UK Thermoelectric Network Meeting, February 2018, Edinburgh

Quantum transport simulations for nanostructured thermoelectric materials

Neophytos Neophytou

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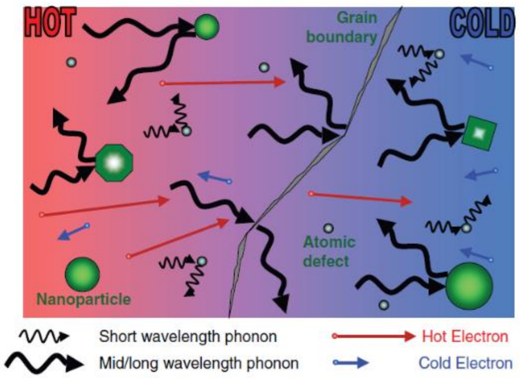
FFG FWF SEVENTH FRAMEWORK PROGRAMME HORIZON 2020 erc

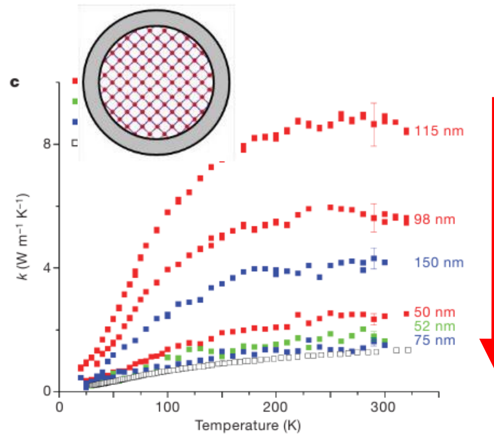
WARWICK

Nanostructuring lowers κ (significantly)



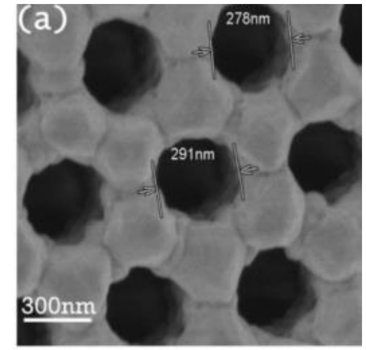
Vineis, ***Adv. Mat.***, 2010

- Nanostructuring
- phonon engineering



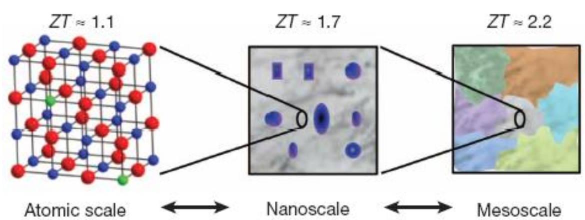
Hochbaum, ***Nature***, 2008

κ = **1-2** W $K^{-1}m^{-1}$

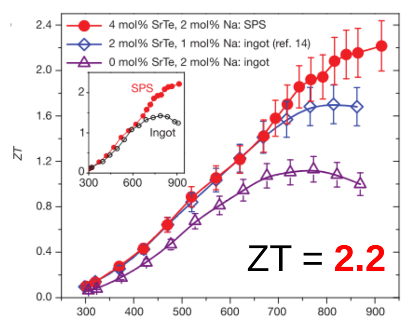


Perez *et* al., ***Scientific Reports*** 6, (2016): 32778.

κ = **0.55** W $K^{-1}m^{-1}$
~30 nm SiGe nanopore film



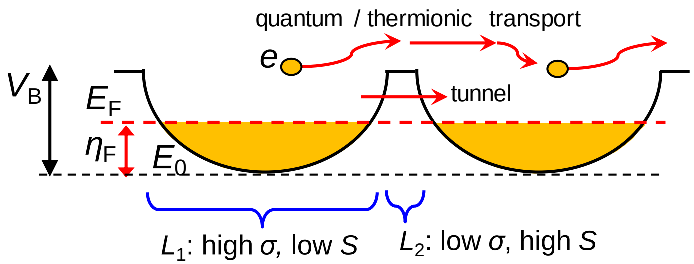
Biswas *et* al, ***Nature***, 2012.
(p-type PbTe)

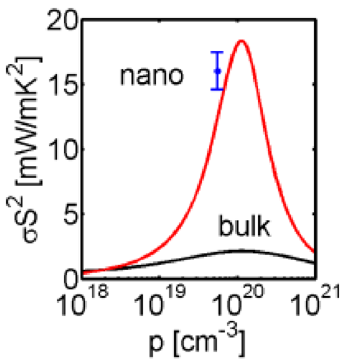


$$ZT = \frac{\sigma S^2 T}{k_e + k_l}$$

How about the power factor ?

with
Dario Narducci
(Milano-Bicocca)



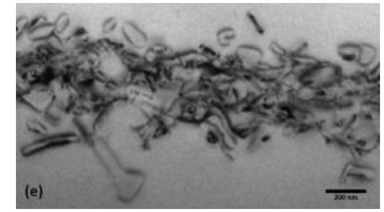


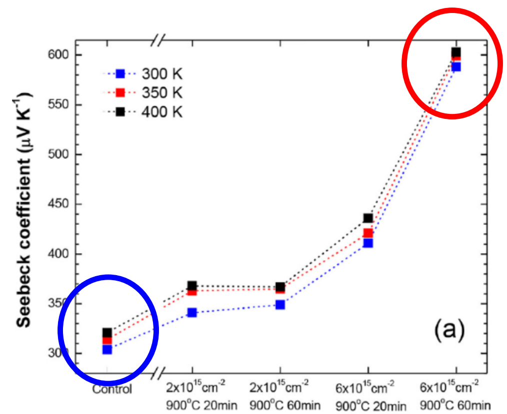
σ ↑ S ↑

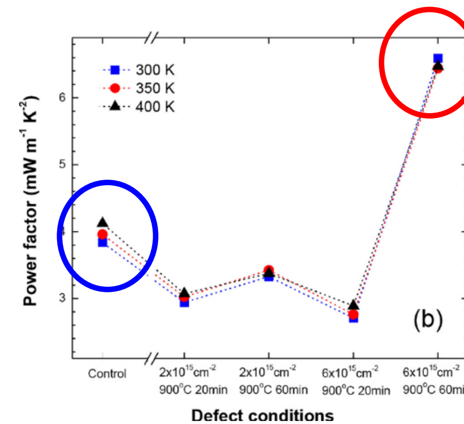
Large σ and S

Very high PF:
poly-c materials: 15 mW/K²m⁻¹
(~5x compared to bulk p-type Si)

with
Nick Bennett
(Heriot Watt, UK)







S ↑ PF ↑

High PF:
Dislocations in Si: 6.5 mW/K²m⁻¹
(~70% higher than bulk n-type Si)

Neophytou, Zianni, Kosina, Frabboni, Lorenzi, Narducci, ***Nanotechnology***, 24, 205402, 2013.
Lorenzi, Narducci, Totini, Frabboni, Gazzadi, Ottaviani, Neophytou, Zianni, ***J. Electr. Mat.***, 43, 3812, 2014
Bennett, Byrne, Cowley, Neophytou, ***Appl. Phys. Lett***., 109, 173905, 2016.

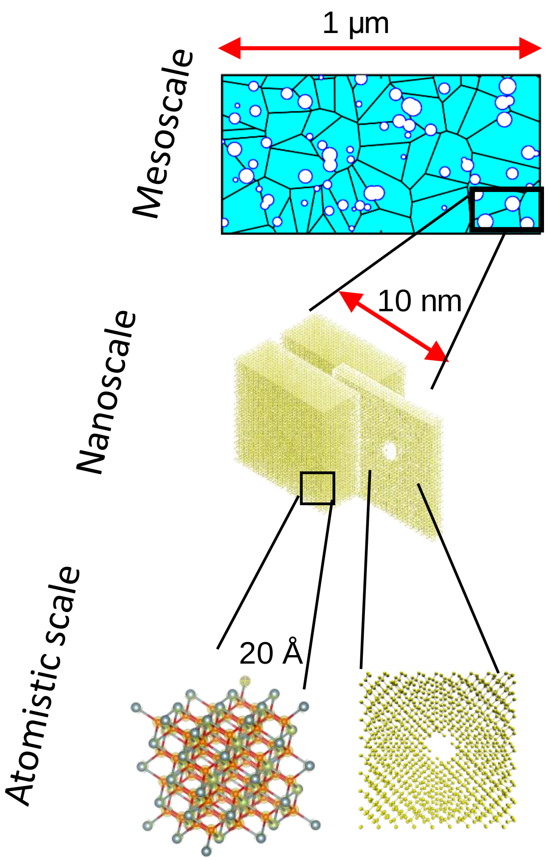
Goals of this talk

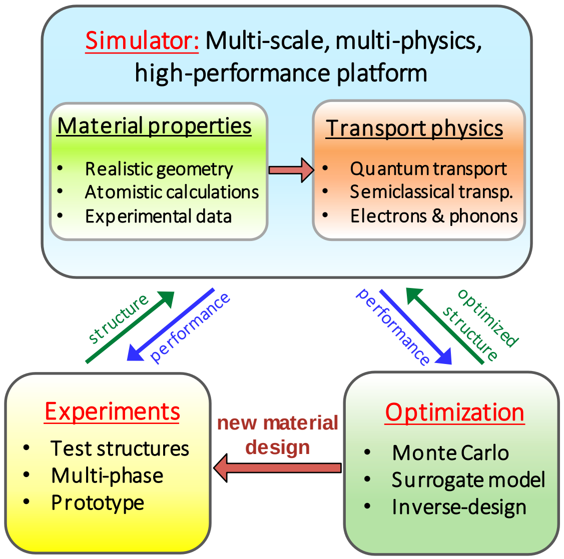
- *The Non-Equilibrium Green's Function (NEGF): Studies on superlattices and nanocomposites*

- *Expansion to multi-physics, multi-scale capabilities*

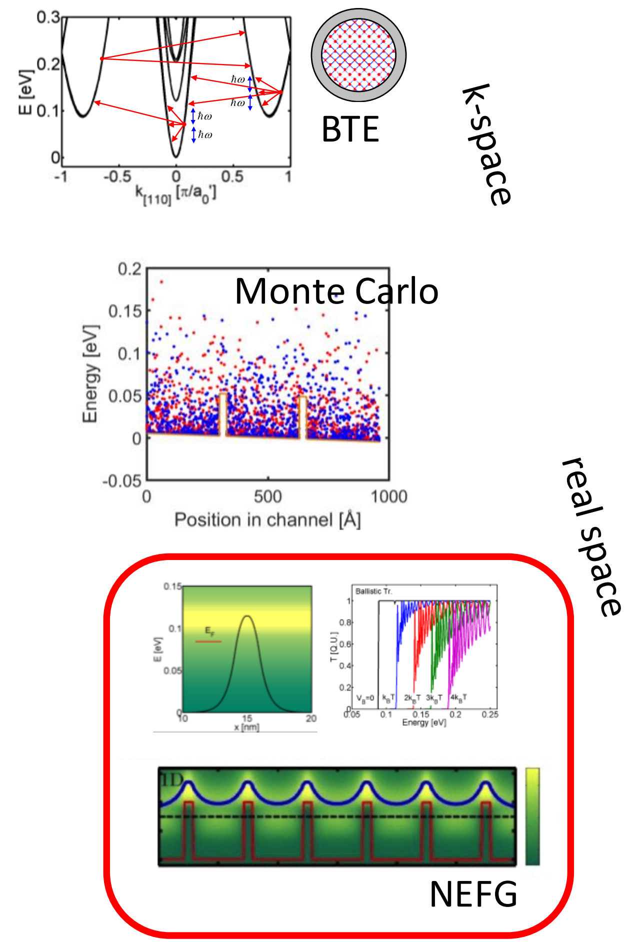
Overview of nanostructure simulation activities

MULTI-SCALE GEOMETRIES
(atomistic to continuum)

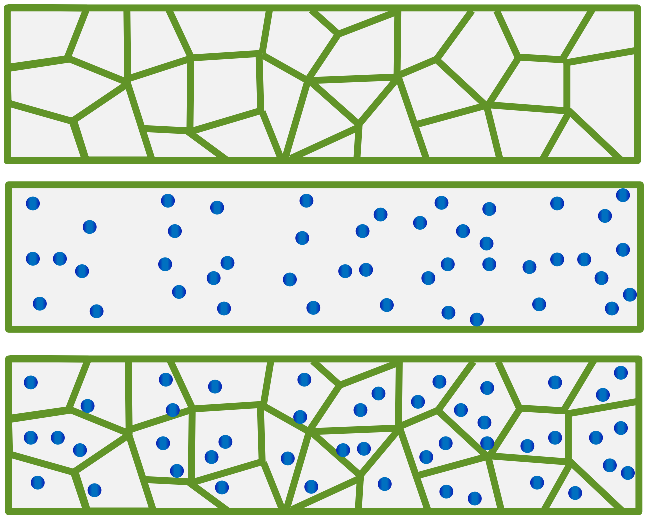




MULTI-PHYSICS TRANSPORT
(semiclassical to quantum)



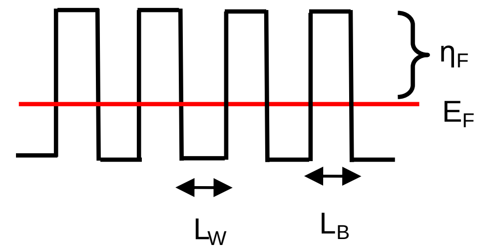
Features for PF improvement

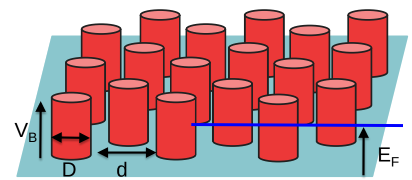


Polycrystalline (PC)

Nanoinclusions (NI)

PC+NI

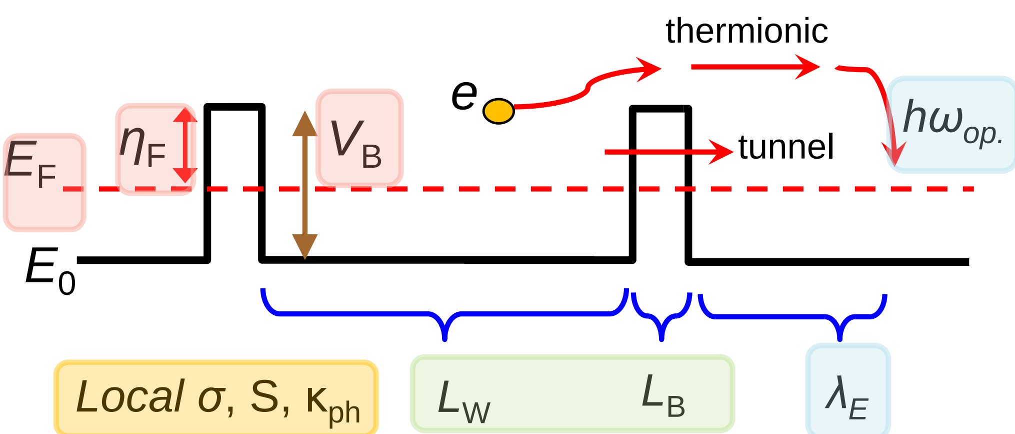


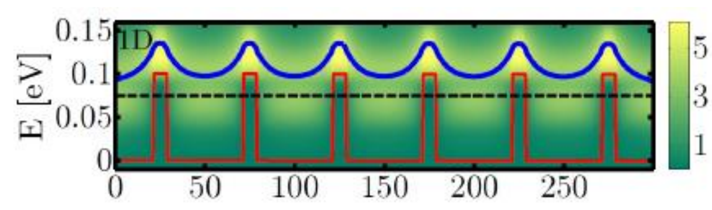


Feature sizes:
a few – 10s – 100s of nanometers

Barriers: $S \sim \eta_F$ ↑ $\sigma \sim \exp(-\eta_F)$ ↓

Wells: $S \sim E_F$ ↓ $\sigma \sim \exp(-E_F)$ ↑



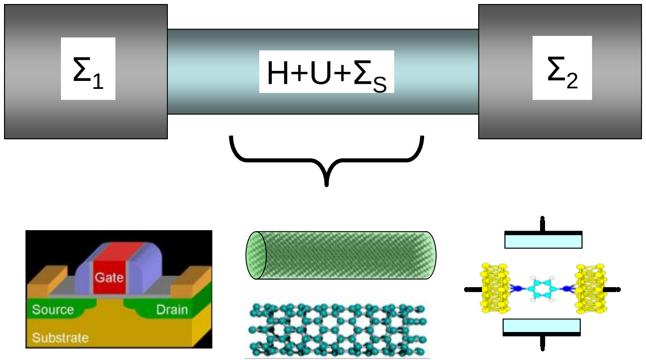


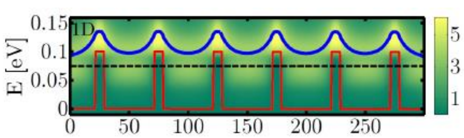
Goals of this talk

- *The Non-Equilibrium Green's Function (NEGF): Studies on superlattices and nanocomposites*

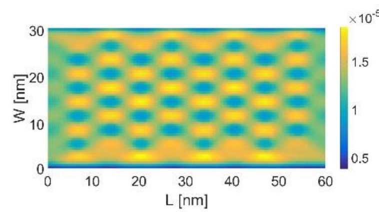
- *Expansion to multi-physics, multi-scale capabilities*

Non-Equilibrium Green's Function (NEGF)

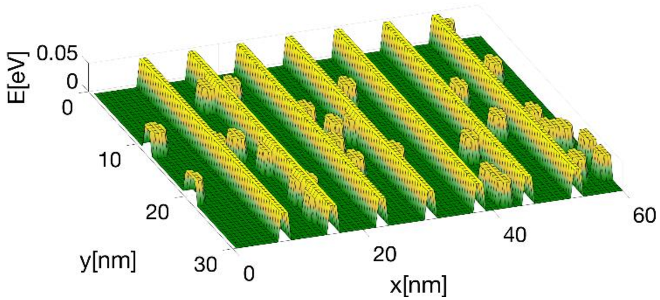




superlattices

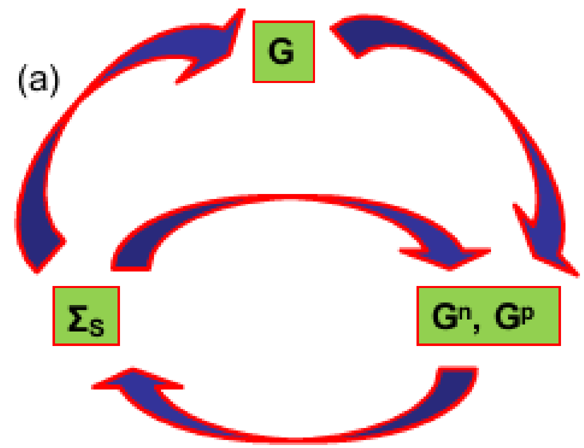


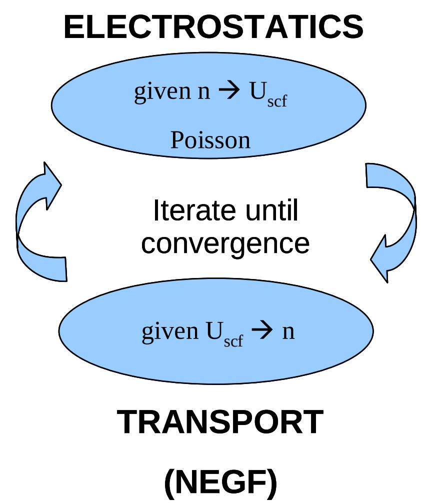
nano-inclusions

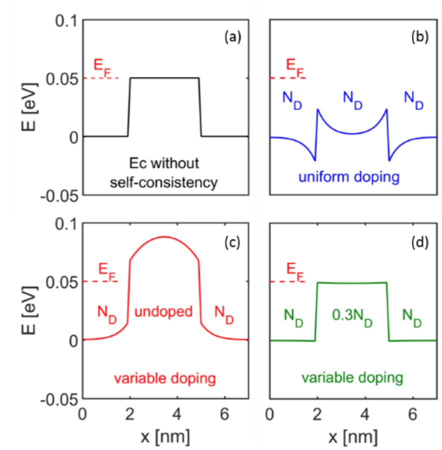


SLs + nano-inclusions

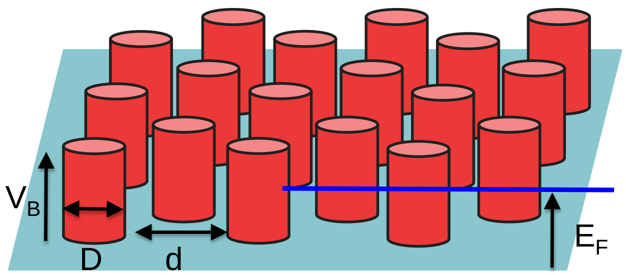
– Self-consistency

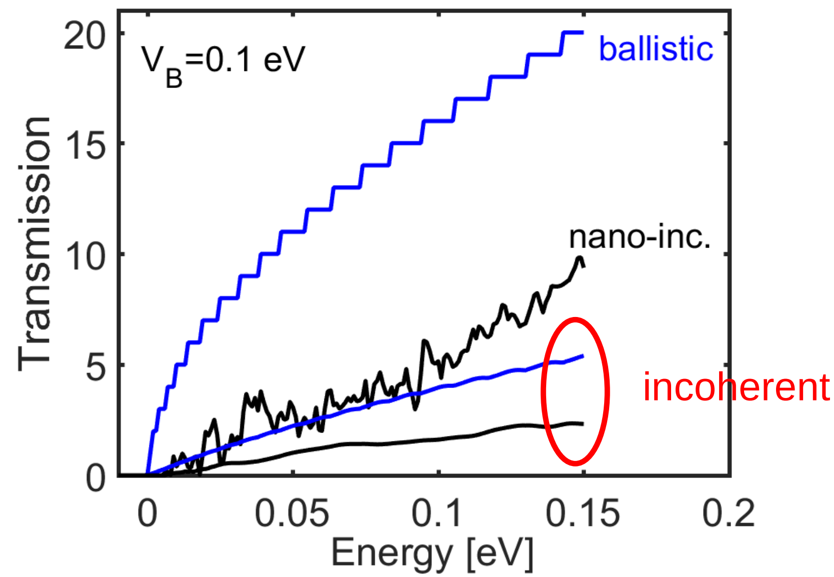






NEGF basic results: coherent vs incoherent





Coherent transport:

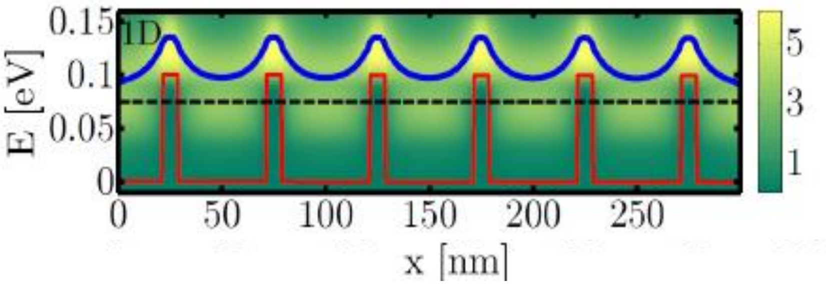
- Usually NOT appropriate – can lead to localization (at room T)
- PF is limited by the *G* of the barrier region – worst case path

Incoherent transport:

- Smoothened resonances
- The different regions can be decoupled

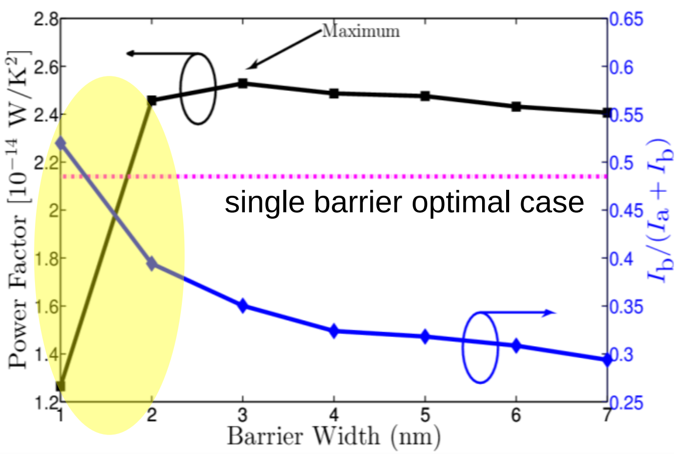
Foster, Thesberg, Neophytou, ***Phys. Rev. B***, 96, 195425, 2017

Example 1: Superlattice - all features captured

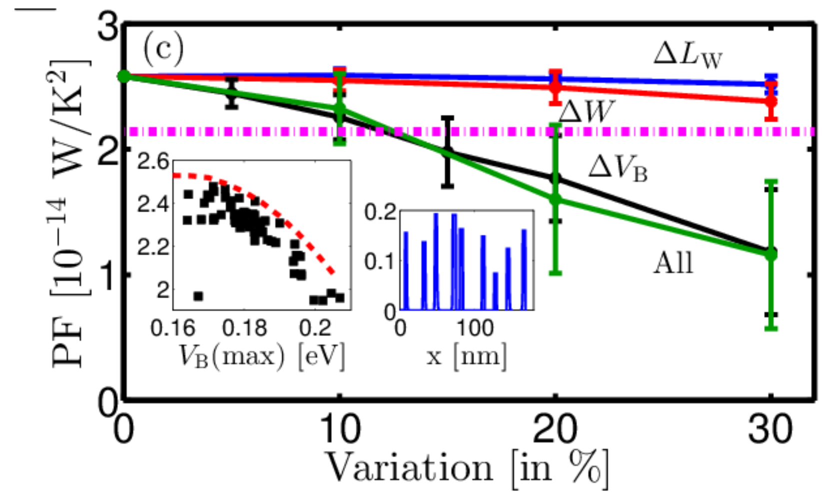


$$S = -\frac{k_B}{q}\left\langle \frac{E - E_F}{k_B T} \right\rangle = -\frac{\langle E - E_F \rangle}{qT}$$

current flow variations and λ_E



Tunneling is detrimental to PF

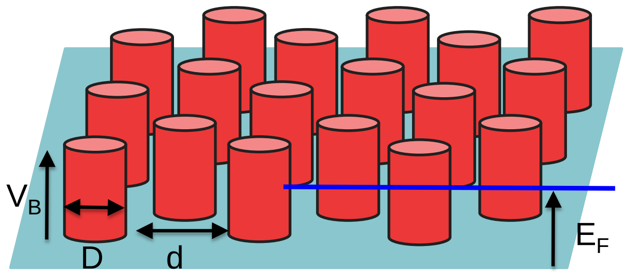


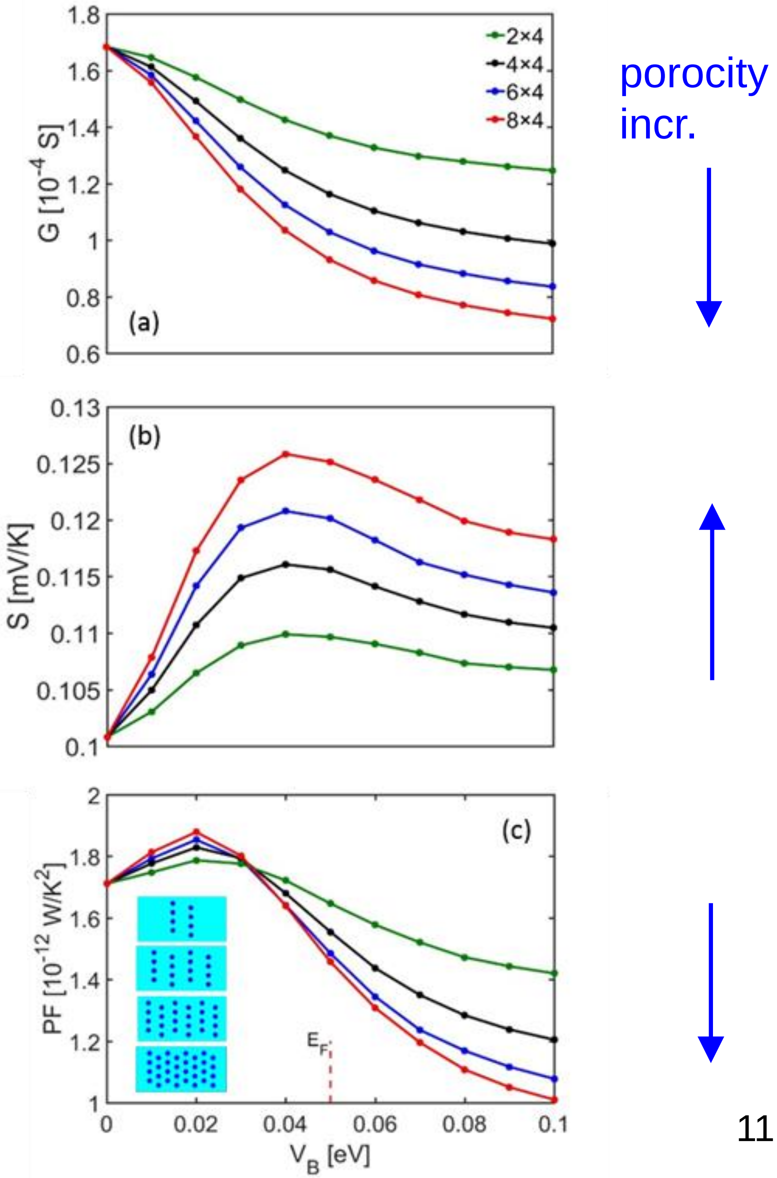
Variation in V_B reduces PF

(could explain why filtering is not yet successful?)

Thesberg, Kosina, Neophytou, ***J. Appl. Phys.***, 118, 224301, 2015

Example 2: nanoinclusions





<u>At small V_B:</u>

PF is independent of desnity

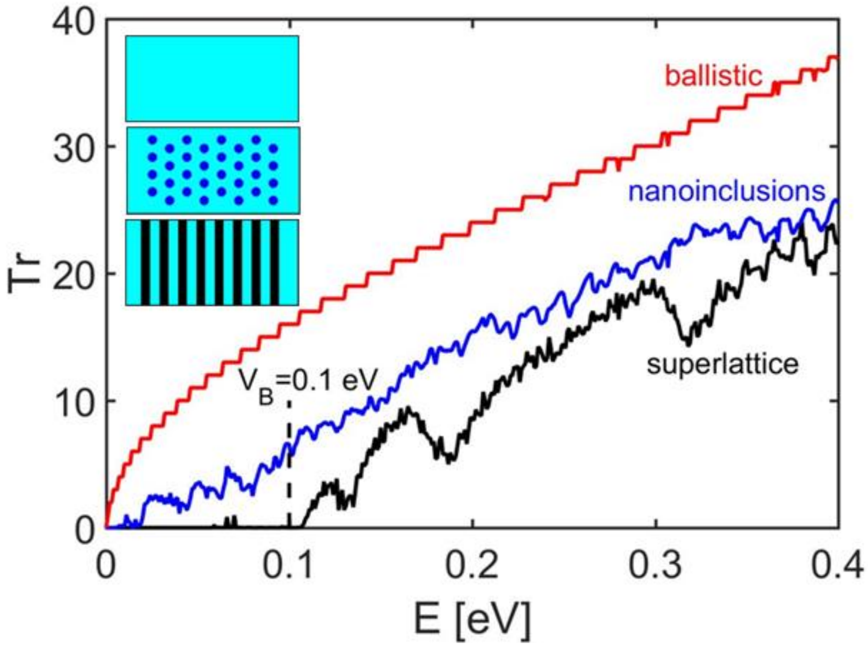
Geometry and randomness as well ?

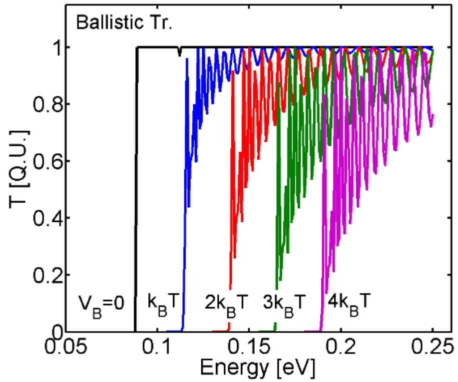
<u>At larger V_B:</u>

Density degrades the PF

Foster, Thesberg, Neophytou, ***Phys. Rev. B***, 96, 195425, 2017

Filtering using Nanoinclusions vs Superlattices



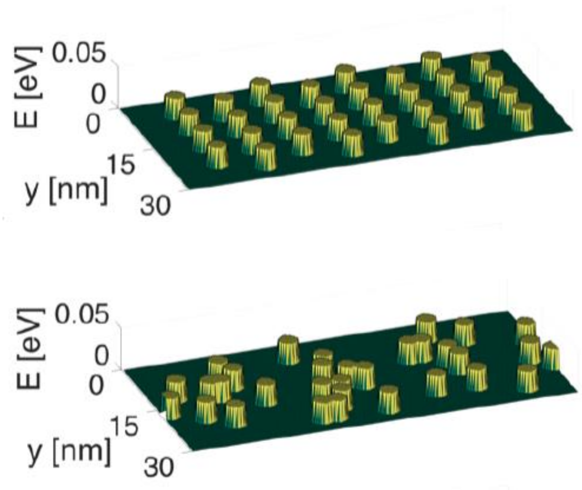


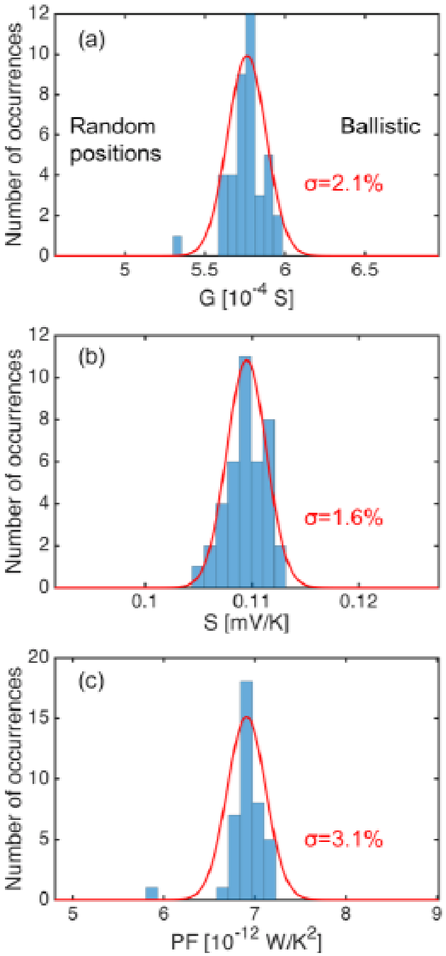
- Superlattices offer filtering capabilities
- *Nanoinclusions offer weak energy filtering capabilities (charge goes around them)*

- The transmission never recovers to the pristine value
- A simple step function transmission is not adequate

Foster, Thesberg, Neophytou, ***Phys. Rev. B***, 96, 195425, 2017

Example 3: variability in complex geometries

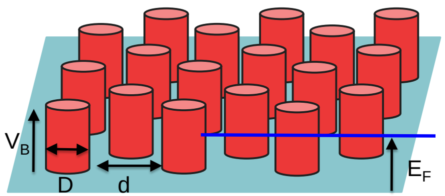


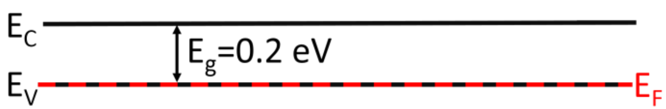


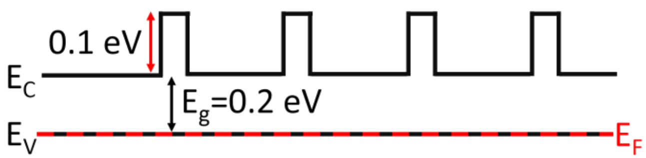
Effect of geometry variations

Vargiamidis, Foster, Neophytou, ***Physica Status Solidi A***, 2018, accepted

Example 4: Suppressing bipolar effects





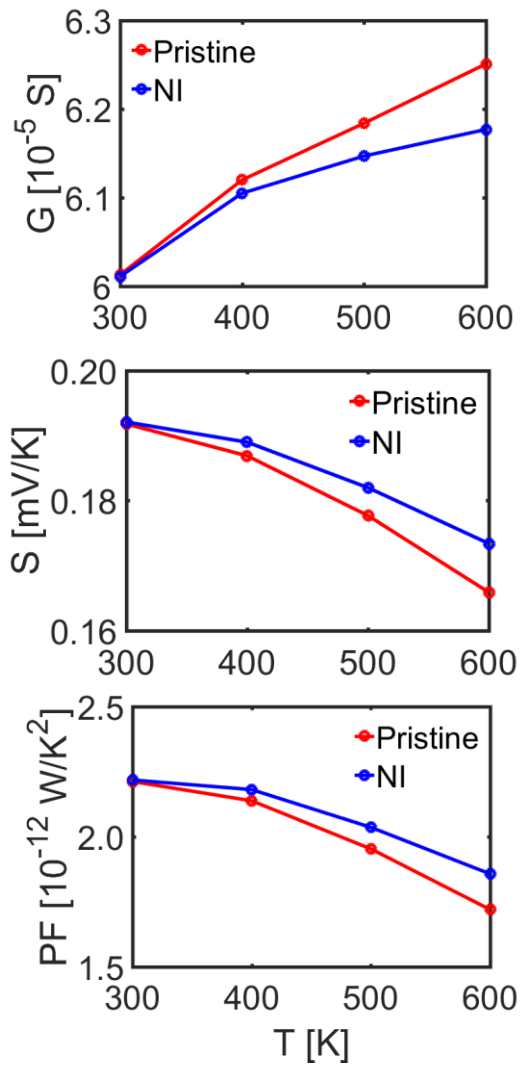


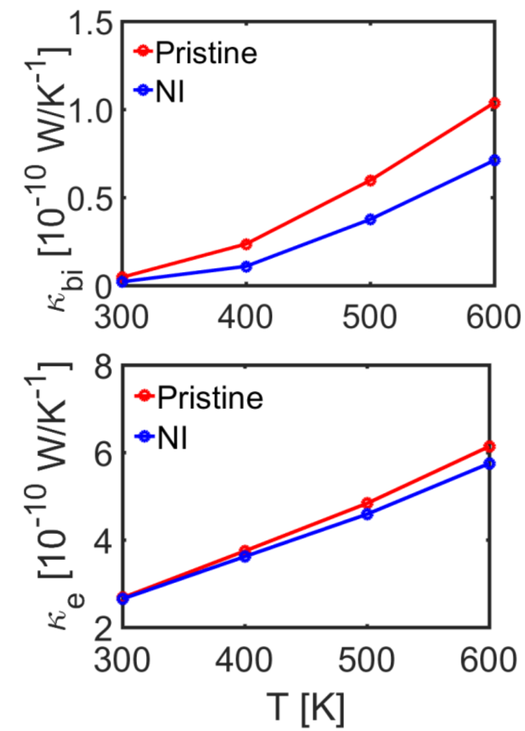
$$ZT = \frac{\sigma S^2 T}{\kappa_e + \kappa_l}$$

$$S = \frac{(G_e S_e + G_h S_h)}{(G_e + G_h)}$$

$$\kappa_{bi} = \frac{G_e G_h}{G_e + G_h}(S_e - S_h)^2 T$$

$$\kappa_{elec} = \kappa_{elec,e} + \kappa_{elec,h} + \kappa_{bi}$$





How should the barriers need to be designed for bipolar suppression?

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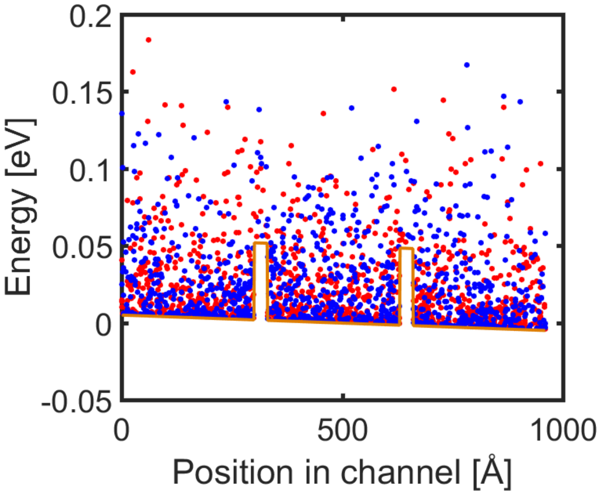
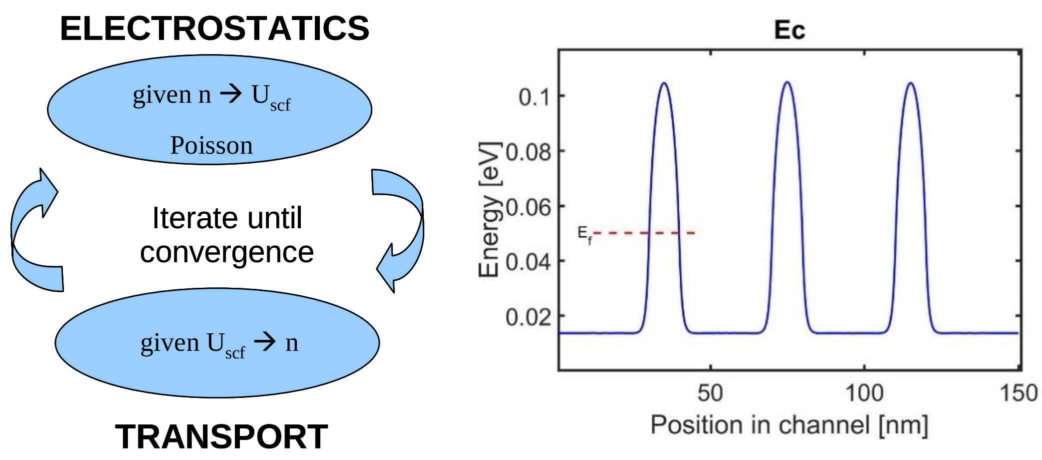
- *Expansion to multi-physics, multi-scale capabilities*

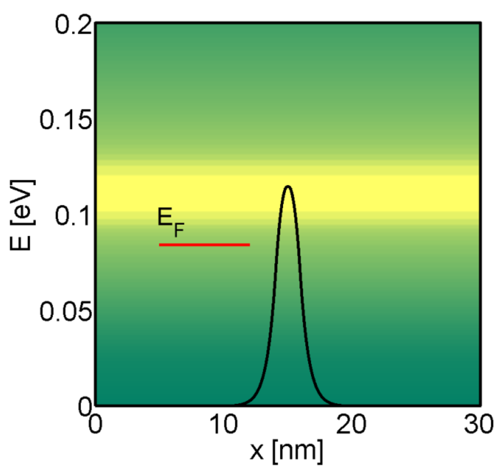
Infrastructure development – multi-physics approach



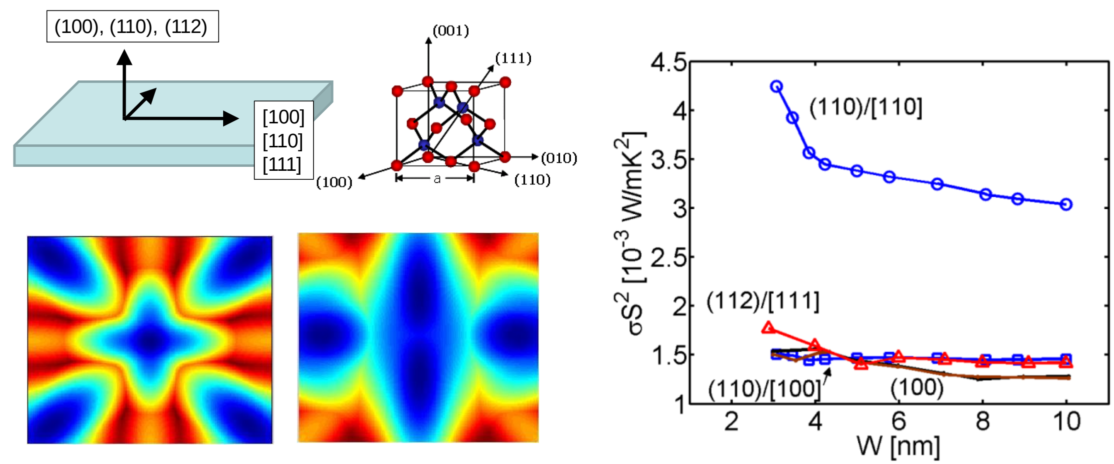
Electronic Monte Carlo



Self-consistent electrostatics



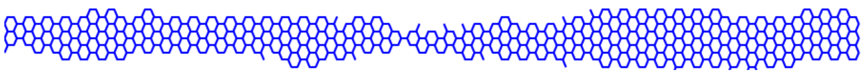
Quantum tunneling

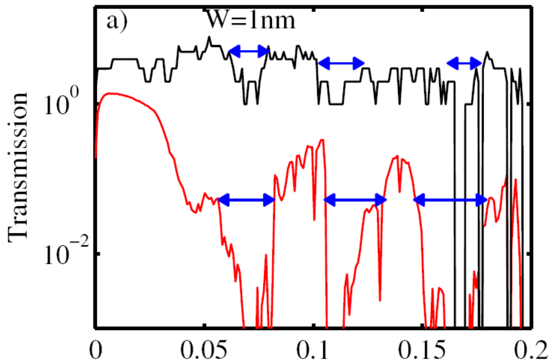


Bandstructure effects

Phonon transport treatment in nanocomposites

Phonon NEGF – coherent effects:

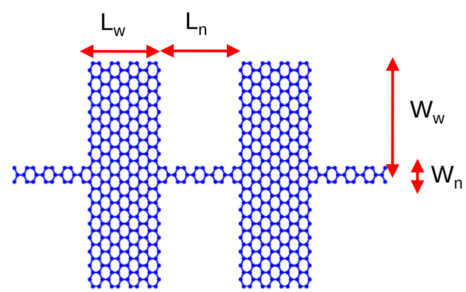




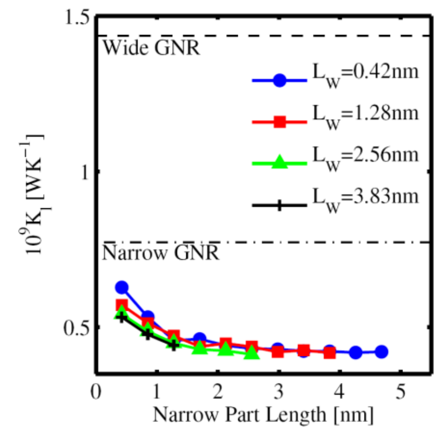
'effective transmission gaps formed'

Karamitaheri et al, ***Phys. Rev. B,*** 91, 165410, 2015

Going below the Casimir limit:

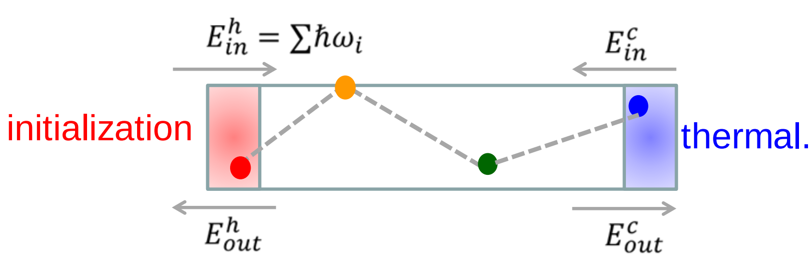


Corrugated structures



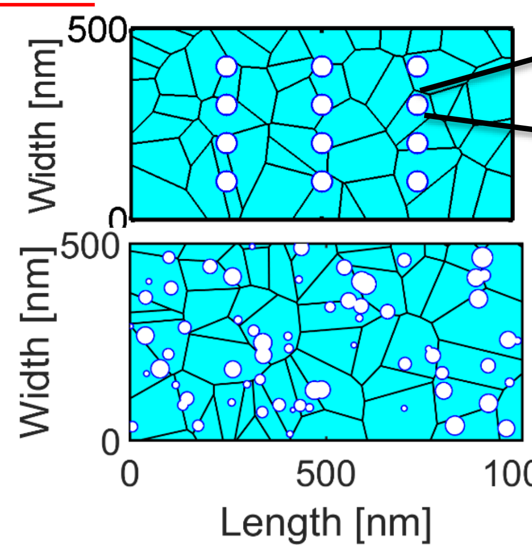
Karamitaheri et al, ***J. Appl. Phys.*** 119, 244302, 2016

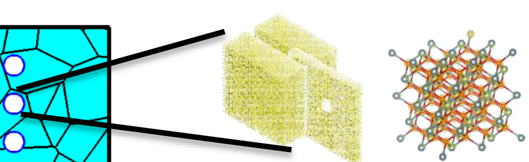
Nanoporous Si: Monte Carlo calculations:

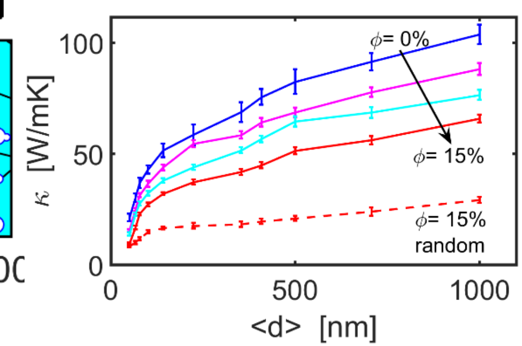


Phonon flux $$\phi = N_{tot}\frac{(E_{in}^h - E_{out}^h) - (E_{in}^c - E_{out}^c)}{N < t_{TOF} >}$$

Fourier's law $$\kappa = -\phi/\nabla T$$



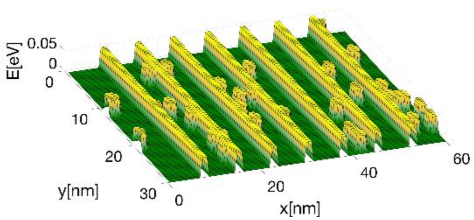


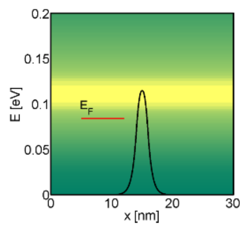


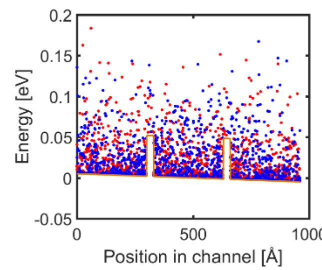
Wolf et al, ***J. Appl. Phys.***, 115, 204306, 2014

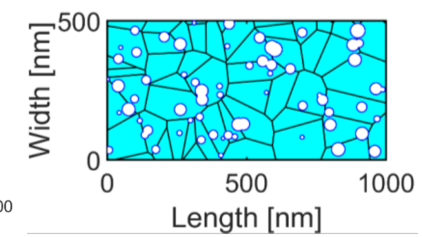
Conclusions

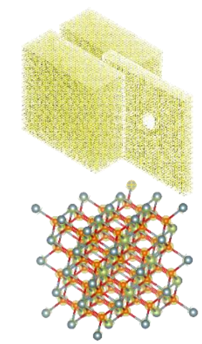
- NEGF transport in nanocomposites
- Importance of multi-physics, multi-scale needs for TE transport











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