Laboratoire de Mécanique des Contacts et des Structures

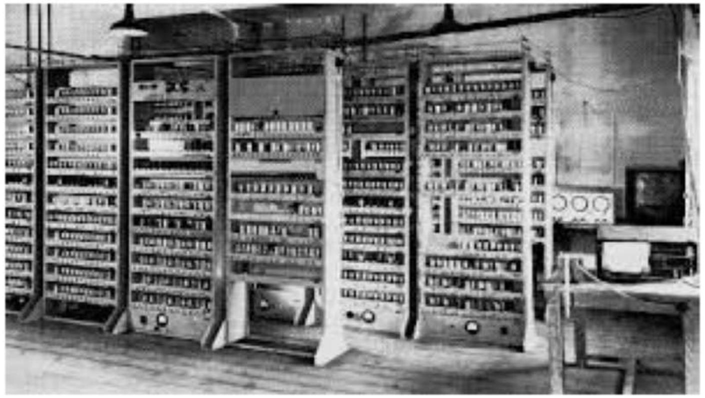
Thermal transfer at the Atomic scale in Amorphous materials and Nanostructured amorphous samples

Anne Tanguy

Tristan Albaret, Yaroslav Beltukov, Tanguy Damart,
Paul Desmarchelier, Valentina Giordano, Anthony Gravouil,
Haoming Luo, Boris Mantisi, Dmitry Parshin, Konstantinos Termentzidis

Institut Lumière Matière, UCBL
&
LaMCoS, UMR CNRS5259 / INSA-LYON
20 Avenue Albert Einstein
69 621 Villeurbanne, France

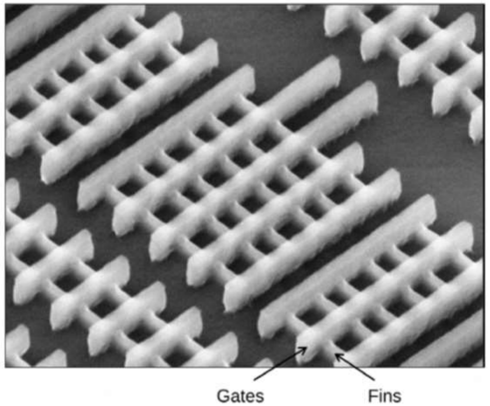
It is important to convert or to evacuate (to manage) the heat at smal scales

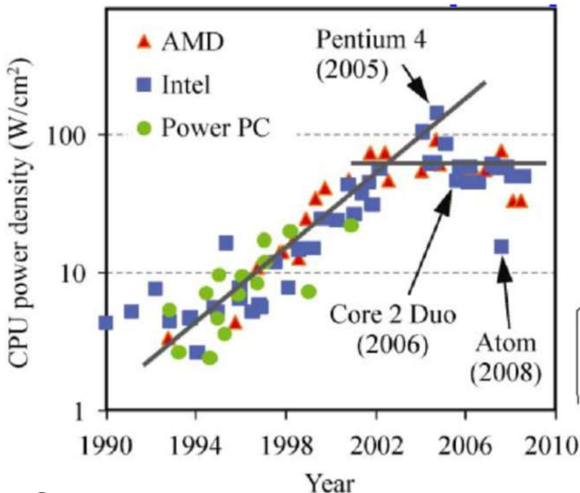


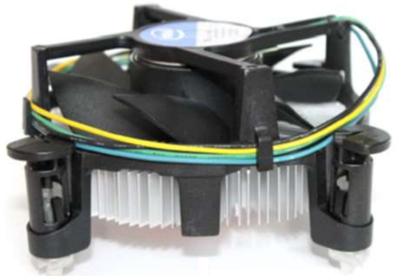
1950's

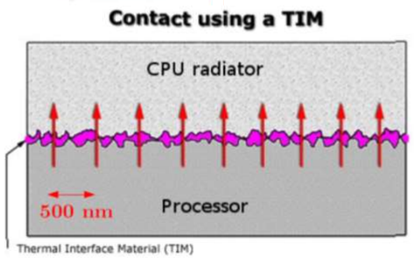
2000's

22 nm Tri-Gate Transistor (Intel, 2011)

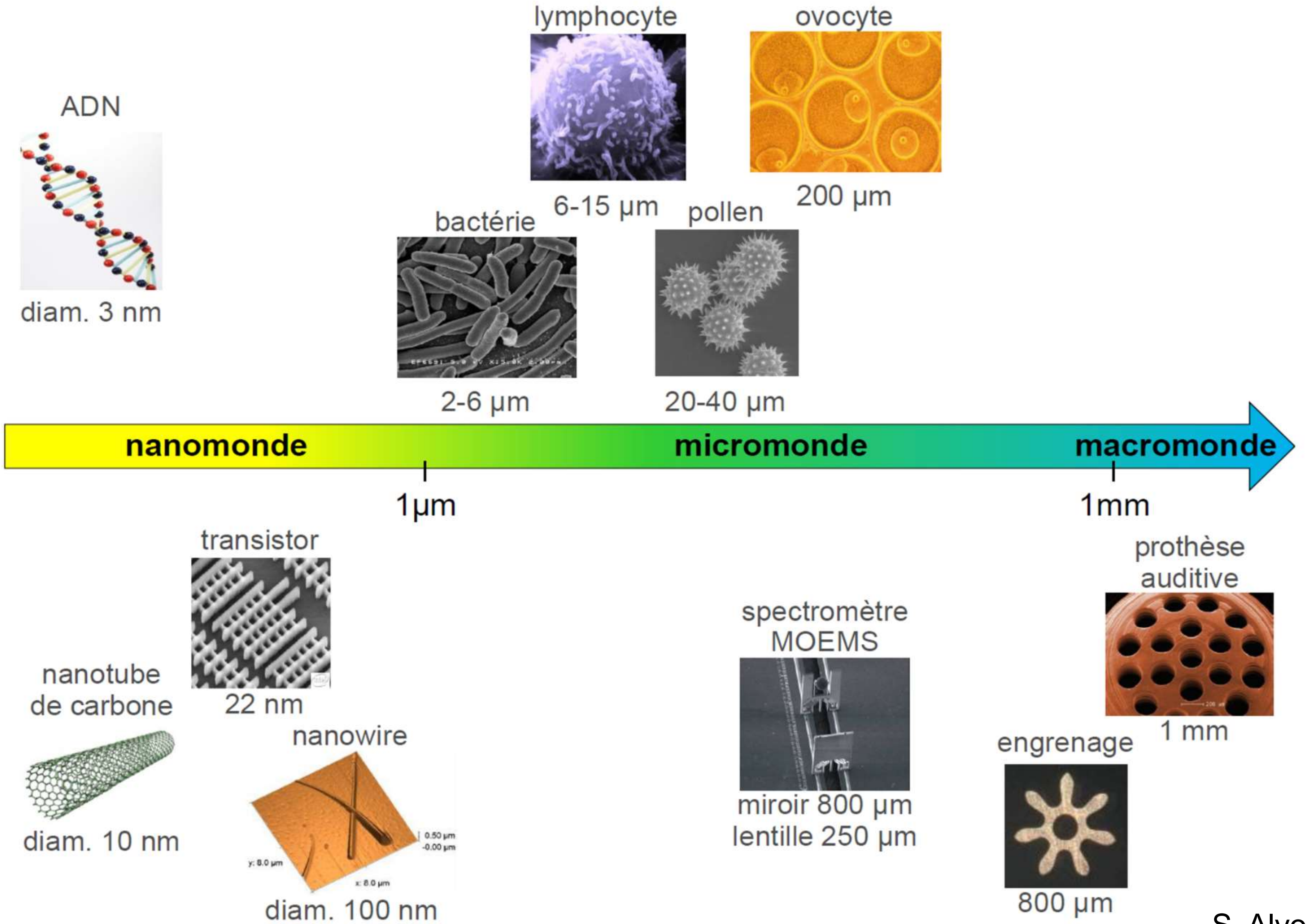








Central processing unit power dissipation


lymphocyte
6-15 µm
ovocyte
200 µm
ADN
diam. 3 nm
bactérie
2-6 µm
pollen
20-40 µm
nanomonde
micromonde
macromonde
1µm
1mm
transistor
22 nm
nanotube
de carbone
diam. 10 nm
nanowire
diam. 100 nm
spectromètre
MOEMS
miroir 800 µm
lentille 250 µm
prothèse
auditive
1 mm
engrenage
800 µm

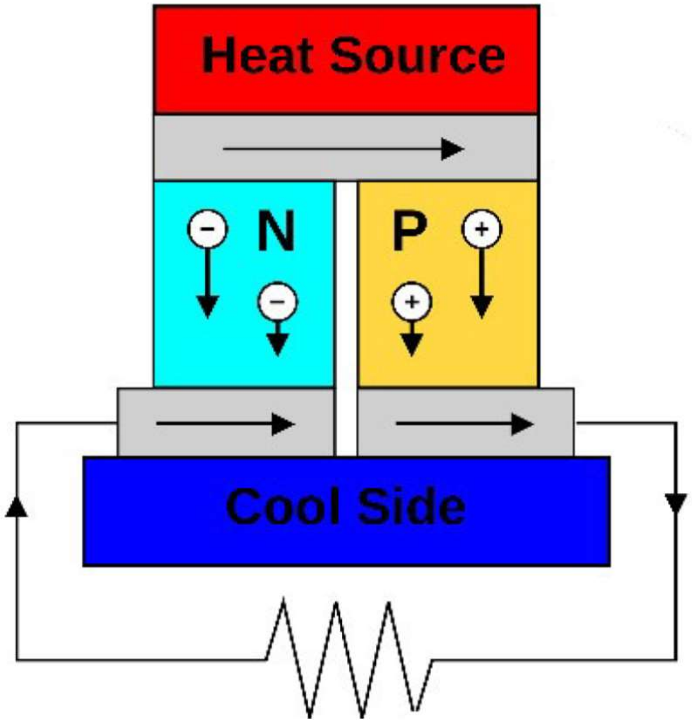
S. Alvo (2012)

Other example: Energy harvesting, Thermoelectric Efficiency: Merit Factor

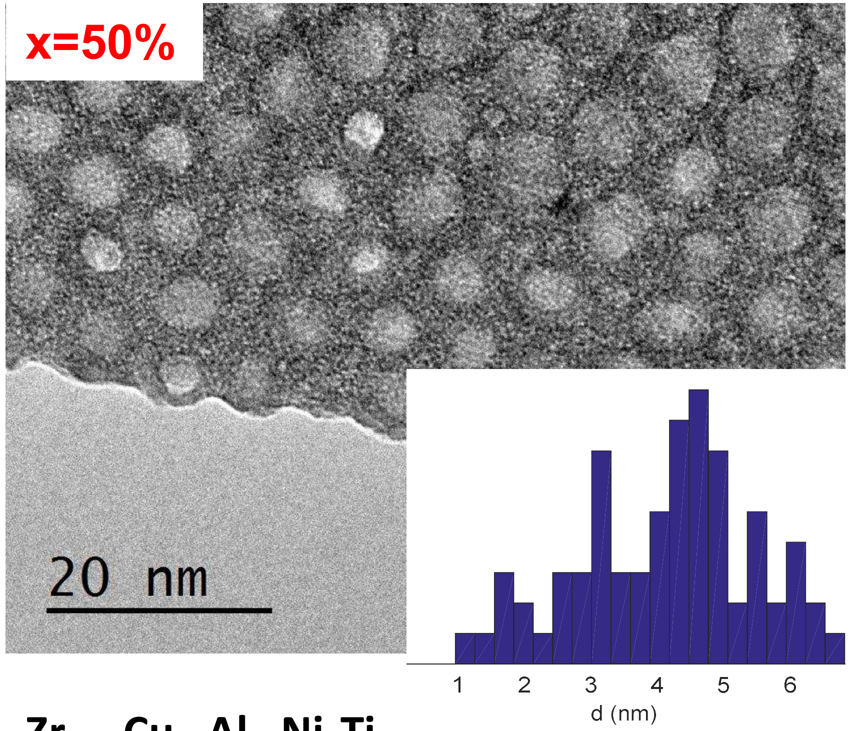
$$Z = \frac{S^2 \sigma}{\lambda_{ph} + \lambda_e}$$

σ — electric conductivity

$\lambda_{ph} + \lambda_e$ — **thermal conductivity**



Property	Metals	Semiconductors	Insulators
S (μVK^{-1})	~ 5	~ 200	~ 1000
σ ($\Omega^{-1}cm^{-1}$)	~ 10^6	~ 10^3	~ 10^{-12}
Z (K^{-1})	~3×10^{-6}	~2×10^{-3}	~5×10^{-17}



V. Giordano et al. (2017)

$Zr_{52.5}Cu_{27}Al_{10}Ni_8Ti_{2.5}$

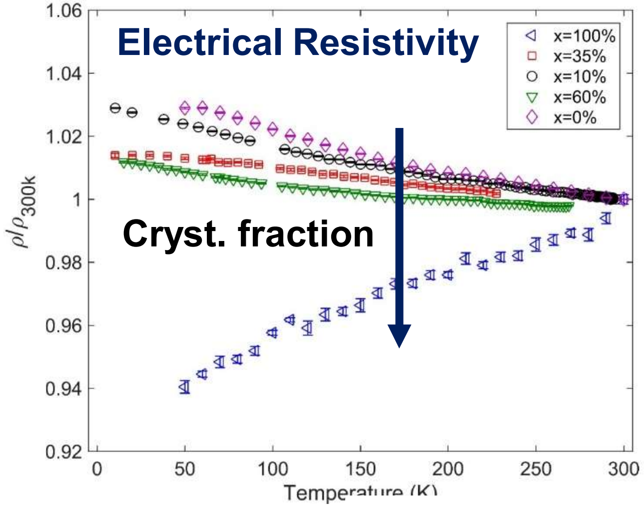
Crystallites mean diameter~4±2 nm

Other example: Energy harvesting, Thermoelectric Efficiency: Merit Factor

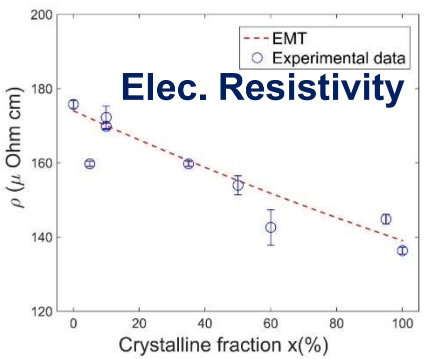
$$Z = \frac{S^2 \sigma}{\lambda_{ph} + \lambda_e}$$

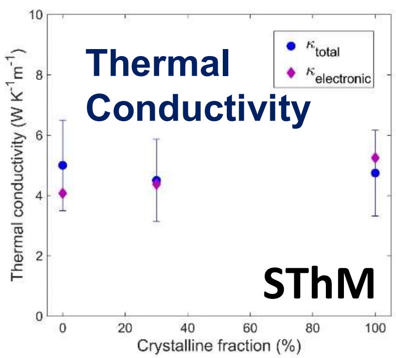
electric conductivity (σ)

thermal conductivity ($\lambda_{ph} + \lambda_e$)

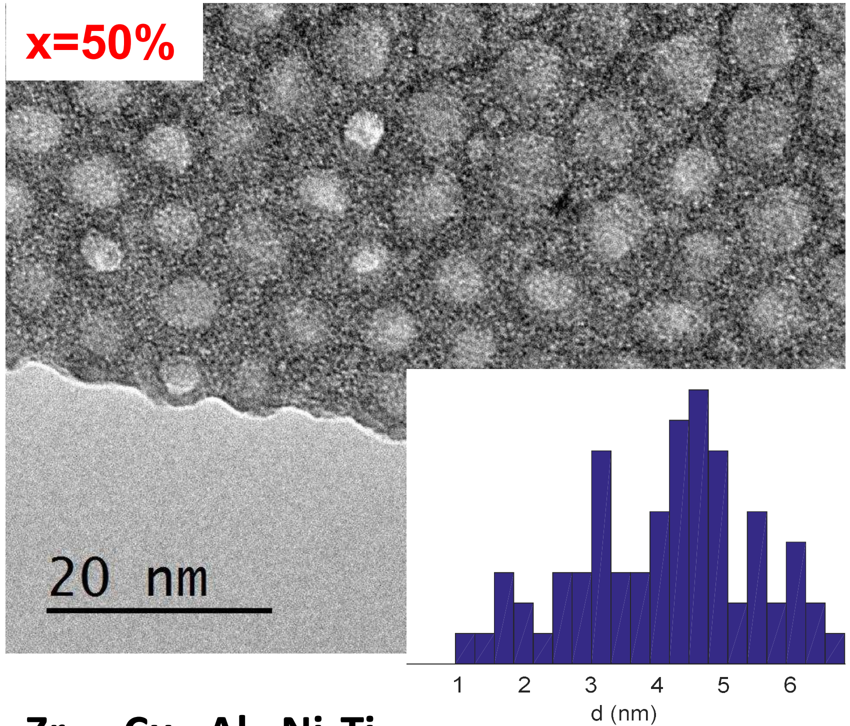


A. Tlili et al. (2017)





Property	Metals	Semiconductors	Insulators
S (μVK^{-1})	~ 5	~ 200	~ 1000
σ ($\Omega^{-1}cm^{-1}$)	~ 10^6	~ 10^3	~ 10^{-12}
Z (K^{-1})	~3×10^{-6}	~2×10^{-3}	~5×10^{-17}



V. Giordano et al. (2017)

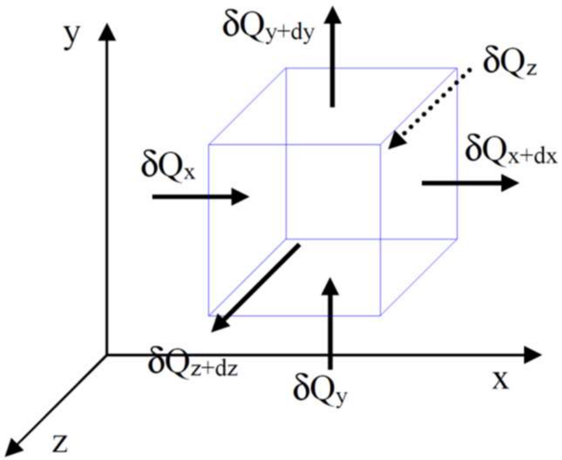
$Zr_{52.5}Cu_{27}Al_{10}Ni_8Ti_{2.5}$

Crystallites mean diameter~4±2 nm

Understanding the Thermal Conductivity at small scales

Out-of-Equilibrium Energy Fluxes:

J.-B. J. Fourier
(1768-1830)

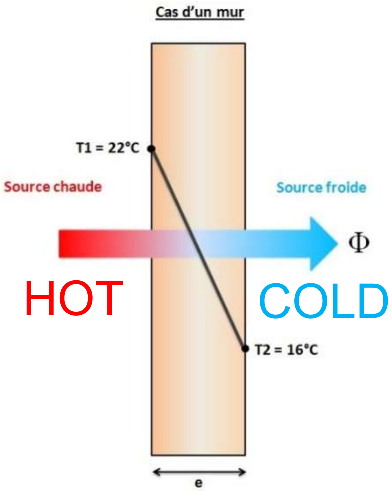


Fourier's Law

$$\delta Q_x = -k_x \frac{\partial T}{\partial x} dydzdt$$

Flux of heat

Thermal Conductivity



Energy Conservation:

$$\delta Q_x + \delta Q_y + \delta Q_z + qdVdt = \delta Q_{x+dx} + \delta Q_{y+dy} + \delta Q_{z+dz} + \rho C.dV.\frac{\partial T}{\partial t}dt$$

$$\frac{\partial}{\partial x}\left(k_x \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(k_y \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(k_z \frac{\partial T}{\partial z}\right) + q = \rho C \frac{\partial T}{\partial t}$$

$$\Delta T + \frac{q}{k} = \frac{1}{D}\frac{\partial T}{\partial t} \qquad D = \frac{k}{\rho C}$$

Diffusivity

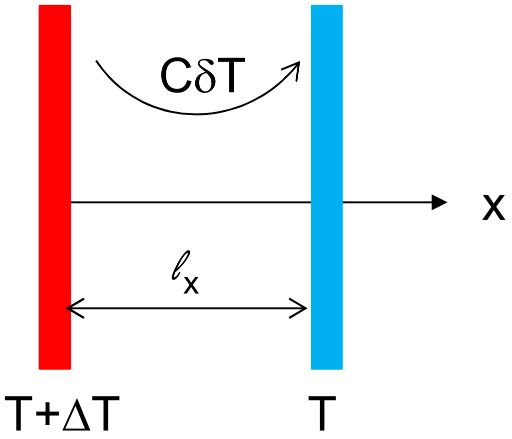
Thermal Conductivity in crystals (Kittel):

Heat carriers = electrons, phonons

In crystals, phonons = quantum of vibrational energy $\left(n+\frac{1}{2}\right)\hbar\omega$

Thermal energy transfer = random diffusive process, « collisions » of heat carriers

Kinetic Theory of Thermal Conductivity:



$$\delta T = \frac{dT}{dx}\ell_X = \frac{dT}{dx}v_x\tau$$

ℓ_X, phonons mean free path

Net flux of Energy: $j_q = -\frac{N}{V}\langle v_x C\delta T\rangle = -\frac{N}{V}\left\langle v_x C\frac{dT}{dx}\ell_X\right\rangle \approx -\frac{1}{3}\frac{N}{V}C\|v\|\ell_X\frac{dT}{dx}$

$$\Rightarrow \boxed{\kappa \approx \frac{1}{3}c\|v\|\ell_X}$$ Heat Conductivity

ℓ_X →

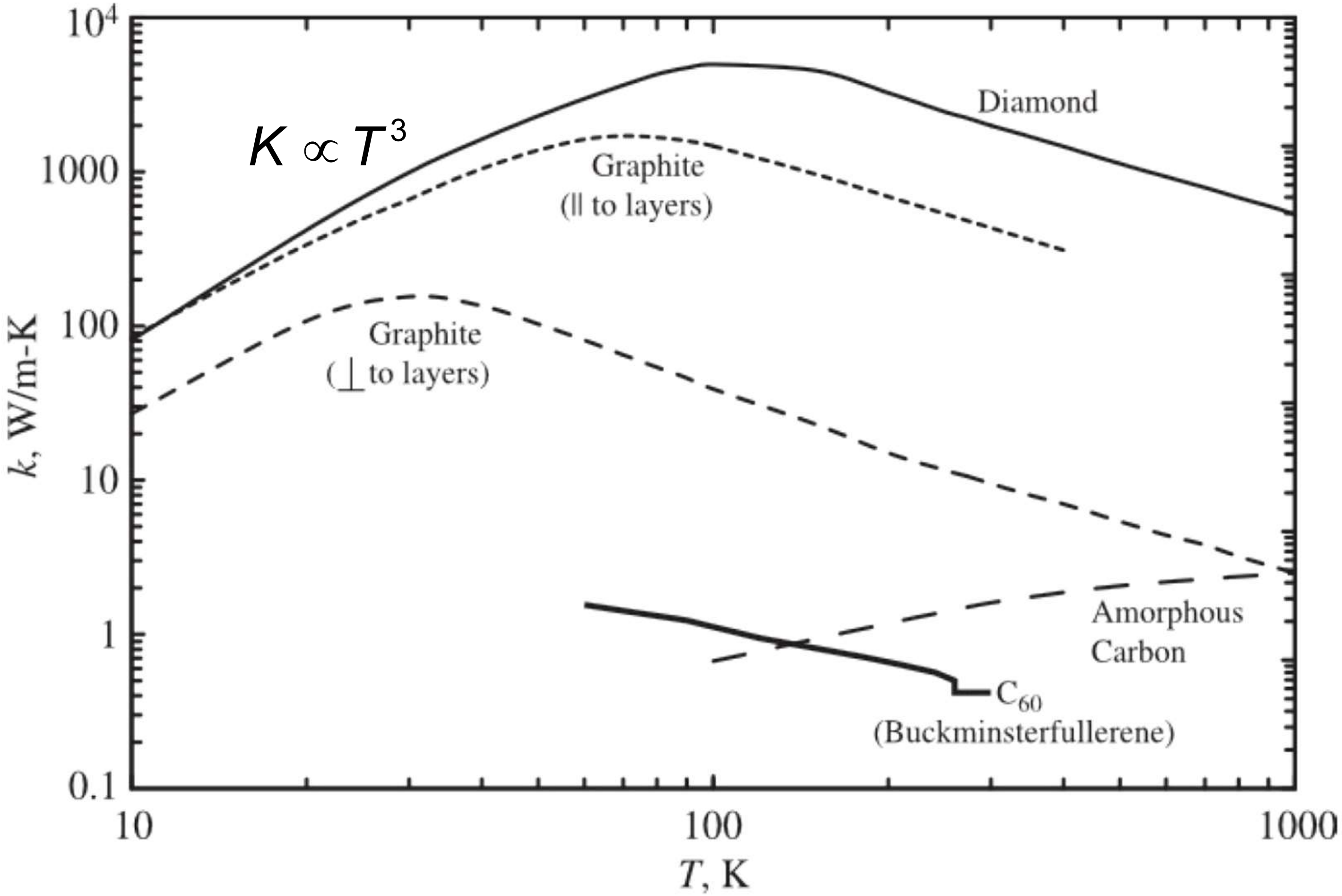
Geometrical scattering: $\ell_X \approx L \quad \kappa \propto c \propto T^3$

crystal boundary, lattice imperfections…

Scattering by other phonons: $\ell_X \propto \frac{1}{n_{phonons}} \propto \frac{1}{T}$

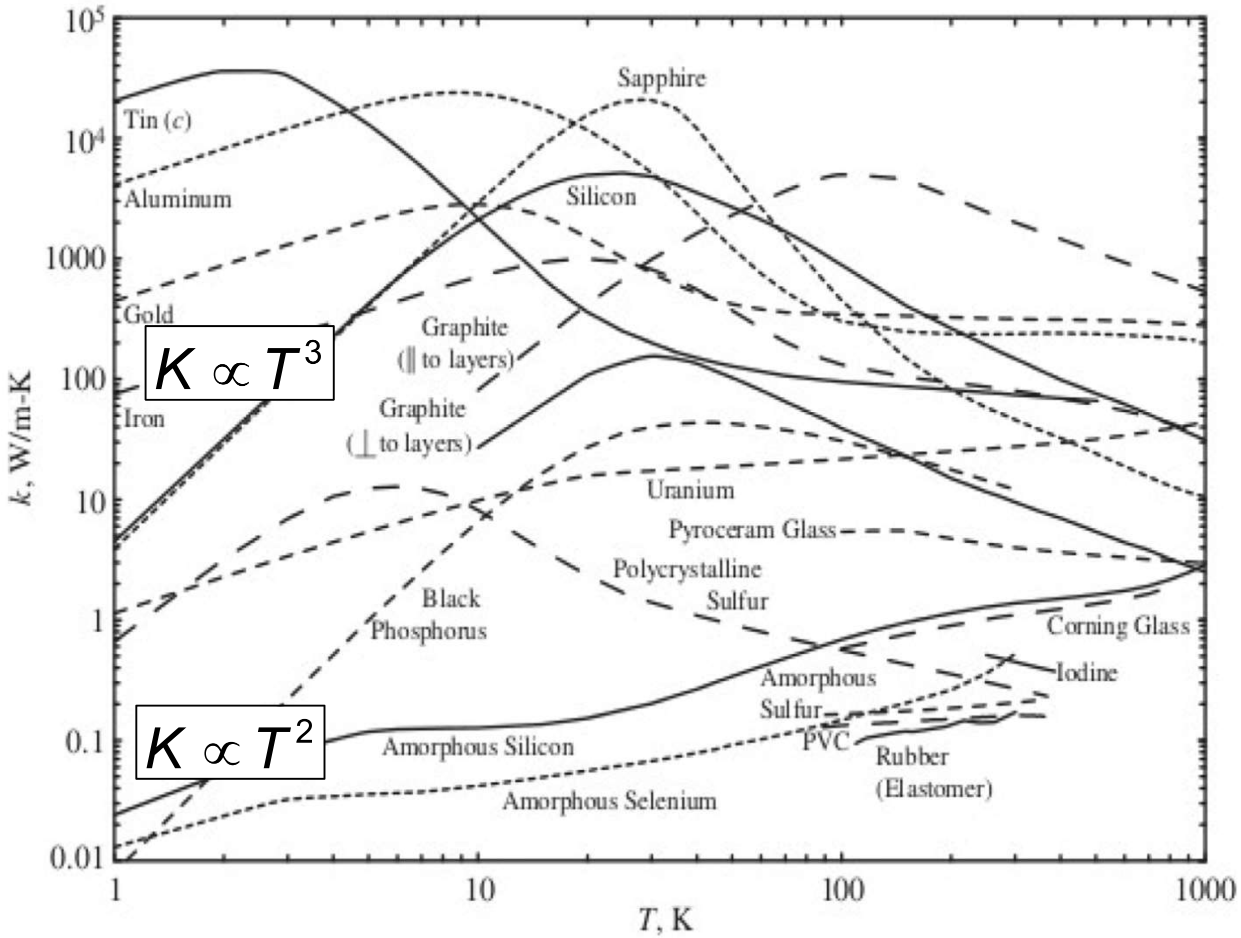
anharmonicity, umklapp processes…

Some Measurements of Thermal Conductivity:



Y. Touloukian, Thermophysical Properties of Matter

Some Measurements of Thermal Conductivity:



Y. Touloukian, Thermophysical Properties of Matter

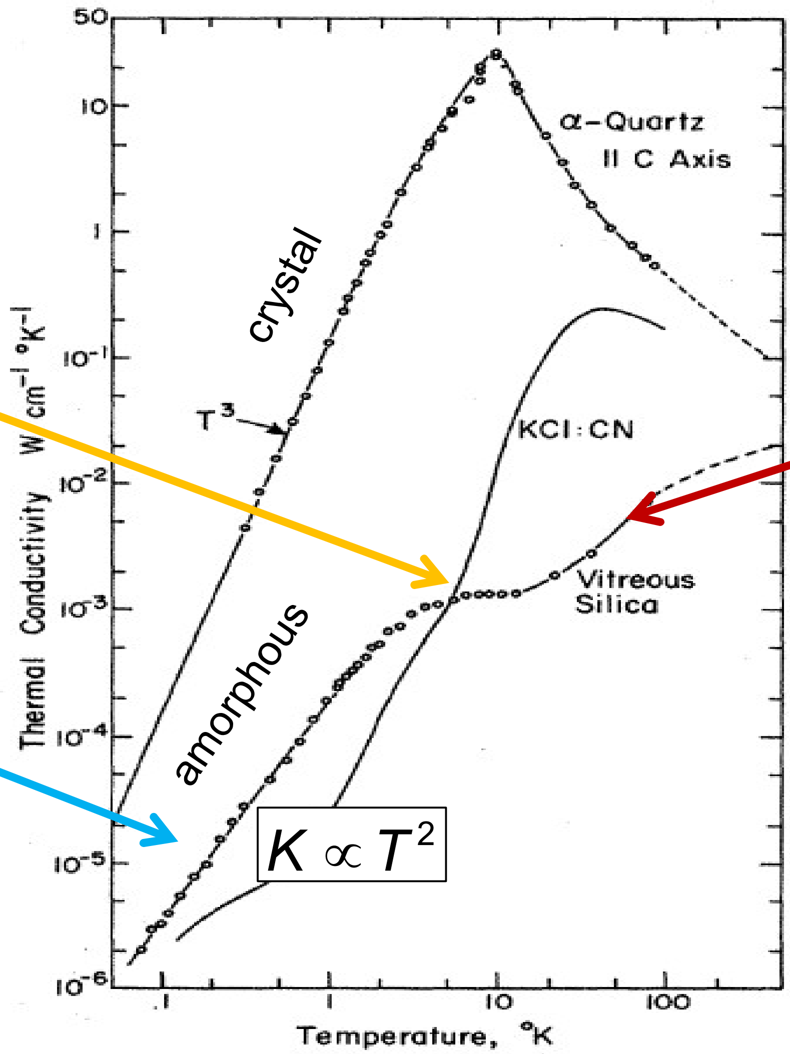
Thermal Conductivity

plateau:

Resonant scattering of phonons by « quasi-local vibrations»?
(Buchenau et al. 1992)

Strongly scattered modes?
(Wyart et al. 2010)

Resonant scattering of phonons on « 2-level systems »?
(Hunklinger et al 1986)



Diffusion mechanism for heat transfer due to clusters vibrating with random phases?
(Cahill et al 1988)

D. Parshin (2013)

What is the role of **disorder** and **interfaces**
in the transportation of energy across the system?

Focus on the **vibrational** energy (phonons)

Interconnection between **Thermal** and **Mechanical** energy?

Introduction

I. Vibrations and Phonons in Amorphous Materials

II. Thermal Conductivity in Nanocomposites

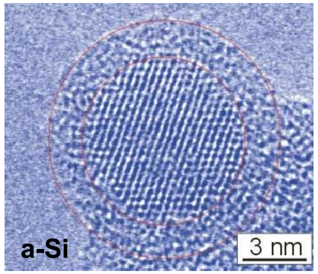
Introduction

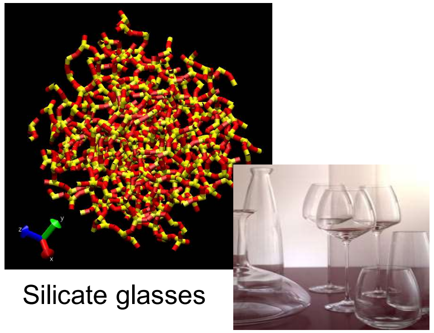
I. Vibrations and Phonons in Amorphous Materials

II. Thermal Conductivity in Nanocomposites

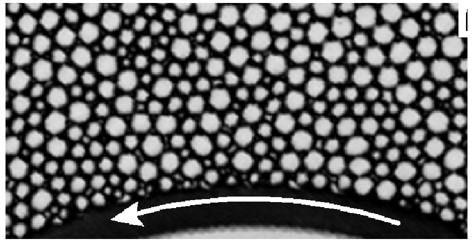
Examples of Amorphous Materials:

Metallic glasses, Vitreloy

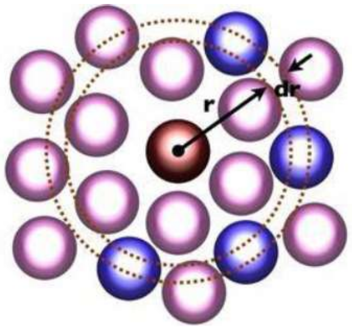


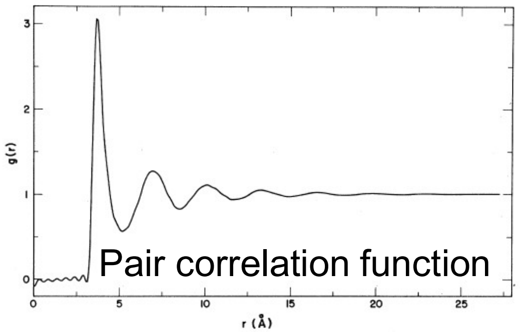


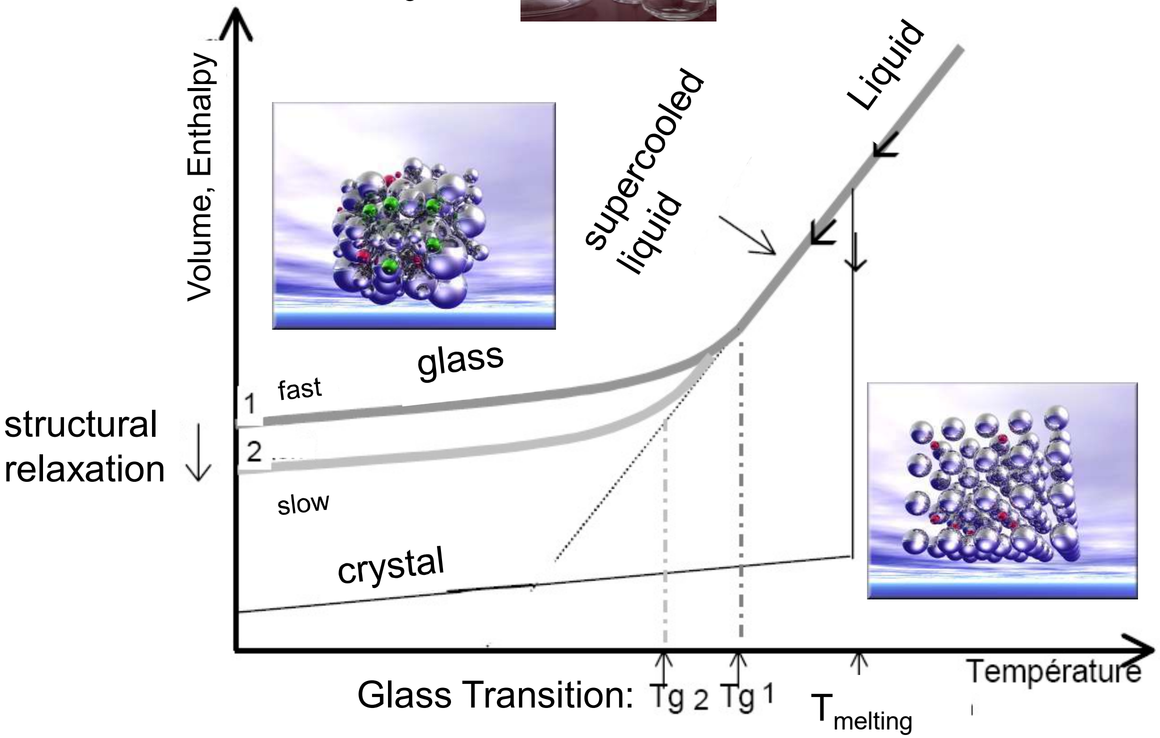
Silicate glasses



Foams







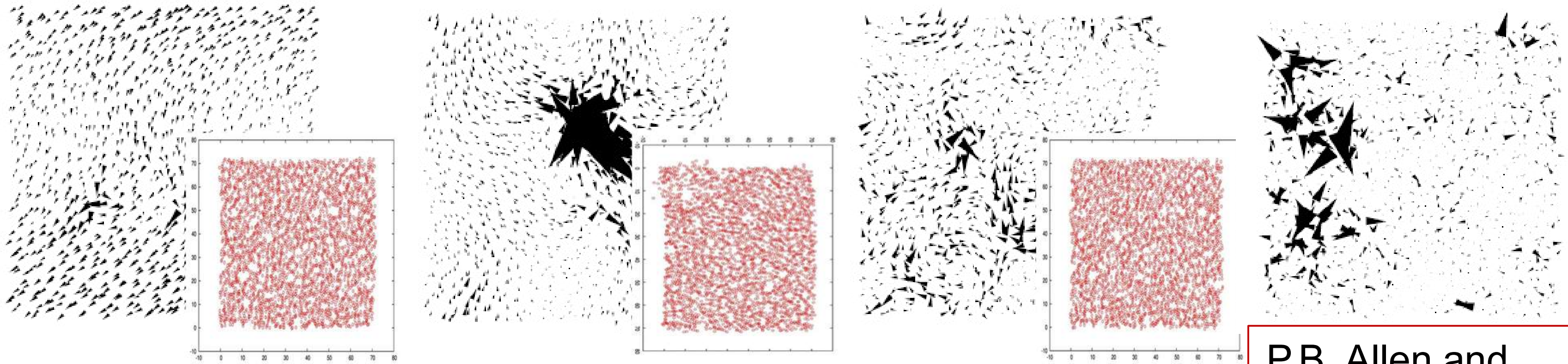
Vibration Modes in Amorphous Materials (spectral analysis)

The example of silica SiO2 glass

$$m_i.\frac{\partial^2 u_\alpha}{\partial t^2}\left(\underline{r_i},t\right) = -\frac{\partial E_{total}}{\partial r_{i\alpha}} \approx -\sum_j \sqrt{m_i m_j} M_{ij}^{\alpha\beta}.u_\beta\left(\underline{r_j},t\right) + f_\alpha\left(\underline{r_i}\right) \text{ with } M_{ij}^{\alpha\beta} \equiv \frac{1}{\sqrt{m_i m_j}}\frac{\partial^2 E_{total}}{\partial r_{i\alpha}\partial r_{j\beta}}$$

$\underline{u}\left(\underline{r_i},t\right) \equiv \underline{r_i}(t) - \underline{r_i}^0 = \underline{U_i}/\sqrt{m_i}.e^{i\omega t}$ Dynamical Matrix: **Eigenvectors** $\underline{\text{U}}$, **eigenvalues** ω^2

Plane Waves **Soft Modes** **Diffusons** **Locons**



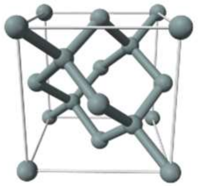
B. Mantisi et al. (2012), N. Shcheblanov et al. (2015)

P.B. Allen and J.L. Feldman (1999)

Vibration Modes in Amorphous Materials (spectral analysis)

The example of amorphous silicon

Y. Beltukov et al. (2016)



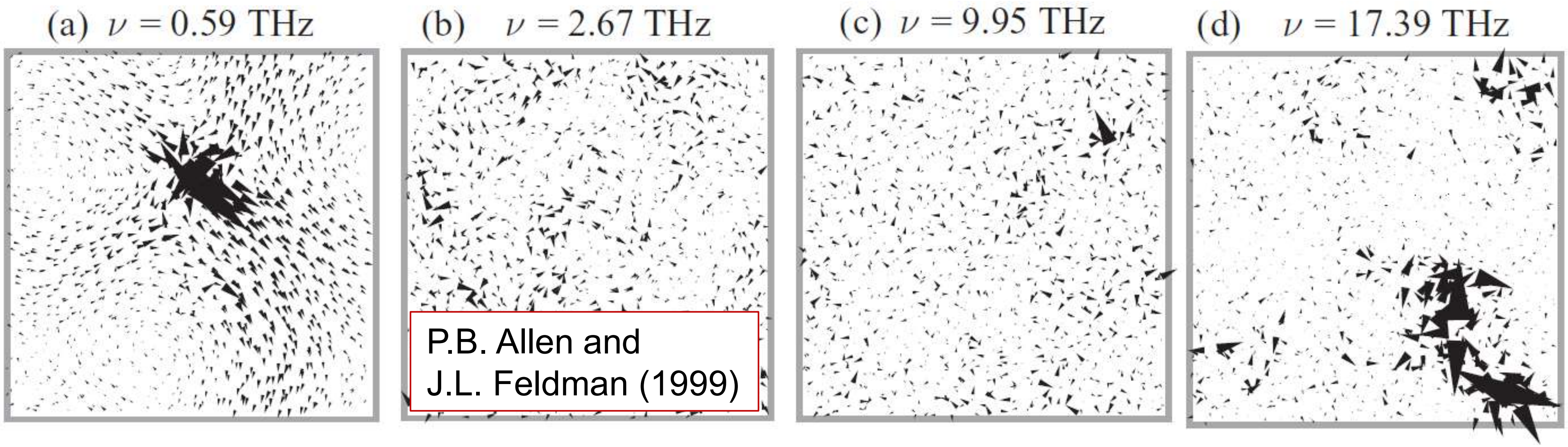
Interatomic interaction energy:

$$E_{total} = \Sigma_{(i,j)} f(r_{ij}) + \Sigma_{i,j,k} \Lambda \left(\cos\theta_{jik} + \frac{1}{3} \right)^2 . e^{\gamma.(r_{ij}-a)^{-1} + \gamma.(r_{ik}-a)^{-1}}$$

Bending energy

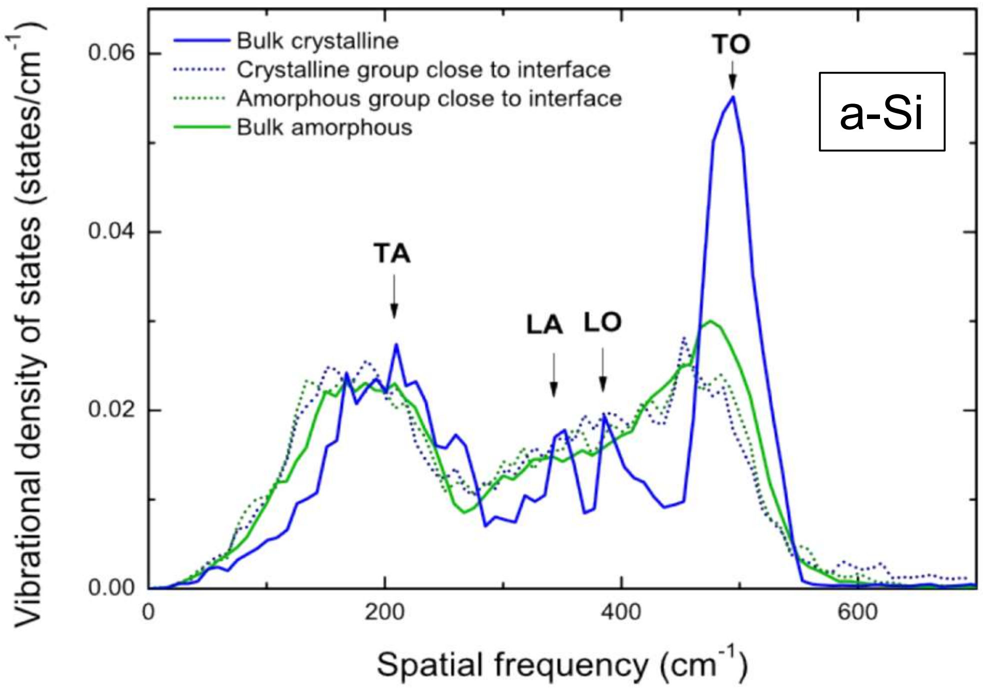
Stillinger-Weber

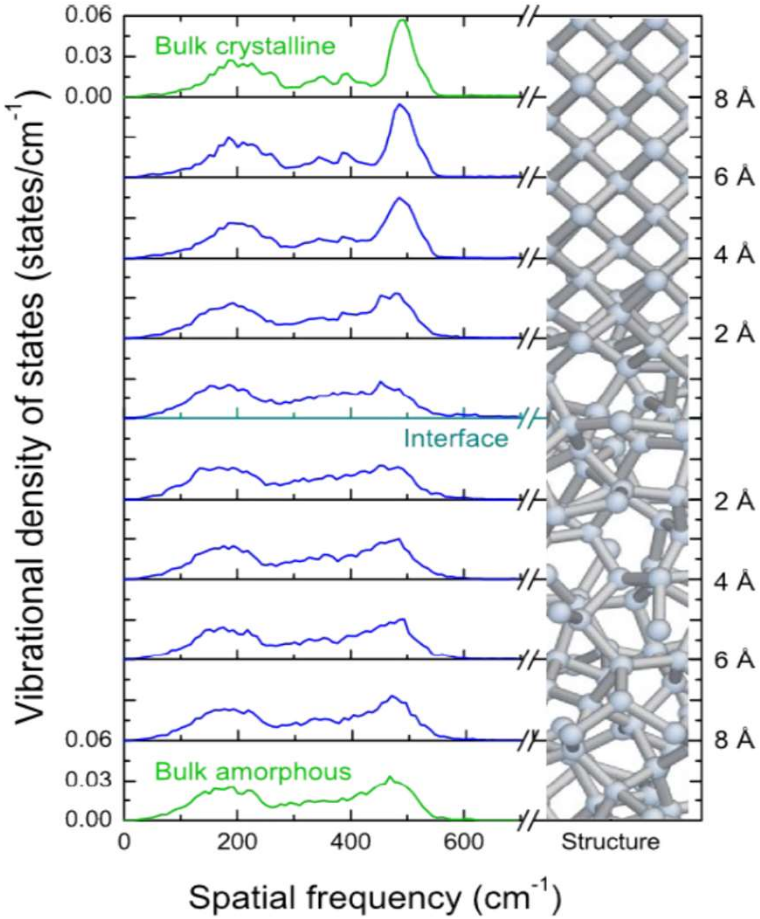
soft modes **diffusons** **locons**



Vibrational Density of States

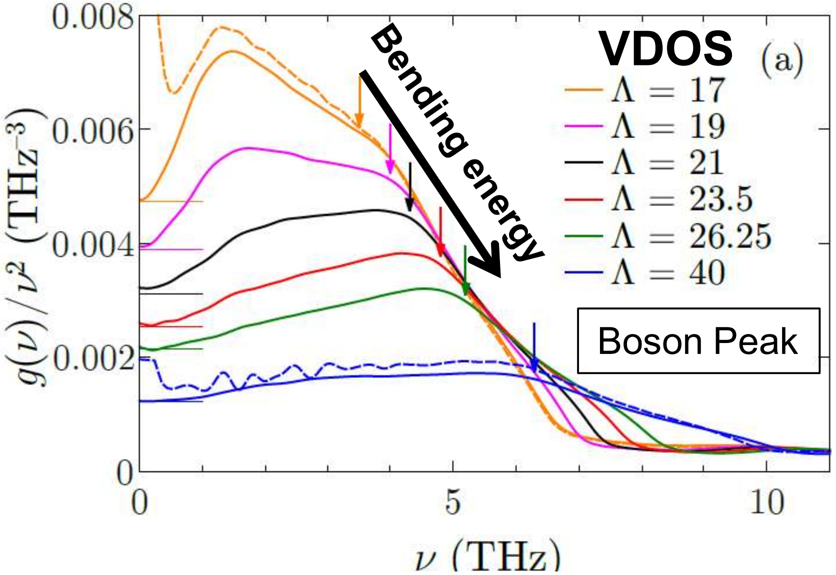
Structural sensitivity, from crystal to amorphous solid





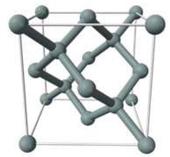
A. France-Lanord et al. (2014)

Normalized Vibrational Density of States in a-Si

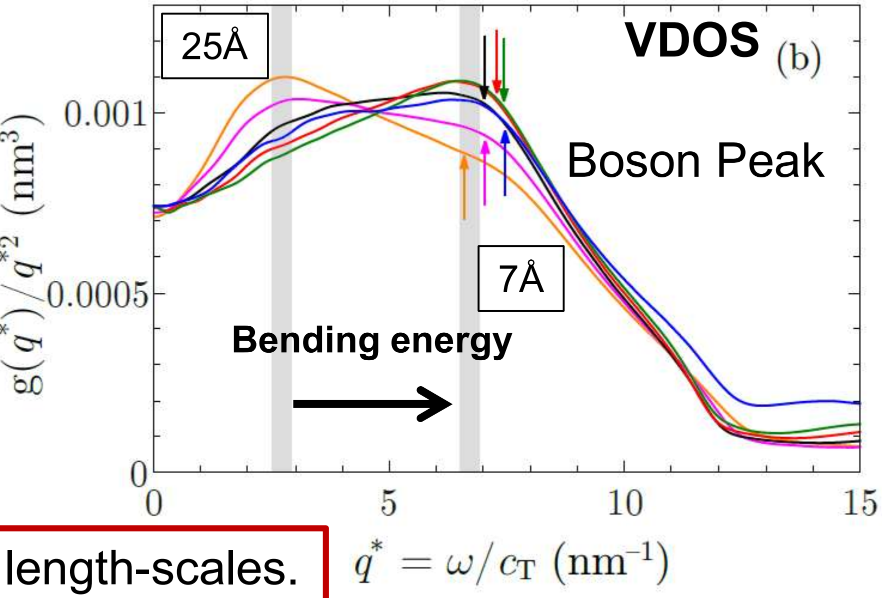


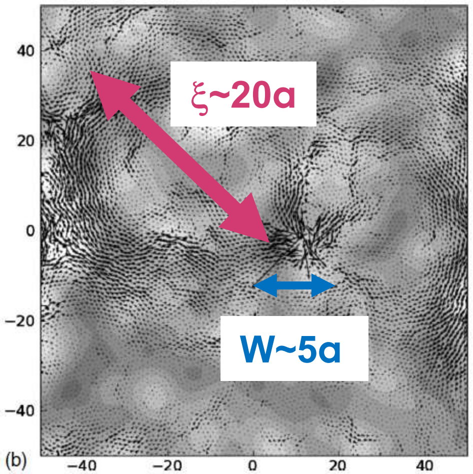
Interatomic interactions:

$$E_{SW} = \Sigma_{(i,j)} f(r_{ij}) + \Sigma_{i,j,k} \Lambda \left(\cos\theta_{jik} + \frac{1}{3} \right)^2 . e^{\gamma.(r_{ij}-a)^{-1} + \gamma.(r_{ik}-a)^{-1}}$$



Λ, bending rigidity

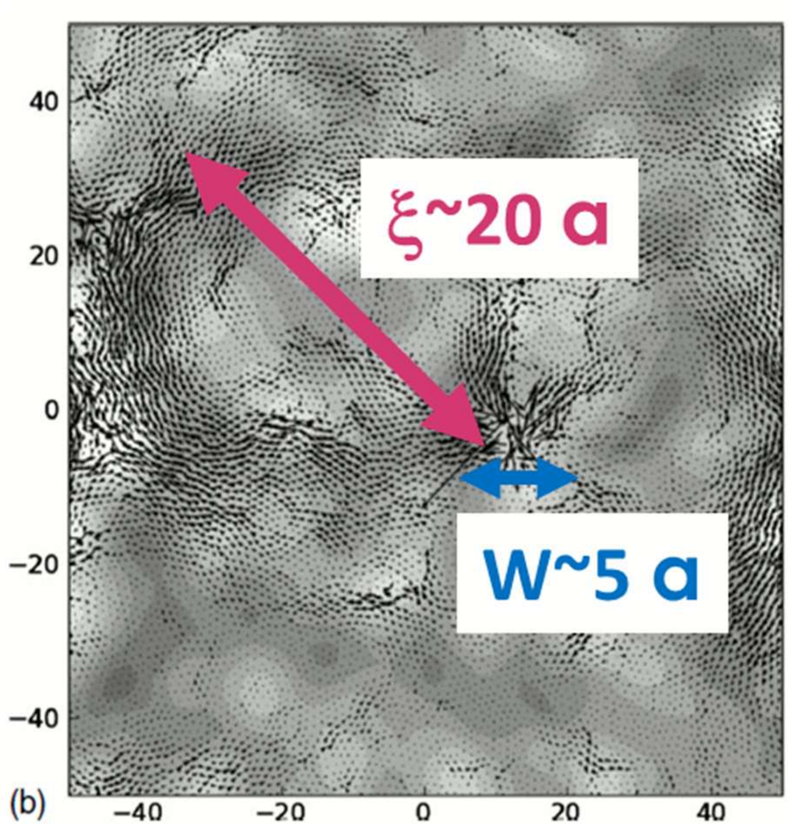




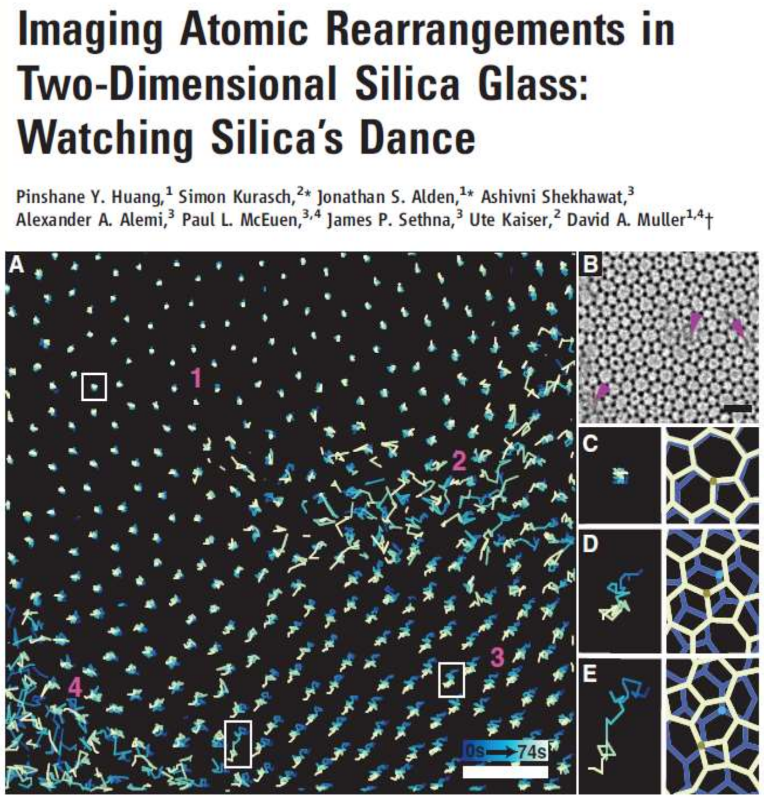
Two different length-scales.
Waves scattering over correlated structures.

Y. Beltukov et al. (2016)
A. Tanguy (2015)

Characteristic Length-scales in Amorphous Solids:



M. Tsamados et al. (2009)

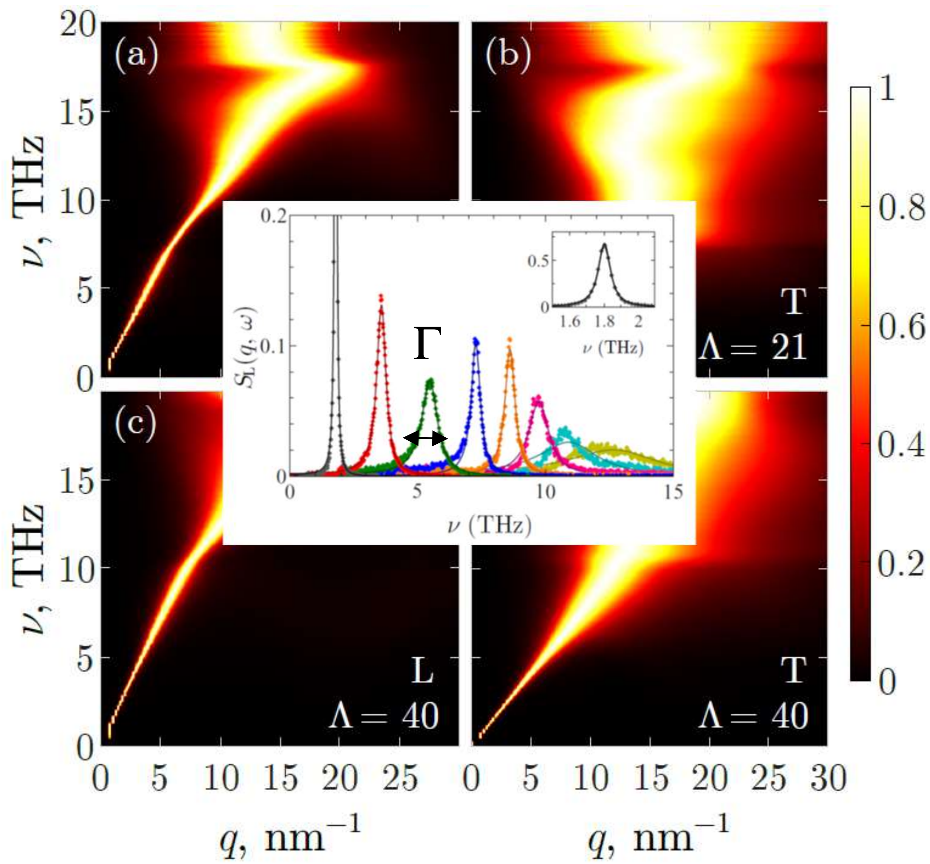


P.Y. Huang et al. Science (October 2013)

Dynamical Structure Factor in **a-Si:**

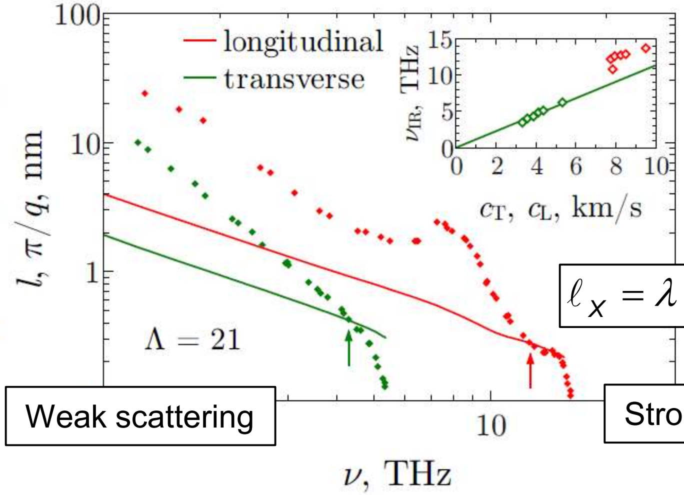
$$S_L(q,\omega) = \int e^{-i\omega t} \left\langle \rho(\bar{q},t).\rho(-\bar{q},0) \right\rangle dt$$

Y. Beltukov et al. (2016)



Damped Harmonic Osc fit:

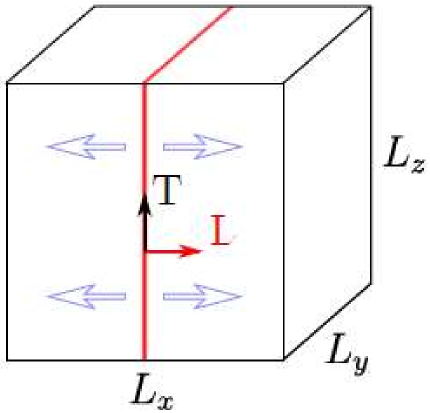
$$S_{L,T}(q,\omega) = \frac{A}{(\omega^2 - \omega_{L,T}^2(q))^2 + \omega^2\Gamma^2}$$

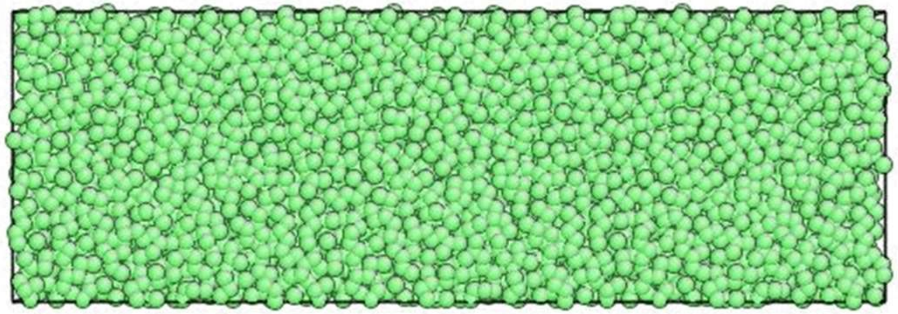


Mean-free Path
$\ell_X = v_g / \Gamma$

Scattering of Transverse and then Longitudinal waves

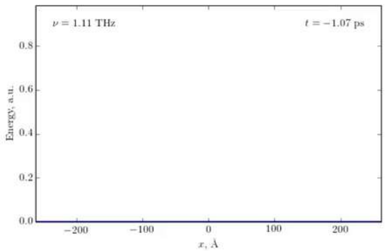
Wave-Packets Dynamics in **a-Si:**



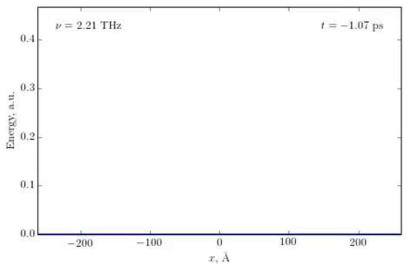


$$f_i^{\mathrm{exc}}(\omega, t) = f_\eta \exp\left(i\omega t - \frac{t^2}{2\tau_{\mathrm{exc}}^2} - \frac{x_i^2}{2w^2}\right)$$

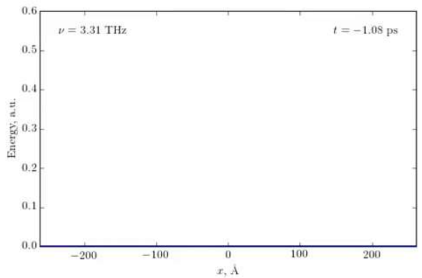
$\omega \nearrow$



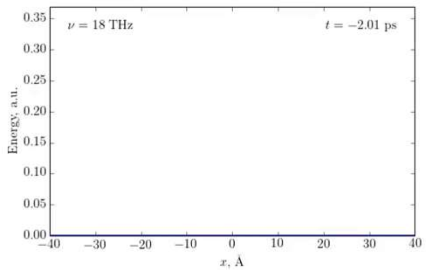
Ballistic Regime
Propagons
$\omega \ll \omega_{IR}$



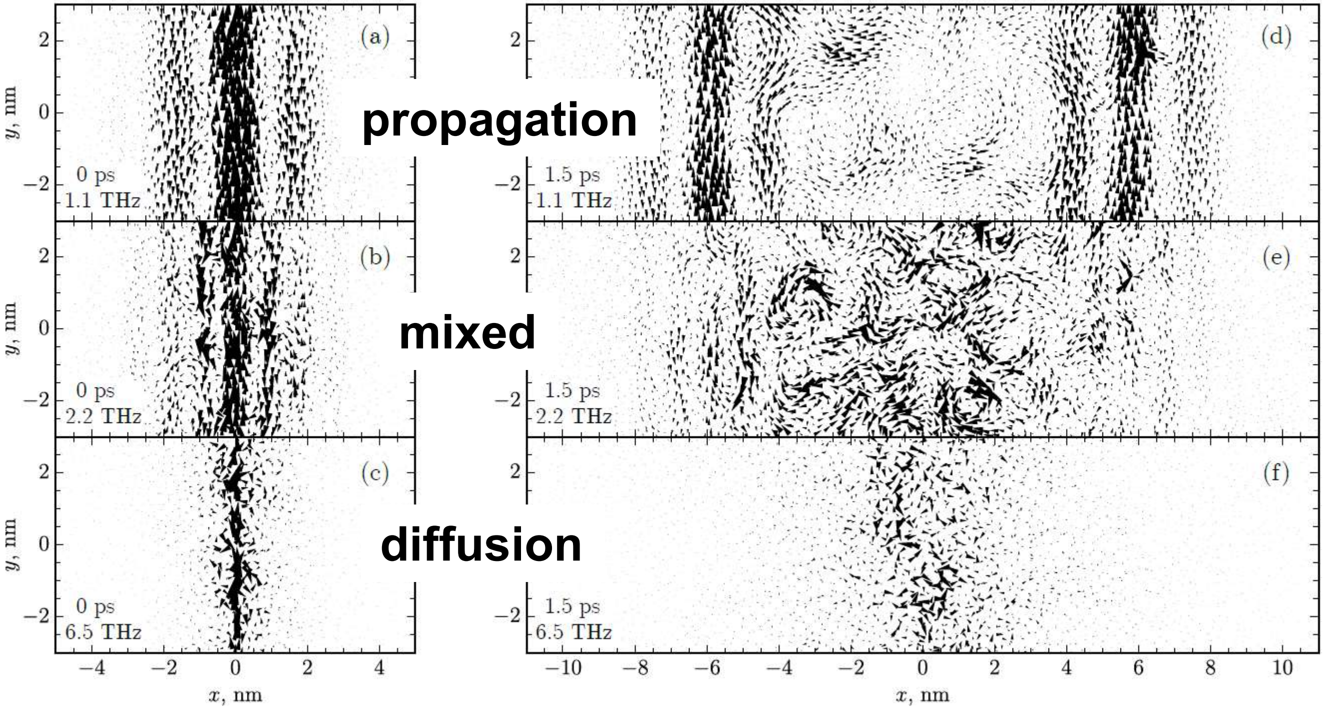
Mixed Regime
(Boson Peak)



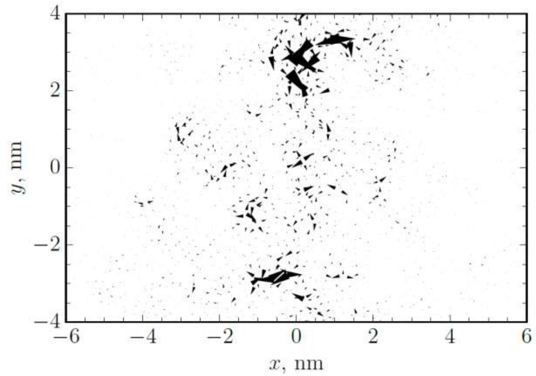
Diffusive Regime
Diffusons
$\omega > \omega_{IR}$



Localisation
Locons
Y. Beltukov et al. (2018)



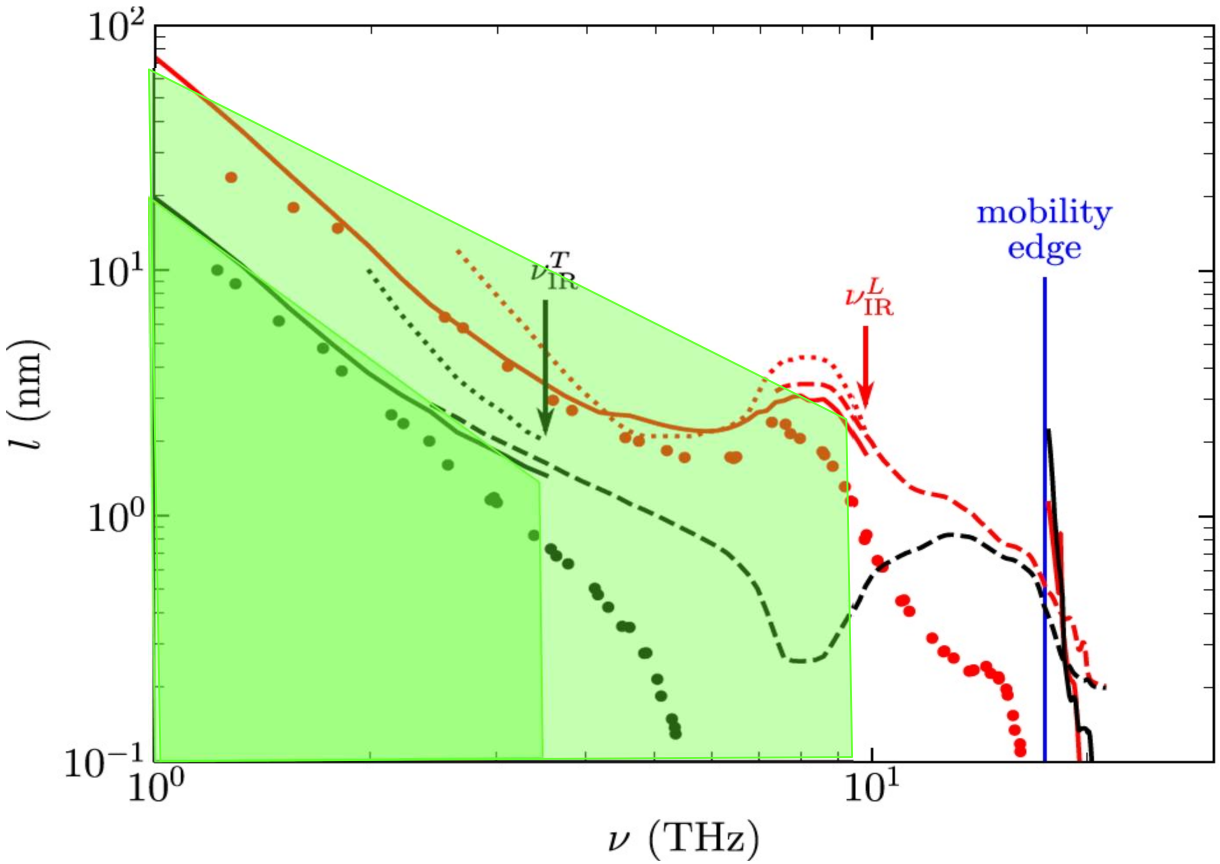
localization



Y. Beltukov et al. (2018)

Attenuation Length

Comparison to the Dynamical Structure Factor



Attenuation length $P_{diff\text{-}prop}(l)=P_{diff\text{-}prop}(0)/e$

Mean-free path (Beer-Lambert)

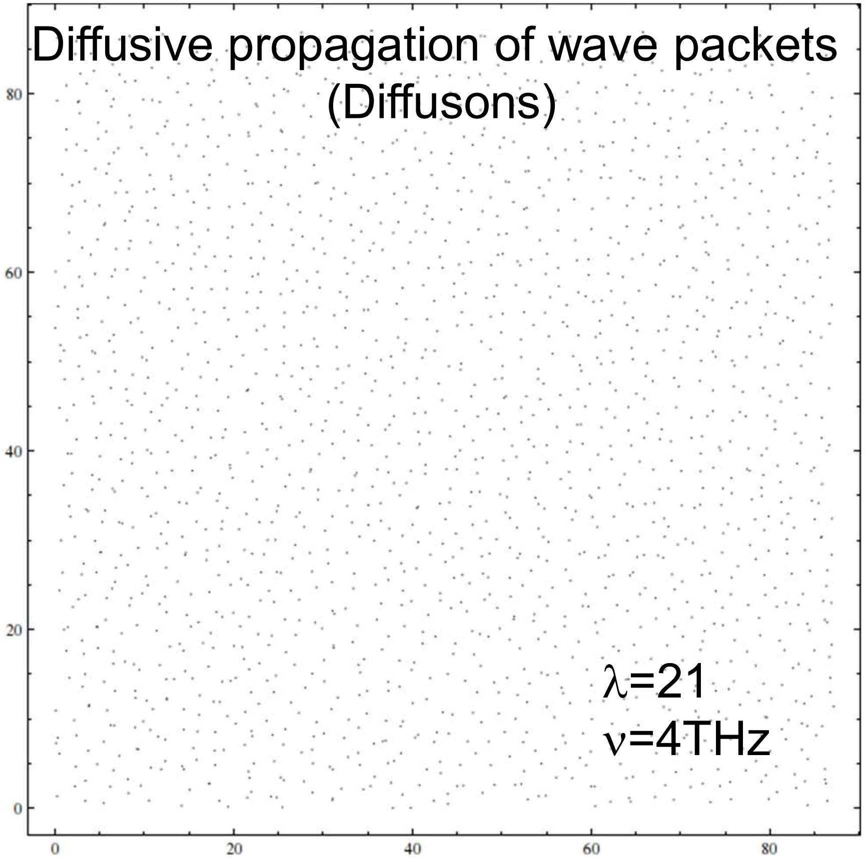
Dyn. Struct. Factor $l = c_{L,T}/\Gamma$

Transportation of Energy even **beyond** the Ioffe-Regel criterion

The Mean-free path (and from the DSF) is defined only in the Ballistic regime

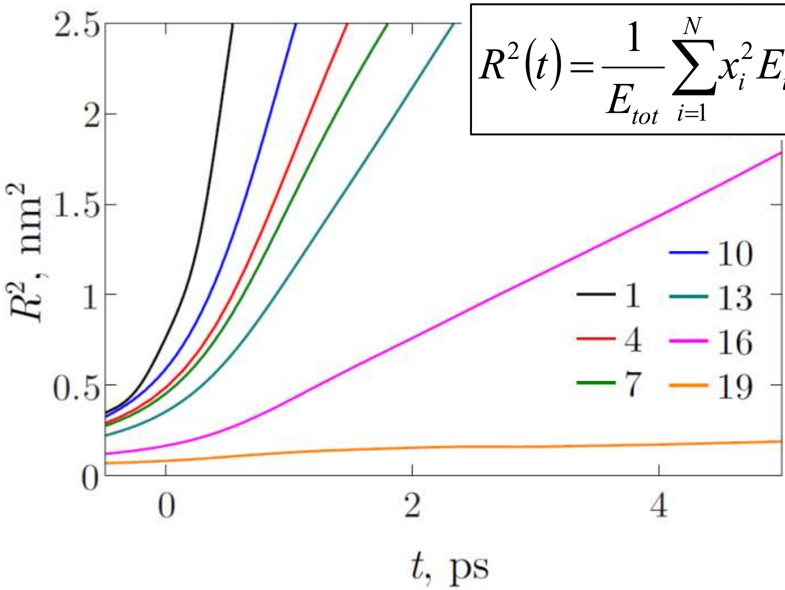
Y. Beltukov et al. (2018)

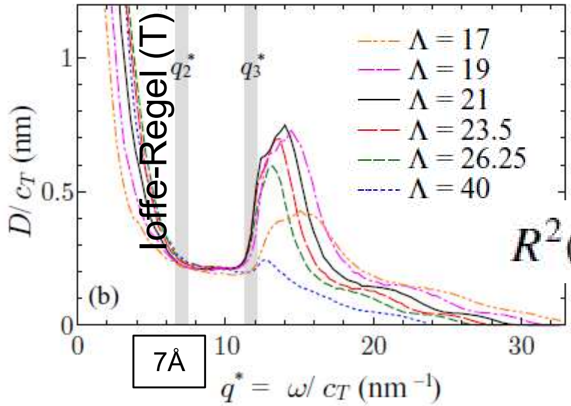
Determination of **Diffusivity** from **Wave-Packets motion**



Minimum **diffusivity** at the Ioffe-Regel criterion of Transverse waves.

$$R^2(t) = \frac{1}{E_{tot}} \sum_{i=1}^{N} x_i^2 E_i(t) \approx D.t$$





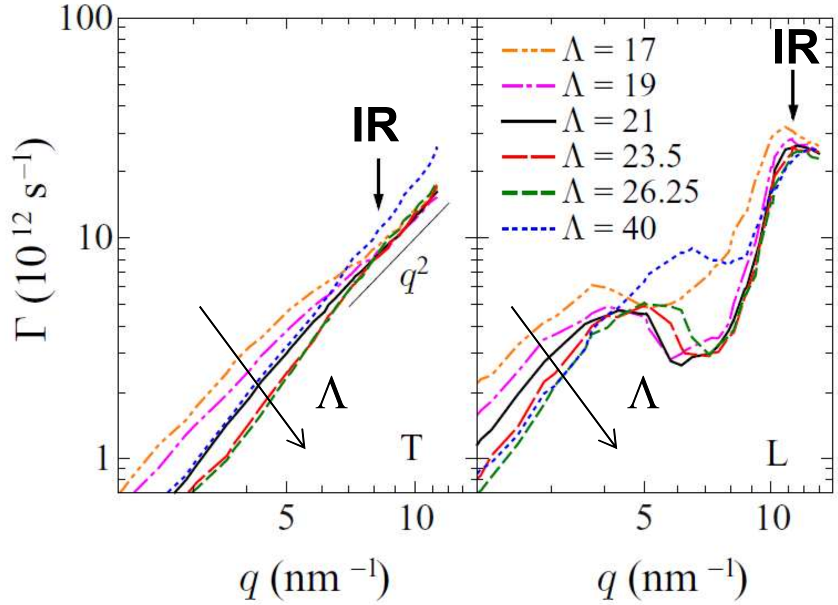
a-Si

$$R^2(t) = 2D(\omega)t$$

Allen and Feldman (1999)

Ballistic Regime

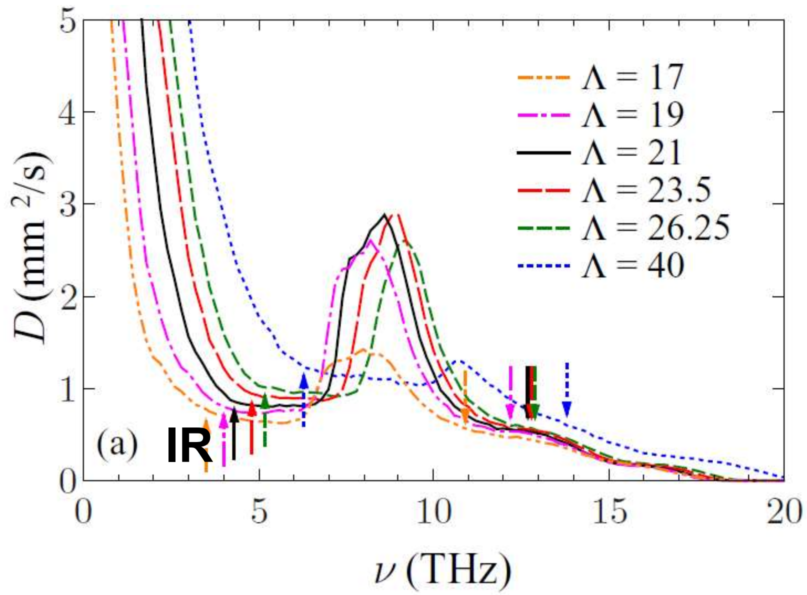
Attenuation for $\omega<\omega_{IR}$



$$\kappa^{prop}(T)=\frac{1}{3}\int_{0}^{\omega_{IR}} C(\omega,T)v_g^{\,2}(\omega)\Gamma^{-1}(\omega)g(\omega)d\omega$$

Diffusive Regime

Diffusivity for $\omega>=\omega_{IR}$

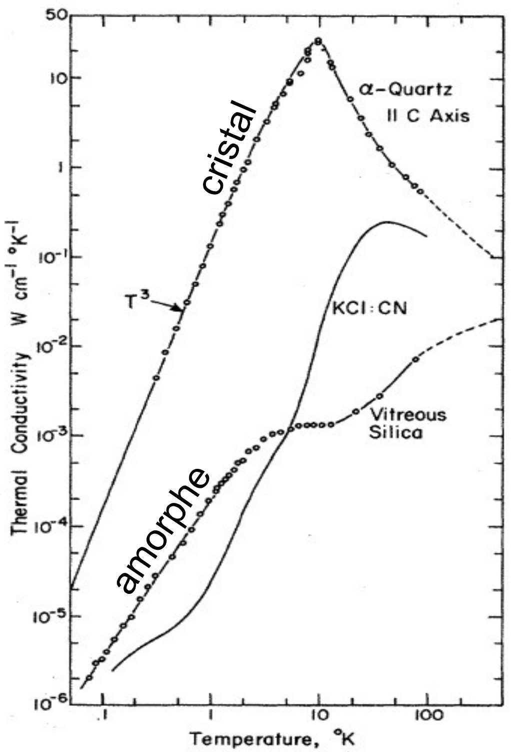


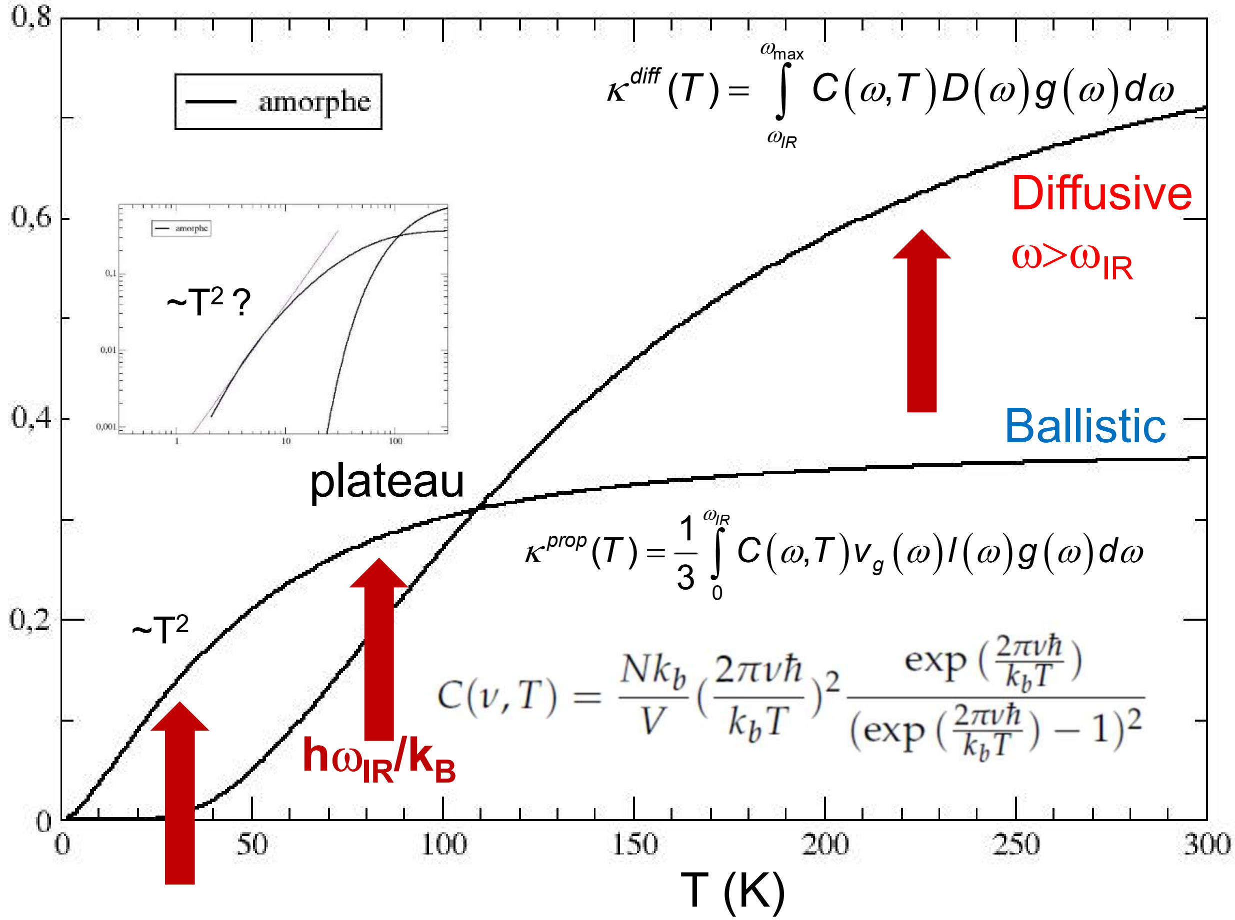
$$\kappa^{diff}(T)=\int_{\omega_{IR}}^{\omega_{max}} C(\omega,T)D(\omega)g(\omega)d\omega$$

J.M. Larkin et al. (2014)

Thermal Conductivity

R.C. Zeller (1971)





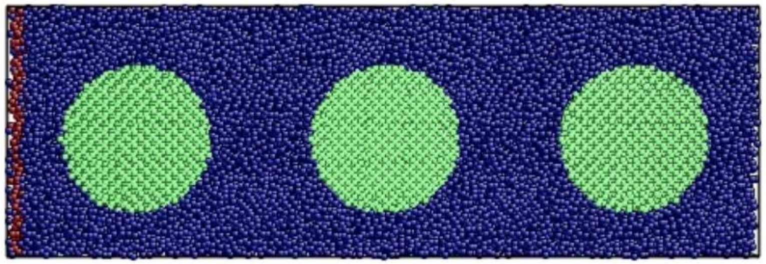
A. Tlili et al. (2019)

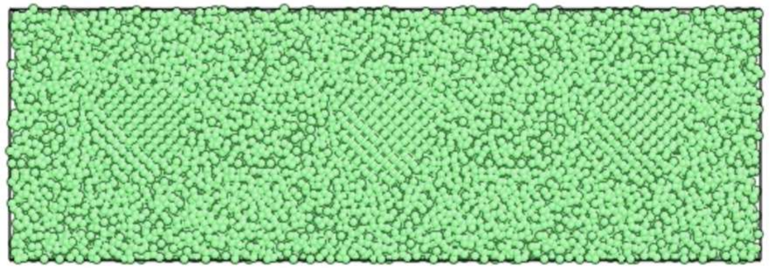
Introduction

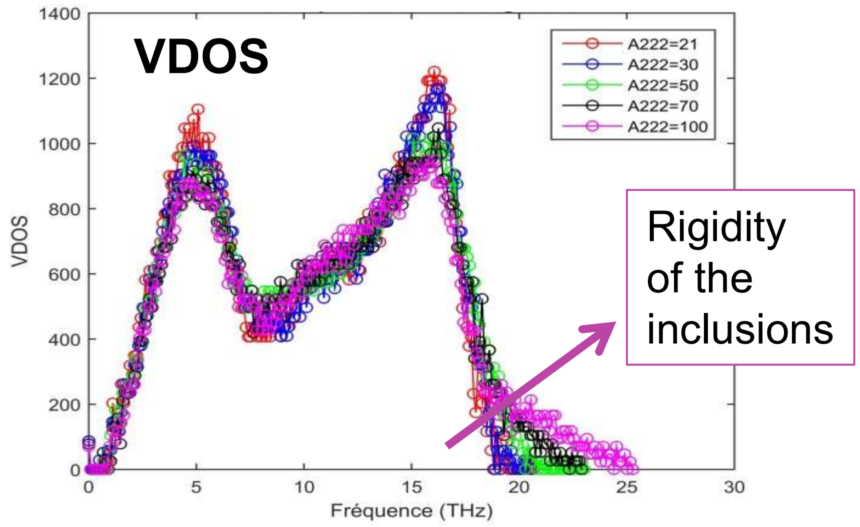
I. Vibrations and Phonons in Amorphous Materials

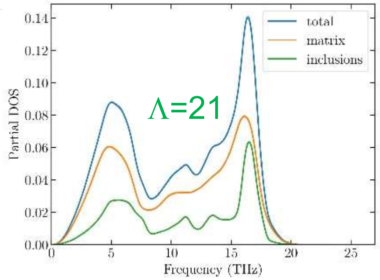
II. Thermal Conductivity in Nanocomposites

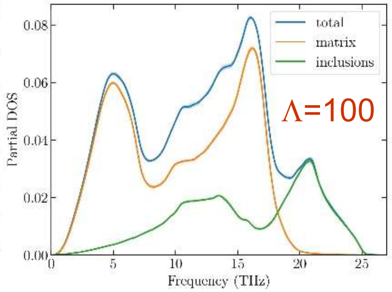
Molecular Dynamics Simulations of the nanocomposite a-Si/c-Si

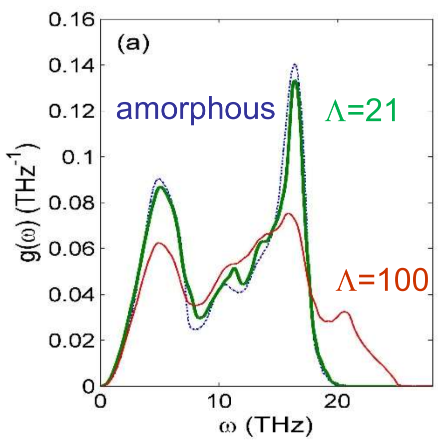


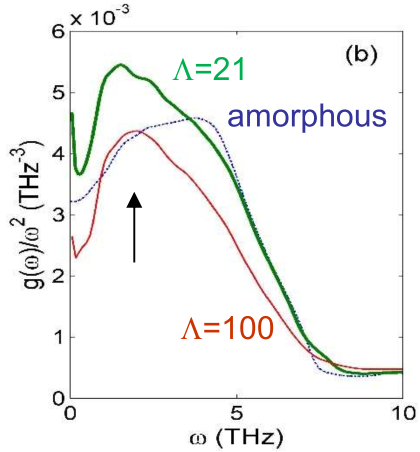










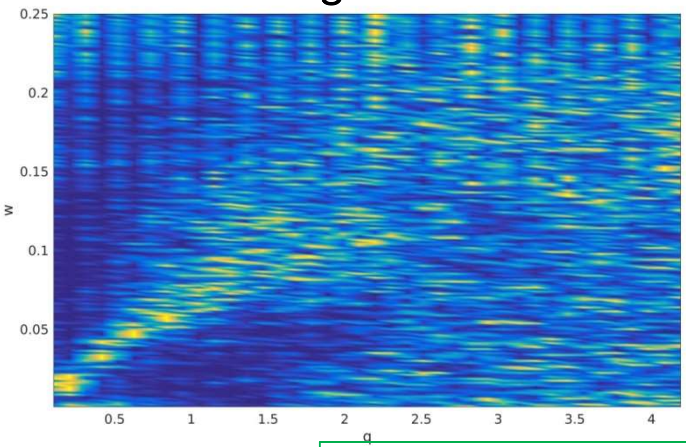


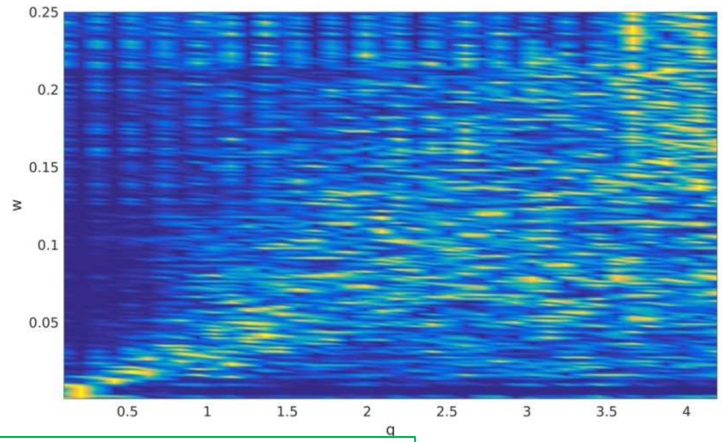
Dynamical Structure Factor in a-Si/c-Si

Longitudinal

Transverse

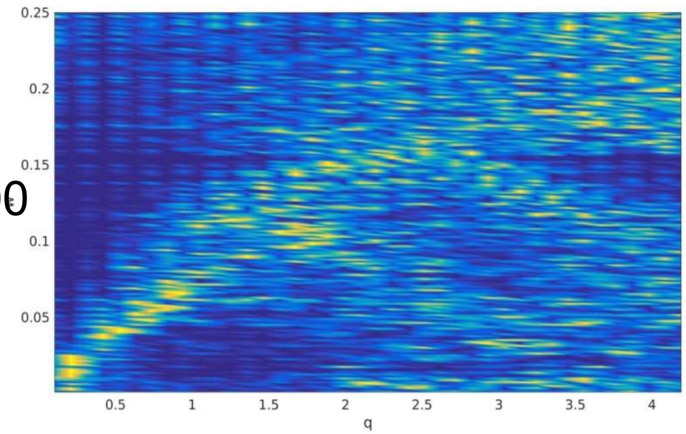
Λ_{Incl}=21

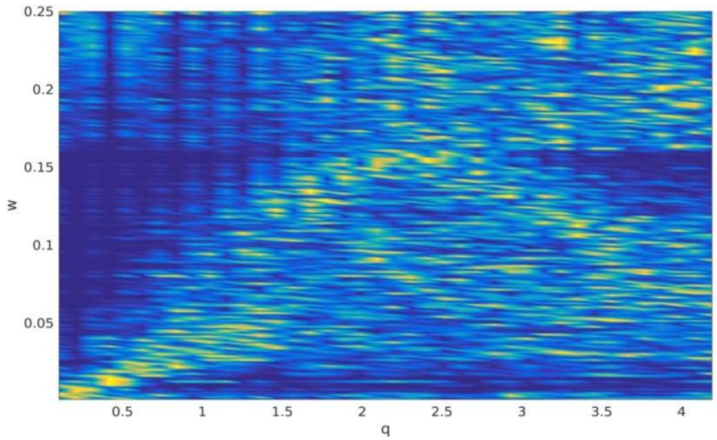




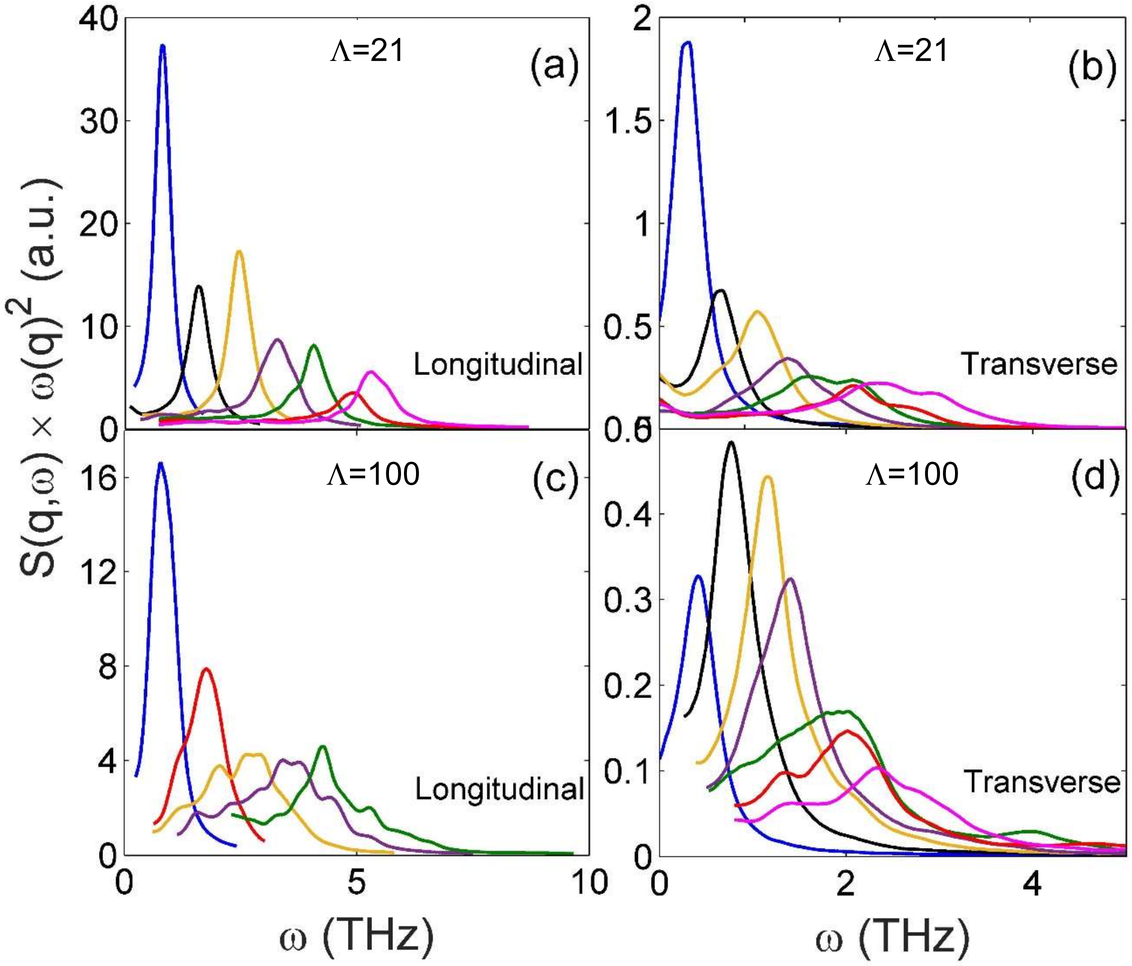
Splitting in the Dynamical Structure Factor

Λ_{Incl}=100

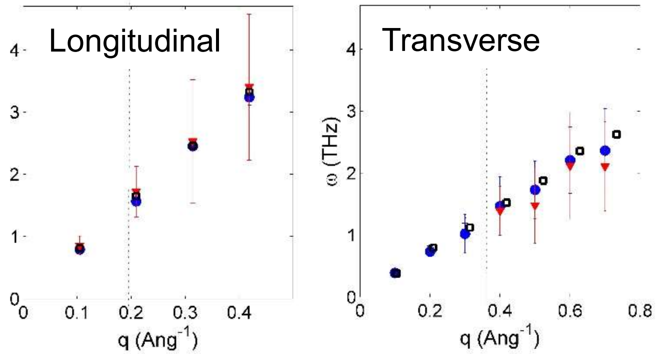




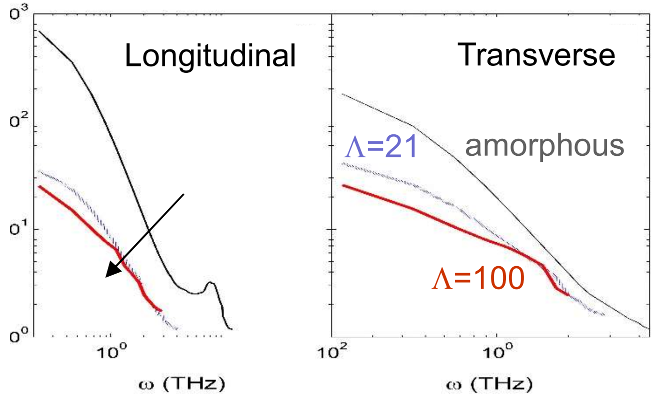
Dynamical Structure Factor in a-Si/c-Si



Dispersion Relation:

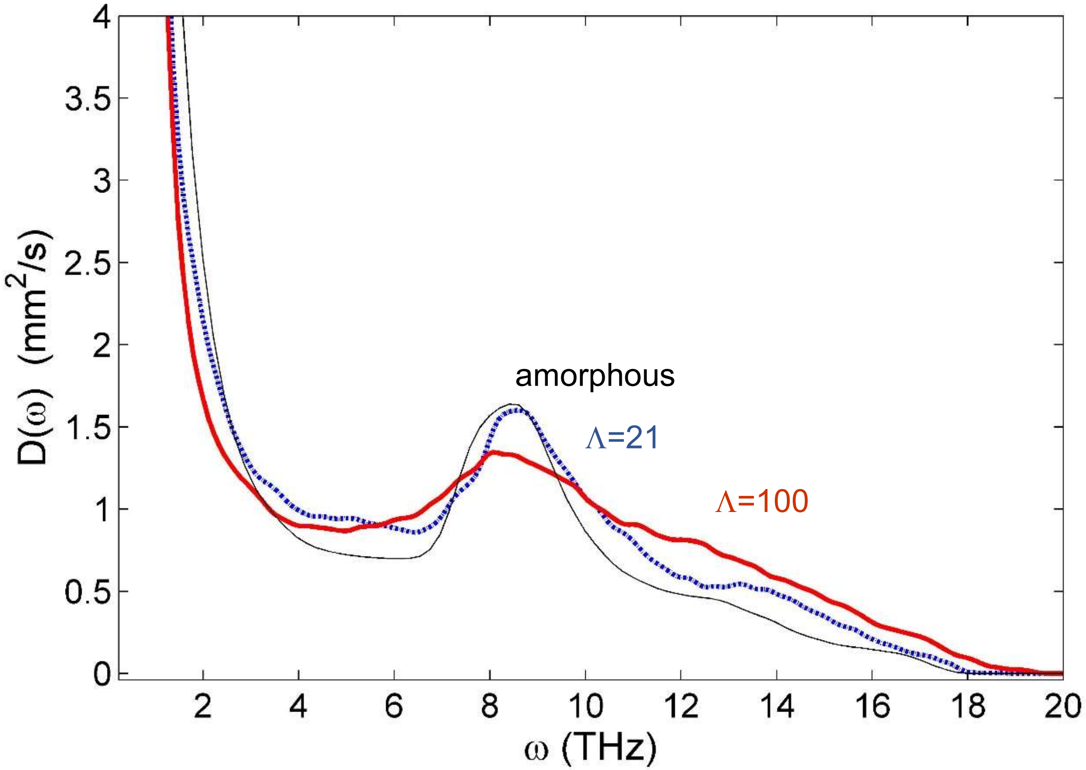


Mean-Free Path:



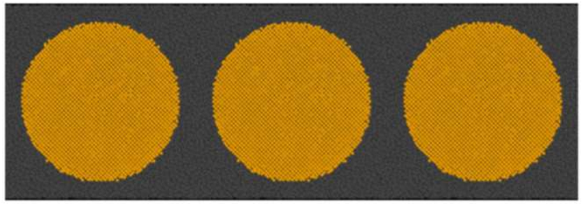
Anticipation of the Ioffe-Regel cross-over

Comparable Diffusivity of Phonons in a-Si/c-Si



Diffusivity dominated by the amorphous matrix

Thermal Conductivity in a-Si / c-Si nanocomposite



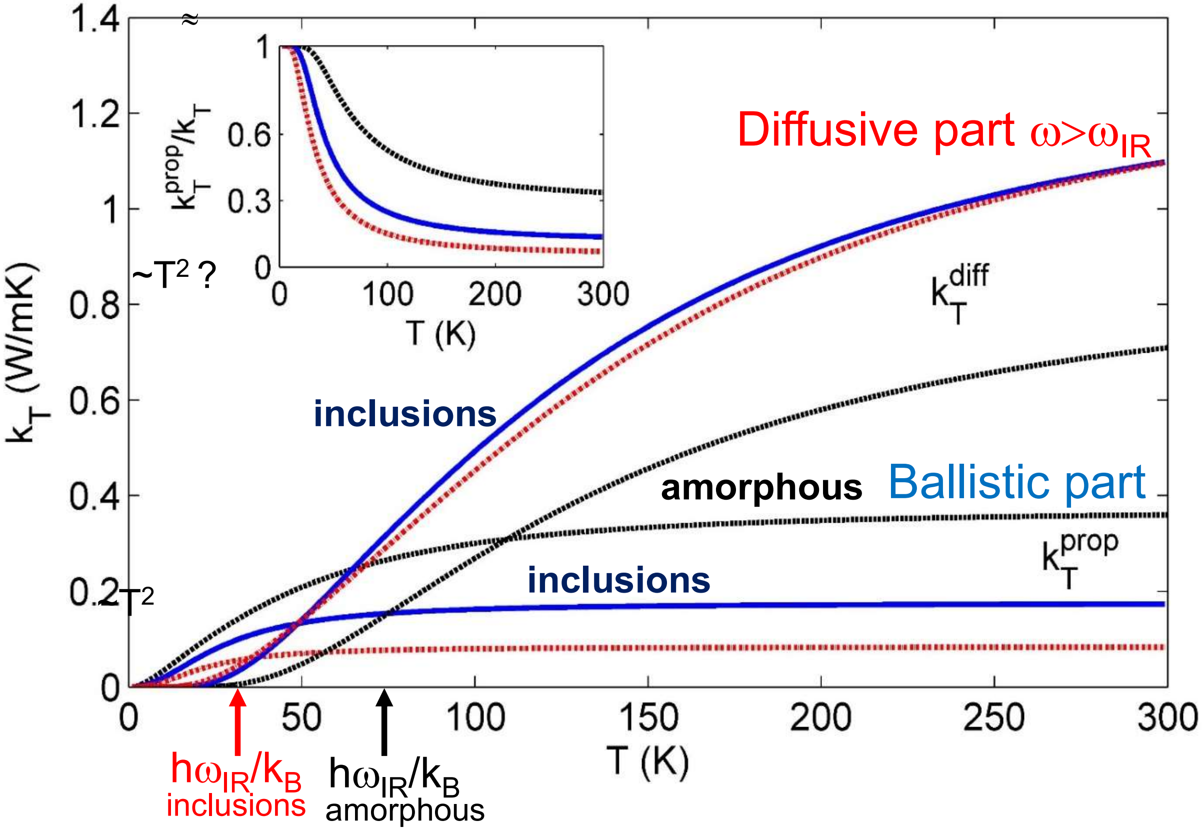
Rigidity contrast:

- (blue line) $E_i / E_m \approx 1$
- (red line) $E_i / E_m \approx 4.6$

The transition frequency

to multiple scattering ω_{IR}

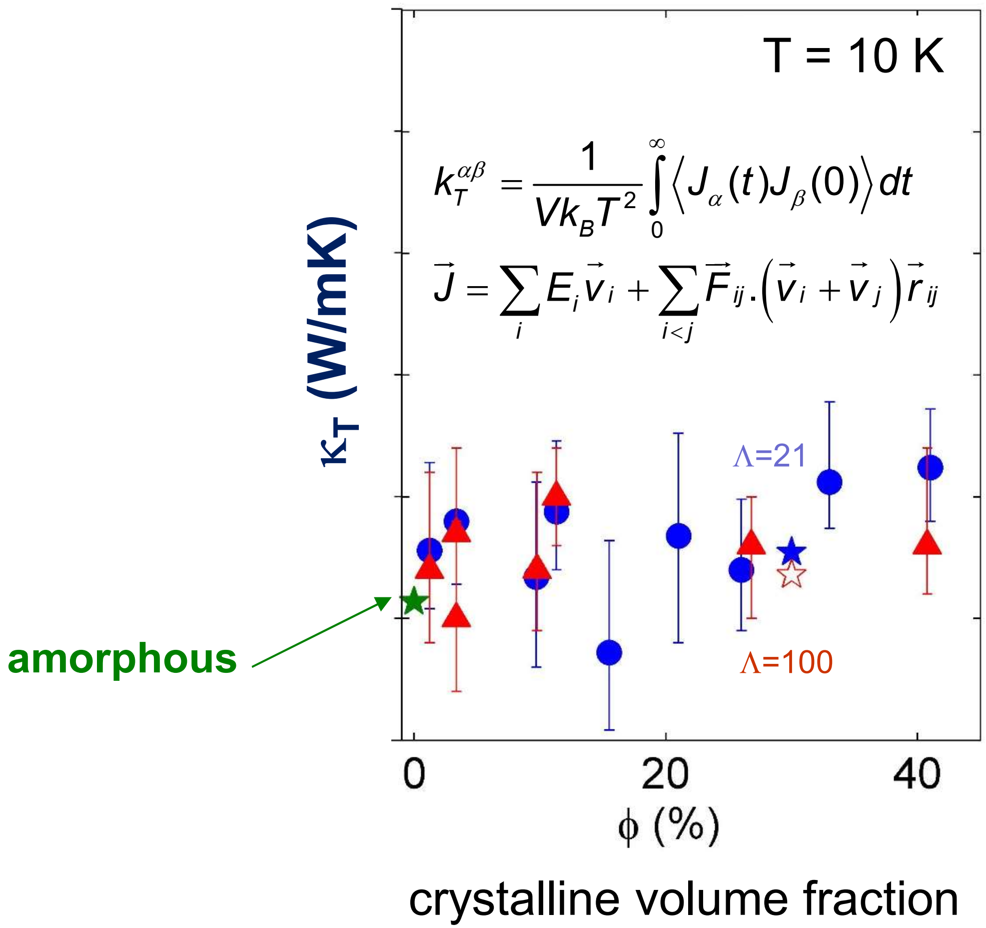
is smaller ↘



Enhancement of the diffusive contribution to Thermal conductivity in the nanocomposite

A. Tlili et al. (2019)

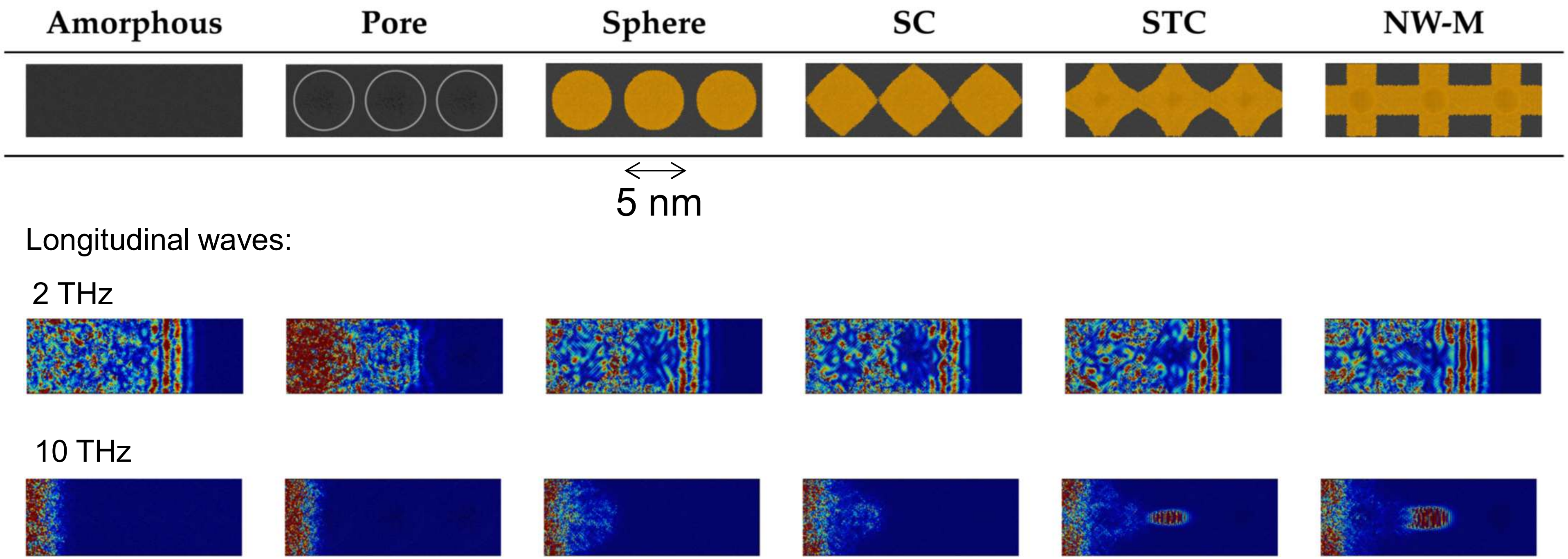
Green-Kubo calculation of Thermal Conductivity



No clear effect
of the Nano-inclusions
on the Thermal Conductivity
in the low T regime

Thermal Conductivity in a-Si / c-Si nanocomposite

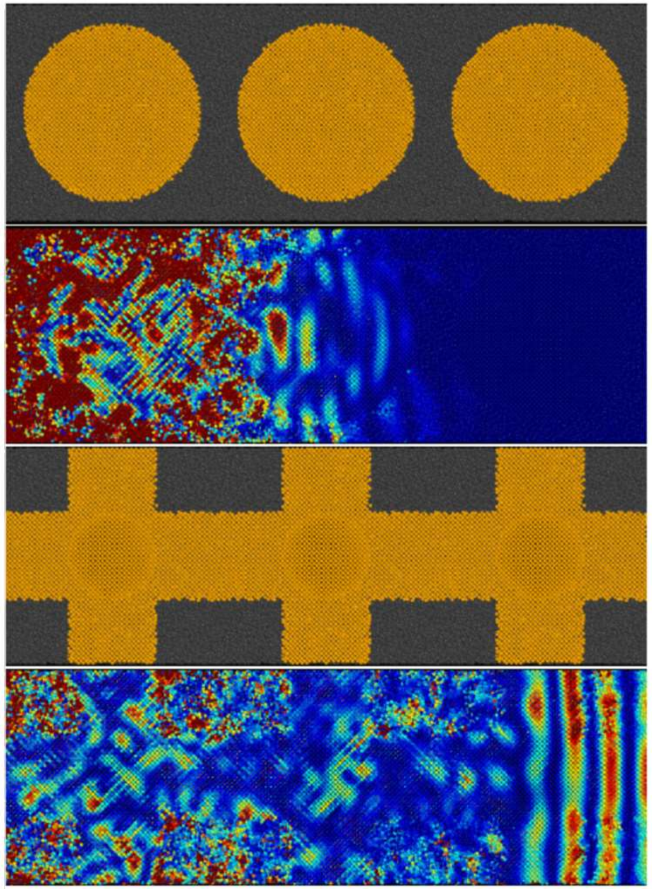
Role of the interconnections between the inclusions



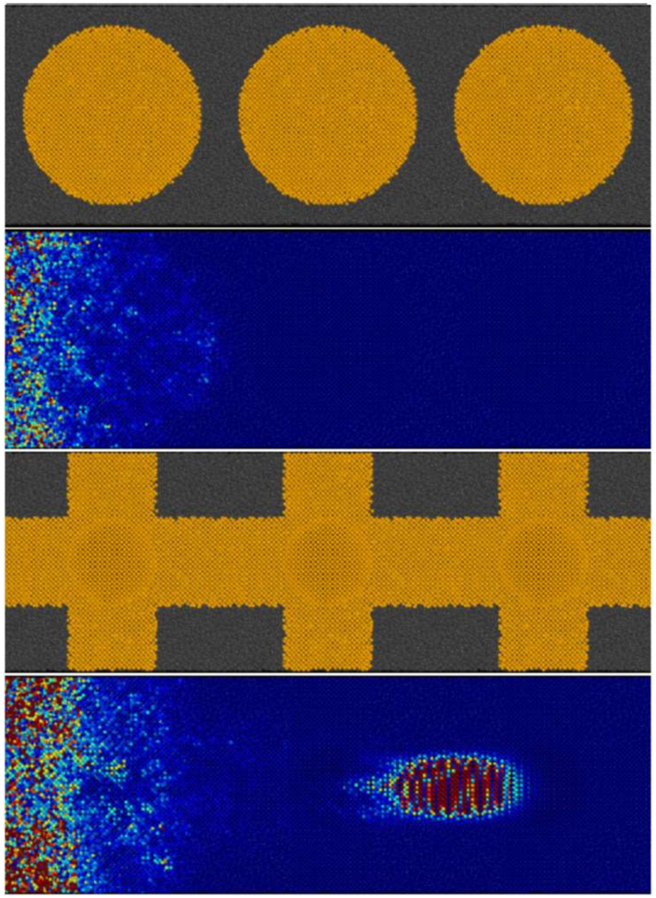
P. Desmarchelier et al. (2021)

Wave Packets propagation in case of percolating vs. Non-percolating nanostructure

2 THz

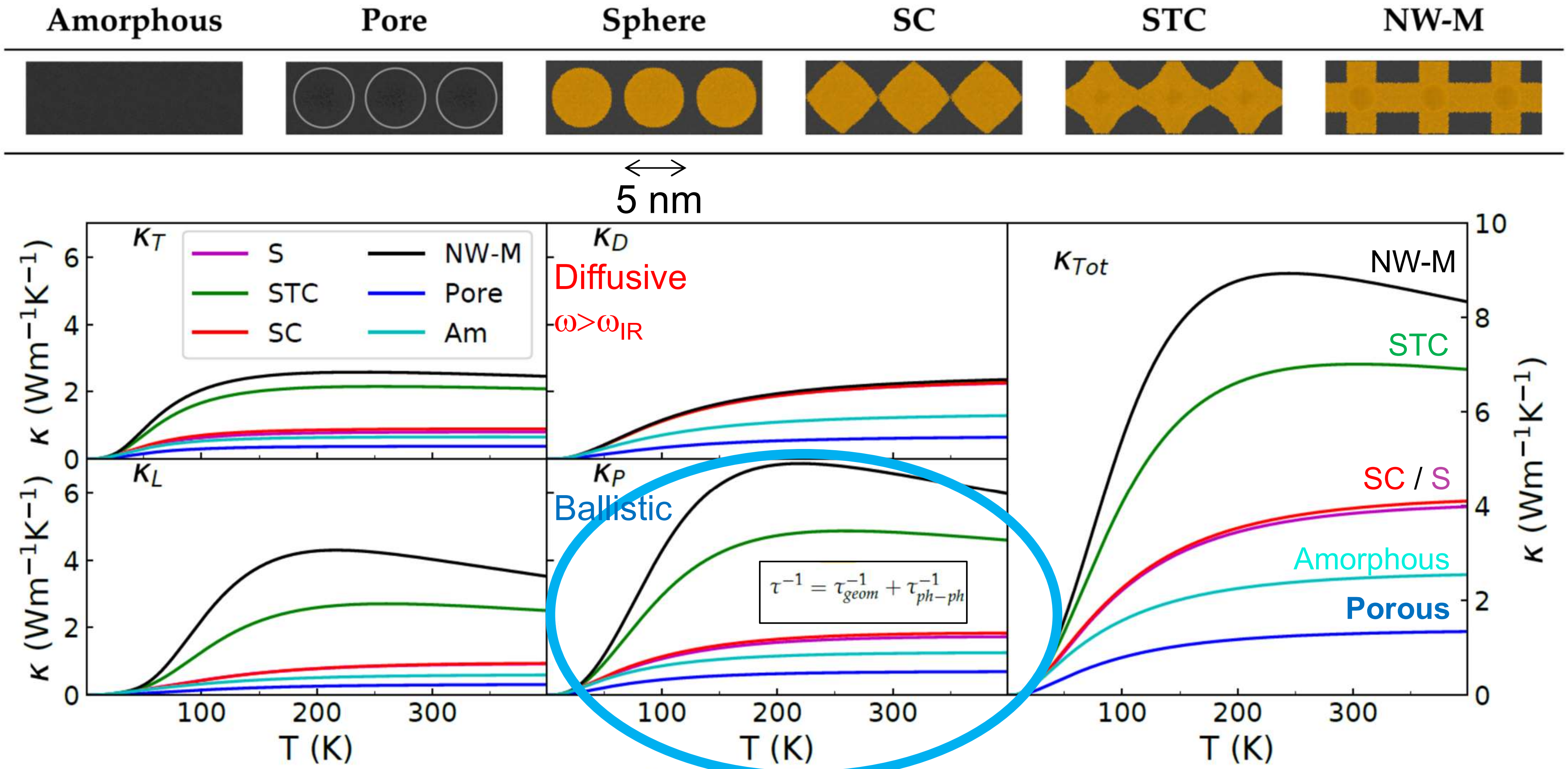


10 THz



P. Demarchelier et al. (2020)

Thermal Conductivity in a-Si / c-Si nanocomposite

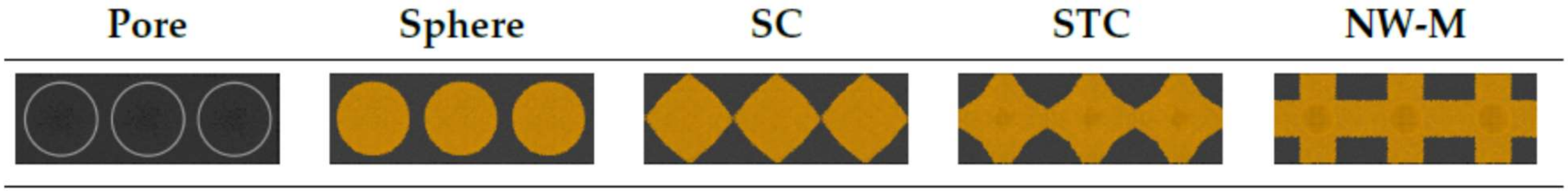


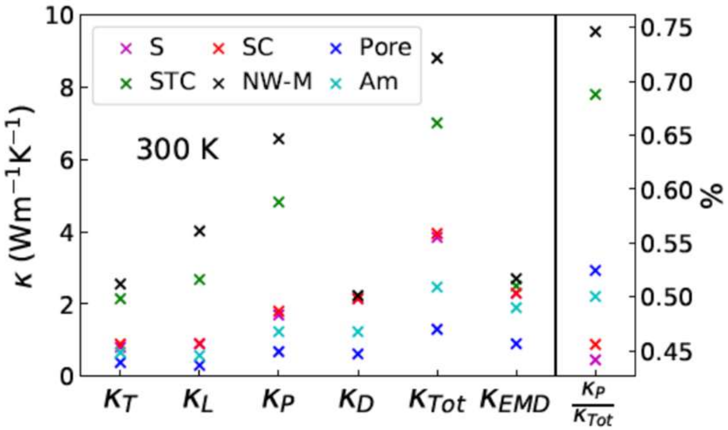
$$\tau_{ph-ph}^{-1} = P(2\pi\nu)^2 T \exp(-C_U/T)$$

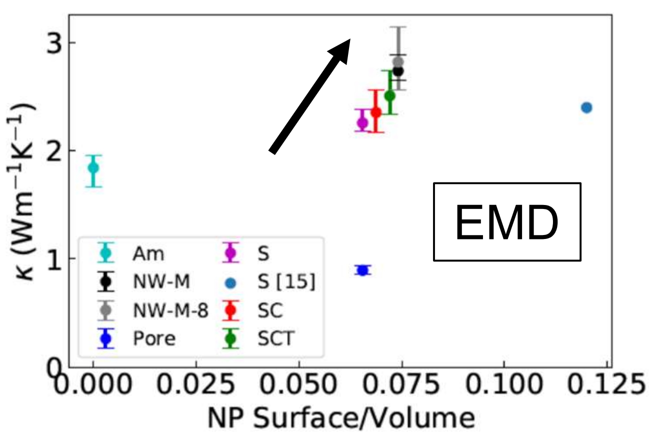
P. Desmarchelier et al. (2021)

Thermal Conductivity in a-Si / c-Si nanocomposite

Role of the interconnections between the inclusions



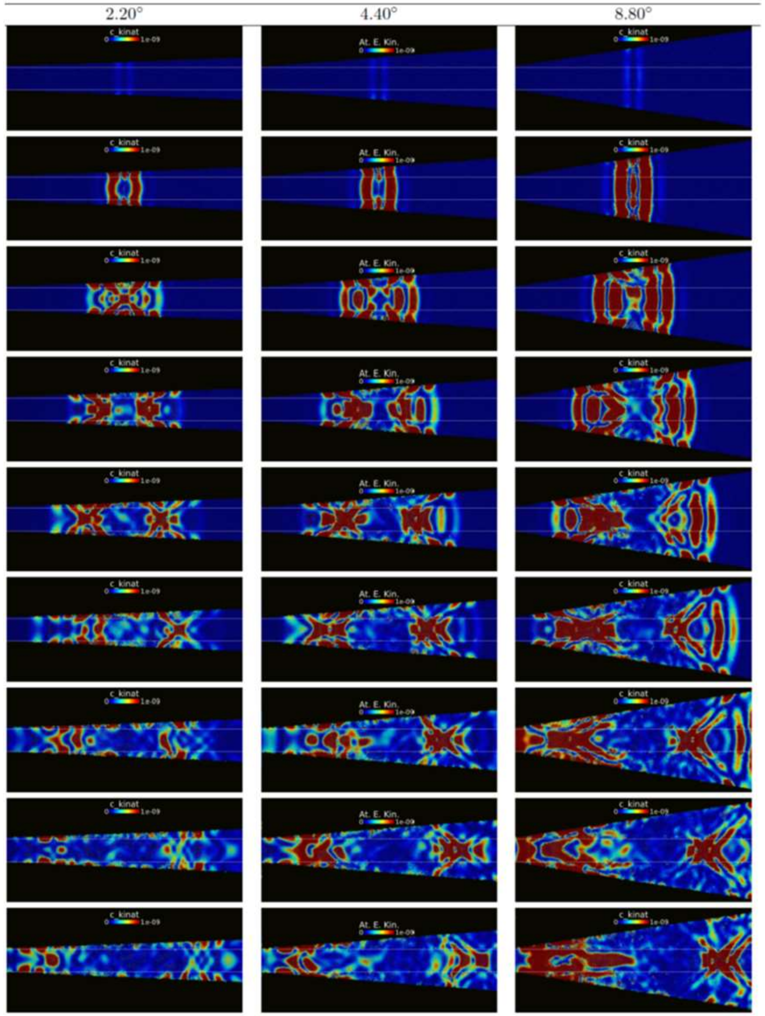


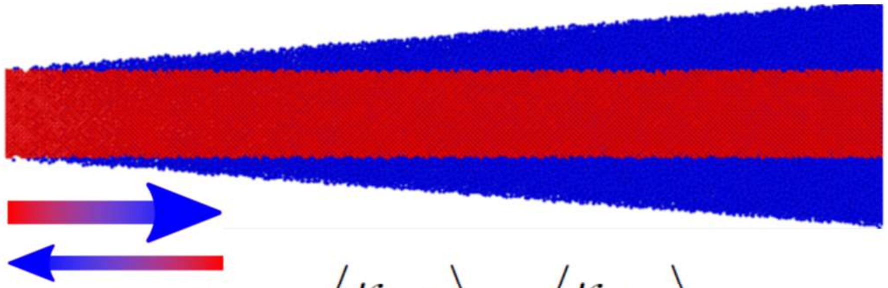


P. Desmarchelier et al. (2021)

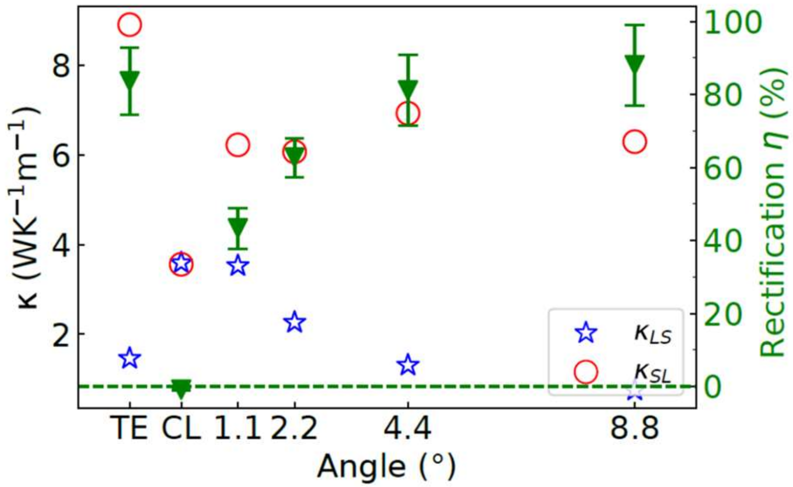
Thermal Rectification in c-Si nanowire with amorphous shell

2 THz





$$\eta = \frac{\langle \kappa_{SL} \rangle - \langle \kappa_{LS} \rangle}{\langle \kappa_{SL} \rangle}$$



P. Demarchelier et al. (2020)

Conclusion

Phononic contribution to Thermal Conductivity in Amorphous Solids:

Low scattering, **strong scattering** and **localization** of wave-packets in amorphous materials:
- the **Ioffe-Regel** cross-over is the transition between low and strong scattering
- the **Mobility Edge** is the transition to localization.

Propagative contribution to **Thermal Conductivity** dominates at low frequencies. Above the Boson Peak frequency, the **diffusive contribution** dominates.

In Nanocomposites:

The presence of **nanoinclusions** changes the **ratio** between the propagative and the diffusive contribution to thermal conductivity.

The **interconnection** (percolation) between the inclusions has a crucial effect.

Playing with **interfaces** paves the way for **thermal management** (rectification)

Bibliography

A. Tanguy, J.P. Wittmer, F. Léonforte and J.-L. Barrat, PRB *66,* 174205-1-17 (2002)
A. Tanguy, B. Mantisi and M. Tsamados, EPL *90* 16004-1-5 (2010)
D. Rodney, A. Tanguy and D. Vandembroucq, MSMSE Topical *19,* 083001 (2011)
A. Tanguy, JOMJ *67*, 1832 (2015)
T. Damart, V. Giordano and A. Tanguy, PRB *92,* 094201 (2015)
Y. Beltukov, C. Fusco, D. Parshin and A. Tanguy, PRE *93*, 023006 (2016)
T. Damart, A. Tanguy and D. Rodney, PRB *95*, 054203 (2017)
A. Tlili et al. Acta Materialia *136*, 425 (2017)
Y. Beltukov, D. Parshin, V. Giordano and A. Tanguy, PRE *98*, 023005 (2018)

H. Luo, V. Giordano, A. Gravouil and A. Tanguy, NanoMaterials *9*, 1471 (2019)
A. Tlili, V. Giordano, Y. Beltukov, S. Merabia, P. Desmarchelier and A. Tanguy, NanoScale *11* (2019)
P. Desmarchelier, A. Tanguy and K. Termentzidis, PRB *103*, 014202 (2021)

Thank You