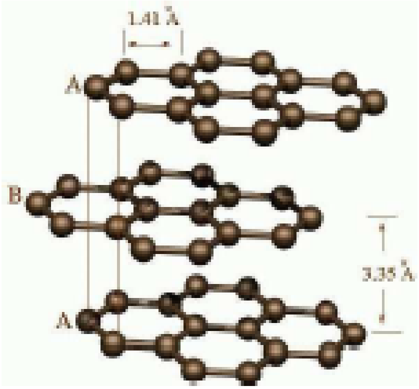
Swift Heavy Ions – Induced Radiation Damage in Graphite

M. Tomut

GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt

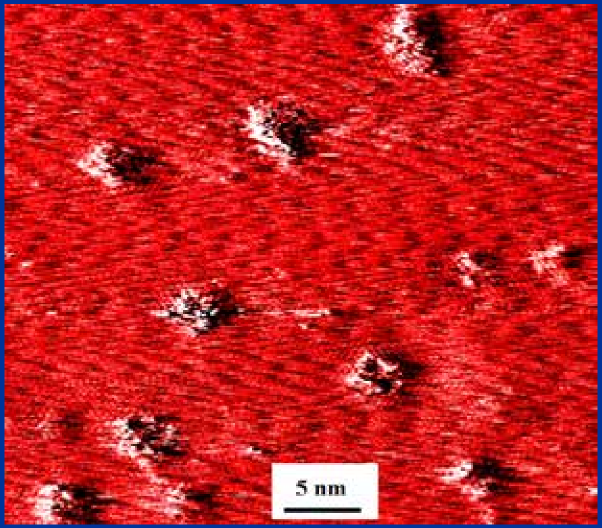
Highly Oriented Pyrolytic Graphite



Heavy ion induced tracks in graphite (HOPG)

STM (HOPG)

U (2710 MeV), at 300 K

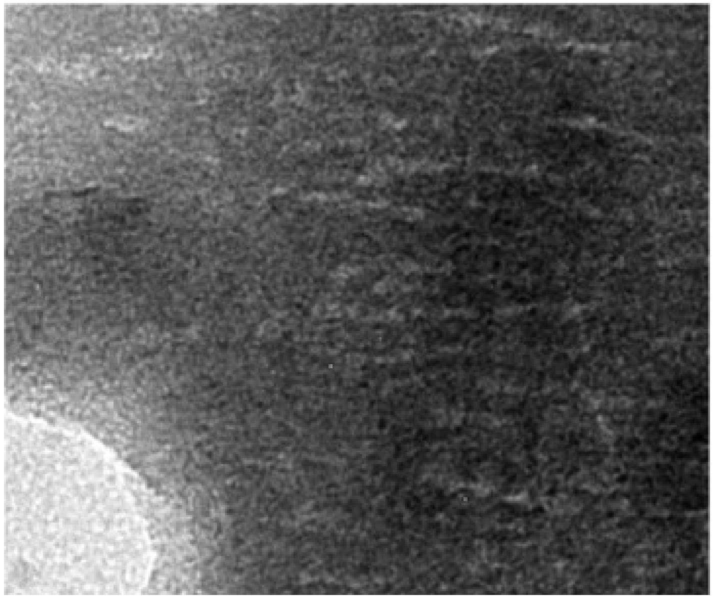


30 nm x 30 nm

Liu et al, PRB 64 (2001) 184115

TEM (natural graphite)

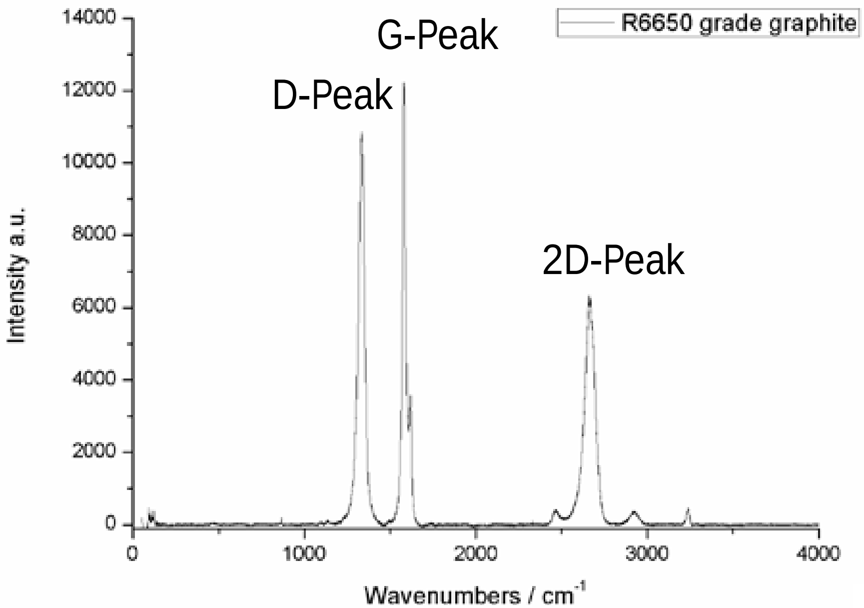
Pb (850 MeV), at 90 K



Dunlop et al, PRB 76 (2007) 155403

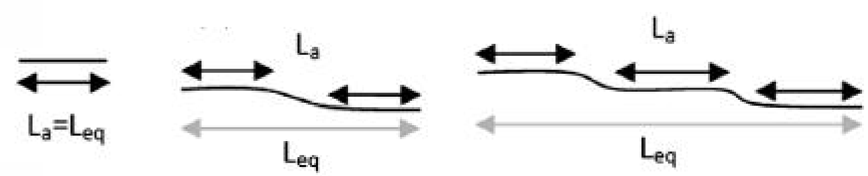
Raman spectra of graphite

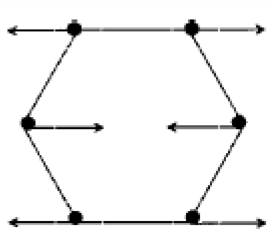
Sensitive to in-plane vacancy type defects



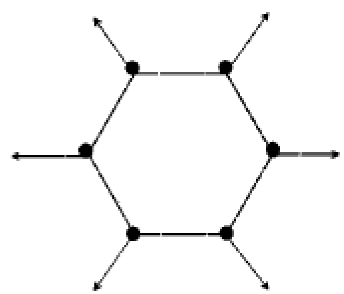
L_a: average in-plane length $L_a = 4.4\left(\frac{A_D}{A_G}\right)^{-1}$ nm

L_{eq}:average graphene length including tortuosity $L_{eq} = 8.8\left(\frac{A_{2D}}{A_D}\right)$ nm





E_{2g} G Mode



A_{1g} D breathing Mode

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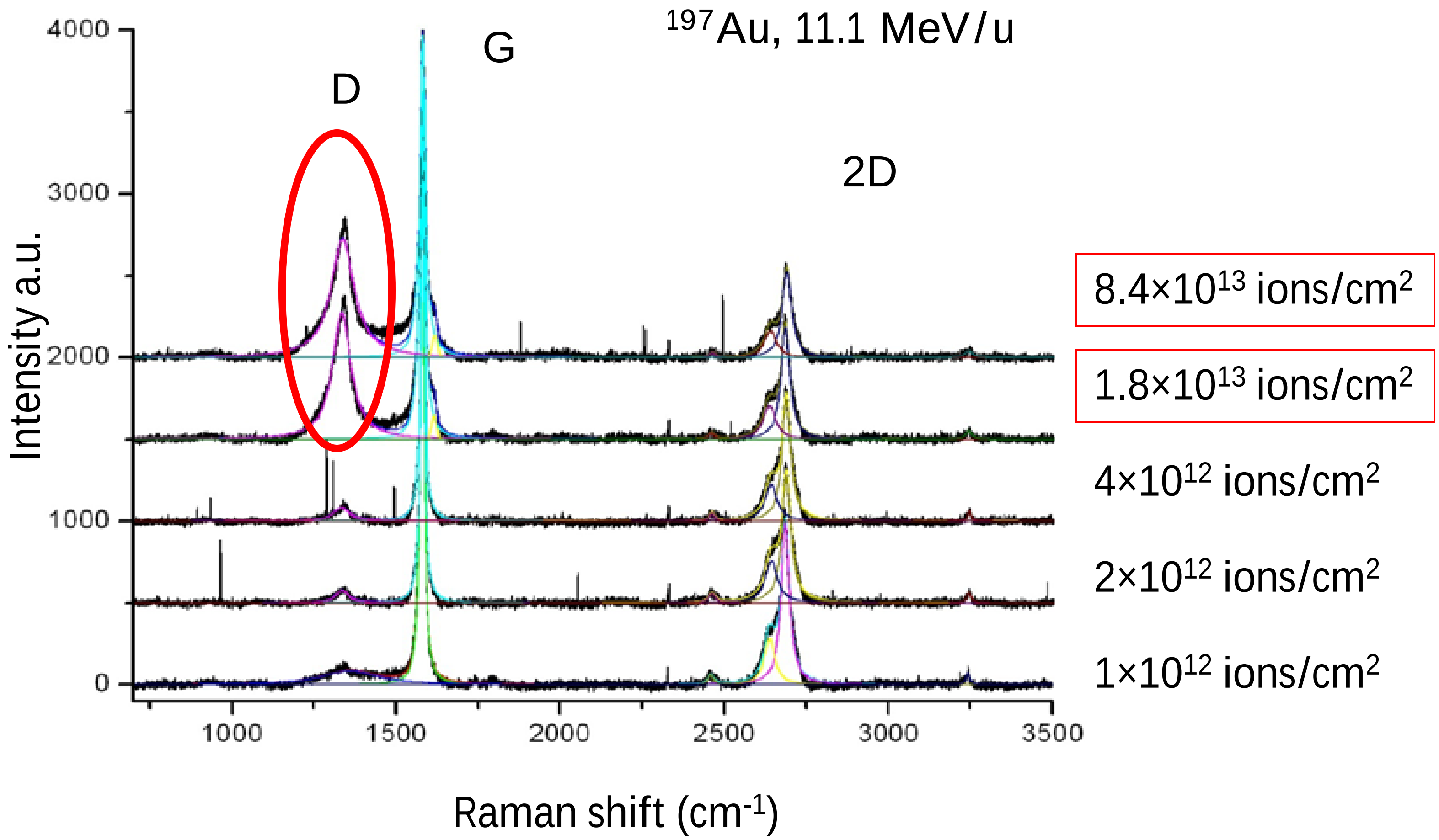
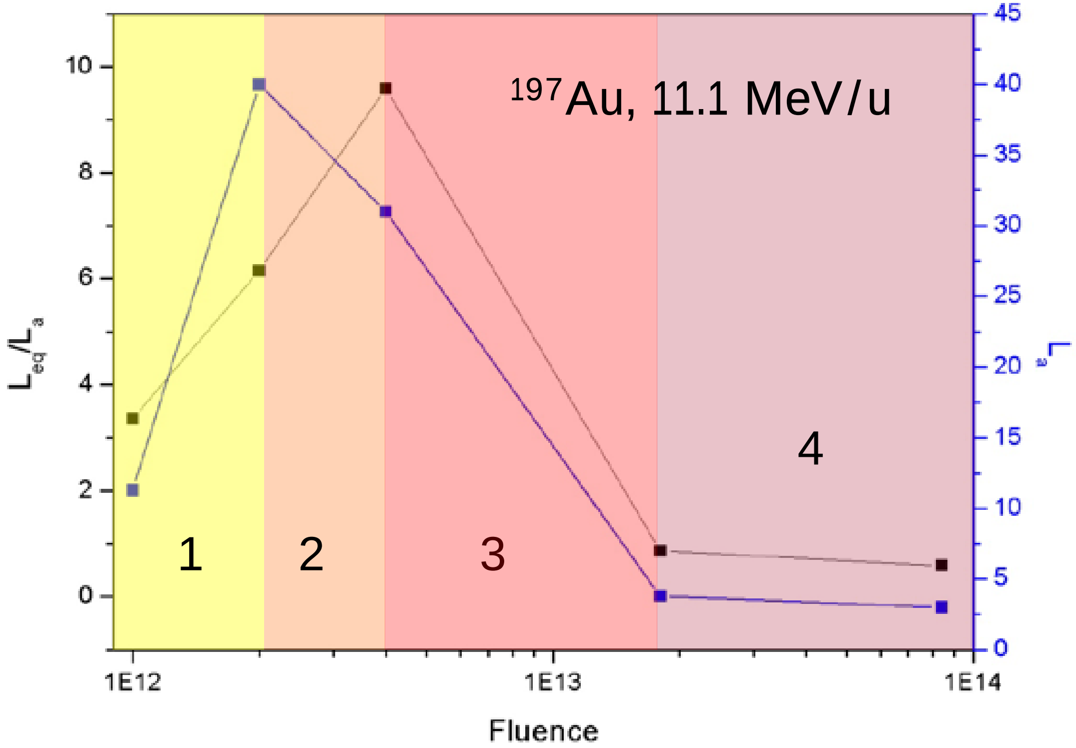
SHI- induced damage in HOPG- Raman spectroscopy



Large increase in damage at fluences where tracks strongly overlap - vacancy clustering

SHI- induced damage in HOPG- Raman spectroscopy

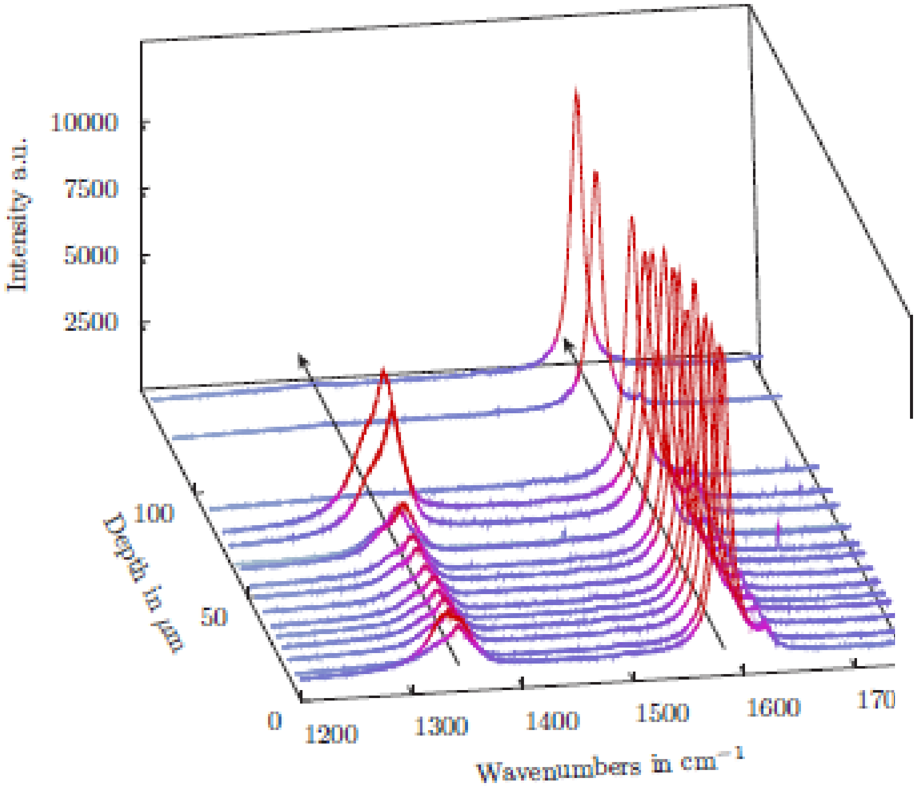
Fluence evolution of Raman graphitic indices in HOPG



1. Annealing of intrinsic defects
2. Bending of graphitic planes
3. Nanostructuring of basal planes
4. Disordering, cross-linking of graphitic planes(accompanied by strong hardening-indentation measurements)

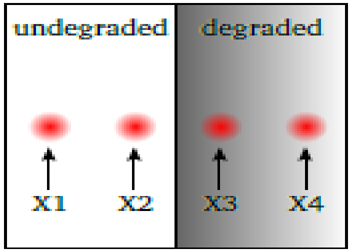
La – in-plane coherence length
Leq/La - characterizes tortuosity of graphitic planes

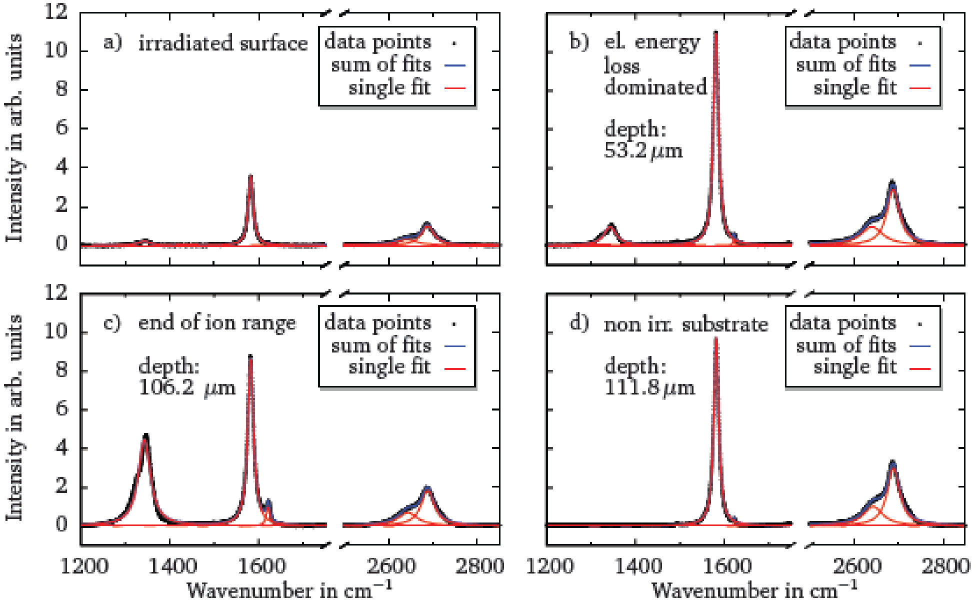
Raman depth profiling of damage in HOPG by succesive cleaving



Damage evolution with depth

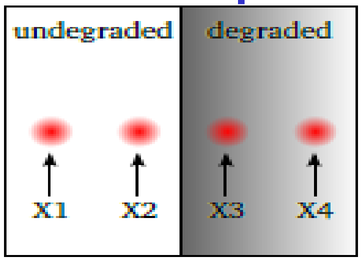
Sample



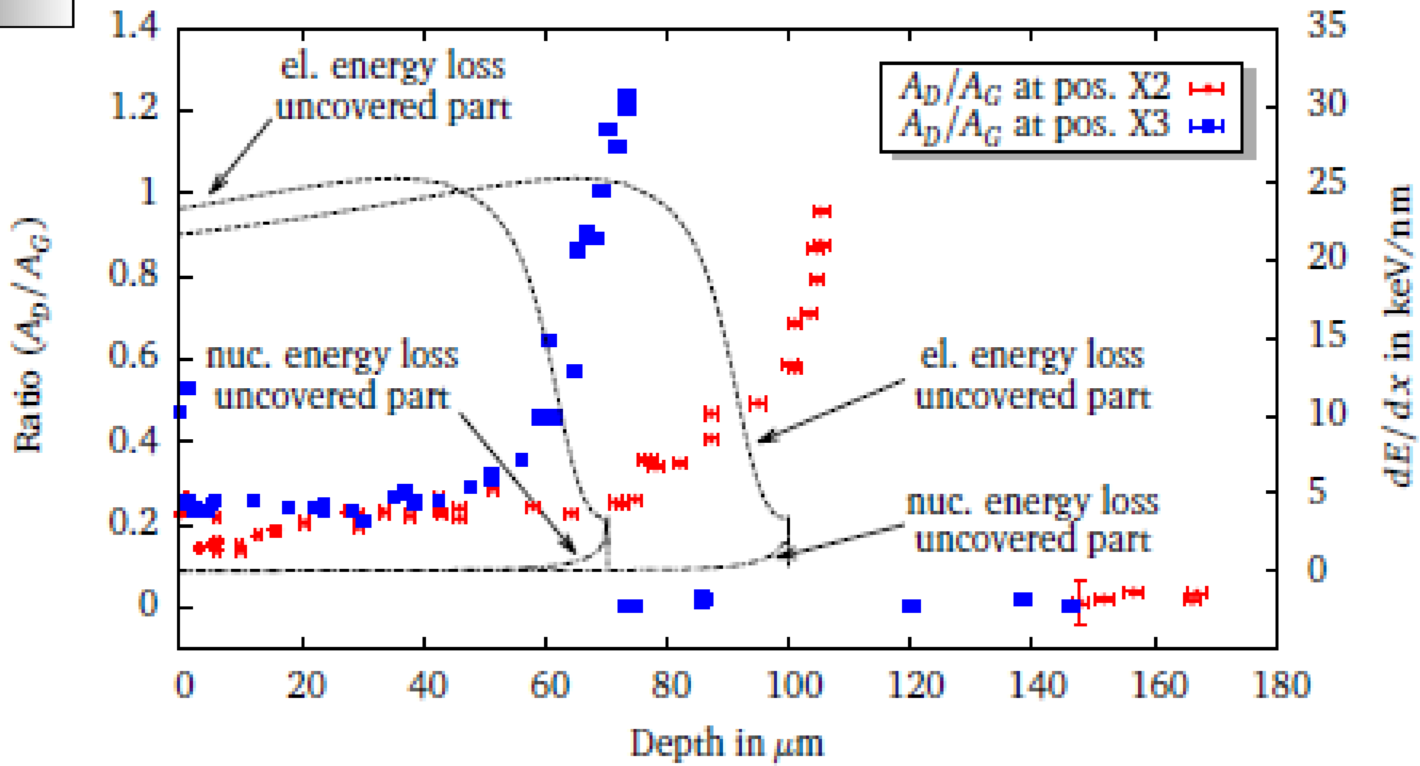


Raman depth profiling of damage in HOPG by succesive cleaving

Sample

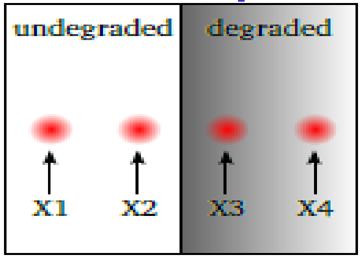


Damage evolution with depth

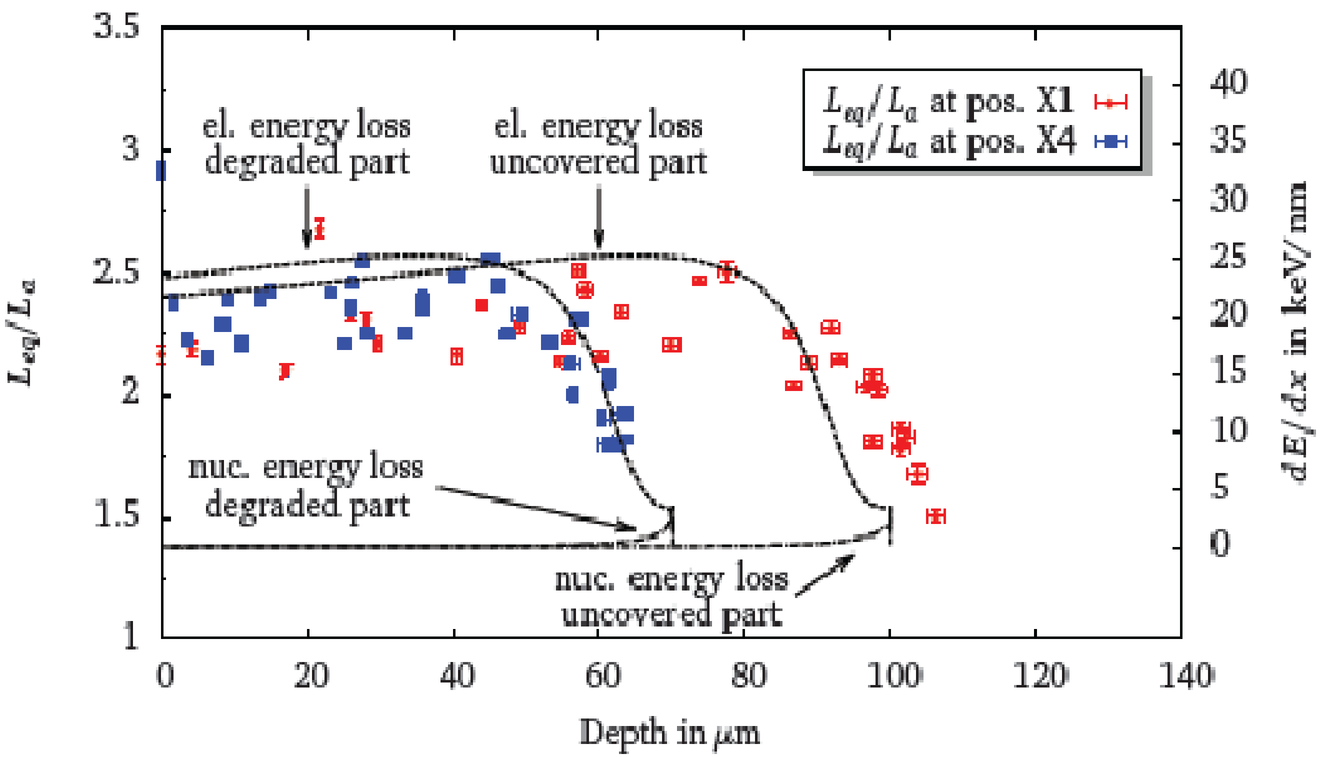


Raman depth profiling of damage in HOPG by succesive cleaving

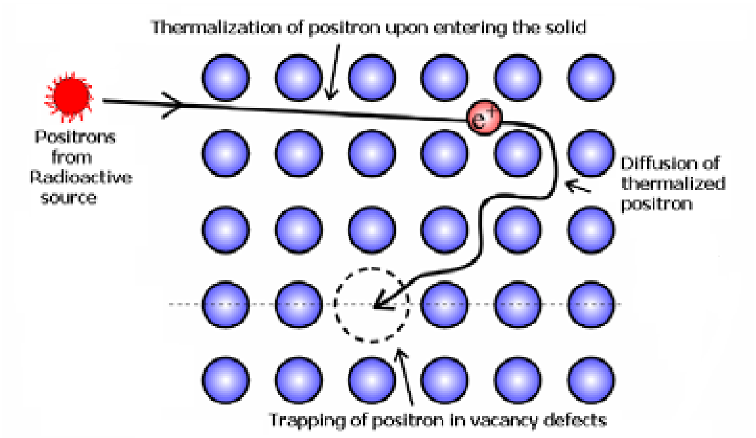
Sample

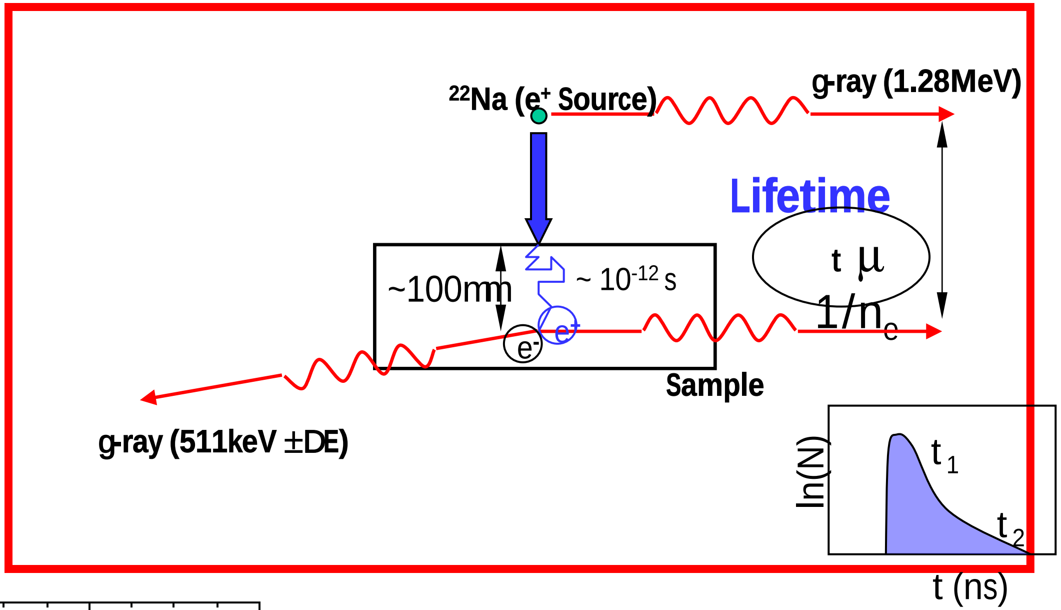


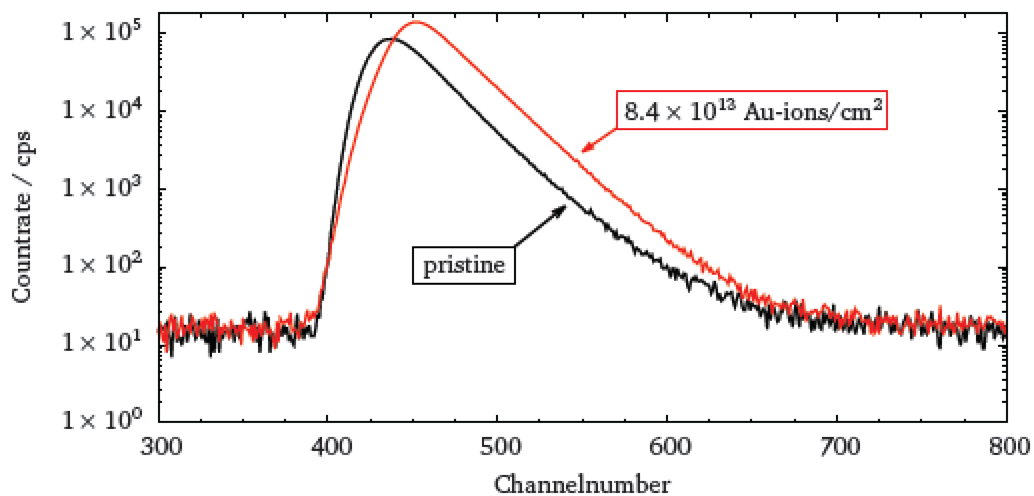
Graphitic layers tortuosity evolution with depth



Positron annihilation spectroscopy

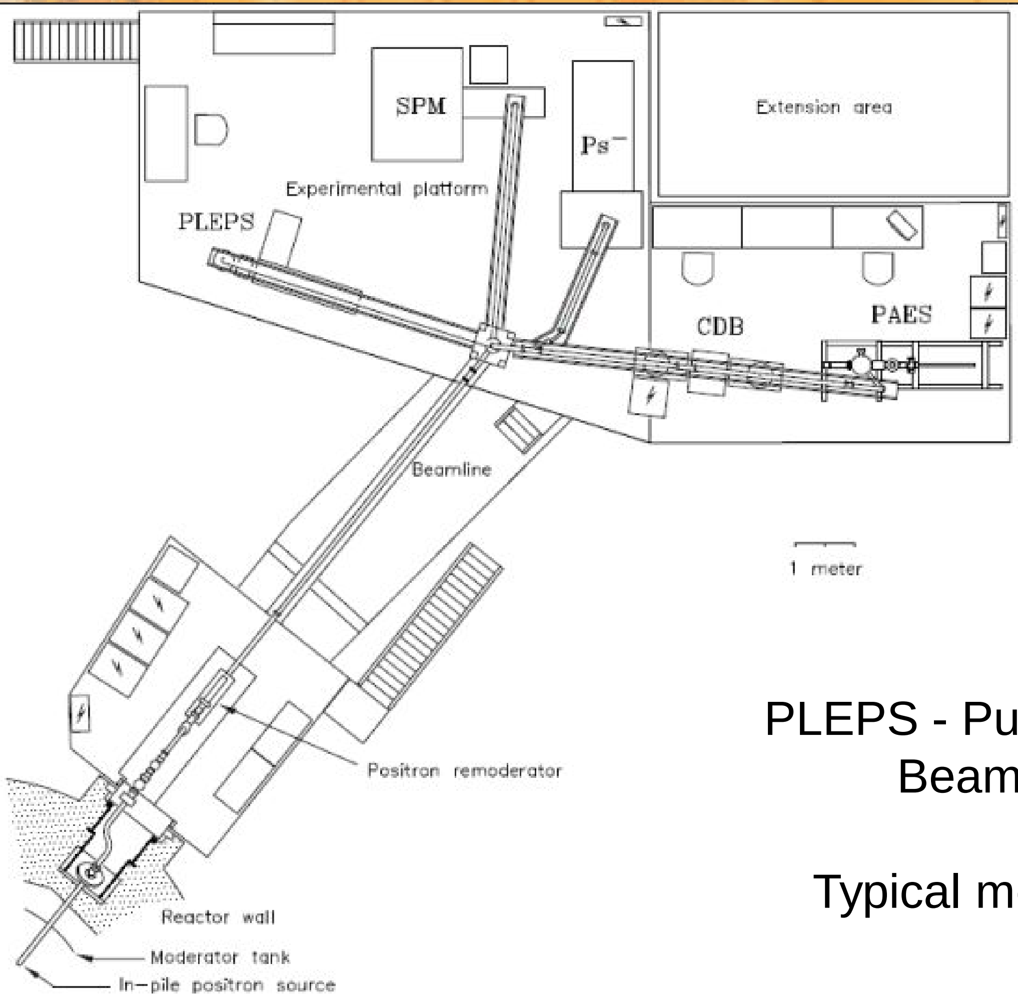






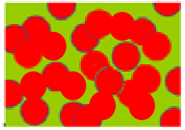
PALS pristine and irradiated HOPG

PLEPS source at FRM II

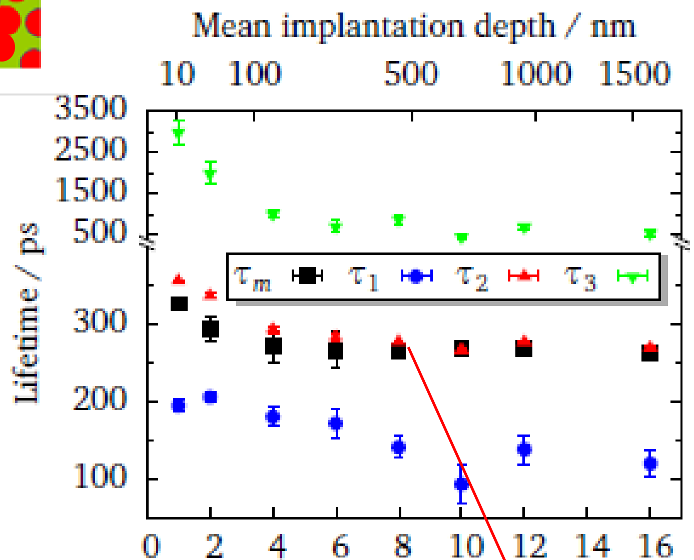


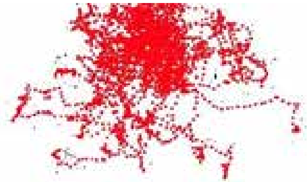
PLEPS - Pulsed Low Energy Positron system
Beam energy at sample position:
E = 0.2-18 keV
Typical measurement time per spectrum:
DBS: 20 min

Vacancy clusters in SHI irradiated HOPG- positron annihilation

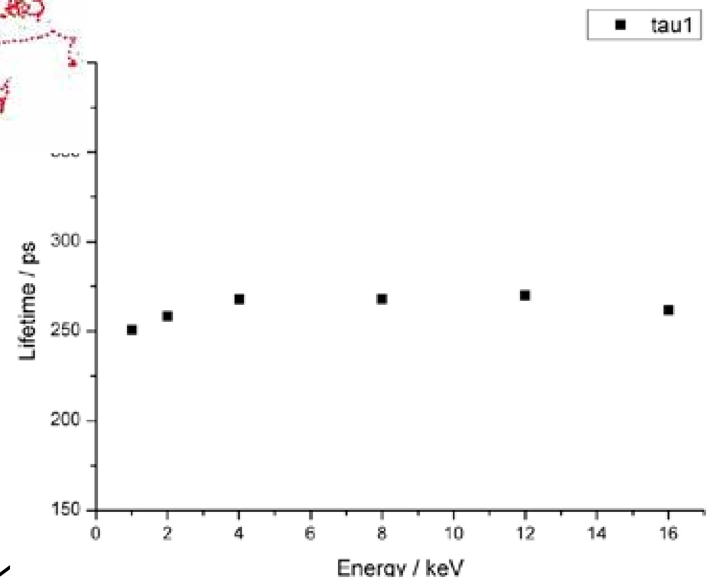


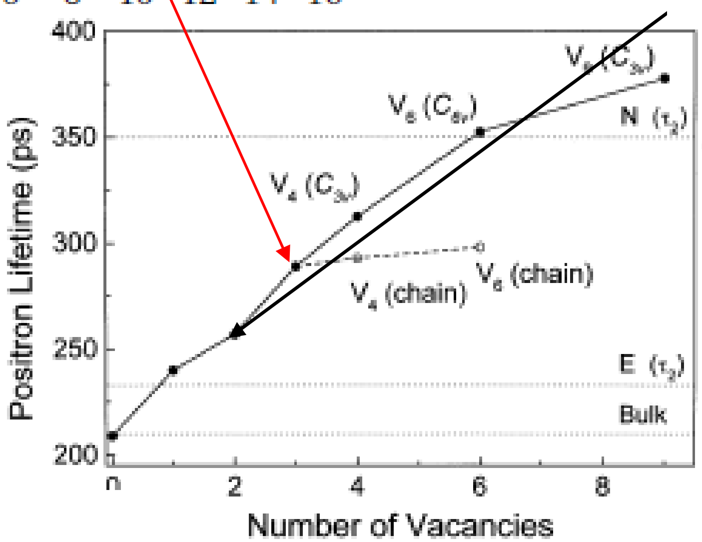
Electronic stopping





Elastic collisions





- τ_1 Reduced bulk lifetime
- T_2 Divacancies and vacancy chains
- τ_3 positronium- cavities?

ÿτ_1 - Divacancies and vacancy chains 100%

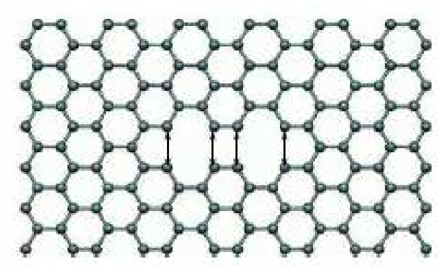
Tang et al,

Phys. Rev. Let . 82 (1999) 2532

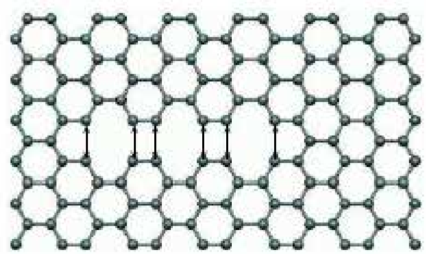
Mechanism of bending of graphitic layers

Unrelaxed vacancy clusters in graphite

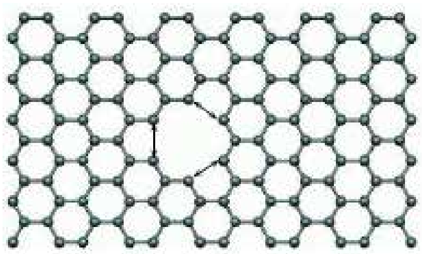
V4 boat



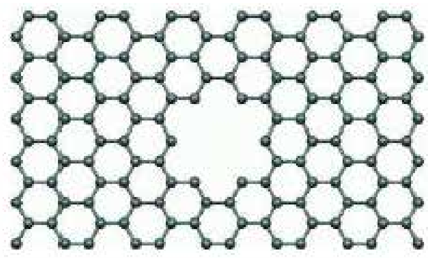
V6 boat



V4 disc

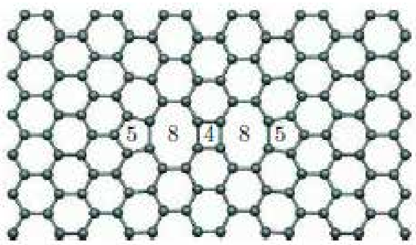


V6 disc

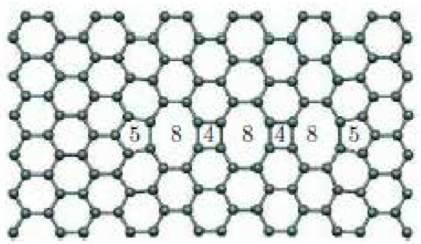


Optimised vacancy clusters in graphite
Formations of pentagons and octagons

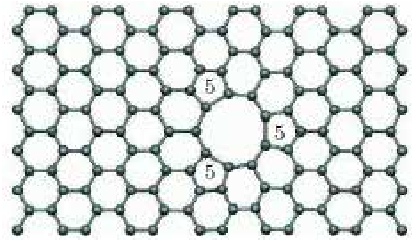
V4 boat



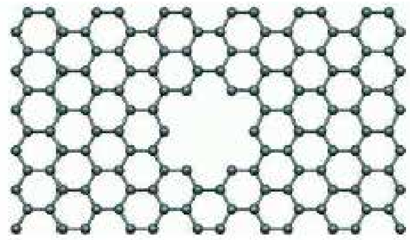
V6 boat



V4 disc

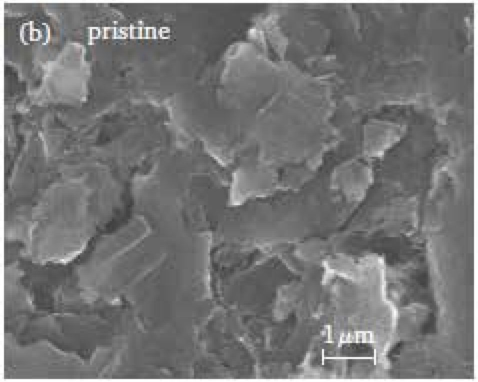


V6 disc



A.A. El-Barbary, Ph.D. Thesis

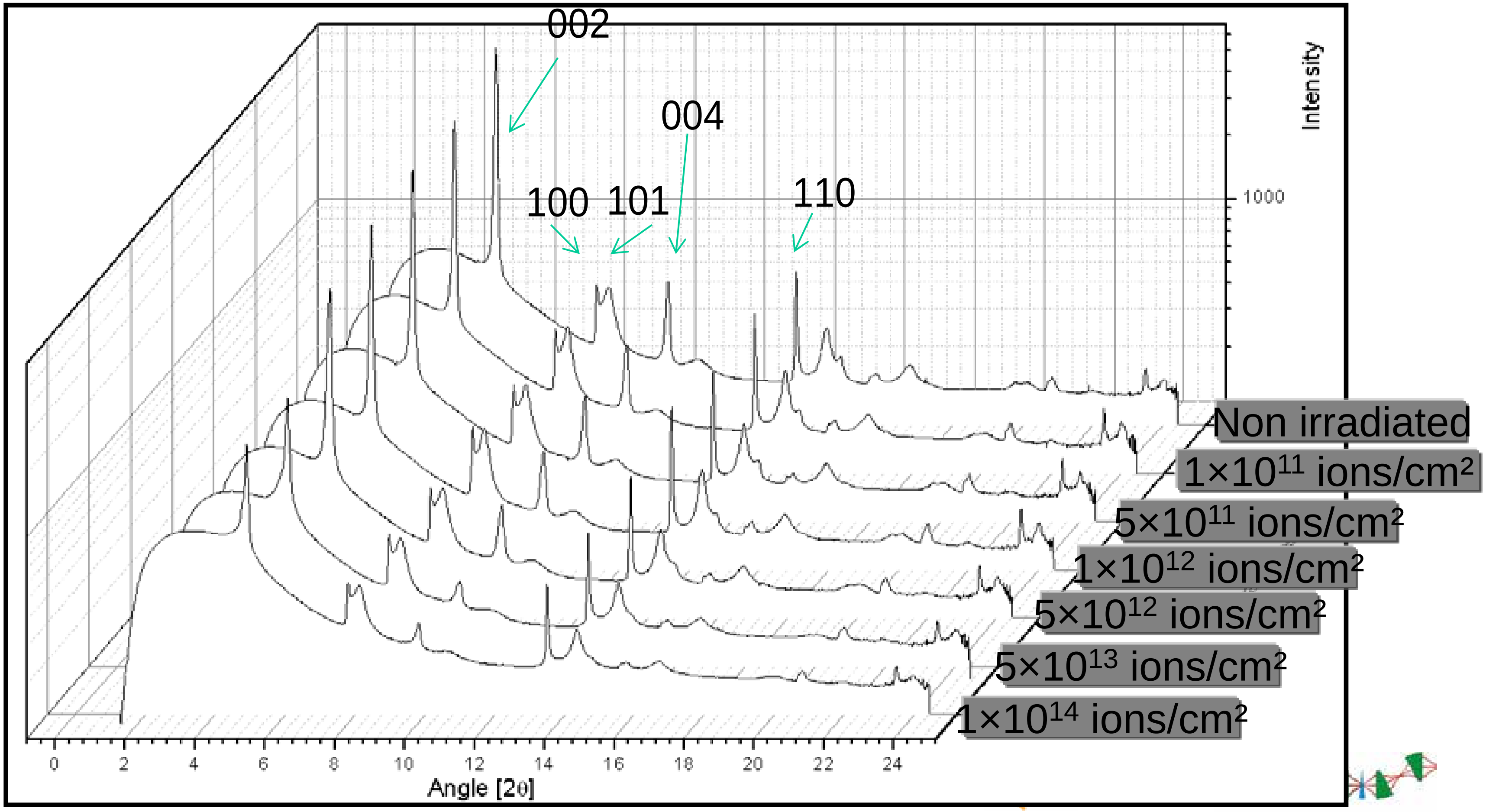
Polycrystalline isotropic graphite



XRD Characterization of Radiation Damage in Graphite Induced by GeV heavy ions

Wavelength: 0,29135Å
Beam spot: 2x2mm

Fluence series: 10^{11} to 10^{14} ions/cm²
Sample: fine grained isotropic graphite
Flux: 10^{10} ions /cm² s
Energy: 3,6MeV/u



Depth profiling of defects in polycrystalline grapite by Raman investigation on sample cross-section

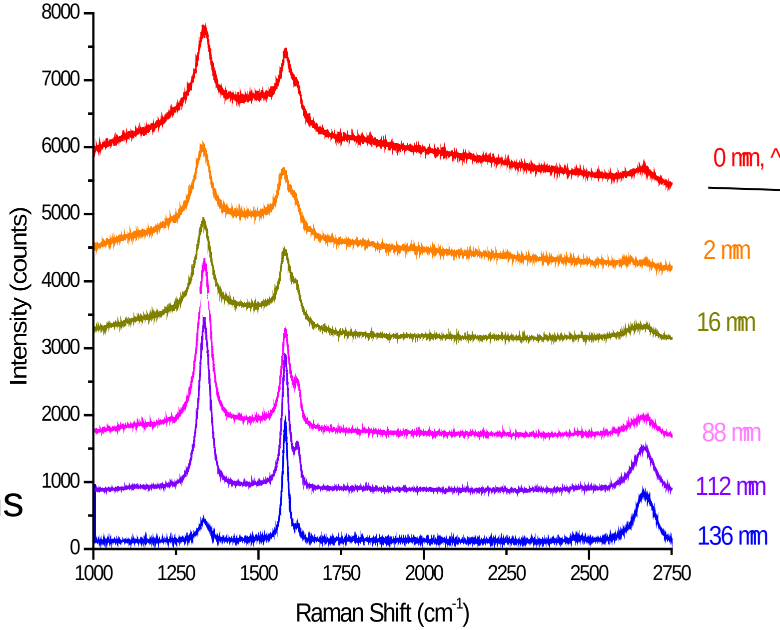
Fine-grained isotropic graphite exposed to $1x10^{13}$ ^{238}U ions/cm^2, 11.1 MeV/u

Raman spectra along the ion trajectory

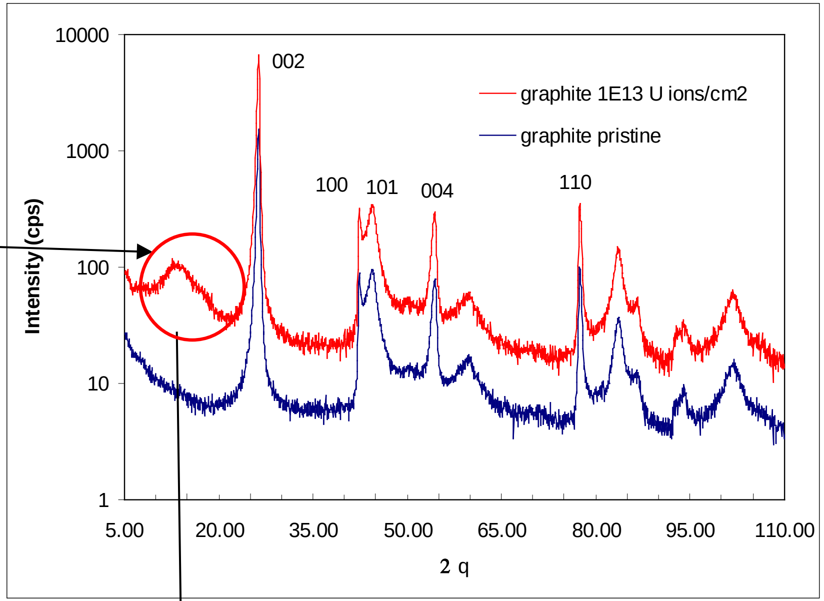
Sample surface

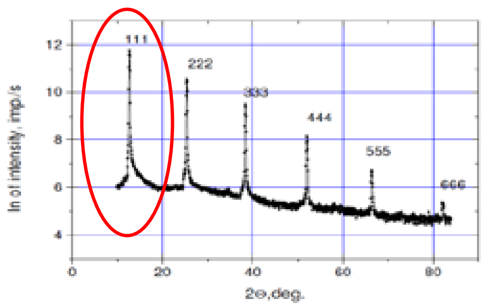
Elastic collisions

Bulk pristine



XRD



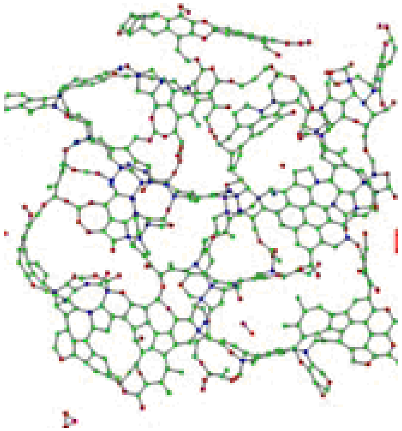


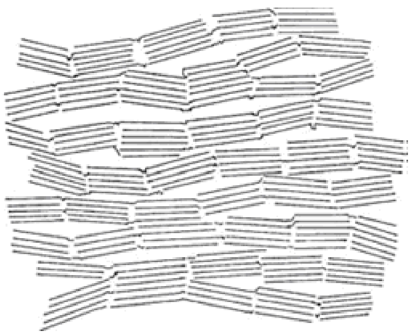
Fullerite

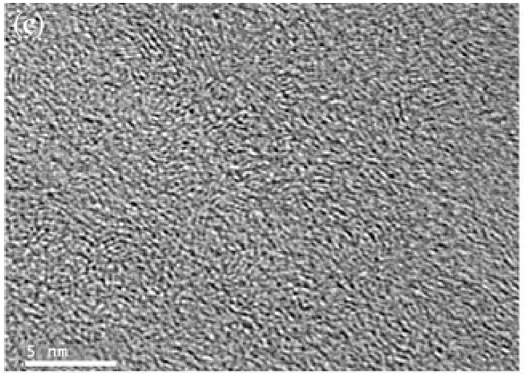
Specific structural changes induced by SHI in graphite

Electronic stopping

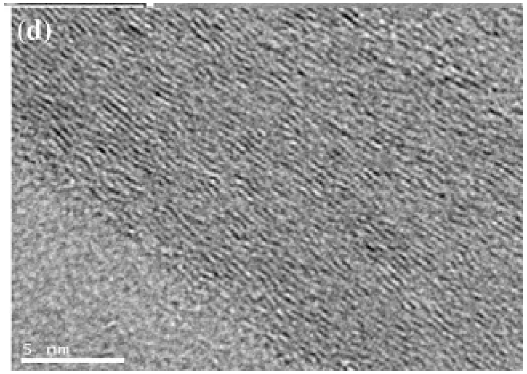
Elastic collisions







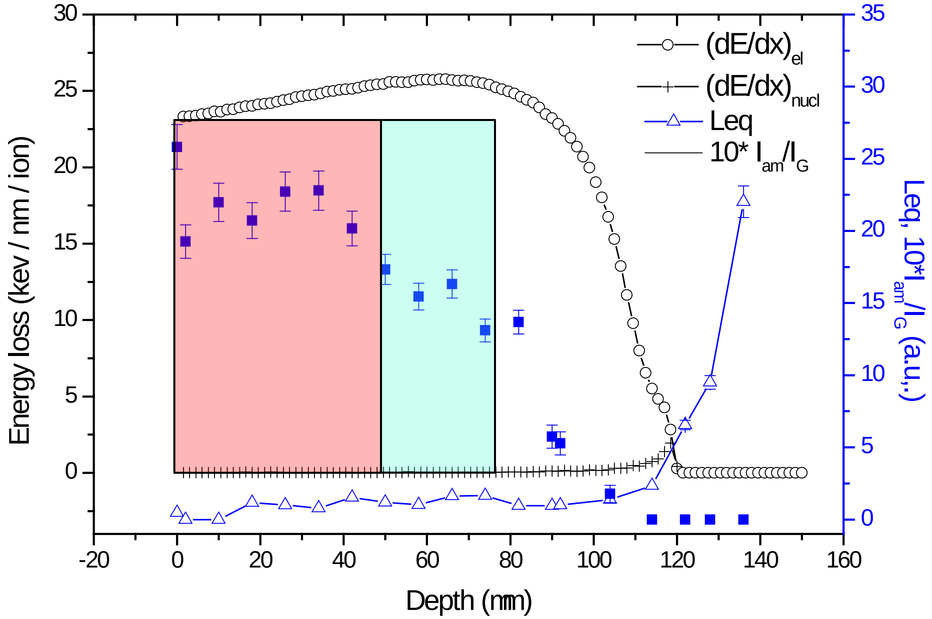
TEM

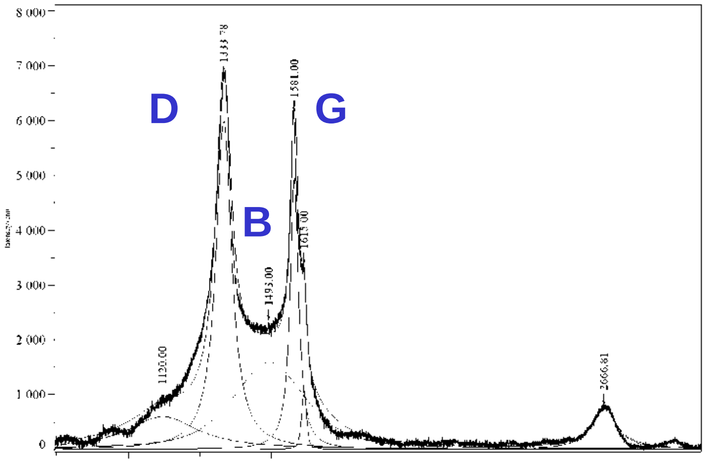


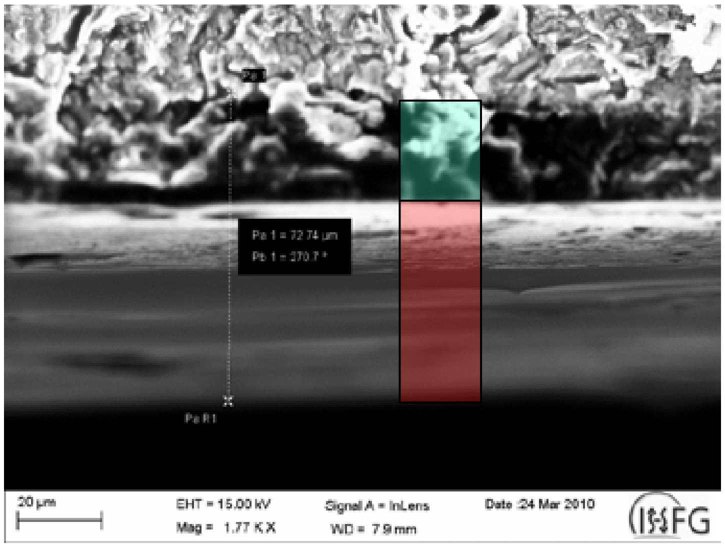
Ammar et. al,Carbon, 2010

Depth profiling of defects in isotropic graphite using Raman graphitic indices

Ib/Ig with depth

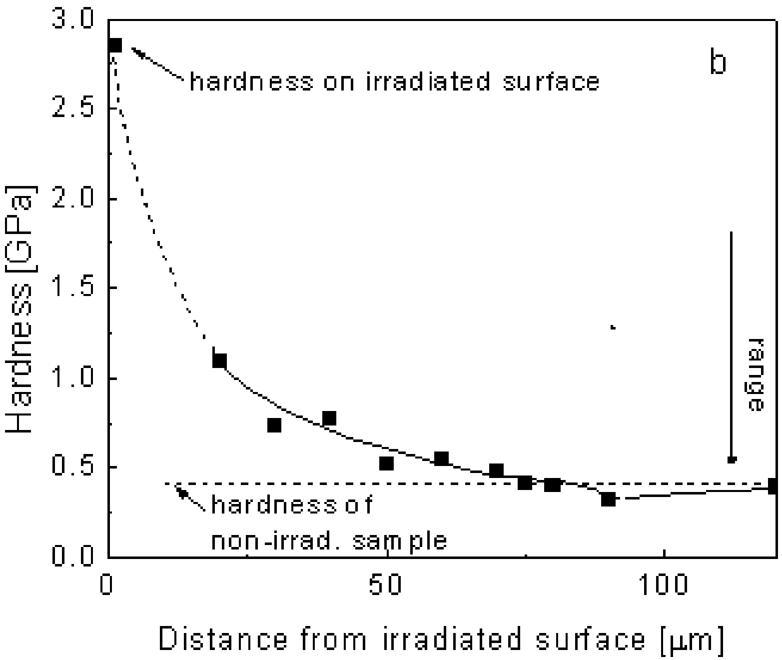




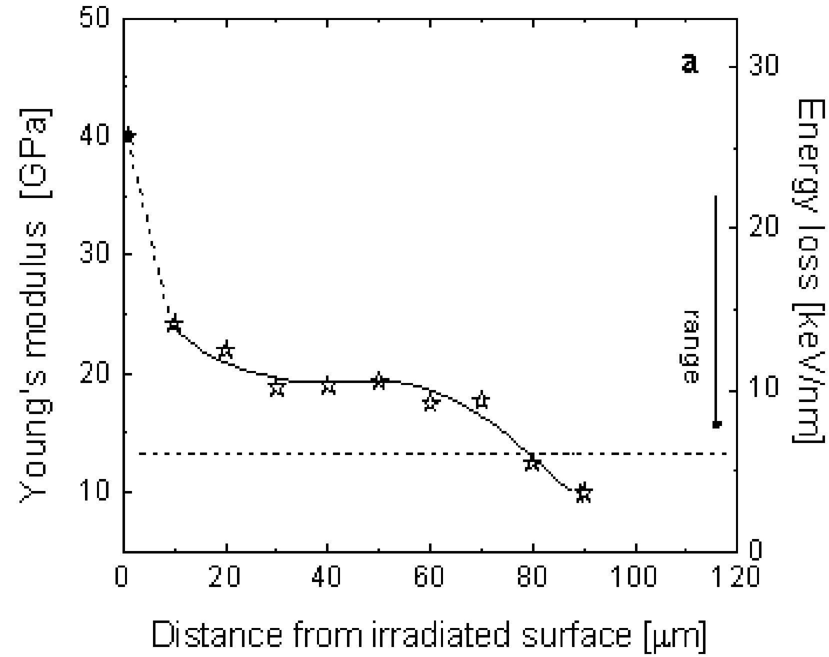


Depth profiling of mechanical properties of U irradiated isotropic graphite- nanoindentation

Hardness with depth



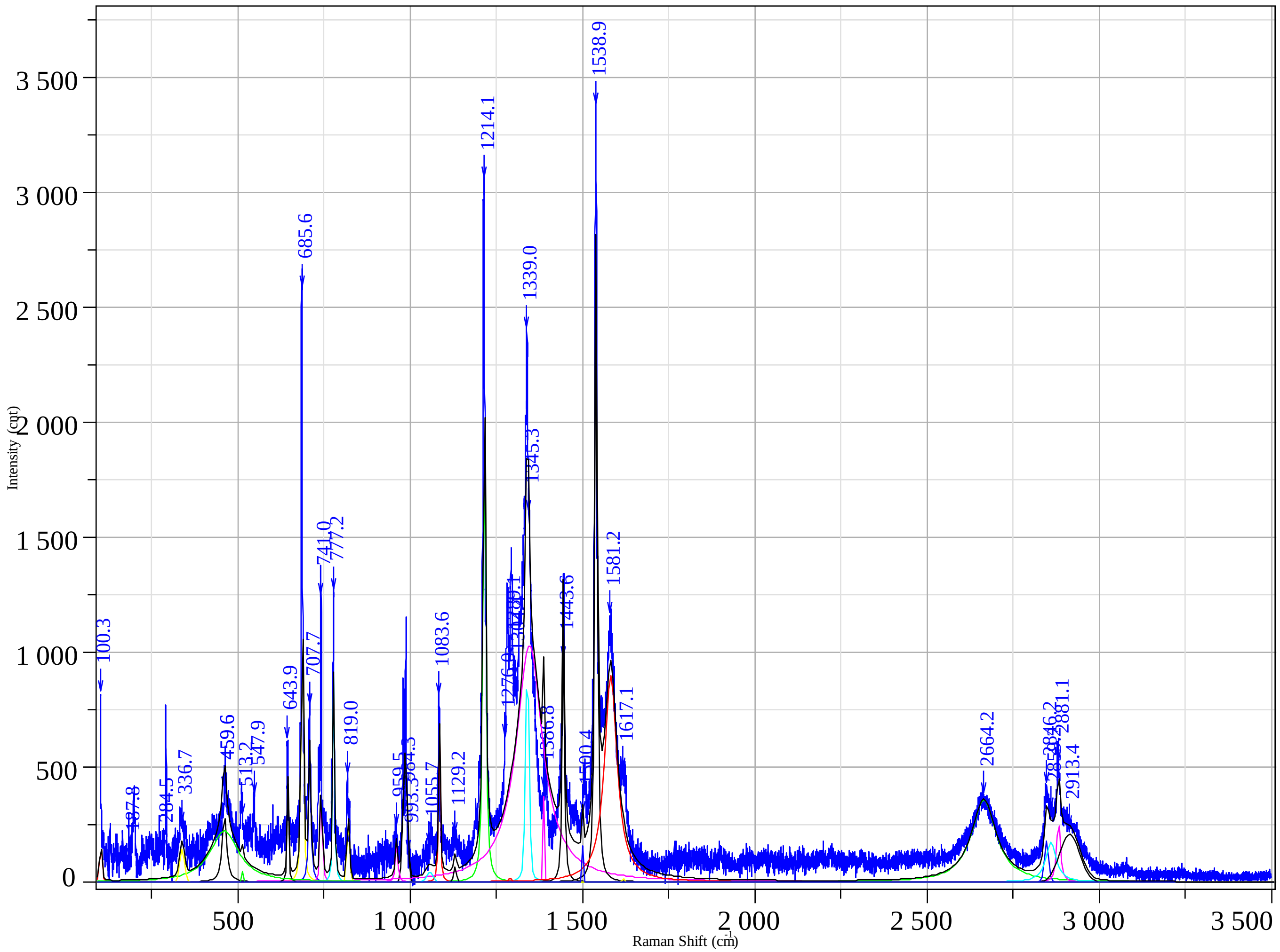
Elastic modulus with depth



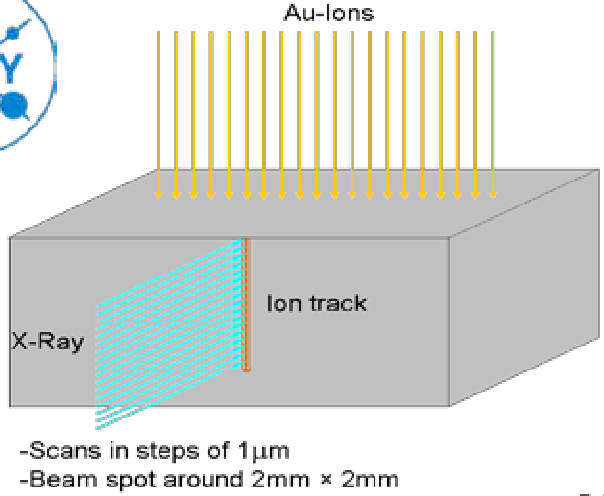
I. Manika et al.
Fullerenes Nanotubes and Carbon Nanostructures, 2012

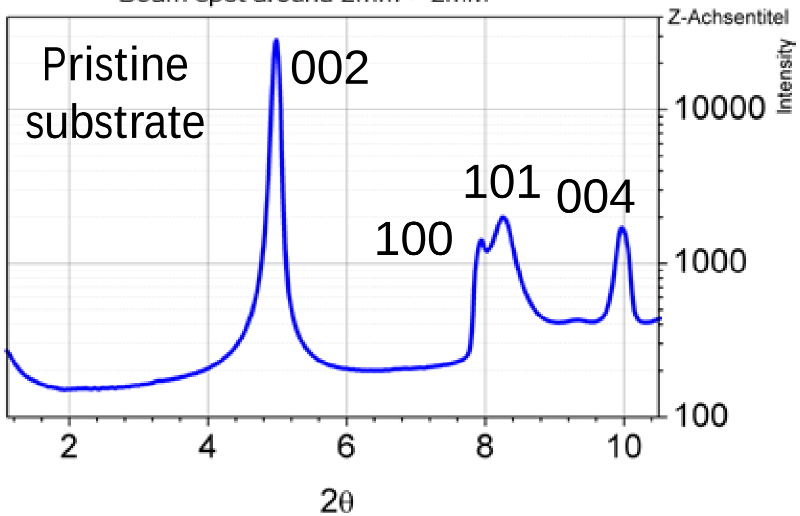
- Strong hardening on the surface
- Hardness and Young modulus lower than virgin at the interface irrad./non-irrad. due to residual elastic stresses

Raman spectrum taken in in cross section on “fresh” U irradiated isotropic graphite



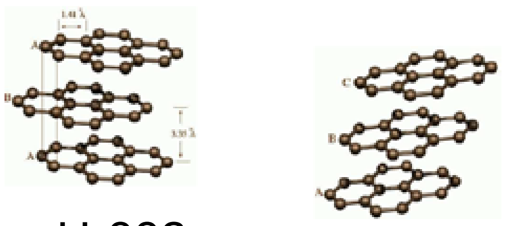
XRD along ion trajectory

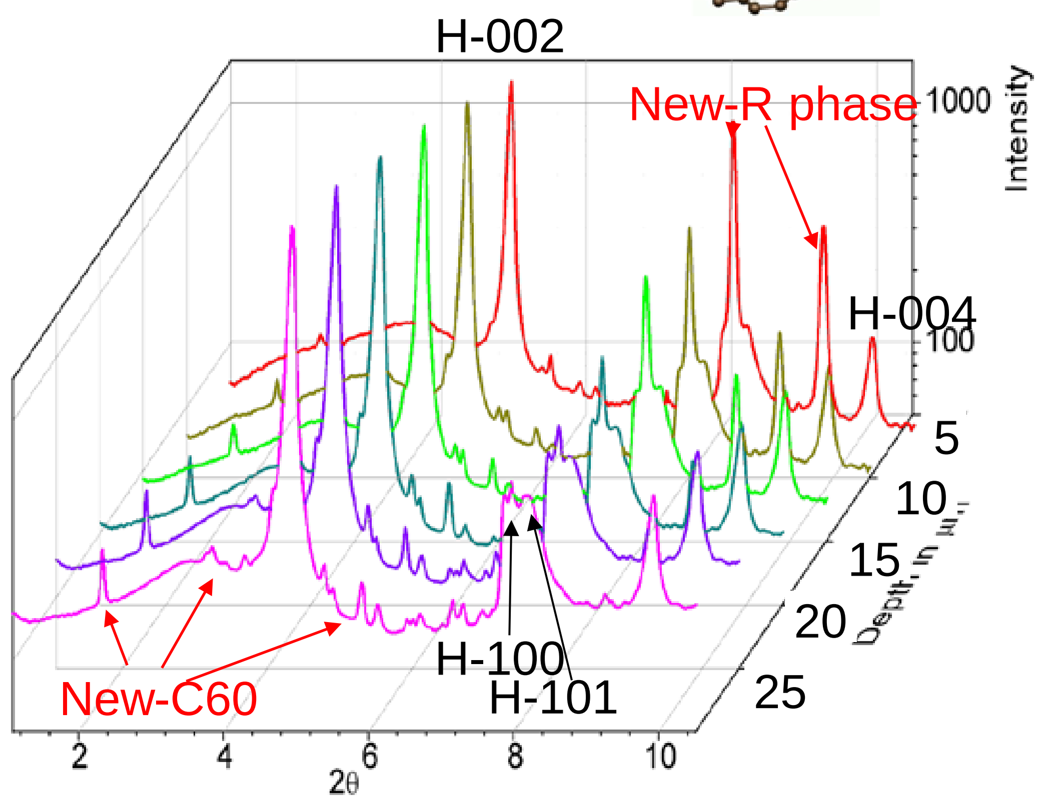




Wavelength: 0,291 Å, Beam spot: 2x2mm

^{238}U, 5.9 MeV/u, 1E13 I/cm^2

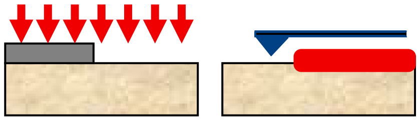




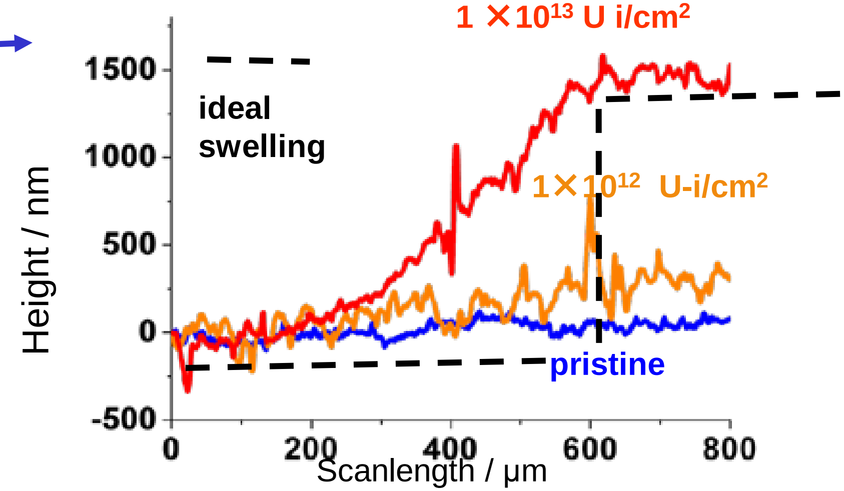
Swift heavy ion -induced property degradation of isotropic graphite

Ion- induced swelling and creep?

Profilometry



Low intensity U beams:
4.8 MeV/u,
flux $1x10^{8}$ i/cm² s

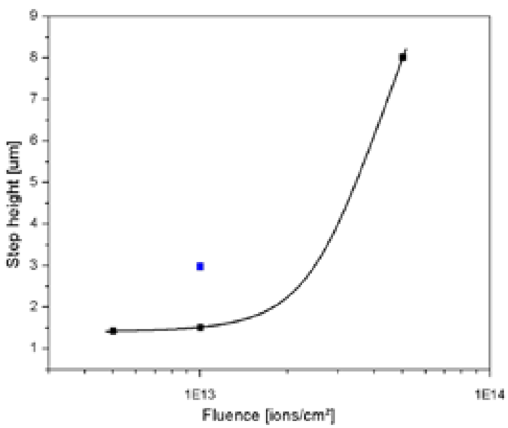


Out-of plane swelling - Profilometry

High intensity Au beams:
4.8 MeV/u,
flux $1x10^{10}$ i/cm² s

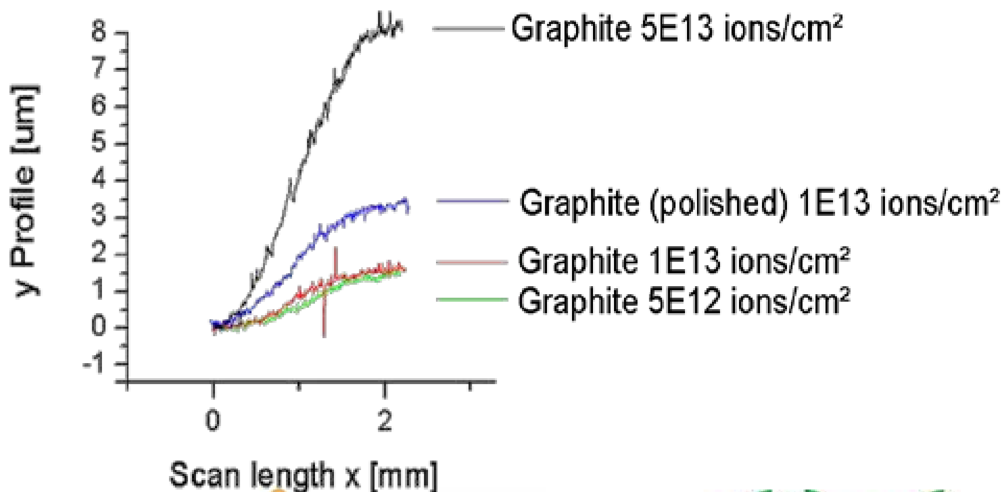
$5x10^{12}$ i/cm²

Step size dependence on fluence



$5x10^{13}$ i/cm²

$1x10^{14}$ i/cm²



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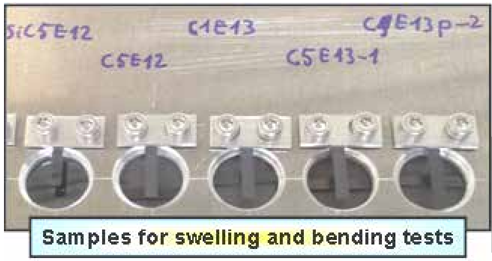
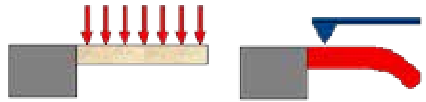
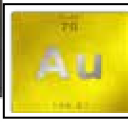
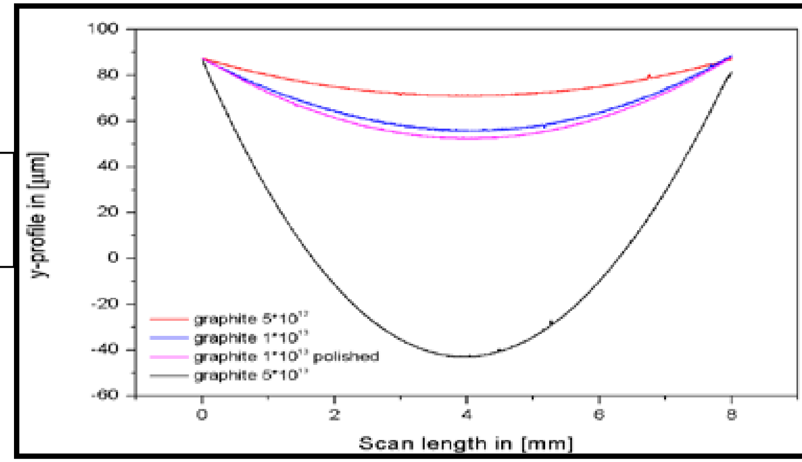
Irradiation- induced stress







Charge state: + 25
Energies: 4.8 MeV/u
Fluences up to 5×10^{13} i/cm²



Swelling ð Stress ð Bending

$$\sigma_{rr}^{f} \simeq -\frac{E_s h_s^2}{6(1-\nu_s)h_f} \frac{1}{R_r}$$

E_s = elastic modulus

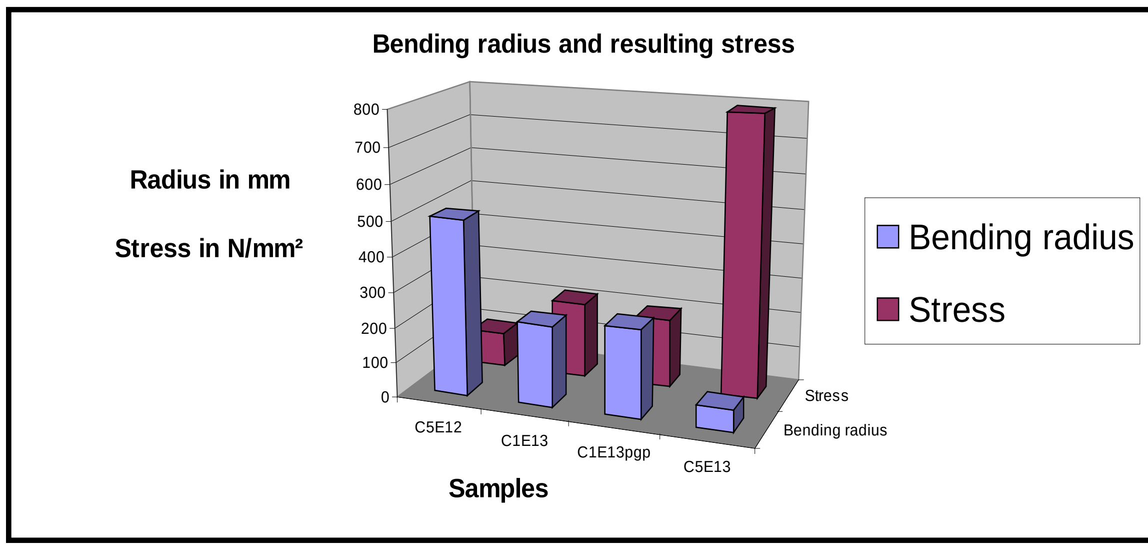
h_s = sample thickness

h_f = irradiated layer thickness

R_r = bending radius

n_s = Poisson number

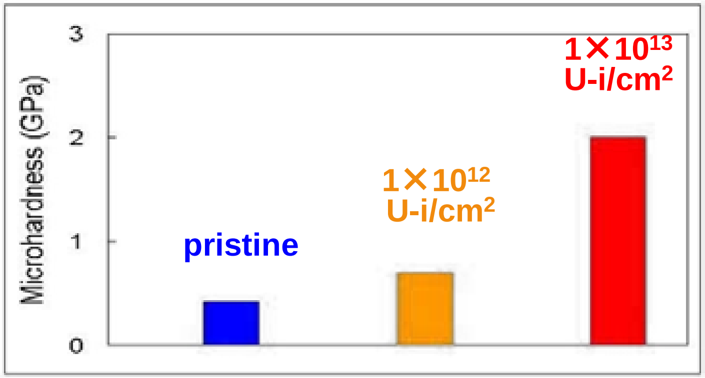
s^f_{rr} = stress



Hardening and embrittlement of ion- irradiated graphite

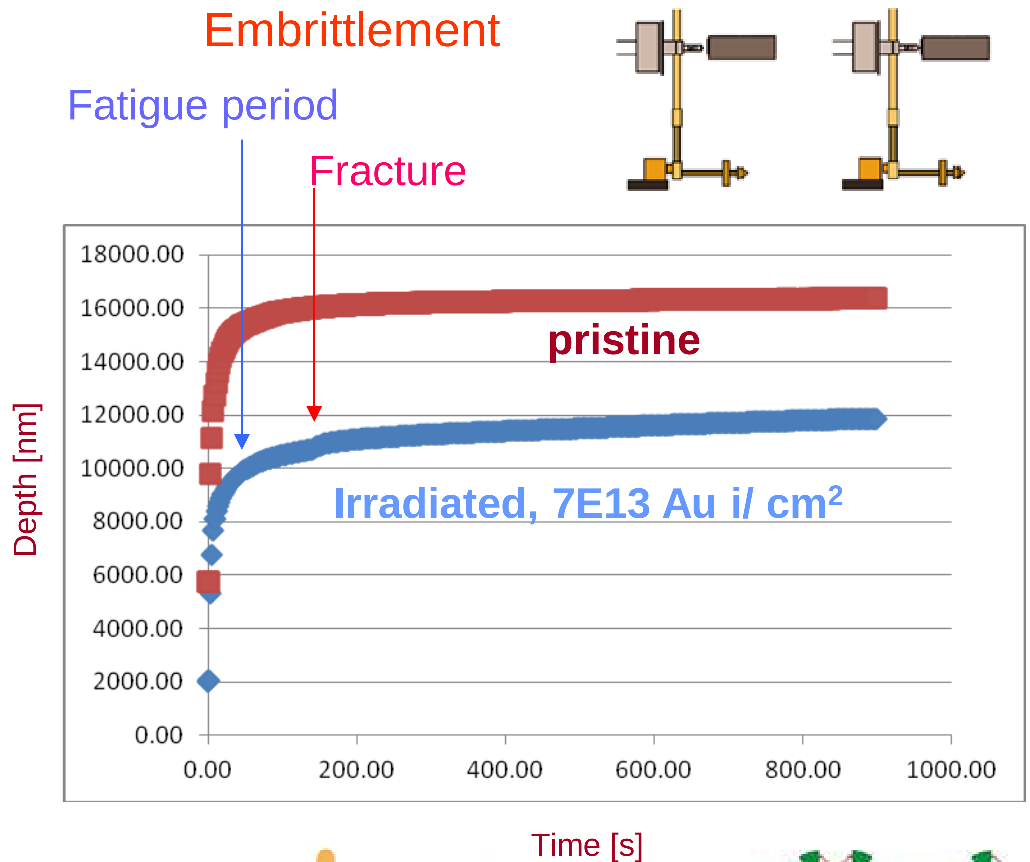
Nanoindentation testing of irradiated graphite

Hardening



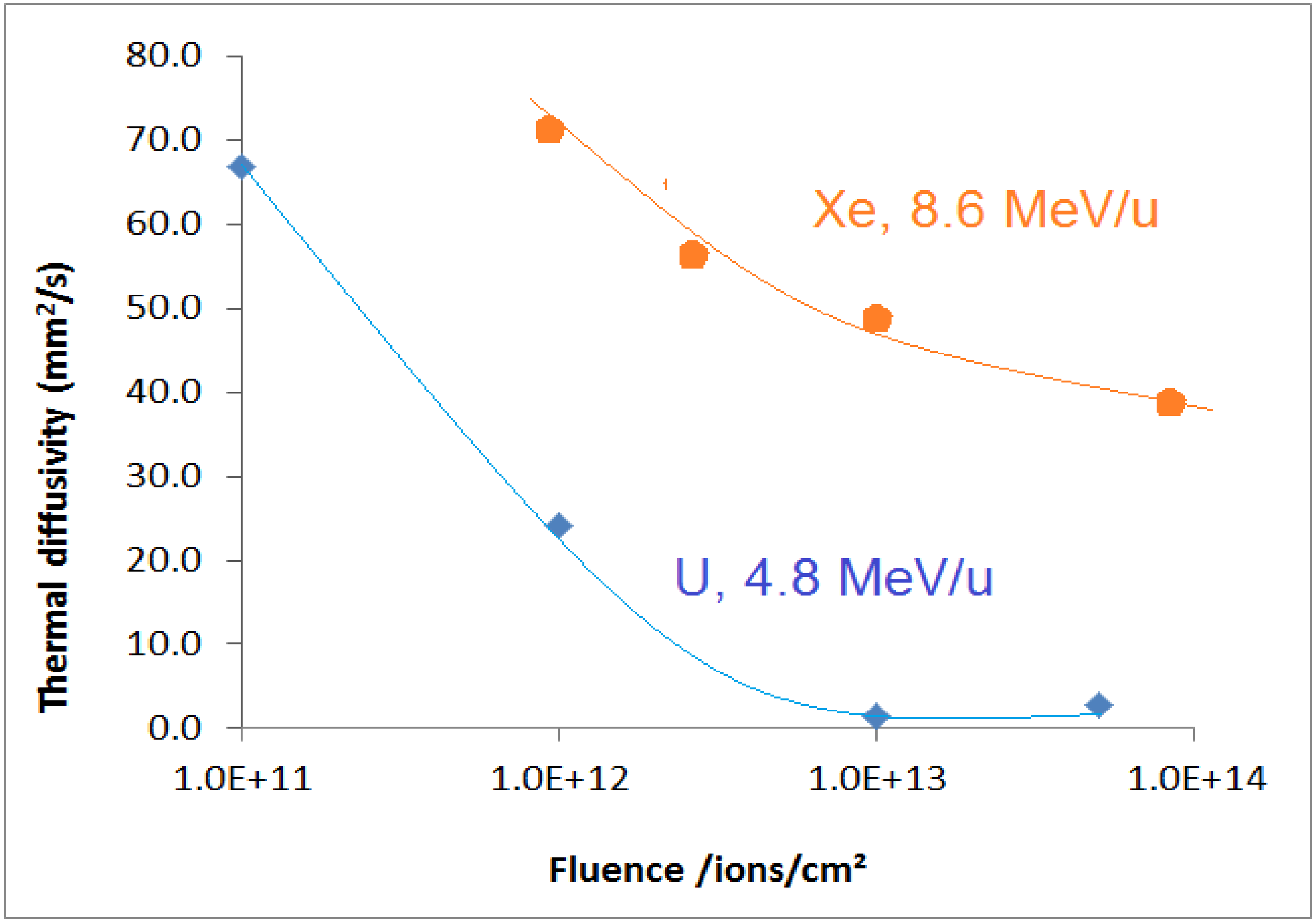
Strong hardening
- Indentation -

Cube Corner 20 mN max load; comparison pristine and irradiated samples



Ion-induced thermal diffusivity degradation of graphite

Comparison U vs Xe irradiation for isotropic graphite



Summary

- For HOPG-defects produced by SHI in graphite in the region dominated by electronic stopping- less efficient than in the nuclear stopping regime
- Increased sensitivity of the surface to damage creation via electronic stopping
- SHI irradiation of isotropic graphite induces a hard disordered sp2 phase in the electronic stopping range- fullerene-related structures in the track core?
- For smaller crystallite (polishing)- SHI- induced damage is more efficient due to the confinement of energy deposition

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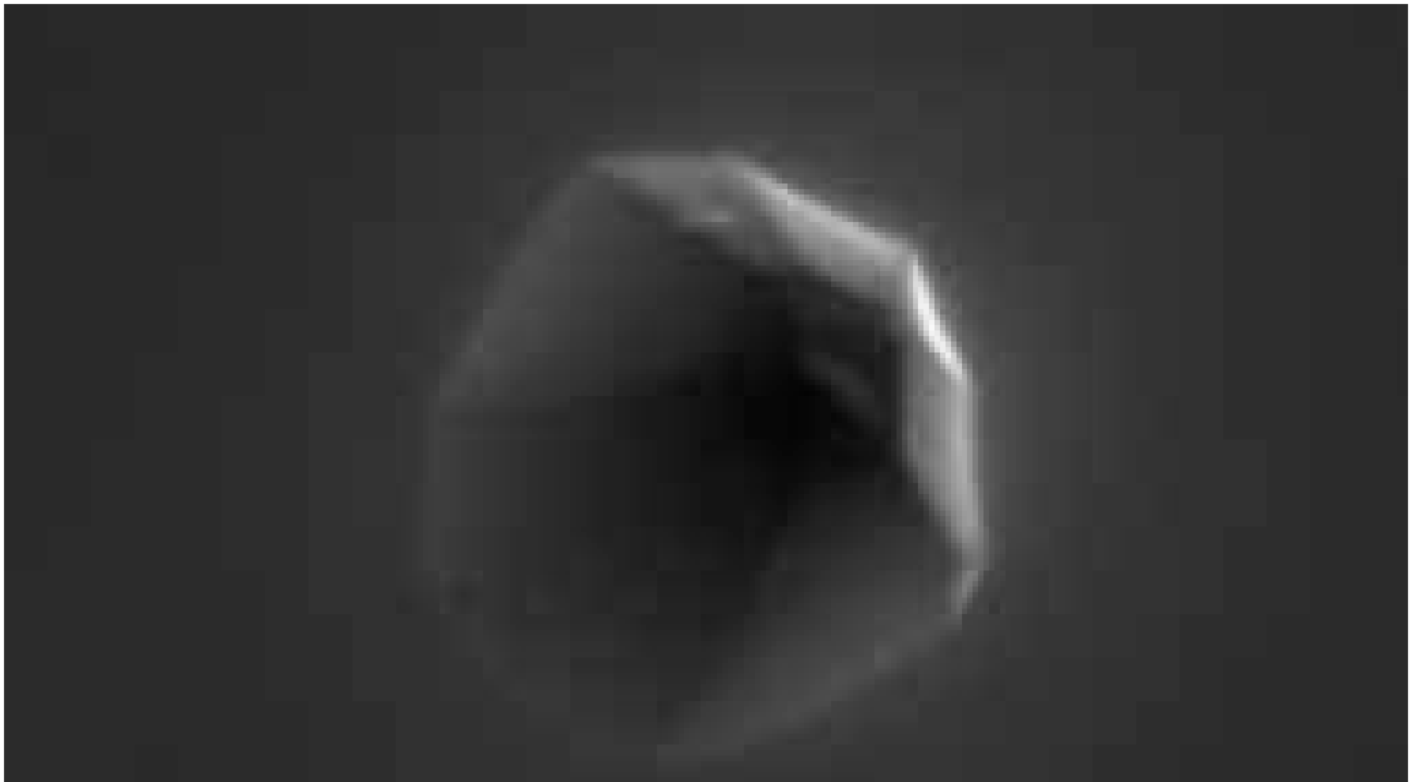

Thank you for your attention!