Thermoelectricity: From Atoms to Systems

Week 2: Thermoelectric Transport Parameters

Lecture 2.7: **Using full band dispersions (ex. of Bi_2Te_3)**

Bonus Lecture

By Dr. Jesse Maassen
Postdoctoral Researcher, Electrical and Computer Engineering
Purdue University

motivation

Why use full band dispersions of materials?

1) Extract meaningful and reliable parameters.

2) Predict the properties of new materials, where little or no experimental data is available.

Bonus lecture: outline

Work through an example of how to extract the TE properties using full band dispersions (case of Bi_2Te_3).

1) Review of TE equations

2) Number of modes (electron & phonon)

3) Electronic TE properties (electrical conductivity, Seebeck, electronic thermal conductivity)

4) Lattice thermal conductivity

review: TE equations

electrical current:

$$E_x = \rho J_x + S\frac{dT}{dx}$$

heat current:

$$J_{Qx} = \pi J_x - (\kappa_e + \kappa_L)\frac{dT}{dx}$$

Differential lattice thermal conductivity

$$\kappa_L = \int_0^{+\infty} \kappa_L'(\hbar\omega)\, d(\hbar\omega)$$

Phonons

$$\sigma = \int_{-\infty}^{+\infty} \sigma'(E)\, dE \qquad \rho = 1/\sigma$$

Differential electrical conductivity

$$S = \frac{-\frac{1}{qT}\int_{-\infty}^{+\infty}(E - E_F)\sigma'(E)\, dE}{\int_{-\infty}^{+\infty}\sigma'(E)\, dE} \qquad \pi = TS$$

$$\kappa_0 = \frac{1}{q^2 T}\int_{-\infty}^{+\infty}(E - E_F)^2\, \sigma'(E)\, dE$$

$$\kappa_e = \kappa_0 - T\sigma S^2$$

Electrons

review: differential conductivity

Boltzmann approach

$$\sigma'(E) = q^2 \Sigma_{\text{el}}(E)\left[-\frac{\partial f}{\partial E}\right] \qquad \kappa'_L(\hbar\omega) = \frac{(\hbar\omega)^2}{T}\Sigma_{\text{ph}}(\hbar\omega)\left[-\frac{\partial n}{\partial(\hbar\omega)}\right]$$

Transport distribution

$$\Sigma(E) = \frac{1}{\Omega}\sum_{\vec{k}}\left[v_x\left(\vec{k}\right)\right]^2 \tau\left(\vec{k}\right)\delta\left(E - E_{\vec{k}}\right) \longrightarrow \Sigma(E) = \frac{1}{h}M(E)\lambda(E)$$

Transport distribution has clear physical interpretation, the product of:

- ✓ Number of modes *(depends only on dispersion, easy to extract).*
- ✓ Mean-free-path for backscattering *(depends on scattering physics and dispersion).*

Bonus lecture: outline

Work through an example of how to extract the TE properties using full band dispersions (case of Bi_2Te_3).

1) Review of TE equations
2) Number of modes (electron & phonon)
3) Electronic TE properties (electrical conductivity, Seebeck, electronic thermal conductivity)
4) Lattice thermal conductivity

number of modes: definition

$$M_{3D}(E) = \frac{1}{A} \sum_{\vec{k}_\perp, n^+} \Theta\left(E - E_{\vec{k}_\perp}\right)$$

Number of conducting channels per cross sectional area *A*, units: m^{-2}

Sum over k-points in the plane perpendicular to transport

Sum over all bands or branches with positive velocity

Heaviside function: If the band has a state at energy *E*, assign one mode

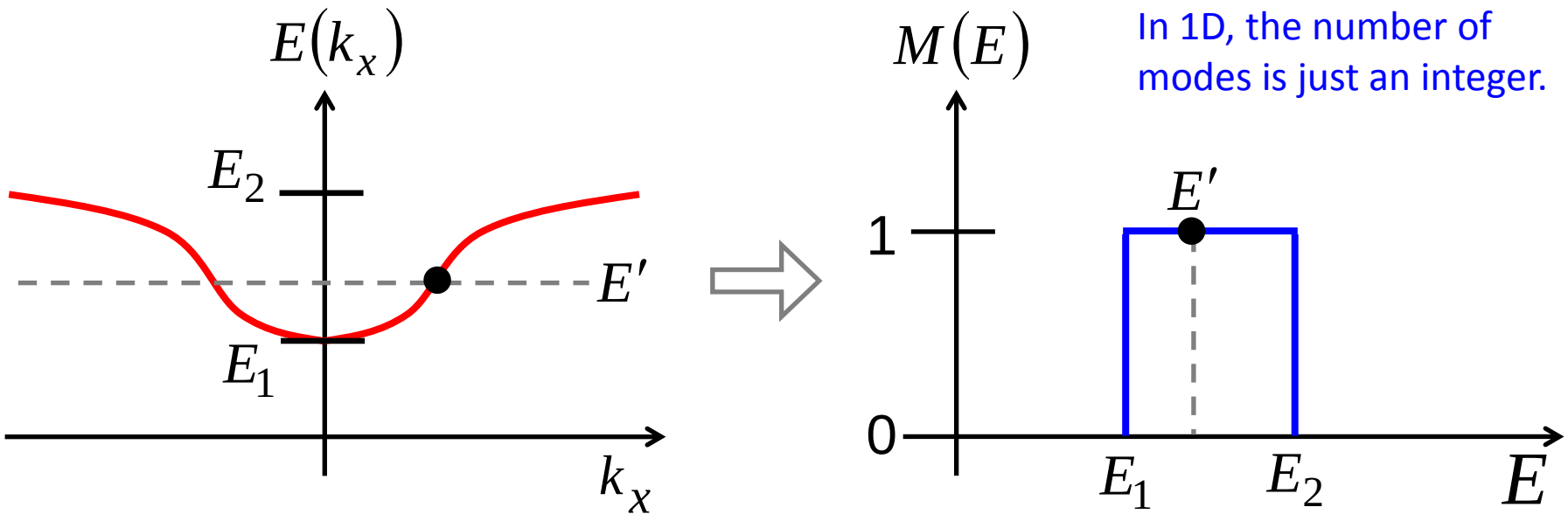
- ✓ Number of modes is easy to calculate *!*
- ✓ "Band counting" is an efficient approach to extract *M*(*E*).

Demonstrate approach with two simple examples (1D and 2D cases) →

number of modes: 1D example

$$M_{1D}(E) = \sum_{n^+} \Theta(E - E_0)$$

Transport along x, no periodicity along y and z, units: $m^{-d+1} = m^0$



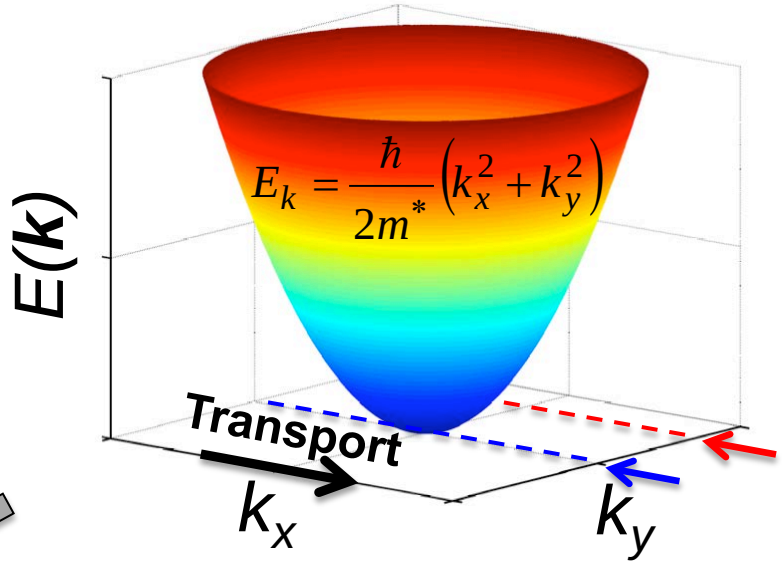
In 1D, the number of modes is just an integer.

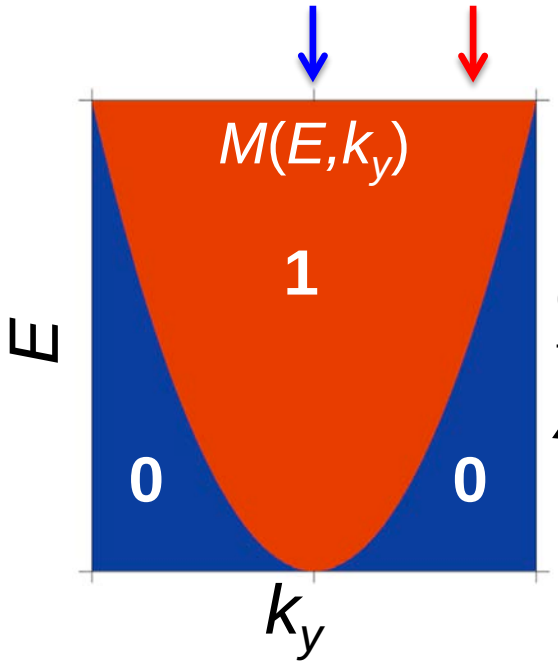
Reference: Jeong, Kim, Luisier, Datta, Lundstrom, J. Appl. Phys. **107**, 023707 (2010).

number of modes: 2D example

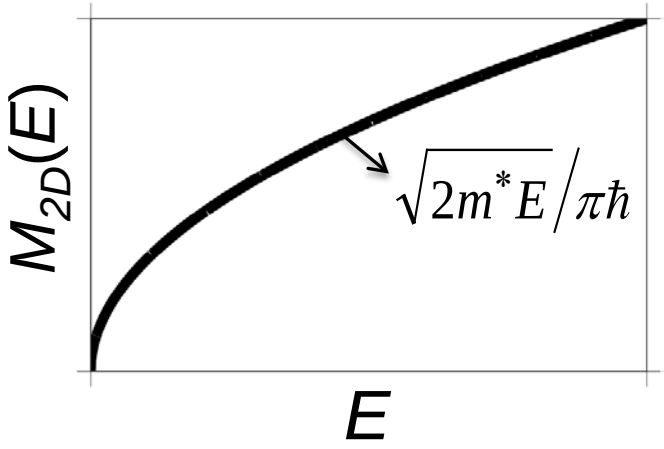
$$M_{2D}(E) = \frac{1}{2\pi}\int \sum_{n^+} \Theta\left(E - E_{k_y}\right) dk_y$$

Transport along x, periodicity along y, units: $m^{-d+1} = m^{-1}$





(Same as 1D example, but for each k_y.)



number of modes: effect of dimensionality

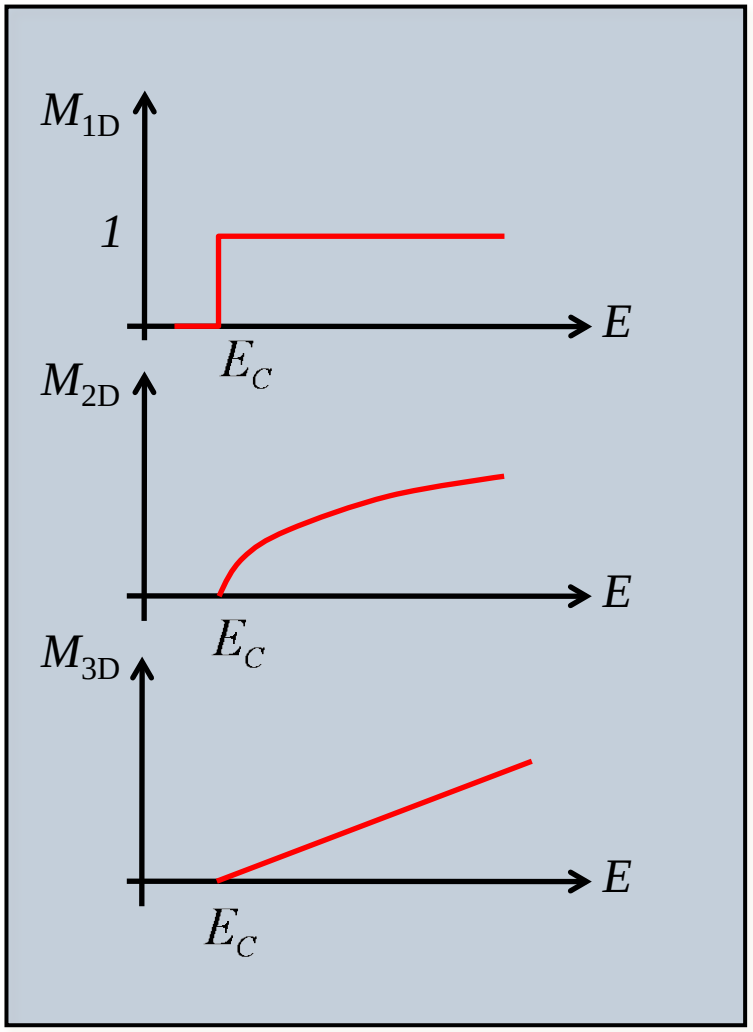
Parabolic bands

$$M(E) = M_{1D}(E) = \Theta(E - E_C)$$

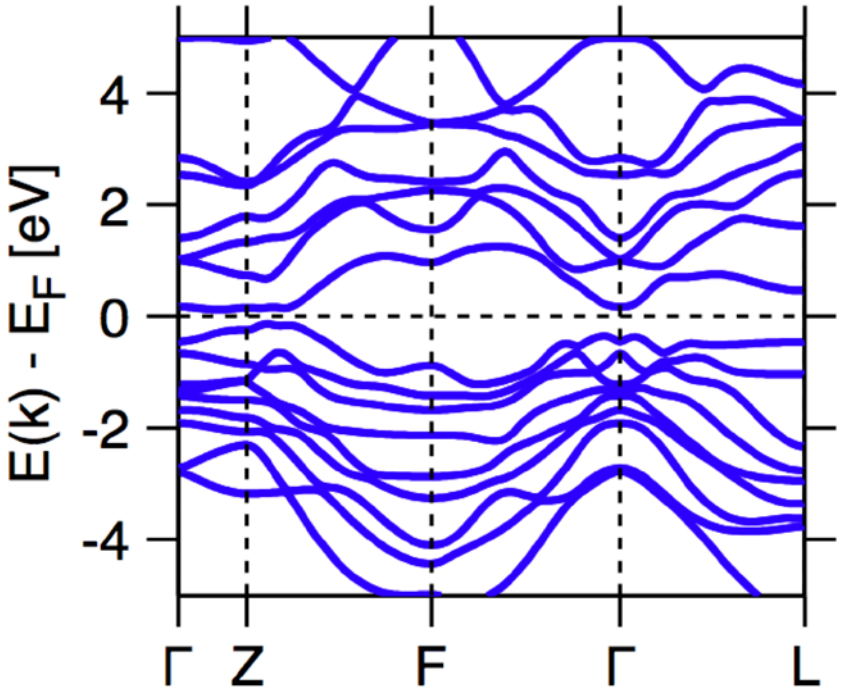
$$M(E) = W\, M_{2D}(E) = W\,\frac{\sqrt{2m^*(E - E_C)}}{\pi\hbar}$$

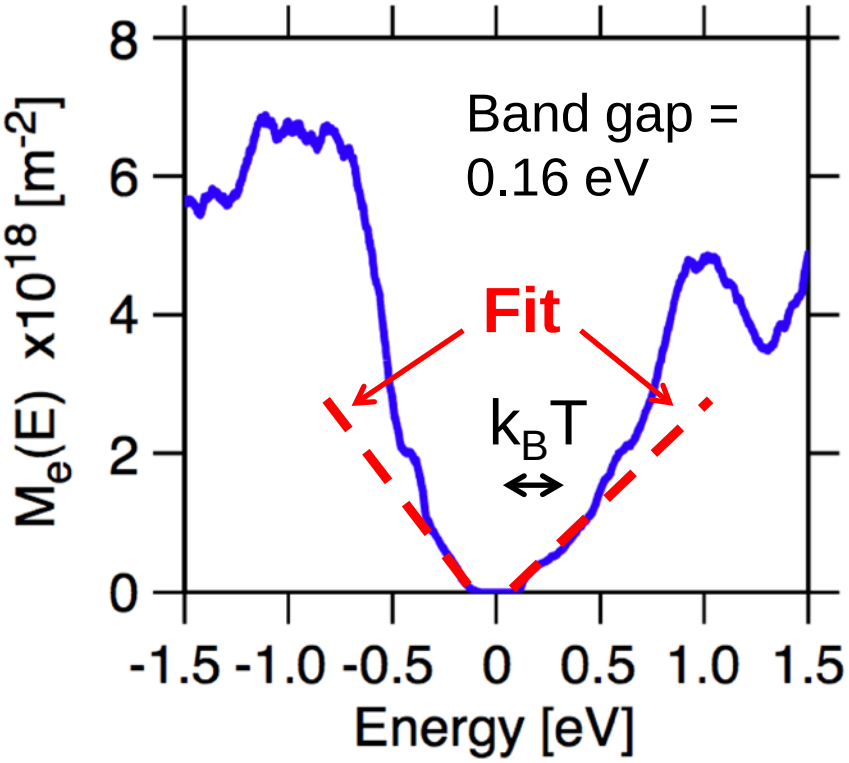
$$M(E) = A M_{3D}(E) = A\frac{m^*(E - E_C)}{2\pi\hbar^2}$$

$$E(\vec{k}) = E_C + \hbar^2 k^2 / 2m^*$$



number of modes: Bi_2Te_3 (*electron*)

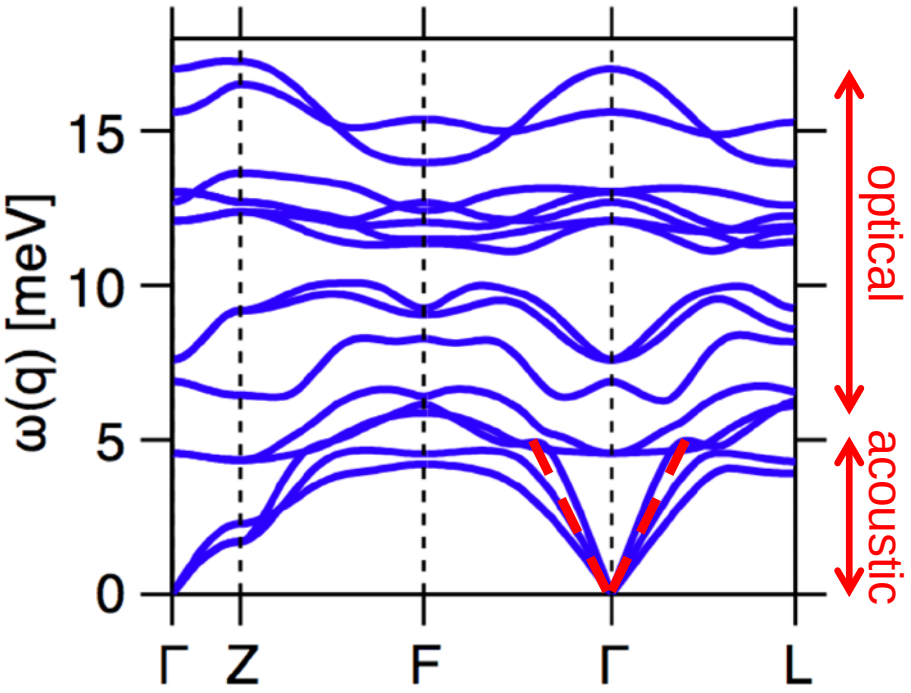


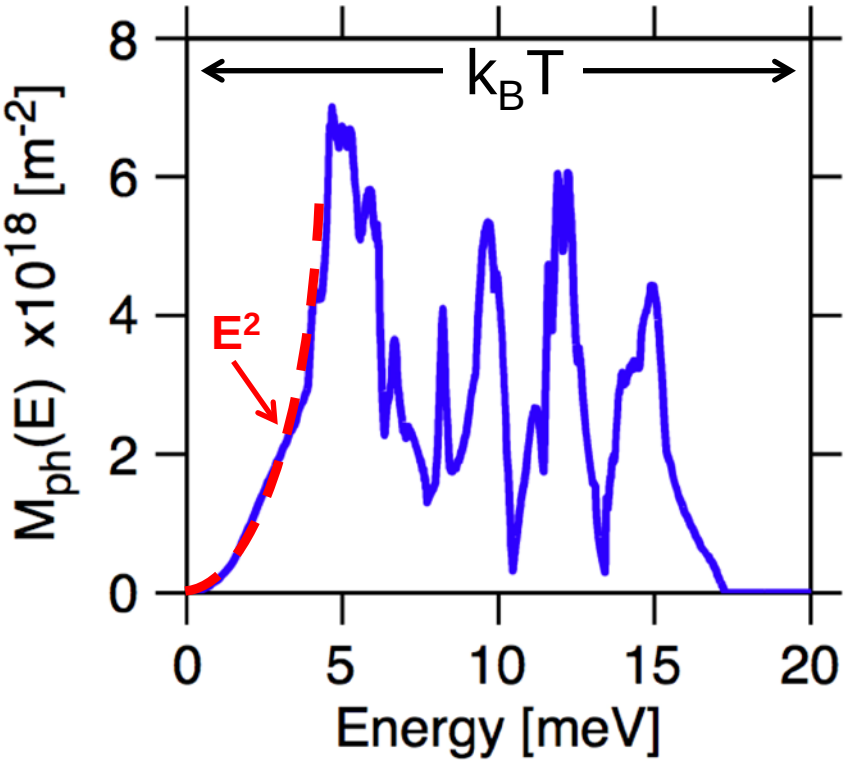


- Complicated band structure.
- Not well represented by the parabolic band approximation.

- Roughly fit by a straight line near band edges (i.e. parabolic band approx.).
- Surprising given the non-parabolic *E*(*k*).
- Only small fraction of modes are thermally active.

number of modes: Bi_2Te_3 (*phonon*)





- Complicated band structure.
- Not well represented by simple dispersion model.

- At low energy, modes fit well with quadratic function (i.e. Debye model).
- At room temperature all modes are thermally active.

Bonus lecture: outline

Work through an example of how to extract the TE properties using full band dispersions (case of Bi_2Te_3).

1) Review of TE equations

2) Number of modes (electron & phonon)

3) Electronic TE properties (electrical conductivity, Seebeck, electronic thermal conductivity)

4) Lattice thermal conductivity

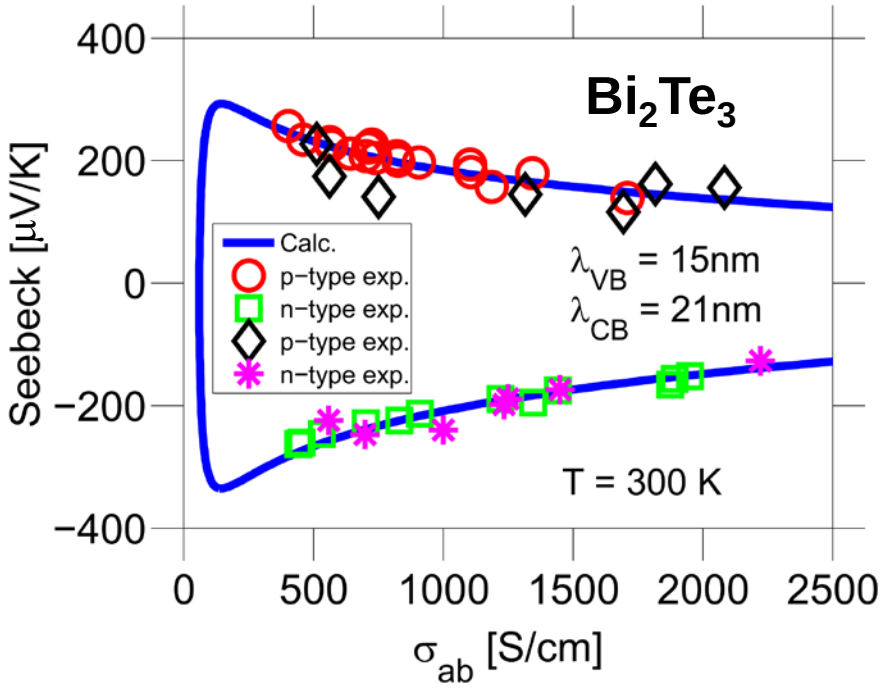
electronic TE properties

$$\sigma'(E) = \frac{2q^2}{h} M_{el}(E) \lambda_{el}(E) \left[-\frac{\partial f}{\partial E} \right]$$

Central quantity to calculate the electronic TE coefficients

Mean-free-path for backscattering

- A constant MFP is reasonable in many cases (exact when assuming parabolic bands and electron-phonon scattering*).
- Calibrate MFP to experimental data:
- Can calculate MFP, but can be difficult.

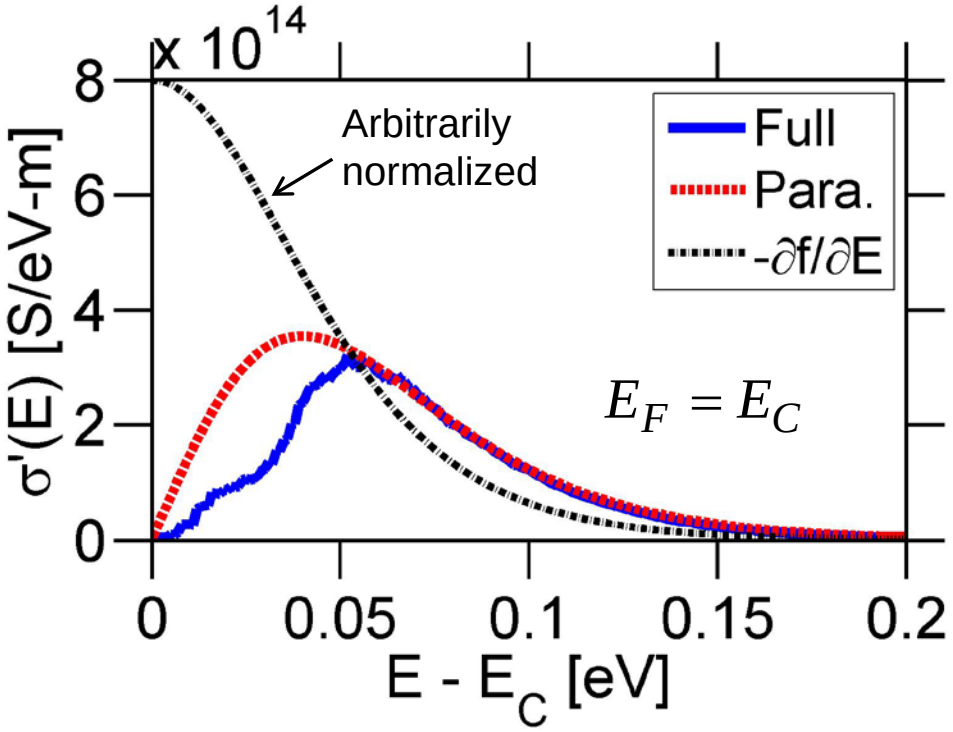


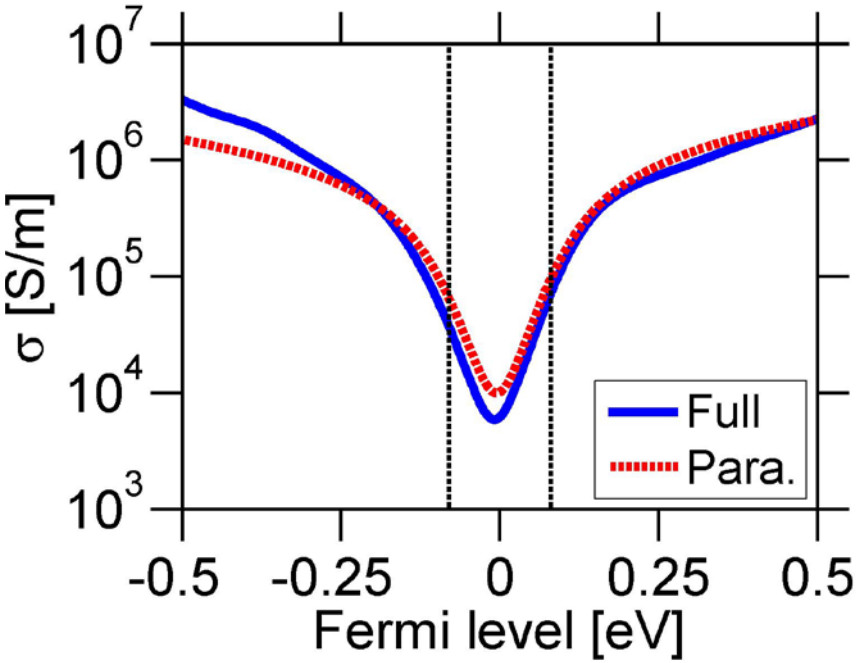
*Mark Lundstrom, *Fundamentals of Carrier Transport*, Cambridge University Press, 2000.

electronic TE properties: *conductivity*

$$\sigma'(E) = \frac{2q^2}{h} M_{el}(E) \lambda_{el}(E) \left[-\frac{\partial f}{\partial E} \right]$$

$$\sigma = \int_{-\infty}^{+\infty} \sigma'(E) dE$$

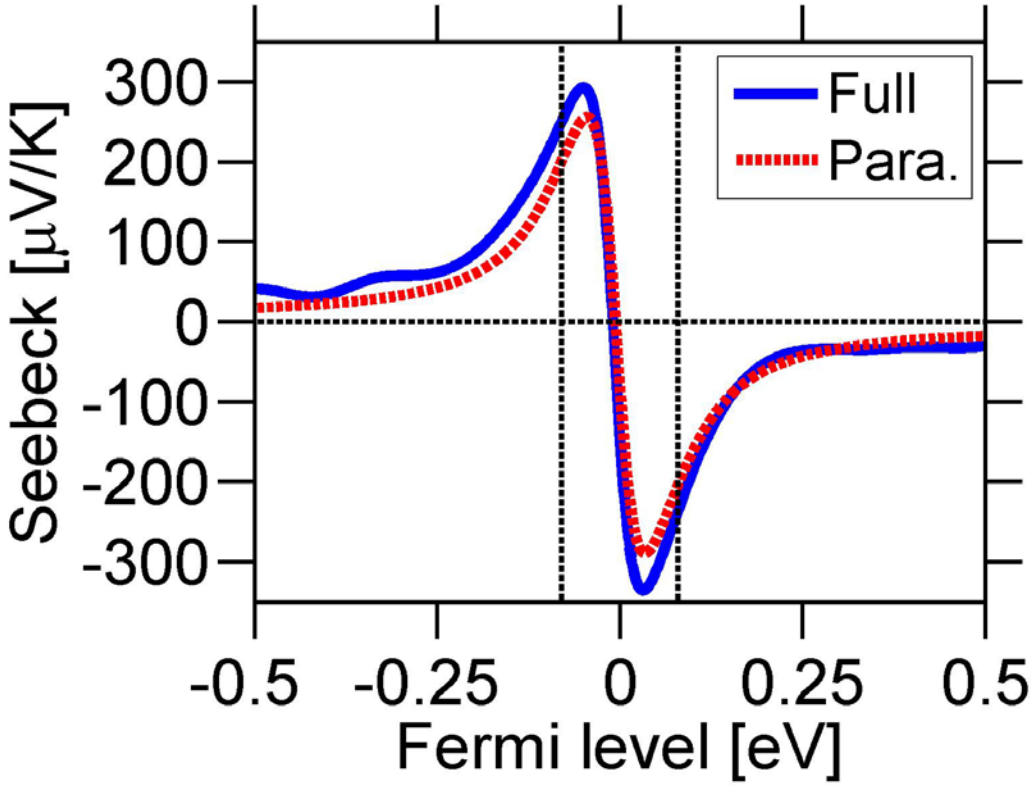




Bi_2Te_3

electronic TE properties: *Seebeck*

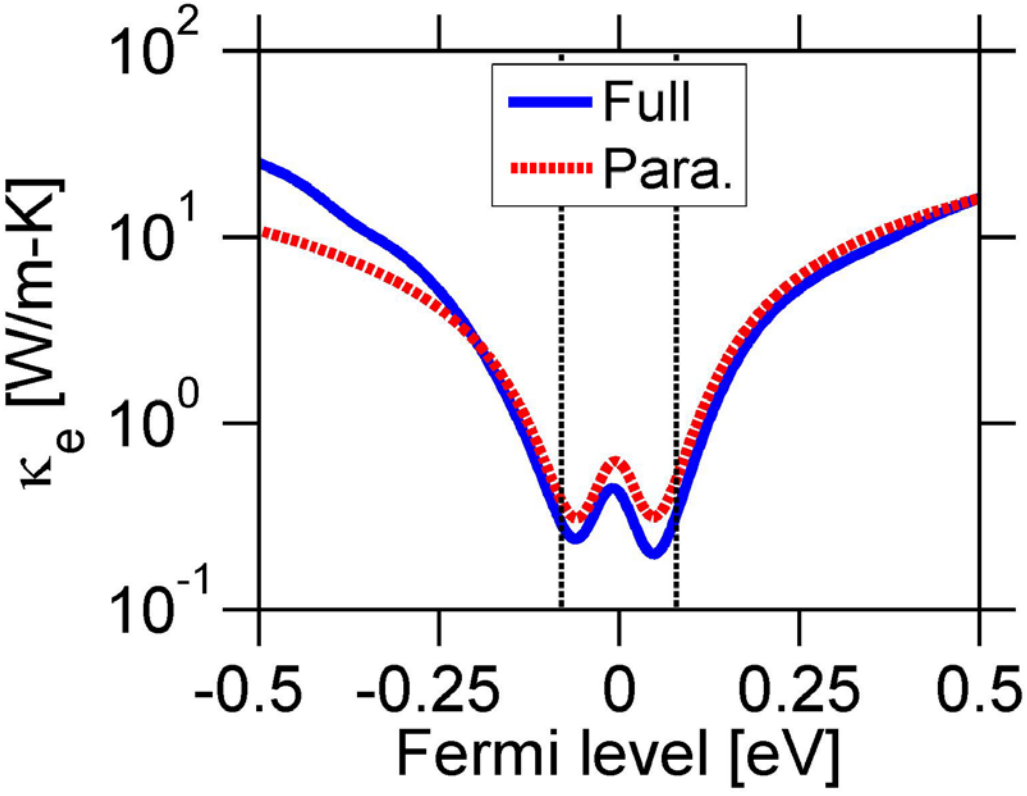
$$S = -\frac{1}{qT}\int_{-\infty}^{+\infty}(E - E_F)\sigma'(E)dE \Big/ \int_{-\infty}^{+\infty}\sigma'(E)dE$$



Bi_2Te_3

electronic TE properties: *thermal conductivity*

$$\kappa_e = \frac{1}{q^2 T}\int_{-\infty}^{+\infty}(E - E_F)^2 \sigma'(E) dE - T\sigma S^2$$



Bi_2Te_3

Bonus lecture: outline

Work through an example of how to extract the TE properties using full band dispersions (case of Bi_2Te_3).

1) Review of TE equations

2) Number of modes (electron & phonon)

3) Electronic TE properties (electrical conductivity, Seebeck, electronic thermal conductivity)

4) Lattice thermal conductivity

Bonus lecture: outline

$$\kappa_L'(\hbar\omega) = \frac{(\hbar\omega)^2}{hT} M_{\text{ph}}(\hbar\omega)\lambda_{\text{ph}}(\hbar\omega)\left[-\frac{\partial n}{\partial(\hbar\omega)}\right] \qquad \kappa_L = \int_0^\infty \kappa_L'(\hbar\omega)d(\hbar\omega)$$

Mean-free-path for backscattering

- Typical scattering mechanisms for phonons*:
  - ✓ Umklapp scattering (phonon-phonon)
  - ✓ Boundary scattering
  - ✓ Defect scattering

$$\lambda = 2\frac{\langle v_x^2 \rangle}{\langle |v_x| \rangle}\tau$$

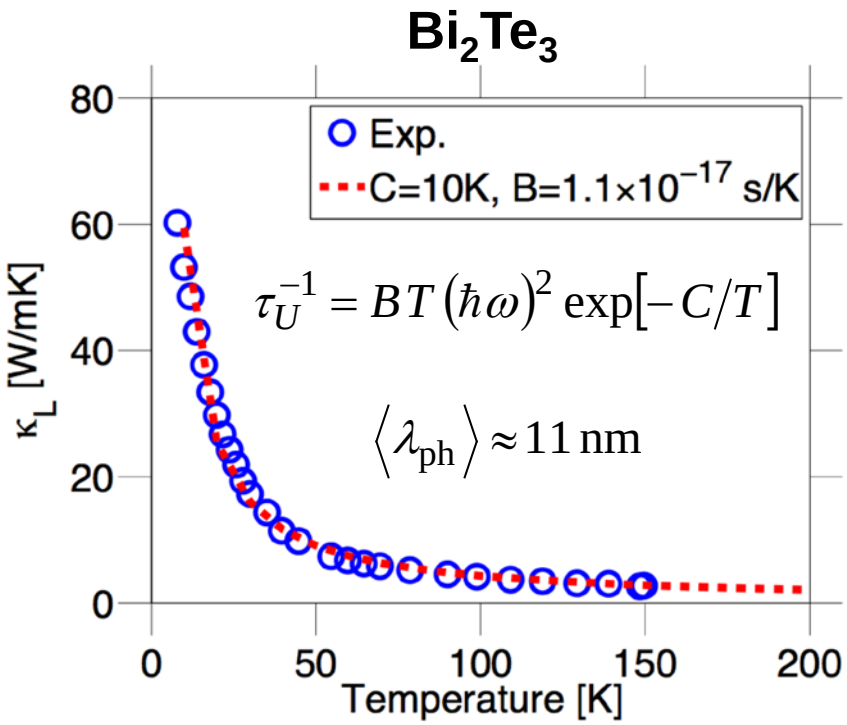
$$\tau^{-1} = \tau_1^{-1} + \tau_2^{-1} + \cdots$$

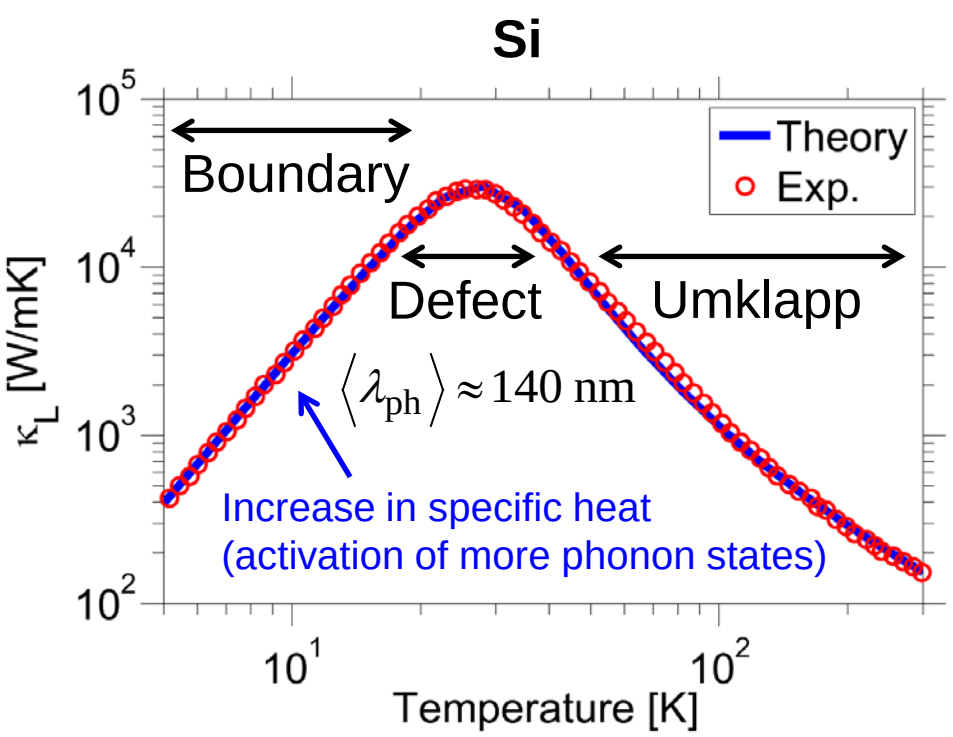
- Calibrate MFP to experimental data (next slide).
- Calculate MFP, but can be difficult.

*Jeong, Datta, Lundstrom, J. Appl. Phys. **109**, 073718 (2011).

lattice thermal conductivity: *calibrate MFP*

$$\kappa_L'(\hbar\omega) = \frac{(\hbar\omega)^2}{hT} M_{\text{ph}}(\hbar\omega)\lambda_{\text{ph}}(\hbar\omega)\left[-\frac{\partial n}{\partial(\hbar\omega)}\right] \qquad \kappa_L = \int_0^\infty \kappa_L'(\hbar\omega)d(\hbar\omega)$$

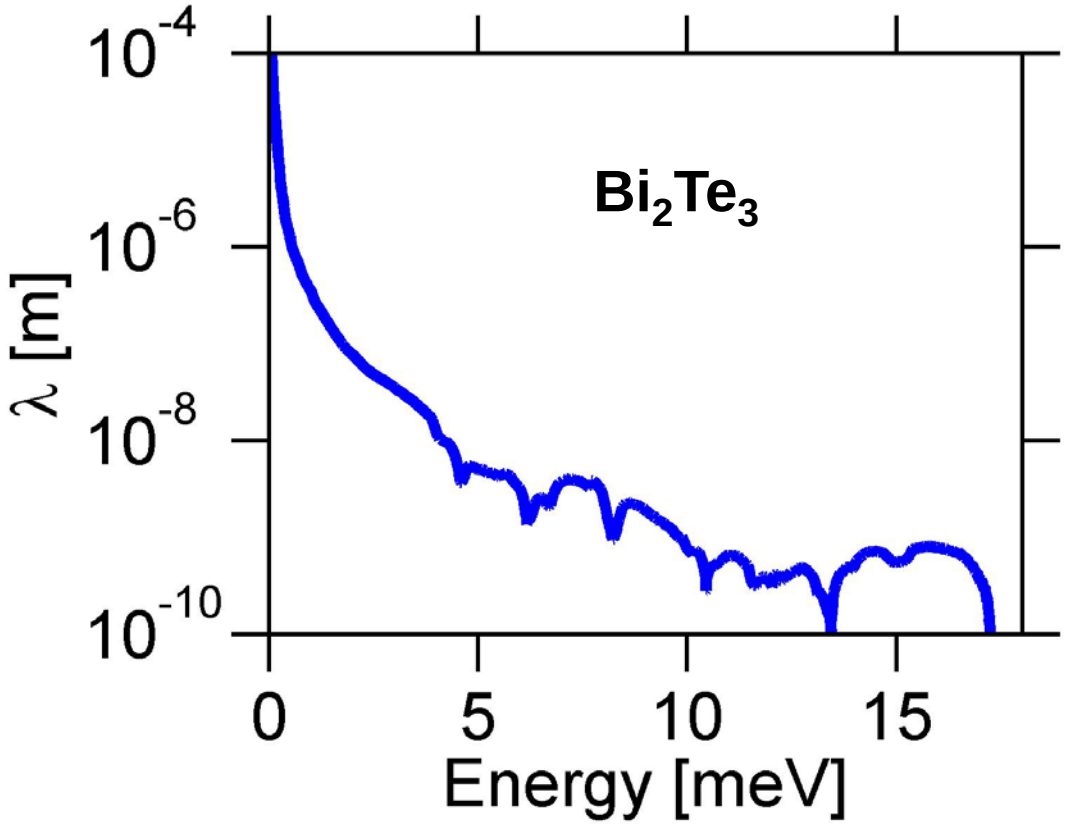




lattice thermal conductivity: *energy-dep. MFP*

$$\kappa_L'(\hbar\omega) = \frac{(\hbar\omega)^2}{hT} M_{\text{ph}}(\hbar\omega)\lambda_{\text{ph}}(\hbar\omega)\left[-\frac{\partial n}{\partial(\hbar\omega)}\right] \qquad \kappa_L = \int_0^\infty \kappa_L'(\hbar\omega)d(\hbar\omega)$$

- The MFP of phonons can vary by many orders of magnitude.
- In stark contrast to the case of electrons, where the MFP is well approximated by a constant.



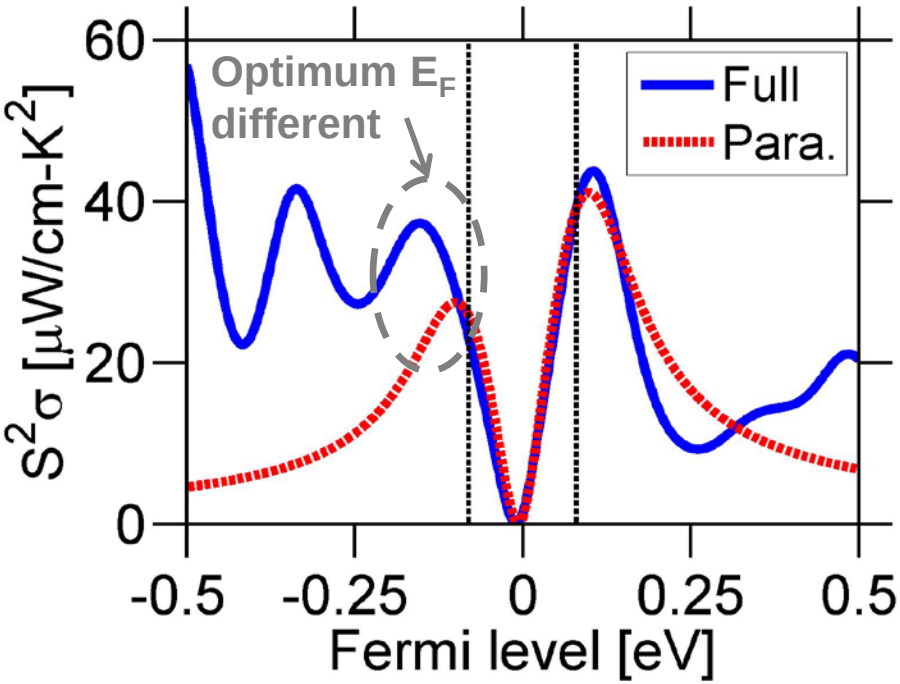
TE performance of Bi_2Te_3

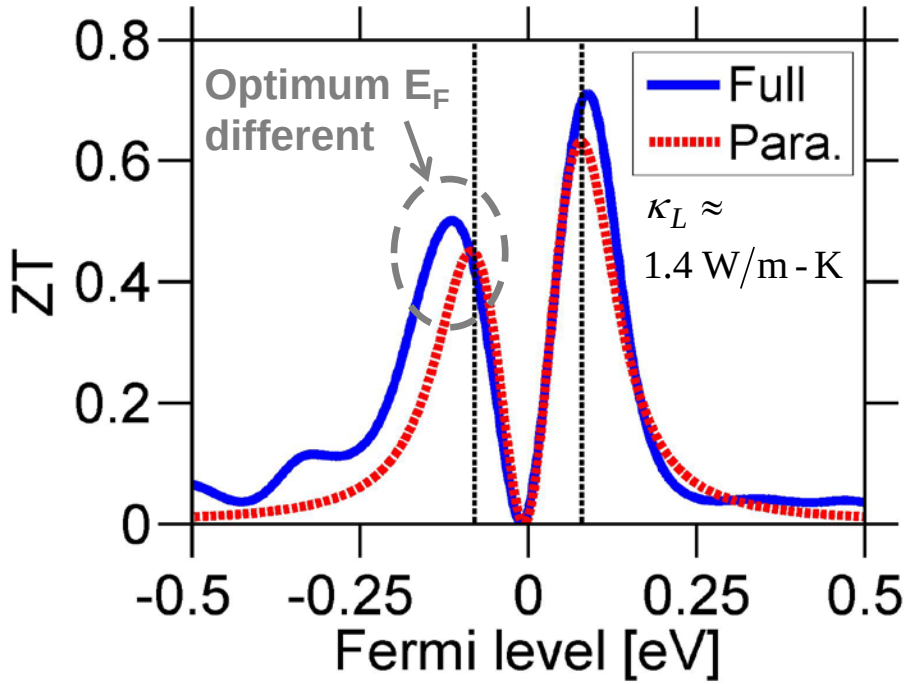
Combine electronic and lattice TE properties to determine TE performance metrics:

Power factor ($S^2\sigma$)

Features not present with simple model *!*

Figure of merit, $ZT=S^2\sigma T / (\kappa_e+ \kappa_L)$





summary: TE properties from full band approach

- Full dispersion-based TE calculations, for electrons and phonons, are particularly useful for
  - ✓ Provide meaningful / accurate physical parameters
  - ✓ Studying / predicting the properties of novel material systems
- Within the Landauer approach, which readily spans ballistic-to-diffusive transport regimes, one needs two things:
  - ✓ **Number of modes**: easy to calculate from general dispersion using the “band counting” method.
  - ✓ **Mean-free-path (MFP) for backscattering**: can calibrate to exp. using phenomenological expressions (easy) or calculate from first principles (difficult).
- Realistic descriptions of TE materials are not always well represented by simple band dispersions. Valuable skill to calculate and predict the TE properties of materials with complex electronic states.

summary: TE properties from full band approach

LanTraP tool available online on *nanoHUB*:

nanohub.org/resources/lantrap