



MAX-PLANCK-INSTITUT
FÜR CHEMISCHE PHYSIK FESTER STOFFE

Thermomagnetic properties of the strongly correlated semimetal CeNiSn

Niels Oeschler

Max Planck Institute for Chemical Physics of Solids, Dresden, Germany



MAX-PLANCK-INSTITUT
FÜR CHEMISCHE PHYSIK FESTER STOFFE

Acknowledgements:

Measurements:

U. Köhler, MPI CPfS, Dresden, Germany

P. Sun, MPI CPfS, Dresden, Germany

S. Paschen, Vienna University of Technology, Austria

F. Steglich, MPI CPfS, Dresden, Germany

Samples:

T. Takabatake, Hiroshima University, Japan



Outline

Introduction

Thermoel. and thermomagn. effects

Exp. setup

Correlated semimetal CeNiSn

Results

Resistivity and Hall effect

Thermopower

Nernst effect and Righi-Leduc effect

Discussion

Field-dependent thermopower

Nernst effect

Summary



Introduction

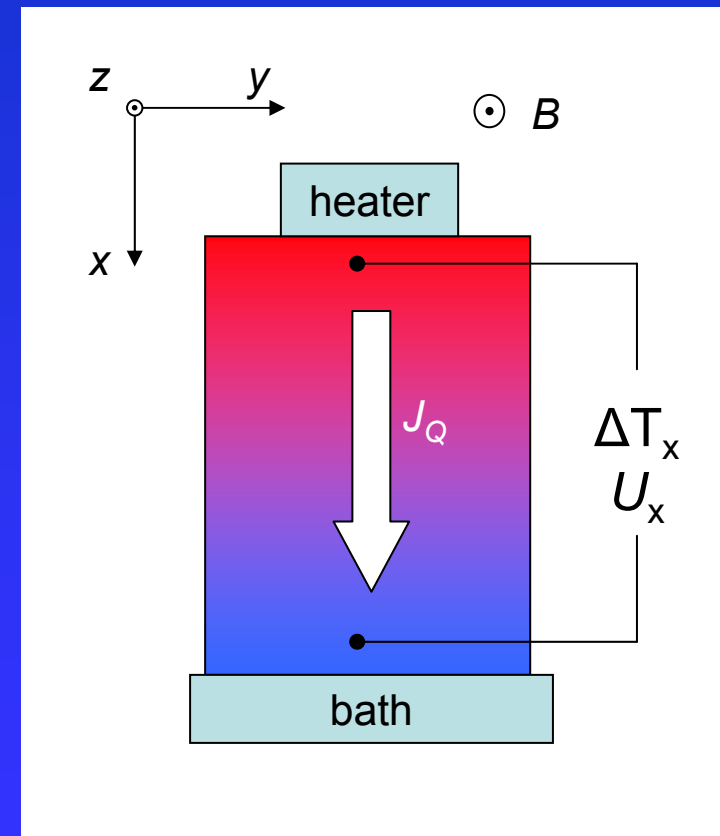
Thermoel. and thermomagn. Effects

Charge transport: $J = \sigma E - \sigma S \Delta T$

Heat transport: $J_Q = \sigma S T E - \kappa \Delta T$

Thermal conductivity: $\kappa = J_Q / \Delta T_x$

Thermopower: $S = -U_x / \Delta T_x$





Introduction

Thermoel. and thermomagn. Effects

Charge transport: $J = \sigma E - \sigma S \Delta T$

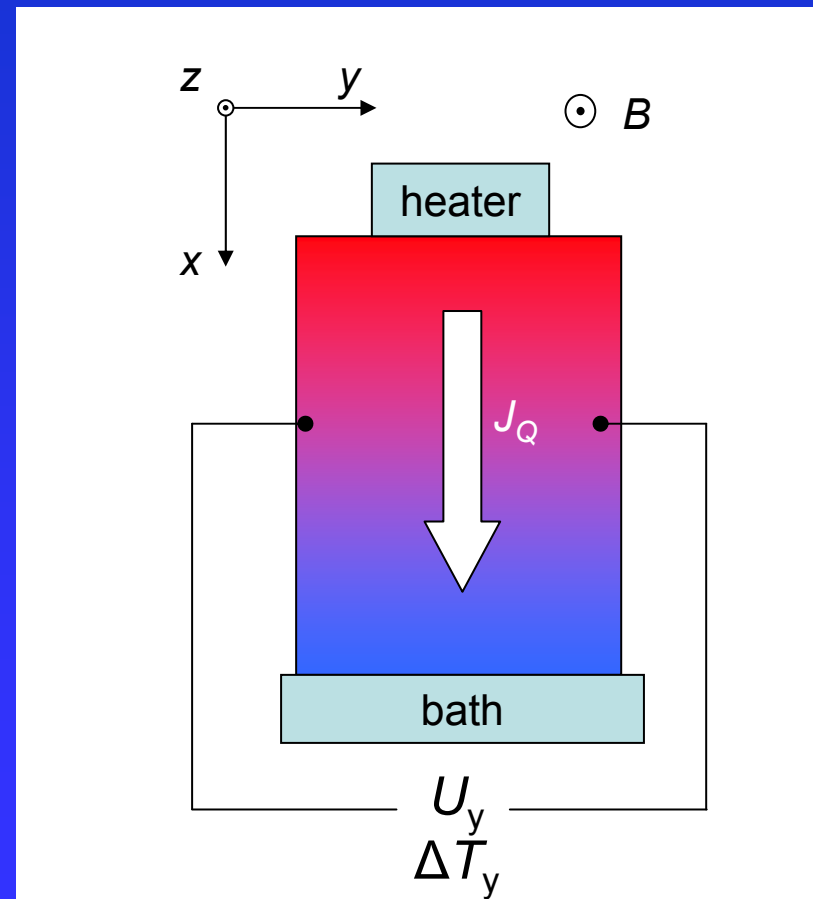
Heat transport: $J_Q = \sigma S T E - \kappa \Delta T$

Thermal conductivity: $\kappa = J_Q / \Delta T_x$

Thermopower: $S = -U_x / \Delta T_x$

Nernst effect: $v = -U_y / \Delta T_x B$

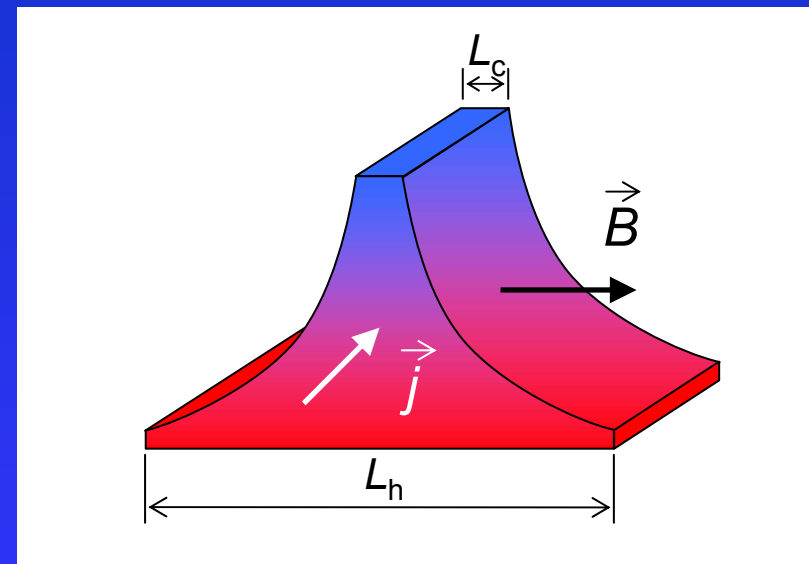
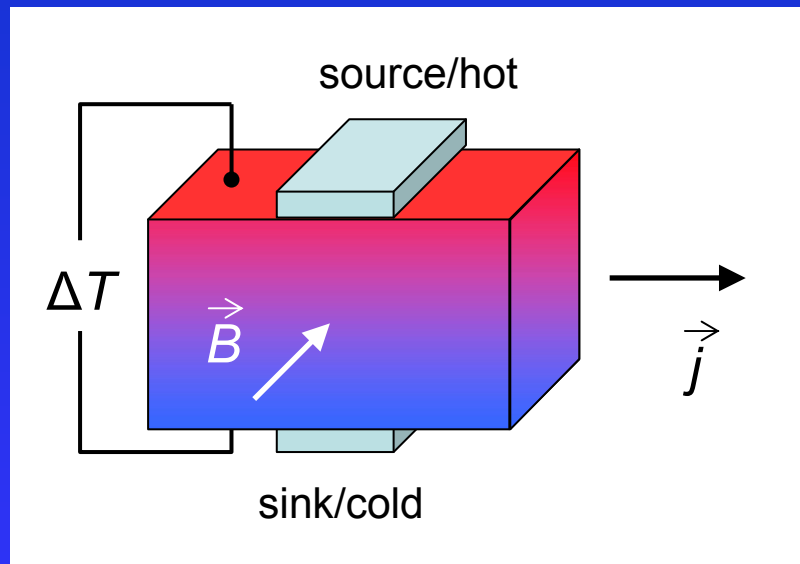
Righi-Leduc effect: $L = -\kappa_y / B \Delta T_y / \Delta T_x$





Introduction

Thermomagn. Effects: Ettingshausen cooling



Thermomagn. figure of merit $Z_{\text{mag}} T$

$$\Delta T_{\text{max}} = \frac{Z_{\text{mag}} T_{\text{cold}}^2}{2}$$

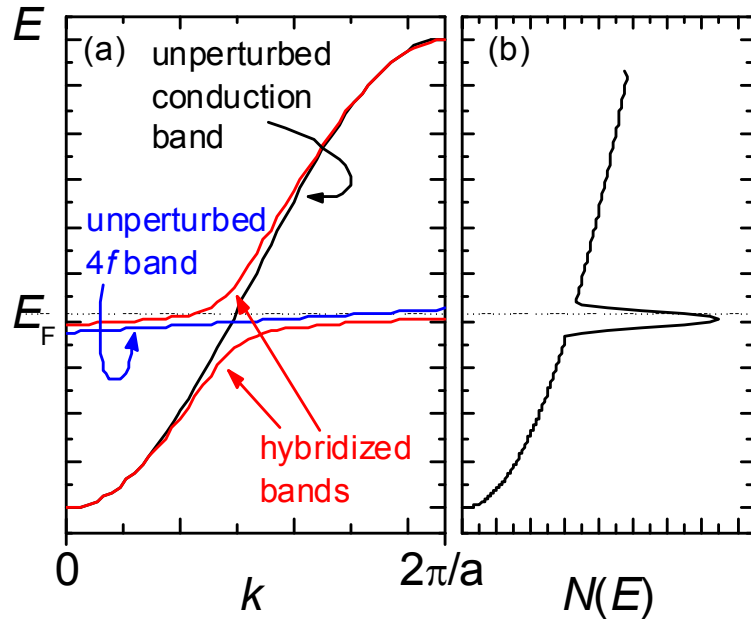
$$Z_{\text{mag}} T = \frac{\sigma(\nu B)^2 T}{\kappa}$$

infinite stage Ettingshausen device

$$\Delta T_{\text{max}} = T_{\text{hot}} \left(1 - \frac{L_c}{L_h} \right)^\alpha$$

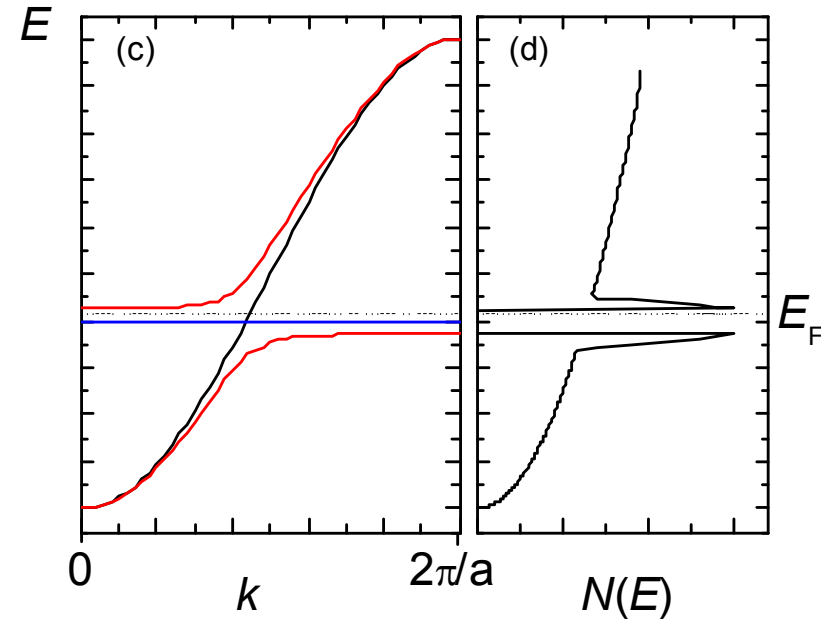


Kondo Insulator



Heavy fermion metals

- $\rho \sim -\ln T$ at $T \approx T_K$
- enhanced DOS at E_F below $\sim T_K$
- metal-like behavior at low T



Kondo insulator

- $\rho \sim -\ln T$ at $T \approx T_K$
- gap below $\sim T_g$
- insulating behavior at low T

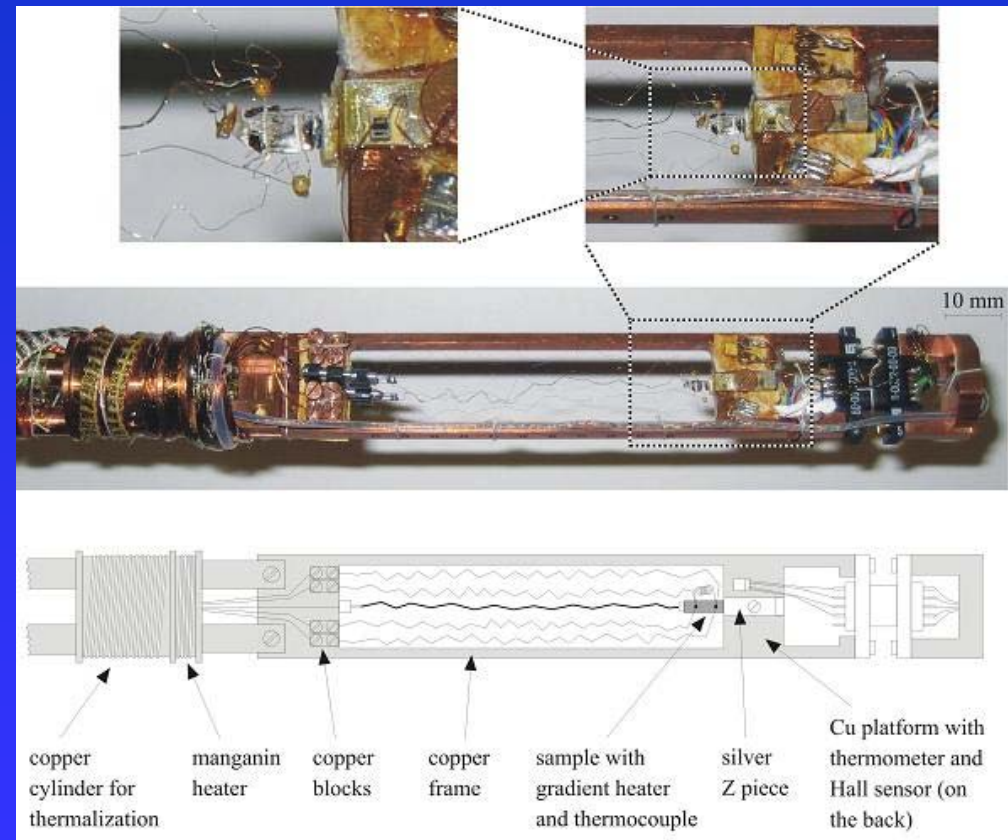


Introduction

Experimental Setup:

^4He cryostat
horizontal 7T magnet
optimized for small samples with
low κ

ΔT :
chromel-AuFe thermocouples
U:
copper wires, nanovoltmeter





CeNiSn

samples

orthorhombic crystal structure
chains of Cerium ions along a
easy a axis

Czochralski method annealed by SSE

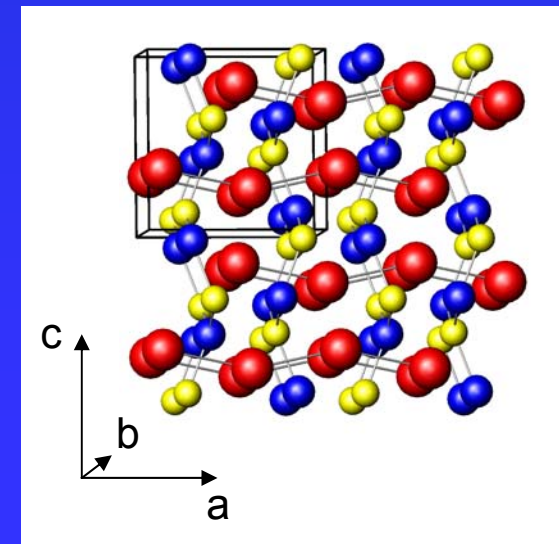
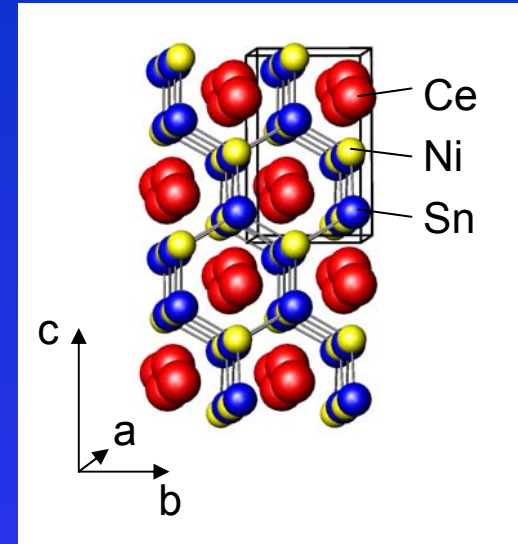
energy scales

crystal field levels: $k_B\Delta_{\text{CEF}} \approx 230\text{K}, 460\text{K}$

Kondo temp.: $T_K \approx 56\text{K}$

pseudogap $\Delta/k_B \approx 10\text{K}$ below $T \approx 10\text{K}$

no ordering down to 25 mK





CeNiSn

gap structure

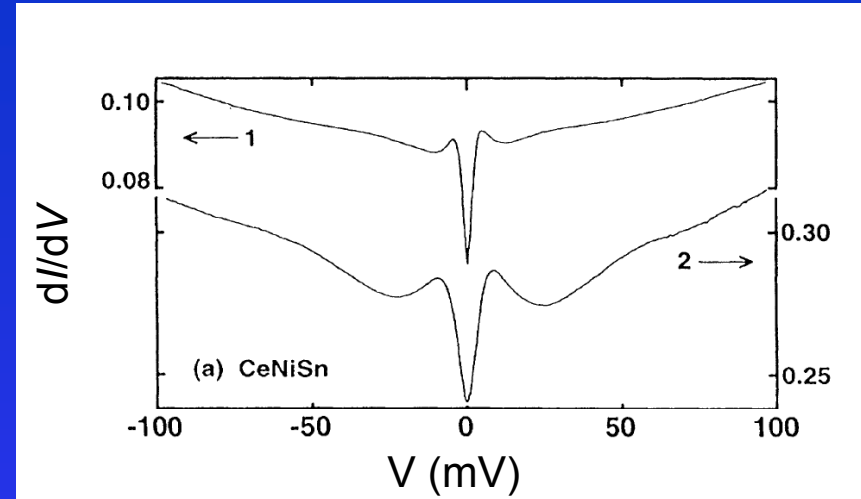
pseudogap opens around 10K

residual states at E_F

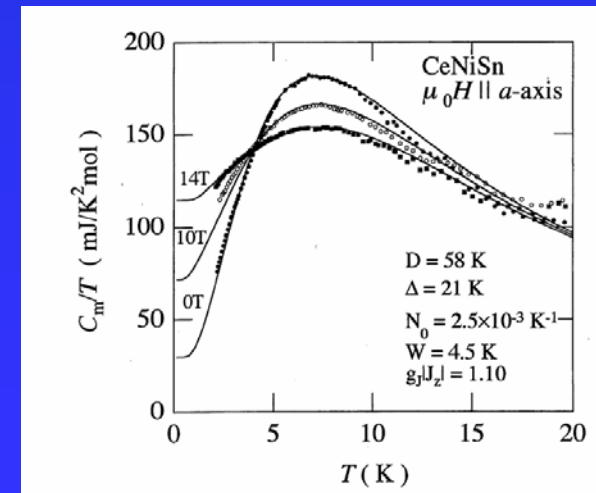
metallic ρ , large Sommerfeld coeff.

gap suppression

- magnetic fields ~ 10 T // a
- pressure ~ 2 GPa
- substitution (Ce/La and Ni/Cu,Co) ~ 10 %



T. Ekino et al., *Phys. Rev. Lett.* **75**, 4262 (1995)



K. Izawa et al., *J. Phys. Soc. Jpn.* **65**, 3119 (1996)

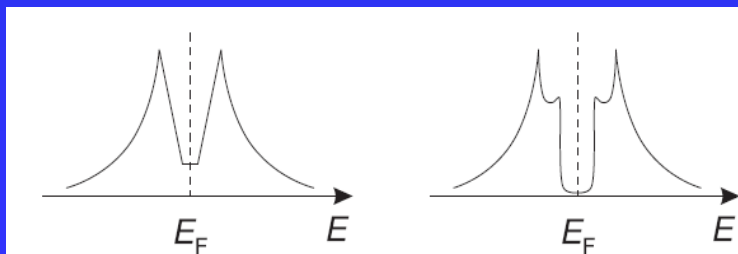


CeNiSn - Kondo Insulator?

sensitive dependence on sample purity
(ρ , MR , R_H)

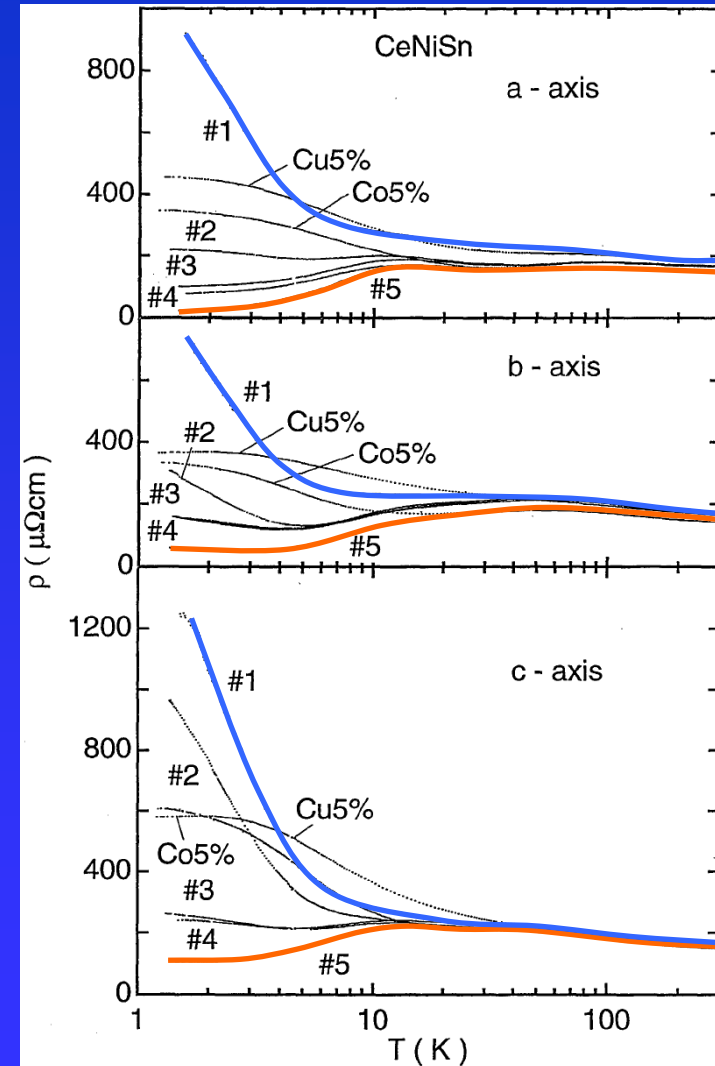
residual DOS near E_F (NMR, c_P , ρ , κ)

⇒ **CeNiSn – Kondo semimetal**



K. Izawa et al., *J. Phys. Soc. Jpn.* **65**, 3119 (1996)

H. Ikeda and K. Miyake, *J. Phys. Soc. Jpn.* **65**, 1769 (1996)



G. Nakamoto et al., *J. Phys. Soc. Jpn.* **64**(12), 4834 (1995)



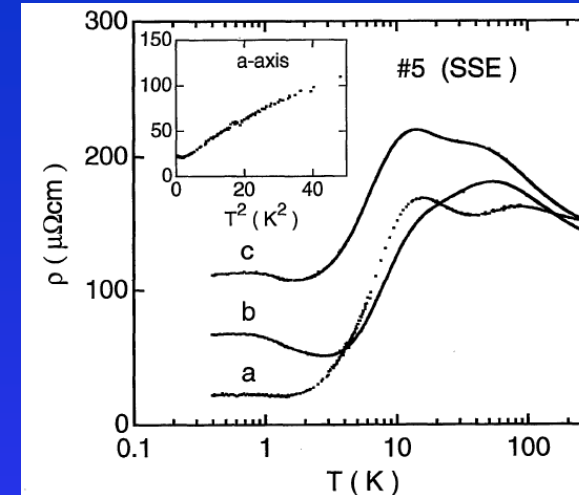
Experimental

Single crystals (#5)

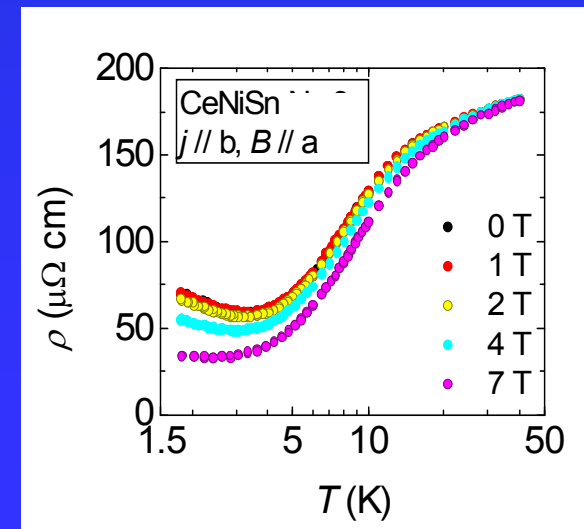
- Czochralski + SSE
- best available samples
- Orientation: Laue, χ , ρ (a/c)
- ca. $4 \times 4 \times 0.8 \text{ mm}^3$
- Measurements: $q \parallel b$; $B \parallel a, c$

Measurements:

- Thermal conductivity
- Thermopower at +B and -B
- Nernst effect
- Thermal Hall effect

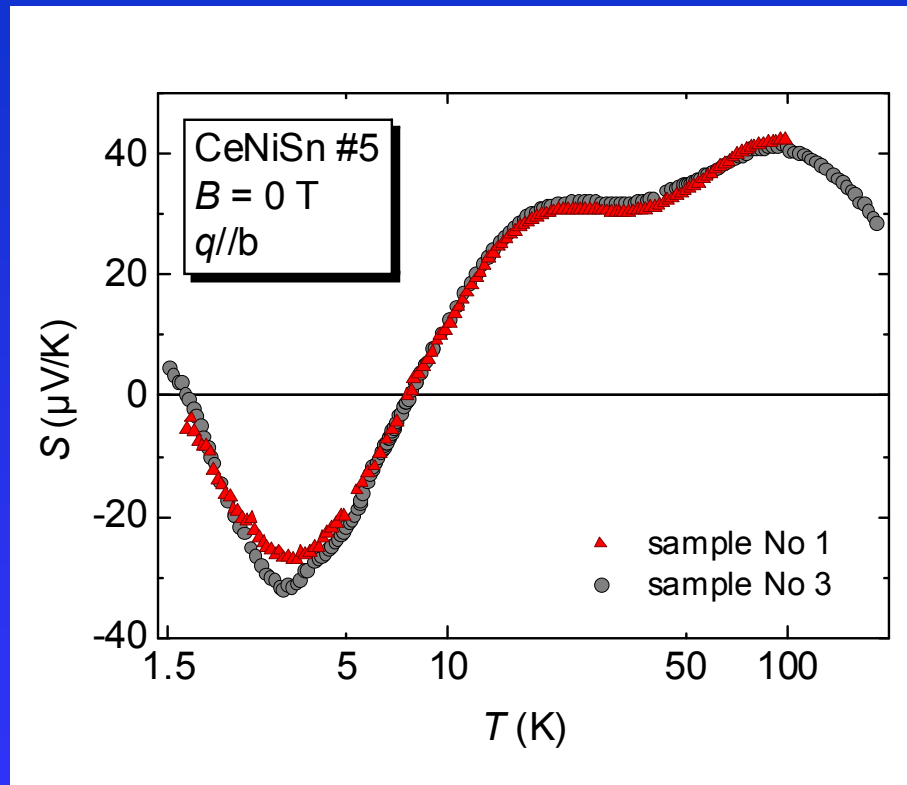


G. Nakamoto et al., *J. Phys. Soc. Jpn.* **64**(12), 4834 (1995)



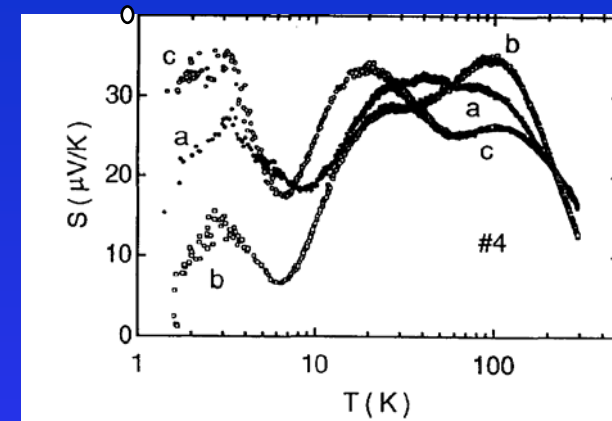


Results: Thermopower



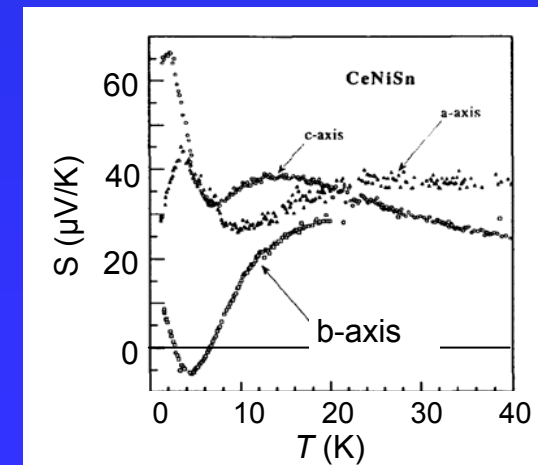
- Kondo system with CEF splitting
- largest negative S ever observed
- very precise orientation !

CeNiSn #4



G. Nakamoto et al., *Physica B* 306 & 307, 840 (1995)

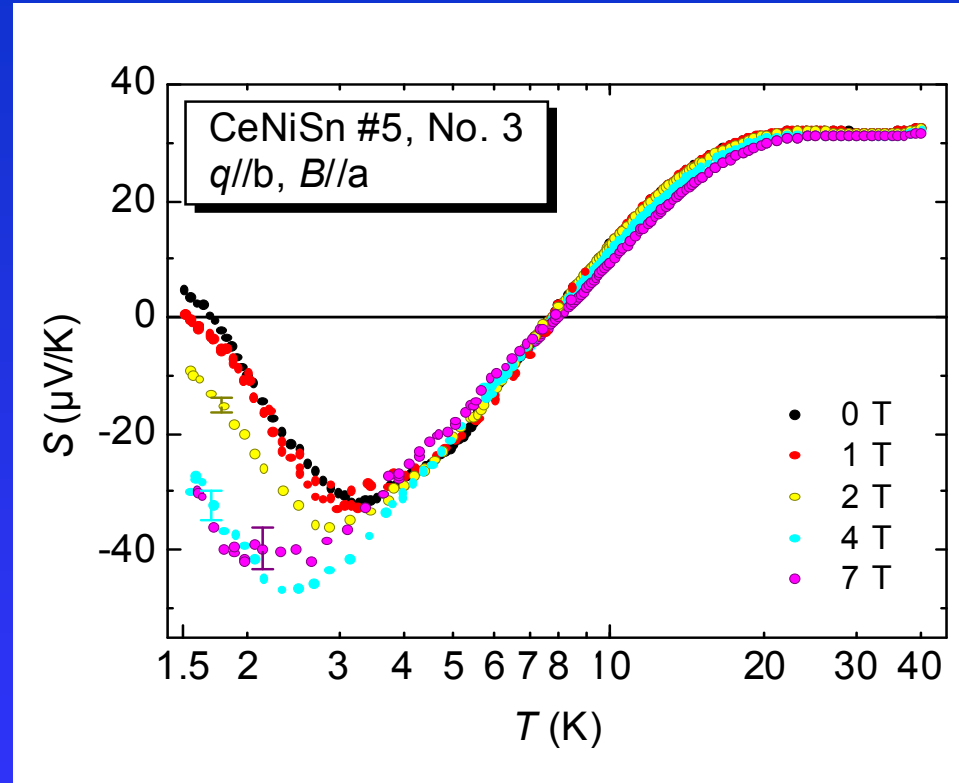
CeNiSn type unknown



J. Sakurai et al., *Physica B* 306 & 307, 834 (1995)



Field-dep. Thermopower



literature:

- strong sample dependence at low T
- no comparable results in field

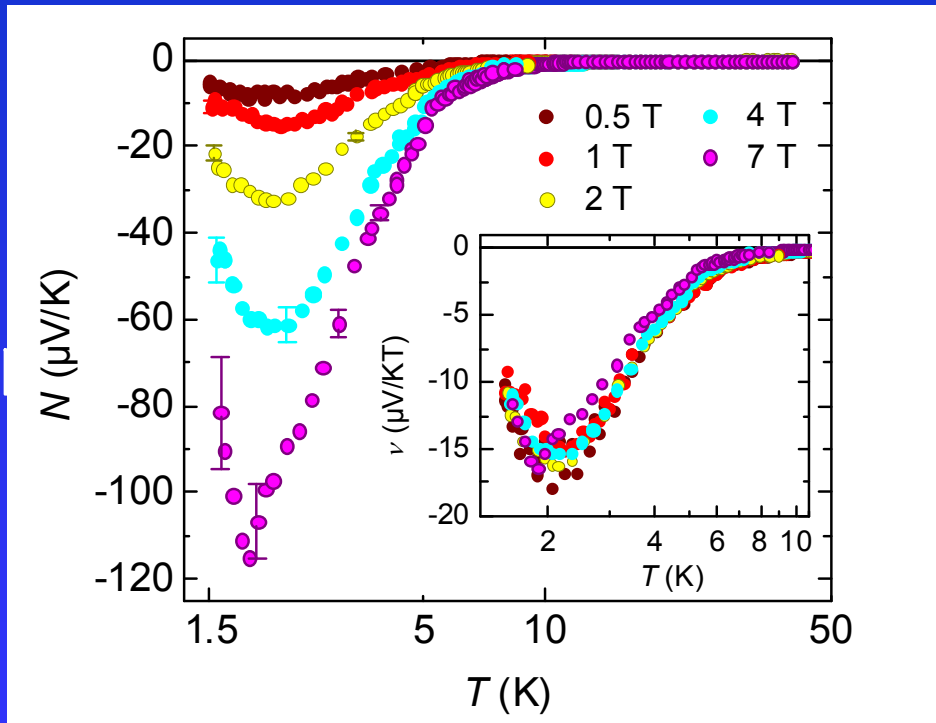
$B // a$: - enhanced values of $|S|$
- shift of the minimum to lower T

$B // c$: - similar, but less pronounced

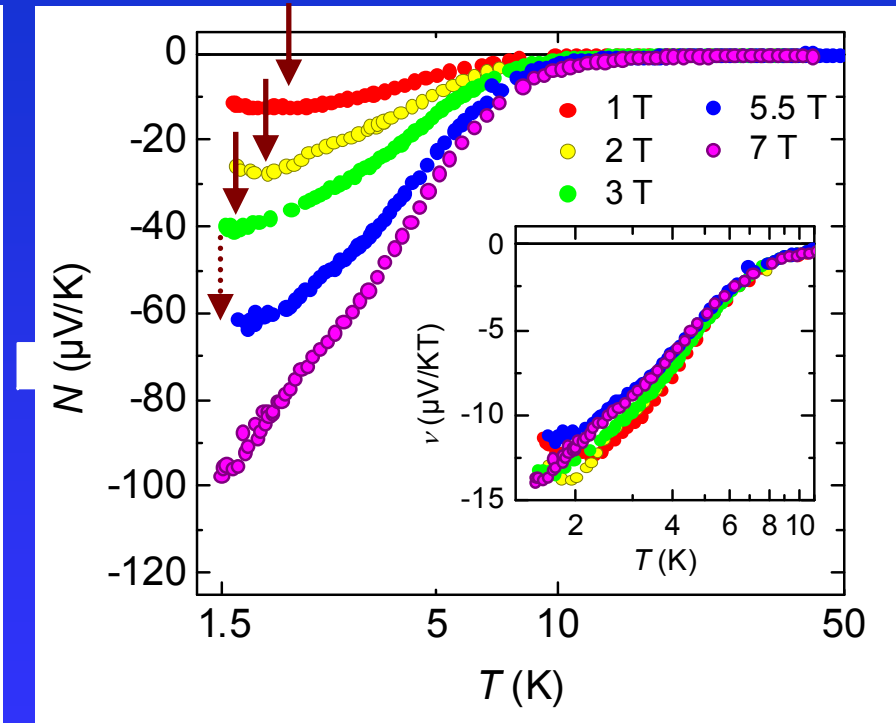


Nernst effect

$q // b, B // a$



$B // c$



large values of N below 10 K (opening of the gap)

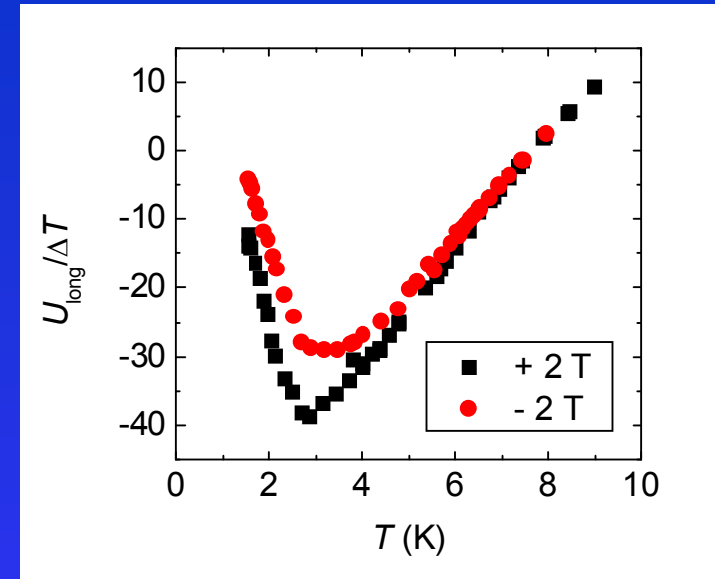
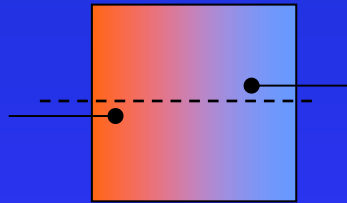
- scaling for $B // a$ (easy axis!)
- shift of minimum for $B // c$



Discussion: Thermopower

variation due to

- sample dependence
- misorientation
- non-negligible Nernst contribution

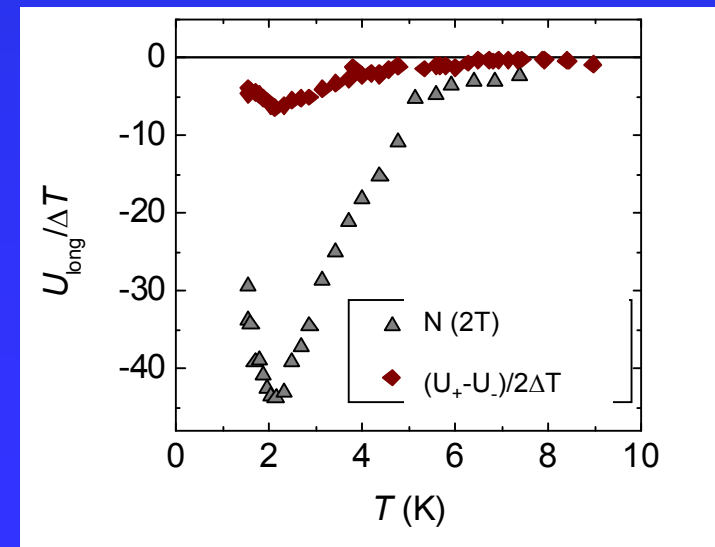
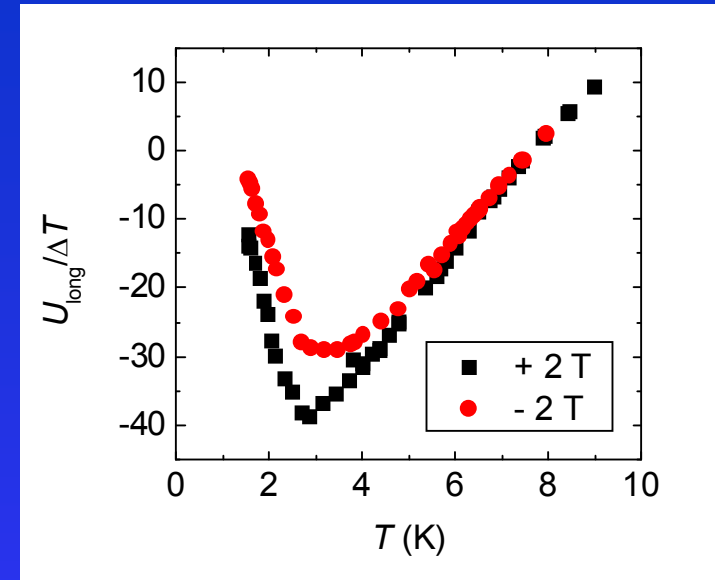
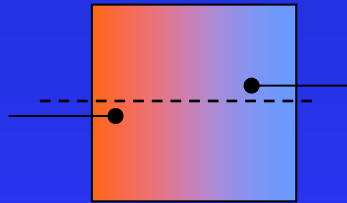




Discussion: Thermopower

variation due to

- sample dependence
- misorientation
- non-negligible Nernst contribution

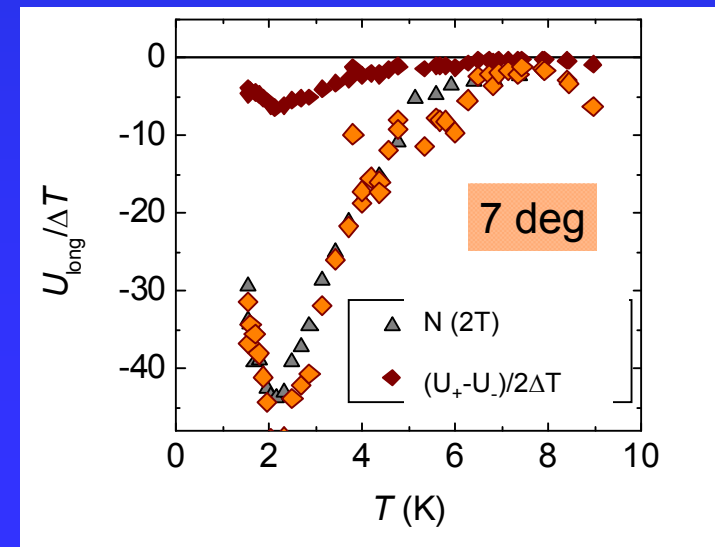
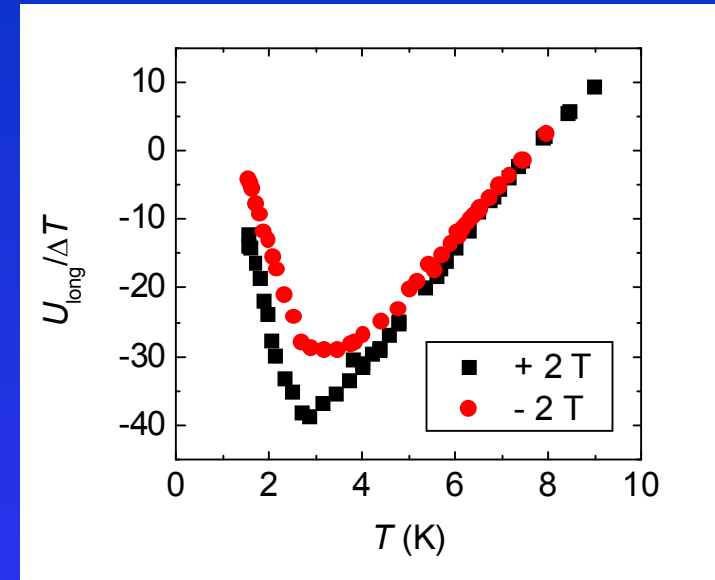
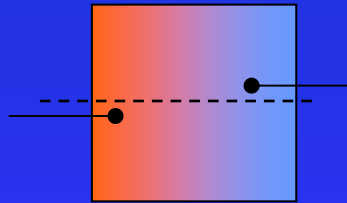




Discussion: Thermopower

variation due to

- sample dependence
- misorientation
- non-negligible Nernst contribution

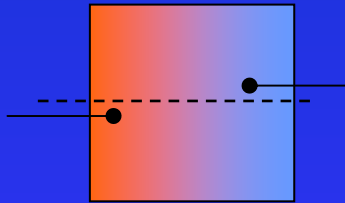




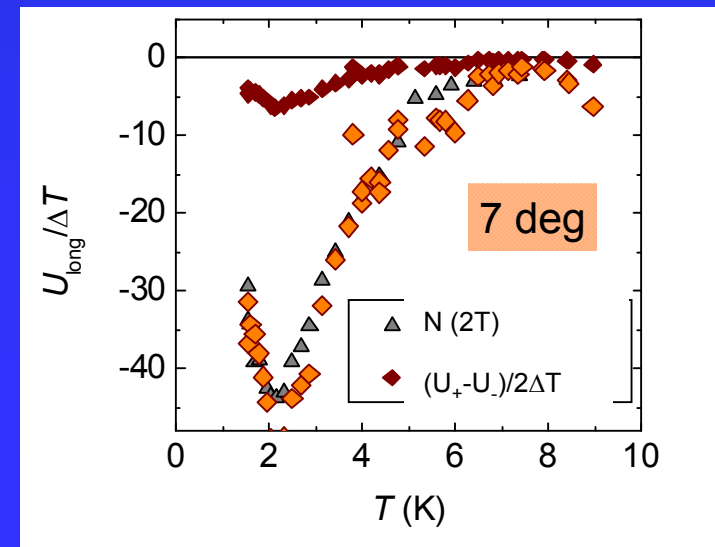
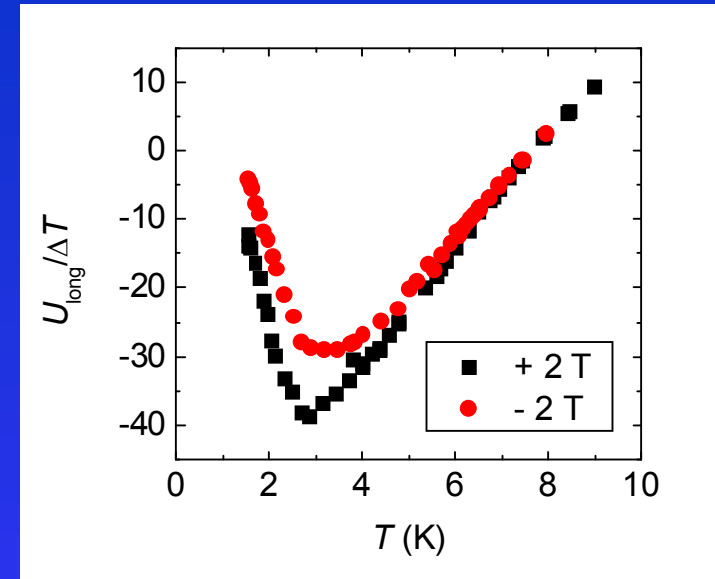
Discussion: Thermopower

variation due to

- sample dependence
- misorientation
- non-negligible Nernst contribution

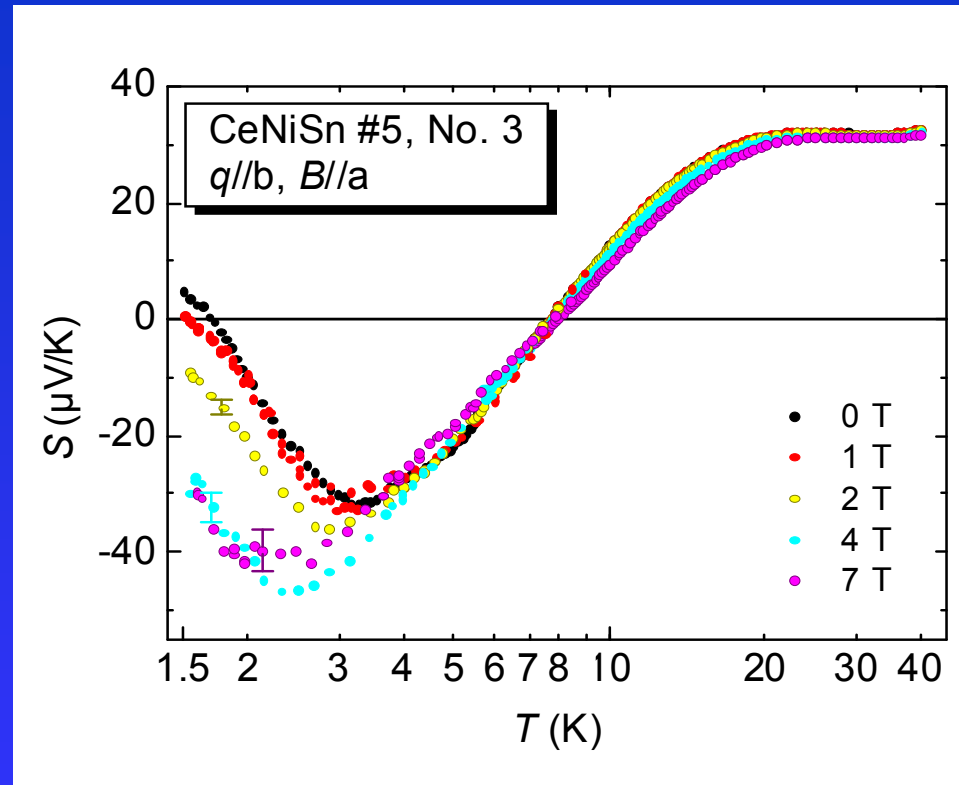


- first systematic study of $S(T, B)$ including:
- best available samples
 - precise orientation
 - correction for the Nernst signal





Field-dep. Thermopower



literature:

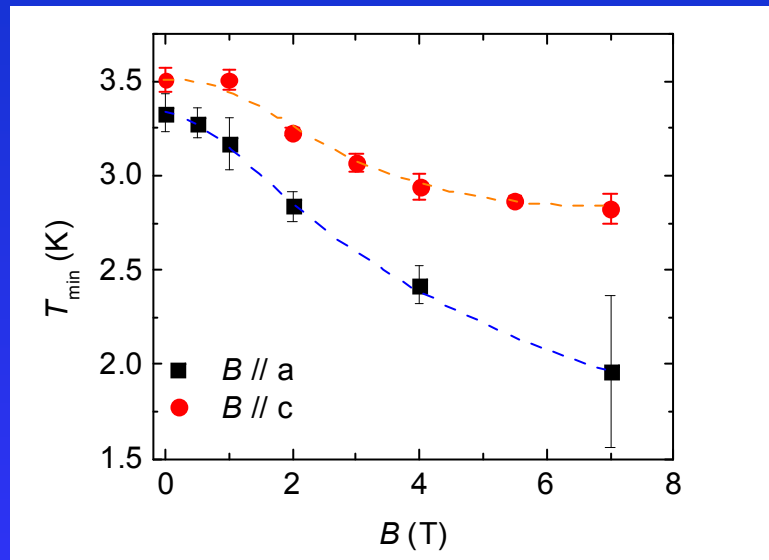
- strong sample dependence at low T
- no comparable results in field

- $B // a$: - enhanced values of $|S|$
- shift of the minimum to lower T
- $B // c$: - similar, but less pronounced



Discussion: Thermopower

position of the minimum

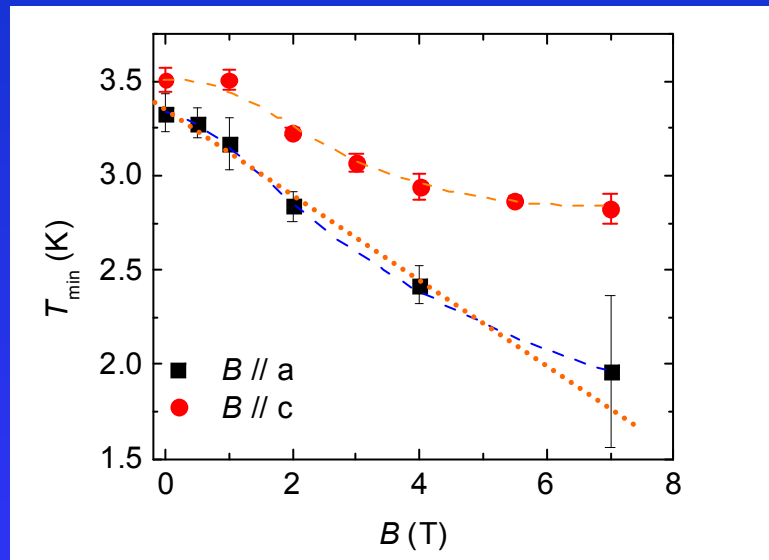


- effect larger for $B // a$



Discussion: Thermopower

position of the minimum

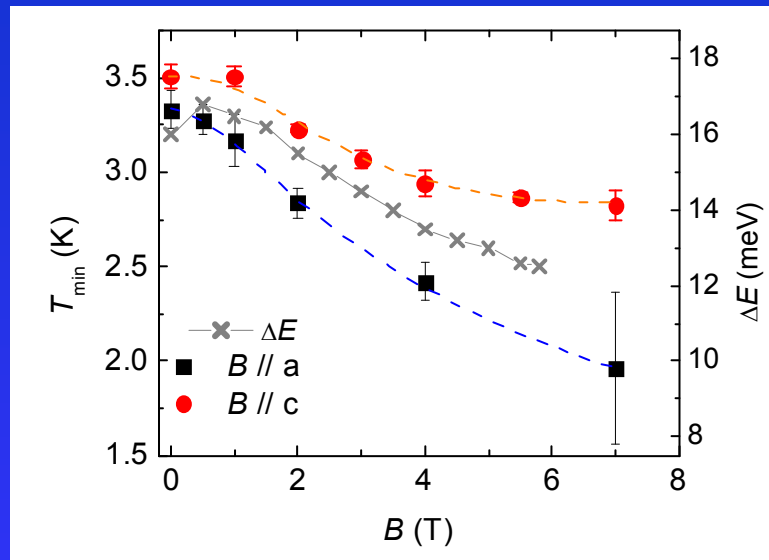


- effect larger for $B // a$
- extrapolation $// a$: $B_c = 14$ T
(MR: 18 T)
- shift \sim closing of the gap



Discussion: Thermopower

position of the minimum

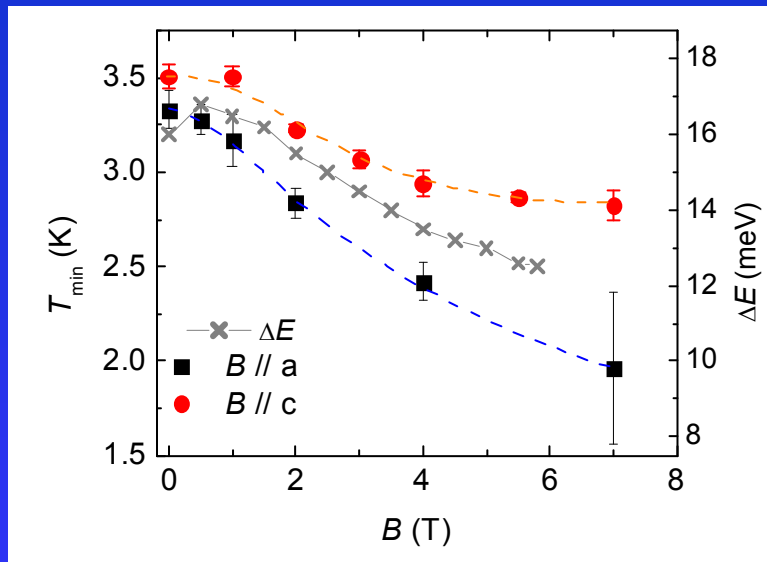


- effect larger for $B // a$
- extrapolation $// a$: $B_c = 14$ T
(MR: 18 T)
- shift \sim closing of the gap
(ΔE from tunneling spectroscopy)



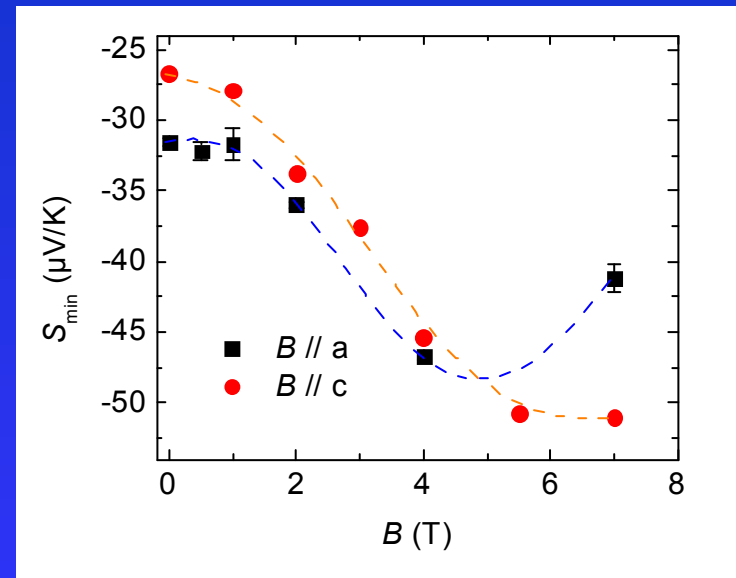
Discussion: Thermopower

position of the minimum



- effect larger for $B // a$
- extrapolation // a: $B_c = 14$ T (MR: 18 T)
- shift ~ closing of the gap (ΔE from tunneling spectroscopy)

value at the minimum

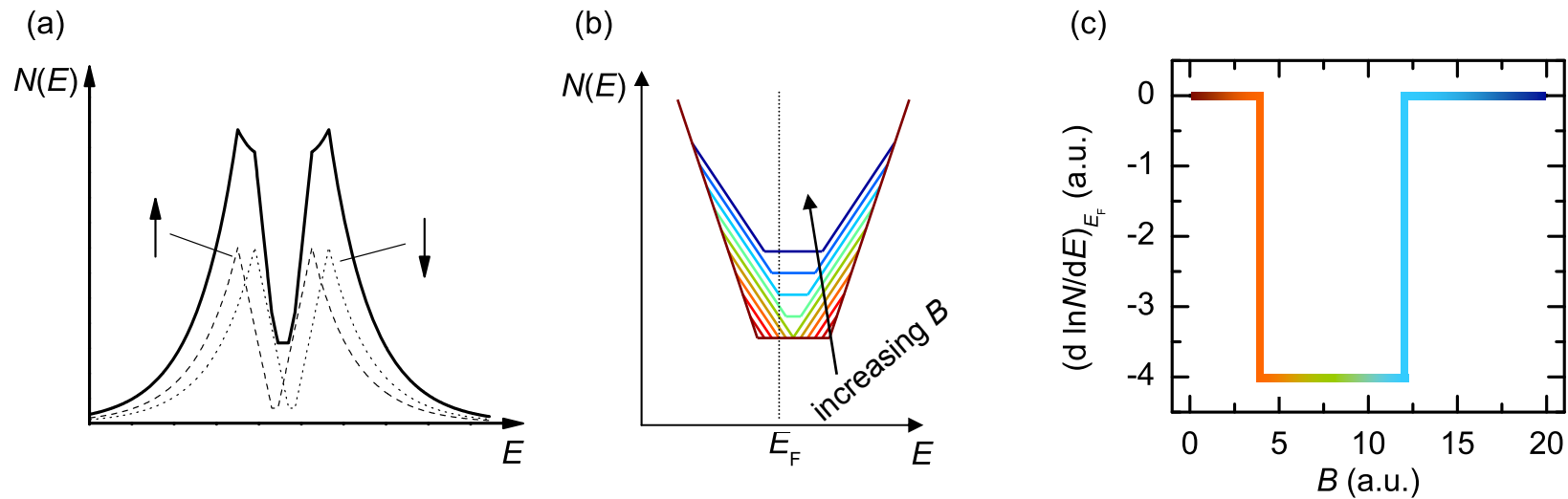


- effect larger for $B // a$
- change of the DOS near E_F due to Zeeman splitting (c_p)
- similar results for $S(T)$ at low T



Discussion: Thermopower

V-shaped DOS in field



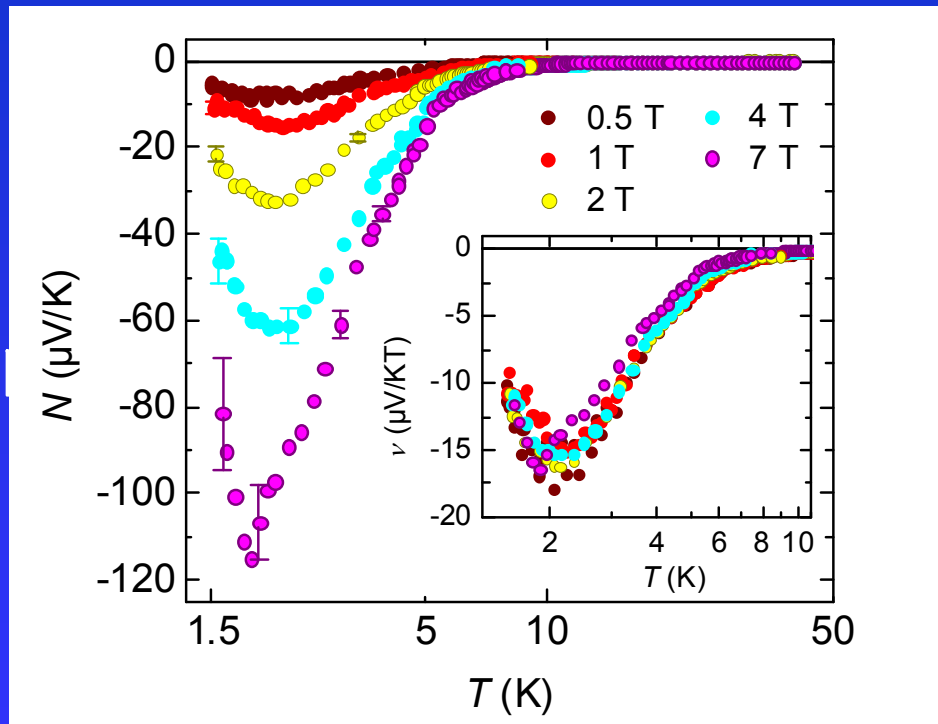
similar analysis for $C_p(T, B)$
(enhanced γ value in field)

$$S \propto \left. \frac{\partial \ln N}{\partial \varepsilon} \right|_{E_F}$$

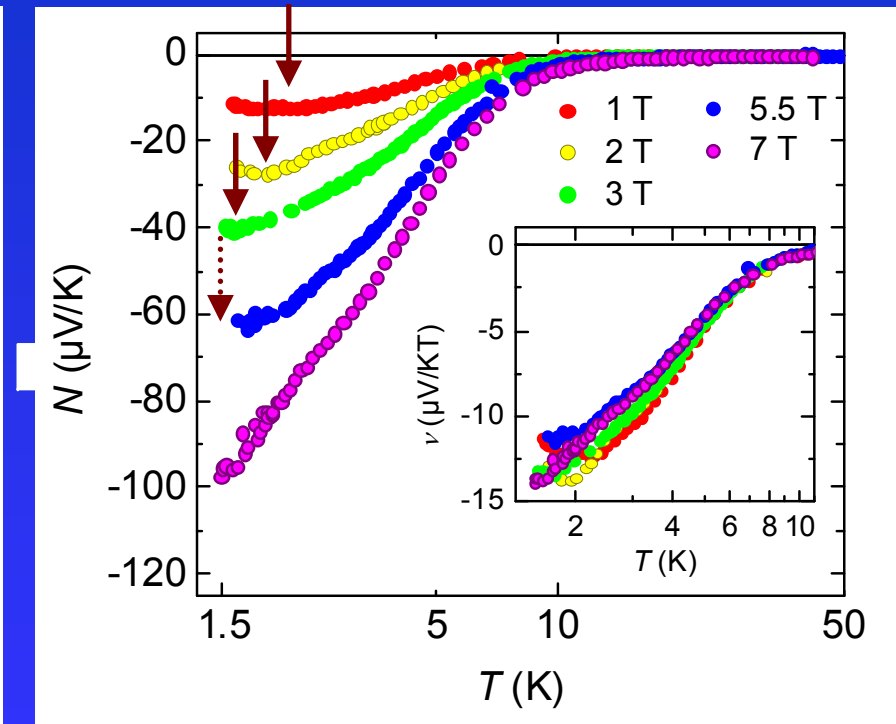


Results: Nernst effect

$q // b, B // a$



$B // c$



large values of N below 10 K (opening of the gap)

- scaling for $B // a$ (easy axis!)

- shift of minimum for $B // c$

→ open question: weak sensitivity to magnetic fields $// a$



Discussion: Nernst effect

Nernst effect:

$$v^a = v^n - \varepsilon_{yy}/\kappa_{yy} L_{xy}$$

v^n : normal Nernst coeff.

v^a : adiabatic Nernst coeff.

due to transverse temp. gradient

For CeNiSn, $\Delta T_y \approx 0$, $L_{xy} \approx 0$

(below resolution limit)

$$\rightarrow v^a = v^n$$

Boltzmann approximation:

$$v^n = \frac{\pi^2}{3} \frac{k_B^2 T}{m^*} \left. \frac{\partial \tau}{\partial \varepsilon} \right|_{E_F}$$

with Hall angle

$$\tan \Theta_H = \frac{\sigma_{yx}}{\sigma_{xx}}$$

$$v^n \approx \frac{\pi^2}{3} \frac{k_B^2 T}{Be} \frac{\tan \Theta_H}{E_F}$$

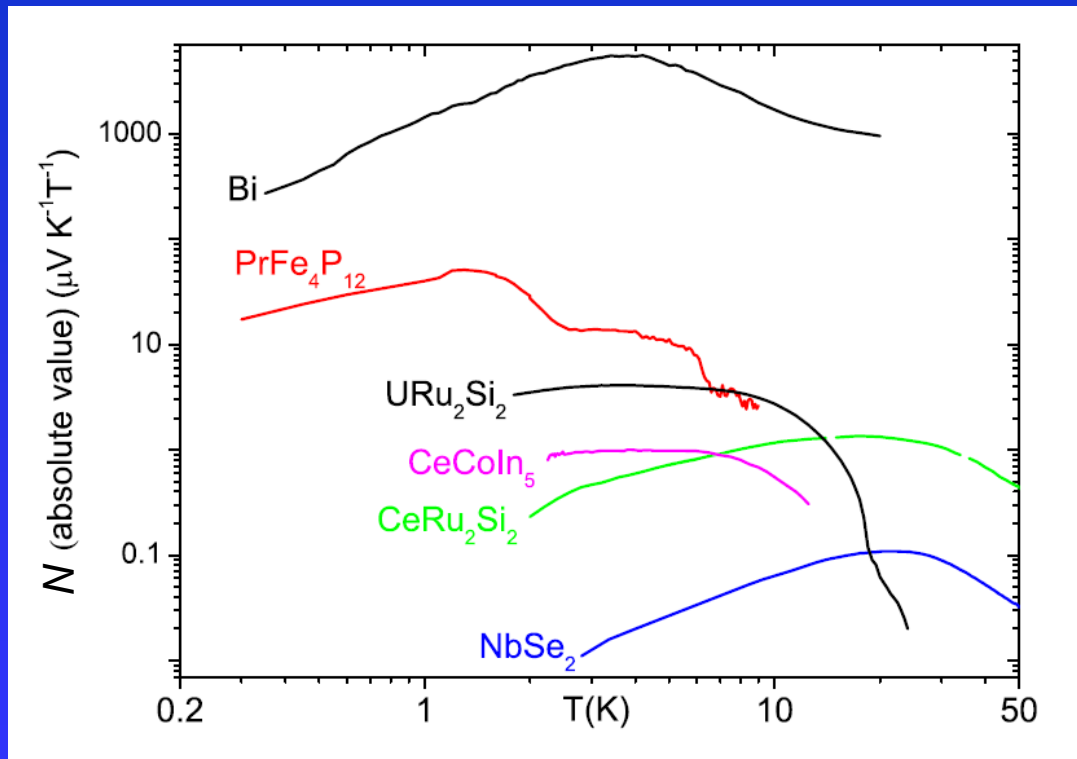
Large Nernst coefficient:

- Low charge carrier concentration
- Small Fermi energy



Discussion: Nernst effect

How to obtain large Nernst coefficients?

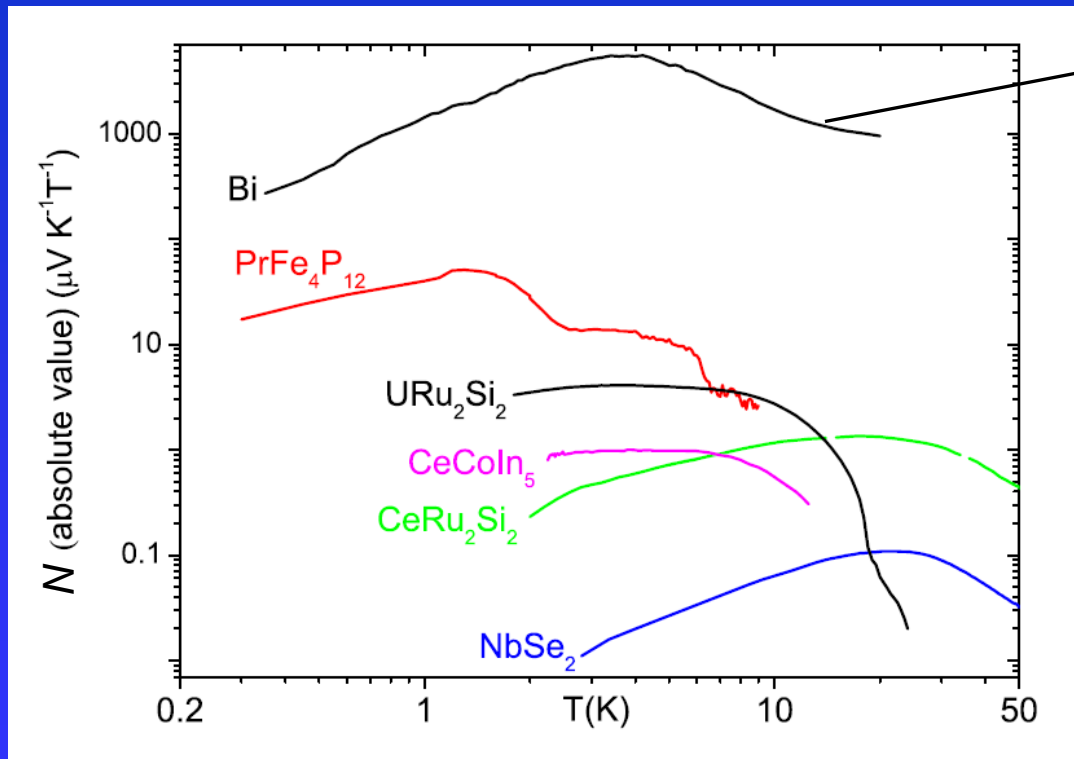


Behnia et al., *Phys. Rev. Lett.* **98**, 076603 (2007)



Discussion: Nernst effect

How to obtain large Nernst coefficients?



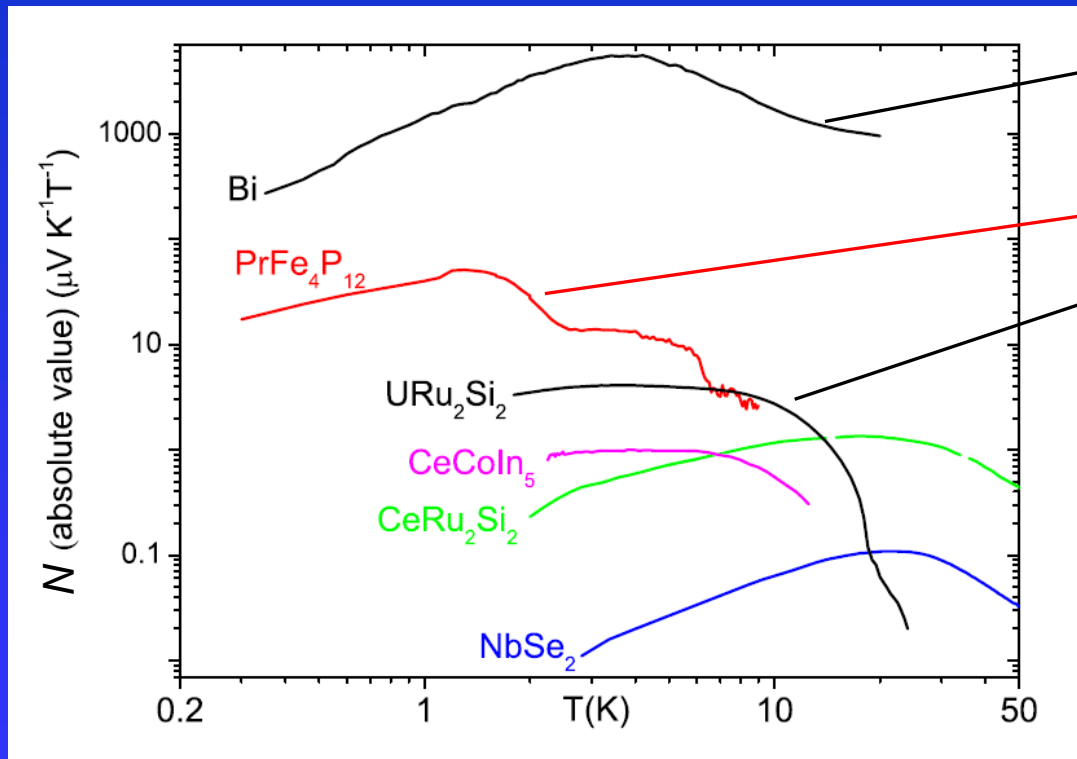
Bi normal semimetal

Behnia et al., *Phys. Rev. Lett.* **98**, 076603 (2007)



Discussion: Nernst effect

How to obtain large Nernst coefficients?



Bi normal semimetal

$\text{PrFe}_4\text{P}_{12}$

correlations,
unconventional
order

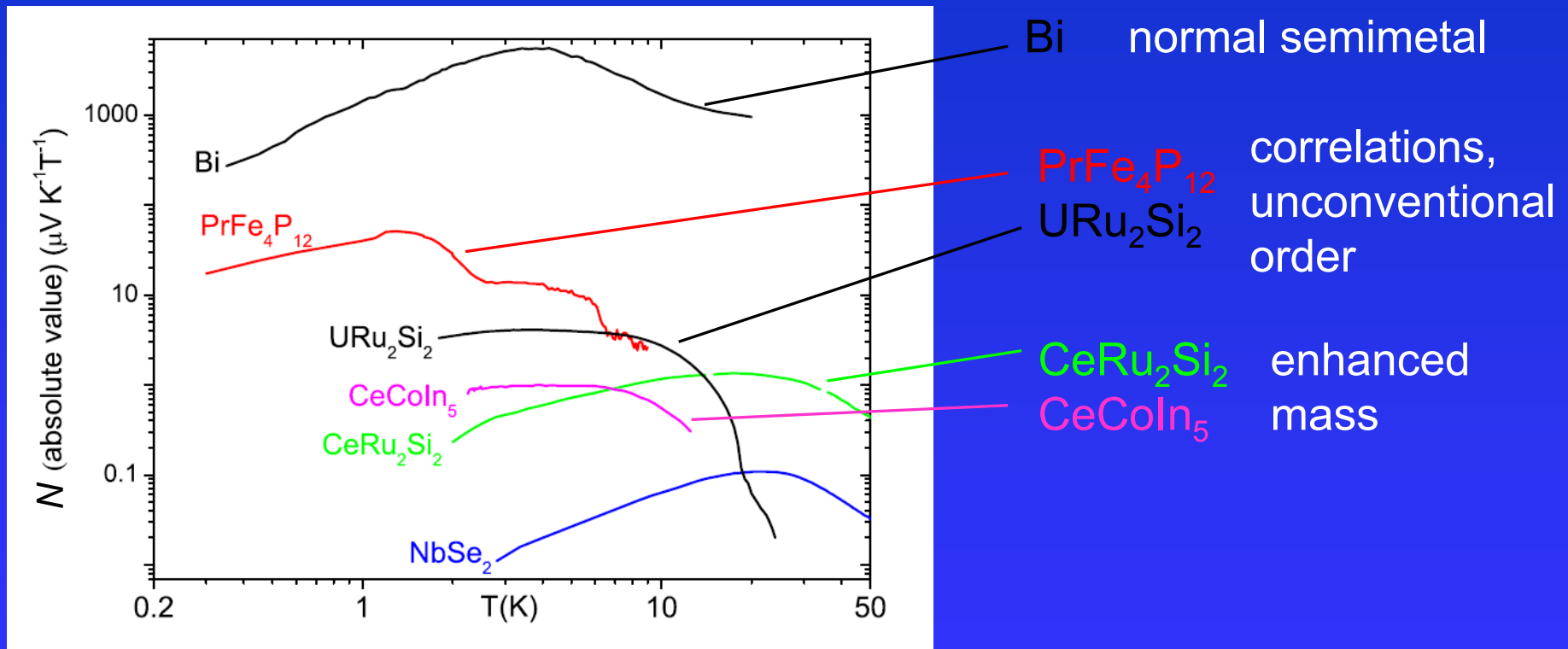
URu_2Si_2

Behnia et al., *Phys. Rev. Lett.* **98**, 076603 (2007)



Discussion: Nernst effect

How to obtain large Nernst coefficients?

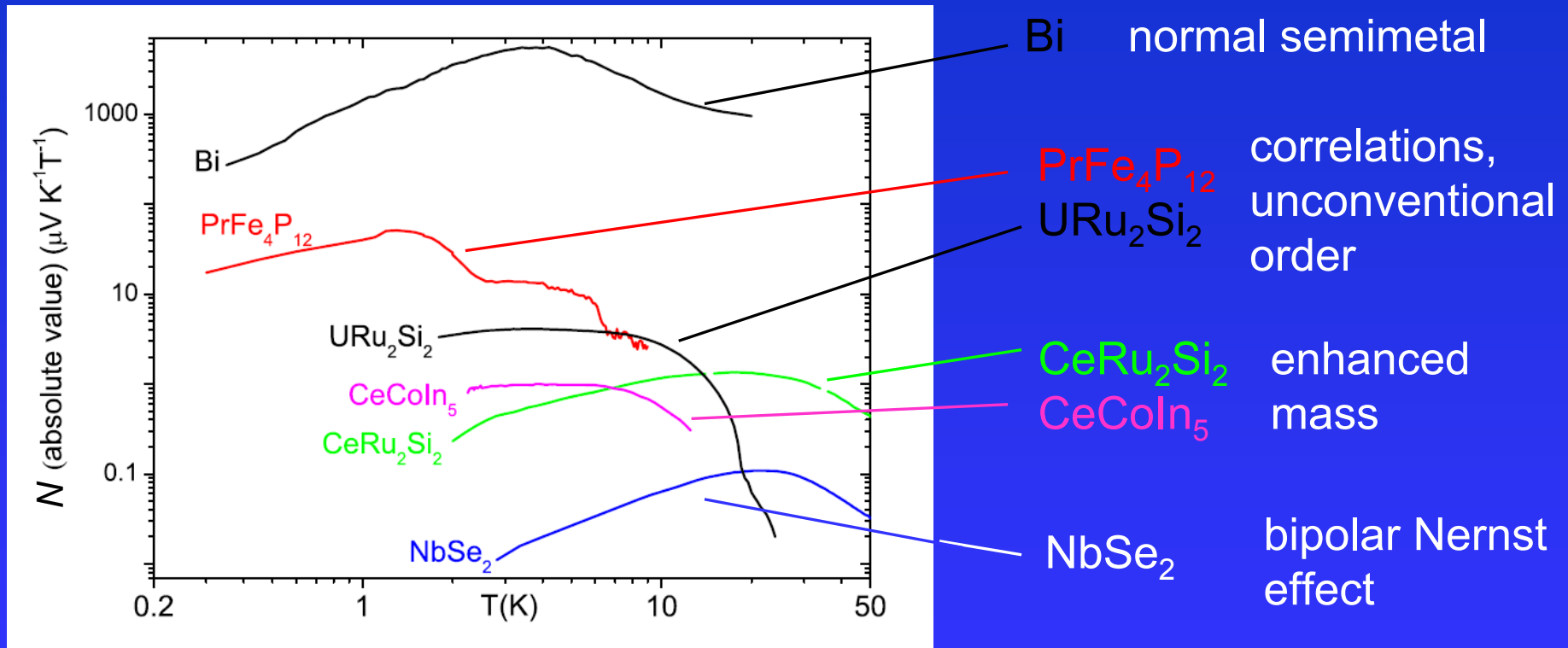


Behnia et al., *Phys. Rev. Lett.* **98**, 076603 (2007)



Discussion: Nernst effect

How to obtain large Nernst coefficients?



Behnia et al., *Phys. Rev. Lett.* **98**, 076603 (2007)



Discussion: Nernst effect

Boltzmann approximation:

$$N^n = \frac{\pi^2}{3} \frac{k_B^2 T}{m^*} \left. \frac{\partial \tau}{\partial \varepsilon} \right|_{E_F}$$

with Hall angle

$$\tan \Theta_H = \frac{\sigma_{yx}}{\sigma_{xx}}$$

$$N^n \approx \frac{\pi^2}{3} \frac{k_B^2 T}{Be} \frac{\tan \Theta_H}{E_F}$$

BUT: CeNiSn with two types of charge carriers



Discussion: Nernst effect

Boltzmann approximation:

$$N^n = \frac{\pi^2}{3} \frac{k_B^2 T}{m^*} \left. \frac{\partial \tau}{\partial \varepsilon} \right|_{E_F}$$

with Hall angle

$$\tan \Theta_H = \frac{\sigma_{yx}}{\sigma_{xx}}$$

$$N^n \approx \frac{\pi^2}{3} \frac{k_B^2 T}{Be} \frac{\tan \Theta_H}{E_F}$$

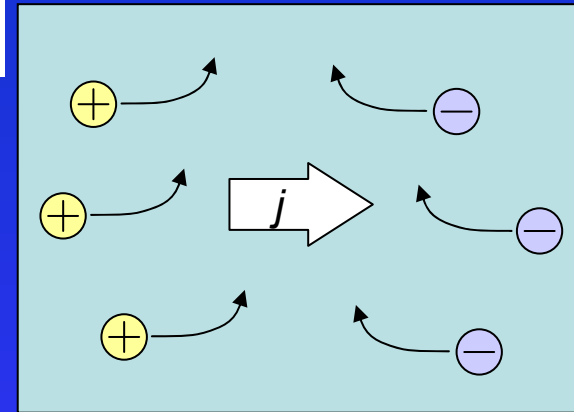
BUT: CeNiSn with two types of charge carriers

V. Oganessian and I. Ussishkin, *Phys. Rev. B* **70**, 054503 (2004)

A. Pourret et al., *Phys. Rev. Lett.* **96**, 176402 (2006)

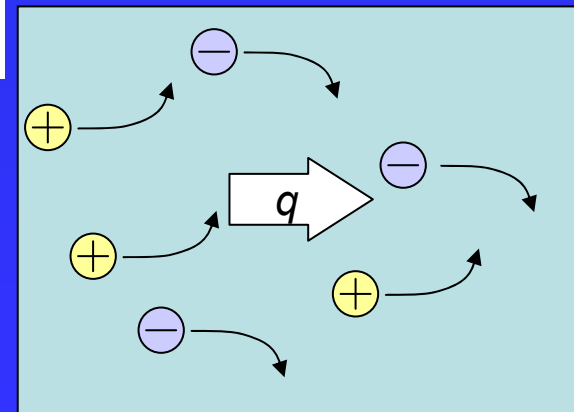
Hall effect

$\otimes \vec{B}$



Nernst effect

$\otimes \vec{B}$





Discussion: Nernst effect

Boltzmann approximation:

$$N^n = \frac{\pi^2}{3} \frac{k_B^2 T}{m^*} \frac{\partial \tau}{\partial \varepsilon} \Big|_{E_F}$$

with Hall angle

$$\tan \Theta_H = \frac{\sigma_{yx}}{\sigma_{xx}}$$

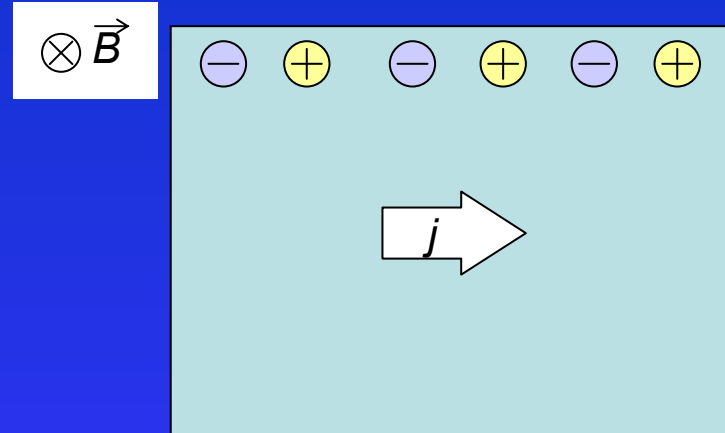
$$N^n \approx \frac{\pi^2}{3} \frac{k_B^2 T}{Be} \frac{\tan \Theta_H}{E_F}$$

BUT: CeNiSn with two types of charge carriers

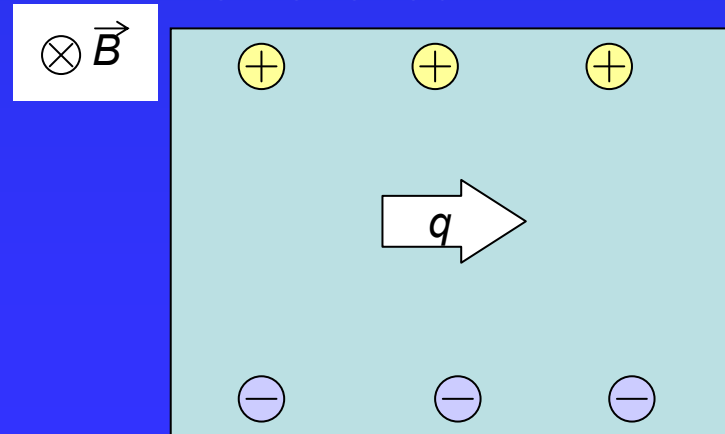
V. Oganeyan and I. Ussishkin, *Phys. Rev. B* **70**, 054503 (2004)

A. Pourret et al., *Phys. Rev. Lett.* **96**, 176402 (2006)

Hall effect



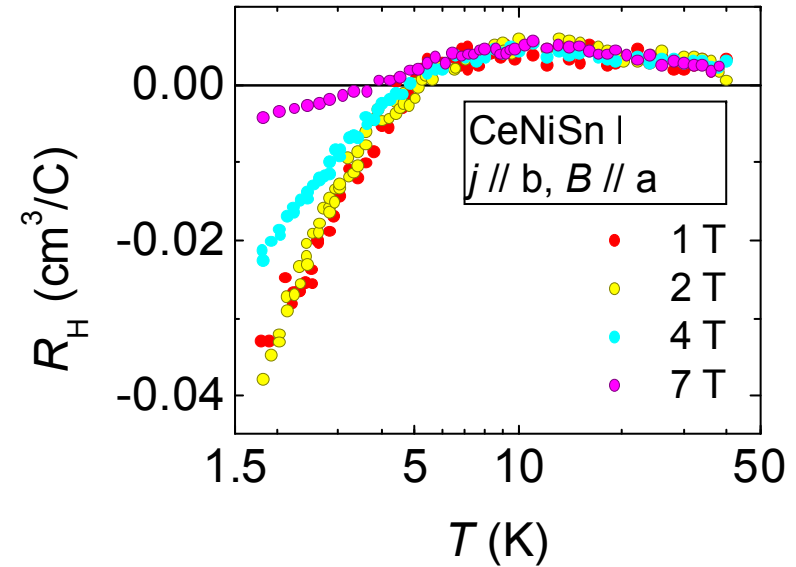
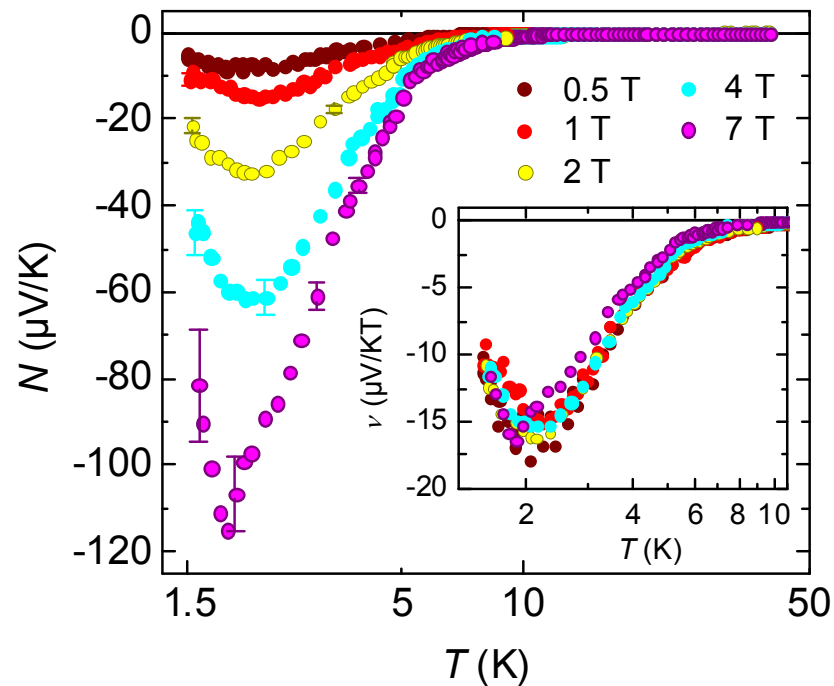
Nernst effect





Discussion: Nernst effect

$q \parallel b, B \parallel a$



sign change in R_H
but not in N



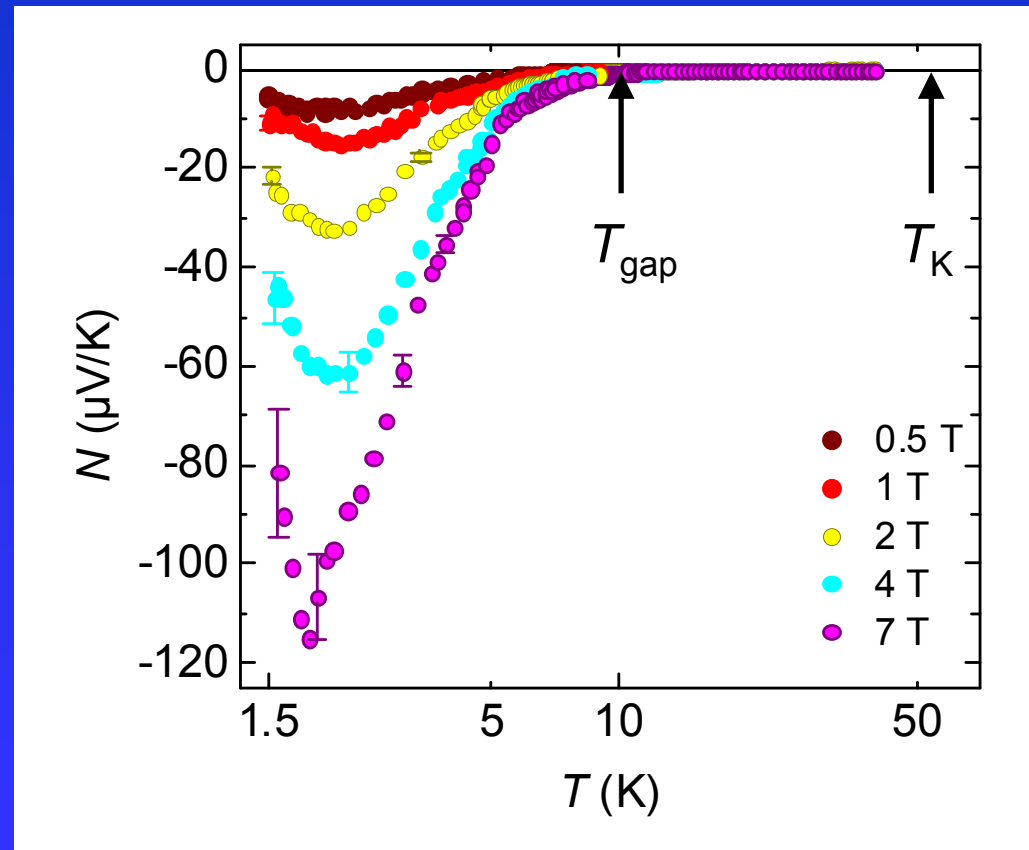
Discussion: Nernst effect

relevant mechanism:

opening of the gap \leftrightarrow
enhanced values of N

low Fermi energy T_K

$q \parallel b, B \parallel a$





Discussion: Nernst effect

$$v^n \approx \frac{\pi^2}{3} \frac{k_B^2 T}{Be} \frac{\tan \Theta_H}{E_F}$$

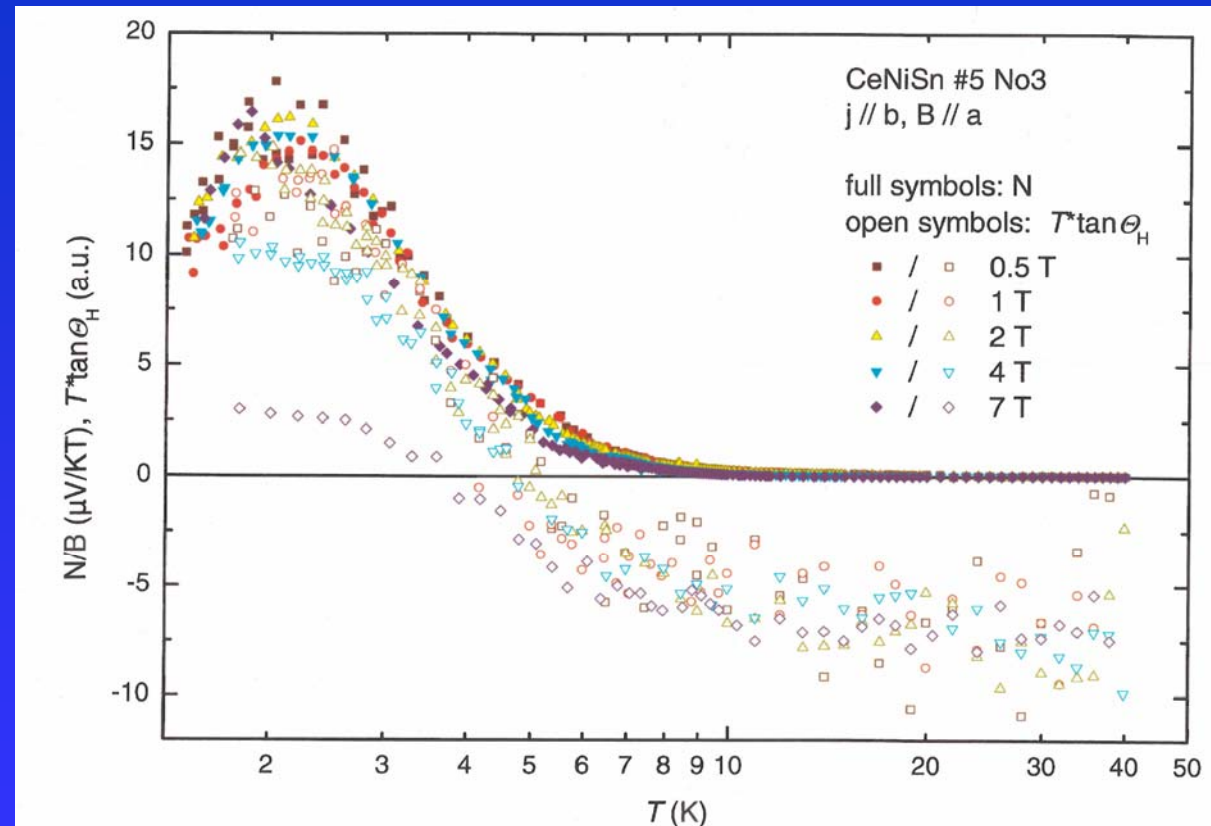
$$\tan \Theta_H = \frac{\sigma_{yx}}{\sigma_{xx}}$$

$$v \sim T/B \tan \Theta_H$$

Increase below T_g

No scaling

Hall coefficient stronger suppressed in field





Discussion: Nernst effect

$$v^n \approx \frac{\pi^2}{3} \frac{k_B^2 T}{Be} \frac{\tan \Theta_H}{E_F}$$

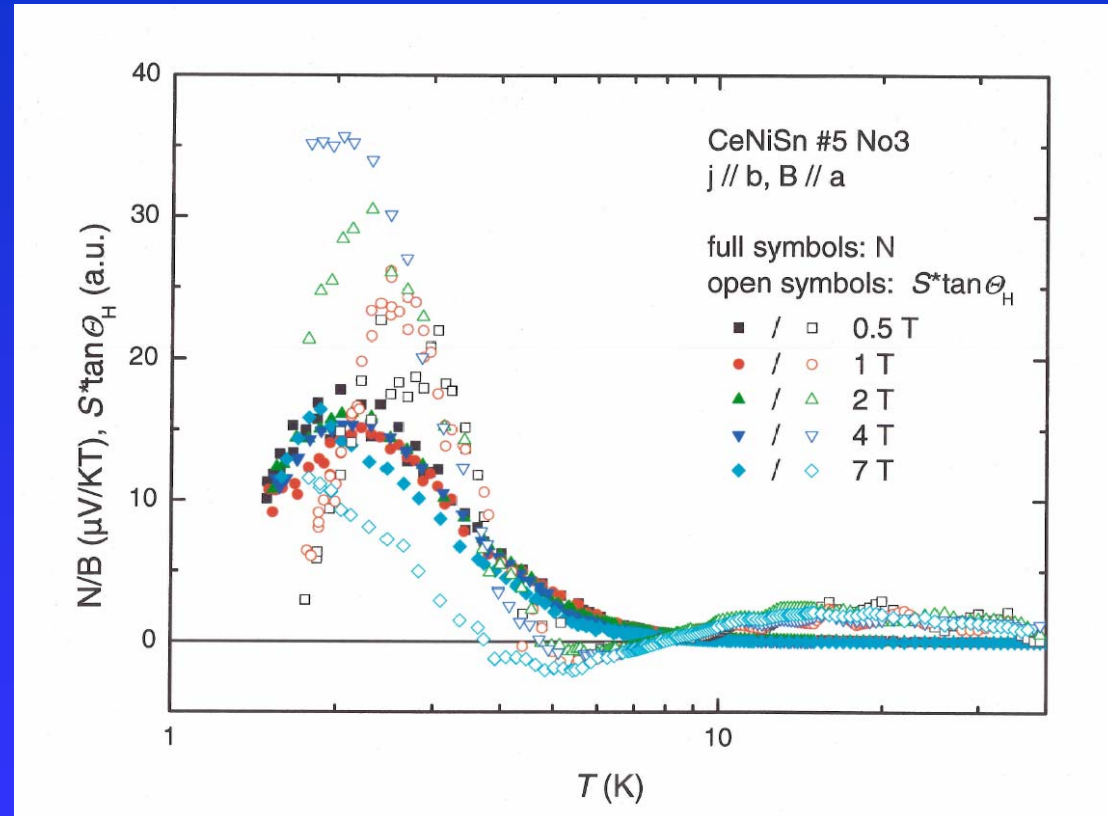
$$v \sim S/B \tan \Theta_H$$

Increase below T_g

No scaling

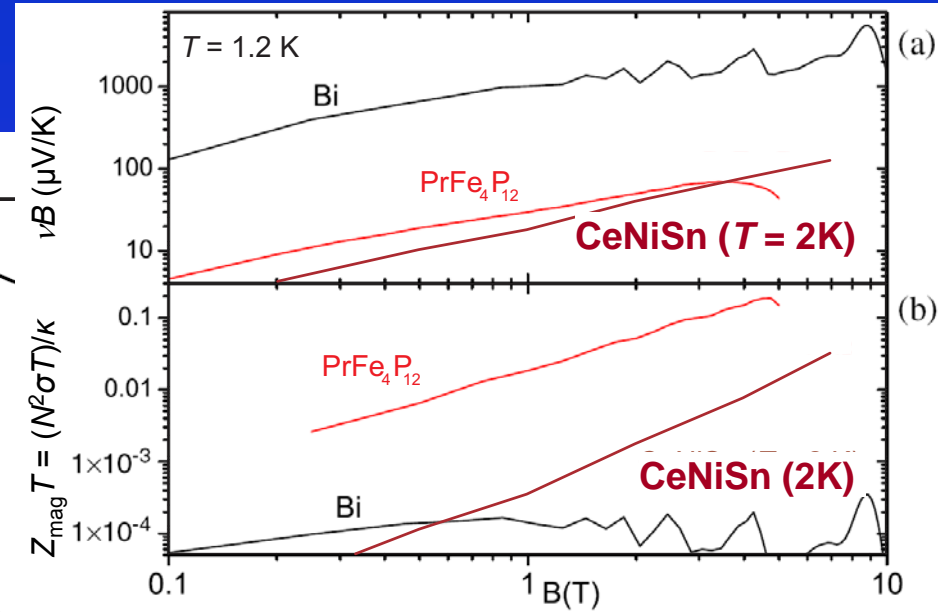
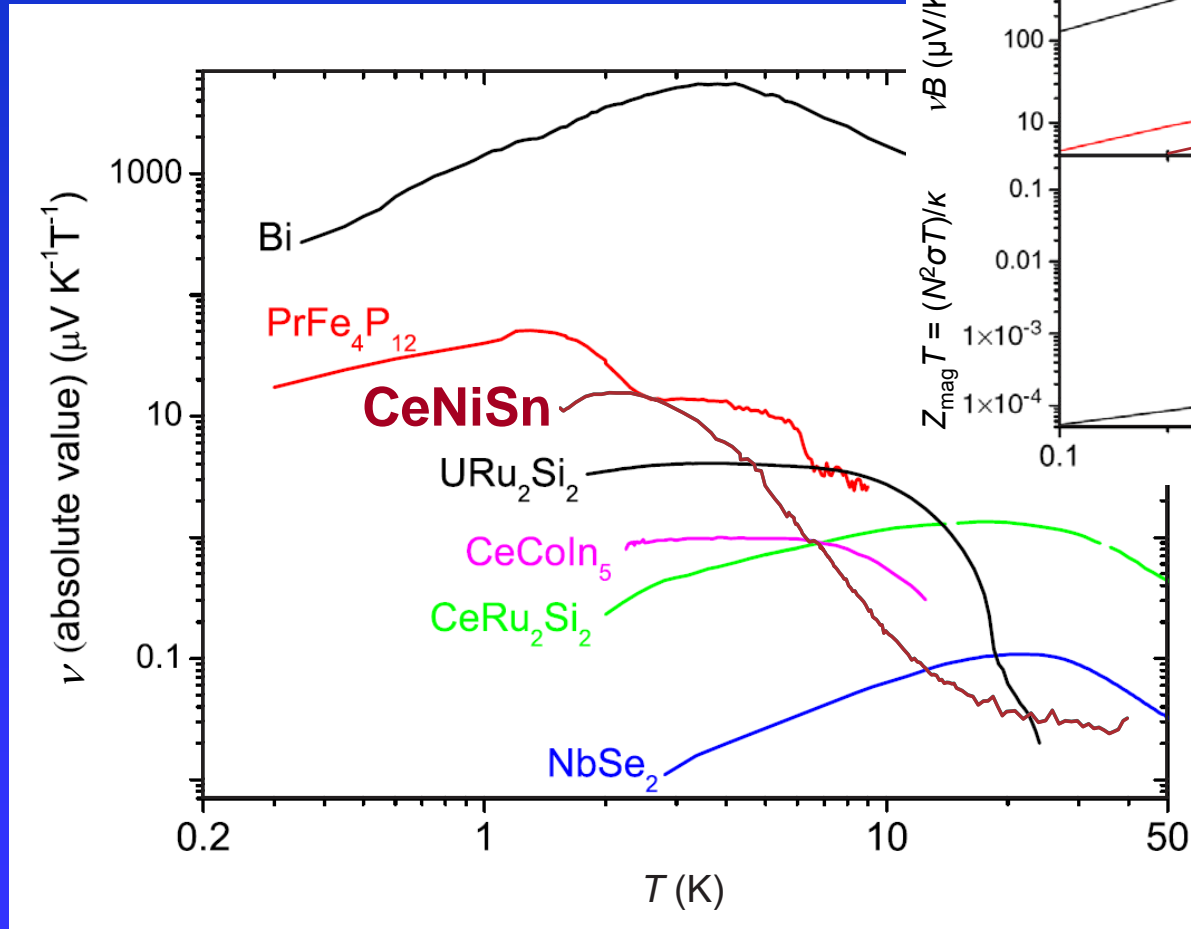
Maximum in $S \tan \Theta_H$ shifts in contrast to N/B

→ Multiband effects must be included





Discussion: Nernst effect



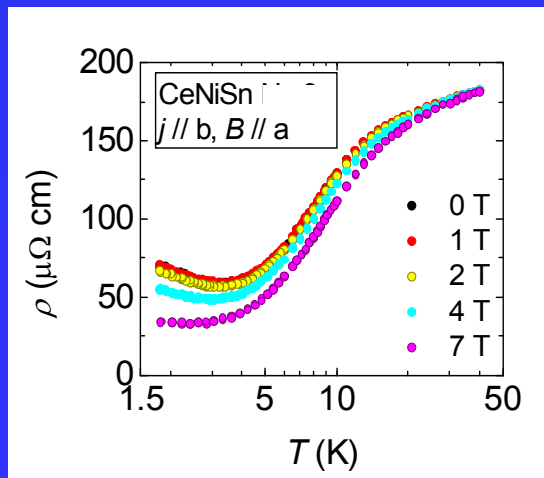
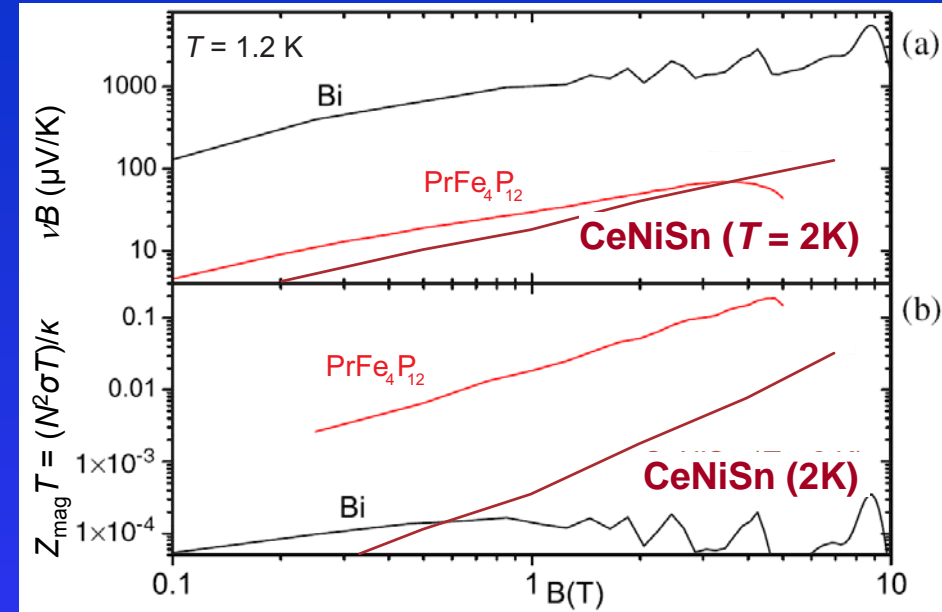
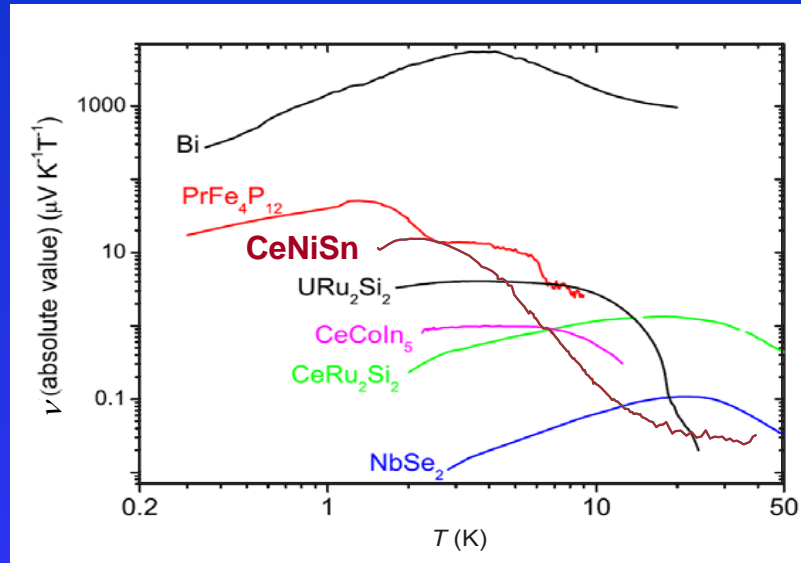
$Z_{\text{mag}} T$:

Low, since high κ
(single crystals)

High, since small MR



Discussion: Nernst effect



$Z_{\text{mag}} T$:

Low, since high κ

(single crystals)

High, since small MR



Summary

thermopower:

- first systematic study of $S(T,B)$ taking into account the Nernst signal
- reported sample dependence partly due to misorientation/ Nernst signal
- low- T thermopower governed by the pseudogap opening
- Unusal field dependence due to gap closing and Zeeman splitting



Summary

thermopower:

- first systematic study of $S(T,B)$ taking into account the Nernst signal
- reported sample dependence partly due to misorientation/ Nernst signal
- low- T thermopower governed by the pseudogap opening
- Unusal field dependence due to gap closing and Zeeman splitting

Nernst coefficient:

- large signal below gap opening
- scaling for $B//a$
- high relevance of the pseudogap
- potential of correlated semiconductors for Ettingshausen cooling (low $\rho(B)$)