Results of GSI irradiation campaign

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9th HL-LHC Collaboration Meeting, Fermilab, Batavia, US – 16.10.2019

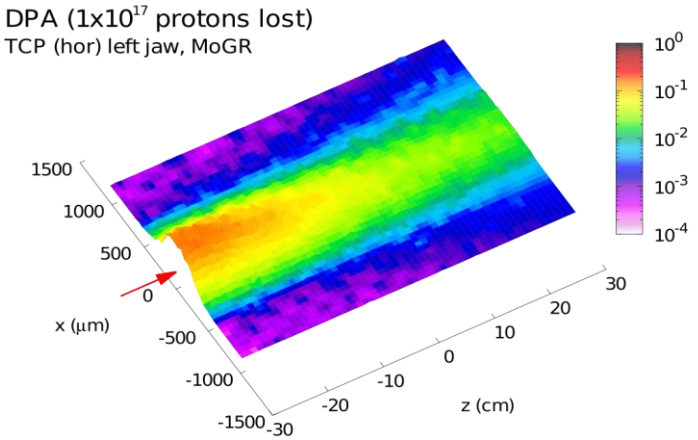
Introduction

- How to simulate irradiated materials properties after ~20 years of HL-LHC operation?
  - **Assess** DPA and H/HE production by Particle Transport simulation code (**FLUKA**)
  - Perform tests in proton and ions **irradiation facilities**, as **BLIP** at BNL, US and **UNILAC** at GSI (DE) at equivalent levels of DPA and gas production

Peak DPA in collimator materials at the end of HL-LHC lifetime (3000 fb^{-1}) ($1x10^{17}$ protons lost in collimation system)

	DPA
Mo coating	$1\text{-}3 \cdot 10^{-3}$
MoGr secondary	$4\text{-}5 \cdot 10^{-4}$
MoGr primary	Up to 0.3

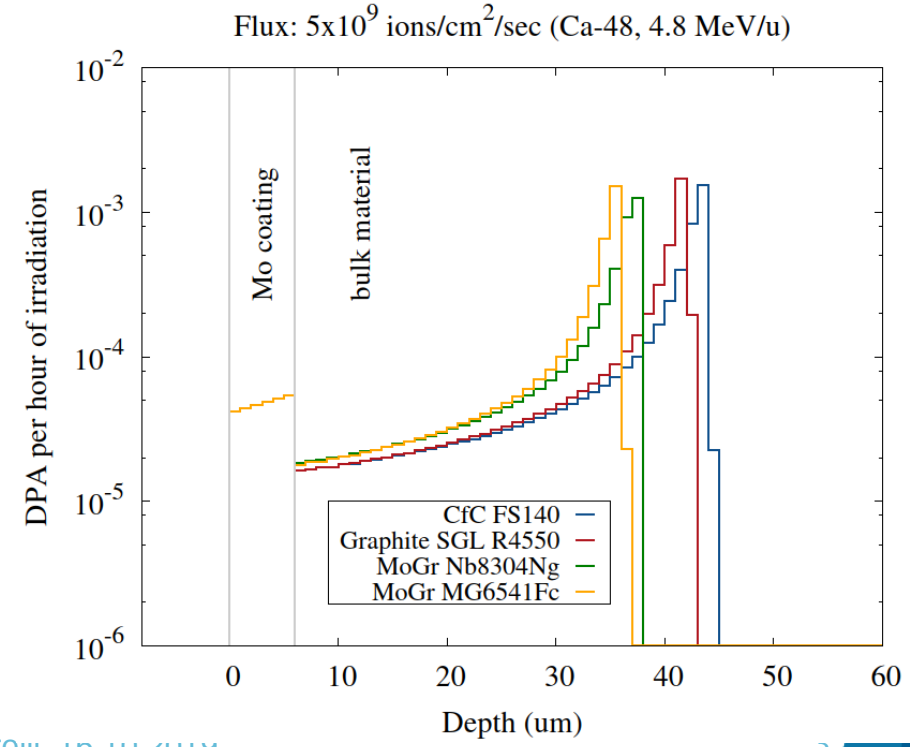
	High energy proton (BNL BLIP)	Low energy light ions (Ca/C) (GSI UNILAC)
Penetration	Deep	Superficial
Gas production	Yes	No
Activation	High	Zero-low
DPA rate	Medium	High



Irradiation Parameters

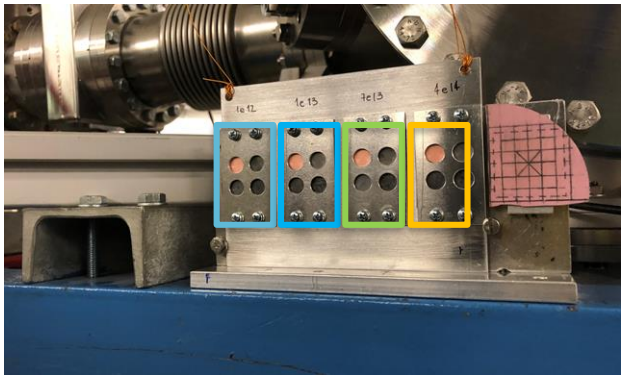
- Irradiation took place between the 27th March and the 1st April 2019 in the M3 line of the M-branch
- 113 h of beamtime granted
- Ca ions of 4.8 MeV/u to reach higher DPA
- Peak DPA in Mo coating equivalent to the one in HL-LHC after 3000 fb-1

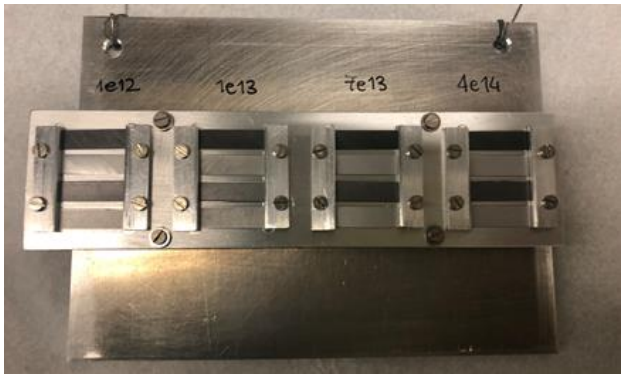
Irradiation Parameters	
DPA rate in Mo coating	**~ $5 \cdot 10^{-5}$ DPA·hr^{-1}**
DPA rate in bulk	**~ $2 \cdot 10^{-3}$ DPA·hr^{-1}**
Species	Ca48
Charge state	+10
Energy	4.8 MeV/u
Flux	5÷8.5 ion·cm^{-2}·s
Time pulse	1.8÷5.2 ms
Frequency	5 Hz
Beam spot	2.5 × 2.5 cm^2 to 2.7 × 2.7 cm^2



Experimental Setup

- Movable sample holder with 4 target stations
- Possible to irradiate 1 single station (4 materials)→ reach 4 different fluences→ relation between fluence (DPA) and properties degradation
- 2 samples geometries (optimized for Post Irradiation Examination)





Fluences [ions/cm^2]	Peak DPA coating	Peak DPA bulk
$1 \cdot 10^{12}$	$\sim 2.8 \cdot 10^{-6}$	$\sim 1.1 \cdot 10^{-4}$
$1 \cdot 10^{13}$	$\sim 2.8 \cdot 10^{-5}$	$\sim 1.1 \cdot 10^{-3}$
$7 \cdot 10^{13}$	$\sim 1.9 \cdot 10^{-4}$	$\sim 7.8 \cdot 10^{-3}$
$4 \cdot 10^{14}$	$\sim 1.1 \cdot 10^{-3}$	$\sim 4.4 \cdot 10^{-2}$

	Peak DPA collimator at 3000 fb^{-1}
Mo coating	$1\text{-}3 \cdot 10^{-3}$
MoGr secondary	$4\text{-}5 \cdot 10^{-4}$
MoGr primary	0.3 *(in limited region)*

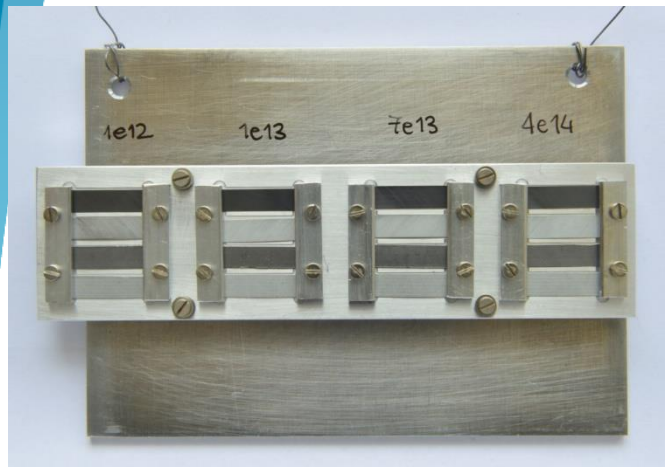
Irradiated Materials

- 8 material combinations tested
  - MoGr Nanoker - Production grade (Nb8304Ng)
  - MoGr Brevetti Bizz – HRMT-36 grade (MG6541Fc)
  - Graphite SGL R4550
  - CFC Tatsuno FS140
  - Nb8304Ng + Mo coating (DTI)
  - Nb8304Ng + Cu coating (DTI)
  - R4550 + Mo coating (DTI)
  - MG6541Fc + Mo coating (CERN)
- 5 Sample holders, including one hybrid.
- 80 samples in total
- All samples irradiated at 4 different fluences (figures in ions/cm^2)

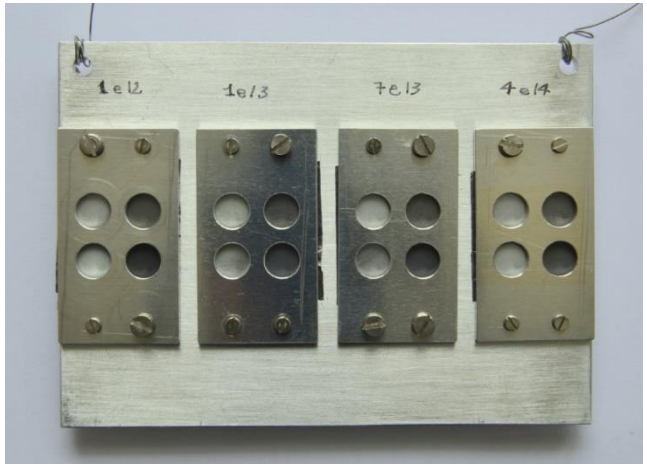
1e12	1e13	7e13	4e14

Visual inspection at the end of irradiation

HOLDER #1

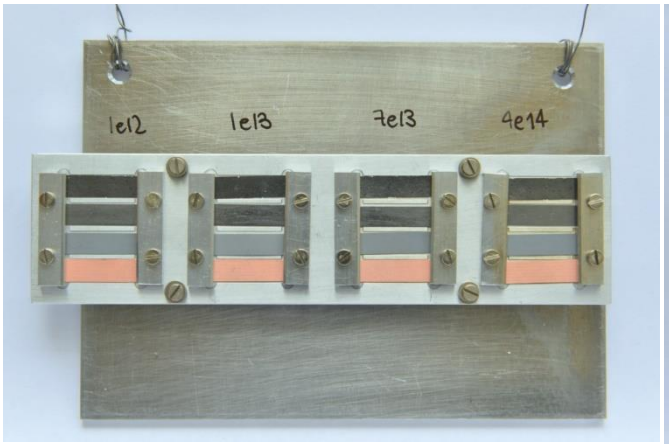


HOLDER #2

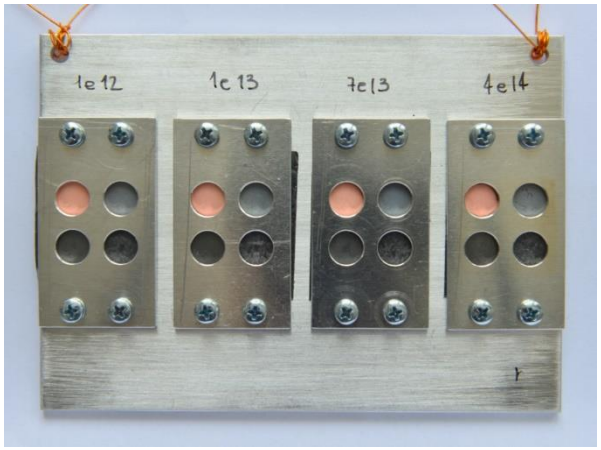


No visual signs of mechanical damage both in coating and bulk

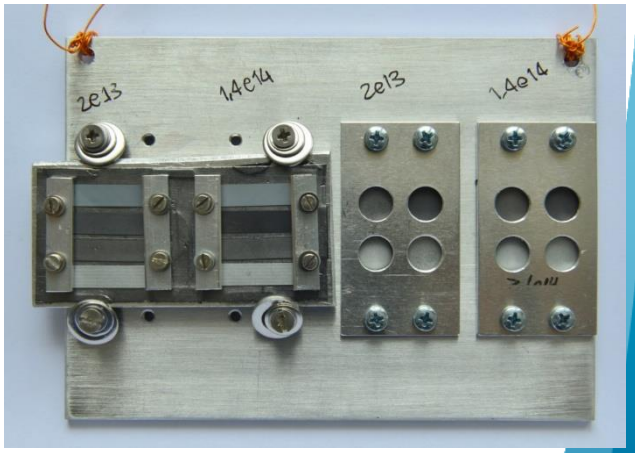
HOLDER #3



HOLDER #4



HOLDER #5



Activation after the test

- Just after the irradiation ~200 µSv/h at ~ 20cm
- Just after the irradiation (inside the box) ~31 µSv/h
- After 1 day (inside the box) ~8 µSv/h

End of irradiation

~ 2 months

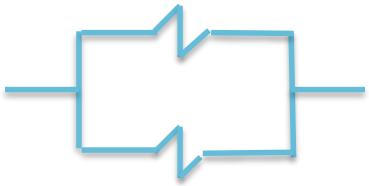
Shipping at CERN

- Sample shipped at CERN on the 11th of June (dose rate < 0.1µSv/h)
- Measurements ongoing in controlled area since the 25th of June

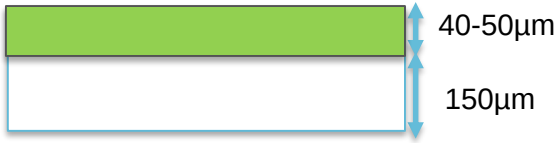
Electrical resistivity measurements

- Four probes method set-up
- Parallel resistance model (2 or 3-layers)
- Minimized sample thickness to see the contribution of the irradiated layer
- Radiation penetration depth from FLUKA simulation

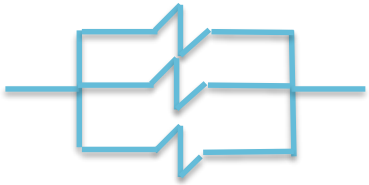
$R_{irradiated} = ?$



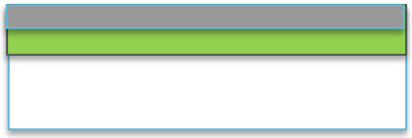
$$R_{bulk} = \rho_{pristine} \frac{L}{d_1 * t_{bulk}}$$

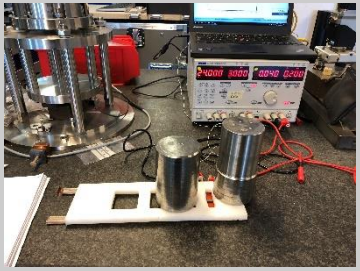


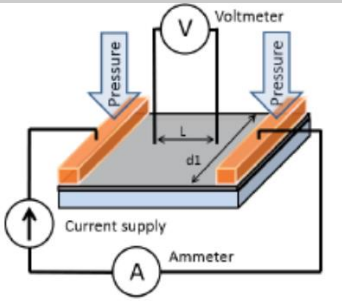
$R_{coating} = ?$

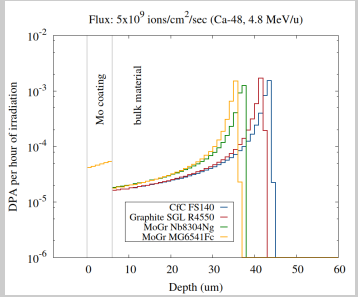


$$R_{bulk} = \frac{R_{bulk\_pristine} \cdot R_{bulk_{irrad.}}}{R_{bulk\_pristine} + R_{bulk_{irrad.}}}$$





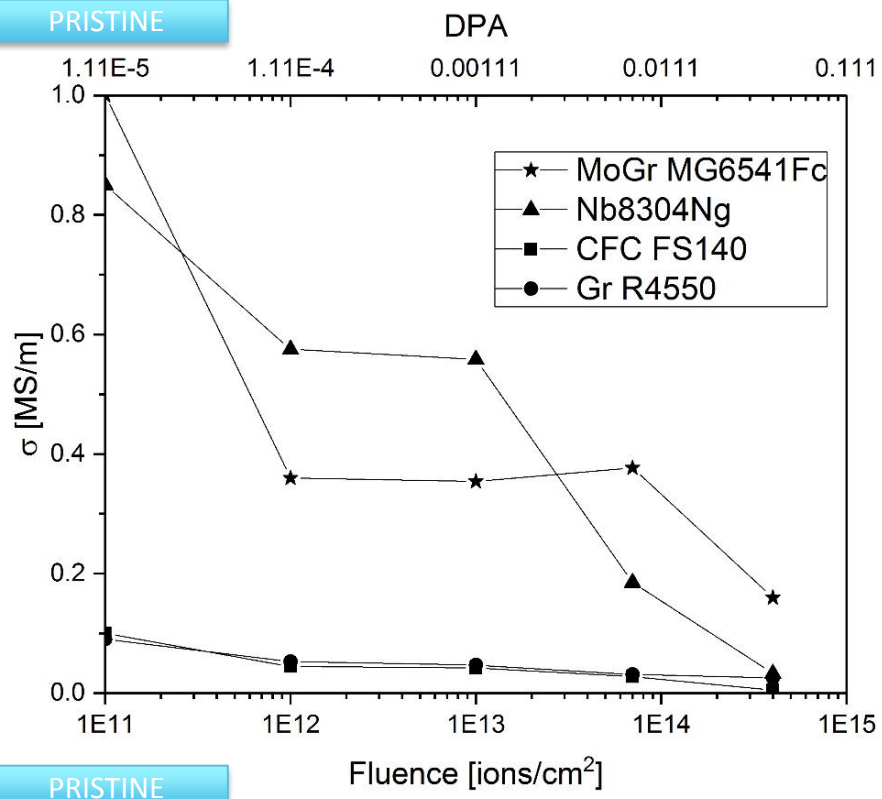




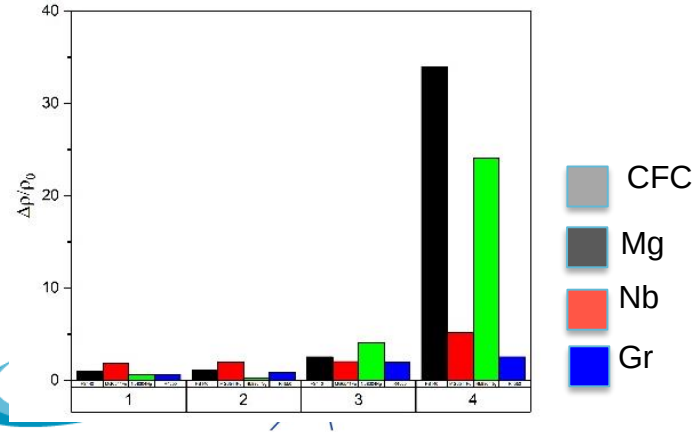
Four probe methods measurements @ Impedance meeting: https://indico.cern.ch/event/816840/

Electrical resistivity measurements-bulks

PRISTINE



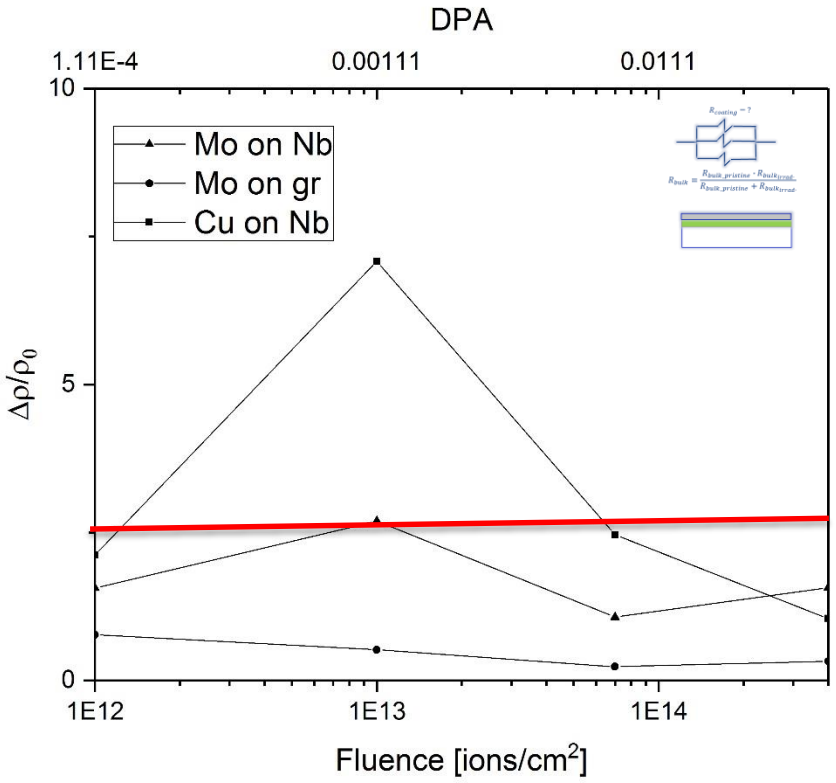
PRISTINE

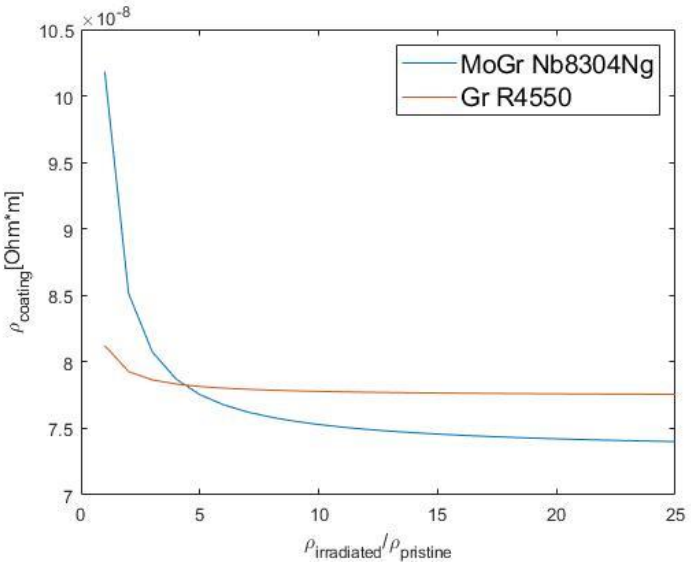


- All the materials increase their resistivity with fluence
- Graphite starts from a much more disordered state → reach saturation before MoGr (and CFC)
- Electrical conductivity in MoGr systematically higher than Gr in spite of stronger DPA effect
- Threshold effect in MoGr up to 1-7e13 ions/cm^2 → for a DPA 10 times higher then in secondary collimators the MoGr will have a conductivity ~0.6-0.7 MS/m
- MoGr with fibers more radiation resistant
- High scattering in CFC values

Electrical resistivity measurements-coatings

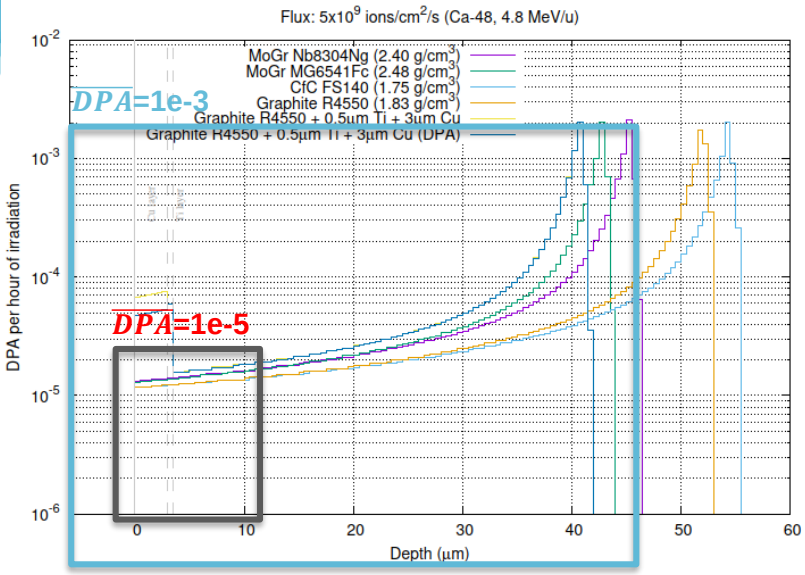
- No clear trend at increasing fluence observed
- Final values for Mo ~ 6-13 MS/m → factor 2-3 worst then pristine values
- Reason not yet explained





Impedance measurement with H011 cavity

- Uncoated samples
  - No relevant changes in the measured Q factor
  - Coherent with DC: average DPA in the layer investigated with the cavity (skin depth ~ 4µm) << then the average DPA in the whole irradiated layer



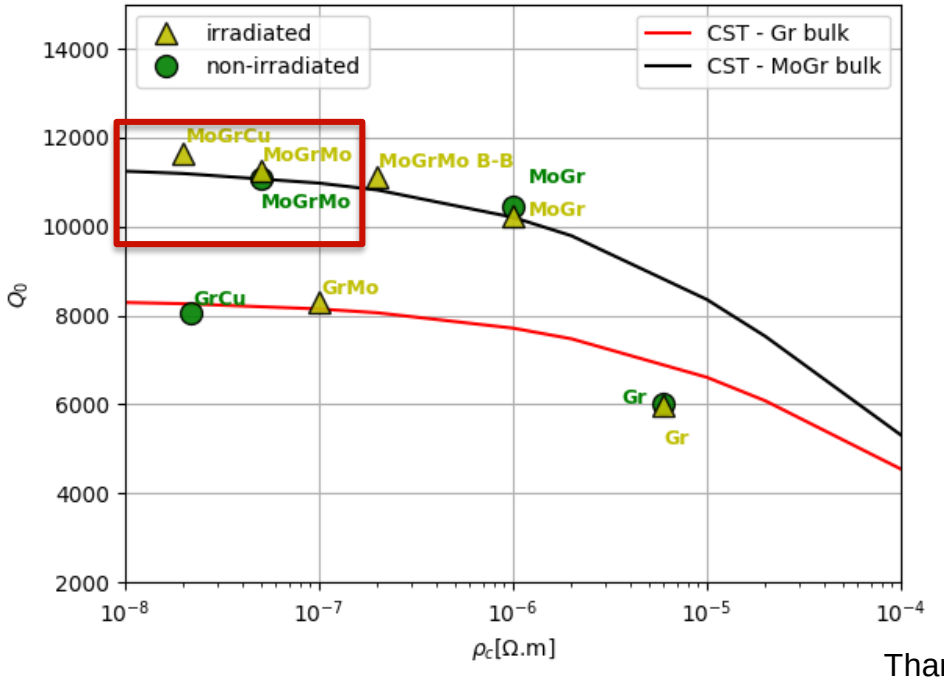
Fluence [ions/cm²]	Average DPA over 50µm (DC)	Average DPA over 10µm (H011)
1e12	5.5E-05	5.5E-07
1e13	5.5E-04	5.5E-06
7e13	3.8E-03	3.8E-05
4e14	2.2E-02	2.2E-04

→ No relevant Δσ for DPA<2e-4 : coherent with DC!

Impedance measurement with H011 cavity

- Coated samples
  - Sensitivity of the cavity too low to detect small Δσ (within the error bar of the measurements) in the region of interest (flat curve)
  - Possible improvement with the smaller cavity

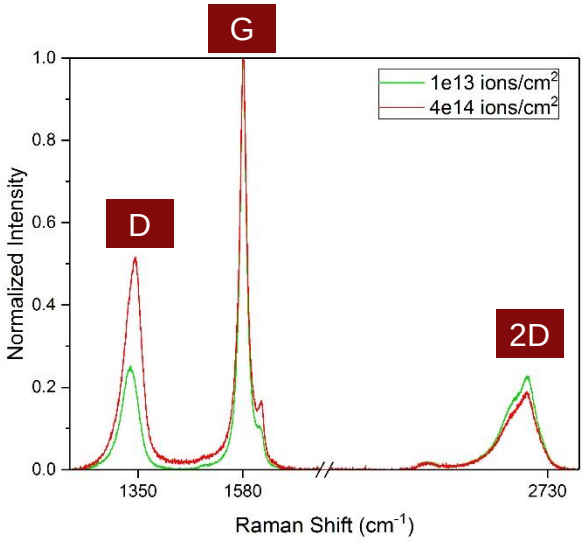
Q factor variation in function of material resistivity (coated and uncoated sample)

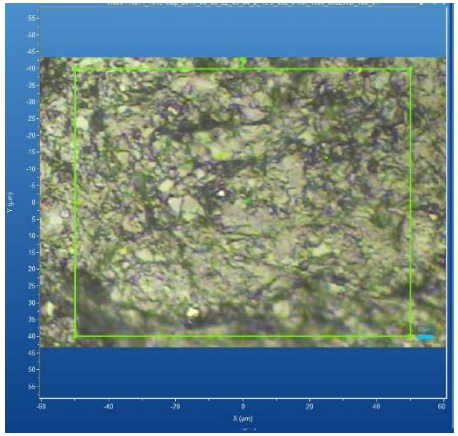


Thanks to Nicolò and Adnan!

Raman Spectroscopy of MoGr

- Penetration of the red laser (λ~632.8nm) in graphite ~100 nm→ investigation of the irradiated region only
- Mapping over 20 points to increase the statistic





Measurements performed at GSI

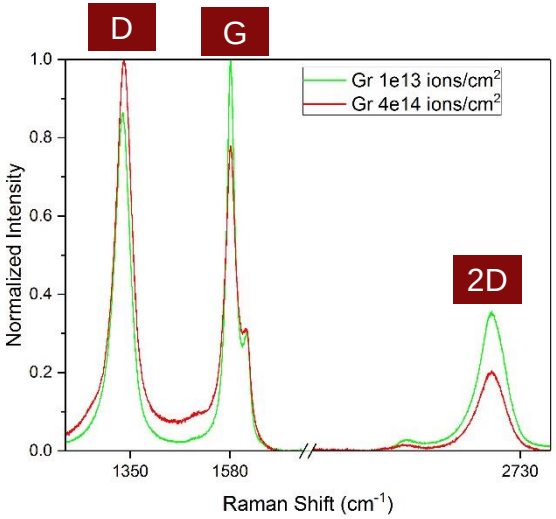
Fluence [ions/cm²]	Grain size before [nm]	Grain size after [nm]	Normalized grain reduction
1.00E+12	146	180	-26%
1.00E+13	140	150	-7.14%
7.00E+13	410	150	63.41%
4.00E+14	350	72	79.43%

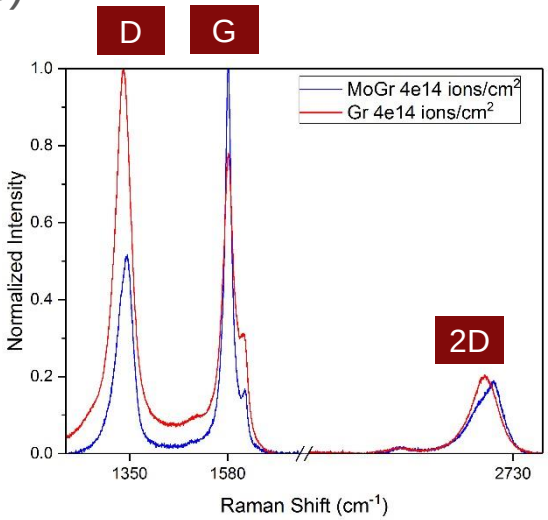
$$L_a[nm] = (2.4x10^{-10})\lambda_l^4 \left(\frac{I_G}{I_D}\right)$$

- Important change of grain at 7e13 (coherent with electrical resistivity threshold)
- Results to be cross-checked with the same device used before irradiation (different laser wavelength)

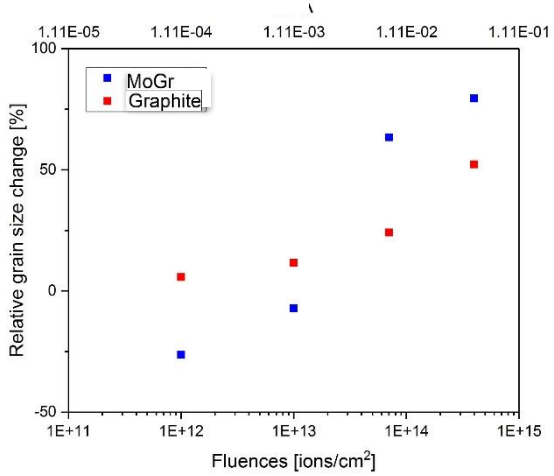
Raman Spectroscopy of Graphite

- Disorder increase with the fluences
- Grain size change less that MoGr, but final grain smaller (consistent with electrical conductivity measurements)





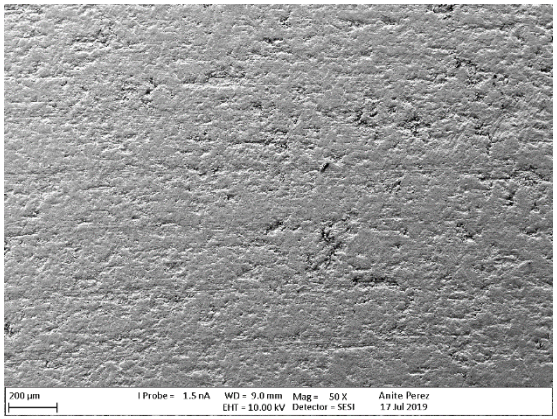
Fluence [ions/cm²]	Grain size before [nm]	Grain size after [nm]	Normalized grain reduction
1.00E+12	47	44	5.7%
1.00E+13	49	43	11.62%
7.00E+13	57	43	24.13%
4.00E+14	60	28	52.25%

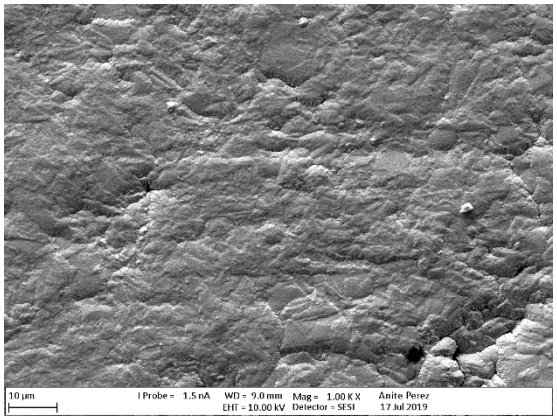


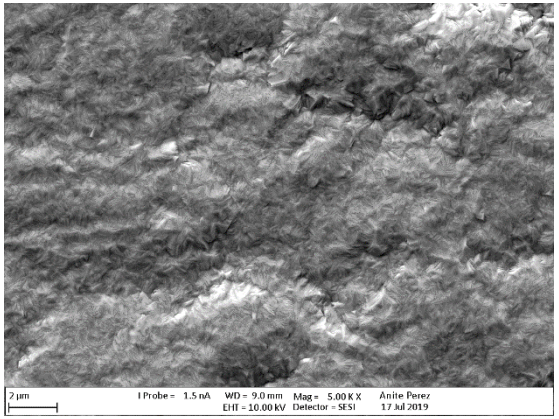
Measurements performed at GSI

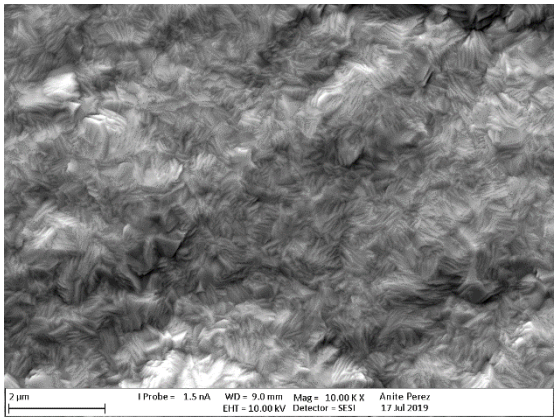
SEM Mo on MoGr

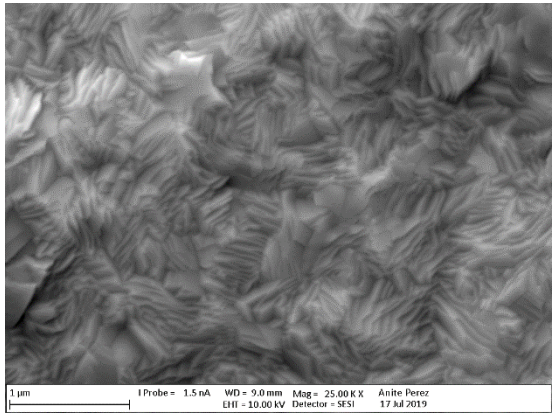
- Mo on MoGr irradiated at 4e14 ions/cm2 → Max DPA in the coating corresponding to the peak DPA in the coating after 3000 fb^{-1}









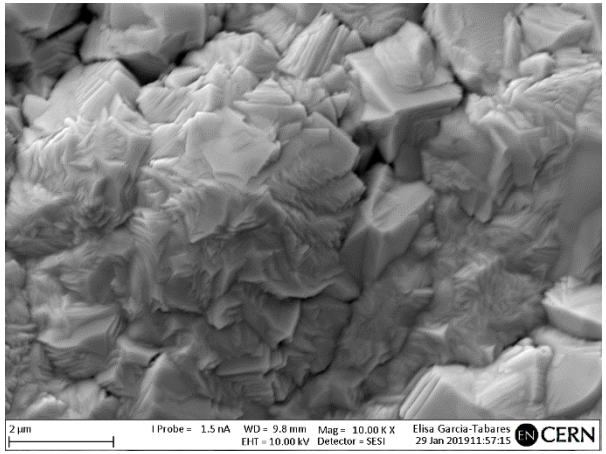


A. Peres, A. Baris (EN/MME)

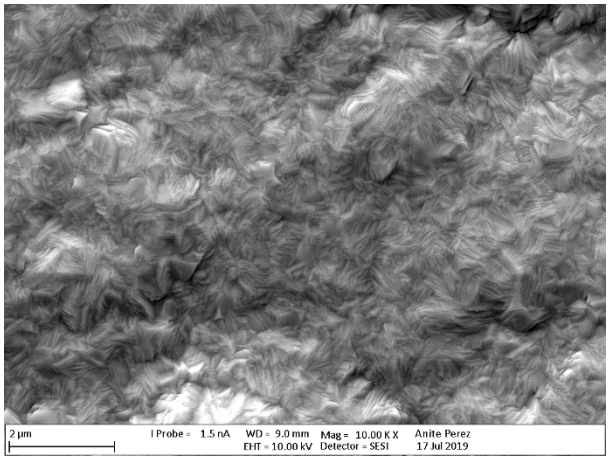
SEM Mo on MoGr

- Mo on MoGr irradiated at 4e14 ions/cm2 → Max DPA in the coating corresponding to the peak DPA in the coating after 3000 fb^{-1}

Before irradiation



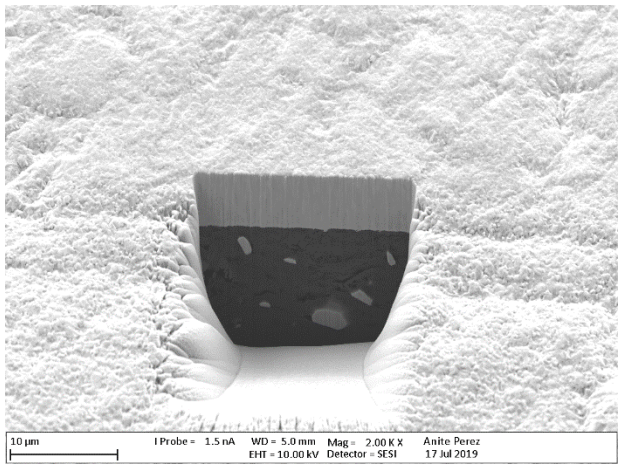
After irradiation

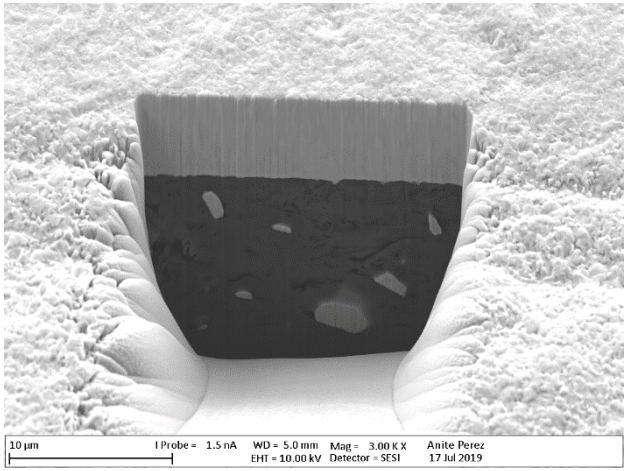


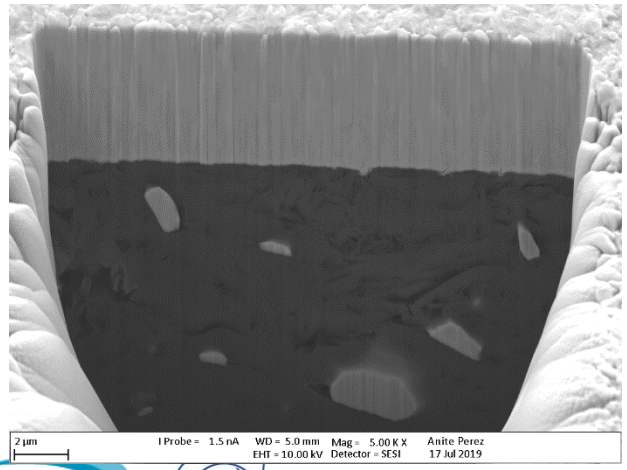
- We are comparing different samples but same bulk, surface preparation and coating process
- Irradiated sample present more discontinuous microstructure: possible indication of grain size reduction→ to be checked with XRD
- Discontinuities could explain the reduction in electrical conductivity

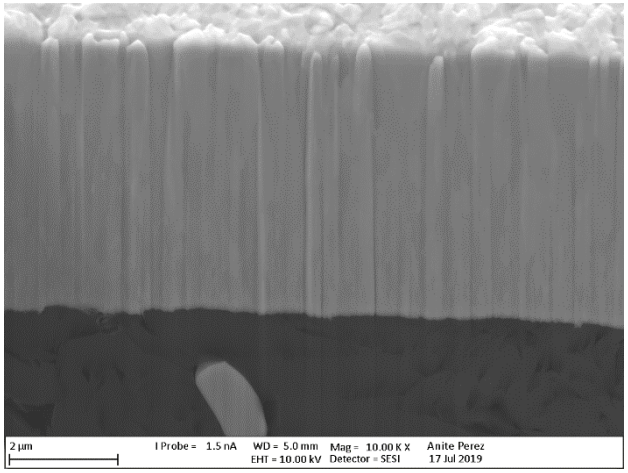
FIB Mo on MoGr

- Mo on MoGr irradiated at 4e14 ions/cm2 → Max DPA in the coating corresponding to the peak DPA in the coating after 3000 fb^{-1}





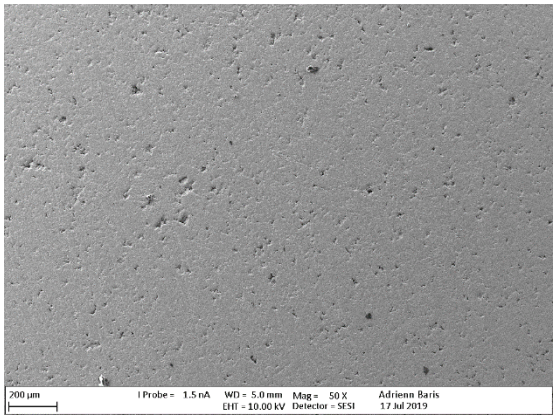


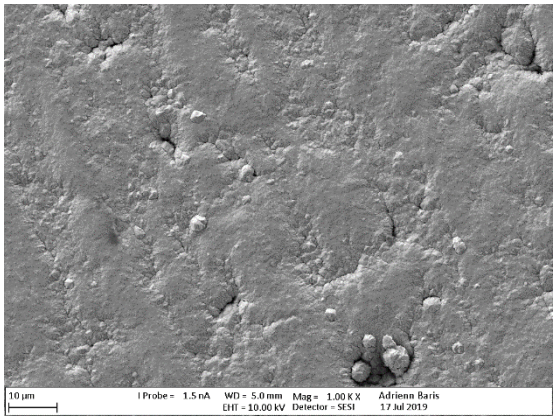


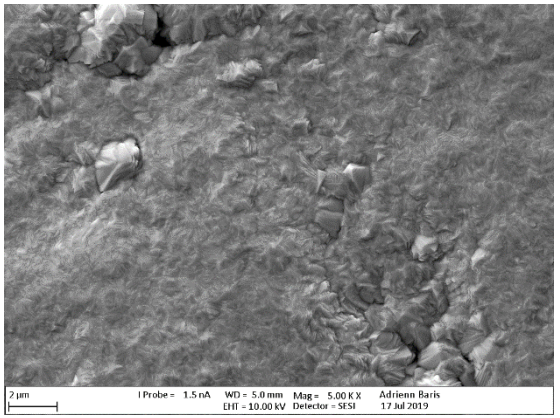
- The coating-bulk interface is continuous, no evidence of coating detachment
- No crack in the coating

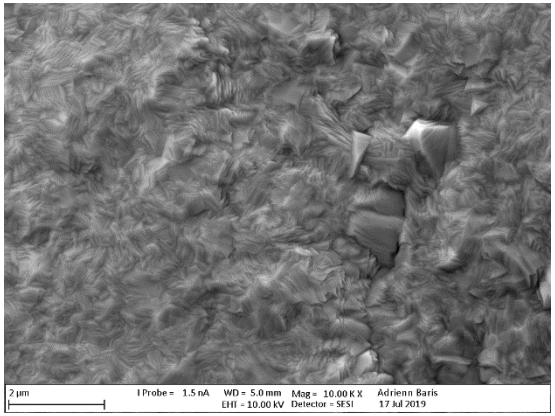
SEM Mo on Graphite

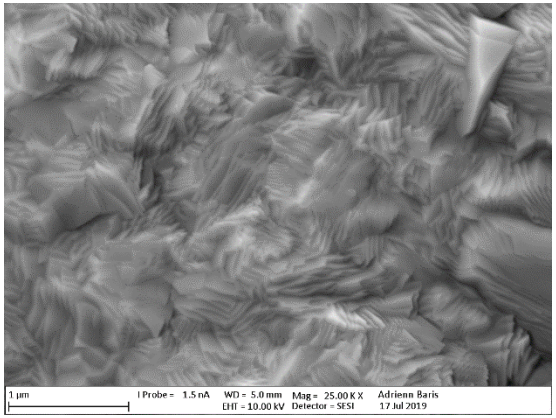
- Mo on Gr irradiated at 4e14 ions/cm2 → Max DPA in the coating corresponding to the peak DPA in the coating after 3000 fb^{-1}





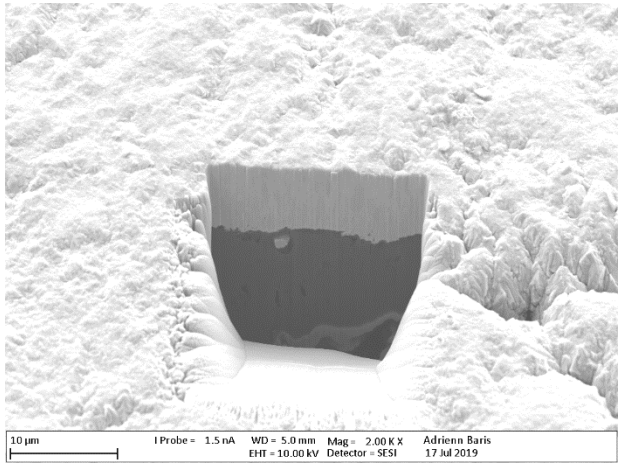


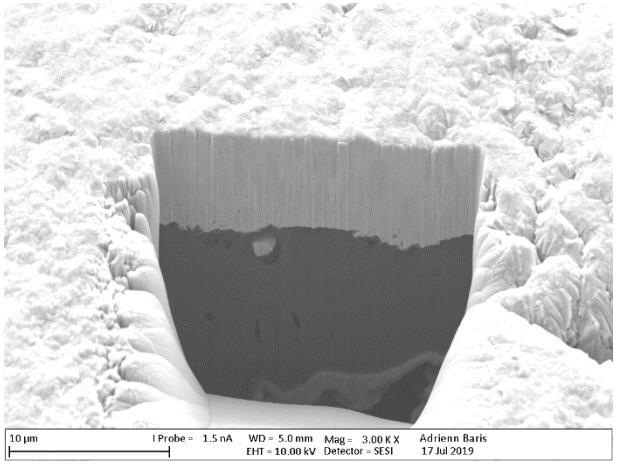


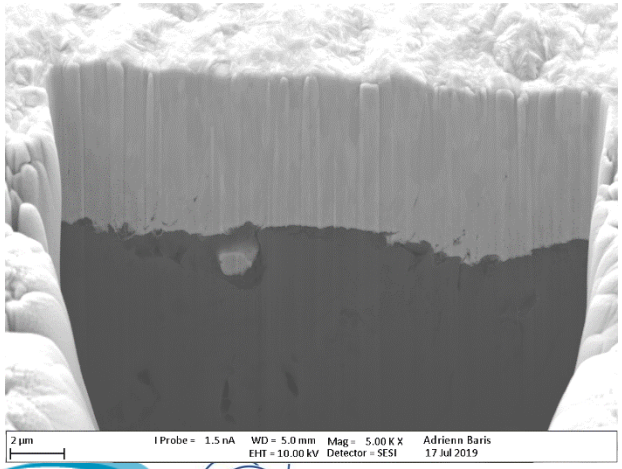


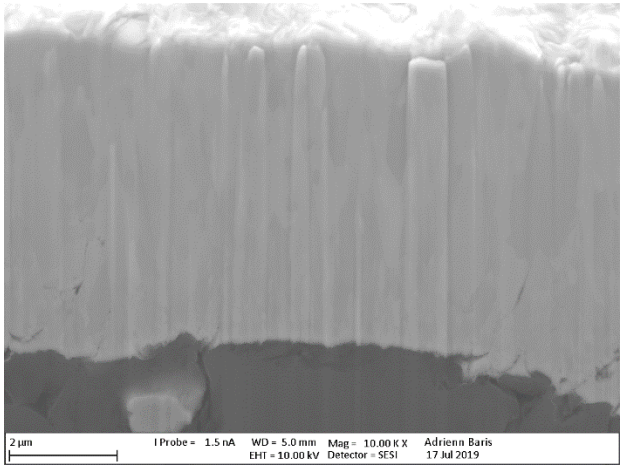
FIB Mo on Gr

- Mo on Gr irradiated at 4e14 ions/cm2 → Max DPA in the coating corresponding to the peak DPA in the coating after 3000 fb^{-1}





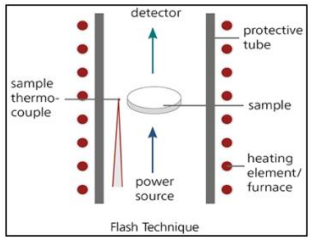


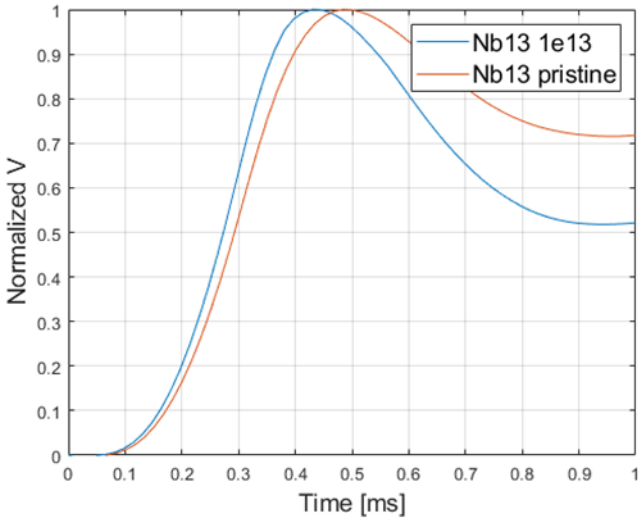


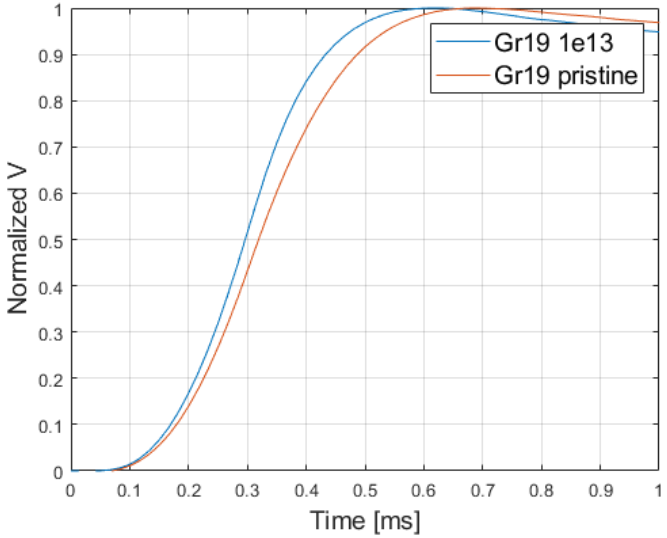
- The coating-bulk interface is continuous, no evidence of coating detachment
- No crack in the coating
- Mo diffusion in the bulk thought the porosities (probably before irradiation)

Thermal diffusivity

- 2-layers model+ reverse engineering
- Short and long measurement to study heating up and cooling down (along plane and through plane conductivity studies)







- The through-plane conductivity seems to be higher after irradiation (faster) → analysis ongoing to interpret this puzzling result

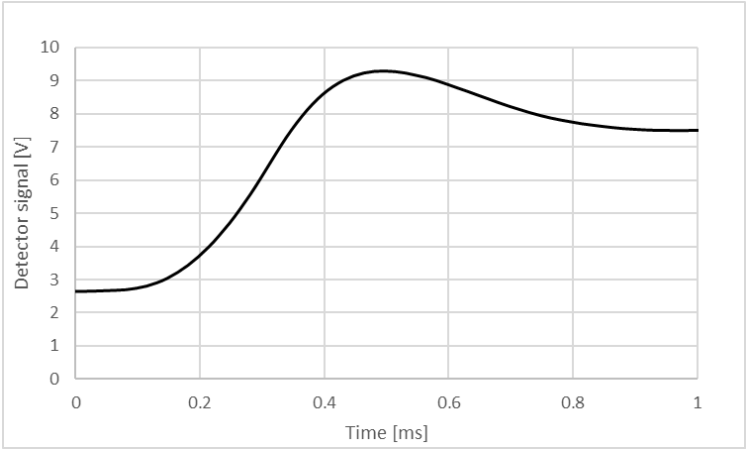
Conclusions

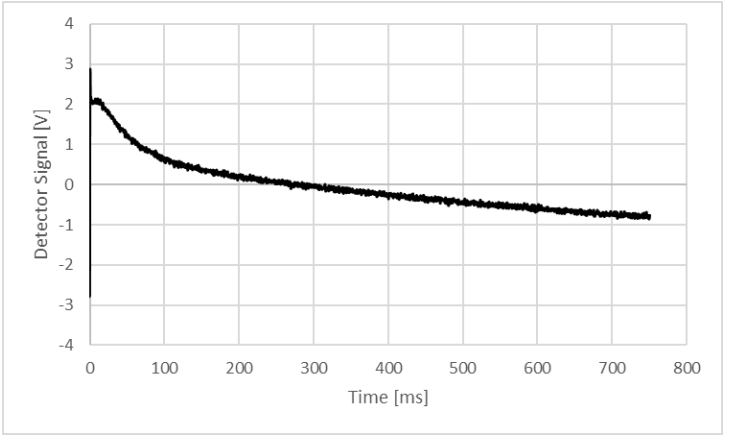
- 80 samples of different materials/coatings and size irradiated at GSI UNILAC with Ca ions
- No visible sign of radiation-induced mechanical damage on both coatings and bulk materials
- Electrical conductivity degradation in the bulk in line with expectations (higher for more ordered materials).
- Threshold effect at ~ 7e17 ions/cm^2 (8e-3 peak DPA) in bulk MoGr highlighted by electrical conductivity measurements and seeming confirmed by Raman spectrography
- Coating degradation seems to be independent of fluence (very low threshold): factor of 2 with respect to the pristine value for both Mo and Cu
- SEM and FIB investigation revealed a possible change in the coating microstructure; adherence seems good also at the maximum fluences (corresponding to DPA expected in HL-LHC)
- XRD measurement completed → data being analyzed to understand microstructural changes

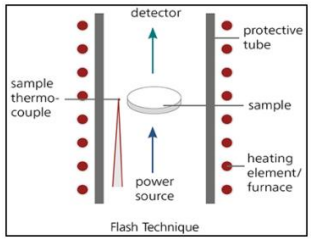
Thank you for the attention …
… and thanks Carlotta for providing the slides

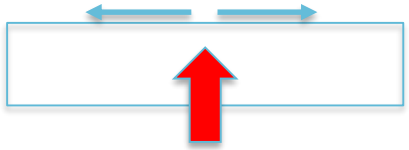
Thermal diffusivity

- 2-layers model+ reverse engineering
- Short and long measurement to study heating up and cooling down (along plane and through plane conductivity studies)









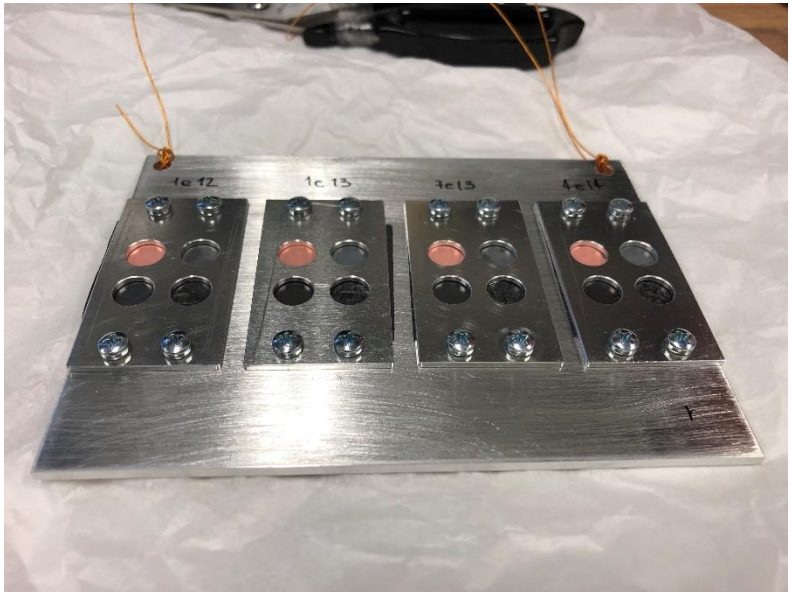
Very thin sample→ lateral conduction

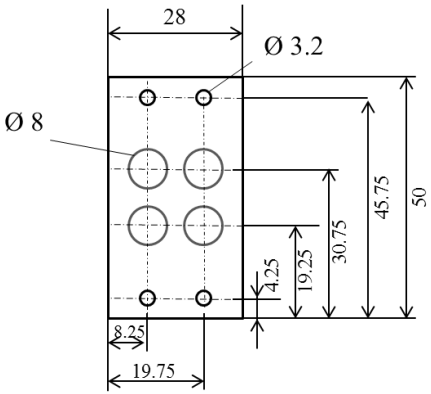
Simulation of gas production

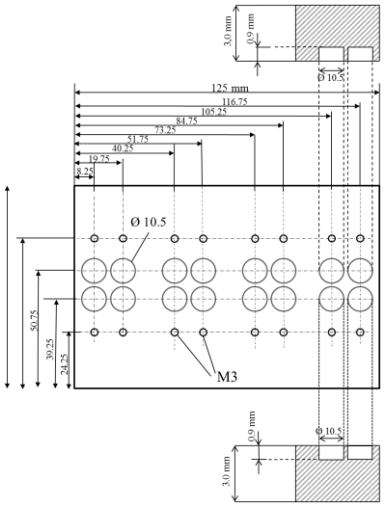
- The ion irradiation is characterized by
  - High dose rate→ conservative with respect p+
  - No gas production→ important to investigate!
- Simulate gas production with H/He implantation
  - Where?→ MIAMI (University of Huddersfield): 10keV, ~100nm
    - Possible to regulate the beam to have certain appm/DPA
    - Online TEM monitoring
    - Thermophysical characterization would not be possible in such a small layer, but a threshold for blistering can be established
  - How much?
    - FLUKA simulation (appm/DPA production)
    - Measurement of diffusion coefficient with TDS
  - With the ion irradiated samples (at different DPA) we could investigate the influence of the DPA on the gas effects

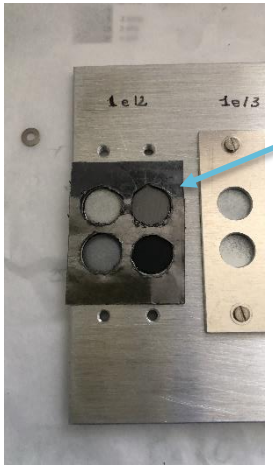
Holder description-geometry

- Disk: 10x1mm
- Geometry chosen for microscopy (FIB, XRD, Raman), indentation and adhesion









100um flexible graphite in the groove and below the mask

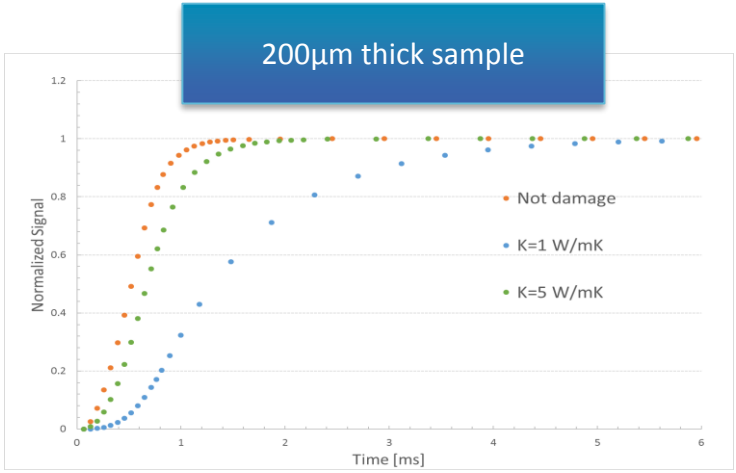
Holder description-materials

- Hybrid holder (beamtime left) to investigate intermediate fluences

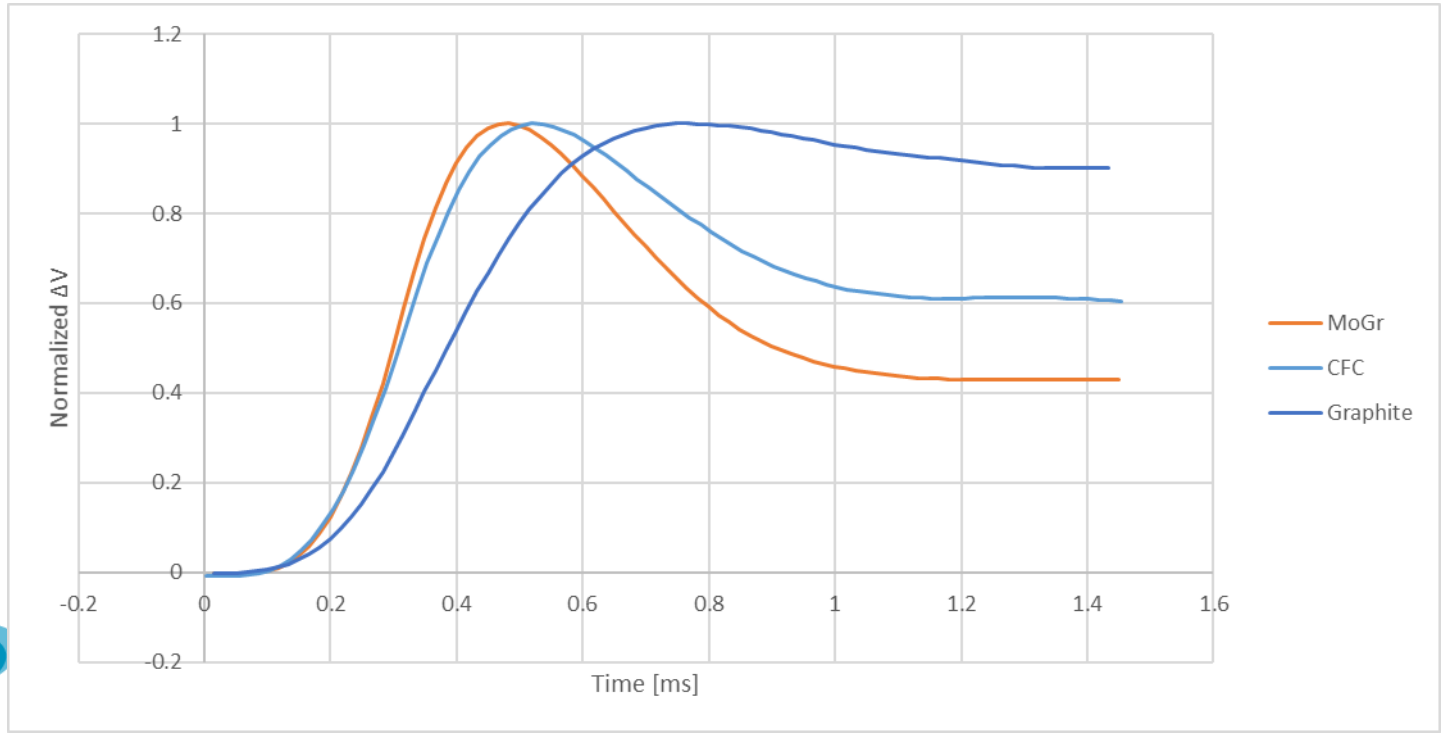
2e13	1.4e14	2e13	1.4e14

- MoGr Nanoker (Nb8304Ng)→ production grade
- Graphite SGL R4550
- Nb8304Ng+ Mo coating DTI
- R4550+ Mo coating CERN (no more spares of DTI coating)

Thermal diffusivity

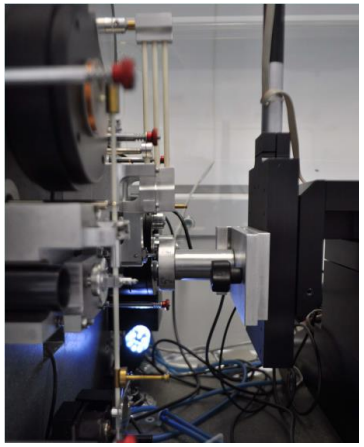


- Simulation indicates that the influence of the damaged layer should be seen with thin sample
- LFA of thin sample required reverse engineering to find the values of conductivity

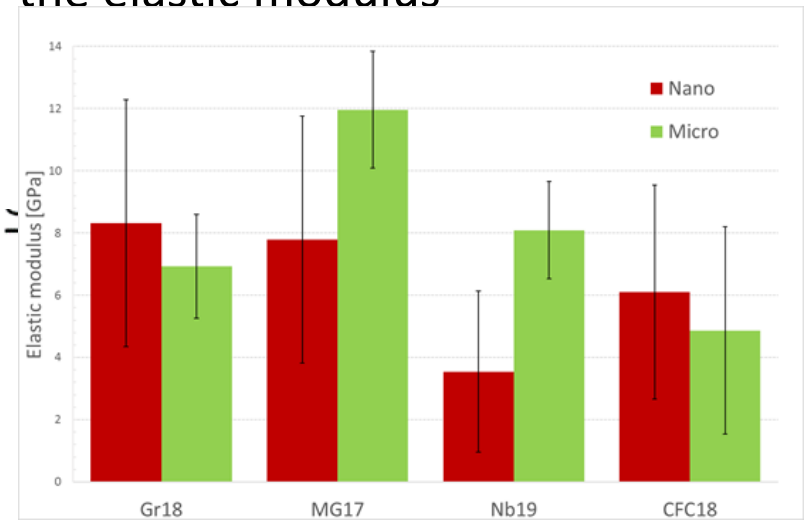


Indentation–Bulk

- Each sample indented in 25 points with penetration-controlled (nano) and load controlled (micro)
- Microindentation allows a reduction of the standard deviation→ useful to observe radiation-induced hardening and increase of the elastic modulus



Nano-Micro indentation set-up (GSI)

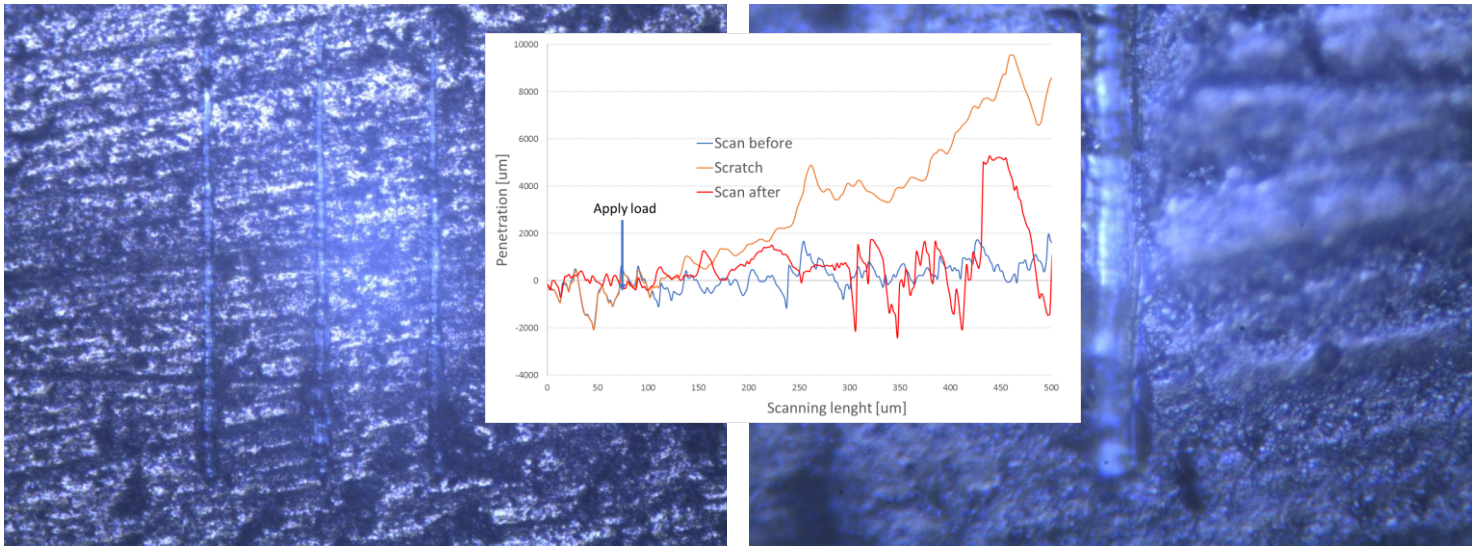


Material	Average E [GPa]	Standard deviation [GPa]	IET [GPa]
Graphite R4550	6.9	1.7	11
MG6541Fc	12	1.9	5
Nb8304Ng	8.1	1.6	4
CFC FS 140	4.9	3.3	3

- Differences with respect other method (e.g. IET) related to factors such as anisotropy and non-linearity.

Coating adhesion-Scratch test

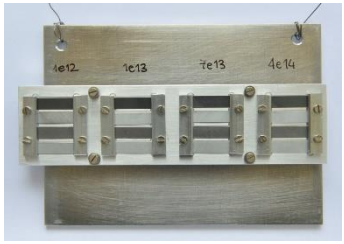
- An increasing load is applied by the nanoindenter, possible coating failure can be detected with:
  - Optical microscopy images→ difficult for material roughness and coating/bulk colours
  - Abrupt variation in the load curve→ irregularities due to roughness already present
  - Acoustic emission→ not available in the used set-up



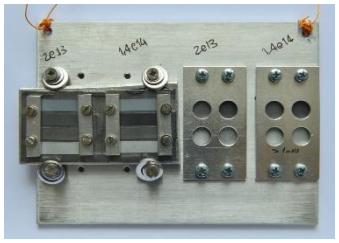
Electrical resistivity measurements

- Bulk resistivity changes in function of fluences (DPA)
- Graphite starts from a much more disordered state→ reach saturation before MoGr
- Electrical conductivity in MoGr systematically higher than Gr in spite of stronger DPA effect
- Values coherent with previous test

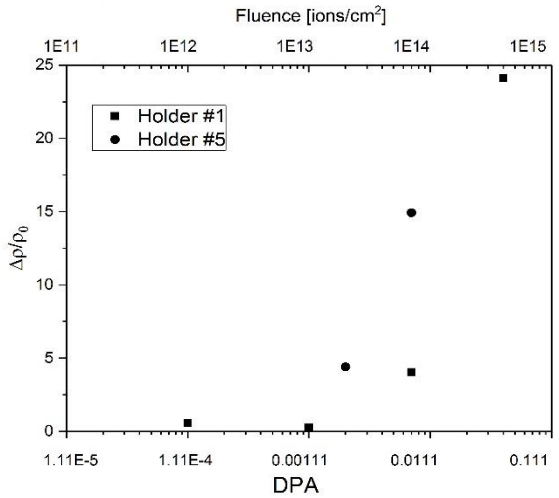
HOLDER #1



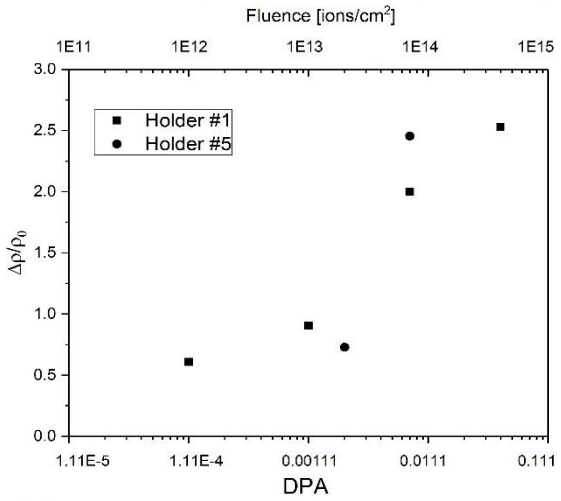
HOLDER #5

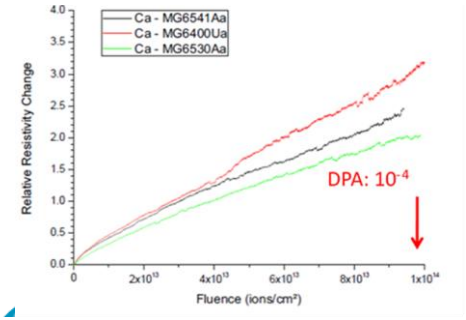


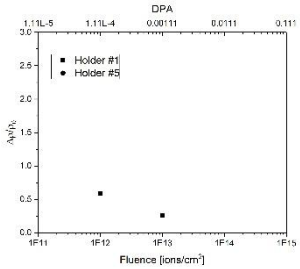
MOLYBDENUM-GRAPHITE



GRAPHITE







- For a DPA 10 times higher then in secondary collimators the MoGr will have a conductivity ~0.6-0.7 MS/m
- Important to check the area affected by 0.3 DPA in TCP

Irradiation test at GSI 2015-2016