Thermoelectricity: From Atoms to Systems

Week 5: Recent Advances in Materials and Physics

Lecture 5.2: Semiconductors with embedded nanoparticles

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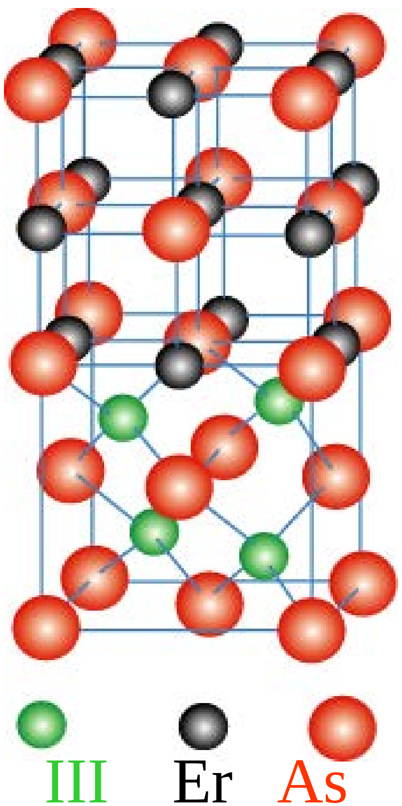
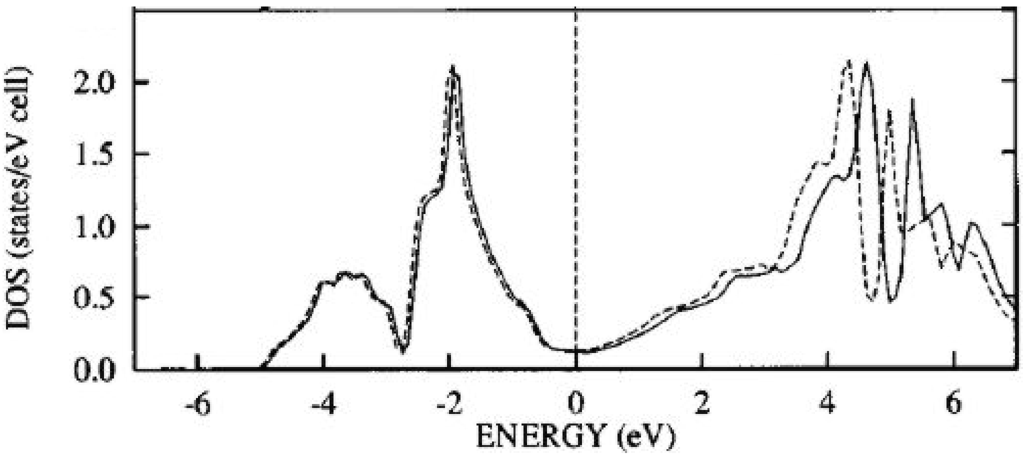
Semimetallic nanoparticles: ErAs/III-V



Semimetal
ErAs
(Rock-salt)
a=5.74 Å

Semiconductor
e.g. InGaAlAs
(Zinc blende)
a=5.85 Å

- Arsenic has a continuous fcc sublattice.
- ErAs is a Rock-salt semi metal, ~ lattice matched to InGaAlAs

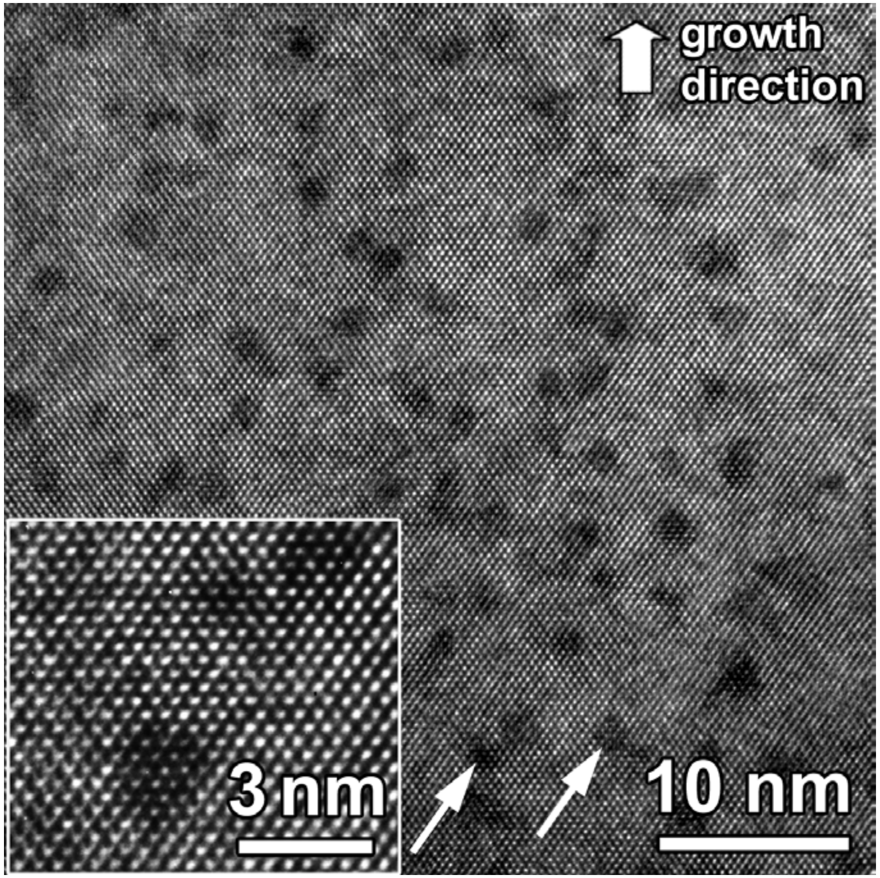


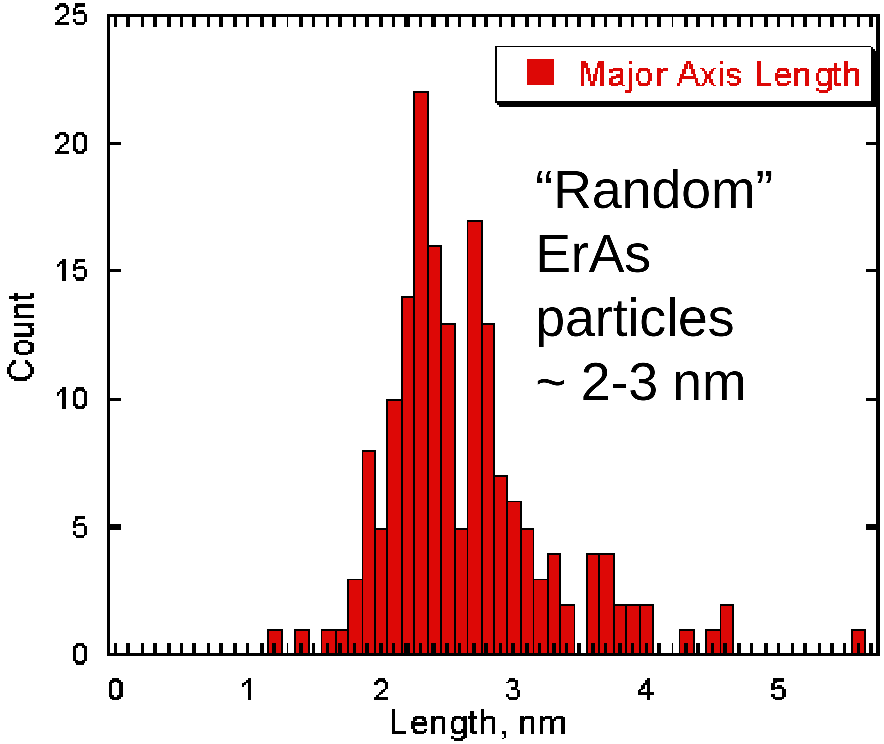
A.G. Petukhov, et. al. *Phys. Rev. B*. **53**(8), 1996.

J. Zide and A. Gossard (UCSB)

ErAs Semi-metal Nanoparticles imbedded in InGaAs Semiconductor Matrix

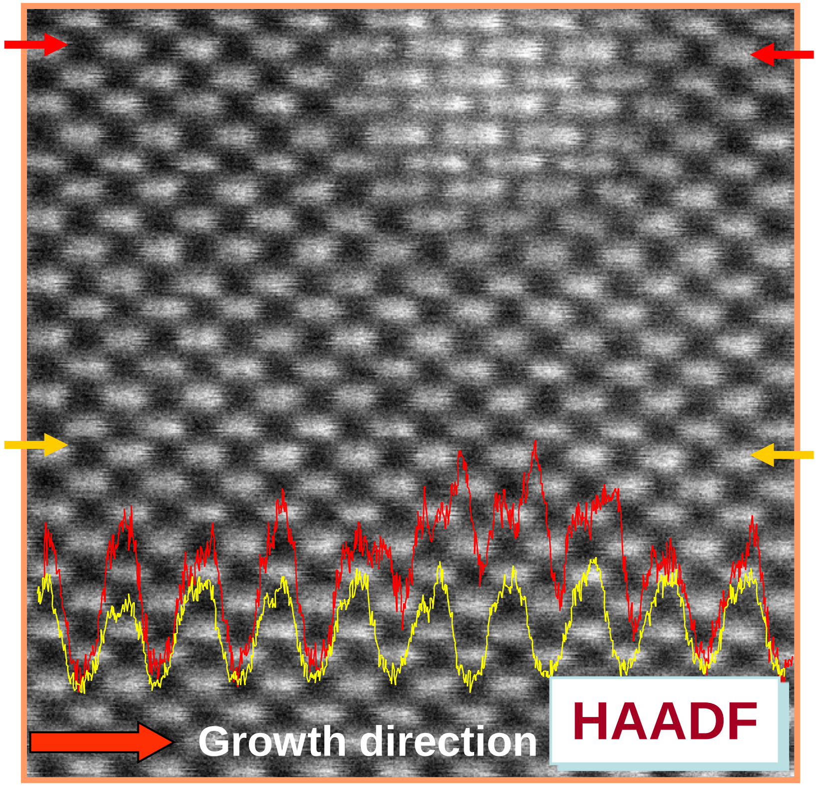
- Erbium is co-deposited at a growth rate which is a fixed fraction of the InGaAs growth rate (MBE growth, 60 microns thick films)
- Solubility limit is exceeded → nanoparticles are formed (2-3nm)

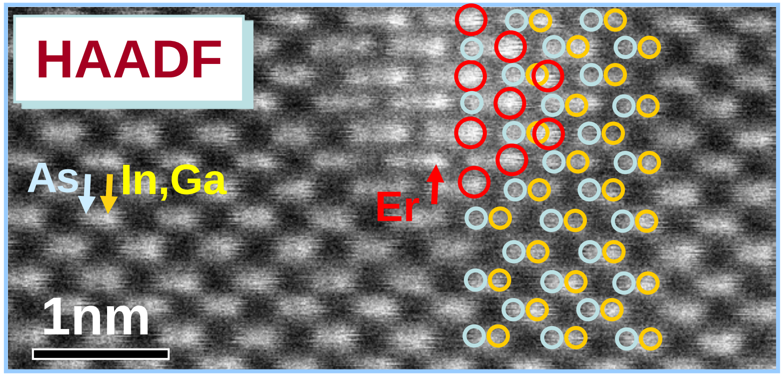


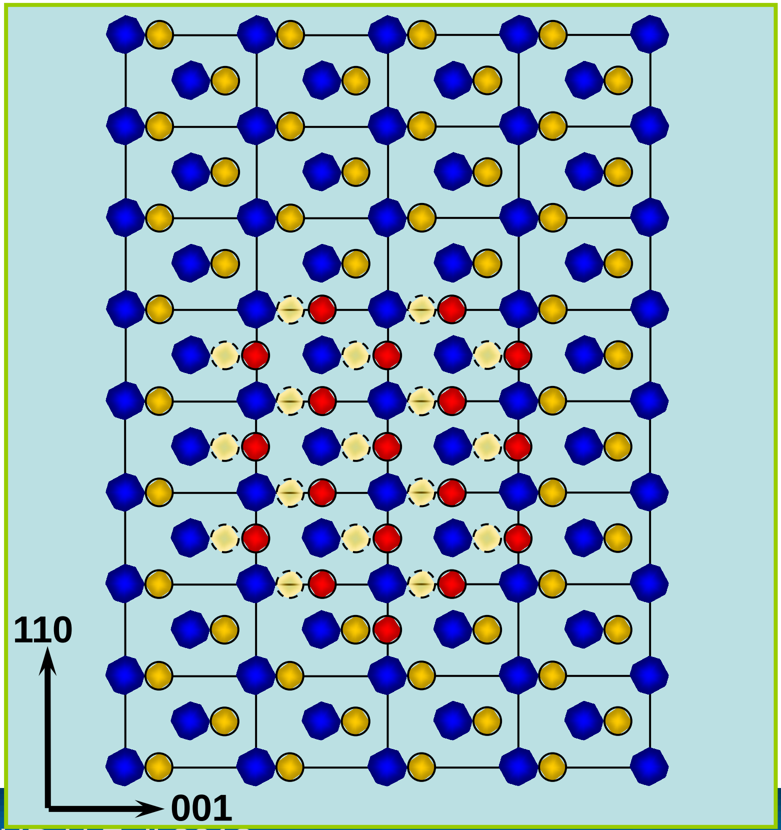


Josh Zide, Hong Lu, Art Gossard, Chris Palmström, Susan Stemmer, John Bowers **UCSB**

HAADF/STEM of ErAs Nanoparticles



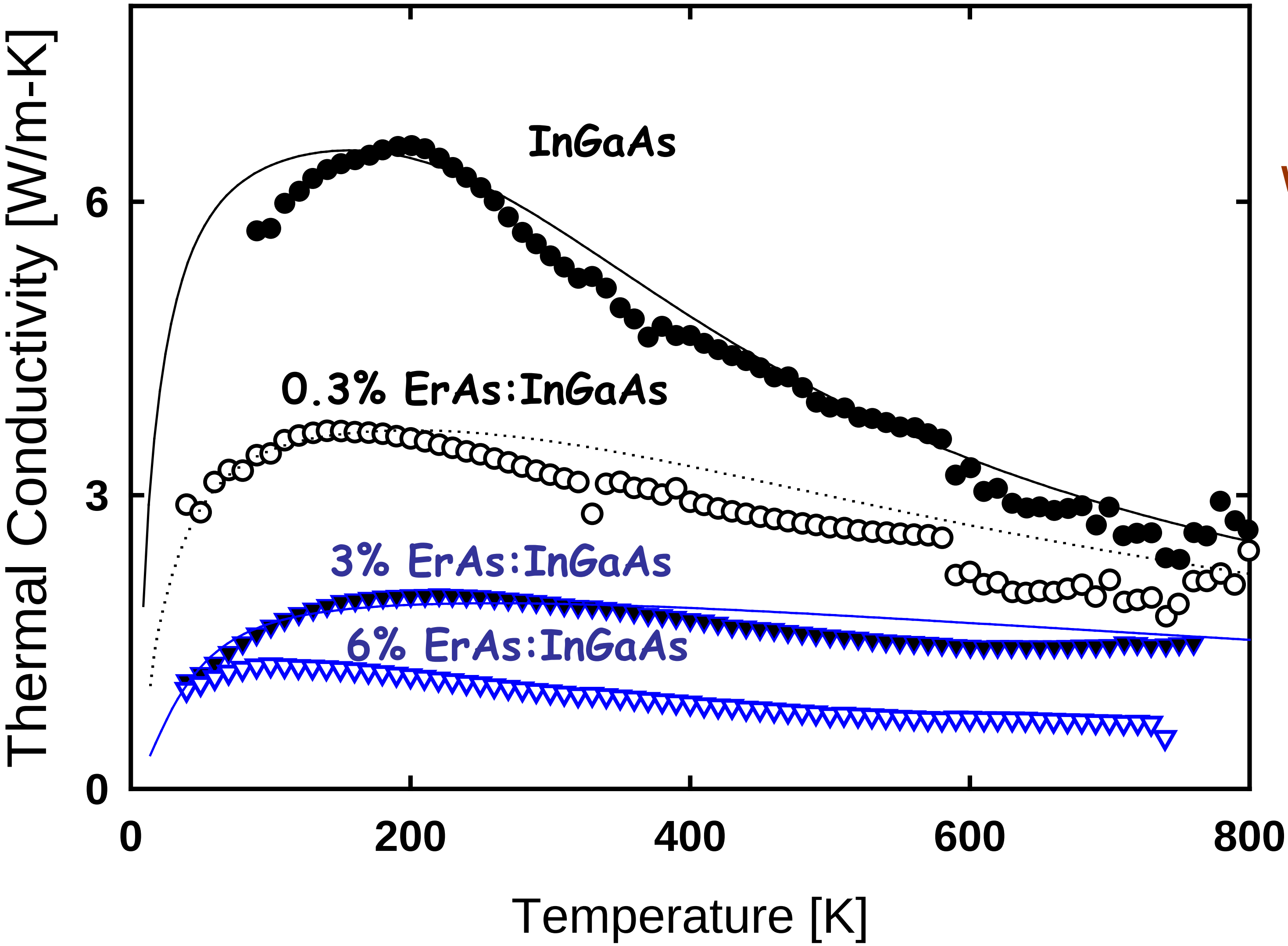




STEM: ErAs particles have the rock salt structure. The As sublattice is continuous across the interface.

D. O. Klenov, D. C. Driscoll, A. C. Gossard, S. Stemmer, Appl. Phys. Lett. 86, 111912 (2005)

Beating the Alloy Limit in Thermal Conductivity



W. Kim, J. Zide et al. Physical Review Letters 2006

Modeling of thermal conductivity

$$k = \frac{1}{3}\int_0^{\omega_{\max}} \underbrace{C_v(\omega)}_{\text{heat capacity}} \underbrace{\upsilon(\omega)}_{\text{phonon group velocity}} \underbrace{\ell(\omega)}_{\text{mean free path}} \, d\omega$$

$$\frac{1}{\ell(\omega)} = \underbrace{\frac{1}{\ell_b(\omega)}}_{\text{boundary scattering}} + \underbrace{\frac{1}{\ell_i(\omega)}}_{\text{impurity scattering}} + \underbrace{\frac{1}{\ell_U(\omega)}}_{\text{Umklapp scattering}}$$

Alloy & Nanostructures

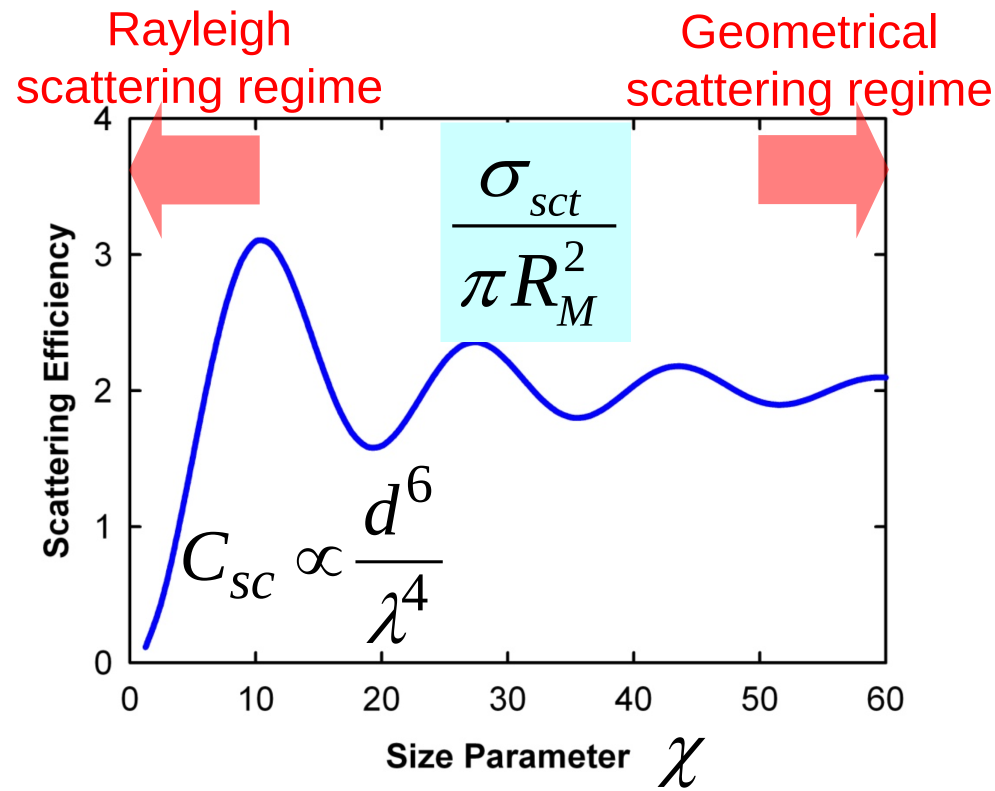
Kim and Majumdar, *Journal of Applied Physics,* **99**, 084306 (2006)

Nanoparticle scattering cross section

$$\ell = \frac{1}{\sigma \eta}$$

- σ : Scattering cross section
- η : Impurity concentration

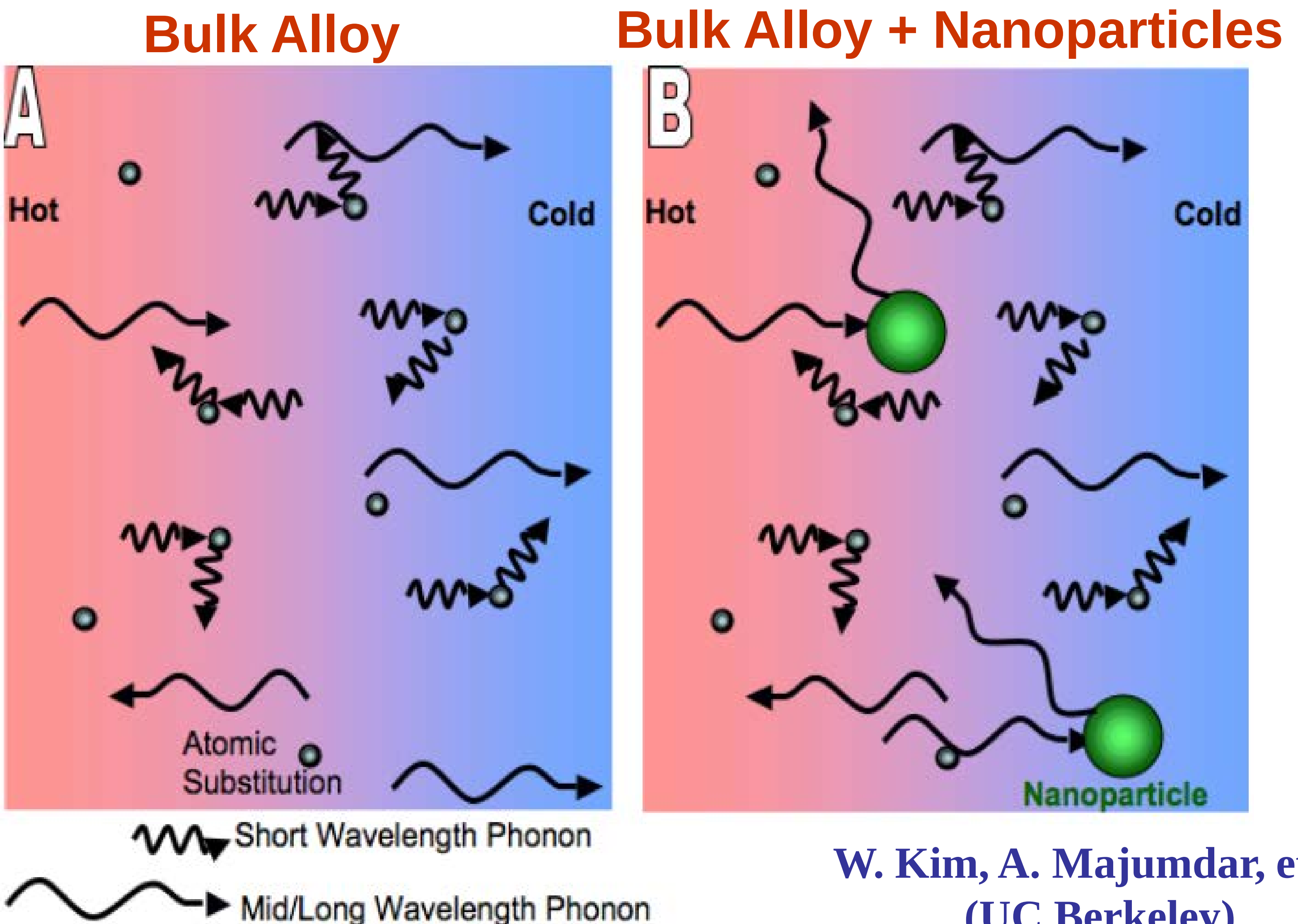
$$\sigma = \int_0^\infty \sigma_{sct} \underbrace{\left(\begin{array}{l} size \\ distribution \\ function \end{array} \right)}_{\substack{\text{mean}(d_M) \\ \text{standard deviation}(\sigma)}} dr$$



- q : Incident wavevector
- R : Size of a scatter
- R_M : Mean radius of the scatter
- qR : Size parameter, χ

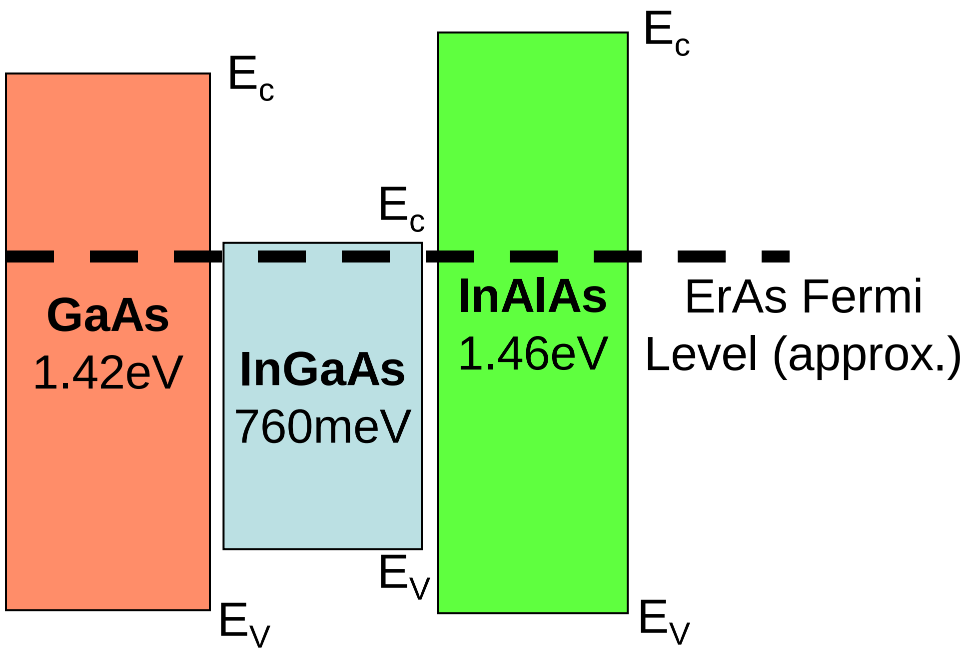
Kim and Majumdar, *Journal of Applied Physics*, **99**, 084306 (2006)

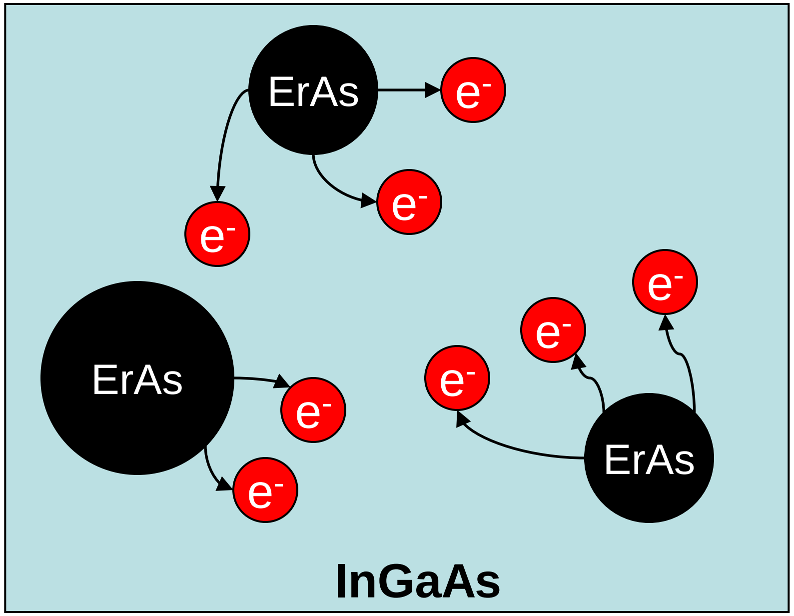
Using Nanoparticles to Reduce Lattice Thermal Conductivity



W. Kim, A. Majumdar, et al. (UC Berkeley)

Electrical Properties of ErAs:InGaAlAs

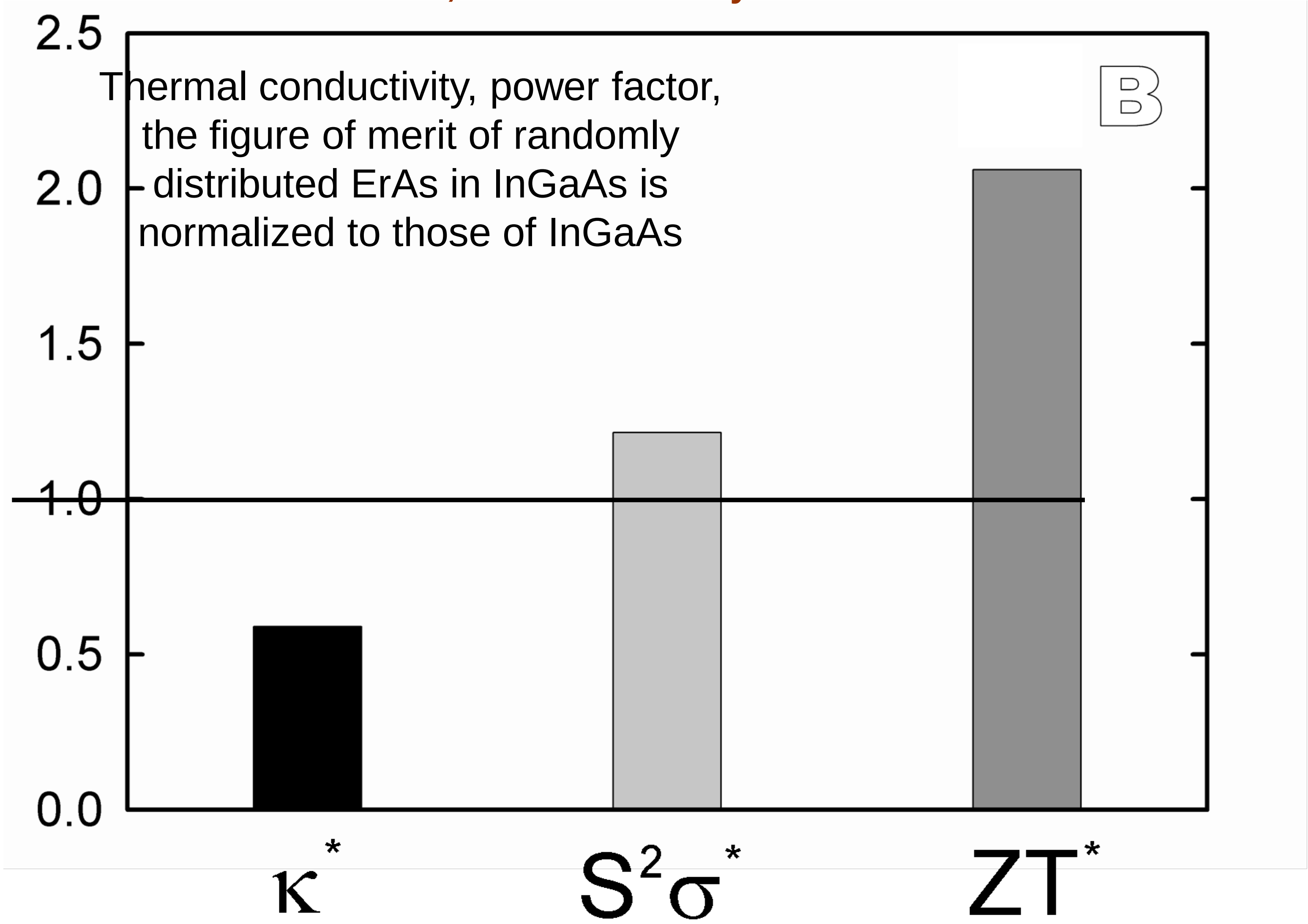




- The Fermi levels of nanocomposites depend on the semiconductor matrix.
  - For example, the Fermi level in ErAs:GaAs is pinned deep within the bandgap.
- ErAs contributes electrons to InGaAs.
- Electron mobilities are high in ErAs:InGaAs.

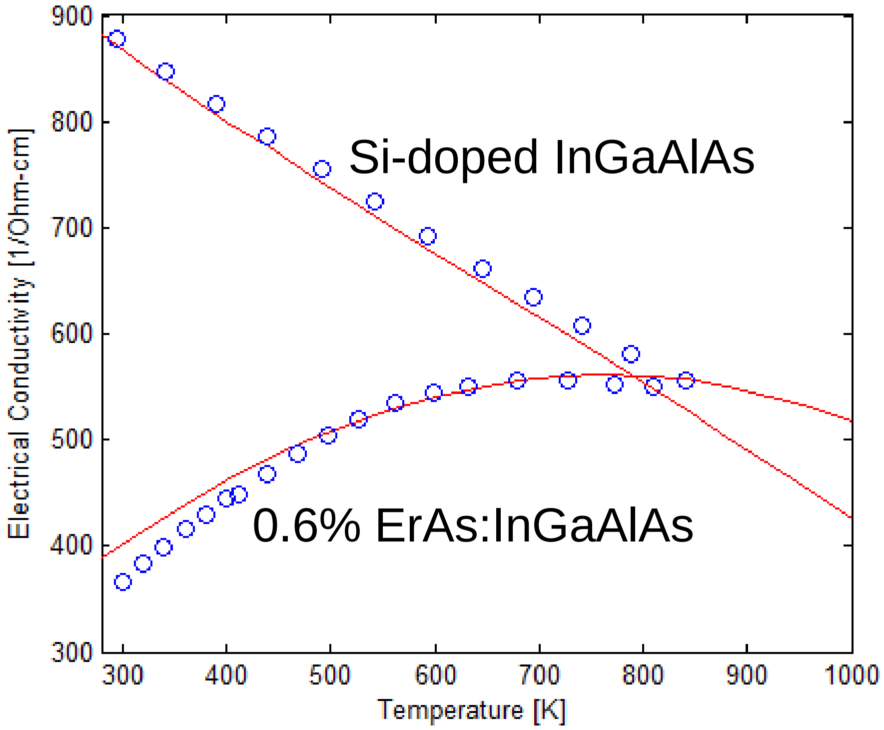
Normalized ZT of 0.3% ErAs: InGaAs (300K)

W. Kim, J. Zide et al. Physical Review Letters 2006

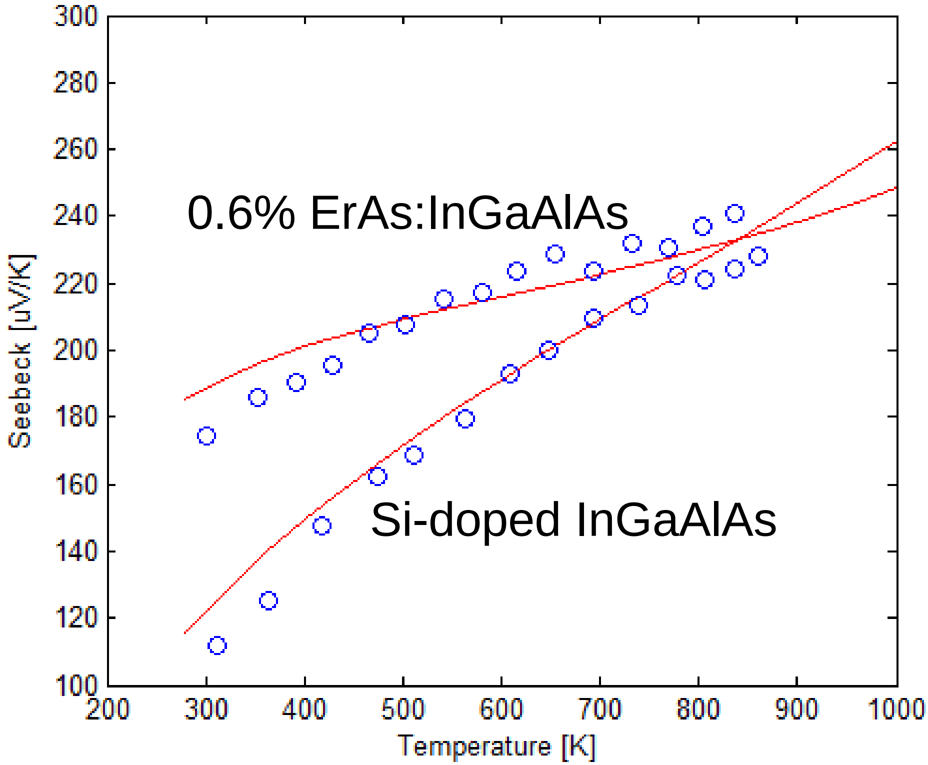


Electrical conductivity and Seebeck (theory/experiment)

Electrical Conductivity



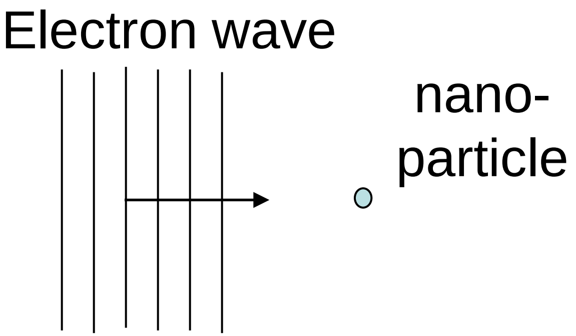
Seebeck



M. Zebarjadi, et al., Appl. Phys. Lett. 2009
J.-H. Bahk et al., Phys. Rev. B 2010 (Si-doped)
J. Zide et al., J. Appl. Phys. 2010

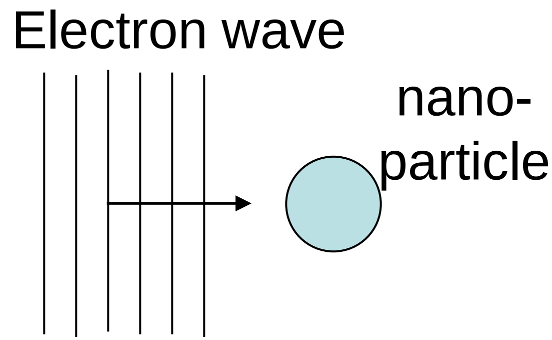
Embedded nanoparticle scattering

Born Approximation



- Perturbation theory
- Point size potentials
- Low barriers

Partial Wave technique

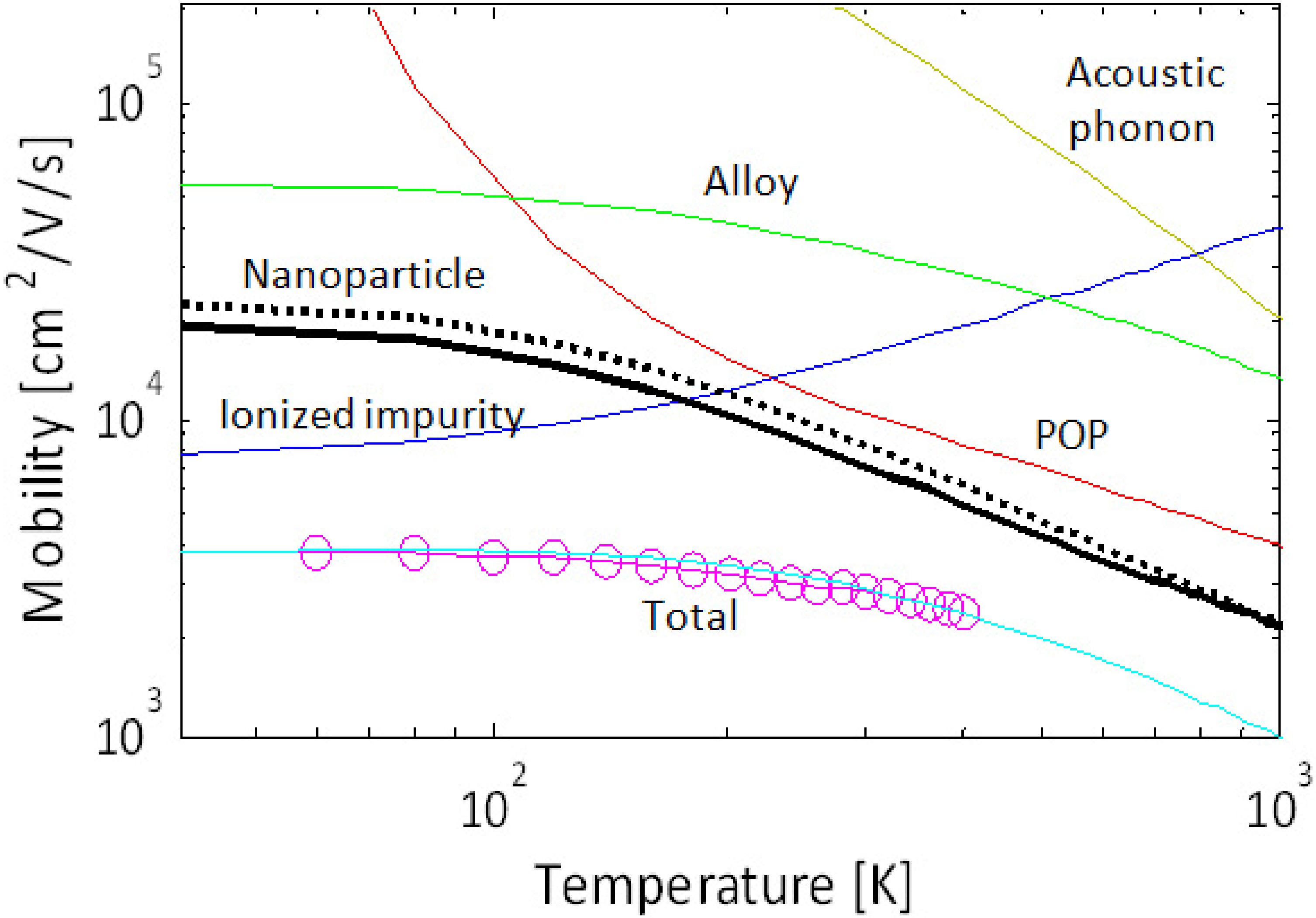


- Exact solution of Schrodinger
- Finite size scattering center
- Harder to implement

Mona Zebarjadi, et al. Appl. Phys. Lett. 94: 202105, 2009

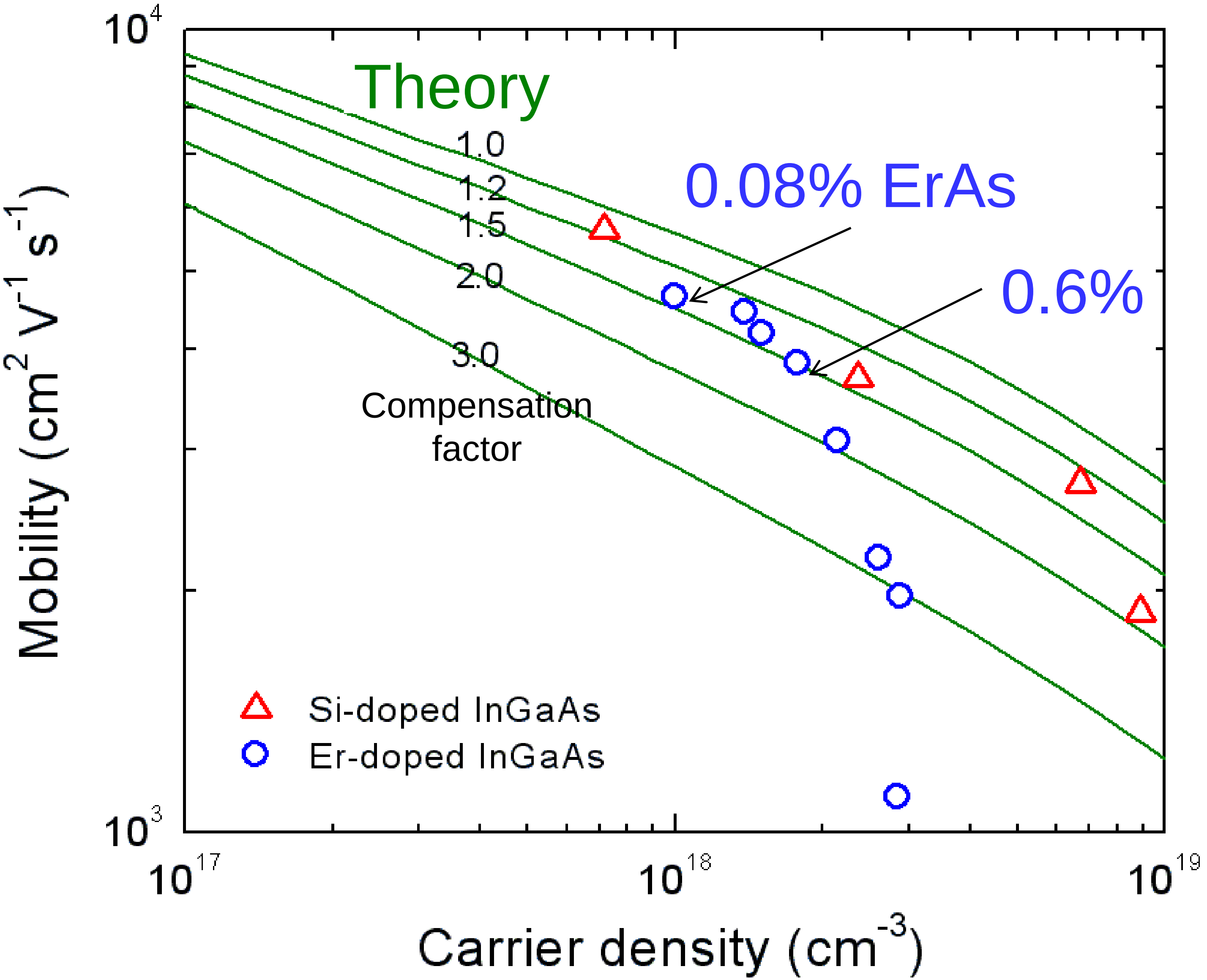
Multiple scatterings => Coherent Potential Approximation
(not described here, M. Zebarjadi, et al. Nano Letters, 11, 225, 2011)

Mobility (Theory vs. Experiment)



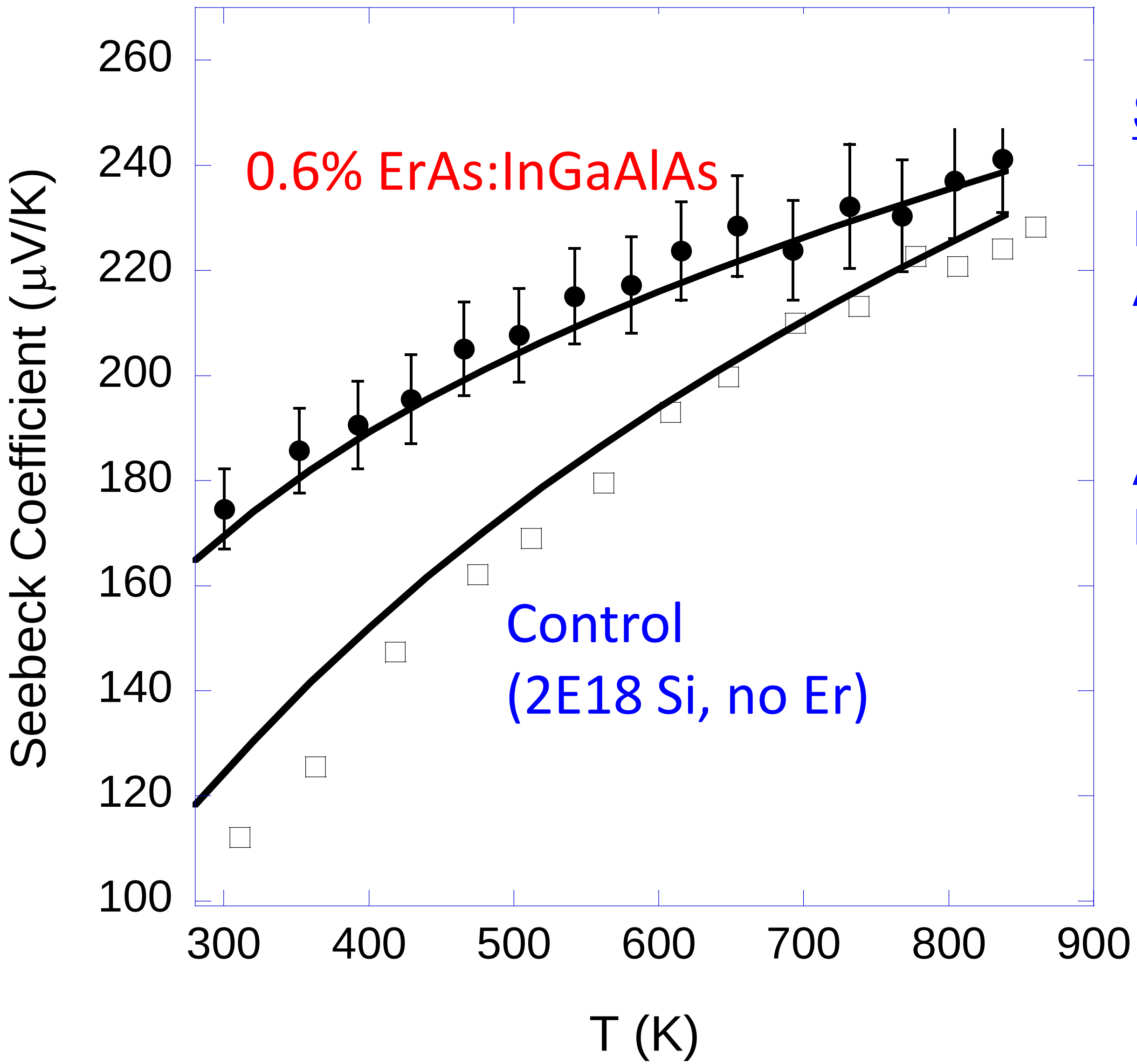
Je-Hyeong Bahk, Mona Zebarjadi et al. (2009)

Electron mobility in embedded nanoparticle material



Je-Hyeong Bahk, Peter Burke, et al. 2013

Seebeck (Theory vs. Experiment)



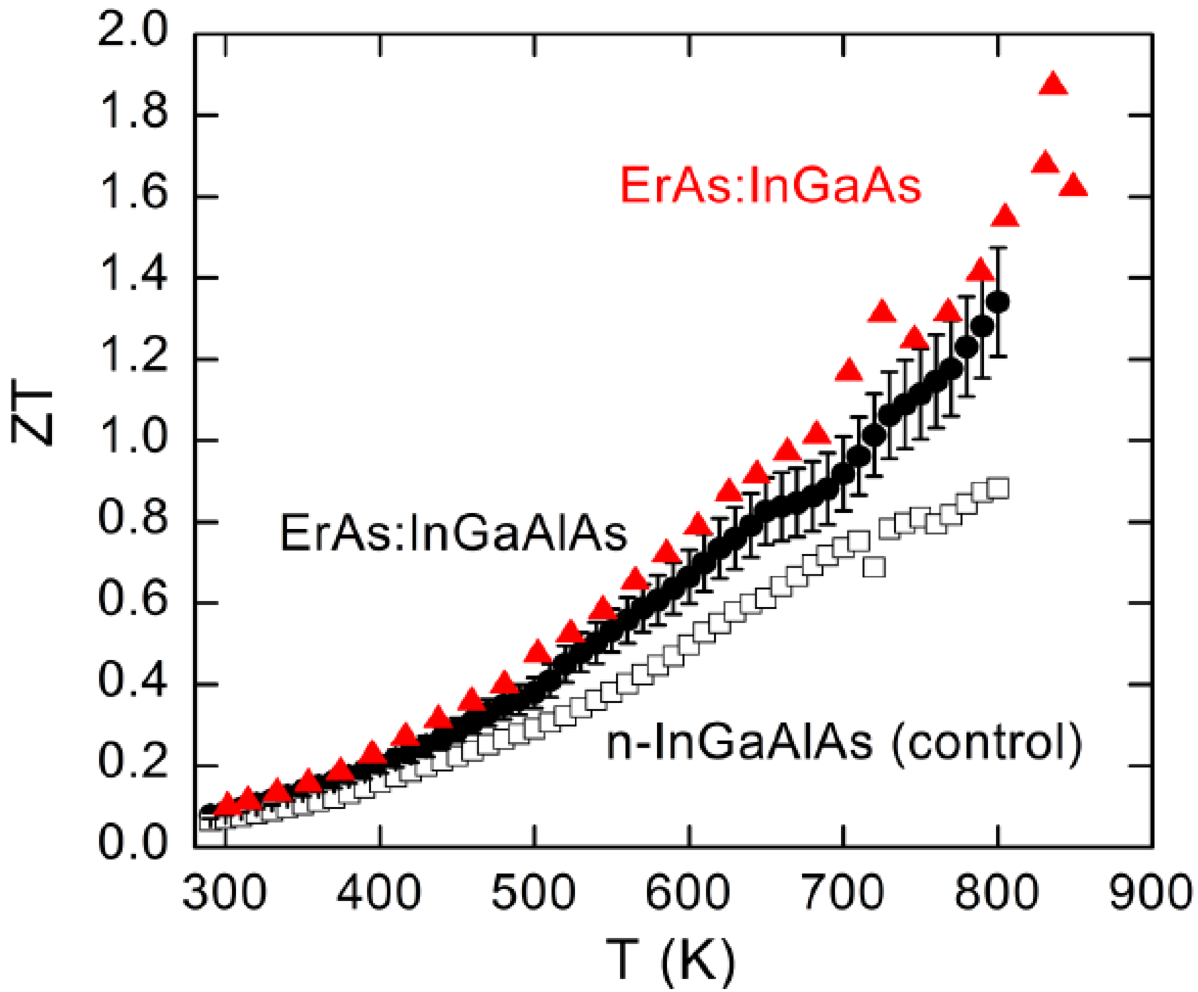
Scattering mechanisms:

Polar optical phonons
Acoustic phonons
Intervalley phonons
Impurity
Alloy scattering
Nano particle scattering:
(Partial Wave technique)

Solid lines are theoretical pr ediction **(no fitting)**

Je-Hyeong Bahk, Mona Zebarjadi et al. (2009)

Thermoelectric figure-of-merit

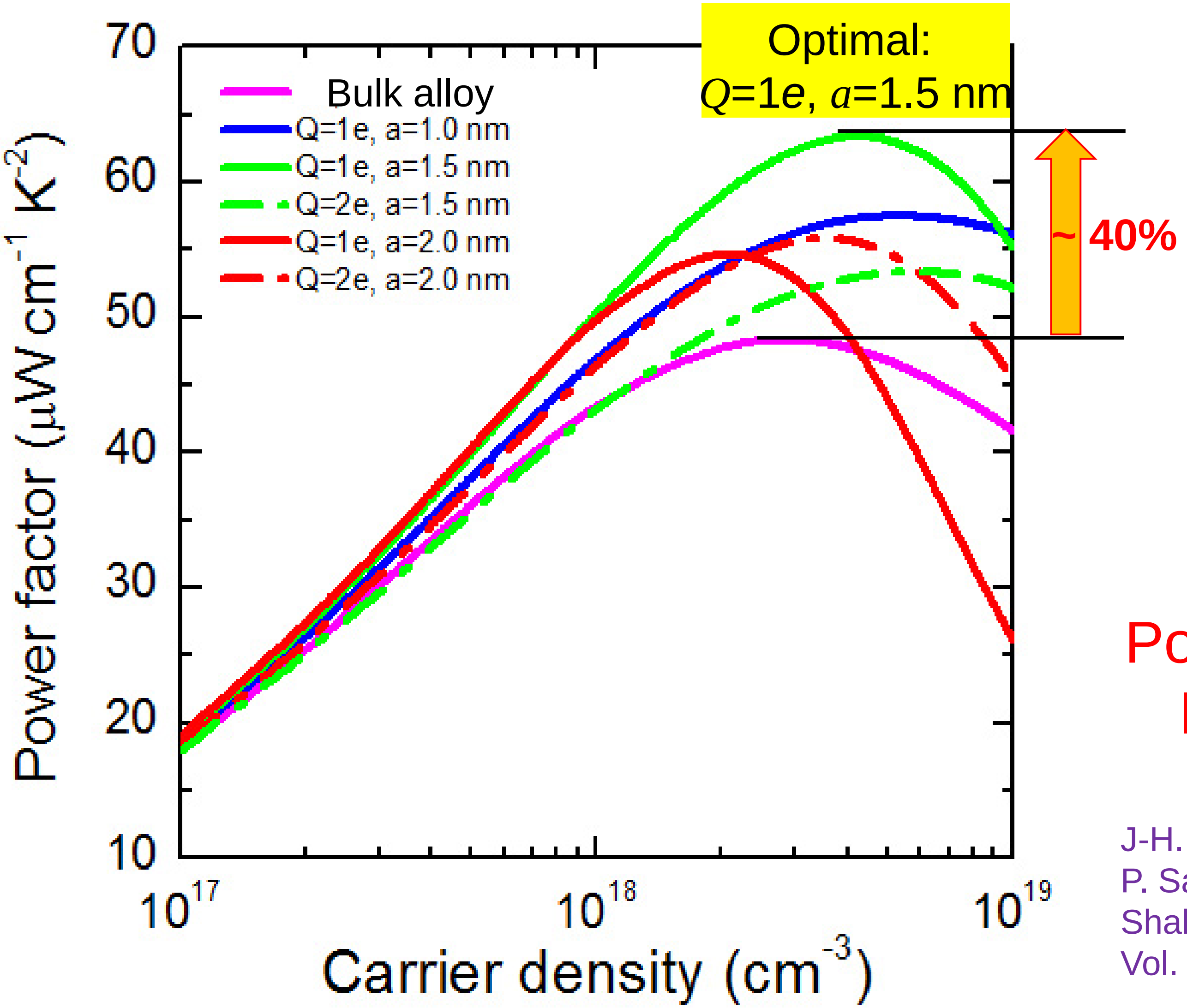


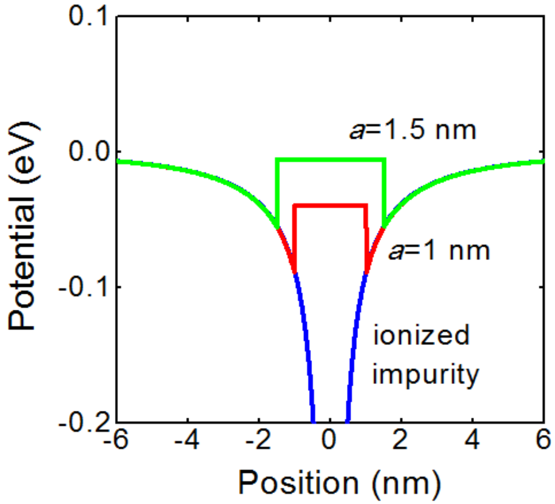
Zide et al. J. of Applied Physics (2010);

Burke, Bahk, et al. (2013)

The majority of ZT enhancement is from thermal conductivity reduction.
5% power factor enhancement at 800K.

Nanoparticle scattering optimization





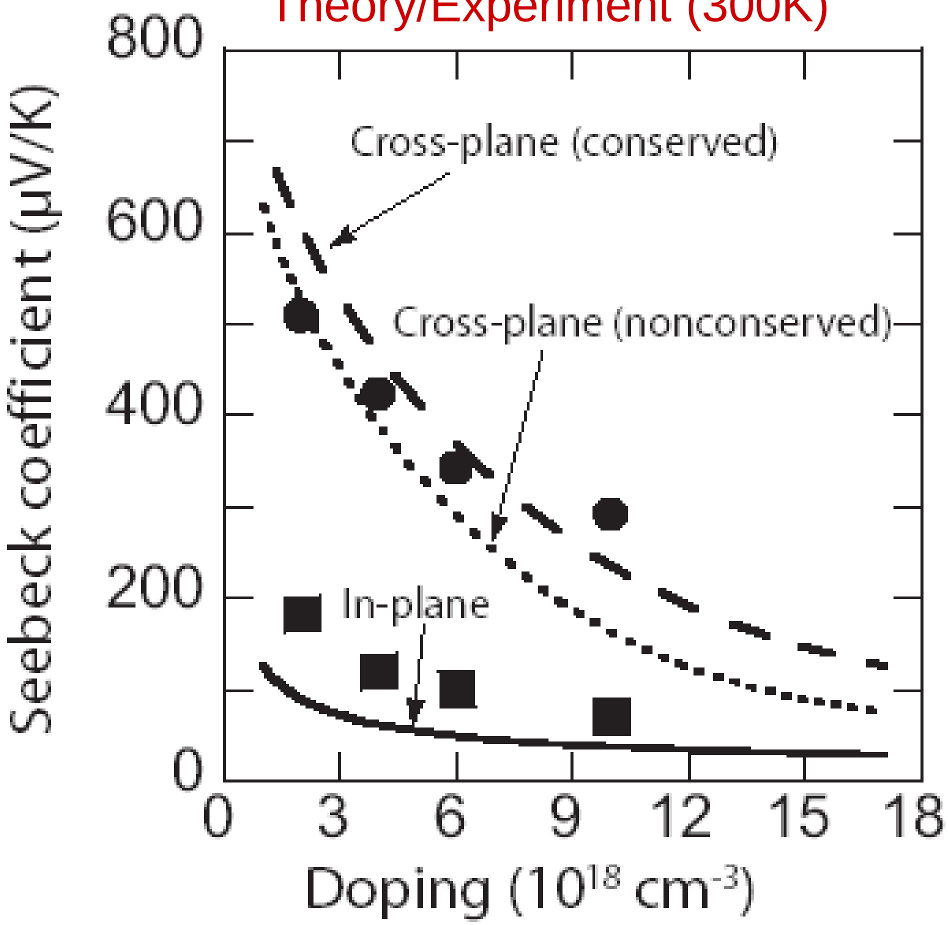
Power factor, n-type InGaAs at 600K

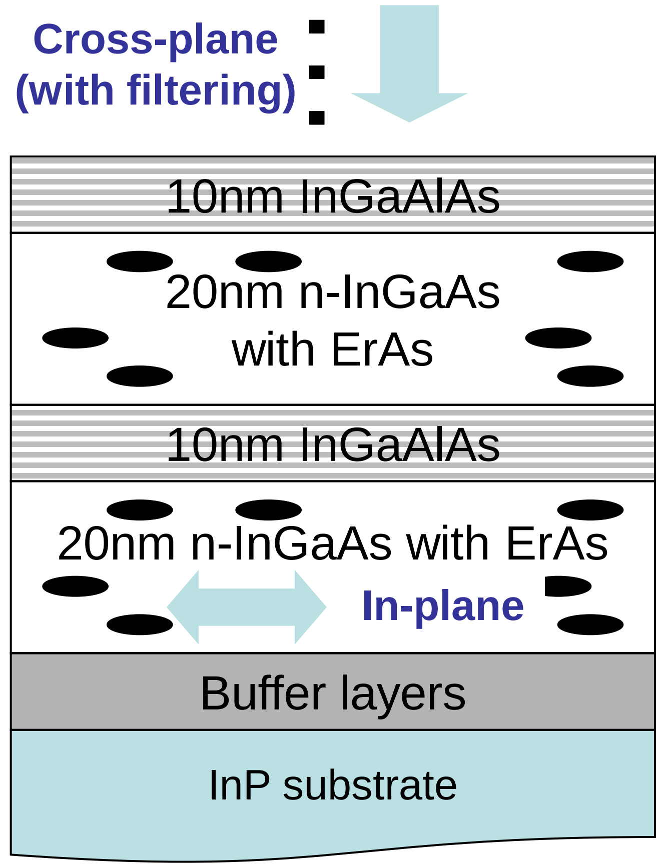
J-H. Bahk, Z. Bian, M. Zebarjadi, P. Santhanam, R. Ram, A. Shakouri, "Applied Physics Letters Vol. 99, Art. No. 072118, 2011.

Cross-plane and in-plane Seebeck in thick barrier superlattices InGaAs:ErAs/InGaAlAs

- Enhance energy filtering by inserting InGa**Al**As barriers inside ErAs:InGaAs can enhance cross-plane Seebeck by a factor of 3

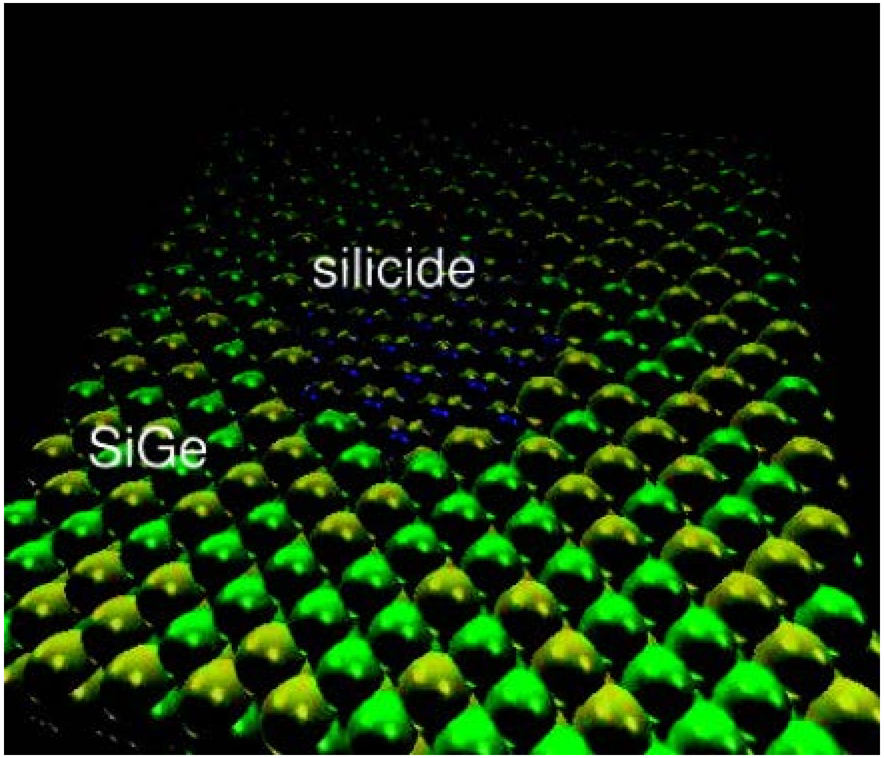
Theory/Experiment (300K)





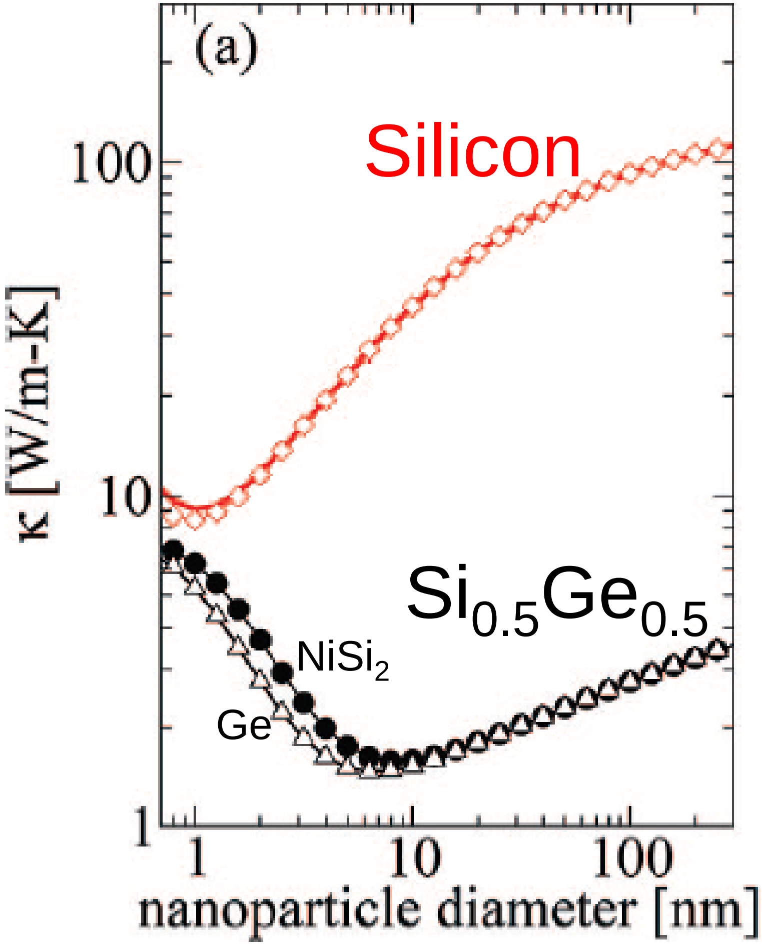
Zide et al, PRB 74, 205335, 2006

Nanoparticle in alloy for thermal conductivity reduction



Natalio Mingo et al. Nano Letters 2009

300K
3.4% nanoparticles



Week 5: Lecture 2 Summary

- Embedded ErAs nanoparticles in InGaAs (endo-epitaxial nanoparticles –see Kanatzidis group)
- Thermal conductivity reduction below alloy limit
  – Long/mid wavelength phonon scattering
- Power factor enhancement
  – improved mobility –modulation doping
  – Energy filtering (?) –scattering time has sharper energy dependence