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Workshop on
Novel States and Phase Transitions in Highly Correlated Matter

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Thermoelectricity of correlated electrons: insights and enigmas

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Thermoelectricity of correlated electrons: insights and enigmas

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In collaboration with:

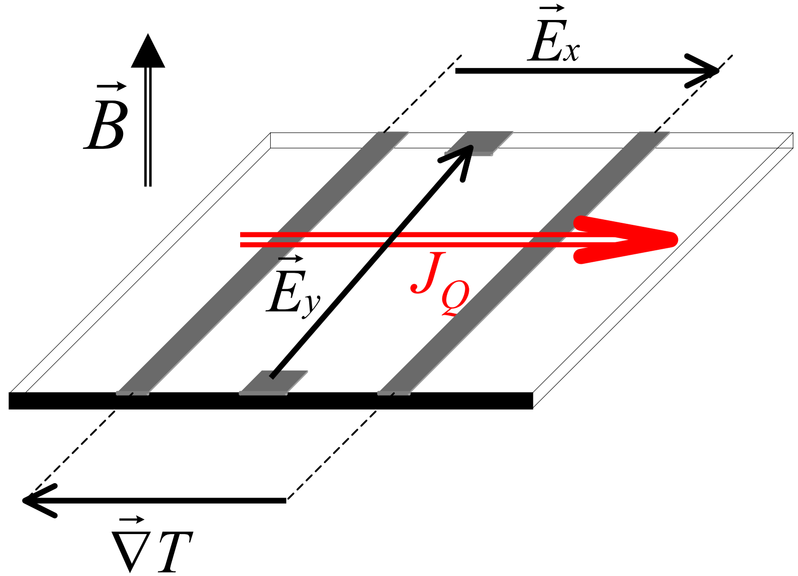
Heavy fermions:
- Didier Jaccard (Geneva),
- Jacques Flouquet (Grenoble),
- Koichi Izawa & Yuji Matsuda (Tokyo)

Overdoped LSCO:
- Saco Nakame (Orsay),
- S. Yaters & Nigel Hussey (Bristol)
- M. Nohara & Hide Takagi(Tokyo)

Underdoped cuprates:
- Cigdem Capan (Los Alamos)
- Z.Z. Li & Hélène Raffy (Orsay)

The 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.



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

Seebeck coefficient of the free electron gas

In the Boltzmann picture thermopower is linked to electric conductivity:

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

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 $\tau = \tau_0 \varepsilon^{\zeta}$:

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

For a proper treatment of the first term, see Miyake and Kohno '04

Thermopower and specific heat

In a free electron gas (with ξ=0):

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

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

- The free-electron-gas picture is inadequate at finite temperature
- Even in simple elemental metals, S(T) is not linear in temperature and displays a complex structure

What about the zero-temperature limit?

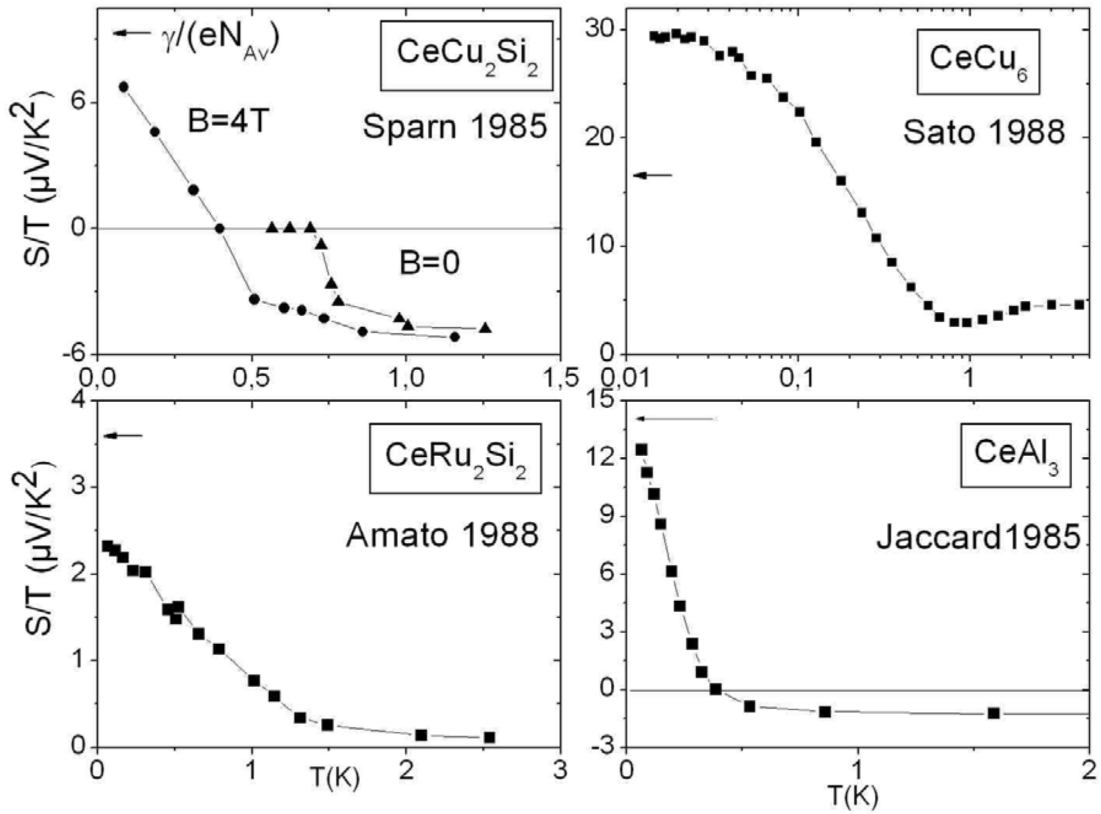
- The issue of carrier density : Is there a single carrier per formula unit?(often yes)

- The sign problem: The contribution of hole-like bands and electron-like bands are expected to be opposite in sign!

The magnitude of S/T has never been put under scrutiny in the zero-temperature limit!

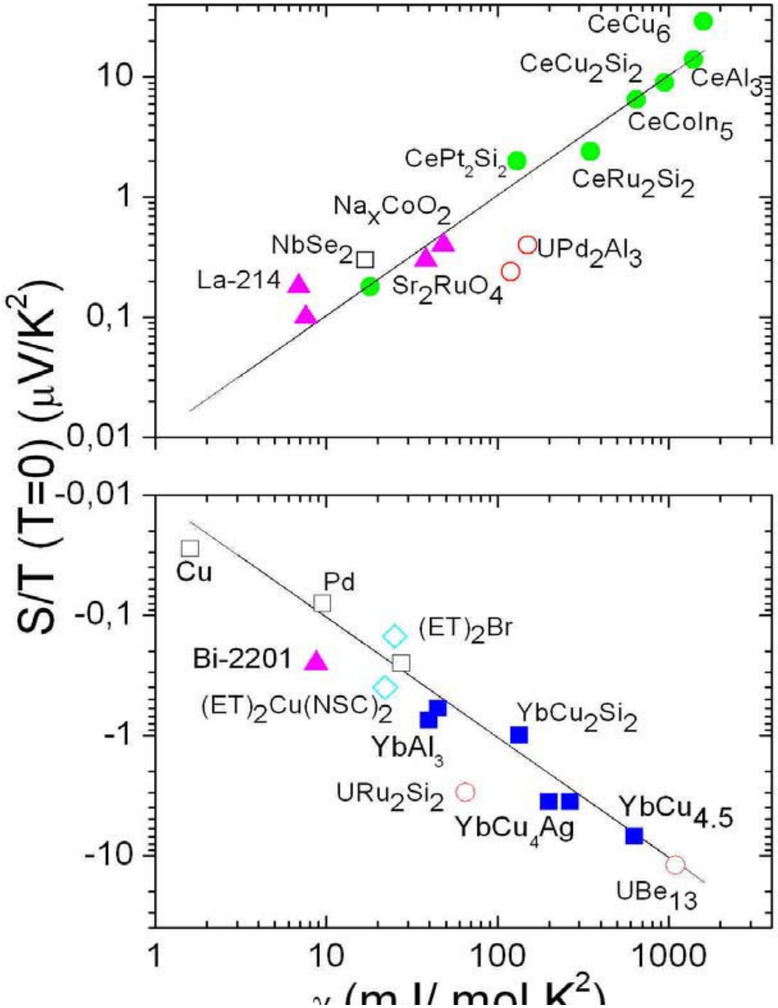
Heavy electrons in the T=0 limit

Two-decades-old data reconsidered!

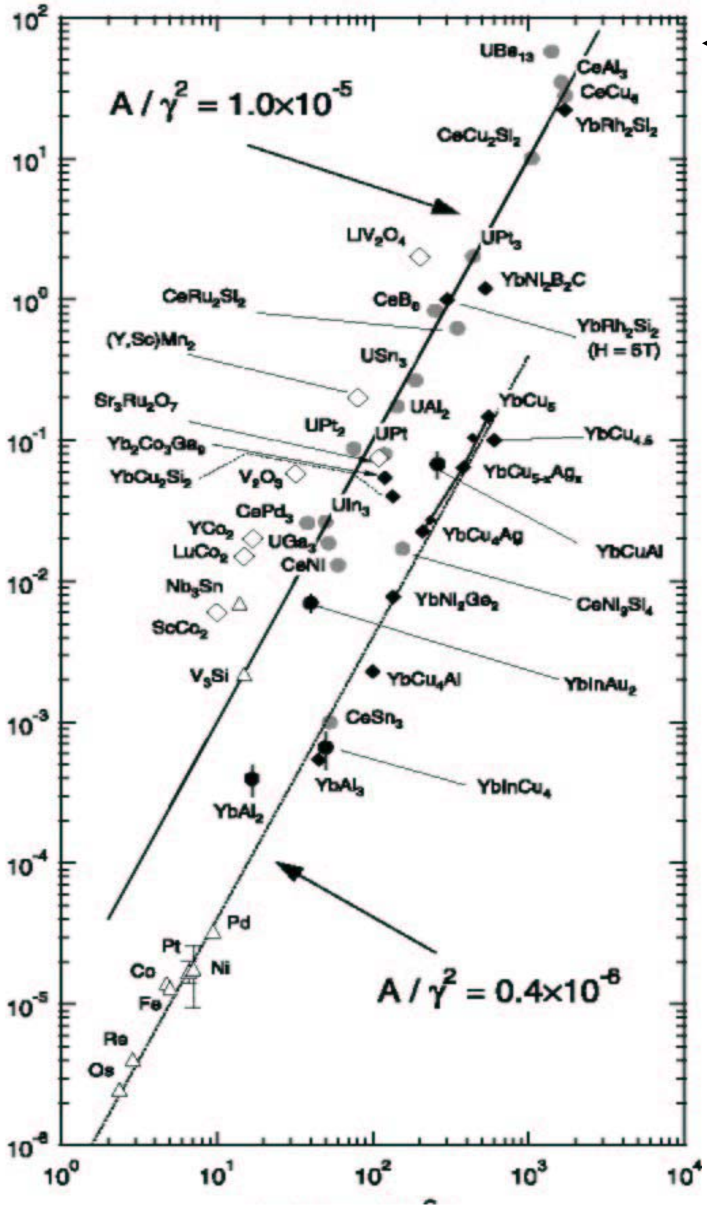


Extrapolated to T=0, data yields a q close to unity!

A new plot linking two distinct signatures of electron correlation



Ratios used to characterize a Fermi liquid

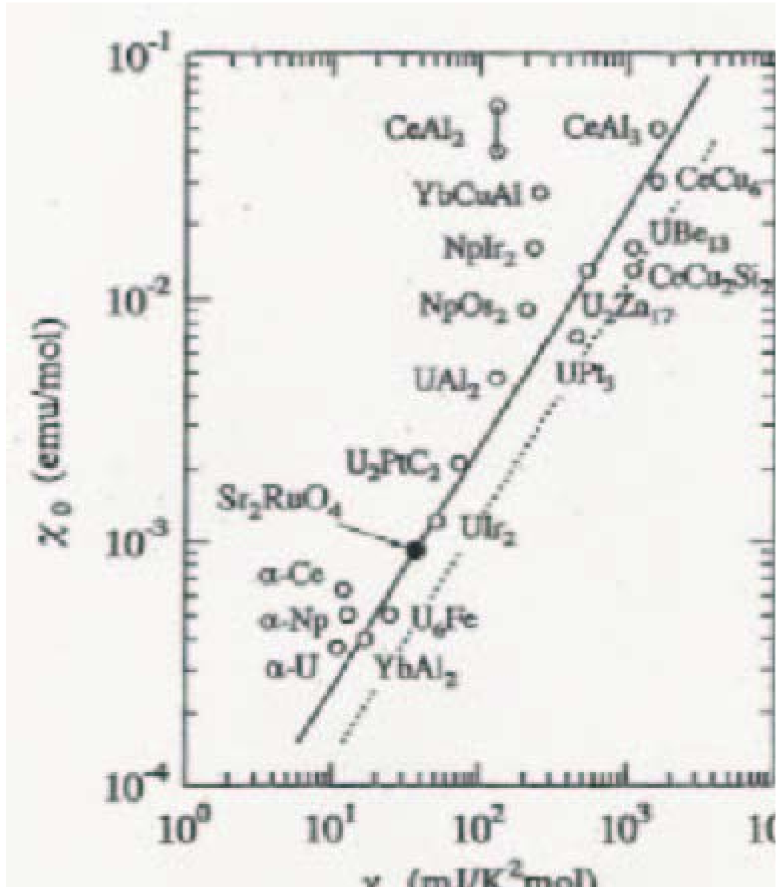


← *Kadowaki-Woods ratio (A/γ^2)*

[$\rho=\rho_0+AT^2$]   Tsujii, 2003

Wilson Ratio (χ/γ)

Maeno 1997

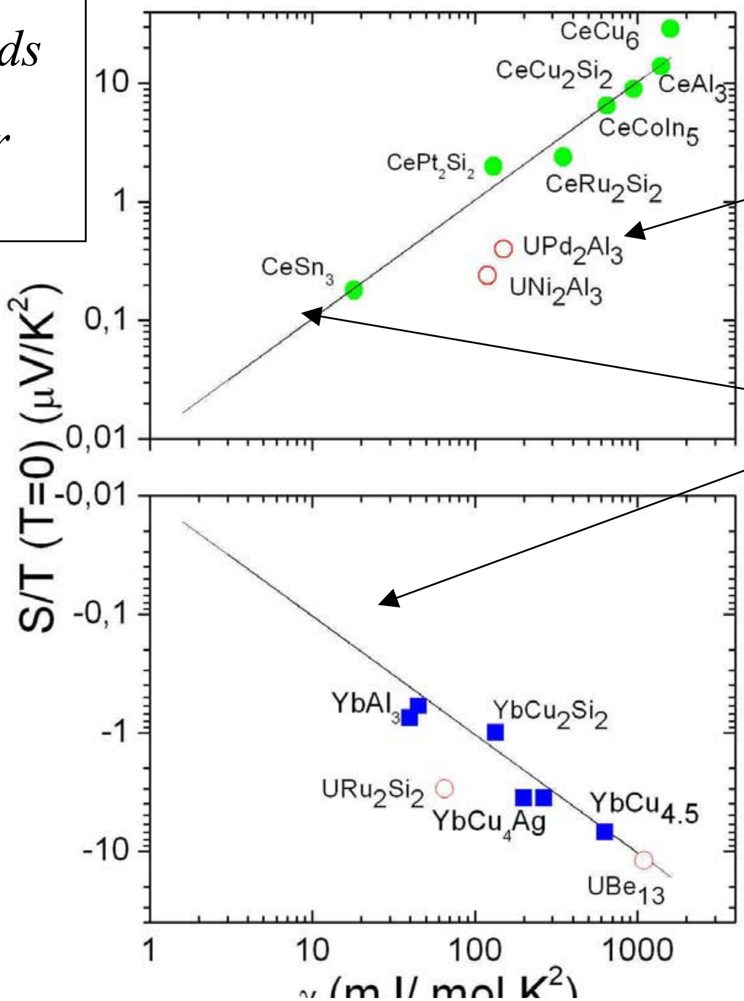


Heavy Fermions

S>0 in Ce compounds

S<0 un Yb Compounds

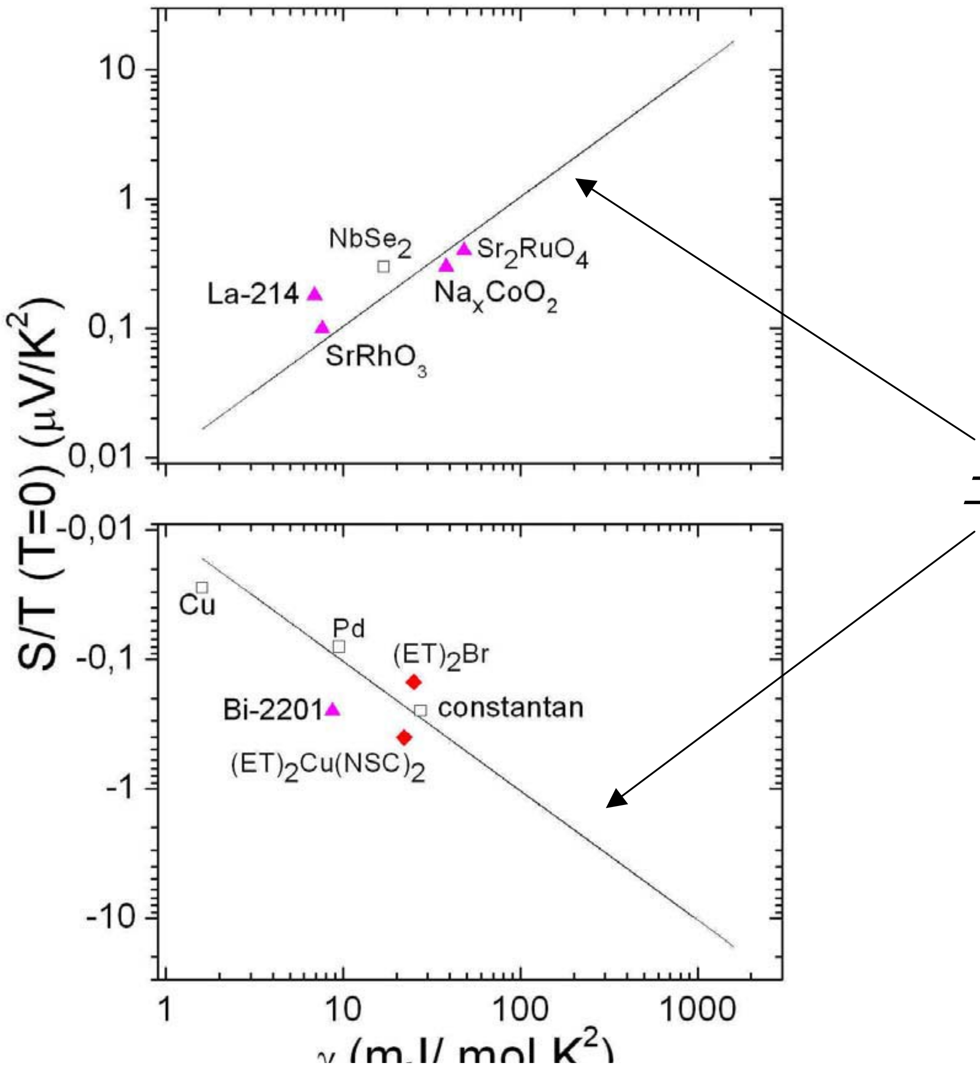
No subkelvin data for most U compounds



No subkelvin data aavailable for these two!

$\pm\gamma/(eN_{Av})$

Other metals



$\pm\gamma/(eN_{Av})$

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
$UPd_2 Al_3$	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 $q = \frac{S}{T}\frac{N_{Av}e}{\gamma}$ is discussed in the text.

Behnia, Jaccard, Flouquet
J. Phys. Condens. Matter
16, 5187(2004)

Sr_2RuO_4: a well-established Fermi liquid

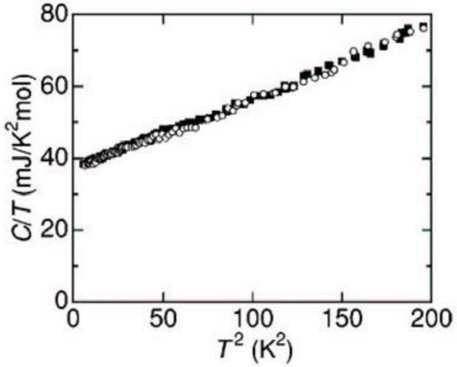


FIG. 7. The total specific heat of Sr_2RuO_4 divided by temperature between T_c and 14 K in zero field (filled squares) and an applied magnetic field of 14 T (open circles) applied parallel to *c*. From Mackenzie, Ikeda, *et al.* (1998).

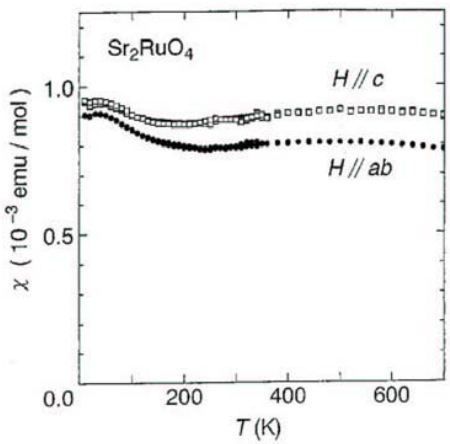
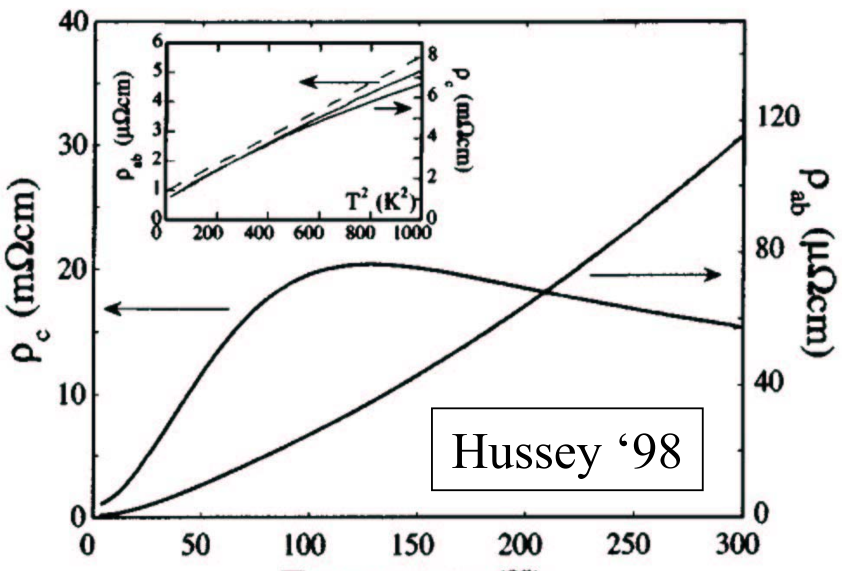
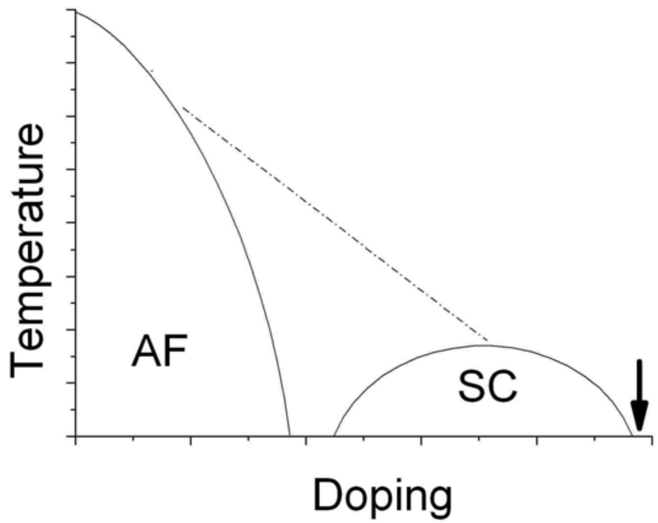
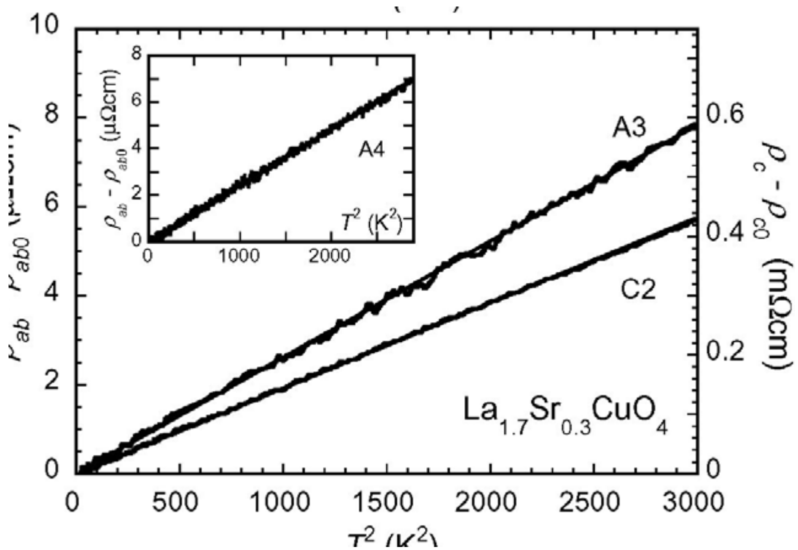


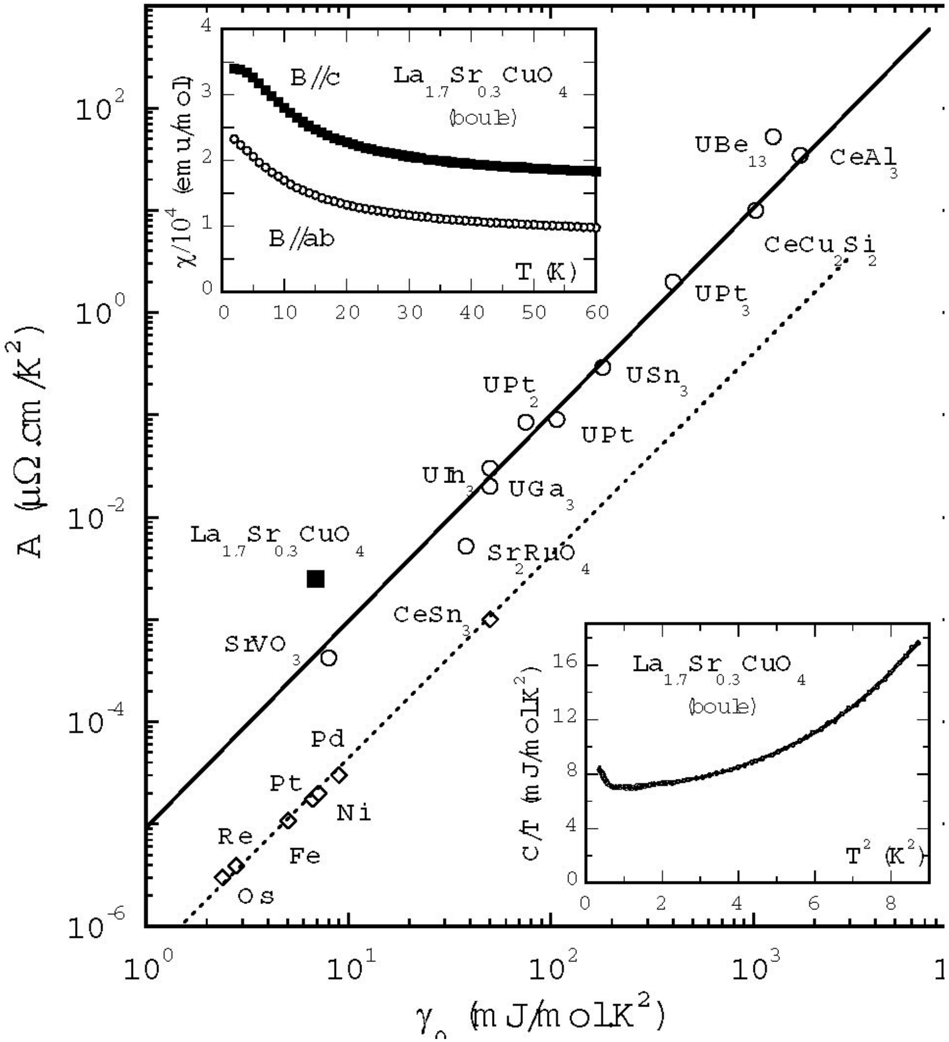
FIG. 8. The static susceptibility of Sr_2RuO_4 for fields of 1 T applied parallel to the *ab* plane and the *c* axis, from Maeno *et al.* (1997).



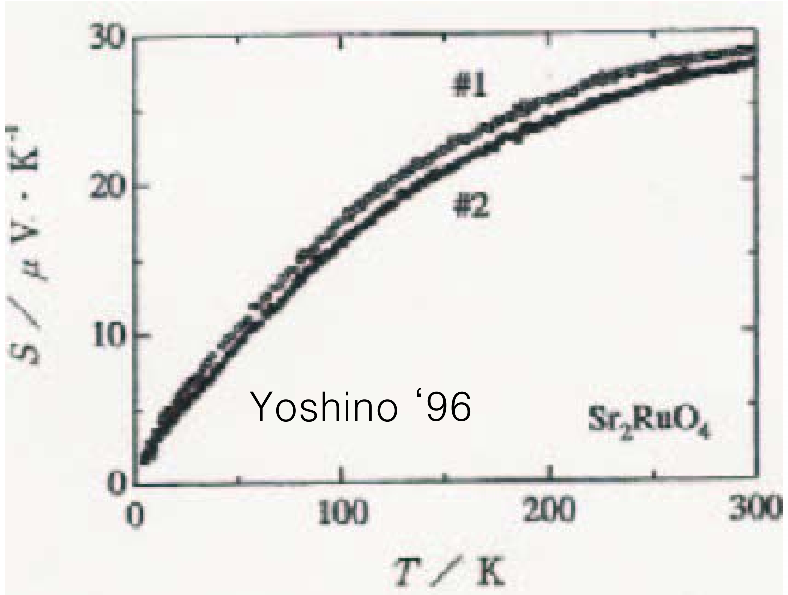
Another remarkable Fermi liquid: $La_{1.7}Sr_{0.3}CuO_4$



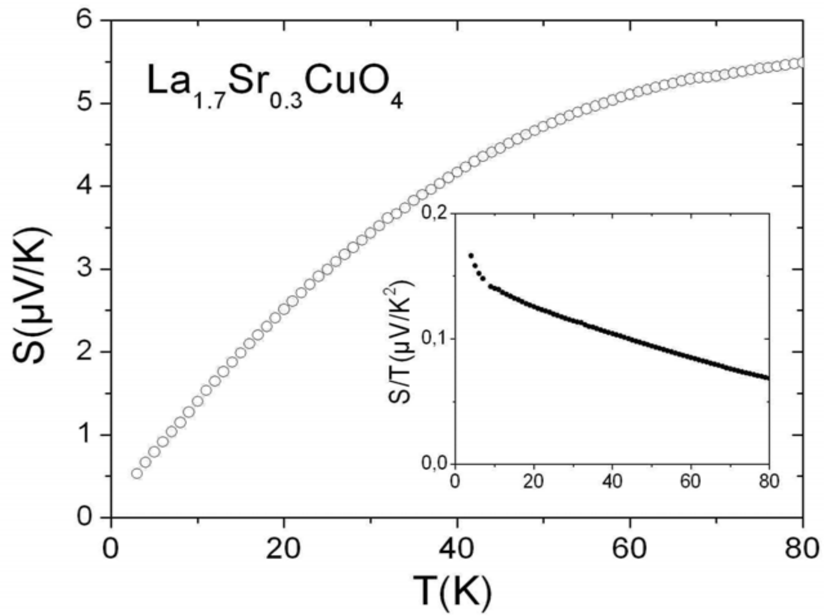




The ratio q is a probe of carrier density!



Jin *et al.* , to be published



$\gamma = 38\ mJ / mol\ K^2$

$S/T = 0.3\ \mu V/K^2$

q= 0.8 (1.2 carrier per site)

$\gamma = 6.9\ mJ / mol\ K^2$

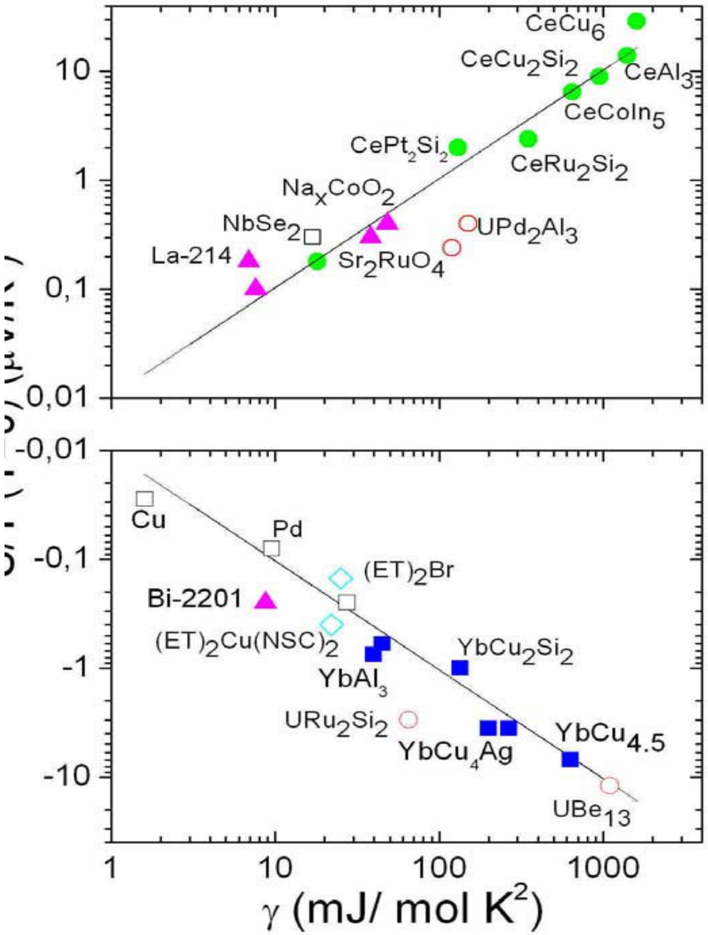
$S/T = 0.18\ \mu V/K^2$

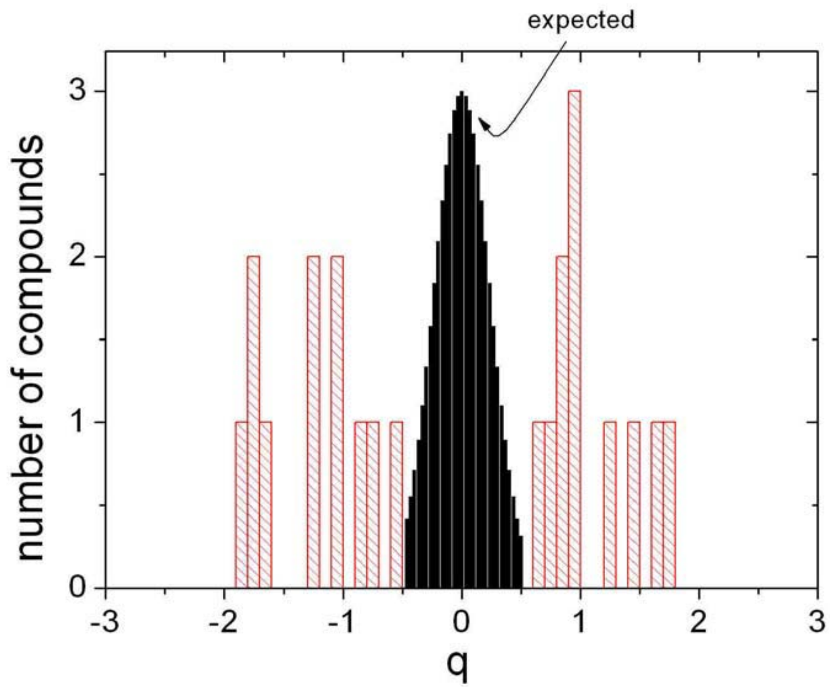
q= 2.5 (0.4 carrier per site)

γ measures entropy per elementary cell!
S/T measures entropy per carrier!

Is then the
FS of LSCO at x=0.3
still hole-like?
What about ARPES?

An enigma!





Asymmetry of mass renormalisation for electrons and holes?

What governs the sign?

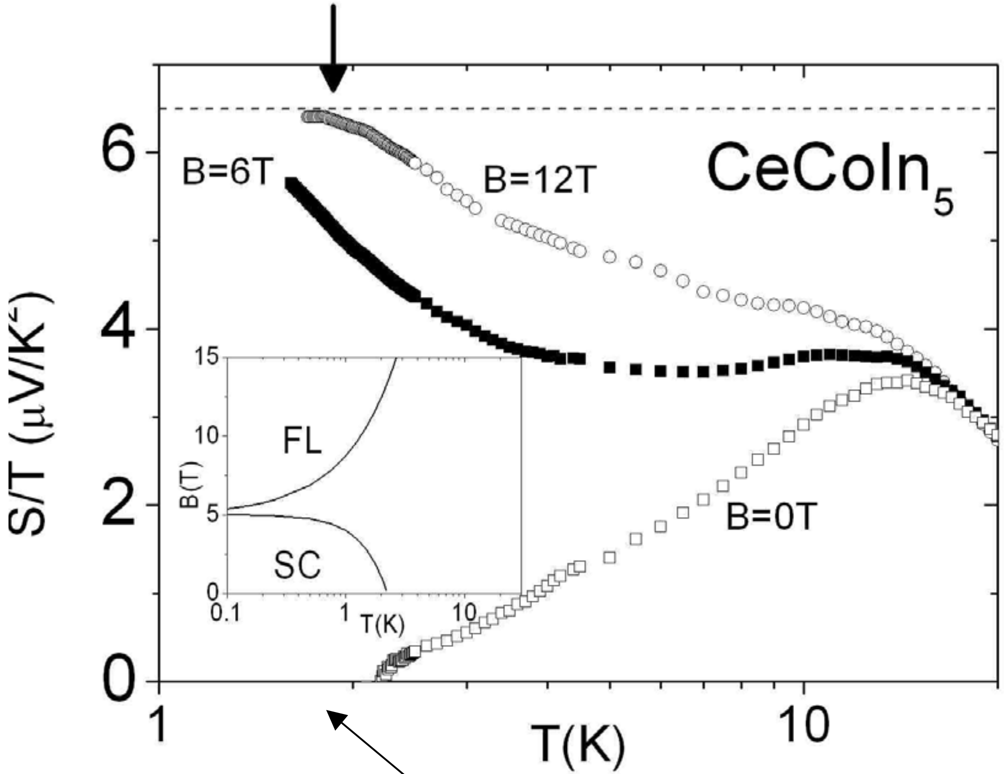
Why does this simple free-electron-gas picture work even in the case of multi-band Fermi surfaces?

An observation

- [Within the resolution of available data], the magnitude of q is the same for different families
- This contrasts with KW plot :A/γ^2 varies from $0.04a_0$ (in Yb compounds)to 50 a_0 (Na_xCoO_2).
- Contrary to KW, q links two zero-energy properties.[Yet it should mirror anomalous $\tau(\varepsilon)$!]

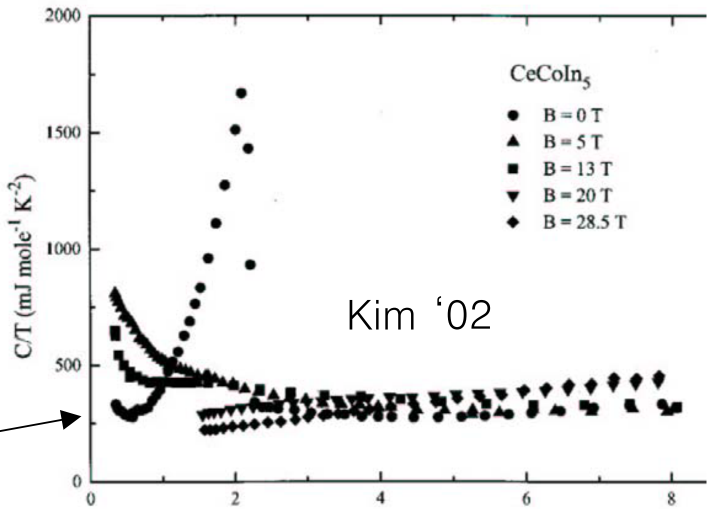
A tool for tracking non-trivial physics!

Example: $CeCoIn_5$ at the onset of superconductivity:

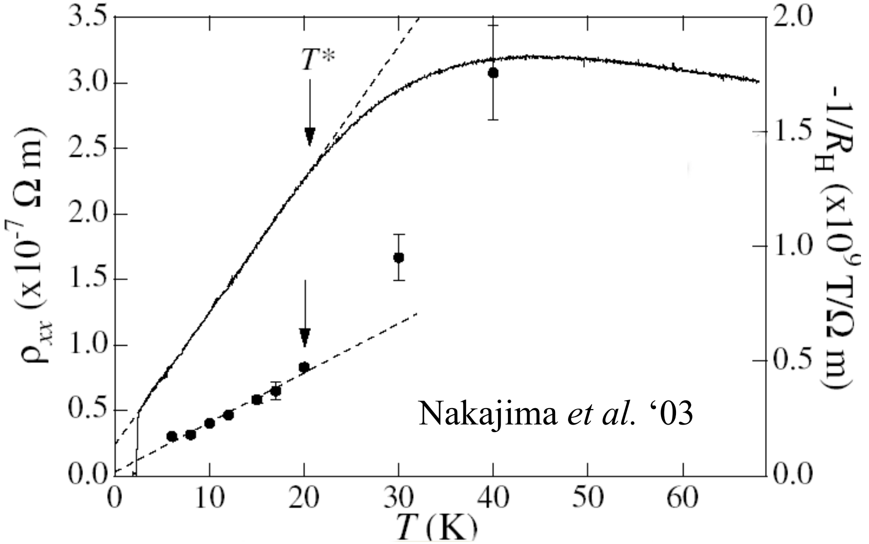


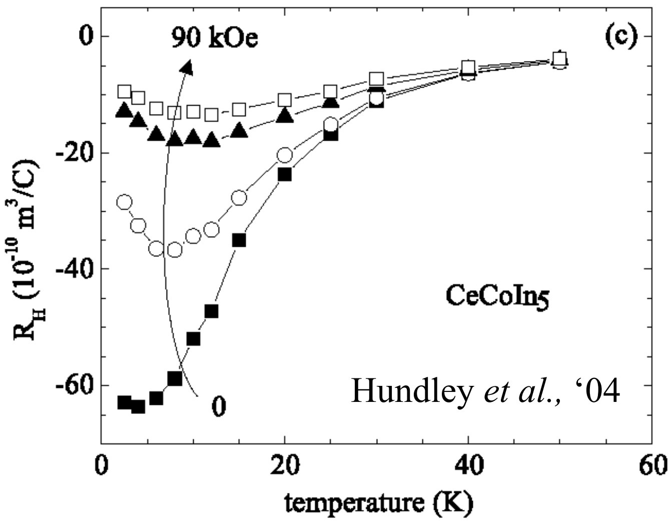
S/T strongly field-dependent!

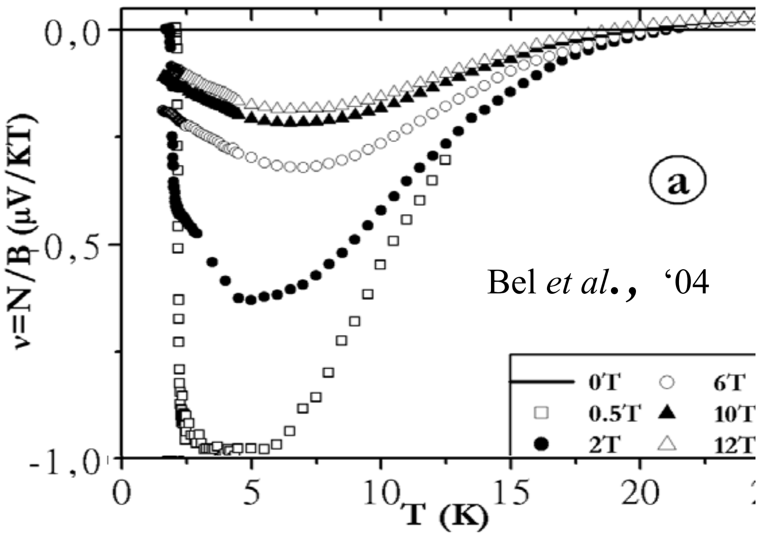
And γ field-independent up to 13T!



Zero-field anomalous transport properties of $CeCoIn_5$







inear resistivity

emperature and field-dependent Hall effect

iant Nernst effect

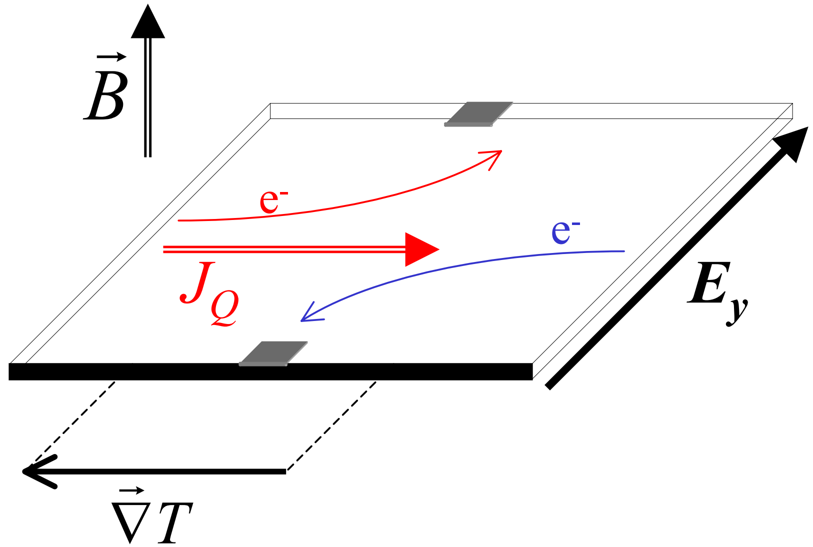
ow-magnitude thermopower

All zero-field anomalies!
ll Pointing to a non-trivial $\tau(\varepsilon)$]

Sondheimer cancellation in single-band metals

✓ Counterflow of hot and cold electrons

$J_Q \neq 0$; $J_e = 0$; $\boldsymbol{E_y} = \mathbf{0}$

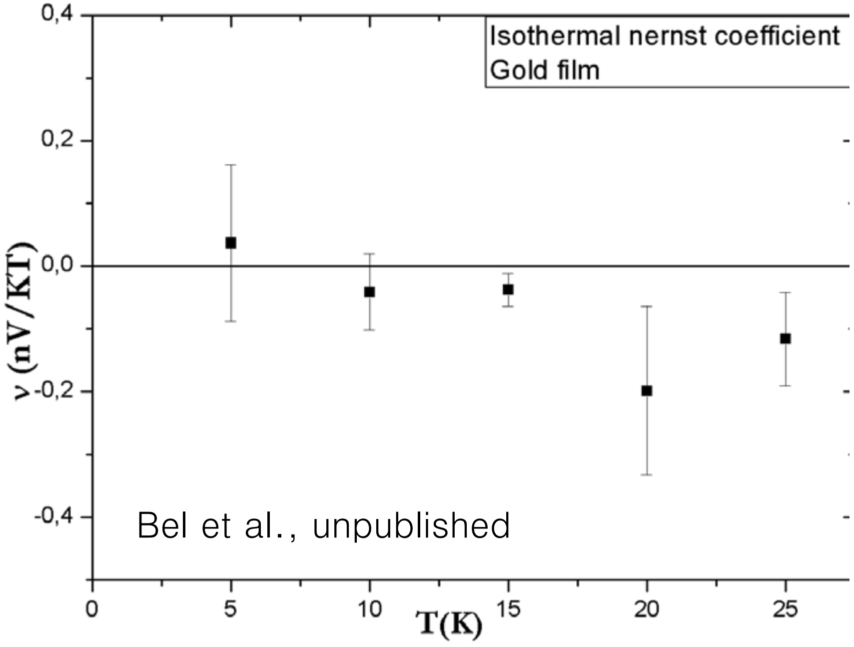


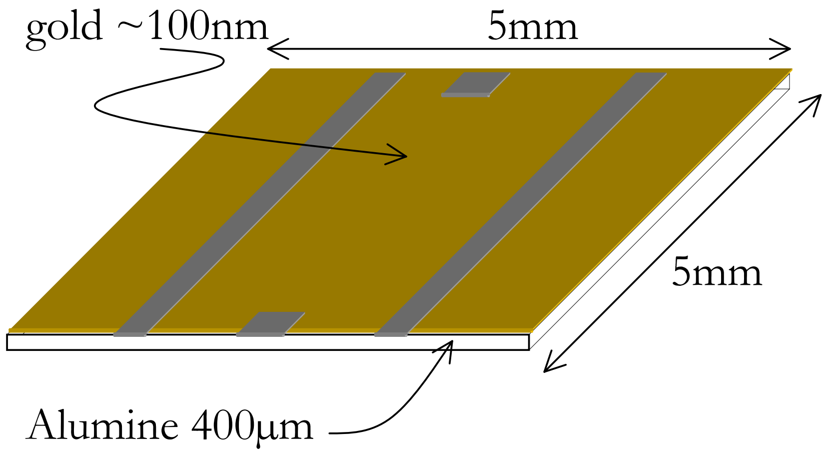
Dans un métal simple, l'effet Nernst est nul

Isothermal gold effect in gold

✓ Gold film evaporated on Alumine

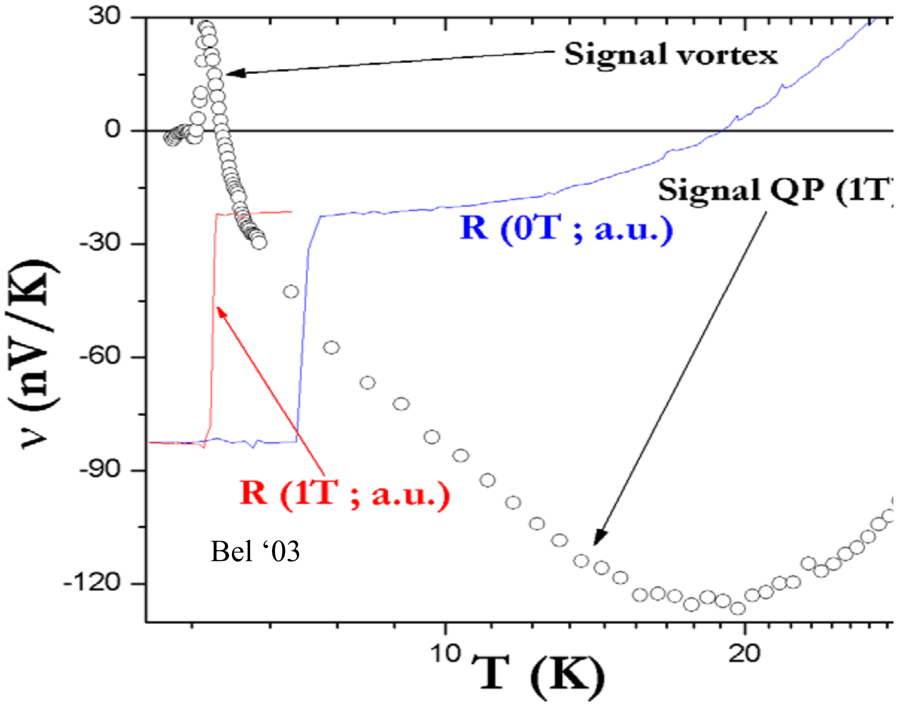
✓ Very small Nernst coefficient ($\nu = N/B \sim 0.1$ nV/KT)

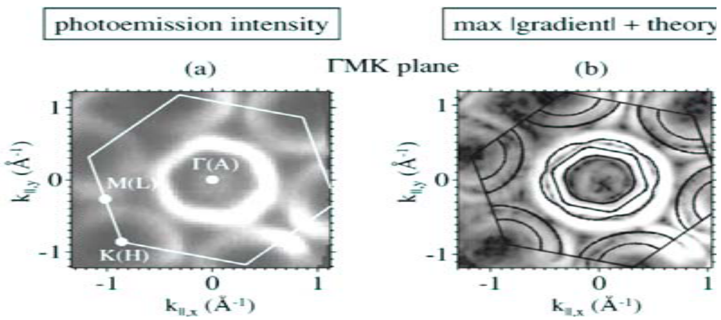




Ambipolar Nernst effect in $NbSe_2$

- $NbSe_2$: superconductor with Tc~ 7,2 K.
- Charge Density wave at ~32K.
- Multi-band metal
- Positive vortex signal
- **A negative Nernst signal of omparable magnitude in the ıetallic state.**



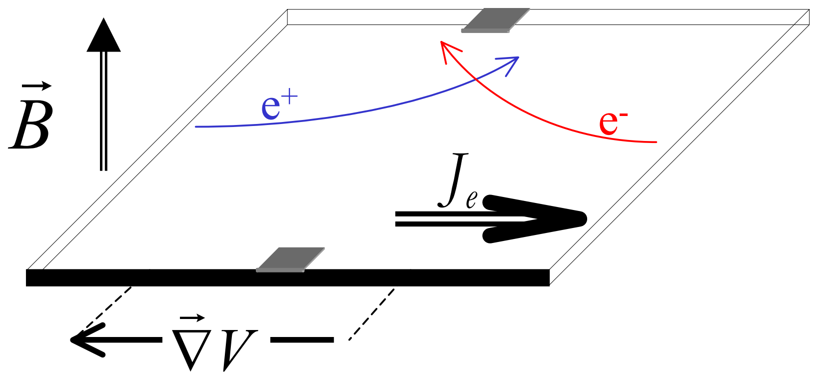


Surface de Fermi
Rossnagel et al. *PRB* 64, 235119 (2001

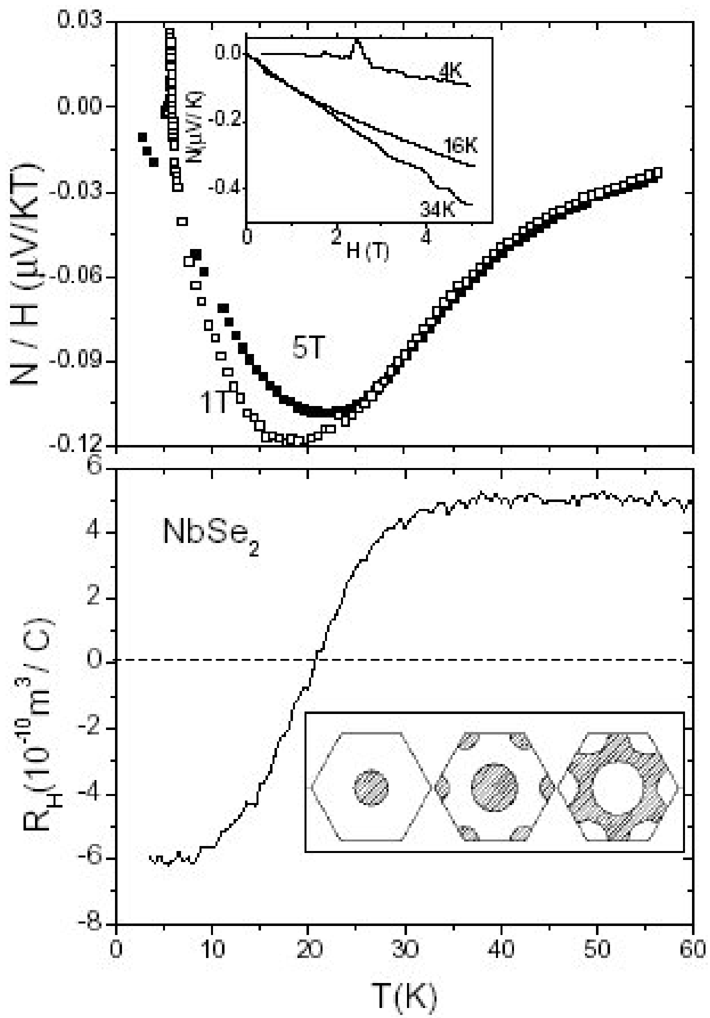
No Sondheimer cancellation in a multi-band metal!

✓ The Nernst signal is maximum when the hall coefficient is zero!

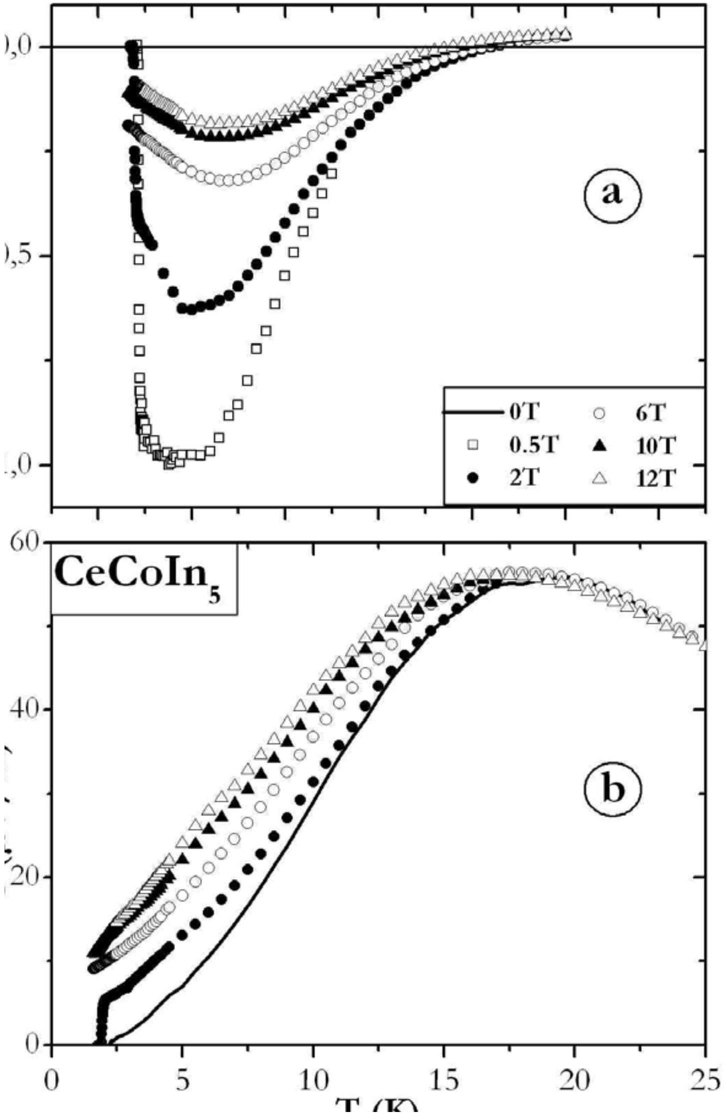
✓Known as the ambipolar Nernst effect!



$J_Q = 0$; $J_e \neq 0$; $\boldsymbol{R_H} = \mathbf{0}$



Giant Nernst effect in $CeCoIn_5$



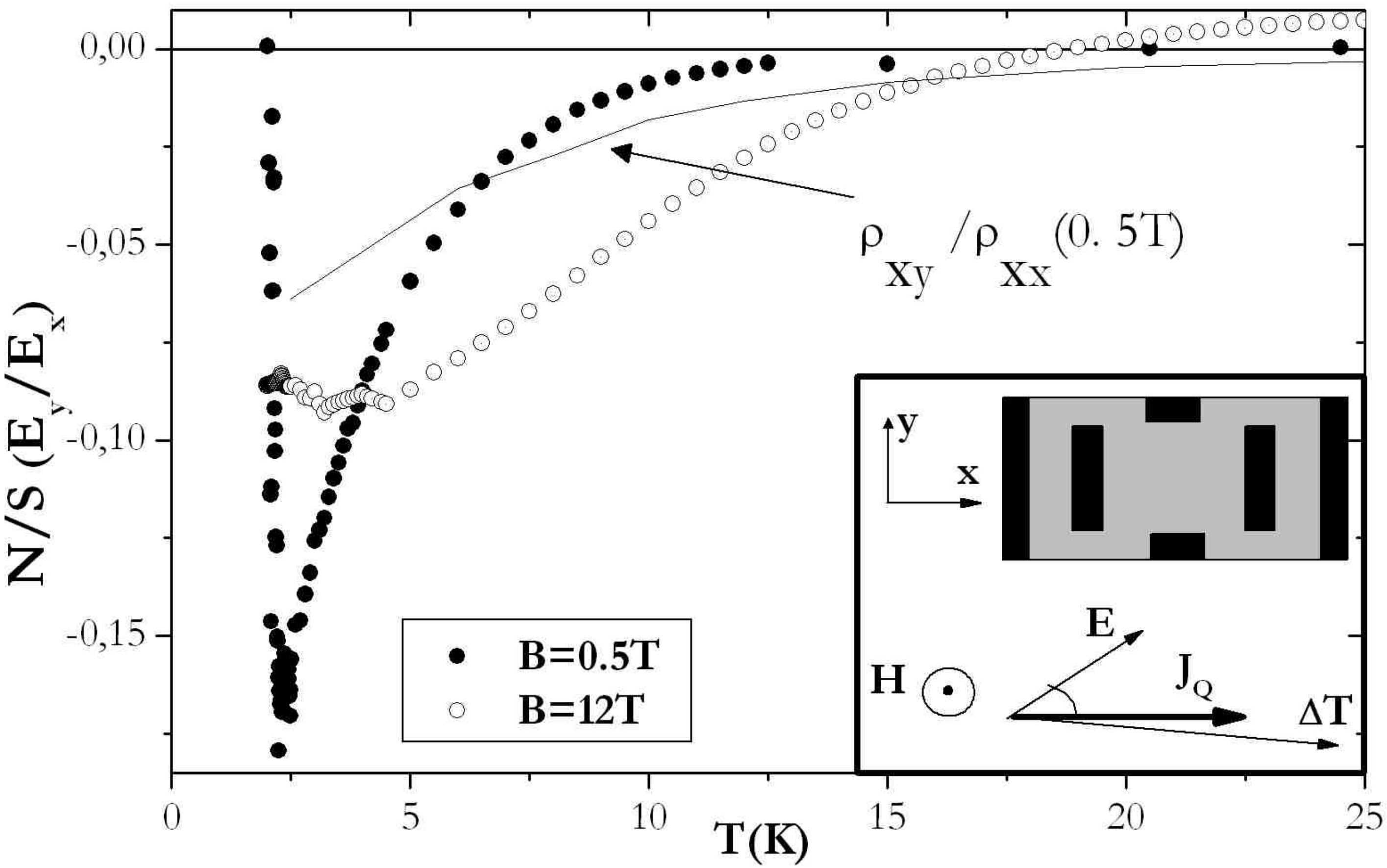
✓One order of magnitude larger

✓Emerges in presence of a large Hall effect

✓Restricted to a limited region in the H,T plane

$$\nu \propto \left(\frac{\partial \tau}{\partial \varepsilon}\right)$$

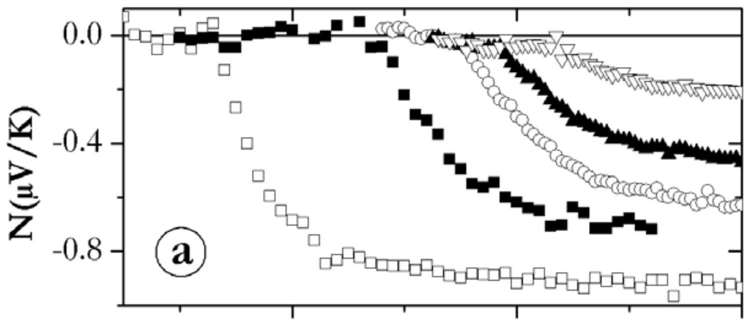
In the T=0 limit, the N/S ratio diverges!



A longitudinal thermal gradient produces a purely transverese electric field!

The vortex Nernst signal in $CeCoIn_5$

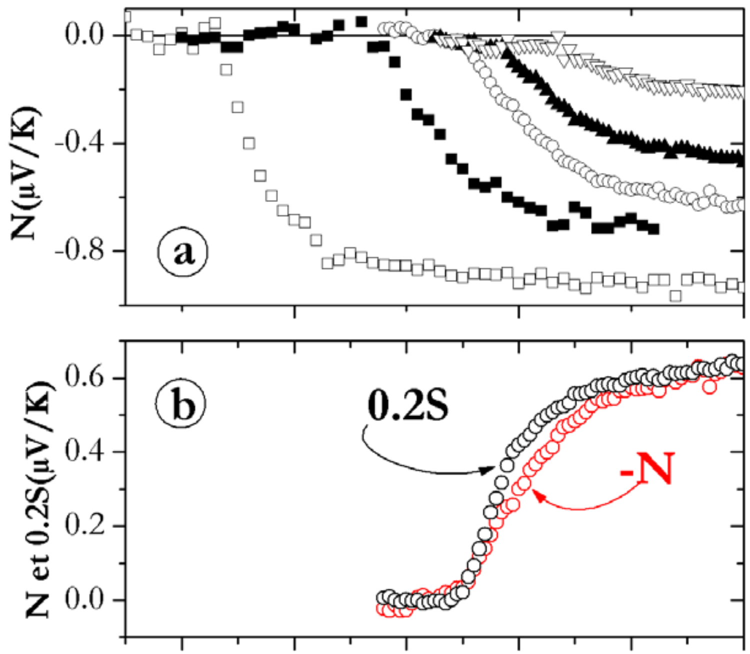
✓ No trace of a positive signal in the vicinity of the superconducting transition



The vortex Nernst signal in CeCoIn5

✓ No trace of a positive signal in the vicinity of the superconducting transition!

✓But Nernst signal colapses faster than the Seebeck signal.

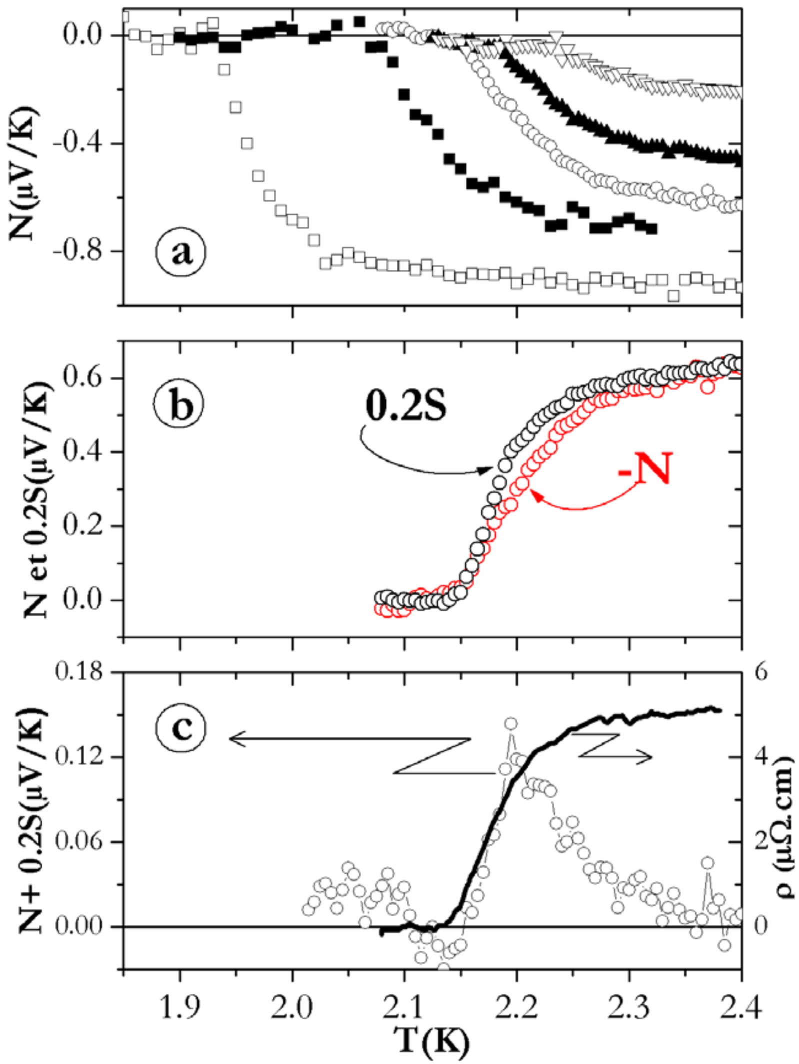


The vortex Nernst signal in CeCoIn5

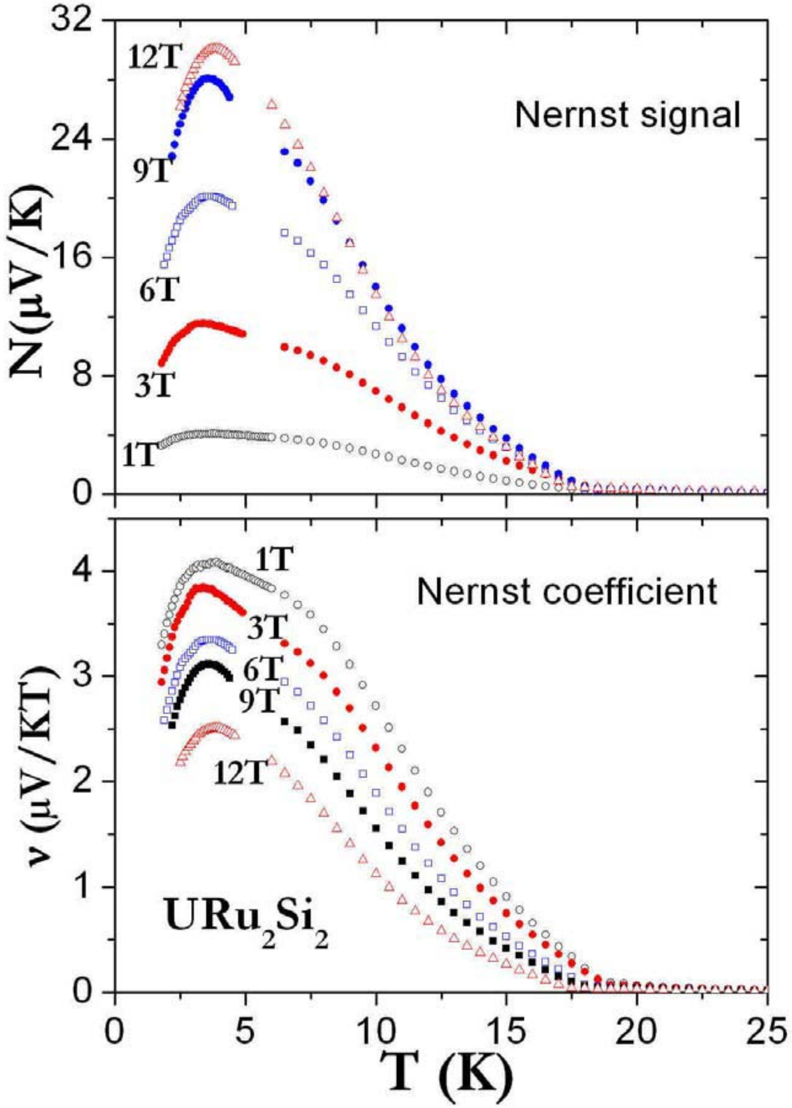
✓ No trace of a positive signal in the vicinity of the superconducting transition

✓But Nernst signal collapses faster than the Seebeck signal.

✓Plotting *aS-N,* the vortex Nernst signal, overwhelmed by the normal-state signal, can be extracted.



Giant Nernst effect in the hidden order phase of URu_2Si_2



Hidden order in URu_2Si_2

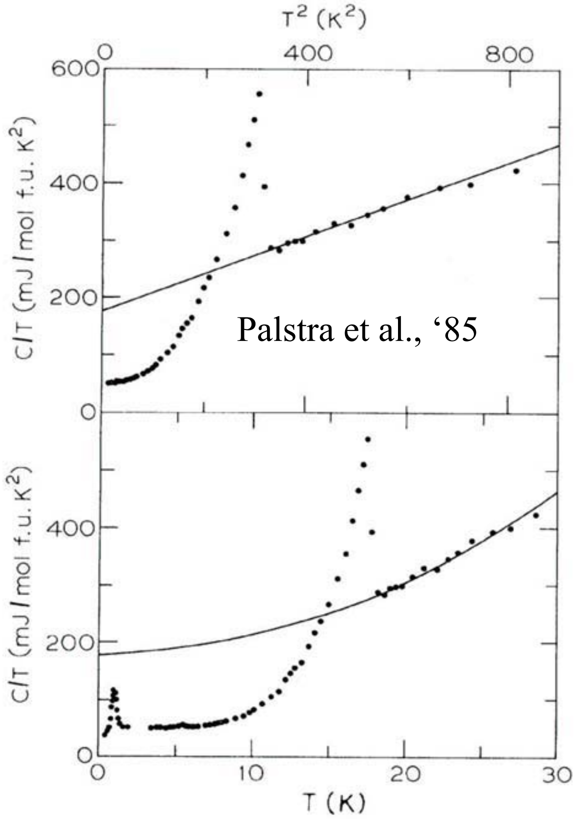


FIG. 1. Specific heat of URu_2Si_2 plotted as C/T vs T^2 bove) yielding γ and Θ_D, and as C/T vs T (below) show- g the entropy balance.

✓Large macroscopic anomalies occur at T_0 ~18K

✓But only a tiny magnetic moment($0.02\mu_B$) appears!

✓The order parameter is yet to be identified!

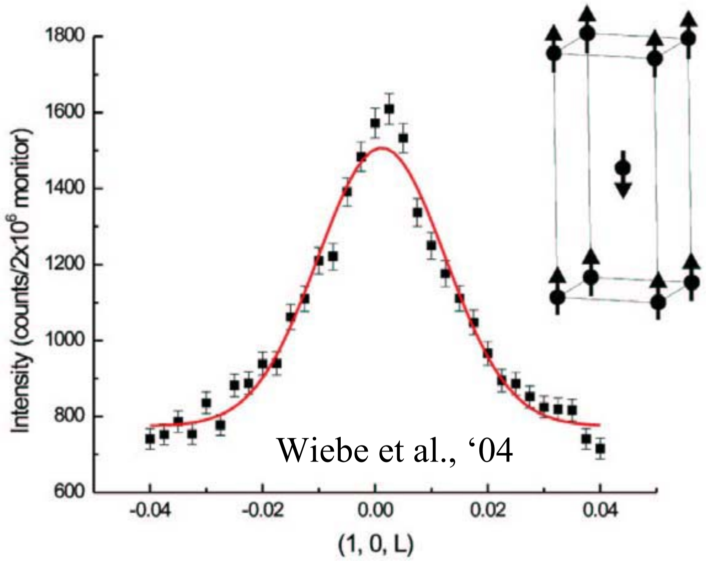
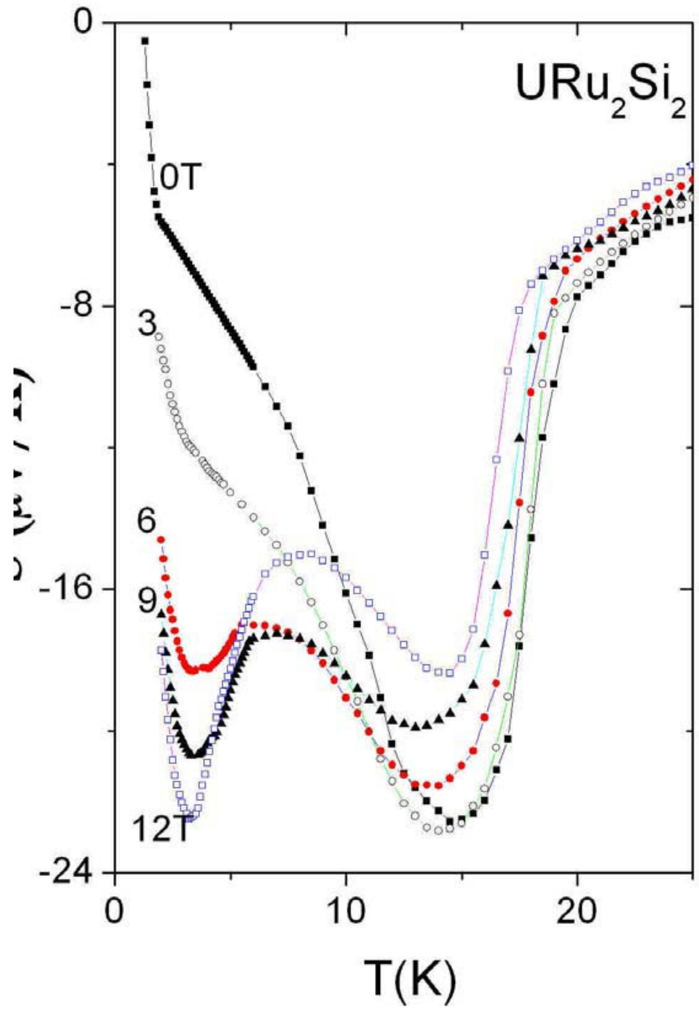
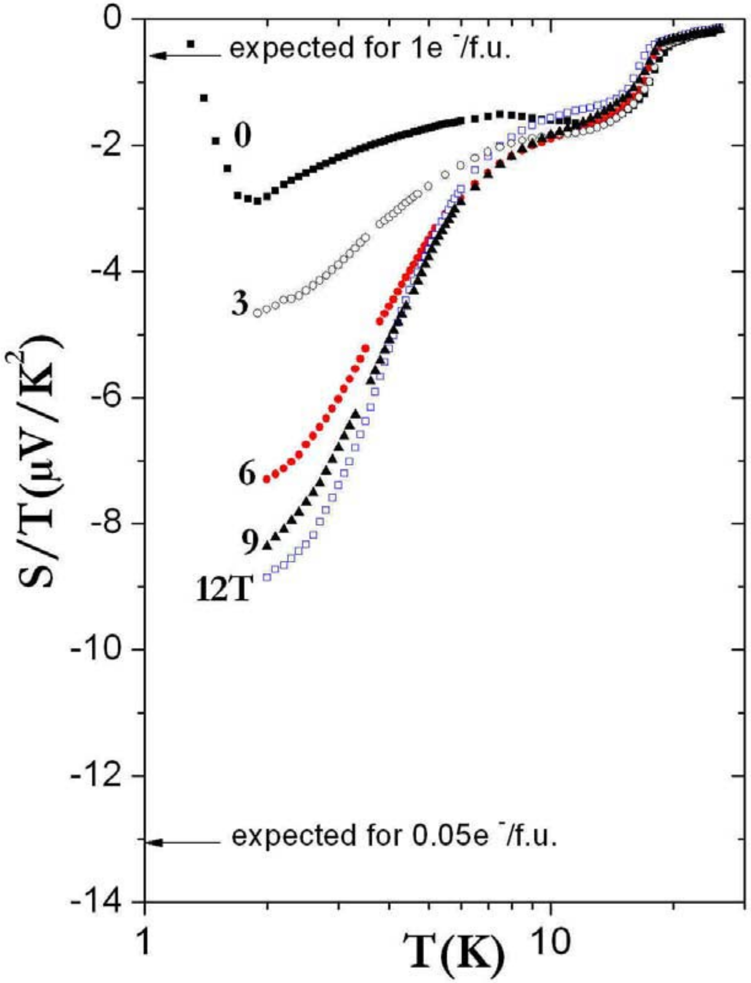


FIG. 1. The antiferromagnetic Bragg peak at (1, 0, 0) and $T = 8$ K (fit is to a Gaussian). The inset shows the corresponding magnetic structure for the U^{4+} moments.

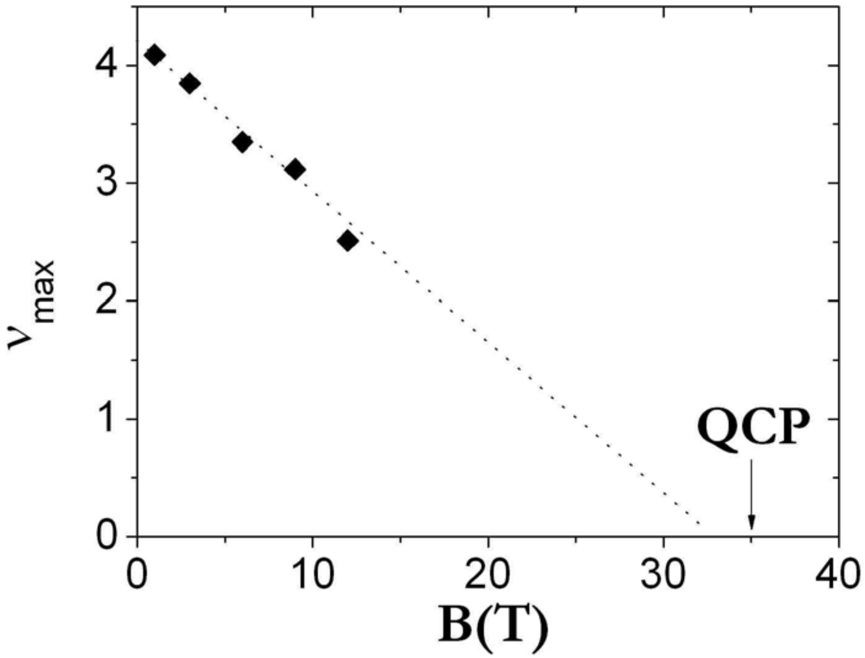
Seebeck coefficient in the hidden-order state



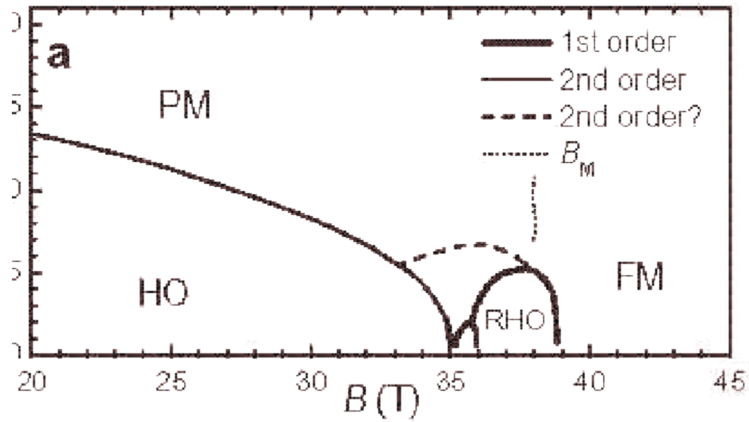


A small number of carriers survive in the hidden-order state!

The giant Nernst effect would not survive the destruction of the hidden order at a high magnetic field!



Decrease of maximum Nernst coefficient with magnetic field



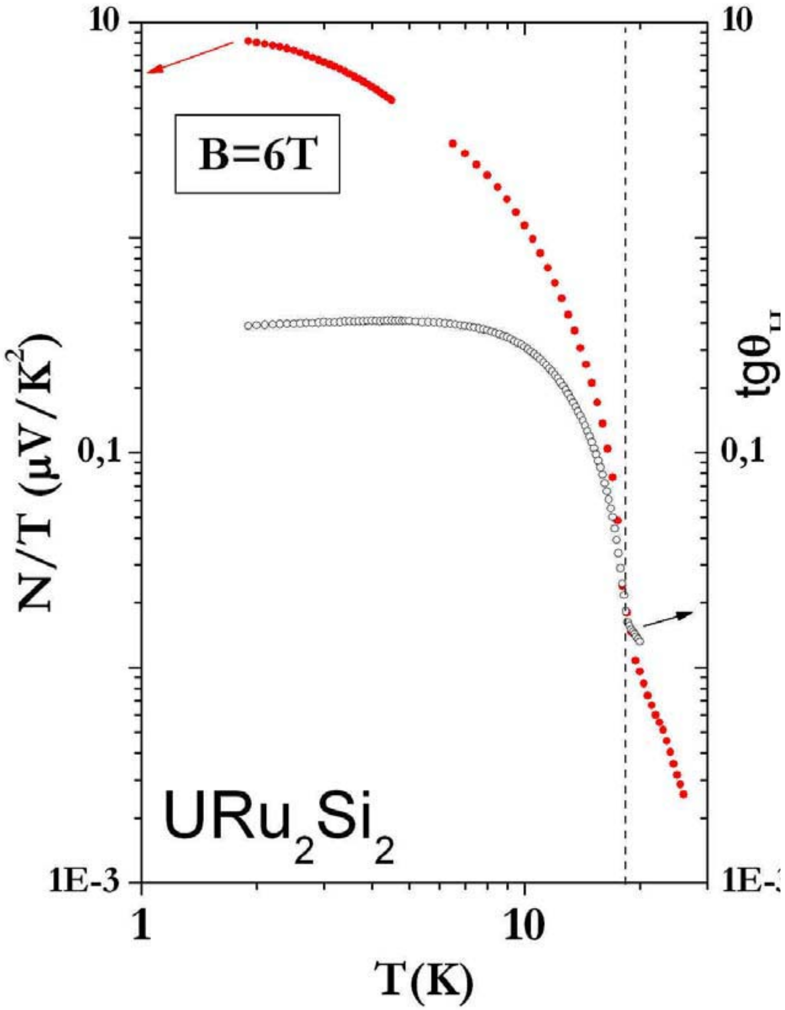
Quantum Critical Point at 35 T (Harisson,Kim, Jaime, Mydosh '03)

Correlation between the Nernst signal and the Hall angle

In a Boltzmann picture:

$$N = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \Theta_H}{\partial \epsilon}|_{\varepsilon F}$$

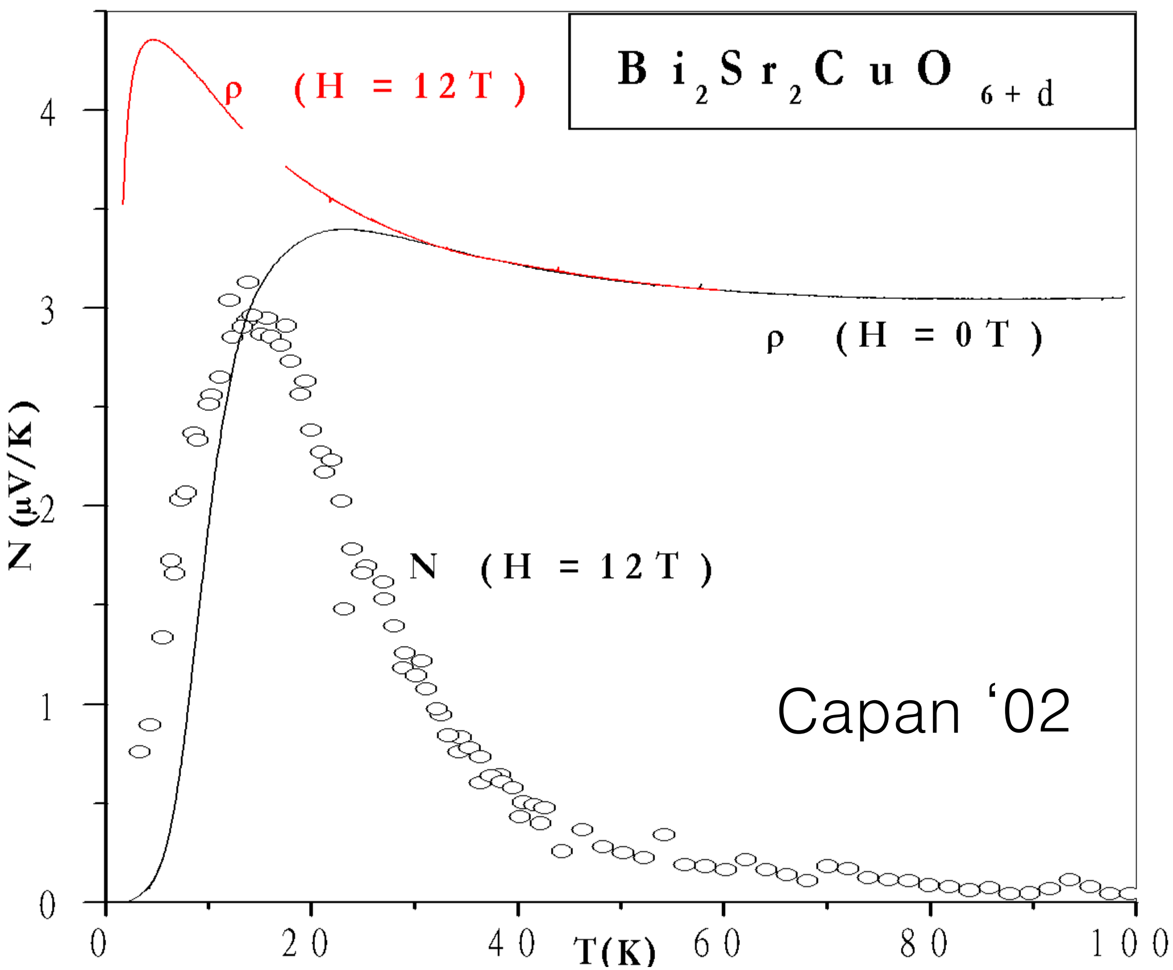
- Small ε_F in Heavy Fermions!
- Large θ_H in the hidden-order state!
- Could this combination explain the large N?



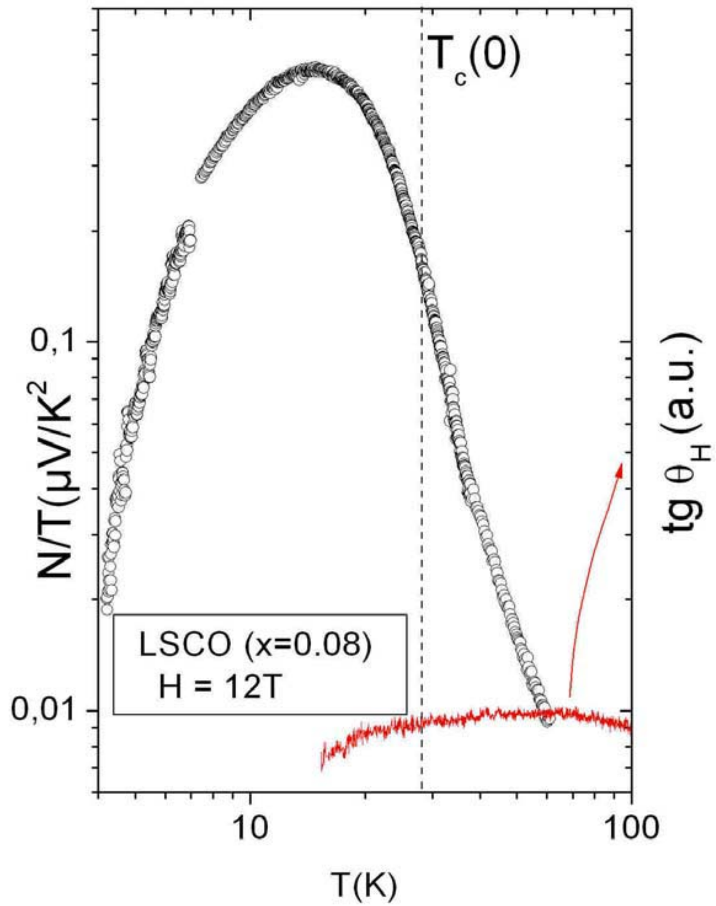
Only qualitatively:

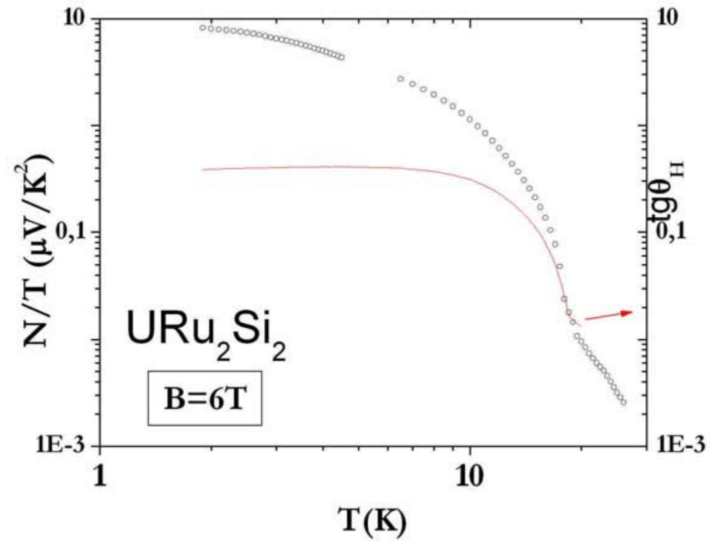
- Implies ε_F ~17K !
- Even when θ_H constant, N/T increases!
- N/S diverges!

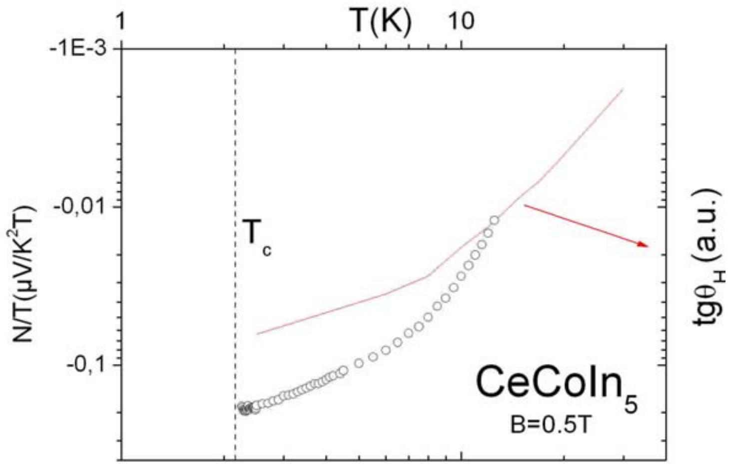
An order of magnitude larger than the vortex Nernst signal in cuprates



But, the anomalous Nernst signal in cuprates does NOT correlate with the Hall angle!







Superconducting fluctuations remain the most plausible source!

Summary

- It is instructive to look at the thermopower-to-specific heat ratio!

- In appropriate units, this ratio is close to unity for a wide range of compounds!

- Anomalously large Nernst signals $CeCoIn_5$ and URu_2Si_2 arise when this is not the case and point to a an unidentified source of transverse thermoelectricity!