Conservation Laws and Thermodynamic Efficiencies

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We show that generic systems with a single relevant conserved quantity reach the Carnot efficiency in the thermodynamic limit. Such a general result is illustrated by means of a diatomic chain of hard-point elastically colliding particles where the total momentum is the only relevant conserved quantity.

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Conservation laws strongly affect transport properties. Conserved quantities may lead to time correlations not decaying with time, so that transport is not diffusive and is described, within the linear response theory, by diverging transport coefficients. This ideal conducting (ballistic) behavior can be firmly established as a consequence of an inequality by Mazur [1–3] which, for a system of size Λ characterized by M conserved quantities Q_n, $n = 1, \ldots, M$, bounds the time-averaged current-current correlation functions as

$$\lim_{t\to\infty} \frac{1}{t} \int_0^t dt' \langle J(t')J(0)\rangle_T \geq \sum_{n=1}^{M} \frac{\langle JQ_n\rangle_T^2}{\langle Q_n^2\rangle_T}, \tag{1}$$

where $\langle \cdots \rangle_T$ denotes the thermodynamic average at temperature T. The constants of motion, Q_n, are orthogonal to each other, i.e., $\langle Q_n Q_m\rangle_T = \langle Q_n^2\rangle_T \delta_{n,m}$, and *relevant*; that is, $\langle JQ_n\rangle_T \neq 0$ for all n. A nonzero right-hand side in Eq. (1) at the thermodynamic limit implies a finite Drude weight for the current J, which in turn indicates ballistic transport [4,5]. The impact of motion constants on the electric and thermal conductivities has been widely investigated [4–7]. In particular, anomalous heat transport has been discussed for momentum conserving interacting systems in low dimensions [8,9]. However, to the best of our knowledge, conservation laws have never been discussed for coupled flows, in particular, in relation to the problem of optimizing thermodynamic efficiencies.

The search for a new technology capable of reducing the environmental impact of electrical power generation and refrigeration has aroused great interest in thermoelectricity, namely, the possibility to build a type of solid-state heat engine capable of converting heat into electricity, or, alternatively, electricity into cooling [10–14]. The main difficulty is connected to the low efficiency of such heat engines. We recall that the maximum thermoelectric efficiency as well as the efficiency at maximum power [15–19] are determined, within the linear response regime and for systems with time-reversal symmetry [20], by the so-called figure of merit ZT, which is a dimensionless quantity, a combination of the three main transport properties of a material—the thermal conductivity κ, the electrical conductivity σ, and the thermopower (Seebeck coefficient) S, as well as of the absolute temperature T:

$$ZT = \frac{\sigma S^2}{\kappa} T. \tag{2}$$

The maximum efficiency is given by

$$\eta_{\max} = \eta_C \frac{\sqrt{ZT+1}-1}{\sqrt{ZT+1}+1}, \tag{3}$$

where η_C is the Carnot efficiency, while the efficiency $\eta(W_{\max})$ at maximum output power $W_{\max}$ reads [15]

$$\eta(W_{\max}) = \frac{\eta_C}{2} \frac{ZT}{ZT+2}. \tag{4}$$

The only restriction imposed by thermodynamics is $ZT \geq 0$, so that both efficiencies are monotonically growing functions of the figure of merit and $\eta_{\max} \to \eta_C$, $\eta(W_{\max}) \to \frac{\eta_C}{2}$, when $ZT \to \infty$. It has been suggested that the value $ZT = 3$ is the target to be achieved in order to make thermoelectric engines economically competitive. In spite of recent progress in material science, present technology is limited to low ZT materials and no clear path has been identified in order to increase efficiency.

A promising new approach, based on the theory of dynamical systems, has been recently introduced [22,23]. The hope is that the analysis of idealized models may lead to some insight regarding the microscopic mechanisms which lead to the high figure of merit in more realistic materials. While for noninteracting systems, even in the classical framework, the energy filtering mechanism [24–26] has been shown to allow us to reach Carnot efficiency, very little is known for interacting particles. Recent numerical and empirical evidence has shown that for a one-dimensional diatomic disordered chain of hard-point elastically colliding particles, the figure of merit ZT diverges as the number of particles increases [27,28]. Since it has been verified that the energy filtering mechanism does not work here [29], it follows that the divergence of ZT in the thermodynamic limit rests on a different, unknown property.

In the present Letter we analyze and solve this problem. Indeed, we show that for systems having a single relevant constant of motion, the electric conductivity is ballistic, i.e., $\sigma \propto \Lambda$, the heat conductivity is subballistic, $\kappa \propto \Lambda^{\alpha}$ with $\alpha < 1$, and the thermopower is size independent, $S \propto \Lambda^0$, so that the figure of merit $ZT \propto \Lambda^{1-\alpha} \to \infty$ in the thermodynamic limit $\Lambda \to \infty$. Our findings are illustrated by the above-mentioned prototype model of an interacting one-dimensional system: a diatomic chain of hard-point elastically colliding particles, where the total momentum is the only relevant constant of motion.

We start from the equations connecting fluxes and thermodynamic forces within linear irreversible thermodynamics [30,31]:

$$J_\rho = L_{\rho\rho}X_1 + L_{\rho u}X_2, \qquad J_u = L_{u\rho}X_1 + L_{uu}X_2, \quad (5)$$

where J_ρ and J_u are the particle and energy currents, and the thermodynamic forces $X_1 = -\nabla(\beta\mu)$, $X_2 = \nabla\beta$, with μ the chemical potential and $\beta = 1/T$ inverse temperature. (We set the Boltzmann constant $k_B = 1$.) The Onsager coefficients L_{ij} (i, $j = \rho$, u) are related to the familiar transport coefficients as follows:

$$\sigma = \frac{L_{\rho\rho}}{T}, \qquad \kappa = \frac{1}{T^2}\frac{\det \boldsymbol{L}}{L_{\rho\rho}}, \qquad S = \frac{1}{T}\left(\frac{L_{\rho u}}{L_{\rho\rho}} - \mu\right), \quad (6)$$

where $\boldsymbol{L}$ denotes the Onsager matrix with matrix elements L_{ij}, and we have set the electric charge of each particle $e = 1$. Thermodynamics imposes $\det \boldsymbol{L} \geq 0$, $L_{\rho\rho} \geq 0$, $L_{uu} \geq 0$, and $L_{u\rho} = L_{\rho u}$. The figure of merit reads

$$ZT = \frac{(L_{u\rho} - \mu L_{\rho\rho})^2}{\det \boldsymbol{L}}. \quad (7)$$

It diverges (thus leading to maximum efficiency) if and only if the Onsager matrix $\boldsymbol{L}$ is ill conditioned, that is, in the so-called strong-coupling condition, for which the energy and particle currents are proportional, $J_u \propto J_\rho$, the proportionality factor is independent of the values of the applied thermodynamic forces.

The Green-Kubo formula expresses the Onsager coefficients in terms of correlation functions of the corresponding current operators, calculated at thermodynamic equilibrium [32,33]:

$$L_{ij} = \lim_{\omega\to 0} \mathrm{Re} L_{ij}(\omega), \quad (8)$$

where

$$L_{ij}(\omega) \equiv \lim_{\epsilon\to 0} \int_0^\infty dt e^{-i(\omega - i\epsilon)t} \times \lim_{\Lambda\to\infty} \frac{1}{\Lambda}\int_0^\beta d\tau \langle J_i J_j(t + i\tau)\rangle_T. \quad (9)$$

The real part of $L_{ij}(\omega)$ can be decomposed into a δ function at zero frequency defining the *generalized Drude weight* $\mathcal{D}_{ij}$ (for $i = j = \rho$ this is the conventional Drude weight) and a regular part $L_{ij}^{\mathrm{reg}}(\omega)$:

$$\mathrm{Re}\, L_{ij}(\omega) = 2\pi \mathcal{D}_{ij}\delta(\omega) + L_{ij}^{\mathrm{reg}}(\omega). \quad (10)$$

Nonzero Drude weights, $\mathcal{D}_{ij} \neq 0$ for i, $j = \rho$, u are a signature of ballistic transport, namely, $L_{ij} \propto \Lambda$ at the thermodynamic limit, and therefore the thermopower $S = L_{\rho u}/(TL_{\rho\rho}) - \mu/T \propto \Lambda^0$.

We now discuss the influence of conserved quantities on the figure of merit ZT. We make use of Suzuki's formula [2] for the currents J_ρ and J_u, which generalizes Mazur's inequality (1) by stating that, for a system of finite size Λ,

$$C_{ij}(\Lambda) \equiv \lim_{t\to\infty} \frac{1}{t}\int_0^t dt' \langle J_i(t') J_j(0)\rangle_T = \sum_{n=1}^{M} \frac{\langle J_i Q_n\rangle_T \langle J_j Q_n\rangle_T}{\langle Q_n^2\rangle_T}, \quad (11)$$

where the summation is extended over all the M orthogonal constants of motion which are relevant for the considered flows, that is, nonorthogonal to the currents J_ρ and J_u, i.e., $\langle J_\rho Q_n\rangle_T \neq 0$ and $\langle J_u Q_n\rangle_T \neq 0$. (Irrelevant constants of motion are not included in the summation since they would give zero contribution.) The presence of relevant conservation laws implies that the *finite-size* generalized Drude weights

$$D_{ij}(\Lambda) \equiv \frac{1}{2\Lambda} C_{ij}(\Lambda) \quad (12)$$

are different from zero. If at the thermodynamic limit the generalized Drude weight

$$\mathcal{D}_{ij} = \lim_{t\to\infty}\lim_{\Lambda\to\infty} \frac{1}{2\Lambda t}\int_0^t dt' \langle J_i(t') J_j(0)\rangle_T \quad (13)$$

is nonzero, then we can conclude that transport is ballistic [34]. Note that in Eq. (13) the thermodynamic limit $\Lambda \to \infty$ must be taken before the long-time limit $t \to \infty$. The below developed theory only applies to the cases in which the two limits commute, that is, $\mathcal{D}_{ij} = \lim_{\Lambda\to\infty} D_{ij}(\Lambda)$ [35].

If there is a *single* relevant constant of motion, $M = 1$, due to Suzuki's formula (11) (and assuming that the two limits $\Lambda \to \infty$ and $t \to 0$ commute), the ballistic contribution to $\det \boldsymbol{L}$ vanishes, since it is proportional to $\mathcal{D}_{\rho\rho}\mathcal{D}_{uu} - \mathcal{D}_{\rho u}^2$, which is zero from (11). Hence, $\det \boldsymbol{L}$ grows due to the contributions involving the regular part in Eq. (10), i.e., slower than Λ^2, thus implying that the thermal conductivity $\kappa \propto \det \boldsymbol{L}/L_{\rho\rho}$ grows subballistically. That is, $\kappa \propto \Lambda^{\alpha}$, with $\alpha < 1$. Since $\sigma \propto L_{\rho\rho}$ is ballistic and $S = L_{\rho u}/TL_{\rho\rho} - \mu/T \propto \Lambda^0$, we can conclude that $ZT = \sigma S^2 T/\kappa \propto \Lambda^{1-\alpha} \to \infty$ when $\Lambda \to \infty$.

The situation is drastically different if $M > 1$. In this case, due to the Schwartz inequality,

$$D_{\rho\rho}D_{uu} - D_{\rho u}^2 = ||\boldsymbol{x}_\rho||^2 ||\boldsymbol{x}_u||^2 - \langle \boldsymbol{x}_\rho, \boldsymbol{x}_u\rangle \geq 0, \quad (14)$$

where

$$\boldsymbol{x}_i = (x_{i1}, \ldots, x_{iM}) = \frac{1}{2\Lambda}\left(\frac{\langle J_i Q_1\rangle_T}{\sqrt{\langle Q_1^2\rangle_T}}, \ldots, \frac{\langle J_i Q_M\rangle_T}{\sqrt{\langle Q_M^2\rangle_T}}\right), \quad (15)$$

and $\langle \boldsymbol{x}_\rho, \boldsymbol{x}_u\rangle = \sum_{k=1}^{M} x_{\rho k} x_{uk}$. The equality arises only in the exceptional case when the vectors $\boldsymbol{x}_\rho$ and $\boldsymbol{x}_u$ are parallel. Hence, for $M > 1$ we expect, in general, $\det \boldsymbol{L} \propto \Lambda^2$, so that heat transport is ballistic and $ZT \propto \Lambda^0$.

In order to illustrate the above general ideas, we consider a one-dimensional, diatomic disordered chain of N hard-point elastically colliding particles with randomly distributed coordinates $z_i \in [0, \Lambda]$, velocities v_i, and masses $m_i \in \{\nu_1, \nu_2\}$. The numerically observed divergence of the figure of merit ZT for $\nu_1 \neq \nu_2$ [27–29] can be understood now in terms of the above developed theory. Indeed, in this system there is a single relevant constant of motion $Q_1 = P$, where $P = \sum_{i=1}^{N} m_i v_i$ is the overall momentum [36]. In this case the particle current $J_\rho = \sum_{i=1}^{N} v_i$ and the energy current $J_u = \sum_{i=1}^{N} \frac{1}{2} m_i v_i^3$. Note that the mass current, $J_m \equiv \sum_{i=1}^{N} m_i v_i$, equals the total momentum P and therefore does not decay, while on the other hand $J_m \sim \bar{m} J_\rho$, where $\bar{m}$ is the average mass per particle; hence, we expect that $\langle J_\rho(t) J_\rho(0)\rangle$ does not decay either, so that $\sigma \sim \Lambda$.

It is easy to compute analytically the time-averaged correlation functions $C_{ij}(\Lambda)$ [from the second line of Eq. (11)] and then the finite-size generalized Drude weights

$$\begin{aligned} D_{\rho\rho}(\Lambda) &= \frac{C_{\rho\rho}(\Lambda)}{2\Lambda} = \frac{TN^2}{2\Lambda(\nu_1 N_1 + \nu_2 N_2)}, \\ D_{uu}(\Lambda) &= \frac{C_{uu}(\Lambda)}{2\Lambda} = \frac{9T^3N^2}{8\Lambda(\nu_1 N_1 + \nu_2 N_2)}, \\ D_{\rho u}(\Lambda) &= \frac{C_{\rho u}(\Lambda)}{2\Lambda} = \frac{3T^2N^2}{4\Lambda(\nu_1 N_1 + \nu_2 N_2)}. \end{aligned} \quad (16)$$

Here, $N = N_1 + N_2$ with N_1 and N_2 the number of particles with mass ν_1 and ν_2, respectively. Note that, as expected from the above theory, $D_{\rho\rho}(\Lambda) D_{uu}(\Lambda) - D_{\rho u}^2(\Lambda) = 0$ for any system size Λ.

To numerically confirm the above results, we compute the autocorrelation functions of J_ρ and J_u, and the cross correlation function between them. In doing so we apply periodic boundary conditions and assign to N_1 (N_2) particles of mass ν_1 (ν_2) random initial positions and random initial velocities derived from the Maxwell-Boltzmann distribution corresponding to temperature T. We then evolve the system and compute $c_{ij}(\Lambda, t) = \frac{1}{t}\int_0^t dt' \langle J_i(t') J_j(0)\rangle_T$ (for $i, j = \rho, u$) up to a time t sufficiently long to obtain stable averages, thus estimating $C_{ij}(\Lambda) = \lim_{t\to\infty} c_{ij}(\Lambda, t)$. We show in Fig. 1 that the current-current correlation functions $\langle J_i(t) J_j(0)\rangle_T$ do not decay to zero as the correlation time is increased, implying that $c_{ij}(\Lambda, t)$ do not decay either and thus indicating ballistic transport. We finally estimate the finite-size generalized Drude weights from the time-averaged correlation functions as $c_{ij}(\Lambda, t)/(2\Lambda)$, with a sufficiently long time t to approximate the asymptotic value $D_{ij}(\Lambda) = C_{ij}(\Lambda)/(2\Lambda)$. As shown in Fig. 2, the numerically determined values are in very good agreement with the theoretical values D_{ij} given by Eq. (16).

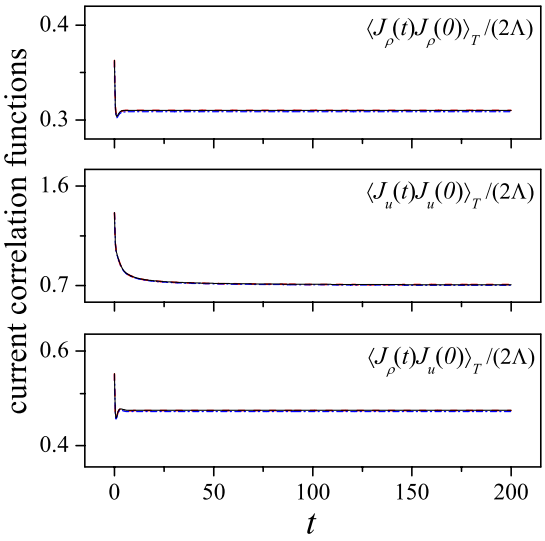

FIG. 1 (color online). The current-current correlation functions for various system sizes with $\Lambda = 256$ (red dashed curve), 512 (blue dash-dotted curve), and 1024 (black solid curve). The temperature $T = 1$, particle masses are $\nu_1 = 1$ and $\nu_2 = \sqrt{5}$. It is seen that all the current-current correlation functions approach a finite nonzero value as the correlation time increases and that the characteristic time scale to approach such value is independent of the system size. Note that in all the figures the particle density is fixed to be $N/\Lambda = 1$ and $N_1 = N_2 = N/2$.

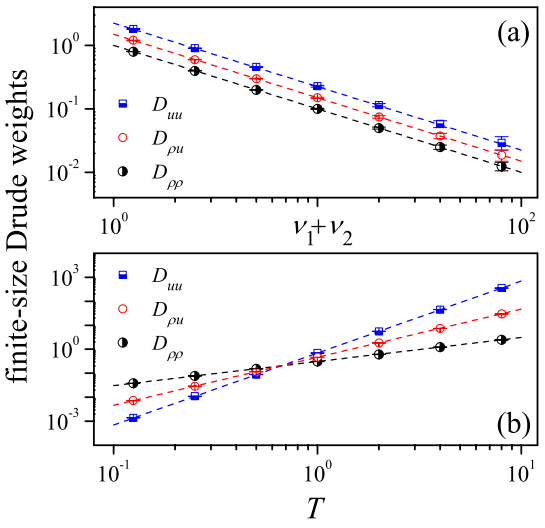

FIG. 2 (color online). Comparison between the numerically determined finite-size generalized Drude weights (symbols) and their analytical values (lines) given by Eq. (16) for $\Lambda = 256$. We set $T = 1$ and $\nu_1 = 1$ in (a) and $\nu_1 = 1$, $\nu_2 = \sqrt{5}$ in (b).

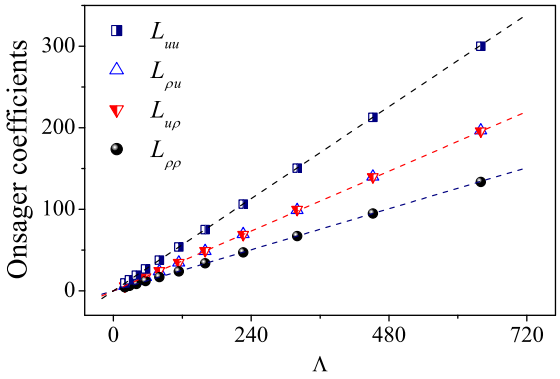

FIG. 3 (color online). Dependence of the Onsager coefficients L_{ij} on the system size Λ. The straight lines are the best linear fitting, $L_{ij} \propto \Lambda$. In these nonequilibrium simulations we set the temperature $T = 1$, the chemical potential $\mu = 0$, and the masses $\nu_1 = 1$ and $\nu_2 = \sqrt{19}$.

Figure 1 provides clear evidence that the convergence of the correlation functions $c_{ij}(\Lambda, t)$ to their asymptotic values $C_{ij}(\Lambda)$ takes place on a time scale independent of the system size Λ. Therefore, Fig. 1 provides a strong indication that for the model under investigation the two limits $\Lambda \to \infty$ and $t \to \infty$ do commute, so that we can compute the generalized Drude weights at the thermodynamic limit as $\mathcal{D}_{ij} = \lim_{\Lambda\to\infty} D_{ij}(\Lambda)$. Note that in taking the thermodynamic limit, we keep constant the particle density N/Λ and the ratio N_1/N_2.

Finally, we perform a nonequilibrium calculation of the various transport coefficients. (For technical details of numerical simulations see Ref. [28].) According to our theory, we expect that all the Onsager coefficients grow linearly with the system size in the thermodynamic limit, since the generalized Drude weights $\mathcal{D}_{ij}$ are all different from zero. This expectation is confirmed by the data shown in Fig. 3. Finally, in Fig. 4 we show the transport coefficients σ, S, κ, and the thermoelectric figure of merit ZT as a function of the system size. In agreement with our theory, we observe that $\sigma \propto \Lambda$, while S saturates to the value predicted from theory for ballistic transport, $S = \frac{1}{T}(\frac{\mathcal{D}_{\rho u}}{\mathcal{D}_{\rho\rho}} - \mu) = \frac{3}{2}$, $\kappa \propto \Lambda^{\alpha}$ with $\alpha \approx 1/3$ [37,38], and the growth of the figure of merit in good agreement with the dependence $ZT \propto \Lambda^{1-\alpha}$.

Note that the above conclusions for the thermal conductivity κ and the figure of merit ZT do not hold in the integrable case $\nu_1 = \nu_2$, where $M > 1$ since all moments of the momentum distribution are conserved quantities. In this case $\kappa \propto \Lambda$ and it is easy to analytically compute $ZT = 1$ [27].

To summarize, we have shown that for systems with a *single relevant* constant of motion, the thermoelectric figure of merit diverges as the system size increases, so that the Carnot efficiency is achieved at the thermodynamic limit. Such a result has been illustrated in the case of a chain of hard-point elastically colliding particles, with a remarkable agreement between analytical results, equilibrium, and out-of-equilibrium numerical simulations. We would like to point out that, while our illustrative model is one dimensional, there are no dimensionality restrictions in our theory, so that it should apply also to two- and three-dimensional systems in which total momentum is the only relevant constant of motion. Therefore, our paper unveils a rather generic mechanism for increasing thermoelectric efficiency in interacting systems.

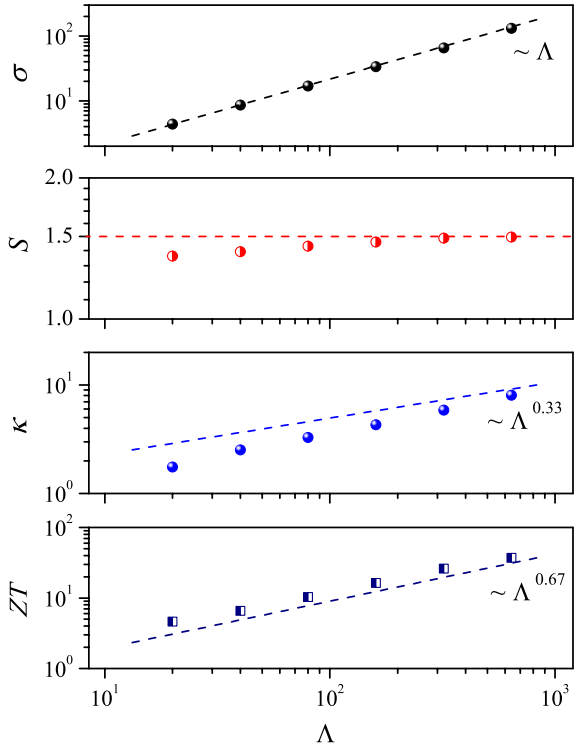

FIG. 4 (color online). Dependence of the transport coefficients σ, S, κ, and ZT on the system size Λ, for the same parameter values as in Fig. 3. Dashed lines are drawn for reference.

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[34] See Ref. [3] for a detailed discussion and derivation of Eq. (13).
[35] See Ref. [3] for a proof of the commutation of the two limits for a class of quantum spin chains.
[36] Total energy E and number of particles N_1 and N_2 are also constants of motion. However, they are not relevant since they are even functions of the velocities v_i and therefore the thermodynamic averages $\langle EJ_i\rangle_T$, $\langle N_1 J_i\rangle_T$, and $\langle N_2 J_i\rangle_T$ ($i = \rho,\ u$) vanish, being J_ρ and J_u odd functions of velocities.
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