## Most Efficient Quantum Thermoelectric at Finite Power Output

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Machines are only Carnot efficient if they are reversible, but then their power output is *vanishingly small*. Here we ask, what is the maximum efficiency of an irreversible device with *finite* power output? We use a nonlinear scattering theory to answer this question for thermoelectric quantum systems, heat engines or refrigerators consisting of nanostructures or molecules that exhibit a Peltier effect. We find that quantum mechanics places an upper bound on both power output and on the efficiency at any finite power. The upper bound on efficiency equals Carnot efficiency at zero power output but decays with increasing power output. It is intrinsically quantum (wavelength dependent), unlike Carnot efficiency. This maximum efficiency occurs when the system lets through all particles in a certain energy window, but none at other energies. A physical implementation of this is discussed, as is the suppression of efficiency by a phonon heat flow.

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Introduction.—Quantum thermodynamics [1] is the physics of thermodynamic processes in quantum systems, such as the conversion of heat to work. This is of particular interest for the thermoelectric response [2–4] of nanostructures [5] or molecules [6,7]. It places fundamental bounds on the efficiency and power output of heat engines and refrigerators made from such systems, such as Carnot's thermodynamic bound on efficiency or Pendry's quantum bound on entropy flow [8].

The efficiencies of heat engines  $\eta_{\rm eng}$  and refrigerators  $\eta_{\rm fri}$  are particularly important ( $\eta_{\rm fri}$  is called the coefficient of performance, COP). These efficiencies are the ratio of power output to power input. For a heat engine, the output is the electrical power  $P_{\rm gen}$  and the input is the heat flow out of a reservoir [the left (L) reservoir in Fig. 1(c)],  $J_L$ . For a refrigerator, it is the inverse. For left (L) and right (R) reservoirs at temperatures  $T_L$  and  $T_R$ , Carnot's bounds on these efficiencies are

$$\eta_{\rm eng}^{\rm Carnot} = 1 - T_R/T_L, \qquad \eta_{\rm fri}^{\rm Carnot} = (T_R/T_L - 1)^{-1}, \qquad (1)$$

where heat flows as in Fig. 1, so  $T_L > T_R$  for heat engines and  $T_R > T_L$  for refrigerators. Proposals exist to achieve these efficiency in bulk [9] or quantum [10–12] systems.

However Carnot efficiency is only achieved in "reversible systems," which have vanishing power output. Any useful device must give a finite power output and so be "irreversible." So what are the equivalents of Carnot efficiencies for such irreversible (entropy-producing) systems? To be more precise, we note that engineers typically need a device to provide a certain power, at the highest possible efficiency. Thus, we ask, what is the maximum allowed efficiency at any *given* power output? As physicists, we can also ask what is the least irreversible system (i.e., that which produces the least entropy) that delivers a given power output? With a little algebra, the

first and second laws of thermodynamics [13] tell us that a heat engine producing power P must also produce entropy at a rate

$$\dot{S}(P) = (P/T_R)(\eta_{\text{eng}}^{\text{Carnot}}/\eta_{\text{eng}} - 1). \tag{2}$$

Similarly, a refrigerator with cooling power J has

$$\dot{S}(J) = (J/T_R)(1/\eta_{\text{fri}} - 1/\eta_{\text{fri}}^{\text{Carnot}}). \tag{3}$$

Thus, the two above questions are the same, since the most efficient system is the least irreversible.

Central results.—We answer these questions for any thermoelectric quantum system that can be modeled with nonlinear Landauer-Büttiker scattering theory.

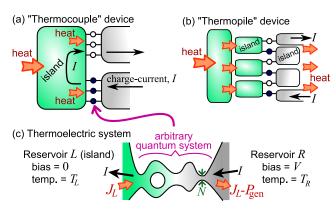


FIG. 1 (color online). Typical thermoelectric devices are shown in (a) and (b), with (c) showing the quantum system with the thermoelectric response. To unify the analysis, heat is always taken to flow as shown; hence, a heat engine has  $T_L > T_R$ , while a refrigerator has  $T_L < T_R$ . In (a) and (b), the filled (open) circles are quantum systems where transport occurs via electron states above the Fermi surface (hole states below the Fermi surface). In (c), N is the number of transverse modes in the narrowest part of the quantum system.

First, we find that quantum mechanics places an upper bound on the power output of such systems,

heat engine: 
$$P_{\text{gen}} \le P_{\text{gen}}^{\text{QB2}} \equiv A_0 \frac{\pi^2}{h} N k_B^2 (T_L - T_R)^2$$
, (4)

refrigerator: 
$$J_L \le \frac{1}{2} J_L^{\text{QB}} \equiv \frac{1}{12} \frac{\pi^2}{h} N k_B^2 T_L^2$$
, (5)

where  $A_0 \simeq 0.0321$ . We refer to  $P_{\rm gen}^{\rm QB2}$  and  $J_L^{\rm QB}$  as quantum bounds (QBs), as they depend on the number of transverse modes in the quantum system N, which scales like the inverse Fermi wavelength.  $J_L^{\rm QB}$  is Pendry's quantum bound on the heat current out of reservoir L [8]. The "2" on  $P_{\rm gen}^{\rm QB2}$  indicates that it is for two-lead systems [14].

Second, we find a fundamental upper bound on the efficiencies at finite power output, which is *lower* than Carnot efficiency. The upper bound for a heat engine is a decaying function of  $P_{\rm gen}/P_{\rm gen}^{\rm QB2}$ , whereas the upper bound for a refrigerator is a decaying function of  $J_L/J_L^{\rm QB}$ . At small output power, these bounds on efficiencies are

$$\eta_{\rm eng}(P_{\rm gen}) = \eta_{\rm eng}^{\rm Carnot} \left( 1 - 0.478 \sqrt{\frac{T_R}{T_L}} \frac{P_{\rm gen}}{P_{\rm gen}^{\rm QB2}} \right), \quad (6)$$

$$\eta_{\text{fri}}(J_L) = \eta_{\text{fri}}^{\text{Carnot}} \left( 1 - 1.09 \sqrt{\frac{T_R}{T_R - T_L}} \frac{J_L}{J_L^{\text{QB}}} \right), \quad (7)$$

to lowest order in  $P_{\rm gen}/P_{\rm gen}^{\rm QB2}$  and  $J_L/J_L^{\rm QB}$ , respectively. In these limits, the least irreversible heat engine produces entropy at a rate  $\dot{S} \propto P_{\rm gen}^{3/2}$ , whereas the least irreversible refrigerator does so at a rate  $\dot{S} \propto J_L^{3/2}$ .

These fundamental upper bounds on efficiencies at finite power are of quantum origin (they are wavelength dependent), unlike Carnot's bounds (which were derived using classical physics). They play the role for irreversible thermoelectric systems that Carnot's bounds do for reversible systems and are more stringent than Carnot's bounds. This upper bound on efficiency is achieved when only particles in a given energy window (determined by the desired power output) traverse the quantum system; see Fig. 2(a). Real systems will have lower efficiencies; improving them would only approach these bounds.

Nonlinear theory.—Linear-response theory works in bulk systems for most  $T_R/T_L$  values [15], but a nonlinear theory is needed for quantum systems whenever  $1-T_R/T_L$  is not small. An example would be getting electricity from a thermoelectric between a diesel motor's exhaust  $\approx$ 700 K and its surroundings  $\approx$ 280 K [in which case the bound in Eq. (4) is  $\sim$ 10 nW per transverse mode].

Interactions are crucial in the nonlinear regime and must be treated in a manner appropriate to the system in question. Here, we use a nonlinear Landauer-Büttiker scattering formula, which was first derived by treating

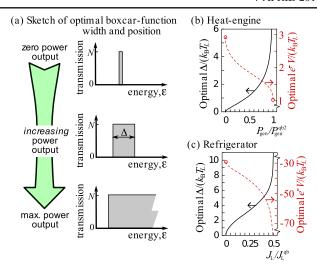


FIG. 2 (color online). (a) Sketch of how the optimal transmission changes as the required power output is increased. Maximum power is when the right-hand edge of the boxcar function goes to  $+\infty$ . The qualitative features always follow this sketch, while the quantitative details depend on  $T_R/T_L$ . (b) Plots of optimal  $\Delta$  (solid curve) and  $e^-V$  (dashed curve) against heatengine power output  $P_{\rm gen}$  at  $T_R/T_L=0.1$ . The first equation in Eq. (12) then gives  $\epsilon_0$ . (c) Plots of optimal  $\Delta$  (solid curve) and  $e^-V$  (dashed curve) against cooling power  $J_L$  at  $T_R/T_L=10$ . The second equation in Eq. (14) then gives  $\epsilon_1$ .

electron-electron interactions as mean-field charging effects [16,17] and recently applied to thermoelectric effects [18–20,26]. Identical equations apply for resonant level models [10,21–24] and have been derived from functional renormalization group (RG) [25] for such models with single-electron charging effects. References [26,27] show that such theories respect thermodynamics. The heat current out of the L reservoir into the quantum system  $J_L$  and the electrical power generated by the system  $P_{\rm gen} = VI_L$  are

$$J_L = \frac{1}{h} \sum_{\mu} \int_0^{\infty} d\epsilon \epsilon \mathcal{T}_{RL}^{\mu\mu}(\epsilon) [f_L^{\mu}(\epsilon) - f_R^{\mu}(\epsilon)], \qquad (8)$$

$$P_{\rm gen} = \frac{1}{h} \sum_{\mu} \int_{0}^{\infty} d\epsilon \mu e^{-V} \mathcal{T}_{RL}^{\mu\mu}(\epsilon) [f_{L}^{\mu}(\epsilon) - f_{R}^{\mu}(\epsilon)], \quad (9)$$

where  $e^-$  is the electron charge  $(e^- < 0)$ . The sum is over  $\mu = 1$  for "electron" states above the L reservoir's chemical potential and  $\mu = -1$  for "hole" states below that chemical potential. Interaction effects mean that the transmission function  $T_{RL}^{\mu\mu}(\epsilon)$  is a self-consistently determined function of  $T_{L,R}$  and V. The Fermi function for electrons entering from reservoir j is

$$f_j^{\mu}(\epsilon) = \{1 + \exp[(\epsilon - \mu e^{-V_j})/(k_B T_j)]\}^{-1}.$$

Scattering theory has been used to find the properties of many thermoelectric systems from their  $T_{RL}^{\mu\mu}(\epsilon)$  value, e.g., Refs. [6,12,18–25,28–37]. Here instead, we find the

 $T^{\mu\mu}_{RL}(\epsilon)$  that maximizes efficiency at given power output. We initially assume only elastic scattering in the quantum system, although decoherence without relaxation is allowed as it does not change the structure of Eqs. (8) and (9). Inelastic effects are briefly discussed at the end of this Letter. We take each island (see Fig. 1) as large enough to be a reservoir in local equilibrium. This differs from the "three-terminal" systems [38–42], in which particles remain coherent in the island. Here, we only discuss electrons dominating transmission (filled circles in Fig. 1). When holes dominate (open circles in Fig. 1), one takes  $T^{\mu,\mu}_{RL}(\epsilon) \to T^{-\mu,-\mu}_{RL}(\epsilon)$  with  $V \to -V$ , then  $I_L \to -I_L$  while  $J_L$  and  $P_{\rm gen}$  are unchanged.

Literature on reversibility and irreversibility.—To be Carnot efficient, systems must be reversible (create no entropy); for a thermoelectric, there are two requirements for this [10]. First, it must have a  $\delta$ -function-like transmission [9,11,12] [ $\Delta/(k_BT_{L,R}) \to 0$  in Fig. 3(b)] for which the figure of merit  $ZT \to \infty$ . Second, the load resistance must be such that  $e^-V = \epsilon_0(1 - T_R/T_L)$  [10] so the reservoirs' occupations are equal at  $\epsilon_0$ . However, then the power output vanishes,  $P_{\rm gen} \propto \Delta^2 \to 0$ .

Larger  $P_{\rm gen}$  requires heat engines that are irreversible (create a finite amount of entropy per unit of work provided). The authors of Ref. [24], motivated by works on classical pumps [43–45], proposed increasing  $P_{\rm gen}$  by keeping  $\Delta \to 0$  ( $ZT \to \infty$ ) but choosing the load to maximize  $P_{\rm gen}$ , rather than achieve reversibility. The resulting Curzon-Alhborn efficiency is significantly below  $\eta_{\rm eng}^{\rm Carnot}$ , yet  $P_{\rm gen} \propto \Delta$  remains very small. Other works on finite power include Refs. [23,25,37,46].

Here, we get an efficiency higher than the Curzon-Alhborn efficiency found in Ref. [24] for the same (or much larger)  $P_{\rm gen}$  by making  $\Delta$  finite (thereby decreasing ZT). Thus,  $ZT \to \infty$  does not give maximal efficiency at given (finite) power output. That said, our work does not consider ZT further, as it has little meaning outside the linear-response regime [19,20,31,47].

Heat engine.—Here, we find the transmission function  $\mathcal{T}_{RL}^{\mu\mu}(\epsilon)$  that maximizes the heat-engine efficiency  $\eta_{\rm eng}(P_{\rm gen}) = P_{\rm gen}/J_L$  for a given power generated  $P_{\rm gen}$ . We treat  $\mathcal{T}_{RL}^{\mu\mu}(\epsilon)$  as a set of slices as in Fig. 3(a) and find

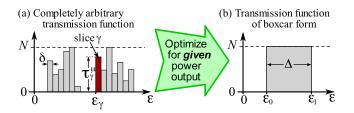


FIG. 3 (color online). Finding the  $T^{\mu\mu}_{RL}(\epsilon)$  function that maximizes the efficiency. In (a)  $T^{\mu\mu}_{RL}(\epsilon)$  is considered as infinitely many slices of width  $\delta \to 0$ , so slice  $\gamma$  has energy  $\epsilon_{\gamma} \equiv \gamma \delta$  and height  $\tau^{\mu}_{\gamma}$ . This gives (b) with a transcendental equation for  $\epsilon_0$  and  $\epsilon_1$ .

optimal values of each slice and of the bias V under the constraint of fixed  $P_{\rm gen}$ . A little algebra shows that  $\eta_{\rm eng}(P_{\rm gen})$  will only grow with increasing  $\tau_{\gamma}^{\mu}$ , if  $\epsilon_{\gamma}$  satisfies

$$\left[\epsilon_{\gamma} - \mu e^{-V} J_{L}^{\prime} / P_{\text{gen}}^{\prime}\right] \partial P_{\text{gen}} / \partial \tau_{\gamma}^{\mu} |_{V} < 0, \tag{10}$$

where the prime symbol indicates  $\partial/\partial V$  for fixed  $\mathcal{T}^{\mu\mu}_{RL}(\epsilon)$ . From this, the optimal  $\mathcal{T}^{\mu\mu}_{RL}(\epsilon)$  is a boxcar function [Fig. 3(b)],

$$\mathcal{T}^{\mu\mu}_{RL}(\epsilon) = \begin{cases} N & \text{for } \mu = 1 & \text{and} & \epsilon_0 < \epsilon < \epsilon_1 \\ 0 & \text{otherwise} \end{cases}$$
 (11)

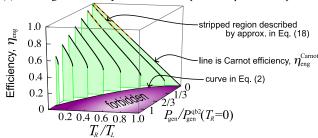
where N is given in Fig. 1(c). Then the integrals in Eqs. (8) and (9) are sums of terms containing logarithmic and dilogarithm functions of  $\epsilon_0$  and  $\epsilon_1$ . Equation (10) gives

$$\epsilon_0 = e^- V / (1 - T_R / T_L), \qquad \epsilon_1 = e^- V J_L / P_{\text{gen}}'.$$
 (12)

Since  $J_L$  and  $P_{\rm gen}$  depend on  $\epsilon_1$ , the second equality is a transcendental equation for  $\epsilon_1$ . Solving this, we get  $J_L(V)$  and  $P_{\rm gen}(V)$  and so  $\eta(V)$ . To get  $\eta(P_{\rm gen})$  from  $\eta(V)$ , we invert  $P_{\rm gen}(V)$  and substitute for V. Below, we do these steps analytically for high power ( $P_{\rm gen} = P_{\rm gen}^{\rm QB2}$ ) and low power ( $P_{\rm gen} \ll P_{\rm gen}^{\rm QB2}$ ). For other cases, a numerical solution is plotted in Figs. 2(b) and 4(a).

The quantum bound on power output, given in Eq. (4), is found by noting that the maximum occurs when  $P'_{\rm gen}=dP_{\rm gen}/d\epsilon_1=0$ . For this  $\epsilon_1\to\infty$ , so  $T^{\mu\mu}_{RL}(\epsilon)$  is a Heaviside  $\theta$ -step function, while  $e^{-\epsilon_0/(k_BT_L)}\simeq 0.318$ . The efficiency at this maximum power is

(a) Heat-engine efficiency as a function of power output and temperature



(b) Refrigerator efficiency as a function of power output and temperature

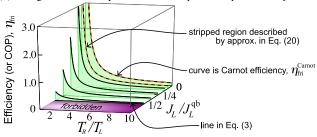


FIG. 4 (color online). Efficiencies of (a) heat engines and (b) refrigerators. The black lines are the maximum allowed efficiency for various power outputs as functions of  $T_R/T_L$ . The light green regions indicate allowed efficiencies for those power outputs.

$$\eta_{\rm eng}(P_{\rm gen}^{\rm QB2}) = \eta_{\rm eng}^{\rm Carnot}/[1 + C_0(1 + T_R/T_L)], \quad (13)$$

with  $C_0 \simeq 0.936$ , so it is always more than  $0.3\eta_{\rm eng}^{\rm Carnot}$ .

For low power output, one can take Eqs. (8) and (9) with Eq. (11) and easily perform a small  $\Delta$  expansion up to order  $(\Delta/k_BT_R)^3$ . In this limit, Eq. (12) is satisfied by  $\epsilon_0=3.2436k_BT_L$ . Similarly, taking  $P_{\rm gen}$  to lowest order in  $\Delta$ , we rewrite  $\eta$  in terms of  $P_{\rm gen}$  to get Eq. (6).

Refrigerator.—A refrigerator's efficiency, or COP, is  $\eta_{\rm fri}(J_L)=J_L/P_{\rm abs},$  where  $P_{\rm abs}=-P_{\rm gen}$  is the electrical power absorbed by the refrigerator. We maximize  $\eta_{\rm fri}(J_L),$  for given cooling power  $J_L$ . This gives the boxcar function in Eq. (11) with

$$\epsilon_0 = -e^- V J_L' / P_{\text{abs}}', \qquad \epsilon_1 = -e^- V / (T_R / T_L - 1), \quad (14)$$

so  $\epsilon_0$  is given by a transcendental equation. This is solved below analytically at high and low  $J_L$  values; otherwise, the numerical results are given in Figs. 2(c) and 4(b).

The quantum bound on cooling power in Eq. (5) occurs when the transmission function is a  $\theta$ -step function ( $\epsilon_0 = 0$ ,  $\epsilon_1 \to \infty$ , and  $-e^-V \to \infty$ ). Then,  $\eta_{\rm fri}(J_L)$  is zero, since V is infinite. However, one gets exponentially close to this limit for  $-e^-V \gg k_B T_R$ , for which  $\eta_{\rm fri}(J_L)$  is finite [see Fig. 4(b)]. In the opposite limit,  $J_L \ll J_L^{\rm QB}$ , an expansion up to third order in  $\Delta/(k_B T_L)$  gives Eq. (7).

Phonons and photons.—These unavoidably carry heat from hot to cold in parallel with the electronic flow. Their heat current  $J_{\rm ph}$  depends nonlinearly on  $T_{L,R}$  (given by a Stefan-Boltzmann law or similar [8,48,49]). Then, maximal efficiencies for heat engines and refrigerators are suppressed and given by

$$\begin{split} \eta_{\rm eng}^{e+{\rm ph}}(P_{\rm gen}) &= [\eta_{\rm eng}^{-1}(P_{\rm gen}) + J_{\rm ph}/P_{\rm gen}]^{-1}, \\ \eta_{\rm fri}^{e+{\rm ph}}(J-J_{\rm ph}) &= (1-J_{\rm ph}/J)\eta_{\rm fri}(J) \quad {\rm for} \ J > J_{\rm ph}, \end{split}$$

where  $\eta_{\text{eng}}(P_{\text{gen}})$ ,  $\eta_{\text{fri}}(J)$  are efficiencies at  $J_{\text{ph}}=0$ .

In many devices, there is a large phonon or photon heat flow  $J_{\rm ph}$ . For a heat engine in a situation where  $J_{\rm ph}\gg \eta_{\rm eng}P_{\rm gen}$ , one has  $\eta_{\rm eng}^{e+{\rm ph}}(P_{\rm gen})=P_{\rm gen}/J_{\rm ph}$ . Thus, the efficiency is maximal when the power is maximal, as given by Eq. (4). For a refrigerator to cool, it needs  $J-J_{\rm ph}>0$ , so one may need the maximum cooling power in Eq. (5) when  $J_{\rm ph}$  is large. In both cases, this corresponds to a  $\theta$ -step transmission function.

Inelastic effects.—Inelastic electron-phonon and electron-electron interactions in the quantum system are not accounted for in the above theory. However, they will be negligible if the quantum system is small enough. At 700 K, electrons typically travel tens of nanometers before an inelastic scattering, so if the quantum system is a few ångstroms across, inelastic effects may be insignificant. Below 1 K, electrons can traverse microsized structures without inelastic scattering. We will address inelastic effects in detail elsewhere [50] using a voltage-probe model [51].



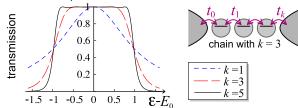


FIG. 5 (color online). Chain of systems with levels at energy  $E_0$  and hoppings  $\{t_i\}$  that will hybridize to form a band centered at  $E_0$ , with a width given by the hopping. If the hoppings are smallest at the chain's middle and larger at its ends [50], the transmission becomes increasingly like a boxcar function as one increases k (the bandwidths in the plots have been set to 1).

We will show that they *cannot* increase the maximum power beyond that in Eqs. (4) and (5). For low powers, we will show that they cannot increase the maximum efficiencies beyond those in Eqs. (6) and (7). For intermediate powers, it remains open whether they could raise the maximum efficiency; however, there is no reason to think so.

Many quantum systems in parallel.—Increasing the number of modes N increases the efficiency at given power output. This is because the quantum bounds in Eqs. (4) and (5) go like N, and the efficiency goes toward Carnot efficiency as these bounds grow.

However, most thermoelectric quantum systems have (exceptions being superconductor-normalmetal–superconductor structures [5]). Then, large N values would require many N = 1 systems in parallel. For a surface covered with a certain density of such systems [12], Eqs. (4) and (5) become bounds on the power per unit area. Carnot efficiency is only approachable when the power per unit area is much less than these bounds. The number of modes per unit area cannot exceed  $\lambda_F^{-2}$ , for Fermi wavelength  $\lambda_F$ . Thus, Eq. (4) tells us that to get 100 W of power output from a semiconductor thermoelectric (with  $\lambda_F \sim 10^{-8}$  m) between reservoirs at 700 and 300 K, one needs a cross section of at least 4 mm<sup>2</sup>. To get this power at 90% of Carnot efficiency, one needs a cross section of at least 0.4 cm<sup>2</sup>. Remarkably, it is quantum mechanics which gives these bounds, even though the cross sections in question are macroscopic.

Concluding remark on implementation.—Whenever  $(1-T_R/T_L)$  is not small, the transmission function for a system must be found self-consistently to capture charging effects. This work has shown that maximum efficiency for given power output occurs when this transmission is a boxcar function (with correct position and width). Figure 5 shows one potential implementation; the energy levels should be chosen so that they align at energy  $E_0$  when the optimal bias is applied. To get maximum power output is simpler, since a good point contact has the required  $\theta$ -step function transmission.

I thank M. Büttiker for proposing a band as a candidate for the boxcar transmission.

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- [15] Linear response fails when the ratio of temperature drop  $\delta T$  to temperature T is significant on the scale of the electron relaxation length  $l_{\rm rel}$ . In bulk systems ( $L\gtrsim 1$  mm,  $l_{\rm rel}\lesssim 0.1~\mu$ m), linear response [9] works well, since the ratio is  $(\Delta T/T)l_{\rm rel}/L$ , which is small even for  $\Delta T/T\sim 1$ . For quantum systems ( $L\ll l_{\rm rel}$ ), it fails whenever  $\Delta T/T$  is not small.
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