Catalytic transformations with finite-size environments: applications to cooling and thermometry

Ivan Henao[1, *] and Raam Uzdin[1, †]

[1]*Fritz Haber Research Center for Molecular Dynamics, Institute of Chemistry,*
The Hebrew University of Jerusalem, Jerusalem 9190401, Israel

The laws of thermodynamics are usually formulated under the assumption of infinitely large environments. While this idealization facilitates theoretical treatments, real physical systems are always finite and their interaction range is limited. These constraints have consequences on important tasks such as cooling, not directly captured by the second law of thermodynamics. Here, we study catalytic transformations that cannot be achieved when a system exclusively interacts with a finite environment. Our core result consists of constructive conditions for these transformations, which include the corresponding global unitary operation and the explicit states of all the systems involved. From this result we present various findings regarding the use of catalysts for cooling. First, we show that catalytic cooling is always possible if the dimension of the catalyst is sufficiently large. In particular, the cooling of a qubit using a hot qubit can be maximized with a catalyst as small as a three-level system. We also identify catalytic enhancements for tasks whose implementation is possible without a catalyst. For example, we find that in a multiqubit setup catalytic cooling based on a three-body interaction outperforms standard (non-catalytic) cooling using higher order interactions. Another advantage is illustrated in a thermometry scenario, where a qubit is employed to probe the temperature of the environment. In this case, we show that a catalyst allows to surpass the optimal temperature estimation attained only with the probe.

I. INTRODUCTION

In the field of Chemistry, catalysts are substances that can be used to assist a chemical reaction without being consumed in the process. This simple but powerful principle has also found applications in areas of quantum information [1–14] and quantum thermodynamics [15–24], where catalysts are quantum systems that enable the implementation of otherwise impossible transformations. For example, transformations that are forbidden under local operations and classical communication (LOCC) become possible once a suitable entangled state is employed as catalyst [1]. Regarding the technical aspect, catalytic transformations have often been addressed using the concept of "catalytic majorization" [2–4] and related extensions [19, 25]. A distinctive feature of this approach is that it provides general conditions for the existence of a catalyst state that enables the transformation. However, typically such state and the corresponding implementation are not explicitly given.

In quantum thermodynamics, it has been shown that catalysts extend the set of state transitions that a system can undergo in the presence of a thermal environment [15, 19]. These transformations are performed through global unitaries that couple the environment with the rest of the system, and preserve the energy of the total setup. Originally introduced without the inclusion of catalysts, such conditions define maps on the system known as "thermal operations" [26–34]. Although a few studies have addressed thermal operations with finite environments [31, 35, 36], the most general transformations

derived within this framework rely on the possibility of interactions with arbitrarily large baths (determined by arbitrary bath Hamiltonians)[26, 32] — an assumption that has also been adopted in the case of catalytic thermal operations [15, 18–20, 24]. Here, we consider catalytic transformations where the main system interacts with a catalyst and a finite environment. The transformations result from the application of non-energy preserving unitaries on the total system, and the enviroment may start in a generic state. Moreover, they are explicit, in the sense that explicit unitaries and the corresponding catalyst state are obtained. From a thermodynamic viewpoint, the main motivation is the characterization of conditions to catalytically circumvent cooling limitations due to the finite character of the environment.

The interest in the formalization and quantification of the fundamental limits for cooling has seen a resurgence in the last years [37, 38]. Explicit bounds on the duration of continuous [39, 40] and discrete cooling processes [31, 41, 42] have been provided, thus casting the famous unattainability principle [37, 43] on more firm grounds. Different physical resources needed to achieve very low temperatures have also been unveiled and characterized [18, 37, 39, 44–47]. These include non-equilibrium states for cooling via catalytic thermal operations [18], the (Hilbert space) dimension of microscopic refrigerators [48, 49], and quantum properties such as coherence [50] and entanglement [51]. Besides asymptotic limits for heath bath algorithmic cooling protocols [52, 53], Ref. [45] presents a bound on the attainable ground population of a generic system, using absorption (heat-driven) or power (work-driven) refrigerators in steady state operation. It has also been shown that in some special cases cooling bounds are saturable using finite environments [35, 45], and enhancements due to non-Markovian effects

have been pointed out [35, 54]. The intrinsic limitations concerning the interaction with a small bath were studied in Ref. [36], regarding thermal operations, and in Ref. [44], in the case of general (non-energy preserving) global unitaries. Similarly, no-go theorems show that it is impossible to achieve an arbitrarily high purity by interacting with a finite system that starts in an mixed state [55–57].

In this work, we establish sufficient conditions for transformations that lift cooling restrictions by employing a *finite-dimensional and single-copy* catalyst. When the environment is small, the maximum achievable cooling can be far from the fundamental bounds previously mentioned. In some extreme situations cooling is not even possible, e.g. if the environment is very hot [58]. We characterize such scenario using the notion of passivity [59–62], for the case where the goal is to reduce the mean energy of the system. However, we show that a catalyst can activate cooling not only by decreasing the system energy, but also by increasing its ground state population. The key property of the associated transformations is that they cannot be reproduced by applying random unitary maps [63] on the system-environment compound. Equivalently, this means that the final joint state cannot be obtained by applying a unital map on the initial state (non-unital transformation). The use of catalysts to perform non-unital transformations has been exemplified with a protocol that allows to extract work from a passive state [17]. More recently, it was also shown that such property makes possible to catalytically bypass the Jarzinsky fluctuation theorem [22] The formal tools introduced here are naturally applicable to cooling. However, they also provide a framework for a more comprehensive analysis of catalytic transformations that do not satisfy unitality.

As an additional application we show that a catalyst can be useful to enhance a thermometric task. In thermometry [64], and metrology in general [65–68], the purpose is to employ probabilistic information from measurements of some obervable, in order to estimate a physical parameter such as the temperature of the environment in the case of thermometry. A possible thermometric strategy consists of coupling the environment with a probe, and then measure the probe to extract temperature information [69–79]. Assuming that optimal measurements can be performed, the minimum estimation error corresponds to an optimal unitary interaction between the probe and the environment. In this context, we show such an error can be further reduced by means of a catalyst.

The paper is organized as follows. After motivating the tackled problem with an example (Sect. II), Sect. III characterizes the limitations on cooling for systems and environments of finite, but otherwise arbitrary dimension. In Sect. IV we introduce the formal tools that are employed in the rest of the paper. In particular, our analysis relies on a subset of global unitaries described in this section. By resorting to the concept of majoriza-

tion [80, 81], we can define non-unital transformations as those where the final state of the system is not majorized by its initial state. The first main result characterizes sufficient conditions to implement these transformations via a catalyst. In the same section, we develop a graphical method that provides an intuitive picture of this finding and subsequent derivations. The applications to catalytic and cooling transformations are addressed in Sect. V, where the main results refer to:

- Catalytic cooling with arbitrarily small environments.

- Catalytic cooling of a qubit, where the catalyst enhances the cooling even if the environment is sufficient to perform it .

We provide upper bounds on the catalyst dimension for these transformations, and illustrate them with several examples. In particular, we optimize the cooling of a qubit using another qubit as environment, for catalysts of arbitrary (finite) dimension. When cooling with the environment alone is possible, we illustrate a catalytic enhancement for a two-level system that couples to a three-level environment. In Sect. VI we extend our findings to a scenario where the environment is composed of an arbitrary but fixed number of qubits. We show that if this system is used to cool single qubits, the inclusion of a two-level catalyst can increase the total extracted heat. Moreover, the catalytic strategy generally requires less control on the environment than cooling without using the catalyst. In Sect. VII we show that a two-level catalyst provides a thermometric advantage, when the temperature of a three-level environment is probed by a two-level system. Finally, we present a discussion and open questions in Sect. VIII, and conclude in Sect. IX.

II. EXAMPLE OF IMPOSSIBILITY FOR COOLING USING A SMALL ENVIRONMENT

In the rest of the paper we will frequently call the system to be cooled and the environment "cold object" and "hot object", respectively. Moreover, the ground state of these systems and the catalyst will be denoted using the label "1" instead of "0". This choice is convenient to simplify the notation of other physical quantities that will be defined later. States that describe the total system (formed by the catalyst, the cold object and the hot object) are written without labels, as well as the corresponding unitary operations. This also simplifies notation and does not generate ambiguity, since this is the only three-partite setup considered.

Figure 1(a) illustrates a situation where cooling with a very small hot object is forbidden. Here, the cold object is a qubit in the initial state $\rho_c = \sum_{i=1}^{2} p_i^c |i_c\rangle\langle i_c|$, and the hot object is a three-level system in the state $\rho_h = \sum_{j=1}^{2} p_j^c |j_h\rangle\langle j_h|$. With the prescription of non-increasing eigenvalues, $p_1^c \geq p_2^c$ and $p_j^h \geq p_{j+1}^c$, cooling is
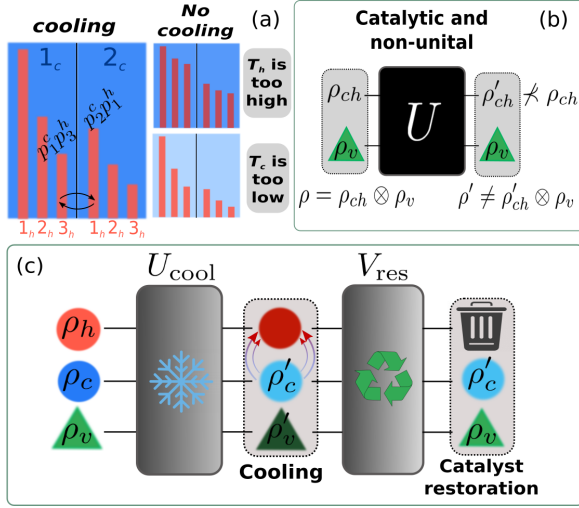
Figure 1. General framework for the studied catalytic transformations. (a) Illustrative example: a three-level system in the initial state ρ_h is used as hot object to cool a (cold) qubit in the initial state ρ_c. The eigenvalues of the joint state $\rho_c \otimes \rho_h$ are obtained by rescaling the eigenvalues of ρ_h (orange bars) with the eigenvalues of ρ_c. When cooling is possible a swap $|1_c 3_h\rangle \rightleftarrows |2_c 1_h\rangle$ (black arrows) suffices to perform it. If T_h is too high or T_c is too low, cooling is impossible with any global unitary U_{ch} or random unitary map $\sum_i \lambda_i U_{ch}^{(i)}$, where $\{\lambda_i\}$ are probabilities [63]. (b) The inclusion of a catalyst in an appropriate initial state ρ_v allows to lift this restriction. Defining $\rho_c \otimes \rho_h \equiv \rho_{ch}$, cooling is enabled only if the corresponding final state ρ'_{ch} is not majorized by ρ_{ch} (non-unital transformation). (c) The transformations are implemented by global unitaries of the form $U = U_{\text{cool}} \oplus V_{\text{res}}$, where U_{cool} is employed to cool the system c and V_{res} returns the catalyst to its initial state. $\rho' \neq \rho'_{ch} \otimes \rho_v$

possible if and only if $p_1^c p_3^h < p_2^c p_1^h$. The case $p_1^c p_3^h \geq p_2^c p_1^h$ constitutes an example of passivity in the context of cooling, characterized for generic systems (with discrete Hamiltonians) in the next section. This restriction can be circumvented by adding a catalyst in a proper initial state ρ_v, such that it allows cooling and also remains unaltered by the transformation. The action of the studied catalytic transformations on the compound hot-cold object is shown in Fig. 1(b). Due to the passivity constraint, the transformation must be non-unital not only on the cold object (to allow cooling), but also on the joint system that includes the hot object (to bypass the passivity restriction). As mentioned in the introduction, this means that the final state cannot be majorized by the initial one. In addition, the generation of correlations between the catalyst and the rest of the total system is allowed. This condition is characteristic of recent works on catalysts [17, 19, 24] and is also natural in our framework, where all the systems involved are finite. A more detailed form of the global unitaries that implement the transformations is illustrated in Fig. 1(c).

III. GENERAL PASSIVITY AND COOLING

The fundamental limits for cooling can be understood using the notion of passivity. Passivity is essentially a condition whereby applying unitary transformations to a system cannot decrease the mean value of certain observables [58, 62]. While traditionally it has been associated with the Hamiltonian and the impossibility of work extraction [60, 82–85], passivity can be extended to any hermitian operator that represents an observable. Consider a bipartite system in the initial state $\rho_c \otimes \rho_h$, where $\rho_c = \frac{e^{-\beta_c H_c}}{\text{Tr}(e^{-\beta_c H_c})}$ and $\rho_h = \frac{e^{-\beta_h H_h}}{\text{Tr}(e^{-\beta_h H_h})}$ are respectively the thermal states of the cold object and the hot object. The corresponding Hamiltonians are $H_c = \sum_{i=1}^{d_c} \varepsilon_i^c |i_c\rangle\langle i_c|$ and $H_h = \sum_{i=1}^{d_h} \varepsilon_i^h |i_h\rangle\langle i_h|$.

For inverse temperatures β_c and β_h such that $\beta_c \geq \beta_h$, cooling occurs if the average value of H_c is reduced. This process requires an interaction with the hot object and possibly an additional external driving, that results in a joint unitary evolution U_{ch}. However, it is possible that

$$\Delta \langle H_c \rangle = \text{Tr}[H_c(U_{ch}\rho_c \otimes \rho_h U_{ch}^\dagger - \rho_c \otimes \rho_h)] \geq 0 \quad (1)$$

for any global unitary U_{ch} (e.g. if the the cold temperature β_c^{-1} is very low). In this case we say that the state $\rho_c \otimes \rho_h$ is passive with respect to the local Hamiltonian H_c. Conversely, if $\Delta \langle H_c \rangle < 0$ for some U_{ch} then $\rho_c \otimes \rho_h$ is non-passive with respect to H_c, and the cold object can be cooled down using a hot object in the state ρ_h. Throughout this text passivity will always refer to initial states that satisfy Eq. (1), unless otherwise stated. Moreover, we note that this kind of passivity is more stringent than the traditional one, since not only ρ_c but also the "extended" state $\rho_c \otimes \rho_h$ is passive with respect to H_c.

A question that follows up naturally is how can we characterize passive states $\rho_c \otimes \rho_h$. Let $\{p_i^c\}_{1 \leq i \leq d_c}$ and $\{p_j^h\}_{1 \leq j \leq d_h}$ denote respectively the eigenvalues of ρ_c and ρ_h, being d_c (d_h) the dimension of the Hilbert space of the cold (hot) object. Using the standard convention of non-decreasing eigenenergies, $\varepsilon_i^c \leq \varepsilon_{i+1}^c$ and $\varepsilon_i^h \leq \varepsilon_{i+1}^h$, passivity is easily expressed by means of the inequalities

$$\frac{p_i^c}{p_{i+1}^c} \geq \frac{p_1^h}{p_{d_h}^h} \, \forall i. \quad (2)$$

From this expression we see that passivity is essentially determined by the ratio between the highest and smallest populations of the hot object. In particular, $p_1^h/p_{d_h}^h = 1$ in the limit of infinite temperature, and the inequalities hold regardless the populations of the cold object. If the hot object is composed of a large of number N of identical subsystems, $p_1^h/p_{d_h}^h = (q_{\max}^h/q_{\min}^h)^N$, where $q_{\max}^h$ and $q_{\min}^h$ denote respectively the highest and smallest populations of each subsystem. Since $q_{\max}^h/q_{\min}^h > 1$ for finite temperature, in the limit $N \to \infty$ the ratio $p_1^h/p_{d_h}^h$ tends to infinity and it is always possible to violate at least one

of the inequalities (2). This explains why cooling is always allowed given unlimited access to a sufficiently large and multipartite hot bath.

In a more general context, two hermitian operators A and B are said to be passive with respect to each other if $[A, B] = 0$ and the eigenvalues of A are non-increasing with respect to those of B [62]. Equation (2) simply translates this condition to the operators $A = \rho_c \otimes \rho_h$ and $B = H_c \otimes \mathbb{I}_h$. According to Eq. (2), the eigenvalues of $\rho_c \otimes \rho_h$ are non-increasing with respect to the index i, while by construction the eigenvalues of H_c are non-decreasing with respect to the same index. Moreover, for i fixed all the eigenstates $|i_c j_h\rangle$ yield the same eigenvalue ε_i^c when H_c is applied on them. This implies that the eigenvalues of $\rho_c \otimes \rho_h$ are non-decreasing with respect to those of $H_c \otimes \mathbb{I}_h$.

IV. CATALYTIC AND NONUNITAL (CNU) TRANSFORMATIONS

A. Catalytic transformations and cooling

Given the passivity condition (2), our goal is to introduce a third system that enables cooling and works as a catalyst. This means that if the catalyst is initially in a state $\rho_v = \sum_{k=1}^{d_v} p_k^v |k_v\rangle\langle k_v|$, at the end of the transformation it must be returned to the same state. In addition, we assume that the catalyst starts uncorrelated from the cold and objects, i.e. the initial total state is $\rho = \rho_c \otimes \rho_h \otimes \rho_v$. The transformation on the cold object is implemented through a global unitary map U that acts on the total system. Denoting the final total state as ρ', a generic catalytic (C) transformation satisfies

$$\rho \xrightarrow{\text{C}} \rho' = U(\rho_c \otimes \rho_h \otimes \rho_v)U^\dagger, \tag{3}$$
$$\mathrm{Tr}_{ch}(\rho') = \rho_v. \tag{4}$$

Note that Eq. (4) guarantees "catalysis" (i.e. the restoration of the catalyst to its initial state) but does not say anything about the final correlations between the catalyst and the rest of the total system. Allowing correlations has proven to be useful in extending the transformations that a system can undergo in the presence of a catalyst [17, 19, 22–24]. Moreover, the access to a broader set of transformations is not the only motivation for using catalysts. Given the condition (4), we can imagine a situation where a fresh copy of the state $\rho_c \otimes \rho_h$ is brought into contact with the catalyst, and the same process performed with the old copy is exactly repeated. This is possible because the initial total state with the new copy is identical to that with the old one, i.e. $\rho_c \otimes \rho_h \otimes \rho_v$. In this way, the catalyst can be harnessed with as many copies as desired, through repeated interactions of the form (3). In Section VI we will see how this possibility can be highly advantageous in a cooling scenario involving many cold objects.

Given a passive state $\rho_c \otimes \rho_h$, the inclusion of the catalyst allows to reduce the mean energy $\langle H_c \rangle$ as long as the total state ρ is non-passive with respect to H_c. Let $\{p_k^v\}_{1 \leq k \leq d_v}$ denote the eigenvalues of ρ_v or "catalyst eigenvalues", for a catalyst of dimension d_v, and let $\{p_i\} = \{p_i^c p_j^h p_k^v\}$ denote the eigenvalues of ρ. Assuming the ordering $p_k^v \geq p_{k+1}^v$ for all k, the catalyst breaks down the initial passivity (2) if and only if there exists i such that

$$\frac{p_i^c}{p_{i+1}^c} < \frac{p_1^h p_1^v}{p_{d_h}^h p_{d_v}^v}. \tag{5}$$

Since the ratio between the highest and smallest eigenvalues of the *composite state* $\rho_h \otimes \rho_v$ (r.h.s. of Eq. (5)) is always larger than $p_1^h/p_{d_h}^h$, by a factor of $p_1^v/p_{d_v}^v$, we can understand why passivity with respect to $\rho_c \otimes \rho_h \otimes \rho_v$ can be violated, even if all the inequalities (2) are satisfied. In particular, this violation always takes place if $\rho_v = |1_v\rangle\langle 1_v|$. However, we will see later that the catalysis condition (4) requires the use of catalysts in initial *mixed* states.

By definition of passivity, if Eq. (5) is satisfied there exists a global unitary that cools down the cold object. We can explicitly consider a two-level unitary U_{swap} that swaps the states $|i_c d_h d_v\rangle$ and $|(i+1)_c 1_h 1_v\rangle$, while acting as the identity on any other eigenstate of ρ. This unitary transfers population $\delta p = p_{i+1}^c p_1^h p_1^v - p_i^c p_{d_h}^h p_{d_v}^v$ from $|(i+1)_c 1_h 1_v\rangle$ to $|i_c d_h d_v\rangle$, thereby reducing the energy of the cold object by $\Delta \langle H_c \rangle = -\delta p(\varepsilon_{i+1}^c - \varepsilon_i^c)$. However, the same operation modifies the state ρ_v by transferring population δp from the eigenstate $|1_v\rangle$ towards the eigenstate $|d_v\rangle$. This illustrates how the possibility of cooling is accompanied by an initial alteration of the catalyst. The restoration of the catalyst involves an additional unitary operation, which in turn potentially spoils the cooling accomplished through U_{swap}.

The characterization of the most general catalytic transformations that also perform cooling is a complex problem that is not addressed in the present work. Instead of that, we shall concentrate on a subset of this class of transformations, determined by sufficient conditions that will be specified later on. Meanwhile, motivated by our previous discussion, we present the general structure of the global unitaries U we will be interested in. These unitaries are composed of two independent unitaries U_{cool} and V_{res}, whose functions are respectively cooling and restoration of the catalyst (cf. Fig. 1). Hence, we will refer to them as the "cooling unitary" and the "restoring unitary". An example of cooling unitary is the swap U_{swap} described before. The restoring unitary is chosen in such a way that the subspaces where U_{cool} and V_{res} are defined are orthogonal. Let $\mathcal{H}_{\text{cool}} \subset \mathcal{H}$ and $\mathcal{H}_{\text{res}} \subset \mathcal{H}$ denote such subspaces, being $\mathcal{H} = \mathrm{span}\{|i_c j_h k_v\rangle\}$ the total Hilbert space. We consider global unitaries that satisfy

Eq. (4) and have the form

$$U = U_{\text{cool}} \oplus V_{\text{res}}, \tag{6}$$

$$U_{\text{cool}} : \mathcal{H}_{\text{cool}} \to \mathcal{H}_{\text{cool}}, \tag{7}$$

$$V_{\text{res}} : \mathcal{H}_{\text{res}} \to \mathcal{H}_{\text{res}}, \tag{8}$$

where $\oplus$ stands for the direct sum.

For the sake of clarity we will always describe the action of U as a sequence $V_{\text{res}}U_{\text{cool}}$, where V_{res} is preceded by U_{cool}. However, due to the direct sum structure (6) we have that $V_{\text{res}}U_{\text{cool}} = U_{\text{cool}}V_{\text{res}}$. Crucially, the commutativity between U_{cool} and V_{res} also allows us to study the effect of V_{res} using directly the initial state ρ, instead of the state obtained after the application of U_{cool}.

B. Majorization, cooling currents and restoring currents

1. Majorization, cooling, and non-unitality

Majorization defines a preorder between quantum states [80, 81], and is intimately connected with different cooling criteria [45]. Let ϱ and σ denote two generic quantum states defined on some Hilbert space of dimension d, with respective eigenvalues $\{r_i\}_{1 \le i \le d}$ and $\{q_i\}_{1 \le i \le d}$. Moreover, let $\{r_i^\downarrow\}$ and $\{q_i^\downarrow\}$ be the same eigenvalues arranged in non-increasing order, i.e. $r_{i+1}^\downarrow \le r_i^\downarrow$ and $q_{i+1}^\downarrow \le q_i^\downarrow$. It is said that "$\varrho$ majorizes σ", formally written as $\varrho \succ \sigma$, if

$$\sum_{i=1}^{j} r_i^\downarrow \ge \sum_{i=1}^{j} q_i^\downarrow \tag{9}$$

for all $1 \le j \le d$. Physically, majorization is useful to compare the degree of purity between two quantum states. For example, according to Eq. (9) a pure state majorizes any other state, while a fully mixed state is majorized by any other state. However, it is possible that for some pair of states ϱ and σ none of the conditions $\varrho \succ \sigma$ or $\sigma \succ \varrho$ takes place, whereby majorization does not constitute an order relation.

An important result of information theory [81, 86] states that $\varrho \succ \sigma$ iff $\sigma = \sum_i \lambda_i U_i \varrho U_i^\dagger$, where $\{\lambda_i\}$ are probabilities and $\{U_i\}$ are unitary operations. Equivalently, $\varrho \succ \sigma$ iff $\sigma = \mathcal{E}_{\text{un}}(\varrho)$, where $\mathcal{E}_{\text{un}}$ is a unital map defined by the condition $\mathcal{E}_{\text{un}}(\mathbb{I}) = \mathbb{I}$ [86]. Based on this observation, we define a "unital transformation" as a transformation $\varrho \to \sigma$ such that $\varrho \succ \sigma$, since it can be implemented through the application of a unital map to ϱ.

The description of majorization in terms of the relation $\sigma = \sum_i \lambda_i U_i \varrho U_i^\dagger$ establishes a link between majorization and the standard definition of cooling. Consider a transformation on the cold object $\rho_c \to \rho_c'$, such that $\rho_c \succ \rho_c'$. Since this implies that ρ_c' can be written as $\rho_c' = \sum_i \lambda_i U_i \rho_c U_i^\dagger$, this transformation can only increase

the value of $\langle H_c \rangle$ because unitary operations cannot lower the mean energy of a thermal state. Therefore, a necessary condition to have $\Delta \langle H_c \rangle < 0$ is that ρ_c does not majorize ρ_c', or equivalently that $\rho_c \to \rho_c'$ is a "non-unital transformation". This occurs in particular if the populations of ρ_c' in the eigenbasis of ρ_c satisfy

$$\sum_{l=1}^{i} p_l^c < \sum_{l=1}^{i} p_l'^c \text{ for some } 1 \le i \le d_c - 1, \tag{10}$$

where $p_l'^c = \text{Tr}(|l_c\rangle\langle l_c|\rho'_c)$ and $\rho_c|l_c\rangle = p_l^c|l_c\rangle$.

Let us see why a transformation satisfying Eq. (10) cannot be achieved through a unital map. First, the eigenvalues $\{p_l^c\}$ are by construction non-increasing and therefore $\sum_{l=1}^{i} p_l^c = \sum_{l=1}^{i} p_l^{c\downarrow}$. In addition, the populations $\{p_l'^c\}$ correspond to the eigenvalues of the state $\mathcal{D}(\rho_c')$, where $\mathcal{D}$ is the map that completely erases the coherence of ρ_c' in the eigenbais of ρ_c. Since this operation can be performed through a random unitary map [11], it follows that $\rho_c' \succ \mathcal{D}(\rho_c')$. Hence, $\sum_{l=1}^{i} p_l'^{c\downarrow} \le \sum_{l=1}^{i} q_l^{c\downarrow}$ for all i, where $\{q_l^{c\downarrow}\}$ are the sorted eigenvalues of ρ_c'. By plugging this inequality in the r.h.s. of Eq. (10), we obtain the relation $\sum_{l=1}^{i} p_l^{c\downarrow} < \sum_{l=1}^{i} q_l^{c\downarrow}$, which means that Eq. (9) is violated for some i.

The catalytic and cooling transformations that we will study are based on a more general class of catalytic transformations that satisfy Eq. (10). We denote a transformation of this kind as $\rho \xrightarrow{\text{CNU}} \rho'$, with the abbreviation CNU standing for catalytic (with respect to the catalyst) and non-unital (with respect to the cold object). A characterization of these transformations shall be provided in Theorem 1. $\{\ln(p_1^c p_j^h)\}_j$

2. Cooling currents and restoring currents

The question we want to answer can be precisely stated in the following way: given an initial state of the form $\rho = \rho_c \otimes \rho_h \otimes \rho_v$, such that $\rho_c \otimes \rho_h$ is passive with respect to H_c, is there a unitary U that satisfies Eq. (6) and that allows to implement a CNU transformation? We will provide sufficient conditions for a positive answer, expressed entirely in terms of the eigenvalues of ρ. To that end we will explicitly construct unitaries U that perform the transformation, which are conveniently characterized using the notion of "population currents", or simply "currents".

A population current is the population transferred between two eigenstates $|\boldsymbol{i}\rangle$ and $|\boldsymbol{j}\rangle$ of ρ, due to the action of a two-level unitary $U^{(2)} : \text{span}\{|\boldsymbol{i}\rangle, |\boldsymbol{j}\rangle\} \to \text{span}\{|\boldsymbol{i}\rangle, |\boldsymbol{j}\rangle\}$. Any population transferred in this way can be described through a unitary of the form

$$U^{(2)}|\boldsymbol{i}\rangle = \sqrt{1-a^2}|\boldsymbol{i}\rangle + a|\boldsymbol{j}\rangle, \tag{11}$$

$$U^{(2)}\boldsymbol{j}\rangle = a|\boldsymbol{i}\rangle - \sqrt{1-a^2}|\boldsymbol{j}\rangle, \tag{12}$$

where $0 \leq a \leq 1$. Importantly, our definition of current refers to a *net population transfer* between two states, rather than a rate of population exchanged per unit of time. However, we will see that such a denomination is helpful to have an intuitive picture for the transformations that will be studied throughout this paper.

Keeping in mind that the initial populations are $p_i = \mathrm{Tr}(|i\rangle\langle i|\rho)$ and $p_j = \mathrm{Tr}(|j\rangle\langle j|\rho)$, after the application of $U^{(2)}$ the state $|j\rangle$ acquires population

$$p'_j = p_i a^2 + p_j(1 - a^2)$$
$$= p_j + a^2(p_i - p_j). \tag{13}$$

In this way, the population current from $|i\rangle$ to $|j\rangle$ is defined as

$$J_{|i\rangle \to |j\rangle} \equiv \begin{cases} a^2(p_i - p_j), & \text{if } p_i > p_j \\ 0, & \text{if } p_i \leq p_j. \end{cases} \tag{14}$$

The definition (14) may at first look a bit artificial, given that it only describes the transferred population $p'_j - p_j$ if the population of $|j\rangle$ increases (cf. Eq. (13)). However, this convention of positive currents has the advantage that a current $J_{|i\rangle \to |j\rangle}$ unambiguously indicates a population flow from $|i\rangle$ to $|j\rangle$, and consequently that $p_i > p_j$. If $p_i < p_j$, the flow occurs in the opposite direction and is characterized by the current $J_{|j\rangle \to |i\rangle} = a^2(p_j - p_i)$. Clearly, $J_{|i\rangle \to |j\rangle}$ varies between 0 and $p_i - p_j$, with its maximum value attained when $U^{(2)}$ performs a swap between the states $|i\rangle$ and $|j\rangle$. This maximum is termed "swap current" and is denoted as

$$\mathcal{J}_{|i\rangle \to |j\rangle} \equiv \max_a J_{|i\rangle \to |j\rangle} = p_i - p_j. \tag{15}$$

Eventually, we will also employ the notation $J_{|i\rangle \to}$ for a current that describes a population flow from $|i\rangle$ to some unknown eigenstate of ρ. Likewise, $J_{\to |i\rangle}$ will denote a population flow from an unknown eigenstate towards $|i\rangle$.

Depending on the states $|i\rangle$ and $|j\rangle$ and their initial populations, the two-level unitaries described by Eqs. (11) and (12) generate different types of currents. In particular, the violation of majorization (10) (necessary for cooling) is possible if there exist a current that we term "cooling current". In the following the "existence of a current $J_{|i\rangle \to |j\rangle}$" signifies that $J_{|i\rangle \to |j\rangle} \neq 0$, which in turn implies that there exists a two-level unitary transferring population from $|i\rangle$ to $|j\rangle$. Moreover, we note that $J_{|i\rangle \to |j\rangle}$ exists iff $\mathcal{J}_{|i\rangle \to |j\rangle}$ exists.

Definition 1 (cooling current). A cooling current is a current $J_{|(i+n)_c j_h k_v\rangle \to |i_c j'_h k'_v\rangle}$ ($n \geq 1$), whose function is to transfer population from an eigenstate of $\rho_c \otimes \mathbb{I}_{hv}$ with eigenvalue p^c_{i+n}, towards an eigenstate with larger or equal eigenvalue p^c_i. From Eq. (15) it is straightforward to check that $J_{|(i+n)_c j_h k_v\rangle \to |i_c j'_h k'_v\rangle}$ exists iff

$$\frac{p^v_k}{p^v_{k'}} > \left(\frac{p^h_{j'}}{p^h_j}\right) \frac{p^c_i}{p^c_{i+n}}$$
$$\Leftrightarrow \frac{p^v_1}{p^v_{d_v}} > \left(\frac{p^h_{d_h}}{p^h_1}\right) \frac{p^c_i}{p^c_{i+1}}, \tag{16}$$

where the second inequality follows by bounding $\frac{p^v_k}{p^v_{k'}}$ from above and $\left(\frac{p^h_{j'}}{p^h_j}\right) \frac{p^c_i}{p^c_{i+n}}$ from below.

Noting that Eqs. (16) and (5) are equivalent, we conclude that the inclusion of a catalyst breaks down the passivity with respect to H_c iff the eigenvalues of ρ_v are such that there exists a cooling current $J_{|(i+n)_c j_h k_v\rangle \to |i_c j'_h k'_v\rangle}$. Such a current has opposite effects on the cold object and the catalyst. First, it increases the partial sum $\sum_{l=1}^i p^c_l$ by transferring population to $|i_c\rangle$, which yields a nonunital transformation of the form (10). On the other hand, notice that the r.h.s. of Eq. (16) must be larger or equal than one due to passivity without the catalyst. As a consequence $p^v_k > p^v_{k'}$, whereby the cooling current also reduces the population of the catalyst eigenstate $|k_v\rangle$, and increases the (smaller) population of $|k'_v\rangle$ by the same amount. This has a mixing effect on such states, which can be readily reproduced through a *local* two-level unitary acting on the subspace $\mathrm{span}\{|k_v\rangle, |k'_v\rangle\}$. By definition, the resulting transformation is unital. The purpose of restoring currents is to counter this effect by transferring population in the opposite sense. That is, from a low-population eigenstate of ρ_v towards a higher-population eigenstate.

Definition 2 (restoring current). A general restoring current is a current $J_{|i_c j_h (k+n)_v\rangle \to |i'_c j'_h k_v\rangle}$ ($n \geq 1$), whose function is to transfer population from an eigenstate of $\mathbb{I}_{ch} \otimes \rho_v$ with eigenvalue p^v_{k+n}, towards an eigenstate with larger or equal eigenvalue p^v_k. However, the study of cooling transformations will be mainly based on restoring currents of the kind $J_{|j_h (k+n)_v\rangle \to |j'_h k_v\rangle}$, which are associated with two-level unitaries that do not involve the cold object. From Eq. (15), it is straightforward to check that $J_{|j_h (k+n)_v\rangle \to |j'_h k_v\rangle}$ exists iff

$$\frac{p^h_j}{p^h_{j'}} > \frac{p^v_k}{p^v_{k+n}}$$
$$\Leftrightarrow \frac{p^h_1}{p^h_{d_h}} > \frac{p^v_k}{p^v_{k+1}}, \tag{17}$$

where the second inequality follows by bounding $\frac{p^v_k}{p^v_{k+n}}$ from below and $\frac{p^h_j}{p^h_{j'}}$ from above.

C. CNU transformations with a single cooling current

Now we specialize to CNU transformations that contain a single cooling current, which amounts to impose that $U_{\rm cool} = U_{\rm cool}^{(2)}$ is given by a single two-level unitary. In Sect. VIII we will discuss how to construct more general transformations, where $U_{\rm cool}$ may be composed of several two-level unitaries. If $U_{\rm cool} = U_{\rm cool}^{(2)}$, we denote the corresponding transformation as $\rho \xrightarrow{\rm CNU1} \rho'$. Moreover, we consider restoring unitaries $V_{\rm res}$ of the form

$$V_{\rm res} = \oplus_k V_k^{(2)}, \tag{18}$$

where each $V_k^{(2)}$ is a two-level unitary giving rise to a restoring current.

Given a cooling current $J_{|(i+n)_c j_h k_v\rangle \to |i_c j_h' k_v'\rangle}$, the goal of $V_{\rm res}$ is to return the populations of the states $|k_v\rangle$ and $|k_v'\rangle$ to their initial values. The most direct way to do that would be through a single restoring current $J_{|l_c' m_h' k_v'\rangle \to |l_c m_h k_v\rangle}$. However, in general such a current may not exist. The following theorem provides necessary and sufficient conditions for the existence of a CNU1 transformation, in terms of appropriate restoring currents. The corresponding proof can be found in Appendix A. In Sect. VC1, we introduce a graphical method that provides an intuitive visualization of the Theorem. In Sects. VC2 and VC3 we establish the relation between restoring currents and general restoring unitaries (18). Finally, in Sect. VC4, we apply this relation to characterize the restoring unitaries that can be used according to Theorem 1. These unitaries are special in the sense that they not only restore the catalyst, but also guarantee the non-unital character of the transformation.

Theorem 1 (existence of CNU1 transformations). Let $\rho = \rho_c \otimes \rho_h \otimes \rho_v \in B(\mathcal{H}_c \otimes \mathcal{H}_h \otimes \mathcal{H}_v)$ be an initial density matrix of the total system, with $\mathcal{H}_{x=c,h,v}$ a Hilbert space of dimension d_x. Moreover, let $\{p_i^x\}$ denote the eigenvalues of ρ_x. A CNU1 transformation $\rho \xrightarrow{\rm CNU1} \rho' = U_{\rm cool}^{(2)} \oplus V_{\rm res} \rho U_{\rm cool}^{(2)\dagger} \oplus V_{\rm res}^\dagger$ exists iff

1. There exists a cooling current $\mathcal{J}_{|(i+1)_c 1_h l_v\rangle \to |i_c d_h (l'+1)_v\rangle}$, determined by the conditions

$$\frac{p_l^v}{p_{l'+1}^v} > \left(\frac{p_{d_h}^h}{p_1^h}\right) \frac{p_i^c}{p_{i+1}^c} > 1, \tag{19}$$

for some $1 \le i \le d_c - 1$ and $1 \le l, l' \le d_v - 1$, and

2. For $l \le k \le l'$ there exist restoring currents $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |i_c d_h k_v\rangle}\}_{l \le k \le l'}$, determined by the conditions

$$\left(\frac{p_1^h}{p_{d_h}^h}\right) \frac{p_1^c}{p_i^c} > \frac{p_k^v}{p_{k+1}^v} > 1, \tag{20}$$

or restoring currents $\{\mathcal{J}_{|(i+1)_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l \le k \le l'}$,

determined by the conditions

$$\left(\frac{p_1^h}{p_{d_h}^h}\right) \frac{p_{i+1}^c}{p_{d_c}^c} > \frac{p_k^v}{p_{k+1}^v} > 1. \tag{21}$$

Remark 1 (Generalization of Theorem 1). The proof given in Appendix A is applicable to general states of the form $\rho_s \otimes \rho_v$, where ρ_s represents an arbitrary state on which the non-unital transformation is performed. From this general proof, the conditions (19)-(21) follow by choosing $\rho_s = \rho_c \otimes \rho_h$, and requiring that the transformation is non-unital not only on ρ_s, but also on the state ρ_c.

On the other hand, we also note that in Theorem 1 no reference is made to the thermal character of the states ρ_c and ρ_h. Similarly, the theorems 2 and 3 that will be presented later are formulated only in terms of the *eigenvalues* of general states ρ_c and ρ_h.

1. Graphical characterization of CNU1 transformations

In order to provide an intuitive understanding of Theorem 1 we introduce the following graphical method to describe cooling currents and catalytic currents:

$\mathbf{ln}(p^{ch}) \times \mathbf{ln}(p^v)$ **Diagram**. Consider an horizontal axis where the values $\{\ln(p_i^c p_j^h)\}_{1 \le i \le d_c, 1 \le j \le d_h}$ are arranged in decreasing order, i.e. the larger the value the more to the left it is placed on this axis. Similarly, we arrange the values $\{\ln(p_k^v)\}_{1 \le k \le d_v}$ in a vertical axis, with larger values at the top and smaller ones at the bottom. A "row" k_v is an horizontal line that intersects the value $\ln(p_k^v)$, and represents also the catalyst eigenstate $|k_v\rangle$. A "column" $i_c j_h$ is a vertical line that passes through the value $\ln(p_i^c p_j^h)$, and represents the eigenstate $|i_c j_h\rangle$. The intersection between a row k_v and a column $i_c j_h$ is associated with the pair $(i_c j_h, k_v)$, which corresponds to the global eigenstate $|i_c j_h k_v\rangle$.

The main purpose of the $\ln(p^{ch}) \times \ln(p^v)$ diagram, illustrated in Fig. 2, is the depiction of cooling currents and catalytic currents. In addition, the non-overlap between gray rectangles (energy eigenspaces of the cold object) stands for the condition of passivity without catalyst. This means that the largest element of a any subset in the low gray boxes is always upper bounded by the smallest element in the next subset at the left. By applying the natural logarithm to the second inequality in Eq. (16), we obtain the relation $\ln(p_1^v) - \ln(p_{d_v}^v) > \ln(p_i^c p_{d_h}^h) - \ln(p_{i+1}^c p_1^h)$. Therefore, the existence of a cooling current (downward-oriented blue arrow) means that the height of the $\ln(p^{ch}) \times \ln(p^v)$ diagram must be larger than the distance between two consecutive columns $i_c d_h$ and $(i + 1)_c 1_h$, for some value of i. In the diagram this relation is represented by enclosing the cooling current inside a vertical rectangle. Analogously, the application of the natural logarithm to Eq. (17) yields the inequality $\ln(p_1^h) - \ln(p_{d_h}^h) > \ln(p_k^v) - \ln(p_{k+1}^v)$, meaning that the
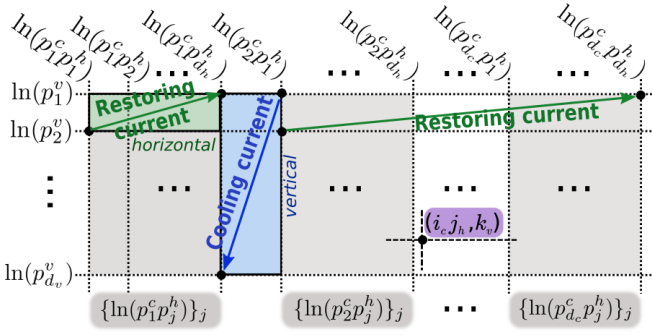
Figure 2. $\ln(p^{ch}) \times \ln(p^v)$ diagram. In this diagram each element $\ln(p_i^c p_j^h) \in \{\ln(p_i^c p_j^h)\}_{i,j}$ is associated with a "column" $i_c j_h$ (vertical line), and each element $\ln(p_k^v) \in \{\ln(p_k^v)\}_k$ is associated with a "row" k_v (horizontal line). The corresponding interesection yields the pair $(i_c j_c, k_v)$ (purple box). The elements $\ln(p_i^c p_j^h)$ are arranged in non-increasing order, from left to right, and $\ln(p_k^v)$ are non-increasing, from top to bottom. The left-most green arrow represents a current that takes place inside an energy eigenspace of the cold object, and is the primary kind of restoring current involved in cooling transformations. The right-most green arrow connects different eigenspaces of ρ_c, and is involved in more general (not necessarily cooling) non-unital transformations on the composite state $\rho_c \otimes \rho_h$.

corresponding restoring current is enclosed by an horizontal rectangle of width $\ln(p_1^h/p_{d_h}^h)$ and height $\ln(p_k^v/p_{k+1}^v)$. The left-most green arrow in Fig. 2 illustrates this type of restoring current for $k = 1$. Moreover, generic restoring currents are always upward-oriented.

The diagrams for the conditions of Theorem 1 are given in Fig. 3. The sides of the vertical cyan rectangle in both diagrams have lenghts obtained from the application of the natural logarithm to Eq. (19). Specifically, its height is given by $\ln(p_l^v/p_{l'+1}^v)$, and its width is given by $\ln(p_i^c p_{d_h}^h/p_{i+1}^c p_1^h)$. Hence, a cooling current exists iff we can identify a vertical rectangle with vertical sides living on *consecutive* columns $i_c d_h$ and $(i+1)_c 1_h$. The restoring currents are enclosed by a set of adjacent horizontal rectangles (light green rectangles). When taken together, they compose a (not necessarily horizontal) total rectangle joining the rows l_v and $(l'+1)_v$. The rectangles shown in Fig. 2(a) result from taking the logarithm of Eq. (20), and are located at the left of the cyan rectangle. Moreover, the rectangles in Fig. 2(b) are located at the right of the cyan rectangle and result from the logarithm of Eq. (21).

2. Effect of currents on the the catalyst

The total population variation for a catalyst eigenstate $|k_v\rangle$ is given by (see Appendix B)

$$p_k'^v - p_k^v = \text{Tr}[|k_v\rangle\langle k_v|(U\rho U^\dagger - \rho)]$$
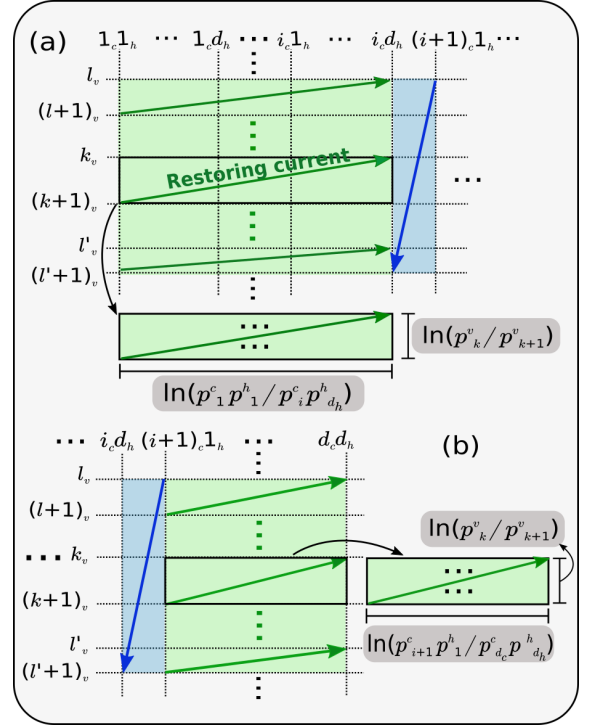$$= J_{\to|k_v\rangle} - J_{|k_v\rangle\to}, \qquad (22)$$

Figure 3. $\ln(p^{ch}) \times \ln(p^v)$ diagram for the conditions stated in Theorem 1. Vertical (horizontal) ellipsis indicate that there can be rows (columns) in between. (a) Depiction of the conditions described by Eqs. (19) and (20). (b) Depiction of the conditions described by Eqs. (19) and (21).

where

$$J_{\to|k_v\rangle} \equiv \sum_{i,j} J_{\to|i_c j_h k_v\rangle} \qquad (23)$$

is the sum of all the currents that transfer population to eigenstates with catalyst eigenvalue p_k^v. Similarly,

$$J_{|k_v\rangle\to} \equiv \sum_{i,j} J_{|i_c j_h k_v\rangle\to} \qquad (24)$$

corresponds to the sum of all the currents that take population from these eigenstates.

It is important to stress that, in the most general case, a current $J_{\to|i_c j_h k_v\rangle}$ could connect $|i_c j_h k_v\rangle$ with another eigenstate that also has catalyst eigenvalue p_k^v. If this occurs such a current cannot contribute to $J_{\to|k_v\rangle}$, since it leaves invariant the population of $|k_v\rangle$. However, we always deal with currents $J_{\to|i_c j_h k_v\rangle}$ and $J_{|i_c j_h k_v\rangle\to}$ connecting eigenstates with different catalyst eigenvalues, which implies that the quantities $J_{\to|k_v\rangle}$ and $J_{|k_v\rangle\to}$ are properly characterized by Eqs. (23) and (24). In the following $J_{\to|k_v\rangle}$ and $J_{|k_v\rangle\to}$ will be termed "catalyst currents", as they describe population flows within the catalyst.

3. Restoration of the catalyst

Equation (22) implies that the population of the state $|k_v\rangle$ remains unchanged as long as the corresponding catalyst currents satisfy $J_{\to|k_v\rangle} = J_{|k_v\rangle\to}$. In the following lemma we provide necessary and sufficient conditions for the existence of a restoring unitary $V_{\rm res}$, given a two-level unitary $U_{\rm cool}^{(2)}$ that generates a single cooling current. Accordingly, the condition $J_{\to|k_v\rangle} = J_{|k_v\rangle\to}$ holds under the action of the total unitary $U = U_{\rm cool}^{(2)} \oplus V_{\rm res}$. Here we will prove sufficiency, leaving the proof of necessity for Appendix C.

Lemma 1 (Existence of restoring unitaries). Let $J_{|(i+1)_c 1_h l_v\rangle \to |i_c d_h (l'+1)_v\rangle}$ be a cooling current, which increasses (decreases) the population of the catalyst eigenstate $|(l'+1)_v\rangle$ ($|l_v\rangle$). A restoring unitary $V_{\rm res}$ that reverses the effect of this current on the catalyst exists iff there exists a set of restoring currents $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l\le k\le l'}$ (see Fig. 4).

The following definitions will be useful to prove this lemma:

Definition 3 (loop and uniform loop). A loop is a set of currents, such that for any catalyst state with an incoming current $J_{\to|k_v\rangle}$ there is an outgoing current $J_{|k_v\rangle\to}$. A "uniform loop" is a loop with the additional property that all the catalyst currents satisfy $J_{\to|k_v\rangle} = J_{|k_v\rangle\to}$. According to Eq. (22), a uniform loop keeps the state of the catalyst unchanged.

Definition 4 (chain). A chain is a set of currents, such that only two of the connected eigenstates are not connected by both types of currents (incoming and outgoing). Let us call such states the "outer links of the chain", while the other eigenstates (connected by an incoming and an outgoing current) will be called "inner links". To understand the role of chains in the existence of restoring unitaries, and at the same time keep consistency with previously introduced notation, it is convenient to denote the outer links as $|l_v\rangle$ and $|(l'+1)_v\rangle$. In this way, a chain is a set

$$\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle} \equiv \{J_{|l_v\rangle \to |k_v^{(1)}\rangle}, J_{|k_v^{(1)}\rangle\to}, ... $$
$$..., J_{\to |k_v^{(n)}\rangle}, J_{|k_v^{(n)}\rangle \to |(l'+1)_v\rangle}\}, \quad (25)$$

where $J_{|l_v\rangle \to |k_v^{(1)}\rangle}$ ($J_{|k_v^{(n)}\rangle \to |(l'+1)_v\rangle}$) is the total catalyst current connecting the states $|l_v\rangle$ and $|k_v^{(1)}\rangle$ ($|k_v^{(n)}\rangle$ and $|(l'+1)_v\rangle$).

Since $J_{|l_v\rangle \to |k_v^{(1)}\rangle}$ constitutes an incoming current for $|k_v^{(1)}\rangle$, Eq. (25) indicates that the inner link $|k_v^{(1)}\rangle$ contains both an incoming current and an outgoing current $J_{|k_v^{(1)}\rangle\to}$. Similarly, $J_{|k_v^{(n)}\rangle \to |(l'+1)_v\rangle}$ constitutes the outgoing current for the inner link $|k_v^{(n)}\rangle$. On the other hand, the outer link $|l_v\rangle$ only has an outgoing current, and the outer link $|(l'+1)_v\rangle$ only has an incomming current. If $\{|k_v^{(2)}\rangle, |k_v^{(3)}\rangle, ..., |k_v^{(n-1)}\rangle\}$ denotes the set of all the remaining inner links, then the ellipsis in Eq. (25) stand

for pairs $\{J_{|k_v^{(i)}\rangle\to}, J_{\to |k_v^{(i)}\rangle}\}_{2\le i\le n-1}$ of incoming and outgoing currents connecting these links. We also note that the simplest chain has the form $\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle} = \{J_{|l_v\rangle \to |(l'+1)_v\rangle}\}$, with a single catalyst current and no inner links.

Definition 5 (restoring chain). When a chain $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$ is joined with a chain $\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$ their union forms a loop $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle} \cup \mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$, since $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$ provides an incomming current for the outer link $|l_v\rangle$, and an outgoing current for the outer link $|(l'+1)_v\rangle$. In this way, $|l_v\rangle$ and $|(l'+1)_v\rangle$ become inner links in the set $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle} \cup \mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$ (note that by definition all the links are inner links in a loop). If the currents contained by $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$ and $\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$ have all the same magnitude, their union yields also a uniform loop. Thus, we can say that $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$ is a "restoring chain" for $\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$ and vice versa, since the populations changes in the outer links are cancelled out once they are joined.

Sufficiency proof for Lemma 1. If $\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$ is the chain that contains only the cooling current $\mathcal{J}_{|(i+1)_c 1_h l_v\rangle \to |i_c d_h (l'+1)_v\rangle}$, the set $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l\le k\le l'}$ is a chain of the type $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$, which generates a loop with $\mathbf{ch}_{|l_v\rangle \to |(l'+1)_v\rangle}$ (see Fig. 4). Let $\mathcal{J}_{\rm loop}^{\rm min}$ denote the minimum swap current in this loop . Moreover, let

$$\tilde{V}_{\rm res} = \oplus_{k=l}^{l'} \tilde{V}^{(2)}_{|1_c 1_h (k+1)_v\rangle \leftrightarrow |d_c d_h k_v\rangle} \quad (26)$$

be a unitary composed of partial swaps $\tilde{V}^{(2)}_{|1_c 1_h (k+1)_v\rangle \leftrightarrow |d_c d_h k_v\rangle}$ between the states $|1_c 1_h (k+1)_v\rangle$ and $|d_c d_h k_v\rangle$, with "intensities" a_k^2 (cf. Eqs. (11) and (12)). Since $\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle} > 0$, each partial swap $\tilde{V}^{(2)}_{|1_c 1_h (k+1)_v\rangle \leftrightarrow |d_c d_h k_v\rangle}$ generates a restoring current $J_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle} > 0$. If the swap intensity of the cooling unitary, denoted by $a_{\rm cool}^2$, and the intensities a_k^2, satisfy

$$a_{\rm cool}^2 = \frac{\mathcal{J}_{\rm loop}^{\rm min}}{\mathcal{J}_{|(i+1)_c 1_h l_v\rangle \to |i_c d_h (l'+1)_v\rangle}}, \quad (27)$$

$$a_{l\le k\le l'}^2 = \frac{\mathcal{J}_{\rm loop}^{\rm min}}{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}}, \quad (28)$$

Eq. (14) implies that all the currents generated by $\tilde{V}_{\rm res}$ and $U_{\rm cool}^{(2)}$ have the same magnitude $\mathcal{J}_{\rm loop}^{\rm min}$. Therefore, a unitary $U = U_{\rm cool}^{(2)} \oplus \tilde{V}_{\rm res}$ that satisfies the previous equations is catalytic (or equivalently $\tilde{V}_{\rm res}$ is restoring), since it generates a uniform loop.

The necessity condition for Lemma 1 is proven in Appendix C. The essential idea is that if the chain $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l\le k\le l'}$ does not exist, no general restoring chain $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$ exists either. Accordingly, it is impossible to form a loop with the cooling current and the catalyst cannot be restored.
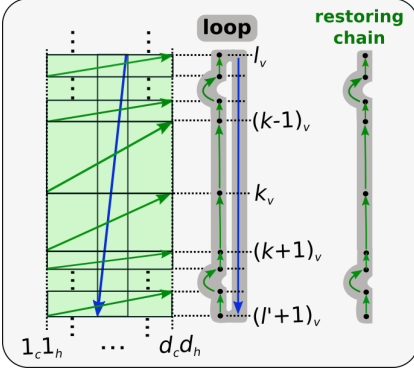
Figure 4. $\ln(p^{ch}) \times \ln(p^v)$ diagram for the existence of a restoring unitary. Given a cooling current that transfers population from $|l_v\rangle$ to $|(l'+1)_v\rangle$ (blue arrow), the set of currents $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l \leq k \leq l'}$ (green arrows) forms a restoring chain $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$ that restores the populations p_l^v and $p_{l'+1}^v$, while keeping the intermediate populations $p_{l+1 \leq k \leq l'}^v$ unchanged. The "vertical projection" of the currents at the right characterizes the population flows within the catalyst. In this depiction, a curved arrow represents a portion of the chain that potentially connects several intermediate (not shown) eigenstates. The top and bottom currents cancel the population changes due to the cooling current. Moreover, any intermediate eigenstate $|k_v\rangle$ ($l+1 \leq k \leq l'$) has incomming and outgoing currents that cancel each other.

4. Relation between restoring chains and restoring unitaries

The connection between restoring unitaries and restoring chains, previously elucidated, allows us to study the structure of restoring unitaries that yield CNU1 and cooling transformations. First, note that the sets of restoring currents characterized by Eqs. (20) and (21) are also restoring chains of the form $\mathbf{ch}_{|(l'+1)_v\rangle \to |l_v\rangle}$, each of which forms a loop with the cooling current $\mathcal{J}_{|(i+1)_c 1_h l_v\rangle \to |i_c d_h (l'+1)_v\rangle}$. Accordingly, they also have the loop structure shown in Fig. 4. The existence of these chains ensures that there exist unitaries

$$V_{\mathrm{res},L} = \oplus_{k=l}^{l'} V^{(2)}_{|1_c 1_h (k+1)_v\rangle \leftrightarrow |i_c d_h k_v\rangle}, \quad (29)$$

$$V_{\mathrm{res},R} = \oplus_{k=l}^{l'} V^{(2)}_{|(i+1)_c 1_h (k+1)_v\rangle \leftrightarrow |d_c d_h k_v\rangle}, \quad (30)$$

where $V_{\mathrm{res},L}$ is a "left" restoring unitary, derived from Eq. (20), and $V_{\mathrm{res},R}$ is a "right" restoring unitary, derived from Eq. (21). Essentially, the partial swaps in $V_{\mathrm{res},L}$ generate the restoring currents illustrated in Fig. 3(a), and the partial swaps in $V_{\mathrm{res},R}$ generate the restoring currents shown in Fig. 3(b). By suitably adjusting the intensities of these swaps, it is possible to obtain a uniform loop that guarantees the restoration of the catalyst. In addition, we stress that either $V_{\mathrm{res},L}$ or $V_{\mathrm{res},R}$ exist only if the chain described in Lemma 1 exists.

A restoring chain of the form $\{\mathcal{J}_{|1_h (k+1)_v\rangle \to |d_h k_v\rangle}\}_{l \leq k \leq l'}$ also allows to reverse the effect that a cooling current $\mathcal{J}_{|(i+1)_c 1_h l_v\rangle \to |i_c d_h (l'+1)_v\rangle}$

has on the catalyst. The currents in this chain exist iff the inequalities $\frac{p_1^h}{p_{d_h}^h} > \frac{p_k^v}{p_{k+1}^v}$ hold for $l \leq k \leq l'$ (Eq. (17)). Crucially, these inequalities guarantee a unitary $V_{\mathrm{res}} = \mathbb{I}_c \otimes V_{hv}$, which restores the catalyst using only the hot object. In particular, a direct sum of appropriate partial swaps

$$V_{hv} = \oplus_{k=l}^{l'} V^{(2)}_{|1_h (k+1)_v\rangle \leftrightarrow |d_h k_v\rangle} \quad (31)$$

generates a uniform restoring chain $\{J_{|1_h (k+1)_v\rangle \to |d_h k_v\rangle}\}_{l \leq k \leq l'}$, where all the currents have the same magnitude of the cooling current. Since $\mathbb{I}_c \otimes V_{hv}$ does not interfere with the cooling effect of $U^{(2)}_{\mathrm{cool}}$, in the rest of the paper we will mainly deal with transformations based on unitaries $U = U^{(2)}_{\mathrm{cool}} \oplus (\mathbb{I}_c \otimes V_{hv})$.

V. CATALYTIC COOLING (CC) TRANSFORMATIONS

In the preceding section we established necessary and sufficient conditions for a CNU1 transformation. Here we show some examples where Theorem 1 can be applied to characterize catalytic cooling (CC) transformations, i.e. transformations that are catalytic and also obey some standard criterion for cooling. To this aim we start by presenting our second main result, which addresses the existence of CNU1 transformations in terms of the dimension of the catalyst.

Theorem 2 (Catalyst size and CNU1 transformations). Let d_v denote the dimension of a finite catalyst, and let $\{p_l^c\}$ and $\{p_j^h\}$ denote the eigenvalues of the cold and hot objects, respectively. If any of the following conditions hold:

1. The initial state of hot object is not fully mixed, i.e. $p_j^h \neq p_{j'}^h$ for some pair j, j',

2. The Hilbert space of the cold object has dimension $d_c \geq 3$, and $p_1^c > p_i^c$ or $p_{i+1}^c > p_{d_c}^c$ for some $1 \leq i \leq d_c$,

there exists a CNU1 transformation that increases the partial sum $\sum_{l=1}^{i} p_l^c$. Defining $\mu_i \equiv \max\left\{ \frac{p_1^h p_1^c}{p_{d_h}^h p_i^c} - \epsilon, \frac{p_1^h p_{i+1}^c}{p_{d_h}^h p_{d_c}^c} - \epsilon \right\}$, with $\epsilon > 0$ and ϵ such that $\mu_i > 0$, the transformation can be implemented by any catalyst of dimension $d_v = d_v^*$ that satisfies $d_v^* > \frac{1}{\ln(\mu_i)} \ln\left(\frac{p_i^c p_{d_h}^h}{p_{i+1}^c p_1^h} \right) + 1 \geq 2$.

Proof: We consider a full-rank catalyst state ρ_v, whose eigenvalues satisfy the condition

$$\frac{p_k^v}{p_{k+1}^v} = \mu_i \text{ for } 1 \leq k \leq d_v - 1. \quad (32)$$

From this equation it readily follows that $\prod_{k=1}^{d_v-1} \left(\frac{p_k^v}{p_{k+1}^v} \right) = \frac{p_1^v}{p_{d_v}^v} = \mu_i^{d_v-1}$.

If condition 1 holds, then $p_1^h > p_{d_h}^h$ and $\mu_i > 1$ for ϵ small enough. Note also that this is true even if ρ_c is fully mixed. If condition 2 holds, then $\max\left\{\frac{p_1^c}{p_i^c}, \frac{p_{i+1}^c}{p_{d_c}^c}\right\} > 1$ and consequently $\mu_i > 1$ (for ϵ sufficiently small). In this case, $\mu_i > 1$ even if ρ_h is fully mixed. The fact that $\mu_i > 1$ implies that there is always a finite dimension $d_v = d_v^*$ that fulfills

$$\mu_i^{d_v^*-1} = \frac{p_1^v}{p_{d_v}^v} > \frac{p_i^c p_{d_h}^h}{p_{i+1}^c p_1^h}. \tag{33}$$

This inequality is equivalent to $d_v^* > \frac{1}{\ln(\mu_i)}\ln\left(\frac{p_i^c p_{d_h}^h}{p_{i+1}^c p_1^h}\right)+1$ (as in the statement of the theorem), and guarantees the existence of a cooling current $\mathcal{J}_{|(i+1)_c 1_h 1_v\rangle \to |i_c d_h d_v^*\rangle}$. Moreover, we show in Appendix D that Eq. (32) guarantees a "left" restoring chain $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle \to |i_c d_h k_v\rangle}\}_{1\le k\le d_v^*-1}$, if $\mu_i = \frac{p_1^h p_1^c}{p_{d_h}^h p_i^c} - \epsilon$, or a "right" restoring chain $\{\mathcal{J}_{|(i+1)_c 1_h (k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{1\le k\le d_v^*-1}$, if $\mu_i = \frac{p_1^h p_{i+1}^c}{p_{d_h}^h p_{d_c}^c} - \epsilon$, with the strict positivity of ϵ being crucial for the proof. The restoring unitaries corresponding to these chains are respectively given by Eqs. (29) and (30), with $l = 1$ and $l' = d_v^* - 1$. In addition, the explicit catalyst eigenvalues can be straightforwardly derived from Eq. (32) (see Appendix E).

According to Theorem 2, CNU1 transformations are possible for almost any initial state $\rho_c \otimes \rho_h$, if a sufficiently large catalyst is available. In particular, condition 1 implies that any hot object with nondegenerate energy spectrum and finite temperature suffices. It is also worth pointing out that a harmonic oscillator constitutes an example of universal catalyst, in two complementary aspects. On the one hand, for a harmonic oscillator $d_v \to \infty$, which makes it suitable to implement a CNU1 transformation on any state $\rho_c \otimes \rho_h$ that adheres to the previous conditions. On the other hand, any CNU1 transformation that can be realized with a catalyst of finite dimension d_v, can also be performed with a harmonic oscillator. This is possible by simply populating d_v levels of the harmonic oscillator with the eigenvalues of the (finite-catalyst) state ρ_v.

From the definition of μ_i we can understand more clearly when a large dimension is required. If the ratio p_i^c/p_{i+1}^c is kept fixed, the quantities $p_{d_h}^h/p_1^h$ and μ_i determine how large must be d_v^* to fulfill Eq. (33). In particular, the r.h.s. of this inequality increases as $p_{d_h}^h/p_1^h$ tends to one. Conversely, μ_i decreases under this condition, and also for $p_1^c/p_i^c \sim 1$ and $p_{i+1}^c/p_{d_c}^c \sim 1$. We can thus conclude that if ρ_h is a very mixed state, and ρ_c is very mixed in the subspaces $\mathrm{span}\{|l_c\rangle\}_{1\le l\le i}$ and $\mathrm{span}\{|l_c\rangle\}_{i+1\le l\le d_c}$, a large value of d_v^* is needed to implement the transformation. In the context of cooling, this is consistent with the physical intuition that such a task involves more resources when performed with a very "hot" environment. On the other hand, in what

follows we show that even for a fully mixed hot object the ground population of the cold object can be catalytically increased. In such a case it is crucial that ρ_c is not completely mixed in at least one of the aforementioned subspaces, which is equivalent to Condition 2 of Theorem 2.

A. Catalytic cooling by reducing the mean energy $\langle H_c \rangle$

Now we are ready to present some examples of cooling transformations. First, we consider cooling transformations that decrease the mean energy of the cold object. If condition 1 of Theorem 2 holds it is possible to implement a CNU1 transformation using a restoring unitary V_{hv}, with $l = 1$ and $l' = d_v - 1$ in Eq. (31). Since this unitary does not affect the cold object, the transformation is guaranteed to cool the cold object via the current $J_{|(i+1)_c 1_h 1_v\rangle \to |i_c d_h d_v\rangle}$. Details on how condition 1 is sufficient to perform the transformation with V_{hv}, and the corresponding catalyst dimension, can be found in Appendix C.

Since the only effect of $U = U_{|(i+1)_c 1_h 1_v\rangle \leftrightarrow |i_c d_h d_v\rangle}^{(2)} \oplus (\mathbb{I}_c \otimes V_{hv})$ on the cold object is due to the cooling current, the mean energy $\langle H_c \rangle = \mathrm{Tr}[\rho_c H_c]$ decreases by

$$\begin{aligned}\Delta\langle H_c\rangle &= \mathrm{Tr}[(\rho_c' - \rho_c)H_c] \\ &= -J_{|(i+1)_c 1_h 1_v\rangle \to |i_c d_h d_v\rangle}(\varepsilon_{i+1}^c - \varepsilon_i^c).\end{aligned} \tag{34}$$

This cooling transformation is illustrated in Fig. 5(a). Importantly, a two-level system with eigenenergies $\varepsilon_1^h \ne \varepsilon_2^h$ and thermalized at finite temperature $\beta_h > 0$ serves as hot object. Moreover, it can be used to cool down any cold object by transferring population between some pair of consecutive eigenstates $|(i+1)_c\rangle$ and $|i_c\rangle$, regardless of its size. In Subsection C we will study the optimization of this transformation, when both ρ_c and ρ_h describe two-level systems.

We also point out that any transformation that reduces the energy of the cold object automatically decreases its von Neumann entropy. This stems from the positivity of the relative entropy $S(\rho_c'||\rho_c) \equiv -S(\rho_c') - \mathrm{Tr}(\rho_c' \ln \rho_c)$ [87], where $S(\rho_c') = -\mathrm{Tr}(\rho_c' \ln \rho_c')$ is the von Neumann entropy of ρ_c'. For a thermal state ρ_c at inverse temperature β_c, the relative entropy can be written as $S(\rho_c'||\rho_c) = \beta_c \Delta\langle H_c\rangle - \Delta S_c$, being $\Delta S_c = S(\rho_c') - S(\rho_c)$. Accordingly, if $\Delta\langle H_c\rangle < 0$ the von Neumann entropy must decrease to have $S(\rho_c'||\rho_c)$ positive. This implies that for any ρ_h not fully mixed we can construct a cooling transformation that reduces the value of S_c, following Eq. (34).

B. Catalytic cooling by increasing the population of the eigenspace with lowest energy

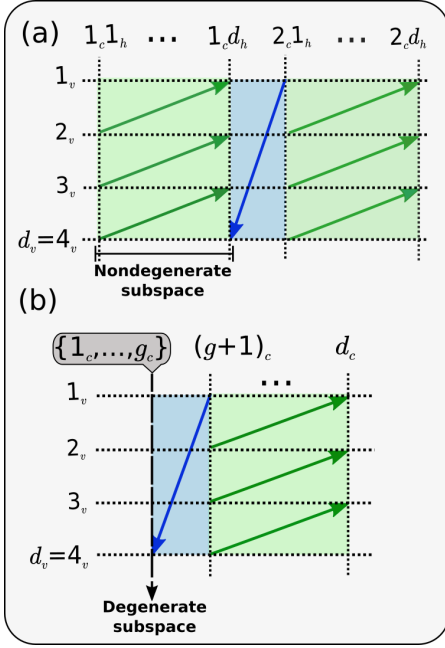Another approach for cooling consists of increasing the ground state population of the cold object. Based on our

Figure 5. Examples of catalytic cooling (CC) transformations. The figure illustrates two transformations where a four-dimensional catalyst suffices, taking into account the conditions of Theorem 2. (a) A CC transformation that cools down a cold qubit, and employs a restoring unitary V_{hv} performed on the catalyst and the hot object. In this case all the restoring currents occur inside the two energy eigenspaces of the the cold qubit (left and right green rectangles). (b) A CC transformation that increases the population in the ground subspace of the cold object, spanned by the minimum-energy eigenstates $\{|i_c\rangle\}_{1\leq i\leq g}$ (these degenerate eigenstates are depicted by the single dashed column). This type of cooling is possible without using the hot object.

previous discussion, if condition 1 of Theorem 2 holds we can generate a cooling current $J_{|1_c1_h1_v\rangle \to |1_c d_h d_v^*\rangle}$ that has this effect. However, it turns out that we can construct a CNU1 transformation that performs this kind of cooling, even if ρ_h is fully mixed. This transformation also has the particularity that the hot object is not needed. That is, $U = U_{cv}^{(2)} \oplus V_{cv}$, where both $U_{cv}^{(2)}$ (cooling unitary) and V_{cv} (restoring unitary) act jointly on the cold object and the catalyst.

For the sake of generality, let us consider that the ground energy ε_1^c is degenerate and that the goal is to increase the population in the corresponding eigenspace. In other words, we aim at increasing the average value of the projector $\Pi_1^c = \sum_{l=1}^g |l_c\rangle\langle l_c|$, where $\{|l_c\rangle\}_{1\leq l\leq g}$ are all the eigenstates with eigenenergy ε_1^c. The nondegenerate case is recovered for $g = 1$. By choosing $i = g$, the cooling current $J_{|(g+1)_c 1_v\rangle \to |g_c d_v^*\rangle}$ yields the increment

$$\Delta \langle \Pi_1^c \rangle = \mathrm{Tr}[(\rho_c' - \rho_c)\Pi_1^c]$$
$$= J_{|(g+1)_c 1_v\rangle \to |g_c d_v^*\rangle}. \tag{35}$$

On the other hand, suppose that $d_c \geq g + 2$, which means that the cold object possesses more than one ex-

cited eigenstate, and that $p_{d_c}^c < p_{g+1}^c$, which means that its highest eigenenergy is larger than the first excited eigenenergy ε_{g+1}^c. In this case condition 2 of Theorem 2 holds, and there exists a restoring unitary

$$V_{cv} = \oplus_{k=1}^{d_v^*-1} V_{|(g+1)_c(k+1)_v\rangle \leftrightarrow |d_c k_v\rangle}^{(2)}.$$

This unitary gives rise to a restoring chain $\mathbf{ch}_{|d_v^*\rangle \to |1_v\rangle} = \{J_{|(g+1)_c(k+1)_v\rangle \to |d_c k_v\rangle}\}$, which in turn forms a loop with $J_{|(g+1)_c 1_v\rangle \to |g_c d_v^*\rangle}$.

The physical mechanism behind the cooling effect is easy to understand. First, the cooling current transfers population from $|(g+1)_c\rangle$ to $|g_c\rangle$, which yields the increment $\Delta \langle \Pi_1^c \rangle = \Delta \langle |g_c\rangle\langle g_c| \rangle$. Since V_{cv} transfers population from $|(g+1)_c\rangle$ to $|d_c\rangle$, the restoring unitary does not interfere with this increment. The corresponding transformation is illustrated in Fig. 5(b).

C. Optimal catalytic cooling of a qubit using another qubit as hot object

Previously we mentioned that a system of infinite dimension constitutes a universal catalyst, in the sense that it enables CNU1 transformations for any state ρ_{ch} that complies with conditions 1 or 2 of Theorem 2. Moreover, we have seen that CC transformations can also be performed for almost any initial state ρ_{ch}. If this state is fixed, it is natural to ask which is the optimal catalytic cooling that can be achieved by using catalysts of different sizes. Here we address this question, regarding the simplest scenario of cooling of a qubit using another qubit as hot object. The cold and hot qubits start in states $\rho_c = p_1^c |1_c\rangle\langle 1_c| + p_2^c |2_c\rangle\langle 2_c|$ and $\rho_h = p_1^h |1_h\rangle\langle 1_h| + p_2^h |2_h\rangle\langle 2_h|$, and satisfy the inequality $p_2^c \leq p_2^h$ (no cooling condition without catalyst). In this case, all the cooling criteria considered before are equivalent to the increasing of the ground population of the cold object.

Without loss of generality, we can focus on the optimization of cooling for an infinite-dimension catalyst. Specifically, we consider the maximization of the cooling current with respect to the eigenvalues of a state ρ_v with *fixed rank* $2 \leq n < \infty$, which has support on a subspace of the infinite Hilbert space $\mathcal{H}_v$. Since a state of rank n is equivalent to a full-rank state for a catalyst of (finite) dimension n, the optimization for a given value of n yields also the maximum cooling using this catalyst. Accordingly, the optimal cooling using a finite catalyst can always be performed with one of infinite dimension. On the other hand, we will see that for certain values of p_2^c and p_2^h larger cooling currents can be obtained if n is small. Such a result is remarkable, as it implies that *in some cases small catalysts can be as effective as larger ones*. In what follows we assume that if ρ_v has rank n then $p_k^v = 0$ for all $k \geq n + 1$. That is, only the levels $1 \leq k \leq n$ are populated.

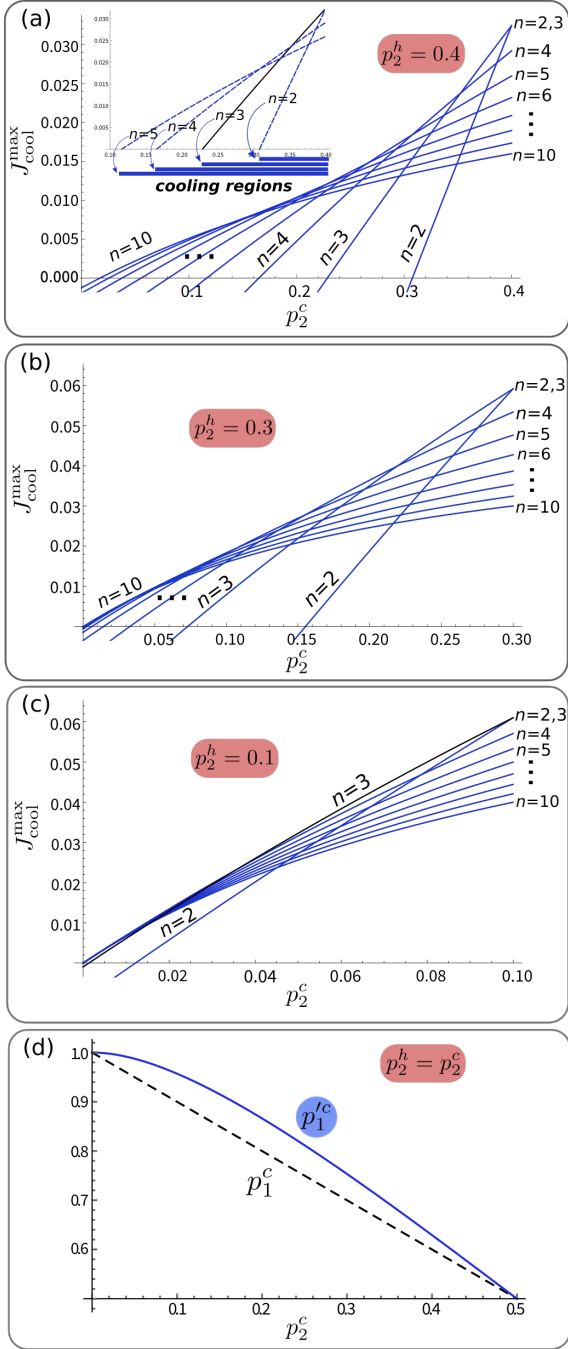In Appendix E we show that for n fixed the optimal

Figure 6. Optimal catalytic cooling of a qubit using another qubit as hot object. (a), (b) and (c) show the cooling currents obtained with catalyst states ρ_v of ranks $2 \leq n \leq 10$, and hot qubits with initial (excited) populations $p_2^h = 0.4$, $p_2^h = 0.3$, and $p_2^h = 0.1$, respectively. The inset in (a) shows the cooling regions corresponding to $2 \leq n \leq 5$, where states having these ranks allow to cool. Since the maximum of $J_{\text{cool}}^{\max}$ is achieved for $p_2^c = p_2^h$ and $n = 2, 3$, in (d) we consider the cooling using a two-level catalyst ($n = 2$) and a hot qubit such that $p_2^h = p_2^c$. The black dashed line depicts the initial ground population of the cold qubit and the blue curve is the corresponding final population.

CC unitary is given by

$$\mathcal{U} = \mathcal{U}_{\text{cool}} \oplus \mathcal{V}_{hv}, \tag{36}$$

$$\mathcal{V}_{hv} = \oplus_{k=1}^{n-1} \mathcal{V}_{|1_h(k+1)_v\rangle \leftrightarrow |2_h k_v\rangle} \tag{37}$$

where $\mathcal{U}_{\text{cool}} = \mathcal{U}_{|2_c 1_h 1_v\rangle \leftrightarrow |1_c 2_h n_v\rangle}$ is the swap between the states $|2_c 1_h 1_v\rangle$ and $|1_c 2_h n_v\rangle$, and $\mathcal{V}_{|1_h(k+1)_v\rangle \leftrightarrow |2_h k_v\rangle}$ is the swap between the states $|1_h(k + 1)_v\rangle$ and $|2_h k_v\rangle$. If $n = 4$ and we adopt the view that ρ_v describes a catalyst of dimension four, the currents generated by $\mathcal{U}$ are illustrated in Fig. 5(a). This structure is characterized by two restoring chains inside the energy eigenspaces of the cold qubit, and describes the effect of $\mathcal{U}$ for general values of n. The maximization of the cooling current $J_{\text{cool}} = J_{|2_c 1_h 1_v\rangle \leftrightarrow |1_c 2_h n_v\rangle}$ also yields catalyst eigenvalues that satisfy

$$\mathcal{J}_k = \mathcal{J}_{|2_c 1_h 1_v\rangle \leftrightarrow |1_c 2_h n_v\rangle} \text{ for } 1 \leq k \leq n - 1, \tag{38}$$

where $\mathcal{J}_k \equiv \mathcal{J}_{|1_h(k+1)_v\rangle \leftrightarrow |2_h k_v\rangle}$ is the current generated by $\mathcal{V}_{|1_h(k+1)_v\rangle \leftrightarrow |2_h k_v\rangle}$.

From Eq. (38), we find that the maximum cooling current reads (see Appendix E)

$$J_{\text{cool}}^{\max} = \frac{(1 - r_h) \left[p_1^{(h)} p_2^{(c)} - p_2^{(h)} p_1^{(c)} r_h^n \right]}{(1 - r_h) r_h^n + (1 - r_h^n) p_2^{(c)}} p_n^{(v)}, \tag{39}$$

where $r_h \equiv \frac{p_2^h}{p_1^h}$ and we have added parentheses to superscripts to distinguish them from powers. Moreover, the optimal eigenvalues $\{p_k^v\}$ are characterized by the equation

$$p_{n-k}^{(v)} = \frac{r_h^{1-k}}{p_2^{(h)}} \left[p_1^{(h)} - \left(\frac{1 - r_h^k}{1 - r_h} \right) \frac{J_{\text{cool}}^{\max}}{p_n^{(v)}} \right] p_n^{(v)}, \tag{40}$$

$1 \leq k \leq n - 1$, with $p_n^{(v)}$ deduced via normalization $\sum_k p_k^{(v)} = 1$.

Figure 5 shows plots of $J_{\text{cool}}^{\max}$ and the final ground population $p_1'^c$, for different values of p_2^h and catalyst states of ranks $2 \leq n \leq 10$. Each solid curve in Figs. 5(a)-(c) depicts the maximum cooling current corresponding to a different rank of ρ_v. Moreover, $J_{\text{cool}}^{\max}$ in Eq. (39) is plotted as a function of $0 \leq p_2^c \leq p_2^h$, which constitutes the interval where cooling without the catalyst is not possible. In Fig. 5(a) we can see that as n increases the interval of p_2^c where $J_{\text{cool}}^{\max}$ is positive also increases. Since $J_{\text{cool}}^{\max} < 0$ means that population would be transferred from the ground state to the excited state of the cold qubit, the "cooling region" is described by the condition $J_{\text{cool}}^{\max} \geq 0$. The inset in Fig. 5(a) shows more clearly the cooling regions (blue bars) corresponding to states of ranks $2 \leq n \leq 5$. The enlargement of these regions as n increases indicates that *larger catalysts may allow cooling in regimes not accesible to small catalysts*, characterized by $p_2^c \ll p_2^h$. On the other hand, for $p_2^c = p_2^h$ it is remarkable that $J_{\text{cool}}^{\max}$ is maximized by $n = 2$ and $n = 3$, and decreases for larger values of n. This implies that in such

a case *the smallest possible catalyst, corresponding to a two-level system, is enough to achieve maximum cooling.* Moreover, it is also worth noting that *the cooling current corresponding to $n = 3$ always surpasses the current corresponding to $n = 2$* (except for $p_2^c = p_2^h$).

Figures 5(b) and 5(c) display the same pattern that characterizes Fig. 5(a). In particular, notice that in both cases a catalyst state of rank $n = 10$ allows to cool for almost any value of p_2^c. In Fig. 5(c) we also see that a state of rank $n = 3$ (black curve) is essentially as effective as any state with rank $4 \leq n \leq 10$. Accordingly, in this case a *three-level catalyst is optimal for almost any value of p_2^c*. Figure 5(d) shows the initial and final ground populations as a function of p_2^c, if the populations of the hot and cold qubits always coincide. The final population is computed as $p_1'^c = p_1^c - J_{\text{cool}}^{\max}$, where $J_{\text{cool}}^{\max}$ is the cooling current attained for $n = 2$ or $n = 3$.

D. Catalyst-aided *enhancement* of cooling

The usefulness of catalysts is not restricted to the implementation of transformations that are forbidden without the utilisation of these systems. Here we show that cooling can be catalytically enhanced, even if the hot object is sufficient to achieve a certain level of cooling. This is formally stated in the following theorem, which constitutes our third main result. The proof is given in Appendix F and consists of two parts. A first one where a global unitary that yields maximum cooling without using the catalyst is derived, and a second part that involves the catalytic transformation. The construction of the catalytic transformation is similar to that employed for Theorem 2, with the explicit expression for the catalyst dimension provided below. We also remark that optimal cooling unitaries for a qubit interacting with a finite environment have been provided in Ref. [44]. However, we present a derivation based on passivity, in order to maintain a self contained structure.

Theorem 3 (cooling enhancement with a catalyst). Let ρ_h be the state of a hot object of dimension $d_h \geq 3$, and ρ_c the state of a cold qubit. If d_h is even and the largest $\frac{d_h}{2}$ eigenvalues or the smallest $\frac{d_h}{2}$ eigenvalues are not fully degenerate (i.e. $p_j^h \neq p_{j'}^h$ for some pair of the referred largest or smallest eigenvalues), or d_h is odd and the largest $\frac{d_h+1}{2}$ eigenvalues of ρ_h or the smallest $\frac{d_h+1}{2}$ eigenvalues of ρ_h are not fully degenerate, then a large enough catalyst *increases* the optimal cooling achieved with the hot object alone. To this end, it suffices to consider a catalyst dimension $d_v = d_v^*$ that satisfies $d_v^* > \frac{1}{\ln(\mu)} \ln\left(\frac{p_1^c p_{j_1}^h}{p_2^c p_{j_2}^h} \right) + 1 \geq 2$, where

$$\mu \equiv \max\left\{ \frac{p_1^h}{p_{j_1}^h} - \epsilon, \frac{p_{j_2}^h}{p_{d_h}^h} - \epsilon \right\}, \epsilon > 0 \text{ is such that } \mu > 1,$$

and j_1, j_2 are indices such that $j_1 \geq \frac{d_h}{2}$ and $j_2 \leq \frac{d_h}{2}$ ($j_1 \geq \frac{d_h+1}{2}$ and $j_2 \leq \frac{d_h+1}{2}$) for d_h even (odd).

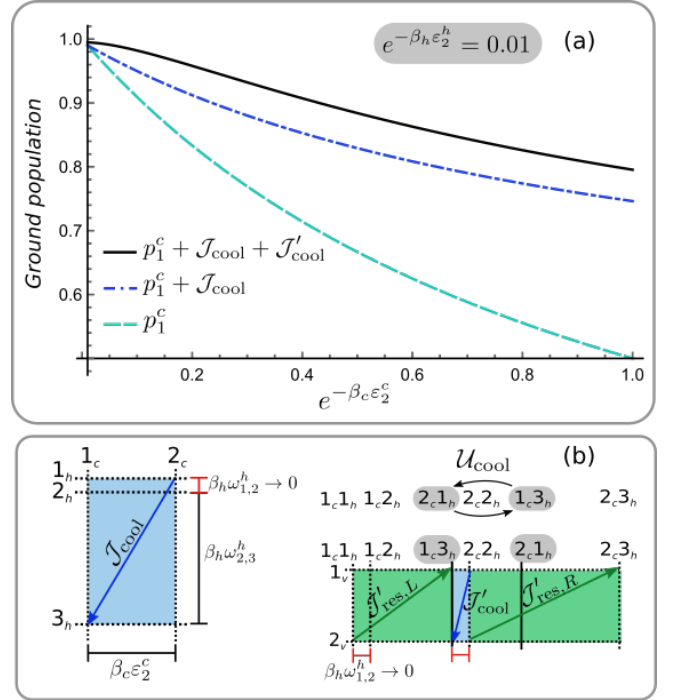To exemplify the catalytic improvement of cooling con-

Figure 7. Catalytic enhancement of cooling using a qubit as catalyst and a three-level system as hot object. (a) Initial ground population of the cold qubit (cyan dashed), ground population after optimal cooling without the catalyst (blue dashed-dotted), and ground population after a subsequent CC transformation (black solid). The parameter $e^{-\beta_h \varepsilon_2^h} = 0.01$ is fixed. (b) Schematic depiction of the optimal transformation without catalyst (left), and the additional CC transformation (right). The top right arrangement contains the labels for the eigenstates of $\rho_c \otimes \rho_h$, sorted according to decreasing eigenvalues. The lower $\ln(p^{ch}) \times \ln(p^v)$ diagram depicts the CC transformation with a two-level catalyst and the state $\mathcal{U}_{\text{cool}}(\rho_c \otimes \rho_h)\mathcal{U}_{\text{cool}}^\dagger$.

sider the minimal hot object that adheres to the hypothesis of Theorem 3, i.e. a three-level system. Let $H_h = \sum_{j=1}^3 \varepsilon_j^h |j_h\rangle\langle j_h|$ be the corresponding Hamiltonian, with $\varepsilon_1^h = \varepsilon_2^h = 0$ and $\omega_{2,3}^h \equiv \varepsilon_3^h - \varepsilon_2^h > 0$. For this energy spectrum a thermal state $\rho_h = \frac{e^{-\beta_h H_h}}{\text{Tr}(e^{-\beta_h H_h})}$ satisfies the hypothesis of the aforementioned theorem, since the two smallest eigenvalues p_2^h and p_3^h are non-degenerate.

In Fig. 7(a) we show the maximum cooling attainable using ρ_h, as well as an additional cooling through a CC transformation that employs a qubit as catalyst. The optimal cooling transformation without the catalyst is characterized in Appendix F, for a generic state ρ_h. The parameter $e^{-\beta_h \varepsilon_3^h}$ is set to $e^{-\beta_h \varepsilon_3^h} = 0.01$, which also fixes the eigenvalues of ρ_h due to the degeneracy of ε_1^h and ε_2^h. The blue dashed-dotted curve depicts the ground population of the cold qubit after the optimal cooling without catalyst, associated with the cooling current $\mathcal{J}_{\text{cool}} = \mathcal{J}_{|2_c 1_h\rangle \rightarrow |1_c 3_h\rangle}$. Moreover, the black solid curve stands for the final population after apply-

ing a suitable catalytic and cooling transformation. The left rectangle in Fig. 6(b) is a $\ln(p^c) \times \ln(p^h)$ diagram employed to illustrate the optimal transformation without the catalyst. In this diagram, we keep a small gap $\beta_h \omega_{1,2}^h = \beta_h(\varepsilon_2^h - \varepsilon_1^h) > 0$ that allows to distinguish the degenerate levels. However, it is indicated that $\beta_h \omega_{1,2}^h$ tends to zero, to comply with the degeneracy condition. Assuming that $\varepsilon_1^c = 0$, in the limit $\beta_h \omega_{1,2}^h \to 0$ the condition of cooling without the catalyst amounts to impose that $\ln(p_2^h/p_3^h) = \beta_h \omega_{2,3}^h > \beta_c \varepsilon_2^c = \ln(p_1^c/p_2^c)$. For $\beta_c \geq \beta_h$, this inequality holds if the energy gap $\omega_{2,3}^h$ is sufficiently large.

The right diagram of Fig. 7(b) illustrates the CC transformation that yields the black curve in Fig. 7(a). In this diagram the columns represent the eigenstates of the state $\mathcal{U}_{\mathrm{cool}}(\rho_c \otimes \rho_h)\mathcal{U}_{\mathrm{cool}}^\dagger$, where $\mathcal{U}_{\mathrm{cool}} = \mathcal{U}_{|2_c 1_h\rangle \leftrightarrow |1_c 3_h\rangle}$ is the swap that maximizes the cooling using only the hot object. By arranging the columns according to decreasing eigenvalues, we can apply the rules that determine cooling currents and restoring currents in a $\ln(p^{ch}) \times \ln(p^v)$ diagram, even if the state $\mathcal{U}_{\mathrm{cool}}(\rho_c \otimes \rho_h)\mathcal{U}_{\mathrm{cool}}^\dagger$ has not a product form. The only feature that we need to keep in mind is that now the values associated with the columns $2_c 1_h$ and $1_c 3_h$ are, respectively, $\ln(p_1^c p_3^h)$ and $\ln(p_2^c p_1^h)$ (see Fig. 7(b)). In this way, the depicted currents are generated by the unitary $\mathcal{U}' = \mathcal{U}_{\mathrm{cool}}' \oplus \mathcal{V}_{\mathrm{res},R}' \oplus \mathcal{V}_{\mathrm{res},L}'$, where $\mathcal{U}_{\mathrm{cool}}'$ and $\mathcal{V}_{\mathrm{res},X=R,L}'$ are swaps between the connected eigenstates.

It is important to mention that $\mathcal{U}'$ does not commute with $\mathcal{U}_{\mathrm{cool}}$, since $[\mathcal{U}_{\mathrm{cool}}', \mathcal{U}_{\mathrm{cool}}] \neq 0$. Therefore, the total transformation $\mathcal{U}'\mathcal{U}_{\mathrm{cool}}$ cannot be written in the direct sum form. This explains also why we require two independent diagrams for the representation of each transformation.

The restoring chain for the CC transformation contains the total current $\mathcal{J}_{\mathrm{res}}' = \mathcal{J}_{\mathrm{res},L}' + \mathcal{J}_{\mathrm{res},R}'$, where

$$\mathcal{J}_{\mathrm{res},L}' = p_1^c p_1^h p_2^v - p_2^c p_1^h p_1^v, \tag{41}$$

$$\mathcal{J}_{\mathrm{res},R}' = p_2^c(p_2^h p_2^v - p_3^h p_1^v). \tag{42}$$

From the condition $\mathcal{J}_{\mathrm{cool}}' = \mathcal{J}_{\mathrm{res}}'$ (uniform loop) and the degeneracy $p_1^h = p_2^h$, it follows that

$$\mathcal{J}_{\mathrm{cool}}' = \left(\frac{p_2^h p_1^c - p_3^h p_2^c}{(1 + p_2^c)p_2^h + p_2^c} \right) p_2^c p_2^h. \tag{43}$$

Remarkably, Fig. 7(a) shows that for low temperatures (β_c large) the increment of p_1^c due to the catalytic transformation is comparable to that achieved via optimal cooling without the catalyst. Moreover, the cooling enhancement provided by the catalyst is significant in all the temperature range.

In Fig. 8 we plot the cooling currents $\mathcal{J}_{\mathrm{cool}}$ and $\mathcal{J}_{\mathrm{cool}} + \mathcal{J}_{\mathrm{cool}}'$, where $\mathcal{J}_{\mathrm{cool}} + \mathcal{J}_{\mathrm{cool}}'$ is the current obtained from the total transformation $\mathcal{U}'\mathcal{U}_{\mathrm{cool}}$. In these plots $e^{-\beta_c \varepsilon_2^c}$ is fixed, and we vary instead the parameter $e^{-\beta_h \varepsilon_3^h}$. The condition $\beta_c \varepsilon_2^c > \beta_h \varepsilon_3^h$ (for cooling without the catalyst) implies that the maximum value of $e^{-\beta_h \varepsilon_3^h}$ must
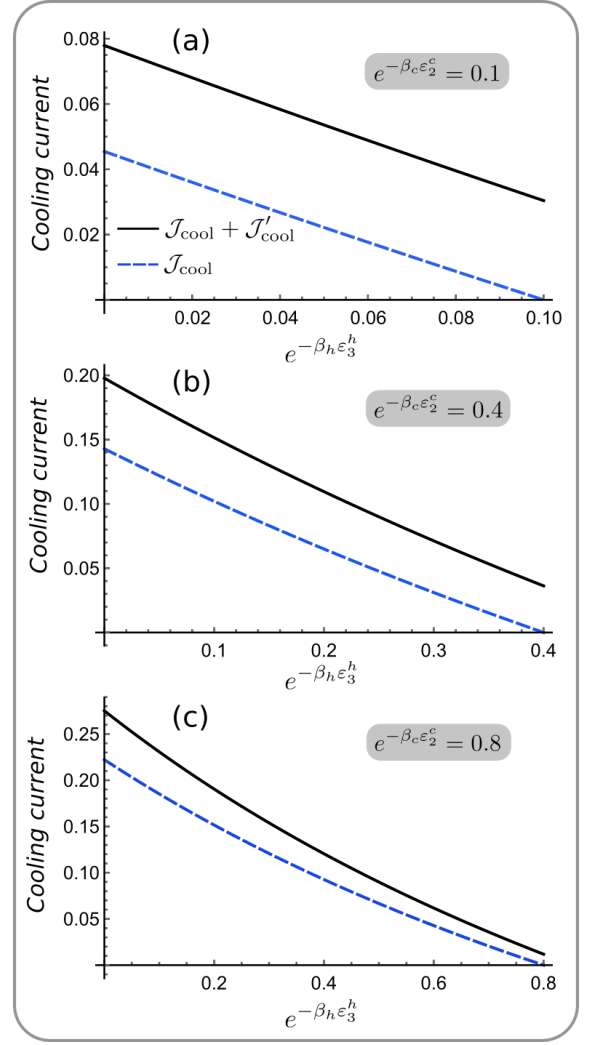

Figure 8. Cooling current obtained from the optimal transformation without the catalyst (dashed blue), and total cooling current after applying the catalytic transformation (solid black). The parameter $e^{-\beta_c \varepsilon_2^c}$ has the fixed values 0.1 (a), 0.4 (b), and 0.8 (c). The plotting variable $e^{-\beta_h \varepsilon_3^h}$ is increased until cooling without the catalyst is not possible, where $\mathcal{J}_{\mathrm{cool}} = 0$.

coincide with $e^{-\beta_c \varepsilon_2^c}$. When $e^{-\beta_h \varepsilon_3^h}$ increases, the hot object is hotter and consequently the cooling current $\mathcal{J}_{\mathrm{cool}}$ decreases, reaching its minimum value $\mathcal{J}_{\mathrm{cool}} = 0$ at $e^{-\beta_h \varepsilon_3^h} = e^{-\beta_c \varepsilon_2^c}$. On the other hand, we see again that the catalytic contribution is more significant the lower the cold temperature. To conclude this section, we remark that CC transformation considered here could be suboptimal, and therefore the advantage derived from the catalyst could be even larger.

VI. CATALYTIC ADVANTAGE WHEN COOLING A LARGE NUMBER OF COLD OBJECTS

In quantum thermodynamics and related research areas, the possibility to implement otherwise forbidden transformations has been one of the main motivations for the introduction of catalysts [1, 15, 17, 19, 22]. However, the restoration of the catalyst per se is not mandatory to achive that goal, and, on the contrary, it is expected that a potentially larger number of transformations can be reached if the restoration constraint is removed. While it is true that such transformations are not technically catalytic, there is no a priori reason for not to consider the catalyst simply as an ancilla, and implement a global unitary that optimizes the transformation on the system of interest.

A practical motivation to preserve the state of the catalyst is that it can be *reused* when necessary. For example, the repeated use of a chemical catalyst can substantially increase the rate of a chemical reaction. In the context of cooling, we can also imagine a situation where a large number of cold objects are cooled down by the repeated application of a catalytic transformation. When taken together, the cold objects and the corresponding hot objects employed for each transformation can bee seen as *larger (yet finite) environments*. A question that arises naturally in this scenario is how catalytic cooling compares to cooling strategies that do not use a catalyst. In particular, the passivity restriction that prevents cooling always breaks down for large enough environments, as explained in Sect. III. Here, we will show that *catalytic cooling can outperform a cooling strategy that uses arbitrary many-body interactions between cold objects and the hot environment* (formed by the hot objects). It is important to stress that the catalytic transformations involve at most three-body interactions. Therefore, the catalytic advantage is two-fold, since larger cooling is achieved with a lower degree of control on the environments.

A. Catalytic cooling vs. cooling using many-body interactions

Consider the scenario schematically depicted in Fig. 9. The goal is to cool as much as possible a group of N_c qubits, using a group of N_h qubits that play the role of a hot environment. All the qubits start at the same inverse temperature β and have identical energy spectrum. Therefore, the Hamiltonians of the ith cold and hot qubits are respectively $H_c^{(i)} = |1_c\rangle_i\langle 1_c|$ and $H_h^{(i)} = |1_h\rangle_i\langle 1_h|$. The total Hamiltonian for the $X = C, H$ group is $H_X = \sum_{i=1}^{N_X} |1_{x=c,h}\rangle_i\langle 1_{x=c,h}|$, and the global initial state is a product of thermal states $\rho_{CH} = \rho_C \otimes \rho_H$, where $\rho_X = \frac{e^{-\beta H_X}}{\text{Tr}(e^{-\beta H_X})}$. Assuming that the total number of qubits $N = N_c + N_h$ is fixed, we now describe two cooling strategies, illustrated in Fig. 9.
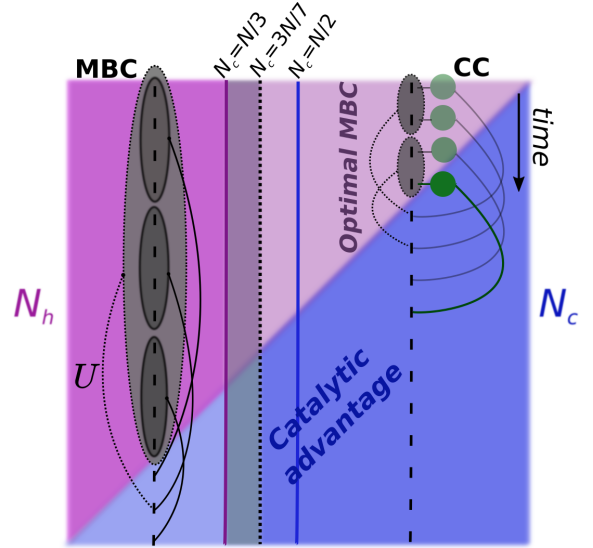

Figure 9. Catalytic cooling (CC) and Many-body cooling (MBC) strategies to cool a group of N_c cold qubits, using N_h hot qubits. The height of the square represents the fixed number $N_c + N_h$. A vertical line corresponds to a certain configuration (N_c, N_h), where N_c (N_h) is associated with the portion of the line in the blue (purple) region. The MBC and CC strategies are illustrated using respectively the left-most dashed line and the right-most dashed line, and described in detail in the main text. The connections between dashes and gray ellipses in the right-most line also represents the corresponding *optimal* MBC.

1. Many-body cooling (MBC) strategy: subsets of $2 \le k \le N_h$ qubits from the hot group are used to *optimally* cool *individual* qubits in the cold group, through *optimal unitary transformations*. Each qubit is cooled down only one time and the hot qubits pertaining to different subsets are all different (this implies that hot qubits are also used only once). Note also that $k \ge 2$, since all the qubits have identical states and therefore cooling is forbidden for $k = 1$.

2. Catalytic cooling (CC) strategy: a catalyst is employed to cool down *single* qubits from the cold group, using only *one* hot qubit per cold qubit. As with the MBC strategy, there is no reusage of hot qubits and each cold qubit is cooled down only one time.

In the MBC strategy the optimal cooling with a subset of k hot qubits involves $(k + 1)$-body interactions between these qubits and the corresponding cold qubit. More specifically, such couplings are described by an interaction Hamiltonian that contains products of the form $\otimes_{i=1}^{k+1} B_i$, where B_i is a non-trivial (i.e. different from the identity) operator on the Hilbert space of the ith qubit. On the other hand, the CC strategy is based on the repeated application of the unitary $\mathcal{U}$ in Eq. (36), for the case $n = 2$. This means that each cycle implements the optimal cooling of a single qubit using a two-level catalyst and one hot qubit. Importantly, the corresponding restoring unitary involves only a two-body interaction be-

tween the catalyst and the hot qubit, while $\mathcal{U}_{\text{cool}}$ requires a three-body interaction. In what follows we show that *even for large values of k, the CC strategy always outperforms the MBC strategy if $N_c \geq 3N/7$.*

The purpose of any of the described strategies is to reduce as much as possible the total average energy $\langle H_C \rangle$ of the cold qubits. Depending on the value of N_c, the number of qubits that can be cooled may be smaller than N_c. This limitation is directly associated with the amount of hot qubits available to perform the cooling. For example, if $N_c = N - 2$ only two hot qubits are available. In this case, two qubits can be cooled using the CC strategy and only one qubit can be cooled through the MBC strategy. That being said, it is important to remark that the following analysis covers all the possible values $1 \leq N_c \leq N-1$. Therefore, it provides a full picture of the task at hand, including also the situations where all the N_c qubits can be cooled. Taking this into account, the total heat extracted is given by

$$Q_C \equiv -\Delta \langle H_C \rangle = \sum_{i=1}^{n_c} \Delta p_1^{(i)}, \qquad (44)$$

where $n_c \leq N_c$.

B. Characterization of MBC

In the case of MBC, the maximum extractable heat Q_C can be conveniently addressed by introducing a coefficient that characterizes how efficient is the cooling of a single qubit, with respect to the number of hot qubits employed. This is a natural figure of merit in our scenario, taking into account that the hot qubits constitute a limited resource. Specifically, we define the "k-cooling coefficient" $\xi_{\text{cool}}^{(k)}$ as

$$\xi_{\text{cool}}^{(k)} \equiv \frac{Q_C^{(k)}}{k}, \qquad (45)$$

where $Q_C^{(k)}$ is the heat extracted by using a subset of $k \leq N_h$ hot qubits.

In the MBC strategy there are many ways in which the N_h hot qubits can be divided into cooling subsets. Two of such possibilities are illustrated in Fig. 9, for the case $N_c = 3$ and $N_h = 12$. Each dash in the leftmost dashed line represents one qubit, with cold qubits occupying the blue region and hot qubits the purple region (recall that both groups of qubits have the same temperature and therefore color difference is only used to distinguish them). One option is to cool each cold qubit using subsets of four hot qubits (darker gray ellipses), through global unitaries that are depicted by the lines joining these subsets with dashes in the cold region. Instead of that, we could use all the hot qubits (lighter gray ellipse) to cool down a single cold qubit. In general,

the heat extracted from this *single* qubit should be larger than the heat extracted by each four-qubit subset, keeping in mind that interactions with more hot qubits are allowed. However, a *larger number* of qubits are cooled down when several cooling subsets are employed. Since we are interested in the total heat Q_C, and not necessarily on maximizing the cooling for single qubits, it is not immediately clear which strategy wins.

By resorting to the cooling coefficient (45), we can express the total extracted heat as

$$Q_C = \sum_{k \in \boldsymbol{K}} Q_C^{(k)} = \sum_{k \in \boldsymbol{K}} \xi_{\text{cool}}^{(k)} k, \qquad (46)$$

where $\boldsymbol{K} = \{k_0, k_1, ...\}$ describes a certain partition of the hot group into cooling subsets. In particular, we note that $\sum_{k \in \boldsymbol{K}} k = N_h$, and that it is perfectly legitimate to have subsets of different sizes $k_i \neq k_j$. Given a *fixed* partition, we also have the bound

$$Q_C \leq \left(\max_{k \in \boldsymbol{K}} \xi_{\text{cool}}^{(k)} \right) \sum_{k \in \boldsymbol{K}} k = \left(\max_{k \in \boldsymbol{K}} \xi_{\text{cool}}^{(k)} \right) N_h. \qquad (47)$$

While the heat $Q_C^{(k)}$ is by construction a non-decreasing function of k, Fig. 10 provides numerical evidence that $\xi_{\text{cool}}^{(k)}$ is maximum for $k = 2$. For very large values of k it is also naturally expected that $\xi_{\text{cool}}^{(k)}$ tends to zero, since otherwise $Q_C^{(k)}$ would be an unbounded quantity. Therefore, we conjecture that

$$\xi_{\text{cool}}^{(k)} \leq \xi_{\text{cool}}^{(2)} = \left(\frac{1 - 2p_2^c}{2} \right) p_1^c p_2^c, \qquad (48)$$

for all $k \geq 2$ and for any β, which is satisfied for $2 \leq k \leq 14$ (see Fig. 10). The explicit expression for $\xi_{\text{cool}}^{(2)}$ is derived in Appendix G.

Although our conjecture and Eq. (47) seem to indicate that to maximize Q_C one should always choose minimal cooling subsets, composed of two qubits, this choice is actually optimal if N_c is above certain value. In this respect, we note that the maximization in Eq. (47) involves values of k characterizing a specific partition $\boldsymbol{K}$, and that only partitions such that all the cooling subsets are employed are meaningful. For example, to cool only one qubit ($N_c = 1$) it is clear that the best strategy consists of using $k = N_h$ qubits, which excludes any partition into cooling subsets. On the other hand, for $N_c \geq N_h/2$ (equivalently $N_c \geq N/3$) we can use $N_h/2$ cooling subsets of two qubits to cool $n_c = N_h/2 \leq N_c$ cold qubits. In this case all the cooling subsets are harnessed and the bound (47) is saturated with the maximum coefficient $\xi_{\text{cool}}^{(2)}$. More generally, we have that

$$\max_{\boldsymbol{K}} Q_C = \xi_{\text{cool}}^{(2)} N_h, \text{ if } N_c \geq \frac{N}{3}, \qquad (49)$$

$$2\xi_{\text{cool}}^{(2)} N_c \leq \max_{\boldsymbol{K}} Q_C \leq \xi_{\text{cool}}^{(2)} N_h, \text{ if } N_c \leq \frac{N}{3} - 1, \quad (50)$$
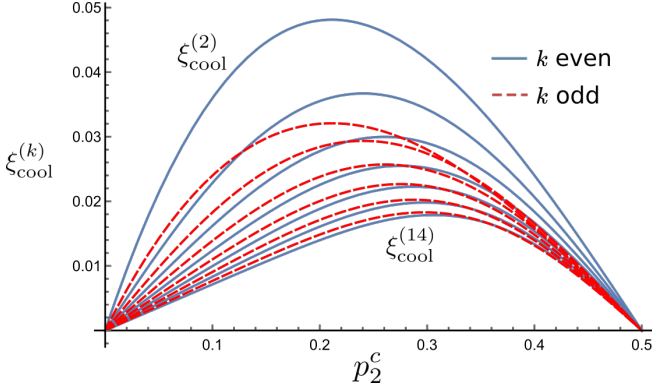
Figure 10. Cooling coefficient (45) curves for the cooling of a cold qubit using $2 \leq k \leq 14$ hot qubits. Blue solid (red dashed) curves stand for k even (odd). The highest and lowest curves correspond respectively to $\xi_{\text{cool}}^{(2)}$ and $\xi_{\text{cool}}^{(14)}$. Since $\xi_{\text{cool}}^{(k)} < \xi_{\text{cool}}^{(2)}$ for any value of p_2^c, this plot shows that the conjecture (48) is true for $2 \leq k \leq 14$.

where Eq. (49) indicates that partitions into subsets having more than two qubits are suboptimal. Importantly, this assertion depends on the validity of the conjecture $\xi_{\text{cool}}^{(2)} \geq \xi_{\text{cool}}^{(k)}$, for all $k \geq 2$.

The upper bound in Eq. (50) generalizes the bound (47) to all the possible partitions of $N_h \geq 2N/3 + 1$ hot qubits. This bound is in general not saturable, as already exemplified with the case $N_c = 1$. The corresponding lower bound follows from the fact that $N_h \geq 2N_c$ for $N_c \leq N/3 - 1$, whereby N_c subsets containing *at least* 2 qubits can be used to cool all the cold qubits. In such a case the heat extracted per cold qubit equals $2\xi_{\text{cool}}^{(2)}$. However, it is clearly more profitable to employ larger cooling subsets that allow to cool more each individual qubit, as illustrated with the four-qubit subsets in Fig. 9. This implies that the left inequality in (50) is in general estrict, and clarifies why in this regime minimal cooling subsets are not the optimal choice.

C. Advantage of the CC strategy

In contrast to the MBC strategy, the CC strategy has a more direct characterization. Let us denote as $Q_C^{(\text{CC})}$ the total extracted heat in this case, to distinguish it from the heat Q_C considered before. First, note that the CC strategy is by construction based on a cyclic operation where each cycle is optimized to maximize the cooling of a qubit, using a single hot qubit and a catalyst. This procedure is depicted in Fig. 9, where the four hot qubits (dashes) in the right-most dashed line are employed in a sequence of four cycles that cool four cold qubits. Since $p_1^{(i)} = p_1^{(j)}$ for all $1 \leq i, j \leq N$, we consider another qubit as catalyst (green circle in Fig. 9). This choice is based on the plots (a), (b) and (c) in Fig. 6, which show that when the hot qubit and the cold qubit have

the same populations a two-level catalyst maximizes the cooling. Accordingly, the maximum heat extracted per cycle is given by the cooling current (39), with $n = 2$ and $p_2^h = p_2^c = \frac{e^{-\beta}}{1 + e^{-\beta}}$. The total extracted heat is thus

$$Q_C^{(\text{CC})} = n_c \left(\frac{1 - 2p_2^c}{1 + 2p_1^c p_2^c} \right) p_1^c p_2^c, \qquad (51)$$

i.e. n_c times the aforementioned cooling current, being n_c the maximum number of cycles that can be implemented.

For $N_c \leq N/2$, all the cold qubits can be cooled down using $N_c \leq N_h$ hot qubits (which corresponds to $n_c = N_c$ cycles). On the other hand, for $N_c \geq N/2 + 1$ only $n_c = N_h < N_c$ qubits are cooled down but all the N_h qubits are consumed. Therefore, from Eq. (51) and the aforementioned conditions it follows that

$$Q_C^{(\text{CC})} = \begin{cases} N_c \left(\frac{1 - 2p_2^c}{1 + 2p_1^c p_2^c} \right) p_1^c p_2^c, & \text{if } N_c \leq \frac{N}{2}, \\ (N - N_c) \left(\frac{1 - 2p_2^c}{1 + 2p_1^c p_2^c} \right) p_1^c p_2^c, & \text{if } N_c \geq \frac{N}{2} + 1, \end{cases} \qquad (52)$$

where in the second line we write $n_c = N_h$ as $N - N_c$.

To perform the comparison between CC and MBC we introduce the relative performance ratio

$$\gamma \equiv \frac{Q_C^{(\text{CC})}}{\max_{\boldsymbol{K}} Q_C}, \qquad (53)$$

where both the numerator and the denominator refer to a fixed number of cold qubits and population p_2^c (which in turn characterizes the inverse temperature β). For $N_c \leq N/3 - 1$, the lower bound in Eq. (50) and Eq. (53) lead to

$$\gamma_{N_c \leq \frac{N}{3} - 1} \leq \frac{1 - 2p_2^c}{2\xi_{\text{cool}}^{(2)}(1 + 2p_1^c p_2^c)} = \frac{1}{1 + 2p_1^c p_2^c}. \qquad (54)$$

Clearly, $\gamma_{N_c \leq \frac{N}{3} - 1}$ is bounded from above by unity and therefore MBC outperforms CC in this regime.

If $N_c \geq N/3$, Eqs. (49) and (52) yield the following expressions for γ:

$$\gamma_{\frac{N}{3} \leq N_c \leq \frac{N}{2}} = \left(\frac{2}{1 + 2p_1^c p_2^c} \right) \frac{N_c}{N - N_c} \geq \frac{1}{1 + 2p_1^c p_2^c}. \qquad (55)$$

$$\gamma_{\frac{N}{2} + 1 \leq N_c \leq N} = \frac{2}{1 + 2p_1^c p_2^c}. \qquad (56)$$

The lower bound at the r.h.s. of Eq. (55) is tight only for $N_c = N/3$, which implies that the performance ratio is strictly larger if $N_c > N/3$. In particular, we are interested in values of N_c such that $\gamma_{N/3 \leq N_c \leq N/2} > 1$, since this means that the CC strategy is better than the MBC strategy. This condition leads to the equivalent inequality

$$\frac{N_c}{N} > \frac{1 + 2p_1^c p_2^c}{3 + 2p_1^c p_2^c}. \qquad (57)$$

The r.h.s. of such inequality varies between $1/3$, for $p_2^c = 0$, and $3/7$, for $p_2^c = 1/2$. Accordingly, in the regime $3N/7 < N_c \leq N/2$ the performance ratio (55) satisfies $\gamma_{3N/7 \leq N_c \leq N/2} > 1$. For the remaining interval $N/3 \leq N_c \leq 3N/7$, Eq. (57) provides an upper bound on p_2^c to have $\gamma > 1$.

On the other hand, from Eq. (56) it follows that $4/3 \leq \gamma_{N/2+1 \leq N_c \leq N} \leq 2$, which implies that in this regime the CC strategy outperforms the MBC cooling strategy, for *any* value of the inverse temperature β. We also stress that in the definition of γ the heat Q_C is optimized with respect to all the many-body interactions involving N_h qubits. Hence, even allowing arbitrary control over the available N_h hot qubits, the CC strategy with low control is more powerful in this regime.

The darker blue region in Fig. 9 depicts the regime (in terms of N_c) where the CC advantage takes place, irrespective of the inverse temperature β. The darker purple region corresponds to the regime (in terms of N_h) where MBC outperforms CC, and the gray-like region in between is the interval where the CC advantage is restricted to temperatures that obey Eq. (57). In particular, we note that in the limit $\beta \to \infty$ the fraction N_c/N satisfies this equation in all the interval $(1/3, 3/7)$. Therefore, for very low temperatures and in the limit $N \to \infty$, where such a fraction behaves approximately as a continuous variable, the CC advantage can be extended to the interval $(1/3, 1]$. The quantitative assessment of the performance ratio γ is provided in Fig. 11, following Eqs. (54)-(56). Importantly, we see once more that the CC advantage is strengthened as the temperature decreases, reaching a maximum value of $\gamma = 2$ for $\beta \to \infty$.

Finally, note that even if only $n_c < N_c$ qubits can be cooled down for a given value of N_c (e.g. if $N_c \geq N/2$), the remaining $N_c - n_c$ qubits are not *heated up* either. This condition guarantees that the temperature of all the N_c qubits remains below a certain treshold (in this case below β^{-1}), and may be important for some applications. In other words, it provides a justification for choosing a fixed value of N_c. On the other hand, one may alternatively be interested in optimizing the extracted heat with respect to N_c. In the case of the CC strategy it readily follows from Eq. (52) that such a maximum is attained for $N_c = N/2$. Regarding the MBC strategy, we can resort to the bound $\max_{\boldsymbol{K}} Q_C \leq \xi_{\mathrm{cool}}^{(2)} N_h$ (cf. Eqs. (49) and (50)) to determine if there are values of N_c such that $\max_{\boldsymbol{K}} Q_C$ *potentially* surpasses the maximum $Q_C^{(\mathrm{CC})}$. By writing N_h as $N_h = N - N_c$, we have that $\max_{\boldsymbol{K}} Q_C > \max_{N_c} Q_C^{(\mathrm{CC})}$ only if

$$\xi_{\mathrm{cool}}^{(2)}(N - N_c) > \frac{N}{2}\left(\frac{1 - 2p_2^c}{1 + 2p_1^c p_2^c}\right) p_1^c p_2^c, \qquad (58)$$

which after a simple algebra leads to the inequality

$$\frac{N_c}{N} < \frac{2p_1^c p_2^c}{1 + 2p_1^c p_2^c}. \qquad (59)$$

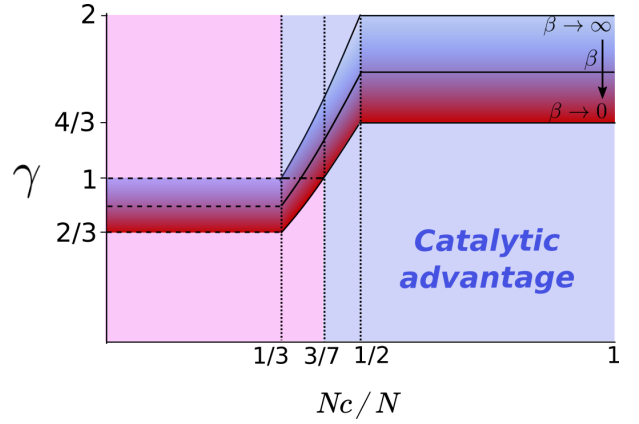The maximum value of the fraction N_c/N that satisfies this bound is achieved in the high temperature limit

Figure 11. Performance ratio γ (cf. Eq. (53)), as a function of the fraction of cold qubits N_c/N. The upper and lower boundaries of the blurred region correspond respectively to the limits $\beta \to \infty$ and $\beta \to 0$. For β finite, γ is described by a curve inside this region that has the same shape as those describing the boundaries, and whose vertical position increases with β. For $N_c/N \geq 1/3$ (continuous segment) the curve gives the exact value of γ, and for $N_c/N < 1/3$ (dashed segment) it yields an upper bound. The light blue (light red) area covers the regime where CC outperforms (is outperformed by) MBC. For $N_c/N \geq 3/7$, $\gamma > 1$ for all β. For $1/3 \leq N_c/N \leq 3/7$, $\gamma > 1$ iff β is contained between the upper boundary and the dash-dotted segment.

$\beta \to 0$, where the bound tends to $1/3$. Conversely, as β increases the bound becomes tighter, and in the limit $\beta \to \infty$ we have that the fraction must go to zero. This implies that for MBC to outperform the optimal (with respect to N_c) CC the number of cold qubits must be lower than $N/3$, and that *for very low temperatures optimal CC outperforms MBC for almost any value of N_c.* In particular, using Eqs. (48) and (49) we have that, *for any temperature,*

$$\frac{\max_{N_c} Q_C^{(\mathrm{CC})}}{\max_{N_c \geq N/3} Q_C} = \frac{3}{2}\left(\frac{1}{1 + 2p_1^c p_2^c}\right) \geq 1. \qquad (60)$$

VII. CATALYTIC THERMOMETRY

In this section we study an example where a catalyst is applied for precision enhancement in thermometry [74], where the goal is to estimate the temperature of a certain environment at thermal equilibrium. Let $\rho_e = \frac{e^{-\beta H_e}}{\mathrm{Tr}(e^{-\beta H_e})}$ denote the state of an environment with Hamiltonian $H_e = \sum_j \varepsilon_j |j_e\rangle\langle j_e|$, equilibrated at inverse temperature β. Essentially, a temperature estimation consists of assigning temperature values $\hat{T}_i$ to the different outcomes of a properly chosen observable O. In this way, the set $\{\hat{T}_i\}$ defines a temperature estimator $\hat{T}$, and the precision is assesed through the estimation error

$$\sqrt{\langle(\hat{T} - T)^2\rangle} \equiv \sqrt{\sum_i p_i(\hat{T}_i - T)^2}, \qquad (61)$$

where $T = \beta^{-1}$ is the actual temperature and p_i is the probability of measuring the outcome i.

The traditional approach to characterize the thermometric precision and also the precision in the estimation of more general physical parameters is based on the Fisher information [67]. This quantity determines a lower bound on the estimation error, known as the Cramer-Rao bound. In the case of thermometry, it is known that the Cramer-Rao bound is always saturated if $O = H_e$ [74]. That is, if the temperature estimation is carried out by directly performing energy measurements on the environment. Here we consider a different scenario, where an auxilliary ancilla or "probe" is used to extract temperature information via an interaction with the environment. Such a technique may be useful for example if the environment is very large and direct energy measurements are hard to implement. However, our main motivation is to show that the estimation error can be reduced below the minimum value attained only with the probe, by including an additional interaction with a catalyst. We consider a three-level environment with degeneracy $\varepsilon_1 = \varepsilon_2 = 0$, which is probed by a two-level system in the initial state $\varrho = p_1 |1_P\rangle\langle 1_P| + p_2 |2_P\rangle\langle 2_P|$, $p_1 > p_2$ (for simplicity we only use the subindex P for the eigenstates of ϱ). Moreover, the catalyst is also a two-level system in the initial state ρ_v. This setup is illustrated in Fig. 12(a). It also corresponds to the physical configuration studied in Sect. V-D, with the probe and the environment taking respectively the roles of the cold qubit and the hot object. As we will see, under suitable conditions the same catalytic transformation that allows cooling enhancement also allows precision enhancement in the temperature estimation.

We assume that $\hat{T}$ is an unbiased estimator, which means that its expectation value coincides with the actual temperature: $\langle \hat{T} \rangle = T$. It is important to mention that the assumption of unbiased estimators is common not only in thermometry but also for metrology in general. In particular, the Cramer-Rao bound limits the precision attained with this kind of estimators. If $\langle \hat{T} \rangle = T$ it follows that $\langle (\hat{T} - T)^2 \rangle = \mathrm{Var}(\hat{T})$, where $\mathrm{Var}(\hat{T}) = \langle \hat{T}^2 \rangle - \langle \hat{T} \rangle^2$ is the variance of $\hat{T}$. Moreover, it can be shown that if the temperature to be estimated belongs to a small interval $(T - \delta T, T + \delta T)$ (this is the so called "local estimation regime", most often studied in thermometry and other areas of metrology [88]), the estimation error using the observable O reads [74]

$$\sigma(\hat{T}) = \sqrt{\mathrm{Var}(\hat{T})} = \frac{\sqrt{\mathrm{Var}(O)}}{|\partial_T \langle O \rangle|},$$

where $\mathrm{Var}(O) = \langle O^2 \rangle - \langle O \rangle^2$ and $\partial_T \langle O \rangle = \frac{\partial}{\partial T} \langle O \rangle$. For the sake of covenience, we shall consider an "inverse temperature estimator" $\hat{\beta}$ instead of $\hat{T}$. The errors $\sigma(\hat{T})$ and $\sigma(\hat{\beta})$ are connected by the simple relation $\sigma(\hat{T}) = T^2 \sigma(\hat{\beta})$, which follows from the chain rule $\partial_\beta \langle O \rangle = -T^2 \partial_T \langle O \rangle$.

A. Optimal precision using only the probe and catalytic enhancement

In our example the observable $O_P = o_1 |1_P\rangle\langle 1_P| + o_2 |2_P\rangle\langle 2_P|$ describes a projective measurement on the probe, with eigenvalues o_1 and o_2. Information about β is encoded in the probe state $\varrho' = \mathrm{Tr}_e \left[U_{Pe}(\varrho \otimes \rho_e) U_{Pe}^\dagger \right]$, which results after a unitary evolution U_{Pe} that couples the probe with the environment. It is straightforward to check that in this case the estimation error reads

$$\sigma'(\hat{\beta}) = \frac{\sqrt{p_1' p_2'}}{|\partial_\beta p_1'|}, \tag{62}$$

where $p_1' = \mathrm{Tr}(|1_P\rangle\langle 1_P| \varrho')$.

The ratio in the r.h.s. of Eq. (62) constitutes the figure of merit in our analysis. On the one hand, under certain conditions one can find a unitary U_{Pe} that minimizes the product $p_1' p_2'$, and at the same time maximizes the quantity $\partial_\beta p_1'$. In such a case, the inequality

$$\min_{U_{Pe}} \sigma'(\hat{\beta}) \geq \frac{\min_{U_{Pe}} \sqrt{p_1' p_2'}}{\max_{U_{Pe}} |\partial_\beta p_1'|} \tag{63}$$

guarantees that the same operation minimizes the error $\sigma'(\hat{\beta})$. This implies that if a unitary U_{Pe} saturates the previous bound it also optimizes the temperature estimation by measuring only the probe.

On the other hand, we will see that when the bound (63) is saturable it is possible to apply a catalytic transformation such that

$$\frac{\sqrt{p_1'' p_2''}}{|\partial_\beta p_1''|} < \min_{U_{Pe}} \left(\frac{\sqrt{p_1' p_2'}}{|\partial_\beta p_1'|} \right), \tag{64}$$

where $p_1'' = \mathrm{Tr}(|1_P\rangle\langle 1_P| \varrho'')$, and ϱ'' is the probe state obtained after an interaction that involves a two-level catalyst. This means that such catalytic transformation further reduces the estimation error. Denoting the corresponding unitary evolution as U', the total transformation $\varrho \to \varrho''$ is implemented by a global unitary of the form $U'U_{Pe}$. Importantly, the optimal U_{Pe} and U' satisfy $U_{Pe} = \mathcal{U}_{\mathrm{cool}}$ and $U' = \mathcal{U}'$, being $\mathcal{U}_{\mathrm{cool}}$ and $\mathcal{U}'$ the cooling and catalytic cooling unitaries defined in Sect. V-D. This is not a coincidence, as we show below that the bound (63) can be saturated by maximally cooling the probe with the environment. In addition, the fact that $\mathcal{U}'$ yields a cooling enhancement for the probe (which here takes the role of the cold qubit) implies that it also reduces the product $p_1' p_2'$. Since we also show that $\partial_\beta p_1'' > \partial_\beta p_1'$, Eq. (64) follows.

B. Maximization of the population sensitivity in terms of passivity

In what follows we will refer to $\partial_\beta p_1'$ as the "population sensitivity", as it quantifies how the final population

p'_1 varies with respect to temperature changes. Defining $\rho_{Pe} = \varrho \otimes \rho_e$ and $\rho'_{Pe} = U_{Pe}\rho_{Pe}U^\dagger_{Pe}$, we can use the fact that U_{Pe} is independent of β to write the population sensitivity as

$$\partial_\beta p'_1 = \partial_\beta \mathrm{Tr}\left[|1_P\rangle\langle 1_P|\rho'_{Pe}\right]$$
$$= \mathrm{Tr}\left[|1_P\rangle\langle 1_P|U_{Pe}(\partial_\beta\rho_{Pe})U^\dagger_{Pe}\right]. \quad (65)$$

The operator $\partial_\beta\rho_{Pe} = \varrho \otimes \partial_\beta\rho_e$ has real eigenvalues $\{\lambda^{Pe}_{i,j}\} \equiv \{p_i\lambda^e_j\}$, where

$$\lambda^e_j \equiv \partial_\beta p^e_j = p^e_j\left(\langle H_e\rangle - \varepsilon_j\right), \quad (66)$$

and $\langle H_e\rangle = \mathrm{Tr}(H_e\rho_e)$. As we show next, this property allows us to analyze the maximization of the population sensitivity by applying the tools of passivity.

Since ρ_{Pe} is hermitian, the operator $A \equiv \partial_\beta\rho_{Pe} - \min\{\lambda^{Pe}_{i,j}\}\mathbb{I}$ is positive semidefinite. In this way, we can rewrite Eq. (65) in the form

$$\partial_\beta p'_1 = \mathrm{Tr}(A)\mathrm{Tr}\left[|1_P\rangle\langle 1_P|U_{Pe}\frac{A}{\mathrm{Tr}(A)}U^\dagger_{Pe}\right] + \min\left\{\lambda^{Pe}_{i,j}\right\},$$

where $\frac{A}{\mathrm{Tr}(A)}$ represents a density matrix (i.e. its eingevalues describe a probability distribution). Accordingly, maximizing $\partial_\beta p'_1$ is equivalent to maximize the expectation value of $|1_P\rangle\langle 1_P|$ over global unitaries that act on this (effective) initial state. Moreover, it is not difficult to see that this maximization is achieved by transforming $\frac{A}{\mathrm{Tr}(A)}$ in a passive state with respect to $-|1_P\rangle\langle 1_P|$. The definition of $\frac{A}{\mathrm{Tr}(A)}$ also implies that the corresponding passive state is obtained by a permutation that transfers the *three largest eigenvalues* of $\partial_\beta\rho_{Pe}$ to the eigenstates $\{|1_P p^e_j\rangle\}_j$. Consequently, the application of such a permutation yields an operator

$$\partial_\beta\rho'_{Pe} = |1_P\rangle\langle 1_P| \otimes \sum_j \lambda'^{Pe}_{1,j}|j_e\rangle\langle j_e|$$
$$+ |2_P\rangle\langle 2_P| \otimes \sum_j \lambda'^{Pe}_{2,j}|j_e\rangle\langle j_e|, \quad (67)$$

where $\{\lambda'^{Pe}_{i,j}\}$ is a rearrangement of the eigenvalues $\{\lambda^{Pe}_{i,j}\}$ that satisfies $\min_j \lambda'^{Pe}_{1,j} \geq \max_j \lambda'^{Pe}_{2,j}$.

C. Results

For an initial state ϱ such that $\beta\varepsilon_3 > \ln(p_1/p_2)$ (cf. Fig. 12(b) and the cooling transformation U_{cool} in Sect. V-D), the swap $U_{|1_P 3_e\rangle\leftrightarrow|2_P 1_e\rangle}$ optimally cools the probe, which amounts to maximize the value of $p'_1 p'_2$. On the other hand, it is easy to check that $U_{|1_P 3_e\rangle\leftrightarrow|2_P 1_e\rangle}$ is also a permutation that satisfies Eq. (67). This implies that in such a case $U_{|1_P 3_e\rangle\leftrightarrow|2_P 1_e\rangle}$ is an optimal unitary U^{op}_{Pe} that saturates the bound (63). However, we stress that *while this swap always maximizes the population sensitivity $\partial_\beta p'_1$, it also minimizes the error $\sigma'(\hat\beta)$ as long as*
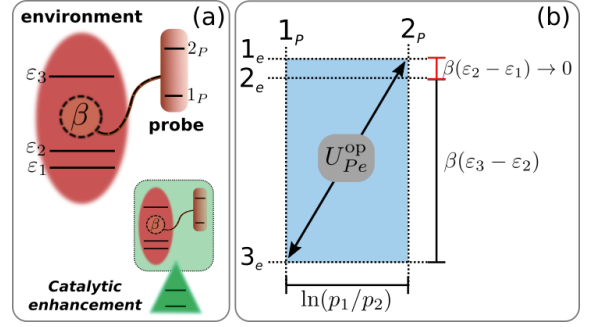

Figure 12. (a) The thermometric setup. Initially a two-level system optimally probes the temperature of a three-level environment. Afterwards, a joint interaction with a two-level catalyst (green triangle) reduces the minimum estimation error previously achieved. (b) $\ln(p) \times \ln(p^e)$ diagram for the initial probe-environment state $\varrho \otimes \rho_e$. If the condition $\beta(\varepsilon_3 - \varepsilon_2) \geq \ln(p_1/p_2)$ (cooling of the probe using only the environment) holds, the swap $U_{|1_P 3_e\rangle\leftrightarrow|2_P 1_e\rangle}$ is an optimal unitary U^{op}_{Pe} that minimizes the error $\sigma'(\hat\beta)$.

cooling is possible with the environment, see Fig. 12(b). Otherwise, any unitary U_{Pe} increases simultaneously the product $p_1 p_2$ and the population sensitivity, and *we cannot be certain that the maximization of $\partial_\beta p'_1$ is accompanied by a minimization of $\sigma'(\hat\beta)$.*

The dashed red curves in Fig. 13 show the estimation error obtained with $U_{|1_P 3_e\rangle\leftrightarrow|2_P 1_e\rangle}$, for three different initial states ϱ. In the same figure, the black (dashed-dotted) curves stand for the corresponding Cramer-Rao bound, which characterizes the minimum error that can be attained under POVMs (positive operator valued measurements) on the environment. Therefore, these curves are below the red ones, as expected. The estimation error achieved after the subsequent interaction with the probe and the catalyst is depicted by the blue curves in Fig. 13. The catalytic transformation has exactly the same form of the one illustrated in Fig. 7(b), if the labels c and h are substituted respectively by P and e. The corresponding final population sensitivity is given by $\partial_\beta p''_1 = \mathrm{Tr}\left[|1_P\rangle\langle 1_P|\partial_\beta\rho''_{Pe}\right]$, where $\rho''_{Pe} = \mathcal{U}'(\rho'_{Pe})\mathcal{U}'^\dagger$.

To understand why the population sensitivity is increased through $\mathcal{U}'$ it is convenient to write explicitly the sensitivity attained before the catalyst is employed. Specifically,

$$\partial_\beta p'_1 = \mathrm{Tr}_P\left[|1_P\rangle\langle 1_P|\mathrm{Tr}_e\partial_\beta\rho'_{Pe}\right]$$
$$= p_1(\lambda^e_1 + \lambda^e_2) + p_2\lambda^e_1,$$

where $\mathrm{Tr}_e\partial_\beta\rho'_{Pe}$ is computed from Eq. (67). Keeping in mind that $\mathcal{U}'$ is composed of (total) swaps between the states connected by the currents in Fig. 7(b), the final sensitivity reads

$$\partial_\beta p''_1 = \mathrm{Tr}_P\left[|1_P\rangle\langle 1_P|\mathrm{Tr}_e\partial_\beta\rho''_{Pe}\right]$$
$$= \partial_\beta p'_1 + p_2\left[p^v_1(\lambda^e_2) - p^v_2(\lambda^e_1)\right],$$

where the contribution $p_2\left[p^v_1(\lambda^e_2) - p^v_2(\lambda^e_1)\right]$ is due to the swap that generates the cooling current. Crucially,
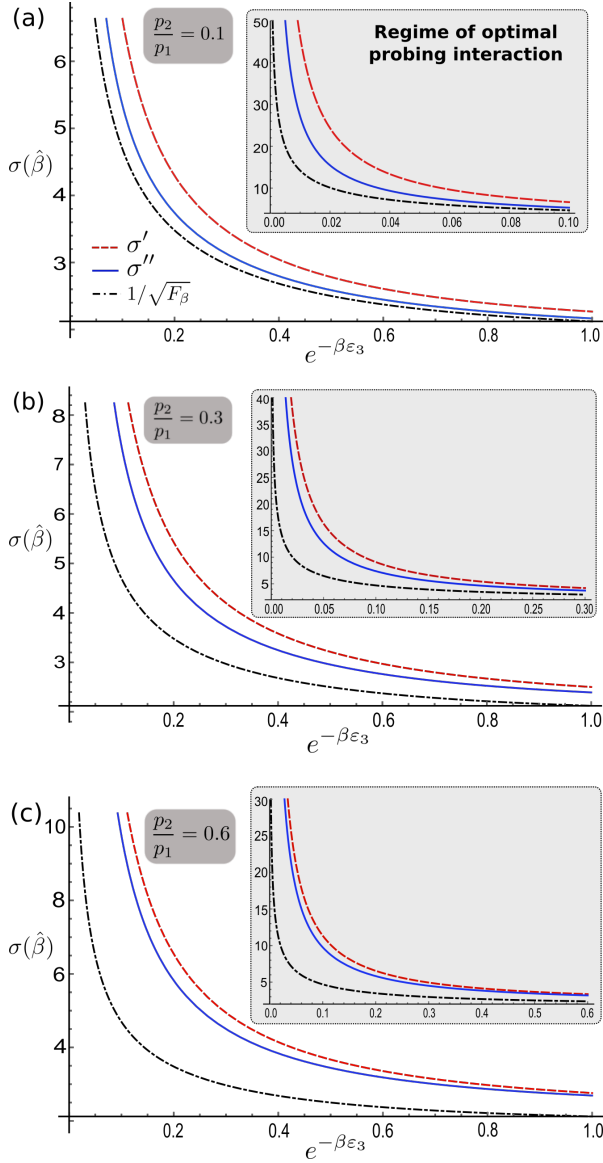
Figure 13. Estimation error $\sigma(\hat{\beta})$ for three initial probe states, characterized by the population ratios $p_2/p_1 = 0.1$ (a), $p_2/p_1 = 0.3$ (b), and $p_2/p_1 = 0.6$ (c). Red dashed and blue solid curves stand respectively for $\sigma'(\hat{\beta})$ and $\sigma''(\hat{\beta})$, which describe the error achieved without using the catalyst and the final error after applying the catalytic transformation. The black (dashed-dotted) curves show the thermal Cramer-Rao bound of the environment $1/\sqrt{F_\beta}$, where $F_\beta = \langle H_e^2 \rangle - \langle H_e \rangle^2$ is the associated Fisher information. The insets correspond to the range $e^{-\beta\varepsilon_3} \leq p_2/p_1$, where $\sigma'(\hat{\beta})$ is the *minimum estimation error* under general unitary evolutions that act jointly on the probe and the environment.

$\partial_\beta p_1'' > \partial_\beta p_1'$ iff this contribution is positive. Noting that $\lambda_1^e \geq 0$ (cf. Eq. (66)), it follows that the catalytic transformation increases the sensitivity iff $\lambda_2^e > 0$ and

$$\frac{p_1^v}{p_2^v} > \frac{\lambda_1^e}{\lambda_2^e}.$$

The degeneracy condition $\varepsilon_1 = \varepsilon_2$ implies that $\lambda_1^e = \lambda_2^e$, and consequently this inequality holds for any catalyst whose intial state is not fully mixed. In this way, the same catalytic transformation studied in Sect. V-D cools down the probe and simultaneously enhances the population sensitivity. Accordingly, the final estimation error $\sigma''(\hat{\beta}) = \frac{\sqrt{p_1'' p_2''}}{\left| \partial_\beta p_1'' \right|}$ is such that $\sigma''(\hat{\beta}) < \sigma'(\hat{\beta})$, which is illustrated by the fact that the blue solid curves are always below the red dashed ones in Fig. 13. The insets stand for the restricted intervals $e^{-\beta\varepsilon_3} \leq p_2/p_1$, where the swap that maximizes the population sensitivity (cf. Fig. 12) also minimizes $\sigma'(\hat{\beta})$ (by optimally cooling the probe). Therefore, in this region Eq. (64) is satisfied, i.e. $\sigma''(\hat{\beta}) < \min_{U_{P_e}} \sigma'(\hat{\beta})$. From Fig. 13 we see that both the precision without the catalyst and the catalytic advantage are more pronounced the purer is the initial state of the probe. In particular, for $p_2/p_1 = 0.1$ the catalytic transformation yields an error very close to the Crammer-Rao bound.

VIII. DISCUSSION AND OPEN PROBLEMS

A. Optimization of CNU and CC transformations

The proofs of Theorems 2 and 3 are constructive, meaning that besides ρ_v they also provide an explicit unitary U that performs the transformation. However, the proofs do not address the optimality of the pair (ρ_v, U), related to the maximization of the corresponding cooling current. As a first step in this direction, we optimized in Appendix E the pair catalyst-unitary for the maximum cooling of a qubit using a hot qubit. We found that the optimal U is given by *total* swaps (permutations) between the eigenstates connected in the loop. The first open question that we want to pose concerns the optimization for states ρ_c and ρ_h that describe higher dimensional systems. We refer specifically to the problem of increasing as much as possible a partial sum $\sum_{l=1}^i p_l^c$, for a given value of i. This is the core principle for CNU1 transformations and for CC transformations in combination with a restoring unitary $V_{\text{res}} = \mathbb{I}_c \otimes V_{hv}$.

We start by illustrating with a simple example the subtleties of this problem. Consider the top $\ln(p^{ch}) \times \ln(p^v)$ in Fig. 14. This diagram characterizes a state $\rho_c \otimes \rho_h \otimes \rho_v$ where both ρ_c and ρ_v are associated with qubits, and ρ_h is the state of a hot object with dimension $d_h = 4$. The solid (green and blue) arrows exemplify the basic configuration of currents for *all* the transformations studied until now (see Figs. 3, 5 and 7(b)), for a two-level catalyst. The hallmark of this structure is the presence of a single cooling current that joins two consecutive columns, and restoring currents that connect the *extreme columns* in the subspaces joined by the cooling current (green rectangles). On the other hand, dashed arrows indicate potential new currents that, if included, also lead to catalyst
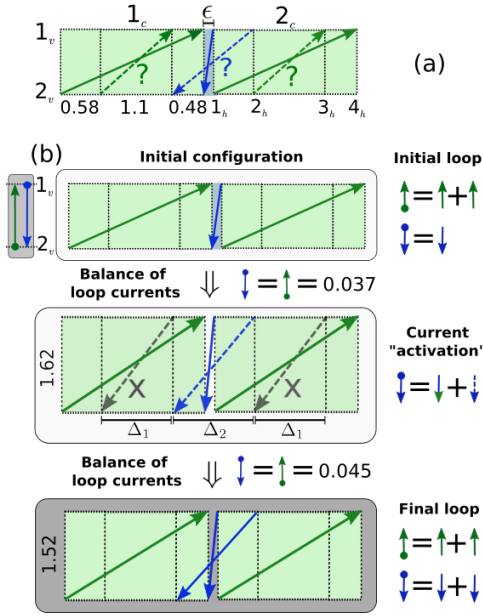
Figure 14. Example of application of the protocol in Appendix H, for the cooling of a qubit using a two-level catalyst and a four-level hot object. (a) $\ln(p^{ch}) \times \ln(p^v)$ diagram for the initial state. The eigenvalues of ρ_c and ρ_h are respectively $p_1^c = 0.897$ and $\{p_1^h, p_2^h, p_3^h\} = \{0.538, 0.3, 0.1\}$, which generate the corresponding distances between columns in the diagram ($\epsilon \sim 0$ implies passivity without the catalyst). Depending on the eigenvalues $\{p_k^v\}$, different currents may be activated (dashed arrows) apart from those associated with basic configuration (solid arrows). (b) The protocol starts from the basic configuration. This produces a loop (arrows with a circle in the tail, left) where the total restoring current is the sum of two currents and there is a single cooling current (right). After two steps (see main text) that yield catalyst eigenvalues with corresponding distances $\ln(p_1^v/p_2^v) = 1.62$ and $\ln(p_1^v/p_2^v) = 1.52$, the protocol finishes with a loop where the cooling current is the sum of two currents.

restoration through a loop. Hence, the question is if the inclusion of more currents increases the *total* cooling current and if so which ones should be added to maximize it.

In Appendix H we develop a protocol for the systematic targeting of new currents that increase the cooling current. On top of such a selection, it provides a recipe to optimize the corresponding catalyst state and singles out a permutation as optimal unitary. A single round of the protocol is illustrated in Fig. 14. To begin with, catalyst eigenvalues $\{p_k^v\}$ that yield the initial configuration are obtained through Eq. (32). The first step, described as "balance of loop currents", provides new eigenvalues $\{p_k^v(1)\}$ such that the total catalyst currents induced by permutations match each other. In a second step a useful new current (dashed blue arrow) is "activated". This current is useful in the sense that it contributes to the cooling, since it can be enclosed by a vertial rectangle in the diagram. Conversely, any current connecting the eigenstates joined by the dashed gray arrows can only

be useful as a restoring current (because it lives in an eigenspace of the cold qubit), and thus it should be enclosed by a horizontal rectangle. While this is the case in the initial configuration, for the new eigenvalues the gray arrows have an opposite effect and therefore they are not activated. After the addition of the new cooling current the resulting loop is not balanced. Thus, the following step produces eigenvalues $\{p_k^v(2)\}$ that balance the total restoring current with the new total cooling current. At this point the protocol ends because no new useful currents are available for activation.

The protocol produces a final (total) cooling current $J_{\text{cool}}(2) = 0.045$ that overcomes the cooling current at the first step, $J_{\text{cool}}(1) = 0.037$, and also the current $J_{\text{cool}} = 0.039$ that would result if all the dashed currents in the top diagram are activated. Although for this elemental example a single round suffices, it is an open question if larger systems require more iterations. Importantly, we demonstrate in Appendix E that the cooling current at each iteration can never decrease. Appart from the speed of convergence to the final loop, another relevant question is if this loop always corresponds to an optimal solution. These questions are specially meaningful when considering that the possible configurations of currents increase quickly as the size of the cold and hot objects increase.

Before moving to the next section some final remarks are in order:

- In the studied example the final loop contains an additional cooling current. However, it is not difficult to see that in some situations the initial loop is actually the best choice. If the eigenvalues p_1^h and p_4^h are kept fixed, the initial configuration (which guarantees that a two-level catalyst suffices) does not change. On the other hand, by reducing p_2^h and increasing p_3^h by the same amount the distance Δ_1 decreases and the distance Δ_2 increases. The maximum change leads to $p_2^h = p_3^h = 0.2$ and $\Delta_2^{\max} = 2.34$. Hence, for $1.62 \leq \Delta_2 \leq 2.64$ the blue dashed arrow reverts its direction and no new cooling current emerges, nor new restoring current.

- Coversely, Δ_1 can be increased up to a maximum $\Delta_1^{\max} = 1.69$ (corresponding to $p_2^h = 0.338$ and $p_3^h = 0.062$). If $1.62 < \Delta_1 \leq 1.69$, restoring currents connected by the dashed gray arrows are activated, since $\Delta_1 > 1.62$. Moreover, the activation of the new cooling current is maintained because Δ_2 decreases. In this case the loop balance including all the currents (as in Fig. 14(a)) potentially leads to a larger $J_{\text{cool}}(2)$, as compared to the balance for the loop without the new restoring currents. However, as explained in step 4 of the general protocol (see Appendix H), there exists the possibility that currents activated *before* the balance are flipped *after* the balance and thus they should be avoided in the next iteration. This may happen because the balance step modifies the catalyst eigenvalues and

consequently the currents themselves.

- The preceding observations indicate that a naive adjustment of the catalyst eigenvalues, e.g. to generate new cooling currents, is not necessarily a good optimization strategy. Instead of that, the protocol operates via a feedback process where first the best eigenvalues for a *fixed* loop are derived, and then the loop is improved (by activating or deactivating currents) keeping fixed such eigenvalues. This procedure is what ultimately ensures that the resulting (total) cooling current never decreases, formally demonstrated in Appendix H.

B. Explicit catalytic transformations

The study of catalytic transformations has been mainly driven by an "implicit" approach, in which the existence of the transformation is deduced but its implementation and the corresponding catalyst state are generally unknown. This approach is especially suited to derive fundamental constraints on state transitions within a resource-theoretic context. Catalytic thermal operations [15, 19] and catalyst-aided LOCC operations for entanglement manipulation [4] constitute paradigmatic examples. On the other hand, explicit catalytic transformations have been applied to specific tasks such as work extraction [17, 22, 23] and randomness generation [11]. Formulating explicit and more general transformations is an important step towards a more complete understanding of the phenomenon of catalysis at the microscopic level. The class of CNU transformations, introduced here, is partially motivated by such a quest. While our focus has been cooling, these transformations can be useful in any context where a catalyst allows to overcome limits imposed by unital evolution. This scenario encompasses themometry using a probe, as illustrated here, as well as the tasks considered in Refs. [17, 22, 23].

The class of CNU transformations addressed in this paper relies on a weak condition for violating majorization. Namely, the increment of the partial sum (10). Although this endows the formalism with a good degree of generality, it is worth mentioning that Eq. (10) is sufficient but not necessary for the violation of majorization, since it refers to the populations of the final state rather than its eigenvalues. Another possible avenue to extend the scope of the transformations is to consider their implementation through global unitaries that do not exhibit the structure $U = U_{\text{cool}} \oplus V_{\text{res}}$. On the other hand, Theorem 2 corroborates that this set of unitaries is general enough to ensure that almost any initial state can be transformed in a non-unital way, given access to a sufficiently large (single-copy) catalyst. Accordingly, a more interesting motivation to further generalize the transformations concerns its optimality (note that the protocol exemplified in the previous section addresses this problem within the set $\{U_{\text{cool}} \oplus V_{\text{res}}\}$). Such an optimality can also
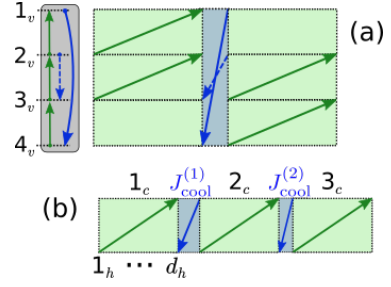

Figure 15. Two examples of CNU transformations that remain to be explored. (a) A transformation where the loop (structure at the left, enclosed by the gray rectangle) contains an additional cooling current between the rows 2_v and 3_v. While this current is not necessary for catalyst restoration (if it is removed we recover the "standard" loop in Fig. 4), it is interesting to verify if it can increase the total cooling current, once the corresponding catalyst state is optimized. (b) A transformation that reproduces the loop structure in Fig. 4 but contains two cooling currents $J_{\text{cool}}^{(1)}$ and $J_{\text{cool}}^{(2)}$ that increase different partial sums of populations. The figure depicts specifically a CC transformation to cool a three-level system, using a two level catalyst and a hot object of dimension d_h.

have a more clear meaning in the context of applications. For example, Ref. [9] shows that it is possible to catalytically increase the purity of a qubit, to a value arbitrarily close to one, if its initial (von Neumann) entropy satisfies $S \leq 1/2$. The setting is reminiscent to that of Fig. 6(d), where another qubit that starts in an identical state is employed as heat sink. However, the catalyst of Ref. [9] is implicit and therefore its dimension is unknown [89]. For $p_2^c \lesssim 0.11$ in Fig. 6(d) (which reproduces the condition $S \leq 1/2$) we see that a two-level catalyst already yields a remarkable increase of purity, with $p_1'^c \sim 1$ for $p_2^c \lesssim 0.06$. The optimal cooling of larger systems using small catalysts is a problem where the tools developed here could prove useful.

On the other hand, it turns out that the set $\{U_{\text{cool}} \oplus V_{\text{res}}\}$ can induce a rich variety of CNU transformations that remain to be explored. In fact, catalyst restoration is guaranteed whenever its eigenvalues allow the generation of a loop (cf. Definition 3). In Fig. 15 we show two examples of transformations that satisfy this condition and differ from those considered before. For the transformation of Fig. 15(a) the loop structure presents a distinctive feature. Specifically, the total outgoing current from the catalyst eigenstate $|2_v\rangle$ has two contributions: one from a restoring current (upper green arrow), and another from a cooling current (dashed blue arrow). The key feature is that these currents transfer population in opposite directions: one towards the upper row $|1_v\rangle$, and the other towards the lower row $|3_v\rangle$. A similar effect takes place with the total incoming current for the eigenstate $|3_v\rangle$. This contrasts with previous transformations, where incoming and outgoing currents only have contributions from a single direction, as in Fig. 4. The transformation of Fig. 15(b) adheres to the "standard" loop

but allows to increase more than one partial sum. That is, $p_1'^c = p_1^c + J_{\text{cool}}^{(1)}$ and $\sum_{l=1}^{2} p_l'^c = \sum_{l=1}^{2} p_l^c + J_{\text{cool}}^{(2)}$. In this context, pertinent questions could be which is the maximum value of $J_{\text{cool}}^{(1)}$ given a fixed value of $J_{\text{cool}}^{(2)} > 0$ (or vice versa), or which is the maximum of $J_{\text{cool}}^{(1)} + J_{\text{cool}}^{(2)}$ given that $J_{\text{cool}}^{(i=1,2)} > 0$. It is worth mentioning that for both transformations in Fig. 15 $U_{\text{cool}} = \oplus_i U_i^{(2)}$ and $V_{\text{res}} = \oplus_k V_k^{(2)}$, where $U_i^{(2)}$ ($V_k^{(2)}$) are partial swaps between states connected by the cooling (restoring) currents. Hence, another question is whether in this case permutations also lead to optimal solutions.

IX. CONCLUSIONS AND OUTLOOK

In this paper, we introduced tools for the systematic construction of catalytic transformations on quantum systems of finite size. Size limitations constrain tasks such as cooling using a finite environment or thermometry with a very small probe. In the case of cooling, we showed that the introduction of a catalyst lifts cooling restrictions in two complementary ways: catalysts enable cooling when it is impossible using only the environment, and enhance it when the environment suffices to cool. These results were illustrated with several examples regarding the cooling of a single qubit. In particular, we found that small catalysts such as three-level systems allow maximum cooling in wide temperature ranges. We also demonstrated that to cool a system of any dimension a large enough catalyst and any environment that starts in a non-fully mixed state are sufficient. Moreover, the ground population of the system can be catalytically increased, without requiring any interaction with an environment. Another advantage of catalytic cooling was shown in a setup consisting of many qubits prepared in identical states, where a subset of qubits is employed as environment to cool another subset. In this system, we found that it is possible to outperform the cooling achieved through many-body interactions with the environment, by including a two-level catalyst that cools using at most three-body interactions.

An application to thermometry was illustrated by considering a three-level environment whose temperature is probed by a two-level system, where we demonstrated that the inclusion of a two-level catalyst enhances the precision of the temperature estimation. It is worth remarking that this is the smallest possible setup where a catalyst may provide an advantage with respect to optimal interactions between the probe and the environment. For example, a two-level environment can be directly swapped with a two-level probe, which allows to saturate the thermal Cramer-Rao bound by performing appropriate measurements on the probe. A similar argument also implies that for larger environments probes that have at least the same dimension allow optimal thermometry. Hence, in contrast to cooling, a catalyst yields a thermometric enhancement by circumventing size lim-

itations that do not refer to the environment, but rather to the probe. Beyond the example mentioned above, an interesting direction for future work is to determine more general conditions for catalytic advantages in thermometry. This includes to further study the roles of initially mixed probes and the sizes of the involved systems.

Is is also pertinent to precisely characterize the role of the catalyst state and its dimension in different transformations. For example, we saw that large catalysts may be necessary to operate on very mixed objects. However, there exists the possibility that catalysts smaller than those descibed in Theorems 2 and 3 also allow to implement the corresponding transformations. Regarding the cooling of many qubits studied in Sect. VI, it woud be interesting to investigate how the catalytic and non-catalytic strategies compare when considering higher-dimensional systems. In this scenario, it is meaningful to vary not only the size of the catalyst but also the size of the subsystems that compose the cold and hot objects, as well as their temperatures. An important aspect of the catalysts states considered here is that they describe a single system. Recently, it was shown that catalytic thermal operations exhibit a feature of "universality" [20], which means that the associated thermodynamic transitions can be performed using sufficient copies of generic catalyst states. The authors also conjecture that this phenomenom could be valid for transformations with single-copy catalysts of high enough dimension, and provide some numerical evidence. Despite being formulated in a different context, we hope that our findings can provide some clues for the solution of this puzzle.

Appendix A: Proof of Theorem 1

Consider a system in an initial state ρ_s, with eigenvalues $\{p_i^s\}_{1 \leq i \leq d_s}$ (where d_s is the dimension of $\mathcal{H}_s$), and a catalyst in an initial state ρ_v, with eigenvalues $\{p_i^v\}_{1 \leq i \leq d_v}$. In this appendix we demonstrate that the conditions:

$$\frac{p_l^v}{p_{l'+1}^v} > \frac{p_i^s}{p_{i+1}^s} > 1, \tag{A1}$$

for some $1 \leq i \leq d_s - 1$ and $1 \leq l, l' \leq d_v - 1$; and, for $l \leq k \leq l'$,

$$\frac{p_1^s}{p_i^s} > \frac{p_k^v}{p_{k+1}^v} > 1, \qquad (A2)$$

$$or \; \frac{p_{i+1}^s}{p_{d_c}^s} > \frac{p_k^v}{p_{k+1}^v} > 1, \qquad (A3)$$

are necessary and sufficient for a CNU1 transformation on the system. The right-most inequalities follow from the convention $p_j^s \geq p_{j+1}^s$ and $p_j^v \geq p_{j+1}^v$ for all j. By choosing $\rho_s = \rho_c \otimes \rho_h$, the necessary and sufficient conditions (19)-(21) for a CNU1 transformation on ρ_c are obtained.

Sufficiency. Equation (A1) is equivalent to

$$p_l^v p_{i+1}^s > p_{l'+1}^v p_i^s,$$

for some i and some l, l'. This guarantees the existence of a cooling current $J_{|(i+1)_s l_v\rangle \to |i_s (l'+1)_v\rangle}$ that increases the partial sum $\sum_{j=1}^i p_j^s$ of the system, by $J_{|(i+1)_s l_v\rangle \to |i_s (l'+1)_v\rangle}$.

If Eq. (A2) holds then there is a restoring chain $\{J_{|1_s(k+1)_v\rangle \to |i_s k_v\rangle}\}_{l \leq k \leq l'}$, such that $J_{|1_s(l'+1)_v\rangle \to |i_s l'_v\rangle}$ cancels the population increment (on the catalyst) generated by $J_{|(i+1)_s l_v\rangle \to |i_s(l'+1)_v\rangle}$, and $J_{|1_s(l+1)_v\rangle \to |i_s l_v\rangle}$ restores the population taken away by the same current. Moreover, for $l+1 \leq k \leq l'-1$ any population variation due to a restoring current is cancelled by another restoring current. For example, the effects of $J_{|1_s(l+2)_v\rangle \to |i_s(l+1)_v\rangle}$ and $J_{|1_s(l+3)_v\rangle \to |i_s(l+2)_v\rangle}$ on the state $|(l+2)_v\rangle$ cancel each other. Since this restoring chain acts within the subspace $\mathrm{span}\{|j_s\rangle\}_{1 \leq j \leq i}$, it does not affect the change on the partial sum $\sum_{j=1}^i p_j^s$ generated by the cooling current.

If Eq. (A3) holds, there is a restoring chain $\{J_{|(i+1)_s(k+1)_v\rangle \to |d_s k_v\rangle}\}_{l \leq k \leq l'}$ that cancels the effect of $J_{|(i+1)_s l_v\rangle \to |i_s(l'+1)_v\rangle}$ (on the catalyst) through the currents $J_{|(i+1)_s(l'+1)_v\rangle \to |d_s l'_v\rangle}$ and $J_{|(i+1)_s(l+1)_v\rangle \to |d_s l_v\rangle}$, and also leaves unchanged the populations corresponding to $l+1 \leq k \leq l'-1$. The partial sum $\sum_{j=1}^i p_j^s$ is neither affected by this chain, given that it only transfers population within the subspace $\mathrm{span}\{|j_s\rangle\}_{i+1 \leq j \leq d_s}$.

Necessity. If Eq. (A1) does not hold, there is no cooling current $J_{|(i+1)_s l_v\rangle \to |i_s(l'+1)_v\rangle}$ and by definition it is impossible to implement a CNU1 transformation. On the other hand, let us see that if Eqs. (A2), (A2) are both violated, any catalytic transformation must be unital (even if the cooling current exists).

Suppose that Eq. (A2) does not hold. This is equivalent to say that $p_1^s p_{k+1}^v - p_i^s p_k^v \leq 0$ for some $l \leq k \leq l'$, and consequently that the *left* restoring chain $\{\mathcal{J}_{|1_s(k+1)_v\rangle \to |i_s k_v\rangle}\}_{l \leq k \leq l'}$ does not exist. Crucially, any restoring unitary mapping the "left subspace" $\mathrm{span}\{|j_s\rangle\}_{1 \leq j \leq i} \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$ into itself exists iff the chain $\{\mathcal{J}_{|1_s(k+1)_v\rangle \to |i_s k_v\rangle}\}_{l \leq k \leq l'}$ exists. This result follows after a straightforward generalization of Lemma 1. This Lemma states that, if $\rho_s = \rho_c \otimes \rho_h$, a restoring unitary exists iff there exists a restoring chain $\{\mathcal{J}_{|1_c 1_h(k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l \leq k \leq l'}$. The particularities in this case are the form of ρ_s (i.e. $\rho_s = \rho_c \otimes \rho_h$) and the fact that the restoring unitary is defined on the "whole" space $\mathcal{H}_s \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$ (by "whole" we mean that the full Hilbert space of the *system* is involved). However, the crucial property of such a chain is independent of these details, and consequently the restoring condition can be easily extended to generic states ρ_s and generic subspaces $\mathcal{G}_s \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$, $\mathcal{G}_s \subseteq \mathcal{H}_v$.

Specifically, the chain $\{\mathcal{J}_{|1_c 1_h(k+1)_v\rangle \to |d_c d_h k_v\rangle}\}_{l \leq k \leq l'}$ is characterized by the fact that each restoring current connects the *maximum-eigenvalue* eigenstate of $\rho_c \otimes \rho_h$ (given by $|1_c 1_h\rangle$) with its *minimum-eigenvalue* eigenstate (given by $|d_c d_h\rangle$). If we consider a general state ρ_s and a subspace $\mathcal{G}_s \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$, the restoring condition can be recast by defining a chain whose restoring currents connect eigenstates of ρ_s, with maximum and minimum eigenvalues restricted by $\mathcal{G}_s$. In other words, let $I_\mathcal{G} = \{j_0, j_1, ...\}$ be the set of indices that label the eigenstates spanning the subspace $\mathcal{G}_s$, i.e. $\mathcal{G}_s = \mathrm{span}\{|j_s\rangle\}_{j \in I_\mathcal{G}}$. Taking into account that $p_{j+1}^s \leq p_j^s$, a restoring unitary acting on $\mathcal{G}_s \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$ exists iff the chain $\{\mathcal{J}_{|\min_{j \in I_\mathcal{G}} j_s(k+1)_v\rangle \to |\max_{j \in I_\mathcal{G}} j_s k_v\rangle}\}_{l \leq k \leq l'}$ exists. In the case of the left subspace, $\mathcal{G}_s = \mathrm{span}\{|j_s\rangle\}_{1 \leq j \leq i}$, and the corresponding chain is $\{\mathcal{J}_{|1_s(k+1)_v\rangle \to |i_s k_v\rangle}\}_{l \leq k \leq l'}$. Since we are also assuming that the *right* chain $\{\mathcal{J}_{|(i+1)_s(k+1)_v\rangle \to |d_s k_v\rangle}\}_{l \leq k \leq l'}$ does not exist (i.e. that Eq. (A3) is also violated), there is no restoring unitary in the subspace $\mathrm{span}\{|j_s\rangle\}_{i+1 \leq j \leq d_s} \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$ either.

The preceding discussion implies that the only way to have a catalytic transformation is by means of a restoring unitary on a subspace $\mathrm{span}\{|j_s\rangle\}_{j \in I_\mathcal{G}} \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$, such that $\max_{j \in I_\mathcal{G}} \geq i+1$ and $\min_{j \in I_\mathcal{G}} \leq i$. Note that a subspace determined by the single condition $\max_{j \in I_\mathcal{G}} \geq i+1$ ($\min_{j \in I_\mathcal{G}} \leq i$) includes the possibility of a right (left) restoring unitary, which has already been discarded. If $\max_{j \in I_\mathcal{G}} \geq i+1$ and $\min_{j \in I_\mathcal{G}} \leq i$, at least one of the partial swaps composing the restoring unitary (cf. Eq. (8)) must connect an eigestate of $\rho_s \otimes \rho_v$ belonging to $\mathrm{span}\{|j_s\rangle\}_{1 \leq j \leq i} \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$, with one eigenstate belonging to $\mathrm{span}\{|j_s\rangle\}_{i+1 \leq j \leq d_s} \otimes \mathrm{span}\{|k_v\rangle\}_{l \leq k \leq l'}$. Otherwise, the restoring unitary could be defined exclusively in the left or in the right subspace. This partial swap has an associated restoring current that transfers population from the (system) subspace $\mathrm{span}\{|j_s\rangle\}_{1 \leq j \leq i}$ towards $\mathrm{span}\{|j_s\rangle\}_{i+1 \leq j \leq d_s}$. Let us denote this current simply as J. Since the cooling current $J_{|(i+1)_s l_v\rangle \to |i_s(l'+1)_v\rangle}$ transfers population in the opposite direction, we have that

$$\delta\left(\sum_{j=1}^i p_j^s\right) \leq -J + J_{|(i+1)_s l_v\rangle \to |i_s(l'+1)_v\rangle} = 0,$$

where the condition $J = J_{|(i+1)_s l_v\rangle \to |i_s(l'+1)_v\rangle}$ is necessary for the restoring unitary to generate a uniform loop. In this way, we conclude that the potential increment of the partial sum $\sum_{j=1}^i p_j^s$ is spoiled if we also demand that

the transformation is catalytic. Hence, the transformation must be unital.

Finally, we note that if $\rho_s = \rho_c \otimes \rho_h$, the substitutions (in Eqs. (A1)-(A3))

$$p_i^s \to p_{d_h}^h p_i^c,$$
$$p_{i+1}^s \to p_0^h p_{i+1}^c,$$
$$p_0^s \to p_0^h p_0^c,$$
$$p_{d_s}^s \to p_{d_h}^h p_{d_c}^c,$$

lead to Eqs. (19)-(21).

Appendix B: Population currents within the catalyst

The population variation for a general eigenstate $|i_c j_h k_v\rangle$ is generated by a two-level unitary that couples $|i_c j_h k_v\rangle$ to some other eigenstate. Therefore, the final population is given by $p_{ijk}^{chv} + J_{\to|i_c j_h k_v\rangle}$, if the population increases, or by $p_{ijk}^{chv} - J_{|i_c j_h k_v\rangle\to}$, if the population decreases.

For k fixed, the total probability to measure $|k_v\rangle$ is the sum of the probabilities to measure it given different values of i, j. That is,

$$\mathrm{Tr}(|k_v\rangle\langle k_v|U\rho U^\dagger) = \sum_{i,j\in\mathcal{L}_+}\left(p_{ijk}^{chv} + J_{\to|i_c j_h k_v\rangle}\right)$$
$$+ \sum_{i,j\in\mathcal{L}_-}\left(p_{ijk}^{chv} - J_{|i_c j_h k_v\rangle\to}\right),$$

where the set $\mathcal{L}_+(\mathcal{L}_-)$ contains the indices i, j such that the population p_{ijk}^{chv} increases (decreases). In this way,

$$\mathrm{Tr}[|k_v\rangle\langle k_v|(U\rho U^\dagger - \rho)] = \sum_{i,j\in\mathcal{L}_+} J_{\to|i_c j_h k_v\rangle}$$
$$- \sum_{i,j\in\mathcal{L}_-} J_{|i_c j_h k_v\rangle\to},$$

as indicated in Eqs. (22)-(24). Note that Eqs. (23) and (24) are written without referring to the sets $\mathcal{L}_\pm$. However, due to the direct sum structure of the unitaries U, a population p_{ijk}^{chv} increases iff there is current $J_{\to|i_c j_h k_v\rangle}$, and it decreases iff there is a current $J_{|i_c j_h k_v\rangle\to}$. This unambiguously determines that the sums (23) and (24) run over $\mathcal{L}_+$ and $\mathcal{L}_-$, respectively.

Appendix C: Proof of necessity for lemma 1

Suppose that the chain $\{\mathcal{J}_{|1_c 1_h (k+1)_v\rangle\to|d_c d_h k_v\rangle}\}_{l\leq k\leq l'}$ does not exist, which means that $\mathcal{J}_{|1_c 1_h (K+1)_v\rangle\to|d_c d_h K_v\rangle} = 0$ for some $l \leq K \leq l'$.

Let us see that this condition implies the non existence of a general chain $\mathbf{ch}_{|(l'+1)_v\rangle\to|l_v\rangle}$. First, note that

$$\mathcal{J}_{|1_c 1_h (K+1)_v\rangle\to|d_c d_h K_v\rangle} \geq \mathcal{J}_{|i_c j_h (K+1)_v\rangle\to|i'_c j'_h K_v\rangle}$$
$$\Leftrightarrow (p_1^c p_1^h)p_{K+1}^v - (p_{d_c}^c p_{d_h}^h)p_K^v \geq (p_i^c p_j^h)p_{K+1}^v - (p_{i'}^c p_{j'}^h)p_K^v,$$

where $1 \leq i, i' \leq d_c$ and $1 \leq j, j' \leq d_h$. This inequality follows directly from the inequalities $p_1^c p_1^h \geq p_i^c p_j^h$ and $p_{d_c}^c p_{d_h}^h \leq p_{i'}^c p_{j'}^h$. Similarly, for $k' \geq K + 1$ and $k'' \leq K$ it holds that

$$\mathcal{J}_{|i_c j_h (K+1)_v\rangle\to|i'_c j'_h K_v\rangle} \geq \mathcal{J}_{|i_c j_h k'_v\rangle\to|i'_c j'_h k''_v\rangle}$$
$$\Leftrightarrow p_i^c p_j^h(p_{K+1}^v) - p_{i'}^c p_{j'}^h(p_K^v) \geq p_i^c p_j^h(p_{k'}^v) - p_{i'}^c p_{j'}^h(p_{k''}^v),$$

which follows from the inequalities $p_{K+1}^v \geq p_{k'}^v$ and $p_K^v \leq p_{k''}^v$. In this way, we can join the previous inequalities to obtain:

$$\mathcal{J}_{|1_c 1_h (K+1)_v\rangle\to|d_c d_h K_v\rangle} \geq \mathcal{J}_{|i_c j_h k'_v \geq (K+1)_v\rangle\to|i'_c j'_h k''_v \leq K_v\rangle}.$$

Therefore, $\mathcal{J}_{|1_c 1_h (K+1)_v\rangle\to|d_c d_h K_v\rangle} = 0$ implies that $\mathcal{J}_{|i_c j_h k'_v\rangle\to|i'_c j'_h k''_v\rangle} = 0$ for $k' \geq K + 1$ and $k'' \leq K$ (and $1 \leq i, i' \leq d_c$ and $1 \leq j, j' \leq d_h$). The non existence (i.e. its nullity) of $\mathcal{J}_{|i_c j_h k'_v\rangle\to|i'_c j'_h k''_v\rangle}$ means that it is impossible to connect any pair catalyst eigenstates $|k'_v \geq (K+1)_v\rangle$ and $|k''_v \leq K_v\rangle$ using a restoring current. Since any chain of the form $\mathbf{ch}_{|(l'+1)_v\rangle\to|l_v\rangle}$ must contain this type of current, the non existence of $\mathcal{J}_{|i_c j_h k'_v\rangle\to|i'_c j'_h k''_v\rangle}$ also implies the non existence of $\mathbf{ch}_{|(l'+1)_v\rangle\to|l_v\rangle}$.

Appendix D: Restoring chains for a catalyst whose dimension complies with theorem 2, and corresponding catalyst eigenvalues

Restoring chains. Let $\{p_k^v\}_{1\leq k\leq d_v^*}$ be the eigenvalues of a full-rank catalyst state ρ_v, where $d_v = d_v^*$ is a dimension that satisfies the hypothesis of Theorem 2. If (Eq. (32) of the main text)

$$\frac{p_k^v}{p_{k+1}^v} = \mu_i \text{ for } \text{ for } 1 \leq k \leq d_v - 1,$$

with $\mu_i \equiv \max\left\{\frac{p_1^h p_1^c}{p_{d_h}^h p_i^c} - \epsilon, \frac{p_1^h p_{i+1}^c}{p_{d_h}^h p_{d_c}^c} - \epsilon\right\} > 1$ (ϵ positive and sufficiently small), we show now that Eq. (32) yields a restoring chain $\mathbf{ch}_{|d_v^*\rangle\to|1_v\rangle}$. This chain ensures the restoration of the catalyst given the cooling current $\mathcal{J}_{|(i+1)_c 1_h 1_v\rangle\to|i_c d_h d_v^*\rangle}$. Let us assume first that $\mu_i = \frac{p_1^h p_1^c}{p_{d_h}^h p_i^c} - \epsilon$. Equation (32) implies that for $1 \leq k \leq d_v^* - 1$.

$$\frac{p_1^h p_1^c}{p_{d_h}^h p_i^c} = \frac{p_k^v}{p_{k+1}^v} + \epsilon > \frac{p_k^v}{p_{k+1}^v}.$$

Accordingly, the chain $\mathbf{ch}_{|d_v^*\rangle\to|1_v\rangle}$ can be composed as $\mathbf{ch}_{|d_v^*\rangle\to|1_v\rangle} = \{\mathcal{J}_{|1_h i_c (k+1)_v\rangle\to|d_h (i+1)_c k_v\rangle}\}_k$. If $\mu_i = \frac{p_1^h p_{i+1}^c}{p_{d_h}^h p_{d_c}^c} - \epsilon$, it is straightforward to check

that $\frac{p_1^h p_{i+1}^c}{p_{d_h}^h p_{d_c}^c} > \frac{p_k^v}{p_{k+1}^v}$, which provides the chain $\{\mathcal{J}_{|1_h(i+1)_c(k+1)_v\rangle \to |d_h d_c k_v\rangle}\}_k$.

On the other hand, for a non fully mixed hot object (condition 1 of Theorem 2) we can also define $\bar\mu \equiv \frac{p_1^h}{p_{d_h}^h} - \epsilon > 1$. If $\frac{p_k^v}{p_{k+1}^v} = \bar\mu$ for for $1 \leq k \leq d_v - 1$, then the minimum dimension $d_v = \bar d_v$ such that

$$\bar\mu^{\bar d_v - 1} = \frac{p_1^v}{p_{\bar d_v}^v} > \frac{p_i^c p_{d_h}^h}{p_{i+1}^c p_1^h}$$

gives rise to the cooling current $\mathcal{J}_{|(i+1)_c 1_h 1_v\rangle \to |i_c d_h \bar d_v\rangle}$. Similarly to the restoring chains deduced from Eq. (32), the condition $\frac{p_k^v}{p_{k+1}^v} = \bar\mu$ guarantees a restoring chain $\{\mathcal{J}_{|1_h(k+1)_v\rangle \to |d_h k_v\rangle}\}_{1 \leq k \leq \bar d_v - 1}$. The corresponding restoring unitary is of the form V_{hv} (cf. Eq. (31)), with $l = 1$ and $l' = \bar d_v - 1$.

Catalyst eigenvalues. The eigenvalues $\{p_k^v\}$ are straightforwardly derived from Eq. (32) or its analogous $\frac{p_k^v}{p_{k+1}^v} = \bar\mu$. Specifically,

$$\frac{p_1^v}{p_k^v} = \prod_{i=1}^{k-1} \frac{p_i^v}{p_{i+1}^v} = t^{k-1} \Rightarrow p_k^v = p_1^v t^{k-1},$$

where $t = \mu_i$ or $t = \bar\mu$. From this relation and the normalization condition $\sum_k p_k^v = 1$ the catalyst eigenvalues are completely characterized.

Appendix E: Optimal catalyst state for the cooling of a qubit using another qubit as hot object

Proof of optimality of the eigenvalues that satisfy Eq. (38). Before deriving the optimal eigenvalues $\{p_k^v\}_{1 \leq k \leq n}$, let us show why the condition

$$\mathcal{J}_k = \mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}, \ \forall 1 \leq k \leq n-1, \quad (E1)$$

leads to the maximization of $J_{\rm cool}$. To this aim we prove that if $\{p_k^v\}$ satisfies this equation, then any other set of eigenvalues $\{q_k^v\} \neq \{p_k^v\}$ is suboptimal. Note that we consider states of fixed rank n, and therefore $p_k^v, q_k^v \neq 0$ for $k \leq n$, while $p_k^v = q_k^v = 0$ for $k \geq n+1$. Let $\mathcal{J}_k'$ and $\mathcal{J}'_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$ denote the counterparts of the currents $\mathcal{J}_k$ and $\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$, corresponding to the eigenvalues $\{q_k^v\}$. That is, the expressions for the primed currents are obtained by simply replacing p_k^v by q_k^v in the expressions for the non-primed currents

$$\mathcal{J}_k = p_1^h p_{k+1}^v - p_2^h p_k^v, \quad (E2)$$

$$\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle} = p_2^c p_1^h p_1^v - p_1^c p_2^h p_n^v. \quad (E3)$$

Moreover, let $J_{\rm cool}^{\max}$ and $(J_{\rm cool}^{\max})'$ denote respectively the maximum cooling currents achieved with $\{p_k^v\}$ and $\{q_k^v\}$.

These currents are given by the minimum swap currents in the corresponding loops:

$$J_{\rm cool}^{\max} = \mathcal{J}_{\rm Loop}^{\min}$$
$$= \min\left\{\min_k \mathcal{J}_k, \mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}\right\}, \quad (E4)$$
$$(J_{\rm cool}^{\max})' = (\mathcal{J}_{\rm Loop}^{\min})'$$
$$= \min\left\{\min_k \mathcal{J}'_k, \mathcal{J}'_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}\right\}. \quad (E5)$$

In other words, the maximum cooling current must satisfy these equations to form a *uniform loop* with the restoring chain.

To prove that $(J_{\rm cool}^{\max})' \leq J_{\rm cool}^{\max}$, we show that any transformation $\{p_k^v\} \to \{q_k^v\}$ must be such that $(\mathcal{J}_{\rm Loop}^{\min})' \leq \mathcal{J}_{\rm Loop}^{\min}$. An arbitrary transformation $\{p_k^v\} \to \{q_k^v\}$ can be described by the equations

$$q_k^v = p_k^v + \delta_k, \quad (E6)$$

with the values of δ_k restricted so that $\{q_k^v\}$ represents a probability distribution. In particular, probability conservation implies that $\sum_{k=1}^n \delta_k = 0$. The population variations $\delta_k = q_k^v - p_k^v$ and $\delta_{k+1} = q_{k+1}^v - p_{k+1}^v$ in turn induce a change in the restoring current $\mathcal{J}_k$, denoted as $\delta\mathcal{J}_k$. Likewise, since $\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$ depends on p_1^v and p_n^v, the variations δ_1 and δ_n change this current by an amount $\delta\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$.

Taking into account condition (E1) and Eq. (E4), the only way to increase $J_{\rm cool}^{\max}$ is to simultaneously increase *all* the swap currents in the loop. Keeping in mind Eq. (E6),

$$\delta\mathcal{J}_k = \mathcal{J}'_k - \mathcal{J}_k$$
$$= p_1^h \delta_{k+1} - p_2^h \delta_k, \quad (E7)$$
$$\delta\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle} = (p_2^c p_1^h)\delta_1 - (p_1^c p_2^h)\delta_n. \quad (E8)$$

Accordingly, $\delta\mathcal{J}_k > 0$ implies that $\delta_k < 0$ or $\delta_{k+1} > 0$, and $\delta\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle} > 0$ implies that $\delta_1 > 0$ or $\delta_n < 0$. Let us see that these conditions cannot be both satisfied.

- If $\delta_k < 0$ for some k, $\delta\mathcal{J}_{k-1} = p_1^h \delta_k - p_2^h \delta_{k-1}$ (cf. Eq. (E7)) can be positive only if $\delta_{k-1} < 0$. Therefore, a simple generalization by induction leads us to conclude that $\delta\mathcal{J}_{l \leq k} \geq 0$ only if $\delta_l < 0$ for all $l \leq k-1$. In particular, $\delta_1 < 0$. On the other hand, from probability conservation it follows that if $\delta_l < 0$ for all $l \leq k$ then $\delta_m > 0$ for some $m \geq k+1$. This in turn implies that $\delta\mathcal{J}_m = p_1^h \delta_{m+1} - p_2^h \delta_m > 0$ only if $\delta_{m+1} > 0$, and by induction that $\delta\mathcal{J}_{l \geq m} > 0$ only if $\delta_l > 0$ for all $l \geq m+1$. In this way we conclude that $\delta_n > 0$ and $\delta_1 < 0$, and following Eq. (E8) that $\delta\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle} < 0$. Therefore, it is not possible to have $\delta\mathcal{J}_k$ positive for all $1 \leq k \leq n-1$ and also $\delta\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$ positive, assuming that $\delta_k < 0$ for some k. This leaves us with the only option $\delta_{k+1} > 0$ to have $\delta\mathcal{J}_k > 0$, which we address below.

- If $\delta_{k+1} > 0$ for some k, we can apply a completely analogous reasoning. In this case $\delta_l > 0$ for all $l \geq k+2$ is necessary for the positivity of $\delta\mathcal{J}_{l \geq k+1}$, which implies that $\delta_n > 0$. Moreover, from the condition $\delta_m < 0$ for some $m \leq k$ (derived from probability conservation) it follows that $\delta\mathcal{J}_{l \leq m-1} > 0$ only if $\delta_l < 0$ for $l \leq m-1$. Once again, we conclude that $\delta_1 < 0$ and $\delta_n > 0$, and consequently that $\delta\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle} < 0$.

Derivation of the optimal eigenvalues. The optimal eigenvalues $\{p_k^v\}$ are obtained by using Eqs. (E2) and (E3) in the following way. By defining the quantity $r_h \equiv \frac{p_2^h}{p_1^h}$, it is straightforward to check the identity

$$\mathcal{J}_{n-1} + r_h \mathcal{J}_{n-2} = p_1^h p_n^v - \frac{\left(p_2^h\right)^2}{p_1^h} p_{n-2}^v. \qquad \text{(E9)}$$

Here, the effect of the factor r_h is to cancel the second term in the current $\mathcal{J}_{n-1}$ (given by $p_1^h p_{n-1}^v$) with the first term of $\mathcal{J}_{n-2}$ (given by $p_1^h p_{n-1}^v$). More generally, the identity $r_h^i p_2^h = r_h^{i+1} p_1^h$ guarantees that in the sum $\sum_{i=0}^{k-1} r_h^i \mathcal{J}_{n-(i+1)}$ only the first and last terms survive. That is,

$$\sum_{i=0}^{k-1} r_h^i \mathcal{J}_{n-(i+1)} = p_1^h p_n^v - r_h^{k-1} p_2^h p_{n-k}^v. \qquad \text{(E10)}$$

On the other hand, from Eq. (E1) it also holds that (by application of the geometric series)

$$\sum_{i=0}^{k-1} r_h^i \mathcal{J}_{n-(i+1)} = \left(\sum_{i=0}^{k-1} r_h^i\right) \mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$$

$$= \left(\frac{1-r_h^k}{1-r_h}\right) \mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}. \qquad \text{(E11)}$$

By equating Eqs. (E10) and (E11) we obtain:

$$p_{n-k}^v = \frac{r_h^{1-k}}{p_2^h}\left[p_1^h - \left(\frac{1-r_h^k}{1-r_h}\right)\frac{\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}}{p_n^v}\right]p_n^v. \qquad \text{(E12)}$$

Given the dependence of $\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$ on p_1^v, this equation provides the eigenvalues p_{n-k}^v, for $1 \leq k \leq n-1$, in terms of p_n^v and p_1^v. To eliminate the dependence on p_1^v we substitute $k = n-1$ and isolate p_1^v, which yields

$$p_1^v = \frac{\left(1-r_h^{n-1}\right)r_h p_1^c + (1-r_h)}{\left(1-r_h^{n-1}\right)p_1^c + (1-r_h)r_h^{n-1}}p_n^v. \qquad \text{(E13)}$$

After inserting this back into Eq. (E12) we can apply the normalization condition $\sum_{k=1}^{n-1} p_{n-k}^v + p_n^v = 1$ to derive p_n^v. In this way, we obtain a complete characterization of all the eigenvalues $\{p_k^v\}_{1 \leq k \leq n}$, in terms of the eigenvalues $\{p_i^c\}$ and $\{p_i^h\}$ of the cold and hot objects. Specifically the normalization condition yields

$$p_n^v = \frac{p_1^h r_h^{n-1}(1-r_h)}{p_1^h(1-r_h^n) + \left[nr_h^{n-1}(1-r_h) - (1-r_h^n)\right]\frac{J_{\text{cool}}^{\max}}{(1-r_h)p_n^v}}, \qquad \text{(E14)}$$

where $J_{\text{cool}}^{\max}$ corresponds to Eq. (39) in the main text. Note that $J_{\text{cool}}^{\max}/p_n^v$ depends only on $\{p_i^c\}$ and $\{p_i^h\}$, and therefore the r.h.s. in the previous equation only depends on these eigenvalues. In addition, Eq. (39) is obtained by replacing p_1^v and p_n^v into $\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$ (Eq. (E3)) by the expressions given in Eqs. (E13) and (E14). This recasting of $\mathcal{J}_{|2_c 1_h 1_v\rangle \to |1_c 2_h n_v\rangle}$ as $J_{\text{cool}}^{\max}$ in turn gives rise to Eq. (40).

Optimal CC unitary. The optimal unitary that yields the current (E4) is composed of swaps between the states connected by the cooling current and by the restoring currents. The reason is that, as already proven, $J_{\text{cool}}^{\max}$ is maximized through a uniform loop formed by swap currents (cf. Eq. (E1)). Besides the swap for the cooling current, which exchanges the states $|2_c 1_h 1_v\rangle$ and $|1_c 2_h n_v\rangle$, the swaps that give rise to the restoring currents (E2) exchange the states $|1_h(k+1)_v\rangle$ and $|2_h k_v\rangle$.

Appendix F: Proof of theorem 3

Lemma 2 (optimal cooling of a cold qubit using only the hot object). Let $\left\{\left(p_j^h\right)^\downarrow\right\}_{1 \leq j \leq \mathfrak{J}}$ be the $\mathfrak{J}$ largest eigenvalues of ρ_h, in non-increasing order, where $\mathfrak{J} = \frac{d_h}{2}$ if d_h is even and $\mathfrak{J} = \frac{d_h-1}{2}$ if d_h is odd. Moreover, let $\{|j_h\rangle_\downarrow\}_{1 \leq j \leq \mathfrak{J}}$ denote the corresponding eigenstates. Likewise, let $\left\{\left(p_j^h\right)^\uparrow\right\}_{1 \leq j \leq \mathfrak{J}}$ be the $\mathfrak{J}$ lowest eigenvalues of ρ_h, in non-decreasing order, with $\{|j_h\rangle_\uparrow\}_{1 \leq j \leq \mathfrak{J}}$ being the corresponding eigenstates. The optimal cooling of a qubit is obtained through a unitary (permutation) $\tilde{U}_{ch}$ that satisfies:

$$\tilde{U}_{ch}|2_c j_h\rangle_\downarrow = |1_c j_h\rangle_\uparrow, \qquad \text{(F1)}$$

$$\tilde{U}_{ch}|1_c j_h\rangle_\uparrow = |2_c j_h\rangle_\downarrow, \qquad \text{(F2)}$$

for all j such that $p_2^c\left(p_j^h\right)^\downarrow > p_1^c\left(p_j^h\right)^\uparrow$, and $\tilde{U}_{ch}|2_c j_h\rangle_\downarrow = |2_c j_h\rangle_\downarrow$, $\tilde{U}_{ch}|1_c j_h\rangle_\uparrow = |1_c j_h\rangle_\uparrow$, for all j such that $p_2^c\left(p_j^h\right)^\downarrow \leq p_1^c\left(p_j^h\right)^\uparrow$.

Proof of Lemma 2. Let us denote as $A_> \equiv \left\{p_2^c\left(p_j^h\right)^\downarrow\right\}$ and $A_< \equiv \left\{p_1^c\left(p_j^h\right)^\uparrow\right\}$ the subsets of eigenvalues of ρ_{ch} that satisfy $p_2^c\left(p_j^h\right)^\downarrow > p_1^c\left(p_j^h\right)^\uparrow$. In other words, $A_>$ and $A_<$ contain the eigenvalues characterizing eigenstates of ρ_{ch} on which $\tilde{U}_{ch}$ acts non-trivially. Note that, by construction, $A_> \subseteq \left\{p_2^c\left(p_j^h\right)^\downarrow\right\}_{1 \leq j \leq \mathfrak{J}}$ and $A_< \subseteq \left\{p_1^c\left(p_j^h\right)^\uparrow\right\}_{1 \leq j \leq \mathfrak{J}}$. The eigenstates of ρ_{ch} that possess eigenvalue ε_1^c with respect to H_c have eigenvalues $\{p_1^c p_j^h\} = A_< \cup A_<^c$, where $A_<^c = \{a \in \{p_1^c p_j^h\} \text{ s.t. } a \notin A_<\}$ is the complement of $A_<$. Similarly, the eigenstates with eigenvalue ε_2^c with respect to H_c have eigenvalues $\{p_2^c p_j^h\} = A_> \cup A_>^c$ with respect to ρ_{ch}, where $A_>^c = \{a \in \{p_2^c p_j^h\} \text{ s.t. } a \notin A_>\}$. The unitary $\tilde{U}_{ch}$ is a permutation that exchanges the eigenvalues $A_>$ and $A_<$,

whence the final eigenvalues corresponding to eigenstates $\{|1_c j_h\rangle\}_j$ and $\{|2_c j_h\rangle\}_j$ are respectively given by $A_> \cup A_<^c$ and $A_< \cup A_>^c$. This is illustrated in Fig. 16.

Keeping in mind that the final state $\rho'_{ch} = \tilde{U}_{ch}(\rho_{ch})\tilde{U}_{ch}^\dagger$ commutes with H_c, this state is also passive with respect to H_c if $\max(a \in A_< \cup A_<^c) \leq \min(a \in A_> \cup A_>^c)$. If this is the case the mean energy of the cold object cannot be further reduced and $\tilde{U}_{ch}$ performs optimal cooling. This inequality holds iff

$$\max(a \in A_<) \leq \min(a \in A_>), \min(a \in A_<^c),$$
$$\max(a \in A_>^c) \leq \min(a \in A_>), \min(a \in A_<^c).$$

The inequality $\max(a \in A_<) \leq \min(a \in A_<^c)$ holds because $A_<$ is a subset of the *smallest* elements of $A_< \cup A_<^c$, and $\max(a \in A_>^c) \leq \min(a \in A_>)$ holds because $A_>$ is a subset of the *largest* elements of $A_> \cup A_>^c$. Moreover, we note that the maximum index $j = j_{\max}$ in the sets $A_<$ and $A_>$ determines the corresponding maximum and minimum elements, and that by definition $p_1^c \left(p_{j_{\max}}^h\right)^\uparrow < p_2^c \left(p_{j_{\max}}^h\right)^\downarrow$. Therefore, $\max(a \in A_<) \leq \min(a \in A_>)$.

This leaves us with the verification of the inequality $\max(a \in A_>^c) \leq \min(a \in A_<^c)$. Noting that we can write $\max(a \in A_>^c) = p_2^c \left(p_{j_{\max}+1}^h\right)^\downarrow$ and $\min(a \in A_<^c) = p_1^c \left(p_{j_{\max}+1}^h\right)^\uparrow$, the assumption $\max(a \in A_>^c) > \min(a \in A_<^c)$ is contradictory because it would imply that $\max(a \in A_>^c)$ and $\min(a \in A_<^c)$) also belong to $A_>$ and $A_<$, respectively. It thus follows that $\max(a \in A_>^c) \leq \min(a \in A_<^c)$, which completes the proof of the inequality $\max(a \in A_< \cup A_<^c) \leq \min(a \in A_> \cup A_>^c)$ and the passivity of ρ'_{ch} with respect to H_c.

Proof of Theorem 3. We construct a CC transformation using a global unitary U' that acts on the eigenstates of ρ_{ch} whose eigenvalues belong to the sets $A_>^c$ and $A_<^c$, on which $\tilde{U}_{ch}$ acts as the identity (see Fig. 16). Let us denote the eigenstates of ρ_{ch} with eigenvalues in $A_>^c$ as $\{|2_c(j_>)_h\rangle\}$, and the eigenstates with eigenvalues in $A_<^c$ as $\{|1_c j_{h,<}\rangle\}$. Using a catalyst in the initial state ρ_v, the unitary U' implements a CC transformation by mapping the subspace $\text{span}(\{|2_c j_{h,>}\rangle\} \cup \{|1_c j_{h,<}\rangle\}) \otimes \mathcal{H}_v$ into itself, as shown in Fig. 16.

Consider a full-rank state ρ_v whose eigenvalues satisfy

$$\frac{p_k^v}{p_{k+1}^v} = \mu \equiv \max\left\{\frac{\max(a \in A_<^c)}{\min(a \in A_<^c)} - \epsilon, \frac{\max(a \in A_>^c)}{\min(a \in A_>^c)} - \epsilon\right\},$$
(F3)

for $1 \leq k \leq d_v - 1$. By definition, the set $A_>^c$ includes the *smallest* $\frac{d_h}{2}$ $\left(\frac{d_h+1}{2}\right)$ elements of $\{p_2^c p_j^h\}_{1 \leq j \leq d_h}$ if d_h is even (odd). Similarly, the set $A_<^c$ includes the *largest* $\frac{d_h}{2}$ $\left(\frac{d_h+1}{2}\right)$ elements of $\{p_1^c p_j^h\}_{1 \leq j \leq d_h}$ if d_h is even (odd). See Fig. 16 to visualize these properties. Accordingly, from the hypothesis of Theorem 3 it follows that $\max(a \in A_>^c) \neq \min(a \in A_>^c)$, or $\max(a \in A_<^c) \neq \min(a \in A_<^c)$. This implies that $\mu > 1$, for ϵ small enough. From Eq.
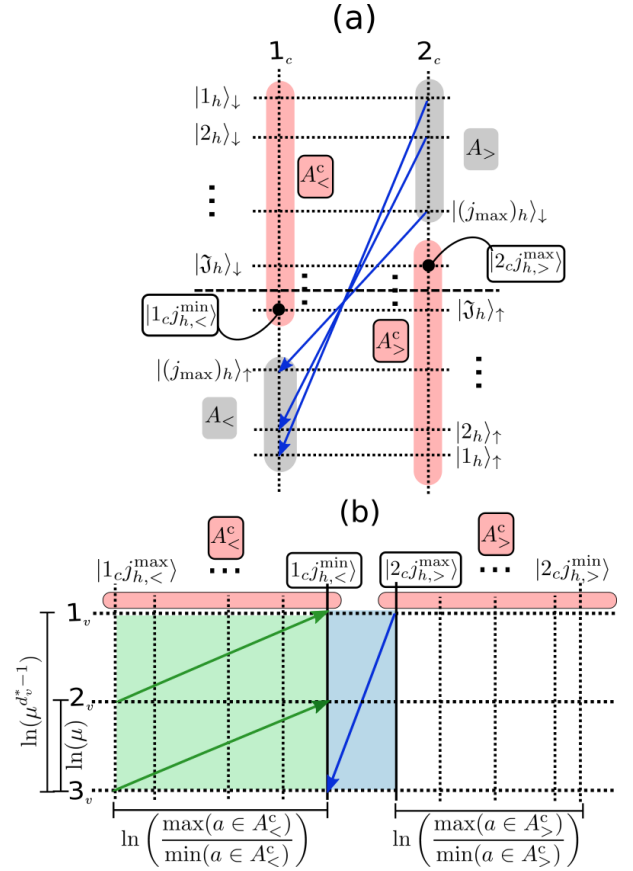

Figure 16. (a) $\ln(p^c) \times \ln(p^h)$ diagram for a unitary $\tilde{U}_{ch}$ that performs optimal cooling of a cold qubit. In this case we assume that d_h is even and hence $\mathfrak{J} = d_h/2$. The dashed line separates the eigenstates $|j_h\rangle_\downarrow$ associated with the largest $\mathfrak{J}$ eigenvalues of ρ_h, from the eigenstates $|j_h\rangle_\uparrow$, associated with the $\mathfrak{J}$ smallest eigenvalues. The index $j_{\max}$ is the maximum j such that $p_1^c\left(p_j^h\right)^\uparrow \leq p_2^c\left(p_j^h\right)^\downarrow \Leftrightarrow \max(a \in A_<) \leq \min(a \in A_>)$. The unitary $\tilde{U}_{ch}$ permutes the eigenstates of $\rho_c \otimes \rho_h$ corresponding to $A_>$ with those corresponding to $A_<$, generating the depicted cooling currents. (b) $\ln(p^{ch}) \times \ln(p^v)$ diagram for the CC transformation implemented after $\tilde{U}_{ch}$. Here, we see that $d_v^* = 3$ is enough for the transformation, meaning that the total height $\ln(\mu^{d_v^*-1}) = 2\ln(\mu)$ allows to form a vertical (cyan) rectangle with the columns $|1_c j_{h,<}^{\min}\rangle$ and $|2_c j_{h,>}^{\max}\rangle$ (which contains the cooling current). The restoring chain is guaranteed by Eq. (F3).

(F3), $\frac{p_1^v}{p_{d_v}^v} = \prod_{k=1}^{d_v-1} \frac{p_k^v}{p_{k+1}^v} = \mu^{d_v-1}$, and consequently

$$\frac{p_1^v}{p_{d_v}^v} = \mu^{d_v^*-1} > \frac{\min(a \in A_<^c)}{\max(a \in A_>^c)} > 1$$

for $d_v = d_v^*$ sufficiently large. This guarantees a cooling current $\mathcal{J}_{|2_c j_{h,>}^{\max}\rangle \otimes |1_v\rangle \to |1_c j_{h,<}^{\min}\rangle \otimes |d_v\rangle}$, where $|2_c j_{h,>}^{\max}\rangle$ is an eigenstate with eigenvalue $\max(a \in A_>^c)$, and $|1_c j_{h,<}^{\min}\rangle$ is an eigenstate with eigenvalue $\min(a \in A_<^c)$.

Suppose first that $\mu = \frac{\max(a \in A_<^c)}{\min(a \in A_<^c)} - \epsilon$ or, equivalently, $\frac{\max(a \in A_<^c)}{\min(a \in A_<^c)} = \frac{p_k^v}{p_{k+1}^v} + \epsilon > \frac{p_k^v}{p_{k+1}^v}$. In this way, the inequalities

$\frac{\max(a\in A^c_<)}{\min(a\in A^c_<)} > \frac{p^v_k}{p^v_{k+1}}$ give rise to a "left" restoring chain

$$\{\mathcal{J}_{|1_c j^{\max}_{h,<}\rangle\otimes|(k+1)_v\rangle\to|1_c j^{\min}_{h,<}\rangle\otimes|k_v\rangle}\}_{1\le k\le d_v-1},$$

where $|1_c j^{\max}_{h,<}\rangle$ is an eigenstate with eigenvalue $\max(a\in A^c_<) = p^c_1 p^h_1$.

If $\mu = \frac{\max(a\in A^c_>)}{\min(a\in A^c_>)} - \epsilon$, an analogous argument leads to deduce the "right" restoring chain

$$\{\mathcal{J}_{|2_c j^{\max}_{h,>}\rangle\otimes|(k+1)_v\rangle\to|2_c j^{\min}_{h,>}\rangle\otimes|k_v\rangle}\}_{1\le k\le d_v-1},$$

where $|2_c j^{\min}_{h,>}\rangle$ is an eigenstate of ρ_{ch} with eigenvalue $\min(a\in A^c_>) = p^c_2 p^h_{d_h}$.

The parameter μ and the condition $\mu^{d^*_v-1} > \frac{\min(a\in A^c_>)}{\max(a\in A^c_>)}$ can be recast as they appear in the statement of the theorem, by defining $p^h_{j_1} \equiv \left(p^h_{j_{\max}+1}\right)^\uparrow$ and $p^h_{j_2} \equiv \left(p^h_{j_{\max}+1}\right)^\downarrow$. In this way, the substitution of $\frac{\max(a\in A^c_<)}{\min(a\in A^c_<)} = \frac{p^h_1}{p^h_{j_1}}$ and $\frac{\max(a\in A^c_>)}{\min(a\in A^c_>)} = \frac{p^h_{j_2}}{p^h_{d_h}}$ in Eq. (F3) yields $\mu = \max\left\{\frac{p^h_1}{p^h_{j_1}} - \epsilon, \frac{p^h_{j_2}}{p^h_{d_h}} - \epsilon\right\}$. Moreover, from $\frac{\min(a\in A^c_>)}{\max(a\in A^c_>)} = \frac{p^c_1 p^h_{j_1}}{p^c_2 p^h_{j_2}}$ we obtain $\mu^{d^*_v-1} > \frac{p^c_1 p^h_{j_1}}{p^c_2 p^h_{j_2}}$. After taking the natural logarithm of this inequality we obtain the inequality for d^*_v in the theorem statement.

Appendix G: Maximum cooling of a qubit using k hot qubits

The optimal cooling of a single qubit using k hot qubits can be obtained by applying Lemma 2. In this case, the k hot qubits constitute a hot environment of dimension $d_h = 2^k$ even. The environment state is given by $\rho_H = \rho_h^{\otimes k}$, where $\rho_h = p^h_1|1_h\rangle\langle 1_h| + p^h_2|2_h\rangle\langle 2_h|$ is the state of a single hot qubit. The eigenstates of ρ_H that describe $l \le k$ excited qubits and $k-l$ qubits in the ground state possess the eigenvalue $(p^h_1)^{k-l}(p^h_2)^l$, and degeneracy $\frac{k!}{l!(k-l)!}$. Moreover, from $p^h_1 > p^h_2$ it readily follows that $(p^h_1)^{k-l}(p^h_2)^l > (p^h_1)^{k-(l+1)}(p^h_2)^{l+1}$. This implies that the largest eigenvalues of ρ_H correspond to $l \le k/2$.

From Lemma 2, the optimal unitary $\tilde{U}_{cH}$ is composed of swaps between eigenstates $|2_c \boldsymbol{j}_H\rangle_\downarrow$ and $|1_c \boldsymbol{j}_H\rangle_\uparrow$, where $|\boldsymbol{j}_H\rangle_\downarrow$ ($|\boldsymbol{j}_H\rangle_\uparrow$) is an eigenstate of ρ_H with $l \le k/2$ excited (non excited) hot qubits. Each of these swaps generates a cooling current

$$J^{(l)}_{\mathrm{cool}} \equiv p^c_2[(p^h_1)^{k-l}(p^h_2)^l] - p^c_1[(p^h_1)^l(p^h_2)^{k-l}]$$
$$= (p^c_1)^{k-l}(p^c_2)^{l+1} - (p^c_1)^{l+1}(p^c_2)^{k-l},$$

where in the second line we use the condition $p^c_i = p^h_i$ (the cold qubit is identical to the hot qubits). The maximum current is achieved for $l = 0$ and $J^{(l)}_{\mathrm{cool}}$ is non-increasing with respect to l.

For k even, choosing $l = k/2$ yields $J^{(k/2)}_{\mathrm{cool}} = (p^c_1 p^c_2)^{k/2}[p^c_2 - p^c_1] < 0$. On the other hand, if $l = k/2 - 1$ we have that $J^{(k/2-1)}_{\mathrm{cool}} = (p^c_1 p^c_2)^{k/2}[p^c_1 - p^c_2] > 0$. This means that the minimum (positive) current corresponds to $l = k/2 - 1$. On the other hand, for a given value of l there are $\frac{k!}{l!(k-l)!}$ swaps that exchange all the pairs of eigenstates with l excited qubits and l non excited qubits. This implies that the total ground population increment generated by $\tilde{U}_{cH}$ is

$$\Delta p^c_1 = \sum_{l=0}^{k/2-1} \frac{k!}{l!(k-l)!} J^{(l)}_{\mathrm{cool}} \text{ for } k \text{ even.}$$

For k odd, choosing $l = (k-1)/2$ yields $J^{((k-1)/2)}_{\mathrm{cool}} = 0$, and choosing $l = (k-3)/2$ yields $J^{((k-3)/2)}_{\mathrm{cool}} = (p^c_1 p^c_2)^{(k-1)/2}[(p^c_1)^2 - (p^c_2)^2] > 0$. Accordingly, the total population increment in this case reads

$$\Delta p^c_1 = \sum_{l=0}^{(k-3)/2} \frac{k!}{l!(k-l)!} J^{(l)}_{\mathrm{cool}} \text{ for } k \text{ odd.}$$

These expressions are employed to plot the cooling coefficient $\xi^{(k)}_{\mathrm{cool}} = \frac{\Delta p^c_1}{k}$ in Fig. 10 of the main text. In the case $k = 2$, it is straightforward to check that

$$\xi^{(2)}_{\mathrm{cool}} = \frac{J^{(0)}_{\mathrm{cool}}}{2} = \frac{(p^c_1)^2(p^c_2)^1 - (p^c_1)^1(p^c_2)^2}{2}$$
$$= \frac{(1 - 2p^c_2)}{2}p^c_1 p^c_2,$$

which coincides with Eq. (48) of the main text.

Appendix H: Optimization of CNU and CC transformations

In this Appendix we present a protocol to systematically enhance CNU and CC transformations. After n steps, the protocol provides a pair $[\rho_v(n), U_n]$ consisting of a catalyst state $\rho_v(n)$ and a global unitary U_n that implements a catalytic transformation

$$\rho(n) \to \rho'(n) = U_n \rho(n) U^\dagger_n,$$

where $\rho(n) \equiv \rho_c \otimes \rho_h \otimes \rho_v(n)$. We remark that $\rho(n)$ is not to be interpreted as a transformed state. Instead, it represents the *initial* state that should be employed according to the nth step of the protocol. Since the cold and hot objects are characterized by a fixed initial state $\rho_c \otimes \rho_h$, the dependence on n is completely contained in $\rho_v(n)$. Following the same approach of the main text, we consider transformations $\rho(n) \to \rho'(n)$ that are non-unital with respect to the cold object. Hence, if the final state $\rho'_c(n) \equiv \mathrm{Tr}_{hv}[\rho'(n)]$ has populations (in the eigenbasis of ρ_c) denoted as $\{p'^{c,n}_l\}_{1\le l\le d_c}$, the non-unitality condition (10) reads $\sum_{l=1}^i p'^{c,n}_l - \sum_{l=1}^i p^c_l > 0$. For a fixed value

of i, the protocol yields transformations $\rho(n) \to \rho'(n)$ such that this difference is monotonically increasing (or at least non-decreasing) with respect to n. This "sequential optimization" can also be applied to catalytic and cooling transformations, as specified later.

For the sake of simplicity we characterize the transformations in terms of loops, instead of unitaries U_n. Keeping in mind that for a loop each catalyst eigenstate has an incoming current and an outgoing current (cf. Definition 3 of the main text), it is always possible to find a unitary $U = U_{\text{cool}} \oplus V_{\text{res}}$ that leaves the catalyst unchanged, by adjusting the corresponding partial swaps so as to generate a uniform loop. The nth step of the protocol generates a loop of the form $\textbf{loop}_n \equiv \textbf{ch}^{(n)}_{|1_v\rangle \to |d_v\rangle} \cup \textbf{ch}^{(n)}_{|d_v\rangle \to |1_v\rangle}$, where $\textbf{ch}^{(n)}_{|d_v\rangle \to |1_v\rangle}$ is a restoring chain (cf. Definition 5 of the main text), and $\textbf{ch}^{(n)}_{|1_v\rangle \to |d_v\rangle}$ is the chain that contains the *total* cooling current $J_{\text{cool}}(n)$. This current quantifies the non-unital effect for the transformation $\rho(n) \to \rho'(n)$, i.e. $J_{\text{cool}}(n) = \sum_{l=1}^{i} p_l'^{c,n} - \sum_{l=1}^{i} p_l^c$. In addition, $J_{\text{cool}}(n)$ may be the sum of *several* individual cooling currents, and consequently the transformations $\rho(n) \to \rho'(n)$ are more general than those analyzed in the main text. Using this notation we can reformulate the essential purpose of the protocol. That is, to sequentially generate transformations $\rho(n) \to \rho'(n)$ such that $J_{\text{cool}}(n+1) \geq J_{\text{cool}}(n)$.

The cold and hot objects are characterized by states ρ_c and ρ_h that adhere to any of the conditions of Theorem 2. In addition, the initial catalyst state $\rho_v(0)$ is chosen such that $\frac{p_k^v}{p_{k+1}^v} = \mu_i$ for $1 \leq k \leq d_v - 1$, with μ_i defined in Theorem 2. This guarantees the existence of a loop $\textbf{loop}_0 = \{\mathcal{J}_0\} \cup \{\mathcal{J}_k\}_{1 \leq k \leq d_v - 1}$, with a cooling current $\mathcal{J}_0 \equiv \mathcal{J}_{|(i+1)_c 1_h 1_v\rangle \to |i_c d_h d_v\rangle}$ and the restoring chain $\{\mathcal{J}_k\}_{1 \leq k \leq d_v - 1}$ associated with μ_i (indicated in the proof of Theorem 2).

1. Protocol for optimization of CNU transformations

Starting from the loop $\textbf{loop}_0$ and fixing the corresponding catalyst dimension, new loops $\{\textbf{loop}_n\}_{n \geq 1}$ are systematically produced by the following protocol:

1. **Balance of loop currents**: The first loop $\textbf{loop}_1 \equiv \{\mathcal{J}_0(1)\} \cup \{\mathcal{J}_k(1)\}_{1 \leq k \leq d_v - 1}$ corresponds to a catalyst state $\rho_v(1) = \sum_{k=1}^{d_v} p_k^{v,1} |k_v\rangle\langle k_v|$ that is the solution to the equations

$$\mathcal{J}_k(1) = \mathcal{J}_{k+1}(1) \text{ for } 0 \leq k \leq d_v - 1, \qquad \text{(H1)}$$

where $\mathcal{J}_0(1) = \left(p_{i+1}^c p_1^h\right) p_1^{v,1} - \left(p_i^c p_{d_h}^h\right) p_{d_v}^{v,1}$, and
$\mathcal{J}_{k \geq 1}(1) = p_1^c p_1^h p_{k+1}^{v,1} - p_i^c p_{d_h}^h p_k^{v,1}$ if $\mu_i = \frac{p_1^h p_1^c}{p_{d_h}^h p_i^c} - \epsilon$, or
$\mathcal{J}_{k \geq 1}(1) = p_{i+1}^c p_1^h p_{k+1}^{v,1} - p_{d_c}^c p_{d_h}^h p_k^{v,1}$ if $\mu_i = \frac{p_1^h p_{i+1}^c}{p_{d_h}^h p_{d_c}^c} - \epsilon$
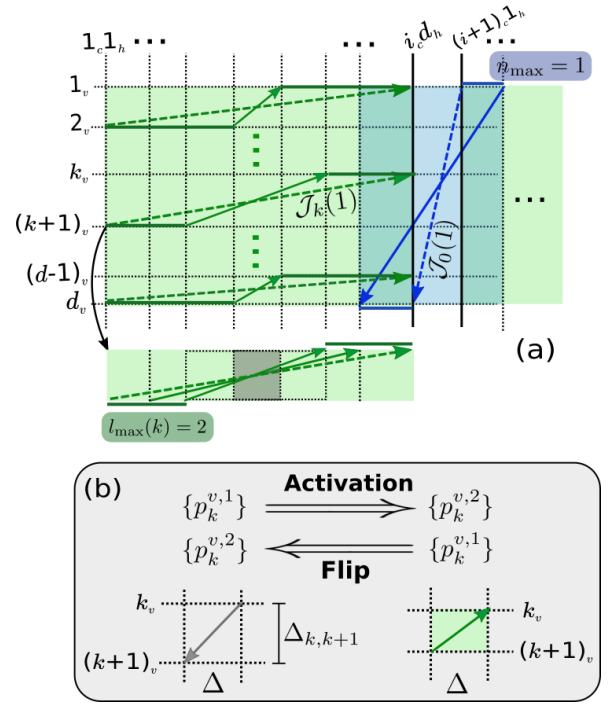$k \geq 1$.

Figure 17. (a) Activation of new cooling and restoring currents by the catalyst state $\rho_v(1)$. The eigenvalues of this state satisfy Eq. (H1), with the currents $\{\mathcal{J}_k(1)\}$ being depicted by the *dashed* green arrows (the length of each arrow must not be interpreted as the magnitud of the corresponding current). The two solid arrows in the lower rectangle are new restoring currents that contribute the new total current $\mathcal{J}_k^L(1)$. Here, $l_{\max}(k) = 2$ because the gray square does not contribute with a new current. For the other rectangles, the thick green lines cover all the eigenstates (columns) of $\rho_c \otimes \rho_h$ that are connected by new restoring currents (not shown). The new total cooling current $\mathcal{J}_0^{\text{tot}}(1)$ includes the current $\mathcal{J}_0(1)$ (dashed arrow), and the new cooling current corresponding to the solid blue arrow. (b) After the loop balance at step 3, the activation of a restoring current between some pair of rows k_v and $(k+1)_v$ can occur if for the new eigenvalues $\{p_k^{v,2}\}$ the distance $\Delta_{k,k+1} \equiv \ln(p_k^v/p_{k+1}^v)$ is sufficiently reduced. Conversely, an increment of $\Delta_{k,k+1}$ can cause that a current active with $\{p_k^{v,1}\}$ is flipped with $\{p_k^{v,2}\}$. A similar situation may take place for the cooling currents.

2. **Check for activation of new currents**: Let $\{p_l^{ch}\}_{1 \leq l \leq d_c d_h}$ denote the eigenvalues of $\rho_c \otimes \rho_h$, sorted in non-increasing order: $p_{l+1}^{ch} \leq p_l^{ch}$ for all l. For example, using this simplified notation $\mathcal{J}_0(1)$ is written as $\mathcal{J}_0(1) = p_{id_h+1}^{ch} p_1^{v,1} - p_{id_h}^{ch} p_{d_v}^{v,1}$. The state $\rho_v(1)$ potentially "activates" new restoring currents $\mathcal{J}_{k,l}^L(1) \equiv p_{l+1}^{ch} p_{k+1}^{v,1} - p_{id_h-l}^{ch} p_k^{v,1} > 0$, for $1 \leq l \leq l_{\max}(k)$, and $\mathcal{J}_{k,m}^R(1) = p_{id_h+m+1}^{ch} p_{k+1}^{v,1} - p_{d_c d_h-m}^{ch} p_k^{v,1} > 0$, for $0 \leq m \leq m_{\max}(k)$. In Fig. 17(a) we illustrate the new "left" currents $\{\mathcal{J}_{k,l}^L(1)\}_{1 \leq l \leq l_{\max}(k)}$. Note that, in general, the number $l_{\max}(k)$ may depend on k. Although the new "right" currents $\mathcal{J}_{k,m}^R(1)$ are not shown, the

only difference with respect to $\mathcal{J}_{k,l}^L(1)$ is that they must be located at the right of the column $(i+1)_c1_h$.

In addition, new cooling currents $\mathcal{J}_{0,n}(1) = p_{id_h+n+1}^{ch}p_1^{v,1} - p_{id_h-n}^{ch}p_{d_v}^{v,1} > 0$, for $1 \le n \le n_{\max}$, may also be activated. This results in a total cooling current $\mathcal{J}_0^{\text{tot}}(1)$ and total restoring currents $\mathcal{J}_k^{\text{tot}}(1) = \mathcal{J}_k^L(1) + \mathcal{J}_k^R(1)$, such that

$$\mathcal{J}_k^L(1) = \sum_{l=1}^{l_{\max}(k)} \mathcal{J}_{k,l}^L(1),$$

$$\mathcal{J}_k^R(1) = \sum_{m=1}^{m_{\max}(k)} \mathcal{J}_{k,m}^R(1),$$

$$\mathcal{J}_0^{\text{tot}}(1) = \sum_{n=1}^{n_{\max}} \mathcal{J}_{0,n}(1).$$

3. **Balance of loop currents**: If the state $\rho_v(1)$ does not activate any new (cooling or restoring) current, the protocol ends with the transformation $\rho(1) \to \rho'(1)$. Conversely, if at least one new current is activated, a new loop **loop**$_2 \equiv \{\mathcal{J}_0^{\text{tot}}(2)\} \cup \{\mathcal{J}_k^{\text{tot}}(2)\}_{1 \le k \le d_v-1}$ is defined by the state $\rho_v(2) = \sum_{k=1}^{d_v} p_k^{v,2}|k_v\rangle\langle k_v|$ whose eigenvalues satisfy the equations

$$\mathcal{J}_k^{\text{tot}}(2) = \mathcal{J}_{k+1}^{\text{tot}}(2) \text{ for } 0 \le k \le d_v - 1. \qquad \text{(H2)}$$

These equations impose that for the new eigenvalues $\{p_k^{v,2}\}$ the total currents in step 2 match each other. Hence, Eqs. H2 form a linear system with unknowns $\{p_k^{v,2}\}$, where

$$\mathcal{J}_0^{\text{tot}}(2) = a_1^{(+)}p_1^{v,2} - a_{d_v}^{(-)}p_{d_v}^{v,2},$$
$$\mathcal{J}_k^{\text{tot}}(2) = a_{k+1}^{(+)}p_{k+1}^{v,2} - a_k^{(-)}p_k^{v,2},$$

for $1 \le k \le d_v - 2$, and coefficients $\{a_k^{(+)}, a_k^{(-)}\}$ such that

$$a_1^{(+)} = \sum_{n=1}^{n_{\max}} p_{(id_h)+n+1}^{ch}, \quad a_{d_v}^{(-)} = \sum_{n=1}^{n_{\max}} p_{(id_h)-n}^{ch},$$

and

$$a_{k+1}^{(+)} = \sum_{l=1}^{l_{\max}(k)} p_{l+1}^{ch} + \sum_{m=1}^{m_{\max}(k)} p_{(id_h)+m+1}^{ch},$$

$$a_k^{(-)} = \sum_{l=1}^{l_{\max}(k)} p_{(id_h)-l}^{ch} + \sum_{m=1}^{m_{\max}(k)} p_{d_cd_h-m}^{ch},$$

for $1 \le k \le d_v - 1$.

4. **Iteration of steps 2 and 3**: After the loop balance at step 3 two situations can occur. If $\rho_v(2)$ does not generate any new current, the protocol

finishes with the transformation $\rho(2) \to \rho'(2)$. On the other hand, it is possible that new currents are activated by the new eigenvalues $\{p_k^{v,2}\}$, but also that currents activated at step 1 (with the eigenvalues $\{p_k^{v,1}\}$) *revert their direction* under $\rho_v(2)$. The second scenario is illustrated in Fig. 17 (b). Such currents are detrimental because they reduce the total cooling current (if a cooling current is flipped) or a total restoring current (if a restoring current is flipped). Hence, step 2 is iterated by including the new activated currents in the loop and removing the flipped currents from it. The new total currents are evaluated and step 3 is repeated by finding a loop **loop**$_3$ that solves the equations $\mathcal{J}_k(3) = \mathcal{J}_{k+1}(3)$, with the coefficients obtained from the previous step.

Let us see now that $J_{\text{cool}}(n+1) > J_{\text{cool}}(n)$. The loops generated at steps 2 and 3 of the protocol have the general structure **loop** $= \{\mathcal{J}_0^{\text{tot}}\} \cup \{\mathcal{J}_k^{\text{tot}}\}_{k \ge 1}$, where $\mathcal{J}_{k \ge 1}^{\text{tot}}$ is a sum of swap restoring currents connecting the catalyst eigenstates $|(k+1)_v\rangle$ and $|k_v\rangle$. Similarly, $\mathcal{J}_0^{\text{tot}}$ is a sum of swap cooling currents that connect the eigenstates $|1_v\rangle$ and $|d_v\rangle$. The maximum cooling current that guarantees the catalyst restoration is $J_{\text{cool}} = \min_{0 \le k \le d_v-1} \mathcal{J}_k^{\text{tot}}$. This current can always be achieved via partial swaps between the eigenstates of ρ for which $\mathcal{J}_{k'}^{\text{tot}} \ge \min_k \mathcal{J}_k^{\text{tot}}$, in such a way that the total current connecting any pair of catalyst eigenstates equals $\min_k \mathcal{J}_k^{\text{tot}}$. Since by construction $\mathcal{J}_k^{\text{tot}}(1) \ge \mathcal{J}_k(1) = \mathcal{J}_0(1)$ for all k, it follows that $\min_k \mathcal{J}_k^{\text{tot}}(1) \ge J_{\text{cool}}(1) = \mathcal{J}_0(1)$ (note that $J_{\text{cool}}(1) = \mathcal{J}_0(1)$ because $\min_k \mathcal{J}_k(1) = \mathcal{J}_0(1)$). This shows that the second step of the protocol produces a loop with a cooling current potentially larger than $J_{\text{cool}}(1)$.

On the other hand, $\min_k \mathcal{J}_k^{\text{tot}}(2) = \mathcal{J}_0(2)$, and therefore $J_{\text{cool}}(2) = \mathcal{J}_0(2)$. The crucial difference between the currents $\mathcal{J}_k^{\text{tot}}(1)$ and $\mathcal{J}_k^{\text{tot}}(2)$ is that the last ones form a uniform loop **loop**$_2$, since $\mathcal{J}_k^{\text{tot}}(2) = \mathcal{J}_{k+1}^{\text{tot}}(2)$. Based on this observation, we demonstrate in the following that $\mathcal{J}_0(2) \ge \min_k \mathcal{J}_k^{\text{tot}}(1)$, which amounts to have $J_{\text{cool}}(2) \ge J_{\text{cool}}(1)$. The proof is completely analogous to that employed in Appendix E to show that the corresponding optimal calalyst eigenvalues generate a uniform loop of swap currents. Here, this means in particular that if the catalyst eigenvalues $\{p_k^{v,2}\}$ are transformed into $\{p_k^{v,1}\}$, the resulting loop is suboptimal. More generally, we can consider the analogous of condition E6,

$$q_k^v = p_k^{v,2} + \delta_k, \qquad \text{(H3)}$$

where δ_k is such that $\{q_k^v\}$ represent valid catalyst eigenvalues. The variations δ_k give rise to corresponding variations in the currents $\mathcal{J}_k^{\text{tot}}(2)$, denoted as $\delta\mathcal{J}_k^{\text{tot}}(2) = \mathcal{J}_k'^{\text{tot}}(2) - \mathcal{J}_k^{\text{tot}}(2)$, where $\mathcal{J}_k'^{\text{tot}}(2)$ is the current that results from replacing $p_k^{v,2}$ by q_k^v in $\mathcal{J}_k^{\text{tot}}(2)$. Using the expressions $\mathcal{J}_0^{\text{tot}}(2) = a_1^{(+)}p_1^{v,2} - a_{d_v}^{(-)}p_{d_v}^{v,2}$ and $\mathcal{J}_{k\ge1}^{\text{tot}}(2) =$

$a_{k+1}^{(+)} p_{k+1}^{v,2} - a_k^{(-)} p_k^{v,2}$, it follows that

$$\delta \mathcal{J}_{k\geq 1}^{\text{tot}}(2) = a_{k+1}^{(+)} \delta_{k+1} - a_k^{(-)} \delta_k, \qquad \text{(H4)}$$

$$\delta \mathcal{J}_0^{\text{tot}}(2) = a_1^{(+)} \delta_1 - a_{d_v}^{(-)} \delta_{d_v}. \qquad \text{(H5)}$$

These equations are the analogous of Eqs. E7 and E8.

As in Appendix E, the fundamental idea is that the minimum current in the loop $\{\mathcal{J}_0'^{\text{tot}}(2)\} \cup \{\mathcal{J}_k'^{\text{tot}}(2)\}$ can be larger than $\mathcal{J}_0^{\text{tot}}(2)$ only if $\delta \mathcal{J}_k^{\text{tot}}(2) > 0$ for $0 \leq k \leq d_v - 1$. We prove that this is impossible by reproducing the argument used in Appendix E, with $\delta \mathcal{J}_k^{\text{tot}}(2)$ taking the role of $\delta \mathcal{J}_k$, d_v taking the role of n, and the replacements

$$p_1^h \to a_{k+1}^{(+)}, \quad p_2^h \to a_k^{(-)},$$
$$p_2^c p_1^h \to a_1^{(+)}, \quad p_1^c p_2^h \to a_{d_v}^{(-)}.$$

Since the proof in Appendix E essentially relies on the positivity of the coefficients $\{p_1^h, p_2^h, p_2^c p_1^h, p_1^c p_2^h\}$, and $a_k^{(+)}$ and $a_k^{(-)}$ are also all positive, it is equally applicable in this case.

From Eqs. (H4) and (H5), $\delta \mathcal{J}_k^{\text{tot}}(2) > 0$ implies that $\delta_k < 0$ or $\delta_{k+1} > 0$, and $\delta \mathcal{J}_0^{\text{tot}}(2) > 0$ implies that $\delta_1 > 0$ or $\delta_{d_v} < 0$. Let us see that these conditions cannot be both satisfied.

- If $\delta_k < 0$ for some k, $\delta \mathcal{J}_{k-1}^{\text{tot}}(2) = a_k^{(+)} \delta_k - a_{k-1}^{(-)} \delta_{k-1}$ (cf. Eq. (H4)) can be positive only if $\delta_{k-1} < 0$. Therefore, a simple generalization by induction leads us to conclude that $\delta \mathcal{J}_{l \leq k}^{\text{tot}}(2) \geq 0$ only if $\delta_l < 0$ for all $l \leq k - 1$. In particular, $\delta_1 < 0$. On the other hand, from probability conservation it follows that if $\delta_l < 0$ for all $l \leq k$ then $\delta_m > 0$ for some $m \geq k + 1$. This in turn implies that $\delta \mathcal{J}_m^{\text{tot}}(2) = a_{m+1}^{(+)} \delta_{m+1} - a_m^{(-)} \delta_m > 0$ only if $\delta_{m+1} > 0$, and by induction that $\delta \mathcal{J}_{l \geq m}^{\text{tot}}(2) > 0$ only if $\delta_l > 0$ for all $l \geq m+1$. In this way we conclude that $\delta_{d_v} > 0$ and $\delta_1 < 0$, and following Eq. (H5) that $\delta \mathcal{J}_0^{\text{tot}}(2) < 0$. Therefore, it is not possible to have $\delta \mathcal{J}_k^{\text{tot}}(2)$ positive for all $1 \leq k \leq d_v - 1$ and also $\delta \mathcal{J}_0^{\text{tot}}(2)$ positive, assuming that $\delta_k < 0$ for some k. This leaves us with the only option $\delta_{k+1} > 0$ to have $\delta \mathcal{J}_k^{\text{tot}}(2) > 0$, which we address below.

- If $\delta_{k+1} > 0$ for some k, we can apply a completely analogous reasoning. In this case $\delta_l > 0$ for all $l \geq k + 2$ is necessary for the positivity of $\delta \mathcal{J}_{l \geq k+1}^{\text{tot}}(2)$, which implies that $\delta_{d_v} > 0$. Moreover, from the condition $\delta_m < 0$ for some $m \leq k$ (derived from probability conservation) it follows that $\delta \mathcal{J}_{l \leq m-1}^{\text{tot}}(2) > 0$ only if $\delta_l < 0$ for $l \leq m - 1$. Once again, we conclude that $\delta_1 < 0$ and $\delta_{d_v} > 0$, and consequently that $\delta \mathcal{J}_0^{\text{tot}}(2) < 0$.

In this way, we show that $\min_k \mathcal{J}_0'^{\text{tot}}(2) \leq \mathcal{J}_0^{\text{tot}}(2)$. Since $\mathcal{J}_0'^{\text{tot}}(2) = \mathcal{J}_0^{\text{tot}}(1)$ for $\{\delta_k\}$ such that $q_k^v = p_k^{v,1}$, it follows that $J_{\text{cool}}(2) \geq J_{\text{cool}}(1)$. Any iteration of steps 2 and 3 of the protocol also produce increasing currents, because: when iterating step 1 the new total currents are by construction all larger or equal than $\mathcal{J}_0^{\text{tot}}(2)$, and the loop generated by these currents is optimized by choosing a catalyst state that makes it uniform, which corresponds to repeat step 3. Hence, $J_{\text{cool}}(n+1) \geq J_{\text{cool}}(n)$ for all n.

2. Protocol for optimization of CC transformations

Since the restoring currents that characterize the loop $\textbf{loop}_2$ in general connect different eigenspaces of the cold object, this loop provides a CNU transformation but not necessarily a CC transformation. However, a straightforward adaptation of the protocol guarantees that $\rho(2) \to \rho'(2)$ and the subsequent transformations derived from step 4 are catalytic and cooling. To this end we only need to impose that the restoring currents in each loop are generated using exclusively the hot object. As shown in Appendix D, this can always be done if the hot object is not fully mixed, using a catalyst that satisfies $\frac{p_k^v}{p_{k+1}^v} = \bar{\mu}$ for $1 \leq k \leq d_v - 1$, with $\bar{\mu} \equiv \frac{p_1^h}{p_{d_h}^h} - \epsilon$. Such a catalyst provides the state for the loop $\textbf{loop}_0 = \{\mathcal{J}_0\} \cup \{\mathcal{J}_k\}_{1 \leq k \leq d_v - 1}$, where $\mathcal{J}_0 \equiv \mathcal{J}_{|(i+1)_c 1_h 1_v\rangle \to |i_c d_h d_v\rangle}$ and $\mathcal{J}_{k \geq 1} \equiv \mathcal{J}_{|1_h (k+1)_v\rangle \to |d_h k_v\rangle}$.

For step 1 of the adapted protocol we only need to replace $\mathcal{J}_{k \geq 1}(1)$ with the currents defined above. That is, $\mathcal{J}_{k \geq 1}(1) = p_1^h p_{k+1}^{v,1} - p_{d_h}^h p_k^{v,1}$. We note also that the cooling current $\mathcal{J}_0$ and the subsequent currents $\mathcal{J}_0(n)$ have the same structure in both protocols. The activation of restoring currents in step 2 yields total currents

$$\mathcal{J}_{k \geq 1}^{\text{tot}}(1) = \sum_{l=1}^{l_{\max}(k)} \mathcal{J}_{k,l}(1),$$

where $\mathcal{J}_{k,l}(1) \equiv p_{l+1}^h p_{k+1}^{v,1} - p_{d_h - l}^h p_k^{v,1}$. By matching these currents between each other and with the total cooling current (step 3 of the adapted protocol) the catalyst eigenvalues $\{p_{k+1}^{v,2}\}$ are obtained.

The analogous of the $\ln(p^{ch}) \times \ln(p^v)$ diagram in Fig. 14(a), for the adapted protocol, is given in Fig. 15. The iteration of steps 2 and 3 is also performed in this case, if new currents are activated or flipped by the state $\rho_v(2)$. As seen in this figure, the restoration using a unitary $\mathbb{I}_c \otimes V_{hv}$ implies that the restoring currents lie inside the eigenspaces of the cold object.

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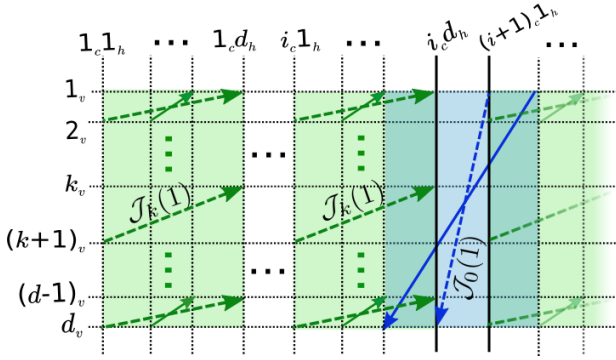
Figure 18. Activation of new currents in the second step of the protocol for optimizing CC transformations. The dashed arrows correspond to the currents $\mathcal{J}_0(1)$ (blue) and $\{\mathcal{J}_k(1)\}_{k \geq 1}$ (green), that define the loop **loop**$_1$. The solid arrows illustrate new currents activated by the state $\rho_v(1)$. The restoring currents are replicated in the eigenspaces $\text{span}\{|m_c j_h\rangle\}_j$, for $1 \leq m \leq d_c$.

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