Title: Quantum Kinetic Approach to the Calculation of Thermal Transport and the Nernst Effect

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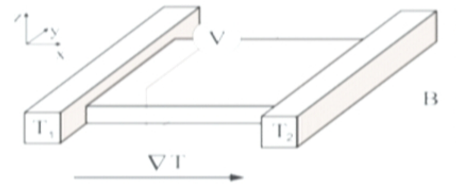
Abstract: Recently, we developed a user friendly scheme based on the quantum kinetic equation for studying thermal transport phenomena in the presence of interactions and disorder . This scheme is suitable for both a systematic perturbative calculation as well as a general analysis. We believe that this method presents an adequate alternative to the Kubo formula, which for thermal transport is rather cumbersome. We have applied this approach in the study of the Nernst signal in superconducting films above the critical temperature. We showed that the strong Nernst signal observed in amorphous superconducting films, far above Tc, is caused by the fluctuations of the superconducting order parameter. We demonstrated a striking agreement between our theoretical calculations and the experimental data at various temperatures and magnetic fields. My talk will include a general description of the quantum kinetic approach, but mainly I will concentrate on the Nernst effect in superconducting films. I will use this example to discuss some subtle issues in the theoretical study of thermal phenomena that we encountered while calculating the Nernst coefficient. In particular, I will explain how the Nernst theorem (the third law of thermodynamics) imposes a strict constraint on the magnitude of the Nernst effect.


PRB 80, 214516 (2009).
PRB. 80, 115111 (2009).
EPL. 86, 27007 (2009).
cations of Thermoelectric Materials, p.213.
Physics

Massachusetts Institute of Technology



Quantum kinetic approach to the calculation of thermal transport and the Nernst effect

Karen Michaeli and Alexander M. Finkel'stein

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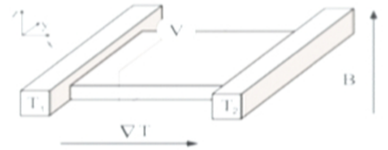
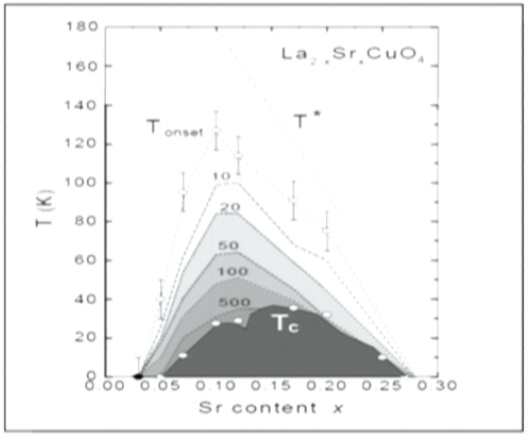
Properties and Applications of Thermoelectric Materials, p.213.

MIT Pappalardo Fellowships in Physics

Nernst Effect- High Tc Materials



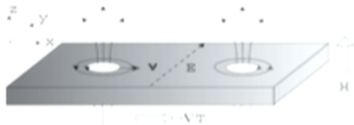
$$\nu = \frac{E_y}{-\nabla_x T \cdot B}$$



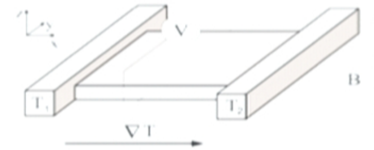
Y. Wang, *et al* 2005

Usually explained by the existence of vortices. → Pairing must survive above T_C. → Disappearance of phase coherence at T_C although the gap is still finite

Anderson, 2007
Raghu, et al, 2008
Mukerjee and Huse 2004



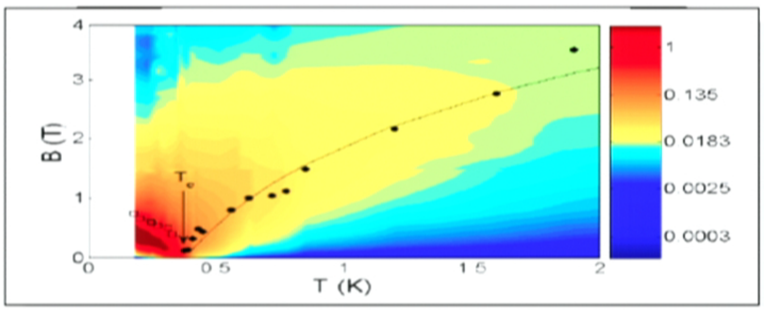
Nernst Effect – Conventional Superconductors



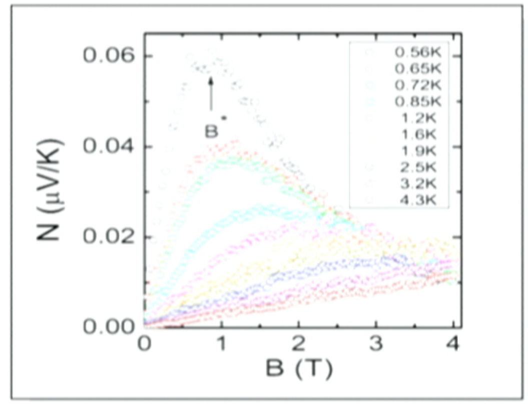
The Nernst signal

$$\nu = \frac{E_y}{-\nabla_x T \cdot B}$$

The strong Nernst signal above Tc can not be explained by the vortex-like fluctuations.



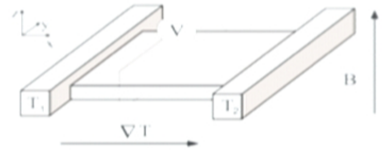
$Nb_{0.15}Si_{0.85}$



A. Pourret, *et al* 2007

It has been suggested that the fluctuations of the order parameter cause the effect.

Nernst Effect - Conventional Superconductors



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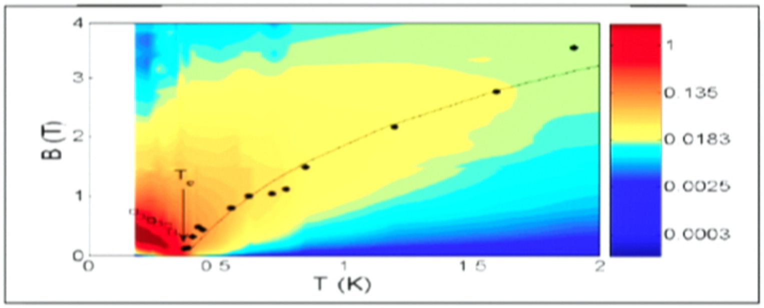

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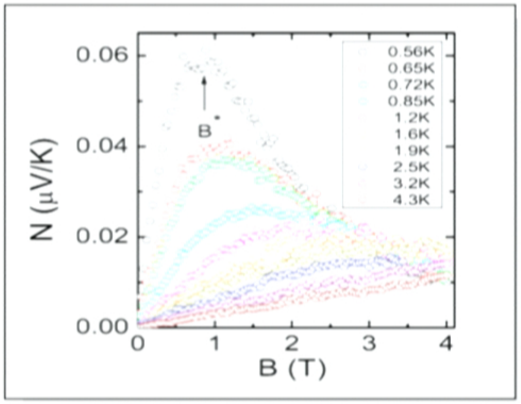

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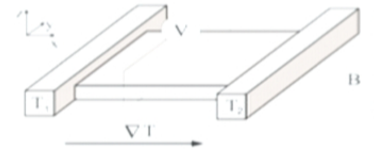
Why Superconducting Fluctuations and Not Quasi-Particles?



The electric current in response to a temperature gradient in a system with two species of particles (electrons and holes):

The Boltzmann equation for the distribution function:

$$\frac{\delta f_{e/h}(\mathbf{k})}{\tau} = \frac{\partial f_0(\varepsilon_{\mathbf{k}})}{\partial T}\mathbf{v}_{\mathbf{k}} \cdot \nabla T \mp \frac{e\mathbf{v}_{\mathbf{k}}}{c} \times \mathbf{B} \cdot \frac{\delta f_{e/h}(\varepsilon_{\mathbf{k}})}{\partial \mathbf{k}}$$

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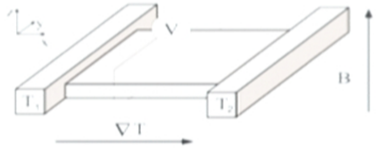

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The electric current :

$$\mathbf{j}_e = -e\int \frac{d\mathbf{k}}{(2\pi)^d}\mathbf{v}_{\mathbf{k}}\delta f_e + e\int \frac{d\mathbf{k}}{(2\pi)^d}\mathbf{v}_{\mathbf{k}}\delta f_h$$

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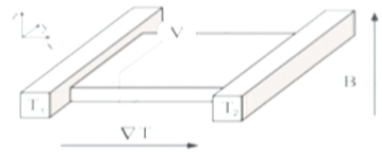

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The ***longitudinal*** electric current:

$$j_e^x = e\int\frac{d\mathbf{k}}{(2\pi)^d}\frac{\partial f_0(\varepsilon_\mathbf{k})}{\partial \varepsilon_\mathbf{k}}\left[\frac{\varepsilon_\mathbf{k}v_\mathbf{k}^2\tau}{d} - \frac{\varepsilon_\mathbf{k}v_\mathbf{k}^2\tau}{d}\right]\frac{\nabla_x T}{T} = 0$$

Vanishes due to particle - hole symmetry

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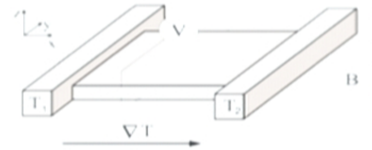

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The ***transverse*** electric current:

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$$\omega_C = \frac{eB}{mc}$$

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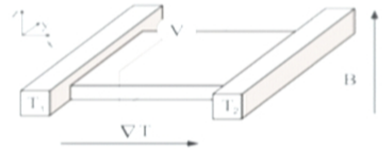

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Particle-hole symmetry does not constrain the magnitude of the Nernst effect.

Under the approximation of a constant density of states:

$$j_e^y = 2e^2(\omega_C\tau)\frac{v_F^2\tau}{d}\frac{\nabla_x T}{T}\int\frac{d\varepsilon_{\mathbf{k}}}{(2\pi)^d}\nu_0\frac{\partial f_0(\varepsilon_{\mathbf{k}})}{\partial\varepsilon_{\mathbf{k}}}\varepsilon_{\mathbf{k}} = 0$$

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For the collective modes the effective density of states is far from being a constant.

The neutral modes are not deflected by the Lorentz force.

The charged modes such as fluctuations of superconducting order parameter generate the Nernst effect.

The Nernst Coefficient

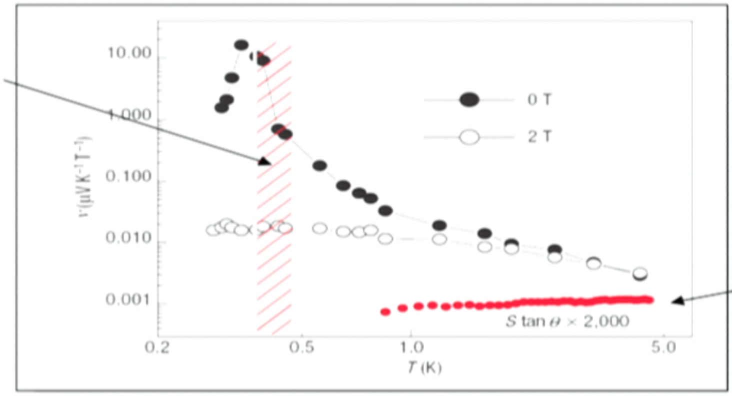
$$\begin{pmatrix} j_e \\ j_h \end{pmatrix} = \begin{pmatrix} \sigma & \alpha \\ \tilde{\alpha} & \kappa \end{pmatrix} \begin{pmatrix} E \\ -\nabla T \end{pmatrix}$$

$$e_N = \frac{E_y}{-\nabla_x T} = \frac{\alpha_{xy}\sigma_{xx} - \alpha_{xx}\sigma_{xy}}{\sigma_{xy}^2 + \sigma_{xx}^2}$$

$$e_N \approx \frac{\alpha_{xy}}{\sigma_{xx}}$$

α_{xx} is negligible in comparison to α_{xy}

$$\nu = \frac{e_N}{H}$$



$$S \tan\theta = \frac{\alpha_{xx}\sigma_{xy}}{\sigma_{xx}^2}$$

A. Pourret, et al 2006

Electric Conductivity - Kubo Formula

Luttinger approach - J. M. Luttinger 1964.

Electric conductivity:

The density matrix at $t = -\infty$ is $\rho_0 = e^{-\beta H_0}$.

Adiabatically switching on a scalar potential $H = H_0 + e\int e^{-st}\phi(\mathbf{r})n(\mathbf{r})d\mathbf{r}$

The E.O.M for the density matrix

$$i\frac{d\rho(t)}{dt} = [H, \rho(t)]$$

Density continuity equation:

$$e\dot{n} + \nabla j = 0$$

The Kubo formula for the linear response to the field

$$\langle j_e(r) \rangle = -\langle e^{-\beta H_0} j_e j_e \rangle \nabla \varphi$$

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The diffusion coefficient:

The electro-chemical potential is kept constant : $\zeta_0 = \mu(r) + e\varphi(r)$

The system is at equilibrium: $\langle j(r)\rangle = -\sigma\nabla\varphi - eD\nabla n = 0$

Einstein's relation

$$D = -\frac{\sigma\nabla\varphi}{e\nabla n} = \sigma\frac{\nabla\mu}{e^2\nabla n}$$

Thermal Conductivity - Kubo Formula

Luttinger approach - J. M. Luttinger 1964.

Thermal conductivity:

Introducing a mechanical force field that is coupled to the Hamiltonian density:

$$H = \int h_0(r)dr + \int e^{-st}\gamma(r)h_0(r)dr$$

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Energy continuity equation:

$$\dot{h} + \nabla j_h = 0$$

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$$H = \int h_0(r)dr + \int e^{-st}\gamma(r)h_0(r)dr$$

To keep the system in equilibrium, two compensating currents are introduced:

$$j_h^i(\mathbf{r}) = \kappa_{ij} T \nabla_j \left(\frac{1}{T}\right) + \tilde{\kappa}_{ij}\left(-\nabla_j \gamma\right) = 0$$

Using thermodynamic identities, one obtains

$$\left(\frac{1}{T}\right)_{\mathbf{q}} = \frac{V}{T}\gamma(\mathbf{q}) \longrightarrow \kappa_{ij} = \tilde{\kappa}_{ij}$$

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Luttinger's expression for the current operator:

J. M. Luttinger 1964.

$$j_h^{\upsilon} = \frac{1}{2}\sum_i \left(h_i j_i^{\upsilon}(\mathbf{r}) + j_i^{\upsilon}(\mathbf{r}) h_i \right) - \frac{1}{8m}\sum_{i,i'} \left[\left(r_i^{\sigma} - r_{i'}^{\sigma} \right) \frac{\partial u(\mathbf{r}_i, \mathbf{r}_{i'})}{\partial r_i^{\sigma}} \delta(\mathbf{r} - \mathbf{r}_i) + \delta(\mathbf{r} - \mathbf{r}_i)\left(r_i^{\sigma} - r_{i'}^{\sigma} \right) \frac{\partial u(\mathbf{r}_i, \mathbf{r}_{i'})}{\partial r_i^{\sigma}} \left(p_i^{\upsilon} + p_{i'}^{\upsilon} \right) \right]$$

where

$$j_i^{\nu} = \frac{1}{2m}\left(p_i^{\nu} \delta(\mathbf{r} - \mathbf{r}_i) + \delta(\mathbf{r} - \mathbf{r}_i) p_i^{\nu} \right)$$

$$h_i = \frac{p_i^2}{2m} + V_{imp}^{i} + \frac{1}{2}\sum_{i' \neq i} u(\mathbf{r}_i, \mathbf{r}_{i'})$$

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$$+ \delta(\mathbf{r} - \text{[illegible]}) \frac{\text{[illegible]}(\mathbf{r}_i, \mathbf{r}_{i'})}{\partial r_i^{\sigma}} \left(p_i^{\nu} + p_{i'}^{\nu}\right) \Big]$$

How exactly to use such a vertex?

where

$$j_i^{\nu} = \frac{1}{2m}\left(p_i^{\nu} \delta(\mathbf{r} - \mathbf{r}_i) + \delta(\mathbf{r} - \mathbf{r}_i) p_i^{\nu}\right)$$

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Thermal Conductivity - Vertex Term

The common expression used in the Kubo formula:

$$j_h^{v}(\mathbf{q},\omega) = \frac{2\varepsilon_m + \omega_m}{2e} j_e^{v}(\mathbf{k}+\mathbf{q},\omega+\varepsilon)$$

Where are the corrections due to the interaction?

Test: The simplified version of the Kubo formula fails to reproduce the phenomenologically known result that the Wiedemann-Franz law is valid for Fermi liquids.

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$$+\delta(\mathbf{r}-\mathbf{r}_i)\left(r_i^{\sigma} - r_{i'}^{\sigma}\right)\frac{\partial u(\mathbf{r}_i,\mathbf{r}_{i'})}{\partial r_i^{\sigma}}\left(p_i^{\upsilon} + p_{i'}^{\upsilon}\right)\Bigg]$$

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Nernst Effect - Magnetization

There has been a long discussion about the contribution of magnetization to the thermoelectric transport currents.

For example:
Obraztsov Sov. Phys. Solid State 1965
Smrcka and Streda J. Phys. C 1977
Cooper, Halperin and Ruzin PRB 1997

In the presence of magnetic field the thermodynamic expression for the heat contains the magnetization term:

$$dQ = TdS = dE - \mu dN + MdB.$$

The Kubo formula is not enough, the contribution from the magnetization must be added.


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N + MdB.
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Quantum Kinetic Approach

Derivations of the transport coefficients using the kinetic equation already exist, for example:

J.-W. Wu, and G. D. Mahan, 1984.
G. Strinati, C. Castellani, C. DiCastro, and G. Kotliar, 1991.
D. V. Livanov, M. Y. Reizer, and A. V. Sergeev, 1991.
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Our Scheme Differs in Few Aspects

The kinetic equations and the currents are derived from the **action** and the conservation laws emerging from the action.

For the Coulomb interaction all the continuity currents share a common simple structure:

$$\mathbf{j}_{e,h} = -i\int \frac{d\varepsilon}{2\pi} \chi_{e,h}(\varepsilon)\left[\hat{\mathbf{v}}(\varepsilon)\hat{G}(\varepsilon)\right]^{<}$$

$$\chi_e(\varepsilon) = -e \quad , \quad \chi_h(\varepsilon) = \varepsilon$$

The renormalized velocity

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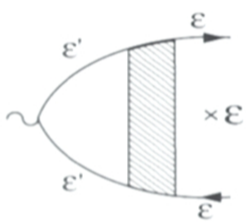

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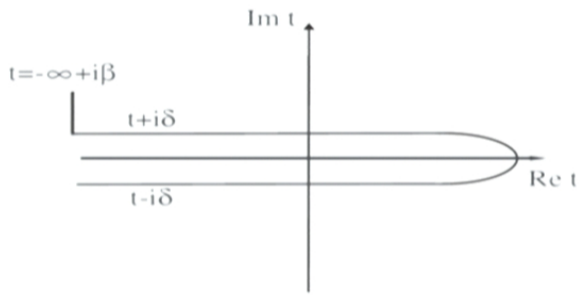

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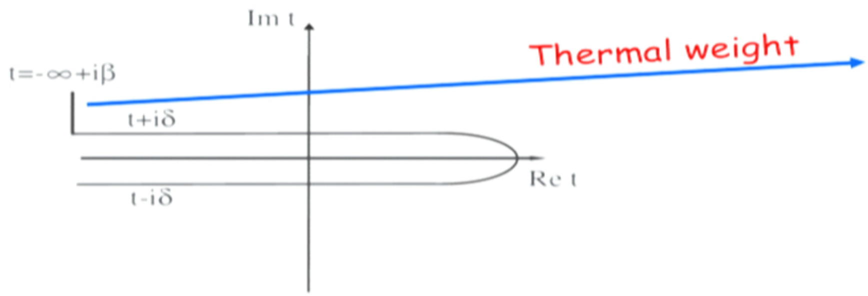

$$\hat{\rho} = \exp\left\{-i\int d\mathbf{r} \int_{-\infty+i\beta(\mathbf{r})}^{-\infty+i\delta} dt\left[h(\mathbf{r}) - \mu n(\mathbf{r})\right]\right\}$$

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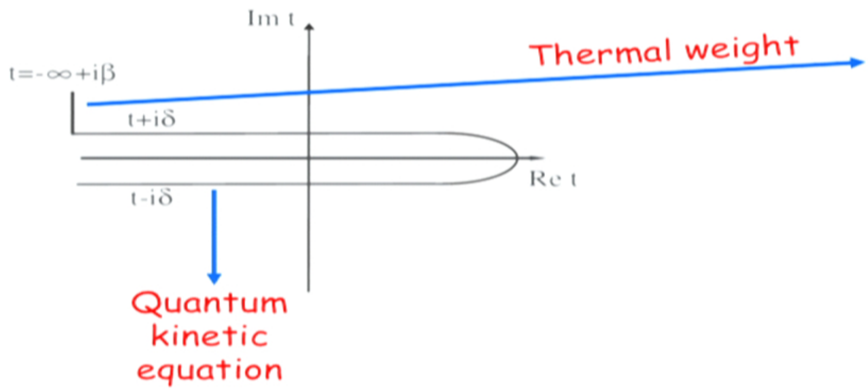

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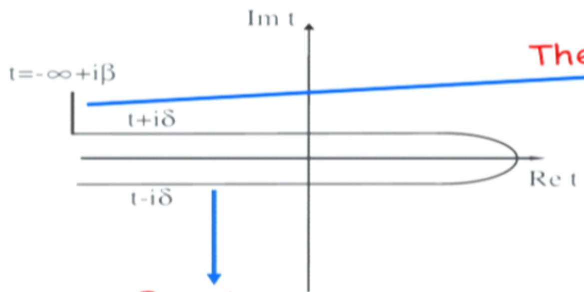


Thermal weight

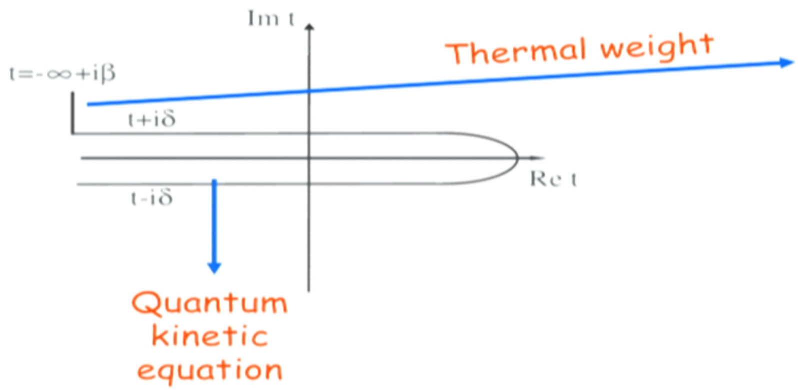
$$\hat{\rho} = \exp\left\{ -i \int d\mathbf{r} \int_{-\infty + i\beta(\mathbf{r})}^{-\infty + i\delta} dt \left[h(\mathbf{r}) - \mu n(\mathbf{r}) \right] \right\}$$

Quantum kinetic equation

We have to find highly non-trivial initial state before we even start to study its time evolution

Quantum Kinetic Approach

The Keldysh Green function: $$\hat{G}(r,t;r't') = \begin{pmatrix} G_T & G^< \\ G^> & G_{\tilde{T}} \end{pmatrix} \Leftrightarrow \begin{pmatrix} G^R & G^K \\ & G^A \end{pmatrix}$$



$$\hat{\rho} = \exp\left\{ -i\int d\mathbf{r} \int_{-\infty+i\beta(\mathbf{r})}^{-\infty+i\delta} dt'\gamma(\mathbf{r})\left[h(\mathbf{r}) - \mu n(\mathbf{r})\right] \right\}$$

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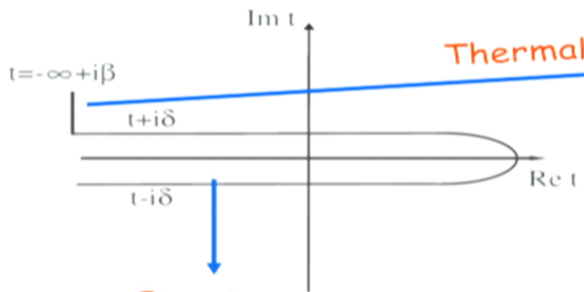

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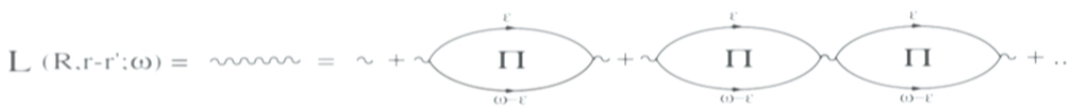
Quantum kinetic equation

We introduce the field γ(r) in such a way that the system is initially in fully equilibrium state due to the balancing between the temperature gradient and gravitation field.

Nernst Effect- The Quantum Kinetic Approach

We introduce two propagators:

1. The electronic Green's function $\hat{G}\left(\mathbf{R}=\frac{\mathbf{r}+\mathbf{r}'}{2},\mathbf{r}-\mathbf{r}';\varepsilon\right)$
2. The propagator of the superconducting fluctuations $\hat{L}\left(\mathbf{R}=\frac{\mathbf{r}+\mathbf{r}'}{2},\mathbf{r}-\mathbf{r}';\varepsilon\right)$



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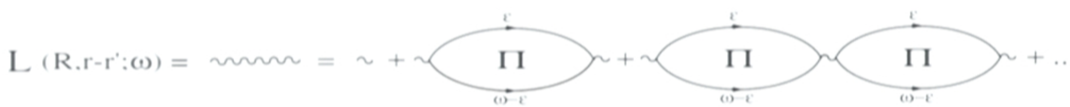

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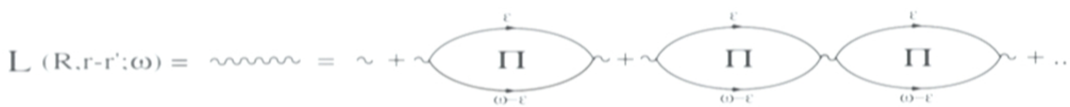


The electric current as a response to a temperature gradient :

$$\mathbf{j}_e = \mathbf{j}_e^{con} + \mathbf{j}_e^{mag}$$

$$\mathbf{j}_e^{con} = ie\int\frac{d\varepsilon}{2\pi}\left[\hat{\mathbf{v}}(\varepsilon)\hat{G}(\varepsilon)\right]^< + 2ie\int\frac{d\omega}{2\pi}\left[\hat{\mathbf{v}}_\Delta(\omega)\hat{L}(\omega)\right]^<$$

$$\mathbf{j}_e = \nabla\times\mathbf{M}G^<(\nabla T)$$

v_Δ is the notation used for $\hat{\mathbf{v}}_\Delta = \frac{\partial\hat{\Pi}(\mathbf{q},\omega)}{\partial\mathbf{q}}$

The ∇T-Dependent Propagators

$\hat{G}\left(\nabla T; \mathbf{R} = \frac{\mathbf{r}+\mathbf{r}'}{2}, \mathbf{r}-\mathbf{r}'; \varepsilon\right)$ consists of two terms:

The first is a straightforward extension of the equilibrium Green's function for a non-uniform temperature.

$$\hat{G}_{loc-eq} = \frac{\mathbf{R}\cdot\nabla T}{T}\varepsilon\frac{\partial \hat{G}_0\left(\mathbf{R}, \mathbf{r}-\mathbf{r}'; \varepsilon\right)}{\partial \varepsilon}$$

The Green's function atequilibrium

The local-equilibrium Green's function is responsible for the non-vanishing contribution of the magnetization current:

$$\mathbf{j}_e = \nabla \times \mathbf{M} G^{<}(\nabla T)$$

The ∇T-Dependent Propagators

$\hat{G}\left(\nabla T;\mathbf{R}=\frac{\mathbf{r}+\mathbf{r}'}{2},\mathbf{r}-\mathbf{r}';\varepsilon\right)$ consists of two terms:

The second term reminds the Green's function in the presence of an electric field

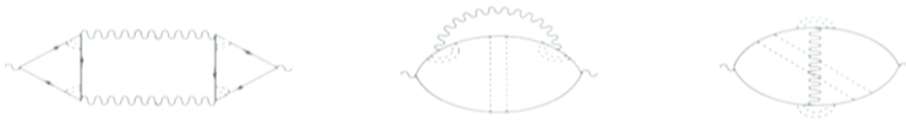
$$\hat{G}_{\nabla T}(\varepsilon)=\hat{g}_{eq}(\varepsilon)\hat{\Sigma}_{\nabla \mathrm{T}}(\varepsilon)\hat{g}_{eq}(\varepsilon)$$
$$-i\frac{\nabla \mathrm{T}}{2T}\varepsilon\left[\frac{\partial\hat{g}_{eq}(\varepsilon)}{\partial\varepsilon}\hat{\mathbf{v}}(\varepsilon)\hat{g}_{eq}(\varepsilon)-\hat{g}_{eq}(\varepsilon)\hat{\mathbf{v}}(\varepsilon)\frac{\partial\hat{g}_{eq}(\varepsilon)}{\partial\varepsilon}\right]$$

The propagator of the fluctuations depends on the temperature gradient only through the polarization operator:

$$\hat{L}(\nabla T)=-\hat{L}_0\hat{\Pi}(\nabla T)\hat{L}_0$$

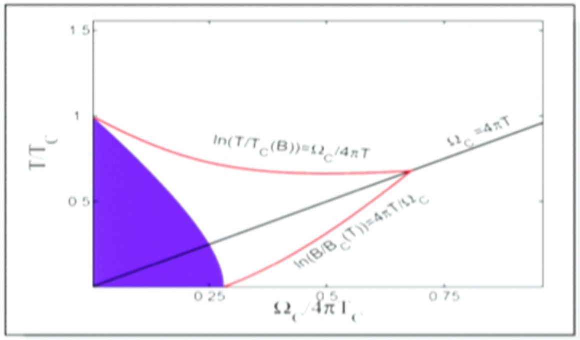
The Peltier Coefficient

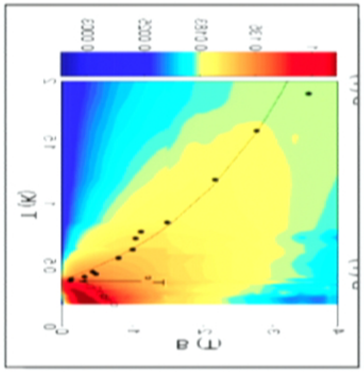
The quantum kinetic equation ≠ diagrammatic method.
The contributions to α_{xy}^{con} can be ***interpreted*** in terms of the following diagrams:



The magnetization current:

$$\alpha_{xy}^{mag} = -\frac{\partial}{\partial B}\frac{eB}{\pi}\sum_{N=0}^{\infty}\sum_{\omega_m}\ln\left\{-\nu\left[\ln\frac{T}{T_C}-\psi\left(\frac{1}{2}+\frac{|\omega_m|+\Omega_C(N+1/2)}{4\pi T}\right)-\psi\left(\frac{1}{2}\right)\right]\right\} \qquad \Omega_c = \frac{4eDH}{c}$$





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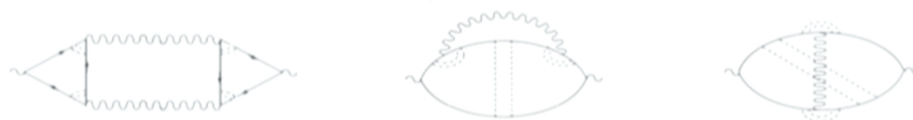

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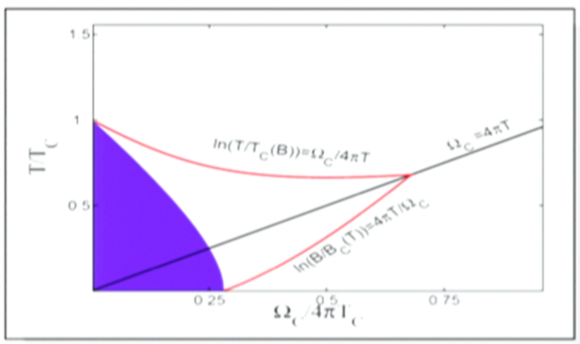
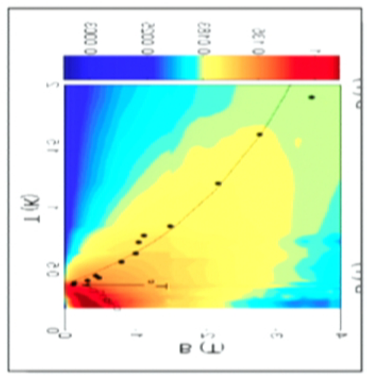
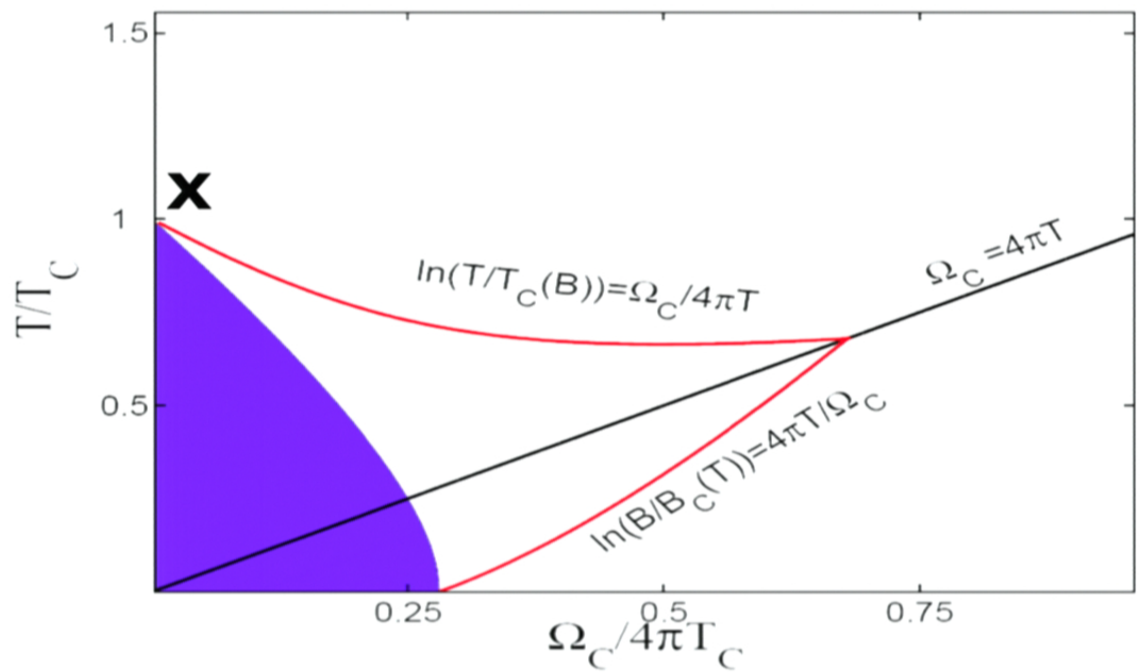




The Peltier coefficient is related to the flow of entropy

The Peltier Coefficient



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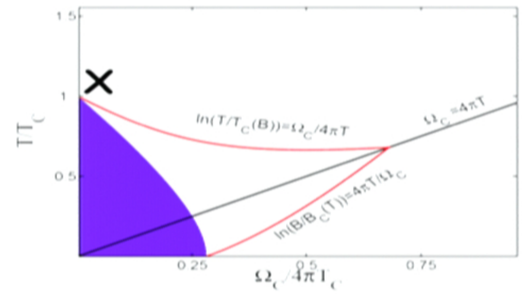

$$\Omega_C << T \qquad \ln\frac{T}{T_C} << 1$$

$$\alpha_{xy} \approx \frac{e\Omega_C}{192T\ln\left(T/T_C(H)\right)}$$

Classical fluctuations - coincide with the phenomenological result of Ussishkin, Sondhi and Huse, 2002

This result is different from the one obtained by the simplified version of the Kubo formula

Serbyn, Skvortsov, Varlamov, and Galitski, 2009

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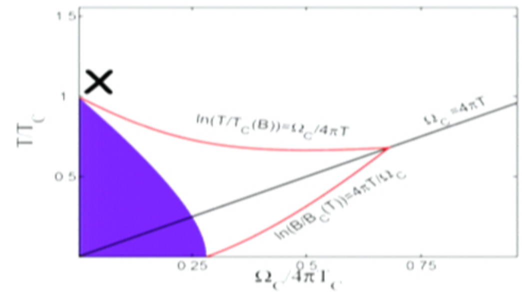

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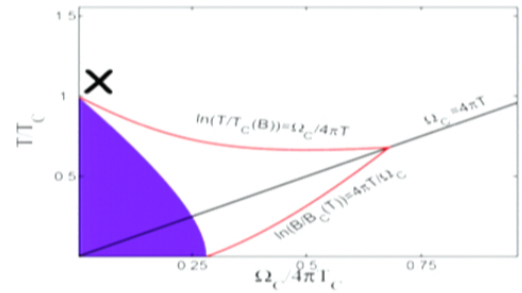

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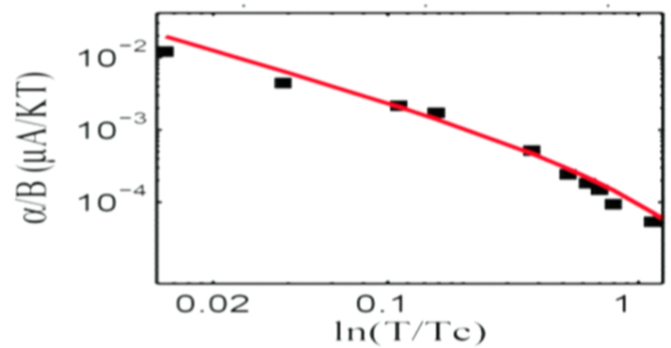
Experimental data from A. Pourret, et al 2007

$Nb_{0.15}Si_{0.85}$ film of thickness $35nm$

and $T_C = 380mK$

$D = 0.187\, cm^2/\mathrm{sec}$ $\qquad T_C^{MF} = 385mK$



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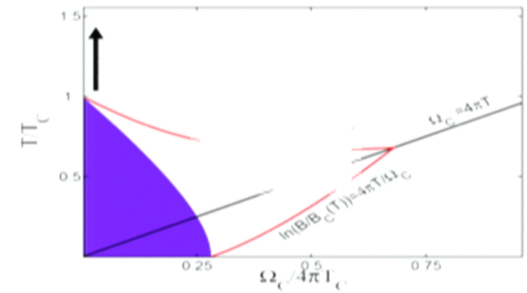

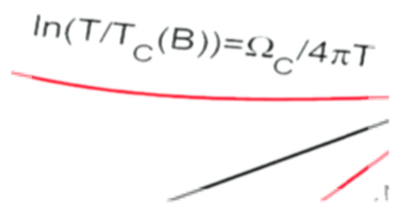

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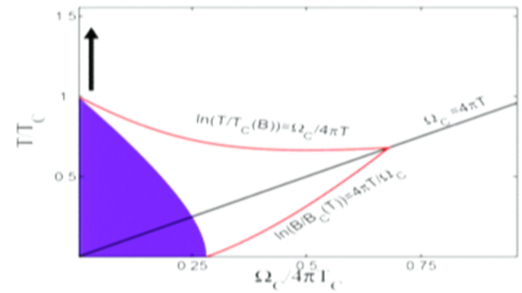

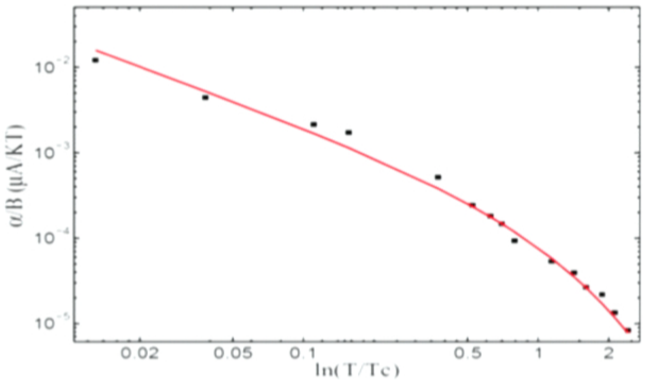

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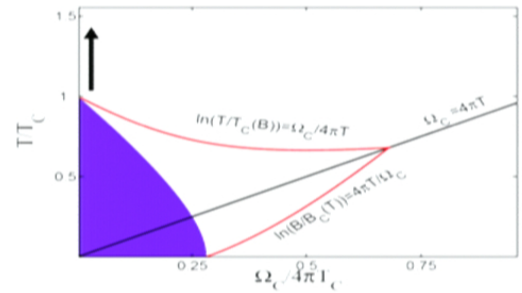

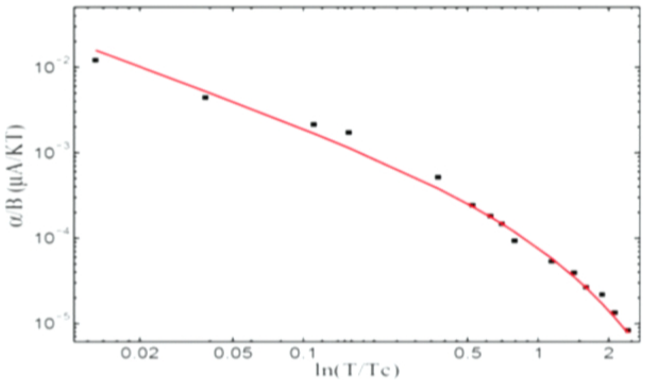

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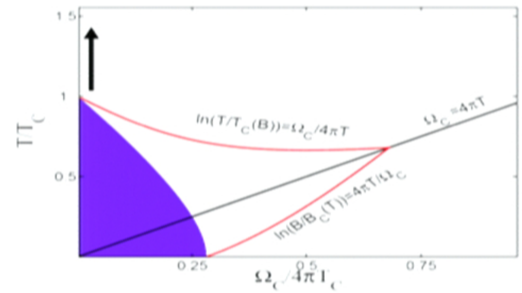

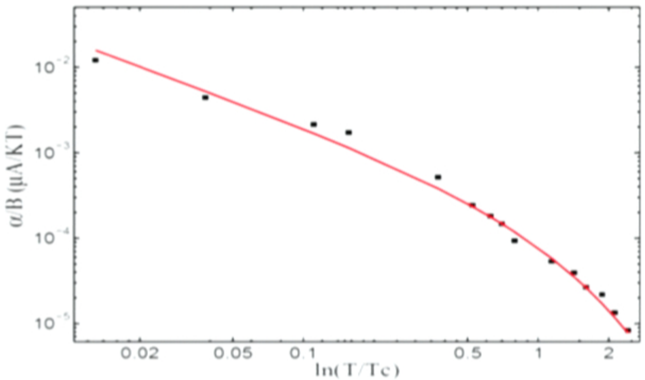

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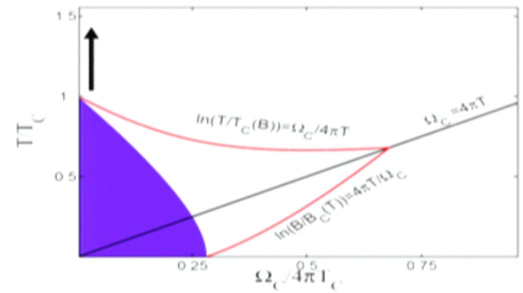
Quantum fluctuations - $T < \omega < 1/\tau$

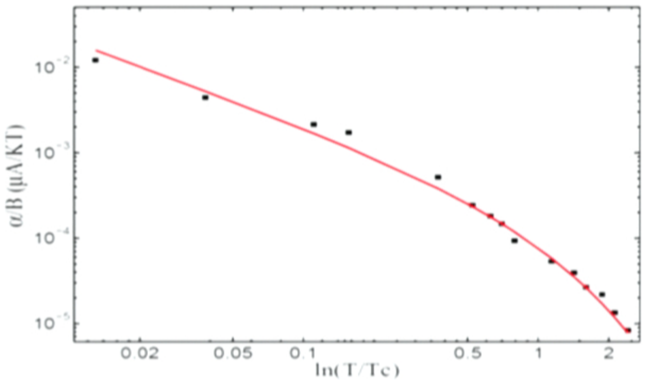
→ $\mathbf{j}_e^{con}$ yield contributions of the order: $\ln\left(\ln\frac{1}{T\tau}\right) - \ln\left(\ln\frac{T}{T_C}\right)$

The logarithmically divergent terms are canceled out by the magnetization current

Trace of the third law of thermodynamics

The Peltier Coefficient





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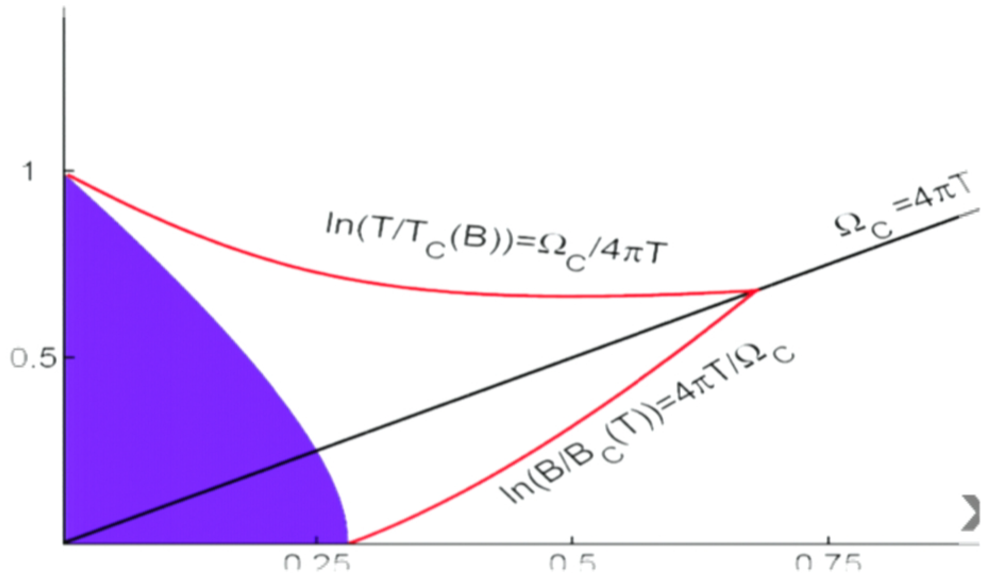
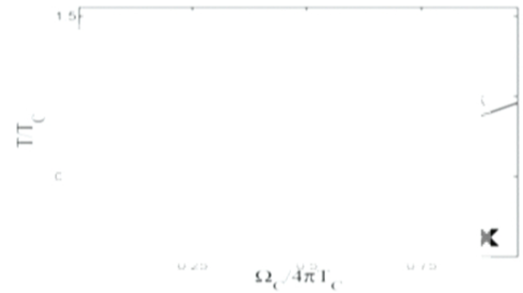
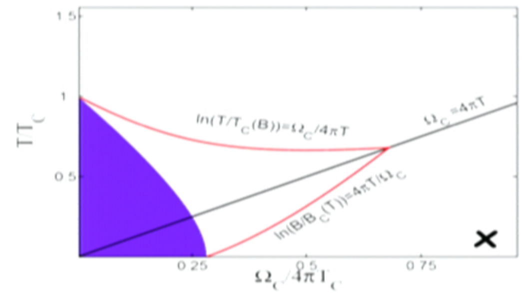




The Peltier Coefficient – High Magnetic field



$\Omega_C >> T \qquad \ln\frac{H}{H_{C2}} >> 1$

$\mathbf{j}_e^{con}$ includes contributions proportional to $\frac{\Omega_C}{T}$.

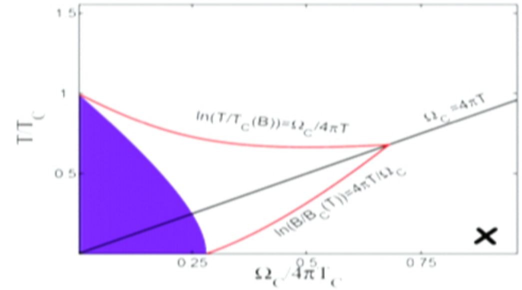
These terms are canceled out by the magnetization current.

$$\alpha_{xy} \approx \frac{2eT}{3\Omega_C \ln\left(H / H_{C2}\right)}$$

The Nernst signal goes to zero at T→0.

Consistent with the third law of thermodynamics.

The Peltier Coefficient – High Magnetic field



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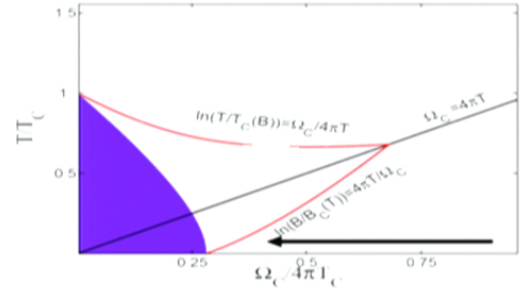

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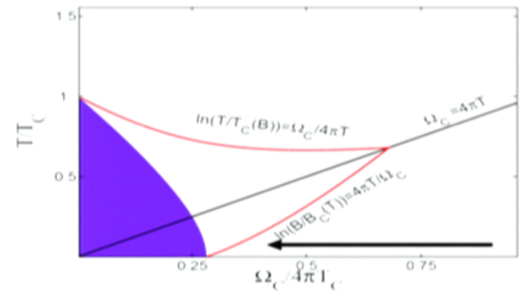

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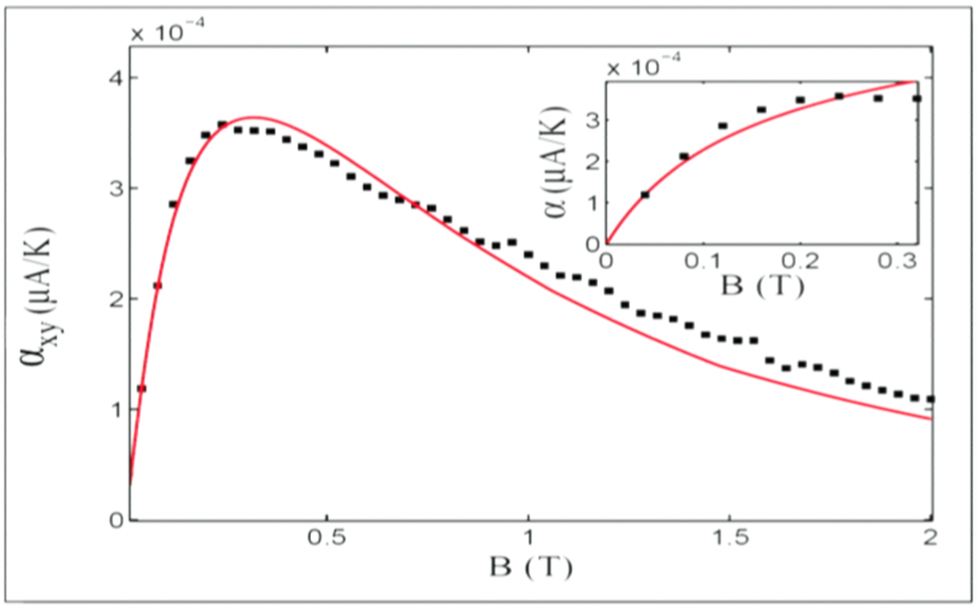
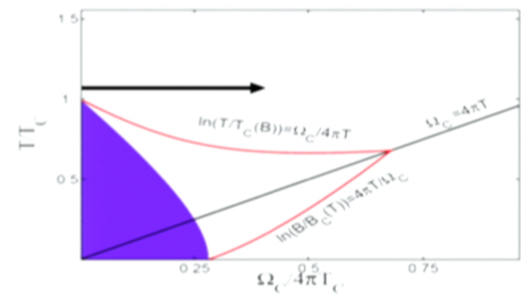
$$\ln\left(\frac{H}{H_C(T)}\right) < \frac{T}{\Omega_C} \qquad \alpha_{xy} \approx -\frac{e\ln 3}{2\pi \ln\left(H / H_C(T)\right)}$$

Since the transverse signal is non-dissipative the sign of the effect is not fixed.

The Peltier Coefficient as a Function of the Magnetic Field





No fitting parameters have been used

Summary

- To overcome the complications associated with the Kubo formula, we developed an alternative scheme for studying thermal transport using the quantum kinetic equation.
- All the currents share a uniform and compact structure:

$$\mathbf{j}^{con}_{e,h} = -i\int \frac{d\varepsilon}{2\pi}\chi_{e,h}(\varepsilon)\left[\hat{\mathbf{v}}(\varepsilon)\hat{G}(\varepsilon)\right]^{<}$$

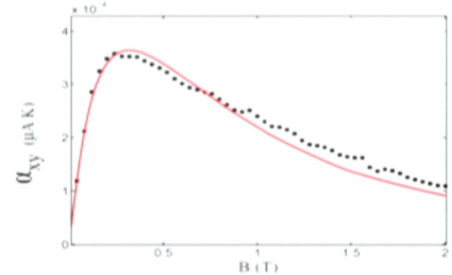
- The Nernst effect provides an excellent opportunity to test the quantum kinetic approach
- The contribution from the fluctuations of the superconducting order parameter to the Nernst effect is dominant and can be observed far away from the transition.
- The magnetization plays an important role in the calculation of the Nernst effect. The magnetization cancels the quantum contributions, thus making the Nernst signal compatible with the third law of thermodynamics.
- The third law of thermodynamics (Nernst theorem) imposes a strong constraint on the magnitude of the Nernst signal not only at low temperature ($T \rightarrow 0$), but also at higher temperatures ($T \gg T_C$). .

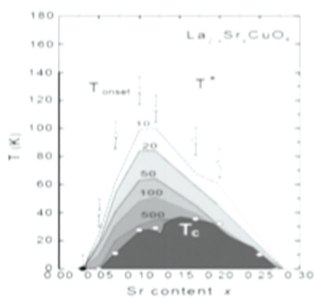
Why Should We Believe the Quantum Kinetic Approach?

a. The Onsager relation naturally emerges in this method.

b. The expression for the Nernst effect at $B \to 0$ and $T \sim T_c$ coincide with the result of the phenemenological calculation using the time dependent Ginzburg Landau equation

c. Agreement with the experiment (no fitting parameters):





Why the Nernst effect in High-Tc superconductors decays so strongly with T ?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