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IOP Institute of Physics

Published in partnership with: Deutsche Physikalische Gesellschaft and the Institute of Physics

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OPEN ACCESS

RECEIVED
2 March 2021

REVISED
29 April 2021

ACCEPTED FOR PUBLICATION
5 May 2021

PUBLISHED
11 June 2021

PAPER

Contributions from populations and coherences in non-equilibrium entropy production

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Keywords: quantum thermodynamics, quantum coherence, many-body physics

Abstract

The entropy produced when a quantum system is driven away from equilibrium can be decomposed in two parts, one related with populations and the other with quantum coherences. The latter is usually based on the so-called relative entropy of coherence, a widely used quantifier in quantum resource theories. In this paper we argue that, despite satisfying fluctuation theorems and having a clear resource-theoretic interpretation, this splitting has shortcomings. First, it predicts that at low temperatures the entropy production will always be dominated by the classical term, irrespective of the quantum nature of the process. Second, for infinitesimal quenches, the radius of convergence diverges exponentially as the temperature decreases, rendering the functions non-analytic. Motivated by this, we provide here a complementary approach, where the entropy production is split in a way such that the contributions from populations and coherences are written in terms of a thermal state of a specially dephased Hamiltonian. The physical interpretation of our proposal is discussed in detail. We also contrast the two approaches by studying work protocols in a transverse field Ising chain, and a macrospin of varying dimension.

1. Introduction and preliminary results

Quantum coherence and quantum correlations play a key role in the thermodynamics of microscopic systems [1, 2]. They can be exploited to extract useful work [3–9], speed-up energy exchanges [10–13], and improve heat engines [14–19]. On a more fundamental level, they alter the possible state transitions in thermodynamic processes [20–22], lead to new forms of work and heat fluctuations [23–28], modify the fluctuation–dissipation relation (FDR) for work [29–31] and may even generate heat flow reversals [32–35]. Understanding the role of coherence in the formulation of the laws of quantum thermodynamics is therefore a major overarching goal in the field, which has been the subject of considerable recent interest.

When a system relaxes to equilibrium, in contact with a heat bath, quantum coherences are known to contribute an additional term to the entropy production [21, 36, 37], which quantifies the amount of irreversibility in the process. A similar effect also happens in unitary work protocols [38, 39]. To be concrete, we focus on the latter and consider a scenario where a system is described by a Hamiltonian $H_t = H(g_t)$, depending on a controllable parameter g_t. The system is initially prepared in thermal equilibrium at a temperature T, such that its initial state is the thermal state $\rho_0^{\mathrm{th}} \equiv \rho^{\mathrm{th}}(g_0) = \mathrm{e}^{-\beta H_0}/Z_0$, where $\beta = 1/T$ and $Z_0 = \mathrm{tr}\,\mathrm{e}^{-\beta H_0}$ is the partition function. At $t = 0$, a work protocol g_t, that lasts for a total time τ, is applied to the system, driving it out of equilibrium [40, 41]. Letting U denote the unitary

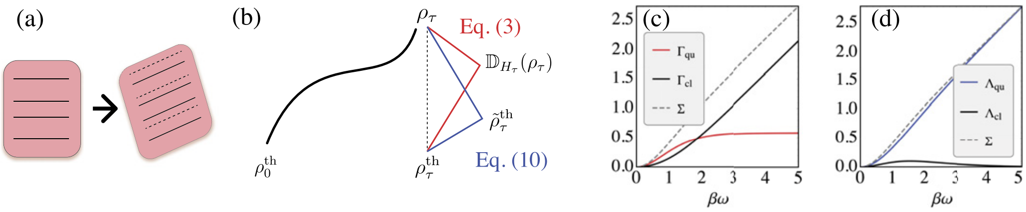

Figure 1. Difficulties in identifying the classical and quantum contributions to the entropy production. (a) A work protocol $H(g_t)$ can modify the Hamiltonian in two ways: altering the energy level spacings, which can be viewed as a semi-classical effect, and rotating the energy eigenbasis, which is a fully quantum property with no classical counterpart. (b) The entropy production (2) compares the final state ρ_τ with the reference thermal state $\rho_\tau^{\rm th}$. To understand its classical and quantum contributions, the splitting (3) uses an intermediate state $\mathbb{D}_{H_\tau}(\rho_\tau)$. Conversely, in this paper we introduce the splitting (10), which uses the state $\tilde{\rho}_\tau^{\rm th}$, in equation (17). (c) The contributions $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$ of equation (3) in a minimal qubit model, as a function of $\beta\omega$, plotted using equations (6) and (7) with $\theta = 1.1$ (see text for details). (d) The new splitting (10) (see also equation (11)), which yields physically more reasonable results at low temperatures.

generated by the drive, the state of the system after a time τ will be

$$\rho_\tau = U\rho_0^{\rm th}U^\dagger. \tag{1}$$

In general, ρ_τ will be very different from the corresponding equilibrium state $\rho_\tau^{\rm th} = {\rm e}^{-\beta H_\tau}/Z_\tau$. This difference is captured by the entropy production (also called non-equilibrium lag in this context) [42–45],

$$\Sigma = S\left(\rho_\tau \| \rho_\tau^{\rm th}\right), \tag{2}$$

where $S\left(\rho\|\sigma\right) = tr\left\{\rho(\ln\rho - \ln\sigma)\right\}$ is the quantum relative entropy. The non-equilibrium lag is directly proportional to the irreversible work [46–48], $\Sigma = \beta\left(\langle W\rangle - \Delta F\right)$, where $\langle W\rangle = {\rm tr}\left(H_\tau\rho_\tau - H_0\rho_0^{\rm th}\right)$ is the work performed in the process and $\mathit{\Delta} F = F(g_\tau) - F(g_0)$ is the change in equilibrium free energy, $F(g) = {\rm tr}\left\{H(g)\rho^{\rm th}(g)\right\} - TS(\rho^{\rm th}(g))$ (with $S(\rho) = -tr\{\rho\ln\rho\}$ being the von Neumann entropy). Due to its clear thermodynamic interpretation, has been widely used as a quantifier of irreversibility, both theoretically [45–53] and experimentally [54–65].

The entropy production Σ in equation (2) contains contributions of both a classical and quantum nature. This is linked with the fact that the work protocol g_t can modify the Hamiltonian $H(g_t)$ in two ways. On the one hand, it may alter the spacing of the energy levels; and, on the other, it may rotate the eigenvectors (figure 1(a)). The latter is directly associated with quantum coherence and to the fact that $[H(g_{t_1}), H(g_{t_2})] \neq 0$, for two different times t_1, t_2. It therefore has no classical counterpart, and corresponds to a fundamental feature distinguishing classical and quantum processes. In general, these two processes will become mixed, and hence identifying how each physical process contributes to Σ is in general a challenging task. In the literature, a popular choice is the splitting put forward in [20, 21, 36, 38]:

$$\Sigma = \Gamma_{\rm cl} + \Gamma_{\rm qu}, \tag{3}$$

where

$$\Gamma_{\rm cl} = S\left(\mathbb{D}_{H_\tau}(\rho_\tau)\|\rho_\tau^{\rm th}\right), \tag{4}$$

$$\Gamma_{\rm qu} = S\left(\rho_\tau\|\mathbb{D}_{H_\tau}(\rho_\tau)\right) = S\left(\mathbb{D}_{H_\tau}(\rho_\tau)\right) - S(\rho_\tau), \tag{5}$$

with $\mathbb{D}_H(\rho)$ being the super-operator that completely dephases the state ρ in the eigenbasis of H (explicitly defined below, in equation (12)). The first term, $\Gamma_{\rm cl}$, measures the entropic distance between the populations of the actual final state ρ_τ and those of the reference thermal state $\rho_\tau^{\rm th}$, and is generally identified with the classical contribution. The term $\Gamma_{\rm qu}$, in turn, is known as the relative entropy of coherence and compares the final state ρ_τ with the dephased state $\mathbb{D}_{H_\tau}(\rho_\tau)$. It hence captures the contribution from coherences in the energy basis. By construction, $\Gamma_{\rm cl}$ and $\Gamma_{\rm qu}$ in equation (3) are both non-negative, which shows that coherences increase the entropy production in the process, as compared to a fully classical (incoherent) scenario. One should also clarify that, since the changes in populations and coherences are inevitably mixed, the terminology 'classical' vs 'quantum' is not entirely precise, nor is there a one-to-one relationship between this and the terms 'populations' and 'coherences'. For instance, while $\Gamma_{\rm qu}$ depends only on the basis rotation (coherences), depends on both the changes in energy eigenvalues, as well as the eigenbasis rotation. Notwithstanding, as we will show, in the case of infinitesimal quenches, these distinctions can be made precise.

The splitting (3), first analyzed in [20], has been studied in the context of the resource theory of thermodynamics [21], relaxation towards equilibrium [36, 37], thermodynamics of quantum optical systems [66] and work protocols in the absence of a bath [9, 38, 39]. At the stochastic level, both $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$ satisfy individual fluctuation theorems [38], which is a very desirable property. Moreover, $\Gamma_{\rm cl}$ has a resource-theoretic interpretation within the resource theory of athermality [67, 68], while $\Gamma_{\rm qu}$ is a natural monotone in the resource theory of coherence [69, 70]. These facts make the splitting (3) a valuable tool in understanding the relative contribution of classical and quantum features to non-equilibrium processes. However, working with various models, we have observed that this splitting behaves strangely, even in some simple protocols. More specifically, we identify two main shortcomings.

The first concerns the relative magnitudes of $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$: at low temperatures, $\Gamma_{\rm cl}$ will always be much larger than $\Gamma_{\rm qu}$. The reason is purely mathematical: $\Gamma_{\rm qu}$ is a special kind of relative entropy because it can be expressed as a difference between two von Neumann entropies, as in the second equality of (5). As $\beta \to \infty$, $\rho_0^{\rm th}$ tends to a pure state and hence $S(\rho_\tau)$ tends to zero, while $S(\mathbb{D}_{H_\tau}(\rho_\tau)) \in [0, \ln\, d]$, where d is the dimension of the Hilbert space. As a consequence, $\Gamma_{\rm qu}$ will always remain finite. The term $\Gamma_{\rm cl}$, on the other hand, generally diverges when the support of ρ_τ is not contained in that of $\rho_\tau^{\rm th}$ [71], meaning $\Gamma_{\rm cl}$ will grow unbounded when $\beta \to \infty$. This implies that it is *impossible* to construct a low-temperature process where the quantum term dominates.

More precisely, consider again the two types of drivings depicted in figure 1(a): one that alters the spacing of the energy levels (associated here to a classical process), and one which may rotate eigenvectors (associated here to a quantum process). At strictly zero temperature, a Gibbs state is invariant under the first class of protocols, and hence we may expect that any entropy production in (2) has a quantum origin. However, the opposite identification arises in the splitting (3). The reason for this apparent contradiction is rather simple: the splitting (3) is not characterising whether the driving generates quantum coherence or not; rather, given a possibly coherent process, it characterises how much the final diagonal and off-diagonal terms contribute to the total entropy production.

This issue can be neatly illustrated by a minimal qubit model. Consider a qubit which starts at $H_0 = \omega\sigma^z$ and is suddenly quenched ($U = 1$) to $H_\tau = \omega(\sigma^z \cos\theta + \sigma^x \sin\theta)$ (where σ^α are Pauli matrices). In this quench the energy levels remain intact and all that happens is that the eigenbasis is rotated by an angle θ. This is thus, by all accounts, a highly quantum process. The entropy production (2) for this model reads

$$\Sigma = 2t\tanh^{-1}(t)\sin^2(\theta/2), \tag{6}$$

where $t = \tanh(\beta\omega) \in [0, 1]$. On the other hand, the coherent contribution $\Gamma_{\rm qu}$ in equation (5), reads

$$\Gamma_{\rm qu} = t\tanh^{-1}(t) - t\,\cos\theta\tanh^{-1}(t\,\cos\theta) - \frac{1}{2}\ln\left(1 + \sinh^2(\beta\omega)\sin^2\theta\right). \tag{7}$$

A plot of $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl} = \Sigma - \Gamma_{\rm qu}$ is shown in figure 1(c) as a function of $\beta\omega$, for $\theta = 1.1$. As can be seen, in general both quantities are comparable in magnitude. But, as the temperature goes down (β goes up), the classical contribution becomes increasingly larger and eventually dominates. Thus, at very low temperatures, most of Σ comes from the population term $\Gamma_{\rm cl}$ and very little from coherences.

The above considerations highlight the fact that splitting the total entropy production (2) in a classical and quantum contribution may be highly non-trivial, and that different splittings might provide different insights. In particular, we argue that the splitting in equation (3) does not appropriately distinguish coherent from non-coherent drivings (see figure 1), but instead characterises how populations and off-diagonal terms contribute to entropy production. In this work, we will propose a new complementary splitting that better incorporates the difference between coherent and non-coherent drivings.

A second issue with the splitting (3) concerns *infinitesimal quenches*. This is a very important scenario, widely studied in the context of critical systems [72–75] and quasi-isothermal processes [29, 30]. The idea is to analyze the entropy production perturbatively, for a small instantaneous quench of the work parameter, from g to $g + \delta g$. The problem with $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$ in this case is that, as will be shown, the parameter δg appears multiplied by a factor that increases exponentially with β. Hence, the radius of convergence of $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$, in δg, tends to zero exponentially fast as $\beta \to \infty$. For Σ, no such issue arises.

This is again well illustrated by the qubit example in equations (6) and (7), where the quench parameter is now the angle θ. We see that Σ in (6) can be readily expanded in powers of θ, for any temperature β (or any $t = \tanh(\beta\omega)$). The same is not true for $\Gamma_{\rm qu}$, however. The problem is in the third term of equation (7), which is a function of $x = \sinh^2(\beta\omega)\sin^2\theta$. This quantity appears inside a logarithm, in the form $\ln(1 + x)$. However, a series expansion of $\ln(1 + x)$ only converges if $|x| < 1$. And since the prefactor $\sinh^2(\beta\omega)$ grows exponentially with β, at low temperatures, extremely small values of are required to validate a series expansion.

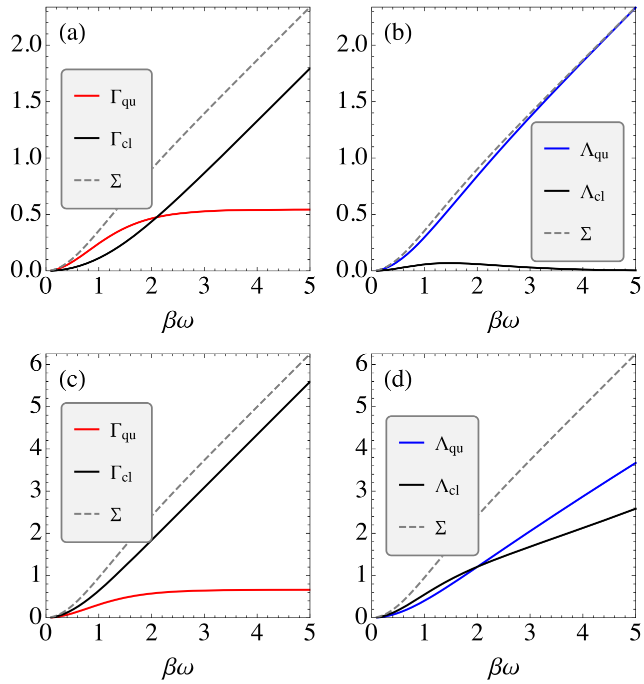

Figure 2. Splitting of the entropy production in a cyclic qubit model, $H_\tau = H_0 = \omega\sigma^z$. The unitary is generated by an x-pulse with a duration τ; that is, $U = e^{-i\tau(H_0 + h_x\sigma^x)}$, where h_x is the pulse intensity. The curves were computed using equations (14)–(16) and (18) and (19), with $\omega = 1$, $h_x = 1.3$ and two different values of τ: in (a) and (b) $\tau = 0.4$ and in (c) and (d) $\tau = 1$.

More generally, one can readily show that for Σ this issue does not arise. If we use $\Sigma = \beta\left(\langle W\rangle - \Delta F\right)$, we find in the case of infinitesimal quenches that

$$\Sigma = \beta \mathrm{tr}\left\{\Delta H \rho^{\mathrm{th}}(g_0)\right\} - \beta\Delta F, \tag{8}$$

where $\Delta H = H(g_0 + \delta g) - H(g_0)$ and $\Delta F = F(g_0 + \delta g) - F(g_0)$. A series expansion of Σ in δg therefore amounts to two things. First, an expansion of ΔH in powers of δg, which is entirely independent of β. And second, an expansion of $F(g)$, which is an analytic and generally smooth function (except possibly at a critical point [73]). Indeed, if $H(g)$ is linear in g, the leading order contribution to the expansion becomes [74]

$$\Sigma \simeq -\frac{1}{2}\beta\delta g^2 \frac{\partial^2 F}{\partial g_0^2}, \tag{9}$$

showing that Σ is simply proportional to the equilibrium susceptibility, a textbook quantity used throughout equilibrium statistical mechanics.

The above results show that, despite its interesting properties (individual fluctuation theorems and resource-theoretic interpretation), the splitting (3) is not a fully satisfying splitting of the entropy production into a classical and quantum contribution (in the sense described in figure 1(a)). In order to capture the difference between coherent and non-coherent drivings, in this paper we propose a different splitting, which is inspired by the recent results of [30]. We label it as

$$\Sigma = \Lambda_{\mathrm{cl}} + \Lambda_{\mathrm{qu}}. \tag{10}$$

The actual definitions of Λ_{qu} and Λ_{cl} will be given below in section 2 and a stochastic trajectories formulation will be given in section 3. A comparison in the case of the minimal qubit example is also presented in figure 1(d). In this case, using the results of section 2, one finds the following elegant expression for Λ_{qu} (to be contrasted with equation (7)):

$$\Lambda_{\mathrm{qu}} = \frac{1}{2}\ln\left(\frac{1 - \tanh^2(\beta\omega\cos\theta)}{1 - \tanh^2(\beta\omega)}\right). \tag{11}$$

As seen in figure 1(d), Λ_{qu} and Λ_{cl} behave as desired: since the process is highly coherent, Λ_{cl} is very small; and as the temperature goes down, Λ_{qu} grows monotonically, showing that cold processes have higher contributions from the coherences.

Table 1. Comparison between $\Lambda_{\rm qu}$, $\Lambda_{\rm cl}$, $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$.

<table>
<tr><th></th><th>$\Lambda_{\rm qu}$</th><th>$\Lambda_{\rm cl}$</th><th>$\Gamma_{\rm qu}$</th><th>$\Gamma_{\rm cl}$</th></tr>
<tr><td>Fluctuation theorem</td><td>✗</td><td>✓</td><td>✓</td><td>✓</td></tr>
<tr><td>Fluctuation theorem when $\Delta H \to 0$</td><td colspan="2">✓</td><td colspan="2">✓</td></tr>
<tr><td>Analytic when $\Delta H \to 0$ and low T</td><td colspan="2">✓</td><td colspan="2">✗</td></tr>
<tr><td>Resource-theoretic interpretation</td><td colspan="2">✗</td><td colspan="2">✓</td></tr>
<tr><td>Vanishing for commuting protocols</td><td>✓</td><td>—</td><td>✓</td><td>—</td></tr>
<tr><td>Dominant for highly coherent protocols</td><td>✓</td><td>—</td><td>✗</td><td>—</td></tr>
<tr><td>Dominant at low temperatures</td><td>✓</td><td>—</td><td>✗</td><td>—</td></tr>
</table>

The features discussed in figure 1 are not restricted to quenches. To illustrate that we show in figure 2 another qubit example, where the process is assumed to be cyclic, with $H_\tau = H_0 = \omega\sigma^z$, and the unitary is taken to be generated by an x-pulse with a duration τ; that is, $U = \mathrm{e}^{-\mathrm{i}\tau(H_0 + h_x\sigma^x)}$, where h_x is the pulse intensity. Figure 2 illustrates the results for $\omega = 1$, $h_x = 1.3$ and two choices of τ: in the upper panels $\tau = 0.4$ and in the lower panels $\tau = 1$. The results show that for (3) the behavior is always roughly the same, with $\Gamma_{\rm cl}$ always eventually dominating at low temperatures. Conversely, for the new splitting (10) a richer competition is observed. Depending on the parameters we may either have $\Lambda_{\rm qu}$ dominating, or $\Lambda_{\rm cl}$, or both.

As we will show in this paper, our new splitting (10) more accurately distinguishes which part of the entropy production is generated by a commuting or non-commuting drive. This provides a complementary approach to the standard splitting in equation (3), which instead describes how populations and coherences in the final state contribute to entropy production. On the other hand, we also note that $\Lambda_{\rm qu}$ and $\Lambda_{\rm cl}$ do not share some of the nice properties of $\Gamma_{\rm qu}$ and $\Gamma_{\rm cl}$. First, $\Lambda_{\rm qu}$ cannot be directly linked with a monotone for coherence or asymmetry [70]. Second, while $\Lambda_{\rm cl}$ always satisfies an individual fluctuation theorem, $\Lambda_{\rm qu}$ only does so in the case of infinitesimal quenches. Different properties of each splitting are highlighted in table 1. We also show that for infinitesimal quenches at high temperatures, both splittings coincide—see section 3.3.

To illustrate the usefulness of our results, we analyze our new splitting in two quantum many-body problems. Previous works have focused on the behavior of the statistics of work and entropy production Σ for quantum quenches [72–77], with emphasis in quantum phase transitions [73, 78–85]. Motivated by this, we analyze in section 4 the transverse field Ising model (TFIM), and discuss the behavior of (10) in the vicinity of the quantum critical point. This is complementary to the analysis put forth in [39], which studied equation (3). Then, in section 5, we consider a macrospin of varying size and focus on the full statistics of $\Lambda_{\rm qu}$ and $\Lambda_{\rm cl}$, including their probability distributions and their first four cumulants. We finish with conclusions and future perspectives in section 6.

2. Splittings of the entropy production

In this section we introduce our alternative splitting of the entropy production (equation (10)). We focus for now at the level of averages; the corresponding stochastic formulation will be presented in section 3.

Let $\mathcal{O}$ denote any Hermitian observable and decompose it as $\mathcal{O} = \sum_\alpha o_\alpha \Pi_\alpha$, where Π_α are projectors onto the subspaces with eigenvalues o_α. We define the dephasing operation

$$\mathbb{D}_{\mathcal{O}}(\bullet) = \sum_\alpha \Pi_\alpha \bullet \Pi_\alpha. \tag{12}$$

The rationale of the splitting equation (3) was to introduce an intermediate step, associated with the state $\mathbb{D}_{H_\tau}(\rho_\tau)$ (figure 1(b)). This represents the final state ρ_τ dephased in the eigenbasis of the final Hamiltonian. If the process generates coherences, this state will differ from the actual final state ρ_τ and their entropic distance will be precisely $\Gamma_{\rm qu}$ in equation (5).

For convenience, we introduce the non-equilibrium free energy, associated with the final Hamiltonian H_τ

$$F(\rho) = \mathrm{tr}\,\{H_\tau \rho\} - TS(\rho). \tag{13}$$

Non-equilibrium free energies depend on two parameters, H and ρ. However, in this paper, we will henceforth only need free energies defined with respect to H_τ, so we write it more simply as $F(\rho)$. In terms of F, the entropy production (2) can be written as

$$\Sigma = \beta\left\{F(\rho_\tau) - F(\rho_\tau^{\rm th})\right\}. \tag{14}$$

Similarly, one can also express Γ_{qu} and Γ_{cl} in terms of free energy differences. Since $\text{tr}\{H_\tau \mathbb{D}_{H_\tau}(\rho_\tau)\} = \text{tr}\{H_\tau \rho_\tau\}$, one finds that

$$\Gamma_{\text{qu}} = \beta \left\{F(\rho_\tau) - F\left(\mathbb{D}_{H_\tau}(\rho_\tau)\right)\right\}, \tag{15}$$

$$\Gamma_{\text{cl}} = \beta \left\{F\left(\mathbb{D}_{H_\tau}(\rho_\tau)\right) - F(\rho_\tau^{\text{th}})\right\}, \tag{16}$$

which clearly add up to Σ.

The splitting (3) uses $\mathbb{D}_{H_\tau}(\rho_\tau)$ as intermediate state. Our new splitting (10) follows a similar logic, but in reverse: instead of working with ρ_τ dephased in the basis of H_τ, we work with H_τ dephased in the basis of ρ_τ. More precisely, we define

$$\tilde{\rho}_\tau^{\text{th}} = \frac{\exp\{-\beta \mathbb{D}_{\rho_\tau}(H_\tau)\}}{\text{tr}\left(\exp\{-\beta \mathbb{D}_{\rho_\tau}(H_\tau)\}\right)}, \tag{17}$$

which is a thermal state based only on the incoherent part of H_τ, in the basis of ρ_τ (as a consequence, $[\tilde{\rho}_\tau^{\text{th}}, \rho_\tau] = 0$). With this in mind, we now define

$$\Lambda_{\text{cl}} = \beta \left\{F(\rho_\tau) - F(\tilde{\rho}_\tau^{\text{th}})\right\}, \tag{18}$$

$$\Lambda_{\text{qu}} = \beta \left\{F(\tilde{\rho}_\tau^{\text{th}}) - F(\rho_\tau^{\text{th}})\right\}, \tag{19}$$

which add up to Σ, as in equation (10). The first term, Λ_{cl}, compares the two *commuting* states ρ_τ and $\tilde{\rho}_\tau^{\text{th}}$ and is hence associated with their population mismatch. The nonnegativity of Λ_{cl} becomes evident by noting that it can also be written as

$$\Lambda_{\text{cl}} = S(\rho_\tau \| \tilde{\rho}_\tau^{\text{th}}). \tag{20}$$

The term Λ_{qu}, on the other hand, compares $\rho_\tau^{\text{th}} \propto \text{e}^{-\beta H_\tau}$ with $\tilde{\rho}_\tau^{\text{th}} \propto \text{e}^{-\beta \mathbb{D}_{\rho_\tau}(H_\tau)}$. Unlike Λ_{cl}, the contribution Λ_{qu} cannot be written as a relative entropy. In fact, written down explicitly, it reads

$$\Lambda_{\text{qu}} = \text{tr}\left\{\rho_\tau \left(\ln\, \tilde{\rho}_\tau^{\text{th}} - \ln\, \rho_\tau^{\text{th}}\right)\right\}. \tag{21}$$

Notwithstanding, as shown in appendix A, it turns out that Λ_{qu} is still non-negative, and zero if and only if $[\rho_\tau, H_\tau] = 0$.

Throughout this paper we will provide several additional justifications as to why the choices (18) and (19) are physically reasonable, starting in section 2.1. But before doing so, let us briefly revisit the minimal qubit model defined above equation (6). The process is a quench ($U = 1$), so $\rho_\tau = \rho_0^{\text{th}}$. Hence, all we need to do in order to compute Λ_{qu} is to dephase the final Hamiltonian $H_\tau = \omega(\sigma^z \cos\theta + \sigma^x \sin\theta)$ in the basis of ρ_0^{th}. Or, what is equivalent, in the basis of H_0. The result is thus simply $\mathbb{D}_{\rho_\tau}(H_\tau) = \omega\cos(\theta)\sigma^z$. Using this in (19) yields equation (11), which is the result plotted in figure 1(d) and discussed in section 1.

2.1. Infinitesimal quenches

The physics of the problem becomes particularly simpler in the case of infinitesimal quenches. We therefore now specialize the above results to this scenario. This will provide strong justifications in favor of the new splitting (10). Furthermore, in this limit the splitting (10) becomes equivalent to the one recently put forward in [30]. More precisely, in [30] the authors describe quasi-isothermal processes as a series of infinitesimal quenches, and in particular consider how Σ splits into a classical and quantum contribution. Focusing on a single infinitesimal quench, both approaches become directly comparable and, as we will show, agree with each other.

We thus analyze what happens if we take $U = 1$, and assume that H changes only by a small amount ΔH(i.e., we write $H_\tau = H_0 + \Delta H$). Since $U = 1$, the state of the system remains unchanged: $\rho_\tau = \rho_0^{\text{th}}$. Therefore, dephasing H_τ in the basis of ρ_τ is equivalent to dephasing in the basis of H_0:

$$\mathbb{D}_{\rho_\tau}(H_\tau) = \mathbb{D}_{\rho_0^{\text{th}}}(H_\tau) = \mathbb{D}_{H_0}(H_\tau). \tag{22}$$

Let us define the dephased (incoherent) and coherent parts of the perturbation ΔH, in the initial energy basis, $\Delta H^{\text{d}} = \mathbb{D}_{H_0}(\Delta H)$ and $\Delta H^{\text{c}} = H_\tau - \mathbb{D}_{H_0}(H_\tau)$. Then, following a procedure detailed in appendix B of reference [30], one may show that,

$$\tilde{\rho}_\tau^{\text{th}} = \rho_0^{\text{th}} - \beta \mathbb{J}_{\rho_0^{\text{th}}}[\Delta H^{\text{d}} - \langle \Delta H^{\text{d}} \rangle_0] + \mathcal{O}(\Delta H^2), \tag{23}$$

$$\rho_\tau^{\text{th}} = \rho_0^{\text{th}} - \beta \mathbb{J}_{\rho_0^{\text{th}}}[\Delta H - \langle \Delta H \rangle_0] + \mathcal{O}(\Delta H^2), \tag{24}$$

where $\langle \dots \rangle_0 = \mathrm{tr}\{\dots \rho_0^{\mathrm{th}}\}$ and $\mathbb{J}_\rho$ is a super-operator defined as

$$\mathbb{J}_\rho[\bullet] = \int_0^1 \rho^t \bullet \rho^{1-t}\, \mathrm{d}t. \tag{25}$$

We see that both ρ_τ^{th} and $\tilde{\rho}_\tau^{\mathrm{th}}$ can be expanded essentially in a power series in $\beta\Delta H$. Conversely, the same is not true for the state $\mathbb{D}_{H_\tau}(\rho_0^{\mathrm{th}})$ entering (16) and (15). In fact, one may show that to order ΔH [86]

$$\mathbb{D}_{H_0+\Delta H}(\rho_0^{\mathrm{th}}) = \rho_0^{\mathrm{th}} + \lim_{s\to\infty} \frac{i}{s} \int_0^s \mathrm{d}t \int_0^1 \mathrm{d}x\, t\, \mathrm{e}^{-\mathrm{i}xH_0t} [\rho_0^{\mathrm{th}}, \Delta H] \mathrm{e}^{\mathrm{i}xH_0t}. \tag{26}$$

Even though this is an expansion in ΔH, the dependence on β enters in a highly non-trivial way. This explains the non-analytic behavior of Γ_{cl} and Γ_{qu} at low temperatures, discussed in section 1.

Plugging (23) and (24) in equations (2), (20) and (21) leads, up to second order, to

$$\Sigma = \frac{\beta^2}{2} \mathrm{tr}\left\{\Delta H\, \mathbb{J}_{\rho_0^{\mathrm{th}}}[\Delta H - \langle \Delta H\rangle_0]\right\} = \Lambda_{\mathrm{cl}} + \Lambda_{\mathrm{qu}}, \tag{27}$$

$$\Lambda_{\mathrm{cl}} = \frac{\beta^2}{2} \mathrm{tr}\left\{\Delta H^{\mathrm{d}}\, \mathbb{J}_{\rho_0^{\mathrm{th}}}[\Delta H^{\mathrm{d}} - \langle \Delta H^{\mathrm{d}}\rangle_0]\right\}, \tag{28}$$

$$\Lambda_{\mathrm{qu}} = \frac{\beta^2}{2} \mathrm{tr}\left\{\Delta H^{\mathrm{c}}\, \mathbb{J}_{\rho_0^{\mathrm{th}}}[\Delta H^{\mathrm{c}}]\right\}, \tag{29}$$

where we used the fact that $\langle \Delta H\rangle_0 = \langle \Delta H^{\mathrm{d}}\rangle_0$. The interesting aspect of these results is that, within this infinitesimal quench limit, Λ_{cl} and Λ_{qu} are found to be related to Σ via the simple separation of the perturbation, , into a dephased and a coherent part. These results also coincide with the splitting proposed in [30].

An additional justification for the splitting (10) can be given in terms of the FDR. As shown in references [29, 30], equation (27) can also be written as

$$\Sigma = \frac{1}{2}\beta^2\, \mathrm{Var}_0[\Delta H] - \beta\mathcal{Q}, \tag{30}$$

where $\mathrm{Var}_0[\Delta H] = \langle \Delta H^2\rangle_0 - \langle \Delta H\rangle_0^2$, is the variance of the perturbation, and

$$\mathcal{Q} = \frac{\beta}{2} \int_0^1 \mathrm{d}y\, I^y(\rho_0^{\mathrm{th}}, \Delta H) \geqslant 0, \tag{31}$$

is a measure of quantum coherence, associated with the so-called Wigner–Yanase–Dyson skew information [87]

$$I^y(\varrho, X) = -\frac{1}{2} \mathrm{tr}\left\{[\varrho^y, X][\varrho^{1-y}, X]\right\}. \tag{32}$$

For incoherent processes one recovers the usual FDR $\Sigma = \frac{\beta^2}{2}\mathrm{Var}_0[\Delta H]$ [46]. But when the process is coherent, the FDR is broken by a term $-\beta\mathcal{Q}$. Repeating the same procedure for Λ_{cl} and Λ_{qu}, one readily finds that

$$\Lambda_{\mathrm{cl}} = \frac{\beta^2}{2}\mathrm{Var}_0[\Delta H^{\mathrm{d}}], \qquad \Lambda_{\mathrm{qu}} = \frac{\beta^2}{2}\mathrm{Var}_0[\Delta H^{\mathrm{c}}] - \beta\mathcal{Q}. \tag{33}$$

Whence, Λ_{cl} always satisfies a standard FDR, and all violations are associated to Λ_{qu}. This provides additional justification as to why Λ_{qu} is referred to as a quantum contribution.

In the case of high temperatures ($\beta \to 0$), one may show that $\mathcal{Q}\mathcal{O}(\beta^3)$. Moreover, the state entering the variances in equation (33) can be replaced with the maximally mixed state $\mathbb{I}/d$. As a consequence, we find that to leading order in β,

$$\Lambda_{\mathrm{cl}} = \frac{\beta^2}{2}\mathrm{Var}_{\mathbb{I}/d}[\Delta H^{\mathrm{d}}], \qquad \Lambda_{\mathrm{qu}} = \frac{\beta^2}{2}\mathrm{Var}_{\mathbb{I}/d}[\Delta H^{\mathrm{c}}]. \tag{34}$$

Both contributions are thus found to scale as β^2 in this limit, which agrees with the observations in figures 1(c) and (d). However, their relative contribution will be determined by the variance of ΔH^{d} and ΔH^{c} in the maximally mixed state; hence, which term will be dominant will depend on the details of the process (either a commuting or a non-commuting drive). This is also expected to remain true for general drives.

3. Stochastic trajectories

We now discuss how to formulate the splittings (3) and (10) at the level of stochastic trajectories, based on a standard two-point measurement (TPM) scheme [48]. Since $\Sigma = \beta\left(\langle W\rangle - \Delta F\right)$, the statistics of Σ can be obtained solely from measurements in the eigenbasis of the initial and final Hamiltonians. As first shown in [38], a major advantage of the original splitting (3) is that this remains true when assessing the individual contributions $\Gamma_{\rm cl}$ and $\Gamma_{\rm qu}$; that is, no additional measurements are necessary. As we will now show, the same is also true for $\Lambda_{\rm cl}$ and $\Lambda_{\rm qu}$ (equation (10)). This means that both splittings can be assessed, at the stochastic level, with the same amount of information as a standard TPM.

Irrespective of the splitting one is interested in, the protocol may therefore be described as follows. Initially the system is in the thermal state $\rho_0^{\rm th}$, associated with the Hamiltonian $H_0 = \sum_i \epsilon_i^0 |i_0\rangle\langle i_0|$. The first measurement is performed in the basis $|i_0\rangle$, which occurs with probability $p_i^0 = {\rm e}^{-\beta\epsilon_i^0}/Z_0$. Conversely, the second measurement is performed at time τ, after the map (1), and in the eigenbasis of the final Hamiltonian $H_\tau = \sum_j \epsilon_j^\tau |j_\tau\rangle\langle j_\tau|$. The bases $\{|i_0\rangle\}$ and $\{|j_\tau\rangle\}$ are, in general, not compatible.

The conditional probability of finding the system in $|j_\tau\rangle$ given that it was initially in $|i_0\rangle$ is $|\langle j_\tau|U|i_0\rangle|^2$. The probability associated with the forward protocol $|i_0\rangle \to |j_\tau\rangle$ is thus $\mathcal{P}_{\rm F}[i,j] = |\langle j_\tau|U|i_0\rangle|^2 p_i^0$. The dynamics is defined as being incoherent when $|\langle j_\tau|U|i_0\rangle|^2 = \delta_{i,j}$, which means U is not able to generate transitions between states of the initial and final Hamiltonians. Similarly, in the backward protocol the system starts in $\rho_\tau^{\rm th}$ and one measures first in the basis of H_τ, yielding $|j_\tau\rangle$ with probability $p_j^\tau = {\rm e}^{-\beta\epsilon_j^\tau}/Z_\tau$. The time-reversed unitary $U^\dagger$ is then applied, after which one measures in the basis $|i_0\rangle$ of H_0. This yields the backward distribution $\mathcal{P}_{\rm B}[i,j] = |\langle i_0|U^\dagger|j_\tau\rangle|^2 p_j^\tau$.

The entropy production associated to the trajectory $|i_0\rangle \to |j_\tau\rangle$ is now defined as usual:

$$\sigma[i,j] = \ln \frac{\mathcal{P}_{\rm F}[i,j]}{\mathcal{P}_{\rm B}[i,j]} = \ln p_i^0/p_j^\tau. \tag{35}$$

The second equality follows from the fact that $|\langle i_0|\, U^\dagger|j_\tau\rangle|^2 = |\langle j_\tau|\, U|i_0\rangle|^2$. As a consequence, $\sigma[i,j]$ depends only on the equilibrium populations p_i^0 and p_j^τ, associated with the initial and final Hamiltonians. As can be readily verified, $\langle\sigma[i,j]\rangle = \sum_{i,j}\sigma[i,j]\mathcal{P}_{\rm F}[i,j] = \Sigma$, returns precisely equation (2). In addition, $\sigma[i,j]$ also satisfies an integral fluctuation theorem $\langle {\rm e}^{-\sigma}\rangle = 1$ (see equation (43) for more details).

3.1. Stochastic definitions for the splittings (3) and (10)

Following [38], we now define stochastic quantities associated to $\Gamma_{\rm cl}$ and $\Gamma_{\rm qu}$. In order to do that, we first write the dephased state $\mathbb{D}_{H_\tau}(\rho_\tau)$ as $\mathbb{D}_{H_\tau}(\rho_\tau) = \sum_j q_j^\tau |j_\tau\rangle\langle j_\tau|$, where

$$q_j^\tau = \langle j_\tau|\rho_\tau|j_\tau\rangle = \sum_i |\langle j_\tau|U|i_0\rangle|^2 p_i^0. \tag{36}$$

In passing, we note that $q_j^\tau = \sum_i \mathcal{P}_{\rm F}[i,j]$, so q_j^τ can also be interpreted as the marginal distribution of the final measurement. As shown in [38], we may now define

$$\gamma_{\rm cl}[i,j] = \ln q_j^\tau/p_j^\tau, \tag{37}$$

$$\gamma_{\rm qu}[i,j] = \ln p_i^0/q_j^\tau. \tag{38}$$

Clearly $\gamma_{\rm cl}[i,j] + \gamma_{\rm qu}[i,j] = \sigma[i,j]$, which is the stochastic analog of (3). Moreover, $\langle\gamma_{\rm cl}[i,j]\rangle = \Gamma_{\rm cl}$ and $\langle\gamma_{\rm qu}[i,j]\rangle = \Gamma_{\rm qu}$.

Similarly, we construct stochastic quantities for the new quantities $\Lambda_{\rm cl}$ and $\Lambda_{\rm qu}$ in equation (10). The central object now is the thermal state $\tilde{\rho}_\tau^{\rm th}$, defined in equation (17) and associated with the Hamiltonian $\mathbb{D}_{\rho_\tau}(H_\tau)$. Since the system evolves unitarily, $\rho_\tau = U\rho_0^{\rm th}U^\dagger = \sum_i p_i^0|\psi_i\rangle\langle\psi_i|$, where $|\psi_i\rangle = U|i_0\rangle$. That is, ρ_τ has the same populations p_i^0 as $\rho_0^{\rm th}$, but a rotated eigenbasis. Based on this, we can now write equation (17) as

$$\tilde{\rho}_\tau^{\rm th} = \sum_i \tilde{p}_i^\tau |\psi_i\rangle\langle\psi_i|, \qquad \tilde{p}_i^\tau = {\rm e}^{-\beta(\tilde{\epsilon}_i^\tau - F(\tilde{\rho}_\tau^{\rm th}))}, \tag{39}$$

where $\tilde{\epsilon}_i^\tau = \langle\psi_i|H_\tau|\psi_i\rangle$ are the eigenvalues of the dephased Hamiltonian $\mathbb{D}_{\rho_\tau}(H_\tau)$ and $F(\tilde{\rho}_\tau^{\rm th})$ is the same free energy as that appearing in equation (18). We then define

$$\lambda_{\rm cl}[i,j] = \ln p_i^0/\tilde{p}_i^\tau, \tag{40}$$

$$\lambda_{\rm qu}[i,j] = \ln \tilde{p}_i^\tau/p_j^\tau. \tag{41}$$

These quantities satisfy $\lambda_{\rm cl}[i,j] + \lambda_{\rm qu}[i,j] = \sigma[i,j]$, as well as $\langle\lambda_{\rm cl}[i,j]\rangle = \Lambda_{\rm cl}$ and $\langle\lambda_{\rm qu}[i,j]\rangle = \Lambda_{\rm qu}$.

3.2. Cumulant generating functions

For all stochastic quantities in the previous section, we can define their corresponding probability distributions or, what is more convenient, their cumulant generating functions (CGFs). For instance, from (35) we define

$$P(\sigma) = \sum_{i,j} \mathcal{P}_{\mathrm{F}}[i,j]\delta(\sigma - \sigma[i,j]), \tag{42}$$

from which we may compute the CGF, $K_\sigma(v) = \ln\langle e^{-v\sigma}\rangle$. With some manipulations, this can be neatly written as [53, 88]

$$\begin{aligned} K_\sigma(v) &= \ln\,\mathrm{tr}\left\{(\rho_\tau^{\mathrm{th}})^v(\rho_\tau)^{1-v}\right\} \\ &= (v-1)S_v(\rho_\tau^{\mathrm{th}}\|\rho_\tau). \end{aligned} \tag{43}$$

The second equality expresses the CGF in terms of the Rényi divergences $S_v(\rho\|\sigma) = (v-1)^{-1}\ln\,\mathrm{tr}\,\left\{\rho^v\sigma^{1-v}\right\}$, which may be convenient in some situations. Setting $v = 1$ yields $K_\sigma(1) = 0$, which is the integral fluctuation theorem [40, 41]

$$\left\langle \mathrm{e}^{-\sigma}\right\rangle = 1. \tag{44}$$

In addition, from the CGF we may compute any cumulant of σ as

$$\kappa_n(\sigma) = (-1)^n \frac{\partial^n K_\sigma}{\partial v^n}\bigg|_{v=0}, \tag{45}$$

with $\kappa_1(\sigma) = \Sigma$ being the mean in equation (2).

We may also compute the joint CGF of γ_{cl} and γ_{qu}, defined as $K_{\gamma_{\mathrm{cl}},\gamma_{\mathrm{qu}}}(v,u) = \ln\langle \mathrm{e}^{-v\gamma_{\mathrm{cl}}-u\gamma_{\mathrm{qu}}}\rangle$. With similar manipulations, it may be written as

$$K_{\gamma_{\mathrm{cl}},\gamma_{\mathrm{qu}}}(v,u) = \ln\,\mathrm{tr}\left\{(\rho_\tau^{\mathrm{th}})^v\left[\mathbb{D}_{H_\tau}(\rho_\tau)\right]^{u-v}(\rho_\tau)^{1-u}\right\}. \tag{46}$$

The CGF of $\sigma = \gamma_{\mathrm{cl}} + \gamma_{\mathrm{qu}}$, equation (43), is recovered by setting $u = v$; that is $K_\sigma(v) = K_{\gamma_{\mathrm{cl}},\gamma_{\mathrm{qu}}}(v,v)$. The reduced CGFs of γ_{cl} and γ_{qu} are found by setting $u = 0$ or $v = 0$, respectively:

$$K_{\gamma_{\mathrm{cl}}}(v) = \ln\,\mathrm{tr}\left\{(\rho_\tau^{\mathrm{th}})^v\left[\mathbb{D}_{H_\tau}(\rho_\tau)\right]^{-v}\rho_\tau\right\}, \tag{47}$$

$$K_{\gamma_{\mathrm{qu}}}(u) = \ln\,\mathrm{tr}\left\{\left[\mathbb{D}_{H_\tau}(\rho_\tau)\right]^u(\rho_\tau)^{1-u}\right\}. \tag{48}$$

From this one may verify that γ_{cl} and γ_{qu} individually satisfy fluctuation theorems

$$\langle \mathrm{e}^{-\gamma_{\mathrm{cl}}}\rangle = \langle \mathrm{e}^{-\gamma_{\mathrm{qu}}}\rangle = 1. \tag{49}$$

Note also that, except in certain particular cases, equation (46) cannot be written as a sum of two CGFs, which means γ_{cl} and γ_{qu} are statistically dependent.

Similarly, we compute the joint CGF of λ_{cl} and λ_{qu}, defined as $K_{\lambda_{\mathrm{cl}},\lambda_{\mathrm{qu}}}(v,u) = \ln\langle \mathrm{e}^{-v\lambda_{\mathrm{cl}}-u\lambda_{\mathrm{qu}}}\rangle$. It reads

$$K_{\lambda_{\mathrm{cl}},\lambda_{\mathrm{qu}}}(v,u) = \ln\,\mathrm{tr}\left\{(\rho_\tau^{\mathrm{th}})^u(\tilde{\rho}_\tau^{\mathrm{th}})^{v-u}(\rho_\tau)^{1-v}\right\}. \tag{50}$$

The reduced CGFs of λ_{cl} and λ_{qu} are again found by setting $u = 0$ an $v = 0$,

$$K_{\lambda_{\mathrm{cl}}}(v) = \ln\,\mathrm{tr}\left\{(\tilde{\rho}_\tau^{\mathrm{th}})^v(\rho_\tau)^{1-v}\right\} \tag{51}$$

$$K_{\lambda_{\mathrm{qu}}}(u) = \ln\,\mathrm{tr}\left\{(\rho_\tau^{\mathrm{th}})^u(\tilde{\rho}_\tau^{\mathrm{th}})^{-u}\rho_\tau\right\}. \tag{52}$$

Once again, λ_{cl} and λ_{qu} are, in general, statistically dependent. Equation (51) implies that λ_{cl} satisfies a fluctuation theorem,

$$\langle \mathrm{e}^{-\lambda_{\mathrm{cl}}}\rangle = 1. \tag{53}$$

But the same is not true for λ_{qu}. Notwithstanding, as we will show, this property is recovered in the limit of infinitesimal quenches.

3.3. Infinitesimal quenches

As before, we now specialize the above expressions to the case of infinitesimal quenches. Since $U = 1$, the path probability reduces to $\mathcal{P}_{\mathrm{F}}[i,j] = |\langle j_\tau|i_0\rangle|^2 p_i^0$. Moreover, since ΔH is assumed to be small, $|j_\tau\rangle$ will be close to $|i_0\rangle$ and ϵ_j^τ will be close to ϵ_j^0. For concreteness, we assume that the spectra of H_0 is non-degenerate. Standard perturbation theory then yields, to order ΔH^2,

$$\epsilon_j^\tau = \epsilon_j^0 + \Delta H_{jj} + E_j^{(2)}, \tag{54}$$

where $\Delta H_{ij} = \langle i_0\,|\Delta H|\,j_0\rangle$ and $E_j^{(2)} = \sum_{l\neq j}|\Delta H_{jl}|^2/(\epsilon_j^0 - \epsilon_l^0)$. Note that if we split $\Delta H = \Delta H^{\mathrm{d}} + \Delta H^{\mathrm{c}}$, the first non-trivial contribution of the former is ΔH_{jj}, while that of the latter is $E_j^{(2)}$. Similarly, the eigenstates $|j_\tau\rangle$ of the final Hamiltonian can be expanded as

$$|\langle j_\tau|i_0\rangle|^2 = \frac{|\Delta H_{ij}|^2}{(\epsilon_j^0 - \epsilon_i^0)^2}, \tag{55}$$

for $i \neq j$ while $|\langle j_\tau j_0\rangle|^2 = 1 - \sum_{\ell\neq j}|\langle j_\tau \ell_0\rangle|^2$.

Using this, we can expand all relevant probabilities $\{\tilde{p}_j^\tau\}$, $\{q_j^\tau\}$ and $\{p_j^\tau\}$ entering in the stochastic definitions (35), (37), (38), (40) and (41):

$$\tilde{p}_i^\tau = p_i^0(1 - \tilde{f}_i), \tag{56}$$

$$p_j^\tau = p_j^0(1 - f_j), \tag{57}$$

$$q_j^\tau = p_j^0(1 - s_j) \tag{58}$$

where

$$\tilde{f}_j = \beta(\Delta H_{jj} - \langle\Delta H^{\mathrm{d}}\rangle_0) + \beta^2\langle\Delta H^{\mathrm{d}}\rangle_0(\Delta H_{jj} - \langle\Delta H^{\mathrm{d}}\rangle_0) + \frac{1}{2}\beta^2[\Delta H_{jj}^2 - \langle(\Delta H^{\mathrm{d}})^2\rangle_0], \tag{59}$$

$$f_j = \tilde{f}_j + \beta[E_j^{(2)} - \langle E^{(2)}\rangle_0], \tag{60}$$

$$s_j = \sum_{\ell\neq j}\frac{1 - \mathrm{e}^{-\beta(\epsilon_\ell^0 - \epsilon_j^0)}}{(\epsilon_j^0 - \epsilon_\ell^0)^2}|\Delta H_{\ell j}|^2, \tag{61}$$

and $\langle E^{(2)}\rangle_0 = \sum_i p_i^0 E_i^{(2)}$. Note how $\tilde{f}_j$ depends only on the diagonal part of the perturbation, ΔH^{d}. This is in line with equation (23). Conversely, f_j, which is associated with the full probabilities p_j^τ, also has an additional contribution from $E_j^{(2)}$, which is the term associated to coherences.

Inserting equations (56) and (57) into equations (35), (37), (38), (40) and (41) we obtain,

$$\sigma[i,j] = \ln p_i^0/p_j^0 - \ln(1 - f_j), \tag{62}$$

$$\gamma_{\mathrm{cl}}[i,j] = \ln(1 - s_j) - \ln(1 - f_j), \tag{63}$$

$$\gamma_{\mathrm{qu}}[i,j] = \ln p_i^0/p_j^0 - \ln(1 - s_j), \tag{64}$$

$$\lambda_{\mathrm{cl}}[i,j] = -\ln(1 - \tilde{f}_i), \tag{65}$$

$$\lambda_{\mathrm{qu}}[i,j] = \ln p_i^0/p_j^0 + \ln(1 - \tilde{f}_i) - \ln(1 - f_j). \tag{66}$$

We are now in the position to discuss the analyticity of the entropy production and its splittings, at the stochastic level. A series expansion of $\ln(1 - x)$ is convergent only for $|x| < 1$. Thus, since $p_i^0/p_j^0 = \mathrm{e}^{-\beta(\epsilon_i^0 - \epsilon_j^0)}$ is a well behaved function, the analyticity of σ, λ_{cl} and λ_{qu} are all conditioned on having $|f_j| < 1$ and $|\tilde{f}_j| < 1$, which is satisfied if $\beta|\Delta H_{ij}| \lesssim 1$, as intuitively expected. Thus, the quantities of our new proposed splitting (10) behave, from an analytical point of view, similarly to the full entropy production.

On the other hand, equations (63) and (64) rely on $|s_j| < 1$. Each s_j in (61) is a weighted contribution from all energies ϵ_ℓ^0, with $\ell \neq j$. At low temperatures, those energies for which $\epsilon_\ell^0 < \epsilon_j^0$ will yield an exponentially large contribution $1 - \mathrm{e}^{-\beta(\epsilon_\ell^0 - \epsilon_j^0)}$ to the sum. Conversely, those with $\epsilon_\ell^0 > \epsilon_j^0$ will contribute negligibly. The expansion is thus not in powers of $\beta\Delta H$, which is also visible from (26). Instead, it is an expansion in powers of ΔH, with coefficients that depend exponentially in β. Violating the condition $|s_j| < 1$ is thus exponentially easier at low temperatures. These results show that the shortcomings illustrated in section 1, are in fact absolutely general.

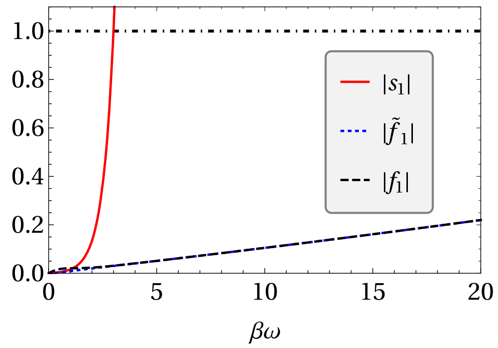

Figure 3. Analyticity of thermodynamic quantities at the stochastic level, for the minimal qubit model of figure 1. The figure compares equation (67) (red-solid), (68) (blue-dashed) and (69), as a function of $\beta\omega$ for $\theta = 0.1$. The condition $|s_1| < 1$, for the analyticity of $\gamma_{\rm cl}$ and $\gamma_{\rm qu}$, is quickly violated. For σ, $\lambda_{\rm cl}$ and $\lambda_{\rm qu}$, on the other hand, the conditions $|f_1| < 1$ and $|\tilde{f}_1| < 1$ are satisfied for a much larger range of temperatures.

To better illustrate this discussion, we revisit the minimal qubit model example treated in section 1. Initially the system's Hamiltonian is $H_0 = \omega\sigma^z$, and after an instantaneous quench it becomes $H_1 = \omega(\sigma^z \cos\theta + \sigma^x \sin\theta)$, where we consider θ to be small. The problematic term in this case is s_1 (equation (61)), which is given by

$$s_1 = \left(1 - \mathrm{e}^{2\beta\omega}\right)\left(\frac{\sin\,\theta}{2}\right)^2. \tag{67}$$

In comparison, we have

$$\tilde{f}_1 = 2\beta\omega\,\sin^2(\theta/2)(1 + \tanh\,\beta\omega)\left[1 + 2\beta\omega\,\sin^2(\theta/2)\,\tanh\,\beta\omega\right] \tag{68}$$

$$f_1 = \tilde{f}_1 + \frac{1}{2}\beta\omega\,\sin\,\theta(1 - \tanh\,\beta\omega). \tag{69}$$

In figure 3 we plot equations (67)–(69) as a function of $\beta\omega$, for $\theta = 0.1$. The condition for analyticity of $\gamma_{\rm cl}$ and $\gamma_{\rm qu}$ in this case, $|s_1| < 1$, is rapidly violated with increasing $\beta\omega$. For σ, $\lambda_{\rm cl}$ and $\lambda_{\rm qu}$, instead, $|f_1| < 1$ and $|\tilde{f}_1| < 1$ for a much larger range of temperatures. It is also interesting to note that, at low temperatures, the excited state thermal probabilities p_1^0, $\tilde{p}_1^\tau$ and $p_1^\tau \propto \mathrm{e}^{-\beta\omega}$ all tend to zero exponentially as $e^{-\beta\omega}$. Conversely, q_1^τ tends to $\sin^2(\frac{\theta}{2})$. This corroborates the use of thermal states, such as (17), as intermediate states for the splitting of Σ, as it ensures that the resulting functions are analytic.

We now move on to discuss what becomes of the CGFs of section 3.2 in the infinitesimal quench regime. We start with the CGFs of σ, $\lambda_{\rm cl}$ and $\lambda_{\rm qu}$ in equations (43), (50)–(52). Using equations (62), (65) and (66), together with the path probability $\mathcal{P}_{\rm F}[i,j] = p_i^0|\langle j_\tau|i_0\rangle|^2$, we find to order $\varDelta H^2$, that

$$K_{\lambda_{\rm cl},\lambda_{\rm qu}}(v,u) = K_{\lambda_{\rm cl}}(v) + K_{\lambda_{\rm qu}}(u), \tag{70}$$

where

$$K_{\lambda_{\rm cl}}(v) = \frac{\beta^2}{2}(v^2 - v)\mathrm{Var}_0[\Delta H^{\rm d}], \tag{71}$$

$$K_{\lambda_{\rm qu}}(u) = \frac{\beta^2}{2}(u^2 - u)\mathrm{Var}_0[\Delta H^{\rm c}] + \frac{\beta^2}{2}\int_0^u \mathrm{d}x\int_x^{1-x}\mathrm{d}y I^y(\rho_0^{\rm th}, \Delta H^{\rm c}). \tag{72}$$

These results are quite illuminating. Equation (70) implies $\lambda_{\rm cl}$ and $\lambda_{\rm qu}$ become *statistically independent* in this limit. Moreover, since $K_\sigma(v) = K_{\lambda_{\rm cl},\lambda_{\rm qu}}(v,v)$, we now find that

$$K_\sigma(v) = K_{\lambda_{\rm cl}}(v) + K_{\lambda_{\rm qu}}(v). \tag{73}$$

This means that all cumulants of σ can be split as a sum of the cumulants of $\lambda_{\rm cl}$ and $\lambda_{\rm qu}$: $\kappa_n(\sigma) = \kappa_n(\lambda_{\rm cl}) + \kappa_n(\lambda_{\rm qu})$. For all intents and purposes, the two channels of entropy production, $\varLambda_{\rm cl}$ and $\varLambda_{\rm qu}$, may thus be regarded as stemming from *independent* processes: $\varLambda_{\rm cl}$ gives the entropy production associated with a quench from $H_0 \to \mathbb{D}_{H_0}(H_\tau)$, while $\varLambda_{\rm qu}$ is associated with a second quench from $\mathbb{D}_{H_0}(H_\tau) \to H_\tau$. We also note that, from equation (73), it can now be seen that in this limit $\lambda_{\rm qu}$ satisfies an integral fluctuation theorem: $\langle \mathrm{e}^{-\lambda_{\rm qu}}\rangle = 1$.

In contrast, for the original splitting (3), we have

$$K_{\gamma_{\rm cl},\gamma_{\rm qu}}(v,u)=\ln\sum_{i,j}(p_j^0/p_i^0)^u(1-s_j)^{u-v}(1-f_j)^v p_i^0|\langle j_\tau|i_0\rangle|^2. \tag{74}$$

Once again, a series expansion of $(1-s_j)^{-x}$ is convergent only if $|s_j|<1$. However, if $|s_j|<1$ is satisfied, which happens for sufficiently high temperatures, one may show that, to order ΔH^2, we can also split $K_{\gamma_{\rm cl},\gamma_{\rm qu}}(v,u)=K_{\gamma_{\rm cl}}(v)+K_{\gamma_{\rm qu}}(u)$. And, what is more important, $K_{\gamma_{\rm cl}}$ and $K_{\gamma_{\rm qu}}$ coincide with $K_{\lambda_{\rm cl}}$ and $K_{\lambda_{\rm qu}}$ respectively. Whence, at sufficiently high temperatures the splittings (3) and (10) coincide, even at the stochastic level. However, this is only true for infinitesimal quenches. Otherwise, the two splittings may differ, even at high temperatures.

4. Transverse field Ising model

We now turn to discuss applications of our framework. We begin with the behavior of the splitting (10) in the one-dimensional TFIM, which is a prototypical model of a quantum critical system. An analysis of (3) for the same model was recently made in [39]. Here we aim to contrast those results with our new splitting (10). We thus restrict the analysis to quench protocols, and study the problem at the level of the averages $\Lambda_{\rm cl}$ and $\Lambda_{\rm qu}$ (equations (18) and (19)). Non-trivial unitaries and higher order statistics will be studied in section 5, for a different model.

We begin by introducing the model and delineating the steps to compute $\Lambda_{\rm cl}$ and $\Lambda_{\rm qu}$. To make the paper self-consistent, some additional details are provided in appendices B and C. Consider a linear chain of N spins, each described by Pauli operators $\sigma_j^{x,y,z}$ and interacting via the Hamiltonian

$$H(g)=-\sum_{j=1}^{N}\left(J\sigma_j^x\sigma_{j+1}^x+g\sigma_j^z\right), \tag{75}$$

where g is the applied magnetic field and J is the coupling strength, which we henceforth set to unity ($J=1$). We consider periodic boundary conditions, $\sigma_{N+1}^{\alpha}=\sigma_1^{\alpha}$. This model presents critical points at $g=\pm1$, where the system changes from a ferromagnetic phase, for $|g|<1$, to a paramagnetic phase, for $|g|>1$.

After a series of transformations (see appendix B) this Hamiltonian can be written as

$$H(g)=\sum_k\epsilon_k(g)\left(2\eta_k^{\dagger}\eta_k-1\right), \tag{76}$$

where $\{\eta_k\}$ are fermionic operators and

$$\epsilon_k(g)=\sqrt{(g-\cos k)^2+\sin^2 k}, \tag{77}$$

are the single-particle energies. Here, $\mathrm{k}=\pm(2\mathrm{n}+1)\frac{\pi}{\mathrm{N}}$, with $n=0,1,\dots,N/2-1$, denotes the system's quasi-momenta. We consider that the system initially has a transverse field g_0 and is prepared in the thermal state $\rho_0^{\rm th}=\mathrm{e}^{-\beta H_0}/Z_0$. The full expression for $\rho_0^{\rm th}$ can be found in appendix C. Due to the structure of (76), it can be decomposed as a product over individual modes, which greatly facilitates the calculation of all thermodynamic quantities.

The system is then decoupled from the reservoir and undergoes an instantaneous quench, where the field is changed to $g_\tau=g_0+\delta g$. Since the quench is instantaneous, the state of the system remains the same, but its Hamiltonian changes, from H_0 to $H_\tau=H_0+\Delta H$, where $\Delta H=-\delta g\sum_j\sigma_j^z$. Full details on the computation of $\Lambda_{\rm cl}$ and $\Lambda_{\rm qu}$ are provided in appendices B and C. The overall contributions of the diagonal vs off-diagonal is described by the Bogoliubov angle $\cos\,\theta_k=(g_0-\cos\,k)/\epsilon_k^0$ and $\sin\,\theta_k=\sin\,k/\epsilon_k^0$. And the state (17), associated with the dephased final Hamiltonian, is described by the modified energies $\tilde{\epsilon}_k^{\tau}=\epsilon_k^0+\delta g\,\cos\,\theta_k$.

We are interested in the thermodynamic limit (N very large), where k sums can be converted to integrals and all quantities become extensive in N. In this case, we ultimately find that

$$\Lambda_{\rm cl}=N\int_0^{\pi}\frac{\mathrm{d}k}{2\pi}\,2\left\{\ln\left[\frac{\cosh\left(\beta\tilde{\epsilon}_k^{\tau}\right)}{\cosh\left(\beta\epsilon_k^0\right)}\right]+\beta\left(\epsilon_k^0-\tilde{\epsilon}_k^{\tau}\right)\,\tanh\left(\beta\epsilon_k^0\right)\right\}, \tag{78}$$

and

$$\Lambda_{\rm qu}=N\int_0^{\pi}\frac{\mathrm{d}k}{2\pi}\,2\,\ln\left[\frac{\cosh\left(\beta\epsilon_k^{\tau}\right)}{\cosh\left(\beta\tilde{\epsilon}_k^{\tau}\right)}\right]. \tag{79}$$

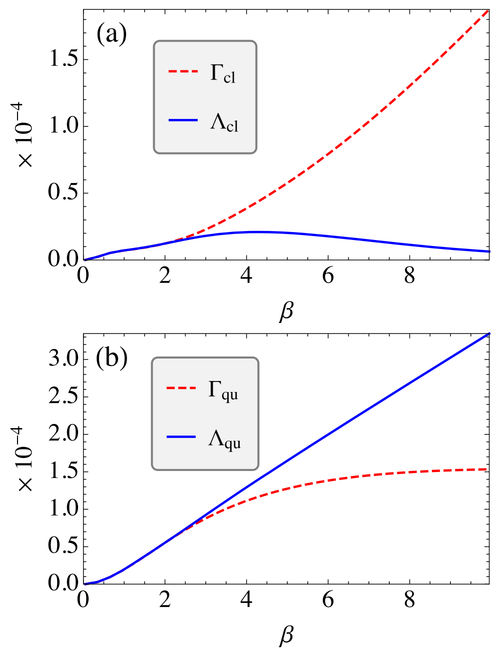

Figure 4. Comparison between (a) Γ_{cl} and Λ_{cl}, and (b) Γ_{qu} and Λ_{qu}, for the TFIM, as a function of β, with $g_0 = 0.75$ and $\delta g = 0.01$.

Adding both contributions recovers the full entropy production Σ, which was computed in [39, 73, 83] and reads

$$\Sigma = N\int_0^{\pi}\frac{\mathrm{d}k}{2\pi}\,2\left\{\ln\left[\frac{\cosh\left(\beta\epsilon_k^{\tau}\right)}{\cosh\left(\beta\epsilon_k^{0}\right)}\right] + \beta\left(\epsilon_k^0 - \tilde{\epsilon}_k^{\tau}\right)\,\tanh\left(\beta\epsilon_k^0\right)\right\}. \tag{80}$$

For comparison, in appendix D we also present the formulas for the splitting (3), which were developed in [39]. We also mention, in passing, that equations (78) and (79) are not perturbative in the quench magnitude. That is, they hold for arbitrary quenches δg. The only assumption is that $U = 1$. For completeness, their behavior in the infinitesimal case is presented in equations (C13) and (C14).

Figure 4 compares the two splittings (3) and (10) as a function of β, with fixed $g_0 = 0.75$ (outside criticality) and $\delta g = 0.01$. At high temperatures, one clearly sees how both splittings coincide. This corresponds to the region of parameters where Γ_{cl} and Γ_{qu} are analytic. But as the system is cooled, they eventually begin to differ. In particular, Γ_{cl} tends to grow linearly with β, while Λ_{cl} tends to zero. For the quantum components the opposite is observed: Γ_{qu} tends to saturate while Λ_{qu} tends to grow. Thus, at very low temperatures Λ_{qu} becomes the dominant contribution in (10), while becomes the dominant one in (3).

Next we turn to the behavior near criticality. In figure 5 we plot Γ_{cl}, Γ_{qu}, Λ_{cl} and Λ_{qu} as a function of the initial field g_0, for different values of β (focusing on low temperatures) and fixed quench magnitude of $\delta g = 0.01$. The full entropy production Σ behaves similarly to Λ_{qu} in figure 5(a); for any finite T it presents a peak at $g_0 = 1$, which eventually tends to a divergence as $\beta \to \infty$. As is clear by comparing figures 5(a) and (b), the dominant contribution to the splitting (10) is Λ_{qu}. Moreover, Λ_{qu} is found to always grow (and eventually diverge) with β at $g_0 = 1$, while Λ_{cl} sharply drops to zero. Conversely, for the splitting (3), we find in figures 5(c) and (d) that the dominant contribution is instead that of the populations Γ_{cl}. Crucially, we find that in this case Γ_{qu} remains finite as $\beta \to \infty$, while Γ_{cl} diverges [39]. We also call attention to the non-monotonic dependence on β, of the quantities in figure 5(c). This is an artifact of the fact that Γ_{qu} is scaled by β. The quantity Γ_{qu} itself is monotonic, but its behavior changes from β^2 at high temperatures, to in low temperatures [39].

As highlighted in [39], the entropy production in this limit results entirely from the changes in occupations, i.e. creation/annihilation of particles, in the modes $\pm k$, when the quench is performed. This enters in Γ_{cl} as a population mismatch between the initial and final equilibrium states. Conversely, in the split (10), it enters in Λ_{qu} as resulting from the rotating energy basis. On the other hand, Λ_{cl} only quantifies the contribution to the entropy production resulting from a change in the energy levels given by $\tilde{\epsilon}_k^{\tau} - \epsilon_k^0 = \delta g\,\cos\,\theta_k$. In the low temperature limit, only the ground and low lying excited states are relevant, and close to the critical point $g_0 = 1$, the latter corresponds to creating excitations with momentum $k \to 0$; but one can easily show that at $g_0 = 1$, $\cos\theta_k = |\sin(k/2)|$, which goes to zero when $k \to 0$. This explains

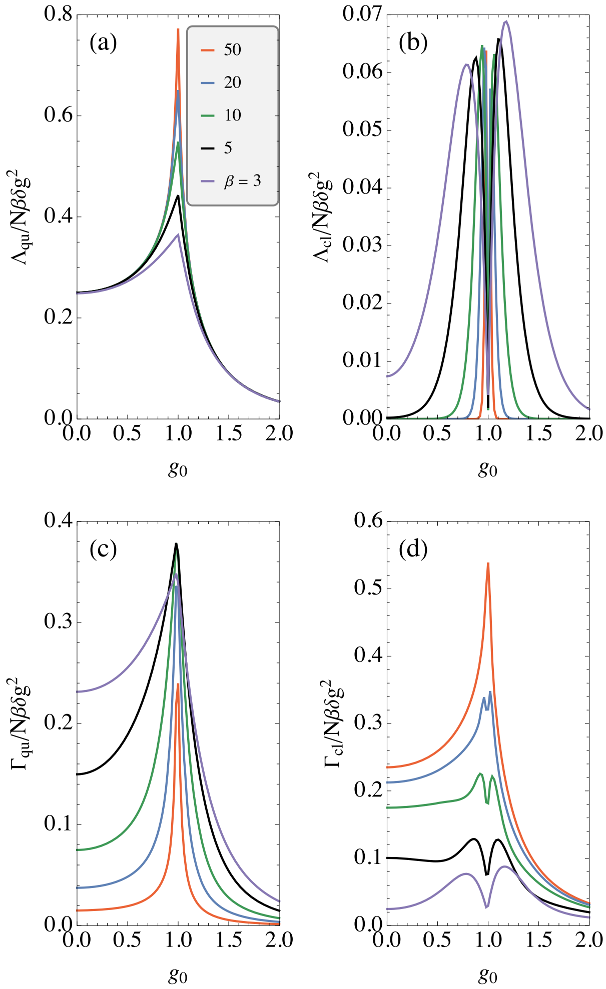

Figure 5. Λ_{qu}, Λ_{cl}, Γ_{qu} and Γ_{cl} for the TFIM as a function of g_0, for different values of β (in the low temperature regime) and $\delta g = 0.01$. All quantities are scaled by $\beta\delta g^2$.

the drop in Λ_{cl} at this point. Overall, figure 5 therefore shows that the critical properties of these quantities depend crucially on the type of splitting used.

5. Macrospin model

Finally, we analyze our framework from the stochastic perspective developed in section 3. To emphasize the generality of our results, we also focus on non-quench scenarios ($U \neq 1$). We consider a macrospin model, with $d = 2S + 1$ levels and spin operators S_x, S_y, S_z [89]. We consider a scenario similar to that of figure 2: the initial and final Hamiltonians are taken to coincide, being given by $H_0 = H_\tau = -h_z S_z$. And the unitary is driven by a magnetic pulse in the x direction, so $U = \exp\{-i\tau(H_0 - h_x S_x)\}$. Since the Hamiltonian is cyclic, the eigenbases $|i_0\rangle$ and $|j_\tau\rangle$ coincide. However, since the unitary is now non-trivial, the final state $\rho_\tau = U\rho_0^{th}U^\dagger$ will contain coherences in the S_z-basis.

A panel summarizing the results for the splitting (10) is shown in figure 6, where we plot the first four cumulants of λ_{cl} (images (a)–(d)) and those of λ_{qu} (images (f)–(i)), as a function of the Hilbert space dimension d and for different values of β. In figures 6(e) and (j), we also show exemplary plots of the full distributions $P(\lambda_{cl})$ and $P(\lambda_{qu})$, for fixed $d = 200$ and two values of β. For comparison, a similar panel, but for the quantities in (3), is shown in figure 7. Note also that some cumulants are scaled by either d or β, whenever a simple scaling rule could be found.

From these plots the following conclusions can be drawn. Concerning figure 6, all cumulants of λ_{cl} are found to be intensive, saturating at a finite value when $d \to \infty$. Conversely, all cumulants of λ_{qu} are extensive, scaling proportionally to d. The cumulants of λ_{qu} also scale with powers of β at low temperatures (figures 6(f)–(i)), but for higher order cumulants this scaling only becomes good at very low temperatures. For the splitting (3) the situation is reversed: now the cumulants of γ_{cl} become extensive (and quite similar to those of λ_{qu}), while those of γ_{qu} tend to saturate. The only exception is $\kappa_1(\gamma_{qu})$, which is found to grow

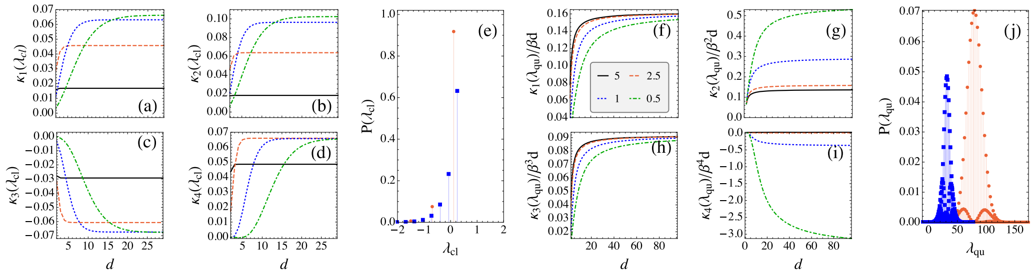

Figure 6. Statistics of the splitting (10) for the macrospin model, as a function of the Hilbert space dimension d and different values of β, as shown in image (f). (a)–(d) First four cumulants of λ_{cl}. (e) $P(\lambda_{cl})$ for $d = 200$, with $\beta = 1$ (blue) and $\beta = 2.5$ (red). (f)–(j) Same, but for λ_{qu}. Some of the cumulants are scaled by powers of β and d, whenever such a scaling law exists. Additional parameters: $h_z = 1$, $h_x = 0.5$ and .

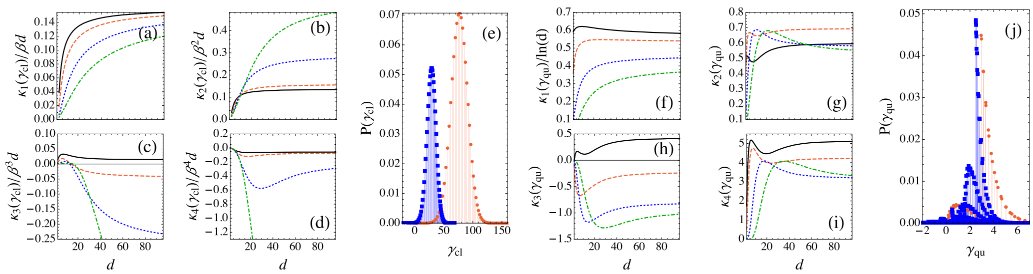

Figure 7. Same as figure 6, but for the splitting (3).

logarithmically with d. Notice that this dependence on the dimensionality is fundamentally different from what was found in the TFIM (section 4), where all quantities were extensive in the number of particles. We also note that the statistics of λ_{cl} (figure 6(e)) has significantly smaller support than that of λ_{qu}. This is a consequence of the fact that, from its definition in equation (40), λ_{cl} depends only on the initial points $|i_0\rangle$, while λ_{qu} depends on both $|i_0\rangle$ and $|j_\tau\rangle$.

The results in figure 6(e) indicate that even when $d \to \infty$, the distribution of λ_{cl} will never tend to a Gaussian. Conversely, for γ_{cl} in figure 7(e), this is clearly the case. This is supported by a comparison of the corresponding cumulants in images (a)–(d), which are intensive for λ_{cl} and extensive for γ_{cl}. As for λ_{qu} and γ_{qu}, even though the histograms in figures 6(j) and 7 (j) do not seem to indicate a Gaussian behavior, this is expected to eventually occur for sufficiently large d. For λ_{qu}, the scaling with d is similar to γ_{cl}, and hence the same argument as above applies. Conversely, the situation for γ_{qu} is more delicate, since the first cumulant scales only logarithmically (and hence very slowly) with d. Extremely large sizes may thus be necessary for a Gaussian behavior to be observed.

One might expect that in the limit $d \to \infty$ one should recover a classical spin model. This does not happen, however, as is evidenced by the fact that the coherent terms Γ_{qu} and Λ_{qu} do not vanish in this limit, but actually increase with d. The explanation for this rests essentially on a coarse-graining argument. Even though we take $d \to \infty$, we continue to assume we have full access to all eigenstates of the system, as appears, for instance, in the dephasing operations involved in constructing the intermediate states.

6. Conclusion

In this article, we studied how entropy production can be divided into a classical and quantum contribution, when a system is driven out of equilibrium. A popular choice in the literature is given in equation (3), see in particular [38]. This splitting has several interesting properties, including individual fluctuation theorems for each term [38] and a resource-theoretic interpretation [20, 21, 36]. However, we here noted it also has two major shortcomings. First, we showed that the classical contribution Γ_{cl} in equation (3) dominates for highly coherent processes and at low temperatures, in contrast with what might be expected. We observed this undesired behavior in all considered systems, from a simple driven qubit to a many-body Ising model at criticality, and identified the divergence of the relative entropy in (4), at low temperatures, as the underlying cause. Second, given a perturbation δg of the Hamiltonian, the radius of convergence of Γ_{qu} and Γ_{cl} tends to zero exponentially fast as $\beta \to \infty$, making this splitting impractical to characterise the entropy production of quenched systems at low temperatures.

In order to overcome these shortcomings, we suggested a new splitting for the entropy production given in equation (10), which was motivated by the developments of reference [30] for infinitesimal quenches. The definition is valid arbitrarily out-of-equilibrium. We also provided a formulation in terms of stochastic trajectories and a physical interpretation, highlighting how it can be obtained following a similar logic to the one behind (3). Indeed, both (3) and (10) can be understood by introducing intermediate states for comparison. The different choices, however, turn out to have crucial consequences, especially for highly coherent processes. Indeed, in the low-temperature regime the quantum term dominates in equation (10), but the classical one does in (3). For high temperatures and infinitesimal quenches, both splittings coincide. A comparison between the two approaches is summarized in table 1.

More generally, our considerations illustrate that it is non-trivial to identify the classical and quantum contributions in entropy production for an arbitrarily out-of-equilibrium process. In analogy with the definition of work for coherent processes [26, 90], the splitting of Σ in a classical and quantum term may not be unique, and will depend on the specific context into consideration. Nevertheless, there are some relevant scenarios where such a splitting is unambiguous. One is in a thermalization process described by either a Markovian master equation or as a resource-theoretic state transformation; in both cases, such a distinction seems to be very well captured by equation (3) [20, 21, 36]. On the other hand, when an equilibrium state is slightly moved out of equilibrium (e.g. by an infinitesimal quench), the splitting (10) provides a more accurate description of the quantum and classical contributions. In fact, in such a scenario, the entropy production can be decomposed into a classical and quantum contribution at all levels of the statistics, as shown in section 3.2 (see also [30, 31, 91]). For general out-of-equilibrium processes, however, classical and quantum contributions become inevitably mixed. Still, our results show that the splitting (10) has a more reasonable behavior (i.e., the quantum term dominates at low temperatures and for highly coherent processes).

In a second part of the article, we applied these ideas to a TFIM, and to a macrospin undergoing finite time dynamics. For the Ising model, we found that the behavior close to criticality is fundamentally different for both splittings, with the quantum component playing a predominant role for (10) and the classical component being dominant in (3). For the macrospin model, we focused not only on the average, but on the full statistics, including the first four cumulants and the corresponding probability distributions. We have found that different cumulants scale with the Hilbert space dimension d in non-trivial ways, some being extensive, others intensive or even logarithmic.

We hope that these results help to motivate further investigations on the non-trivial way in which populations and coherences intermix in quantum thermodynamic processes. We are particularly interested in further understanding how this unfolds for many-body systems in general. In particular, the analysis of higher order cumulants for these models has been seldom explored in the literature, even for Σ itself. It would also be interesting to generalize the present results for open systems, undergoing generic interactions with a heat bath. This can be done for quasi-static processes, following the approach in [30]. Or it can be constructed in a controllable way using collisional models [92]. Finally, these ideas could also be extended to describe quantum correlations in bipartite systems. For instance, instead of studying the entropy production in a work protocol, one may analyze it in the context of heat exchange between two quantum correlated systems, which are locally thermal, as studied in [35].

Acknowledgments

We thank M Scandi for insightful discussions. We acknowledge financial support from the Brazilian agencies Conselho Nacional de Desenvolvimento Científico e Tecnológico and Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) - Finance Code 001. GTL acknowledges the financial support of the São Paulo Funding Agency FAPESP (Grants Nos. 2017/50304-7, 2017/07973-5 and No. 2018/12813-0). MP-L acknowledges funding from Swiss National Science Foundation (Ambizione PZ00P2-186067).

Data availability statement

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

Appendix A. Nonnegativity of Λ_{qu}

In this appendix we show that Λ_{qu} defined in equation (19) is also non-negative, even though it cannot be written as a relative entropy. The proof is essentially based on the Bogoliubov variational theorem [93]. The first term in equation (19) reads explicitly $F(\tilde{\rho}_\tau^{\mathrm{th}}) = \mathrm{tr}\left\{\tilde{\rho}_\tau^{\mathrm{th}} H_\tau\right\} - TS(\tilde{\rho}_\tau^{\mathrm{th}})$. At first sight, this is not an equilibrium free energy, because the Hamiltonian H_τ is not the same as the one appearing in the exponent of $\tilde{\rho}_\tau^{\mathrm{th}}$ (equation (17)). However, due to the presence of the trace, we can equivalently write this as $F(\tilde{\rho}_\tau^{\mathrm{th}}) = \mathrm{tr}\left\{\tilde{\rho}_\tau^{\mathrm{th}} \mathbb{D}_{\rho_\tau}(H_\tau)\right\} - TS(\tilde{\rho}_\tau^{\mathrm{th}})$, which shows that it is actually an equilibrium free energy. Next, we note that the final Hamiltonian can be rewritten as $H_\tau = \mathbb{D}_{\rho_\tau}(H_\tau) + H_\tau^{\mathrm{c}}$, where $H_\tau^{\mathrm{c}} = H_\tau - \mathbb{D}_{\rho_\tau}(H_\tau)$. The Bogoliubov variational theorem [93] then yields

$$F(\rho_\tau^{\mathrm{th}}) \leqslant F(\tilde{\rho}_\tau^{\mathrm{th}}) + \mathrm{tr}\left\{\tilde{\rho}_\tau^{\mathrm{th}} H_\tau^{\mathrm{c}}\right\}. \tag{A1}$$

But, by construction, $H_\tau^{\mathrm{c}} = H_\tau - \mathbb{D}_{\rho_\tau}(H_\tau)$ has only off-diagonal elements in the common eigenbasis of $\tilde{\rho}_\tau^{\mathrm{th}}$ and ρ_τ. Thus, the second term in equation (A1) vanishes. Plugging the resulting inequality back into equation (19), we finally conclude that $\Lambda_{\mathrm{qu}} \geqslant 0$.

We can also show that for any finite temperature Λ_{qu} is zero if and only if ρ_τ is incoherent in the eigenbasis of H_τ. The *if* part of this statement is easy: when ρ_τ is incoherent in the final energy eigenbasis, $\mathbb{D}_{\rho_\tau}(H_\tau) = \mathbb{D}_{H_\tau}(H_\tau) = H_\tau$, which leads to $\Lambda_{\mathrm{qu}}(\rho_\tau) = 0$. Conversely, if we assume that $\Lambda_{\mathrm{qu}}(\rho_\tau) = 0$ and $\beta > 0$, we must have $F(\tilde{\rho}_\tau^{\mathrm{th}}) - F(\rho_\tau^{\mathrm{th}}) = 0$. This implies that

$$\sum_{k=0}^{+\infty} \frac{(-\beta)^k}{k!}\, \mathrm{tr}\left\{H_\tau^k - \mathbb{D}_{\rho_\tau}(H_\tau)^k\right\} = 0, \tag{A2}$$

which means $\mathrm{tr}\, H_\tau^k - \mathbb{D}_{\rho_\tau}(H_\tau)^k = 0,\ \forall\, k \in \mathbb{N}$. The case $k = 0$ is trivial and the case $k = 1$ follows directly from the definition of $\mathbb{D}_{\rho_\tau}(H_\tau)$. For the case $k = 2$, we use that

$$\mathrm{tr}\left\{H_\tau^2\right\} = \mathrm{tr}\left\{\left(\mathbb{D}_{\rho_\tau}(H_\tau) + H_\tau^{\mathrm{c}}\right)^2\right\} = \mathrm{tr}\left\{\mathbb{D}_{\rho_\tau}(H_\tau)^2 + 2\mathbb{D}_{\rho_\tau}(H_\tau)H_\tau^{\mathrm{c}} + (H_\tau^{\mathrm{c}})^2\right\}.$$

Again, using the definition of $\mathbb{D}_{\rho_\tau}(H_\tau)$ one may verify that $\mathrm{tr}\left\{\mathbb{D}_{\rho_\tau}(H_\tau)H_\tau^{\mathrm{c}}\right\} = 0$. Therefore we are left with

$$\mathrm{tr}\left\{H_\tau^2 - \mathbb{D}_{\rho_\tau}(H_\tau)^2\right\} = \mathrm{tr}\left\{\left(H_\tau - \mathbb{D}_{\rho_\tau}(H_\tau)\right)^2\right\} = 0. \tag{A3}$$

But since $H_\tau - \mathbb{D}_{\rho_\tau}(H_\tau)$ is also Hermitian, we must have $H_\tau - \mathbb{D}_{\rho_\tau}(H_\tau) = 0$. Then, since $\mathbb{D}_{\rho_\tau}(H_\tau) = H_\tau$, for $k \geqslant 3$, $\mathrm{tr}\left\{H_\tau^k - \mathbb{D}_{\rho_\tau}^k(H_\tau)\right\} = 0$ follows trivially, and ρ_τ must be incoherent in the eigenbasis of H_τ, i.e., we must have $[\rho_\tau, H_\tau] = 0$.

Appendix B. Diagonalization of the transverse field Ising model

The TFIM Hamiltonian in equation (75) can be diagonalized by a series of transformations, as shown in [94]. Our notation follows closely that of reference [39], which contains a self-contained derivation of these results. The first step is the introduction of a Jordan–Wigner transformation, that maps the spin chain onto an equivalent system of spinless fermions,

$$\sigma_j^x = (c_j^\dagger + c_j)\prod_{i<j}(1 - 2c_i^\dagger c_i), \qquad \sigma_j^z = 1 - 2c_j^\dagger c_j, \tag{B1}$$

where $c_j^\dagger$ and c_j are canonical creation and annihilation fermionic operators. We assume N is large and even. We may then ignore boundary terms [95], and introduce the Fourier transform

$$c_j = \frac{\mathrm{e}^{-\imath\pi/4}}{\sqrt{N}} \sum_k c_k\, \mathrm{e}^{\imath kj}, \tag{B2}$$

where $k = \pm(2n+1)\frac{\pi}{N}$ and $n = 0, 1, \dots, N/2 - 1$. Equation (75) is then transformed to

$$H(g) = \sum_{k>0}\left[(g - \cos k)(c_k^\dagger c_k - c_{-k} c_{-k}^\dagger) + \sin k(c_k^\dagger c_{-k}^\dagger + c_{-k} c_k)\right]. \tag{B3}$$

Next, we introduce a new set of Fermionic operators η_k through the Bogoliubov transformation

$$\eta_k = \cos(\theta_k/2)c_k + \sin(\theta_k/2)c^\dagger_{-k}. \tag{B4}$$

With the definitions

$$\begin{aligned} \epsilon_k(g) &= \sqrt{(g - \cos\, k)^2 + \sin^2 k}, \\ (\sin\, \theta_k, \cos\, \theta_k) &= \left(\frac{\sin(k)}{\epsilon_k}, \frac{g - \cos(k)}{\epsilon_k}\right), \end{aligned} \tag{B5}$$

we then finally obtain

$$H(g) = \sum_k \epsilon_k(g)\left(2\eta^\dagger_k \eta_k - 1\right). \tag{B6}$$

Exploring the fact that $\epsilon_{-k}(g) = \epsilon_k(g)$, we can rewrite $H(g)$ as a sum over only positive values of k,

$$H(g) = \sum_{k>0} 2\epsilon_k(g)(\eta^\dagger_k \eta_k + \eta^\dagger_{-k}\eta_{-k} - 1). \tag{B7}$$

This is useful because, as we will see, a perturbation δg couples pairs of modes $+k$ and $-k$. Finally, if we let $|n_{-k}n_k\rangle$ be the joint eigenstates of $\eta^\dagger_{-k}\eta_{-k}$ and $\eta^\dagger_k\eta_k$, where $n_{\pm k} = 0, 1$, we may also write

$$H(g) = \sum_{k>0} 2\epsilon_k(g)\left(-|0_{-k}0_k\rangle\langle 0_{-k}0_k| + |1_{-k}1_k\rangle\langle 1_{-k}1_k|\right). \tag{B8}$$

If we consider now a perturbation δg in the field, we have

$$\Delta H = -\delta g \sum_{j=1}^N \sigma^z_j = \delta g \sum_{j=1}^N (2c^\dagger_j c_j - 1) = \delta g \sum_k (2c^\dagger_k c_k - 1), \tag{B9}$$

where we used equations (B1) and (B2). Finally, using equations (B4) and (B5) we obtain

$$\Delta H = 2\delta g \sum_{k>0}\left[\cos\, \theta_k(\eta^\dagger_k\eta_k + \eta^\dagger_{-k}\eta_{-k} - 1) + \sin\, \theta_k(\eta^\dagger_{-k}\eta^\dagger_k - \eta_{-k}\eta_k)\right], \tag{B10}$$

where the coupling between $+k$ and $-k$ modes is clear from the second term. Alternatively, this can be written as $\Delta H = \Delta H^\mathrm{d} + \Delta H^\mathrm{c}$, with

$$\Delta H^\mathrm{d} = 2\delta g \sum_{k>0} \cos\, \theta_k\left(-|0_{-k}0_k\rangle\langle 0_{-k}0_k| + |1_{-k}1_k\rangle\langle 1_{-k}1_k|\right), \tag{B11}$$

$$\Delta H^\mathrm{c} = 2\delta g \sum_{k>0} \sin\, \theta_k\left(|0_{-k}0_k\rangle\langle 1_{-k}1_k| + |1_{-k}1_k\rangle\langle 0_{-k}0_k|\right), \tag{B12}$$

where ΔH^d and ΔH^c are the dephased and coherent parts of the perturbation, respectively.

Appendix C. Λ_cl and Λ_qu for the TFIM

Using the results from (B), we now show how to compute Λ_cl and Λ_qu using equations (18) and (19). Since we consider the initial field to be g_0, the initial Hamiltonian is given by

$$\begin{aligned} H_0 &= \sum_k \epsilon^0_k(2\eta^\dagger_k\eta_k - 1), \\ &= \sum_{k>0} 2\epsilon^0_k\left(-|0_{-k}0_k\rangle\langle 0_{-k}0_k| + |1_{-k}1_k\rangle\langle 1_{-k}1_k|\right) \end{aligned} \tag{C1}$$

where $\epsilon^0_k = \epsilon_k(g_0)$ and $|n_{-k}n_k\rangle$ are the joint eigenstates of $\eta^\dagger_k\eta_k$ and $\eta^\dagger_{-k}\eta_{-k}$. Thus, the initial state $\rho^\mathrm{th}_0 = \mathrm{e}^{-\beta H_0}/Z_0$ can be written as

$$\rho^\mathrm{th}_0 = \prod_{k>0} \rho^\mathrm{th}_{0\,|\pm k}, \tag{C2a}$$

$$\rho^\mathrm{th}_{0\,|\pm k} = \sum_{n_k=0,1,\ n_{-k}=0,1} \frac{\mathrm{e}^{2\beta\epsilon^0_k(1-n_k-n_{-k})}}{4\cosh^2(\beta\epsilon^0_k)}|n_{-k}n_k\rangle\langle n_{-k}n_k|. \tag{C2b}$$

After the instantaneous quench, which changes the field to its final value $g_\tau = g_0 + \delta g$, we have the final Hamiltonian,

$$\begin{aligned} H_\tau &= \sum_k \epsilon_k^\tau (2\xi_k^\dagger \xi_k - 1), \\ &= \sum_{k>0} 2\epsilon_k^\tau \left(-|0_{-k}^\tau 0_k^\tau\rangle\langle 0_{-k}^\tau 0_k^\tau| + |1_{-k}^\tau 1_k^\tau\rangle\langle 1_{-k}^\tau 1_k^\tau|\right) \end{aligned} \tag{C4}$$

where $\epsilon_k^\tau = \epsilon_k(g_\tau)$ and $|n_{-k}^\tau n_k^\tau\rangle$ are the joint eigenstates of the post-quench fermionic operators $\xi_k^\dagger \xi_k$ and $\xi_{-k}^\dagger \xi_{-k}$, which are related to the pre-quench operators $\{\eta_k\}$ according to [73]

$$\xi_k = \cos(\Delta_k/2)\eta_k + \sin(\Delta_k/2)\eta_{-k}^\dagger, \tag{C5}$$

where $\sin\,\Delta_k = -\delta g\,\sin(k)/\epsilon_k^\tau \epsilon_k^0$. As discussed in appendix B, equation (C5) shows us that the perturbation couples pairs of modes $+k$ and $-k$. This is why it is more convenient to write all quantities as $\rho_0^{\text{th}} = \prod_{k>0} \rho_{0|\pm k}^{\text{th}}$ and $H_0 = \sum_{k>0} H_{0|\pm k}$, instead of a product/sum over the negative values of k.

The corresponding final equilibrium state $\rho_\tau^{\text{th}} = \mathrm{e}^{-\beta H_\tau}/Z_\tau$ is given by

$$\rho_\tau^{\text{th}} = \prod_{k>0} \rho_{\tau|\pm k}^{\text{th}}, \tag{C5a}$$

$$\rho_{\tau|\pm k}^{\text{th}} = \sum_{n_k^\tau=0,1,\ n_{-k}^\tau=0,1} \frac{\mathrm{e}^{2\beta\epsilon_k^\tau(1-n_k^\tau-n_{-k}^\tau)}}{4\cosh^2(\beta\epsilon_k^\tau)} |n_{-k}^\tau n_k^\tau\rangle\langle n_{-k}^\tau n_k^\tau|. \tag{C5b}$$

We can proceed now to calculate $\tilde{\rho}_\tau^{\text{th}}$ in equation (17). We first compute the dephased Hamiltonian $H_0 + \varDelta H^{\text{d}}$, where $\varDelta H^{\text{d}}$ is given in equation (B11). That is,

$$\mathbb{D}_{\rho_0^{\text{th}}}(H_\tau) = \sum_{k>0} 2\tilde{\epsilon}_k^\tau \left(-|0_{-k}0_k\rangle\langle 0_{-k}0_k| + |1_{-k}1_k\rangle\langle 1_{-k}1_k|\right), \tag{C8}$$

where $\tilde{\epsilon}_k^\tau = \epsilon_k^\tau\,\cos\,\Delta_k = \epsilon_k^0 + \delta g\,\cos\,\theta_k$. From this, one then finds the associated thermal state (equation (17))

$$\tilde{\rho}_\tau^{\text{th}} = \prod_{k>0} \tilde{\rho}_{\tau|\pm k}^{\text{th}}, \tag{C7a}$$

$$\tilde{\rho}_{\tau|\pm k}^{\text{th}} = \sum_{n_k=0,1,\ n_{-k}=0,1} \frac{\mathrm{e}^{2\beta\tilde{\epsilon}_k^\tau(1-n_k-n_{-k})}}{4\cosh^2(\beta\tilde{\epsilon}_k^\tau)} |n_{-k}n_k\rangle\langle n_{-k}n_k|. \tag{C7b}$$

We have all we need to compute $\varLambda_{\text{cl}}$ and $\varLambda_{\text{qu}}$ now. We just have to plug equations (C2), (C6) and (C9) into (18) and (19). Because all states ρ_0^{th}, $\tilde{\rho}_\tau^{\text{th}}$ and ρ_τ^{th} are separable in terms of $\pm k$ modes, $\varLambda_{\text{cl}}$ and $\varLambda_{\text{qu}}$ will be given as sums over k. Hence, we find

$$\Lambda_{\text{cl}} = \sum_{k>0} 2\left\{\ln\left[\frac{\cosh\left(\beta\tilde{\epsilon}_k^\tau\right)}{\cosh\left(\beta\epsilon_k^0\right)}\right] + \beta\left(\epsilon_k^0 - \tilde{\epsilon}_k^\tau\right)\tanh\left(\beta\epsilon_k^0\right)\right\}, \tag{C11}$$

and

$$\Lambda_{\text{qu}} = \sum_{k>0} 2\,\ln\left[\frac{\cosh\left(\beta\epsilon_k^\tau\right)}{\cosh\left(\beta\tilde{\epsilon}_k^\tau\right)}\right]. \tag{C12}$$

Finally, in the limit of very large N, all k-sums can be converted to integrals and all quantities become extensive in N. In particular, we can substitute $\sum_{k>0} \to N\int_0^\pi \frac{\mathrm{d}k}{2\pi}$ in equations (C11) and (C12) to obtain equations (78) and (79).

We note that equations (78) and (79) do not assume that the quench is infinitesimal. All they assume is that $U = 1$. If, in particular, we are interested in infinitesimal quenches, then we may series expand these expressions in powers of δg, leading to

$$\Lambda_{\text{cl}} = N\beta^2\,\delta g^2 \int_0^\pi \frac{\mathrm{d}k}{2\pi}\,\text{sech}^2(\beta\epsilon_k^0)\cos^2\theta_k, \tag{C10}$$

$$\Lambda_{\text{qu}} = N\beta^2\,\delta g^2 \int_0^\pi \frac{\mathrm{d}k}{2\pi}\,\frac{\tanh(\beta\epsilon_k^0)}{\beta\epsilon_k^0}\,\sin^2\theta_k, \tag{C11}$$

where it is clear the relation of $\varLambda_{\text{cl}}$ and $\varLambda_{\text{qu}}$ with the dephased and coherent parts of the perturbation in equations (B11) and (B12). Furthermore, it is easy to check that they satisfy equation (33).

Appendix D. Γ_{cl} and Γ_{qu} for the TFIM

For completeness, in this appendix we write down the expressions for Γ_{cl} and Γ_{qu} for the TFIM, computed in [39]:

$$\Gamma_{\mathrm{qu}} = N\int_0^{\pi} \frac{\mathrm{d}k}{2\pi}\left\{\frac{1}{2}\tanh\left(\beta\epsilon_k^0\right)\left[\ln\left[\frac{1+\tanh\left(2\beta\epsilon_k^0\right)}{1-\tanh\left(2\beta\epsilon_k^0\right)}\right] - \cos(\Delta_k)\ln\left[\frac{1+\tanh\left(2\beta\epsilon_k^0\right)\cos(\Delta_k)}{1-\tanh\left(2\beta\epsilon_k^0\right)\cos(\Delta_k)}\right]\right]\right.$$
$$\left. - \frac{\cosh\left(2\beta\epsilon_k^0\right)}{4\cosh^2\left(\beta\epsilon_k^0\right)}\ln\left[1+\sinh^2\left(2\beta\epsilon_k^0\right)\sin^2(\Delta_k)\right]\right\}, \tag{D1}$$

$$\Gamma_{\mathrm{cl}} = N\int_0^{\pi} \frac{\mathrm{d}k}{2\pi}\left\{2\ln\left[\frac{\cosh\left(\beta\epsilon_k^{\tau}\right)}{\cosh\left(\beta\epsilon_k^0\right)}\right] - \frac{1}{2}\tanh\left(\beta\epsilon_k^0\right)\cos(\Delta_k)\right.$$
$$\times\left[\ln\left[\frac{1+\tanh\left(2\beta\epsilon_k^{\tau}\right)}{1-\tanh\left(2\beta\epsilon_k^{\tau}\right)}\right] - \ln\left[\frac{1+\tanh\left(2\beta\epsilon_k^0\right)\cos(\Delta_k)}{1-\tanh\left(2\beta\epsilon_k^0\right)\cos(\Delta_k)}\right]\right]$$
$$\left. + \frac{\cosh\left(2\beta\epsilon_k^0\right)}{4\cosh^2\left(\beta\epsilon_k^0\right)}\ln\left[1+\sinh^2\left(2\beta\epsilon_k^0\right)\sin^2(\Delta_k)\right]\right\}. \tag{D2}$$

These expressions were used in plotting figures 5(c) and (d). The problem in the analyticities of these quantities stem from the last term in both integrals: in order to series expand them we need to satisfy the condition $\sinh^2(2\beta\epsilon_k^0)\sin^2(\Delta_k) < 1$. This is prohibitive at low temperatures, since this function scales exponentially with β, through $\sinh^2(2\beta\epsilon_k^0)$, but only polynomially with the perturbation, through $\sin^2(\Delta_k)$.

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$$\mathbb{D}_H(\rho) = \lim_{s\to\infty} \frac{1}{s}\int_0^s \mathrm{d}t e^{-iHt}\rho e^{iHt}.$$

We then use that $e^{t(H_0+\Delta H)} = e^{tH_0} + t\mathbb{J}_{e^{tH_0}}[\Delta H] + \mathcal{O}(\Delta H^2)$ and $[\rho_0^{\mathrm{th}}, H_0] = 0$. To order ΔH this gives equation (26).
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