

Proposed Model of the Giant Thermal Hall Effect in Two-Dimensional Superconductors: An Extension to the Superconducting Fluctuation Regime

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We extend the thermodynamic approach for the description of the thermal Hall effect in two-dimensional superconductors above the critical temperature, where fluctuation Cooper pairs contribute to the conductivity, as well as in disordered normal metals where the particle-particle channel is important. We express the Hall heat conductivity in terms of the product of temperature derivatives of the chemical potential and of the magnetization of the system. Based on this general expression, we derive the analytical formalism that qualitatively reproduces the superlinear increase of the thermal Hall conductivity with the decrease of temperature observed in a large variety of experimentally studied systems [Grissonanche *et al.*, *Nature (London)* **571**, 376 (2019)]. We also predict a nonmonotonic behavior of the thermal Hall conductivity in the regime of quantum fluctuations, in the vicinity of the second critical field and at very low temperatures.

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The thermal Hall effect consists of a generation of a heat flow by a combined action of the temperature gradient ∇T and magnetic field \mathbf{H} perpendicular to it [1–3]. 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 $[\mathbf{H} \times \nabla T]$ 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 κ_{yx} violating the Wiedemann-Franz law has been reported in several pseudogap cuprates such as $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$, $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, and $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$ [6].

The increase of the absolute value of κ_{yx} by about 2 orders of magnitude and its negative sign in these systems seems puzzling, at first glance. Note, that the studied materials [6] included both up-critical superconductors, where the conductivity is strongly influenced by fluctuation Cooper pairs, and normal metals where no Cooper pairs could be formed. While the mechanism of electric conductivity in the studied materials varied strongly, their

thermal Hall conductivity demonstrated the same features, namely, the negative sign and the superlinear increase with the decrease of temperature. These discoveries have triggered the interest to comparatively large values of κ_{yx} 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 [9–11]. A multitude of possible reasons of the effect have been proposed for each studied system, while no unified approach to the interpretation of a giant increase of thermal Hall conductivity in up-critical superconductors and disordered metals is available till now, to the best of our knowledge.

Here, we attempt to formulate 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 κ_{yx} 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 characterized by a constant electrochemical potential. This assumption will allow us to express κ_{yx} through the temperature derivatives of the chemical potential and magnetization. Analyzing the recent experimental data on pseudogap cuprates we conclude that the giant Hall

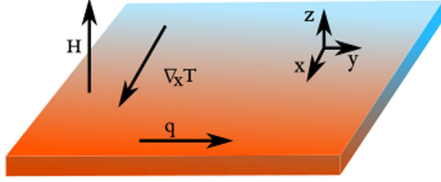


FIG. 1. The schematic showing the geometry of a thermal Hall effect measurement. The Hall bar is studied in the broken circuit geometry. The thermal flow in the y direction is measured as a function of the temperature gradient applied in the x direction and the magnetic field parallel to the z axis.

thermoconductivity found in these systems might take place because the temperature derivative of the magnetization shows a strong increase with the decrease of temperature, especially in the vicinity of the phase transition, while the temperature derivative of the chemical potential does not contain the smallness characteristic of the noninteracting degenerate Fermi gas (T/E_F). Together, these two factors might be responsible for the giant magnitude of the effect. In this Letter, in the framework of the fluctuation theory approach, we derive the analytical expressions for κ_{yx} both for a superconductor in various regimes and for a normal metal. These expressions qualitatively describe the giant increase of (negative) thermal Hall conductivity reported in cuprates [6]. 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 predict that the effect vanishes in zero temperature limit in 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 \mathbf{E} and temperature gradient ∇T with use of the conductivity $\hat{\sigma}(H)$, thermoelectric $\hat{\beta}(H)$, and heat conductivity $\hat{\kappa}(H)$ tensors as follows:

$$\begin{pmatrix} \mathbf{j} \\ \mathbf{q} \end{pmatrix} = \begin{pmatrix} \hat{\sigma} \\ \hat{\gamma} \end{pmatrix} \mathbf{E} - \begin{pmatrix} \hat{\beta} \\ \hat{\kappa} \end{pmatrix} \nabla T, \quad (1)$$

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

The thermal Hall effect consists of the buildup of the off-diagonal elements of $\hat{\kappa}$ in the presence of a magnetic field, as the scheme in Fig. 1 shows.

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

$$\bar{\mu} = \mu + e^* \phi \quad (2)$$

(μ is the chemical potential, ϕ 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 μ becomes dependent on the coordinate and, consequently, the internal electric field \mathcal{E} is generated:

$$\mathcal{E}_x = -\nabla_x \phi = -\frac{1}{e^*} \left(\frac{d\mu}{dT} \right) \nabla_x T. \quad (3)$$

Under these conditions the diagonal components of the thermoelectric tensor $\hat{\beta}$ 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 a 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):

$$\hat{\beta} = \begin{pmatrix} -\frac{\sigma_{xx}}{e^*} \frac{d\mu}{dT} & c \frac{dM_z}{dT} \\ -c \frac{dM_z}{dT} & -\frac{\sigma_{yy}}{e^*} \frac{d\mu}{dT} \end{pmatrix}. \quad (4)$$

Using these relations one can express the Hall thermal flow as

$$q_y = -\kappa_{yx} \nabla_x T = \gamma_{yx} \mathcal{E}_x. \quad (5)$$

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:

$$\kappa_{yx} = \frac{cT}{e^*} \left(\frac{dM}{dT} \right) \left(\frac{d\mu}{dT} \right). \quad (6)$$

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 the y direction is frequently measured rather than the thermal flow. This quantity, also known as the Righi-Leduc coefficient [4], is dependent on both diagonal and nondiagonal components of the tensor $\hat{\kappa}$ and cannot be directly described by the expression proposed here for the thermodynamic contribution to κ_{yx} . However, we believe that the thermodynamic formula (6) grasps the essential physics that is behind the observed effect.

To start with, using the general relation Eq. (6) obtained above, 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 a 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 as well as in the high temperature limit, far above the critical temperature. Then, we shall address the thermal Hall effect in a normal metal where no Cooper pairs can be formed but the repulsive interaction in a particle-particle channel leads to the renormalization of the electron effective mass. We show that also in this case the temperature derivative of the chemical potential is much larger than one of a degenerate Fermi gas of noninteracting electrons, which induces that superlinear increase of the thermal Hall conductivity with the decrease of temperature.

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 fluctuation superconductor in the 2D case that one can find in Ref. [15]:

$$F_{(2)}^{(fl)}(\epsilon, h) = -\frac{T_{c0}S}{4\pi\xi^2} \left[\epsilon \ln \frac{1}{2h} - 2h \ln \frac{\Gamma(\frac{1}{2} + \frac{\epsilon}{2h})}{\sqrt{2\pi}} \right]. \quad (7)$$

Here, S is the sample cross section and $\xi^2 = \pi\mathcal{D}/8T_{c0}$ is the superconducting coherence length, \mathcal{D} is the electron diffusion coefficient, $\epsilon = \ln T/T_{c0} \approx (T - T_{c0})/T_{c0} \ll 1$ is the reduced temperature, T_{c0} is critical temperature of the superconducting phase transition at zero magnetic field. The dimensionless magnetic field $h = H/\tilde{H}_{c2}(0) \ll 1$ is normalized with the second critical field $\tilde{H}_{c2}(0) = \Phi_0/2\pi\xi^2$, introduced as the linear extrapolation to zero temperature 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]:

$$M_{(2)}^{(fl)}(\epsilon, h) = \frac{T_{c0}}{\Phi_0} \left\{ \ln \frac{\Gamma(\frac{1}{2} + \frac{\epsilon}{2h})}{\sqrt{2\pi}} - \frac{\epsilon}{2h} \left[\psi \left(\frac{1}{2} + \frac{\epsilon}{2h} \right) - 1 \right] \right\}, \quad (8)$$

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

$$\begin{aligned} \frac{dM_{(2)}^{(fl)}(\epsilon, h)}{dT} &= -\frac{1}{2h\Phi_0} \left[\frac{\epsilon}{2h} \psi' \left(\frac{1}{2} + \frac{\epsilon}{2h} \right) - 1 \right] \\ &= \frac{h}{\Phi_0} \begin{cases} 1/6\epsilon^2, & h \ll \epsilon \ll 1, \\ 1/2h^2, & \epsilon \ll h \ll 1, \\ 1/\epsilon_h^2, & \epsilon_h \ll h, \end{cases} \end{aligned} \quad (9)$$

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 both in the vicinity of T_{c0} and at high temperatures [18], far from 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 $N_{(2)}^{(fl)}$. The latter can be easily obtained by means of integration of the distribution function of the Cooper pairs over momenta. This procedure yields

$$\mu_{(2)}^{(fl)}(\epsilon, h) = -T_{c0}e \frac{\ln \frac{1}{2h} - \frac{2h}{\epsilon} \ln \frac{\Gamma(1/2 + \epsilon/2h)}{\sqrt{2\pi}}}{\ln \frac{1}{2h} - \psi(\frac{1}{2} + \frac{\epsilon}{2h})}. \quad (10)$$

The details of this derivation are given in the Supplemental Material [19].

Thermal Hall conductivity due to fluctuating Cooper pairs close to T_{c0} .—Now, the thermal Hall conductivity can be represented explicitly:

$$\begin{aligned} \tilde{\kappa}_{yx(2)}^{(fl)}(\epsilon, h) &= -\frac{T_{c0}}{4\pi h} \left[1 - \frac{\epsilon}{2h} \psi' \left(\frac{1}{2} + \frac{\epsilon}{2h} \right) \right] \\ &\times \left[1 - \frac{\epsilon}{2h} \psi' \left(\frac{1}{2} + \frac{\epsilon}{2h} \right) \frac{\ln \frac{1}{2h} - \frac{2h}{\epsilon} \ln \frac{\Gamma(1/2 + \epsilon/2h)}{\sqrt{2\pi}}}{[\ln \frac{1}{2h} - \psi(\frac{1}{2} + \frac{\epsilon}{2h})]^2} \right]. \end{aligned} \quad (11)$$

It is instructive to express Eq. (11) in its asymptotic form

$$\tilde{\kappa}_{yx(2)}^{(fl)}(\epsilon, h) = -\frac{e\mathcal{D}H}{64c} \begin{cases} 1/3\epsilon^2, & h \ll \epsilon \ll 1, \\ 1/h^2, & \epsilon \ll h \ll 1, \\ \epsilon_h^{-2} \ln \frac{2h}{\epsilon_h}, & \epsilon_h \ll h. \end{cases} \quad (12)$$

Here, we operate with the true magnetic field $H = h\tilde{H}_{c2}(0)$. We took advantage of the relation between the GL extrapolation of the second critical field and the BCS one: $\tilde{H}_{c2}(0) = (8\gamma_E/\pi^2)H_{c2}^{\text{BCS}}(0)$. We note that in the BCS theory $H_{c2}(0) = (2/\gamma_E)\Phi_0(T_{c0}/\mathcal{D})$, where $\gamma_E = 1.78$ is the Euler constant.

Fluctuation superconductor: the high temperature limit $T \gg T_{c0}$.—In this limit, fluctuation Cooper pairs can still be formed, and their concentration can be estimated based on the fluctuation theory [19]. Assuming that the Cooper pairs obey the Bose-Einstein statistics, we obtain a relation between the chemical potential and the concentration of Cooper pairs that allows us to obtain the temperature derivative of the chemical potential. Expressing the magnetization as in Refs. [20,21], we obtain the thermal Hall conductivity as

$$\kappa_{(2)}^{(\text{fl})}(T \gg T_{c0}) = -\frac{2\pi^2 eDH}{3c} \left[1 - \frac{\ln \left(\ln \ln \frac{1}{T_{c0}\tau} - \ln \ln \frac{T}{T_{c0}} \right)}{\ln(T/T_{c0})} \right].$$

Here, τ is a characteristic scattering time. We conclude that the temperature dependence of the thermal Hall conductivity becomes less pronounced as one gets farther from the phase transition point, however, the superlinear character of this dependence and the negative sign of κ_{yx} are maintained.

Normal metal. Contribution to the thermal Hall effect due to electron-electron interactions.—We consider a normal metal, where no fluctuation Cooper pairs are formed but the repulsive Coulomb interaction between electrons is important. This is the regime that is observed in some of the cuprates where no superconducting phase transition is observed. We analyze the temperature dependencies of the magnetization and the chemical potential here. It is important to emphasize the role of interelectron interaction in the particle-particle channel that may be considered as a counterpart of Cooper pairing and leads to the renormalization of the electron effective mass [22]. Because of this renormalization that is strongly temperature dependent, the derivative of the chemical potential over temperature strongly varies as a function of temperature, which is crucial for the understanding of the temperature dependence of the thermal Hall conductivity. As shown in the Supplemental Material, the following expression for κ_{yx} is valid in this case:

$$\kappa_{(2)}^{(g)} = -\frac{\pi eDH}{3c} \frac{1}{T\tau \ln^2(T_K/T)}. \quad (13)$$

Note, that the sign of the thermal Hall conductivity is negative, and it increases with the temperature decrease in a qualitative similarity to the behavior characteristic of up-critical superconductors.

Effect of quantum fluctuations on the thermal Hall conductivity above $H_{c2}(0)$.—Using the general

thermodynamic relation (6) one can predict the behavior of thermal Hall conductivity above $H_{c2}(0)$ also in the limit of very low temperatures, in the domain of quantum fluctuations (QF). The behavior of the fluctuation magnetization in this regime was studied in Ref. [23]:

$$M_{(2)}^{(\text{fl})}(t, \tilde{h}) = \frac{T_{c0}}{\gamma_E \Phi_0} \left[\ln \frac{1}{2\gamma_E t} - \frac{\gamma_E t}{\tilde{h}} - \psi \left(\frac{\tilde{h}}{2\gamma_E t} \right) \right], \quad (14)$$

with $t = T/T_{c0} \ll 1$ and $\tilde{h} = [H - H_{c2}(T)]/H_{c2}(T) \ll 1$. The differentiation of Eq. (14) results in

$$\begin{aligned} \frac{dM_{(2)}^{(\text{fl})}(t, \tilde{h})}{dT} &= \frac{1}{\gamma_E \Phi_0} \left[\frac{\tilde{h}}{2\gamma_E} \frac{\psi'}{t^2} \left(\frac{1}{2\gamma_E} \frac{\tilde{h}}{t} \right) - \frac{1}{t} - \frac{\gamma_E}{\tilde{h}} \right] \\ &= \frac{1}{\Phi_0} \begin{cases} 2\gamma_E t / 3\tilde{h}^2, & t \ll \tilde{h} \ll 1, \\ 1/\tilde{h}, & \tilde{h} \ll t \ll 1. \end{cases} \end{aligned} \quad (15)$$

In the vicinity of $H_{c2}(0)$, the chemical potential of fluctuation Cooper pairs can be written as $\mu^{(\text{QF})} = -\Delta_{\text{BCS}}\tilde{h}$ (similarly to the expression valid at T_{c0} , see Ref. [24]. Its temperature derivative differs from zero due to the temperature dependence of $H_{c2}(T)$ (see Ref. [25]):

$$\frac{d\mu^{(\text{QF})}}{dT} = \frac{\Delta_{\text{BCS}}}{H_{c2}(0)} \left(\frac{dH_{c2}(T)}{dT} \right) = -\frac{2\gamma_E}{\pi} t. \quad (16)$$

Substituting Eqs. (15) and (16) into Eq. (6) and taking into account that $T_{c0} = (\pi/\gamma_E)\Delta_{\text{BCS}}$ one finally finds

$$\begin{aligned} \tilde{\kappa}_{yx(2)}^{(\text{fl})}(t, \tilde{h}) &= \frac{\Delta_{\text{BCS}}}{\pi} \left[t + \frac{\gamma_E t^2}{\tilde{h}} - \frac{\tilde{h}}{2\gamma_E} \psi' \left(\frac{1}{2\gamma_E} \frac{\tilde{h}}{t} \right) \right] \\ &= -\frac{\Delta_{\text{BCS}}}{\pi} t^2 \begin{cases} 2\gamma_E t / 3\tilde{h}^2, & t \ll \tilde{h} \ll 1, \\ 1/\tilde{h}, & \tilde{h} \ll t \ll 1. \end{cases} \end{aligned} \quad (17)$$

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

Results and discussion.—Figure 2 shows the thermal Hall conductivity as a function of the reduced temperature and magnetic field with enlargements of the areas corresponding to the low magnetic field and quantum fluctuation regimes. One can see that, while the specific shape of the dependence may vary, $\kappa_{yx}^{(\text{fl})}(\epsilon, h)$ always has a negative sign, and its absolute value increases rapidly with the temperature decrease. The same universal behavior is found in the high temperature limit and in normal metals, as discussed above.

Now one can compare the predictions of our theory with the experimental results reported in Ref. [6] for four cuprates. 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

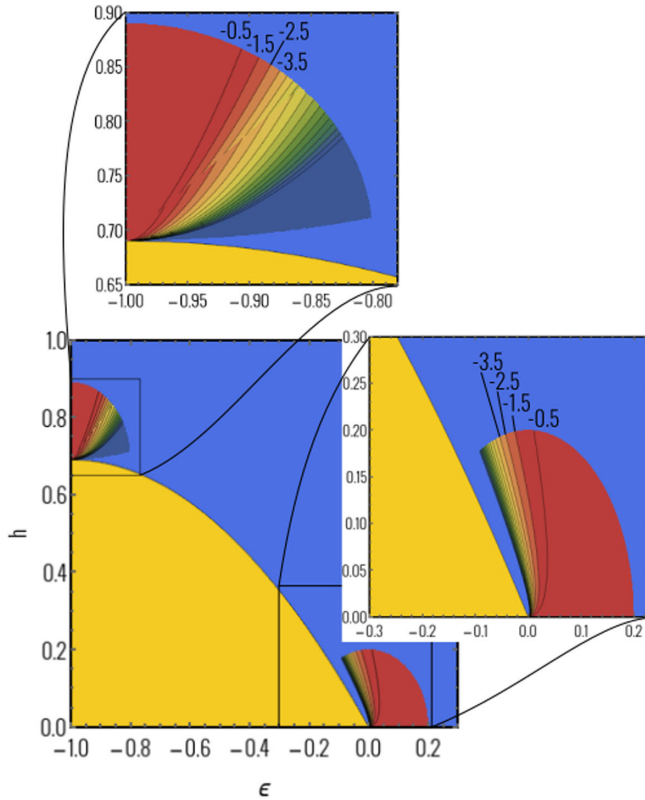


FIG. 2. Fluctuation induced thermal Hall conductivity $\tilde{\kappa}_{yx}^{(n)}$ (shown by the color scale and numbers in arbitrary units) as a function of dimensionless temperature and magnetic field. The blue area shows the normal phase above the superconducting transition, the yellow area corresponds to the superconducting state. Our consideration is valid in the blue area in the domain close to the critical temperature T_{c0} [see Eq. (11)] and in the vicinity of the second critical field $H_{c2}(0)$ [see Eq. (17)].

decrease. We believe that the qualitative agreement of such a simple model with the large variety of 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 for the description of up-critical superconductors 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 studied 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 the ones of a conventional superconductor. This is confirmed in the recent study Ref. [26], that went beyond the weak-fluctuation formalism, applied the precursor-pairing approach within the BCS to Bose-Einstein condensation crossover scheme [27,28] 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.

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