Effet Nernst et la figure de mérite thermomagnétique dans les semi-métaux

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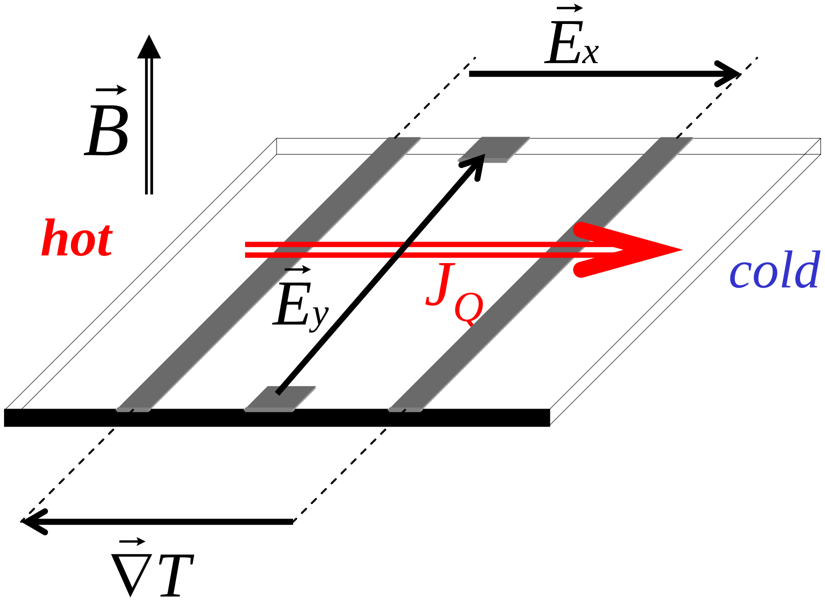
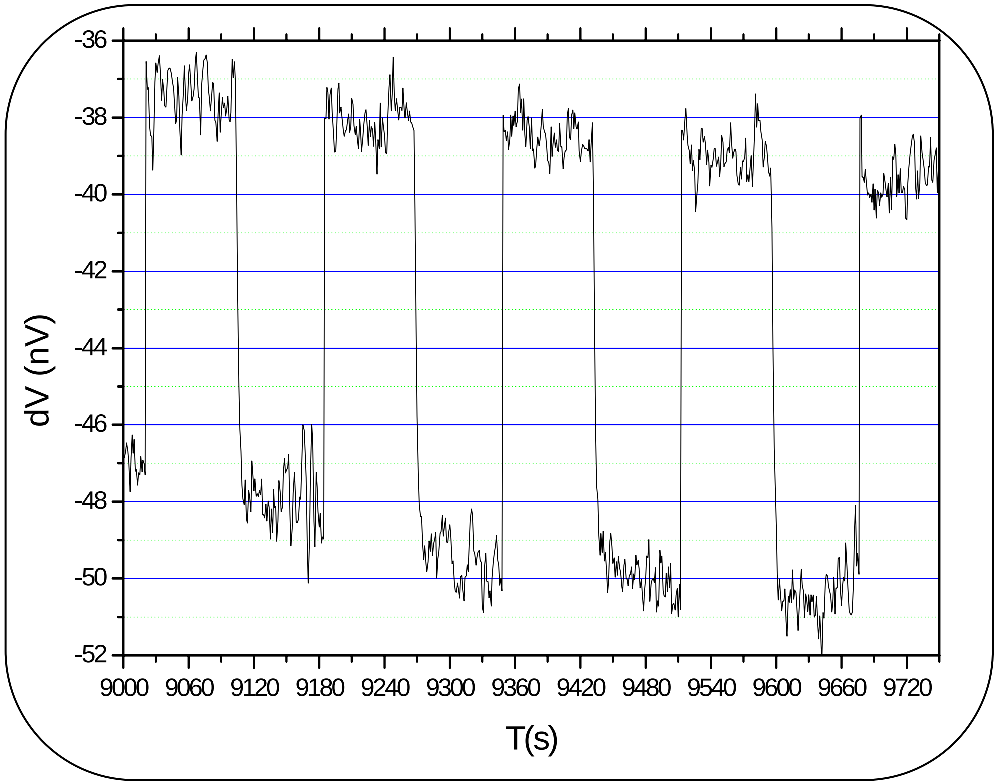
Thermoelectric coefficients

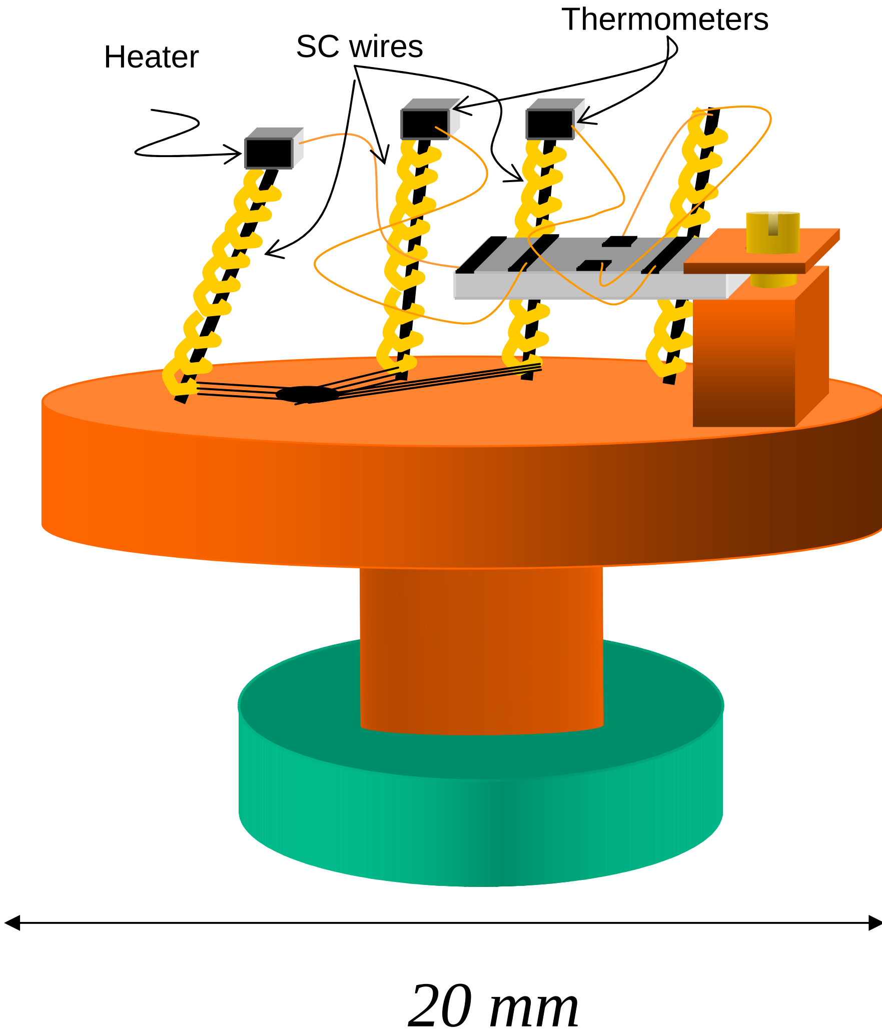
- In presence of a thermal gradient, electrons produce an electric field.
- Seebeck and Nernst effect refer to the longitudinal and the transverse components of this field.



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

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!

Thermoelectricity in extreme conditions (a network)

- LPEM-ESPCI, Paris

(down to 0.15 K, up to 12 T)

- SPSMPS-CEA, Grenoble (J. Flouquet)

(down to 0.1 K, up to 16 T)

- LNCMP, Toulouse (C. Proust)

(down to 1.5 K up to 62 T[pulsed field])

- LCMI, Grenoble (user facility)

(down to 0.15 K up to 28 T)

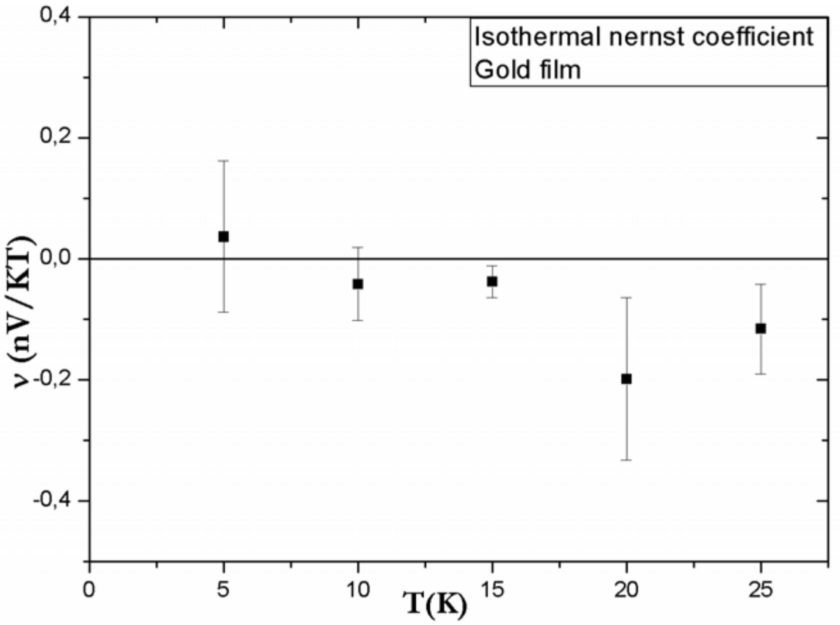
- NHMFL, Tallahassee (user facility)

(down to 0.35 K up to 33 T[45T planned])

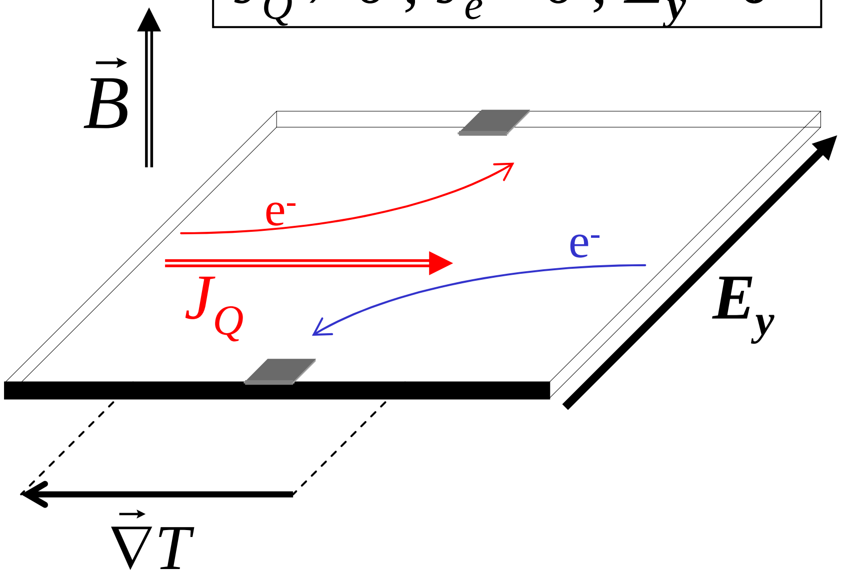
Nernst effect in a single-band metal

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

Almost undetectable in gold!(<0.5 nV/KT)



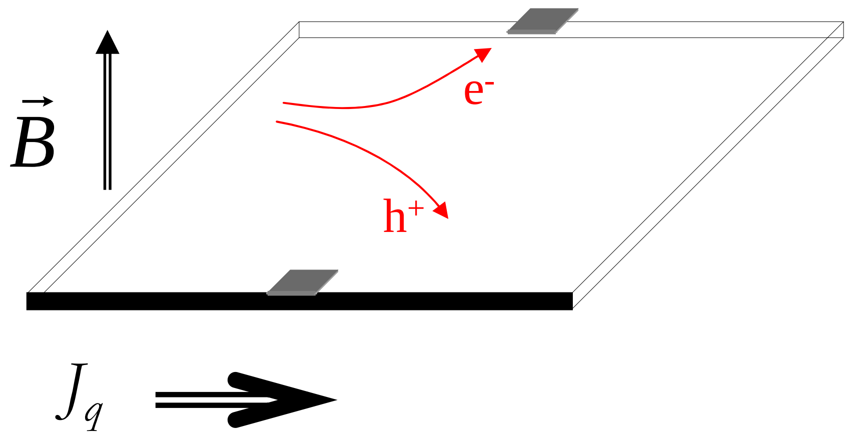
$J_Q \neq 0$; $J_e = 0$; $\boldsymbol{E_y} = \mathbf{0}$



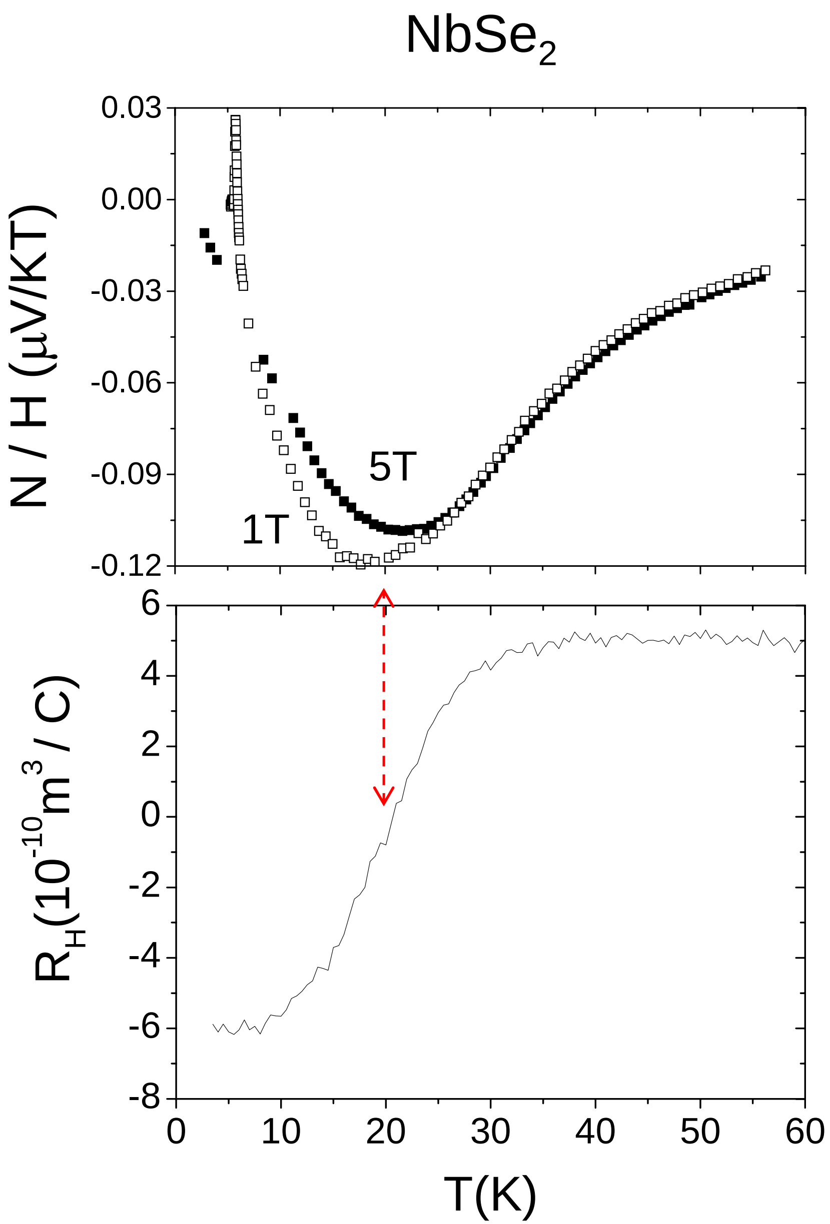
In an ideally simple metal, the Nernst effect vanishes!
(« Sondheimer cancellation », 1948)

Ambipolar Nernst effect

✓ The Nernst contribution os of hole-like and electron-like carriers add up!



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



What does « Sondheimer cancellation » mean?

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

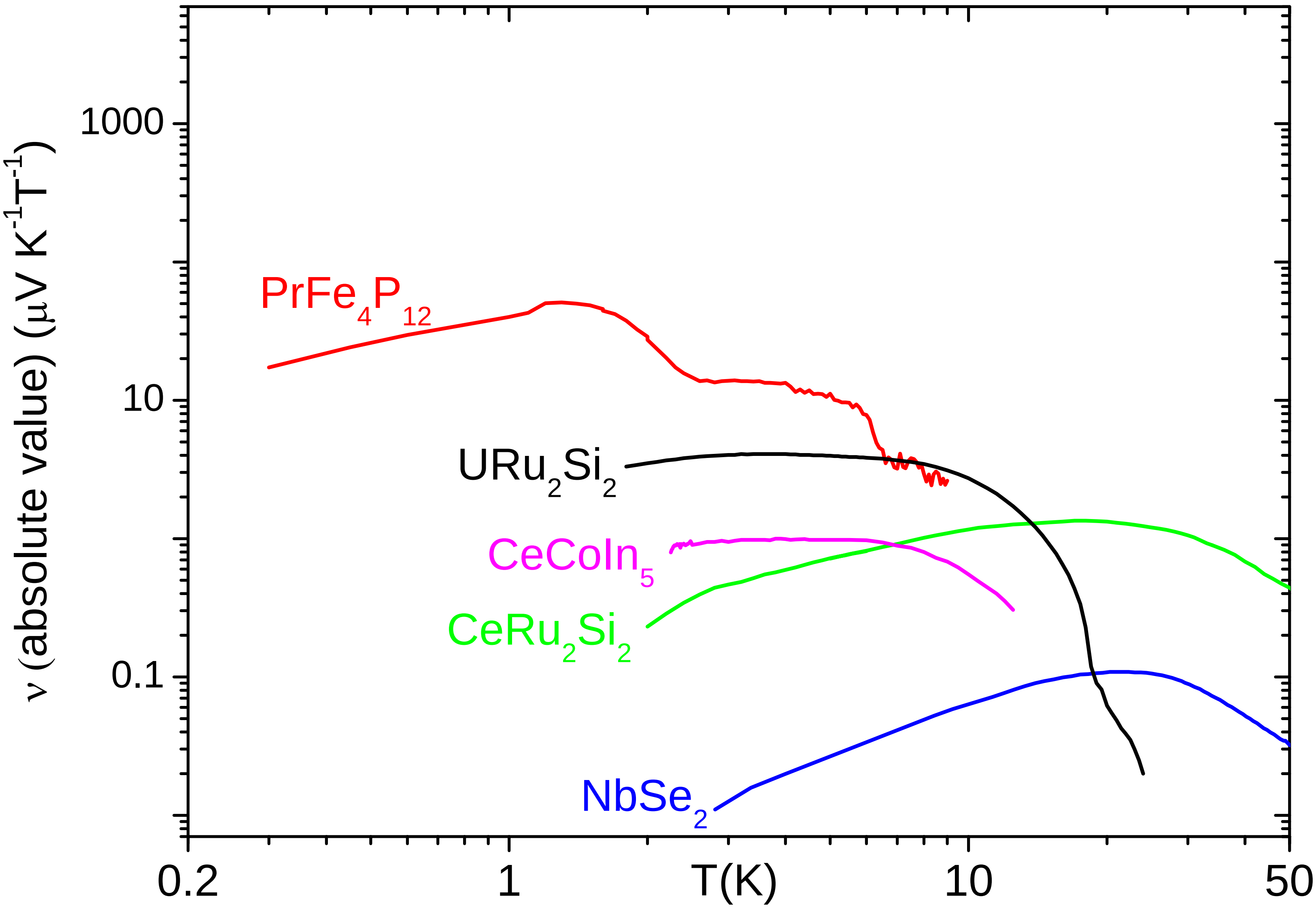
$J_e=0$

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

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

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

The Nernst coefficient can be large in some metals



The enigmatic order of URu_2Si_2!

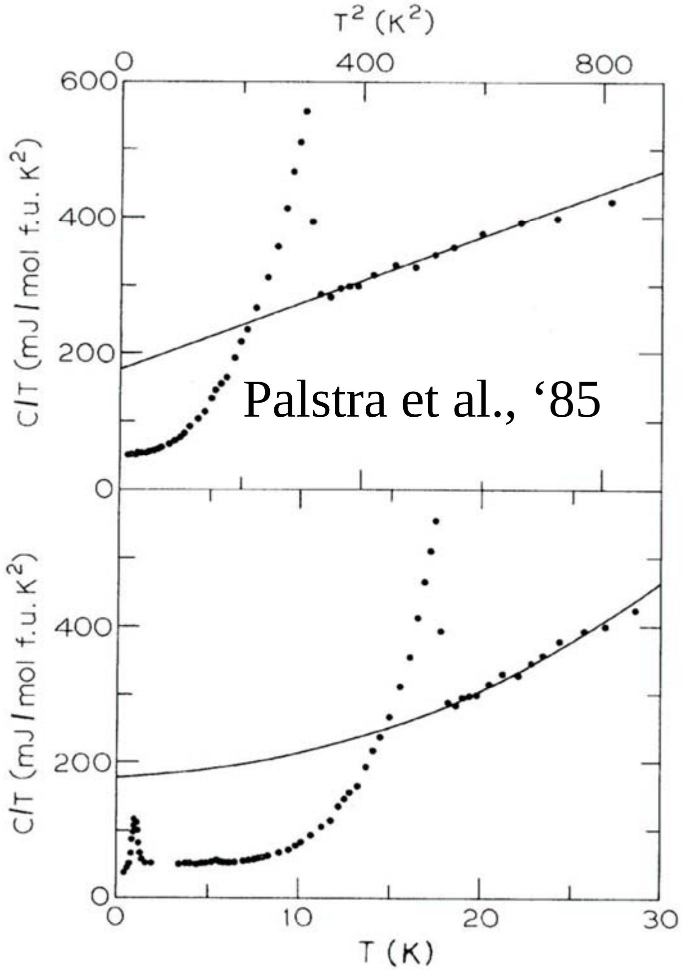


FIG. 1. Specific heat of URu_2Si_2 plotted as C/T vs T^2 (above) yielding γ and Θ_D, and as C/T vs T (below) showing the entropy balance.

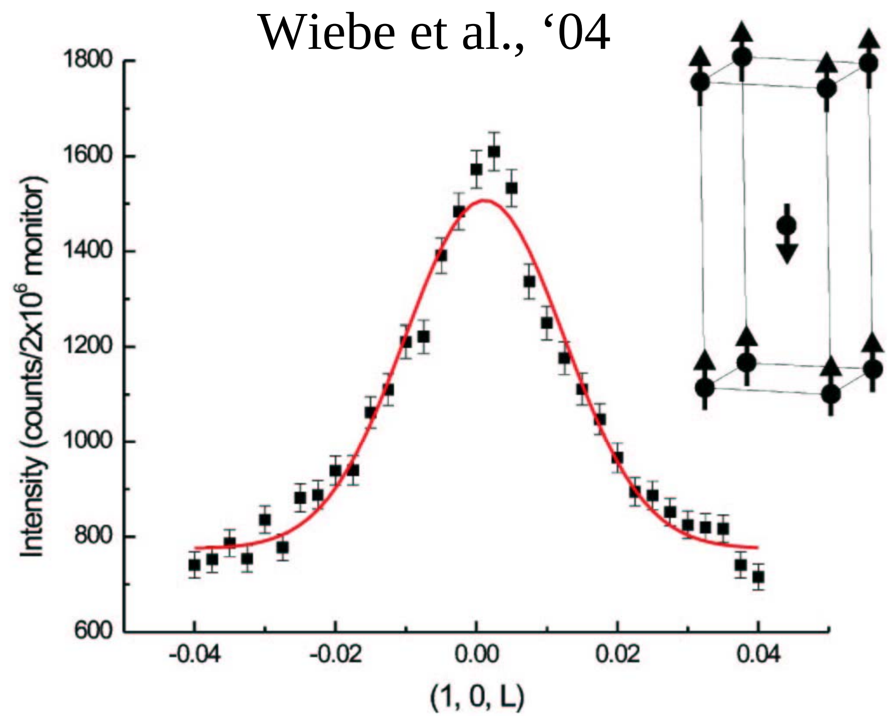
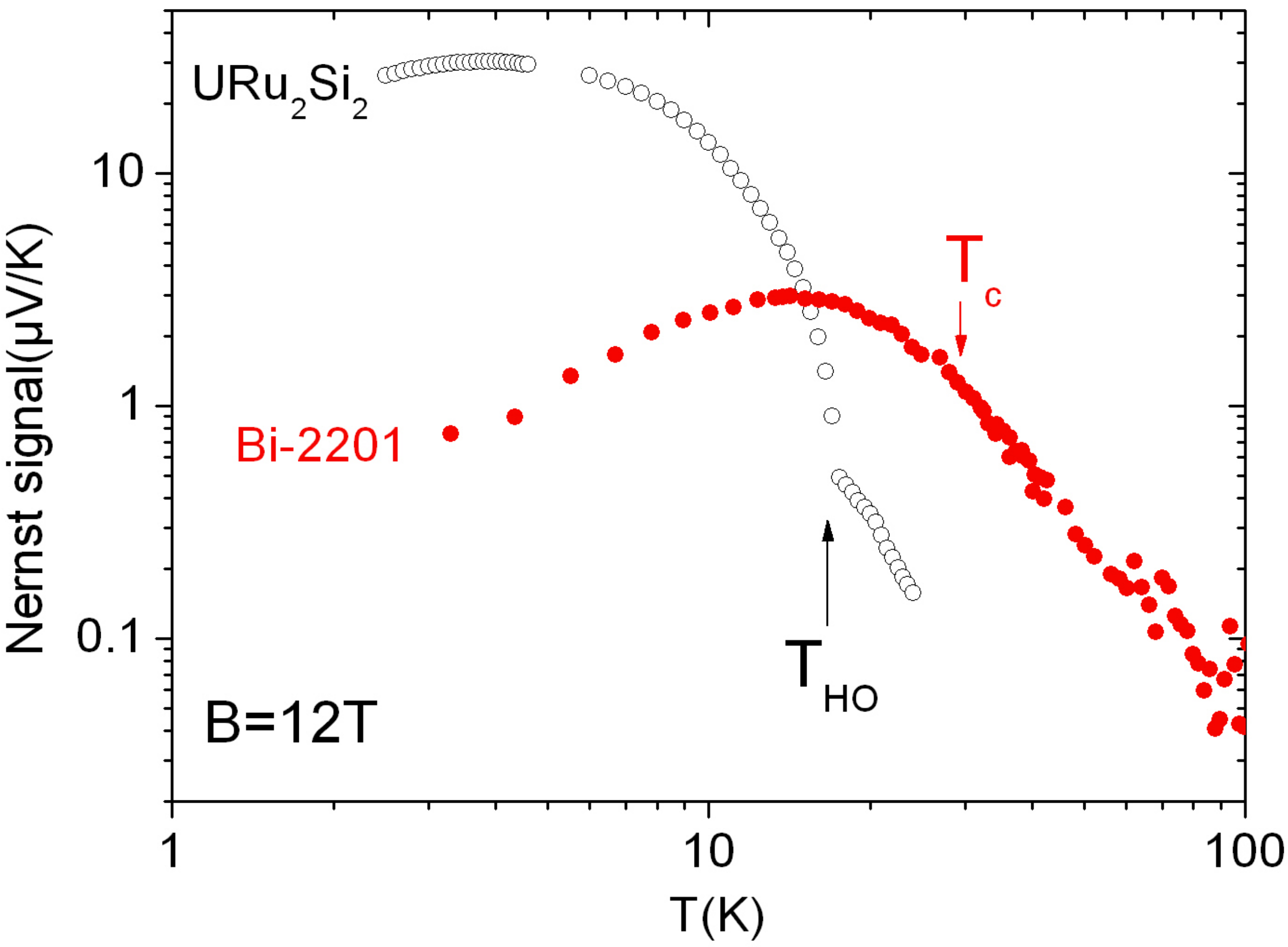


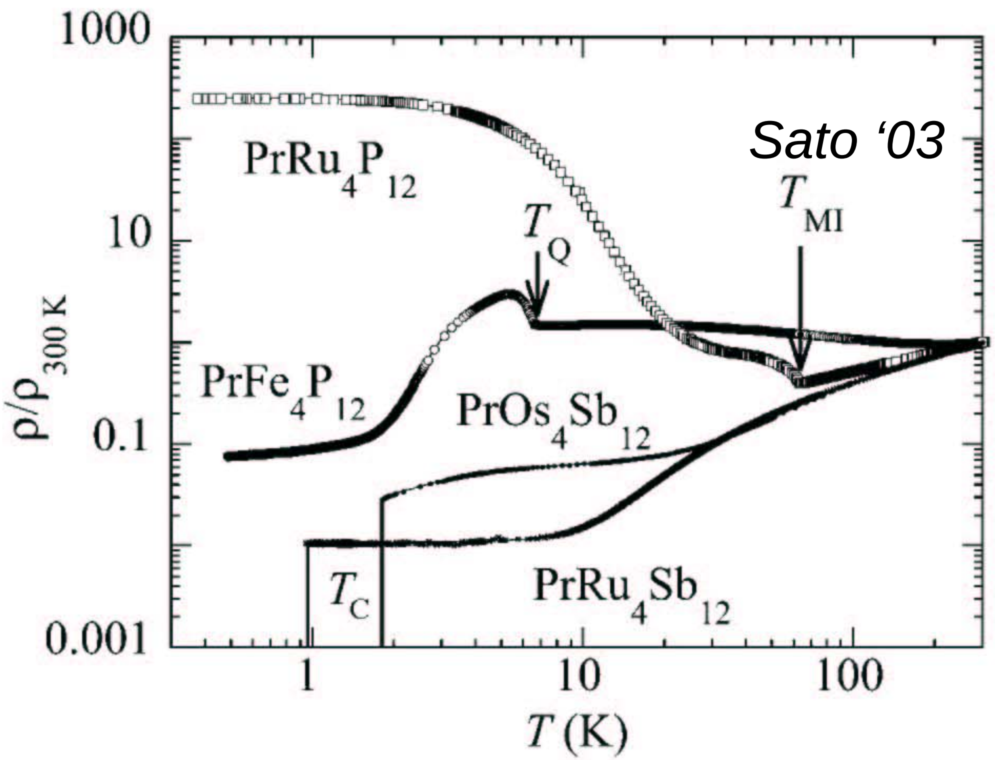
FIG. 1. The antiferromagnetic Bragg peak at (1, 0, 0) and $T = 8$ K (fit is to a Gaussian). The inset shows the corresponding magnetic structure for the U^{4+} moments.

A lot of entropy is lost, but only a tiny magnetic moment appears!

Exceeds by an order of magnitude the signal in high-T_c superconductors!



Pr-filled Skutterudites: a dazzling variety of ground states!



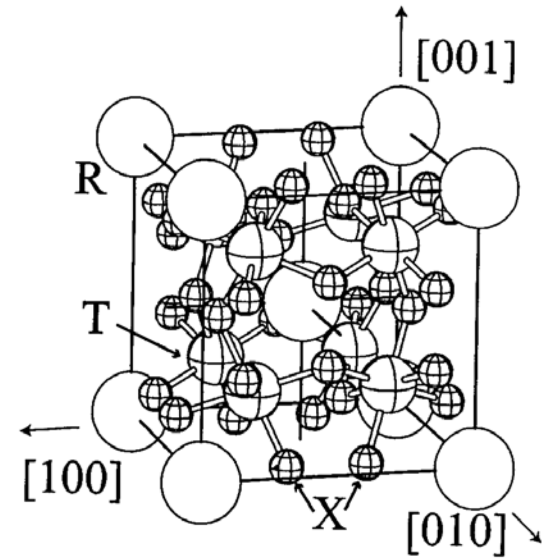


Fig. 1. Crystal structure of the filled skutterudite RT_4X_{12}.

Figure 1. Temperature dependence of electrical resistivity for the Pr-based filled skutterudites. T_{MI}, MI transition temperature; T_Q, transition temperature to the LOP; T_C, superconducting temperature.

What happens at T_Q in $PrFe_4P_{12}$?

How can quasi-particles produce a Nernst coefficient of this size?

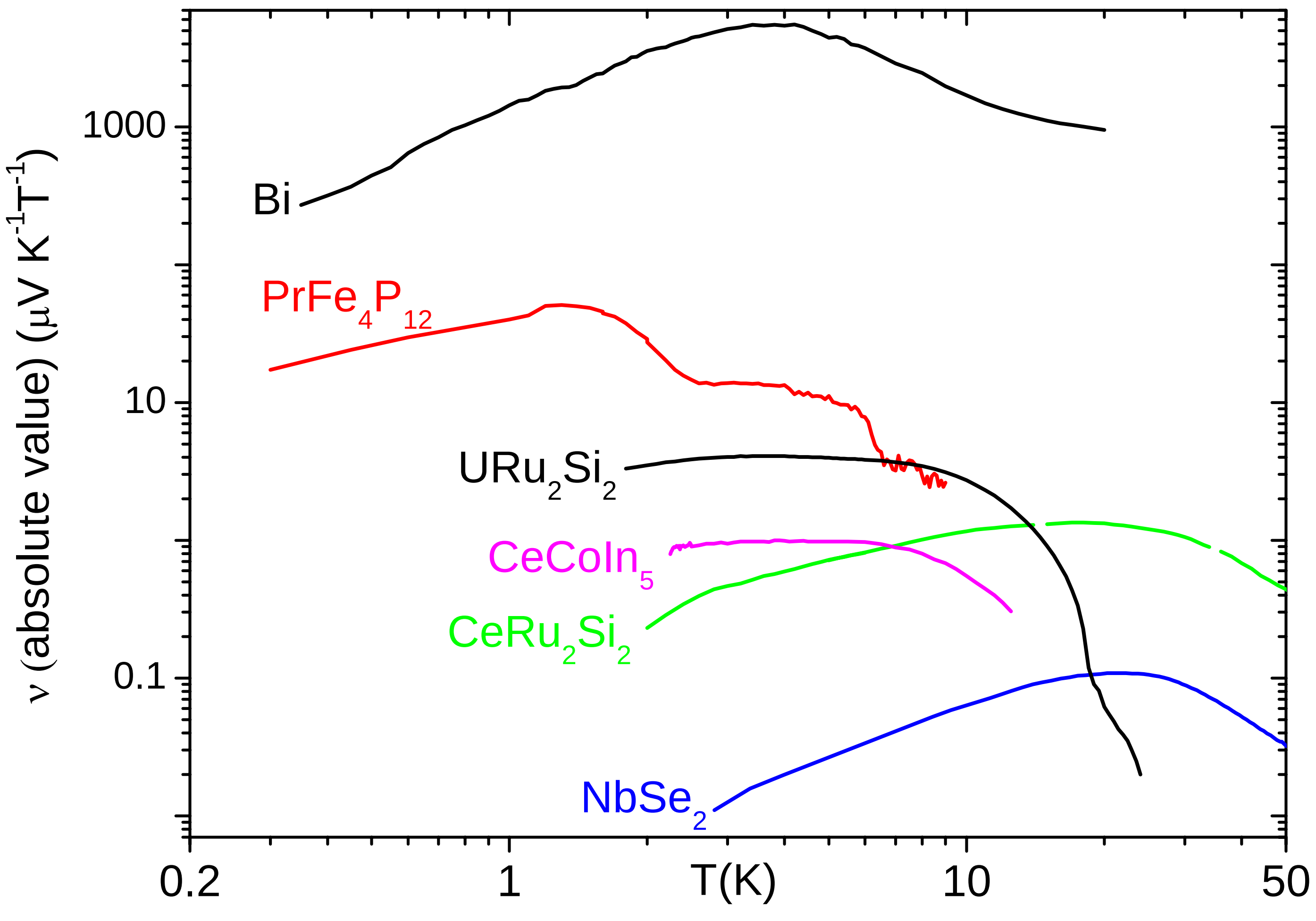
$$N = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \Theta_H}{\partial \epsilon}\Big|_{\varepsilon_F}$$

A crude estimation : $N = 285\ \mu V/K \times \Theta_H \times k_B T/\varepsilon_F$

A low Fermi energy and a large Hall mobility produce a giant Nernst effect!

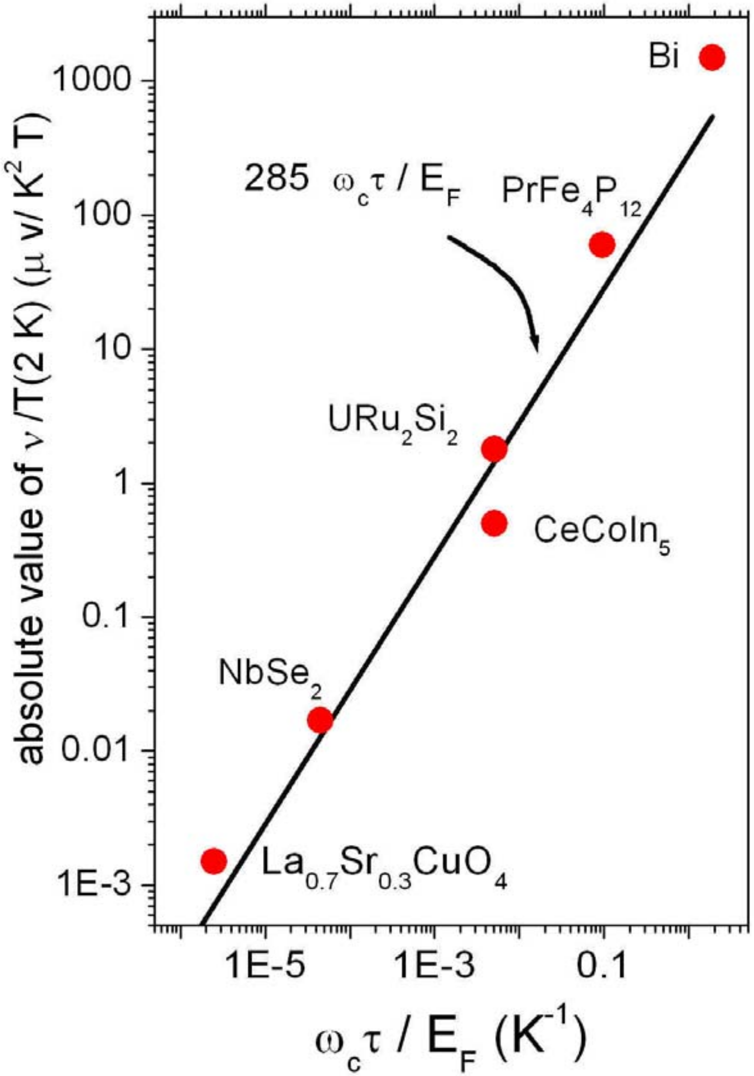
$$\Theta_H = \omega_c \tau = \frac{eB}{\hbar}\frac{\ell}{k_F}$$

Back to 1886!



- The Nernst effect in semi-metallic Bismuth is still more than one order of magnitude larger!

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



$N \sim \pi^2/3\ k_B/e\ \omega_c\tau / E_F$

	Bismuth	URu_2Si_2	$PrFe_4P_{12}$
k_F (nm^{-1})	0.14	1.1	0.7
m* (m_e)	0.06	25	10
n(per f.u.)	10^{-5}	0.005-0.018	0.03-0.05
$\omega_c\tau$ (1T)	420	0.08	0.85
E_F (K)	310	22	9
$283\omega_c\tau /E_F$	1400	40	1.9

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Ettingshausen Effect and Thermomagnetic Cooling

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(Received December 31, 1957)

The use of the Ettingshausen effect in refrigeration is considered. The phenomenological similarity of this cooling effect with the cooling by a cascade of Peltier couples is treated. Theoretical expressions are derived for the maximum cooling that is possible with the Ettingshausen effect, and it is shown that the optimum shape of the cooling element is an exponential of a given form. Preliminary experiments with bismuth alloys have given a cooling of about 0.25°C.

Thermomagnetic figure of merit:

$$ZT_{\varepsilon} = \frac{N^2 \sigma T}{\kappa}$$

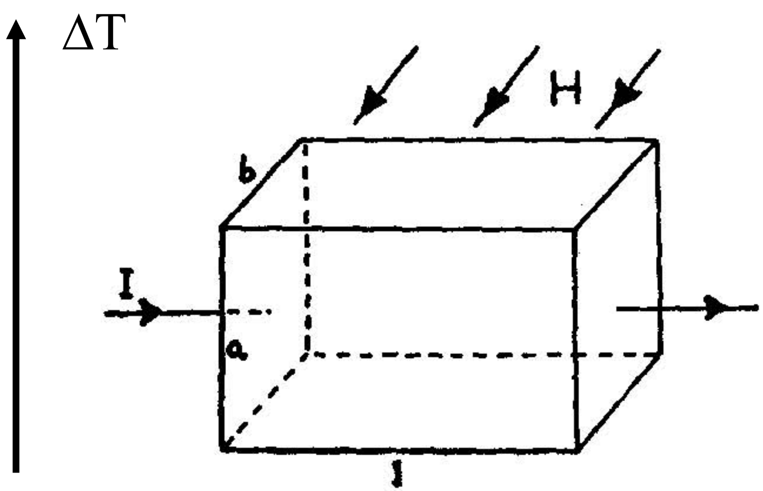


FIG. 1. Elementary configuration required for production of Ettingshausen cooling.

$$\Delta T_{\max} = \frac{ZT_{\varepsilon}}{2}$$

Infinite stage Ettingshausen cooler

T. C. Herman et al., Applied Physics Lett. 4, 77 (1964)

$$z(x) = z(0)\left[\frac{z(L_x)}{z(0)}\right]^{\frac{x}{L_x}}$$

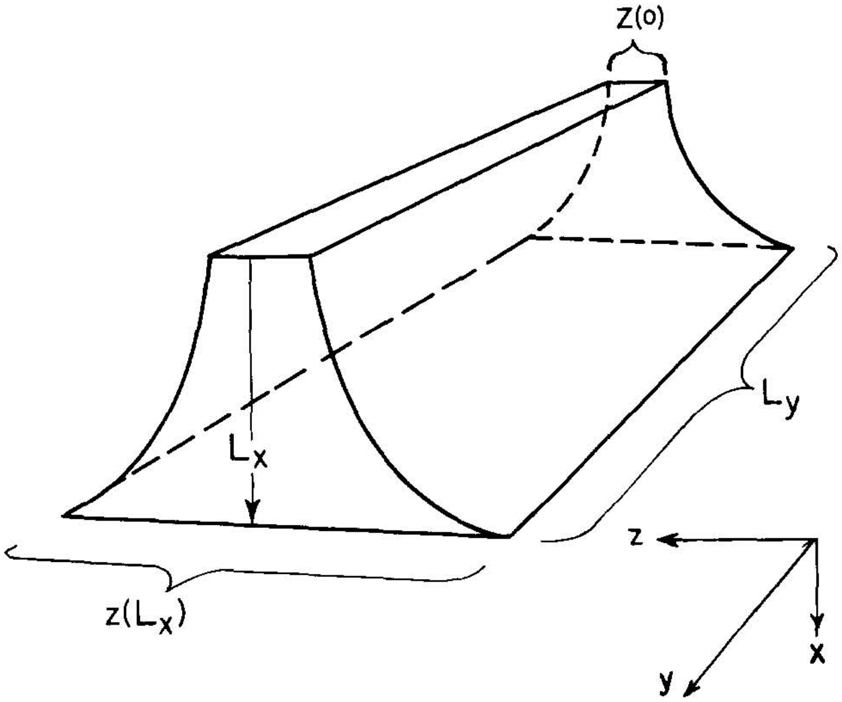


Fig. 1. Schematic diagram of a shaped Bi crystal along with the coordinate system and dimension symbols.

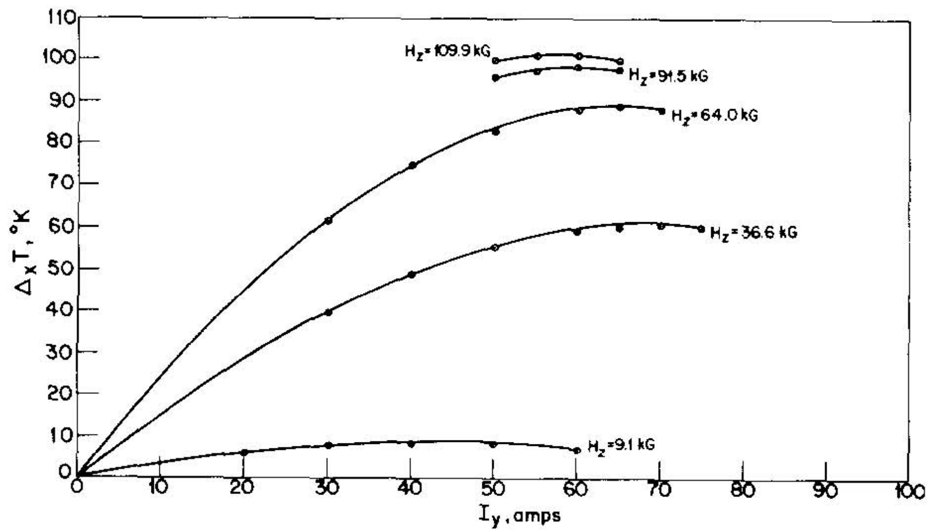


Fig. 2. Experimental curves of $\Delta_x T$ (temperature difference) vs I_y (current) for various H_z (magnetic field) for a shape ratio of 128 and a hot junction temperature of 302°K.

100 degrees of cooling! But, with B =

Ettingshausen cooling with a permanent magnet

K. Scholz *et al.* J. Appl. Phys. **75**, 5406 (1994)

$Bi_{0.97}Sb_{0.03}$

B=0.75 T

ΔT= 42 K at T=160K

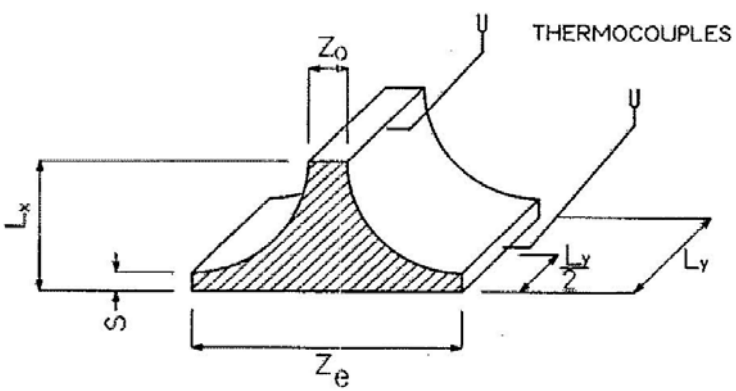


FIG. 2. Schematic drawing of the shaped cooler.

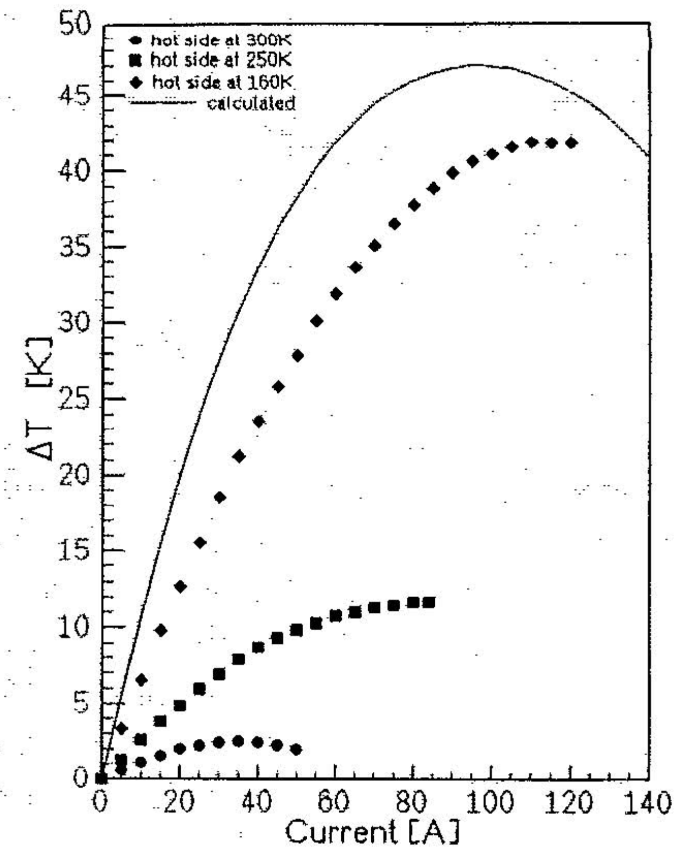


FIG. 3. Temperature difference ΔT as function of the absolute current for various heat sink temperatures, using an Ettingshausen cooler with exponential shape consisting of an oriented single crystal of $Bi_{97}Sb_3$. The line represents the result of a computer simulation.

What sets the magnitude of ZT_ε in a metal at low temperatures?

- Let us forget phonons!

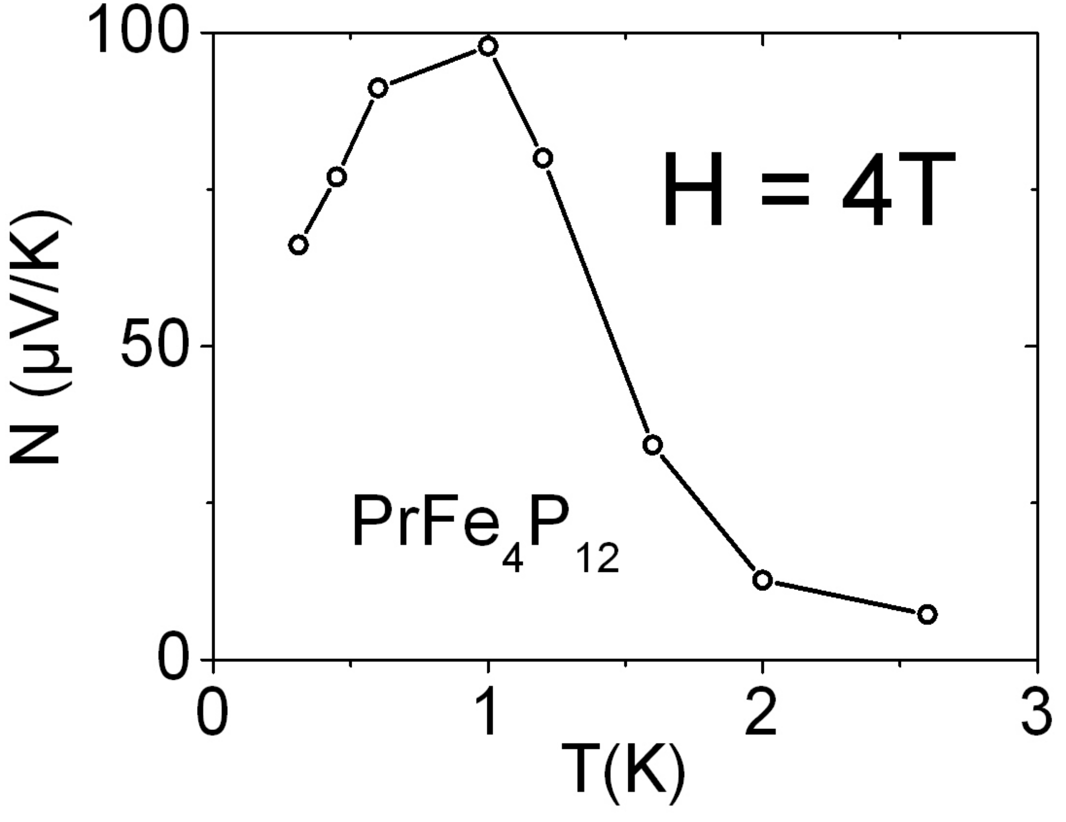
$$ZT_\varepsilon = \frac{N^2 \sigma T}{\kappa}$$

$$L = \frac{\kappa}{\sigma T} = 2.44 \times 10^{-8} \frac{V^2}{K^2}$$

[The Wiedemann-Franz law]

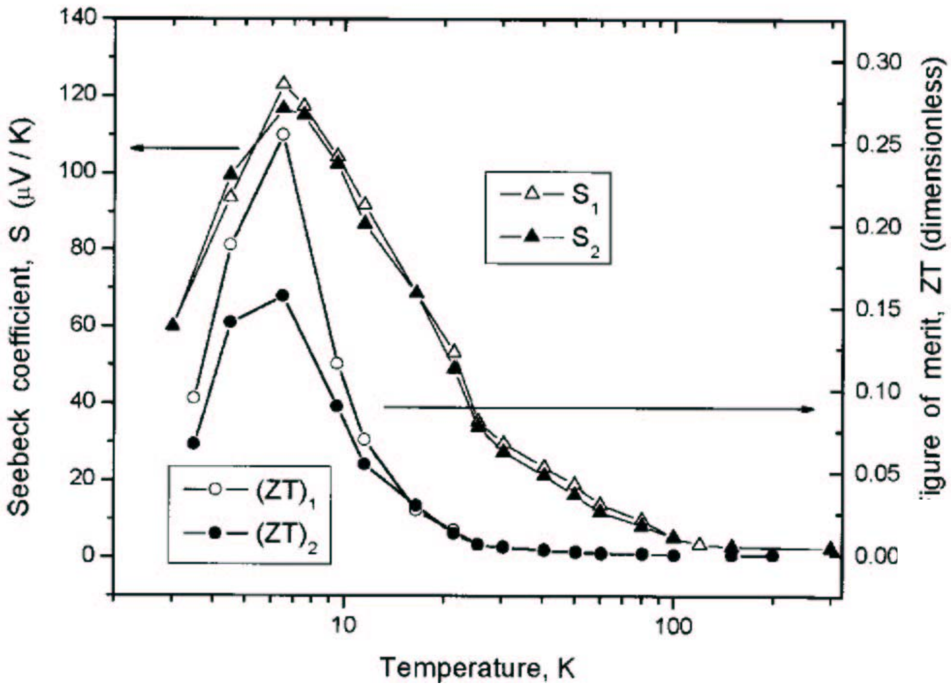
$$N \geq \sqrt{L_0}[= 156 \mu V / K] \Rightarrow ZT_\varepsilon \geq 1$$

In $PrFe_4P_{12}$, this threshold is approached!

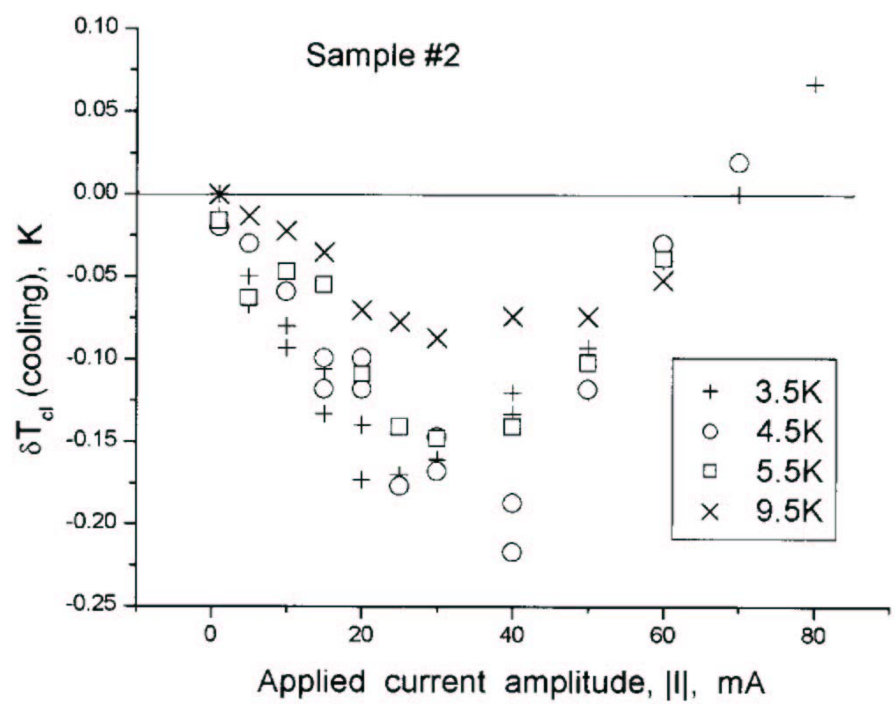


An interesting thermomagnetic material at cryogenic temperatures!

Peltier cooling with CeB_6 at cryogenic temperatures



Harutyanyan *et al.,* 2003

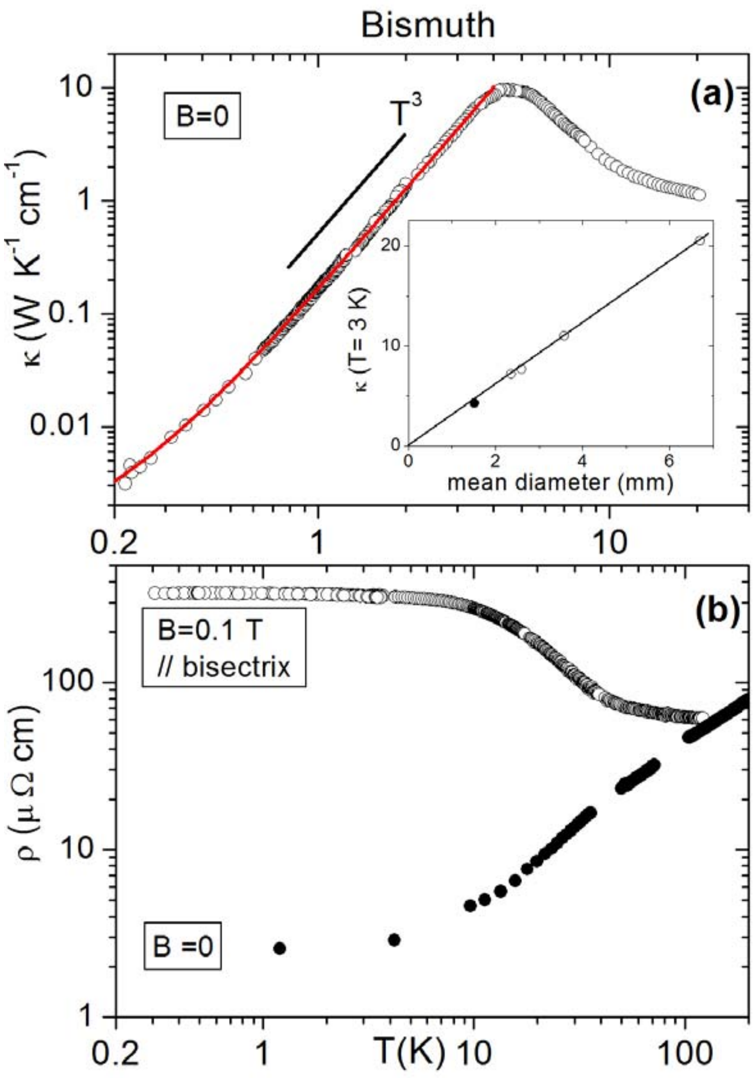


Why Bismuth does NOT qualify [for low T Ettingshausen cooling]?

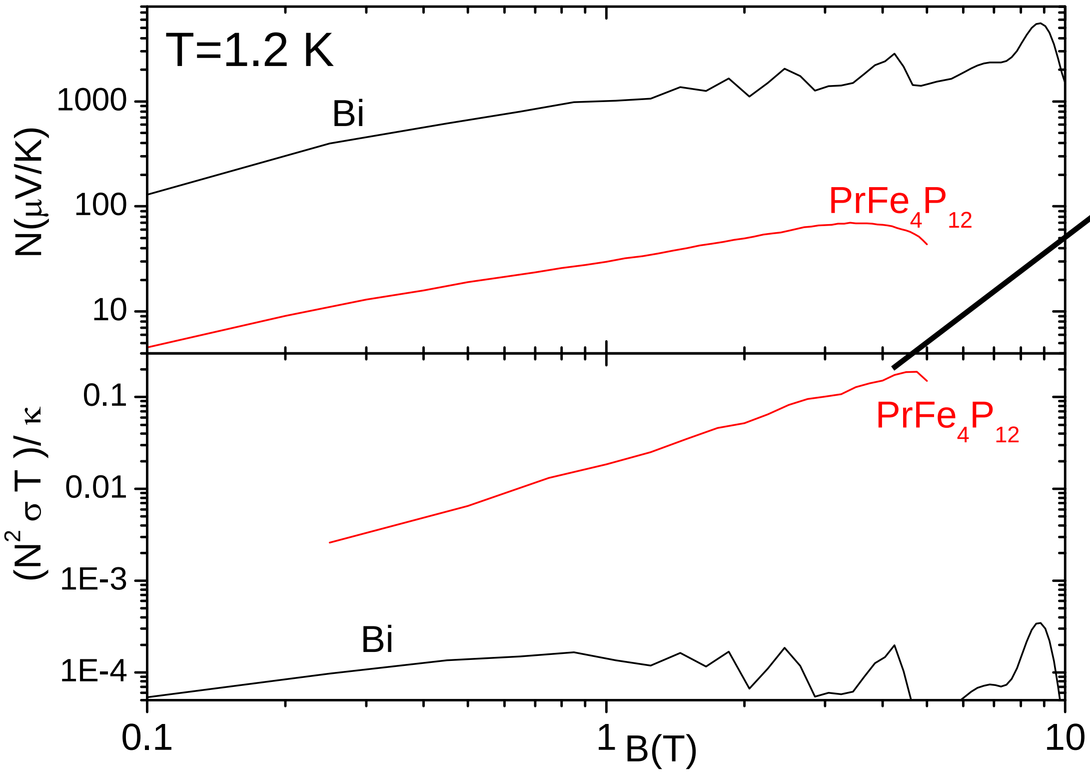
- In presence of a magnetic field, Bismuth behaves like an insulator!
- Magnetoresistance is very large!
- Heat is almost entirely carried by phonons!

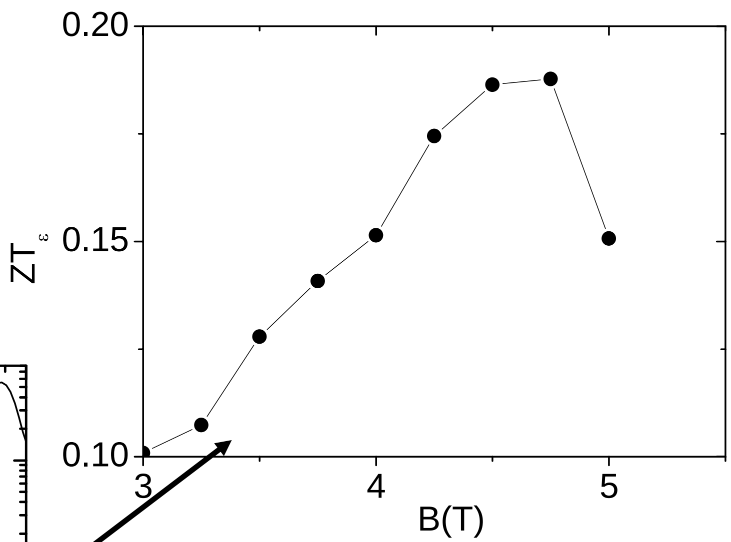
The ultimate fault:
The lightness of electrons leading to a large cyclotron frequency!

$$\omega_c = \frac{eB}{m^*} \qquad \Delta\rho \propto (\omega_c \tau)^2$$



A comparison of the thermomagnetic figures of merit at 1.2 K





Maximum ZT_ε ~ 0.19!

Heavy-electron semi-metals

- The combination of small k_F and large m* is not very common

CeNiSn

$PrFe_4 P_{12}$

URu_2Si_2

Carrier density 10^{-3}-10^{-2} /f.u.

Effective mass : 10-100 m_e

…emerge as promising candidates for Ettingshausen cooling!

Ettingshausen vs. Peltier cooling

Possibility of using an infinite stage cooler

ZT_{ε} depends on the electronic mean-free-path

But needs magnetic field!

1T (the field provided by permanent magnets) is an important threshold!

Summary

- Low Fermi energy and high electronic mobility are the ingredients to produce a giant Nernst effect!
- Semi-metals with heavy electrons may prove to be useful for thermomagnetic cooling at low temperatures
- The thermomagnetic figure of merit of $PrFe_4P_{12}$ appears large enough to attain subkelvin temperatures without He^3