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Fluctuating local thermoelectric heat in dirty metals

David P. DiVincenzo
IBM Research Division, Thomas J. Watson Research Center, Yorktown Heights, New York 10598

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Using a recently developed multilead theory of dephasing in mesoscopic conductors, the mean-squared magnitude of the local Peltier heat in a uniform disordered metal is calculated diagrammatically. A heuristic estimate based on conductance fluctuation theory is also developed, and gives the same results. The generation and absorption of local thermoelectric heats require both phase-coherent elastic scattering to produce local conductance fluctuations and phase-breaking inelastic scattering to transport heat to and from the reservoirs. This phenomenon can cause substantial spatial variations in the electron temperature of low-carrier-density, clean, quasi-two-dimensional metals.

INTRODUCTION

After some years of intense effort, it is now well understood how wave-mechanical effects radically alter the phenomenology of electric conduction in metals in which elastic scattering dominates. Electric resistances are strongly sample specific, are spatially nonlocal, and fluctuate rapidly with electron density or magnetic field, all because of random wave-interference effects.[1] A key parameter is the inelastic scattering length $L_{\rm in}$; on scales shorter than $L_{\rm in}$ wave effects prevail, while a classical description is correct on longer scales. As I will demonstrate in this paper, a good test of our understanding of inelastic scattering is provided by the local thermoelectric response of a metal, because it is sensitive to the real energy flows in and out of the electron system and not just to phase-breaking effects alone. Here a recent formulation of inelastic-scattering theory[2,3] is used to calculate the magnitude of local Peltier heats. They turn out to be strongly fluctuating, spatially inhomogeneous, and sample specific in the quantum regime.

Figure 1(a) reviews the classical Peltier effect, which results when an electric current J^e is passed through a couple between two metals M_1 and M_2 under isothermal conditions.[4] As I discuss below [see Eq. (10)], the particle current is accompanied by a heat current J^Q. In general J^Q is different in the two metals, so that an excess or deficit of heat $dQ/dt = \dot{Q}$ appears at the boundary between the two metals; this is the *Peltier heat.* It should be imagined that constant-temperature conditions are established by there being good contact, e.g., with a high thermal-conductivity substrate. The Peltier heat $\dot{Q}$ is supplied or carried away by that substrate, and in principle it can be measured there.

Existing quantum-transport theories[5–7] model the Peltier effect poorly. In particular, as stated explicitly by Serota, Ma, and Goodman,[5] these previous treatments obey the Chester-Thellung relation between the electric current density $\mathbf{j}^e(\mathbf{r})$ and the heat current density $\mathbf{j}^Q(\mathbf{r})$:

$$\mathbf{j}^Q(\mathbf{r}) = (\pi^2/3e)(k\Theta)^2 \partial \mathbf{j}^e(\mathbf{r})/\partial E_F. \qquad (1)$$

(Here Θ is the temperature, E_F the Fermi energy.) This equation plus particle conservation $\nabla \cdot \mathbf{j}_e = 0$ implies conservation of heat current: $\nabla \cdot \mathbf{j}_Q = \dot{Q} = 0$. Therefore, in this approximation there is no Peltier effect.

The problem is that while most of these previous treatments include a cutoff from dephasing scattering in the particle-hole and particle-particle propagators (diffusons and cooperons), they do not permit the actual transport of energy in or out of the electron system. Some recent formulations of inelastic scattering within a quantum-kinetic-equation approach[2,3] cure this problem. This re-

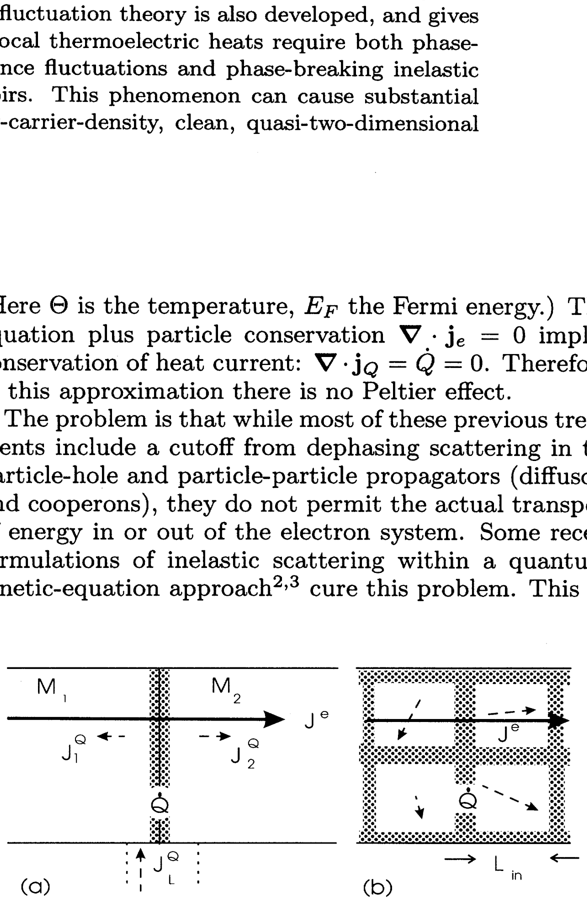

FIG. 1. (a) Classical Peltier effect. When a uniform electric current J^e (solid arrow) is passed through the junction between two different metals M_1 and M_2, Peltier heat $\dot{Q}$ is absorbed (or emitted) at the boundary. This happens because in each metal, the electric current is accompanied by a uniform heat current J^Q (dashed arrows), but its magnitude and sign is a materials-dependent property, and will differ in the two metals. In a quantum-transport description, this heat $\dot{Q}$ can be extracted from a reservoir as a heat current J_L^Q from an additional lead (shown dotted). (b) Heuristic description of the mesoscopic Peltier effect. If the system is considered to be divided into phase-coherent subregions of size $L_{\rm in}$, a uniform electric current J^e (solid arrow) will cause heat currents (dashed arrows) to flow. The size of these heat flows is determined by the universal conductance fluctuations, and because of the sample-specific nature of quantum transport, their magnitude differs in magnitude and direction in each (statistically equivalent) region. Thus, as in (a), Peltier heats $\dot{Q}$ appear at the boundaries.

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cent work is based on an artifice originally suggested by Büttiker:[8] he pointed out that if an additional lead is connected to a quantum conductor [see Fig. 1(a)], the chemical potential of this lead can be tuned such that no particle current passes through the lead. Thus, this lead has no effect on the overall current flows in the conductor — it is a "voltage lead" in the experimentalists' language. Nevertheless, the presence of this lead both produces the effect of dephasing scattering and permits nonzero energy flows in or out of the electron system, mimicking the effect of real inelastic scattering. The dephasing results because an electron which passes into this lead is replaced by a reservoir electron with uncorrelated phase.[3] The thermal effect results because the injected electron need not have exactly the same energy as the original. Rather, its energy will be within $\pm k\Theta$ of E_F, which is an important part of the physics embodied in the Chester-Thellung formula Eq. (1).

THEORY

This observation of Büttiker's has been extended into a full theory of inelastic scattering by the simple expedient of attaching a fictitious voltage lead to every point $\mathbf{r}$ of a quantum conductor.[3] Formally, the heat current flowing into the lead attached at point $\mathbf{r}$, $J_L^Q(\mathbf{r})$ [$\equiv I^Q(\mathbf{r})$], can be taken to be the local Peltier heat $\dot{Q}(\mathbf{r})$ evolved (or absorbed) at that point. This multilead approach has recently been explored by Butcher;[9] it goes back to ideas of Sivan and Imry.[10] Of course, in a more realistic microscopic description, $\dot{Q}$ can only be defined on length scales of order $L_{\rm in}$; thus, to be interpreted realistically, the multilead results should be averaged over a coherence volume.

Extending the development of Hershfield,[3] a full diagrammatic treatment of the local Peltier heat in a disordered metal, which has a close parallel to the usual diagrammatic treatment of the nonlocal Kubo conductivity,[11,12] can be formulated. The basic linear-response equation for the local heat current is[9,3]

$$I^Q(\mathbf{r}) = \frac{\pi^2}{3}\frac{(k\Theta)^2}{h}\int d\mathbf{r}' T'(\mathbf{r},\mathbf{r}')\mu(\mathbf{r}'),$$

$$T(\mathbf{r},\mathbf{r}') = \Gamma_{\rm in}^2 |g(\mathbf{r},\mathbf{r}')|^2. \qquad (2)$$

Here $T(\mathbf{r},\mathbf{r}')$ is the transmission between fictitious leads at $\mathbf{r}$ and $\mathbf{r}'$, T' denotes its derivative with respect to Fermi energy, g is the one-particle Green function, $\Gamma_{\rm in}$ is the inelastic scattering rate determined by the coupling strength to the fictitious leads,[3] and $\mu(\mathbf{r})$ is the Fermi energy of the fictitious lead at $\mathbf{r}$, chosen such that the electric current into the lead is zero.

Equation (2) comes directly from applying the Chester-Thellung relation [Eq. (1)] to the basic constitutive equation of the multilead formalism:

$$I(\mathbf{r}) = \frac{e}{h}\int d\mathbf{r}'\ T(\mathbf{r},\mathbf{r}')\mu(\mathbf{r}'). \qquad (3)$$

The desired correlation function is easily constructed:

$$\langle I^Q(\mathbf{r}_1)I^Q(\mathbf{r}_3)\rangle = \frac{\pi^4}{9}\frac{(k\Theta)^4}{h^2}\int\int d\mathbf{r}_2 d\mathbf{r}_4 \langle T'(\mathbf{r}_1,\mathbf{r}_2)[\mu(\mathbf{r}_2)-\mu(\mathbf{r}_1)]T'(\mathbf{r}_3,\mathbf{r}_4)[\mu(\mathbf{r}_4)-\mu(\mathbf{r}_3)]\rangle. \qquad (4)$$

This equation is abtained simply by squaring Eq. (2). The $\mu(\mathbf{r}_1)$ and $\mu(\mathbf{r}_3)$ terms can be introduced because of the indentity

$$\int d\mathbf{r}_2 T(\mathbf{r}_1,\mathbf{r}_2) = 0, \qquad (5)$$

which follows from the equilibrium condition applied to Eq. (3), that the currents are zero if all the chemical potentials are equal.

Equation (4) contains an ensemble average over the product of four fluctuating quantities. We can argue that the leading contribution to this quantity is a factorized average over the separate pieces of this product:

$$\langle I^Q(\mathbf{r}_1)I^Q(\mathbf{r}_3)\rangle \approx \frac{\pi^4}{9}\frac{(k\Theta)^4}{h^2}\int\int d\mathbf{r}_2 d\mathbf{r}_4 \langle T'(\mathbf{r}_1,\mathbf{r}_2)T'(\mathbf{r}_3,\mathbf{r}_4)\rangle\langle\mu(\mathbf{r}_2)-\mu(\mathbf{r}_1)\rangle\langle\mu(\mathbf{r}_4)-\mu(\mathbf{r}_3)\rangle. \qquad (6)$$

We cannot factorize the average any further, because $\langle T'\rangle = 0$. The Appendix argues that corrections to the factorized average in Eq. (6) are of order $\lambda_F^2/(L_{\rm in}l_e)$ times the contribution in Eq. (6), so we are justified in ignoring them.

The chemical-potential averages in Eq. (6) are simply obtained in terms of the classical potential drop $\boldsymbol{\nabla}\mu$ along the conductor:

$$\langle\mu(\mathbf{r}_2)-\mu(\mathbf{r}_1)\rangle = (\mathbf{r}_2-\mathbf{r}_1)\cdot\boldsymbol{\nabla}\mu. \qquad (7)$$

The T' correlation function inside Eq. (6) is very similar to the correlation functions for the nonlocal Kubo conductivity $\langle\sigma(\mathbf{r}_1,\mathbf{r}_2)\sigma(\mathbf{r}_3,\mathbf{r}_4)\rangle$ studied by Kane, Serota, and Lee[11] and by Hershfield.[12] The lowest-order diagrams [in the usual expansion parameter for a diffusive metal, $(k_F l_e)^{-1}$, with k_F the Fermi wavevector and l_e the elastic mean free path] for the two problems have the same form. One of these diagrams is illustrated in Fig. 2; for a complete discussion, consult Hershfield.[12] The only differences between the evaluation of the diagrams

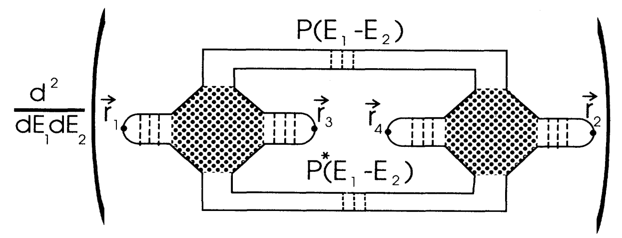

FIG. 2. One of the diagrams for the local thermoelectric fluctuations $\langle T'(\mathbf{r}_1,\mathbf{r}_2)T'(\mathbf{r}_3,\mathbf{r}_4)\rangle$ in Eq. (6). The solid lines denote single-particle Green functions, the dashed lines indicate the ladder sums of impurity scattering (i.e., diffusion propagators P), the dots indicate the scalar vertices, and the shaded regions are Hikami boxes. [This is very similar to a conductance fluctuation diagram; cf. Fig. 4(a) of Ref. 11.] The calculation requires the overall energy derivatives as shown, with $E_1 = E_2 = E_F$ at the end; I have indicated where the energy dependence appears inside the diagram by giving the energy arguments of the diffusion propagators. The other four diffusion ladders in the figure are evaluated at zero energy (i.e., on the Fermi surface).

for $\langle T'T'\rangle$ and $\langle\sigma\sigma\rangle$ are the following: (1) There are energy derivatives on the T's, which act on the diffusons in the diagrams as indicated in the figure. These derivatives cure some of the ultraviolet divergence problems encountered in the previous conductivity calculation. (2) The vertices are scalar (density) vertices rather than vector (current) vertices. A consequence of this is that the vertex dressing is important even in the translationally invariant case, unlike in the conductivity problem.[13] (3) Related to this is that the diffusons in the vertex dressing are cut off on the scale of the inelastic length; in the conductivity calculation they are not cut off. This can also be viewed as a consequence of the fact that the conductivity calculation satisfies a current-conservation condition, $\nabla \cdot \sigma(\mathbf{r},\mathbf{r}') = 0$, which does not apply to $T'(\mathbf{r},\mathbf{r}')$.

The result of evaluating the diagrams (for zero magnetic field) is the following:

$$\langle T'(\mathbf{r}_1,\mathbf{r}_2)T'(\mathbf{r}_3,\mathbf{r}_4)\rangle \approx \frac{1}{L_{\text{in}}^{4(d-1)}(hD)^2} \times f\left(\frac{\mathbf{r}_{12}}{L_{\text{in}}},\frac{\mathbf{r}_{13}}{L_{\text{in}}},\frac{\mathbf{r}_{14}}{L_{\text{in}}}\right). \tag{8}$$

This expression is correct for dimension $d = 2$ or 3. D is the diffusion constant of the metal, and f is a dimensionless function with the properties $f(0,0,0) \sim 1$; $f(x,y,z) << 1$ if x, y or $z > 1$. Unlike in the conductivity calculation, f is long-ranged in all of its variables (i.e., it is not proportional to any δ function), and it has not proved possible to obtain an analytic form for this function. Nevertheless, the parametric dependence of the mean-squared Peltier heat can be given; putting together Eqs. (6)–(8):

$$\langle I^Q(\mathbf{r}_1)I^Q(\mathbf{r}_3)\rangle \approx \frac{(k\Theta)^4}{h^2}\frac{1}{(hD)^2}L_{\text{in}}^{2(3-d)} \times \nabla\mu_i\nabla\mu_j g_{ij}\left(\frac{\mathbf{r}_{13}}{L_{\text{in}}}\right). \tag{9}$$

Here g_{ij} is another dimensionless function with the same properties as f. It should be noted that g_{ij} varies smoothly on the scale of L_{in}, which is consistent with the requirement that the physical I_Q cannot vary on scales much shorter than L_{in}.

SIMPLE PICTURE

The result of Eq. (9) is consistent with a very simple picture of the quantum conductor that has been developed by Imry and others[14] to explain the crossover to classical behavior in the presence of inelastic scattering. As Fig. 1(b) shows, in this picture the conductor is imagined to be divided into regions of size L_{in}. Inside each region, the conductor is perfectly phase coherent, with only elastic scattering; different regions add classically. Suppose that there is an overall potential gradient $\nabla\mu$ across the conductor, so that there is a potential drop $L_{\text{in}}\nabla\mu/e$ across a quantum-coherent domain. Then the conductance tensor G_{ij} of a domain is defined by $J_i^e = G_{ij}(L_{\text{in}}\nabla\mu_j/e)$; here $\mathbf{J}^e$ is the total electric current in a single domain. From the Chester-Thellung relation Eq. (1),[5] the corresponding relation for the total heat current $\mathbf{J}^Q$ in one region is

$$J_i^Q = \frac{\pi^2}{3e^2}(k\Theta)^2 L_{\text{in}}\frac{dG_{ij}}{d\mu}\nabla\mu_j, \qquad \frac{dG}{d\mu} \sim \frac{e^2/h}{hD/L_{\text{in}}^2}. \tag{10}$$

The tensor character of G_{ij} is retained to emphasize that the heat current need not be in the same direction as the potential gradient, as Fig. 1(b) illustrates. The estimate for the typical value of $dG/d\mu$ in Eq. (10) follows from universal conductance fluctuation theory:[1] the conductance is expected to fluctuate by e^2/h over the Thouless energy scale $\Gamma_{\text{in}} = hD/L_{\text{in}}^2$.

An estimate for the local Peltier heat is given by the continuity equation $I^Q(\mathbf{r}) = \nabla \cdot \mathbf{j}^Q$. The heat current density j^Q is related to the total heat current of Eq. (10) by $j^Q \approx J^Q/L^{d-1}$. J^Q varies on the scale of L_{in}, so that the divergence operator in the continuity equation gives another factor of $1/L_{\text{in}}$. Putting this together gives an estimate for I^Q:

$$I^Q \sim \frac{(k\Theta)^2}{h}\frac{L_{\text{in}}^{3-d}}{hD}\nabla\mu. \tag{11}$$

Comparing (11) with (9) shows that the heuristic argument agrees with the diagrammatic calculation, supporting this picture of the physics of the local Peltier heat.

Preliminary numerical studies also tend to confirm the above results. The simulations are on a two-dimensional strip using a lattice Hamiltonian and one-dimensional leads attached to every lattice site as suggested by Hershfield.[3] The simulations are done not for fixed L_{in} but rather for fixed inelastic scattering rate Γ_{in}. Using the free-electron relation $D = \hbar G/m$, where G is the two-dimensional conductance in units of e^2/h, Eq. (11) predicts

$$I^Q \propto \Gamma_{\rm in}^{-1/2} G^{-1/2} \nabla\mu. \tag{12}$$

The best fit to the dependences in the simulations is

$$I^Q \propto \Gamma_{\rm in}^{-0.9} G^{-0.4} \nabla\mu. \tag{13}$$

The exponents in Eq. (13) should not be taken too seriously; certain systematic errors, for example those due to the departures from the free-electron density of states in the lattice model, have not been fully corrected. Nevertheless, the agreement in the trends between the analytic and the preliminary numerical results is encouraging.

EXPERIMENTAL CONSEQUENCES

Finally, I comment on the effect which the local Peltier heat described in this paper would produce in an experimental geometry which has been used to study mesoscopic thermopower.[15] In this work a uniform current flowing along a channel is assumed to produce a spatially uniform Joule heating; I wish to point out that for certain length scales and excitation levels, the electron temperatures will be nonuniform because of the local Peltier effect. (At this point I relax the condition that the Peltier effect is isothermal, assuming that partial thermal equilibration occurs solely because of electronic thermal conductivity.) In Ref. 15, the elastic mean free path l_e is given as $l_e = 500$ Å, and from the reported electron-electron scattering time of 6×10^{-12} s, I deduce that $L_{\rm in}$ is also on the order of 500 Å. From these and the equation for the diffusion constant $D = v_F l_e/d$, I can use Eq. (11) to determine the local Peltier heat density for a given applied potential $\nabla\mu$. It is most useful to compare the result with the expression for the Joule heat density, $I^J \approx \langle G\rangle(\nabla\mu)^2$. The thickness of the semiconductor in Ref. 15 is reported to be 500 Å; Since this is comparable to $L_{\rm in}$, it is reasonable to use the two-dimensional versions of the equations with $\langle G\rangle \approx k_F l_e$. I find that when the applied potential is about $\nabla\mu/e \sim 640$ V/m, the Joule heat and the Peltier heat are comparable, being about 40 W/m^2.

Reference 15 is not explicit about the applied potential along the channel; it seems that theirs is substantially higher than 640 V/m. Nevertheless, this would not be an unreasonable applied potential in a mesoscopic experiment. The Peltier heat flows that this potential sets up will generate thermal gradients; counterflows will be set up because of the electronic thermal conductivity, and a steady state will be reached. Using the classical expression $\kappa = c_V v_F l_e/3$ for the thermal conductivity and the standard expression for the electronic specific heat c_V,[16] I can estimate the temperature variations that will result in steady state:

$$\frac{\Delta\Theta}{\Theta} \approx \frac{e^4 m^2}{h^8} \frac{(k_B\Theta)^2 L_{\rm in}^4}{\langle G\rangle^4}. \tag{14}$$

This ratio is quite small ($\sim 10^{-6}$) for the parameters of Ref. 15, but because of the strong dependence on the parameters $L_{\rm in}$ and $\langle G\rangle$, it could be made appreciable in a clean metallic film with low carrier density. The thermal gradients of Eq. (14) vary on a length scale of order $L_{\rm in}$, which is the shortest scale on which the temperature is well defined.

In conclusion, I have obtained predictions for the magnitude of local fluctuating Peltier heats using a recent multilead theory of inelastic scattering in metals. These predictions agree with a heuristic description in which this heat appears only at the boundaries between phase-coherent segments of the conductor. Estimates show that the local variations of the electron temperature that result from these heat flows would be large enough to have experimental consequences, especially for relatively clean, quasi-two-dimensional semimetals.

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APPENDIX: FACTORIZATION IN Eq. (6)

Here I present the arguments which lead to the conclusion that it is legitimate to factorize the average in Eq. (4), as done in Eq. (6). That is, I consider the approximations which can be made to the quantity

$$\langle T'_{12} T'_{34} \Delta\mu_{21} \Delta\mu_{43} \rangle, \tag{A1}$$

where I have introduced some obvious simplifications in the notation of Eq. (4). First, I introduce a decomposition of the chemical-potential differences into an average part and a fluctuating part:

$$\Delta\mu_{ij} \equiv \langle \Delta\mu_{ij} \rangle + \delta\Delta\mu_{ij}. \tag{A2}$$

With this the quantity in Eq. (A1) can be rewritten exactly:

$$\begin{aligned}
&\langle [\langle T'_{12}T'_{34}\rangle + \delta(T'_{12}T'_{34})](\langle\Delta\mu_{21}\rangle + \delta\Delta\mu_{21})(\langle\Delta\mu_{43}\rangle + \delta\Delta\mu_{43})\rangle \\
&\quad = \langle T'_{12}T'_{34}\rangle\langle\Delta\mu_{21}\rangle\langle\Delta\mu_{43}\rangle + \langle T'_{12}T'_{34}\rangle\langle\delta\Delta\mu_{21}\delta\Delta\mu_{43}\rangle + \langle\delta(T'_{12}T'_{34})\delta\Delta\mu_{43}\rangle\langle\Delta\mu_{21}\rangle \\
&\qquad + \langle\delta(T'_{12}T'_{34})\delta\Delta\mu_{21}\rangle\langle\Delta\mu_{43}\rangle + \langle\delta(T'_{12}T'_{34})\delta\Delta\mu_{21}\delta\Delta\mu_{43}\rangle.
\end{aligned} \tag{A3}$$

Now, the object of the present excercise is to show that the first term is dominant in the sum on the right-hand side of Eq. (A3). I have not done a detailed analysis of most of the terms written out here. I can, however, give a convincing argument that the second term is much smaller than the first. This I proceed with now.

Using the Schwarz inequality, we can set a bound on this term:

$$\langle T'_{12}T'_{34}\rangle\langle\delta\Delta\mu_{21}\delta\Delta\mu_{43}\rangle \geq \langle T'_{12}T'_{34}\rangle\sqrt{\langle\delta\Delta\mu_{21}^2\delta\Delta\mu_{43}^2\rangle}. \tag{A4}$$

Now, if the fluctuations of the local chemical potential are Gaussian (a reasonable assumption, according to conductance fluctuation theory), then the fourth-power average in Eq. (A4) is factorizable into second-power averages. A representative term after this separation will be

$$\langle T'_{12}T'_{34}\rangle\sqrt{\langle\delta\Delta\mu_{21}^2\rangle}\sqrt{\langle\delta\Delta\mu_{43}^2\rangle}. \tag{A5}$$

The typical mean-squared fluctuations of the chemical potential may be estimated using universal-conductance-fluctuation arguments. Suppose we have a three-dimensional (3D) conductor in a quasi-1D geometry, with the maximum cross-sectional area A consistent with there being phase coherence across the width of the conductor, namely $A \sim L_{\rm in}^2$. Following the results of Refs. 11 and 12, the fluctuation in the resistance from one point along the conductor to the other is given by

$$\Delta R \approx \frac{e^2}{h}R_\phi^2. \tag{A6}$$

Here R_ϕ is the average resistance of a length $L_{\rm in}$ of the conductor:

$$R_\phi \sim \frac{L_{\rm in}}{A}\frac{1}{\bar\sigma} \sim \frac{1}{L_{\rm in}\bar\sigma}. \tag{A7}$$

Here $\bar\sigma$ is the average bulk resistivity of the 3D conductor. Now, suppose there is a steady current I running through the conductor, which is related to the average potential gradient by

$$I = A\bar\sigma\langle\nabla\mu\rangle \sim L_{\rm in}^2\bar\sigma\langle\nabla\mu\rangle. \tag{A8}$$

Putting these together, we arrive at an estimate for the chemical-potential fluctuations:

$$\begin{aligned}\delta\Delta\mu &\sim \nabla\mu\frac{e_2}{h}\frac{1}{\bar\sigma},\\ &\sim \nabla\mu\frac{\lambda_F^2}{l_e}.\end{aligned} \tag{A9}$$

I have used the standard Boltzmann expression for the average conductivity. Finally, we compare Eq. (A9) with Eq. (7). Upon integration in Eq. (4), the factor $\mathbf{r}_2 - \mathbf{r}_1$ turns into a contribution of, order $L_{\rm in}$. Thus we see that the fluctuating-potential contribution in Eq. (A3) is smaller than the average-potential contribution of, e.g., Eq. (6) by a number of order $\lambda_F^2/l_eL_{\rm in}$, which is much less than unity in the cases that we are interested in. Thus we conclude that the first term is dominant over the second in Eq. (A3); since all the other terms also contain the fluctuating part of the chemical potential drop, we can argue heuristically that all the other terms are also small by the same factor(s).

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