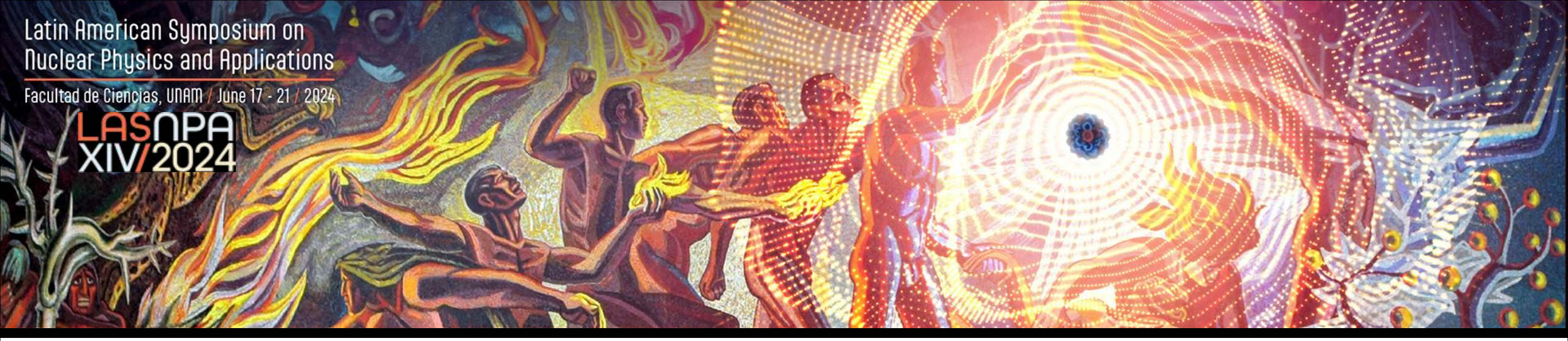

Endurance of HOPG to Ionizing Radiation: Advancements in Target Development for Nuclear Reactions

Marcilei Aparecida Guazzelli

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M.A. Guazzelli, L.H. Avanzi, V.A.P. Aguiar, A. C. Villas-Boas, S. Alberton, S.H. Masunaga, M. T. Escote, E.F. Chinaglia, K. Araki, M. Nakamura, M.M. Toyama, F.F. Ferreira, M. T. Escote, R.B.B. Santos, N.H. Medina, J.R.B. Oliveira, F. Cappuzzello, M. Cavallaro

MOTIVATION

NUMEN project - Nuclear Matrix Elements in Neutrinoless Double Beta Decay - nuclear double charge exchange (DCE) reactions are investigated at LNS/INFN (*Laboratori Nazionali del Sud, Catania, Italy*). This project has important implications for neutrino physics and possibly for the Standard Model of elementary particles.

The NUMEN project: NUclear Matrix Elements for Neutrinoless double beta decay. EUROPEAN PHYSICAL JOURNAL A. , v.54, p.1/54 - 46, 2018.

NUMEN TDR review article
International Journal of Modern Physics A
Vol. 36, No. 30 (2021) 2130018

MOTIVATION

- NUCLEAR REACTIONS PRODUCED IN SELECTED TARGETS BY HEAVY AND HIGHLY ENERGETIC IONS;
- THE LARGE AMOUNT OF HEAT PRODUCED IN NUCLEAR REACTIONS MUST SOMEHOW BE DISSIPATED;
- THE SUBSTRATE FOR THE TARGET MUST HAVE EXCELLENT THERMAL CONDUCTIVITY;

Ion beams will have energy range 15-60 MeV/u and intensity up to $I_{beam} \approx 20 - 30\ \mu A$

The NUMEN experiment plans to use mainly beams of Carbon, Oxygen and Neon with intensity up to 10^{13} pps.

FOR THIS, THE ACCELERATOR IS BEING UPDATED.

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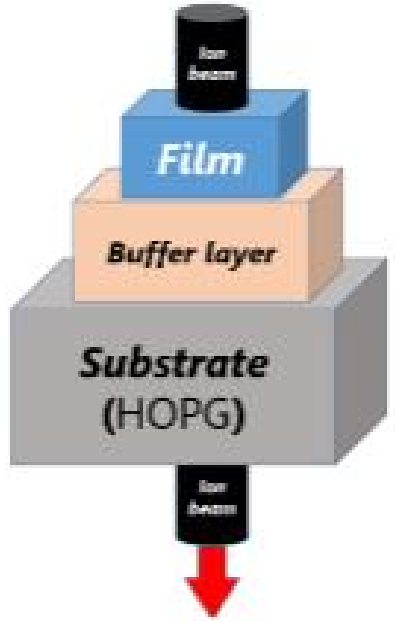
The accelerator physics community and many other areas are researching materials that can be used as heat sinks.

NUMEN

Several targets of interest in this project are metallic elements with a low melting point.

HOPG (HIGH ORIENTED PYROLYTIC GRAPHITE) as a substrate for the targets may be essential to avoid their melting.



THE SELECTED MATERIAL

HOPG – ***H****ighly* ***O****riented* ***P****yrolytic* ***G****raphite*

- **VACANCIES PRODUCED DURING IRRADIATION CONSIDERABLY DECREASE THERMAL CONDUCTIVITY;**
- **THE THERMAL CONDUCTIVITY OF THIS MATERIAL IS DUE TO MAINS PHONONS.**

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APPLIED NUCLEAR PHYSICS + SOLID STATE PHYSICS

+ INSTRUMENTATION + MATERIALS SCIENCE

SOLVING COMPLEX PROBLEMS

SCIENTIFIC KNOWLEDGE

IDEAS

STUDY OF THE EFFECTS CAUSED BY IONIZING RADIATION ON THE TARGET, PARTICULARLY the DEGRADATION IN THE HIGH ORIENTED PYROLYTIC GRAPHITE (HOPG)

Characterization of thin **HOPG targets; t**his study proposes the characterization of samples of Highly Oriented Pyrolytic Graphite (**HOPG**), observing the possible variations in their structural properties, which can interfere in chemical-physical properties after being exposed to different radiation sources ionizing.

simulations of the interaction of different particle beams and composite targets, according to the reactions of interest for carrying

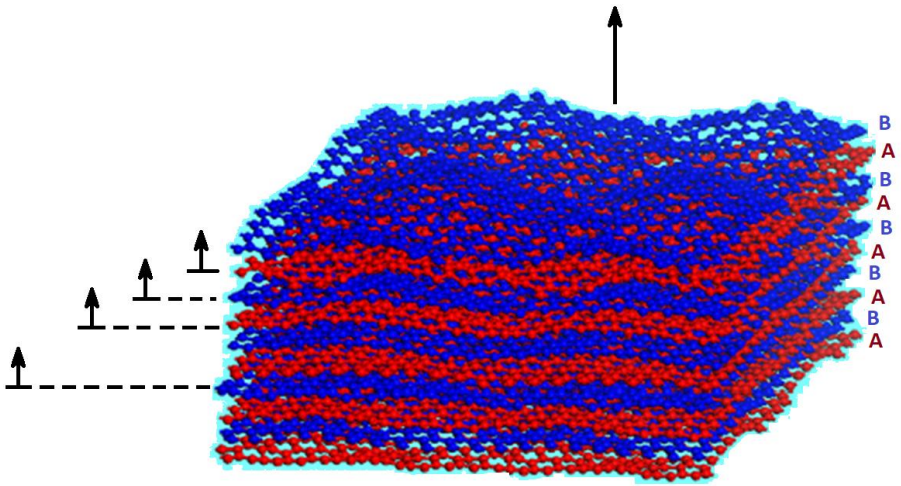
experiments that simulate the damage to HOPG generated in nuclear reactions.

WHAT IS HOPG?

CONSISTING ONLY OF CARBON ATOMS THAT REMAIN GEOMETRICALLY ARRANGED IN A HEXAGONAL SHAPE, HIGHLY ORIENTED PYROLYTIC GRAPHITE (HOPG) BELONGS TO **LAMELLAR MATERIALS.**

WITH ATOMS ARRANGED IN PARALLEL AND STACKED LAYERS, IT HAS A CRYSTALLINE STRUCTURE AND IS CHARACTERIZED BY ITS HIGH DEGREE OF THREE-DIMENSIONAL ORDERING (SIMILAR TO THIS ONE, BUT ORGANIZED IN TWO-DIMENSIONAL FORM, IT IS CALLED GRAPHENE), IN OTHER WORDS, THE

HOPG IS A SUCCESSION OF STACKED LAYERS OF GRAPHENE.



graphene foils, one on top of the other, forming the HOPG structure

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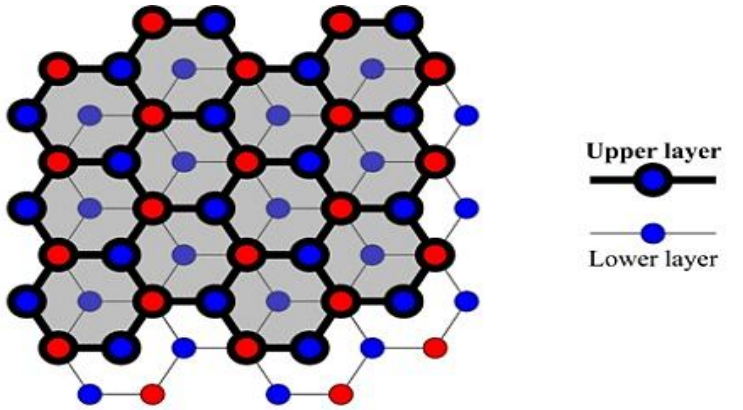
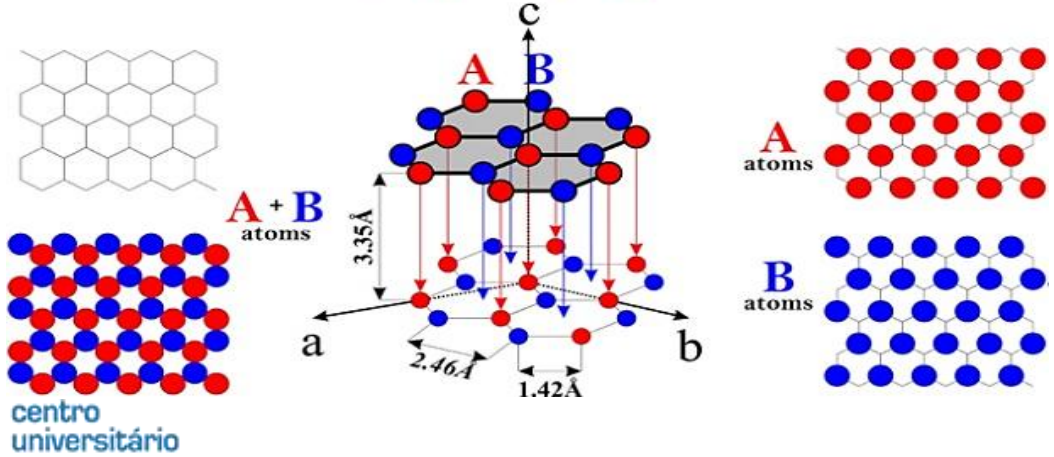
For **the HOPG** to act with thermal energy dissipation functionality during the experiments proposed by the NUMEN project, **IT IS NECESSARY TO VERIFY WHETHER POSSIBLE CHANGES CAUSED BY EXPOSURE TO THE RADIATION BEAM HAVE A DIRECT OR INDIRECT INFLUENCE ON ITS THERMAL PROPERTIES.**





Physical properties of HOPG are characteristic of its crystal lattice structure and it presents different results when measured along the basal plane (A-B) and along the main axis (c) (perpendicular to the basal plane).

THE MATERIAL IS HIGHLY STABLE AT TEMPERATURES FROM 500°C TO TO 3000 °C DEGREES CELSIUS IN A VACUUM OR INERT ENVIRONMENT.

CASE STUDY - HOPG: 2 μm thick **FROM OPTIGRAPH**.

Density: 2.2 g/cm^3

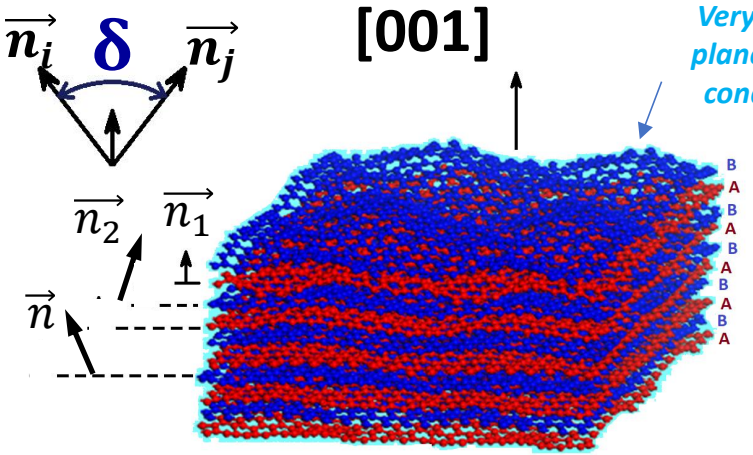
Thermal conductivity: **κ = 1700 W/m*K in plane**

Interlayer distance: 3.35 Å

distance between atoms (in the hexagonal plane):0.1415 nm

Highly Oriented Pyrolytic Graphite

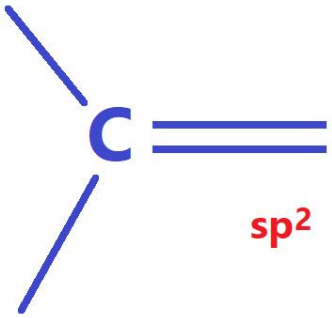
HOPG is a stacking of graphene foils with a slight disorientation between each other



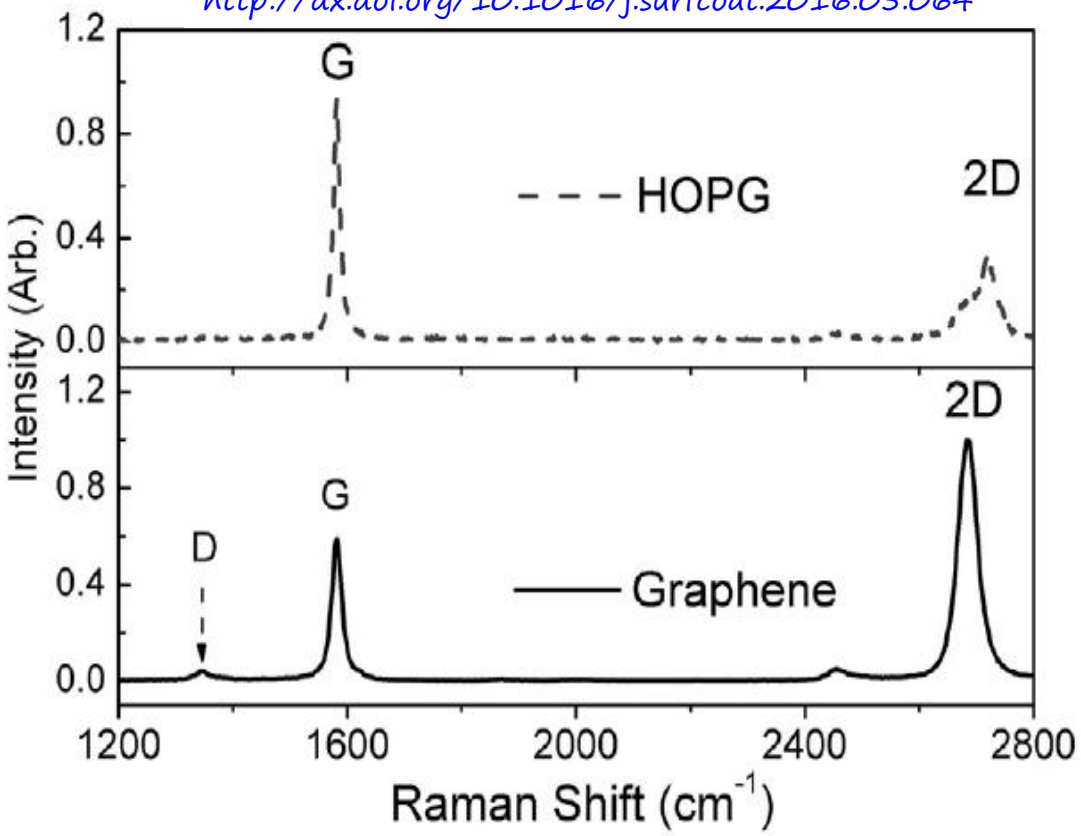
Grade*	Maximun Disorientantion (δ)	
ZYA	$(0.4 \pm 0.1)^o$	Used in the NUMEN project
ZYB	$(0.8 \pm 0.2)^o$	
ZYD	$(1.2 \pm 0.2)^o$	
ZYH	$(3.5 \pm 1.5)^o$	

Zhikun Zhang et. al., ***Materials Letters*** **115** (2014), 261-264

RAMAN *for HOPG and Graphene: the differences in intensities is due to the number of graphene sheets in the HOPG*



H. Peng, et al., *Surf. Coat. Technol.* (
http://dx.doi.org/10.1016/j.surfcoat.2016.05.064



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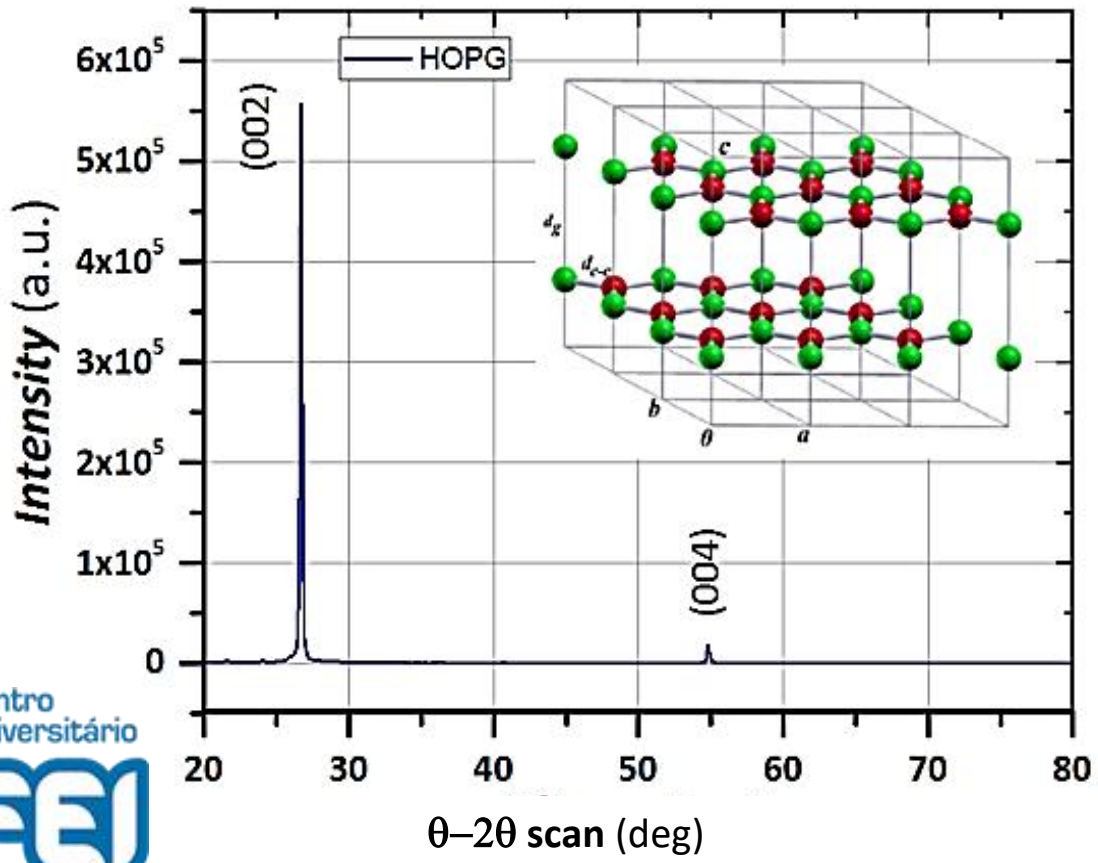
The ideal condition would be for the graphene layers to have the same normal vector

Characterization

- **X-ray diffraction** → FEI



X-ray diffraction *for HOPG used as substrate for targets in the NUMEN project*

High degree of purity and high degree of ordering of atoms in the crystal lattice of pyrolytic graphite. After irradiating: possible changes are expected, justifying the changes in physical properties.

X-ray diffraction can indicate changes in the crystal lattice, for example:

i) change of the position of diffraction peaks and

ii) change in the width of these peaks

iii) Change in intensity

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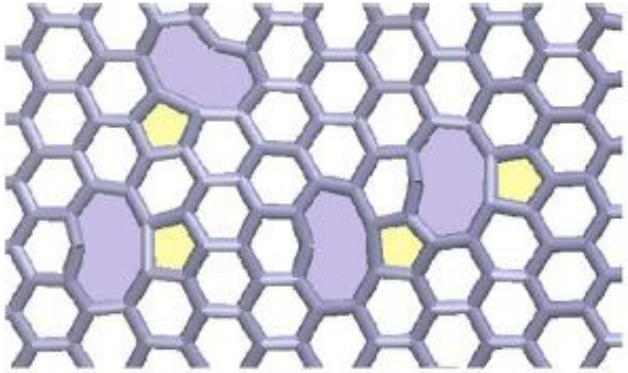
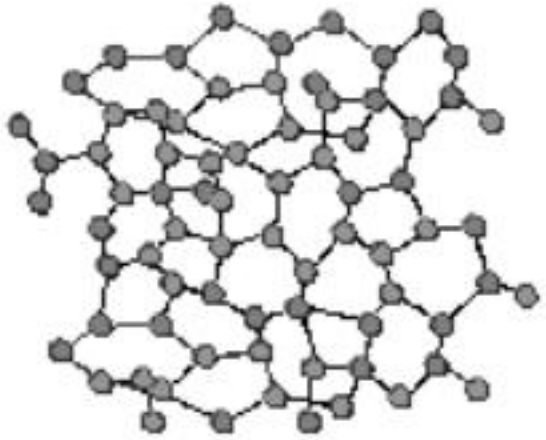
DIAGRAM BASED ON AVERAGE VALUES REPORTED IN LITERATURE. THERMAL CONDUCTIVITY OF BULK CARBON ALLOTROPES AS A FUNCTION OF *T*.

The *K* value for pyrolytic graphite constitutes the bulk graphite limit of **~2000 W/mK at RT**.

At low *T*, ***K* is proportional to T^{γ} where γ varies in a wide range depending on graphite's quality.**



vacancy



amorphous carbon

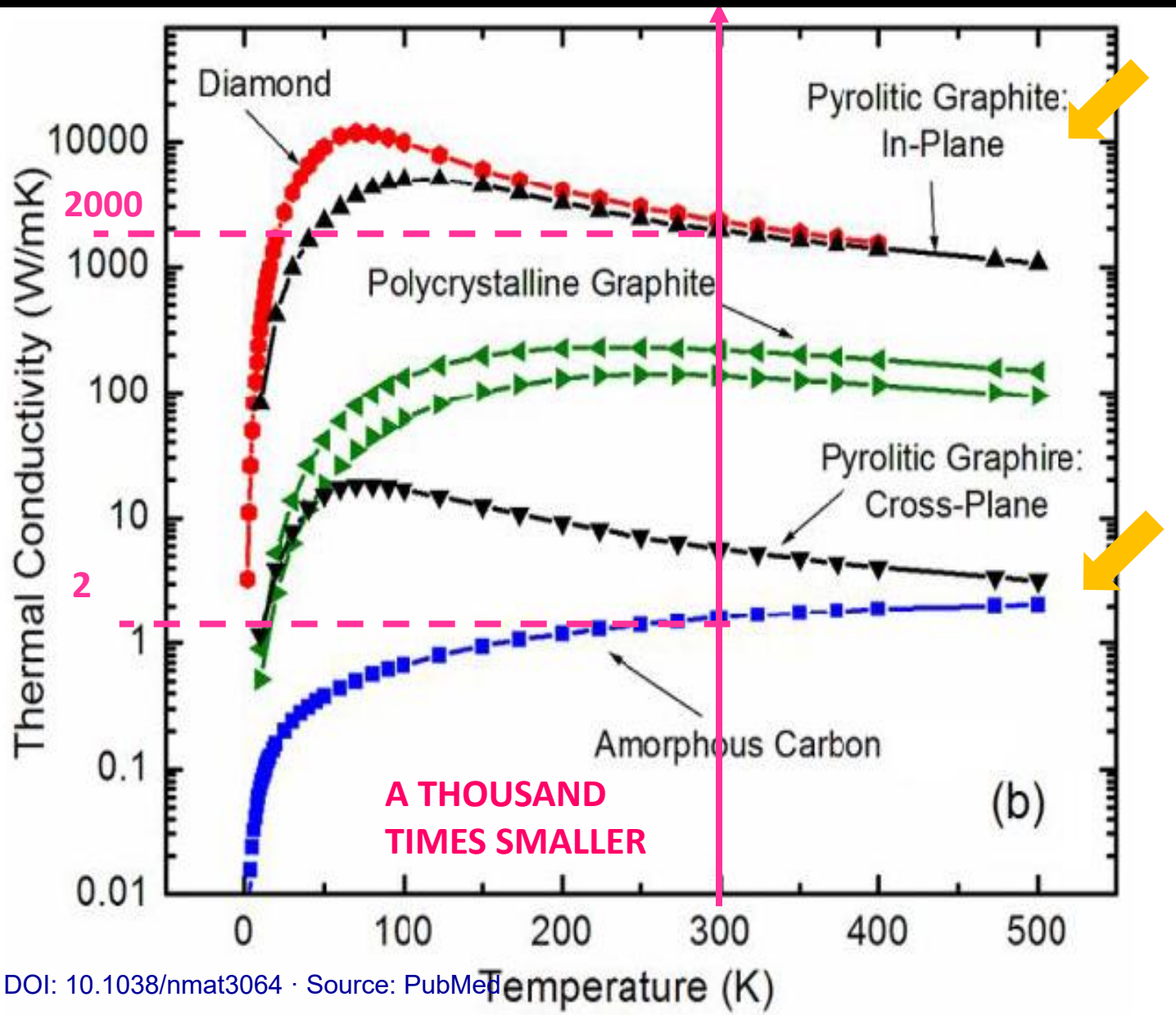
K, THERMAL CONDUCTIVITY of the different allotropic forms of carbon is due to **phonons of the lattice.**

K: THERMAL CONDUCTIVITY

DIAGRAM BASED ON AVERAGE VALUES REPORTED IN LITERATURE.

THERMAL CONDUCTIVITY OF BULK CARBON ALLOTROPES AS A FUNCTION OF *T*.

- diamond (sp^3)
- amorphous carbon (disordered mixture of sp^2/sp^3)



Article in Nature Materials · July 2011 DOI: 10.1038/nmat3064 · Source: PubMed

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Thermal conductivity and tensile response of defective graphene: A molecular dynamics study

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[b] *ICube laboratory, University of Strasbourg/CNRS, 2 Rue Boussingault, 67000 Strasbourg, France*
[c] *Georgia Institute of Technology, School of Materials Science & Engineering, Atlanta, GA 30332, USA*

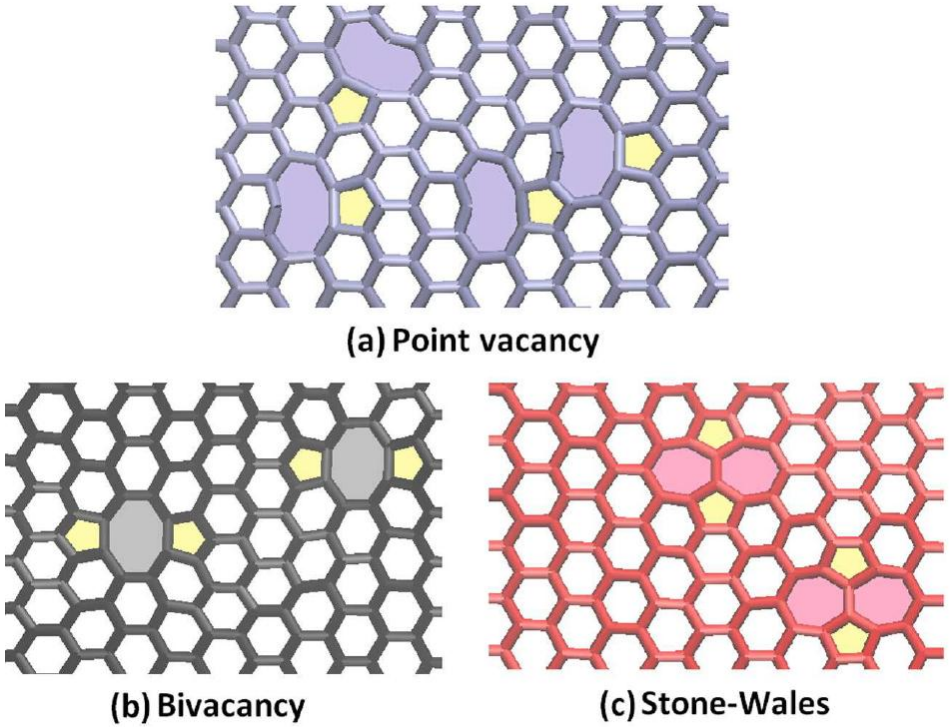


Fig. 1 – Different types of defects in equilibrated graphene sheets.

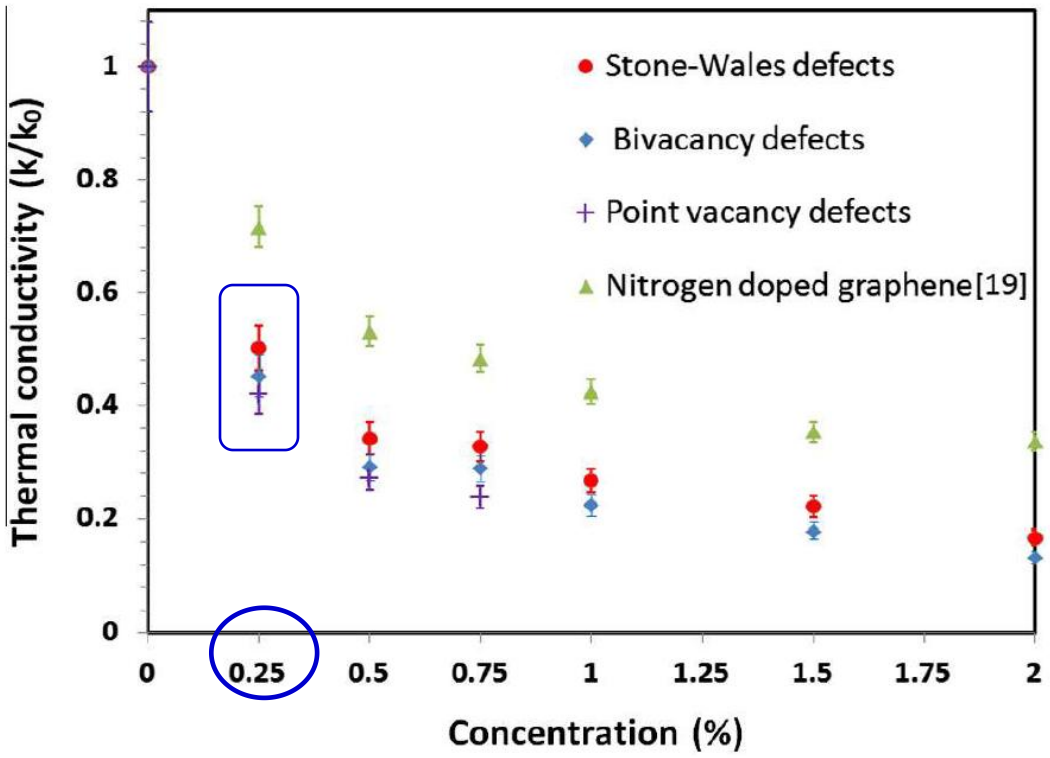


Fig. 8 – Normalized thermal conductivity of graphene as a function of defects concentrations.

– We found exponential trends in reduction of graphene thermal conductivity as results of defects concentrations. **Our results suggest that only 0.25% concentration of defects in graphene result in significant reduction of thermal conduction property of graphene by around 50%.** We also found that the effects of studied defects on the thermal conductivity of graphene are considerably close to each other.

FEW STRUCTURAL DEFECTS IN HIGHLY ORDERED MATERIAL CAN GREATLY COMPROMISE ITS FUNCTIONALITY

Linear Energy Transfer to target
Vacancies formed by Ion beam
Vacancies formed by target ions, scattered by Ion beam, in HOPG
Total Single Vacancies
Dose rate and total dose accumulated in target – related with Total Energy

- **SRIM simulations** → vacancy formation processes in target and HOPG substrate
- **Irradiation** → several ions and doses

Ziegler & Biersack

Transport of Ions in Matter - TRIM

is part of a program group which calculates the stopping and range of ions into matter using a quantum mechanical treatment of ion-atom collisions (assuming a moving atom as an ***"ion"***, and all target atoms as ***"atoms"***): **The Stopping and Range of Ions in Matter – SRIM.**

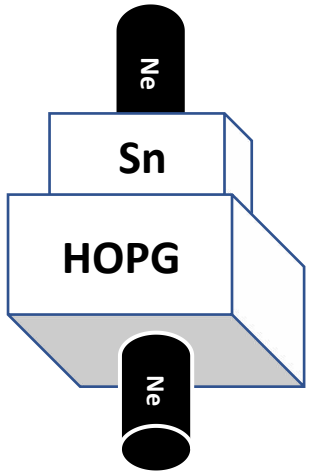
TRIM - accept complex targets made of compound materials
It will calculate both the final 3D distribution of the ions and also all kinetic phenomena associated with the ion's energy loss: target damage, sputtering, ionization, and phonon production.

SRIM results from the original work by J. P. Biersack on range algorithms (see J. P. Biersack and L. Haggmark, Nucl. Instr. and Meth., vol. 174, 257, 1980) and the work by J. F. Ziegler on stopping theory (see "***The Stopping and Range of Ions in Matter***", *volumes 2 - 6*, Pergamon Press, 1977-1985).

Total vacancies on HOPG *x* Irradiation time (*for 3 possibles ions fluxes*)

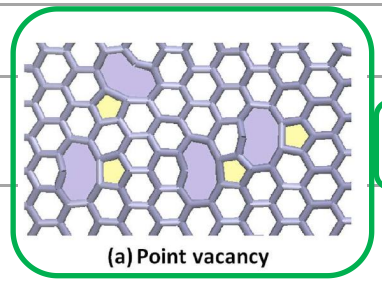
^{20}Ne Beam parameters

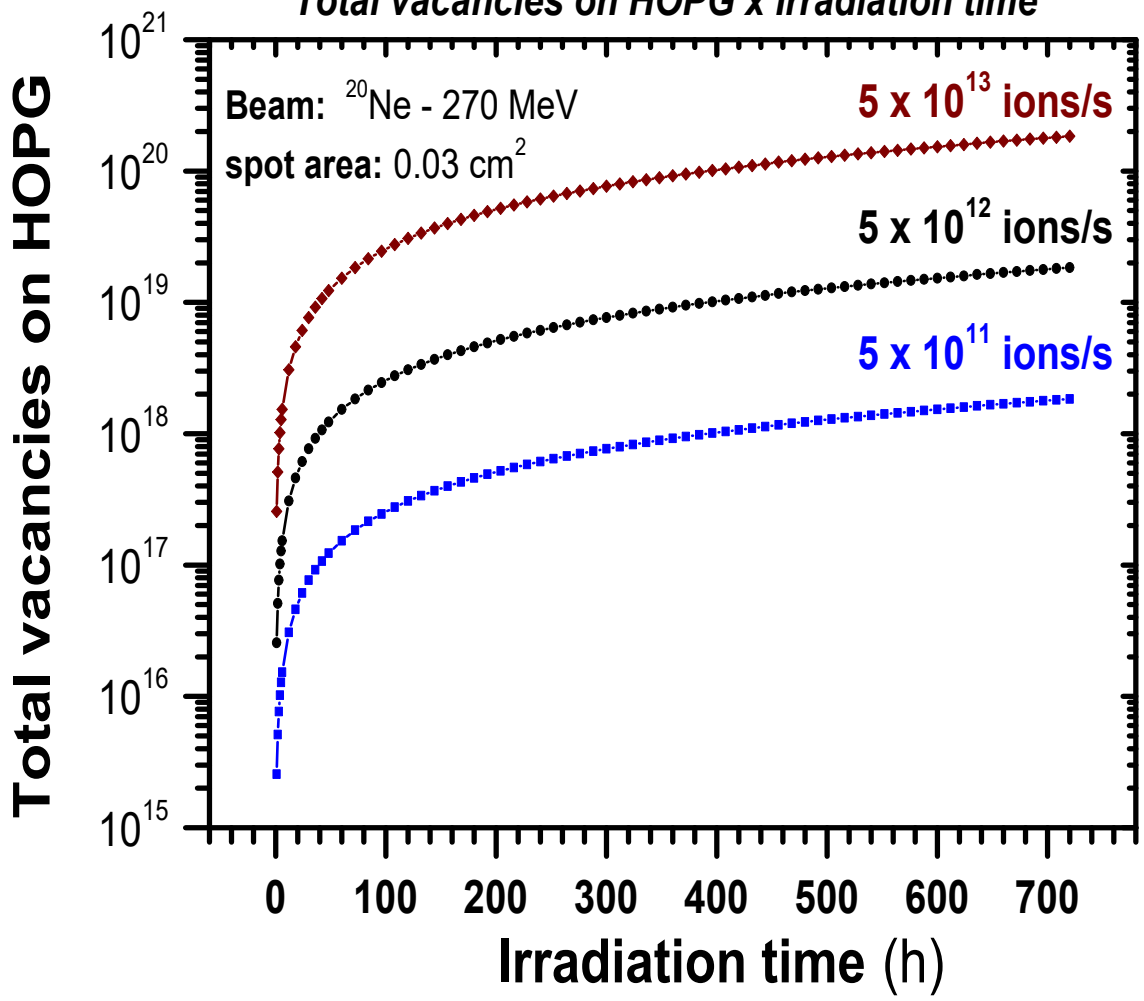
Energy (MeV)	270
Spot area (cm²)	0.03
Current (μA)	*variable*
Charge state	10
Flux (ions/s)	*variable*
Normal incidence	



Target parameters

	Sn film	buffer	HOPG substrate
Thickness (μm)	0.4	-	2.635
Density (g/cm³)	7.3	-	2.22
a.m.u.	118.7		12
Surface binding energy (eV)	3.12		7.41
Lattice binding energy (eV)	3.0		3.0
Irradiated volume (cm³)	$1.2 \cdot 10^{-6}$	-	$7,905 \cdot 10^{-6}$

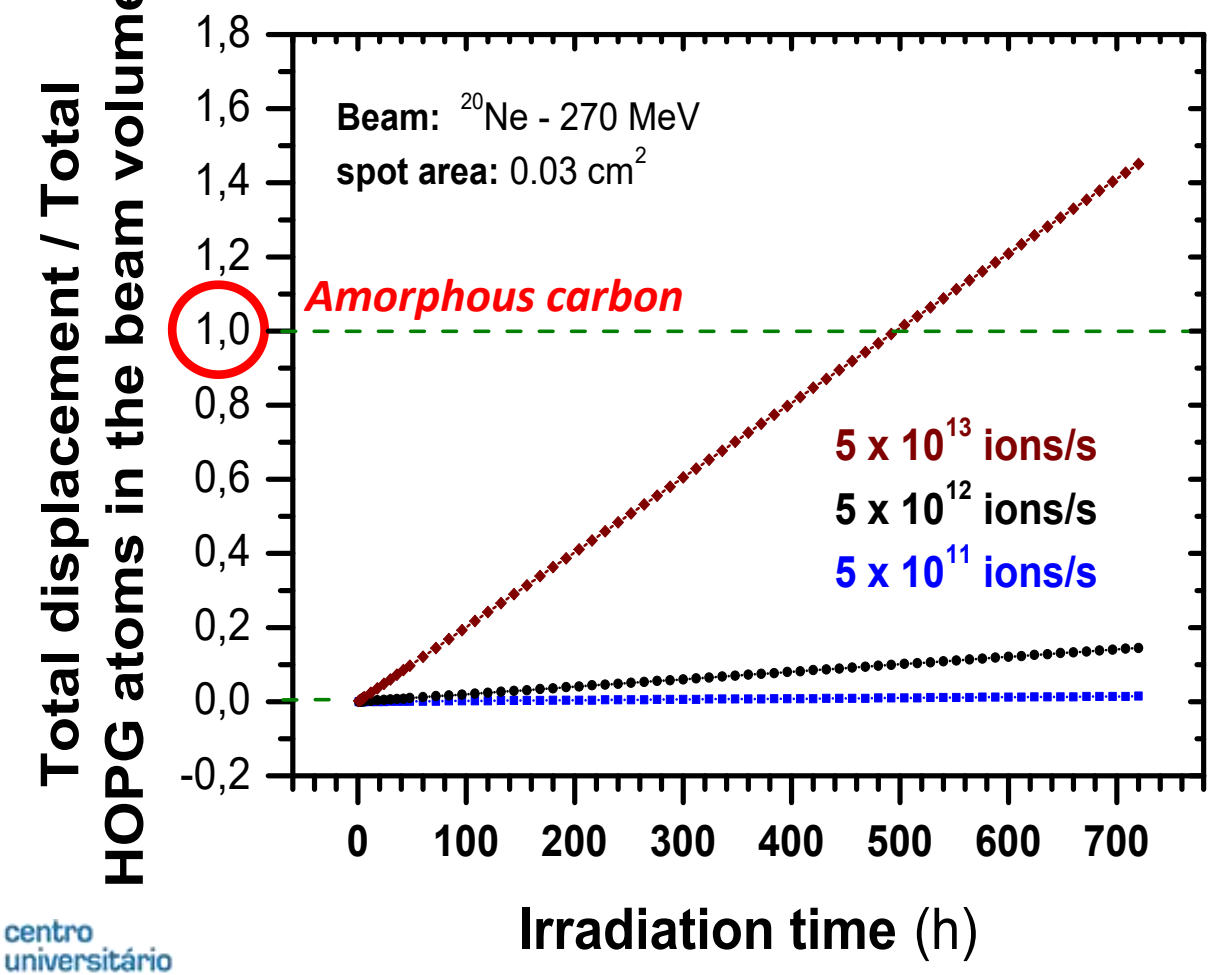




JOURNAL OF PHYSICS. CONFERENCE SERIES, v.2340, p.012002, 2022.Using TRIM-SRIM code simulations to determine defect density produced in HOPG irradiated with high energy heavy ions

Defect concentration on HOPG *x* Irradiation time (*for 3 possibles ions fluxes*)

Transport of Ions in Matter - TRIM

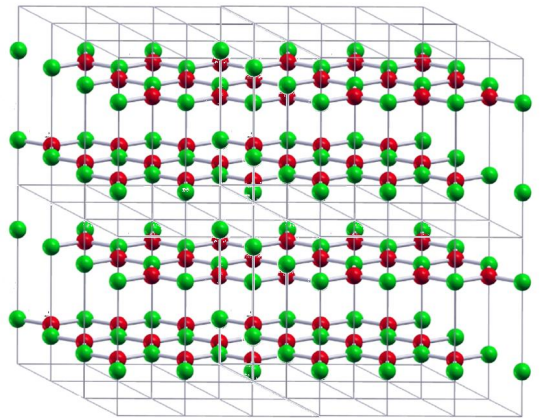


When deffect concentration = 1
All the HOPG in the beam volume has become amorphous carbon

Degradation in the HOPG crystallographyc lattice perfection due to irradiation

Thermal conductivity changing from:

HOPG – *2000 W/m.K*
to
amorphous C – *2 W/m.K*



HOPG

Before irradiation
HOPG is a stacking of graphene sheets

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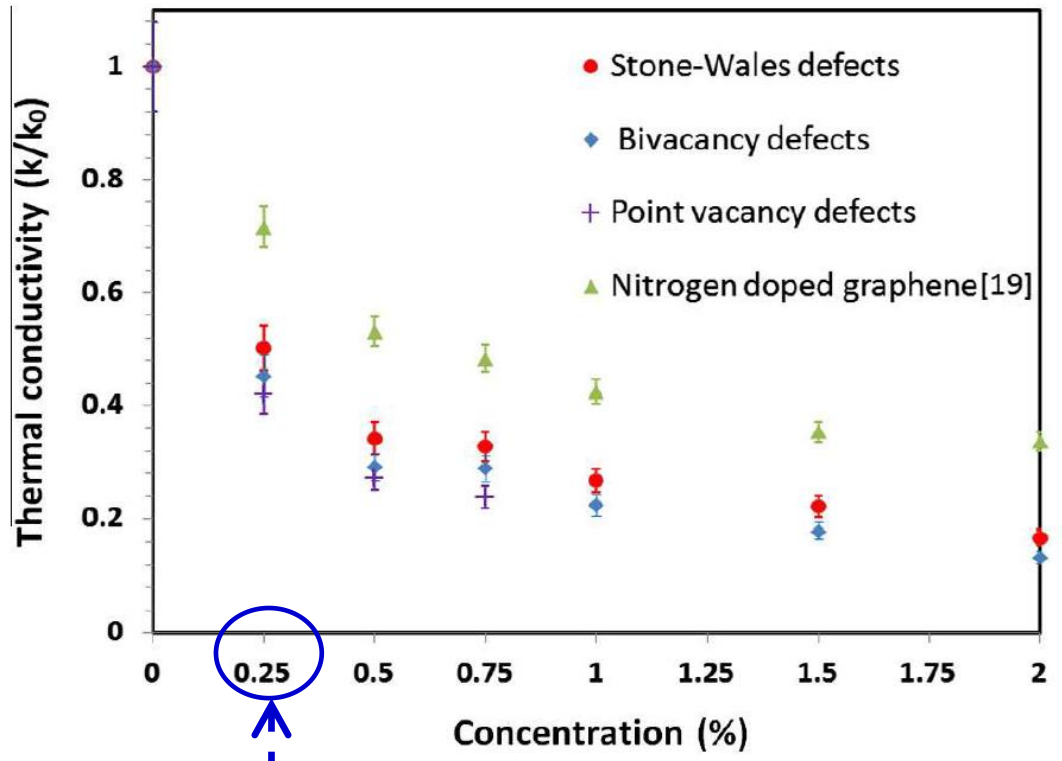


Fig. 8 – Normalized thermal conductivity of graphene as a function of defects concentrations.

– We found exponential trends in reduction of graphene thermal conductivity as results of defects concentrations. **Our results suggest that only 0.25% concentration of defects in graphene result in significant reduction of thermal conduction property of graphene by around 50%.** We also found that the effects of studied defects on the thermal conductivity of graphene are considerably close to each other.

~3.5 hours

In this case, through the simulation, in just 3.5 hours we would have 0.25% damage to the crystalline structure, reducing conductivity by 50%.

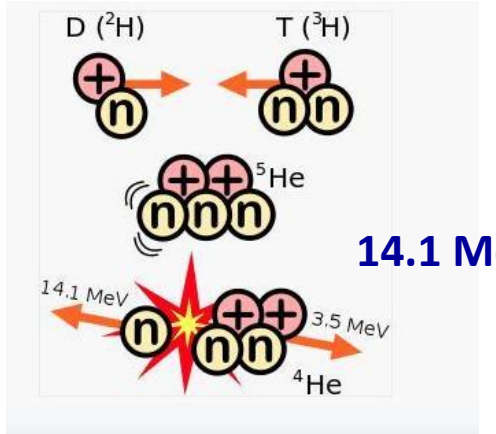
THE EXPERIMENT

NEUTRONS

- Neutrons cause damage to the crystalline structure
- Nuclear reactions generate neutrons

WE MUST REMEMBER THAT THE DAMAGE IS NOT CAUSED BY NEUTRONS, BUT BY THE PRODUCT OF THE NEUTRON REACTION IN THE HOPG

NEUTRON IS ALSO A CONCERN FOR NUCLEAR REACTION EXPERIMENTS



IN THIS EXPERIMENT WE USE A MONOENERGETIC BEAM FROM A GENERATOR Neutrons are produced by Deuterium–Tritium (DT) reactions obtained from deuterium ions accelerated and impinging on a tritiated titanium target

Available

Beam – target

HOPG *- NUMEN*

.

Neutron beam:

we irradiated the sample with a neutron beam
to verify the properties of HOPG
before and after irradiation damage

Energy: $14\ MeV$

Characterization

i. X-ray diffraction (θ-2θ scan)

ii. Raman "mapping"

iii. AFM

iv. SEM

v. Thermal conductivity

vi. Electrical Resistivity

vii. and Magnetoresistance

NEUTRON GENERATOR

IEAv – DT NEUTRON GENERATOR

Technical informations:

Pressure: 120 PSIG / 6200 Torr (99.995% SF_6)

Neutron yield: ~ 1×10^8 n/s

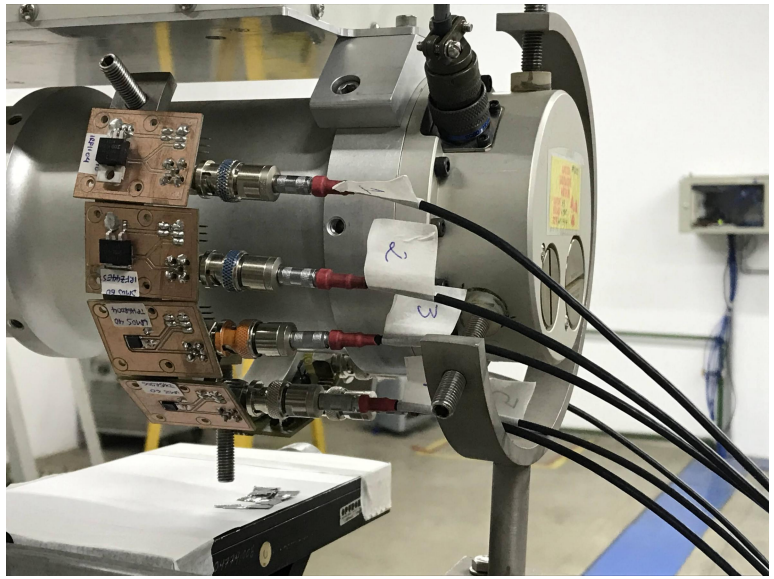
Neutron Energy: ~ 14 MeV

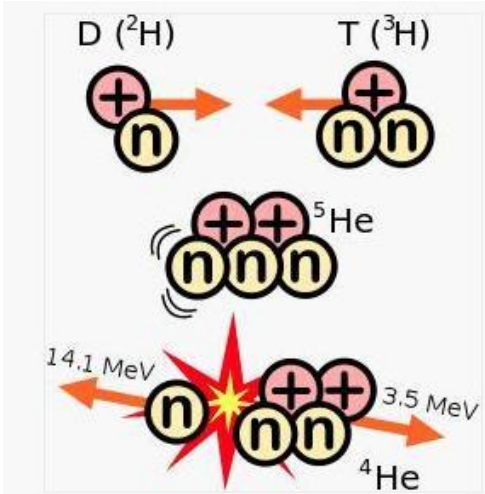
Frequency: 250 – 20 kHz

DD: $^2H + ^2H \rightarrow ^3He + n + 3.266$ MeV

DT: $^2H + ^3H \rightarrow ^4He + n + 17.586$ MeV

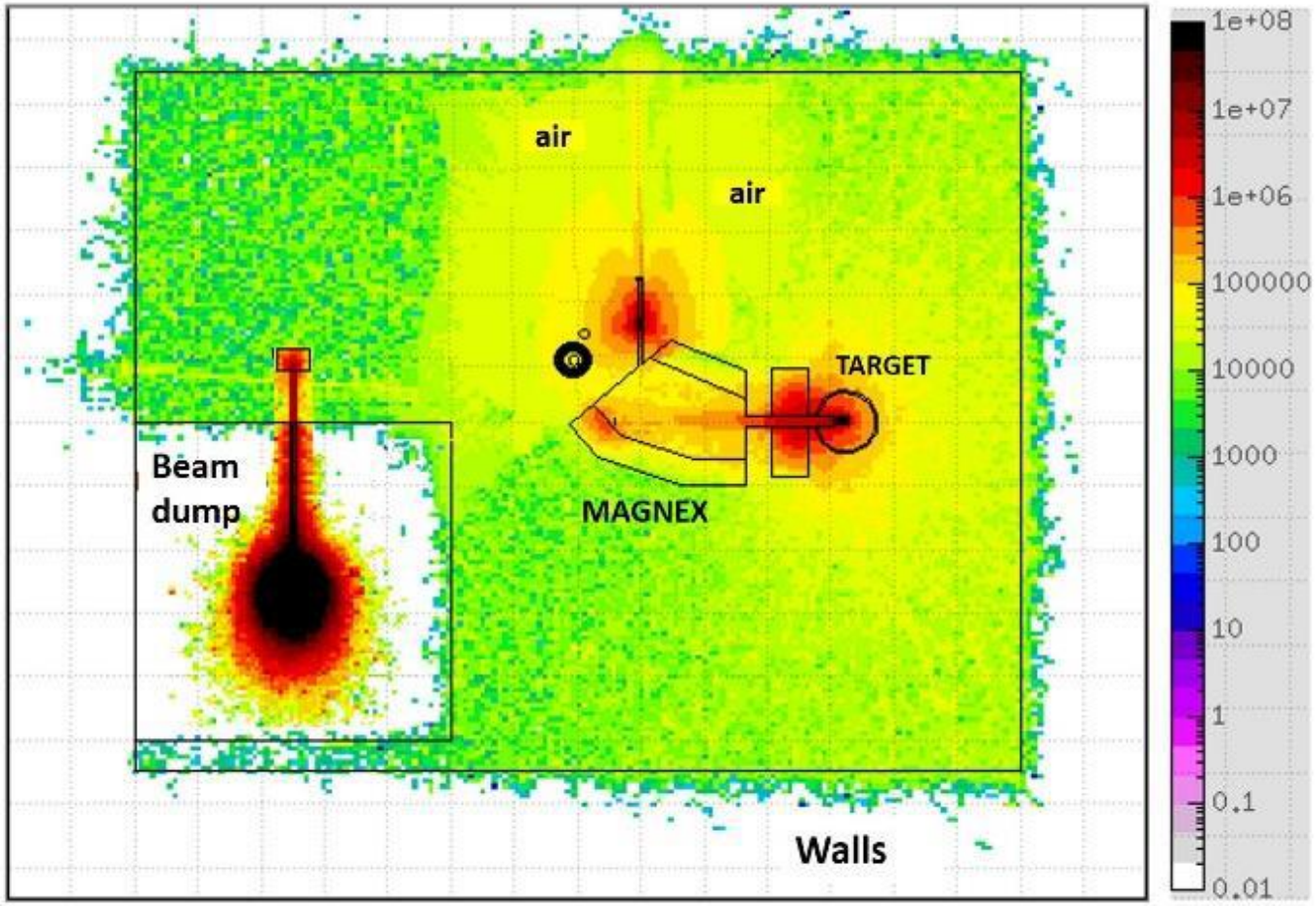
Neutron Energy: ~ 14 MeV





14.1 MeV

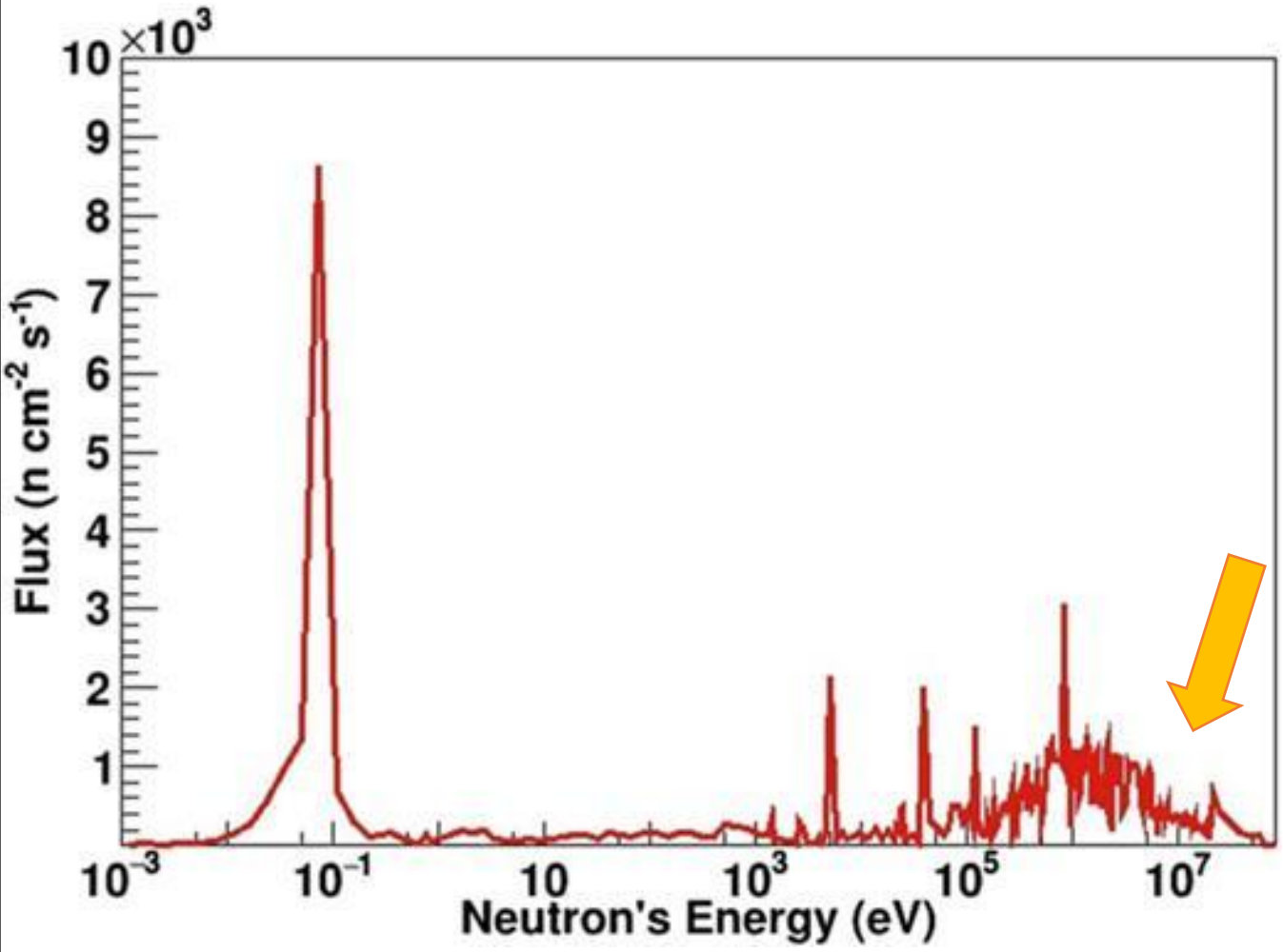
Sectioned accelerator axial DD NG, with three stage accelerator section, puller electrode for the initial beam extraction and acceleration and the RF induction ion source with an external.



FLUKA simulations of the neutron flux in MAGNEX experimental hall. The target chamber is placed just before the MAGNEX quadrupole. The region where the focal plane detector is located is denoted with an open black circle.

The ^{20}Ne beam interaction with a ^{76}Ge target with ^{12}C backing was simulated and the analysis of the distribution of secondary particles was made to derive the dose, energy spectra, and the flux of neutrons in the experimental hall.

A ^{20}Ne beam with an energy of 60 MeV/A and a current of 85 eμA was considered. The highest neutron flux inside the experimental hall is expected near the focal plane detector region. The estimated neutron flux near the focal plane detector is 1.3×10^4 thermal neutrons/cm²/s (En < 0.1 eV), 1.7×10^4 epithermal neutrons/cm²/s (0.1 eV < En < 100 keV), **and 9.0×10^4 fast neutrons/cm²/s (En > 100 keV),** totaling approximately 1.2×10^5 neutrons/cm²/s.



Neutron energy spectrum obtained via FLUKA simulations for the region where the MAGNEX focal plane detector is located. MAGNEX is the dipole that deflects the beam

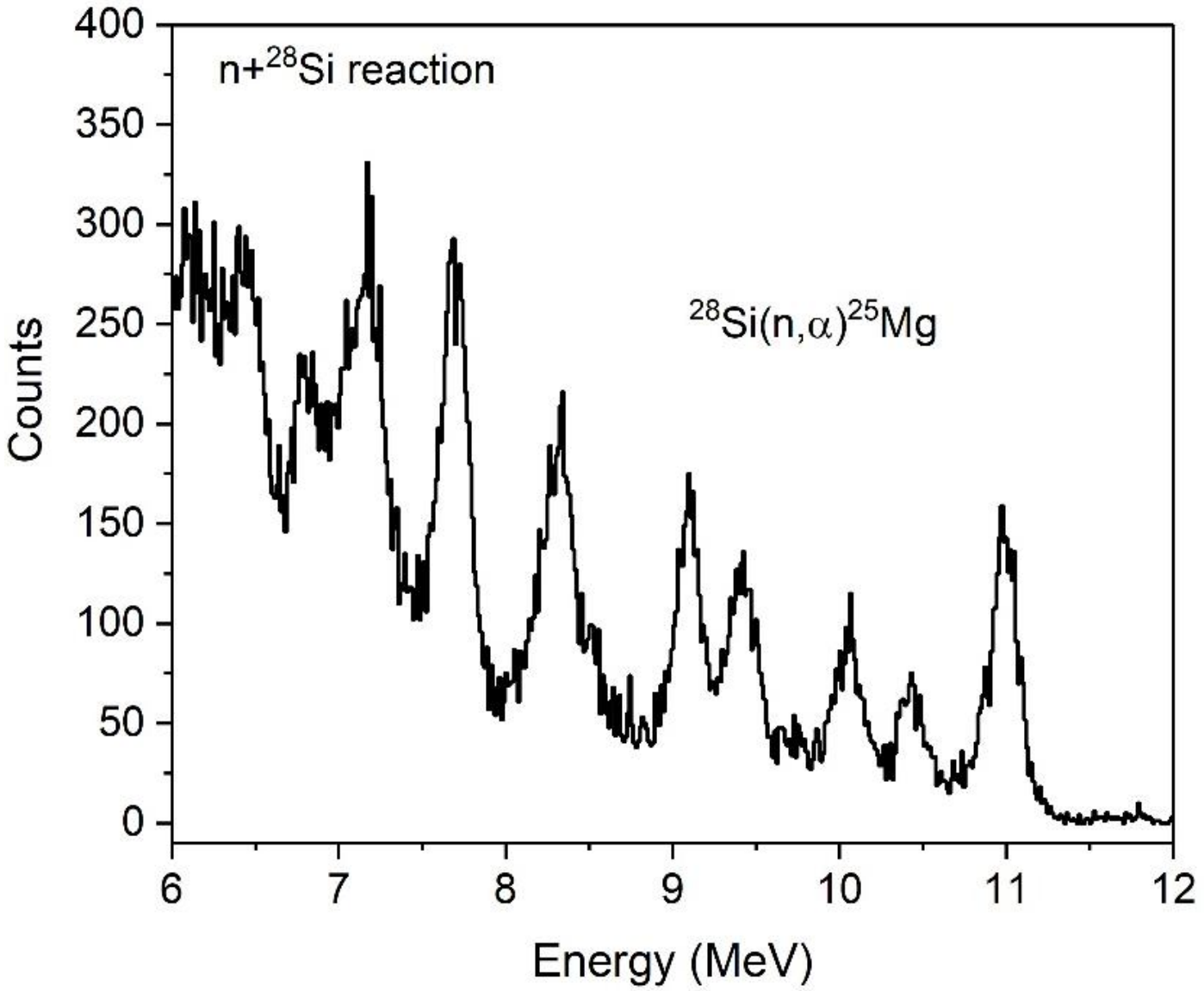
It is important to highlight that during the NUMEN experiments, an estimated neutron production of 10^5 neutrons/cm^2/s is expected, with the total spectrum consisting of thermal and epithermal neutrons, ranging from 10^{-3} eV to 10^8 eV.

In this study, monochromatic neutrons of 1.4 x 10^7 eV were used, which may be representative of a portion of neutrons produced in NUMEN.

Rev. Sci. Instrum. 91, 083301 (2020)
https://doi.org/10.1063/5.0010968

To monitor the reaction, a 100 µm-thick fully depleted Si surface barrier, which detected the 28Si(n, α)25Mg, and 28Si(n, p)28Al nuclear reactions, was used. In Figure the Response spectrum for Si surface barrier detector exposed to 14 MeV neutrons is shown.

Through the detection of **α particles and 25Mg nuclei** in the silicon (Si) detector, we can ensure that by-products were generated resulting from the interaction of neutrons also with 12C nuclei (HOPG).



WE VERIFIED IN THE PACE4 (fusion evaporation program) PROGRAM THE PRODUCTS OF THE NEUTRON REACTIONS OF 14.1 MEV IN CARBON

→ statistical model evaporation code that employs Monte Carlo simulation for the de-excitation of the compound nucleus

Starting conditions

	Z	N	A	Spin
Projectile	0	1	1	0.5
Target	6	6	12	0
Compound nucleus	6	7	13	

1. Yields of residual nuclei

Yelds of residual nuclei from the n + ^{12}C reaction at 14 MeV

Z	N	A	events	percent	x-section(mb)	
6	7	13 C	13	1.3%	5.39	
6	6	12 C	590	59%	245	neutron channel
4	5	9 Be	397	39.7%	165	Alpha channel
TOTAL			1000	100	414.468	

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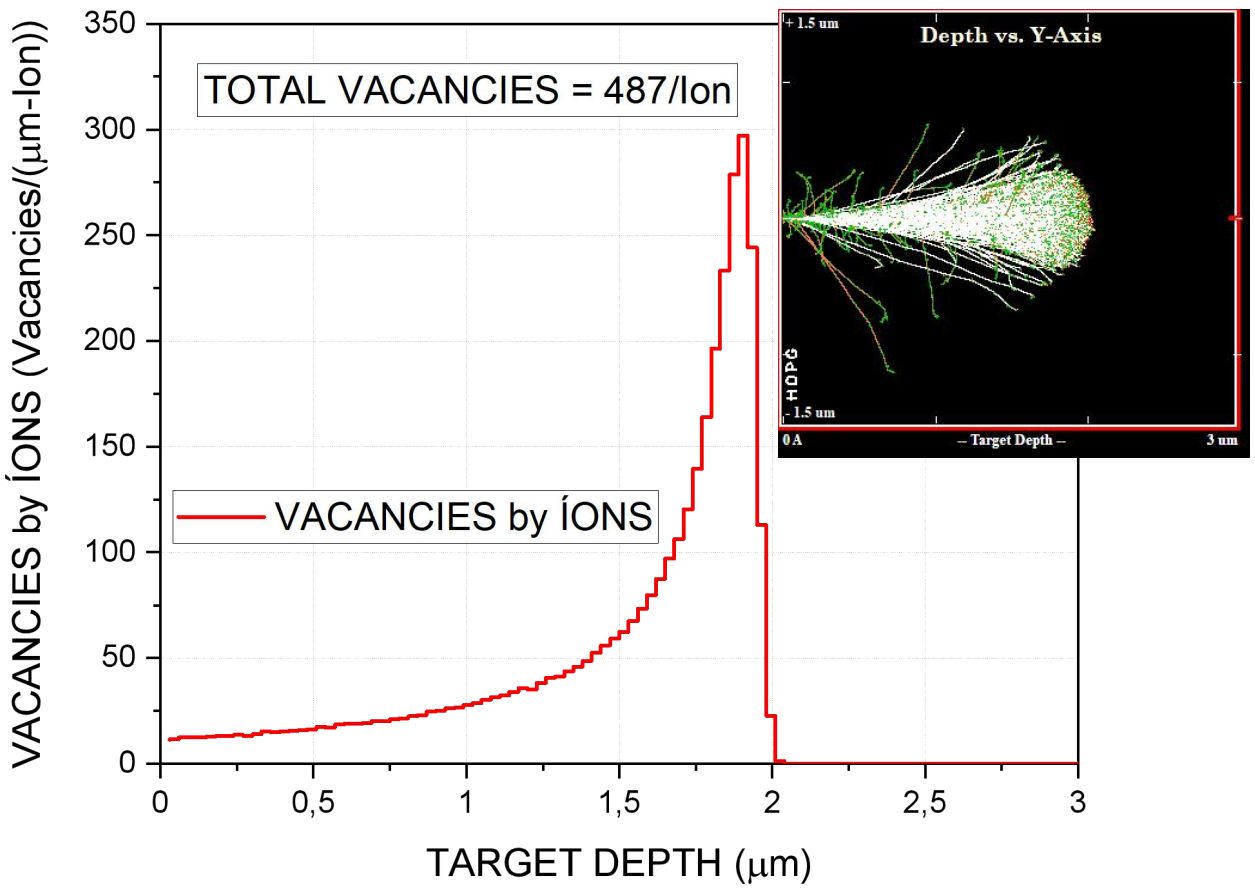
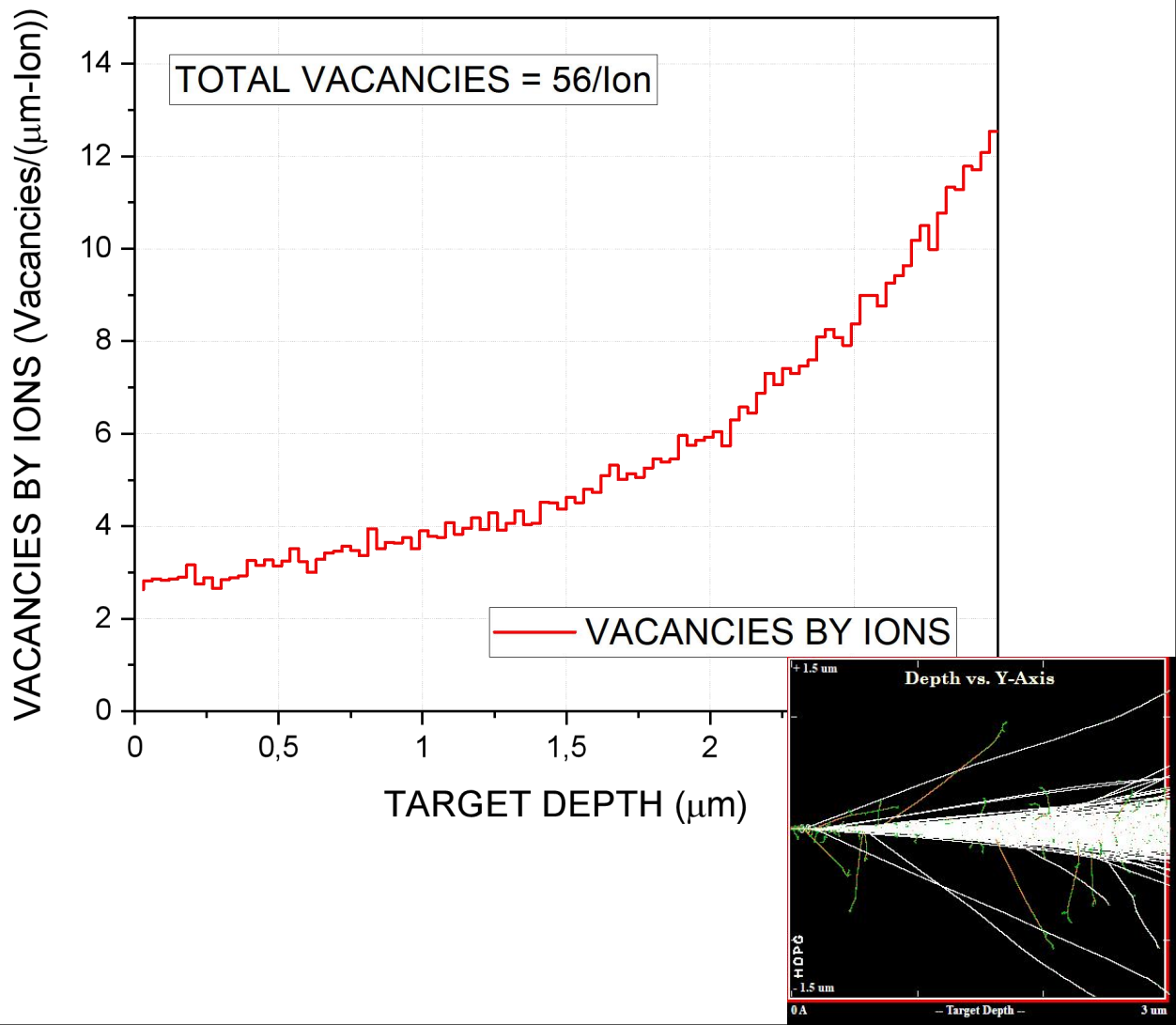
TRIM SIMULATIONS





sample	IRRADIATION	EXPOSURE TIME	FLUENCE (n/cm^2)	Max # of ^{12}C nuclei	Max # of ^{9}Be nuclei	Max # of vacancies/cm^2
1N	I	4.7 h	$1.84\,(4)\times10^{9}$	$\sim1\times10^{5}$	$\sim8\times10^{4}$	$\sim6\times10^{7}$
2N	II	10.48 h	$5.18\,(13)\times10^{9}$	$\sim4\times10^{5}$	$\sim2\times10^{5}$	$\sim2\times10^{8}$
3N	III	24.40 h	$1.27(2)\times10^{10}$	$\sim9\times10^{5}$	$\sim6\times10^{5}$	$\sim4\times10^{8}$
4N	IV	30.50 h	$1.70(3)\times10^{10}$	$\sim1\times10^{6}$	$\sim8\times10^{5}$	$\sim6\times10^{8}$

1 cm x 1 cm samples were irradiated for each irradiation time

IT IS EQUIVALENT TO 27 HOURS OF EXPOSURE TO FAST NEUTRONS GENERATED IN THE NUMEN EXPERIMENT.

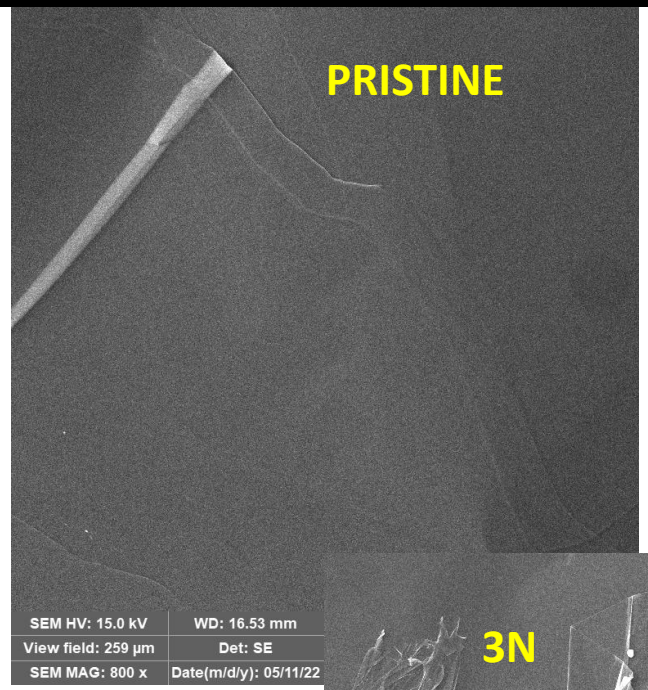
RAMAN, XRD, SEM, AFM, THERMAL CONDUCTIVITY, ELECTRICAL RESISTIVITY, AND MAGNETORESISTANCE.

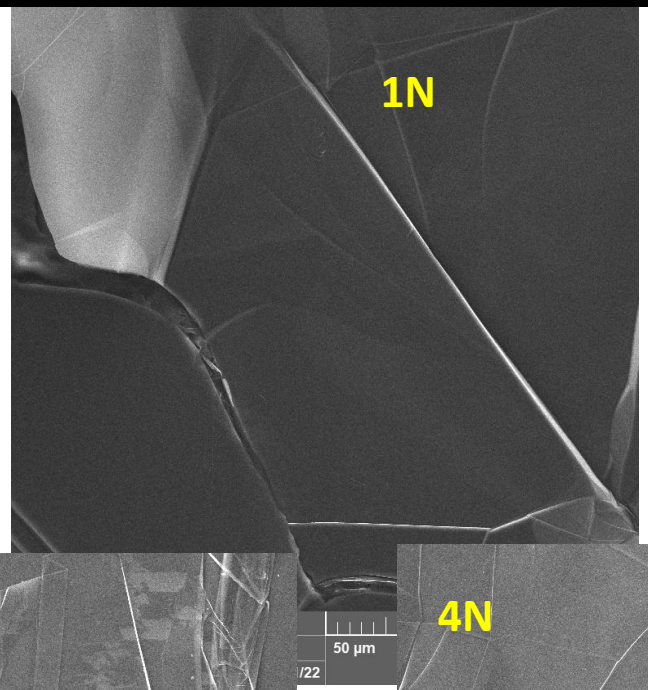
The microstructural characterizations were done with scanning electron microscopy (SEM) and atomic force microscopy (AFM).

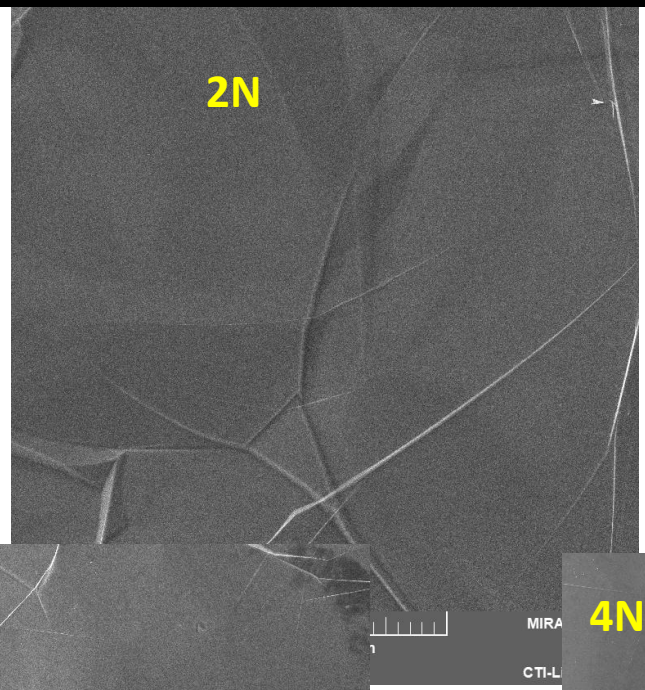
The characterization of the orientation and disorder of the HOPG crystal lattice of these three foils and a pristine foil were performed using Raman spectroscopy and X-ray Diffraction (XRD).

The effects of the loss of crystalline structure on the properties of HOPG were observed through Thermal Conductivity, Electrical Resistivity, and Magnetoresistance tests.

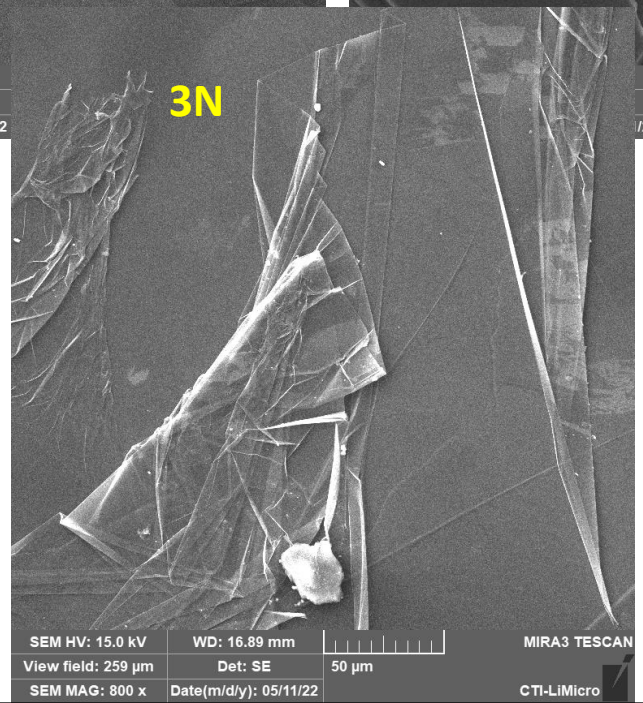
CHARACTERIZATION

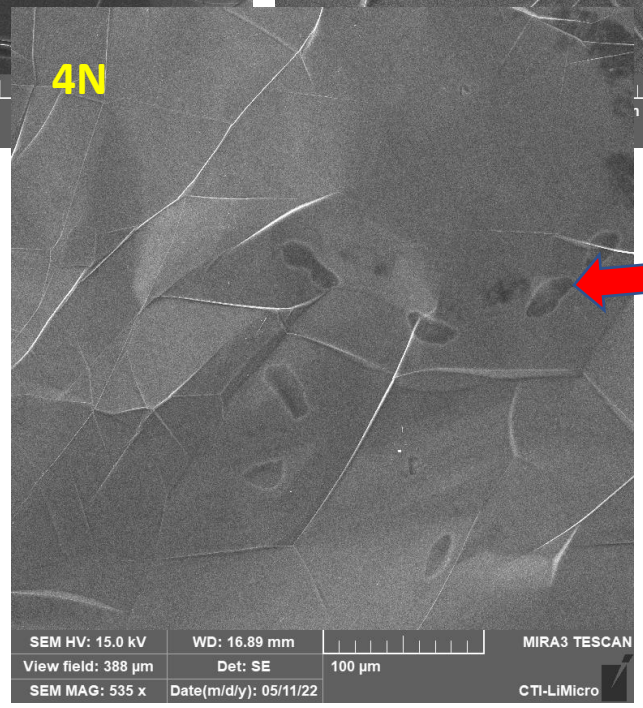


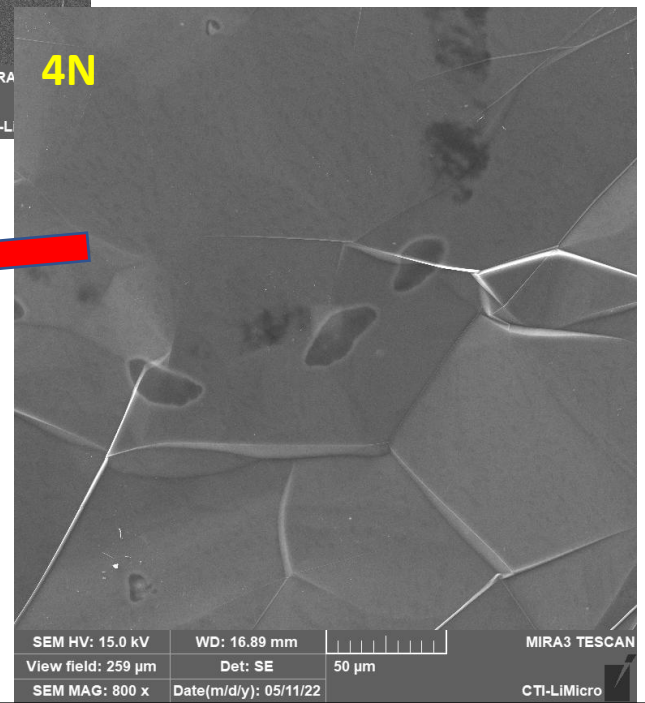




THE GRAPHENE FOILS APPEAR IN GREATER NUMBERS, AS IF THEY were PEELED OFF, AS AN EFFECT OF THE LAMELLAR STRUCTURE.



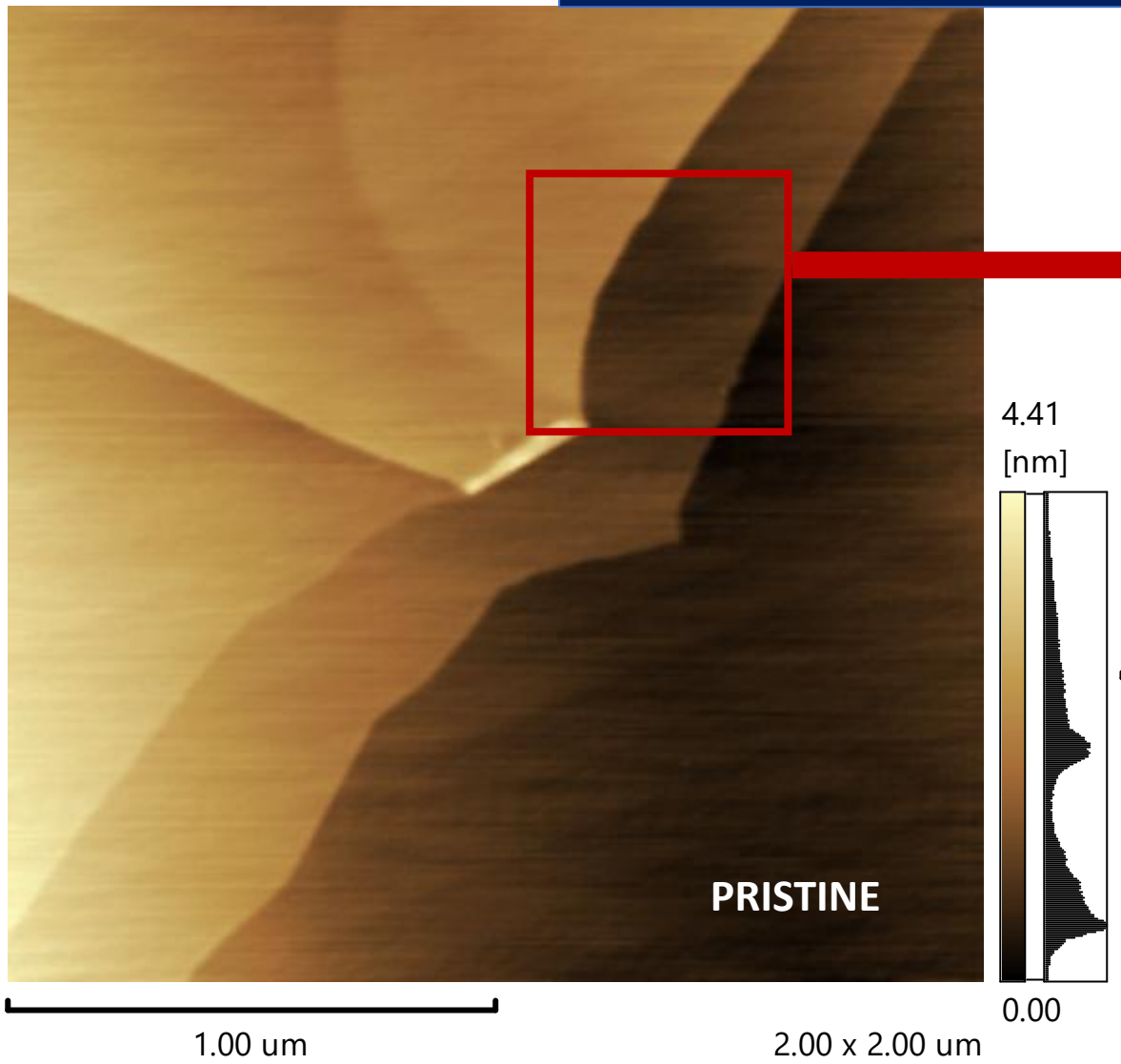


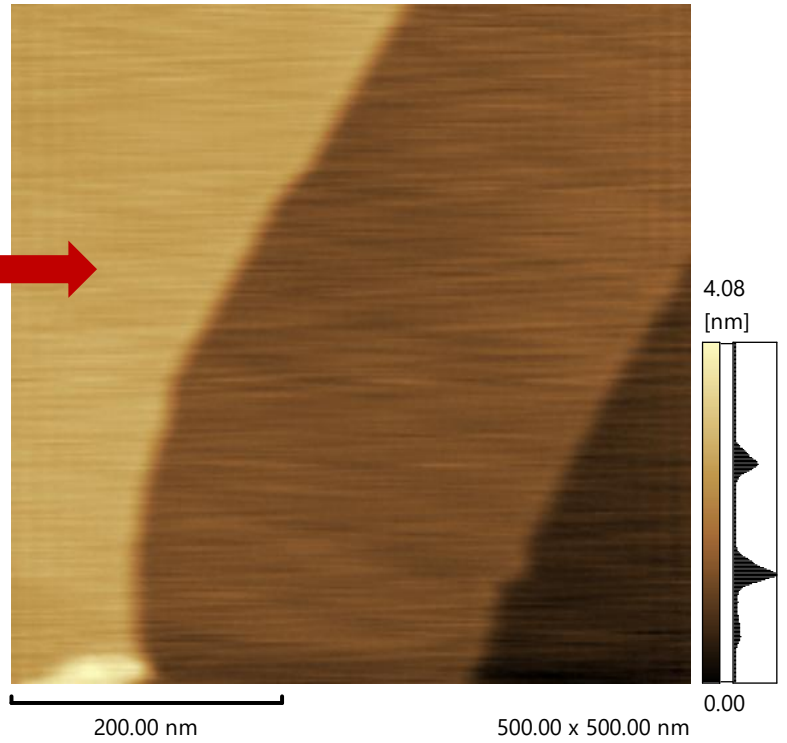


SEM
Scanning Electron Microscopy

Atomic Force Microscopes

HOPG Steps after removing a surface layer and before n-irradiation





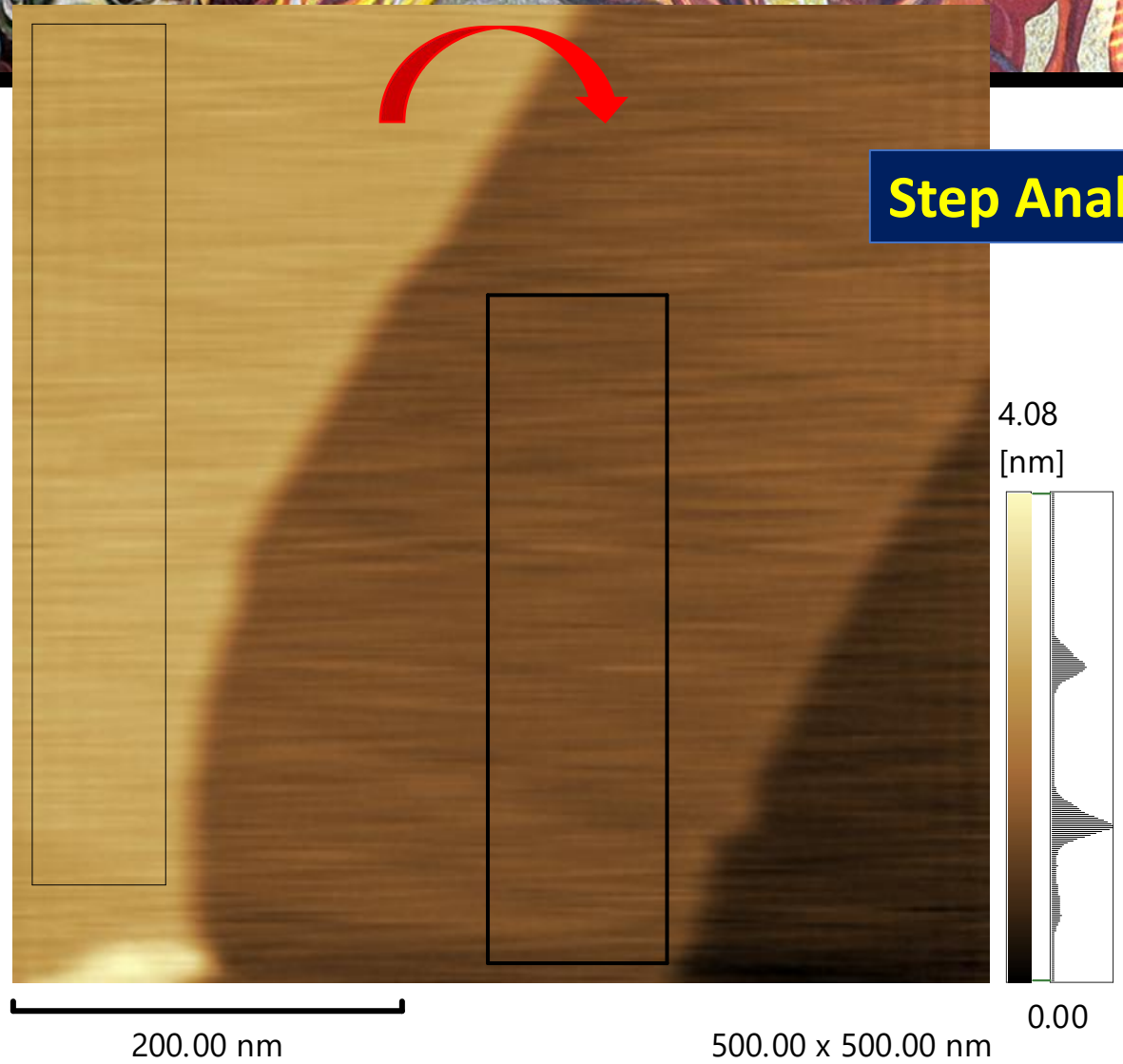
HOPG - AFM

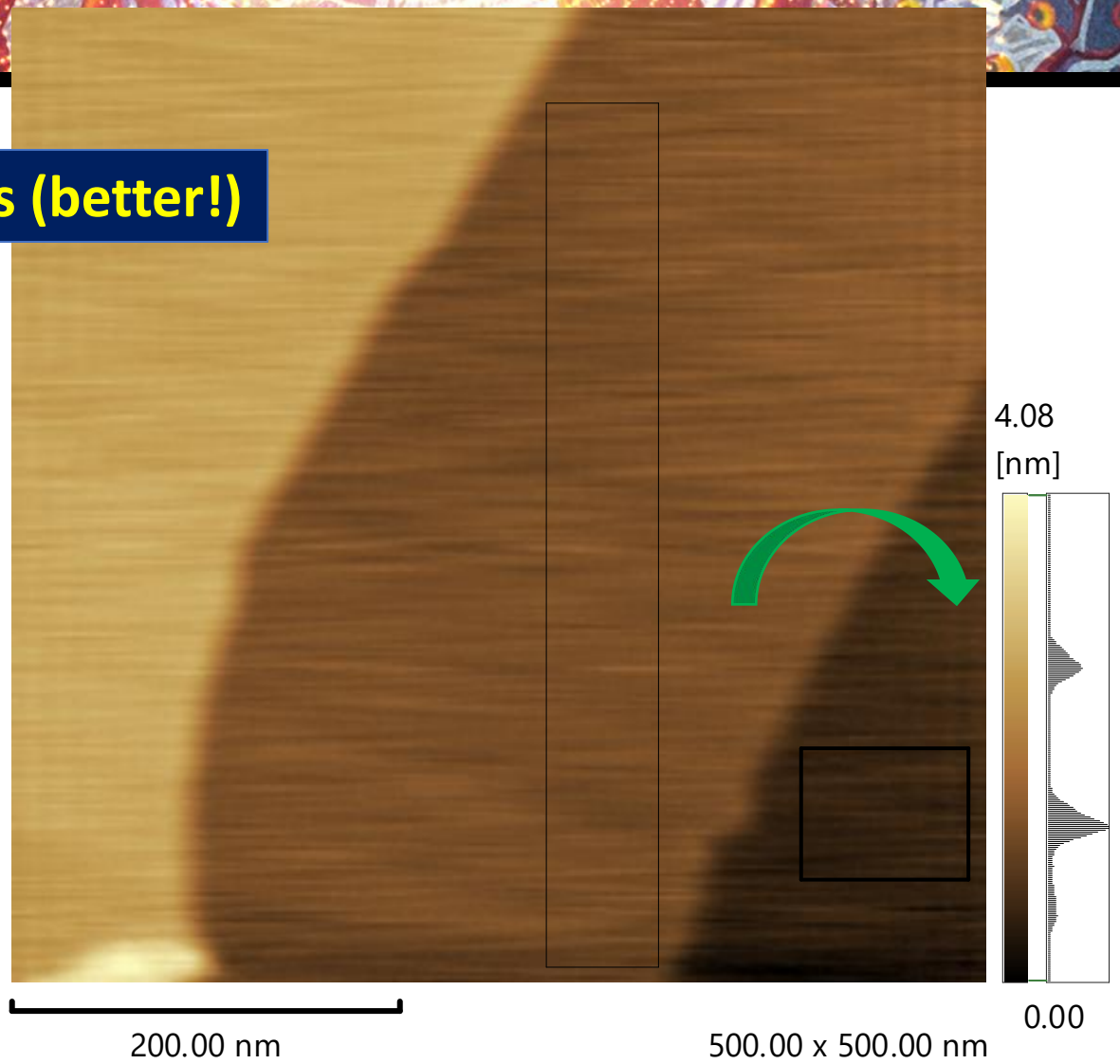
Surface morphology:
Atomic Force Microscopy,
dynamic mode (Si tip) in air.
(Shimadzu SPM9700)

RMS roughness and Step analysis:
at least 4 images, scanning areas of
(0.5 x 0.5) μm^2 (1.0 x 1.0) μm^2 (2.0 x 2.0) μm^2
(4.0 x 4.0) μm^2 (6.0 x 6.0) μm^2

HOPG - AFM – VIRGIN

Step Analysis (better!)



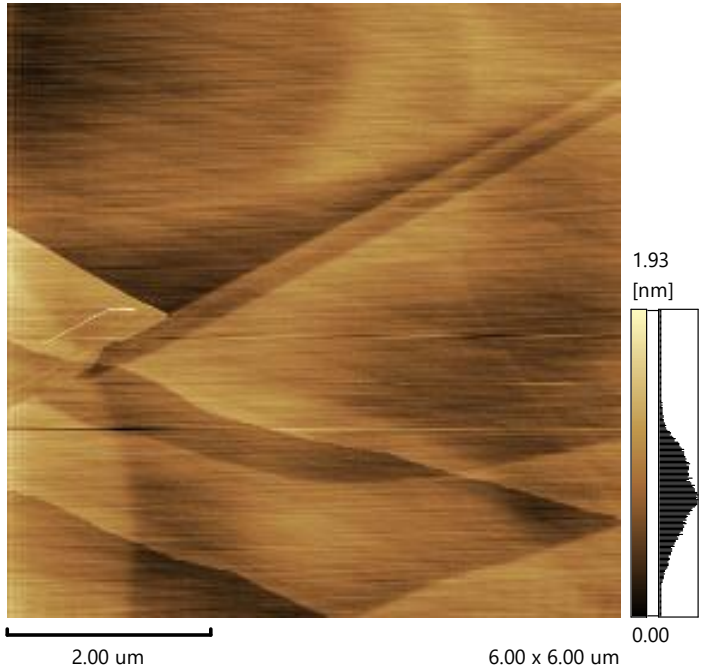


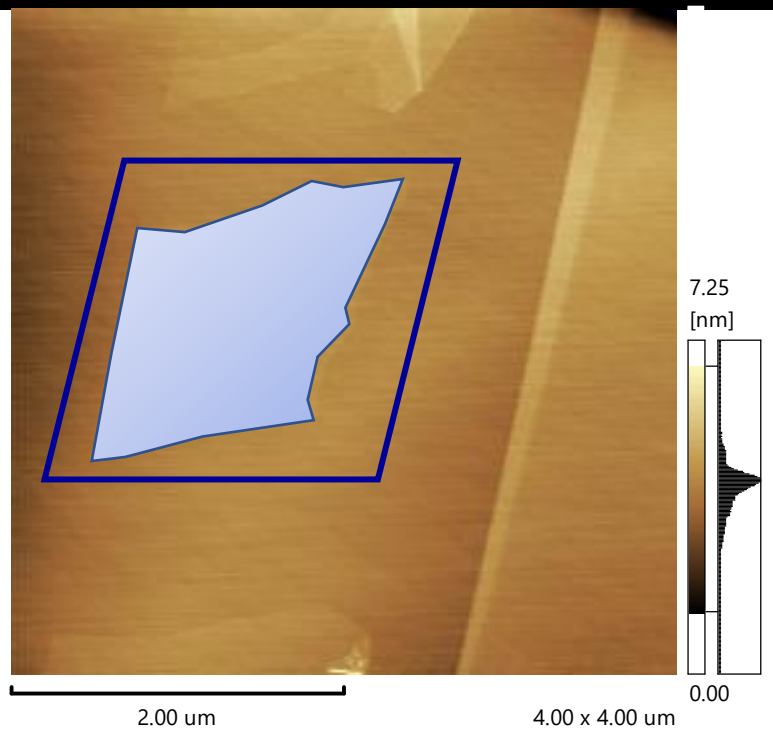
Step:1.571[nm] Area A:30107.50[(nm)2] Area B:31375.88[(nm)2]

Step:0.631[nm] Area A:25545.12[(nm)2] Area B:5790.71[(nm)2]

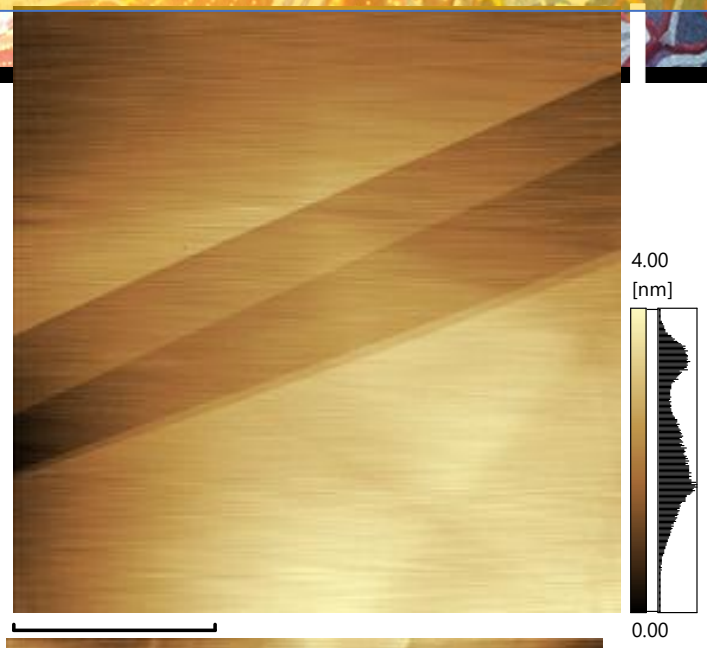
Similiar analysis after neutron irradiation

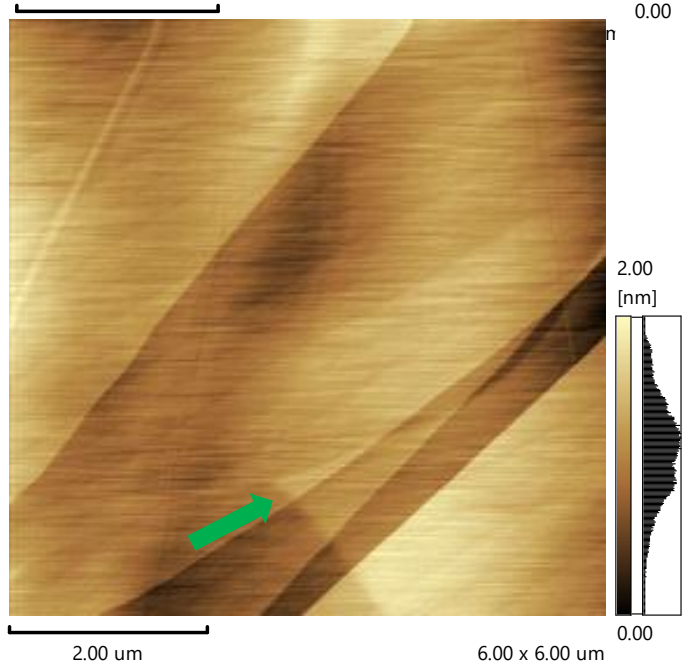
PRISTINE





AMOSTRA 3 - APOS IRRADIACAO RETIRADA CAMADA SUPERFICIAL REGIAO CENTRAL 2





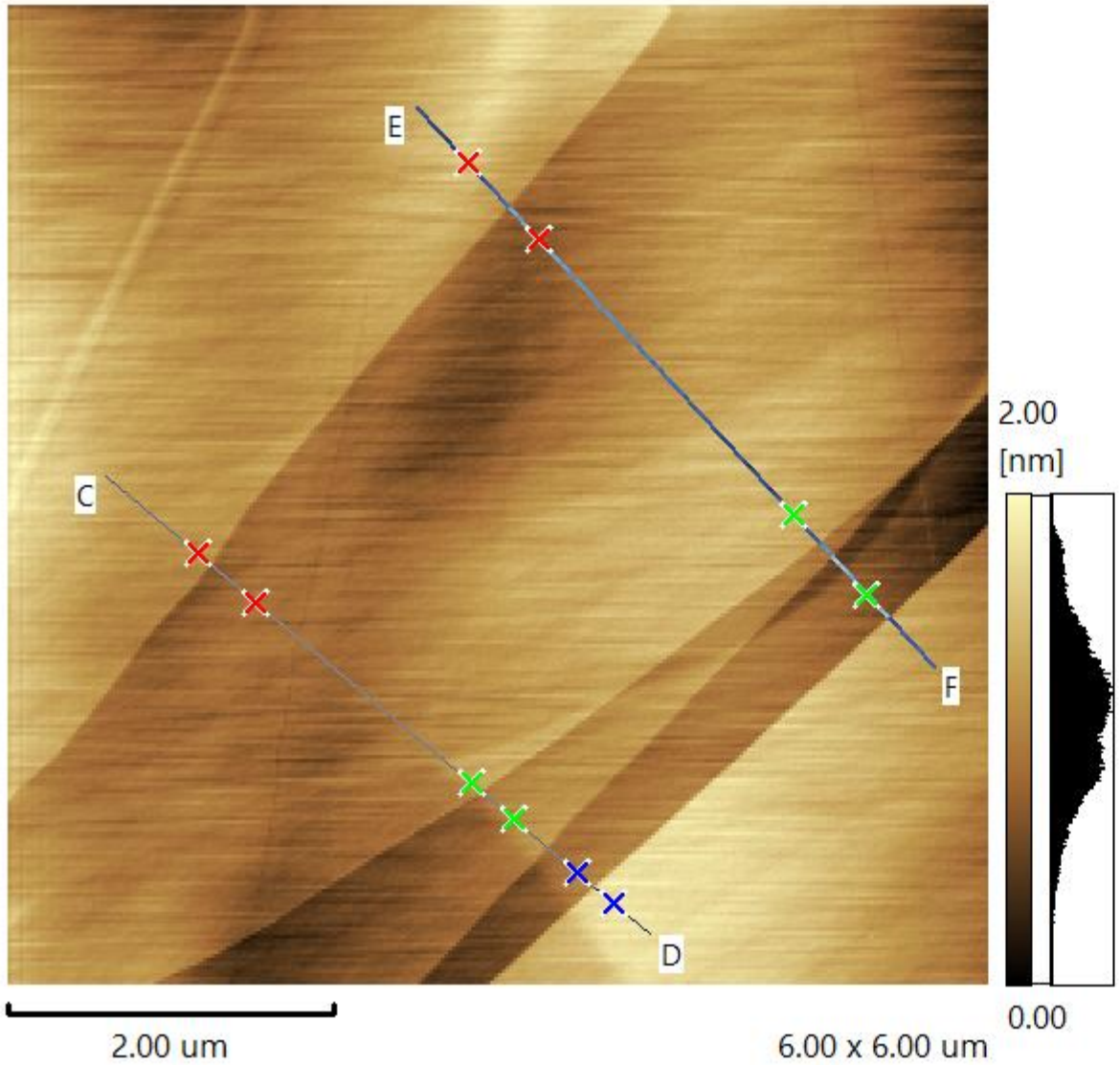
HOPG - AFM – 1N

SURFACES APPEAR WITH DIFFERENT NUMBER OF LAYERS AND IMPERFECTIONS

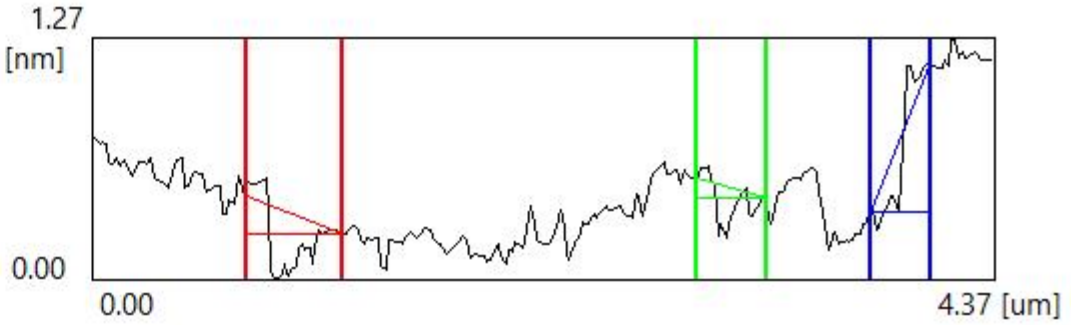
IMPERFECTIONS ARISE THAT WERE NOT SEEN IN THE PRISTINE

Sample 1N

HOPG - AFM – 1N

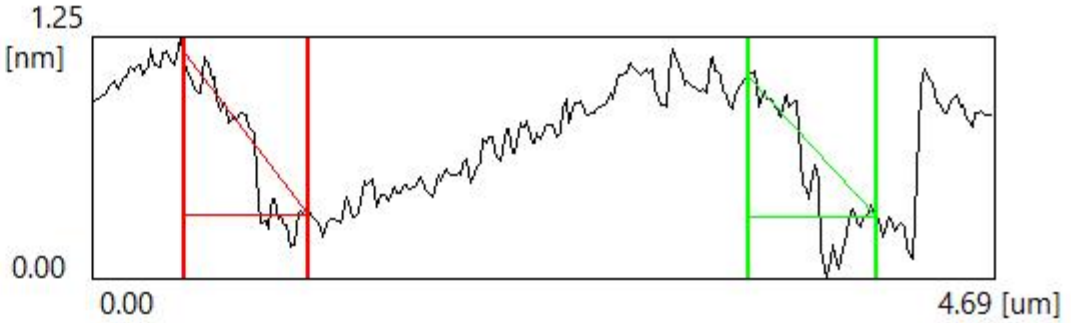


C-D



	Width [um]	Height [nm]	Angle [deg]
	0.47	0.21	0.03
	0.34	0.14	0.02
	0.29	0.77	0.15

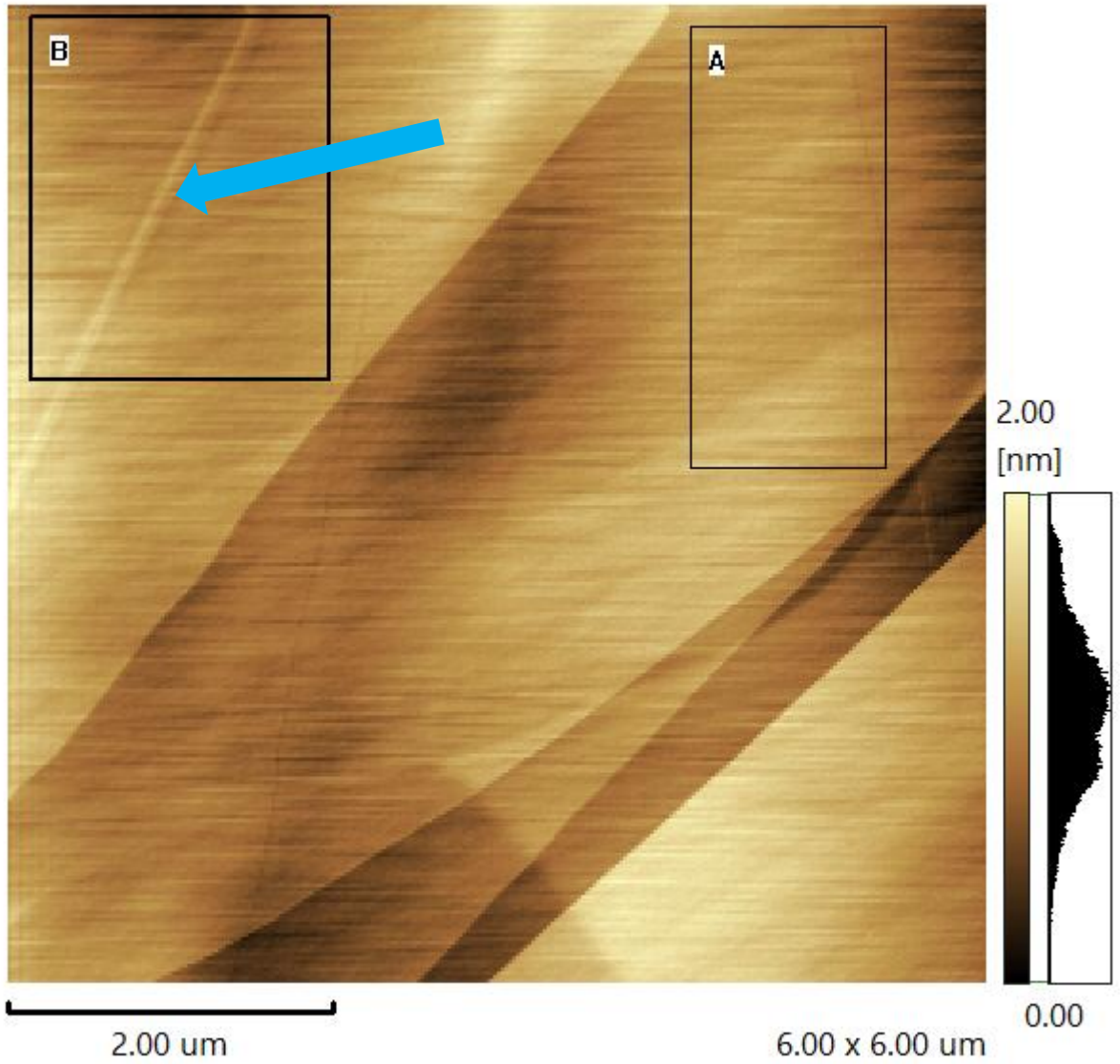
E-F



	Width [um]	Height [nm]	Angle [deg]
	0.65	0.85	0.07
	0.67	0.76	0.06

Sample 1N

HOPG - AFM – 1N



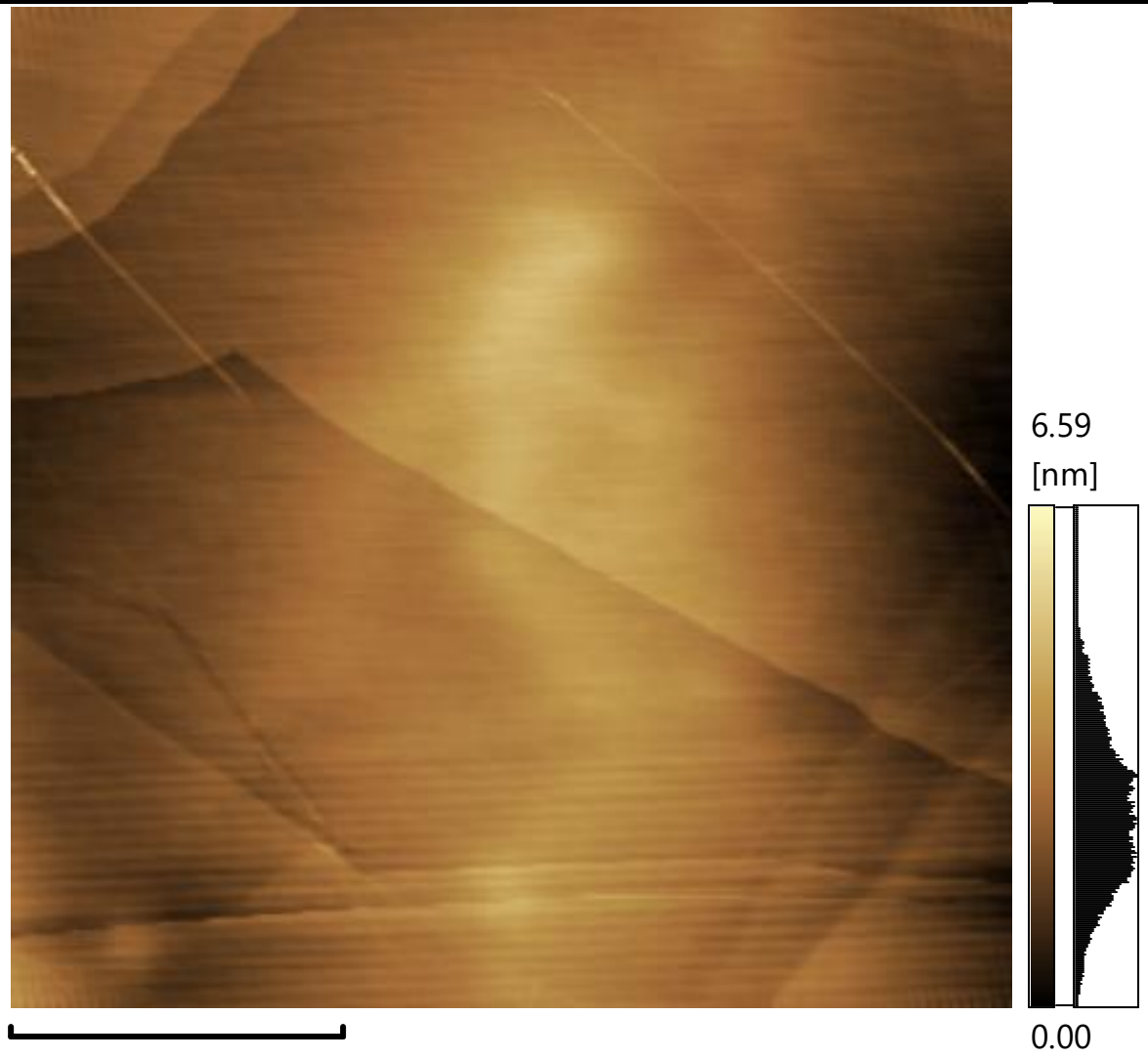
RMS – Roughness Analysis

All Area		A		B	
LengthX	6.000[um]	LengthX	1.195[um]	LengthX	1.828[um]
LengthY	6.000[um]	LengthY	2.707[um]	LengthY	2.227[um]
Area	36.000[(um)2]	Area	3.236[(um)2]	Area	4.070[(um)2]
Ra	0.252[nm]	Ra	0.281[nm]	Ra	0.783[nm]
Rz	2.013[nm]	Rz	0.955[nm]	Rz	1.622[nm]
Rzjis	0.971[nm]	Rzjis	0.413[nm]	Rzjis	0.000[nm]
Rq	0.313[nm]	Rq	0.320[nm]	Rq	0.841[nm]
Rp	0.896[nm]	Rp	0.239[nm]	Rp	0.061[nm]
Rv	1.117[nm]	Rv	0.716[nm]	Rv	1.562[nm]

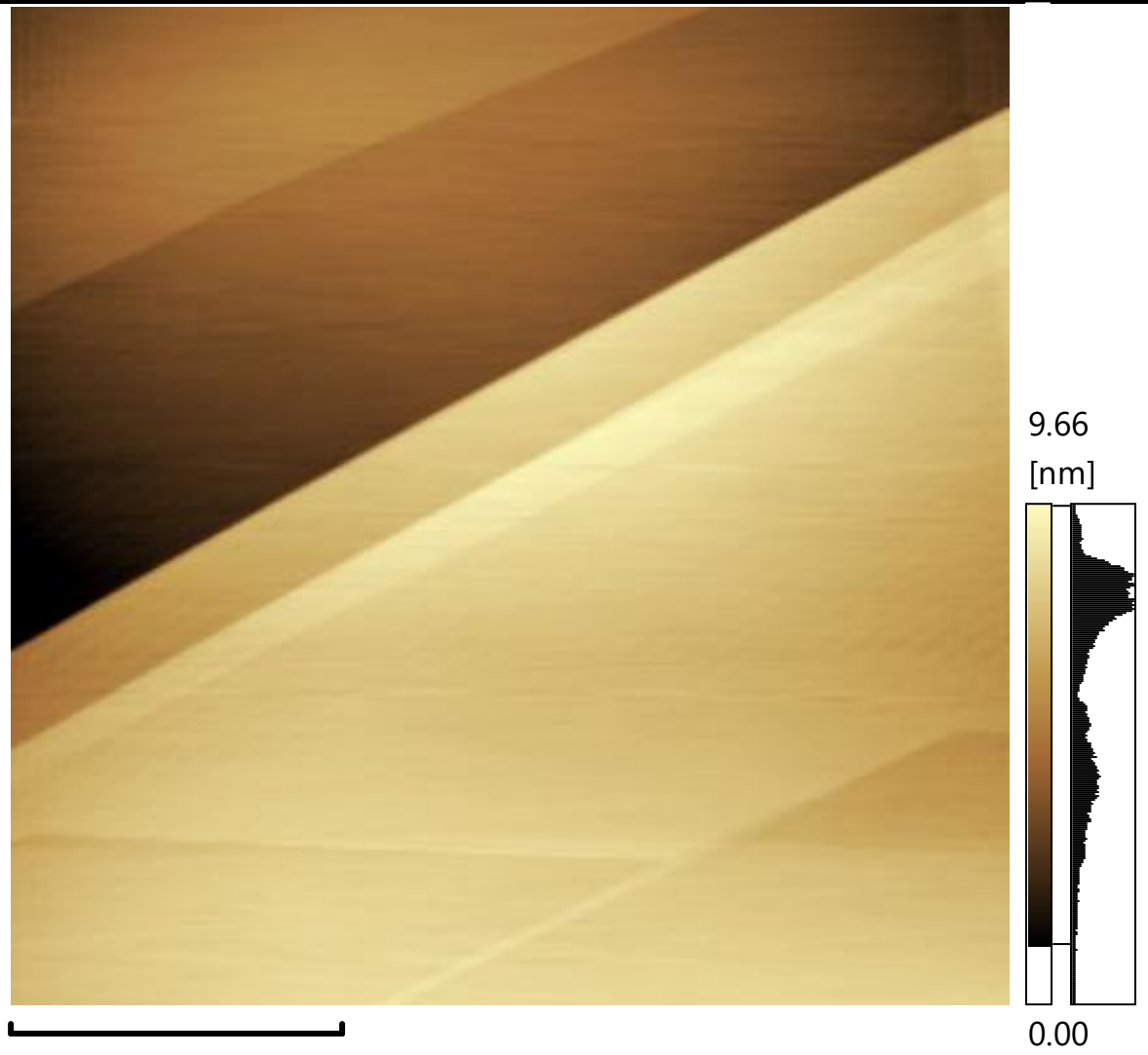
STEP BETWEEN LAYERS (PLANES) **0.1 nm to 0.9 nm**

LAYER ROUGHNESS (PLANE) **0.3 nm to 0.8 nm**

HOPG before n-irradiation



HOPG After n-irradiation 4N



HOPG before n-irradiation

STEP BETWEEN LAYERS (PLANES)	0.914 nm
LAYER ROUGHNESS (PLANE)	0.65 nm

HOPG After 4N

STEP BETWEEN LAYERS (PLANES)	1.318 nm
LAYER ROUGHNESS (PLANE)	0.66 nm

The average values estimated by AFM for the RMS roughness and heights between the graphene layers are also strong indicators of the weakening of the bonds across the planes, caused by the effect of radiation. The RMS roughness and distance between layer values were 0.65(14) nm and 0.914(87) nm, respectively, for the pristine sample without irradiation. After exposure to the neutron beam for 30.5 hours (sample N4), these values increased to 0.66(21) nm and 1.318(85) nm, respectively.

STOE *X-ray diffractometer* (*model* **STADI-P**)

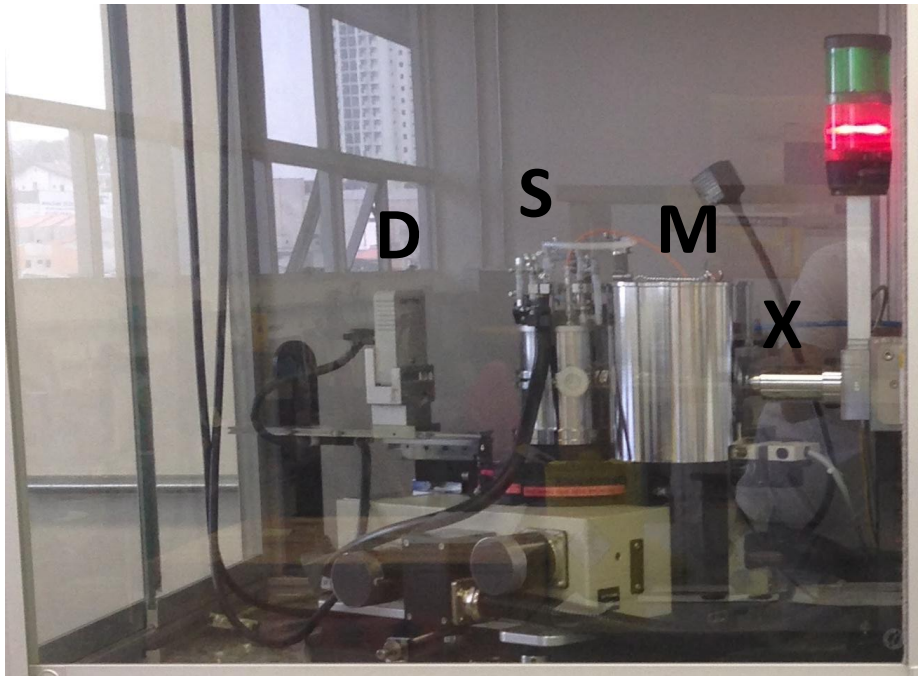
Transmission mode

D: solid state detector (Mythen 1K)

S: HOPG sample

M: curved Ge(111) monochromator

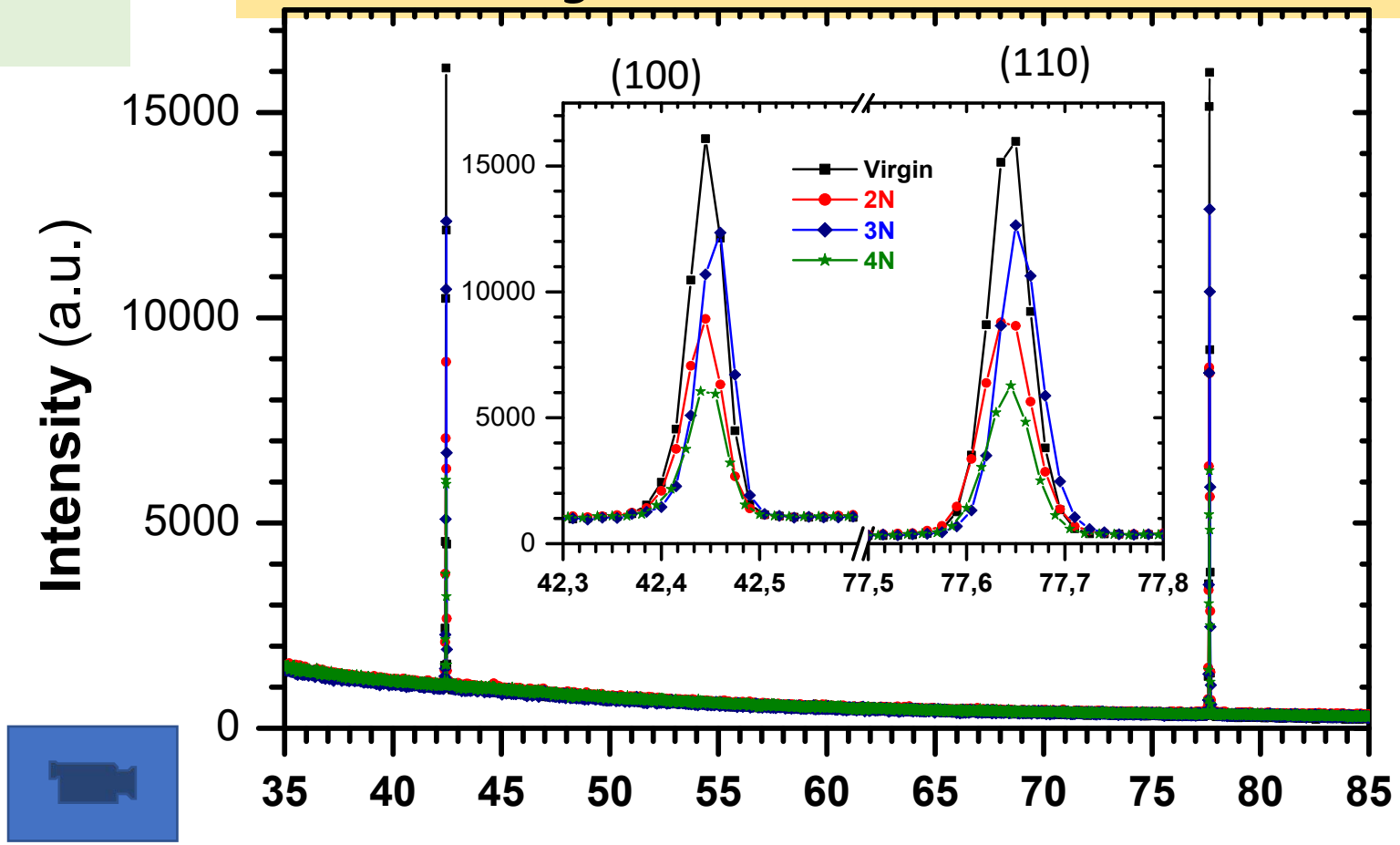
X: incoming X-ray



https://lccem.ufabc.edu.br/

Results from X-ray transmission measurements

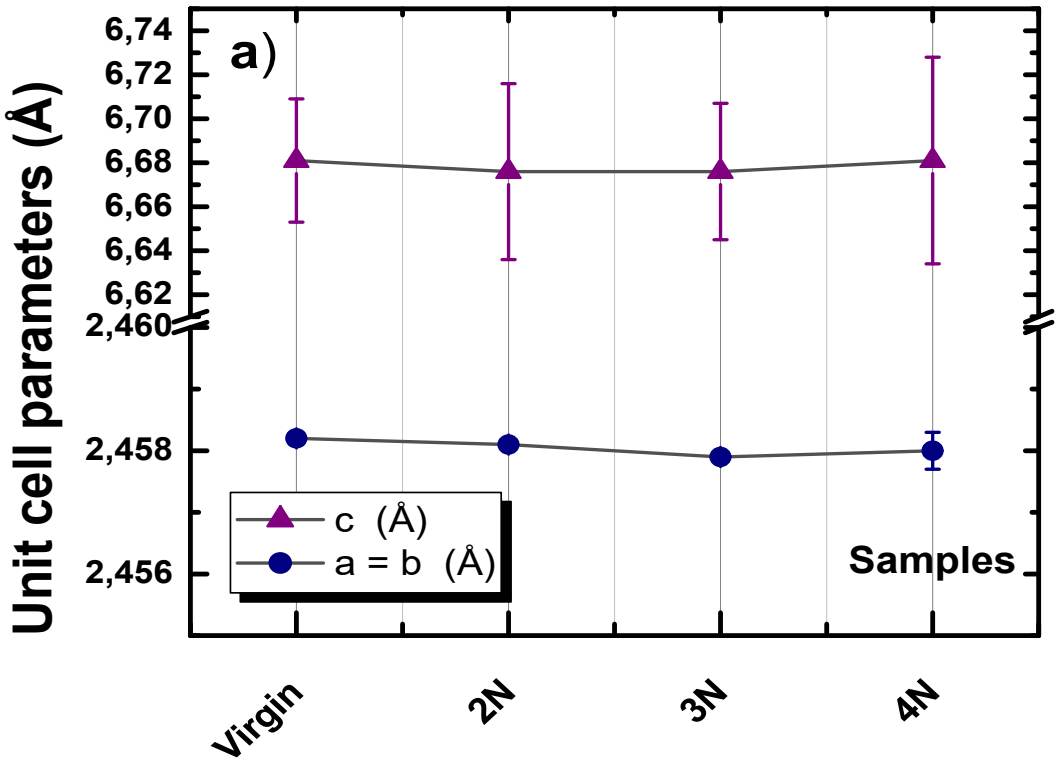
Rietveld using TOPAS code



X Ray Diffraction

UFABC

Results for a = b and c lattice parameters



Sample	FWHM (deg)	$\frac{\Delta a}{a}$	$\frac{\Delta c}{c}$
PRISTINE	0.0348 (4)	($a_o = 2.4582$ Å)	($c_o = 6.681$ Å)
2N	0.0376 (8) = 8.0%	$4.07(65) \cdot 10^{-5}$	$7.48(49) \cdot 10^{-4}$
3N	0.0335 (7) = - 3.7 %	$12.20(12) \cdot 10^{-5}$	$7.48(27) \cdot 10^{-4}$
4N	0.0368 (9) = 5.7 %	$8.14(18) \cdot 10^{-5}$	–

the variation in crystal lattice parameters is small

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MICRO RAMAN
EXPERIMENTAL SETUP

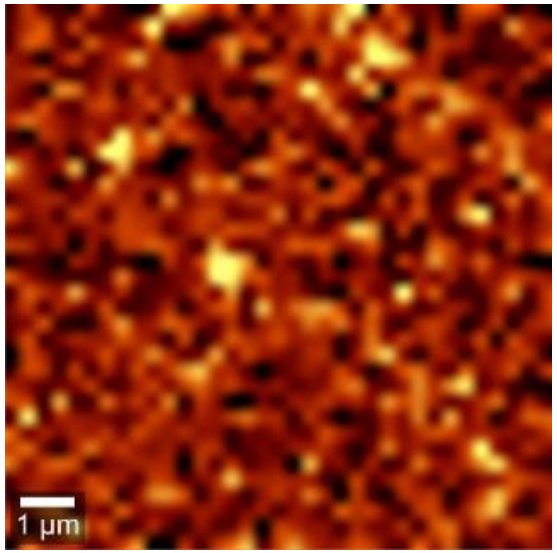
For the acquisition of Raman mappings, a confocal microscope RamanAlpha 300R (Witec) was used with a laser at a power of 30 mW, and integration time of 1 s for each pixel of the acquired image, with a collector fiber of 100 µm.

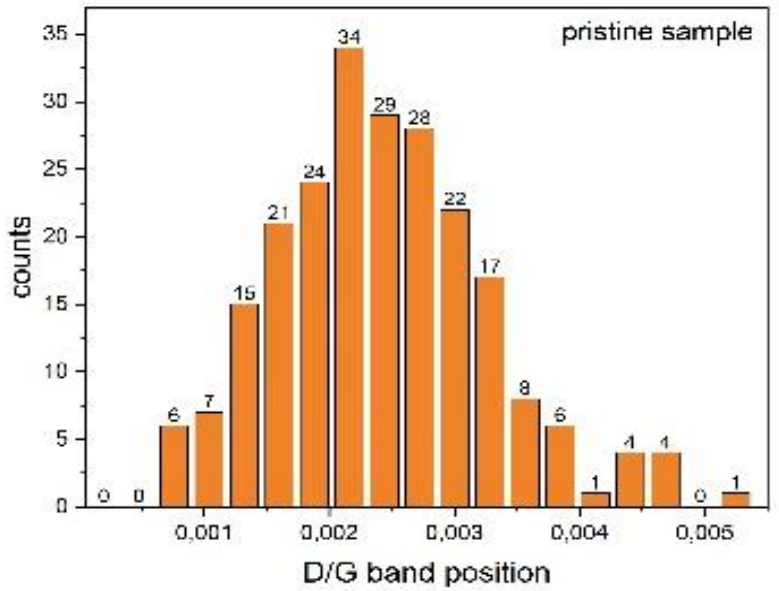
The pixel size used in all mappings was 286 nm.

The chosen size of the Raman mapping was 10 um, with the intention of better observing the presence of defects. The optical lens used in these experiments was 100x (NA=0.8, Nikon).

An average of 15 Raman mappings were performed at different points of the same sample, considering the two surfaces of the sample: front and back

HOPG - MICRO RAMAN



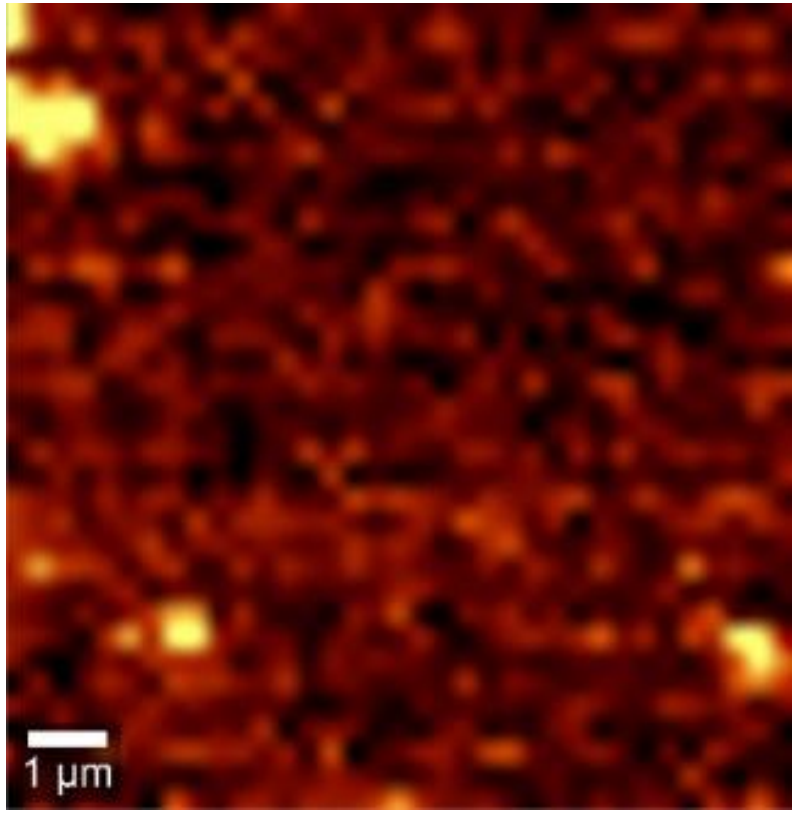


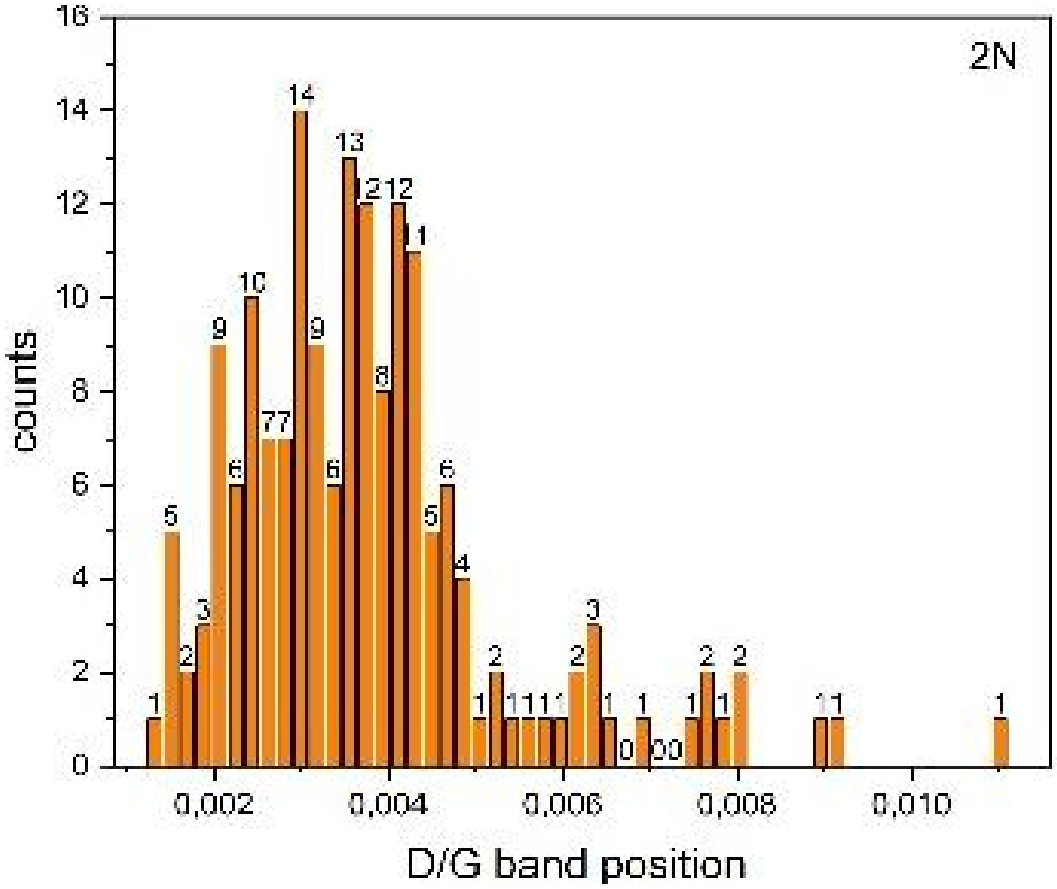
PRISTINE

D/G = 0.0035 ± 0.0008

The histograms represent the distributions of the intensity ratios of the D/G bands in the area analyzed by Raman mapping. Despite the sample before being exposed to the neutron beam already presenting a small number of defects, represented by the D/G = 0.0035(8) ratio, the results of this analysis reinforce that, as the exposure time to the neutron beam increases, the D/G ratio also increases, and the samples exhibit a subtle but noticeable loss of crystallinity. This gradual increase in the D/G ratio indicated that the structural order of the materials was transforming. This phenomenon can be attributed to the interaction of neutrons with the atomic structure of the samples, leading to structural modifications and a decrease in the degree of crystallinity.

D/G = 0.0042 ± 0.0007





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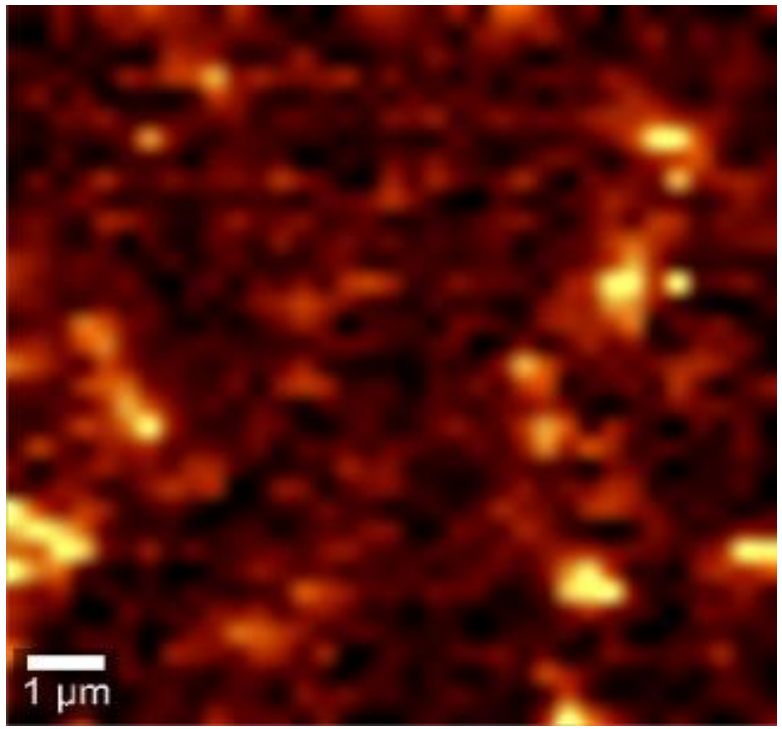
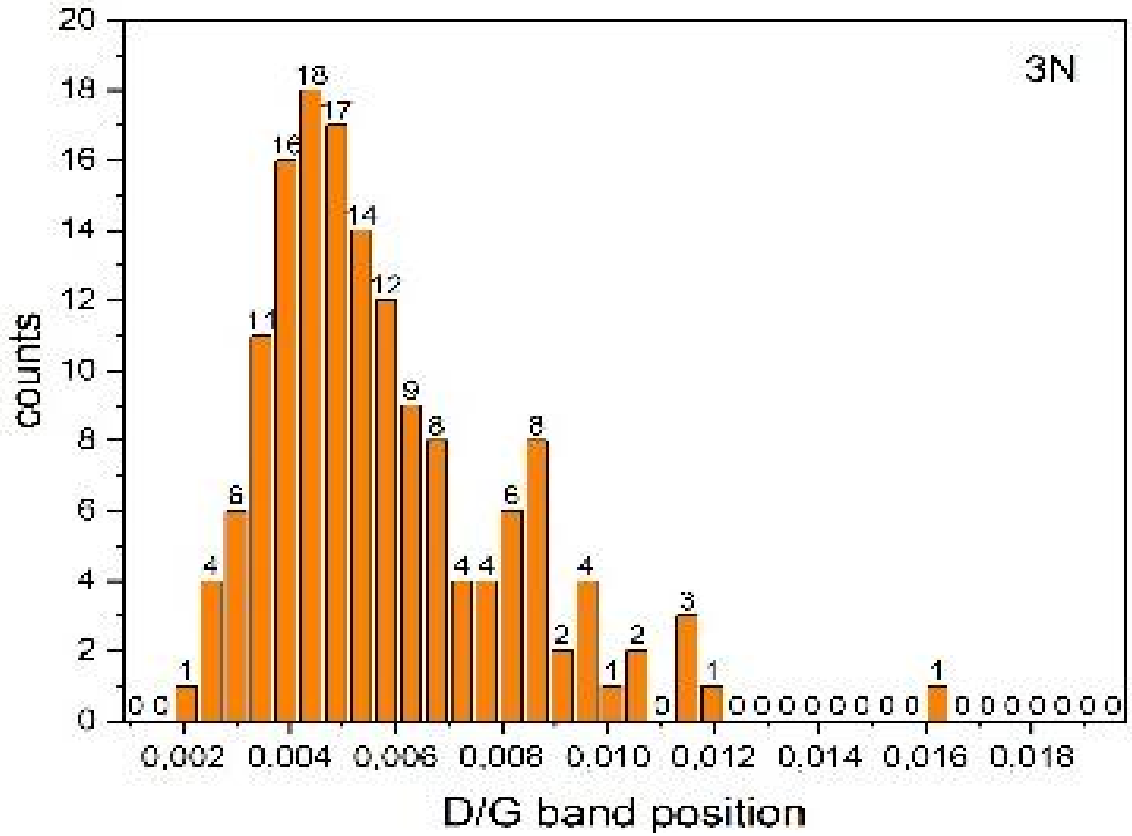
D/G = 0.0055 ± 0.0014





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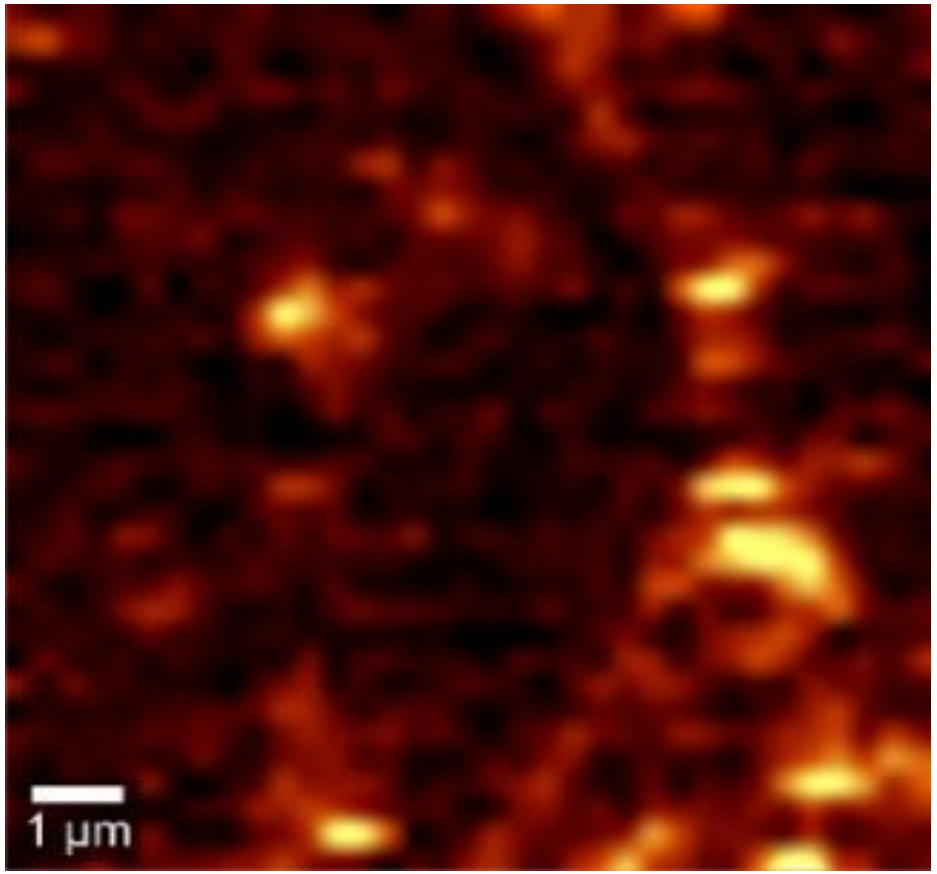
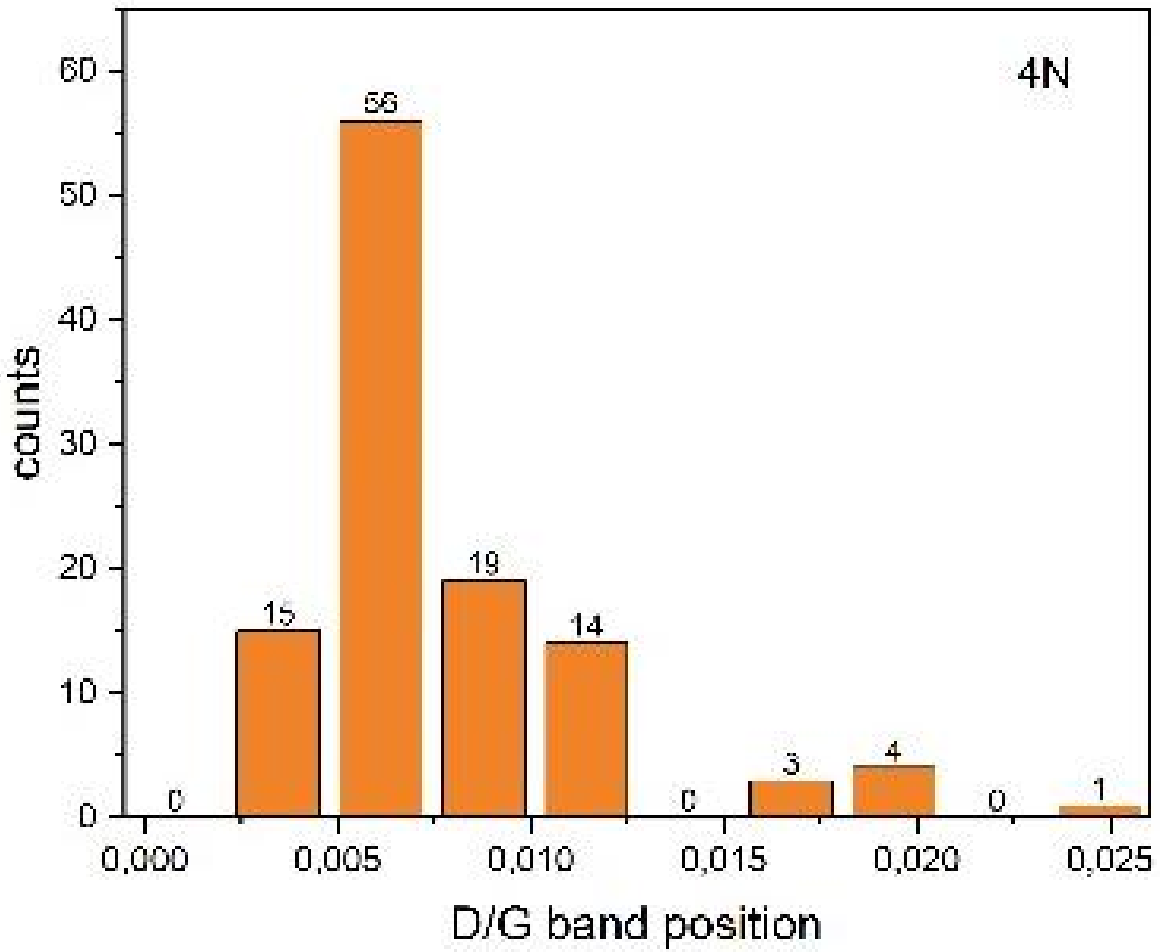
D/G = 0.0092± 0.0021





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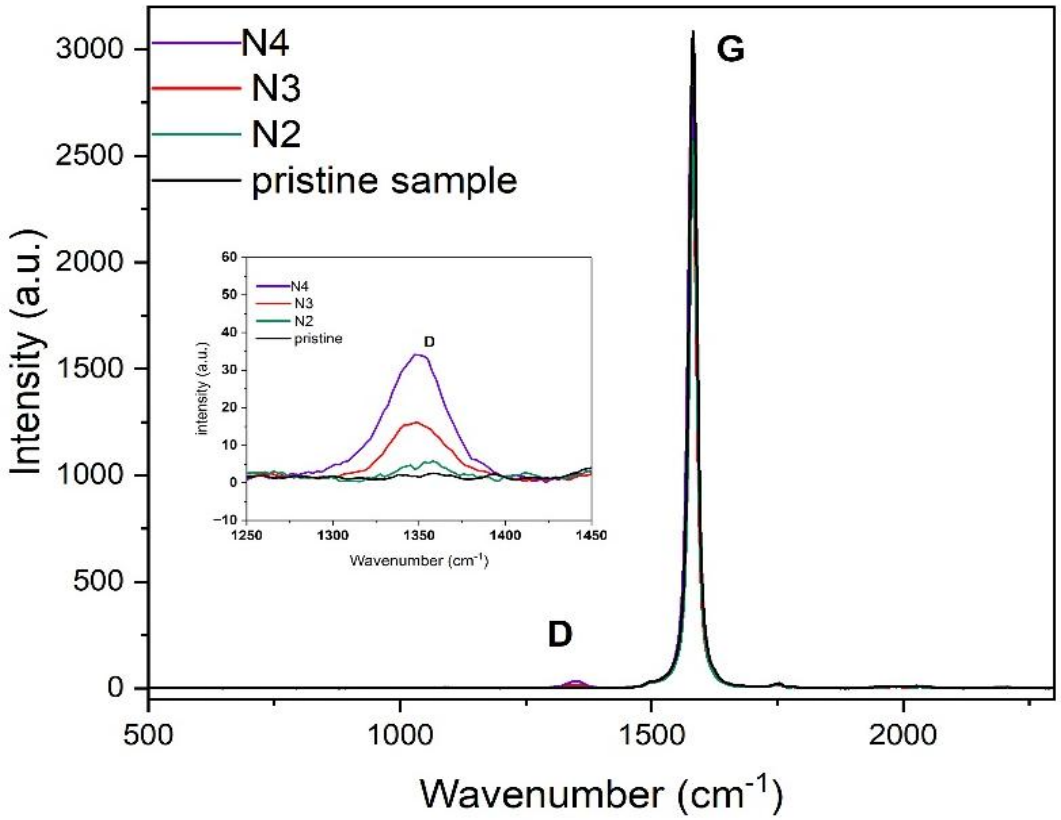
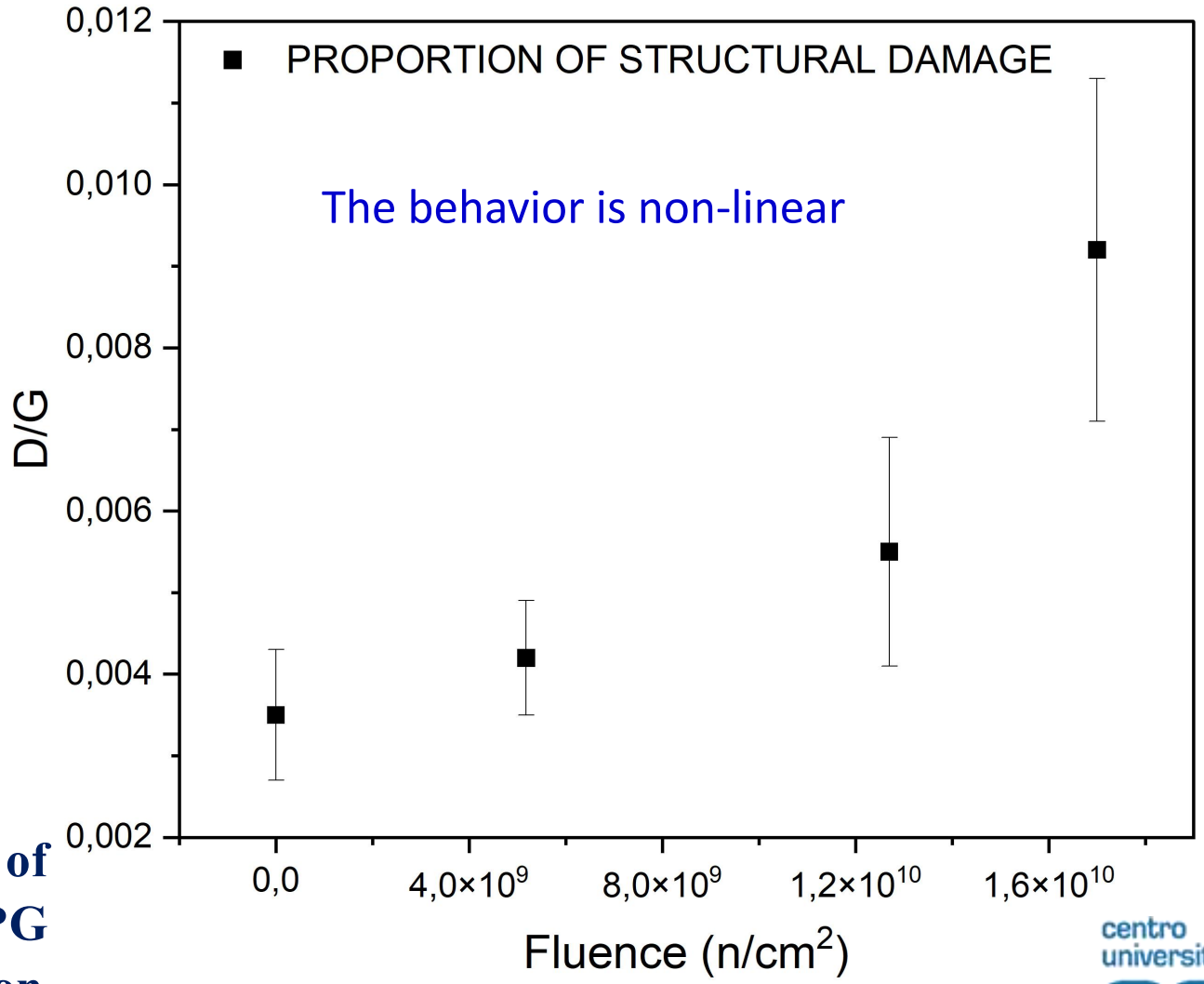
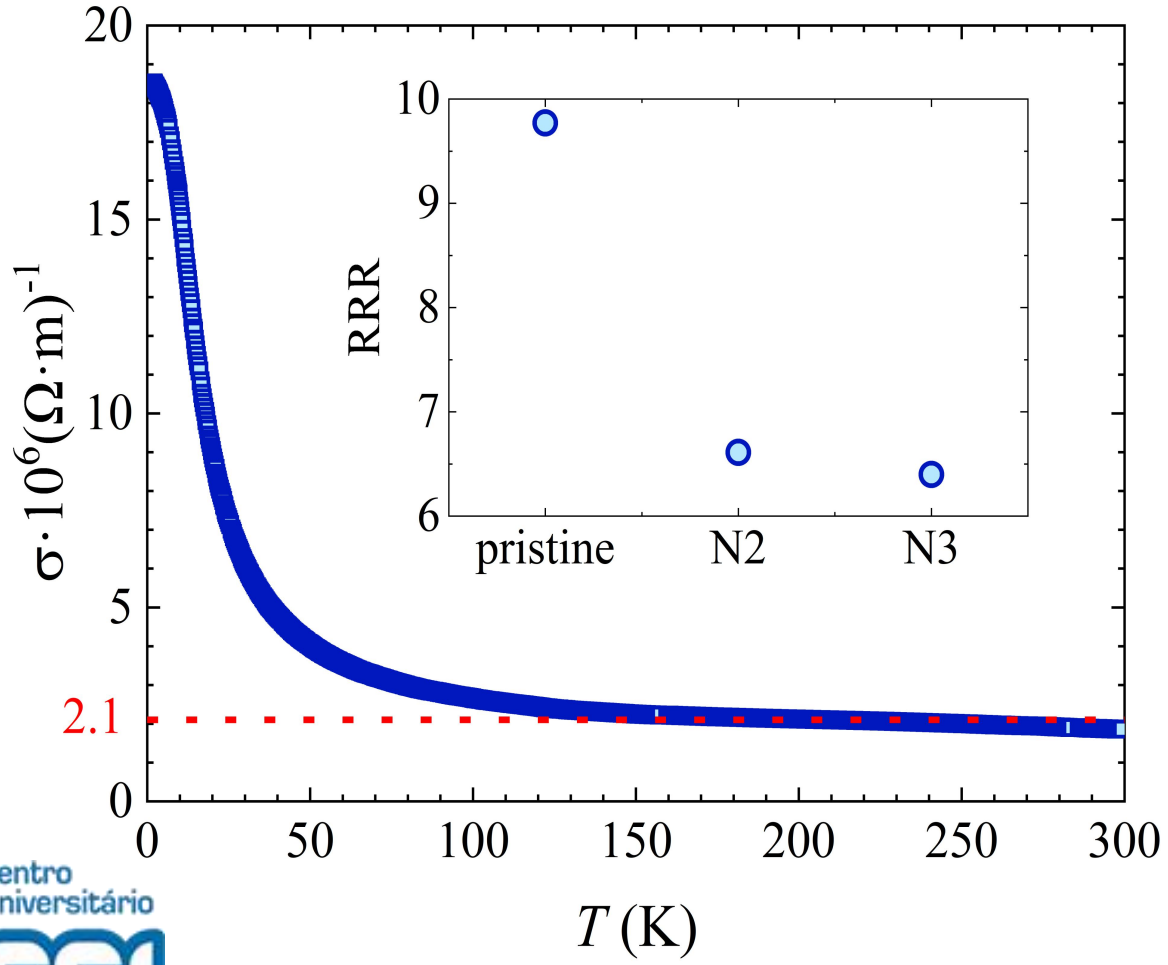
HOPG - MICRO RAMAN





D band intensities, associated with the presence of defects, increase with exposure time of the HOPG foil to the neutron beam. This suggests a degradation in the thermal properties of the material.

Electrical Resistivity and Magnetoresistance



In-plane **electrical conductivity (σ)** of pristine HOPG sample as a function of temperature.

The σ value at room temperature (RT) aligns with the manufacturer's provided value of $2.1 \cdot 10^6$ $(\Omega \cdot m)^{-1}$, as indicated by the red dashed line on the graph.

RESIDUAL RESISTIVITY RATIO (RRR)

The inset displays the **residual resistivity ratio**, which serves as an indicator of the crystalline quality of the materials under investigation. The RRR values decrease from 9.8 for the pristine sample to 6.6 and 6.4 for irradiated samples N2 and N3, respectively. This decline suggests a reduction in the degree of crystallinity corroborating the findings of Raman spectroscopy and XRD analysis.

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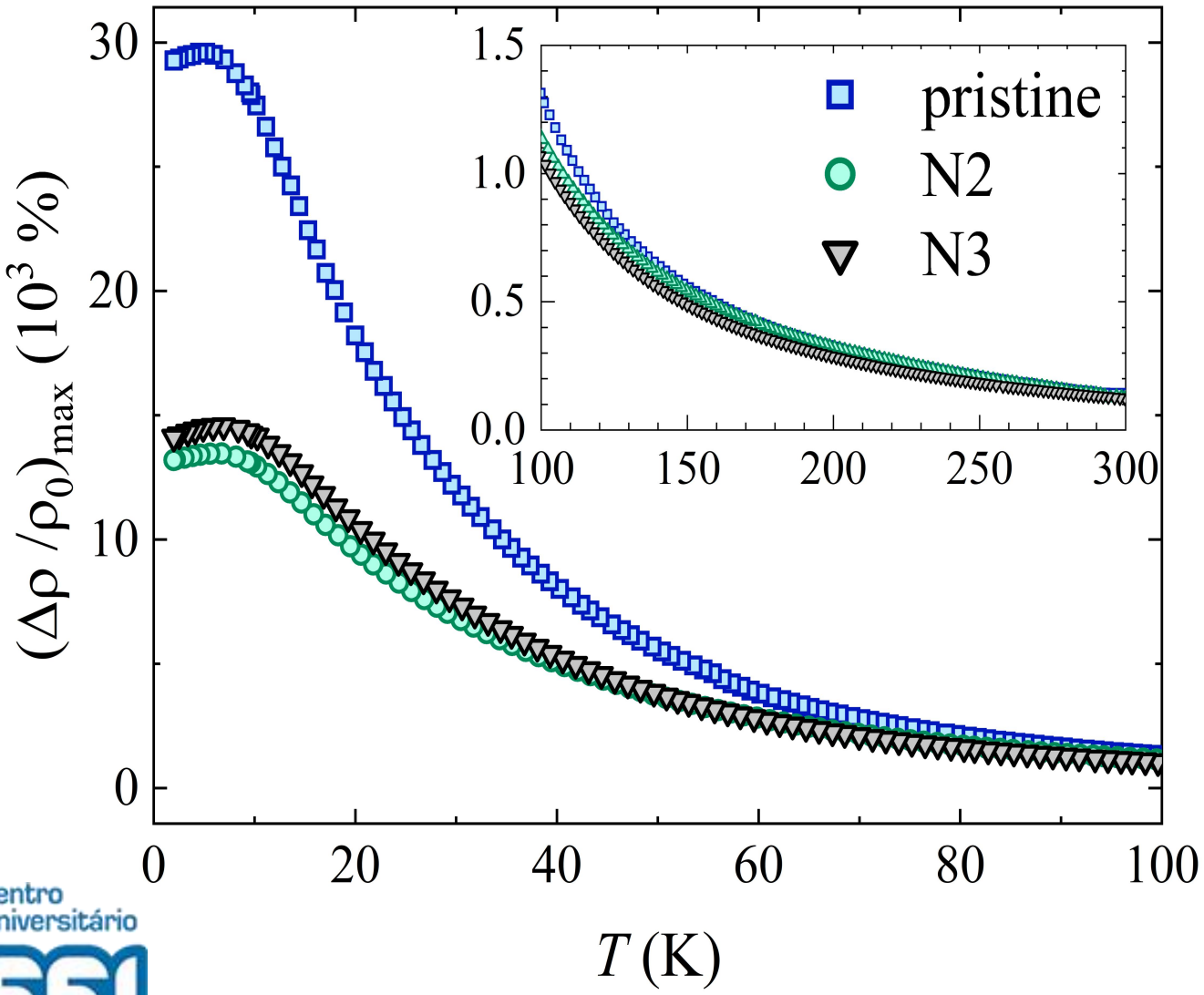

The temperature-dependent maximum magnetoresistance, $(\Delta\rho/\rho)_{max}$, measured under an applied magnetic field of 1 T, is shown in Figure. At low temperatures, defect scattering significantly influences the electrical resistivity, thus resulting in notable variations in the $(\Delta\rho/\rho)_{max}$ relative to crystal perfection.

At 4 K, $(\Delta\rho/\rho)_{max}$ values for samples N2 and N3 decreases to 45% and 49%, respectively, of the maximum magnetoresistance observed for the pristine sample. In contrast, the $(\Delta\rho/\rho)_{max}$ curves for the pristine HOPG, as well as samples N2 and N3, demonstrate convergence near RT.

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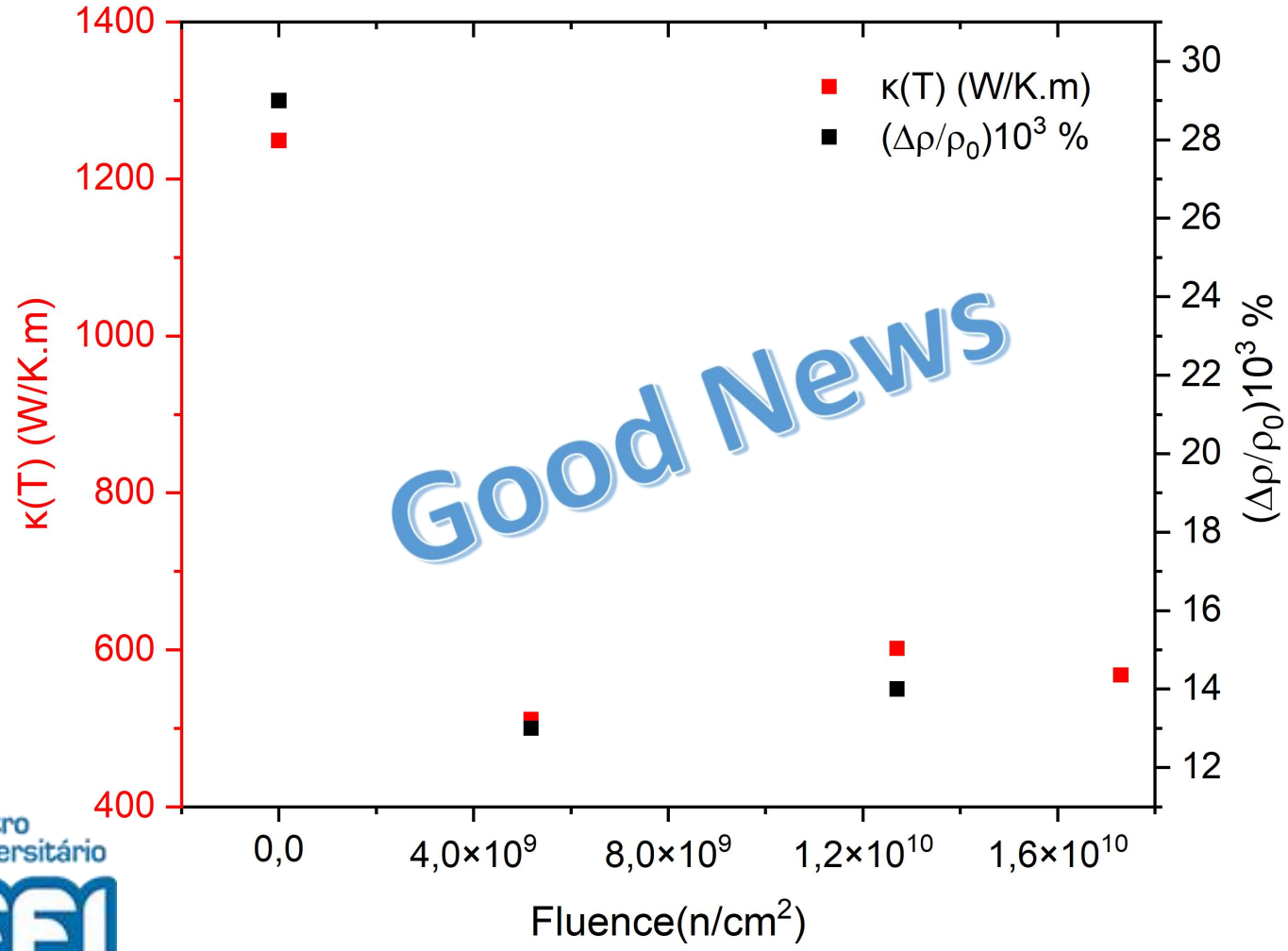
Thermal Conductivity

κ(T) reduction of ~ 45 % related to the increased defect



The steady-state thermal conductivity κ(T) measurements were taken using a homemade apparatus based on the parallel heat-flow technique.

Rectangular HOPG samples (typically 0.2 μm x 4 mm x 6 mm) were glued to this apparatus with silver epoxy, where the upper and low edges of the sample were fixed to the hot and cold terminals of the sample holder. All measurements were performed at room temperature (300 K) inside a close-cycle cryostat (ARS -system) at high vacuum (~10^{-5} mbar).

$(\Delta\rho/\rho)_{max}$ decreases ~ 45%

κ(T) decreases ~ **45%**

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AFM and SEM indicate that the bond between graphene layers becomes weaker with the effects of radiation. The average roughness value remains practically the same, but the height between layers increases.

Raman analysis showed a tendency of disorder increases as a function of irradiation, making it possible to calculate the disorder fraction presented in the HOPG. The ratio of disorder to HOPG's ordered structure, D/G, almost tripled.

XRD showed alterations in the lattice parameters, indicating alterations in the degree of disorder of the atomic structure - loss of crystal orientation.

Magnetoresistance has a strong correlation with thermal conductivity, indicating that even minor damage to the crystalline structure of HOPG significantly degrades its physical properties. This highlights a notable decrease in heat dissipation efficiency.

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WITH THE RESULTS OBTAINED, ACCORDING TO THE NEUTRON FLUENCE THAT REACHES THE TARGET IN A NUCLEAR REACTION, IT IS POSSIBLE TO ESTIMATE THE DEGRADATION IN THE THERMAL AND ELECTRICAL PROPERTIES OF THE MATERIAL.

- AFTER **ABOUT 10 HOURS OF EXPOSURE**, THERE WAS ALREADY AROUND A **45%** DECREASE IN THE THERMAL CONDUCTIVITY OF HOPG.

ALTHOUGH HOPG AND GRAPHENE MATERIALS HAVE EXCELLENT PROPERTIES, SIGNIFICANT DEGRADATION OCCURS AFTER INTERACTION WITH IONIZING RADIATION DUE TO MINOR CHANGES IN THEIR CRYSTALLINE STRUCTURE.

HOPG 1700 W/m ·K, after irradiation 935 W/m ·K,
Copper = 410 W/m ·K

EVEN SO, AFTER DEGRADATION, THE CONDUCTIVITY OF HOPG IS STILL HIGHER THAN THAT OF COPPER.

But…. this is only due to fast neutrons!

Heavy ion beam damage remains to be checked!

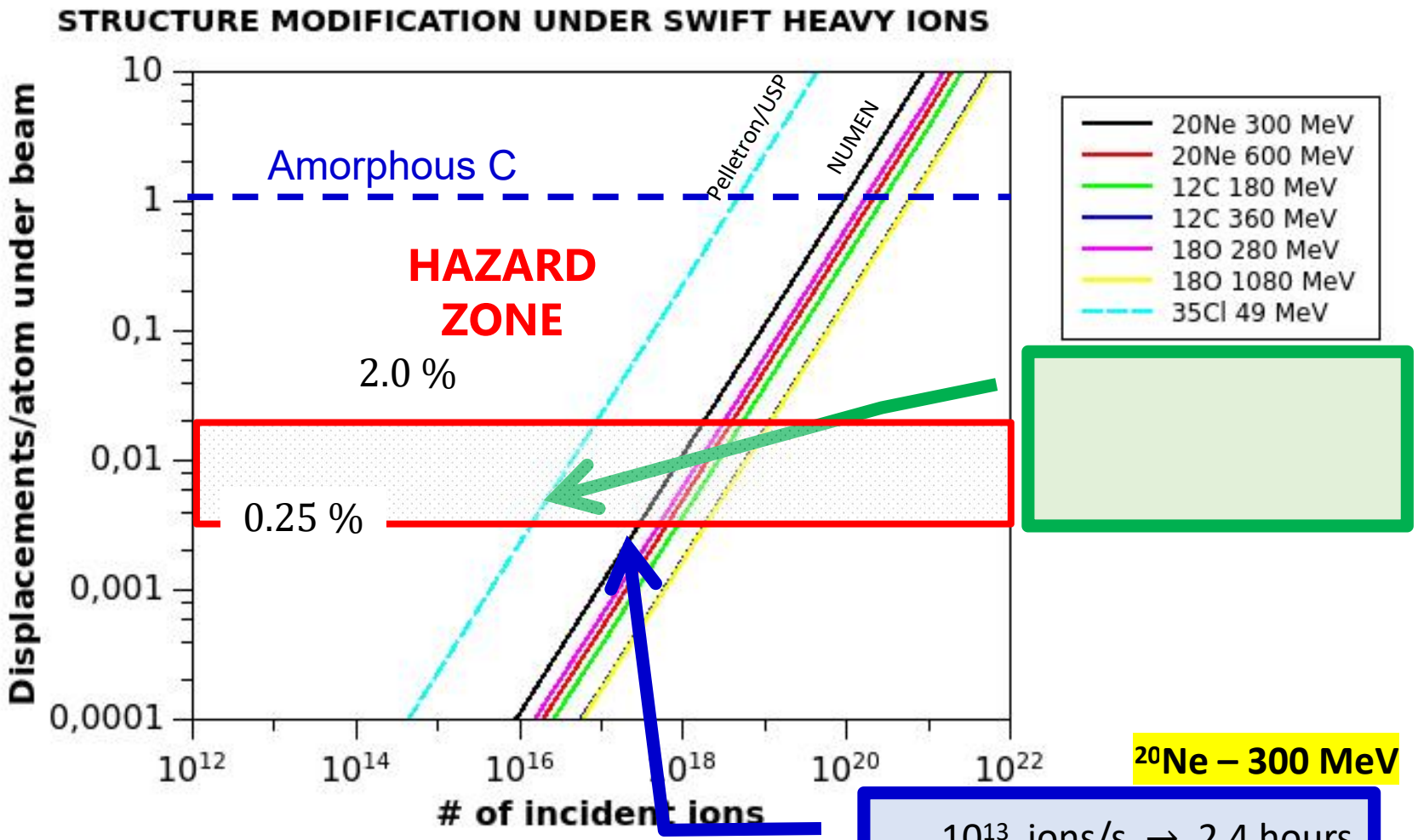
RUNNING...

*"The results suggest that only **0.25%** concentration of defects in **graphene** result in significant reduction of termal conduction property of graphene by around **50%**"*

CARBON 63 (2013) 460–470

35-Clorine of 49 MeV

which simulates the same damage obtained by intense beams from NUMEN



^{20}Ne – 300 MeV

10^{13} ions/s → 2.4 hours
10^{12} ions/s → 24 hours
10^{11} ions/s → 240 hours

SUMMARY OF THE NEXT STEPS IN THE EXPERIMENTAL PROCEDURE

1.The experiment has already been conducted on the Pelletron with ^{35}Cl.

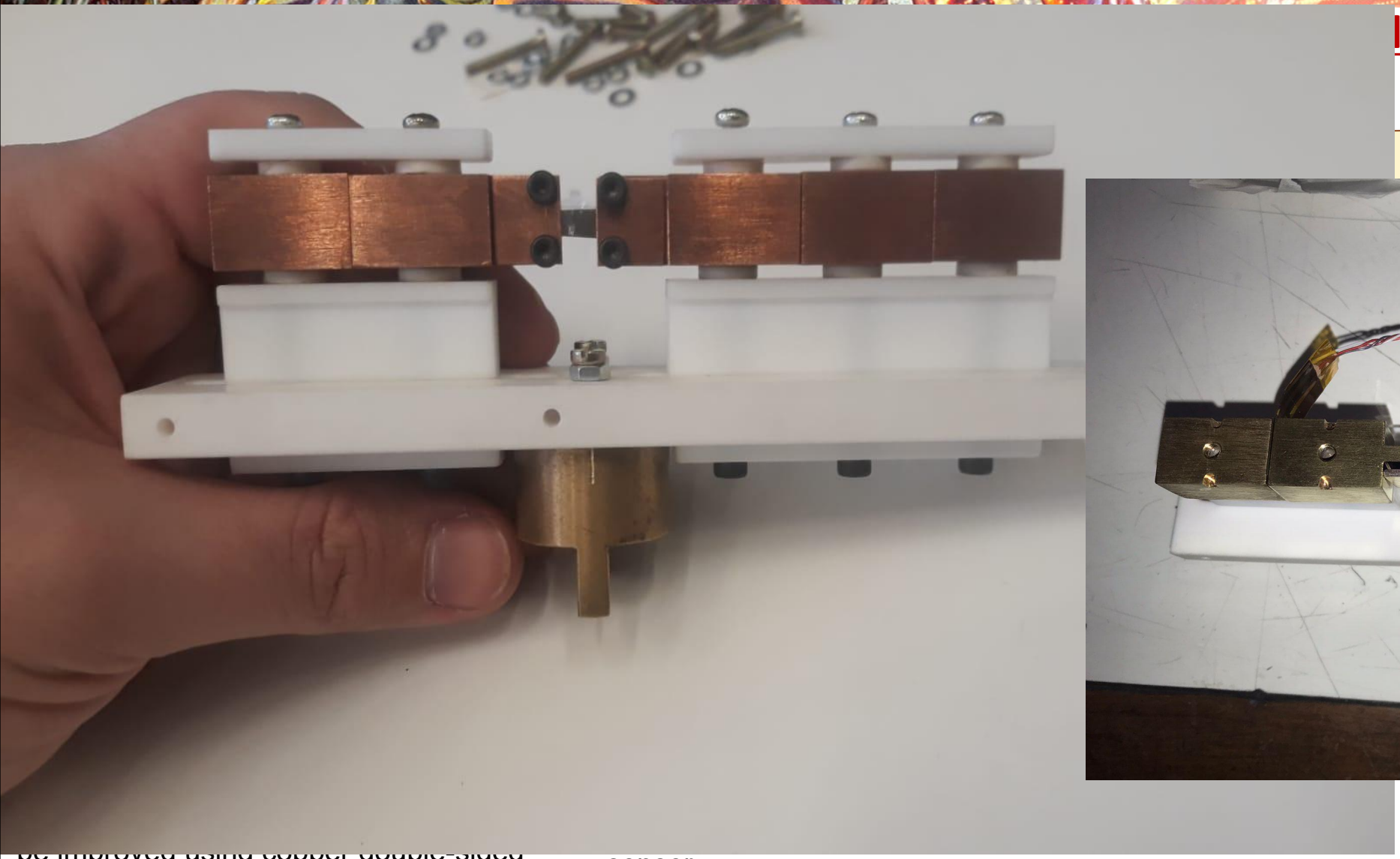
2.Samples from this experiment are being analyzed.

3.A second experiment will be conducted in August 2024 using the same reaction but with different HOPG targets from various manufacturers.

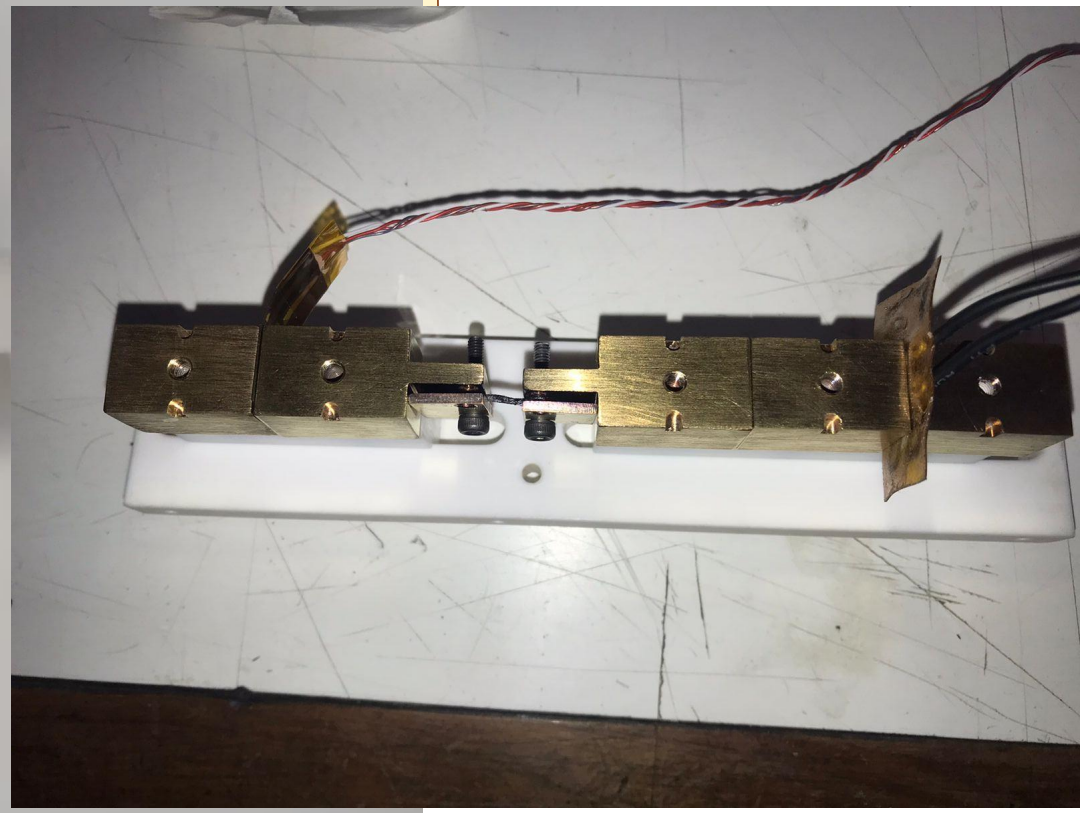
PROBLEMS FACED IN THIS MULTIDISCIPLINARY RESEARCH

Test Methodology

1.Sample Analysis Before and After Irradiation: The irradiated sample has a small area.
2.Use of Small Pieces for Various Analyses: The various analyses must use a small piece of the irradiated target sample.



lear reaction



be improved using copper double-sided adhesive tape (used for SEM sample preparation)

sensor

Kapton planar heater

centro universitário FEI

M.A. Guazzelli[1], L.H. Avanzi[1], V.A.P. Aguiar[2],, S.H. Masunaga[1], M. T. Escote[3], E.F. Chinaglia[1], K. Araki[4], M. Nakamura[4], M.M. Toyama[4], F.F. Ferreira[3], R.B.B. Santos[1], N.H. Medina[2], J.R.B. Oliveira[2], F. Cappuzzello[5,6], M. Cavallaro[6].

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"MORE CRUCIAL THAN KNOWING A BIT EVERYTHING IS KNOWING WHO EXCELS IN EACH PART"

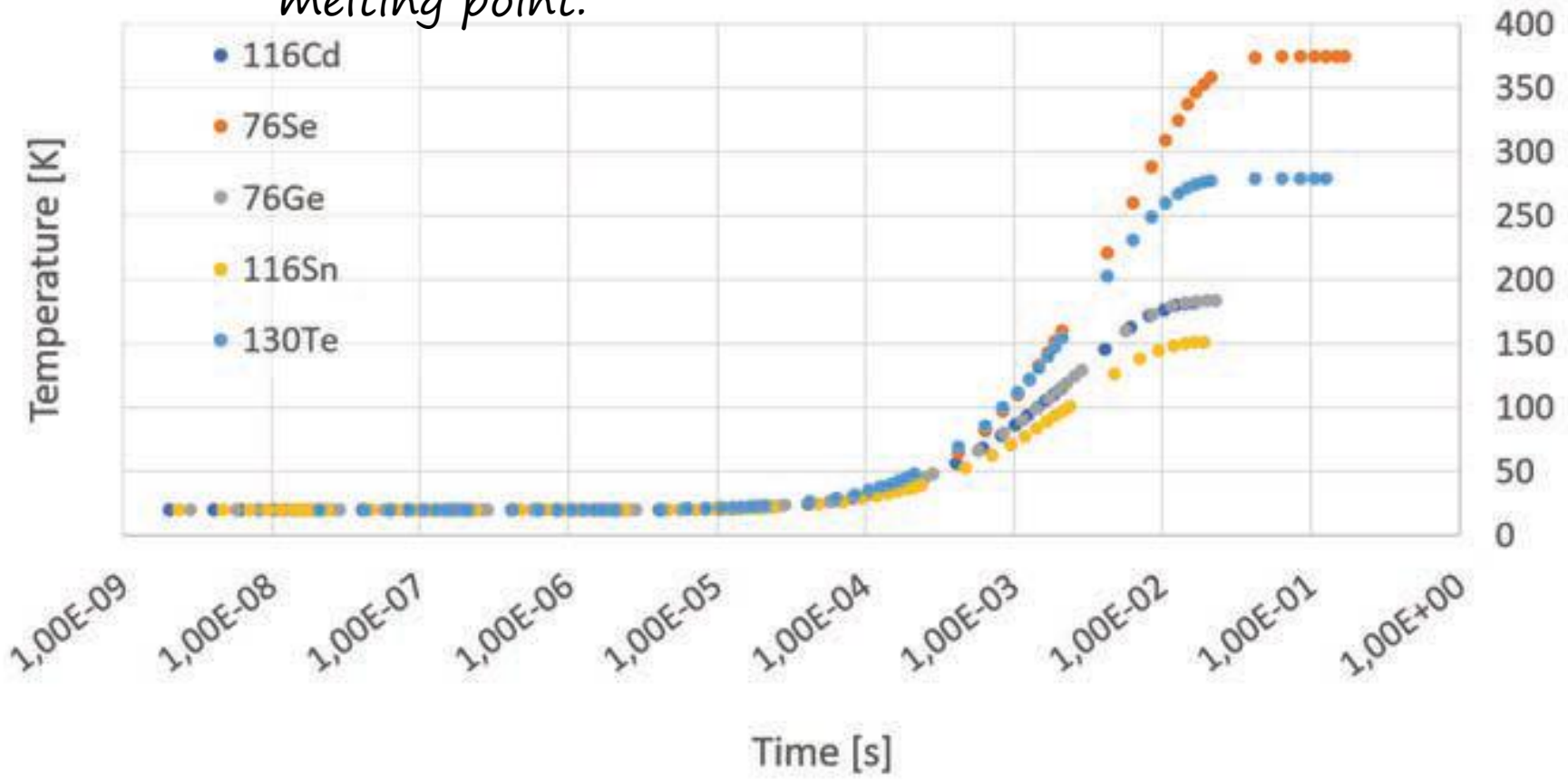
CNPq Proc.number: 408800/2021-6

Thank you very much for your attention

Marcilei Aparecida Guazzelli

centro universitário FEI

Evolution of the temperature as a function of time at the center of a 400 nm thick target of various materials irradiated by a 20 A beam with an energy of 15 MeV/u. Each target remains safely below the melting point.



In practice, the heat can be dissipated from the graphite by a cold sample holder made of copper, shown in Fig. 118, and maintained at low temperature by a cryo-cooler. The detailed numerical calculations presented above are realistic but can

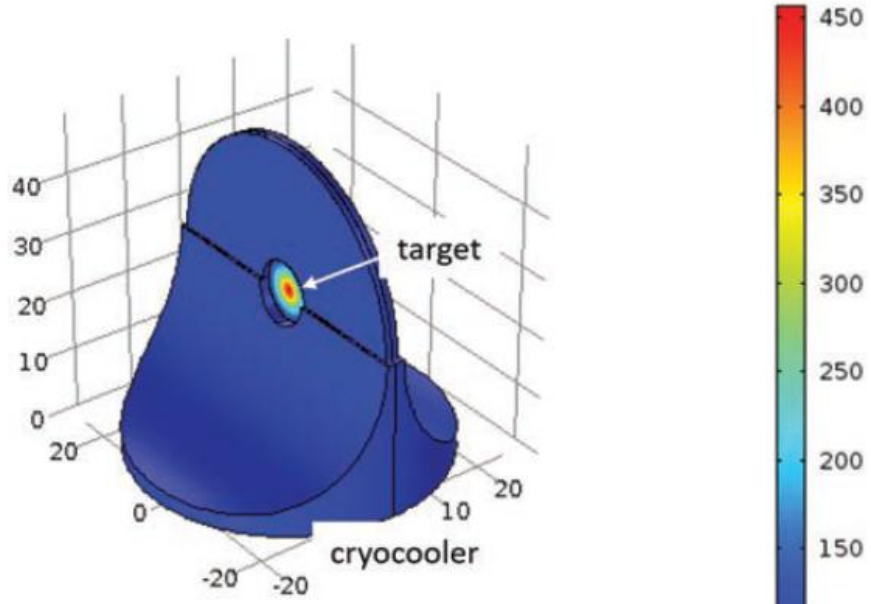


Fig. 118. (Color online) Output of a simulation performed using the software COMSOL; the copper sample holder, kept at ≈100 K, is blue-colored. A Sn film is here used as target.

Table 18. Melting and steady-state temperatures for different targets backed by a HOPG substrate under a beam with an intensity of 20 μA (∗ lowered to 13 μA for Se) and an energy of 15 MeV/u. The surface densities of the targets are also indicated.

Target isotope	^{116}Sn	^{76}Se*	^{76}Ge	^{116}Cd	^{130}Te
Surface density [μg/cm^2]	219	144	213	519	250
Beam	^{18}O	^{18}O	^{20}Ne	^{20}Ne	^{20}Ne
Melting T [K]	505	494	1211	594	722
Steady T [K]	213	432.4	249	251	381.8