Elastic versus Inelastic Scattering at Interfaces

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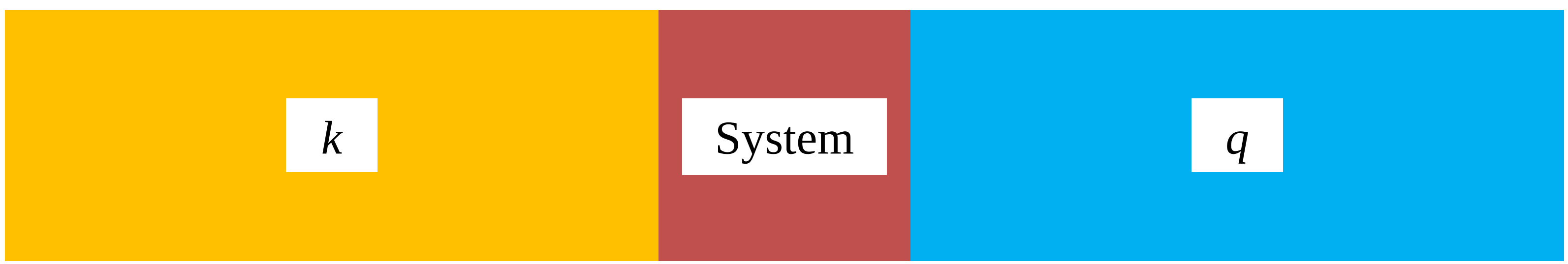
GENERIC INTERFACE PROBLEM

Leads are bulk and have well defined dispersion relation

Isolated Bulk-Bulk Interface: AMM, DMM, Atomistic (SBM/Green’s Function) (see Eric’s thesis)

$\xrightarrow[z]{}$



Bulk-Bulk Interface “System”: DMM, Atomistic (SBM/Green’s Function)

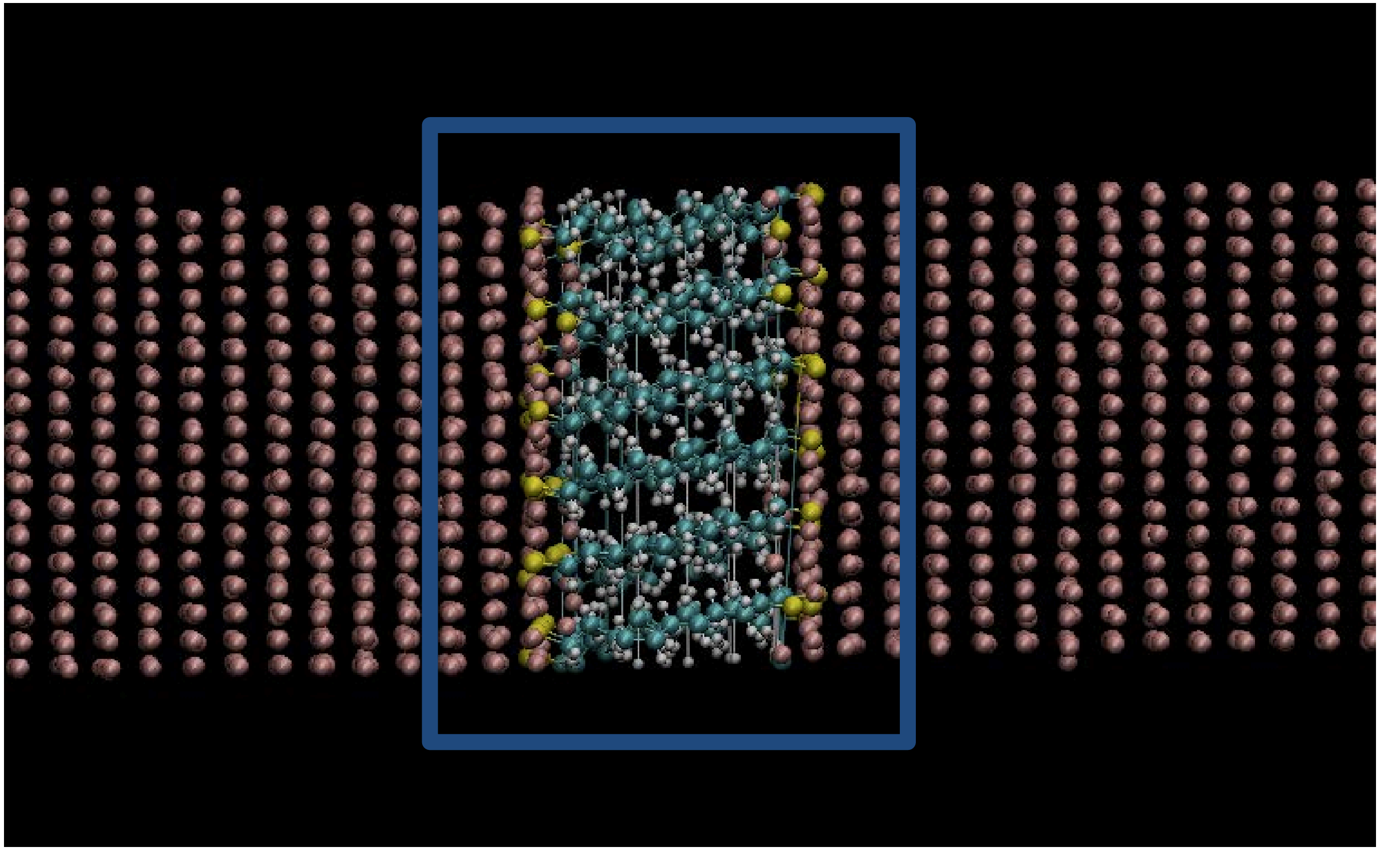
Landauer-Buttiker:
Phonons treated as particles with MFP/coherence length

$$\dot{Q} = \sum_{k_z>0} n_k v_k \epsilon_k \alpha_k + \sum_{q_z<0} n_q v_q \epsilon_q \alpha_q$$

ORGANIC INTERFACE

Specific case: Interface system is organic molecule.

Complication: Discrete vibrational states



ELASTIC VERSUS INELASTIC SCATTERING AT ISOLATED BULK-BULK INTERFACE

This is what I think: the generic Landauer energy flux expression can be written in terms of orders of phonon interactions at the interface

$$\dot{Q}_{L\to R} = \overbrace{\boxed{\sum_{k,q} n_k \alpha_{k\to q} v_q \epsilon_q \delta_{\omega_k,\omega_q} \delta_{k,q+nG}}}^{\text{TWO-PHONON PART}} + \overbrace{\boxed{\sum_{k,q,q'} n_k \alpha_{k\to q,q'} (v_q \epsilon_q + v_{q'} \epsilon_{q'}) \delta_{\omega_k,\omega_q+\omega_{q'}} \delta_{k,q+q'+nG}}}^{\text{1}^{\text{st}}\text{ THREE-PHONON PART}}$$

$$+ \underbrace{\boxed{\sum_{k,k',q} (n_k + n_{k'}) \alpha_{k,k'\to q} v_q \epsilon_q \delta_{\omega_k+\omega_{k'}\omega_q} \delta_{k+k',q+nG}}}_{\text{2}^{\text{nd}}\text{ THREE-PHONON PART}} + \dots$$

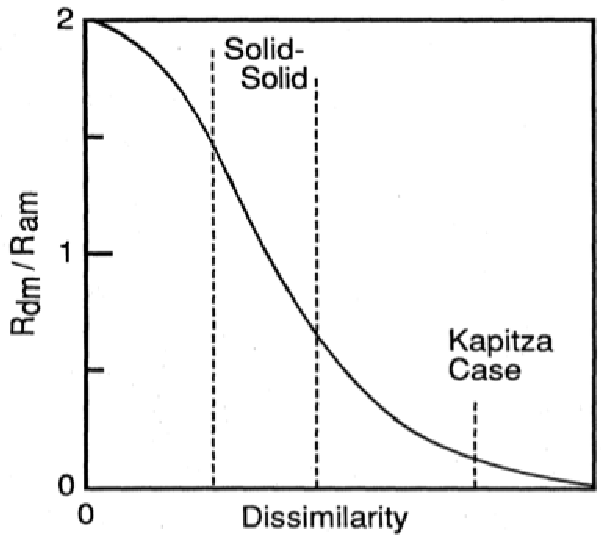
ELASTIC SCATTERING

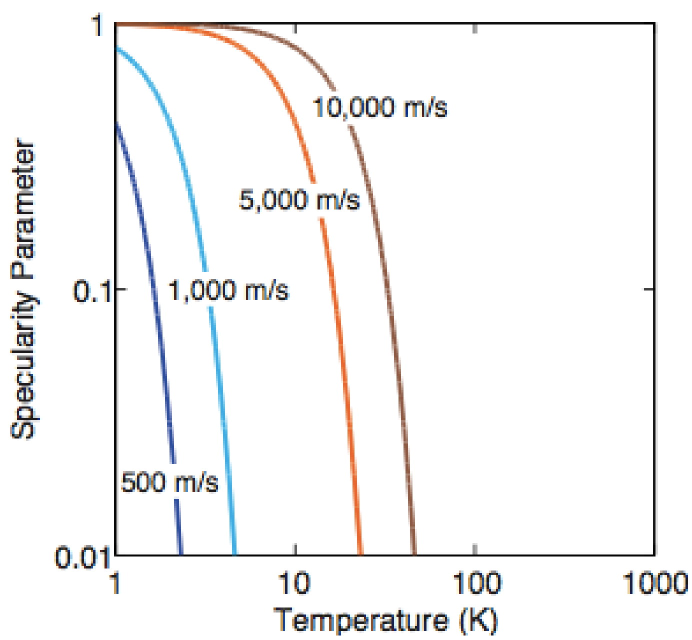
- Incident and outgoing phonons have the same energy (frequency):
  TWO PHONON PROCESSES AT THE INTERFACE

INELASTIC SCATTERING

- Incident and outgoing phonons mix energies, but total energy is still conserved:
  THREE OR MORE PHONONS PROCESSES AT THE INTERFACE
- Derek Stewart: Three-phonon processes at the interface are “rare”

ELASTIC VERSUS INELASTIC SCATTERING AT ISOLATED BULK-BULK INTERFACE: DMM



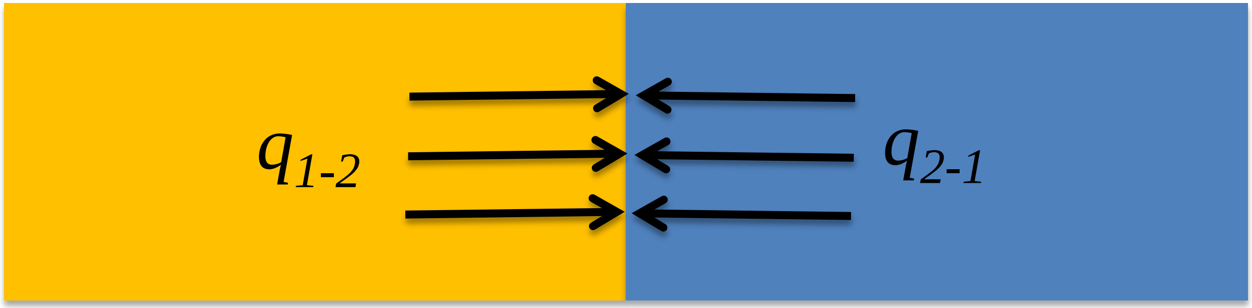


- Higher temperatures than AMM
- All phonons scatter diffusely
  - Lose all memory of its state before scattering event
- Transmission function dictated by difference in density of states
  - Principle of detailed balance

[1] Swartz E.T and Pohl R.O, RMP **61**(3), 1989

[2] Duda JC et al. JAP **108**(073515), 2010

ELASTIC VERSUS INELASTIC SCATTERING AT ISOLATED BULK-BULK INTERFACE: DMM



Detailed balance

$$q_{1\text{-}2} = q_{2\text{-}1}$$

Diffuse scattering

$$\alpha_{1\text{-}2} = 1 - \alpha_{2\text{-}1}$$

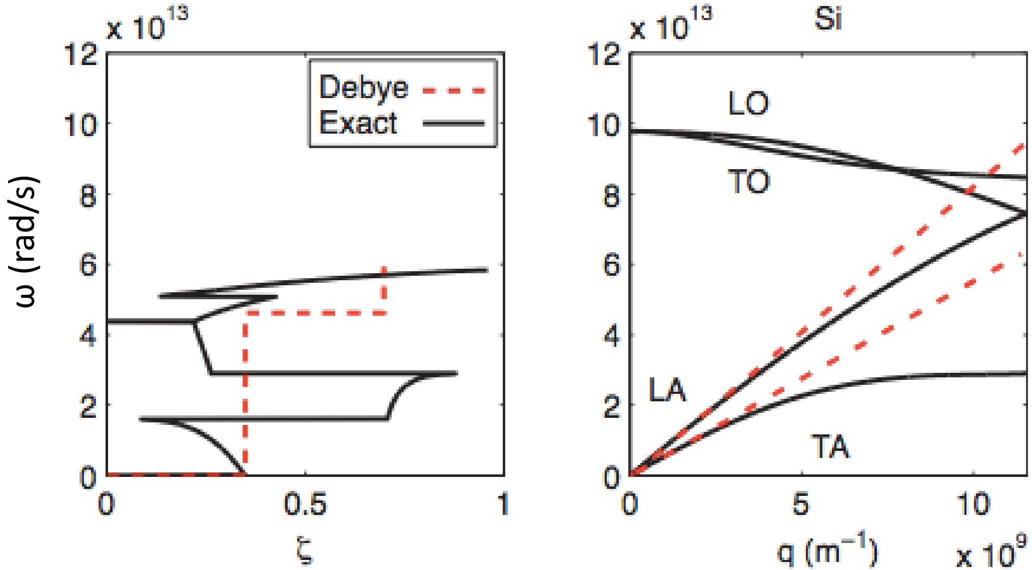
Elastic scattering assumption

$$\omega_1 = \omega_2$$

$$\zeta^{1\to 2}(k_1) = \frac{\Sigma_j \hbar\omega_{j,2} k_{j,2}^2 v_{j,2} f_0 dk_{j,2}}{\Sigma_j \hbar\omega_{j,2} k_{j,2}^2 v_{j,2} f_0 dk_{j,2} + \Sigma_j \hbar\omega_{j,1} k_{j,1}^2 v_{j,1} f_0 dk_{j,1}}.$$

$$\zeta^{1\to 2}(\omega) = \frac{\Sigma_j [k_{j,2}(\omega)]^2}{\Sigma_j [k_{j,2}(\omega)]^2 + \Sigma_j [k_{j,1}(\omega)]^2}.$$

ELASTIC VERSUS INELASTIC SCATTERING AT ISOLATED BULK-BULK INTERFACE: DMM



- Mode-dependent transmission function
- For inelastic, no mode dependence
  - Single transmission coefficient

[1] Duda JC et al. JAP **108**(073515), 2010

ELASTIC SCATTERING AT ISOLATED INTERFACE

Only two-phonon processes, so we can just deal with generic Landauer formula

KHALATNIKOV

$$\dot{Q} = \sum_{k_z>0} n_k v_k \epsilon_k \alpha_k + \sum_{q_z<0} n_q v_q \epsilon_q \alpha_q$$

- Distribution functions on both sides are Bose-Einstein distributions at different temperatures
- Incorrectly predicts finite Kapitza conductance when no interface exists

SIMONS/VISSCHER

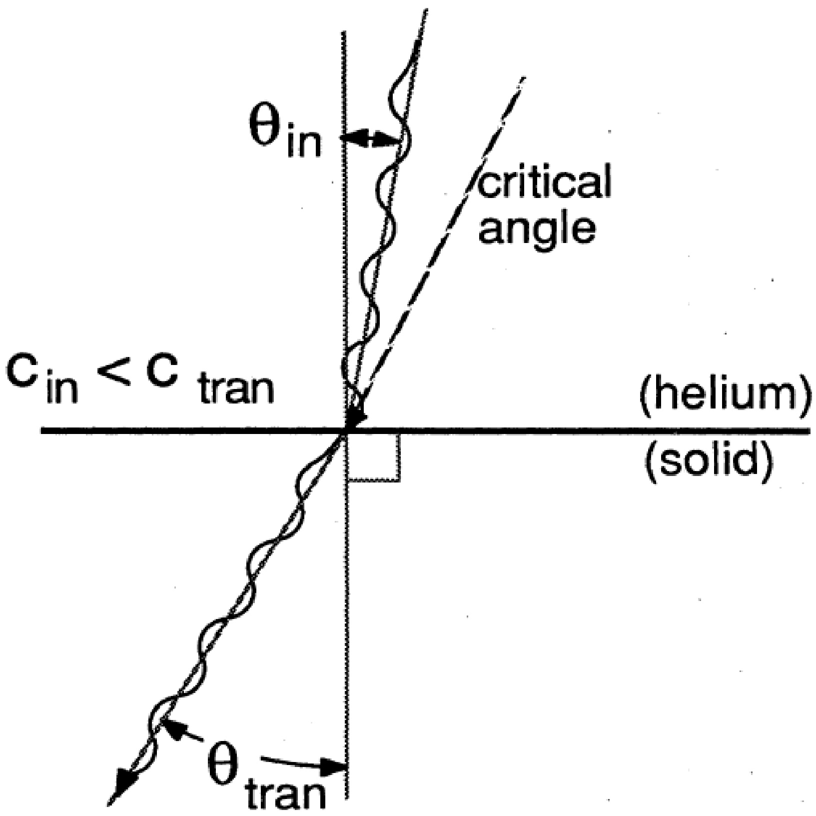
- Distributions are Bose-Einstein modified by a non-equilibrium portion responsible for the heat

$$n_k = \overline{n}_k + \hat{n}_k \rightarrow \dot{Q}_{bulk} = 2\sum_{k_z>0} v_k \epsilon_k \hat{n}_k$$

Lumpkin *et al*, Phys. Rev. B **17** 4295 (1977)

ELASTIC VERSUS INELASTIC SCATTERING AT ISOLATED BULK-BULK INTERFACE: AMM

Throw away the Landauer picture. Adopt a picture where the two materials are continua and we can talk about displacement waves. Plane waves impinge on the interface and refract or reflect



$$c_{top} \sin\theta_{top} = c_{bot} \sin\theta_{bot}$$

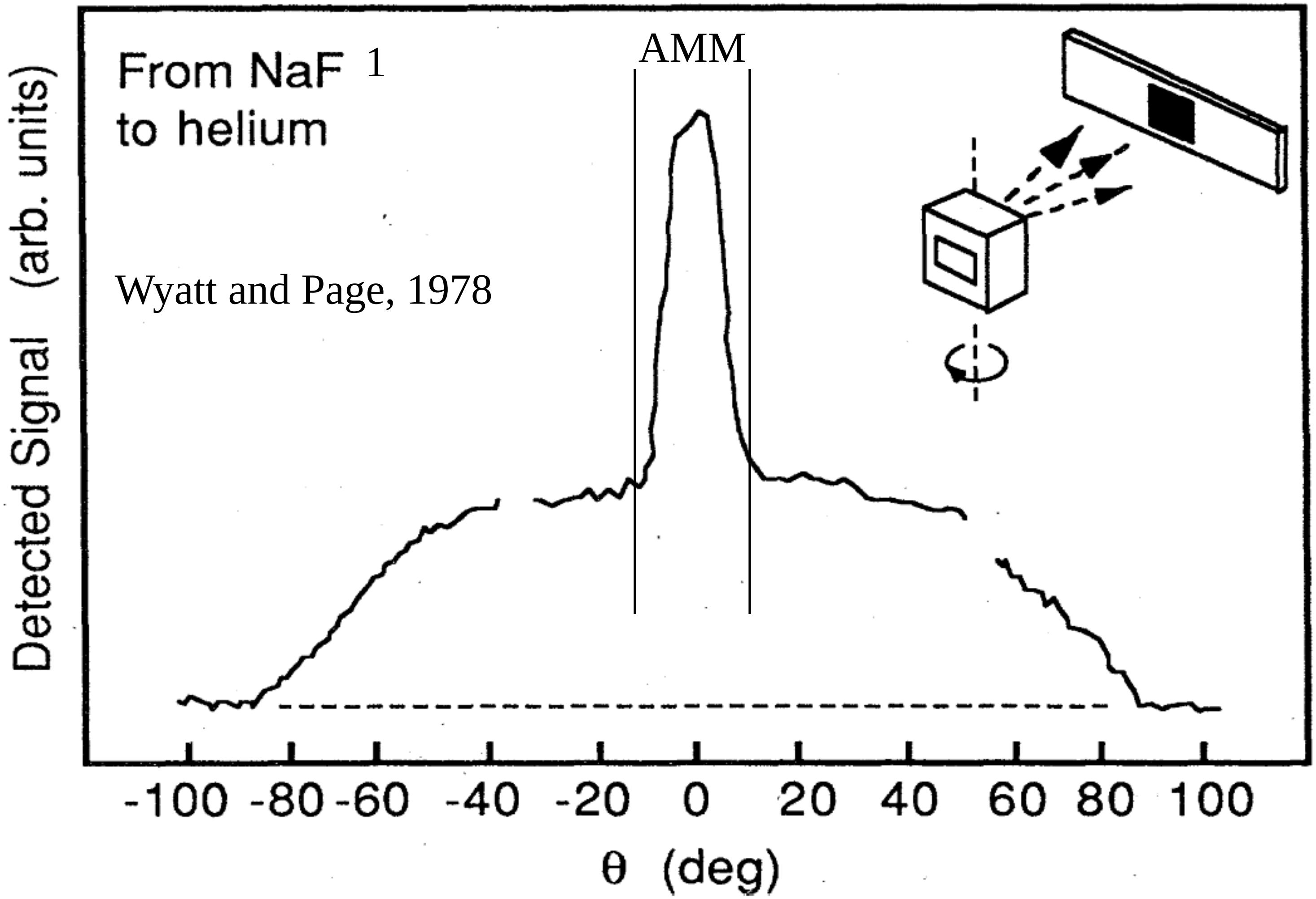
For a mode k

$$\alpha_k = \frac{4\rho_{top}\rho_{bot}c_{k,top}c_{k,bot}}{(\rho_{top}c_{k,top} + \rho_{bot}c_{k,bot})^2}$$

Typically Debye approximation is taken, so that there is one group velocity for a branch.

Swartz and Pohl, Rev. Mod. Phys. **61**, 605 (1989)

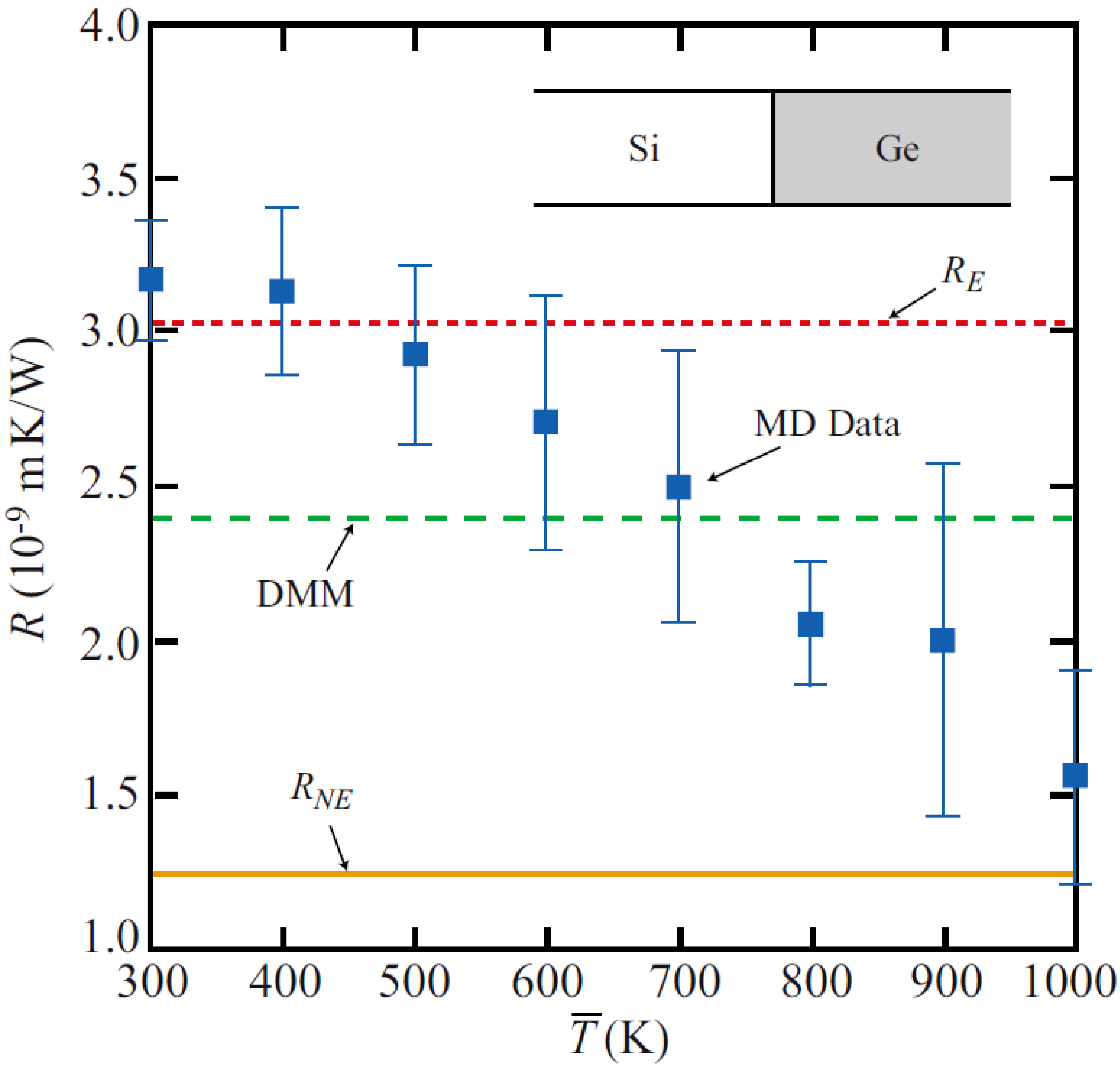
EVIDENCE FOR INELASTIC SCATTERING: CASE SPECIFIC TO AMM



"In the solid-liquid situation, neither of these conditions [for elastic scattering] is close to being satisfied; in the solid-solid case the forces may be nearly harmonic, but due to boundary conditions the [phonon] density matrix near the interface is not expected to be diagonal."[2]

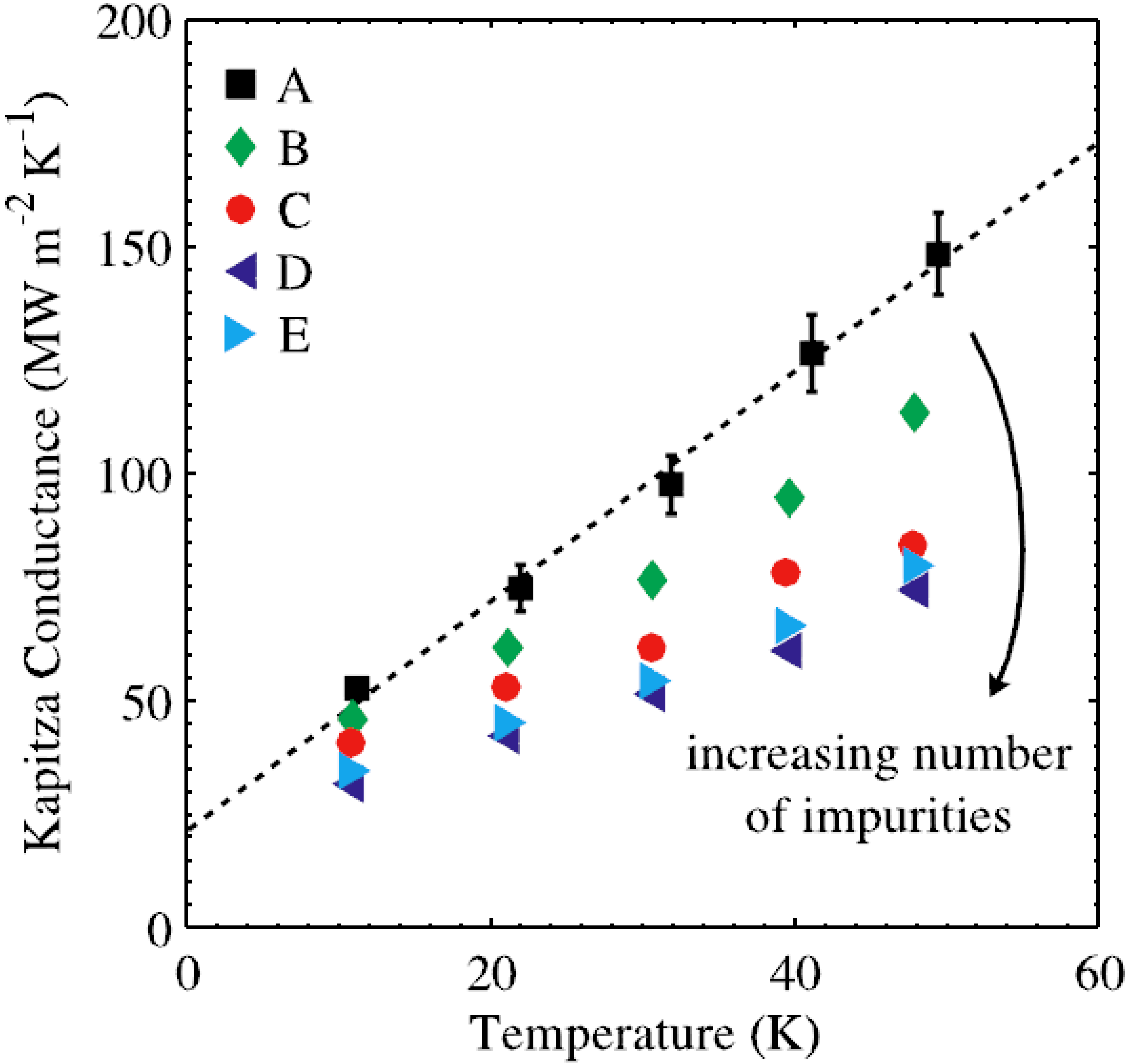
1. Swartz and Pohl, Rev. Mod. Phys. **61**, 605 (1989)
2. Lumpkin *et al*, Phys. Rev. B **17** 4295 (1977)

EVIDENCE FOR INELASTIC SCATTERING: MD ISOLATED INTERFACE



Landry and McGaughey, Phys. Rev. B **80**, 165304 (2009)

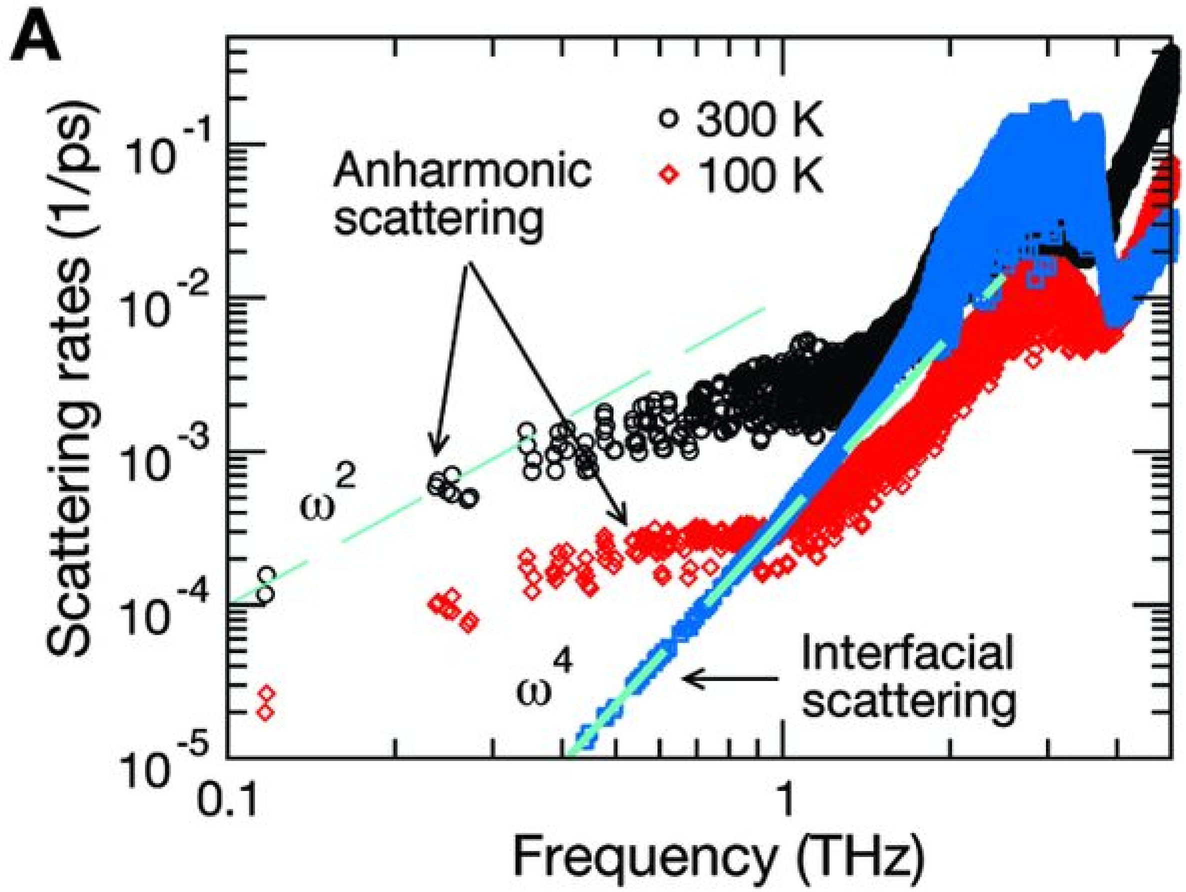
EVIDENCE FOR INELASTIC SCATTERING: ANOTHER MD ISOLATED INTERFACE



Duda *et al*, J. Appl. Phys. **112**, 073519 (2012)

SCATTERING RATES: LIFETIMES IN BULK ALD IS EQUIVALENT TRANSMISSION IN INTERFACE

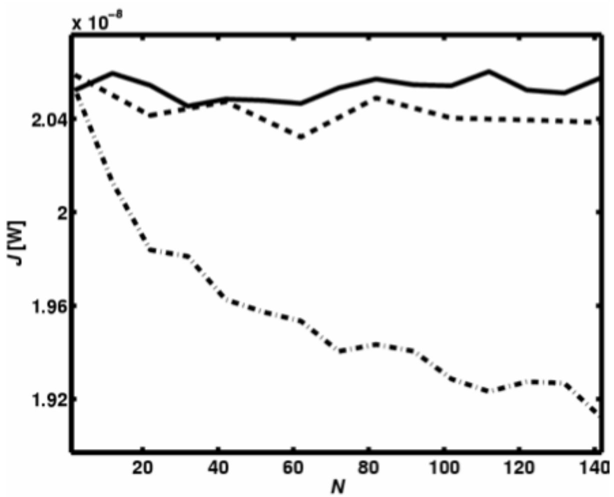
GaAs/AlAs Superlattice

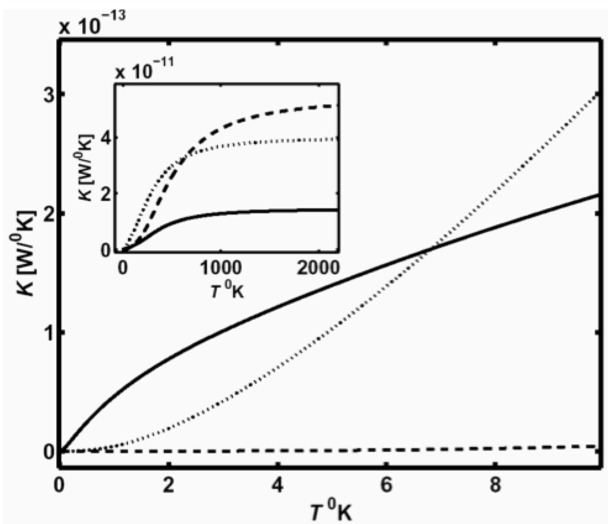


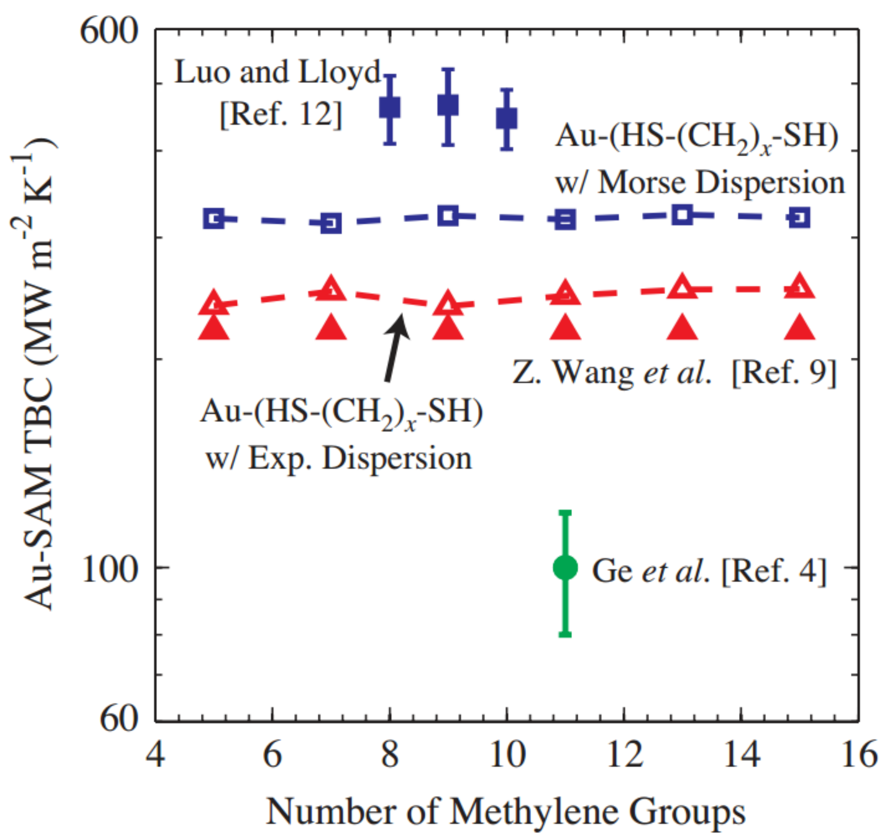
"In this work, we include both interface roughness as the random mixing of Ga and Al atoms in a narrow region around the interface and three-phonon processes from first-principles without introducing any fitting parameters. Scattering caused by this mixing was computed from Fermi's golden rule."[1]

1. Luckyanova *et al*, Science **338** 6109 (2012)

- Very small anharmonic effects







[1] Segal D et al. JCP **119**:6840, 2003
[2] Duda JC et al. JAP **108**(073515), 2010