Replicating the Second Law by Macroscopic Thermodynamic Theory derived from Quantum Mechanics

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We construct a macroscopic thermodynamical theory that satisfies the second law of thermodynamics, based on quantum mechanics. More concretely, our macroscopic theory satisfies the following two features: (1) A macroscopic equilibrium state is expressed as a set of macroscopic variables $\vec{a} = (a_1, \cdots, a_L)$, which represents coarse-grained values of a set of physical quantities, and (2) An adiabatic transformation from an equilibrium state $\vec{a}$ to another state $\vec{a}'$ is possible if and only if $s_{\vec{a}} \leq s_{\vec{a}'}$ holds. Here, $s_{\vec{a}}$ is a real-valued function corresponding to the thermodynamical entropy, which depends only on macroscopic variables $\vec{a}$. We show that such a macroscopic thermodynamical theory can be consistently constructed if the von-Neumann entropy of a generalized microcanonical state satisfies a generalized central limit theorem. We also show that even when the von-Neumann entropies do not satisfy the condition, a macroscopic thermodynamic-like theory with a similar structure exists, which could be at the foundations of nonequilibrium thermodynamics. Finally, we make a replica of (U, V, N) expression of thermodynamics satisfying (1) and (2), using a toy model of free particles in a box.

I. INTRODUCTION

Thermodynamics is one of the most successful phenomenologies in physics, and it has a huge application from chemical reactions [1] to black holes [2]. Reconstruction of macroscopic thermodynamics from the mechanical laws of microscopic physics, be it classical or quantum, is one of the major goals of statistical mechanics. To accomplish this goal, it is necessary to construct a theory, based on the microscopic mechanical laws, to replicate the following two aspects of thermodynamics [1, 3, 4]:

(1) *Expression of Equilibrium States*: Macroscopic equilibrium states are represented by a set of macroscopic variables $(a_1, \cdots, a_L)$, such as (U, V, N) denoting the internal energy, the volume and the number of particles.

(2) *The Second Law*: There exists a real-valued function $s_{\vec{a}}$, which depends only on macroscopic variables $\vec{a}$, such that an adiabatic transformation $\vec{a} \to \vec{a}'$ is possible if and only if $s_{\vec{a}} \leq s_{\vec{a}'}$.

Theories constructed from the ordinary statistical mechanics satisfies Property (1) completemly and Property (2) partially [5–13]. However, such approaches has not succeeded in deriving the necessary and sufficient condition for possibility of state transformation as stated in (2). Recently, approaches to this goal from quantum information theory [14–23] have succeeded in deriving several thermodynamic inequalities, which characterize possibility and impossibility of state transformations by a set of restricted operations. In their approaches, however, conditions for possibility of state transformations are represented by functions which depends microscopic quantum state of the system. This is in contrast to thermodynamics, in which microscopic parameters does not appear in the theory.

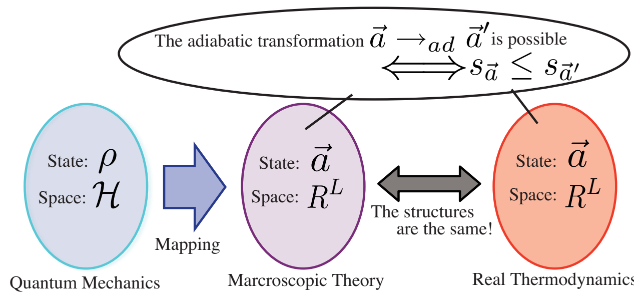

FIG. 1: Schematic diagram of our result

In this article, we integrate the above two streams, and propose a general method to construct a macroscopic theory that satisfies both of the two conditions (1) and (2) (Fig.1). To this end, we first introduce a microcanonocal state as an equivalence class of sequences of generalized microcanonical states which is characterized by a set of macroscopic values $\vec{a} = (a_1, \cdots, a_L)$. Next, we investigate conditions for the existence of a function $s_{\vec{a}}$, which depends only on the macroscopic variables $\vec{a}$ and does not depend on any microscopic parameters, such that an adiabatic transformation from $\vec{a}$ to $\vec{a}'$ is possible if and only if $s_{\vec{a}} \leq s_{\vec{a}'}$ holds. We prove that a sufficient condition for the existence of such a function is that the von Neumann entropies of the generalized microcanonical states satisfy a generalized central limit theorem. We also generalize this result to the case where the von-Neumann entropies do not satisfy this condition, and propose a macroscopic theory which could be at the foundations of nonequilibrium thermodynamics. Finally, we give three examples of our theory, including a replica of (U, V, N) expression of Thermodynamics satisfying (1) and (2), using a toy model of free particles in a box. Although the construction is based on quantum mechanics, our method also applies for discrete classical systems.

II. CONCEPTS AND PURPOSE

Let us introduce key concepts in our construction of macroscopic thermodynamical theories.

- *macroscopic variables*: Variables that characterize macroscopic behavior of a quantum system is called macroscopic variables. They are classified into three types: observable variables, controllable variables and constraint variables. We denote them by $\vec{a}_{\rm o} = (a_{\rm o}^{[1]}, \cdots, a_{\rm o}^{[L]})$, $\vec{a}_{\rm cl} = (a_{\rm cl}^{[1]}, \cdots, a_{\rm cl}^{[L']})$ and $\vec{a}_{\rm cs} = (a_{\rm cs}^{[1]}, \cdots, a_{\rm cs}^{[L'']})$, respectively. In the (U, V, N) expression of thermodynamics, U corresponds to $\vec{a}_{\rm o}$, V corresponds to $\vec{a}_{\rm cl}$, and N corresponds to $\vec{a}_{\rm cs}$.

- *macroscopic state space*: A macroscopic state space corresponding to a constraint $\vec{a}_{\rm cs}$ is denoted by $\mathcal{A}_{\vec{a}_{\rm cs}}$. Elements of $\mathcal{A}$ are $(L + L')$-dimensional real vectors $\vec{a} = (\vec{a}_{\rm o}, \vec{a}_{\rm cl})$ that represent observable variables and controllable variables. We regard constraint variables as fixed in advance. Hence we simply denote $\mathcal{A}_{\vec{a}_{\rm cs}}$ as $\mathcal{A}$, and refer to $\vec{a}$ as explicit variables. A macroscopic state is represented by $\vec{a}$.

- *order structure*: We express as $\vec{a} \succ \vec{a}'$ the fact that a transformation between two macroscopic states $\vec{a} \to \vec{a}'$ is possible by a restricted set of operations. $\succ$ induces a partial order on $\mathcal{A}$, if the set of operations is closed under composition.

- *macro-thermodynamic-like theory*: A pair $\{\mathcal{A}, \succ\}$ is called a macro-thermodynamic-like theory if there exists a set of functions, which only depends on $\vec{a}$ and does not depend on microscopic parameters, that determines if $\vec{a} \succ \vec{a}'$ or not.

- *macro-thermodynamic theory*: A macro-thermodynamic-like theory $\{\mathcal{A}, \succ\}$ is called a macro-thermodynamic theory if there exists a single function $s_{\vec{a}}$, which depends only on $\vec{a}$, such that $\vec{a} \succ \vec{a}'$ holds if and only if $s_{\vec{a}} \leq s_{\vec{a}'}$.

The main goal in this paper is to propose a method to construct a thermodynamic-like theory $\{\mathcal{A}, \succ\}$ based on quantum mechanics, and to give a sufficient condition for $\{\mathcal{A}, \succ\}$ to be a thermodynamic theory.

III. FORMULATION AND MAIN RESULTS

We provide a method to construct the macroscopic state space $\mathcal{A}$, each element of which is represented by macroscopic variables $\vec{a}$. We regard $\vec{a}$ as defining an equivalence class of sequences of generalized microcanonical states. The physical idea is as follows:

Basic Idea 1 *In thermodynamics, the statement "an isolated system is in an equilibrium state $\vec{a}$" means that we have no information about the system except that physical quantities take values $\vec{a}$ stably, within the range of small but nonzero fluctuation that vanishes in a "macroscopic limit". Moreover, it is not possible for us to know the range exactly.*

Let us formulate the above idea rigorously. Since we are concerning a macroscopic limit, we describe a physical system by a sequence of Hilbert spaces $\{\mathcal{H}_{\vec{a}_{\rm cs}}^{(n)}\}_{n=1}^{\infty}$ for each value of constraint variables $\vec{a}_{\rm cs}$, instead of a single Hilbert space $\mathcal{H}$. Because the constraint variables $\vec{a}_{\rm cs}$ are fixed, hereafter we do not write $\vec{a}_{\rm cs}$ explicitly. Hence, we denote $\{\mathcal{H}_{\vec{a}_{\rm cs}}^{(n)}\}_{n=1}^{\infty}$ as $\{\mathcal{H}^{(n)}\}_{n=1}^{\infty}$. A "macroscopic observable" is represented by a sequence of Hermitian operators $A_{\vec{a}_{\rm cl}}^{(n)}$ on $\mathcal{H}^{(n)}$ for each value of controllable variables $\vec{a}_{\rm cl}$, denoted as $\{A_{\vec{a}_{\rm cl}}^{(n)}\}_{n=1}^{\infty}$. As an example for the case where $L' = 0$ (i.e., there is no controllable parameter), consider an n-particle system described by a Hilbert space $\mathcal{H}^{(n)} = \mathcal{H}^{\otimes n}$. An arbitrary observable $A^{(n)}$ which takes the form of

$$A^{(n)} = A^{\langle 1 \rangle} + \cdots + A^{\langle n \rangle}, \tag{1}$$

satisfies the above assumption, where each $A^{\langle k \rangle}$ is a Hermitian operator A acting on the k-th Hilbert space in $\mathcal{H}$ of $\mathcal{H}^{\otimes n}$. Composite systems can also be described in our framework by defining $\mathcal{H}^{(n)}$, e.g., as $\mathcal{H}^{(n)} = \mathcal{H}_1^{(n)} \otimes \mathcal{H}_2^{(n)}$. An example for $L' \geq 1$ will be given in Section IV.

We introduce the generalized microcanonical state as follows. For each n, let $\vec{A}_{\vec{a}_{\rm cl}}^{(n)} := (A_{\vec{a}_{\rm cl}}^{(n),[1]}, ..., A_{\vec{a}_{\rm cl}}^{(n),[L]})$ be a set of commuting Hermitian operators on $\mathcal{H}_{\vec{a}_{\rm cl}}^{(n)}$, each element of which corresponds to an observable variable $a^{[l]}$ $(l = 1, \cdots, L)$. The "δ_n-*window*" $\mathcal{H}_{\vec{a},\delta_n}^{(n)}$ is then defined as

$$\mathcal{H}_{\vec{a},\delta_n}^{(n)} := \left\{ |\psi\rangle \in \mathcal{H}^{(n)} \,\middle|\, \exists \lambda^{[l]} \in [n(a^{[l]} - \delta_n), n(a^{[l]} + \delta_n)) \text{ s.t. } A_{\vec{a}_{\rm cl} + \delta_n \vec{1}_{\rm cl}}^{(n)[l]} |\psi\rangle = \lambda^{[l]} |\psi\rangle \text{ for } 1 \leq l \leq L \right\}, \tag{2}$$

where $\vec{1}_{\rm cl}$ is a L'-dimensional vector $(1, \cdots, 1)$. The parameter δ_n represents the fluctuation of observable variables and controllable variables, i.e., the values of $\vec{a}_o$ and $\vec{a}_{\rm cl}$ are regarded as "macroscopically the same" within the range of fluctuation δ_n, as they are in the case of U and V in thermodynamics. The "generalized microcanonical state" is then defined as the maximally mixed state on the δ_n-window, i.e.,

$$\hat{\pi}_{\vec{a},\delta_n}^{(n)} := \hat{\Pi}_{\vec{a},\delta_n}^{(n)} / D_{\vec{a},\delta_n}^{(n)} \tag{3}$$

where $\hat{\Pi}_{\vec{a},\delta_n}^{(n)}$ is the projection onto $\mathcal{H}_{\vec{a},\delta_n}^{(n)}$ and $D_{\vec{a},\delta_n}^{(n)} := \dim \mathcal{H}_{\vec{a},\delta_n}^{(n)}$.

Now, we define a macroscopic equilibrium state as a vector $\vec{a}$ which characterizes an equivalence class of sequences of generalized microcanonical states. First we introduce a sequence of positive real numbers $\{\delta_n\}_{n=1}^{\infty}$ to represent the asymptotic behavior of the fluctuation of macroscopic variables. Physically, it would be natural to expect that the fluctuation monotonically decreases for the system size, and vanishes in the limit of infinite system size. The speed of convergence should, however, be slower than $1/\sqrt{n}$, because otherwise the higher order fluctuation like the Brownian motion of macroscopic observables becomes macroscopically visible. Note that the small fluctuations like the Brownian motion is outside the scope of equilibrium thermodynamics. Thus it is physically plausible to assume that $\{\delta_n\}_{n=1}^{\infty}$ satisfies

$$O(1/\sqrt{n}) < \delta_n < o(1), \ \ \delta_n > \delta_{n+1}, \tag{4}$$

which is equivalent to

$$\lim_{n\to\infty} \delta_n = 0, \ \lim_{n\to\infty} \sqrt{n}\cdot\delta_n = \infty, \ \ \delta_n > \delta_{n+1}. \tag{5}$$

Hereafter, we denote the set of sequences satisfying (4) by Δ.

As stated in Basic Idea 1, it is physically plausible to assume that two states are macroscopically the same if any macroscopic variables of the states are the same, up to a vanishingly small errors $\{\delta_n\}_{n=1}^{\infty} \in \Delta$. In addition, there is no way for us to know the range δ_n exactly. Therefore, we define macroscopic states of a system as an equivalence class of sequences of the generalized microcanonical states as follows:

$$\vec{a} \equiv \left\{ \{\hat{\pi}^{(n)}_{\vec{a},\delta_n}\}_{n=1}^{\infty} \middle| \ \{\delta_n\}_{n=1}^{\infty} \in \Delta \right\} \tag{6}$$

Let us introduce a mathematical definition of the condition that "an adiabatic transformation $\vec{a} \to \vec{a}'$ is possible". According to Basic Idea 1, we cannot distinguish any two elements of the same equivalence class characterized by $\vec{a}$. Therefore, if we can transform from an arbitrary sequence $\{\hat{\pi}^{(n)}_{\vec{a},\delta_n}\}_{n=1}^{\infty}$ belonging to the equivalence class characterized by $\vec{a}$ to a sequence $\{\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\}_{n=1}^{\infty}$ belonging to the equivalence class characterized by $\vec{a}'$, then we can judge that the transformation from $\vec{a}$ to $\vec{a}'$ is possible (Fig.2). Hence, we model an adiabatic transformation from $\vec{a}$ to $\vec{a}'$ by a sequence of quantum operations $\{\mathcal{E}_n\}_{n=1}^{\infty}$, which maps an element of $\vec{a}$ to that of $\vec{a}'$ with a vanishingly small error.

As the quantum operations $\{\mathcal{E}_n\}_{n=1}^{\infty}$, we employ unital CPTP (completely positive and trace preserving) maps which transform the identity operator to the identity operator, i.e., $\mathcal{E}(\hat{1}) = \hat{1}$. A remarkable feature of the unital operation on $\mathcal{S}(\mathcal{H})$ is that the unital operation does not decrease the von-Neumann entropy of the system [24];

$$S(\mathcal{E}(\rho)) \geq S(\rho), \ \ \forall \rho \in \mathcal{S}(\mathcal{H}), \tag{7}$$

where $\mathcal{S}(\mathcal{H})$ is the state space on $\mathcal{H}$. Because this feature is similar to the adiabatic transformation in thermodynamics, many researches have treat the unital operation as a quantum counterpart of the adiabatic transformation in thermodynamics [12, 20–22]. Using the unital

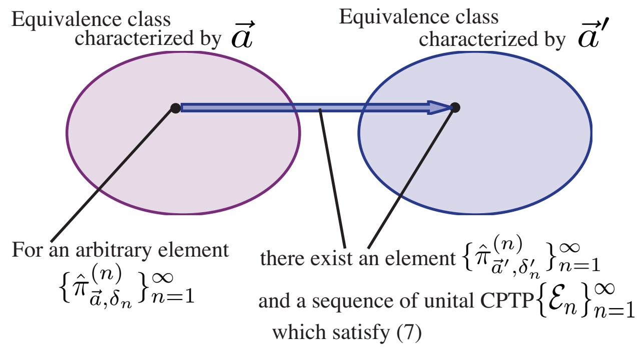

FIG. 2: Schematic diagram of Definition 1

operations, we define a macroscopic adiabatic transformation as follows;

Definition 1 *An adiabatic transformation $\vec{a} \to \vec{a}'$ is possible if, for any $\{\delta_n\}_{n=1}^{\infty} \in \Delta$, there exist $\{\delta'_n\}_{n=1}^{\infty} \in \Delta$ and a sequence $\{\mathcal{E}_n\}_{n=1}^{\infty}$ of unital CPTP maps on $\{\mathcal{S}(\mathcal{H}^{(n)})\}_{n=1}^{\infty}$ such that*

$$\lim_{n\to\infty} \left\| \mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{n}_{\vec{a}',\delta'_n} \right\| = 0. \tag{8}$$

Here, $\|\rho - \sigma\|$ is the trace distance defined by $\|\rho-\sigma\| := \frac{1}{2}\mathrm{Tr}|\rho - \sigma|$. We express as $\vec{a} \succ \vec{a}'$ when an adiabatic transformation $\vec{a} \to \vec{a}'$ is possible.

Clearly, if $\vec{a} \succ \vec{a}'$ and $\vec{a}' \succ \vec{a}''$ hold, then $\vec{a} \succ \vec{a}''$ also holds.

In general, a partially ordered set $(\mathcal{A}, \succ)$ constructed in this way is not necessarily a macro-thermodynamic theory or macro-thermodynamic-like theory. We prove that a sufficient condition for $(\mathcal{A}, \succ)$ to be a macro-thermodynamic theory is given by the following assumption. It states that the von Neumann entropies of the generalized microcanonical states $\{\hat{\pi}^{(n)}_{\vec{a},\delta_n}\}_{n=1}^{\infty}$ satisfies a generalized central limit theorem.

Assumption 1 *The states $\{\hat{\pi}^{(n)}_{\vec{a},\delta_n}\}_{n=1}^{\infty}$ satisfy conditions that*

1. *There exist a positive function $s_{\vec{a}}$ of $\vec{a}$, and a function $f(n)$ which does not depend on $\vec{a}$ and δ_n, such that*

$$S(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - f(n) = ns_{\vec{a}} + o(n) \tag{9}$$

for any $\{\delta_n\}_{n=1}^{\infty} \in \Delta$.

2. *There exist a real number $0 < c < 1$ and a positive function $t_{\vec{a}}$ such that*

$$S(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - f(n) = \begin{cases} ns_{\vec{a}} + o(1) & (\text{if } s_{\vec{a}} = \sup_{\vec{a}_o,\vec{a}_m} s_{\vec{a}}) \\ ns_{\vec{a}} + n\delta_n t_{\vec{a}} + o(n\delta_n) & (\text{otherwise}) \end{cases} \tag{10}$$

for any $\{\delta_n\}_{n=1}^{\infty} \in \Delta$ satisfying $O(n^{c-1}) < \delta_n$.

Theorem 1 *Under Assumption 1, it holds that*

$$s_{\vec{a}} \leq s_{\vec{a}'} \Leftrightarrow \vec{a} \succ \vec{a}'. \tag{11}$$

When Assumption 1 does not hold, $(\mathcal{A}, \succ)$ is not necessarily macro-thermodynamic. In such cases, we define positive functions which serve as $s_{\vec{a}}$ as follows:

$$\overline{s}_{\vec{a},\mathcal{D}} := \limsup_{n\to\infty} \frac{1}{n}\left[S(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - f(n)\right] \tag{12}$$

$$\underline{s}_{\vec{a},\mathcal{D}} := \liminf_{n\to\infty} \frac{1}{n}\left[S(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - f(n)\right] \tag{13}$$

$$\overline{s}_{\vec{a}} := \sup_{\mathcal{D}\in\Delta} \overline{s}_{\vec{a},\mathcal{D}},\ \underline{s}_{\vec{a}} := \sup_{\mathcal{D}\in\Delta} \underline{s}_{\vec{a},\mathcal{D}}. \tag{14}$$

Here, we denote a sequence $\{\delta_n\}_{n=1}^{\infty}$ of positive real numbers as $\mathcal{D}$, and $f(n)$ is a function that only depends on n. If both (12) and (13) are bounded for a function $f(n)$ and any $\mathcal{D} \in \Delta$, so are (14). Then a similar statement as (11) holds regardless of whether Assumption 1 holds or not. Precisely, we have the following theorem, a proof of which is given in Appendix B.

Theorem 2 *When there exists a real valued function $f(n)$ such that (12) and (13) are bounded for any $\mathcal{D} \in \Delta$, it holds that*

$$\overline{s_{\vec{a}}} < \underline{s_{\vec{a}'}} \ \Rightarrow \vec{a} \succ \vec{a}', \tag{15}$$

$$\underline{s_{\vec{a}}} \leq \overline{s_{\vec{a}'}} \ \Leftarrow \vec{a} \succ \vec{a}'. \tag{16}$$

Theorem 1 states that Assumption 1 is a sufficient condition on the choice of controllable variables and macroscopic observables, in order that a macro-thermodynamic theory is constructed based on quantum mechanics. When the sequence $\{S(\hat{\pi}^{(n)}_{\vec{a},\delta_n})\}_{n=1}^{\infty}$ satisfies the generalized central limit theorem, the vector space of macroscopic variables $\vec{a}$ has the same structure as the space of macroscopic equilibrium states in thermodynamics, in terms of convertibility by adiabatic transformations. The function $s_{\vec{a}}$ plays the same role as that of thermodynamic entropy in equilibrium thermodynamics: An adiabatic transformation $\vec{a} \to \vec{a}'$ is possible if and only if $s_{\vec{a}} \leq s_{\vec{a}'}$.

Theorem 2 states that a macro-thermodynamic-like theory can be constructed in general, regardless of whether Assumption 1 holds or not. When the sequence $\{S(\hat{\pi}^{(n)}_{\vec{a},\delta_n})\}_{n=1}^{\infty}$ does not satisfy Assumption 1, a macroscopic state $\vec{a}$ does *not* necesarily have a definite value $s_{\vec{a}}$. Thus we can interpret $\vec{a}$ as representing a non-equilibrium state. Convertibility of macroscopic states is in this case characterized by two functions $\overline{s_{\vec{a}}}$ and $\underline{s_{\vec{a}}}$, not by a single function as in the case of thermodynamic theories. Theorem 2 gives an order structure implied by [4].

We emphasize that our thermodynamic theory and thermodynamic-like theory do not depend on any microscopic parameters, including δ_n that we have introduced to define the generalized microcanonical state $\hat{\pi}_{\vec{a},\delta_n}$. This is in contrast to previous approaches from quantum information theory [14–23], in which convertibility of states are characterized by functions that depends on microscopic parameters. Note that a replica of thermodynamics should not depend on microscopic parameters, since no microscopic parameter appears in thermodynamics. Theories we have constructed satisfy this condition, because macroscopic states are defined as macroscopic variables that characterize the equivalence classes of the sequences of microcanonical states. To our knowledge, this is the first time that a theory satisfying both Conditions (1) and (2) has been constructed from the mechanical laws of microscopic physics.

IV. EXAMPLES

In this section, we describe three examples of macro-thermodynamic theories.

A. Example 1: system of many spins

Consider a system consisting of d-level particles, and suppose that $L' = L'' = 0$. In this case, we only need to consider observable variables $\vec{a}_o$. We have $\mathcal{H}^{(n)} := \mathcal{H}^{\otimes n}$ for each n, where $\mathcal{H}$ is a d-dimensional Hilbert space. A set of observables corresponding to observable variables $\vec{a}_o$ is defined as $\vec{A}^{(n)} := (A_n^{[1]}, \cdots, A_n^{[L]})$. Each l-th component $A_n^{[l]}$ is an arbitrary observable on $\mathcal{H}^{\otimes n}$ such that $a_n^{\max} - a_n^{\min} = O(n)$ holds, where $a_n^{\max}$ and $a_n^{\min}$ are the maximum and minimum eigenvalues of A_n^l, respectively. Then the following lemma holds:

Lemma 1 *Suppose each l-th component $\{A_n^{[l]}\}_{n=1}^{\infty}$ of $\{\vec{A}_n\}_{n=1}^{\infty}$ satisfies the following two conditions:*

1. *There exists a real number $0 < c < 1$ and a real-valued function $I(a)$ which is positive, differentiable, convex for $a \neq 0$ and satisfies*

$$\begin{aligned}\Pr_{\pi^{(n)}}[A_n \geq na] &:= \mathrm{Tr}[\pi^{(n)}\Pi^{(n)}_{[a,\infty)}] \\ &= e^{-nI(a)+o(n^c)} \ (a > 0),\end{aligned} \tag{17}$$

$$\begin{aligned}\Pr_{\pi^{(n)}}[A_n \leq na] &:= \mathrm{Tr}[\pi^{(n)}\Pi^{(n)}_{[-\infty,a)}] \\ &= e^{-nI(a)+o(n^c)} \ (a < 0).\end{aligned} \tag{18}$$

2. *For any $\{\delta_n\}_{n=1}^{\infty} \in \Delta$,*

$$\begin{aligned}&\lim_{n\to\infty} \Pr_{\pi^{(n)}}[-n\delta_n \leq A_n \leq n\delta_n] \\ &:= \lim_{n\to\infty} \mathrm{Tr}[\pi^{(n)}\Pi^{(n)}_{0,\delta_n}] = 1.\end{aligned} \tag{19}$$

Then $\{\hat{\pi}^{(n)}_{\vec{a},\delta_n}\}_{n=1}^{\infty}$ satisfies Assumption 1, and thus a pair $\{\mathcal{A}, \succ\}$ corresponding to $\{\vec{A}_n\}_{n=1}^{\infty}$ and $\mathcal{H}^{(n)}$ is a thermodynamic theory.

We prove this lemma in Appendix A. Inequalities (17) and (18) are equivalent to the large deviation principle when $c = 1$. Hence Condition 1 requires that convergence speed of $\Pr_{\pi^{(n)}}[A_n \geq na]$ and $\Pr_{\pi^{(n)}}[A_n \leq na]$ is faster than required by the large deviation principle. An arbitrary observable of the form (1) satisfies Condition 1 due to the strong large deviation theory [27, 28]. Condition 2 is satisfied by such an observable as well, since it satisfies the central limit theorem.

B. Example 2: a toy model of free particles in a box - a replica of (U, V, N) expression of thermodynamics

Let us consider a toy model of free particles in a box. We assume that the particles are distinguishable, in order to avoid difficulty in counting dimensions of the micro-canonical states. Let $\mathcal{H}$ be a Hilbert space describing the position of a particle in the free space of three dimensions. We introduce a constraint variable N so that nN is equal to the particle number, and define a sequence of Hilbert spaces by $\mathcal{H}^{(n)} := \mathcal{H}^{\otimes nN}$ $(n \in \mathbb{N})$. We regard the volume V of the box as a controllable variable, and the total energy U of the particles as a observable variable. Hence we have $L = L' = L'' = 1$ for this example. An observable corresponding to the total energy is given by the total Hamiltonian $A_V^{(n)} := \sum_{k=1}^{nN} A_V^{\langle k\rangle}$, where $A_V^{\langle k\rangle}$ is the Hamiltonian acting on the k-th Hilbert space of $\mathcal{H}^{\otimes nN}$, defined by

$$A_V^{\langle k\rangle} := \frac{\vec{p_k}^{\,2}}{2m} + f_{nV}(x, y, z) \tag{20}$$

$$f_{nV}(x, y, z) := \begin{cases} 0 & (\text{when } (x, y, z) \in [0, (nV)^{1/3}]^3) \\ \infty & (\textit{otherwise}) \end{cases} \tag{21}$$

Here, $\vec{p}_k$ is the momentum of the k-th particle. Define a sequence $\{\hat{\pi}^{(n)}_{(U,V),\delta_n}\}_{n=1}^{\infty}$ as (2), for which $\vec{a}_{\mathrm{o}} = U$, $\vec{a}_{\mathrm{cl}} = V$ and $\vec{A}^{(n)}_{\vec{a}_{\mathrm{cl}}} = A_V^{(n)}$. As we prove below, the sequence $\{\hat{\pi}^{(n)}_{(U,V),\delta_n}\}_{n=1}^{\infty}$ satisfies Assumption 1. Hence $\{\mathcal{A}, \succ\}$ of this example is a macro-thermodynamic theory.

Proof that $\{\hat{\pi}^{(n)}_{(U,V),\delta_n}\}_{n=1}^{\infty}$ satisfies Assumption 1: Define a subspace $\mathcal{H}^{(n)}_{U_\geq,V} \subset \mathcal{H}^{(n)}$ by

$$\mathcal{H}^{(n)}_{U_\geq,V} := \left\{ |\psi\rangle \in \mathcal{H}^{(n)} \mid \exists \lambda \leq U \text{ s.t. } A_V^{(n)}|\psi\rangle = \lambda|\psi\rangle \right\}, \tag{22}$$

and let $\tilde{D}^{(n)}_{U,V} := \dim \mathcal{H}^{(n)}_{U_\geq,V}$. Then we have

$$\begin{aligned} &\frac{\pi^{3nN/2}}{2^{3nN}\Gamma(\frac{3nN}{2}+1)} \left(n^{5/3} \frac{UV^{2/3}}{\epsilon} \right)^{3nN/2} \geq \tilde{D}^{(n)}_{U,V} \\ &\geq \frac{\pi^{3nN/2}}{2^{3nN}\Gamma(\frac{3nN}{2}+1)} \left(n^{5/3} \frac{UV^{2/3}}{\epsilon} - \sqrt{3nN} \right)^{3nN/2}, \end{aligned} \tag{23}$$

where ϵ is a real positive constant[26]. Using

$$\Gamma(\alpha) = \sqrt{\frac{2\pi}{\alpha}} \left(\frac{\alpha}{e}\right)^{\alpha} \left(1 + \frac{1}{\alpha} + O\left(\frac{1}{\alpha^2}\right) \right), \tag{24}$$

we obtain the following asymptotic expansion by a simple calculation:

$$\log \tilde{D}^{(n)}_{U,V} = f(n) + 3nN \log \frac{U^{3/2}V}{\epsilon^{3/2}} + O(n^{2/3}), \tag{25}$$

where $f(n)$ is a real valued function which depends only on n and N. (Note that N is a constraint variable.) Therefore, defining $I(U, V) := 3N \log \frac{U^{3/2}V}{\epsilon^{3/2}}$, we have

$$\begin{aligned} S(\hat{\pi}^{(n)}_{(U,V),\delta_n}) &= \log(\tilde{D}^{(n)}_{U+\delta_n,V+\delta_n} - \tilde{D}^{(n)}_{U-\delta_n,V+\delta_n}) \\ &= f(n) + nI(U, V) + \left(\frac{\partial I(U, V)}{\partial U} + \frac{\partial I(U, V)}{\partial V} \right) n\delta_n \\ &\quad + o(\delta_n n) + O(n^{2/3}). \end{aligned} \tag{26}$$

Hence the sequence $\{\hat{\pi}^{(n)}_{(U,V),\delta_n}\}_{n=1}^{\infty}$ satisfies Assumption 1. ■

C. Example 3: Another treatment for the toy model of free particles in a box

In Example 2, we have regarded V as a controllable variable. Here, we see that a similar macro-thermodynamic theory can also be constructed in such a way that V is regarded as an observable variable. We perform this translation by using the idea which describes the time-dependent Hamiltonian as the time-independent Hamiltonian of the extended system in Ref.[14]. Define $\mathcal{H}$ in the same way as Example 2, and a sequence of Hilbert spaces by

$$\tilde{\mathcal{H}}^{(n)} = \mathcal{H}^{\otimes nN} \otimes \mathcal{H}_R \quad (n \in \mathbb{N}), \tag{27}$$

where R is a reference system describing the position of a particle in the free space of 1-dimension. The system R plays the role of the controller of the size of the box. Using $A_V^{(n)}$ in Example 2, we introduce a Hamiltonian $A^{[1](n)}$ on $\tilde{\mathcal{H}}^{(n)}_N$ defined by

$$A^{[1](n)} := \sum_{v=1}^{\infty} A^{(n)}_{v/n} \otimes |v\rangle\langle v|. \tag{28}$$

An observable $A^{[2](n)}$ corresponding to the "*size of the box*" is then given by

$$A^{[2](n)} := I \otimes \sum_{v=1}^{\infty} v|v\rangle\langle v|. \tag{29}$$

Define a sequence $\{\hat{\tilde{\pi}}^{(n)}_{(U,V),\delta_n}\}_{n=1}^{\infty}$ as (2), for which $\vec{A}_0^{(n)} = (A^{[1](n)}, A^{[2](n)})$ and $\vec{a}_o = (U, V)$. In the same way as Example 2, the sequence $\{\hat{\tilde{\pi}}^{(n)}_{(U,V),\delta_n}\}_{n=1}^{\infty}$ satisfies Assumption 1. Hence $\{\mathcal{A}, \succ\}$ in this example is a macro-thermodynamic theory as well.

V. DERIVATION

Finally, we prove Theorem 1.

Proof of Theorem 1: The forward implication of Proposition (11) is proved as follows. Let us first consider the case where $s_{\vec{a}} < \sup_{\vec{a}} s_{\vec{a}}$. In general, there exists a unital CPTP map $\mathcal{E}$ such that $\sigma = \mathcal{E}(\rho)$, if and only if ρ and σ satisfies the majorization relation $\rho \succ \sigma$ defined as follows [23, 25];

$$\rho \succ \sigma \underset{\text{def}}{\Longleftrightarrow} \sum_{k=1}^{m} p_k^{\downarrow} \geq \sum_{k=1}^{m} q_k^{\downarrow} \text{ for any } m. \tag{30}$$

Here, $\{p_k^{\downarrow}\}_k$ and $\{q_k^{\downarrow}\}_k$ are eigenvalues of ρ and σ, respectively, sorted in decreasing order[25]. Hence it suffices to show that, for any $\{\delta_n\}_{n=1}^{\infty}$ satisfying $\delta_n \to 0\,(n \to \infty)$, there exists another sequence $\{\delta'_n\}_{n=1}^{\infty}$ satisfying $\delta'_n \to 0\,(n \to \infty)$, such that the following relation holds for any sufficiently large n:

$$\hat{\pi}^{(n)}_{\vec{a},\delta_n} \succ \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}. \tag{31}$$

We prove that (31) is indeed satisfied for any sufficiently large n when we choose $\{\delta'_n\}_{n=1}^{\infty}$ so that $O(\max\{\delta_n, n^{c-1}, 1/\sqrt{n}\}) < \delta'_n < o(1)$. From (10), there exists a sequence of positive real numbers $\{\gamma_n\}_{n=1}^{\infty}$, satisfying $\gamma_n \to 0\,(n \to \infty)$, such that

$$\begin{aligned} ns_{\vec{a}} + \tilde{\delta}_n n(t_{\vec{a}} + \gamma_n) &\geq S(\hat{\pi}^{(n)}_{[\vec{a} \pm \vec{\tilde{\delta}}_n)}) - f(n) \\ &\geq ns_{\vec{a}} + \tilde{\delta}_n n(t_{\vec{a}} - \gamma_n) \end{aligned} \tag{32}$$

holds for any $\{\tilde{\delta}_n\}_{n=1}^{\infty}$ satisfying $O(\max\{n^{c-1}, 1/\sqrt{n}\}) < \tilde{\delta}_n < o(1)$. Since $S(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) = \log D^{(n)}_{\vec{a},\delta_n}$, it follows that

$$e^{-(f(n)+ns_{\vec{a}'}+n\delta'_n(t_{\vec{a}'}-\gamma'_n))} \geq \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} \tag{33}$$

for a sequence of positive real numbers $\{\gamma'_n\}_{n=1}^{\infty}$ satisfying $\gamma'_n \to 0\,(n \to \infty)$. Consider another sequence $\{\delta''_n\}_{n=1}^{\infty}$ such that $O(\max\{n^{c-1}, \delta_n, 1/\sqrt{n}\}) < \delta''_n < o(\delta'_n)$, which necessarily satisfies $O(\max\{n^{c-1}, 1/\sqrt{n}\}) < \delta''_n < o(1)$. Due to (32), there exists a sequence of positive real numbers $\{\gamma''_n\}_{n=1}^{\infty}$ satisfying $\gamma''_n \to 0\,(n \to \infty)$ such that

$$\frac{1}{D^{(n)}_{\vec{a},\delta_n}} \geq \frac{1}{D^{(n)}_{\vec{a},\delta''_n}} \geq e^{-(f(n)+ns_{\vec{a}}+n\delta''_n(t_{\vec{a}}+\gamma''_n)} \tag{34}$$

for any sufficiently large n. From (33), (34) and $O(\delta''_n) < \delta'_n$, it follows that

$$\begin{aligned} \frac{1}{D^{(n)}_{\vec{a},\delta_n}} &\geq e^{-(f(n)+ns_{\vec{a}}+n\delta''_n(t_{\vec{a}}+\gamma''_n)} \\ &\geq e^{-(f(n)+ns_{\vec{a}'}+n\delta'_n(t_{\vec{a}'}-\gamma'_n))} \geq \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} \end{aligned} \tag{35}$$

for any sufficiently large n, which implies (31).

Next we consider the case where $s_{\vec{a}} = \sup_{\vec{a}} s_{\vec{a}}$. We prove that a sequence of unital CPTP maps $\{\mathcal{E}_n\}_{n=1}^{\infty}$, satisfying (8), exists for $\delta'_n = \delta_n$. We prove this by showing that there exists a sequence of positive real numbers $\{\alpha_n\}_{n=1}^{\infty}$, satisfying $\alpha_n \to 0\,(n \to \infty)$, and a sequence of states $\{\rho_n\}_{n=1}^{\infty}$ on $\mathcal{H}_n$, such that

$$\hat{\pi}^{(n)}_{\vec{a},\delta_n} \succ (1-\alpha_n)\hat{\pi}^{(n)}_{\vec{a}',\delta_n} + \alpha_n \rho_n \tag{36}$$

holds for sufficiently large n. Note that we have $\|(1-\alpha_n)\hat{\pi}^{(n)}_{\vec{a}'} + \alpha_n\rho_n - \hat{\pi}^{(n)}_{\vec{a}'}\| \leq \alpha_n$.

The existence of such $\{\alpha_n\}_{n=1}^{\infty}$ and $\{\rho_n\}_{n=1}^{\infty}$ is proved as follows. Since we have $s_{\vec{a}'} = \log d$ when $s_{\vec{a}} \leq s_{\vec{a}'}$, it follows from (10) that there exists a sequence of positive real numbers $\{\beta_n\}_{n=1}^{\infty}$, satisfying $\beta_n \to 0\,(n \to \infty)$, such that we have

$$-\beta_n \leq S(\hat{\pi}^{(n)}_{\vec{a}',\delta_n}) - S(\hat{\pi}^{(n)}_{\vec{a},\delta_n}). \tag{37}$$

Defing α_n by $\log(1-\alpha_n) = -\beta_n$, we have

$$\log(1-\alpha_n) \leq \log \frac{D^{(n)}_{\vec{a}',\delta_n}}{D^{(n)}_{\vec{a},\delta_n}}, \tag{38}$$

which leads to

$$\frac{1-\alpha_n}{D^{(n)}_{\vec{a}',\delta_n}} \leq \frac{1}{D^{(n)}_{\vec{a},\delta_n}}. \tag{39}$$

Therefore, for ρ_n being the maximally mixed state on $\mathcal{H}_n - \mathcal{H}^{(n)}_{\vec{a}',\delta_n}$, Relation (36) holds because $d^n - D^{(n)}_{\vec{a}',\delta_n} \geq D^{(n)}_{\vec{a},\delta_n}$.

We prove the backward implication of Proposition (11) by showing that for arbitrary $\{\delta_n\}_{n=1}^{\infty} \in \Delta$, $\{\delta'_n\}_{n=1}^{\infty} \in \Delta$ and $\{\mathcal{E}_n\}_{n=1}^{\infty}$, the following inequality holds for sufficiently large n;

$$\|\mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\| \geq \frac{1}{3} \tag{40}$$

We refer to the projection to the support $\mathrm{supp}[\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}]$ as $P^{(n)}_{\vec{a}',\delta'_n}$, and define a unital CPTP map $\mathcal{P}^{(n)}_{\vec{a}',\delta'_n}$ as follows;

$$\mathcal{P}^{(n)}_{\vec{a}',\delta'_n}(\rho) = P^{(n)}_{\vec{a}',\delta'_n}\rho P^{(n)}_{\vec{a}',\delta'_n} + (I - P^{(n)}_{\vec{a}',\delta'_n})\rho(I - P^{(n)}_{\vec{a}',\delta'_n}) \tag{41}$$

We also refer to the diagonal elements of $\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$ as $\{p_i\}$, where $\mathcal{E}'_n := \mathcal{P}^{(n)}_{\vec{a}',\delta'_n} \circ \mathcal{E}_n$. Then, because the states $\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}$ and $\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$ can be diagonalized simultaneously, we have

$$\begin{aligned} &\|\mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\| \geq \|\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\| \\ &= \frac{1}{2}\sum_{i=1}^{D^{(n)}_{\vec{a}',\delta'_n}} |p_i - \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}}| + \frac{1}{2}\sum_{i>D^{(n)}_{\vec{a}',\delta'_n}} p_i \\ &\geq \frac{1}{2}\sum_{i=1}^{D^{(n)}_{\vec{a}',\delta'_n}} \left| p_i - \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} \right|. \end{aligned} \tag{42}$$

Here, we defined p_i $(i = 1, \cdots, D^{(n)}_{\vec{a}',\delta'_n})$ as eigenvalues of $\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$ on supp$[\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}]$. Because of $\hat{\pi}^{(n)}_{\vec{a},\delta_n} \succ \mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) \succ \mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$, we have

$$\frac{1}{D^{(n)}_{\vec{a},\delta_n}} \geq p_i, \ \ \forall i. \tag{43}$$

Moreover, because $\delta_s := s_{\vec{a}} - s_{\vec{a}'}$ is positive, the following inequality holds for sufficiently large n

$$\log D^{(n)}_{\vec{a},\delta_n} - \log D^{(n)}_{\vec{a}',\delta'_n} > \frac{n\delta_s}{2} \tag{44}$$

Therefore, we have

$$\frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} > \frac{e^{-n\delta_s/2}}{D^{(n)}_{\vec{a}',\delta'_n}} > \frac{1}{D^{(n)}_{\vec{a},\delta_n}} \geq p_i \tag{45}$$

for sufficiently large n. Hence, we obtain

$$\begin{aligned} \|\mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\| &\geq \frac{1}{2}\sum_{i=1}^{D^{(n)}_{\vec{a}',\delta'_n}} \left| \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} - p_i \right| \\ &> \frac{1}{2} D^{(n)}_{\vec{a}',\delta'_n} \left(\frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} - \frac{e^{-n\delta_s/2}}{D^{(n)}_{\vec{a}',\delta'_n}} \right) > \frac{1}{3} \end{aligned} \tag{46}$$

for sufficiently large n.

■

Conclusion: We have constructed a macro-thermodynamic theory based on quantum mechanics. The theory is described only by macroscopic observables, and does not include microscopic parameters. We firstly expresses a macroscopic state as a set of macroscopic variables $\vec{a}$ which characterizes the equivalence classes of the sequences of the generalized microcanonical states $\{\hat{\pi}^{(n)}_{\vec{a},\delta_n}\}_{n=1}^{\infty}$. Next, we have shown that when the sequence of von-Neumann entropy $\{S(\hat{\pi}^{(n)}_{\vec{a},\delta_n})\}_{n=1}^{\infty}$ satisfies the generalized central limit theorem, the theory replicates the second law of thermodynamics. More precisely, we prove the existence of a real-valued function $s_{\vec{a}}$ depending only on the macroscopic variables $\vec{a}$, such that an adiabatic transformation $\vec{a} \to \vec{a}'$ is possible if and only if $s_{\vec{a}} \leq s_{\vec{a}'}$. The function thus plays the same role in our theory as that of the thermodynamical entropy in thermodynamics.

Even when the observables do not satisfy the central limit theorem or the large deviation principle, we have proven that a thermodynamic-like theory with a second law-like condition can be constructed. Such a theory could be a candidate for a quantum mechanical foundation of nonequilibrium thermodynamics.

We emphasize that our thermodynamic and thermodynamic-like theories do not depend on any microscopic parameters, including δ_n that we introduced to define the generalized microcanonical state $\hat{\pi}_{\vec{a},\delta_n}$. This is the biggest advantage of our approach on the previous results which have given several types of thermodynamic-like theories[5–22].

Finally, we give three examples of our theory, including a replica of (U, V, N) expression of thermodynamics which has the same totally ordered structure as thermodynamics, using a toy model of free particles in a box.

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Appendix A: Proof of Lemma 1

We consider the case where $L = 1$ for simplicity. Generalization to an arbitrary $L > 1$ is straightforward.

For $a > 0$, noting that

$$\begin{aligned} D^{(n)}_{a,\delta_n} &= \Pr_{\hat{\pi}^{(n)}}[n(a+\delta_n) > A_n \geq n(a-\delta_n)]d^n \\ &= (\Pr_{\hat{\pi}^{(n)}}[A_n \geq n(a-\delta_n)] - \Pr_{\hat{\pi}^{(n)}}[A_n \geq n(a+\delta_n)])d^n, \end{aligned} \tag{A1}$$

we have, from (17),

$$\begin{aligned} S(\hat{\pi}^{(n)}_{a,\delta_n}) &= \log D^{(n)}_{a,\delta_n} \\ &= n\log d + \log e^{-nI(a-\delta_n)+o(n^c)} \\ &\quad + \log(1 - e^{-n(I(a+\delta_n)-I(a-\delta_n))+o(n^c)}) \\ &= n\log d + \log e^{-nI(a)+n\delta_n I'(a)+o(n\delta_n)+o(n^c)} \\ &\quad + \log(1 - e^{-2n\delta_n I'(a)+o(n\delta_n)+o(n^c)}). \end{aligned} \tag{A2}$$

Consequently, we obtain

$$S(\hat{\pi}^{(n)}_{a,\delta_n}) = n(\log d - I(a)) + o(n\delta_n) + o(n^c) \tag{A3}$$

for any $\{\delta_n\}_{n=1}^{\infty} \in \Delta$. Due to (18), Equality (A3) holds for $a < 0$ as well. Noting that $I(a) > 0$ and thus $s_a < \log d$ when $a \neq 0$, we obtain (9) and (10) for $a \neq 0$.

For $a = 0$, it immediately follows from (19) that

$$\begin{aligned} S(\hat{\pi}^{(n)}_{0,\delta_n}) &= \log D^{(n)}_{a,\delta_n} \\ &= \log \Pr_{\hat{\pi}^{(n)}}[-n\delta_n \leq A_n \leq n\delta_n]d^n \\ &= n\log d + \log(1+o(1)) = n\log d + o(1). \end{aligned} \tag{A4}$$

Thus we have $s_a = \log d$ and hence (9) and (10), which competes the proof. ■

Appendix B: Proof of Theorem 2

Let us denote $\vec{a}$ and $\vec{a}'$ simply by a and a', respectively. We first introduce a generalized adiabatic process.

Definition 2 *An adiabatic transformation $(a,\mathcal{D}) \to (a',\mathcal{D}')$ is possible if there exists a sequence $\{\mathcal{E}_n\}_{n=1}^{\infty}$ of unital CPTP maps $\mathcal{E}_n$ on $\mathcal{S}(\mathcal{H}^{\otimes n})$ that satisfies*

$$\lim_{n\to\infty} \left\| \mathcal{E}_n\left(\hat{\pi}^{(n)}_{a,\delta_n}\right) - \hat{\pi}^{(n)}_{a',\delta'_n} \right\| = 0.$$

Definition 1 is then reformulated as follows:

Definition 1 *An adiabatic transformation $a \to a'$ is possible if, for any $\mathcal{D} \in \Delta$, there exists another sequence $\mathcal{D}' \in \Delta$, such that an adiabatic transformation $(a,\mathcal{D}) \to (a',\mathcal{D}')$ is possible.*

We prove the following lemma regarding the possibility and impossibility of generalized adiabatic processes.

Lemma 2 *Suppose $\mathcal{D},\mathcal{D}' \in \Delta$. An adiabatic transformation $(a,\mathcal{D}) \to (a',\mathcal{D}')$ is possible if $\overline{s}_{a,\mathcal{D}} < \underline{s}_{a',\mathcal{D}'}$. Conversely, an adiabatic transformation $(a,\mathcal{D}) \to (a',\mathcal{D}')$ is possible only if $\underline{s}_{a,\mathcal{D}} \leq \overline{s}_{a',\mathcal{D}'}$.*

Proof of Lemma 2: In general, there exists a unital map $\mathcal{E}$ satisfying $\mathcal{E}(\rho) = \sigma$, if and only if $\rho \succ \sigma$ holds, i.e., σ is majorized by ρ. Thus we prove the first statement of Theorem 2 by showing that, for sufficiently large n, we have

$$\hat{\pi}^{(n)}_{a,\delta_n} \succ \hat{\pi}^{(n)}_{a',\delta'_n}. \tag{B1}$$

Define

$$\begin{aligned} \overline{\gamma}_{a,\mathcal{D},n} &:= \max\left\{\frac{1}{n}S(\hat{\pi}^{(n)}_{a,\delta_n}) - \overline{s}_{a,\mathcal{D}} - \frac{1}{n}f(n),\ 0\right\}, \\ \underline{\gamma}_{a,\mathcal{D},n} &:= \min\left\{\frac{1}{n}S(\hat{\pi}^{(n)}_{a,\delta_n}) - \underline{s}_{a',\mathcal{D}'} - \frac{1}{n}f(n),\ 0\right\}. \end{aligned}$$

By definition, we have

$$\underline{s}_{a',\mathcal{D}'} + \underline{\gamma}_{a,\mathcal{D},n} \leq \frac{S(\hat{\pi}^{(n)}_{a,\delta_n}) - f(n)}{n} \leq \overline{s}_{a,\mathcal{D}} + \overline{\gamma}_{a,\mathcal{D},n} \tag{B2}$$

for any a, $\mathcal{D}$ and n, as well as

$$\lim_{n\to\infty} \overline{\gamma}_{a,\mathcal{D},n} = \lim_{n\to\infty} \underline{\gamma}_{a,\mathcal{D},n} = 0 \tag{B3}$$

due to the definitions of $\overline{\gamma}_{a,\mathcal{D},n}$ and $\underline{\gamma}_{a,\mathcal{D},n}$. From (B2), we have

$$\begin{aligned} \frac{1}{n}\log D^{(n)}_{a,\delta_n} &= \frac{1}{n}S(\hat{\pi}^{(n)}_{a,\delta_n}) \leq \overline{s}_{a,\mathcal{D}} + \overline{\gamma}_{a,\mathcal{D},n} + \frac{1}{n}f(n), \\ \frac{1}{n}\log D^{(n)}_{a',\delta'_n} &= \frac{1}{n}S(\hat{\pi}^{(n)}_{a',\delta'_n}) \geq \underline{s}_{a',\mathcal{D}'} + \underline{\gamma}_{a,\mathcal{D},n} + \frac{1}{n}f(n). \end{aligned}$$

Hence we obtain

$$\begin{aligned} &\frac{1}{n}\log D^{(n)}_{a',\delta'_n} - \frac{1}{n}\log D^{(n)}_{a,\delta_n} \\ &\geq (\underline{s}_{a',\mathcal{D}'} - \overline{s}_{a,\mathcal{D}}) + (\underline{\gamma}_{a,\mathcal{D},n} - \overline{\gamma}_{a,\mathcal{D},n}). \end{aligned} \tag{B4}$$

Suppose $\overline{s}_{a,\mathcal{D}} < \underline{s}_{a',\mathcal{D}'}$. Due to (B3) and (B4), we have

$$\log D^{(n)}_{a',\delta'_n} - \log D^{(n)}_{a,\delta_n} \geq 0$$

for sufficiently large n, which implies (B1). This completes the proof of the first part of Theorem 2.

We prove the second statement by showing that an adiabatic transformation $(a,\mathcal{D}) \to (a',\mathcal{D}')$ is *not* possible if $\delta s := \underline{s}_{a',\mathcal{D}'} - \overline{s}_{a,\mathcal{D}} > 0$. From (B2), we have

$$\begin{aligned} & \log D^{(n)}_{a,\delta_n} - \log D^{(n)}_{a',\delta'_n} \\ = \ & S(\hat{\pi}^{(n)}_{a,\delta_n}) - S(\hat{\pi}^{n}_{a',\delta'_n}) \\ \geq \ & n(\underline{s}_{a',\mathcal{D}'} - \overline{s}_{a,\mathcal{D}}) + n(\underline{\gamma}_{a,\mathcal{D},n} - \overline{\gamma}_{a,\mathcal{D},n}) \\ = \ & n(\delta s + \delta\gamma_n). \end{aligned}$$

for any n, where we defined

$$\delta\gamma_n := \underline{\gamma}_{a,\mathcal{D},n} - \overline{\gamma}_{a,\mathcal{D},n}.$$

Hence we have

$$\frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} > \frac{e^{-n\delta_s/2}}{D^{(n)}_{\vec{a}',\delta'_n}} \geq \frac{1}{D^{(n)}_{\vec{a},\delta_n}} \tag{B5}$$

for sufficiently large n. Note that $\lim_{n\to\infty}\delta\gamma_n = 0$ from (B3). Let $P^{(n)}_{\vec{a}',\delta'_n}$ be the projection onto $\mathrm{supp}[\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}]$, and define a unital CPTP map $\mathcal{P}^{(n)}_{\vec{a}',\delta'_n}$ by

$$\mathcal{P}^{(n)}_{\vec{a}',\delta'_n}(\rho) = P^{(n)}_{\vec{a}',\delta'_n}\rho P^{(n)}_{\vec{a}',\delta'_n} + (I - P^{(n)}_{\vec{a}',\delta'_n})\rho(I - P^{(n)}_{\vec{a}',\delta'_n}).$$

Let $\mathcal{E}'_n := \mathcal{P}^{(n)}_{\vec{a}',\delta'_n} \circ \mathcal{E}_n$ for any unital CPTP map $\mathcal{E}_n$. Since $\mathcal{E}'_n$ is a untal CPTP map as well, we have

$$\hat{\pi}^{(n)}_{\vec{a},\delta_n} \succ \mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) \succ \mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}). \tag{B6}$$

Consequently, all eigenvalues p_i of $\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$ satisfies

$$\frac{1}{D^{(n)}_{\vec{a},\delta_n}} \geq p_i. \tag{B7}$$

From (B5) and (B7), we obtain

$$\frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} > \frac{e^{-n\delta_s/2}}{D^{(n)}_{\vec{a}',\delta'_n}} \geq p_i. \tag{B8}$$

The distance between $\mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$ and $\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}$ is then bounded as follows. Due to the monotonicity of the trace distance, we have

$$\begin{aligned} & \left\|\mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\right\| \geq \|\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\| \\ = \ & \frac{1}{2}\sum_{i=1}^{D^{(n)}_{\vec{a}',\delta'_n}} \left|p_i - \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}}\right| + \frac{1}{2}\sum_{i > D^{(n)}_{\vec{a}',\delta'_n}} p_i \\ \geq \ & \frac{1}{2}\sum_{i=1}^{D^{(n)}_{\vec{a}',\delta'_n}} \left|p_i - \frac{1}{D^{(n)}_{\vec{a}',\delta'_n}}\right| \\ \geq \ & \frac{1}{2} D^{(n)}_{\vec{a}',\delta'_n}\left(\frac{1}{D^{(n)}_{\vec{a}',\delta'_n}} - \frac{e^{-n\delta_s/2}}{D^{(n)}_{\vec{a}',\delta'_n}}\right) \\ = \ & \frac{1}{2}\left(1 - e^{-n\delta_s/2}\right). \end{aligned} \tag{B9}$$

Here, we defined p_i $(i = 1, \cdots, D^{(n)}_{\vec{a}',\delta'_n})$ as eigenvalues of $\mathcal{E}'_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n})$ on $\mathrm{supp}[\hat{\pi}^{(n)}_{\vec{a}',\delta'_n}]$. Thus we obtain

$$\left\|\mathcal{E}_n(\hat{\pi}^{(n)}_{\vec{a},\delta_n}) - \hat{\pi}^{(n)}_{\vec{a}',\delta'_n}\right\| \geq \frac{1}{3} \tag{B10}$$

for sufficiently large n and any unital CPTP map $\mathcal{E}_n$, which implies that an adiabatic transformation $(a,\mathcal{D}) \to (a',\mathcal{D}')$ is not possible. ■

Proof of Theorem 2: The first statement is proved as follows. Due to (14), we have

$$\begin{aligned} & \forall\mathcal{D} \in \Delta\,; \ \ \overline{s}_{a,\mathcal{D}} \leq \overline{s}_a, \\ & \forall\epsilon > 0, \exists\mathcal{D}' \in \Delta\,; \ \ \underline{s}_{a'} - \epsilon \leq \underline{s}_{a',\mathcal{D}'}. \end{aligned} \tag{B11}$$

By choosing $\epsilon = (\underline{s}_{a'} - \overline{s}_a)/2 > 0$, it follows that for any $\mathcal{D}$, there exists $\mathcal{D}'$ such that

$$\overline{s}_{a,\mathcal{D}} \leq \overline{s}_a < \frac{\underline{s}_{a'} + \overline{s}_a}{2} \leq \underline{s}_{a',\mathcal{D}'}. \tag{B12}$$

Thus an adiabatic transformation $a \to a'$ is possible. To prove the second statement, assume we have $\underline{s}_a > \overline{s}_{a'}$. Equivalently to (B11), we have

$$\begin{aligned} & \forall\epsilon > 0, \exists\mathcal{D} \in \Delta\,; \ \ \underline{s}_a - \epsilon \leq \underline{s}_{a,\mathcal{D}} \\ & \forall\mathcal{D}' \in \Delta\,; \ \ \overline{s}_{a',\mathcal{D}'} \leq \overline{s}_{a'}. \end{aligned}$$

By choosing $\epsilon = (\underline{s}_a - \overline{s}_{a'})/2 > 0$, we find that there exists $\mathcal{D}$, such that for any $\mathcal{D}'$ we have

$$\underline{s}_{a,\mathcal{D}} \geq \frac{\underline{s}_a + \overline{s}_{a'}}{2} > \overline{s}_{a'} \geq \overline{s}_{a',\mathcal{D}'}. \tag{B13}$$

Thus an adiabatic transformation $a \to a'$ is not possible, which completes the proof. ■

Remark. Theorem 2 also holds when we define $\underline{s}_a$ by

$$\underline{s}_a := \inf_{\mathcal{D}} \underline{s}_{a,\mathcal{D}}, \tag{B14}$$

instead of (14). In that case, however, the statement of Theorem 2 becomes weaker.