PHYSICAL REVIEW B **84**, 201306(R) (2011)

Thermopower with broken time-reversal symmetry

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(Received 10 July 2011; revised manuscript received 8 October 2011; published 14 November 2011)

We show that when inelastic scattering effects are taken into account, the thermopower is, in general, asymmetric under magnetic field reversal, even for noninteracting systems. Our findings are illustrated in the example of a three-dot ring structure pierced by an Aharonov-Bohm flux.

DOI: 10.1103/PhysRevB.84.201306 PACS number(s): 72.20.Pa, 05.70.Ln

In power generation and refrigeration by means of thermal engines, efficiency plays a basic theoretical and practical role. The Carnot bound on efficiency lies at the foundations of thermodynamics: For a heat engine functioning between hot and cold reservoirs at temperatures T_h and T_c, the efficiency η, defined as the ratio of the output power over the heat extracted per unit time from the high-temperature reservoir, is upper bounded by the Carnot efficiency η_C: $\eta \leqslant \eta_C = 1 - T_c/T_h$.

For systems with time-reversal symmetry, thermoelectric power generation and refrigeration is governed, within linear response, by a single parameter, the dimensionless figure of merit $ZT = (\sigma S^2/\kappa)T$, where σ is the electric conductivity, S is the thermopower (Seebeck coefficient), κ is the thermal conductivity, and $T \approx T_h \approx T_c$ is the temperature. The maximum efficiency is given by

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

Thermodynamics only imposes $ZT \geqslant 0$ and the Carnot limit is reached when $ZT \to \infty$.

On the other hand, we have recently shown[1] that for systems with broken time-reversal symmetry the efficiency depends on two parameters: a "figure of merit" and an asymmetry parameter. In contrast to the time-symmetric case, the figure of merit is bounded from above, yet the Carnot efficiency can be reached at lower and lower values of the figure of merit as the asymmetry parameter increases. According to the expression for the efficiency, large asymmetry of the thermopower can be responsible for highly nontrivial effects,[1] and potentially can be a useful tuning parameter to control thermoelectric efficiency of the material. Hence, finding general conditions for asymmetry of the thermopower is of general interest both from a practical and purely fundamental point of view.

If time-reversal symmetry is broken, e.g., by means of a magnetic field $\boldsymbol{B}$, then one does not expect the Seebeck coefficient to be, in general, symmetric with respect to the magnetic field. Yet for the particular case of noninteracting systems, one has $S(\boldsymbol{B}) = S(-\boldsymbol{B})$ as a consequence of the symmetry properties of the scattering matrix.[2] Even though this constraint does not apply when interactions or inelastic scattering are taken into account, and even though there are no general results imposing the symmetry of the Seebeck coefficient, the latter has always been found to be an even function of the magnetic field in purely metallic two-terminal mesoscopic systems.[3] On the other hand, Andreev interferometer experiments[4] and recent theoretical studies indicate that systems in contact with a superconductor[5] or with a heat bath[6] can exhibit nonsymmetric thermopower. However, accurate numerical simulations of various models of two-terminal purely Hamiltonian *interacting* dynamical systems, which violate time-reversal symmetry, such as a two-dimensional anisotropic and inhomogeneous system of interacting particles in a perpendicular magnetic field,[7] systematically failed to find a nonsymmetric thermopower $S(\boldsymbol{B}) \neq S(-\boldsymbol{B})$. Therefore, it remains a completely open and interesting problem to understand what requirements must be fulfilled in order to *actually* lead to a thermopower which is asymmetric in the magnetic field.

In this Rapid Communication we show that the thermopower is, in general, asymmetric when inelastic scattering is added to the system, even though the system is noninteracting. Indeed, in the noninteracting case the symmetry of the thermopower is a consequence of the unitarity of the scattering matrix, which is broken when noise is added. A very convenient way to introduce noise is by means of a third terminal, whose parameters (temperature and chemical potential) are chosen self-consistently so that there is no average flux of particles and heat between the terminal and the system. In mesoscopic physics, such a third terminal, or a "conceptual probe," is commonly used to simulate phase-breaking processes in partially coherent quantum transport, since it introduces phase relaxation without energy damping.[8] We also show that, as a consequence of the asymmetry of the Seebeck coefficient, a weak magnetic field generally improves either the efficiency of thermoelectric power generation or of refrigeration, the efficiencies of the two processes being no longer equal when a magnetic field is added. Our findings are illustrated by the example of a realistic, asymmetric three-dot ring structure pierced by an Aharonov-Bohm flux. A main advantage of this model is that it can be analyzed exactly, without resorting to approximations.

General setup. The model we consider is sketched in Fig. 1. A system is in contact with left (L) and right (R) reservoirs (terminals) at temperatures $T_L = T + \Delta T$, $T_R = T$

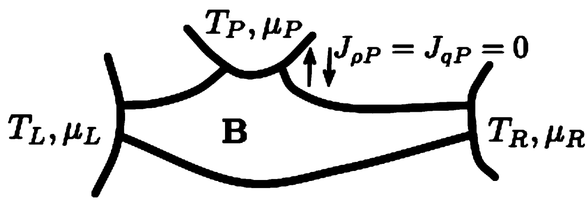

FIG. 1. Schematic drawing of the model. The third (probe) reservoir mimics inelastic scattering.

(without loss of generality, we assume $T_L > T_R$) and chemical potentials $\mu_L = \mu + \Delta\mu$, $\mu_R = \mu$. Both electric and heat currents flow along the horizontal axis. Inelastic scattering effects are simulated by means of a third (probe) reservoir (P) at temperature $T_P = T + \Delta T_P$ and chemical potential $\mu_P = \mu + \Delta\mu_P$. Let $J_{\rho k}$ and J_{Ek} denote the particle and energy currents from the kth reservoir ($k = L,R,P$) into the system, with the steady-state constraints of charge and energy conservation $\sum_k J_{\rho k} = 0, \sum_k J_{Ek} = 0$. The sum of the entropy production rates at the reservoirs reads $\dot{S} = \sum_k (J_{Ek} - \mu_k J_{\rho k})/T_k$. Within a linear response, $\dot{S} = \boldsymbol{J} \cdot \boldsymbol{X} \equiv \sum_{i=1}^4 J_i X_i$, where we have defined the four-dimensional vectors $\boldsymbol{J}$ and $\boldsymbol{X}$ as

$$\boldsymbol{J} = (eJ_{\rho L}, J_{qL}, eJ_{\rho P}, J_{qP}), \tag{2}$$

$$\boldsymbol{X} = \left(\frac{\Delta\mu}{eT}, \frac{\Delta T}{T^2}, \frac{\Delta\mu_P}{eT}, \frac{\Delta T_P}{T^2} \right), \tag{3}$$

and where the heat currents $J_{qk} \equiv J_{Ek} - \mu J_{\rho k}$ and e is the electron charge. The equation connecting the fluxes J_i and the thermodynamic forces X_i within linear irreversible thermodynamics is[9]

$$\boldsymbol{J} = \boldsymbol{L}\boldsymbol{X}, \tag{4}$$

where $\boldsymbol{L}$ is a 4×4 Onsager matrix, and $\boldsymbol{J}$, $\boldsymbol{X}$ must be written as column vectors.

The probe reservoir is adjusted in such a way that $J_3 = J_4 = 0$, that is, the net particle and heat flow from the probe into the system vanishes. It is convenient to write Eq. (4) in the block matrix form

$$\begin{pmatrix} \boldsymbol{J}_\alpha \\ \boldsymbol{J}_\beta \end{pmatrix} = \begin{pmatrix} \boldsymbol{L}_{\alpha\alpha} & \boldsymbol{L}_{\alpha\beta} \\ \boldsymbol{L}_{\beta\alpha} & \boldsymbol{L}_{\beta\beta} \end{pmatrix} \begin{pmatrix} \boldsymbol{X}_\alpha \\ \boldsymbol{X}_\beta \end{pmatrix}, \tag{5}$$

where $\boldsymbol{\alpha}$ stands for (1,2) and $\boldsymbol{\beta}$ for (3,4). The self-consistency condition $\boldsymbol{J}_\beta = (J_3, J_4) = 0$ implies $\boldsymbol{X}_\beta = -{\boldsymbol{L}_{\beta\beta}}^{-1}\boldsymbol{L}_{\beta\alpha}\boldsymbol{X}_\alpha$, so that

$$\boldsymbol{J}_\alpha = \boldsymbol{L}'\boldsymbol{X}_\alpha, \quad \boldsymbol{L}' \equiv \boldsymbol{L}_{\alpha\alpha} - \boldsymbol{L}_{\alpha\beta}{\boldsymbol{L}_{\beta\beta}}^{-1}\boldsymbol{L}_{\beta\alpha}. \tag{6}$$

The problem has then been reduced to two coupled fluxes

$$\begin{pmatrix} J_1 \\ J_2 \end{pmatrix} = \begin{pmatrix} L'_{11} & L'_{12} \\ L'_{21} & L'_{22} \end{pmatrix} \begin{pmatrix} X_1 \\ X_2 \end{pmatrix}, \tag{7}$$

where the reduced 2×2 Onsager matrix matrix $\boldsymbol{L}'$ fulfills the Onsager-Casimir relations

$$L'_{ij}(\boldsymbol{B}) = L'_{ji}(-\boldsymbol{B}) \quad (i,j = 1,2). \tag{8}$$

We would like to draw the reader's attention to the fact that the matrix $\boldsymbol{L}'$ is the Onsager matrix for two-terminal noisy transport, with noise modeled by means of a self-consistent reservoir. In particular, the Seebeck and the Peltier coefficients are given by $S = L'_{12}/(eTL'_{11})$ and $\Pi = L'_{21}/(eL'_{11})$. The thermopower is asymmetric when $L'_{12}(\boldsymbol{B}) \neq L'_{21}(\boldsymbol{B})$, i.e., $\Pi \neq ST$.

A key point is that, since $J_3 = J_4 = 0$, J_1 is the charge current from the left to right reservoir and the heat is extracted from (for power generation) or dissipated to (for refrigeration) the left (or right) reservoir only. Therefore, we can apply the analysis developed in Ref. 1. In particular, the efficiency depends on the asymmetry parameter x and on the "figure of merit" parameter y

$$x = \frac{L'_{12}}{L'_{21}}, \quad y = \frac{L'_{12}L'_{21}}{\det \boldsymbol{L}'}. \tag{9}$$

For power generation ($J_2 > 0$ and output power $\omega = -J_1\Delta\mu = -J_1 eTX_1 > 0$) the efficiency $\eta = \omega/J_2$ has a maximum value

$$\eta_{\max} = \eta_C x \frac{\sqrt{y+1} - 1}{\sqrt{y+1} + 1}, \tag{10}$$

while for refrigeration ($J_2 < 0$, $\omega < 0$) the maximum of the efficiency $\eta^{(r)} = J_2/\omega$ is

$$\eta^{(r)}_{\max} = \eta_C \frac{1}{x} \frac{\sqrt{y+1} - 1}{\sqrt{y+1} + 1}. \tag{11}$$

Noninteracting systems. Exact calculation of thermopower and efficiencies is possible for noninteracting models by means of the Landauer-Büttiker approach. We start from the bilinear Hamiltonian $H = H_S + H_R + H_C$, where the different terms correspond, respectively, to the nanoscale electronic system, the reservoirs, and the reservoir-system coupling. The tight-binding N-site system Hamiltonian reads

$$H_S = \sum_{n,n'=1}^{N} H_{nn'} c_n^\dagger c'_n, \tag{12}$$

where c_n and $c_n^\dagger$ are fermionic annihilation and creation operators. The reservoirs are modeled as ideal Fermi gases $H_R = \sum_{k,q} E_q c_{kq}^\dagger c_{kq}$, where $c_{kq}^\dagger$ creates an electron in the state q in the kth reservoir. The coupling (tunneling) Hamiltonian

$$H_C = \sum_{k,q} (t_{kq} c_{kq}^\dagger c_{i_k} + t_{kq}^* c_{kq} c_{i_k}^\dagger) \tag{13}$$

establishes the contact between site i_k and reservoir k.[10]

The charge and heat currents from the left terminal (reservoir) are given by[11]

$$J_1 = \frac{e}{h} \int_{-\infty}^{\infty} dE \sum_k [T_{kL}(E) f_L(E) - T_{Lk}(E) f_k(E)], \tag{14}$$

$$J_2 = \frac{1}{h} \int_{-\infty}^{\infty} dE (E - \mu_L) \sum_k [T_{kL}(E) f_L(E) - T_{Lk}(E) f_k(E)], \tag{15}$$

where $f_k(E) = \{\exp[(E - \mu_k)/k_B T_k] + 1\}^{-1}$ is the Fermi function and T_{kl} is the transmission probability from terminal l to terminal k. Analogous expressions can be written for J_3 and J_4, provided the terminal L is substituted by P.

The Onsager coefficients L_{ij} are obtained from the linear response expansion of the currents J_i. We have

$$L_{11} = \frac{e^2}{h}\int_{-\infty}^{\infty} dE \sum_{k\neq L} T_{Lk}(E)F(E), \tag{16}$$

$$L_{12} = L_{21} = \frac{e}{h}\int_{-\infty}^{\infty} dE(E-\mu) \sum_{k\neq L} T_{Lk}(E)F(E), \tag{17}$$

$$L_{22} = \frac{1}{h}\int_{-\infty}^{\infty} dE(E-\mu)^2 \sum_{k\neq L} T_{Lk}(E)F(E), \tag{18}$$

where $F(E) \equiv -Tf'(E) = 1/4k_B\cosh^2[(E-\mu)/2k_BT]$. Analogous formulas are obtained for L_{33}, $L_{34} = L_{43}$, and L_{44}, with the P terminal used instead of L. Note that, for the noninteracting three-terminal model, L_{12} is still an even function of the magnetic field, that is, $L_{12} = L_{21}$. On the other hand, the symmetry of the off-diagonal matrix elements is broken for the reduced Onsager matrix $\boldsymbol{L}'$. Indeed, reduction (6) involves other off-diagonal matrix elements of $\boldsymbol{L}$—between "left" (1,2) and "probe" (3,4) sectors—which, in general, are not even functions of an applied magnetic field. The block $\boldsymbol{L}_{\alpha\beta}$ of matrix $\boldsymbol{L}$ is given by

$$\boldsymbol{L}_{\alpha\beta} = -\frac{e^2}{h}\int_{-\infty}^{\infty} dE \begin{pmatrix} 1 & \frac{E-\mu}{e} \\ \frac{E-\mu}{e} & \left(\frac{E-\mu}{e}\right)^2 \end{pmatrix} T_{LP}(E)F(E), \tag{19}$$

and $\boldsymbol{L}_{\alpha\beta} \neq \boldsymbol{L}_{\beta\alpha}$, since $\boldsymbol{L}_{\beta\alpha}$ is obtained from $\boldsymbol{L}_{\alpha\beta}$ after substitution of T_{LP} with T_{PL} and, in general, $T_{LP} \neq T_{PL}$.

The transmission probabilities are given by[2]

$$T_{pq} = \mathrm{Tr}[\Gamma_p(E)G(E)\Gamma_q(E)G^\dagger(E)], \tag{20}$$

where the broadening matrices Γ_k are defined in terms of the self-energies Σ_k: $\Gamma_k(E) \equiv i[\Sigma_k(E) - \Sigma_k^\dagger(E)]$ and the (retarded) system Green function $G(E) \equiv [E - H_S - \sum_k \Sigma_k(E)]^{-1}$.

Aharonov-Bohm interferometer. As an illustrative, realistic example we consider a three-dot ring structure pierced by an Aharonov-Bohm flux, with dot k coupled to reservoir k, as sketched in Fig. 2. The system Hamiltonian reads

$$H_S = \sum_k \epsilon_k c_k^\dagger c_k + (t_{LR}c_R^\dagger c_L e^{i\phi/3} + t_{RP}c_P^\dagger c_R e^{i\phi/3} + t_{PL}c_L^\dagger c_P e^{i\phi/3} + \mathrm{H.c.}), \tag{21}$$

and the broadening matrices are $\Gamma_k = \gamma_k c_k^\dagger c_k$. We apply the Landauer-Büttiker approach to this model, numerically computing the Onsager coefficients following Eqs. (16)–(20).

As expected, we obtain asymmetric off-diagonal reduced Onsager matrix elements, that is, $L'_{12} \neq L'_{21}$, as far as the Aharonov-Bohm flux ϕ is nonvanishing and there is anisotropy in the systems, for instance, when $\epsilon_L \neq \epsilon_R$. Since the thermopower is not symmetric with respect to the magnetic field, i.e., $L'_{12}(\boldsymbol{B}) \neq L'_{12}(-\boldsymbol{B}) = L'_{21}(\boldsymbol{B})$, then, in general, the ratio $x = L'_{12}/L'_{21} \neq 1$. The asymmetry parameter x can be made arbitrarily small when $L'_{12} \to 0$ or arbitrarily large when $L'_{21} \to 0$—see, for instance, Fig. 3.

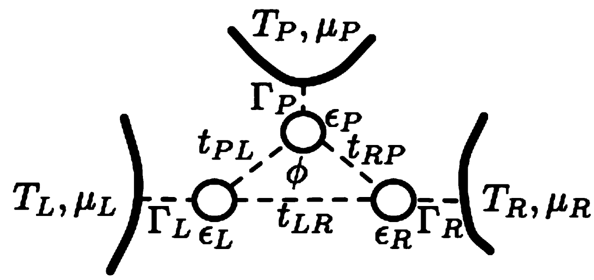

FIG. 2. Schematic drawing of the three-dot model.

Remarks. Large asymmetries do not imply, *ipso facto*, large efficiencies, for example, in the case of Fig. 3 when x diverges the figure of merit y and the efficiency tend to zero. It is, however, interesting to compare the efficiencies of power generation and refrigeration. While in the time-symmetric case the two efficiencies coincide $\eta_{\max}(\phi=0) = \eta_{\max}^{(r)}(\phi=0)$, this is no longer the case when $x \neq 1$. For small fields, x is, in general, a linear function of the field $x(\phi) = 1 + \alpha\phi + O(\phi^2)$, while y is by construction an even function of the field, so that $y(\phi) = y(0) + \beta\phi^2 + O(\phi^4)$. From Eqs. (10) and (11) we obtain $\eta_{\max}(\phi) = \eta_{\max}(0)[1 + \alpha\phi + O(\phi^2)]$ and $\eta_{\max}^{(r)}(\phi) = \eta_{\max}(0)[1 - \alpha\phi + O(\phi^2)]$. Therefore, a small external magnetic field either improves power generation and worsens refrigeration or vice versa, while the average efficiency $\bar{\eta} \equiv [\eta_{\max}(\phi) + \eta_{\max}^{(r)}(\phi)]/2 = \eta_{\max}(0)$ up to second-order corrections. Due to the Onsager-Casimir relations, $x(-\phi) = 1/x(\phi)$, and therefore by inverting the direction of the magnetic field one can improve either power generation or refrigeration.

In conclusion, we have shown that inelastic scattering generally leads to a thermopower which is a nonsymmetric function of the magnetic field. Such a general result has been illustrated by means of a realistic three-dot Aharonov-Bohm

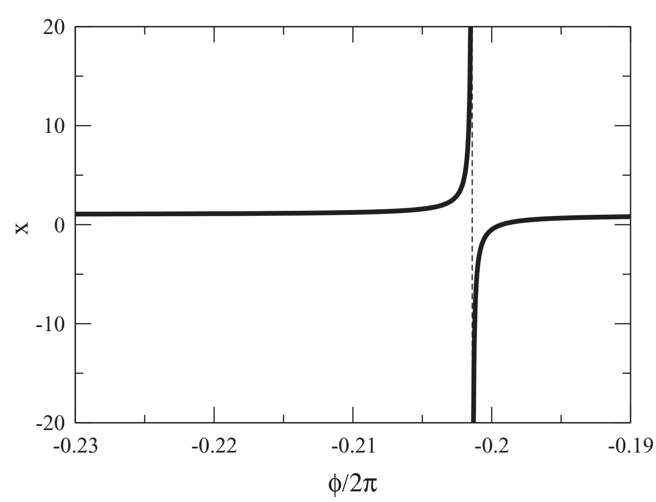

FIG. 3. Ratio x of the off-diagonal matrix elements L'_{12} and L'_{21} of the reduced Onsager matrix at $T=1$, $\mu=0.3$, $\epsilon_L=0$, $\epsilon_R=0.5$, $\epsilon_P=1$, all hopping terms $t_{pq}=-1$, and broadenings $\gamma_k=0.1$, independently of energy (wide-band limit). Hereafter we set $e=\hbar=k_B=1$.

interferometer model, which appears suitable for experimental investigations by means of three-terminal mesoscopic devices. The asymmetry of the Seebeck coefficient with respect to the magnetic field allows, in principle, *in the linear response regime*, to obtain a finite power at Carnot efficiency. Whether this is actually the case remains an interesting open problem. An additional interesting open problem is whether noiseless interacting systems might exhibit asymmetric thermopower. In our Rapid Communication, we have introduced the third lead to take dissipation into account phenomenologically. We hope that our work motivates further studies in order to gain a better understanding of the mechanisms leading to large asymmetry of the thermopower in realistic situations.

Recently, we became aware of a related work,[12] showing that in a setup with inelastic scattering mimicked by a third (probe) reservoir, the Seebeck coefficient is magnetic field asymmetric only when the Sommerfeld expansion is carried out beyond leading order.

Acknowledgments. K.S. was supported by MEXT, Grant No. (23740289), G.B. and G.C. by the MIUR-PRIN 2008 and by Regione Lombardia, and T.P. by the Grant Nos. J1-2208 and P1-0044 of the Slovenian Research Agency.

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