Supplementary Information

In what follows we present the full technical details of all our findings. Section A provides the definition of the basic quantities used in our work, namely the Rényi entropies and divergences and useful relations between them. In Section B, we briefly summarize the theory of majorization and trumping, which we will later generalise and use to derive our set of exact second laws. We then introduce the connection between Rényi divergences (which our generalised free energies are based on), and trumping, and show how to reduce the number of trumping relations required. We then show that this provides the basis of catalytic transformations in the simple setting where the Hamiltonian is trivial.

In Section C, we provide the overview description of catalytic operations in the context of full thermodynamics with general Hamiltonians. We then proceed in Section D to show that only a unique set of states (namely Gibbs states) can be used as free resources, to obtain non-trivial state transition conditions. This allows us to define equivalence classes between state-Hamiltonian pairs, giving rise to the zeroeth law of thermodynamics. Our main technical result is Theorem 17 of Section E, which generalises trumping relations from the bistocastic case, to the case of arbitrary channels which take a subset of states to some other subset of states. We expect this to be of independent interest with applicability in a broad range of contexts. This provides the mathematical tools to formulate necessary and sufficient conditions for thermodynamic state transformations for states block diagonal in the energy eigenbasis. These are detailed in Section F. Here, we also observe the elimination of conditions for ranges of α, when certain states are allowed to be used and returned with close fidelity. In particular, borrowing a pure state allows us to eliminate conditions where $\alpha < 0$, which significantly simplifies the conditions on state transformations. We also apply our second laws to the case of Landauer erasure of states while preserving a memory in Subsection F 5. In Subsection F 4 we go beyond classical states, and present a family of fully quantum second laws.

Section G makes an extensive presentation on the case where inexact catalysis is allowed, and we show that for different regimes of closeness between the catalyst's initial and final state, we obtain different second laws. The different regimes include exact catalysis, returning the catalyst such that a small amount of work is required to bring it to it's original state, returning the catalyst with small error per particle, returning it such that only a small amount of work on average is required to bring it back to its original state, and returning it with arbitrary good fidelity, where we find that all state transformations are possible. In Supplementary Information H we show how for larger systems, one can often check just two quantities to determine if a state transformation is possible. In I, we discuss in more detail how our paradigm incorporates changing Hamiltonians. Since we have conditions for state transformations using ancillary systems, we are in a position to prove the optimality of the procedure for changing a Hamiltonian used in [1]. The work storage system is also an ancillary system, and we further discuss the equivalence of the work bit, to other batteries.

Appendix A: Preliminaria: Rényi relative entropies and their properties

1. Rényi divergence

Consider probability distributions $p = \{p_1, p_2, ..., p_n\}$ and $q = \{q_1, q_2, ..., q_n\}$. The Rényi divergences are defined for $\alpha \in [-\infty, \infty]$ as follows

$$D_\alpha(p\|q) = \frac{\mathrm{sgn}(\alpha)}{\alpha - 1} \log \sum_i^n p_i^\alpha q_i^{1-\alpha}, \tag{A1}$$

where

$$\mathrm{sgn}(\alpha) = \begin{cases} 1 & (\alpha \geq 0); \\ -1 & (\alpha < 0). \end{cases} \tag{A2}$$

We use the conventions $\frac{0}{0} = 0$ and $\frac{a}{0} = \infty$ for $a > 0$. The cases $\alpha = 0, 1, \infty, -\infty$ are defined via the suitable limit, namely

$$D_0(p\|q) = \lim_{\alpha \to 0^+} D_\alpha(p\|q) = -\log \sum_{i:p_i \neq 0}^n q_i, \qquad D_1(p\|q) = \lim_{\alpha \to 1} D_\alpha(p\|q) = \sum_i^n p_i(\log p_i - \log q_i),$$

$$D_\infty(p\|q) = \lim_{\alpha \to \infty} D_\alpha(p\|q) = \log \max_i \frac{p_i}{q_i}, \qquad D_{-\infty}(p\|q) = -\lim_{\alpha \to -\infty} D_\alpha(p\|q) = D_\infty(q\|p).$$

Note that for $\alpha \to \infty$, $D_\alpha(p\|q)$ can also be expressed as:

$$D_\infty(p\|q) = \log \min \left\{ \lambda | \forall i, \lambda \geq \frac{p_i}{q_i} \right\} \tag{A3}$$

Also, there exists a useful relation between two divergences for $\alpha \notin \{0,1\}$:

$$\alpha \operatorname{sgn}(1-\alpha) D_{1-\alpha}(p\|q) = (1-\alpha)\operatorname{sgn}(\alpha) D_\alpha(q\|p). \tag{A4}$$

For some properties of Rényi divergence, the reader can refer to [2, 3]. Note that discussions in other literatures define Rényi divergence for only non-negative alphas. However, the relative entropy D_α we define satisfies the data processing inequality for all $\alpha \in [-\infty, \infty]$:

$$D_\alpha(\Lambda(p)\|\Lambda(q)) \leq D_\alpha(p\|q), \tag{A5}$$

where Λ is a stochastic map. The range $\alpha \in [0, \infty]$ is proven in [3], while the negative regime is an immediate consequence of Eq. (A4). For $\alpha \in [0, \infty]$ the Rényi divergence is nondecreasing in α:

$$D_\alpha(p\|q) \leq D_\gamma(p\|q) \tag{A6}$$

for $\alpha \leq \gamma$ (see Theorem 3 of Ref. [2]).

2. Rényi entropy

The Rényi entropies are defined for $\alpha \in R \setminus \{0,1\}$ as

$$H_\alpha(p) = \frac{\operatorname{sgn}(\alpha)}{1-\alpha} \log \sum_{i=1}^{n} p_i^\alpha, \tag{A7}$$

where $\operatorname{sgn}(\alpha)$ has been defined in (A2). Again, for $\alpha \in \{-\infty, 0, 1, \infty\}$ we define $H_\alpha(p)$ by taking limits. Explicitly we have

$$H_0(p) = \log \operatorname{rank}(p), \quad H_1(p) = -\sum_{i=1}^{n} p_i \log p_i, \quad H_\infty(p) = -\log p_{\max} \quad H_{-\infty}(p) = \log p_{\min} \tag{A8}$$

where $\operatorname{rank}(p)$ is the number of nonzero elements of p, and $p_{\max}$, $p_{\min}$ are the maximal and minimal element of p, respectively. The Rényi entropies can be recovered from the relative Rényi entropies as follows

$$H_\alpha(p) = \operatorname{sgn}(\alpha) \log n - D_\alpha(p\|\eta) , \tag{A9}$$

with $\eta = \{\frac{1}{n}, \frac{1}{n}, ..., \frac{1}{n}\}$ is the uniform probability distribution.

It is worth noting that the Rényi divergences and entropies have generally been defined only for positive alphas. However, for completeness, we have generalized the definitions to negative alphas so that conditions for state transformations can be described fully by these quantities.

3. Quantum Rényi divergence

The Rényi divergence can be generalized to the quantum case in many different ways (see e.g. [4, 5]) due to noncommutativity of quantum states. The most straightforward generalization is the following candidate:

$$\tilde{S}_\alpha(\rho\|\sigma) = \frac{\operatorname{sgn}(\alpha)}{\alpha - 1} \log \operatorname{tr} \rho^\alpha \sigma^{1-\alpha}. \tag{A10}$$

For $\alpha = 0$ it reduces to the min-relative entropy given by $\tilde{S}_{\min}(\rho\|\sigma) = -\log \operatorname{tr}(\Pi_\rho \sigma)$, where π_ρ is the projector onto the support of ρ, and for $\alpha \to 1$ it reduces to the standard quantum relative entropy:

$$\lim_{\alpha \to 1^+} \tilde{S}_\alpha(\rho\|\sigma) = S(\rho\|\sigma) = \operatorname{tr}(\rho \log \rho - \rho \log \sigma). \tag{A11}$$

As proved in Lemma B.4 of [6], this version of Rényi divergence is known to be monotonic under quantum operations for $\alpha \in [0,2]$, i.e. for any completely positive trace-preserving (CPTP) map Λ and states ρ, σ we have

$$\tilde{S}_\alpha(\Lambda(\rho)||\Lambda(\sigma)) \leq \tilde{S}(\rho||\sigma), \tag{A12}$$

for $0 \leq \alpha \leq 2$.

In [7, 8] (see also [9]) another version of the quantum divergence was introduced for $\alpha \in (0,\infty]$:

$$S_\alpha(\rho||\sigma) = \frac{1}{\alpha - 1} \log \left(\mathsf{tr}(\sigma^{\frac{1-\alpha}{2\alpha}} \rho \sigma^{\frac{1-\alpha}{2\alpha}})^\alpha \right) \tag{A13}$$

The cases $\alpha = 1$ and $\alpha = \infty$ are obtained by limits so that $S_1(\rho||\sigma)$ is the standard relative entropy (A11) and $S_\infty(\rho||\sigma) = \log ||\sigma^{-\frac{1}{2}} \rho \sigma^{-\frac{1}{2}}||_\infty$ where $||\cdot||_\infty$ is the operator norm. Recently it was proven in [5] (see also [10]) that this entropy is monotonic under quantum completely positive trace preserving maps for $\alpha \geq 1/2$, namely for any CPTP map Λ and states ρ, σ we have

$$S_\alpha(\Lambda(\rho)||\Lambda(\sigma)) \leq S(\rho||\sigma), \tag{A14}$$

for $\alpha \geq 1/2$.

Note, that if ρ and σ commute, then both types of quantum Rényi divergences reduce to the classical version.

For both of these quantities, we can further define a Rényi-skew-divergence generalising the skew divergence of [11]. I.e.

$$S_\alpha^s(\rho||\sigma) := S_\alpha^s(\rho||\gamma) \tag{A15}$$

with $\gamma = s\rho + (1-s)\sigma$ and $s \in [0,1]$. The definition is similar for $\tilde{S}$, and they will both turn out to be a monotone under thermal operations when σ is the thermal state.

4. Majorization and Schur convexity

There is a partial order between probability distributions called majorization, which is defined for arbitrary vectors $x, y \in R_k^+$. We say that $x = (x_1, \ldots x_k)$ majorizes $y = (y_1, \ldots y_k)$ if for all $l = 1, \ldots k$

$$\sum_{i=1}^l x_i^\downarrow \geq \sum_{i=1}^l y_i^\downarrow, \quad \text{and} \quad \sum_{i=1}^k x_i = \sum_{i=1}^k y_i, \tag{A16}$$

where $x^\downarrow$ is a vector obtained by arranging the components of x in decreasing order: $x^\downarrow = (x_1^\downarrow, \ldots, x_k^\downarrow)$ where $x_1^\downarrow \geq \ldots \geq x_k^\downarrow$. We write

$$x \succ y \tag{A17}$$

to indicate that x majorizes y.

A function f is called Schur convex if it always preserves the majorization order, i.e. if $x \succ y$ implies $f(x) \geq f(y)$. If the majorization order is always reversed, the function is called Schur concave. A function is called *strictly* Schur convex if $x \succ y$ implies $f(x) > f(y)$ except when $x^\downarrow = y^\downarrow$, then $f(x) = f(y)$. A useful criterion for strict Schur convexity is stated in the following lemma:

Lemma 1. *A function $f : R_+^k \to R$ of the form $f(x) = \sum_i g(x_i)$ is (strictly) Schur convex/concave, iff $g(x)$ is (strictly) convex/concave.*

This lemma follows from the general criterion of Schur convexity [12]. However it is easy to prove directly, using the Birkhoff-von Neumann theorem, which states that if $p \succ q$, then q is a convex combination of permutations on p. (cf. Theorem 11). Using this, and the strict monotonicity of the logarithm, we see that the Rényi entropies reverse the majorization order:

Lemma 2. *For $\alpha \in (-\infty, 0) \cup (0, \infty)$, the Rényi entropies H_α are strictly Schur concave. For $\alpha = 0, \pm\infty$, the Rényi entropies are Schur concave. The function $\sum_i \log p_i$ is also strictly Schur concave.*

Appendix B: Exact catalysis with trivial Hamiltonian

We are interested in the interplay of energy and entropy, which is the essence of thermodynamics. However, before we approach this problem, it is instructive to first consider the case where the Hamiltonian is trivial in the sense that $H = 0$, and thermodynamics is reduced to bare information theory.

This toy model for thermodynamics was described in [13] and it has its roots in the problem of exorcising Maxwell demon [14, 15]. It is constructed within a framework of so called resource theories fo thermodynamics [1, 13, 16, 17] having its roots in research on entanglement manipulations [18–21], where one is interested in transformations between states by means of an allowed class of operations. Some states can be brought in for free, and they constitute a free resource. The others cannot be created, but only manipulated, i.e. we may transform one resource state into some other one. In the mentioned toy thermodynamics, which is perhaps the simplest known resource theory, the free resource are just maximally mixed states, and all unitary transformations as well as partial trace are allowed operations. The emerging class of operations was called *noisy operations*. It was shown that in the case of systems of the same size, the class of noisy operations is equivalent to mixtures of unitaries. Therefore the condition that ρ can be transformed into ρ' is equivalent to majorization: ρ can be transformed into ρ' iff the spectrum of ρ majorizes the spectrum of ρ' [22]. The noisy operations are equivalent to thermal operations applied to a system with a trivial Hamiltonian.

As we mentioned, in this toy thermodynamics, the law that governs state-to-state transitions is majorization. However, this is only so when we do not allow ancillary systems that are then returned in the same state. It is interesting to analyse how the laws change if we allow catalytic transitions between states. Since the transitions without catalysis are governed by majorization, the catalytic transitions will be governed by *trumping*. We say that x can be trumped into y if there exists some finite-dimensional z such that

$$x \otimes z \succ y \otimes z. \tag{B1}$$

The condition that z be finite dimensional is physically reasonable, since the experimenter is working within a finite volume and below some maximum energy. In [23] it was for the first time shown that using catalysis, one can perform transitions otherwise impossible. In particular the following explicit example of states that do not majorize one another, but allow catalytic transition was given

$$p = \left(\frac{4}{10}, \frac{4}{10}, \frac{1}{10}, \frac{1}{10}\right), \quad q = \left(\frac{1}{2}, \frac{1}{4}, \frac{1}{4}, 0\right) \tag{B2}$$

One checks that $p_1 < q_1$ but $p_1 + p_2 > q_1 + q_2$. Now, if one takes the catalyst $r = \left(\frac{6}{10}, \frac{4}{10}\right)$, then $q \otimes r \succ p \otimes r$, so that q can be trumped into p.

Recently Klimesh and Turgut [24, 25] independently provided necessary and sufficient conditions for x to be trumped into y. These were in terms of functions closely related to the Rényi entropies. We present here a set of conditions, equivalent to the Klimesh-Turgut ones, written in terms of Rényi divergences w.r.t. the uniform distribution η. Let us first state the result of Klimesh's work in the following lemma.

Lemma 3 (Theorem 2, [24]). *Let $x \in R_k^+$ and $y \in R_k^+$ be probability vectors which do not both contain components equal to zero, where $x \neq y$. Then x can be trumped into y if, and only if, for all $\alpha \in (-\infty, \infty)$:*

$$f_\alpha(x) > f_\alpha(y), \tag{B3}$$

where

$$f_\alpha(x) = \begin{cases} \log \sum_{i=1}^k x_i^\alpha & (\alpha > 1); \\ \sum_{i=1}^k x_i \log x_i & (\alpha = 1); \\ -\log \sum_{i=1}^k x_i^\alpha & (0 < \alpha < 1); \\ -\sum_{i=1}^k \log x_i & (\alpha = 0); \\ \log \sum_{i=1}^k x_i^\alpha & (\alpha < 0), \end{cases} \tag{B4}$$

Subsequently, we prove that when x is only required to trump y up to some arbitrarily high precision (meaning that x trumps another state y_ε arbitrarily close to y), then the conditions originally stated in Lemma 3 can be equivalently written in terms of non-strict inequalities.

Proposition 4. *Let $x \in R_k^+$ and $y \in R_k^+$ be probability vectors. Then the following conditions are equivalent:*

1. For arbitrary $\varepsilon > 0$, there exists y_ε such that $||y - y_\varepsilon|| \leq \varepsilon$ and x can be trumped into y_ε.

2. *The following inequality:*

$$D_\alpha(x||\eta) \geq D_\alpha(y||\eta) \tag{B5}$$

holds for all $\alpha \in (-\infty, \infty)$, with $\eta = (1/k, \ldots, 1/k)$ being the uniform distribution.

Proof. Let us first consider the trivial case where $x = y$. Then condition 1 holds, since for any $\varepsilon > 0$, we can define $y_\varepsilon = y = x$ itself, and we know that $x \succ x$ is true. Condition 2 also holds by definition.

Next, we divide the proof into two parts:

"Condition 1 implies 2": Let us first note that without loss of generality we can assume that y_ε is of full rank. Indeed, if it is not, we can perturb the state to add a small amount of noise (which is a valid noisy operation), obtaining $\tilde{y}_{\varepsilon'}$ which has full rank, i.e. no zeros, and satisfies $||\tilde{y}_{\varepsilon'} - y|| \leq \varepsilon'$, with ε' arbitrarily small. With this assumption, the condition of numbers of zeroes in x and y need not be stated.

Let us then look at the case where if $x \neq y$. Assume that condition 1 holds. Then by Lemma 3 we know that for any $\varepsilon > 0$, there exists y_ε such that for all $\alpha \in (-\infty, \infty)$,

$$f_\alpha(x) > f_\alpha(y_\varepsilon). \tag{B6}$$

We also know that the functions f_α are continuous, and this is true even for the case when the functions diverge to infinity. For example, consider y which does not have full rank, $\lim_{\varepsilon \to 0+} f_0(y_\varepsilon) = \infty$ even if y_ε is assumed to be of full rank. Therefore, we conclude that Eq. (B6) implies that $f_\alpha(y) = \lim_{\varepsilon \to 0} f_\alpha(y_\varepsilon) \leq f_\alpha(x)$.

Now, all the functions f_α, excluding $\alpha = 0$ are proportional to $-H_\alpha(x)$. In turn, the Rényi entropy is related to $D_\alpha(x||\eta)$ by Equation (A9). Since D_0 is obtained by taking D_α in the limit of $\alpha \to 0+$, according to Remark 5 we have that $D_0(x||\eta) \geq D_0(y||\eta)$.

"Condition 2 implies 1": Now, suppose that condition 2 holds instead. From the argument above we know that this translates to $f_\alpha(x) \geq f_\alpha(y)$ for all $\alpha \neq 0$. For any $\varepsilon > 0$, define $y_\varepsilon = (1 - \varepsilon)y + \varepsilon\eta$, and we know that $y \succ y_\varepsilon$, which also means that y trumps y_ε. Hence by Lemma 2, $\forall \alpha \neq 0$, $f_\alpha(x) > f_\alpha(y_\varepsilon)$.

Before we proceed, note that for any $\varepsilon > 0$, $f_0(y_\varepsilon) < \infty$. If x has full rank, then it can be verified that

$$-\frac{1}{k}\sum_{i=1}^{k}\log x_i - \log k = \lim_{\alpha \to 0^+}\frac{1-\alpha}{\alpha}D_\alpha(x||\eta). \tag{B7}$$

We can now analyse the following cases.

A) x is not of full rank : In this case, $f_0(x) = \infty > f_0(y_\varepsilon)$, so the condition on f_0 holds trivially.

B) x is of full rank : In this case, Eq. (B7) holds. Therefore we know that if condition 2 implies $D_0(x||\eta) \geq D_0(y||\eta)$, then this also implies that $f_0(x) \geq f_0(y) > f_0(y_\varepsilon)$. This independently verifies that condition 2 implies that the Klimesh conditions for trumping corresponding to $\alpha = 0$ is satisfied.

We have seen that assuming condition 2, we can construct y_ε such that the Klimesh trumping conditions in Lemma 3 holds for x and y_ε. Therefore by Lemma 3 implies that for any $\varepsilon > 0$, there exists y_ε such that x trumps y_ε (condition 1).

□

Remark 5. *Knowing that non-strict inequalities are enough, one can remove any discrete number of trumping conditions, as they can be obtained in the limit from the other conditions.*

The Klimesh-Turgut conditions have a peculiar feature. Namely, if there are zeros in both x and y we have to truncate the zeros and compute the conditions on smaller vectors. This means that we do not simply compare two functions, but which function we will choose depends on some relation between the vectors. Second, the condition of having zeros in both vectors is very unstable. If we slightly perturb y so that we remove zeros, then we do not need to truncate anymore. Note that the problem here is only with negative α, as for positive α the functions $D_\alpha(p||\eta)$ do not depend on additional zeros, while the functions with negative α are infinite, when at least one component vanishes.

We note that earlier, Aubrun and Nechita [26] gave conditions for trumping, in which only H_α with $\alpha > 1$ were needed. This is because they considered a special kind of closure, where one is allowed to add an arbitrary number of zeros to the initial vector x while returning an (arbitrarily good) approximation of the needed output y. This is a kind of embezzling (see section G 2 for definition of embezzling): One adds an ancilla and returns it with arbitrary small error, but the size of ancilla needs to grow in order to make the error smaller. In thermodynamics this is not allowed, as according to the second law, we should consider processes which do not change the environment. Proposition 4 already suggests how the conditions for thermodynamics should look like: the maximally mixed state should get replaced by the Gibbs state. In section E we will prove that this is indeed the case.

1. Restricting to $\alpha \geq 0$ by investing a small amount of extra work

In this section, we will argue that if we are allowed to invest an arbitrary small amount of work, only the conditions with positive α are relevant. This was considered by Aubrun and Nechita in [26], and the way we do that below is very similar.

We have three cases

(i) p has more zeros than q.

(ii) p has fewer zeros than q.

(iii) p and q have the same number of zeros (in particular, can both have no zeros).

In case (i), after truncation p will still have zeros, and so relative entropies for p with negative α will be infinite. Therefore the conditions with negative α are always satisfied. In case (ii) the transition cannot be realised, but this is reported by comparing ranks, which can be obtained using $H_0 = \lim_{\alpha \to 0^+} H_\alpha$. So again already the conditions with $\alpha > 0$ report the impossibility of transition, and negative α's are not needed. Finally, in case (iii), if we consider the transition $p \to q$, with a small amount of work invested in addition:

$$p \otimes |0\rangle_{d+1}\langle 0| \otimes \eta_d \to q \otimes |0\rangle_d\langle 0| \otimes \eta_{d+1}, \tag{B8}$$

where $|0\rangle_k\langle 0|$ stands for the distribution $\underbrace{(1, 0, \ldots, 0)}_{k}$ (equivalently, a pure state on C^k) and η_k stands for the uniform distribution with k elements. The invested amount of work is $\log \frac{d+1}{d}$, hence it is arbitrary small, when $d \to \infty$. Now, on the left hand side, we have more zeros than on the other side (as initially, we had the same number of zeros). Therefore, we are back to case (i).

Note however, that we can rule out negative α in a different way, if instead of insisting on preparing the exact output state, we allow for preparing an ε-approximation, with arbitrary accuracy (as discussed in Prop. 4). Therefore, in case (iii) considered above we can add to both sides an ancilla in a pure state of the same dimension (which is actually needed only when p and q both have no zeros). Then both input and ouptut will have the same number of zeros. But as the desired output we can take an approximation of it which is full rank. Then the transition is governed solely by the conditions with positive α. The returned state of ancilla is now only approximately pure, but the accuracy is arbitrarily good. Note that we can choose the approximation in such a way that it affects only the ancilla; the original output state is not changed and will be produced exactly.

The above two methods of ruling out negative α do not differ very much: in the first one we input some pure state of large dimension and return a pure state of dimension smaller by 1, while in the second we input a qubit in a pure state and return it with arbitrarily good approximation. However, in both cases one has to borrow a perfectly pure state, which in thermodynamic context would mean initially investing an infinite amount of work.

Appendix C: Catalytic thermal operations

In Sec. B, we considered catalytic noisy operations, where apart from using noisy operations, one can add any catalyst, provided it is returned in the same state. To obtain a full picture of thermodynamic interactions, we consider a generalisation of these operations to the setting where we have non-trivial Hamiltonians, and a class of *thermal operations* introduced in [1, 16, 17]. The relevant objects now come in pairs (ρ, H) - the system's state and its Hamiltonian.

Consider a fixed temperature T. Then given a system S in some initial state ρ, thermal operations include the following procedures:

- adding any system ρ_β^R which is the thermal state w.r.t. an arbitrary Hamiltonian H_R at temperature T,

- applying arbitrary unitary that commutes with the joint Hamiltonian of system and reservoir $H_S + H_R$,

- removing a system by performing partial trace.

It is clear that thermal operations preserve the thermal state. Conversely, for states diagonal in energy basis, any operation which preserves the thermal state belongs to the class of thermal operations as proved in [1, 16].

We now consider catalytic thermal operations, where in addition to thermal operations, one can add an ancilla called the catalyst (ρ_C, H_C), provided it is returned in the same state (in product form with the other systems). In other

words, (ρ, H) can be catalytically transformed into (ρ', H') if there exists catalyst (ρ_C, H_C) such that $(\rho \otimes \rho_C, H + H_C)$ can be transformed into $(\rho' \otimes \rho_C, H' + H_C)$. Demanding that the catalyst be returned uncorrelated from the system is natural, since one is likely to want to re-use the catalyst over many cycles, and having correlations between the catalyst and the system could result in inefficiencies later on. One could also imagine that the purification of the catalyst is held somewhere, and require that the catalyst and its purification remain close to its original state.

Appendix D: The zeroeth law and the emergence of temperature as equivalence relations required for thermodynamics to be a non-trivial resource theory

In this section, we spell the proof of the zeroeth law in this framework of thermal operations, which singles out Gibbs states as the unique free resource in thermodynamics. More precisely, if we consider the resource theory framework of thermal operations, which allows arbitrary energy preserving unitaries across the global system, then the only resource states that, when allowed for free, does not give rise to trivial state conversion conditions is that of the Gibbs state. It is clear that this is directly related to work extraction via thermal operations, since one should make sure that work cannot be extracted from a freely allowed resource. Otherwise, one could, by using indefinite amounts of free resources, extract enough work to facilitate any state transformation. Therefore, we want to show that given a system Hamiltonian H_A, and the corresponding thermal state τ_A with some fixed temperature T, then by allowing arbitrarily many copies of any other state $\rho_A \neq \tau_A$, one can always extract work deterministically by thermal operations.

Let us first show that this is true for states diagonal in the energy eigenbasis. The case where coherences between energy eigenstates exist can be dealt with, as shown in [27], by decohering multiple copies of the state in its energy subspace, and invoking Landauer's principle to extract work. We shall base on the result of Pusz and Woronowicz [28] who introduced the notion of *passive* states, i.e. the states whose energy cannot be decreased by arbitrary cyclic Hamiltonian evolution. They further introduced a notion of *completely passive* states:

Definition 6 (Completely passive states). *A state ρ is completely passive iff for all $n \in \mathbb{Z}^+$, $\rho^{\otimes n}$ is also passive.*

They proved for general quantum systems described by a C^* algebra that the only completely passive states are either so called KMS states or ground states. Lenard [29] has translated their results to the case of finite-dimensional systems. For such systems one has:

Fact 1. *Consider a state ρ corresponding to some Hamiltonian H, and let $\{p_i\}$ and $\{E_i\}$ be the eigenvalues of ρ and H respectively. Then ρ is passive, iff $[\rho, H] = 0$, and for any i, j, $E_i > E_j$ implies that $p_i \leq p_j$.*

Moreover a state which is completely passive is either ground state or a Gibbs state (since in finite dimensional state, the KMS state is unique and it is just the Gibbs state).

Either from the very definition or from the above characterization one sees that given any non-passive state ρ_{np}, one can extract on *average* a non-zero amount of work by performing a population inversion (a switch of energy levels). By invoking typicality arguments, we show that by performing extraction over many identical copies of ρ_{np}, an amount of work closely related to average work can always be extracted except with small probability. Then to conclude, one uses the mentioned result, that any state which is not a Gibbs state or a ground state, becomes non-passive, once we take sufficiently many copies.

Before beginning the proof, we state for completeness Theorem 7 which is the main tool we use to invoke typicality.

Theorem 7 (Hoeffding inequality). *Consider $x = \sum_{i=1}^{N} h_i$, where $h_1, \cdots, h_N$ are independent random variables such that for every $i, h_i \in [a_i, b_i]$. Denote $R_i = b_i - a_i$. Then for any $\alpha > 0$, the probability*

$$P\left[|x - E[x]| \geq \alpha N\right] \leq e^{-\frac{2\alpha^2 N^2}{\sum_i R_i^2}}. \tag{D1}$$

Theorem 8. *Given any non-passive, diagonal state ρ corresponding to some Hamiltonian H. Then for any probability $\varepsilon > 0$, there exists m such that given m copies of ρ, it is possible to extract a non-zero amount of work, except with probability ε.*

Proof. There are two main steps in this proof: first we construct the unitary that performs work extraction from a single copy of ρ, while storing the work in a battery system B similar to that of [27]. Subsequently, we form the joint unitary over the m systems and the battery system, and use the Hoeffding inequality to bound the amount of extracted work, albeit with some small failure probability.

Consider a non-passive ρ_{A_k} on system A_k, $1 \leq k \leq m$. Then by the definition of passivity, there exists some i, j where $E_i > E_j$, $p_i > p_j$ holds. Construct the battery as a harmonic osciilator system with Hamiltonian $H_B =$

$\sum_{n=1}^{N} n\hbar\omega|n\rangle\langle n|$, where $\hbar\omega = E_i - E_j$. Define the lowering operator $\hat{a}_B = \sum_{i=1}^{N} |i-1\rangle\langle i|$ and the joint energy-preserving unitary over system A_k and B,

$$U_k = |i\rangle\langle j|_{A_k} \otimes \hat{a}_B + |j\rangle\langle i|_{A_k} \otimes \hat{a}_B^\dagger + |i\rangle\langle i|_{A_k} \otimes |N\rangle\langle N|_B + |j\rangle\langle j|_{A_k} \otimes |0\rangle\langle 0|_B + \sum_{r\neq i,j} |r\rangle\langle r|_{A_k} \otimes \mathbb{I}_B. \tag{D2}$$

Clearly, this unitary acted upon the initial joint state $\rho_{A_k} \otimes |m\rangle\langle m|_B$ extracts $\hbar\omega \geq 0$ amount of work into the battery system with probability p_i, and $-\hbar\omega$ with probability p_j, while the expectation value of work extracted is given by the difference in expected energy in the battery system, $\langle W\rangle = \hbar\omega(p_i - p_j) > 0$.

Now, similarly, consider the initial state $\rho_{A_1} \otimes \cdots \otimes \rho_{A_m} \otimes |m\rangle\langle m|_B$, and the unitary transformation $U_m \cdots U_1$. Each unitary raises, lowers or leave unchanged the battery state with certain probabilities; hence this operation can be represented with a string $x = x_1 \cdots x_m$ numbers, where for all $y = 1, \cdots, m$,

$$p(x_y = c) = \begin{cases} p_i & (c=1); \\ p_j & (c=-1); \\ 1 - p_i - p_j & (c=0). \end{cases} \tag{D3}$$

The total amount of work extracted in this process is equal to $W_T = x_t\hbar\omega$, where $x_T = \sum_{i=1}^{m} x_i$, while the expectation value of x_T is $\langle x_T\rangle = m(p_i - p_j) > 0$. Since $x_1, \cdots, x_m$ are i.i.d. random variables with bounded values between -1 and 1, we can invoke Theorem 7 for some small number $\alpha > 0$, obtaining

$$P[x_T \leq \langle x_T\rangle - \alpha m] \leq P[|x_T - \langle x_T\rangle| \geq \alpha m] \leq \exp\left[\frac{-2\alpha^2 m^2}{\sum_{i=1}^{m} 2^2}\right] = \exp\left[\frac{-\alpha^2 m}{2}\right] = \varepsilon, \tag{D4}$$

Therefore, we conclude that $W_T \geq (\langle x_T\rangle - \alpha m)\hbar\omega > 0$ except with probability ε. For any $\varepsilon > 0$, pick some small α and $m = \frac{2}{\alpha^2}\log\frac{1}{\varepsilon}$ suffices. $\square$

Theorem 9. *Given a fixed Hamiltonian H, if ρ' is not the Gibbs state or the ground state corresponding to the Hamiltonian H at a certain temperature T, then for any $\varepsilon > 0$, there exists n such that given n copies ρ', it is possible to extract a non-zero amount of work, except with probability at most ε.*

Proof. The proof comes from a straightforward realisation that any such ρ' is not *completely* passive, hence there exists some positive integer a such that $\rho'^{\otimes a}$ is non-passive. One can then invoke Theorem 8 with $\rho = \rho'^{\otimes a}$. For any $\varepsilon > 0$, pick some small α and $n = am = a\frac{2}{\alpha^2}\log\frac{1}{\varepsilon}$ suffices. $\square$

Appendix E: Catalysis with general Hamiltonians

We now turn to the case of the full theory of thermodynamics, where we have an interplay between energy and entropy. In this case, the Hamiltonian of the system and reservoir are arbitrary. To derive the conditions for state transformations, we will need a generalisation of the majorisation condition, known as d-majorisation, which we describe in E 1. We then derive the catalytic version of this, in E 3, theorem 17. Next we show that it is sufficient to consider catalysts diagonal w.r.t its Hamiltonian, when the initial state is already block diagonal in the energy eigenbasis. We can then apply our results to the case of thermodynamics to derive second laws – these are stated seperately in Section F. We will later in Section G discuss the case where the thermodynamic processes do not return the catalyst in exactly the same state, but only approximately close.

1. d-majorization

Definition 10. *Given probability distributions p, q, p', q', we say that (p, q) d-majorizes (p', q') if and only if for any convex function g,*

$$\sum_i q_i g\left(\frac{p_i}{q_i}\right) \geq \sum_i q_i' g\left(\frac{p_i'}{q_i'}\right). \tag{E1}$$

We denote this as $d(p||q) \succ d(p'||q')$.

The Birkhoff-von Neumann theorem relates majorization to transition between states, which we state here:

Theorem 11. *For two probability distributions p and p' the following two conditions are equivalent:*

(i) p majorizes p'.

(ii) there exists a channel Λ such that

$$\Lambda(p) = p', \quad \Lambda(\eta) = \eta, \tag{E2}$$

where η is the uniform distribution.

The generalization of Birkhoff-von-Neumann theorem to d-majorization is as follows:

Proposition 12 (Theorem 2, [30])**.** *For probability distributions p, p', q, q' the following two conditions are equivalent:*

(i) (p, q) d-majorizes (p', q'),

$$d(p\|q) \succ d(p'\|q'). \tag{E3}$$

(i) There exists a channel Λ such that

$$\Lambda(p) = p', \quad \Lambda(q) = q'. \tag{E4}$$

In fact it has been shown that a particular limited set of convex functions is sufficient. In the case when $q = q'$, the conditions can be expressed by the so-called *thermo-majorization* diagrams identified in [1]. The thermo-majorization diagrams can be easily extended also to the case $q \neq q'$, although this is not relevant for our application to thermodynamics in which $q = q'$ is the Gibbs state.

One can see that Proposition 12 implies Theorem 11, by taking $q = q' = \eta$ and noting that

$$p \succ p' \quad \leftrightarrow \quad d(p\|\eta) \succ d(p'\|\eta). \tag{E5}$$

A natural question is whether there is a trumping analogue for d-majorization. We show that this is the case in section E 3, where we prove a version of Prop. 12 allowing catalysis. We will recover analogous relations to the Klimesh-Turgut ones in the case where $q = q' = \eta$. We illustrate the situation in Table I.

	standard majorization	d-majorization
no catalysis	$d(p\|\eta) \succ d(p\|\eta)$	$d(p\|q) \succ d(p'\|q')$
	$\exists \Lambda: \ \Lambda(p) = p', \quad \Lambda(\eta) = \eta$	$\exists \Lambda: \ \Lambda(p) = p', \quad \Lambda(q) = q'$
	$\forall l \sum_{i=1}^{l} p_i \geq \sum_{i=1}^{l} p_i'$	comparing diagrams (Fig. **??**)
catalysis	$D_\alpha(p\|\eta) \geq D_\alpha(p'\|\eta)$	$D_\alpha(p\|q) \geq D_\alpha(p'\|q')$
	$\exists \Lambda, r: \ \Lambda(p \otimes r) = p' \otimes r, \quad \Lambda(\eta \otimes \tilde{\eta}) = \eta \otimes \tilde{\eta}$	$\exists \Lambda, r, s: \ \Lambda(p \otimes r) = p'_\epsilon \otimes r, \quad \Lambda(q \otimes s) = q' \otimes s$

TABLE I: Partial orderings as criteria for state transformations. p'_ϵ is an arbitrarily good approximation of p'.

2. Notations and technical tools

Before setting out to prove the main result in Section E 3, we list several technical tools and lemmas that we will use.

We will start by describing an embedding channel, which will be used later to prove the above results. Consider the simplex of probability distributions $\{p_i\}_{i=1}^{k}$ and a set of natural numbers $\{d_i\}_{i=1}^{k}, i = 1, \ldots, k$, and let $N = \sum_{i=1}^{k} d_i$. We define the embedding

$$\Gamma(p) = \oplus_i p_i \eta_i, \tag{E6}$$

where $\eta_i = \{\frac{1}{d_i}, \ldots, \frac{1}{d_i}\}$ is the uniform distribution. More clearly, the image can be writen as the following N-dimensional probability distribution:

$$\Gamma(p) = \left\{ \underbrace{\frac{p_1}{d_1}, \ldots, \frac{p_1}{d_1}}_{d_1}, \ldots, \underbrace{\frac{p_k}{d_k}, \ldots, \frac{p_k}{d_k}}_{d_k} \right\}. \tag{E7}$$

The inverse map Γ^* acts on the space of N-dimensional probability distributions $\tilde{p}$, where

$$\tilde{p} = \oplus_{i=1}^{k} \tilde{p}^{(i)}, \tag{E8}$$

with each $\tilde{p}^{(i)} = \{\tilde{p}_1^{(i)}, \cdots, p_{d_i}^{(i)}\}$ being a (unnormalized) d_i-dimensional probability distribution. Γ^* can be written as

$$\Gamma^*(\tilde{p}) = r, \tag{E9}$$

with $r = \{r_i\}_{i=1}^{k}$ being a normalized probability distribution with the form $r_i = \sum_{j=1}^{d_i} \tilde{p}_j^{(i)}$. The maps Γ and Γ^* are channels, and for all probability distributions p we have $\Gamma^*(\Gamma(p)) = p$. Moreover, for the specific state $\gamma = \{\frac{d_1}{N}, \ldots, \frac{d_k}{N}\}$,

$$\Gamma(\gamma) = \mathbb{I}/N \tag{E10}$$

is the uniform N-dimensional distribution. Recall that $\sum_{i=1}^{k} \frac{d_i}{N} = N \cdot \frac{1}{N} = 1$, and γ is normalized.

Also, we will be making use of the following simple lemmas, which we state here for the reader's convenience. Lemma 13 relates the Rényi divergence $D_\alpha(p\|q)$ to $D_\alpha(\Gamma(p)\|\Gamma(q))$. Lemma 14 describes the change in $D_\alpha(p\|q)$ when p is replaced by a mixture of distributions p and q. Lemma 15 is a technical tool that enables us to work with distributions containing irrational probability values, by introducing small corrections such that we need only to consider rational values. Lastly, Lemma 16 shows us when a channel can be writen as a direct sum of two channels acting disjointly on partitions of the total input/output space.

Lemma 13. *Let $p = \{p_i\}_{i=1}^{k}$ be an ordered probability distribution, and $\{d_i\}_{i=1}^{k}$ be natural numbers with $\sum_{i=1}^{k} d_i = N$. Define the following fine-grained, N-dimensional probability distribution*

$$\tilde{p} = \Big\{ \underbrace{\frac{p_1}{d_1}, \ldots, \frac{p_1}{d_1}}_{d_1}, \ldots, \underbrace{\frac{p_k}{d_k}, \ldots, \frac{p_k}{d_k}}_{d_k} \Big\}, \tag{E11}$$

and let γ be the k-dimensional probability distribution

$$\gamma = \Big\{ \frac{d_1}{N}, \ldots, \frac{d_k}{N} \Big\}. \tag{E12}$$

Then for $\alpha \in [-\infty, \infty]$ we have

$$D_\alpha(p\|\gamma) = D_\alpha(\tilde{p}\|\eta_N), \tag{E13}$$

with $\eta_N = (1/N, \ldots, 1/N)$ the uniform distribution. By (E10), this means that equivalently

$$D_\alpha(p\|\gamma) = D_\alpha(\Gamma(p)\|\Gamma(\gamma)). \tag{E14}$$

Proof. Let us first assume that $\alpha \notin \{-\infty, 0, 1, \infty\}$. Then

$$D_\alpha(p\|\gamma) = \frac{1}{\alpha - 1} \log \sum_{i=1}^{k} p_i^\alpha \left(\frac{d_i}{N} \right)^{1-\alpha} \tag{E15}$$

$$= \frac{1}{\alpha - 1} \log \sum_{i=1}^{k} d_i \left(\frac{p_i}{d_i} \right)^\alpha \left(\frac{1}{N} \right)^{1-\alpha} \tag{E16}$$

$$= \frac{1}{\alpha - 1} \log \sum_{i=j}^{N} \tilde{p}_j^\alpha \left(\frac{1}{N} \right)^{1-\alpha} = D_\alpha(\tilde{p}\|\eta_N), \tag{E17}$$

where note that in the third inequality we sum over the fine-grained distribution $\tilde{p}$, and for each $i \in \{1, \cdots, k\}$, $\tilde{p}$ contains d_i number of degenerate values $\frac{p_i}{d_i}$. For $\alpha \in \{-\infty, 0, 1, \infty\}$ one can obtain the relation by considering limits. Here we show this explicitly. For $\alpha = 0$ we have

$$D_0(p\|\gamma) = -\log \sum_{i:p_i \neq 0} \gamma_i = -\log \sum_{j:p_j \neq 0} \frac{d_i}{N} = -\log \sum_{i:\tilde{p}_i \neq 0} \frac{1}{N} = D_0(\tilde{p}\|\eta_N). \tag{E18}$$

For $\alpha \to \infty$, by using (A3) we have

$$D_\infty(p\|\gamma) = \log \min \left\{ \lambda : \forall i, \lambda \geq \frac{p_i}{d_i/N} \right\} = \log \min \left\{ \lambda : \forall i, \lambda \geq \frac{p_i/d_i}{1/N} \right\} = D_\infty(\tilde{p}\|\eta_N), \tag{E19}$$

and similarly for $\alpha \to -\infty$. Finally, for $\alpha \to 1$,

$$D_1(p\|\gamma) = \sum_{i=1}^{k} p_i \log \frac{p_i N}{d_i} = \sum_{i=1}^{k} d_i \frac{p_i}{d_i} \log \frac{p_i N}{d_i} = D_1(\tilde{p}\|\eta_N). \tag{E20}$$

$\square$

Lemma 14. *Let $p \neq q$ be distributions with q full rank. Then for all $\alpha \in (-\infty, \infty)$ and all $0 < \delta < 1$,*

$$D_\alpha((1-\delta)p + \delta q\|q) < D_\alpha(p\|q). \tag{E21}$$

Proof. The proof is given separately for different ranges of α.

In the range of $\alpha \in [0,1]$, (E21) is obtained by the joint convexity of the Rényi divergence, as proven in Theorem 11 of [3]. The special case of $\alpha = 1$ is also proven in [31]. For any two sets of distributions (A_1, B_1) and (A_2, B_2), and a parameter $0 < \delta < 1$, the Rényi divergence in this range of α satisfies

$$D_\alpha((1-\delta)A_1 + \delta A_2\|(1-\delta)B_1 + \delta B_2) \leq (1-\delta)D_\alpha(A_1\|B_1) + \delta D_\alpha(A_2\|B_2). \tag{E22}$$

Setting $B_1 = B_2 = q$, $A_1 = p$ and $A_2 = q$, then

$$D_\alpha((1-\delta)p + \delta q\|q) \leq (1-\delta)D_\alpha(p\|q) + \delta D_\alpha(q\|q) < D_\alpha(p\|q) \tag{E23}$$

since $D(q\|q) = 0$, $\delta > 0$ and $D_\alpha(p\|q) > 0$ for $p \neq q$.

For $\alpha > 1$ joint convexity does not hold. However, denote $r = (1-\delta)p + \delta q$, note that (E21) is equivalent to

$$\sum_i r_i^\alpha q_i^{1-\alpha} > \sum_{i=1}^{k} p_i^\alpha q_i^{1-\alpha}. \tag{E24}$$

We now make use of the fact that the function $f(x) = x^\alpha$ is convex for $\alpha > 1$, over all $x > 0$ (This be checked since the second derivative of $f(x)$ is positive over the range of $x > 0$). and since each $q_i^{1-\alpha} > 0$, the linear combination

$$F(p) = \sum_i q_i^{1-\alpha} p_i^\alpha \tag{E25}$$

is convex w.r.t. p. For any $p \neq q$, note that $F(p) > 1$ comes from the positivity of the Rényi divergences, since $D_\alpha(p\|q) = \frac{\text{sgn}(\alpha)}{\alpha-1} \log F(p)$. Also, note that for a fixed q, $F(q) = \sum_i q_i = 1$. This implies that for (E24),

$$\text{L.H.S.} \equiv F(r) \leq (1-\delta)F(p) + \delta F(q) \tag{E26}$$
$$= F(p) - \delta[F(p) - F(q)] \tag{E27}$$
$$< F(p) \equiv \text{R.H.S.} \tag{E28}$$

For $\alpha < 0$, whenever the kernel of p is non-empty, $D_\alpha(p\|q) = \infty$, However $D_\alpha((1-\delta)p + \delta q\|q)$ is finite since q has full rank, hence the desired inequality holds always. When p also has full rank, the same approach can be used as in $\alpha > 1$, since $f(x) = x^\alpha$ is also convex for positive x, and since $\frac{\text{sgn}(\alpha)}{\alpha-1} > 0$, (E21) is also equivalent to (E24). $\square$

Lemma 15. *Let $q = \{q_i\}_{i=1}^k$ be an ordered (descendingly) k-dimensional probability distribution of full rank, possibly containing irrational values. Then for any $\varepsilon > 0$, there exists a state $\tilde{q}$ such that*

(i) $\|q - \tilde{q}\| \leq \varepsilon$,

(ii) *Each entry of $\tilde{q}$ is a rational number, i.e. there exists a set of natural numbers $\{d_i\}_{i=1}^k$ such that $\tilde{q} = \{\frac{d_i}{N}\}_{i=1}^k$, with $\sum_{i=1}^k d_i = N$,*

(iii) *There exists a valid channel E such that $E(q) = \tilde{q}$ and for any other distribution p, $\|p - E(p)\| \leq O(\sqrt{\varepsilon})$.*

Proof. We prove this by constructing a specific $\tilde{q}$ that satisfies (i) and (ii), then we construct the corresponding channel E that maps q to $\tilde{q}$. Note that since q is ordered, the minimum value $\min_i q_i = q_k$. Firstly, choose any integer N such that $q_k > \frac{k}{\sqrt{N}} > \frac{k}{N}$. Now, for $i \in \{1, \cdots, k-1\}$, define

$$\tilde{q}_i = \frac{\lceil q_i N \rceil}{N} = \frac{d_i}{N}. \tag{E29}$$

Lastly, choose

$$\tilde{q}_k = 1 - \sum_{i=1}^{k-1} \tilde{q}_i = 1 - \frac{\sum_{i=1}^{k-1} d_i}{N} = \frac{d_k}{N}. \tag{E30}$$

It is clear that

$$\begin{aligned} 1 - \tilde{q}_k = \sum_{i=1}^{k-1} \tilde{q}_i &= \sum_{i=1}^{k-1} \frac{\lceil q_i N \rceil}{N} \\ &\leq \sum_{i=1}^{k-1} \frac{q_i N}{N} + \frac{k-1}{N} \\ &\leq 1 - q_k + \frac{k}{N}, \end{aligned} \tag{E31}$$

which means that $\tilde{q}_k \geq q_k - \frac{k}{N} > 0$ from our choice of N, hence $\tilde{q}$ is a valid distribution with rational probability values, and condition (ii) is satisfied. Note also that the statistical distance between q and $\tilde{q}$ is lower bounded by

$$\begin{aligned} \|q - \tilde{q}\| = \frac{1}{2} \sum_{i=1}^{k} |q_i - \tilde{q}_i| &= \frac{1}{2} \left[\sum_{i:q_i > \tilde{q}_i} (q_i - \tilde{q}_i) + \sum_{i:q_i \leq \tilde{q}_i} (\tilde{q}_i - q_i) \right] \\ &= \sum_{i:q_i > \tilde{q}_i} q_i - \tilde{q}_i \\ &= q_k - \tilde{q}_k \leq \frac{k}{N} = \varepsilon \end{aligned} \tag{E32}$$

which satisfies condition (i) by setting $\varepsilon = \frac{k}{N}$, which can be arbitrarily small by choice of N. The third inequality holds because both distributions are normalized,

$$\begin{aligned} \sum_{i:q_i > \tilde{q}_i} q_i + \sum_{i:q_i \leq \tilde{q}_i} q_i &= \sum_{i:q_i > \tilde{q}_i} \tilde{q}_i + \sum_{i:q_i \leq \tilde{q}_i} \tilde{q}_i = 1 \\ \sum_{i:q_i > \tilde{q}_i} q_i - \tilde{q}_i &= \sum_{i:q_i \leq \tilde{q}_i} \tilde{q}_i - q_i, \end{aligned} \tag{E33}$$

while the fourth inequality occurs since for $i = \{1, \cdots, k-1\}, q_i \leq \tilde{q}_i$ is not included in the summation.

Now, to prove (iii) we need to construct a valid channel, which is characterized by the transition probabilities $P(i \to j)$ going from state i to state j, where for all $i \in \{1, \cdots, k\}$,

$$\sum_{j=1}^{k} P(i \to j) = 1. \tag{E34}$$

To understand how we construct an appropriate channel, note that the channel has to increase slightly the probabilities q_i to $\tilde{q}_i$ for all $i \in \{1, \cdots, k-1\}$, while decreasing $\tilde{q}_k$ accordingly for the normalization to hold.

Bearing this in mind, let us denote the set $\mathcal{I} = \{i | 1 \leq i \leq k-1\}$ which contains the indices where $\tilde{q}_i \geq q_i$. Also define $\Delta_i = \tilde{q}_i - q_i$ and the sum $\Delta = \sum_{i \in \mathcal{I}} \Delta_i$. Note that $\tilde{q}_k = q_k - \Delta$ comes from the normalization of $\tilde{q}$.

Now, let us consider the channel E with the following transition probabilities:

$$p(i \to j) = \begin{cases} 1 & \text{for } i = j, & i, j \in \mathcal{I} \\ 0 & \text{for } i \neq j, & i, j \in \mathcal{I} \\ 1 - \frac{\Delta}{q_k} & \text{for } i = j = k \\ 0 & \text{for } j = k, & i \in \mathcal{I} \\ \frac{\Delta_j}{q_k}, & \text{for } i = k, & j \in \mathcal{I}. \end{cases} \tag{E35}$$

One can verify that the above transition probabilities satisfies (E34), hence they characterize a valid channel. The last step is to show that this channel causes only a small perturbation to any other state p.

$$||p - E(p)|| = p_k - \tilde{p}_k = \frac{\Delta}{q_k} p_k \leq \frac{\Delta}{q_k}. \tag{E36}$$

Note that $\Delta = q_k - \tilde{q}_k \leq \frac{k}{N}$, hence

$$||p - E(p)|| \leq \frac{k}{Nq_k} \leq \varepsilon \frac{\sqrt{N}}{\sqrt{k}} \frac{1}{\sqrt{k}} = \frac{\sqrt{\varepsilon}}{\sqrt{k}}, \tag{E37}$$

which holds since $\varepsilon = \frac{k}{N}$, and we have chosen N so that $q_k > \frac{k}{\sqrt{N}}$. $\square$

Lemma 16. *Consider a channel such that for some fixed n-dimensional probability distribution $t = (t_1, \ldots, t_l, 0, \ldots 0)$ the channel gives output $t' = (t'_1, \ldots, t'_l, 0, \ldots 0)$. Moreover, $\Lambda(w) = w$ holds for some full rank distribution w. Then this channel can be decomposed as $\Lambda = \Lambda_1 \oplus \Lambda_2$, where Λ_1 acts only on the first l-elements, mapping them onto the same group of elements, while Λ_2 acts similarly on the remaining $n - l$ elements.*

Proof. Consider the joint probability of two random variables (X, Y), given by the preserved distribution w and the channel. Let $X = 0$ denote the event that the inputs are from the first group (items from 1 to l), and $X = 1$ that they are from the second group (items from $l + 1$ to n), and Y denotes similar events for the outputs. Since the channel preserves w, we have that $P(Y = 0) = P(X = 0)$. Moreover, since the channel sends t into t', this means that $p(Y = 0|X = 0) = 1$. We then have

$$p(Y = 0) = p(Y = 0|X = 0)p(X = 0) + p(Y = 0|X = 1)p(X = 1) = p(Y = 0) + p(Y = 0|X = 1)p(X = 1) \tag{E38}$$

so that either $P(X = 1) = 0$ or $p(Y = 0|X = 1) = 0$. However, since w is of full rank, we know that $P(X = 1) > 0$. Therefore $p(Y = 0|X = 1) = 0$ must hold. We have therefore that $P(Y = 0|X = 0) = P(Y = 1|X = 1) = 1$ which means that the channel is direct sum of two channels, acting on two disjoint groups of elements $\{1, \cdots, l\}$ and $\{l + 1, \cdots, n\}$. $\square$

3. Catalytic d-majorization

In this section, we prove a crucial result (Theorem 17), which relates monotonicity of Rényi divergences to catalytic transformations. This can be viewed as both a generalization of trumping relations [24, 25] and the d-majorization result [30]. It also gives an operational interpretation to the Rényi divergences, answering the question posed in [2].

With the tools listed in Section E 2 in place, we can now proceed to state and prove the main theorem of this section.

Theorem 17. *Given probability distributions p, p', q, q', where q, q' has full rank. The following conditions are equivalent:*

(i) For all $\alpha \in (-\infty, \infty), D_\alpha(p||q) \geq D_\alpha(p'||q')$.

(ii) For any $\varepsilon > 0$, there exists probability distributions r, s of full rank, a distribution p'_ε and a stochastic map Λ such that

    1. $\Lambda(p \otimes r) = p'_\varepsilon \otimes r$,

    2. $\Lambda(q \otimes s) = q' \otimes s$,

    3. $||p' - p'_\varepsilon|| \leq \varepsilon$.

    Moreover, we can take $s = \eta$, where η is the uniform distribution onto the support of r.

Proof. "(i) → (ii)". To prove in this direction, we suppose that condition (i) holds, and construct a channel Λ that satisfies (ii), with some ε that can be chosen arbitrarily small. Let us consider the following two cases separately:

(A) The probabilities in q and q' are rational. Without loss of generality, they can be written as $q = (d_1/N, \ldots d_k/N)$ and $q' = (d'_1/N, \ldots, d'_k/N)$ for some sets of integers $\{d_i\}_{i=1}^k, \{d'_i\}_{i=1}^k$, such that $\sum_{i=1}^k d_i = \sum_{i=1}^k d'_i = N$.

With this, define two embedding channels Γ and Γ' associated with $\{d_i\}_{i=1}^k$ and $\{d_i'\}_{i=1}^k$ respectively. The fine-grained distributions of p and p' are given by

$$\tilde{p} = \Gamma(p) = \oplus_i \; p_i\eta_i$$
$$\tilde{p}' = \Gamma'(p'') = \oplus_i \; p_i'\eta_i',$$

where η_i, η_i' are maximally mixed distributions of dimensions d_i and d_i' respectively. Condition 1 together with Lemma 13 tells us that

$$D_\alpha(\tilde{p}||\eta_N) = D_\alpha(p||q) \geq D_\alpha(p'||q') = D_\alpha(\tilde{p}'||\eta_N), \tag{E39}$$

and hence by Proposition 4, we know that for any $\varepsilon > 0$, there exists a distribution $\tilde{p}_\varepsilon'$ such that $\|\tilde{p}' - \tilde{p}_\varepsilon'\|_1 \leq \varepsilon$ and $\tilde{p}$ can be trumped into $\tilde{p}_\varepsilon'$. Equivalently, there exists a probability distribution r (the catalyst) and a bistochastic map Φ such that

$$\Phi(\tilde{p} \otimes r) = \tilde{p}_\varepsilon' \otimes r. \tag{E40}$$

Note that by Lemma 16, r can be without loss of generality be of full rank, or in other words, the zeros in r do not affect finding such a bistochastic map. More precisely, let us consider the case where r does not have full rank, but has some rank d instead. Then by setting $t = \tilde{p} \otimes r$, $t' = \tilde{p}'' \otimes r$ and $w = \eta_N \otimes \eta$, we can use Lemma 16 to show that $\Phi = \Phi_1 \oplus \Phi_2$, where the channel Φ_1 gives us

$$\Phi_1(\tilde{p} \otimes r) = \tilde{p}'' \otimes r, \quad \Phi_1(\eta_N \otimes \eta_d) = \eta_N \otimes \eta_d, \tag{E41}$$

where η_d is the uniform distribution on the support of r. On the other hand if r is of full rank, then $\Phi = \Phi_1$.

Now, consider the following mapping

$$\tilde{\Lambda} = (\Gamma'^* \otimes \mathbb{I}) \circ \Phi_1 \circ (\Gamma \otimes \mathbb{I}). \tag{E42}$$

This map $\tilde{\Lambda}$ transforms $p \otimes r$ into $p'' \otimes r$, setting $\varepsilon = \delta$ satisfies conditions 1 and 3 in (ii). To satisfy condition 2, we want $\Lambda(q \otimes s) = q' \otimes s$ for some s. This is achieved by taking $s = \eta$, a uniform distribution of any dimension. Indeed, since $\Gamma(q) = \mathbb{I}/N$, $(\Gamma \otimes \mathbb{I})(q \otimes \eta)$ is also a maximally mixed distribution. Since Φ_1 is bistochastic, it preserves this distribution. Finally, by definition of Γ' we have $\Gamma'^*(\mathbb{I}/N) = q'$.

(B) The distributions q or q' contain irrational values. We show that in such cases, a similar approach in (A) can be used, by considering distributions $\tilde{q}$ which are rational and close to the original distributions. Note that for any real $\alpha > 0$, $D_\alpha(p||q)$ is a continuous function of both arguments p and q, whenever q is of full rank. For $\alpha < 0$, whenever p does not have full rank, both $D_\alpha(p||q)$ and $D_\alpha(p||\tilde{q})$ diverge to infinity. When p has full rank, continuity can be obtained by noting that $D_\alpha(p||q) = c \cdot D_{1-\alpha}(q||p)$ and $D_\alpha(p||\tilde{q}) = c \cdot D_{1-\alpha}(\tilde{q}||p)$ for some positive c, and $1 - \alpha > 0$, hence $D_{1-\alpha}(q||p)$ is continuous.

By Lemma 15, we use the channel E that maps q into $\tilde{q}$ while not perturbing p too much. More precisely, for any $\varepsilon > 0$, one can define a stochastic map E such that

$$E(q) = \tilde{q}, \quad \|\tilde{q} - q\| \leq \varepsilon, \tag{E43}$$

and $\|E(p) - p\| \leq O(\sqrt{\varepsilon})$ for any other state p. From the above discussion, we conclude that when $\|q - \tilde{q}\| \leq \varepsilon$, in the limit of $\varepsilon \to 0$, condition 1 implies that $D_\alpha(p||\tilde{q}) \geq D_\alpha(p'||\tilde{q}')$ holds as well. Since we can choose ε to be arbitrarily small, in the limit of $\varepsilon \to 0$,

$$D_\alpha(E(p)||\tilde{q}) = D_\alpha(p||q) \geq D_\alpha(p'||q') = D_\alpha(p'||\tilde{q}'). \tag{E44}$$

Following the first part of the proof (which established the result for rational q and q') we find that there is a catalyst r and a stochastic operation Λ such that

$$\Lambda(E(p) \otimes r) = p'' \otimes r, \quad \Lambda(E(q) \otimes \eta) = \tilde{q}' \otimes \eta, \tag{E45}$$

with η the maximally mixed distribution.

If q' also contains irrational values, we can similarly define a second correction map that maps q' into $\tilde{q}'$ while not perturbing p'' too much. By invoking Lemma 15 on q', we construct E' such that

$$E'(\tilde{q}') = q', \quad \text{and for any probability distribution } r, \|E'(r) - r\| \leq O(\sqrt{\varepsilon}). \tag{E46}$$

Denoting the inverse of E' as E'^*, hence $E'^*(\tilde{q}') = q'$. Choose an $r = E'^*(p'')$, then $\|E'(r) - r\| = \|p'' - E'^*(p'')\| \leq O(\sqrt{\varepsilon})$.

Our final stochastic map is given by $E'^* \circ \Lambda \circ E$, where

$$
\begin{aligned}
(E'^* \otimes \mathbb{I}) \circ \Lambda \circ (E \otimes \mathbb{I})(q \otimes \eta) &= (E'^* \otimes \mathbb{I}) \circ \Lambda(\tilde{q} \otimes \eta) \\
&= (E'^* \otimes \mathbb{I})(\tilde{q}' \otimes \eta) \\
&= q' \otimes \eta,
\end{aligned}
\tag{E47}
$$

and

$$
\begin{aligned}
(E'^* \otimes \mathbb{I}) \circ \Lambda \circ (E \otimes \mathbb{I})(p \otimes r) &= (E'^* \otimes \mathbb{I}) \circ \Lambda(E(p) \otimes r) \\
&= (E'^* \otimes \mathbb{I})(p'' \otimes r) \\
&= p''' \otimes r,
\end{aligned}
\tag{E48}
$$

with $p''' = E'^*(p'')$ such that

$$
\|p''' - p'\| \leq \|p'' - p'\| + \|p''' - p''\| \leq \varepsilon + O\left(\sqrt{\varepsilon}\right).
\tag{E49}
$$

Since ε can be chosen arbitrarily, the first part of the theorem follows.

"(ii) $\to$ (i)". Suppose that for all $\varepsilon > 0$ there exist probability distributions r, s, p'_ε and a stochastic map Λ such that

$$
\|p' - p'_\varepsilon\|_1 \leq \varepsilon,
\tag{E50}
$$

and

$$
\Lambda(p \otimes r) = p'_\varepsilon \otimes r, \quad \Lambda(q \otimes s) = q' \otimes s,
\tag{E51}
$$

and the support of s includes the support of r. Then by monotonicity of the Rényi divergences,

$$
D_\alpha(p'_\varepsilon \otimes r \| q' \otimes s) \leq D_\alpha(p \otimes r \| q \otimes s),
\tag{E52}
$$

which equals

$$
D_\alpha(p'_\varepsilon \| q') + D_\alpha(r \| s) \leq D_\alpha(p \| q) + D_\alpha(r \| s),
\tag{E53}
$$

by additivity. Since both r and s are full rank, $D_\alpha(r \| s)$ is finite, and can be subtracted from both sides. Lastly, we consider the limit $p'_\varepsilon \to p'$. Recall that as long as the second argument q' has full rank, for any $\alpha > 0$, $D_\alpha(p' \| q')$ is continuous w.r.t. both p' and q'. For $\alpha < 0$, whenever p' with full rank, continuity holds. If p' does not have full rank, $\lim_{\varepsilon \to 0} D_\alpha(p'_\varepsilon \| q') = \infty = D_\alpha(p' \| q')$. Hence we obtain

$$
D_\alpha(p' \| q') \leq D_\alpha(p \| q)
\tag{E54}
$$

for all $\alpha > 0$ and $\alpha < 0$. Since $D_0(p \| q) = \lim_{\alpha \to 0^+} D_\alpha(p \| q)$, the above inequality holds also for $\alpha = 0$.

$\square$

4. For diagonal input state of the system, a diagonal catalyst is enough

Here we will show that if the initial state of the system is block diagonal in the energy eigenbasis, then the diagonal of the output state does not depend on coherences of the catalyst but only on its diagonal. This means that if we are interested only in the diagonal of the output state of the system, we can replace the catalyst with its dephased version (i.e. put a diagonal catalyst that has the same diagonals as the original one). The conditional probabilities form the channel, which maps the initial diagonal to the final diagonal of the state of the system. By block diagonal, we mean that the state can be written as

$$
\rho = \sum \sigma_{ijk} |E_i, j\rangle \langle E_i, k|
\tag{E55}
$$

where $|E_i, j\rangle$ has energy E_i and has degenerate levels labled by j.

To see that only block diagonal catalysts are needed, we write the initial state as

$$\rho_{RSC}^{in} = \rho_R^{\text{in}} \otimes \rho_S^{\text{in}} \otimes \rho_C^{\text{in}}, \tag{E56}$$

where ρ_R^{in} is the heat bath, which is of course diagonal, ρ_C^{in} is the state of an arbitrary catalyst, and ρ_S^{in} is the state of the system which we assume to be diagonal. We then act with an energy-preserving unitary U and get the output state

$$\rho_{RSC}^{out} = U \rho_{RSC}^{in} U^\dagger. \tag{E57}$$

We now compute the diagonals element of ρ_S^{out}, and will see that they depend only on the diagonal elements of ρ_C^{in}. We have

$$\langle E_S | \rho_S^{\text{out}} | E_S \rangle = \sum_{E_R, E_C} \langle E_R, E_S, E_C | \rho_{RSC}^{out} | E_R, E_S, E_C \rangle \tag{E58}$$

(here and in the following we sum over energies as well as degeneracies). This can be written as

$$\langle E_S | \rho_S^{\text{out}} | E_S \rangle = \sum_{E_R, E_C} \sum_{E_R', E_C', E_S', E_C''} \langle E_R, E_S, E_C | U | E_R' E_S' E_C' \rangle \langle E_R', E_S', E_C' | \rho_{RSC}^{in} | E_R', E_S', E_C'' \rangle \times$$

$$\times \langle E_R', E_S', E_C'' | U^\dagger | E_R, E_S, E_C' \rangle$$

where we used that ρ_R^{in} and ρ_S^{in} are diagonal. Since U preserves energy, we have $E_R + E_S + E_C = E_R' + E_S' + E_C'$ as well as $E_R + E_S + E_C = E_R' + E_S' + E_C''$. This implies that $E_C' = E_C''$. We thus obtain

$$\langle E_S | \rho_S^{\text{out}} | E_S \rangle = \sum_{E_S'} p(E_S | E_S') \langle E_S' | \rho_S^{\text{out}} | E_S' \rangle, \tag{E59}$$

where the conditional probabilities are given by

$$p(E_S | E_S') = \sum_{E_R, E_C} \sum_{E_R', E_C'} |\langle E_R, E_S, E_C | U | E_R', E_S', E_C' \rangle|^2 \langle E_R' | \rho_R^{\text{in}} | E_R' \rangle \langle E_C' | \rho_C^{\text{in}} | E_C' \rangle. \tag{E60}$$

One easily finds that indeed this is a valid conditional probability distribution.

Appendix F: The Second Laws

In this section we formulate the state transformation conditions, namely the second laws of thermodynamics. We will first do this for states diagonal in the energy eigenbasis, and then for general state transformations. The former states are those ρ which satisfy $[\rho, H] = 0$. The conditions are given in terms of generalized free energies, which are defined as follows:

$$F_\alpha(\rho, H) = -kT[\ln Z + D_\alpha(\rho \| \rho_\beta)] = F(\rho_\beta, H) + kT D_\alpha(\rho \| \rho_\beta) \tag{F1}$$

where Z is the partition function for the Hamiltonian H, and ρ_β is the thermal state. Note that F_1 is the standard free energy, and that for thermal states and pure eigenenergy states, all free energies are equal to the standard one.

In Subsection F 4 we will present the quantum limitations which apply to all state transformations.

1. The second laws for states block diagonal in energy

Theorem 18 (Second laws for block diagonal states). *Consider a system with Hamiltonian H. Then a state ρ block diagonal in the energy eigenbasis can be transformed with arbitrary accuracy into another block diagonal state ρ' under catalytic thermal operations if and only if, for all $\alpha \in (-\infty, \infty)$,*

$$F_\alpha(\rho, H) \geq F_\alpha(\rho', H). \tag{F2}$$

Remark 19. *By continuity, the conditions can equivalently include $\alpha = \infty, -\infty$.*

Proof. We will prove it using Theorem 17. Consider initial and final states of the system ρ_S and ρ'_S. Suppose first that for $\alpha \in (-\infty, \infty)$, equation (F2) holds, which is equivalent to

$$D_\alpha(\rho_S \| \rho_S^\beta) \geq D_\alpha(\rho'_S \| \rho_S^\beta). \tag{F3}$$

We now need to show that one can transform the state ρ_S into an arbitrary good version of ρ'_S by catalytic thermal operations.

We denote p and p' to be eigenvalues of ρ_S and ρ'_S respectively, and $q = q' = \rho_S^\beta \otimes \rho_{\mathrm{anc}}^\beta$. Then, using Theorem 17, we get that there exists a channel Λ and uniform distribution η such that (i) Λ preserves the state $q \otimes \eta$, (ii) Λ sends $p \otimes r$ into $p'_\epsilon \otimes r$, where p'_ϵ approximates p'. Condition (i) means that we can take the catalyst system with trivial Hamiltonian. Thus we have

$$\Lambda(\rho_S \otimes \rho_C) = \rho_S^{\mathrm{out}} \otimes \rho_C, \tag{F4}$$

where $\|\rho_S^{\mathrm{out}} - \rho'_S\| \leq \epsilon$ and

$$\Lambda(\rho_S^\beta \otimes \rho_{\mathrm{anc}}^\beta \otimes \rho_C^\beta) = \rho_S^\beta \otimes \rho_{\mathrm{anc}}^\beta \otimes \rho_C^\beta, \tag{F5}$$

where ρ_C^β is the maximally mixed state on catalyst system C, i.e. Λ preserves the thermal state of the system SC and the ancilla. However thermal operations are precisely the operations that preserve the thermal state [1, 16]. Thus the required transition can be made by catalytic thermal operations.

Conversely, let us assume that for given states ρ_S and ρ'_S there exists a quantum channel Λ, and a system C with the hamiltonian H_C, and state ρ_C such that

$$\Lambda(\rho_S^\beta \otimes \rho_C^\beta) = \rho_S^\beta \otimes \rho_C^\beta, \quad \Lambda(\rho_S \otimes \rho_C) = \rho_S^{out} \otimes \rho_C. \tag{F6}$$

where $\|\rho_S^{\mathrm{out}} - \rho'_S\| \leq \epsilon$. Now, let us note that by thermal operations, one can make any state diagonal in the energy basis, namely one can apply random phases (by using the Birkhoff primitive of [17] which supplies random noise and unitaries that change phases). In Sec. E 4 we have shown that since the input and output states are diagonal, we can take the state ρ_C to be diagonal too, and the above condition reads

$$\Lambda_{\mathrm{cl}}(q \otimes s) = q \otimes s, \quad \Lambda_{\mathrm{cl}}(p \otimes r) = p'_\epsilon \otimes r, \tag{F7}$$

Thus we can apply theorem 17, obtaining that

$$D_\alpha(\rho_S \| \rho_S^\beta) \geq D_\alpha(\rho'_S \| \rho_S^\beta) \tag{F8}$$

for all real α which is equivalent to (F2). $\square$

2. Getting rid of second laws with negative α

In the paradigm of catalytic thermal operations it is assumed that the catalyst is returned exactly. If we had required that the catalyst is returned only with good fidelity, all restrictions would be lifted, and there will be no conditions for thermodynamic transitions at all. This happens when the catalyst's dimension is free to be arbitrarily large. In such cases, fidelity is not a good criterion for closeness any more (we discuss this in more detail in Section G of the Supplementary Information).

However, in addition to the catalyst, we can consider borrowing some system with fixed size (e.g. a qubit) in a pure state, given that we will return it with arbitrary good fidelity. As we will argue now, this will lift all the conditions on negative α. However since creating a pure state from a thermal state without additional resource requires infinite work, we will also consider approximate versions.

Here is the result, where we allow the use of an exactly, pure qubit.

Theorem 20. *If we can use catalytic thermal operations, and are allowed to borrow a qubit with a trivial Hamiltonian in the state $|0\rangle\langle 0|$ and have to return it with arbitrary accuracy, then a state ρ block diagonal in the energy eigenbasis can be transformed with arbitrary accuracy into another block diagonal state ρ' if and only if, for all $\alpha \geq 0$*

$$F_\alpha(\rho, H) \geq F_\alpha(\rho', H). \tag{F9}$$

Proof. Suppose first that one can transform the state $\rho_S \otimes |0\rangle\langle 0|$ into arbitrary good version of $\rho'_S \otimes |0\rangle\langle 0|$. Then we proceed as in the proof of Theorem 17 using monotonicity, additivity of D_α and noticing that D_α is finite for $|0\rangle\langle 0|$ for $\alpha > 0$, we get

$$D_\alpha(\rho_S||\rho_S^\beta) \geq D_\alpha(\rho'_S||\rho_S^\beta) \tag{F10}$$

for $\alpha \geq 0$, so that conditions (F9) are satisfied. Conversely, assume that the latter conditions are satisfied (hence equivalently (F10) hold). However for $\alpha < 0$, D_α is infinite on the left side, and finite on the right, if for the output side we return the qubit in a state which is of full rank, but arbitrarily close to it's original state. Moreover $D_\alpha(\rho_S \otimes |0\rangle\langle 0| \,||\rho_{\beta\,SC}) = D_\alpha(\rho_S||\rho_{\beta\,S}) + D_\alpha(|0\rangle\langle 0| \,||\rho_{\beta\,C})$ and same for S'. Thus we get that for all real α

$$D_\alpha(\rho_S||\rho_S^\beta) \geq D_\alpha(\rho'_S||\rho_S^\beta). \tag{F11}$$

and by Theorem 18 we can transform the state $\rho_S \otimes |0\rangle\langle 0|$ into arbitrary good version of $\rho'_S \otimes |0\rangle\langle 0|$. $\qquad\square$

We thus borrow a pure state qubit, and return it with full rank, but arbitrarily close to being pure, to get rid of the conditions for negative α. However, let us note, that we might borrow a noisy version of $|0\rangle\langle 0|$ namely $(1-\epsilon)|0\rangle\langle 0| + \epsilon|1\rangle\langle 1|$. It costs a finite amount of work to create it from the thermal state. If $F_\alpha(\rho_S^{\rm in}, H) \geq F_\alpha(\rho_S^{\rm out}, H)$ for $\alpha \geq 0$, we can pretend that the ancilla is in pure state $|0\rangle\langle 0|$ and apply the map from Theorem 20. We will obtain the output state arbitrarily close to

$$(1 - \epsilon)\rho_S^{\rm out} \otimes |0\rangle\langle 0|_{\rm anc} + \epsilon\rho'_{\rm S,anc}. \tag{F12}$$

We can then return the ancilla in the same state (by depolarizing a bit if needed), and obtain the output state $\rho_S^{\rm out}$ with accuracy ϵ in trace norm. However as we will discuss in section G, if there are no further restrictions on the available catalyst, then closeness in trace distance is not a suitable demand on the returned catalyst in thermodynamic transformations.

3. Borrowing ancilla with a nontrivial Hamiltonian

There is another approach, that might eliminate some of the conditions F2, yet is again not acceptable. Namely, we can borrow a qubit with nontrivial Hamiltonian $H = E|E\rangle\langle E|$ in state $\rho_\epsilon = (1-\epsilon)|0\rangle\langle 0| + \epsilon|E\rangle\langle E|$ and return thermal state ρ_β. If we take $\epsilon \geq \frac{e^{-\beta E}}{Z}$, we have $||\rho_\epsilon - \rho_\beta||_1 \leq \epsilon$. On the other hand, for any $\alpha > 1$, if we set e.g. $\epsilon = 1/E^2$ and use the approximation that E is large, then we have

$$
\begin{aligned}
D_\alpha(\rho_\epsilon||\rho_\beta) &= \frac{1}{\alpha-1}\log\left[\left(1 - \frac{1}{E^2}\right)^\alpha\left(1 - \frac{e^{-\beta E}}{Z}\right)^{1-\alpha} + \left(\frac{1}{E^2}\right)^\alpha\left(\frac{e^{-\beta E}}{Z}\right)^{1-\alpha}\right] \\
&\gtrsim \frac{1}{\alpha-1}\log\left[\left(\frac{1}{E^2}\right)^\alpha\left(\frac{e^{-\beta E}}{Z}\right)^{1-\alpha}\right] \\
&\geq \frac{\alpha}{\alpha-1}\log\frac{1}{E^2} - \log\frac{e^{-\beta E}}{Z} \\
&\geq \frac{1}{\ln 2}\beta E - \frac{2\alpha}{\alpha-1}\log E,
\end{aligned}
\tag{F13}
$$

which diverges for large E. Here, we used the approximation that the first term in the logarithm does not contribute much, since $\left(\frac{e^{-\beta E}}{Z}\right)^{1-\alpha}$ is large for $\alpha > 1$. Furthermore, $Z = 1 + \frac{e^{-\beta E}}{Z} \approx 1$. So, if we consider the transition

$$\rho_S^{\rm in} \otimes \rho_\epsilon \to \rho_S^{\rm out} \otimes \rho_\beta, \tag{F14}$$

then for any fixed $\alpha_0 > 1$, if we pick high energy E, we obtain that the D_α of the left hand side can be arbitrarily large, and the transition is possible, once conditions for $\alpha < \alpha_0$ are satisfied. However in this approach, we are allowed to borrow something very expensive, in the sense that it requires a large amount of work to be created from the thermal state, and return something useless - a thermal state. In other words there is a large *work distance* between the state we return and the state we have borrowed (see section G 3).

It is interesting to compare the two situations: to eliminate negative α we borrow a pure ancilla state. The Hamiltonian of the ancilla is trivial, and the state is much more pure than the thermal one. That's why it is

(infinitely) expensive. To eliminate $\alpha > 1$, we have the opposite: we take a Hamiltonian of the ancilla, such that thermal state is very pure, and we borrow a state which is *more mixed* than the thermal state, and for this reason also expensive. If we borrow both ancillas, we are left with conditions $\alpha \in [0, 1+\delta]$, with δ depending on how large an energy we will have in one of the ancillas.

4. Quantum second laws: limitations for states that are not diagonal in energy basis

For states that are not diagonal in energy basis, we report the following limitations:

Proposition 21 (Quantum limitations). *For a state ρ to be transformed with arbitrary accuracy into another state ρ' (both not necessary diagonal in energy basis) we require*

i) For $\alpha \geq \frac{1}{2}$,

$$S_\alpha(\rho||\rho_\beta) \geq S_\alpha(\rho'||\rho_\beta), \tag{F15}$$

ii) For $\frac{1}{2} \leq \alpha \leq 1$,

$$S_\alpha(\rho_\beta||\rho) \geq S_\alpha(\rho_\beta||\rho'), \tag{F16}$$

iii) For $-1 \leq \alpha \leq 2$,

$$\tilde{S}_\alpha(\rho||\rho_\beta) \geq \tilde{S}_\alpha(\rho||\rho_\beta). \tag{F17}$$

This proposition follows directly from monotonicity under completely positive and trace-preserving maps, and additivity of the quantum Rényi divergences associated with the generalised free energies, as discussed in Section A 3. E.g. we have

$$\begin{aligned} S_\alpha(\rho \, ||\rho_\beta) &\geq S_\alpha(\Lambda(\rho)||\Lambda(\rho_\beta)) \\ &\geq S_\alpha(\rho'||\rho_\beta) \end{aligned} \tag{F18}$$

with the first inequality due to monotonicity of S_α under completely positive trace-preserving maps Λ, and the second line following from the fact that if $\Lambda(\rho) = \rho'$ is a thermal operation, it preserves the thermal state. The reason for considering condition (F16) only for $\frac{1}{2} \leq \alpha \leq 1$, is that for $\alpha \geq 1$, the entropies would diverge for non full rank catalyst, so may not be monotones if we use such catalysts. As before, by using a pure state which is returned arbitrarily close to pure, we will automatically satisfy Equation (F17) for $\alpha \leq 0$ and we may thus only consider $0 \leq \alpha \leq 2$.

These conditions are like second laws, in that they are necessary conditions which must be respected during a thermodynamic transition, however, they are not sufficient. This follows from the fact that there are operations which preserve the thermal state, which are not thermal operations, and which can transform a state which is block diagonal in the energy eigenbasis, to one which is not [32]. Such a transition is impossible by thermal operations since thermal operations cannot transform a block diagonal state to one which is not. However, such a transition will respect any second law which also hold for operations which preserve the thermal state, as is the case with our quantum second laws.

5. Application: Landauer erasure with a quantum Maxwell demon

As an application of the above second laws, we consider a special case of a Maxwell demon with a memory Q in state ρ_Q, who wants to reset a system S in state ρ_S to some pure state. One imagines that the demon's memory is correlated in some way with the system so that the total state of demon plus system is ρ_{SQ}. The demon wishes to reset the state of the system by transforming it to some pure state, but the state of the demon's memory should not change. This can be seen as a fully quantum version of the standard Maxwell demon/Landauer erasure scenario, and has been considered in the case of a trivial Hamiltonian[33, 34] and when the Maxwell demon does not have access to ancilliary systems. The result of [33] gave a thermodynamic interpretation to the notion of negative information [35]. We will see here, that the amount of work which is required for such an operation, is quantitatively different when all thermodynamical operations are considered. This suggests that in the single-shot scenario with ancillas, the notion of partial information takes on a different form, which we shall now derive.

In fact, the result follows immediately from the work distance of Equation (**??**), namely, we have that the cost of resetting the system W_{reset} to the pure state ψ_S^E with energy E is given by

$$W_{\text{reset}} = -\inf_\alpha kT[D_\alpha(\rho_{QS}\|\rho_{\beta QS}) - D_\alpha(\rho_Q \otimes \psi_S^E\|\rho_{\beta QS})]$$
$$= -\inf_\alpha kT[D_\alpha(\rho_{QS}\|\rho_{\beta QS}) - D_\alpha(\rho_Q\|\rho_{\beta Q})] + E + kT \log Z_S \tag{F19}$$

In the case when the Hamiltonian is trivial, this reduces to

$$W_{\text{reset}} = kT \sup_\alpha [H_\alpha(\rho_{QS}) - H_\alpha(\rho_Q)] \tag{F20}$$

When this quantity is negative, the demon can reset the system to a pure state, and not only does this not cost work, but the corresponding amount of work is actually gained. Not only can thermodynamical work be negative, as shown in [33], but for the single shot case, it can be even more negative! In the sense that catalytic operations can be used to gain more work than would be otherwise possible. Note that this gives the difference of Rényi entropies of Equation (F20) an operational interpretation, very similar to that enjoyed by the conditional von-Neumann entropy in the case of identically and independently distributed states. Finally, in the case of standard Landauer erasure of a qubit with a non-trivial Hamiltonian (where we don't have a memory and start in the thermal state), Equation (F19) gives what you might expect, namely $W_{\text{reset}} = E + kT \log Z_S$.

Appendix G: Approximate catalysis

So far we considered exact catalysis, i.e. the process is perfectly cyclic and the catalyst should be in the same state as it initially was. However, this is usually an unreasonable demand because in physical processes there are some unavoidable inaccuracies. Therefore, we ask what are the conditions for transformations, when the catalyst ρ_C^{in} can be returned in a state ρ_C^{out} that is merely "close" to the initial state.

1. How to quantify "approximate"

As already discussed in the introduction, it turns out that what is meant by "close" matters greatly. At first glance, one might be tempted to demand that close should mean that ρ_C^{in} is close to ρ_C^{out} in terms of the fidelity, or essentially equivalently, the trace norm distance. The trace norm distance between two states ρ and σ can be written as

$$\|\rho - \sigma\|_1 = \max_{0 \leq M \leq \mathbb{I}} \text{tr}\left[M\left(\rho - \sigma\right)\right] . \tag{G1}$$

We say that ρ and σ are ϵ-close if $\|\rho - \sigma\|_1 \leq \epsilon$. It enjoys an appealing operational interpretation as being ϵ-close in trace norm distance means that if we were given states ρ and σ with probability $1/2$ each, then our probability of correctly distinguishing them by any physically allowed measurement is bounded by $1/2 + \epsilon/2$. In other words, being close in the trace distance means that the two states cannot be distinguished well by any physical process [45]. In terms of the catalyst, one might hence ask that $\|\rho_C^{\text{in}} - \rho_C^{\text{out}}\|_1 \leq \epsilon$ for some arbitrary small ϵ. This would be a mistake as we will see.

2. The embezzling state dilemma: when there is no second law

As mentioned already in the introduction, there is the phenomenon of embezzling [36] in entanglement theory, where at an expense of increasing the size of the catalyst, one can perform arbitrary transformation with arbitrary good fidelity, while returning the catalyst in a state arbitrarily close to the initial state. More specifically, for $H = 0$ we can adapt the results of [36] to show that for any ϵ, there exists a dimension d such that using the catalyst

$$\rho_C^{\text{in}} = \frac{1}{C(n)} \sum_{j=1}^n \frac{1}{j}|j\rangle\langle j| , \tag{G2}$$

allows us to transform any state ρ ϵ-close to any state ρ' such that $\|\rho_C^{\text{out}} - \rho_C^{\text{in}}\|_1 \leq \epsilon$. Here, $C(d)$ is a normalisation constant. In particular, there exists an n large enough, such that for any ϵ, we can use ρ_C^{in} to erase a state $\varphi = \sum_{i=1}^m p_i|i\rangle\langle i|$ to $|0\rangle$ via a unitary transformation taking $\rho_C^{\text{in}} \otimes \varphi$ to a state close to $\rho_C^{\text{in}} \otimes |0\rangle\langle 0|$. We first apply a unitary

on the state $\rho_C^{\text{in}} \otimes \varphi$ to produce a state $\omega = \sum_{i=1}^{nm} q_i |i\rangle\langle i|$ such that the eigenbasis of ω is the same as $\rho_C^{\text{in}} \otimes \varphi$, and the probabilities q_i are the same as the eigenvalues of $\rho_C^{\text{in}} \otimes \varphi$, but in decreasing order. One can then show [36] that $\sum_j \sqrt{q_j/jC(n)}$ goes to 1 as n goes to infinity, which implies that the fidelity $F(\omega, \rho_C^{\text{in}}) = \text{tr}\sqrt{\sqrt{\omega}\sqrt{\rho_C^{\text{in}}}} \to 1$. Since the fidelity upperbounds the trace distance via $F(\omega, \rho_C^{\text{in}}) \geq 1 - \frac{1}{2}\|\omega - \rho_C^{\text{in}}\|$ we have the desired result.

Another example of embezzling work occurs with $H = J$ and the pure state coherence resource used in [17]

$$|\psi\rangle = \frac{1}{\sqrt{d}} \sum_{j=1}^{d} |j\rangle \tag{G3}$$

with $|j\rangle$ energy eigenstates. If we apply the energy conserving operation which takes $|0\rangle_S |j\rangle_C \to \||1\rangle_S |j-1\rangle_C$ to the state $|0\rangle_S|\psi\rangle_C$, then by increasing d, the catalyst will be left in a state arbitrarily close to it's original state $|\psi\rangle$ in trace distance, even though one unit of work (energy) has been transferred to the system.

Demanding that the catalyst be returned close in the trace distance is thus too weak a condition. Intuitively, the reason why it is too weak is that we may still need to consume much work to obtain the original catalyst from its returned version.

If we therefore concieve of an approximately cylic process, as one which the working body is returned in a state which is ϵ close in fidelity to it's original form, then there is no second law. All state transformations are possible.

3. The work distance

We thus take an operational perspective on the problem of inexact catalysis. More precisely, we propose to take as a reference point, exact catalysis, and require that the catalyst should be returned in a form such that, only a small amount of work is needed in order restore it to the original form. This is natural, in the sense that if someone loans you a catalyst, then they would want it returned in such a way that it would not require a large amount of work to return it back into it's original state.

We thus consider the inexact transition

$$\rho_S^{\text{in}} \otimes \rho_C^{\text{in}} \to \rho_{SC}^{\text{out}} \tag{G4}$$

and require that to obtain from ρ_{SC}^{out} the original state of the catalyst and the required output state of the system, we need not input more than a small amount of work.

This prompts our definition of the work distance below. To make this precise, let us take a closer look how one can derive upper bounds on the amount of work needed for state transformation, namely to restore the catalyst to its original form. To input this amount of work, we can append a battery to either provide or extract work, to facilitate this transformation. We then apply our conditions for state transformation to the state and the battery together. The battery we use is a two level system called a *wit*, or *work bit*, introduced previously in [1]. The wit initially starts out in the energy eigenstate $\omega_i = |0\rangle\langle 0|$ which has energy $E_i = 0$. At the end of the process, the battery is in another energy eigenstate $\omega_f = |W\rangle\langle W|$, having energy W. W can be either negative or positive, depending on whether work is used from or stored in the battery system. This means that the following transition is possible

$$\rho_{SC}^{\text{out}} \otimes |0\rangle\langle 0|_W \to \rho_S^{\text{out}} \otimes \rho_C^{\text{in}} \otimes |W\rangle\langle W|_W, \tag{G5}$$

while the thermal state of the battery system is given by

$$\tau_W = \frac{e^{-\beta W}|W\rangle\langle W| + |0\rangle\langle 0|}{1 + e^{-\beta W}}, \tag{G6}$$

with $\beta = \frac{1}{kT}$, k being the Boltzmann constant and T being the temperature of the bath that is in contact with the system and battery. For this transformation to be possible, it is required in Theorem 17 that the following conditions hold for all $\alpha \geq 0$,

$$F_\alpha(\rho_S^{\text{in}} \otimes \rho_C^{\text{in}} \otimes \omega_i \| \rho_{\beta S} \otimes \rho_{\beta C} \otimes \tau_W) \geq F_\alpha(\rho_S^{\text{out}} \otimes \rho_C^{\text{in}} \otimes \omega_f \| \rho_{\beta S} \otimes \rho_{\beta C} \otimes \tau_W). \tag{G7}$$

Since the initial, final, and thermal state of the battery is known and depends only on the parameter W, we can then derive an upper bound for W from (G7) and (G6) as

$$F_\alpha(\rho_S^{\text{in}} \| \rho_{\beta S}) + \frac{kT}{\alpha - 1} \log\left[\frac{1}{1 + e^{-\beta W}}\right]^{1-\alpha} \geq F_\alpha(\rho_S^{\text{out}} \| \rho_{\beta S}) + \frac{kT}{\alpha - 1} \log\left[\frac{e^{-\beta W}}{1 + e^{-\beta W}}\right]^{1-\alpha} \tag{G8}$$

Rearranging yields

$$F_\alpha(\rho_S^{\mathrm{in}}\|\rho_{\beta S}) \geq F_\alpha(\rho_S^{\mathrm{out}}\|\rho_{\beta S}) + W \ , \tag{G9}$$

which yields the following upper bound on W

$$W \leq F_\alpha(\rho_S^{\mathrm{in}}\|\rho_{\beta S}) - F_\alpha(\rho_S^{\mathrm{out}}\|\rho_{\beta S}). \tag{G10}$$

In essence, the possible transitions are governed by Rényi divergences, up to tolerance W, which we choose. Since W has to be smaller than the above bounds for all positive alpha, the maximal amount of work extractable for such a process, going from ρ to ρ' will be given as

$$\mathrm{D}_{\mathrm{work}}(\rho \succ \rho') = \inf_{\alpha>0} \ [\mathrm{F}_\alpha(\rho\|\rho_\beta) - \mathrm{F}_\alpha(\rho'\|\rho_\beta)]. \tag{G11}$$

where we refer to $\mathrm{D}_{\mathrm{work}}(\rho \succ \rho')$ as the *work distance* from ρ to ρ'.

It is interesting to see how the conditions presented in [1] arise as special cases of our conditions, and are hence independent of a catalyst. In [1], the extractable work from a state (by thermalizing it) and the work cost for its formation (starting with a thermal state) via thermal operations have been given by

$$\hat{W}_{\mathrm{ext}}(\rho) = -kT \log \mathsf{tr}(\Pi_\rho \rho_\beta) = kT D_0(\rho\|\rho_\beta) \tag{G12}$$

$$\hat{W}_{\mathrm{cost}}(\rho) = kT \log \min\{\lambda : \rho \leq \lambda\rho_\beta\} = kT D_\infty(\rho\|\rho_\beta), \tag{G13}$$

The subsequent corollary shows that the work distance reduces to these quantities.

Corollary 22. *Consider initial and final states ρ and ρ':*

- *If ρ' is the thermal state, then the maximum extractable work $\hat{W}_{\mathrm{ext}}(\rho) = \mathrm{D}_{\mathrm{work}}(\rho \succ \rho')$.*

- *If ρ is the thermal state, then the minimum work cost $\hat{W}_{\mathrm{cost}}(\rho') = -\mathrm{D}_{\mathrm{work}}(\rho \succ \rho')$.*

Proof. If $\rho' = \rho_\beta$, then $\forall\alpha, \ D_\alpha(\rho'\|\rho_\beta) = 0$, hence

$$\mathrm{D}_{\mathrm{work}}(\rho \succ \rho') = kT \cdot \inf_{\alpha>0} \ D_\alpha(\rho\|\rho_\beta) = \hat{W}_{\mathrm{ext}}(\rho),$$

where the last equality holds due to the fact that the Rényi divergences are non-decreasing in positive α. If $\rho = \rho_\beta$, then similarly

$$-\mathrm{D}_{\mathrm{work}}(\rho \succ \rho') = -kT \cdot \inf_{\alpha>0} \ [-D_\alpha(\rho'\|\rho_\beta)] = kT \cdot \sup_{\alpha>0} \ [D_\alpha(\rho'\|\rho_\beta)] = \hat{W}_{\mathrm{cost}}(\rho').$$

$\square$

Let us now return to the discussion of catalysis, where we demand that the catalyst is returned, such that the work distance for resetting the catalyst to it's original state is small. In the case where inexact catalysis occurs, we are allowed to borrow a catalyst ρ_C^{in} and conduct the transformation $\rho_S^{\mathrm{in}} \otimes \rho_C^{\mathrm{in}} \to \rho_{SC}^{\mathrm{out}}$. If this transformation is allowed via thermal operations, then we know for all $\alpha \geq 0$,

$$F_\alpha(\rho_S^{\mathrm{in}} \otimes \rho_C^{\mathrm{in}}, \rho_{\beta S} \otimes \rho_{\beta C}) \geq F_\alpha(\rho_{SC}^{\mathrm{out}}, \rho_{\beta S} \otimes \rho_{\beta C}). \tag{G14}$$

Consider the restrictions as follows: if $\rho_{SC}^{\mathrm{out}} = \rho_S^{\mathrm{out}} \otimes \rho_C^{\mathrm{out}}$ is of product form(see our previous discussion at the start of Section C on why this is required)[46] , and if the cost of restoring the catalyst has to be small, $-\mathrm{D}(\rho_C^{\mathrm{out}} \succ \rho_C^{\mathrm{in}}) \leq \varepsilon$, namely $\mathrm{D}(\rho_C^{\mathrm{out}} \succ \rho_C^{\mathrm{in}}) \geq -\varepsilon$, then

$$F_\alpha(\rho_S^{\mathrm{in}} \otimes \rho_C^{\mathrm{in}}, \rho_{\beta S} \otimes \rho_{\beta C}) \geq F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta S}) + F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta C}) \tag{G15}$$

$$F_\alpha(\rho_S^{\mathrm{in}}, \rho_{\beta S}) \geq F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta S}) + F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta C}) - F_\alpha(\rho_C^{\mathrm{in}}, \rho_{\beta C}) \tag{G16}$$

$$\geq F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta S}) + \inf_{\alpha>0} \left[F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta C}) - F_\alpha(\rho_C^{\mathrm{in}}, \rho_{\beta C}) \right] \tag{G17}$$

$$\geq F_\alpha(\rho_S^{\mathrm{out}}, \rho_{\beta S}) - \varepsilon, \tag{G18}$$

which tells us that our second laws are recovered.

4. Small error per particle – recovering the free energy

We now consider the regime where we allow a catalyst to be returned with accuracy $\|\rho_C^{\mathrm{in}} - \rho_C^{\mathrm{out}}\|_1 \leq \epsilon/\log(N)$ where N is the dimension of the catalyst. I.e. the error per particle is small. We will see in such a case, we recover the ordinary second law.

a. Trivial Hamiltonians – recovering the Shannon entropy

Let us again first consider the case of a trivial Hamiltonian. In particular, we will see that in the extensive regime only the Shannon entropy matters, and the Rényi entropies are no longer relevant. This shows that if we relax the conditions on how cyclic the process is, by allowing relatively large inaccuracies in the returned catalyst, then we recover the usual second law.

Theorem 23. *Let $\varepsilon \geq 0$ and let $p = \mathrm{spec}(\rho)$ and $q = \mathrm{spec}(\rho')$ be the spectra of the input and output state respectively which are diagonal in the same basis and have dimension d.*
If there exists a catalyst with spectrum $r = r_N$ of dimension N such that

$$p \otimes r \succ s, \qquad \|s - q \otimes r\|_1 \leq \frac{\varepsilon}{\log(N)}, \tag{G19}$$

then

$$H(p) \leq H(q) - \varepsilon - \frac{\varepsilon \log(d)}{\log(N)} - h\left(\frac{\varepsilon}{\log(N)}\right), \tag{G20}$$

with $h(x) := -x\log(x) - (1-x)\log(1-x)$ the binary entropy.
Conversely, if

$$H(p) < H(q), \tag{G21}$$

then for all N sufficiently large there exists a catalyst with spectrum $r = r_N$ of dimension N such that

$$p \otimes r \succ s, \qquad \|s - q \otimes r\|_1 = \exp(-c\sqrt{\log(N)}). \tag{G22}$$

for some constant c.

Proof. Suppose there is a catalyst with spectrum r such that Eq. (G19) holds true. Then by Fannes inequality,

$$|H(s) - H(q \otimes r)| \leq \varepsilon + \frac{\varepsilon \log(d)}{\log(N)} + h\left(\frac{\varepsilon}{\log(N)}\right) \tag{G23}$$

and Eq. (G20) follows from monotonicity of entropy under stochastic maps.
Conversely, suppose Eq. (G21) holds. Then we know that for all n sufficiently large we have

$$p^{\otimes n} \succ q_n \tag{G24}$$

with

$$\|q_n - q^{\otimes n}\|_1 \leq \exp(-c\sqrt{n}). \tag{G25}$$

Let us consider the following catalyst introduced in [37]:

$$\omega = p^{\otimes(n-1)} \oplus p^{\otimes(n-2)} \otimes q \oplus \ldots \oplus p \otimes q^{\otimes(n-2)} \oplus q^{\otimes(n-1)}/n. \tag{G26}$$

We have

$$p \otimes \omega = p^{\otimes n} \oplus p^{\otimes(n-1)} \otimes q \oplus \ldots \oplus p^{\otimes 2} \otimes q^{\otimes(n-2)} \oplus p \otimes q^{\otimes(n-1)}/n. \tag{G27}$$

Then by Eq. (G24),

$$p \otimes \omega \succ s \tag{G28}$$

with

$$s = q_n \oplus p^{\otimes(n-1)} \otimes q \oplus \ldots \oplus p^{\otimes 2} \otimes q^{\otimes(n-2)} \oplus p \otimes q^{\otimes(n-1)}/n. \tag{G29}$$

The result follows from the bound

$$\|s - q \otimes \omega\|_1 = \|q_n - q^{\otimes n}\|_1/n \leq \exp(-c\sqrt{n}), \tag{G30}$$

and the fact that the dimension of ω is $N = n2^{n-1}$. $\qquad\square$

Note that in the above we do not have a condition that the states are diagonal in the energy eigenbasis because all states are diagonal in the energy eigenbasis of the trivial Hamiltonian.

b. General Hamiltonians

Here we prove a result analogous to the one of Section G 4 a in terms of the free energy and for systems with a non-trivial Hamiltonian As above, we find that if we relax the condition on how cyclic the process must be, then we recover the usual second law. Below by $p \xrightarrow{TO} q$ we mean that one can go from p to q by thermal operations.

Theorem 24. *Let $\varepsilon \geq 0$ and let H_S be the Hamiltonian. Let $p = \mathrm{spec}(\rho)$, $q = \mathrm{spec}(\rho')$ be the spectra of the input and output state, respectively, which are diagonal in the energy eigenbasis. If there exists a catalyst with spectrum $r = r_N$ of dimension N (with some Hamiltonian H_C) such that*

$$p \otimes r \xrightarrow{TO} s, \qquad \|s - q \otimes r\|_1 \leq \frac{\varepsilon}{\max\{\log(N), E_{\max}\}}, \tag{G31}$$

where $E_{\max}$ is maximal energy of the Hamiltonian H_S, then

$$F(\rho) \geq F(\rho') - 2\varepsilon - \frac{\varepsilon \log(d)}{\log(N)} - h\left(\frac{\varepsilon}{\log(N)}\right), \tag{G32}$$

where F is the standard free energy $F = E - TS$.
   Conversely, if

$$F(p) > F(q), \tag{G33}$$

then for all N sufficiently large there exists a catalysts with spectrum $r = r_N$ of dimension N such that

$$p \otimes r \xrightarrow{TO} s, \qquad \|s - q \otimes r\|_1 = \exp(-\Omega(\sqrt{\log(N)})). \tag{G34}$$

Proof. Suppose there is a catalyst r such that Eq. (G31) holds true. Then by Fannes inequality,

$$|H(s) - H(q \otimes r)| \leq \varepsilon + \frac{\varepsilon \log(d)}{\log(N)} + h\left(\frac{\varepsilon}{\log(N)}\right). \tag{G35}$$

Also,

$$|E(p) - E(q)| \leq \varepsilon \tag{G36}$$

Therefore $|F(p) - F(q)| \leq 2\varepsilon + \frac{\varepsilon \log(d)}{\log(N)} + h\left(\frac{\varepsilon}{\log(N)}\right)$ which gives (G32).
   Conversely, suppose that (G33) holds. Then from the main result of Ref. [17] we know that for n sufficiently large

$$p^{\otimes n} \xrightarrow{TO} q_n, \tag{G37}$$

with

$$\|q_n - q^{\otimes n}\|_1 \leq \exp(-\Omega(\sqrt{\log(N)})). \tag{G38}$$

We then consider a catalyst with the following Hamiltonian:

$$H_C = \oplus_{k=1}^n \sum_{i=1}^n H_S^{(i)} \tag{G39}$$

where $H_S^{(i)} = \overbrace{\mathbb{I} \ldots \otimes \underbrace{H_S}_{i\text{-th site}} \otimes \mathbb{I} \otimes \ldots \otimes \mathbb{I}}^{n\text{terms}}$ and the state of catalyst of the form (G26). We have

$$p \otimes \omega = \frac{1}{n} p^{\otimes n} \oplus \tilde{\omega}, \quad \omega \otimes q = \tilde{\omega} \oplus \frac{1}{n} q^{\otimes n} \qquad (G40)$$

Now, let us note that

$$\frac{1}{n} p^{\otimes n} \oplus \tilde{\omega} \overset{TO}{\to} \tilde{\omega} \oplus \frac{1}{n} q_n. \qquad (G41)$$

Indeed we first apply the operation that switches the energy levels from the first term of the direct sum with the levels of its last term. Then we apply to the levels of the last term the operation that transforms $p^{\otimes n}$ into q_n, which was shown to exists in Ref. [17], as mentioned above. Then we reverse the above switching operation. Now, the result follows by

$$||\omega \otimes q - \tilde{\omega} \oplus \frac{1}{n} q_n|| \leq \frac{1}{n} ||q^{\otimes n} - q_n||, \qquad (G42)$$

(G38), and the fact that the dimension of the catalyst is $N = n2^{n-1}$. $\qquad\qquad\square$

c. Small average work

In Ref. [38] a paradigm of drawing *average work* was put forward. One starts with system in state ρ_S^{in} and work register in some initial state ρ_W^{in}, and applies thermal operations to get the work register in a new state ρ_W^{out}. The average work is the difference of average energy of the initial and final state of work register. This is because one can show that the protocol of drawing work in Ref. [38] is such that while repeating it with many independent systems onto the same work register, its state becomes highly peaked around some fixed energy without degeneracies.

In [38] it was shown that if ρ and σ satisfy $F(\rho) \geq F(\sigma)$ then one can perform the transition, without spending average work (that is, such transition, may need sometimes a lot of deterministic work, but sometimes one gets the work back). It is reasonable to assume that in the case where a catalyst is used many times over many cyclic processes, then one might not care how much work is required to return the catalyst to it's original state, but rather, one might only care about how much work is required on average. Indeed, if the catalyst is used many times independently, then by the law of large numbers, the averages work with high probability will converge to some deterministic value.

In such scenario, we are allowed to return the catalyst close to the initial catalyst, in the sense that by spending an ϵ amount of average work, one can go back to the original catalyst. According to [38] this means that

$$F(\rho_C^{\text{in}}) \geq F(\rho_C^{\text{out}}) - \epsilon. \qquad (G43)$$

We shall now show, that within such paradigm, if for two states we have $F(\rho_S^{\text{in}}) > F(\rho_S^{\text{out}})$ one can make transition $\rho_S^{\text{in}} \to \rho_S^{\text{out}}$. Indeed, suppose now that $F(\rho_S^{\text{in}}) > F(\rho_S^{\text{out}})$. From the previous section we know that the following transition is possible

$$\rho_S^{\text{in}} \otimes \rho_C^{\text{in}} \to \pi \qquad (G44)$$

where $||\pi - \rho_S^{\text{out}} \otimes \rho_C^{\text{in}}|| \leq \exp(-\Omega(\sqrt{(\log N)})$, with N being dimension of the catalyst. Let us argue, that therefore the free energy of the states π and $\rho_S^{\text{out}} \otimes \rho_C^{\text{in}}$ are close. First, the entropies are close, by Fannes inequality: $|S(\pi) - S(\rho_S^{\text{out}} \otimes \rho_C^{\text{in}})| \lesssim \log N \exp(-\Omega(\sqrt{\log(N)}))$. Let us now estimate the energy difference. Applying the reasoning from section G 4 b we have that diagonal of π is given by $\tilde{\omega} \oplus \frac{1}{n} \sigma_n$ and diagonal of $\rho_S^{\text{out}} \otimes \rho_C^{\text{in}}$ is $\omega \otimes \rho_S^{\text{out}} = \tilde{\omega} \oplus \rho_S^{\text{out} \otimes n}$, where σ_n is a state which can be obtained from $\rho_S^{\text{in} \otimes n}$ by thermal operations (as proved in [17]). The state σ_n satisfies $||\sigma_n - \rho_S^{\text{out} \otimes n}|| \leq \exp(-\Omega(\sqrt{\log(N)}))$ and both σ_n, and $\rho_S^{\text{out} \otimes n}$ have Hamiltonian $\sum_{i=1}^{n} H_S^{(i)}$. Thus we have

$$E(\pi) - E(\rho_S^{\text{out}} \otimes \rho_C^{\text{in}}) = \frac{1}{n} \left(E(\rho_S^{\text{out} \otimes n}) - E(\sigma_n) \right) \leq E_S^{\text{max}} ||\rho_S^{\text{out} \otimes n} - \sigma_n||_{\text{tr}} \leq E_S^{\text{max}} \exp(-\Omega(\sqrt{\log(N)})) \qquad (G45)$$

Where E_S^{max} is maximal energy of a single system S, which is some constant, independent of the size of catalyst.

Now, since the free energies of the real output state π, and the ideal output are close, then one can convert π into $\rho_S^{\text{out}} \otimes \rho_C^{\text{in}}$, by investing an amount of work exponentially small in the catalyst dimension N. Thus, if the customer hands to the bank the state π the bank can regain the original catalyst and return the required output state of the system to the customer.

Note, that we assumed that the in order to return the catalyst to it's original state, one can act on the whole state of the system and ancilla, while more naturally, one should act only on the catalyst. It is an interesting open question whether these two assumptions are equivalent.

Appendix H: Proofs of properties of Rényi entropies and divergences

In addition to collecting and proving useful properties of the Rényi divergences, we show in this section that by allowing error terms which are independent of the dimension of our system, the Rényi divergences needed for our second laws can collapse to just two quantities.

1. Smoothing of Rényi entropies and divergences

Note that the entropies and divergences are monotonic in α (decreasing for Rényi entropies, and increasing for Rényi divergences). The inequality, however, can be reversed if smoothing to a nearby state is allowed. It should be noted that we have explicit methods for smoothing in the regions $\alpha < 1$ and $\alpha > 1$, but otherwise independent of the value of α. Nevertheless, our result give general relations in terms of so-called smoothed entropies and we will hence include them here for completeness.

a. Definitions of smoothing

Besides the exact Rényi entropies, their smoothed versions have also been considered in [39, 40], such that continuity with regard to small changes in probability distribution is preserved, and these quantities have more physical interpretations in terms of operational tasks. Their definitions are as follows:

$$
H_\alpha^\epsilon(p) = \begin{cases} \max_{\tilde{p}} \ H_\alpha(\tilde{p}) & (\alpha < 0); \\ \min_{\tilde{p}} \ H_\alpha(\tilde{p}) & (0 \leq \alpha \leq 1); \\ \max_{\tilde{p}} \ H_\alpha(\tilde{p}) & (\alpha > 1). \end{cases} \tag{H1}
$$

where optimization occurs over sub-normalized states that are ϵ-close to p in terms of trace distance. Mathematically, $\tilde{p} \in \mathcal{B}^\epsilon(p)$, for $\mathcal{B}^\epsilon(p) = \{\tilde{p} : \frac{1}{2} \sum_i |p_i - \tilde{p}_i| \leq \epsilon\}$.

The smoothed Rényi divergences are similarly defined, by smoothing over the ϵ-ball of subnormalized states for the first argument, where maximization/minimization is taken depending on α. Formally,

$$
D_\alpha^\epsilon(p\|q) = \begin{cases} \min_{\tilde{p}} \ D_\alpha(\tilde{p}\|q) & (\alpha < 0); \\ \max_{\tilde{p}} \ D_\alpha(\tilde{p}\|q) & (0 \leq \alpha \leq 1); \\ \min_{\tilde{p}} \ D_\alpha(\tilde{p}\|q) & (\alpha > 1). \end{cases} \tag{H2}
$$

where optimization occurs over sub-normalized states that are ϵ-close to p.

b. Technical lemmas

These quantities will be useful in considering approximate state transformations, where getting into any state close to the target state is sufficient. Also, smoothing allows us to reformulate an infinite families of conditions on the α-Rényi entropies to only two conditions, if states in the ϵ-ball of the original state are allowed. We express this in terms of the following lemma.

Lemma 25. *Given any distribution p, for $0 < \alpha < 1$ and $\epsilon > 0$, there exists a (sub-normalized) distribution $p' \in \mathcal{B}^\epsilon(p)$, such that*

$$
H_0(p) \geq H_\alpha(p) \geq H_0(p') - \frac{\log \frac{1}{\epsilon}}{1 - \alpha}.
$$

For $\alpha > 1$, there exists another smoothed distribution $p'' \in \mathcal{B}^\epsilon(p)$, such that

$$
H_\infty(p'') + \frac{\log \frac{1}{\epsilon}}{\alpha - 1} \geq H_\alpha(p) \geq H_\infty(p).
$$

Proof. To prove this statement, we construct such a smoothed state p' which is within the ϵ-ball of p, and show that the inequality above holds. A construction of p'' can be found in [41] for $\alpha > 1$.

Note that the elements $p_i = 0$ do not contribute in our calculations, furthermore the quantities we calculate are invariant under permutations of i. Hence, without loss of generality we can arrange them such that $p_1 \leq p_2 \leq \cdots \leq p_n$. For any ϵ, denote j as the maximum number such that

1. $\sum_{i=1}^{j} p_i \leq \epsilon$

2. $\sum_{i=1}^{j+1} p_i > \epsilon$.

The smoothed probability distribution p' is then obtained by cutting all probabilities p_i where $i \leq j$. Also note that for $i \leq j$, $p_i \leq p_{j+1}$. Now, we evaluate a lower bound for the following quantity:

$$\sum_{i=1}^{n} p_i^\alpha \geq \sum_{i=1}^{j+1} p_i^\alpha \geq \sum_{i=1}^{j+1} p_i^{\alpha-1} p_i \geq p_{j+1}^{\alpha-1} \sum_{i=1}^{j+1} p_i > p_{j+1}^{\alpha-1} \cdot \epsilon \geq [\text{rank}(p')]^{1-\alpha} \cdot \epsilon.$$

where the third inequality holds because $\alpha - 1 < 0$, and the last inequality holds because p'_{j+1} is the minimum value in the smoothed distribution p', and therefore must be smaller than $\frac{1}{\text{rank}(p')}$. Taking the logarithm and dividing by $1 - \alpha > 0$,

$$H_\alpha(p) = \frac{1}{1-\alpha} \log \sum_{i=1}^{n} p^\alpha(x_i)$$

$$\geq \log \text{rank}(p') - \frac{\log \frac{1}{\epsilon}}{1-\alpha}$$

$$\geq H_0(p') - \frac{\log \frac{1}{\epsilon}}{1-\alpha}.$$

It is worth noting that in extreme cases, where smoothing cannot be performed, the bound becomes trivial. To see this, note that the minimum value $p_1 \leq \frac{1}{n}$, where n is the rank of p. If one cannot smooth at all, then $\epsilon < \frac{1}{n}$. The bound then translates to

$$H_0(p') - \frac{\log \frac{1}{\epsilon}}{1-\alpha} \leq \log n - \frac{1}{1-\alpha} \log n \leq 0. \tag{H3}$$

This means that for values of $\epsilon < \frac{1}{n}$, the bound simply becomes trivially $H_\alpha(p) \geq 0$.

$\square$

From this lemma, combining with the fact that the smoothed entropy $H_0^\epsilon(p)$ is obtained by minimizing over all subnormalized states in the ϵ-ball of p. More precisely, $H_0^\epsilon(p) = \min_{\tilde{p} \in \mathcal{B}^\epsilon(p)} H_0(\tilde{p}) \leq H_0(p')$. Hence, it is easy to obtain a corollary that corresponds to Lemmas 4.2 and 4.3 as stated in [41].

Corollary 26. *Given any distribution p, for $0 < \alpha < 1$ and $\epsilon > 0$,*

$$H_\alpha(p) \geq H_0^\epsilon(p) - \frac{\log \frac{1}{\epsilon}}{1-\alpha}.$$

For $\alpha > 1$,

$$H_\alpha(p) \leq H_\infty^\epsilon(p) + \frac{\log \frac{1}{\epsilon}}{\alpha-1}.$$

From Lemma 25, we know that the Rényi entropies collapse to two smoothed quantities, the $H_0(p)$ and $H_\infty(p)$ for $0 < \alpha < 1$ and $\alpha > 1$ respectively. In the next two lemmas, we show similar results for the smoothing of Rényi divergences. Our proofs will be constructed in a similar way as the above proof, namely, we will specify the smoothed states within some ϵ-ball such that the desired inequality holds, then link this to the smooth min- or max- divergence.

Lemma 27. *Given probability distributions p, q such that $\text{supp}(p) \subseteq \text{supp}(q)$. Then for $\alpha > 1$ and $\epsilon > 0$, there exists a smoothed distribution p'*

$$D_\alpha(p\|q) \geq D_\infty(p'\|q) - \frac{\log \frac{1}{\epsilon}}{\alpha-1}.$$

Proof. Note that we require $\text{supp}(p) \subseteq \text{supp}(q)$ so that $D_\infty(p\|q)$ does not diverge to infinity. Now, let us consider the set $Z_\delta = \{i : \frac{p_i}{q_i} \geq \delta\}$. Then the smoothed probability distribution p' is defined by having $p'_i = \delta \cdot q_i$ for all $i \in Z_\delta$. The statistical distance between p and p' is

$$d = \sum_{i \in Z_\delta} [p_i - p'_i] \leq \sum_{i \in Z_\delta} p_i, \tag{H4}$$

note that d can be made equal to ϵ, by tuning δ in a continuous manner. Then, we have

$$D_\infty(p'\|q) = \log \max_i \frac{p_i}{q_i} = \log \delta. \tag{H5}$$

We now evaluate a lower bound on

$$\sum_i p_i^\alpha q_i^{1-\alpha} \geq \sum_{i \in Z_\delta} p_i^\alpha q_i^{1-\alpha} \geq \sum_{i \in Z_\delta} p_i \cdot \left[\frac{p_i}{q_i}\right]^{\alpha-1} \geq \delta^{\alpha-1} \cdot \sum_{i \in Z_\delta} p_i \geq \delta^{\alpha-1} \cdot \epsilon,$$

where the first inequality holds because of the positivity of p_i and q_i for all i, the third inequality holds since $\alpha - 1 > 0$, and the last inequality holds due to (H4). Then, taking the logarithm, and dividing the whole equation by $\alpha - 1$, we have

$$D_\alpha(p\|q) = \frac{1}{\alpha - 1} \log \sum_x p_i^\alpha q_i^{1-\alpha} \geq \log \delta + \frac{\log \epsilon}{\alpha - 1} = D_\infty(p'\|q) + \frac{\log \epsilon}{\alpha - 1}. \tag{H6}$$

$\square$

Lemma 28. *Given probabilty distributions p and q. Then for $\alpha < 1$ and $\epsilon > 0$, there exists a smoothed distribution p' such that*

$$D_\alpha(p\|q) \leq D_0(p'\|q) + \frac{\log \frac{1}{\epsilon}}{1 - \alpha}. \tag{H7}$$

Proof. Similarly in the proof of Lemma 27, we define a particular smoothing of p, called p'. Note also, that terms $p_{x_i} = 0$ or $q_{x_1} = 0$ are automatically discarded because they do not affect our calculations in any way. Firstly, we order the values $\frac{p_1}{q_1} \leq \frac{p_2}{q_2} \leq \cdots \leq \frac{p_n}{q_n}$. Subsequqently, we find an integer j such that the two conditions below are satisfied:

1. $\sum_{i=1}^{j} p_i \leq \epsilon$,

2. $\sum_{i=1}^{j+1} p_i > \epsilon$.

Note that $D_0(p'\|q) = -\log \sum_{i=j+1}^{n} q_i$. Also, for any $i \geq j+1$, $\frac{p_i}{q_i} \geq \frac{p_{j+1}}{q_{j+1}}$.

With this, we can evaluate the following bound:

$$\sum_{i=j+1}^{n} q_i \leq \frac{q_{j+1}}{p_{j+1}} \cdot \sum_{i=j+1}^{n} p_i \leq \frac{q_{j+1}}{p_{j+1}}.$$

subsequently, we can evaluate a lower bound on the following summation:

$$\sum_{i=1}^{n} p_i^\alpha q_i^{1-\alpha} \geq \sum_{i=1}^{j+1} p_i^\alpha q_i^{1-\alpha}$$

$$\geq \sum_{i=1}^{j+1} p_i \cdot \left[\frac{p_i}{q_i}\right]^{\alpha-1}$$

$$\geq \left[\frac{p_{j+1}}{q_{j+1}}\right]^{\alpha-1} \cdot \sum_{i=1}^{j+1} p_i$$

$$> \left[\frac{p_{j+1}}{q_{j+1}}\right]^{\alpha-1} \cdot \epsilon$$

$$\geq \left[\frac{1}{\sum_{i=j+1}^{n} q_i}\right]^{\alpha-1} \cdot \epsilon,$$

where the second and fifth inequality holds since $\alpha - 1 < 0$, and others by srtaightforward manipulation.

It is useful to see that even for the case where $j = 0$ where no smoothing is possibly done, the proof still holds since now we know that $\epsilon < p_1$, where p_1 is the value that corresponds to the minimum in $\frac{p_i}{q_i}$. Note that in this case,

$$\sum_{i=1}^{n} p_i^{\alpha} q_i^{1-\alpha} \geq \left[\frac{p_1}{q_1}\right]^{\alpha-1} p_1 \geq \epsilon. \tag{H8}$$

This comes from two facts: $p_1 > \epsilon$, and the fact that $\frac{p_1}{q_1} \leq 1$. To see why this is the case, let us prove by contradiction, i.e. assume that $\frac{p_1}{q_1} > 1$ and all other $\frac{p_i}{q_i} > 1$ also. Then for each i, $p_i > q_i$, and $\sum_{i=1}^{n} p_i > \sum_{i=1}^{n} q_i = 1$ which is impossible. $\qquad \square$

With these two lemmas, two simple corollaries with regard to smooth divergences can be obtained:

Corollary 29. *Given distributions p and q. Then for $\alpha > 1$ and $\epsilon > 0$,*

$$D_{\alpha}(p\|q) \geq D_{\infty}^{\epsilon}(p\|q) - \frac{\log \frac{1}{\epsilon}}{\alpha - 1}. \tag{H9}$$

Corollary 30. *Given distributions p and q. Then for $\alpha < 1$ and $\epsilon > 0$,*

$$D_{\alpha}(p\|q) \leq D_{0}^{\epsilon}(p\|q) + \frac{\log \frac{1}{\epsilon}}{1 - \alpha}. \tag{H10}$$

We have also investigated the Rényi entropies for the $\alpha < 0$ regime, and shown that they are monotonically increasing in α. Although they are not required for our work upon elimination of negative α, we also state them here for completeness.

Theorem 31 (Jensen's inequality). *For any convex function f, the following inequality holds:*

$$f\left(\frac{\sum_i a_i y_i}{\sum_i a_i}\right) \leq \frac{\sum_i a_i f(y_i)}{\sum_i a_i}. \tag{H11}$$

The inequality is reversed for concave functions.

Using Theorem 31, we will prove the following lemma about the monotonicity of $\mathrm{H}_{\alpha}(p)$ in the negative α regime.

Lemma 32. *For $\forall \alpha' \leq \alpha < 0$, $\mathrm{H}_{\alpha'}(p) \leq \mathrm{H}_{\alpha}(p)$ for any probability distribution p.*

Proof. For $\alpha < 0$ and probability distribution p, $\mathrm{H}_{\alpha}(p) = \frac{1}{\alpha - 1} \log \sum_i p^{\alpha}$. For convenience of dealing with positive numbers always, we rewrite this expression by defining a variable $\kappa = -\alpha$, and for $\kappa \in (0, \infty)$, define

$$\hat{\mathrm{H}}_{\kappa}(p) = \frac{-1}{1 + \kappa} \log \sum_i x_i^{\kappa} = \mathrm{H}_{\alpha}(p), \tag{H12}$$

where $x_i = \frac{1}{p_i}$ for all i. We then prove that $\frac{\partial \hat{\mathrm{H}}_{\kappa}(p)}{\partial \kappa} \leq 0$.

$$\begin{aligned}
\frac{\partial \hat{\mathrm{H}}_{\kappa}(p)}{\partial \kappa} &= \frac{1}{(1+\kappa)^2} \log \sum_i x_i^{\kappa} - \frac{1}{1+\kappa} \frac{1}{\sum_i x_i^{\kappa}} \sum_i x_i^{\kappa} \log x_i \\
&= \frac{1}{(1+\kappa)^2} \frac{1}{\sum_i x_i^{\kappa}} \left[\sum_i x_i^{\kappa} \log\left(\sum_j x_j^{\kappa}\right) - (1+\kappa) \sum_i x_i^{\kappa} \log x_i\right] \\
&= \frac{1}{(1+\kappa)^2} \frac{1}{\sum_i x_i^{\kappa}} \left[\sum_i x_i^{\kappa} \log\left(\sum_j x_j^{\kappa}\right) - \sum_i x_i^{\kappa} \log x_i^{(1+\kappa)}\right].
\end{aligned} \tag{H13}$$

By defining the function $f(x) = x \log x$ which is convex in $\mathcal{R}^+$, we can apply Theorem 31 by setting $a_i = x_i^{-1} = p_i$ and $y_i = x_i^{1+\kappa}$. This implies that

$$\left[\sum_i x_i^{\kappa} \log\left(\sum_j x_j^{\kappa}\right) - \sum_i x_i^{\kappa} \log x_i^{(1+\kappa)}\right] \leq 0 \tag{H14}$$

and hence (H13) is upper bounded by 0 due to the positivity of the first two terms. $\qquad \square$

2. Smoothing relations

It turns out that our infinite set of conditions can often be verified by checking just two conditions in an approximate sense. This applies to the case where only the conditions for $\alpha \geq 0$ are relevant. However, as we will argue later this is generally sufficient.

Let us first explain how this works by considering only the Rényi entropies, which are the relevant quantities when the Hamiltonian is trivial. Note that if $\alpha \geq \beta$ then for all distributions p we have $\mathrm{H}_\alpha(p) \leq \mathrm{H}_\beta(p)$. The key to approximately reducing the number of conditions is to note that there exists a distribution quite close to p such that up to some error terms the entropies can also be related in the opposite direction. Closeness is thereby measured in terms of the statistical distance and we use $\mathcal{B}^\epsilon(p) = \{p' : \frac{1}{2}\sum_i |p_i - p'_i|\}$ to denote the ϵ ball of (sub-normalized) distributions p' around p. We will also call such a p' a *smoothed* distribution [39, 40].

Specifically, we will show (see Lemma 25) that for any $0 < \alpha < 1$, any distribution p and any $\epsilon > 0$, there exists a smoothed distribution $p' \in \mathcal{B}^\epsilon(p)$ such that

$$\mathrm{H}_0(p) \geq \mathrm{H}_\alpha(p) \geq \mathrm{H}_0(p') - \frac{\log \frac{1}{\epsilon}}{1 - \alpha}. \tag{H15}$$

Similarly, whenever $\alpha > 1$ there exists another distribution $p'' \in \mathcal{B}^\epsilon(p)$ such that

$$\mathrm{H}_\infty(p'') + \frac{\log \frac{1}{\epsilon}}{\alpha - 1} \geq \mathrm{H}_\alpha(p) \geq \mathrm{H}_\infty(p). \tag{H16}$$

This means that whenever we demand that $\mathrm{H}_\alpha(\rho) \leq \mathrm{H}_\alpha(\rho')$ for all values of $\alpha \geq 0$, we can reduce the set of conditions in an approximate sense by relating H_α to H_0 or H_∞. More precisely, given probability distributions p and q and $\epsilon > 0$ we can construct smoothed distributions $p' \in \mathcal{B}^\epsilon(p)$ and $q'' \in \mathcal{B}^\epsilon(q)$ according to explicit smoothing strategies as in Lemma 25. If the following conditions are satisfied

- For $0 < \alpha < 1$, $\mathrm{H}_0(p') - \frac{\log \frac{1}{\epsilon}}{1-\alpha} \geq \mathrm{H}_0(q)$

- For $\alpha > 1$, $\mathrm{H}_\infty(p) \geq \mathrm{H}_\infty(q'') + \frac{\log \frac{1}{\epsilon}}{\alpha-1}$,

then $\forall \alpha > 0$, it holds that $\mathrm{H}_\alpha(p) \geq \mathrm{H}_\alpha(q)$. As we will see these conditions can also be expressed in terms of smoothed entropies [39], however, we would like to emphasize that there are in fact only two smoothing strategies, one for $\alpha \geq 1$ and one for $\alpha < 1$. This means that one could apply these smoothing strategies, and only verify the two conditions stated above. It should also be emphasized that this allows a verification in one direction only. Namely, if we find that the conditions above are satisfied, then we can conclude that also the original conditions are satisfied for all α. However, due to the approximations above the converse does not hold in the sense that the original conditions may be satisfied and yet the fudge terms in the conditions above no longer allow for a verification.

A similar statement can be made for the Rényi divergences, which are relevant for the case of a non-trivial Hamiltonian. Here we want to check whether given initial and final states ρ and ρ' we have that for all $\alpha \geq 0$, $D_\alpha(\rho\|\rho_\beta) \geq D_\alpha(\rho'\|\rho_\beta)$. Again, we have a bound in one direction as the Rényi divergences are monotonically increasing in α, hence $D_\alpha(\rho'\|\rho_\beta) \geq D_\beta(\rho'\|\rho_\beta)$ whenever $\alpha \geq \beta$ [6]. As we show below, one can again obtain a bound in the other direction by considering smoothed distributions. We show (Lemma 27), that for any distribution p there exists a particular smoothed distribution $p' \in \mathcal{B}^\epsilon(p)$ such that $D_\alpha(p\|q) \geq D_\infty(p'\|q) - c$ for all $\alpha > 1$. c is a logarithmic fudge factor that depends on the smoothing parameter. A similar statement holds for $\alpha < 1$, by relating D_α to D_0 (see Lemma 28). Again, the smoothed distributions only depend on ϵ and whether $\alpha > 1$ or $\alpha < 1$.

These relations now again allow us to simplify our conditions in an approximate sense. Consider probability distributions p, q and r where q has full rank $\mathrm{rank}(q) = n$. For $\epsilon > 0$, apply the ϵ-smoothing strategy in proof of Lemma 27 for p to obtain $p' \in \mathcal{B}^\epsilon(p)$. Then if

$$D_\infty(p'\|r) \gtrsim D_\infty(q\|r), \tag{H17}$$

we have $D_\alpha(p\|r) \geq D_\alpha(q\|r)$ for $\alpha > 1$. Similarly, apply the ϵ-smoothing strategy in Lemma 28 for q to obtain $q' \in \mathcal{B}^\epsilon(q)$. Then if the following conditions are satisfied

$$D_0(p\|r) \gtrsim D_0(q'\|r),$$

then $\forall \alpha > 0$, it holds that $D_\alpha(p\|r) \geq D_\alpha(q\|r)$.

Appendix I: Comparison to other models

1.   Thermal operations and time dependent Hamiltonians

As shown in [1], one can incorporate changing Hamiltonians into thermal operations by introducing an ancillary system. Now that we have conditions for state transformations using ancillary systems we are in a position to prove the optimality of this procedure.

In the standard thermodynamics, one usually deals with time dependent Hamiltonian. For example, expanding a gas in a container by drawing a piston (and thereby changing the Hamiltonian, one goes from the thermal state of volume V_1 to the thermal state of volume V_2). This results in obtaining work.

In the paradigm of thermal operations, we have been discussing the case of a fixed Hamiltonian. There is a possibility of changing the Hamiltonian of the system in the following sense: given a system in state ρ with Hamiltonian H, one brings in another system in the thermal state with Hamiltonian H'. Then one can apply thermal operations to this compound system, and finally trace out the initial system, obtaining some output state σ with Hamiltonian H'. In particular, in this way, one can go for free from the thermal state with one Hamiltonian to a thermal state with another Hamiltonian, which may seem to contradict a common thermodynamical paradigm, where changing the Hamiltonian is related to performing work. However, the above "change" of Hamiltonian is not really a change: the Hamiltonian of the universe is fixed, and we simply have turned to another system with the required new Hamiltonian, and removed the system with the old Hamiltonian. Thus in this case, while going from the thermal state (ρ_β, H) to another thermal state (ρ'_β, H') one does not gain any work, nor does one need any work while performing such an operation.

In standard thermodynamics, in the above transition, work is performed equal to the difference of free energy of the initial and final state. The way to reconcile the paradigm of thermal operations with the more common picture of changing the Hamiltonian was given in [1] (see also [17]). Suppose one wants to go from ρ to σ and change Hamiltonian form H to H', however not in the trivial way described above, which does not cost any work, but in a traditional sense. As noted, the total Hamiltonian of the universe is constant, so such a change of Hamiltonian means that we actually use some other system, usually a clock system, such that the total Hamiltonian is

$$\sum_{t=t_i}^{t=t_f} H(t) \otimes |t\rangle\langle t| \tag{I1}$$

where $|t\rangle$ are orthogonal states. Then in more standard thermodynamics, we set the initial state to be $\rho \otimes |t_i\rangle\langle t_i|$, and let such a system (but not the clock) weakly interact with the heat bath. Due to the weakness of interactions, during the system's evolution, the state is approximately in product form $\rho(t) \otimes |t\rangle\langle t|$ Then the final state is $\sigma \otimes |t_f\rangle\langle t_f|$, and we say we have changed state ρ to $\sigma = \rho(t_f)$ and $H(t_i)$ into $H(t_f)$. The work is then equal to $\Delta W = \int_{t_i}^{t_f} \text{tr } H(t)\rho(t)$.

In our present approach, the weak interaction with the heat bath is replaced by a unitary transformation, that commutes with the total Hamiltonian. We require only, that the final state is of product form $\rho(t_f) \otimes |t_f\rangle\langle t_f|$. The state t_f we take orthogonal to t_i. However, now one can greatly simplify it, as for ideal processes, the possibility of a transition does not depend on the intermediate times, and one can use just two values of t: $|t_i\rangle \equiv |0\rangle$ and $|t_f\rangle \equiv |1\rangle$.

Therefore, to mimic the change of Hamiltonian, we consider a fixed Hamiltonian

$$H = H_0 \otimes |0\rangle\langle 0| + H_1 \otimes |1\rangle\langle 1| \tag{I2}$$

where we have an additional system, that plays the role of a switch bit, and its state changes from $|0\rangle$ to $|1\rangle$, so that we have a transition between $\rho \otimes |0\rangle\langle 0|$ and $\sigma \otimes |1\rangle\langle 1|$ and Hamiltonian H_0 to H_1 acting on the system. As shown in [1], if ρ and σ are thermal states then the amount of deterministic work we need to perform is equal precisely to the difference of standard free energies. In this picture, the variety of paths that might lead from H_0 to H_1 is replaced by the variety of thermal operations that may be applied to the initial state, and give the final state $\sigma \otimes |1\rangle\langle 1|$.

As an example, let us observe, that by using the paradigm of thermal operations, for ρ and σ being thermal states, we will obtain the same answer as in the traditional paradigm: the amount of work is equal to the difference of free energies.

We start with $(\rho_\beta^{(0)} \otimes |0\rangle\langle 0| \otimes |E\rangle_W\langle E|, H) \equiv (\rho_{\text{in}}, H)$ and want to end up with $(\rho_\beta^{(1)} \otimes |1\rangle\langle 1| \otimes |E'\rangle_W\langle E'|, H) \equiv (\rho_{\text{out}}, H)$ where $\rho_\beta^{(0)}$ and $\rho_\beta^{(1)}$ are thermal states for Hamiltonians H_0 and H_1, respectively. The total Hamiltonian is of the form

$$H = H_0 \otimes |0\rangle\langle 0| + H_1 \otimes |1\rangle\langle 1| + H_W \tag{I3}$$

where H_W is the Hamiltonian of the work system. One then computes that $kTD_\alpha(\rho_{\text{in}}) = kT\ln(Z_0/Z) + E$ and $kTD_\alpha(\rho_{\text{out}}) = kT\ln(Z_1/Z) + E'$ where Z_0, Z_1 are partition functions for H_0, H_1 in temperature T. Thus the transition is possible, whenever $F(\rho_\beta^{(0)}) - F(\rho_\beta^{(1)}) \geq \Delta W$, where $\Delta_W = E' - E$ is the amount of performed work.

For general states, we obtain, that $(\rho \otimes |0\rangle\langle0|, H)$ can be transformed into $(\sigma \otimes |1\rangle\langle1|, H)$, with H of the form (I2) if and only if the *generalized free energies* satisfy

$$F_\alpha(\rho, H_0) \geq F_\alpha(\sigma, H_1) \tag{I4}$$

where F_α is defined as follows, for any given state and Hamiltonian:

$$F_\alpha(\rho, H) = -kT\ln Z + kTD_\alpha(\rho\|\rho_\beta) \tag{I5}$$

where ρ_β is the thermal state for the Hamiltonian H.

Finally, one can ask, whether the Hamiltonian of the form (I2) is optimal for the transition from (ρ_0, H_0) to (ρ_1, H_1)?

More generally one could do the following. We consider initial state $\rho_0 \otimes \sigma_0$ and $\rho_1 \otimes \sigma_1$, where σ_0, and σ_1 are arbitrary states of ancillas (replacing the switch states $|0\rangle$ and $|1\rangle$). Then we are to choose an arbitrary total Hamiltonian which satisfies the following conditions: for any state from the support of $\rho_0 \otimes \sigma_0$ it acts as H_0, while for any state with support σ_0, and σ_1 it acts as H_1. We also demand that σ_0 and σ_1 have to be related unitarily, otherwise, going from σ_0 and σ_1 can be exploited as a resource(if e.g. σ_0 is pure, while σ_1 is not).

Now one computes that

$$D_\alpha(\rho_0 \otimes \sigma_0, H) = -\ln Z_0/Z + D_\alpha(\rho_0\|\rho_\beta^{(0)}) + D_\alpha(\sigma_0\|I/d)$$

$$D_\alpha(\rho_0 \otimes \sigma_0, H) = -\ln Z_1/Z + D_\alpha(\rho_1\|\rho_\beta^{(1)}) + D_\alpha(\sigma_1\|I/d)$$

Using that $D_\alpha(\sigma_0\|I/d) = D_\alpha(\sigma_1\|I/d)$ we obtain that the condition of monotonicity of D_α is equivalent to (I4). Thus the simple switch mechanism of Hamiltonian (I2) is optimal.

2. Universality of the work bit

Although we have used a specific form of work system to invest/extract work (the work bit), it was claimed to be equivalent to other work systems in [1]. However, as the work system is an ancilla, one needs to check that this universality continues to hold in the context where catalysts are allowed. One wants to show that the derived work distance is general, i.e. whether by considering other proposed forms of battery systems, we arrive at the same quantity. In fact, we will see that the second laws impose a constraint on how one defines work, if one demands reversibility of the work system. We see that this is satisfied when work is defined in terms of raising and lowering the energy of a pure state (or a system with highly peaked energy). This is because transitions between pure states are not effected by catalysis. However, in models where the Hamiltonian is trivial, i.e. $H = 0$, one needs an alternative way to define work. For example, one can define it in terms of the number of maximally mixed states which are erased into pure states, as was done in [34]. In this case, we can again apply our second laws to the initial and final state, including the battery, to derive an upper bound on W.

More generally, let us take the initial state of the work system W to be W_i and the final state to be W_f. Considering the conditions we derived for state transformation on the joint system SW, from ρ to ρ' on system S, $\forall \alpha \geq 0$ (since the probability distributions have zeros, conditions for $\alpha \langle 0$ become redundant), we have

$$F_\alpha(\rho' \otimes W_f\|\rho_{\beta\,SW}) \leq F_\alpha(\rho \otimes W_i\|\rho_{\beta\,SW}) \tag{I6}$$

This implies

$$F_\alpha(W_f\|\rho_{\beta\,W}) - F_\alpha(W_i\|\rho_{\beta\,W}) \leq F_\alpha(\rho\|\rho_{\beta\,S}) - F_\alpha(\rho'\|\rho_{\beta\,S}) \tag{I7}$$

It is reasonable that for any definition of work system and work extraction, we require that one can reversibly go from state W_i to W_f and visa versa. This is equivalent to requiring that

$$\tilde{W} = F_\alpha(W_f\|\rho_{\beta\,W}) - F_\alpha(W_i\|\rho_{\beta\,W}) \ \forall\alpha \tag{I8}$$

This is because if $F_\alpha(W_f\|\rho_{\beta\,W}) - F_\alpha(W_i\|\rho_{\beta\,W})$ were different for two values of α, then it would require a different amount of pure energy to go from the initial state to the final state, than from the final state to the initial. We

can then just define this constant $\tilde{W}$ to be the amount of work W, and thus the amount of work obeys $W = F_\alpha(W_f \| \rho_{\beta_W}) - F_\alpha(W_i \| \rho_{\beta_W})$ for all α.

Now, in the case of close to pure energy states, $F_\alpha(W_f \| \rho_{\beta_W}) - F_\alpha(W_i \| \rho_{\beta_W}) = E_f - E_i$ and so we recover that the work is just the change in energy of the work system, regardless of what type of system it is. For the battery used in [34] this is also the case: denote the work system W as consisting of n qubits, where the Hamiltonian $\hat{H}_W = \mathbb{I}$ is trivial. n can be arbitrarily large. Its initial state is described as $W_i = |0\rangle\langle 0| \otimes 2^{-\lambda_1}\mathbb{I}$. Physically this just implies that the state consists of λ_1 maximally mixed qubits, and the remaining $n - \lambda_1$ qubits are pure. Its final state is similarly defined as $W_f = |0\rangle\langle 0| \otimes 2^{-\lambda_2}\mathbb{I}$. The thermal state of system W is $\rho_{\beta_W} = 2^{-n}\mathbb{I}$, which is maximally mixed.

What is interesting about this comparison is that while in our model, work is stored in the form of energy, in [13, 34] the work is quantified by means of purity, i.e. how many pure qubits we invest/create during the process of state transformation. The significance of information to work extraction has been discussed by various works [13, 42, 43]. In particular, Landauer's principle [15, 44] states that any physical process that erases one bit of information (i.e., creating purity) in an environment of temperature T has a fundamental average work cost of $kT \ln(2)$. Similarly, by ulitizing one bit of information stored (i.e., consuming purity) in a physical system, and allowing it to interact with a thermal bath at temperature T, one can draw an average work of $kT \ln(2)$.

We will now use this battery system W. From Equation (I7) we can derive an upper bound for the quantity $\lambda = \lambda_1 + \lambda_2$

$$kT(\lambda_1 - \lambda_2) \leq F_\alpha(\rho | \rho_{\beta_S}) - F_\alpha(\rho' \| \rho_{\beta_S}) \tag{I9}$$

Since the work is defined via the process of Landauer erasure as $kT(\lambda_1 - \lambda_2)$, we see that this bound is equivalent to the bounds in (G8). The quantity $\lambda = \lambda_1 - \lambda_2$ denotes the *net gain of pure qubits* in the process. By Landauer's principle λ gives a bound on the maximum amount of work extractable, as no more than $kT \ln(2)$ amount of work can be extracted given one bit of pure information. We thus see that our condition yields the same upper bound on W

$$W \leq \inf_{\alpha > 0}[F_\alpha(\rho \| \rho_{\beta_S}) - F_\alpha(\rho' \| \rho_{\beta_S})]$$

There are, however, a few more subtle differences between these two models for work systems. For instance, note that this battery consists of qubits, and hence λ_1 and λ_2 take integer values. [34] has shown that they can be further generalized to take values of rational numbers, however, one has to postulate the connection between work and information. In our wit model, however, W can take any value, including irrational values.

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[45] Finally, we note that when considering how cyclic a process is, we might also ask about preserving correlations between the catalyst ρ_C^{in} and its purifying systems. Clearly, there exist simple operations that preserve the catalyst even exactly, and yet do not preserve correlations with separated reference systems holding the purification. Consider for example a catalyst of the form $\rho_C^{\text{in}} = \rho_1 \otimes \rho_2$ where $\rho_1 = \rho_2$ and the purifications of ρ_1 and ρ_2 are held separately by systems 1 and 2 respectively. Returning the catalyst as $\rho_2 \otimes \rho_1$ preserves the catalyst, but it requires interaction between the two purifying systems of ρ_1 and ρ_2 in order to restore the correlations with the catalyst. Such a paradigm is more restrictive than the one considered here.

[46] For non-product output distributions, this argument does not hold since the α free energies are not superadditive, namely $F_\alpha(\rho_{AB}, \sigma_{AB}) \geq F_\alpha(\rho_A, \sigma_A) + F_\alpha(\rho_B, \sigma_B)$ is generally not true.