Quantum Carnot thermal machines reexamined: Definition of efficiency and the effects of strong coupling

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Whether the strong coupling to thermal baths can improve the performance of quantum thermal machines remains an open issue under active debate. Here we revisit quantum thermal machines operating with the quasistatic Carnot cycle and aim to unveil the role of strong coupling in maximum efficiency. Our analysis builds upon definitions of excess work and heat derived from an exact formulation of the first law of thermodynamics for the working substance, which captures the non-Gibbsian thermal equilibrium state that emerges at strong couplings during quasistatic isothermal processes. These excess definitions differ from conventional ones by an energetic cost for maintaining the non-Gibbsian characteristics. With this distinction, we point out that one can introduce two different yet thermodynamically allowed definitions for efficiency of both the heat engine and refrigerator modes. We dub them excess and hybrid definitions, which differ in the way of defining the gain for the thermal machines at strong couplings by either just analyzing the energetics of the working substance or instead evaluating the performance from an external system upon which the thermal machine acts, respectively. We analytically demonstrate that the excess definition predicts that the Carnot limit remains the upper bound for both operation modes at strong couplings, whereas the hybrid one reveals that strong coupling can suppress the maximum efficiency rendering the Carnot limit unattainable. These seemingly incompatible predictions thus indicate that it is imperative to first gauge the definition for efficiency before elucidating the exact role of strong coupling, thereby shedding light on the ongoing investigation on strong-coupling quantum thermal machines.

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I. INTRODUCTION

Quantum thermal machines (QTMs) [1–3] perform energetic tasks such as energy conversion and extraction at the nanoscale. With quantum systems as working substances, QTMs are capable of harnessing quantum resources to facilitate the operation process and achieve unparalleled capabilities that are provably impossible with classical counterparts [4–7]. As such, QTMs are becoming potential platforms for demonstrating the quantumness of energetic tasks and contrasting classical and quantum thermodynamics, as highlighted by recent intriguing theoretical proposals [4,5,7–18] and delicate experimental realizations [6,19–30] to name just a few.

Concerning the thermodynamics of QTMs, it is recognized that the description of QTMs does not permit a complete adoption of the well-established classical framework as QTMs can display distinct features beyond the capability of the classical description. One feature that attracts recent attention asserts that working substances of QTMs can experience possible strong system-bath couplings as their surface area can become comparable to their volume [6,15,19,21,23,24,27,31], rendering the weak-coupling assumption inapplicable. Investigating strong-coupling QTMs becomes urgent. On the one hand, the field of strong-coupling quantum thermodynamics has witnessed significant progress over the decades with a number of self-consistent strategies formulated (see a recent review [32] and references therein). Nevertheless, no consensus on a universal framework for strong-coupling quantum thermodynamics has been reached as one lacks prior knowledge of thermodynamic behavior of systems at strong couplings. Applying the existing strong-coupling quantum thermodynamic frameworks to QTMs, one can reveal their own thermodynamic signatures of strong coupling that allow for verification with current experimental capabilities, thereby providing benchmarks for establishing a universal framework for strong-coupling quantum thermodynamics. On the other hand, strong coupling can enable non-negligible system-bath correlation and entanglement, which are useful operational resources for QTMs. In this regard, a recent study [33] suggests that the strong coupling is inevitable when devising steady-state entanglement QTMs.

To date, substantial efforts have been put into the investigation of strong-coupling QTMs [5,11,13,18,34–45]. To circumvent the theoretical and numerical challenges imposed by strong couplings, one typically focuses on either specific models and methodologies [5,11,34,36,37,39,42,43] or specific thermodynamic settings [13,18,40,41,44,45], yielding somewhat contradictory conclusions on the role of strong couplings. Whereas some studies claim that strong coupling could bring up operational advantages to potentially enhance the performance of QTMs [5,18,33,37,38,41,44,45],

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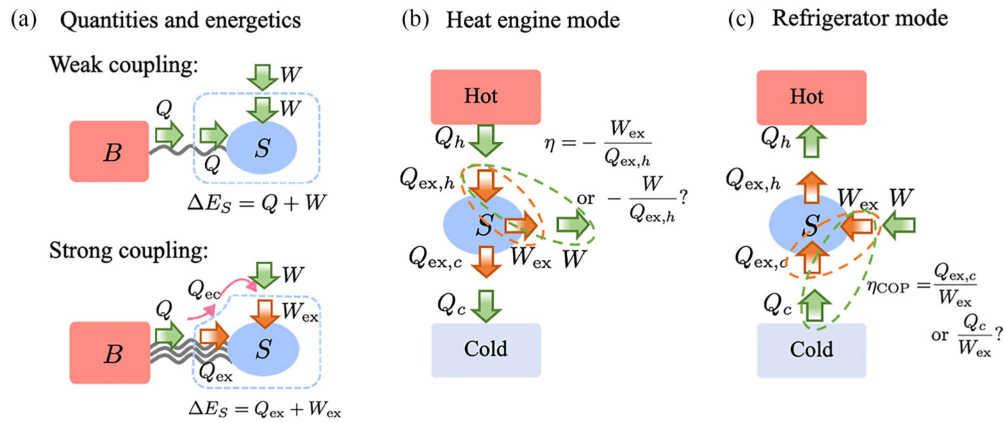

FIG. 1. Sketch of the study. (a) Quantities and energetics during a quasistatic isothermal process. Upper panel: Weak coupling limit with conventional heat definition Q due to bath energy loss and conventional work definition W due to external drivings which are not shown in the figure. The first law of thermodynamics for the system reads $\Delta E_S = Q + W$ [see Eq. (3)]. Lower panel: Strong coupling scenario with both conventional quantities Q, W and excess quantities $Q_{\rm ex} = Q - Q_{\rm ec}$ [see Eq. (4)], $W_{\rm ex} = W + Q_{\rm ec}$ [see Eq. (5)] with $Q_{\rm ec}$ an energetic cost for maintaining the non-Gibbsian thermal state. In this scenario, the first law of thermodynamics for the system reads $\Delta E_S = Q_{\rm ex} + W_{\rm ex}$ [Eq. (5)]. Directions of arrows reflect our positive sign convention of thermodynamic quantities. (b) Energy flow representation of a heat engine mode at strong couplings. Due to the difference between W and $W_{\rm ex}$, one may come up with different definitions for thermodynamic efficiency η which differ in gain as highlighted by the orange and green dashed circles (see details in Sec. IV). (c) Energy flow representation of a refrigerator mode at strong couplings. Due to the difference between Q_c and $Q_{{\rm ex},c}$, one may carry out different definitions for the coefficient of performance $\eta_{\rm COP}$ which differ in gain as highlighted by the orange and green dashed circles (see details in Sec. V).

there are opposite perspectives indicating that strong coupling is merely detrimental to the operation of QTMs [11,13,34,36,39,40,42,43]. Thus, elucidating the exact role of strong coupling in the performance of QTMs still warrants further investigation. Here we focus on QTMs operating with a quasistatic Carnot cycle and analyze the strong-coupling effect on their maximum efficiency.

Noting that the thermal equilibrium state can deviate from a canonical Gibbsian form at strong couplings [46,47], we utilize the excess heat and work definitions proposed in Ref. [35] which naturally account for such a scenario through an exact formulation of the first law of thermodynamics for the working substances at strong couplings. Remarkably, these excess definitions ensure thermodynamic relations between quantities to satisfy the same forms across the weak to strong couplings in the quasistatic limit, thereby providing a promising strategy to lift the ambiguity in defining heat and work at strong couplings. Compared with conventional heat and work definitions based on the change of system density matrix and Hamiltonian, respectively, the excess counterparts differ by an energetic cost term indispensable for maintaining the non-Gibbsian characteristics.

We note that excess work and heat sufficiently determine the energetics of the working substances, whereas the conventional ones, when applying at strong coupling, are instead associated with external work and heat sources [see Fig. 1(a) for an illustration]. With this distinction, we point out that there is a flexibility in defining a thermodynamic figure of merit for QTMs at strong couplings, depending on how one defines the gain of applying the QTMs in that limit. For clarity, here we just consider the efficiency [coefficient of performance (COP)] as the corresponding figure of merit for the heat engine (refrigerator) mode.

Specifically, we propose that one can define two thermodynamically allowed definitions for the chosen figure of merit at strong couplings. The first definition involves just the excess quantities in an analogy with the weak-coupling scenario where one utilizes quantities in the first law of thermodynamics to define the figure of merit. We dub it an "excess" definition for later convenience. The excess definition treats the excess work (excess heat from the cold bath) as the gain of applying a heat engine (refrigerator), thereby evaluating the performance from solely the system energetics in a way similar to the weak-coupling scenario; see the orange dashed circles in Figs. 1(b) and 1(c). Alternatively, one can recall the design purpose of QTMs and evaluate their performance from the standpoint of the external systems upon which the QTMs act. In this spirit, one should take the conventionally defined work (conventionally defined heat out of the cold bath) as the gain for a heat engine (refrigerator), thereby yielding a "hybrid" definition which involves both excess and conventionally defined quantities; see green dashed circles in Figs. 1(b) and 1(c).

These two definitions for the figure of merit coincide only at weak couplings where the energetic cost for maintaining the non-Gibbsian feature vanishes and both reduce to the Carnot limit as expected in the quasistatic limit. However, a distinction between the two definitions emerges at strong couplings. We demonstrate that the excess definition still recovers the Carnot limit for both quasistatic quantum heat engine and quantum refrigerator modes at strong couplings, whereas the hybrid definition leads one to a conclusion that strong coupling plays a negative role as it suppresses the maximum efficiency, rendering the Carnot limit unattainable at strong couplings. Therefore, our results indicate that an uncertainty in the definition for the figure of merit would hinder a thorough evaluation of the strong-coupling effect on the optimal performance. Nevertheless, focusing on the quasistatic limit, we suggest that the hybrid definition is more preferable than the excess one in practical applications as it takes into account the overall effect of QTMs exerted on external systems and thus closely reflects the designed purpose of QTMs.

This paper is organized as follows. For the sake of completeness, we first recap excess heat and work definitions [35] in Sec. II by considering an arbitrary quasistatic isothermal process during which the system can strongly couple to the thermal bath. In Sec. III we present details of the adopted quantum Carnot cycle and expressions for excess quantities using relations carried out in Sec. II. Then in Secs. IV and V we analyze the heat engine mode and refrigerator mode, respectively. We explicitly propose two different yet thermodynamically allowed definitions for the figure of merit of both operation modes. We further demonstrate the strong-coupling effect on optimal performance using both definitions. Finally, we present a few remarks and conclude the study in Sec. VI.

II. EXCESS HEAT AND WORK AT STRONG COUPLINGS

To analyze thermodynamic performance of QTMs, one should first settle the definitions for heat and work. At weak couplings, one usually starts from the changes of system energy and identify the part associated with an entropic change as the heat [48,49] and attribute the remaining part to the work. However, definitions for heat and work are subtle at strong couplings, and no consensus on their definitions has been reached [32]. Here we adopt the framework developed in Ref. [35], which guarantees the applicability of the aforementioned thermodynamic interpretations of heat and work [48,49] at strong couplings. For the sake of completeness and to fix the notation, we will briefly recap the definitions for the excess quantities defined in Ref. [35] with a particular emphasis on clarifying their physical meanings, which will be the basis for proposing and constrasting different definitions for the figure of merit at strong couplings.

Consider a *quasistatic* isothermal process in which a quantum system coupled to a single thermal bath at a temperature $T = \beta^{-1}$ (setting $\hbar = 1$ and $k_B = 1$ hereafter) can stay in an instantaneous thermal equilibrium state. When the system-bath coupling strength deviates from the weak-coupling regime, the system equilibrium state ρ_S is no longer of a Gibbsian form $\rho_{th} = e^{-\beta H_S}/Z_S$ with the system Hamiltonian H_S and $Z_S = \mathrm{Tr}[e^{-\beta H_S}]$ [46,47],

$$\rho_S \;=\; \mathrm{Tr}_B\left[\frac{e^{-\beta H_{\rm tot}}}{Z_{\rm tot}}\right] \;\neq\; \rho_{th}. \tag{1}$$

Here $H_{\rm tot}$ denotes the total Hamiltonian including the system, the bath (B), and the system-bath interaction, $Z_{\rm tot} = \mathrm{Tr}_{\rm tot}[e^{-\beta H_{\rm tot}}]$, and Tr_B denotes a trace operation over bath degrees of freedom.

We consider the von Neumann entropy of the system state $S = -\mathrm{Tr}[\rho_S \ln \rho_S]$ which can become a thermodynamic entropy when one has perfect knowledge of the system [50]. The system von Neumann entropy S satisfies [35,51]

$$S \;=\; \beta(E_S - \mathcal{F}). \tag{2}$$

Here $E_S = \mathrm{Tr}[H_S\rho_S]$ denotes the internal energy of the system, $\mathcal{F} = F + TD(\rho_S||\rho_{th})$ is the nonequilibrium free energy [51–53] with $F = -T\ln Z_S$ the Helmholtz free energy, and $D(\rho_S||\rho_{th}) = \mathrm{Tr}[\rho_S(\ln\rho_S - \ln\rho_{th})]$ the quantum relative entropy between ρ_S and ρ_{th} accounting for the non-Gibbsian nature of ρ_S at strong couplings. We remark that in the quantum domain the strong-coupling equilibrium state ρ_S can contain nonzero coherence in the energy basis of the system Hamiltonian [47]; therefore the quantum relative entropy involved in the nonequilibrium free energy definition cannot reduce to the Kullback-Leibler divergence between classical distributions. Nevertheless, the same form of the first law holds as well in classical systems described in terms of distributions at strong couplings. Taking S as the thermodynamic entropy [50], we can then regard $\mathcal{F}$ as a thermodynamic free energy at strong couplings. It is evident that $\mathcal{F} \geqslant F$ with the equality attained at weak couplings where $\rho_S = \rho_{th}$, implying an additional capacity to perform work using information of relative entropy [53] at strong couplings. We remark that Eq. (2) defines an exact form of the first law of thermodynamics for isothermal processes under an arbitrary coupling strength.

To define heat and work applicable at strong couplings, one can start from considering infinitesimal changes of thermodynamic quantities at hand. We first have

$$\delta E_S = \mathrm{Tr}[\delta\rho_S H_S] + \mathrm{Tr}[\rho_S\delta H_S] \equiv \delta Q + \delta W. \tag{3}$$

Here $\delta W \equiv \mathrm{Tr}[\rho_S\delta H_S]$ represents the work generated by external driving fields in the system Hamiltonian, and $\delta Q \equiv \mathrm{Tr}[\delta\rho_S H_S]$ represents the heat exchanged with the thermal bath [35]. To see this, we denote the total Hamiltonian $H_{\rm tot} = H_B + V + H_S$ with H_B and V the bath Hamiltonian and system-bath interaction, respectively. We first have $\delta W = \delta\mathrm{Tr}[H_{\rm tot}\rho] = \mathrm{Tr}[\rho\delta H_S]$ with ρ the total density matrix by noting $\mathrm{Tr}[H_{\rm tot}\delta\rho] = 0$ as a result of the quantum Liouville equation for ρ. We then find $\delta\mathrm{Tr}[H_{\rm tot}\rho] = \delta\mathrm{Tr}[H_B\rho] + \delta\mathrm{Tr}[H_S\rho] + \delta\mathrm{Tr}[V\rho] \simeq \delta\mathrm{Tr}[H_B\rho_B] + \delta E_S = \delta W$ with $\delta\mathrm{Tr}[V\rho]$ being negligible for a constant coupling in the quasistatic limit and ρ_B the reduced bath state. Comparing it with Eq. (3), we can find $\delta Q = -\delta\mathrm{Tr}[H_B\rho_B]$. One recalls that the usual semiclassical modelings of cyclic QTMs do not explicitly include pure work sources and loads in the Hamiltonian but assume their presence through external driving fields in the system Hamiltonian [54]. In this sense, we emphasize that W defines the amount of work that can be harnessed by external work loads even at strong couplings. Hereafter, we set the sign convention that positive heat and work increase the internal energy of the system.

At weak couplings, Q and W unambiguously define the heat and work, respectively, and quantify the energetics of the working substance according to Eq. (3) [see the upper panel of Fig. 1(a)]. In a quasistatic isothermal process, we have $\rho_S = \rho_{th}$ diagonal in the energy basis, hence the heat Q induces a pure entropic change. However, this is no longer the case at strong couplings as we will show below. Nevertheless, we remark that one can still use Q and W at strong couplings as they are still well-defined thermodynamic quantities in that regime [see the lower panel of Fig. 1(a)].

Inserting Eq. (3) into Eq. (2), we find

$$\delta S \;=\; \beta(\delta Q - \delta Q_{\rm ec}) \;\equiv\; \beta\delta Q_{\rm ex}. \tag{4}$$

Here $\delta Q_{\rm ec} \equiv \delta\mathcal{F} - \delta W$ denotes an energetic cost for maintaining the non-Gibbsian characteristics [35] and corresponds to an energy part of δQ that does not induce an entropic change. $\delta Q_{\rm ex} \equiv \delta Q - \delta Q_{\rm ec}$ thus defines an excess heat accompanying with a pure entropic change, in accordance with the definition for a thermodynamic heat [48,49]. Hence, we take $Q_{\rm ex}$ as the appropriate heat definition for the working substance at strong couplings in the quasistatic limit. Notably, $Q_{\rm ex}$ reduces to Q at weak couplings as $Q_{\rm ec}$ vanishes in that limit.

With the definition of excess heat, we can rewrite Eq. (3) as

$$\delta E_S \;=\; \delta W_{\rm ex} + \delta Q_{\rm ex}. \tag{5}$$

Here the excess work $\delta W_{\rm ex} \equiv \delta W + \delta Q_{\rm ec} = \delta \mathcal{F}$, reducing to δW at weak couplings, precisely corresponds to the nonequilibrium free energy change, in accordance with the conventional expectation for the work in the quasistatic limit.

From the first law of thermodynamics, Eq. (2), one generally finds $\Delta\mathcal{F}(t) = \Delta E_S(t) - T\Delta S(t)$ with $\Delta A(t) = A(t) - A(0)$ for an arbitrary quantity A. Meanwhile, we have $\Delta E_S(t) = Q_{\rm ex}(t) + W_{\rm ex}(t)$ in terms of excess quantities, which, together with the second law $T\Delta S(t) \geqslant Q_{\rm ex}(t)$, implies $\Delta E_S(t) \leqslant T\Delta S(t) + W_{\rm ex}(t)$. Altogether, we arrive at a principle of maximum work applicable at strong couplings $-\Delta\mathcal{F}(t) \geqslant -W_{\rm ex}(t)$ with the equality taken in the quasistatic limit. Hence, one can regard $-W_{\rm ex}$ as the maximum work that the working substance can provide during a quasistatic isothermal process according to our sign convention. As such, we take $W_{\rm ex}$ as the work definition for the working substance at strong couplings which together with the excess heat $Q_{\rm ex}$ completely determines the energetics of the working substance [see the lower panel of Fig. 1(a) for an illustration]. However, as we remarked before, only the part W of $W_{\rm ex}$ can be harnessed by external systems. Noting $Q_{\rm ec} = W_{\rm ex} - W$, one can also interpret $Q_{\rm ec}$ as the amount of work that cannot be harnessed by external systems.

III. QUANTUM CARNOT CYCLE

To operate QTMs, we consider a *quasistatic* Carnot cycle consisting of two quasistatic isothermal strokes and two quasistatic adiabatic strokes. Taking the heat engine mode as an example, we have four strokes following the given order.

(i) A quasistatic hot isothermal expansion stroke. During this stroke, a quantum working substance is attaching to a hot thermal bath at temperature T_h with the system Hamiltonian changing from H_S^A to H_S^B; We use superscripts to distinguish quantities at different stages induced by, e.g., tuning control parameters. To realize expansion of quantum working substances with discrete energy levels $\{E_n\}$, one needs to change all energy levels $E_n^A \to E_n^B = \xi E_n^A$ with the same ratio $0 < \xi < 1$ [8]. During this stroke, we can use definitions in the previous section to express excess work and heat in terms of the nonequilibrium free energy and entropy changes, respectively,

$$W_{{\rm ex},h} = \mathcal{F}^B - \mathcal{F}^A, \quad Q_{{\rm ex},h} = T_h(S^B - S^A). \tag{6}$$

We also have a formal definition $W_h \equiv \int_A^B {\rm Tr}[\rho_S(t)\frac{d}{dt}H_S(t)]\,dt$ which denotes the work generated by driving fields during this stroke. In practice, once $H_S(t)$ and the governing equation of motion for $\rho_S(t)$ are specified during an isothermal process, we can estimate the nonequilibrium free energy and entropy changes directly, thereby obtaining the values of the excess work and heat, respectively. W_h can also be estimated using its formal definition, which, together with the relation $Q_{{\rm ec},h} = W_{{\rm ex},h} - W_h$ and the value of $W_{{\rm ex},h}$, further gives an evaluation of the energetic cost. We take a two-level system with a time-dependent Hamiltonian $H_S(t) = \frac{\Omega(t)}{2}\sigma_z$, for example, to illustrate how to implement the calculation. To realize an expansion stroke, one should first specify a detailed driving protocol $\Omega(t)$ such that $\Omega^B < \Omega^A$ while ensuring the quasistatic requirement. Then one utilizes numerical methodologies such as the one in [55] to simulate the evolution dynamics of $\rho_S(t)$ at strong couplings. With $\rho_S(t)$ and $H_S(t)$, one can then calculate W_h according to the aforementioned definition. The same dynamical information also allows us to directly compute the entropy S and the nonequilibrium free energy $\mathcal{F}$, and consequently, the values of $W_{{\rm ex},h}$ and $Q_{{\rm ex},h}$ based on the definitions in Eq. (6). The energetic cost $Q_{{\rm ec},h}$ is obtained as the difference between $W_{{\rm ex},h}$ and W_h.

(ii) A quasistatic adiabatic expansion stroke. During this stroke, the working substance is detached from the hot thermal bath, rendering heat exchanges impossible and leaving the system entropy unchanged, $S^C = S^B$. The temperature of the working substance drops from T_h to T_c accomplished by an internal energy change which comes in the form of work. By requiring that all energy gaps of the working substance change by the same ratio T_c/T_h, one can ensure that the end points of the adiabatic expansion stroke coincide with those of an isothermal processes [8]. Hence we can still adopt the first law of thermodynamics given in the previous section to rewrite the work,

$$W_{{\rm ex},a1} = W_{a1} \;=\; E_S^C - E_S^B = \mathcal{F}^C - \mathcal{F}^B - (T_h - T_c)S^B. \tag{7}$$

To simulate this stroke with the aforementioned two-level system, one needs to choose a driving protocol $\Omega(t)$ such that $\Omega^C = (T_c/T_h)\Omega^B$. The evolution of $\rho_S(t)$ is now governed by the quantum Liouville equation $d\rho_S(t)/dt = -i[H_S(t), \rho_S(t)]$ with initial condition ρ_S^B. With $\rho_S(t)$ and $H_S(t)$, one can calculate the entropy S and the nonequilibrium free energy $\mathcal{F}$ with their changes during this stroke to determine the value of $W_{{\rm ex},a1}$.

(iii) A quasistatic cold isothermal compression stroke. During this stroke, the working substance is attaching to a cold thermal bath at temperature T_c with all energy levels increasing $E_n^C \to E_n^D = \xi' E_n^C$ by the same ratio $\xi' > 1$. For a two-level system, the compression is thus realized by increasing the energy gap. We have

$$W_{{\rm ex},c} = \mathcal{F}^D - \mathcal{F}^C, \quad Q_{{\rm ex},c} = T_c[S^D - S^B], \tag{8}$$

and similarly, $W_c \equiv \int_C^D {\rm Tr}[\rho_S(t)\frac{d}{dt}H_S(t)]\,dt$. Similar to the hot isothermal stroke, one can calculate the excess quantities together with the energetic cost for the two-level system by just devising a quasistatic driving protocol with the requirement $\Omega^D > \Omega^C$.

(iv) A quasistatic adiabatic compression stroke. During this stroke, the system is detached from the cold thermal bath, and the temperature of the working substance increases from T_c to T_h along with all energy gaps of the working substance increasing by the same ratio T_h/T_c. Similar to the second

adiabatic stroke, we find

$$W_{ex,a2} = W_{a2} = E_S^A - E_S^D = \mathcal{F}^A - \mathcal{F}^D + (T_h - T_c)S^A. \quad (9)$$

Here we have used the fact that $S^D = S^A$. For the two-level system, one requires a driving protocol with $\Omega^A = (T_h/T_c)\Omega^D$ and then follows the procedures in the adiabatic expansion stroke to calculate the corresponding thermodynamic quantities. After this stroke, we couple the working substance to the hot thermal bath again, thus completing the thermodynamic cycle. The Carnot cycle for the refrigerator mode can be obtained by reversing the aforementioned orderings of strokes as well as each stroke's operational direction, and the associated thermodynamic quantities can be expressed in an analogous way.

In the following section we will distinguish between two possible sets of definitions for the figure of merit of quantum Carnot thermal machines [see Figs. 1(b) and 1(c)]. The first definition, which we dub an *excess* one, involves just excess quantities, amounting to characterizing the performance from solely the system energetics in complete analogy with the weak-coupling scenario. The second definition instead evaluates the performance from the action on an external system and inevitably contains both the conventional and excess quantities, thereby bearing the name of a *hybrid* definition.

IV. QUANTUM CARNOT HEAT ENGINE

We first focus on the cyclic quantum Carnot heat engine operating with the Carnot cycle stated in Sec. III. At weak couplings, one utilizes Q_h and total work $W \equiv \sum_{i=h,a1,c,a2} W_i$ to define the efficiency, which, however, cannot be directly applied to the strong-coupling regime [18,35]. Hence, to unveil the strong-coupling effect on the efficiency, one needs to decipher an appropriate definition for the efficiency first. In this regard, one should bear in mind that a thermodynamic efficiency quantifies how worth a gain at the expense of a cost is.

For a heat engine operated at strong couplings, the actual cost should be the excess heat Q_{ex}^h during the hot isothermal process that the working substance absorbs. As for the gain, we point out that one can have two possible choices at strong couplings due to the distinction between $W_{ex} \equiv \sum_{i=h,a1,c,a2} W_{ex,i}$ and W.

Through introducing excess heat and work that have the same interpretations as their weak-coupling counterparts, one actually builds up complete analog between weak and strong couplings in terms of the first law of thermodynamics [Eq. (3) for weak coupling and Eq. (5) for strong coupling extended to a two-bath scenario]. At weak couplings, one can just utilize W and Q_h involved in the first law of thermodynamics to define a thermodynamic efficiency which recovers the Carnot bound in the quasistatic limit. Following this spirit and noting the analog, one can define a thermodynamic efficiency using just excess quantities at strong couplings

$$\eta^{ex} \equiv -\frac{W_{ex}}{Q_{ex,h}}. \quad (10)$$

This definition amounts to evaluating the performance of heat engines from solely the energetics of the working substance and treating W_{ex} as the gain in analogy with the weak-coupling scenario. We dub it an *excess* definition. Using results in Sec. III, one identifies

$$W_{ex} = (T_h - T_c)[S^A - S^B]. \quad (11)$$

Inserting Eq. (11) into Eq. (10), one finds that the inside efficiency definition exactly coincides with the Carnot efficiency

$$\eta^{ex} = \frac{T_h - T_c}{T_h} = \eta_c. \quad (12)$$

Hence, one concludes that the strong couplings have no impact on the maximum efficiency when defining the efficiency of a heat engine from a complete analogy with the weak-coupling scenario. This result was first obtained in Ref. [35] and was stated as a manifestation of the thermodynamic universality of the Carnot efficiency.

Alternatively, as we remarked before, only the part W can be directly harnessed by external systems as it is associated with external driving fields. Hence, if one evaluates the performance of a quantum Carnot heat engine from the standpoint of external systems, one then naturally refers to W as the gain of the machine since a part of the excess work manifests as an energetic cost which cannot be harnessed by external systems. This viewpoint brings up a thermodynamic efficiency definition

$$\eta^{hyb} \equiv -\frac{W}{Q_{ex,h}} = \eta_c + \frac{\sum_{i=h,c} Q_{ec,i}}{Q_{ex,h}}. \quad (13)$$

In arriving at the second equality, we have used the relation $W = W_{ex} - \sum_{i=h,c} Q_{ec,i}$ and Eq. (12). As this efficiency definition involves both conventional and excess quantities, we refer to it as a *hybrid* definition. It is evident that η^{hyb} and η^{ex} are identical only at weak couplings where $Q_{ec,i}$ vanishes. At strong couplings, the two definitions η^{ex} and η^{hyb} are no longer equivalent. We note that the second term on the right-hand side of Eq. (13) arises when $Q_{ec,i}$ becomes nonzero. Recalling that $Q_{ec,i}$ quantifies the energetic cost for maintaining the non-Gibbsian state during the isothermal strokes, we can attribute the second term to the effect of strong coupling.

To ensure that the hybrid efficiency η^{hyb} is thermodynamically allowed, we need to check whether it can surpass the Carnot efficiency due to the presence of an extra term. To this end, we can analyze the sign of this extra term, which is solely determined by the numerator $\sum_{i=h,c} Q_{ec,i}$ as the denominator $Q_{ex,h}$ is always positive in a heat engine. We first note that $-W_{ex} > 0$ and $-W > 0$ in a heat engine according to our sign convention. With the principle of maximum work, we know that $-W_{ex}$ corresponds to the maximum work that the engine can generate as it is just the nonequilbrium free energy change during a cycle, while $-W$ is the amount of the work that can be harnessed by an external work load eventually. It is reasonable to expect that the external work load cannot gain more work than the engine can provide, hence one should have $|W_{ex}| \geqslant |W|$ or equivalently, $-W_{ex} \geqslant -W$. From this inequality, we can infer that $-\sum_{i=h,c} Q_{ec,i} = W - W_{ex} \geqslant 0$ with the equality attained at weak couplings. Hence the second term on the right-hand side of Eq. (13) is always nonpositive, leading to a constraint on the hybrid efficiency definition η^{hyb},

$$\eta^{hyb} \leqslant \eta_c. \quad (14)$$

Here the equality is met at weak couplings where $\sum_i Q_{\mathrm{ec},i} = 0$. At strong couplings with $\sum_i Q_{\mathrm{ec},i} < 0$, one generally expects $\eta^{\mathrm{hyb}} < \eta_c$, rendering η^{hyb} a valid thermodynamic efficiency consistent with the second law of thermodynamics. More importantly, adopting η^{hyb} as the efficiency definition would lead one to conclude that the strong coupling has a negative effect in the sense that it prevents the maximum efficiency of the strong-coupling heat engine from reaching the Carnot efficiency, unlike their weak-coupling counterparts.

V. QUANTUM CARNOT REFRIGERATOR

We now turn to a cyclic quantum Carnot refrigerator operating with a reversed Carnot cycle. The corresponding expressions for the excess work and heat are obtained by reversing the sign of those listed in Sec. III.

To define a coefficient of performance (COP) for the refrigerator, one similarly needs to specify the cost and the resulting gain. Here the cost should be the excess work W_{ex}, which directly induces the free energy change of the working substance in the quasistatic limit. As for the gain, one also has two different choices, similar to the heat engine mode.

Here one still has an analogy between the weak and strong-coupling scenarios after introducing excess thermodynamic quantities. Inspired by the COP definition Q_c/W at weak couplings using just quantities involved in the first law of thermodynamics, one can also evaluate the performance of the refrigerator mode using just the excess quantities describing the energetics of the working substance at strong coupling,

$$\eta_{\mathrm{COP}}^{\mathrm{ex}} \equiv \frac{Q_{\mathrm{ex},c}}{W_{\mathrm{ex}}}. \tag{15}$$

Here the excess heat $Q_{\mathrm{ex},c}$ during the cold isothermal stroke is identified to be the gain of the refrigerator mode by noting the analogy with the weak coupling scenario. We dub it an *excess* COP definition. For the reversed Carnot cycle, one has $Q_{\mathrm{ex},c} = T_c[S^B - S^A]$ and $W_{\mathrm{ex}} = (T_h - T_c)[S^B - S^A]$, which are opposite in sign to those shown in Sec. III, yielding

$$\eta_{\mathrm{COP}}^{\mathrm{ex}} = \eta_{\mathrm{COP}}^{c}. \tag{16}$$

Here $\eta_{\mathrm{COP}}^{c} \equiv T_c/(T_h - T_c)$ is just the classical Carnot limit of the COP. Namely, the optimal COP of the refrigerator at strong couplings can reach the Carnot limit just as was the case for the weak-coupling scenario if adopting the excess definition for the COP, thereby implying no strong-coupling effect on the optimal performance.

Nevertheless, during the cold isothermal expansion stroke, one recognizes that the total amount of energy extracted out of the cold bath should be Q_c instead of $Q_{\mathrm{ex},c}$, which induces a system entropic change. Recall that the design purpose of a quantum absorption refrigerator is to extract energy out of a cold bath, and one should instead treat Q_c as the gain of the refrigerator. Therefore, one can have an alternative COP definition

$$\eta_{\mathrm{COP}}^{\mathrm{hyb}} \equiv \frac{Q_c}{W_{\mathrm{ex}}} = \eta_{\mathrm{COP}}^{c} + \frac{Q_{\mathrm{ec},c}}{W_{\mathrm{ex}}}. \tag{17}$$

In arriving at the second equality, we have used Eq. (16) and the relation $Q_c = Q_{\mathrm{ex},c} + Q_{\mathrm{ec},c}$. Similar to the heat engine mode, we refer to $\eta_{\mathrm{COP}}^{\mathrm{hyb}}$ as a *hybrid* COP definition. We note that $\eta_{\mathrm{COP}}^{\mathrm{ex}}$ and $\eta_{\mathrm{COP}}^{\mathrm{hyb}}$ become equivalent only at weak couplings where the energetic cost $Q_{\mathrm{ec},c}$ vanishes. At strong couplings, these two COP definitions $\eta_{\mathrm{COP}}^{\mathrm{ex}} \neq \eta_{\mathrm{COP}}^{\mathrm{hyb}}$ due to a nonzero energetic cost $Q_{\mathrm{ec},c}$ for maintaining the non-Gibbsian state during the cold isothermal stroke. We thus take the presence of the second term on the right-hand side of Eq. (17) as a sign of strong coupling in the quasistatic limit.

With the presence of an extra term on top of η_{COP}^{c}, examining whether the hybrid COP definition $\eta_{\mathrm{COP}}^{\mathrm{hyb}}$ is consistent with the second law of thermodynamic becomes necessary. We note that the denominator W_{ex} of the extra term in Eq. (17) is positive in a refrigerator, hence the sign of the extra term is fully determined by the numerator $Q_{\mathrm{ec},c}$. To analyze the sign of $Q_{\mathrm{ec},c}$, let us focus on the cold isothermal expansion stroke of the reversed Carnot cycle. During this stroke, the working substance generates work output, implying $W_{\mathrm{ex},c} < 0$ and $W_c < 0$. Since $|W_{\mathrm{ex},c}|$ defines the maximum work, we should have $|W_{\mathrm{ex},c}| \geqslant |W_c|$, or equivalently, $W_{\mathrm{ex},c} \leqslant W_c$. We thus find $Q_{\mathrm{ec},c} = W_{\mathrm{ex},c} - W_c \leqslant 0$. Hence, the hybrid COP definition is upper bounded by the Carnot COP, rendering it a valid COP definition,

$$\eta_{\mathrm{COP}}^{\mathrm{hyb}} \leqslant \eta_{\mathrm{COP}}^{c}. \tag{18}$$

Here the equality is attained only at weak couplings where $Q_{\mathrm{ec},c} = 0$. At strong couplings, we generally expect $\eta_{\mathrm{COP}}^{\mathrm{hyb}} < \eta_{\mathrm{COP}}^{c}$. Therefore, similar to the heat engine mode, the hybrid COP definition also unveils a negative effect of strong coupling by noting that only a weak-coupling setup can reach the Carnot COP in the quasistatic limit.

VI. DISCUSSION AND CONCLUSION

We remark that both the excess and hybrid definitions for the figure of merit are thermodynamically valid, and one cannot definitively exclude one definition from the two based on pure thermodynamic principles at strong couplings. Nevertheless, we recommend that the hybrid definition for the figure of merit should be used in actual applications of quantum thermal machines as it faithfully recognizes the design capability of quantum thermal machines, that is, exerting a thermodynamic useful action on the targeted external systems.

We also emphasize that the present results are specifically obtained in the quasistatic limit. Turning to the finite-time operation regime where the efficiency at maximum power is of special interest [56], we note that non-Gibbsian states can arise even at weak couplings due to the finite-time driving fields during isothermal strokes. Hence, we cannot simply associate strong-coupling effects with the emergence of the non-Gibbsian character of the system state in finite time cycles. We further remark that one can still define a nonequilibrium free energy to account for an arbitrary finite time isothermal process; however, additional dissipation induced by finite-time drivings prevents us from identifying excess heat and work from the entropy and nonequilibrium free energy changes, respectively. Extending the present framework to the finite-time regime and addressing strong-coupling effects on the efficiency at maximum power therein remains an open question that warrants further investigation.

Considering that the Gibbs state is completely passive, one may naturally expect that a non-Gibbsian thermal equilibrium state has a finite ergotropy [57] that allows for work extraction. To see this is indeed the case, we adopt the total ergotropy definition describing scenarios with many identical system copies, $\mathcal{E}_{\rm tot}=E_S-E_S^R$ [58]. Here $E_S^R={\rm Tr}[H_S\rho_R]$ is the mean system energy evaluated with respect to a Gibbsian reference state $\rho_R=e^{-H_S/T_R}/Z_R$ that has the same von Neumann entropy S of ρ_S. Since $E_S^R=F_R+T_RS$ with $F=-T_R\ln Z_R$ and $E_S=\mathcal{F}+T_RS$, we can express the total ergotropy as $\mathcal{E}_{\rm tot}=\mathcal{F}-F_R$, which vanishes only when ρ_S reduces to a Gibbsian form. In the quasistatic limit, one can further replace the changes of $\mathcal{F}$ and S with the excess work and heat, respectively, thereby obtaining a relation between total ergotropy change and excess thermodynamic quantities.

Furthermore, it is well known in classical thermodynamics that a quasistatic cycle implies a vanishing total entropy production, which in turn, combined with the usual first law of thermodynamics, leads to the celebrated Carnot theorem. With this result in mind, it seems that our outside view that strong coupling can suppress the maximum efficiency is incompatible with the quasistatic cycle we adopted. To clarify, we first note that delivering a definition for the total entropy production over a cycle at strong couplings faces an ambiguity as well. Following the excess definition, it is evident that the total entropy production over a cycle should read $\Sigma^{\rm ex}=-Q_{{\rm ex},h}/T_h-Q_{{\rm ex},c}/T_c$. Then the result $\eta^{\rm ex}=\eta_c$ for the heat engine follows from the combination of conditions $\eta^{\rm ex}=-W_{\rm ex}/Q_{{\rm ex},h}$, $W_{\rm ex}+Q_{{\rm ex},h}+Q_{{\rm ex},c}=0$ and $\Sigma^{\rm ex}=0$. However, inspired by the hybrid definition, one can argue that the total entropy production over a cycle should be defined as $\Sigma^{\rm hyb}=-Q_h/T_h-Q_c/T_c$ which quantifies the entropy change associated with the heat out of the baths. Taking the heat engine mode as an example, we have $\eta^{\rm hyb}=-W/Q_{{\rm ex},h}=(Q_h+Q_c)/(Q_h-Q_{{\rm ec},h})$ with the equality $W+Q_h+Q_c=0$ [cf. Eq. (3)]. Adapting the rationale that leads to Eq. (18), we can infer $Q_h-Q_{{\rm ec},h}\geqslant Q_h$ such that $\eta^{\rm hyb}\leqslant(Q_h+Q_c)/Q_h=\eta_c$ with the last equality obtained using the quasistatic condition $\Sigma^{\rm hyb}=0$. Hence, our result is fully compatible with the quasistatic condition.

In conclusion, we addressed strong-coupling quantum Carnot thermal machines in the quasistatic limit with a pure analytical treatment, with the consideration that it is challenging to numerically implement a proper quasistatic thermodynamic cycle. We revealed that the quantum thermal machine allows for multiple definitions for the figure of merit at strong couplings. As a result, one cannot reach conclusive results regarding the strong-coupling effect on maximum efficiency. Specifically, we proposed two possible definitions, dubbed excess and hybrid ones. We have analytically shown that the excess definition which just considers the energetics of the working substance exactly recovers the Carnot limit at strong couplings, thereby implying that there is no strong-coupling effect on the maximum efficiency. In contrast, we demonstrated that the hybrid definition which evaluates the gain from the perspective of the external system upon which the quantum thermal machine acts predicts a suppression effect of the strong coupling, rendering the Carnot limit unattainable at strong couplings. Notably, the latter result extends that in Ref. [43] to regimes beyond the linear response. Our results thus point out that it is necessary to first gauge the definition for the figure of merit before trying to elucidate the strong-coupling effect on the performance. We believe this work highlights an important issue in the field and will lead to further investigations on the nature of strong-coupling quantum thermodynamics and quantum thermal machines.

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