Tailoring the optical properties of silica irradiated with swift heavy ions

A. Rivera,[1] A. Prada,[1] O. Peña-Rodríguez,[1]
J. Manzano-Santamaría,[2,3] M. Crespillo,[3]
J. Olivares[3,4] and F. Agulló-López[3]

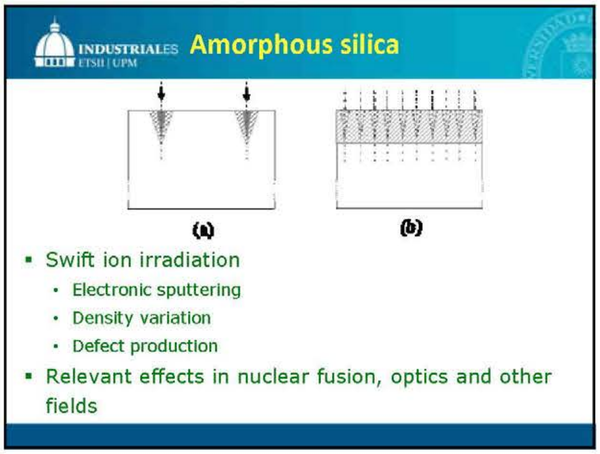
[1]Instituto de Fusión Nuclear, Universidad Politécnica de Madrid
[2]Euratom/CIEMAT Fusion Association
[3]Centro de Micro-Análisis de Materiales, Universidad Autónoma de Madrid
[4]Instituto de Óptica, Consejo Superior de Investigaciones Científicas

Outline

- Introduction
- Molecular dynamics
    - Electronic excitation
- Linking simulations with experiments
    - Optical measurements
- Conclusions

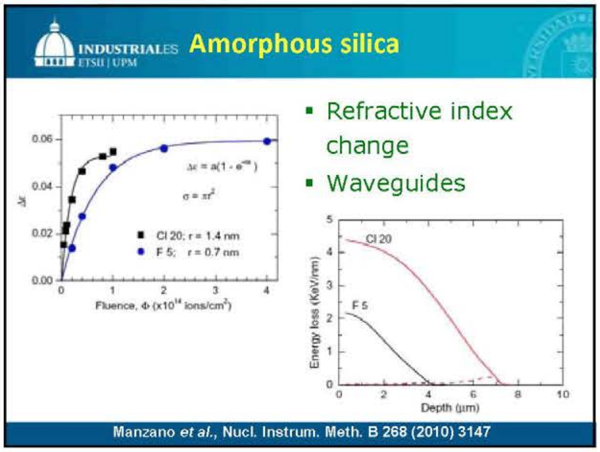
Outline

- **Introduction**
- Molecular dynamics
    - Electronic excitation
- Linking simulations with experiments
    - Optical measurements
- Conclusions



Amorphous silica

- Swift ion irradiation
    - Electronic sputtering
    - Density variation
    - Defect production
- Relevant effects in nuclear fusion, optics and other fields



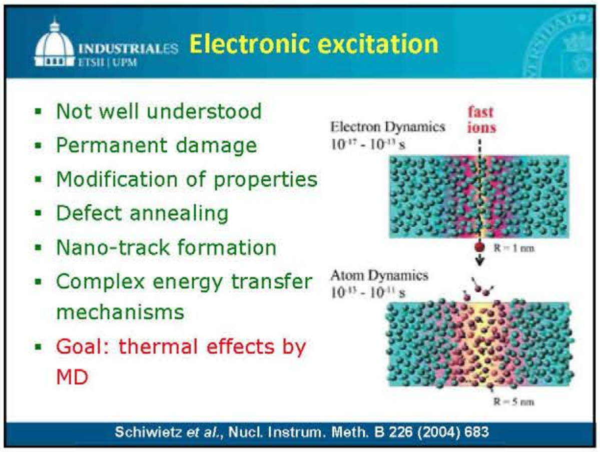
Amorphous silica

- Refractive index change
- Waveguides

Manzano *et al.*, Nucl. Instrum. Meth. B 268 (2010) 3147

Outline

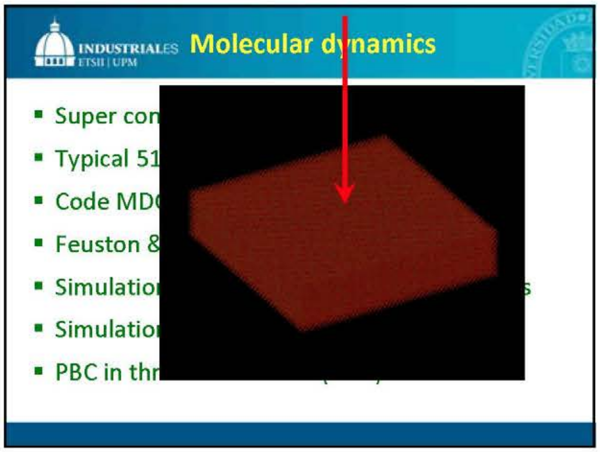
- Introduction
- **Molecular dynamics**
    - Electronic excitation
- Linking simulations with experiments
    - Optical measurements
- Conclusions



Electronic excitation

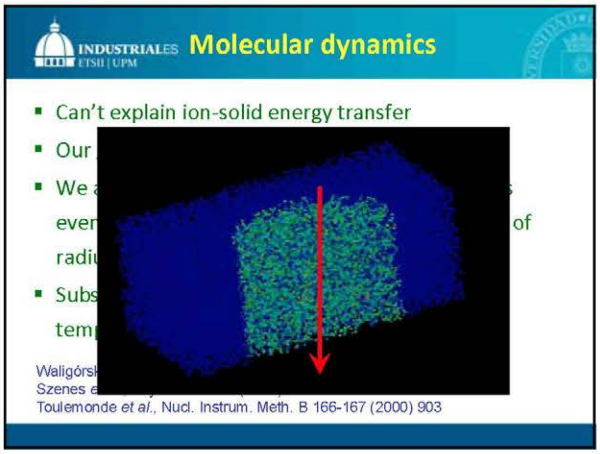
- Not well understood
- Permanent damage
- Modification of properties
- Defect annealing
- Nano-track formation
- Complex energy transfer mechanisms
- Goal: thermal effects by MD

Schiwietz *et al.*, Nucl. Instrum. Meth. B 226 (2004) 683



Molecular dynamics

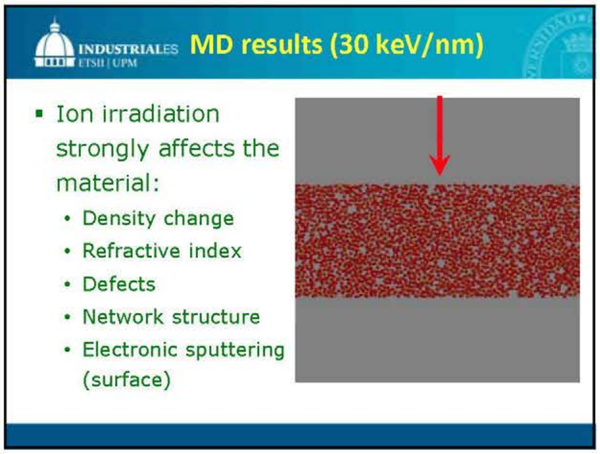
- Super con
- Typical 51
- Code MD
- Feuston &
- Simulation s
- Simulation
- PBC in thr)



Molecular dynamics

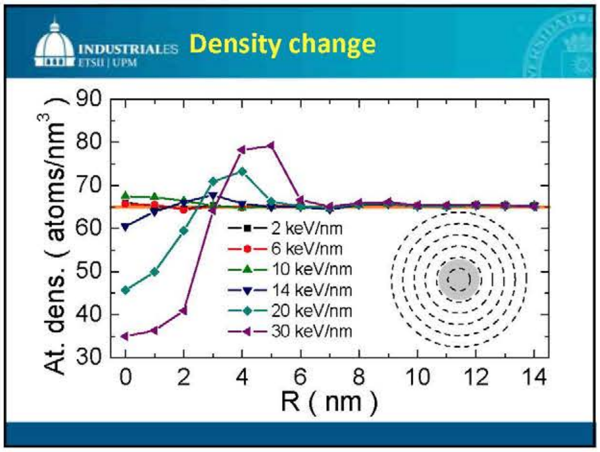
- Can’t explain ion-solid energy transfer
- Our
- We a s
  even of
  radiu
- Subs
  temp

Waligórsk
Szenes e
Toulemonde *et al.*, Nucl. Instrum. Meth. B 166-167 (2000) 903

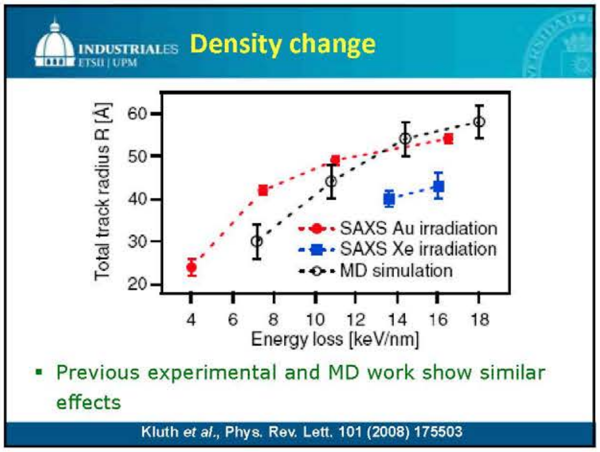


MD results (30 keV/nm)

- Ion irradiation strongly affects the material:
  - Density change
  - Refractive index
  - Defects
  - Network structure
  - Electronic sputtering (surface)



Density change

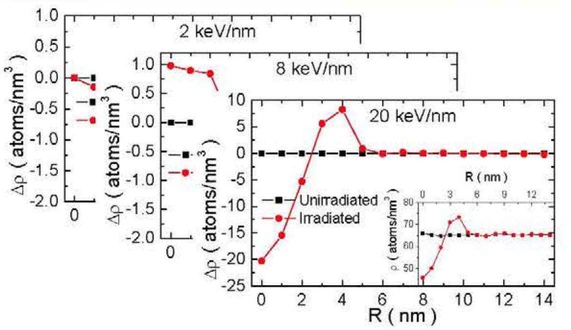


Density change

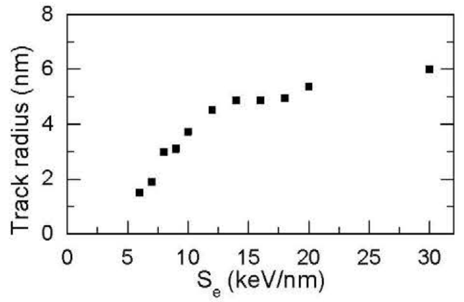
- Previous experimental and MD work show similar effects

Kluth *et al.*, Phys. Rev. Lett. 101 (2008) 175503

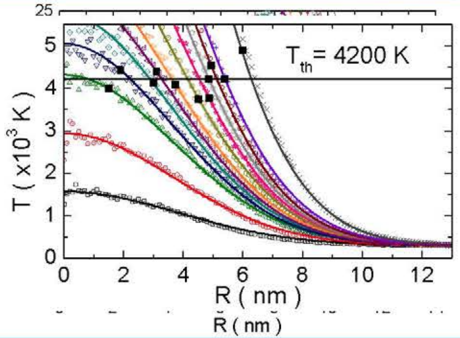
Track radius (direct determination)



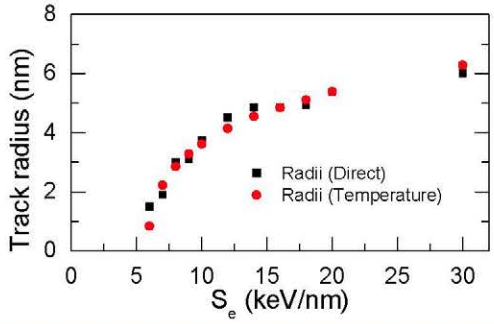
"Measured" radii



Track radius (from Temp. profiles)



"Measured" radii



In summary

- Tree clearly-defined regions
  - Below threshold (~4-6 keV/nm) there are no tracks
  - For low S_e (< 12 keV/nm) we observed compact cylindrical tracks
  - For high S_e (> 12 keV/nm) we obtained a core-shell track. The core region has a lower density whereas for the shell the opposite is true

Outline

- Introduction
- Molecular dynamics
  - Electronic excitation
- **Linking simulations with experiments**
  - Optical measurements
- Conclusions

Linking simulations with experiments

- Comparing the simulations with experimental results is very desirable but it is not an easy task
- Microscopic measurements
  - Directly comparable
  - Hard to perform
- Macroscopic measurements
  - Easier to obtain
  - Comparison with simulations not always obvious

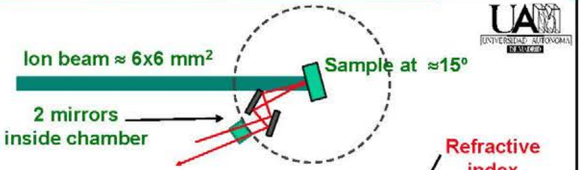
Optical measurements

- “Macroscopic” measurement
- Very fast (we could follow the kinetics at one-second intervals)
- Easy to do
- Easy to analyze
- Direct conversion from track density/stoichiometry to refractive index are possible
- Effective medium approximations (EMA) provide an straightforward mean to represent the refractive index kinetics

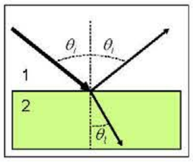
In situ reflectance

CMAM

UAM UNIVERSIDAD AUTONOMA DE MADRID



Refractive index

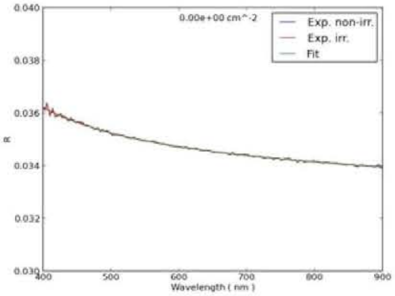


$n_1 \sin(\theta_i) = n_2 \sin(\theta_t)$ Snell's law

$r = r_s = r_p = \dfrac{n_1 - n_2}{n_1 + n_2}$ Fresnel coefficients

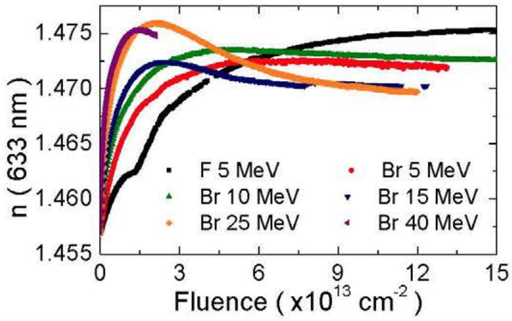
$R = |r|^2,\ T = 1 - R$ Reflectance

In situ reflectance

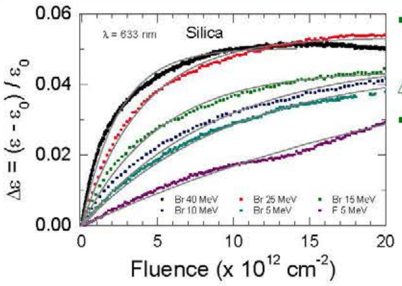


Br 40 MeV, S_e = 7.2 keV/nm

In situ reflectance



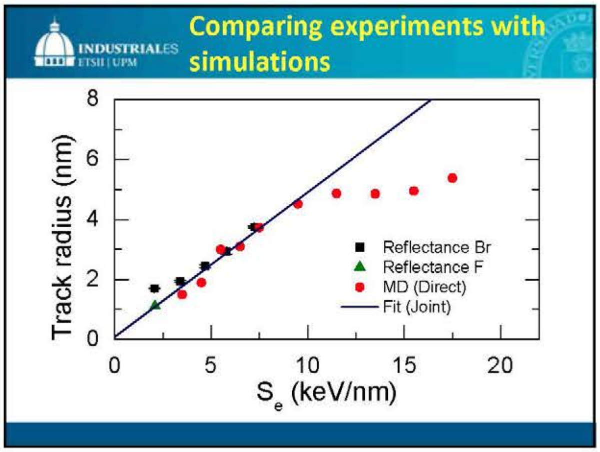
Track radius



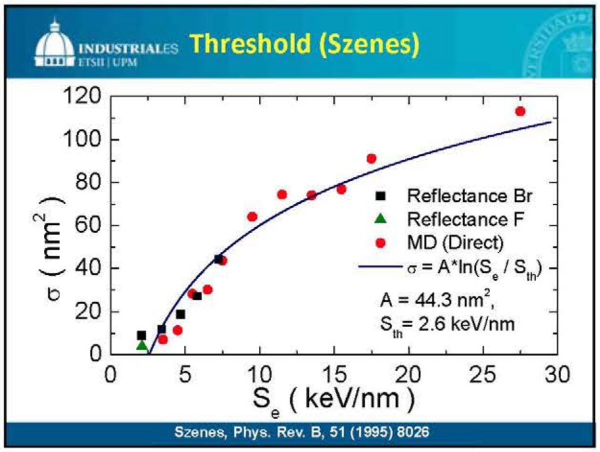
- Good Poisson fittings:

$\Delta\varepsilon = A[1 - e^{-\sigma\phi}]$

- We obtain:
  - Cross sections (σ)
  - Track radius ($R = \pi\sigma^2$)

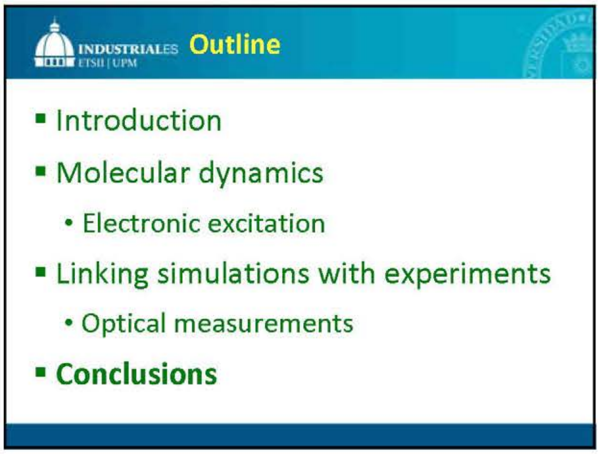


Comparing experiments with simulations



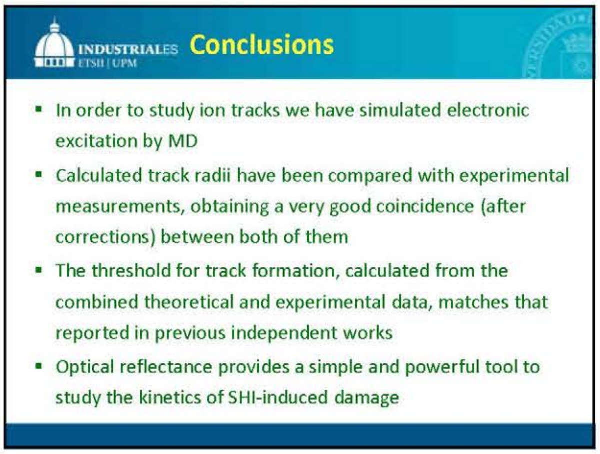
Threshold (Szenes)

Szenes, Phys. Rev. B, 51 (1995) 8026



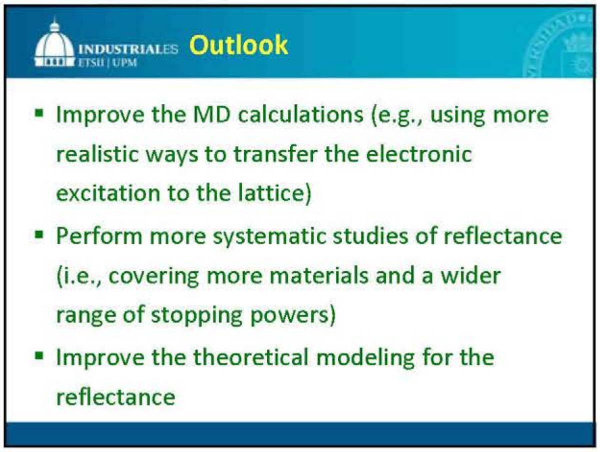
Outline

- Introduction
- Molecular dynamics
  - Electronic excitation
- Linking simulations with experiments
  - Optical measurements
- **Conclusions**



Conclusions

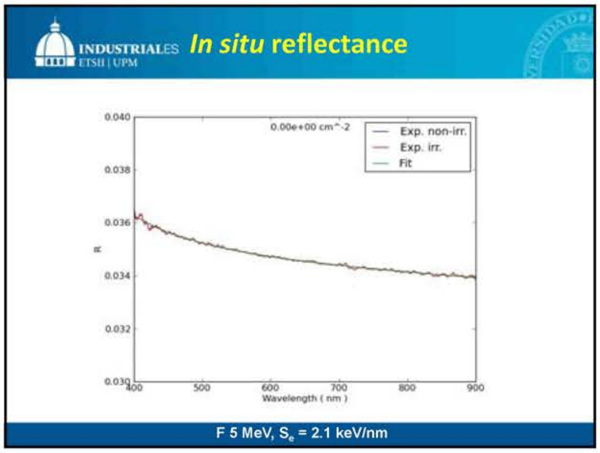
- In order to study ion tracks we have simulated electronic excitation by MD
- Calculated track radii have been compared with experimental measurements, obtaining a very good coincidence (after corrections) between both of them
- The threshold for track formation, calculated from the combined theoretical and experimental data, matches that reported in previous independent works
- Optical reflectance provides a simple and powerful tool to study the kinetics of SHI-induced damage



Outlook

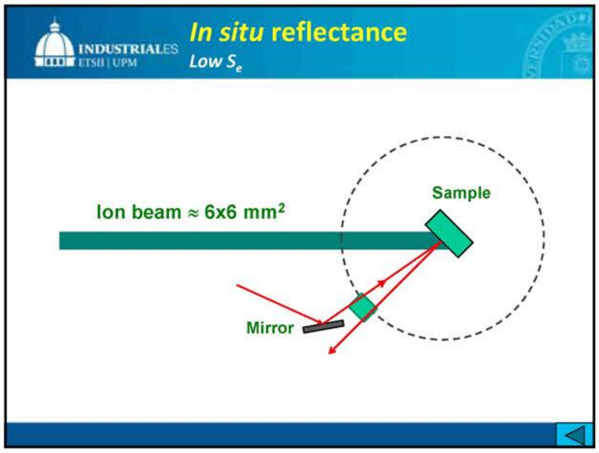
- Improve the MD calculations (e.g., using more realistic ways to transfer the electronic excitation to the lattice)
- Perform more systematic studies of reflectance (i.e., covering more materials and a wider range of stopping powers)
- Improve the theoretical modeling for the reflectance

Thank you
for your attention



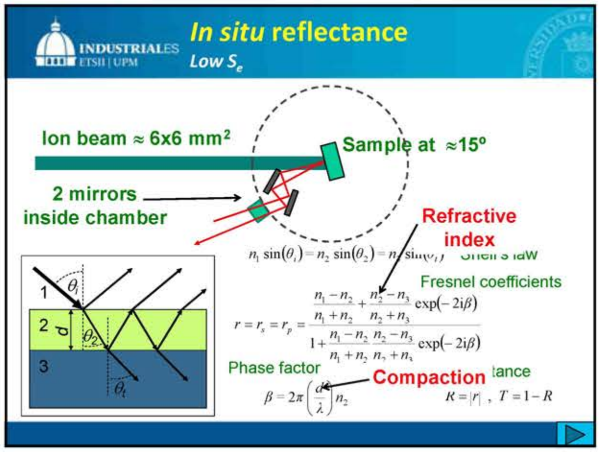
In situ reflectance

F 5 MeV, S_e = 2.1 keV/nm



In situ reflectance

Low S_e



In situ reflectance

Low S_e

Refractive index

$n_1 \sin(\theta_i) = n_2 \sin(\theta_2) = n_3 \sin(\theta_t)$ Snell's law

Fresnel coefficients

$$r = r_s = r_p = \frac{\dfrac{n_1 - n_2}{n_1 + n_2} + \dfrac{n_2 - n_3}{n_2 + n_3}\exp(-2i\beta)}{1 + \dfrac{n_1 - n_2}{n_1 + n_2}\dfrac{n_2 - n_3}{n_2 + n_3}\exp(-2i\beta)}$$

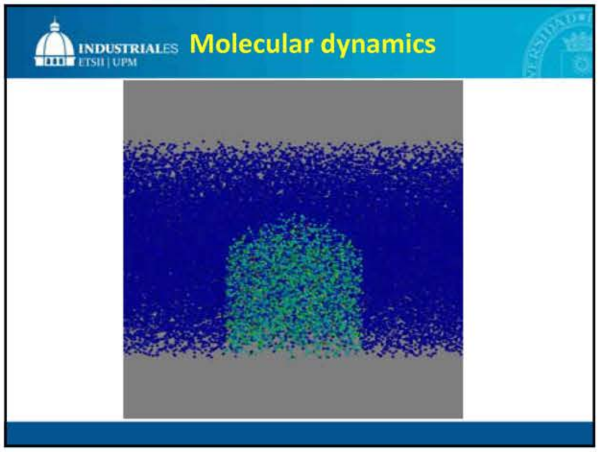
Phase factor

$$\beta = 2\pi\left(\frac{d}{\lambda}\right)n_2$$

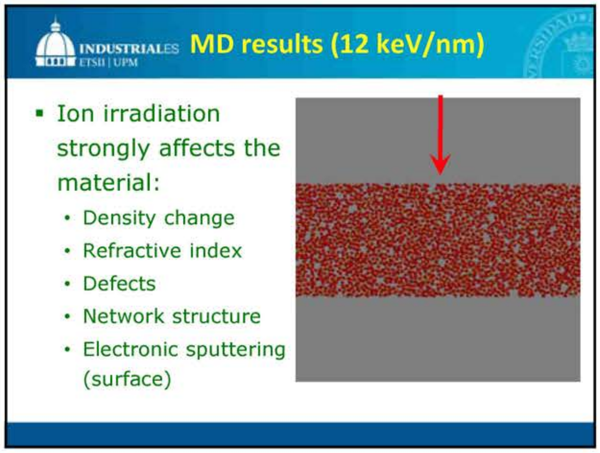
Compaction

tance

$$R = |r|, \quad T = 1 - R$$

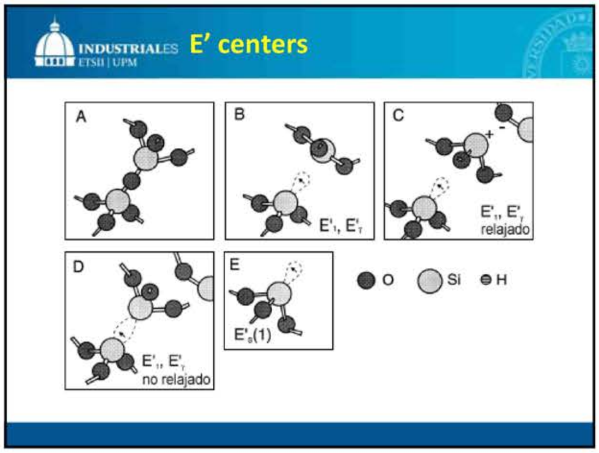


Molecular dynamics



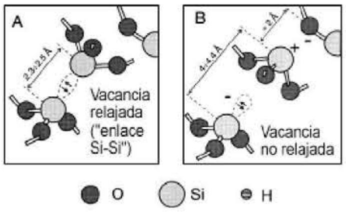
MD results (12 keV/nm)

- Ion irradiation strongly affects the material:
  - Density change
  - Refractive index
  - Defects
  - Network structure
  - Electronic sputtering (surface)



E' centers

ODC



NBOHC

$$\equiv \mathrm{Si{-}O{-}Si} \equiv \;\rightarrow\; \equiv \mathrm{Si{-}O}\bullet \;\; \bullet \mathrm{Si} \equiv$$