A giant thermal Hall effect in the vicinity of the superconducting phase transition

A. V. Kavokin,1,2,3 Y. M. Galperin,4,5 and A. A. Varlamov6

1Westlake University, 18 Shilongshan Road, Hangzhou 310024, Zhejiang Province, China
2Institute of Natural Sciences, Westlake Institute for Advanced Study, 18 Shilongshan Road, Hangzhou 310024, Zhejiang Province, China
3Spin Optics Laboratory, St. Petersburg State University, Ulanyoenskaya 1, 198504 St. Petersburg, Russia
4A. F. Ioffe Physico-Technical Institute of Russian Academy of Sciences, Polytekhnicheskaya 26, 194021 St. Petersburg, Russia
5Department of Physics, University of Oslo, 0316 Oslo, Norway
6CNR-SPIN, DICHI-University of Rome Tor Vergata, Via del Politecnico 1, 00133 Rome, Italy

We extend the thermodynamic approach for the description of the thermal Hall effect in the vicinity of a superconducting phase transition, in the fluctuation dominated regime. We show that the Hall heat conductivity is proportional to the product of temperature derivatives of the chemical potential and of the magnetization of the system. We argue that the latter derivative shows the strong singularity in the vicinity of the phase transition, while the former does not contain the characteristic for fermionic systems smallness (T/E_F), what additionally increases the effect. We derive the analytical formula predicting the temperature dependence of the thermal Hall conductivity in the vicinity of the critical temperature for different magnetic fields. Moreover, we study the phenomenon in the regime of quantum fluctuations, in the vicinity of the second critical field and at very low temperatures. We demonstrate how it fades away in a full agreement with the third law of thermodynamics. The developed approach qualitatively explains the recently observed giant thermal Hall effect in cuprates [1].

The thermal Hall effect consists in a generation of a heat flow by a combined action of the temperature gradient ∇T and magnetic field H perpendicular to it [2, 3]. The heat current is generated in the direction that is perpendicular both to the magnetic field and the temperature gradient applied. This phenomenon is cognate to the Leduc-Righi effect [4], well known in metals and semiconductors, where the temperature gradient induced in the direction [H × ∇T] is measured as a function of ∇T. In agreement with the Wiedemann-Franz law, the thermal Hall effect in metals is usually very weak. This is clearly understandable as heat flows carried by phonons are weakly sensitive to magnetic fields [5]. However, recently, in a number of publications, a giant increase of the thermal Hall conductivity κyx violating the Wiedemann-Franz law has been reported in several pseudogap cuprates such as La_{1.6−x}Nd_{0.4}Sr_xCuO_4, La_{1.8−x}Eu_{0.2}Sr_xCuO_4, La_{2−x}Sr_xCuO_4, and Bi_2Sr_{2−x}La_xCuO_{6+δ} [6].

The increase of the absolute value of κyx by about two orders of magnitude and its negative sign in the vicinity of the phase transition temperature in these systems seems puzzling, at first glance. These discoveries have triggered the interest to comparatively large values of κyx found also in antiferroics [7] and the nearly ferroelectric insulator SrTiO_3 [8]. First experimental works were followed by a number of publications aimed at the further study and interpretation of the observed effects [9, 10]. A multitude of possible reasons of the effect has been proposed for each studied system, while no unified approach to the interpretation of a giant increase of thermal Hall conductivity in the vicinity of phase transition points is available till now, to the best of our knowledge.

Here we attempt at formulating a simple model that reveals the mechanism behind the observed effects and may be adapted to each particular experimental system. We develop a general thermodynamic approach that links κyx to the equilibrium characteristics of the systems under study. We consider an open circuit geometry where there is no electric current in the system (see Fig. 1). We shall assume that the system is in the stationary state that may be characterised by a constant electro-chemical potential. This assumption will allow us to express κyx through the temperature derivatives of the chemical potential and magnetization. Analyzing the recent experimental data on pseudo-gap cuprates we conclude that the giant Hall thermoconductivity found in these systems might take place because the temperature derivative of the magnetization shows a strong singularity in the vicinity of the phase transition, while that one of the chemical potential does not contain the smallness characteristic of the degenerate Fermi gas (T/E_F). Together, these two factors might be responsible for the giant magnitude of the effect. Here, in the framework of the Ginzburg-Landau approach, we derive the analytical formula for κyx that qualitatively describes the giant increase of (negative) thermal Hall conductivity reported in cuprates [1]. Furthermore, we study the thermal Hall effect in the regime of quantum fluctuations: in the vicinity of the second critical magnetic field and in the limit of very low temperatures. We show that the effect vanishes in zero temperature limit in a full agreement with the third law of thermodynamics.

Basic definitions and the thermodynamic approach. To start with, let us recall that the electric and heat currents can be linked to the external electric field E and
temperature gradient $\nabla T$ with use of the conductivity $\sigma(H)$, thermolectric $\beta(H)$, and heat conductivity $\kappa(H)$ tensors as follows:

$$\left( \frac{\mathbf{j}}{q} \right) = \left( \frac{\sigma}{\gamma} \right) \mathbf{E} - \left( \frac{\beta}{\kappa} \right) \nabla T,$$

(1)

with the Onsager relation $\dot{\gamma}(H) = -T \dot{\beta}(-H)$.

The thermal Hall relation consists in the build up of the off-diagonal elements of $\kappa$ in the presence of a magnetic field, as the scheme in Figure 1 shows.

In the stationary regime, where the external circuit is broken, no electric current is flowing through the system and the electrochemical potential of the charge carriers

$$\tilde{\mu} = \mu + e^* \phi$$

(2)

($\mu$ is the chemical potential, $\phi$ is the electrostatic potential, $e^*$ is the carrier’s charge) remains constant. This statement is valid also if a temperature gradient is present in the sample. In this case, the chemical potential $\mu$ becomes dependent on the coordinate and, consequently, the internal electric field, $\mathbf{E}$, is generated:

$$E_x = -\nabla x = -\frac{1}{e^*} \left( \frac{d\mu}{dT} \right) \nabla_x T.$$  

(3)

Under these conditions the diagonal components of the thermolectric tensor $\beta$ can be related to the temperature derivative of the chemical potential by the Kelvin formula [12], while the off-diagonal components of this tensor (arising if magnetic field is applied) are governed by the appearance of uncompensated magnetization currents. They can be expressed in terms of the temperature derivative of the magnetization (see, e. g., [13, 14] and references therein):

$$\dot{\beta} = \left( \begin{array}{c} -\frac{\sigma_{xx}}{c^*} \frac{d\mu}{dT} - \frac{c_{xx}}{c^*} \frac{dM_z}{dT} \\ -\frac{c_{xx}}{c^*} \frac{dM_z}{dT} - \frac{\sigma_{xx}}{c^*} \frac{d\mu}{dT} \end{array} \right).$$  

(4)

Using these relations one can express the Hall thermal flow as

$$q_y = -\kappa_{yx} \nabla_x T = \gamma_{yx} E_x.$$  

(5)

We note that the second equality in Eq. (5) is by no means universal. It is valid only in the open circuit geometry ($\mathbf{j} = 0$, $\mathbf{q} = 0$) in the stationary regime, where the effect of a temperature gradient can be fully accounted for by the introduction of an induced electric field (3). Using this substitution, one can write down the relation linking the thermal Hall conductivity to the temperature derivatives of the chemical potential and magnetization:

$$\kappa_{yx} \equiv \frac{e^*}{c^*} \left( \frac{dM_z}{dT} \right) \left( \frac{d\mu}{dT} \right).$$  

(6)

One can see that the thermal Hall effect is governed by the product of the chemical potential and magnetization derivatives over temperature. This simple relation sheds light on the physics that is behind the recently observed giant thermal Hall effect in cuprates. We also note that, experimentally, the temperature gradient in $y$-direction is frequently measured rather than the thermal flow. This quantity, also known as the Righi-Leduc coefficient [5], is dependent on both diagonal and non-diagonal components of the tensor $\kappa$ and cannot be directly described by the proposed here expression for the thermodynamic contribution to $\kappa_{yx}$. However, we believe that the thermodynamic formula [6] grasps the essential physics that is behind the observed effect.

Now, using the obtained above general relation Eq. (6), we will focus on the role of fluctuating Cooper pairs in the thermal Hall effect above the superconducting phase transition. Even before doing any calculations one can expect that the effect will be huge here since the fluctuation diamagnetism, being precursor of the Meissner effect, is giant [15]. An additional reinforcing factor is the large value of the temperature derivative of the chemical potential of fluctuating Cooper pairs. We start from the detailed study of the domain of the phase diagram close to the critical temperature using the Ginzburg-Landau formalism. We shall estimate the magnitude and the temperature dependence of the thermal Hall effect in the domain of quantum fluctuations: above $H_{c2}(0)$ and at very low temperatures.

The free energy, magnetization, and chemical potential of fluctuating Cooper pairs. We shall use the expression for the Ginzburg-Landau (GL) free energy for a fluctuating superconductor in the 2D case that one can find in Ref. [16]:

$$F_{(2)}^{(H)}(\epsilon, h) = -\frac{1}{4\pi^2} \ln \left[ \ln \left( \frac{1}{T - T_{c0}} \right) \right].$$

(7)

Here $S$ is the sample cross-section and $\xi = \pi D/2\xi_{c0}$ is the superconducting coherence length, $D$ is the electron diffusion coefficient, $\epsilon = (T - T_{c0})/T_{c0}$ is the reduced temperature, $T_{c0}$ is critical temperature of the superconducting phase transition at zero magnetic field. The dimensionless magnetic field $h = H/H_{c2}(0)$ is normalized with the second critical field $H_{c2}(0) = \Phi_0/(2\pi\xi^2)$,
introduced as the linear extrapolation to zero tempera-
ture of the GL formula and \( \Phi_0 = \pi c/e \) as the magnetic flux quantum. Note that superconducting fluctuations behave as 2D objects since the characteristic size of the fluctuating Cooper pairs, \( \xi(\epsilon) = \xi/\sqrt{\epsilon} \), exceeds the thickness \( d \) of the film.

The expression for 2D fluctuation magnetization per unit square of the film can be obtained just differentiating the expression for the free energy over magnetic field and taking this derivative with the opposite sign \[16\]:

\[
M_{(2)}^{(a)}(\epsilon, h) = \frac{T_{c0}}{\Phi_0} \left\{ \ln \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) - 1 \right\},
\]

where \( \psi(z) = d \ln \Gamma(x)/dx \) is the logarithmic derivative of the Euler gamma function. This formula describes the crossover from the weak field linear regime to the saturation of the fluctuation magnetization in strong fields \[17\]. The temperature derivative of the fluctuation magnetization is given by

\[
\frac{dM_{(2)}^{(a)}(\epsilon, h)}{dT} = -\frac{1}{2h\Phi_0} \left[ \frac{\epsilon}{2h} \psi' \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) - 1 \right] \quad (9)
\]

where \( \epsilon_h \equiv \epsilon + h \).

The last ingredient which we need in order to be able to calculate Eq. \[6\] explicitly is the chemical potential of fluctuating Cooper pairs. We are interested in the expression that would be valid for an arbitrary relation between the temperature and the magnetic field in the vicinity of \( T_{c0} \). It can be found from the definition of the chemical potential in terms of the derivative of the free energy, see Eq. \[7\] over the fluctuation Cooper pairs concentration \( N_{(2)}^{(a)} \). The latter can be easily obtained by means of integration of the distribution function of the Cooper pairs over momenta. In the presence of an external magnetic field, for a 2D superconductor the integration is replaced by the summation over Landau levels. Performing the calculus analogous to the derivation of Eq. \[7\], see Ref. \[15\], one finds

\[
N_{(2)}^{(a)}(\epsilon, H) = \frac{1}{4\pi c^2} \left[ \ln \left( \frac{1}{2h} - \psi \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) \right) \right].
\]

Finally, using Eqs. \[7\] and \[10\] one finds

\[
\mu_{(2)}^{(a)}(\epsilon, h) = \left( \frac{\partial F_{(2)}^{(a)}}{\partial N_{(2)}^{(a)}} \right) = -T_{c0} \epsilon \frac{1}{2} \ln \frac{1}{2h} - \psi \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) \quad (11)
\]

At zero field, it acquires a simple form \( \mu_{(2)}^{(a)}(0) = T_{c0} \epsilon \). Now, in particular, the temperature derivative of the fluctuation Cooper pair chemical potential can be obtained as:

\[
\frac{d\mu_{(2)}^{(a)}(\epsilon, h)}{dT} = -1 + \frac{\epsilon}{2h} \psi' \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) \ln \frac{1}{2h} - 2h \ln \left( \frac{1}{2h} - \psi' \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) \right) \quad (12)
\]

Thermal Hall conductivity due to fluctuating Cooper pairs. Now, the thermal Hall conductivity can be represented explicitly:

\[
\kappa_{yx}^{(a)}(\epsilon, h) = -\frac{T_{c0}}{4\pi h} \left[ 1 - \frac{\epsilon}{2h} \psi' \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) \right] \ln \frac{1}{2h} - 2h \ln \left( \frac{1}{2h} - \psi' \left( \frac{1}{2} + \frac{\epsilon}{2h} \right) \right) \quad (13)
\]

It is instructive to express Eq. \[13\] in its asymptotic form:

\[
\kappa_{yx}^{(a)}(\epsilon, h) = -\frac{eD \kappa}{64c} \left\{ \begin{array}{l} 1/3\epsilon^2, \quad \epsilon \ll 1, \\ 1/2\epsilon^2, \quad \epsilon \ll 1, \\ 1/2\epsilon^2, \quad \epsilon \ll h, \end{array} \right. \quad (14)
\]

Here we have introduced the true magnetic field, \( H = hH_{c2}(0) \). It is also used the relation between the GL extrapolation of the second critical field with the BCS one: \( H_{c2}(0) = (8\gamma_E/\pi^2)H_{c2}^{BCS}(0) \). We note that in the BCS theory \( H_{c2}(0) = (2/\gamma_E)\Phi_0 (T_{c0}/D) \), where \( \gamma_E = 1.78 \) is the Euler constant.

Effect of quantum fluctuations on thermal Hall conductivity above \( H_{c2}(0) \). Using the general thermodynamic relation \[6\] one can study the behavior of thermal Hall conductivity above \( H_{c2}(0) \) in the limit of very low temperatures. We shall consider the domain of quantum fluctuations, where one approaches the superconducting state by reducing the magnetic field toward \( H_{c2}(0) \) at sufficiently low temperatures. The behaviour of the fluctuation magnetization in this regime was found by Galitski and Larkin in Ref. \[20\]:

\[
M_{(2)}^{(a)} \left( t, \tilde{h} \right) = \frac{T_{c0}}{\gamma_E \Phi_0} \left[ \ln \frac{1}{2\gamma_E t} - \psi \left( \frac{1}{2\gamma_E t} \right) \right] \quad (15)
\]

with \( t = T/T_{c0} \ll 1 \) and \( \tilde{h} = (H - H_{c2}(T))/H_{c2}(T) \ll 1 \). The differentiation of Eq. \[15\] results in

\[
\frac{dM_{(2)}^{(a)}(t, \tilde{h})}{dT} = \frac{1}{\gamma_E \Phi_0} \left[ \frac{\tilde{h}}{2\gamma_E t} \psi' \left( \frac{1}{2\gamma_E t} \right) \right] \quad (16)
\]
Results and discussion. Figure 2 shows the thermal Hall conductivity as a function of the reduced temperature and magnetic field. One can see that $\kappa_{yx}(\epsilon, h)$ has a negative sign, and its absolute value increases rapidly as one approaches the phase transition boundary.

Now one can compare the predictions of our analytical theory with the experimental results reported in Ref. [1] for four cuprate superconductors. One can notice that the theory correctly reproduces both the sign of the thermal conductivity and the dramatic increase of its magnitude with the temperature decrease. We believe that the qualitative agreement of such a simple model with the experimental results is significant as it hints at the essentially thermodynamic nature of the giant thermal Hall effect.

We note that the approach we used is based on the conventional theory of fluctuations [15] applicable to superconductors above the phase transition boundary $H_{c2}(T)$. It may not account for all the specifics of the experimentally studies cuprate superconductors. Yet, it turns out that the main ingredients required for application of Eq. (6), i.e., temperature dependencies of the fluctuation magnetization and chemical potential of the preformed Cooper pairs in the pseudogap state, qualitatively do not differ much from that ones of a conventional superconductor. This is confirmed in the recent study Ref. [22], that went beyond the weak-fluctuation formalism, applied the precursor-pairing approach within the BCS-BEC crossover scheme [23, 24] and found a large singular diamagnetic response for the temperatures much higher than the transition temperature side by side with the strong temperature dependence of the pair chemical potential in a striking similarity to the effects predicted by the simple model developed here.

Note, that the thermodynamic approach to the thermal Hall effect can be universally applied to a large variety of systems undergoing a topological phase transition of the Lifshitz type, while the specific expressions for a magnetization and chemical potential we used in the calculation are applicable for 2D superconductors in the fluctuation dominated regime.

Finally, we would like to point out that the effect described here is closely related to the Leduc-Righi effect [5]. Indeed, at weak magnetic fields and in the absence of an electric current ($j_x = j_y = 0$) the heat flow can be written in the form

$$\mathbf{q} = -\tilde{k}\nabla T + L[\mathbf{H} \times \nabla T].$$  \hspace{1cm} (19)

If the additional condition $q_y = 0$ is implied, the temperature gradient applied along the $x$–axis induces the temperature gradient along the $y$–axis:

$$\nabla_y T = \frac{L_H}{\kappa_{yy}} \nabla_x T.$$  \hspace{1cm} (20)

One can see that the thermal Hall conductivity vanishes at zero temperature, in a full agreement with the third law of thermodynamics.

In the vicinity of $H_{c2}(0)$, the chemical potential of fluctuation Cooper pairs can be written as $\mu^{(QF)} = -\Delta_{BCS} \tilde{h}$ (similarly to the expression valid at $T_c$ see Ref. [19]). Its temperature derivative differs from zero due to the temperature dependence of $H_{c2}(T)$ (see Ref. [21]):

$$\frac{d\mu^{(QF)}}{dT} = \frac{\Delta_{BCS}}{H_{c2}(0)} \left( \frac{dH_{c2}(T)}{dT} \right) = -\frac{2\gamma_E}{\pi} \tilde{t}. \hspace{1cm} (17)$$

Substituting Eqs. (16) and (17) into Eq. (6) and taking into account that $T_{c0} = (\pi/\gamma_E)\Delta_{BCS}$ one finally finds

$$\kappa_{yx}(\epsilon, \tilde{h}) = \frac{\Delta_{BCS}}{\pi} \left[ t + \frac{\gamma_E t^2}{\tilde{h}} - \frac{\tilde{h}}{2\gamma_E} \psi' \left( \frac{1}{2\gamma_E t} \right) \right], \hspace{1cm} (18)$$

$$= -\frac{\Delta_{BCS}}{\pi} \tilde{t}^2 \left\{ 2\gamma_E t/3\tilde{h}^2, \hspace{0.5cm} t \ll \tilde{h} \ll 1, \hspace{1cm} \frac{1}{\tilde{h}}, \hspace{0.5cm} \tilde{h} \ll t \ll 1. \right.$$ 

One can see that the thermal Hall conductivity vanishes at zero temperature, in a full agreement with the third law of thermodynamics.
pressed as:

\[
\frac{L}{\kappa_{yy}} = -\frac{1}{\kappa_{yy}} c^* H \left( \frac{dM}{dT} \right) \left( \frac{d\mu}{dT} \right).
\]

(21)

Let us stress here, that the diagonal component of the thermal conductivity tensor \(\kappa_{yy}\) depends on the kinetic properties of the system.

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