Thermodynamic limits of dynamic cooling

Armen E. Allahverdyan,[1] Karen V. Hovhannisyan,[1] Dominik Janzing,[2] and Guenter Mahler[3]

[1]*Yerevan Physics Institute, Alikhanian Brothers Street 2, Yerevan 375036, Armenia*
[2]*MPI for Intelligent Systems, Spemannstrasse 38, D-72076 Tuebingen, Germany*
[3]*Institute of Theoretical Physics I, University of Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany*

We study dynamic cooling, where an externally driven two-level system is cooled via reservoir, a quantum system with initial canonical equilibrium state. We obtain explicitly the minimal possible temperature $T_{min} > 0$ reachable for the two-level system. The minimization goes over all unitary dynamic processes operating on the system and reservoir and over the reservoir energy spectrum. The minimal work needed to reach T_{min} grows as $1/T_{min}$. This work cost can be significantly reduced, though, if one is satisfied by temperatures slightly above T_{min}. Our results on $T_{min} > 0$ prove unattainability of the absolute zero temperature without ambiguities that surround its derivation from the entropic version of the third law. We also study cooling via a reservoir consisting of $N \gg 1$ identical spins. Here we show that $T_{min} \propto \frac{1}{N}$ and find the maximal cooling compatible with the minimal work determined by the free energy. Finally we discuss cooling by reservoir with an initially microcanonic state and show that although a purely microcanonic state can yield the zero temperature, the unattainability is recovered when taking into account imperfections in preparing the microcanonic state.

I. INTRODUCTION

Some physical systems have to be cooled before they can demonstrate interesting features, for example, quantum properties of matter are typically displayed only after suitable cooling. There are various cooling methods; we distinguish here the "brute force method," the Nernst setup, and the so-called dynamic cooling. The brute force method of cooling amounts to bringing the system in contact with a low-temperature thermal bath, so that it relaxes to this lower temperature.

When trying to do without a pre-existing cold bath different strategies are needed: Low temperatures can alternatively be produced dynamically from an initially equilibrium system via cyclic action of an external field [1–18]. In this case the resulting cooled state should be exploited before the system has time to relax back to equilibrium.

Within macroscopic quasiequilibrium thermodynamics some of these methods have been summarized in the Nernst setup of cooling, which supports a formulation of the third law [1–7]. In fact, this law controls the cooling of a macroscopic target initially in contact with an appropriate macroscopic reservoir. The target is then typically subject to a two-step cooling process in which an external field cycle is executed under (quasiequilibrium) isothermal and adiabatic conditions, respectively. A well-known realization of this Nernst setup is the magnetocaloric effect, which was first observed in 1880, but still attracts attention [8].

There are, however, other methods of cooling that became important with the rise of low-temperature physics [9–18]. Here the target of cooling (or the reservoir, or both) is generally not a macroscopic system, while the process that produces low temperatures ceases to be a quasiequilibrium one. Hence, the understanding of these methods should rely on the actual dynamics rather than on quasiequilibrium thermodynamics. We therefore refer to them as dynamic cooling. A notable example of this is the dynamical nuclear polarization in nuclear magnetic resonance (NMR) [9–14]. Within this method the nuclear spins are cooled via transferring polarization

from electron spins by means of external microwave fields [9–14]. It was originally employed in the solid-state NMR, but since recently it is also applied for the liquid-state NMR in view of its medical and biophysical applications [14]. Other examples of dynamic cooling are algorithmic cooling [15,16], bath-assisted cooling [17], and laser cooling of motional state in atoms [18].

Dynamic nuclear polarization illustrates the basic ingredients of dynamic cooling processes that are seen already in the Nernst setup: the target system to be cooled (nuclear spins), the reservoir, which plays the role of a polarization source or the entropy sink (electrons), and external fields that couple these two together (microwave radiation at suitable frequency). Recall that no cooling is possible without reservoir [19]; that is, one can never cool the whole system (=target + reservoir) by coherent fields. As already indicated, the operation of cooling and the concept of the third law are intimately interrelated. While we do not attempt to dwell on the general ideas behind operationalism as originally promoted by Bridgman [20], it is nevertheless worthwhile to stress the potential benefits of such an approach even to modern thermodynamics, and, in particular, for reaching the objectives of this paper. Our aim here is threefold: to formalize cooling schemes in terms of imposed limited resources, to parametrize specific reservoir models, and to investigate the resulting minimal temperature as a (scaling) function of those operational parameters.

In particular, we intend to study the minimal temperature T_{min} reachable within a sufficiently general setup of dynamic cooling, where both the target and the reservoir are finite quantum systems and where the final state of the reservoir can generally be far from equilibrium. We determine how T_{min} depends on resources of the setup and note that the minimal work necessary to achieve T_{min} grows as $\frac{1}{T_{min}}$ whenever $T_{min} \to 0$. Hence, the work is a diverging resource of dynamic cooling, in contrast to the Nernst setup and the third law, where the work done for cooling is always well bounded. We also determine the minimal temperature (and the minimal work needed to attain for it) for several concrete reservoir models.

Our results on $T_{\min} > 0$ extend (to dynamic cooling) the unattainability formulation of the third law. Hence, Sec. II reviews the (frequently disguised) assumptions of the Nernst setup and of the third law in its two formulations. (The reader who is well aware of the third law may just consult Sec. II D for relevant assumptions.)

The dynamic cooling setup is defined in Sec. III. Section III C shows that dynamic cooling with a reservoir in an initial canonical state does not allow to reach the absolute zero of temperature [unattainability]. In Sec. III E we summarize relevant conditions of the dynamic cooling setup.

The following three sections are concerned with reservoir models. Section IV studies the maximal cooling possible within the dynamic setup. It confirms that the work is a relevant resource for cooling and determines the minimal work necessary to achieve the maximal cooling. Section V studies a reservoir with the homogeneous, nondegenerate spectrum, while in Sec. VI the reservoir is modeled as a thermal bath consisting of $N \gg 1$ spins. This model is complemented by a discussion on the dynamic cooling process for an initially microcanonic state of the reservoir.

In the last section we summarize our results and discuss their relations with other microscopic approaches for studying thermodynamic limits of cooling: The approach of Ref. [21], where the cooling process is restricted to operations within degenerate subspaces of the joint Hamiltonian of the system and nonequilibrium reservoir, and the approach based on quantum refrigerators [22–26]. We close by pointing out some open issues.

II. OPERATIONAL ANALYSIS OF THE NERNST SETUP AND THE THIRD LAW

A. Description of the Nernst setup

We are given a macroscopic system in contact with a much larger thermal bath [1]. Both have initial temperature T_{in}. One now switches on an external field g acting on the system and changes it *slowly* from its initial value g_{in} to its final value g_{fin}. The contact with the bath is held fixed. Hence, this is an isothermal process. Let g_{in} and g_{fin} be chosen such that the entropy $S[T,g]$ of the system *decreases* (see Fig. 1):

$$S[T_{\mathrm{in}}, g_{\mathrm{in}}] > S[T_{\mathrm{in}}, g_{\mathrm{fin}}]. \qquad (1)$$

The entropy difference is transferred to the bath.

Next, the system is *thermally* isolated (i.e., isolated from the bath) and the field g is *slowly* returned back to its original value g_{in}, thereby completing the cooling cycle. (Note: The cyclic condition only refers to the external field, that is, the time-dependent Hamiltonian; it would be meaningless to require the target to return to its initial state.) Hence, this part of the process is thermally isolated and reversible (adiabatic). Since the system is macroscopic, the adiabatic part is characterized by constant thermodynamic entropy [1]:

$$S[T_{\mathrm{fin}}, g_{\mathrm{in}}] = S[T_{\mathrm{in}}, g_{\mathrm{fin}}], \qquad (2)$$

where T_{fin} is the final temperature of the system. Due to (1) and $\frac{\partial S}{\partial T} \geqslant 0$, we get $T_{\mathrm{fin}} < T_{\mathrm{in}}$: some cooling has been achieved [1–8]. Recall that $\frac{\partial S}{\partial T} \geqslant 0$ is one of basic conditions of local thermodynamic stability (positivity of the specific heat) [1].
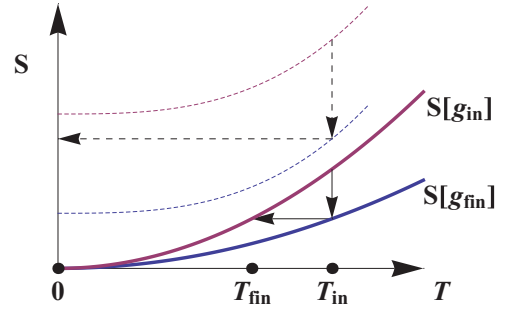

FIG. 1. (Color online) A schematic representation of the relation between the entropic formulation of the third law and the attainability of $T = 0$. Here S and T denote entropy and temperature, respectively; g is the external field. Bold (dashed) lines represent a situation, where the entropic formulation holds (is violated). Arrows represent the isothermal and adiabatic parts of the cooling cycle. We see that the final temperature T_{fin} is greater than zero. However, it is formally possible to attain $T = 0$ (dashed arrows) when the entropic formulation is violated in the sense that $S[T = 0]$ still depends on the field g.

The system will stay cold as long as it is well isolated from the bath.

This cooling cycle has a work cost. In terms of the free energy,

$$F[T,g] = U[T,g] - T S[T,g], \qquad (3)$$

of the system, where $U[T,g]$ is its energy, the work done in the isothermal step is $F[T_{\mathrm{in}}, g_{\mathrm{fin}}] - F[T_{\mathrm{in}}, g_{\mathrm{in}}]$. The work in the adiabatic step is $U[T_{\mathrm{fin}}, g_{\mathrm{in}}] - U[T_{\mathrm{in}}, g_{\mathrm{in}}]$. Summing them up and using (2) one has for the work W invested per cooling cycle[1]:

$$W = F[T_{\mathrm{fin}}, g_{\mathrm{in}}] - F[T_{\mathrm{in}}, g_{\mathrm{in}}] > 0, \qquad (4)$$

which is always positive due to $\frac{\partial F}{\partial T} = -S \leqslant 0$[2] and $T_{\mathrm{in}} > T_{\mathrm{fin}}$. Below, in Secs. III and VI, we relate (4) to the minimal work cost of the dynamic cooling setup. Note that W according to (4) is always finite, even for $T_{\mathrm{fin}} = 0$.

B. Entropic formulations of the third law

The entropic formulation states that for each fixed and finite value of the field g, the entropy $S[T,g] \geqslant 0$ *smoothly* goes to zero with temperature T: $S[T,g] \to 0$ for $T \to 0$ [1–7] (see Fig. 1). The entropic formulation is frequently deduced from the nondegeneracy of the system's ground state. However, this does not suffice for a derivation, because for a macroscopic system the appropriate order for taking limits is that first the thermodynamic limit is taken, the entropy density is calculated, and then after that the low-temperature limit goes [4,5]. Then the zero-temperature entropy starts to depend on global features of the energy spectrum. The full derivation within statistical mechanics was attempted several times [4–7] but is regarded to be an open problem, because there are

---

[1]We use the sign convention $W > 0$, if the work source loses energy.
[2]We assume the convention where the entropy is defined to be non-negative.

different classes of theoretical Hamiltonian systems violating the entropic formulation: The zero-temperature entropy for them is not zero and depends on external fields [4,6,27–32].

It is believed that Hamiltonians which would violate the entropic formulation are unstable with respect to small perturbations [2,6]. Such perturbations would have to be generated by some appropriate noise model. Unfortunately, a general recipe for selecting such a model does not seem to be known (or even exist).[3] There are results showing how one should select the noise model for particular cases only [2,6,27,30,33,34].

C. Unattainability formulation of the third law

Already the entropic formulation of the third law can be brought in contact with operational requirements. This is even more so for the unattainability formulation. This formulation states that it is impossible to reach absolute zero of temperature, $T = 0$, by finite means, that is, via a *tractable* physical process. Note that the unattainability formulation is clearly different from the entropic formulation that refers to a limiting feature of a definite function of temperature (entropy). The unattainability formulation relies on the notion of tractability, whose *general* formalization appears to be hard to come by. Hence, the two formulations cannot be completely equivalent [3].

Within the Nernst setup one naturally assumes that for a tractable physical process the strength g of the external field should not assume infinite values [1,2]. This suffices for a heuristic derivation of the unattainability formulation from the entropic formulation (see Fig. 1 and [1]). Indeed, Fig. 1 also shows that the unattainability is violated together with counterexamples of the entropic formulation.[4] However, there are also examples that would violate the unattainability formulation only [2,37].[5] Again, one way out of this ambiguous situation is to look for "sufficiently" stable Hamiltonians [2].

D. Summary of conditions

We finally try to summarize the Nernst setup in terms of two lists of conditions: The first list **N1**–**N6** specifies

---

[3]The reader may consult controversies with the validity of the entropic formulation in the field of the Casimir effect [27,28,33] or for black holes [29], criteria of this formulation for spin systems [30,34], and classes of one-dimensional counterexamples to the formulation [31,32]. The authors of [32] ensured robustness of their counterexamples to a class of noise models.

[4]This would not imply that one can ever verify reaching $T = 0$, since any temperature measurement has a finite precision. (The situation with reaching a positive temperature, say 10 K, is different, because one can arrange for passing through this temperature at some [uncertain] time.) In particular, temperature fluctuations can prevent a precise determination of low temperatures [35,36]. According to the standard thermodynamic approach [35] these fluctuations will grow for $T \rightarrow 0$ due to the vanishing heat capacity. The issues of temperature fluctuations was recently reconsidered and clarified in [36] from the first principles of quantum mechanics.

[5]The mechanism of these counterexamples is that the entropy nullifies at some $T_c(g) > 0$ and stays zero for $T \leqslant T_c(g)$.

details of the task: It reflects our choice for implementing a satisfactory cooling process. In part these conditions may be tied to the actual technology. The second list **T1**–**T3** introduces fundamental tractability conditions. These stress the unavoidable finiteness of the resources at our disposal and should remain intact within any future technology. The respective process will be called tractable insofar as these requirements can be fulfilled:

(**N1**) initial state, equilibrium for the system and bath;

(**N2**) system (target of cooling), macroscopic;

(**N3**) cooling process, quasiequilibrium;

(**N4**) bath, macroscopic, much larger than the system;

(**N5**) external field, changes cyclically;

(**N6**) the Hamiltonian of the target, stable with respect to perturbations.

N1 recalls that we intend to start from a given finite temperature, while **N2**–**N4** are assumed because the process is to be embedded into macroscopic equilibrium thermodynamics. **N2** and **N3** reflect the technology of Nernst's times. **N5** is assumed, because the cooled system has to be *autonomous*; that is, once it was cooled, it can be used in other places, without being kept under a constant external field. **N6** is included here because a general noise model for perturbations is not known: The concrete stability requirement thus becomes a matter of choice.

We now turn to the tractability conditions; this list is not necessarily complete (cf. Sec. VI C):

(**T1**) external field has finite strength;

(**T2**) the work cost necessary for cooling is finite;

(**T3**) duration of the cooling cycle is finite.

T1 turns out to be essential for deducing the unattainability formulation of the third law. It is also relevant for the entropic formulation. (It is easy to find examples where the entropy does not nullify with the temperature if simultaneously the field goes to infinity [2].) Due to (4), **T2** is guaranteed by **N1**–**N4**. Typically, **T3** becomes essential for certain scenarios: Indeed, in frustrated systems reaching the true equilibrium may demand unrealistically long observation times; on practically relevant observation times the system may find itself trapped in quasistationary states, where the nonequilibrium entropy does not go to zero with the ambient temperature, though the true equilibrium state does satisfy the entropic formulation. This effect was observed experimentally [38]. It is also well known for glassy systems [39] (residual entropy) (see [40] for a review).

III. SETUP FOR DYNAMIC COOLING

A. System and reservoir

A reasonably general setup for dynamic cooling requires to specify the class of Hamiltonians for the target system **S** and the reservoir **R**, together with their initial state. Here both subsystems will be taken to be finite-level quantum systems with the overall Hamiltonian $H_{\mathbf{S+R}}$. The initial state of $\mathbf{S} + \mathbf{R}$ is a *canonical* equilibrium one with density matrix

$$\rho_{\mathbf{S+R}} = \frac{e^{-\beta H_{\mathbf{S+R}}}}{\mathrm{tr}\,[\,e^{-\beta H_{\mathbf{S+R}}}\,]}, \tag{5}$$

and the initial temperature $T = 1/\beta$ ($k_B = 1$). We additionally assume that in the initial state the coupling between **S** and **R** is negligible:

$$H_{\mathbf{S+R}} = H_{\mathbf{S}} + H_{\mathbf{R}}, \quad \rho_{\mathbf{S+R}} = \rho_{\mathbf{S}} \otimes \rho_{\mathbf{R}}. \tag{6}$$

The action of external fields is described by an interaction term in the time-dependent Hamiltonian of **S** + **R**:

$$H_{\mathbf{S+R}}(t) = H_{\mathbf{S}} + H_{\mathbf{R}} + V(t), \tag{7}$$

which is switched on at time $t = 0$ and switched off at time $t = \tau$: $V(t) = 0$ for $t < 0$ and for $t > \tau$ (*cyclic* motion of the external field). $V(t)$ acts on both **S** and **R**; if it acts only on **S**, no cooling is possible [19]. $H_{\mathbf{S+R}}(t)$ generates a unitary operator U that takes the initial state (5) to the final state of **S** + **R**:

$$\rho_{\mathbf{S+R}}(\tau) = U\rho_{\mathbf{S+R}}U^{\dagger}, \tag{8}$$

$$\rho_{\mathbf{S}}(\tau) = \mathrm{tr}_{\mathbf{R}}\rho_{\mathbf{S+R}}(\tau), \quad \rho_{\mathbf{R}}(\tau) = \mathrm{tr}_{\mathbf{S}}\rho_{\mathbf{S+R}}(\tau), \tag{9}$$

where we defined also marginal final states of **S** and **R**.

B. Definition of cooling

How to define cooling of **S** in the nonequilibrium setting? Note that the maximal cooling is always well defined, because it means that the final ground state probability of **S** is [$|0\rangle$ is the ground state of $H_{\mathbf{S}}$]

$$p_0(\tau) \equiv \langle 0|\rho_{\mathbf{S}}(\tau)|0\rangle = 1 \quad (\text{or} \quad T = 0). \tag{10}$$

Defining a nonmaximal cooling via $p_0(\tau)$ is reasonable for two-level systems, since a larger $p_0(\tau)$ means that the energy distribution in the final state is more shifted toward the ground state.[6] If in addition the state of **S** is diagonal in the energy representation, one can equivalently express the cooling via temperature defined as in (5). For a multilevel system **S** one needs to take care in defining the meaning of a possibly nonequilibrium state of **S** being colder than a given equilibrium state.[7]

_____

[6]One may define cooling via the maximal eigenvalue of the density matrix and not the ground-state probability. In the optimal regime of our setup both definitions agree with each other, because the final state of **S** is energy-diagonal; see Sec. IV A.

[7]When comparing two systems with the same energy levels—e.g., the target of cooling before and after the cooling process realized via a cyclically changing Hamiltonian—one can define cooling by requiring that the whole energy distribution is shifted toward the ground state. In effect, this amounts to using *majorization* as a measure of cooling; see [41] and Appendix A for the definition of this concept, see also [21] for a related approach. The drawback (or viewing differently an advantage) of this definition is that when the number of energy levels is larger than two, not every two states can be said to be cold or hot relative to each other. Another (less preferred by us) approach to cooling would be to employ global measures such as entropy.

For simplicity we assume that **S** is a two-level system with energies[8]

$$0 < \epsilon \tag{11}$$

and equilibrium probabilities

$$p_0 \equiv \langle 0|\rho_{\mathbf{S}}(0)|0\rangle = \frac{1}{1 + e^{-\beta\epsilon}}, \quad p_1 = \frac{e^{-\beta\epsilon}}{1 + e^{-\beta\epsilon}}. \tag{12}$$

The reservoir **R** is a M-level system with energies

$$0 = \mu_0 \leqslant \mu_1 \leqslant \cdots \leqslant \mu_{M-1} \equiv \mu, \tag{13}$$

and initial equilibrium probabilities [see (5), (6)]

$$\pi_l = \frac{e^{-\beta\mu_l}}{1 + \sum_{k=1}^{M-1} e^{-\beta\mu_k}}, \quad l = 0, \ldots, M-1. \tag{14}$$

The eigenvalues of the initial density matrix $\rho_{\mathbf{S+R}}$ read

$$\{\omega_k\}_{k=0}^{2M-1} = (p_0\pi_0, \, p_1\pi_0, \, p_0\pi_1, \, p_1\pi_1, \ldots). \tag{15}$$

C. Unattainability of the absolute zero

The above setup suffices for showing the unattainability of the absolute zero, $p_0(\tau) > 1$, for the target of cooling **S**, given the initial state (5) of the reservoir. The final ground-state probability $p_0(\tau)$ of **S** follows from (5)–(14),

$$1 - p_0(\tau) = \sum_{i=0}^{1} \sum_{\alpha,\gamma=0}^{M-1} p_i\pi_\gamma [1 - |\langle 0,\alpha|U|i,\gamma\rangle|^2], \tag{16}$$

where $\{|i\rangle\}_{i=0}^{1}$ and $\{|\alpha\rangle\}_{i=0}^{M-1}$ are the eigenbases of, respectively, $H_{\mathbf{S}}$ and $H_{\mathbf{R}}$ in (6), and $\{\pi_\alpha\}_{\alpha=0}^{M-1}$ is given by (14). Since $\sum_{i=0}^{1}\sum_{\gamma=0}^{M-1}|\langle 0,\alpha|U|i,\gamma\rangle|^2 = 1$, some inequlities $1 \geqslant |\langle 0,\alpha|U|i,\gamma\rangle|^2$ must be strict. Noting also $p_i\pi_\gamma > 0$ we deduce from (16)

$$p_0(\tau) < 1 \quad (\text{strict inequality}). \tag{17}$$

This argument applies for $M = \infty$, where, for example, $\sum_{i=0}^{1}\sum_{\gamma=0}^{\infty}|\langle 0,\alpha|U|i,\gamma\rangle|^2 = 1$ and $\sum_{\gamma=0}^{\infty}\pi_\gamma = 1$ are convergent series, and where U is unitary for $M = \infty$. Note that the argument leading to (17) is essentially based on $p_i\pi_\gamma > 0$ and hence $\pi_\gamma > 0$ [see (14)].

With trivial changes the derivation of (17) applies for a multilevel system **S**.

D. Lower bounds for work

Here we summarize restrictions imposed by the second law on the work needed for cooling. Our presentation uses the same ideas and methods as [42,43]; see also [44] in this context.

_____

[8]Consider a multilevel system whose energy gap between the ground state and the first excited state is smaller than the gap between the first and second excited state. For sufficiently low initial temperatures this system can be regarded as effectively two-level system, because populations of the second and higher energy levels can be neglected.

Recall that for the considered unitary (thermally isolated) process the work done on the system is equal to the [average] energy difference [42,43]:

$$W = \mathrm{tr}\{[\rho_{\mathbf{S+R}}(\tau) - \rho_{\mathbf{S+R}}]H_{\mathbf{S+R}}\}, \qquad (18)$$

where we recall that the interaction $V(t)$ with external fields is switched off after the final time [see (7)].

Since the initial state (5) is at a Gibbsian equilibrium, any unitary operator that changes this state costs some work. Indeed, given the initial state (5) and the unitary dynamics implemented by a cyclically changing Hamiltonian (7), (8), the work (18) invested in $\mathbf{S} + \mathbf{R}$ amounts to

$$\beta W = \mathrm{tr}[\rho_{\mathbf{S+R}} \ln \rho_{\mathbf{S+R}} - \rho_{\mathbf{S+R}}(\tau) \ln \rho_{\mathbf{S+R}}] \qquad (19)$$

$$= \mathrm{tr}[\rho_{\mathbf{S+R}}(\tau) \ln \rho_{\mathbf{S+R}}(\tau) - \rho_{\mathbf{S+R}}(\tau) \ln \rho_{\mathbf{S+R}}] \qquad (20)$$

$$\equiv S[\rho_{\mathbf{S+R}}(\tau)||\rho_{\mathbf{S+R}}] \geqslant 0, \qquad (21)$$

where in moving from (19) to (20) we used the unitarity of dynamics [see (8)]. The relative entropy $S[\rho_{\mathbf{S+R}}(\tau)||\rho_{\mathbf{S+R}}]$ is non-negative and nullifies only if $\rho_{\mathbf{S+R}}(\tau) = \rho_{\mathbf{S+R}}$; see Ref. [45] for further features of the relative entropy and its role in (quantum) information theory. Thus, any change of the initially equilibrium state via a cyclic Hamiltonian process costs some work. This work is also a resource of dynamic cooling.

In the context of cooling one can derive more stringent lower bounds on the work. Using (6) we rearrange (20):

$$\beta W = S[\rho_{\mathbf{S}}(\tau)||\rho_{\mathbf{S}}] + S[\rho_{\mathbf{R}}(\tau)||\rho_{\mathbf{R}}] + I_{\mathbf{SR}}(\tau), \qquad (22)$$

$$I_{\mathbf{S+R}}(\tau) \equiv \mathrm{tr}[\rho_{\mathbf{S+R}}(\tau) \ln \rho_{\mathbf{S+R}}(\tau)]$$
$$- \mathrm{tr}[\rho_{\mathbf{S}}(\tau) \ln \rho_{\mathbf{S}}(\tau)] - \mathrm{tr}[\rho_{\mathbf{R}}(\tau) \ln \rho_{\mathbf{R}}(\tau)] \geqslant 0. \qquad (23)$$

$I_{\mathbf{S+R}}(\tau)$ is the mutual information between $\mathbf{S}$ and $\mathbf{R}$ in the final state. $I_{\mathbf{S+R}}(\tau) \geqslant 0$ due to the subadditivity of the entropy.

Now (5), (6), (22) and (21), (23) produce

$$W \geqslant T S[\rho_{\mathbf{S}}(\tau)||\rho_{\mathbf{S}}]$$
$$= \mathcal{F}[\rho_{\mathbf{S}}(\tau)] - \mathcal{F}[\rho_{\mathbf{S}}] \equiv \Delta\mathcal{F} \geqslant 0, \qquad (24)$$

where for any density matrix σ and the initial temperature T we defined

$$\mathcal{F}[\sigma] \equiv \mathrm{tr}(H_{\mathbf{S}}\sigma) + T\mathrm{tr}(\sigma \ln \sigma). \qquad (25)$$

If σ is a Gibbsian density matrix at temperature T, $\mathcal{F}[\sigma]$ coincides with equilibrium free energy [cf. (3)].[9]

---

[9]If the reservoir is a thermal bath at temperature T, then—as advocated in [46]—$\mathcal{F}$ in (25) can be interpreted as a nonequilibrium generalization of the equilibrium free energy. However, in contrast to the equilibrium free energy, $\mathcal{F}$ is not a state function, since it is defined via the bath temperature T. Nevertheless, it has several useful features; for example, it determines the maximal work extractable from a nonequilibrium system in contact with a (canonic equilibrium) thermal bath at temperature T [46]. From that viewpoint, one can interpret (22) as follows. The work separates into three components: the (nonequilibrium) free energy difference for $\mathbf{S}$, the equilibrium free energy difference for the bath $\mathbf{R}$, and the energy stored in the mutual information between $\mathbf{S}$ and $\mathbf{R}$.

In general, however, $\Delta\mathcal{F}$ in (24) is different from the free energy difference (4) of the Nernst setup, where both the initial and final states are at equilibrium. Hence, in (4) the temperatures T_{in} and T_{fin} refer to the initial and final states, respectively. For dynamic cooling only the initial states are at Gibbsian equilibrium (5), so that $\mathcal{F}$ in (24) and (25) exclusively refers to the corresponding initial temperature.

To compare quantitatively (24) with (4), we note that g_{in} in (4) refers within dynamic cooling to the $\mathbf{S}$-$\mathbf{R}$ (system-reservoir) interaction that is zero both initially and finally. Assume that the final state $\rho_{\mathbf{S}}(\tau)$ of $\mathbf{S}$ is Gibbsian (canonic equilibrium) at temperature T_{fin}. Then the thermodynamic entropy in (3) is the von Neumann entropy from (25). Recalling that $T_{\mathrm{fin}} < T_{\mathrm{in}} \equiv T$ we obtain from (4), (24)

$$F[T_{\mathrm{fin}}, g_{\mathrm{in}}] - F[T_{\mathrm{in}}, g_{\mathrm{in}}] - T S[\rho_{\mathbf{S}}(\tau)||\rho_{\mathbf{S}}]$$
$$= -\mathrm{tr}[\rho_{\mathbf{S}}(\tau) \ln \rho_{\mathbf{S}}(\tau)] (T_{\mathrm{fin}} - T_{\mathrm{in}}) \geqslant 0. \qquad (26)$$

Thus, the difference between (24) and (4) tends to disappear with the final entropy of $\mathbf{S}$. Such a situation will be met in Sec. VI.

E. Summary of conditions

In analogy to Sec. II D we summarize the dynamical cooling setup in terms of the following task list:

(**D1**) initial state, canonical equilibrium system and reservoir (they are uncoupled;

(**D2**) system (target of cooling), microscopic;

(**D3**) cooling dynamics, unitary in the space of the system + reservoir (not necessarily quasiequilibrium);

(**D4**) reservoir, possibly microscopic (not necessarily larger than the system);

(**D5**) system-reservoir interaction (driven by external field), changes cyclically.

D2 makes obsolete the need for the entropic formulation of the third law, since a thermally isolated process done on a finite $\mathbf{S}$ is not uniquely characterized by its entropy [47] (instead the full spectrum of the density matrix determines the set of states that can be reached by unitary processes). In this paper we assumed **D2**[*] rather than **D2**, that is, that the system $\mathbf{S}$ is two-dimensional. This more restrictive condition should be relaxed in future studies. **D5** is motivated in the same way as **N5**. To grasp the difference between **N4** and **D4**, we study in Sec. VI the dynamic cooling setup under condition **N4**.

As for the tractability we keep the conditions **T1**–**T3** from Sec. II D. The unattainability of the absolute zero is proven in Sec. III C via **D1** and **D3**, without explicit reference to **T1**–**T3**. Nevertheless, **T2** is needed below for understanding limitations on reaching lowest (nonzero) temperatures, while **T3** is inherently demanded for applications of the dynamical cooling: The cycle time now refers to the time needed to realize the unitary operator (7) via a suitable Hamiltonian.

IV. OPTIMAL RESERVOIR

The task list of Sec. III E does not yet fully specify the reservoir. In this section and the following two we investigate three different models; their respective parameters are then shown to characterize the way in which cooling can be achieved.

A. Max-min cooling scenario

The purpose of dynamic cooling is to increase the ground-state probability of **S** subject to the constraints listed in Sec. III E. Obviously, some setups work better for the desired task than others. For studying principal limitations we should thus optimize the design, that is, try to maximize this very probability. This has to be done (i) over all unitary transformations U in (8) and (ii) over the energy level distribution of the reservoir assuming that they are bounded from above; that is, μ in (13) is a finite, fixed number. This parameter of the upper energy level of the reservoir turns out below to control dynamic cooling. We denote these maximization strategies as $\max_U$ and $\max_{\mu_k}$, respectively. The overall maximization is $\max_{U,\mu_k}$.

Generally, the above maximization procedure will specify a set of equivalent scenarios only, that is, leave some free parameters (see below). Additional requirements may be imposed to reduce this ambiguity. Here we attempt to minimize the work needed for given cooling effect. This is why our intended goal requires a "max-min" scenario: First the ground-state probability of **S** is maximized and only after that the work is minimized.

B. Maximal cooling

Assume that **R** has an even number of energy levels:

$$M = 2n. \tag{27}$$

Appendix A shows that the unitary operator, which leads to the largest final ground-state probability for **S**, amounts to permuting the elements (15) of the initial density matrix $\rho_{\mathbf{S}+\mathbf{R}}$. We recall that it is diagonal in the energy representation. Hence, in the maximal cooling regime the final state of the two-level target is diagonal in the energy representation: It has a well-defined temperature, which is hence not imposed, but emerges out of optimization.

The fact that the two-level target of cooling ends up in an energy diagonal density matrix (with the ground-level probability greater than the excited level probability) is straightforward to establish: Otherwise, there will be a unitary operator acting only on the two-level system such that its ground-state probability is increased.

Consider the eigenvalue vector of the final state of $\mathbf{S} + \mathbf{R}$ with the largest ground-state probability $p_0(\tau)$ of **S**. In this vector the largest $2n$ elements of the vector (15) are at the odd places [counting starts with 1], and the final, maximized ground-state probability for **S** reads

$$\max_U[p_0(\tau)] = \mathbf{smax}_{2n}\big[\{\omega_k\}_{k=0}^{4n-1}\big], \tag{28}$$

where $\mathbf{smax}_k[\mathbf{a}]$ returns the sum of k largest elements of vector **a**. Finding the $2n$ maximal elements of $\{\omega_k\}_{k=0}^{4n-1}$ leaves some freedom in U. One represents (28) as

$$\max_U[p_0(\tau)] = p_0 \sum_{k=0}^{n-1} \pi_k + p_1\pi_0$$
$$+ \mathbf{smax}_{n-1}[p_1\pi_1, \ldots, p_1\pi_{n-1}, p_0\pi_n, \ldots, p_0\pi_{2n-2}], \tag{29}$$

where we used (15) and straightforward induction over n.

Maximizing (28) over the energy levels (13) amounts to maximizing it over the Boltzmann weights $v_k = e^{-\beta\mu_k}$ [see (13), (14)] under constraints

$$1 \geqslant v_1 \geqslant \ldots \geqslant v_{2n-1} \geqslant v = e^{-\beta\mu}. \tag{30}$$

Since the left-hand side of (28) is a ratio of linear functions of v_k, it can maximize only at the borders of the allowed range of v_k; that is, some of v_k's are equal to 1, while others are equal to v. The physical meaning of this result is that *the optimal reservoir is an effective two-level system*, a fact that greatly simplifies searching for the maximal (final) ground-state probability of **S**. To illustrate this result, consider an example in (29): $n = 2$, $\mathbf{smax}_1[p_1\pi_1, p_0\pi_2] = \max[p_1\pi_1, p_0\pi_2] = p_1\pi_1$. This reduces (29) to $\pi_0 + \pi_1$, which maximizes for $v_1 = 1, v_2 = v$.

It now remains to check all possible arrangements of energy levels that render **R** an effectively two-level system. This check produces the maximum of (29) for

$$\mu_0 = \cdots = \mu_{n-1} = 0, \quad \mu_n = \cdots = \mu_{2n-1} = \mu. \tag{31}$$

Hence, the optimal reservoir for cooling a two-level system has to have a degenerate ground state for $n \geqslant 2$. The maximal ground-state probability now reads

$$\max_{U,\mu_k}[\, p_0(\tau)\,] = \sum_{k=0}^{n-1} \pi_k = \frac{1}{1 + e^{-\beta\mu}} = \frac{1}{1 + e^{-\epsilon/T_{\min}}}, \tag{32}$$

where

$$T_{\min} = T\epsilon/\mu. \tag{33}$$

Thus, (32) is the maximal probability for a reservoir with maximal energy μ; $T_{\min}$ is the minimal temperature. Note that (33) is consistent with the unattainability argument (17). For $\mu \to \infty$, $T_{\min}$ would approach zero.

We get cooling, that is, $\max_{U,\mu_k}[p_0(\tau)] > p_0$, only when the largest energy of **R** is larger than the energy of **S**:

$$\mu > \epsilon. \tag{34}$$

This asymmetry ensures that the two-level is cooled in presence of the reservoir.

C. Minimal work

We turn to calculating the minimal work given the maximal probability (32). Write (15) under (31):

$$\underbrace{p_0\pi, p_1\pi, \ldots, p_0\pi, p_1\pi}_{2n \text{ elements}}, \underbrace{p_0\tilde{\pi}, p_1\tilde{\pi}, \ldots, p_0\tilde{\pi}, p_1\tilde{\pi}}_{2n \text{ elements}}, \tag{35}$$

where we defined $\pi = \frac{1}{n(1+v)}$ and $\tilde{\pi} = \frac{v}{n(1+v)}$.

Equation (32) is found after the first $2n$ elements in (35) are distributed over the odd places in the eigenvalue list of $\rho_{\mathbf{S}+\mathbf{R}}^{\text{fin}}$ [counting starts from 1]. Concrete places they occupy are not important for obtaining the optimal cooling effect (32): Those various orderings constitute an equivalence class. Concrete places, however, become important for minimizing the work. Note that for a fixed final ground-state probability of the two-level system minimizing the work amounts to maximizing the

overall probability of the lowest level $\mu_0 = \cdots = \mu_{n-1} = 0$ for the final state of the reservoir [see (31)]. Then (35) implies that the minimal work is obtained for a permutation that does not touch the underlined elements in (35), but interchanges those that are not underlined: each $p_1\pi$ with some element $p_1\tilde{\pi}$; see Appendix A for the argument reducing the considered unitary to a permutation. Recalling that the initial energy of the system + reservoir is $\frac{\mu e^{-\beta\mu}}{1+e^{-\beta\mu}} + p_1\epsilon$ and using (18) we get

$$W = (\mu - \epsilon)\left[\frac{1}{1 + e^{-\beta\mu}} - p_0\right], \qquad (36)$$

for the work. Now (36) shows that reaching the lowest possible temperature $T_{\min}$ requires the work

$$W \simeq \mu p_1 \sim 1/T_{\min}. \qquad (37)$$

This constitutes a parametrization of the attainability constraints for dynamical cooling: Work W and and the inverse temperature reached, $1/T_{\min}$, are proportional. Accepting that W cannot be infinite (see condition **T2** in Secs. II D and III E), we have to accept that the minimal temperature reached cannot be zero.

Let now the number of energy levels of **R** be an odd number: $M = 2n + 1$. Instead of (29) we get

$$\max_U[p_0(\tau)] = p_0 \sum_{k=0}^{n-1}\pi_k + p_1\pi_0$$
$$+ \mathbf{smax}_n[p_1\pi_1, \ldots, p_1\pi_{n-1}, p_0\pi_n, \ldots, p_0\pi_{2n-1}]. \qquad (38)$$

The final ground-state probability is maximized for

$$\mu_0 = \cdots = \mu_{n-1} = 0, \quad \mu_n = \cdots = \mu_{2n} = \mu, \qquad (39)$$

meaning again that the optimal reservoir has to have a degenerate ground state for $n \geqslant 2$. The maximal ground-state probability reads

$$\max_{U,\mu_k}[p_0(\tau)] = \frac{n + p_0 e^{-\beta\mu}}{n + (n+1)e^{-\beta\mu}}. \qquad (40)$$

Condition (34) is still needed, and the qualitative conclusion from studying the work cost is the same as above.

In contrast to (32), expression (40) already depends on the initial probability p_0 of **S**. Equations (32) is recovered from (40) for a many-level reservoir $n \gg 1$. Instead of assuming such a many level reservoir with the optimal unitary operating on the joint Hilbert space of this reservoir and the target system, we can relate (40) to (32) under a weaker condition. Apply the cooling protocol repeatedly with the reservoir—having a finite, odd number of energy levels—reprepared in its equilibrium state, for example, via fast relaxation, as it happens with electronic spins in solid state NMR [9]. Then the ground-state probability increases iteratively as [see (40)]:

$$p_0^{[l+1]} = \frac{n + p_0^{[l]}e^{-\beta\mu}}{n + (n+1)e^{-\beta\mu}}, \quad l = 1,2,\ldots. \qquad (41)$$

For $l \gg 1$ the result of this iteration converges to (32).

D. Trade-off between maximizing cooling and minimizing work

The existence of the above work cost for dynamic cooling raises the following question: To what extent can we reduce this cost if, given the upper bound on the reservoir energy spectrum [see (13)], we sacrifice some ground-state probability of the target system, that is, instead of reaching (32) we agree to reach a somewhat lower final probability. As compared to (32), this will require a different reservoir and a different unitary transformation for cooling. The answer to this question is that even a small decrease in the achieved ground-state probability can significantly reduce the work cost. We illustrate this fact via an example.

Let us take $M = 2n = 4$, and postulate a unitary operator that permutes the eigenvalues of the initial state (15) so that the final-state eigenvalues read

$$(p_0\pi_0, p_0\pi_2, p_1\pi_0, p_1\pi_2, p_0\pi_1, p_0\pi_3, p_1\pi_1, p_1\pi_3). \qquad (42)$$

The logic of obtaining (42) from (15) is as follows. The first four probabilities $p_0\pi_0, p_1\pi_0, p_0\pi_1, p_1\pi_1$ in (15) are moved to odd places, producing

$$\hat{p}_0(\tau) = \pi_0 + \pi_1 = \frac{1 + e^{-\beta\mu_1}}{1 + e^{-\beta\mu_1} + e^{-\beta\mu_2} + e^{-\beta\mu_3}}, \qquad (43)$$

which is similar to $\pi_0 + \pi_1$ in (32). The remaining four probabilities $p_0\pi_2, p_1\pi_2, p_0\pi_3, p_1\pi_3$ [last four elements in (15)] are then arranged in between without changing their mutual order. The work and the final probabilities for the four reservoir energy levels read

$$W = \epsilon[p_0 - \hat{p}_0(\tau)] + \sum_{k=1}^{3}\mu_k[\pi_k(\tau) - \pi_k], \qquad (44)$$

$$\pi_0(\tau) = p_0(\pi_0 + \pi_2), \quad \pi_1(\tau) = p_1(\pi_0 + \pi_2), \qquad (45)$$

$$\pi_2(\tau) = p_0(\pi_1 + \pi_3), \quad \pi_3(\tau) = p_1(\pi_1 + \pi_3). \qquad (46)$$

Let us assume that

$$\beta\mu_2 = \beta\mu_3 \equiv \beta\mu \gg 1 \qquad (47)$$

is a fixed number. Hence, μ_1 is the only parameter over which we can minimize (44). In the limit $\beta\mu \to \infty$ both (32) and (43) converge to 1:

$$\max_{U,\mu_k}[\,p_0(\tau)\,] \to 1, \quad \hat{p}_0(\tau) \to 1, \qquad (48)$$

while their ratio is a finite number,

$$\frac{1 - \max_{U,\mu_k}[\,p_0(\tau)\,]}{1 - \hat{p}_0(\tau)} = \frac{1}{2}(1 + e^{-\beta\mu_1}) + \mathcal{O}[e^{-\beta\mu}], \qquad (49)$$

where $\mathcal{O}[e^{-\beta\mu}]$ can be neglected due to (47).

We now show that although (32) and (43) are quite close to each other, the work needed to obtain (43) deviates significantly from (36). Note that π_2 and π_3 are exponentially small with $\beta\mu \to \infty$. We neglect such terms in (44) and obtain

$$W = -\epsilon p_1 + \mu_1(p_1\pi_0 - \pi_1) + \mu\pi_1, \qquad (50)$$

$$\beta W = p_1 \ln\frac{p_1}{1 - p_1} + \frac{\beta\mu_1(p_1 - e^{-\beta\mu_1}) + \beta\mu e^{-\beta\mu_1}}{1 + e^{-\beta\mu_1}}. \qquad (51)$$

To get (51) and (50) from (44)–(46) we employed (12) and (14). It should now be clear that for $\beta\mu \gg 1$ we get a logarithmically growing work, $\beta W = p_1 \ln[\frac{p_1\beta\mu}{1-p_1}]$, if we choose $\beta\mu_1 = \ln(\beta\mu)$. The minimization of the right-hand side of (51) over $\beta\mu_1$ satisfying $\mu_1 \leqslant \mu$ produces a very similar result. For $\beta\mu \gg 1$ neglect in (51) all terms with

$e^{-\beta\mu_1}$ except the factor $\beta\mu e^{-\beta\mu_1}$, where a small term $e^{-\beta\mu_1}$ is multiplied by the large $\beta\mu$. Now substitute $\beta\mu_1 = \ln[\frac{\beta\mu}{yp_1}]$ into (51), where y is a new variable. This produces $\beta W = p_1 \ln[\frac{p_1}{1-p_1}] + p_1(y + \ln[\frac{\beta\mu}{yp_1}])$. After differentiating over y we obtain for the minimum: $y = 1$, or

$$\beta W = p_1 \ln\left[\frac{\beta\mu e}{1-p_1}\right]. \tag{52}$$

and thus a logarithmic rather than linear increase of W with μ, that is, with β'. In the limit $\beta\mu \to \infty$ the corresponding ground-state probability is only slightly reduced:

$$\frac{1 - \max_{U,\mu_k}[\,p_0(\tau)]}{1 - \hat{p}_0(\tau)} = \frac{1}{2}\left(1 + \frac{p_1}{\beta\mu}\right). \tag{53}$$

This equation can be rewritten in terms of temperatures. Recall (48) and (32), and introduce a temperature $\hat{T}$ via $\hat{p}_0(\tau) = (1 + e^{-\epsilon/\hat{T}})^{-1}$. Then (53) reads

$$\frac{1}{\hat{T}} - \frac{1}{T_{\min}} = \frac{1}{\epsilon}\ln\left[\frac{1}{2}\left(1 + \frac{p_1}{\beta\mu}\right)\right]. \tag{54}$$

For $\beta\mu \to \infty$ the right-hand side of this equation goes to $-\frac{\ln 2}{\epsilon}$.

If one allows further deviations from the maximal probability (32) [more than (53)], then it is possible to reduce the work even below the logarithmic dependence (52). Appendix B shows that the example (42) generalized to $n = 4$ (eight-level reservoir) produces [for $\beta\mu \gg 1$] the double-logarithmic scaling of the minimal work, $W \simeq \ln[\ln(\beta\mu)]$ provided that the deviation from the maximal probability scales as

$$\frac{1 - \max_{U,\mu_k}[\,p_0(\tau)]}{1 - \hat{p}_0(\tau)} = \frac{1}{4}\left(1 + \mathcal{O}\left[\frac{1}{\ln(\beta\mu)}\right]\right). \tag{55}$$

Realistic reservoirs tend to deviate from the optimal model studied above. In the following we investigate models subject to additional constraints.

V. HOMOGENEOUS SPECTRUM OF RESERVOIR

A. Finite number of levels

We saw above that the maximal energy μ [see (13)] is an essential parameter for dynamical cooling via a reservoir with a finite number of energy levels: Cooling to ground-state probability 1 is possible only for $\mu \to \infty$. In the optimal scenario (32) (with two levels only) μ conincides with the energy gap.

Here we study the simplest cooling scenario, where μ can be large without increasing the level spacing. This scenario also illustrates the limit of the infinite-dimensional Hlibert space for the reservoir.

We assume that **R** has M equidistant energy levels:

$$\mu_k = \delta k, \quad k = 0, 1, \ldots, M-1, \tag{56}$$

where $\delta > 0$ is the reservoir energy gap. We realize with respect to this reservoir the same max-min scenario. However, no optimization over the energy gaps of the reservoir is carried out; that is, δ is a fixed parameter.

The initial-state eigenvalues of $\mathbf{S} + \mathbf{R}$ read

$$\frac{1}{Z}[p_0, p_1, p_0 v, p_1 v, p_0 v^2, p_1 v^2, \ldots, p_0 v^{M-1}, p_1 v^{M-1}], \tag{57}$$

where we defined

$$v = e^{-\beta\delta}, \quad Z = \sum_{k=0}^{M-1} v^k = (1 - v^M)/(1 - v). \tag{58}$$

We define α from

$$p_0 v^\alpha \geqslant p_1 \geqslant p_0 v^{\alpha+1}, \quad \alpha = \left\lfloor \frac{\ln(p_0/p_1)}{\ln(1/v)} \right\rfloor = \left\lfloor \frac{\epsilon}{\delta} \right\rfloor, \tag{59}$$

where $\lfloor x \rfloor$ is the floor integer part of x; for example, $\lfloor 0.99 \rfloor = 0$. Let us reorder the elements of vector (57) (first the largest element, then next to the largest, etc.):

$$\vec{\omega} = \frac{1}{Z}\Bigg[\underbrace{p_0, p_0 v, \ldots, p_0 v^\alpha}_{\alpha+1}, \tag{60}$$

$$\underbrace{p_1, p_0 v^{\alpha+1}, p_1 v, p_0 v^{\alpha+2}, \ldots, p_1 v^{M-\alpha-2}, p_0 v^{M-1}}_{2(M-\alpha-1)}, \tag{61}$$

$$\underbrace{p_1 v^{M-\alpha-1}, p_1 v^{M-\alpha}, \ldots, p_1 v^{M-1}}_{\alpha+1} \Bigg], \tag{62}$$

where the curly bracket shows the number of elements in each group. In (60)–(62) we select the first M elements; their sum will give the maximal final ground-state probability of the target two-level system. Then the optimal unitary transformation amounts to distributing those M largest elements over the odd places in the eigenvalue list of the final density matrix. Clearly, $M = \alpha + 1$ means that no cooling is possible with the considered reservoir.

Let $M - (\alpha + 1)$ be an even number:

$$M - (\alpha + 1) = 2s, \tag{63}$$

where s is an integer. Now (61) is factorized as

$$(61) = \underbrace{p_1, p_0 v^{\alpha+1}, \ldots, p_1 v^{s-1}, p_0 v^{\alpha+s}}_{M-\alpha-1},$$

$$\underbrace{p_1 v^s, p_0 v^{\alpha+s+1}, \ldots, p_1 v^{M-\alpha-2}, p_0 v^{M-1}}_{M-\alpha-1} \tag{64}$$

Thus, the final ground-state probability of the target two-level system reads[10]

$$\max_U[p_0(\tau)] = \frac{1}{Z}\left[p_0 \sum_{k=0}^{\alpha+s} v^k + p_1 \sum_{k=0}^{s-1} v^k \right]$$

$$= \frac{1 - p_1 v^s - p_0 v^{\alpha+s+1}}{1 - v^M}. \tag{65}$$

As expected, (65) is smaller than the bound (32) with $\mu = \delta(M-1)$.

B. Infinite number of levels

For finite δ and α we get from (65):

$$\max_U[p_0(\tau)] \to 1 \quad \text{when} \quad M \to \infty \quad \text{and} \quad s \to \infty. \tag{66}$$

The minimal work necessary for (66) is *finite*. In showing this let us restrict ourselves with the simplest case $\alpha = 0$

---

[10]Likewise, for an odd $M - (\alpha + 1)$ we introduce $M - \alpha - 1 = 2u + 1$, where u is an integer. Instead of (65) we get $\max_U[p_0(\tau)] = \frac{1 - p_1 v^{u+1} - p_0 v^{\alpha+u+1}}{1 - v^M}$, which implies the same consequence (66).

in (63). Equations (60)–(62) now read

$$\underbrace{p_0, p_1, \ldots, p_1 v^{(M-3)/2}, p_0 v^{(M-1)/2}}_{M},$$

$$\underbrace{p_1 v^{(M-1)/2}, p_0 v^{(M+1)/2}, \ldots, p_0 v^{M-1}, p_1 v^{M-1}}_{M}. \quad (67)$$

Equation (67) implies for the minimal work in the limit (66)

$$W = \epsilon[p_0 - p_0(\tau)] + \frac{\delta}{Z}\left(p_1 + p_0 v^{\frac{M+1}{2}}\right) \sum_{k=0}^{(M-3)/2} (2k+1)v^k$$

$$+ \frac{\delta}{Z}\left(p_0 v + p_1 v^{\frac{M+1}{2}}\right) \sum_{k=0}^{(M-3)/2} (2k+2)v^k - \frac{\delta}{Z} \sum_{k=0}^{M-1} k v^k$$

$$= (\delta - \epsilon)p_1 + \frac{\delta v}{1-v}, \quad (68)$$

which is clearly finite, positive and is larger than the bound $\Delta F = -\frac{1}{\beta}\ln(1 + e^{-\beta\epsilon})$ demanded by (24).

The convergence (66) does not mean that for $M = \infty$ (harmonic oscillator spectrum) we would get $\max_U[p_0(\tau)] = 1$ (or $T_{\min} = 0$), because for $M = \infty$ the transformation that leads to (65) is not even bijective, let alone unitary (i.e., the limit of unitary processes for $M \to \infty$ is not unitary); see in this context our discussion after (17). Let us take $\alpha = 0$ in (60)–(62). For a finite M the optimal unitary—which we recall amounts to a permutation—distributes the first half of the vector (60)–(62) over the odd places in the final vector. The elements from the second half are distributed into even places of the final vector. For $M = \infty$ this second (sub)process disappears in infinity. Hence, the limit of the above permutation for $M \to \infty$ is not even bijective.

Cooling with the harmonic oscillator reservoir ($M = \infty$) is a well-defined problem provided that one ensures that the operator U stays unitary for $M = \infty$. For each such unitary we have $p_0(\tau) < 1$ [see (17)]. However, the maximum $\max_U[p_0(\tau)]$ now does not exist. It can be substituted by supremum

$$\sup_U[p_0(\tau)] = 1, \quad (69)$$

as follows from (66). Equation (69) points out on an important difference between the infinite and finite-level situations.

VI. RESERVOIR CONSISTING OF N IDENTICAL SPINS

We study this modular case for two reasons: First, in the *thermodynamic limit* $N \to \infty$ the reservoir will become a standard thermal bath; hence, one expects to establish connections between the Nernst setup (Sec. II) and the dynamic cooling setup (Sec. III). Second, the model is relevant for polarization transfer experiments, and thus was widely studied—albeit for the high-temperature limit only—in the NMR literature [11,16].

A. Maximal cooling

Now reservoir **R** consists of N identical spins, each one with energies 0 and $\delta > 0$. The energy levels of the reservoir,

$E_A = \delta A$, are equidistant with degeneracy

$$d_A = \frac{N!}{(N-A)!\, A!}. \quad (70)$$

One can apply (60)–(62) to this situation—with α being defined as in (59)—but now each element $p_i v^A$ has to be repeated d_A times. The indices under the curly brackets in (60)–(62) indicate now the number of distinct elements. Again, we need to divide the vector $\vec{\omega}$ in (60)–(62) into two equal parts. For simplicity [and without altering the asymptotic formulas (75), (76)] assume that N is an odd number.

If α is an even number, dividing $\vec{\omega}$ into two equal parts amounts to finding an integer m that satisfies

$$\sum_{k=0}^{\alpha} d_k + \sum_{k=1}^{m} d_{\alpha+k} + \sum_{k=0}^{m} d_k = 2^N, \quad (71)$$

since now the first half of $\vec{\omega}$ ends up with element $p_1 v^m$. This leads to $m = \lfloor \frac{N}{2} \rfloor - \frac{\alpha}{2}$, where $\lfloor x \rfloor$ is defined after (59).

If α is an odd number, the first half of $\vec{\omega}$ ends up with element $p_0 v^{\alpha+m}$, and $m = \lfloor \frac{N}{2} \rfloor - \lfloor \frac{\alpha}{2} \rfloor$ is found from

$$\sum_{k=0}^{\alpha} d_k + \sum_{k=1}^{m} d_{\alpha+k} + \sum_{k=0}^{m-1} d_k = 2^N.$$

For simplicity we focus on the even α case (71). Then taking in (60)–(62) all the initial elements (together with their degeneracies) up to the index m means dividing it into two equal parts. Thus, the largest ground-state probability achievable is equal to the sum of the elements from the first half of $\vec{\omega}$:

$$\max_U[p_0(\tau)] = \frac{1}{(1+v)^N}\left[p_0 \sum_{k=0}^{\alpha+m} d_k v^k + p_1 \sum_{k=0}^{m} d_k v^k \right],$$

$$m = \left\lfloor \frac{N}{2} \right\rfloor - \frac{\alpha}{2}. \quad (72)$$
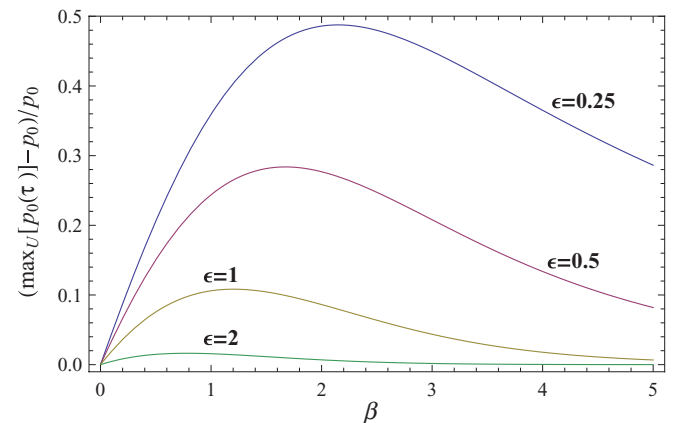
Examples of (72) are presented in Fig. 2.

FIG. 2. (Color online) The relative cooling $\frac{\max_U[p_0(\tau)]-p_0}{p_0}$ calculated according to (12) and (72) versus the initial inverse temperature $\beta = 1/T$ for various values of ϵ and $N = 201$, $\delta = 0.1$. The relative cooling is maximal at some intermediate $\beta(\epsilon)$. The maximum is sharper for smaller values of ϵ.

Let us now take the limit $N \gg 1$. Despite this limit both ϵ and δ are finite; we neglect α in (72) by taking there $m = \lfloor \frac{N}{2} \rfloor$ [recall that $\alpha = 0$ for $\epsilon < \delta$] and transform (72) via

$$\frac{1}{(1+v)^N} \sum_{k=0}^{m} d_k v^k = \frac{\sum_{k=0}^{N} d_k v^k - \sum_{k=m+1}^{N} d_k v^k}{(1+v)^N}$$

$$= 1 - \frac{v^{m+1} d_{m+1}}{(1+v)^N} \sum_{k=0}^{N-m-1} v^k \frac{d_{k+m+1}}{d_{m+1}}. \quad (73)$$

Since $m = \lfloor \frac{N}{2} \rfloor > Nv/(1+v)$, $v^k d_{k+m+1}$ is a decaying function of k; hence, $\sum_{k=0}^{N-m-1} v^k \frac{d_{k+m+1}}{d_{m+1}}$ is dominated by its first few terms and is bounded from above for $N \to \infty$.[11] We are left with estimating $\frac{v^{m+1} d_{m+1}}{(1+v)^N}$ for $m = \frac{N}{2}$. Using the Stirling's formula (74)

$$N! \simeq \sqrt{2\pi N}(N/e)^N, \quad (74)$$

we get for $N \gg 1$

$$\frac{1}{N} \ln\{1 - \max_U [p_0(\tau)]\} = -S\left[\frac{1}{2} I || \sigma\right] + \mathcal{O}\left(\frac{\ln N}{N}\right), \quad (75)$$

where the relative entropy $S[\frac{1}{2} I || \sigma]$ between the probability vectors $(\frac{1}{2}, \frac{1}{2})$ and $(\frac{v}{1+v}, \frac{1}{1+v}) = \sigma$ satisfies

$$S\left[\frac{1}{2} I || \sigma\right] = \ln \frac{1+v}{2\sqrt{v}} = \ln \cosh\left[\frac{\beta\delta}{2}\right] \quad (76)$$

[see also (21)]. Note that σ is the state of the single reservoir spin [the reservoir consists of N such identical spins]. Note that for $N \gg 1$ the asymptotic behavior of $p_0(\tau)$ does not depend on ϵ.

Thus, for $N \to \infty$, the (small) deviation of $p_0(\tau)$ from 1 is controlled by the relative entropy (76). The final temperature of the two-level system reads from (75) and (76)

$$T_{\min} = \frac{\epsilon}{N S[\frac{1}{2} I || \sigma]}. \quad (77)$$

Note that (77) is consistent with the unattainability argument (17).

Let us now turn to calculating the minimal work needed for this cooling.

B. The minimal work

The message of the following calculations is to show that the minimal work required for cooling (75) and (76) *can* converge to its thermodynamic lower bound (24) in the thermodynamic limit $N \gg 1$.

We start by the case $\epsilon \to 0$. Conceptually, this case of degenerate target system energy levels is interesting, because no brute force method (i.e., a low-temperature bath) can ever increase the occupation of one of the energy levels. In addition, the work done for cooling is not blurred by the energy released from the two-level system. Starting with this case is also useful for technical reasons, moreover that $\epsilon > 0$ will not bring essential news. The reader not interested in technical details can immediately turn to (93) and the discussion afterward.

As follows from the discussion around (71), the vector (60)–(62) can be written via its two halves as

$$\vec{\omega} = (\vec{a}, \vec{b}), \quad (78)$$

$$\vec{a} = \frac{1}{2(1+v)^N}[v^0, v^0, \ldots, v^{[N/2]}, v^{[N/2]}], \quad (79)$$

$$\vec{b} = \frac{1}{2(1+v)^N}[v^{[N/2]+1}, v^{[N/2]+1}, \ldots, v^N, v^N], \quad (80)$$

$$\vec{c} = [0, 1, 2, \ldots, N], \quad (81)$$

and where $\vec{c}$ is the vector of the reservoir energies. Recall that in (79) and (80) each factor v^k is repeated d_k times. Likewise, in (81) each symbol k is repeated d_k times. Vectors $\vec{a}$, $\vec{b}$ and $\vec{c}$ have equal number of components 2^N.

The discussion around (71) implies that $\vec{a}$ contains the largest 2^N elements of the vector (60)–(62). Thus, the minimal final reservoir energy needed for the optimal cooling (75) is the following inner product:

$$\delta(\vec{a} + \vec{b})\vec{c}. \quad (82)$$

Once reservoir's initial energy $\delta N v/(1+v)$ is known, the initial and final energies of the target spin are known as well, the minimal work is determined via (82).

Note that $d_k v^k$ is peaked around

$$k = \left\lfloor N \frac{v}{1+v} \right\rfloor \equiv N\xi, \quad (83)$$

where $\lfloor x \rfloor$ is defined after (59), $v = e^{-\beta\delta}$ and the difference $|\xi - \frac{v}{1+v}|$ fluctuates for different $N \gg 1$ only by the amount $1/N$. Hence, in $\vec{a}$ we can focus on dominant energy levels

$$\{\delta(N\xi + m)\}_{m=-\Delta}^{\Delta}, \quad (84)$$

where Δ is determined by requiring that the ratio $\frac{d_{N\xi \pm \Delta} v^{N\xi \pm \Delta}}{d_{N\xi} v^{N\xi}}$ of the central (maximal) probability to the probability at the edge of the interval (84) is exponentially (over N) small. This guarantees that considering only (84) suffices for calculating quantities that are not exponentially small. Employing for $N \gg 1$ Stirling's formula (74), we get

$$\frac{d_{N\xi \pm \Delta} \ v^{N\xi \pm \Delta}}{d_{N\xi} \ v^{N\xi}} = e^{-\frac{\Delta^2}{2\xi(1-\xi)N} + \mathcal{O}(\frac{\Delta^3}{N^2}) + \mathcal{O}(\frac{\Delta}{N})}. \quad (85)$$

For achieving the sought exponential smallness we need to require $\Delta = N^{a+1/2}$, where a is a fixed small number; for example, $a = 1/10$. Hence, the total probability of energy levels that do not fall in the interval (84) is $\mathcal{O}[Ne^{-N^{2a}}] \to 0$. Now in (82) we can neglect $\vec{b}\vec{c}$ and keep in $\vec{a}\vec{c}$ only those elements of $\vec{a}$ whose energies lay in the interval (84).

Within interval (84) we select a segment consisting of $v^{m+N\xi}$'s only. In $\vec{a}$ it occupies positions with numbers from

---

[11]For numerics one can use the hypergeometric function $\sum_{k=0}^{N-m-1} v^k \frac{d_{k+m+1}}{d_{m+1}} = {}_2F_1[1, m-N+1, 2+m, -v]$ that holds for any $N - m - 1 > 0$. This formula is derived by expressing $\frac{d_{k+m+1}}{d_{m+1}}$ via Euler's γ functions [recall (70)], extending the summation to infinity: $\sum_{k=0}^{N-m-1} = \sum_{k=0}^{\infty}$, and employing the standard definition ${}_2F_1(a, b, c; z) = \sum_{k=0}^{\infty} \frac{(a)_k(b)_k}{(c)_k} \cdot \frac{z^k}{k!}$ of the hypergeometric function [48]. Here $(a)_k = \frac{\Gamma(a+k)}{\Gamma(a)}$ and $(-a)_k = (-1)^k \frac{\Gamma(a+1)}{\Gamma(a-k+1)}$ are the Pochhammer symbols, and $\Gamma(z)$ denotes the γ function.

$1 + \sum_{k=0}^{N\xi+m-1} 2d_k$ to $\sum_{k=0}^{N\xi+m} 2d_k$. Its length is $2d_{m+N\xi}$. It appears that there are only two energies $\Phi_m - 1$ and Φ_m that correspond to that segment in $\vec{c}$. We write $\Phi_m = N\xi + m + \ell_m$, where ℓ_m is the minimal integer that satisfies

$$\sum_{k=0}^{N\xi+m+\ell_m-1} d_k - \sum_{k=0}^{N\xi+m-1} 2d_k \equiv D_m \geqslant 0. \quad (86)$$

The reason for having only two energies for each segment in the dominant interval (84) is that ℓ_m does not depend on m, as we see below.

Each sum in (86) is dominated by its largest summand. Recalling from (70)

$$\frac{d_{k+1}}{d_k} = \frac{1 - \frac{k}{N}}{\frac{k}{N}} \left[1 + \mathcal{O}\left(\frac{1}{N}\right) \right], \quad (87)$$

we get a general pattern for approximating such sums[12]:

$$\sum_{k=0}^{N\xi} d_k = \frac{d_{N\xi}}{1-v} \left[1 + \mathcal{O}\left(\frac{1}{N}\right) \right]. \quad (88)$$

Equation (86) rewrites as [neglecting factors $\mathcal{O}(\frac{1}{N})$]

$$\sum_{k=N\xi+m+1}^{N\xi+m+\ell_m} d_k = d_{N\xi+m} \sum_{k=1}^{\ell_m} \frac{1}{v^k} \geqslant \sum_{k=0}^{N\xi+m} d_k = \frac{d_{N\xi+m}}{1-v}. \quad (89)$$

These relations imply

$$\ell_m = \ell = \left\lceil \frac{\ln 2}{\ln \frac{1}{v}} \right\rceil = \left\lceil \frac{\ln 2}{\beta\delta} \right\rceil, \quad (90)$$

where $\lceil x \rceil$ is the ceiling integer part of x; for example, $\lceil 0.99 \rceil = 1$, $\lceil 1.1 \rceil = 2$. As anticipated, ℓ_m does not depend on m.[13] Hence, we write for the minimal work (82)

$$\delta \vec{a}\vec{c} = \frac{\delta}{2(1+v)^N} \sum_{m=-\Delta}^{\Delta} v^{N\xi+m}[D_m(N\xi + m + \ell - 1) \\ + (2d_{N\xi+m} - D_m)(N\xi + m + \ell)], \quad (91)$$

where D_m is defined in (86). Equation (89) implies

$$D_m = d_{N\xi+m} \frac{v(v^{-\ell} - 2)}{1-v} \left[1 + \mathcal{O}\left(\frac{1}{N}\right) \right]. \quad (92)$$

Note that $2d_{N\xi+m} - D_m > 0$ for the considered range of m.[14] Using (92) we get for (91)

$$\delta \sum_{m=-\Delta}^{\Delta} \frac{d_{N\xi+m} v^{N\xi+m}}{(1+v)^N} \left[N\xi + \ell - \frac{v(v^{-\ell} - 2)}{2(1-v)} + m \right]$$
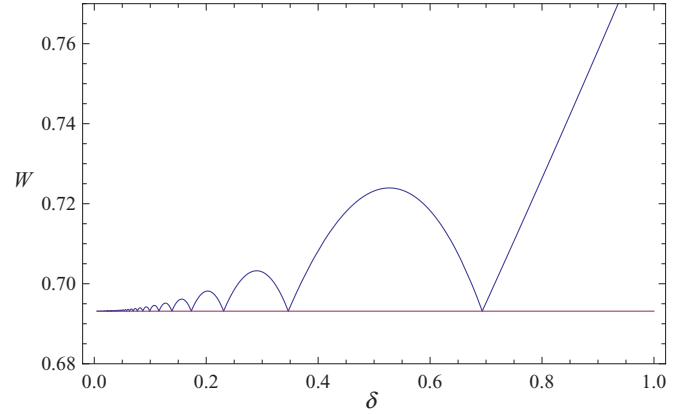

FIG. 3. (Color online) The minimal work $W(\delta)$ for $\epsilon = 0$ and $T = 1$. The horizontal line is $T \ln 2$, the lower bound (24) for $W(\delta)$.

Neglecting[15] $\sum_{m=-\Delta}^{\Delta} \frac{md_{N\xi+m}v^{N\xi+m}}{(1+v)^N}$ we get for the work

$$W = \delta \left[\ell - \frac{v(v^{-\ell} - 2)}{2(1-v)} \right], \quad (93)$$

where $v = e^{-\beta\delta}$ and ℓ is defined in (90). Equation (93) implies several important conclusions.

(i) The initial and final reservoir energy differ from each other by a factor $W = \mathcal{O}(1)$; hence, the state of each reservoir particle changes by a quantity of order $\mathcal{O}(1/N)$, negligible for $N \gg 1$.

(ii) When $\frac{\ln 2}{\beta\delta}$ approaches to an integer number from below, (90) and (93) predict

$$W \to T \ln 2. \quad (94)$$

According to (4) and (24) [and recalling that $\epsilon \to 0$] $T \ln 2$ is the minimal possible work—both within the Nernst setup and within the dynamic cooling—necessary to cool from the absolutely disordered state [the equilibrium state with $\epsilon \to 0$] to an almost ordered state (75). Hence, the thermodynamic bound (24) is reachable for certain *finite* δ's. The behavior of $W(\delta)$ is shown in Fig. 3. For a finite N, W is larger than $T \ln 2$ even if $\frac{\ln 2}{\beta\delta}$ is an integer (see Table I for numerical results).

(iii) For $\delta > T \ln 2$, (93) yields $W(\delta) = \frac{\delta}{2(1-v)}$: Now the minimal work monotonically increases with δ (reservoir energy gap). Recall from (75) and (76) that for a large but finite N the deviation of $\max_U [p_0(\tau)]$ from 1 is controlled by $S[\frac{1}{2}I||\sigma]$, which also grows with δ. Thus, enhanced cooling demands more work. Moreover, one observes

$$W \approx \Delta F + T S[\rho||\sigma], \quad \text{for} \quad T \ln 2 \leqslant \delta, \quad (95)$$

---

[12]Note that both (87) and (88) contain error $\mathcal{O}(\frac{1}{N})$. The error $\mathcal{O}(\frac{1}{N})$ from (87) does not accumulate in (88), because the sum $\sum_{k=0}^{N\xi} d_k$ is dominated by few (smaller than N) terms around $k = N\xi$.

[13]This fact together with (90) and the reasoning above it makes it possible to guess an upper bound $\delta\ell$ for the work W. This bound is confirmed by (93) and makes it possible to deduce quickly the fact of (94).

[14]In view of (92) this reduces to $\frac{2}{v} > \frac{v^{-\ell} - 2}{1-v}$.

[15]Recall (70) and denote $\widetilde{d}_A = \frac{(N-1)!}{(N-A-1)!A!}$. Put $m = N\xi + m - N\xi$ in $\sum_{m=-\Delta}^{\Delta} \frac{md_{N\xi+m}v^{N\xi+m}}{(1+v)^N}$ and using $kd_k = N\widetilde{d}_{k-1}$ write it as

$$\xi N \left\{ \sum_{m=-\Delta-\widetilde{\xi}}^{\Delta-\widetilde{\xi}} \frac{\widetilde{d}_{\widetilde{N}\xi+m} v^{\widetilde{N}\xi+m}}{(1+v)^{\widetilde{N}}} - \sum_{m=-\Delta}^{\Delta} \frac{d_{N\xi+m}v^{N\xi+m}}{(1+v)^N} \right\},$$

where $\widetilde{N} = N - 1$ and $\widetilde{\xi} = 1 - \xi$. Each sum in the curly brackets is equal to 1 minus exponentially small terms [cf with (85)].

TABLE I. The minimal work W as a function of N for $\delta = \ln 2 = 0.693\,147$, $\epsilon = 0$, and $T = 1$.

N	W
$N = 601$	$0.716\,891$
$N = 1001$	$0.711\,592$
$N = 1401$	$0.708\,766$
$N = 2001$	$0.706\,23$

where $\Delta F = T \ln 2$ is the free-energy difference from (4) and (24), $\rho = \frac{1}{2} I$ [for $\epsilon = 0$] is the initial state of the target two-level system, and σ is the initial state of any reservoir spin. Equation (95) becomes exact for $\delta \gg T \ln 2$.

The case $\epsilon > 0$ does not require any new idea as compared to the above derivation; only algebraic steps are more tedious. Hence, we quote only the final result for the minimal work:

$$W = \begin{cases} \delta\left[\ell - \frac{f - u\alpha}{1+u}\right] - \frac{\epsilon u}{1+u} & \text{for} \quad f \leqslant 1, \\[2mm] \delta\left[\ell - \frac{1 + u v^{-\alpha}(f-1) - u\alpha}{1+u}\right] - \frac{\epsilon u}{1+u} & \text{for} \quad f > 1, \end{cases} \quad (96)$$

where $v = e^{-\beta\delta}$, $u = e^{-\beta\epsilon}$, α is defined in (59), and where

$$\ell = \left\lceil \frac{\ln(1 + v^\alpha)}{\ln \frac{1}{v}} \right\rceil, \quad f = \frac{v(v^{-\ell} - 1 - v^\alpha)}{1 - v}. \quad (97)$$

For $\epsilon = 0$ both branches of (96) are equal, so no conditioning is needed.

Our conclusion on the reachability of the thermodynamic lower bound (24) remains valid also for $\epsilon > 0$: Consider $\frac{\ln(1 + v^\alpha)}{\ln \frac{1}{v}}$ reaching an integer number from below and $\frac{\epsilon}{\delta}$ reaching an integer number from above. Then (96) produces $W = T \ln(1 + e^{-\beta\epsilon})$. This is the bound (24) [recall (75), (76)].

C. Microcanonical initial state of the reservoir

1. Unattainability

So far we considered the reservoir starting its evolution from a canonical equilibrium state (5) and (6). Another notion of equilibrium is provided by the microcanonical density matrix, where all the energy levels in the vicinity $[E - \kappa, E + \kappa]$ of a given energy E have equal probability, while all other energy levels have zero occupation. While the canonical state describes a system prepared via a thermal bath, the microcanonical state refers to a closed macroscopic system whose energy is fixed up to a small uncertainty 2κ [46,49]. Hence, κ should be large enough for $[E - \kappa, E + \kappa]$ to incorporate many energy levels and account for unavoidable environmental perturbations, but small compared with E [46,49].

The canonical and microcanonical notions of equilibria are normally equivalent for macroscopic observables of systems containing sufficiently many particles (see [50] for a review). The microcanonical state can apply for systems with a finite number particles provided that the interaction Hamiltonian does have certain chaoticity features (see [49,51] and references therein). There are situations—such as systems in the vicinity of phase transitions, or systems with long-range interactions–where the equivalence between canonical and microcanonical state is broken even for macroscopic observables

(see, e.g., [50,52,53] and [54] for review). Generally, these states are different regarding some important aspects [52–54]. In particular, the local stability conditions (i.e., stability with respect to small perturbations) for the microcanonic state are less restrictive than for the canonic state (with the same temperature) [54]. Hence, in several physically interesting situations the canonic state is unstable, but the microcanonic state is stable and serves as the only description of the equilibrium [52–54].

For the reservoir consisting of $N \gg 1$ identical spins the microcanonical state concentrated at the energy δK with $K \gg 1$ is especially easy to define: The d_K degenerate energy levels δK [see (70)] have equal probability $\frac{1}{d_k}$, while all other energies have zero probability. Because the energy level δK is already exponentially degenerate we restricted ourselves with the minimal width $\kappa \to 0$.

Given this initial state of the reservoir it is easy to see using the same construction as in (60)–(62) [see our discussion after (70)] that the maximal final ground-state probability for the two-level system interacting with such a reservoir is just equal to 1:

$$\max_U [\, p_0(\tau) \,] = 1. \quad (98)$$

The result (98) does not require $N \to \infty$ or $\kappa \to 0$. It is also not specific to the microcanonical density matrix. Any density matrix for an M-level reservoir that is diagonal in the energy representation, and that has at least $\lfloor \frac{M}{2} \rfloor$ zero eigenvalues would lead to (98) (cf. with our discussion in Sec. III C). For example, it also applies to the so-called θ ensemble, where all the energy levels below a certain E are equally populated, while the energies above E have zero probability.

Equation (98) was obtained via the standard definition of the microcanonical state, but it contains an essential idealization: It is assumed that at least $\lfloor \frac{M}{2} \rfloor$ energies have exactly zero probability. Consider the emergence of the microcanonic state. Classically, any single system is in a state with definite energy. Weak and inevitable interactions with environment smear this energy over the interval 2κ. This, however, does not ensure that energies outside $[E - \kappa, E + \kappa]$ have strictly zero probability.

Quantum mechanically, we cannot know the state of a single system unless we prepared it, for example, by measurement. One general method of preparing a microcanonical state is to do a selective measurement of energy and then isolate the system.[16] Generally, after a selective measurement all energies will be populated, because the measurement itself may be noisy due to our inability to control the measurement interaction and/or the system Hamiltonian. Alternatively, the system may be weakly coupled to its environment prior to the energy measurement, which then necessarily refers to measuring the local energy value. For an open system

---

[16]The selection will produce (Luders' postulate) a density matrix $\propto \Pi(E, \kappa) \rho_{\text{in}} \Pi(E, \kappa)$, where ρ_{in} is the initial state of the system and where $\Pi(E, \kappa)$ is the projector on the Hilbert subspace with energies $[E - \kappa, E + \kappa]$. If ρ_{in} was a sufficiently smooth function of the Hamiltonian (e.g., a Gibbs state), a microcanonic state results. If ρ_{in} was an arbitrary state we face an additional problem of relaxation toward the microcanonic state.
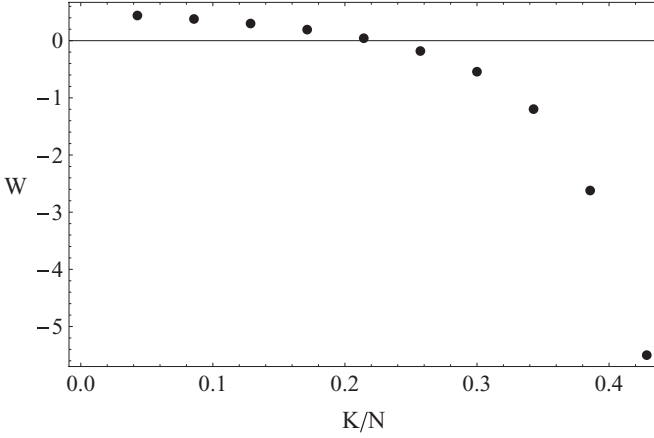
FIG. 4. The minimal work W (needed for maximal cooling) versus the initial energy δK of the microcanonical state, where the initial state of the N-spin reservoir is the mixture of $\frac{N!}{K!(N-K)!}$ energy eigenstates with the energy δK. We considered $N = 7 \times 10^4$, $\delta = 1$, $\epsilon = 0$.

the latter observable is not strictly conserved.[17] Again, both these mechanisms will generally populate energies outside the interval $[E - \kappa, E + \kappa]$. It now suffices to give a small, but nonzero probability to more than $\lfloor \frac{M}{2} \rfloor$ energy levels, and $\max_U [p_0(\tau)]$ in (98) will be smaller than one, as we saw in Sec. III C.

Hence, the unattainability of $T = 0$ for the microcanonic state of the reservoir is recovered provided the microcanonic state contains such tails. Admittedly, we currently lack any general condition for how small these tails of the microcanonic state could be made. It is also unclear which *generic* (not *ad hoc*) conditions one has to impose on the coupling to environment or on the measurement noise to ensure the tails needed for unattainability. In this context one anticipates an additional tractability condition **T4** – referring to a finite environmental coupling or a finite measurement noise—to be added to conditions **T1**–**T3** in Secs. II D and III E.[18]

2. Differences between canonic and microcanonic reservoir

Note the difference: The reachability of low temperatures for a canonical reservoir with a fixed upper energy would require an ever increasing energy gap (related to an ever increasing work cost), while for a microcanonic reservoir (with a fixed upper energy again) reaching low temperatures requires small tails around the central energy.

Another essential difference is that there is no work cost associated with the microcanonic reservoir: The minimal work

---

[17] If the environmental coupling is not weak, the local energy has to be properly defined; see [49] for a concrete proposal (LEMBAS principle).

[18] Conversely, if the unattainability of the absolute zero is regarded to be a law of nature, it can constrain the microscopic processes of environmental coupling and selective measurement. Although this possibility contradicts to the current paradigm of deriving the laws of thermodynamics from microscopic theories, it needs to be taken seriously.

necessary for the maximal cooling need not be positive (i.e., no work has to be consumed) (see Fig. 4). This is related to the general fact that the microcanonical ensemble is not passive [43,55]: There exists for it a class of unitary operations (generated by a suitable cyclically changing Hamiltonians) that leads to sizable work extraction even from a macroscopic microcanonic reservoir [56]. Importantly, the unitary realizing the maximal cooling for the microcanonical reservoir can be put into that class, as Fig. 4 shows.

VII. SUMMARY

In this paper we have studied dynamical cooling of a two-level system (as target) in contact to various types of quantum reservoirs prepared in various types of initial states. Based on operational ideas these schemes have been placed within the context of given resource constraints; the operational meaning of the third law can thus be clarified.

A. Canonical initial state: Complementarity between cooling and work

Unattainability of $T = 0$ is a direct consequence of the dynamic cooling setup (see Sec. III C). In contrast to the unattainability formulation of the third law, it does not involve unproven or disguised assumptions. For a given reservoir with a fixed upper energy starting its evolution from a canonical equilibrium, and for a two-level system as the target of cooling, we explicitly predict the lowest nonzero temperature (32) achievable within dynamic cooling. The lowest temperature (32) would approach zero whenever the upper energy of the reservoir goes to infinity.

We have been able to reinterpret this functional dependence in terms of the work intake: Reaching the lowest temperature demands work growing as the inverse of this temperature [see (36)]. However, this linear relation only applies for reaching that nominal lowest temperature possible: Compromising for a slightly increased final target temperature would allow to change the work growth regime from the linear to (multiple) logarithmic (see Sec. IV D).

Next, we studied concrete models of reservoir. For them we found the minimal temperature attainable (and the minimal work necessary to reach it) by optimizing over cooling dynamics. In Sec. VI we modeled the reservoir via a thermal bath consisting of a large number, $N \gg 1$, of identical spins and established relations between the resources of cooling and certain relative entropies. The lowest temperature $T_{\min}$ reachable with such a reservoir scales as the inverse of N and the relative entropy $S[\frac{1}{2} I || \sigma]$, where I is the 2×2 unit matrix and σ is the initial state of a bath spin [see (77)]. The minimal work W needed to reach this temperature has basically two different expressions. If the reservoir gap is sufficiently larger than temperature, $\delta \gtrsim T$, we get $W \to \Delta F + S[\rho || \sigma]$, where ΔF is the free energy difference of the cooled two-level system, and where ρ is its initial (hot) state. Recall that ΔF is the minimal work required by thermodynamics for achieving cooling [see (24) and (4)]. If $\delta \lesssim T \ln 2$, W is close to $\Delta F = T \ln 2$. Hence, enhancing cooling demands a larger work, but a substantial cooling can already be achieved with the minimal work ΔF demanded by the second law. We

should thus stress that approaching $T = 0$, while using only the amount of work ΔF, is feasible for several limits of reservoir parameters.

Note that the existence of the lowest temperature is related to a finite upper energy of the reservoir. Either it really has this feature, or one considers only that class of cooling operations, where only finite reservoir energies couple to the target of cooling. If the upper energy of the reservoir is infinite (and one allows system-reservoir interactions coupling to arbitrary high reservoir energies), the unattainability of $T = 0$ is still there, but the minimum temperature does not exist. Instead, the infimum temperature is zero (see Sec. V B).

B. Microcanonical initial state

The unattainability of $T = 0$ can be recovered also for a reservoir starting its evolution from a microcanonical state (see Sec. VI C). However, the origin of the strictly nonzero minimal temperature is different: It relates to tails of the energy distribution resulting from a weak interaction with the environment (and thus our inability to avoid such marginal couplings). Put differently, the difference between canonical and microcanonical reservoir concerns the physics of the $T \rightarrow 0$ limit. Low temperatures in the canonical situation require a large energy gap in the reservoir. For the microcanonical case, low temperatures require a sufficiently large system which can be prepared in a state with an almost definite energy. There is no work cost involved in cooling via a microcanonic reservoir.

C. Comparison with other approaches

Reference [21] studies a cooling setup with three components: the system **S** to be cooled, the resource **R**, and the environment **E** with Hamiltonians $H_{\mathbf{S}}$, $H_{\mathbf{R}}$, and $H_{\mathbf{E}}$, respectively. **S** and **E** start in, respectively, the Gibbs states $\gamma_{\mathbf{S}}$ and $\gamma_{\mathbf{E}}$ with the same fixed initial temperature T_{in}. Then [21] allows only for cooling operations that are unitary on the Hilbert space of $\mathbf{S} + \mathbf{R} + \mathbf{E}$ and commute with the interaction free joint Hamiltonian $H_{\mathbf{S}} + H_{\mathbf{R}} + H_{\mathbf{E}}$. Hence, no work cost is related to implementing this unitary and cooling is only possible if the resource deviates from its Gibbs state $\gamma_{\mathbf{R}}$ at the same temperature T_{in}; that is, a resource being in its Gibbs state is "worthless" [see in this context (22)–(24)]. If the resource $\mathbf{R}_N := \mathbf{R}^{\times N}$ consists of N identical copies, the lowest temperature that can be achieved for **S** is asymptotically determined by an analog of (77), where instead of $S[\frac{1}{2}I||\sigma]$ one has $S[\gamma_{\mathbf{R}}||\rho_{\mathbf{R}}]$ [21]. Here $\gamma_{\mathbf{R}}$ is the Gibbs state and $\rho_{\mathbf{R}}$ the actual state of one resource copy. This relation between (77) and the results of [21] relates to the fact that in our present setting the reservoir that consists of N identical subsystems can (asymptotically) be used for cooling if and only if the subsystems are not in their maximally mixed state $\frac{1}{2}I$ (see Sec. VI). Hence, relative entropies other than free energies determine the value of a resource (reservoir) both in the present paper and in [21]. Note that the relative entropy $S[\gamma_{\mathbf{R}}||\rho_{\mathbf{R}}]$ must not to be confused with $S[\rho_{\mathbf{R}}||\gamma_{\mathbf{R}}]$, which is essentially the free energy up to constants. As opposed to free energy, $S[\gamma_{\mathbf{R}}||\rho_{\mathbf{R}}]$ diverges if $\rho_{\mathbf{R}}$ gets a zero eigenvalue. Thus, the absolute zero is reachable only for a resource state having a zero eigenvalue, an aspect closely related to the microcanonic treatment of Sec. VI C.

Alternatively, the features of the limit $T \rightarrow 0$ can be studied via refrigerators that cyclically operate between two thermal baths at temperatures T_c and T_h ($T_c < T_h$) and cool the (finite) low-temperature bath at the expense of consuming work from an external source [22–26]. If the refrigerator works at a finite efficiency, then for $T_c \rightarrow 0$ the heat taken per unit of time from the low-temperature bath scales as T_c^{a+1} with $a \geqslant 0$ [23,24], showing that cooling the low-temperature bath is progressively slowed down. The optimal behavior $a = 0$ is reached for the refrigerator model studied in [26].

D. Open issues

An important aspect of practical implications is the issue of time required to complete the cooling transformation. In our present operational analysis we have dealt with this problem rather formally, assuming that once the needed unitary transformation is constructed, there is always a (time-dependent) Hamiltonian that realizes it. Provided that we do not restrict the magnitude of the external fields realizing the Hamiltonian, this realization could, in principle, take an arbitrary short time; that is, the external fields can be assumed to function in the pulsed regime. However, if we demand that the optimal cooling unitary transformations are implemented in terms of a well-defined base of *realizable* unitary operations, the cooling process may turn out to be complex: It may take a long sequence of the realizable operations to construct the needed unitary. This complexity (and hence time) resource needs further studies.

Another open issue concerns target systems with more than two energy levels. This should be interesting especially with respect to nonequilibrium aspects. Moreover, cooling to very low temperatures may, in fact, demand considering many-body targets of cooling, because at such low temperatures the standard assumptions of the weak coupling between the target and the reservoir—as well as between different parts of the target – may be broken [57].

ACKNOWLEDGMENTS

APPENDIX A

Here we show that the unitaries implementing optimal cooling can be chosen to be permutations of energy eigenstates.

We are given two sequences $\Lambda = (\lambda_1, \ldots, \lambda_N)$ and $D = (d_1, \ldots, d_N)$ of real numbers. Let $\{\lambda_k^{\downarrow}\}_{k=1}^{N}$ and $\{d_k^{\downarrow}\}_{k=1}^{N}$ be the nonincreasing arrangements of their elements:

$$\lambda_1^{\downarrow} \geqslant \lambda_2^{\downarrow} \cdots \geqslant \lambda_n^{\downarrow}, \quad d_1^{\downarrow} \geqslant d_2^{\downarrow} \cdots \geqslant d_n^{\downarrow}. \quad \text{(A1)}$$

Λ *majorizes* D if the following N conditions hold [41]:

$$\sum_{k=1}^{m} \lambda_k^{\downarrow} \geqslant \sum_{k=1}^{m} d_k^{\downarrow} \quad \text{for} \quad m = 1, \ldots, N-1, \quad \text{(A2)}$$

$$\sum_{k=1}^{m} \lambda_k^{\downarrow} = \sum_{k=1}^{m} d_k^{\downarrow}. \quad \text{(A3)}$$

In words, for each $m = 1, \ldots, N$, the sum of the m largest elements of Λ is at least as large as the sum of the k largest elements of D, with equality for $m = N$.

Birkhoff showed that Λ majorizes D if and only if there is a double-stochastic matrix S such that

$$D = S\Lambda, \tag{A4}$$

where double-stochastic means that $S_{ij} \geqslant 0$, $\sum_{j=1}^{N} S_{ij} = \sum_{i=1}^{N} S_{ij} = 1$ [41].

Let Λ be the sequence (15) of initial eigenvalues. Denote by $\{|i\rangle\}_{i=0}^{2M-1}$ the set of eigenvectors for the initial Hamiltonian (6). Let $D = \{\langle i|\rho_{\mathbf{S+R}}(\tau)|i\rangle\}_{i=0}^{2M-1}$ be the probability vector of energy level occupations in the final state (8). Equation (8) implies (A4) with a double-stochastic matrix $S_{ij} = |\langle i|U|j\rangle|^2$, where the unitary operator U is defined by (7) and (8).

The Birkhof theorem implies that the sum of the largest k elements of D is not larger than that of Λ. For the purpose of cooling we want to make the sum of M largest elements of D as big as possible, and thus it has to be equal to the sum of M largest elements of Λ. This is realized if S *permutes* the M largest elements of Λ.

APPENDIX B

The example (42) was developed for $M = 2n = 4$. It can be generalized to arbitrary n:

$$\hat{p}_0 = \pi_0 + \pi_1 + \cdots + \pi_{n-1}, \quad k = 0, \ldots, n-1, \tag{B1}$$

$$\hat{\pi}_{2k} = p_0(\pi_k + \pi_{n+k}), \quad \hat{\pi}_{2k+1} = p_1(\pi_k + \pi_{n+k}), \tag{B2}$$

$$W = \epsilon(\hat{p}_1 - p_1) + \sum_{k=1}^{2n-1} \mu_k(\hat{\pi}_k - \pi_k). \tag{B3}$$

We assume

$$\beta\mu_k \equiv \beta\mu \gg 1 \quad \text{for} \quad k \geqslant n. \tag{B4}$$

Hence,

$$\pi_{k \geqslant n} \to 0, \quad \hat{p}_0 \to 1, \tag{B5}$$

$$W = -\epsilon p_1 + \sum_{k=1}^{n-1} \mu_k(\hat{\pi}_k - \pi_k) + \sum_{k=n}^{2n-1} \mu_k \hat{\pi}_k \tag{B6}$$

$$= -\epsilon p_1 + \sum_{k=1}^{n-1} \mu_k(\hat{\pi}_k - \pi_k) + \mu_n \sum_{k=n}^{2n-1} \hat{\pi}_k. \tag{B7}$$

Consider in detail the case $n = 4$, that is, eight-level reservoir:

$$\beta W = p_1 \ln \frac{p_1}{1 - p_1} + \beta\mu_1(p_1\pi_0 - \pi_1)$$
$$+ \beta\mu_2(p_0\pi_1 - \pi_2) + \beta\mu_3(p_1\pi_1 - \pi_3) + \beta\mu_4(\pi_2 + \pi_3)$$
$$= \left[1 + e^{-\beta\mu_1} + e^{-\beta\mu_2} + e^{-\beta\mu_3}\right]^{-1} \left[\beta\mu_1(p_1 - e^{-\beta\mu_1})\right.$$
$$+ \beta\mu_2(p_0 e^{-\beta\mu_1} - e^{-\beta\mu_2}) + \beta\mu_3(p_1 e^{-\beta\mu_1} - e^{-\beta\mu_3})$$
$$+ \left.\beta\mu_4(e^{-\beta\mu_2} + e^{-\beta\mu_3})\right], \tag{B8}$$

where we used (12) and (14). We now minimize this expression over $\mu_1 \leqslant \mu_2 \leqslant \mu_3$ assuming that $\beta\mu_4 \gg 1$ is fixed [see (B4)]. First we introduce new variables y_2 and y_3,

$$\beta\mu_2 = \ln\left[\frac{\beta\mu_4}{y_2}\right], \quad \beta\mu_3 = \ln\left[\frac{\beta\mu_4}{y_3}\right], \tag{B9}$$

substitue them into (B8) and obtain after minimization:

$$y_2 = p_0 e^{-\beta\mu_1}, \quad y_3 = p_1 e^{-\beta\mu_1}. \tag{B10}$$

Note that $y_2 > y_3$ seen from (B10) due to $p_0 > p_1$ is consistent with $\mu_2 < \mu_3$. Next, we put (B10) back into (B8):

$$\beta W = p_1 \ln \frac{p_1}{1 - p_1}$$
$$+ \frac{\beta\mu_1 p_1 + e^{-\beta\mu_1} \ln[\beta\mu_4 e^{1 + h[p_0]}]}{1 + e^{-\beta\mu_1}}, \tag{B11}$$

where $h[p] \equiv -p \ln p - (1 - p)\ln(1 - p)$. Now the right-hand side of (B11) is to be minimized over μ_1. This is done similarly to (51) and (52). Provided that $\ln[\beta\mu_4 e^{1 + h[p_0]}]$ is sufficiently large, the maximization over $\beta\mu_1$ produced

$$\beta\mu_1 = \ln\left[\frac{1}{p_1} \ln[\beta\mu_4 e^{1 + h[p_0]}]\right], \tag{B12}$$

$$\beta W = p_1 \ln \frac{p_1}{1 - p_1} + p_1 \ln\left[\frac{e}{p_1} \ln[\beta\mu_4 e^{1 + h[p_0]}]\right]. \tag{B13}$$

Hence, the work needed for cooling scales double-logarithmically with maximal energy gap μ_4. The deviation of the ground-state probability (B1) from its maximal value (32) is controlled by (55) [see (B9), (B10), and (B12) in this context].

Equations (B9) and (B12) show that the reservoir spectrum is self-similar: $\beta\mu_3$ and $\beta\mu_2$ depend logarithmically on $\beta\mu_4$, while μ_1 depend on $\beta\mu_4$ doubly logarithmically.

Continuing this reasoning one can show that for a 2^{m-1} level reservoir the work can scale $\ln[\cdots[\ln[\beta\mu]]\cdots]$ (logarithm is repeated m times) as a function of the gap.

[1] H. B. Callen, *Thermodynamics* (Wiley, New York, 1985).

[2] J. C. Wheeler, Phys. Rev. A **43**, 5289 (1991); **45**, 2637 (1992).

[3] P. Landsberg, Am. J. Phys. **65**, 269 (1997).

[4] R. B. Griffiths, J. Math. Phys. **6**, 1447 (1965); in *A Critical Review of Thermodynamics*, edited by E. B. Stuart, B. Gal-Or, and A. J. Mainard (Mono, Baltimore, 1970).

[5] M. J. Klein, in *Thermodynamics of Irreversible Processes, Scuola Internazionale di Fisica "Enrico Fermi,"* edited by S. R. de Groot (Societa Italiana di fisica, Bologna, 1960).

[6] H. B. Callen, in *Modern Developments in Thermodynamics*, edited by B. Gal-Or (Wiley & Sons, New York, 1974).

[7] G. Falk, Phys. Rev. **115**, 249 (1959).

[8] K. A. Gschneidner Jr., V. K. Pecharsky, and A. O. Tsokol, Rep. Prog. Phys. **68**, 1479 (2005).

[9] A. Abragam and M. Goldman, Rep. Prog. Phys. **41**, 395 (1978).

[10] R. R. Ernst, G. Bodenhausen, and A. Wokaun, *Principles of Nuclear Magnetic Resonance in One and Two Dimensions* (Oxford University Press, London, 1987).

[11] O. W. Sorensen, Prog. Nucl. Magn. Reson. Spectrosc. **21**, 503 (1989).

[12] C. P. Slichter, *Principles of Magnetic Resonance* (Springer, Berlin, 1990).

[13] D. Suter, J. Chem. Phys. **128**, 052206 (2008).

[14] D. A. Hall *et al.*, Science **276**, 930 (1997); J. H. Ardenkjær-Larsen *et al.*, Proc. Natl. Acad. Sci. USA **100**, 10158 (2003).

[15] P. O. Boykin *et al.*, Proc. Natl. Acad. Sci. USA **99**, 3388 (2002); J. M. Fernandez *et al.*, Int. J. Quantum Inf. **2**, 461 (2004); F. Rempp, M. Michel, and G. Mahler, Phys. Rev. A **76**, 032325 (2007).

[16] L. J. Schulman, T. Mor, and Y. Weinstein, Phys. Rev. Lett. **94**, 120501 (2005).

[17] A. E. Allahverdyan, R. S. Gracia, and T. M. Nieuwenhuizen, Phys. Rev. Lett. **93**, 260404 (2004).

[18] J. Eschner *et al.*, J. Opt. Soc. Am. B **20**, 1003 (2003).

[19] W. Ketterle and D. E. Pritchard, Phys. Rev. A **46**, 4051 (1992); A. Bartana, R. Kosloff, and D. J. Tannor, J. Chem. Phys. **106**, 1435 (1997); **99**, 196 (1993).

[20] P. W. Bridgman, *The Nature of Thermodynamics* (Harvard University Press, Cambridge, 1941).

[21] D. Janzing, P. Wocjan, R. Zeier, R. Geiss, and Th. Beth, Int. J. Theor. Phys. **39**, 2217 (2000).

[22] Z. Yan and J. Chen, J. Phys. D **23**, 136 (1990); S. Velasco, J. M. M. Roco, A. Medina, and A. C. Hernandez, Phys. Rev. Lett. **78**, 3241 (1997).

[23] R. Kosloff, E. Geva, and J. M. Gordon, J. Appl. Phys. **87**, 8093 (2000).

[24] Y. Rezek *et al.*, Europhys. Lett. **85**, 30008 (2009).

[25] D. Segal, Phys. Rev. Lett. **101**, 260601 (2008).

[26] A. E. Allahverdyan, K. Hovhannisyan, and G. Mahler, Phys. Rev. E **81**, 051129 (2010).

[27] V. B. Bezerra, G. L. Klimchitskaya, V. M. Mostepanenko, and C. Romero, Phys. Rev. A **69**, 022119 (2004).

[28] M. Bordag and I. G. Pirozhenko, Phys. Rev. D **82**, 125016 (2010).

[29] R. M. Wald, Phys. Rev. D **56**, 6467 (1997); G. Chirco, S. Liberati, and T. P. Sotiriou, *ibid.* **82**, 104015 (2010).

[30] Y. Chow and F. Y. Wu, Phys. Rev. B **36**, 285 (1987).

[31] U. Behn and V. A. Zagrebnov, J. Phys. A **21**, 2151 (1988).

[32] G. Watson, G. Canright, and F. L. Somer, Phys. Rev. E **56**, 6459 (1997).

[33] S. A. Ellingsen, Phys. Rev. E **78**, 021120 (2008).

[34] H. S. Leff, Phys. Rev. A **2**, 2368 (1970).

[35] J. Wu and A. Widom, Phys. Rev. E **57**, 5178 (1998).

[36] T. Jahnke, S. Lanery, and G. Mahler, Phys. Rev. E **83**, 011109 (2011).

[37] B. Derrida, Phys. Rev. B **24**, 2613 (1981).

[38] C. Nisoli, R. Wang, J. Li, W. F. McConville, P. E. Lammert, P. Schiffer, and V. H. Crespi, Phys. Rev. Lett. **98**, 217203 (2007); G. C. Lau *et al.*, Nat. Phys. **2**, 249 (2006).

[39] M. Huang and J. P. Sethna, Phys. Rev. B **43**, 3245 (1991); J. J. Brey and A. Prados, *ibid.* **43**, 8350 (1991); D. A. Parshin and A. Wurger, *ibid.* **46**, 762 (1992).

[40] S. M. Stishov, Sov. Phys. Usp. **31**, 52 (1988).

[41] A. W. Marshall and I. Olkin, *Inequalities: Theory of Majorization and its Applications* (Academic Press, New York, 1979).

[42] G. Lindblad, *Non-Equilibrium Entropy and Irreversibility* (D. Reidel, Dordrecht, 1983).

[43] A. Lenard, J. Stat. Phys. **19**, 575 (1978); I. M. Bassett, Phys. Rev. A **18**, 2356 (1978); W. Thirring, *Quantum Mechanics of Large Systems*, Vol. 4 of *A Course in Mathematical Physics* (Springer-Verlag, Wien, 1980).

[44] J. M. R. Parrondo, C. Van den Broeck, and R. Kawai, New J. Phys. **11**, 073008 (2009).

[45] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, UK, 2000); V. Vedral, Rev. Mod. Phys. **74**, 197 (2002).

[46] L. D. Landau and E. M. Lifshitz, *Statistical Physics I* (Pergamon Press, Oxford, 1978).

[47] A. E. Allahverdyan, R. Balian, and Th. M. Nieuwenhuizen, Europhys. Lett. **67**, 565 (2004).

[48] A. F. Nikiforov and V. B. Uvarov, *Special Functions of Mathematical Physics* (Birkhaeuser, Basel, Switzerland, 1988).

[49] J. Gemmer, M. Michel, and G. Mahler, *Quantum Thermodynamics* (Springer, New York, 2004).

[50] H. Touchette, R. S. Ellis, and B. Turkington, Physica A **335**, 518 (2004).

[51] F. Borgonovi and F. M. Izrailev, Phys. Rev. E **62**, 6475 (2000).

[52] M. Kastner and O. Schnetz, J. Stat. Phys. **122**, 1195 (2006).

[53] A. Ramirez-Hernandez, H. Larralde, and F. Leyvraz, Phys. Rev. E **78**, 061133 (2008).

[54] A. Campa, T. Dauxois, and S. Ruffo, Phys. Rep. **480**, 57 (2009).

[55] A. E. Allahverdyan and Th. M. Nieuwenhuizen, Physica A **305**, 542 (2002).

[56] A. E. Allahverdyan and K. V. Hovhannisyan, Europhys. Lett. **95**, 60004 (2011).

[57] Th. M. Nieuwenhuizen and A. E. Allahverdyan, Phys. Rev. E **66**, 036102 (2002).