



THE OHIO STATE UNIVERSITY

# *The Thermal Chiral Anomaly in ideal field-induced Weyl semimetals.*

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SPICE

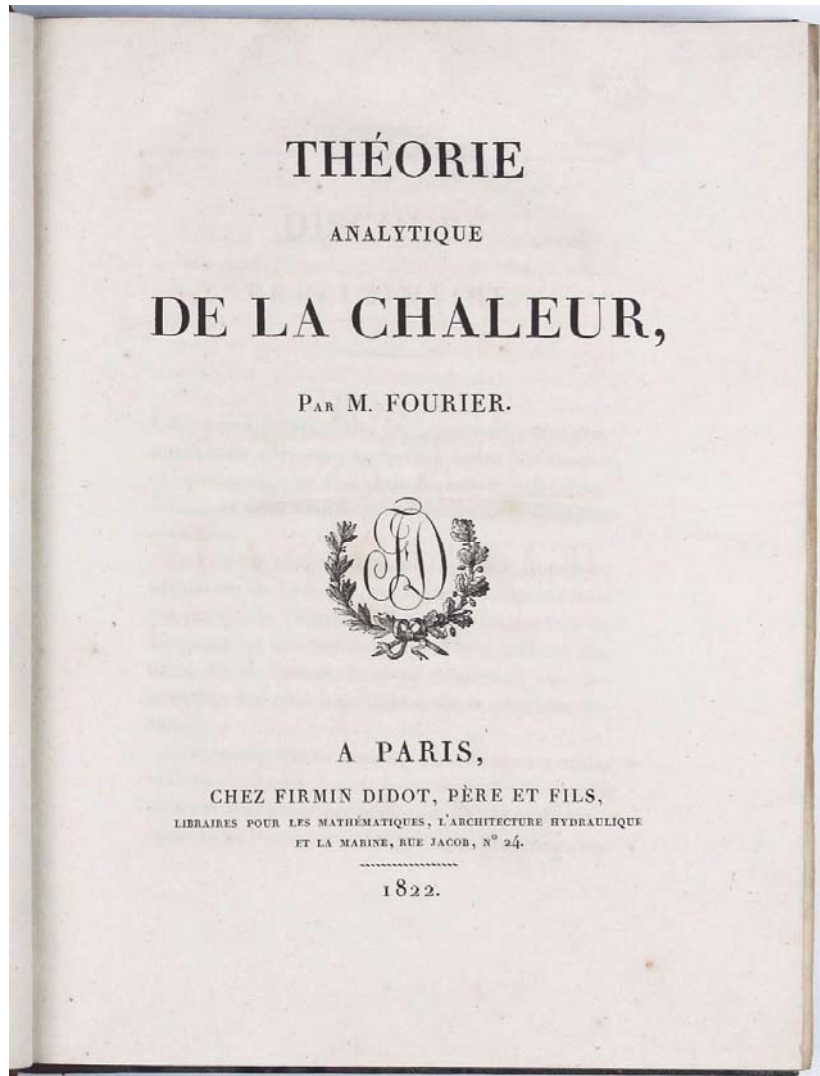
University of Mainz, Germany

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Grant No. DMR-2011876



## 1822: Fourier's Thermal Conductivity



*“La chaleur pénètre, comme la gravité, toutes les parties de l’univers.”*

“Heat, like gravity, permeates all parts of the universe.”

+ Thermal transport can be measured on quasi-particles that have neither charge nor spin

- Heat goes everywhere => measurements difficult.



1. Introduction:

- Weyl semimetals: chiral anomaly and and thermal conductivity
- Experimental difficulties
- Bi-Sb semiconductors alloys and TI's
- In the ultra-quantum limit: field-induced Weyls

2. Thermal conductivity

- Experimental evidence
- Robustness to phonons and defects
- Decay only via inter-Weyl point scattering
- The Wiedemann-Franz law

[arXiv:1906.02248](https://arxiv.org/abs/1906.02248)

# Weyl semimetals: 3-dimensional topological solids

$$E(\vec{k}) = \pm v_F \hbar \vec{k}$$

$$\vec{k} = (k_x, k_y, k_z)$$

Fermi level at the Dirac points:

- At  $T > 0$ , same amount of electrons and holes
- We work at temperatures

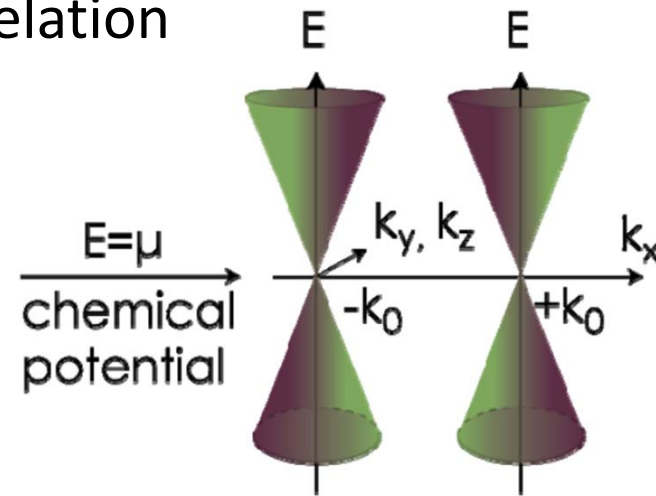
$$\mu \ll k_B T < E_{BW}$$

- Mass is not defined
- Electron has **velocity**  $v_F$
- Electron has **chirality**  
 $\chi = 1$  at  $\vec{k}_0$ ,  $\chi = -1$  at  $-\vec{k}_0$
- Berry phase  
 $\vec{\Omega}_{\pm}(\vec{k}) = \pm \frac{\vec{k}}{k^3}$
- Equation of motion

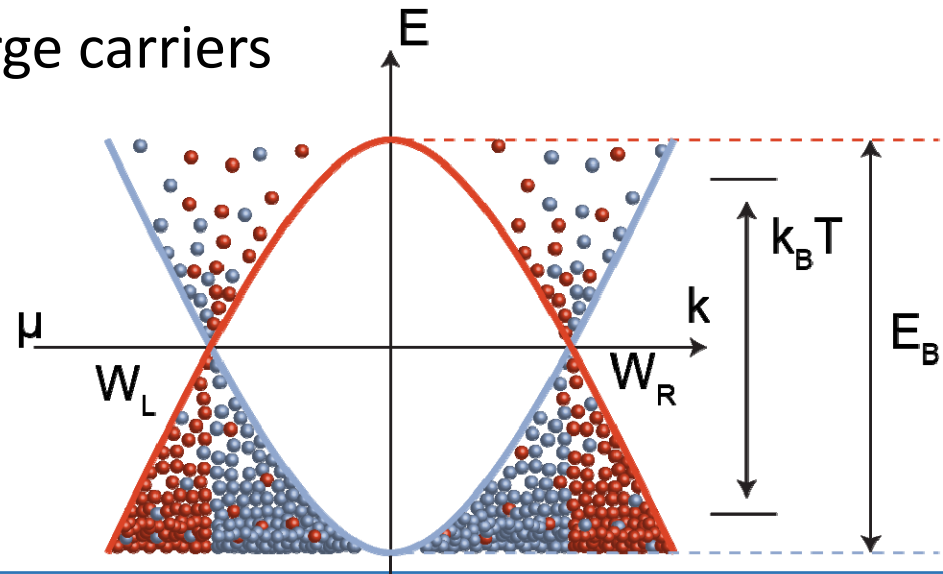
$$\frac{\partial \vec{r}}{\partial t} = \nabla_{\vec{k}} \varepsilon + \underbrace{\frac{\partial \vec{k}}{\partial t} \times \vec{\Omega}}_{\text{Anomalous velocity}}$$

Anomalous velocity

Dispersion relation

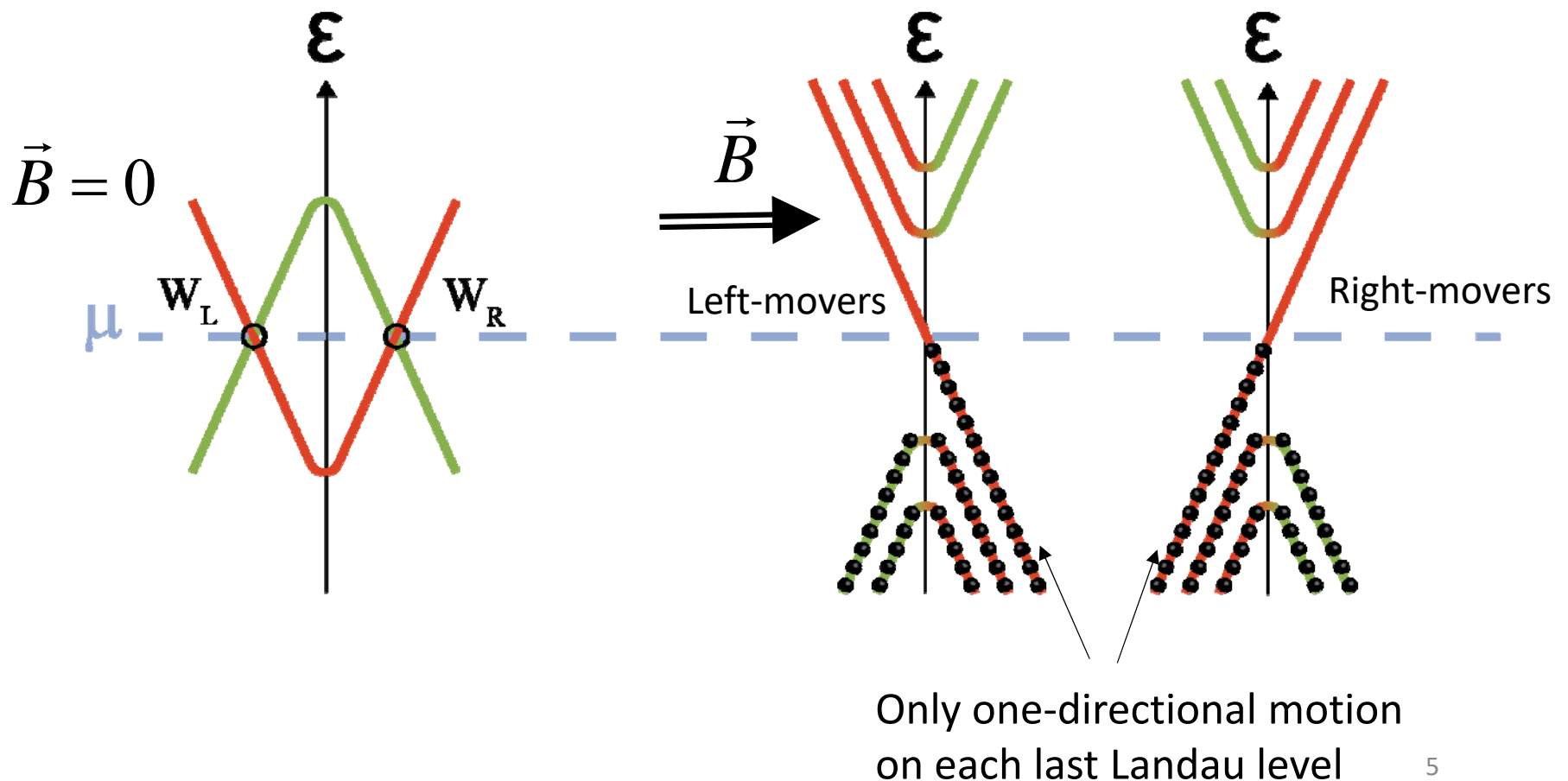


Charge carriers

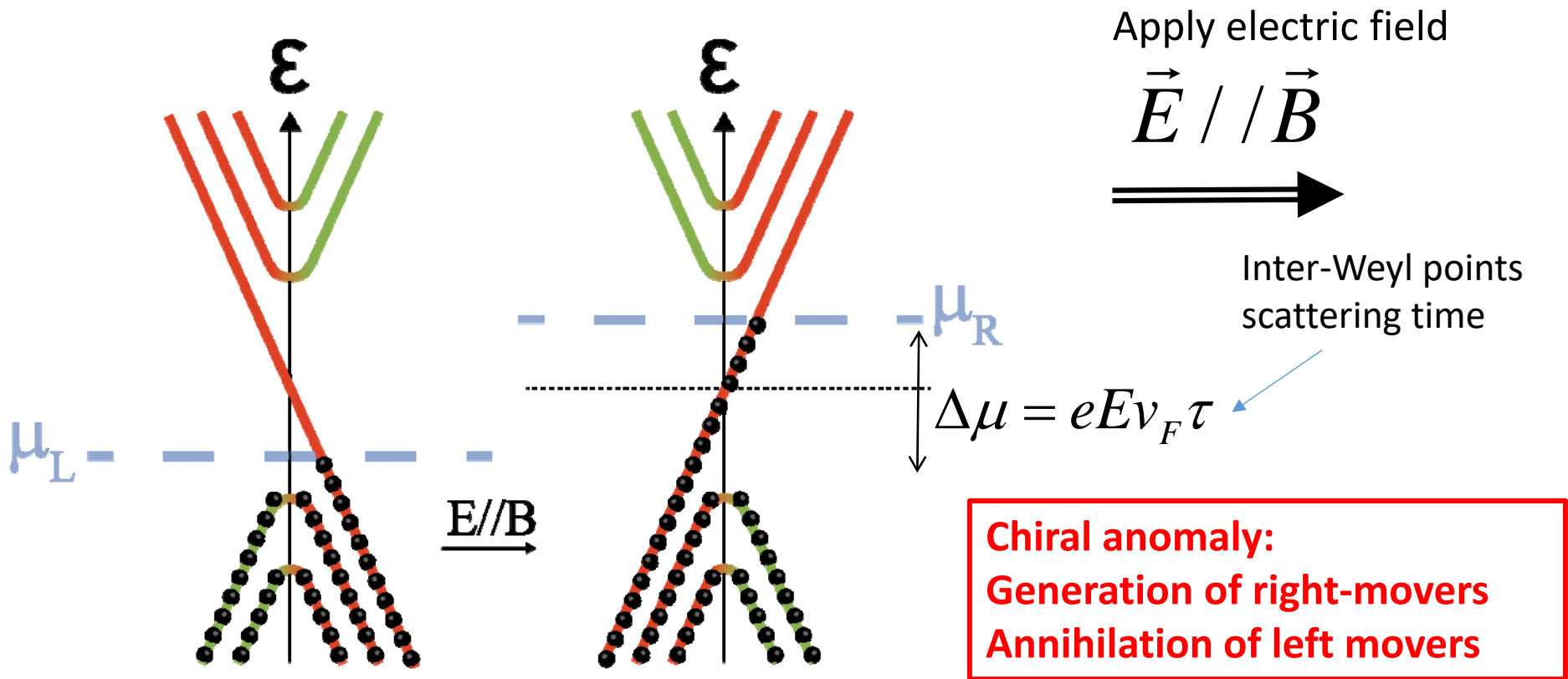


## Landau levels, extreme-quantum limit (EQL)

- Apply quantizing magnetic field => Landau levels
- Extreme quantum limit: only last Landau level crosses chemical potential
- Effect of  $\Omega$ : only one velocity allowed for the last Landau level
- => carriers separate into left-movers and right-movers



## The chiral anomaly: apply electric field



Anomalous Current  $j_A \propto DOS_{2D} ev_F \tau E \propto \vec{E} \cdot \vec{B}$

Anomalous conductivity:  $\sigma_A = \frac{e^2 v_F \tau}{2\pi h \ell_B^2}$

**Negative magnetoresistance**

## Effects of the anomalous velocity on transport

Solve Boltzmann transport equations with both  $\mathbf{E} // \mathbf{B}$  &  $\nabla_r T // \mathbf{B}$

=> Change in carrier concentration between left and right movers:

$$\delta n_\chi = \frac{\chi e^2 \tau}{4\pi^2 \hbar^2} [\vec{B} \cdot \vec{E}] C_0 + \frac{\chi e \tau}{4\pi^2 \hbar^2} \left[ \vec{B} \cdot \frac{-\nabla_r T}{T} \right] C_1$$

$$C_n = \int_{-\infty}^{\infty} (\varepsilon - \mu)^n \left( -\frac{\partial f_0}{\partial \varepsilon} \right) d\varepsilon; \quad n \in \{0, 1, 2, \dots\}; \quad \chi = \pm 1$$

In an ideal Weyl semimetal  $\mu = 0 \Rightarrow C_0 = 1; C_1 = 0$

$$\delta n_\chi = \frac{\chi e^2 \tau}{4\pi^2 \hbar^2} [\vec{B} \cdot \vec{E}]$$

Anomalous electrical conductivity, negative magnetoresistance

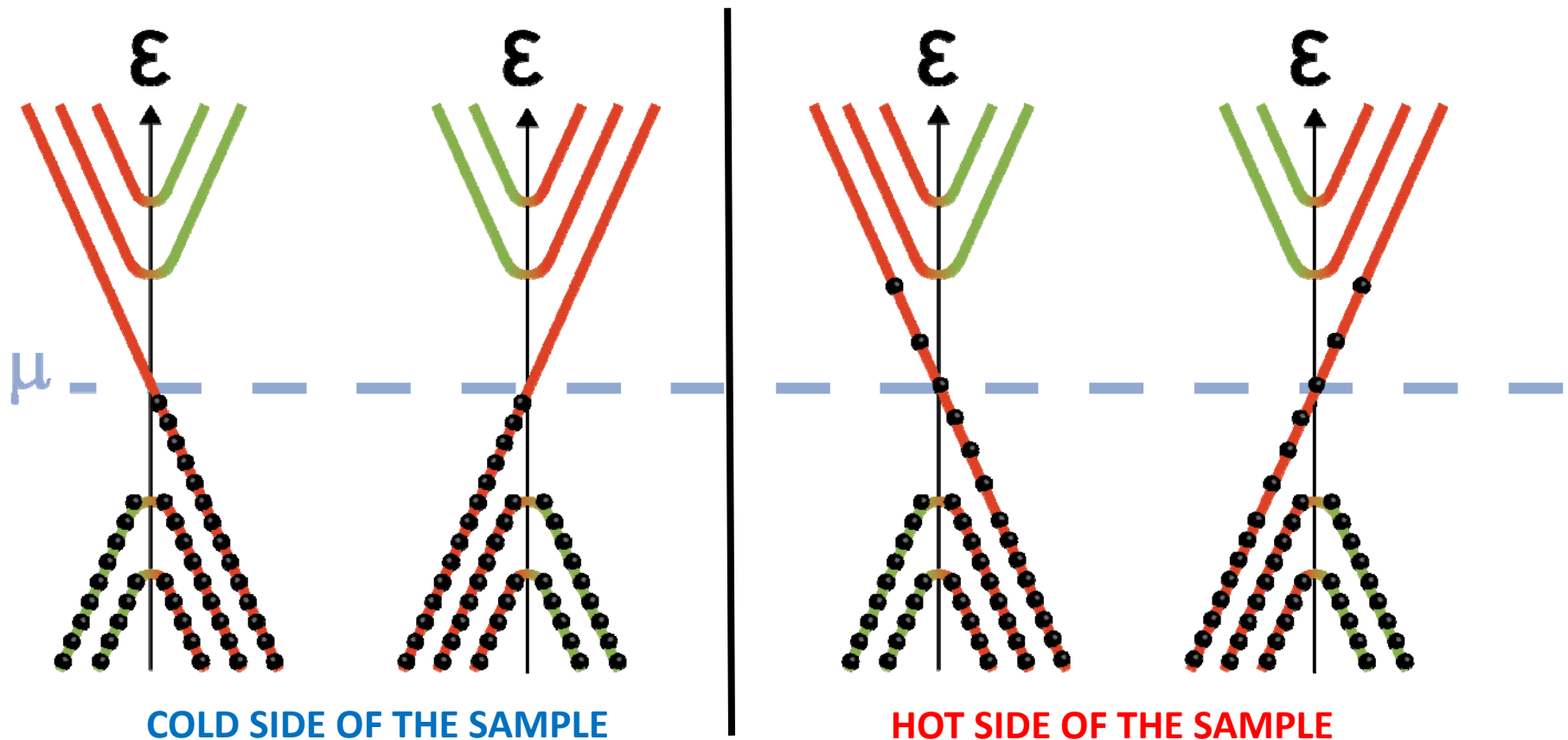
$$\sigma_A = N_w \frac{e^2 v \tau}{4\pi \hbar \ell_B^2} = N_w \frac{e^3 v \tau}{4\pi \hbar^2} B_z$$

$N_w$  = number of degenerate pairs of Weyl points

N. Trivedi and W. Zhang; *Spivak, N. Z. and Andreev, A. V., Phys. Rev. B* **93**, 085107 (2016)]

## Apply a temperature gradient $\nabla_r T \parallel B$

Thermal gradient BY ITSELF does NOT produce an imbalance between left movers and right movers.



The chiral anomaly requires an electric field: anomalous current  $j_A \propto \vec{E} \cdot \vec{B}$



## Effects of the anomalous velocity on thermal transport

Solve Boltzmann transport equations with both  $\mathbf{E} // \mathbf{B}$  &  $\nabla_r T // \mathbf{B}$

=> Change in carrier energy between left and right movers:

$$\delta\varepsilon_\chi = \frac{\chi e^2 \tau}{4\pi^2 \hbar^2} [\vec{B} \cdot \vec{E}] (\mu C_0 + C_1) + \frac{\chi e \tau}{4\pi^2 \hbar^2} \left[ \vec{B} \cdot \frac{-\nabla_r T}{T} \right] (\mu C_1 + C_2)$$

In an ideal Weyl semimetal:  $\mu = 0 \Rightarrow C_0 = 1; C_1 = 0; C_2 = \frac{\pi^2}{3} k_B T$

If  $\mathbf{E} = \mathbf{0}$  &  $\nabla_r T // \mathbf{B}$

$$\delta\varepsilon_\chi = -\frac{\pi^2}{3} k_B \frac{\chi e \tau}{4\pi^2 \hbar^2} [\vec{B} \cdot \nabla_r T]$$

$$\delta n_\chi = 0$$

Anomalous thermal conductivity, positive magneto-thermal conductivity

$$\kappa_A = N_w \frac{\pi^2}{3} \frac{v \tau k_B^2 T}{4\pi \hbar \ell_B^2} = N_w \frac{\pi^2}{3} \frac{e v \tau k_B^2 T}{4\pi \hbar^2} B_z$$

## Summary Ideal Weyl semimetal ( $\mu = 0$ )

1. Electric field only: 
$$\delta n_\chi = \frac{\chi e^2 \tau}{4\pi^2 \hbar^2} [\vec{B} \cdot \vec{E}]$$
$$\delta \varepsilon_\chi = 0$$

⇒ **Change in carrier density** between left and right movers

⇒ **No change in carrier energy**

2. Thermal gradient only: 
$$\delta \varepsilon_\chi = -\frac{\pi^2}{3} k_B \frac{\chi e \tau}{4\pi^2 \hbar^2} [\vec{B} \cdot \nabla_r T]$$
$$\delta n_\chi = 0$$

⇒ **Change in carrier energy** between left and right movers

⇒ **No change in carrier density**

3. Ratio of the two: 
$$\kappa_A = LT \sigma_A \quad L = L_0 = \frac{\pi^2}{3} \left( \frac{k_B}{e} \right)^2$$

⇒ **The Wiedemann-Franz law holds with the free electron Lorenz ratio**

if the inelastic scattering rate is dominated by the helicity and  $\mu=0$  (ideal Weyl)



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2. Thermal conductivity

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[arXiv:1906.02248](https://arxiv.org/abs/1906.02248)

# Problem #1 with magnetoresistance: current jetting

High-mobility materials:

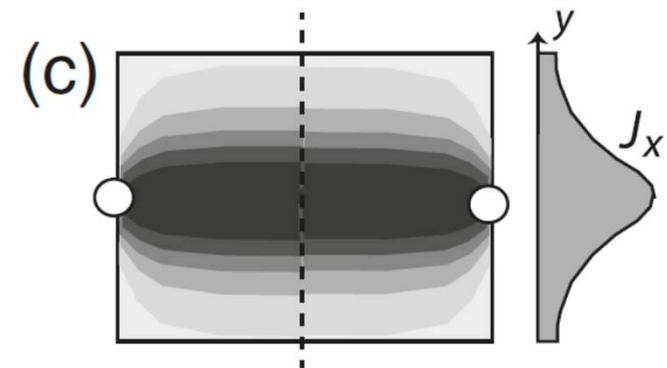
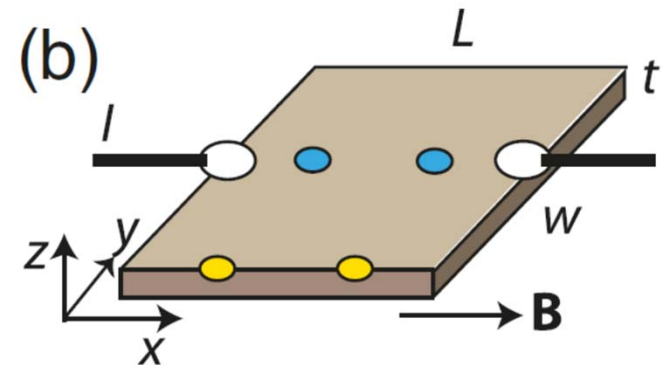
4-probe longitudinal magnetoresistance is OFTEN negative due to the distortions of the current lines in the sample.

Purely geometrical effect

Current bundles up in the middle of the sample

Voltage wires loose contact to the region of the sample with the current

=> looks like negative longitudinal MR, but it isn't.



*Liang, ..., Cava, Ong, PRX 8, 031002 (2018)*  
*Felser group, arXiv 1606.03389*

## Problem #2 : Geometrical magnetoresistance

In high-mobility nanoscale materials and transverse magnetic fields: current lines follow the Hall angle.

High field: diameter of the helix  $<$  sample size **(b)**  
=> surface scattering is suppressed  
=> resistivity goes down

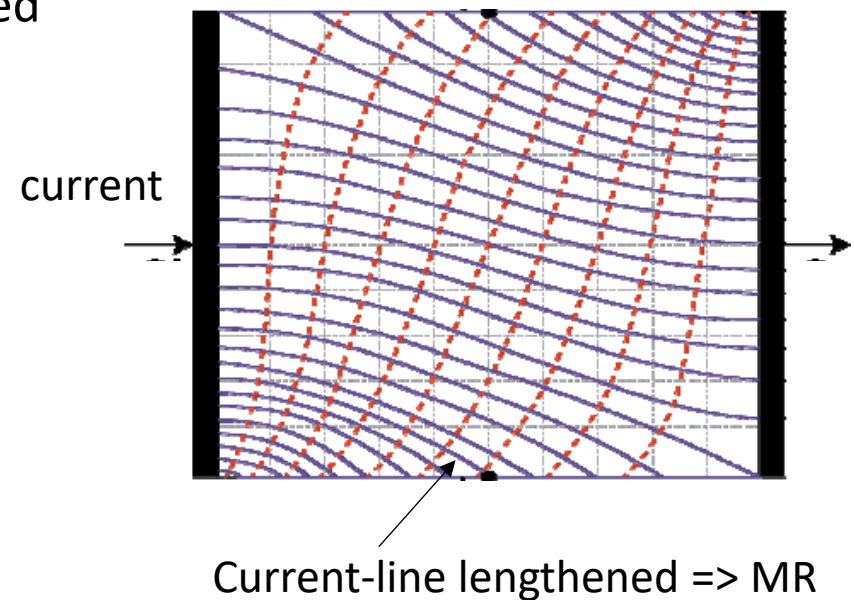
Purely geometrical effect

Apparent positive transverse MR,  
But is a side-effect of very slight  
Field misalignments

Appears even in 2-wire measurements.

Order of magnitude:  $R(B) / R_0 = 1 + \mu^2 B^2$

In our samples,  $\mu \sim 10^6 \text{ cm}^2/\text{Vs}$  => 1 degree misalignment 1 Tesla gives 200% MR



## *Thermal conductivity measurements*

Experimentally much easier than magnetoresistance measurements:

1. No electrical contacts => no circulating currents  
=> no Lorentz force of the current lines

2. Thermal conductivity has an electronic contribution and a lattice contribution.

The physics we seek to measure arises from the electronic contribution

The lattice thermal conductivity little affect by magnetic field  
=> The lattice conductivity redistributes the heat flux lines

3. Summary: thermal measurements much less sensitive to problems.



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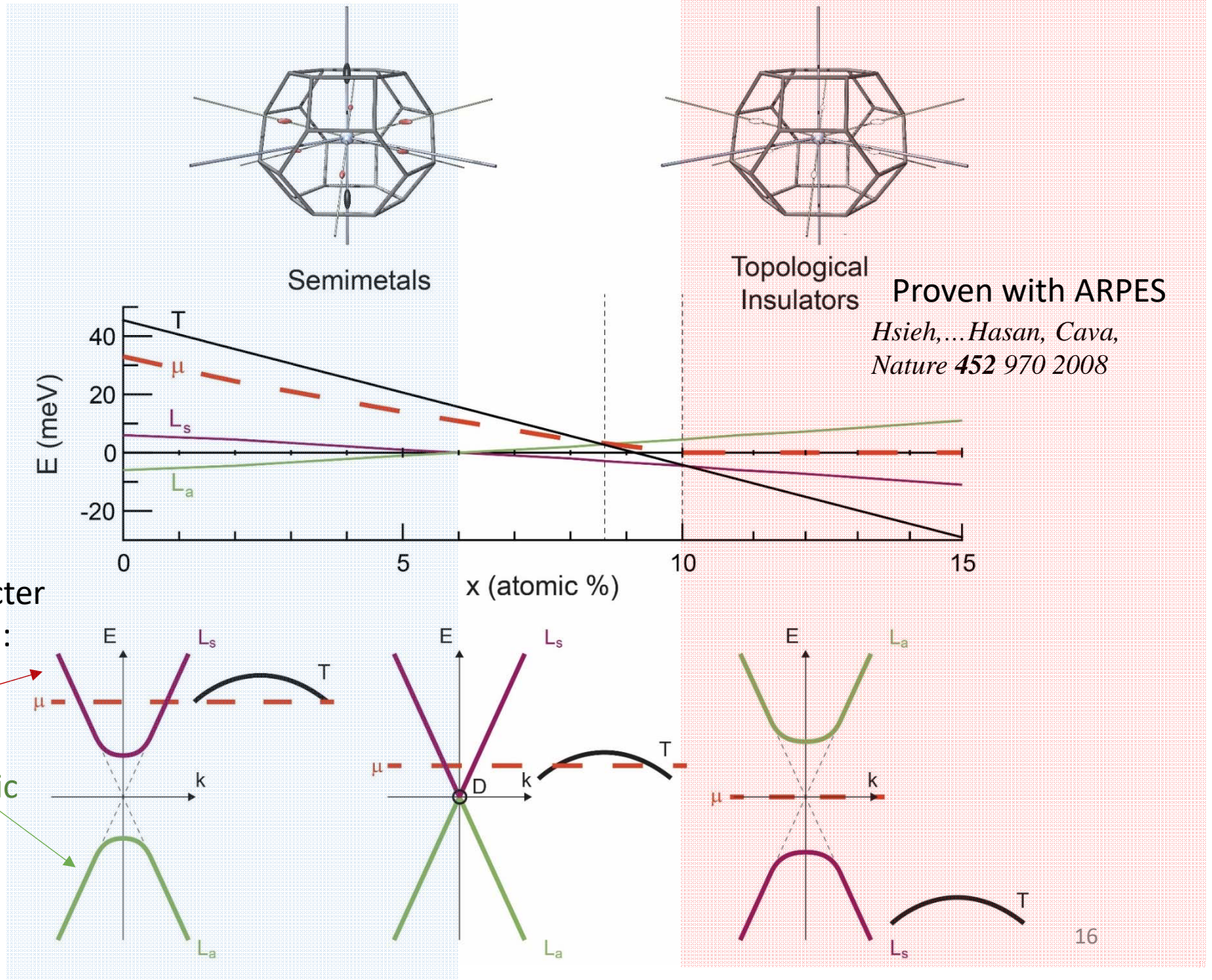
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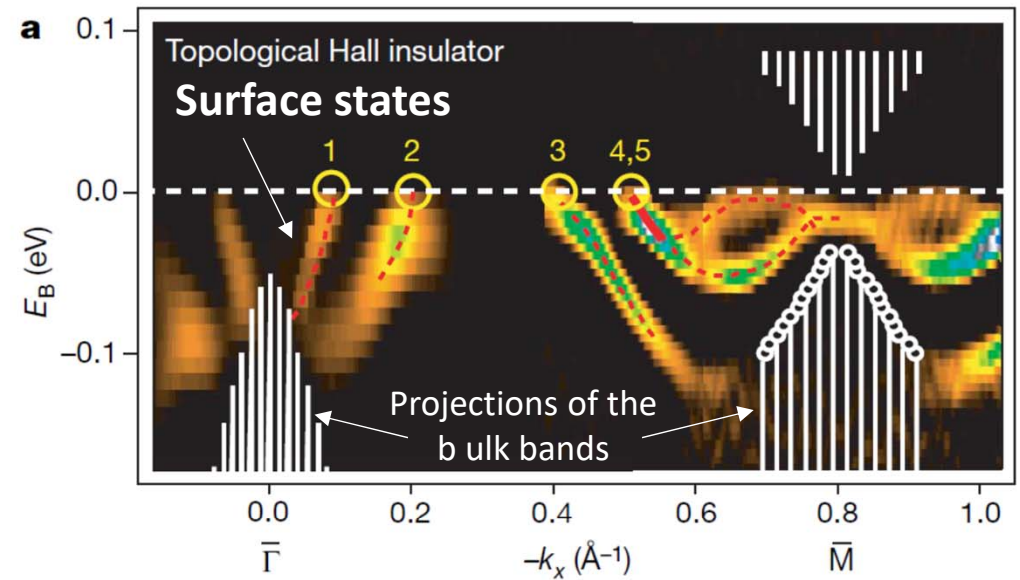
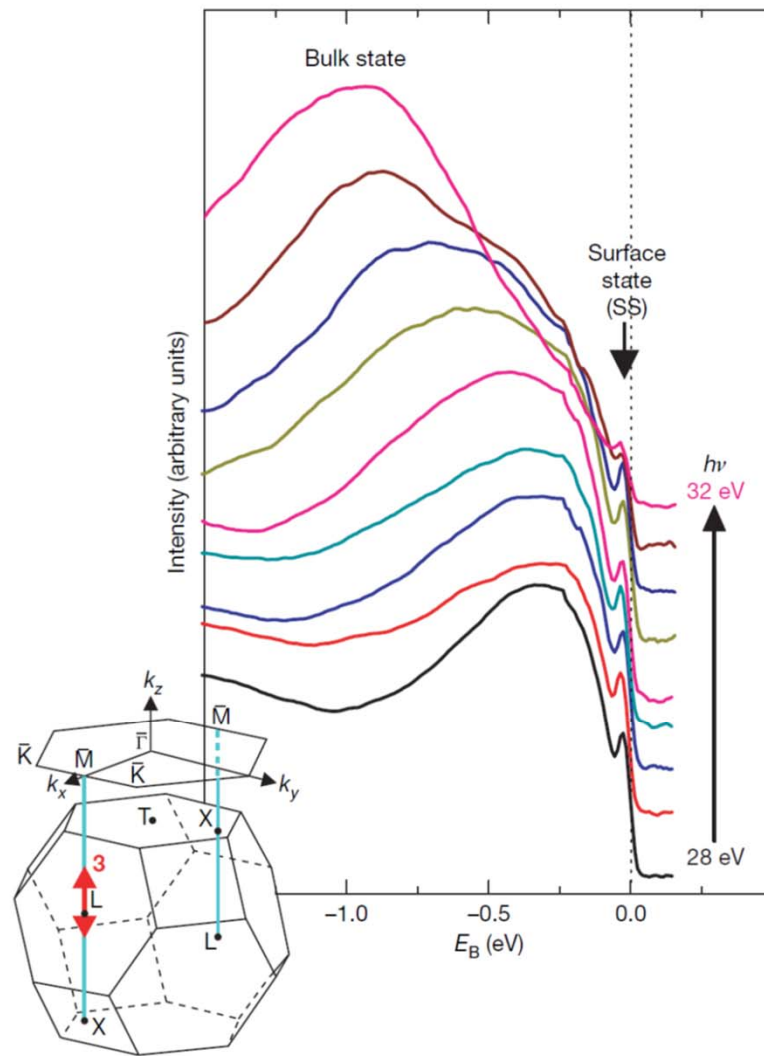


# The $Bi_{1-x}Sb_x$ semiconductors and semimetals





# ARPES Surface and bulk states in TI $\text{Bi}_{88}\text{Sb}_{12}$



Hsieh, ... Hor, Ong, Hasan, Cava, *Nature* **452** 970 2008

## Last Landau level CLOSES the gap at L-point

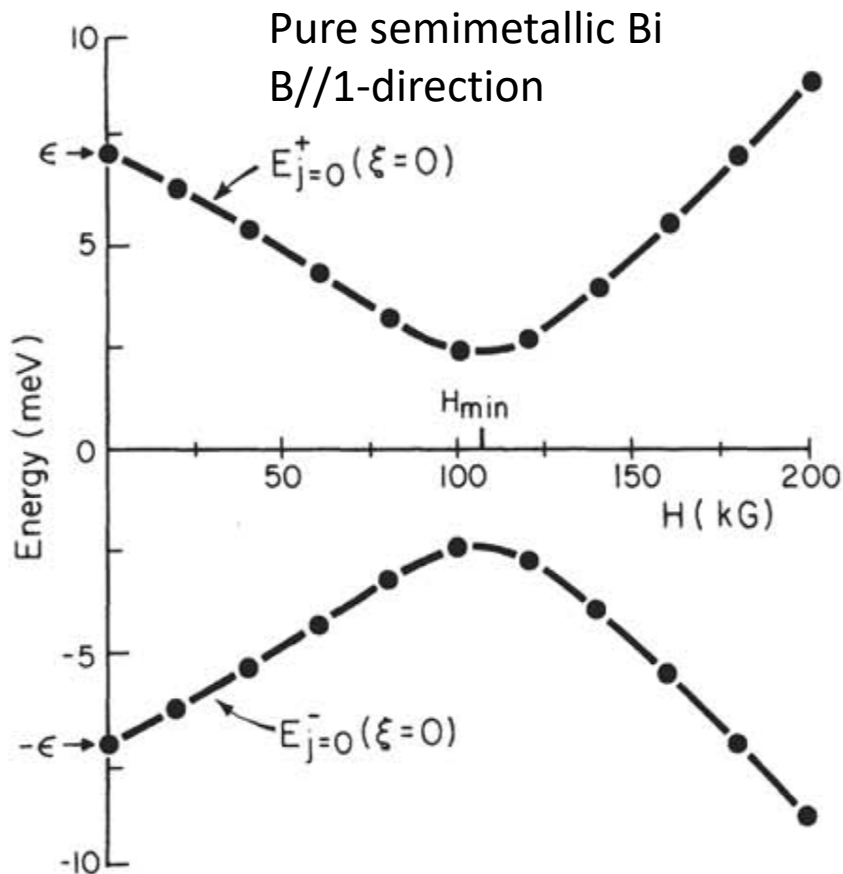


FIG. 2. Magnetic field dependence of the  $j = 0$  energy levels at  $\xi = 0$  ( $k_3 = 0$ ) for the light binary electrons of Bi.

Magnetic field dependence of the Landau level energies at  $k_z = 0$

$$E(k_z = 0) = (n + \frac{1}{2})\hbar\omega_C + sg\mu_B B$$

The Landé factors of  $\text{Bi}_{1-x}\text{Sb}_x$  are enormous, diamagnetic and anisotropic.

Pure semimetal Bi: **bands closing observed experimentally** with magneto-optics

Same effect is calculated for  $\text{Bi}_{1-x}\text{Sb}_x$  alloys

Effect small in binary (1) field, much larger in trigonal (3) field => crossing near 1 T.

# Ultraquantum TI's become field-driven Weyls

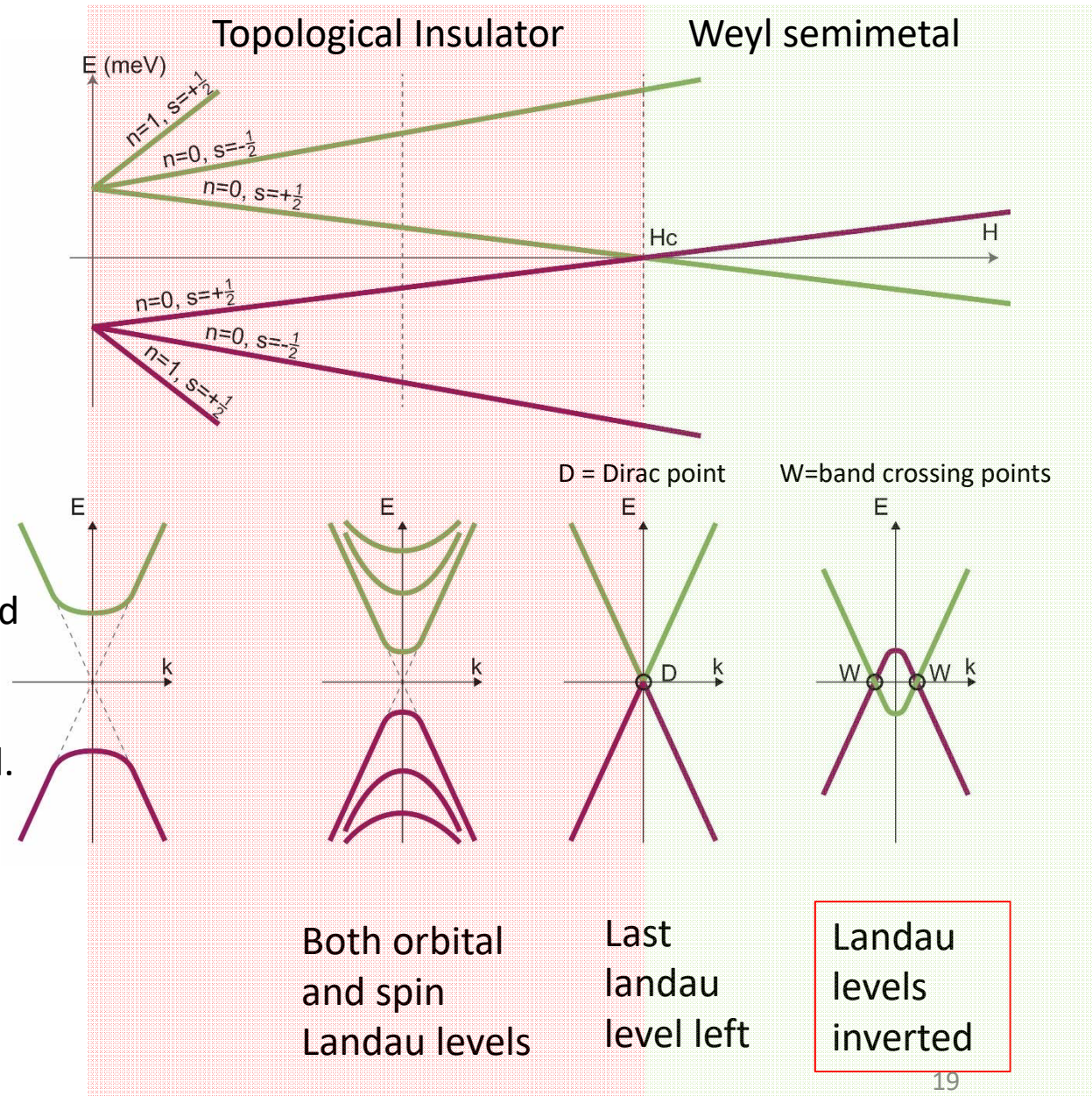
The Landé factors of  $\text{Bi}_{1-x}\text{Sb}_x$  are enormous, diamagnetic and anisotropic.

⇒ The Landau levels can be made to close with field

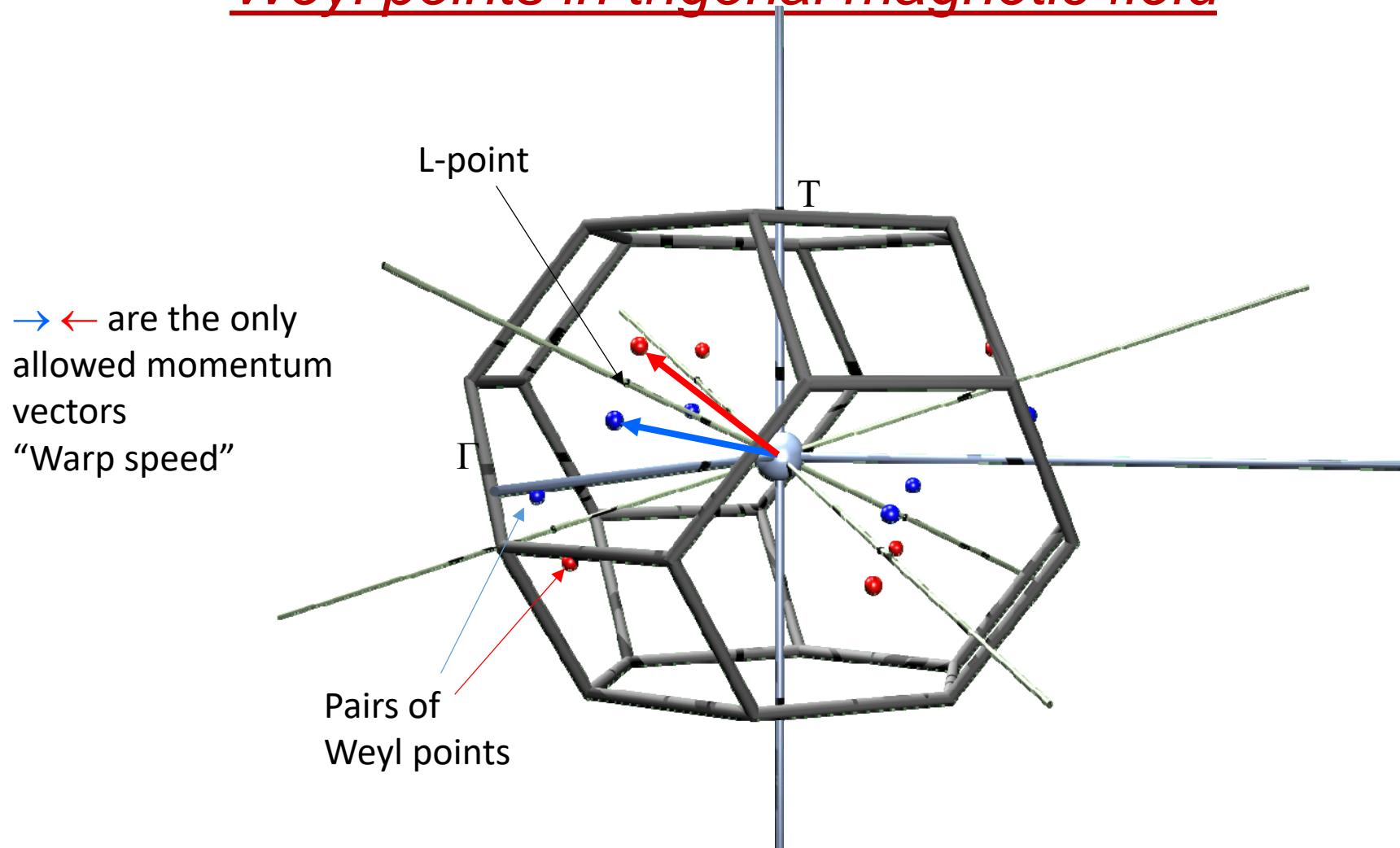
⇒ The bands can be made to invert a second time

⇒ These are single spin-polarized Landau levels

⇒ Second inversion gives a Weyl.



## Weyl points in trigonal magnetic field



- Painstakingly identified, calculation in supplemental slides
- No trivial pockets to the Fermi surface.
- 3 degenerate pairs of Weyl points per unit cell
- If we can pin the electrochemical potential to the Weyl points we have the ideal Weyl



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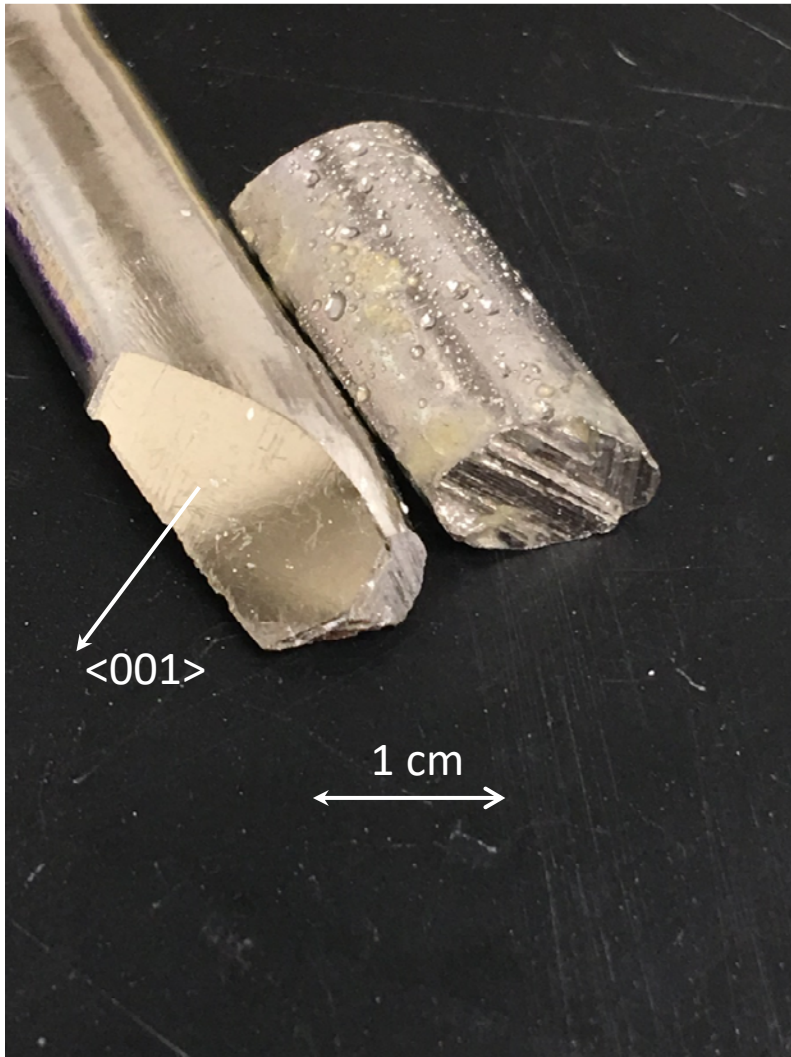
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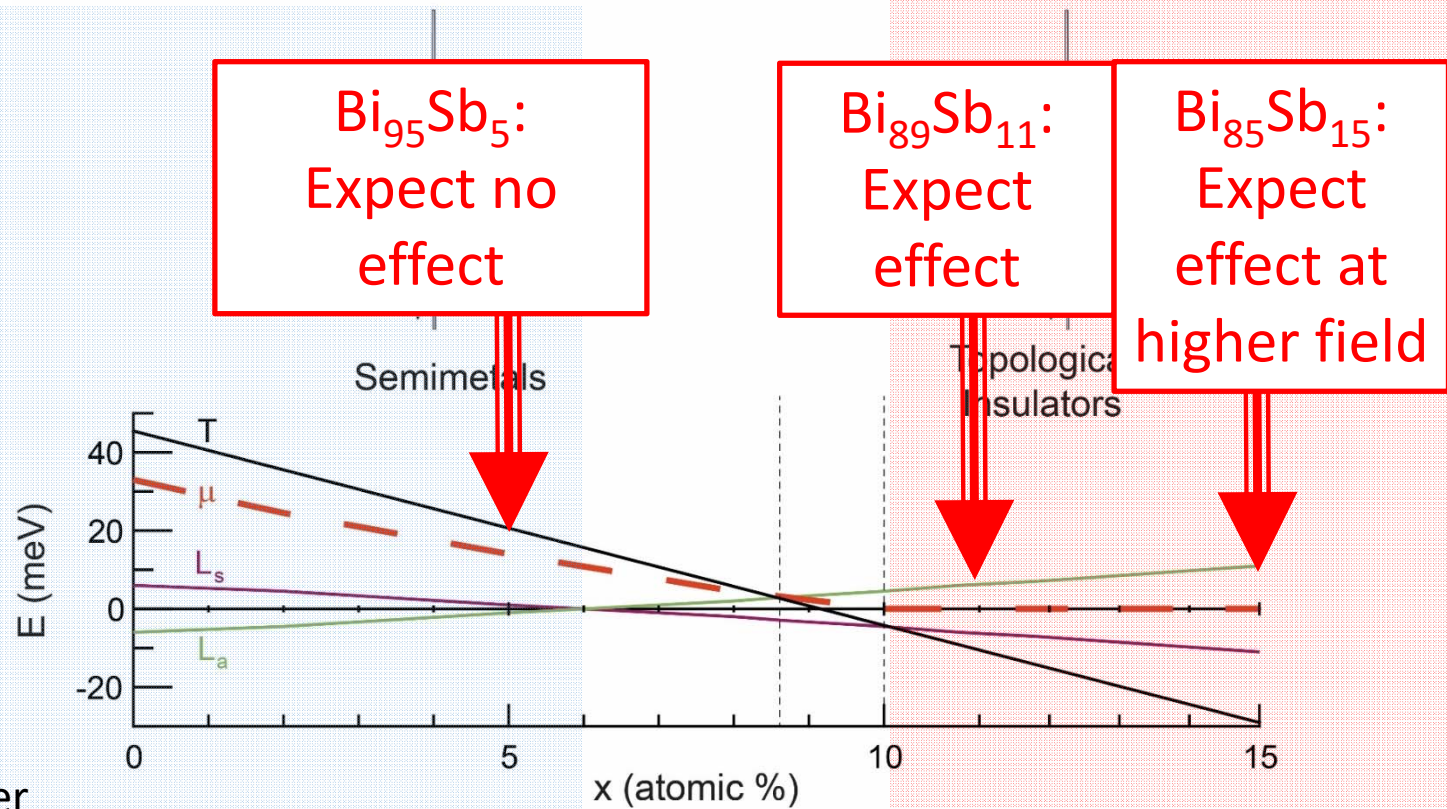


# Samples



- Bi and Sb are isoelectronic  
=> no problem with stoichiometry control
- Full solid solution Bi<sub>1-x</sub>Sb<sub>x</sub> ( $0 \geq x \geq 1$ )  
=> Can be prepared with exquisite purity and perfection  
=> => Extraordinary mobility ( $> 10,000,000$  cm<sup>2</sup>/V s at 4K for pure Bi)
- Starting material must be purified in-house by zone-melting
- Crystals grown in-house
- Measured 6 samples  $x=11\%$  with consistent results, and  $x=15$ ,  $x=5\%$ .
- Uniformity  $x \sim 1\%$ (nominal) checked by XRD and XRF
- Extremely low carrier concentrations => electrochemical potential at Weyl points

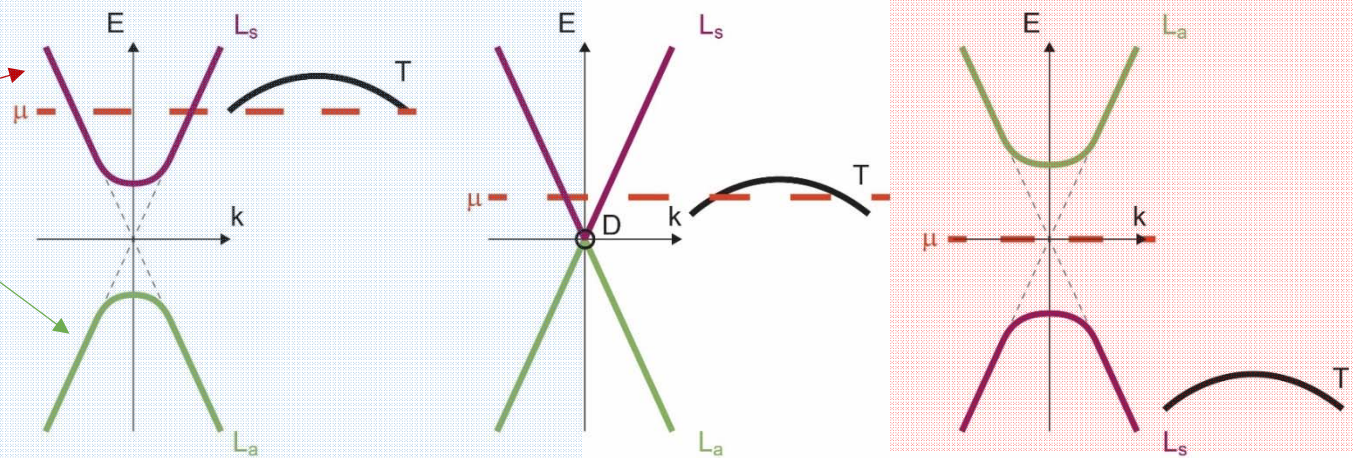
# Samples for this study



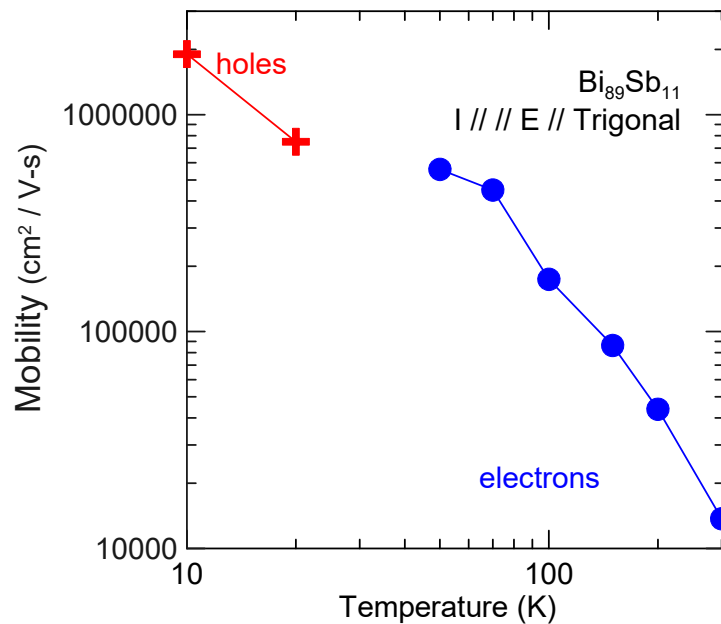
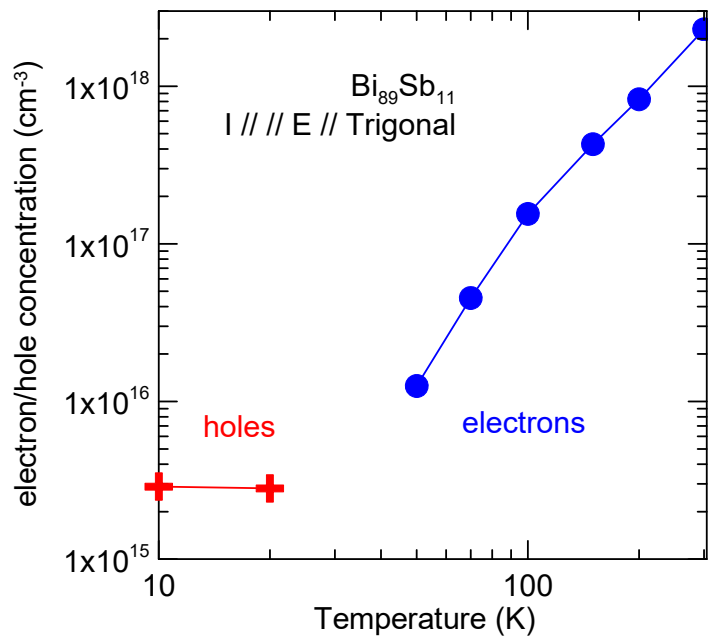
Orbital character  
Wavefunction:

Symmetric

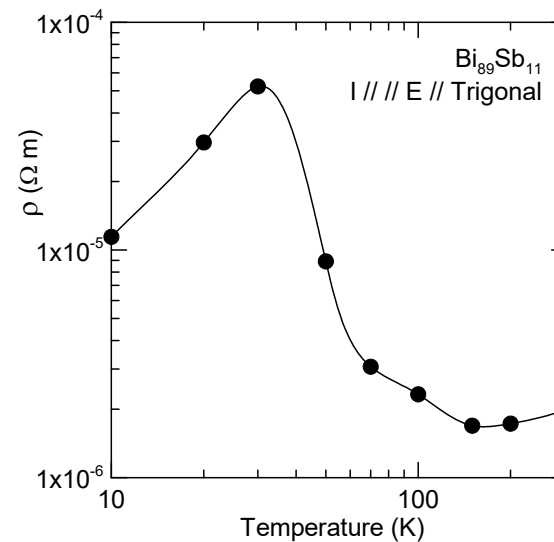
Antisymmetric



# Superb single crystals

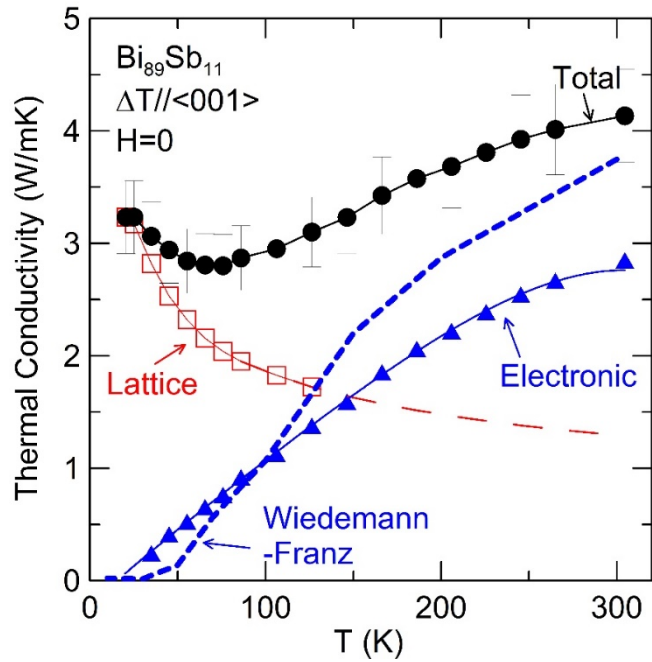


- Very low-field Hall measurements,  $B //$  bisectrix (no band movement)
- Freeze-out achieved: carrier concentration reduced to  $2 \times 10^{15} \text{ cm}^{-3}$  ( $T < 20\text{-}40\text{K}$ )
- Chemical potential at energy of minimum DOS, i.e. midgap at zero field, at Weyl point in field.
- Mobility
  - at 10K:  $2,000,000 \text{ cm}^2 / \text{V s}$
  - At 100K:  $200,000 \text{ cm}^2 / \text{V s}$





# Thermal conductivity $\kappa_{33}$ trigonal direction, zero field (TI)

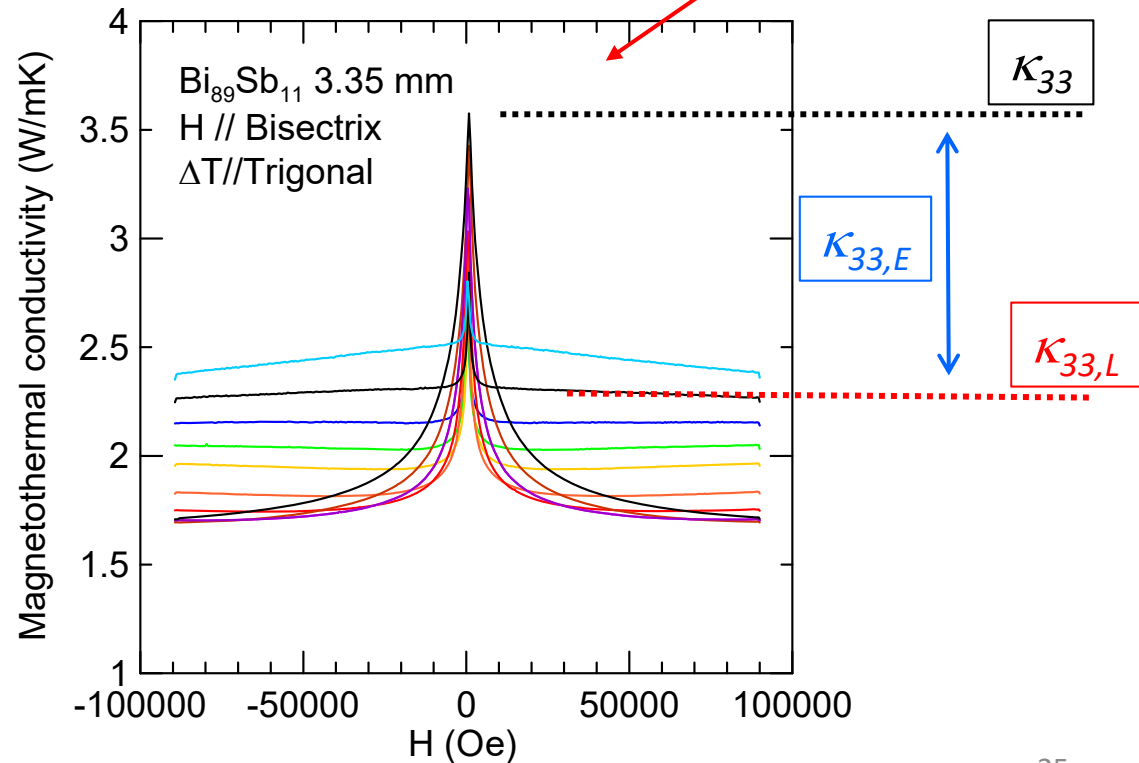


- T (K):
- 34
  - 44
  - 54
  - 64
  - 75
  - 85
  - 105
  - 125
  - 145
  - 164

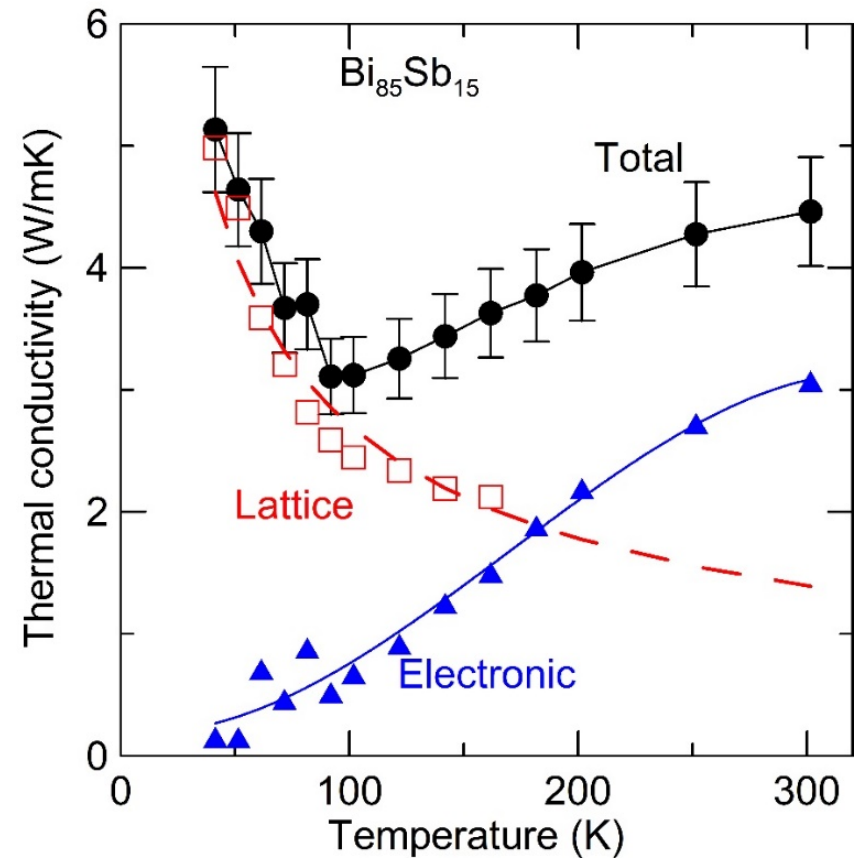
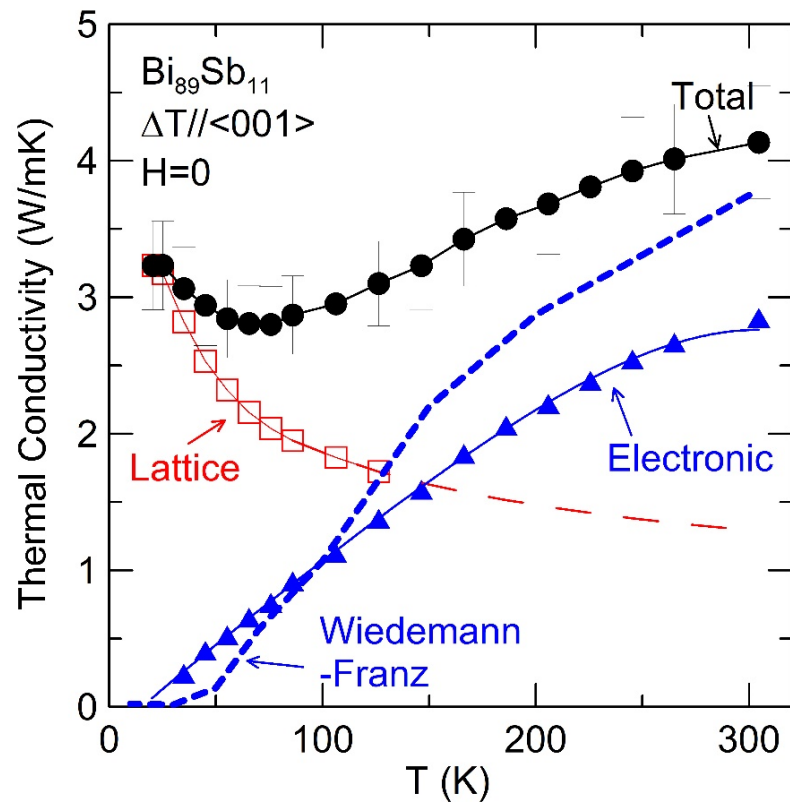
$$\kappa_{33} = \kappa_{33,E} + \kappa_{33,L}$$

Measure      Isolate      Subtract

Obtain  $\kappa_{33,L}$  from measurements of  $\kappa_{33}(B_2)$



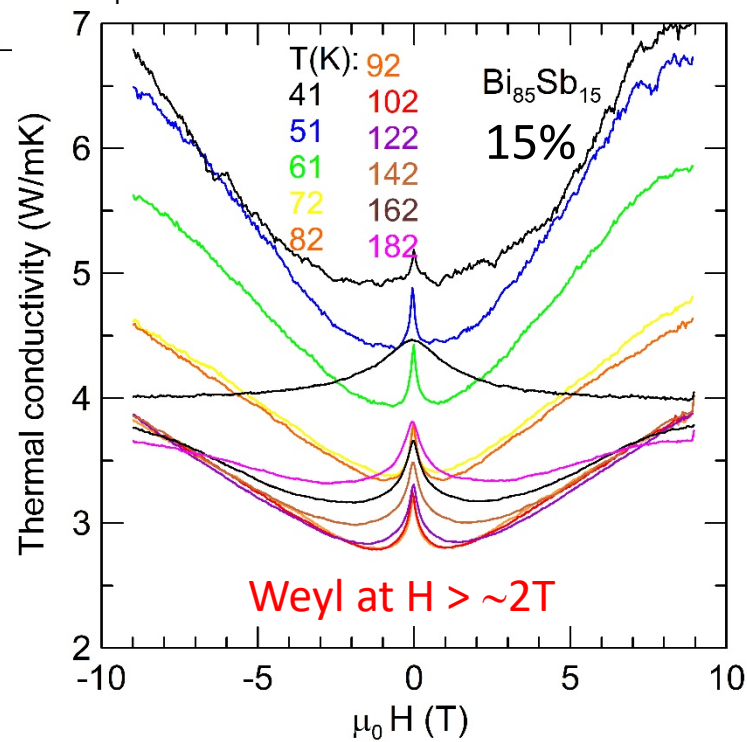
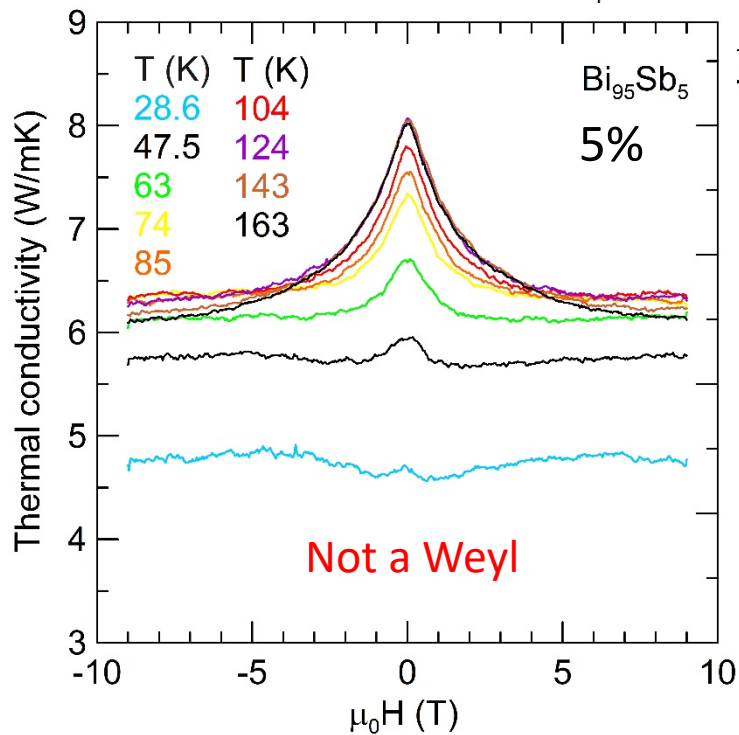
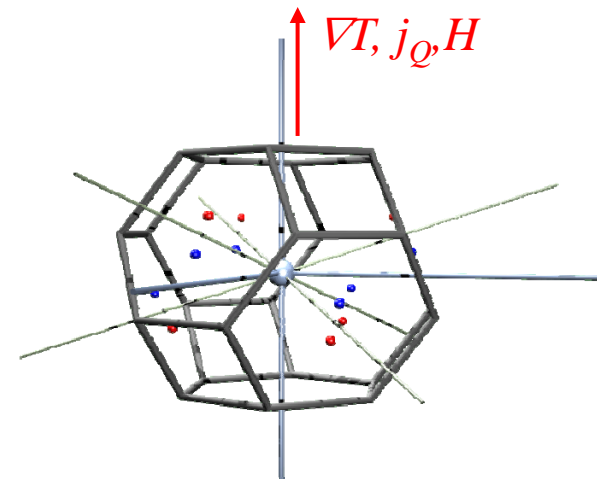
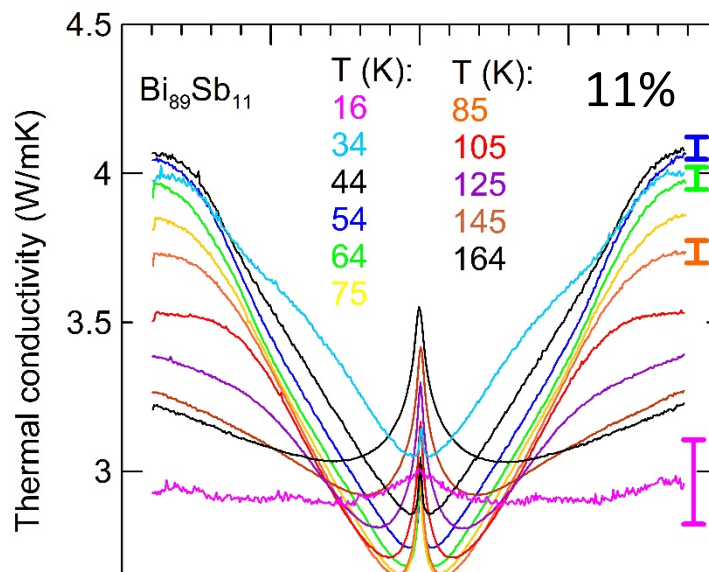
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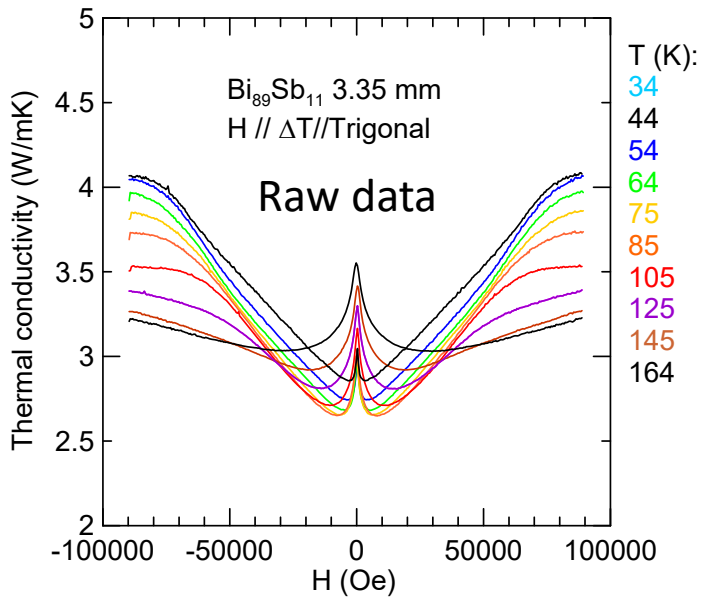
Dashed line: Wiedemann-Franz law, for later

# Data: $\kappa_{33}$ ( $B_3$ )

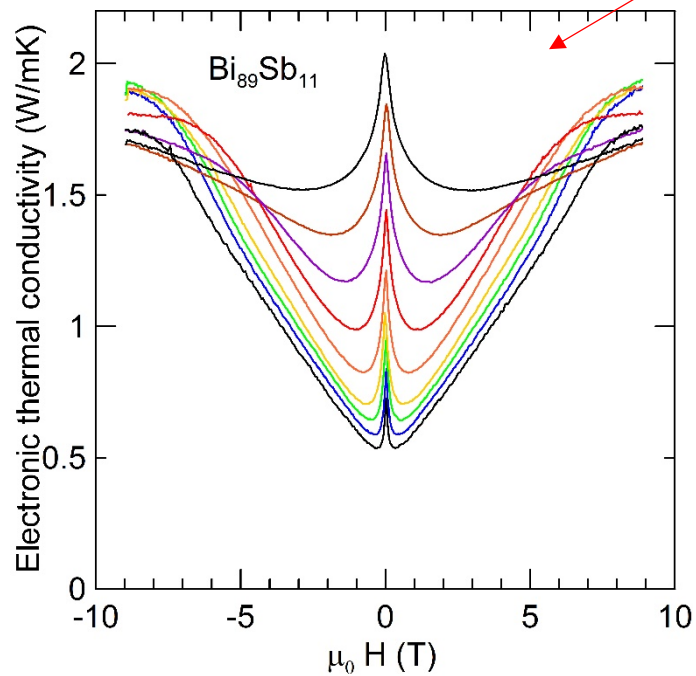
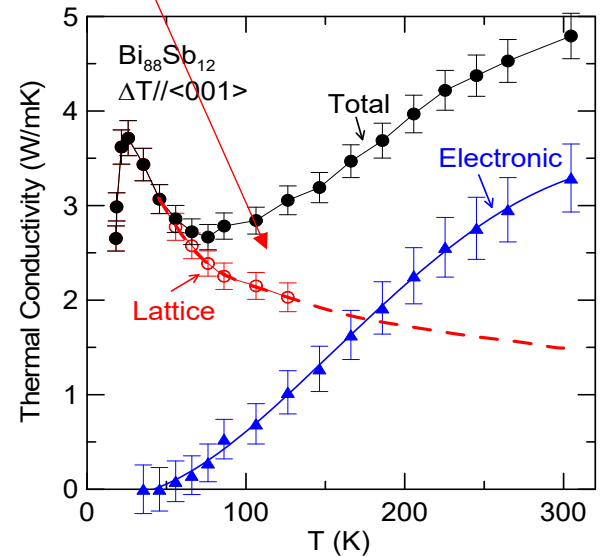
Weyl  
at  $H > \sim 1$  T



# Electronic contribution to $\kappa_{33}$ ( $B_3$ )



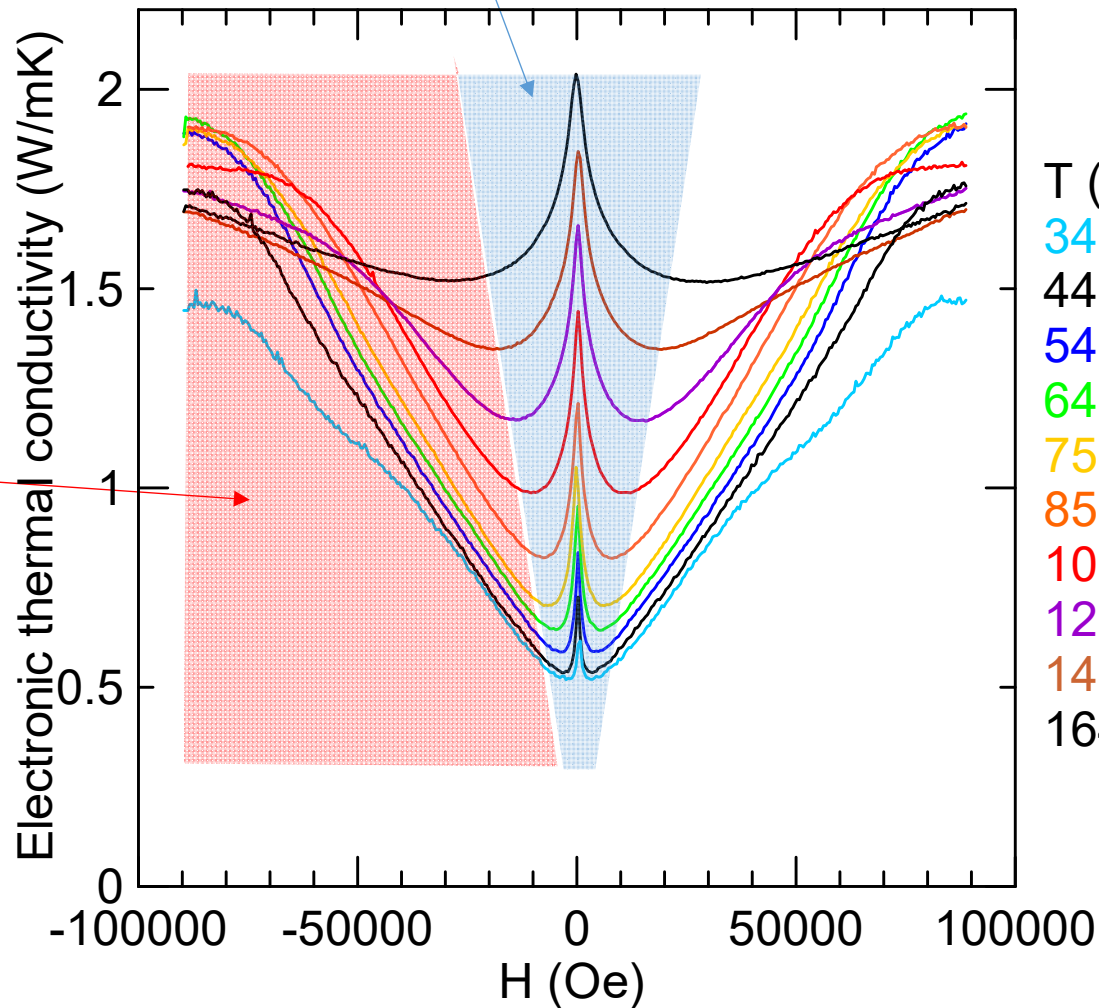
Subtract the lattice part



# Electronic thermal conductivity

Low-field: conventional TI

Thermal conductivity decreases with field: normal magnetoresistance  
decreases electronic contribution to thermal conductivity



Thermal conductivity increases:

Claim: the Chiral energy unbalance term

Analyzed next

T (K):

34

44

54

64

75

85

105

125

145

164

Effect persists to  $T > 160$  K  
=> Effect is robust to phonon scattering

Sample 1



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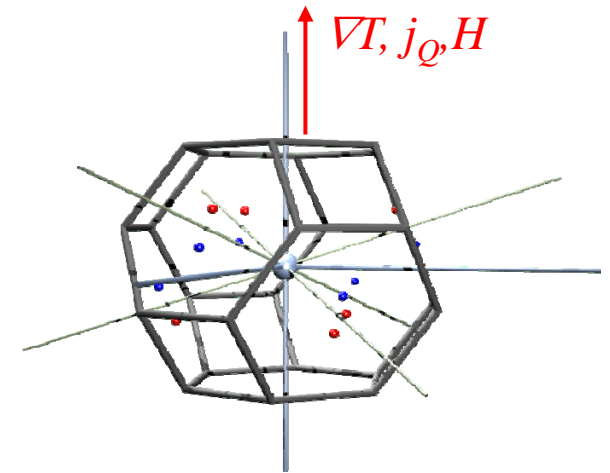
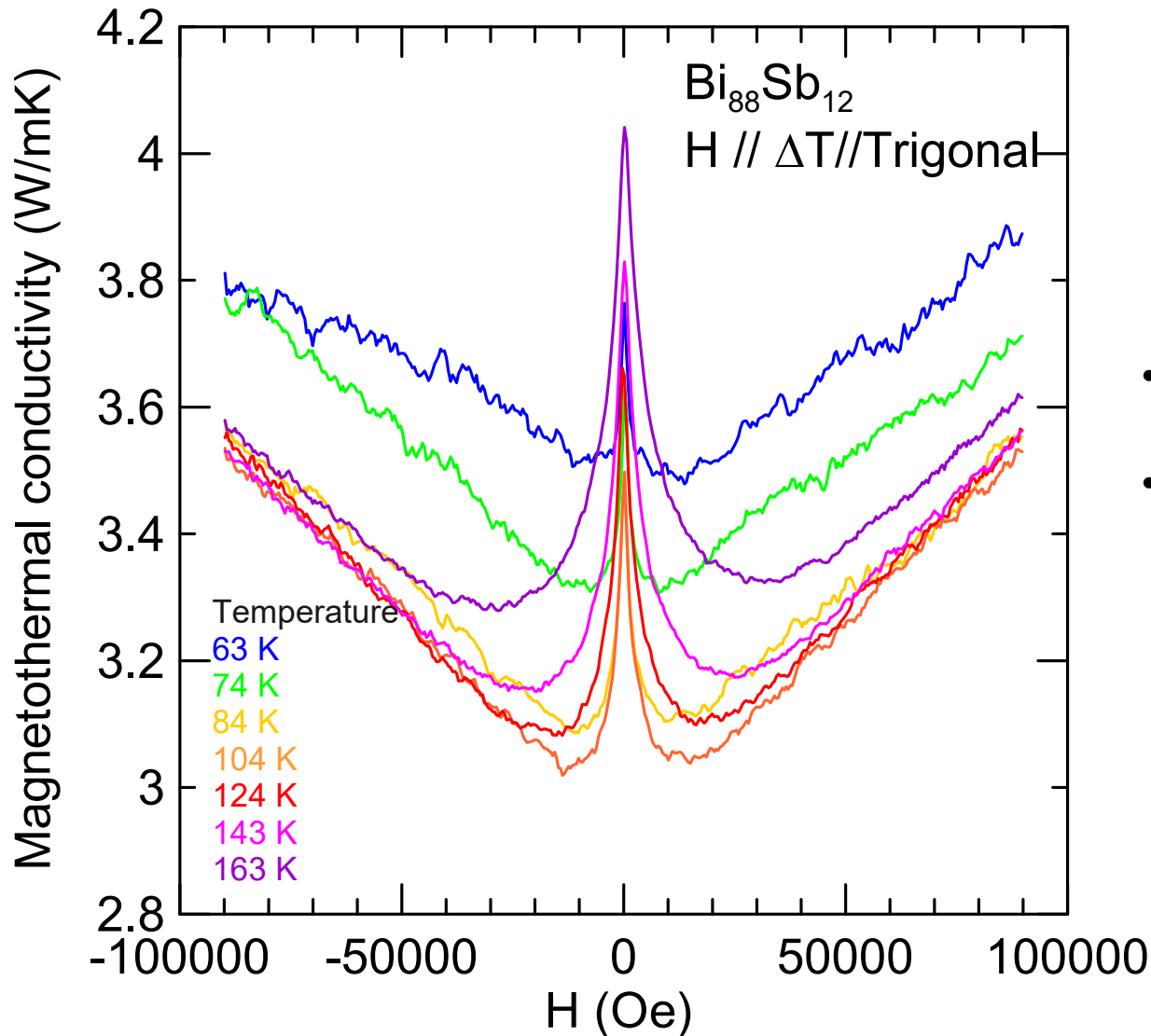
## *Rules for experimentalists*

If you think you see an effect, you must also be able to:

1. Reproduce it on multiple samples.
2. Deliberately make it go away.

## ✓ Reproduce on Sample 2: $\text{Bi}_{88}\text{Sb}_{12}$

And sample 3 and sample 4



- Lower mobility  $2 \times 10^4 \text{ cm}^{-3}$  at 10K
- Residual doping n-type  $1 \times 10^{16} \text{ cm}^{-3}$

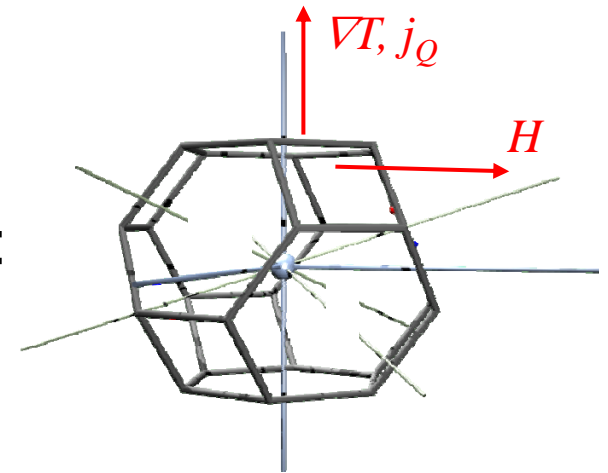
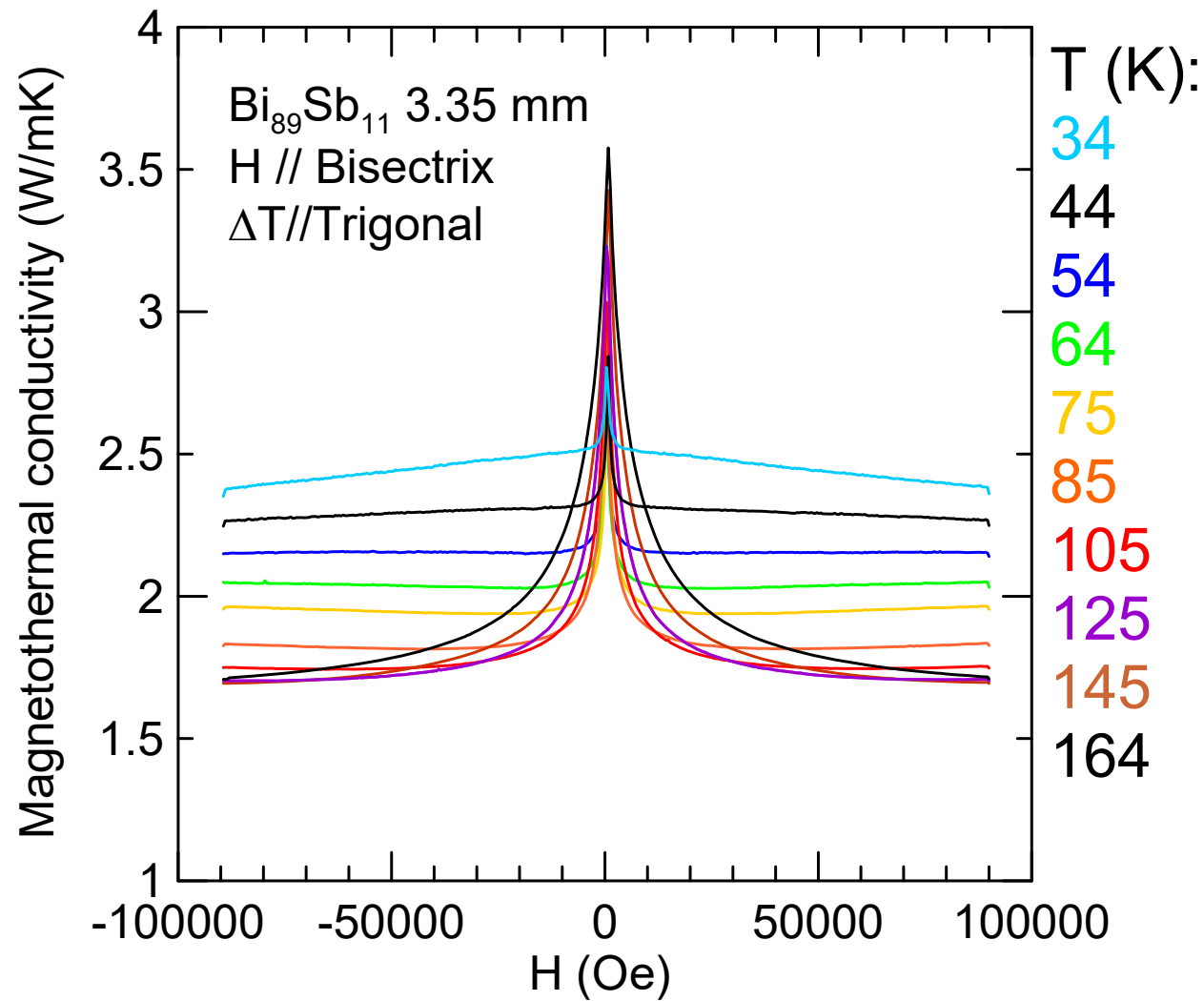
Effect persists in samples with 100 times lower mobility  
=> Effect is robust to defect scattering



✓ Can make the effect go away:

Transverse field

Trigonal thermal conductivity sample 1



Field in wrong  
direction =>

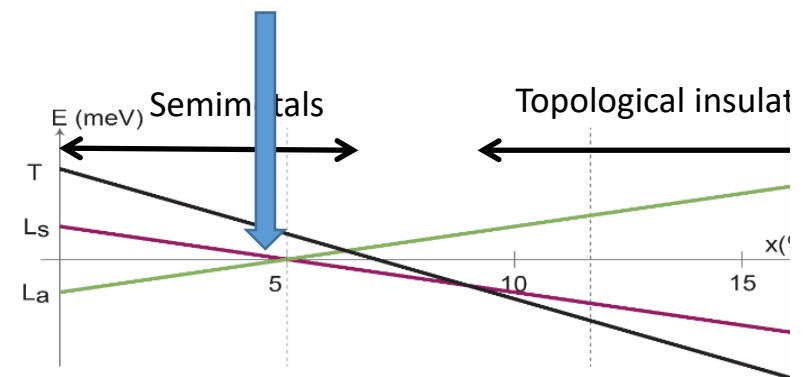
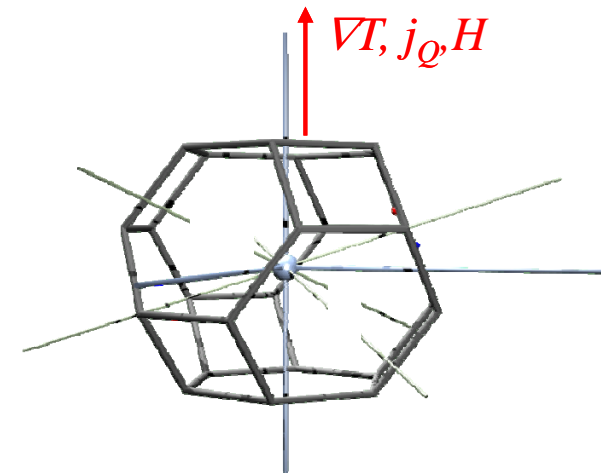
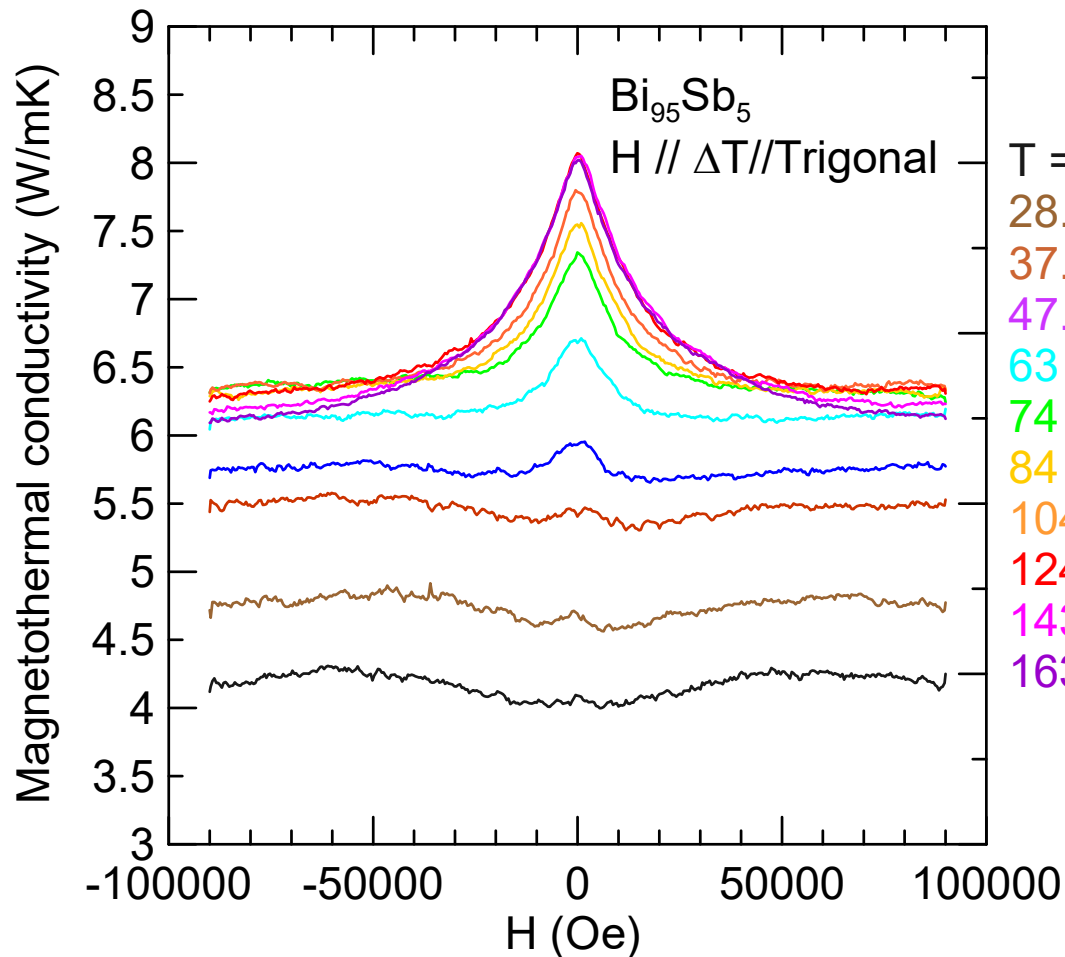
No chiral anomaly

Should **NOT** show the  
effect

✓ Can make the effect go away:

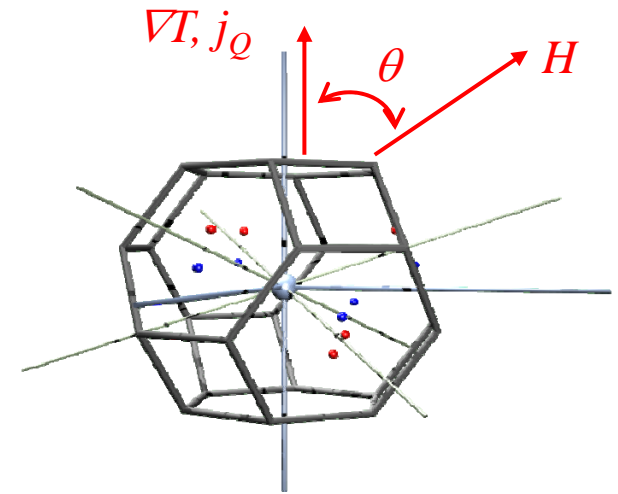
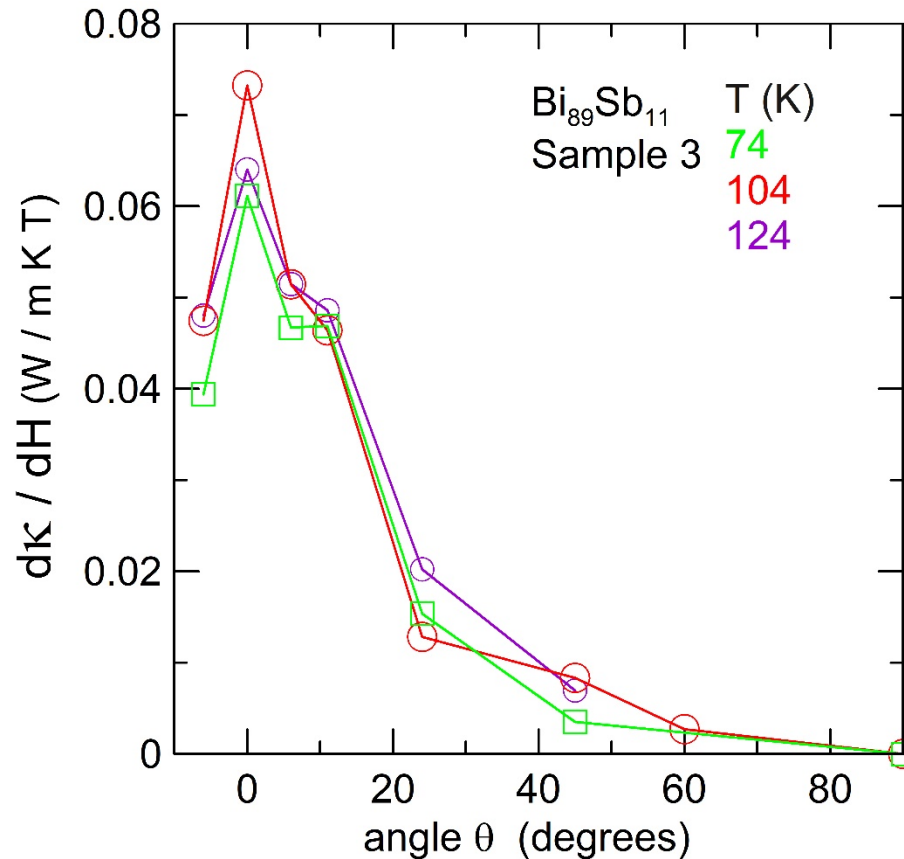
Composition at the Dirac point

Trigonal thermal conductivity  $\text{Bi}_{95}\text{Sb}_5$



- Geometry is correct
- Bands are not inverted
- $\Rightarrow$  no Weyl points
- Should **NOT** show the effect

## Angular dependence: angle from 3-axis in 3-2 plane

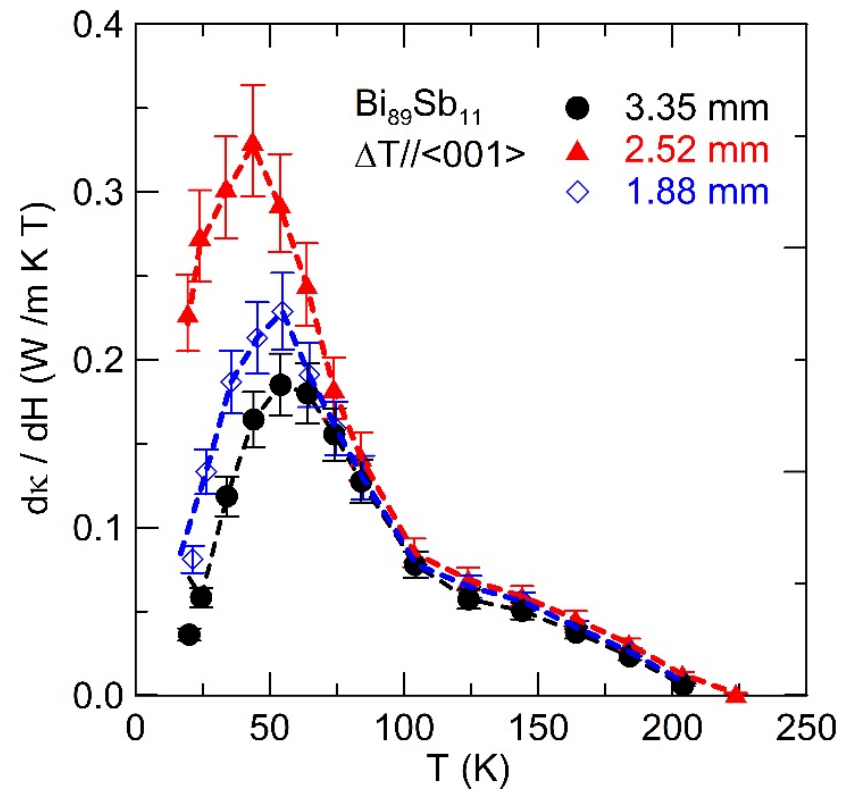


- Much sharper dependence than  $\cos(\theta)$  or  $\cos^2(\theta)$ , possibly  $\cos^N(\theta)$  with  $N > 6$
- Signature feature of electrical chiral anomaly

## Temperature and sample length dependence

- Slope of thermal conductivity at 7 Tesla
- No length dependence
- Temperature dependence analyzed next

Sample 1





1. Introduction:

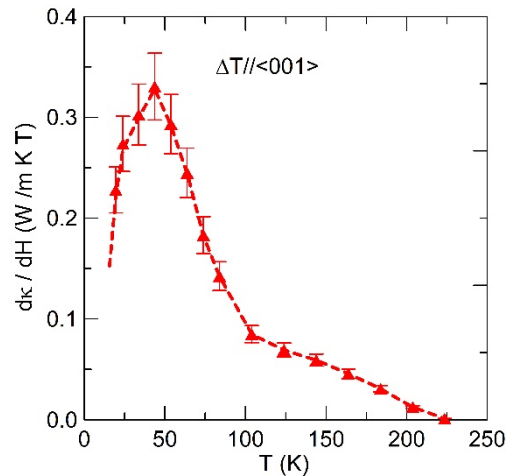
- Weyl semimetals: chiral anomaly and and thermal conductivity
- Experimental difficulties
- Bi-Sb semiconductors alloys and TI's
- In the ultra-quantum limit: field-induced Weyls

2. Thermal conductivity

- Experimental evidence
- Robustness to phonons and defects
- **Decay only via inter-Weyl point scattering**
- The Wiedemann-Franz law

[arXiv:1906.02248](https://arxiv.org/abs/1906.02248)

## Analysis of temperature-dependence, $k_B T > \mu$



1. Take these raw data for experimental temperature dependence at 7 T of  $d\kappa_{zz}/dB_z$

2. Compare to the formula for the anomalous conductivity  $d\kappa_{zz}/dB_z = \frac{\pi e v_F k_B^2}{\hbar^2} T \tau$

3. Solve for  $\tau$  and plot versus  $1/T$

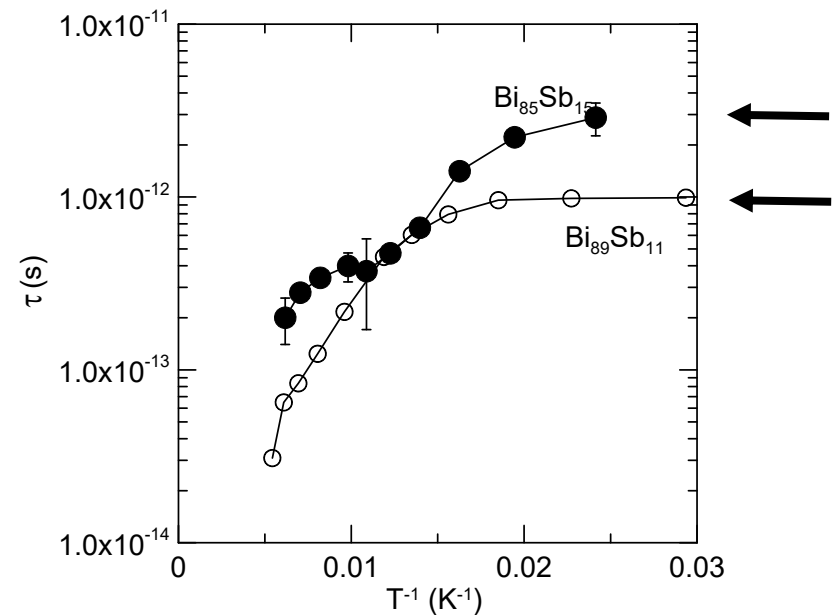
4. For  $T < 50 K$ ,

$Bi_{89}Sb_{11}$   $\tau \sim 1 \times 10^{-12} s$

$Bi_{85}Sb_{15}$   $\tau \sim 3 \times 10^{-12} s$

$\tau$  Is T-independent

$\tau$  Is 10x larger than the resistivity scattering time

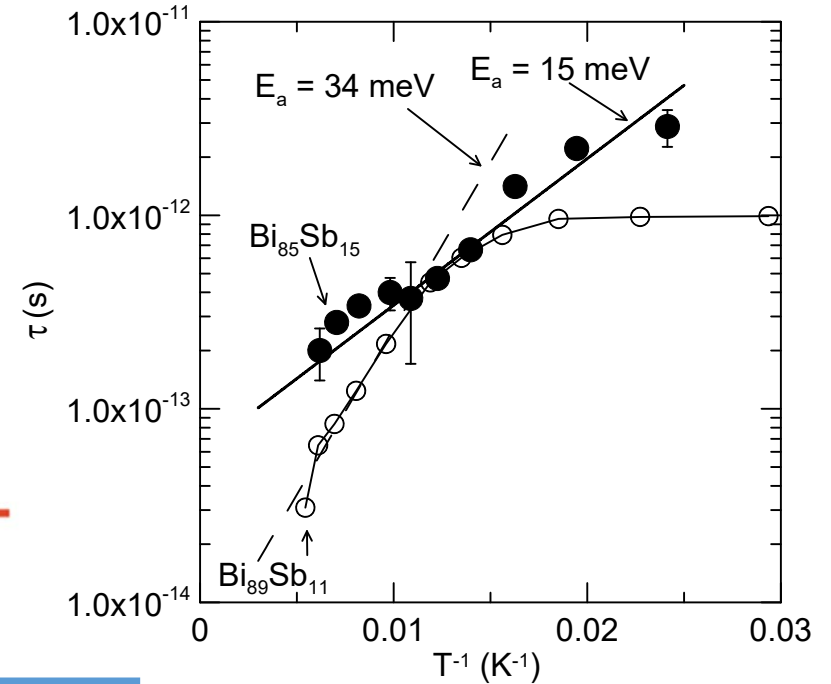
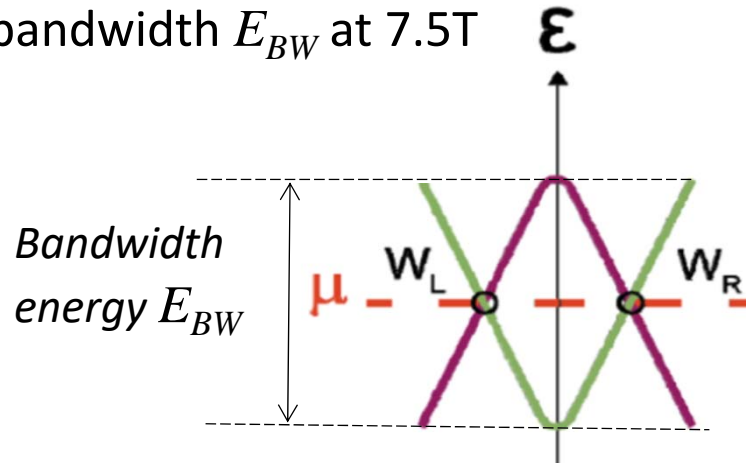


## Analysis of temperature-dependence, high-T

6.  $T > 100$  K: fit an activation-energy to  $\tau(T)$ :

$$\tau(T) = \tau_0 e^{E_A/k_B T}$$

7. Calculated bandwidth  $E_{BW}$  at 7.5T



Composition	Experimental $E_A$	Calculated $E_{BW}$
	meV	meV@7.5T
$Bi_{89}Sb_{11}$	$34 \pm 2$	35
$Bi_{85}Sb_{15}$	$15 \pm 2$	20

- $\Rightarrow \tau$  is the inter-Weyl point scattering time, thermal activation
- $\Rightarrow$  The only energy scale in the observation is the width of the Weyl bands
- $\Rightarrow$  The Wiedemann-Franz law is expected to hold



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• The Wiedemann-Franz law:

$$K = K_E + K_L$$

Measure      Isolate      Subtract

• Can we prove experimentally

- That the electronic contribution  $K_E = LT\sigma$

- with  $L = L_o = \frac{\pi^3}{3} \left( \frac{k_B}{e} \right)^2 = 2.45 \cdot 10^{-8} \left( \frac{V}{K} \right)^2$



# Wiedemann-Franz law (WFL) $\kappa_E = LT\sigma$

Predictions for regime of chiral anomaly:

- If inelastic relaxation rate ( $\tau_{eff}^{-1}$ ) is dominated by the helicity ( $\tau_h^{-1}$ ):  $L = L_o$   
*Spivak, N. Z. and Andreev, A. V., Phys. Rev. B 93, 085107 (2016)*

- If there are other inelastic relaxation mechanisms,  $L = \tau_h^{-1} / \tau_{eff}^{-1} L_o < L_o$

- If there is ambipolar conduction:  $\kappa_E = L_o T \sigma (1 + ZT); \quad L > L_o$

- Chiral zero sound:  $L \gg L_o$

# At zero field Wiedemann-Franz holds

Below 50 K

$$\kappa \sim \kappa_L$$

⇒ extreme error

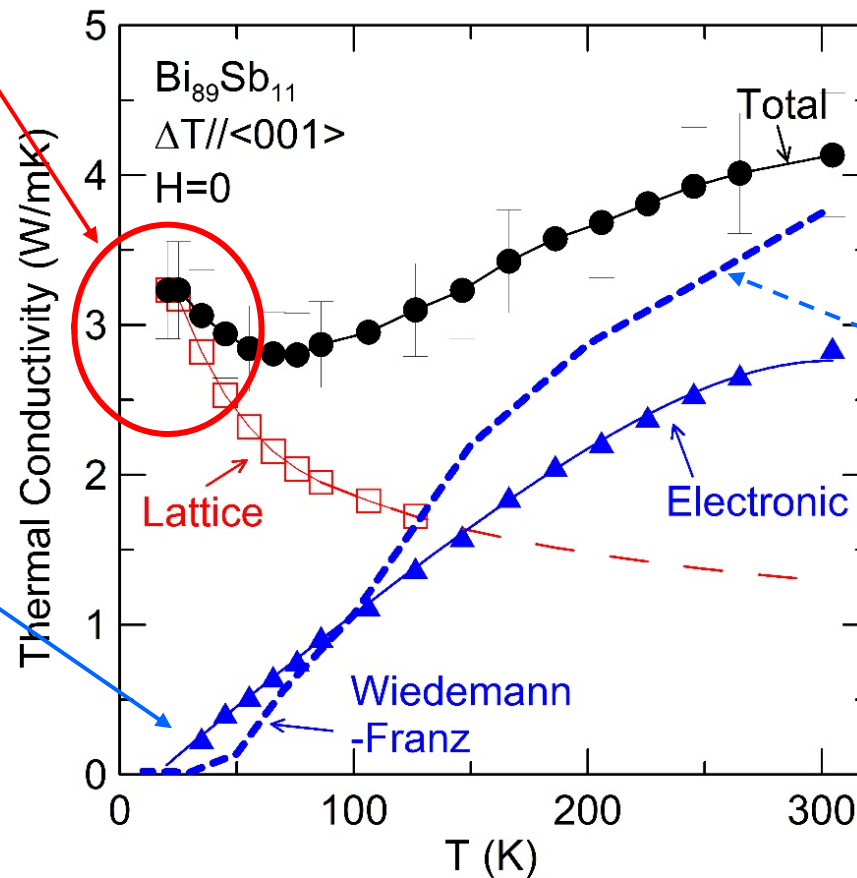
bars on  $\kappa_E$

⇒ Procedure impossible

⇒ Example: at zero field one would get unphysical result  $L \gg L_0$

$$\kappa = \kappa_E + \kappa_L$$

Measure      Isolate      Subtract



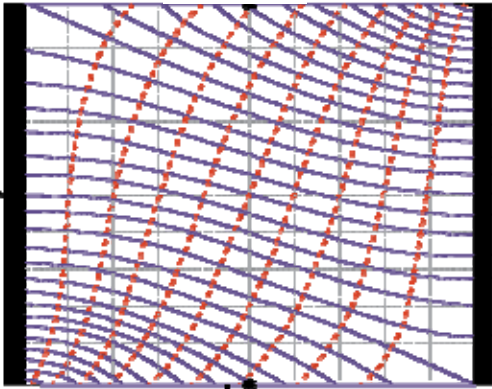
- Take experimental values of electrical resistivity
- Calculate

$$\kappa_{WF} = L_0 T \sigma$$

- Reproduces  $\kappa_E$
- Wiedemann-Franz holds

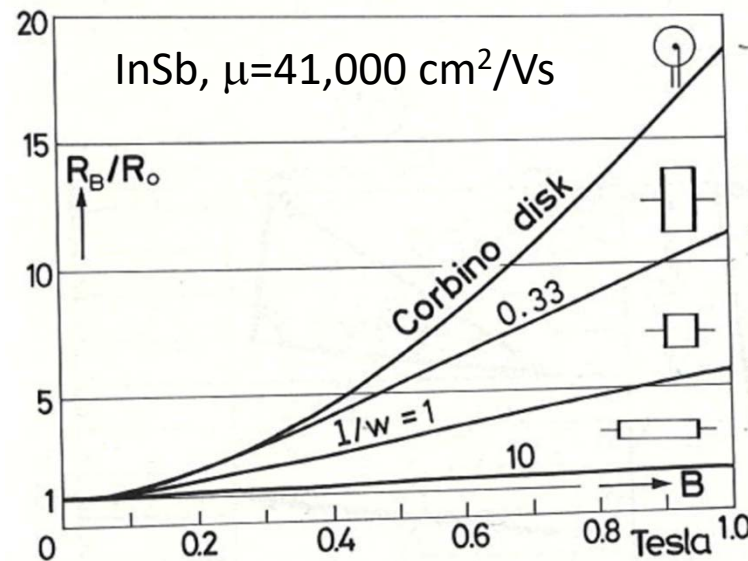
# Error due to geometrical magnetoresistance in $\sigma_{33}(B_3)$

Distortions of current lines by Lorentz force =>  
Geometrical magnetoresistance in transverse field



$$R(B) / R_0 = 1 + g \mu^2 B^2$$

↑  
Geometrical factor



Length / Width	$g$
0	1
0.33	0.6
1	0.28
10	0.05
$\infty$	0

In our samples,  $\mu \sim 0.7 \cdot 10^6 \text{ cm}^2/\text{Vs}$  @ 60 K =>  
1T transverse gives, for square sample  $R(B)/R(0)=1400$

*Baker, D. R. and Heremans, J. P., Phys. Rev. B 59, 13927 (1999), InSb data from Weiss, Zeitschrift für Physik, 13d. t38, S. 322--329 (1954).*

# Problem: geometrical magnetoresistance in $\sigma_{33}(B_3)$

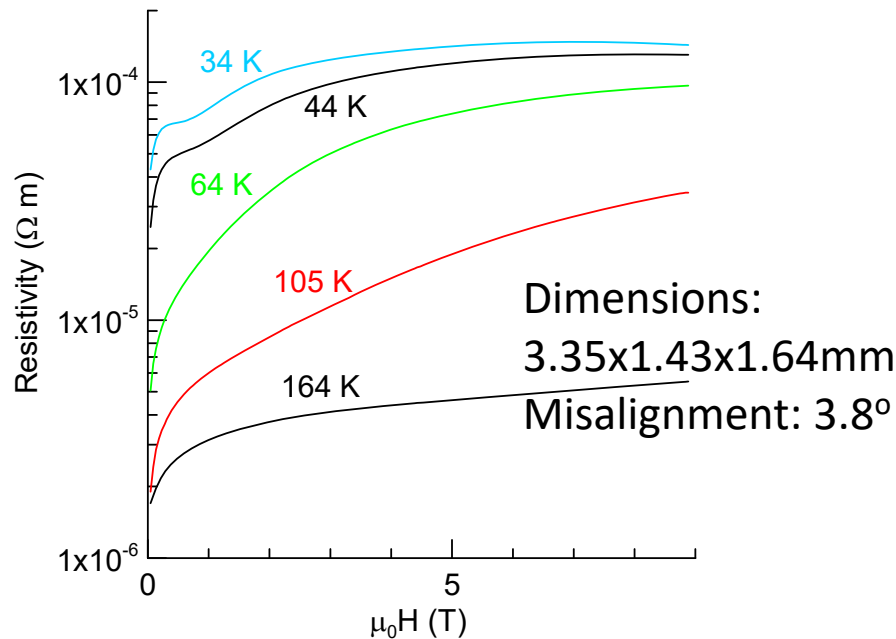
Source of error: misalignment of the longitudinal field, angle  $\theta$

$$\Delta R / R|_{ERROR} = g \mu^2 B^2 \sin^2 \theta$$

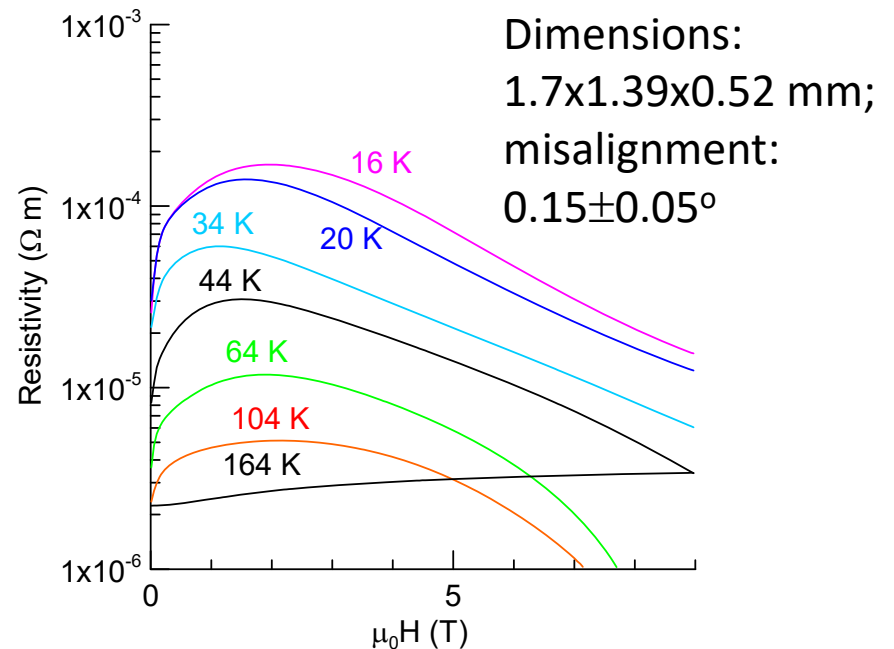
In our samples,  $\mu \sim 0.7 \cdot 10^6 \text{ cm}^2/\text{Vs}$  @60 K  
 $\Rightarrow$  1 degree misalignment 1 Tesla gives  
 200% relative error

In Bi,  $\mu \sim T^{-2}$  so error  $\sim T^{-4}$  5% error at 60 K gives 45% error at 35 K

Example: same  $\text{Bi}_{89}\text{Sb}_{11}$  sample as used for thermal conductivity



Geometrical MR error at 60 K & 9T: factor 400  
 Chiral anomaly masked



Geometrical MR error at 60 K & 9T: 25%  
 Chiral anomaly visible

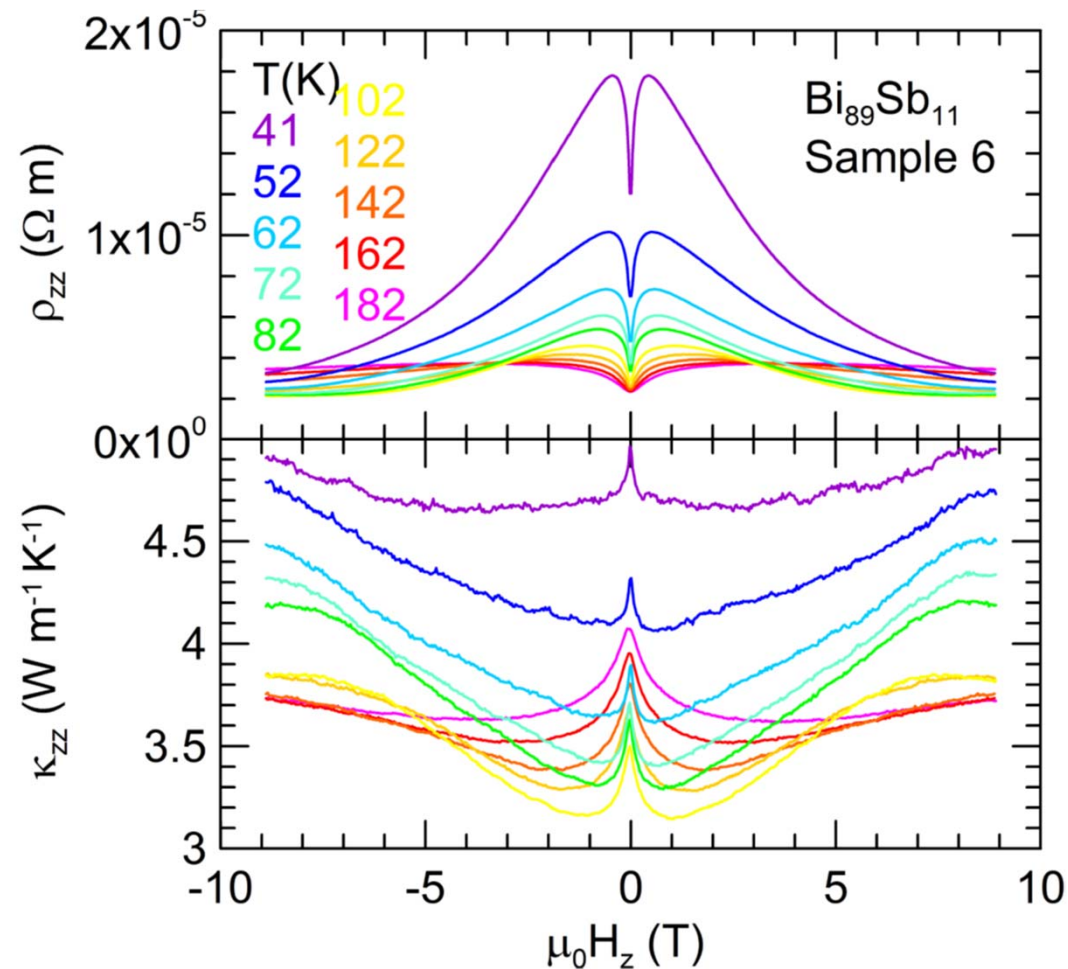
Wiedemann-Franz law (WFL)  $\kappa_E = LT\sigma$

Predictions for the effect of measurement errors:

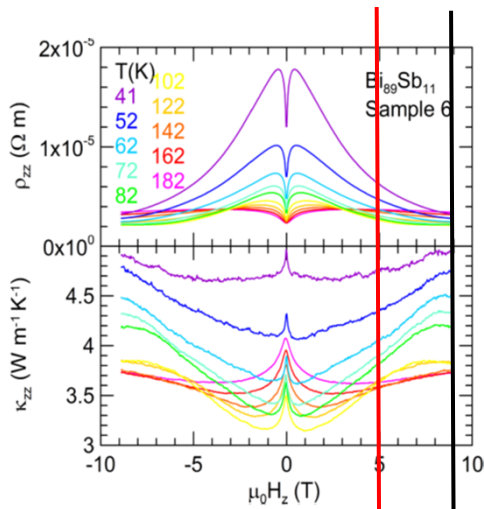
- If we underestimate the lattice contribution:  $L \gg L_o$
- If current jetting effect contaminates measurement:  $L < L_o$
- Small misalignment and geometrical MR contamination:  $L \gg L_o$

## *Bi<sub>89</sub>Sb<sub>11</sub> sample 6 specially cut & etched*

1. Cut to  $3 \times 0.6 \times 0.4$  mm  $L/W=7.5/1$  ( $g=0.06$ )
2. Sides etched (surface roughness also gives geometrical MR)
3. Mounted with misalignment angle  $< 0.1$  degree (goniometer + guides)
4. MR error at  $T=60$  K and  $B=9T$ :  $< 7\%$ .



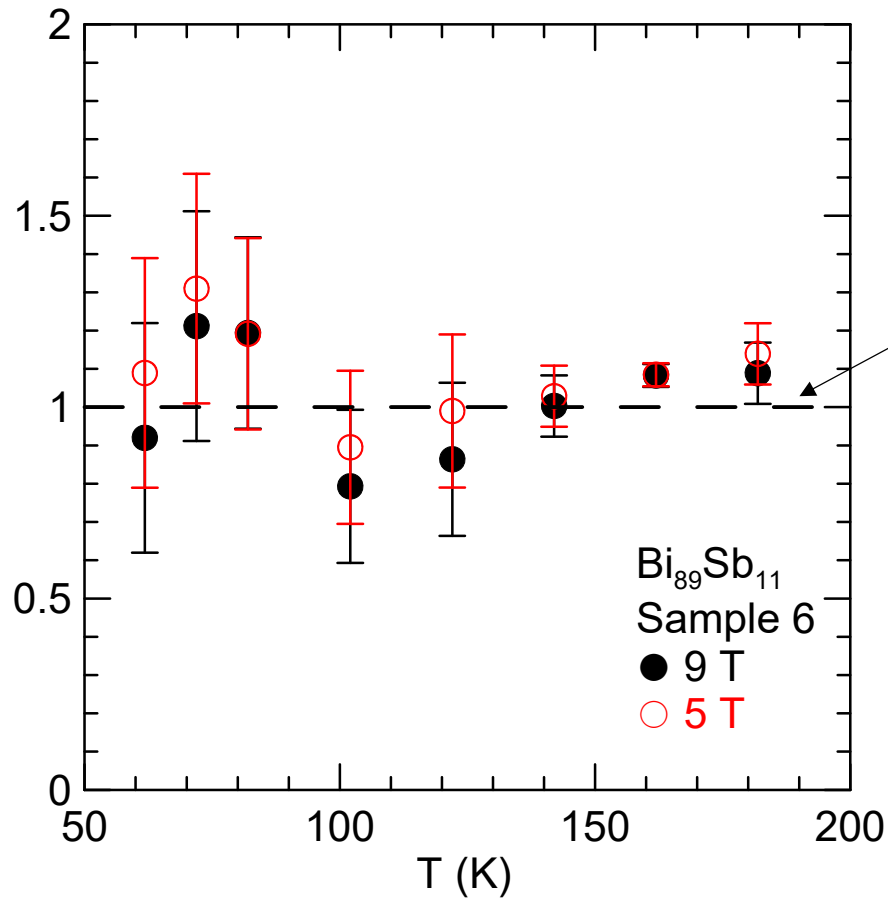
Wiedemann-Franz is obeyed at  $T > 50$  K in the chiral anomaly regime



- Take  $\kappa_{zz}$
- Subtract  $\kappa_{zz,L}$
- Take  $\rho_{zz}$
- Calculate

$$\frac{(\kappa_{zz} - \kappa_{zz,L}) \rho_{zz}}{L_0 T}$$

$\kappa_e \rho / L_0 T$



$$L = L_0 = \frac{\pi^2}{3} \left( \frac{k_B}{e} \right)^2$$

Below 50 K, error on MR and error on  $(\kappa_{zz} - \kappa_{zz,L})$  are prohibitive => no data



## Conclusions

1. “Chiral anomaly” in Weyl semimetals is a charge carrier density unbalance between Weyl points when  $\vec{E} // \vec{B}$
2. There exists an equivalent energy unbalance when  $\nabla T // \vec{B}$
3. Both effects are related by the Wiedemann-Franz law
4.  $\text{Bi}_{1-x}\text{Sb}_x$  alloys ( $x > 10\%$ ) are ideal Weyl semimetals in magnetic field
5. In them, the anomalies are:
  - 300 % at 9T
  - Robust against phonon scattering
  - Robust against defect scattering
  - Governed only by one energy scale: the width of the Weyl bands