Thermal Spin Transport: Spin-Seebeck and Magnon-drag Effects

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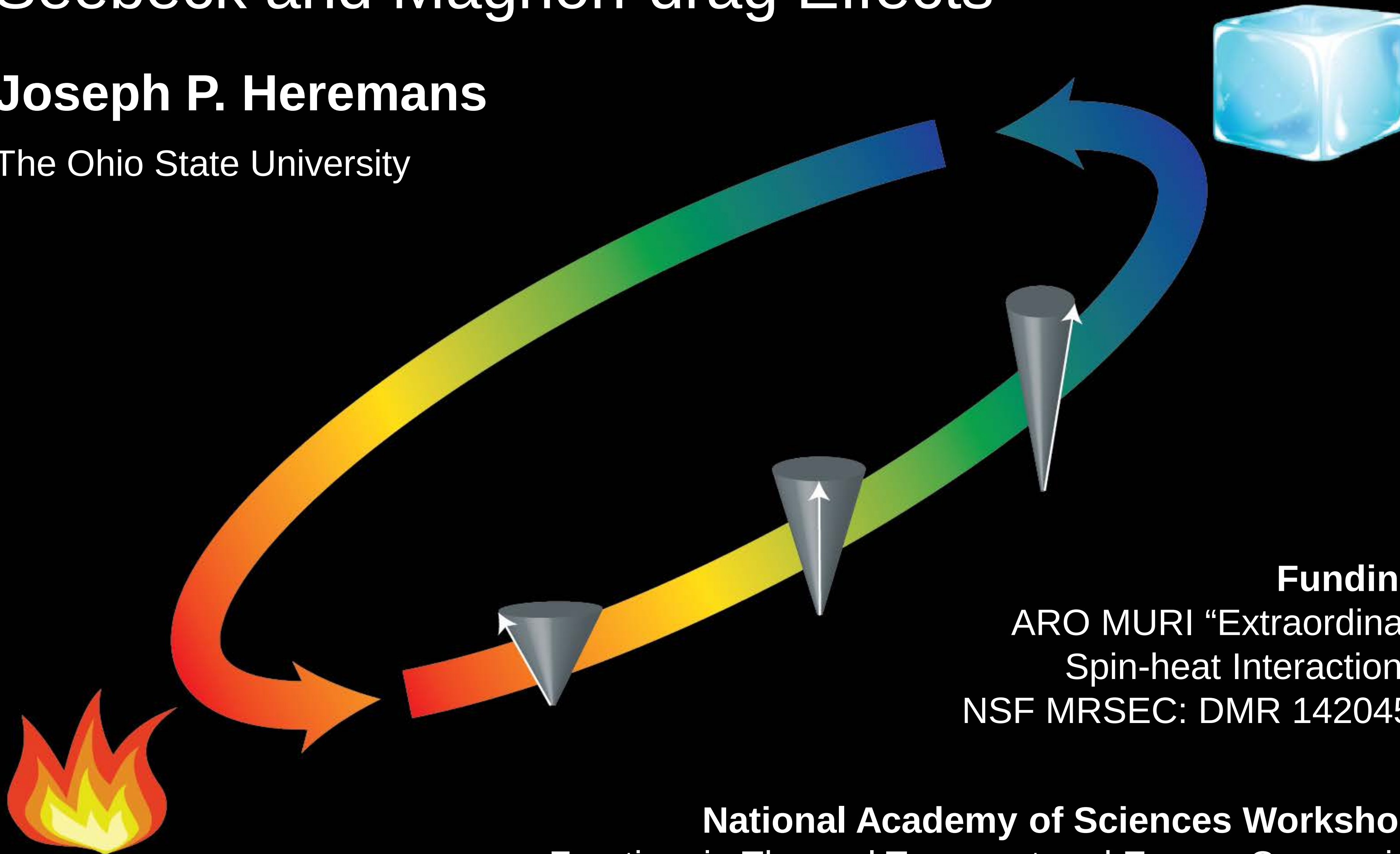

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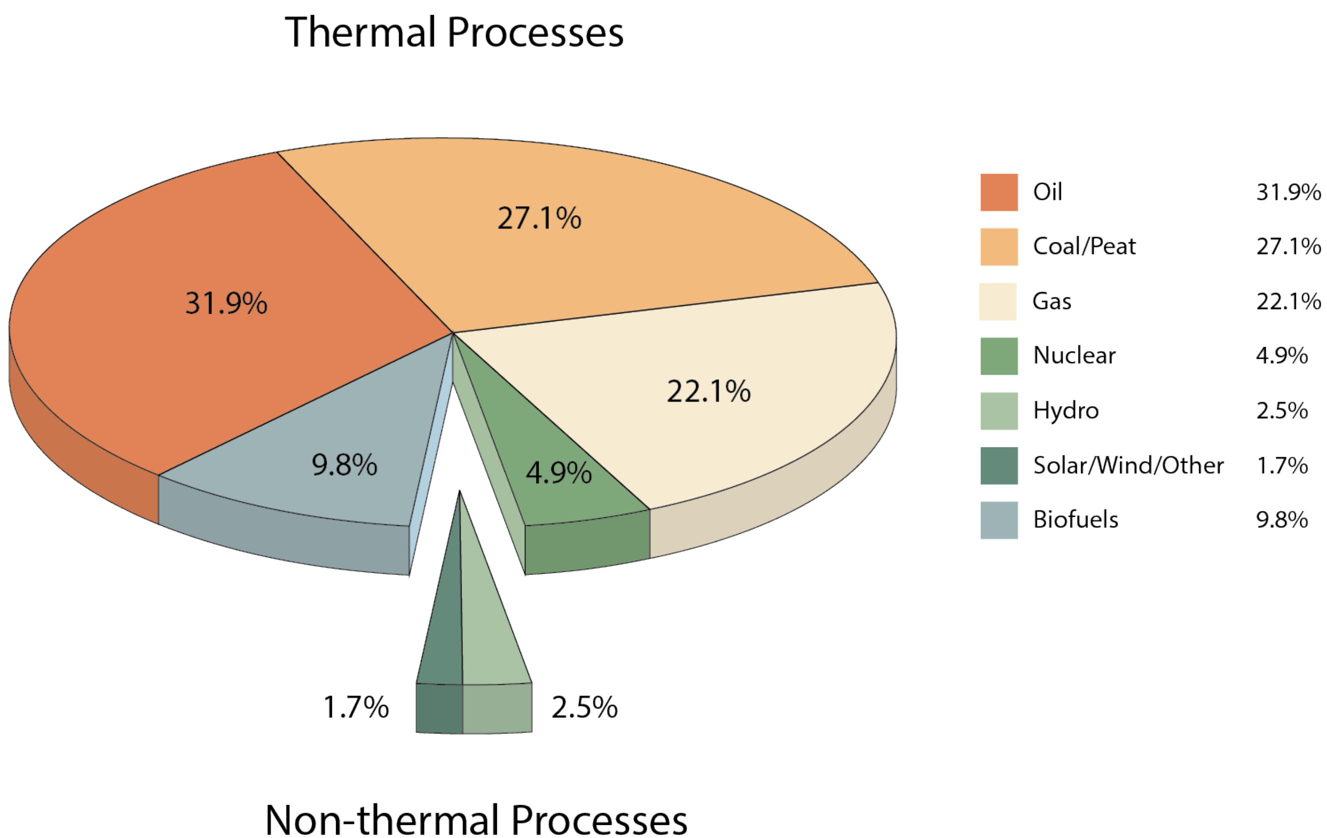
Motivation

- Over 95% of world energy comes from thermal processes
- Improvements in efficiency impact energy conservation and CO_2 emissions



International Energy Agency (2016)
https://www.iea.org/statistics/?country=WORLD&year=2016&category=Energy%20supply&indicator=TPESbySource&mode=chart&dataTable=BALANCES

Solid State Heat Engines

- Advantages:
  - No moving parts
  - Robust, infinite lifetime, no maintenance
- Technologies
  - Magnetocaloric engines, adiabatic demagnetization:
    - Require heat switches
    - Require a magnetic field
  - Electrocaloric engines:
    - Require heat switches
    - Very low power
  - Thermoelectrics:
    - Very high power density (specific power) → Light weight
    - Easy to control
    - Suitable to power levels up to 1 kW

Heremans & al., Nature Nano. ***8****, 471-(2013)*

Thermoelectrics: Fundamentally new approaches are necessary

- Thermoelectrics have low efficiency, low ZT
- Since ~2013, thermoelectric research has stalled at ZT ~ 2.2 $\pm$ 0.4
- Projected limit of ZT ~ 4 (*G. A. Slack, CRC Handbook on Thermoelectricity, 1997*)
- **Need approaches based on 21st century physics**

Increase the thermopower of solids with high electron density.
Break Mott relation based on Fermi statistics

Lower lattice thermal conductivity
Amorphous limit is ~ reached

$$ZT = \frac{S^2\sigma}{\kappa_E + \kappa_L} T = \frac{S^2}{L} \frac{1}{1 + \kappa_L / \kappa_E}$$

$$\kappa_E = LT\sigma$$

Lower Lorenz ratio
Break quasi-particle picture

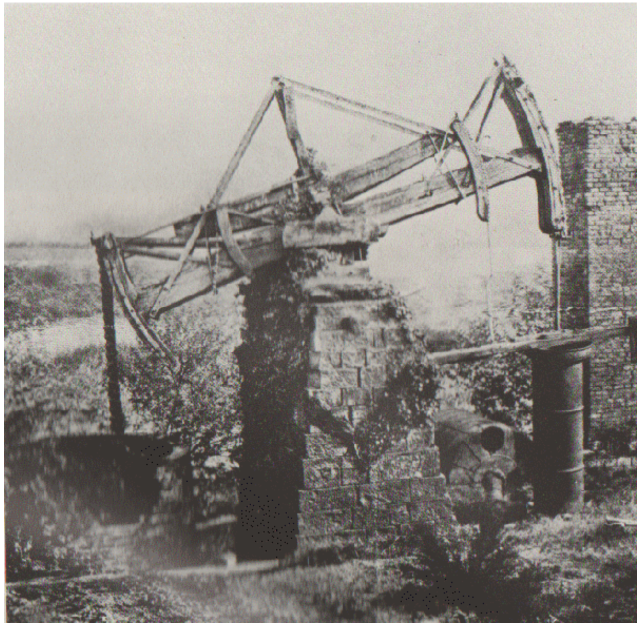
Structure of This Lecture

1. **Spin-based Energy Conversion**
    1. Introduction: Solid-state Thermal Energy Conversion
    2. The Spin-Seebeck Effect
    3. Magnon-drag and Paramagnon-drag Thermopower
2. **Phonons and Diamagnetism**
3. **Outlook**

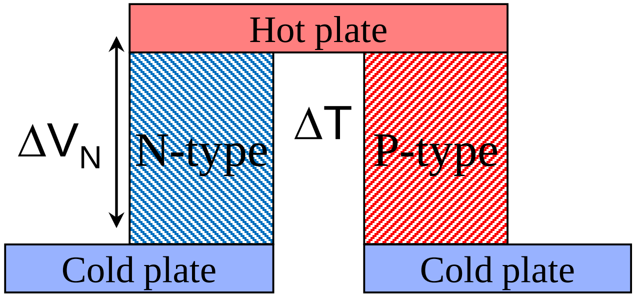
Heat Engines as Rectifiers

- Heat is the random motion of (quasi-) particles:
  - Electrons, magnons, atoms (phonons), molecules
- Work is unidirectional motion → A heat engine is some form of rectifier

Hero of Alexandria
10-70 AD



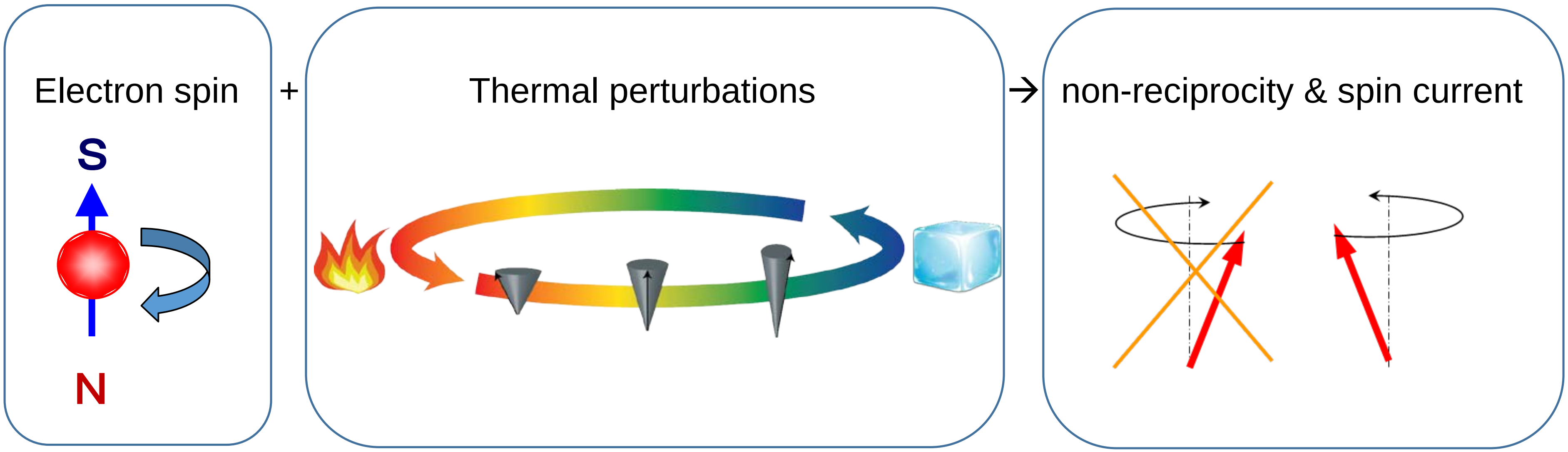
Newcomen engine
1720



Thermoelectric generator
1960's

1.1 Introduction: Solid-state Thermal Energy Conversion

- Precession waves in the magnetization (magnons): precession is counter-clockwise



- Thermally driven spin effects that produce work:
    - Spin-Seebeck Effect
    - Magnon drag
    - Mixed spin-Seebeck/Anomalous Nernst Effects
    - Paramagnon drag

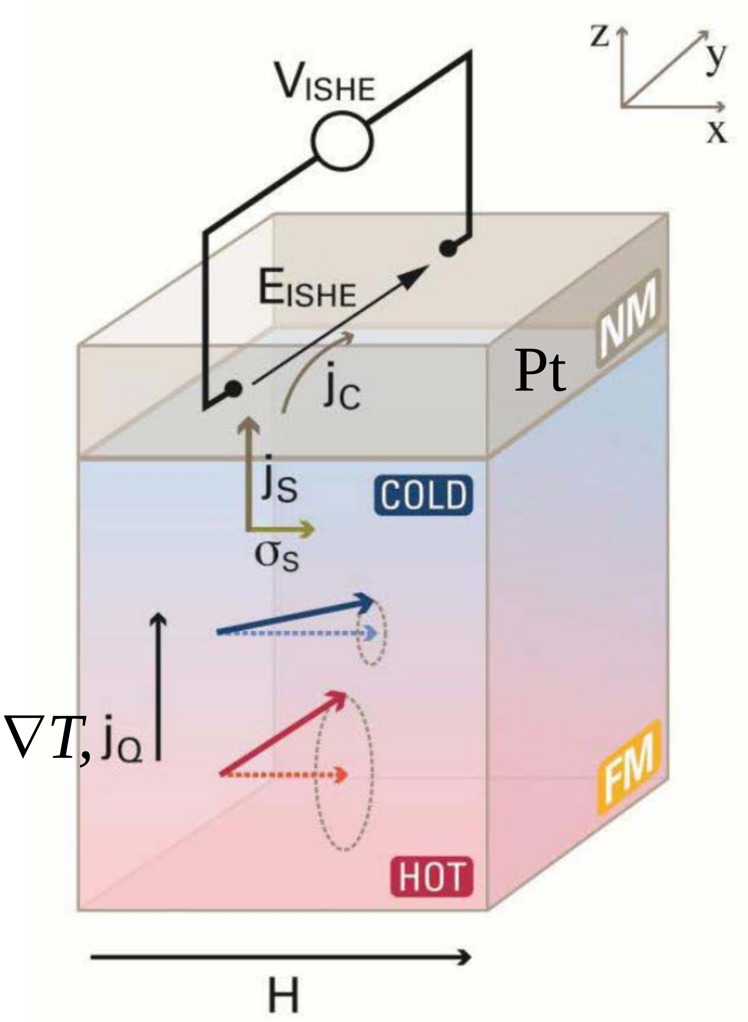
1.2 The Spin-Seebeck Effect (SSE)

- ∇T drives the magnon spin current
- Transfer of spin angular momentum across interfaces spin-polarizes Pt electrons
- Spin-orbit interactions in PT convert

$$J_S => E_{ISHE}$$

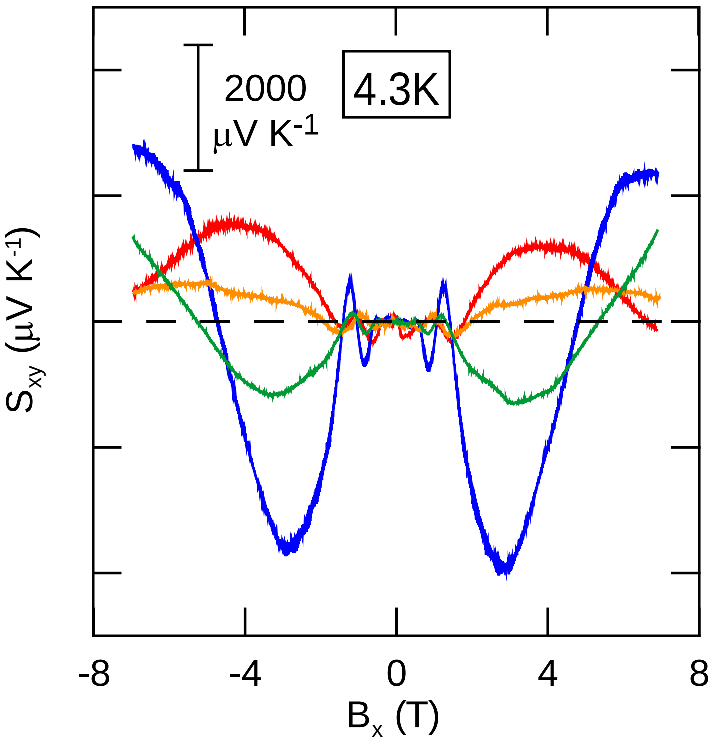
- Overall spin-Seebeck coefficient:

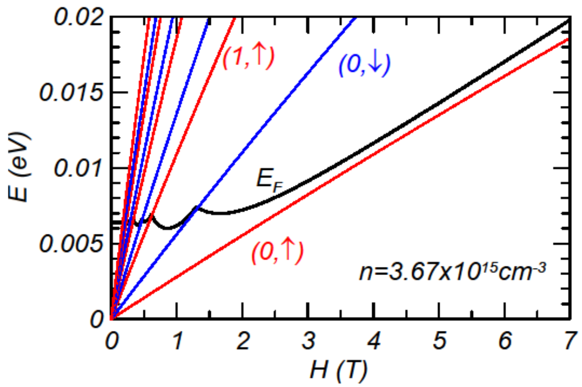
$$\boxed{S_{xy} \equiv \frac{E_{ISHE,y}}{\nabla_x T}}$$



The Spin-Seebeck Effect (SSE)

- Spin-Seebeck Effect has been observed in:
  - Ferromagnetic metals and semiconductors
    - Permalloy: *Uchida & al. (Saitoh group, Tohoku U.), Nature **455** 778 (2008)*
    - GaMnAs: *Jaworski & al. (OSU), Nature Materials **9** 898 (2010)*
    - YIG: *Uchida & al., Nature Materials **9** 894 (2010)*
  - Antiferromagnets
    - MnF_2: *Wu & al. (A. Hoffman, ANL), Phys. Rev. Lett. **116**, 097204 (2016)*
  - Paramagnets
    - Paramagnetic garnets: *Wu & al., Phys. Rev. Lett. **114**, 186602 (2015)*
  - Semiconductors with electrons on Zeeman-split Landau levels InSb (diamagnet): *Jaworski et al. (OSU), Nature **487** 213 (12)*





Thermopower of Fermions versus Bosons

- Thermopower is (single-particle picture) the entropy per particle *H. Callen, Thermodynamics, Wiley 1960*

- Thermopower of electrons is small at high concentration because electrons are fermions:

$$\alpha_{diff} = \frac{(\pi k_B)^2 T}{3eE_F}$$

- Thermopower of bosons does not have this limitation:

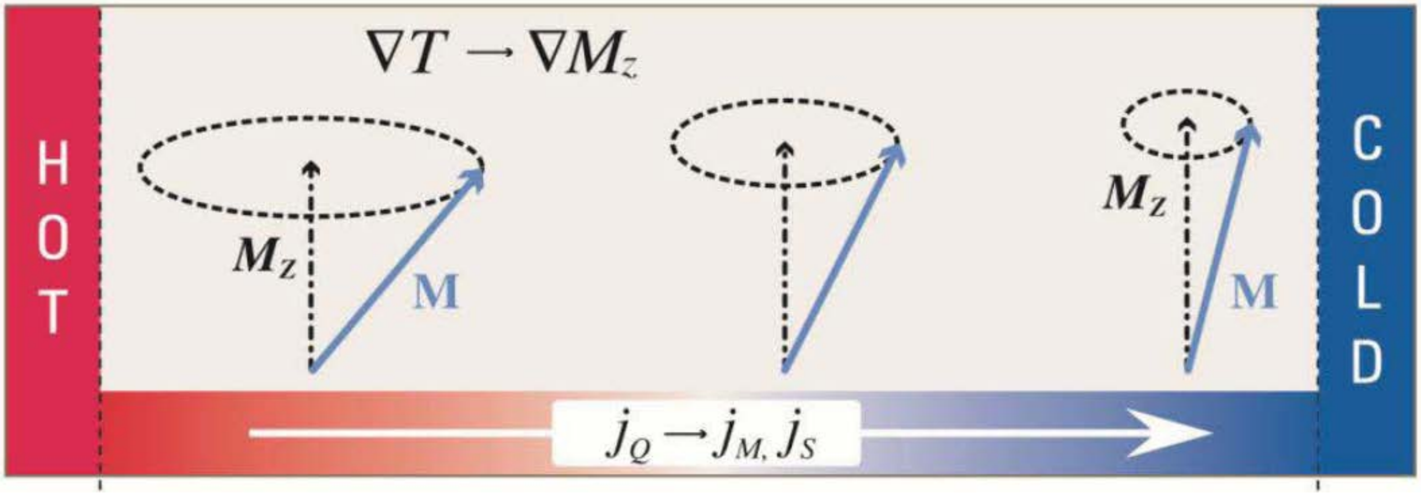
Magnon chemical potential, measures out-of-equilibrium magnon accumulation

$$\alpha_{magnonic} \equiv \frac{\nabla \mu}{\nabla T} \approx \frac{C_m}{n_m}$$

Specific heat per particle, not limited by the density of particles

Vandaele, …JPH, Materials Today Physics (2017).

Magnon Heat Current → Spin Current



Heat current of spin-wave implies a spin current → A temperature gradient implies a magnetization gradient.

$$j_Q = \kappa_{MAGNON} \nabla T$$

$$j_Q = k_B T . j_{MAGNON}$$

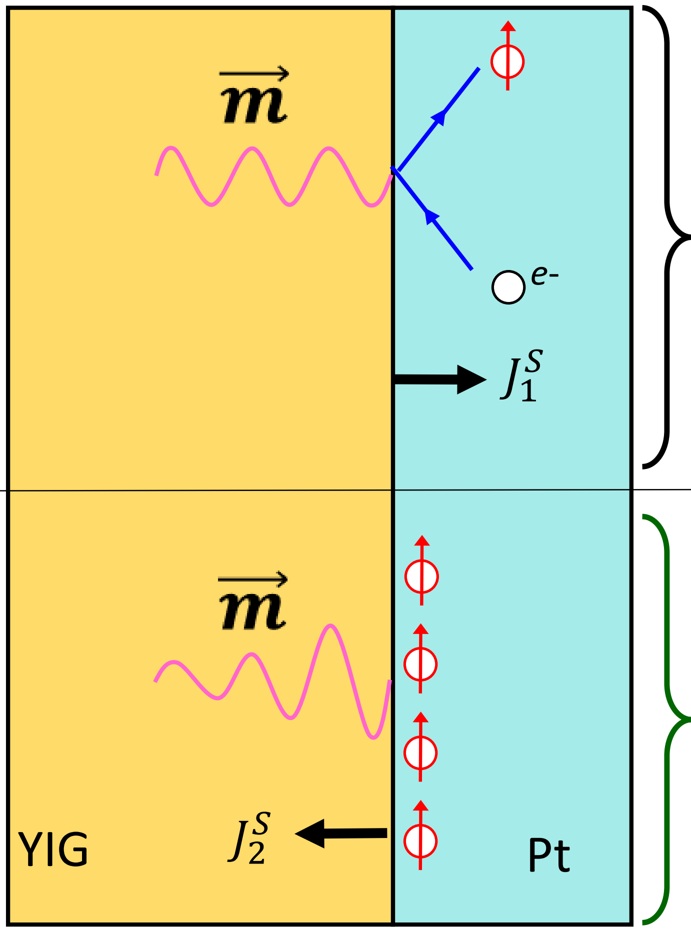
$$j_S \propto \hbar . j_{MAGNON} \propto \hbar / k_B T \, \kappa_{MAGNON} \nabla T$$

$$j_M \propto g \mu_B . j_{MAGNON} \propto g \mu_B / k_B T \, \kappa_{MAGNON} \nabla T$$

Magnon thermal conductivity measurements in YIG:

Boona & Heremans, Phys. Rev. B ***90****, 064421 (2014)*

Onsager Reversibility: Spin-Peltier Effect



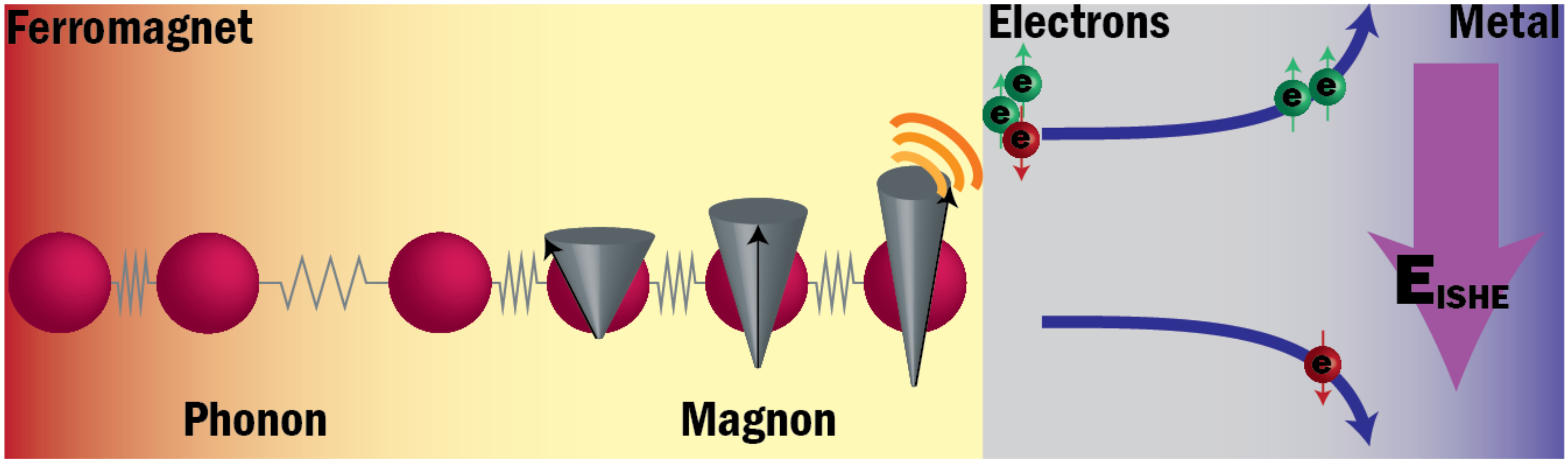
- Spin flux in ferromagnet
  - carried by magnons or spin-polarized electrons
  - pushed by heat current
- Spin transfer FM→Pt at the interface: J_1^S
  - exchange interaction between free electrons in the metal ("s") and "d"-electrons on Fe atoms in FM
  - Exponential decay of spin-polarization in Pt

- Reversible effect follows Onsager reciprocity
  - Spin accumulation in Pt (e.g., by spin-Hall Effect)
  - Spin transfer Pt→FM, generating magnons or spin-polarization of electrons in FM
  - Exponential decay of spin current J_2^S in FM

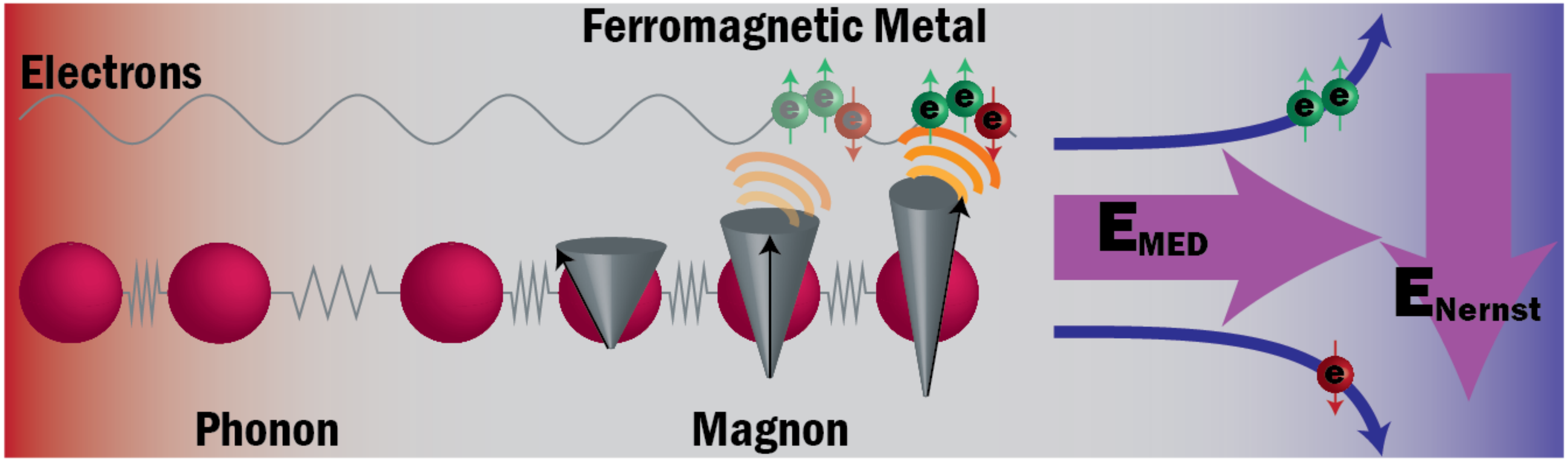
Flipse... B. van Wees, Phys. Rev. Lett. 113, 027601 (2014)

1.3 Advective Transport: Spin-Seebeck Effect and Magnon Drag

Spin Seebeck Effect



Magnon-Drag Effect



Boona...JPH, APL Materials ***4****, 104502 (2016)*

Magnon Drag (MD)

- DRAG is advective transport inside a bulk sample
  - Thermal force is exerted on magnons via phonon-magnon coupling
  - Magnon flux drags electron flux via s-d interactions (very strong)
  - 1 to 2 orders of magnitude stronger than diffusion thermopower
  - Rare example of a spin-mediated effect larger than a charge-mediated effect

- History:
  - The thermopower of Fe is due to MD: *Blatt & al., Phys. Rev. Lett.* ***18****, 395 (1967)*
  - Relation MD/SSE: *Lucassen & al., Appl. Phys. Lett.* ***99****, 262506 (2011)*
  - Hydrodynamic theory and fit to Fe, Co, & Ni: *Watzman, ... JPH, Phys. Rev. B* ***94****, 144407 (2016)*
  - Landau-Lifshitz theory: *Flebus et al., Euro Phys. Lett.* ***115****, 57004 (2016).*

Hydrodynamic Model

- **Hydrodynamic theory of magnon-drag thermopower**

magnon specific heat $C_M \propto T^{3/2}$ for FM

$\propto T^3$ for AFM

$$\alpha_{MD} = \pm \frac{2C_M}{3n_e e} \cdot \frac{1}{1 + \tau_m^{-1} / \tau_{em}^{-1}}$$

Concentration of free electrons

τ_m	Total momentum relaxation time for magnons.
τ_{em}	Magnon momentum relaxation time limited by magnon-electron interactions.
τ_L	Magnon lifetime in the AFM ($\tau_{L,AFM}$) and PM ($\tau_{L,PM}$) regimes
τ_e	Total momentum relaxation time for electrons.
τ_{me}	Electron momentum relaxation time limited by electron-magnon interactions

*S. J. Watzman,& al. (OSU) Phys. Rev. B **94** 144407 (2016)*

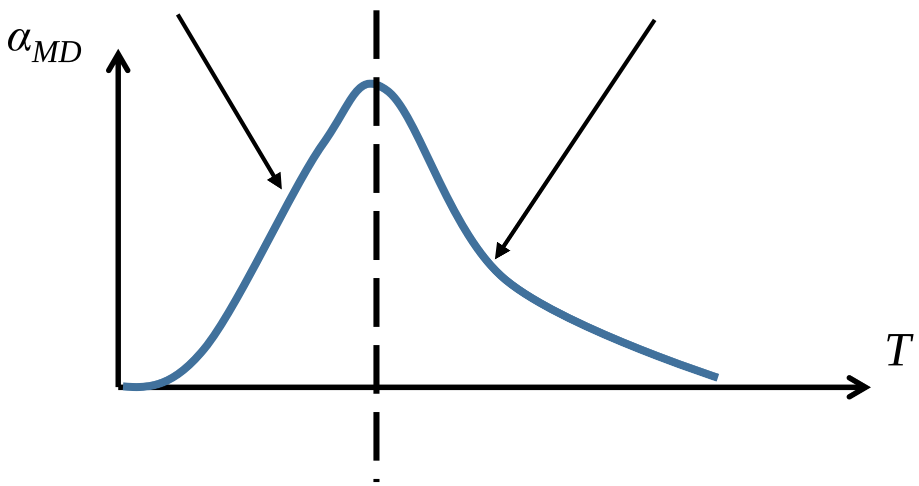
Hydrodynamic Model

$C_M \propto T^{3/2}$ for ferromagnet

$\propto T^3$ for antiferrmomagnet

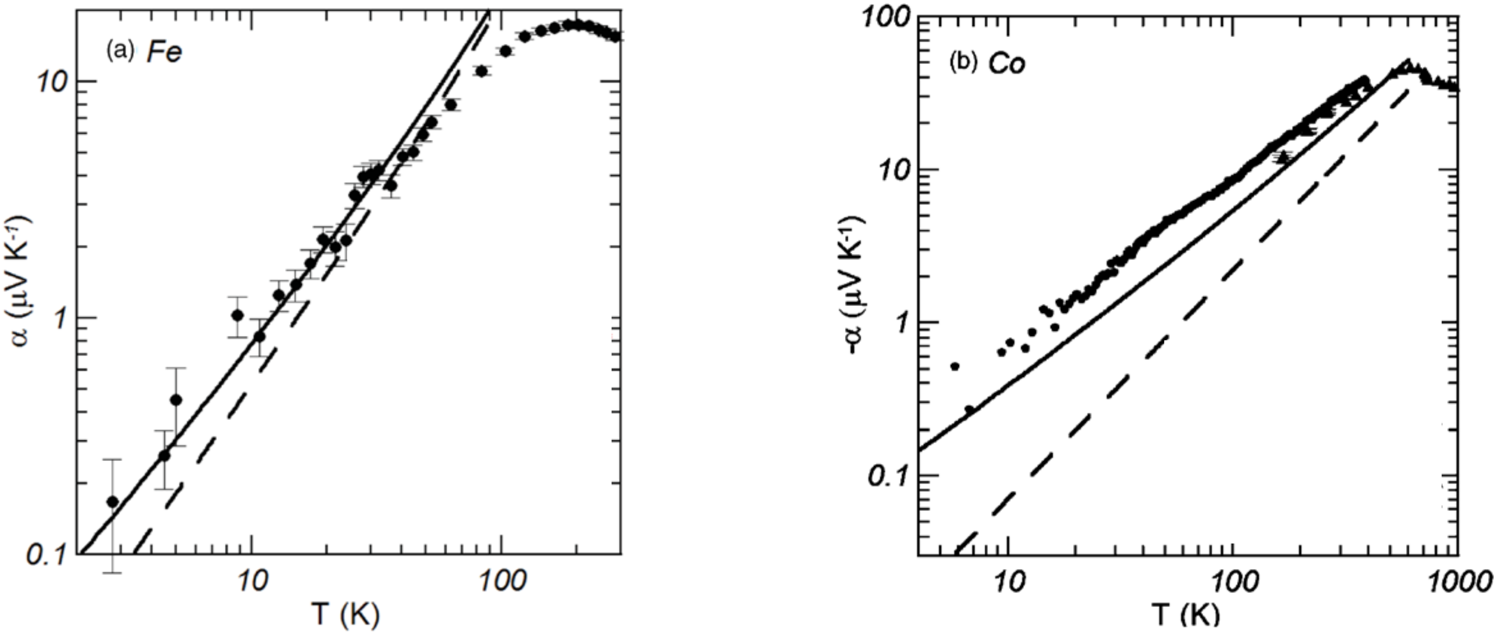
$$\alpha_{MD} = \pm \frac{2C_M}{3n_e e} \cdot \frac{1}{1+\tau_m^{-1}/\tau_{em}^{-1}}$$

$T^{3/2 or 3}$ at low temperature



Hydrodynamic Model

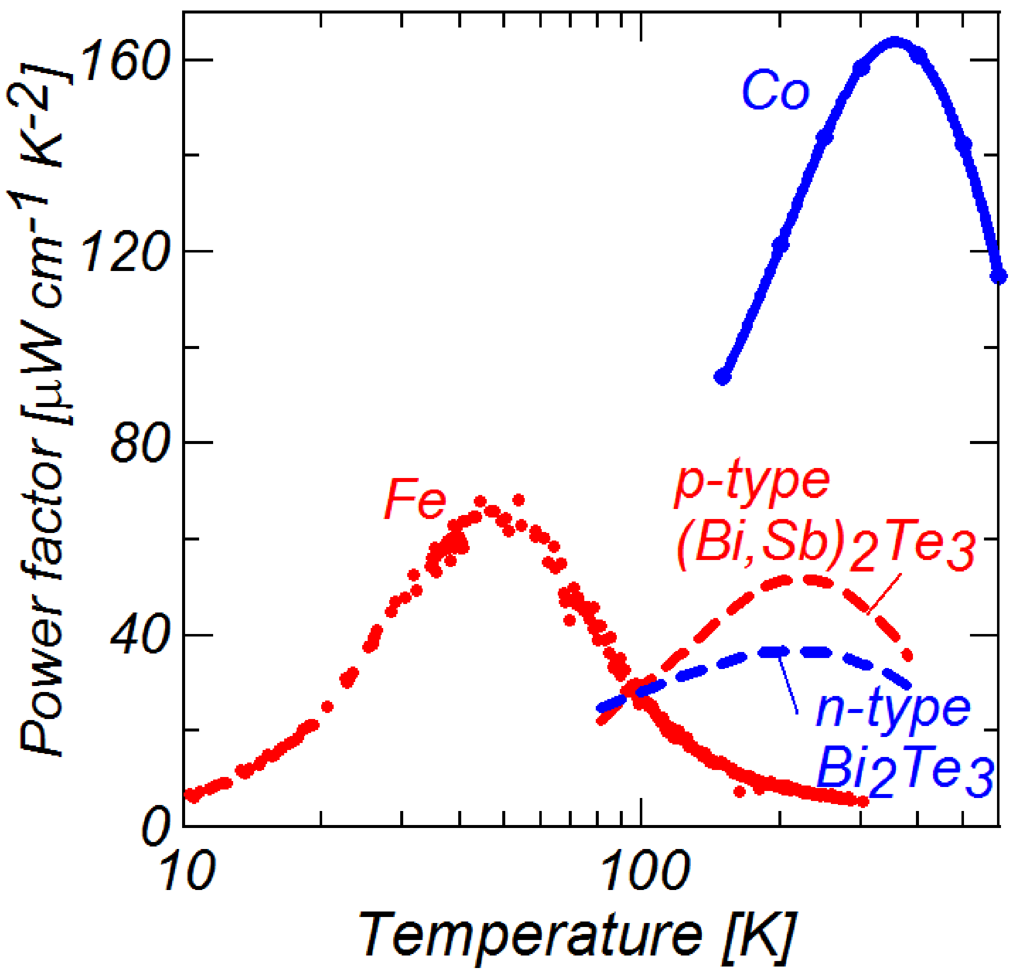
- Elemental Fe and Co



- Magnon dispersions and C_M are known
- Charge carrier concentrations are known
- S-d scattering dominates → assume all magnon scattering is with electrons → No adjustable parameters
- $\alpha_{MD}$is 10 times larger than α_{diff}!

S. J. Watzman,& al. (OSU) Phys. Rev. B ***94*** *144407 (2016)*

Thermoelectric Power Factors from Magnon Drag



- But these are metals. Can this be applied to thermoelectric semiconductors?
- No known ferromagnetic semiconductors at room temperature

MnTe

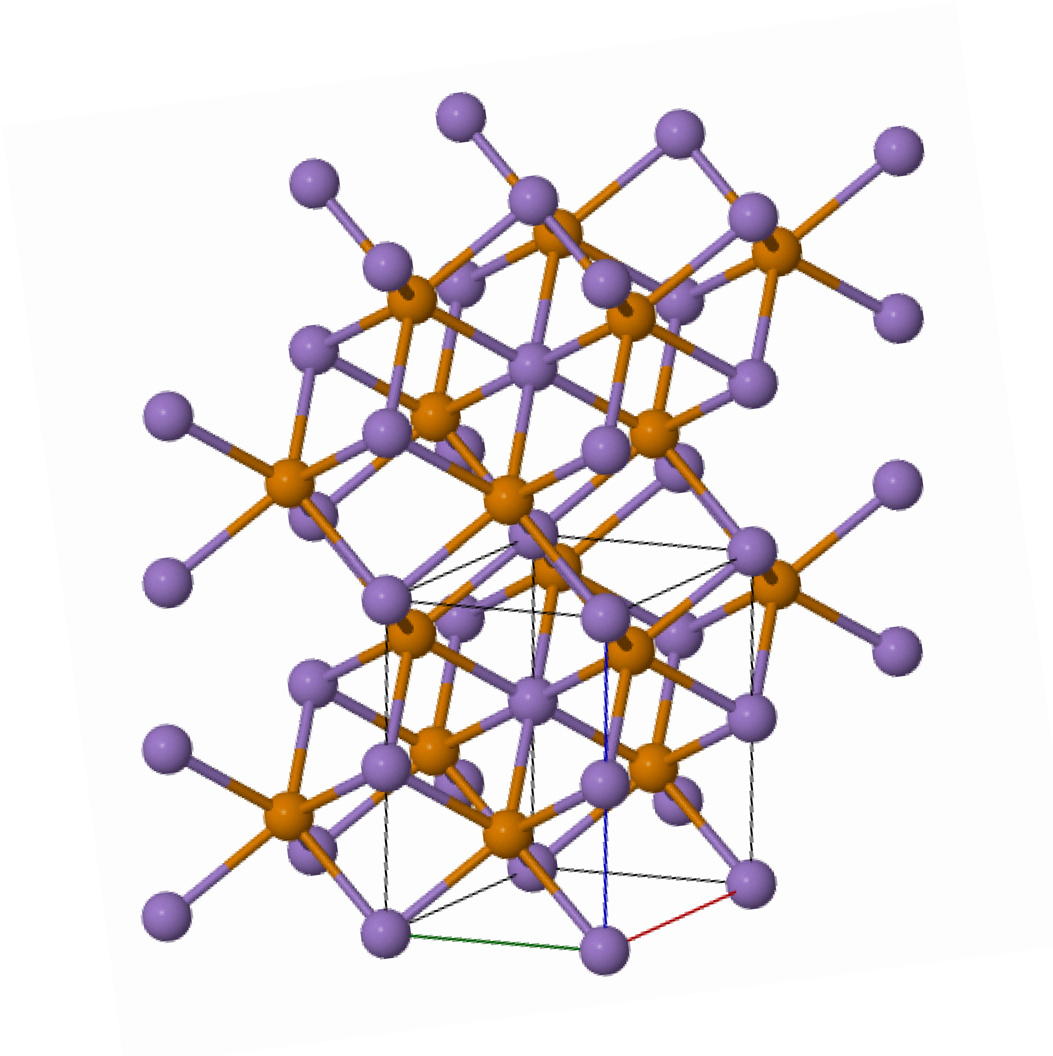
- Antiferromagnetic semiconductor
- Hexagonal NiAs structure
- Neel temperature TN ~ 307 K
- Curie-Weiss temperature ϑ = -585 for single-crystal MnTe

- Lithium = aliovalent acceptor

- This study: $Li_xMn_{1-x}Te$

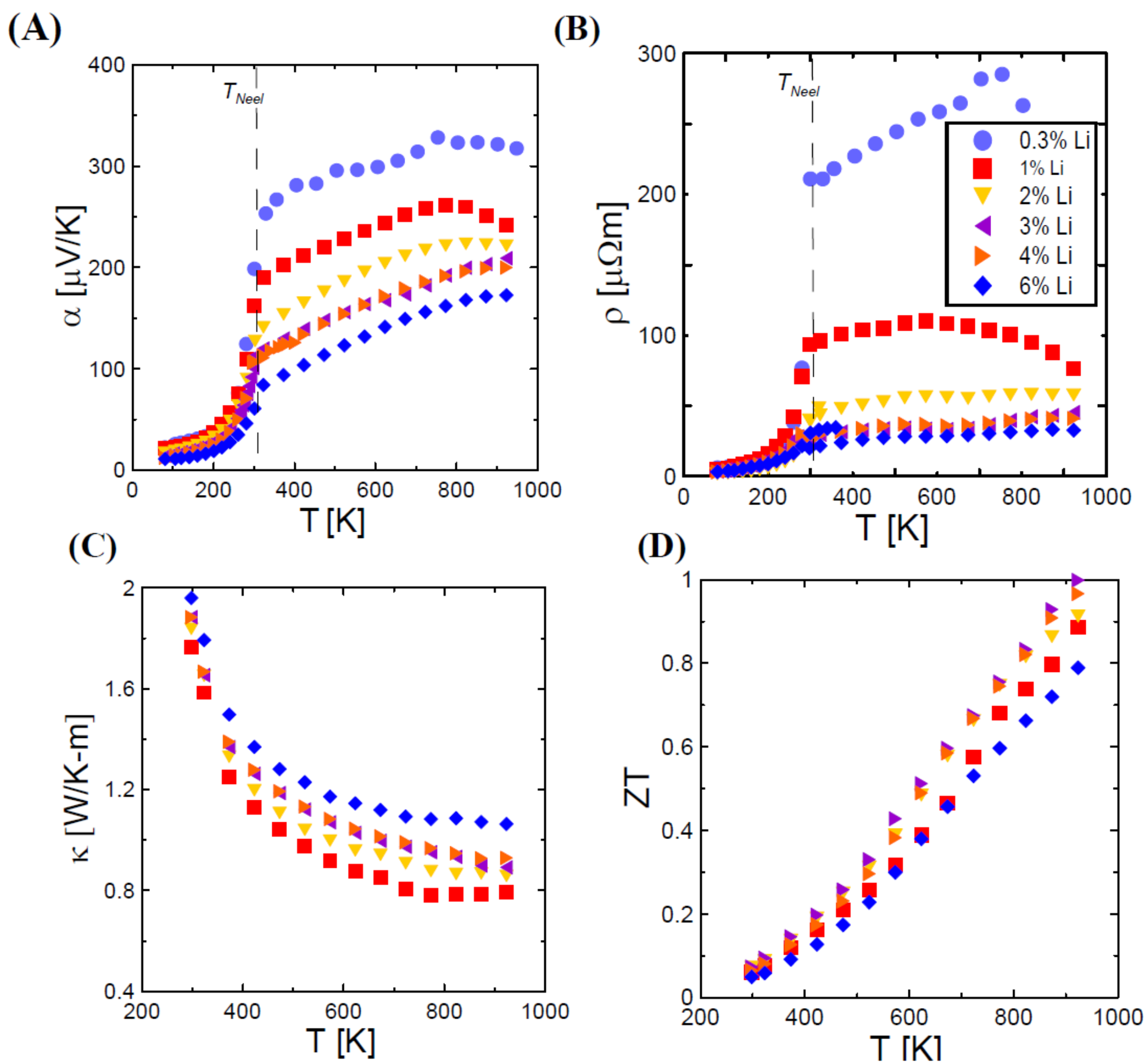
x (at%) = 0.3, 1,2,3,4,6%

Hole concentration 5×10^{19} to 10^{21} cm^{-3}



*J. D. Wasscher and C. Haas. Physics Letters **8** 302-304 (1964)*
P. Villars, PAULING FILE in: Inorganic Solid Phases, SpringerMaterials.

Thermopower, Resistivity, Thermal Conductivity, and ZT >1 in Simple Binary Semiconductor



Y. Zheng, et al. collaboration OSU/NCS/ONRL/Chinese Acad. Sci, Science Adv. (under review)

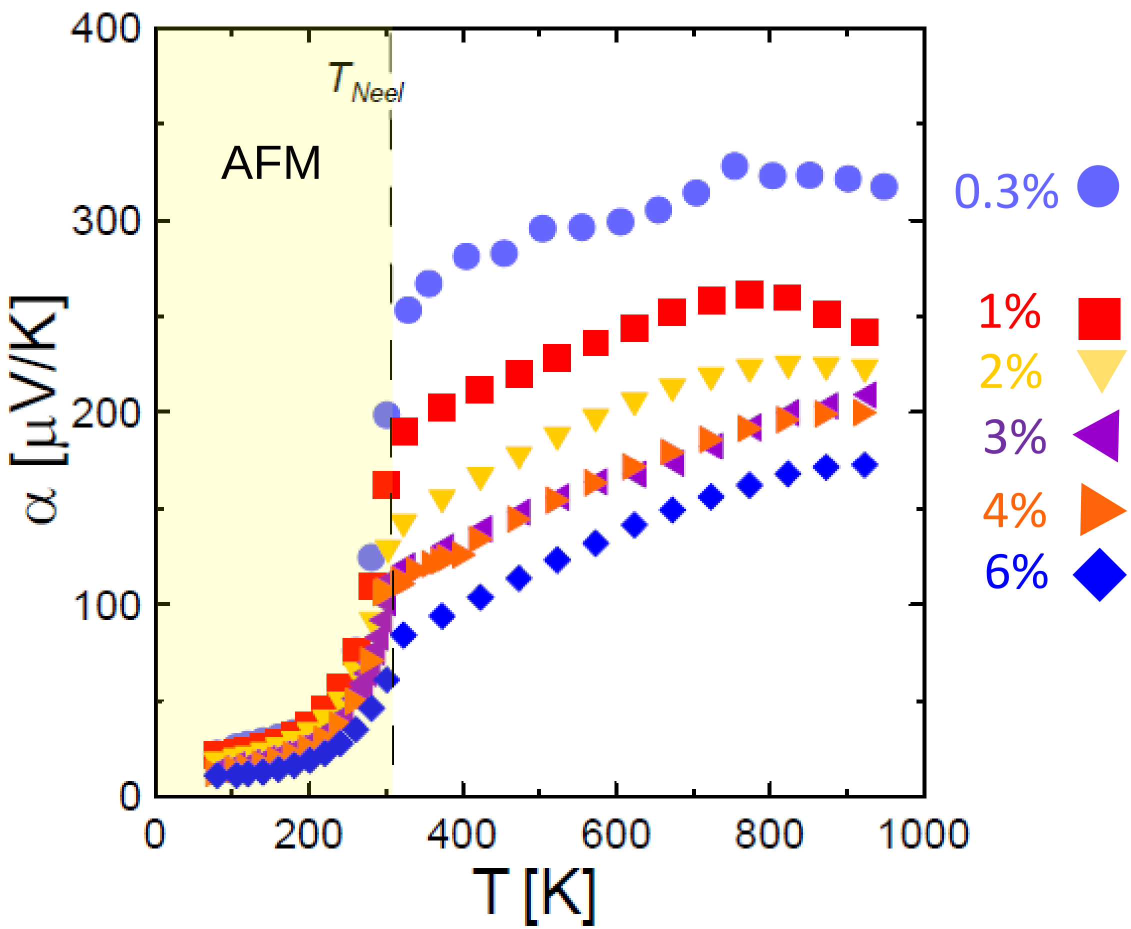
Thermopower

$T < T_{Néel}$: Magnon-drag

$$\alpha_{MD} = \pm \frac{2C_M}{3n_e e} \cdot \frac{1}{1+\tau_m^{-1}/\tau_{em}^{-1}}$$

$$C_{magnon} \propto T^3$$

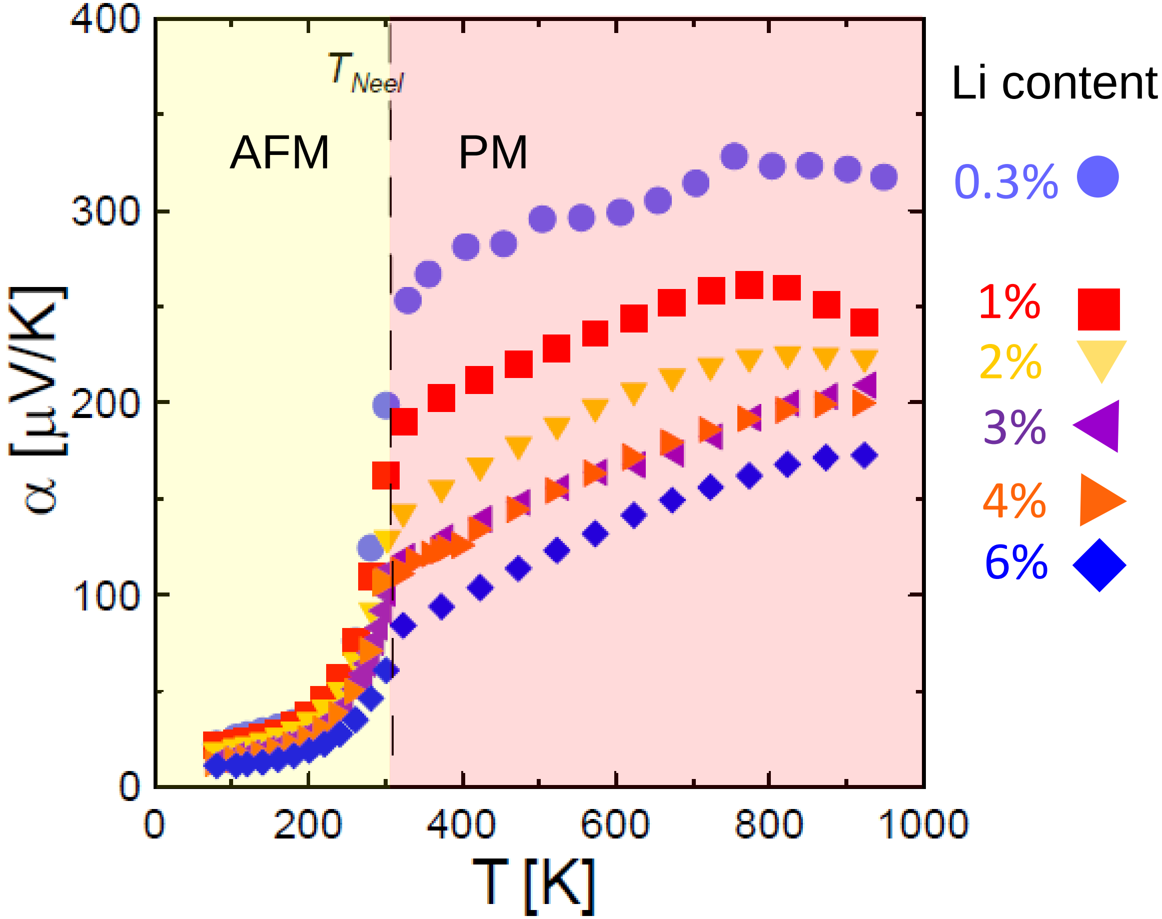
- Quantitative calculation (upon request) reproduces the data well



Thermopower

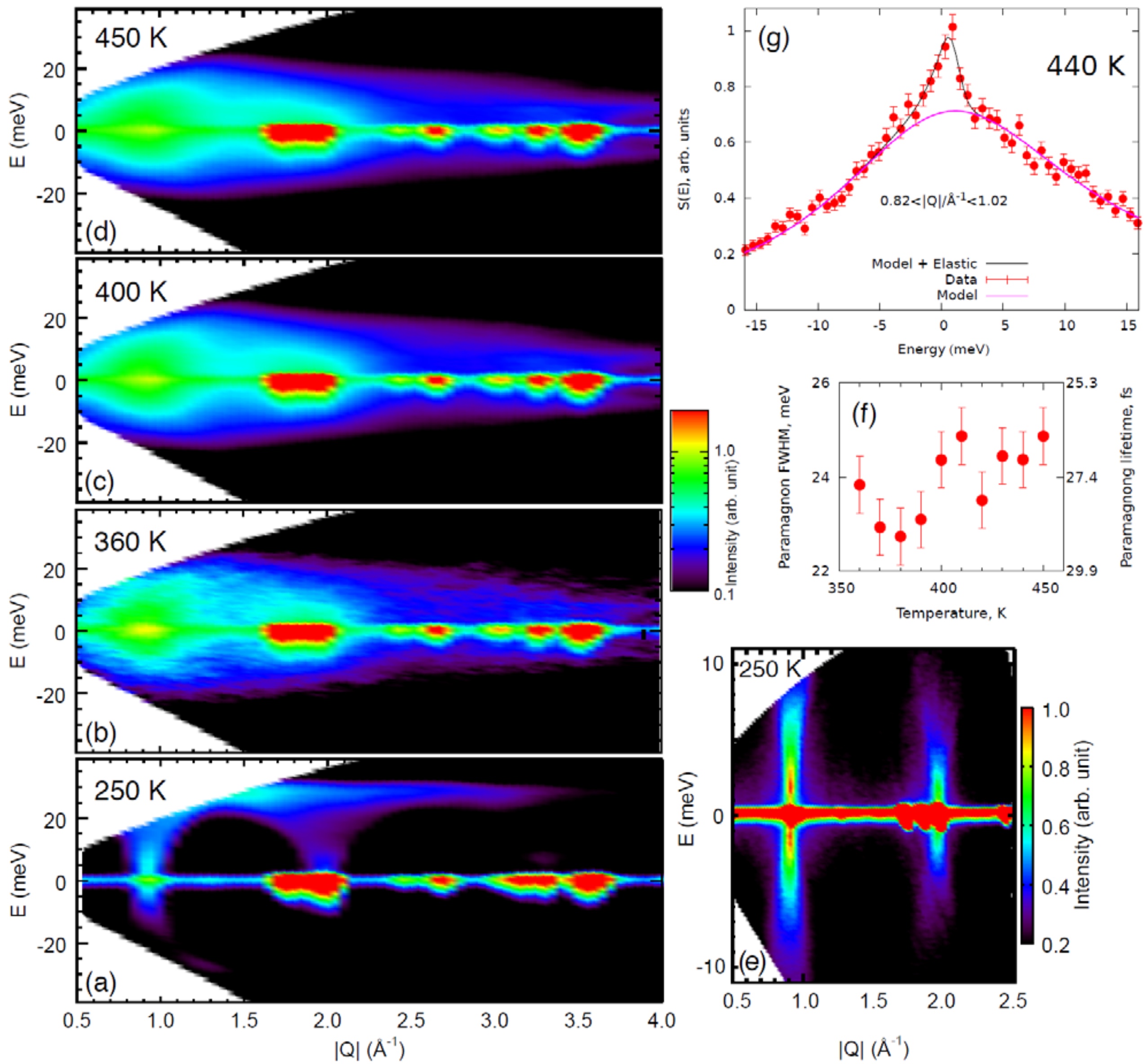
$T > T_{Néel}$: Hypothesis: paramagnon-drag

- Vision:
  - Local thermal fluctuations of the magnetization
  - Have spin-spin correlations
  - Have a finite lifetime
  - Drag electrons



Y. Zheng, et al. Science Adv. (under review)

Ineleastic Neutron Scattering



R. Hermann, ONRL, Science Adv. (under review)

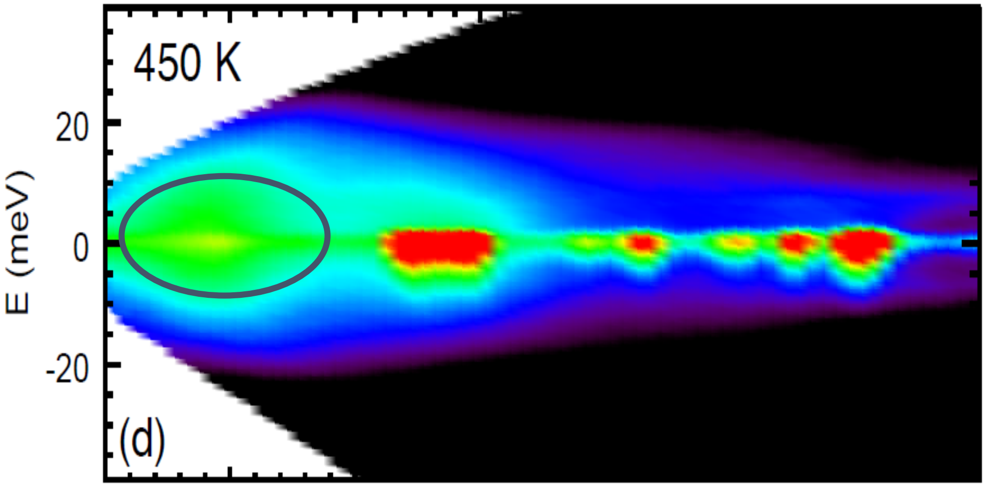
PM: Paramagnon Drag

- Paramagnons are short-lived, locally ordered spin fluctuations
- ΔE → Paramagnon lifetime

$\tau_{L,PM}$ = 27±1 fs

- ΔQ → Spin-spin correlation length

$\xi \approx$ 2.3±0.3 nm



$\tau_{L,PM}$ ≈ 27±1 fs >> $\tau_e \approx$ 3±2 fs (electron scattering)

$\xi \approx$ 2.3±0.3 nm >> $\lambda_{dB} \approx$ 0.6 nm & $a_B^* \approx$ 0.5 nm

electron de Broglie wavelength | effective Bohr radius

To electrons, paramagnons look like magnons

Opens the possibility of using dilute magnetic semiconductors as thermoelectrics

2. Phonons and Diamagnetism

- Lattice distortion due to phonons locally perturbs the valance band
- This creates a local change in diamagnetic susceptibility
- In the presence of a magnetic field → local variation of magnetization

→Magnetic force on atoms

→Perturbs interatomic force

→Change in Grüneisen parameter γ

$$\omega = \omega(x_0, k, m_{atoms})$$

$$k = k(x, x_0)$$

$$\gamma \equiv {d \ln(\omega)} \Big/ {d \ln(V)}$$

→**Magnetoresistance in the lattice thermal conductivity**

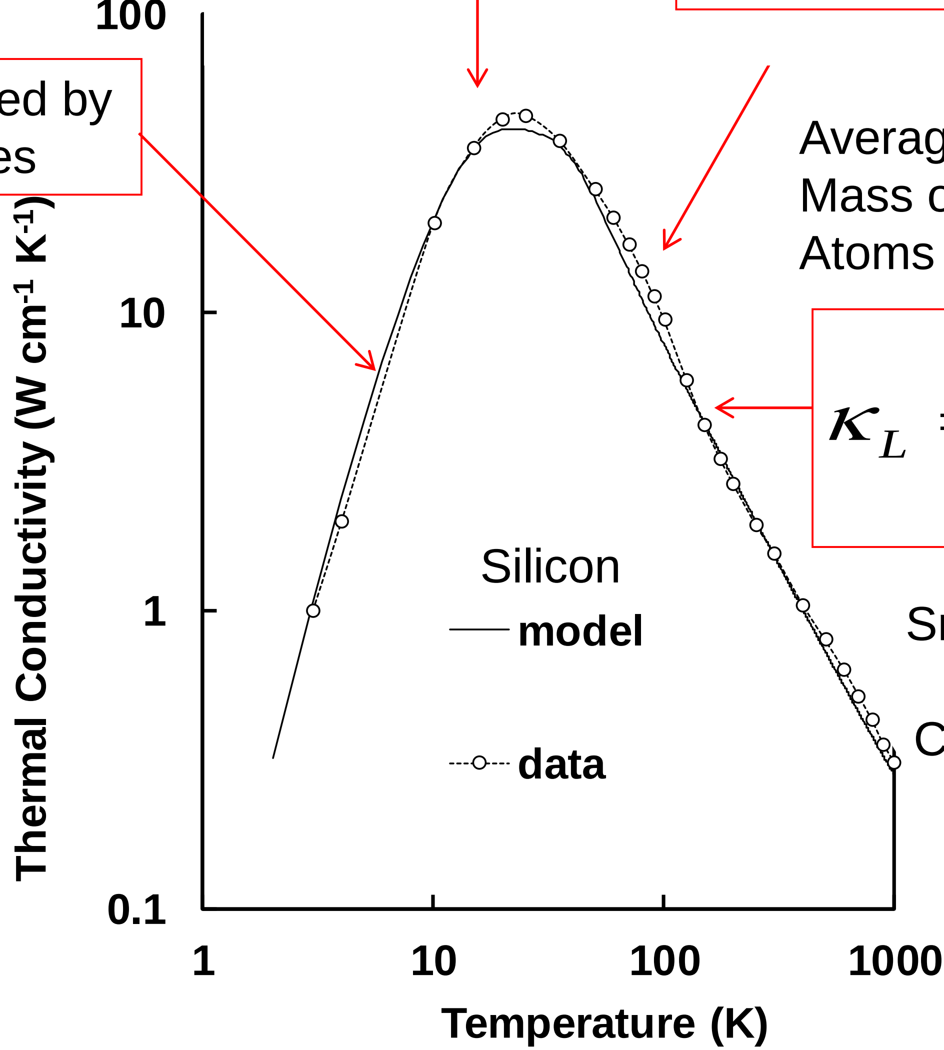
H. Jin & al. (OSU): Nature Materials ***14****, 601-606 (2015)*

Anharmonicity and Lattice Thermal Conductivity

Phonons scattered by defects (isotopes, …)

Phonons scattered by other phonons

Phonons scattered by crystal boundaries



Average Mass of Atoms

Debye Temp

Volume per atom

$$\kappa_L = A \frac{\overline{M}\theta^3 V_{atom}^{1/3}}{\gamma^2 n^{2/3} T}$$

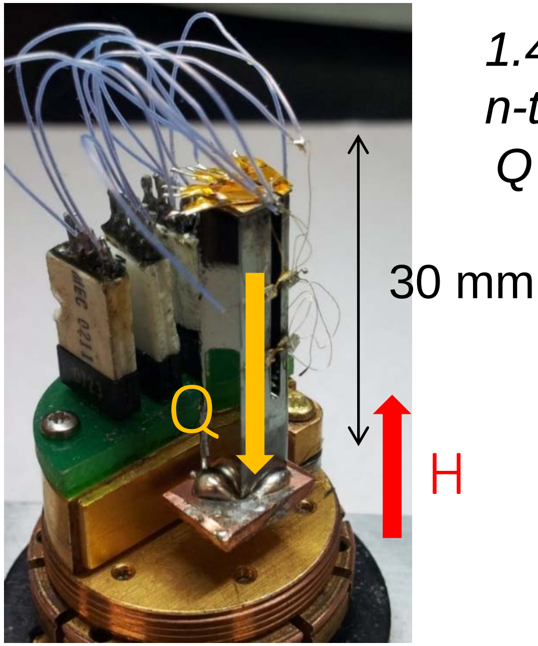
Smattering of Constants

atoms in cell

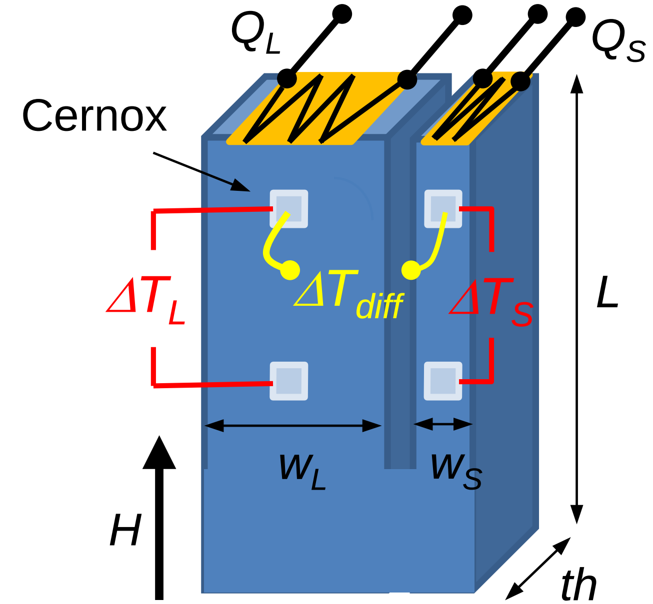
Grüneisen Parameter

Morelli, Heremans & Slack Physical Review B ***66****, 195304 (2002)*

Experimental Setup: Thermal Potentiometer



1.4×10^{15} cm^{-3} doped
n-type InSb single crystal
Q // H // [100]



w_L (3mm) ~ $3w_S$ (1mm) $\Rightarrow$ $A_L \sim 3A_S$

Measure the ratio of the heat it takes to keep the same gradient on both arms →

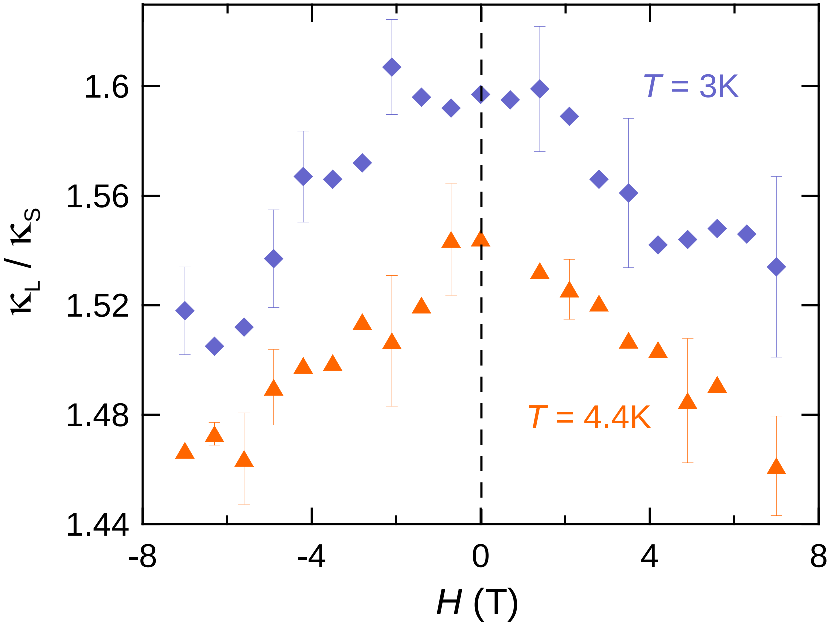
Ratio κ_L/κ_S

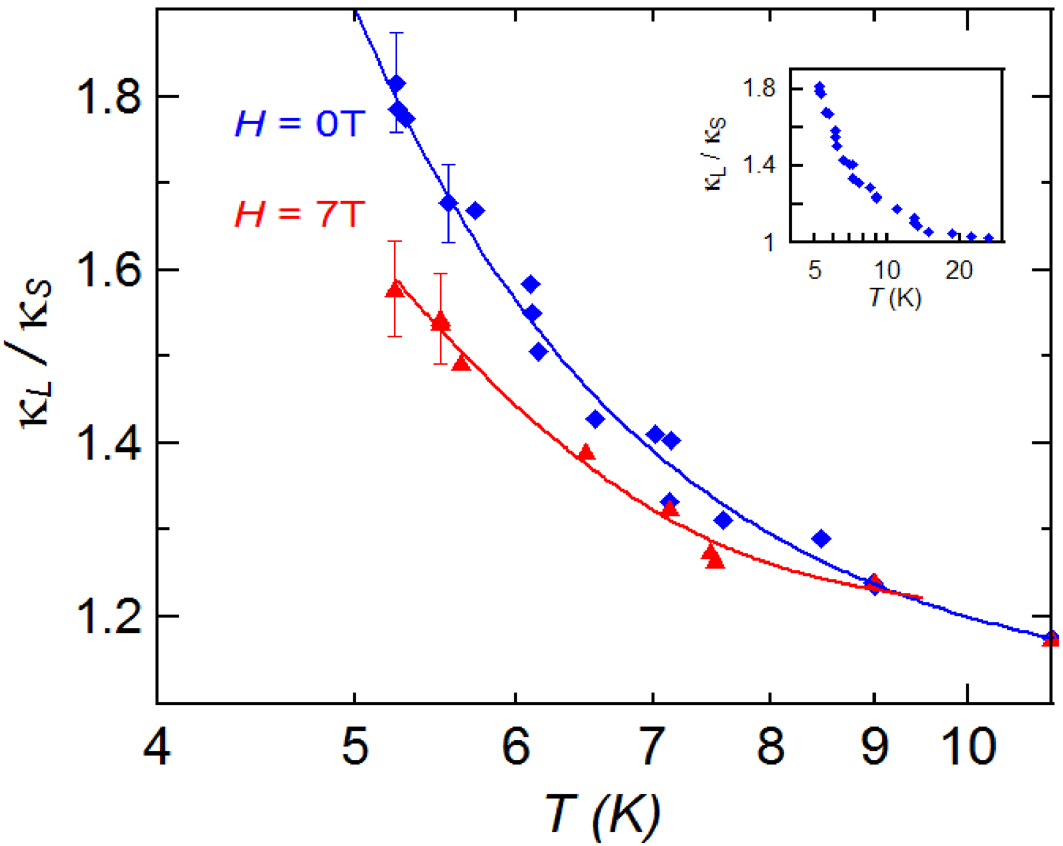
- Measurement is one of thermal flux, not temperature
- Independent of the magnetic field dependence of thermometry
- Choose temperature so that the small arm is in the size-scattering regime; the large arm is in the anharmonic regime
- The small arm serves as a reference to probe anharmonic scattering in the large arm

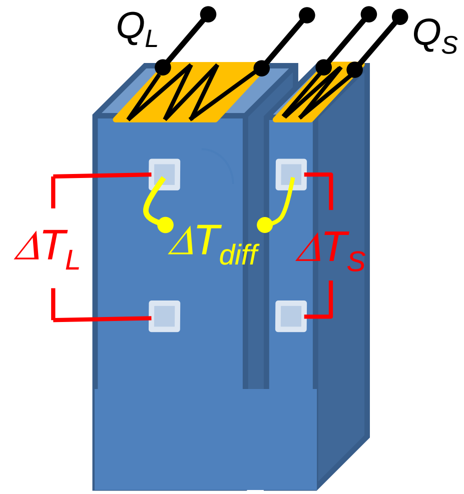
H. Jin & al. (OSU): Nature Materials ***14****, 601-606 (2015)*

Temperature and Magnetic Field Dependence

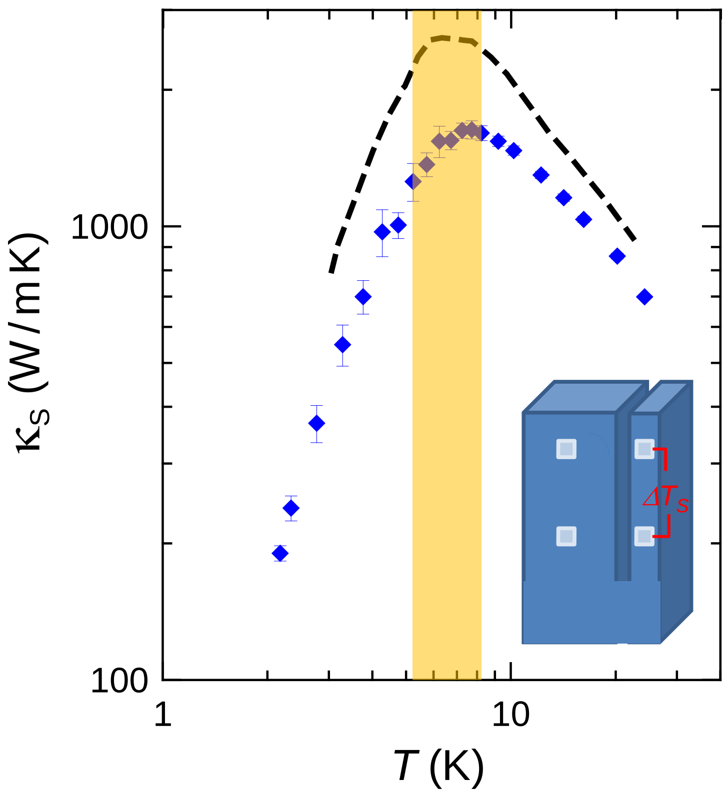
- Ratio κ_L/κ_S is magnetic-field dependent and quadratic in field







Physical Meaning of κ_L/κ_S



1. Effect occurs where the transport is still ballistic, but starts picking up a phonon scattering component.

$$\kappa = \tfrac{1}{3} C_V v \ell = \tfrac{1}{3} C_V v^2 \tau$$

$$\tau^{-1} = \tau_B^{-1} + \tau_\phi^{-1}$$

$$\frac{\kappa_L}{\kappa_S} = \frac{\tfrac{1}{3} C_V v^2 \left[\tau_{BL}^{-1} + \tau_\phi^{-1}\right]^{-1}}{\tfrac{1}{3} C_V v^2 \left[\tau_{BS}^{-1} + \tau_\phi^{-1}\right]^{-1}} = \frac{\tau_{BS}^{-1} + \tau_\phi^{-1}}{\tau_{BL}^{-1} + \tau_\phi^{-1}}$$

2. Thermal potentiometer eliminates specific heat and sound velocities from the physics of the problem.
3. If we consider the limiting case where $\tau_{BS}^{-1} >> \tau_\phi^{-1} >> \tau_{BL}^{-1}$ $\quad \dfrac{\kappa_L}{\kappa_S} = \dfrac{\tau_{BS}^{-1} + \tau_\phi^{-1}}{\tau_{BL}^{-1} + \tau_\phi^{-1}} \approx \dfrac{\tau_{BS}^{-1}}{\tau_\phi^{-1}}$
4. For long-wavelength phonons ($T << \Theta_D$)

Grüneisen

$$\tau_\phi^{-1} = A\gamma^2 T^a$$

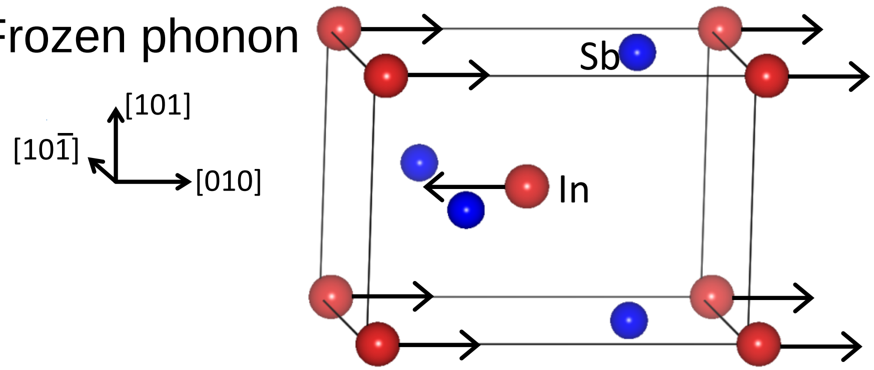
Proportionality constant

a=3 for cubic crystals (1)
a=2 for trigonal crystals (2)

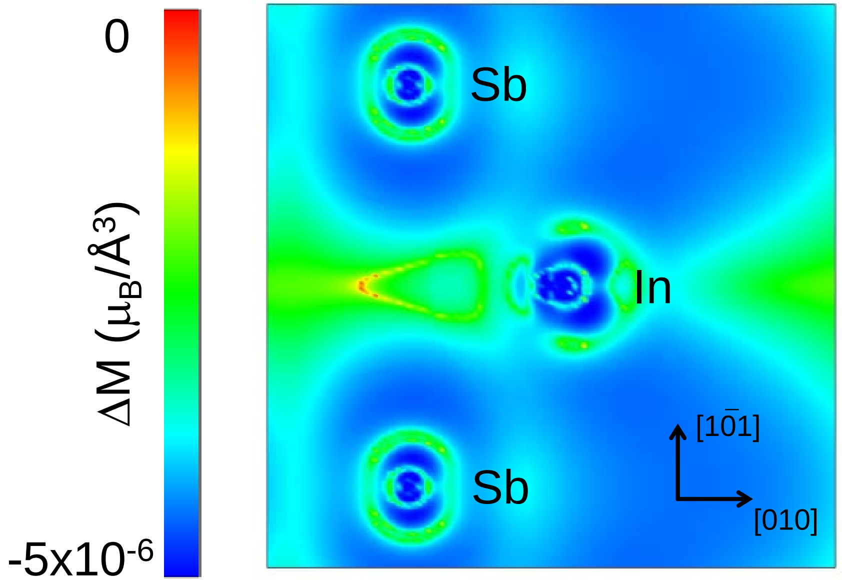
$$\boxed{\frac{\kappa_L}{\kappa_S} \approx \frac{\tau_{BS}^{-1}}{\tau_\phi^{-1}} \propto \frac{1}{\gamma^2}}$$

Theory of Phonon-perturbated Diamagnetism

Frozen phonon

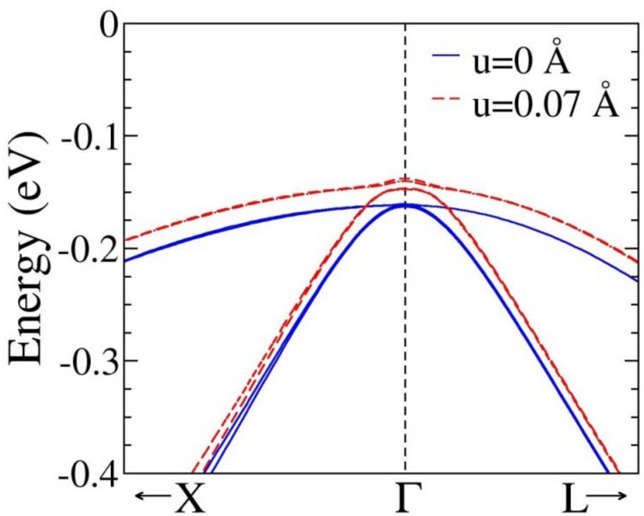


Magnetization around frozen phonon



H=7 T // [010]

Valence band structure frozen phonon



- Orbital magnetism in the valence band
- Local moments weak
- Gradients very sharp
- Magnetic force on atoms:

$$F_M(r) = \nabla\left[M(r)\cdot H\right] = H^2 \nabla \chi(r)$$

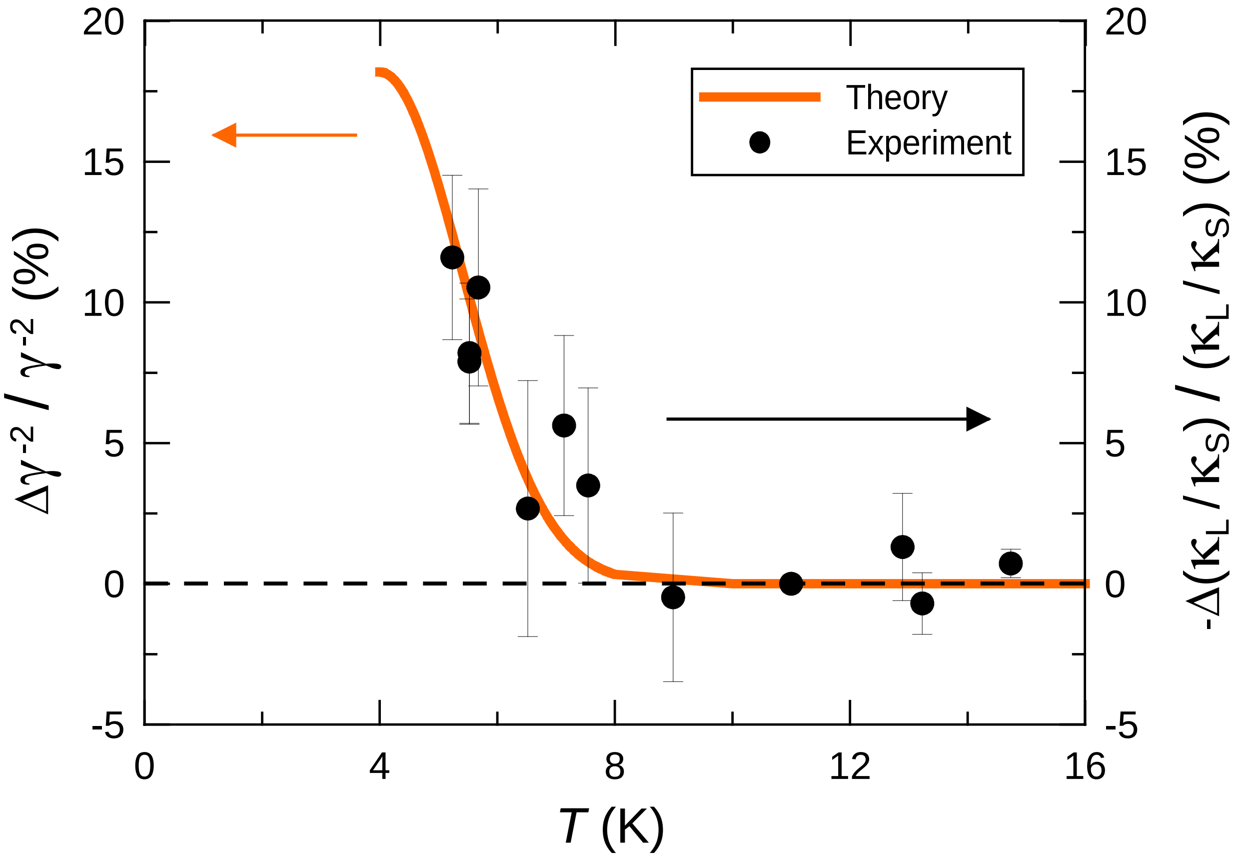
- Magnetic force is anharmonic $\rightarrow$ alters thermal conductivity

Comparison Theory – Experiment (no adjustable parameters)

$$\frac{\kappa_L}{\kappa_S} \approx \frac{\tau_{BS}^{-1}}{\tau_\phi^{-1}} \propto \frac{1}{\gamma^2}$$

=>

$$\frac{\left[\kappa_L/\kappa_S\right]_{H_{APPL}=7T} - \left[\kappa_L/\kappa_S\right]_{H_{APPL}=0T}}{\left[\kappa_L/\kappa_S\right]_{H_{APPL}=0T}} = \frac{\gamma_{H_{APPL}=7T}^{-2} - \gamma_{H_{APPL}=0T}^{-2}}{\gamma_{H_{APPL}=0T}^{-2}}$$



No adjustable parameters

H. Jin & al. (OSU): Nature Materials ***14****, 601-606 (2015)*

Summary: Major Advances in the Topic

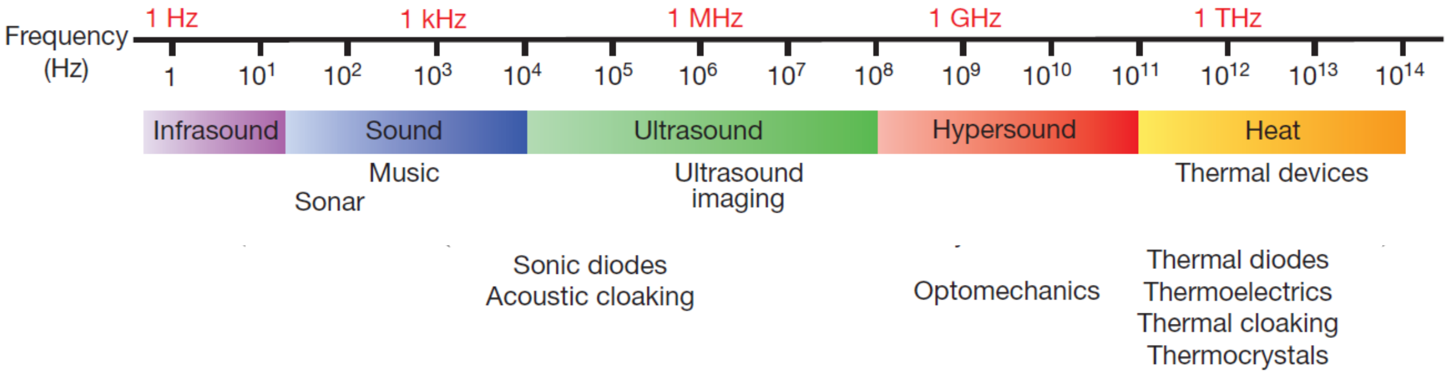
Thermal measurements access transport properties of entities that are not electrically charged:

1. Magnons and spins
2. Phonons

More Examples:

"Advances in sonic and thermal diodes, optomechanical crystals, acoustic and thermal cloaking, hypersonic phononic crystals, thermoelectrics, and thermocrystals herald the next technological revolution in phononics" (verbatim)

M. Moldovan, "Sound and heat revolutions in phononics", Nature ***503*** *209 (2013)*



Scientific Questions for the Near Future

- Measure/harness topological protection in electrical insulators

- Magnon and phonon dispersions and hybridized magnon/phonon band crossings
  - Identify topological invariants
  - Weyl points

- Mobility enhancement?
- Unidirectional heat propagation?
- Thermally activated mechanical (phononic) structures

E.g.:
Kane-Lubensky invariant: very-low-symmetry flexible structures
C. L. Kane & T. C. Lubensky, "Topological boundary modes in isostatic lattices", Nature Physics ***10****, pages 39–45 (2014)*

N.B. Brownian motion is observable on pollen ≈ 100 μm across (via molecular motion).

Technologies with Game-changing Potential

- **Present:**
    - Heat management is what limits the density of integrated circuits today.
    - Technological importance of non-linear thermal circuit elements:
        - Solid-state refrigeration
        - Waste-heat scavenging using non-linear thermal devices
        - Thermal diode applications
        - Thermal switch applications
        - Thermal regulator and negative differential resistance circuits

G. Wehmeyer, T. Yabuki, C. Monachon, J. Wu, and C. Dames, "Thermal diodes, regulators, and switches: physical mechanisms and potential applications," Appl. Phys. Rev. ***4****, 041304 (2017).*

- **Future:**

- Imagine and develop fundamentally new principles for the operation of heat-to-work engines, based on advances in thermal rectification and switching