Multiscale Simulation of Heat Transport and Phonon Dynamics

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Outline

1. Introduction to phonon-mediated thermal transport
2. Thermal Conductivity in Yttria-Stabilized Zirconia
   - Experiment
   - Simulation
3. Molecular-dynamics simulation of phonon dynamics
4. Multiscale simulation of phonon processes
5. *Nanofluids*
6. Conclusions and outlook

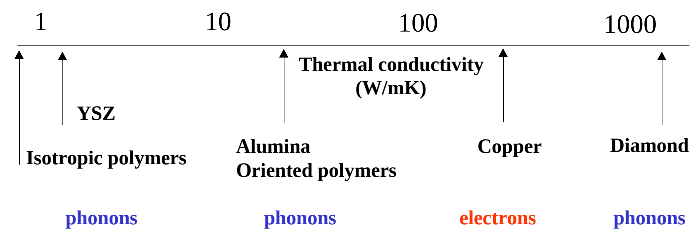
Heat Transfer Mechanisms

Three fundamental mechanisms of heat transfer:

- Convection
- Radiation
- Conduction

- Convection is a mass movement of fluids (liquid or gas) rather than a real heat transfer mechanism (heat transfer is <u>with</u> convection rather than <u>by</u> convection)
- Radiative heat transfer is important at high temperature
- Conduction is heat transfer by molecular or atomic motion

Thermal Conductivity



Electrical conductivity

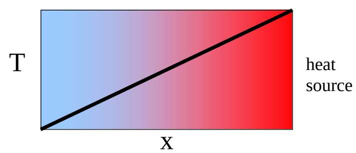
$\sigma(\text{Cu}) \sim 5\ 10^{5}\ (\Omega\ \text{cm})^{-1}$

$\sigma(\text{diamond}) \sim 10^{-10}\ (\Omega\ \text{cm})^{-1}$

Thermal conductivity

Fourier's Law

$J = -\kappa \, dT/dx$



$J = \int J(\omega) \, d\omega$

Phonon-mediated

$J(\omega) = h\omega \, v(\omega) \, D(\omega) \, n(\omega)$

energy

velocity

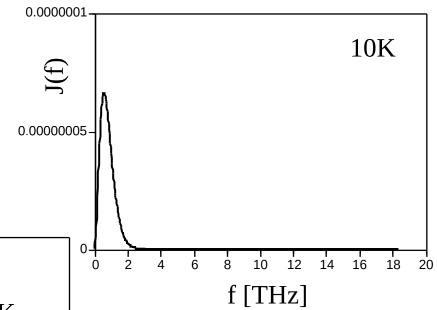
Density of states

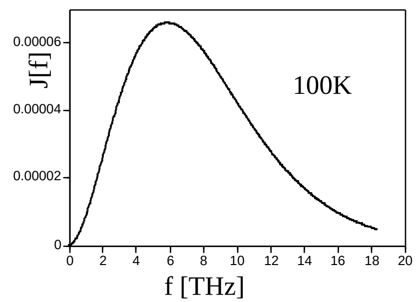
Bose-Einstein distribution

Phonons and thermal conductivity

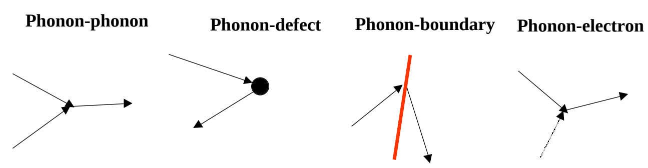
Dominant phonon

$f_D = 2.82 \ kT/h$



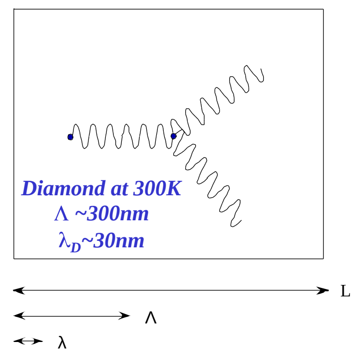


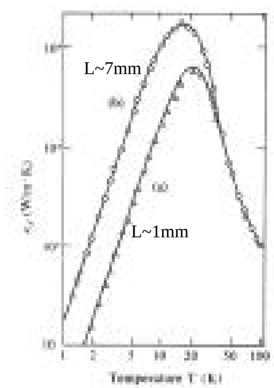
Phonon Scattering Mechanisms



Macroscale ***** *** *

Phonon Length Scales



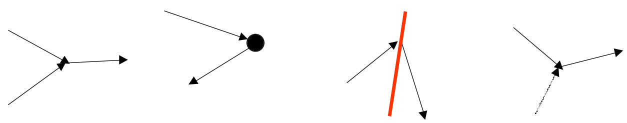


Sample-size dependent thermal conductivity of single crystal LiF

- Usually l < Λ < L
- New physics arises when L is reduced to nanometers

from J.S. Blakemore, Solid State Physics, p. 138 (1989)

Phonon Scattering Mechanisms



	Phonon-phonon	Phonon-defect	Phonon-boundary	Phonon-electron
Macroscale	*****	***	*	
Nanoscale	**	***	*****	

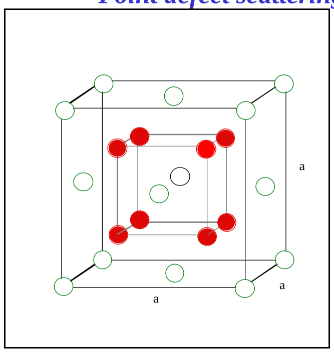
Thermal Conductivity in Yttria-Stabilized Zirconia

Simulation: Effect of stoichiometry

Experiment: Effect of interfaces

Thermal Transport in Yttria-Stabilized Zirconia

Point defect scattering and interface scattering



Oxygen Site

Zirconium/Yttrium site

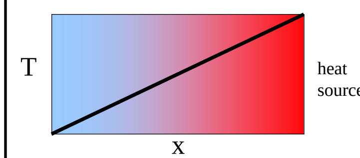
Doping with Y_2O_3:

1. Remove two ZrO_2

2. Replace with one Y_2O_3

Introduces Y^{3+} substitional ions and O vacancies, resulting in point defect scattering.

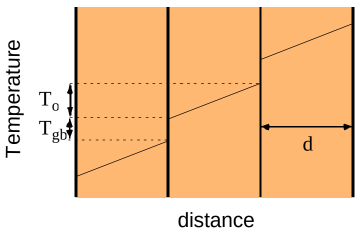
Fourier's Law



$$q = -k \frac{\partial T}{\partial x}$$

- Fourier's Law

Interfacial (Kapitza) Thermal Resistance

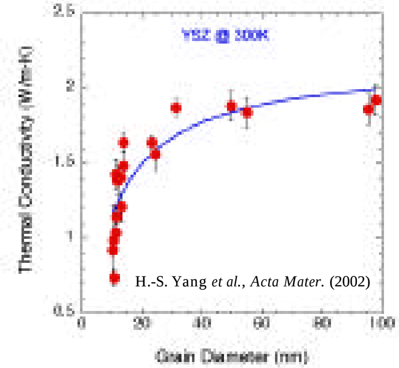


- Kapitza resistance results in temperature discontinuities at interfaces

$$k = \frac{k_o}{1 + \frac{k_o R_k}{d}}$$

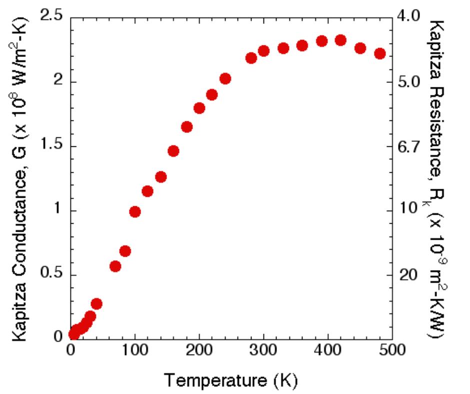
- R_k and k_o are obtained by a 2-parameter fit to $k(d)$
- Nanocrystalline materials

Kapitza resistance in YSZ



- $\boldsymbol{R_k}$ = 4.5E-9 m^2-K/W @ 300 K

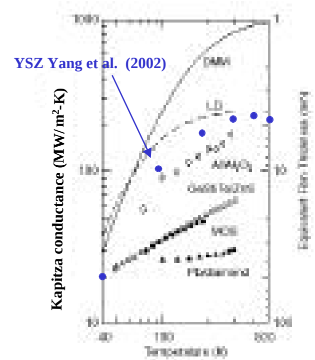
R_k temperature dependence



- At low T, $\boldsymbol{R_k}$ is dominated by C_p, but is ~ constant above Θ_D

H.-S. Yang et al., Acta Mater. (2002)

Kapitza Conductance



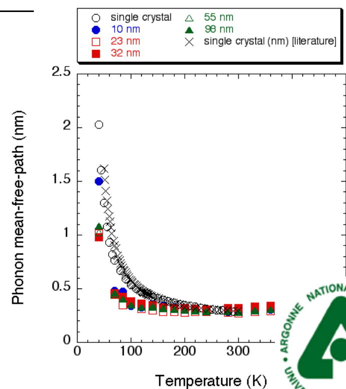
Cahill, Goodson, and Majumdar, *Jn. Heat Transfer*, **124**, 223, (2002)

- data for nanocrystalline YSZ similar to that for several heterophase systems

Phonon mean-free-path length

	Phonon mean-free-path	
	10 K	300 K
diamond	1 mm	3000 Å
quartz	75 µm	100 Å

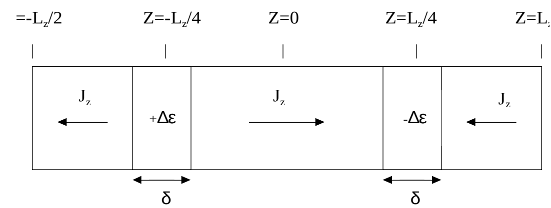
$$\Lambda = \frac{3k}{C_p v}$$



- Why is mean-free path in YSZ so small?

Simulation of Thermal Conductivity

- **Non-equilibrium: Add and remove heat**

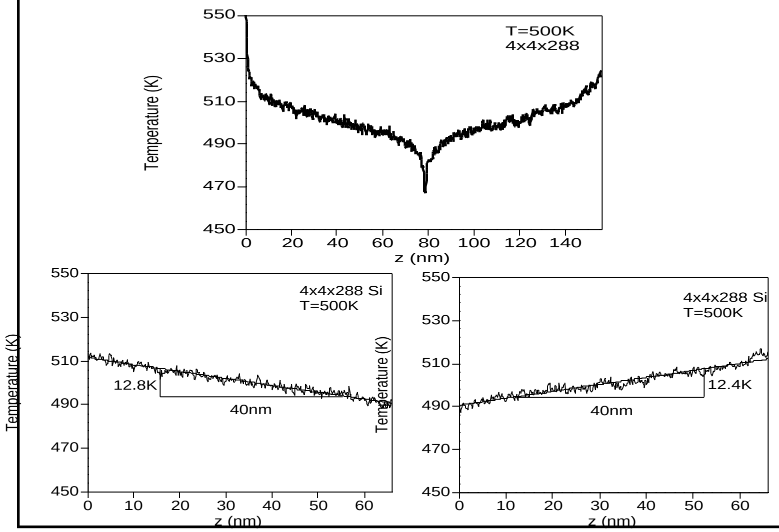


$J = -\kappa \, (dT/dz)$

- **Equilibrium: Green-Kubo Method**

$$\kappa_{\mu\nu}(\omega,T) = (1/\Omega k T^2) \langle J_\mu(\omega)\, J_\nu(\omega) \rangle$$

Temperature profile

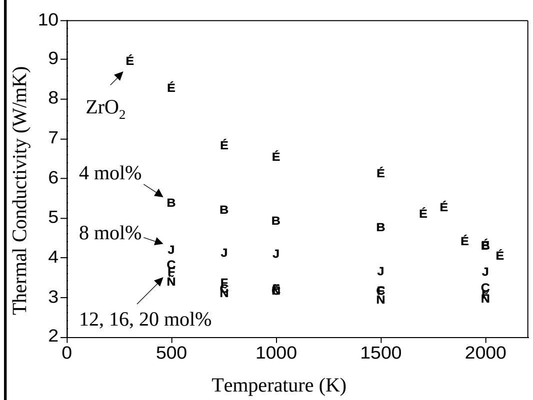


κ (W/mK) for Si

	500K	1000K
Add/Remove heat	119±40	65±16
Equilibrium Sim.	-	62±16
Expt.(extrapolated)	~120	~50

- Two method agree
- Large error bars
- No microscopic information

Temperature and Concentration Dependence of κ



	Sim. (W/mK)	Expt. (W/mK)
ZrO_2 (300K)	κ=8.92	κ=8.22
YSZ (>500K)	κ= 3.0	κ= 2.3

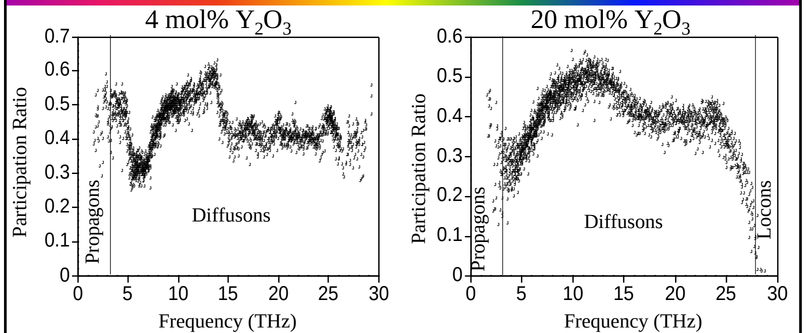
Normal Modes in Amorphous Materials

- Nature of normal modes reveals thermal conduction mechanism
- Classification scheme due to Feldman and Allen (PRB **48**, 1993):

Locons:	Localized modes
Diffusons:	Delocalized mode, but with no **k**-vector
Propagons:	Delocalized mode, can define a **k**-vector

•For a-Si 93% of normal modes are diffusons

Participation Ratios



- Localization a small effect, like a-Si
- Like a-Si, diffusons are dominant transport mechanism
- Hence concentration and temperature independence of κ

Thermal conductivity

Experiment

- currently little information on interfaces
- interpretation is indirect

Simulation

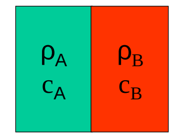
- computationally costly
- 10-20% error bars
- one number
- no microscopic information

Elucidate elementary processes:

Simulate phonon-interface interactions

Interface Scattering

Acoustic mismatch model

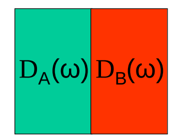


$Z = \rho c$

$t_{AB} = 4Z_AZ_B/ (Z_A + Z_B)^2$

ρ -density
c -speed of sound

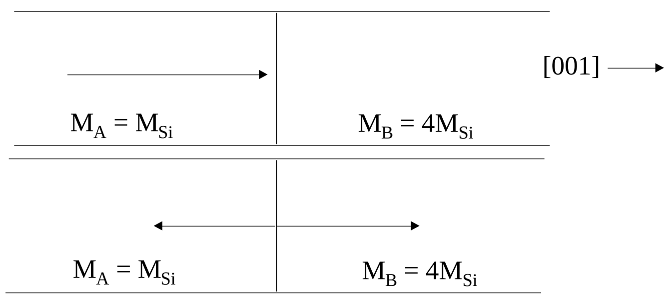
Diffuse mismatch model



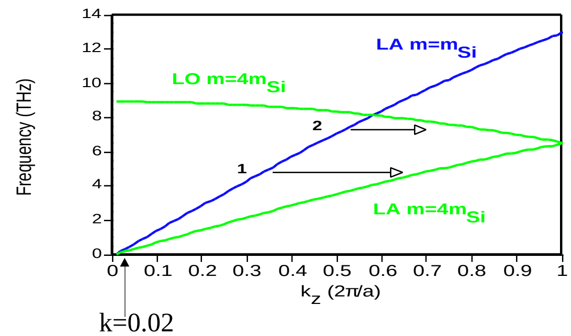
$t_{AB}(\omega) = D_B(\omega)/ (D_A(\omega)+D_B(\omega))$

$D(\omega)$ - density of states

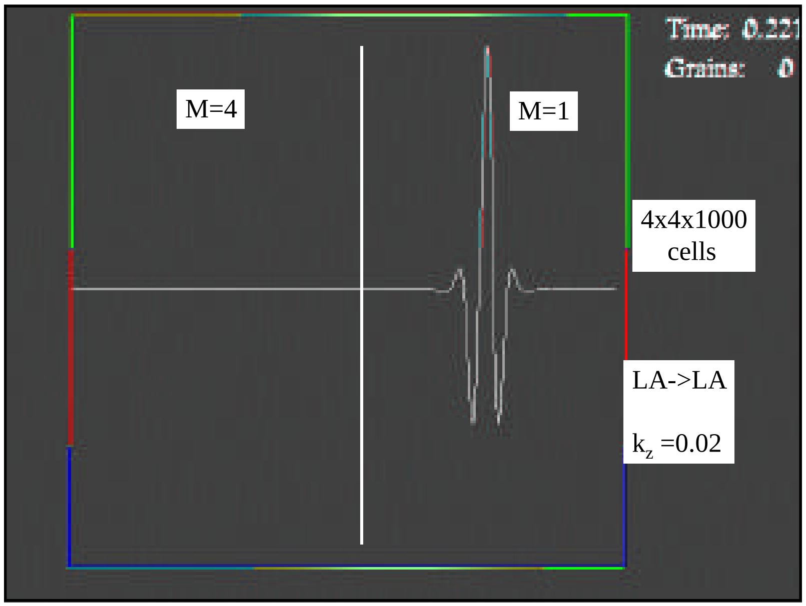
Elementary Process of Thermal Conductivity in Interfacial Systems: Phonon/Interface Scattering

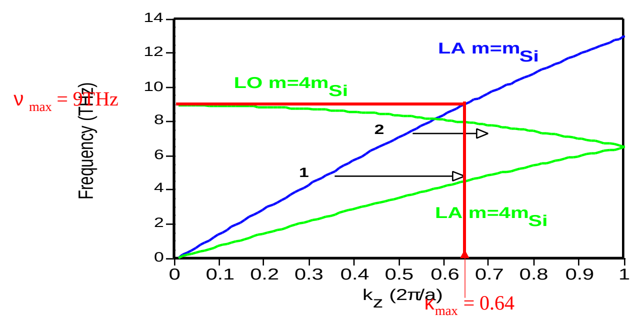


- Interface created from a simple mass discontinuity
- All interactions: Stillinger-Weber Si potential

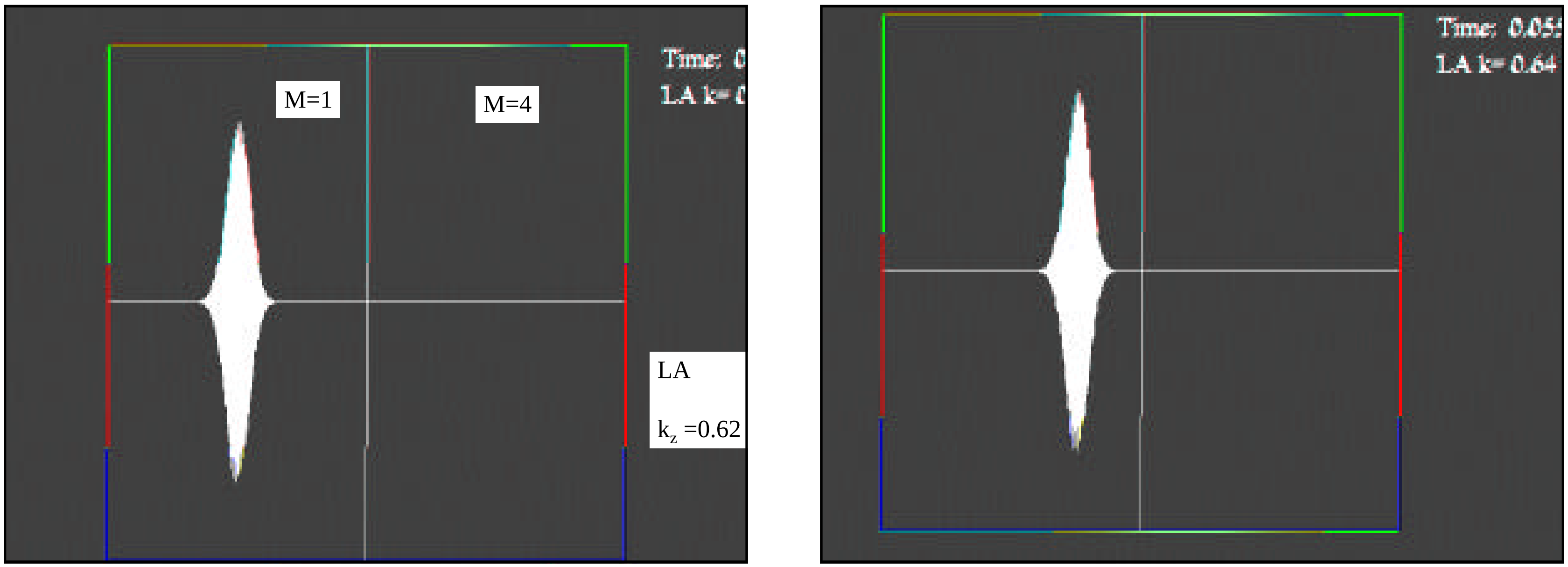


1. LA LA transmission
2. LA LO transmission

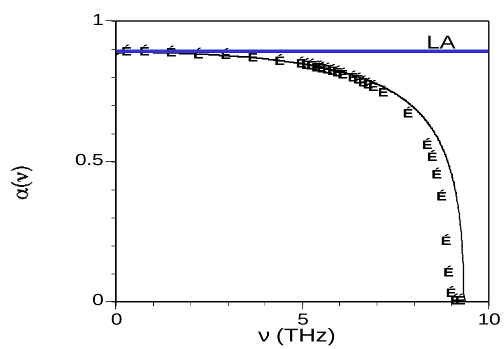




1. LA LA transmission
2. LA LO transmission


Time: 0
LA k= 0
M=1
M=4
LA
k_z =0.62
Time: 0.055
LA k= 0.64

Interfacial scattering rates

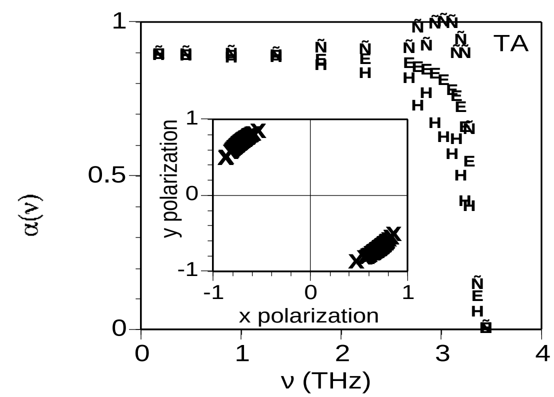


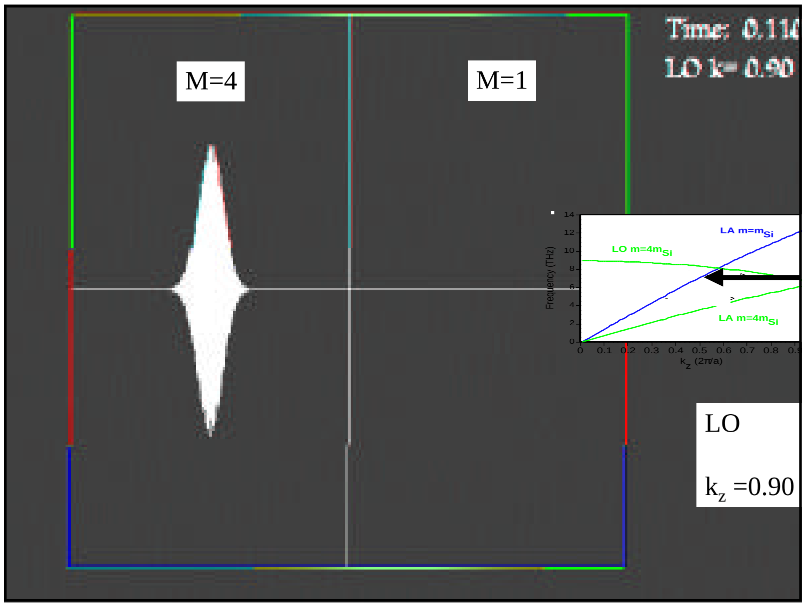
Acoustic mismatch model

1. α(ν) represents fraction of energy transmitted at frequen

2. Low ν regime described by continuum model

Transverse Acoustic Phonons



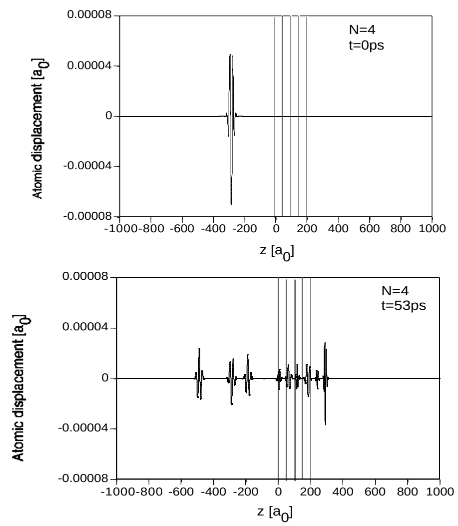


N=4 Superlattice

A	B	A	B	A	B

· $M_A = M_{Si}$ $M_B = 4M_{Si}$

MD simulation of phonons through N=4 superlattice



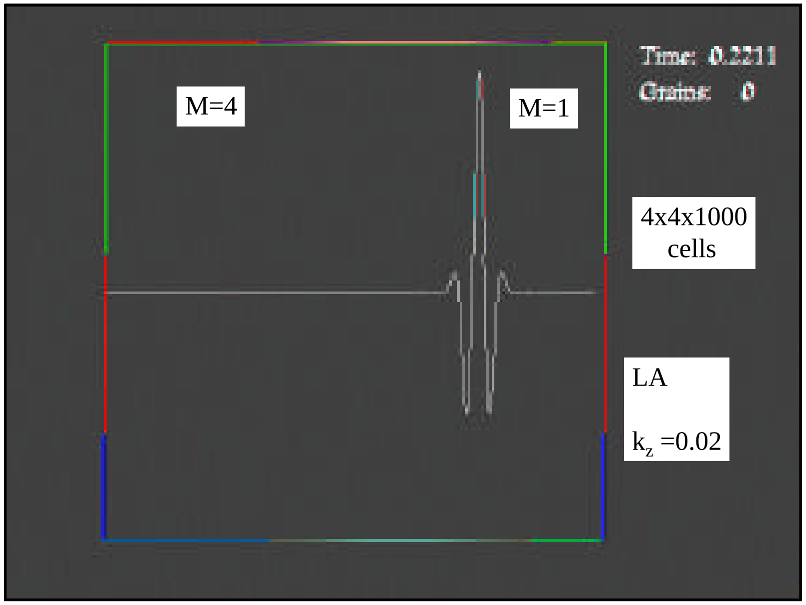
N=4 Superlattice Structure

A	B	A	B	A	B

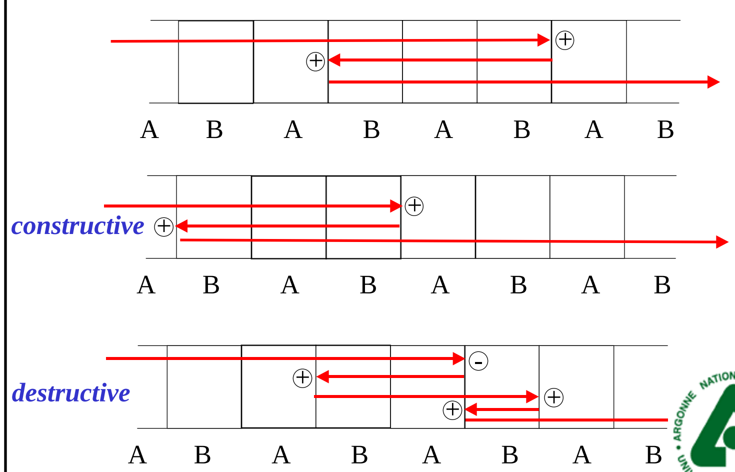
· $M_A = M_{Si}$ $M_B = 4M_{Si}$

Scattering at interfaces

- Energy transmission coefficient: α
- Amplitude
  - transmission $\sqrt{\alpha}$
  - reflection B-A $\sqrt{1-\alpha}$
  - reflection A-B $-\sqrt{1-\alpha}$ phase change



Constructive vs. Destructive Interference



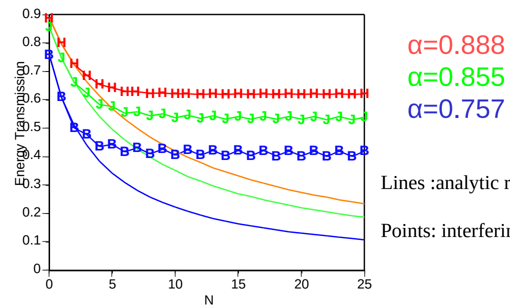
MD simulation and particle model agree quantitatively

Wave packet	time [t_0]	E/E_0 MD	E/E_0 particles with interference	E/E_0 particles without interference
1	1.00	0.70015	0.70023	0.70023
2	1.67	0.00880	0.00878	0.00878
3	2.33	0.01083	0.01086	0.00889
4	3.00	0.00505	0.00512	0.00704

Interference effects essential in superlattices

· N=2, N=4 results of particle model agree exactly with MD results

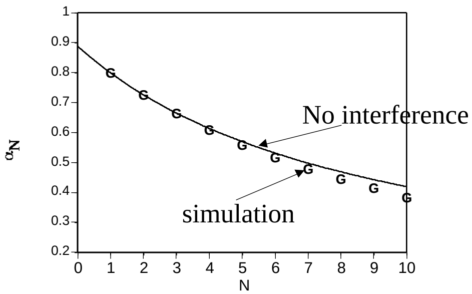
· Interference effects *increase* the amount of transmitted energy



Lines :analytic results with no interference.

Points: interfering particle model.

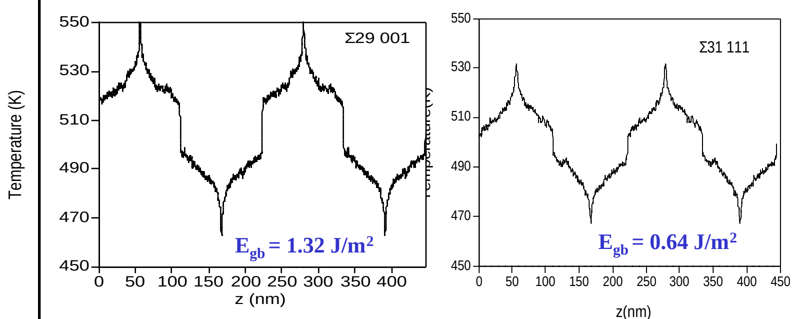
Random layer thickness



Interference effects small but destructive interference dominates

Thermal Transport at grain boundaries

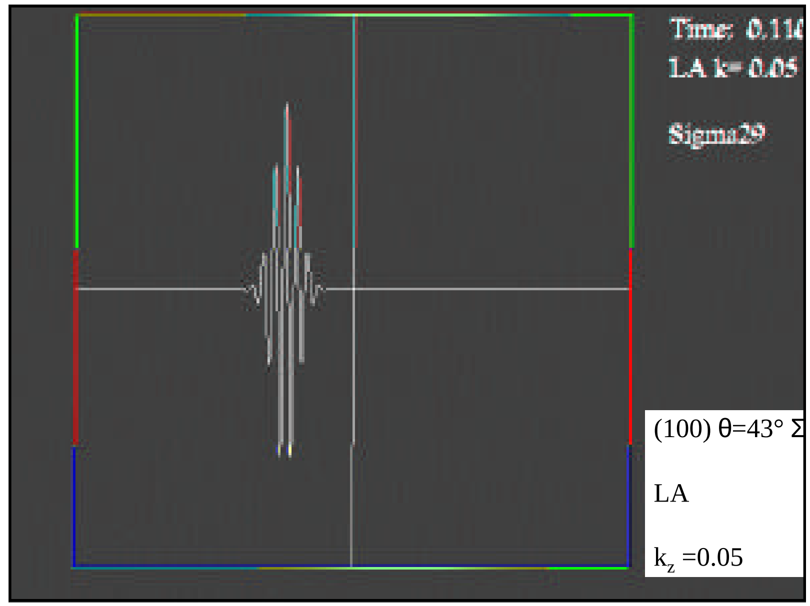
Interfacial scattering at twist grain boundaries in Si

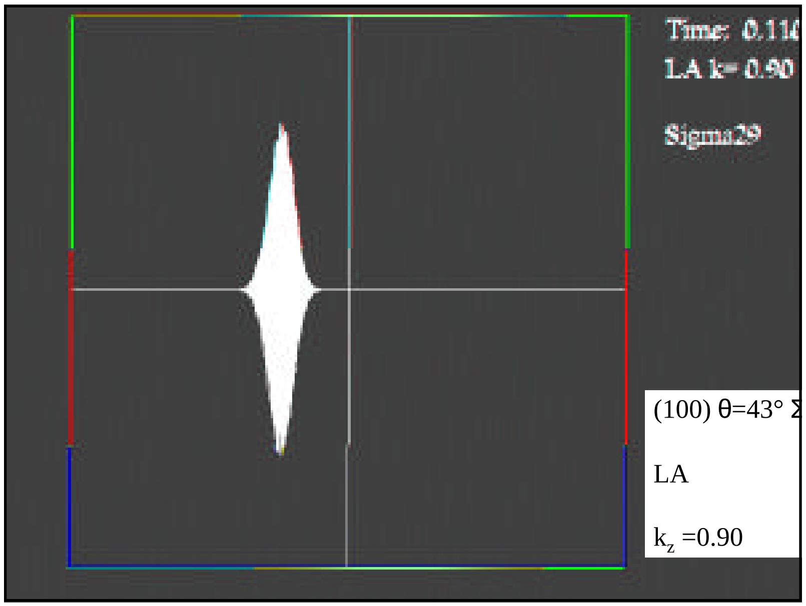


$$J = \sigma_K \, \Delta T$$

σ_K = 0.85GW/m²K for (001) Σ29 boundary - high energy

σ_K = 1.53GW/m²K for (111) Σ31 boundary - low energy



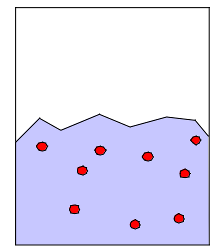


Nanofluids

- Fluids have low ***k*** compared to most solids

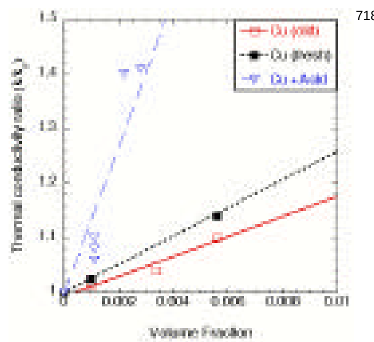
Material		Room Temperature Thermal Conductivity (W/m-K)
Metallic Solids:	Silver	429
	Copper	401
	Aluminum	237
Nonmetallic Solids:	Diamond	3300
	Silicon	148
	Alumina (Al_2O_3)	40
Metallic Liquids:	Sodium @ 644K	72.3
Nonmetallic Liquids:	Water	0.613
	Ethylene Glycol	0.253
	Engine Oil	0.145

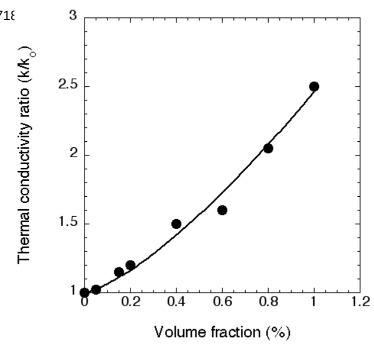
U.S. Choi and J.A. Eastman, "Enhanced Heat Transfer Using Nanofluids," U.S. Patent #6,221,275



- Goal is to enhance effective fluid thermal conductivity and heat transfer coefficient by suspending solid nanoparticles
- Nanoparticles provide advantages due to better dispersion behavior, less clogging and abrasion, and much larger total surface area

Thermal conductivity of nanofluids

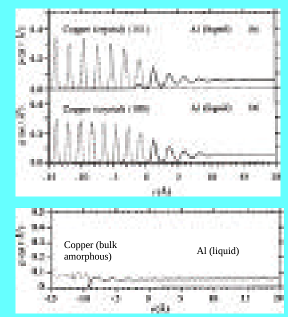




J. A Eastman, S. Choi ANL

Possible mechanisms

- Possible microscopic mechanisms:
  (P. Keblinski *et al.*, *ASME Journal of Heat Transfer* **45**, pp. 855-863 (2002))
  - Brownian motion (but thermal motion is expected to be faster than expected particle motion)
  - Effect of particles on liquid local ordering (effectively decreases average spacing between particles)
  - Ballistic rather than diffusive thermal transport in the particles (but isn't expected to affect transport between particles)
  - Nanoparticle clustering (would probably lead to poor dispersion properties)
- Simulation studies are in progress
  (L. Xue, P. Keblinski and S.R. Phillpot)



P. Geysermans *et al.*, Jn. Chem. Phys.. **113**, 6382, 2000.

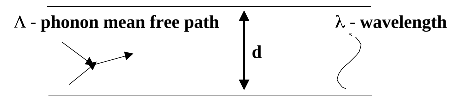
Conclusions

- MD simulation of thermal conductivity
- MD simulation of phonon dynamics
- Particle model of phonon dynamics
  - interference effects important
  - can quantitatively reproduce MD results

Outlook

- MD simulation of phonon dynamics at grain boundaries
  - mode coupling
  - diffusive scattering at GBs
- Particle model
  - finite mean free path, mode coupling,
  - diffusive scattering
  - -2-d and 3-d systems

Outlook- Multiscale Simulation of Thermal Transport



Atomistic Simulations

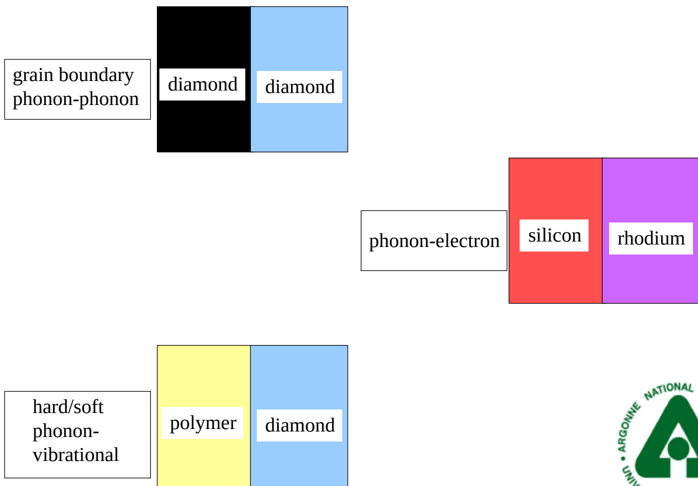
Continuum Simulations of Heat Transfer

d ~ λ

d ~ Λ

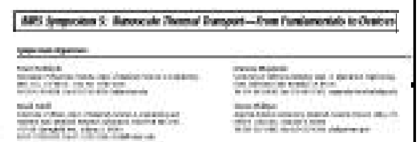
Mesoscale Models of Phonon Transport

Outlook - Different mechanisms

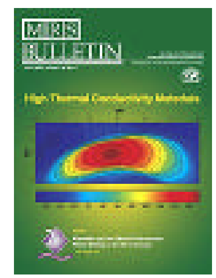


Outlook - Thermal Transport in Nanostructures

Report commissioned by DOE Council for Materials, 2001
(to be published in Applied Physics Reviews)



MRS Spring Meeting, 2003



MRS Bulletin June 2001