Low-temperature thermoelectric response of correlated electrons: an experimental survey

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Outline

1. On the thermoelectricity of Fermi liquids in the zero-temperature limit

2. Probing quantum criticality

3. Semi-metals across the quantum limit

Heat and charge current in a solid

$$\vec{J}_e = \sigma \vec{E} - \alpha \vec{\nabla} T$$

$$\vec{J}_Q = \beta \vec{E} - \kappa \vec{\nabla} T$$

$$\beta = \alpha T$$

$$\beta = \alpha T$$ *Kelvin relation(1860)*
Onsager relation (1930)

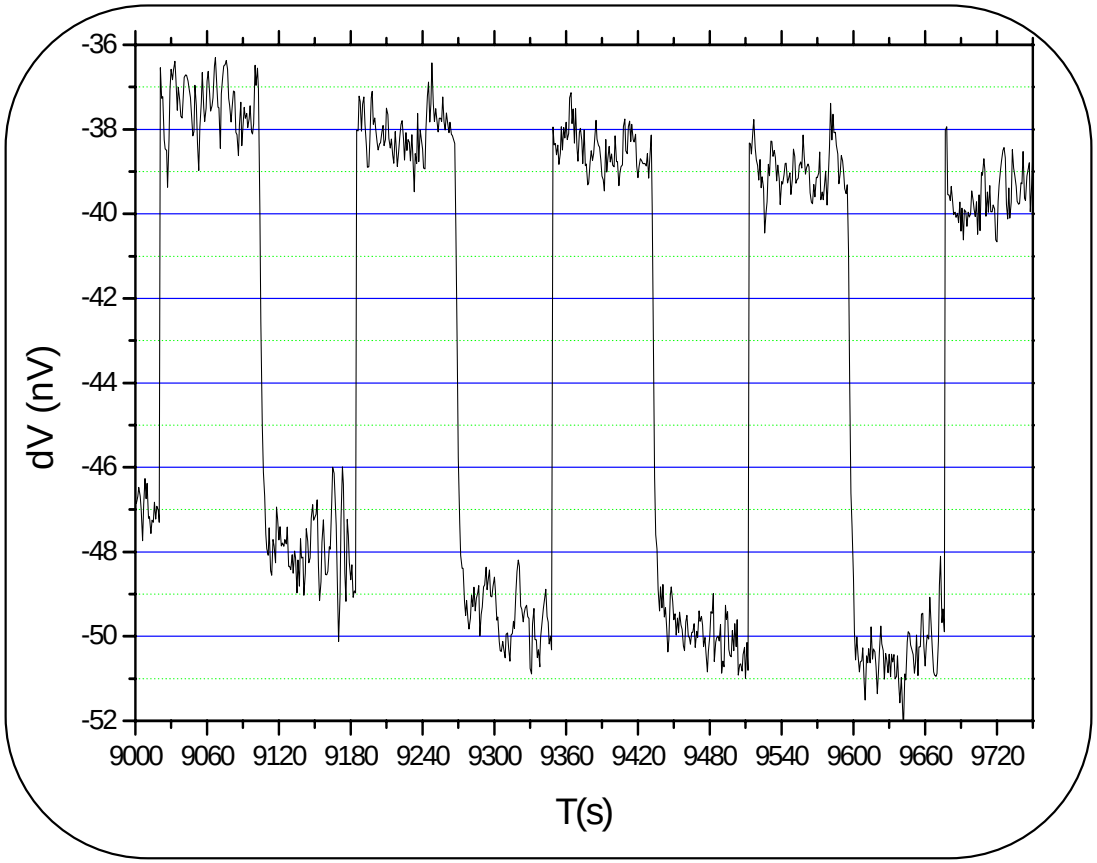
Four vectors

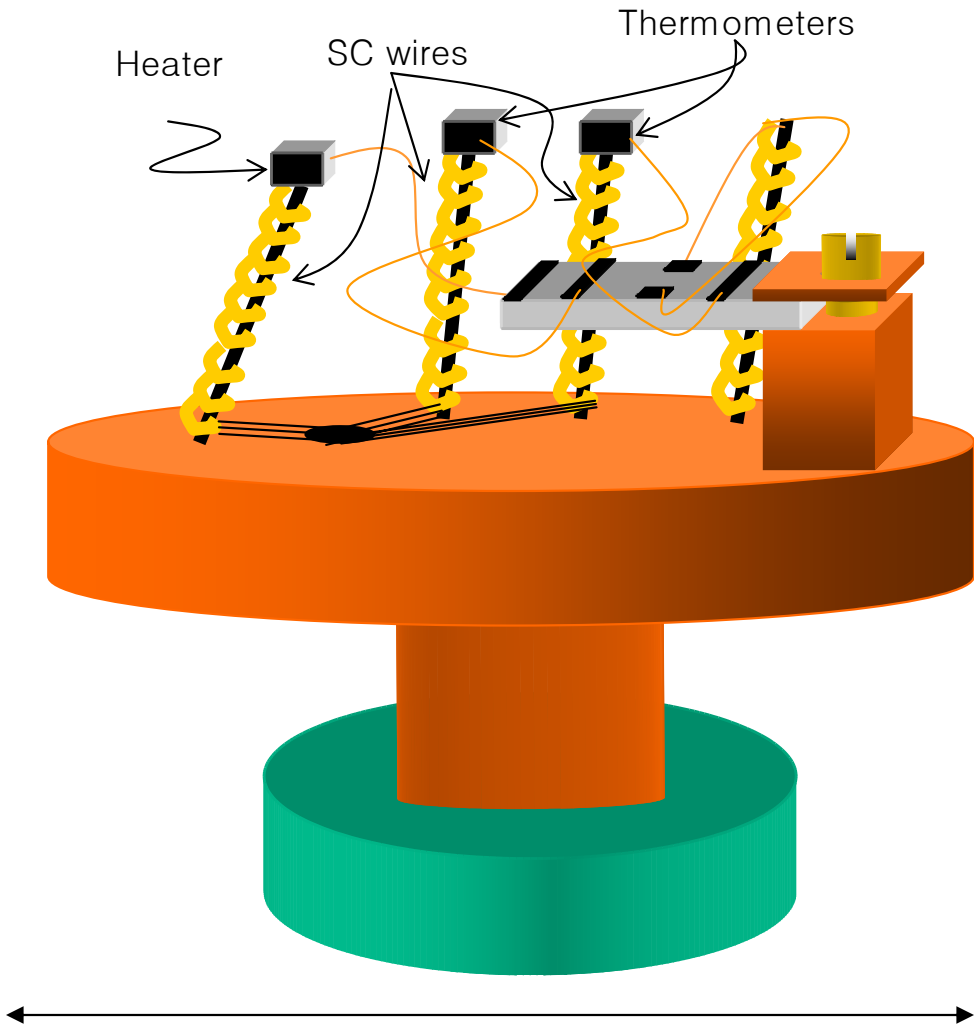
J_e : charge current density
J_Q : heat current density
E : electric field
DT : thermal gradient

Three tensors

σ electric conductivity
κ thermal conductivity
α thermoelectric conductivity

Set-up for monitoring thermal(κ_{xx}, κ_{xy}), thermo-electric (S, N) and electric (σ_{xx}, σ_{xy}) conductivity tensors

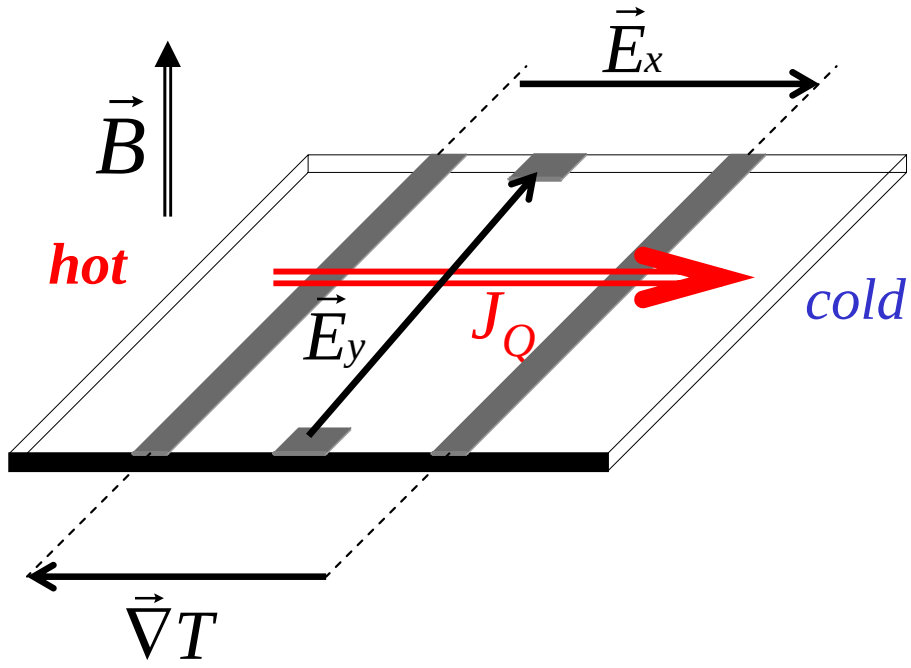




DC voltages of the order of 1 nV resolved!

Definition of thermoelectric coefficients

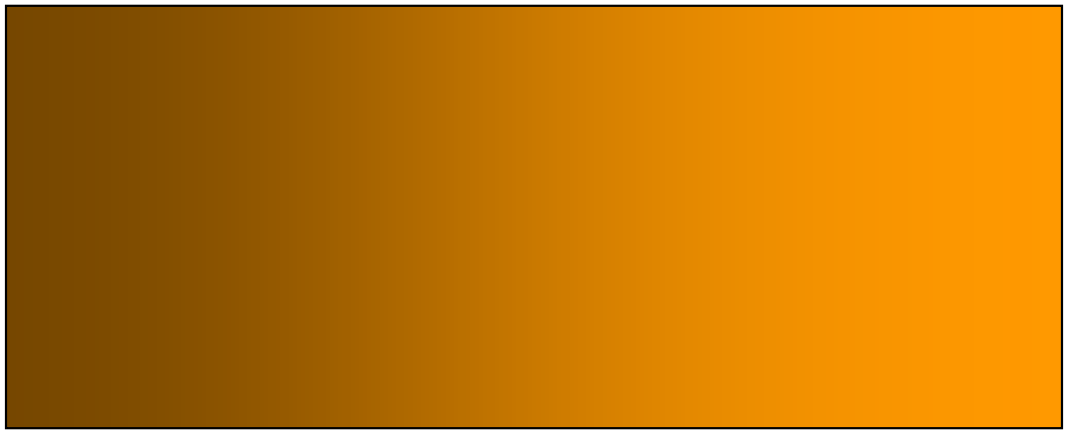
- In presence of a thermal gradient, electrons produce an electric field.
- Seebeck and Nernst effect refer to the longitudinal and the transverse components of this field.



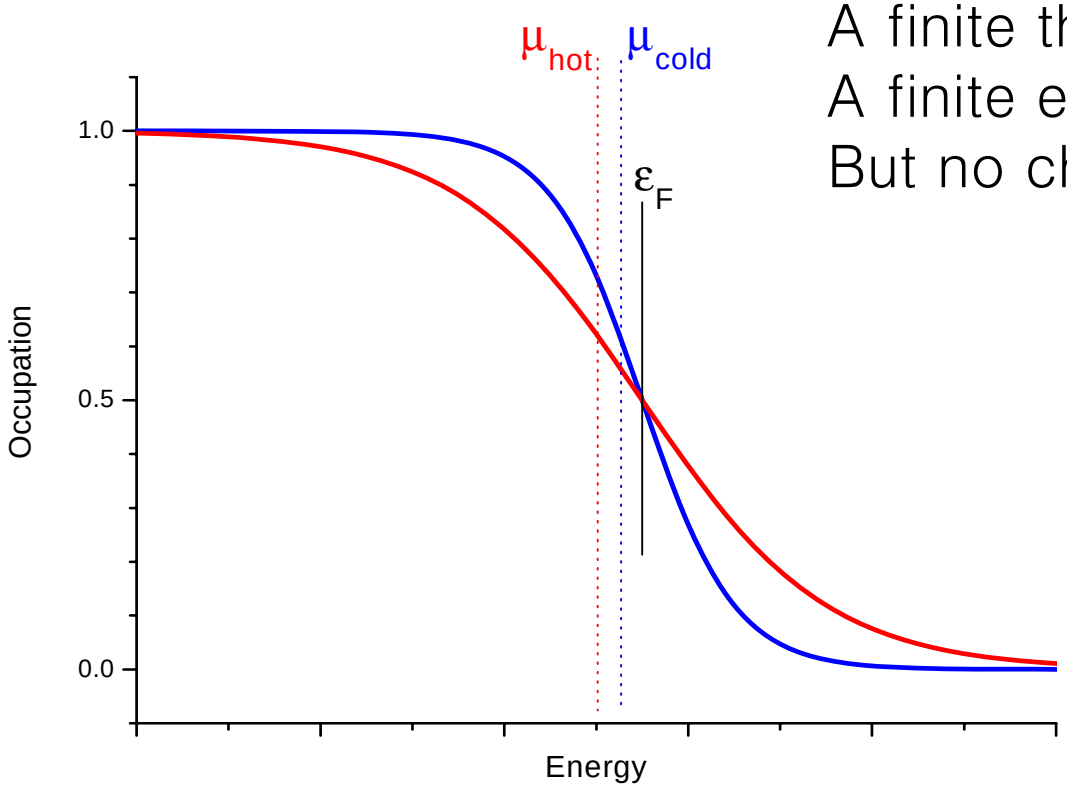
$$S = \frac{-E_x}{\nabla_x T} \qquad N = S_{xy} = \frac{-E_y}{\nabla_x T} \qquad [\nu = \frac{-E_y}{B_z \nabla_x T}]$$

Thermoelectric response

hot



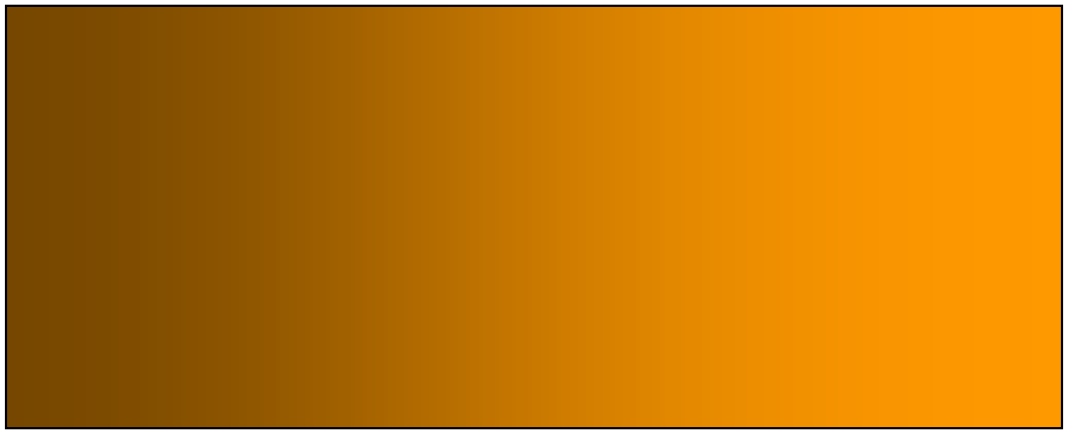
cold



A finite thermal gradient!
A finite electric field!
But no charge current!

Response to a gradient in chemical potential!

Thermoelectric response



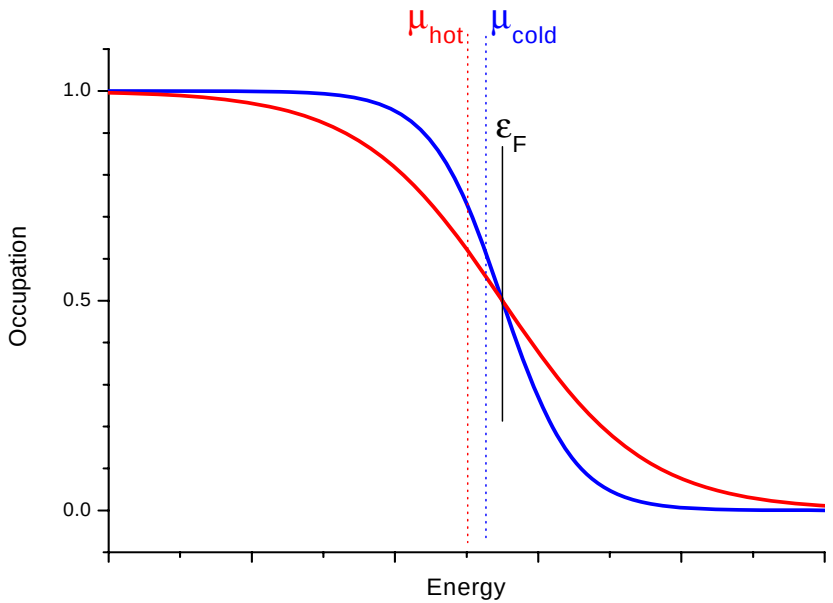
Carriers with a charge, q, and an entropy, S_{ex} will suffer two forces:

- Electric force: $F = E\,q$
- Thermal force: $F = S_{ex}\,\nabla T$

$S = E/\nabla T = S_{ex}/q$

Thermopower measures entropy per [charged] carrier.

$S = (k_B\,T/\,E_F)/\,e$



Heavy Fermi liquids

- Enhanced specific heat $\gamma = \frac{\pi^2}{3} k_B^2 N(\varepsilon_F)$
- Enhanced Pauli Susceptibility $\chi = \mu_B^2 N(\varepsilon_F)$
- Enhanced T^2-resistivity $\rho = \rho_0 + A\ T^2$

$$A \propto N(\varepsilon_F)^2$$

Fermi liquid ratios

The Wilson ratio

$$R_W = \frac{\pi^2}{3}\frac{k^2{}_B}{\mu_B^2}\frac{\chi}{\gamma}$$

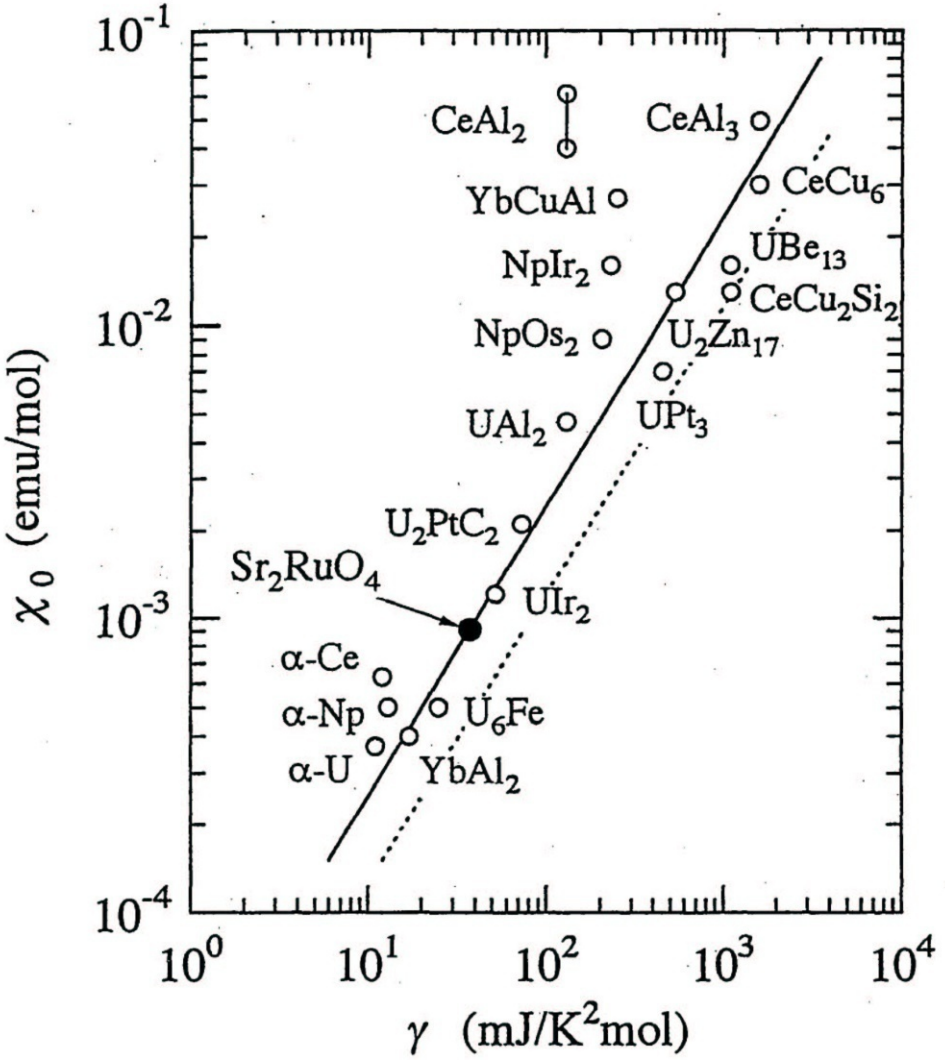
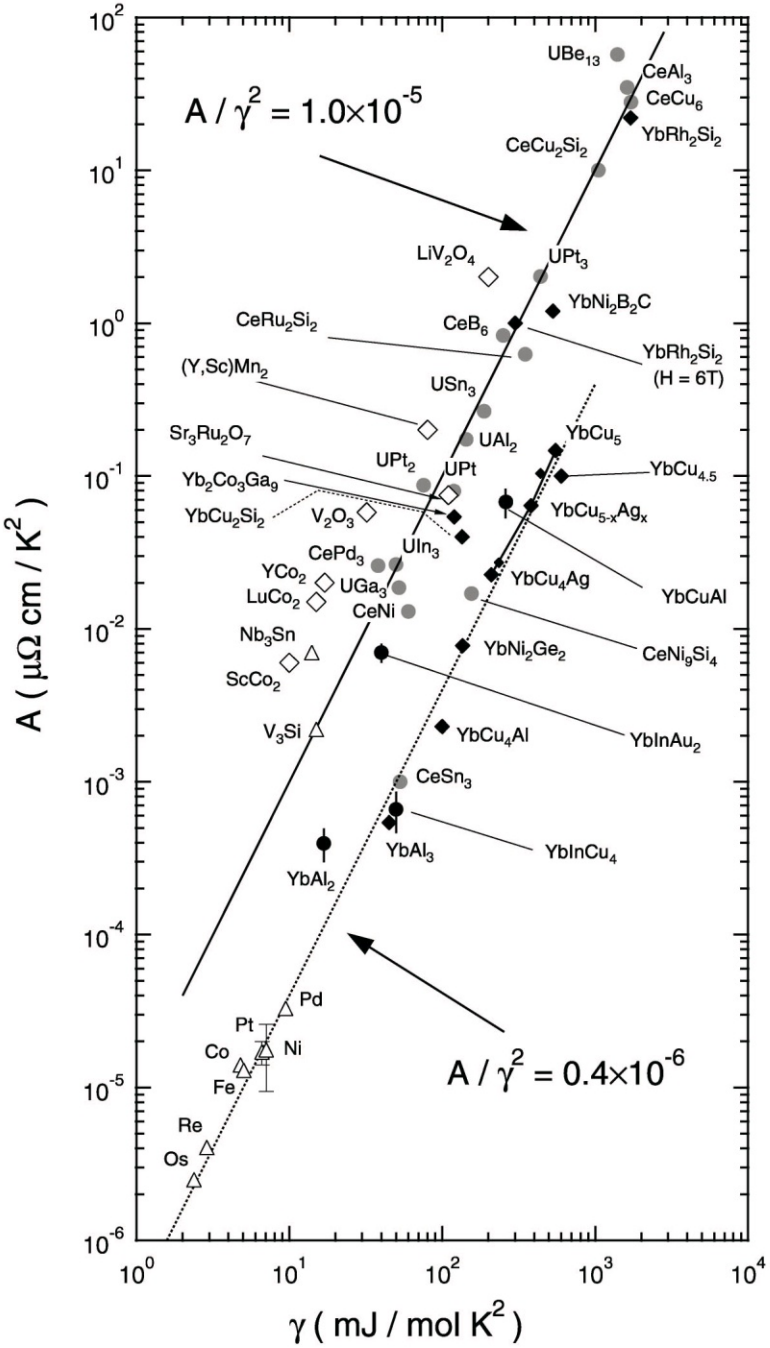


Figure credit
Maeno et al. ‘96

Fermi liquid ratios



The Kadowaki-Woods ratio

$$KW = \frac{A}{\gamma^2}$$

Figure credit
Kontani‘02

Seebeck coefficient of the free electron gas

In the Boltzmann picture thermopower is linked to electric conductivity [the Mott formula]:

$$S = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\left(\frac{\partial \ln \sigma(\epsilon)}{\partial \epsilon}\right)_{\epsilon_F}$$

This yields:

$$S = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\left[\left(\frac{\partial \ln \tau(\epsilon)}{\partial \epsilon}\right)_{\epsilon_F} + \frac{\int d\mathbf{k}\delta(\epsilon_F - \epsilon(\mathbf{k}))\mathbf{M}^{-1}(\mathbf{k})}{\int d\mathbf{k}\delta(\epsilon_F - \epsilon(\mathbf{k}))v(\mathbf{k})v(\mathbf{k})}\right]$$

transport Thermodynamic

For a free electron gas, with $\tau = \tau_0 \varepsilon^{\xi}$, this becomes:

$$S = -\frac{\pi^2}{3}\frac{k_B^2}{e}\frac{T}{\epsilon_F}\left(\frac{3}{2} + \xi\right)$$

Thermopower and specific heat

In a free electron gas (with ξ=0):

$$S = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{N(\epsilon_F)}{n}$$

$$C_{el} = \frac{\pi^2}{3} k_B^2 T N(\epsilon_F)$$

Thermopower is a measure of specific heat per carrier

The dimensionless ratio: $q = N_{Av} e \frac{S}{T\gamma}$

is equal to −1 (+1) for free electrons (holes)

Thermoelectricity in real metals

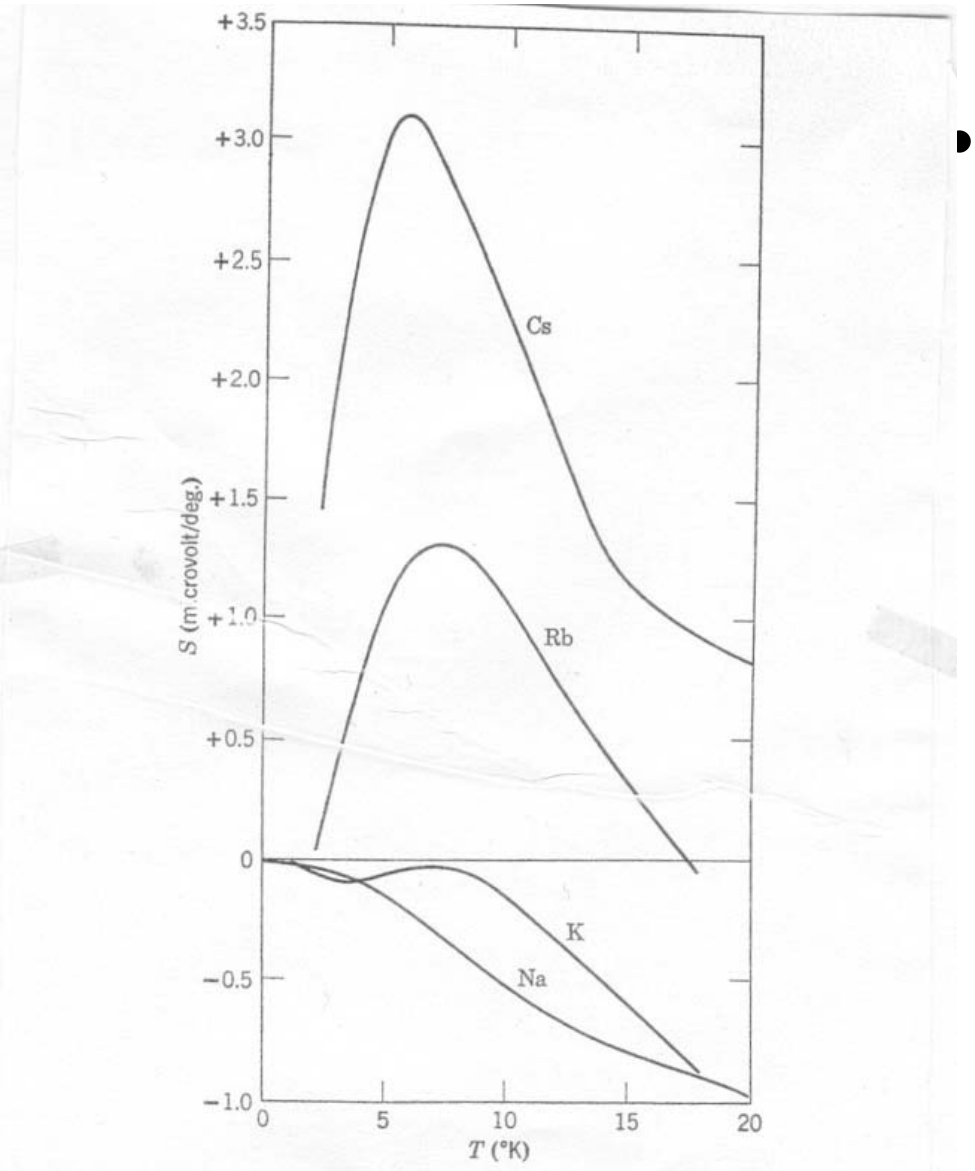
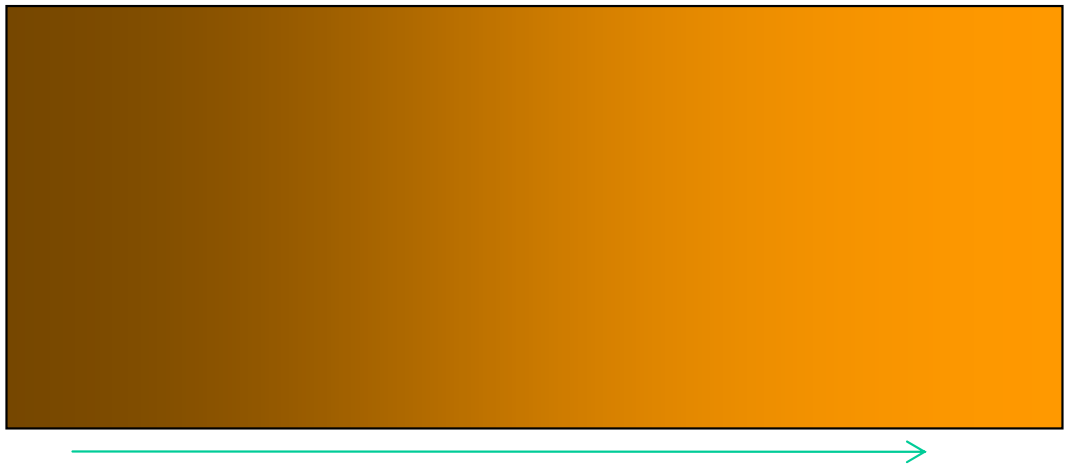


Fig. 28. Absolute thermoelectric power of alkali metals at very low temperatures. The data are taken from MacDonald, Pearson, and Templeton (1958) and, in general, the purest specimen has been selected in each case here. The positive hump in the thermoelectric power due to phonon-drag is particularly striking at the low temperatures in rubidium and caesium and already is making its presence felt in potassium between 5 to 10° K.

- Even in simplest metals, the free-electron-gas picture does not work at finite temperature !

Structure in the thermopower of AlKali metals (MacDonald 1961) !

Phonon Dragg



•If electrons and phonons exchange energy then electrons would be dragged by phonons

Phonon dragg thermopower $S_g = \frac{C_L}{Ne}\alpha$

Lattice specific heat

Carrier density

e^- – ph coupling ($0<\alpha<1$)

Order of magnitude of phonon dragg

$$S_g / S_{diff} = \frac{C_L}{C_e \beta} \alpha$$

$(T/\Theta_D)^3$

Frequency of ph–e⁻ scattering events

T/T_F

e^- per atom

- Expected to vanish in the zero-temperature limit
- Becomes negligible when $C_e >> C_L$

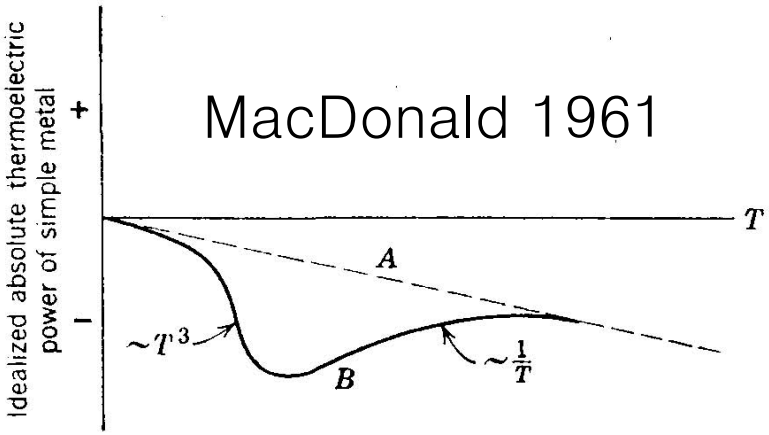


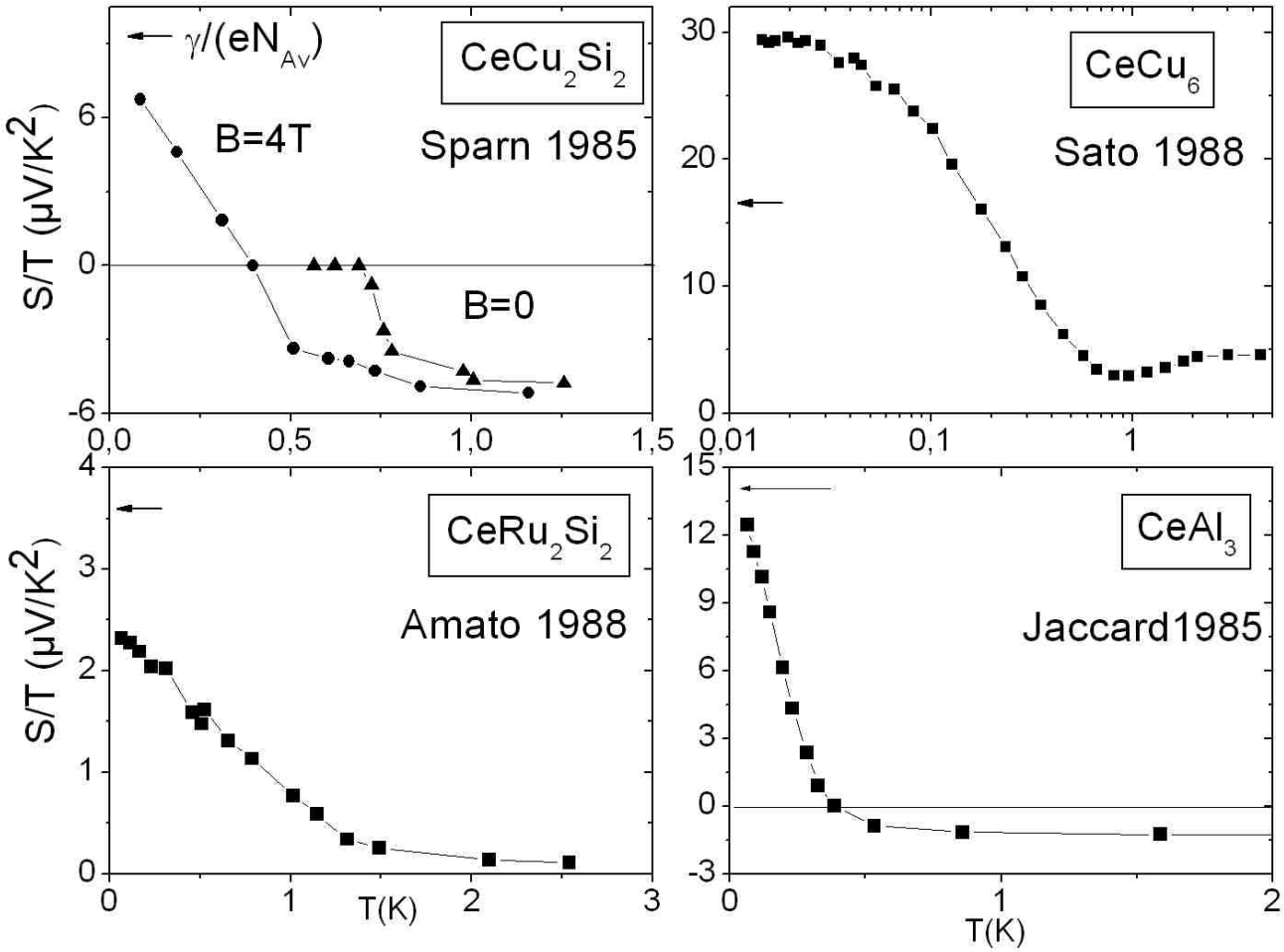
Fig. 8. Sketch of idealized absolute thermoelectric power of a simple quasi-free electron metal.

A: Electron diffusion component of thermoelectric power approximately proportional to T.

B: Phonon-drag component with magnitude increasing as T^3 at very low temperatures ($T \ll \theta$), and decaying as $1/T$ at "high" temperatures ($T \gtrsim \theta$).

Heavy electrons in the T=0 limit

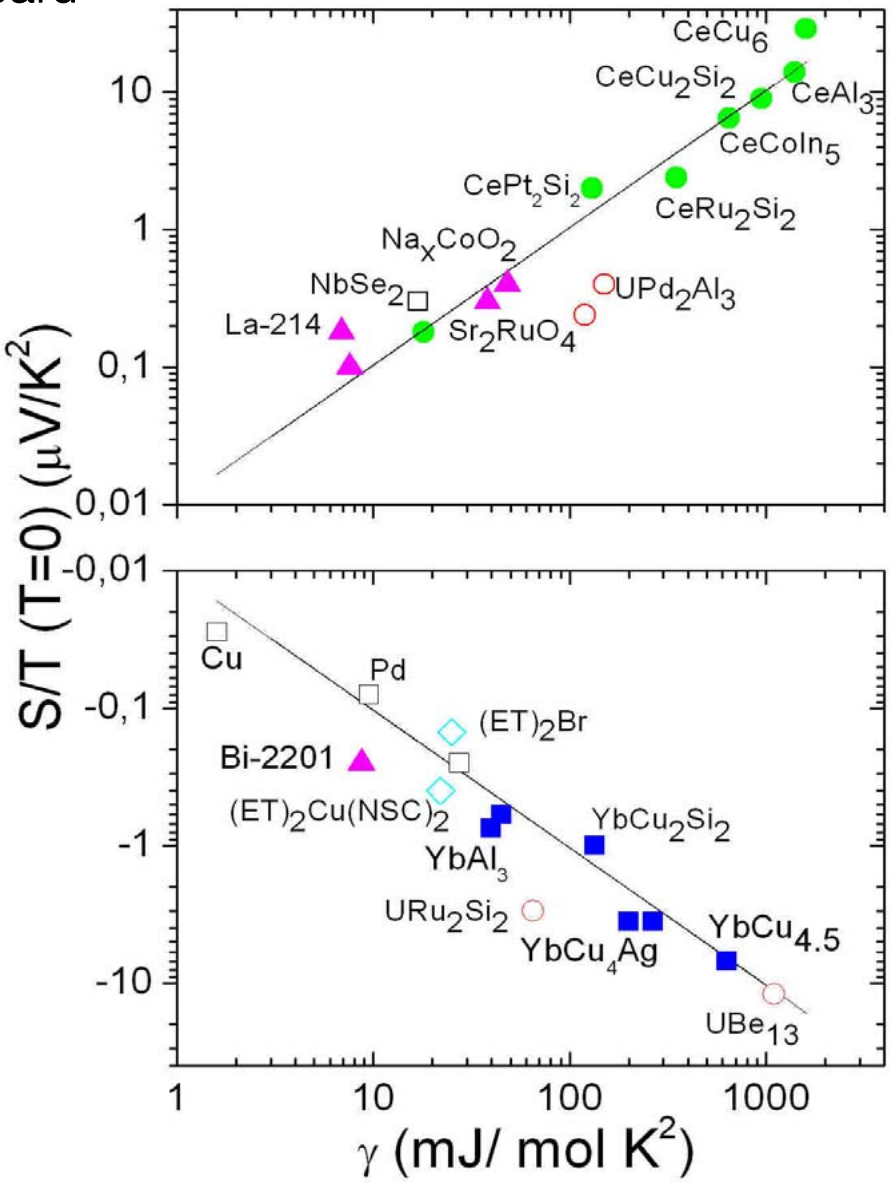
Replotting two-decades-old data!



Extrapolated to T=0, the extracted S/T yields a q close to unity!

Another plot linking two distinct signatures of electron correlation

Behnia, Flouquet, Jaccard
JPCM '04



Compound	S/T (μ V / K^2)	Remarks	γ (mJ /mol K^2)	q
$CeCu_2Si_2(B = 4T)$	9[30]	polycrystal	950[52]	0.9
$CeCu_6$	29[33]	along [010]	1600[53]	1.7
$CeAl_3$	14[32]	polycrystal	1400[32]	1..0
$CeRu_2Si_2$	2.4[31]	in-plane	350[54]	0.7
$CeCoIn_5$ (B=6T)	6[55]	in-plane	650[56]	0..9
$CePt_2Si_2$	2[57]	along [110]	130[58]	1.5
$CeSn_3$	0.18[59]	polycrystal	18[60]	1.0
CeNiSn	50[61]	polycrystal	45[62]	107
$YbCu_{4.5}$	-7[63]	polycrystal	635[64]	-1.1
YbCuAl	-3.6[65]	polycrystal	267[66]	-1.3
$YbCu_4Ag$	-3.6[67]	polycrystal	200[68]	-1.7
$YbCu_2Si_2$	-1[20, 69]	polycrystal	135[70]	-0.7
$YbAl_3$	-0.6[20]	polycrystal	45[71]	-1.3
$YbInAu_2$	-0.75[69]	polycrystal	40[72]	-1.8
UPt_3	unknown	none observed[35]	430[35]	–
UBe_{13}(B=7.5T)	-12[36]	polycrystal	1100[73]	-1.1
UNi_2Al_3	0.24[37]	polycrystal	120[74]	0.2
UPd_2Al_3	0.4[37]	S $\perp c$	150[75]	0.3
URu_2Si_2	-3[38]	S $\perp c$	65[76]	-4.5
κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br	-0.4[41]	in-plane	22[77]	-1.7
κ-(BEDT-TTF)$_2$Cu(NSC)$_2$	-0.15[42]	in-plane	25[78]	-0.6
$(TMTSF)_2ClO_4$	unknown	No report found	11[79]	–
Sr_2RuO_4	0.3[44]	in-plane	38[4]	0..8
$SrRuO_3$	unknown	No report found	30[80]	–
$Sr_3Ru_2O_7$	unknown	No report found	38[81]	–
$SrRhO_3$	0.03[82]	polycrystal	7.6[83]	1..3
Na_xCoO_2	0.4[45]	in-plane	48[46]	0..8
$La_{1.7}Sr_{0.3}CuO_4$	0.18[47]	ceramic	6.9[6]	2.5
$Bi_2Sr_2CuO_{6+\delta}$	-0.25[48]	ceramic	8.7[84]	-2.8
$NbSe_2$	0.3[51]	in-plane	17[85]	1.7
Pd	-0.08[16]	polycrystal	9.5[86]	-0.8
Cu	-0.028[50]	along [231]	1.6[11]	-1.7
constantan (%43Ni-%57Cu)	-0.25[14]	wire	27.4[87]	-0.9

Table 1. Reported magnitudes of linear thermopower and specific heat for a number of metals. The significance of the coefficient $q = \frac{\mathcal{S}}{T}\frac{N_{Ave}}{\gamma}$ is discussed in the text.

Data since 2004

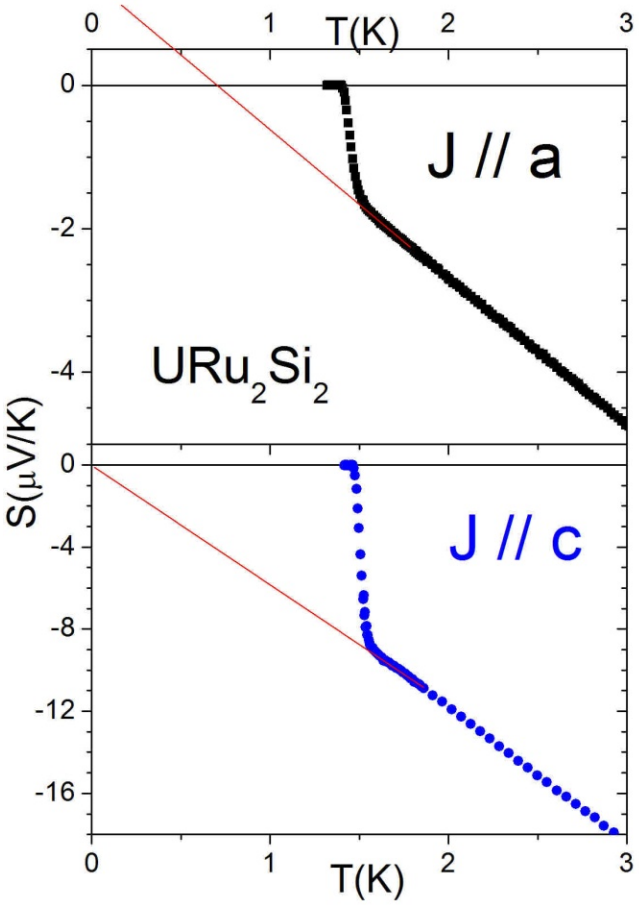
	γ	S/T	q
UPt_3	430	2.5	0.6
$NpPd_5Al_2$	200	-1.3	-0.65
$CeNi_2Al_3$	30	0.27	0.9
$PrFe_4As_{12}$	340	-1	0.3
$YbRh_2Si_2$	750	-6.7	-0.8
FeTe	34	-0.3	-0.9
MgB_2	3	0.04	-1.3
$SrRu0_3$	30	0.24	0.8
$SrRh_2O_4$	10	0.22	2.2

In semi-metals q is large

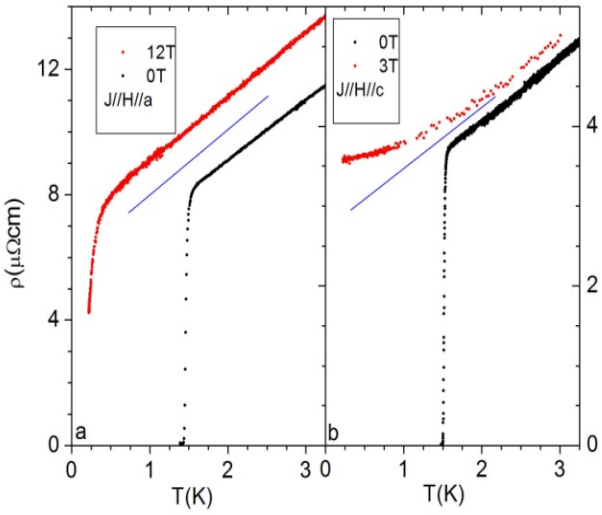
- URu_2Si_2 (q=-11)
- $PrFe_4P_{12}$ (q=-58)
- $PrRu_4P_{12}$ (q=-43)
- CeNiSn (q=107)
- $Bi_{0.96}Sb_{0.04}$ ($q=10^4$)

In these systems the FS occupies a small fraction (~1/2q) of the BZ.

In URu_2Si_2 hidden orde opens a gap and destroys nine-tenth of the FS



FL behavior for J// a does not set down to the lowest T neither in S nor in ρ
(Zhu et al. PRB '09)



Theory on correlation between S and γ

Miyake & Kohno, JPSJ (2005)

$$\lim_{\varepsilon \to \mu} \frac{\partial \ln \tau_{imp}(\epsilon)}{\partial \epsilon} \propto \left(\frac{\partial \ln N^*(\epsilon)}{\partial \epsilon}\right)_{\varepsilon=\mu} \sim \frac{1}{\widetilde{\epsilon}_F}$$

In both unitary and Born limits, q ~ ±1

Zlatic, Monnier, Freericks & Becker, PRB 2007

(Single-impurity Anderson model)

Paul & Kotliar, PRB 2001
Near a QCP both expected to diverge logarithmically

Haule and Kotliar, CorrelatedThermoelectricity workshop (2008)
DMFT

Measuring the Fermi energy of a 3D electronic system

- Resistivity: $A = \frac{h}{e^2} a \frac{1}{\varepsilon_F^2}$

A material-depenedent length scale (model-dependent combination of interatomic and interelectronic distances)

- Specfic heat $\gamma = \frac{\pi^2}{3} k_B^2 n \frac{1}{\varepsilon_F}$

Carrier density

- Thermopower $\frac{S}{T} = \frac{\pi^2}{3} \frac{k_B}{e} \frac{1}{\varepsilon_F}$

Near a QCP, thermoelectric response becomes large !

Nernst and Seebeck Coefficients of the Cuprate Superconductor $YBa_2Cu_3O_{6.67}$: A Study of Fermi Surface Reconstruction

J. Chang,[1] R. Daou,[1,*] Cyril Proust,[2,3] David LeBoeuf,[1] Nicolas Doiron-Leyraud,[1] Francis Laliberté,[1] B. Pin B. J. Ramshaw,[4] Ruixing Liang,[4,3] D. A. Bonn,[4,3] W. N. Hardy,[4,3] H. Takagi,[5] A. B. Antunes,[6] I. Sheikin,[6] K. Behnia,[7] and Louis Taillefer[1,3,†]

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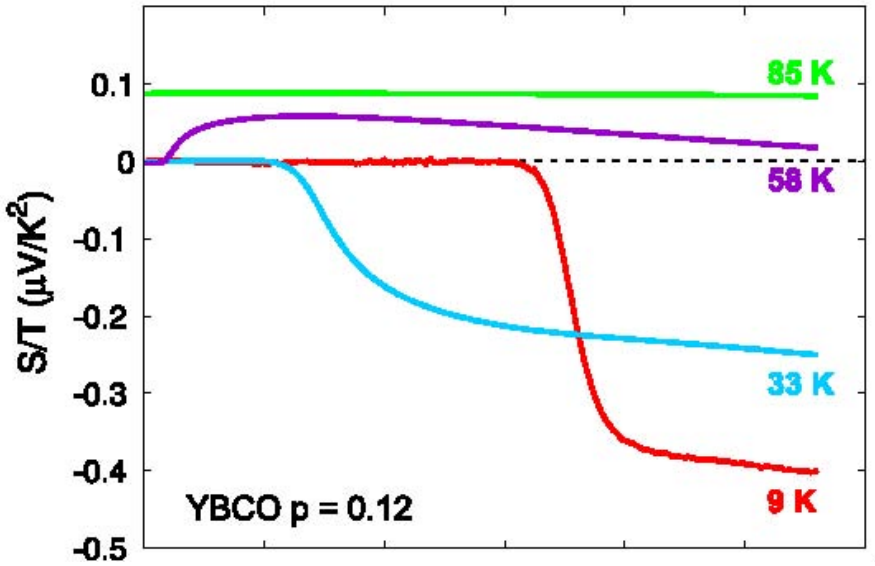
The Seebeck and Nernst coefficients S and ν of the cuprate superconductor $YBa_2Cu_3O_y$ (YBCO) were measured in a single crystal with doping $p = 0.12$ in magnetic fields up to $H = 28$ T. Down to $T = 9$ K, ν becomes independent of field by $H \simeq 30$ T, showing that superconducting fluctuations have become negligible. In this field-induced normal state, S/T and ν/T are both large and negative in the $T \to 0$ limit, with the magnitude and sign of S/T consistent with the small electronlike Fermi surface pocket detected previously by quantum oscillations and the Hall effect. The change of sign in $S(T)$ at $T \simeq 50$ K is remarkably similar to that observed in $La_{2-x}Ba_xCuO_4$, $La_{2-x-y}Nd_ySr_xCuO_4$, and $La_{2-x-y}Eu_ySr_xCuO_4$, where it is clearly associated with the onset of stripe order. We propose that a similar density-wave mechanism causes the Fermi surface reconstruction in YBCO.



The large S/T implies a small Fermi energy within a factor of 2 of the pocket detected by quantum oscillations!

The expected γ_0 given the size of the FS

Loram et al. JPCS, '01

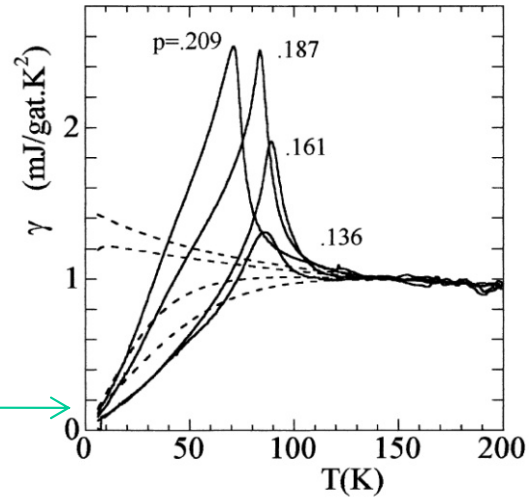
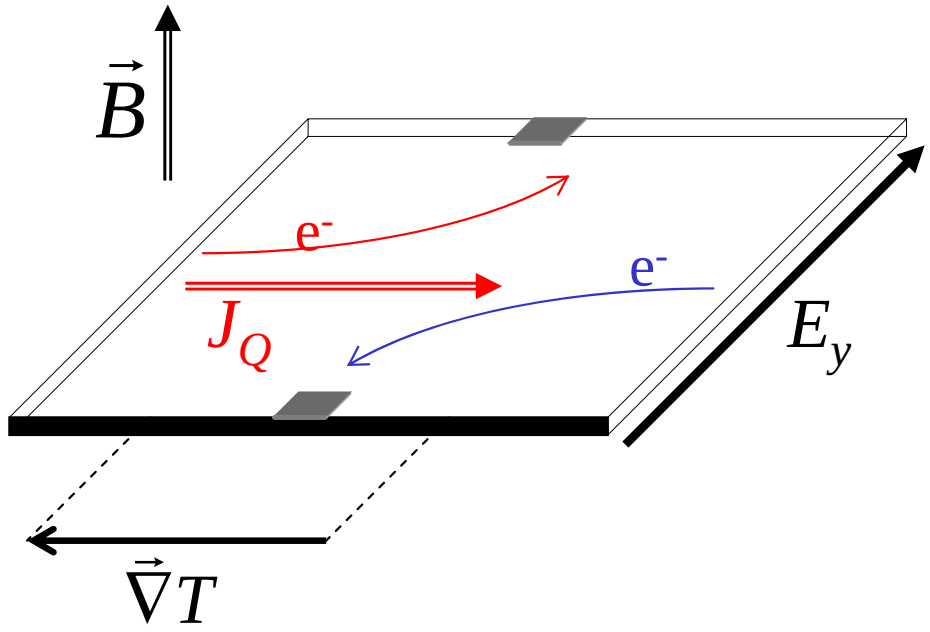


Fig. 11. Broken lines show γ_n for $T < T_c$ deduced from the entropy above T_c. The condition $S_n(T_c) = S_s(T_c)$ can only be satisfied if there is a pseudogap in γ_n for $p < 0.187$ and no pseudogap for $p > 0.187$.

Nernst effect in a single-band metal

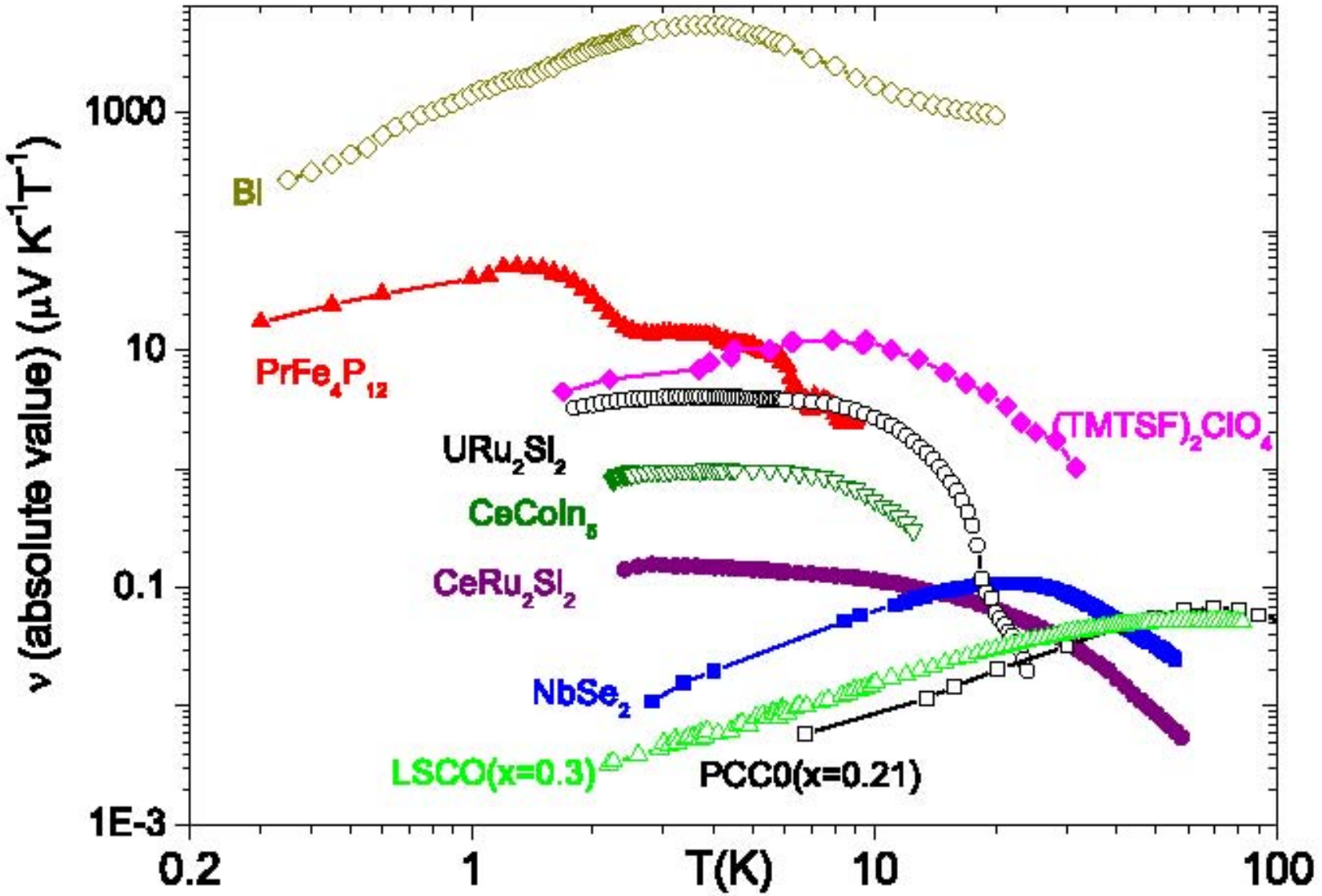
Absence of charge current leads to a counterflow of hot and cold electrons:

$$J_Q \neq 0 \; ; \; J_e = 0 \; ; \; E_y = 0$$

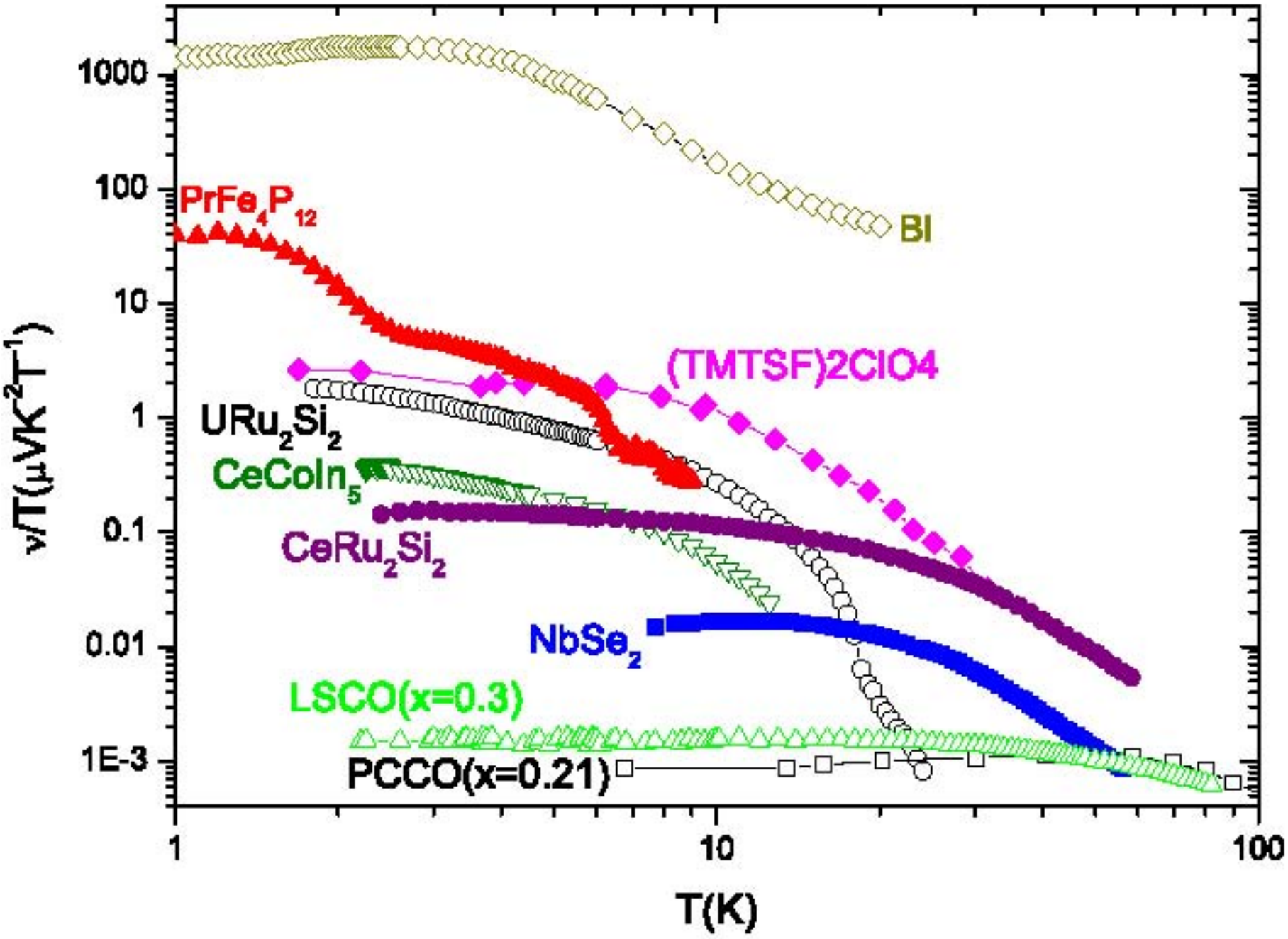


In an ideally simple metal, the Nernst effect vanishes!
(« Sondheimer cancellation », 1948)

Nernst coefficient in remarkable metals!



A large diffusive component in the zero-temperature limit!



Close-up on Sondheimer cancellation

$$\vec{J}_e = \sigma \vec{E} - \alpha \vec{\nabla} T$$

$$\vec{J}_Q = \alpha T \vec{E} - \kappa \vec{\nabla} T$$

$J_e=0$

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

Boltzmann picture: $\overline{\alpha} = \frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \overline{\sigma}}{\partial \epsilon}\Big|_{\varepsilon_F} \longrightarrow N = \frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \Theta_H}{\partial \epsilon}\Big|_{\varepsilon_F}$

If the Hall angle, Θ_H, does not depend on the position of the Fermi level, then the Nernst signal vanishes!

Roughly, the Nernst coefficient tracks $\omega_c\tau$ / E_F…

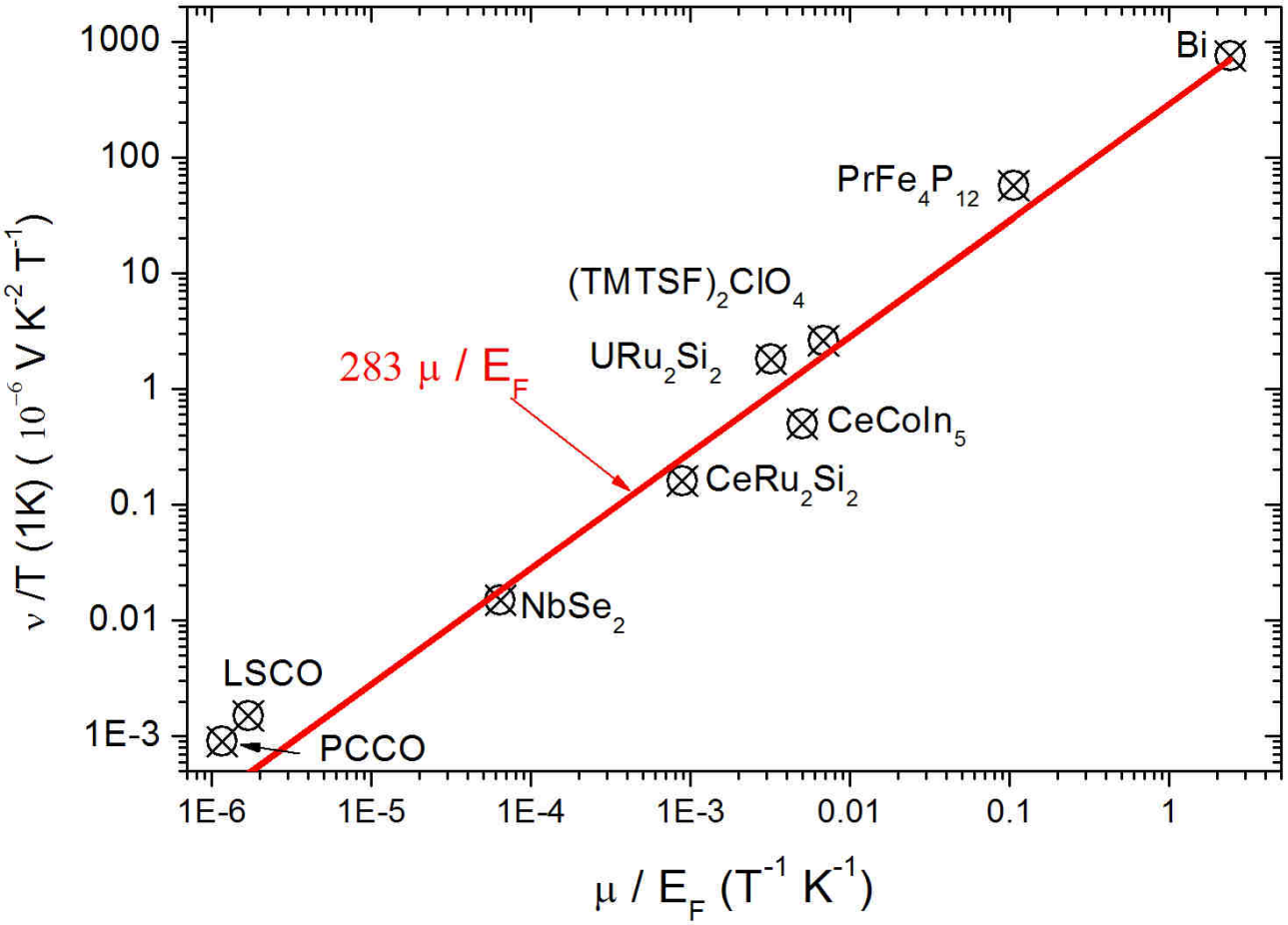
$$N = \frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \Theta_H}{\partial \epsilon}\Big|_{\epsilon_F}$$

$N \sim \pi^2/3\ k^2_B T/e\ \omega_c\tau\ /\ E_F$

Table 1. The magnitude of the Nernst coefficient divided by temperature at low temperature, together with estimations of the electronic mobility and the Fermi energy in various metals. The fourth column yields the expected magnitude of ν/T according to equation (10).

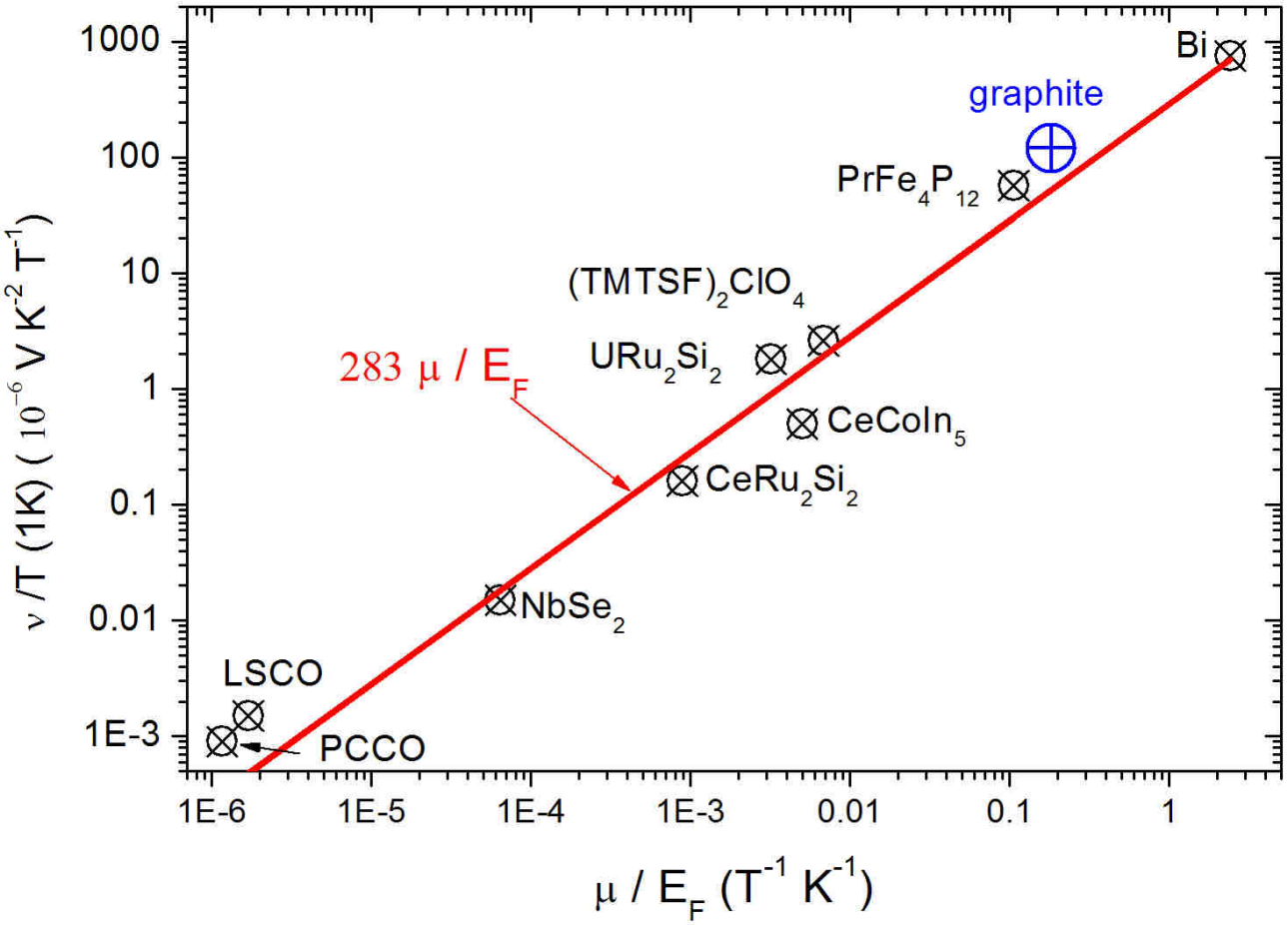
System	ν/T (μV K^{-2} T^{-1})	μ (T^{-1})	ϵ_F (K^{-1})	$\frac{\pi^2}{3}\frac{k_B}{e}\frac{\mu}{\epsilon_F}$ (μV K^{-2} T^{-1})
Bi	750	420	130	914
$CeRu_2Si_2$	0.16	0.2	180	0.25
$CeCoIn_5$	0.5	0.3	60	1.4
URu_2Si_2	1.8	0.08	25	0.9
$PrFe_4P_{12}$	57	0.85	8	30
$(TMTSF)_2ClO_4$	2.6	0.75	110	1.9
$La_{1.7}Sr_{0.3}CuO_4$	0.0015	0.01	5900	4.8×10^{-4}
$Pr_{1.79}Ce_{0.21}CuO_4$	9×10^{-4}	0.005	4300	3.3×10^{-4}
$NbSe_2$	0.015	0.09	1400	0.018

… and becomes large in clean semi-metals!



	Bismuth	URu_2Si_2	$PrFe_4P_{12}$
n (per f.u.)	10^{-5}	$3\ 10^{-2}$	$2\ 10^{-3}$

… and becomes large in clean semi-metals!

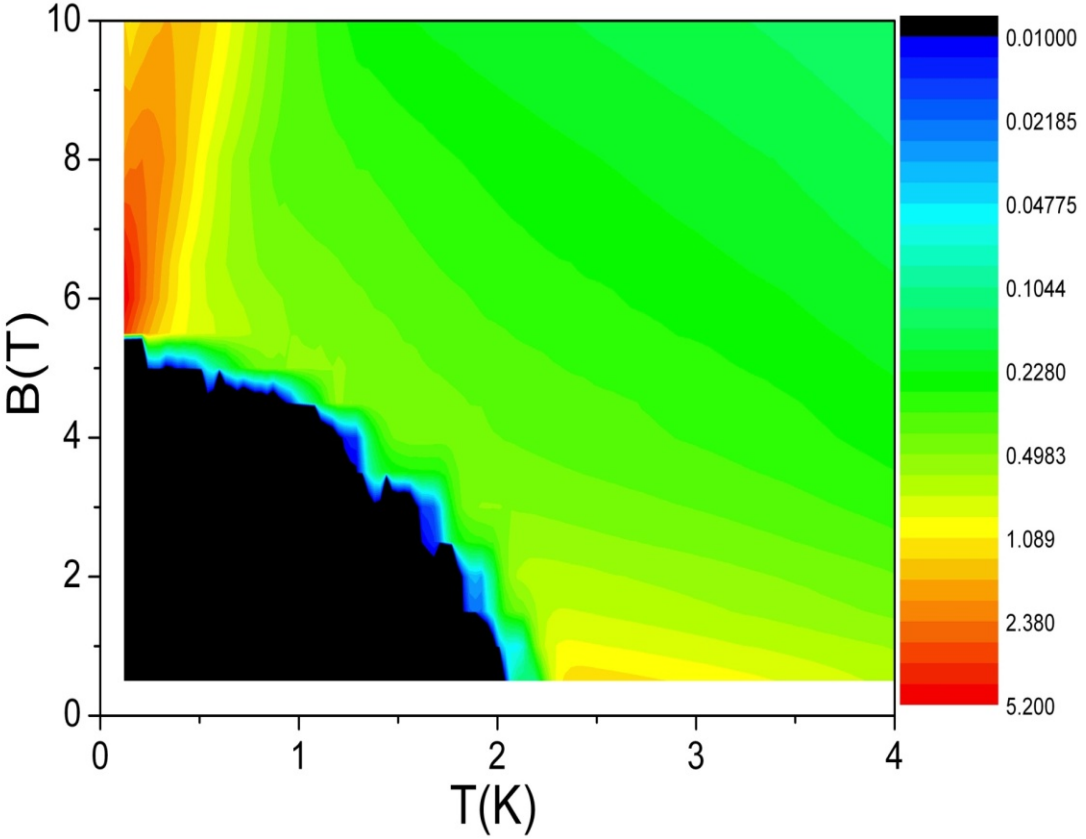


	Bismuth	URu_2Si_2	$PrFe_4P_{12}$
n (per f.u.)	10^{-5}	$3\ 10^{-2}$	$2\ 10^{-3}$

Nernst effect as a probe of quantum criticality

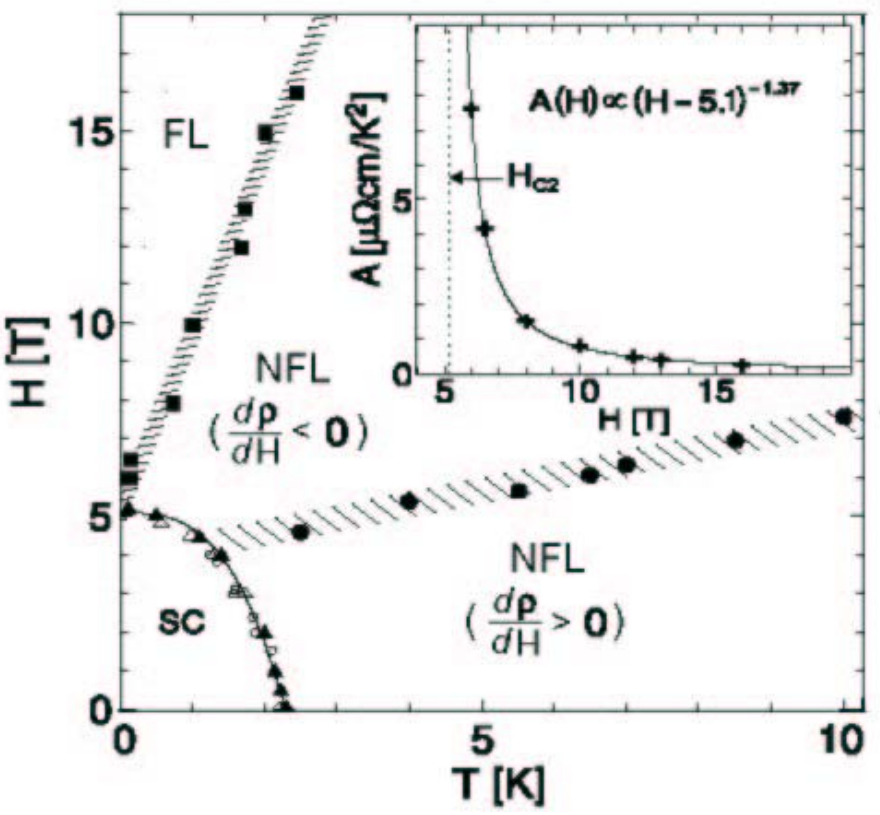
The case of $CeCoIn_5$

Izawa et al. 2007



logarithmic color plot of ν/T

Paglione *et al. ,2003,* Bianchi *et al. 2003*



Nernst effect directly reveals the quantum critical point!

Nernst effect in semi-metals across the quantum limit

Semi-metals

J.–P. Issi, Aust. J. Phys. (1979)

Bismuth crystal structure
Credit: Liu & Allen, PRB '95

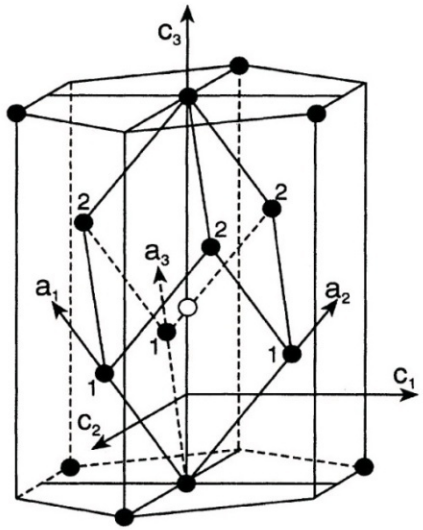
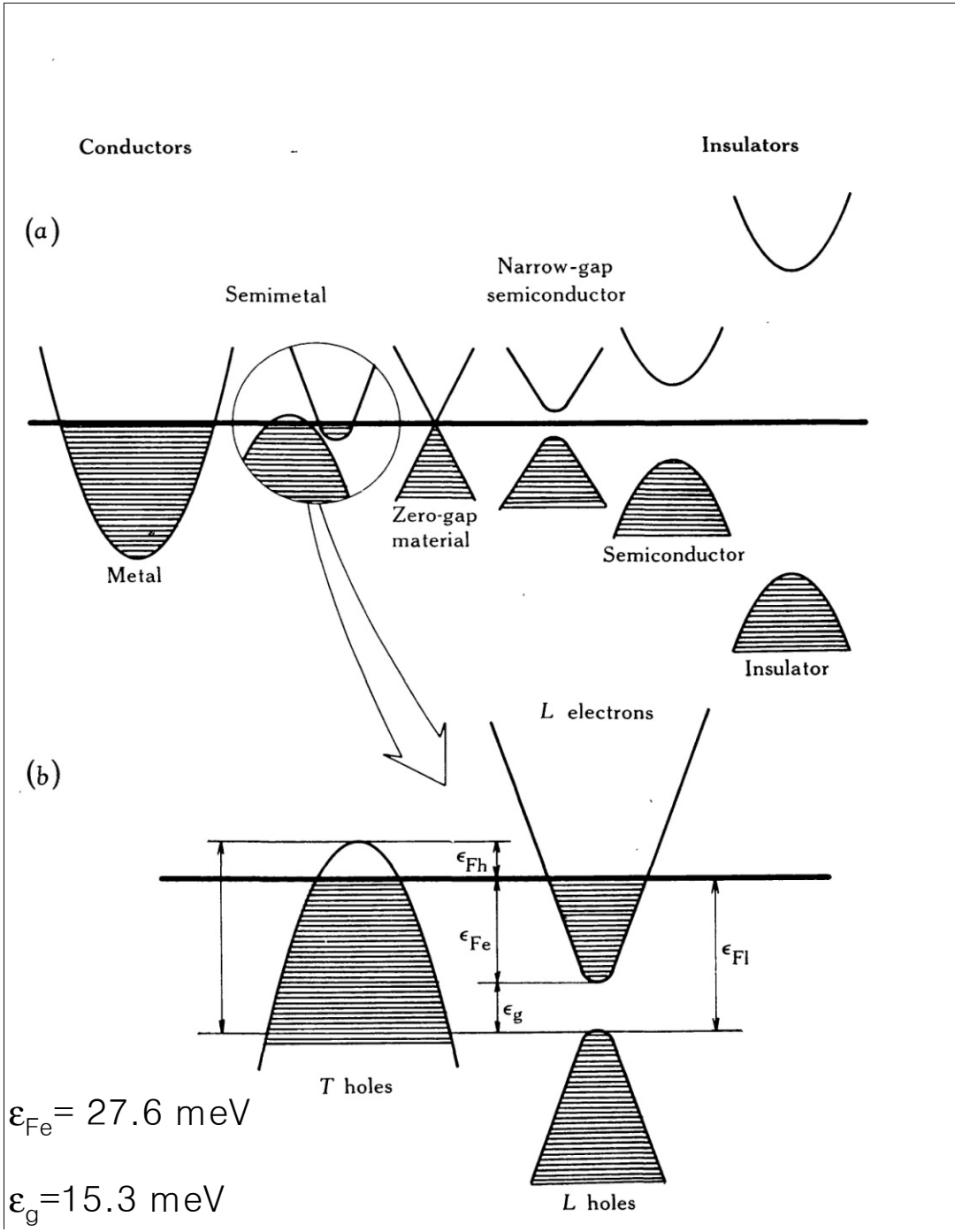


FIG. 1. Crystal structure of Sb and Bi, showing first and second neighbors (labeled 1 and 2) to the central atom represented by an open circle. The bisectrix (C_1), binary (C_2) and trigonal (C_3) axes, and primitive translation vectors (a_1, a_2, a_3) are also shown.

Cubic bismuth would have been a simple metal!



ε_{Fe}= 27.6 meV

ε_g=15.3 meV

Small fermi surfaces

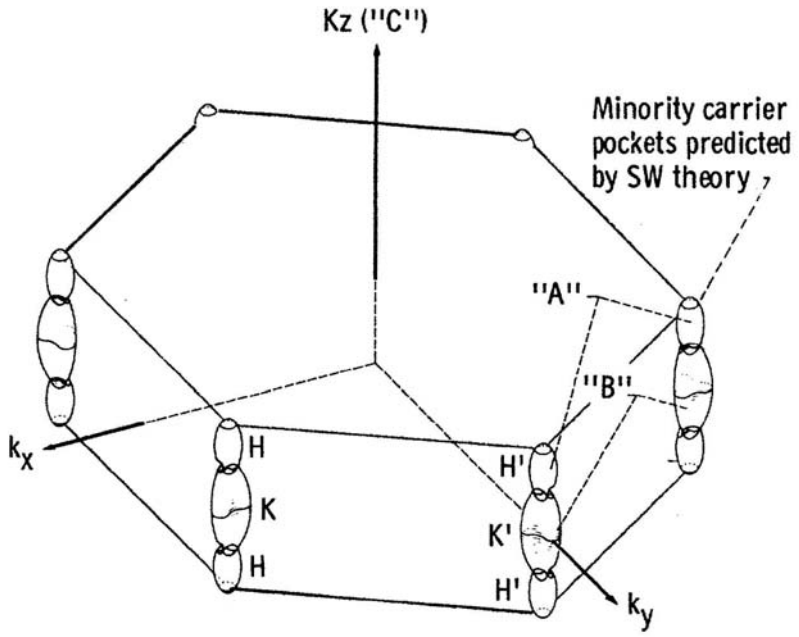
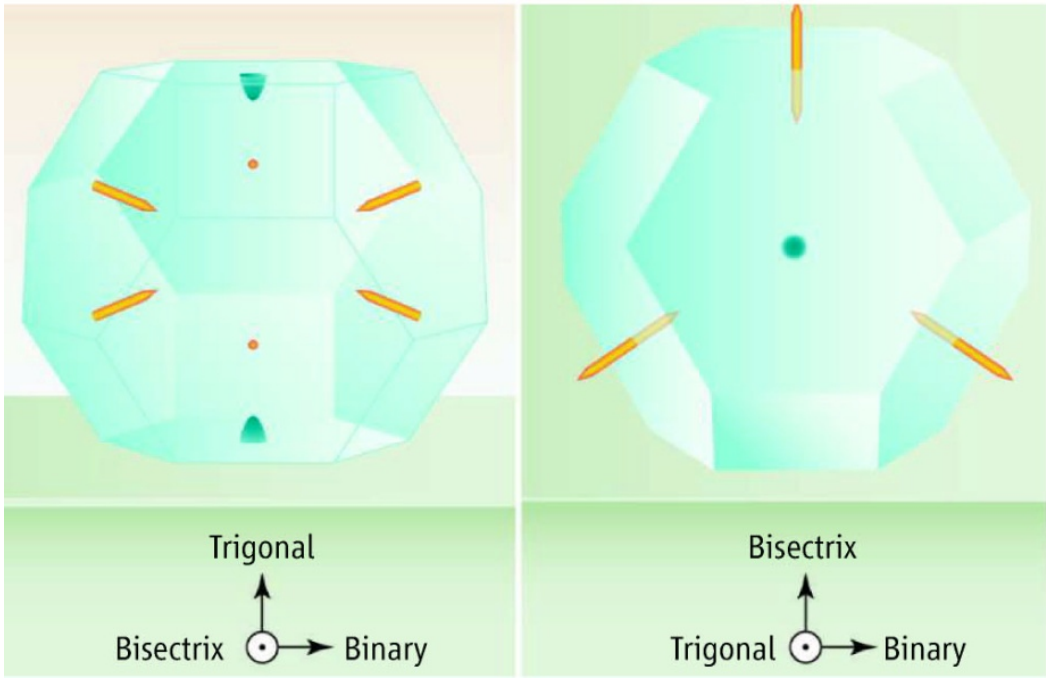


FIG. 4. Brillouin zone of graphite. The Fermi surfaces (located along H, K, H) are labeled A and B.

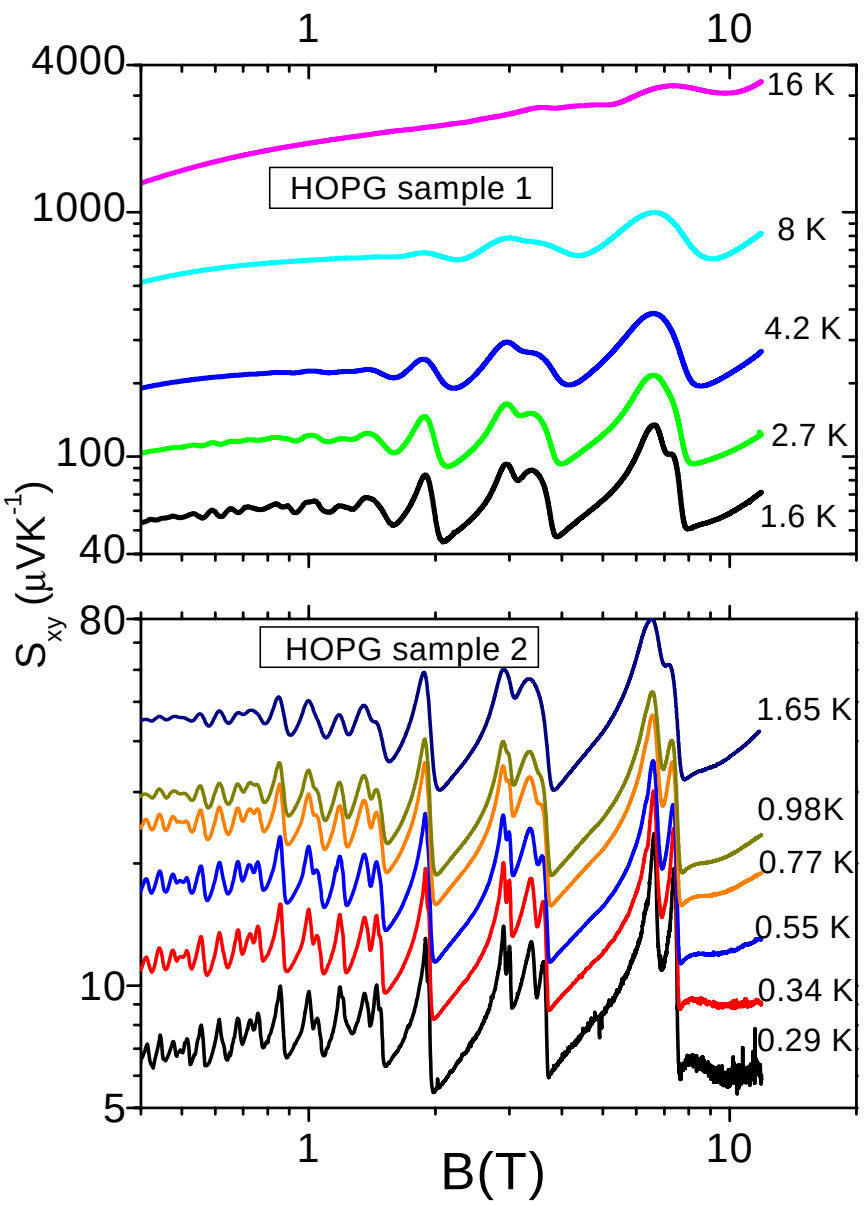
graphite



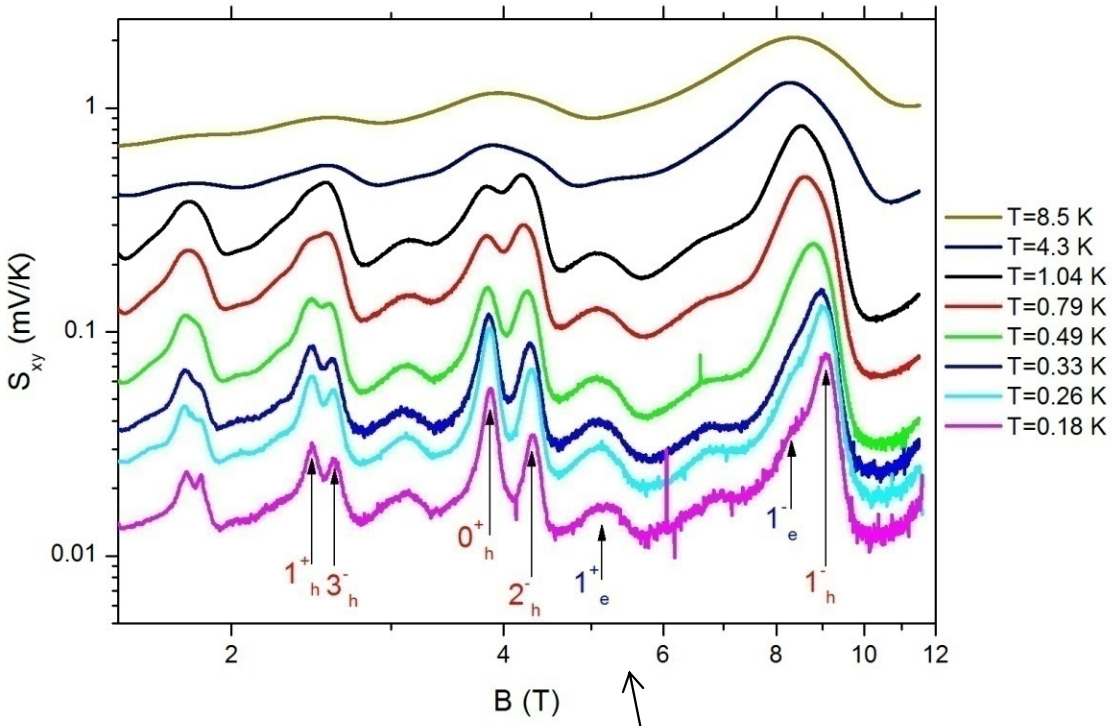
bismuth

Giant quantum oscillations in bismuth and graphite

Zhu et al., Nature Physics '09



KB, M.A-Méasson & Y. Kopelevitch, PRL 2007



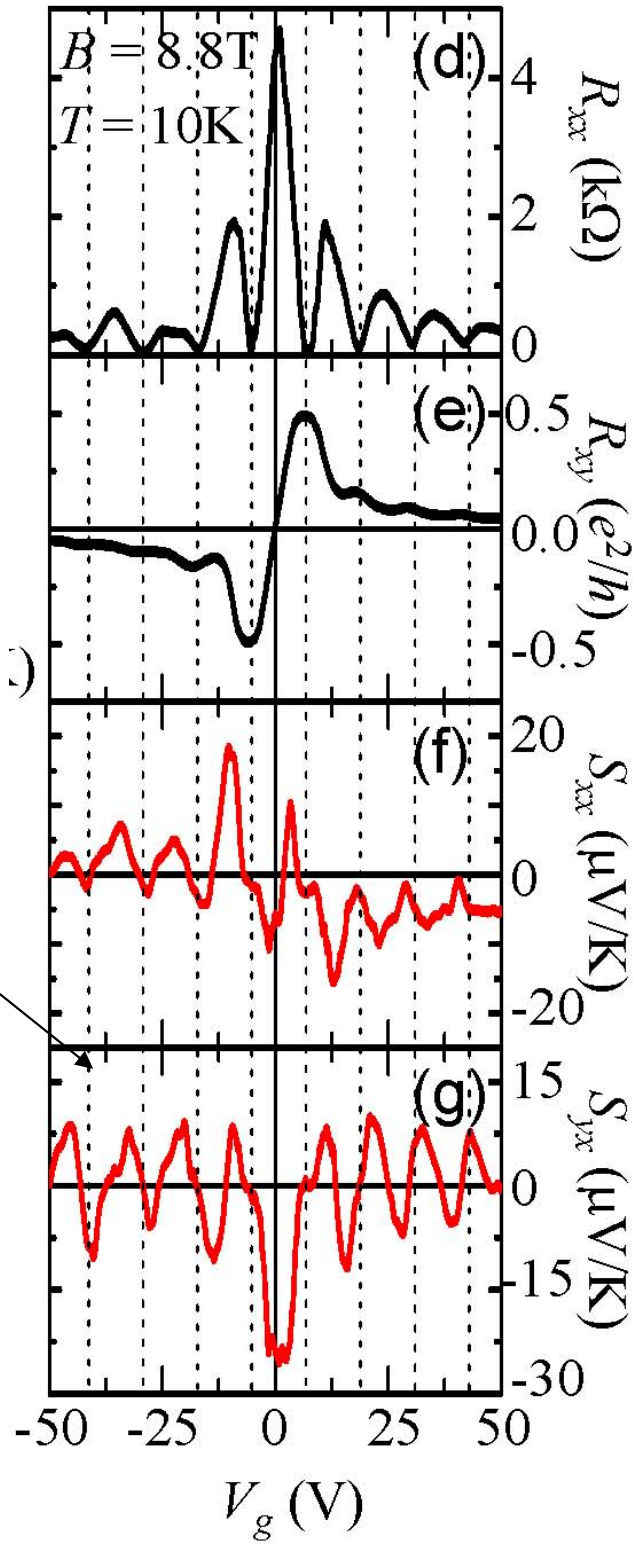
Bismuth B// trigonal

Graphite B//c

Quantum oscillations in Graphene

Zuev, Chang & Kim, PRL'09

When a Landau level meets the Fermi level, S_{xy} vanishes!



See also:

Wei et al., PRL'09
Checkelsky and Ong, arXiv: 0812.2866

2D electron gas

THEORY

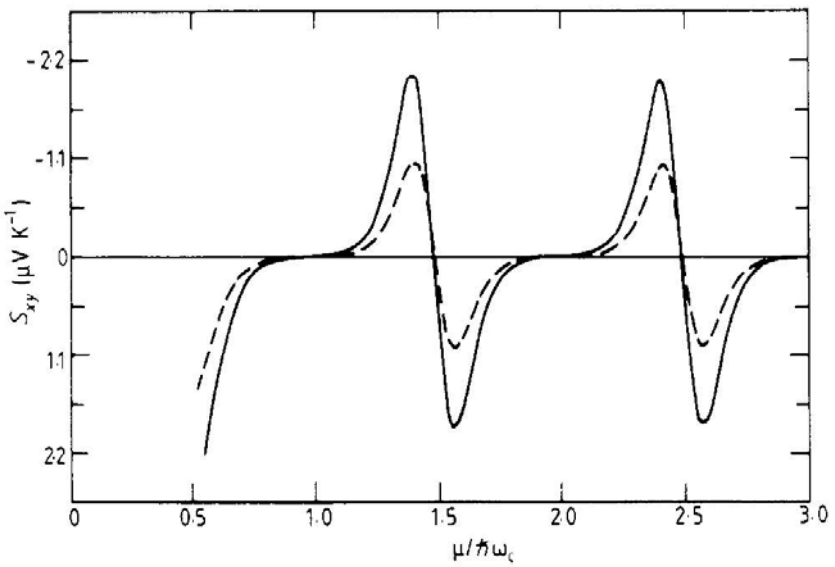
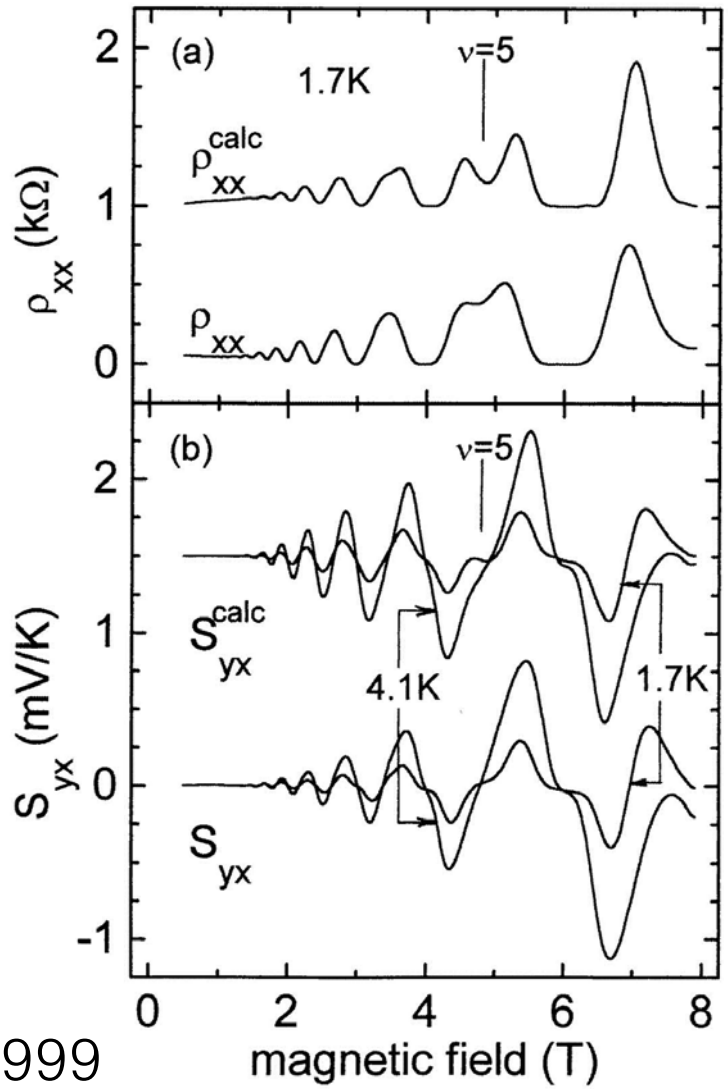


Figure 2. Non-diagonal component of the thermopower S_{xy} as a function of $\mu/\hbar\omega_c$ for $k_B T/\hbar\omega_c = 0.05$, $\Gamma/\hbar\omega_c = 0.05$ (full curve) and $\Gamma/\hbar\omega_c = 0.025$ (broken curve).

Jonson & Girvin, PRB '84

Oji, J. Phys. C '84

EXP.

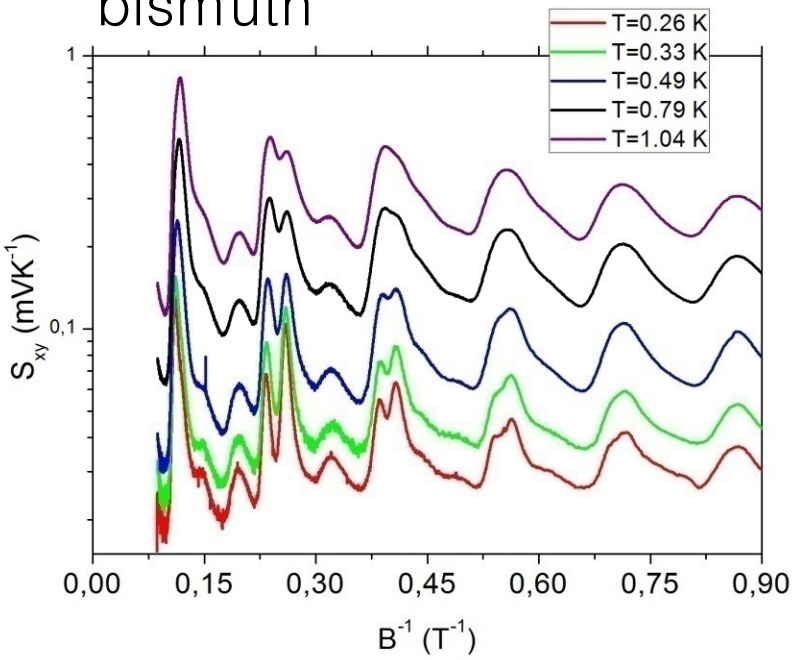


Fletcher 1999

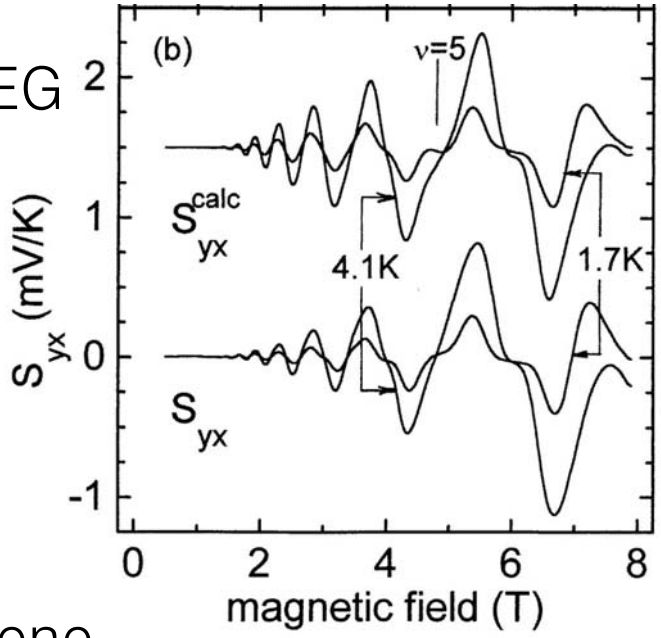
Figure 6. The upper panel shows ρ_{xx} for a 2DEG at a GaAs/$Ga_{1-x}Al_xAs$ heterojunction in the integer quantum Hall region. The lower trace is experimental and the upper trace is calculated using equation (20). Similarly the lower panel shows experimental S_{yx} and the calculated curves are from equation (19). The data are believed to be dominated by S_{yx}^{g}. In both panels the calculated curves are offset from zero for clarity. Taken from [45]

Summary: empirical correlation between the Nernst profile and dimensionality!

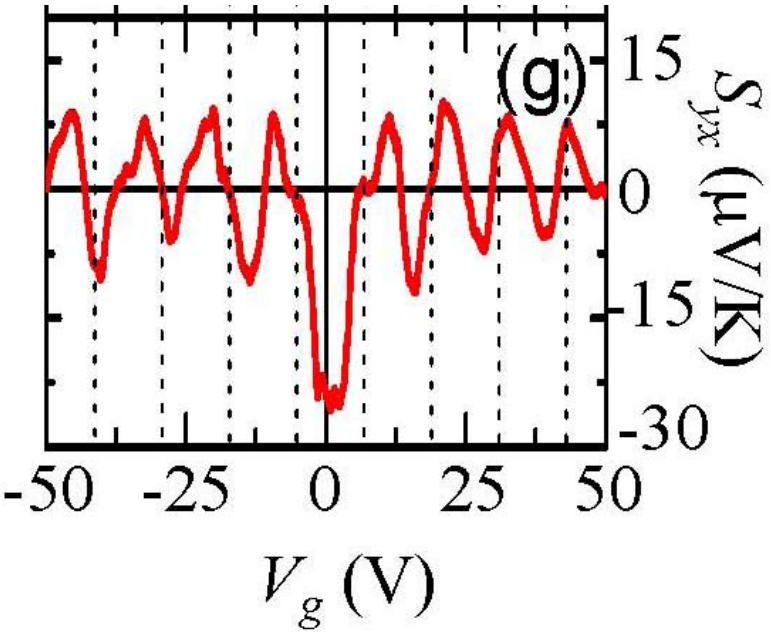
bismuth

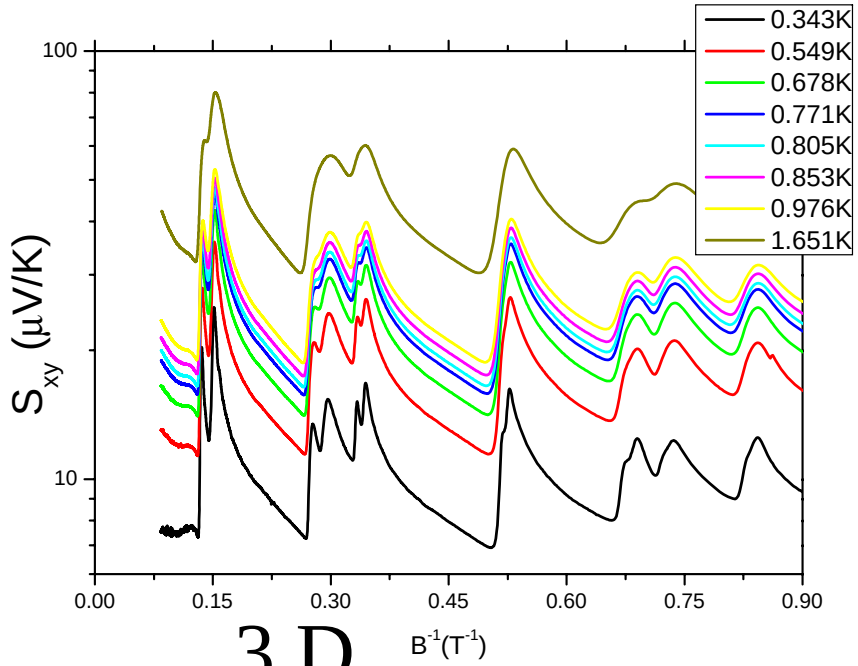


GaAs 2DEG



graphene

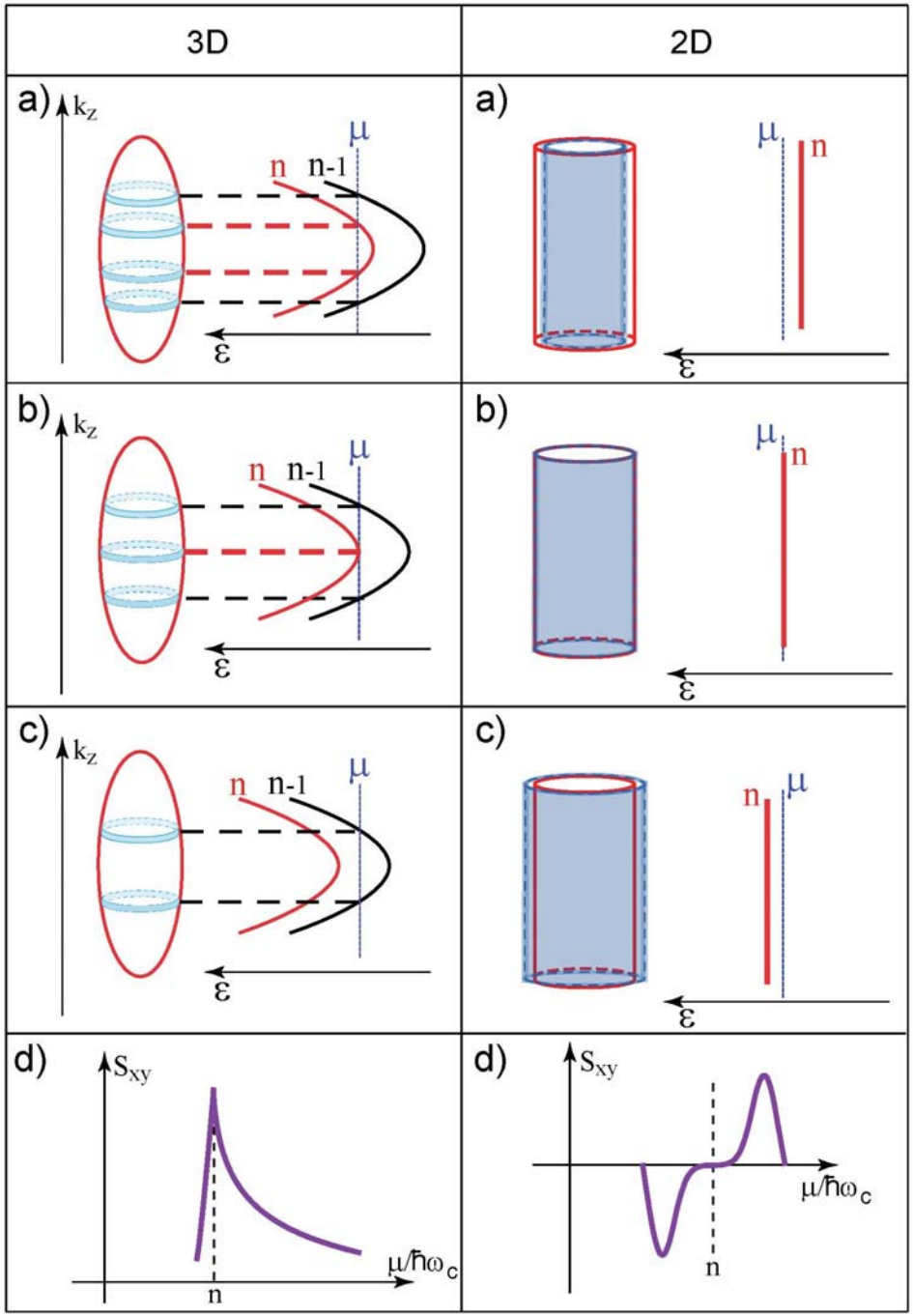


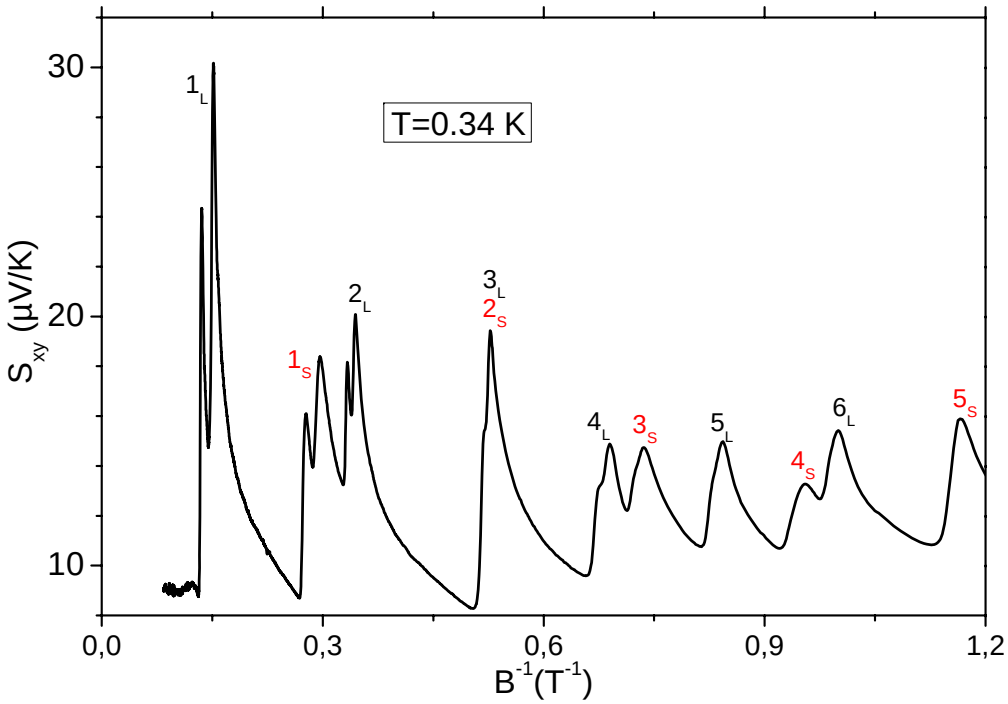


3 D

2D

Topological phase transition exclusive to 3D





Zhu et al., Nature Physics '09

For a review paper on topological phase transitions, see: Blanter, Kaganov, Pantsulaya and Varlamov Phys. Rep. 245, 159 (1994).

For a recent treatment of the sinigularity , see: Bergman & Oganesyan, Phys. Rev. Lett. 104, 066601 (2010).

J. Phys. F: Met. Phys. **18** (1988) 49–61. Printed in the UK

Theory of the de Haas–van Alphen effect

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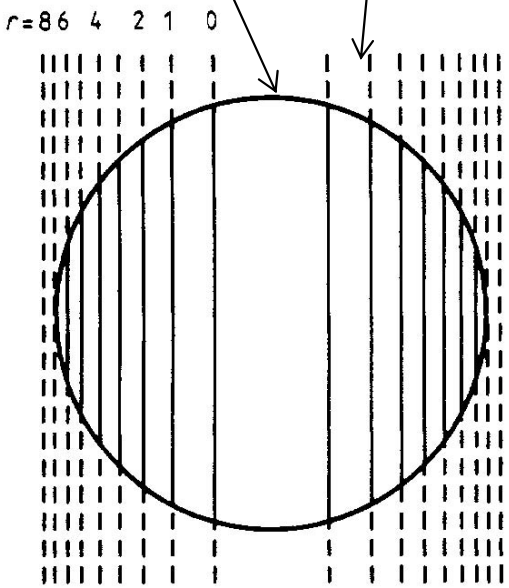
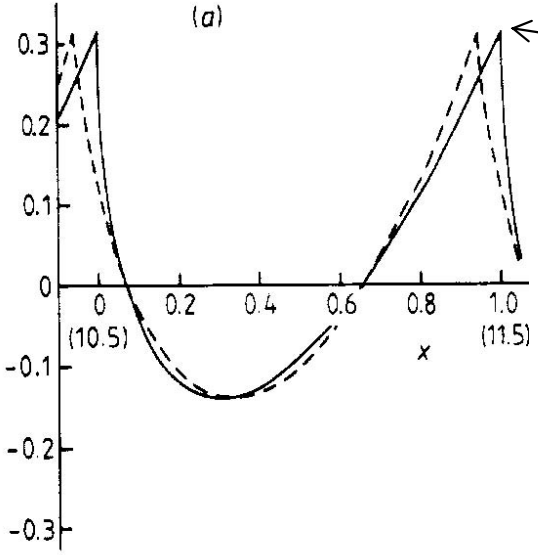
Received 2 April 1987, in final form 6 July 1987

Abstract. The oscillatory magnetisation (de Haas–van Alphen effect) of a free electron gas at $T=0$ can be expressed explicitly as the sum over p of terms proportional to $(p+x)^{1/2}$, where p is an integer going from 0 to n (the quantum number of the highest occupied Landau tube) and x is the oscillation phase $(0<x<1)$. This summation is evaluated by the Euler–Maclaurin formula and the result provides a simple algebraic expression describing the oscillation lineshape as a function of x. It is shown that this result is equivalent to the Lifshitz–Kosevich expansion in harmonics and also that, with appropriate reinterpretation of the parameters involved, the result is valid for an arbitrary electronic structure, provided the extremal cross-sectional area of the Fermi surface is a maximum. The modification needed for a minimum area is deduced by considering a hypothetical hyperboloidal Fermi surface. Modifications associated with oscillations in the Fermi energy, magnetic interaction and electron spin are also considered and an illustrative example is worked out in detail.

Landau tubes

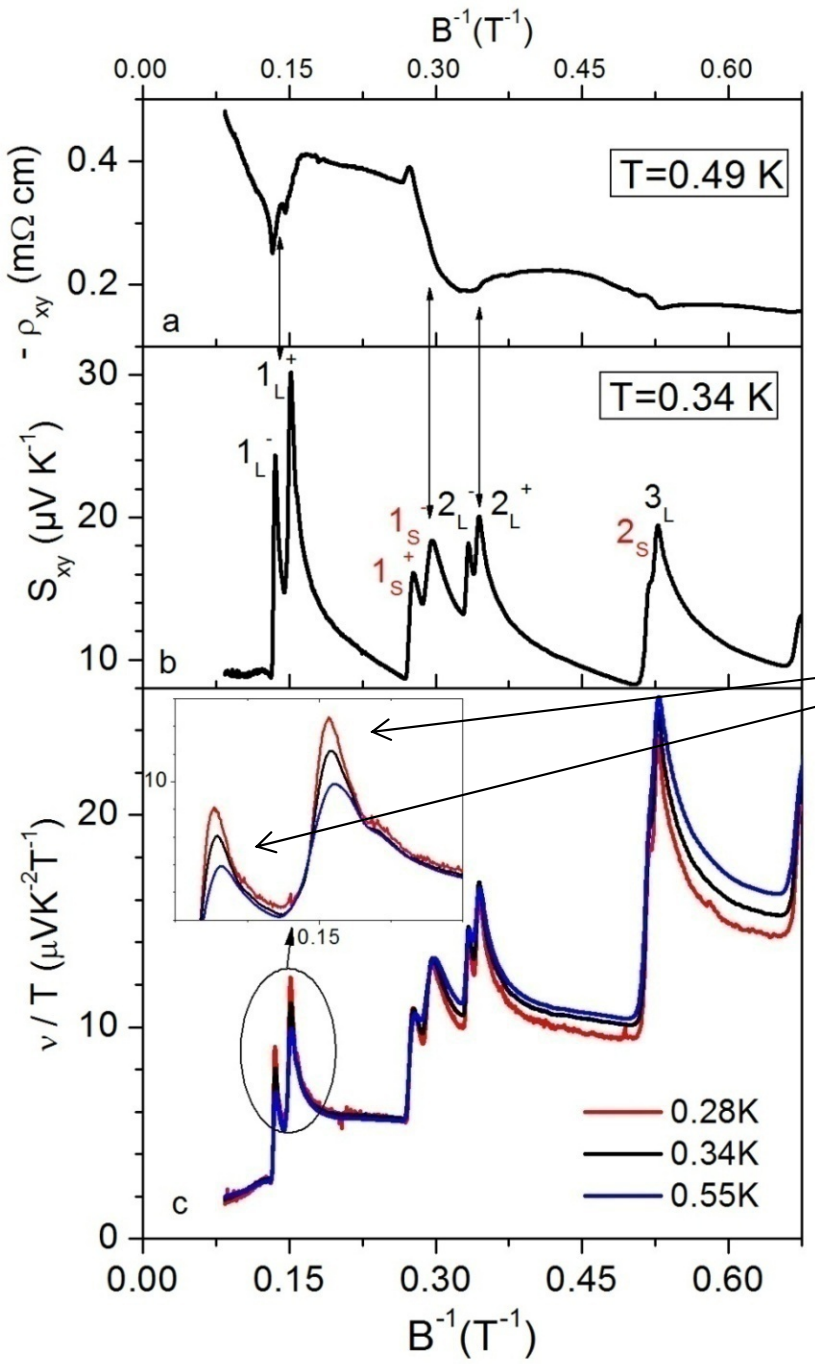
Fermi surface





- A singularity in DOS whenever a Landau tube leaves the Fermi surface!
- In presence of many Landau tubes this singularity smears out!

Nernst coefficient in graphite



Penultimate Landau tubes leave the Fermi surface

Divergent ν/T

Summary

- Thermoelectricity is a sensitive and underexplored probe of electronic correlations!
- In 3D there is a singularity in transverse thermoelectric response whenever a Landau level meets the Fermi level.
- Beyond the quantum limit, in bismuth there is a cascade of additional topological transitions, while graphite undergoes a thermodynamic phase transition.

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- *Koichi Izawa & Jacques Flouquet, CEA (Grenoble)*
- *Marie-Aude Méasson (Paris)*

Samples

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- Bertrand Lenoir *(Nancy)*
- Valerii Egorov *(Moscow)*

Theory

- Yuriy Sharlai & Grigorii Mikitik (Kharkov)