Efficiency and dissipation in a two-terminal thermoelectric junction, emphasizing small dissipation

O. Entin-Wohlman,1, 2 J-H. Jiang,3, 4 and Y. Imry3

1 Raymond and Beverly Sackler School of Physics and Astronomy, Tel Aviv University, Tel Aviv 69978, Israel
2 Department of Physics and the Ilse Katz Center for Meso- and Nano-Scale Science and Technology, Ben Gurion University, Beer Sheva 84105, Israel
3 Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 76100, Israel
4 Present address: Department of Physics, University of Toronto, 60 St. George St., Toronto, Canada ON M5S 1A7

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The efficiency and cooling power of a two-terminal thermoelectric refrigerator are analyzed near the limit of vanishing dissipation (ideal system), where the optimal efficiency is the Carnot one, but the cooling power then unfortunately vanishes. This limit, where transport occurs only via a single sharp electronic energy, has been referred to as “strong coupling” or “the best thermoelectric”. It follows however, that “parasitic” effects that make the system deviate from the ideal limit, and reduce the efficiency from the Carnot limit, are crucial for the usefulness of the device. Among these parasitics, there are: parallel phonon conduction, finite width of the electrons’ transport band and more than a single energy transport channel. In terms of a small parameter characterizing the deviation from the ideal limit, the efficiency and power grow linearly, and the dissipation quadratically. The results are generalized to the case of broken time-reversal symmetry, and the major nontrivial changes are discussed. Finally, the recent universal relation between the thermopower and the asymmetry of the dissipation between the two terminals is briefly discussed, including the small dissipation limit.

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I. INTRODUCTION

There has been major interest in thermoelectric energy conversion1–4. An important application is converting wasted thermal energy into, say, useful and storable electrical one. Also, deriving electricity from the more easily available thermal energy in remote environments is often relevant.5–7 Thermoelectric cooling is a real possibility. Obviously, there exist rigid thermodynamic limitations to such processes. However, the efficiency of existing devices is severely further limited by material and device properties so that getting a sizable fraction of the thermodynamic Carnot efficiency (especially with a reasonable power) is still an unattained goal.

A key idea in this connection is that of trying to obtain a “strongly-coupled” thermoelectric in which there would be a well-defined ratio between electrical and thermal current. Mahan and Sof08 suggested a way to achieve that, for electrons, by funneling the electrical transport to a very narrow energy-band. The deviation from the ideal Carnot efficiency would then be determined by “parasitic” effects, such as the small width of the abovementioned electronic band and the non-electronic thermal conductivity.

For definiteness, we shall consider here cooling in a two-terminal electronic setup, whose terminals are kept at different temperatures, TL and TR, and different chemical potentials, μL and μR. The total entropy production of the electrons in such a device is

\[ \dot{S}_\text{tot} = \frac{\dot{E}_L - \mu_L \dot{N}_L}{T_L} + \frac{\dot{E}_R - \mu_R \dot{N}_R}{T_R} = \frac{\dot{Q}_L}{T_L} + \frac{\dot{Q}_R}{T_R}. \]  

(1)

Here, \( \dot{E}_{L,R} \) is the rate of energy change in the L, R reservoir, \( \dot{N}_{L,R} \) is minus the particle current leaving that reservoir, and \( \dot{Q}_{L,R} \) is the heat current entering it. Current conservation implies \( \dot{N}_L + \dot{N}_R = 0 \), and energy conservation requires \( \dot{E}_L + \dot{E}_R = 0 \). Consequently,

\[ \dot{S}_\text{tot} = J^Q_L \left( \frac{1}{T_R} - \frac{1}{T_L} \right) + J^Q_L (\mu_L - \mu_R), \]  

(2)

where \( J^Q_L \equiv -\dot{E}_L + \mu_L \dot{N}_L \) is the heat current and \( J_L \equiv -\dot{N}_L \) is the particle current leaving the left reservoir.

Assuming for concreteness that the left terminal is to be cooled, then \( T_L - T_R < 0 \) and the working conditions for a thermoelectric refrigerator configuration are positiveness of the used power \( W \)

\[ W = \frac{\mu_L - \mu_R}{e} c J_L \equiv V J_L > 0, \]  

(3)

(\( e \) being the charge of the carriers and \( V \) is the voltage drop) and positiveness of the cooling power \( P \),

\[ P \equiv J^Q_L > 0. \]  

(4)

The efficiency \( \eta \) of the cooling process (sometimes called “coefficient of performance”) is defined as the ratio between the cooling power and the used one; it is advantageous to express it in terms of the entropy production...
It modifies this conclusion.

for a junction obeying time-reversal symmetry; breaking and the cooling power will also vanish. This happens true, but, as is well-known, in that limit both the used lead to a full Carnot efficiency, an interesting question in Sec. III.

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It is found that the dissipation occurs symmetrically in the two reservoirs only in the limit of full electron-hole symmetry (i.e., when the energy-dependent conductivity or transmission of the junction is symmetric around the average chemical potential) and then the thermopower vanishes. The larger is this asymmetry (and the ther-
mpower) the more asymmetric is the distribution of the dissipation between the two reservoirs. Thus the thermopower and the asymmetry of the dissipation are mono-
tonic functions of one another! We will come back to this interesting question in Sec. III

Equation (5) suggests that a vanishing dissipation can lead to a full Carnot efficiency, $\eta = \eta_C$. This is indeed true, but, as is well-known, in that limit both the used and the cooling power will also vanish. This happens for a junction obeying time-reversal symmetry; breaking it modifies this conclusion. One might be tempted to consider the possibility that the two terms forming the dissipation, Eq. (2), cancel one another while each remains finite. This might have been possible, since the thermal part of the dissipation, $J_L^D(T_L - T_R)/T_L$, is in fact negative (the cooling heat current flows against the temperature drop). However, the above is only apparent: at the zero-dissipation limit all currents and input/output powers vanish. One might have hoped to overcome this caveat by breaking time-reversal symmetry; however, at least in a Landauer-type formulation, it has been shown that there are in fact strong limitations (beyond the Carnot one) on $\eta$. Nevertheless, if one considers, as we shall do here, a system very close (characterized by a small parameter $\zeta$) to the ideal limit of Carnot efficiency, both the used and the cooling powers will vanish proportional to $\zeta$, while the dissipation will vanish like $\zeta^2$.

To appreciate this point, it is instructive to relate the dissipation to the properties of the Onsager matrix $\mathcal{M}$, which connects the currents and the thermodynamic “driving” forces (sometimes called affinities),

$$
\begin{bmatrix}
\frac{eJ_F}{J_L^D} \\
\frac{\mu_L - \mu_R}{eT_L}
\end{bmatrix}
= \mathcal{M} \left[
\begin{bmatrix}
\frac{\eta_C - 1}{eT_L} \\

\end{bmatrix}
\right].
$$

In the linear-response regime $\mathcal{M}$ does not depend on the driving forces and is determined by the equilibrium properties of the setup. (For a system invariant to time-reversal the matrix $\mathcal{M}$ is symmetric.) The dissipation, Eq. (2), can be written in the form

$$
T_R \hat{S}_{\text{tot}} = \left[
\begin{bmatrix}
\frac{\mu_L - \mu_R}{e} \\

\end{bmatrix}
\right] \mathcal{M} \left[
\begin{bmatrix}
\frac{\eta_C - 1}{eT_L} \\

\end{bmatrix}
\right].
$$

As the Onsager matrix $\mathcal{M}$ is positive definite, it follows that the dissipation vanishes when the lowest eigenvalue of $\mathcal{M}$ is zero. A $2 \times 2$ symmetric matrix whose determinant is zero is necessarily of the form

$$
\mathcal{M} \propto \begin{bmatrix}
1 & \alpha \\
\alpha & \alpha^2
\end{bmatrix}
$$

Such a matrix implies that the charge and the heat currents are proportional to one another. This was termed by Kedem and Caplan “strong coupling”. The strong-coupling configuration is the limit of the “best thermoelectric” of Mahan and Sofo. Thus, if one could achieve the ideal limit of Refs. 3 and 8, the “driving force” vector $\{((\mu_L - \mu_R)/e, -\eta_C^{-1})\}$ would be proportional to $(-\alpha, 1)$. One would then get the Carnot efficiency, but both currents would vanish, along with zero power in the linear-response regime.

Mahan and Sofo achieved the strong-coupling configuration by channeling the electronic transport into an infinitely-narrow band. There, (see also Ref. 3) the Onsager matrix (again limited to electronic processes) takes the form

$$
\frac{\mathcal{M}}{T} = G \begin{bmatrix}
1 & E/e \\
E/e & E^2/e^2
\end{bmatrix},
$$

where $T$ is the common temperature of the setup, $G$ is the conductance, and $E$ is essentially the unique, well-defined, energy transferred by each charge carrier between the two terminals. In real life there are always “parasitic” effects, such as phonon heat conductivity, transport in a finite-width energy band, more energy channels, etc. As we have now just discovered, such “parasitics” can be crucial for the operation of thermoelectric heat conversion! One is thus led to consider what these parasitics do to the dissipation, to the power, and to the efficiency.

From this line of reasoning it follows that we need to consider the situation when the (nonnegative) determinant of the Onsager matrix is positive, but still very small. In the configuration of Refs. 3 and 8 the two-terminal setup is further connected to a non-electronic...
which "measures" the distance of the determinant from forces at small dissipation, we introduce a parameter its relation to the thermopower section II A. The question of the symmetry of the dissipation of the eigenvectors of the Onsager matrix is presented in The analysis of the optimum driving-force vectors and that parameter are different: the total dissipation is affected far less than the efficiency.

II. CALCULATIONS AND RESULTS

As is explained above, the dissipation can vanish when the determinant of the Onsager matrix $\mathcal{M}$ is zero. In order to study the efficiency and the optimal driving forces at small dissipation, we introduce a parameter which "measures" the distance of the determinant from the strong-coupling limit condition

$$\zeta = \sqrt{1 - \frac{\mathcal{M}_{12}\mathcal{M}_{21}}{\mathcal{M}_{11}\mathcal{M}_{22}}}, \quad \zeta \geq 0.$$  \hfill (11)

When time-reversal symmetry holds the Onsager matrix is symmetric, and $\zeta = (1 + (zT))^{-1/2}$, where $zT$ is the figure of merit. In that case $0 \leq \zeta \leq 1$. These bounds on $\zeta$ persist also when $\mathcal{M}$ is not symmetric, with $\mathcal{M}_{12}/\mathcal{M}_{21} > 0$. However, when that ratio is negative (the example considered below will be $\mathcal{M}_{21} < 0$) then $\zeta$ exceeds 1. Namely, a setup lacking time-reversal symmetry and for which $\mathcal{M}_{12}/\mathcal{M}_{21} < 0$ is always far away from the strong-coupling limit.

Adding a notation for the deviation of $\mathcal{M}$ from complete symmetry

$$\gamma_a = \sqrt{\frac{\mathcal{M}_{12}/\mathcal{M}_{21}}{\mathcal{M}_{11}/\mathcal{M}_{22}}}, \quad \gamma_a \geq 0.$$  \hfill (12)

the positiveness of the Onsager matrix, $\mathcal{M}_{11} \mathcal{M}_{22} - (\mathcal{M}_{12} + \mathcal{M}_{21})^2/4 \geq 0$, imposes a relation between $\zeta$ and the asymmetry parameter $\gamma_a$

$$\sqrt{\frac{1 - \zeta}{1 + \zeta}} \leq \gamma_a \leq \sqrt{\frac{1 + \zeta}{1 - \zeta}}.$$  \hfill (13)

[For a time-reversal invariant system, $\gamma_a = 1$ and condition (13) is always satisfied.] With this parametrization of the Onsager matrix, to which we add for convenience

$$\gamma = \sqrt{\mathcal{M}_{11}/\mathcal{M}_{22}},$$  \hfill (14)

we obtain for the cooling power

$$P = \frac{\eta_C^{-1}\mathcal{M}_{21}}{T_R} \left( (\eta_C V) - s \frac{\gamma_a/\gamma}{\sqrt{1 - \zeta^2}} \right),$$  \hfill (15)

where $s \equiv \text{sgn}[\mathcal{M}_{21}]$, and for the used power

$$W = \frac{\eta_C^{-2}\mathcal{M}_{11}}{T_R} \left( (\eta_C V)((\eta_C V) - \frac{\gamma_a}{\gamma} \sqrt{1 - \zeta^2}) \right).$$  \hfill (16)

Hence, when $\mathcal{M}_{21}$ is positive, both used and cooling powers are positive for

$$\langle \eta_C V \rangle \geq \frac{\gamma_a/\gamma}{\sqrt{1 - \zeta^2}},$$  \hfill (17)

while if it is negative then the voltage drop $V$ must be negative as well, with

$$-\langle \eta_C V \rangle \geq \frac{\gamma_a/\gamma}{\sqrt{\zeta^2 - 1}}.$$  \hfill (18)

Finally, the efficiency is

$$\eta_C = \frac{s \sqrt{1 - \zeta^2}}{\gamma_a \sqrt{\mathcal{M}_{11}/\mathcal{M}_{22}}} \left( (\eta_C V) - s \frac{\gamma_a/\gamma}{\sqrt{1 - \zeta^2}} \right).$$  \hfill (19)

The efficiency is optimized for a given $\eta_C$, by controlling $V \eta_C$, the ratio of the two components of the driving-force vector. To do that, we first show in Fig. II graphs of the efficiency $\eta/\eta_C$ as a function of $V \eta_C$ in the whole range, and with $\gamma_a = 1$, for which the physical bound on $V \eta_C$ is given in Eq. (17). The mathematical function of the efficiency has a "pole" at $V \eta_C = \sqrt{1 - \zeta^2}/\gamma$ and a zero at $V \eta_C = (\gamma \sqrt{1 - \zeta^2})^{-1}$. It is large in the non-physical region on the left of the pole and negative between the pole and the zero. We emphasize that although the efficiency can be larger in the non-physical regions, we have to choose the maximum within the physical domain, given by Eq. (17). The plots in Fig. II help us to do that. The physical region is on the right of both zero and pole.
When the system tends to the “ideal” limit \( \zeta \to 0 \), the zero and the pole merge together and the maximum of the efficiency, \( \eta/\eta_C \to 1 \) is infinitesimally above \( V\eta_C = 1/\gamma \). This rather nontrivial behavior is clearly seen in Fig. 1a, and Eqs. 15 and 16 show how both the cooling and used powers approach zero at the ideal point. The pole and the zero get further apart as \( \zeta \) increases, Figs. 1b and 1c, and the value of \( V\eta_C \) for which the efficiency is maximal increases as well (see below).

The efficiency is optimal for a given \( \eta_C \) at \( V = V_m \), where

\[
V_m\eta_C = \frac{\gamma_a}{\gamma} \sqrt{\left|\frac{1 + \zeta}{1 - \zeta}\right|},
\]

showing that the sign of the bias is determined by the \( s \equiv \text{sgn}M_{21} \), as expected. At the voltage drop corresponding to the optimal efficiency, the cooling power is

\[
P_m = \frac{\eta_C^{-1} |M_{21}| \gamma_a}{T_R} \frac{\zeta}{\gamma \sqrt{1 - \zeta^2}},
\]

the used power is

\[
W_m = \frac{\eta_C^2 M_{11} \gamma_a^2}{T_R^2} \frac{1 + \zeta}{1 - \zeta},
\]

and the optimal efficiency \( \eta_m \) is

\[
\eta_m = \eta_C \frac{1}{\gamma_a^2} \frac{1 - \zeta}{1 + \zeta}.
\]

Note that the optimal efficiency never exceeds the Carnot one: this is ensured by the condition (13). Finally, the total dissipation at optimal efficiency is

\[
T_R \dot{S}_m = \left(W_m - \eta_C^{-1} P_m\right) = \eta_C^2 M_{22} - \frac{\zeta}{1 - \zeta}\left(\gamma_a^2(1 + \zeta) - (1 - \zeta)\right).
\]

Hence we see that as a time-reversal symmetric junction \( (\gamma_a = 1) \) approaches the strong-coupling limit, its efficiency, cooling power, and the used power all deviate from the ideal limit as \( \zeta \); the total dissipation, however, deviates much more weakly from zero, as \( \zeta^2 \), due to the cancellation of the \( \zeta \) term between the thermal and the electrical dissipations.

The effect of time-reversal symmetry breaking (for a positive ratio of the two off-diagonal elements of the matrix \( \mathcal{M} \)) is examined in Figs. 2. The figures show the dependence of the efficiency on the driving force in the physical region of the latter [see Eq. (17)]. The four curves in each panel are for various values of the asymmetry parameter \( \gamma_a \), according to condition (13), with the efficiency decreasing as \( \gamma_a \) is increasing [see Eq. (23), for the parameters used here]. These show that increasing \( \gamma_a \) is detrimental to the coefficient of performance of cooling, the more so as the deviation from the strong-coupling limit increases. This asymmetry has another outcome. Whereas for \( \gamma_a = 1 \) a subtle cancellation of the order \( \zeta \)-terms in the dissipation causes it to deviate less from the ideal limit as compared to the powers, this ceases to hold as \( \gamma_a \neq 1 \). This is a notable result of our analysis.

A. Analysis of the eigenvalues and eigenvectors of the Onsager matrix with time-reversal symmetry, \( (\gamma_a = 1) \).

It is interesting to study the behavior of the driving-force vector close to the ideal limit, where the dissipation is very small. As the only variable is \( \eta_C V \), it suffices to consider just the ratio of the first to the second component of the eigenvectors. Writing the Onsager matrix for \( \gamma_a = 1 \) in the parametrized form (for brevity, we take both off-diagonal elements of \( \mathcal{M} \) to be positive)

\[
\mathcal{M} = \begin{bmatrix}
\gamma & \sqrt{1 - \zeta^2} \\
\sqrt{1 - \zeta^2} & 1/\gamma
\end{bmatrix},
\]

FIG. 1: The mathematical function of the efficiency, Eq. (19), as a function of \( V\eta_C \), for \( \gamma_a = 1, \gamma = 10, \) and \( s = 1 \), for various values of \( \zeta \). (a)-(c) \( \zeta = 0.01, \) (b)-(c) \( \zeta = 0.1, \) and (c)-(c) \( \zeta = 0.5. \)
We have found that at optimal efficiency our driving-force vector is [see Eq. (20)]

\[ v_m \propto \frac{1}{\gamma} \sqrt{\frac{1}{1 - \zeta} - \frac{\zeta}{1 - \zeta}} \cdot \frac{1}{1 - \zeta} \cdot \frac{1}{1 - \zeta} \].

Hence \( v_m \) is directed along the eigenvector belonging to the lower eigenvalue at \( \zeta \to 0 \), and rotates away from it as the Onsager matrix deviates from the ideal strong-coupling limit.

**III. DISCUSSION OF THE ASYMMETRY OF THE DISSIPATION VS. THE THERMOPower**

The treatment of the heat exchange between transport electrons and an electron reservoir is based on the principle that a particle with an energy \( E \) exiting (entering) a large system extracts from (delivers to) it a heat \( \Delta Q \) and an entropy \( \Delta S \) given by

\[ \Delta Q = (E - \mu); \quad \Delta S = (E - \mu) / T, \]

where \( \mu \) is the chemical potential in the reservoir. This is proven thermodynamically, for example, in Ref. [17], adopted to the mesoscopic regime within the Landauer-transport formulation in Ref. [18] and given a statistical-mechanics proof in Ref. [19]. For two reservoirs with chemical potentials \( \mu_L \) and \( \mu_R \) connected by a single-channel “wire” having a transmission coefficient \( T(E) \), this yields [18] for the heat

\[ -J_{\alpha}^Q = \Delta Q = T \Delta S, \]

delivered to reservoir \( \alpha \) (\( \alpha = L \) or \( R \))

\[ J_{\alpha}^Q = \frac{2}{R} \int_{-\infty}^{\infty} (\mu_\alpha - E) \frac{f_\alpha(E) - f_\beta(E)}{T} dE, \]

with \( \beta \neq \alpha \) and \( f_{L,R}(E) \) being the corresponding Fermi function. We limit ourselves to the linear-response regime with a single driving field \( V = (\mu_L - \mu_R) / e \). The electrical current between the reservoirs gives the conductance \( G \) and the heat current gives the Onsager coefficient \( \mathcal{L} = \mathcal{M}_{21} / T \), [cf. Eq. (7)]. The Seebeck thermopower \( S \) is then given by \( S = \mathcal{L} / (GT) \). We consider here only the case where time-reversal symmetry prevails.

The first crucial observation here from Eq. (30) is the well-known one that in order to have nonzero thermoelectric response, \( T(E) \) must have an odd component as a function of \( E - \mu \). Next, we note that if such “electron-hole (\( e - h \))” symmetry breaking” exists, the two powers \( J_{L,R}^Q \) are each linear in \( V \). However, by adding them, one finds that the total dissipation (heat generated per unit time) is indeed (see Ref. [18]) the good old Joule heating, \( IV \), which is second-order in \( V \) (\( I \) is the net electrical current).

The novel observation of Ref. [12] is that it is the same symmetry breaking which also leads to an asymmetry of the dissipation between the two reservoirs. In fact, by
subtracting Eq. (30) for $L$ and $R$, one readily obtains, to order $V$,

$$J_R^L(V) - J_R^Q(V) = 2LV = 2GSTV.$$  \hspace{1cm} (31)

Thus the dissipation asymmetry and the thermopower are proportional to one another, at least within linear response. The physical reason for this is clear: within linear response, the energy flow from, say, $L$ to $R$, which differs by just a constant from the heat flow, can be regarded as less energy (and heat) dumped in $L$, and more dumped in $R$. This transfers energy from $L$ to $R$.

An interesting feature is that the heat flow/dissipation asymmetry, while small for weak $e - h$ symmetry breaking, is first order in $V$. The total dissipation, which exists also with that symmetry valid, is second order—i.e. much smaller! As shown in Ref. 12, this thermopower—dissipation-asymmetry relationship can be used to determine the former quantity by measuring the latter. We note that when the Seebeck coefficient, $S = 0$, vanishes the dissipation is symmetric and independent of the nature of the two reservoirs (and of any asymmetry between them).

Although in linear response the currents are odd in $V$, the dissipation in each reservoir is not. In fact, by subtracting Eq. (30) for $L$, with $V$ and $-V$, it is found\(^\text{12}\) that, e.g.,

$$J_L^Q(-V) - J_L^Q(V) = 2LV = 2GSTV,$$  \hspace{1cm} (32)

as well. These relationships should hold for any meso- and nano-scale system, including molecular bridges. They were confirmed by both electronic-structure numerical computations and by experiments using an innovative nano-scale temperature detection.\(^\text{12}\)

We conclude by discussing some simple particular cases, demonstrating the relevance to our main subject in the present paper—the case of small dissipation. Having full $e - h$ symmetry is of no interest here, since both $S$ and the dissipation asymmetry vanish. A very interesting case is that a almost full $e - h$ symmetry breaking, when almost all charge carriers are e.g. electrons. A simple realisation of this is a narrow Lorentzian resonance of width $\Gamma$ much above the Fermi level (i.e when its center energy $E_0$ satisfies $E_0 - \mu \gg \Gamma, k_B T$). In the low-temperature and narrow-resonance limit this becomes the strong-coupling configuration\(^\text{2}\) or ‘best’\(^\text{2}\) thermoelectric, having a singular transport matrix. The system then has vanishing dissipation and power. The new insight gained from the thermopower–dissipation-asymmetry relationship is that the dissipation is negative in one terminal and positive in the other, such that they fully cancel each other (while heat is still carried by the electron current from $L$ to $R$).

A finite width of the resonance, or a low but significant temperature will create the almost singular transport matrix discussed in this paper. A more complicated case is that of a mobility edge far above the Fermi level. Again, most charge carriers will be electrons. This case was treated in Refs. 18 and 19, and sizeable values of the Seebeck coefficient $S$ were indeed found. This entails large dissipation asymmetry.

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