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Quantum refrigerators in quest of the absolute zero

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The second and third laws of thermodynamics can be used to establish a fundamental bound for the maximum possible cooling rate in approaching the absolute zero of temperature. In modeling the behavior of the molecular refrigerators geared toward attaining ultralow temperatures, only quantum mechanical, as opposed to classical physics, models can be admissible. As a simple model, we analyze a three-level quantum refrigerator, and in particular its irreversible thermodynamic performance as absolute zero is approached.
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I. INTRODUCTION

Part of the intrigue in cooling close to the absolute zero of temperature is the unraveling of the quantum nature of matter and associated applications. Many methods have been invented which depend on the particulars of the system being cooled. The theoretical analyses employed have been complex and often case specific. Our interest lies in searching for simple, basic underlying principles that allow an analysis of these cooling processes yet obviate the need for specific mechanistic details. One fundamental question is: What are the restrictions imposed on the cooling rate as absolute zero is approached. Combining the fields of thermodynamics and quantum mechanics is the key to such an endeavor.

In the quest for temperatures approaching absolute zero, the second and third laws of thermodynamics provide fundamental bounds on attainable cooling rates. Two schemes often considered are: (a) evaporation and (b) refrigeration cycles. The analysis of evaporative cooling near absolute zero is reserved for a separate treatment. This article is limited to the study of the cooling characteristics of quantum refrigerators.

As an illustration, consider a cyclic endoreversible refrigerator (illustrated schematically in Fig. 1) that is driven by a power input P, removes heat at a rate $\dot{Q}_c$ from a cold reservoir of temperature T_c, and rejects heat at a rate $\dot{Q}_h$ to a hot reservoir of temperature T_h. The rates P, $\dot{Q}_c$, and $\dot{Q}_h$ represent cycle-average quantities and are defined as positive. (Even if irreversibilities beyond finite-rate heat exchange are introduced, the arguments expounded immediately below remain valid.) Our objective is to cool the cold reservoir arbitrarily close to the absolute zero.

One equivalent statement of the third law is that no system can be cooled to the absolute zero in finite time. The second law requires that the entropy production in the universe due to refrigerator operation must be nonnegative. Say we can express $\dot{Q}_c$ as a function of T_c. Then in the limit of vanishing T_c, the third law requires that $\dot{Q}_c$ must, at the very least, be proportional to a positive power of T_c, i.e.,

$$\dot{Q}_c \propto T_c^{\alpha}, \tag{1}$$

with $\alpha > 0$. The rate of entropy production in the universe $\dot{S}$ is

$$\dot{S} = (\dot{Q}_h / T_h) - (\dot{Q}_c / T_c). \tag{2}$$

The second law insures that the negative entropy production at the cold bath is more than offset by the positive entropy production at the hot bath. Hence, if the entropy production due to finite-rate heat exchange with the hot bath is bounded, the second law implies an even stronger condition: $\dot{Q}_c$ must increase at least linearly with T_c (i.e., $\alpha \geq 1$), independent of the model invoked.

The problem in examining cooling to ultralow temperatures with refrigeration models based on classical physics is that in the limit of absolute zero, device behavior is dominated by quantum-mechanical effects. This provides the incentive to develop a simple quantum refrigerator model with which these basic bounds can be explored rigorously.

II. A REVERSIBLE THREE-LEVEL QUANTUM REFRIGERATION CYCLE

An external driving field (coherent radiation) is the work input to the proposed quantum refrigeration cycle depicted schematically in Fig. 2. Heat is pumped from the cold to the hot bath $(2 \rightarrow 3)$. Heat rejection $(3 \rightarrow 1)$ results from the coupling of levels 3 and 1 to the hot bath, while cooling (heat removal, $1 \rightarrow 2$) derives from the coupling of levels 1 and 2 to the cold bath. The hot and cold baths are macroscopic objects which maintain a constant temperature during the

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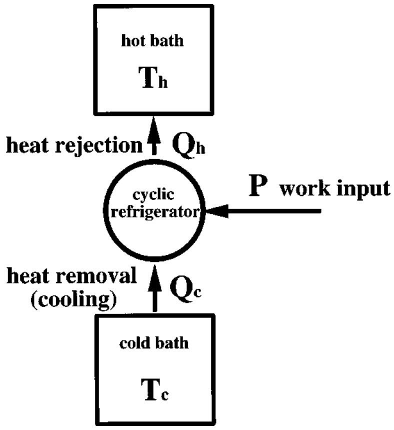

FIG. 1. Schematic of an endoreversible refrigeration cycle.

refrigeration cycle. Steady-state (rather than transient) cyclic operation will be analyzed. Accordingly, the change in the state variables of the refrigerator itself is zero over the cycle, independent of operating details.

The second law requires that the population of level 3, N_3, not exceed that of level 2, N_2. A reversible mode of operation is considered first (in the following section, an irreversible dynamical mode will be analyzed). Equilibrium is maintained between: (a) levels 1 and 3 with the hot bath; and (b) levels 1 and 2 with the cold bath. Since $N_3/N_1 = \exp(-\Delta E_{31}/T_h)$ and $N_2/N_1 = \exp(-\Delta E_{21}/T_c)$, the requirement that $N_3 \leqslant N_2$ leads to a lower bound for T_c

$$T_c \geqslant T_h \Delta E_{21}/\Delta E_{31}, \tag{3}$$

where ΔE_{ij} denotes the energetic difference between levels i and j.

Refrigerator efficiency is commonly cited as the coefficient of performance (COP), defined as the cooling energy delivered relative to the work input. In this instance

$$\mathrm{COP} = \Delta E_{21}/\Delta E_{32} \leqslant T_c/(T_h - T_c), \tag{4}$$

where $T_c/(T_h - T_c)$ is the reversible limit or Carnot COP and is the absolute upper bound for *any* refrigeration cycle.

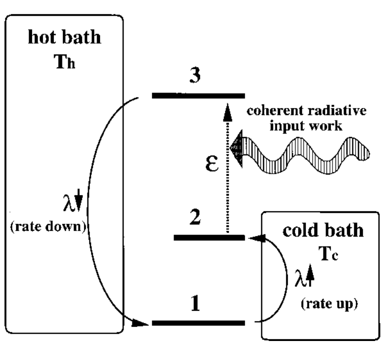

FIG. 2. Schematic of a three-level quantum refrigeration cycle. The driving input power is coherent radiation, which induces the transition from level 2 to 3. Heat rejection from level 3 to 1 is effected by coupling levels 1 and 3 to a hot bath of temperature T_h. Cooling (heat removal) derives from coupling levels 1 and 2 to a cold bath of temperature T_c. λ denotes bath pumping rate. The radiative input is coupled to levels 2 and 3 with coupling parameter ε and frequency $\omega_0 = \Delta E_{32}$. (Units are used in which $k_B = 1$ and $\hbar = 1$.)

In principle, a thermodynamic cycle can be equally well run in reverse. Namely, by reversing the direction of all energy flows, a refrigeration cycle is converted to a heat engine. Although our major thrust is an examination of the unique aspects of the cooling mode, we digress momentarily to note the operation of the three-level scheme portrayed in Fig. 2 in heat engine mode.

Heat from the hot bath creates a population inversion by pumping the $1 \rightarrow 3$ transition. The amplification or work produced is the stimulated emission of the $3 \rightarrow 2$ transition. Coupling to the cold bath induces the $2 \rightarrow 1$ transition, and the cycle is complete.

Population inversion requires $N_3 > N_2$, which in turn implies $T_c/T_h \leqslant \Delta E_{21}/\Delta E_{31}$. Heat engine efficiency η can be viewed as the ratio of the photon work generated ΔE_{32} to the heat input ΔE_{31}. From these observations, we arrive at the familiar Carnot limit for heat engine efficiency: $\eta = \Delta E_{32}/\Delta E_{31} \leqslant 1 - (T_c/T_h)$. This analogy between the quantum amplifier and the Carnot heat engine was recognized in Refs. 1–4.

Returning to consideration of the cooling operation, we note that the reversible three-level quantum refrigerator is an idealized model of the laser cooling of solids[5–8] or of laser cooling of dyes.[9] Cooling is based on an anti-Stokes process where the pump is identified as the $2 \rightarrow 3$ transition and fluorescence as the $3 \rightarrow 1$ transition. For reversible operation, the cooling rate is zero. However, laser cooling proceeds at a finite rate and is therefore inherently irreversible. This requires a dynamical version of the model.

III. DYNAMICAL MODEL OF THE QUANTUM REFRIGERATOR

The dynamical model requires equations of motion for the thermodynamic observables. A reduced description of the dynamics in terms of the operators of the three-level system is sufficient for identifying the quantum analogs of the thermodynamic variables. Energy is associated with the Hamiltonian of the working medium, $\mathbf{H} = \mathbf{H_0} + \mathbf{V}(t)$. It contains the bare level projections, $\mathbf{H_0} = E_1 \mathbf{P_1} + E_2 \mathbf{P_2} + E_3 \mathbf{P_3}$ and a time-dependent interaction $\mathbf{V}(t) = \varepsilon \mathbf{P_{23}} \exp(i\omega_0 t) + \varepsilon^* \mathbf{P_{32}} \exp(-i\omega_0 t)$, where $\mathbf{P_{ij}} = |i\rangle\langle j|$ ($\mathbf{P_i} = \mathbf{P_{ii}}$). This time-dependent interaction is responsible for the energy transfer with an external power source.

The dynamics are influenced by the heat transfer with the baths, represented by the Heisenberg equation of motion

$$\dot{\mathbf{A}} = i\hbar[\mathbf{H},\mathbf{A}] + \mathcal{L}_h(\mathbf{A}) + \mathcal{L}_c(\mathbf{A}) \tag{5}$$

where $\mathcal{L}_h$ and $\mathcal{L}_c$ are the dissipative Liouville operators corresponding to the hot and cold baths, respectively.

The reduced dynamical description of the system is based on a thermodynamic perspective in which the bulk terms are dominant relative to the surface interaction terms with the bath. The interface between the system and the bath becomes a thermodynamically isothermal partition which allows energy transfer, but does not destroy the integrity of the system. That is, no quantum entanglement is created between the system and the baths. This requirement has been shown to be equivalent to the conditions of the quantum weak-coupling limit,[10] for which explicit forms of the dissipative superoperators $\mathcal{L}$ have been derived.[11,12]

The external periodic driving force causes a perturbation in the energy level structure of the system. As a result, levels 2 and 3 are split by the field amplitude ε. Thermodynamic consistency requires accounting for this effect by dressing the system with the field prior to the weak coupling reduction procedure.[13] The result is field dependent expressions for coupling terms with the baths $\mathcal{L}_h$ and $\mathcal{L}_c$. These expressions are completely determined by the bath correlation functions.[13,14]

The quantum thermodynamic observables are identified by substituting the Hamiltonian $\mathbf{H}$ in the Heisenberg equations of motion Eq. (5), which leads to the time derivative of the first law of thermodynamics

$$\frac{d\langle \mathbf{H}\rangle}{dt}=\left\langle \frac{\partial \mathbf{V}}{\partial t}\right\rangle-\langle \boldsymbol{\mathcal{L}}_h(\mathbf{H})\rangle+\langle \boldsymbol{\mathcal{L}}_c(\mathbf{H})\rangle \tag{6}$$

(recall that all energy flows are defined as positive). The power P is identified as $\langle \partial \mathbf{V}/\partial t\rangle$, and the heat currents are identified as $\dot{Q}_h=\langle \mathcal{L}_h(\mathbf{H})\rangle$ for the hot bath and $\dot{Q}_c=\langle \mathcal{L}_c(\mathbf{H})\rangle$ for the cold bath.[14–17] Since internal energy is a state function and device operation is cyclic (so transients can be ignored), it follows that $P-\dot{Q}_h+\dot{Q}_c=0$.

Under operating conditions where the field-induced splitting of levels 2 and 3 is much larger than k_BT, the general expression for the thermodynamic currents is simplified. The steady-state power P^{ss} and heat flow from the cold bath $\dot{Q}_c^{ss}$ become a product of three terms, respectively

$$P^{ss}=\omega_0 k_{\mathrm{eff}}(n_c^- - n_h^-), \tag{7}$$

$$\dot{Q}_c^{ss}=k_{\mathrm{eff}}(\Delta E_{21}-\varepsilon)(n_c^- - n_h^-) \tag{8}$$

$(n_c^- \geqslant n_h^-)$. The population difference term in Eqs. (7) and (8) refers to the unnormalized lower dressed Boltzmann terms $n_i^{\pm}$

$$n_h^{\pm}=\exp[-\beta_h(\Delta E_{31}\pm\varepsilon)],\quad n_c^{\pm}=\exp[-\beta_c(\Delta E_{21}\pm\varepsilon)], \tag{9}$$

where $\beta_h=1/(k_BT_h)$ and $\beta_c=1/(k_BT_c)$.

The energy quanta of the transition are ω_0 for the power input and $(\Delta E_{21}-\varepsilon)$ for the heat flow. k_{eff} is an effective rate of transition. For strong fields, defined by $\varepsilon \gg \Delta E_{32}-\omega_0$, only the lower level of the Rabi split levels 2 and 3 is relevant to cooling, where the effective rate is given by[14] $k_{\mathrm{eff}}=\lambda_h^-\lambda_c^-/[2(\lambda_h^-+\lambda_c^-)]$. The parameters λ_i are determined by the bath correlation functions $\lambda_i^{\pm}=\tilde{C}^i_{\Lambda\Lambda^\dagger}(\Delta E_{32}\pm\varepsilon)$, with $\tilde{C}^i_{\Lambda\Lambda^\dagger}(\nu)$ being the Fourier transform of the bath correlation function corresponding to the system–bath coupling operator Λ.[14] Detailed balance implies the Kubo relation $\tilde{C}^i_{\Lambda\Lambda^\dagger}(\nu)=\tilde{C}^i_{\Lambda\Lambda^\dagger}(-\nu)\exp(-\beta_i\nu)$. In the limits of (1) a weak driving field, (2) $\varepsilon\rightarrow 0$, and (3) zero detuning, ω_0 approaches ΔE_{32}, and the effective rate becomes

$$k_{\mathrm{eff}}=\left[\frac{4\varepsilon^2}{\lambda_h^-\lambda_c^-+4\varepsilon^2}\right]\left[\frac{\lambda_h^-\lambda_c^-}{2(\lambda_h^-+\lambda_c^-)}\right].$$

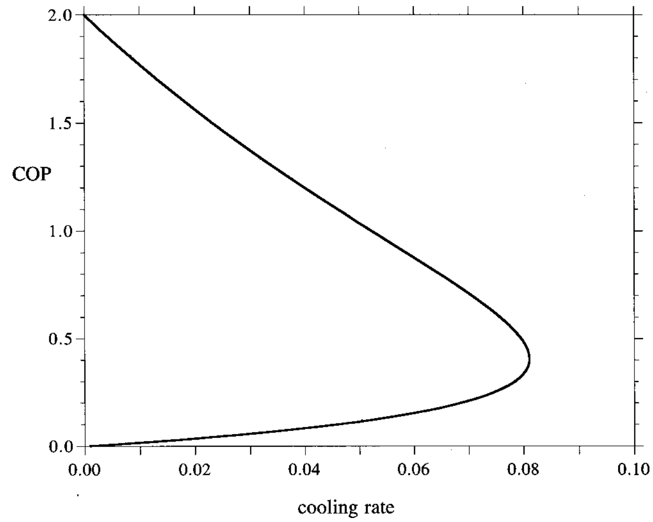

FIG. 3. The efficiency-cooling rate characteristic of the modeled refrigeration cycle, COP as a function of $\dot{Q}_c$, plotted for parameter values: $\beta_h=1$, $\beta_c=1.5$, $V_{\max}=1$, and $k_{\mathrm{eff}}=1$.

As the cold bath grows colder, the population of level 2, n_c^-, decreases until the cooling stops. Eventually, the cold bath temperature reaches the limit of Eq. (3), $T_c=T_h(\Delta E_{21}-\varepsilon)/(\Delta E_{31}-\varepsilon)$. To maintain cooling, one must lower level 2 as the temperature of the cold bath decreases.

The position of level 2 is our control parameter: the "knob" we turn to vary the cooling rate. Shifting the effective energy difference $\Delta E_{21}^{\mathrm{eff}}=\Delta E_{21}-\varepsilon$ allows us to scan all possible cooling rates, and thereby to determine the maximum possible cooling rate. An effective scheme to lower level 2 is either to increase the field ε or to change the position of the level via some external influence. This is a well established strategy when extremely low temperatures need to be reached.[18] Lowering level 2 lessens the rate of cooling since it decreases the quantum of energy exchange. As a result, the cooling rate $\dot{Q}_c$ reaches a maximum between the two points: (1) where $\dot{Q}_c$ vanishes at $\Delta E_{21}^{\mathrm{eff}}=0$, and (2) the equilibrium point where $n_c^-=n_h^-$ (see Fig. 3).

IV. CURRENT–VOLTAGE ANALOGY

The variables of our quantum refrigerator model can be cast in terms of the current and voltage of a simple analogous electrical device. The current (I) is the analog of the population difference $n_c^- - n_h^-$. The voltage (V) is the analog of the energy level difference $\Delta E_{32}^{\mathrm{eff}}=\Delta E_{31}^{\mathrm{eff}}-\Delta E_{21}^{\mathrm{eff}}$, with its maximum value being $V_{\max}=\Delta E_{31}^{\mathrm{eff}}$.

The formulas for the power input, cooling rate, heat rejection rate, and COP can then, respectively, be expressed as

$$P=k_{\mathrm{eff}}VI, \tag{10}$$

$$\dot{Q}_c=k_{\mathrm{eff}}(V_{\max}-V)I, \tag{11}$$

$$\dot{Q}_h=k_{\mathrm{eff}}V_{\max}I, \tag{12}$$

$$\mathrm{COP}=(V_{\max}-V)/V. \tag{13}$$

The current–voltage characteristic is

$$I = \exp[-\beta_c(V_{\max} - V)] - \exp(-\beta_h V_{\max}). \quad (14)$$

The minimum voltage (in the reversible limit of zero current) is $V_{\min} = V_{\max}(1 - T_c/T_h)$, for which the COP approaches $\mathrm{COP}_{\mathrm{Carnot}} = T_c/(T_h - T_c)$. At the other extreme of maximum voltage (and maximum current), the COP vanishes. The efficiency-cooling rate relation for this refrigeration cycle, i.e., COP as a function of $\dot{Q}_c$, is illustrated in Fig. 3.

In real molecular systems, the reversible limit cannot be realized due to additional irreversibility sources that are not incorporated in the simplified model offered here. For example, a refined (future) model should include spontaneous emission. These additional irreversibilities will militate against low-current operation. Namely, COP will vanish in both the high and low current limits, and Fig. 3 will become a loop-shaped curve with distinct points of maximum cooling rate and maximum COP (well below the Carnot limit and at nonzero cooling rate). This is comparable to the familiar cooling performance curve of the thermoelectric refrigerator.[19]

V. MODEL PREDICTIONS VIS-À-VIS THE SECOND AND THIRD LAWS

Because entropy is a state function, cyclic operation means that the cycle-averaged change in the entropy of the working medium is zero. Since no internal dissipation has been introduced in the model, the entire entropy production resides solely at the interface between the system and the baths

$$\dot{S} = \frac{\dot{Q}_h}{T_h} - \frac{\dot{Q}_c}{T_c} = k_{\mathrm{eff}}(\beta_h \Delta E_{31}^{\mathrm{eff}} - \beta_c \Delta E_{21}^{\mathrm{eff}})[\exp(-\beta_c \Delta E_{21}^{\mathrm{eff}}) - \exp(-\beta_h \Delta E_{31}^{\mathrm{eff}})] \geq 0. \quad (15)$$

Since k_{eff} is positive, the product in Eq. (15) is also positive, as required by the second law.

The control variable for changing the cooling rate

$$\dot{Q}_c = k_{\mathrm{eff}} \Delta E_{21}^{\mathrm{eff}}[\exp(-\beta_c \Delta E_{21}^{\mathrm{eff}}) - \exp(-\beta_h \Delta E_{31}^{\mathrm{eff}})] \quad (16)$$

is $\Delta E_{21}^{\mathrm{eff}} = \Delta E_{21} - \varepsilon$. Namely, all other parameters in Eq. (16) are viewed as fixed and known. From threshold scattering laws at low temperature, k_{eff} is only weakly dependent on $\Delta E_{21}^{\mathrm{eff}}$ (Ref. 20). In this model, no mechanism militates against operation in the limit of the maximum permissible $\Delta E_{21}^{\mathrm{eff}}$. Hence $\dot{S} \to 0$ (and $\dot{Q}_c \to 0$) in the reversible limit of $\Delta E_{21}^{\mathrm{eff}} = T_c \Delta E_{31}^{\mathrm{eff}}/T_h$, and $\mathrm{COP} \to \mathrm{COP}_{\mathrm{Carnot}} = T_c/(T_h - T_c)$.

Figure 4 is a plot of cooling rate against $\Delta E_{21}^{\mathrm{eff}}$ at assorted cold bath temperatures as the absolute zero is approached. From Eq. (16), it follows that the maximum cooling rate is of the form

$$\dot{Q}_c^{\max} = (\mathrm{const.}) T_c, \quad (17)$$

where the constant in Eq. (17) depends on k_{eff}, T_h, and $\Delta E_{31}^{\mathrm{eff}}$. Note that the validity of Eqs. (15)–(17) is not restricted to the limit $T_c \to 0$.

The maximum obtainable cooling rate vanishes precisely linearly with T_c, which, as noted in Sec. I, is the weakest functional dependence commensurate with the second and third laws. Similarly, because the power input does not vanish in the limit of the absolute zero, the COP of the quantum refrigerator operating at maximum cooling rate also vanishes linearly in T_c (as does the Carnot COP).

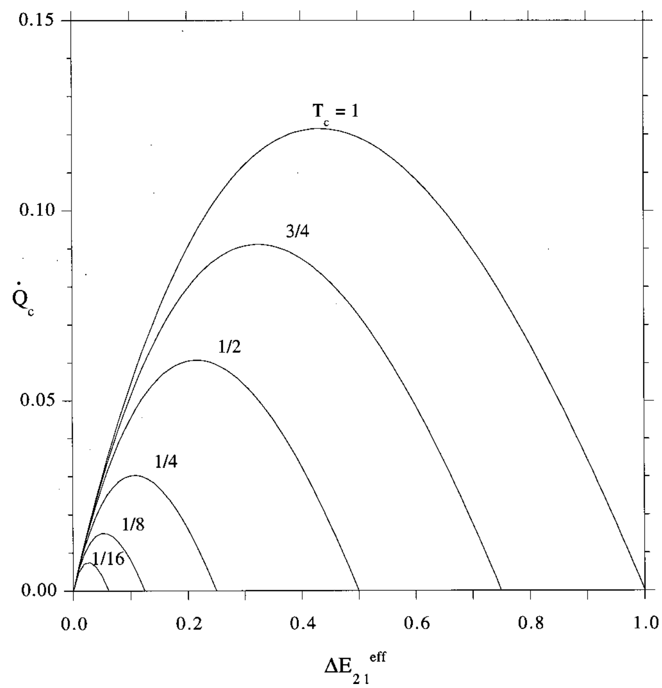

FIG. 4. Plot of cooling rate $\dot{Q}_c$ as a function of the control parameter $\Delta E_{21}^{\mathrm{eff}} = \Delta E_{21} - \varepsilon$ (the effective energy quantum), at different cold bath temperatures ($T_c = 1/\beta_c$). The other fixed parameters used are $\Delta E_{31}^{\mathrm{eff}} = 1$ and $\beta_h = 1$. Note that the equilibrium point and the point of maximum $\dot{Q}_c$ shift to lower $\Delta E_{21}^{\mathrm{eff}}$ as the cold bath temperature is lowered.

Closed-form formulas emerge in the limit of vanishing T_c, where the population difference $n_c^- - n_h^-$ grows small

$$\dot{Q}_c^{\max} = (k_{\mathrm{eff}}/4)(\Delta E_{31}^{\mathrm{eff}}/T_h)^2 \exp(-\beta_h \Delta E_{31}^{\mathrm{eff}}) T_c, \quad (18)$$

$$\mathrm{COP}_{\dot{Q}_c = \dot{Q}_c^{\max}} = \left(\frac{\Delta E_{31}^{\mathrm{eff}}}{2\omega_0 T_h}\right) T_c, \quad (19)$$

$$\dot{S}_{\dot{Q}_c = Q_c^{\max}} = \frac{k_{\mathrm{eff}}}{4}\left(\frac{\Delta E_{31}^{\mathrm{eff}}}{T_h}\right)^2 n_h. \quad (20)$$

The third law is sometimes stated as the entropy of a system approaching zero at the absolute zero, provided the final state is completely ordered. Because the final state in our scheme is a pure state (and hence of zero entropy), the model and its predictions are commensurate with this alternative formulation of the third law.

In summary, the basic result that, in nearing the absolute zero, the maximum attainable cooling rate must vary at least proportionally to the cold bath temperature, is model independent. Only with a quantum-mechanical (as opposed to a classical physics) model, however, can this limit be properly explored. The simple dynamical three-level cyclic quantum refrigerator is a rigorous example of a system for which the fundamental bound on cooling imposed by the second and third laws can be realized.

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