Quantum refrigerators and the third law of thermodynamics

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The rate of temperature decrease of a cooled quantum bath is studied as its temperature is reduced to absolute zero. The third law of thermodynamics is then quantified dynamically by evaluating the characteristic exponent ζ of the cooling process $\frac{dT(t)}{dt} \sim -T^{\zeta}$ when approaching absolute zero, $T \to 0$. A continuous model of a quantum refrigerator is employed consisting of a working medium composed either by two coupled harmonic oscillators or two coupled two-level systems. The refrigerator is a nonlinear device merging three currents from three heat baths: a cold bath to be cooled, a hot bath as an entropy sink, and a driving bath which is the source of cooling power. A heat-driven refrigerator (absorption refrigerator) is compared to a power-driven refrigerator. When optimized, both cases lead to the same exponent ζ, showing a lack of dependence on the form of the working medium and the characteristics of the drivers. The characteristic exponent is therefore determined by the properties of the cold reservoir and its interaction with the system. Two generic heat bath models are considered: a bath composed of harmonic oscillators and a bath composed of ideal Bose/Fermi gas. The restrictions on the interaction Hamiltonian imposed by the third law are discussed. In the Appendices, the theory of periodically driven open systems and its implication for thermodynamics are outlined.

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I. INTRODUCTION

Thermodynamics was initially formed as a phenomenological theory, with the fundamental rules assumed as postulates based on experimental evidence. The well-established part of the theory concerns quasistatic macroscopic processes near thermal equilibrium. Quantum theory, on the other hand, treats the dynamical perspective of systems at atomic and smaller length scales. The two disciplines rely upon different sets of axioms. However, one of the first developments, namely Planck's law, which led to the basics of quantum theory, was achieved thanks to consistency with thermodynamics. Einstein, following the ideas of Planck on blackbody radiation, quantized the electromagnetic field [1].

With the establishment of quantum theory, quantum thermodynamics emerged in the quest to reveal the intimate connection between the laws of thermodynamics and their quantum origin [2–19]. In this tradition, the present study is aimed toward the quantum study of the third law of thermodynamics [20–24], in particular quantifying the unattainability principle. Apart from the fundamental interest in the emergence of the third law of thermodynamics from a quantum dynamical system, cooling mechanical systems reveal their quantum character. As the temperature decreases, degrees of freedom freeze out, leaving a simplified dilute effective Hilbert space. Ultracold quantum systems contributed significantly to our understanding of basic quantum concepts. In addition, such systems form the basis for emerging quantum technologies. The necessity to reach ultralow temperatures requires a focus on the cooling process itself, namely quantum refrigeration.

The minimum requirement for constructing a continuous refrigerator is a system connected simultaneously to three reservoirs [25]. These baths are termed hot, cold, and work reservoir, as described in Fig. 1. This framework has to be translated to a quantum description of its components, which includes the Hamiltonian of the system H_s and the implicit description of the reservoirs. We present a careful study on the influence of different components and cooling mechanisms on the cooling process itself. Namely, we consider a working medium composed of two harmonic oscillators or two two-level systems (TLSs). Two generic models of the cold heat bath are considered: a phonon and an ideal Bose/Fermi gas heat bath. Another classification of the refrigerator is due to the character of the work reservoir. The first studied example is a heat-driven refrigerator, an absorption refrigerator model proposed in Ref. [24], where $T_w \gg T_h \geqslant T_c$.[1] In a power-driven refrigerator, the work reservoir represents zero entropy mechanical work, which is modeled as a periodic time-dependent interaction Hamiltonian.

The models studied contain universal quantum features of such devices. The third law of thermodynamics is quantified by the characteristic exponent ζ of the change in temperature of the cold bath $\frac{dT_c(t)}{dt} \sim -T_c^{\zeta}$ when its temperature approaches absolute zero, $T_c \to 0$. The exponent ζ is determined by a balance between the heat capacity of the cold bath and the heat current $\mathcal{J}_c$ into the cooling device. When the performance of the refrigerator is optimized, the final third-law characteristics are found to be independent of the refrigerator type.

The analysis is based on a steady-state operational mode of the refrigerator. Then the first and second laws of thermodynamics have the form

$$\tilde{\mathcal{J}}_h + \tilde{\mathcal{J}}_c + \mathcal{P} = 0, \qquad -\frac{\tilde{\mathcal{J}}_h}{T_h} - \frac{\tilde{\mathcal{J}}_c}{T_c} - \frac{\mathcal{P}}{T_w} \geqslant 0, \tag{1}$$

where $\tilde{\mathcal{J}}_k$ are the stationary heat currents from each reservoir. The first equality represents conservation of energy (first law) [3,4], and the second inequality represents non-negative entropy production in the Universe, $\Sigma_u \geqslant 0$ (second law). The

[1]A similar idea was also proposed in Phys. Rev. Lett. **108**, 120603 (2012) by Cleuren *et al*. However, one can show that this model violates the third law. The reason for this will be discussed elsewhere.

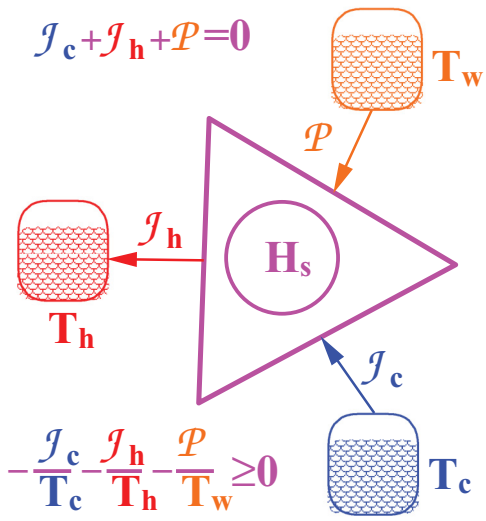

FIG. 1. (Color online) A quantum heat pump designated by the Hamiltonian H_s coupled to a work reservoir with temperature T_w, a hot reservoir with temperature T_h, and a cold reservoir with temperature T_c. The heat and work currents are indicated. In steady state, $\mathcal{J}_h + \mathcal{J}_c + \mathcal{P} = 0$.

fulfillment of the thermodynamic laws is employed to check the consistency of the quantum description. Inconsistencies can emerge either from wrong definitions of the currents $\mathcal{J}_k$ or from erroneous derivations of the quantum master equation. In Appendix A, we present a short and heuristic derivation of such a consistent Markovian master equation based on the rigorous weak coupling [26] or low density [27] limits for a constant system's Hamiltonian. Its generalization to periodic driving proposed in Ref. [28] and based on the Floquet theory is briefly discussed in Appendix B. In Appendix C the definition of heat currents is proposed which satisfies the second law of thermodynamics, not only for the stationary state but also during the evolution from an arbitrary initial state of the system. It allows us also to compute an averaged power in the stationary state. Finally, in Appendix D we discuss the condition on the interaction with a bosonic bath, to assure the existence of the ground state.

II. QUANTUM ABSORPTION REFRIGERATORS

We develop and discuss in detail the model of a quantum absorption refrigerator proposed in Ref. [24]. We extend the results of Ref. [24] treating in the same way the original model with two harmonic oscillators and its two two-level systems counterpart to stress the universality of the proposed cooling mechanism. The advantage of the absorption refrigerator is its underlying microscopic model with a time-independent Hamiltonian.

A. Absorption refrigerator model

The model consists of two harmonic oscillators or two TLSs (A and B) which are described by two pairs of annihilation and creation operators satisfying the commutation or anticommutation relations

$$aa^\dagger + \epsilon a^\dagger a = 1, \quad aa + \epsilon aa = 0,$$
$$bb^\dagger + \epsilon b^\dagger b = 1, \quad bb + \epsilon bb = 0 \tag{2}$$

with $\epsilon = 1$ for the TLS and $\epsilon = -1$ for oscillators. Each subsystem A (B) is coupled to a hot (cold) bath at the temperature T_h (T_c). A collective coupling of the system $A + B$ to the third "work bath" at the temperature $T_w \gg T_h > T_c$ generates heat transport. The nonlinear coupling to the "work bath" is essential. A linearly coupled working medium cannot operate as a refrigerator.[2] The Hamiltonian of the working medium $A + B$ is given by

$$H = \omega_h a^+ a + \omega_c b^+ b, \quad \omega_h > \omega_c, \tag{3}$$

and the interaction with the three baths (hot, cold, and work) is assumed to be of the following form:

$$H_{\rm int} = (a + a^+) \otimes R_h + (b + b^+) \otimes R_c + (ab^+ + a^+ b) \otimes R_w, \tag{4}$$

with $R_{(\cdot)}$ being the corresponding bath operator. The third term in Eq. (4) contains the generator of a swap operation between A and B subsystems [29].

Applying now the derivation of the Markovian dynamics based on the weak-coupling limit (see Appendix A), one obtains the following Markovian master equation involving three thermal generators:

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho] + \mathcal{L}_h\rho + \mathcal{L}_c\rho + \mathcal{L}_w\rho, \tag{5}$$

where

$$\mathcal{L}_h\rho = \tfrac{1}{2}\gamma_h([a,\rho a^\dagger] + e^{-\beta_h\omega_h}[a^\dagger,\rho a] + \text{H.c.}), \tag{6}$$

$$\mathcal{L}_c\rho = \tfrac{1}{2}\gamma_c([b,\rho b^\dagger] + e^{-\beta_c\omega_c}[b^\dagger,\rho b] + \text{H.c.}), \tag{7}$$

$$\mathcal{L}_w\rho = \tfrac{1}{2}\gamma_w([ab^\dagger,\rho a^\dagger b] + e^{-\beta_w(\omega_h-\omega_c)}[a^\dagger b,\rho ab^\dagger] + \text{H.c.}), \tag{8}$$

and $\beta_c > \beta_h \gg \beta_w$ are inverse temperatures for the cold, hot, and work bath, respectively.

The values of relaxation rates $\gamma_h,\gamma_c,\gamma_w > 0$ depend on the particular models of heat baths, and their explicit form is discussed in Appendix A. Notice that one can add also the generators describing pure decoherence (dephasing) in the form

$$\mathcal{D}_h\rho = -\tfrac{1}{2}\delta_h[a^\dagger a,[a^\dagger a,\rho]],$$
$$\mathcal{D}_c\rho = -\tfrac{1}{2}\delta_c[b^\dagger b,[b^\dagger b,\rho]], \quad \delta_h,\delta_c > 0, \tag{9}$$

which, however, do not change the evolution of diagonal matrix elements and therefore have no influence on the cooling mechanism at the stationary state. The generator $\mathcal{L}_w$ is not ergodic in the sense that it does not drive the system $A + B$ to a Gibbs state because it preserves a total number of excitations $a^\dagger a + b^\dagger b$. This fault can be easily repaired by adding to $\mathcal{L}_w$ a term of the form (6) or/and (7) but with the temperature T_w. However, we assume that the processes described by Eqs. (6)–(8) dominate and additional contributions can be neglected.

B. The cooling mechanism

The stationary "cold current" describing heat flux from the cold bath to the working medium can be computed using the

[2]E. Martinez and J. P. Paz (unpublished).

definitions presented in Appendix C. The cooling of the cold bath takes place if this current is positive,

$$\tilde{\mathcal{J}}_c = \omega_c \mathrm{Tr}[(\mathcal{L}_c\tilde{\rho})b^\dagger b] > 0. \tag{10}$$

To compute $\tilde{\mathcal{J}}_c$, we need the following equations for the mean values of the relevant observables $\bar{n}_h = \mathrm{Tr}(\rho a^\dagger a)$ and $\bar{n}_c = \mathrm{Tr}(\rho b^+ b)$, which can be derived using the explicit form of the generators (6)–(8):

$$\frac{d}{dt}\bar{n}_h = -\gamma_h(1+\epsilon e^{-\beta_h\omega_h})\bar{n}_h + \gamma_h e^{-\beta_h\omega_h} + \gamma_w(\bar{n}_c - \bar{n}_h) - R, \tag{11}$$

$$\frac{d}{dt}\bar{n}_c = -\gamma_c(1+\epsilon e^{-\beta_c\omega_c})\bar{n}_c + \gamma_c e^{-\beta_c\omega_c} + \gamma_w(\bar{n}_h - \bar{n}_c) + R, \tag{12}$$

where R is the nonlinear rate

$$R = \gamma_w(1 - e^{-\beta_w(\omega_h-\omega_c)})\mathrm{Tr}[\rho(b^+b)(aa^+)]. \tag{13}$$

Equations (11)–(13) can be solved analytically in the high-temperature limit for the work bath $\beta_w \to 0$, which implies $R \to 0$. Under this condition, the stationary cold current reads

$$\tilde{\mathcal{J}}_c = \omega_c\gamma_w \times \frac{(e^{\beta_c\omega_c}+\epsilon)^{-1} - (e^{\beta_h\omega_h}+\epsilon)^{-1}}{1+\gamma_w[\gamma_h^{-1}(1+\epsilon e^{-\beta_h\omega_h})^{-1} + \gamma_c^{-1}(1+\epsilon e^{-\beta_c\omega_c})^{-1}]}. \tag{14}$$

The cooling condition $\tilde{\mathcal{J}}_c > 0$ is equivalent to a very simple one,

$$\frac{\omega_c}{\omega_h} < \frac{T_c}{T_h}. \tag{15}$$

One can similarly compute the other heat currents to obtain the coefficient of performance (COP),

$$\mathrm{COP} = \frac{\mathcal{J}_c}{\mathcal{J}_w} = \frac{\omega_c}{\omega_h - \omega_c}, \tag{16}$$

which becomes the Otto cycle COP [14,30].

We are interested in the final stage of the cooling process when the temperature T_c is close to absolute zero and hence we can assume that $\gamma_c(T_c) \ll \gamma_h(T_h)$. Optimizing the cooling current means keeping essentially constant the value of ω_c/T_c [31]. This leads to the following simplification of the formula (14):

$$\tilde{\mathcal{J}}_c \simeq \omega_c\gamma_c e^{-\omega_c/k_BT_c}. \tag{17}$$

III. PERIODICALLY DRIVEN REFRIGERATOR

An alternative to driving the refrigerator by a "very hot" heat bath is to apply a time-dependent perturbation to the system of the two harmonic oscillators. One can repeat the derivation for two TLSs, but the final expressions for the currents are more intricate and therefore we restrict ourselves to the oscillator working medium. The time-dependent Hamiltonian reads

$$H(t) = \omega_h a^\dagger a + \omega_c b^\dagger b + \lambda(e^{-i\Omega t}a^\dagger b + e^{i\Omega t}ab^\dagger), \tag{18}$$

where Ω denotes the driving frequency which is chosen to be in resonance $\Omega = \omega_h - \omega_c$ and $\lambda > 0$ measures the strength of the coupling to the external field. Interaction with the baths is given by

$$H_{\mathrm{int}} = (a + a^\dagger) \otimes R_h + (b + b^\dagger) \otimes R_c. \tag{19}$$

The general derivation of the weak-coupling limit Markovian master equation with periodic driving is discussed in Appendix B and is essential for consistency with the second law of thermodynamics [6]. The master equation has the form

$$\frac{d}{dt}\rho(t) = -i[H(t),\rho(t)] + \mathcal{L}_h(t)\rho(t) + \mathcal{L}_c(t)\rho(t) \tag{20}$$

with $\mathcal{L}_{h(c)}(t) = U(t,0)\mathcal{L}_{h(c)}U(t,0)^\dagger$, which under resonance conditions can be derived directly without applying the full Floquet formalism.

The main ingredients of the derivation are as follows:

(i) Transformation to interaction picture. The bath operators transform according to the free baths Hamiltonian, and the system operators transform according to the unitary propagator (under resonance conditions),

$$U(t,0) = \mathcal{T}\exp\left\{-i\int_0^t H(s)ds\right\} = e^{-iH_0t}e^{-iVt}, \tag{21}$$

where

$$H_0 = \omega_h a^\dagger a + \omega_c b^\dagger b, \quad V = \lambda(a^\dagger b + ab^\dagger). \tag{22}$$

(ii) Fourier decomposition of the interaction part,

$$a(t) = U(t,0)^\dagger a U(t,0) = e^{iVt}[e^{iH_0t}ae^{-iH_0}]e^{-iVt} = \cos(\lambda t)e^{-i\omega_h t}a - i\sin(\lambda t)e^{-i\omega_h t}b, \tag{23}$$

which gives the Fourier decomposition [compare with Eq. (B3)]

$$a(t) = \frac{1}{\sqrt{2}}(e^{-i(\omega_h^+)t}d_+ + e^{-i(\omega_h^-)t}d_-) \tag{24}$$

and

$$b(t) = \frac{1}{\sqrt{2}}(e^{-i(\omega_c^+)t}d_+ - e^{-i(\omega_c^-)t}d_-), \tag{25}$$

where $d_+ = \frac{a+b}{\sqrt{2}}$, $d_- = \frac{a-b}{\sqrt{2}}$, and $\omega_{h(c)}^\pm = (\omega_{h(c)} \pm \lambda)$. Similarly, we can calculate $a^\dagger(t), b^\dagger(t)$.

(iii) Performing the weak-coupling approximation, the total time-independent (interaction picture) generator has the form

$$\mathcal{L} = \mathcal{L}_h^{(+)} + \mathcal{L}_h^{(-)} + \mathcal{L}_c^{(+)} + \mathcal{L}_c^{(-)}, \tag{26}$$

where

$$\mathcal{L}_{h(c)}^{(+)}\rho = \tfrac{1}{4}\gamma_{h(c)}^{(+)}([d_+,\rho d_+^\dagger] + e^{-\beta_{h(c)}\omega_{h(c)}^+}[d_+^\dagger,\rho d_+] + \mathrm{H.c.}) \tag{27}$$

and

$$\mathcal{L}_{h(c)}^{(-)}\rho = \tfrac{1}{4}\gamma_{h(c)}^{(-)}([d_-,\rho d_-^\dagger] + e^{-\beta_{h(c)}\omega_{h(c)}^-}[d_-^\dagger,\rho d_-] + \mathrm{H.c.}) \tag{28}$$

with the relaxation rates $\gamma_{h(c)}^{(\pm)} = \gamma_{h(c)}(\omega_{h(c)} \pm \lambda)$ discussed explicitly in Appendices A and B. Any such generator and any sum of them possess a unique stationary state (under condition $\omega_{h(c)} \pm \lambda > 0$),

$$\tilde{\rho}_{h(c)}^{(+)} = Z^{-1}\exp[-\beta_{h(c)}\omega_{h(c)}^+ d_+^\dagger d_+] \tag{29}$$

and

$$\tilde{\rho}_{h(c)}^{(-)} = Z^{-1}\exp[-\beta_{h(c)}\omega_{h(c)}^{-} d_{-}^{\dagger} d_{-}]. \tag{30}$$

The steady (time-independent) heat currents can be computed using the definitions of Appendix C. For example, the heat current from the cold bath is given by the sum of entropy flows, which are related to the quasienergies $\omega_c \pm \lambda$, times the bath temperature,

$$\tilde{\mathcal{J}}_c = -k_B T_c\{\mathrm{Tr}[(\mathcal{L}_c^{(+)}\tilde{\rho})\ln\tilde{\rho}_c^{(+)}] + \mathrm{Tr}[(\mathcal{L}_c^{(-)}\tilde{\rho})\ln\tilde{\rho}_c^{(-)}]\}. \tag{31}$$

This current can be calculated analytically. The result is the following:

$$\tilde{\mathcal{J}}_c = \frac{1}{2}\left[\omega_c^{-}\frac{(e^{\beta_c\omega_c^{-}}-1)^{-1}-(e^{\beta_h\omega_h^{-}}-1)^{-1}}{[\gamma_h^{(-)}(1-e^{-\beta_h\omega_h^{-}})]^{-1}+[\gamma_c^{(-)}(1-e^{-\beta_c\omega_c^{-}})]^{-1}} + \omega_c^{+}\frac{(e^{\beta_c\omega_c^{+}}-1)^{-1}-(e^{\beta_h\omega_h^{+}}-1)^{-1}}{[\gamma_h^{(+)}(1-e^{-\beta_h\omega_h^{+}})]^{-1}+[\gamma_c^{(+)}(1-e^{-\beta_c\omega_c^{+}})]^{-1}}\right]. \tag{32}$$

Similarly to Sec. II B when the temperature T_c is close to absolute zero, we can assume $\gamma_c^{(-)} \ll \gamma_h^{(-)}$ and $\gamma_c^{(+)} \ll \gamma_h^{(+)}$ while keeping $\lambda/T_c < \omega_c/T_c$ as constants. This simplifies formula (32),

$$\tilde{\mathcal{J}}_c \simeq \tfrac{1}{2}[\omega_c^{+}\gamma_c^{(+)}e^{-\omega_c^{+}/k_BT_c} + \omega_c^{-}\gamma_c^{(-)}e^{-\omega_c^{-}/k_BT_c}]. \tag{33}$$

Notice that the cold current does not vanish when λ tends to zero, which obviously should be the case. This is due to the fact that the derivation of master equations in the weak-coupling regime involves time-averaging procedures eliminating certain oscillating terms. This procedure makes sense only if the corresponding Bohr frequencies are well-separated. In our case, it means that ω_c^{-} should be well separated from ω_c^{+}, which implies that $\lambda \sim \omega_c$. Indeed, if both ω_c and λ vanish, the cold current vanishes as well. This problem of time scales in the weak-coupling Markovian dynamics has been discussed, for constant Hamiltonians, in Ref. [32] (see also [33] for the related "dynamical symmetry breaking" phenomenon).

IV. THE DYNAMICAL THIRD LAW OF THERMODYNAMICS

There exist two seemingly independent formulations of the third law of thermodynamics, both originally stated by Nernst [20,22]. The first is a purely static (equilibrium) one, also known as the Nernst heat theorem, and can simply be phrased as follows:

(a) The entropy of any pure substance in thermodynamic equilibrium approaches zero as the temperature approaches zero.

The second is a dynamical one, known as the unattainability principle:

(b) It is impossible by any procedure, no matter how idealized, to reduce any assembly to absolute zero temperature in a finite number of operations [34].

Different studies investigating the relation between the two formulations have led to different answers regarding which of these formulations implies the other, or if neither does. Although interesting, this question is beyond the scope of this paper. For further considerations regarding the third law, we refer the reader to Refs. [23,34–38]. In particular, in Refs. [37,38] the validity of the static formulation (a) has been confirmed for a large class of open quantum systems. We shall use a more concrete version of the dynamical third law, which can be expressed as follows:

(b′) No refrigerator can cool a system to absolute zero temperature at finite time.

This formulation enables us to quantify the third law, i.e., evaluating the characteristic exponent ζ of the cooling process $\frac{dT(t)}{dt} \sim -T^{\zeta}$ for $T \to 0$. Namely, for $\zeta < 1$ the system is cooled to zero temperature at finite time. As a model of the refrigerator, we use the above-discussed continuous refrigerators with a cold bath modeled either by a system of harmonic oscillators (bosonic bath) or the ideal gas at low density, including the possible Bose-Einstein condensation effect. To check under what conditions the third law is valid, we consider a finite cold bath with the heat capacity $c_V(T_c)$ cooled down by the refrigerator with the optimized time-dependent parameter $\omega_c(t)$ and the additional parameter $\lambda(t)$ for the case of a periodically driven refrigerator. The equation which describes the cooling process reads

$$c_V[T_c(t)]\frac{dT_c(t)}{dt} = -\mathcal{J}_c[\omega_c(t),T_c(t)], \quad t \geqslant 0. \tag{34}$$

The third law would be violated if the solution $T_c(t)$ reached zero at finite time t_0. Now we can consider two generic models of the cold heat bath.

A. Harmonic oscillator cold heat bath

This is a generic type of quantum bath including, for example, an electromagnetic field in a large cavity or a finite but macroscopic piece of solid described in the thermodynamic limit. We assume the linear coupling to the bath and the standard form of the bath's Hamiltonian,

$$H_{\mathrm{int}} = (b+b^{\dagger})\left(\sum_k [g(k)a(k) + \bar{g}(k)a^{\dagger}(k)]\right),$$
$$H_B = \sum_k \omega(k)a^{\dagger}(k)a(k), \tag{35}$$

where $a(k)$,$a^{\dagger}(k)$ are annihilation and creation operators for mode k. For this model, the weak-coupling limit procedure leads to the generator (7) with the cold bath relaxation rate

$$\gamma_c \equiv \gamma_c(\omega_c) = \pi\sum_k |g(k)|^2\delta(\omega(k)-\omega_c)[1-e^{-\omega(k)/k_BT_c}]^{-1}. \tag{36}$$

For the bosonic field in d-dimensional space, where k is a wave vector, and with the linear low-frequency dispersion law $[\omega(k) \sim |k|]$, we obtain the following scaling properties at low frequencies (compare Appendix D):

$$\gamma_c \sim \omega_c^{\kappa}\omega_c^{d-1}[1-e^{-\omega_c/k_BT_c}]^{-1}, \tag{37}$$

where ω_c^{κ} represents scaling of the coupling strength $|g(\omega)|^2$, and ω_c^{d-1} is the density of modes scaling. This implies the

following scaling of the cold current:

$$\mathcal{J}_c \sim T_c^{d+\kappa}\left[\frac{\omega_c}{T_c}\right]^{d+\kappa}\frac{1}{e^{\omega_c/T_c}-1}. \tag{38}$$

Optimization of Eq. (38) with respect to ω_c leads to the frequency tuning $\omega_c \sim T_c$ and the final current scaling,

$$\mathcal{J}_c^{\rm opt} \sim T_c^{d+\kappa}. \tag{39}$$

Consider that for low temperatures the heat capacity of the bosonic systems scales like

$$c_V(T_c) \sim T_c^d, \tag{40}$$

which finally produces the following scaling of the dynamical equation (34):

$$\frac{dT_c(t)}{dt} \sim -(T_c)^\kappa. \tag{41}$$

Notice that in a similar way the same scaling (41) is achieved for the periodically driven refrigerator (33), with the optimization tuning $\omega_c,\lambda \sim T_c$. As a consequence, the third law implies a rather unexpected constraint on the form of interaction with a bosonic bath,

$$\kappa \geqslant 1. \tag{42}$$

For standard systems such as electromagnetic fields or acoustic phonons with the linear dispersion law $\omega(k) = v|k|$ and the form factor $g(k) \sim |k|/\sqrt{\omega(k)}$, the parameter $\kappa = 1$, as for low ω, $|g(\omega)|^2 \sim |k|$. However, the condition (42) excludes exotic dispersion laws $\omega(k) \sim |k|^\alpha$ with $\alpha < 1$, which nevertheless produce the infinite group velocity forbidden by the relativity theory. Moreover, the popular choice of Ohmic coupling is excluded for systems in dimension $d > 1$. The condition (42) can also be compared with the condition

$$\kappa > 2 - d, \tag{43}$$

which is necessary to assure the existence of the ground state for the bosonic field interacting by means of the Hamiltonian (35) (see Appendix D).

B. Ideal Bose/Fermi gas cold heat bath

We consider now a model of a cooling process where part B of the working medium is an (infinitely) heavy particle with the internal structure approximated (at least at low temperatures) by a TLS immersed in a low density gas at temperature T_c. The Markovian dynamics of such a system was rigorously derived by Dumcke [27] in the low density limit and N-level internal structure. The form of the corresponding LGKS generator is presented in Appendix A. For our case of TLS, we have only one Bohr frequency ω_c, because elastic scattering corresponding to $\omega = 0$ does not influence the cooling process. Cooling occurs due to the nonelastic scattering, giving the relaxation rate (Appendix A)

$$\gamma_c = 2\pi n \int d^3\vec{p} \int d^3\vec{p}\,'\delta(E(\vec{p}\,') - E(\vec{p}) - \hbar\omega_c) \times f_{T_c}(\vec{p}_g)|T(\vec{p}\,',\vec{p})|^2 \tag{44}$$

with n the particle density, $f_{T_c}(\vec{p}_g)$ the probability distribution of the gas momentum strictly given by the Maxwell distribution, and $\vec{p}$ and $\vec{p}\,'$ the incoming and outgoing gas particle momentum, respectively. $E(\vec{p}) = p^2/2m$ denotes the kinetic energy of gas particle.

At low energies (low temperature), scattering of neutral gas in three dimensions can be characterized by the s-wave scattering length a_s, having a constant transition matrix, $|T|^2 = (\frac{4\pi a_s}{m})^2$. For our model, the integral (44) is calculated as

$$\gamma_c = (4\pi)^4\left(\frac{\beta_c}{2\pi m}\right)^{\frac{1}{2}} a_s^2 n\omega_c \mathcal{K}_1\left(\frac{\beta_c\omega_c}{2}\right)e^{\frac{\beta_c\omega_c}{2}}, \tag{45}$$

where $\mathcal{K}_p(x)$ is the modified Bessel function of the second kind. Note that formula (45) is also valid for a harmonic oscillator instead of TLS, assuming only linear terms in the interaction and using the Born approximation for the scattering matrix.

Optimizing formula (17) with respect to ω_c leads to $\omega_c \sim T_c$ and to scaling of the heat current,

$$\mathcal{J}_c^{\rm opt} \sim n(T_c)^{\frac{3}{2}}. \tag{46}$$

When the Bose gas is above the critical temperature for the Bose-Einstein condensation, the heat capacity c_V and the density n are constants. Below the critical temperature, the density n in formula (44) should be replaced by the density $n_{\rm ex}$ of the exited states, having both $c_V, n_{\rm ex} \sim (T_c)^{\frac{3}{2}}$, which finally implies

$$\frac{dT_c(t)}{dt} \sim -(T_c)^{\frac{3}{2}}. \tag{47}$$

In the case of Fermi gas at low temperatures, only the small fraction $n \sim T_c$ of fermions participates in the scattering process and contributes to the heat capacity; the rest is "frozen" in the "Dirac sea" below the Fermi surface. Again, this effect modifies in the same way both sides of Eq. (34), and therefore (47) is still valid. Similarly, a possible formation of Cooper pairs below the critical temperature does not influence the scaling (47).

V. CONCLUSIONS

We have introduced and analyzed two types of continuous quantum refrigerators, namely an absorption refrigerator and a periodically driven refrigerator. The latter required us to present new definitions for heat flow for periodically driven open systems. These definitions are in line with the second law and are applicable for a time-independent Hamiltonian as well. Unlike the first and second laws, the third law of thermodynamics does not define a new state function. In its first formulation (cf. Sec. IV), the third law provides a reference point for scaling the entropy and becomes intuitive when thinking in terms of quantum states or levels. The second formulation, (b′) in Sec. IV, which states that no refrigerator can cool a system to absolute zero temperature at finite time, provides information on the characteristic exponent ζ, the speed of cooling, and gives an insight and restriction on the properties of realistic systems.

Universal behavior of the final scaling near absolute zero is obtained. The third law does not depend on the bath dimension. The type of refrigerator, either absorption or a periodically driven refrigerator, does not influence the characteristic exponent, nor does a different medium, i.e., a

harmonic oscillator and a TLS produce the same scaling. The characteristic exponent is governed only by the feature of the heat bath and its interaction with the system. For a harmonic oscillator heat bath, the third law imposes a restriction on the form of the interaction between the system and the bath, $\kappa \geqslant 1$, allowing only physical coupling and dispersion relations, thus for phonons with a linear dispersion relation $\zeta = \kappa = 1$. For an ideal Bose/Fermi gas heat bath, $\zeta = 3/2$, which implies faster cooling of the phonon bath than the gas bath. This distinction between the two baths may occur due to particle conservation for the gas, indicating a more efficient extraction of heat by eliminating particles from the system. The key component of a realistic refrigerator is the heat transport mechanism between the heat bath and the working medium. This mechanism determines the third-law scaling. The working medium is a nonlinear device combining three currents. If it is optimized properly by adjusting its internal structure, it does not pose a limit on cooling.

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APPENDIX A: THERMAL GENERATORS FOR A CONSTANT HAMILTONIAN

Consider a system and a reservoir (bath), with a "bare" system Hamiltonian H^0 and the bath Hamiltonian H_R, interacting via the Hamiltonian $\lambda H_{\rm int} = \lambda S \otimes R$. Here, S (R) is a Hermitian system (reservoir) operator and λ is the coupling strength (a generalization to more complicated $H_{\rm int}$ is straightforward). We assume also that

$$[\rho_R, H_R] = 0, \quad \mathrm{Tr}(\rho_R\, R) = 0. \tag{A1}$$

The reduced, system-only dynamics in the interaction picture is defined as a partial trace,

$$\rho(t) = \Lambda(t,0)\rho \equiv \mathrm{Tr}_R[U_\lambda(t,0)\rho \otimes \rho_R U_\lambda(t,0)^\dagger], \tag{A2}$$

where the unitary propagator in the interaction picture is given by the ordered exponential,

$$U_\lambda(t,0) = \mathcal{T}\exp\left\{\frac{-i\lambda}{\hbar}\int_0^t S(s)\otimes R(s)\,ds\right\}, \tag{A3}$$

where

$$S(t) = e^{(i/\hbar)Ht} S e^{-(i/\hbar)Ht}, \quad R(t) = e^{(i/\hbar)H_R t} R e^{-(i/\hbar)H_R t}. \tag{A4}$$

Notice that $S(t)$ is defined with respect to the renormalized, *physical*, H and not H^0, which can be expressed as

$$H = H^0 + \lambda^2 H_1^{\rm corr} + \cdots. \tag{A5}$$

The renormalizing terms containing powers of λ are *Lamb-shift* corrections due to the interaction with the bath, which cancel afterward the uncompensated term $H - H^0$, which, in principle, should also be present in Eq. (A3). The lowest-order (Born) approximation with respect to the coupling constant λ yields $H_1^{\rm corr}$, while the higher-order terms $(\cdots)$ require going beyond the Born approximation.

A convenient, albeit not used in the rigorous derivations, tool is a cumulant expansion for the reduced dynamics,

$$\Lambda(t,0) = \exp\sum_{n=1}^{\infty}[\lambda^n K^{(n)}(t)]. \tag{A6}$$

One finds that $K^{(1)} = 0$ and the Born approximation (weak coupling) consists of terminating the cumulant expansion at $n = 2$, hence we denote $K^{(2)} \equiv K$:

$$\Lambda(t,0) = \exp[\lambda^2 K(t) + O(\lambda^3)]. \tag{A7}$$

One obtains

$$K(t)\rho = \frac{1}{\hbar^2}\int_0^t ds \int_0^t du\, F(s-u) S(s)\rho S(u)^\dagger + (\text{similar terms}), \tag{A8}$$

where $F(s) = \mathrm{Tr}[\rho_R R(s) R]$. The *similar terms* in Eq. (A8) are of the form $\rho S(s) S(u)^\dagger$ and $S(s)S(u)^\dagger\rho$.

The Markov approximation (in the interaction picture) means in all our cases that for long enough time, one can use the following approximation:

$$K(t) \simeq t\mathcal{L}, \tag{A9}$$

where $\mathcal{L}$ is a Linblad-Gorini-Kossakowski-Sudarshan (LGKS) generator. To find its form, we first decompose $S(t)$ into its Fourier components,

$$S(t) = \sum_{\{\omega\}} e^{i\omega t} S_\omega, S_{-\omega} = S_\omega^\dagger, \tag{A10}$$

where the set $\{\omega\}$ contains *Bohr frequencies* of the Hamiltonian

$$H = \sum_k \epsilon_k |k\rangle\langle k|, \quad \omega = \epsilon_k - \epsilon_l. \tag{A11}$$

Then we can rewrite the expression (A8) as

$$K(t)\rho = \frac{1}{\hbar^2}\sum_{\omega,\omega'} S_\omega \rho S_{\omega'}^\dagger \int_0^t e^{i(\omega-\omega')u} du \int_{-u}^{t-u} F(\tau) e^{i\omega\tau} d\tau + (\text{similar terms}) \tag{A12}$$

and use two crucial approximations:

$$\int_0^t e^{i(\omega-\omega')u} du \approx t\delta_{\omega\omega'},$$
$$\int_{-u}^{t-u} F(\tau) e^{i\omega\tau} d\tau \approx G(\omega) = \int_{-\infty}^{\infty} F(\tau) e^{i\omega\tau} d\tau \geqslant 0. \tag{A13}$$

This makes sense for $t \gg \max\{1/(\omega-\omega')\}$. Applying these two approximations, we obtain $K(t)\rho_S = (t/\hbar^2)\sum_\omega S_\omega \rho_S S_\omega^\dagger G(\omega) + (\text{similar terms})$, and hence it follows from Eq. (A9) that $\mathcal{L}$ is a special case of the LGKS generator derived for the first time by Davies [26]. Returning to the Schrödinger picture, one obtains the following Markovian master equation:

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho] + \mathcal{L}\rho,$$
$$\mathcal{L}\rho \equiv \frac{\lambda^2}{2\hbar^2}\sum_{\{\omega\}} G(\omega)([S_\omega, \rho S_\omega^\dagger] + [S_\omega\rho, S_\omega^\dagger]). \tag{A14}$$

Several remarks are in order:

(i) The absence of off-diagonal terms in Eq. (A14), compared to Eq. (A12), is the crucial property of the Davies generator which can be interpreted as a coarse-graining in time of fast oscillating terms. It implies also the commutation of $\mathcal{L}$ with the Hamiltonian part $[H,\cdot]$.

(ii) The positivity $G(\omega) \geqslant 0$ follows from Bochner's theorem and is a necessary condition for the complete positivity of the Markovian master equation.

(iii) The presented derivation showed implicitly that the notion of *bath's correlation time*, often used in the literature, is not well-defined—Markovian behavior involves a rather complicated cooperation between system and bath dynamics. In other words, contrary to what is often done in phenomenological treatments, *one cannot combine arbitrary H's with a given LGKS generator*. This is particularly important in the context of thermodynamics of controlled quantum open system, where it is common to assume Markovian dynamics and apply arbitrary control Hamiltonians. Erroneous derivations of the quantum master equation can easily lead to violation of the laws of thermodynamics.

If the reservoir is a quantum system at a thermal equilibrium state, the additional Kubo-Martin-Schwinger (KMS) condition holds,

$$G(-\omega) = \exp\left(-\frac{\hbar\omega}{k_B T}\right) G(\omega), \tag{A15}$$

where T is the bath's temperature. As a consequence of Eq. (A15), the Gibbs state

$$\rho_\beta = Z^{-1} e^{-\beta H}, \quad \beta = \frac{1}{k_B T} \tag{A16}$$

is a stationary solution of Eq. (A14). Under mild conditions (e.g., "the only system operators commuting with H and S are scalars"), the Gibbs state is a unique stationary state and any initial state relaxes toward equilibrium ("zeroth law of thermodynamics"). A convenient parametrization of the corresponding *thermal generator* reads

$$\mathcal{L}\rho = \frac{1}{2}\sum_{\{\omega\geqslant 0\}} \gamma(\omega)\{([S_\omega,\rho S_\omega^\dagger] + [S_\omega\rho, S_\omega^\dagger]) + e^{-\hbar\beta\omega}([S_\omega^\dagger,\rho S_\omega] + [S_\omega^\dagger\rho, S_\omega])\}, \tag{A17}$$

where finally

$$\gamma(\omega) = \frac{\lambda^2}{\hbar^2}\int_{-\infty}^{+\infty} \mathrm{Tr}(\rho_R\, e^{iH_R t/\hbar}\, R\, e^{-iH_R t/\hbar} R)\, dt. \tag{A18}$$

A closer look at the expressions (A17) and (A18) shows that the transition ratio from the state $|k\rangle$ to the state $|l\rangle$ is exactly the same as that computed from the Fermi Golden Rule,

$$W(|\mathrm{in}\rangle \to |\mathrm{fin}\rangle) = \frac{2\pi}{\hbar}|\langle \mathrm{in}|V|\mathrm{fin}\rangle|^2 \delta(E_{\mathrm{fin}} - E_{\mathrm{in}}). \tag{A19}$$

Namely, one should take as a perturbation $V = \lambda S \otimes R$, an initial state $|\mathrm{in}\rangle = |k\rangle \otimes |E\rangle$, a final state $|\mathrm{fin}\rangle = |l\rangle \otimes |E'\rangle$ ($|E\rangle$ denotes the reservoir's energy eigenstate), and integrate over the initial reservoir's states with the equilibrium distribution and over all the final reservoir's states.

The above interpretation allows us to justify the extension of the construction of a thermal generator to the case of a heat bath consisting of noninteracting particles at low density n and thermal equilibrium (see [27] for a rigorous derivation). In this case, a fundamental relaxation process is a scattering of a single bath particle with the system described by the scattering matrix T. The scattering matrix can be decomposed as $T = \sum_{\{\omega\}} S_\omega \otimes R_\omega$, where now R_ω are single-particle operators. Then the structure of the corresponding master equation is again given by Eq. (A17) with

$$\gamma(\omega) = 2\pi n \int d^3\vec{p} \int d^3\vec{p}\,'\delta(E(\vec{p}\,') - E(\vec{p}) - \hbar\omega) M(\vec{p})| \times T_\omega(\vec{p}\,',\vec{p})|^2 \tag{A20}$$

resembling a properly averaged expression (A19). Here the initial (final) state has a structure $|k\rangle \otimes |\vec{p}\rangle$ ($|l\rangle \otimes |\vec{p}\,'\rangle$), $M(\vec{p})$ is the equilibrium (Maxwell) initial distribution of particle momenta, with $|\vec{p}\rangle$ being the particle momentum eigenvector, and $E(\vec{p})$ is the kinetic energy of a particle. The perturbation V in Eq. (A19) is replaced by the scattering matrix T (equal to V for the Born approximation) and finally

$$T_\omega(\vec{p}\,',\vec{p}) = \langle \vec{p}\,'|R_\omega|\vec{p}\rangle. \tag{A21}$$

APPENDIX B: THERMAL GENERATORS FOR PERIODIC DRIVING

In order to construct models of quantum heat engines or powered refrigerators, we have to extend the presented derivations of the Markovian master equation to the case of periodically driven systems. Fortunately, we can essentially repeat the previous derivation with the following amendments:

(i) The system (physical, renormalized) Hamiltonian is now periodic,

$$H(t) = H(t+\tau), \quad U(t,0) \equiv \mathcal{T}\exp\left\{-\frac{i}{\hbar}\int_0^t H(s)\, ds\right\}, \tag{B1}$$

and the role of constant Hamiltonian is played by H defined as

$$H = \sum_k \epsilon_k |k\rangle\langle k|, \quad U(\tau,0) = e^{-iH t/\hbar}. \tag{B2}$$

(ii) The Fourier decomposition (A10) is replaced by the following one:

$$U(t,0)^\dagger\, S\, U(t,0) = \sum_{q\in\mathbf{Z}}\sum_{\{\omega\}} e^{i(\omega+q\Omega)t} S_{\omega q}, \tag{B3}$$

where $\Omega = 2\pi/\tau$ and $\{\omega\} = \{\epsilon_k - \epsilon_l\}$. The decomposition of the above follows from the Floquet theory, however for our model we can obtain it directly using the manifest expressions for the propagator $U(t,0)$.

(iii) The generator in the interaction picture has the form

$$\mathcal{L} = \sum_{q\in\mathbf{Z}}\sum_{\{\omega\}} = \mathcal{L}_{\omega q}, \tag{B4}$$

where

$$\mathcal{L}_{\omega q}\rho = \tfrac{1}{2}\gamma(\omega + q\Omega)\{([S_{\omega q},\rho S_{\omega q}^\dagger] + [S_{\omega q}\rho, S_{\omega q}^\dagger]) + e^{-\hbar\beta(\omega+q\Omega)}([S_{\omega q}^\dagger,\rho S_{\omega q}] + [S_{\omega q}^\dagger\rho, S_{\omega q}])\}. \tag{B5}$$

Returning to the Schrödinger picture, we obtain the following master equation:

$$\frac{d\rho(t)}{dt} = -\frac{i}{\hbar}[H(t),\rho(t)] + \mathcal{L}(t)\rho(t), \quad t \geqslant 0, \tag{B6}$$

where

$$\begin{aligned} \mathcal{L}(t) &= \mathcal{L}(t+\tau) = \mathcal{U}(t,0)\mathcal{L}\mathcal{U}(t,0)^\dagger, \\ \mathcal{U}(t,0)\cdot &= U(t,0)\cdot U(t,0)^\dagger. \end{aligned} \tag{B7}$$

In particular, one can represent the solution of Eq. (B6) in the form

$$\rho(t) = \mathcal{U}(t,0)e^{\mathcal{L}t}\rho(0), \quad t \geqslant 0. \tag{B8}$$

Any state satisfying $\mathcal{L}\tilde{\rho} = 0$ defines a periodic steady state (limit cycle),

$$\tilde{\rho}(t) = \mathcal{U}(t,0)\tilde{\rho} = \tilde{\rho}(t+\tau), \quad t \geqslant 0. \tag{B9}$$

Finally, one should notice that in the case of multiple couplings and multiple heat baths, the generator $\mathcal{L}$ can always be represented as an appropriate sum of the terms like (A17).

APPENDIX C: HEAT FLOWS AND POWER FOR PERIODICALLY DRIVEN OPEN SYSTEMS

We consider a periodically driven system coupled to several heat baths with the additional index j labeling them. Then the generator in the interaction picture has the form

$$\mathcal{L} = \sum_{j=1}^{M}\sum_{q\in\mathbf{Z}}\sum_{\{\omega\geqslant 0\}} \mathcal{L}^j_{\omega q}, \tag{C1}$$

where any single $\mathcal{L}^j_{\omega q}$ has a structure of Eq. (B5) with the appropriate $\gamma_j(\omega)$. Notice that a single component $\mathcal{L}^j_{\omega q}$ is also a LGKS generator and possesses a Gibbs-like stationary state written in terms of the averaged Hamiltonian H,

$$\tilde{\rho}^j_{\omega q} = Z^{-1}\exp\left\{-\frac{\omega+q\Omega}{\omega}\frac{H}{k_B T_j}\right\}. \tag{C2}$$

The corresponding time-dependent objects satisfy

$$\begin{aligned} \mathcal{L}^j_{q\omega}(t)\tilde{\rho}^j_{q\omega}(t) = 0, \quad \mathcal{L}^j_{q\omega}(t) &= \mathcal{U}(t,0)\mathcal{L}^j_{q\omega}\mathcal{U}(t,0)^\dagger, \\ \tilde{\rho}^j_{q\omega}(t) = \mathcal{U}(t,0)\tilde{\rho}^j_{q\omega} &= \tilde{\rho}^j_{q\omega}(t+\tau). \end{aligned} \tag{C3}$$

Using the decomposition (C1), one can define a *local heat current* which corresponds to the exchange of energy $\omega + q\Omega$ with the jth heat bath for any initial state,

$$\mathcal{J}^j_{q\omega}(t) = \frac{\omega+q\Omega}{\omega}\mathrm{Tr}\{[\mathcal{L}^j_{q\omega}(t)\rho(t)]\tilde{H}(t)\}, \quad \tilde{H}(t) = \mathcal{U}(t,0)H, \tag{C4}$$

or in the equivalent form,

$$\mathcal{J}^j_{q\omega}(t) = -k_B T_j \mathrm{Tr}\{[\mathcal{L}^j_{q\omega}(t)\rho(t)]\ln\tilde{\rho}^j_{q\omega}(t)\}. \tag{C5}$$

The heat current associated with the jth bath is a sum of the corresponding local ones,

$$\mathcal{J}^j(t) = -k_B T_j \sum_{q\in\mathbf{Z}}\sum_{\{\omega\geqslant 0\}} \mathrm{Tr}\{[\mathcal{L}^j_{q\omega}(t)\rho(t)]\ln\tilde{\rho}^j_{q\omega}(t)\}. \tag{C6}$$

In order to prove the second law, we use Spohn's inequality [3],

$$\mathrm{Tr}([\mathcal{L}\rho][\ln\rho - \ln\tilde{\rho}]) \leqslant 0, \tag{C7}$$

which is valid for any LGKS generator $\mathcal{L}$ with a stationary state $\tilde{\rho}$.

Computing now the time derivative of the entropy $S(t) = -k_B\mathrm{Tr}\rho(t)\ln\rho(t)$ and applying (C7), one obtains the second law in the form

$$\frac{d}{dt}S(t) - \sum_{j=1}^{M}\frac{\mathcal{J}^j(t)}{T_j} \geqslant 0, \tag{C8}$$

where $S(t) = -k_B\mathrm{Tr}[\rho(t)\ln\rho(t)]$.

The heat currents in the steady state $\tilde{\rho}(t)$ are time-independent and given by

$$\tilde{\mathcal{J}}^j = -k_B T_j \sum_{q\in\mathbf{Z}}\sum_{\{\omega\geqslant 0\}} \mathrm{Tr}\big[\big(\mathcal{L}^j_{q\omega}\tilde{\rho}\big)\ln\tilde{\rho}^j_{q\omega}\big]. \tag{C9}$$

They satisfy the second law in the form

$$\sum_{j=1}^{M}\frac{\tilde{\mathcal{J}}^j}{T_j} \leqslant 0 \tag{C10}$$

while, according to the first law,

$$-\sum_{j=1}^{M}\tilde{\mathcal{J}}^j = -\tilde{\mathcal{J}} = \bar{\mathcal{P}} \tag{C11}$$

is the averaged power (negative when the system acts as a heat engine). Notice that in the case of a single heat bath, the heat current is always strictly positive except for the case of no driving, when it is equal to zero.

Notice that for the constant Hamiltonian, the above formulas are also applicable after removing the index q, which implies also that $\sum_{j=1}^{M}\tilde{\mathcal{J}}^j = 0$.

APPENDIX D: VAN HOVE PHENOMENON

A natural physical stability condition which should be satisfied by any model of an open quantum system is that its total Hamiltonian should be bounded from below and should possess a ground state. In the case of systems coupled linearly to bosonic heat baths, it implies the existence of the ground state for the following bosonic Hamiltonian [compare with Eq. (35)]:

$$H_{\mathrm{bos}} = \sum_k\{\omega(k)a^\dagger(k)a(k) + [g(k)a(k) + \bar{g}(k)a^\dagger(k)]\}. \tag{D1}$$

Introducing a formal transformation to a new set of bosonic operators,

$$a(k) \mapsto b(k) = a(k) + \frac{\bar{g}(k)}{\omega(k)}, \tag{D2}$$

we can write

$$H_{\mathrm{bos}} = \sum_k \omega(k)b^\dagger(k)b(k) - E_0, \quad E_0 = \sum_k\frac{|g(k)|^2}{\omega(k)} \tag{D3}$$

with the formal ground state $|0\rangle$ satisfying

$$b(k)|0\rangle = 0 \quad \text{for all} \quad k. \tag{D4}$$

For the interesting case of an infinite set of modes $\{k\}$, labeled by the d-dimensional wave vectors, two problems can appear:

(i) The ground state energy E_0 can be infinite, i.e., it does not satisfy

$$\sum_k \frac{|g(k)|^2}{\omega(k)} < \infty. \tag{D5}$$

(ii) The transformation (D2) can be implemented by a unitary one, i.e., $b(k) = Ua(k)U^\dagger$ if and only if

$$\sum_k \frac{|g(k)|^2}{\omega(k)^2} < \infty. \tag{D6}$$

Nonexistence of such a unitary implies nonexistence of the ground state (D4) (in the Fock space of the bosonic field), and this is called the *van Hove phenomenon* [39].

While the divergence of the sums (D5) and (D6) (or integrals for the infinite volume case) for large $|k|$ can be avoided by applying the *ultraviolet cutoff*, the stronger condition (D6) puts restrictions on the form of $g(k)$ at low frequencies. Assuming that $\omega(k) = v|k|$ and $g(k) \equiv g(\omega)$, the condition (D6) is satisfied for the following low-frequency scaling in the d-dimesional case:

$$|g(\omega)|^2 \sim \omega^\kappa, \quad \kappa > 2 - d. \tag{D7}$$

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