ELI-NP Young Scientist and Young Engineer Days

• January 10th-12th, 2022 •

ELI-NP (IFIN-HH), Măgurele

TITLE:

NANOMETER THIN DIAMOND-LIKE CARBON FILMS OPTIMIZED FOR LASER TARGETS FOR ELI-NP

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OTHER TEAM MEMBERS

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INFLPR: *PhD Bogdana Mitu, PhD Veronica Sătulu, PhD Valentina Mărăscu*

LABORATORY SUPPORT

• Target Laboratory from ELI-NP •

• Plasma Processes, Materials and Surfaces Group from INFLPR •

- Motivation for laser-driven acceleration of carbon ions and protons using targets which consist in free-standing carbonic thin film
- Requirements for free-standing carbonic thin films for this application
- Suitability of diamond-like carbon
- Optimization of growth of diamond-like carbon thin films with required properties
- Characterization methods applied to check compatibility of grown films for free-standing target application
- Conclusions

MOTIVATION

Applications of **irradiation** with **ACCELERATED C/H ions**:

- testing of materials to ionizing radiation conditions.
- radiotherapy
  - due to high Linear Energy Transfer (LET) of high-energy ions/protons leading to high LET-dependent Relative Radiobiological Effectiveness (RBE). **[1] [2]**
- diagnostics and imagery. **[3] [4] [5] [6]**
- isotopes composition changing.
- generating of fluxes of neutrons and nuclei from secondary targets, subsequently usable.

LASER-DRIVEN ACCELERATION of C/H ions from carbonic solid film targets
(H from film impurities / residual composition)

⇓

Fluxes of accelerated C/H ions much higher than classical accelerators

[1] G. Milluzzo & D. Doria, et al., 'Dosimetry of laser-accelerated carbon ions for cell irradiation at ultra-high dose rate', Journal of Physics: Conference Series, vol. 1596, p. 012038, July 2020.

[2] D. Schardt, 'Tumor therapy with high-energy carbon ion beams', Nuclear Physics A 787 (2007) 633c–641c.

[3] M. Barberio, et al., 'Laser-Accelerated Proton Beams as Diagnostics for Cultural Heritage', Scientific Reports, 7:40415 (2017).

[4] H.-J. Ziock, et al., 'The proton radiography concept', LA-UR-98-1368, http://lib-www.lanl.gov/la-pubs/00460235.pdf.

[5] G. Poludniowski, et al., 'Proton radiography and tomography with application to proton therapy', BJR 2015, 88:105320150134.

[6] Lina Sheng, et al., 'Heavy-ion radiography facility at the Institute of Modern Physics', Laser and Particle Beams (2014), 32, 651-655.

MOTIVATION

Fluxes of C/H ions **without other** elements than C and H

⇓

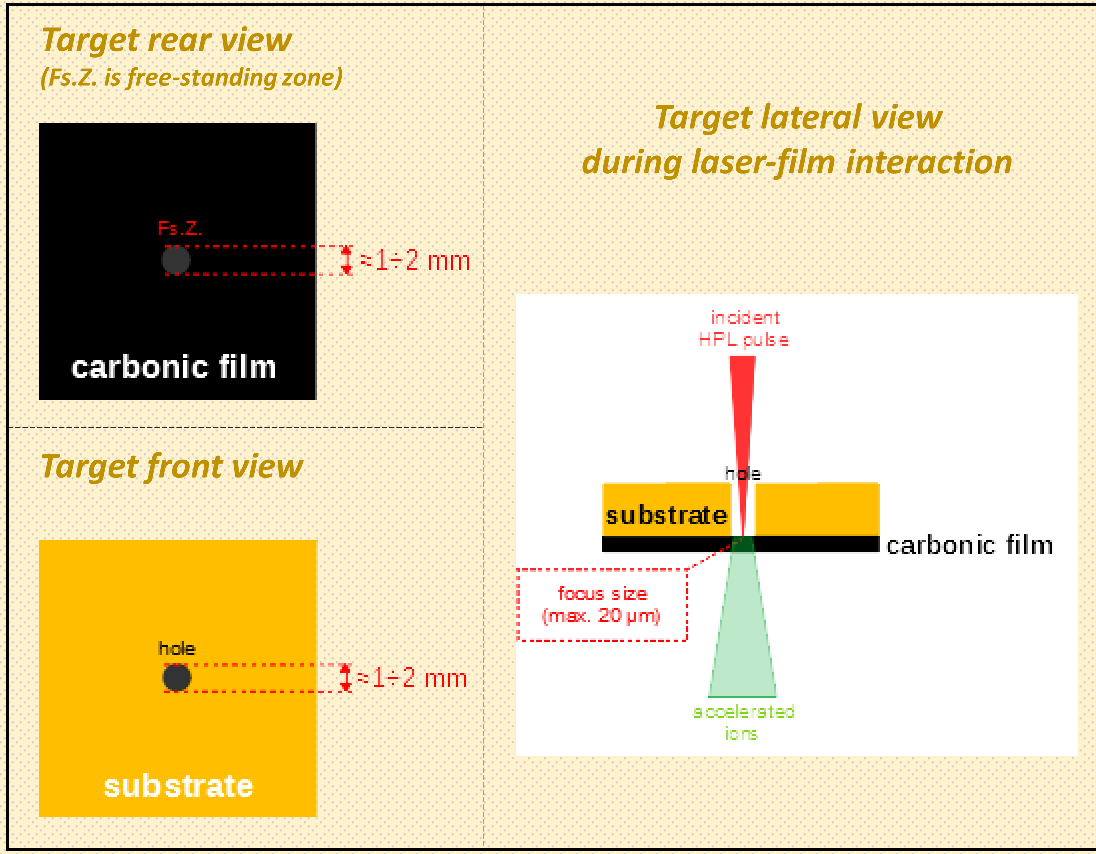
Carbonic solid film **target** **without other** elements than C and H

⇓

Target consisting by carbonic solid **film** with **free-standing** pattern on **substrate** with **holes** pattern

<=

Carbonic solid **film** will be **grown on substrate**



REQUIREMENTS

THICKNESS of carbonic target:

- From **nanometers** to **micrometers** to accelerate **C** and/or **H ions** by **two main mechanisms** or **combination of these**:
  - **RPA (ultrathin/thin films)** which allows **highest kinetic energies** of **laser-driven** acceleration technique;
  - **TNSA (thin/thick films)** which has **less requirements** than **RPA** about **laser pulse** (duration, contrast, intensity), **advantageous** in applications which **don't need high kinetic energies**.

[7] [8] [9] [10] [11] [12]

ROUGHNESS of carbonic target:

- **Bellow** of **λ/10** on surface zones which have sizes **higher** than laser spot diffraction limit **1.2λ**

λ=820 nm for **ELI-NP** HPL pulses **=>** { **λ/10=82 nm**; **1.2λ=984 nm** }

MECHANICAL STRENGTH of carbonic target:

- **High resistance** to mechanical tensile to avoid breaking by **target manipulation**, because of **high ratio aspect** of **free-standing zones**.

[7] D. Sangwan & D. Stutman & B. Diaconescu, et al., 'Simulations of carbon ion acceleration by 10 PW laser pulses on ELI-NP', Laser and Particle Beams, volume 37, issue 4, 2019, pp. 346-353.

[8] C. Scullion, D. Doria, et al., 'Polarization Dependence of Bulk Ion Acceleration from Ultrathin Foils Irradiated by High-Intensity Ultrashort Laser Pulses', Phys. Rev. Lett., vol. 119, no. 5, p. 054801, 2017.

[9] Bulanov S. S., et al., 'Optimized laser pulse profile for efficient radiation pressure acceleration of ions', Physics of Plasmas 19 (2012) 093112.

[10] D. Jung, et al., 'Laser-driven 1 GeV carbon ions from preheated diamond targets in the break-out afterburner regime', Physics of Plasmas, vol. 20, p. 083103, 2013.

[11] B. M. Hegelich, et al., 'Laser-driven ion acceleration from relativistically transparent nanotargets', New Journal of Physics, vol. 15, p. 085015, 2013.

[12] D. Jung, et al., 'Efficient carbon ion beam generation from laser-driven volume acceleration', New Journal of Physics, vol. 15, p. 023007, 2013.

DIAMOND-LIKE CARBON (DLC)

Suitable carbonic materials for such target films to C/H ions acceleration:

- Crystalline structures:

  sp^2 hybridized C

  - ***graphene***: **film** having **only ultrathin** thicknesses => **suitable only** for **some** acceleration mechanisms (if target doesn't contain a film from below carbonic materials, supplementary to graphene film)

  sp^3 hybridized C:

  - ***diamond***: **film** growth **requires very high** temperature/pressure for **one step** fabrication*)

- Amorphous structures:

  sp^3 and **sp^2** hybridized C:

  - ***DLC***: **film** growth **can be** performed at **room/low** temperature in **vacuum** conditions for **one step** fabrication,*)
    e.g. radio-frequency plasma-enhanced chemical vapor deposition (**RF-PECVD**)

*) **one step** fabrication = **synthesis** and **film deposition** in **same time**

DLC:

- **DLC** contains **sp^3** hybridized C in diamond (**cubic diamond**) and lonsdaleite (**hexagonal diamond**) **local** crystalline structures **without to form clearly separated crystallites** => **DLC** is **amorphous**

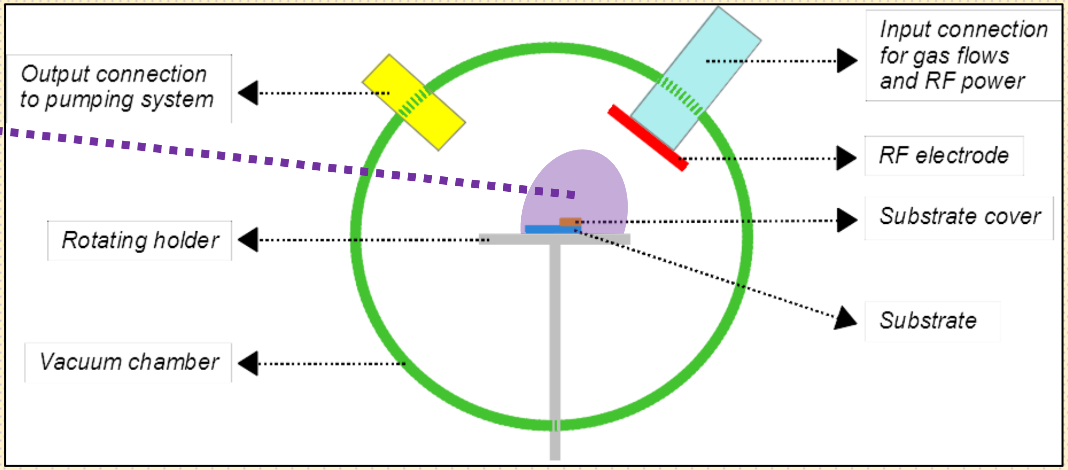
- **real DLC** contains **sp^2** hybridized C (*in graphite form*), **in addition** to sp^3 C, because of **inherent imperfections** of **growth** processes => **mechanical characteristics** of **DLC** are **worsened if graphite content is too high**, **relative to requirements** of DLC **specific application** => **growth process optimization to reduce** sp^2 C content

DLC GROWTH

RF plasma ions:
$[CH_3]^{n+}$, $[CH_2]^{n+}$, $[CH]^{n+}$, $[C]^{n+}$,
$[H_2]^{+}$, $[H]^{+}$, $[Ar]^{n+}$

- **Carbon** from **plasma constituents** is deposited, forming **sp^3 & sp^2 carbonic** structures in film.
- Hydrogen **more preferentially etches sp^3** than **sp^2 carbon**, from film.
- Globally, **sp^3** structures **are favorized to grow**
- **More** hydrogen content in plasma leads to **more sp^3 C** concentration in film (better **DLC**).

[13]



DLC growth experimental set-up (RF-PECVD)

substrate cover => **portion** of substrate **uncoated** by **DLC =>** *<u>determination of DLC film thickness</u>*

[13] Peter K. Bachmann, et al., 'Towards a general concept of diamond chemical vapour deposition', Diamond and Related Materials, vol. 1, issue 1 (1991) pp. 1-12.

DLC GROWTH

Clean silicon wafers as substrates **=>**
=> Very smooth substrates **=>**
=> Worsening of **DLC film** roughnesses by **substrates** roughnesses **is excluded =>**
=> IF such **worsening occurs**, **find** of **cause will focus** on **deposition** process

METHOD 1			
Precursor gas mixture	CH_4 + Ar	**Total pressure**	*~10^{-3} mbar*
C:H ratio	***1:4***	**RF power**	*100 W*
Gas flows	*25 sccm CH_4* ***50 sccm Ar***	**Deposition times**	***200 min ;*** ***300 min***

METHOD 2			
Precursor gas mixture	CH_4 + H_2	**Total pressure**	*~1.5×10^{-3} mbar*
C:H ratio	***1:9 (increased H content)***	**RF power**	*100 W*
Gas flows	*25.0 sccm CH_4* ***62.5 sccm H_2***	**Deposition times**	***23 min ;*** ***34 min ;*** ***45 min ;*** ***60 min***

DLC GROWTH

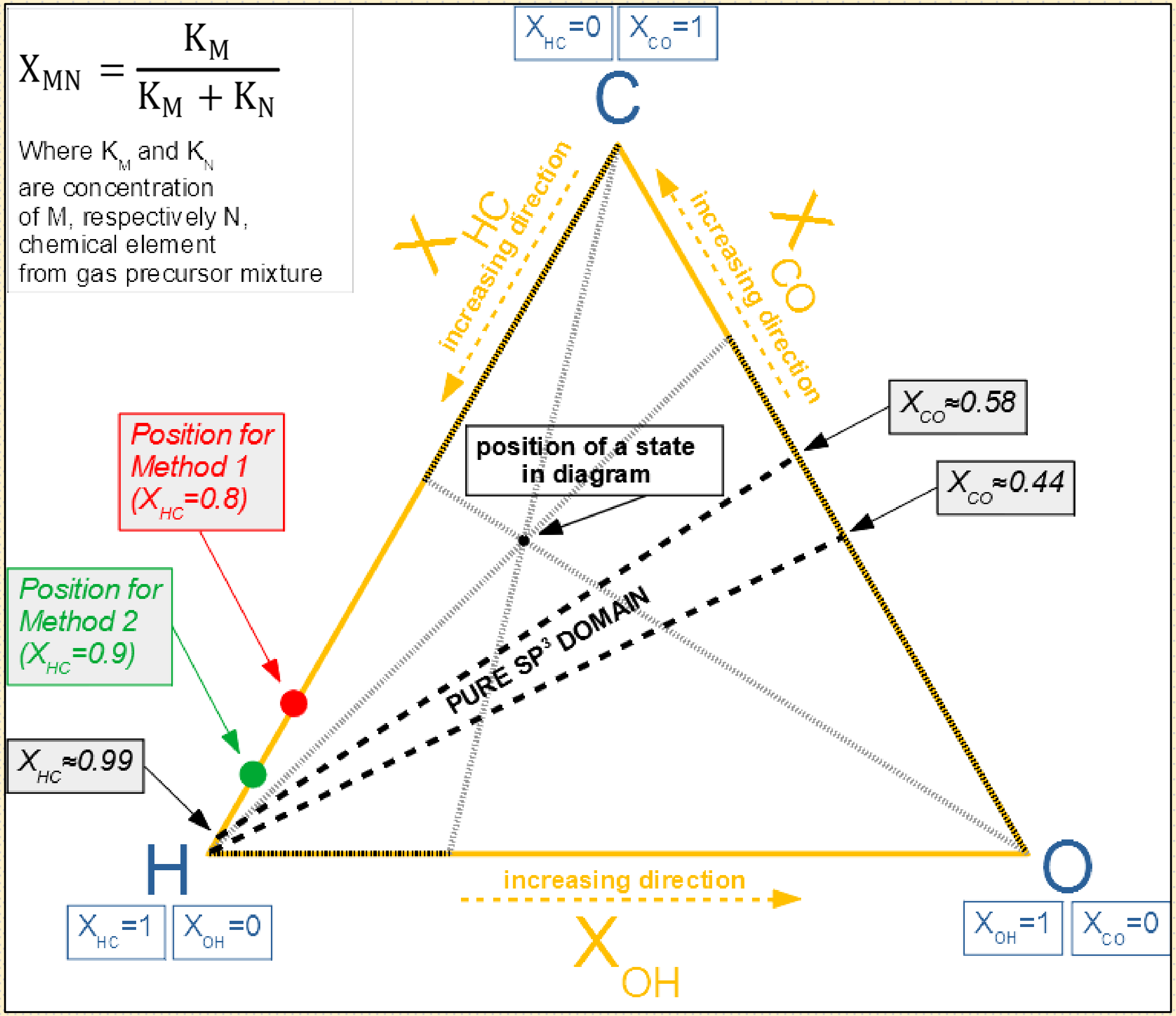
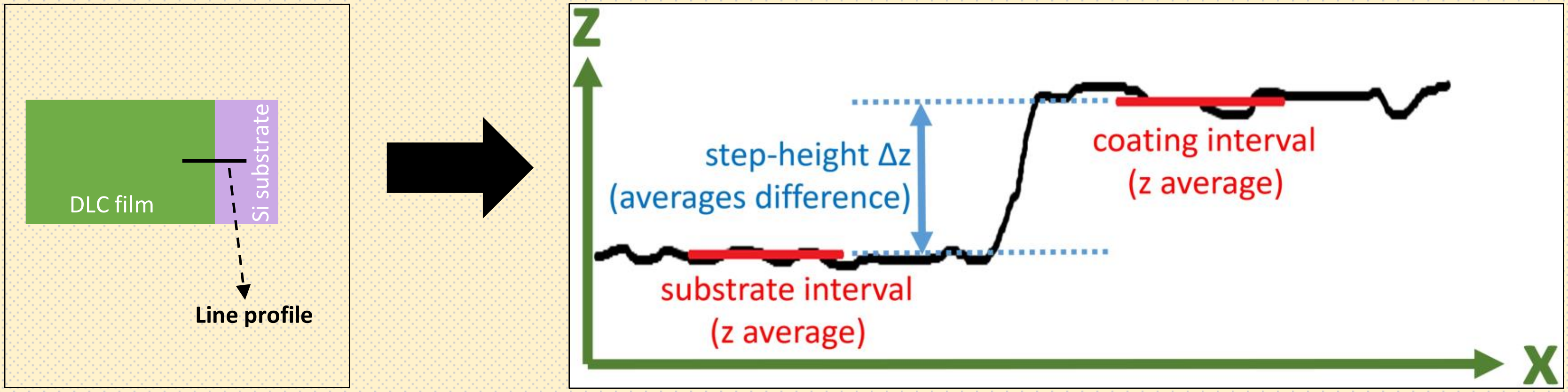


Diagram of pure sp³ deposited carbonic films,
related to C/H/O-composition of precursor gas mixture, for CVD processes **[13]**

[13] Peter K. Bachmann, et al., 'Towards a general concept of diamond chemical vapour deposition', Diamond and Related Materials, vol. 1, issue 1 (1991) pp. 1-12.

DLC CHARACTERIZATION

CONTACT (MECHANICAL) PROFILOMETRY



• *Step-height procedure for thickness determination* •

METHOD 1 SAMPLES		
Deposition time Δt [min]	**200**	**300**
Thickness Δz [nm]	381.7	535.5

Δz(Δt) linear fit

Deposition rate (slope)
1.8 nm/min

METHOD 2 SAMPLES				
Deposition time [min]	**23**	**34**	**45**	**60**
Thickness [nm]	21.8	26.3	50.6	57.0

Δz(Δt) linear fit

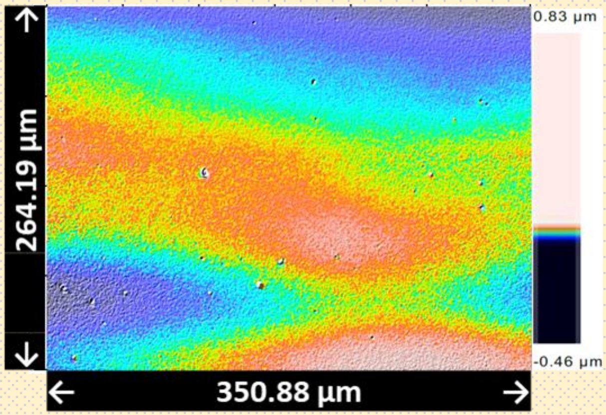
Deposition rate (slope)
1.0 nm/min

DLC CHARACTERIZATION

METHOD 1 samples

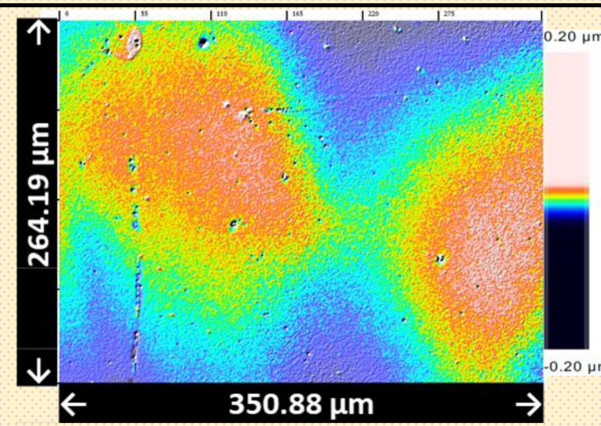
200 min deposition time

roughness **S_q=18.6nm**



300 min deposition time

roughness **S_q=14.8nm**



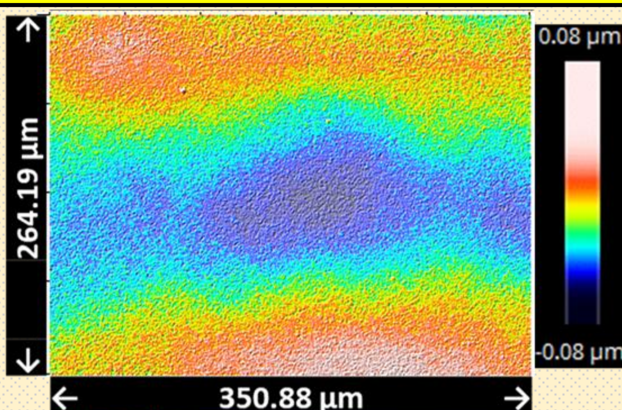
HPL focus spot diffraction limit: 1.2λ=0.98 µm

OPTICAL PROFILOMETRY → Roughness determination → Placement under of λ/10=82 nm

METHOD 2 samples

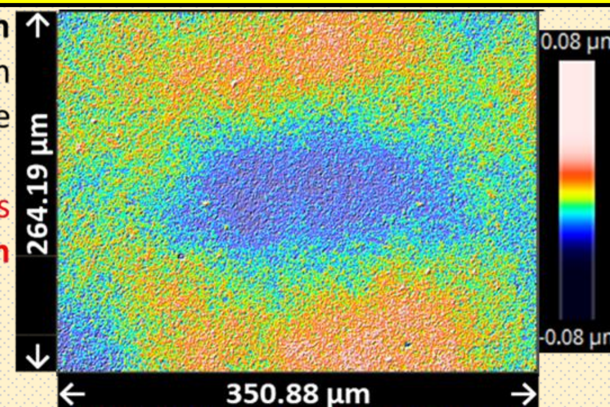
23 min deposition time

roughness **S_q=2.9 nm**



45 min deposition time

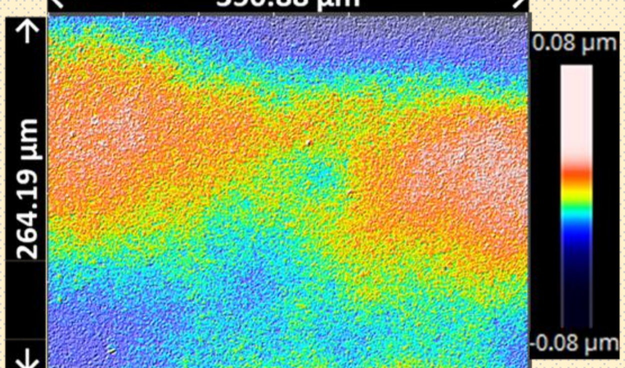
roughness **S_q=1.6 nm**



HPL focus spot diffraction limit: 1.2λ=0.98 µm

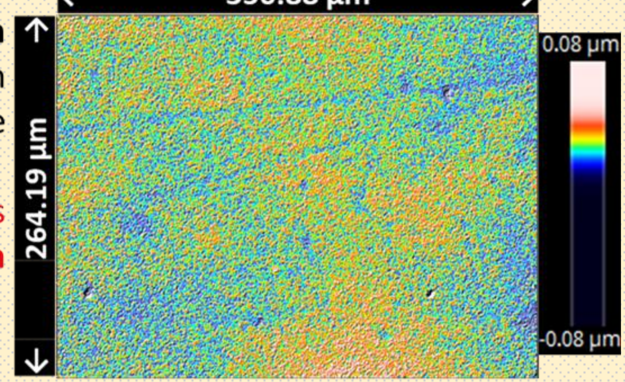
34 min deposition time

roughness **S_q=2.7 nm**



60 min deposition time

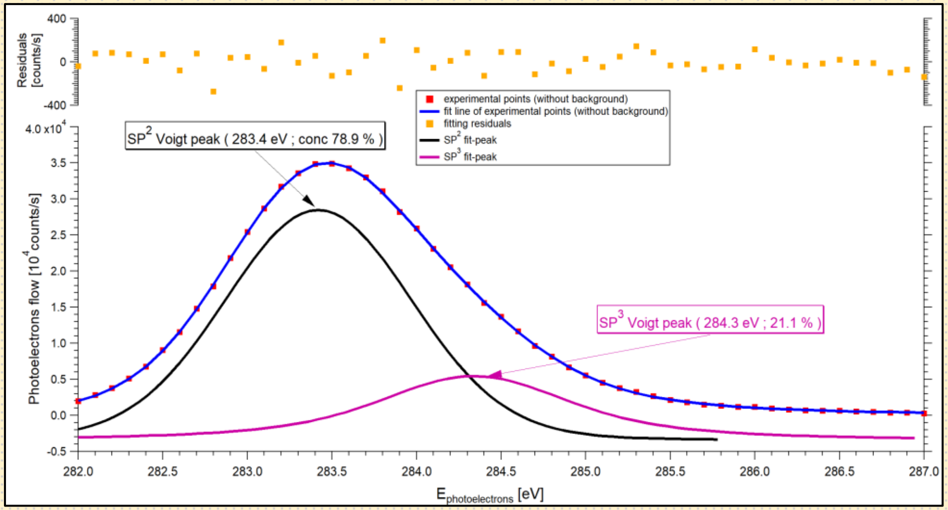
roughness **S_q=1.9 nm**



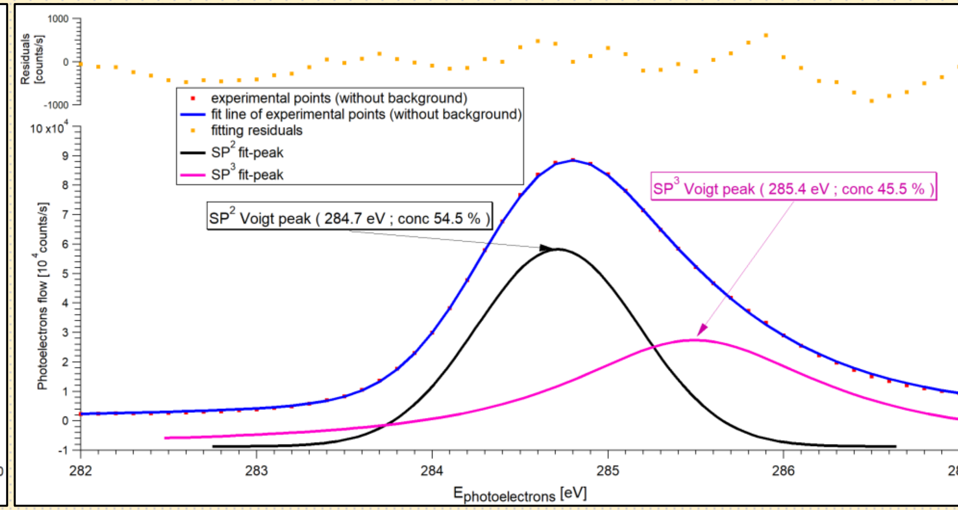
DLC CHARACTERIZATION

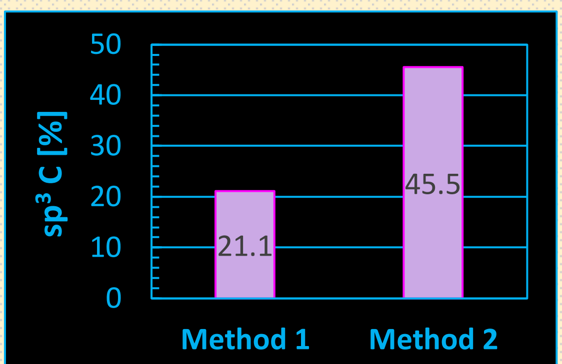
X-RAY PHOTOEMISSION SPECTROMETRY (XPS)

DLC film from **method 1** deposition
(*XPS spectra of C-C bonds*)



DLC film from **method 2** deposition
(*XPS spectra of C-C bonds*)





Improving of DLC film **mechanical strength** by **method 2**, comparing to **method 1**

CONCLUSIONS

CONCLUSION

Characterization data *shows* ***compliance*** *of grown DLC films with* ***HPL target requirements****:*

- *λ/10 limit of roughnesses on large area (≈92700 μm^2)*
- *sp^3 C content assures mechanical strength for application concerned*

NEXT STEPS

- ***Implementation*** *of developed growth method on* ***copper*** *substrates (much cheaper than silicon wafers)*
- ***Patterned etching*** *of copper substrates* ***to achieve free-standing zones*** *of deposited DLC film*
- ***HPL acceleration*** *of* ***carbon*** *&* ***hydrogen*** *ions using* ***DLC*** *targets*

THANK YOU

FOR ATTENTION

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