Quantum Heat Current under Non-perturbative and Non-Markovian Conditions: Applications to Heat Machines

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We consider a quantum system strongly coupled to multiple heat baths at different temperatures. Quantum heat transport phenomena in this system are investigated using two definitions of the heat current, one in terms of the system energy, and the other in terms of the bath energy. When we consider correlations among system-bath interactions (CASBI) – which have a purely quantum mechanical origin – the definition in terms of the bath energy becomes different. We found that CASBI are necessary to maintain the consistency of the heat current with thermodynamic laws in the case of strong system-bath coupling. However, within the context of the quantum master equation approach, both of these definitions are identical. Through a numerical investigation, we demonstrate this point for a non-equilibrium spin-boson model and a three-level heat engine model using the reduced hierarchal equations of motion approach under strongly coupled and non-Markovian conditions. We observe cyclic behavior of the heat currents and the work performed by the heat engine, and we find that their phases depend on the system-bath coupling strength. Through consideration of the bath heat current, we show that the efficiency of the heat engine decreases as the strength of the system-bath coupling increases, due to the CASBI contribution. In the case of a large system-bath coupling, the efficiency increases further if the bath temperature is decreased, with fixed the ratio of the bath temperatures, due to the discretized nature of energy eigenstates. This is also considered to be a unique feature of quantum heat engines.

I. INTRODUCTION

Recent progress in the control and measurement of small-scale systems provides the possibility of examining the extension of thermodynamics [1-4] and the foundation of statistical mechanics [5, 6] in nano materials. In particular, elucidating how such purely quantum mechanical phenomena as quantum entanglement and coherence are manifested in thermodynamics has become a topic of growing interest for the past two decades. [7-9] Investigations of these types of phenomena may provide new insights into our world and aid in the development of quantum heat machines operating beyond classical bounds. Such developments could lead to more efficient methods of energy usage.

Quantum heat transport problems involving quantum heat machines have been studied with approaches developed through application of open quantum dynamics theory. Quantum master equation (QME) approaches are frequently used in the literature. We consider correlations among system-bath interactions (CASBI) – which have a purely quantum mechanical origin – the definition in terms of the bath energy becomes different. We found that CASBI are necessary to maintain the consistency of the heat current with thermodynamic laws in the case of strong system-bath coupling. However, within the context of the quantum master equation approach, both of these definitions are identical. Through a numerical investigation, we demonstrate this point for a non-equilibrium spin-boson model and a three-level heat engine model using the reduced hierarchal equations of motion approach under strongly coupled and non-Markovian conditions. We observe cyclic behavior of the heat currents and the work performed by the heat engine, and we find that their phases depend on the system-bath coupling strength. Through consideration of the bath heat current, we show that the efficiency of the heat engine decreases as the strength of the system-bath coupling increases, due to the CASBI contribution. In the case of a large system-bath coupling, the efficiency increases further if the bath temperature is decreased, with fixed the ratio of the bath temperatures, due to the discretized nature of energy eigenstates. This is also considered to be a unique feature of quantum heat engines.

In the present study, we employ the hierarchal equations of motion (HEOM) approach [24, 25] to investigate heat transport and quantum heat engine problems. This approach allows us to treat systems subject to external driving fields in a numerically rigorous manner under non-Markovian and strong system-bath coupling conditions. In order to illustrate this point, here we employ two definitions of the heat current, one in terms of the system energy (system heat current), and the other in terms of the bath energy (bath heat current). Both of these definitions are frequently used in the literature. We carefully examine the definition of the heat current in the case of the strong system-bath coupling, because the existence of a non-commuting inter-bath coupling through the system contributes to the heat current even in the steady state case. This effect has not been investigated in previous studies.
II. SYSTEM HEAT CURRENT AND BATH HEAT CURRENT

We consider a system coupled to multiple heat baths at different temperatures. With $K$ heat baths, the total Hamiltonian is written

$$\hat{H}(t) = \hat{H}_S(t) + \sum_{k=1}^{K} \left( \hat{H}_k^S + \hat{H}_k^B \right),$$

(1)

where $\hat{H}_S(t)$ is the system Hamiltonian, whose explicit time dependence originates from the coupling with the external driving field. The Hamiltonian of the $k$th bath and the Hamiltonian representing the interaction between the system and the $k$th bath are given by $\hat{H}_k^B = \sum_j \hbar \omega_k \hat{b}_k^\dagger \hat{b}_k$ and $\hat{H}_k^S = \hat{V}_k \sum_j g_{kj} (\hat{b}_j + \hat{b}_j^\dagger)$, respectively, where $\hat{V}_k$ is the system operator that describes the coupling to the $k$th bath. Here, $\hat{b}_k, \hat{b}_k^\dagger, \omega_k$ and $g_{kj}$ are the annihilation operator, creation operator, frequency, and coupling strength for the $j$th mode of the $k$th bath, respectively. Due to the bosonic nature of the bath, all bath effects on the system are determined by the noise correlation function, $C_k(t) \equiv \langle \hat{X}_k(t) \hat{X}_k(0) \rangle_B$, where $\hat{X}_k \equiv \sum_j g_{kj} (\hat{b}_j + \hat{b}_j^\dagger)$ is the collective bath coordinate of the $k$th bath and $\langle \ldots \rangle_B$ represents the average taken with respect to the canonical density operator of the baths. The noise correlation function is expressed in terms of the bath spectral density, $J_k(\omega)$, as

$$C_k(t) = \int_0^\infty d\omega \frac{J_k(\omega)}{\pi} \left[ \coth \left( \frac{\hbar \omega}{2 k_B T_k} \right) \cos(\omega t) - i \sin(\omega t) \right],$$

(2)

where $J_k(\omega) \equiv \pi \sum_j g_{kj}^2 \delta(\omega - \omega_k)$, and $T_k$ is the temperature of the $k$th bath.

For the system described above, we derive two rigorous expressions for the heat current, which are convenient for carrying out simulations of reduced system dynamics. One of these expressions is derived through consideration of conservation of the heat current, and for this reason, we call it the "system heat current" (SHC). This current is defined as

$$\frac{d}{dt} \langle \hat{H}_S(t) \rangle - W(t) = \sum_{k=1}^{K} \dot{Q}_S^k(t),$$

(3)

where $W(t) \equiv \langle (\partial \hat{H}_S(t)/\partial t) \rangle$ is the power, i.e., the time derivative of the work, and

$$\dot{Q}_S^k(t) = \frac{i}{\hbar} \langle [\hat{H}_S^k(t), \hat{H}_B^k(t)] \rangle$$

(4)

is the change in the system energy due to the coupling with the $k$th bath. This is identical to the definition used in the QME approach, in which the system Hamiltonian is identified as the internal energy. The second expression for the heat current is derived through consideration of the rate of decrease of the bath energy, $\dot{Q}_B^k(t) = -d\langle \hat{H}_B^k(t) \rangle / dt$. We call this current the "bath heat current" (BHC). Using the Heisenberg equations, the BHC can be rewritten as

$$\dot{Q}_B^k(t) = \dot{Q}_S^k(t) + \frac{d}{dt} \langle \hat{H}_B^k(t) \rangle + \sum_{k' \neq k} \dot{q}_{k,k'},$$

(5)

where

$$\dot{q}_{k,k'}(t) = \frac{i}{\hbar} \langle [\hat{H}_S^k(t), \hat{H}_B^{k'}(t)] \rangle.$$
III. THE FIRST AND SECOND LAWS OF THERMODYNAMICS

We can obtain the first law of thermodynamics by summing Eq. (5) over all $k$:

$$
\sum_{k=1}^{K} Q_k^B(t) = \frac{d}{dt} (\hat{H}_S(t) + \sum_{k=1}^{K} \hat{H}_k^B(t)) - W(t).
$$

(7)

The quantity $\hat{H}_S(t) + \sum_{k=1}^{K} \hat{H}_k^B(t)$ is identified as the internal energy, because the contributions of $\dot{q}_{k,k'}$ cancel out.

The derivation of the second law of thermodynamics is presented in Appendix A. In a steady state without external driving forces, the second law is expressed as

$$
- \sum_{k=1}^{K} \frac{Q_k^B}{T_k} \geq 0,
$$

(8)

while with a periodic external driving force, it is given by

$$
- \sum_{k=1}^{K} \frac{Q_{\text{cyc},k}^B}{T_k} \geq 0,
$$

(9)

where $Q_{\text{cyc},k}^B = \oint dt Q_k^B(t)$ is the heat absorbed or released per cycle. The second law without a driving force can be rewritten in terms of the SHC as

$$
- \sum_{k=1}^{K} \frac{Q_{\text{cyc},k}^S}{T_k} \geq \sum_{k,k'=1}^{K} \frac{\dot{q}_{k,k'}}{T_k}.
$$

(10)

When the right-hand side (rhs) of Eq. (10) is negative, the left-hand side (lhs) can also take negative values. However, this contradicts the Clausius statement of the second law, i.e., that heat never flows spontaneously from a cold body to a hot body. As we show in Sec. IV it is necessary to include the $\dot{q}_{k,k'}$ terms to have a thermodynamically valid description.

IV. REDUCED DESCRIPTION OF HEAT CURRENTS

In order to calculate the heat current from the reduced system dynamics, we must evaluate the expectation value of the collective bath coordinate. To do so, we adapt a generating functional approach by adding the source term, $f_k(t)$, for the $k$th interaction Hamiltonian as

$$
\hat{V}_k \hat{X}_k \to \hat{V}_k f(t) \hat{X}_k \equiv (\hat{V}_k + f_k(t)) \hat{X}_k.
$$

(11)

We add the source term only to the ket (left) side of the density operator. The interaction representation of any operator, $\hat{A}$, with respect to the non-interacting Hamiltonian, $\hat{H}_S(t) + \sum_{k} \hat{H}_k^B$ is expressed as $\hat{A}(t)$. The total density operator, $\hat{\rho}_{\text{tot}}(t)$, with the source term is then denoted by $\hat{\rho}_{\text{tot},f}$. This source term enables us to have a collective bath coordinate with the functional derivative as

$$
\left. \hat{X}_k(t) \hat{\rho}_{\text{tot}}(t) = \frac{i\hbar}{\delta f_k(t)} \hat{\rho}_{\text{tot},f}(t) \right|_{f=0},
$$

(12)

Then, for example, the SHC and the interaction Hamiltonian are expressed in terms of the operators in the system space as

$$
\hat{Q}_k^S(t) = \text{Tr}_S \left\{ \left[ \hat{H}_S(t), \hat{V}_k \right] \frac{\delta}{\delta f_k(t)} \hat{\rho}_f(t) \right|_{f=0} \right\}
$$

(13)

and

$$
\left\langle \hat{H}_k^B(t) \right\rangle = i\hbar \text{Tr}_S \left\{ \hat{V}_k \frac{\delta}{\delta f_k(t)} \hat{\rho}_f(t) \right|_{f=0} \right\},
$$

(14)

where $\hat{\rho}(t) \equiv \text{Tr}_B \{ \hat{\rho}_{\text{tot}(t)} \}$ is the reduced density operator of the system obtained by tracing out all bath degrees of freedom. This is expressed in the form of a second-order cumulant expansion, which is exact in the present case, due to the bosonic nature of the bath:

$$
\hat{\rho}_f(t) = \mathcal{T}_+ \{ \mathcal{U}_f(t) \} \hat{\rho}(0).
$$

(15)

Here, $\mathcal{U}_f(t) = \prod_{k=1}^{K} \exp[\int_0^t ds \hat{W}_k(s)]$ is the Feynman-Vernon influence functional in operator form, and $\mathcal{T}_+ \{ \ldots \}$ is the time-ordering operator, where the operators in $\{ \ldots \}$ are arranged in chronological order. Here, the initial state is taken to be the product state of the reduced system and the bath density operators, $\hat{\rho}_{\text{tot}}(0) = \hat{\rho}(0) \prod_{k=1}^{K} \hat{\rho}_{\text{eq},k}^B$, where $\hat{\rho}_{\text{eq},k}^B$ is the canonical density operator for the $k$th bath. Note that, by generalizing the influence functional, we can extend the present result to the case of a mixed initial state of the reduced system and the bath. The operators of the influence phase are defined by

$$
\hat{W}_k(s) = \int_0^s du \hat{\Psi}_k(s) \left[ C_k^R(s-u) \hat{\Psi}_k(u) - C_k^I(s-u) \right. \left. \hat{\Psi}_k(u) \right],
$$

(16)

where we have introduced the two superoperators $\hat{\Phi}_k \hat{\rho} = (i/\hbar) \{ \hat{V}_k, \hat{\rho} \}$ and $\hat{\Psi}_k \hat{\rho} = (1/\hbar) \{ \hat{V}_k, \hat{\rho} \}$, and $C_k(s)$ and $C_k^I(s)$ are the real and imaginary parts of $C_k(s)$. Thus, by applying the functional derivative with respect to $f_k(t)$, to the reduced density operator, we obtain the following relation:

$$
\left. \frac{i\hbar}{\delta f_k(t)} \hat{\rho}_f(t) \right|_{f=0} = - \mathcal{T}_+ \left\{ \int_0^t ds \left[ \frac{C_k^R(t-s)}{C_k} \hat{\Phi}_k(s) - C_k^I(t-s) \hat{\Psi}_k(s) \right] \mathcal{U}_f(t) \right\} \hat{\rho}(0).
$$

(17)
From the generating functional, for the SHC, we obtain the expression

\[
\dot{Q}_S^k(t) = \int_0^t ds \left( C_{IR}^k(t-s) \frac{i}{\hbar} \langle [\dot{A}_k(t), \dot{V}_k(s)] \rangle \right. \\
- C_{Ik}(t-s) \frac{1}{\hbar} \langle [\dot{A}_k(t), \dot{V}_k(s)] \rangle \left. \right),
\]

(18)

where \( \dot{A}_k(t) \equiv (i/\hbar) [\dot{H}_S(t), \dot{V}_k(t)] \). In order to obtain an explicit expression for the BHC given in Eq. (13), we need to evaluate the expectation value of the interaction energy. From the generating functional, this is obtained as

\[
\langle \dot{H}_S^k(t) \rangle = - \int_0^t ds \left( C_{IR}^k(t-s) \frac{i}{\hbar} \langle [\dot{V}_k(t), \dot{V}_k(s)] \rangle \\
- C_{Ik}(t-s) \frac{1}{\hbar} \langle [\dot{V}_k(t), \dot{V}_k(s)] \rangle \right).
\]

(19)

In order to obtain this expression, we have divided the real part of the noise correlation function into a short-time correlated part, expressed by a delta-function, and the remaining part, as \( C_{IR}^k(s) = C_{Ik}^k(s) + 2 \Delta \delta(s) \). The imaginary part of the noise correlation function is similarly divided into a delta-function part and the remaining part, but in this case, we incorporate this delta-function part into the system Hamiltonian by renormalizing the frequency. Taking the time derivative of Eq. (19), we obtain the following:

\[
\frac{d}{dt} \langle \dot{H}_S^k(t) \rangle = - \int_0^t ds \left( C_{IR}^k(t-s) \frac{i}{\hbar} \langle [\dot{V}_k(t), \dot{V}_k(s)] \rangle \\
- C_{Ik}(t-s) \frac{1}{\hbar} \langle [\dot{V}_k(t), \dot{V}_k(s)] \rangle \right) \\
+ \frac{i}{\hbar} \Delta_i \left( \left[ \frac{d\dot{V}_k(t)}{dt}, \dot{V}_k(t) \right] \right) \\
+ \frac{2}{\hbar} C_{Ik}^k(0) \langle \dot{V}_k^2(t) \rangle.
\]

(20)

The lhs of the above equation is identical to \( \dot{Q}_S^k(t) \), because we have

\[
\left[ \left\langle \frac{d\dot{V}_k(t)}{dt} \right\rangle, \dot{V}_k(t) \right] = -\dot{Q}_S^k(t) - \sum_{k' \neq k} \dot{q}_{k,k'}(t),
\]

(21)

which is derived from the Heisenberg equation of motion for \( \dot{V}_k \),

\[
\frac{d\dot{V}_k(t)}{dt} = \dot{A}_k(t) - \sum_{k' \neq k} \frac{i}{\hbar} [\dot{V}_k(t), \dot{V}_{k'}(t) \dot{X}_{k'}(t)].
\]

(22)

Accordingly, using the relation

\[
\frac{i}{\hbar} \left\langle \frac{d\dot{V}_k(t)}{dt}, \dot{V}_k(t) \right\rangle = \sum_{k' \neq k} \int_0^t ds \left( C_{IR}^k(t-s) \frac{i}{\hbar} \langle [\dot{B}_{k,k'}(t), \dot{V}_{k'}(s)] \rangle \\
- C_{Ik}(t-s) \frac{1}{\hbar} \langle [\dot{B}_{k,k'}(t), \dot{V}_{k'}(s)] \rangle \right) \\
+ \frac{i}{\hbar} \langle [\dot{A}_k(t), \dot{V}_k(t)] \rangle,
\]

(23)

where \( \dot{B}_{k,k'} \equiv (i/\hbar)^2 [\dot{V}_k, \dot{V}_{k'}], \dot{V}_k \), we obtain the following as the final expression for the BHC:

\[
\dot{Q}_B^k(t) = - \int_0^t ds \left( C_{IR}^k(t-s) \frac{i}{\hbar} \langle [\dot{V}_k(t), \dot{V}_k(s)] \rangle \\
- C_{Ik}(t-s) \frac{1}{\hbar} \langle [\dot{V}_k(t), \dot{V}_k(s)] \rangle \right) \\
+ \frac{2}{\hbar} C_{Ik}^k(0) \langle \dot{V}_k^2(t) \rangle \\
+ \frac{1}{\hbar} \langle [\dot{A}_k(t), \dot{V}_k(t)] \rangle.
\]

(24)

Now that we have obtained the explicit expressions for the SHC and BHC, the remaining task is to evaluate these expressions in a numerically rigorous manner. This was carried out using the HEOM approach.

V. HIERARCHICAL EQUATIONS OF MOTION
APPROACH

When the noise correlation function, Eq. (2), is written as a linear combination of exponential functions and a delta function, \( C_{kk}(t) = \sum_{j=0} J_j c_{kk}^j e^{-\gamma_j t} + 2 \Delta \delta(t) \), which is realized for the Drude, Lorentz, and Brownian cases (and combinations thereof), we can obtain the reduced equations of motion as the HEOM. The HEOM consist of the following set of equations of motion for the auxiliary density operators (ADOs):

\[
\rho_{\vec{n}_1,...,\vec{n}_k}(t) \equiv \mathcal{T} \left\{ \exp \left[ - \frac{i}{\hbar} \int_0^t ds l(s) \right] \right\} \\
\times \mathcal{T} \left\{ \prod_{k=1}^K \prod_{j=0}^{J_k} \left[ - \int_0^t ds e^{-\gamma_j (t-s)} \hat{O}_{kj}(s) \right] \hat{O}_{kj}(0) \right\} \rho(0).
\]

(25)

Here, we have \( \hat{O}_{kj} \equiv c_{kj}^k \hat{F}_k - c_{kj}^k \hat{F}_k \) and \( l(t) \rho = [\hat{H}_S(t), \rho] \). Each ADO is specified by the index \( \vec{n}_k = \).
For the ADOs given in Eq. (25), we can evaluate the noise correlation functions in Eqs. (18) for a static system coupled to a single bath (tangled state of the system and the baths; for example, that the steady-state solution of the HEOM is an ensemble of density operators, which are obtained by integrating Eq. (26) with respect to time until convergence to the steady state is realized. It is important to note that the steady-state solution of the HEOM takes the form $$\rho_{\text{ss}} = \sum_{\{\vec{n}\}} \rho_{\{\vec{n}\}}$$

where $$\vec{c}_j$$ is the unit vector along the jth direction. The HEOM consist of an infinite number of equations, but they can be truncated at finite order by ignoring all $$k_j$$ beyond the value at which $$\sum_{k_j} n_{k_j}$$ first exceeds some appropriately large value N.

Employing the noise decomposition of the HEOM approach for the noise correlation functions in Eqs. (18) and (22), and comparing the resulting expressions with the definition of the ADOs given in Eq. (25), we can evaluate the SHC and BHC in terms of the ADOs as

$$\hat{Q}^S_k(t) = - \sum_{j=0}^{J_k} \text{Tr}\{\hat{A}_k(t)\hat{\rho}_{\{\vec{n}\},\{\vec{e}\}}(t)\}$$

and

$$\hat{Q}^B_k(t) = - \sum_{j=0}^{J_k} \gamma_j \text{Tr}\{\hat{V}_k\hat{\rho}_{\{\vec{e}\}}(t)\}$$

where $$\gamma_j$$ is the coupling strength, and the spectral density of each bath takes the Drude form, $$\gamma_j/\omega$$.

The non-equilibrium spin-Boson model studied here consists of a two-level system coupled to two bosonic baths at different temperatures. This model has been employed extensively as the simplest heat-transport model. The system Hamiltonian is given by $$\hat{H}_S = (\hbar\omega_0/2)\sigma_z$$.

We consider the case in which the system is coupled to the first bath through $$\hat{V}_1 = \sigma_x$$ and to the second bath through $$\sigma_x$$ and $$\sigma_z$$ in the form $$\hat{V}_2 = (s_x\sigma_x - s_z\sigma_z)/s^2_x + s^2_z$$). In order to investigate the difference between the SHC and BHC, we consider the case $$s_z \neq 0$$, because otherwise the CASBI term vanishes, and thus the SHC coincides with the BHC. (This is the case that most of previous investigations have considered.) It should be noted that the CASBI has a purely quantum origin, as can be seen from its definition. We chose $$T_1 = 2.0\omega_0/k_B, T_2 = 1.0\omega_0/k_B, \gamma = 2.0\omega_0$$ and $$s_z = 1$$.

Figure 2 depicts the role of non-commuting component of the heat currents, which we carried out numerical simulations for a non-equilibrium spin-boson model and a three-level heat engine model with the HEOM approach (Fig. 1). Although, with this approach, we can study transient behavior without any difficulty, here we focus on investigating physical quantities in the steady state, because the definition of the heat current is ambiguous in the non-steady-state case. We assume that the spectral density of each bath takes the Drude form, $$\gamma_j/\omega$$.

VI. NUMERICAL ILLUSTRATION

To demonstrate the role of the CASBI in the heat current, we carried out numerical simulations for a non-equilibrium spin-boson model and a three-level heat engine model with the HEOM approach (Fig. 1). Although, with this approach, we can study transient behavior without any difficulty, here we focus on investigating physical quantities in the steady state, because the definition of the heat current is ambiguous in the non-steady-state case. We assume that the spectral density of each bath takes the Drude form, $$\gamma_j/\omega$$.

A. Non-equilibrium spin-Boson model

The non-equilibrium spin-boson model studied here consists of a two-level system coupled to two bosonic baths at different temperatures. This model has been employed extensively as the simplest heat-transport model. The system Hamiltonian is given by $$\hat{H}_S = (\hbar\omega_0/2)\sigma_z$$.

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FIG. 2: The heat currents of the non-equilibrium spin-boson model are plotted as functions of $s_x$ in order to illustrate the effect of the fact that $V_1$ and $V_2$ do not commute. The solid (blue) and the dashed (black) curves represent $\dot{Q}_B$ and $\dot{Q}_S$, which correspond to the BHC and SHC.

BHC processes in the steady state as functions of $s_x$. Here, the system-bath coupling strengths are set to $\eta_1 = \eta_2 = 0.01\omega_0$. Even when the system Hamiltonian commutes with the second interaction Hamiltonian in the case $s_x = 0$ (i.e., even when the system couples to the second bath in a non-dissipative manner with the interaction $s_x\sigma_z$ as $[H_S, H_2^2] = 0$), non-zero heat current arises due to the CASBI contribution, $\dot{q}_{1,2}$. This is because the Hamiltonian of the system plus system-bath interactions does not commute with the second interaction (i.e., $[H_S + \sum_{k=1,2} H_k^2, H_2^1] = [H_1^1, H_2^1] \neq 0$), while the system Hamiltonian and the second interaction Hamiltonian do commute.

Figure 3 depicts the heat currents as functions of the system-bath coupling strength for the case $s_x = s_z = 1$. In the weak system-bath coupling regime, the SHC and BHC increase linearly with the coupling strength in similar manners. It is thus seem that in this case, the CASBI contribution is minor. As the strength of the system-bath coupling increases, the difference between the SHC and BHC becomes large: While $\dot{Q}_S^1$ decreases after reaching a maximum value near $\eta_1 = \eta_2 = 0.2\omega_0$, the CASBI contribution, $\dot{q}_{1,2}$, dominates the BHC, and as a result, it remains relatively large. Thus, in this regime, the SHC becomes much smaller than the BHC. In the very strong coupling regime, the SHC eventually becomes negative, which indicates the violation of the second law.

Each curve is properly adjusted as $\dot{Y} \rightarrow \dot{Y} - \frac{1}{2}(\max_t\{\dot{Y}(t)\} + \min_t\{\dot{Y}(t)\})$ for $Y = \dot{Q}_S^1, \dot{Q}_S^2$, and $W$.

B. Three-level heat engine model

The three-level heat engine model considered here consists of three states, denoted by $|0\rangle$, $|1\rangle$, and $|2\rangle$, coupled to two bosonic baths. The system is driven by a periodic external field with frequency $\Omega$. The system Hamiltonian

$$H = H_0 + \sum_{\alpha \in \{1,2\}} \eta_{\alpha} a_\alpha^\dagger a_\alpha + i\hbar\Omega a_1^\dagger a_2.$$ 

The Hamiltonian $H_0$ contains the system states $|0\rangle$, $|1\rangle$, and $|2\rangle$, with energies $E_0$, $E_1$, and $E_2$, respectively. The two baths are described by the Hamiltonian

$$H_\alpha = \sum_n \omega_n a_\alpha^\dagger a_n + \text{H.c.},$$ 

where $\omega_n$ are the frequencies of the bath modes, and $a_\alpha^\dagger, a_\alpha$ are the creation and annihilation operators for the bath modes. The coupling between the system and the baths is given by the terms $\eta_{\alpha} a_\alpha^\dagger a_\alpha$, which describe the interaction between the system and the $\alpha$-th bath. The system Hamiltonian $H_0$ is coupled to the $\alpha$-th bath via the interaction

$$H_{\alpha,\beta} = i\hbar\Omega a_\alpha^\dagger a_\beta,$$ 

where $\alpha, \beta = 1, 2$. The time evolution of the system is governed by the Schrödinger equation

$$i\hbar\partial_t |\psi(t)\rangle = H|\psi(t)\rangle.$$

The system is initially prepared in the state $|\psi(0)\rangle = |0\rangle$, and it is driven by a periodic external field of frequency $\Omega$. The goal is to study the thermodynamic performance of the heat engine, such as the power output $W$ and the efficiency $\eta$. The power output is given by the average heat current $\dot{W} = \langle \dot{Q}_B \rangle$, where $\dot{Q}_B$ is the heat current from the bath to the system.

The system is in a state $|\psi(t)\rangle$ at time $t$, and the power output $W(t)$ is defined as

$$W(t) = \langle \dot{Q}_B(t) \rangle = \frac{1}{2}\hbar\Omega |\langle \psi(t) | a_1^\dagger a_2 | \psi(t) \rangle|^2.$$ 

The efficiency $\eta$ is defined as the ratio of the power output to the maximum work that can be done by the system, which is given by the Clausius inequality

$$\eta = \frac{\dot{W}}{\dot{Q}_B}.$$ 

The three-level heat engine model is a generalization of the two-level heat engine model, where the system can absorb heat from two different baths, and the power output can be controlled by the driving frequency $\Omega$. The efficiency of the heat engine is a function of the driving frequency, and it is possible to optimize the efficiency by adjusting $\Omega$. The optimal driving frequency is given by

$$\Omega = \sqrt{\sum_{\alpha = 1,2} \eta_{\alpha}}.$$ 

The optimal efficiency is achieved when $\eta_1 = \eta_2$, which is a consequence of the principle of maximum work. The efficiency of the heat engine is limited by the second law of thermodynamics, and it cannot exceed the Carnot efficiency for a heat engine with the same temperature differences between the baths.

The three-level heat engine model is a promising candidate for realizing efficient heat engines in quantum systems, where the system can absorb heat from two different baths, and the power output can be controlled by the driving frequency. The optimal performance of the heat engine is achieved when the system absorbs heat from the baths at the optimal driving frequency, which is given by $\Omega = \sqrt{\sum_{\alpha = 1,2} \eta_{\alpha}}$. The efficiency of the heat engine is a function of the driving frequency, and it is possible to optimize the efficiency by adjusting $\Omega$. The optimal driving frequency is achieved when $\eta_1 = \eta_2$, which is a consequence of the principle of maximum work. The efficiency of the heat engine is limited by the second law of thermodynamics, and it cannot exceed the Carnot efficiency for a heat engine with the same temperature differences between the baths.
is expressed as
\[
\hat{H}_S(t) = \sum_{i=0,1,2} \hbar \omega_i |i\rangle \langle i| + g(e^{-\Omega t}|1\rangle \langle 2| + \text{H.c.}) \tag{29}
\]
with $\omega_1 > \omega_2 > \omega_0$. The system-bath interactions are defined as $\hat{V}_1 = |0\rangle \langle 1| + |1\rangle \langle 0|$ and $\hat{V}_2 = |2\rangle \langle 0| + |0\rangle \langle 2|$. We set $\omega_0 = 0$ without loss of generality. Roughly stated, this model acts as a quantum heat engine when population inversion between the two excited states, $|1\rangle$ and $|2\rangle$, occurs. This can be realized in the case that the temperature of the first bath, $T_1$, is sufficiently higher than that of the second bath, $T_2$. Using this model, we analyze the work and the heat per cycle, i.e., $Y_{\text{cyc}} = \lim_{T\rightarrow \infty} \int_t^{t + 2\pi/|\Omega|} Y(t') dt'$ for $Y = W, Q_S^1, Q_B^1$ with $k = 1$ or 2. We set $\omega_2 = 0.5\omega_1$, $\Omega = 0.5\omega_1$, $\gamma = 2.0\omega_1$, and $\Omega = 0.1\omega_1$.

In Figure 4, we illustrate the time dependences of the two SHC, $Q_S^1$ and $Q_S^2$, and the power, $W$, that arise from transitions between $|0\rangle$ and $|1\rangle$, $|0\rangle$ and $|2\rangle$, and $|1\rangle$ and $|2\rangle$, respectively, for several values of the first bath coupling, with the second bath coupling fixed ($\eta_2 = 0.001\omega_1$). The figure depicts $Q_S^1, Q_S^2$, and $W$ for one cycle of the external force. We set $t = 0$ and $t = 1$ to correspond to the maxima of the cyclic driving field. The time delays observed for $Q_S^1, Q_S^2$, and $W$ imply that the transition $|0\rangle \rightarrow |1\rangle \rightarrow |2\rangle$ is cyclic. This behavior can be regarded as a microscopic manifestation of a quantum heat engine. The periods of the currents and power are, however, half as long as the period of the driving field. This is because, while the transition for the work production is induced by an even number of system-field interactions, the second-order interaction that involves components with frequency $2\Omega$, which is twice that of the system-field interaction, becomes dominant.

The phases of $Q_S^1, Q_S^2$ and $W$ depend on the first bath coupling. Because the strength of the second bath coupling and the external fields are weak, all of these changes are a consequence of the change in $Q_S^1$. When the first bath coupling is weak, the first SHC, $Q_S^1$, which is the current from the high-temperature heat bath, cannot follow the change of the external field and hence exhibits a delay in its response to the decrease of the heat current that occurs at the maxima of the field intensity. As a result, the power, $W$, and the heat current for the low-temperature bath, $Q_S^2$, exhibit successively delayed response. When the first bath coupling is strong, $Q_S^1$ closely follows the variation of the field. The time delays of $Q_S^2$ and $W$ also decrease as first bath coupling increases.

In Fig. 5, we depict the system efficiency, $\epsilon_S \equiv -W_{\text{cyc}}/Q_S^{\text{cyc,1}}$, and the bath efficiency, $\epsilon_B \equiv -W_{\text{cyc}}/Q_B^{\text{cyc,1}}$, as functions of the strength of the coupling to the first bath with fixed strength of the coupling to the second bath, $\eta_2 = 0.001\omega_1$. Here, $W_{\text{cyc}}$ and $Q_{\text{cyc}}$ represent the time average of $W$ and $Q$ per cycle. We consider a high temperature case with $T_2 = \hbar\omega_1/k_B$ and a low temperature case with $T_2 = 0.1\hbar\omega_1/k_B$, with the fixed ratio $T_2/T_1 = 0.1$.

While the system efficiency is weakly dependent on the strength of the first bath coupling, regardless of the temperature, the bath efficiency decreases as the strength of the first bath coupling increases, in particular in the low temperature case. The reason for this can be understood from Fig. 6. There, it is seen that $Q_S^{\text{cyc,1}}$ decreases as the strength of the first bath coupling increases, as a result of the strong suppression of the thermal activation by dissipation. Because we set the strength of the second bath coupling and the external fields are weak, the work $-W_{\text{cyc}}$ tends to follow the behavior of $Q_S^{\text{cyc,1}}$, as illustrated in Fig. 4 whereas $Q_B^{\text{cyc,1}}$ increases as the strength of the coupling to the first bath increases, due to the CASBI contribution, $\dot{q}_{1,2}$. As a result, the system efficiency, $-W_{\text{cyc}}/Q_S^{\text{cyc,1}}$, does not change significantly, whereas the bath efficiency, $-W_{\text{cyc}}/Q_B^{\text{cyc,1}}$, decreases as a function of the first coupling strength.

In the strong coupling regime, the bath efficiency in the low temperature case is larger than that in the high temperature case, as depicted in Fig. 5 because as the temperature decreases, the system coupled to the low temperature bath becomes less activated. Note that if the strength of the coupling to the second bath is sufficiently large that the system is in the non-perturbative regime, the system efficiency decreases as the strength of the coupling to the first bath increases, because in this case, a part of $-W$ goes to $Q_S^{\text{cyc,2}}$.

Unlike in the non-equilibrium spin-boson case, the system efficiency is physically meaningful. We believe, however, that the bath efficiency is more appropriate as the rigorous definition of the heat efficiency, because the system efficiency does not include the contribution to the energy from the system-bath interactions, which must be regarded as a part of the system. The decrease of efficiency can be regarded as a quantum effect, because it originates from the discretization of the energy eigenstates.

VII. CONCLUDING REMARKS

In this paper, we introduced an explicit expression for the bath heat current (BHC), which includes contributions from the correlations among the system-bath interactions (CASBI). The BHC reduces to the widely used system heat current (SHC) derived in terms of the system energy under conditions of a weak system-bath coupling or in the case that all system-bath interactions commute. Our definition of the BHC can be applied to any system with any driving force and any strength of the system-bath coupling. We numerically examined the role of the CASBI using the HEOM approach. We demonstrated this approach in the case of a non-equilibrium spin-boson system in which the CASBI contribution is necessary to maintain consistency with thermodynamic laws in the strong system-bath coupling regime. In the three-level heat-engine model, we observed cyclic time evolution of the high-temperature heat current, the work, and the
Here, we consider $\epsilon$ (dash-dotted line) in the low temperature case, with $T$ with fixed weak coupling to the second bath ($\eta_2 = 0.001\omega_1$). Here, we consider $\epsilon_S = -W^{\text{cyc}}/Q_S^{\text{cyc},1}$ (solid line) and $\epsilon_B = -W^{\text{cyc}}/Q_B^{\text{cyc},1}$ (curve with circles) in the high temperature case, with $T_2 = 1.0\hbar\omega_1/k_B$, and $\epsilon_S$ (dashed curve) and $\epsilon_B$ (dash-dotted line) in the low temperature case, with $T_2 = 0.1\hbar\omega_1/k_B$.

![Diagram](image)

FIG. 5: The efficiencies of the three-level heat engine calculated as functions of the coupling to the first bath, $\eta_1$, with fixed weak coupling to the second bath ($\eta_2 = 0.001\omega_1$). Here, we consider only the high temperature case, with $T_2 = 1.0\hbar\omega_1/k_B$, and $\epsilon_S$ (dashed curve) and $\epsilon_B$ (dash-dotted line) in the low temperature case, with $T_2 = 0.1\hbar\omega_1/k_B$.

low-temperature heat current, as in a classical heat engine. When the system-bath coupling is weak, there is a time delay between the variation of the external field and the heat current of the high-temperature bath, because this bath cannot follow variations of the system, due to the weakness of the system-bath coupling. Contrastingly the heat current does not exhibit any time delay in the strong system-bath coupling case. The efficiency defined using the BHC, which is regarded as physically more appropriate than that defined using the SHC, decrease as the strength of the system-bath coupling increases.

Although our present analysis is restricted to the steady-state case, we can also apply our formulation to analysis of transient behavior, in which the variation of the bath energy in time is experimentally measurable. Because the HEOM approach is capable of calculating various physical quantities in non-equilibrium situations, it would also be interesting to extend the present investigation to other quantum transport problems by calculating higher-order cumulants [49] and non-linear optical signals [50–52] to reveal the detailed physical properties of the dynamics. We leave such problems to future studies.

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Appendix A: Derivation of the second law

In this Appendix, we derive the Clausius inequality as the second law of thermodynamics for quantum steady states by extending the result of Defner and Jarzynski [53] for the classical case to the quantum case. Note that this derivation is not limited to the case of reduced dynamics. Because extension to the case of multiple heat baths is straightforward, here we consider the case of a single bath, with the Hamiltonian $\hat{H}(t) = \hat{H}_S(t) + \hat{H}_1 + \hat{H}_B$. We consider the von Neumann entropy $\mathcal{H}(t)$ defined as

$$\mathcal{H}(t) \equiv -\text{Tr}\{\hat{\rho}(t) \ln \hat{\rho}(t)\},$$

(A1)

where $\hat{\rho}(t)$ is the total density operator. The reduced density operator for the system is $\hat{\rho}_S(t) \equiv \text{Tr}_B\{\hat{\rho}(t)\}$, while that for the bath is $\hat{\rho}_B(t) \equiv \text{Tr}_S\{\hat{\rho}(t)\}$. Then we introduce the von Neumann entropies of the system and bath as $\mathcal{H}_S(t) = -\text{Tr}\{\hat{\rho}_S(t) \ln \hat{\rho}_S(t)\}$ and $\mathcal{H}_B(t) = -\text{Tr}\{\hat{\rho}_B(t) \ln \hat{\rho}_B(t)\}$, respectively. Without loss of generality, we assume that the total system is initially in the factorized state $\hat{\rho}(0) = \hat{\rho}_S(0)\hat{\rho}_B^\text{eq}$, where $\hat{\rho}_B^\text{eq} = e^{-\beta B}/Z_B$ is the equilibrium density operator of the bath, in which $Z_B$ is the partition function of the bath. In this case, the total entropy is merely the sum of the system and bath entropies:

$$\mathcal{H}(0) = \mathcal{H}_S(0) + \mathcal{H}_B(0).$$

(A2)

Because the von Neumann entropy is invariant under unitary evolution (i.e. $\mathcal{H}(t) = \mathcal{H}(0)$), and because the von Neumann entropy is sub-additive (i.e. $\mathcal{H}(t) \leq \mathcal{H}_S(t) + \mathcal{H}_B(t)$), we have the following inequality for $\Delta \mathcal{H}_S(t) \equiv \mathcal{H}_S(t) - \mathcal{H}_S(0)$ and $\Delta \mathcal{H}_B(t) \equiv \mathcal{H}_B(t) - \mathcal{H}_B(0)$ :

$$\Delta \mathcal{H}_S(t) + \Delta \mathcal{H}_B(t) \geq 0.$$ 

(A3)
We now rewrite the von Neumann entropy of the bath as
\[ \mathcal{H}_B(t) = -\text{Tr}\left(\hat{\rho}_B(t) \ln \hat{\rho}_B(t)\right) - \text{Tr}\left(\hat{\rho}_B(t) \ln \hat{\rho}_B(t)\right) - \text{Tr}\left(\hat{\rho}_B(t) \ln \hat{\rho}_B(t)\right) \]
\[ = \beta E_B(t) - \beta F_B - D(\hat{\rho}_B(t)||\hat{\rho}_B^{eq}), \quad (A4) \]
where \( E_B(t) = \text{Tr}\left(\hat{H}_B\hat{\rho}_B(t)\right), F_B = -\beta^{-1} \ln Z_B, \) and \( D(\hat{\rho}||\hat{\sigma}) = \text{Tr}(\hat{\rho} \ln \hat{\rho}) - \text{Tr}(\hat{\rho} \ln \hat{\sigma}) \geq 0 \) are the bath energy, the bath free energy, and the quantum relative entropy, respectively. This leads to the condition
\[ \Delta \mathcal{H}_B(t) = \beta \Delta E_B(t) - D(\hat{\rho}_B(t)||\hat{\rho}_B^{eq}) \leq \beta \Delta E_B(t). \quad (A5) \]
Then, using Eq. (A3), we obtain the inequality
\[ \Delta \mathcal{H}_S(t) + \beta \Delta E_B(t) \geq 0. \quad (A6) \]

A quantum heat machine is subject to a periodic perturbation, \( \hat{H}_S(t) = \hat{H}_S(t+T) \), where \( T \) is the period of the perturbation. After a sufficiently long time, the system relaxes to a periodic steady state, with \( \hat{\rho}_S(t) = \hat{\rho}_S(t+T) \). We separate the time of observation into a transient part (from \( t = 0 \) to \( n_0 T \)) and a steady part (from \( t = n_0 T \) to \( nT \)), where \( n_0 \) is an integer sufficiently large to ensure that the system is in the periodic steady state at \( n_0 T \). The change in a variable \( A \) from time \( n_0 T \) to \( nT \) is written \( \Delta A^{n_0 \rightarrow n} = A(nT) - A(n_0 T) \). The inequality Eq. (A6) is then partitioned as
\[ \Delta \mathcal{H}^{0 \rightarrow n_0} + \Delta \mathcal{H}^{n_0 \rightarrow n} + \beta \Delta E_B^{0 \rightarrow n_0} + \beta \Delta E_B^{n_0 \rightarrow n} \geq 0. \quad (A7) \]

This is the result presented in Sec. III.

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