

## Particle-hole asymmetry in fluctuating thermoelectric and Hall effects

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Thermoelectric and Hall currents arise due to particle-hole asymmetry (PHA) in the electron system. Quadratic electron spectrum alone is a source of PHA. This asymmetry, not associated with expansion of the density of states (DOS) near the Fermi surface, leads to singular fluctuation thermopower and Hall conductivity in electron systems with two-dimensional spectrum. In impure films new contributions dominate over the contributions due to DOS expansion by parameter  $(T_c\tau)^{-1}$ , where  $\tau$  is the electron-impurity scattering time. In agreement with experimental observations in high- $T_c$  cuprates, sign of the fluctuation Hall correction is opposite to the sign of the Hall conductivity in the normal state.

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In recent years fluctuations effects in superconductors near the critical temperature  $T_c$  attracted much attention in strongly anisotropic high- $T_c$  cuprates with two-dimensional electron transport along layers and weak coupling between layers. Near  $T_c$  the fluctuation conductivity of ordinary superconductors is described by the Aslamazov-Larkin (AL) correction.<sup>1</sup> The excess conductivity of high- $T_c$  cuprates is also well described by AL theory if anisotropy effects are included.<sup>2-4</sup> The current situation with the fluctuations thermopower (TEP) and Hall effect is rather confusing. The most intriguing problem is that the sign of the calculated fluctuation Hall conductivity<sup>5-7</sup> contradicts to experimental data on high- $T_c$  superconductors.<sup>8-11</sup>

All theories to date predict that AL correction to thermoelectric coefficient and Hall conductivity in two-dimensional conductors are absent.<sup>3,5-7</sup> Electrons and holes drifting in perpendicular magnetic and electric fields or in the temperature gradient give contributions to the electric current of opposite signs. Therefore, thermoelectric and Hall currents arise due to the difference between electron and hole states, i.e., due to the particle-hole asymmetry (PHA). To obtain nonzero fluctuations thermopower and Hall coefficient one should extract the PHA terms in the fluctuation propagator. Previous works took into account PHA in the electron density of states (DOS), which is constant in two-dimensional conductors. Therefore, one could conclude that the AL-corrections to the thermopower and Hall conductivity is absent. Interlayer coupling results in nonzero fluctuation TEP and Hall conductivity along layers due to modification of two-dimensional electron spectrum. However, experimental measurements in various cuprates exhibit no significant dependence on the interlayer coupling.<sup>8-11</sup>

We show that complete treatment of all sources of PHA can resolve the problems of sign and magnitude of fluctuations TEP and Hall effects. To elucidate main idea of our paper, we remind that for noninteracting electrons TEP requires expansion of electron parameters,  $v^2\nu\tau$  ( $v$  is the velocity,  $\nu$  is the density of states, and  $\tau$  is the momentum relaxation rate) near the Fermi surface to extract PHA. For the Hall conductivity no such expansion is necessary, and PHA due to quadratic electron spectrum alone results in nonvanishing effect.<sup>12</sup> Note, that for the linear (Dirac) spectrum

there is no PHA and the Hall effect is absent. For TEP and Hall conductivity of interacting electrons, both sources of PHA mentioned above should be taking into account on the same footing. However, PHA effects from the quadratic electron spectrum have been lost in all previous calculations of fluctuation TEP and Hall effect.

In the present paper we show that PHA from the electron spectrum results in singular fluctuation thermopower and Hall conductivity in 2D systems. The effects under consideration are also important for 3D conductors. In particular, we show that, in impure thin films the obtained TEP and Hall corrections are  $T_c\tau$  times larger than found in earlier papers.<sup>5-7,13,14</sup> In pure films our corrections are of the same order as corrections due to expansion of DOS.<sup>6,13</sup>

First, we calculate the PHA terms in the fluctuation propagator, which describes the interaction in the Cooper channel. In equilibrium the retarded propagator is

$$L_0^R(q, \omega) = [\lambda^{-1} - P^R(q, \omega)]^{-1}, \quad (1)$$

where  $\lambda$  is a constant of the electron-electron interaction and  $P(q, \omega)$  is the polarization operator

$$P^R(q, \omega) = i \int \frac{d\epsilon}{2\pi} S_0(\epsilon) \frac{\pi\nu\tau\zeta(q, \omega)}{1 - \zeta(q, \omega)}, \quad (2)$$

where  $S_0(\epsilon) = -\tanh(\epsilon/2T)$ . The function  $\zeta(q, \omega)$  is given by

$$\begin{aligned} \zeta(q, \omega) &= \frac{1}{\pi\nu\tau} \int \frac{d\mathbf{p}}{(2\pi)^2} G_0^A(\mathbf{p}, \epsilon) G_0^R(\mathbf{q} - \mathbf{p}, \omega - \epsilon) \\ &= -\frac{i}{\tau} \int \frac{d\phi}{2\pi} \frac{1}{2\epsilon - \omega + \xi_{q-p} - i/\tau}, \end{aligned} \quad (3)$$

where  $\phi$  is the angle between  $\mathbf{p}$  and  $\mathbf{q}$ , and the retarded ( $R$ ) and advanced ( $A$ ) electron Green functions are

$$G_0^R(\mathbf{p}, \epsilon) = [G_0^A(\mathbf{p}, \epsilon)]^* = (\epsilon - \xi_p + i/2\tau)^{-1}, \quad (4)$$

where  $\xi_p = (p^2 - p_F^2)/2m$ .

Equations (1)–(4) are well known and widely used in the microscopic theory of fluctuations.<sup>1,5-7,13,14</sup> An important

point of our calculations is that in Eq. (3) we will take into account PHA associated with electron spectrum  $\xi_p$ . Any deviation from the linear spectrum  $\xi_p = v_F(p - p_F)$  results in asymmetric terms. For the quadratic spectrum we have

$$\xi_{q-p_F} = -v_F q \cos \phi + q^2/(2m), \quad (5)$$

and PHA is taken into account by the small term  $q^2/2m$ . As seen, this term enters into Eq. (3) in the same way as  $\omega$ . Therefore, the expression for  $P(q, \omega)$  accounting for the PHA is given by

$$P^R(q, \omega) = -\frac{\nu}{2} \left[ \ln \frac{2\gamma\omega_D}{\pi T_c} - \alpha q^2 + \frac{i\pi}{8T_c} \omega + \frac{\omega}{2\epsilon_F} \frac{\partial \ln \nu}{\partial \ln \epsilon_F} \ln \frac{2\gamma\omega_D}{\pi T_c} - \frac{i\pi}{8T_c} \frac{q^2}{2m} \right], \quad (6)$$

where  $\nu$  is the electron density of states at the Fermi energy,  $\omega_D$  is the Debye frequency, and  $\gamma$  is the Euler constant.

Particle-hole symmetry is directly connected with properties of the retarded and advanced functions, it requires that  $P^R(-\mathbf{q}, -\omega) = P^A(\mathbf{q}, \omega)$ . Taking into account that  $[P^R(\mathbf{q}, \omega)]^* = P^A(\mathbf{q}, \omega)$ , the asymmetric term in  $P^R(\mathbf{q}, \omega)$  may be extracted as

$$P_{\text{PHA}}^R(\mathbf{q}, \omega) = \frac{1}{2} \{P^R(\mathbf{q}, \omega) - [P^R(-\mathbf{q}, -\omega)]^*\}. \quad (7)$$

Therefore, last two terms in Eq. (6) represent PHA. The first of them obtained from expansion of DOS has been taken into account in foregoing works.<sup>5-7,13,14</sup> Note that this term vanishes for 2D system. The last term is new, it has been overlooked in all previous publications. This term has the same form in all dimensions and plays a key role in 2D systems.

For an arbitrary electron momentum relaxation time the coefficient  $\alpha$  in Eq. (6) is<sup>1</sup>

$$\alpha = -\frac{v_F^2 \tau^2}{d} \left[ \psi \left( \frac{1}{2} + \frac{1}{4\pi T_c \tau} \right) - \psi \left( \frac{1}{2} \right) - \frac{1}{4\pi \tau T_c} \psi' \left( \frac{1}{2} \right) \right], \quad (8)$$

where  $d$  is the dimensionality of the electron system,  $\psi(x)$  is the logarithmic derivative of the gamma function. In the limiting cases

$$\alpha = \begin{cases} \pi v^2 \tau / (8dT), & T\tau \ll 1, \\ 7\zeta(3)v^2 / (16\pi^2 dT^2), & T\tau \gg 1. \end{cases} \quad (9)$$

Let us estimate relative contributions of the asymmetric terms. According to Eq. (6) the PHA term due to the DOS expansion is of the order of  $\omega/\epsilon_F$  and our new term is  $\sim q^2/(mT_c)$ . Taking into account that  $\alpha q^2 \sim \omega/T_c \sim (T - T_c)/T_c$ ,<sup>1</sup> we expect that for pure films ( $T_c \tau \gg 1$ ) the new contributions to the thermoelectric coefficient and Hall conductivity are of the same order as contributions from DOS expansion. In impure films ( $T_c \tau \ll 1$ ) the new contributions dominate over the old ones by parameter  $(T_c \tau)^{-1}$ . Calculations below support these estimations.

To investigate effect of superconducting fluctuations on the electron transport we use the quantum transport

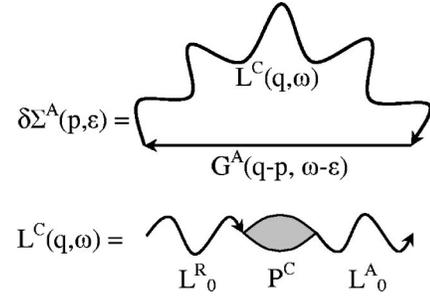


FIG. 1. Electron self-energy  $\delta\Sigma_{\text{AL}}$  corresponding to the Aslamazov-Larkin correction and the equilibrium fluctuation propagator  $L^C(\mathbf{q}, \omega)$ .

equation,<sup>13</sup> which is equivalent to the linear response method.<sup>1,5,7,14</sup> Below, we will present calculations taking into account only the new PHA term. Our purpose is to calculate the electric current initiated by the temperature gradient or external electric and magnetic fields

$$\mathbf{J}_e = 2e \int \frac{d\mathbf{p}d\epsilon}{(2\pi)^{d+1}} \mathbf{v} S(\mathbf{p}, \epsilon) \text{Im} G^A(\mathbf{p}, \epsilon). \quad (10)$$

According to Eq. (10), the electric current may be associated with the nonequilibrium distribution function  $S(\mathbf{p}, \epsilon)$ , as well as with various nonequilibrium corrections to the electron density of states  $\text{Im} G^A(\mathbf{p}, \epsilon)$ .

In the transport equation method the AL-term corresponds to the correction to  $\text{Im} G^A$ ,<sup>13</sup>

$$\delta G^A = (G_0^A)^2 \delta\Sigma_{\text{AL}}^A, \quad (11)$$

where the electron self-energy  $\delta\Sigma_{\text{AL}}^A$  shown in Fig. 1 is given by

$$\delta\Sigma_{\text{AL}}^A = \frac{i}{2} \int \frac{d\mathbf{q}d\omega}{(2\pi)^3} \frac{G_0^R(\mathbf{q}-\mathbf{p}, \omega-\epsilon)}{[1-\zeta(q, \omega)]^2} \delta L^C(\mathbf{q}, \omega), \quad (12)$$

where  $\delta L^C(\mathbf{q}, \omega)$  is the kinetic (Keldysh) component of the nonequilibrium fluctuation propagator, which will be discussed latter.

Substituting Eq. (12) into Eq. (11), and then into Eq. (10), we get

$$\mathbf{J}_e = e \int \frac{d\mathbf{p}d\mathbf{q}d\epsilon d\omega}{(2\pi)^{d+4}} \mathbf{v} S_0(\epsilon) \frac{\delta L^C(\mathbf{q}, \omega)}{[1-\zeta(q, \omega)]^2} \times [G_0^A(\mathbf{p}, \epsilon)]^2 G_0^R(\mathbf{q}-\mathbf{p}, \omega-\epsilon). \quad (13)$$

Integrating over  $\mathbf{p}$  and  $\epsilon$ , we find

$$\mathbf{J}_e = e\nu\alpha \int \frac{d\mathbf{q}d\omega}{(2\pi)^3} \mathbf{q} \text{Im} \delta L^C(\mathbf{q}, \omega). \quad (14)$$

Now we calculate the nonequilibrium fluctuation propagator, when the electron system is disturbed by the thermal gradient. The equilibrium propagator may be presented as (see Fig. 2)

$$L_0^C(q, \omega) = L_0^R(q, \omega) P^C(q, \omega) L_0^A(q, \omega), \quad (15)$$

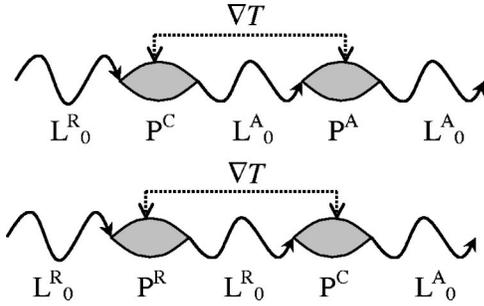


FIG. 2. Nonequilibrium fluctuation propagator  $\delta L_T^C$  with the Poisson bracket due to the temperature gradient.

where

$$P^C(q, \omega) = \coth\left(\frac{\omega}{2T}\right) [P^R(q, \omega) - P^A(q, \omega)]. \quad (16)$$

When the system is driven out of equilibrium by the temperature gradient, the corresponding Poisson bracket<sup>13</sup>

$$\{A, B\}_T = \nabla T \left( \frac{\partial A}{\partial T} \frac{\partial B}{\partial \mathbf{q}} - \frac{\partial B}{\partial T} \frac{\partial A}{\partial \mathbf{q}} \right), \quad (17)$$

applied to Eq. (15), according to Fig. 2, results in the nonequilibrium propagator

$$\begin{aligned} \delta L_T^C &= i[L_0^R(L_0^A)^2 - (L_0^R)^2 L_0^A] (P^R - P^A) \\ &\times \frac{\partial}{\partial T} \left( \coth\frac{\omega}{2T} \right) \nabla T \frac{\partial}{\partial \mathbf{q}} [\text{Re}P^R(q, \omega)]. \end{aligned} \quad (18)$$

Expanding  $\delta L_T^C$  to the first order in  $q^2/2m$  and integrating it over  $\omega$ , we get

$$\begin{aligned} \int \frac{d\omega}{2\pi} \delta L_T^C(q, \omega) &= i\nu\alpha\mathbf{q}\nabla T \frac{q^2}{2m} \int \frac{d\omega}{2\pi} \frac{2}{\omega} \frac{\partial}{\partial \omega} (L_0^R L_0^A) \\ &\times \frac{\partial}{\partial T} \left( \coth\frac{\omega}{2T} \right) \\ &= \frac{i\pi\alpha}{\nu T} \frac{\mathbf{q}\nabla T}{(\Delta T/T + \alpha q^2)^3} \frac{q^2}{2m}, \end{aligned} \quad (19)$$

where  $\Delta T = T - T_c$ . Finally, integrating Eq. (19) over  $\mathbf{q}$ , we find the electric current  $\mathbf{J}_e$  [Eq. (14)]. Keeping in mind that  $\mathbf{J}_e = \eta \nabla T$ , we get the thermoelectric coefficient in 2D system,

$$\delta\eta = \frac{e}{16m\alpha T} \ln\left(\frac{T_c}{T - T_c}\right). \quad (20)$$

Next we consider the fluctuation correction to the Hall coefficient. The corresponding nonequilibrium propagator  $\delta L_{E \times H}^C$  in crossed electric and magnetic fields is calculated with electric and magnetic Poisson brackets

$$\{A, B\}_E = e\mathbf{E} \left( \frac{\partial A}{\partial \omega} \frac{\partial B}{\partial \mathbf{q}} - \frac{\partial B}{\partial \omega} \frac{\partial A}{\partial \mathbf{q}} \right), \quad (21)$$

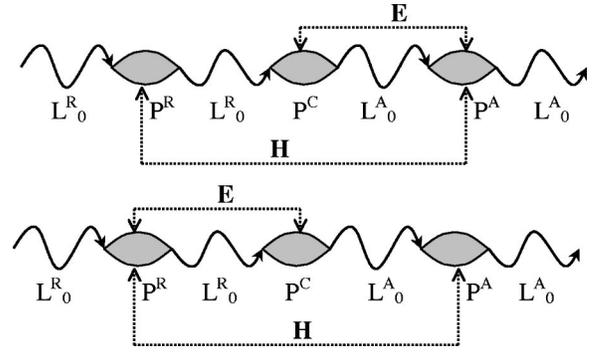


FIG. 3. Nonequilibrium fluctuation propagator  $\delta L_{E \times H}^C$  with the Poisson brackets due to electric and magnetic fields.

$$\{A, B\}_H = \frac{e}{c} \mathbf{H} \left( \frac{\partial A}{\partial \mathbf{q}} \times \frac{\partial B}{\partial \mathbf{q}} \right), \quad (22)$$

according to Fig. 3. The electric bracket [Eq. (21)] applied to the first diagram gives

$$\delta L_E^C = i(P^R - P^A)L_0^R(L_0^A)^2 \frac{\partial}{\partial \omega} \left( \coth\frac{\omega}{2T} \right) \frac{\partial P^A}{\partial \mathbf{q}} \mathbf{E}. \quad (23)$$

Then the magnetic bracket [Eq. (22)] gives

$$\begin{aligned} \delta L_{E \times H}^C &= (i)^2 (P^R - P^A) (L_0^R L_0^A)^2 \frac{\partial}{\partial \omega} \left( \coth\frac{\omega}{2T} \right) \\ &\times \frac{\partial^2 P^A}{\partial q^2} \frac{e}{c} \mathbf{H} \left( \frac{\partial P^R}{\partial \mathbf{q}} \times \mathbf{E} \right). \end{aligned} \quad (24)$$

The second diagram in Fig. 3 is calculated in the same way. Total contribution of both diagrams is

$$\begin{aligned} \delta L_{E \times H}^C &= -2(\nu\alpha)^2 (P^R - P^A) (L_0^R L_0^A)^2 \frac{\partial}{\partial \omega} \left( \coth\frac{\omega}{2T} \right) \\ &\times \frac{e}{c} \mathbf{q} \cdot (\mathbf{E} \times \mathbf{H}). \end{aligned} \quad (25)$$

Next we perform expansion over  $q^2/2m$  in Eq. (25),

$$(P^R - P^A)(L_0^R L_0^A)^2 = -\frac{q^2}{2m} \frac{\partial}{\partial \omega} (P^R - P^A)(L_0^R L_0^A)^2. \quad (26)$$

Substituting  $\delta L_{E \times H}^C$  into Eq. (14) and integrating it over  $\omega$ , we get

$$\int \frac{d\omega}{2\pi} \delta L_{E \times H}^C = -\frac{\pi^2 e}{8} \frac{\alpha^2}{c \nu T} \frac{\mathbf{q} \cdot (\mathbf{E} \times \mathbf{H})}{(\Delta T/T + \alpha q^2)^5} \frac{q^2}{2m}. \quad (27)$$

Finally, integrating Eq. (27) over  $\mathbf{q}$ , we find the electric current proportional to  $\mathbf{E} \times \mathbf{H}$ . Defined from  $\mathbf{J}_e = \sigma_H(\mathbf{E} \times \mathbf{H})/H$ , the Hall conductivity in a 2D system is

$$\delta\sigma_{xy} = -\frac{\pi}{384} \frac{e^2 \omega_c}{T} \left( \frac{T_c}{T - T_c} \right)^2, \quad (28)$$

where  $\omega_c$  is the cyclotron frequency. Eqs. (20) and (28) are presented for 2D conductors. For a quasi-two-dimensional system (thin films with thickness less than the coherence length) these expressions should be divided by the film thickness. Note that  $\delta\sigma_{xy}$  has a sign opposite to the sign of  $\sigma_{xy}$  ( $\sigma_{xy} = \omega_c \tau \sigma_{xx}$ ).

Now we discuss our results and compare them with contribution obtained from expansion of DOS. Taking into account PHA due to quadratic electron spectrum, we find the fluctuation thermoelectric coefficient [Eq. (20)] and Hall conductivity [Eq. (28)]. The obtained results predict the singular behavior of the transport coefficients near  $T_c$  in 2D electron system. Our results have identical form for 2D systems and thin films. Contributions to the fluctuation thermoelectric coefficient and Hall conductivity from DOS expansion vanish in 2D systems. For thin films the corresponding correction to  $\eta$  is<sup>13</sup>

$$\delta\eta^{\text{DOS}} = 0.1 \frac{eT}{\epsilon_F a} \frac{\partial \ln \nu}{\partial \ln \epsilon_F} \ln \frac{2\gamma\omega_D}{\pi T_c} \ln \left( \frac{T_c}{T - T_c} \right). \quad (29)$$

Taking into account PHA due to expansion of DOS [asymmetric  $\omega$ -dependent term in Eq. (6)] and repeating our calculations, we obtain the corresponding correction to the Hall conductivity

$$\delta\sigma_{xy}^{\text{DOS}} = - \frac{e^2 \alpha m \omega_c T}{2\pi \epsilon_F a} \frac{\partial \ln \nu}{\partial \ln \epsilon_F} \ln \frac{2\gamma\omega_D}{\pi T_c} \left( \frac{T_c}{T - T_c} \right)^2. \quad (30)$$

Note, that authors of previous works<sup>5-7</sup> reported  $\sigma_{xy}^{\text{DOS}}$ , which is of the same order as our result [Eq. (30)], but with

a wrong sign. The result of Ref. 5 comes from erroneous sign of the  $\omega$ -dependent PHA term in the fluctuation propagator [see Eq. (6)]. This mistake has already been corrected for fluctuation TEP in our previous paper.<sup>13</sup> The authors of Refs. 6,7 also mentioned the wrong sign of the  $\omega$ -dependent PHA term in Ref. 5, but arrived at the same sign of the Hall conductivity as in Ref. 5.

Comparing Eqs. (20), (28), (29), and (30), we see that for pure films ( $T_c \tau \gg 1$ ) the new contributions and the contribution from expansion of DOS are of the same order. In impure films ( $T_c \tau \ll 1$ ) new contributions to the thermoelectric coefficient and Hall conductivity dominates over the old ones by parameter  $(T_c \tau)^{-1}$ .

According to Eqs. (28) and (30), both fluctuation corrections to the Hall conductivity are opposite by sign to the Hall conductivity of noninteracting electrons ( $\sigma_{xy}$ ). We would like to emphasize that, according to Refs. 1,13 and our current paper, AL corrections provide monotonic changes of all transport coefficients (conductivity, thermoelectric coefficient, and Hall conductivity) from normal to superconducting states.

While in the current paper we have considered *s*-wave pairing, it may be shown that accounting for *d*-wave pairing does not change our results for pure superconductors ( $T_c \tau \gg 1$ ),<sup>15</sup> and our conclusions are applicable to high- $T_c$  cuprates. Thus, the monotonic change of the Hall conductivity above  $T_c$  is in agreement with experimental observations in cuprates.<sup>8-11</sup> This resolves a long-standing problem of the sign of fluctuation Hall conductivity.<sup>6,7</sup>

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