Thermodynamics of Mesoscopic Thermoelectric Heat Engine beyond Linear-Response Regime

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Mesoscopic thermoelectric heat engine is much anticipated as a new device which allows us to utilize wasted heat inaccessible by the conventional heat engine with high efficiency. Most theoretical studies so far, however, have been limited to the linear-response regime; its thermodynamics beyond the regime still remains unclear. In this Letter, we give a clear-cut definition of the heat current of the engine beyond the linear-response regime. It resolves the confusion in the definition of the heat current in the linear-response regime. After verifying its thermodynamic consistency, we find the following two interesting results: the efficiency of the mesoscopic thermoelectric engine reaches the Carnot efficiency if and only if the transmission function is a delta function at a specific energy; the unitarity of the scattering matrix guarantees the second law of thermodynamics, invalidating Benenti et al.’s argument in the linear-response regime that one could obtain a finite power with the Carnot efficiency under broken time-reversal symmetry.

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Introduction — Thermolectric heat engine [1–15] is much anticipated as a new device which allows us to utilize wasted heat inaccessible by the conventional heat engine. This engine operates in a non-equilibrium steady state and converts heat to useful electrical power steadily, so that we do not need non-steady processes used in cyclic engines, such as adiabatic compression, isothermal expansion, and so forth. Its efficiency, however, so far has been too low to use in terms of the figure of merit $Z(T)$, which is a serious problem in the field of thermoelectricity [16–18].

The mesoscopic thermoelectric heat engine [2, 4, 7–12, 14, 15, 19–21] has emerged as a possible solution. For example, it has been argued that the heat current can be bounded because of the uncertainty principle [20–22] and that the unitarity of the scattering matrix gives a new bound for the Onsager coefficients in the linear-response regime [10, 11].

However, researches on this engine have been limited mostly to the linear-response regime [23]; its thermodynamics beyond the regime is yet to be clarified. Although some researchers have already used the definition of the heat current which we will derive in this Letter beyond the linear-response regime [2, 4, 19, 21, 24], few discussions have been publicized regarding even its origin and validity. This is in contrast to the study of the cyclic heat engine, where we can apply thermodynamics regardless of the difference of temperatures and chemical potentials between hot and cold reservoirs.

The most fundamental problem is how we go beyond the linear-response regime; in experiments, nonlinear effects often cannot be ignored [25–27]. We, in the present Letter, construct thermodynamic formalism beyond the linear-response regime under reasonable assumptions. We first give a clear-cut definition of the heat current of the mesoscopic thermoelectric heat engine by taking in the non-linear terms. After verifying its thermodynamic consistency, we find the following two interesting results using this definition: the efficiency of the mesoscopic thermoelectric engine reaches the Carnot efficiency if and only if the transmission function is a delta function at a certain energy; the unitarity of the scattering matrix guarantees the second law of thermodynamics, invalidating Benenti et al.’s argument in the linear-response regime that one could obtain a finite power with the Carnot efficiency under broken time-reversal symmetry.

Thermoelectric heat engine — Let us explain the steady-state heat engine. Consider a system attached to two reservoirs on both sides; see Fig. 1. We then make the following three assumptions: the reservoirs are so much larger than the system that it is always at equilibrium if they interact with the system, and hence we can define thermodynamic quantities of each reservoir, such as the temperature and the chemical potential; the system has reached a non-equilibrium steady state in which there are constant flows from the left reservoir to the right one; there is no dissipation in the system because electrons undergo only elastic scattering there.

This system is regarded as a heat engine under the following conditions. We set the chemical potential of the right reservoir higher than that of the left, while the
Its efficiency $\eta$ can thus consider this system as a heat engine. Because and then dump heat $J$ heat. This is certainly not correct in the situation in electrons did not do work and hence all energy became $\mu_R = \mu_L$. The choices do not make difference in the linear response of the voltage difference $(\mu_R - \mu_L)/e$ between $J_Q^L$ and $J_Q^R$ but differ in higher orders.

We here define the heat currents $J_Q^L$ and $J_Q^R$ thermodynamically consistently. The first law of thermodynamics gives the relations

$$dU^\alpha = dQ^\alpha + dW^\alpha,$$

where $\alpha = L, R$, $dU^\alpha$ and $dQ^\alpha$ denote the energy and heat flowing into the left (right) reservoir, and $dW^\alpha = \mu_\alpha dN^\alpha$ is the work done to the reservoir. Using (3), we can express $dQ^\alpha$ in the form

$$dQ^\alpha = dU^\alpha - \mu_\alpha dN^\alpha.$$  

Note that we managed to define the heat flowing to a nonequilibrium system thanks to the first assumption above.

Since we treat the nonequilibrium steady state, we can define the change of the particle number and energy in a reservoir as a steady current, which enables us to define the particle and energy currents as

$$J_N^L = \frac{dN^L}{dt}, \quad J_N^R = \frac{dN^R}{dt}.$$  

which is the same as the standard cyclic heat engine. Note that when the directions of all currents are inverted, we can see this engine as a refrigerator, whose coefficient of performance is given by $\eta_{\text{cop}} = J_Q^L/(IV)$.

**Heat current** — Before defining the heat current, we mention the confusion in the definition of the heat current in the linear-response regime. We first note that the energy current $J_E$ is often referred to as a ‘heat’ current [28, 29]. This would be correct, though confusing, if the electrons did not do work and hence all energy became heat. This is certainly not correct in the situation in Fig. [1].

Another definition $J_Q = J_E - \mu J_N$ was often used in the dawn of the research of heat current in mesoscopic systems [20]. This definition may have been taken from Eq. (17.8) in Callen’s textbook [23]. Since this ‘heat’ current was not microscopically derived, we do not clearly know where it flows. It is indeed ambiguous of which part of the system in Fig. [1] the chemical potential $\mu$ of the heat current $J_Q = J_E - \mu J_N$ is. We should probably choose $\mu$ so that $J_Q$ may satisfy Onsager’s reciprocal theorem. For example, in ref. [30] the authors chose $\mu$ as $(\mu_L + \mu_R)/2$ and in ref. [31] the author chose $\mu$ as $\mu_L$. The choices do not make difference in the linear response of the voltage difference $(\mu_R - \mu_L)/e$ between $J_Q^L$ and $J_Q^R$ but differ in higher orders.

We then show that the upper limit of this efficiency is the Carnot efficiency as is expected from the theory of the standard cyclic heat engine. Let $S_L dt = dQ^L/T_L$ and $S_R dt = dQ^R/T_R$ denote the entropy productions in the left and right reservoirs, respectively. Note that the entropy production of the system is zero because we assume that the system has reached a non-equilibrium steady state. Using (6), we can relate these entropy productions to the conventional heat flows as

$$J_Q^L = -T_L \dot{S}_L, \quad J_Q^R = T_R \dot{S}_R.$$  

FIG. 1. Schematic picture of a mesoscopic heat engine. We set the chemical potential of the right reservoir higher than the left, while the temperature of the left reservoir higher than the right so that an electric current may go from left to right against the difference of the chemical potential.
Imposing the condition that the net entropy production $\dot{S} = \dot{S}_L + \dot{S}_R$ is positive, that is, $\dot{S}_L \geq -\dot{S}_R$, we have with \cite{1}, \cite{2}, and \cite{7}

$$\eta = \frac{T_L \dot{S}_L + T_R \dot{S}_R}{T_L \dot{S}_L} \leq 1 - \frac{T_R}{T_L} = \eta_c,$$  \hspace{1cm} (8)

where $\eta_c$ is the Carnot efficiency. We can achieve the equality if and only if $\dot{S}_L = -\dot{S}_R$, that is, $\dot{S} = 0$. We conclude that the heat currents (6) is thermodynamically consistent because of Eqs. (1) and (8), which are the same formulas as those of the cyclic engine.

**Heat current in the mesoscopic heat engine** — Here we derive the heat current in the mesoscopic thermoelectric heat engine. We assume that the system in Fig. 1 is a coherent conductor which accommodates the Landauer-Büttiker formalism \cite{32}. We can obtain $J_N$ and $J_E$ in a microscopically as

$$J_N = \frac{1}{\hbar} \int_{-\infty}^{\infty} d\epsilon \tau(\epsilon)(f_L(\epsilon) - f_R(\epsilon)), \hspace{1cm} (9)$$

$$J_E = \frac{1}{\hbar} \int_{-\infty}^{\infty} d\epsilon \tau(\epsilon)c(\epsilon)f_L(\epsilon) - f_R(\epsilon)), \hspace{1cm} (10)$$

where $\hbar$ is the Planck constant, $f_\alpha(\epsilon) = [1 + \exp(\beta_\alpha(\epsilon - \mu_\alpha))]^{-1}$ is the Fermi distribution function of the reservoir ($\alpha = L,R$), $\beta_\alpha = T_\alpha^{-1}$ is the inverse temperature, and $\tau(\epsilon)$ is the transmission probability at energy $\epsilon$. Substituting (9) and (10) into (6), we arrive at

$$J_Q^N = \frac{1}{\hbar} \int_{-\infty}^{\infty} d\epsilon \tau(\epsilon)(\epsilon - \mu_\alpha)(f_L(\epsilon) - f_R(\epsilon)), \hspace{1cm} (11)$$

where $\alpha = L,R$.

Note that these heat currents satisfy Onsager’s reciprocal theorem when we expand them in terms of appropriate affinities; we can verify the theorem by expanding $J_N$ and $J_Q^N$ in terms of $A_{NQ}^\alpha = \beta_\alpha(\mu_L - \mu_R)$ and $A_{Q}^{\alpha} = -\beta_\alpha$ or $J_N$ and $J_Q^N$ in terms of $A_{NQ}^\alpha = -\beta_\alpha$ and $A_{Q}^{\alpha} = -\beta_\alpha$ \cite{33}.

**Transmission function for the Carnot efficiency** — We then show that the transmission function with which the efficiency reaches the Carnot efficiency is only a delta function. We now know that we can achieve the Carnot efficiency when the total entropy production of the system is zero. The Landauer-Büttiker formalism gives the total entropy production as

$$\dot{S} = \dot{S}_L + \dot{S}_R = -\frac{J_Q^L}{T_L} + \frac{J_Q^R}{T_R}$$

$$= \int_{-\infty}^{\infty} d\epsilon \tau(\epsilon)(f_L(\epsilon) - f_R(\epsilon)) \log \left[ \frac{f_L(\epsilon)(1 - f_R(\epsilon))}{f_R(\epsilon)(1 - f_L(\epsilon))} \right] .$$  \hspace{1cm} (12)

Because $\tau(\epsilon) \geq 0$, the integrand in Eq. (12) is always non-negative. This leads to the positivity of the entropy production, which guarantees the validity of the heat currents (11) in this engine.

Let us then consider the condition for $\dot{S} = 0$. We easily find the following condition: for each value of $\epsilon$, $\tau(\epsilon) = 0$ or $f_L(\epsilon) = f_R(\epsilon)$ if $\tau(\epsilon) = 0$ for any $\epsilon$ or if $f_L(\epsilon) = f_R(\epsilon)$ for any $\epsilon$, however, the transport would not happen, the engine would not work, and the efficiency $\eta$ would be trivially 0/0. The only nontrivial condition is given by $\tau(\epsilon) \neq 0$ and $f_L(\epsilon) - f_R(\epsilon) = 0$ at one value of any energy; if we demanded $f_L(\epsilon) = f_R(\epsilon)$ at two values of energy, they would be equal at any values of energy.

In order to achieve the above condition, the transmission function should be of the form \cite{12, 34}

$$\tau(\epsilon) = \delta(\epsilon - \epsilon_c),$$  \hspace{1cm} (13)

where $\epsilon_c = (T_L\mu_R - T_R\mu_L)/(T_L - T_R)$ is given by the condition $f_L(\epsilon_c) = f_R(\epsilon_c)$. Using \cite{2}, \cite{6}, \cite{9}, and \cite{11}, we indeed find that this transmission function is a necessary and sufficient condition for the Carnot efficiency:

$$\eta = \frac{\mu_L - \mu_R}{\epsilon_c - \mu_L} = 1 - \frac{T_R}{T_L} = \eta_c.$$  \hspace{1cm} (14)

Let us consider the transmission function (13) in the limit of $\Delta \to 0$ of the energy window shown in Fig. 2 in order to see that the same phenomenon as the Carnot cycle happens; the efficiency reaches the Carnot efficiency but the power vanishes in the quasi-static limit. The power $J_N \Delta \mu$ and the heat current $J_Q^N$ vanish in the limit $\Delta \to 0$ as follows:

$$J_N \Delta \mu = \frac{\Delta \mu}{\hbar} \int_{\epsilon_c}^{\epsilon_c + \Delta} d\epsilon (f_L(\epsilon) - f_R(\epsilon))$$

$$= \frac{\Delta \mu}{2\hbar} F(\epsilon_c)\Delta^2 + O(\Delta^3),$$  \hspace{1cm} (15)$$

and

$$J_Q^N = \frac{1}{\hbar} \int_{\epsilon_c}^{\epsilon_c + \Delta} d\epsilon (\epsilon - \mu_L)(f_L(\epsilon) - f_R(\epsilon))$$

$$= \frac{\epsilon_c - \mu_L}{2\hbar} F(\epsilon_c)\Delta^2 + O(\Delta^3),$$  \hspace{1cm} (16)
where $F(\epsilon) = \partial / \partial \epsilon(f_L(\epsilon) - f_R(\epsilon))$. The efficiency becomes the Carnot efficiency as $\Delta \to 0$ as follows:

$$
\eta = \frac{\Delta \eta}{2h} F(\epsilon_c) \Delta^2 + O(\Delta^3) \xrightarrow{\Delta \to 0} \frac{\mu_L - \mu_R}{\epsilon_c - \mu_L} = \eta_c. \quad (17)
$$

Note that the power vanishes in the Carnot limit, which is the same as in the standard heat engine: the Carnot cycle produces zero power. Since $\Delta \sim \hbar t^{-1}$, where $t$ is the transmission time of electrons, it takes infinite time for electrons to transmit from the left reservoir to the right reservoir in the limit $\Delta \to 0$. This corresponds to the quasi-static limit of the cyclic heat engine, in which it takes infinite time for us to operate the engine.

**Second law, reciprocity, and unitarity** — We here remark on the relation of the entropy production with the reciprocity and the unitarity. Using Onsager’s reciprocal theorem, Benenti et al. [10] recently proposed an interesting argument that an engine could have a finite power with the Carnot efficiency under a magnetic field in the linear-response regime. As this proposal is only in the linear-response regime, let us consider its possibility with the entropy production [12], which is valid in nonlinear-response regimes.

For the purpose, we rewrite the entropy production [12] under a magnetic field $B$ in the form

$$
\dot{S}(B) = \int_{-\infty}^{\infty} d\epsilon (\tau_{L \to R}(\epsilon, B)f_L(\epsilon) - \tau_{R \to L}(\epsilon, B)f_R(\epsilon)) 
\times \log \left[ \frac{f_L(\epsilon)(1 - f_R(\epsilon))}{f_R(\epsilon)(1 - f_L(\epsilon))} \right], \quad (18)
$$

where $\tau_{L \to R}(\epsilon, B)$ is the transmission probability for the electrons from left to right at energy $\epsilon$ and $\tau_{R \to L}(\epsilon, B)$ that for the electrons from right to left at energy $\epsilon$. Only under the reciprocity $\tau_{L \to R}(\epsilon, B) = \tau_{R \to L}(\epsilon, -B)$ [32], the entropy production could be zero with a finite power $J_N \neq 0$ when $f_L(\epsilon) = f_R(\epsilon) \neq f_R(\epsilon)\tau_{L \to R}(\epsilon, B)/\tau_{R \to L}(\epsilon, B)$. Since the reciprocity corresponds to Onsager’s reciprocal relation [32], this is the nonlinear version of Benenti et al.’s argument in the linear-response regime [6].

However, the unitarity of the scattering matrix prohibits this situation. The unitarity condition of the scattering matrix yields $\tau_{L \to R}(\epsilon, B) = \tau_{R \to L}(\epsilon, B)$, which takes the entropy production back to [12], and hence we find no power at the Carnot efficiency. This corresponds to the fact that Brandner et al. [10] found a new bound among Onsager’s coefficients from the unitarity of the scattering matrix considering the three-terminal model in the linear-response regime. This bound prevents the power from being finite with the Carnot efficiency. We also note that the unitarity guarantees the positivity of the entropy production in the mesoscopic transport theory.

Since the unitarity comes from the conservation of the particle current, there should be no possibility to obtain the Carnot efficiency at a finite power. Brandner and Seifert [11] argued the attainability of the Carnot efficiency at a finite power in a multi-terminal model when the number of probes (a probe is a special reservoir in which we set the temperature and chemical potential so that the net particle and heat currents flowing into the probe may be zero) is infinite, but they themselves denied it later in ref. [15] because of a numerically conjectured inequality for the Onsager coefficients. From our point of view, we expect that the inequality found in ref. [15] is based on the unitarity.

**Conclusion** — In this Letter, we defined the heat currents [6], which is thermodynamically consistent with [1] and [3]. These heat currents gave us interesting results when we applied them to the mesoscopic thermoelectric heat engine. We found that the heat currents in this engine [11] correctly give the positivity of the entropy production. We also found that the transmission function to achieve the Carnot efficiency is the delta function [13] and that the unitarity of the scattering matrix guarantees the positivity of the entropy production.

It will be interesting to incorporate an electron-electron interaction to our theory. When the interaction is elastic, Eq. (6) is still valid because it does not break our assumptions. We may also expand our theory to the inelastic scattering case, such as electron-hole and electron-phonon interactions, by introducing a third bosonic reservoir that represents the energy dissipation [35].

It is also interesting to look for stronger bounds than the second law of thermodynamics in some systems, which makes it impossible to reach the Carnot efficiency. For a system with many probes, for example, especially under the broken time-reversal symmetry, the transmission probability may not have enough degrees of freedom to be a delta function.

We finally mention the possibility of experimental realization of the mesoscopic heat engine. It may be easy to make the setup of the steady-state heat engine, particularly the mesoscopic thermoelectric heat engine, thanks to the improvement of experimental techniques in mesoscopic transport systems. The results in this Letter can be used from quantum point contacts to one-dimensional nanowires, as well as to cold atoms [36] if we utilize the Landauer-Büttiker formula. A possible difficulty is how we experimentally observe the heat currents and the efficiency of the heat engine. This problem may be solved in near future because the technique of observing the energy current has been improved recently [37].

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