UNIVERSITÉ DE LORRAINE

Lemta

Monte Carlo modeling of phonon transport in nanostructures

David LACROIX

Outline

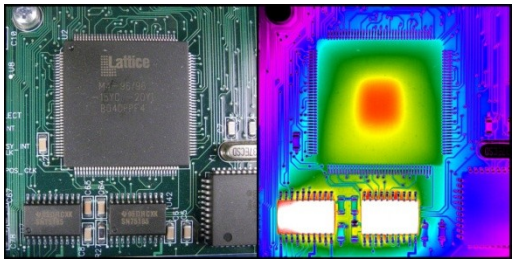
- Heat transport in nano & microstructures, basics, models and challenges
- Using Monte Carlo technique to solve the Boltzmann transport equation for phonons
- Application of MC-BTE tool to appraise thermal properties in nanostructures
- Improvement of MC-BTE by coupling with ab-initio calculations
- Summary and perspectives

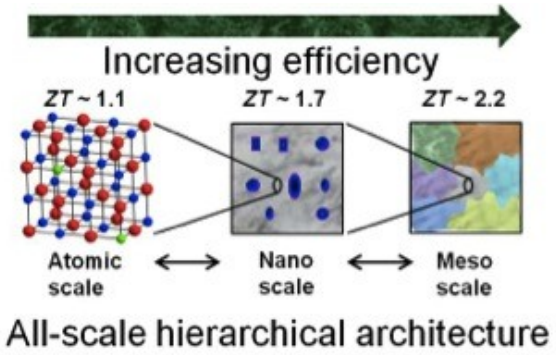
Heat transport in nano & microstructures, basics, models and challenges

Tailoring heat transport in nano & microstructures

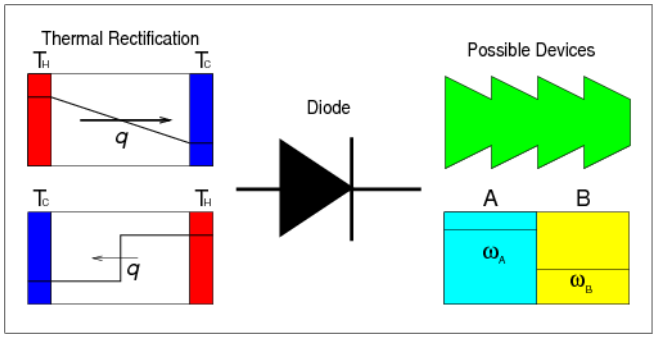
Challenges

- **Control of overheating**
  - minimize failure,
  - hotspots occurrence
  - improve performance of materials
  - etc
- **Tailoring heat transport properties**
  - thermoelectric material improvement
  - thermal cloaking, thermal rectification
  - etc
- **Thermal management applications**
  - electronic, optical, optoelectronic and thermoelectric devices,
  - thermal diode
  - etc



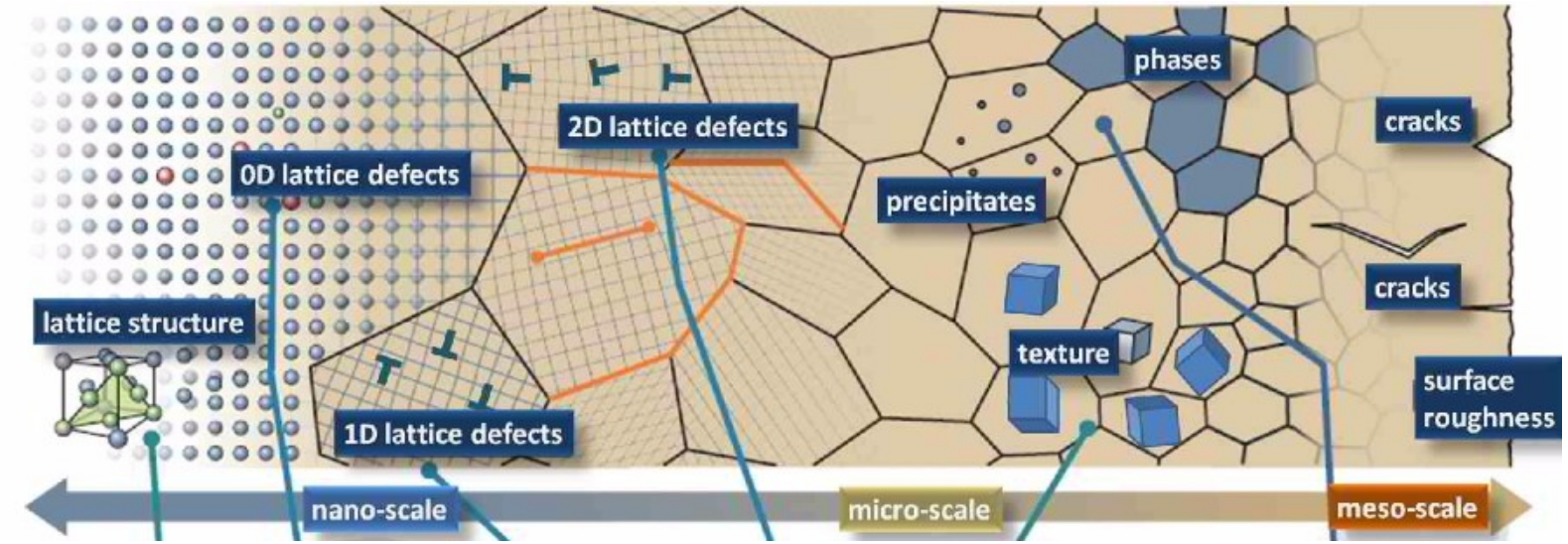


Nature 489, 414–418 (2012)



http://telab.vuse.vanderbilt.edu/research.html

Heat transport modeling, a multiscale issue



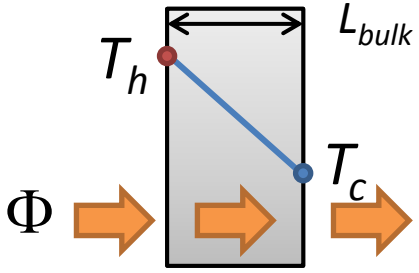
http://www.dierk-raabe.com/

DFT	MD	MC	FEM, FVM
few atoms & electrons	thousands of atoms	crystals	structures
~ 1nm	10~100nm	100nm ~ 100µm	100µm ~ above

Heat transport in nano & microstructures

Basics

- Heat transfer at the nanoscale differs from what occur in bulk material:
  - Fourier's law needs to be considered with caution
  - Heat transport equation is no longer valid
  - Thermal properties of materials depend on **length scales** and temperature
- Heat transport varies between diffusive and ballistic regimes



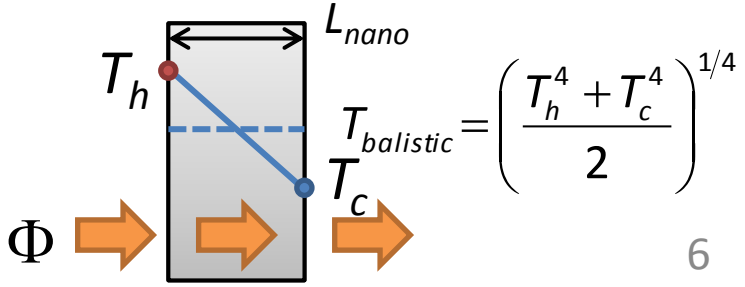
$$\Phi = -k\nabla T$$

$$\rho C \frac{dT}{dt} = \nabla.(k\nabla T) + q_{vol}$$

k_{nano} ???

dT/dt ???

Approach based on heat carriers transport: "phonons"

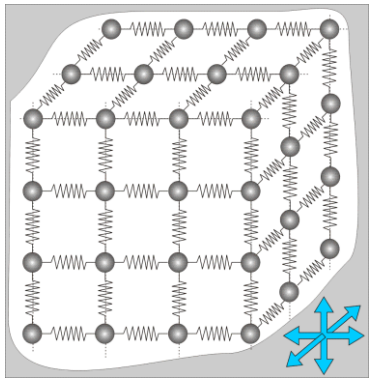


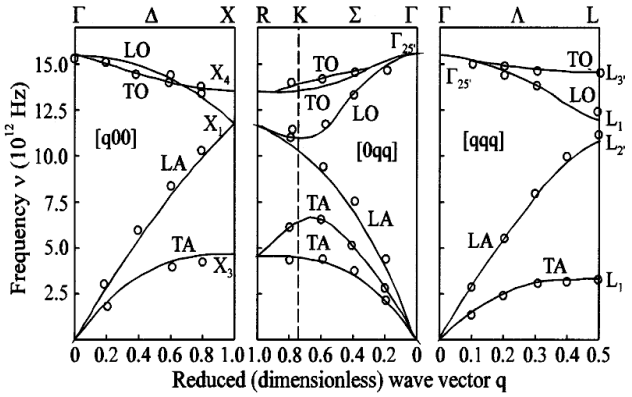
Phonon and heat propagation

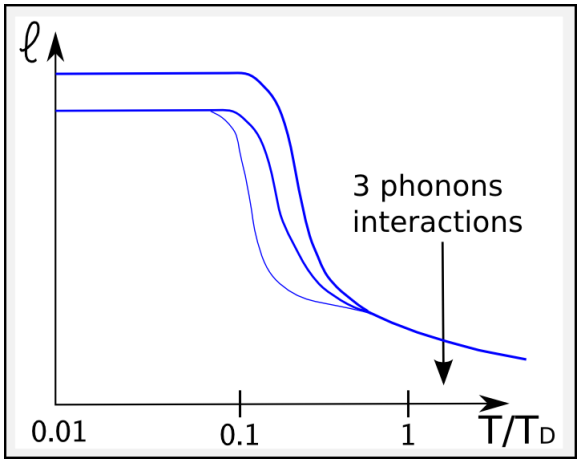
In semiconductors “phonons” are quasiparticles that characterize the vibrational motions of a lattice. They propagate heat and can be either considered as wave or particles.

$\Lambda \equiv \ell$: phonon mean free path

Vibration ↔ Dispersion relations



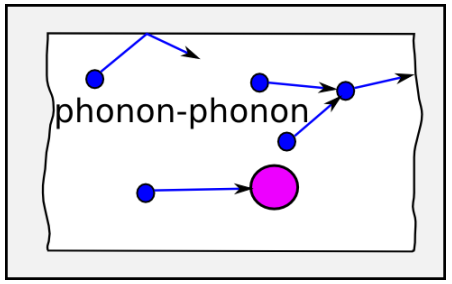




Kinetic model for thermal conductivity:

$$k = \frac{1}{3}\rho C v_g \Lambda \quad ; \quad \Lambda = \frac{v_g}{\tau}$$

Scattering mechanisms



Boltzmann transport equation for phonons

Phonons obey to the **Boltzmann Transport Equation (BTE)**. When considered as particles their motion and interactions (scattering) in nanostructures depends on: temperature, dispersion properties and scattering lifetimes of the considered material.

The BTE is solved in the frame of the relaxation time approximation by a Monte Carlo method.

BTE

$$\frac{\partial \mathrm{f}}{\partial t} + \nabla_{\mathrm{K}}\,\omega \cdot \nabla_{\mathrm{r}}\mathrm{f} + F \cdot \nabla_{\mathrm{p}}\mathrm{f} = \left.\frac{\partial \mathrm{f}}{\partial t}\right|_{scat}$$

Relaxation time approximation

$$\left.\frac{\partial \mathrm{f}}{\partial t}\right|_{scat} = \frac{\mathrm{f}_{\omega} - \mathrm{f}_{\omega}^{0}}{\tau(\omega)}$$

$\mathbf{f}$ is the distribution function
$\leftrightarrow$ number of phonons

$$\mathrm{N}(t,\mathrm{r}) = \frac{1}{V}\sum_{\mathrm{K}} \mathrm{f}(t,\mathrm{r},\mathrm{K})$$

Problem: $\tau(\omega)$ appraisal is not explicit. Analytic expressions can be found for some bulk materials (Si, Ge, GaN, C, etc) but littérature is poor for complex compounds

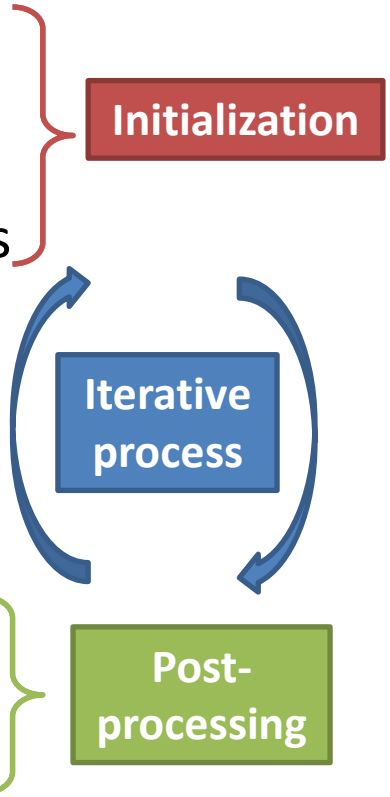
Solution? Ab-initio calculation of τ

Using Monte Carlo technique to solve the Boltzmann transport equation for phonons

Monte Carlo solution of the BTE

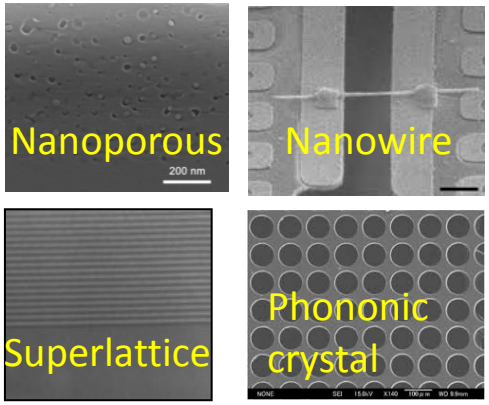
The MC solution of the BTE for phonon lies on several steps:

- Design of the nanostructure geometry and discretization
- Prescription of boundary conditions
- Initialization of the phonon state in the discretized cells
- Motion of phonon during a time step
- Scattering of phonon to restore thermodynamic equilibrium
- Calculation of local temperature and heat flux
- Derivation of thermal conductivity
- Assessment of other quantities (phonon spectrum vs mfp, phonon phase function, etc)

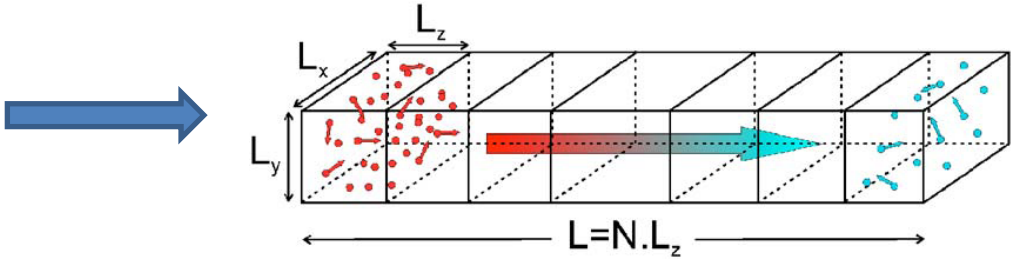


Monte Carlo solution of the BTE - initialization

Nanostructures



Structures are discretized taking into account periodicities
Temperature are prescribed in first and last cells (blackbodies)

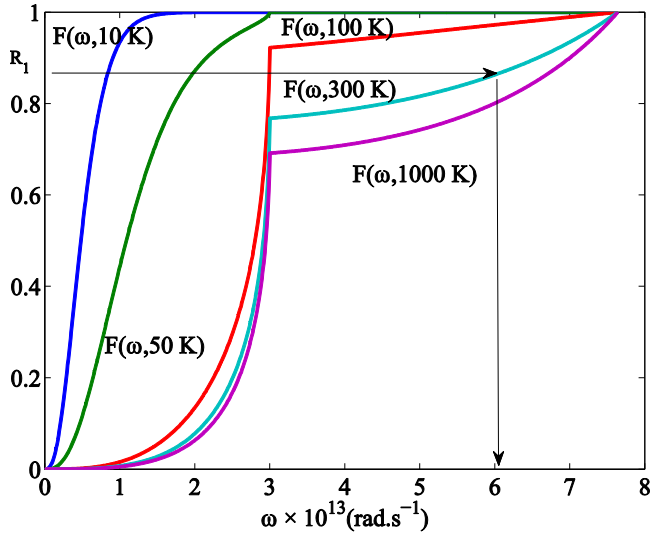


Energy within a cell

$$E = \int_{\omega} \sum_{p} \frac{1}{\exp(\hbar\omega / k_B T) - 1} D_p(\omega) g_p \hbar\omega \, d\omega$$

Cumulative distribution function

$$F(X) = \frac{\int_0^X \sum_p f^0(\omega,T) D_p(\omega) g_p \hbar\omega \, d\omega}{\int_0^{\omega max} \sum_p f^0(\omega,T) D_p(\omega) g_p \hbar\omega \, d\omega}$$



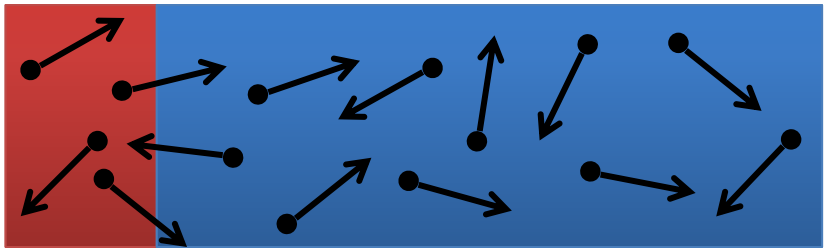
1. Sampling of a phonon population (energy bundles) at a given temperature T, according to dispersion relations (isotropic). Random location of phonons.

Monte Carlo solution of the BTE – transport and scattering

2. Follow phonon displacement according to their group velocity and boundary conditions

$$\frac{\partial \mathrm{f}}{\partial t}+\nabla_{\mathrm{K}}\omega\cdot\nabla_{\mathrm{r}}\mathrm{f}+F\cdot\nabla_{\mathrm{p}}\mathrm{f}=\left.\frac{\partial \mathrm{f}}{\partial t}\right|_{scat}$$

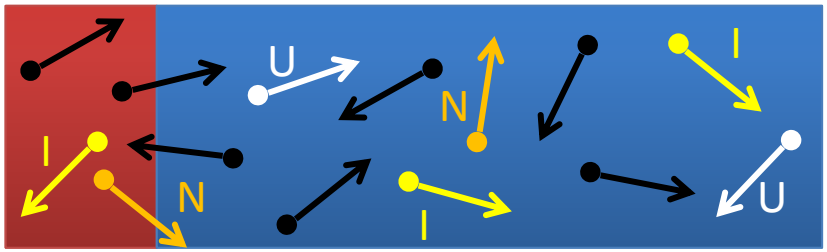
$$r(t+\delta t)=r(t)+v_g(\omega,p)\,\delta t$$



3. Proceed to phonon scattering with respect to the Matthiesen rule, calculation of E and T

$$P_{scat}=1-\exp\left[\frac{-\delta t}{\tau(\omega,p,T)}\right]$$

$$\tau(\omega,p,T)^{-1}=\sum_{process}\tau_{process}(\omega,p,T)^{-1}$$



Lifetimes of scattering processes (Normal, Umklapp, Impurity) are derived from M.G. Holland for Si and Ge

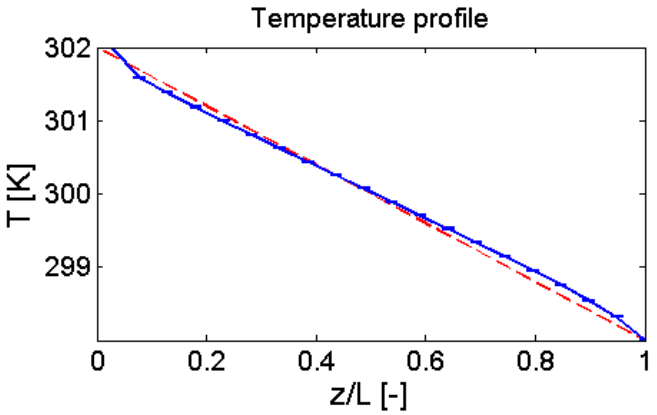
*M.G. Holland, PR **132**, 2461-2471*

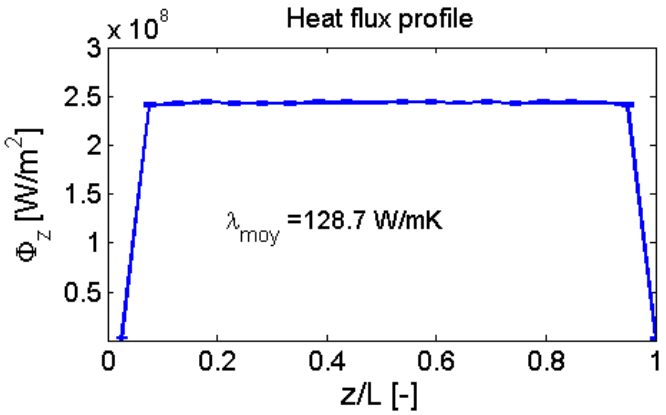
Monte Carlo solution of the BTE – post processing

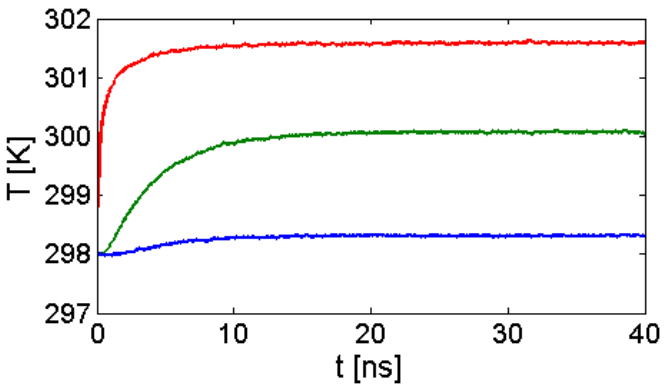
4. Extract T and Φ according to the local phonon distribution in the nanostructure

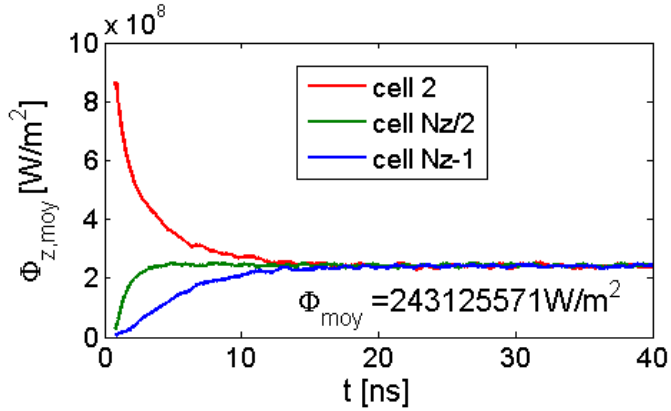
$$\Phi_z = \sum_{i=1}^{N} \frac{\hbar \omega_i V_{gz}}{V}$$

Silicon nanofilm
Lz = 2µm
δt = 1ps
Nz = 20 cells
40000 time steps
8 cores / 6 hours
Cross plane TC
k = 128.7 W/m K





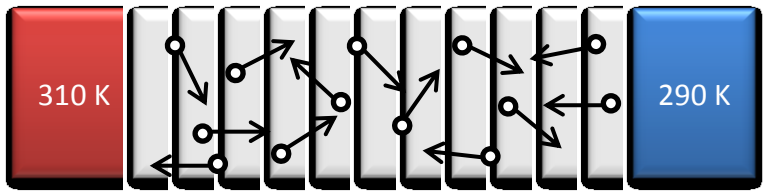




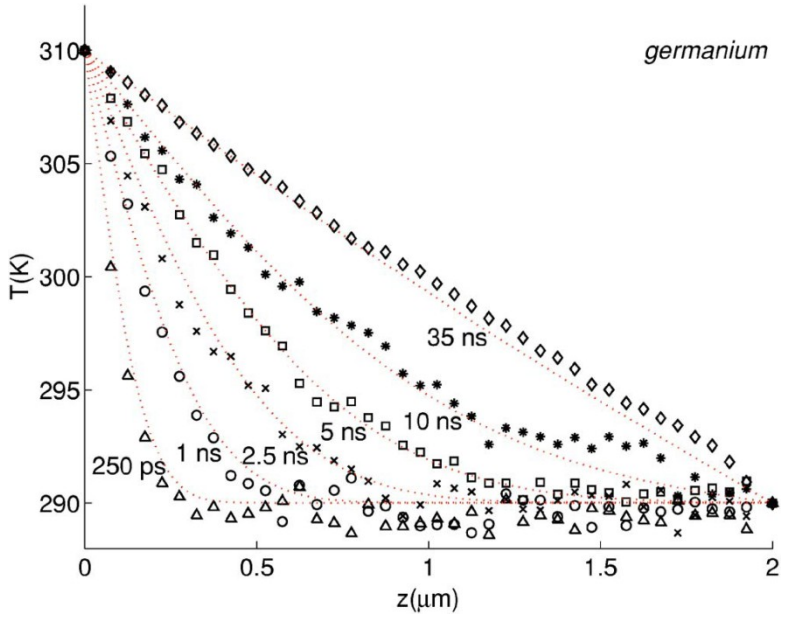
Application of MC-BTE tool to appraise thermal properties in nanostructures

Monte Carlo simulations – Nanofilms I

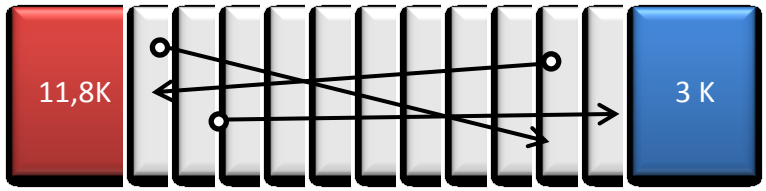
Diffuse regime



L = 2 μm

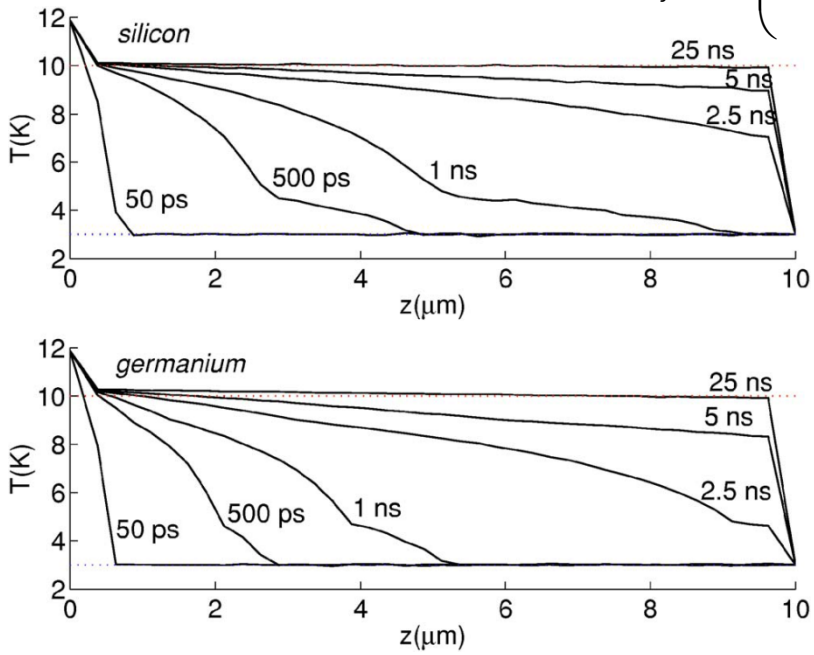


Ballistic regime



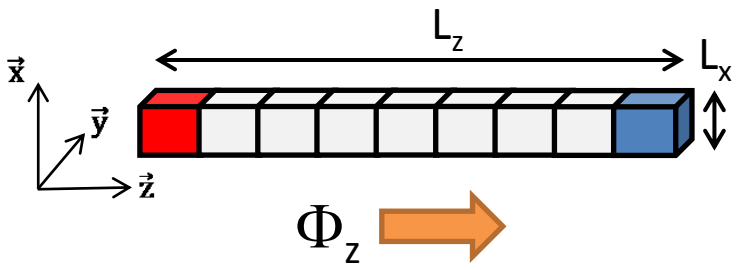
L = 10 μm

$$T_{final} = \left(\frac{T_c^4 + T_f^4}{2} \right)^{1/4}$$



*D. Lacroix, PRB **72**, 064305*

Monte Carlo simulations – Nanofilms II



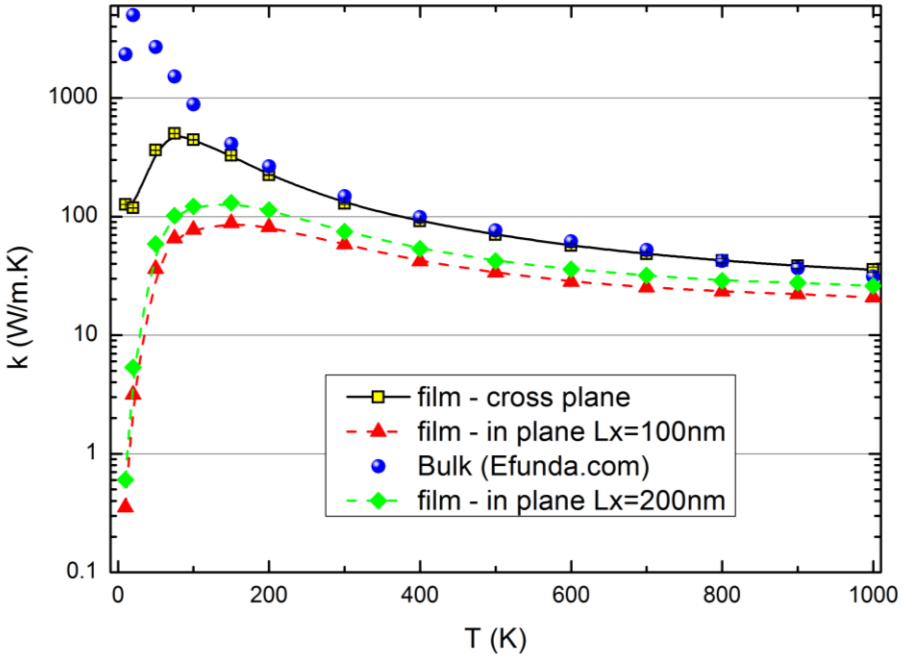
Rx, Ry: reflections
D: diffuse
S: Specular

Wire: Rx = D, Ry = D

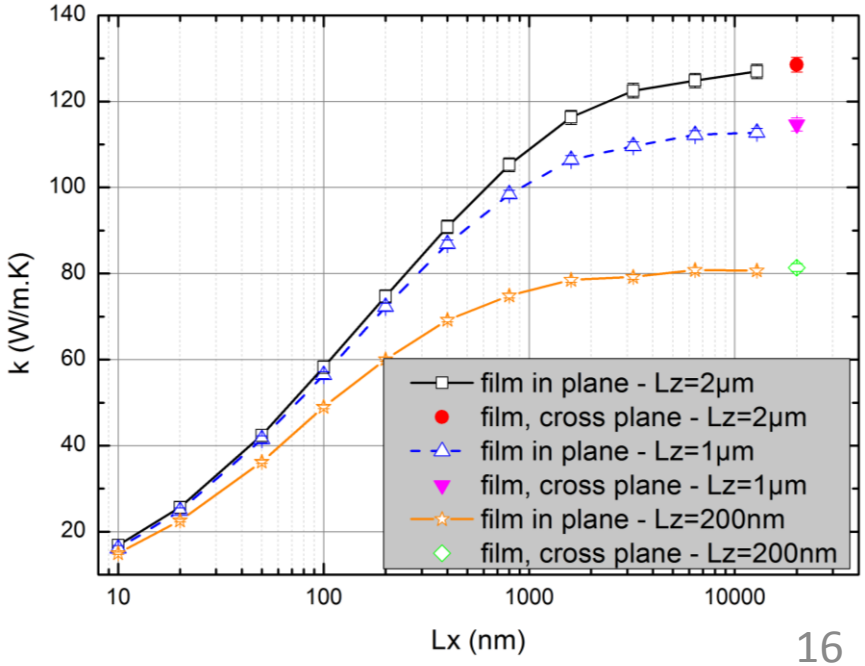
Film In-plane: Rx = D, Ry = S

Bulk /film Cross-plane: Rx = S, Ry = S

Thermal conductivity in Si vs T; Lz = 2µm

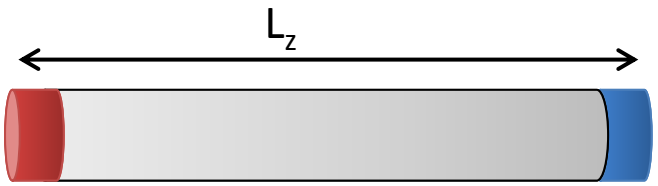


In-plane Thermal conductivity in Si film vs Lx

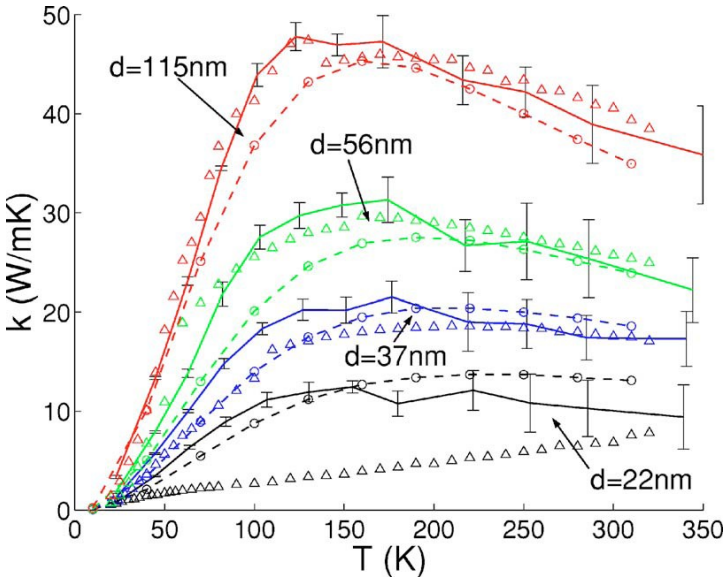


Monte Carlo simulations – Nanowires I

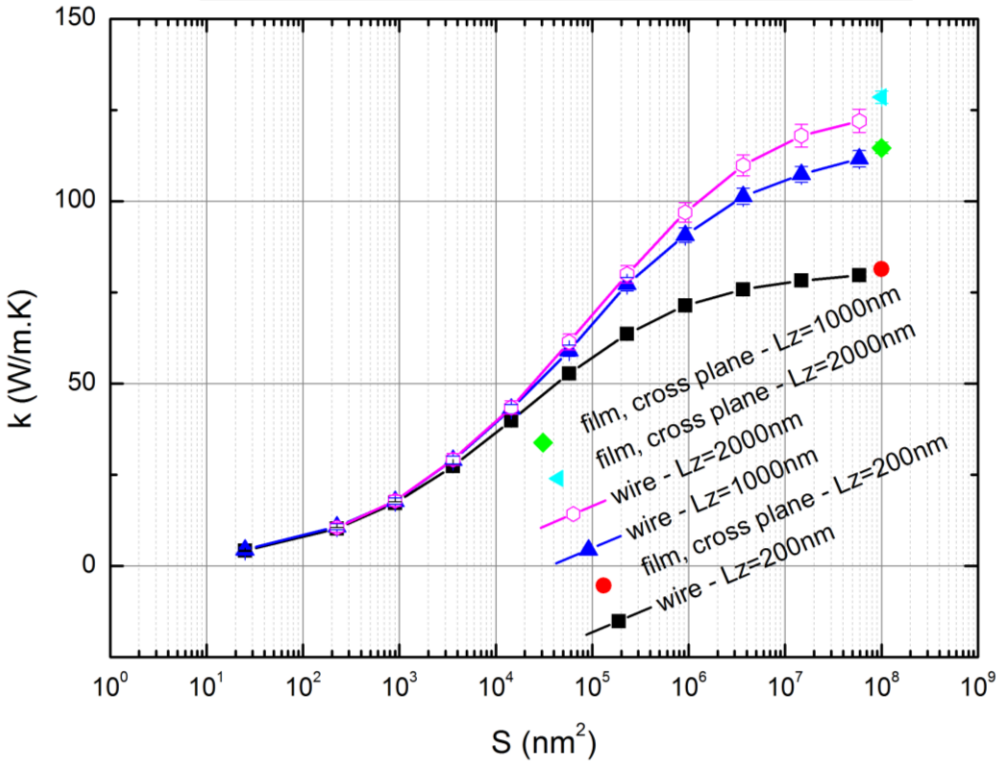
Smooth nanowires



Si nanowire TC vs T



Si nanowire TC vs cross-section

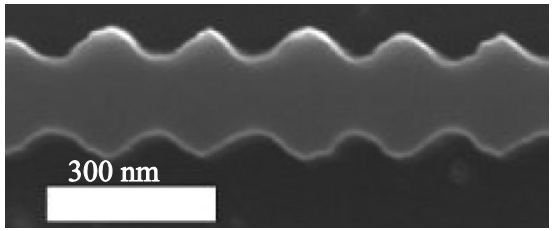


Simulations match experiments, except for very thin diameters (bulk limit)

*D. Lacroix, APL **89**, 103104*
*D. Li, APL **83**, 2934*

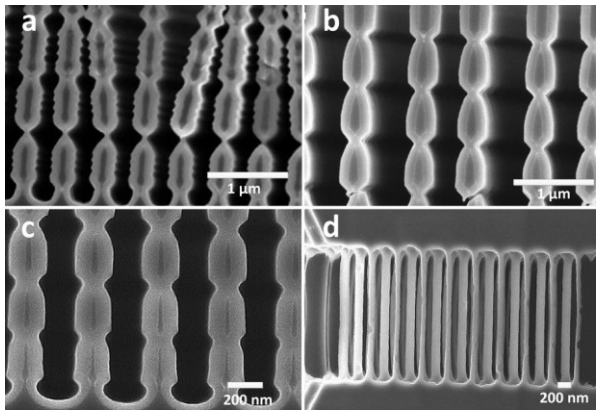
Monte Carlo simulations – Nanowires II

Corrugated and modulated nanowires



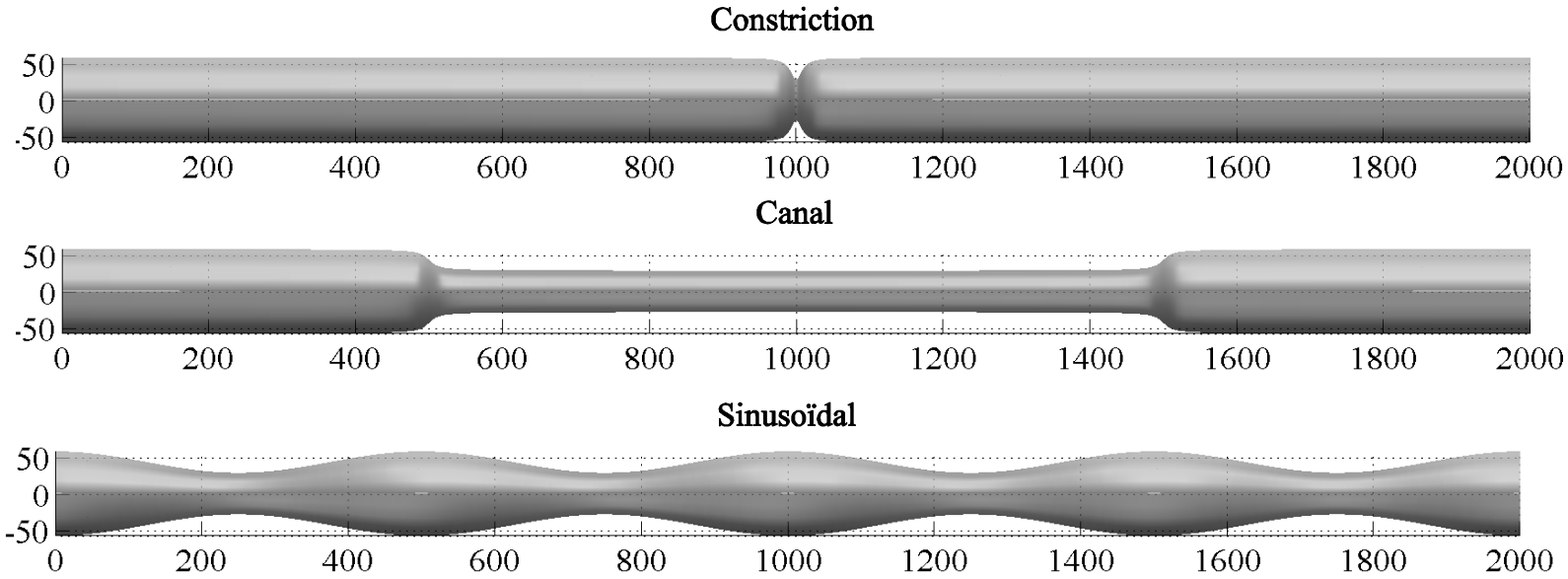
*C. Blanc, APL **103**, 043109*

Si Nanowires with shape modulation



*E. Buitrago, Microelectronic Engineering **97**, 345–348*

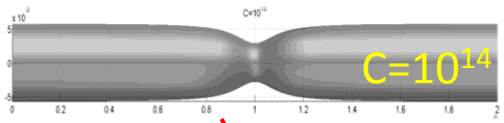
MC design of modulated nanowires

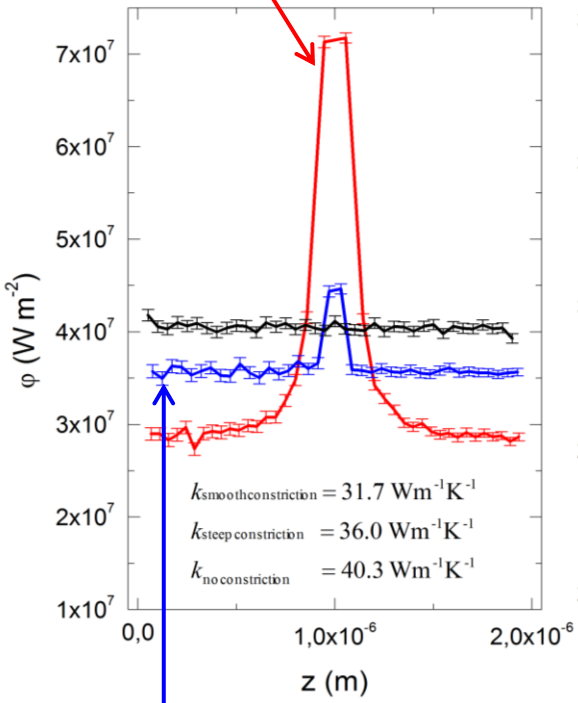


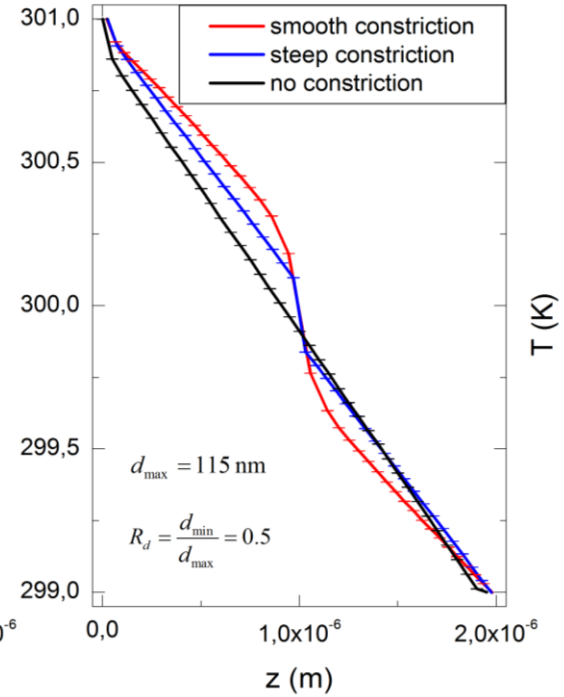
Monte Carlo simulations – Nanowires III

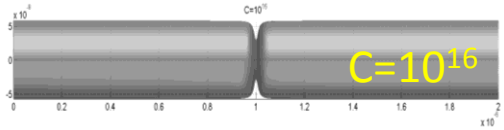
Smooth and steep constrictions

$$d(z) = d_{\max}\left[1 - \frac{(1-R_d)}{1+C(z-z_0)^2}\right] \quad ; \quad R_d = \frac{d_{\min}}{d_{\max}}$$

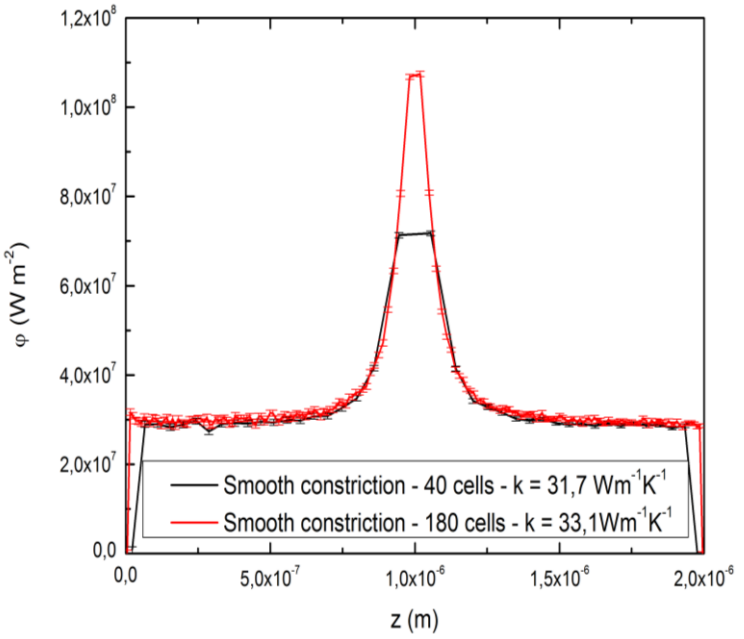








Spatial discretization impact

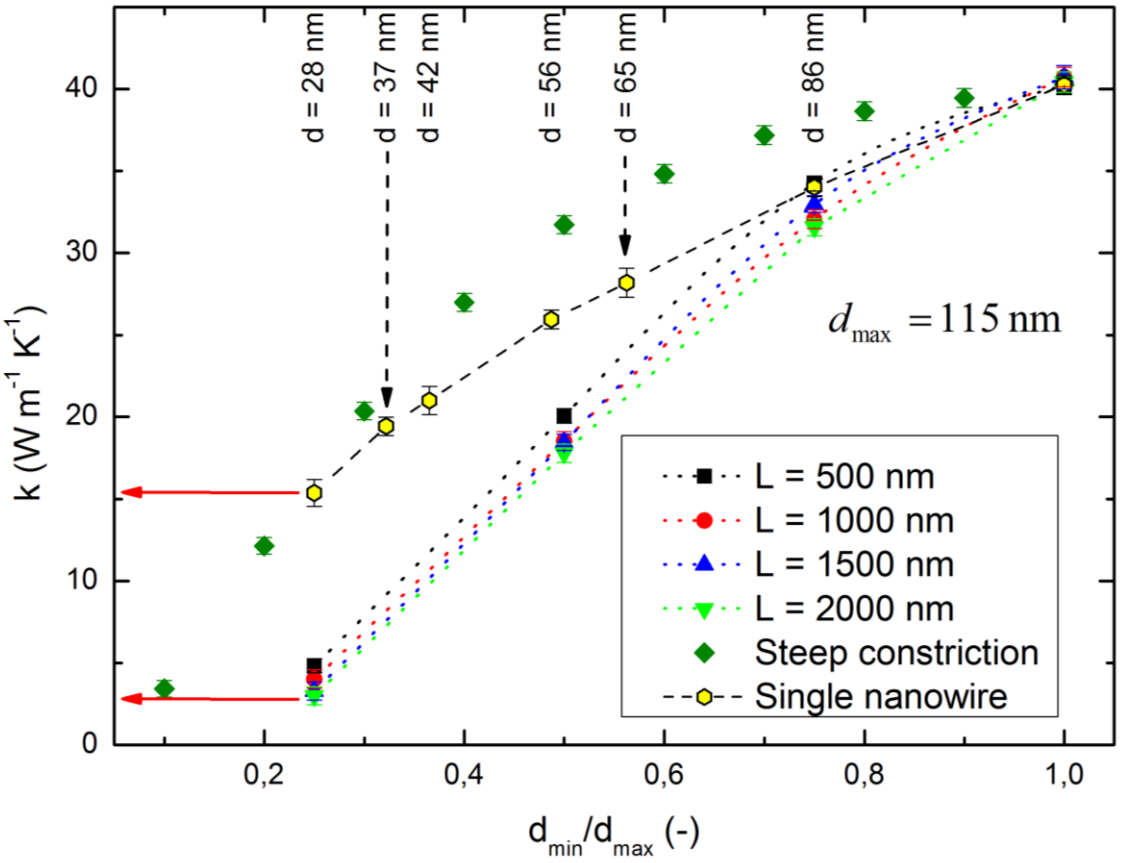


$k_{smooth} < k_{steep}$ ⇒ importance of Λ_b

*V. Jean, IJHMT **86**, 648-645*

Monte Carlo simulations – Nanowires IV

Long constriction 'canal shape'

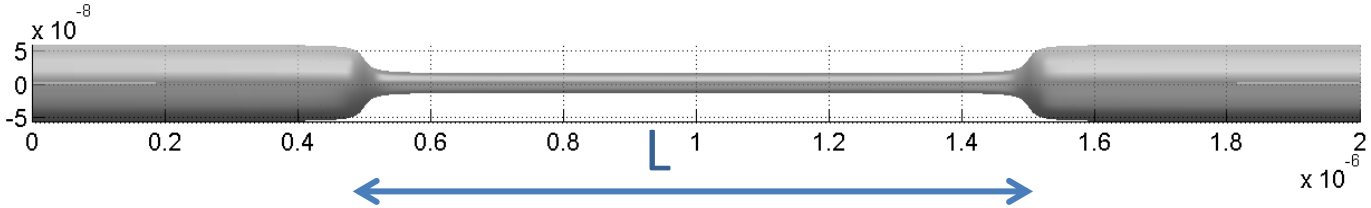


TC in canal NW smaller than in single NW with the minimal diameter

No significant effect of 'canal' length,

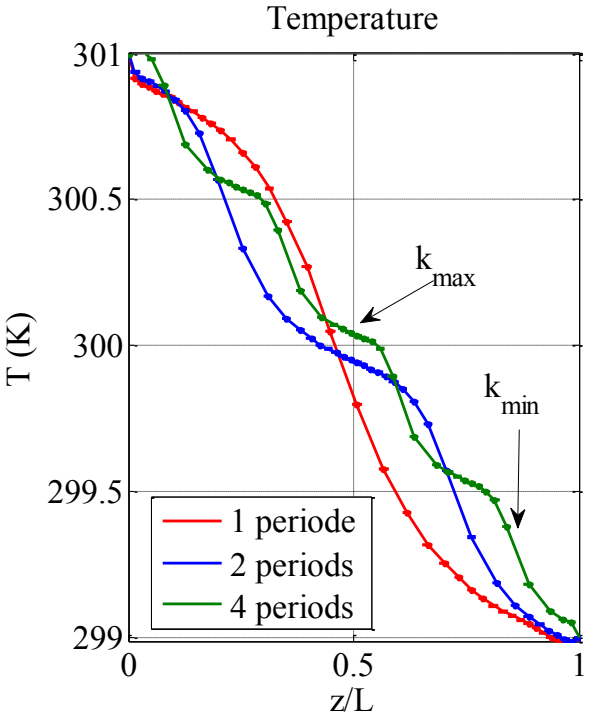
Constriction resistance

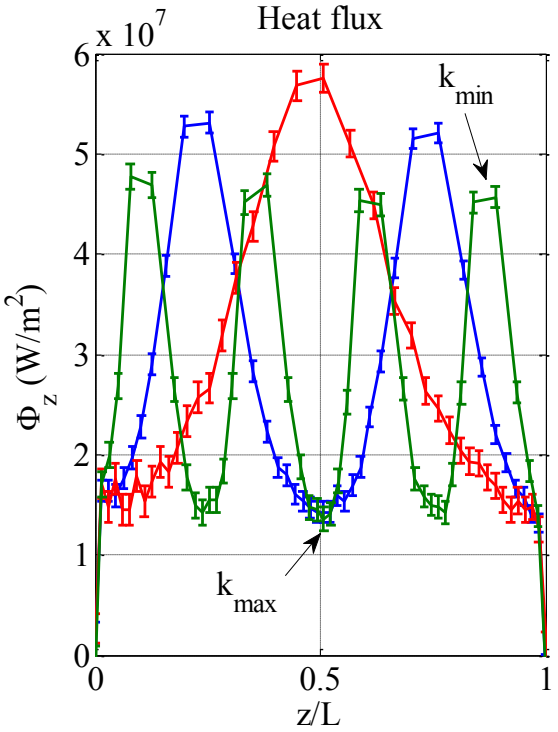
*V. Jean, IJHMT **86**, 648-645*

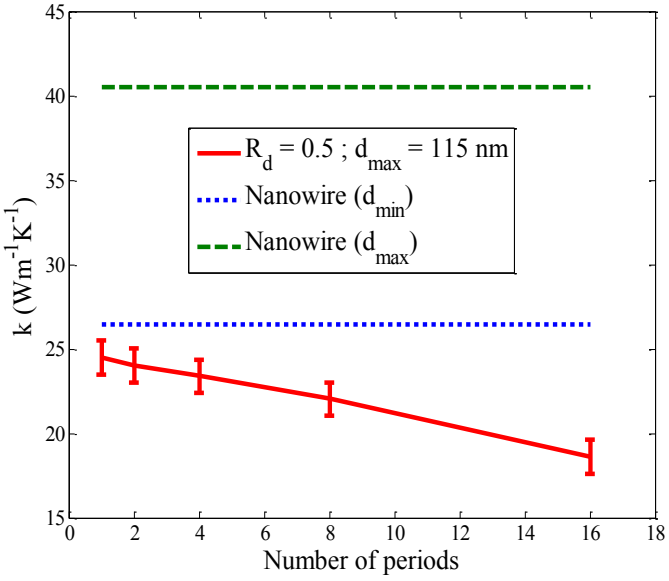


Monte Carlo simulations – Nanowires V

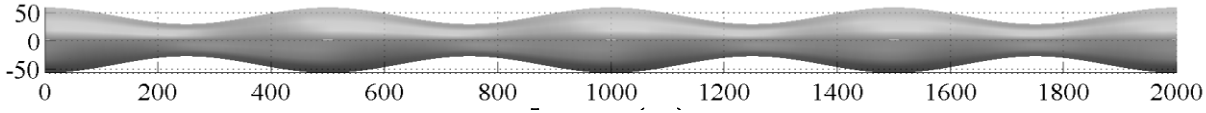
Modulated nanowire







4 periods nanowire

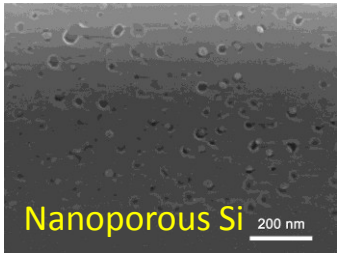


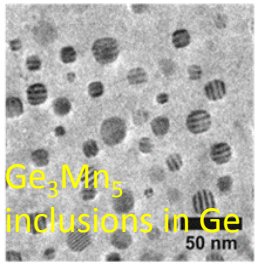
Increase of period number leads to a decrease of the thermal conductivity

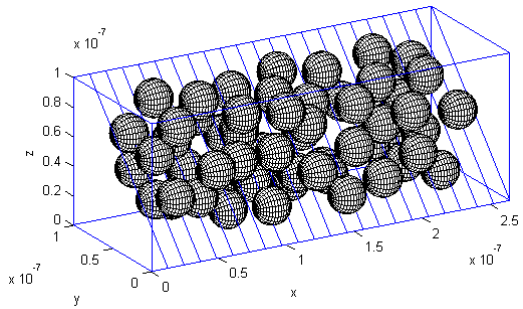
*V. Jean, IJHMT **86**, 648-645*

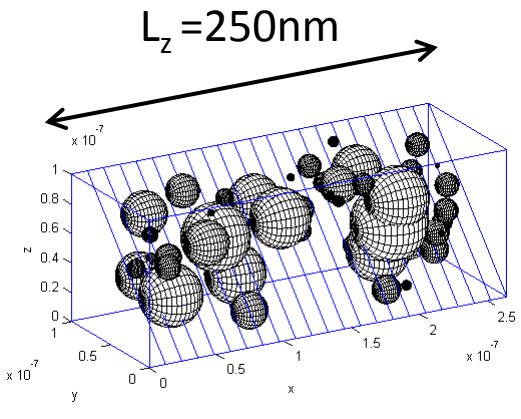
Monte Carlo simulations – Porous membranes

Porous membranes/membranes with inclusions



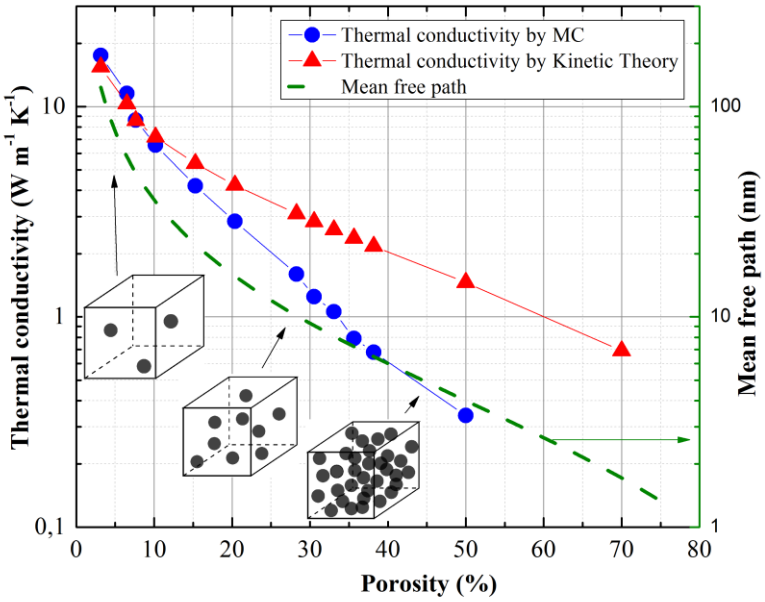




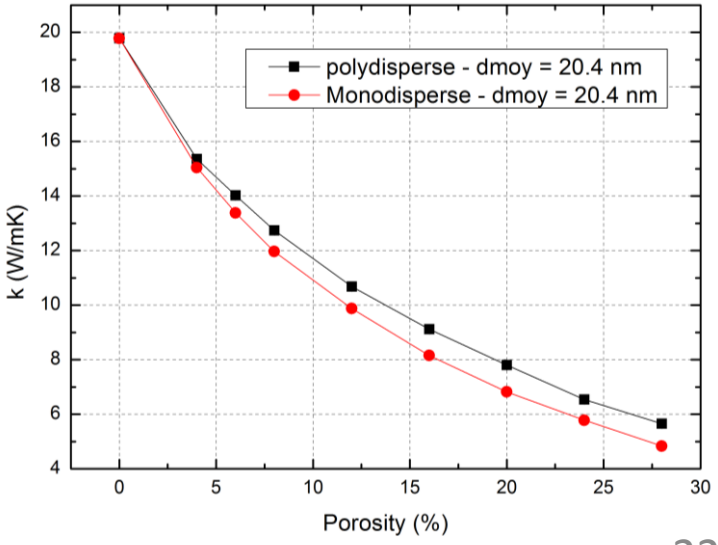


Monodisperse inclusions

Polydisperse inclusions



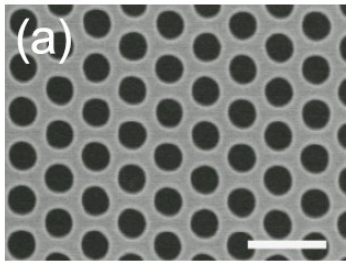
*V. Jean, JAP **115**, 024304*

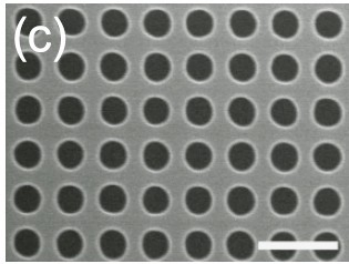


ANR Grant - Mesophon

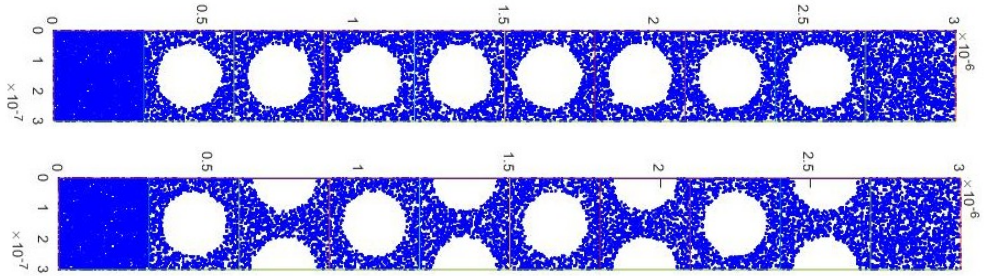
Monte Carlo simulations – PnC membranes I

Phononic (PnC) Si membranes

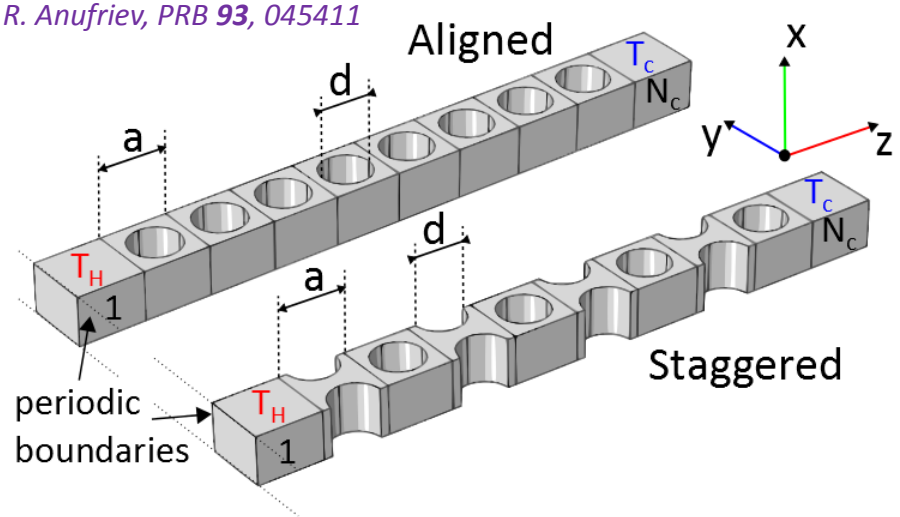


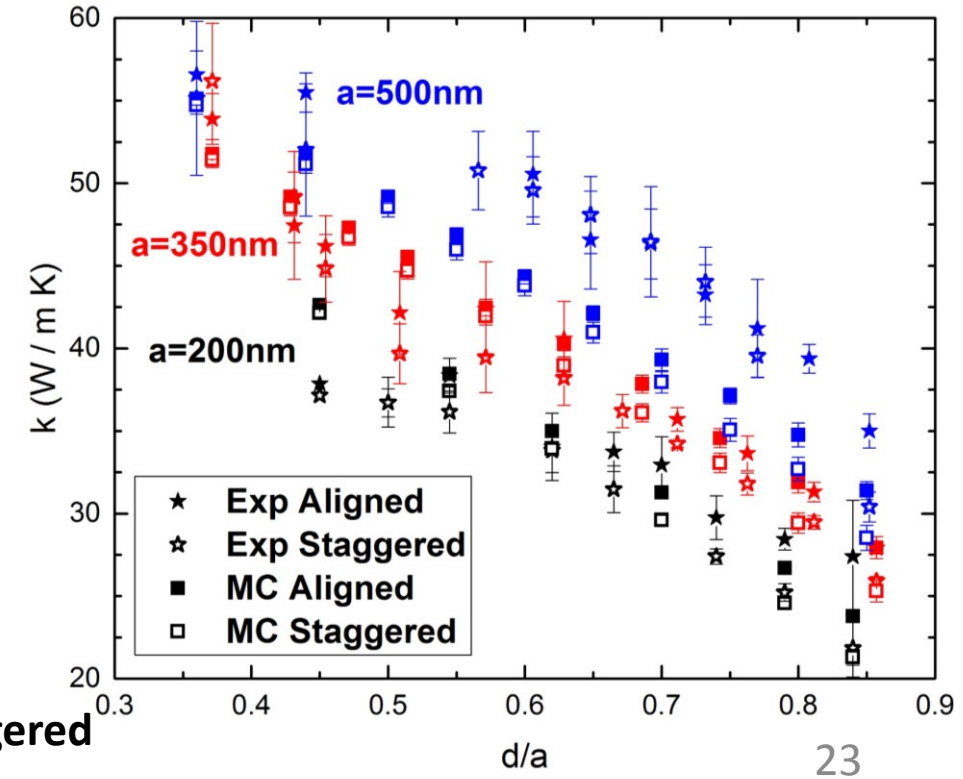


'Staggered' 'Aligned'



*R. Anufriev, PRB **93**, 045411*





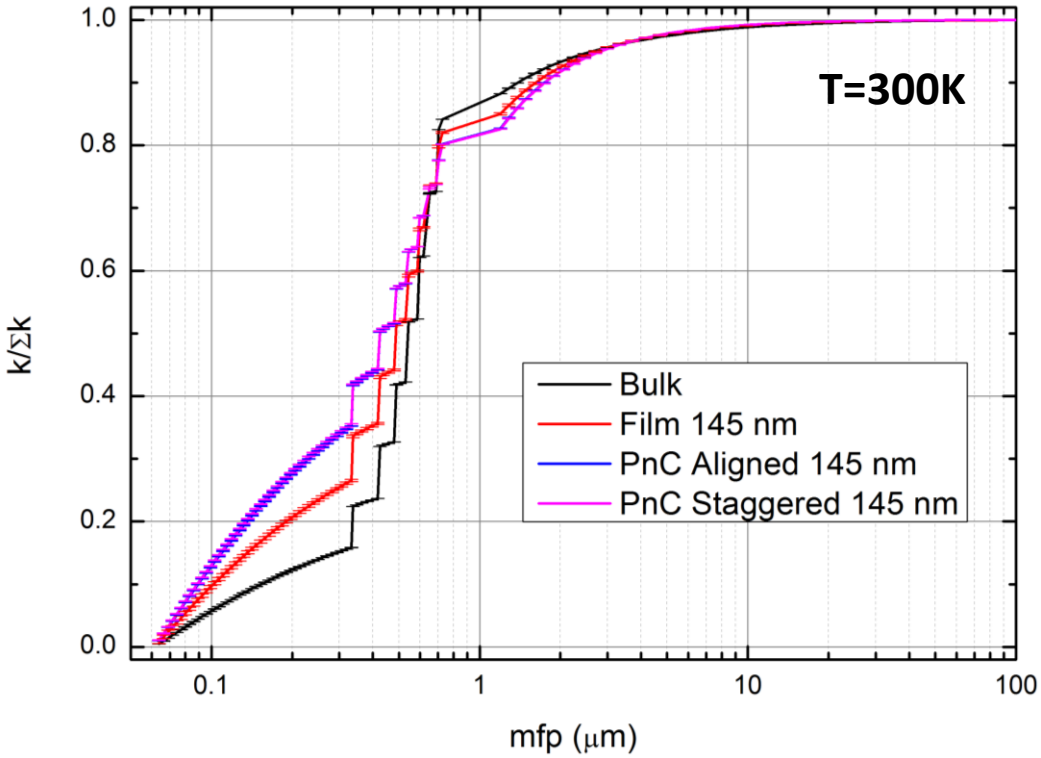
Good agreement between experiments & simulations

The TC is always lower when pores are in staggered configuration for a same S/V ratio

Monte Carlo simulations – PnC membranes II

Accumulated TC in PnC Si membranes

Extract from MC the contribution discrete phonon fequencies to the TC

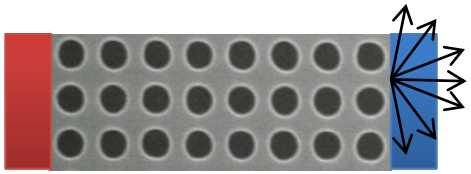


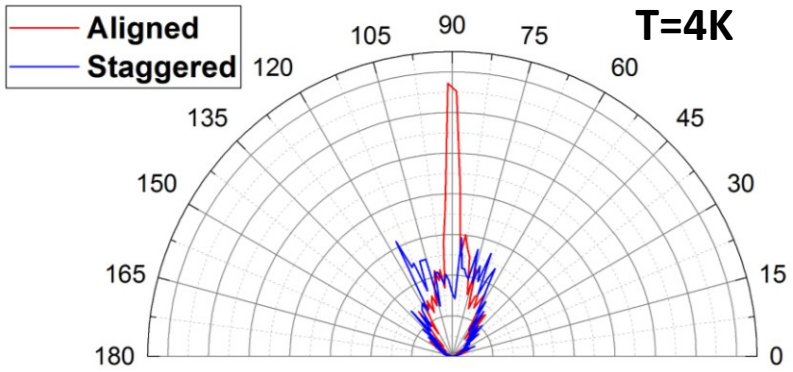
Bulk : 50% of TC due to phonon with mfp < 500nm

PnC : 65% of TC due to phonon with mfp < 500nm

Ballistic transport in PnC membranes

Distribution of the exit angle of phonons just before the cold thermostat for a=160nm and d=126nm





Some phonons experiment ballistic transport in aligned PnC

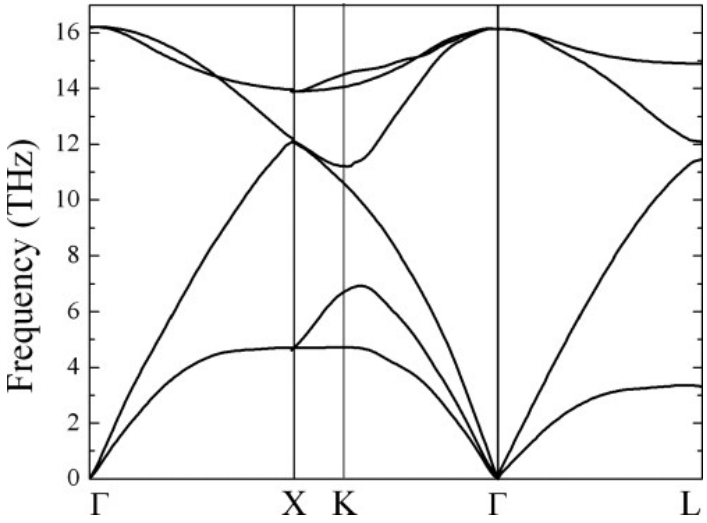
Improvement of MC-BTE by coupling with ab-initio calculations

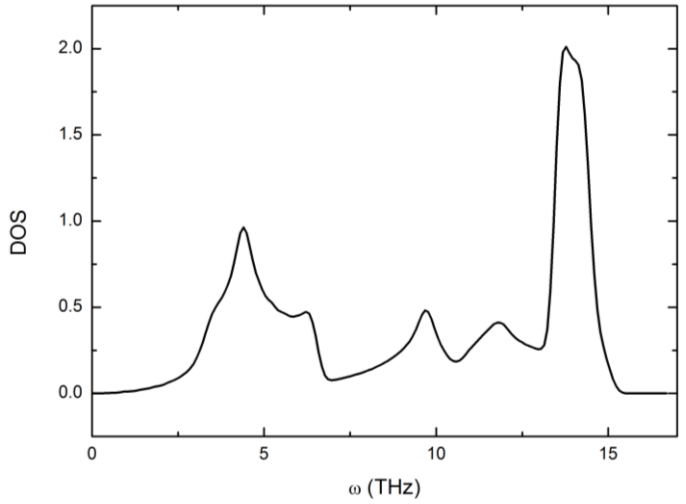
Monte Carlo simulations, ab-initio coupling I

The MC solution of the BTE for phonon has a main weakness, the necessity to have an explicit formulation of phonon lifetimes of the studied material.

- **Idea**: replace analytic lifetime expressions of basic material (Si, Ge, etc) by the one provided by DFT calculation
- Use the real dispersions properties of the material (frequency, polarization branches including optical mode, group velocities)

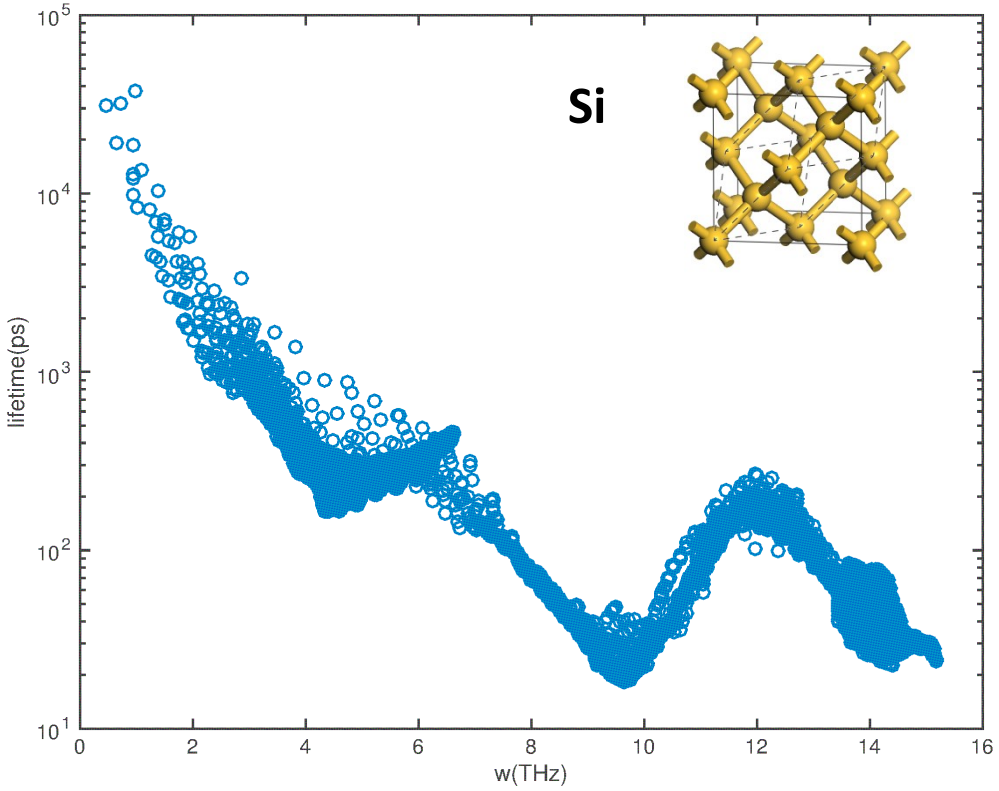
Monte Carlo simulations, ab-initio coupling II





DFT calculations: Dispersion properties & phonon lifetimes

Discretization of the first Brillouin zone on a 31x31x31 K grid

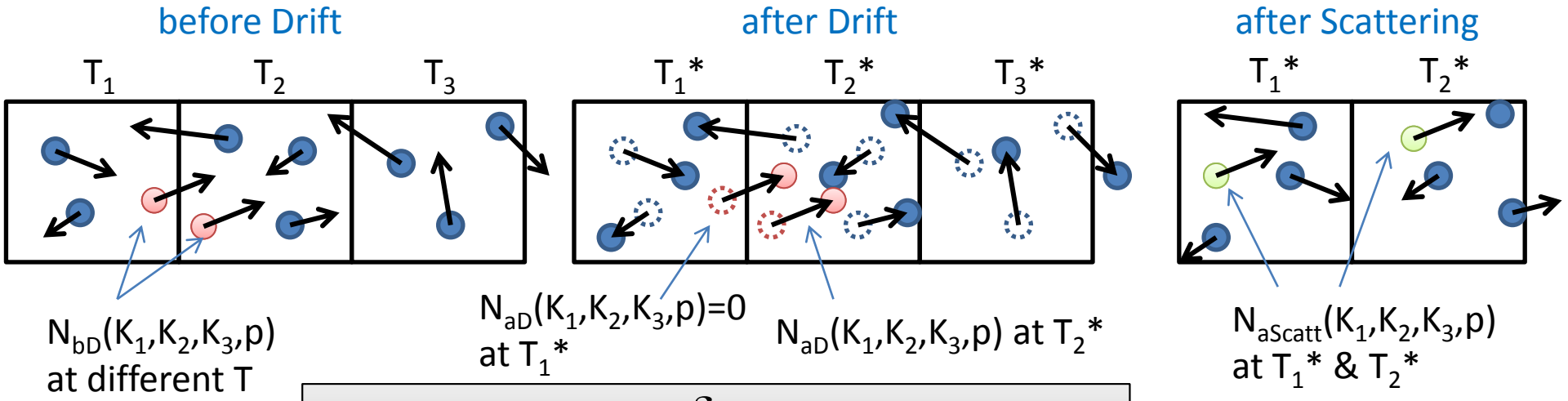


Monte Carlo simulations, ab-initio coupling III

The MC-ab initio solution of the BTE for phonon lies on the same principle as above: discretization, BC, initialization, phonon drift and phonon scattering. Changes are related to:

- Use of a K space discretization instead of frequency one
- Calculation of scattering term of the BTE

For each K point, a phonon is sampled in a cell and caries a given number of modes N.



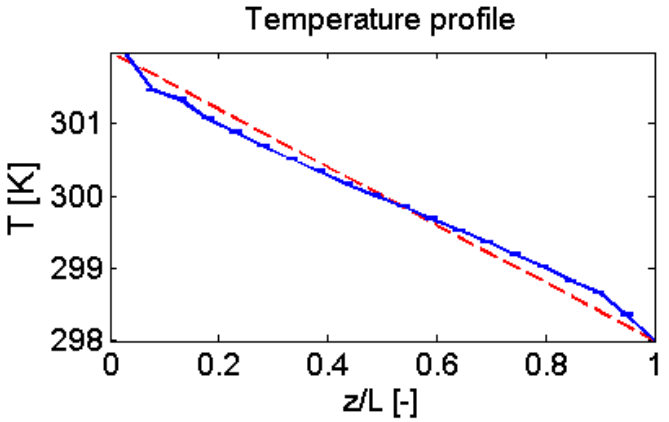
$$N_{aScatt} = N_{aD} + \frac{\delta t}{\tau(K_1,K_2,K_3,p)}\left(N_{BE}(T^*) - N_{aD}\right)$$

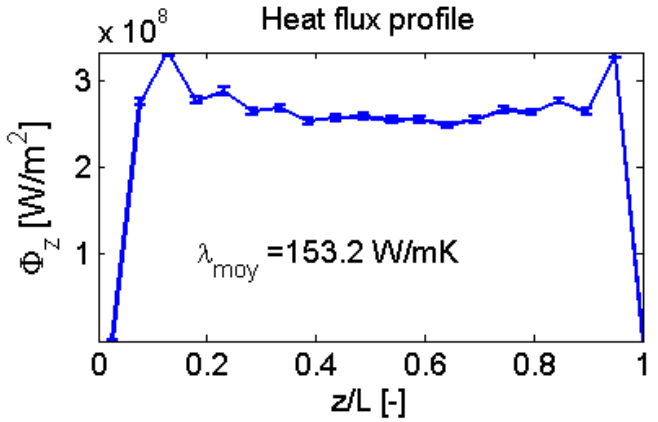
Monte Carlo simulations, ab-initio coupling IV

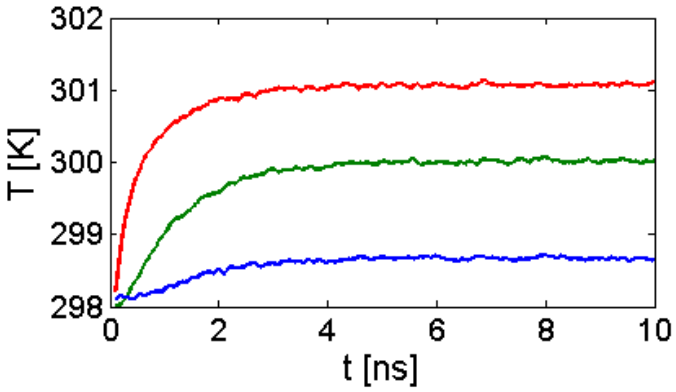
Calculation of T and Φ according to the local phonon distribution in the nanostructure

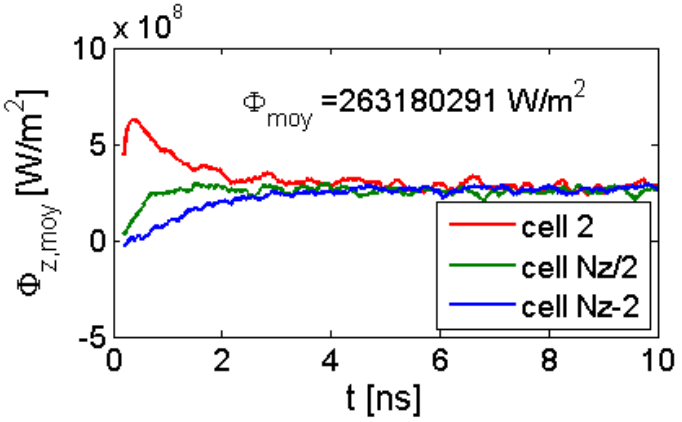
$$\Phi_z = \sum_{i=1}^{N} \frac{\hbar \omega_i V_{gz}}{V}$$

Silicon nanofilm
Lz = 2µm
δt = 1ps
Nz = 20 cells
10000 time steps
1 core / 12 hours
Cross plane TC
k = 153.2 W/m K







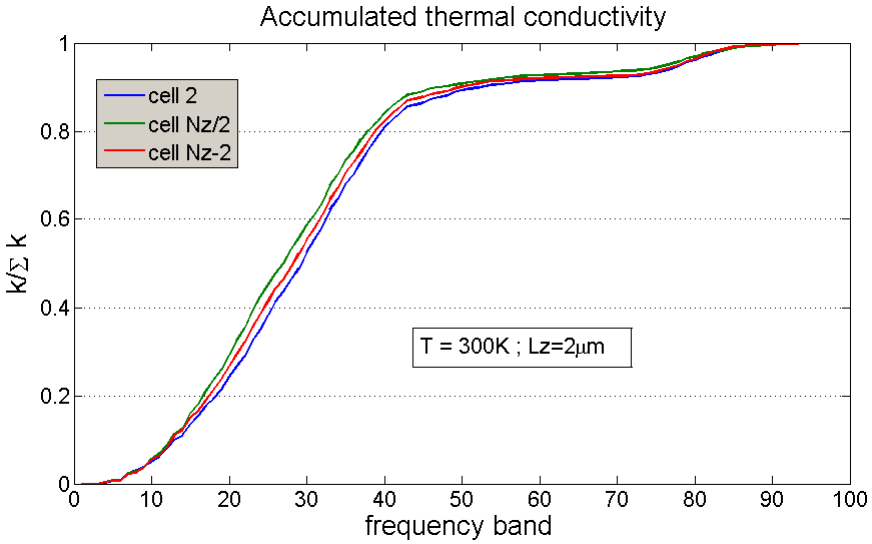


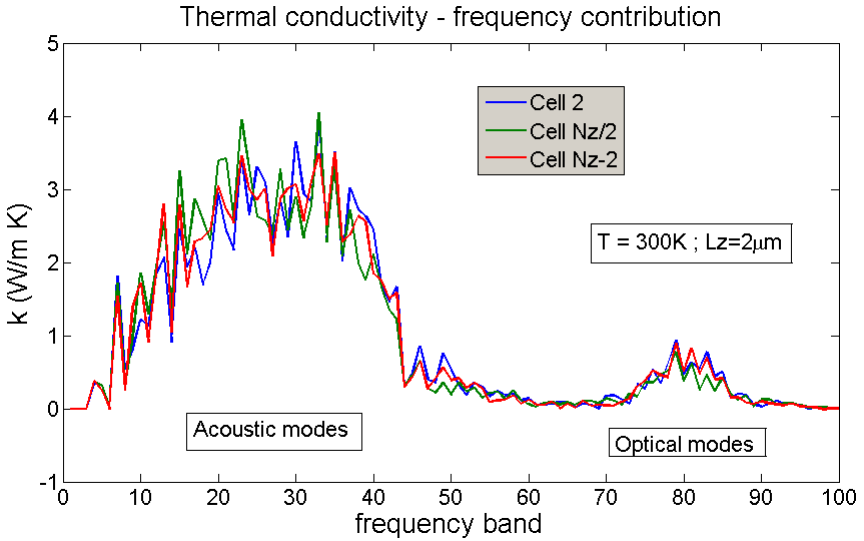
Holland relaxation time : k = 128.7 W/m K !!! Overestimation of the TC with MC-ab-initio !?!

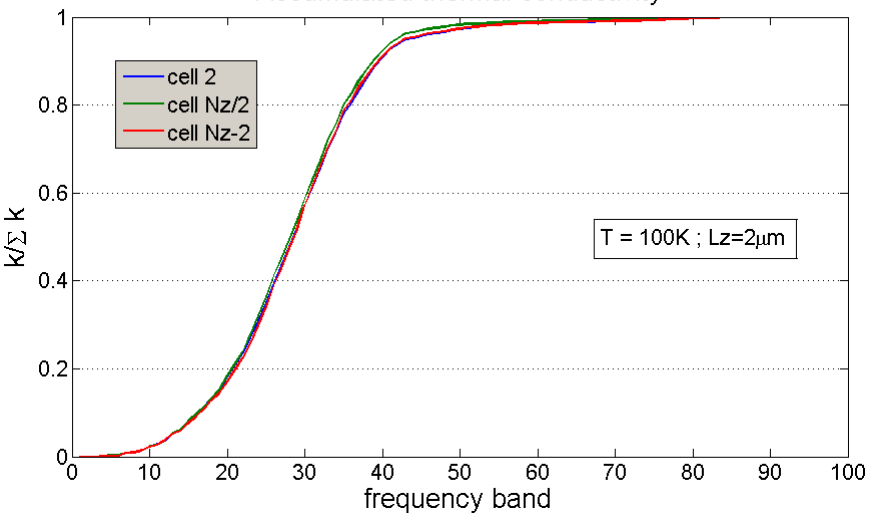
Calculation without $\tau_l(\omega)$: $k_{MC\text{-}NU}$ = 155.2 W/m K

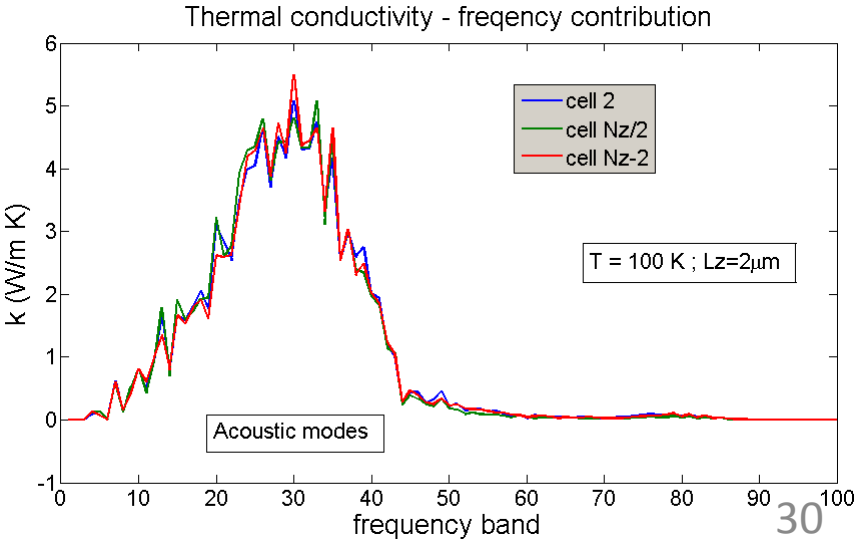
Monte Carlo simulations, ab-initio coupling IV

Monte Carlo post-processing, modes contribution to Thermal Conductivity



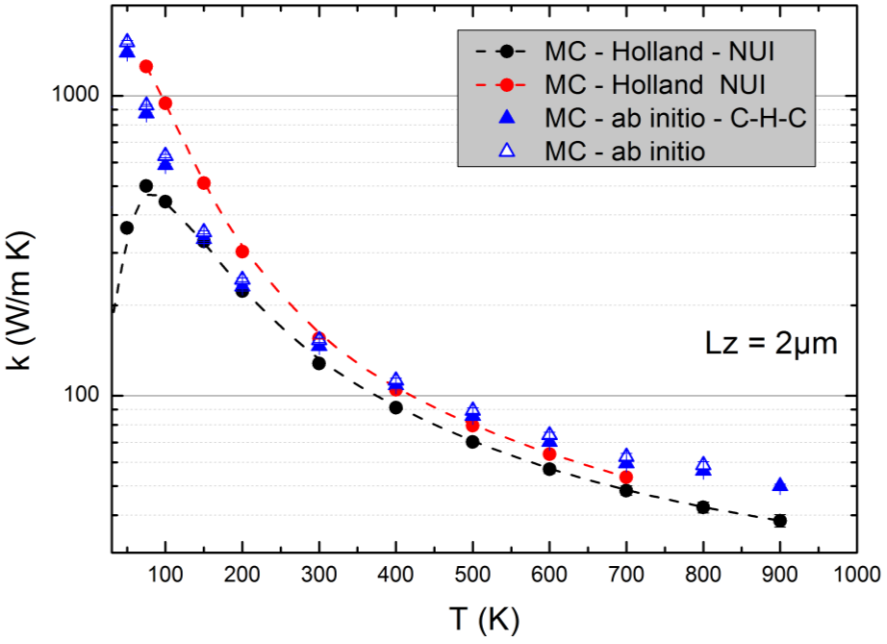




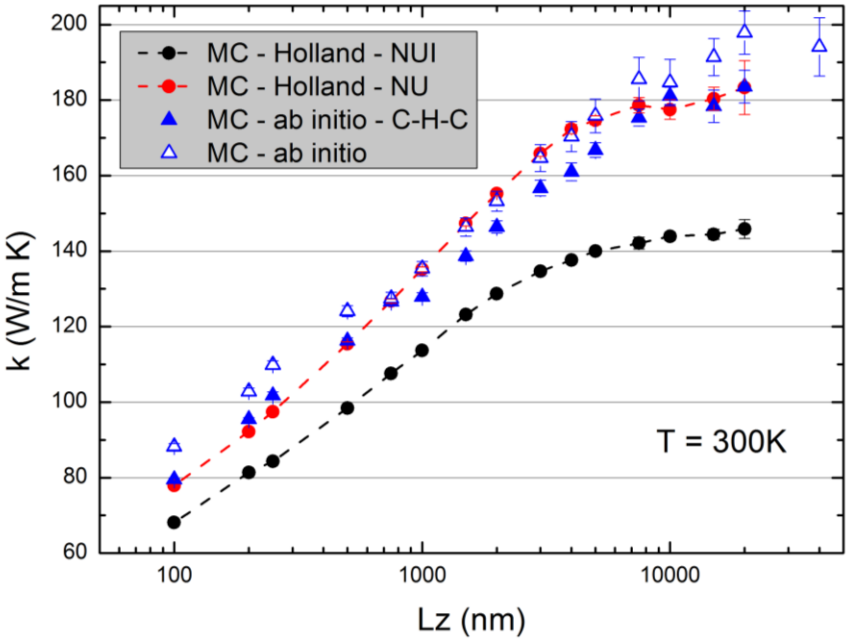


Monte Carlo simulations, ab-initio coupling V

Thermal conductivity in Si vs T; Lz = 2µm



Cross-plane Thermal conductivity in Si film vs Lz



✓ Temperature and thickness well recovered

✓ Impurity scattering explain the light overestimation; need to be implemented in MC-ab-initio

Summary and perspectives

Summary

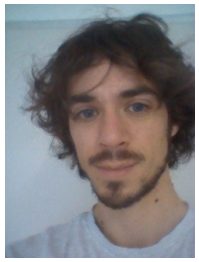
- MC solution of BTE offers a good flexibility to model heat transport in different kind of nanostructures close to the real ones
- Computational cost is reasonable (max 1 week)
- Possibility to include real material dispersion properties
- Gives transient information on heat transport
- Gives spectral information on phonon contribution to thermal properties
- Actually coherent effect are not considered

Colleagues et collaborations

L. Chaput, A. Pr., LEMTA

H. Chaynes, IE, LEMTA

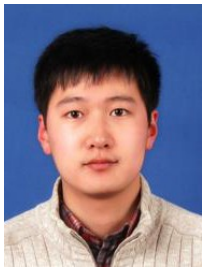
M. Isaev, Post-doc, LEMTA



V. Jean, PhD, LEMTA

Y. Han, Post-doc LEMTA

D. Lacroix, Pr, LEMTA



S. Li, Post-doc, LEMTA

G. Pernot, A. Pr, LEMTA

N. Stein, A. Pr., IJL

K. Termentzidis, CR 1, LEMTA

M. Verdier, PhD, LEMTA

X. Zianni, invited Pr, DEMOKRITOS

Master students!

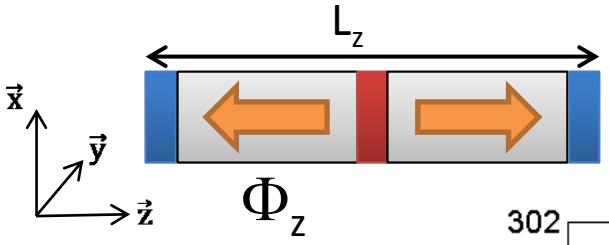
Aristotle University of Thessaloniki

KYOTO UNIVERSITY

DEMOKRITOS
NATIONAL CENTER FOR SCIENTIFIC RESEARCH

Monte Carlo simulations, ab-initio coupling VI

Calculation of T and Φ using Cold-Hot-Cold boundaries



Silicon nanofilm
Lz = 2x2µm
δt = 1ps
Nz = 25 cells
10000 time steps
1 core / 18 hours
Cross plane TC
k = 146.3 W/m K