Finite-size effects in the phonon density of states of nanostructured germanium

TECHNISCHE UNIVERSITÄT DARMSTADT

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Material Modeling

tud mm mawi

About me

Material science building

Our group

Introduction

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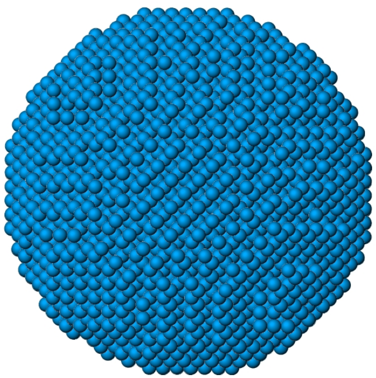
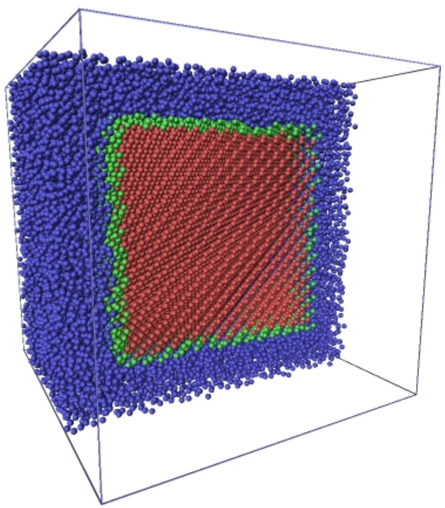
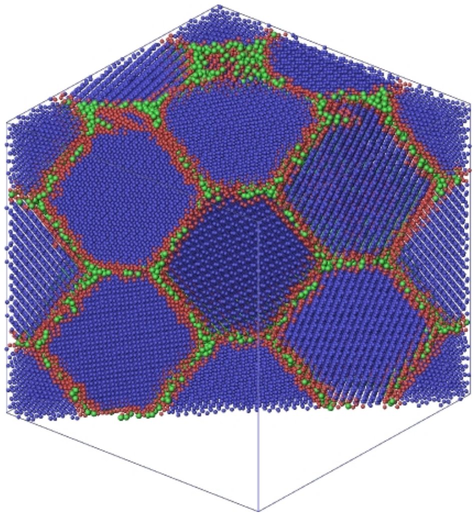
The studied nanostructures



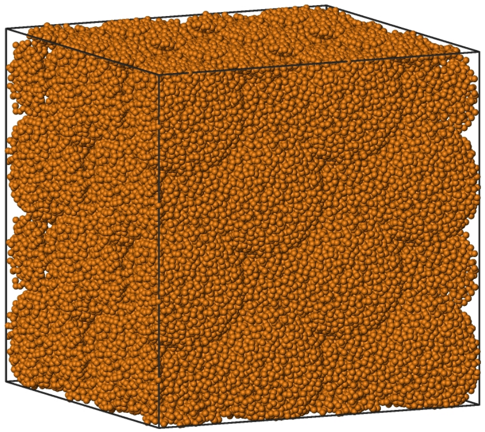
Nanoparticles



Embedded nanoparticles



Nanocrystals

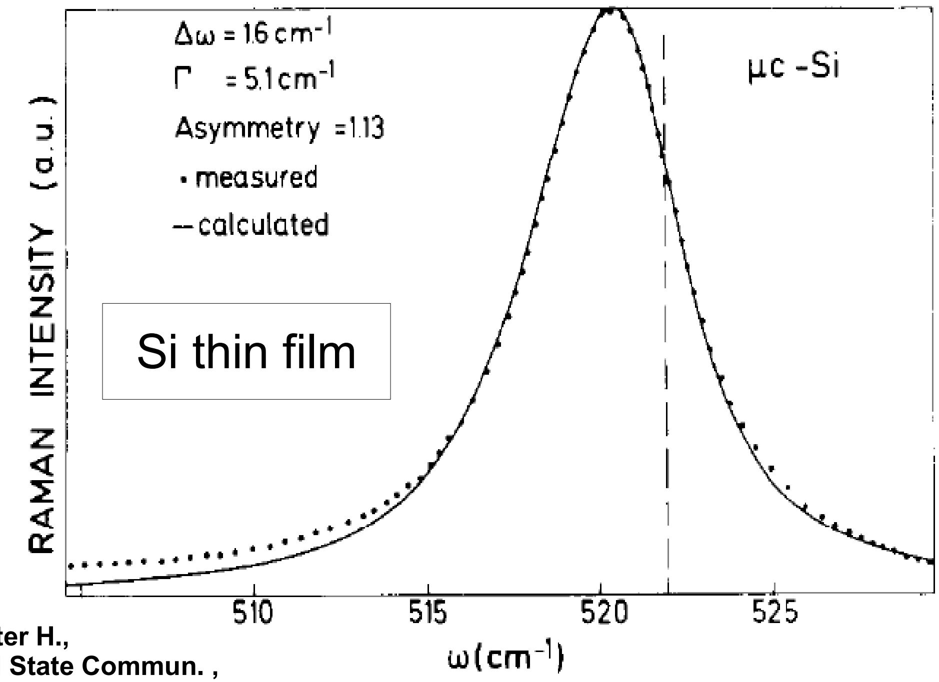


Nanoglasses

Literature review

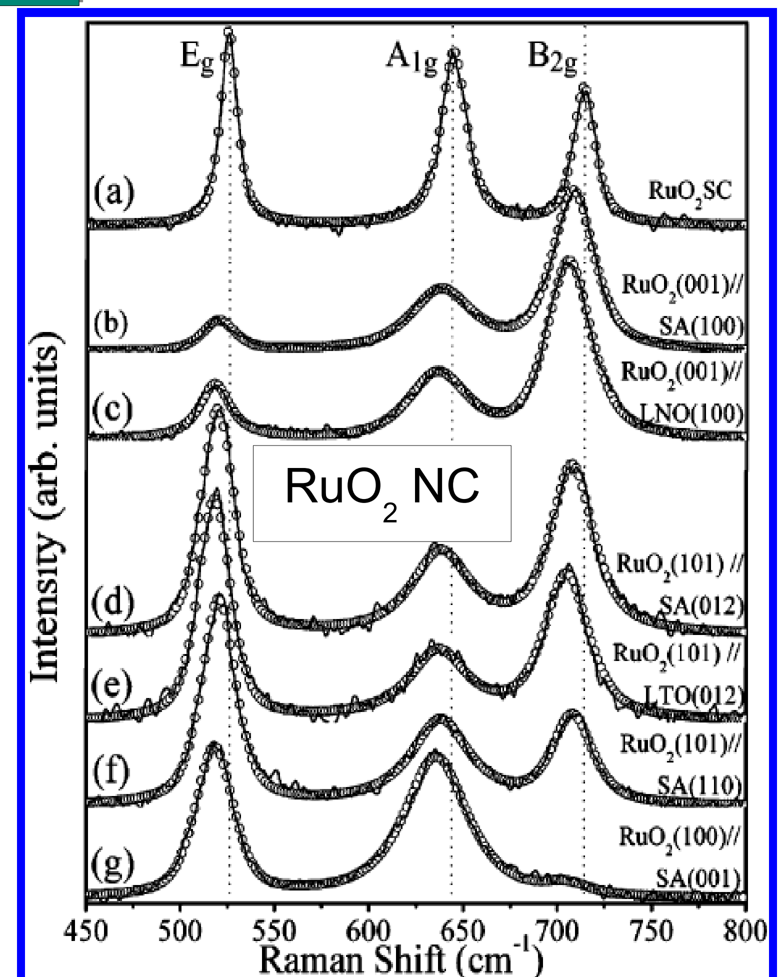
TECHNISCHE UNIVERSITÄT DARMSTADT

Raman red-shift



Richter H.,
Solid State Commun. ,
39, 625, (1981)

Raman red-shift due to the confinement effect



Korotcov A.,
Crystal Growth & Design,
2501, 42, (2006)

Raman red-shift due to the surface stress

Literature review

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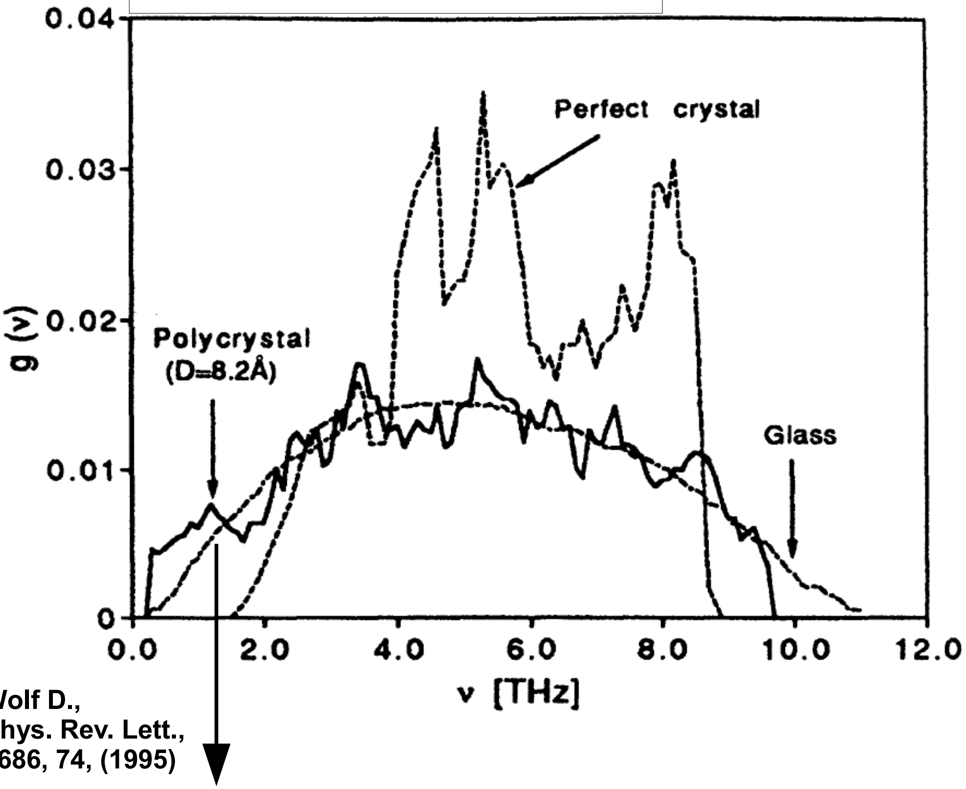
Size effects in the PDOS

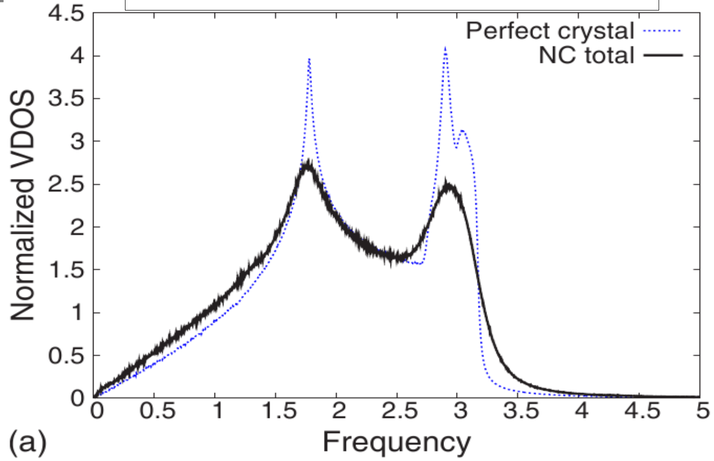
NC Cu - LJ-potential

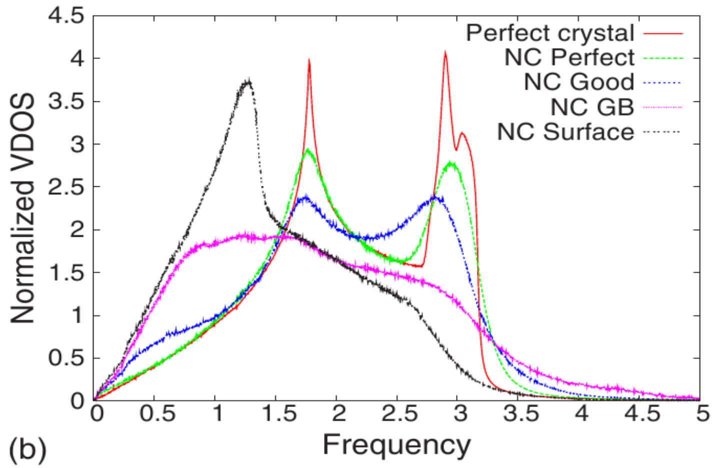


Wolf D., Phys. Rev. Lett., 4686, 74, (1995)

Finite system size effects

NC models - LJ-potential





Hudon C., Phys. Rev. B, 76, 046409, (2007)

PDOS changes only due to the GBs

Literature review

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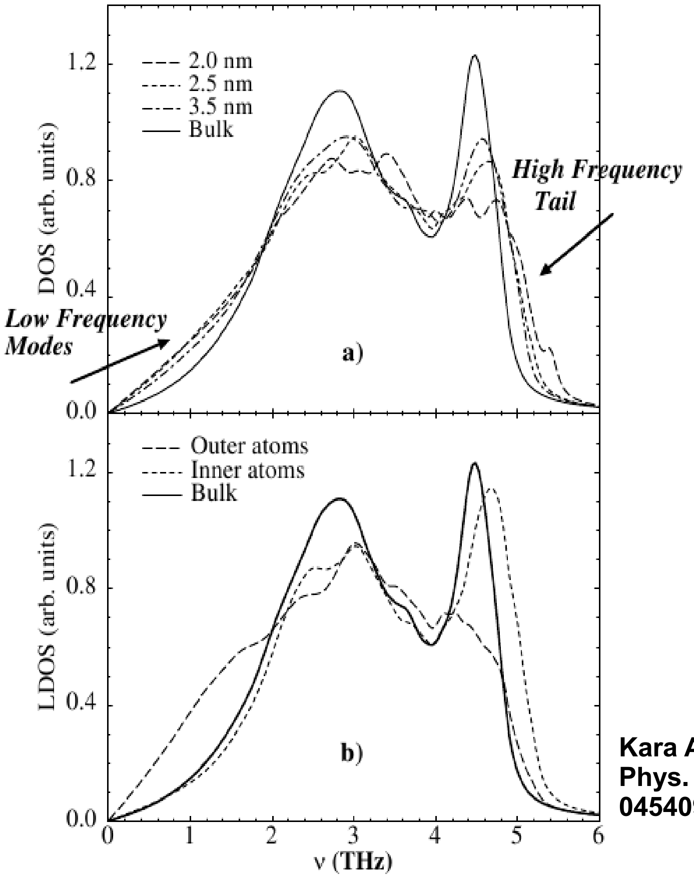
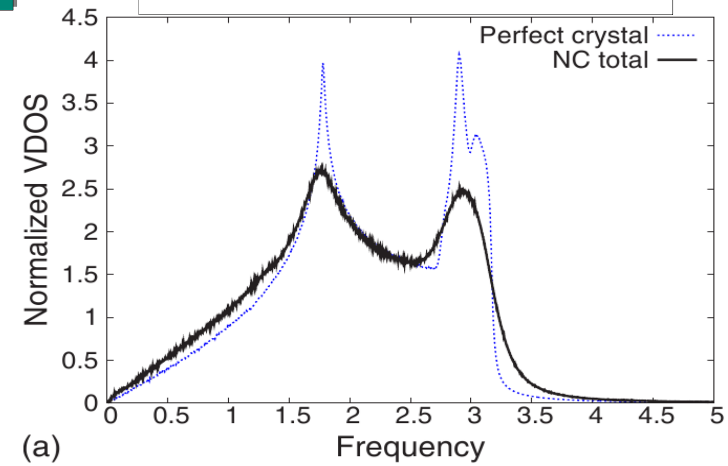
Size effects in the PDOS

Ag free particle - EAM potential

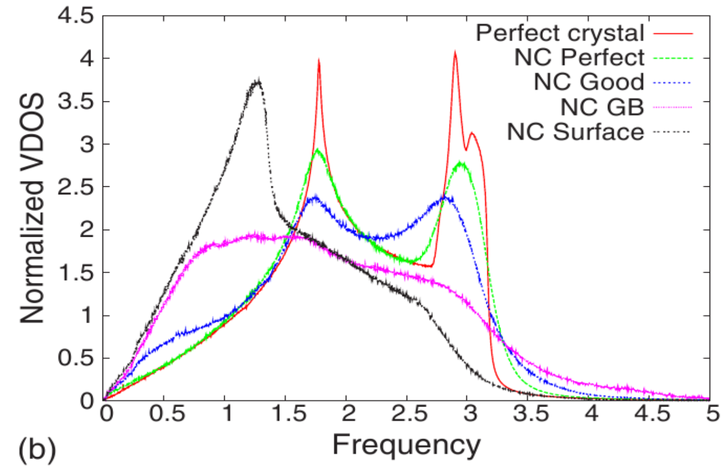


Kara A., Phys. Rev. Lett, 81, 045409, (1998)

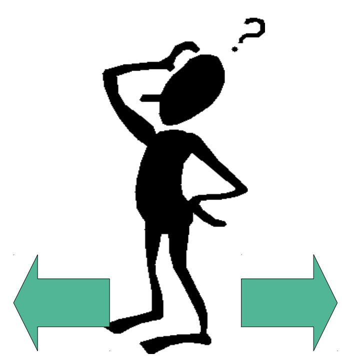
NC models - LJ-potential



Hudon C., Phys. Rev. B, 76, 046409, (2007)



PDOS changes due to the free surface



PDOS changes due to the GBs

Open questions

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- What are the finite size effects that lead to changes in the PDOS of nanostructured materials?
- How do these size effect influence the PDOS?
- Is it possible to obtain information about structure and properties of nanoglasses by studying vibrational properties?

Methods

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• PDOS of nanoparticles, nanocrystals, embedded nanoparticles and nanoglasses are studied by means of MD simulations.

• Ge covalently bonded material using theTersoff interatomic potential [1].

• PDOS, G(ω), was calculated by the Fourier transform of the velocity autocorrelation Z(t) [2],

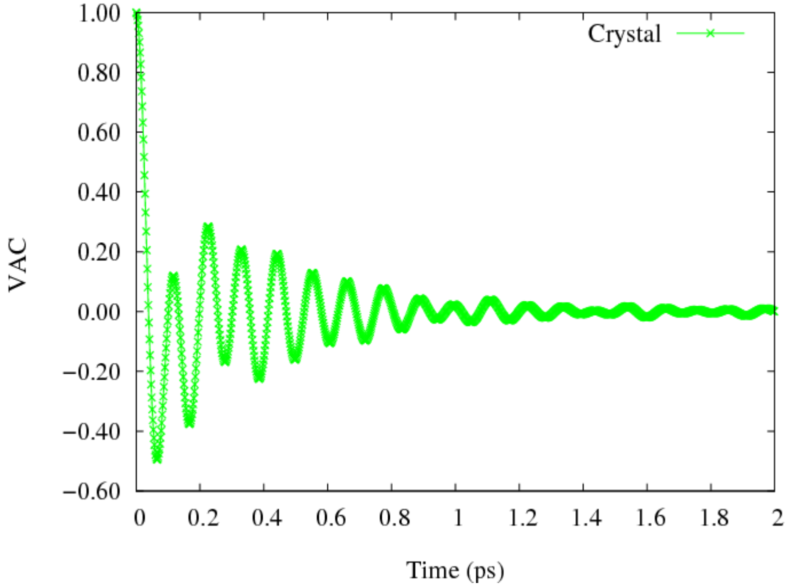
$$G(\omega)=\int_0^\infty Z(t)\cos(\omega t)\,dt \qquad Z(t)=\frac{<v(t)v(0)>}{<v(0)v(0)>}$$

• PDOS calculation were performed at 50 K.

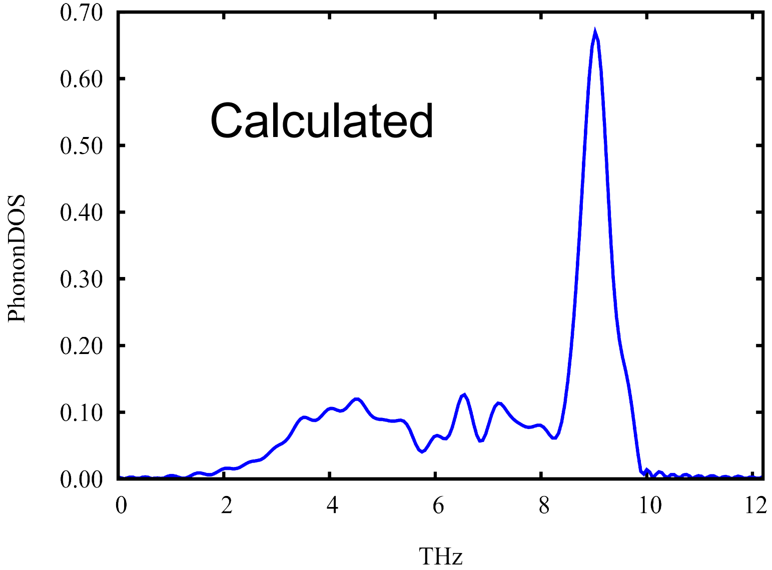
[1] Tersoff J., Phys. Rev. B, 39, 5566, (1989)
[2] Dickey J., Phys. Rev., 188, 1407, (1069)

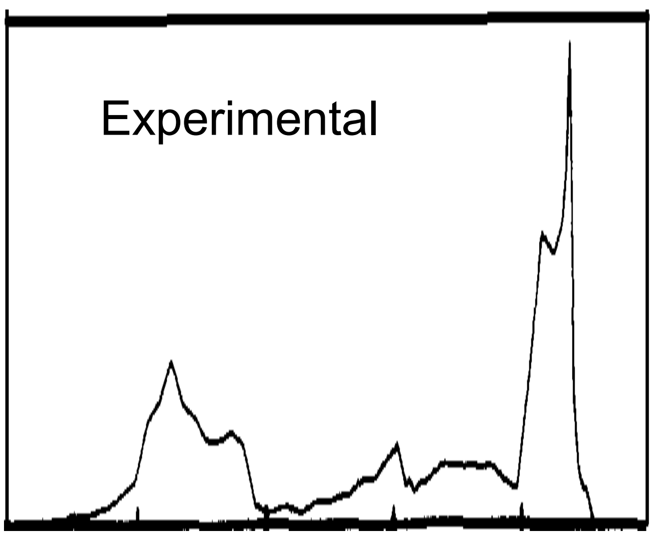
Methods

PDOS of Ge bulk crystal



Fourier transform

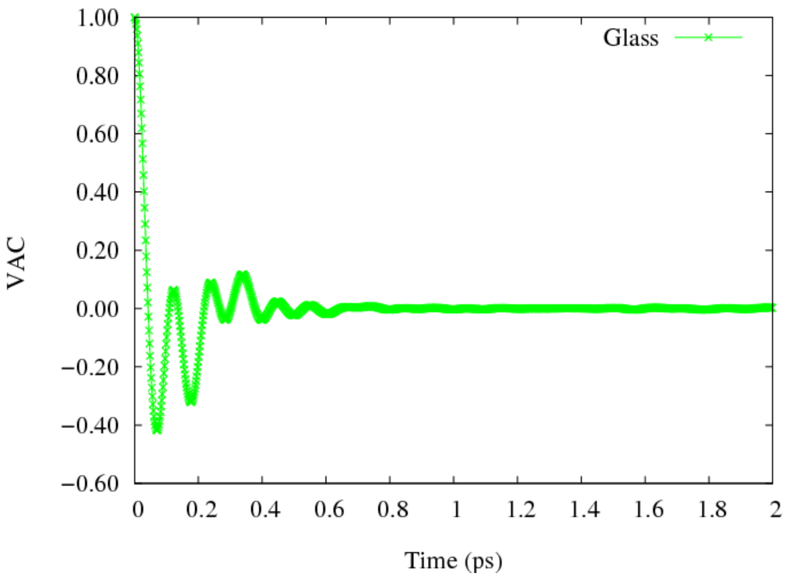




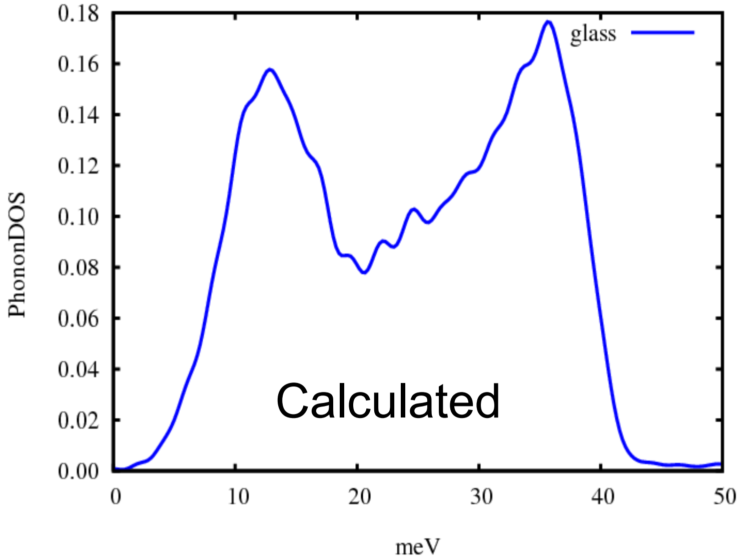
Siging W., Phys. Rev. B, 2221, 50, (1993)

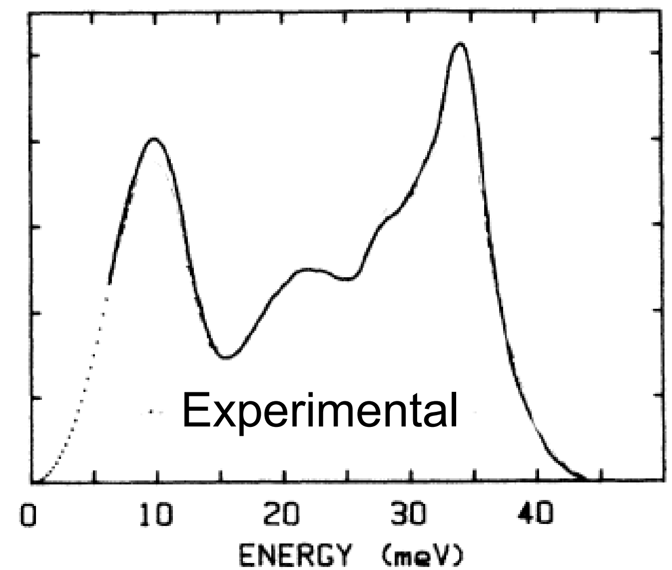
Methods

PDOS of Ge bulk glass



Fourier transform



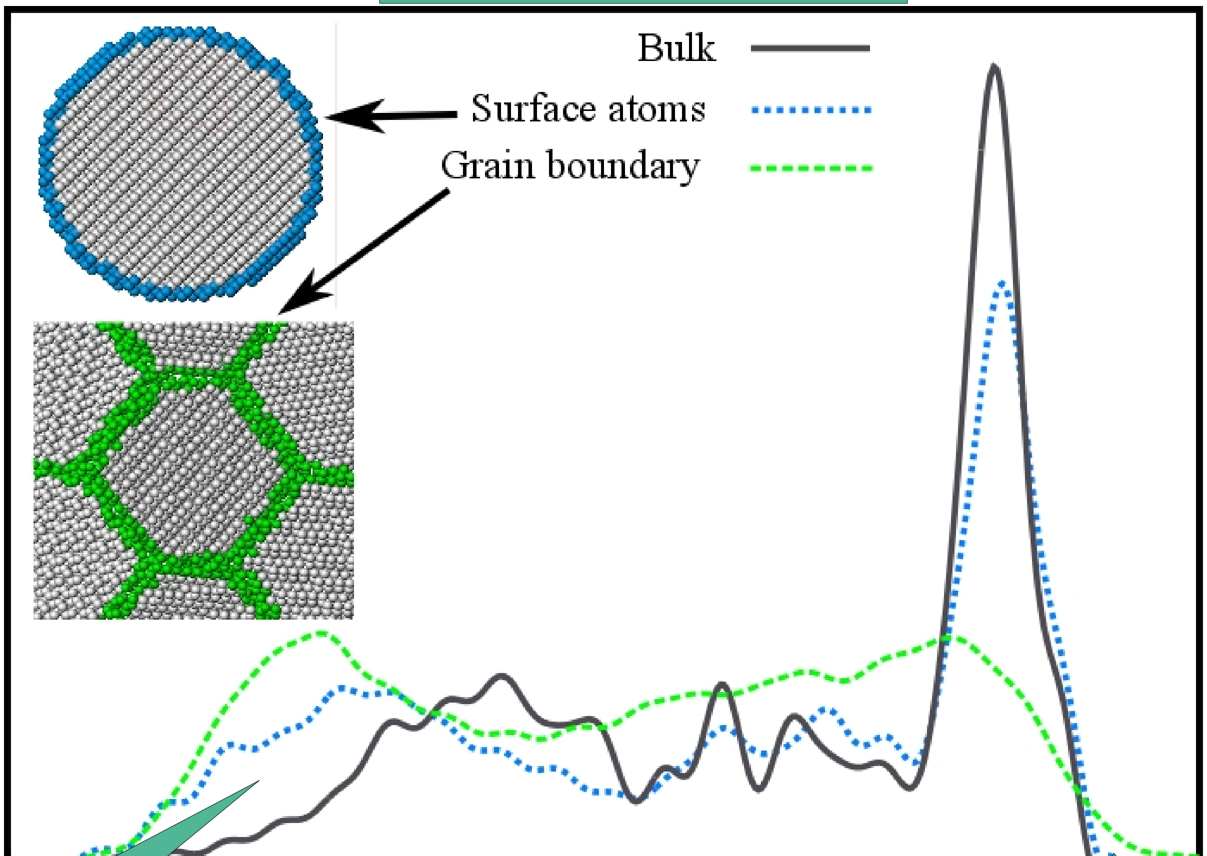


Maley N.,
Phys. Rev. Lett., 56,
1720, (1986)

Size effects which change the PDOS

1

Inhomogeneities



Low density or low coordinated atoms

High atomic disorder

Size effects which change the PDOS

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2

Surface stresses

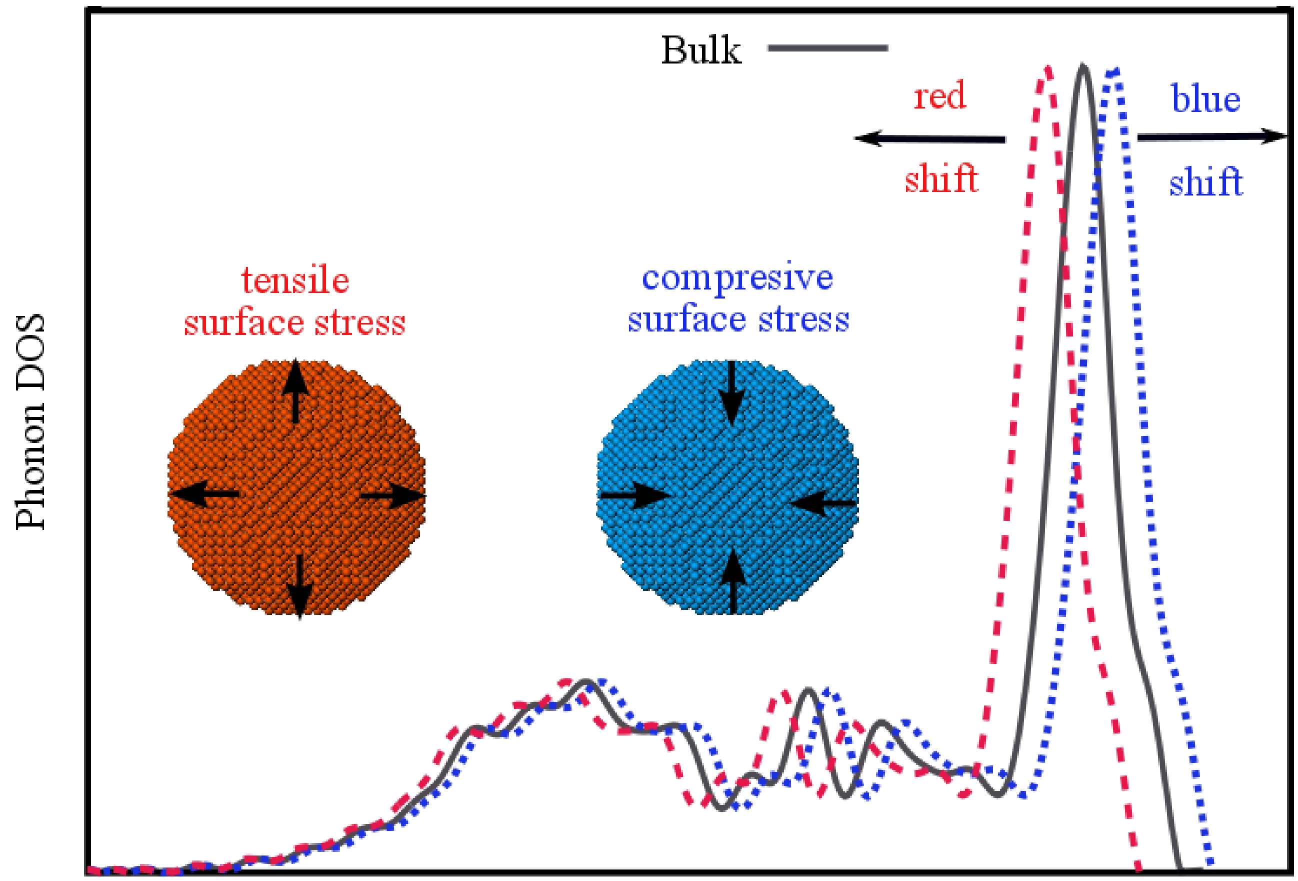
$f_{ij}=\delta_{ij}\sigma+(\partial\sigma/\partial e_{ij})$

$\partial\sigma/\partial e_{ij}<0$

compresive surface stress

$\partial\sigma/\partial e_{ij}>0$

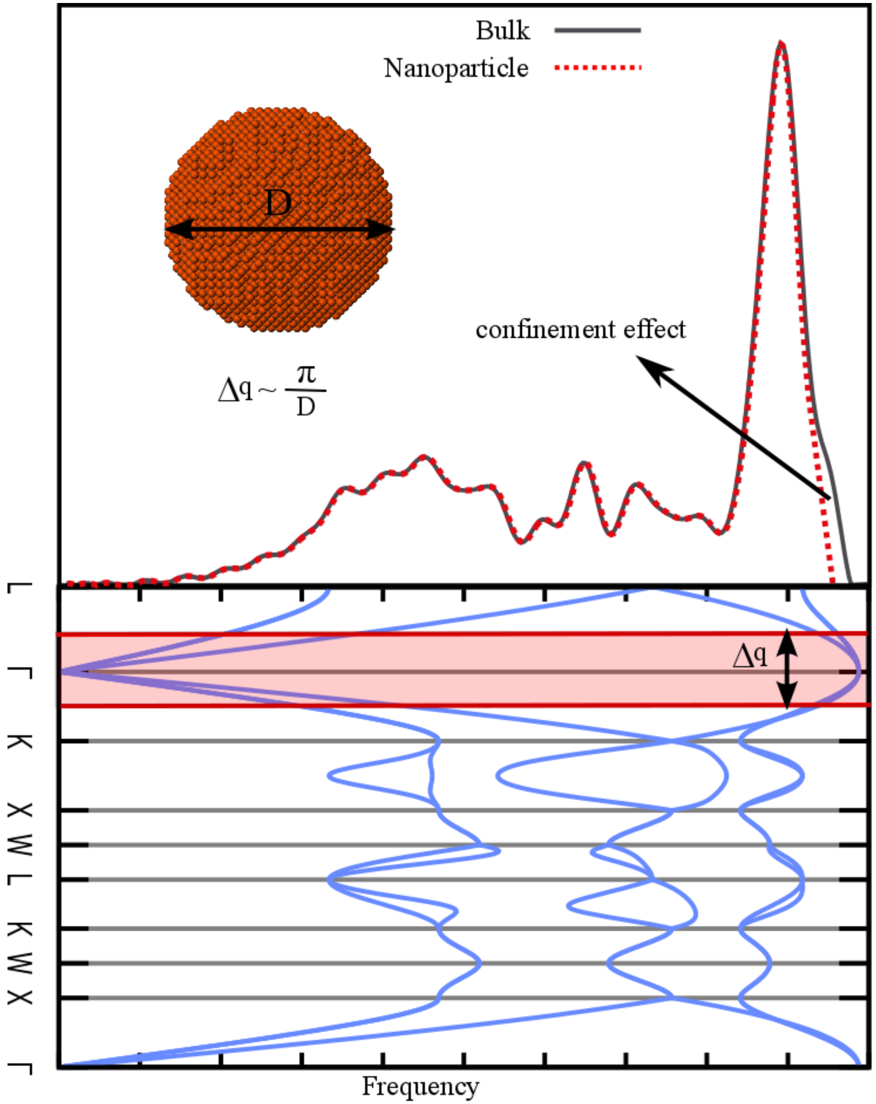
tensile surface stress



Size effects which change the PDOS

3

Confinement effect



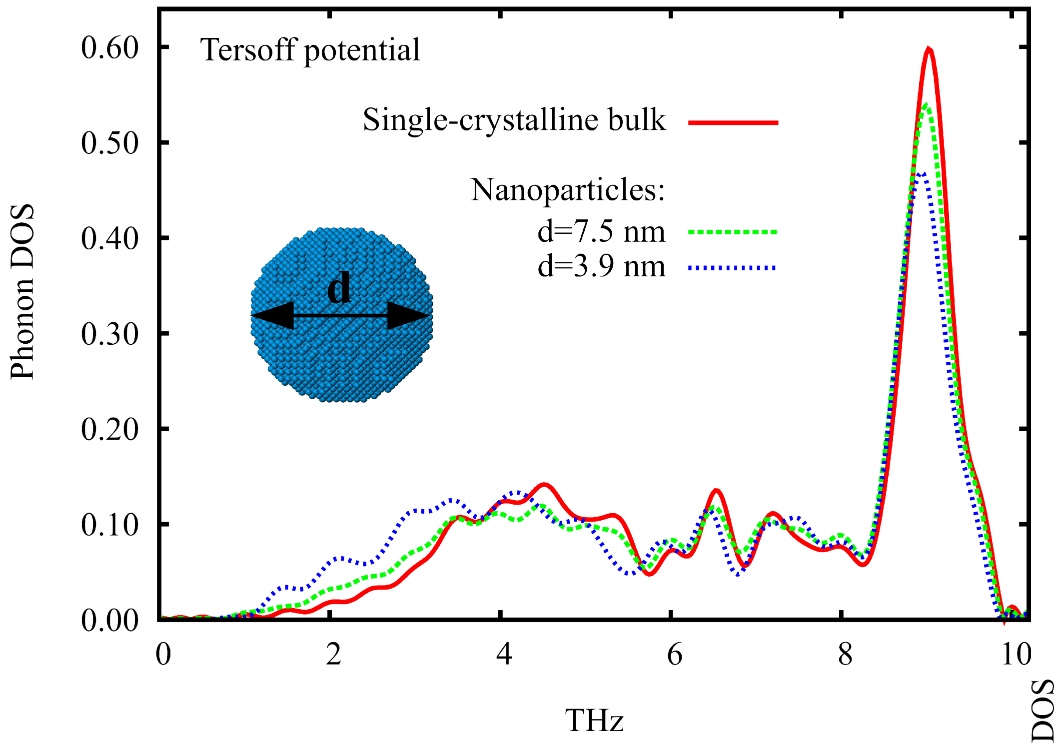
$|\vec{q}| \approx \pi/D$

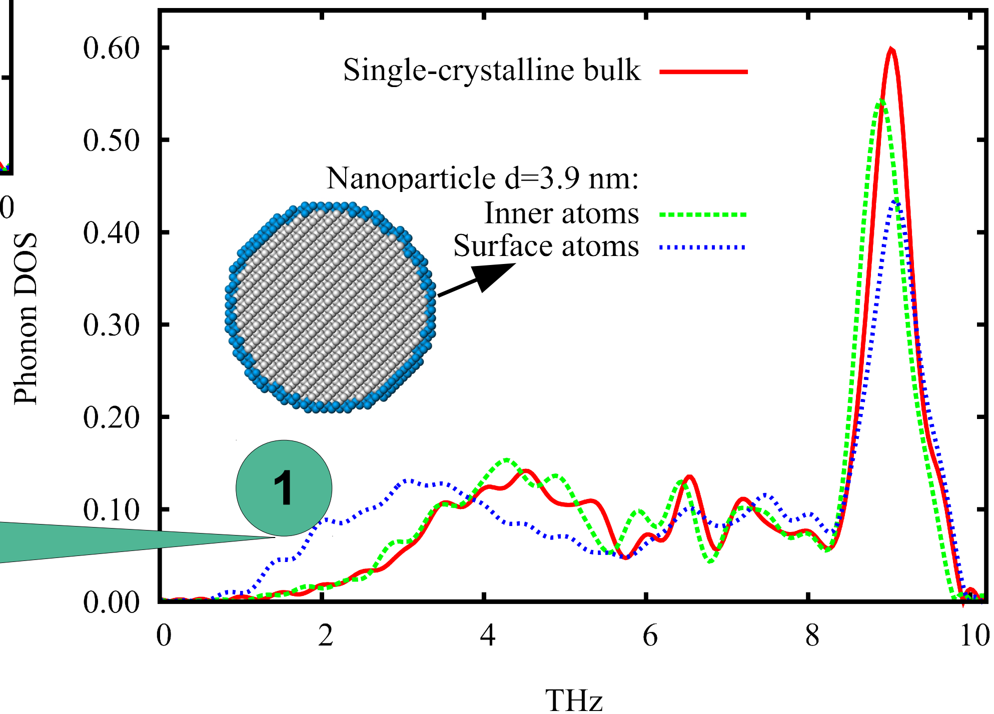
$D \approx nm$

$\rightarrow q \neq 0$

Sopu D et al., Phys. Rev. B, 83, 245416, (2011)

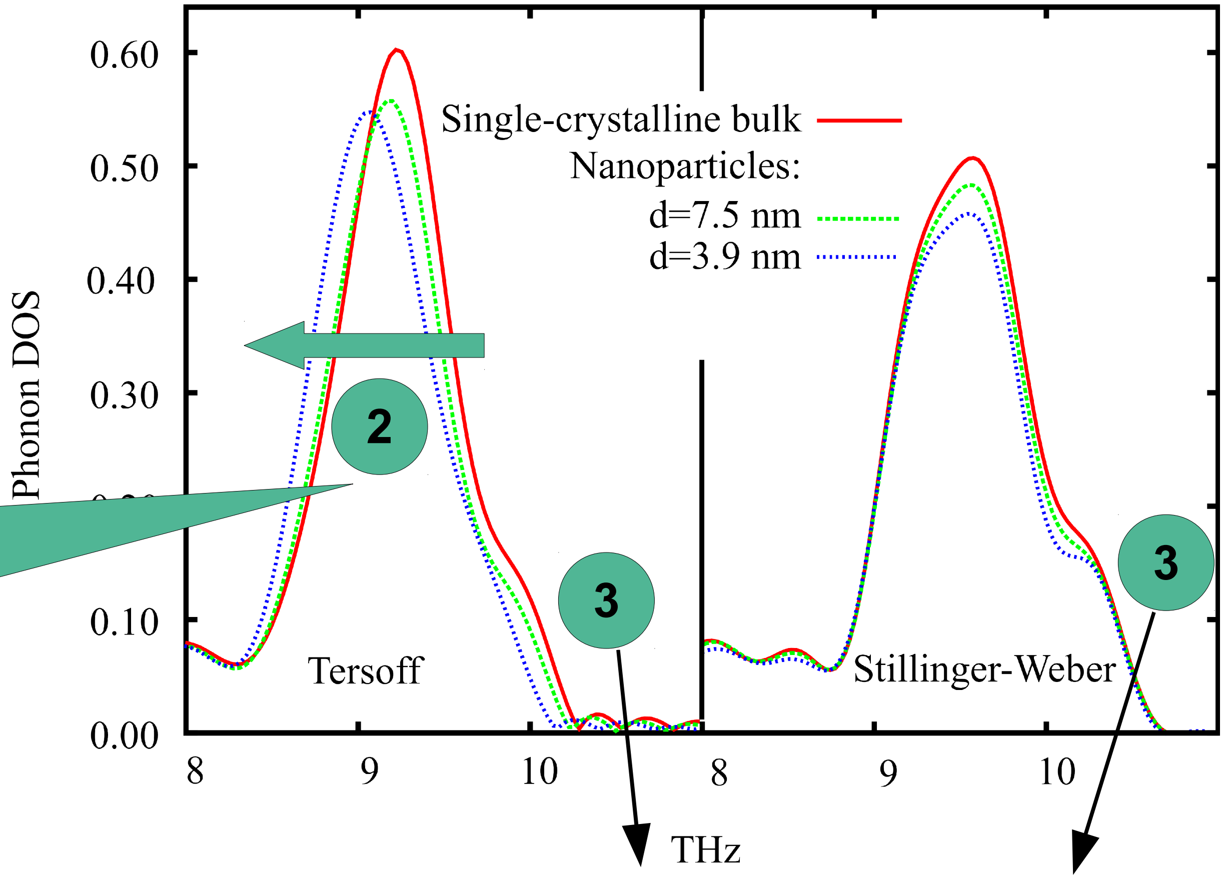
Free Nanoparticles





Attributed to the weakly bonded surface atoms, with lower coordination

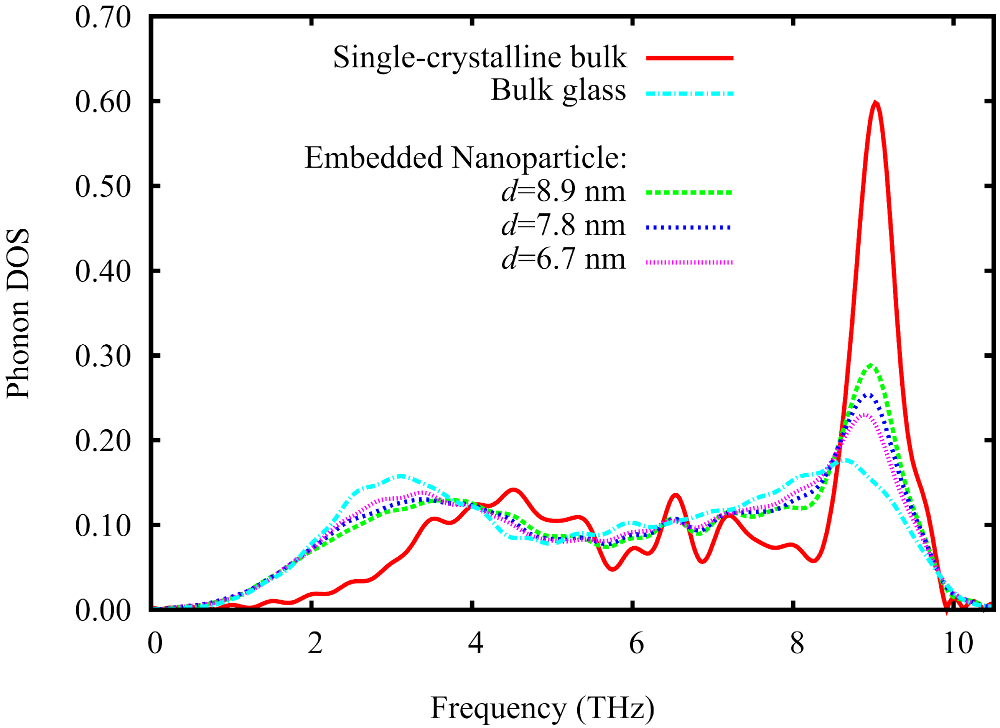
Free Nanoparticles

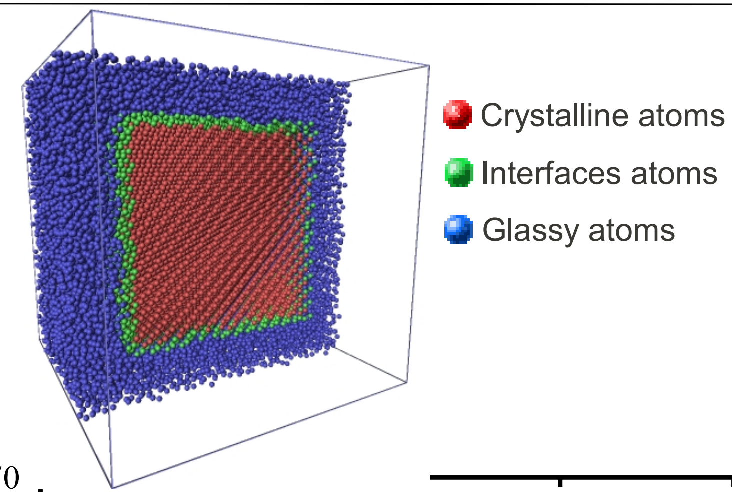


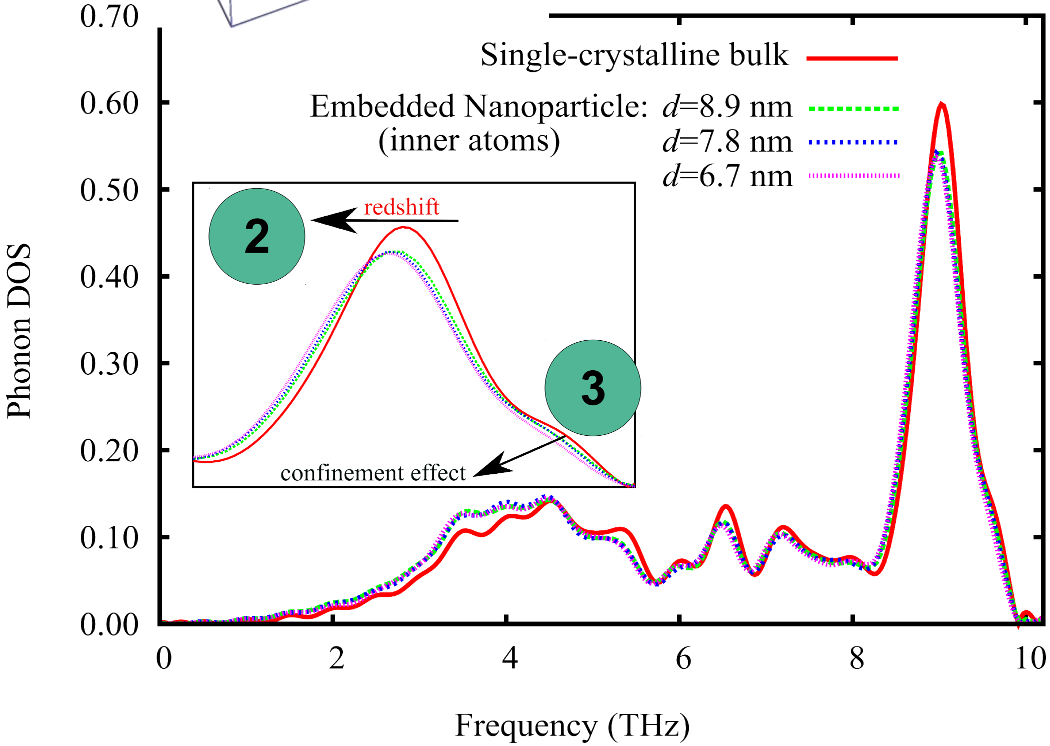
Red-shift can be attributed to strain effects due to surface stress

The shoulder at high frequency is shifted due to the confinement effect.

Embedded nanoparticles







Free vs. embedded nanoparticles

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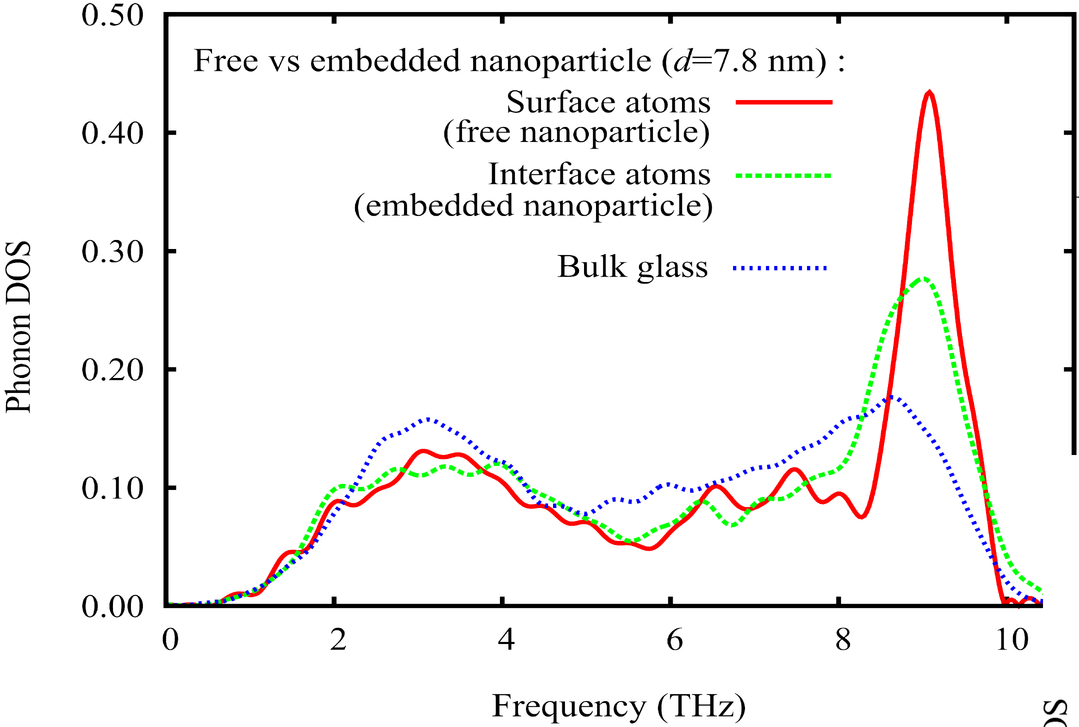
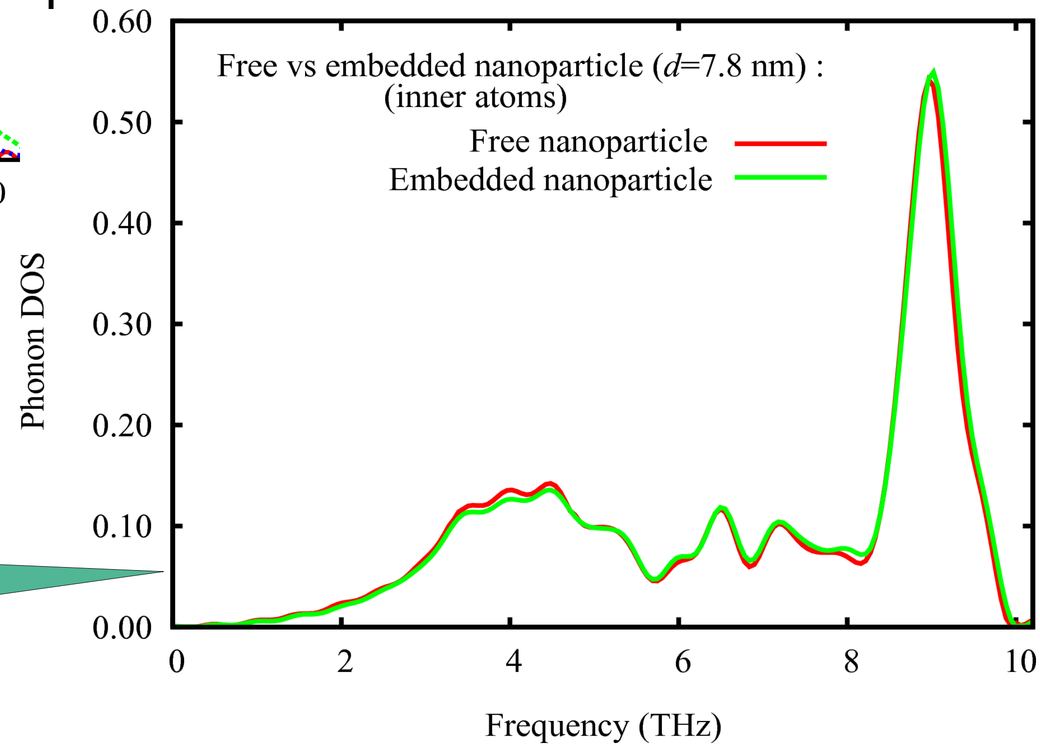


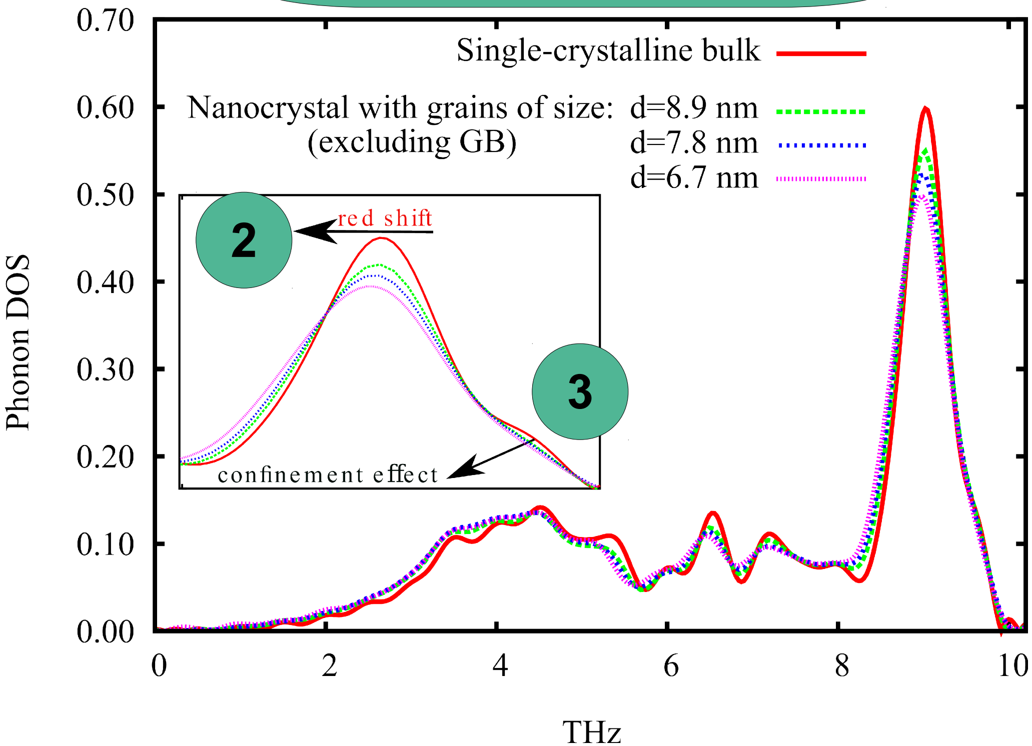
Glassy matrix affects the partial PDOS of the crystal-glass interface.

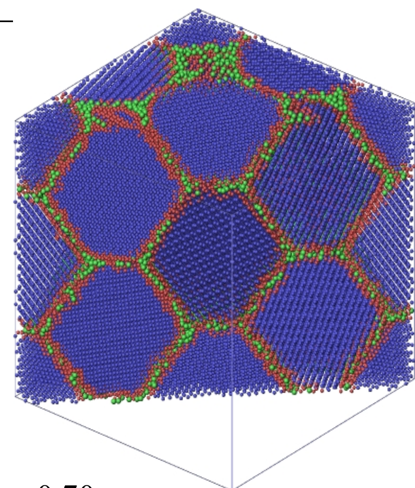


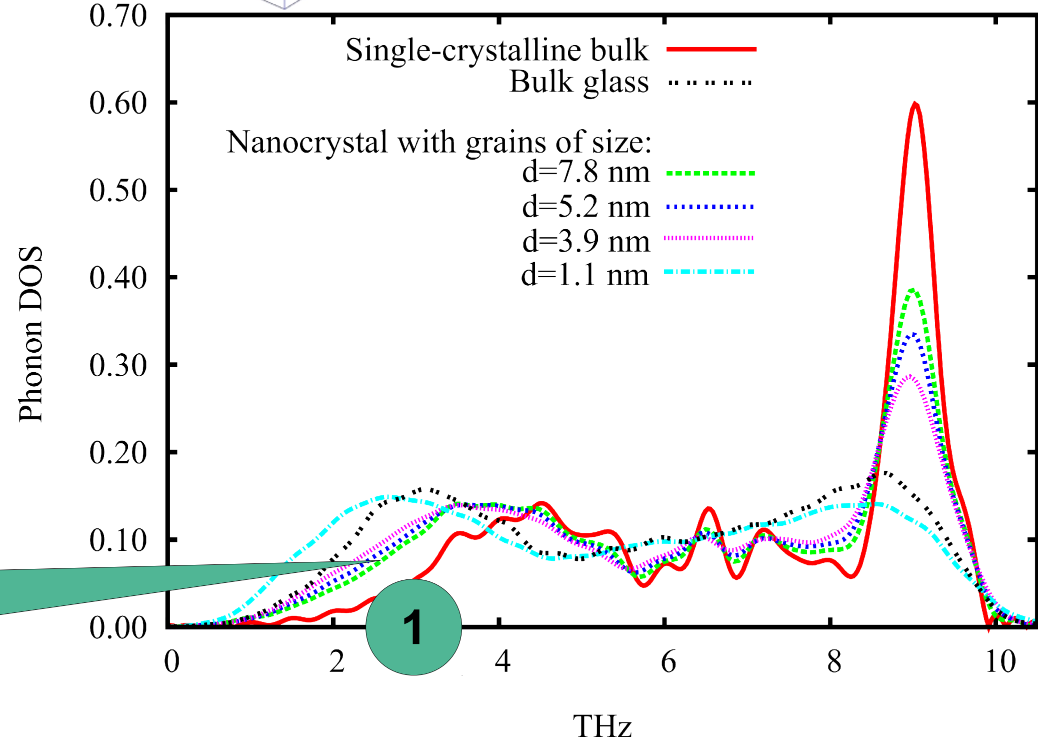
Vibrational modes of glassy matrix do not extend in the inner part of the embedded particle

Nanocrystalline Ge

Excluding GB



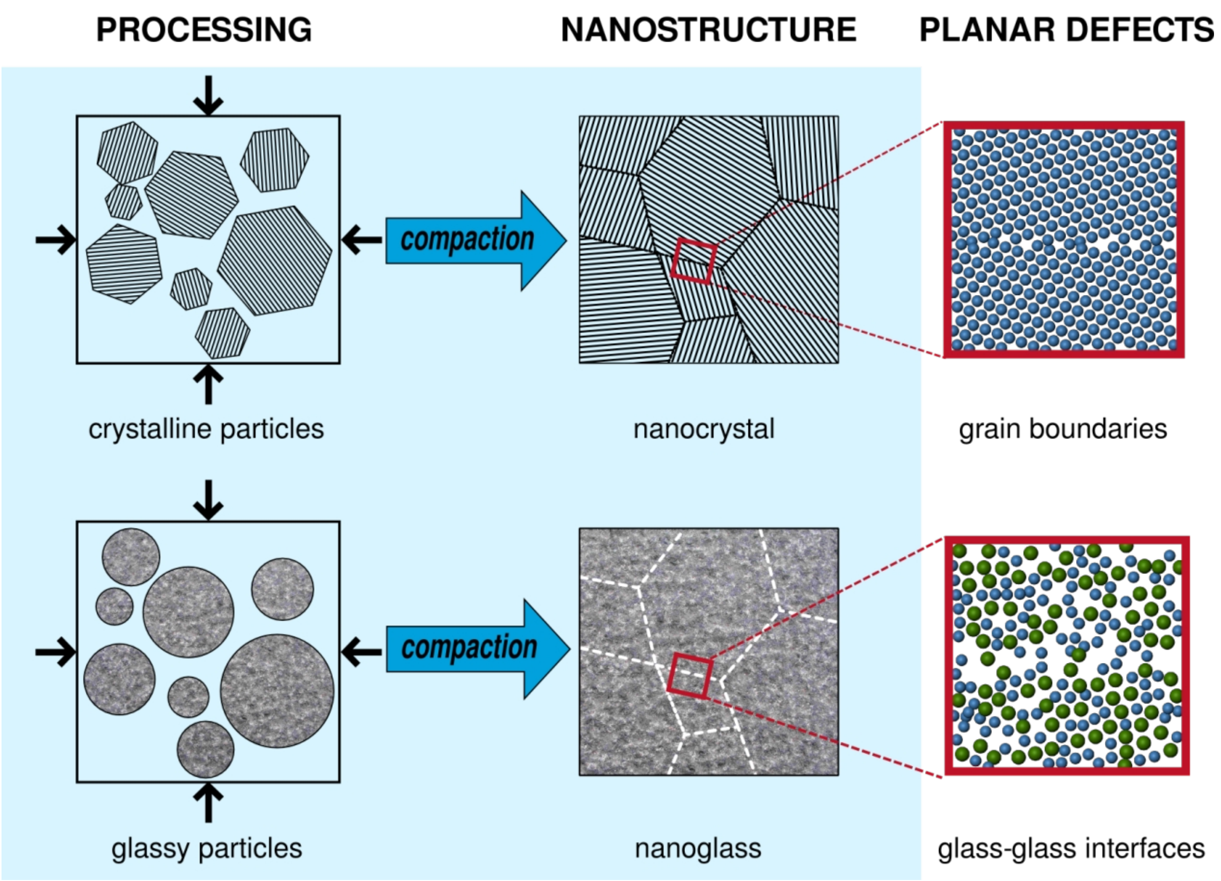




Low frequency modes increase with the increasing of GB fraction

Nanoglass-introduction

Nanoglasses



Motivation

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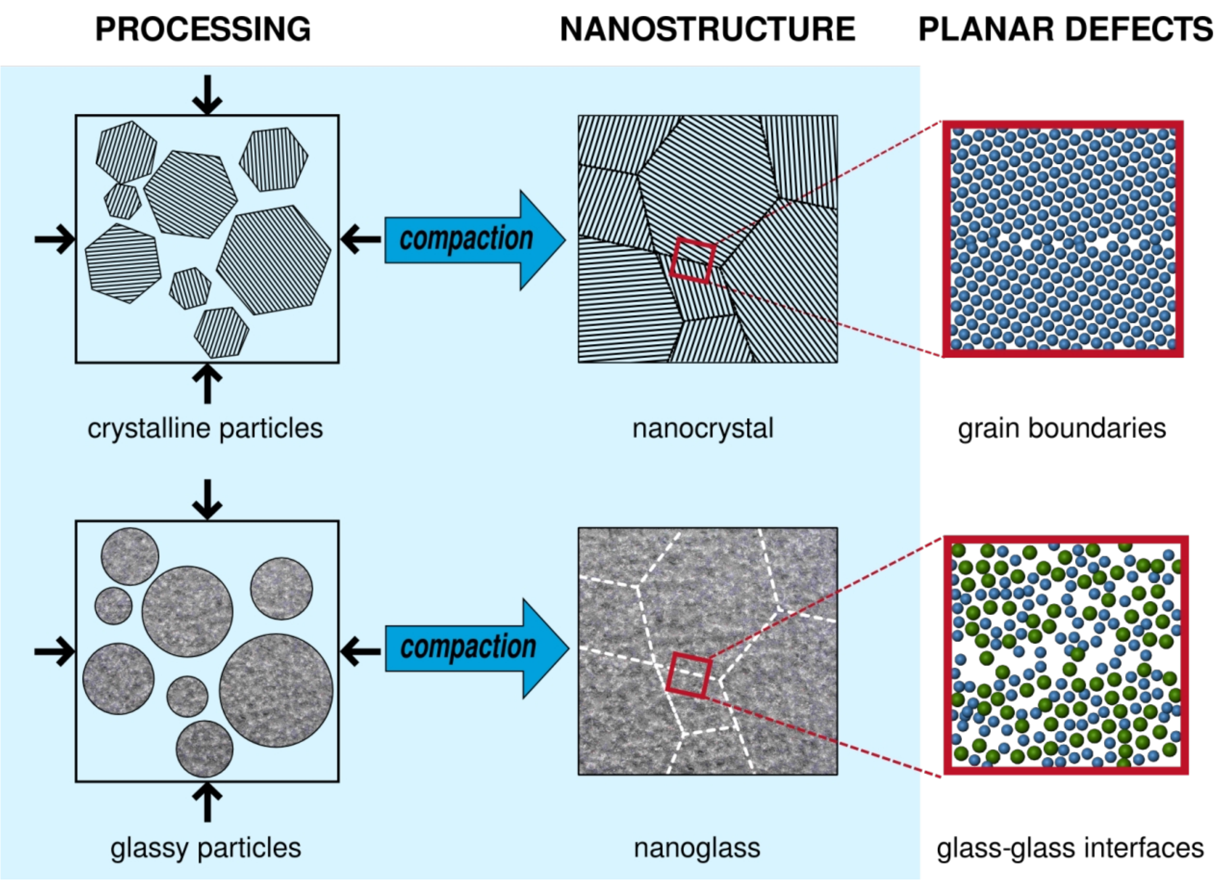
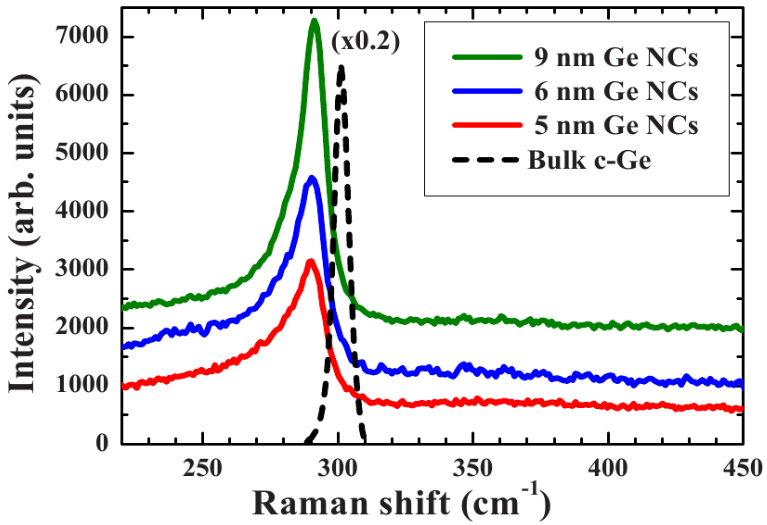
Nanoglasses



PROPERTIES



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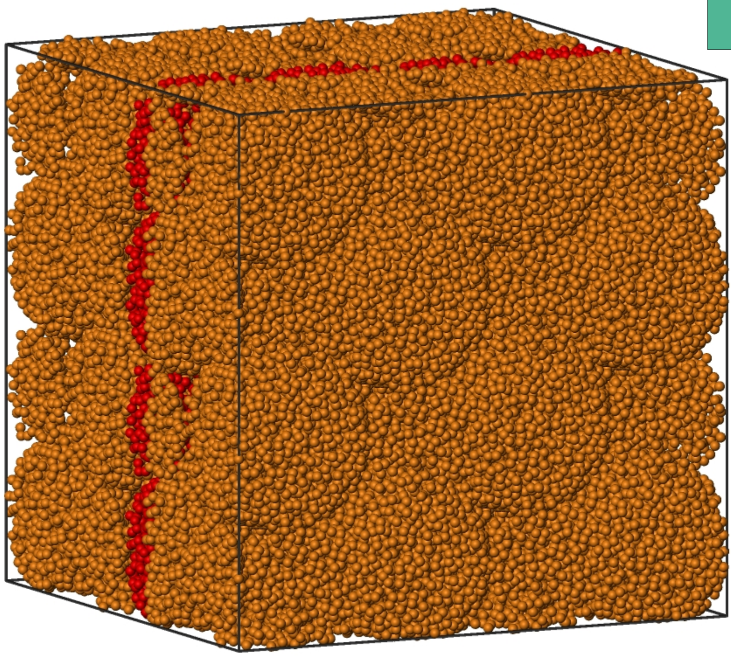
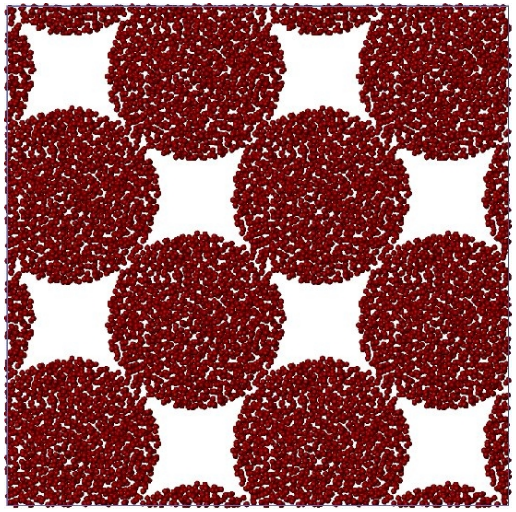
Manley *et al*., Phys. Rev. B, 78, 020101, (2008)

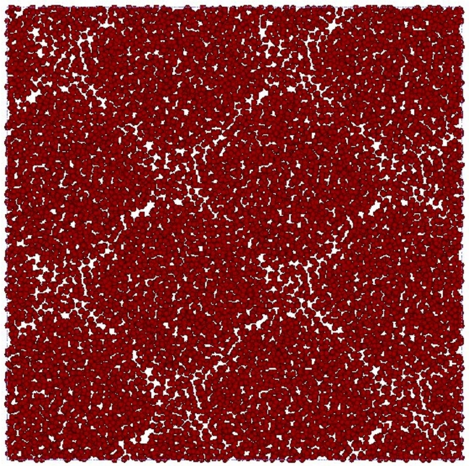
Nanoglass Ge



Thin slab



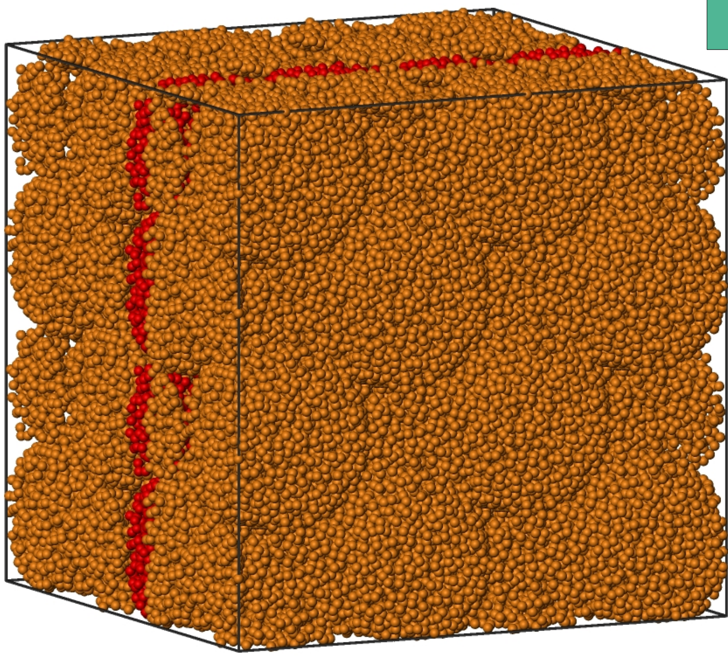
Compaction 5 GPa



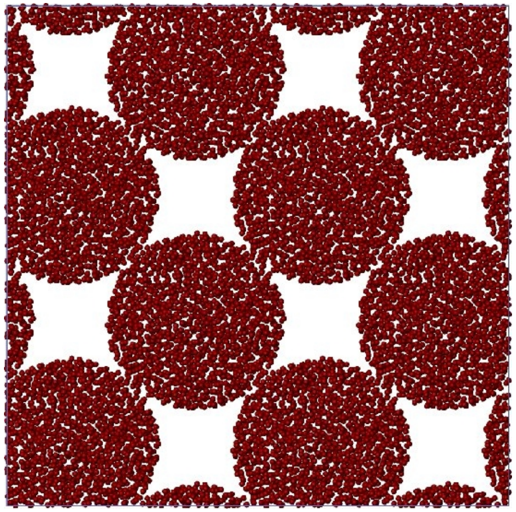
32 glassy sphere (5 nm)

Sopu D., Appl. Phys. Lett., 94, 191911, (2009)

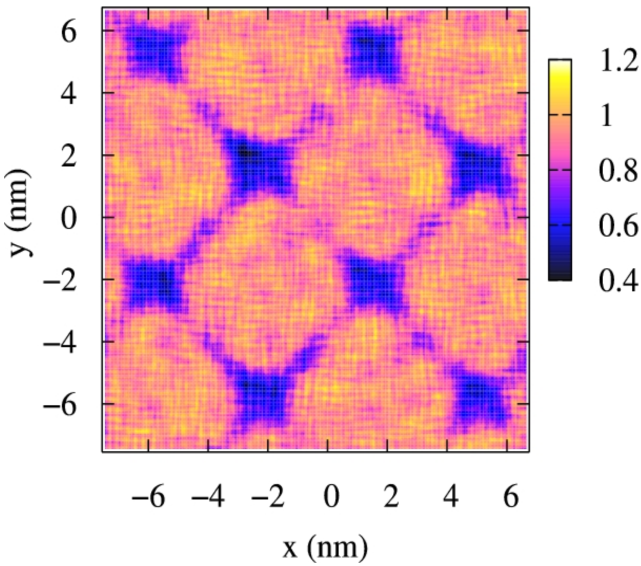
Nanoglass Ge



Thin slab



Compaction 5 GPa



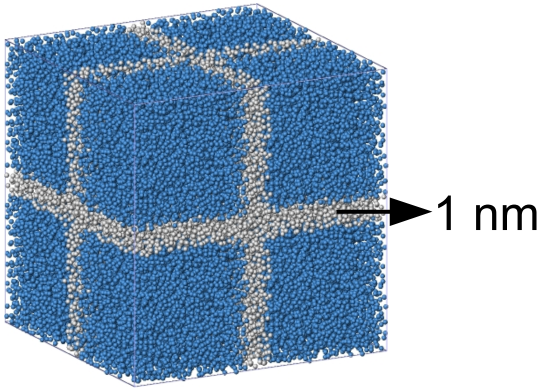
32 glassy sphere (5 nm)

Nanoglass characterized by interfaces with enhanced free volume

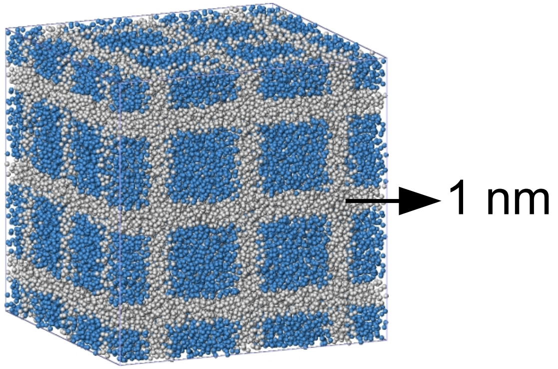
Sopu D., Appl. Phys. Lett., 94, 191911, (2009)

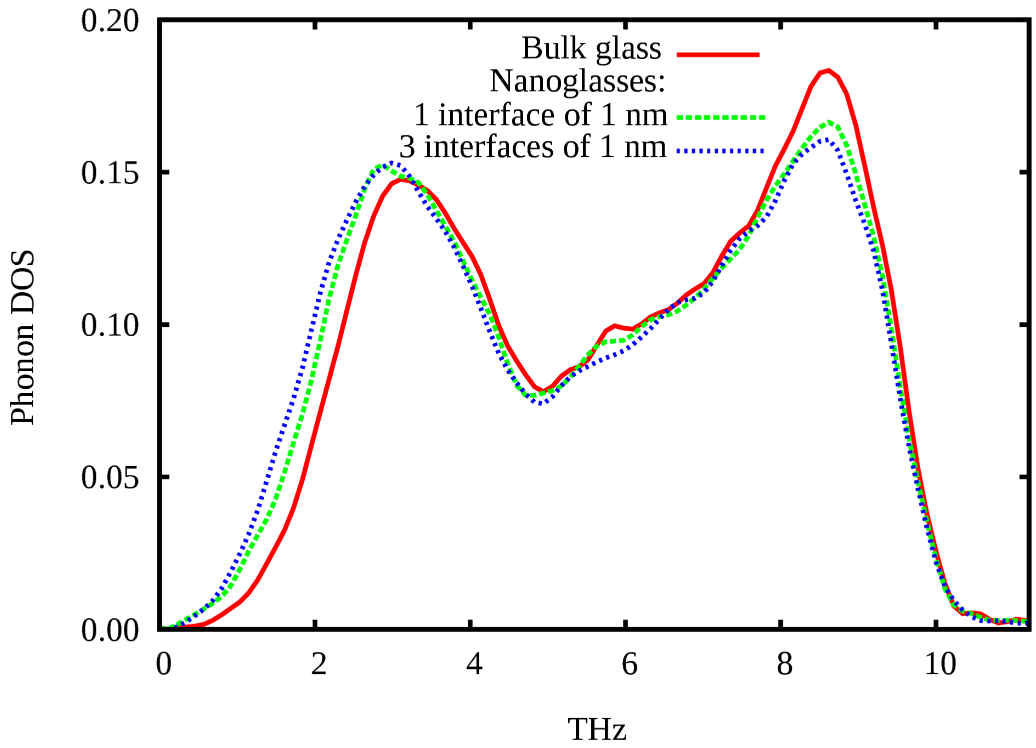
Nanoglass Ge

PDOS function of the number of interfaces



Interfaces with
8% excess free volume

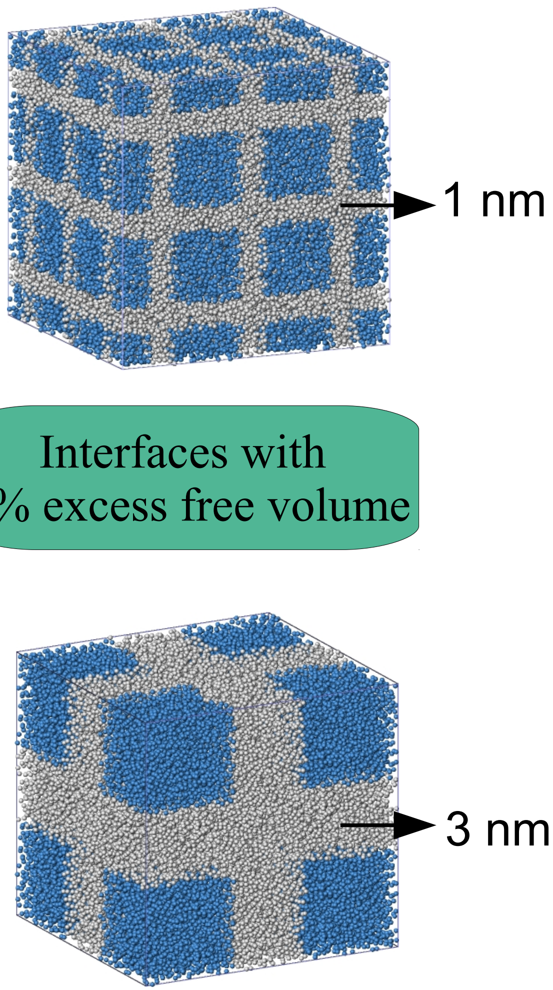




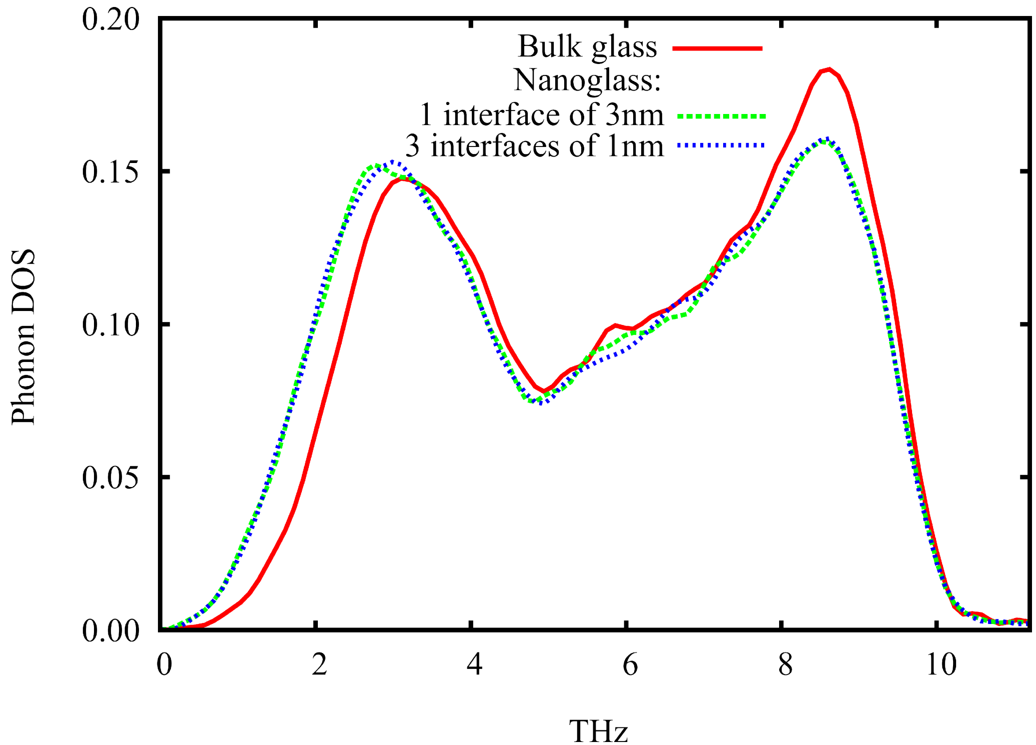
The acoustic peak shift to a lower frequency
With increasing number of interfaces

Nanoglass Ge

PDOS depending on the interface width

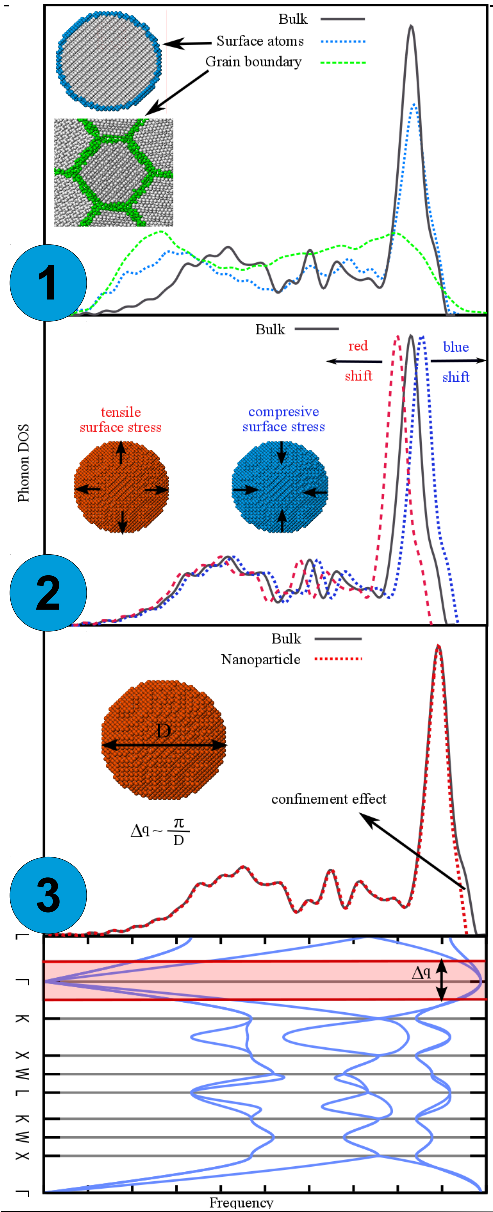


Interfaces with
8% excess free volume



Same free volume in both NGs leads to
no difference in the PDOS

Conclusions



In case of nanocrystalline materials the PDOS changes as follow:

(1) Nanostructural discontinuities (surface atoms, GBs, interfaces) cause an enhanced population modes with low frequencies.

(2) Tensile (compresive) surface stress result in a shift of the entire PDOS to lower (higher) frequencies

(3) Confinement due to the particle size cause the disappearance of several optical modes at Brillouin zone (q=0)

In case of nanoglasses the PDOS changes only due to the excess free volume in the interfaces.

Sopu D et al., Phys. Rev. B, 83, 245416, (2011)

Thank you for your attention!

Funded by DFG AL-578-5