}$  (orange stars, dashed), both of which count energetic changes in the FE Hamiltonian. We see excellent agreement between these two CFs close to  $\chi = 0$ , with deviations at larger values of  $\chi$  shown in the insets. The analytic result shows decaying repetitions of the main feature centred around  $\chi = 0$ , whereas the HC-RCME captures this main feature but lacks the subsequent decay for larger  $\chi$  values, due to the approximations made during its derivation. Also shown in figure 2 is the CF which counts energetic changes in the RE Hamiltonian,  $\Phi_{\text{R}}^{\text{rc}}$  (purple circles, dotted). We see similar qualitative behaviour between  $\Phi_{\text{F}}^{\text{rc}}$  and  $\Phi_{\text{R}}^{\text{rc}}$ , in that they repeat their feature centered at  $\chi = 0$ , with no decay as we increase  $\chi$ .

It is clear that there are quantitative differences in the CFs when counting only on the RE rather than the FE. The physical meaning behind these differences is not immediately obvious from these results alone. To gain physical insight, in the following section we calculate the first two moments of the probability distributions associated with these CFs.

#### 3.2. Statistical moments

The  $n^{\text{th}}$  moment of the probability distribution associated with a CF is given by

$$\langle Q^n \rangle(t) = (-i)^n \left. \frac{d^n}{d\chi^n} \Phi(\chi, t) \right|_{\chi=0}. \quad (30)$$



**Figure 2.** Real (left) and imaginary (right) parts of the CFs for different definitions of heat transfer in the IBM, calculated with different methods. We compare an exact analytic solution  $\Phi_F^{\text{ex}}$  (green, solid) with the approximate CF which uses the HC-RCME method  $\Phi_F^{\text{rc}}$  (orange stars, dashed) both of which define heat as changes in the energy of the full environment. Together with this we show the CF which defines heat as energetic changes in the Hamiltonian of the residual environment,  $\Phi_R^{\text{rc}}$  (purple circles, dotted), which also relies on the HC-RCME. The CFs are evaluated at a time when the TLS coherence is in the steady state. The main curves show the CFs close to  $\chi = 0$ , where we see excellent agreement between the analytic ( $\Phi_F^{\text{ex}}$ ) and HC-RCME ( $\Phi_F^{\text{rc}}$ ) solutions, suggesting the HC-RCME can capture the energy statistics of the full environment. The insets show a larger range of counting parameters, where we see deviations between the analytic and HC-RCME results for the full environment. We set  $\Delta = 0$ , the TLS energy splitting is  $\epsilon = 2$  eV, and the initial system state is  $\rho_S(0) = |+\rangle\langle+|$ . The environment spectral density parameters are  $\alpha = 0.1$  eV,  $\Gamma = 0.001$  eV,  $\omega_0 = 0.05$  eV, and the environment temperature is  $T = 300$  K, with  $N_{\text{RC}} = 20$  states simulated in the RC. These parameters are used throughout the rest of this work.

Thus, the excellent agreement between  $\Phi_F^{\text{ex}}$  and  $\Phi_F^{\text{rc}}$  around  $\chi = 0$  shown in figure 2 implies that the HC-RCME can accurately capture the lower-order statistical moments of the probability distributions studied here [43].

We can find analytic forms for the moments of the FE heat distribution by applying equation (30) to equation (29), giving

$$\langle Q_F^{\text{ex}}(t) \rangle = 2 \int_0^{\infty} d\omega \frac{J_{\text{FE}}(\omega)}{\omega} (1 - \cos(\omega t)), \quad (31)$$

for the mean, and

$$\text{var}[Q_F^{\text{ex}}(t)] = 2 \int_0^{\infty} d\omega J_{\text{FE}}(\omega) \coth\left(\frac{\beta\omega}{2}\right) (1 - \cos(\omega t)), \quad (32)$$

for the variance.

To calculate the mean and variance predicted by the approximate CFs,  $\Phi_F^{\text{rc}}$  and  $\Phi_R^{\text{rc}}$ , we follow the finite-difference method used by Popovic *et al* [41]. By choosing a small value of the counting parameter  $\chi_\delta$ , we find the mean as

$$\langle Q_b^{\text{rc}}(t) \rangle = \frac{\text{Im}[\Phi_b^{\text{rc}}(\chi_\delta, t)]}{\chi_\delta} + \mathcal{O}(\chi_\delta), \quad (33)$$

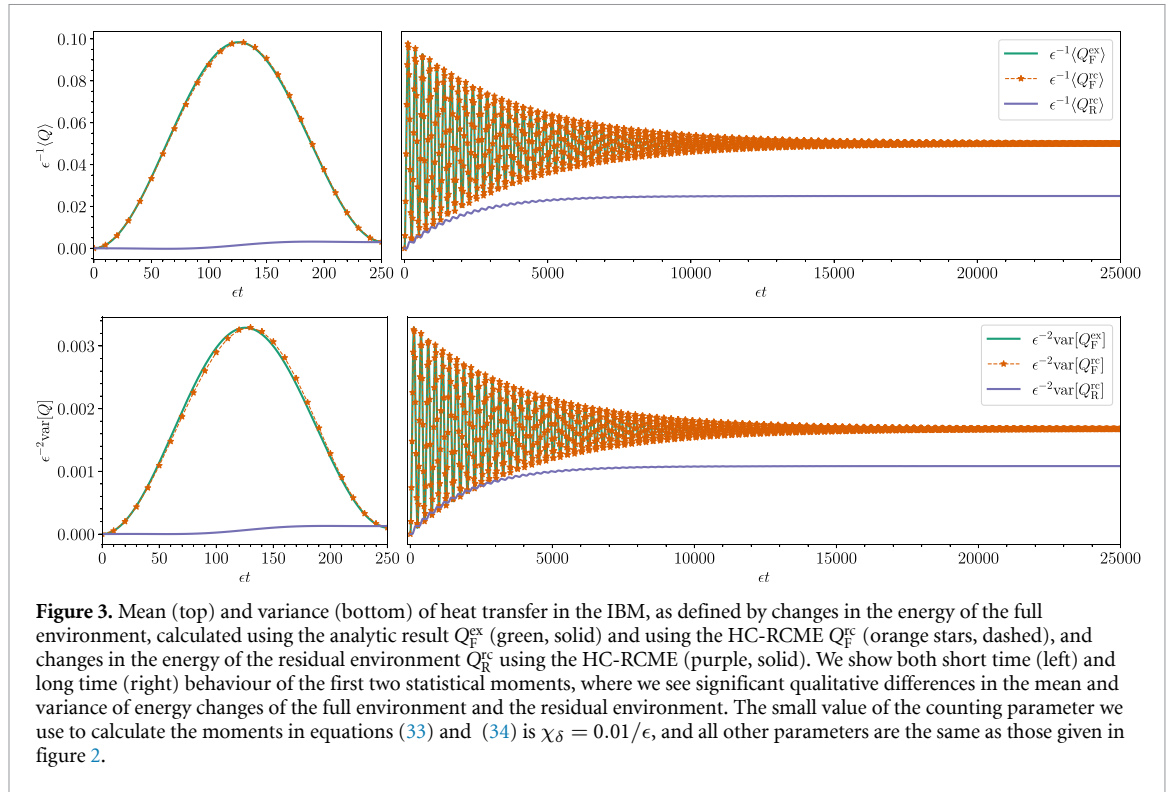
and the variance as

$$\text{var}[Q_b^{\text{rc}}(t)] = \frac{2 - 2\text{Re}[\Phi_b^{\text{rc}}(\chi_\delta, t)]}{\chi_\delta^2} - \langle Q_b^{\text{rc}}(t) \rangle^2 + \mathcal{O}(\sqrt{2}\chi_\delta), \quad (34)$$

where  $b = F$  for the FE definition and  $b = R$  for the RE definition. In the above  $\mathcal{O}(x)$  represents error of order  $x$ .

In figure 3 (top) we plot the mean energy change of the FE, calculated using the exact method  $\langle Q_F^{\text{ex}} \rangle$  (green, solid) and the HC-RCME method  $\langle Q_F^{\text{rc}} \rangle$  (orange stars, dashed), as well as the mean energy change of the RE  $\langle Q_R^{\text{rc}} \rangle$  (purple, solid), showing both short (left) and long (right) timescales. We clearly see that the HC-RCME accurately describes the mean energy change of the FE. Interestingly, this definition of heat has large oscillations present, which can be intuitively explained using the RCM. The TLS strongly couples to the RC, which captures the long memory effects of the FE. This leads to a coherent exchange of energy and information between the TLS and RC, suggesting that ‘heat’, as defined by the change in energy of the FE, is not irreversibly lost to the environment, but can re-excite [44] the TLS leading to coherent oscillations.

This contrasts with the mean heat predicted by counting on only the RE, which contains heavily suppressed oscillations, suggesting that energy and information that leave the ES is lost irreversibly to the RE.



This behaviour is more in keeping with the classical definition of heat, which is understood to be the changes in internal energy which lead to a monotonic entropy change, and which flows in a unidirectional manner from a hot body to a cold body.

These differences are also reflected in figure 3 (bottom), where we plot the variances associated with the different CFs. Once again, we see excellent agreement between the exact treatment and the HC-RCME when counting on the FE. Similar to the case of the mean, we see both quantitative and qualitative differences in the variance when counting only on the RE, with coherent oscillations suppressed. The variance of heat transfer is expected to monotonically increase with time, as seen in the RE definition, suggesting it is most suitable for the non-Markovian regime.

### 3.3. Changes in ergotropy and entropy

The coherent oscillations observed in figure 3 for both methods of calculating the mean energy change of the full environment,  $\langle Q_F^{ex} \rangle$  and  $\langle Q_F^{rc} \rangle$ , suggests that there is a work-like contribution within this definition of heat. The RC method provides us with a unique insight into this behaviour, providing an avenue to calculate the ergotropy [24, 45, 46] of the TLS, which then treats all energy emitted into the environment as heat (i.e. the full environment paradigm referenced above), as well as the ergotropy of the ES, which treats the RC as a potential work source.

Ergotropy is defined as the maximum amount of work we can extract unitarily from a quantum state, and for the two environment partitions can be defined as,

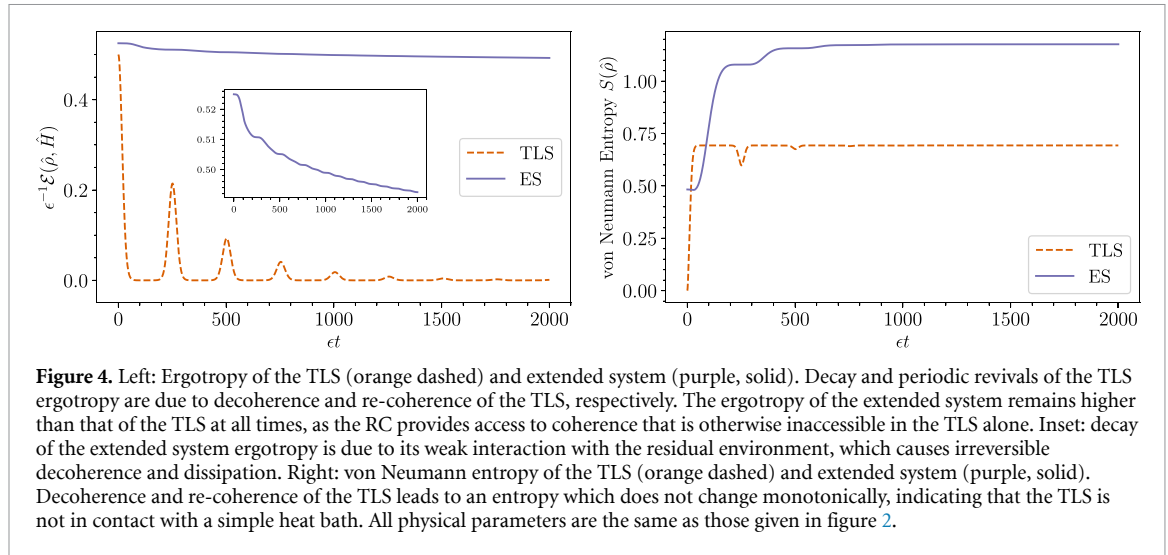
$$\mathcal{E}(\hat{\rho}_S, \hat{H}_S) = \text{Tr}[\hat{H}_S \hat{\rho}_S] - \min_{\hat{U}} \text{Tr}[\hat{H}_S \hat{U} \hat{\rho}_S \hat{U}^\dagger], \quad (35)$$

for the TLS, and

$$\mathcal{E}(\hat{\rho}_{ES}, \hat{H}_{ES}) = \text{Tr}[\hat{H}_{ES} \hat{\rho}_{ES}] - \min_{\hat{V}} \text{Tr}[\hat{H}_{ES} \hat{V} \hat{\rho}_{ES} \hat{V}^\dagger], \quad (36)$$

for the ES. The operators  $\hat{U}$  and  $\hat{V}$  act on the Hilbert spaces of the TLS and ES, respectively [47]. The minimisation is satisfied through a unitary transformation which takes the state to its passive counterpart, from which no further work can be extracted [24]. By diagonalising the state as

$$\hat{\rho} = \sum_j r_j |r_j\rangle\langle r_j|, \quad (37)$$



such that  $r_j > r_{j+1}$ , and diagonalising the Hamiltonian as

$$\hat{H} = \sum_k h_k |h_k\rangle\langle h_k|, \quad (38)$$

such that  $h_k < h_{k+1}$  the ergotropy is given by

$$\mathcal{E}(\hat{\rho}, \hat{H}) = \sum_{jk} r_j h_k \left( |\langle r_j | h_k \rangle|^2 - \delta_{jk} \right). \quad (39)$$

In figure 4 (left) we plot the ergotropies of the TLS state and ES state, when initialising the TLS in  $\hat{\rho}_S(0) = |+\rangle\langle +|$  and the RC in the Gibbs state. At  $t = 0$  the TLS ergotropy (orange, dashed) is the amount of extractable energy by unitarily transforming the state from  $|+\rangle$  to the ground state  $|g\rangle$ , as expected. We then see a sharp decrease in ergotropy followed by decaying recurrences, which are in phase with the decoherence and re-coherences of the TLS induced by the non-Markovian interaction with the FE, as seen in figure 5. This is explained by coherence in the energy eigenbasis contributing to ergotropy [25, 48, 49]. When calculating the ergotropy of the TLS state, we assume we have complete control over the TLS, but no control over the FE. During decoherence, the TLS cannot use coherence in the energy eigenbasis as a source of extractable work, and hence the ergotropy decreases. During the re-coherence process, the FE generates coherence in the TLS (in ever smaller quantities), providing an increase in the extractable work (also in ever smaller quantities).

When considering the ergotropy of the ES, we see that the presence of the RC increases ergotropy by providing access to coherence that are otherwise lost to the FE. Notably, the ergotropy of the ES begins at a larger value than that of the TLS, despite the RC beginning in the Gibbs state. While the Gibbs state of the RC is passive with respect to the RC Hamiltonian, the presence of the TLS-RC interaction Hamiltonian means the Gibbs state is not passive with respect to the ES Hamiltonian, thus providing an increase in the amount of extractable work for the ES state. This interaction maintains coherence within the ES, explaining the (relatively) constant ergotropy. However, a gradual decay occurs in the ES's ergotropy resulting from the weak interaction between the RC and the (infinite) RE, causing slow, and irreversible decoherence and dissipation in the ES, as seen in the inset of figure 4 (left).

Next, we calculate the von Neumann entropy for the TLS and the ES states. In the IBM, if we begin the TLS in a pure state (i.e.  $|+\rangle$ ), we know it will dephase due to its interaction with the FE. We therefore expect the entropy of the TLS to increase as the system loses coherence and becomes more mixed, and to decrease during any re-coherence process as it becomes purer. However, were the system to interact with a heat sink, its entropy should change monotonically.

In figure 4 (right), we see that the von Neumann entropy of the TLS (orange dashed) does not change monotonically, which adds evidence to the argument that the TLS is not in contact with a heat bath (i.e. changes in the FE cannot be characterised solely as heat in this regime). Notice that the changes in entropy of the TLS are in phase with changes in its ergotropy, both of which are in phase with the coherence of the TLS shown in figure 5. As the TLS gains coherence via a non-Markovian interaction, its state becomes purer, reducing entropy and increasing ergotropy. Meanwhile, the ES's entropy increases monotonically [50], adding further evidence to the fact that the RE acts as a heat bath.

These results appear to show that the RC is a viable work source, at least in principle [51]. Therefore, if work and heat are to be defined as separate and distinct components of a system's internal energy change, definitions of heat that do not include energetic changes within the RC may be more appropriate than those that do. This gives further evidence to support using the definition of heat as energetic changes in the RE Hamiltonian when studying the non-Markovian regime.

#### 4. Discussion

We have used the TPMP to study the full counting statistics of energetic changes in the IBM within the non-Markovian regime. We employed the RC formalism, which not only accounts for TLS-environment correlations through the RC—enabling us to study energetic changes in the environment in strong coupling and non-Markovian regimes—but also allows us to investigate how different system-environment partitions impact the definition of heat in open quantum systems. In future work it would be interesting to study the similarities and differences between secular and non-secular versions of the HC-RCME, as well as the behaviour of the heat definition when performing the RCM multiple times.

Notably, we find that a naive definition of heat, in which all energy changes of an environment is categorised as heat, overlooks significant work-like contributions present in the TLS-environment interactions, which leads to coherent oscillations in both the average and variance of these energetic changes. The RC formalism, however, allows us to use an alternative definition in which only energetic changes of the RE are considered as heat. With this definition, coherent oscillations in the mean and variance of heat are heavily suppressed, suggesting that heat transfer within the IBM in this case is truly an irreversible process. This is supported by considering the ergotropy and von Neumann entropy for both the TLS and ES.

This work demonstrates that defining heat as changes in the energy of the RE, as characterized by the RCM, aligns with the classical intuition of heat: monotonic entropy non-conserving changes in internal energy that are distinct from work transfer. Recent research by Colla and Breuer [52] supports this conclusion, revealing that peaks in the spectral density of a supposed heat bath can enable it to partially function as a work reservoir. Combined with our findings, this suggests that assumptions about the interaction between an open quantum system and a thermal environment must be made carefully when considering quantum thermodynamics. It is thus particularly crucial to consider the structure of the environment's spectral density when evaluating energy transfer and distinguishing between heat and work.

#### Data availability statement

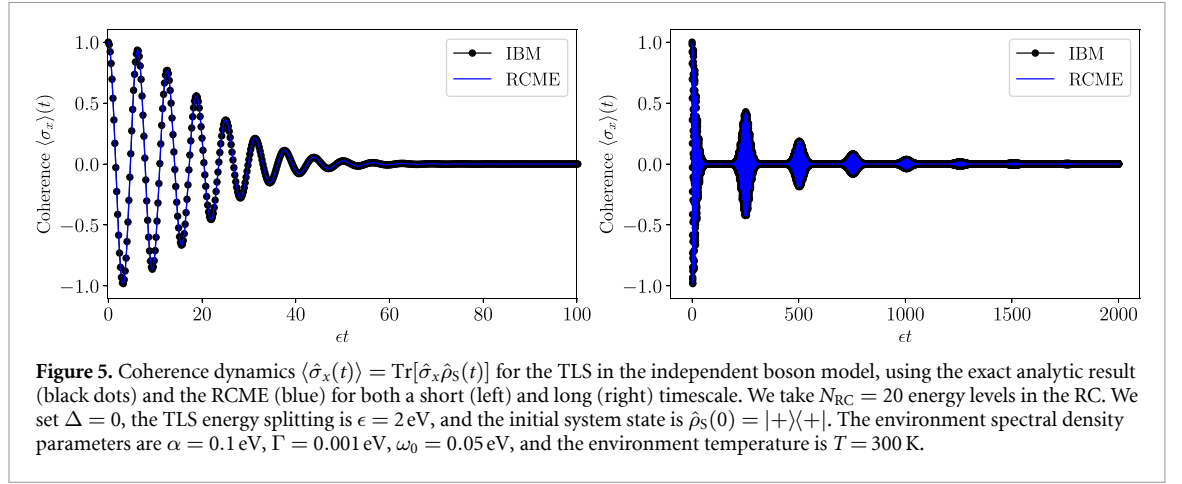
All data that support the findings of this study are included within the article (and any supplementary files).

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#### Appendix A. Benchmarking dynamics

Here we show that the RCME (HC-RCME with  $\chi = 0$ ) is able to track TLS properties accurately. In figure 5 we plot the dynamics of the TLS coherence  $\langle \hat{\sigma}_x(t) \rangle$  on both a short (left) and long (right) timescale, by solving the RCME (blue) and from the analytic result for the IBM (black dots). We see that the RCME is able to track the TLS coherence dynamics very well with  $N_{RC} = 20$  energy levels being simulated in the RC. The TLS energy splitting is  $\epsilon = 2$  eV, the environment spectral density parameters are  $\alpha = 0.1$  eV,  $\Gamma = 0.001$  eV,  $\omega_0 = 0.05$  eV, and the environment temperature is  $T = 300$  K (as elsewhere). The TLS coherence shows decaying oscillations on a short timescale, figure 5 (left), with periodic re-coherences on longer timescales, which themselves decay in time, figure 5 (right). These re-coherences are a result of the sharply peaked form of the FE spectral density, and indicate that we are working in a regime where the standard Born–Markov master equation would fail, as it is unable to predict re-coherences of the TLS.



## Appendix B. Generalised master equation in the RC frame

The interaction Hamiltonian in equation (10) can be written as  $\hat{H}_I = \hat{A} \otimes \hat{B}$ , where  $\hat{A}$  and  $\hat{B}$  are operators on the Hilbert spaces of the ES and RE, respectively. We ignore the counter term since it does not contribute to the master equation we are about to derive [15, 20]. Dressing this interaction Hamiltonian with the counting parameter and moving into the interaction picture with respect to  $\hat{H}_{\text{ES}} + \hat{H}_{\text{RB}}$  gives

$$\tilde{H}_I(\chi, t) = e^{i\hat{H}_{\text{ES}}t} \hat{A} e^{-i\hat{H}_{\text{ES}}t} \otimes e^{i\hat{H}_{\text{RE}}t} e^{i\frac{\chi}{2}\hat{H}_{\text{RE}}} \hat{B} e^{-i\frac{\chi}{2}\hat{H}_{\text{RE}}} e^{-i\hat{H}_{\text{RE}}t} = \tilde{A}(t) \otimes \tilde{B}(\chi, t). \quad (\text{B1})$$

Upon applying the Born–Markov approximations between the ES and RE we arrive at the ‘HC-RCME’

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_{\text{ES}}(\chi, t) &= -i[\hat{H}_{\text{ES}}, \hat{\rho}_{\text{ES}}(\chi, t)] - \hat{A}\hat{A}_1\hat{\rho}_{\text{ES}}(\chi, t) + \hat{A}\hat{\rho}_{\text{ES}}(\chi, t)\hat{A}_2(\chi) + \hat{A}_3(\chi)\hat{\rho}_{\text{ES}}(\chi, t)\hat{A} - \hat{\rho}_{\text{ES}}(\chi, t)\hat{A}_4\hat{A} \\ &= \mathcal{L}_{\text{ES}}(\chi)[\hat{\rho}_{\text{ES}}(\chi, t)]. \end{aligned} \quad (\text{B2})$$

The terms in the above equation are given by

$$\hat{A}_1 = \int_0^\infty d\tau \tilde{A}(-\tau) \langle \tilde{B}(\chi, \tau) \hat{B}(\chi) \rangle, \quad (\text{B3})$$

$$\hat{A}_2(\chi) = \int_0^\infty d\tau \tilde{A}(-\tau) \langle \tilde{B}(-\chi, -\tau) \hat{B}(\chi) \rangle, \quad (\text{B4})$$

$$\hat{A}_3(\chi) = \int_0^\infty d\tau \tilde{A}(-\tau) \langle \tilde{B}(-\chi, \tau) \hat{B}(\chi) \rangle, \quad (\text{B5})$$

$$\hat{A}_4 = \int_0^\infty d\tau \tilde{A}(-\tau) \langle \tilde{B}(-\chi, -\tau) \hat{B}(-\chi) \rangle, \quad (\text{B6})$$

with

$$\tilde{A}(-\tau) = \sum_{j,k=1}^{2N_{\text{RC}}} e^{-i\lambda_{jk}\tau} A_{jk} |\lambda_j\rangle\langle \lambda_k|, \quad (\text{B7})$$

where the ES Hamiltonian in its spectral form is given by  $\hat{H}_{\text{ES}} = \sum_{k=1}^{2N_{\text{RC}}} \lambda_k |\lambda_k\rangle\langle \lambda_k|$ , and where the  $\chi$ -dependent RE correlation functions are given by

$$\langle \tilde{B}(\chi, \tau) \hat{B}(\chi) \rangle = \int_0^\infty d\omega J_{\text{RE}}(\omega) \left( N(\omega) e^{i\omega\tau} + (1 + N(\omega)) e^{-i\omega\tau} \right), \quad (\text{B8})$$

$$\langle \tilde{B}(-\chi, \pm\tau) \hat{B}(\chi) \rangle = \int_0^\infty d\omega J_{\text{RE}}(\omega) \left( N(\omega) e^{\pm i\omega\tau} e^{-i\chi\omega} + (1 + N(\omega)) e^{\mp i\omega\tau} e^{i\chi\omega} \right), \quad (\text{B9})$$

$$\langle \tilde{B}(-\chi, -\tau) \hat{B}(-\chi) \rangle = \int_0^{\infty} d\omega J_{\text{RE}}(\omega) \left( N(\omega) e^{-i\omega\tau} + (1 + N(\omega)) e^{i\omega\tau} \right). \quad (\text{B10})$$

In the above,  $N(\omega) = (e^{\beta\omega} - 1)^{-1}$  is the thermal occupation number for bosons. By making use of the Sokhotski–Plemelj theorem [26]

$$\int_0^{\infty} d\tau e^{-i(\omega \pm \nu)\tau} = \pi \delta(\omega \pm \nu) - i\mathcal{P} \left( \frac{1}{\omega \pm \nu} \right), \quad (\text{B11})$$

we can evaluate the time and frequency integrals. In the above,  $\delta(x - a)$  represents a Dirac delta function centred at  $x = a$ , and  $\mathcal{P}$  is the Cauchy principal value. After evaluating the integrals, ignoring the principal value terms, and taking into consideration the three possible cases for  $\lambda_{mn}$ , we obtain

$$\hat{A}_1 = \sum_{mn} \begin{cases} \pi A_{mn} J_{\text{RE}}(\lambda_{mn}) N(\lambda_{mn}) |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} > 0, \\ \pi A_{mn} J_{\text{RE}}(|\lambda_{mn}|) (1 + N(|\lambda_{mn}|)) |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} < 0, \\ \pi A_{mn} \gamma \beta^{-1} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} = 0, \end{cases} \quad (\text{B12})$$

$$\hat{A}_2(\chi) = \sum_{mn} \begin{cases} A_{mn} J_{\text{RE}}(\lambda_{mn}) (1 + N(\lambda_{mn})) e^{i\chi \lambda_{mn}} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} > 0, \\ \pi A_{mn} J_{\text{RE}}(|\lambda_{mn}|) N(|\lambda_{mn}|) e^{-i\chi |\lambda_{mn}|} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} < 0, \\ \pi A_{mn} \gamma \beta^{-1} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} = 0, \end{cases} \quad (\text{B13})$$

$$\hat{A}_3(\chi) = \sum_{mn} \begin{cases} \pi A_{mn} J_{\text{RE}}(\lambda_{mn}) N(\lambda_{mn}) e^{-i\chi \lambda_{mn}} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} > 0, \\ \pi A_{mn} J_{\text{RE}}(|\lambda_{mn}|) (1 + N(|\lambda_{mn}|)) e^{i\chi |\lambda_{mn}|} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} < 0, \\ \pi A_{mn} \gamma \beta^{-1} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} = 0, \end{cases} \quad (\text{B14})$$

$$\hat{A}_4 = \sum_{mn} \begin{cases} \pi A_{mn} J_{\text{RE}}(\lambda_{mn}) (1 + N(\lambda_{mn})) |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} > 0, \\ \pi A_{mn} J_{\text{RE}}(|\lambda_{mn}|) N(|\lambda_{mn}|) |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} < 0, \\ \pi A_{mn} \gamma \beta^{-1} |\lambda_m\rangle \langle \lambda_n| & \text{if } \lambda_{mn} = 0. \end{cases} \quad (\text{B15})$$

If we set  $\chi = 0$  we recover the standard RCME.

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- [45] Perarnau-Llobet M, Hovhannisyan K V, Huber M, Skrzypczyk P, Brunner N and Acín A 2015 Extractable work from correlations *Phys. Rev. X* **5** 041011
- [46] Sone A and Deffner S 2021 Quantum and classical ergotropy from relative entropies *Entropy* **23** 1107
- [47] The time dependence of the ergotropies and states have been omitted from the above definition for ease of reading
- [48] Çakmak B 2020 Ergotropy from coherences in an open quantum system *Phys. Rev. E* **102** 042111
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- [50] Except for a slight decrease within the first few time steps
- [51] We are not suggesting a method of extracting work from the RC, but are using the fact that work can in principle be extracted from it to make arguments about the most appropriate definition of heat
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