Journal of Mathematical Physics

RESEARCH ARTICLE | NOVEMBER 04 2022

Which bath Hamiltonians matter for thermal operations?

Frederik vom Ende

J. Math. Phys. 63, 112202 (2022)
https://doi.org/10.1063/5.0117534

Journal of Mathematical Physics ARTICLE scitation.org/journal/jmp

Which bath Hamiltonians matter for thermal operations?

Cite as: J. Math. Phys. 63, 112202 (2022); doi: 10.1063/5.0117534
Submitted: 1 August 2022 • Accepted: 14 October 2022 •
Published Online: 4 November 2022

View Online Export Citation CrossMark

Frederik vom Ende[a)]

AFFILIATIONS

Department of Chemistry, Technische Universität München, Lichtenbergstraße 4, 85737 Garching, Germany
and Munich Centre for Quantum Science and Technology (MCQST) and Munich Quantum Valley (MQV), Schellingstraße 4, 80799 München, Germany

[a)]Author to whom correspondence should be addressed: frederik.vom-ende@tum.de

ABSTRACT

In this article, we explore the set of thermal operations from a mathematical and topological point of view. First, we introduce the concept of Hamiltonians with a resonant spectrum with respect to some reference Hamiltonian, followed by proving that when defining thermal operations, it suffices to only consider bath Hamiltonians, which satisfy this resonance property. Next, we investigate the continuity of the set of thermal operations in certain parameters, such as energies of the system and temperature of the bath. We will see that the set of thermal operations changes discontinuously with respect to the Hausdorff metric at any Hamiltonian, which has the so-called degenerate Bohr spectrum, regardless of the temperature. Finally, we find a semigroup representation of (enhanced) thermal operations in two dimensions by characterizing any such operation via three real parameters, thus allowing for a visualization of this set. Using this, in the qubit case, we show commutativity of (enhanced) thermal operations and convexity of thermal operations without the closure. The latter is done by specifying the elements of this set exactly.

I. INTRODUCTION

Over the last decade, sparked by Brandão *et al.*[1] and Horodecki and Oppenheim,[2] as well as Renes[3]—and further pursued by others[4–9]—thermo-majorization and, in particular, its resource theory approach have been widely discussed and researched topics in quantum physics. Here, the central question is given a fixed background temperature and initial and target states of a quantum system, can the former be mapped to the latter by means of a thermal operation? These channels are the fundamental building block of the resource theory approach to quantum thermodynamics as they, roughly speaking, are the operations that are assumed to be performable in an arbitrary number without any cost; for a precise definition, cf. Sec. II. Thus, arguably, studying and understanding the thermal operations, their structure, and their properties are of crucial importance.

The concept of thermal operations is an attempt to formalize which operations can be carried out at no cost (with respect to some resource, e.g., work). Recall that in macroscopic systems, a state transformation is thermodynamically possible if and only if the free energy decreases. In the quantum realm—using the currently accepted definition of thermal operations—this is at least necessary: the non-equilibrium system free energy[10] $F = \operatorname{tr}(H(\cdot)) - k_B TS$ cannot increase under any thermal operation. Here, H is the system's Hamiltonian, S is the von Neumann entropy, T is the temperature of the environment, and k_B is the Boltzmann constant. This property of not increasing actually holds for both the free energy of the classical (diagonal) part and the so-called asymmetry (relative entropy of the coherences); together, these add up to the free energy.[11] However, the decrease of the free energy is not sufficient to guarantee state conversion via thermal operations (Example 6 in Ref. 12). This changes once one relaxes the set of operations to those that leave not the energy, but the *average* energy of the system plus bath invariant as these are precisely the channels that decrease the free energy.[13] For the interconversion of classical states, considering not only the free energy but also a collection of generalized free energies leads to a characterization of this problem when allowing for *catalytic* thermal operations (Theorem 18 in Ref. 1). For a comprehensive introduction to this topic, we refer to the review article by Lostaglio.[12]

These conditions imposed by the generalized free energies have been called "the second laws of quantum thermodynamics" in the past. On a related note, there is also a third law of quantum thermodynamics, at least for qubits. Scharlau and Mueller gave a lower bound on the population of the lowest energy level when applying any thermal operation (Theorem 9 in Ref. 14). In particular, this implies that no non-ground state can be mapped exactly to the ground state by means of thermal operations with finite heat baths. This is a refinement of the related result that no state with a trivial kernel (i.e., 0 is not an eigenvalue of the state) can be mapped to the ground state—or any pure state for that matter—by means of a Gibbs-preserving channel (Corollary 4.7 in Ref. 15).

The problem of characterizing state conversions as mentioned in the beginning is fully solved in the classical regime. This has to do with the observation made early on that thermal operations and general Gibbs-preserving quantum maps are (approximately) indistinguishable on quasi-classical states. Indeed, given a system described by $\mathrm{diag}(E_1,\ldots,E_n)$ with background temperature $T\in(0,\infty]$, transforming $\mathrm{diag}(y)$ into $\mathrm{diag}(x)$ via thermal operations is possible if and only if $\|x-\frac{y_i}{d_i}d\|_1\le\|y-\frac{y_i}{d_i}d\|_1$ holds for all $i=1,\ldots,n$, where $d:=(e^{-E_j/T})_{j=1}^n$ is the vector of Gibbs weights.[16] Equivalently, the so-called "thermo-majorization curve" (a piecewise linear bijection on the interval [0, 1]) corresponding to y must not lie below the curve corresponding to x anywhere.[2] This reduces the classical state conversion problem to a finite list of conditions, that is, n simple 1-norm inequalities or, using thermo-majorization curves, to $n-1$ inequalities each involving a minimum over a set of n elements (Theorem 4 in Ref. 17). For more details and further characterizations, we refer to Proposition 1 in Ref. 16.

Be aware that it was also noticed early on that the thermal operations form a strict subset of the Gibbs-preserving maps as soon as coherences come into play.[4] This is one of the reasons why the state conversion problem becomes much more complicated in the quantum case: While there exists a characterization via infinitely many inequalities involving the conditional min-entropy,[18] a simple characterization—such as in the classical case—beyond qubits is still amiss; refer also to Sec. 4.2 in Ref. 15. There have been different ways to deal with this problem in the past: While some authors constrained the set of thermal operations to simpler subsets, e.g., such which are experimentally implementable using the current technology,[6,19] others[14,20,21] focused on learning more about the role of the bath Hamiltonian in the action of thermal operations. In this article, we follow the second line of thought.

This work is organized as follows. In Sec. II, we introduce the concept of bath Hamiltonians having a "resonant spectrum" with respect to a given system, and we show that these are everything one needs to generate (approximate) all thermal operations (Proposition 4). As a special case, we recover and refine a result about the structure of thermal operations if the system in question is a spin system, that is, if the Hamiltonian has equidistant eigenvalues (Corollary 5 and 6). These corollaries suggest that the set of thermal operations may in some sense change discontinuously at certain Hamiltonians; this we investigate in Sec. III. There, we look at two particular systems where this discontinuity manifests (Example 7). These examples can be generalized to arbitrary dimensions and Hamiltonians with certain properties, thus revealing a structural problem rather than being singled-out counter-examples. Finally, in Sec. IV, we visualize the set of qubit thermal operations as a three-dimensional shape (Fig. 2). Using this and our results regarding baths with a resonant spectrum, we give a full answer to what elements the qubit thermal operations consist of and what role degenerate bath Hamiltonians play (Theorem 10).

II. THERMAL OPERATIONS: THE BASICS

We start by reviewing how thermal operations are defined and what basic properties they have. Consider an n-level system described by some $H_S\in\mathbb{C}^{n\times n}$ Hermitian ("system's Hamiltonian") and some $T>0$ ("fixed background temperature"). Given any $m\in\mathbb{N}$, we define

$$\begin{aligned}\Phi_{T,m}: i\mathfrak{u}(m)\times \mathsf{U}(mn)&\to \mathrm{CPTP}(n)\\ (H_B,U)&\mapsto \mathrm{tr}_B\bigg(U\bigg((\cdot)\otimes\frac{e^{-H_B/T}}{\mathrm{tr}(e^{-H_B/T})}\bigg)U^*\bigg),\end{aligned}$$

where $\mathsf{U}(m)$ is the unitary group in m dimensions, $\mathfrak{u}(m)$ is its Lie algebra [so $i\mathfrak{u}(m)$ is the collection of all Hermitian $m\times m$ matrices], and $\mathrm{CPTP}(n)$ is the set of all completely positive, trace preserving, linear maps on $\mathbb{C}^{n\times n}$. Thus, $\Phi_{T,m}(H_B,U)$ represents first coupling the system described by H_S to an m-dimensional bath described by H_B at temperature T, then applying the unitary channel $\mathrm{Ad}_U=U(\cdot)U^*$ to the full system, and finally discarding the bath. Using this notation, following Lostaglio,[12] we define the thermal operations with respect to H_S, T as

$$\mathsf{TO}(H_S,T):=\bigcup_{m\in\mathbb{N}}\Big\{\Phi_{T,m}(H_B,U):\ {\scriptstyle H_B\in i\mathfrak{u}(m),U\in\mathsf{U}(mn) \atop U(H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B)U^*=H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B}\Big\} \tag{1}$$

$$=\bigcup_{m\in\mathbb{N}}\Big\{\Phi_{T,m}(H_B,e^{iH_{\mathtt{tot}}}):\ {\scriptstyle H_B\in i\mathfrak{u}(m),H_{\mathtt{tot}}\in i\mathfrak{u}(mn) \atop [H_{\mathtt{tot}},H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B]=0}\Big\} \tag{2}$$

$$=\bigcup_{m\in\mathbb{N}}\Big\{\Phi_{T,m}(H_B,e^{iH_{\mathtt{tot}}}):\ {\scriptstyle H_B=\mathrm{diag}(E_1,\ldots,E_m)\text{ with }E_1\le\ldots\le E_m \atop H_{\mathtt{tot}}\in i\mathfrak{u}(mn),[H_{\mathtt{tot}},H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B]=0}\Big\}. \tag{3}$$

Physically, $H_{\mathtt{tot}}$ includes the system–bath interaction H_{SB}, that is, $H_{\mathrm{tot}}=H_S\otimes 1_B+1_S\otimes H_B+H_{SB}$, where the commutator condition then reduces to $[H_{SB},H_S\otimes 1_B+1_S\otimes H_B]=0$; cf. also Ref. 22.

Now, to see that sets (1) and (2) are equal, note that the subgroup of $\mathsf{U}(n)$, which stabilizes $H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B$, is compact and connected [because $\mathsf{U}(n)$ is compact and the stabilized element is Hermitian]. Therefore, $\exp$ maps onto this subgroup and we can replace the stabilizing condition on U by the equivalent condition on the level of generators on $H_{\mathtt{tot}}$.

For the equality of (2) and (3) in the definition of $\mathsf{TO}(H_S, T)$ (henceforth, TO for short), i.e., the fact that there is some unitary degree of freedom on the ancilla despite energy-conservation, note $\Phi_{T,m}(H_B, U) = \Phi_{T,m}(\mathrm{Ad}_V(H_B), \mathrm{Ad}_{\mathbb{1}\otimes V}(U))$ for all $H_B \in i\mathfrak{u}(m)$, $U \in \mathsf{U}(mn)$, and $V \in \mathsf{U}(m)$; this follows from the partial trace identity $\mathrm{tr}_2((A \otimes B)C(D \otimes B^{-1})) = A\, \mathrm{tr}_2(C)D$. Moreover, $\mathrm{Ad}_{\mathbb{1}\otimes V}(U)$ is energy-conserving with respect to $(H_S, \mathrm{Ad}_V(H_B))$, so, in particular, we can choose V such that it diagonalizes H_B. What this implies is that the only relevant information coming from the bath is the spectrum of the associated Hamiltonian together with its degeneracies.

Remark 1 (thermal operations in the high temperature limit). Observing that the Gibbs state of any finite-dimensional system becomes the maximally mixed state in the limit $T \to \infty$, one can extend the definition of thermal operations to $T = \infty$ via

$$\mathsf{TO}(H_S, \infty) := \bigcup_{m\in\mathbb{N}} \Big\{ \Phi_{1,m}(\mathbb{1}_m, U) : \begin{smallmatrix} H_B \in i\mathfrak{u}(m), U \in \mathsf{U}(mn) \\ U(H_S\otimes\mathbb{1}_B + \mathbb{1}\otimes H_B)U^* = H_S\otimes\mathbb{1}_B + \mathbb{1}\otimes H_B \end{smallmatrix} \Big\}$$

$$= \bigcup_{m\in\mathbb{N}} \Big\{ \Phi_{1,m}(\mathbb{1}_m, e^{iH_{\mathtt{tot}}}) : \begin{smallmatrix} H_B = \mathrm{diag}(E_1,\ldots,E_m) \text{ with } E_1 \le \cdots \le E_m \\ H_{\mathtt{tot}} \in i\mathfrak{u}(mn), [H_{\mathtt{tot}}, H_S\otimes\mathbb{1}_B + \mathbb{1}\otimes H_B] = 0 \end{smallmatrix} \Big\}.$$

While it might seem that the bath Hamiltonian is redundant as it does not appear in the argument of $\Phi_{1,m}$, waiving it from the energy conservation condition (i.e., setting $H_B = 0$) would reduce the set to only dephasing thermalizations [sometimes called "Hadamard channels" because they are of the Hadamard product form $\rho \mapsto P * \rho := (p_{ij}\rho_{ij})_{i,j=1}^n$, for some positive semi-definite $P \in \mathbb{C}^{n\times n}$, which has only ones on the diagonal; cf. Chap. 1.2 in Ref. 23]. In particular, this would disallow any non-trivial action on diagonal matrices, which is, however, certainly possible within TO.

Having explained how TO is defined for infinite temperatures, let us illustrate how for all $0 < T \le \infty$ the set of thermal operations changes under some elementary transformations of the system's Hamiltonian.

Lemma 2. Given $H_S \in i\mathfrak{u}(n)$, $T \in (0, \infty]$, and $U \in \mathsf{U}(n)$, the following statements hold:

(i) $\mathsf{TO}(\lambda H_S + \mu\mathbb{1}, \lambda T) = \mathsf{TO}(H_S, T)$ *for all* $\lambda > 0, \mu \in \mathbb{R}$.
(ii) $\mathsf{TO}(\mathrm{Ad}_U(H_S), T) = \mathrm{Ad}_U \circ \mathsf{TO}(H_S, T) \circ \mathrm{Ad}_{U^*}$.

These statements continue to hold when replacing TO by its closure.

This is straightforward to show, so we omit the proof.

Now, let us have a closer look at the condition $[H_{\mathtt{tot}}, H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B] = 0$ from Eq. (2), which encodes energy-conservation: this imposes the block-diagonal structure on $H_{\mathtt{tot}}$ in some eigenbasis of $H_S \otimes H_B$, and the sizes of the blocks correspond to how degenerate the eigenvalues of $H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B$ are. Letting $\sigma(\cdot)$, henceforth, denote the spectrum of any matrix: because $\sigma(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B) = \sigma(H_S) + \sigma(H_B)$, this means that $H_{\mathtt{tot}}$ acts non-trivially on an energy level E of the full system only if E can be decomposed into a sum of elements from $\sigma(H_S)$ and $\sigma(H_B)$ in more than one way, i.e., $E = E_i + E_l' = E_j + E_k'$ for some pairwise different $E_i, E_j \in \sigma(H_S)$, $E_k', E_l' \in \sigma(H_B)$. However, this is equivalent to $E_i - E_j = E_k' - E_l'$, which is the necessary condition for the diagonal entries ρ_{ii}, ρ_{jj} of the state of the system ρ to mix by means of a thermal operation (cf. Remark 3 for details). Because the spectrum of $\mathrm{ad}_{H_S} := [H_S, \cdot\,]$ is given by $\{E_i - E_j : i, j\}$, this motivates the following definition: Given $m, n \in \mathbb{N}$, $H_S \in i\mathfrak{u}(n)$, $H_B \in i\mathfrak{u}(m)$ define an undirected graph with vertices being the eigenenergies of H_B, and two vertices are connected if the difference of the corresponding energies appears in the spectrum of ad_{H_S}. We say that H_B has a *resonant* or *absorbing spectrum with respect to H_S* if this graph is connected.[24] To illustrate this definition, we refer to the examples shown in Fig. 1.

Remark 3. A word of warning: The definition of a Hamiltonian having a resonant spectrum is similar to—but should not be confused with—one of the assumptions on heat baths from the early works on thermal operations.[2] There, it was assumed that for any two energies E_i, E_j of the system and any energy E_k' of the bath, there exists some energy E_l' of the bath such that $E_i - E_j = E_k' - E_l'$. However, no finite size heat bath can satisfy this; violations of this condition appear always at the edge and, in some cases, even in the bulk of the energy band. These problems are discussed comprehensively yet in detail in Appendix A in Ref. 25.

This condition is related to a necessary criterion for "interaction" between different entries of a quantum state: Given any Hamiltonian $H_S = \sum_{j=1}^n E_j |g_j\rangle\langle g_j|$, which describes a system currently in the state ρ—represented for now in the eigenbasis of H_S, i.e., $(\rho_{jk})_{j,k=1}^n$ with $\rho_{jk} := \langle g_j, \rho g_k\rangle$—a thermal operation $\Phi_{T,m}(H_B, U)$ can mix ρ_{ij} and ρ_{kl} only if[26] $E_i - E_k = E_j - E_l \in \sigma(\mathrm{ad}_{H_B})$. Equivalently, it is necessary that the transitions corresponding to ρ_{ij} and ρ_{kl} coincide (that is, $E_i - E_j = E_k - E_l$) and that the difference between these transitions appears as a difference in H_B [i.e., $E_i - E_k \in \sigma(\mathrm{ad}_{H_B})$]. Be aware that simply scaling entries ρ_{ij} using TO is independent of either of these notions.

Now, the concept of resonance allows us to restrict the set of bath Hamiltonians necessary for describing the set of thermal operations. This and some fundamental topological properties of TO are summarized in the following.

Proposition 4. Let $H_S \in i\mathfrak{u}(n)$ and $T \in (0, \infty]$ be given, and let $\overline{(\cdot)}$, henceforth, denote the closure. The following statements hold:

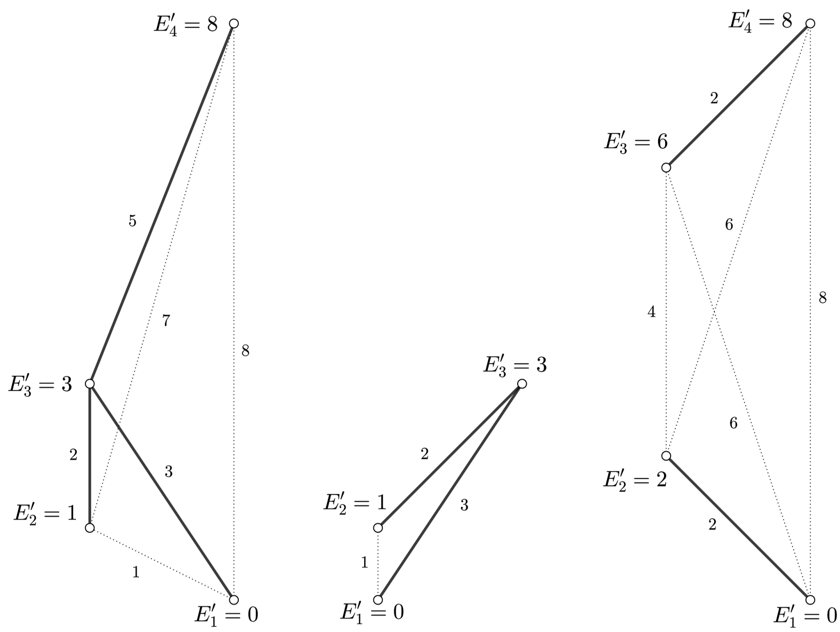

FIG. 1. Let us investigate whether the following bath Hamiltonians have a resonant spectrum with respect to $H_S = \text{diag}(0,2,5)$, that is, $|\sigma(\text{ad}_{H_S})| = \{0,2,3,5\}$. Left: $H_{B,1} = \text{diag}(0,1,3,8)$ has a resonant spectrum with respect to H_S because its graph is connected. Middle: $H_{B,2} = \text{diag}(0,1,3)$ also has a resonant spectrum with respect to H_S for the same reason. Right: $H_{B,3} = \text{diag}(0,2,6,8)$ does not have a resonant spectrum with respect to H_S as it decomposes into the connected components $\{0,2\}$, $\{6,8\}$. These do not "interact" with each other because none of the energy differences between them is in $\sigma(\text{ad}_{H_S})$.

(i) $\mathsf{TO}(H_S,T)$ *is a bounded, path-connected semigroup with identity.*
(ii) $\overline{\mathsf{TO}(H_S,T)}$ *is a convex, compact semigroup with identity.*
(iii) $\overline{\mathsf{TO}(H_S,T)}$ *is a subset of all* CPTP *maps with common fixed point* $e^{-H_S/T}$.
(iv) *For describing the closure of all thermal operations, it suffices to only consider bath Hamiltonians with a resonant spectrum with respect to* H_S*, that is,*

$$\overline{\mathsf{TO}(H_S,T)} = \overline{\bigcup_{m\in\mathbb{N}}\left\{\Phi_{T,m}(H_B, e^{iH_{\texttt{tot}}}) : \begin{array}{c} H_B=\text{diag}(E_1,\ldots,E_m) \text{ with } E_1\le\cdots\le E_m, \\ H_B \text{ has resonant spectrum with respect to } H_S \\ H_{\texttt{tot}}\in i\mathfrak{u}(mn), [H_{\texttt{tot}}, H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B]=0 \end{array}\right\}}. \tag{4}$$

The only non-trivial statements in this lemma are convexity in (ii) as first shown in Appendix C in Ref. 27 and (iv) [respectively, Eq. (4)]. The intuition for the latter is as follows: Given some bath Hamiltonian with a non-resonant spectrum (with respect to H_S), one can partition the said spectrum into different components, which cannot interact with each other because of energy-conservation. This implies that the full unitary is of a similar block structure and that the associated thermal operation can be written as a convex combination of thermal operations generated by bath Hamiltonians with resonant spectra (i.e., the connected components). The full proof is given in Appendix A.

Working with $\overline{\mathsf{TO}}$ instead of TO is advantageous for two reasons: On the one hand, it is unknown whether TO itself is convex (we will answer this in the affirmative for qubits later on). Indeed, a necessary step in showing that statement (iv) from Proposition 4 holds without the closure, i.e., the somehow "intuitive" result that bath Hamiltonians with a non-resonant spectrum are not needed for describing TO, would be a proof of convexity of TO, which continues to hold when considering the right-hand side of (4) (without the closure).

On the other hand, more gravely, TO is not closed. The simplest counter-example corresponds to transforming an energy eigenstate; this is not thermally allowed,[11] meaning the map

$$\begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \mapsto \begin{pmatrix} (1-e^{-1/T})a_{11} + a_{22} & 0 \\ 0 & e^{-1/T}a_{11} \end{pmatrix}$$

is not in $\mathsf{TO}(-\sigma_z,T)$. Yet, this map can be approximated arbitrarily well by thermal operations, so it is an element of $\overline{\mathsf{TO}(-\sigma_z,T)}$; cf. Sec. IV. One way to fix this is to use baths of infinite size, e.g., single-mode bosonic baths (cf. Lemma 1 in Ref. 6 and Ref. 20). However, while such baths are able to implement the above operation, using them to implement full dephasing (even approximately) becomes impossible once the temperature is too low; cf. Theorem 10 (iv).

Either way, the closure guarantees a "reasonable mathematical structure." This can also be motivated from an application or engineering point of view: At least for some questions (e.g., reachability in control theory), it does not matter whether one can implement an operation exactly or "only" with arbitrary precision. However, figuring out which results continue to hold after waiving the closures could reveal more of the structure of the thermal operations (cf. also Sec. V).

An important consequence of Proposition 4 (iv) is that if H_S is a spin-Hamiltonian, i.e., H_S has equidistant eigenvalues, then one can reduce the set of bath Hamiltonians used in the definition of TO to spin-Hamiltonians "of the same structure" without changing the set (after taking the closure). This continues to hold even if H_S only is of spin-form "up to potential gaps." The precise statement—a weaker version of which first appeared in Lemma 1 of Ref. 21— reads as follows.

Corollary 5. Given $H_S \in i\mathfrak{u}(n)$, assume that there exist $E_1 \in \mathbb{R}$ and an energy gap $\Delta E > 0$ such that $\sigma(H_S) \subseteq \{E_1 + j\Delta E : j \in \mathbb{N}_0\}$. Define $\mathsf{TO}^{\Delta E}_{\mathsf{Spin}}(H_S, T)$ as the collection of all thermal operations, where H_B is any spin-Hamiltonian with the same gap ΔE as H_S, that is,

$$\mathsf{TO}^{\Delta E}_{\mathsf{Spin}}(H_S,T) := \bigcup_{m\in\mathbb{N}} \left\{ \Phi_{T,\sum_{j=1}^m \beta_j}(H_B, e^{iH_{\mathtt{tot}}}) : \substack{H_B = \oplus_{j=1}^m j\Delta E\, \mathbb{1}_{\beta_j} \text{ with } \beta_1,\ldots,\beta_m \in \mathbb{N} \\ H_{\mathtt{tot}} \in i\mathfrak{u}(n\sum_j \beta_j), [H_{\mathtt{tot}}, H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B]=0} \right\}$$

for all $T > 0$ and

$$\mathsf{TO}^{\Delta E}_{\mathsf{Spin}}(H_S,\infty) := \bigcup_{m\in\mathbb{N}} \left\{ \Phi_{1,\sum_{j=1}^m \beta_j}(\mathbb{1}, e^{iH_{\mathtt{tot}}}) : \substack{H_B = \oplus_{j=1}^m j\Delta E\, \mathbb{1}_{\beta_j} \text{ with } \beta_1,\ldots,\beta_m \in \mathbb{N} \\ H_{\mathtt{tot}} \in i\mathfrak{u}(n\sum_j \beta_j), [H_{\mathtt{tot}}, H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B]=0} \right\}.$$

One finds

$$\begin{aligned} \forall_{T>0} \quad \overline{\mathsf{TO}^{\Delta E}_{\mathsf{Spin}}(H_S,T)} &= \overline{\bigcup_{m\in\mathbb{N}} \left\{ \Phi_{T,\sum_{j=1}^m \beta_j}(H_B, e^{iH_{\mathtt{tot}}}) : \substack{H_B = \oplus_{j=1}^m j\Delta E\, \mathbb{1}_{\beta_j} \text{ with } \beta_1,\ldots,\beta_m \in \mathbb{N}_0, \sum_{i=1}^m \beta_i > 0 \\ H_{\mathtt{tot}} \in i\mathfrak{u}(n\sum_j \beta_j), [H_{\mathtt{tot}}, H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B]=0} \right\}}, \\ \overline{\mathsf{TO}^{\Delta E}_{\mathsf{Spin}}(H_S,\infty)} &= \overline{\bigcup_{m\in\mathbb{N}} \left\{ \Phi_{1,\sum_{j=1}^m \beta_j}(\mathbb{1}, e^{iH_{\mathtt{tot}}}) : \substack{H_B = \oplus_{j=1}^m j\Delta E\, \mathbb{1}_{\beta_j} \text{ with } \beta_1,\ldots,\beta_m \in \mathbb{N}_0, \sum_{i=1}^m \beta_i > 0 \\ H_{\mathtt{tot}} \in i\mathfrak{u}(n\sum_j \beta_j), [H_{\mathtt{tot}}, H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B]=0} \right\}}, \end{aligned} \tag{5}$$

and $\overline{\mathsf{TO}(H_S,T)} = \overline{\mathsf{TO}^{\Delta E}_{\mathsf{Spin}}(H_S,T)}$ for all $T \in (0,\infty]$.

While this result—for the most part—is a corollary of Proposition 4, we, nevertheless, present a proof in Appendix B. This immediately yields the following.

Corollary 6. Given $T \in (0,\infty]$, if $H_S \in i\mathfrak{u}(n)$ has a rational Bohr spectrum up to a global constant—i.e., there exists real $r > 0$ such that $\sigma(\mathrm{ad}_{H_S}) \in r\mathbb{Z}$—then $\overline{\mathsf{TO}(H_S,T)} = \overline{\mathsf{TO}^{r}_{\mathsf{Spin}}(H_S,T)}$ from (5).

This is a direct application of Corollary 5 because if H_S (with eigenvalues $E_1 \le \cdots \le E_n$) has a rational Bohr spectrum, then

$$\sigma(H_S) = \left\{ E_1 + r\cdot \frac{E_j - E_1}{r} : j = 1,\ldots,n \right\} \subseteq \{E_1 + rj : j \in \mathbb{N}_0\}.$$

There does not seem to be an obvious generalization of the previous two results to arbitrary Hamiltonians. For this, consider $H_S = \mathrm{diag}(0,1,\sqrt{2})$ as a system and $H_B = \mathrm{diag}(0,\sqrt{2}-1,1)$ as a bath Hamiltonian; then, H_B does not have a rational Bohr spectrum up to any constant, yet H_B has a resonant spectrum with respect to H_S, so there is no "obvious" decomposition as in Proposition 4/Corollary 5 into baths $H_{B,1}, H_{B,2}$ of the spin form.

However, one may ask whether the (somewhat unphysical) condition of the Bohr spectrum being rational up to a constant can be waived if one only demands approximation instead of equality in Corollary 6. This essentially boils down to whether TO is continuous in the system's Hamiltonian.

III. THERMAL OPERATIONS AND CONTINUITY—OR LACK THEREOF

A natural question from a physics perspective is how robust the set of thermal operations is to small changes in temperature or in the energy levels of the system. This question already has a partial answer for inhomogeneous reservoirs and diagonal states from the perspective of work generation and α-free energies.[28] Others also have studied characterizing approximate thermodynamic state transitions via smoothed generalized free energies,[29] as well as the general effect of imperfections (such as finite-time and finite-size) on work extraction and the second law.[30,31] However, it seems that a rigorous study of how the *set* of all thermal operations depends on parameters, such as the temperature or (the spectrum of) the system's Hamiltonian, is still amiss.

For this, we introduce a notion of distance between sets of quantum maps. One way to do this is to use the Hausdorff metric δ [here with respect to $\|\cdot\|_{1\to1}$, that is, the usual operator norm if the domain and range are equipped with the trace norm $\|\cdot\|_1 := \mathrm{tr}(\sqrt{(\cdot)^*(\cdot)})$], which—given non-empty, compact sets $A, B \subset \mathscr{L}(\mathbb{C}^{n\times n})$—is defined to be

$$\delta(A,B) := \max\Big\{\max_{S_1\in A}\min_{S_2\in B}\|S_1 - S_2\|_{1\to1}, \max_{S_2\in B}\min_{S_1\in A}\|S_1 - S_2\|_{1\to1}\Big\}. \tag{6}$$

Here and henceforth, given any vector space V, we write $\mathscr{L}(V)$ for the collection of all linear maps: $V \to V$. Expression (6), indeed, is a metric on $\mathscr{P}_c(\mathscr{L}(\mathbb{C}^{n\times n}))$, with the latter denoting the space of all non-empty compact subsets of $\mathscr{L}(\mathbb{C}^{n\times n})$; cf. Sec. 21.VII in Ref. 32. In particular, this allows one to define a distance between any non-empty sets $A, B \subset \mathrm{CPTP}(n)$ via $\delta(\overline{A}, \overline{B})$.

Based on this definition, we will show that whenever $H_S \in i\mathfrak{u}(n)$ has a degenerate Bohr spectrum (see supplementary note 2 in Ref. 33)—i.e., ad_{H_S} has less than $n^2 - n + 1$ different eigenvalues—then the map

$$\begin{aligned} \mathsf{TO}_n : i\mathfrak{u}(n) \times (0,\infty] &\to \big(\mathscr{P}_c(\mathscr{L}(\mathbb{C}^{n\times n}), \|\cdot\|_{1\to1}), \delta\big) \\ (H,T) &\mapsto \overline{\mathsf{TO}(H,T)} \end{aligned} \tag{7}$$

is discontinuous in (H_S, T) for all temperatures $T \in (0,\infty]$. Note that H_S has a degenerate Bohr spectrum iff either H_S itself is degenerate ($|\sigma(H_S)| < n$) or—assuming $\sigma(H_S) = \{E_1,\dots,E_n\}$ for some $E_1 < \dots < E_n$—if some of the energy transitions that H_S admits coincide, i.e., if the map $(j,k) \mapsto E_j - E_k$ with domain $\{(j,k) : 1 \le j < k \le n\}$ is not injective. With this in mind, we will present two examples, which illustrate how map (7) can fail to be continuous.

Example 7.

(i) *First, we consider the simplest case of a degenerate system's Hamiltonian, that is, $n = 2$ and $H_S = 0$. Given arbitrary $T \in (0,\infty]$, we will show that $\delta(\overline{\mathsf{TO}(0,T)}, \overline{\mathsf{TO}(\mathrm{diag}(0,\varepsilon),T)}) \ge 1$ for all $\varepsilon > 0$, which clearly violates the continuity of TO_2 in $(0,T)$. The reason for this, roughly speaking, is that no thermal operation corresponding to a non-degenerate Hamiltonian can mix diagonal and off-diagonal elements. This is prohibited by the known fact that it has to commute with $\mathrm{ad}_{\mathrm{diag}(0,\varepsilon)}$.*

While it is easy to see that $\overline{\mathsf{TO}(0,T)}$ equals the set of all unital qubit maps (that is, all CPTP maps on $\mathbb{C}^{2\times2}$ for which $\mathbb{1}$ is a fixed point; cf. Appendix D and related footnotes), for our purposes, it suffices to define a map $S \in \mathscr{L}(\mathbb{C}^{2\times2})$ via $S := \Phi_{T,2}(H_S, U)$, where

$$U := \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & -1 \\ 1 & 0 & -1 & 0 \\ 0 & 1 & 0 & 1 \end{pmatrix} \in \mathsf{U}(4).$$

Indeed, $S \in \mathsf{TO}(0,T)$ for all $T \in (0,\infty]$, and one readily verifies that the action of S is given by

$$\begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \mapsto \frac{1}{2}\begin{pmatrix} a_{11} + a_{22} & a_{11} - a_{22} \\ a_{11} - a_{22} & a_{11} + a_{22} \end{pmatrix}.$$

Thus, for all $\varepsilon > 0$, $T \in (0,\infty]$, we compute

$$\begin{aligned} \delta(\overline{\mathsf{TO}(0,T)}, \overline{\mathsf{TO}(\mathrm{diag}(0,\varepsilon),T)}) &\ge \min_{\tilde{S}\in\overline{\mathsf{TO}(\mathrm{diag}(0,\varepsilon),T)}} \ \sup_{A\in\mathbb{C}^{2\times2}, \|A\|_1 = 1} \|S(A) - \tilde{S}(A)\|_1 \\ &\ge \min_{\tilde{S}\in\overline{\mathsf{TO}(\mathrm{diag}(0,\varepsilon),T)}} \|S(|e_1\rangle\langle e_1|) - \tilde{S}(|e_1\rangle\langle e_1|)\|_1 \\ &= \min_{\lambda\in[0,1]} \left\| \frac{1}{2}\begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} - \begin{pmatrix} 1 - \lambda e^{-\varepsilon/T} & 0 \\ 0 & \lambda e^{-\varepsilon/T} \end{pmatrix} \right\|_1 \\ &\ge \min_{\lambda'\in\mathbb{R}} \left\| \begin{pmatrix} \lambda' & \frac{1}{2} \\ \frac{1}{2} & -\lambda' \end{pmatrix} \right\|_1 = \min_{\lambda'\in\mathbb{R}} \sqrt{4(\lambda')^2 + 1} = 1. \end{aligned}$$

In the third step, we used Eq. (8) and Theorem 10 (i).

16 April 2024 10:49:23

(ii) *Having dealt with degenerate Hamiltonians, let us now look at the other possible case: Hamiltonians that are non-degenerate but have a degenerate Bohr spectrum. This can only occur in 3 or more dimensions, so consider* $H_S = \mathrm{diag}(0,1,2)$*. Given arbitrary* $T \in (0,\infty]$*, we will show that* $\delta(\overline{\mathsf{TO}(H_S,T)}, \overline{\mathsf{TO}(H_S+\varepsilon|e_3\rangle\langle e_3|,T)}) \geq \frac{2}{3}$ *for all* $\varepsilon > 0$*, which again violates continuity—the reason for this being similar to the reason from (i). Choose* $H_B := H_S$*, and define*

$$U := \begin{pmatrix} 1&0&0&0&0&0&0&0&0\\ 0&0&0&1&0&0&0&0&0\\ 0&0&0&0&1&0&0&0&0\\ 0&1&0&0&0&0&0&0&0\\ 0&0&0&0&0&0&1&0&0\\ 0&0&0&0&0&0&0&1&0\\ 0&0&1&0&0&0&0&0&0\\ 0&0&0&0&0&1&0&0&0\\ 0&0&0&0&0&0&0&0&1 \end{pmatrix},$$

which corresponds to the permutation (in cycle notation) $(1)\,(24)\,(357)\,(68)\,(9)$*. Therefore,* U *is unitary and satisfies the stabilizer condition* $U(H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B)U^* = H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B$ *because matching diagonal entries of* $H_S\otimes \mathbb{1}_B + \mathbb{1}\otimes H_B$ *precisely correspond to the cycles of* U*. With this,* $S := \Phi_{T,3}(H_S,U)$ *acts on any* $A \in \mathbb{C}^{3\times 3}$ *as follows:*

$$\frac{1}{1+e^{-1/T}+e^{-2/T}}\begin{pmatrix} a_{11}+a_{22}(1+e^{-1/T}) & a_{23}(1+e^{-1/T}) & 0\\ a_{32}(1+e^{-1/T}) & a_{11}e^{-1/T}+a_{33}(1+e^{-1/T}) & 0\\ 0 & 0 & (a_{11}+a_{22}+a_{33})e^{-2/T} \end{pmatrix}.$$

For all $\varepsilon > 0$, $T\in(0,\infty]$*, one finds [similar to (i)]*

$$\delta(\overline{\mathsf{TO}(H_S,T)}, \overline{\mathsf{TO}(H_S+\varepsilon|e_3\rangle\langle e_3|,T)}) \geq \min_{\tilde S\in\overline{\mathsf{TO}(H_S+\varepsilon|e_3\rangle\langle e_3|,T)}} \|S(|e_2\rangle\langle e_3|) - \tilde S(|e_2\rangle\langle e_3|)\|_1.$$

Now, $H_S+\varepsilon|e_3\rangle\langle e_3|$ *has a non-degenerate Bohr spectrum for all* $\varepsilon>0$*, meaning the only thing* $\overline{\mathsf{TO}(H_S+\varepsilon|e_3\rangle\langle e_3|,T)}$ *can do to off-diagonal entries is scale them by a factor* $\gamma\in\mathbb{C}$, $|\gamma|\leq 1$*—this follows from the fact that every thermal operation (with respect to* $H_S+\varepsilon|e_3\rangle\langle e_3|$ *and any* T*) has to commute with* $\mathrm{ad}_{H_S+\varepsilon|e_3\rangle\langle e_3|}$*.*[11] *With this, one obtains the lower bound*

$$\begin{aligned}\delta(\overline{\mathsf{TO}(H_S,T)}, \overline{\mathsf{TO}(H_S+\varepsilon|e_3\rangle\langle e_3|,T)}) &\geq \min_{|\gamma|\leq 1}\left\|\frac{1+e^{-1/T}}{1+e^{-1/T}+e^{-2/T}}|e_1\rangle\langle e_2| - \gamma|e_2\rangle\langle e_3|\right\|_1\\ &= \min_{|\gamma|\leq 1}\left(\frac{1+e^{-1/T}}{1+e^{-1/T}+e^{-2/T}} + |\gamma|\right)\\ &= 1 - \frac{e^{-2/T}}{1+e^{-1/T}+e^{-2/T}} \geq 1 - \sup_{T>0}\frac{1}{e^{2/T}+e^{1/T}+1} = \frac{2}{3}.\end{aligned}$$

It is not difficult to generalize these examples to any Hamiltonians with a degenerate Bohr spectrum in arbitrary dimensions.

The reason for discontinuity in either example was the condition $[S,\mathrm{ad}_{H_S}] = 0$ for all $S\in\overline{\mathsf{TO}}$, which comes solely from $U(H_S\otimes\mathbb{1}_B + \mathbb{1}\otimes H_B)U^* = H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B$. This suggests two things: first, to restore the (physically reasonable) requirement of continuity, one has to somehow relax or alter this condition—more on this in Sec. V. Second, as the temperature does not appear, here, it seems reasonable to conjecture the following.

Conjecture 8. For all $H_S\in i\mathfrak{u}(n)$*, the map* $T\mapsto\overline{\mathsf{TO}(H_S,T)}$ *is continuous if the domain* $(0,\infty]$ *is equipped with the metric* $d_{-1}(T,T') := |\frac{1}{T}-\frac{1}{T'}|$*, and the co-domain is equipped with the Hausdorff metric* δ *with respect to* $(\mathscr{L}(\mathbb{C}^{n\times n}), \|\cdot\|_{1\to 1})$*.*

For a simple yet (so far) unsuccessful attempt to prove this, see Appendix C. Either way, this property would be necessary for the role of the temperature in the definition of thermal operations to be "correct" in the sense that it accurately models the behavior of real physical systems. As a final remark, the case $T = 0$ is excluded from the above continuity considerations for two reasons: first, the concept of zero temperature and achieving it with finite resources (e.g., time, heat baths) is problematic in the classical[34,35] and the quantum case (at least for qubits, cf. Lemma 9 in Ref. 14). Second, letting the temperature tend to zero reveals a lack of continuity already in the classical case; cf. Appendix D, Example 3 in Ref. 16. More precisely, there exist classical states (probability vectors) x such that the map $T \mapsto \{Ax : A$ Gibbs − stochastic[36,48] with respect to $H_S, T\}$ is discontinuous in $T = 0$.

IV. THE QUBIT CASE: OVERVIEW, SEMIGROUP REPRESENTATION, AND VISUALIZATION

Two core features of thermal operations are preservation of the Gibbs state and the covariance law (in generator form) $[S, \mathrm{ad}_{H_S}] = 0$ for all $S \in \overline{\mathsf{TO}(H_S,T)}$.[11] This motivates the following definition:[33] Given $H_S \in i\mathfrak{u}(n)$ and some $T > 0$, the set of all covariant Gibbs-preserving maps is defined to be

$$\mathsf{EnTO}(H_S,T) := \{S \in \mathrm{CPTP}(n) \,:\, S(e^{-H_S/T}) = e^{-H_S/T} \wedge [S, \mathrm{ad}_{H_S}] = 0\},$$

where EnTO is short for "enhanced thermal operations." This definition naturally extends to $T = \infty$ by replacing the fixed point $e^{-H_S/T}$ by $\mathbb{1}_n$. It is straightforward to see that for all $H_S \in i\mathfrak{u}(n)$, $T \in (0, \infty]$, EnTO is a convex, compact semigroup with identity, and EnTO satisfies the same transformation rules as TO and $\overline{\mathsf{TO}}$ (Lemma 2). Moreover, $\overline{\mathsf{TO}} \subseteq \mathsf{EnTO}$, and the action of $\overline{\mathsf{TO}}$ and EnTO on any classical state ρ (i.e., on any state with $[H_S, \rho] = 0$) even coincides; cf. Sec. 3 in Ref. 25.

A set of necessary and sufficient (implicit) conditions for state conversion under enhanced thermal operations was given by Gour *et al.*[18] However, for general systems, TO and EnTO do not agree, even in closure and when restricted to their respective action: Choosing $H_S = \mathrm{diag}(0,1,2)$, there exist temperature $T > 0$, quantum states ρ, ρ', and $S \in \mathsf{EnTO}(H_S,T)$ such that $S(\rho) = \rho'$ but $\rho' \notin \overline{\mathsf{TO}(H_S,T)}(\rho)$.[21] This, however, is only true beyond two dimensions because for qubits, it is known that the two sets coincide. Before we review the many results on qubit thermal operations, let us investigate the basic structure of TO in two dimensions; this will simplify things later on.

The qubit case is particularly nice because there EnTO is characterized by three real parameters (i.e., one real and one complex number), so, in particular, we can visualize it. Indeed, given $H_S \in i\mathfrak{u}(2)$ non-degenerate (i.e., $H_S = \sum_{i=1}^2 E_i |g_i\rangle\langle g_i|$ with $E_1 < E_2$ for some orthonormal basis $\{g_1, g_2\}$ of $\mathbb{C}^2$) and $T \in (0, \infty]$, one finds that a linear map $S : \mathbb{C}^{2\times 2} \to \mathbb{C}^{2\times 2}$ is in $\mathsf{EnTO}(H_S,T)$ if and only if there exist $\lambda \in [0,1]$, $r \in \left[0, \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}\right]$, and $\phi \in [-\pi, \pi)$ such that the Choi matrix[37] of S (with respect to $\{g_1, g_2\}$) reads

$$\begin{pmatrix} 1-\lambda e^{-\Delta E/T} & 0 & 0 & re^{i\phi} \\ 0 & \lambda e^{-\Delta E/T} & 0 & 0 \\ 0 & 0 & \lambda & 0 \\ re^{-i\phi} & 0 & 0 & 1-\lambda \end{pmatrix}. \tag{8}$$

Here, $\Delta E := E_2 - E_1 > 0$, and if $T = \infty$, then $e^{-\Delta E/T}$ gets replaced by 1. The basic structure of (8)—meaning the position of the zeros—is solely due to $[S, \mathrm{ad}_{H_S}] = 0$, while the preservation of the Gibbs state is encoded in the diagonal action being a Gibbs-stochastic 2×2 matrix, where $d = (e^{-E_1/T}, e^{-E_2/T})$. In two dimensions, the latter set is well known to equal

$$\mathrm{conv}\left\{\begin{pmatrix} 1-\dfrac{d_2}{d_1} & 1 \\ \dfrac{d_2}{d_1} & 0 \end{pmatrix}, \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}\right\} = \mathrm{conv}\left\{\begin{pmatrix} 1-e^{-\Delta E/T} & 1 \\ e^{-\Delta E/T} & 0 \end{pmatrix}, \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}\right\},$$

so it is characterized by one parameter $\lambda \in [0,1]$. Finally, the scaling of S on the off-diagonal is only restricted by complete positivity [so positive semi-definiteness of (8); cf. again Ref. 37], which is equivalent to $|r|^2 \le (1-\lambda)(1-\lambda e^{-\Delta E/T})$. This allows us to define the linear map

$$\begin{aligned} \Psi_T : \mathscr{L}(\mathbb{C}^{2\times 2}) &\to \mathbb{R} \times \mathbb{C} \\ S &\mapsto \begin{pmatrix} \langle g_1, S(|g_2\rangle\langle g_2|) g_1\rangle \\ \langle g_1, S(|g_1\rangle\langle g_2|) g_2\rangle \end{pmatrix}, \end{aligned} \tag{9}$$

which maps (8) to $(\lambda, re^{i\phi})$. This becomes a faithful semigroup representation if the domain of Ψ_T is restricted to $D(\Psi_T) :=$ "all linear maps on $\mathbb{C}^{2\times 2}$ whose Choi matrix is given by (8) for some $\lambda \in \mathbb{R}$, $r \ge 0$, $\phi \in [-\pi, \pi)$," and if the codomain of Ψ_T is equipped with the associative operation,

$$\circ_T : (\mathbb{R}\times\mathbb{C})\times(\mathbb{R}\times\mathbb{C}) \to (\mathbb{R}\times\mathbb{C})$$
$$\left(\begin{pmatrix}\lambda_1\\ c_1\end{pmatrix},\begin{pmatrix}\lambda_2\\ c_2\end{pmatrix}\right) \mapsto \begin{pmatrix}\lambda_1+\lambda_2-\lambda_1\lambda_2(1+e^{-1/T})\\ c_1c_2\end{pmatrix},$$

which has neutral element $(0,1)^{\top}$. The image of $\Psi_T : D(\Psi_T) \to (\mathbb{R}\times\mathbb{C},\circ_T)$ is depicted in Fig. 2.

Interestingly, $\circ_T$ operates commutatively, which—because Ψ_T is a faithful semigroup representation of EnTO—yields the following.

Corollary 9. Let $T\in(0,\infty]$, and let $H_S\in i\mathfrak{u}(2)$ be non-degenerate. For all $S_1,S_2\in\mathsf{EnTO}(H_S,T)$, one has $S_1\circ S_2=S_2\circ S_1$. In particular, every subset of $\mathsf{EnTO}(H_S,T)$ is commutative, as well.

Be aware that this result does not hold if H_S is degenerate: for example, the group $\mathrm{Ad}_{\mathsf{SU}(2)}$ is a non-commutative subset of $\mathsf{TO}(\mathbb{1}_2,T)\subseteq\mathsf{EnTO}(\mathbb{1}_2,T)$. Moreover, unsurprisingly, this result does not generalize to higher dimensions because already the Gibbs-stochastic matrices form a non-commutative semigroup in three and more dimensions (Appendix A in Ref. 16). In addition, this semigroup representation turns into a group representation if points of the form $(\frac{1}{1+e^{-1/T}},*)$, $(*,0)$ are excluded from the domain of Ψ_T. Then, the inverse of any (λ,c) from the restricted domain of Ψ_T under $\circ_T$ is given by $(\frac{\lambda}{\lambda(1+e^{-1/T})-1},\frac{1}{c})$. Indeed, if one defines the map $x\circ_a y := x+y-xya$ on $\mathbb{R}\times\mathbb{R}$ for any non-zero a, then

$$\mathfrak{I}_a : (\mathbb{R}\backslash\{a^{-1}\},\circ_a)\to(\mathbb{R}\backslash\{0\},\cdot)$$
$$x\mapsto 1-ax$$

is a group isomorphism because $(1-ax)(1-ay)=1-a(x\circ_a y)$. In particular, the map $\mathfrak{I}_a$ transfers commutativity of $(\mathbb{R}\backslash\{0\},\cdot)$ over to $(\mathbb{R}\backslash\{a^{-1}\},\circ_a)$ and, thus, ultimately to $\circ_T$ because $\circ_T\equiv\circ_a|_{a=1+e^{-1/T}}\times\cdot|_{\mathbb{C}}$. Thus, in a way, Corollary 9 is of the same fundamental structure as the statement that multiplying real numbers is commutative.

With this, we are ready to show how TO sits inside EnTO in two dimensions. It has first been shown by Ćwikliński *et al.* that for all $H_S\in i\mathfrak{u}(2)$ and all $T\in(0,\infty]$, the sets $\overline{\mathsf{TO}(H_S,T)}$ and $\mathsf{EnTO}(H_S,T)$ coincide.[33] Using the above semigroup representation, we outlined their proof in Appendix D. Be aware that their proof relied on bath Hamiltonians with exponentially growing degeneracies of the energy levels. This requirement was eventually shown to be unnecessary: the observation that it suffices to consider truncated single-mode bosonic baths [i.e., $H_B=\Delta E\,\mathrm{diag}(0,1,\ldots,m)$] was first done by Scharlau and Mueller for classical states (Sec. IV in Ref. 14), followed by Hu and Ding[20] for

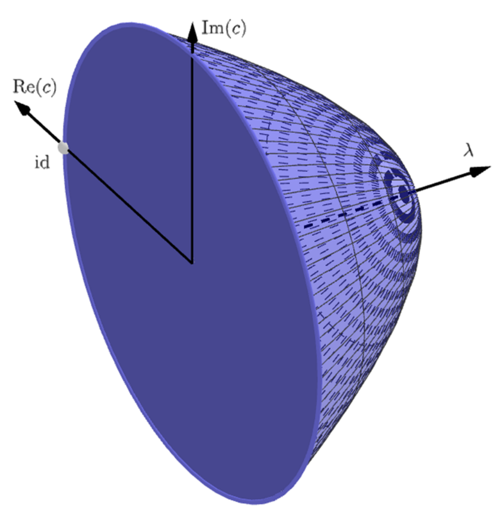

FIG. 2. Graph of Ψ_T from (9) for non-degenerate H_S when restricting the domain to $\mathsf{EnTO}(H_S,T)$. The identity is mapped to $(0,1)^{\top}$. The "classical" channels, i.e., the channels that set all coherences to zero are located on the λ axis. The outer curve in the λ-direction is described by $\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}$, here, depicted for $\Delta E=1$ and $e^{-1/T}=0.2$.

the general case. We summarize and extend on their results in the following theorem. Most notably, the only advantage degenerate baths give over non-degenerate ones is generating full dephasing at low temperatures.

Theorem 10. *Let $H_S \in i\mathfrak{u}(2)$ and $T \in (0, \infty]$ be given. Defining $\Delta E := \max \sigma(H_S) - \min \sigma(H_S)$, the following statements hold:*

(i) $\overline{\mathsf{TO}(H_S, T)} = \mathsf{EnTO}(H_S, T)$.

(ii) *If H_S has a non-degenerate spectrum, then $\mathsf{TO}(H_S, T)$ is convex as it equals*

$$\mathsf{EnTO}(H_S, T) \setminus \Psi_T^{-1}\left(\left\{\begin{pmatrix} \lambda \\ \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}e^{i\phi} \end{pmatrix} : \lambda \in (0,1], \phi \in [-\pi, \pi)\right\}\right). \tag{10}$$

In other words, an enhanced thermal operation can be implemented via a thermal operation if and only if it is a dephasing map ($\lambda = 0$) or if it lies in the relative interior[38] of $\mathsf{EnTO}(H_S, T)$. In particular, the difference between $\mathsf{TO}(H_S, T)$ and $\mathsf{EnTO}(H_S, T)$ occurs only on the relative boundary of the enhanced thermal operations.

(iii) *If H_S has a non-degenerate spectrum, then the semigroup generated by thermal operations with bath Hamiltonians $\mathrm{diag}(0, \Delta E, \ldots, m\Delta E)$ for some $m \in \mathbb{N}_0$ equals $\mathsf{TO}(H_S, T)$ if and only if $T \in (\frac{\Delta E}{\ln 2}, \infty]$. Should the two sets not be the same (i.e., if $T \le \frac{\Delta E}{\ln 2}$), their difference is a subset of $\Psi_T^{-1}(\{(\lambda, 0)^\top : \lambda \in [0,1]\})$ (i.e., the λ axis in Fig. 2); in particular, the two sets coincide after taking the closure.*

(iv) *Every enhanced thermal operation can be realized by a thermal operation with a single-mode bosonic bath, that is, $H_B = \mathrm{diag}\,(j\Delta E)_{j=0}^{\infty}$, if and only if $T \in [\frac{\Delta E}{\ln 2}, \infty]$. Indeed, if $T < \frac{\Delta E}{\ln 2}$, the difference between the two sets has measure strictly larger than zero. This is due to the fact that for small enough λ, the action of thermal operations on the off-diagonal elements cannot become arbitrarily small anymore.*

For convenience, proofs of these results are given in Appendix D. Be aware that most of these results do not generalize to more than two dimensions.

One conclusion "hidden" in the Proof of Theorem 10 concerns the dimension of bath Hamiltonians. Recall that the Stinespring dilation $S = \mathrm{tr}_B(U((\cdot) \otimes |\psi\rangle\langle\psi|)U^*)$ of an arbitrary quantum channel $S \in \mathrm{CPTP}(n)$ can always be chosen such that $\psi \in \mathbb{C}^k$ for some $k \le n^2$ (Theorem 6.18 in Ref. 39). This result breaks down for thermal operations, that is, if $|\psi\rangle\langle\psi|$ is replaced by a Gibbs state and U is required to be energy-preserving: For every $\lambda \in (0,1)$, $m \in \mathbb{N}$, there exist $\varepsilon_m > 0$ and $r \in (\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})} - \varepsilon_m, \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})})$ such that the thermal operation $\Psi_T^{-1}(\lambda, r)$ can be implemented by a bath Hamiltonian H_B only if it is of size $m \times m$ or larger. However, ε_m goes to zero as $m \to \infty$ because $\mathsf{TO}(H_S, T)$ gets arbitrarily close to the (relative) boundary of EnTO for all $T \in (0, \infty]$.

The final observation we want to make is that the "geometry" of the qubit thermal operations pertains to the set $\{\Phi(\rho) : \Phi \in \overline{\mathsf{TO}(H_S, T)}\}$—which is sometimes referred to as the (future) thermal cone[11,40,41]—as is depicted in Fig. 3. This recovers what has already been observed in Ref. 11, specifically Figs. 1 and 6 in the said article. In particular, the boundary of the thermal cone is not linear, meaning that even after factoring out the rotational symmetry inflicted by the thermal operations $\rho \mapsto \mathrm{diag}(1, e^{i\phi})\rho\,\mathrm{diag}(1, e^{-i\phi})$, the set of extreme points is still infinite.

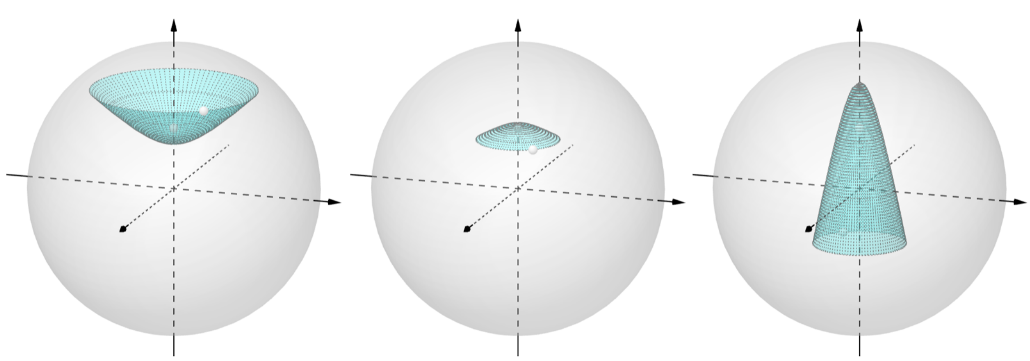

FIG. 3. Future thermal cones of initial states with Bloch vector $c \cdot (0.5, 0.4, \sqrt{0.59})^\top$ for different values of c. Left: $c = 0.9$. Middle: $c = 0.45$. Right: $c = -0.5$. The "system parameter" we chose is $e^{-H_S/T} = 0.45$. Here, the point on the boundary of the cone is the Bloch vector of the initial state, and the point in the interior of the cone (on the z axis) corresponds to the Gibbs state.

V. CONCLUSION AND OPEN QUESTIONS

We reduced the set of bath Hamiltonians needed in the definition of thermal operations to those that have the so-called "resonant spectrum" with respect to the system. This resonance condition is about the spectrum of the bath forming a connected graph with respect to the possible energy transitions of the system (cf. Fig. 1). We saw that the action coming from any bath that does not satisfy this condition decomposes into the convex sum of two or more thermal operations with a resonant bath. Be aware that this notion is logically independent of a bath Hamiltonian containing *all* possible transitions of the system [i.e., $\sigma(\mathrm{ad}_{H_S}) \subseteq \sigma(\mathrm{ad}_{H_B})$]. The latter is a necessary condition for the diagonal action of a thermal operation to be represented by a strictly positive Gibbs-stochastic matrix (cf. also Remark 3).

Either way, as a consequence of the new-found resonance, we showed that if any multiple of the system's Hamiltonian has a rational Bohr spectrum, then there exists an energy gap such that the set of thermal operations is fully characterized by spin Hamiltonians with respect to this energy gap (Corollary 6). As rational numbers are a key concept of this statement, this suggests that the thermal operations behave discontinuously at certain Hamiltonians. Indeed, we were able to show that if either the spectrum or the transitions of the system's Hamiltonian are degenerate, then the set of thermal operations changes discontinuously with respect to the Hausdorff metric (Example 7). Taking the nature of our two counter-examples into account, it seems reasonable to conjecture that TO is at least continuous in the temperature (for fixed Hamiltonian), as well as TO admitting some form of semi-continuity in the joint argument (H_S, T).

An idea to restore continuity—inspired by the concept of average energy conservation[13]—could be to allow for an error in the energy conservation condition: Given any $\varepsilon \geq 0$, define $\mathsf{TO}^\varepsilon(H_S, T)$ by adjusting $U(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^* = H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B$ in the definition of $\mathsf{TO}(H_S, T)$ to $\|U(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^* - H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B\|_\infty \leq 2\varepsilon\|U - \mathbb{1}\|_\infty(\|H_S\|_\infty + \|H_B\|_\infty)$. This is motivated by the simple estimate

$$\begin{aligned}
&\|U(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^* - H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B\|_\infty \\
&\qquad \leq \|U(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^* - (H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^*\|_\infty \\
&\qquad\quad + \|(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^* - H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B\|_\infty \\
&\qquad \leq 2\|U - \mathbb{1}\|_\infty\|H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B\|_\infty = 2\|U - \mathbb{1}\|_\infty(\|H_S\|_\infty + \|H_B\|_\infty).
\end{aligned}$$

Note that TO^0 recovers TO, while TO^ε for $\varepsilon \geq 1$ renders energy conservation obsolete because, then, every unitary satisfies the condition in question.

In some way, introducing such ε (small enough) "smoothens out" the binary nature of energy-conservation by allowing for unitaries, which are ε-close to conserving the energy of the full uncoupled system. As a result, new transitions that were previously forbidden do not appear instantly once the Bohr spectrum becomes degenerate, but the norm of the corresponding diagonal block in the unitary correlates with the error ε. Now, in order to get a collection of maps, which is "physically reasonable," one may have to intersect TO^ε with the Gibbs-preserving maps or maybe consider the semigroup generated by TO^ε.

Finally, we reviewed what is known about thermal operations in the qubit case and, using our results on baths with a resonant spectrum, extended on this knowledge by specifying how the set of thermal qubit operations looks *exactly*. We did so by means of a faithful semigroup representation, which translates the (enhanced) thermal operations into a subset of ordinary 3D space, thus allowing for a visualization from which intuition benefits as well. Interestingly, our proof of the main qubit results (Theorem 10) gave two different families of energy-preserving unitaries for approximating the extreme points of the enhanced thermal operations depending on whether the temperature is finite or infinite. For now, finding a (temperature-dependent) family of unitaries, which continues to do the job in the limit $T \to \infty$, is an open problem.

In any case, these qubit results readily lead us to a number of open questions for general (finite-dimensional) systems, two of the more obvious ones being the following:

- Is $\mathsf{TO}(H_S, T)$ convex for all $H_S \in i\mathfrak{u}(n)$, $T \in (0, \infty]$? We showed that this holds true in two dimensions; however, our proof—as well as the proof of convexity of $\overline{\mathsf{TO}}$—is very much unsuited to tackle this question in higher dimensions because they are either too complicated or they fundamentally rely on rational numbers and approximations. Should convexity hold, in general, (without the closure), it seems likely that proving so requires some deeper knowledge about thermal operations.
- Can one specify an upper bound for how degenerate the energy levels of the bath need to be? For qubits, our Proof of Proposition 4 (i) shows that every qubit thermal operation is the composition of something "close to an extreme point" (i.e., non-degenerate bath) and a partial dephasing, which can always be implemented by a trivial two-level bath. Then, the proof of the semigroup property shows that the energy levels of the bath Hamiltonian of the composite operation have degeneracy at most two. Thus, one may conjecture that the definition of TO can be restricted to such (resonant) bath Hamiltonians, which have degeneracy at most dimension of the system. This claim is further supported by the fact that full dephasing in n dimensions can always be realized by choosing $H_B = \mathbb{1}_n$ [together with $U = \oplus_{j=1}^n \mathrm{diag}\,(e^{2\pi ijk/n})_{k=0}^{n-1}$]; cf. p. 88 in Ref. 23.

Generally speaking, settling which results regarding $\overline{\mathsf{TO}}$ continue to hold once the closure is waived should be a future line of research. We expect that any progress in this direction will reveal more of the intrinsic structure the set of thermal operations has.

ACKNOWLEDGMENTS

I would like to thank Emanuel Malvetti, Gunther Dirr, Thomas Schulte-Herbrüggen, and Amit Devra for valuable and constructive comments during the preparation of the manuscript. Moreover, I am grateful to the anonymous referee for the valuable comments, which led to an improved presentation of the material. This research was part of the Bavarian excellence network ENB via the International Ph.D. Program of Excellence Exploring Quantum Matter (EXQM) and the Munich Quantum Valley of the Bavarian State Government with funds from Hightech Agenda Bayern Plus.

AUTHOR DECLARATIONS

Conflict of Interest

The author has no conflicts to disclose.

Author Contributions

Frederik vom Ende: Conceptualization (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (equal); Writing – original draft (equal); Writing – review and editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

APPENDIX A: PROOF OF PROPOSITION 4

We will only prove the case $T \in (0,\infty)$ as the case $T = \infty$ is done analogously.

(i) While the semigroup property is well-known, we still give a proof here for the sake of completeness. Given thermal operations S_i, $i = 1, 2$, with associated bath Hamiltonian $H_{B,i} \in \mathbb{C}^{m_i\times m_i}$ and energy-preserving unitary $U_i \in \mathbb{C}^{m_i n\times m_i n}$, respectively, we claim that $S_1 \circ S_2 = \Phi_{T,m_1m_2}(H_B, U)$ with $H_B := H_{B,1} \otimes \mathbb{1}_{m_2} + \mathbb{1}_{m_1} \otimes H_{B,2}$ and

$$U := (U_1 \otimes \mathbb{1}_{m_2})(\mathbb{1}_n \otimes \mathbb{F}^*)(U_2 \otimes \mathbb{1}_{m_1})(\mathbb{1}_n \otimes \mathbb{F}) \in \mathbb{C}^{n\times n} \otimes \mathbb{C}^{m_1\times m_1} \otimes \mathbb{C}^{m_2\times m_2}. \tag{A1}$$

Here, $\mathbb{F} : \mathbb{C}^{m_1} \otimes \mathbb{C}^{m_2} \to \mathbb{C}^{m_2} \otimes \mathbb{C}^{m_1}$ is the flip operator, i.e., the unique linear operator, which satisfies $\mathbb{F}(x \otimes y) = y \otimes x$ for all $x \in \mathbb{C}^{m_1}$, $y \in \mathbb{C}^{m_2}$. Note that $\mathbb{F}$ also "generates" the matrix flip, that is, $\mathbb{F}^*(B \otimes A)\mathbb{F} = A \otimes B$ for all $A \in \mathbb{C}^{m_1\times m_1}, B \in \mathbb{C}^{m_2\times m_2}$. Now, the idea as to why $S_1 \circ S_2$ can be described in such a way is depicted in Fig. 4.

The key tool one uses in this proof is the following partial trace identity: Given Hilbert spaces $\mathscr{H}_1, \mathscr{H}_2$, a trace-class operator A on $\mathscr{H}_1 \otimes \mathscr{H}_2$, and bounded linear operators C, D on $\mathscr{H}_1$ and B on $\mathscr{H}_2$ such that B is invertible, one readily verifies

$$\mathrm{tr}_{\mathscr{H}_2}\big((C \otimes B)A(D \otimes B^{-1})\big) = C\,\mathrm{tr}_{\mathscr{H}_2}(A)D. \tag{A2}$$

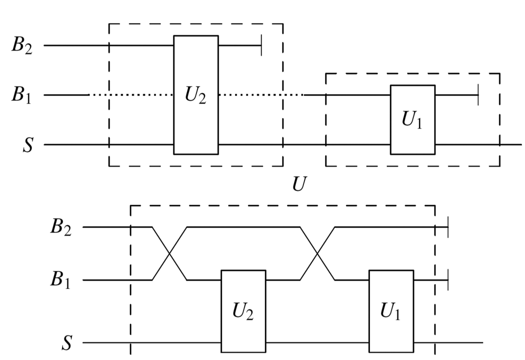

FIG. 4. Top: Circuit of first applying S_2 followed by applying S_1 (i.e., $S_1 \circ S_2$). Bottom: Circuit of $\Phi_{T,m_1m_2}(H, U)$ with U from (A1). The idea as to why the action of these circuits coincides is that tracing out the bath B_2 commutes with applying U_1 because the action of the latter on B_2 is trivial.

Consequently, given Hilbert spaces $\mathscr{H}_1, \mathscr{H}_2, \mathscr{H}_3$ and trace-class operators X on $\mathscr{H}_1 \otimes \mathscr{H}_2$ and Y on $\mathscr{H}_3$, one finds

$$\operatorname{tr}_{\mathscr{H}_2}(X) \otimes Y = \operatorname{tr}_{\mathscr{H}_2}\big((\mathbb{1} \otimes \mathbb{F}^*)(X \otimes Y)(\mathbb{1} \otimes \mathbb{F})\big), \tag{A3}$$

where $\operatorname{tr}_{\mathscr{H}_2}$ on the right-hand side of Eq. (A3) is the partial trace on $(\mathscr{H}_1 \otimes \mathscr{H}_3) \otimes \mathscr{H}_2$ and, again, $\mathbb{F} : \mathscr{H}_3 \otimes \mathscr{H}_2 \to \mathscr{H}_2 \otimes \mathscr{H}_3$ is the flip operator. Note that (A2) implies (A3) by choosing $C = D = \mathbb{1}$, $B = \mathbb{F}$, and $A = X \otimes Y$. With all of this, we compute that $S_1 \circ S_2$ equals

$$\begin{aligned}
&\operatorname{tr}_{B_1} \circ \operatorname{Ad}_{U_1} \circ \left((\cdot) \otimes \frac{e^{-H_{B,1}/T}}{\operatorname{tr}(e^{-H_{B,1}/T})}\right) \circ \operatorname{tr}_{B_2} \circ \operatorname{Ad}_{U_2} \circ \left((\cdot) \otimes \frac{e^{-H_{B,2}/T}}{\operatorname{tr}(e^{-H_{B,2}/T})}\right)\\
&\overset{(A3)}{=} \operatorname{tr}_{B_1} \circ \operatorname{Ad}_{U_1} \circ \operatorname{tr}_{B_2} \circ \operatorname{Ad}_{\mathbb{1}\otimes\mathbb{F}^*} \circ \left((\cdot) \otimes \frac{e^{-H_{B,1}/T}}{\operatorname{tr}(e^{-H_{B,1}/T})}\right) \circ \operatorname{Ad}_{U_2} \circ \left((\cdot) \otimes \frac{e^{-H_{B,2}/T}}{\operatorname{tr}(e^{-H_{B,2}/T})}\right)\\
&\overset{(A2)}{=} \operatorname{tr}_{B_1,B_2} \circ \operatorname{Ad}_{U_1\otimes\mathbb{1}} \circ \operatorname{Ad}_{\mathbb{1}\otimes\mathbb{F}^*} \circ \left((\cdot) \otimes \frac{e^{-H_{B,1}/T}}{\operatorname{tr}(e^{-H_{B,1}/T})}\right) \circ \operatorname{Ad}_{U_2} \circ \left((\cdot) \otimes \frac{e^{-H_{B,2}/T}}{\operatorname{tr}(e^{-H_{B,2}/T})}\right)\\
&= \operatorname{tr}_{B_1,B_2} \circ \operatorname{Ad}_{U_1\otimes\mathbb{1}} \circ \operatorname{Ad}_{\mathbb{1}\otimes\mathbb{F}^*} \circ \operatorname{Ad}_{U_2\otimes\mathbb{1}} \circ \left((\cdot) \otimes \frac{e^{-H_{B,1}/T}}{\operatorname{tr}(e^{-H_{B,1}/T})}\right) \circ \left((\cdot) \otimes \frac{e^{-H_{B,2}/T}}{\operatorname{tr}(e^{-H_{B,2}/T})}\right)\\
&\overset{(A1)}{=} \operatorname{tr}_{B_1,B_2} \circ \operatorname{Ad}_{U} \circ \operatorname{Ad}_{\mathbb{1}\otimes\mathbb{F}^*} \circ \left((\cdot) \otimes \frac{e^{-H_{B,2}/T}}{\operatorname{tr}(e^{-H_{B,2}/T})} \otimes \frac{e^{-H_{B,1}/T}}{\operatorname{tr}(e^{-H_{B,1}/T})}\right) = \Phi_{T,m_1m_2}(H,U).
\end{aligned}$$

Finally, let us sketch why U is energy-preserving by tracking how each of the three components of $H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_B = H_S \otimes \mathbb{1} \otimes \mathbb{1} + \mathbb{1} \otimes H_{B,1} \otimes \mathbb{1} + \mathbb{1} \otimes \mathbb{1} \otimes H_{B,2}$ change with the factors of U,

$$\begin{bmatrix} H_S \otimes \mathbb{1} \otimes \mathbb{1} \\ \mathbb{1} \otimes H_{B,1} \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,2} \end{bmatrix} \xrightarrow{\operatorname{Ad}_{\mathbb{1}\otimes\mathbb{F}}} \begin{bmatrix} H_S \otimes \mathbb{1} \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,1} \\ \mathbb{1} \otimes H_{B,2} \otimes \mathbb{1} \end{bmatrix} \xrightarrow{\operatorname{Ad}_{U_2\otimes\mathbb{1}}} \begin{bmatrix} U_2(H_S \otimes \mathbb{1})U_2^* \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,1} \\ U_2(\mathbb{1} \otimes H_{B,2})U_2^* \otimes \mathbb{1} \end{bmatrix}.$$

However, the sum of these three matrices is equal to $H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_B$ because U_2 is energy-preserving with respect to $(H_S, H_{B,2})$, that is, $U_2(H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_{B,2})U_2^* = H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_{B,2}$. Similarly, one finds

$$\begin{bmatrix} H_S \otimes \mathbb{1} \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,1} \\ \mathbb{1} \otimes H_{B,2} \otimes \mathbb{1} \end{bmatrix} \xrightarrow{\operatorname{Ad}_{\mathbb{1}\otimes\mathbb{F}^*}} \begin{bmatrix} H_S \otimes \mathbb{1} \otimes \mathbb{1} \\ \mathbb{1} \otimes H_{B,1} \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,2} \end{bmatrix} \xrightarrow{\operatorname{Ad}_{U_1\otimes\mathbb{1}}} \begin{bmatrix} U_1(H_S \otimes \mathbb{1})U_1^* \otimes \mathbb{1} \\ U_1(\mathbb{1} \otimes H_{B,1})U_1^* \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,2} \end{bmatrix} \simeq \begin{bmatrix} H_S \otimes \mathbb{1} \otimes \mathbb{1} \\ \mathbb{1} \otimes H_{B,1} \otimes \mathbb{1} \\ \mathbb{1} \otimes \mathbb{1} \otimes H_{B,2} \end{bmatrix}.$$

Boundedness of TO comes from the known fact[42] that the CPTP maps form a subset of the unit sphere with respect to $\|\cdot\|_{1\to1}$. Path-connectedness follows from the fact that every thermal operation can be connected to the identity in a continuous manner: Given $S = \operatorname{tr}_B(e^{iH_{\mathtt{tot}}}((\cdot) \otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})})e^{-iH_{\mathtt{tot}}})$ with $H_{\mathtt{tot}}$ energy-preserving,

$$t \mapsto \operatorname{tr}_B\left(e^{itH_{\mathtt{tot}}}\left((\cdot) \otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})}\right)e^{-itH_{\mathtt{tot}}}\right)$$

is a continuous curve in $\mathsf{TO}(H_S, T)$, which connects S $(t = 1)$ with id $(t = 0)$.

(ii) The only non-trivial things here are convexity and the semigroup property. First, $\overline{\mathsf{TO}}$ is a semigroup because it is the closure of a semigroup in a space where left- and right-multiplication are continuous. This is a general fact: Given any Hausdorff topological space (X, τ) with a binary operation $\circ : X \times X \to X$, which is left- and right-continuous, i.e., $x \mapsto x \circ y$ and $x \mapsto y \circ x$ are continuous for all $y \in X$, if $S \subset X$ is a semigroup (with respect to $\circ$), then $\overline{S}$ is a semigroup, as well. The idea is to first show that $\overline{S} \circ S \subseteq \overline{S}$ by means of nets using continuity of right-multiplication. Based on this, one sees that $\overline{S} \circ \overline{S} \subseteq \overline{\overline{S}} = \overline{S}$ in a similar fashion.

For convexity, one first shows that for any two thermal operations S_1, S_2 and any $\lambda \in (0,1) \cap \mathbb{Q}$, the convex combination $\lambda S_1 + (1-\lambda)S_2$ again is a thermal operation; cf. Appendix C in Ref. 27. Indeed, let such λ and $S_i \in \mathsf{TO}(H_S, T)$ generated by $H_{B,i} \in \mathbb{C}^{m_i \times m_i}$ Hermitian and $U_i \in \mathsf{U}(m_i n)$ energy-preserving, respectively, for $i = 1, 2$, be given. There exist $k, d \in \mathbb{N}$, $k < d$ such that $\lambda = \frac{k}{d}$. We claim that $\lambda S_1 + (1-\lambda)S_2 = \Phi_{T,m_1m_2d}(H_B, U)$, where $H_B = H_{B,1} \otimes \mathbb{1} \otimes \mathbb{1}_d + \mathbb{1} \otimes H_{B,2} \otimes \mathbb{1}_d$ and $U = U_1 \otimes \mathbb{1} \otimes \Pi + (\mathbb{1} \otimes \mathbb{F}^* \otimes \mathbb{1})(U_2 \otimes \mathbb{1} \otimes (\mathbb{1}_d - \Pi))(\mathbb{1} \otimes \mathbb{F} \otimes \mathbb{1})$. Here, Π is any orthogonal projection on $\mathbb{C}^d$ of rank k and $\mathbb{F}$ is the flip operator from earlier. Using (A3) and the fact that $\Pi(1-\Pi) = 0$, one directly computes that U is unitary and energy-preserving and that

$$\Phi_{T,m_1m_2d}(H_B,U)=\operatorname{tr}\bigg(\frac{e^{-H_{B,2}/T}}{\operatorname{tr}(e^{-H_{B,2}/T})}\bigg)\frac{\operatorname{tr}(\Pi)}{d}S_1+\operatorname{tr}\bigg(\frac{e^{-H_{B,1}/T}}{\operatorname{tr}(e^{-H_{B,1}/T})}\bigg)\frac{\operatorname{tr}(\mathbb{1}_d-\Pi)}{d}S_2$$

as desired. This intermediate result will carry over to $\overline{\mathsf{TO}}$ simply by combining it with two approximation arguments. Indeed, let T_1, $T_2\in\overline{\mathsf{TO}(H_S,T)}$, $\lambda\in(0,1)$, and $\varepsilon>0$ be given. On the one hand, there exist thermal operations S_1,S_2 with $\|T_i-S_i\|_{1\to1}<\min\{\frac{\varepsilon}{8\lambda},\frac{\varepsilon}{2}\}$ for $i=1,2$, and on the other hand, one finds $\mu\in(0,1)\cap\mathbb{Q}$ with $|\mu-\lambda|<\frac{\varepsilon}{8}$. By our previous considerations, we know that $\mu S_1+(1-\mu)S_2$ is a thermal operation, which—as we will compute now—is ε-close to $\lambda T_1+(1-\lambda)T_2$. Because ε is arbitrary, this would show $\lambda T_1+(1-\lambda)T_2\in\overline{\mathsf{TO}(H_S,T)}$. Indeed,

$$\begin{aligned}
&\|(\lambda T_1+(1-\lambda)T_2)-(\mu S_1+(1-\mu)S_2)\|_{1\to1}\\
&\quad\le\|\lambda T_1-\mu S_1\|_{1\to1}+\|T_2-S_2\|_{1\to1}+\|\lambda T_2-\mu S_2\|_{1\to1}\\
&\quad<\|\lambda T_1-\lambda S_1\|_{1\to1}+\|\lambda S_1-\mu S_1\|_{1\to1}+\frac{\varepsilon}{2}+\|\lambda T_2-\lambda S_2\|_{1\to1}+\|\lambda S_2-\mu S_2\|_{1\to1}\\
&\quad<\lambda\cdot\frac{\varepsilon}{8\lambda}+\frac{\varepsilon}{8}\|S_1\|_{1\to1}+\frac{\varepsilon}{2}+\lambda\cdot\frac{\varepsilon}{8\lambda}+\frac{\varepsilon}{8}\|S_2\|_{1\to1}=\varepsilon.
\end{aligned}$$

Here, we used that $\|S_1\|_{1\to1},\|S_2\|_{1\to1}=1$ as stated previously.

(iii) This is implied by energy conservation because $e^{-(H_S\otimes\mathbb{1}_B+\mathbb{1}\otimes H_B)/T}=e^{-H_S/T}\otimes e^{-H_B/T}$. In addition, the subset of all CPTP maps, which have $e^{-H_S/T}$ as a common fixed point, is closed, so the inclusion continues to hold when replacing TO by its closure.

(iv) There are two steps to this proof: first, we show that the rhs of (4) is convex, followed by proving the more important fact that $\mathsf{TO}(H_S,T)$ is a subset of the convex hull of the right-hand side of (4). The statement in question then follows from

$$\begin{aligned}
\overline{\mathsf{TO}(H_S,T)}\subseteq\overline{\operatorname{conv}(\text{ rhs of }(4))}&=\operatorname{conv}(\overline{\text{ rhs of }(4)})\\
&=\operatorname{conv}(\text{ rhs of }(4))=\text{ rhs of }(4)\subseteq\overline{\mathsf{TO}(H_S,T)}.
\end{aligned}$$

In the first equality, we used that every bounded subset A of a finite-dimensional vector space satisfies $\operatorname{conv}\overline{A}=\overline{\operatorname{conv}A}$.

Step 1: Convexity is proven just like the convexity of $\overline{\mathsf{TO}}$ in (ii): first, one shows that any rational convex combination is in the set exactly, and for irrational convex coefficients, one at least ends up in the closure. The only thing that changes about the proof: one has to show that if $H_{B,1}\in i\mathfrak{u}(m_1)$, $H_{B,2}\in i\mathfrak{u}(m_2)$ have a resonant spectrum with respect to H_S, then so does $H_{B,1}\otimes\mathbb{1}+\mathbb{1}\otimes H_{B,2}$. Indeed, if $H_{B,i}=\operatorname{diag}((E_1')_i,\ldots,(E_{m_i}')_i)$, $i=1,2$, let us write out $\sigma(H_{B,1}\otimes\mathbb{1}+\mathbb{1}\otimes H_{B,2})=\sigma(H_{B,1})+\sigma(H_{B,2})$ in the following way:

$$\big((E_{i_1}')_1+(E_{i_2}')_2\big)_{i_1=1,i_2=1}^{m_1,m_2}=\begin{pmatrix}(E_1')_1+(E_1')_2&(E_1')_1+(E_2')_2&\cdots&(E_1')_1+(E_{m_2}')_2\\(E_2')_1+(E_1')_2&(E_2')_1+(E_2')_2&\ddots&\vdots\\\vdots&\ddots&\ddots&\vdots\\(E_{m_1}')_1+(E_1')_2&\cdots&\cdots&(E_{m_1}')_1+(E_{m_2}')_2\end{pmatrix}.$$

Now, given an arbitrary proper non-empty subset I of $\{1,\ldots,m_1\}\times\{1,\ldots,m_2\}$, there certainly exists either a row or a column in the above matrix, which features indices (i_1,i_2) from I and indices from the complement I^{C} of I. If we assume w.l.o.g. that this property is satisfied by the row $k\in\{1,\ldots,m_2\}$, this means that

$$I_k:=I\cap\{(j,k):j=1,\ldots,m_1\}\neq\emptyset\quad\text{ and }\quad\{(j,k):j=1,\ldots,m_1\}\not\subset I.$$

In particular, I_k is a proper non-empty subset of $\{1,\ldots,m_1\}\times\{k\}$, so because H_{B_1} is resonant with respect to H_S, there exist $(i_1,k)\in I_k$, $(j_1,k)\in I_k^{\mathsf{C}}$ such that $(E_{i_1}')_1-(E_{j_1}')_1\in\sigma(\operatorname{ad}_{H_S})$. Therefore,

$$\big((E_{i_1}')_1+(E_k')_2\big)-\big((E_{j_1}')_1+(E_k')_2\big)=(E_{i_1}')_1-(E_{j_1}')_1\in\sigma(\operatorname{ad}_{H_S}),$$

which— because $(i_1,k)\in I$, $(j_1,k)\in I^{\mathsf{C}}$—shows that $H_{B,1}\otimes\mathbb{1}+\mathbb{1}\otimes H_{B,2}$ is resonant with respect to H_S as claimed.

Step 2: By Lemma 2, we can assume w.l.o.g. that H_S is diagonal in the standard basis, i.e., $H_S = \mathrm{diag}(E_1,\dots,E_n)$. This will make defining certain objects less tedious. Now, let $H_B = \mathrm{diag}(E'_1,\dots,E'_m) \in i\mathfrak{u}(m)$, $U \in \mathrm{U}(mn)$ be given such that H_B does not have a resonant spectrum with respect to H_S (else we would be done). Hence, there exists $I \subsetneq \{1,\dots,m\}$, $I \neq \emptyset$ such that

$$\{E'_i - E'_j : i \in I, j \in \{1,\dots,m\}\backslash I\} \cap \sigma(\mathrm{ad}_{H_S}) = \emptyset.$$

We will show that this partition of the index set $\{1,\dots,m\}$ implies a decomposition of H_B, U into smaller submatrices such that the resulting thermal operations recreate the original map via a convex combination. More precisely, we define

$$H_{B,1} := \mathrm{diag}\,(E'_i)_{i\in I}, \quad H_{B,2} = \mathrm{diag}\,(E'_j)_{j\in\{1,\dots,m\}\backslash I}$$

and

$$U_1 := (\langle e_i, Ue_j\rangle)_{i,j\in I'}, \quad U_2 := (\langle e_i, Ue_j\rangle)_{i,j\in\{1,\dots,mn\}\backslash I'},$$

where $I' := \{j + (k-1)m : j \in I, k = 1,\dots,n\}$. We claim that

$$\Phi_{T,m}(H_B, U) = \frac{\mathrm{tr}(e^{-H_{B,1}/T})}{\mathrm{tr}(e^{-H_B/T})}\Phi_{T,|I|}(H_{B,1}, U_1) + \frac{\mathrm{tr}(e^{-H_{B,2}/T})}{\mathrm{tr}(e^{-H_B/T})}\Phi_{T,m-|I|}(H_{B,2}, U_2). \tag{A4}$$

The easiest way to see this is by decomposing H_B into blocks. Define $\mathbf{\Pi}_I := \sum_{i\in I}|e_i\rangle\langle e_i|$, and note that $[\mathbf{\Pi}_I, H_B] = 0$. We compute

$$\begin{aligned} H_B &= (\Pi_I + (\mathbb{1} - \Pi_I))H_B(\Pi_I + (\mathbb{1} - \Pi_I)) \\ &= \Pi_I H_B \Pi_I + [H_B, \Pi_I]\Pi_I + \Pi_I[\Pi_I, H_B] + (\mathbb{1} - \Pi_I)H_B(\mathbb{1} - \Pi_I) \\ &= \Pi_I H_B \Pi_I + (\mathbb{1} - \Pi_I)H_B(\mathbb{1} - \Pi_I). \end{aligned}$$

This "block structure" of H_B carries over to U via energy conservation: Given $a, b \in \{1,\dots,n\}$ and $i \in I$, $j \in \{1,\dots,m\}\backslash I$, the energy-conservation condition implies

$$\begin{aligned} 0 &= \langle e_a \otimes e_i, [U, H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_B]e_b \otimes e_j\rangle \\ &= \big((E_b - E_a) - (E'_i - E'_j)\big)\langle e_a \otimes e_i, U(e_b \otimes e_j)\rangle. \end{aligned}$$

However, $E'_i - E'_j \notin \sigma(\mathrm{ad}_{H_S})$ by assumption, while $E_b - E_a \in \sigma(\mathrm{ad}_{H_S})$, meaning that the prefactor is non-zero; hence, $\langle e_a \otimes e_i, U(e_b \otimes e_j)\rangle = 0$. Therefore,

$$(\mathbb{1} \otimes \Pi_I)U(\mathbb{1} - (\mathbb{1} \otimes \Pi_I)) = \sum_{a,b=1}^{n} \sum_{\substack{i\in I \\ j\in\{1,\dots,m\}\backslash I}} \langle e_a \otimes e_i, U(e_b \otimes e_j)\rangle |e_a \otimes e_i\rangle\langle e_b \otimes e_j| = 0,$$

and similarly for $(\mathbb{1} - (\mathbb{1} \otimes \Pi_I))U(\mathbb{1} \otimes \Pi_I)$. This—just as for H_B—yields the block-decomposition $U = \Pi_{I'}U\Pi_{I'} + (\mathbb{1} - \Pi_{I'})U(\mathbb{1} - \Pi_{I'})$, where

$$\begin{aligned} \Pi_{I'} &:= \sum_{i\in I'}|e_i\rangle\langle e_i| = \sum_{j\in I}\sum_{k=1}^{n}|e_{j+(k-1)m}\rangle\langle e_{j+(k-1)m}| \\ &= \sum_{j\in I}\sum_{k=1}^{n}|e_k \otimes e_j\rangle\langle e_k \otimes e_j| \\ &= \left(\sum_{k=1}^{n}|e_k\rangle\langle e_k|\right) \otimes \left(\sum_{j\in I}|e_j\rangle\langle e_j|\right) = \mathbb{1} \otimes \Pi_I. \end{aligned}$$

Inserting this decomposition of U, H_B into the definition of the associated thermal operation yields

$$\Phi_{T,m}(H_B,U) = \mathrm{tr}_B\circ \mathrm{Ad}_{\Pi_{I'}U\Pi_{I'}+(\mathbb{1}-\Pi_{I'})U(\mathbb{1}-\Pi_{I'})}\circ\Big((\cdot)\otimes\frac{\Pi_I e^{-H_B/T}\Pi_I+(\mathbb{1}-\Pi_I)e^{-H_B/T}(\mathbb{1}-\Pi_I)}{\mathrm{tr}(e^{-H_B/T})}\Big)$$
$$= \mathrm{tr}_B\circ \mathrm{Ad}_{\Pi_{I'}U\Pi_{I'}}\circ\Big((\cdot)\otimes\frac{\Pi_I e^{-H_B/T}\Pi_I}{\mathrm{tr}(e^{-H_B/T})}\Big)+\mathrm{tr}_B\circ \mathrm{Ad}_{(\mathbb{1}-\Pi_{I'})U(\mathbb{1}-\Pi_{I'})}\circ\Big((\cdot)\otimes\frac{(\mathbb{1}-\Pi_I)e^{-H_B/T}(\mathbb{1}-\Pi_I)}{\mathrm{tr}(e^{-H_B/T})}\Big)$$
$$= \frac{\mathrm{tr}(\Pi_I e^{-H_B/T}\Pi_I)}{\mathrm{tr}(e^{-H_B/T})}\mathrm{tr}_B\circ \mathrm{Ad}_{\Pi_{I'}U\Pi_{I'}}\circ\Big((\cdot)\otimes\frac{\Pi_I e^{-H_B/T}\Pi_I}{\mathrm{tr}(\Pi_I e^{-H_B/T}\Pi_I)}\Big)$$
$$+\Big(1-\frac{\mathrm{tr}(\Pi_I e^{-H_B/T}\Pi_I)}{\mathrm{tr}(e^{-H_B/T})}\Big)\mathrm{tr}_B\circ \mathrm{Ad}_{(\mathbb{1}-\Pi_{I'})U(\mathbb{1}-\Pi_{I'})}\circ\Big((\cdot)\otimes\frac{(\mathbb{1}-\Pi_I)e^{-H_B/T}(\mathbb{1}-\Pi_I)}{\mathrm{tr}((\mathbb{1}-\Pi_I)e^{-H_B/T}(\mathbb{1}-\Pi_I))}\Big),$$

where we used again that $\Pi_{I'}(\mathbb{1}\otimes(\mathbb{1}-\Pi_I)) = 0 = (\mathbb{1}-\Pi_{I'})(\mathbb{1}\otimes\Pi_I)$. However, now, the second-to-last channel (without the pre-factor) is indistinguishable from $\Phi_{T,|I|}(H_{B,1},U_1)$, and the same holds for the channel in the last line and $\Phi_{T,m-|I|}(H_{B,2},U_2)$. The reason for this is that $\Pi_I e^{-H_B/T}\Pi_I$ and $e^{-H_{B,1}/T}$ and $\Pi_I'U\Pi_I'$ and U_1 have the same non-zero entries in the same "order;" hence, everything else in $\Pi_I e^{-H_B/T}\Pi_I$, $\Pi_I'U\Pi_I'$ can be disregarded without changing the map. More precisely, one may use the decompositions

$$H_{B,1} = \mathrm{diag}\,(\langle e_i, H_B e_i\rangle)_{i\in I} = \sum_{j=1}^{|I|}\langle e_{\iota(j)}, H_B e_{\iota(j)}\rangle |e_j\rangle\langle e_j|$$

and

$$U_1 = \sum_{a,b=1}^{n}\sum_{i,j=1}^{|I|}\langle e_a\otimes e_{\iota(i)}, U(e_b\otimes e_{\iota(j)})\rangle |e_a\rangle\langle e_b|\otimes|e_i\rangle\langle e_j|$$

when enumerating $I = \{\iota(1),\dots,\iota(|I|)\}$, $\iota(1)<\dots<\iota(|I|)$, so $\iota:\{1,\dots,|I|\}\to I$ is bijective and order-preserving. The same is done for $H_{B,2}, U_2$. In total, this proves (A4) by means of a direct computation. The proof is concluded by the observation that U_1, U_2 are unitary because $\mathbf{\Pi}_{I'}U\mathbf{\Pi}_{I'}$, $(\mathbb{1}-\Pi_{I'})U(\mathbb{1}-\Pi_{I'})$ are partial isometries due to the "block-form" of U, as well as the fact that $\Pi_{I'}[U, H_S\otimes\mathbb{1}+\mathbb{1}\otimes H_B]\Pi_{I'} = [\Pi_{I'}U\Pi_{I'}, H_S\otimes\mathbb{1}+\mathbb{1}\otimes H_B]$ has the same non-zero entries as $[U_1, H_S\otimes\mathbb{1}+\mathbb{1}\otimes H_{B,1}]$. However, the former is zero due to energy conservation, meaning that U_1 is energy-conserving with respect to $H_{B,1}$ (similarly for $U_2, H_{B,2}$). ◻

APPENDIX B: PROOF OF COROLLARY 5

W.l.o.g., $H_S = \bigoplus_{j=0}^{n-1}(E_1 + j\Delta E)\,\mathbb{1}_{\alpha_j}$ for some $E_1\in\mathbb{R}$, $\Delta E>0$, and $\alpha_0, n\in\mathbb{N}$, $\alpha_1,\dots,\alpha_{n-1}\in\mathbb{N}_0$; the rest is just a basis change, which Lemma 2 takes care of.

All we have to prove is identity (5) for $T\in(0,\infty)$ (as the case $T=\infty$ is done analogously) because the second statement of the proposition is a special case of Proposition 4 (iv): if something has a resonant spectrum with respect to a spin Hamiltonian (with gaps), then it has to be of the same "spin form." Indeed, if $H_B\in i\mathfrak{u}(m)$ is resonant with respect to the above H_S, then the difference of any pair of eigenvalues of H_B is a multiple of ΔE, so there exist $\alpha_1,\dots,\alpha_m\in\mathbb{N}_0$, $c\in\mathbb{R}$ such that $\sigma(H_B) = \{\alpha_j\cdot\Delta E : j=1,\dots,m\}+c$ (the global shift c can be disregarded as such a shift does not change the corresponding thermal operation). We will show this via contraposition: Assume that $H_B\in i\mathfrak{u}(m)$ has two eigenvalues E_a', E_b', the difference of which is not a multiple of ΔE. Then, the set $I := \{1,\dots,m : \exists_{\alpha\in\mathbb{Z}}\ E_i'-E_b' = \alpha\Delta E\}$ is a proper $(a\notin I)$ non-empty $(b\in I)$ subset of $\{1,\dots,m\}$. However, by definition of I, for all $i\in I$, $j\in\{1,\dots,m\}\backslash I$, one finds that

$$E_i'-E_j' = \underbrace{E_i'-E_b'}_{\text{multiple of }\Delta E} + \underbrace{E_b'-E_j'}_{\text{not a multiple of }\Delta E}$$

is not a multiple of ΔE. Hence, $E_i'-E_j'$ cannot be an element of $\sigma(\mathrm{ad}_{H_S})$, which shows that H_B is not resonant with respect to H_S.

As for the first equation in (5): while "⊆" is obvious, for "⊇," we have to show that it is possible to approximate any thermal operation with bath Hamiltonian $H_B = \bigoplus_{j=1}^m j\Delta E\,\mathbb{1}_{\beta_j}$ (where some of β_j can be zero) using a Hamiltonian where $\beta_j\geq 1$ for all j. Given arbitrary $\alpha\in\mathbb{N}$, define $J := \{j\in\{1,\dots,m\} : \beta_j = 0\}$ and $H_{B,\alpha}' := \underline{\tau_\alpha}((\oplus_{k=1}^\alpha H_B)\oplus\mathrm{diag}\,(j\Delta E)_{j\in J})\underline{\tau_\alpha^{-1}}$, where τ_α is any permutation[43] such that the diagonal of $H_{B,\alpha}'$ is sorted increasingly; thus, $H_{B,\alpha}'$ is of the required form. The total Hamiltonian will be decomposed into $n\times n$ blocks[44] of equal size $(\sum_{j=1}^m\beta_j)\times(\sum_{j=1}^m\beta_j)$, that is, $H_{\mathtt{tot}} = \sum_{i,j=1}^n |e_i\rangle\langle e_j|\otimes(H_{\mathtt{tot}})_{ij}$. With this, we define

$$H_{\mathtt{tot},\alpha}' := \sum_{i,j=1}^n |e_i\rangle\langle e_j|\otimes\underline{\tau_\alpha}\Big(\Big(\bigoplus_{k=1}^\alpha(H_{\mathtt{tot}})_{ij}\Big)\oplus 0_{|J|}\Big)\underline{\tau_\alpha^{-1}}$$

and claim that the thermal operations generated by $H_B, H_{\mathtt{tot}}$ and by $H'_{B,\alpha}, H'_{\mathtt{tot},\alpha}$, respectively, coincide in the limit $\alpha \to \infty$. To see that the latter actually generates a thermal operation, one readily verifies

$$\big([H'_{\mathtt{tot},\alpha}, H_S \otimes \mathbb{1} + \mathbb{1} \otimes H'_{B,\alpha}]\big)_{ij} = \underline{\tau_\alpha}\bigg(\bigoplus_{k=1}^{\alpha}([H_{\mathtt{tot}}, H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_B])_{ij} \oplus 0_{|J|}\bigg)\underline{\tau_\alpha^{-1}}$$

for all $i, j = 1, \ldots, n$, so $[H'_{\mathtt{tot},\alpha}, H_S \otimes \mathbb{1} + \mathbb{1} \otimes H'_{B,\alpha}] = 0$ is equivalent to $[H_{\mathtt{tot}}, H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_B] = 0$. Moreover,

$$\begin{aligned}
S_\alpha &:= \operatorname{tr}_{B'}\left(e^{iH'_{\mathtt{tot},\alpha}}\left((\cdot) \otimes \frac{e^{-H'_{B,\alpha}/T}}{\operatorname{tr}(e^{-H'_{B,\alpha}/T})}\right)e^{-iH'_{\mathtt{tot},\alpha}}\right) \\
&= \frac{\sum_{i,j,k,l=1}^n \operatorname{tr}_{B'}\big((|e_i\rangle\langle e_j| \otimes (e^{iH'_{\mathtt{tot},\alpha}})_{ij})((\cdot) \otimes e^{-H'_{B,\alpha}/T})(|e_k\rangle\langle e_l| \otimes (e^{iH'_{\mathtt{tot},\alpha}})_{kl})\big)}{\alpha\big(\sum_{j=1}^m e^{-j\Delta E/T}\beta_j\big) + \sum_{j\in J} e^{-j\Delta E/T}} \\
&= \frac{\sum_{i,j,k,l=1}^n \langle e_j, (\cdot)e_k\rangle \operatorname{tr}\big((e^{iH'_{\mathtt{tot},\alpha}})_{ij} e^{-H'_{B,\alpha}/T}(e^{iH'_{\mathtt{tot},\alpha}})_{kl}\big)|e_i\rangle\langle e_l|}{\alpha\big(\sum_{j=1}^m e^{-j\Delta E/T}\beta_j\big) + \sum_{j\in J} e^{-j\Delta E/T}} \\
&= \frac{\sum_{i,j,k,l=1}^n \langle e_j, (\cdot)e_k\rangle \Big(\alpha \operatorname{tr}\Big((e^{iH_{\mathtt{tot}}})_{ij} e^{-H_B/T}(e^{-iH_{\mathtt{tot}}})_{kl} + \sum_{j\in J} e^{-j\Delta E/T}\Big)|e_i\rangle\langle e_l|}{\alpha\big(\sum_{j=1}^m e^{-j\Delta E/T}\beta_j\big) + \sum_{j\in J} e^{-j\Delta E/T}},
\end{aligned}$$

which in the limit $\alpha \to \infty$ yields

$$\lim_{\alpha\to\infty} S_\alpha = \frac{\sum_{i,j,k,l=1}^n \langle e_j, (\cdot)e_k\rangle \operatorname{tr}((e^{iH_{\mathtt{tot}}})_{ij} e^{-H_B/T}(e^{-iH_{\mathtt{tot}}})_{kl})|e_i\rangle\langle e_l|}{\sum_{j=1}^m e^{-j\Delta E/T}\beta_j} = \cdots = \operatorname{tr}_B\left(e^{iH_{\mathtt{tot}}}\left((\cdot) \otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})}\right)e^{-iH_{\mathtt{tot}}}\right)$$

as claimed. Here, we used that $(e^{iH'_{\mathtt{tot},\alpha}})_{ij} = \underline{\tau_\alpha}((\oplus_{k=1}^{\alpha}(e^{iH_{\mathtt{tot}}})_{ij}) \oplus \mathbb{1}_{|J|})\underline{\tau_\alpha^{-1}}$, which follows from the identity $(H'_{\mathtt{tot},\alpha})^l_{ij} = \underline{\tau_\alpha}((\oplus_{k=1}^{\alpha}(H_{\mathtt{tot}})^l_{ij}) \oplus 0^l_{|J|})\underline{\tau_\alpha^{-1}}$ for all $l \in \mathbb{N}_0$. The latter is readily verified by means of the block structure of $H'_{\mathtt{tot},\alpha}$. □

APPENDIX C: ATTEMPTED PROOF OF CONJECTURE 8

It would suffice to find a map $c : (0, \infty] \times (0, \infty] \to [0, \infty)$ such that the following holds:

- $c(T, T) = 0$ for all $T \in (0, \infty]$.
- c is continuous with respect to d_{-1}.
- Given any $\varepsilon > 0$, $T, T' \in (0, \infty]$, there, for all $S \in \overline{\mathsf{TO}(H_S, T)}$ exists $S' \in \overline{\mathsf{TO}(H_S, T')}$ such that $\|S - S'\|_{1\to1} < \varepsilon + c(T, T')$ (and vice versa).

  Then, by definition of the Hausdorff metric, $\delta\big(\overline{\mathsf{TO}(H_S, T)}, \overline{\mathsf{TO}(H_S, T')}\big) \le c(T, T')$, which for all $T \in (0, \infty]$, would imply

$$\delta\big(\overline{\mathsf{TO}(H_S, T)}, \overline{\mathsf{TO}(H_S, T')}\big) \to 0 \qquad \text{as} \qquad d_{-1}(T, T') \to 0$$

as desired. Now, given $\varepsilon > 0$, $S \in \overline{\mathsf{TO}(H_S, T)}$, there exist $H_B \in i\mathfrak{u}(m)$ and $U \in \mathsf{U}(mn)$ such that

$$\left\| S - \operatorname{tr}_B\left(U\left((\cdot) \otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})}\right)U^*\right)\right\|_{1\to1} < \varepsilon,$$

where U satisfies $U(H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B)U^* = H_S \otimes \mathbb{1}_B + \mathbb{1} \otimes H_B$. The simplest way of picking an element in $\mathsf{TO}(H_S, T')$, which is "close to" this approximation of S, is to define the channel $S' := \operatorname{tr}_B(U((\cdot) \otimes \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})})U^*)$. This yields the estimate

$$
\begin{aligned}
\|S - S'\|_{1\to 1} &\leq \left\| S - \operatorname{tr}_B\left(U\left((\cdot) \otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} \right) U^* \right) \right\|_{1\to 1} \\
&\quad + \left\| \operatorname{tr}_B\left(U\left((\cdot) \otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} \right) U^* \right) - \operatorname{tr}_B\left(U\left((\cdot) \otimes \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})} \right) U^* \right) \right\|_{1\to 1} \\
&< \varepsilon + \left\| \operatorname{tr}_B\left(U\left((\cdot) \otimes \left(\frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} - \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})} \right) U^* \right) \right\|_{1\to 1} \\
&< \varepsilon + \|\operatorname{tr}_B\|_{1\to 1} \|\mathrm{Ad}_U\|_{1\to 1} \left\| \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} - \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})} \right\|_1 .
\end{aligned} \tag{C1}
$$

Keeping in mind that tr_B, Ad_U have operator norm one (with respect to the trace norm) because they are (completely) positive and trace-preserving (Theorem 2.1 in Ref. 42), it seems reasonable to consider

$$
\begin{aligned}
c : (\,0,\infty\,] \times (\,0,\infty\,] &\to [0,\infty) \\
(T, T') &\mapsto \sup_{m\in\mathbb{N}} \sup_{H_B \in i\mathfrak{u}(m)} \left\| \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} - \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})} \right\|_1
\end{aligned} \tag{C2}
$$

for an upper bound. In other words, the above estimate *would* reduce the problem of continuity of the thermal operations to the continuity of certain Gibbs states in the temperature. However, (C2) already looks unsuited for the task as it does not feature the system's Hamiltonian anymore. Indeed, as soon as T, T' do not coincide, then $c(T, T')$ takes the largest possible value.

Lemma 11. For all $T, T' \in (\,0,\infty\,]$, one has $c(T,T') = 2\delta_{T,T'}$.

Proof. W.l.o.g., $T < T'$, so one has $e^{-E/T} < e^{-E/T'}$ for all $E > 0$. Now, given any $E > 0$, define the Hamiltonian $H_B(E) := 0 \oplus (E \cdot \mathbb{1}_{\lfloor e^{E/2T} e^{E/2T'} \rfloor}) \in i\mathfrak{u}(1 + \lfloor e^{E/2T} e^{E/2T'} \rfloor)$. We claim that

$$
\lim_{E\to\infty} \left(2 - \left\| \frac{e^{-H_B(E)/T}}{\operatorname{tr}(e^{-H_B(E)/T})} - \frac{e^{-H_B(E)/T'}}{\operatorname{tr}(e^{-H_B(E)/T'})} \right\|_1 \right) = 0. \tag{C3}
$$

Indeed, a straightforward computation shows

$$
\begin{aligned}
2 - \left\| \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} - \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})} \right\|_1 &= 2\left(1 - \frac{\lfloor e^{E/2T} e^{E/2T'} \rfloor (e^{-E/T'} - e^{-E/T})}{(1 + \lfloor e^{E/2T} e^{E/2T'} \rfloor e^{-E/T})(1 + \lfloor e^{E/2T} e^{E/2T'} \rfloor e^{-E/T'})} \right) \\
&= 2\left(\frac{1}{1 + \lfloor e^{E/2T} e^{E/2T'} \rfloor e^{-E/T'}} + \frac{1}{\lfloor e^{E/2T} e^{E/2T'} \rfloor^{-1} e^{E/T} + 1} \right) \\
&\leq 2\left(\frac{1}{1 + e^{E/2T} e^{-E/2T'} - e^{-E/T'}} + \frac{1}{e^{E/2T} e^{-E/2T'} + 1} \right).
\end{aligned}
$$

However, $e^{E/2T} e^{-E/2T'} = e^{\frac{E}{2}(\frac{1}{T} - \frac{1}{T'})} \to \infty$ as $E \to \infty$ because $\frac{1}{T} - \frac{1}{T'} > 0$ by assumption. Moreover, the expression in (C3) is non-negative (by the triangle inequality), so because the upper bound we found vanishes as $E \to \infty$, (C3) holds. This concludes the proof. $\square$

There are two ways out of this dilemma: On the one hand, one could restrict the supremum in (C2) to a smaller generating set of the thermal operations (respectively, its closure), for example, $\mathbf{H} := \bigcup_{m\in\mathbb{N}} \{H_B \in i\mathfrak{u}(m) : H_B \text{ is resonant with respect to } H_S\}$. This would invalidate the current Proof of Lemma 11, the key to which was to let the gaps between neighboring eigenvalues of H_B become arbitrarily large—but the resonance condition prohibits this. However, modifying $c(T,T')$ to be $\sup_{H_B\in\mathbf{H}} \| \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})} - \frac{e^{-H_B/T'}}{\operatorname{tr}(e^{-H_B/T'})} \|_1$—while seeming more suited to be an upper bound as H_S now appears at least implicitly in c—does make it more difficult to study c due to the more complicated structure of $\mathbf{H}$. On the other hand, (C1) might be a too poor estimate for studying the continuity of $T \mapsto \overline{\mathsf{TO}(H,T)}$: Although T only ever appears in the Gibbs state, it may be that separating the fundamental building blocks of the thermal operations—as done in (C1)—loses too much of its structure, even if one is only interested in the effect of the temperature. Either way, it seems that proving Conjecture 8—if true at all—requires a more careful analysis of the effect, which changing the temperature can have on the set of thermal operations.

APPENDIX D: PROOF OF THEOREM 10

The following lemma, which is indispensable for qubit computations, is verified directly.

Lemma 12. Let $T \in (0, \infty]$, $\Delta E > 0$, $m \in \mathbb{N}$, *and* $\alpha_0, \ldots, \alpha_{m-1} \in \mathbb{N}$, *as well as unitaries* $U_0 \in \mathbb{C}^{\alpha_0 \times \alpha_0}$, $U_m \in \mathbb{C}^{\alpha_{m-1} \times \alpha_{m-1}}$, *and* $U_j \in \mathbb{C}^{(\alpha_{j-1}+\alpha_j)\times(\alpha_{j-1}+\alpha_j)}$, $j = 1, \ldots, m-1$, *be given. Decompose*

$$U_j = \begin{pmatrix} A_j & B_j \\ C_j & D_j \end{pmatrix} \tag{D1}$$

with $A_j \in \mathbb{C}^{\alpha_j \times \alpha_j}, B_j \in \mathbb{C}^{\alpha_j \times \alpha_{j-1}}, C_j \in \mathbb{C}^{\alpha_{j-1}\times\alpha_j}, D_j \in \mathbb{C}^{\alpha_{j-1}\times\alpha_{j-1}}$ *for all* $j = 1, \ldots, m-1$. *Defining* $H_B := \bigoplus_{j=0}^{m-1} j\Delta E \cdot \mathbb{1}_{\alpha_j}$,

$$U := \begin{pmatrix} U_0 & 0 & 0 & 0 \\ 0 & \bigoplus_{j=1}^{m-1} A_j & \bigoplus_{j=1}^{m-1} B_j & 0 \\ 0 & \bigoplus_{j=1}^{m-1} C_j & \bigoplus_{j=1}^{m-1} D_j & 0 \\ 0 & 0 & 0 & U_m \end{pmatrix} \in \mathbb{C}^{(2\alpha_0+\cdots+2\alpha_{m-1})\times(2\alpha_0+\cdots+2\alpha_{m-1})},$$

and $S_U := \operatorname{tr}_B\big(U((\cdot)\otimes \frac{e^{-H_B/T}}{\operatorname{tr}(e^{-H_B/T})})U^*\big)$ *[respectively,* $S_U := \operatorname{tr}_B\big(U((\cdot)\otimes \frac{\mathbb{1}}{\sum_{j=0}^{m-1}\alpha_j})U^*\big)$ *if* $T = \infty$*], one finds that* U *is unitary,* $S_U \in \mathsf{TO}(\operatorname{diag}(0,1), T)$*, and the Choi matrix of* S_U *reads*

$$\begin{pmatrix} 1-\lambda e^{-\Delta E/T} & 0 & 0 & c \\ 0 & \lambda e^{-\Delta E/T} & 0 & 0 \\ 0 & 0 & \lambda & 0 \\ c^* & 0 & 0 & 1-\lambda \end{pmatrix} \tag{D2}$$

with

$$\lambda = \frac{\sum_{j=0}^{m-2}\operatorname{tr}(B_{j+1}B_{j+1}^*)e^{-j\Delta E/T}}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}},$$

$$c = \frac{\operatorname{tr}(U_0 D_1^*) + \sum_{j=1}^{m-2}\operatorname{tr}(A_j D_{j+1}^*)e^{-j\Delta E/T} + \operatorname{tr}(A_{m-1}U_m^*)e^{-(m-1)\Delta E/T}}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}}.$$

If $T = \infty$, *then Eq. (D2) and the succeeding formulas continue to hold if* $e^{-\Delta E/T}$ *is replaced by* 1.

With this, we are ready to prove Theorem 10.

Proof. (i): First, let us review how Ćwikliński *et al.* showed (in supplementary note 4, Sec. IV in Ref. 33) that $\mathsf{EnTO}(H_S, T) = \overline{\mathsf{TO}(H_S, T)}$ for all non-degenerate Hamiltonians[45,49] in two dimensions. This will allow us to highlight how one gets around using (highly) degenerate bath Hamiltonians [statements (ii) and (iii) of this theorem].

By Lemma 2, w.l.o.g., $H_S = \operatorname{diag}(0, \Delta E)$ with $\Delta E > 0$. Given $T \in (0, \infty)$ (we treat $T = \infty$ separately), $\lambda \in [0, 1]$, what they do is construct a family $(S_m^\mu)_{m\in\mathbb{N},\mu\in(1,e^{1/T})\cap\mathbb{Q}} \in \mathsf{TO}(H_S, T)$ such that

$$\Big(\lim_{\mu\to(e^{\Delta E/T})^-}\lim_{m\to\infty} S_m^\mu\Big)(A) = \begin{pmatrix} a_{11}(1-\lambda e^{-\Delta E/T}) + \lambda a_{22} & \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}\,a_{12} \\ \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}\,a_{21} & \lambda e^{-\Delta E/T}a_{11} + (1-\lambda)a_{22} \end{pmatrix}$$

for all $A \in \mathbb{C}^{2\times 2}$. What this means is that—together with the fact that the channel that only applies a phase to the off-diagonals is in TO ($m = 1$ in the definition)—the extreme points of EnTO (i.e., the boundary in Fig. 2 without the inner area of the circle at the bottom) are in $\overline{\mathsf{TO}}$.

From this, one can deduce that the two sets have to coincide: either one uses that EnTO, $\overline{\mathsf{TO}}$ are convex and compact, so

$$\mathsf{EnTO} = \mathrm{conv}\big(\mathrm{ext}(\mathsf{EnTO})\big) \subseteq \mathrm{conv}\Big(\mathrm{ext}\big(\overline{\mathsf{TO}}\big)\Big) = \overline{\mathsf{TO}} \subseteq \mathsf{EnTO}$$

by Minkowski's theorem (Theorem 5.10 in Ref. 46, where ext is the set of extreme points of a convex set), or one can show that any dephasing channel

$$A \mapsto \begin{pmatrix} a_{11} & \overline{\gamma} a_{12} \\ \gamma a_{21} & a_{22} \end{pmatrix} \tag{D3}$$

for $\gamma \in \mathbb{C}$, $|\gamma| \le 1$ is in TO because then every thermal operation can be written as a composition of an extreme point of EnTO and a dephasing channel: simply choose $U \in \mathsf{U}(2)$ such that $\mathrm{tr}(U) = 2\gamma$ because, then, $\Phi_{T,2}(\mathbb{1}_2, \mathbb{1}_2 \oplus U)$ is in $\mathsf{TO}(H_S, T)$ for all $T \in (0, \infty]$. Then, its action is precisely given by (D3); cf. also Chap. 8.3.6 in the work of Nielsen and Chuang.[47]

Now, the construction of the maps S_m^μ goes as follows: Given $T \in (0, \infty)$, $\mu \in (1, e^{\Delta E/T}) \cap \mathbb{Q}$, and $m \in \mathbb{N}$ define the following:

- $\alpha_0 = \alpha_0(m, \mu) \in \mathbb{N}$ is the smallest integer such that $\alpha_0 \mu^{m-1} \in \mathbb{N}$. The only role of α_0 is to ensure that the ratio of the size of consecutive blocks, which make up the unitary matrix, equals μ, thus approximating $e^{\Delta E/T}$. Indeed, α_0 will not appear in the explicit action of S_m^μ,
- $H_{B,m} := \bigoplus_{j=0}^{m-1} j\Delta E \cdot \mathbb{1}_{\alpha_0 \mu^j}$,
- $D_1 := \mathbb{1}_{\alpha_0}$ and, recursively, $A_j := D_j \oplus \mathbb{1}_{\alpha_0 \mu^{j-1}(\mu - 1)}$ for all $j = 1, \dots, m-1$ and $D_j := \sqrt{\frac{1-\lambda}{1-\frac{\lambda}{\mu}}} A_{j-1}$ for all $j = 2, \dots, m-1$.

  Because $\|A_j\|_\infty, \|D_j\|_\infty \le 1$ (where $\|\cdot\|_\infty$ is the usual operator norm, that is, the largest singular value), it is easy to see that for all $j = 1, \dots, m-1$, one can choose B_j, C_j such that

  $$U_j := \begin{pmatrix} A_j & B_j \\ C_j & D_j \end{pmatrix}$$

  is unitary, i.e., $U_j \in \mathsf{U}(\alpha_0 \mu^{j-1}(\mu + 1))$. With this, one defines

  $$U_m^\mu := \begin{pmatrix} \mathbb{1}_{\alpha_0} & 0 & 0 & 0 \\ 0 & \oplus_{j=1}^{m-1} A_j & \oplus_{j=1}^{m-1} B_j & 0 \\ 0 & \oplus_{j=1}^{m-1} C_j & \oplus_{j=1}^{m-1} D_j & 0 \\ 0 & 0 & 0 & \mathbb{1}_{\alpha_0 \mu^{m-1}} \end{pmatrix}$$

  and $S_m^\mu := \Phi_{T,(\alpha_0 \sum_{j=0}^{m-1} \mu^j)}(H_{B,m}, U_m^\mu)$. All one has to do now is compute the limit as stated above, which using the representation Ψ_T from Sec. IV comes out to be

  $$\lim_{m\to\infty} \Psi_T(S_m^\mu) = \begin{pmatrix} \dfrac{\lambda\mu(\mu-1)e^{-\Delta E/T}}{\mu - \lambda - \mu e^{-\Delta E/T}(1-\lambda)} \\ \sqrt{\mu(1-\lambda)(\mu-\lambda)}e^{-\Delta E/T} + (1 - \mu e^{-\Delta E/T})\left(1 + \dfrac{\sqrt{\mu(1-\lambda)(\mu-\lambda)}\lambda e^{-\Delta E/T}}{\mu - \lambda - \mu e^{-\Delta E/T}(1-\lambda)}\right) \end{pmatrix},$$

  so, as claimed,

  $$\lim_{\mu \to (e^{\Delta E/T})^-} \lim_{m\to\infty} \Psi_T(S_m^\mu) = \begin{pmatrix} \lambda \\ \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})} \end{pmatrix}.$$

  A particularly useful identity for verifying this is $\frac{\mu(1-\gamma^2)}{\mu-\gamma^2} = \lambda$ with $\gamma := \sqrt{\frac{1-\lambda}{1-\frac{\lambda}{\mu}}}$.

This construction breaks down once T is infinite for two reasons: first, the interval $(1, e^{\Delta E/T})$ from which we pick the rational approximation μ becomes empty, and more importantly, even if we just set $\mu = 1$, then $U_m^\mu = U_m^1 = \mathbb{1}$ for all m; thus, the corresponding thermal operation becomes trivial. This is why we have to treat the case $T = \infty$ separately. Indeed, given any $\lambda \in [0,1]$, $\phi \in [0,2\pi)$, choose $H_{B,m} := \operatorname{diag}(j\Delta E)_{j=0}^{m-1}$ and

$$U_m^\phi := \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \sqrt{1-\lambda}\mathbb{1}_{m-1} & \sqrt{\lambda}\mathbb{1}_{m-1} & 0 \\ 0 & -\sqrt{\lambda}e^{-i\phi}\mathbb{1}_{m-1} & \sqrt{1-\lambda}e^{-i\phi}\mathbb{1}_{m-1} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \tag{D4}$$

Obviously, U_m^ϕ is energy-preserving with respect to $(H_S, H_{B,m})$ for all $m \in \mathbb{N}$, and

$$\lim_{m\to\infty} \Psi_T(\Phi_{1,m}(\mathbb{1}, U_m^\phi)) = \lim_{m\to\infty} \begin{pmatrix} \dfrac{\lambda(m-1)}{m} \\ (1-\lambda)e^{i\phi} + \dfrac{\sqrt{1-\lambda}(1+e^{i\phi}(1-2\sqrt{1-\lambda}))}{m} \end{pmatrix} = \begin{pmatrix} \lambda \\ (1-\lambda)e^{i\phi} \end{pmatrix}. \tag{D5}$$

Thus, $\Psi_T^{-1}(\lambda, (1-\lambda)e^{i\phi}) \in \overline{\mathsf{TO}(H_S, \infty)}$ for all $\phi \in [0,2\pi)$ as desired.

(ii): We only have to prove (10) because, then, the convexity of $\mathsf{TO}(H_S,T)$ follows directly. First, let us see that $\mathsf{TO}(H_S,T)$ is a subset of (10). For this, we present a slight modification of the proof of Theorem 1 from Ref. 20: The idea is to find a family of subsets $(\mathscr{S}_m)_m$ of $\mathsf{TO}(H_S,T)$ such that in the limit $m \to \infty$, their convex hull $(\mathsf{conv}(\mathscr{S}_m))_m$ exhausts the cone of enhanced thermal operations from Fig. 2. The exact form of $\mathscr{S}_m$ will let us conclude that for every $S \in \mathsf{EnTO}(H_S,T)$ *not* on the boundary, there exist m and $S_m \in \mathscr{S}_m$ such that S is the composition of S_m and a partial dephasing map. Hence, $S \in \mathsf{TO}(H_S,T)$ as it is the composition of two thermal operations [Proposition 4 (i)].

Now, for the details, given $T \in (0,\infty)$ (a note on the case $T = \infty$ later), $\lambda \in [0,1]$, $m \in \mathbb{N}\backslash\{1\}$, $\phi \in [-\pi,\pi)$ define a thermal operation as follows: $H_{B,m} := \operatorname{diag}(0, \Delta E, \dots, (m-1)\Delta E) \in i\mathfrak{u}(m)$ is the bath Hamiltonian, and the energy-preserving unitary $U_m^{\lambda,\phi} \in \mathsf{U}(2m)$ is given by

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \operatorname{diag}\left(\left(\dfrac{1-\lambda}{1-\lambda e^{-\Delta E/T}}\right)^{j/2}\right)_{j=1}^{m-1} & \operatorname{diag}\left(i\left(1-\left(\dfrac{1-\lambda}{1-\lambda e^{-\Delta E/T}}\right)^{j}\right)^{1/2}\right)_{j=1}^{m-1} & 0 \\ 0 & \operatorname{diag}\left(ie^{-i\phi}\left(1-\left(\dfrac{1-\lambda}{1-\lambda e^{-\Delta E/T}}\right)^{j}\right)^{1/2}\right)_{j=1}^{m-1} & \operatorname{diag}\left(e^{-i\phi}\left(\dfrac{1-\lambda}{1-\lambda e^{-\Delta E/T}}\right)^{j/2}\right)_{j=1}^{m-1} & 0 \\ 0 & 0 & 0 & e^{-i\phi} \end{pmatrix}.$$

Be aware that a variation of this unitary has also appeared in Appendix B of Ref. 6 (cf. also references therein). However, the unitary matrix that Lostaglio *et al.* use leads to a cone that—while containing all classical channels $\{\Psi_T^{-1}(\lambda,0) : \lambda \in [0,1)\}$ (which was their goal)—is always a strict subset of $\mathsf{TO}(H_S,T)$, even in the closure.

Now, let us collect all maps with the same ϕ via $\mathscr{S}_{m,\phi} := \left\{\Phi_{T,m}(H_{B,m}, U_m^{\lambda,\phi}) : \lambda \in [0,1]\right\}$, so the set we are looking for which exhausts $\mathsf{EnTO}(H_S,T)$ in the convex hull as m goes to infinity is $\mathscr{S}_m := \bigcup_{\phi\in[-\pi,\pi)} \mathscr{S}_{m,\phi}$.

Claim: For any $m \in \mathbb{N}$, $\phi \in [-\pi,\pi)$, applying Ψ_T to the set $\mathscr{S}_{m,\phi}$ yields a strictly convex curve with end points,

$$\begin{pmatrix} 0 \\ e^{i\phi} \end{pmatrix} (\text{ for } \lambda = 0) \qquad \text{and} \qquad \begin{pmatrix} \dfrac{1-(e^{-\Delta E/T})^{m-1}}{1-(e^{-\Delta E/T})^m} \\ 0 \end{pmatrix} (\text{ for } \lambda = 1), \tag{D6}$$

and $\Psi_T(\mathscr{S}_{m,\phi})$ converges to $\{(\lambda, e^{i\phi}\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}) : \lambda \in [0,1]\}$ in the Hausdorff metric. This follows from a direct computation using Lemma 12,

$$\Psi_T\left(\Phi_{T,m}(H_{B,m}, U_m^{\lambda,\phi})\right) = \begin{pmatrix} \dfrac{1-(e^{-\Delta E/T})^{m-1}}{1-(e^{-\Delta E/T})^m} - \gamma^2 \dfrac{1-e^{-\Delta E/T}}{1-\gamma^2 e^{-\Delta E/T}} \dfrac{1-(\gamma^2 e^{-\Delta E/T})^{m-1}}{1-(e^{-\Delta E/T})^m} \\ e^{i\phi}(1-e^{-\Delta E/T})\left(\dfrac{\gamma}{1-\gamma^2 e^{-\Delta E/T}} \dfrac{1-(\gamma^2 e^{-\Delta E/T})^{m-1}}{1-(e^{-\Delta E/T})^m} - \dfrac{(\gamma e^{-\Delta E/T})^{m-1}}{1-(e^{-\Delta E/T})^m}\right) \end{pmatrix},$$

where $\gamma := \sqrt{\frac{1-\lambda}{1-\lambda e^{-\Delta E/T}}}$. Note that γ is strictly monotonically decreasing in λ and $\gamma|_{\lambda=0} = 1$, $\gamma|_{\lambda=1} = 0$; hence, γ is bijective on $[0, 1]$ as a function of λ. In particular, setting $\lambda \in \{0,1\}$ ($\gamma \in \{0,1\}$) reproduces (D6). Taking the limit $m \to \infty$ yields

$$\lim_{m\to\infty} \Psi_T\Big(\Phi_{T,m}(H_{B,m}, U_m^{\lambda,\phi})\Big) = \begin{pmatrix} 1-\gamma^2 \dfrac{1-e^{-\Delta E/T}}{1-\gamma^2 e^{-\Delta E/T}} \\ e^{-i\phi} \dfrac{1-e^{-\Delta E/T}}{1-\gamma^2 e^{-\Delta E/T}}\gamma \end{pmatrix} = \begin{pmatrix} 1-\gamma^2 \dfrac{1-\lambda}{\gamma^2} \\ e^{i\phi} \dfrac{1-\lambda}{\gamma^2}\gamma \end{pmatrix} = \begin{pmatrix} \lambda \\ e^{i\phi}\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})} \end{pmatrix}$$

as claimed. Here, we used the readily verified identity $\frac{1-e^{-\Delta E/T}}{1-\gamma^2 e^{-\Delta E/T}} = \frac{1-\lambda}{\gamma^2}$. Note that the case $T = \infty$ is proven analogously once the unitary $U_m^{\lambda,\phi}$ is given by (D4) [as the computation in (D5) shows].

Now, let $\lambda \in (0,1)$, $r \in [0, \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})})$, and $\phi \in [-\pi,\pi)$ be given, that is, $(\lambda, re^{i\phi})$ does not lie on the relative boundary of $\Psi_T(\mathsf{EnTO}(H_S,T))$ [we can exclude the case $\lambda = 0$ as we already know that all partial dephasings are elements of TO; cf. (D3)]. Because $\mathrm{conv}(\mathscr{S}_m)$ is strictly monotonically increasing in m and because $\lim_{m\to\infty}\delta(\mathrm{conv}(\mathscr{S}_m), \mathsf{EnTO}(H_S,T)) = 0$, there exists $m \in \mathbb{N}$ such that $\Psi_T^{-1}(\lambda, re^{i\phi}) \in \mathrm{conv}(\mathscr{S}_m)$. However, by construction of $\mathscr{S}_m$, this means that $\Psi_T^{-1}(\lambda, r'e^{i\phi}) \in \mathscr{S}_m$ for some $r' \geq r$, so

$$\begin{aligned} \Psi_T^{-1}\begin{pmatrix} \lambda \\ re^{i\phi} \end{pmatrix} = \Psi_T^{-1}\begin{pmatrix} 0 \\ \frac{r}{r'} \end{pmatrix} \circ \Psi_T^{-1}\begin{pmatrix} \lambda \\ r'e^{i\phi} \end{pmatrix} &\in \mathsf{TO}(H_S,T) \circ \mathscr{S}_m \\ &\subseteq \mathsf{TO}(H_S,T) \circ \mathsf{TO}(H_S,T) = \mathsf{TO}(H_S,T). \end{aligned}$$

Here, we used again that all partial dephasings are thermal operations [(D3), as $\frac{r}{r'} < 1$].

Conversely, to see that (10) is a subset of $\mathsf{TO}(H_S,T)$, we have to show that

$$\Psi_T^{-1}\begin{pmatrix} \lambda \\ e^{i\phi}\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})} \end{pmatrix} \notin \mathsf{TO}(H_S,T) \tag{D7}$$

for all $\lambda \in (0,1]$, $\phi \in [-\pi,\pi)$, and $T \in (0,\infty)$ [proving (D7) for $T = \infty$ is done analogously].

Assume to the contrary that (D7) is false. Hence, there exist $m \in \mathbb{N}$ and $\alpha_0,\ldots,\alpha_{m-1} \in \mathbb{N}$ such that $\Phi_{T,m}(H_B,U) = \Psi_T^{-1}(\lambda, e^{i\phi}\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})})$ for some energy-preserving unitary $U \in \mathsf{U}(2(\sum_{j=0}^{m-1}\alpha_j))$, where $H_B := \oplus_{j=0}^{m-1} j\Delta E \cdot \mathbb{1}_{\alpha_j}$. The reason for choosing resonant H_B is that $\Psi_T^{-1}(\lambda, e^{i\phi}\sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})})$ is an extreme point of $\mathsf{EnTO}(H_S,T)$, and the Proof of Proposition 4 (iv) [cf. (A4)] shows that any thermal operation with a bath Hamiltonian, which is *not* of this form, can be written as a convex combination of two thermal operations with bath Hamiltonians of the above form. However, this contradicts the extreme point property, so H_B has to have a resonant spectrum with respect to H_S.

Due to H_B being of the spin form, we may apply Lemma 12 to get an explicit form of U and, more importantly, $\Psi_T(\Phi_{T,m}(H_B,U))$. Define an inner product $\langle\cdot,\cdot\rangle_T$ on $\mathbb{C}^{\alpha_0\times\alpha_0} \times \cdots \times \mathbb{C}^{\alpha_{m-1}\times\alpha_{m-1}}$ via

$$\left(\begin{pmatrix} X_1 \\ \vdots \\ X_m \end{pmatrix}, \begin{pmatrix} Y_1 \\ \vdots \\ Y_m \end{pmatrix}\right) \mapsto \sum_{j=0}^{m-1} \langle X_j, Y_j\rangle_{\mathsf{HS}}\, e^{-j\Delta E/T},$$

where $\langle A,B\rangle_{\mathsf{HS}} = \mathrm{tr}(A^*B)$ is the Hilbert–Schmidt inner product on complex square matrices of any dimension. Note that $\langle\cdot,\cdot\rangle_T$ is, indeed, an inner product because it is a sum of inner products with positive weights. This lets us rewrite c from Lemma 12 as

$$c = \frac{\langle (D_1,\ldots,D_{m-1},U_m),(U_0,A_1,\ldots,A_{m-1})\rangle_T}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}}.$$

In particular, we can apply the Cauchy–Schwarz inequality to obtain

$$|c| \le \frac{\|(D_1,\ldots,D_{m-1},U_m)\|_T \|(U_0,A_1,\ldots,A_{m-1})\|_T}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}}$$
$$= \frac{\sqrt{\Big(\sum_{j=0}^{m-2}\|D_{j+1}\|_{\mathrm{HS}}^2 e^{-j\Delta E/T} + \|U_m\|_{\mathrm{HS}}^2 e^{-\frac{(m-1)\Delta E}{T}}\Big)\Big(\|U_0\|_{\mathrm{HS}}^2 + \sum_{j=1}^{m-1}\|A_j\|_{\mathrm{HS}}^2 e^{-j\Delta E/T}\Big)}}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}}.$$

Now, unitarity of U comes into play: On the one hand, U_0, U_m are itself unitary, so $\|U_0\|_{\mathrm{HS}}^2 = \alpha_0$, $\|U_{m-1}\|_{\mathrm{HS}}^2 = \alpha_{m-1}$. Moreover, unitarity of the blocks U is made up of [i.e., (D1) being unitary] implies $A_jA_j^* + B_jB_j^* = \mathbb{1}_{\alpha_j}$ and $B_j^*B_j + D_j^*D_j = \mathbb{1}_{\alpha_{j-1}}$ for all $j = 1,\ldots,m-1$. Taking the trace yields

$$\begin{aligned} \|D_{j+1}\|_{\mathrm{HS}}^2 &= \alpha_j - \|B_{j+1}\|_{\mathrm{HS}}^2 \qquad &&\text{for all } j = 0,\ldots,m-2, \text{ and} \\ \|A_j\|_{\mathrm{HS}}^2 &= \alpha_j - \|B_j\|_{\mathrm{HS}}^2 \qquad &&\text{for all } j = 1,\ldots,m-1. \end{aligned}$$

With this, the upper bound we found for $|c|$ is equal to

$$\sqrt{\left(1 - \frac{\sum_{j=0}^{m-2}\mathrm{tr}(B_{j+1}B_{j+1}^*)e^{-j\Delta E/T}}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}}\right)\left(1 - \frac{\sum_{j=0}^{m-2}\mathrm{tr}(B_{j+1}B_{j+1}^*)e^{-j\Delta E/T}}{\sum_{j=0}^{m-1}\alpha_j e^{-j\Delta E/T}}e^{-\Delta E/T}\right)},$$

that is, $|c| \le \sqrt{(1-\lambda)(1-\lambda e^{-\Delta E/T})}$ with equality if and only if there is equality in the Cauchy–Schwarz inequality because that was the only estimate we used in our calculation. However, it is well known that equality in the Cauchy–Schwarz inequality is equivalent to the two arguments being a scalar multiple of each other. Hence, there exists $\xi \in \mathbb{C}$ such that

$$\begin{pmatrix} D_1 \\ \cdots \\ D_{m-1} \\ U_m \end{pmatrix} = \xi \begin{pmatrix} U_0 \\ A_1 \\ \cdots \\ A_{m-1} \end{pmatrix}. \tag{D8}$$

$\|U_0\|_\infty, \|U_m\|_\infty = 1$ and $\|D_j\|_\infty, \|A_j\|_\infty \le 1$ for all $j = 1,\ldots,m$. Therefore, $|\xi| = \|\xi U_0\|_\infty = \|D_1\|_\infty \le 1$ and $1 = \|U_m\|_\infty = |\xi|\|A_{m-1}\|_\infty \le |\xi|$, so $|\xi| = 1$. However, with this, (D8) forces all B_j to vanish: first, $D_1^*D_1 = |\xi|^2 U_0^*U_0 = \mathbb{1}$, so $B_1 = 0$, and thus, $A_1^*A_1 = \mathbb{1}$ because (D1) is unitary. Then, considering the second element of (D8) implies that $D_2^*D_2 = |\xi|^2 A_1^*A_1 = \mathbb{1}$, so $B_2 = 0$; repeating this argument inductively shows $B_j = 0$ for all $j = 1,\ldots,m-1$. However, this is problematic because, then, $\lambda = 0$ (again by Lemma 12), which contradicts our assumption that $\lambda \in (0,1]$.

(iii): This result is a truncated version of (iv) (i.e., of Theorem 1 from Ref. 20), so the proof we present is inspired by the arguments of Ding *et al.* We showed in (ii) that for qubits, every $S \in \mathrm{TO}(H_S,T)$ is the composition of a thermal operation generated by $H_{B,m} := \mathrm{diag}(0,\Delta E,\ldots,(m-1)\Delta E)$ for some $m \in \mathbb{N}$ and a (partial) dephasing. Hence, if $T > \frac{\Delta E}{\ln 2}$, it suffices to show that each partial dephasing can be implemented using some $H_{B,m}$.

For this, note that given $m \in \mathbb{N}$ and arbitrary phases $\phi_1,\ldots,\phi_m \in [-\pi,\pi)$, the unitary $U := \mathbb{1}_m \oplus U_\phi$ with $U_\phi := \mathrm{diag}(e^{i\phi_1},\ldots,e^{i\phi_m})$ is energy-preserving because it is diagonal. A straightforward computation yields

$$\Phi_{T,m}(H_{B,m},U_\phi)(A) = \begin{pmatrix} a_{11} & a_{12}\left(\frac{\mathrm{tr}(e^{-H_{B,m}/T}U_\phi)}{\mathrm{tr}(e^{-H_{B,m}/T})}\right)^* \\ a_{21}\frac{\mathrm{tr}(e^{-H_{B,m}/T}U_\phi)}{\mathrm{tr}(e^{-H_{B,m}/T})} & a_{22} \end{pmatrix}$$

for all $A \in \mathbb{C}^{2\times 2}$. Thus, all we have to see is that for m "large enough," the map $(\phi_1,\ldots,\phi_m) \mapsto \frac{\mathrm{tr}(e^{-H_{B,m}/T}U_\phi)}{\mathrm{tr}(e^{-H_{B,m}/T})}$ maps surjectively onto the closed unit disk. The key observation here is that given numbers $0 < c_2 < c_1$, the (pointwise) sum of a circle with radius c_2 to a circle with radius c_1 both centered around the origin (i.e., $\{c_1e^{i\phi_1} + c_2e^{i\phi_2} : \phi_1,\phi_2 \in [-\pi,\pi)\}$) is equal to the annulus $\{re^{i\phi} : c_1 - c_2 \le r \le c_1 + c_2, \phi \in [-\pi,\pi)\}$ with inner radius $c_1 - c_2$ and outer radius $c_1 + c_2$. We visualize this fact in Fig. 5, which makes a proof superfluous.

This implies that the expression

$$\frac{\mathrm{tr}(e^{-H_{B,m}/T}U_\phi)}{\mathrm{tr}(e^{-H_{B,m}/T})} = \frac{\sum_{j=0}^{m-1}e^{-j\Delta E/T}e^{i\phi_j}}{\sum_{k=0}^{m-1}e^{-k\Delta E/T}} = \sum_{j=0}^{m-1}\frac{(1-e^{-\Delta E/T})e^{-j\Delta E/T}}{1-e^{-m\Delta E/T}}e^{i\phi_j}$$

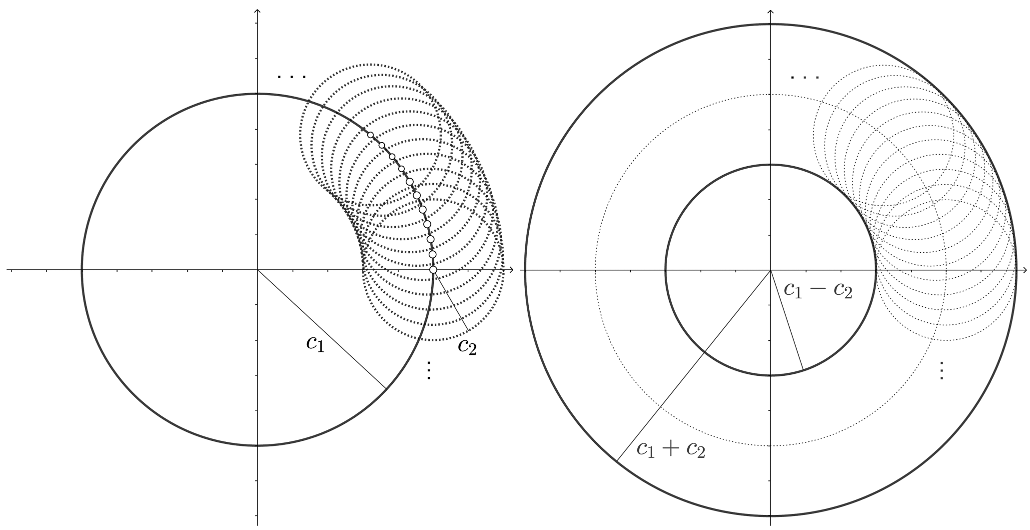

FIG. 5. Visual proof of the equality of $\{c_1 e^{i\phi_1} + c_2 e^{i\phi_2} : \phi_1, \phi_2 \in [-\pi, \pi)\}$ and $\{re^{i\phi} : c_1 - c_2 \le r \le c_1 + c_2, \phi \in [-\pi, \pi)\}$ for all $0 < c_2 < c_1$. Left: Sketch of how each individual set $c_1 e^{i\phi_1} + \{c_2 e^{i\phi_2} : \phi_2 \in [-\pi, \pi)\}$ looks like. Right: The union of these individual sets exhausts the full annulus.

can take any value in the annulus $\{re^{i\phi} : \max\{r_m, 0\} \le r \le 1, \phi \in [-\pi, \pi)\}$, where

$$r_m = \frac{1 - e^{-\Delta E/T}}{1 - e^{-m\Delta E/T}} - \sum_{j=1}^{m-1} \frac{(1 - e^{-\Delta E/T})e^{-j\Delta E/T}}{1 - e^{-m\Delta E/T}} = 2\frac{1 - e^{-\Delta E/T}}{1 - e^{-m\Delta E/T}} - 1.$$

However, $\lim_{m\to\infty} r_m = 1 - 2e^{-\Delta E/T}$, which is smaller than zero if and only if $T > \frac{\Delta E}{\ln 2}$; thus, by assumption, there exists $m \in \mathbb{N}$ such that $r_m < 0$, so $(\phi_1, \ldots, \phi_m) \mapsto \frac{\operatorname{tr}(e^{-H_{B,m}/T} U_\phi)}{\operatorname{tr}(e^{-H_{B,m}/T})}$ maps surjectively onto the closed unit disk. In other words, for this m, all partial dephasings can be implemented via relative phases, which is what we had to show.

Now, if $T \le \frac{\Delta E}{\ln 2}$, we will prove that the two sets in question do not coincide by showing that full dephasing is cannot be implemented using $H_{B,m}$. Indeed, given arbitrary $m \in \mathbb{N}$ and any $U \in \mathrm{U}(2m)$ such that $[U, H_S \otimes \mathbb{1}_B + \mathbb{1}_S \otimes H_{B,m}] = 0$, partitioning

$$U = \begin{pmatrix} U_{11} & U_{12} \\ U_{21} & U_{22} \end{pmatrix}$$

with $U_{11}, U_{12}, U_{21}, U_{22} \in \mathbb{C}^{m\times m}$ leads to

$$\Psi_T(\Phi_{T,m}(H_{B,m}, U)) = \frac{1}{\operatorname{tr}(e^{-H_{B,m}/T})} \begin{pmatrix} \operatorname{tr}(U_{12}^* U_{12} e^{-H_{B,m}/T}) \\ \operatorname{tr}(U_{22}^* U_{11} e^{-H_{B,m}/T}) \end{pmatrix}$$

as is verified by direct computation. We want this expression to be equal to $(0,0)^\top$. This means $\operatorname{tr}(U_{12}^* U_{12} e^{-H_{B,m}/T}) = 0$, which implies $U_{12} = 0$: this is due to the fact that $(A, B) \mapsto \operatorname{tr}(A^* B e^{-H_{B,m}/T})$ is an inner product on $\mathbb{C}^{m\times m}$ because $e^{-H_{B,m}/T}$ is positive definite. Thus, $A \mapsto \operatorname{tr}(A^* A e^{-H_{B,m}/T})$ is a norm on $\mathbb{C}^{m\times m}$, so it takes the value zero if and only if the input is zero. However, as U is unitary, $U_{12} = 0$ implies $U_{21} = 0$, so $U = U_{11} \oplus U_{22}$ for some $U_{11}, U_{22} \in \mathrm{U}(m)$. Moreover, U being energy-conserving yields $[U_{11}, H_B] = [U_{22}, H_B] = 0$, and as H_B is non-degenerate, by assumption, U_{11}, U_{22} (and thus U) have to be diagonal. However, for diagonal U, we already showed that full dephasing can be implemented if and only if $T > \frac{\Delta E}{\ln 2}$, which concludes this part of the proof.

Finally, the statement regarding the closure. By our previous argument (cf. Fig. 5), regardless of the temperature, at least some range of dephasing maps can be implemented (e.g., using diagonal unitaries U), that is, for all $T > 0$, there exists $r_0 < 1$ such that for all $c \in \mathbb{C}$, $|c| \in [r_0, 1]$,

$$S_c : \mathbb{C}^{2\times 2} \to \mathbb{C}^{2\times 2}$$
$$\begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \mapsto \begin{pmatrix} a_{11} & c^* a_{12} \\ c a_{21} & a_{22} \end{pmatrix}$$

is a thermal operation with bath Hamiltonian $\mathrm{diag}(0, \Delta E, \ldots, (m-1)\Delta E)$ for some m. On the other hand, we know that every qubit thermal operation S is the composition of a thermal operation S_m with associated $H_{B,m} := \mathrm{diag}(0, \Delta E, \ldots, (m-1)\Delta E)$ for some $m \in \mathbb{N}$ and a partial dephasing D_S (i.e., $\Psi_T(D_S) = (0, c_S)$ for some $c_S \in [0, 1]$). However, applying any S_c, $|c| < 1$ enough times approximates any degree of dephasing, i.e., $\lim_{k\to\infty} \Psi_T(S_{r_0}^k) = \lim_{k\to\infty}(0, r_0^k) = (0, 0)$. Thus, there are two cases: If $c_S > 0$, then there exist $k \in \mathbb{N}_0$ and $c \in [r_0, 1]$ such that $D_S = S_{r_0}^k \circ S_c$. Therefore, $S = S_{r_0}^k \circ S_c \circ S_m$, so S can be implemented exactly using finitely many truncated single-mode bosonic baths. However, if $c_S = 0$, then this can (only) be done approximately, i.e., $\lim_{k\to\infty} \| S - S_{r_0}^k \circ S_m \| = 0$. Therefore, $\mathsf{TO}(H_S, T)$ is a subset of the closure of the semigroup of thermal operations with bath Hamiltonian $H_{B,m} := \mathrm{diag}(0, \Delta E, \ldots, (m-1)\Delta E)$ [which itself is a subset of $\overline{\mathsf{TO}(H_S, T)}$], meaning that the two sets coincide in the closure.

(iv): This is Theorem 1.(2) in Ref. 20. Because our proof of (iii) (the "truncated version") is similar to their proof, we will omit the details, and we simply refer to Appendix B in their paper. □

REFERENCES

[1] F. Brandão, M. Horodecki, N. Ng, J. Oppenheim, and S. Wehner, "The second laws of quantum thermodynamics," Proc. Natl. Acad. Sci. U. S. A. **112**, 3275–3279 (2015).
[2] M. Horodecki and J. Oppenheim, "Fundamental limitations for quantum and nanoscale thermodynamics," Nat. Commun. **4**, 2059 (2013).
[3] J. M. Renes, "Work cost of thermal operations in quantum thermodynamics," Eur. Phys. J. Plus **129**, 153 (2014).
[4] P. Faist, J. Oppenheim, and R. Renner, "Gibbs–preserving maps outperform thermal operations in the quantum regime," New J. Phys. **17**, 043003 (2015).
[5] G. Gour, M. P. Müller, V. Narasimhachar, R. W. Spekkens, and N. Yunger Halpern, "The resource theory of informational nonequilibrium in thermodynamics," Phys. Rep. **583**, 1–58 (2015).
[6] M. Lostaglio, Á. Alhambra, and C. Perry, "Elementary thermal operations," Quantum **2**, 52 (2018).
[7] T. Sagawa, P. Faist, K. Kato, K. Matsumoto, H. Nagaoka, and F. G. S. L. Brandão, "Asymptotic reversibility of thermal operations for interacting quantum spin systems via generalized quantum Stein's lemma," J. Phys. A: Math. Theor. **54**, 495303 (2021).
[8] P. Mazurek, "Thermal processes and state achievability," Phys. Rev. A **99**, 042110 (2019).
[9] Á. M. Alhambra, M. Lostaglio, and C. Perry, "Heat-bath algorithmic cooling with optimal thermalization strategies," Quantum **3**, 188 (2019).
[10] M. Esposito and C. van den Broeck, "Second law and Landauer principle far from equilibrium," Europhys. Lett. **95**, 40004 (2011).
[11] M. Lostaglio, D. Jennings, and T. Rudolph, "Description of quantum coherence in thermodynamic processes requires constraints beyond free energy," Nat. Commun. **6**, 6383 (2015).
[12] M. Lostaglio, "An introductory review of the resource theory approach to thermodynamics," Rep. Prog. Phys. **82**, 114001 (2019).
[13] P. Skrzypczyk, A. J. Short, and S. Popescu, "Work extraction and thermodynamics for individual quantum systems," Nat. Commun. **5**, 4185 (2014).
[14] J. Scharlau and M. P. Mueller, "Quantum Horn's lemma, finite heat baths, and the third law of thermodynamics," Quantum **2**, 54 (2018).
[15] F. vom Ende, "Strict positivity and D-majorization," Linear Multilinear Algebra (published online, 2020).
[16] F. vom Ende and G. Dirr, "The d-majorization polytope," Linear Algebra Appl. **649**, 152–185 (2022).
[17] Á. Alhambra, J. Oppenheim, and C. Perry, "Fluctuating states: What is the probability of a thermodynamical transition?," Phys. Rev. X **6**, 041016 (2016).
[18] G. Gour, D. Jennings, F. Buscemi, R. Duan, and I. Marvian, "Quantum majorization and a complete set of entropic conditions for quantum thermodynamics," Nat. Commun. **9**, 5352 (2018).
[19] C. Perry, P. Ćwikliński, J. Anders, M. Horodecki, and J. Oppenheim, "A sufficient set of experimentally implementable thermal operations," Phys. Rev. X **8**, 041049 (2018).
[20] X. Hu and F. Ding, "Thermal operations involving a single-mode bosonic bath," Phys. Rev. A **99**, 012104 (2019).
[21] Y. Ding, F. Ding, and X. Hu, "Exploring the gap between thermal operations and enhanced thermal operations," Phys. Rev. A **103**, 052214 (2021).
[22] R. Dann and R. Kosloff, "Open system dynamics from thermodynamic compatibility," Phys. Rev. Res. **3**, 023006 (2021).
[23] R. Bhatia, *Positive Definite Matrices* (Princeton University Press, Princeton, 2007).
[24] Equivalently, H_B having a resonant spectrum with respect to H_S is characterized by the following condition: For all proper (non-empty) subsets I of $\{1, \ldots, m\}$, there exist $i \in I$ and $j \in \{1, \ldots, m\}\backslash I$ such that $E_i' - E_j' \in \sigma(\mathrm{ad}_{H_S})$. This means that the above graph cannot be written as the union of two (or more) disconnected components.
[25] N. Shiraishi, "Two constructive proofs on d-majorization and thermo-majorization," J. Phys. A: Math. Theor. **53**, 425301 (2020).
[26] W.l.o.g., let both H_S, H_B be diagonal in the standard basis. When expressed mathematically, the "mixing property" in question reads $\langle e_k, \Phi_{T,m}(H_B, U)(|e_i\rangle\langle e_j|)e_l\rangle \neq 0$. This readily implies the existence of indices α, β such that neither $\langle e_k \otimes e_\alpha, U(e_i \otimes e_\beta)\rangle$ nor $\langle e_l \otimes e_\alpha, U(e_j \otimes e_\beta)\rangle$ vanish. However, $\langle e_k \otimes e_\alpha, [U, H_S \otimes \mathbb{1} + \mathbb{1} \otimes H_B](e_i \otimes e_\beta)\rangle = (E_i + E_\beta' - E_k - E_\alpha')\langle e_k \otimes e_\alpha, U(e_i \otimes e_\beta)\rangle$ does always vanish due to U being energy-conserving; therefore, $E_i - E_k = E_\alpha' - E_\beta'$ (and similarly $E_j - E_l = E_\alpha' - E_\beta'$ for the second term).
[27] M. Lostaglio, K. Korzekwa, D. Jennings, and T. Rudolph, "Quantum coherence, time-translation symmetry, and thermodynamics," Phys. Rev. X **5**, 021001 (2015).
[28] A. Shu, Y. Cai, S. Seah, S. Nimmrichter, and V. Scarani, "Almost thermal operations: Inhomogeneous reservoirs," J. Phys. A: Math. Theor. **100**, 042107 (2019).
[29] R. van der Meer, N. Ng, and S. Wehner, "Smoothed generalized free energies for thermodynamics," Phys. Rev. A **96**, 062135 (2017).
[30] E. Bäumer, M. Perarnau-Llobet, P. Kammerlander, H. Wilming, and R. Renner, "Imperfect thermalizations allow for optimal thermodynamic processes," Quantum **3**, 153 (2019).
[31] J. G. Richens, Á. M. Alhambra, and L. Masanes, "Finite-bath corrections to the second law of thermodynamics," Phys. Rev. E **97**, 062132 (2018).

[32] K. Kuratowski, *Topology* (Academic Press, New York, 1966), Vol. I.
[33] P. Ćwikliński, M. Studziński, M. Horodecki, and J. Oppenheim, "Towards fully quantum second laws of thermodynamics: Limitations on the evolution of quantum coherences," Phys. Rev. Lett. **115**, 210403 (2015).
[34] W. Nernst, "Thermodynamik und spezifische wärme," in *Sitzungsbericht der Königlich Preussischen Akademie der Wissenschaften* (Verlag der Königlichen Akademie der Wissenschaften, Berlin, 1912), Vol. 134.
[35] E. M. Loebl, "The third law of thermodynamics, the unattainability of absolute zero, and quantum mechanics," J. Chem. Educ. **37**, 361–363 (1960).
[36] Recall that for $n \in \mathbb{N}$ and some positive $d \in \mathbb{R}^n_{++}$, the set of Gibbs-stochastic (or "d-stochastic" in the mathematics literature; cf. Chap. 14.B in Ref. 48) $n \times n$ matrices is defined as the set of all $A \in \mathbb{R}^{n\times n}$ with non-negative entries such that $Ad = d$ and ${}^{\top}A = {}^{\top}$ where $:= (1, \ldots, 1)^{\top}$.
[37] M.-D. Choi, "Completely positive linear maps on complex matrices," Linear Algebra Appl. **10**, 285–290 (1975).
[38] By this, we mean the interior relative to the subset $\Psi_T^{-1}(\mathbb{R}^3)$ of $\mathscr{L}(\mathbb{C}^{2\times 2})$, that is, the usual interior when considering Fig. 2.
[39] A. Holevo, *Quantum Systems, Channels, Information: A Mathematical Introduction*, De Gruyter Studies in Mathematical Physics Vol. 16 (De Gruyter, Berlin, 2012).
[40] K. Korzekwa, "Structure of the thermodynamic arrow of time in classical and quantum theories," Phys. Rev. A **95**, 052318 (2017).
[41] A. de Oliveira Junior, J. Czartowski, K. Życzkowski, and K. Korzekwa, "Geometric structure of thermal cones," arXiv:2207.02237 (2022).
[42] D. Pérez-García, M. M. Wolf, D. Petz, and M. B. Ruskai, "Contractivity of positive and trace–preserving maps under L_p–norms," J. Math. Phys. **47**, 083506 (2006).
[43] Given some permutation $\pi \in S_n$, the corresponding permutation matrix $\underline{\pi}$ is given by $\sum_{i=1}^n |e_i\rangle\langle e_{\pi(i)}|$. In particular, the identities $(\underline{\pi}x)_j = x_{\pi(j)}$ and $\underline{\pi \circ \tau} = \underline{\tau} \cdot \underline{\pi}$ hold for all $\pi, \tau \in S_n$, $x \in \mathbb{C}^n$, $j \in \{1, \ldots, n\}$.
[44] Given $m, n \in \mathbb{N}$, $A \in \mathbb{C}^{nm\times \mathrm{nm}}$, and any orthonormal basis $(g_j)_{j=1}^n$ of $\mathbb{C}^n$, one can decompose $A = \sum_{i,j=1}^n |g_i\rangle\langle g_j| \otimes A_{ij}$, where $A_{ij} := \mathrm{tr}_1((|g_j\rangle\langle g_i| \otimes \mathbb{1})A)$ for all $i, j = 1, \ldots, n$.
[45] The non-degenerate case, that is, $H_S = \mathbb{1}_2$, is a direct consequence of the fact that in two dimensions, the set of all unital quantum maps $[\mathsf{EnTO}(\mathbb{1}, T)]$ equals the convex hull of all unitary channels $[\mathrm{conv}\overline{\mathsf{TO}(\mathbb{1}, T)} = \overline{\mathsf{TO}(\mathbb{1}, T)}$, Proposition 4]; cf. Ref. 49.
[46] A. Brøndsted, *An Introduction to Convex Polytopes*, Graduate Texts in Mathematics Vol. 90 (Springer, New York, 1983).
[47] M. Nielsen and I. Chuang, *Quantum Computation and Quantum Information*, 10th ed. (Cambridge University Press, Cambridge, 2010).
[48] A. Marshall, I. Olkin, and B. Arnold, *Inequalities: Theory of Majorization and its Applications*, 2nd ed. (Springer, New York, 2011).
[49] L. J. Landau and R. F. Streater, "On Birkhoff's theorem for doubly stochastic completely positive maps of matrix algebras," Linear Algebra Appl. **193**, 107–127 (1993).