



## Quantum Bath Refrigeration towards Absolute Zero: Challenging the Unattainability Principle

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(Received 21 May 2012; published 27 August 2012)

A minimal model of a quantum refrigerator, i.e., a periodically phase-flipped two-level system permanently coupled to a finite-capacity bath (cold bath) and an infinite heat dump (hot bath), is introduced and used to investigate the cooling of the cold bath towards absolute zero ( $T = 0$ ). Remarkably, the temperature scaling of the cold-bath cooling rate reveals that it does not vanish as  $T \rightarrow 0$  for certain realistic quantized baths, e.g., phonons in strongly disordered media (fractons) or quantized spin waves in ferromagnets (magnons). This result challenges Nernst's third-law formulation known as the unattainability principle.

DOI: [10.1103/PhysRevLett.109.090601](https://doi.org/10.1103/PhysRevLett.109.090601)

PACS numbers: 05.70.Ln, 03.65.Yz, 07.20.Pe

*Introduction.*—One of the generally unsettled fundamental problems of thermodynamics is the nature of the ultimate limitations on cooling to absolute zero,  $T = 0$ . Attaining  $T = 0$  in a finite number of steps, or, more generally, in finite time, is prohibited by Nernst's unattainability principle which is the dynamical formulation of the third law of thermodynamics [1–3]. However, the universality of this principle has been postulated rather than proven. It is also debatable whether this formulation is always equivalent to Nernst's heat theorem, whereby the entropy vanishes at  $T = 0$ . Do both formulations of the third law hold for all quantum scenarios? The investigation of this open fundamental problem, pertaining to quantum refrigerator (QR) schemes [4–8], raises several principal questions: (i) How does the cooling rate scale with the bath temperature and does it necessarily vanish as  $T \rightarrow 0$ ? (ii) Does a QR differ from its classical counterpart regarding compliance with the third law and to what extent is such compliance model dependent? Answers to these questions are important not only to the understanding of the foundations of quantum thermodynamics [9], but also to the design of novel QR schemes compatible with the needs of quantum nanotechnologies that require compact (nano-size) coolers capable of ultrafast cooling [10].

Here, we propose and explore the simplest QR design thus far that allows us to address the fundamental issues raised above. The working medium is a single two-level system (qubit), permanently (rather than intermittently, as is done in traditional cycles [4]) coupled to a finite-capacity bath to be cooled and to another, much larger and hotter, heat dump. The pumping operation consists of fast modulation of the qubit energy by means of periodic  $\pi$ -flips of the qubit phase. We find that this QR can cool down a finite-capacity (yet macroscopic and spectrally-continuous) bath only if the modulation-period is within the bath-memory (non-Markovian) time. Hence, the cold-bath spectrum is crucial in determining the cooling

condition and rate. Our most striking finding is that for certain experimentally realizable baths, such as quantized spin-waves in ferromagnets (magnons) [11] or acoustic phonons in strongly disordered media (fractons) [12], the cooling rate remains finite as  $T \rightarrow 0$ , in apparent violation of the dynamical formulation of the third law.

*Model and analysis.*—A control qubit is weakly coupled to two baths via the system-bath interaction Hamiltonian:  $H_{\text{SB}} = \sigma_x(B_H + B_C)$ , where  $\sigma_x$  is the spinor  $x$  component,  $B_C$  is the operator of a finite cold bath ( $C$ ) which we wish to refrigerate, and  $B_H$  that of a much larger hot bath ( $H$ ) that remains nearly unchanged. The qubit energy is periodically modulated by an external field  $\nu(t)$  via the Hamiltonian  $H_{\text{ext}} = \frac{1}{2}\sigma_z\nu(t)$ . An illustration (Fig. 1-inset) is that of a charged quantum particle in a double-well potential that is periodically phase-flipped by off-resonant pulses and is coupled to a spatially-confined (macroscopic)  $C$ -bath to be cooled, as well as to a nearly-infinite  $H$  environment into which the heat is dumped. This scheme bears analogy to radiative (sideband) cooling in solids and molecules [13], if one visualizes the red- and blue-shifted qubit frequencies as Stokes and anti-Stokes lines, respectively.

Our analysis reveals the crucial role of the quantized characteristics of system-bath coupling in determining the attainability of  $T \rightarrow 0$ . By contrast, the results are insensitive to the QR scheme chosen (see Discussion).

The general condition for steady-state refrigeration, under periodic, off-resonant, modulation, is positive heat current from  $C$  to  $H$  via the qubit. The sign and magnitude of the current is determined by the steady-state solution of a non-Markovian master equation (ME) for the qubit density matrix [14]. The ME, which is accurate to second order in the system-bath coupling, allows for time-dependent modulation of the system that is much faster than the bath-memory time  $t_c$ . It is valid at any  $T$ , as shown both theoretically [14] and experimentally [15,16]. Deviations

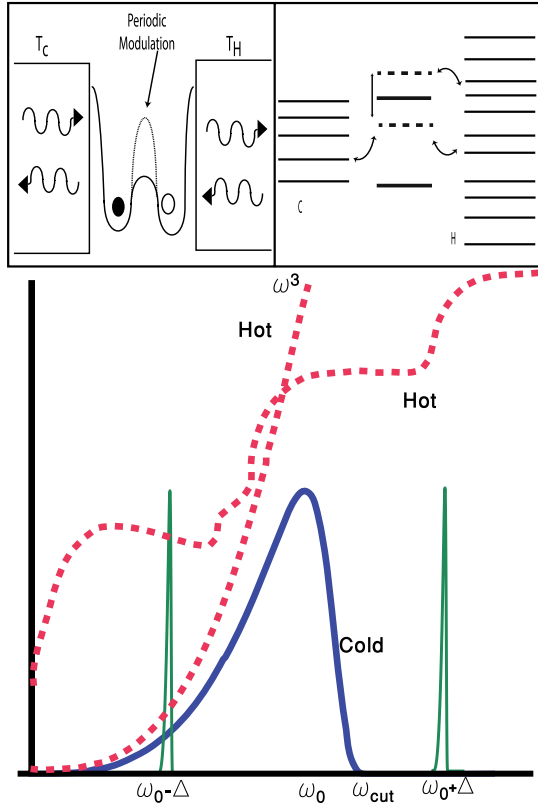


FIG. 1 (color online). Main panel: Schematic depiction of the required bath spectra and the qubit frequency shifts due to periodic phase flips. Inset: Schematic realization of the modulated qubit and its coupling to the baths.

of the evolution (Supplemental Material A, [17]) from that described by the non-Markovian ME include system-bath entanglement (correlations) effects that can be compensated by readjusting the qubit excitation, as well as bath dynamics effects (violation of the Born approximation whereby the bath is constantly in a thermal state) [18]. Yet such deviations are of fourth-order in the system-bath coupling and thus negligible for weak coupling. The cooling of a finite-capacity bath is the result of infinitesimal temperature changes over many modulation cycles, consistently with the Born approximation underlying the ME. Since the Born approximation is the more accurate the larger the bath [19], we assume that the finite-capacity bath is macroscopic and has a continuous spectrum, which does not exhibit mode discreteness or recurrences that may otherwise invalidate this approximation and bath thermalization altogether [19].

*Only the diagonal elements.*—of the qubit's density matrix  $\rho_S$  (energy-state populations) play a part here, although the ME also allows for coherences (off-diagonal elements) [14], but these are absent at  $t = 0$  (starting at equilibrium) and remain so under the modulation. The quantumness of the ME, even when it is diagonal in the energy basis, is embodied by the qubit interlevel transition rate and their non-Markovian time-dependence (Supplemental Material B, [17]). Periodic phase shifts of

the qubit at intervals  $\tau$  dynamically control its coupling to the baths and the resulting transition rates. When  $\tau$  is comparable to the bath memory-time  $t_c$ , these phase shifts modify the detailed balance of the transition rates and thereby either heat or cool the qubit depending on  $\tau$  [16,18]. In what follows, we analyze the steady state and the slow changes of the bath temperature as a result of these periodic perturbations.

Under weak-coupling conditions, the qubit evolution caused by the baths is much slower than  $\tau \sim t_c$ . Hence, in steady state, we can use time-averaged level populations and transition rates between the periodically-perturbed qubit levels (Supplemental Material B, [17]). These time-averaged (steady-state) equations can be recast, upon introducing the polarization of the qubit  $S \equiv (\rho_{ee} - \rho_{gg})/2$ , into

$$\dot{S} = -[\bar{R}_g + \bar{R}_e]S + \frac{\bar{R}_g - \bar{R}_e}{2}, \quad (1)$$

Here, the  $|e\rangle \rightarrow |g\rangle$  averaged transition rate from the excited ( $e$ ) to the ground ( $g$ ) state is  $\bar{R}_e$  and its  $|g\rangle \rightarrow |e\rangle$  counterpart is  $\bar{R}_g$ . The averaged transition rates for  $t \gg \tau$  are found, upon expanding the qubit energy under periodic frequency modulation  $\nu(t)$  into the harmonic (Floquet) series (Supplemental Material B, [17], [20])

$$\bar{R}_e(g) \equiv 2\pi \sum_m P_m G_T[\pm(\omega_0 + m\Delta)]; \quad P_m = |\varepsilon_m|^2, \\ \varepsilon_m = \frac{1}{\tau} \int_0^\tau e^{i \int_0^t (\nu(t') - \omega_0) dt'} e^{im\Delta t} dt, \quad (2)$$

Here  $m$  are all (positive and negative) integers,  $P_m$  are the probabilities of shifting  $G_T(\omega)$  by  $m\Delta$ ,  $\Delta = \frac{2\pi}{\tau}$ , from the qubit average frequency  $\omega_0$ ,  $G_T(\omega)$  being the temperature-dependent bath-coupling spectrum, i.e., the Fourier transform of the bath autocorrelation function:  $G_T(\omega) = \int_{-\infty}^{+\infty} e^{i\omega t} \langle B(t)B(0) \rangle dt = e^{\omega/k_B T} G_T(-\omega)$ . For a bosonic bath:

$$G_T(\omega) = G_0(\omega)(n(\omega) + 1); \\ G_0(\omega) = |g(\omega)|^2 \rho(\omega); \\ n(\omega) = \frac{1}{e^{\omega/T} - 1}, \quad (3)$$

$g(\omega)$  being the system-bath coupling,  $\rho(\omega)$  the bath -mode density and  $n(\omega)$  the  $\omega$ -mode thermal occupancy. These expressions are also obtainable by Floquet (harmonic) expansion of the periodically-driven Markovian (Lindblad) Liouvillian [20,21].

In the presence of hot ( $H$ ) and cold ( $C$ ) baths, under the assumption of a qubit weakly coupled to both baths, the transition rates are split into additive harmonic contributions:  $\bar{R}_{e(g)} \equiv \sum_m (R_{e(g)}^{C(m)} + R_{e(g)}^{H(m)})$ . Hence, Eq. (1) is also split into (Supplemental Material B, [17])  $\dot{S} = \sum_m (\bar{S}_m^C + \bar{S}_m^H)$ , where  $\bar{S}_m^{C(H)}$  is the  $m$ -harmonic polarization flow caused by the cold (hot) bath only. The averaged heat

flow,  $\dot{Q}$ , through the qubit is correspondingly divided into the  $C$  and  $H$  bath contributions. The steady-state Eq. (1) then gives rise to

$$J_{C(H)} = \dot{Q}_{C(H)} = \sum_m (\omega_0 + m\Delta) \overline{S_m^{C(H)}} \quad (4)$$

which is the sum of rates of heat-exchange with the respective baths via  $\omega_0 + m\Delta$  quanta. Positive  $J_C$  implies refrigeration, i.e., heat flow from the cold bath to the hot bath via the modulated qubit.

It is advantageous to use periodic, alternating,  $\pi$ -phase shifts (phase flips) as they give rise, to leading order, to two symmetrically opposite frequency shifts of  $G_T$  at  $\omega_0 \pm \Delta$ . Then, by the Floquet expansion we obtain the probability distribution wherein  $P_0 = 0$  and  $P_{\pm 1} \approx (2/\pi)^2$  are the leading terms [14,22].

Let us choose sufficiently large  $\Delta$ , of the order of the spectral width  $\Gamma = 1/t_c$ , which is the inverse memory time of the cold bath, such that at  $\omega \approx \omega_0 + \Delta$  the qubit is coupled only to the hot bath, while at  $\omega \approx \omega_0 - \Delta$  it is coupled to both the cold and the hot baths. More precisely, we require that

$$\begin{aligned} G_T(\omega_0 + \Delta) &\approx G_T^H(\omega_0 + \Delta) \gg G_T^C(\omega_0 + \Delta); \\ G_T^H(\omega_0 + \Delta) &\gg G_T^H(\omega_0 - \Delta), \quad G_T^C(\omega_0 - \Delta) \end{aligned} \quad (5)$$

where  $G_T^{C(H)}(\omega)$  is the respective temperature-dependent bath-coupling spectrum. This requirement can be satisfied if the cold bath ( $C$ ) is spectrally localized with upper cutoff  $\omega_{\text{cut}} < \omega_0 + \Delta$ . By contrast, for the hot bath ( $H$ ), the required rise of  $G_T^H$  with  $\omega$  is obtained for most common bath spectra, provided the cutoff of  $G_T^H(\omega)$  is much higher than  $\omega_{\text{cut}}$  of  $G_T^C(\omega)$ : e.g., for phonons in bulk media or photons in open space,  $G_T^H(\omega) \propto \omega^3$  satisfies Eq. (5).

Under the conditions of Eq. (5) we find that the steady-state heat current from  $C$  to  $H$  is (Supplemental Material B, [17])

$$\begin{aligned} J_C &= (\omega_0 - \Delta) \overline{S_S^C} \\ &\approx (\omega_0 - \Delta) \frac{G_0^C(\omega_0 - \Delta) [n^C(\omega_0 - \Delta) - n^H(\omega_0 + \Delta)]}{[2n^H(\omega_0 + \Delta) + 1]}. \end{aligned} \quad (6)$$

The balance of  $J_C$  and  $J_H$  (cold and hot) currents (4) obeys the second law [2,23]: It can be verified that the entropy production rate  $\frac{dS}{dt}$  satisfies:  $\frac{dS}{dt} - \left(\frac{J_C}{T_C} + \frac{J_H}{T_H}\right) \geq 0$  for any initial state.

From Eq. (6), the heat pump (QR) condition  $J_C > 0$  amounts to

$$n^C(\omega_0 - \Delta) > n^H(\omega_0 + \Delta) \Leftrightarrow \frac{\omega_0 + \Delta}{T_H} > \frac{\omega_0 - \Delta}{T_C}. \quad (7)$$

An analogous relation holds if  $n^{C(H)}(\omega)$  are Boltzmann rather than Bose factors (occupancies).

Equation (7) reveals the crux of the heat pumping (QR) effect: although by definition  $n^C(\omega_0) < n^H(\omega_0)$ , heat can flow from the cold to the hot bath if the  $C$ -bath thermal occupancy at  $\omega_0 - \Delta$  is higher than that of the  $H$  bath at  $\omega \approx \omega_0 + \Delta$ . If  $\Delta$  is too small for Eq. (7) to hold, we recover the natural heat-flow direction  $H \rightarrow S \rightarrow C$  at steady-state. In addition, Eq. (5) implies that the heat pump requires the qubit to be simultaneously coupled to the  $C$  and  $H$  baths at  $\omega_0 - \Delta$  and  $\omega_0 + \Delta$ , respectively.

*Cooling rate scaling with temperature.*—In what follows, we investigate the QR action (heat pumping from  $C$  to  $H$ ) under the assumptions that the hot bath is practically infinite; hence,  $T_H = \text{const}$ , whereas the macroscopic cold bath has finite heat capacity,  $c_V < \infty$ , resulting in slow evolution of  $T_C(t)$  under the QR action. To estimate this evolution we use the standard thermodynamic definition [24]

$$c_V \frac{dT_C(t)}{dt} = J_C = \dot{Q}_C \quad (8)$$

which presumes that  $T_C$  is well-defined at all  $t$  (since the bath has a continuous spectrum and is large enough to thermalize at finite times).

In order to infer the temperature dependence of the cooling rate  $\frac{dT_C}{dt}$  we shall examine the scaling of  $c_V$  and  $J_C$  with  $T_C$ : (a) The constant-volume heat capacity of the cold-bath,  $c_V$ , depends on the dimensionality of the bosonic bath. If  $\rho(\omega) \approx \omega^{d-1}$  is the  $d$ -dimensional density of modes and  $T_C \ll \omega_{\text{cut}}$  ( $k_B = \hbar = 1$ ), then

$$\begin{aligned} \lim_{T_C \rightarrow 0} c_V &= \frac{d}{dT} \left. \frac{\langle H_B \rangle}{V} \right|_{T_C} \\ &\approx \frac{d}{dT} \int d\omega \omega \rho(\omega) (n_C(\omega) + 1) \Big|_{T_C} \sim T_C^d. \end{aligned} \quad (9)$$

(b) The scaling of the cold-bath heat current,  $J_C$ , in Eq. (7) can be deduced if we maximize the heat flow [25] with respect to  $\Delta$  (our control parameter). This gives the dependence of  $\omega_0 - \Delta \approx T_C$  [7,26]. Hence, to maintain the maximum heat flow, we have to slowly increase  $\Delta$  with time, so as to approach  $T_C \rightarrow 0$ . The closer to  $T_C \rightarrow 0$ , the lower is  $\omega_0 - \Delta$ ; hence, the steady-state dynamics (7) and its slow change (8) become increasingly more accurate. Correspondingly, we parametrize  $J_C$  in Eq. (6) using Eq. (3) and assuming the low-frequency range of the cold bath  $0 \leq \omega = \omega_0 - \Delta \ll \omega_{\text{cut}}$ :  $\lim_{\omega \rightarrow 0} |g(\omega)|^2 \propto \omega^\gamma$ ,  $\rho(\omega) \approx \omega^{d-1}$ . Here,  $|g(\omega)|^2$  is the  $\gamma$ -dependent system coupling to the bosonic bath (discussed below). The heat current, maximized for  $\omega_0 - \Delta \approx T_C$ , then obeys the scaling

$$J_C(T_C) \propto -T_C^{\gamma+d}. \quad (10)$$

(c) Upon substituting Eqs. (9) and (10) in Eq. (8) we observe that the  $T_C^d$  scaling of  $c_V$  is canceled by a similar scaling of the density of modes in Eq. (10). The resulting scaling yields

$$dT_C/dt = -AT_C^\gamma. \quad (11)$$

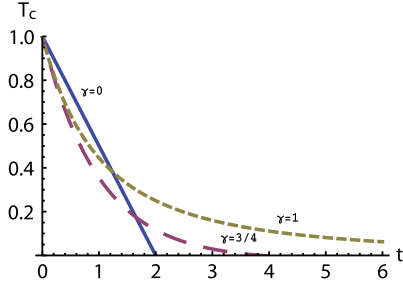


FIG. 2 (color online).  $T_C$  change with time (cooling) for three different system-bath coupling-strength dispersion laws:  $\gamma = 1$  (acoustic phonons),  $\gamma = 3/4$  (fractons),  $\gamma = 0$  (magnons).

Here the constant  $A \propto 1/V$ : the larger the bath the slower its cooling.

Remarkably, the  $\frac{dT_C}{dt}$  scaling only depends on the  $\gamma$ th scaling power of the system-bath coupling strength  $|g(\omega)|^2$ . For different  $\gamma$  the time dependence of  $T_C$ , starting from the same  $T_C(0)$ , is plotted in Fig. 2. For  $\gamma = 1$  we have exponentially slow convergence to  $T_C \rightarrow 0$ , conforming to the third law. Yet, strikingly, for  $0 \leq \gamma < 1$ ,  $T_C(t) \rightarrow 0$  at finite time, thus violating the accepted dynamical formulation of the third law [1–3], if the frequency-dependent coupling  $|g(\omega)|^2$  is sublinear in  $\omega$ .

In what follows, we examine the possibility of such scaling for different bosonic baths. To this end, consider a qubit immersed in a periodic medium, whose local displacement is a linear combination of normal-mode creation and annihilation operators (bath excitations or deexcitations)  $\hat{B}(\vec{x}) = \frac{1}{\sqrt{V}} \sum_{\vec{k}} \frac{1}{\sqrt{\omega(\vec{k})}} (\phi_{\vec{k}}(\vec{x}) a^\dagger(\vec{k}) + \text{H.c.})$ . The normal-mode functions are labeled by the wave vectors  $\vec{k}$  that belong to a reciprocal lattice bounded by the Debye cutoff ( $\omega(\vec{k}) \leq \omega_{\text{cut}} = \omega_D$ ). The couplings of a charged or dipolar system to bath excitations or deexcitations are to leading order determined by the gradient of the displacement operator

$$\nabla \hat{B}(\vec{x}) = \frac{-i}{\sqrt{V}} \sum_{\vec{k}} \frac{1}{\sqrt{\omega(\vec{k})}} (\nabla \phi_{\vec{k}}(\vec{x}) a^\dagger(\vec{k}) - \text{H.c.}) \quad (12)$$

When  $\phi_{\vec{k}}(\vec{x}) = e^{-i\vec{k}\cdot\vec{x}}$  the corresponding coupling constant scales as  $|g(\omega(\vec{k}))| \sim \frac{\vec{k}}{\sqrt{\omega(\vec{k})}}$ . We can discern three generic types of scaling of the coupling constant: (i) For acoustic phonons  $\omega(\vec{k}) \simeq v|\vec{k}|$ , where  $v$  is a sound velocity and the coupling strength satisfies  $|g(\omega)|^2 \sim \omega$ , i.e.,  $\gamma = 1$ . Therefore, acoustic phonons used as a cold bath do not violate the dynamical third-law formulation: the optimal cooling to zero temperature is exponential in time.

(ii) Amorphous (glass) materials may exhibit effects of fractal disorder. These effects imply different scaling of the displacement of the mode function  $\phi_{\vec{k}}(\vec{x})$ ,  $|\nabla \phi_{\vec{k}}(\vec{x})| \sim \omega^\gamma |\phi_{\vec{k}}(\vec{x})|$ : normal phonons are replaced by fractons for which  $\gamma$  takes fractional values. In particular, for some

materials  $\gamma < 1$  [12]. Hence, for a cold bath composed of such fractons the violation of the third law is expected.

(iii) Another system which leads to a violation of the third law is the magnon (spin-wave) bath in a ferromagnetic spin lattice with nearest-neighbor interactions, below the critical temperature. The Holstein-Primakoff transformation of the  $j$ th spin Pauli matrix [11],  $S_j^+ = S_{jx} + iS_{jy} = (2S)^{1/2}(1 - a_j^\dagger a_j/2S)^{1/2} a_j$  to boson annihilation and creation operators  $a_j, a_j^\dagger$ , allows us to represent the system as a set of interacting harmonic oscillators. Introducing the collective spin-wave (magnon) variables  $a(\vec{k}), a^\dagger(\vec{k})$  satisfying  $a_j = \frac{1}{\sqrt{N}} \sum_{\vec{k}} e^{-i\vec{k}\cdot\vec{x}_j} a(\vec{k})$ , we can rewrite its Hamiltonian in the form  $H_0 = \sum_{\vec{k}} \omega(\vec{k}) a^\dagger(\vec{k}) a(\vec{k}) + \text{higher-order terms}$ . At low temperatures the nonlinearity in the Holstein-Primakoff transformation can be neglected and the system becomes equivalent to a bosonic system governed by the Hamiltonian  $H_0$ , whereby the dispersion law is quadratic in the low-frequency region,  $\omega(\vec{k}) \sim (|\vec{k}|^2 + \text{constant})$ . The local spin variable  $a_j$  can then be directly coupled to the qubit by a dipole-dipole (spin-spin) interaction. Hence, the main difference between the dipolar coupling to acoustic phonons and magnons is the absence of the dispersive-coupling coefficient  $\frac{\vec{k}}{\sqrt{\omega(\vec{k})}}$  for the latter.

Therefore, the coupling strength to magnons satisfies  $|g(\omega)|^2 \sim 1$  ( $\gamma = 0$ ), which implies the violation of the third law for magnons.

*Discussion.*—We have analyzed the cooling process of a bosonic bath towards the absolute zero using a new minimal model of a quantum refrigerator: a single two-level system (qubit) permanently coupled to a spectrally restricted cold bath with finite heat capacity and a hot bath with infinite heat capacity has been shown to act as a heat pump, under appropriate modulation. The heat flow is proportional to the population-difference of a pair of oppositely shifted bath modes that are selected by the qubit modulation (phase-flip) rate, analogously to sideband cooling [13]. The attainable cooling rate challenges the third law of thermodynamics, in the sense that arbitrarily low temperature of the cold bath may be reached in finite time by the heat pump for certain quantized cold-bath spectra: e.g., magnon and fracton baths.

In solid-state ferromagnets or glasses, interactions of control qubits with other baths unaccounted by the model, as well as tiny deviations from the predicted weak-coupling, steady-state dynamics (discussed in the Supplemental Material A,B [17]) may restore the third law. Nevertheless, surprisingly fast cooling ( $\gamma < 1$ ) may still be observed down to some (material-dependent) temperature. It would be preferable to demonstrate this effect for quantum dots coupled to controllable baths composed of nuclear spins in solids [27] or for atomic dipoles in optical lattices [28]: in both cases the systems are highly shielded from other baths, while the lattices can be

engineered to conform to the nearest-neighbor ferromagnetic model that engenders magnons.

This study of the compliance with the third law for quantized non-Markovian baths indicates that the temperature scaling of the cooling rate is not specific to the chosen QR model; it is similar to the scaling obtained for the very different noise-driven QR [26]. Namely, the scaling is not sensitive to the form of driving, nor to the method of treating the steady-state dynamics. Hence, the dependence of the scaling on the system-bath coupling dispersion is general. It provides new insights into the bounds of bath cooling in quantum thermodynamics. It shows that Nernst's principle of unattainability of the absolute zero in finite time [1–3] may fail and is not always equivalent to Nernst's heat theorem (see Introduction): the latter holds true since a bosonic bath has a unique ground state whose entropy must vanish.

The support of ISF, DIP, the Humboldt-Meitner Award (G. K.), the Weston Visiting Professorship (R. A.), CONACYT (D. G.) and the Czech Science Foundation, GAP205/10/1657 (M. K.) are acknowledged. Useful discussions are acknowledged with Nir Gov and Ronnie Kosloff. M. K. and D. G. contributed equally to this work.

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