Irreversible thermodynamics of thermoelectric devices: From local framework to global description

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Thermoelectricity is traditionally explained via Onsager’s irreversible, flux-force framework. The coupled flows of heat and electric charge are modeled as steady-state flows, driven by the thermodynamic forces defined in terms of the gradients of local, intensive parameters like temperature and electrochemical potential. A thermoelectric generator is a device with a finite extension, and its performance is measured in terms of total power output and total entropy generation. These global quantities are naturally expressed in terms of discrete or global forces derived from their local counterparts. We analyze the thermodynamics of thermoelectricity in terms of global flux-force relations. These relations clearly show the additional quadratic dependence of the driver flux on global forces, corresponding to the process of Joule heating. We discuss the global kinetic coefficients defined by these flux-force relations and prove that the equality of the global cross-coefficients is derived from a similar property of the local coefficients. Finally, we clarify the differences between the global framework for thermoelectric energy conversion and the recently proposed minimally nonlinear irreversible thermodynamic model.

I. INTRODUCTION

In many transport phenomena, interesting effects arise due to coupling between different thermodynamic forces present simultaneously in the system. Thermoelectricity provides a physically transparent picture of this coupling between flows of heat and electric charge, which gives rise to the well-known Seebeck, Peltier, and Thomson effects [1–5]. These interference phenomena can be successfully described within the linear flux-force formalism of Onsager [1]. The aforesaid linear relationship between fluxes and forces derives from the usually small magnitudes of the thermodynamic forces driving the system. The latter are expressed as gradients of the locally defined intensive parameters and this framework invokes the local-equilibrium hypothesis [2].

Although, the thermodynamic explanation of the above phenomena treats them as steady-state processes at the local level, actual devices have macroscopic extensions and so their performance needs to be analyzed by scaling up the local description. This leads to the study of global quantities like total power output and total entropy generation by the device. Due to the increasing worldwide demand for efficient and environment-friendly energy convertors, many works have pursued optimization of the material properties as well as power output/cooling power of thermoelectric devices [6–19]. It is noteworthy that a local, linear-irreversible model gives rise to nonlinear (quadratic) dissipation terms in the flux equations at the global level [20]. In thermoelectricity, Joule heating plays the role of this dissipation term. Thus, despite the apparent presence of nonlinearities in thermal flux equations, the locally linear character of the underlying framework for thermoelectricity has been emphasized [20 21].

With the advent of finite-time thermodynamics, the flux-force framework for irreversible phenomena has been extended to macroscopic heat devices too [22–24], where the fluxes still are linear functions of the global or discrete forces. Further, the so-called minimally nonlinear irreversible thermodynamic (MNLIT) model [25, 26] proposes generalized heat flux equations by incorporating a nonlinear, phenomenological term at the global level, akin to the dissipation term in thermoelectric devices. This model may be adapted for both autonomous as well as cyclic heat devices operating in finite time [27 28].

In this paper, our focus is on the thermoelectric flux-force relations at the global level. As with the standard Onsager-Callen framework, consideration of the total rate of entropy generation as a bilinear form leads to the identification of global thermodynamic forces. We show how the scale-up from a local description naturally leads us to express the driven flux as a linear function of these global or discrete forces. In consequence, we will see that the driven flux additionally follows a nonlinear dependence on these forces, equivalent to the presence of a nonlinear dissipation term discussed above. We also define global kinetic coefficients from these flux relations. A hallmark of the underlying local framework is the equality of kinetic cross-coefficients based on principle of microscopic time reversibility [29]. We observe a similar property of the global kinetic coefficients, which can be traced to its local counterpart.

The plan of the paper is as follows. In Section II, Onsager-Callen framework for a thermoelectric generator is discussed. In Section III, we identify the discrete or global forms of forces and express the fluxes in terms of these forces. We also obtain expressions for the global kinetic coefficients and prove Onsager reciprocity at the global level. In Section IV, our approach is compared with the MNLIT model. Finally, we conclude in Section V.
Thermodynamics. By defining the electric current density \( \vec{J} \) in order to satisfy the second law of thermodynamics, the local rate of entropy production per unit volume, \( \dot{s} \), is defined as the divergence of the entropy flux \( \vec{J}_s = -\vec{\nabla}s/T \), where \( \mu \) and \( T \) are the local chemical potential and the temperature, respectively. Assuming small magnitudes for such affinities, Onsager expressed the local fluxes as linear combination of these affinities:

\[
\begin{pmatrix}
\vec{J}_N \\
\vec{J}_Q
\end{pmatrix} = \begin{pmatrix}
L_{11} & L_{12} \\
L_{21} & L_{22}
\end{pmatrix} \begin{pmatrix}
-\vec{\nabla}\mu/T \\
-\vec{\nabla}T/T^2
\end{pmatrix},
\]

where the Onsager coefficients, \( L_{ij} \), obey certain conditions \( \text{[1]} \) in order to satisfy the second law of thermodynamics. By defining the electric current density \( \vec{J} = e\vec{J}_N \), and noting that \( \vec{\nabla}\mu = e\vec{\nabla}V \), where \( e \) is the charge on electron and \( V \) is the electrostatic potential, we can write \( \text{[1]} \) as

\[
\begin{pmatrix}
\vec{J} \\
\vec{J}_Q
\end{pmatrix} = \begin{pmatrix}
e^2L_{11} & eL_{12} \\
eL_{21} & L_{22}
\end{pmatrix} \begin{pmatrix}
-\vec{\nabla}V/T \\
-\vec{\nabla}T/T^2
\end{pmatrix}.
\]

It is known that the knowledge of Onsager coefficients \( L_{ij} \) is equivalent to the knowledge of the quantities \( \rho, \kappa \) and \( \alpha \), whereby we have the following expressions \( \text{[2]} \):

\[
\begin{align*}
L_{11} &= \frac{T}{\rho e^2}, \\
L_{12} &= \frac{\alpha T^2}{\rho e} = L_{21}, \\
L_{22} &= T^2 \left( \frac{\alpha^2 T + \kappa}{\rho} \right).
\end{align*}
\]

Note that the cross-coefficients are equal by virtue of the principle of microscopic reversibility \( \text{[1]} \). Secondly, even if the quantities \( \rho, \kappa, \) and \( \alpha \) are assumed to be constant, the Onsager coefficients depend on local temperature \( T \).

III. FROM LOCAL TO GLOBAL AFFINITIES

Now, the Onsager-Callen framework was formulated for the steady-state description of irreversible phenomena in terms of forces and fluxes at the local level. In this section, we analyze how the flux-force relations are extended to the macroscopic level i.e. applied to the full length of the thermoelectric material.

The local rate of entropy production per unit volume, \( \dot{s} \), is defined as the divergence of the entropy flux \( \vec{J}_s = -\vec{\nabla}s/T \), so that \( \dot{s} = \vec{\nabla}.(\dot{Q}_Q/T) \), which can be written as the sum of products of fluxes and conjugate forces, as follows.

\[
\dot{s} = \vec{J}.\left(-\frac{\vec{\nabla}V}{T} \right) + \vec{J}_Q(x) \vec{\nabla}\left(\frac{1}{T}\right),
\]

where we have used \( \text{[2]} \)

\[
\vec{\nabla}.\vec{J}_Q = -\vec{J}.\vec{\nabla}V.
\]

Then, the rate of total entropy production per unit area (p.u.a) along the whole length of the material (1-d case) is \( \dot{S} = \int_0^L \dot{s} \, dx \), and given by

\[
\dot{S} = \frac{J_Q(l)}{T_c} - \frac{J_Q(0)}{T_h},
\]

where \( x = 0(l) \) corresponds to the hot (cold) end of the material.

Now, the global power output p.u.a is given by

\[
\mathcal{P} = -\int_0^L \vec{\nabla}.\vec{J}_Q \, dx, \quad \text{(7)}
\]

\[
= J_{Q}(0) - J_{Q}(l). \quad \text{(8)}
\]

Further, the use of Eq. \( \text{[3]} \) in \( \text{[7]} \), and the constant magnitude \( |\vec{J}| \equiv J \) implies that

\[
\mathcal{P} = J(V_c - V_h) \equiv J\Delta V. \quad \text{(9)}
\]

From Eqs. \( \text{[8]} \) and \( \text{[9]} \), we can write Eq. \( \text{[6]} \) as

\[
\dot{S} = J \left( -\frac{\Delta V}{T_c} \right) + J_{Q}(0) \left( \frac{1}{T_c} - \frac{1}{T_h} \right).
\]

Figure 1. Schematic of thermoelectric heat engine. Two legs (yellow online) of a constant properties thermoelectric material (TEM) in simultaneous contact with a hot \((x = 0)\) and a cold \((x = l)\) reservoir. Electric power, \( P = Q_h - Q_c \), is drawn against an external load.
Thus, the global rate of entropy production p.u.a is expressed in a bilinear form, \( \dot{S} = J_1 X_1 + J_2 X_2 \), where the corresponding flux-force pairs are identified as follows.

\[
\begin{align*}
J_1 &= J, \\
X_1 &= -\frac{\Delta V}{T_c}, \\
J_2 &= J_Q(0), \\
X_2 &= 1 \frac{\Delta V}{T_c - 1 T_h}.
\end{align*}
\]

Comparing Eqs. (4) and (10), we observe how the pair of local affinities take up a discrete form, relevant for the global description. It is interesting to analyze this bilinear form further from irreversible thermodynamic point of view. For future comparison, we consider the rate of total entropy production, \( \dot{S} = J \Delta A + J_2 X_2 \), where

\[
\begin{align*}
J_1 &= I, \\
X_1 &= -\frac{\Delta V}{T_c}, \\
J_2 &= \dot{Q}_h, \\
X_2 &= 1 \frac{\Delta V}{T_c - 1 T_h}.
\end{align*}
\]

with \( I = J \Delta A \) and \( \dot{Q}_h = J_Q(0) \Delta A \).

A. Flux-force relations at the global level

Corresponding to linear flux-force relations at the local level, we now enquire into the relations between fluxes and forces at the macroscopic level. Note that the local flux-force relations are postulated to be linear within Onsager’s approach. Here, we wish to see how the fluxes, \( J_i \), in Eqs. (13) and (14), are expressed in terms of the global or discrete affinities, \( X_i \).

Firstly, from Eqs. (2) and (3), we can write

\[
\dot{J} = \frac{1}{\rho} \nabla V - \frac{\alpha}{\rho} \nabla T.
\]

For the case of a 1-d thermoelectric element, we have

\[
J = -\frac{1}{\rho} \frac{\partial V}{\partial x} - \frac{\alpha}{\rho} \frac{\partial T}{\partial x}.
\]

Thus, we have expressed the fluxes \( (J_1, J_2) \) in terms of the global affinities \( (X_1, X_2) \). Whereas \( J_1 \) is linear in the global forces, \( J_2 \) is explicitly non-linear in these forces. Alternately, nonlinearity lies in the quadratic dissipation term in Eq. (22), due to Joule heating [21].

Now, since the current density \( J \) as well as \( \rho \) and \( \alpha \) are constant along the length of the material, integrating the above equation over \( x \in [0, l] \), we obtain:

\[
J I = -\frac{\Delta V}{\rho} + \frac{\alpha}{\rho} \Delta T.
\]

In the above, \( \Delta V = V_e - V_h > 0 \), whereas \( \Delta T = T_h - T_c > 0 \). Since \( J = I/\Delta A \) and the resistance of the material is \( R = \rho/\Delta A \), we can write Eq. (17) as

\[
I = -\frac{\Delta V}{R} + \frac{\alpha}{R} \Delta T.
\]

which can be rewritten in the form

\[
J_1 = \frac{T_c}{R} X_1 + \frac{\alpha T_c T_h}{R} X_2,
\]

following the definitions in Eqs. (13) and (14).

Thus, we note that the flux \( J_1 = I \) is linear in the discrete forces \( X_1 \) and \( X_2 \).

Similarly, from Eqs. (2) and (3), the local thermal flux inside the thermoelectric material is given by:

\[
J_Q(x) = -\frac{\alpha T(x)}{\rho} \frac{\partial V}{\partial x} - \left( \frac{\alpha^2 T(x)}{\rho} + \kappa \right) \frac{\partial T}{\partial x},
\]

which can be cast in the following form [21]:

\[
J_Q(x) = \alpha T(x) J + \kappa \frac{\Delta T}{l} - \frac{\rho (l - 2x) J^2}{2}.
\]

Then, thermal currents at the end points of the thermoelectric material are evaluated to be

\[
\dot{Q}_h = \alpha T_h I + K \Delta T - \frac{1}{2} RI^2,
\]

\[
\dot{Q}_c = \alpha T_c I + K \Delta T + \frac{1}{2} RI^2,
\]

where \( \dot{Q}_c = J_Q(l) \). Also, \( K = \kappa A/l \) is the thermal conductance. Then, the total power output of thermoelectric generator, \( P = \dot{Q}_h - \dot{Q}_c \) is given by: \( P = \alpha \Delta T I - RI^2 \).

We close this subsection by expressing the thermal flux \( J_2 \equiv \dot{Q}_h \) in terms of the forces \( (X_1, X_2) \). Substituting for \( I = J_1 \) from Eq. (19) into Eq. (22), we obtain

\[
J_2 = \frac{\alpha T_c T_h}{R} X_1 + \left( \frac{\alpha^2 T_c}{R} + K \right) T_h T_c X_2 - \frac{T^2_c}{2R} X_1^2 - \frac{\alpha \alpha^2 T_c^2 T_c}{2R} X_2^2 - \frac{\alpha T_c T_h^2}{R} X_1 X_2.
\]

B. Global kinetic coefficients

We may formally define a set of kinetic coefficients by assuming expansion of the fluxes in terms of the global
affinities, as follows. We write
\[ J_1 = \mathcal{L}_{11} X_1 + \mathcal{L}_{12} X_2, \quad (25) \]
\[ J_2 = \mathcal{L}_{21} X_1 + \mathcal{L}_{22} X_2 + \mathcal{O}[X_j^2]. \quad (26) \]

Then, comparing Eq. (25) with (19), and Eq. (26) with (24), we obtain
\[ \mathcal{L}_{11} = \frac{T_c}{R}, \]
\[ \mathcal{L}_{12} = \frac{\alpha T_c T_h}{R} = \mathcal{L}_{21}, \quad (27) \]
\[ \mathcal{L}_{22} = T_c T_h \left( \frac{\alpha^2 T_h}{R} + K \right). \]

Additionally, we have higher-order coefficients related to the quadratic terms in Eq. (24). The above kinetic coefficients may be compared with their local counterparts in Eq. (3). Now, here we also observe the equality of the cross-coefficients. Recall, that the corresponding equality at the local level can be argued on the basis of microscopic time-reversibility. Interestingly, the equality at the macroscopic level can also be traced to the equality at the local level, as we show below.

From Eq. (2), we have
\[ \tilde{J}_Q(x) = \frac{L_{21}}{eL_{11}} \tilde{J} - \frac{D}{L_{11} T^2} \nabla T, \quad (28) \]
where \( D = L_{11} L_{22} - L_{12} L_{21} \). If we do not invoke Eqs. (3), then, equivalent to Eq. (22), we can write
\[ Q_h = \frac{L'_{21}}{eL_{11}} I + \frac{D'A}{L_{11} T_h^2} \Delta T - \frac{T_h}{2e^2 L_{11}^2} A^2, \quad (29) \]
where the primed Onsager coefficients are evaluated at \( x = 0 \), or \( T = T_h \). Alternately, in terms of the forces \( X_i \), we can write \( J_2 = \tilde{Q}_h \) as
\[ J_2 = \frac{L'_{21} T_c}{eL_{11} R} X_1 + T_c T_h \left( \frac{D'A}{L_{11} T_h^2} I + \frac{\alpha L'_{21} T_c}{eL_{11} R} \right) X_2 + \mathcal{O}[X_j^2]. \quad (30) \]

Comparing the above equation with Eq. (26), we identify \( L_{21} = L_{21} T_c/eL_{11} R \), which turns out to be equal to \( L_{12} \), upon substituting from Eq. (3). Thus, we see that the equality \( L_{21} = L_{12} \) is consistent with the equality of Onsager coefficients (\( L_{21} = L_{12} \)) at the local level.

V. CONCLUDING REMARKS

In the above, we have analyzed the flux-force relations for a thermoelectric generator at global level, by scaling up from the local, linear-irreversible thermodynamic framework of Onsager and Callen. An important step is the identification of global affinities or thermodynamic forces and fluxes from the bilinear expression for the rate of total entropy generation. The flux-force relation is linear in case of the driven flux (electric current) while it is quadratic in the global forces for the driven flux (hot thermal flux). The nonlinear dissipation terms are known to be due to Joule heating. Further, these flux-force relations also yield expressions for the effective kinetic coefficients. We observe the equality of cross-coefficients and show that this is derived from the Onsager reciprocity at the local level. Although, the traditional picture of thermoelectricity is adequate to explain the global features of power generation and so on, we believe the global flux-force relations expressed in terms of discrete or global equivalent of the local forces, is also a valid depiction of the same phenomenon. Our analysis also clarifies the comparison with the other nonlinear phenomenological models such as MNLIT model. One can similarly formulate the global relations in the presence of magnetic field which breaks the time-reversal symmetry [24]. As may be expected, the global cross-coefficients are also not equal in this case. Finally, in the present paper, we have assumed ideal thermal contacts between the thermoelectric material and the reservoirs. In principle, one can also
include the effect of finite-conductance heat exchangers in the performance analysis \[6\]. It will be interesting, albeit more involved, to investigate the global force-flux picture in the presence of internal as well as external irreversibilities.

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**Appendix A: Choice of fluxes and affinities**

We recall that the choice of fluxes and conjugate affinities is not unique within the Onsager framework \[2\]. Thus, if the particle current density \(J_N\) and thermal current density \(J_Q\) are the fluxes of our description, then \(- (\nabla \mu)/T\) and \(\nabla (1/T)\), respectively, are the affinities. On the other hand, if \(J_N\) along with the energy current density, defined as: \(J_E(x) = J_Q(x) + \mu(x) J_N\), are chosen as the fluxes, then the corresponding affinities are, \(- \nabla (\mu/T)\) and \(\nabla (1/T)\), respectively. Analogously, the rate of entropy production at local level is given by:

\[
\dot{s} = -J_N \cdot \nabla \left( \frac{\mu}{T} \right) + J_E(x) \cdot \nabla \left( \frac{1}{T} \right),
\]

(A1)

The corresponding global rate of entropy production is then given by:

\[
\dot{S} = J_N \left( \frac{\mu_h}{T_h} - \frac{\mu_c}{T_c} \right) + J_E(0) \left( \frac{1}{T_c} - \frac{1}{T_h} \right),
\]

(A2)

which is in a bilinear form, expressed in terms of the corresponding fluxes and discrete forces.

**Appendix B: Thermoelectric refrigerator**

In this case, local force-flux relations are expressed as:

\[
\begin{pmatrix}
\frac{\nabla}{T}
\end{pmatrix}
\begin{pmatrix}
J
\end{pmatrix}
= \begin{pmatrix}
\frac{e^2 L_{11}}{e L_{21}} & \frac{e L_{12}}{e L_{21}}
\end{pmatrix}
\begin{pmatrix}
\frac{\nabla V}{T}
\end{pmatrix},
\]

(B2)

where the Onsager coefficients in terms of the properties of thermoelectric material have the same expressions as in Eq. \(3\).

Now, the rate of entropy production p.u.a along the 1-d thermoelectric material is given as

\[
\dot{S} = -\frac{J_Q(l)}{T_c} + \frac{J_Q(0)}{T_h},
\]

(B3)

which can be rewritten as

\[
\dot{S} = J \left( \frac{\Delta V}{T_h} \right) + J_Q(l) \left( \frac{1}{T_h} - \frac{1}{T_c} \right).
\]

(B4)

The rate of total entropy production, \(\dot{S} = \dot{S}_A = J_1 X_1 + J_2 X_2\), where

\[
\begin{align*}
J_1 &= I, \quad X_1 = \frac{\Delta V}{T_h}, \\
J_2 &= \dot{Q}_c, \quad X_2 = \frac{1}{T_h} - \frac{1}{T_c}.
\end{align*}
\]

(B5) (B6)

The above fluxes \(J_i\) may be expressed in terms of macroscopic affinities \(X_i\). By using Eqs. \(3\) and \(B2\), we get

\[
\dot{J} = \frac{\nabla V}{\rho} + \alpha \frac{\nabla T}{\rho}.
\]

(B7)

Integrate it over the whole length, i.e, \(x \in [0,l]\), we obtain

\[
I = \frac{\Delta V}{R} - \alpha \Delta T/R, \text{ which can be written as}
\]

\[
J_1 = \frac{T_h}{R} X_1 + \frac{\alpha T_c T_h}{R} X_2.
\]

(B8)

Also, thermal flux \(J_2\) in terms of the global forces is given as

\[
J_2 = \frac{\alpha T_c T_h}{R} X_1 + \left( \frac{2 T_c}{R} - K \right) T_h T_c X_2 + O[X_j^2].
\]

(B9)

It shows that \(J_2\) is not a linear function of the global forces. Next, we compare Eqs. \(B8\) and \(B9\) with Eqs. \(25\) and \(26\) to get global kinetic coefficients:

\[
\begin{align*}
L_{11} &= \frac{T_h}{R}, \\
L_{12} &= \frac{\alpha T_h T_c}{R} = L_{21}, \\
L_{22} &= T_c T_h \left( \frac{2 T_c}{R} - K \right).
\end{align*}
\]

(B10)

These coefficients of thermoelectric refrigerator may be compared with Eq. \(27\) for thermoelectric generator. We observe the difference in the form of diagonal coefficients. Further, it also shows that cross-coefficients preserve their equality which emerges from time-reversal symmetry at the microscopic level.
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