Entropy plays a central role both in thermodynamics and in information theory. This is remarkable, as the two theories appear to be fundamentally different. Thermodynamics is a phenomenological theory, concerned with the description of large physical systems, such as steam engines or fridges. It relies on concepts like work or heat, which are defined in terms of macroscopic observables. Information theory, on the other hand, deals with “knowledge” on a rather abstract level. Like statistical mechanics, it refers to the microscopic states of a system, such as the values of the individual bits stored in a memory device.

Accordingly, the notion of entropy is rather different in the two theories. In thermodynamics, entropy is a function of the macroscopic state of a physical system which describes phenomenologically which processes are possible independently of any microscopic model. Following Clausius, it is conventionally defined in terms of the heat that flows into a system at a given temperature, and it lends its operational significance from the second law [1, 2]. In information theory, entropy was originally introduced by Shannon to quantify the information content of data or, equivalently, the uncertainty one has about them [3]. Operationally, it characterises properties such as the compression length, i.e., the minimum number of bits needed to store the data. Mathematically, the Shannon entropy is a function of the probability distribution of the random variable that models the data. The von Neumann entropy [4], which is also known from quantum statistical mechanics, provides a generalisation of this concept to the case where information is represented by the state of a quantum-mechanical system.

The connections between thermodynamics and information theory have been systematically studied in the context of Landauer’s principle [5–7]. The principle entails that deleting information requires work, which is in turn dissipated as heat. It thus relates information-theoretic entropy to heat flow and, therefore, to thermodynamic entropy. Derivations of Landauer’s principle often start from microscopic considerations, for example using standard tools from statistical mechanics [8–10], or explicit microscopic models inspired from information theory [11–21].

In this Letter, we take a different, more axiomatic, approach. It relies on a framework proposed by Lieb and Yngvason [22–24] for the study of thermodynamic entropy. Within this framework thermodynamic entropy is obtained via a small set of axioms that naturally characterise adiabatic processes. Our work is based on the observation that the framework can as well be used to capture key concepts from information theory. It therefore provides a common ground for the study of entropy measures both in phenomenological thermodynamics and in information theory. This allows us to establish a novel connection between them.

Furthermore, we extend this connection to “single-shot” generalizations of the Shannon and von Neumann entropy. While the latter are only applicable to study data that has a particular structure (e.g., that has been produced by infinitely many independent invocations of a source), single-shot entropies do not suffer from this limitation (see [25, 26] for examples of how these entropies are used in applications). These entropy measures, in fact, naturally arise from Lieb and Yngvason’s axiomatic framework for nonequilibrium thermodynamics.

The study of entropy has a long tradition both in physics and in information theory. In information theory, several axiomatizations of the Shannon or von Neumann entropy have been proposed [3, 27–30]. Our approach is different, as it relies on an axiomatization of the thermodynamic entropy. Our work also bears some resemblance to the study of entanglement transformation using similar tools [31–33].

The remainder of this Letter is organised as follows. We start with a brief summary of the Lieb-Yngvason framework for thermodynamics and describe how entropy measures are defined within this framework. As a first technical contribution, we show that the framework is as well applicable to an information-theoretic description of thermal systems (Proposition 1). We then show that the corresponding entropy measures defined within the framework coincide with information-theoretic single-shot entropies (Proposition 2). Subsequently, we extend these considerations to various classes of thermo-
dynamic processes involving reservoirs and relate them to information-theoretic counterparts.

Lieb and Yngvason’s approach.—In their axiomatic framework, Lieb and Yngvason [22–24] consider the set $\Gamma$ of all equilibrium states of a thermodynamic system and equip this space with an order relation, denoted $\prec$. For $X$ and $Y \in \Gamma$, $X \prec Y$ means that $Y \in \Gamma$ is “adiabatically accessible” from the state $X \in \Gamma$ “by means of an interaction with some device consisting of some auxiliary system and a weight in such a way that the auxiliary system returns to its initial state at the end of the process, whereas the weight may have risen or fallen” [22] (see Figure 1). The framework also describes the composition of systems as well as their scaling, corresponding to taking an arbitrary amount of a substance. Within this framework, an entropy measure, $S$, is a function on $\Gamma$ that has certain natural properties. In particular, Lieb and Yngvason demand that $S$ should be monotonic with respect to the order relation $\prec$, additive with respect to the composition operation, and extensive in the scaling (we refer to the Appendix for a more detailed description of these properties). They show that, provided the order $\prec$ obeys thermodynamically reasonable axioms, $S$ is determined uniquely by these properties, up to an affine transformation that may be fixed by choosing the entropy of two (arbitrary) reference states $X_0$ and $X_1$. Specifically, $S$ is given by

$$S(X) = \sup \{ \lambda : ((1 - \lambda)X_0, \lambda X_1) \prec X \} \quad (1a)$$

$$= \inf \{ \lambda : X \prec ((1 - \lambda)X_0, \lambda X_1) \}, \quad (1b)$$

where $\lambda X$ denotes the state of a system obtained by scaling a system in state $X$ by a factor $\lambda$, and $(X, X')$ denotes the state of the system obtained by composing systems in state $X$ and $X'$, respectively. Intuitively, if the state $X$ can be reached adiabatically from $X_0$ and $X_1$ can be attained from $X$, then the entropy $S(X)$ is defined as the optimal $\lambda$ such that the state $X$ can be created from $X_0$ and $X_1$ combined at a ratio $(1 - \lambda) : \lambda$ by an adiabatic process. Physically, $S$ corresponds to the usual thermodynamic entropy as defined by Clausius via the reversible heat dissipated $\delta Q_{\text{rev}}$ as $S = \delta Q_{\text{rev}}/T$. This can be intuitively understood from Clausius’ explanation of the second law [1, 2] combined with the uniqueness result in [23]; Clausius’ physical processes agree with the picture of an adiabatic process in Lieb and Yngvason’s framework. The connection is formally established through Lieb and Yngvason’s re-derivation of thermodynamics [23].

The framework can be extended to include non-equilibrium states [24]. The states of the corresponding extended state space, $\Gamma_{\text{ext}}$, generally obey weaker axioms than those of $\Gamma$. For instance, they may not be scaleable. The entropy $S$ can thus not be uniquely extended to $\Gamma_{\text{ext}}$. Instead, it can be shown that all monotonic extensions $S_{\text{ext}}$ of $S$ to the space $\Gamma_{\text{ext}}$ are contained within two bounds, namely

$$S_-(X) = \sup \{ S(X') : X' \in \Gamma, X' \prec X \} \quad (2a)$$

$$S_+(X) = \inf \{ S(X'') : X'' \in \Gamma, X \prec X'' \}. \quad (2b)$$

These entropy measures can be used to characterise the (im)possibility of state transformations by adiabatic processes (according to the definition above). In particular, they provide a sufficient condition for the possibility to transform $X \in \Gamma_{\text{ext}}$ to $Y \in \Gamma_{\text{ext}}$.

$$S_+(X) \leq S_-(Y) \implies X \prec Y. \quad (3)$$

Similarly, they also provide a necessary condition for such a transformation,

$$X \prec Y \implies S_-(X) \leq S_-(Y) \text{ and } S_+(X) \leq S_+(Y). \quad (4)$$

We can also use Equations (1a) and (1b) to define two further entropy measures as

$$\tilde{S}_-(X) = \sup \{ \lambda : ((1 - \lambda)X_0, \lambda X_1) \prec X \} \quad (5a)$$

$$\tilde{S}_+(X) = \inf \{ \lambda : X \prec ((1 - \lambda)X_0, \lambda X_1) \}. \quad (5b)$$

These have the advantage that their definition does not involve neighbouring states $X'$ and $X''$. While in thermodynamics the intervals $[\tilde{S}_-, \tilde{S}_+]$ and $[S_-, S_+]$ coincide, the interval $[S_-, \tilde{S}_+]$ is generally — and especially for quantum states — smaller. A detailed analysis is provided in the Appendix.

Information-theoretic entropy measures.—Information theory is concerned with data and their processing. In quantum information theory, which we consider here for generality, data is encoded in quantum systems (which include classical systems as a special case), whose states we describe by the usual density operator formalism. In order to quantify the information content of data, we appeal to a family of information measures, the Rényi entropies [34] (cf. Appendix). Two important members of this family are the min- and the max-entropy. The min-entropy is defined as

$$H_{\text{min}}(\rho) = -\log \| \rho \|_\infty, \quad (6)$$

where $\| \rho \|_\infty$ denotes the maximal eigenvalue of $\rho$. Operationally, it describes the amount of randomness that can be extracted deterministically from data in state $\rho$ [35, 36]. The max-entropy is defined as

$$H_{\text{max}}(\rho) = \log \text{rank } \rho, \quad (7)$$

and quantifies the number of (qu)bits needed to store data in state $\rho$ [25].

An Information-Theoretic Description of Adiabatic Processes.—In order to apply Lieb and Yngvason’s framework to an information-theoretic description of thermodynamic systems, we need to formally specify the various ingredients (such as the order relation) which the abstract framework requires. First, we identify the set of “equilibrium states” of an information-bearing quantum system. We define these as those quantum states represented by flat density operators (i.e., operators whose non-zero eigenvalues are all equal). This, in turn, allows us to derive the notion of an “adiabatic process,” in
the sense of Lieb and Yngvason, for microscopic systems. Their notion of an adiabatic process (see Figure 1(a)), here, translates to the following three quantum operations:

- addition of an extra ancilla system (“device”), in an equilibrium state;
- interaction of the system and the extra device with a weight system with a joint, energy-preserving unitary;
- removal of the extra device, restored to its original state.

Consider for simplicity a system with a trivial, i.e. fully degenerate Hamiltonian: energy and thus the weight can only have changed its relative height. The change of the state of the system itself is represented as a deformation of the cube here. (b) The system is connected to a heat bath at temperature T. Adiabatic processes with the aid of an auxiliary system and a weight are applied to this combined system.

Figure 1. (a) Adiabatic process according to Lieb and Yngvason’s definition. A system, represented as a cube, interacts with some device and a weight. After the process the device has to be in its initial state again and the weight can only have changed its relative height. The change of the state of the system itself is represented as a deformation of the cube here. (b) The system is connected to a heat bath at temperature T. Adiabatic processes with the aid of an auxiliary system and a weight are applied to this combined system.

Thermodynamics of our Information-Theoretic Model.—We now seek to apply Lieb and Yngvason’s framework to quantum information systems. As we have just seen, an adiabatic process, which defines the order relation \( \prec \), corresponds to accessibility by noisy operations, i.e. \( \rho \prec \rho' \) if and only if \( \rho' \) can be reached from \( \rho \) via noisy operations.

Noisy operations are characterized by the mathematical notion of majorization: the existence of a noisy operation transforming a state \( \rho \) to a state \( \rho' \) is equivalent to the condition that \( \rho \) majorizes \( \rho' \) \(^{[37]}\). This is defined as follows. Let \( \{p_i\}_i \) and \( \{p'_i\}_i \) be the eigenvalues of \( \rho \) and \( \rho' \) arranged in decreasing order. Then \( \rho \) majorizes \( \rho' \), denoted \( \rho \prec_M \rho' \), iff \( \sum_{i=1}^k p_i \geq \sum_{i=1}^k p_i' \) for all \( k = 1, 2, \ldots, \dim(\mathcal{H}) \). (Note that we use a different notation than the conventional ‘\( \succ \)” used e.g. in Ref. \(^{[39]}\), to avoid confusion with Lieb and Yngvason’s adiabatic processes.)

We define the composition of states naturally as their tensor product; establishing a reasonable scaling operation is more involved. We assume scaling a quantum system by a natural factor \( \lambda \in \mathbb{N} \) to mean combining \( \lambda \) such systems. Thus, the scaling coincides with the composition operation. We generalize this concept to non-integer scaling factors by extending the space of equilibrium states to be continuous. For simplicity, consider only states \( \rho \in \mathcal{S}(\mathcal{H}) \) which are diagonal in a common eigenbasis, which does not restrict the generality of our considerations, as explained in the Appendix. Represent \( \rho \)’s eigenvalues \( p_1 \geq p_2 \geq \ldots \geq p_{\dim \mathcal{H}} \) as a step function

\[
 f_\rho(x) = \begin{cases} 
 p_i & \text{if } i - 1 \leq x \leq i \\
 0 & \text{otherwise} 
\end{cases} 
\]

For an equilibrium state \( \sigma \), for which by assumption all non-zero eigenvalues are equal, the step function

\[
 f_\sigma(x) = \begin{cases} 
 \frac{1}{\text{rank}(\sigma)} & \text{if } 0 \leq x \leq \text{rank}(\sigma) \\
 0 & \text{otherwise} 
\end{cases} 
\]

is scaled as \( f_{\lambda \sigma}(x) = f_\sigma(x^\lambda) \), which coincides with the composition for \( \lambda \in \mathbb{N} \) but allows for a formal continuation to any \( \lambda \in \mathbb{R}_{\geq 0} \). This scaled state might be unphysical if \( \lambda \) is not an integer. However, we show in the Appendix that, by considering larger systems, the framework can be reformulated in such a way that only scaling by an integer factor is needed.

Main Results.—Consider a Hilbert space \( \mathcal{H} \), and consider the order relation \( \prec_M \) which corresponds to accessibility by noisy operations, as defined above. We may now state our main results.

Proposition 1. The ordering \( \prec_M \) on \( \mathcal{H} \) obeys Lieb and Yngvason’s axioms with respect to the operations of composition and scaling defined above.

The axioms can be checked through a straightforward calculation, which we carry out in the Appendix.
Proposition 2. For states on $\mathcal{H}$ ordered by means of $\prec_M$, the unique entropy function $S$ for equilibrium states coincides with the von Neumann entropy $H(\rho) = -\text{tr}(\rho \log \rho)$. Furthermore, the two entropic quantities $S_-$ and $\tilde{S}_+$ are equal to $H_{\text{min}}$ and $H_{\text{max}}$.

We provide the proof in the Appendix. The bounds $\tilde{S}_-$ and $\tilde{S}_+$ and their relation to $S_-$ and $S_+$ are visualized in Figure 2. While $S_+$ and $\tilde{S}_+$ coincide, the discrete quantity $S_-$ differs from $\tilde{S}_-$.

Other thermodynamic processes.—We have, up to this point, considered only isolated systems. It seems however natural to ask for analogues of thermodynamic scenarios describing other types of processes, for instance systems interacting with reservoirs. In the most obvious case of a system connected to a heat bath, depicted in Figure 1(b), we assume our system to be in thermal contact with a heat reservoir at temperature $T$, which in statistical mechanics would be described by a canonical ensemble. A system in equilibrium is thus in a thermal state with respect to the heat bath.

On the system and the reservoir we still allow for the above adiabatic processes, i.e. we can describe the allowed state transformations by means of noisy operations. As long as the states of the system alone are block diagonal in the energy eigenbasis, our noisy operations on system and reservoir can be reduced to thermal operations on the states of the system, as one can understand from [15]. Thermal operations [13] consist of the following steps:

- addition of an ancillary system in a thermal state relative to the heat bath;
- unitary transformation of the system and the ancilla commuting with the total Hamiltonian;
- removal of any subsystem.

For states which are diagonal in the energy eigenbasis, the existence of a state transformation by a thermal operation is expressed in terms of thermo-majorization [15]. Let $\rho = \sum \lambda_i p_i |E_i\rangle\langle E_i| \in \mathcal{S}(\mathcal{H})$ be a density matrix diagonal in the energy eigenbasis and let $d = \dim \mathcal{H}$.

Represent its spectrum as a step function

$$f_\rho(x) = \begin{cases} p_i, & i - 1 < x \leq i, \\ 0, & \text{otherwise} \end{cases}$$

with ordered eigenvalues $p_1 \geq p_2 \geq \ldots \geq p_d$. The Gibbs-rescaled version of this step function is defined as

$$f_\rho^T(x) = \begin{cases} \frac{p_1}{e^{\beta E_1}}, & \sum_{k=1}^{x-1} e^{-\beta E_j} \leq x \leq \sum_{k=1}^x e^{-\beta E_j}, \\ 0, & \text{otherwise} \end{cases}$$

with differently ordered terms $\frac{p_1}{e^{\beta E_1}} \geq \frac{p_2}{e^{\beta E_2}} \geq \ldots \geq \frac{p_d}{e^{\beta E_d}}$. The idea of Gibbs-rescaling was introduced in [40–42] and applied in [15, 16]. For states $\rho$ and $\sigma \in \mathcal{S}(\mathcal{H})$ which are block diagonal in the energy eigenbasis, the order relation of thermo-majorization $\prec_T$ can be defined as

$$\rho \prec_T \sigma \iff \int_0^T f_\rho^T(x)dx \geq \int_0^T f_\sigma^T(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}.$$  

It obeys Lieb and Yngvason’s axioms (shown in the Appendix), which enables calculating the unique potential for thermal states as well as bounds on the extension of the potential function to arbitrary block diagonal states. In fact, we recover the “single-shot free energies” $F_{\text{min}}$ and $F_{\text{max}}$ introduced in [15, 20] to describe the work needed for the formation of a state as well as the extractable work.

Proposition 3. For states ordered by means of thermal operations through the relation $\prec_T$ the unique potential $S$ for thermal states coincides with the Helmholtz free energy $F$, and the two quantities $\tilde{S}_-$ and $\tilde{S}_+$ correspond to $F_{\text{max}}$ and $F_{\text{min}}$.

A detailed calculation can be found in the Appendix. Scenarios including other types of reservoirs [21], such as a particle or an angular momentum reservoir, yield analogous results. Various settings, along with their corresponding order relation, equilibrium states and resulting state functions are summarized in Table I.

Conclusions.—We have shown that, with minor adaptations, Lieb and Yngvason’s approach is directly applicable to information theory, allowing us to put thermodynamic and information theoretic entropy on the same footing. Compared to other works based on Landauer’s

![Figure 2. Visualization of the entropic bounds for non-equilibrium states. The spectrum of a state $\rho$ is depicted as a step function (blue). (a) Two equilibrium states $(1 - \lambda\rho_0, \lambda\rho_1)$ and $(1 - \lambda\rho_0, \lambda\rho_1)$ with entropies $\tilde{S}_-$ and $\tilde{S}_+$ are shown (orange). Note that $(1 - \lambda\rho_0, \lambda\rho_1)$ does not describe an actual physical state, as a corresponding state would have non-integer rank. (b) Lieb and Yngvason’s bounds $S_-$ and $\tilde{S}_+$ correspond to $\rho'$ and $\rho''$ are shown. While $S_-$ and $\tilde{S}_+$ are equal the quantity $\tilde{S}_-$ is a discretised version of $\tilde{S}_-$ and describes an actual physical state.](image-url)
Table I. An overview on the application of Lieb and Yngvason’s framework to various scenarios. In the first line we describe Lieb and Yngvason’s original scenario in the case of quantum states. The rest of the table contains adapted scenarios, where the systems have additional interactions with reservoirs, as denoted in the first column. In the second column, we present the corresponding resource theoretic scenarios, the selected class of processes as well as their associated order relation. The remaining columns detail the equilibrium states and the entropic quantities $S$, $S_-$ and $S_+$ corresponding to each setting.

| Setting                          | Processes                      | Equilibrium States                        | Potential $S$ | Bounds $\bar{S}_-, \bar{S}_+$ |
|---------------------------------|--------------------------------|-------------------------------------------|---------------|---------------------------------|
| Isolated system                 | Noisy operations, $\prec_M$   | $\sum_i \frac{1}{\text{rank } \rho} |X_i \rangle \langle X_i|$ | Entropy $H$   | $H_{\text{min}}$, $H_{\text{max}}$ |
| Interaction with a heat bath    | Thermal operations, $\prec_T$ | $\sum_i e^{-\beta E_i \frac{Z}{Z}} |E_i \rangle \langle E_i|$ | Free Energy $F$ | $F_{\text{min}}$, $F_{\text{max}}$ |
| Interaction with a heat bath and a particle reservoir | N.T.-operations, $\prec_{N,T}$ | $\sum_i e^{-\beta (E_i - N_i \mu)} \times |E_i, N_i \rangle \langle E_i, N_i|$ | Grand potential $\Omega$ | $\Omega_{\text{min}}$, $\Omega_{\text{max}}$ |
| Interaction with an angular momentum reservoir | J.-operations, $\prec_J$ | $\sum_i e^{-\beta \gamma J_i \frac{Z}{Z}} |J_i \rangle \langle J_i|$ | Potential $S_J$ | $\bar{S}_{J-}$, $\bar{S}_{J+}$ |

We expect that this approach can be extended further; by slightly changing the order relation to a “smoothed majorization relation”, we presume it to yield the corresponding smoothed entropy measures. Also, for a non-isolated system, our approach is limited to states that are block diagonal in a corresponding eigenbasis, e.g. in the particular case of a heat bath, in the energy eigenbasis [15]. We leave the question of generalizing our results for interacting systems to states with nonzero off-diagonal entries open for further investigation. In addition, we have not allowed the agent carrying out the processes to be assisted by side information about the system, which could be useful for performing thermodynamic operations [12]. We might expect that an appropriate extension of Lieb and Yngvason’s framework would provide an axiomatic and operationally well-justified definition of the conditional entropy.

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APPENDIX

Appendix A: Revisiting Lieb and Yngvason’s axiomatic approach

Lieb and Yngvason have contributed an axiomatic approach to derive an entropy function for thermodynamic equilibrium states [22, 23]. Recently, they have extended their approach to non-equilibrium states [24], which also enables them to make predictions relevant for non-equilibrium thermodynamics.

Lieb and Yngvason consider a preorder \( \prec \) on a set \( \Gamma \); physically \( \Gamma \) is the space of all equilibrium states of a system. A preorder is reflexive, transitive, but in contrast to a partial order not antisymmetric. This means that if two elements denoted by \( X \) and \( Y \) in \( \Gamma \) satisfy \( X \prec Y \) as well as \( Y \prec X \), this does not imply that they are the same element of the set \( \Gamma \). In line with Lieb and Yngvason’s terminology, we will call \( \prec \) an “order relation” or just “order” in the following. Whenever both relations \( X \prec Y \) and \( Y \prec X \) hold, this is denoted by \( Y \sim X \), while \( X \prec Y \) but not \( Y \prec X \) is denoted by \( X \prec Y \).

Elements \( X_1 \in \Gamma_1 \) and \( X_2 \in \Gamma_2 \) of possibly different sets \( \Gamma_1 \) and \( \Gamma_2 \) can be composed, physically meaning that they can be set next to each other and considered as one combined system. The arising composed state is symbolically denoted as \( (X_1, X_2) \in \Gamma_1 \times \Gamma_2 \). Note that the cartesian product \( \Gamma_1 \times \Gamma_2 \) denotes the space of composed systems \( (X_1, X_2) \), where the composition operation is associative and commutative.

In addition, any element \( X \in \Gamma \) can be scaled, i.e. for any \( \lambda > 0 \) one can define a scaled element denoted as \( \lambda X \in \lambda \Gamma \). The scaling is required to obey \( 1X = X \) as well as \( \lambda_1 ( \lambda_2 X) = ( \lambda_1 \lambda_2 )X \). For the sets \( \Gamma \), the required properties are \( 1 \Gamma = \Gamma \) and \( \lambda_1 ( \lambda_2 \Gamma) = ( \lambda_1 \lambda_2 )\Gamma \), where \( \lambda \Gamma \) symbolically denotes the space of scaled elements \( \lambda X \). Scaling a system by a factor \( \lambda \) means taking \( \lambda \) times the amount of substance contained in the original system.

The order relation \( \prec \) satisfies by assumption the following six axioms E1 to E6 as well as the Comparison Hypothesis.

- Reflexivity (E1): \( X \sim X \).
- Transitivity (E2): \( X \prec Y \) and \( Y \prec Z \Rightarrow X \prec Z \).
- Consistent composition (E3): \( X \prec X' \) and \( Y \prec Y' \Rightarrow (X, Y) \prec (X', Y') \).
- Scaling invariance (E4): \( X \prec Y \Rightarrow \lambda X \prec \lambda Y \forall \lambda > 0 \).
- Splitting and recombination (E5): For \( 0 < \lambda < 1 \), \( X \sim (\lambda X, (1 - \lambda )X) \).
- Stability (E6): If \( (X, \varepsilon Z_0) \prec (Y, \varepsilon Z_1) \) for a sequence of scaling factors \( \varepsilon \in \mathbb{R} \) tending to zero, then \( X \prec Y \).

- Comparison Hypothesis: Any two elements in a set \( (1 - \lambda )\Gamma \times \lambda \Gamma \) with \( 0 \leq \lambda \leq 1 \) are related with \( \prec \).

Note that Lieb and Yngvason do not regard the Comparison Hypothesis as an axiom but rather derive it from additional reasonable axioms about thermodynamical systems.

Lieb and Yngvason’s contribution concerns possible “entropy functions” on \( \Gamma \). More precisely, they show that there is a (essentially) unique function \( S \) on \( \Gamma \) which satisfies the following properties:

- Additivity: For any two states \( X \in \Gamma \) and \( X' \in \Gamma' \), \( S((X, X')) = S(X) + S(X') \) holds.
- Extensivity: For any \( \lambda > 0 \) and any \( X \in \Gamma \), \( S(\lambda X) = \lambda S(X) \) holds.
- Monotonicity: If two states \( X \) and \( \tilde{X} \) are comparable with \( \prec \), then \( X \prec \tilde{X} \Leftrightarrow S(X) \leq S(\tilde{X}) \).

Lieb and Yngvason’s second law is restated in the following theorem.

**Theorem 4** (Lieb & Yngvason). Provided that the six axioms E1 to E6 as well as the Comparison Hypothesis are fulfilled, there exists a function \( S \) that is additive under composition, extensive in the scaling and monotonic with respect to \( \prec \). Furthermore, this function \( S \) is unique up to affine transformations.

For a state \( X \in \Gamma \), the unique function \( S \) is given as

\[
S(X) = \sup \{ \lambda : ((1 - \lambda )X_0, \lambda X_1) \prec X \} = \inf \{ \lambda : X \prec ((1 - \lambda )X_0, \lambda X_1) \},
\]

where arbitrary elements \( X_0 \prec X_1 \) in \( \Gamma \) can be chosen, defining a gauge. Since the scaling is continuous, the supremum and the infimum are attained, and the function \( S \) can be conveniently expressed as

\[
S(X) = \{ \lambda : ((1 - \lambda )X_0, \lambda X_1) \sim X \}.
\]

To describe non-equilibrium states of a thermodynamic system, additional elements, not satisfying the scaling property, are introduced. Adding such non-scalable elements to the set \( \Gamma \) we obtain an extended set \( \Gamma_{ext} \) for which we require the following.

- N1: For any \( X' \in \Gamma_{ext} \) there exist \( X_0 \) and \( X_1 \in \Gamma \) with \( X_0 \prec X' \prec X_1 \).
- N2: Axioms E1, E2, E3 and E6, where \( Z_0 \) and \( Z_1 \in \Gamma \) in axiom E6, hold on \( \Gamma_{ext} \).

The first requirement ensures that the non-scalable elements are comparable to at least two elements \( X_0 \) and \( X_1 \) of the set \( \Gamma \), and hence via transitivity to all of them. Furthermore, it assures that the non-scalable elements are not at the boundary of the extended set \( \Gamma_{ext} \) with respect to the preorder \( \prec \). For such non-equilibrium states, Lieb and Yngvason [24] come to the following conclusion.
Proposition 5. On condition that $N1$ and $N2$ hold for any non-equilibrium state $X \in \Gamma_{\text{ext}}$, the two functions $S_-$ and $S_+$ defined as
\begin{align}
S_-(X) &= \sup \{S(X') : X' \in \Gamma, X' \prec X\} \tag{A4} \\
S_+(X) &= \inf \{S(X'') : X'' \in \Gamma, X \prec X''\} \tag{A5}
\end{align}
bound all possible extensions $S_{\text{ext}}$ of $S$ to the set $\Gamma_{\text{ext}}$ which are monotonic with respect to the relation $\prec$.

This implies that for a state $X \in \Gamma_{\text{ext}}$, the attained value $S_{\text{ext}}(X)$ of such an extension always lies in between the values $S(X')$ and $S(X'')$ of its neighboring scalable elements according to the order relation $\prec$: $S_-(X) \leq S_{\text{ext}}(X) \leq S_+(X)$.

We will prefer to work with the following alternative quantities, which only rely on the state $X$ and not on any “neighbouring” equilibrium states $X'$ and $X''$:
\begin{align}
\tilde{S}_-(X) &= \sup \{\lambda : ((1-\lambda)X_0, \lambda X_1) \prec X\} \tag{A6} \\
\tilde{S}_+(X) &= \inf \{\lambda : X \prec ((1-\lambda)X_0, \lambda X_1)\}, \tag{A7}
\end{align}
rather than using Lieb and Yngvason’s bounds (A4) and (A5). Operationally, $\tilde{S}_-$ specifies the portion of the system that can maximally be in state $X_1$ if one wants to create the state $X$ by composing subsystems in states $X_0$ and $X_1$. The minimal portion of $X_1$ one can obtain by transforming $X$ into a composition of two smaller systems in states $X_0$ and $X_1$ is characterized by $\tilde{S}_+$.

Note that in thermodynamics the two sets of bounding quantities $\{S_- , S_+\}$ and $\{\tilde{S}_-, \tilde{S}_+\}$ coincide, as due to the continuity of the thermodynamic quantities an equilibrium state $X' \in \Gamma$ with $X' \sim ((1-\lambda)X_0, \lambda X_1)$ exists for any $\lambda$. However, this does not directly follow from the axioms and the interval $[\tilde{S}_-, \tilde{S}_+]$ is generally smaller than $[S_-, S_+]$ and may not contain all monotonic extensions $S_{\text{ext}}$ of the entropy function $S$. We will see in Appendix C that our quantities $\tilde{S}_-$ and $\tilde{S}_+$ are advantageous in the quantum case.

Appendix B: A short introduction to selected resource theoretic aspects

In general, a resource theory deals with an agent that is only allowed to execute operations of a predefined class, and investigates which tasks can be accomplished.

For quantum resource theories, the state space on which these operations act consists of density operators $\rho$, i.e. positive semidefinite operators of unit trace on a Hilbert space $\mathcal{H}$. We denote the set of all density operators on $\mathcal{H}$ as $S(\mathcal{H})$.

In a resource theory, the value of a state as a means to achieve a certain task can be quantified by a monotone, a measure that is monotonic under the selected class of operations. A state is said to be a resource if it can not be prepared with operations of the allowed class only.

1. The resource theory of noisy operations

The resource theory of noisy operations [37, 43, 44] can be defined by allowing the following operations:

- Addition of an ancillary system in a maximally mixed state.
- Unitary transformation of the system.
- Removal of any subsystem (by taking the partial trace).

Horodecki and Oppenheim [37] have shown that, for finite-dimensional systems, a state $\rho \in S(\mathcal{H})$ can be transformed into a state $\tilde{\rho} \in S(\mathcal{H})$ by noisy operations if and only if the spectrum of $\rho$ majorizes the spectrum of $\tilde{\rho}$.

Definition 6. Let $\rho, \tilde{\rho} \in S(\mathcal{H})$ with $d = \dim \mathcal{H}$ and eigenvalues $p_1 \geq p_2 \geq \ldots \geq p_d$ and $\tilde{p}_1 \geq \tilde{p}_2 \geq \ldots \geq \tilde{p}_d$. The spectrum of $\rho$ majorizes the spectrum of $\tilde{\rho}$, denoted as $\rho \prec_M \tilde{\rho}$, iff for all $k \in \{1, 2, \ldots, d\}$
\begin{equation}
\sum_{i=1}^{k} p_i \geq \sum_{i=1}^{k} \tilde{p}_i \tag{B1}
\end{equation}
with equality for $k = d$.

Note that to avoid confusion with Lieb and Yngvason’s order relation later, we use a non-standard notation for the majorization $\prec_M$, which in particular differs from the notational convention from Bahtia [39].

Majorization can just as well be expressed by means of the step functions $f_\rho$ introduced in the main text:
\begin{equation}
\rho \prec_M \tilde{\rho} \iff \int_0^k f_\rho(x)dx \geq \int_0^k f_{\tilde{\rho}}(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \tag{B2}
\end{equation}

As $f_\rho(x)$ is monotonically decreasing in $x$ and due to the normalization $\int_0^\infty f_\rho(x)dx = 1$, the condition $k \in \{1, 2, \ldots, d\}$ from Definition 6 is equivalently replaced with $k \in \mathbb{R}_{\geq 0}$.

Noisy operations and majorization are furthermore related to the unital operations [44–46].

Definition 7. A unital quantum operation on $S(\mathcal{H})$ is a completely positive trace preserving map (CPTPM) that preserves the identity operator $\mathbb{I}_{\dim \mathcal{H}}$.

Proposition 8. For two density operators $\rho$ and $\tilde{\rho} \in S(\mathcal{H})$ the following are equivalent:

- There exists a noisy operation achieving the transition $\rho \rightarrow \tilde{\rho}$.
- There exists a unital quantum operation achieving the transition $\rho \rightarrow \tilde{\rho}$.
- The spectrum of $\rho$ majorizes the spectrum of $\tilde{\rho}$, denoted by $\rho \prec_M \tilde{\rho}$.
A proof can be found for instance in [44, 45].

The resource theory of noisy operations has numerous monotonic functions [44]. One of the most popular families of monotones under these operations are the so-called Rényi entropies [34].

**Definition 9.** The $\alpha$-Rényi entropy of a density operator $\rho \in S(\mathcal{H})$ is defined as

$$H_\alpha(\rho) = \frac{1}{1-\alpha} \log \text{tr}(\rho^\alpha). \quad (B3)$$

For $\alpha \to \infty$ and $\alpha = 0$ we recover two quantities from the smooth entropy framework, the min-entropy $H_{\min}$ and the max-entropy $H_{\max}$ [25, 47]. Note that taking the limit $\alpha \to 1$ leads to the von Neumann entropy $H = -\text{tr}(\rho \log \rho)$.

**Definition 10.** For a density operator $\rho \in S(\mathcal{H})$ the **min- and max-entropies** are defined as

$$H_{\min}(\rho) = -\log \|\rho\|_\infty \quad (B4)$$
$$H_{\max}(\rho) = \log \text{rank} \rho, \quad (B5)$$

where $\|\rho\|_\infty$ denotes the maximal eigenvalue of $\rho$.

Note that our definition does not coincide with the usual terminology [47], instead we resume the notation from [25], where $H_{\max}$ is defined as $H_0$. Our non-standard choice is well justified, as we regard the max-entropy as a physical concept, which can be quantified by different entropy measures all attaining very similar values.

2. The resource theory of thermal operations

The resource theory of thermal operations describes quantum systems interacting with a heat bath at a temperature $T$ [13, 15, 48]. The allowed operations in this framework are:

- Addition of an ancillary system in a thermal state relative to the heat bath.
- Application of any unitary operation which commutes with the total Hamiltonian on system and ancilla.
- Removal of any subsystem (by taking the partial trace).

Thermal states, also called Gibbs states, are of the form

$$\tau = \sum_i \frac{e^{-\beta E_i}}{Z} |E_i\rangle \langle E_i|, \quad (B6)$$

where $Z$ is the partition function and the $E_i$ denote the energy eigenstates; the constant $\beta = \frac{1}{k_B T}$ is inversely proportional to the temperature $T$ and $k_B$ denotes the Boltzmann constant. These states are preserved under thermal operations, while all athermal states are resource states [13, 15]. Note that for a system with a trivial Hamiltonian, i.e. a system where all energy levels are degenerate, the resource theory of thermal operations is equivalent to the resource theory of noisy operations.

Closely connected to the resource theory of thermal operations is the order relation of thermo-majorization, which is defined as a Gibbs-rescaled majorization [15, 16].

**Definition 11.** Let $\rho = \sum_i p_i |E_i\rangle \langle E_i| \in S(\mathcal{H})$ be a density matrix block diagonal in the energy eigenbasis and let $d = \dim \mathcal{H}$. Represent its spectrum as a step function

$$f_\rho(x) = \begin{cases} p_i & i-1 \leq x \leq i \\ 0 & \text{otherwise} \end{cases} \quad (B7)$$

with eigenvalues $p_1 \geq p_2 \geq \ldots \geq p_d$. The **Gibbs-rescaled** version of this step function is given as

$$f_\rho^T(x) = \begin{cases} \frac{p_i}{e^{\beta E_k}} & \sum_{k=1}^{i-1} e^{-\beta E_k} \leq x \leq \sum_{k=1}^{i} e^{-\beta E_k} \\ 0 & \text{otherwise} \end{cases} \quad (B8)$$

with reordered eigenvalues $\frac{p_1}{e^{\beta E_k}} \geq \frac{p_2}{e^{\beta E_k}} \geq \ldots \geq \frac{p_d}{e^{\beta E_k}}$.

Thermo-majorization can be defined in terms of Gibbs-rescaled step functions, analogous to the formulation of majorization $\prec_M$ in Equation (B2).

**Definition 12.** Let $\rho$ and $\sigma \in S(\mathcal{H})$ be two quantum states which are block diagonal in the energy eigenbasis. We define the order relation of **thermo-majorization** $\prec_T$ as

$$\rho \prec_T \sigma \iff \int_0^k f_\rho^T(x) dx \geq \int_0^k f_\sigma^T(x) dx \quad \forall k \in \mathbb{R}_{\geq 0}. \quad (B9)$$

Note that this definition is different but equivalent to the one in [15].

Horodecki and Oppenheim [15] have shown that thermal operations can be characterized in terms of this order relation of thermo-majorization $\prec_T$, at least for states that are block diagonal in the energy eigenbasis.¹

**Proposition 13.** Let $\rho$ and $\sigma \in S(\mathcal{H})$ be two states which are (block) diagonal in the energy eigenbasis. Then there exists a thermal operation achieving the transition $\rho \rightarrow \sigma$ if and only if the state $\rho$ thermo-majorizes the state $\sigma$, denoted as $\rho \prec_T \sigma$.

¹ Note that Horodecki and Oppenheim extended this result to initial states $\rho$ that are not block diagonal in the energy eigenbasis. A thermal operation $\rho \rightarrow \sigma$ is then equivalent to an ordering $\rho_{\text{deph}} \prec_T \sigma$, where $\rho_{\text{deph}}$ denotes the state $\rho$ dephased in the energy eigenbasis. The reason for this is that thermal operations commute with a dephasing operation in the energy eigenbasis. Therefore, the transitions $\rho_{\text{deph}} \rightarrow \sigma$ is equivalent to $\rho \rightarrow \sigma_{\text{deph}} = \sigma$. 
As shown by Brandao et al. [20], a family of measures which are monotonic under thermal operations for block diagonal states $\rho$ and for all $\alpha \geq 0$ is given as

$$F_\alpha(\rho) = k_B T D_\alpha(\rho|\tau) \ln(2) + F(\tau), \quad (B10)$$

where $\tau$ is the thermal state of the system and $F(\tau) = -k_B T \ln Z_\tau$ its free energy. The R"enyi divergences $D_\alpha(\rho|\tau)$, $\alpha \geq 0$ for states $\rho$ and $\tau$ which commute are

$$D_\alpha(\rho|\tau) = \frac{1}{\alpha - 1} \log \sum_i p_i^\alpha t_i^{1-\alpha}, \quad (B11)$$

where $p_i$ are the eigenvalues of $\rho$ and $t_i$ are the eigenvalues of $\tau$. These measures contain for $\alpha = 0$ the $F_{\text{min}}$ and in the limit $\alpha \to \infty$ the $F_{\text{max}}$, given as

$$F_{\text{min}}(\rho) = k_B T D_0(\rho|\tau) \ln(2) + F(\tau) \quad (B12)$$

$$F_{\text{max}}(\rho) = k_B T D_\infty(\rho|\tau) \ln(2) + F(\tau), \quad (B13)$$

where $D_0(\rho|\tau) = -\log_2 \text{Tr} \rho \tau$ and $D_\infty(\rho|\tau) = \log_2 \min \left\{ \lambda : \rho \leq \lambda \tau \right\}$ correspond to the relative entropies, introduced in [36, 49], with respect to the thermal state of the system. Note that for a thermal state $\tau$ the equality $F_{\text{min}}(\tau) = F(\tau) = F_{\text{max}}(\tau)$ holds.

Horodecki and Oppenheim [15] originally introduced these two quantities to describe the extractable work as well as the work needed to form a state. Assuming to have no errors for these two processes and that the states $\rho \in \mathcal{S}(\mathcal{H})$ are diagonal in the energy eigenbasis, the extractable work under thermal operations is given as

$$W_{\text{ext}} = F_{\text{min}}(\rho) - F_{\text{min}}(\tau), \quad (B14)$$

whereas the work of formation is

$$W_{\text{form}} = F_{\text{max}}(\rho) - F_{\text{max}}(\tau). \quad (B15)$$

In the thermodynamical limit Horodecki and Oppenheim recover the extractable work of a state $\rho \in \mathcal{S}(\mathcal{H})$ to be $W(\rho) = F(\rho) - F(\tau)$. In this limit, the same quantity is used to describe the work of formation.

### Appendix C: Lieb and Yngvason’s entropy for quantum states

Lieb and Yngvason’s framework is based on abstract axioms, which admits its application to other physical contexts. We apply their approach to quantum states, by considering the space $\mathcal{S}(\mathcal{H})$ of all density operators on a Hilbert space $\mathcal{H}$ as $\Gamma_{\text{ext}}$. The equilibrium states, forming the set $\Gamma_{\text{eq}}$, are defined as all those states characterized by a spectrum for which all non-zero eigenvalues are equal.

We show in the following how the noisy operations, characterized by the majorization relation $\prec_M$, arise as the information-theoretic counterpart to adiabatic accessibility in phenomenological thermodynamics.

We express the adiabatic processes including a device and a weight as introduced in Appendix A, denoted by $\rightarrow^A$, by concrete physical operations consisting of the steps:

- Adding a device to the system, formally expressed as taking the tensor product with an ancillary system in an equilibrium state.
- Letting the system and the device interact with the weight in an adiabatic, i.e. energy conserving, process.
- Removing the device in its initial state, which means taking the partial trace of the ancillary system in the state it has been added initially.

To describe the weight, a system which according to Lieb and Yngvason’s framework should only change its height, i.e. provide or remove energy, we rely on a particular model by Aber [38] describing a coherent weight $W$ with a Hamiltonian

$$H_W = s \sum_w |w\rangle \langle w| \quad (C1)$$

corresponding to an energy ladder, where the $\{|w\rangle\}_w$ are orthonormal states and the constant $s \in \mathbb{R}_{\geq 0}$ defines the energy level spacing of the Hamiltonian. The weight, assumed to be in a state $\sigma = |\eta_{l_0}\rangle \langle \eta_{l_0}|$ with $|\eta_{l_0}\rangle = \frac{1}{\sqrt{l}} \sum_{w=0}^{l-1} |w + l_0\rangle$, is connected to a quantum system $S$ with Hamiltonian $H_S$, in our case consisting of the system and the ancilla. We allow operations on the system and the weight which commute with translations of the weight, i.e. do not depend on the absolute energetic state of the weight, and which at the same time conserve the total energy, i.e. commute with $H_S \otimes \mathbb{1}_W + \mathbb{1}_S \otimes H_W$. This realizes the idea that only the relative change in energy of the weight, being the energy that is added to or removed from system $S$, influences the system, giving the weight the role of a work reservoir. For large $L$, these operations on the system and the weight allow the implementation of any unitary operation on the system $S$ alone, catalytically. Thus, the interaction of system and ancilla with the weight reduces to the application of arbitrary unitaries to the system and the ancilla. Intuitively, this weight can be imagined as eliminating the role of energy from the framework or just as allowing to change the Hamiltonian at will.

Compared to other models of weights [19] this particular model allows us to obtain superpositions of energy eigenstates of a system which was initially in such an eigenstate. It thus allows us to include the treatment of states which are not block diagonal in the energy eigenbasis, which could not be created otherwise.

The processes $\rightarrow^A$ resemble intuitively the noisy operations introduced in Appendix B 1. However, the processes $\rightarrow^A$ allow the addition of any equilibrium ancilla yet require us to return the ancillas in their initial state.
The following proposition shows that these processes are also characterized by the majorization relation \( \preceq_M \), and are thus equivalent to noisy operations.

Proposition 14. For two states \( \rho \) and \( \sigma \in \mathcal{S}(\mathcal{H}_S) \) the following are equivalent:

- (A): The spectrum of \( \rho \) majorizes the spectrum of \( \sigma \), i.e. \( \rho \prec_M \sigma \).
- (B): There exists a process \( \rho \xrightarrow{A} \sigma \).

Proof. (A) \( \Rightarrow \) (B): Horodecki and Oppenheim [37] show that if \( \rho \prec_M \sigma \), then there exists a noisy operation bringing \( \rho \) to \( \sigma \), i.e. there exists a unitary \( U_{SA} \) acting on an additional ancillary system \( A \) such that

\[
\text{tr}_A \left( U_{SA} \left( \rho \otimes \frac{1}{\text{dim}(\mathcal{H}_A)} I_A \right) U_{SA}^\dagger \right) = \sigma. \tag{C2}
\]

where \( I_A \in \mathcal{S}(\mathcal{H}_A) \) is a maximally mixed ancilla and \( U_{SA}^\dagger \) denotes the adjoint of \( U_{SA} \). In the noisy operations that Horodecki and Oppenheim construct explicitly, one can see that the unitary \( U_{SA} \) does not change the reduced state on the ancillary system, i.e.

\[
\text{tr}_A \left( U_{SA} \left( \rho \otimes \frac{1}{\text{dim}(\mathcal{H}_A)} \right) U_{SA}^\dagger \right) = \frac{1}{\text{dim}(\mathcal{H}_A)}. \tag{C3}
\]

Thus, the ancilla is removed in the maximally mixed state in which it was added and the process is an adiabatic process \( \rho \xrightarrow{A} \sigma \) according to our definition.

(B) \( \Rightarrow \) (A): As we know from Proposition 8, \( \rho \prec_M \sigma \) is equivalent to the existence of a unital map from \( \rho \) to \( \sigma \). In the following, we show that the processes \( \rho \xrightarrow{A} \sigma \) are unital and thus also imply the ordering \( \rho \prec_M \sigma \). Now let \( \chi \in \mathcal{S}(\mathcal{H}_A) \) be a state with a flat spectrum, i.e. it can be written as \( \chi = \sum_{l=1}^d \frac{1}{2} |l\rangle \langle l| \) with \( d \leq \text{dim}(\mathcal{H}_A) \). Our operations \( \xrightarrow{A} \) are the subset of the operations \( \text{tr}_A(U_{SA}(\cdot) \otimes \chi)U_{SA}^\dagger \) acting on a state \( \rho \), where \( U_{SA} \) denotes an arbitrary unitary, for which the partial trace removes the reduced state \( \chi \), i.e. \( \text{tr}_S \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \right) U_{SA}^\dagger \right) \chi. \)

Consider the function \( h(\rho) = -\text{tr}(\rho \log \rho) \). Now look at the maximally mixed state \( \rho = \frac{1}{\text{dim}(\mathcal{H}_S)} I_{SA} \). We know that

\[
\begin{align*}
\rho(\rho \otimes \chi) &= h \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) \\
&= h \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) U_{SA}^\dagger \right) \leq h \left( \text{tr}_A \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) U_{SA}^\dagger \right) \right) + h \left( \text{tr}_S \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) U_{SA}^\dagger \right) \right). \tag{C4}
\end{align*}
\]

The inequality follows by subadditivity of \( h \). We know

\[
\left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) = \log(\text{dim}(\mathcal{H}_S)) \cdot d. \tag{C5}
\]

Since the ancillary system has to be in state \( \chi \) at the end of the process,

\[
\left( \text{tr}_A \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) U_{SA}^\dagger \right) \right) = \log d. \tag{C6}
\]

From (C4) we conclude that

\[
\text{tr}_A \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) U_{SA}^\dagger \right) \geq \log \text{dim}(\mathcal{H}_S). \tag{C7}
\]

As this inequality can only be satisfied if \( \text{tr}_A \left( U_{SA} \left( \frac{1}{\text{dim}(\mathcal{H}_S)} \otimes \chi \right) U_{SA}^\dagger \right) = \frac{1}{\text{dim}(\mathcal{H}_S)} \), the processes \( \xrightarrow{A} \) are unital.

Recall that for two states \( \rho \) and \( \tilde{\rho} \in \mathcal{S}(\mathcal{H}) \) the majorization condition \( \rho \prec_M \tilde{\rho} \) can be equivalently expressed in terms of the spectral step functions \( f_\rho \) and \( f_{\tilde{\rho}} \), as introduced in (B2). Even though the functions \( f_\rho \) are not in one-to-one correspondence with the states \( \rho \) but rather represent all states having a certain spectrum, this description is enough for this purpose. As the corresponding processes \( \xrightarrow{A} \) include the application of an arbitrary unitary, it is perspicuous that the order relation \( \prec_M \) is independent of the eigenbases of the compared states.

For an equilibrium state \( \rho \), which has a flat spectrum, the step function \( f_\rho \) has the simple form

\[
f_\rho(x) = \begin{cases} 1 & \text{rank } \rho \\ 0 & \text{otherwise} \end{cases}, \tag{C8}
\]

Thus, for two equilibrium states \( \rho \) and \( \tilde{\rho} \) the rank alone determines which one majorizes the other, as

\[
\begin{align*}
\rho \prec_M \tilde{\rho} \iff & \int_0^k f_\rho(x) \text{d}x \geq \int_0^k f_{\tilde{\rho}}(x) \text{d}x \quad \forall \ k \in \mathbb{R}_\geq 0 \\
& \int_0^k \frac{1}{\text{rank } \rho} \text{d}x \geq \int_0^k \frac{1}{\text{rank } \tilde{\rho}} \text{d}x \quad 0 \leq k \leq \text{rank } \rho \\
& \text{rank } \rho \leq \text{rank } \tilde{\rho}. \tag{C9}
\end{align*}
\]

---

2 Note that in order to achieve this an arbitrarily large ancillary system has to be allowed.

3 Note that even though this function corresponds to the von Neumann entropy, we regard it as a purely mathematical function with the mathematical properties that it is subadditive, unitarily invariant, additive for product states, and it reaches its maximum of \( \log \text{dim}(\mathcal{H}) \) if and only if \( \rho \) is maximally mixed. The reason why we consider this quantity is that we know rather well how it behaves; however any other mathematical function that satisfies these properties could have been used instead.
We define the composition of two states \( \rho \in S(\mathcal{H}) \) and \( \tilde{\rho} \in S(\mathcal{H}) \) as their tensor product \( \rho \otimes \tilde{\rho} \in S(\mathcal{H} \otimes \mathcal{H}) \). The scaling of an equilibrium state \( \rho \) is assumed to coincide with its composition for scaling factors \( \lambda \in \mathbb{N} \). The step function of a scaled state \( \lambda \rho \in \lambda S(\mathcal{H}) \), which is thus defined as \( \rho^{\otimes \lambda} \in S(\mathcal{H}^{\otimes \lambda}) \), is

\[
f_{\lambda \rho}(x) = \begin{cases} 
\left( \frac{1}{\text{rank} \rho} \right)^\lambda & 0 \leq x \leq (\text{rank} \rho)^\lambda \\
0 & \text{otherwise} \end{cases} \tag{C10}
\]

To obtain a continuous scaling operation, this way of scaling the step function is applied for any \( \lambda \). For most values of \( \lambda \) the scaled copies \( \lambda \rho \) do not represent physical states and no actual space \( \lambda S(\mathcal{H}) \) exists. However, any normalized, but possibly unphysical, function \( f(x) \), can be turned into a physical state by actually considering the function \( \frac{1}{\lambda} f(x/\lambda) \) for a large enough \( \lambda \): the step now coincides to a good approximation with an integer abscissa and the function now represents to a good approximation a physical state on a larger system. Now, notice that we can always combine states with a fully mixed state of a given rank, and that the following rules apply:

\[
\lambda \left( \rho, \frac{\| n \|}{n} \right) = \left( \lambda \rho, \frac{\| n \|}{n} \right); \tag{C11a}
\]

\[
\left( (\rho \otimes \frac{\| m \|}{m}), \tilde{\rho} \otimes \frac{\| m \|}{m} \right) = \left( (\rho, \tilde{\rho}), \frac{\| n \|}{n} \right). \tag{C11b}
\]

These rules are easily seen with the representation in terms of the step function. They allow now to give a precise signification to any relative statements between states which would be required to be scaled in an unphysical way: for example, we have \((\lambda \rho, \frac{1}{\lambda} n) \prec \sigma \) if and only if \((\lambda (\rho, \frac{1}{\lambda} n), \mu (\tilde{\rho}, \frac{1}{\lambda} m)) \prec \sigma, \frac{1}{\lambda} n, \frac{1}{\lambda} m \). Thus, if \( \lambda \rho \) does not actually correspond to a physical state, then the second expression should in fact be considered; indeed for large enough \( n \) the state \( \lambda (\rho, \frac{1}{\lambda} n) \) is actually physical to a good approximation.

The entropy function (A1) can thus be equivalently rewritten as

\[
S(\rho) = \sup \{ \lambda : (1 - \lambda) \rho_0, \lambda \rho_1 \prec \rho \} = \sup \{ \lambda : \left( 1 - \lambda \right) \left( \rho_0, \frac{\| n \|}{n} \right), \lambda \left( \rho_1, \frac{\| n \|}{n} \right) \prec \left( \rho, \frac{\| n \|}{n} \right) \}. \tag{C12}
\]

The last expression, for \( n \) large enough, only involves physical states.

We now proceed to show that this order relation along with its scaling operation fulfills Lieb and Yngvason’s axioms.

**Proposition 15.** Consider the majorization relation \( \prec_M \) between the ordered spectra of states \( \rho \in S(\mathcal{H}) \), and define the composition operation as well as the scaling of non-equilibrium states as introduced above. Then, for equilibrium states, i.e. states with a flat spectrum, the six axioms E1 to E6 as well as the Comparison Hypothesis hold. Moreover, for non-equilibrium states axioms N1 and N2 are satisfied.

**Proof.** As axiom N2 requires that axioms E1 to E3 as well as E6 also hold for non-equilibrium states, we directly show that they hold for non-equilibrium states as well.

**Reflexivity (E1):** \( \prec_M \) is clearly reflexive: For \( \rho \in S(\mathcal{H}) \)

\[
\int_0^k f_\rho(x) dx = \int_0^k f_\rho(x) dx \quad \forall \, k \in \mathbb{R}_{\geq 0} \tag{C13}
\]

and \( \rho \prec_M \rho \).

**Transitivity (E2):** Let \( \rho, \sigma \) and \( \chi \in S(\mathcal{H}) \). Then, for \( l = 1, 2, \ldots, \dim(\mathcal{H}) \dim(\mathcal{H}') \) one can find \( m_1 \geq m_2 \geq \ldots \geq m_{\dim(\mathcal{H})} \geq 0 \) such that

\[
\int_0^k f_{(\rho, \sigma)}(x) dx = \sum_{j=1}^{\dim(\mathcal{H}')} q_j \int_0^{m_j} f_\rho(x) dx \tag{C15}
\]

for \( l = 1, 2, \ldots, \dim(\mathcal{H}) \dim(\mathcal{H}') \). Then one can find \( m_1 \geq m_2 \geq \ldots \geq m_{\dim(\mathcal{H})} \geq 0 \) such that

\[
\int_0^k f_{(\rho, \sigma)}(x) dx = \sum_{j=1}^{\dim(\mathcal{H}')} q_j \int_0^{m_j} f_\rho(x) dx \tag{C16}
\]
The inequality holds as \( \rho \prec_M \rho' \). One can now find \( n_1 \geq n_2 \geq \ldots \geq n_{\dim(H)} \geq 0 \) such that this sum can be rewritten in terms of integrals in \( f_\sigma \) as

\[
\sum_{j=1}^{\dim(H')} q_j \int_0^{a_j} f_{\rho'}(x) dx = \sum_{i=1}^{\dim(H)} p_i \int_0^{a_i} f_\sigma(x) dx \\
\leq \sum_{i=1}^{\dim(H)} p_i \int_0^{a_i} f_{\sigma'}(x) dx \quad \text{(C17)}
\]

The first inequality holds as \( \rho \prec_M \rho' \) and the second one is fulfilled as the \( \{n_i\} \) might not select the highest \([k]\) eigenvalues \( x'_i = p'_i / q'_i \) of \( \rho' \otimes \sigma' \).

**Stability (E6):** Note that \( \chi_0 \) and \( \chi_1 \) are necessarily equilibrium states, as in Lieb and Yngvason’s framework only those can be scaled. Let \( \rho \in \mathcal{S}(H) \) have eigenvalues \( p_1 \geq p_2 \geq \ldots \geq p_{\dim(H)} \). Assume that \( (\rho, \varepsilon \chi_0) \prec_M (\sigma, \varepsilon \chi_1) \) for a sequence of \( \varepsilon \)'s tending to zero. Let this sequence be denoted by \( (\varepsilon_i) \). Then for all \( \varepsilon_i \) we have

\[
\int_0^k f_{(\rho, \varepsilon \chi_0)}(x) dx \geq \int_0^k f_{(\sigma, \varepsilon \chi_1)}(x) dx \quad \forall k \in \mathbb{R}_{\geq 0}.
\]

Taking the limit \( i \to \infty \) leads to

\[
\lim_{i \to \infty} \int_0^k f_{(\rho, \varepsilon \chi_0)}(x) dx \geq \lim_{i \to \infty} \int_0^k f_{(\sigma, \varepsilon \chi_1)}(x) dx \quad \forall k \in \mathbb{R}_{\geq 0},
\]

and thus

\[
\int_0^k \lim_{i \to \infty} f_{(\rho, \varepsilon \chi_0)}(x) dx \geq \int_0^k \lim_{i \to \infty} f_{(\sigma, \varepsilon \chi_1)}(x) dx \quad \forall k \in \mathbb{R}_{\geq 0},
\]

which follows by dominated convergence. Note that

\[
f_{(\rho, \varepsilon \chi_0)}(x) = \begin{cases} P \cdot \left( \frac{1}{\rank \chi_0} \right)^{\varepsilon_i} & (l - 1) \cdot (\rank \chi_0)^{\varepsilon_i} \leq x \leq l \cdot (\rank \chi_0)^{\varepsilon_i} \\ 0 & \text{otherwise} \end{cases}
\]

and similar for \( f_{(\sigma, \varepsilon \chi_1)} \). Thus, taking the limit leads to

\[
\int_0^k f_{\rho}(x) dx \geq \int_0^k f_{\sigma}(x) dx \quad \forall k \in \mathbb{R}_{\geq 0},
\]

which is equivalent to \( \rho \prec_M \sigma \).

Since for equilibrium states the rank alone suffices to assert majorization, it is straightforward to see that axioms E4 and E5 as well as the Comparison Hypothesis hold:

**Scaling invariance (E4):** We see that

\[
\rho \prec_M \sigma \iff \rank \sigma \geq \rank \rho \\
\iff (\rank \sigma)^{\lambda} \geq (\rank \rho)^{\lambda} \quad \forall \lambda > 0 \quad \text{(C23)}
\]

**Splitting and recombination (E5):** Let \( 0 < \lambda < 1 \). Then,

\[
f_{(\lambda \rho, (1 - \lambda) \rho)}(x) = \begin{cases} \left( \frac{\rank \rho}{\rank \rho} \right)^{\lambda} \left( \frac{\rank \rho}{\rank \rho} \right)^{1 - \lambda} & 0 \leq x \leq (\rank \rho)^{\lambda} (\rank \rho)^{1 - \lambda} \\ 0 & \text{otherwise} \end{cases}
\]

\[
= \begin{cases} \left( \frac{\rank \rho}{\rank \rho} \right)^{\lambda} \left( \frac{\rank \rho}{\rank \rho} \right)^{1 - \lambda} & 0 \leq x \leq \rank \rho \\ 0 & \text{otherwise} \end{cases}
\]

\[
= f_{\rho}(x).
\]

**Comparison Hypothesis:** Let \( 0 \leq \lambda \leq 1 \) and \( 0 \leq \mu \leq 1 \) and let \( \rho, \tilde{\rho}, \sigma, \tilde{\sigma} \in \mathcal{S}(H) \) be arbitrary equilibrium states. Then the object \((\lambda \rho, (1 - \lambda) \sigma)\) with step function

\[
f_{(\lambda \rho, (1 - \lambda) \sigma)}(x) = \begin{cases} \left( \frac{\rank \rho}{\rank \sigma} \right)^{\lambda} \left( \frac{\rank \rho}{\rank \sigma} \right)^{1 - \lambda} & 0 \leq x \leq (\rank \rho)^{\lambda} (\rank \sigma)^{1 - \lambda} \\ 0 & \text{otherwise} \end{cases}
\]

\[
\text{(C25)}
\]
Axiom (N1): Choose $\rho_0$ to be a pure state and $\rho_1 = \frac{1}{\dim(H)}$ maximally mixed. Then, for any state $\rho \in S(H)$ we know that $\rho_0 \preceq_M \rho$ and $\rho \preceq_M \rho_1$. □

Corollary 16. For a system with a trivial Hamiltonian on which processes $A$ can be performed, there exists a unique entropy function $S$ for equilibrium states, as well as bounds $S_-$ and $S_+$ on the entropy of non-equilibrium states, all up to an affine transformation.

Proof. Proposition 15 allows the application of Lieb and Yngvason’s Theorem 4 and Proposition 5 to quantum states, all up to an affine transformation. □

Proposition 17. For an equilibrium state $\rho$ the unique entropy function $S(\rho)$, defined in Equation (A3), corresponds to the von Neumann entropy $H(\rho) = \log \text{rank} \rho$ up to affine transformations.

Proof. For $\lambda = S(\rho)$ we have

\[
((1-\lambda)\rho_0, \lambda \rho_1) \sim_M \rho \iff \int_0^k f_0((1-\lambda)\rho_0, \lambda \rho_1)(x)dx = \int_0^k f_\rho(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \tag{C26}
\]

Exploiting that $\rho_0$, $\rho$ and $\rho_1$ are equilibrium states, for which the ordering $\preceq_M$ only depends on their rank, this is equivalent to

\[
(rank \rho_0)^{1-\lambda}(rank \rho_1)^{\lambda} = \text{rank} \rho, \tag{C27}
\]

which can be reformulated as

\[
\lambda = \frac{1}{\log \text{rank} \rho_1} \log \text{rank} \rho - \frac{1}{\log \text{rank} \rho_0} \log \text{rank} \rho_0 \tag{C28}
\]

\[
= a \cdot \log \text{rank} \rho + b.
\]

The parameters $a$ and $b$ are defined as $a = \frac{1}{\log \text{rank} \rho_1}$, $b = -\frac{1}{\log \text{rank} \rho_0} \log \text{rank} \rho_0$ and depend on the gauge states $\rho_0$ and $\rho_1$. □

In particular, $S(\rho) = H(\rho)$ is obtained with the choice $\rho_0 = 1$ and rank $\rho_1 = 2$. As Lieb and Yngvason [22, 23] have defined the handling of negative scaling factors, this choice of $\rho_0$ and $\rho_1$ is not problematic in order to reasonably define the entropy for an equilibrium state $\rho$ obeying $\rho_1 \preceq_M \rho$: such a state $\rho$ is associated with a negative scaling factor $(1-\lambda)$ and thus characterized by an entropy larger than 1.

Proposition 18. The quantities $\tilde{S}_-$ and $\tilde{S}_+$ from (A6) and (A7) correspond to the information theoretic $H_{\min}$ and $H_{\max}$ defined in (B4) and (B5).

Proof. For any state $\rho \in S(H)$ with eigenvalues $p_1 \geq p_2 \geq \ldots \geq p_{\dim(H)}$ and for any equilibrium states $\rho_0 \preceq_M \rho_1$, let $\lambda$ be such that $((1-\lambda)\rho_0, \lambda \rho_1) \sim_M \rho$. Then,

\[
\int_0^k f_0((1-\lambda)\rho_0, \lambda \rho_1)(x)dx \geq \int_0^k f_\rho(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \tag{C29}
\]

Let $\tilde{k} = (rank \rho_0)^{1-\lambda}(rank \rho_1)^{\lambda}$. As $\rho_0$ and $\rho_1$ are equilibrium states, we know that for $0 \leq k \leq \tilde{k}$,

\[
\int_0^k f_0((1-\lambda)\rho_0, \lambda \rho_1)(x)dx = \int_0^k \left( \frac{1}{\text{rank} \rho_0} \right)^{1-\lambda} \left( \frac{1}{\text{rank} \rho_1} \right)^{\lambda} dx. \tag{C30}
\]

Thus, by considering $k \leq 1$, Equation (C29) directly implies

\[
\left( \frac{1}{\text{rank} \rho_0} \right)^{1-\lambda} \left( \frac{1}{\text{rank} \rho_1} \right)^{\lambda} \geq p_1, \tag{C31}
\]

which can be rewritten as

\[
a \cdot \log \frac{1}{p_1} + b \geq \lambda, \tag{C32}
\]

with $a = \frac{1}{\log \text{rank} \rho_1}$ and $b = -\frac{1}{\log \text{rank} \rho_0} \log \text{rank} \rho_0$. On the other hand, Equation (C31) implies

\[
\left( \frac{1}{\text{rank} \rho_0} \right)^{1-\lambda} \left( \frac{1}{\text{rank} \rho_1} \right)^{\lambda} \min \left\{ k, \tilde{k} \right\} \geq p_1 \cdot \min \left\{ k, \frac{1}{p_1} \right\} \geq \int_0^k f_\rho(x)dx. \tag{C33}
\]

for all $k \in \mathbb{R}_{\geq 0}$, i.e. implies (C29); the second inequality follows as the step function $f_\rho(x)$ is monotonously decreasing and normalised. Thus, taking the supremum over $\lambda$ in (C32) concludes the proof for $\tilde{S}_+$, as $H_{\min}(\rho) = -\log \|\rho\|_{\infty}$, where $\|\rho\|_{\infty}$ denotes the maximal eigenvalue of the state $\rho$, i.e. equals $p_1$.

For $\tilde{S}_-$ we proceed similarly. Let $\rho \in S(H)$ and let $\rho_0 \preceq_M \rho_1$ be two equilibrium states, as above. Now let $\lambda$ be such that $\rho \preceq_M ((1-\lambda)\rho_0, \lambda \rho_1)$ and thus

\[
\int_0^k f_0((1-\lambda)\rho_0, \lambda \rho_1)(x)dx \leq \int_0^k f_\rho(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \tag{C34}
\]

First, we show that

\[
(rank \rho_0)^{1-\lambda}(rank \rho_1)^{\lambda} \geq \text{rank} \rho \tag{C35}
\]

by contradiction. Assume for now that $(rank \rho_0)^{1-\lambda}(rank \rho_1)^{\lambda} < \text{rank} \rho$. For $\tilde{k} =$
(rank ρ₀)₁−λ(rank ρ₁)λ, we therefore find
\[ \int_0^k \rho f_\rho(x)dx < 1. \] (C36)

This contradicts Equation (C34), as
\[ \int_0^k \rho f((1-λ)ρ₀, λρ₁)dx = 1. \] (C37)

Thus we have \( k \geq \text{rank } ρ \), which can be rewritten as
\[ λ \geq a \cdot \log \text{rank } ρ + b \] (C38)

with \( a \) and \( b \) defined as above. Moreover, (C35) implies
\[
\left( \frac{1}{\text{rank } ρ₀} \right)^{1-λ} \left( \frac{1}{\text{rank } ρ₁} \right)^λ \min \{ k, \tilde{k} \} \leq \frac{1}{\text{rank } ρ} \min \{ k, \text{rank } ρ \} \leq \int_0^k f_\rho(x)dx \] (C39)

for all \( k \in \mathbb{R}_{>0} \), i.e. implies (C34); the second inequality holds as \( f_\rho(x) \) is monotonously decreasing and normalised. As \( H_{\text{max}}(ρ) = \log \text{rank } ρ \), taking the infimum over \( λ \) in (C38) concludes the proof. \( \Box \)

In the case of quantum states considered here, the bounds \( \{ \tilde{S}_-, \tilde{S}_+ \} \) do not equal Lieb and Yngvason’s original \( \{ S_-, S_+ \} \). This can be most easily shown with the following example.

**Example.** Consider a qubit in the state \( ρ = \frac{3}{4} |0\rangle \langle 0| + \frac{1}{2} |1\rangle \langle 1| \). Take \( ρ₀ = |0\rangle \langle 0| \) and \( ρ₁ = \frac{1}{2} |0\rangle \langle 0| + \frac{1}{2} |1\rangle \langle 1| \). Then
\[ \tilde{S}_-(ρ) = \sup \{ λ : ((1-λ)ρ₀, λρ₁) ≺ ρ \} = \log \frac{4}{3}, \] (C40)

whereas
\[ S_-(ρ) = \sup \{ λ : ((1-λ)ρ₀, λρ₁) ≪ ρ' \} < ρ' ≺ ρ \} = \log 1 = 0, \] (C41)
as \( ρ' = ρ₀ \) is the next equilibrium state from which \( ρ \) is accessible, which is visualized in Figure 2 of the main text.

The intuitive explanation is that in the case of quantum states — contrary to thermodynamics — there does not exist an equilibrium state for each value of \( λ \), but for a quantum system of a particular size, the equilibrium states are discretely distributed and give only discrete entropy values. Note that for \( \tilde{S}_- \) and \( \tilde{S}_+ \) no such difference is observed.

The quantities \( \tilde{S}_- \) and \( \tilde{S}_+ \) are reasonable, as for two states \( ρ, σ \in S(H) \) they have the desirable properties
\[ \tilde{S}_-(ρ ⊗ σ) = \tilde{S}_-(ρ) + \tilde{S}_-(σ), \] (C42)
\[ \tilde{S}_+(ρ ⊗ σ) = \tilde{S}_+(ρ) + \tilde{S}_+(σ). \] (C43)

Thus, if \( σ \) is an equilibrium state, we observe
\[ \tilde{S}_-(ρ ⊗ σ) = \tilde{S}_-(ρ) + S(σ), \] (C44)

which for \( S_- \) is generally not the case. Moreover, \( \tilde{S}_- \) enables us to distinguish for instance a state \( ρ' = \frac{3}{4} |0\rangle \langle 0| + \frac{1}{2} |1\rangle \langle 1| \) from the state \( ρ \) considered in the previous example, while with \( S_- \) this is impossible.

**Appendix D: System interacting with a heat bath**

In typical laboratory experiments, the systems of interest interact with a thermal environment. Considering systems connected to a heat reservoir is thus a natural and relevant extension of the scenario of an isolated system, which was described in Appendix C.

Assume that a heat bath at a temperature \( T \) is connected to an isolated system \( S \), shown in Figure 1 of the main text. As the system \( S \) now thermalizes with the reservoir, the equilibrium states are thermal states, as introduced in (B6), which can also be constrained to a subsystem; this means that the eigenvalues \( \{ p_i \} \) of such an equilibrium state \( τ = \sum p_i |E_i\rangle \langle E_i| \) are either \( e^{-βE_i}/Z \) or zero, where \( Z \) is the normalisation.

System \( S \) and reservoir \( R \) together still form an isolated system \( SR \), which we assume to undergo adiabatic processes with the aid of an ancillary system and a weight. For an isolated system such a process between two states of the system \( SR \) is expressed by means of the majorization relation \( ≪_M \). We follow Horodecki and Oppenheim’s treatment [15], that majorization for states of the system \( SR \) can be expressed as thermo-majorization \( ≪_τ \) on the corresponding states on \( S \), at least as long as the final state on \( S \) is block diagonal in the energy eigenbasis. Further, the system and the reservoir are assumed to be initially in a product state. The operations that correspond to this order relation \( ≪_τ \) are the thermal operations introduced in Appendix B.2.

As this treatment is only fully applicable as long as the compared states on \( S \) are block diagonal in the energy eigenbasis, the relation \( ≪_τ \) captures whether equilibrium states — which are always diagonal in the energy eigenbasis — can be interconverted. For those non-equilibrium states which are not diagonal in the energy eigenbasis, however, the order relation \( ≪_τ \) is not sufficient to express whether two states can be interconverted by a thermal operation. In the following, we therefore restrict our treatment to equilibrium states as well as to non-equilibrium states which are block diagonal in the energy eigenbasis.

---

4 More precisely, \( ≪_τ \) can only relate such non-equilibrium states to states which are block diagonal in the energy eigenbasis and only as long as the non diagonal state is the preceding element in the order. Consult [15] for further clarification.
The following proposition ensures that we can apply Lieb and Yngvason’s results to quantum states ordered with the relation $\prec_T$.

**Proposition 19.** Consider the order relation of thermo-majorization $\prec_T$. Then, for equilibrium states $\tau$ the six axioms E1 to E6 as well as the Comparison Hypothesis hold, whereas all block diagonal athermal states satisfy axioms N1 and N2.

**Proof.** By Definition 12, block diagonal states $\rho \in \mathcal{S}(\mathcal{H})$ can be represented by their Gibbs-rescaled step functions $f^\rho_T$. For equilibrium states $\tau$ the functions $f^\tau_T$ assume the simple form

$$f^\tau_T(x) = \begin{cases} \frac{1}{Z} & 0 \leq x \leq Z \\ 0 & \text{otherwise} \end{cases}, \quad (D1)$$

where $Z$ is the partition function considering all occupied states. We can define the composition of two arbitrary states as their tensor product, like before. For scaling factors $\lambda \in \mathbb{N}$ the scaling of an equilibrium state is again assumed to coincide with its composition. This scaling operation can be formally extended to any scaling factor $\lambda \in \mathbb{R}_{\geq 0}$ on the level of the functions $f^\tau_T$; the Gibbs-rescaled step function of a thermal state $\tau$ has the form

$$f^\tau_{\lambda\tau}(x) = \left(\frac{1}{Z}\right)^\lambda \frac{1}{Z^\lambda}dx \quad (D2)$$

Note that because these functions are flat, we can again, as we did in Appendix C, give a meaning to scaling with a non-integer factor $\lambda$ if we consider these states on a larger system.

The Gibbs-rescaled functions $f^\rho_T$ are normalized monotonically decreasing step functions like the $f_\rho$. Furthermore, they are flat for equilibrium states, where the partition function $Z$ alone determines which state thermo-majorizes the other; for two equilibrium states $\tau$ and $\tilde{\tau}$

$$\tau \prec_T \tilde{\tau} \Leftrightarrow \int_0^k f^\tau_T(x)dx \geq \int_0^k f^{\tilde{\tau}}_T(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}$$

$$\Leftrightarrow \int_0^k \frac{1}{Z^\tau}dx \geq \int_0^k \frac{1}{Z^{\tilde{\tau}}}dx \quad 0 \leq k \leq Z^{\tau}$$

$$\Leftrightarrow Z^{\tilde{\tau}} \geq Z^{\tau}. \quad (D3)$$

Thus for the $f^\rho_T$ the partition function takes the role of the rank in $f^\rho_\rho$.

Substituting the $f_\rho$ with the $f^\rho_T$ and the rank with the partition function $Z$, we can apply the proof of Proposition 15 for the majorization relation $\prec_M$ to prove the axioms E1 to E6 as well as the Comparison Hypothesis for the order relation of thermo-majorization $\prec_T$. Note that when adapting the proof of E3, the choice of the $\{m_i\}$, and the $\{n_i\}$ is not problematic, as all energies are positive and thus the factors $e^{-\beta E_i}$ are always smaller or equal to one.

Axiom N1 holds, as a state $\rho = \sum_i p_i |E_i\rangle \langle E_i|$ in $\mathcal{S}(\mathcal{H})$ always thermo-majorizes the equilibrium state $\tau_1 = \sum_i \frac{e^{-\beta E_i}}{Z^\tau_1} |E_i\rangle \langle E_i|$ and is thermo-majorized by the equilibrium state $\tau_0 = |E_1\rangle \langle E_1|$. \qed

As all axioms are satisfied by the order relation $\prec_T$, Theorem 4 implies that there is a unique additive and extensive function

$$S_T(\rho) = \{\lambda : ((1 - \lambda)\tau_0, \lambda\tau_1) \sim_T \tau\} \quad (D4)$$

which is monotonic under thermal operations.

**Proposition 20.** The unique function $S_T(\tau)$ for equilibrium states $\tau \in \mathcal{S}(\mathcal{H})$ is given by $S_T(\tau) = \ln Z_\tau$ up to affine transformations, where $Z_\tau$ is the partition function.

**Proof.** For $\lambda = S_T(\tau)$ we have

$$((1 - \lambda)\tau_0, \lambda\tau_1) \sim_T \tau \quad \Leftrightarrow \quad \int_0^k f_{((1 - \lambda)\tau_0, \lambda\tau_1)}^\tau(x)dx = \int_0^k f_T^\tau(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \quad (D5)$$

Recalling that for equilibrium states $\tau \prec_T \tilde{\tau} \Leftrightarrow Z_\tilde{\tau} \geq Z_\tau$ according to (D3), this is equivalent to

$$Z_1^{\tau_0 - \lambda} \sim Z_\lambda \tau_1 = Z_\tau \quad (D6)$$

and can be rewritten as

$$\lambda = \frac{1}{\ln \frac{Z_\tau}{Z_{\tau_0}}} \ln Z_\tau - \ln \frac{Z_\tau}{Z_{\tau_0}} \quad (D7)$$

$$= a_T \cdot \ln Z_\tau + b_T,$n

where $a_T = \frac{1}{\ln \frac{Z_\tau}{Z_{\tau_0}}}$ and $b_T = -a_T \cdot \ln Z_{\tau_0}$. This concludes the proof. \qed

The function $S_T(\tau)$ obtained for a system in contact with a heat bath is therefore equal to the Helmholtz free energy

$$F(\tau) = -k_B T \ln Z_\tau \quad (D8)$$

up to affine transformations. For a choice of two states with partition functions $Z_{\tau_0} = 1$ and $Z_{\tau_1} = e^{-\beta}$ one has exactly $S_T(\tau) = -k_B T \ln Z_\tau = F(\tau)$. Note that this choice of parameters obeys $\tau_0 \prec_T \tau_1$ instead of the usual $\tau_0 \prec_T \tau_1$. With this gauge the sign of $S_T$ is thus reversed and the quantity $F$ decreases under thermal operations.

The quantities $\tilde{S}_T^-$ and $\tilde{S}_T^+$, calculated for block diagonal non-equilibrium states, are not only bounds for the potential $F$, but are also directly related to the work of formation $F_{\text{max}}$ as well as to the extractable work $F_{\text{min}}$ introduced by Horodecki and Oppenheim [15] and described in Appendix B 2.
**Proposition 21.** For block diagonal states \( \rho \in \mathcal{S}(\mathcal{H}) \) the quantities \( \tilde{S}_{\uparrow} \) and \( \tilde{S}_{\uparrow\downarrow} \) correspond to the quantities \( F_{\text{max}} \) and \( F_{\text{min}} \) defined in Equations (B13) and (B12) up to affine transformations.

**Proof.** Let \( \rho \in \mathcal{S}(\mathcal{H}) \) be block diagonal state with rescaled eigenvalues \( p_1^{\text{res}} = \frac{p_1}{\rho_{\text{max}}} \) ordered as \( p_1^{\text{res}} \geq p_2^{\text{res}} \geq \ldots \geq p_{\text{dim}(\mathcal{H})}^{\text{res}} \) and let \( \tau_0 \prec \tau \tau_1 \) be two equilibrium states. Let \( \lambda \) be such that \( ((1 - \lambda)\tau_0, \lambda \tau_1) \prec \lambda \rho \). Then,

\[
\int_0^k \rho f_{((1 - \lambda)\tau_0, \lambda \tau_1)}(x) dx \geq \int_0^k f^T_H(x) dx \quad \forall \lambda \geq 0. \tag{D9}
\]

As \( \tau_0 \) and \( \tau_1 \) are equilibrium states, for any \( 0 \leq \lambda \leq 1 \),

\[
\int_0^k f_{((1 - \lambda)\tau_0, \lambda \tau_1)}(x) dx = \int_0^k \left( \frac{1}{Z_{\tau_0}} \right)^{1 - \lambda} \left( \frac{1}{Z_{\tau_1}} \right)^\lambda dx. \tag{D10}
\]

Therefore, (D9) implies

\[
\left( \frac{1}{Z_{\tau_0}} \right)^{1 - \lambda} \left( \frac{1}{Z_{\tau_1}} \right)^\lambda \geq p_1^{\text{res}}, \tag{D11}
\]

which can be rewritten as

\[
a T \cdot \ln \left( \frac{1}{p_1^{\text{res}}} \right) + b_T \geq \lambda. \tag{D12}
\]

with \( a_T = \frac{1}{\ln Z_{\tau_0}} \) and \( b_T = -a_T \cdot \ln Z_{\tau_0} \) depending on the gauge states \( \tau_0 \) and \( \tau_1 \). On the other hand, (D11) implies

\[
\left( \frac{1}{Z_{\tau_0}} \right)^{1 - \lambda} \left( \frac{1}{Z_{\tau_1}} \right)^\lambda \min \left\{ k, k \right\}
\geq p_1^{\text{res}} \cdot \min \left\{ k, \frac{1}{p_1^{\text{res}}} \right\} \geq \int_0^k f^T_H(x) dx. \tag{D13}
\]

for all \( k \in \mathbb{R}_{\geq 0} \); the second inequality holds as \( f^T_H(x) \) is monotonously decreasing and normalized. Therefore, the conditions (D9) and (D11) are equivalent.

For a state \( \rho \) that is block diagonal in the energy eigenbasis

\[
F_{\text{max}}(\rho) = -k_B T \ln Z_\tau + k_B T D_\infty(\rho||\tau) \ln(2)
\]

\[
= -k_B T \ln Z_\tau + k_B T \ln \min \{ \lambda : \rho \leq \lambda \tau \}
\]

\[
= k_B T \ln \min \{ \mu : \rho \leq Z_\tau \mu \tau \}
\]

\[
= k_B T \ln p_{\text{max}}^{\text{res}}, \tag{D14}
\]

where \( p_{\text{max}}^{\text{res}} \) is the maximal rescaled eigenvalue of the state \( \rho \). Thus, taking the supremum over \( \lambda \) in (D12) implies that \( \tilde{S}_{\uparrow\downarrow}(\rho) = F_{\text{max}}(\rho) \) up to affine transformations.

For \( \tilde{S}_{\uparrow} \) the proof works similarly. Let \( \rho \in \mathcal{S}(\mathcal{H}) \) and let \( \tau_0 \prec \tau \tau_1 \) be two equilibrium states. Now let \( \lambda \) be such that \( \rho \prec \tau ((1 - \lambda) \tau_0, \lambda \tau_1) \) and thus

\[
\int_0^k f_{((1 - \lambda)\tau_0, \lambda \tau_1)}(x) dx \geq \int_0^k f^T(x) dx \quad \forall \lambda \in \mathbb{R}_{\geq 0}. \tag{D15}
\]

First we show by contradiction that

\[
Z_{\tau_0}^{1 - \lambda} Z_{\tau_1}^\lambda \geq Z_\rho. \tag{D16}
\]

Assume for now that \( Z_{\tau_0}^{1 - \lambda} Z_{\tau_1}^\lambda < Z_\rho \). For \( \tilde{k} = Z_{\tau_0}^{1 - \lambda} Z_{\tau_1}^\lambda \), we therefore find

\[
\int_0^k f^T_H(x) dx < 1. \tag{D17}
\]

This contradicts Equation (D15) as

\[
\int_0^k f_{((1 - \lambda)\tau_0, \lambda \tau_1)}(x) dx = 1. \tag{D18}
\]

Thus we have \( \tilde{k} \geq Z_\rho \), which can be rewritten as

\[
\lambda \geq a_T \cdot \ln Z_\rho + b_T, \tag{D19}
\]

with \( a_T \) and \( b_T \) defined as above. Moreover, (D16) implies

\[
\left( \frac{1}{Z_{\tau_0}} \right)^{1 - \lambda} \left( \frac{1}{Z_{\tau_1}} \right)^\lambda \min \left\{ k, Z_{\tau_0}^{1 - \lambda} Z_{\tau_1}^\lambda \right\}
\leq \frac{1}{Z_\rho} \min \left\{ k, Z_\rho \right\} \leq \int_0^k f^T_H(x) dx, \tag{D20}
\]

for all \( k \in \mathbb{R}_{\geq 0} \), i.e., implies (D15); the second inequality holds as \( f^T_H(x) \) is monotonously decreasing and normalized. Therefore, the conditions (D15) and (D16) are equivalent.

For \( F_{\text{min}} \) we find

\[
F_{\text{min}}(\rho) = -k_B T \ln Z_\tau + k_B T D_\infty(\rho||\tau) \ln(2)
\]

\[
= -k_B T \ln Z_\tau - k_B T \ln \text{tr} \Pi_\rho \tau \tag{D21}
\]

\[
= -k_B T \ln (Z_\tau \text{ tr} \Pi_\rho \tau) \tag{D22}
\]

\[
= -k_B T \ln Z_\rho. \tag{D23}
\]

where \( \Pi_\rho \) is the projector onto the support of \( \rho \). Taking the infimum over \( \lambda \) in (D19) implies that \( \tilde{S}_{\uparrow}(\rho) = F_{\text{min}}(\rho) \) up to affine transformations and concludes the proof. \( \square \)

In particular, for constants \( a_T = -k_B T \) and \( b_T = 0 \) corresponding to states with partition functions \( Z_{\tau_0} = 1 \) and \( Z_{\tau_1} = e^{-\beta} \) as above, we obtain precisely

\[
\tilde{S}_{\uparrow} - (\rho) = -k_B T \ln \frac{1}{p_{\text{max}}^{\text{res}}} = F_{\text{max}}(\rho) \tag{D25}
\]

\[
\tilde{S}_{\uparrow}(\rho) = -k_B T \ln Z_\rho = F_{\text{min}}(\rho). \tag{D26}
\]
Appendix E: Connecting other types of reservoirs to the system

Adding a reservoir of any kind to a physical system leads to a mathematically equivalent situation, even though the underlying physics differs. Here, we outline two more scenarios.

1. System connected to a heat and a particle reservoir

Considering a heat and a particle reservoir is a common practice in statistical physics. Our framework can be extended to cover this scenario.

Connecting a particle reservoir to the system and the heat bath allows for particle exchange with this reservoir. Systems in contact with both, a heat and a particle reservoir, have equilibrium states of the form

\[ \rho = \sum_i \frac{e^{-\beta (E_i - N_i)}}{Z} |E_i, N_i \rangle \langle E_i, N_i| \text{ in the eigenbasis } \{|E_i, N_i\}\rangle, \]

where \(Z\) is the grand canonical partition function. For simplicity we assume that there are only particles of one kind.

The system and the two reservoirs together form an isolated system exposed to adiabatic processes assisted by an ancillary system and a weight, expressed by the order relation of majorization \(\prec_M\). As in the case of a heat bath these processes can be described as processes on the system alone. For quantum states \(\rho = \sum_i p_i |E_i, N_i \rangle \langle E_i, N_i|\), which are (block) diagonal in the energy-particle eigenbasis, the possibility of such processes can be expressed by an order relation \(\prec_{N,T}\), which consists again of a rescaling followed by majorization. Analogous to the Gibbs-rescaling, we can define a \(N,T\)-rescaled step function \(f_{\rho}^{N,T}(x)\) as follows.

**Definition 22.** Let \(\rho = \sum_i p_i |E_i, N_i \rangle \langle E_i, N_i|\) be a density matrix. Denote its spectrum by a step function

\[ f_{\rho}(x) = \begin{cases} p_i & 0 \leq x \leq i-1 \leq x \leq i \\ 0 & \text{otherwise.} \end{cases} \]

(E1)

The \(N,T\)-rescaled version of this step function is given as

\[ f_{\rho}^{N,T}(x) = \begin{cases} \frac{1}{Z} & 0 \leq x \leq Z \\ 0 & \text{otherwise.} \end{cases} \]

(E3)

For an equilibrium state \(\rho\) this can be written as

\[ f_{\rho}^{N,T}(x) = \begin{cases} 1 & 0 \leq x \leq Z \\ 0 & \text{otherwise.} \end{cases} \]

Proposition 24. For an equilibrium state \(\rho\) the unique function \(S_{N,T}(\rho)\) equals \(\ln Z\) up to affine transformations.

**Proof.** Let \(\rho \in \mathcal{S}(\mathcal{H})\) be an equilibrium state and let the equilibrium states \(\rho_0 \prec_{N,T} \rho_1 \in \mathcal{S}(\mathcal{H})\) define a gauge. Then for \(\lambda = S_{N,T}(\rho)\),

\[ ((1-\lambda)\rho_0, \lambda \rho_1) \prec_{N,T} \rho \iff \int_0^k f_{\rho_0}^{N,T}(x)dx = \int_0^k f_{\rho_1}^{N,T}(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \]

(E6)

As for \(\prec_T\), the relation \(\prec_{N,T}\) fulfills Lieb and Yngvason’s axioms. According to Theorem 4 there is thus a unique potential

\[ S(\rho)_{N,T} = \{ \lambda : (1-\lambda)\rho_0, \lambda \rho_1 \prec_{N,T} \rho \} \]

(E5)

for equilibrium states.

For equilibrium states \(\rho \prec_{N,T} \rho \prec \mathcal{Z} \geq \mathcal{Z}_\rho\) this is equivalent to

\[ \mathcal{Z}_\rho^{1-\lambda} \mathcal{Z}_\rho^\lambda = \mathcal{Z}_\rho \]

(E7)

and can be written as

\[ \lambda = \frac{1}{\ln \frac{Z_\rho}{Z_\rho^0}} \ln \mathcal{Z}_\rho = \frac{1}{\ln \frac{Z_\rho^0}{Z_\rho}} \ln \mathcal{Z}_\rho \]

(E8)

\[ = a_{N,T} \ln \mathcal{Z}_\rho + b_{N,T}, \]

where \(a_{N,T}\) and \(b_{N,T}\) are constants.
where \( a_{N,T} = \frac{1}{\ln \frac{2^N}{Z_0}} \) and \( b_{n,T} = -a_{N,T} \cdot \ln Z_{\rho_0} \).

The “entropy function”, i.e. the potential for a system in contact with a heat bath and a particle reservoir is thus related to the grand potential \( \Omega \), since

\[
\Omega = -k_B T \ln Z. \tag{E9}
\]

For non-equilibrium states, the bounding functions \( S_{N,T^-} \) and \( S_{N,T^+} \) can be calculated analogously to the scenario including only a heat bath. They define bounds \( \Omega_{\text{max}} \) and \( \Omega_{\text{min}} \) on the potential \( \Omega \) for non-equilibrium states. These are operationally related to the formation and destruction of a state in this scenario, again analogous to the case of a heat bath.

2. System in contact with an angular momentum reservoir

In the study of Landauer’s principle [5], the question whether energy should obtain a special role among the conserved quantities or whether processes such as erasure could as well be realized at an angular momentum instead of an energy cost was brought up by Barnett and Vaccaro [50, 51] and answered in the affirmative. We investigate here whether Lieb and Yngvason’s framework is applicable to systems connected to angular momentum reservoirs, i.e. whether it does not attribute a special role to energy.

As the consideration of an angular momentum reservoir is not common practice, Barnett and Vaccaro’s concrete model of a spin reservoir is presented here. The reservoir consists of \( N \) mobile spin-\( \frac{1}{2} \) particles, for which the possible spin states are denoted by \( |0\rangle \) and \( |1\rangle \). The spin states are assumed to be degenerate in energy and thus decoupled from the spatial degrees of freedom, which are in equilibrium with a heat bath. The equilibrium probability for the reservoir to be in a particular state with \( n \) particles in state \( |1\rangle \) and \( N - n \) particles in state \( |0\rangle \) is

\[
p_n = \frac{e^{-nh \gamma}}{(1 + e^{-h \gamma})^N}, \tag{E10}
\]

where \( \gamma \) is an appropriate parameter analogous to the inverse temperature \( \beta \) for a heat bath. As for each value \( n \) there are \( \binom{N}{n} \) such reservoir states, the normalization is given as 

\[
Z_{\text{res}} = (1 + e^{-h \gamma})^N \quad \text{and has the form of a partition function for the angular momentum reservoir.}
\]

This construction allows us to consider an angular momentum reservoir of arbitrary size \( N \). In the following, we consider a reservoir in the limit \( N \to \infty \).

The state of a system \( S \) in contact with a spin angular momentum reservoir can be described by a density operator. As for systems in contact with a heat reservoir, we restrict ourselves to the treatment of states \( \rho = \sum_i p_i |J_i\rangle \langle J_i| \) that are block diagonal in the eigenbasis of the z-component of the spin operator, denoted as \( \{ |J_i\rangle \} \). To ensure that energy does not affect our considerations, we assume all spin-levels \( |J_i\rangle \) to be degenerate in energy. A system in equilibrium with the reservoir is described by a density operator of the form \( \rho = \sum_i \frac{Z_i e^{-J_i h \gamma}}{Z_0} |J_i\rangle \langle J_i| \) with partition function \( Z_1 = \sum_i e^{-J_i h \gamma} \).

On system \( S \) and reservoir \( R \) we consider again adiabatic processes assisted by an auxiliary system in an equilibrium state and an “angular momentum weight”, which we describe again according to Åberg’s model, just considering the z-component of the spin angular momentum instead of energy. As for energy, the existence of such processes is expressed by the majorization relation \( \prec_M \). The processes occurring on system \( S \) alone are formally determined as in the case of a heat bath, but by exchanging the energy with the z-component of the spin angular momentum. Following again Horodecki and Oppenheim [15], the processes on the system \( S \) for block diagonal states can be expressed with a rescaled order relation \( \prec_J \).

**Definition 25.** Let \( \rho = \sum_i p_i |J_i\rangle \langle J_i| \) be a density matrix diagonal in the eigenbasis of the z-component of the spin operator. Denote its spectrum as

\[
f_\rho(x) = \begin{cases} p_i & i - 1 \leq x \leq i \\ 0 & \text{otherwise} \end{cases}. \tag{E11}
\]

The \( J \)-rescaled step function of \( \rho \) is defined as

\[
f^J_\rho(x) = \begin{cases} p_i & \sum_{k=1}^{i-1} e^{-J_k h \gamma} \leq x \leq \sum_{k=1}^{i} e^{-J_k h \gamma} \\ 0 & \text{otherwise} \end{cases}. \tag{E12}
\]

For an equilibrium state \( \rho \) this simplifies to

\[
f^J_\rho(x) = \begin{cases} \frac{1}{Z_1} & 0 \leq x \leq Z_1 \\ 0 & \text{otherwise} \end{cases}. \tag{E13}
\]

**Definition 26.** Let \( \rho, \sigma \in \mathcal{S}(H) \) be two states which are block diagonal in the basis \( \{ |J_i\rangle \} \). The relation of \( J \)-rescaled majorization \( \prec_J \) is defined as

\[
\rho \prec_J \sigma \iff \int_0^k f^J_\rho(x)dx \geq \int_0^k f^J_\sigma(x)dx \quad \forall k \in \mathbb{R}_{\geq 0}. \tag{E14}
\]

We can describe the processes on system \( S \) corresponding to the order relation \( \prec_J \) as:

- Addition of ancillary systems in an equilibrium state.
- Unitary transformation of system and ancilla conserving angular momentum.
- Removal of any subsystem.
As in the case of a heat bath, the rescaled majorization relation $\prec_J$ fulfills Lieb and Yngvason’s axioms, which gives rise to a unique potential

$$S_J \propto \ln Z_J \quad \text{(E15)}$$

for equilibrium states. For non-equilibrium states, the two bounds on the potential denoted as $S_J^-$ and $S_J^+$ are calculated analogously as in the case of a heat bath, but do not correspond to known quantities.

We have thus found an angular momentum based resource theory corresponding to the order relation $\prec_J$, which has $S_J^-$, $S_J^+$ and $S_J^\pm$ as monotones. In agreement with Barnett’s and Vaccaro’s conclusion [50, 51] that energy can be achieved at an angular momentum instead of an energy cost, energy can be substituted with angular momentum in our resource theoretic picture and does not obtain a special role among the conserved quantities.

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