Molecular Dynamics Study Of The Change Of Silcion Carbide Thermal Conductivity With Irradiation Damage And Grain Size

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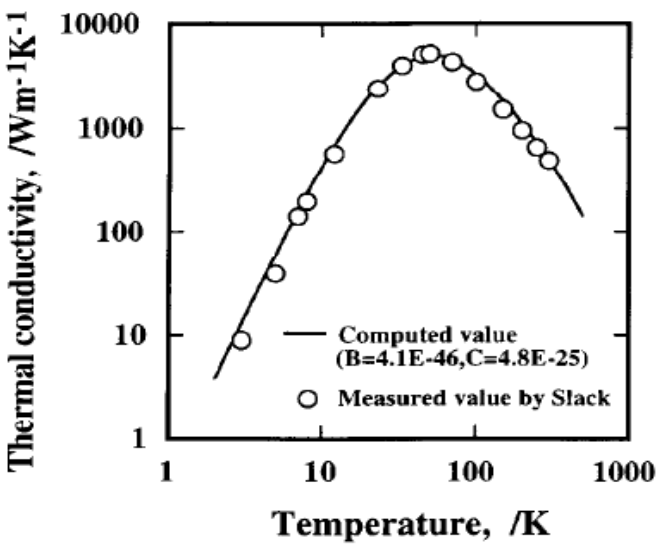
SiC thermal conductivity

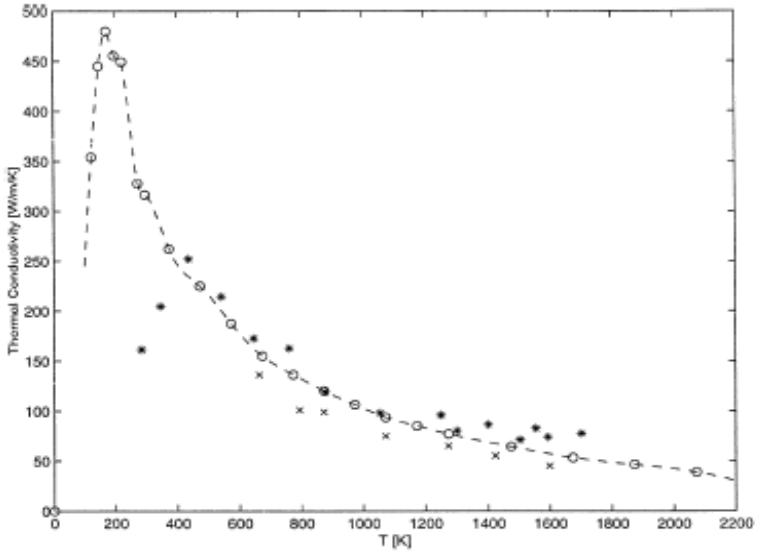
Thermal conductivity = Ratio between heat flux and temperature gradient

$$\vec{J}_Q = -\kappa \vec{\nabla} T$$

Important property for nuclear applications of SiC
(coatings in fusion reactors, matrix in high temperature fission reactors)

Perfect monocrystalline SiC has a very good thermal conductivity





In real materials the conductivity is much lower
achieving high enough values is a strong challenge

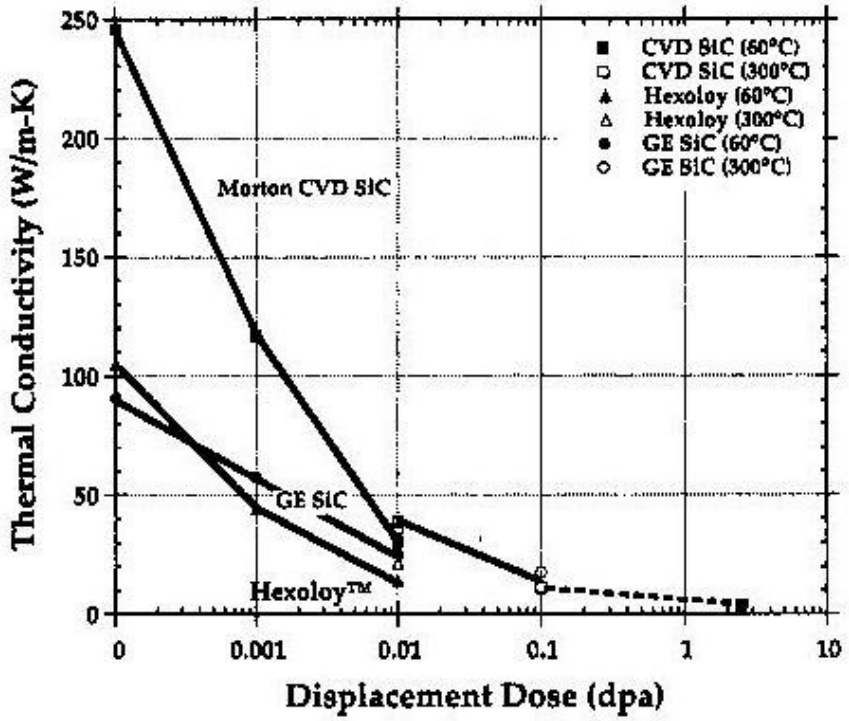
Requirements for fusion reactors
$\kappa \geq 20 W.m^{-1}.K^{-1}$

Sources of degradation of SiC thermal conductivity

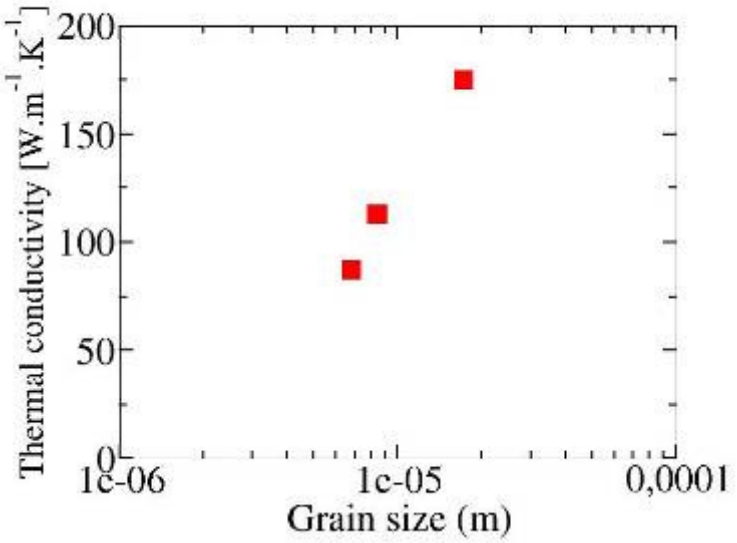
Irradiation damage,
Polycristallinity,
Composite material (fiber, matrix, interphase),
Impurities, dislocations, etc...

Resistivities are additive

$$\frac{1}{\kappa} = \frac{1}{\kappa_{p-p}} + \frac{1}{\kappa_{p-def}} + \frac{1}{\kappa_{p-dis}} + \frac{1}{\kappa_{p-gr}}$$



Exp. by Snead, JNM (2005)



Exp. data from Collins, JAP (1990)

What can we learn from Molecular Dynamics at the atomic scale for pure SiC ?

Molecular Dynamics Simulations at the atomic scale

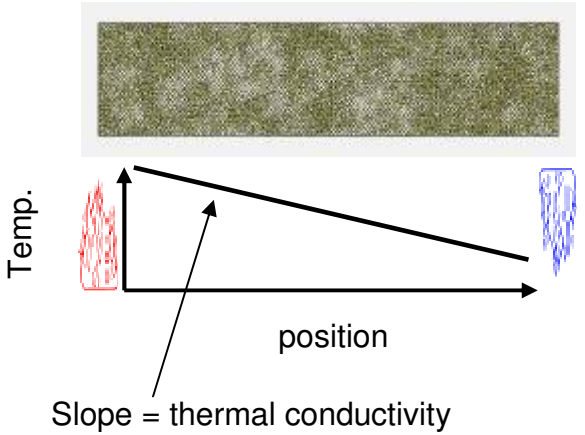
SiC is a semi-conductor ➔ heat conduction through atomic vibrations (phonons)
Can be modeled at by Molecular Dynamics

Simulation box containing atoms with a given arrangement : crystalline, amorphous, defects, grain boundary..
Specific inter-atomic interactions: present work : many-body Tersoff potential, perfect for thermal conductivity
Finite temperature calculations with Newtonian dynamics

Calculation of the thermal conductivity

Direct Method

Heating on one side cooling on the other
Measure of temperature gradient
OK for *inhomogenous* systems



Non equilibrium Molecular Dynamics

Fictitious force ➔ heat current proportional to the thermal cond. without temperature gradient
OK for *homogenous* systems

$$m\vec{a}_i = \vec{F}_i + M^i \vec{F}^{ext.}$$

$$\kappa = \lim_{t\to\infty} \lim_{\vec{F}^{ext}\to 0} \frac{J_q(t)}{TF^{ext}}$$

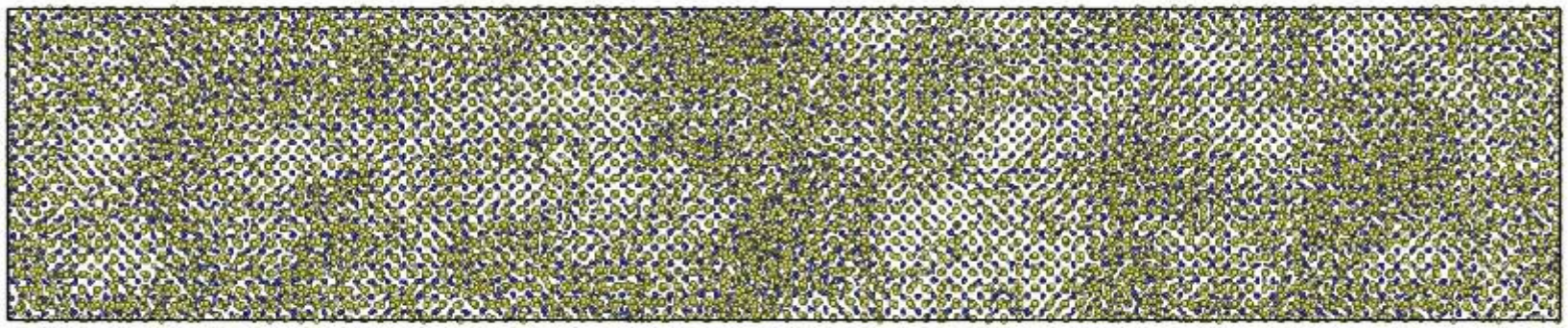
Effect of irradiation damage

Under irradiation (e^-, ions, neutrons) SiC sustain microstructural damage
and even amorphizes (at low temperature T<300 to 500K)

Molecular Dynamics modeling : cascade accumulation

Images after 0, 5, 20, 40, 140 10keV cascades in a 40000 atom box

Gao, PRB (2002)

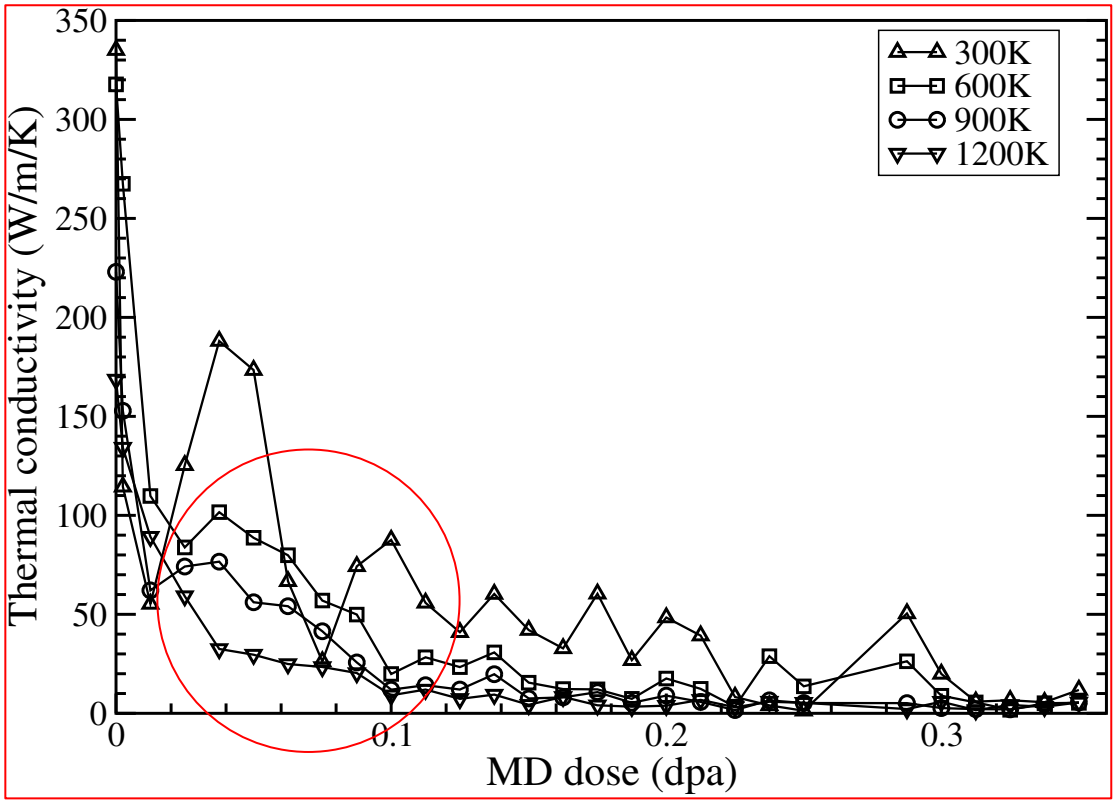


40 casc.
0.1 dpa

Structural evolution from 0 to 0.3 displacement per atom (dpa)
lower doses mainly point defects and clusters ; higher doses: amorphization

Calculation of the thermal conductivity at every stage of accumulation of damage

ıermal conductivity



Lower doses (<0.1dpa):

fast decrease

to values > 10 $Wm^{-1}K^{-1}$

mainly point defects

~high temperature irradiation

exp. 10-50 $Wm^{-1}K^{-1}$

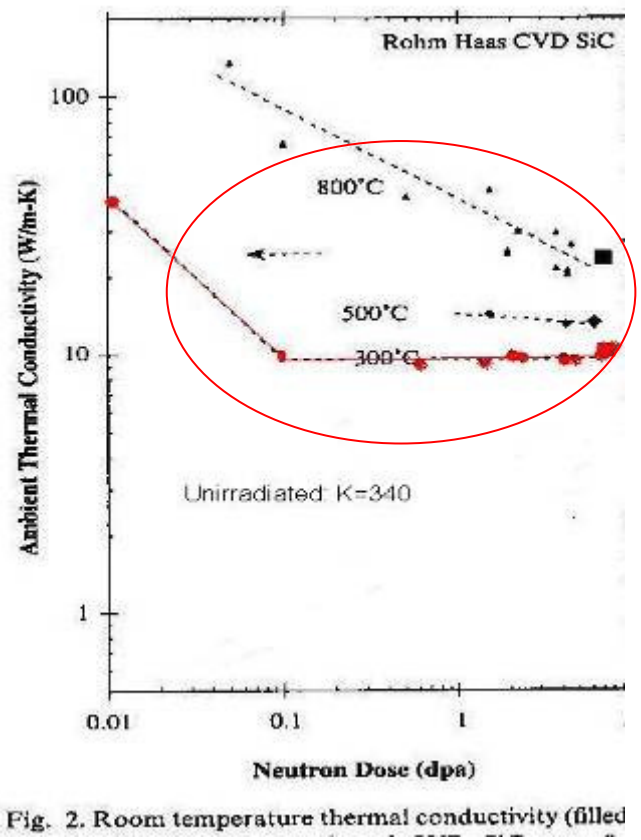
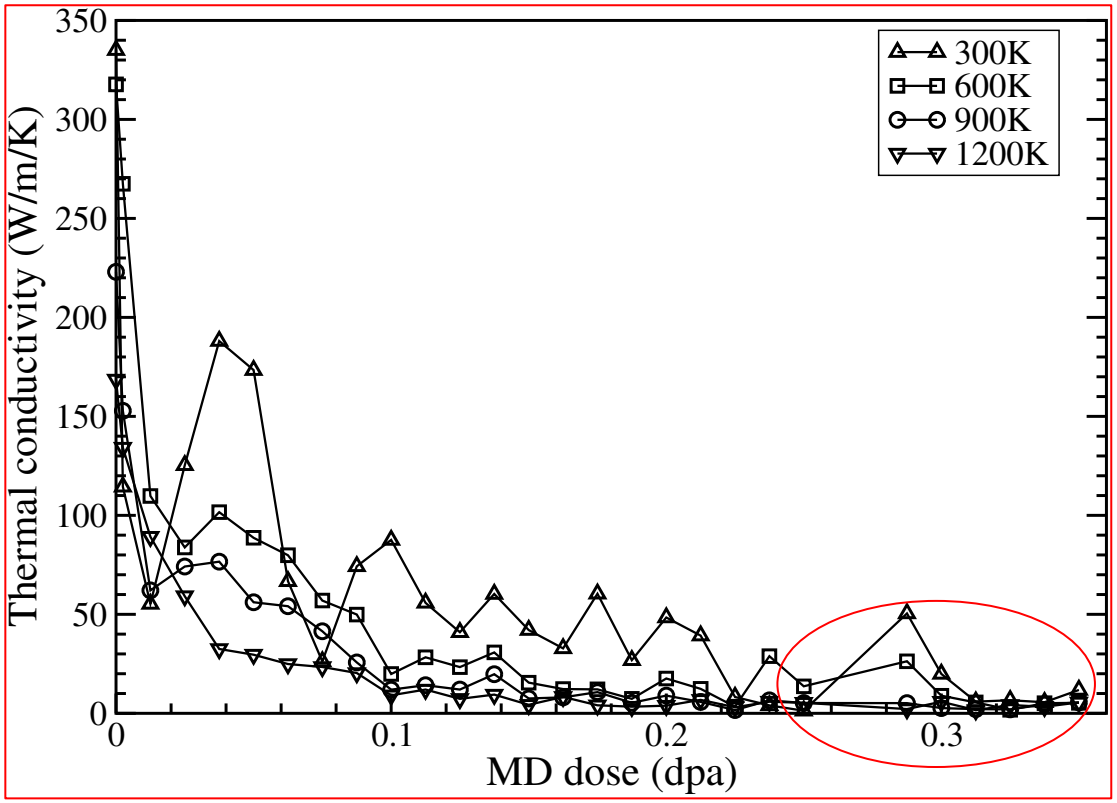
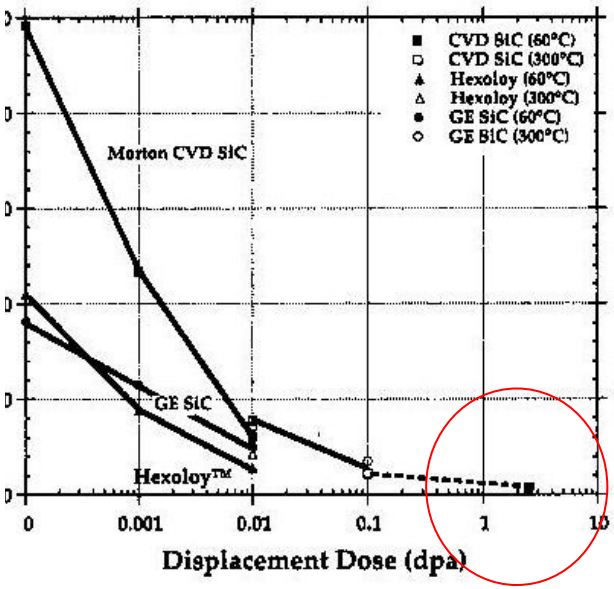


Fig. 2. Room temperature thermal conductivity (filled

Crocombette, JAP 10, 023527 (2007)

ıermal conductivity





Higher doses : further decrease

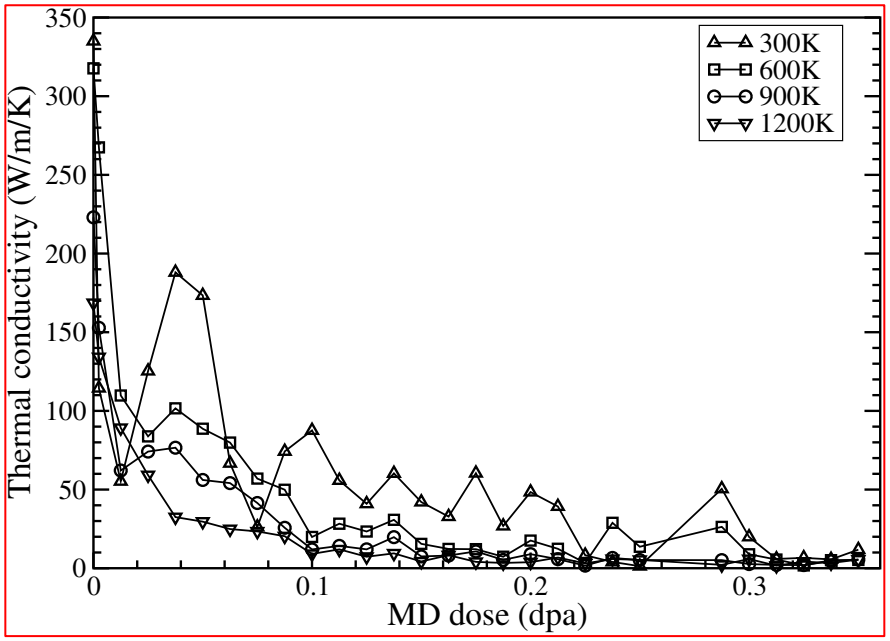
to around 5 $Wm^{-1}K^{-1}$

amorphous phase

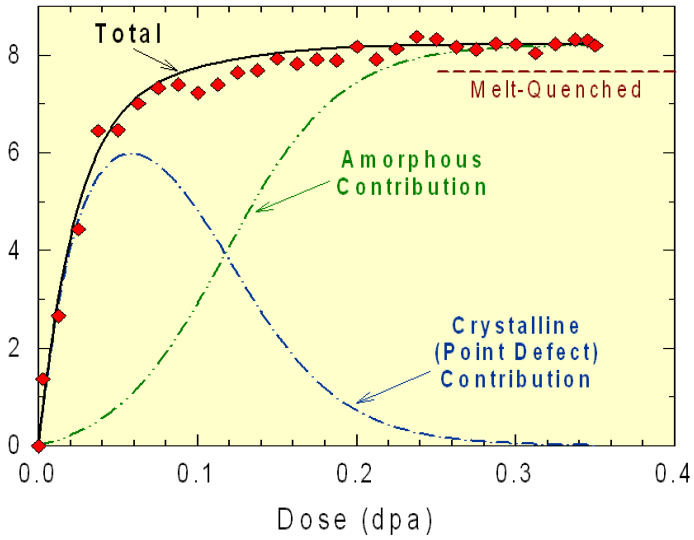
exp (3.8 $Wm^{-1}K^{-1}$)

Crocombette, JAP 10, 023527 (2007)

Thermal conductivity and atomic disorder



swelling



Validation of the structural analysis of the accumulation of damage
Relationship between thermal conductivity decrease and structural disorder/atomic defect state
Thermal conductivity as a scale for atomic defect state

Prospects

Cascade acumulations lacks annealing
High temperature behavior, (irradiation at high temperature + annealing = only point defects
multiscale modeling approach of the post-irradiation kinetics (ab initio, KMC, MD)

Thermal conductivity degradation in polycrystalline SiC

Monocrystals have a higher conductivity than polycrystals

Interfacial Kapitza resistance of the grain boundaries

Temperature drop at the grain boundary, interface effect

$$R_K = \frac{\Delta T}{J}; G_K = \frac{J}{\Delta T}$$

MD calculation of the Kapitza resistances of selected grain boudaries

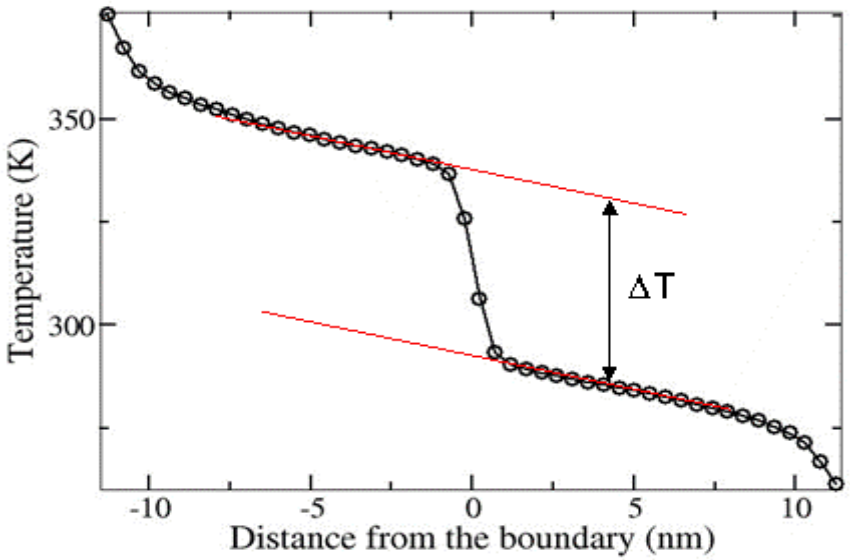
build a bi-crystal, introduce a temperature difference

➔ heat flow, temperature gradient in the two crystals and drop at the interface

The family of grain boundaries : (111) tilt in cubic SiC

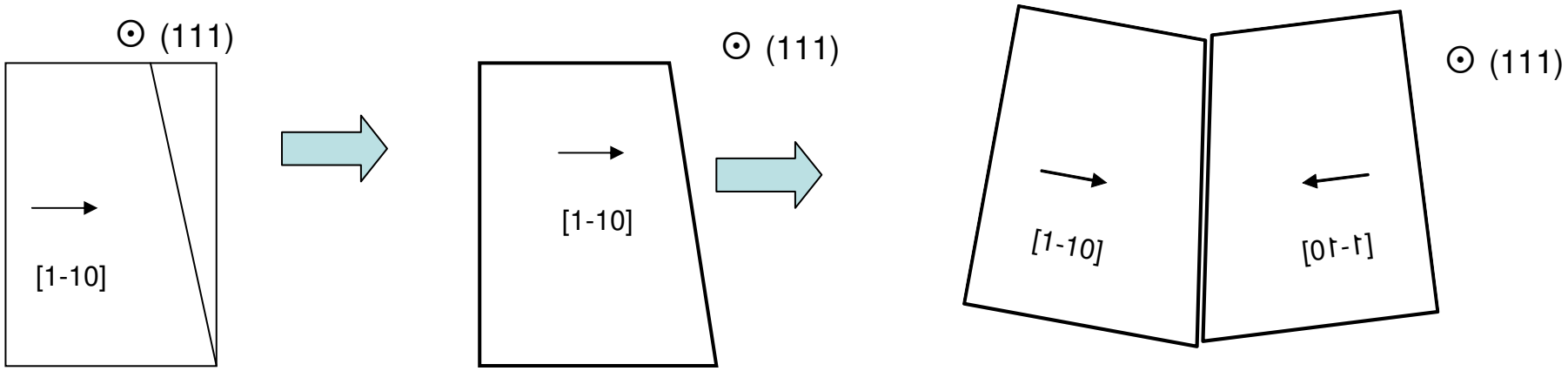
(111) axis : direction of growth for columnar grains in SiC/SiC_f

in the matrix at the interface with the fiber

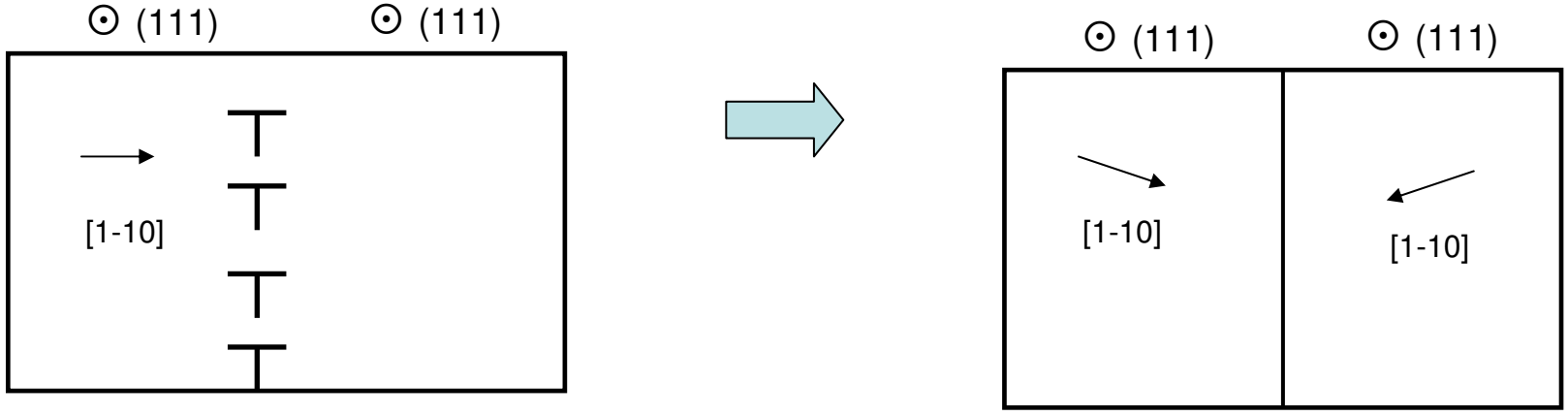


Generation of grain boundaries

Symmetry and shift : cut a grain with a surface including (111), turn one side, flip the other, paste



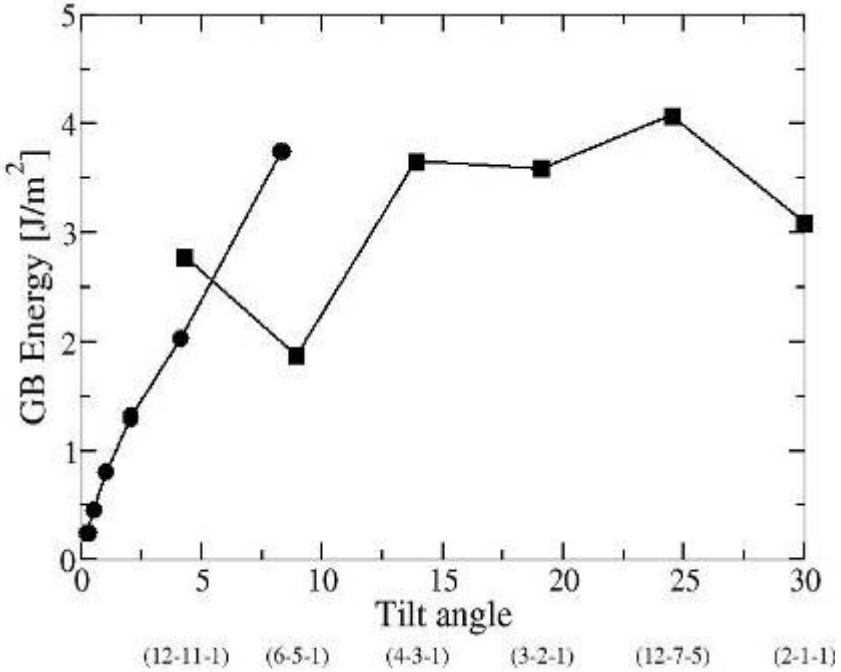
Array of edge dislocations



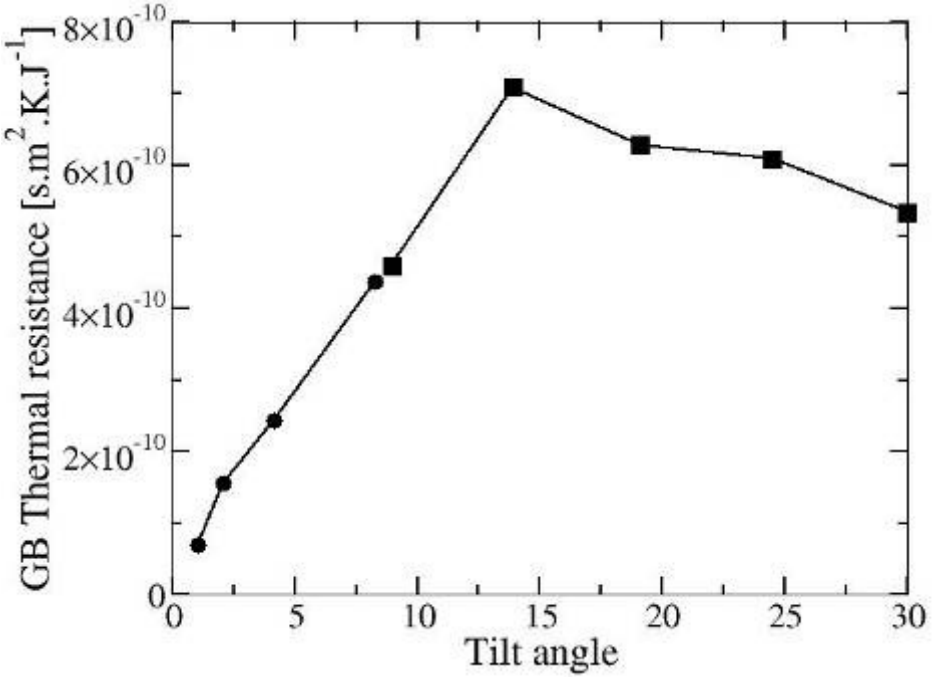
Initial atomic displacements by elasticity theory then relaxation by Molecular Dynamics

GB energies and Interfactial resistances

● dislocations ; ■ symmetry and shift



GB energies



GB interfacial resistances

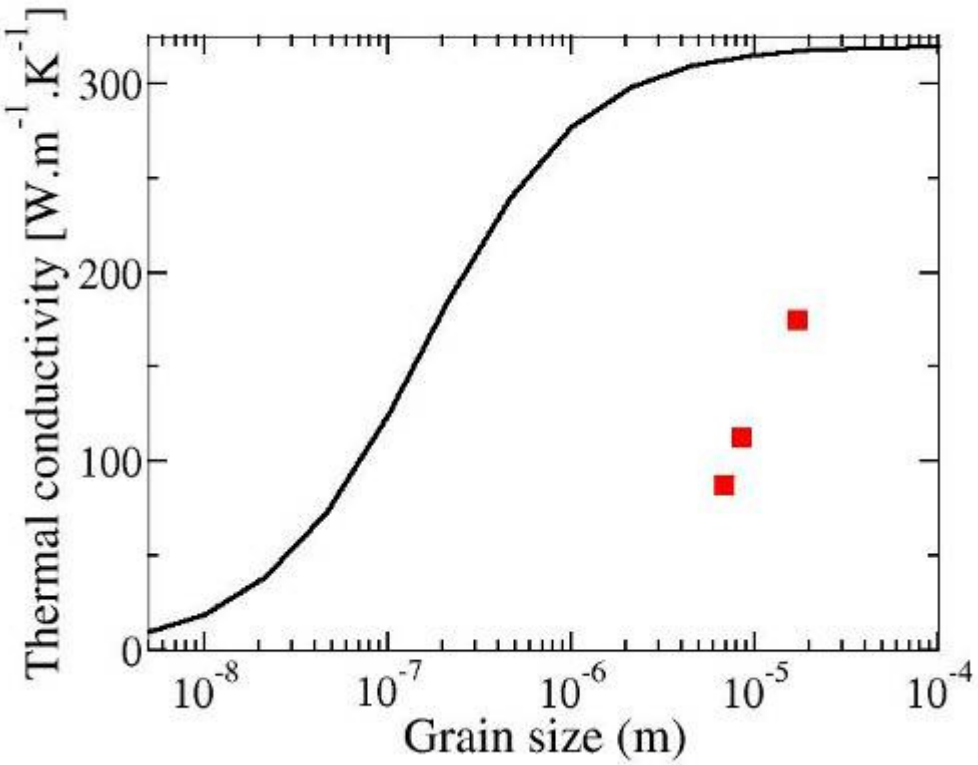
Energy and κ go to zero at low angles then increase to sdaturation

GB from dislocations have a smaller resistance than GB from symmetry-shift

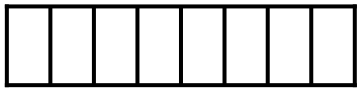
No one to one correspondance between energy and resistance

Quantitative comparison with experiments

Can be fitted with a serial model based on average values of grain sizes and resistances



$$\kappa^S = \kappa_0 / (1 + \kappa_0 \bar{R}_K / \bar{d})$$



Correct qualitative behavior

Wrong quantitatively

■ = Exp. values for SiC

— = calculated values

The decrease of the conductivity kicks in for too small grains

➔ Kapitza resistances are under-estimated by about 50...

Our GBs are too perfect : not enough disorder, no impurities

General conclusions

Molecular Dynamics calculations of the degradation of the thermal conductivity of SiC w.r. perfect crystal

Irradiation damage

- quantitatively correct
- allows a correspondence between atomic defect sate and thermal cond.

Grain boundaries interfacial resistances

- qualitatively correct but vast overestimation
- GB are very far from perfect in real SiC materials