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

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The laws of thermodynamics are usually formulated without setting restrictions on the size of the environment, and not much is known about machines that operate with small environments. We provide explicit protocols for the operation of machines that circumvent certain thermodynamic restrictions, which arise naturally in this context. Such machines are more precisely described as catalysts, as they can be used to perform otherwise impossible transformations. We establish sufficient conditions for catalytic transformations that cannot be achieved when the system interacts with a finite environment. From this key 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. For the optimal catalytic cooling involving a cold qubit and a hot qubit, it is demonstrated that maximum cooling can be attained with a catalyst as small as a three-level system. Moreover, we show that in a multiqubit setup catalytic cooling based on a three-body interaction outperforms standard (non-catalytic) cooling with higher order interactions. We also demonstrate that even transformations that are allowed without catalysts can be enhanced through their use. Besides cooling, such 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 chemical compounds 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 [1–9] and quantum thermodynamics [10–15], where catalysts correspond to quantum systems that enable the implementation of otherwise impossible transformations. For example, transformations that are forbidden if only local operations and classical communication (LOCC) on a bipartite state are available, become possible once a suitable entangled state is employed as catalyst [1]. It is also worth mentioning that the mathematical framework for these transformations relies on the notion of “catalytic majorization” [2], which applies to scenarios beyond entangled states and the LOCC paradigm.

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 [10, 14]. These results refer to transformations where the system is coupled to a catalyst and an arbitrarily large environment, and the interaction preserves the energy of the total system [16–24]. Such conditions define a thermodynamic framework known as “thermal operations”. Here, we consider catalytic transformations performed through general unitary evolutions that act jointly on the system, the catalyst, and a finite-size environment. This setup is illustrated in Fig. 1. Our main motivation is the study of transformations where the inclusion of the catalyst is crucial to cool the system. In addition, realistic systems interact with environments of limited size, or at most with finite portions of very large environments.

Cooling has been studied using techniques of open quantum systems [25] and information theory [23], with traditional approaches that include the analysis of quantum refrigerators [26–30] and algorithmic cooling [31–35]. Recently, an important effort has been devoted to understand and formalize the fundamental limits for this task [36–40]. These limits are intimately connected with the resources at hand. For example, in the framework of thermal operations, catalytic cooling is possible only in combination with an additional system that starts in a non-equilibrium state [13]. Within the context of energy-preserving interactions, it has also been shown that finite environments limit the probability for (non-catalytic) transitions to the ground state [41].

On the other hand, we are interested in transformations where work exchange is possible and the key physical constraint is related to the finite character of the environment. It is important to stress that by “work” we refer to the energy injected (or extracted) by a classical driving, which differs from the definition adopted in the case of thermal operations [16, 42, 43]. We also allow for the arbitrary generation of correlations between the catalyst and the other systems involved in the transformation (cf. Fig. 1(b)). This contrasts with the assumption, made in previous works [10, 11], of a final state where the catalyst is uncorrelated from the main system (see Ref. [14] for results concerning the removal of this constraint).

As illustrated in Fig. 1(a), cooling may be forbidden in situations where the environment is too small. More formally, such a limitation implies that joint unitary operations on the system and the environment cannot lower
Figure 1. General framework for the studied catalytic transformations. (a) Illustrative example: a three-level system in the initial state $\rho_s$ is used as (hot) environment to cool a (cold) qubit in the initial state $\rho_c$. The eigenvalues of the joint state $\rho_c \otimes \rho_h$ are obtained by rescaling the eigenvalues of $\rho_h$ (orange bars) with the eigenvalues of $\rho_c$. When cooling is possible a swap $|0,2\rangle \leftrightarrow |1,0\rangle$ (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 $U_{ch}$. (b) The inclusion of a catalyst in an appropriate initial state $\rho_c$ allows to lift this restriction. If $\rho_c = \rho_s \otimes \rho_h$, cooling is enabled only if the corresponding final state $\rho'_c$ is not majorized by $\rho_s$ (non-unital transformation). (c) The transformations are implemented by global unitaries of the form $U = U_{\text{cool}} \otimes V_{\text{res}}$, where $U_{\text{cool}}$ is employed to cool the system and $V_{\text{res}}$ returns the catalyst to its initial state.

the average energy of the system. A catalyst that enables to circumvent this restriction plays the role of an additional environment, which not only allows cooling but also remains unaltered by the global interaction in which it takes part. This kind of catalytic transformation belongs to a broader class that we term “non-unital transformations”, which are illustrated in Fig. 1(b). From a physical viewpoint, non-unital transformations represent state transitions that cannot be driven by classical electromagnetic fields. Cooling is an example of transformation that satisfies this property, as it requires reducing the energy of a thermal state, and any thermal state is passive [44, 45]. The example in Fig. 1(a) depicts the conditions that prevent to cool a two-level system using a three-level environment. If $\rho_c = \sum_{i=0}^{1} \rho_i ^c |i_c \rangle \langle i_c|$ and $\rho_h = \sum_{j=0}^{2} \rho_j ^h |j_h \rangle \langle j_h|$ denote respectively the states of the (‘cold’) system and the (‘hot’) environment, with $\rho_0 ^h \geq \rho_j ^h$ and $\rho_0 ^h \geq \rho_{j+1} ^h$, cooling is possible if and only if $\rho_0 ^h \rho_2 ^h < \rho_1 ^h \rho_0 ^h$. When this inequality is not fulfilled, any joint unitary $U_{ch}$ has the effect of a mixture of local unitary operations (corresponding to the probabilistic application of different unitaries) on the system.

The usefulness of catalytic non-unital transformations is not limited to cooling. Their applicability is further illustrated by considering an example where the use of a catalyst yields an advantage for thermometry. In thermometry [46], and metrology in general [47, 48], various results refer to the optimization of the so called the Cramer-Rao bound [49, 50], which constitutes a lower bound for the estimation error of some physical parameter. In the case of thermometry this error quantifies the precision of a temperature estimation [51–55, 57]. If the environment interacts with a probe, measurements on the probe contain information about the temperature of the environment [57–59]. In this context, we show that a catalytic transformation allows to reduce the minimum estimation error achieved through optimal interactions using only the probe.

Our results differentiate from previous works in two key aspects. On the one hand, we provide explicit protocols for the construction of catalytic transformations, which include the explicit global unitary $U$ and the initial states of the catalyst and the environment. The unitaries investigated have the structure indicated in Fig. 1(c). While some results exist about explicit catalyst states [11, 12], no general methods to construct catalytic transformations are known beyond very specific cases [12]. On the other hand, we consider transformations where the initial and final catalyst states exactly coincide. In the context of thermodynamics it has been shown that inexact catalysis can lead to extreme physical consequences, even when the final state of the catalyst deviates little from its initial configuration. Essentially, under inexact catalysis the catalyst can become a source of energy or purity that allows any state transition [10, 11]. By considering exact transformations, we prevent this possibility and also guarantee that the catalyst is never degraded (within practical limitations), irrespective of the number of times it is used.

The paper is structured as follows. After some general comments on notation and definitions (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. Our first main finding consists in the establishment of necessary and sufficient conditions for a class of catalytic non-unital transformations. In the same section, we also develop a graphical method that provides an intuitive picture for this result and subsequent derivations. The applications to catalytic and cooling transformations are addressed in Sect. V. This section is based on two fundamental results, which refer to:

- Catalytic cooling with arbitrarily small environments.
- Catalytic cooling where the catalyst enhances the cooling, even if the environment is large enough to cool the system.

Such results imply that a sufficiently large catalyst enables the aforementioned transformations, if the initial
states of the system and the environment satisfy certain conditions. As one of the main applications we derive the catalyst state that maximizes the cooling of a qubit using another qubit as environment, for catalysts of arbitrary (finite) dimension. In addition, we study a catalytic transformation that enhances the optimal cooling obtained by coupling a two-level system to a three-level environment. In Sect. VI we extend our findings to a scenario where the environment can be arbitrarily large. We show that, even without the size constraint, larger cooling can be achieved with less control on the environment by employing a two-level catalyst. In Sect. VII we demonstrate 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 the conclusions and perspectives in Sect. VIII.

II. BRIEF REMARKS ON NOTATION AND DEFINITIONS

In what follows we will term the system to be cooled and the environment “cold object” and “hot object”, respectively. Moreover, the ground state of these systems will be denoted using the label “0” 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, cold and hot objects 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.

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 [60, 61]. While traditionally it has been associated to the Hamiltonian and the impossibility of work extraction [12, 45], 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 $\rho_c = e^{-\beta_c H_c} / \text{Tr}[e^{-\beta_c H_c}]$ and $\rho_h = e^{-\beta_h H_h} / \text{Tr}[e^{-\beta_h H_h}]$ are respectively the thermal states of the cold object and the hot object. The corresponding Hamiltonians are $H_c = \sum_{i=1}^{d_c} \epsilon_i^c |i_c\rangle\langle i_c|$ and $H_h = \sum_{i=1}^{d_h} \epsilon_i^h |i_h\rangle\langle i_h|$.

For inverse temperatures $\beta_c$ and $\beta_h$ 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

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

for any global unitary $U_{ch}$ (e.g. if the the cold temperature $\beta_c^{-1}$ 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 $\rho_h$. 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 $\rho_c$ 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 $\rho_c$ and $\rho_h$, being $d_c$ ($d_h$) the dimension of the Hilbert space of the cold (hot) object. Using the standard convention of non-decreasing eigenenergies, $\epsilon_i^c \leq \epsilon_{i+1}^c$ and $\epsilon_j^h \leq \epsilon_{j+1}^h$, passivity is easily expressed by means of the inequalities

$$\frac{p_i^c}{p_{i+1}^c} \geq \frac{p_j^h}{p_{j+1}^h} \forall i, j.$$  \hspace{1cm} (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, $p_i^c / p_{i+1}^c = 1$ 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, $p_i^h / p_{i+1}^h = (q_{max} / q_{min})^N$, where $q_{max}$ and $q_{min}$ denote respectively the highest and smallest populations of each subsystem. Since $q_{max} / q_{min} > 1$ for finite temperature, in the limit $N \to \infty$ the ratio $p_i^h / p_{i+1}^h$ 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 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 $B$ [60]. Equation (2) simply translates this condition to the operators $A = \rho_c \otimes \rho_h$ and $B = H_c$. 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 $\epsilon_i^c$ 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 $H_c$.

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 $\rho_c = \sum_{k=1}^{d} |\psi_k^c\rangle \langle \psi_k^c|$, 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 $\rho = \rho_c \otimes \rho_h \otimes \rho_o$. The transformation on the total system is implemented through a global unitary map $U$ that acts on the total system. Denoting the final total state as $\rho'$, a generic catalytic (C) transformation satisfies
\[
\rho \xrightarrow{C} \rho' = U (\rho_c \otimes \rho_h \otimes \rho_o) U^\dagger, \quad (3)
\]
\[
\text{Tr}_{ch} (\rho') = \rho_o. \quad (4)
\]

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. Contrary to previous works on catalysts, we do not impose any restriction on the nature and strength of these correlations. This additional degree of freedom naturally extends the set of transformations that become possible once the catalyst is introduced [14]. 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, allowing to repeat exactly the same process performed with the old copy. This is possible because the initial total state with the new copy is identical to that with the old one, i.e. $\rho_c \otimes \rho_h \otimes \rho_o$. 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 ($H_c$) as long as the total state $\rho$ is non-passive with respect to $H_c$. Let $\{p_k^h\}_{1 \leq k \leq d_h}$ denote the eigenvalues of $\rho_h$ or “catalyst eigenvalues”, for a catalyst of dimension $d_h$, and let $\{p_k\} = \{p_k^c p_k^h\}$ denote the eigenvalues of $\rho$. Assuming the ordering $p_k^c \geq p_k^h$ for all $k$, the catalyst breaks down the initial passivity (2) if and only if there exists $i$ such that
\[
\frac{p_i^c}{p_{i+1}^h} < \frac{p_i^h}{p_{i+1}^h}. \quad (5)
\]

Since the ratio between the highest and smallest eigenvalues of the composite state $\rho_h \otimes \rho_c$ (r.h.s. of Eq. (5)) is always larger than $p_i^h / p_{i+1}^h$, by a factor of $p_i^h / p_{i+1}^h$, 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 $\rho_c = |1\rangle \langle 1|$. 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 $U_{\text{swap}}$ that swaps the states $|i,d,h\rangle$ and $(i+1), \langle 1\rangle, |1\rangle, \langle 1\rangle$ respectively, acting as the identity on any other eigenstate of $\rho$. This unitary transfers population $\delta p = p_i^h \delta p_{i+1}^h - p_i^c \delta p_{i+1}^c$ from $|i,d,h\rangle$ to $(i+1), \langle 1\rangle, |1\rangle, \langle 1\rangle$, thereby reducing the energy of the cold object by $\Delta (H_c) = -\delta p (\varepsilon_{i+1} - \varepsilon_i)$. However, the same operation modifies the state $\rho_o$ by transferring population $\delta p$ from the eigenstate $|1\rangle$ towards the eigenstate $|d_o\rangle$. 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 $U_{\text{swap}}$.

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 $U_{\text{cool}}$ and $V_{\text{res}}$, 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 $U_{\text{swap}}$ described before. The restoring unitary is chosen in such a way that the subspaces where $U_{\text{cool}}$ and $V_{\text{res}}$ are defined are orthogonal. Let $\mathcal{H}_{\text{cool}} \subset \mathcal{H}$ and $\mathcal{H}_{\text{res}} \subset \mathcal{H}$ denote such subspaces, being $\mathcal{H} = \text{span} \{i, j, h, k_*\}$ the total Hilbert space. We consider global unitaries that satisfy Eq. (4) and have the form
\[
U = U_{\text{cool}} \oplus V_{\text{res}}, \quad (6)
\]
\[
U_{\text{cool}} : \mathcal{H}_{\text{cool}} \rightarrow \mathcal{H}_{\text{cool}}, \quad (7)
\]
\[
V_{\text{res}} : \mathcal{H}_{\text{res}} \rightarrow \mathcal{H}_{\text{res}}, \quad (8)
\]
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 $V_{\text{res}}$ is preceded by $U_{\text{cool}}$. 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 $U_{\text{cool}}$ and $V_{\text{res}}$ also allows us to study the effect of $V_{\text{res}}$ using directly the initial state $\rho_o$ instead of the state obtained after the application of $U_{\text{cool}}$.

B. Majorization, cooling currents and restoring currents

1. Majorization, cooling, and non-unitality

Majorization defines a preorder between quantum states [62, 63], and is intimately connected with different cooling criteria [38]. Let $\varrho$ and $\sigma$ denote two generic quantum states defined on some Hilbert space of dimension $d$, with respective eigenvalues $\{r_i\}_{1 \leq i \leq d}$ and $\{q_i\}_{1 \leq i \leq d}$. Moreover, let $\{r_i^c\}$ and $\{q_i^c\}$ be the
same eigenvalues arranged in non-increasing order, i.e. $r_{i+1}^+ \leq r_i^+$ and $q_{i+1}^+ \leq q_i^+$. It is said that “$\varrho$ majorizes $\sigma$”, formally written as $\varrho \succ \sigma$, if
\begin{equation}
\sum_{i=1}^{j} r_i^+ \geq \sum_{i=1}^{j} q_i^+
\end{equation}
for all $1 \leq j \leq d$. 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 $\varrho$ and $\sigma$ 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 [63, 64] 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 $\mathcal{E}_\text{un}(|I\rangle) = |I\rangle$ [64]. 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 $\varrho$.

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 $\rho_c'$ 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 $\rho_c$ does not majorize $\rho_c'$, or equivalently that $\rho_c \to \rho_c'$ is a “non-unital transformation”. This occurs in particular if the populations of $\rho_c'$ in the eigenbasis of $\rho_c$ satisfy
\begin{equation}
\sum_{i=1}^{d_c} p_i^{c'} < \sum_{i=1}^{d_c} q_i^{c'} \quad \text{for some } 1 \leq i \leq d_c - 1,
\end{equation}
where $p_i^{c'} = \text{Tr}(|i\rangle \langle i| \rho_c')$ and $q_i^{c'} = p_i^c |i\rangle \langle i|$. Let us see why a transformation satisfying Eq. (10) cannot be achieved through a unital map. First, the eigenvalues $\{p_i^{c'}\}$ are by construction non-increasing and therefore $\sum_{i=1}^{d_c} p_i^{c'} = \sum_{i=1}^{d_c} p_i^c$. In addition, the populations $\{p_i^{c'}\}$ correspond to the eigenvalues of the state $\mathcal{D}(\rho_c')$, where $\mathcal{D}$ is the map that completely erases the coherence of $\rho_c'$ in the eigenbasis of $\rho_c$. This implies that $\sum_{i=1}^{d_c} p_i^{c'} \leq \sum_{i=1}^{d_c} q_i^{c'}$ for all $i$ [Ref.], where $\{q_i^{c'}\}$ are the sorted eigenvalues of $\rho_c'$ (i.e. $\rho_c' \succ \mathcal{D}(\rho_c')$). By plugging this inequality in the r.h.s. of Eq. (10), we obtain the relation $\sum_{i=1}^{d_c} p_i^{c'} < \sum_{i=1}^{d_c} q_i^{c'}$, 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.

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 $\rho$. 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 $|i\rangle$ and $|j\rangle$, due to the action of a two-level unitary $U^{(2)} : \text{span}\{|i\rangle, |j\rangle\} \to \text{span}\{|i\rangle, |j\rangle\}$. Any population transferred in this way can be described through a unitary of the form
\begin{equation}
U^{(2)}(i) = \sqrt{1 - a^2} |i\rangle + a |j\rangle,
\end{equation}
\begin{equation}
U^{(2)}(j) = a |i\rangle - \sqrt{1 - a^2} |j\rangle,
\end{equation}
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 in the construction of an intuitive picture for the transformations that will be studied throughout this paper.

Keeping in mind that the initial populations are $p_i = \text{Tr}(|i\rangle \langle i| \rho)$ and $p_j = \text{Tr}(|j\rangle \langle j| \rho)$, after the application of $U^{(2)}$ the state $|j\rangle$ acquires population
\begin{equation}
p_j' = p_j a^2 + p_j (1 - a^2) = 2 p_j - a^2 (p_j - p_i).
\end{equation}
In this way, the population current from $|i\rangle$ to $|j\rangle$ is defined as
\begin{equation}
J_{|i\rangle \to |j\rangle} = \begin{cases} a^2 (p_i - p_j), & \text{if } p_i > p_j \\ 0, & \text{if } p_i \leq p_j. \end{cases}
\end{equation}

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 $U^{(2)}$
By definition, the resulting transformation is unital. The two-level unitary acting on the subspace span implies that there exists a two-level unitary transferring population from an eigenstate of $\rho_c \otimes \rho_h$ with eigenvalue $p_k^{c+n}$, towards an eigenstate with larger or equal eigenvalue $p_j^{v}$. However, the study of cooling transformations will be mainly based on restoring currents of the kind $J_{|j_v, (k+n)_v\rangle \rightarrow |j'_v, k_v\rangle}$, which are associated with two-level unitaries that do not involve the cold object. From Eq. (15), it is straightforward to check that $J_{|j_v, (k+n)_v\rangle \rightarrow |j'_v, k_v\rangle}$ exists iff

$$
\frac{p_j^{h}}{p_j^{v}} > \frac{p_k^{v}}{p_k^{h+n}} \nonumber
$$

which reduces the population of the catalyst eigenstate $|k_v\rangle$.

Eventually, we will also employ the notation $J_{|i\rangle \rightarrow |j\rangle}$ for a current that describes a population flow from $|i\rangle$ to some unknown eigenstate of $\rho$. Likewise, $J_{|i\rangle \rightarrow |j\rangle}$ 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 $J_{|i\rangle \rightarrow |j\rangle}$” signifies that $J_{|i\rangle \rightarrow |j\rangle} \neq 0$, which in turn implies that there exists a two-level unitary transferring population from $|i\rangle$ to $|j\rangle$. Moreover, we note that $J_{|i\rangle \rightarrow |j\rangle}$ exists iff $J_{|j\rangle \rightarrow |i\rangle}$ exists.

**Definition 1 (cooling current).** A cooling current is a current $J_{|(i+n)_c,j_h,k_v\rangle \rightarrow |i,j'_v,k'_v\rangle}$ ($n \geq 1$), whose function is to transfer population from an eigenstate of $\rho_c \otimes \rho_h \otimes \rho_v$ with eigenvalue $p_k^{c+n}$, towards an eigenstate with larger or equal eigenvalue $p_j^{v}$. From Eq. (15) it is straightforward to check that $J_{|(i+n)_c,j_h,k_v\rangle \rightarrow |i,j'_v,k'_v\rangle}$ exists iff

$$
\frac{p_k^{v}}{p_k^{h+n}} > \frac{p_j^{h}}{p_j^{v}} \nonumber
$$

where the second inequality follows by bounding $\frac{p_k^{v}}{p_k^{h+n}}$ from above and $\frac{p_j^{h}}{p_j^{v}}$ 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 $\rho_c$ are such that there exists a cooling current $J_{|(i+n)_c,j_h,k_v\rangle \rightarrow |i,j'_v,k'_v\rangle}$. Such a current has opposite effects on the cold object and the catalyst. First, it increases the partial sum $\sum_{l=1}^{i} p_l^{c}$ by transferring population to $|i_{c}\rangle$, which yields a non-unitary 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 $p_k^{c+n} > p_k^{h+n}$, 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 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 $\rho_c$ towards a high-population eigenstate.

**Definition 2 (restoring current).** A general restoring current is a current $J_{|i_{c},j_h,(k+n)_v\rangle \rightarrow |i_{c},j'_v,k_v\rangle}$ ($n \geq 1$), whose function is to transfer population from an eigenstate of $\rho_c \otimes \rho_h$ with $p_k^{c+n}$, towards an eigenstate with larger or equal eigenvalue $p_j^{v}$. However, the study of cooling transformations will be mainly based on restoring currents of the kind $J_{|j_v, (k+n)_v\rangle \rightarrow |j'_v, k_v\rangle}$, which are associated with two-level unitaries that do not involve the cold object. From Eq. (15), it is straightforward to check that $J_{|j_v, (k+n)_v\rangle \rightarrow |j'_v, k_v\rangle}$ exists iff

$$
\frac{p_j^{h}}{p_j^{v}} > \frac{p_k^{v}}{p_k^{h+n}} \nonumber
$$

where the second inequality follows by bounding $\frac{p_k^{v}}{p_k^{h+n}}$ from below and $\frac{p_j^{h}}{p_j^{v}}$ 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 $U_{\text{cool}} = U_{\text{cool}}^{(2)}$ is given by a single two-level unitary. We denote these transformations as $\rho \xrightarrow{\text{CNU1}} \rho'$. Moreover, we consider restoring unitaries $V_{\text{res}}$ of the form

$$
V_{\text{res}} = \otimes_k V_k^{(2)},
$$

where each $V_k^{(2)}$ is a two-level unitary giving rise to a restoring current. Given a cooling current $J_{|(i+n)_c,j_h,k_v\rangle \rightarrow |i,j'_v,k'_v\rangle}$, the goal of $V_{\text{res}}$ 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 $J_{|(i+n)_c,j_h,k_v\rangle \rightarrow |i,j'_v,k'_v\rangle}$. However, in general such a current may not exist. The following theorem provides necessary and sufficient conditions for the existence of a CNU1 transformation, by characterizing the currents that lead to a proper restoring unitary $V_{\text{res}}$. The proof of this theorem is given in Appendix A.

**Theorem 1 (existence of CNU1 transformations).** Let $\rho = \rho_c \otimes \rho_h \otimes \rho_v \in \mathcal{B}(\mathcal{H}_c \otimes \mathcal{H}_h \otimes \mathcal{H}_v)$ 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 $\{p_l^{c}\}$ denote the eigenvalues of $\rho_c$. A CNU1 transformation $\rho \xrightarrow{\text{CNU1}} \rho' = U_{\text{cool}}^{(2)} \otimes V_{\text{res}} \rho U_{\text{cool}}^{(2)} \otimes V_{\text{res}}^\dagger$ exists iff

$$
\sum_{l=1}^{i} p_l^{c} > \frac{p_k^{v}}{p_k^{h+n}},
$$

for some $1 \leq i \leq d_c - 1$ and $1 \leq l, l' \leq d_v - 1$, and
2. For $l \leq k \leq l'$ there exist restoring currents \( \{J_{[l,k(l+1)_v]} \rightarrow [l,d_h k_v] \}_{l \leq k \leq l'} \), determined by the conditions
\[
\left( \frac{p^h_l}{p^h_{l_d}} \right) \frac{p^v_i}{p^v_{l_d}} > \left( \frac{p^v_{l+1}}{p^v_{l_d+1}} \right) > 1, \tag{20}
\]
or restoring currents \( \{J_{(l+1)_v, k(l+1)_v} \rightarrow [d_h, d_h k_v] \}_{l \leq k \leq l'} \), determined by the conditions
\[
\left( \frac{p^h_l}{p^h_{d_h}} \right) \frac{p^v_i}{p^v_{d_h}} > \left( \frac{p^v_{l+1}}{p^v_{d_h+1}} \right) > 1. \tag{21}
\]

**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 \( \rho_s \) 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 \( \rho_s \), but also on the state \( \rho_v \).

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

### 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:

**\( \ln(p^h) \times \ln(p^v) \) Diagram.** Consider an horizontal axis where the values \( \{\ln(p^h_{i,j} \rho^h_{i,j})\}_{1 \leq i \leq d_h, 1 \leq j \leq d_v} \) 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 \( \{\ln(p^v_{i,j})\}_{1 \leq k \leq d_v} \) 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 \( \ln(p^v_{i,j}) \), and represents also the catalyst eigenstate \( |k_v\rangle \). A “column” \( i_c, j_h \) is a vertical line that passes through the value \( \ln(p^h_{i,j} \rho^h_{i,j}) \), 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 \( \ln(p^h) \times \ln(p^v) \) 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 \( \ln(p^v_{1,l}) - \ln(p^v_{d_h}) > \ln(p^h_{i,j} \rho^h_{i,j}) - \ln(p^v_{l+1,1}) \). Therefore, the existence of a cooling current (downward-oriented blue arrow) means that

\[\text{the height of the } \ln(p^h) \times \ln(p^v) \text{ diagram must be larger than the distance between two consecutive columns } i, d_h \text{ and } (i+1), d_h, \text{ for some value of } i. \]

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 \( \ln(p^h_{i}) - \ln(p^h_{d_h}) > \ln(p^v_{k}) - \ln(p^v_{k+1}) \), meaning that the corresponding restoring current is enclosed by a horizontal rectangle of width \( \ln(p^h_{1,l})/\ln(p^h_{d_h+1}) \) and height \( \ln(p^v_{k})/\ln(p^v_{k+1}) \). 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 lengths obtained from the application of the natural logarithm to Eq. (19). Specifically, its height is given by \( \ln(p^h_{i}/p^h_{i+1}) \), and its width is given by \( \ln(p^h_{i}/p^v_{d_h+1}/p^v_{d_h+1}) \). Hence, a cooling current exists iff we can identify a vertical rectangle with vertical sides living on consecutive columns \( i, d_h \) and \( (i+1), d_h \). 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).
Figure 3. \( \ln(p^v) \times \ln(p^v) \) diagram for the conditions stated in Theorem 1. Vertical (horizontal) ellipses 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).

2. Effect of currents on the catalyst

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

\[
p^v_k - p^v_k = \text{Tr}[|k_v\rangle \langle k_v| (U \rho U^\dagger - \rho)] = J_{\rightarrow |k_v\rangle} - J_{|k_v\rangle \rightarrow}, \tag{22}
\]

where

\[
J_{\rightarrow |k_v\rangle} = \sum_{i,j} J_{\rightarrow |i,jk_v\rangle} \tag{23}
\]

is the sum of all the currents that transfer population to eigenstates with catalyst eigenvalue \( p^v_k \). Similarly,

\[
J_{|k_v\rangle \rightarrow} = \sum_{i,j} J_{|i,jk_v\rangle \rightarrow} \tag{24}
\]

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 \( J_{\rightarrow |i,jk_v\rangle} \) could connect \( |i,jk_v\rangle \) with another eigenstate that also has catalyst eigenvalue \( p^v_k \). If this occurs such a current cannot contribute to \( J_{\rightarrow |k_v\rangle} \), since it leaves invariant the population of \( |k_v\rangle \). However, we always deal with currents \( J_{\rightarrow |i,jk_v\rangle} \) and \( J_{|i,jk_v\rangle \rightarrow} \) connecting eigenstates with different catalyst eigenvalues, which implies that the quantities \( J_{\rightarrow |k_v\rangle} \) and \( J_{|k_v\rangle \rightarrow} \) are properly characterized by Eqs. (23) and (24). In the following \( J_{\rightarrow |k_v\rangle} \) and \( J_{|k_v\rangle \rightarrow} \) 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 \( J_{\rightarrow |k_v\rangle} = J_{|k_v\rangle \rightarrow} \). In the following lemma we provide necessary and sufficient conditions for the existence of a restoring unitary \( V_{\text{res}} \), given a two-level unitary \( U_{\text{cool}}^{(2)} \) that generates a single cooling current. Accordingly, the condition \( J_{\rightarrow |k_v\rangle} = J_{|k_v\rangle \rightarrow} \) holds under the action of the total unitary \( U = U_{\text{cool}}^{(2)} \otimes V_{\text{res}} \). Here we will prove sufficienty, leaving the proof of necessity for Appendix C.

**Lemma 1** (Existence of restoring unitaries). Let \( J_{\rightarrow |l_v\rangle, \rightarrow |l_v\rangle \rightarrow} \) be a cooling current, which increases (decreases) the population of the catalyst eigenstate \( |l_v\rangle \). A restoring unitary \( V_{\text{res}} \) that reverses the effect of this current on the catalyst exists iff there exists a set of restoring currents \( \{J_{\rightarrow |l_v\rangle, \rightarrow |l_v\rangle \rightarrow} \}_{1 \leq k \leq V} \) (see Fig. 4).

To 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 \( J_{\rightarrow |k_v\rangle} \) there is an outgoing current \( J_{|k_v\rangle \rightarrow} \). A “uniform loop” is a loop with the additional property that all the catalyst currents satisfy \( J_{\rightarrow |k_v\rangle} = J_{|k_v\rangle \rightarrow} \). 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 \( |l_v + 1\rangle \). In this way, a chain is a set

\[
\text{ch}_{|l_v\rangle \rightarrow |l_v + 1\rangle} = \{J_{|l_v\rangle \rightarrow |k_v^{(1)}\rangle}, J_{|k_v^{(1)}\rangle \rightarrow |l_v\rangle}, \ldots, J_{|l_v\rangle \rightarrow |k_v^{(n)}\rangle}, J_{|k_v^{(n)}\rangle \rightarrow |l_v + 1\rangle} \}, \tag{25}
\]

where \( J_{|l_v\rangle \rightarrow |k_v^{(1)}\rangle} \) is the total catalyst current connecting the states \( |l_v\rangle \) and \( |k_v^{(1)}\rangle \) \( \langle k_v^{(1)}| \) and \( |l_v + 1\rangle \).

Since \( J_{|l_v\rangle \rightarrow |k_v^{(1)}\rangle} \) 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.
Similarly, \( J_{k_v^{(1)}} \rightarrow \) 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 \( |(l' + 1)\rangle \) only has an incoming current. If \( \{ |k_v^{(2)}\rangle, |k_v^{(3)}\rangle, \ldots, |k_v^{(n-1)}\rangle \} \) denotes the set of all the remaining inner links, then the ellipsis in Eq. (25) stand for pairs \( \{ J_{k_v^{(i)}} \rightarrow J_{k_v^{(i)}} \} \) for \( 2 \leq i \leq n-1 \) of incoming and outgoing currents connecting these links. We also note that the simplest case has the form \( ch_{|l_v\rangle \rightarrow |(l'+1)\rangle} = \{ J_{l_v} \rightarrow |(l'+1)\rangle \} \), with a single catalyst current and no inner links.

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

**Sufficiency proof for Lemma 1.** If \( ch_{|l_v\rangle \rightarrow |(l'+1)\rangle} \) is the chain that contains only the cooling current \( \mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle} \) and the set \( \{ \mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle} \} \) is a chain of the type \( ch_{|(l'+1)\rangle \rightarrow |l_v\rangle} \), which generates a loop with \( ch_{|l_v\rangle \rightarrow |(l'+1)\rangle} \) (see Fig. 4). Let \( \mathcal{J}^\min_{\text{loop}} \) denote the minimum swap current in this loop. Moreover, let

\[
\tilde{V}_{\text{res}} = \bigoplus_{k=1}^{l'} \tilde{V}_{[(k+1)\rangle \rightarrow |l_v\rangle}^{(2)}
\]

be a unitary composed of partial swaps \( \tilde{V}_{[(k+1)\rangle \rightarrow |l_v\rangle}^{(2)} \) between the states \( |1, k\rangle (k + 1)\rangle \) and \( |d, d_k\rangle \), with “intensities” \( a_k^2 \) (cf. Eqs. (11) and (12)). Since \( \mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle} \) generates a restoring current \( \mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle} > 0 \). If the swap intensity of the cooling unitary, denoted by \( a_k^2 \), and the intensities \( a_k^2 \), satisfy

\[
a_k^2 = \frac{\mathcal{J}^\min_{\text{loop}}}{\mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle}}
\]

Eq. (14) implies that all the currents generated by \( \tilde{V}_{\text{res}} \) and \( \tilde{V}_{[(k+1)\rangle \rightarrow |l_v\rangle}^{(2)} \) have the same magnitude \( \mathcal{J}^\min_{\text{loop}} \). Therefore, a unitary \( U = U_{\text{cool}}^{(2)} \oplus \tilde{V}_{\text{res}} \) that satisfies the previous equations is catalytic (or equivalently \( \tilde{V}_{\text{res}} \) 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

\[
\{ \mathcal{J}_{[(l'+1)\rangle \rightarrow |d, d_k\rangle} \} \]

Figure 4. \( \ln(p^n) \times \ln(p^2) \) diagram for the existence of a restoring unitary. Given a cooling current that transfers population from \( |l_v\rangle \) to \( |(l'+1)\rangle \) (blue arrow), the set of currents \( \{ \mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle} \} \) forms a restoring chain \( ch_{|(l'+1)\rangle \rightarrow |l_v\rangle} \) that restores the populations \( p^l \) and \( p^{l+1} \), while keeping the intermediate populations \( p^l \) 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 \( |k\rangle \) (\( 1 \leq k \leq l' \)) has incoming 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 \( Ch_{|(l'+1)\rangle \rightarrow |l_v\rangle} \), each of which forms a loop with the cooling current \( \mathcal{J}_{[(l'+1)\rangle \rightarrow |l_v\rangle} \). Accordingly, they also have the loop structure shown in Fig. 4. The existence of these chains ensures that there exist unitaries

\[
V_{\text{res},L} = \bigoplus_{k=1}^{l'} V_{[(k+1)\rangle \rightarrow |d, d_k\rangle}^{(2)}
\]

where \( V_{\text{res},L} \) is a “left” restoring unitary, derived from Eq. (20), and \( V_{\text{res},R} \) is a “right” restoring unitary, derived from Eq. (21). Essentially, the partial swaps in \( V_{\text{res},L} \) generate the restoring currents illustrated in Fig. 3(a), and the partial swaps in \( V_{\text{res},R} \) 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, it is worth remarking that either $V_{\text{res},L}$ or $V_{\text{res},R}$ exist only if the chain described in Lemma 1 exists.

A restoring chain of the form \{\((\mathcal{J}_{I(k+1),c})_{1 \leq k \leq l'}\) also allows to reverse the effect that a cooling current \((\mathcal{J}_{I^+},\mathcal{I}_{c^+})_{1 \leq k \leq l'}\) has on the catalyst. The currents in this chain exist iff the inequalities $\frac{p_{kn}^h}{p_{kn}^l} > \frac{p_{l'm}^h}{p_{l'm}^i}$ hold for $l \leq k \leq l'$. Crucially, these inequalities guarantee a unitary $V_{\text{res}} = I_c \otimes V_{hv}$, which restores the catalyst using only the hot object. In particular, a direct sum of appropriate partial swaps

$$V_{hv} = \oplus_{k=l}^{l'} V^{(2)}_{I(k+1),c} \rightarrow |d_k,c_k\rangle$$

generates a uniform restoring chain \{\((\mathcal{J}_{I(k+1),c})_{1 \leq k \leq l'}\) \}, where all the currents have the same magnitude of the cooling current. Since $I_c \otimes V_{hv}$ does not interfere with the cooling effect of $U^{(2)}_{\text{cool}}$, in the rest of the paper we will mainly deal with transformations based on unitaries $U = U_{\text{cool}} \oplus (I_c \otimes V_{hv})$.

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. The corresponding proof is given in Appendix D.

**Theorem 2 (Catalyst size and CNU1 transformations).** Let $d_c$ denote the dimension of the catalyst Hilbert space $\mathcal{H}_c$. For $d_c$ large enough, a CNU1 transformation exists if any of the following conditions hold:

1. The initial state of hot object is not fully mixed, i.e. $p_j^h \neq p_j^l$ for some pair $j,j'$.
2. The Hilbert space of the cold object has dimension $d_c \geq 3$, and $\rho_c$ is not fully mixed.

According to Theorem 2, CNU1 transformations are possible for almost any initial state $\rho_c \otimes \rho_h$. 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 $d_v \rightarrow \infty$, 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 $\rho_c$. In addition, note that the pivotal property behind this advantage is the dimension of the catalyst, whereby any infinite-dimensional system is universal irrespective of its Hamiltonian.

**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 holds we can implement a cooling transition whose restoring effect relies on the non-disturbing (with respect to the cold object) unitary $V_{hv}$. According to the proof given in Appendix C, if $d_v$ is sufficiently large there exists a cooling unitary $U^{(2)}_{\text{cool}}$ that partially swaps the states $|i+1,c\rangle$ and $|i,d_c\rangle$, and a restoring unitary obtained by substituting $l = 1$ and $l' = d_v - 1$ in Eq. (31). The eigenvalues of the state $\rho_c$ are also given in the appendix. These eigenvalues in turn determine the swap intensities of $U^{(2)}_{\text{cool}}$ and $V_{hv}$, by using equations analogous to Eqs. (27) and (28).

Since the only effect of $U = U^{(2)}_{\text{cool}} \oplus (I_c \otimes V_{hv})$ on the cold object is due to the cooling current, the mean energy $\langle H_c \rangle = \text{Tr}[\rho_c H_c]$ decreases by

$$\Delta \langle H_c \rangle = \text{Tr}[(\rho_c' - \rho_c)H_c] = -J_{i+1,c} \rightarrow |i,d_c\rangle (\varepsilon_{i+1} - \varepsilon_i).$$

This cooling transformation is illustrated in Fig. 5(a). Importantly, a two-level system with eigenenergies $\varepsilon_i^h \neq \varepsilon_i^l$ and thermalized at finite temperature $\beta_c > 0$ serves as hot object. Moreover, it can be used to cool down any cold object by transferring population between some pair of consecutive eigenstates $|i\rangle$ and $|i\rangle$, regardless of its size. In Subsection C we will study the optimization of this transformation, when both $\rho_c$ and $\rho_h$ 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 $S(\rho_c' || \rho_c) = -S(\rho_c') - \text{Tr}(\rho_c' \text{ln} \rho_c')$ [Ref. Sagawa], where $S(\rho_c') = -\text{Tr}(\rho_c' \text{ln} \rho_c')$ is the von Neumann entropy of $\rho_c'$. For a thermal state $\rho_c$ at inverse temperature $\beta_c$, 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 $\rho_h$ not fully mixed we can construct a cooling transformation that reduces the value of $S_c$, following Eq. (32).
A CC transformation that cools down a cold qubit, and employs a restoring unitary $U$, can generate a cooling current with respect to the eigenvalues of a state $\rho$. By choosing $i = g$, the cooling current $J_{[(g+1)_c, 1_c)\rightarrow|g_d, d_c\rangle}$ yields the increment

$$\Delta \langle \Pi^g_1 \rangle = \text{Tr}[(\rho_c - \rho_v)\Pi^g_1] = J_{[(g+1)_c, 1_c)\rightarrow|g_d, d_c\rangle}. \quad (33)$$

It is important to stress that $J_{[(g+1)_c, 1_c)\rightarrow|g_d, d_c\rangle}$ always exists if $d_c$ is large enough. On the other hand, suppose that $d_c \geq g + 2$, which means that the cold object possesses more than one excited eigenstate, and that $p^v_d < p^c_{g+1}$, which means that its highest eigenenergy is larger than the first excited eigenenergy $\varepsilon^c_{g+1}$. In this case condition 2 of Theorem 2 holds, and there exists a restoring unitary

$$V_{cv} = \oplus_{k=1}^{d_c-1}V^{(2)}_{[(g+1)_c,(k+1)_c)\rightarrow|d_k, k_c\rangle}.$$ 

This unitary gives rise to a restoring chain $\text{Ch}_{(d_c)\rightarrow|1_c\rangle} = \{J_{[(g+1)_c,(k+1)_c)\rightarrow|d_k, k_c\rangle}\}$, which in turn forms a loop with $J_{[(g+1)_c, 1_c)\rightarrow|g_d, d_c\rangle}$.

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 $\Delta \langle \Pi^g_1 \rangle = \Delta \langle |g_c\rangle\langle g_c| \rangle$. Since $V_{cv}$ transfers population from $(g+1)_c$ to $d_c$, 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 $\rho_{ch}$ 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 $\rho_{ch}$. 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 $\rho_c = p^c_1|1_c\rangle\langle 1_c| + p^c_2|2_c\rangle\langle 2_c|$ and $\rho_h = p^h_1|1_h\rangle\langle 1_h| + p^h_2|2_h\rangle\langle 2_h|$, and satisfy the inequality $p^h_2 \leq p^c_1$ (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 $\rho$, with fixed rank $2 \leq n < \infty$, which has support on a subspace of the infinite Hilbert space $H_c$. 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 \( p_2 \) and \( p_2^3 \) 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 \( \rho_v \) 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 CC unitary is given by

\[
U = U_{cool} \oplus V_{hv},
\]

\[
V_{hv} = \oplus_{k=1}^{n-1} |\psi_{1k(k+1)v}^+\rangle \langle 2k|,
\]

where \( U_{cool} = U_{(2,1,n,v)}^+ \) is the swap between the states \( |2,1,n,v\rangle \) and \( |1,2,n,v\rangle \), and \( V_{1k(k+1)v}^+ \rangle \langle 2k| \) is the swap between the states \( |1,k(k+1)v\rangle \) and \( |2,k,v\rangle \). If \( n = 4 \) and we adopt the view that \( \rho_v \) describes a catalyst of dimension four, the currents generated by \( U \) 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 \( U \) for general values of \( n \).

The maximization of the cooling current \( J_{cool}^\max = J_{12,v}^\max \) also yields catalyst eigenvalues that satisfy

\[
J_k = J_{12,v}^\max \forall 1 \leq k \leq n - 1,
\]

where \( J_k \equiv J_{1k(k+1)v}^+ \langle 2k| \) is the current generated by \( V_{1k(k+1)v}^+ \rangle \langle 2k| \).

From Eq. (36), we find that the maximum cooling current reads (see Appendix E)

\[
J_{cool}^\max = \frac{(1 - r_h) \left[ p_h(1) p_2^c - p_h(2) p_1^c r_h \right]}{(1 - r_h) r_h + (1 - r_h) r_h} p_h(2)^c,
\]

where \( r_h \equiv \frac{2}{p_1^c} \), and we have added parentheses to superscripts to distinguish them from powers. Moreover, the optimal eigenvalues \( \{ p_k \} \) are characterized by the equation

\[
p_h^{n-k} = \left( \frac{r_h}{p_2(2)} \right) \left[ \frac{1}{p_h(1)} - \frac{(1 - r_h) J_{cool}^\max}{1 - r_h} \frac{p_h(n-c)}{p_h(n-v)} \right] p_h(n-v),
\]

1 \( \leq k \leq n - 1 \), with \( p_h(n-v) \) deduced via normalization \( \sum_k p_k(n-v) = 1 \).

Figure 5 shows plots of \( J_{cool}^\max \) and the final ground population \( p_2^n \), for different values of \( p_2^3 \) and catalyst states of ranks \( 2 \leq n \leq 10 \). Each solid curve in Figs. 5(a)-5(c) depicts the maximum cooling current corresponding to a different rank of \( \rho_v \). Moreover, \( J_{cool}^\max \) in Eq. (37) is plotted as a function of \( 0 \leq p_2^3 \leq p_2^3 \), 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 \( p_2^3 \) where \( J_{cool}^\max \) is positive also increases. Since \( J_{cool}^\max < 0 \) 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

\[
\sum_k p_k(n-v) = 1.
\]
$J_{\text{cool}}^{\max} \geq 0$. 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 accessible to small catalysts, characterized by $p_2^c \ll p_3^c$. On the other hand, for $p_3^c = p_2^c$ 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 $p_3^c = p_2^c$).

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.

**Theorem 3 (cooling enhancement with a catalyst).** Let $\rho_c$ be the state of a hot object of dimension $d_h \geq 3$, and $\rho_h$ the state of a cold qubit. If $d_h$ is odd and the largest $d_h/2$ eigenvalues of $\rho_h$ or the smallest $d_h/2$ eigenvalues of $\rho_h$ 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 even and the largest $d_h/2$ eigenvalues or the smallest $d_h/2$ eigenvalues are not fully degenerate, then a large enough catalyst increases the optimal cooling achieved with the hot object alone.

To exemplify the catalytic improvement of cooling consider 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 $\varepsilon_2^h = \varepsilon_3^h = 0$ and $\omega_{2,3}^h = \varepsilon_1^h - \varepsilon_2^h > 0$. For this energy spectrum a thermal state $\rho_h = \frac{1}{\text{Tr}(e^{-\beta_{\varepsilon_2^h} H_h})}$ 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 $\rho_h$, 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 $\rho_h$. The parameter $e^{-\beta_{\varepsilon_2^h}^c} = 0.01$, which also fixes the eigenvalues of $\rho_h$ due to the degeneracy of $\varepsilon_1^h$ and $\varepsilon_2^h$. The blue dashed-dotted curve depicts the ground population of the cold qubit after optimal cooling without catalyst, associated with the cooling current $J_{\text{cool}} = J_{1(2,1_2)} + J_{3(1_2)}$. Moreover, the black solid curve stands for the final population after applying a suitable catalytic and cooling transformation. The left rectangle in Fig. 6(b) is a $\text{ln}(p^c) \times \text{ln}(p^h)$ diagram employed to illustrate the optimal transformation without the catalyst. In this diagram, we keep a small gap $\beta_{\varepsilon_2^c} \omega_{1,2}^c = \beta_{\varepsilon_2^h} (\varepsilon_2^c - \varepsilon_1^c) > 0$ that allows to distinguish the degenerate levels. However, it is indicated that $\beta_{\varepsilon_2^c} \omega_{1,2}^c$ tends to zero, to comply with the degeneracy condition. Assuming that $\varepsilon_1^c = 0$, in the limit $\beta_{\varepsilon_2^c} \omega_{1,2}^c \rightarrow 0$ the condition of cooling without the catalyst amounts to impose that $\text{ln}(p_2^c/p_3^c) = \beta_{\varepsilon_2^c} \omega_{2,3}^c > \varepsilon_2^c = \text{ln}(p_2^c/p_3^c)$. For

\[ e^{-\beta_{\varepsilon_2^c} \omega_{2,3}^c} = 0.01 \]
\(\beta_c \geq \beta_h\), 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 \(\mathcal{U}_{\text{cool}}(\rho_c \otimes \rho_h)\mathcal{U}_{\text{cool}}^\dagger\), where \(\mathcal{U}_{\text{cool}} = \mathcal{U}^{(2,1_k)\oplus[1,3_h]}\) is the swap that maximizes the cooling with 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 \(\ln(p^h) \times \ln(p^v)\) diagram, even if the state \(\mathcal{U}_{\text{cool}}(\rho_c \otimes \rho_h)\mathcal{U}_{\text{cool}}^\dagger\) has not a product form. The only feature that we need to keep in mind is that now the values associated with the columns \(2, 1_k\) and \(1, 3_h\) are, respectively, \(\ln(p_{2}^h p_{3}^v)\) and \(\ln(p_{3}^h p_{2}^v)\) (see Fig. 7(b)). In this way, the depicted currents are generated by the unitary \(\mathcal{U}' = \mathcal{U}_{\text{cool}} \oplus \mathcal{V}_{\text{res}, R} \oplus \mathcal{V}_{\text{res}, L}\), where \(\mathcal{U}'\) and \(\mathcal{V}_{\text{res}, X = R,L}\) are swaps between the connected eigenstates.

It is important to mention that \(\mathcal{U}'\) does not commute with \(\mathcal{U}_{\text{cool}}\), since \([\mathcal{U}_{\text{cool}}' \mathcal{U}_{\text{cool}}] \neq 0\). Therefore, the total transformation \(\mathcal{U}'\mathcal{U}_{\text{cool}}\) 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 \(\mathcal{J}'_{\text{res}} = \mathcal{J}'_{\text{res}, L} + \mathcal{J}'_{\text{res}, R}\), where
\[
\begin{align*}
\mathcal{J}'_{\text{res}, L} &= p_1^h p_2^v - p_2^h p_1^v, \\
\mathcal{J}'_{\text{res}, R} &= p_3^h p_2^v - p_2^h p_3^v.
\end{align*}
\]

From the condition \(\mathcal{J}'_{\text{cool}} = \mathcal{J}_{\text{res}}\) (uniform loop) and the degeneracy \(p_1^h = p_2^h\), it follows that
\[
\mathcal{J}'_{\text{cool}} = \left(\frac{p_3^h p_1^v - p_1^h p_3^v}{1 + p_2^h p_2^v} p_2^v p_2^h\right).
\]

Remarkably, Fig. 7(a) shows that for low temperatures (\(\beta_c\) large) the increment of \(p_1^h\) 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 \(\mathcal{J}_{\text{cool}}\) and \(\mathcal{J}_{\text{cool}} + \mathcal{J}'_{\text{cool}}\), where \(\mathcal{J}_{\text{cool}} + \mathcal{J}'_{\text{cool}}\) is the current obtained from the total transformation \(\mathcal{U}'\mathcal{U}_{\text{cool}}\). In these plots \(e^{-\beta_c \varepsilon_2^h}\) is fixed, and we vary instead the parameter \(e^{-\beta_h \varepsilon_3^h}\). The condition \(\beta_c \varepsilon_2^h > \beta_h \varepsilon_3^h\) (for cooling without the catalyst) implies that the maximum value of \(e^{-\beta_h \varepsilon_3^h}\) must coincide with \(e^{-\beta_c \varepsilon_2^h}\). When \(e^{-\beta_h \varepsilon_3^h}\) increases, the hot object is hotter and consequently the cooling current \(\mathcal{J}_{\text{cool}}\) decreases, reaching its minimum value \(\mathcal{J}_{\text{cool}} = 0\) at \(e^{-\beta_h \varepsilon_3^h} = e^{-\beta_c \varepsilon_2^h}\). 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.

**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 \(e^{-\beta_h \varepsilon_3^h}\) has the fixed values 0.1 (a), 0.4 (b), and 0.8 (c). The plotting variable \(e^{-\beta_c \varepsilon_2^h}\) is increased until cooling without the catalyst is not possible, where \(\mathcal{J}_{\text{cool}} = 0\).

**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, 10, 14]. However, the restoration of the catalyst per se is not mandatory to achieve 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 be seen as environments of big size. 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 \( \beta \) and have identical energy spectrum. Therefore, the Hamiltonians of the \( i \)th cold and hot qubits are respectively \( H_c^{(i)} = \ket{1_c}\bra{1_c} \) and \( H_h^{(i)} = \ket{1_h}\bra{1_h} \). The total Hamiltonian for the \( X = C,H \) group is \( H_X = \sum_{i=1}^{N_X} \ket{1_{xc,ci}}\bra{1_{xc,ci}} \), and the global initial state is a product of thermal states \( \rho_C H = \rho_C \otimes \rho_H \), where \( \rho_X = e^{-\beta H} / Z_X \). Assuming that the total number of qubits \( N = N_c + N_h \) is fixed, we now describe two cooling strategies, illustrated in Fig. 9.

1. Many-body cooling (MBC) strategy: subsets of \( 2 \leq k \leq N_h \) 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 \( k \geq 2 \), 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 once 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 \( \bigotimes_{i=1}^{k+1} B_i \), where \( B_i \) is a non-trivial (i.e. different from the identity) operator on the Hilbert space of the \( i \)th qubit. On the other hand, the CC strategy is based on the repeated application of the unitary \( \mathcal{U} \) in Eq. (34), 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 between 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 follow-
ing 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

$$Q_C \equiv -\Delta \langle H_C \rangle = \sum_{i=1}^{n_c} \Delta p_1^{(i)}, \quad (42)$$

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” $\xi_{cool}^{(k)}$ as

$$\xi_{cool}^{(k)} = \frac{Q_C^{(k)}}{\sum_{k \in K} k}, \quad (43)$$

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 (43), we can express the total extracted heat as

$$Q_C = \sum_{k \in K} Q_C^{(k)} = \sum_{k \in K} \xi_{cool}^{(k)} k, \quad (44)$$

where $K = \{k_0, k_1, \ldots\}$ describes a certain partition of the hot group into cooling subsets. In particular, we note that $\sum_{k \in K} k = N_h$, 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

$$Q_C \leq \left( \max_{k \in K} \xi_{cool}^{(k)} \right) \sum_{k \in K} k = \left( \max_{k \in K} \xi_{cool}^{(k)} \right) N_h. \quad (45)$$

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

$$\xi_{cool}^{(k)} \leq \xi_{cool}^{(2)} = \left( 1 - \frac{2p_2^\xi}{2} \right) p_1^\xi p_2^\xi, \quad (46)$$

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

Although our conjecture and Eq. (45) 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. (45) involves values of $k$ characterizing a specific partition $K$, 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 (45) is saturated with the maximum coefficient \( \xi_{\text{cool}}^{(2)} \). More generally, we have that

\[
\max_K Q_C = \xi_{\text{cool}}^{(2)} N_h, \text{ if } N_c \geq \frac{N}{3}, \quad (47)
\]

\[
2\xi_{\text{cool}}^{(2)} N_c \leq \max_K Q_C \leq \xi_{\text{cool}}^{(2)} N_h, \text{ if } N_c \leq \frac{N}{3} - 1, \quad (48)
\]

where Eq. (47) 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. (48) generalizes the bound (45) 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 (48) is in general strict, 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 \( Q_C^{(CC)} \) 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 \( p^{(i)}_1 = p^{(j)}_1 \) 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 (37), with \( n = 2 \) and \( p^0_2 = p^2 = \frac{\gamma - \beta}{1 + \gamma - \beta} \). The total extracted heat is thus

\[
Q_C^{(CC)} = n_c \left( \frac{1 - 2p^2}{1 + 2p^1_1p^2_2} \right) p^1_1p^2_2, \quad (49)
\]

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. (49) and the aforementioned conditions it follows that

\[
Q_C^{(CC)} = \begin{cases} 
N_c \left( \frac{1 - 2p^2}{1 + 2p^1_1p^2_2} \right) p^1_1p^2_2, & \text{if } N_c \leq \frac{N}{2}, \\
(N - N_c) \left( \frac{1 - 2p^2}{1 + 2p^1_1p^2_2} \right) p^1_1p^2_2, & \text{if } N_c \geq \frac{N}{2} + 1,
\end{cases} \quad (50)
\]

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

\[
\gamma = \frac{Q_C^{(CC)}}{\max_K Q_C}, \quad (51)
\]

where both the numerator and the denominator refer to a fixed number of cold qubits and population \( p^2_2 \) (which in turn characterizes the inverse temperature \( \beta \)). For \( N_c \leq N/3 - 1 \), the lower bound in Eq. (48) and Eq. (51) lead to

\[
\gamma_{N_c, \leq N/3 - 1} \leq \frac{1 - 2p^2}{2\xi_{\text{cool}}^{(2)}(1 + 2p^1_1p^2_2)} = \frac{1}{1 + 2p^1_1p^2_2}. \quad (52)
\]

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

If \( N_c \geq N/3 \), Eqs. (47) and (50) yield the following expressions for \( \gamma \):\[ \gamma_{N/3 \leq N_c \leq N} = \left( \frac{2}{1 + 2p^1_1p^2_2} \right) \frac{N_c}{N - N_c} \geq \frac{1}{1 + 2p^1_1p^2_2}. \quad (53) \]

\[
\gamma_{N/3 + 1 \leq N_c \leq N} = \frac{2}{1 + 2p^1_1p^2_2}. \quad (54)
\]

The lower bound at the r.h.s. of Eq. (53) 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

\[
N_c > \frac{1 + 2p^1_1p^2_2}{3 + 2p^1_1p^2_2}. \quad (55)
\]

The r.h.s. of such inequality varies between \( 1/3 \), for \( p^2_2 = 0 \), and \( 3/7 \), for \( p^2_2 = 1/2 \). Accordingly, in the regime \( 3N/7 < N_c \leq N/2 \) the performance ratio (53) satisfies \( \gamma_{3N/7 \leq N_c \leq N/2} > 1 \). For the remaining interval \( N/3 \leq N_c \leq 3N/7 \), Eq. (55) provides an upper bound on \( p^2_2 \) to have \( \gamma > 1 \).

On the other hand, from Eq. (54) 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 \( \beta \). We also stress that in the definition of \( \gamma \) the heat \( Q_C \) is optimized with respect to all the many-body interactions involving \( N_h \) hot 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 \( N_e \)) where the CC advantage takes place, irrespective of the inverse temperature \( \beta \). 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. (55). In particular, we note that in the limit \( \beta \to \infty \) the fraction \( N_e/N \) 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 \( \gamma \) is provided in Fig. 11, following Eqs. (52)-(54). 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 \( n_c < N_e \) qubits can be cooled down for a given value of \( N_e \) (e.g. if \( N_e \geq N/2 \)), the remaining \( N_e - n_c \) qubits are not heated up either. This condition guarantees that the temperature of all the \( N_e \) qubits remains below a certain threshold (in this case below \( \beta^{-1} \)), and may be important for some applications. In other words, it provides a justification for choosing a fixed value of \( N_e \). On the other hand, one may alternatively be interested in optimizing the extracted heat with respect to \( N_e \). In the case of the CC strategy it readily follows from Eq. (50) that such a maximum is attained for \( N_e = N/2 \). Regarding the MBC strategy, we can resort to the bound \( \text{max}_K Q_C \leq \xi_{\text{cool}} Q_h \) (cf. Eqs. (47) and (48)) to determine if there are values of \( N_c \) such that \( \text{max}_K Q_C \) potentially surpasses the maximum \( Q_C^{(\text{CC})} \). By writing \( N_h = N - N_c \), we have that \( \text{max}_K Q_C > \text{max}_N Q_C^{(\text{CC})} \) only if

\[
\xi_{\text{cool}}^2 \left( N - N_c \right) > \frac{N}{2} \left( 1 + 2p_1^c p_2^c \right) p_1^c p_2^c.
\]

which after a simple algebra leads to the inequality

\[
\frac{N_c}{N} < \frac{2p_1^c p_2^c}{1 + 2p_1^c p_2^c}.
\]

The maximum value of the fraction \( N_c/N \) that satisfies this bound is achieved in the high temperature limit \( \beta \to 0 \), where the bound tends to \( 1/3 \). Conversely, as \( \beta \) 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 \( N_h \)) 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. (46) and (47) we have that, for any temperature,

\[
\frac{\text{max}_N Q_C^{(\text{CC})}}{\text{max}_{N_c \geq N/3} Q_C} = \frac{3}{2} \left( 1 + 2p_1^c p_2^c \right) \geq 1.
\]

VII. CATALYTIC THERMOMETRY

In this section we study an example where a catalyst is applied for precision enhancement in thermometry [51], where the goal is to estimate the temperature of a certain environment at thermal equilibrium. Let \( \rho_c = \frac{e^{-\beta H_e}}{Tr(e^{-\beta H_e})} \) denote the state of an environment with Hamiltonian \( H_e = \sum_j \epsilon_j |j_e\rangle \langle j_e| \), equilibrated at inverse temperature \( \beta \). Essentially, a temperature estimation consists of assigning temperature values \( T_i \) to the different outcomes of a properly chosen observable \( O \). In this way, the set \( \{T_i\} \) defines a temperature estimator \( T \), and the precision is assessed through the estimation error

\[
\sqrt{\left< (T - \bar{T})^2 \right>} \equiv \sqrt{\sum_i p_i (T_i - \bar{T})^2},
\]

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 [49]. 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 \( O = H_e \). 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
auxiliary 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 \( \varrho \)). Moreover, the catalyst is also a two-level system in the initial state \( \rho_c \). 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 [Refs.]. In particular, the Cramer-Rao bound limits the precision attained with this kind of estimators. If \( \langle \hat{T} \rangle = T \) it follows that \( \langle (\hat{T} - T)^2 \rangle = \text{Var}(\hat{T}) \), where \( \text{Var}(\hat{T}) = \langle T^2 \rangle - \langle T \rangle^2 \) is the variance of \( \hat{T} \). Moreover, it can be shown [51] that if the temperature to be estimated belongs to a small interval \( \langle T - \delta T, T + \delta T \rangle \), the estimation error using the observable \( O \) reads

\[
\sigma(\hat{T}) = \sqrt{\frac{\text{Var}(O)}{\partial_T \langle O \rangle}},
\]

where \( \text{Var}(O) = \langle O^2 \rangle - \langle O \rangle^2 \) and \( \partial_T \langle O \rangle = \frac{\partial}{\partial T} \langle O \rangle \).

For the sake of covariance, 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 \( O_P = \alpha_1 |1_p\rangle\langle 1_p| + \alpha_2 |2_p\rangle\langle 2_p| \) describes a projective measurement on the probe, with eigenvalues \( \alpha_1 \) and \( \alpha_2 \). Information about \( \beta \) is encoded in the probe state \( \varrho' = \text{Tr}_c \left[ U_{P_e} (\varrho \otimes \rho_c) U_{P_e}^\dagger \right] \), which results after a unitary evolution \( U_{P_e} \) that couples the probe with the environment. It is straightforward to check that in this case the estimation error reads

\[
\sigma'(\hat{\beta}) = \sqrt{\frac{\partial\beta \rho_p}{T \rho_p}} \left| \frac{\partial\beta \rho_p}{T \rho_p} \right|, \tag{60}
\]

where \( \rho'_p = \text{Tr}[|1_p\rangle\langle 1_p| \varrho' \rho_e] \).

The ratio in the r.h.s. of Eq. (60) constitutes the figure of merit in our analysis. On the one hand, under certain conditions one can find a unitary \( U_{P_e} \) that minimizes the product \( \rho'_p \rho'_p \), and at the same time maximizes the quantity \( \partial\beta \rho_p \). In such a case, the inequality

\[
\min_{U_{P_e}} \sigma'(\hat{\beta}) \geq \min_{U_{P_e}} \frac{\sqrt{\rho'_p \rho'_p}}{\max_{U_{P_e}} \partial\beta \rho_p}, \tag{61}
\]

guarantees that the same operation minimizes the error \( \sigma'(\hat{\beta}) \). This implies that if a unitary \( U_{P_e} \) 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 (61) is saturable it is possible to apply a catalytic transformation such that

\[
\frac{\sqrt{\rho'_p \rho'_p}}{\partial\beta \rho_p} < \min_{U_{P_e}} \left( \frac{\sqrt{\rho'_p \rho'_p}}{\partial\beta \rho_p} \right), \tag{62}
\]

where \( \partial\beta \rho_p = \text{Tr}[|1_p\rangle\langle 1_p| \varrho'' \rho_e] \), and \( \varrho'' \) 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 \( \varrho \rightarrow \varrho'' \) is implemented by a global unitary of the form \( U' U_{P_e} \). Importantly, the optimal \( U_{P_e} \) and \( U' \) satisfy \( U_{P_e} = U_{cool} \) and \( U' = U'_{cool} \), being \( U_{cool} \) and \( U' \) the cooling and catalytic cooling unitaries defined in Sect. V-D. This is not a coincidence, as we show below that the bound (61) can be saturated by maximally cooling the probe with the environment. In addition, the fact that \( U \) yields a cooling enhancement for the probe (which here takes the role of the cold qubit) implies that it also reduces the final population \( \rho'_p \rho'_p \). Since we also show that \( \partial\beta \rho_p > \partial\beta \rho_p \), Eq. (62) follows.

### B. Maximization of the population sensitivity in terms of passivity

In what follows we will refer to \( \partial\beta \rho_p \) as the “population sensitivity”, as it quantifies how the final population \( \rho'_p \) varies with respect to temperature changes. Defining \( \rho_{P_e} = \varrho \otimes \rho_e \) and \( \partial\beta \rho_{P_e} = U_{P_e} \partial\beta \rho_e U_{P_e}^\dagger \), we can use the fact that \( U_{P_e} \) is independent of \( \beta \) to write the population sensitivity as

\[
\partial\beta \rho'_p = \partial\beta \text{Tr}[|1_p\rangle\langle 1_p| \varrho' \rho_e] = \text{Tr}[|1_p\rangle\langle 1_p| U_{P_e} \partial\beta \rho_e U_{P_e}^\dagger]. \tag{63}
\]

The operator \( \partial\beta \rho_{P_e} = \varrho \otimes \partial\beta \rho_e \) has real eigenvalues \( \{ \lambda_{i,j} \} \equiv \{ p_i, \lambda^c \} \), where

\[
\lambda^c = \partial\beta \rho^c = p_j^c (\langle H \rangle - \varepsilon_j), \tag{64}
\]
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) $\ln(p) \times \ln(p')$ diagram for the initial probe-environment state $\rho \otimes \rho_e$. 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 $U_{1|p_3 \beta \rightarrow 2|p_1 \varepsilon}$ is an optimal unitary $U^\text{op}_{pe}$ that minimizes the error $\sigma'(\beta)$.

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

Since $\rho_{pe}$ is hermitian, the operator $A = \partial_\beta \rho_{pe} - \min \{ \lambda_{i,j}^P \}$ is positive semidefinite. In this way, we can rewrite Eq. (63) in the form

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

where $\frac{A}{\text{Tr}(A)}$ represents a density matrix (i.e. its eigenvalues 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 $\frac{A}{\text{Tr}(A)}$ in a passive state with respect to $-|1_P\rangle \langle 1_P|$. The definition of $\frac{A}{\text{Tr}(A)}$ 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 $\{|1_P^\text{op}_{pe}\rangle\}$. Consequently, the application of such a permutation yields an operator

$$\partial_\beta \rho_{pe} = |1_P\rangle \langle 1_P| \otimes \sum_j \lambda^P_{1,j} |j_e\rangle \langle j_e|$$

$$+ |2_P\rangle \langle 2_P| \otimes \sum_j \lambda^P_{2,j} |j_e\rangle \langle j_e|,$$

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

C. Results

For an initial state $\rho$ such that $\beta(\varepsilon_3 - \varepsilon_2) > \ln(p_1/p_2)$ (cf. Fig. 12(b) and the cooling transformation $U_{\text{cool}}$ in Sect. V-D), the swap $U_{1|p_3 \beta \rightarrow 2|p_1 \varepsilon}$ 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 $U_{1|p_3 \varepsilon \rightarrow 2|p_1 \beta}$ is also a permutation that satisfies Eq. (65). This implies that in such a case $U_{1|p_3 \varepsilon \rightarrow 2|p_1 \beta}$ is an optimal unitary $U^\text{op}_{pe}$ that saturates the bound (61). However, we stress that

Figure 13. Estimation error $\sigma(\beta)$ 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 $\sigma'(\beta)$ and $\sigma''(\beta)$, 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 $e^{-\beta\varepsilon_3} < p_2/p_1$, where $\sigma'(\beta)$ is the minimum estimation error under general unitary evolutions that act jointly on the probe and the environment.
while this swap always maximizes the population sensitivity $\partial_\beta p'_1$, it also minimizes the error $\sigma'(\hat{\beta})$ as long as cooling is possible with the environment, see Fig. 12(b). Otherwise, any unitary $U_{\rho_e}$ 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 $U_{[1_p,3_e]}\mapsto[2_p,1_e]$, for three different initial states $\rho$. 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 $\partial_\beta p''_1 = Tr[|1_P\rangle\langle1_P| Tr_c \partial_\beta \rho''_P c]$, where $\rho''_P = U'(\rho_P)U'^\dagger$.

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,

$$\partial_\beta p'_1 = Tr_P [|1_P\rangle\langle1_P| Tr_c \partial_\beta \rho' P c] = p_1 (\lambda'_1 + \lambda'_2) + p_2 \lambda'_1,$$

where $Tr_c \partial_\beta \rho' P c$ is computed from Eq. (65). 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

$$\partial_\beta p''_1 = Tr_P [|1_P\rangle\langle1_P| Tr_c \partial_\beta \rho''_P c] = \partial_\beta p'_1 + p_2 [p'_1 (\lambda'_2) - p'_2 (\lambda'_1)],$$

where the contribution $p_2 [p'_1 (\lambda'_2) - p'_2 (\lambda'_1)]$ is due to the swap that generates the cooling current. Crucially, $\partial_\beta p'_1 > \partial_\beta p''_1$ iff this contribution is positive. Noting that $\lambda'_1 \geq 0$ (cf. Eq. (64)), it follows that the catalytic transformation increases the sensitivity iff $\lambda'_2 > 0$ and

$$\frac{p'_1}{p'_2} > \frac{\lambda'_1}{\lambda'_2}.$$

The degeneracy condition $\epsilon_1 = \epsilon_2$ implies that $\lambda'_1 = \lambda'_2$, and consequently this inequality holds for any catalyst whose initial 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 $\sigma''(\hat{\beta}) = \sqrt{p''_1 p''_2}$ 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 $e^{-\beta \epsilon_2} \leq p_2/p_1$, 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. (62) is satisfied, i.e. $\sigma''(\hat{\beta}) < \min_{U_{\rho_e}}\sigma'(\hat{\beta})$. 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. 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 limitations 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 thermom-
etry. This includes to further study the roles of initially mixed probes and the sizes of the involved systems. We have seen that the dimension of the catalyst is crucial to bypass thermodynamic restrictions imposed by the finiteness of the other systems. This observation is related to a question recently posed in [15], where the authors ask if certain transformations achieved with multipartite catalysts can be performed with a single, and sufficiently large catalyst. The findings presented here may contribute to elucidate this puzzle, since they are based on single-copy catalysts. To that end, the first step is to examine how our results can be extended to include the possibility of energy-preserving interactions, which is the framework considered in [15]. We also remark that the characterization of catalytic transformations provided here is valid for systems that do not necessarily start in thermal states. Hence, it can be useful for studying the role of catalysts in scenarios beyond thermodynamics. Finally, we hope that our explicit description of catalytic transformations paves the way to experimental realisations in the near future.

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Appendix A: Proof of Theorem 1

Consider a system in an initial state \( \rho_s \), 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 \( \rho_c \), with eigenvalues \( \{p_i^c\}_{1 \leq i \leq d_c} \). In this appendix we demonstrate that the conditions:

\[
\frac{p_i^c}{p_i^c} > \frac{p_i^s}{p_i^s} > 1, \quad (A1)
\]

for some \( 1 \leq i \leq d_s - 1 \) and \( 1 \leq l, l' \leq d_c - 1 \); and, for \( l \leq k \leq l' \),

\[
\frac{p_i^c}{p_j^c} > \frac{p_k^c}{p_k^c} > 1, \quad (A2)
\]

\[
or \quad \frac{p_{i+1}^c}{p_d} > \frac{p_{k+1}^c}{p_d} > 1, \quad (A3)
\]

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

**Sufficiency.** Equation (A1) is equivalent to

\[
\frac{p_i^c}{p_i^s} > \frac{p_i^s}{p_i^s},
\]

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

If Eq. (A2) holds then there is a restoring chain \( \{J_{(l+1),(l') \rightarrow (l,l')}\}_{l \leq l' \leq l'} \) such that \( J_{(l+1),l') \rightarrow (l,l')} \) cancels the population increment (on the catalyst) generated by \( J_{(l+1),l') \rightarrow (l,l')} \), and \( J_{(l+1),l') \rightarrow (l,l')} \) 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 \( J_{(l+2),l') \rightarrow (l+1,l')} \) and \( J_{(l+1),l') \rightarrow (l+2,l')} \) on the state \( |(l+2)\rangle \) cancel each other. Since this restoring chain acts within the subspace span \( \{\ket{j_s}\}_{l+1 \leq j \leq l'} \), 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 \( \{J_{(l+1),(l') \rightarrow (l,l')}\}_{l \leq l' \leq l'} \) that cancels the effect of \( J_{(l+1),l') \rightarrow (l,l')} \) (on the catalyst) through the currents \( J_{(l+1),l') \rightarrow (l,l')} \) and \( J_{(l+1),(l') \rightarrow (l,l')} \), 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 span \( \{\ket{j_s}\}_{l+1 \leq j \leq l'} \).

**Necessity.** If Eq. (A1) does not hold, then there is no cooling current \( J_{(l+1),(l') \rightarrow (l,l')} \) and therefore no CNU1 transformation can be implemented (keeping in mind that by definition CNU1 transformations include a cooling current). On the other hand, let us see that if Eqs. (A2), (A3) 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 \( p_i^c > p_i^s \) for some \( 1 \leq i \leq d_s - 1 \) and \( 1 \leq l, l' \leq d_c - 1 \); and, for \( l \leq k \leq l' \),

\[
\frac{p_i^c}{p_j^c} < \frac{p_k^c}{p_k^c} < 1, \quad (A4)
\]

\[
or \quad \frac{p_{i+1}^c}{p_d} < \frac{p_{k+1}^c}{p_d} < 1, \quad (A5)
\]

are necessary and sufficient for a CNU1 transformation on the system. The right-most inequalities follow from the convention \( p_j^c \geq p_{j+1}^c \) and \( p_k^c \geq p_{k+1}^c \) for all \( j \). By choosing \( \rho_s = \rho_c \otimes \rho_h \), the necessary and sufficient conditions (19)-(21) for a CNU1 transformation on \( \rho_c \) are obtained.
ing currents connect eigenstates of $\rho_s$, with maximum and minimum eigenvalues restricted by $G_s$. In other words, let $I_q = \{j_0, j_1, \ldots\}$ be the set of indices that label the eigenstates spanning the subspace $G_s$, i.e., $G_s = \text{span}\{\{j\}\}_{j \in I_q}$. Taking into account that $p_{ij}^{c+1} \leq p_{ij}^c$, a restoring unitary acting on $G_s \otimes \text{span}\{\{k\}\}_i \leq k \leq k'$ exists iff the chain $\{j_{\min_j \in I_q}\} \leq k \leq k' \leq j_{\max_j \in I_q}$ exists in the left case of the partial sum, $G_s = \text{span}\{\{j\}\}_{j \leq j' \leq j'}$, and the corresponding chain is $\{j_{\max_j \in I_q}\} \leq k \leq k' \leq j_{\min_j \in I_q}$.

Since we are also assuming that the right chain $\{j_{\max_j \in I_q}\} \leq k \leq k' \leq j_{\min_j \in I_q}$ does not exist (i.e. that Eq. (A3) is also violated), there is no restoring unitary in the subspace $\text{span}\{\{j\}\}_{j = 1 \leq j \leq d} \otimes \text{span}\{\{k\}\}_i \leq k \leq k'$ either.

The preceding discussion implies that the only way to have a catalytic transformation is by means of a restoring unitary on a subspace $\text{span}\{\{j\}\}_{j \in I_q} \otimes \text{span}\{\{k\}\}_i \leq k \leq k'$, such that $\max_j \in I_q \geq i + 1$ and $\min_j \in I_q \leq i$. Note that a subspace determined by the single condition $\max_j \in I_q \geq i + 1$ ($\min_j \in I_q \leq i$) includes the possibility of a right (left) restoring unitary, which has already been discarded. If $\max_j \in I_q \geq i + 1$ and $\min_j \in I_q \leq i$, at least one of the partial swaps composing the restoring unitary (cf. Eq. (8)) must connect an eigenstate of $\rho_s \otimes \rho_c$ belonging to $\text{span}\{\{j\}\}_{j \leq j' \leq j'} \otimes \text{span}\{\{k\}\}_{i \leq k \leq k'}$, with one eigenstate belonging to $\text{span}\{\{j\}\}_{i \leq i' \leq d} \otimes \text{span}\{\{k\}\}_{i \leq k \leq k'}$. 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 $\text{span}\{\{j\}\}_{j \leq j' \leq j'}$ towards $\text{span}\{\{j\}\}_{j = 1 \leq j \leq d}$. Let us denote this current simply as $J$. Since the cooling current $J_{([i + 1], i_v)} \rightarrow |i_v, (l + 1)\rangle$ transfers population in the opposite direction, we have that

$$\delta \left( \sum_{j=1}^{i} p_{ij}^c \right) \leq -J + J_{([i + 1], i_v)} \rightarrow |i_v, (l' + 1)\rangle = 0,$$

where the condition $J = J_{([i + 1], i_v)} \rightarrow |i_v, (l' + 1)\rangle$ is necessary for the restoring unitary to generate a uniform loop.

In this way, we conclude that the potential increment of the partial sum $\sum_{j=1}^{i} p_{ij}^c$ 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))

$$p_s^c \rightarrow p_{da}^c p_{hc}^c,$$

$$p_{i+1}^c \rightarrow p_{d(a+1)}^c p_{hc}^c,$$

$$p_a^c \rightarrow p_{da}^c p_{hc}^c,$$

$$p^c_{d(a+1)} \rightarrow p_{d(a+1)}^c p_{hc}^c,$$

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

**Appendix B: Population currents within the catalyst**

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

$$\text{Tr}[|k_v\rangle\langle k_v| \rho \rho^U] = \sum_{i,j \in \mathcal{L}_+} (p^{ch}_{ijk} + J_{|i_v j_h k_v\rangle \rightarrow |i_v j_h k_v\rangle})$$

$$+ \sum_{i,j \in \mathcal{L}_-} (p^{ch}_{ijk} - J_{|i_v j_h k_v\rangle \rightarrow |i_v j_h k_v\rangle}),$$

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

$$\text{Tr}[|k_v\rangle\langle k_v| \rho \rho^U] - \rho = \sum_{i,j \in \mathcal{L}_+} J_{|i_v j_h k_v\rangle \rightarrow |i_v j_h k_v\rangle}$$

$$- \sum_{i,j \in \mathcal{L}_-} J_{|i_v j_h k_v\rangle \rightarrow |i_v j_h k_v\rangle},$$

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 $p^{ch}_{ijk}$ increases iff there is current $J_{|i_v j_h k_v\rangle \rightarrow |i_v j_h k_v\rangle}$, and it decreases iff there is a current $J_{|i_v j_h k_v\rangle \rightarrow |i_v j_h k_v\rangle}$. This unambiguously determines 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 $\{J_{1,1}\rightarrow |i_d d_k\rangle \rightarrow |i_d d_k\rangle \rightarrow \cdots \rightarrow |i_d d_k\rangle \rightarrow 1\}$ does not exist, which means that $J_{1,1}\rightarrow |i_d d_k\rangle \rightarrow |i_d d_k\rangle = 0$ for some $l \leq k \leq l'$. Let us see that this condition implies the non existence of a general chain $\chi_{i'\rightarrow i'}$. First, note that

$$J_{1,1}\rightarrow |i_d d_k\rangle \rightarrow |i_d d_k\rangle \geq J_{i_d, i_d} \rightarrow |i_d j_{l'} k_{l'}\rangle$$

$$\Leftrightarrow (p^{ch}_{ik} p^{ch}_{ik}) P_{K+1} + (p^{ch}_{ik} p^{ch}_{ik}) P_{K+1} \geq (p^{ch}_{ik} p^{ch}_{ik}) P_{K+1} - (p^{ch}_{ik} p^{ch}_{ik}) P_{K+1},$$

where $1 \leq i, i' \leq d_c$ and $1 \leq j, j' \leq d_h$. This inequality follows directly from the inequalities $p^{ch}_{ik} P_{K+1} \geq p^{ch}_{ik} P_{K+1}$ and $p^{ch}_{ik} P_{K+1} \leq p^{ch}_{ik} P_{K+1}$. Similarly, for $k' \geq K + 1$ and $k'' \leq K$ it holds that

$$J_{i_d, i_d} \rightarrow |i_d j_{l'} k_{l'}\rangle \geq J_{i_d, i_d} \rightarrow |i_d j_{l'} k_{l'}\rangle$$

$$\Leftrightarrow p^{ch}_{ik} (p^{ch}_{ik}) P_{K+1} - p^{ch}_{ik} (p^{ch}_{ik}) P_{K+1} \geq p^{ch}_{ik} (p^{ch}_{ik}) P_{K+1} - p^{ch}_{ik} (p^{ch}_{ik}) P_{K+1},$$

which follows from the inequalities $p^{ch}_{ik} P_{K+1} \geq p^{ch}_{ik} P_{K+1}$ and $p^{ch}_{ik} P_{K+1} \leq p^{ch}_{ik} P_{K+1}$. In this way, we can join the previous inequalities to obtain:

$$J_{1,1}\rightarrow |i_d d_k\rangle \rightarrow |i_d d_k\rangle \geq J_{i_d, i_d} \rightarrow |i_d j_{l'} k_{l'}\rangle \geq |i_d j_{l'} k_{l'}\rangle \rightarrow |i_d j_{l'} k_{l'}\rangle \geq \cdots \rightarrow |i_d j_{l'} k_{l'}\rangle \rightarrow 1.$$

Therefore, $J_{1,1}\rightarrow |i_d d_k\rangle \rightarrow |i_d d_k\rangle = 0$ implies that $J_{i_d, i_d} \rightarrow |i_d j_{l'} k_{l'}\rangle = 0$ for $k' \geq K + 1$ and $k'' \leq K$ (and
The key ingredient is to show that, for large enough $d_v$, one can always find a cooling current and a suitable restoring chain that forms a loop with the cooling current. This amounts to find suitable catalyst eigenvalues $\{p_k\}_{1 \leq k \leq d_v}$ that enable the construction of the loop. By replacing $t$ by 1 and $t'$ by $d_v - 1$ in Eq. (19), we get the maximum ratio $\frac{p_k}{p_{k+1}}$ between catalyst populations. Clearly, this ratio can be increased as much as wished by choosing a sufficiently pure catalyst, which ensures that a cooling current exists. On the other hand, Eqs. (20) and (21) can be satisfied for all $1 \leq k \leq d_v - 1$ only if the state $\rho_n$ is full rank. This implies that $\rho_n$ must be pure enough to have a cooling current, but at the same time mixed enough to have a restoring chain. Here is where its dimension comes into play.

Let us assume that $\frac{p_k}{p_{k+1}} = \frac{p_1}{p_2}$ for all $1 \leq k \leq d_v - 1$. In this way,

$$\frac{p_k}{p_{d_v}} = \prod_{k=1}^{d_v-1} \frac{p_k}{p_{k+1}} = \left( \frac{p_1}{p_2} \right)^{d_v-1}. \quad (D1)$$

Since $\frac{p_1}{p_2} > 1$ by construction, for $d_v$ large enough we can have a cooling current guaranteed by the inequality

$$\left( \frac{p_1}{p_2} \right)^{d_v-1} = \left( \frac{p_1}{p_2} \right)^{d_v-1} \geq \left( \frac{p_k}{p_{k+1}} \right) \geq \left( \frac{p_1}{p_2} \right)^{d_v-1}.$$  

This is true regardless of the eigenvalues of $\rho_c$ and $\rho_h$.

Now suppose that condition 1 of Theorem 2 holds. In this case, $\frac{p_k}{p_{d_v}} > \frac{p_k}{p_{d_v+1}} = \frac{p_1}{p_2}$ for all $1 \leq k \leq d_v - 1$ (where the assumption $\frac{p_k}{p_{k+1}} = \frac{p_1}{p_2}$ is used again).

Since $\frac{p_1}{p_2} > 1$ (by hypothesis of condition 1), we can choose $p_k$ small enough to obtain a restoring chain $\{J_{1,(k+1),1} \rightarrow \{d_v \}}$ whose existence is guaranteed by the inequality $\frac{p_k}{p_{d_v}} > \frac{p_k}{p_{d_v+1}}$. Noting that this restoring chain only involves the hot object, the corresponding restoring unitary is of the form $V_{hr}$.

If condition 2 holds, a CNU1 transformation exists even if $\rho_h$ is fully mixed. Since $\rho_c$ is not fully mixed we can always choose a value of $i$ such that $\frac{p_i}{p_{i+1}} > 1$ or $\frac{p_i}{p_{d_v}} > 1$. This suffices to have a left restoring chain described by the inequalities $\frac{p_i}{p_{i+1}} > \frac{p_i}{p_{k+1}} = \frac{p_1}{p_2}$, or a right restoring chain described by the inequalities $\frac{p_i}{p_{i+1}} > \frac{p_i}{p_{k+1}} = \frac{p_1}{p_2}$, where once again $\frac{p_1}{p_2}$ has to be sufficiently small. Moreover, any of such chains only involves the cold object, which implies that the corresponding restoring unitaries have the form $V_{cv}$.

We stress that in both cases the ratio $\frac{p_v}{p_{k+1}}$ can be adjusted to enable the existence of the restoring chains, while at the same time a sufficiently large dimension $d_v$ always ensures the existence of the cooling current. The condition $\frac{p_v}{p_{k+1}} = \frac{p_1}{p_2}$ yields the relation

$$\frac{p_v}{p_k} = \prod_{i=1}^{k-1} \frac{p_i}{p_{i+1}} = \left( \frac{p_1}{p_2} \right)^{k-1} \Rightarrow p_v = p_1^{v+1},$$

where $t \equiv \frac{p_1}{p_2}$. This relation, together with the normalization $\sum_k p_k p_v = 1$, completely characterize the catalyst eigenvalues that allow to perform the CNU1 transformation.

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. (36). Before deriving the optimal eigenvalues $\{p_k\}_{1 \leq k \leq n}$, let us show why the condition

$$J_k = J_{[2,1,1,v]} \rightarrow [1,2,n,v], \quad \forall 1 \leq k \leq n - 1,$$

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

$$J_k = p_1 p_{k+1} - p_1 p_k,$$

$$J_{[2,1,1,v]} \rightarrow [1,2,n,v] = p_2 p_{k+1} - p_2 p_k.$$

Moreover, let $J_{cool}^{max}$ and $(J_{cool})'$ denote respectively the maximum cooling currents achieved with $\{p_k\}$ and $\{q_k\}$. These currents are given by the minimum swap currents in the corresponding loops:

$$J_{cool}^{max} = J_{Loop}^{min} \left\{ J_k, J_{[2,1,1,v]} \rightarrow [1,2,n,v] \right\},$$

$$(J_{cool})' = (J_{Loop})'^{min} \left\{ J_k, J'_{[2,1,1,v]} \rightarrow [1,2,n,v] \right\}.$$

In other words, the maximum cooling current must satisfy these equations to form a uniform loop with the restoring chain.
To prove that $J^\text{max}_\text{comp}' \leq J^\text{max}_\text{comp}$, we show that any transformation $\{p^v_k\} \rightarrow \{q^v_k\}$ must be such that $(J^\text{max}_\text{comp})' \leq J^\text{max}_\text{comp}$. An arbitrary transformation $\{p^v_k\} \rightarrow \{q^v_k\}$ can be described by the equations

$$\delta_q^v = p^v_k + \delta_k,$$  \hspace{1cm} (E6)

with the values of $\delta_k$ restricted so that $\{q^v_k\}$ represents a probability distribution. In particular, probability conservation implies that $\sum_{k=1}^n \delta_k = 0$. The population variations $\delta_k = q^v_k - p^v_k$ and $\delta_{k+1} = q^v_{k+1} - p^v_{k+1}$ in turn induce a change in the restoring current $\delta J$, denoted as $\delta J_k$. Likewise, since $J_{[2,1\ldots k] \rightarrow [1,2\ldots n]}$ depends on $p^v_1$ and $p^v_n$, the variations $\delta_1$ and $\delta_n$ change this current by an amount $\delta J_{[2,1\ldots k] \rightarrow [1,2\ldots n]}$. In this case, taking into account condition (E1) and Eq. (E4), the variations $\delta_1$ and $\delta_n$ are obtained by using Eqs. (E2) and (E3) in the following way. By defining the quantity $r_h \equiv p^v_n / p^v_1$, it is straightforward to check the identity

$$J_n-1 + r_h J_{n-2} = p^v_1 p^v_n - \frac{(p^h_2)^2}{p^h_1} p^v_{n-2}. \hspace{1cm} (E9)$$

Here, the effect of the factor $r_h$ is to cancel the second term in the current $J_{n-1}$ (given by $p^h_2 p^v_{n-1}$) with the first term of $J_{n-2}$ (given by $p^h_1 p^v_{n-1}$). More generally, the identity $r_h n^h_2 = r_h p^v_{n-1}$ guarantees that in the sum $\sum_{i=0}^{k-1} r_h^i J_{n-(i+1)}$ only the first and last terms survive. That is,

$$\sum_{i=0}^{k-1} r_h^i J_{n-(i+1)} = p^h_1 p^v_n - r_h^{k-1} p^h_2 p^v_{n-k}. \hspace{1cm} (E10)$$

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

$$\sum_{i=0}^{k-1} r_h^i J_{n-(i+1)} = \left(\sum_{i=0}^{k-1} r_h^i\right) J_{[2,1\ldots k] \rightarrow [1,2\ldots n]} = \left(1 - \frac{r_h}{1-r_h}\right) J_{[2,1\ldots k] \rightarrow [1,2\ldots n]}. \hspace{1cm} (E11)$$

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

$$p^v_{n-k} = \frac{p^v_1 - p^h_2}{p^h_1} \left[ p^h_1 \left(1 - \frac{r_h}{1-r_h}\right) J_{[2,1\ldots k] \rightarrow [1,2\ldots n]} \right] p^v_n. \hspace{1cm} (E12)$$

Given the dependence of $J_{[2,1\ldots k] \rightarrow [1,2\ldots n]}$ on $p^v_1$, this equation provides the eigenvalues $p^v_{n-k}$, for $1 \leq k \leq n-1$, in terms of $p^v_n$ and $p^v_1$. To eliminate the dependence on $p^v_1$ we substitute $k = n-1$ and isolate $p^v_n$, which yields

$$p^v_n = \frac{(1-r_h^{n-1}) r_h p^v_1 + (1-r_h)}{(1-r_h^{n-1}) p^h_1 + (1-r_h) r_h} p^v_n. \hspace{1cm} (E13)$$

After inserting this back into Eq. (E12) we can apply the normalization condition $\sum_{k=1}^{n-1} p^v_{n-k} + p^v_n = 1$ to derive $p^v_n$. In this way, we obtain a complete characterization of all the eigenvalues $\{p^v_k\} \leq k \leq n$, in terms of the eigenvalues $\{p^v_k\}$ and $\{p^h_k\}$ of the cold and hot objects. Specifically the normalization condition yields

$$p^v_n = \frac{p^h_1 r_h^{n-1} (1-r_h)}{p^h_1 (1-r_h) + [nr_h^{n-1} (1-r_h) - (1-r_h)] J^\text{max}_\text{comp} (1-\mathcal{R}_h) p^v_n}, \hspace{1cm} (E14)$$

where $J^\text{max}_\text{comp}$ corresponds to Eq. (37) in the main text. Note that $J^\text{max}_\text{comp}/p^v_n$ depends only on $\{p^v_k\}$ and $\{p^h_k\}$, whereby the r.h.s. in the previous equation only depends on these eigenvalues. In addition, Eq. (37) is obtained by replacing $p^v_1$ and $p^v_n$ into $J_{[2,1\ldots k] \rightarrow [1,2\ldots n]}$ (Eq. (E3)) by the expressions given in Eqs. (E13) and (E14). This recasting of $J_{[2,1\ldots k] \rightarrow [1,2\ldots n]}$ as $J^\text{comp}_\text{cool}$ in turn gives rise to Eq. (38).
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, $r_{\text{cool}}^{\text{max}}$ is maximized through a uniform loop formed by swap currents (cf. Eq. (E1)). Besides the swap for the cooling current, which exchanges the states $|2,1,h\rangle$ and $|1,2,h\rangle$, the swaps that give rise to the restoring currents (E2) exchange the states $|1,h(k+1)\rangle$ and $|2,h\rangle$.

Appendix F: Proof of theorem 3

Lemma 2 (optimal cooling of a cold qubit using only the hot object). Let $\{(p^h_j)^\dagger\}_{1 \leq j \leq 3}$ be the 3 largest eigenvalues of $\rho_{ch}$, in non-increasing order, where $\lambda = \frac{d^h}{d_h}$ if $d_h$ is even and $\lambda = \frac{d^h-1}{d_h}$ if $d_h$ is odd. Moreover, let $\{(j_h)\}_{1 \leq j \leq 3}$ denote the corresponding eigenstates. Likewise, let $\{(p^c_j)^\dagger\}_{1 \leq j \leq 3}$ be the 3 + 1 lowest eigenvalues of $\rho_{ch}$ in non-decreasing order, with $\{(j_h)\}_{1 \leq j \leq 3}$ being the corresponding eigenstates. The optimal cooling of a qubit is obtained through a unitary $\hat{U}_{ch}$ that satisfies:

$$\hat{U}_{ch}|2,ch\rangle = |1,ch\rangle^\dagger,$$  \hspace{1cm} (F1)

$$\hat{U}_{ch}|1,ch\rangle = |2,ch\rangle^\dagger,$$ \hspace{1cm} (F2)

for all $j$ such that $p^c_j (p^c_j)^\dagger > p^h_j (p^h_j)^\dagger$, and $\hat{U}_{ch}|2,ch\rangle = |2,ch\rangle^\dagger$, $\hat{U}_{ch}|1,ch\rangle = |1,ch\rangle^\dagger$, for all $j$ such that $p^c_j (p^c_j)^\dagger \leq p^h_j (p^h_j)^\dagger$.

Proof of Lemma 2. Let us denote as $A_j \equiv \{p^c_j (p^c_j)^\dagger\}$ and $A_\subset \equiv \{p^h_j (p^h_j)^\dagger\}$ the subsets of eigenvalues of $\rho_{ch}$ that satisfy $p^c_j (p^c_j)^\dagger > p^h_j (p^h_j)^\dagger$. In other words, $A_j$ and $A_\subset$ contain the eigenvalues characterizing eigenstates of $\rho_{ch}$ on which $\hat{U}_{ch}$ acts non-trivially. Note that, by construction, $A_\subset \leq \{p^c_j (p^c_j)^\dagger\}_{1 \leq j \leq 3}$ and $A_j \subseteq \{p^h_j (p^h_j)^\dagger\}_{1 \leq j \leq 3}$. The eigenstates of $\rho_{ch}$ that possess eigenvalue $\epsilon_j$ with respect to $H_c$ have eigenvalues $\{p^c_j (p^c_j)^\dagger\} = A_j \cup A_\subset$, where $A_\subset = \{a \in \{p^c_j (p^c_j)^\dagger\} \text{ s.t. } a \notin A_j\}$ is the complement of $A_j$. Similarly, the eigenstates with eigenvalue $\epsilon_j$ with respect to $H_e$ have eigenvalues $\{p^e_j (p^e_j)^\dagger\} = A_j \cup A_e$, where $A_e = \{a \in \{p^c_j (p^c_j)^\dagger\} \text{ s.t. } a \notin A_j\}$. The unitary $\hat{U}_{ch}$ is a permutation that exchanges the eigenvalues $A_j$ and $A_\subset$, whence the final eigenvalues corresponding to eigenstates $\{1,j_h\}_1$ and $\{2,j_h\}_1$ are respectively given by $A_j \cup A_\subset$ and $A_j \cup A_e$.

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

$$\max(a \in A_j) \leq \min(a \in A_\subset), \min(a \in A_e),$$

$$\max(a \in A_\subset) \leq \min(a \in A_j), \min(a \in A_e).$$

The inequality $\max(a \in A_j) \leq \min(a \in A_e)$ holds because $A_\subset$ is a subset of the smallest elements of $A_j \cup A_e$, and $\max(a \in A_\subset) \leq \min(a \in A_j)$ holds because $A_j$ is a subset of the largest elements of $A_j \cup A_e$. Moreover, we note that the maximum index $j = j_{\text{max}}$ in the sets $A_j$ and $A_e$ determines the corresponding maximum and minimum elements, and that by definition $p^e_j (p^e_j)^\dagger < p^h_j (p^h_j)^\dagger$. Therefore, $\max(a \in A_j) \leq \min(a \in A_e)$.

This leaves us with the verification of the inequality $\max(a \in A_\subset) \leq \min(a \in A_e)$. Noting that we can write $\max(a \in A_\subset) = p^c_j (p^c_j)^\dagger$ and $\min(a \in A_e) = p^h_j (p^h_j)^\dagger$, the assumption $\max(a \in A_\subset) > \min(a \in A_e)$ is contradictory because it would imply that $\max(a \in A_\subset)$ and $\min(a \in A_e)$ also belong to $A_j$ and $A_\subset$, respectively. In this way, it follows that $\max(a \in A_\subset) \leq \min(a \in A_e)$, which completes the proof of the inequality $\max(a \in A_j \cup A_\subset) \leq \min(a \in A_e \cup A_\subset)$ and the passivity of $\rho'_{ch}$ with respect to $H_e$.

Proof of Theorem 3. Let us see that, after the application of $\hat{U}_{ch}$, a subsequent cooling can always be achieved through a CC transformation. Such a transformation operates with the eigenstates of $\rho_{ch}$ whose eigenvalues belong to the sets $A_j$ and $A_\subset$, on which $\hat{U}_{ch}$ acts as the identity (cf. Eqs. (F1) and (F2)). Let us denote the eigenstates with eigenvalues in $A_j$ as $\{1,j_h\}_1$ and the eigenstates with eigenvalues in $A_\subset$ as $\{2,j_h\}_1$. What we show below is that there exists a catalyst state $\rho_{cv} \in H_{ch}$ that allows to implement a CC transformation, through a global unitary $U'$ that maps the enlarged subspace $\operatorname{span}(\{1,j_h\}_1 \cup \{0,j_h\}_1) \otimes H_e$ into itself. This is a consequence of the hypothesis concerning the non-degeneracy of the largest or the smallest eigenvalues of $\rho_{ch}$, referred to in Theorem 3. Specifically, we will show that there exists a restoring chain that forms a loop with the cooling current

$$J_{\text{cool}} = J_{[2,j_{\text{max}}^c] \otimes \{1,\} + [1,j_{\text{min}}^c] \otimes \{d_h\}},$$

where $2,j_{\text{max}}^c$ is an eigenstate with eigenvalue $\max(a \in A_j)$, and $1,j_{\text{min}}^c$ is an eigenstate with eigenvalue $\min(a \in A_\subset)$. Since the sets $A_j$ and $A_\subset$ have cardinalities $|A_j| = |A_\subset| \leq 3$, the cardinalities of the complementary sets $A_{\text{c}}$ and $A_{\text{e}}$ are such that $|A_{\text{c}}| = |A_{\text{e}}| \geq d_h - 3$. Taking into account the definition of $\hat{3}$ in Lemma 2, this implies that $|A_{\text{c}}| = |A_{\text{e}}| \geq \frac{d_h}{2}$ if $d_h$ is even, and $|A_{\text{c}}| = |A_{\text{e}}| \geq \frac{d_h+1}{2}$ if $d_h$ is odd. Recalling that $A_{\text{c}} = \{a \in \{p^c_j (p^c_j)^\dagger\} \text{ s.t. } a \notin A_j\}$, it follows that the set $A_{\text{c}}$ includes the smallest $d_h$ elements of $\{p^c_j (p^c_j)^\dagger\}_{1 \leq j \leq d_h}$ if $d_h$ is even (odd).

Similarly, the set $A_{\text{e}}$ includes the largest $\frac{d_h}{2}$ (or $\frac{d_h+1}{2}$) elements of $\{p^c_j (p^c_j)^\dagger\}_{1 \leq j \leq d_h}$ if $d_h$ is even (odd). Moreover, the smallest elements of $\{p^c_j (p^c_j)^\dagger\}_{1 \leq j \leq d_h}$ and the largest
elements of \(\{p_j^h\}_{1 \leq j \leq 3}\) are respectively determined by the smallest and largest eigenvalues of \(\rho_h\), referred to in Theorem 3. Accordingly, if the hypothesis of this theorem holds, at least two elements of either of the sets \(A^\varepsilon\) or \(A^\varphi\) are different. Suppose first that such elements belong to \(A^\varepsilon\), and let us denote as \(|1_cJ^\max_h\rangle\) and \(|1_dJ^\min_h\rangle\) two eigenstates with eigenvalues \(\max(a \in A^\varepsilon)\) and \(\min(a \in A^\varepsilon)\) respectively. By choosing a sufficiently mixed catalyst, we can construct a restoring chain

\[
\{J_{|1_cJ^\max_h\rangle \otimes |(k+1)_v\rangle \rightarrow |1_dJ^\min_h\rangle \otimes |k_v\rangle\}_{1 \leq k_v \leq d_v - 1},
\]

characterized by the inequalities \(\max(a \in A^\varepsilon)p^v_{k+1} > \min(a \in A^\varepsilon)p^v_k\) for \(1 \leq k_v \leq d_v - 1\). Likewise, if the set \(A^\varphi\) contains at least two different elements, the loop can be closed with the chain

\[
\{J_{|2_cJ^\max_h\rangle \otimes |(k+1)_v\rangle \rightarrow |2_dJ^\min_h\rangle \otimes |k_v\rangle\}_{1 \leq k_v \leq d_v - 1},
\]

where \(|2_cJ^\min_h\rangle\) is an eigenstate of \(\rho_{ch}\) with eigenvalue \(\min(a \in A^\varepsilon)\) not \(\max(a \in A^\varepsilon)\).

Since any of the previous chains is composed of currents that take inside energy eigenspaces of the cold qubit, they do not spoil the cooling effect due to \(J_{\text{cool}}\). In addition, note that the cooling current is determined by the inequality \(p^v_{2\varepsilon}\max(a \in A^\varepsilon)p^v_0 > p^v_{\min}(a \in A^\varepsilon)p^v_0\), which can always be satisfied if the \(d_v\) is large enough, even for a very mixed state \(\rho_v\).

**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 \(\rho_H\) that describe \(l \leq k\) 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 \((p^h_1)^{k-l}(p^h_2)^l > (p^h_2)^{k-l}(p^h_1)^l\). This implies that the largest eigenvalues of \(\rho_H\) correspond to \(l \leq k/2\).

From Lemma 2, the optimal unitary \(\tilde{U}_{ch}\) is composed of swaps between eigenstates \([2_cJ^\uparrow_h]_l\) and \([1_dJ^\downarrow_h]_l\), where \([J^\uparrow_h]_l\) is an eigenstate of \(\rho_{ch}\) with \(l \leq k/2\) excited (non excited) hot qubits. Each of these swaps generates a cooling current

\[
J_{\text{cool}}^{(l)} = p^l_2[(p^h_1)^{k-l}(p^h_2)^l] - p^l_1[(p^h_1)^l(p^h_2)^{k-l}] = (p^l_2)^l - (p^l_1)^l + (p^l_2)^{k-l},
\]

where in the second line we use the condition \(p^l_c = p^h_c\) (the cold qubit is identical to the hot qubits). The maximum current is achieved for \(l = 0\) and \(J_{\text{cool}}^{(l)}\) is non-increasing with respect to \(l\).

For \(k\) even, choosing \(l = k/2\) yields \(J^{(k/2)}_{\text{cool}} = (p^l_2)^{k/2}(p^h_2) - (p^l_1)^{k/2}(p^h_1) < 0\). On the other hand, if \(l = k/2 - 1\) we have that \(J^{(k/2-1)}_{\text{cool}} = (p^l_2)^{k/2}(p^h_1) > 0\). 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

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

For \(k\) odd, choosing \(l = (k-1)/2\) yields \(J^{(k-1)/2}_{\text{cool}} = 0\), and choosing \(l = (k-3)/2\) yields \(J^{(k-3)/2}_{\text{cool}} = (p^l_1)^{k/2}(p^h_1) - (p^l_2)^{k/2}(p^h_2) > 0\). Accordingly, the total population increment in this case reads

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

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

\[
\xi_{\text{cool}}^{(2)} = \frac{J^{(0)}_{\text{cool}}}{2} = \frac{(p^l_2)^2(p^h_2)^1 - (p^l_1)^1(p^h_1)^2}{2} = \frac{1 - 2p^l_2}{2} p^l_1 p^h_2,
\]

which coincides with Eq. (46) of the main text.

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