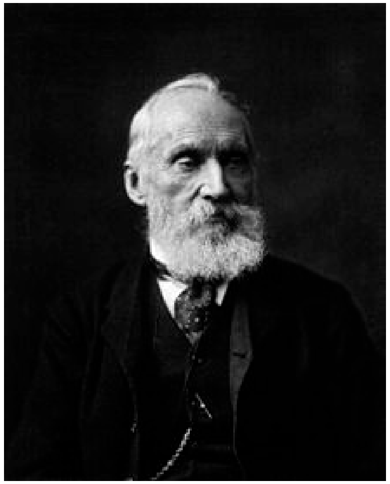
Review: Nanostructuring thermoelectrics to improve device performance

By: Bryan R. Goldsmith

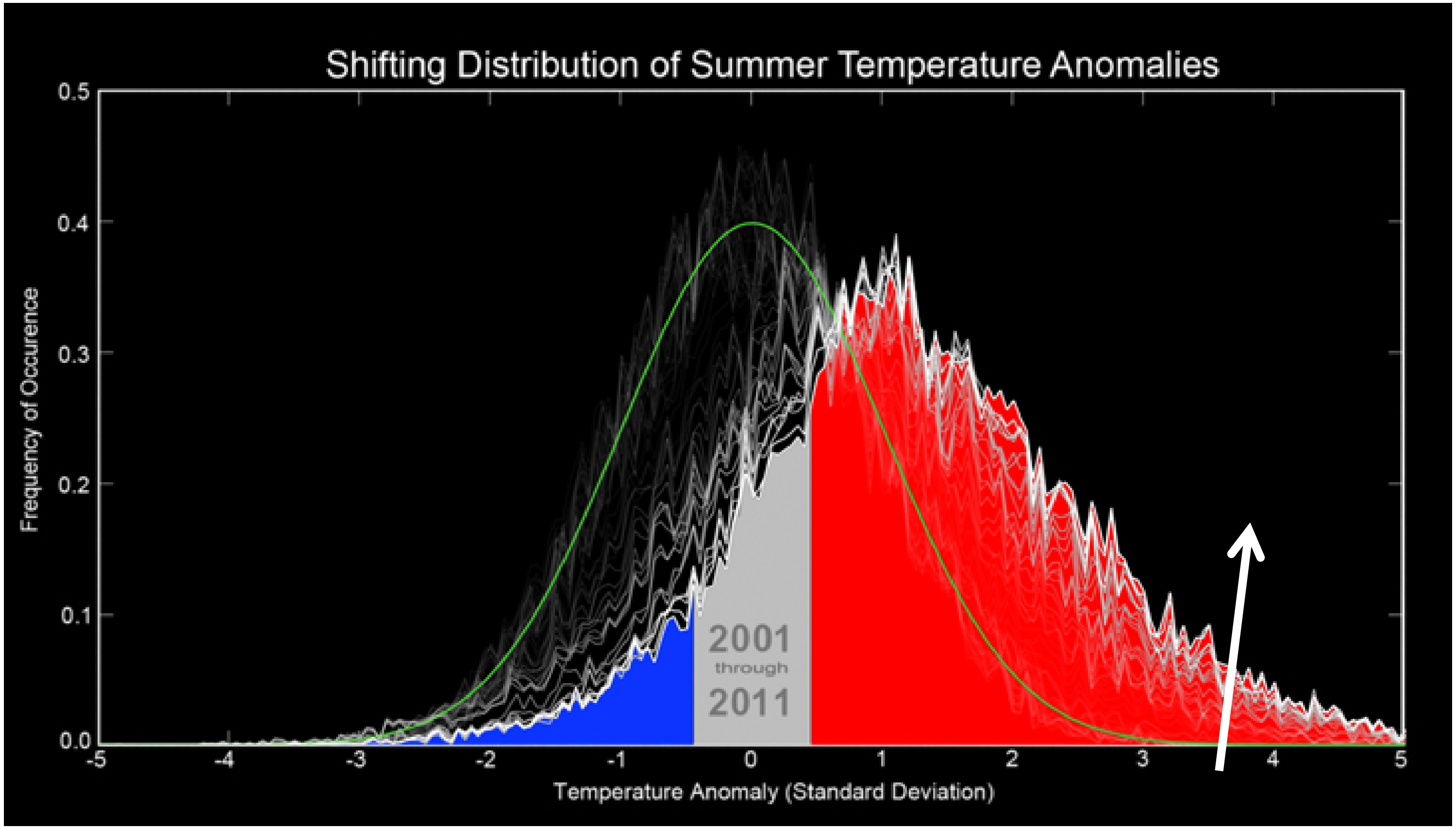
Seebeck

Peltier

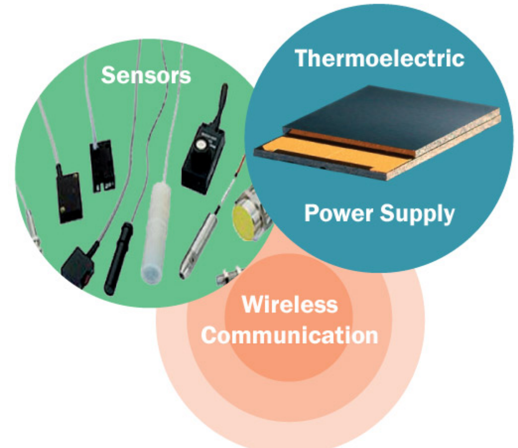


Lord Kelvin

Unprecedented probability of extreme temperatures

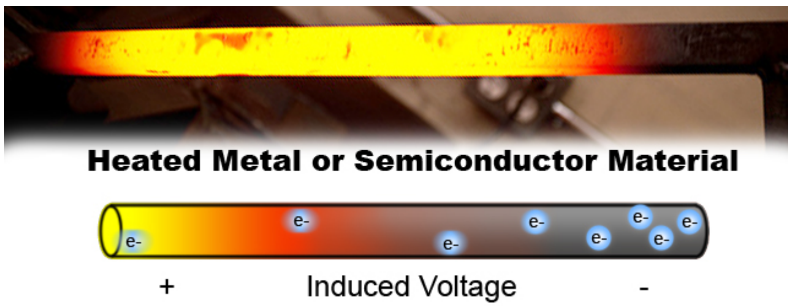


Thermoelectrics are part of the energy solution



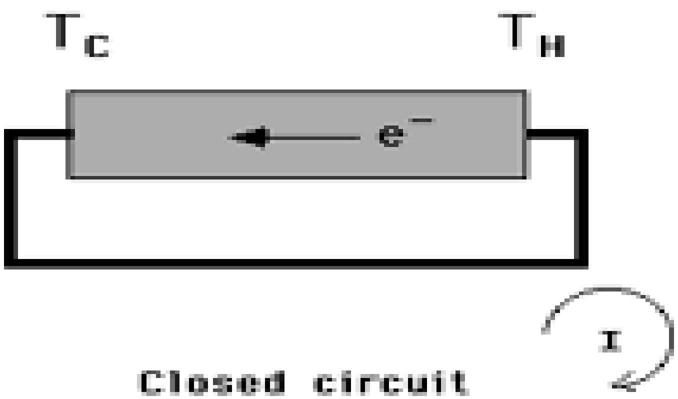
Junction of dissimilar metals produces an electric current when exposed to a temperature gradient

Seebeck effect (1821)



Seebeck coefficient

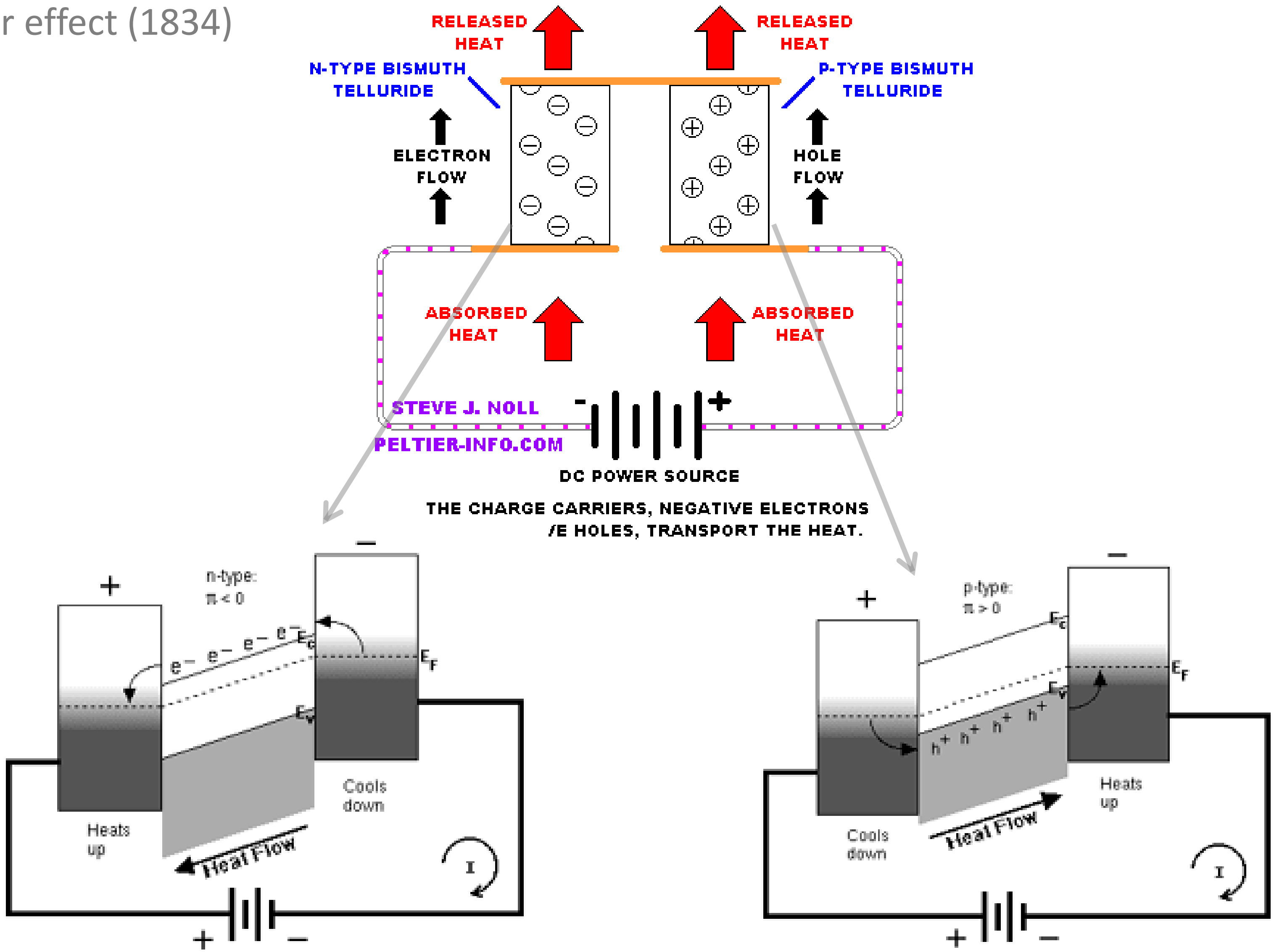
$\alpha = \Delta V / \Delta T$



http://maeweb.ucsd.edu/node/29

Convert electrical energy into a temperature gradient

Peltier effect (1834)



The Figure of Merit (ZT) relates to device efficiency

Seebeck coefficient

Electrical conductivity

$$ZT = \frac{\alpha^2 \sigma T}{\kappa} = \frac{\alpha^2 \sigma T}{\kappa_E + \kappa_L}$$

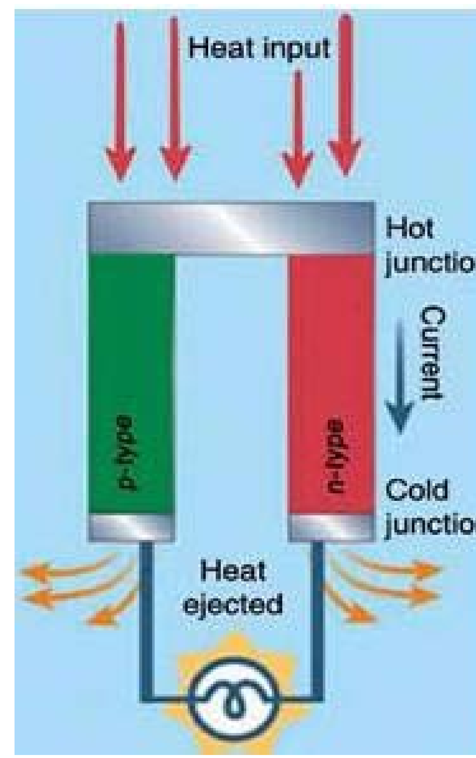
Lattice thermal conductivity (from phonons)

Electronic thermal conductivity

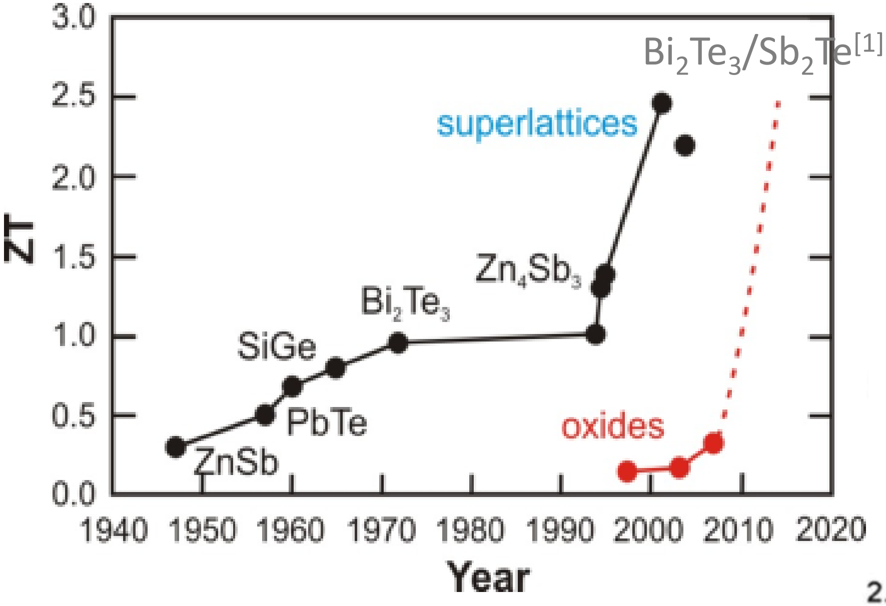
Thermoelectric efficiency

$$\eta = \frac{\Delta T}{T_H} \cdot \frac{\sqrt{1 + ZT} - 1}{\sqrt{1 + ZT} + \frac{T_C}{T_H}}$$

Seebeck effect



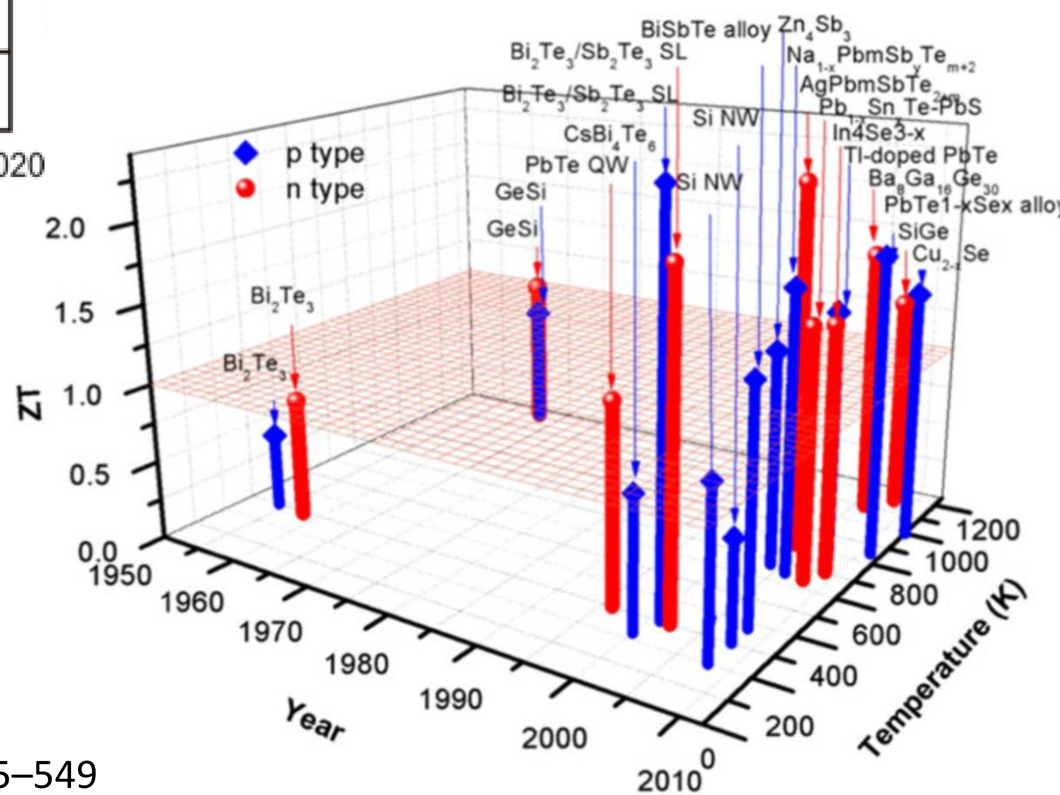
Thermoelectric efficiency has improved steadily, but not enough



To match a refrigerator need ZT= 4

Efficiently recover waste heat from car want $ZT > 2$

Why so difficult to reach zT > 2?



[1] Venkatasubramanian et al. *Nature* 413, 597

[2] Adv. Mater. 2007, 19, 1043, 1053

[3] PNS: Materials International 2012;22(6):535–549

Thermoelectrics have correlated properties

$$ZT = \frac{\alpha^2 \sigma T}{\kappa} = \frac{\alpha^2 \sigma T}{\kappa_E + \kappa_L}$$

$$\alpha = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left(\frac{\pi}{3n}\right)^{2/3}$$

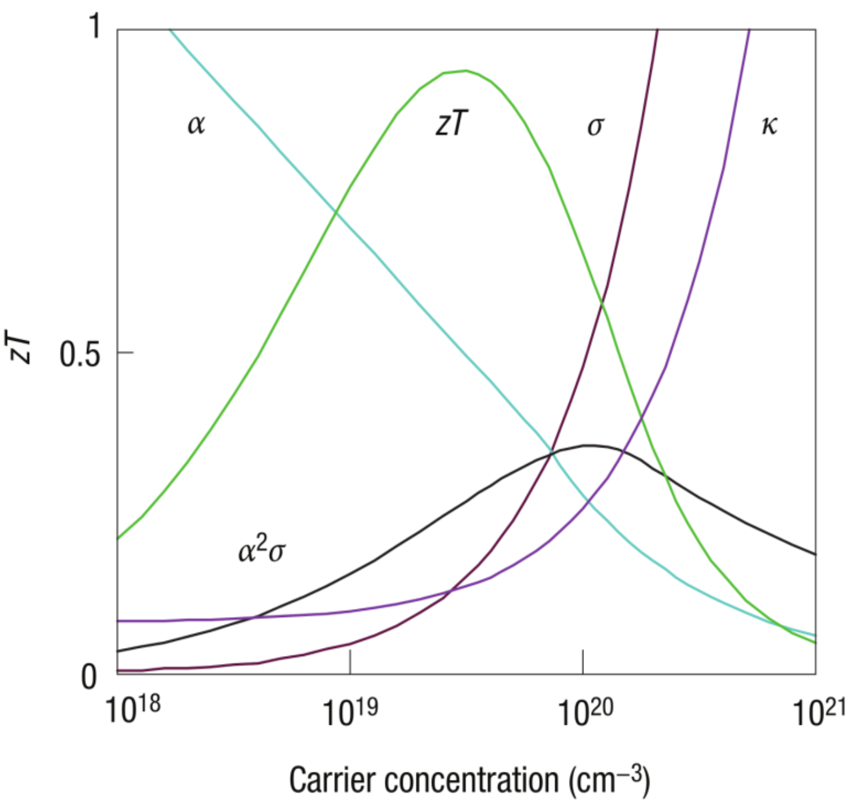
charge carriers

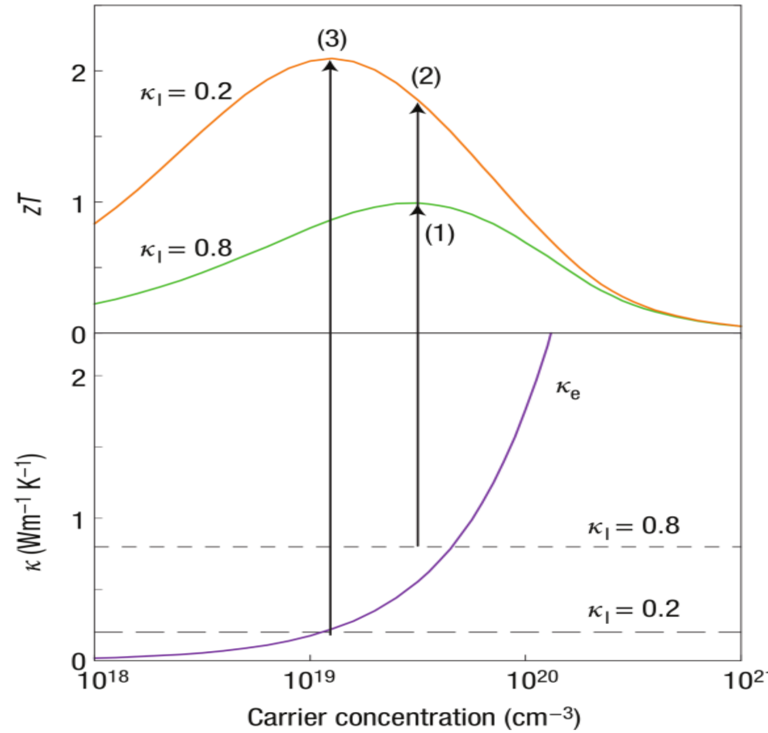
mobility

$$1/\rho = \sigma = ne\mu$$

Wiedemann–Franz law

$$\kappa_E = L\sigma T = ne\mu LT$$



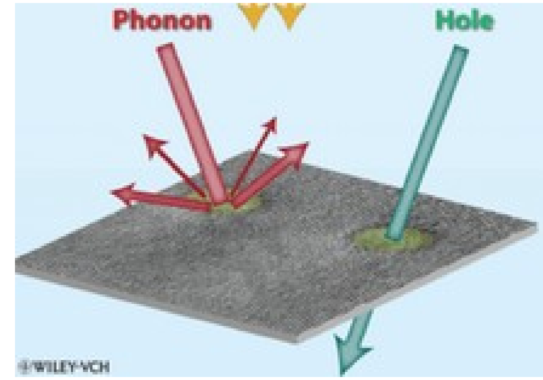


G. Jeffrey Snyder* and Eric S. Toberer Nature. Mater. 7, 2008

Phonon glass – electron crystal

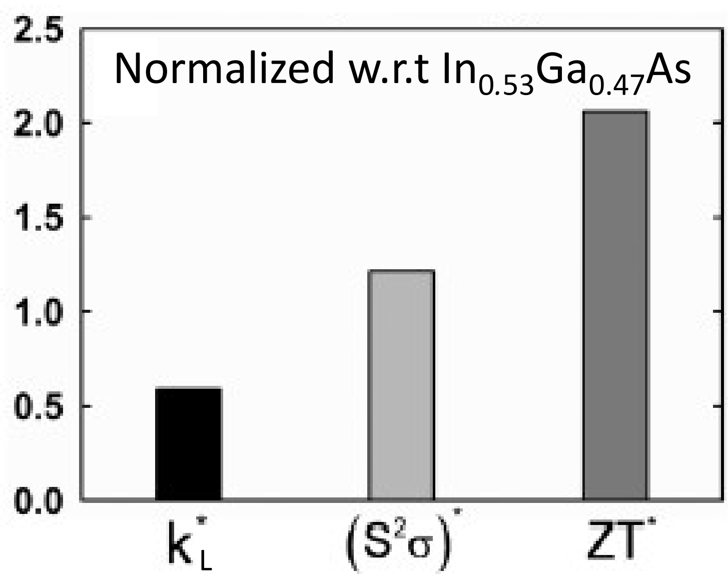
$$ZT = \frac{\alpha^2 \sigma T}{\kappa_E + \kappa_L}$$

Minimize lattice conductivity but do not disturb electronic transport



Make use of embedded nanostructures or random alloys (e.g. Si_xGe_{1-x})!

Example: $In_{0.53}Ga_{0.47}As$ containing ErAs nanoparticles



- κ_L reduced by 2 below alloy limit
- ZT enhanced by 2
- Power unchanged

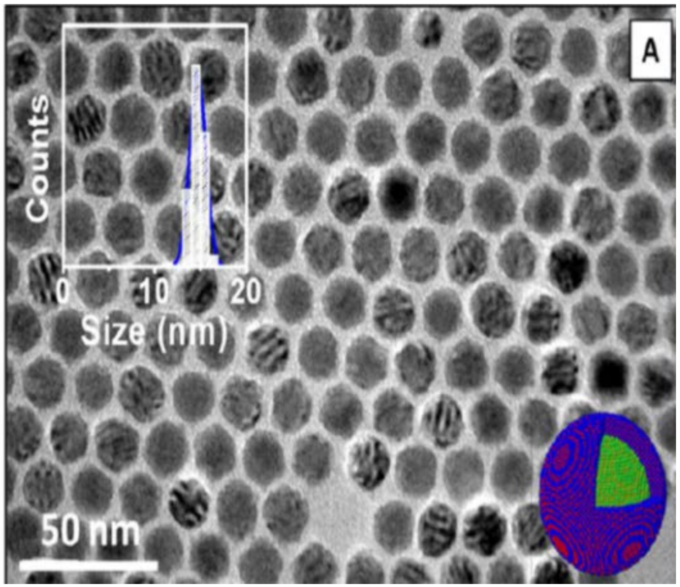
ErAs NP (1-4 nm) scatter mid to long λ phonons

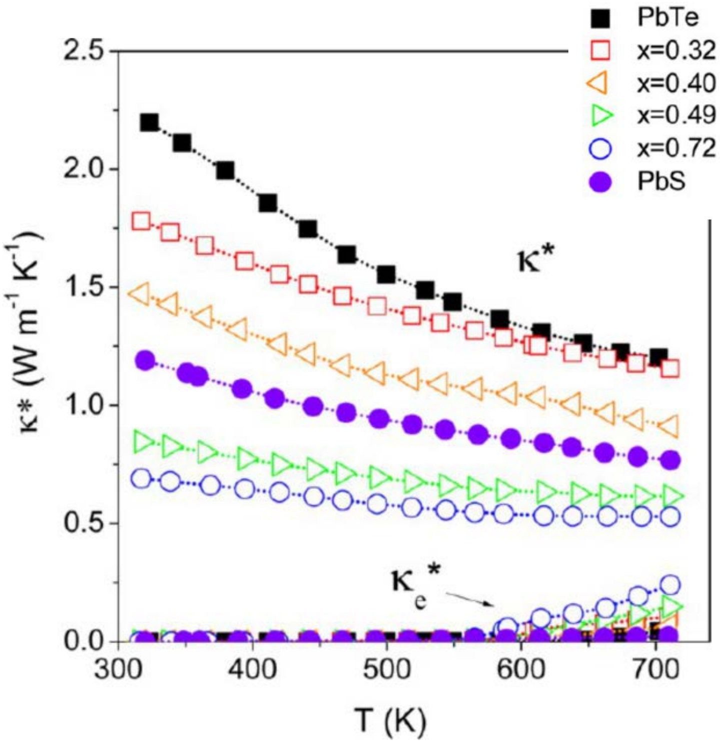
Atomic scale (1Å) defect scatter short λ phonons

W. Kim et. al. PRL, 96, 2006

Mismatched phases in nanoparticles can further scatter phonons

PbTe@PbS coreshell nanoparticles

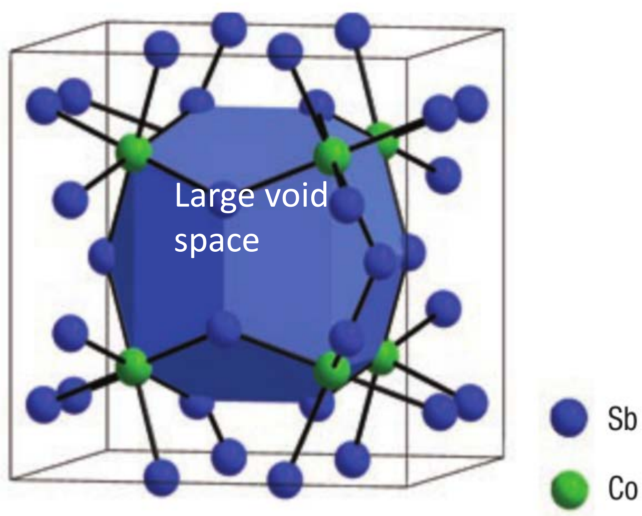


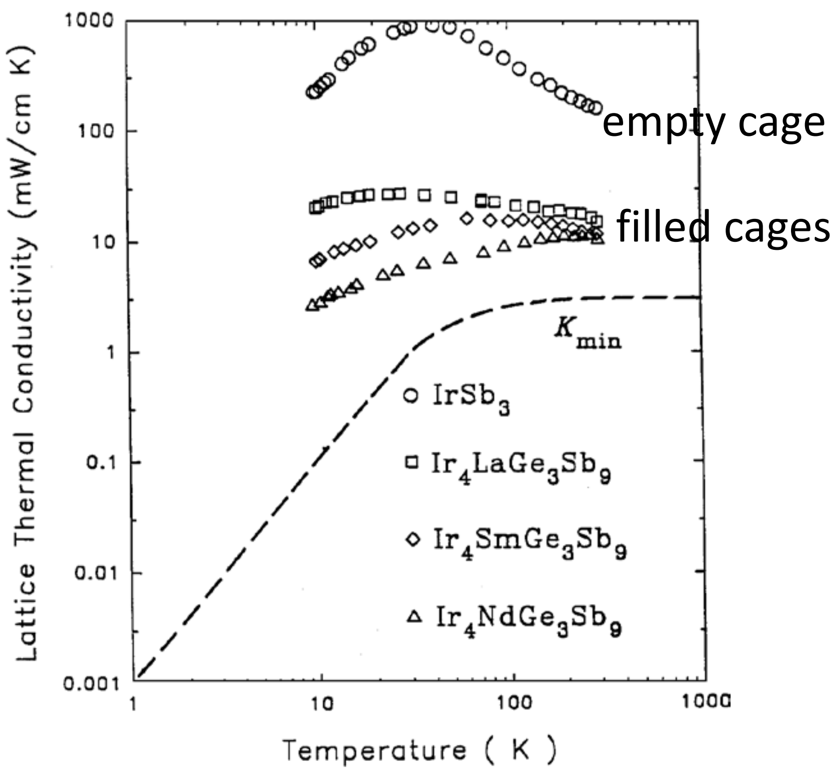


Ibanez, ACS NANO, 7, 3, 2573, 2013

Filling cages scatters phonons due to rattling or local strain

Skutterudite





Annu. Rev. Mater. Sci. 1999. 29,89

Nucleation/spinodal decomposition used as mechanisms to create nanostructured alloys

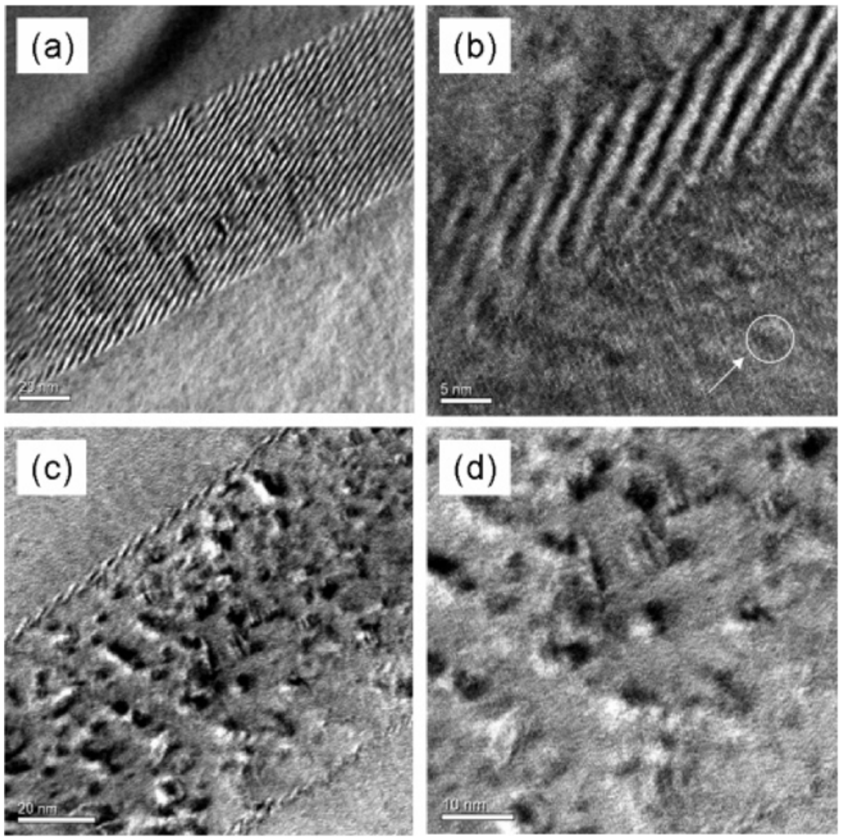
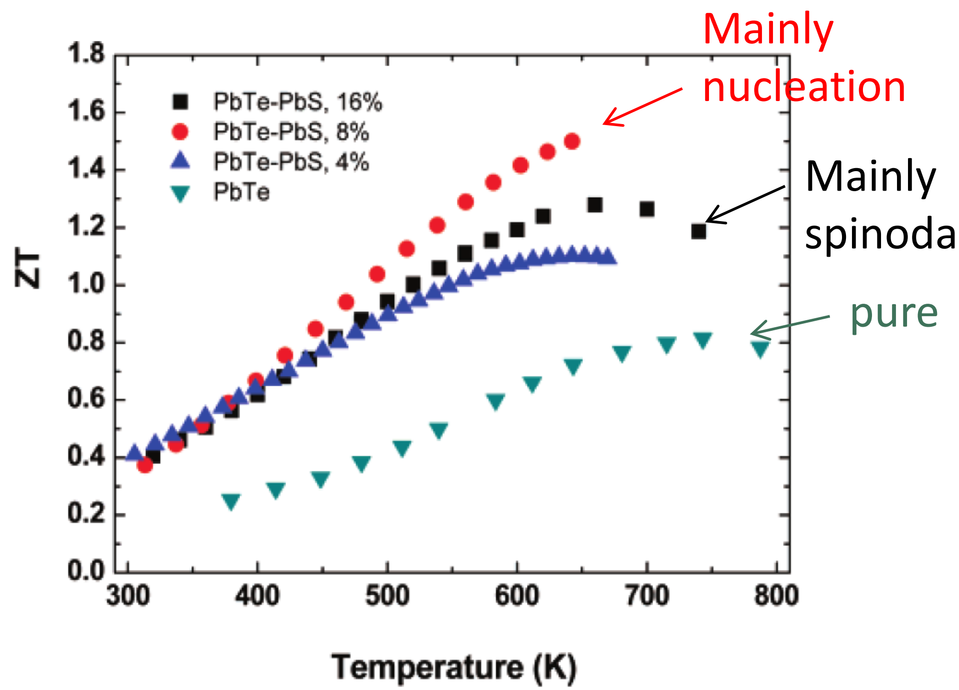


Figure 3. Characteristic HRTEM pictures of the investigated sample of PbS 16%. All pictures show parts of the same patch. (a) Spinodal decomposition of PbTe into PbS. The parallel lines correspond to periodical compositional fluctuations of PbS (bright) and PbTe (dark) phases. (b) Depending on the local S concentration, parts of the patch exit the spinodal region of the phase diagram and the characteristic compositional fluctuation fades away. (c and d) Outside the spinodal region the PbTe−PbS mixture is still unstable, but the two phases separate by nucleation and growth; thus, embedded nanoparticles of arbitrary shape appear.



J. AM. CHEM. SOC. ■ VOL. 129, NO. 31, 2007

PHYSICAL REVIEW B VOLUME 47, NUMBER 19 15 MAY 1993-I

Effect of quantum-well structures on the thermoelectric figure of merit

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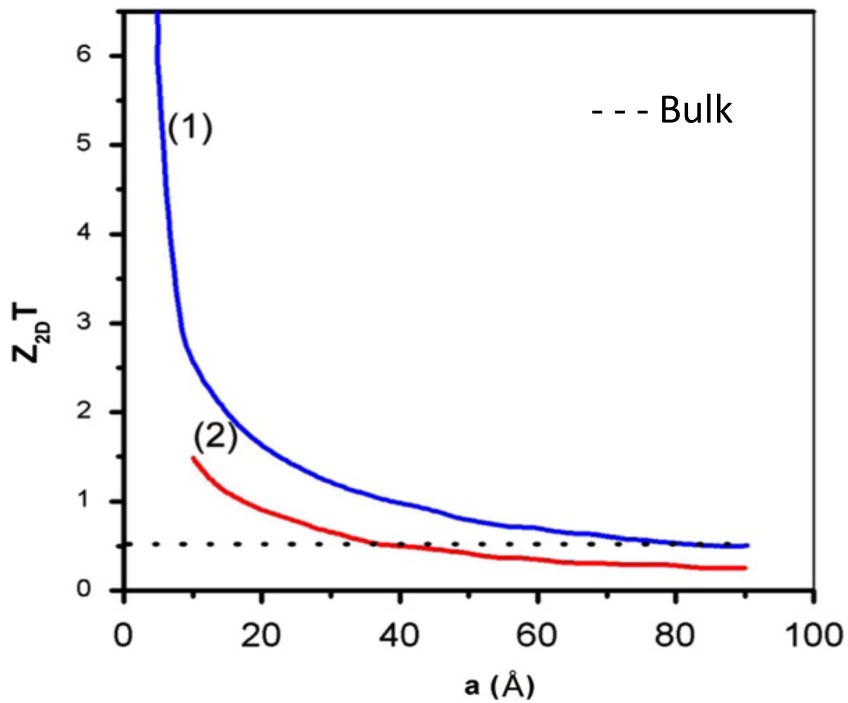
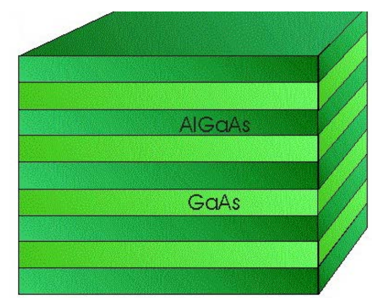
M. S. Dresselhaus

Predicted between 3-15 ZT enhancement for Bi_2Te_3 superlattices

thin film super lattices





A Superlattice Structure

orientation of superlattice is crucial

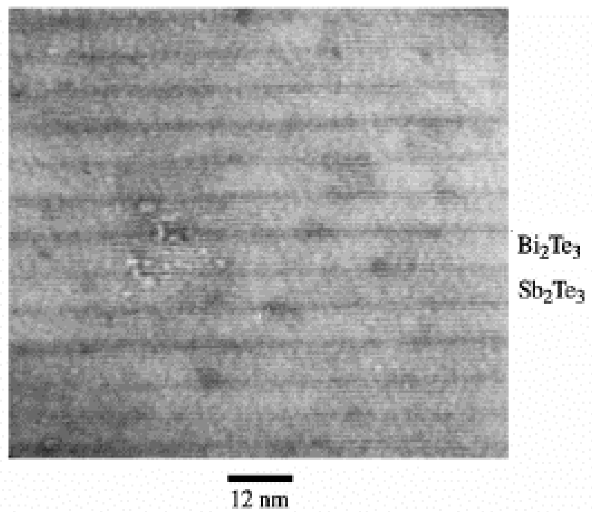
Calculated ZT as a function of layer thickness a in a quantum well structure for layers parallel to the a–b plane (1) and b–c plane (2)

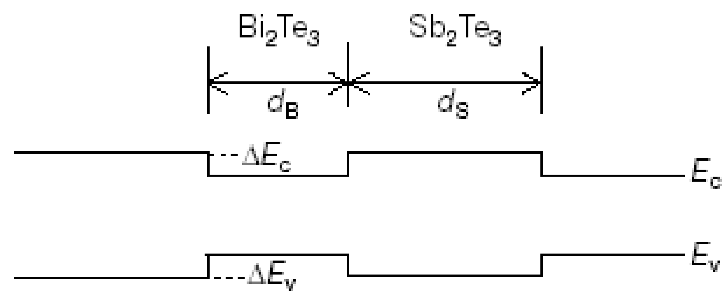
Thin-film thermoelectric devices with high room-temperature figures of merit

Rama Venkatasubramanian, Edward Siivola, Thomas Colpitts & Brooks O'Quinn

8 years later, thin-film with zT = 2.4!

Bi_2Te_3/Sb_2Te[1]





"phonon band gaps"

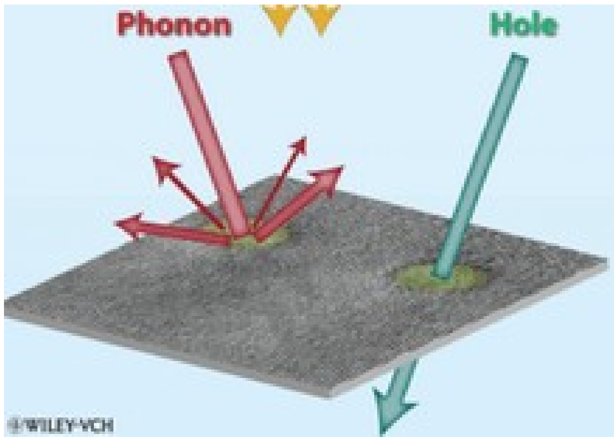
Not too thin layers since tunneling can occur!

- When all the phonons have a mean free path equal to the lattice spacing κ_{min} is expected!
- Frequent phonon-boundary scattering: low κ
- High density of states near E_F: high $S^2\sigma$

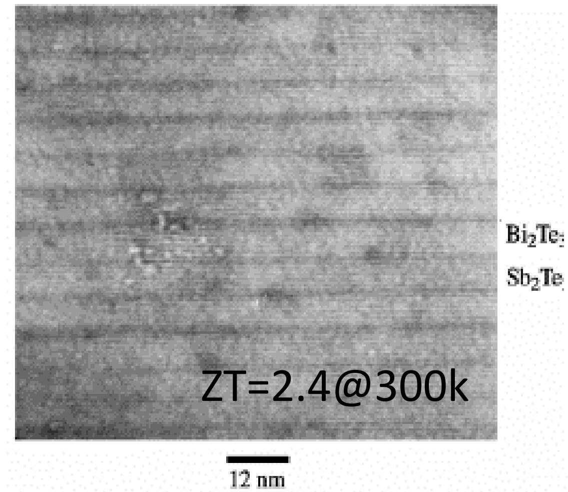
Nature **413**, 597-602 (11 October 2001)

Nanostructuring has greatly enhanced thermoelectrics ZT

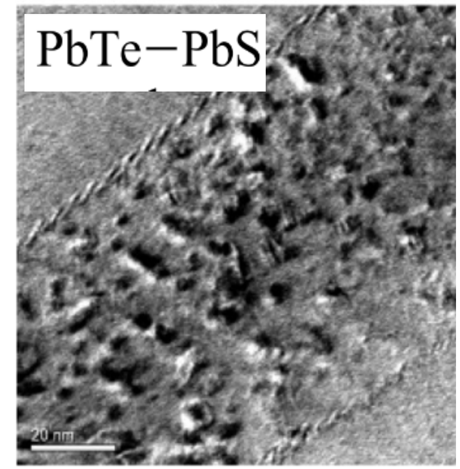
Phonon glass-electron crystal

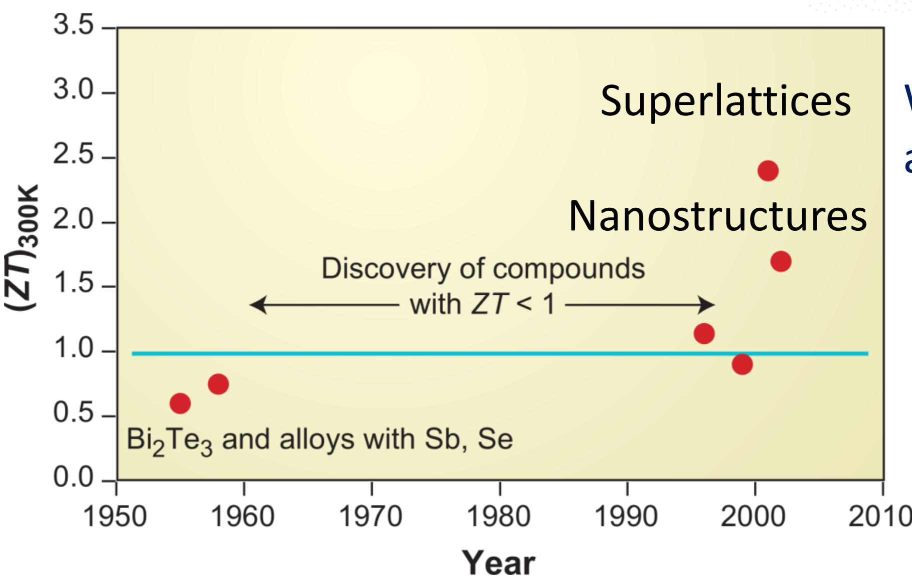


Thin film super lattice



Nucleation of grains





We need to further control nanoscale, mesoscale, and macroscale structures to further improve ZT

Improved device processing is needed to lower cost of high ZT lab created devices

Science **303**, 777 (2004);

Acknowledgements

Dr. Christian Carbogno
Prof. Matthias Scheffler
Prof. Baron Peters
Peters group

UC Santa Barbara

Fritz-Haber Institute