


# Exploring the gap between thermal operations and enhanced thermal operations

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The gap between thermal operations (TO) and enhanced thermal operations (EnTO) is an open problem raised in [Phys. Rev. Lett. **115**, 210403 (2015)]. It originates from the limitations on coherence evolutions. Here we solve this problem by analytically proving that a state transition induced by EnTO cannot be approximately realized by TO. It confirms that TO and EnTO lead to different laws of state conversions. Our results can also contribute to the study of the restrictions on coherence dynamics in quantum thermodynamics.

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## I. INTRODUCTION

In the resource theory of quantum thermodynamics [1], the main problem is to figure out the allowed state conversions under a set of quantum operations known as thermal operations (TO). A thermal operation is generally constructed as follows [2–5]. A quantum system, previously isolated, is brought into contact with a heat bath at a fixed inverse temperature  $\beta$ , and then decoupled from it after some time.

As in Refs. [6,7], two important properties of TO are identified: (i) the time-translation symmetry, and (ii) the Gibbs-preserving property. Conversely, these two conditions may not be sufficient for a quantum operation to be a thermal operation. Thus the set of quantum operations which satisfy these properties is defined as enhanced thermal operations (EnTO) [6,7], or thermodynamic processes in some literature [8]. By definition, TO is a subset of EnTO. Compared to TO, EnTO is easier to be dealt with mathematically. Further, the two properties in the definition of EnTO correspond to the first and second laws of thermodynamics, respectively. Thus, EnTO is widely employed to study the constraints on state conversions.

When only population dynamics is concerned, it has been proven that state conversions induced by TO are equivalent to those induced by EnTO [3,9]. This elegant result leads to the necessary and sufficient conditions on population dynamics under TO [3,4,9–11]. In general quantum systems, where coherence between energy levels may exist, one should also take into account the constraints on coherence evolution [6,7,12–18]. A complete set of necessary and sufficient conditions for arbitrary quantum state conversions under EnTO are provided recently [8], while it is unclear whether these conditions are sufficient for state conversion under TO.

Although EnTO and TO acting on qubit systems are equivalent, it is not true for high-dimensional systems. In Ref. [6], it is discovered that there are state conversions under EnTO which cannot be realized *exactly* by TO. However, it remains an open problem whether this gap can be closed

*approximately*. This open problem attracts wide attentions from researchers working on quantum thermodynamics and resource theories [1]. Here we formally state two versions of the closure conjecture.

*Conjecture 1.* (Closure conjecture, v1) For any enhanced thermal operation  $\mathcal{E}^{\text{EnTO}}$ , there exists a thermal operation  $\mathcal{E}^{\text{TO}}$ , such that the distance between these two operations is small, i.e.,  $|\mathcal{E}^{\text{EnTO}} - \mathcal{E}^{\text{TO}}| \leq \epsilon$ .

*Conjecture 2.* (Closure conjecture, v2) For any given input state  $\rho_0$ , if the state conversion  $\rho_0 \rightarrow \rho$  is realizable by EnTO, then there exists a state  $\rho'$  such that  $|\rho' - \rho| \leq \epsilon$  and the conversion  $\rho_0 \rightarrow \rho'$  is achievable by TO.

Apparently, Conjecture 1 implies Conjecture 2. Thus disproof of Conjecture 2 is sufficient for disproving Conjecture 1. In this paper, we will disprove Conjecture 2 with an analytical counterexample, and hence confirm that TO and EnTO lead to different laws of state conversions. Precisely, we consider a qutrit system with Hamiltonian  $H_S = \sum_{m=0}^2 m\hbar\omega |m\rangle\langle m|$  and in initial state  $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ , and a heat bath at a fixed inverse temperature  $\beta$ . We will show that the state conversion from  $\rho_0 = |\psi_0\rangle\langle\psi_0|$  to

$$\rho = \frac{1}{2} \begin{pmatrix} 1 - e^{-2\beta\hbar\omega} & \sqrt{1 - e^{-2\beta\hbar\omega}} & 0 \\ \sqrt{1 - e^{-2\beta\hbar\omega}} & 1 & 0 \\ 0 & 0 & e^{-2\beta\hbar\omega} \end{pmatrix} \quad (1)$$

is realizable by EnTO. Then we will prove analytically that any state  $\rho'$  satisfying  $|\rho' - \rho| \leq \epsilon$  is not achievable by TO, if the inverse temperature  $\beta$  is chosen such that  $0 \ll e^{-\beta\hbar\omega} \ll \frac{\sqrt{5}-1}{2}$  holds.

## II. THERMAL OPERATIONS AND RELATED CONCEPTS

Here we briefly review some related concepts and results. We consider a quantum system  $S$  with Hamiltonian  $H_S$  and in a state  $\rho_S$ . A thermal operation  $\mathcal{E}^{\text{TO}}$  on  $S$  is defined as [3]

$$\mathcal{E}^{\text{TO}}(\rho_S) = \text{Tr}_R[U(\rho_S \otimes \gamma_R)U^\dagger], \quad (2)$$

where  $\gamma_R = e^{-\beta H_R} / \text{Tr}(e^{-\beta H_R})$  is the Gibbs state of the heat bath  $R$ ,  $H_R$  is the Hamiltonian of  $R$ ,  $\beta$  is the inverse

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temperature, and  $U$  is a joint unitary which preserves the total energy, i.e.,  $[U, H_S + H_R] = 0$ .

An enhanced thermal operation  $\mathcal{E}^{\text{EnTO}}$  on  $S$  is defined as a quantum operation which satisfies both of the following conditions [6]:

(i) Time-translation symmetric condition,

$$\mathcal{E}^{\text{EnTO}}(e^{-iH_S t} \rho_S e^{iH_S t}) = e^{-iH_S t} \mathcal{E}^{\text{EnTO}}(\rho_S) e^{iH_S t}; \quad (3)$$

(ii) Gibbs-preserving condition,

$$\mathcal{E}^{\text{EnTO}}(\gamma_S) = \gamma_S. \quad (4)$$

If a quantum operation belongs to TO, it satisfies these two conditions [6]. However, the converse may not be true. Hence, TO is a subset of EnTO.

Let  $\mathbf{p}_0$  and  $\mathbf{p}$  be the vectors of population distributions of states  $\rho_0$  and  $\rho$ , respectively. Each element of the vector  $\mathbf{p}$  is  $p_k = \langle k | \rho | k \rangle$  (and similar for  $\mathbf{p}_0$ ), where  $|k\rangle$  are energetic eigenstates of the system. The population dynamics induced by an enhanced thermal operation  $\mathcal{E}^{\text{EnTO}}$  can be written as

$$\rho = \mathcal{E}^{\text{EnTO}}(\rho_0) \Rightarrow \mathbf{p} = G\mathbf{p}_0. \quad (5)$$

Here  $G$  is a matrix of transition probabilities  $G_{k'k} = p_{k'|k} = \langle k' | \mathcal{E}(|k\rangle\langle k|) | k' \rangle$  from state  $|k\rangle$  to  $|k'\rangle$ . From property (ii), the population dynamics  $G$  induced by EnTO is a stochastic matrix that preserves the Gibbs distribution. Such matrices, also called the Gibbs-stochastic matrices, can be realized by TO [3,9]. Hence, when only population dynamics is concerned, TO is equivalent to EnTO.

As shown in Refs. [6,7], the coherence dynamics between energy levels depends on both initial coherence of quantum state and transition probabilities. For a quantum state  $\rho$  expanded in its energy eigenbasis  $\rho = \sum_{i,j} \rho_{ij} |i\rangle\langle j|$ , (where  $H_S |j\rangle = E_j |j\rangle$ ) a mode of coherence is defined as an operator  $\rho^{(\omega)}$  composed of coherence terms between degenerate gaps:

$$\rho^{(\omega)} = \sum_{i,j: E_i - E_j = \hbar\omega} \rho_{ij} |i\rangle\langle j|. \quad (6)$$

The output coherence term after the action of an enhanced thermal operation is bounded as [6,7]

$$|\rho_{ij}| \leq \sum'_{c,d} |\rho_{0,cd}| \sqrt{P_{ic} P_{jd}}, \quad (7)$$

where the primed sum  $\sum'$  refers to the summation over the indices  $c, d$  which satisfy  $E_c - E_d = E_i - E_j$ , and  $\rho_{0,cd} = \langle c | \rho_0 | d \rangle$ .

If  $\rho_0$  and  $\rho$  are qubit states, TO can saturate the bound in Eq. (7). However, for higher dimension systems, there exist situations where TO cannot achieve the bound in Eq. (7) exactly, while EnTO can [6].

### III. SETUP

In this paper, the main system we focus on is a three-dimensional quantum system whose Hamiltonian reads

$$H_S = \sum_{m=0}^2 m \hbar\omega |m\rangle\langle m|. \quad (8)$$

The heat bath is at a fixed inverse temperature  $\beta$ . In the following, we will label  $q \equiv e^{-\beta\hbar\omega}$ .

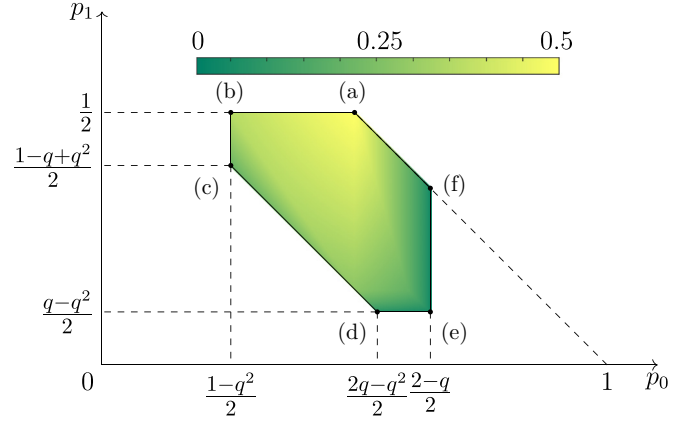


FIG. 1. EnTO cone of the qutrit state  $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ . The maximum value of  $|\rho_{01}|$  is presented as the color bar (gray-scale bar). Here the parameter  $q \equiv e^{-\beta\hbar\omega}$ .

Because the main purpose of this paper is to disprove the closure conjecture, a counterexample is sufficient. Thus in the rest of the paper, we mainly consider the initial state  $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ , though most of our discussions can be applied to general input states.

The set of states which can be obtained from a given input state  $\rho$  via a set of operations  $X$  is called the  $X$  cone of  $\rho$ , labeled as  $C^X(\rho) := \{\rho' : \rho' = \mathcal{E}(\rho), \mathcal{E} \in X\}$ . The gap between two sets of operations  $X_1$  and  $X_2$  can be indicated from a gap between  $X_1$  cone and  $X_2$  cone of a given state. In the following, we explicitly calculate the EnTO cone of the state  $\rho_0 = |\psi_0\rangle\langle\psi_0|$ , and show that the gap between  $C^{\text{TO}}(\rho_0)$  and  $C^{\text{EnTO}}(\rho_0)$  is non-negligible.

From Eqs. (5) and (7), any state in the EnTO (or TO) cone of  $\rho_0$  is in the following form:

$$\rho = \begin{pmatrix} p_0 & |\rho_{10}|e^{i\phi_1} & 0 \\ |\rho_{10}|e^{-i\phi_1} & p_1 & |\rho_{21}|e^{i\phi_2} \\ 0 & |\rho_{21}|e^{-i\phi_2} & 1 - p_0 - p_1 \end{pmatrix}. \quad (9)$$

Further, all states in the above form are equivalent to states with  $\phi_1 = \phi_2 = 0$  by covariant unitary operators  $U(\phi_1, \phi_2) = \text{diag}(e^{-i\phi_1}, 1, e^{i\phi_2})$ . Therefore, the EnTO (or TO) cone of  $\rho_0$  is fully described by  $(p_0, p_1, |\rho_{10}|, |\rho_{21}|)$ , and hence can be presented in a four-dimensional parameter space. In the following, we will focus on the projections of the cones on the three-dimensional parameter space  $(p_0, p_1, |\rho_{10}|)$ . Clearly, a gap between the projections of cones indicates a gap between the cones.

### IV. THE GAP BETWEEN TO AND EnTO

#### A. EnTO cone

Here we analytically calculate the EnTO cone of the initial state  $\rho_0$ , and visualize it in the three-dimensional parameter space spanned by  $(p_0, p_1, |\rho_{10}|)$ ; see Fig. 1. The range of the output populations is derived from the thermo-majorization relation [3].

The maximum value of  $|\rho_{10}|$  for given  $(p_0, p_1)$  is obtained as follows. According to Eq. (7), we have  $|\rho_{10}| \leq \frac{1}{2}\sqrt{G_{00}G_{11}}$ .

Thus the upper bound of  $|\rho_{10}|$  reads

$$\begin{aligned} & \max \frac{1}{2} \sqrt{G_{00} G_{11}} \\ & \text{s.t. } G\boldsymbol{\gamma} = \boldsymbol{\gamma}, G\mathbf{p}_0 = \mathbf{p}, G^T \mathbb{I} = \mathbb{I}, G_{ij} \in [0, 1], \end{aligned} \quad (10)$$

where  $\boldsymbol{\gamma} = (1, q, q^2)^T$ ,  $\mathbf{p}_0 = (\frac{1}{2}, \frac{1}{2}, 0)^T$ ,  $\mathbf{p} = (p_0, p_1, 1 - p_0 - p_1)^T$ , and  $\mathbb{I} = (1, 1, 1)^T$ . This bound is reached by the EnTO with Kraus operators,

$$K^{(n)} = \sum_{i,j=0:i-j=n}^2 \sqrt{G_{ij}^*} |i\rangle\langle j|, \quad (11)$$

where  $n = -2, -1, 0, 1, 2$  and  $G_{ij}^*$  denotes the elements of the optimal transition matrix  $G^*$  which reach the maximum in Eq. (10). We analytically solve the problem in Eq. (10) (see Appendix A for details) and present the result in Fig. 1. As shown in this figure, the extreme states of the EnTO cone is continuous, namely, a small perturbation in the output population would only result in a small variance of the maximum value of  $|\rho_{10}|$ .

### B. Optimal output coherence via TO

In order to derive the maximal value of  $|\rho_{10}|$  via TO, we start from the general form of the Hamiltonian of the heat bath,

$$H_R = \sum_{E_R} E_R \Pi_{E_R}, \quad (12)$$

where  $E_R$  are the eigenvalues of energy, and  $\Pi_{E_R}$  is the projector to the eigenspace of  $E_R$ . Here we assume that the degeneracy of  $E_R$  is monotonically nondecreasing with  $E_R$ .

Without loss of generality, each eigenvalue  $E_R$  can be expressed as  $E_R = n\hbar\omega + \xi \equiv E(\xi, n)$ , where  $n$  is a non-negative integer and  $\xi \in [0, \hbar\omega)$ . Accordingly,  $\Pi_{E_R} \equiv \Pi_{\xi, n}$ . Now we divide the Hilbert space of  $R$  as  $\mathcal{H}_R = \bigoplus_{\xi} \mathcal{H}_{\xi}$  with  $\mathcal{H}_{\xi} = \bigoplus_n \mathcal{H}_{\xi, n}$  and  $\mathcal{H}_{\xi, n}$  the eigenspace of  $E(\xi, n)$ . The Hamiltonian of the heat bath is then rewritten as  $H_R = \bigoplus_{\xi} H_{\xi}$ , where  $H_{\xi} = \sum_n (n\hbar\omega + \xi) \Pi_{\xi, n}$  is the Hamiltonian acting on  $\mathcal{H}_{\xi}$ . It follows that the thermal state of  $R$  reads

$$\gamma_R = \bigoplus_{\xi} p_{\xi} \gamma_{\xi}, \quad (13)$$

where  $\gamma_{\xi} = e^{-\beta H_{\xi}} / \text{Tr}(e^{-\beta H_{\xi}})$  is the thermal state of  $H_{\xi}$ , and  $p_{\xi} = \text{Tr}(e^{-\beta H_{\xi}}) / [\sum_{\xi'} \text{Tr}(e^{-\beta H_{\xi'}})]$  satisfies  $\sum_{\xi} p_{\xi} = 1$ .

Further, the Hamiltonian of the total system reads

$$H_{SR} = \bigoplus_{\xi} H_{SR_{\xi}}, \quad (14)$$

where  $H_{SR_{\xi}} = H_S \otimes \mathbb{I}_{\xi} + \mathbb{I}_S \otimes H_{\xi}$  is the Hamiltonian acting on subspace  $\mathcal{H}_S \otimes \mathcal{H}_{\xi}$ . Importantly, for  $H_S = \sum_m m\hbar\omega |m\rangle\langle m|$ , the eigenvalues of  $H_{SR_{\xi}}$  and those of  $H_{SR_{\xi'}}$  (where  $\xi' \neq \xi$ ) does not have an overlap. Therefore, any joint unitary which satisfies  $[U, H_{SR}] = 0$  is in a block-diagonal form,

$$U = \bigoplus_{\xi} U_{\xi}, \quad (15)$$

where  $U_{\xi}$  acts on  $\mathcal{H}_S \otimes \mathcal{H}_{\xi}$  and satisfies  $[U_{\xi}, H_{SR_{\xi}}] = 0$ . Now we introduce a lemma which will simplify our subsequent analysis.

*Lemma 1.* For a system with Hamiltonian  $H_S = \sum_m m\hbar\omega |m\rangle\langle m|$ , any thermal operation can be written as  $\mathcal{E}^{\text{TO}} = \sum_{\xi} p_{\xi} \mathcal{E}_{\xi}$ , where  $\mathcal{E}_{\xi}$  is a thermal operation induced by a heat bath with Hamiltonian  $H_{\xi} = \sum_n (n\hbar\omega + \xi) \Pi_{\xi, n}$ .

*Proof.* From Eqs. (13) and (15), a thermal operation acting on  $\rho_S$  reads

$$\begin{aligned} \mathcal{E}^{\text{TO}}(\rho_S) &= \text{Tr}_R[U(\rho_S \otimes \gamma_R)U^{\dagger}] \\ &= \text{Tr}_R \left[ \bigoplus_{\xi} U_{\xi} \left( \rho_S \otimes \bigoplus_{\xi} p_{\xi} \gamma_{\xi} \right) \bigoplus_{\xi} U_{\xi}^{\dagger} \right] \\ &= \sum_{\xi} p_{\xi} \text{Tr}_{R_{\xi}}[U_{\xi}(\rho_S \otimes \gamma_{\xi})U_{\xi}^{\dagger}] \\ &= \sum_{\xi} p_{\xi} \mathcal{E}_{\xi}(\rho_S). \end{aligned} \quad (16)$$

Here  $\mathcal{E}_{\xi}(\rho_S) = \text{Tr}_{R_{\xi}}[U_{\xi}(\rho_S \otimes \gamma_{\xi})U_{\xi}^{\dagger}]$  is a thermal operation induced by a heat bath with Hamiltonian  $H_{\xi}$ , because  $\gamma_{\xi}$  is the thermal state of  $H_{\xi}$ , and  $U_{\xi}$  satisfies  $[U_{\xi}, H_{SR_{\xi}}] = 0$ . This completes the proof.

Next, we will first consider the output population at point (b) in Fig. 1, and show a gap between the maximum values of  $|\rho_{10}|$  which can be preserved by EnTO and by TO. Then we will show that this gap cannot be closed approximately, namely, the extreme state at point (b) of the EnTO cone cannot be reached by TO, even approximately.

#### 1. Maximum coherence via TO for output population of point (b)

At point (b) in Fig. 1, the output population reads  $p_0 = \frac{1-q^2}{2}$  and  $p_1 = \frac{1}{2}$ . For this output population, the transition matrix is uniquely fixed as

$$G^{(b)} = \begin{bmatrix} 1-q^2 & 0 & 1 \\ 0 & 1 & 0 \\ q^2 & 0 & 0 \end{bmatrix}. \quad (17)$$

Due to the equivalence between TO and EnTO in terms of population dynamics, this transition matrix can also be realized by TO. In the following, we will maximize the value of  $|\rho_{10}|$  over all thermal operations which achieve the transition matrix as in Eq. (17). The maximum value of  $|\rho_{10}|$  is denoted as  $|\rho_{10}^{\text{TO}}|$ .

From Lemma 1, any thermal operation acting on our qutrit system can be written as a convex roof of thermal operations  $\mathcal{E}_{\xi}$  based on heat baths with a fixed energetic gap  $H_{\xi} = \sum_n (n\hbar\omega + \xi) \Pi_{\xi, n}$ . Thus we have  $G = \sum_{\xi} p_{\xi} G_{\xi}$ , where  $G_{\xi}$  is the transition matrix corresponding to  $\mathcal{E}_{\xi}$ . Meanwhile, it is easy to check that for  $G^{(b)}$  as in Eq. (17),  $G^{(b)} = \sum_{\xi} p_{\xi} G_{\xi}$  if and only if  $G_{\xi} = G^{(b)}$  for all  $\xi$ . It means that in order to realize the transition matrix as in Eq. (17) by TO, each  $\mathcal{E}_{\xi}$  should also achieve this transition matrix. Hence, for any thermal operation  $\mathcal{E}^{\text{TO}}$  which can realize  $G^{(b)}$ , we have

$$\begin{aligned} |\langle 1 | \mathcal{E}^{\text{TO}}(\rho_0) | 0 \rangle| &\leq \sum_{\xi} p_{\xi} |\langle 1 | \mathcal{E}_{\xi}(\rho_0) | 0 \rangle| \\ &\leq \max_{\xi} |\langle 1 | \mathcal{E}_{\xi}(\rho_0) | 0 \rangle|. \end{aligned} \quad (18)$$

Therefore, the maximum value of  $|\rho_{10}|$  which can be achieved by TO equals to the maximum value realized by thermal operations based on a heat bath with Hamiltonian  $H_\xi$ . Further, the value of  $\xi$  does not affect the state transition of  $S$ , so we set  $\xi = 0$  without loss of generality. The effective Hamiltonian of the heat bath then reads  $H_R = \sum_n n\hbar\omega\Pi_n$ , and the Gibbs state of  $R$  is

$$\gamma_R = \sum_n \gamma_n \Pi_n. \quad (19)$$

The corresponding thermal operations are denoted as  $\mathcal{E}_0^{\text{TO}}$ .

The joint unitary  $U$  is in the block-diagonal form  $U = \bigoplus_{k=0}^{\infty} U^{(k)}$ , where each block  $U^{(k)}$  lives in a subspace with total energy  $k\hbar\omega$ . Precisely,  $U^{(k)}$  is written as

$$U^{(k)} = \sum_{i,j=0}^{\min\{2,k\}} |i\rangle\langle j| \otimes u_{ij}^k, \quad (20)$$

where  $|i\rangle$  and  $|j\rangle$  are energetic states of the system  $S$ ,  $u_{ij}^k$  is a matrix of dimension  $d_{k-i} \times d_{k-j}$ , and  $d_n$  denotes the degeneracy of the energy level  $E_R = n\hbar\omega$  of the heat bath.

Now we define vectors  $\vec{U}_{ij}$ , whose  $k$ th entry is  $(\vec{U}_{ij})_k = \sqrt{\gamma_{k-j}} u_{ij}^k$  with  $k = \max\{i, j\}, \max\{i, j\} + 1, \dots$  and  $i, j = 0, 1, 2$ . For example,

$$\vec{U}_{00} = (\sqrt{\gamma_0} u_{00}^0, \sqrt{\gamma_1} u_{00}^1, \dots), \quad (21)$$

$$\vec{U}_{11} = (\sqrt{\gamma_0} u_{11}^1, \sqrt{\gamma_1} u_{11}^2, \dots). \quad (22)$$

Further, for two such vectors  $\vec{A}$  and  $\vec{B}$  which satisfy that for all  $l$ ,  $A_l$  and  $B_l$  are matrices of the same size, we define the inner product  $(\vec{A}, \vec{B}) \equiv \sum_l \text{Tr}(A_l^\dagger B_l)$ . Then the inner product of vectors  $\vec{U}_{ij}$  and  $\vec{U}_{i'j'}$  (where  $i - i' = j - j'$ ) reads

$$(\vec{U}_{i'j'}, \vec{U}_{ij}) = \sum_{k=\max\{i,j\}}^{\infty} \gamma_{k-j} \text{Tr}(u_{i'j'}^{(k-j+j')\dagger} u_{ij}^k). \quad (23)$$

By substituting Eqs. (19) and (20) to Eq. (2), we obtain that for  $i - i' = j - j'$ ,

$$\langle i | \mathcal{E}_0^{\text{TO}}(|j\rangle\langle j'|) | i' \rangle = (\vec{U}_{i'j'}, \vec{U}_{ij}). \quad (24)$$

In particular, when  $i = i'$  and  $j = j'$ , Eq. (24) reduces to the transition probability from  $j$  to  $i$ ,

$$p_{ij} = G_{ij} = (\vec{U}_{ij}, \vec{U}_{ij}). \quad (25)$$

Moreover, when  $i = j = 1$  and  $i' = j' = 0$ , we have  $\langle 1 | \mathcal{E}_0^{\text{TO}}(|1\rangle\langle 0|) | 0 \rangle = (\vec{U}_{00}, \vec{U}_{11})$ . Remembering that  $|\langle 1 | \mathcal{E}_0^{\text{TO}}(\rho_0) | 0 \rangle| = \frac{1}{2} |\langle 1 | \mathcal{E}_0^{\text{TO}}(|1\rangle\langle 0|) | 0 \rangle|$ , we obtain the following:

$$|\rho_{10}^{\text{TO}}| = \max \frac{1}{2} |(\vec{U}_{00}, \vec{U}_{11})|, \quad \text{s.t. } (\vec{U}_{00}, \vec{U}_{00}) = 1 - q^2, \quad (\vec{U}_{11}, \vec{U}_{11}) = 1. \quad (26)$$

As we prove in Appendix B, the maximum in Eq. (26) is reached by joint unitary operators whose main blocks  $u_{jj}^k$  ( $j = 0, 1, 2, k \geq j$ ) are diagonal matrices  $M_{jj}^k$  whose entries  $\sigma_s(M_{jj}^k)$  ( $s = 0, \dots, d_{k-j} - 1$ ) satisfy  $\sigma_s(M_{jj}^k) \in [0, 1]$  and are in a nonincreasing order.

From Eq. (25), if  $G_{ij} = 0$  for some  $i$  and  $j$ , then  $\vec{U}_{ij} = 0$ , which means that  $u_{ij}^k = \mathbf{0}, \forall k$ . Therefore, the joint unitary in

the thermal operation which realizes the transition matrix as in Eq. (17) and can reach the maximum as in Eq. (26) should be in the following form:  $U^{(0)} = M_{00}^0$ ,

$$U^{(1)} = \left[ \begin{array}{c|c} M_{00}^1 & \mathbf{0} \\ \hline \mathbf{0} & M_{11}^1 \end{array} \right], \quad (27)$$

and for  $k \geq 2$ ,

$$U^{(k)} = \left[ \begin{array}{c|c|c} M_{00}^k & \mathbf{0} & u_{02}^k \\ \hline \mathbf{0} & M_{11}^k & \mathbf{0} \\ \hline u_{20}^k & \mathbf{0} & \mathbf{0} \end{array} \right]. \quad (28)$$

Because each block  $U^{(k)}$  is a unitary matrix, we have  $M_{00}^0 = \mathbb{I}_{d_0}, M_{00}^1 = \mathbb{I}_{d_1}$ , and  $M_{11}^k = \mathbb{I}_{d_{k-1}}$  for  $k \geq 1$ . Further, for  $k \geq 2$ , we prove in Appendix C that

$$\sigma_s(M_{00}^k) = \begin{cases} 1, & s = 0, \dots, d_k - d_{k-2} - 1, \\ 0, & s = d_k - d_{k-2}, \dots, d_k - 1. \end{cases} \quad (29)$$

It follows that

$$\begin{aligned} \text{Tr}(M_{00}^{k\dagger} M_{00}^k) &= \text{Tr}(M_{00}^{k\dagger} M_{11}^{k+1}) \\ &= \begin{cases} d_k & k = 0, 1, \\ d_k - d_{k-2} & k \geq 2. \end{cases} \end{aligned} \quad (30)$$

It can be directly checked that this unitary can indeed achieve the transition matrix as in Eq. (17). For example,

$$\begin{aligned} G_{00} &= (\vec{U}_{00}, \vec{U}_{00}) = \sum_{k=0}^{\infty} \gamma_k \text{Tr}[M_{00}^{k\dagger} M_{00}^k] \\ &= \gamma_0 d_0 + \gamma_1 d_1 + \sum_{k=2}^{\infty} \gamma_k (d_k - d_{k-2}) = 1 - q^2. \end{aligned} \quad (31)$$

For the last equality, we use the fact that  $\gamma_k = q^2 \gamma_{k-2}$  and hence  $\sum_{k=2}^{\infty} \gamma_k d_{k-2} = q^2 \sum_{k=2}^{\infty} \gamma_{k-2} d_{k-2} = q^2$ .

Now we are ready to calculate the following:

$$\begin{aligned} |\rho_{10}^{\text{TO}}| &= \frac{1}{2} (\vec{U}_{00}, \vec{U}_{11}) \\ &= \frac{1}{2} \sum_{k=0}^{\infty} \gamma_k \text{Tr}[M_{00}^{k\dagger} M_{11}^{k+1}] \\ &= \frac{1}{2} G_{00} = \frac{1}{2} (1 - q^2). \end{aligned} \quad (32)$$

However, from Eq. (10), the maximal output coherence via EnTO reads

$$|\rho_{10}^{\text{EnTO}}| = \frac{1}{2} \sqrt{1 - q^2}. \quad (33)$$

Comparing Eqs. (32) and (33), we observe the non-negligible gap between  $|\rho_{10}^{\text{EnTO}}|$  and  $|\rho_{10}^{\text{TO}}|$ ,

$$\begin{aligned} \Delta_{10} &= |\rho_{10}^{\text{EnTO}}| - |\rho_{10}^{\text{TO}}| \\ &= \frac{1}{2} [\sqrt{1 - q^2} - (1 - q^2)]. \end{aligned} \quad (34)$$

It means that the states with  $p_0 = \frac{1-q^2}{2}, p_1 = \frac{1}{2}$  and  $|\rho_{10}| \in (\frac{1-q^2}{2}, \frac{\sqrt{1-q^2}}{2}]$  are in the EnTO cone but not in the TO cone of  $\rho_0$ . This extends the example in Ref. [6], where only one state in  $\mathcal{C}^{\text{EnTO}}(\rho_0) \setminus \mathcal{C}^{\text{TO}}(\rho_0)$  is found.

## 2. Disproof of closure conjecture

Here we prove that the extreme state at point (b) of Fig. 1 in the EnTO cone of  $\rho_0$ , i.e., the state with  $(p_0, p_1, |\rho_{10}|) = \frac{1}{2}(1 - q^2, 1, \sqrt{1 - q^2})$ , cannot be reached by TO approximately, if the temperature is proper such that  $1 - q - q^2 \gg 0$  and  $q \gg 0$  (or equivalently,  $0 \ll q \ll \frac{\sqrt{5}-1}{2}$ ).

Precisely, we consider states in the TO cone of  $\rho_0$  with  $(p_0^\epsilon, p_1^\epsilon, |\rho_{10,\epsilon}|)$ . Here  $p_0^\epsilon, p_1^\epsilon$  is a valid population distribution in the neighbourhood of point (b), i.e.,  $p_0^\epsilon - \frac{1}{2}(1 - q^2) \leq \epsilon_0$  and  $\frac{1}{2} - p_1^\epsilon \leq \epsilon_0$ , where  $\epsilon_0$  is small. We will prove a non-negligible gap between the maximum value of  $|\rho_{10,\epsilon}|$  and  $|\rho_{10}^{\text{EnTO}}| = \frac{1}{2}\sqrt{1 - q^2}$ .

From the linearity of the population dynamics, a perturbation in the output population distribution results from a perturbation in the transition matrix. Together with Lemma 1, it is sufficient to set the Gibbs state of  $R$  as in Eq. (19) and restrict the entries of the perturbed transition matrix  $G_\epsilon^{(b)}$  as  $|G_{ij,\epsilon}^{(b)} - G_{ij}^{(b)}| \leq \epsilon$ . Therefore, we define

$$\begin{aligned} |\rho_{10,\epsilon}^{\text{TO}}| &= \max \frac{1}{2} |(\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)|, \\ \text{s.t. } & |(1 - q^2) - (\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{00}^\epsilon)| \leq \epsilon, \\ & 1 - (\bar{\mathcal{U}}_{11}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon) \leq \epsilon. \end{aligned} \quad (35)$$

Here, the two restrictions come from  $|G_{00,\epsilon}^{(b)} - G_{00}^{(b)}| \leq \epsilon$  and  $|G_{11,\epsilon}^{(b)} - G_{11}^{(b)}| \leq \epsilon$ , respectively. The following inequality is the main result of this section:

$$\begin{aligned} \Delta_{10}^\epsilon &\equiv |\rho_{10}^{\text{EnTO}}| - |\rho_{10,\epsilon}^{\text{TO}}| \\ &> \frac{1}{4}(1 - \sqrt{1 - q^2})^2(1 - q - q^2)(1 - \delta) - 2\epsilon, \end{aligned} \quad (36)$$

where  $\delta$  is small. It means that when the temperature is chosen properly such that  $q$  and  $1 - q - q^2$  are not small, the output coherence  $|\rho_{10}^{\text{EnTO}}| = \frac{1}{2}\sqrt{1 - q^2}$  cannot be reached approximately by thermal operations which realizes a perturbed population dynamics.

In the following, we sketch the proof of Eq. (36), and leave the rigorous proof to Appendix D.

From Lemma 2 in Appendix B, the maximum in Eq. (35) is reached by joint unitary operators with  $u_{jj,\epsilon}^k = M_{jj,\epsilon}^k$ , where  $M_{jj,\epsilon}^k$  are diagonal matrices with non-negative entries in a nonincreasing order. Hence  $|(\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)| = (\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon) = (\bar{\mathcal{U}}_{11}^\epsilon, \bar{\mathcal{U}}_{00}^\epsilon)$ . Now we define

$$\alpha = \frac{(\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)}{(\bar{\mathcal{U}}_{11}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)}, \quad \bar{\mu} = \bar{\mathcal{U}}_{00}^\epsilon - \alpha \bar{\mathcal{U}}_{11}^\epsilon. \quad (37)$$

Direct calculation leads to

$$p_{010}^\epsilon p_{111}^\epsilon - (\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)^2 = p_{111}^\epsilon (\bar{\mu}, \bar{\mu}), \quad (38)$$

where  $p_{010}^\epsilon = (\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{00}^\epsilon) \in [1 - q^2 - \epsilon, 1 - q^2 + \epsilon]$  and  $p_{111}^\epsilon = (\bar{\mathcal{U}}_{11}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon) \in [1 - \epsilon, 1]$ . Clearly,  $\sqrt{p_{010}^\epsilon p_{111}^\epsilon} \geq (\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)$ . Together with  $p_{010}^\epsilon < p_{111}^\epsilon$ , we obtain

$$\frac{1}{2}\sqrt{p_{010}^\epsilon p_{111}^\epsilon} - \frac{1}{2}(\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon) > \frac{1}{4}(\bar{\mu}, \bar{\mu}). \quad (39)$$

Because  $\frac{1}{2}\sqrt{p_{010}^\epsilon p_{111}^\epsilon}$  is  $\epsilon$ -close to  $|\rho_{10}^{\text{EnTO}}|$ , and the maximum value of  $\frac{1}{2}(\bar{\mathcal{U}}_{00}^\epsilon, \bar{\mathcal{U}}_{11}^\epsilon)$  equals to  $|\rho_{10,\epsilon}^{\text{TO}}|$ , Eq. (39) means that the gap  $\Delta_{10}^\epsilon$  cannot be closed if  $(\bar{\mu}, \bar{\mu})$  is not small.

From the definition as in Eq. (37) and the fact that  $p_{111}^\epsilon \in [1 - \epsilon, 1]$ , we obtain the following:

$$(\bar{\mu}, \bar{\mu}) > \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [\sigma_s(M_{00,\epsilon}^k) - \alpha]^2 - 2\alpha\epsilon. \quad (40)$$

In order to evaluate the right-hand side of Eq. (40), we assume the heat bath is large and satisfies the following.

(i) There is a set of energy levels  $\mathcal{R}$ , such that  $\sum_{E_R \in \mathcal{R}} \text{Tr}(\gamma_R \Pi_{E_R}) = 1 - \delta$ , where  $\delta$  is small.

(ii) For any  $E_R \in \mathcal{R}$ ,  $E_R \pm m\hbar\omega \in \mathcal{R}$ , where  $m = 1, 2$ .

(iii) For  $E_R \in \mathcal{R}$ , the degeneracies  $g(E_R)$  satisfy  $|\frac{g(E_R - m\hbar\omega)}{g(E_R)q^m} - 1| \leq \delta$ , where  $m = 1, 2$ .

These assumptions have been employed in Ref. [3] to derive the famous thermo-majorization relation.

By assumption (iii) and the condition  $1 - q - q^2 \gg 0$ , we have

$$d_k - d_{k-1} - d_{k-2} \geq [(1 - q - q^2) - (q + q^2)\delta]d_k > 0, \quad (41)$$

for  $k\hbar\omega \in \mathcal{R}$ . The unitarity of the block  $U_\epsilon^{(k)}$  then ensures that at least  $d_k - (d_{k-1} + d_{k-2})$  singular values of  $M_{00,\epsilon}^k$  are equal to 1. By subtracting some non-negative terms from the right-hand side of Eq. (40), we obtain the following:

$$\begin{aligned} (\bar{\mu}, \bar{\mu}) &> \sum_{k:k\hbar\omega \in \mathcal{R}} \gamma_k \sum_{s:\sigma_s(M_{00,\epsilon}^k)=1} [\sigma_s(M_{00,\epsilon}^k) - \alpha]^2 - 2\alpha\epsilon \\ &= \sum_{k:k\hbar\omega \in \mathcal{R}} \gamma_k (d_k - d_{k-1} - d_{k-2})(1 - \alpha)^2 - 2\alpha\epsilon \\ &\geq (1 - \alpha)^2(1 - q - q^2)(1 - \delta) - 2\alpha\epsilon. \end{aligned} \quad (42)$$

Here  $\alpha \leq \sqrt{\frac{p_{010}^\epsilon}{p_{111}^\epsilon}} \leq \sqrt{\frac{1 - q^2 + \epsilon}{1 - \epsilon}} \equiv \alpha(\epsilon)$  by definition. Clearly,  $\alpha(\epsilon)$  is  $\epsilon$ -close to  $\sqrt{1 - q^2}$ . Together with Eq. (39), we arrive at Eq. (36).

It is worth mentioning that, although our derivation heavily depends on the condition  $1 - q - q^2 \gg 0$ , this condition is by no means necessary for the gap. Moreover, the bound to the gap as in Eq. (36) is not tight. Nevertheless, these results are sufficient for the purpose of this paper, which is to disprove the closure conjecture with a counterexample. We will leave the explicit problems, such as necessary conditions and tighter bounds on the gap, to future work.

## V. CONCLUSIONS

We have disproved the closure conjecture with an analytic counterexample. We derive the EnTO cone of a given qutrit state, and calculate the optimal coherence preserved by TO for a given output population distribution. We also evaluate the upper bound on the coherence preserved by TO, if a small perturbation on the output population distribution is allowed. By doing so, we discover a state conversion under EnTO which cannot be approximated by TO.

Our findings show that thermal operations and enhanced thermal operations can lead to different laws of coherence

evolution. Further, the methods we developed here can be used to evaluate the upper bound on the output coherence via thermal operations. Thus our results can contribute to studying the restrictions on coherence dynamics under TO.

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### APPENDIX A: ANALYTIC EXPRESSION FOR ENTO CONE OF $\rho_0$

In this section, we analytically solve the optimization problem in Eq. (10). By observing the objective function in Eq. (10), we notice that the maximum can be taken at the point  $(G_{00}, G_{11})$  where both  $G_{00}$  and  $G_{11}$  are maximal in the feasible region.

Here we first derive the feasible region of optimization. The transition conditions  $G\boldsymbol{\gamma} = \boldsymbol{\gamma}$ ,  $G\boldsymbol{p}_0 = \boldsymbol{p}$  give us the following equations:

$$G_{01} = 2p_0 - G_{00}, \quad (\text{A1})$$

$$G_{10} = 2p_1 - G_{11}, \quad (\text{A2})$$

$$G_{02} = [1 - G_{00} - q(2p_0 - G_{00})]/q^2, \quad (\text{A3})$$

$$G_{12} = [1 - qG_{11} - (2p_1 - G_{11})]/q^2, \quad (\text{A4})$$

where  $(p_0, p_1)$  is a fixed point in the hexagon in Fig. 1. By applying the condition that above entries lie in  $[0,1]$ , we give the bounds of  $G_{00}$  and  $G_{11}$  as

$$G_{00}^{\text{lb}} \leq G_{00} \leq G_{00}^{\text{ub}} := \min \left\{ 2p_0, 1, \frac{1 - 2qp_0}{1 - q} \right\}, \quad (\text{A5})$$

$$G_{11}^{\text{lb}} \leq G_{11} \leq G_{11}^{\text{ub}} := \min \left\{ 2p_1, 1, \frac{q^2 + 2p_1 - 1}{1 - q} \right\}. \quad (\text{A6})$$

Here we omit the expressions for the lower bounds  $G_{00}^{\text{lb}}$  and  $G_{11}^{\text{lb}}$ , because the central question here is to find the upper bound for  $\sqrt{G_{00}G_{11}}$ . Further, by applying the condition  $G^T \mathbb{I} = \mathbb{I}$ , we get that  $G_{00} + G_{10}$ ,  $G_{01} + G_{11}$  and  $G_{02} + G_{12}$  also lie in  $[0,1]$ . It follows that

$$\text{lb} \leq G_{00} - G_{11} \leq \text{ub}, \quad (\text{A7})$$

where

$$\text{lb} := \max \left\{ -2p_1, 2p_0 - 1, \frac{1 - 2(qp_0 + p_1)}{1 - q} + q \right\}, \quad (\text{A8})$$

$$\text{ub} := \min \left\{ 1 - 2p_1, 2p_0, \frac{1 + q - 2(qp_0 + p_1)}{1 - q} \right\}. \quad (\text{A9})$$

The combination of Eqs. (A5), (A6), and (A7) gives the necessary and sufficient condition for entries  $G_{00}$  and  $G_{11}$  in a feasible transition matrix  $G$ . The reason for sufficiency is as follows. If one starts from a given pair of  $G_{00}$  and  $G_{11}$  which satisfies these three equations, other entries of  $G$  are fixed by Eqs. (A1)–(A4) and  $G^T \mathbb{I} = \mathbb{I}$ . Further, the transition matrix  $G$  as such satisfies all of the four conditions in Eq. (10).

Next, we calculate the maximum in Eq. (10) for the following three cases.

Case 1.  $\text{lb} \leq G_{00}^{\text{ub}} - G_{11}^{\text{ub}} \leq \text{ub}$ .

Case 2.  $\text{lb} > G_{00}^{\text{ub}} - G_{11}^{\text{ub}}$ .

Case 3.  $G_{00}^{\text{ub}} - G_{11}^{\text{ub}} > \text{ub}$ .

For Case 1, it is easy to see that  $G_{00}^* = G_{00}^{\text{ub}}$  and  $G_{11}^* = G_{11}^{\text{ub}}$ . For Case 2, the upper bound  $G_{11}^{\text{ub}}$  cannot be reached by  $G_{11}^*$  because of Eq. (A7), so we have  $G_{00}^* = G_{00}^{\text{ub}}$  and  $G_{11}^* = G_{00}^{\text{ub}} - \text{lb}$ . Similarly, for Case 3, we have  $G_{00}^* = G_{11}^{\text{ub}} + \text{ub}$  and  $G_{11}^* = G_{11}^{\text{ub}}$ . To sum up, we arrive at the following analytic solution to Eq. (10).

$$\frac{1}{2} \begin{cases} \sqrt{G_{00}^{\text{ub}} G_{11}^{\text{ub}}}, & \text{lb} \leq G_{00}^{\text{ub}} - G_{11}^{\text{ub}} \leq \text{ub}; \\ \sqrt{G_{00}^{\text{ub}} (G_{00}^{\text{ub}} - \text{lb})}, & \text{lb} > G_{00}^{\text{ub}} - G_{11}^{\text{ub}}; \\ \sqrt{(G_{11}^{\text{ub}} + \text{ub}) G_{11}^{\text{ub}}}, & G_{00}^{\text{ub}} - G_{11}^{\text{ub}} > \text{ub}. \end{cases} \quad (\text{A10})$$

This solution is visualized in Fig. 1.

### APPENDIX B: OPTIMAL JOINT UNITARY

In this section, we will show that, among all of the joint unitary operators which achieve a given transition matrix, the maximum value of  $|\rho_{10}|$  is reached by the unitary operators whose main blocks are diagonal with non-negative entries in a nonincreasing order. Precisely, we will prove the following lemma.

*Lemma 2.* Let  $H_S = \sum_{m=0}^2 m\hbar\omega |m\rangle \langle m|$  and  $H_R = \sum_n n\hbar\omega \Pi_n$  be the Hamiltonian of the main system  $S$  and that of the heat bath  $R$ , respectively, and  $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$  be the initial state of  $S$ . For any (energy-preserving) joint unitary  $U$ , one can always find another (energy-preserving) joint unitary  $\tilde{U}$ , which satisfies the following.

(a)  $\tilde{u}_{jj}^k = M_{jj}^k$ , where  $M_{jj}^k$  are diagonal matrices with non-negative entries in a nonincreasing order.

(b)  $(\tilde{u}_{ij}, \tilde{u}_{ij}) = (\tilde{u}_{ij}, \tilde{u}_{ij})$ , which means that  $U$  and  $\tilde{U}$  lead to the same transition matrix.

(c)  $|\langle \tilde{u}_{00}, \tilde{u}_{11} \rangle| \leq |\langle u_{00}, u_{11} \rangle|$ , which means that the output coherence  $|\rho_{10}|$  achieved by  $\tilde{U}$  is no less than that achieved by  $U$ .

The rest of this section will be devoted to the proof of this lemma. Following the method as in Ref. [6], we perform the singular value decomposition (SVD) to the main blocks:

$$u_{jj}^k = A_{jj}^k M_{jj}^k B_{jj}^{k\dagger}, \quad (\text{B1})$$

where  $A_{jj}^k$  and  $B_{jj}^k$  are unitary matrices, and  $M_{jj}^k$  are diagonal matrices with singular values (which are non-negative numbers) in a nonincreasing order as entries. By introducing two unitary matrices  $A \equiv \bigoplus_{k=0}^{\infty} [\bigoplus_{j=0}^{\min(k,2)} A_{jj}^k]$  and  $B \equiv \bigoplus_{k=0}^{\infty} [\bigoplus_{j=0}^{\min(k,2)} B_{jj}^k]$ , we define a new joint unitary,

$$\tilde{U} = A^\dagger U B. \quad (\text{B2})$$

Similar to Eq. (15),  $\tilde{U}$  can be expressed as  $\tilde{U} = \bigoplus_{k=0}^{\infty} \tilde{U}^{(k)}$  with  $\tilde{U}^{(k)} = \sum_{i,j=0}^{\min(k,2)} |i\rangle \langle j| \otimes \tilde{u}_{ij}^k$ . Then Eq. (B2) leads to

$$\tilde{u}_{ij}^k = A_{ii}^{k\dagger} u_{ij}^k B_{jj}^k. \quad (\text{B3})$$

Now we are ready to prove that  $\tilde{U}$  satisfies the conditions (a), (b), and (c) as mentioned above.

By substituting Eq. (B1) to Eq. (B3) with  $i = j$ , we have  $\tilde{u}_{jj}^k = A_{jj}^{k\dagger} A_{jj}^k M_{jj}^k B_{jj}^{k\dagger} B_{jj}^k = M_{jj}^k$ . Thus, condition (a) holds.

As for condition (b), we employ the definition of inner product as in Eq. (23), and obtain the following:

$$\begin{aligned}
 & (\tilde{u}_{ij}, \tilde{u}_{ij}) \\
 &= \sum_{k=\max\{i,j\}}^{\infty} \gamma_{k-j} \text{Tr}(\tilde{u}_{ij}^{k\dagger} \tilde{u}_{ij}^k) \\
 &= \sum_{k=\max\{i,j\}}^{\infty} \gamma_{k-j} \text{Tr}(B_{jj}^{k\dagger} u_{ij}^{k\dagger} A_{ii}^k A_{ii}^{k\dagger} u_{ij}^k B_{jj}^k) \\
 &= \sum_{k=\max\{i,j\}}^{\infty} \gamma_{k-j} \text{Tr}(u_{ij}^{k\dagger} u_{ij}^k) \\
 &= (\tilde{u}_{ij}, \tilde{u}_{ij}). \tag{B4}
 \end{aligned}$$

Then from Eq. (25), this equality means that  $U$  and  $\tilde{U}$  leads to the same transition matrix.

In order to prove condition (c), we employ the following lemma, which was proved in Refs. [19,20] and introduced to this problem in Ref. [6].

*Lemma 3.* If  $X$  and  $Y$  are  $d \times d$  complex matrices,  $W$  and  $V$  are  $d \times d$  unitary matrices, and  $\sigma_1 \geq \sigma_2 \geq \dots \geq \sigma_d \geq 0$  denotes ordered singular values, then

$$|\text{Tr}(WXYV)| \leq \sum_{s=0}^{d-1} \sigma_s(X) \sigma_s(Y)$$

and the equality always exist for some  $W$  and  $V$ .

From this lemma, we have  $|\text{Tr}[u_{00}^{(k-1)\dagger} u_{11}^k]| \leq \sum_{s=0}^{d_k-1} \sigma_s(u_{00}^{(k-1)\dagger}) \sigma_s(u_{11}^k) = \text{Tr}[M_{00}^{(k-1)\dagger} M_{11}^k]$ , and hence,

$$\begin{aligned}
 |(\tilde{u}_{00}, \tilde{u}_{11})| &\leq \sum_{k=1}^{\infty} \gamma_{k-1} |\text{Tr}[u_{00}^{k\dagger} u_{11}^{k-1}]| \\
 &\leq \sum_{k=1}^{\infty} \gamma_{k-1} \text{Tr}[M_{00}^{(k-1)\dagger} M_{11}^k] \\
 &= |(\tilde{u}_{00}, \tilde{u}_{11})|. \tag{B5}
 \end{aligned}$$

This completes the proof of Lemma 2.

### APPENDIX C: PROOF OF EQ. (29)

In this section, we will prove Eq. (29) in the main text. For  $k \geq 2$ , we have

$$U^{(k)} = \begin{bmatrix} M_{00}^k & \mathbf{0} & u_{02}^k \\ \mathbf{0} & M_{11}^k & \mathbf{0} \\ u_{20}^k & \mathbf{0} & \mathbf{0} \end{bmatrix}, \tag{C1}$$

where  $M_{11}^k = \mathbb{I}_{d_{k-1}}$  and  $M_{00}^k$  is a diagonal matrix with non-negative entries in a nonincreasing order. We will first prove that the number of zero rows in  $u_{02}^k$  equals exactly to  $d_k - d_{k-2}$ , and then show that there are  $d_k - d_{k-2}$  entries of  $M_{00}^k$  which equal to one, while the rest of the entries of  $M_{00}^k$  all equal to zero.

Here and following, we denote the  $s$ th row of a matrix  $X$  as  $\vec{r}_s(X)$ , and the  $l$ th column of  $X$  as  $\vec{c}_l(X)$ . From

the unitarity of  $U^{(k)}$ , we have  $\vec{r}_s(U^{(k)})^* \cdot \vec{r}_{s'}(U^{(k)}) = \delta_{ss'}$  and  $\vec{c}_l(U^{(k)})^* \cdot \vec{c}_{l'}(U^{(k)}) = \delta_{ll'}$ . Our derivations are based on these two equations.

For  $d_k + d_{k-1} \leq l, l' < d_k + d_{k-1} + d_{k-2}$ , we have  $\delta_{ll'} = \vec{c}_l(U^{(k)})^* \cdot \vec{c}_{l'}(U^{(k)}) = \vec{c}_l(u_{02}^k)^* \cdot \vec{c}_{l'}(u_{02}^k)$ . It means that the  $d_{k-2}$  columns of  $u_{02}^k$  are nontrivial and linearly independent. Therefore, at least  $d_{k-2}$  rows of  $u_{02}^k$  are nontrivial.

For  $0 \leq s, s' \leq d_k - 1$  and  $s \neq s'$ , we have  $0 = \vec{r}_s(U^{(k)})^* \cdot \vec{r}_{s'}(U^{(k)}) = \vec{r}_s(u_{02}^k)^* \cdot \vec{r}_{s'}(u_{02}^k)$ . Hence, there are at most  $d_{k-2}$  nontrivial rows in  $u_{02}^k$ , and the rest of the rows of  $u_{02}^k$  have to be zero.

Therefore, the number of zero rows in  $u_{02}^k$  is  $d_k - d_{k-2}$ . Then we have

$$\vec{r}_s(u_{02}^k) \begin{cases} = 0, & 0 \leq s \leq d_k - d_{k-2} - 1, \\ \neq 0, & d_k - d_{k-2} \leq s \leq d_k - 1. \end{cases} \tag{C2}$$

It follows that for  $0 \leq s \leq d_k - d_{k-2} - 1$ , we have  $1 = \vec{r}_s(U^{(k)})^* \cdot \vec{r}_s(U^{(k)}) = [\sigma_s(M_{00}^k)]^2$ , and hence  $\sigma_s(M_{00}^k) = 1$ .

For  $d_k - d_{k-2} \leq s \leq d_k - 1$  and  $d_k + d_{k+1} \leq l \leq d_k + d_{k+1} + d_{k+2}$ , the scalar product  $\vec{c}_s(U^{(k)})^* \cdot \vec{c}_l(U^{(k)}) = 0$  gives  $\sigma_s(M_{00}^k) \cdot \vec{r}_s(u_{02}^k) = 0$ . Meanwhile,  $\vec{r}_s(u_{02}^k) \neq 0$ , so we have  $\sigma_s(M_{00}^k) = 0$ .

### APPENDIX D: DETAILED PROOF OF EQ. (36)

In this section, we prove Eq. (36) under the condition  $1 - q - q^2 \gg 0$  (or equivalently,  $q \ll \frac{\sqrt{5}-1}{2}$ ) and the assumptions (i)–(iii) on the heat bath.

Here we start from Eq. (38) in the main text. It follows that  $\sqrt{p_{010}^\epsilon p_{111}^\epsilon} \geq (\tilde{u}_{00}^\epsilon, \tilde{u}_{11}^\epsilon)$  and hence,

$$\frac{1}{2} \sqrt{p_{010}^\epsilon p_{111}^\epsilon} - \frac{1}{2} (\tilde{u}_{00}^\epsilon, \tilde{u}_{11}^\epsilon) \geq \frac{1}{4} \sqrt{\frac{p_{111}^\epsilon}{p_{010}^\epsilon}} (\bar{\mu}, \bar{\mu}). \tag{D1}$$

Because  $\sqrt{p_{010}^\epsilon p_{111}^\epsilon} \leq \sqrt{1 - q^2 + \epsilon} = \sqrt{1 - q^2} + \frac{\epsilon}{2\sqrt{1 - q^2}} + O(\epsilon^2)$ , and  $\sqrt{\frac{p_{111}^\epsilon}{p_{010}^\epsilon}} \geq \sqrt{\frac{1 - \epsilon}{1 - q^2 + \epsilon}} = \frac{1}{\sqrt{1 - q^2}} - \frac{2 - q^2}{2(1 - q^2)^{3/2}} \epsilon + O(\epsilon^2)$ , the above equation becomes

$$\Delta_{10}^\epsilon \geq \frac{1}{4} \left( 1 - \frac{2 - q^2}{2(1 - q^2)} \epsilon \right) \frac{(\bar{\mu}, \bar{\mu})}{\sqrt{1 - q^2}} - \frac{\epsilon}{4\sqrt{1 - q^2}} + O(\epsilon^2). \tag{D2}$$

From the definition as in Eq. (37) and the fact that  $p_{111}^\epsilon \in [1 - \epsilon, 1]$ , we obtain the following:

$$(\bar{\mu}, \bar{\mu}) > \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [\sigma_s(M_{00,\epsilon}^k) - \alpha]^2 - 2\alpha\epsilon. \tag{D3}$$

The reason is as follows. First, because  $1 - p_{111}^\epsilon \leq \epsilon$ ,  $\sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} 1 = 1$ , and  $p_{111}^\epsilon = \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [\sigma_s(M_{11,\epsilon}^k)]^2$ , we have the following:

$$\begin{aligned}
 & \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [1 - \sigma_s(M_{11,\epsilon}^k)] \\
 & \leq \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [1 - \sigma_s(M_{11,\epsilon}^k)]^2 \leq \epsilon. \tag{D4}
 \end{aligned}$$

By the definition as in Eq. (37),

$$\begin{aligned}
 (\bar{\mu}, \bar{\mu}) &= \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [\sigma_s(M_{00,\epsilon}^k) - \alpha \sigma_s(M_{11,\epsilon}^k)]^2 \\
 &\geq \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} \{[\sigma_s(M_{00,\epsilon}^k) - \alpha]^2 - 2\alpha[1 - \sigma_s(M_{11,\epsilon}^k)]\} \\
 &\geq \sum_{k=0}^{\infty} \gamma_k \sum_{s=0}^{d_k-1} [\sigma_s(M_{00,\epsilon}^k) - \alpha]^2 - 2\alpha\epsilon. \tag{D5}
 \end{aligned}$$

Here for the second line, we have used  $\sigma_s(M_{11,\epsilon}^k) \in [0, 1]$  and  $\sigma_s(M_{00,\epsilon}^k) - \alpha \geq -1$ .

Now we consider the  $k$ th block of  $U^\epsilon$ ,

$$U_\epsilon^{(k)} = \begin{bmatrix} M_{00,\epsilon}^k & u_{01,\epsilon}^k & u_{02,\epsilon}^k \\ u_{10,\epsilon}^k & M_{11,\epsilon}^k & u_{12,\epsilon}^k \\ u_{20,\epsilon}^k & u_{21,\epsilon}^k & u_{22,\epsilon}^k \end{bmatrix}, \tag{D6}$$

where  $k\hbar\omega \in \mathcal{R}$ . By assumption (iii) and the condition  $1 - q - q^2 \gg 0$ , we have  $d_k > d_{k-1} + d_{k-2}$ . For  $0 \leq s, s' \leq d_k - 1$  and  $s \neq s'$ , the unitarity of  $U_\epsilon^{(k)}$  implies

$$\begin{aligned}
 0 &= \bar{r}_s(U_\epsilon^{(k)})^* \cdot \bar{r}_{s'}(U_\epsilon^{(k)}) \\
 &= \bar{r}_s([u_{01,\epsilon}^k | u_{02,\epsilon}^k])^* \cdot \bar{r}_{s'}([u_{01,\epsilon}^k | u_{02,\epsilon}^k]). \tag{D7}
 \end{aligned}$$

Here  $[u_{01,\epsilon}^k | u_{02,\epsilon}^k]$  is a rectangle matrix of dimension  $d_k \times (d_{k-1} + d_{k-2})$ , which is composed of two blocks  $u_{01,\epsilon}^k$  and  $u_{02,\epsilon}^k$ . Hence there are at most  $(d_{k-1} + d_{k-2})$  nontrivial rows of  $[u_{01,\epsilon}^k | u_{02,\epsilon}^k]$  satisfying Eq. (D7), and the rest of the  $d_k - (d_{k-1} + d_{k-2})$  rows are zero. For  $\bar{r}_s([u_{01,\epsilon}^k | u_{02,\epsilon}^k]) = 0$ , the equality  $\bar{r}_s(U_\epsilon^{(k)})^* \cdot \bar{r}_s(U_\epsilon^{(k)}) = 1$  leads to  $\sigma_s(M_{00,\epsilon}^k) = 1$ . Therefore, at least  $d_k - (d_{k-1} + d_{k-2})$  singular values of  $M_{00,\epsilon}^k$  are equal to 1. By subtracting some non-negative terms from the right-hand side of Eq. (D3), we obtain the

following:

$$\begin{aligned}
 (\bar{\mu}, \bar{\mu}) &> \sum_{k:k\hbar\omega \in \mathcal{R}} \gamma_k \sum_{s:\sigma_s(M_{00,\epsilon}^k)=1} [\sigma_s(M_{00,\epsilon}^k) - \alpha]^2 - 2\alpha\epsilon \\
 &= \sum_{k:k\hbar\omega \in \mathcal{R}} \gamma_k (d_k - d_{k-1} - d_{k-2})(1 - \alpha)^2 - 2\alpha\epsilon \\
 &\geq (1 - \alpha)^2(1 - q - q^2)(1 - \delta) - 2\alpha\epsilon. \tag{D8}
 \end{aligned}$$

For the last line, we have used  $\gamma_k = \gamma_{k-m} q^m$  with  $m = 0, 1, 2$ , and hence,  $\sum_{k:k\hbar\omega \in \mathcal{R}} \gamma_k d_{k-m} = q^m \sum_{k:k\hbar\omega \in \mathcal{R}} \gamma_{k-m} d_{k-m} = q^m(1 - \delta)$ . By definition,  $\alpha \leq \sqrt{p_{0|0}^\epsilon/p_{1|1}^\epsilon} \leq \sqrt{\frac{1-q^2+\epsilon}{1-\epsilon}} \equiv \alpha(\epsilon)$ , and hence  $\alpha \leq \sqrt{1 - q^2} + O(\epsilon)$ , and

$$\begin{aligned}
 (1 - \alpha)^2 &\geq [1 - \alpha(\epsilon)]^2 \\
 &= [1 - \alpha(\epsilon)]^2|_{\epsilon=0} - 2[1 - \alpha(\epsilon)] \frac{d\alpha(\epsilon)}{d\epsilon} \Big|_{\epsilon=0} \epsilon + O(\epsilon^2) \\
 &= [1 - \sqrt{1 - q^2}]^2 \\
 &\quad - \frac{(1 - \sqrt{1 - q^2})(2 - q^2)}{\sqrt{1 - q^2}} \epsilon + O(\epsilon^2). \tag{D9}
 \end{aligned}$$

Then Eq. (D8) becomes

$$\begin{aligned}
 (\bar{\mu}, \bar{\mu}) &> [1 - \sqrt{1 - q^2}]^2(1 - q - q^2)(1 - \delta) - f_1(q)\epsilon + O(\epsilon^2), \tag{D10}
 \end{aligned}$$

where  $f_1(q) = 2\sqrt{1 - q^2} + \frac{(1 - \sqrt{1 - q^2})(2 - q^2)(1 - q - q^2)}{\sqrt{1 - q^2}}$ . Together with Eq. (D2), we obtain

$$\begin{aligned}
 \Delta_{10}^\epsilon &> \frac{[1 - \sqrt{1 - q^2}]^2(1 - q - q^2)(1 - \delta)}{4\sqrt{1 - q^2}} \\
 &\quad - \frac{1}{4}f(q)\epsilon + O(\epsilon^2), \tag{D11}
 \end{aligned}$$

where  $f(q) = 2 + \frac{[1 - (1 - q^2)^2](1 - q - q^2)}{2(1 - q^2)^{3/2}} + \frac{1}{\sqrt{1 - q^2}}$ . For  $0 < q < \frac{\sqrt{5}-1}{2}$ , it holds that  $1 < \frac{1}{\sqrt{1 - q^2}} < 2$  and  $f(q) < 8$ . Then we obtain the simpler form as in Eq. (36).

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