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Thermoelectric power fluctuations

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We consider the fluctuations of the thermoelectric transport coefficients due to quantum interference. At low temperatures, the relative fluctuations in these coefficients are substantially enhanced over the relative conductance fluctuations and result in random sign changes in the coefficients. We suggest a simple experiment to measure these effects for $L > L_{\text{in}}$.

It has been recognized recently that quantum transport is inherently non-self-averaging due to intrinsic interference effects.[1,2] Thus conductance fluctuations are size independent at low temperature,[1,2] in violation of simple statistical arguments. Experimental systems confirming this have included metallic wires and rings[3,4] and metal-oxide-semiconductor field-effect transistors (MOSFET's).[5] It is of interest to investigate how quantum interference manifests itself in other transport properties. Already this has led to the observation of a zero-field transverse (Hall-like) conductance[6] and anomalous magnetic field asymmetry of the conductance.[7-9] In this paper, we will investigate non-self-averaging aspects of thermoelectric effects.

Our calculation is based on the fact that in the absence of inelastic scattering, thermoelectric coefficients, like the Peltier coefficient, the thermopower, and others, can be related to the conductance at finite temperature,[10,11] which in turn can be related to the dependence of the conductance on the chemical potential. Thus, fluctuations in the thermoelectric coefficients are closely tied to the correlation in the conductance fluctuations between states of different chemical potential. We will find that these thermoelectric fluctuations grow with temperature until either the inelastic lifetime τ_{in} is less than the time τ_D taken for the electrons to diffuse across the sample or $k_B T > \hbar \tau_D^{-1}$.

We begin with the current response to an electric field and a thermal gradient

$$\mathbf{j} = \sigma \mathbf{E} - \eta \nabla T \ . \tag{1}$$

Since we are dealing with random samples, the local equation (1) is not well defined; instead, we use the conductance G and the corresponding quantity for η denoted by N. We then have

$$I = GV - N\Delta T \ , \tag{2}$$

with I as the current, V as the voltage drop, and ΔT as the temperature difference between the two ends.

Provided the quasiparticles are noninteracting and all scatterings are elastic, it has been shown that[10,11]

$$N = -\frac{1}{eT} \int \frac{df}{d\varepsilon} (\varepsilon - \mu) G(\varepsilon) d\varepsilon \ , \tag{3}$$

where $G(\epsilon)$ is the zero-temperature conductance at chemical potential ϵ, μ is the physical chemical potential, and f the Fermi distribution. If, in addition, the Sommerfeld expansion is valid, (3) becomes

$$N = \frac{\pi^2}{3e} k_B (k_B T) G'(\mu) \ , \tag{4}$$

that is, N is proportional to the derivative of the conductance with respect to the chemical potential. Note that common textbook proofs notwithstanding, (3) and (4) are not based on the relaxation time approximation.

Following the reasoning of Ref. 1, we define

$$(\delta N)^2 = \langle (N - \langle N \rangle)^2 \rangle \ , \tag{5}$$

with $\langle \ \rangle$ implying the impurity average. This gives the fluctuations of N within an ensemble of macroscopically "identical" samples, and via the ergodic assumption, the fluctuations under changing magnetic fields or chemical potentials in the same sample.[1] Using Eq. (3), we have

$$(\delta N)^2 = \frac{1}{e^2 T^2} \int \int \frac{df}{d\varepsilon} \frac{df}{d\varepsilon'} (\varepsilon - \mu)(\varepsilon' - \mu) \times F(\varepsilon - \varepsilon') d\varepsilon d\varepsilon' \ , \tag{6}$$

where

$$F(x) = \langle G(\mu) G(\mu + x) \rangle - \langle G(\mu) \rangle \langle G(\mu + x) \rangle \tag{7}$$

is the conductance correlation function at zero temperature. In Ref. 1, the function $F(x)$ was calculated in order to study the chemical potential correlation in the conductance. F behaves like

$$F(x) \approx \left(\frac{e^2}{\hbar} \right)^2 a_d \left[1 - \left(\frac{x^2}{E_c} \right) \right], \ x < E_c \ , \tag{8a}$$

$$\approx 0, \ x > E_c \ , \tag{8b}$$

with a_d of order unity and weakly dependent on the effective dimension d. The chemical-potential change at which conductances become uncorrelated is given by

$$E_c \approx \frac{\hbar \pi^2 D}{L_x^2} \ . \tag{9}$$

In (9), D is the diffusion constant and L_x is the sample size in the direction of the current. The behavior of $(\delta N)^2$ is as follows.

(i) $k_B T < E_c$: In this case $F(\varepsilon - \varepsilon')$ varies little on the scale of $k_B T$. The Sommerfeld expansion is valid, and yields

$$\delta N = \frac{\pi^2}{3} \frac{k_B^2}{e} T[-F''(0)]^{1/2} = \frac{\pi^2}{3e} k_B^2 T (\delta G)' \ , \tag{10}$$

where the last equality follows from (4). Then from Eq. (8a),

$$\delta N \approx \frac{\pi^2}{3e} k_B \frac{e^2}{\hbar} \sqrt{2a_d} \left(\frac{k_B T}{E_c} \right) \ . \tag{11}$$

Unlike δG, δN depends on the size and the degree of disorder, δN going as L_x^2/D. Since in this regime $\langle N \rangle \sim \langle G' \rangle$, and $\langle G' \rangle \sim \langle G \rangle / E_F$,

$$\frac{\delta N}{N} \approx \left(\frac{E_F}{E_c} \right) \frac{\delta G}{\langle G \rangle} \ . \tag{12}$$

Thus, the relative fluctuation in N is enhanced over that of G by a factor E_F/E_c. From reported experimental values of E_F, $\langle G \rangle$, etc., $\delta N / \langle N \rangle$ ranges from ~ 1 (MOSFET) to $\sim 10^2$ (gold rings and wires). That $\delta N / \langle N \rangle$ is large is not surprising since $\langle N \rangle$ must have the same sign for a given material, while $N \sim G'$ fluctuates in sign as the chemical potential is changed.

(ii) $k_B T > E_c$: The linear increase of N with T and L_x^2 ceases when $k_B T \gtrsim E_c$. In this case, in Eq. (6), $F(\varepsilon - \varepsilon')$ varies rapidly on the scale of $k_B T$, and the Sommerfeld expansion breaks down. Since $F(\varepsilon - \varepsilon')$ is nonnegligible only for $|\varepsilon - \varepsilon'| < E_c$, this leads to $\delta N \sim (E_c / k_B T)^{1/2}$ for $k_B T > E_c$. Thus, δN increases as $k_B T / E_c$ until $k_B T > E_c$, whence it decreases as $(E_c / k_B T)^{1/2}$. The peak value of δN, at $k_B T \approx E_c$, is

$$(\delta N)_{\max} \approx \frac{1}{e} \frac{\pi^2}{3} k_B \frac{e^2}{\hbar} \sqrt{2a_d} \ . \tag{13}$$

Note that $(\delta N)_{\max}$ is independent of size, dimension, and degree of disorder. Samples with larger temperature slopes, due to smaller D or larger L_x^2, peak at lower temperatures. The discussion so far has been confined to elastic scattering. Although Eq. (3) has strictly been shown to be correct only for that case, we will assume its applicability, even with elastic scattering, our view being that the principle effect of such scattering is to destroy quantum coherence. The relevant length scale is L_{in}, the inelastic diffusion length.

For $L_x \gg L_{\text{in}}$ or, equivalently, $\hbar \tau_{\text{in}}^{-1} \gg E_c$, $\delta N / \langle N \rangle$ should scale[12] as $(L_{\text{in}}/L_x)^{d/2}$. Since $\langle N \rangle \sim L_x^{d-2}$, this implies

$$\delta N = (\delta N)_{\max} (T \tau_{\text{in}}) (E_c \tau_{\text{in}})^{1-d/4} \ , \tag{14}$$

with $(\delta N)_{\max}$ given by (13). Because $\tau_{\text{in}} \sim T^{-p}$, there is a crossover behavior from $\delta N \sim T$ to $\delta N \sim T^{1-2p+(dp/4)}$ as T increases. For example, in one dimension p is expected to be $\frac{2}{3}$, hence $\delta N \approx T^{-1/6}$, while in two dimensions $p = \frac{1}{2}$ and $\delta N \sim T^{1/4}$.

Depending on whether $k_B T$ or $\hbar \tau_{\text{in}}^{-1}$ becomes greater than E_c first, the temperature dependence of δN is given by $\delta N \sim (E_c / k_B T)^{1/2}$ or by Eq. (14). For a highly diffusive system τ_{in} should be very large ($\tau_{\text{in}} \sim D^{1/3}$ in one dimension, $\sim D$ in two dimensions); the former case applies and the universal $(\delta N)_{\max}$ can be observed. This is the situation for the gold rings[3,4] but not for the MOSFET's.[5]

Next we discuss the Peltier coefficient Π. For $\nabla T = \mathbf{0}$, the heat current due to an electric field is $I_H = N_H V$. Onsager's relation requires $N_H(B) = TN(-B)$, with B being the external magnetic field. The Peltier coefficient is defined as

$$\Pi = TN_H(B)/G(B) = TN_H(-B)/G(B) \ , \tag{15}$$

where G is now the conductance at temperature T, and for longitudinal conductance $G(B) = G(-B)$.

For a given G, $N \sim G'$ can be randomly positive or negative and relative fluctuations of G are small, so that $\langle N/G \rangle \approx \langle G^{-1} \rangle \langle N \rangle$. Assuming that $\langle G^{-1} \rangle \approx \langle G \rangle^{-1}$, we find that

$$\langle \Pi \rangle \approx T \langle N \rangle / \langle G \rangle \ . \tag{16}$$

Since $\delta N / \langle N \rangle \gg \delta G / \langle G \rangle$, the fluctuation in Π is dominated by the fluctuation in N, yielding

$$\frac{\delta \Pi}{\langle \Pi \rangle} \approx \frac{\delta N}{\langle N \rangle} \ . \tag{17}$$

For the thermopower Q, we use Kelvin's theorem, $\Pi = QT$, which is based on thermodynamic considerations and holds generally (but see the following discussion).

The analysis thus far has relied on Eqs. (4) and (15) which are unaveraged relations and hold for a given sample. If the same experimental setup is used to measure G, L, and Q then, while they will all undergo random fluctuations with, say, changing chemical potential, the fluctuations are not independent and these equations hold at a given chemical potential. If different setups are used, then the fluctuations become independent. Also, if the thermopowers Q_1 and Q_2 of two samples are measured, their relative thermopower when placed together (actually three samples are needed) is

$$Q_{12} = Q_1 - Q_2 + O(\delta Q) \neq Q_1 - Q_2 \ .$$

In particular, if $Q_1 = Q_2$, Q_{12} will not be zero.

In summary, we have considered the fluctuations of thermoelectric coefficients. These fluctuations may be due to a chemical-potential or magnetic field change in one sample, or they may be fluctuations between samples.[1,2] We have restricted ourselves to diagonal or longitudinal components. We note here that, since $G_{xy} \neq 0$ in zero field,[6] N_{xy} will be nonzero also. The consequence of this will be reported elsewhere. In the regime where $L_x < L_{\text{in}}$, the maximum fluctuation, at $k_B T = E_c = \hbar \Pi^2 D / L_x^2$, of N is "Universal," while that of $Q \sim 1/\langle G \rangle$, and of $\Pi \sim E_c / \langle G \rangle$. In all cases, they are anomalous for $k_B T \lesssim E_c$, and do not vary with size or dimension as they do in classical statistics. Since the relative fluctuations are much enhanced over the conductance fluctuations $[\delta N/N \sim \delta\Pi/\Pi \sim \delta Q/Q \sim (E_F/E_c)(\delta G/G)]$, and can result in random sign changes of the thermoelectric effects, they should be observable even in spite of the difficulties associated with such measurements in small systems.

While it is difficult to set up a controlled temperature difference between the ends of samples of size greater than L_{in}, the thermopower fluctuations in samples of size less than L_{in} can be effectively enhanced over those in the smaller samples, as we will now explain. To probe these thermoelectric effects, we suggest that fluctuations in the induced magnetic moment, M, be measured rather than the fluctuations in the current. This is because $\delta M \sim (\delta I)L^2$, while $\delta I \sim L^{-1/2}$ for $L > L_{\text{in}}$. An idealized configuration for this measurement is presented in Fig. 1. Here, wires a and b of the same material and length L $(L > L_{\text{in}})$ are connected by large electrodes which are maintained at a temperature difference ΔT. If G is the conductance of each electrode between the a and b contacts, then the circulating thermoelectric current is

$$\frac{I}{\Delta T} = \frac{N_a f_b - N_b f_a}{1 + f_a f_b (G_a + G_b)/G} ,$$

where $f_a = G_a(G_a + G_b)^{-1}$ and similarly for f_b. For $G \gg G_a \sim G_b$, and because $\delta G/G \ll \delta N/N$, Eq. (15) becomes

$$\frac{I}{\Delta T} = \tfrac{1}{2}(N_a - N_b) .$$

This circulating current creates a magnetic moment $M = \zeta I L L'$, where $\zeta \sim 1$. The fluctuation of M about zero is then

$$\delta M = \zeta (L_{\text{in}} L L'^2)^{1/2} \delta N_1 \Delta T , \tag{18}$$

where δN_1 is the fluctuation of δN when $L = L_{\text{in}}$. This experiment is subject to less stringent size restrictions than measurements of conductance fluctuations.

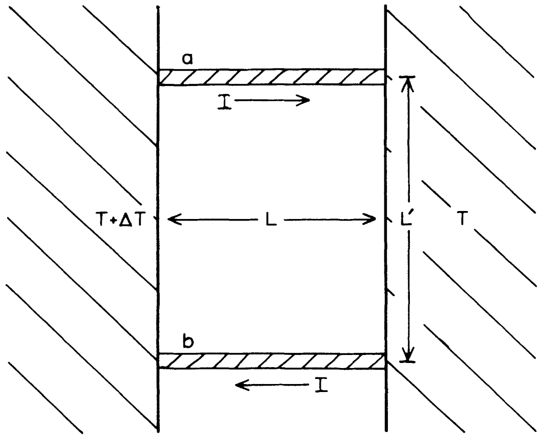

FIG. 1. Schematic of a setup to observe thermopower fluctuations, more specifically, fluctuations of the coefficient N. If the bridges a and b are equivalent on average, the variation of the moment with change of chemical potential or external magnetic field will be equally often positive as negative.

Note added in proof. For $k_B T \gg E_c$, the important contribution to δN comes from the large-x regime of $F(x)$ and the approximation in Eq. (8) is insufficient. Instead, the relatively slow power decay of $F(x)$ should be used. While the $T^{-1/2}$ falloff in δN given above Eq. (13) is still correct for $d \leq 2$, it changes to a slower $T^{(d-4)/4}$ for $d \geq 2$. We thank B. Serota for pointing this out to us.

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[12] Actually this is only correct if $\delta N/N \ll 1$, which is not true in the present case. Thus, there should be an intermediate regime, when $L_x \gtrsim L_{\text{in}}$, where the drop off in $\delta N/N$ differs from $(L_x)^{d/2}1$.