Selecting the proper substrate for targets in NUMEN Experiment

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Majorana 2023 Workshop
July 12-14 2023, Modica

Outline

- Introduction
- Raman measurements
- Evaluating the density
- Evaluating the thermal conductivity
- Thickness uniformity
- Conclusions

NUMEN: a Double Charge Exchange(DCE) Experiment

The NUMEN collaboration is interested to extract the **Double Charge Exchange** nuclear matrix elements to put constraints on **neutrino-less double beta decay matrix elements (the theoretical work done demonstrated that they are proportional)**

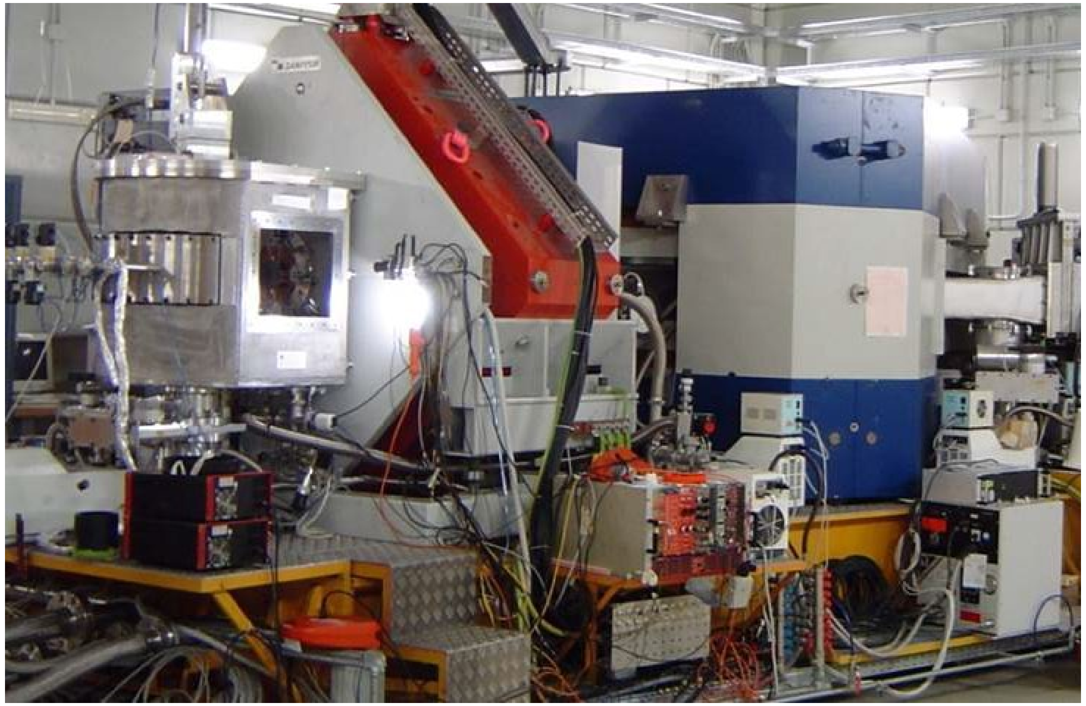
DCE

$$
\begin{cases}
{}^{A}_{Z}X + {}^{a}_{z}x \rightarrow {}^{\;\;\;\;A}_{Z+2}Y + {}^{\;\;\;\;a}_{z-2}y \\
{}^{A}_{Z}X + {}^{a}_{z}x \rightarrow {}^{\;\;\;\;A}_{Z-2}Y + {}^{\;\;\;\;a}_{z+2}y
\end{cases}
$$

Canditates isotopes: ^{48}Ca, ^{82}Se, ^{100}Mo, ^{124}Sn, ^{128}Te, ^{130}Te, ^{136}Xe, ^{148}Nd, ^{150}Nd, ^{154}Sm, ^{160}Gd, ^{198}Pt .

NUMEN Requirements

- DCE reactions have a low cross section (shown by Diana Carbone) (few nb) and a lot of data are required for having a good statistics: need of high intensity ion beam of ^{18}O and ^{20}Ne (more than 13 μA up to about 50 μA)
- A good energy resolution is required too, in order to clearly distinguish the energy levels of recoiling nuclei: targets must be thin, below 1 μm
- High intensity beams produce a lot of heat by energy loss, which standalone thin targets cannot withstand

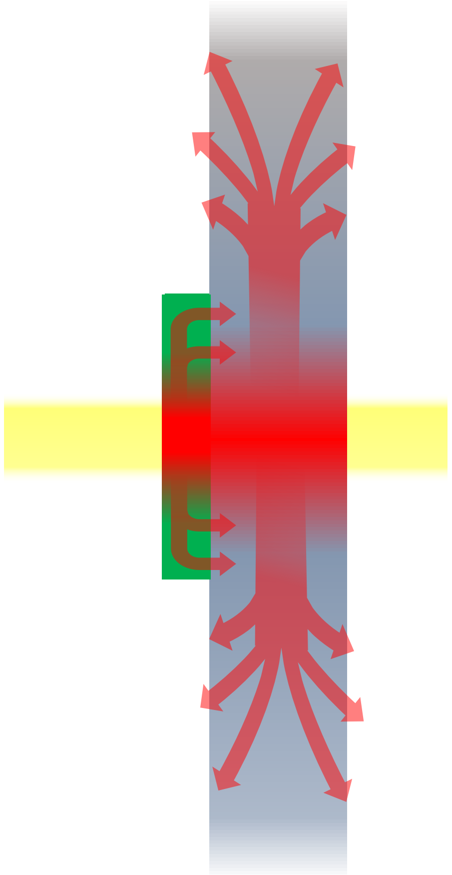


NUMEN challenges

- Thermal stress is addressed by depositing the targets on a high thermally conductive substrate of carbon foils made of multilayers of graphene (MLG)
- **The ideal MLG substrate should have the following chraracteristics:**

1) High in-plane thermal conductivity ($k_{//}$=1400-2300 $Wm^{-1}K^{-1}$) to dissipate heat efficiently toward the cooling system

2) Thickness around 2 µm and good **thickness uniformity** to minimize the impact on the energy resolution of the reaction products

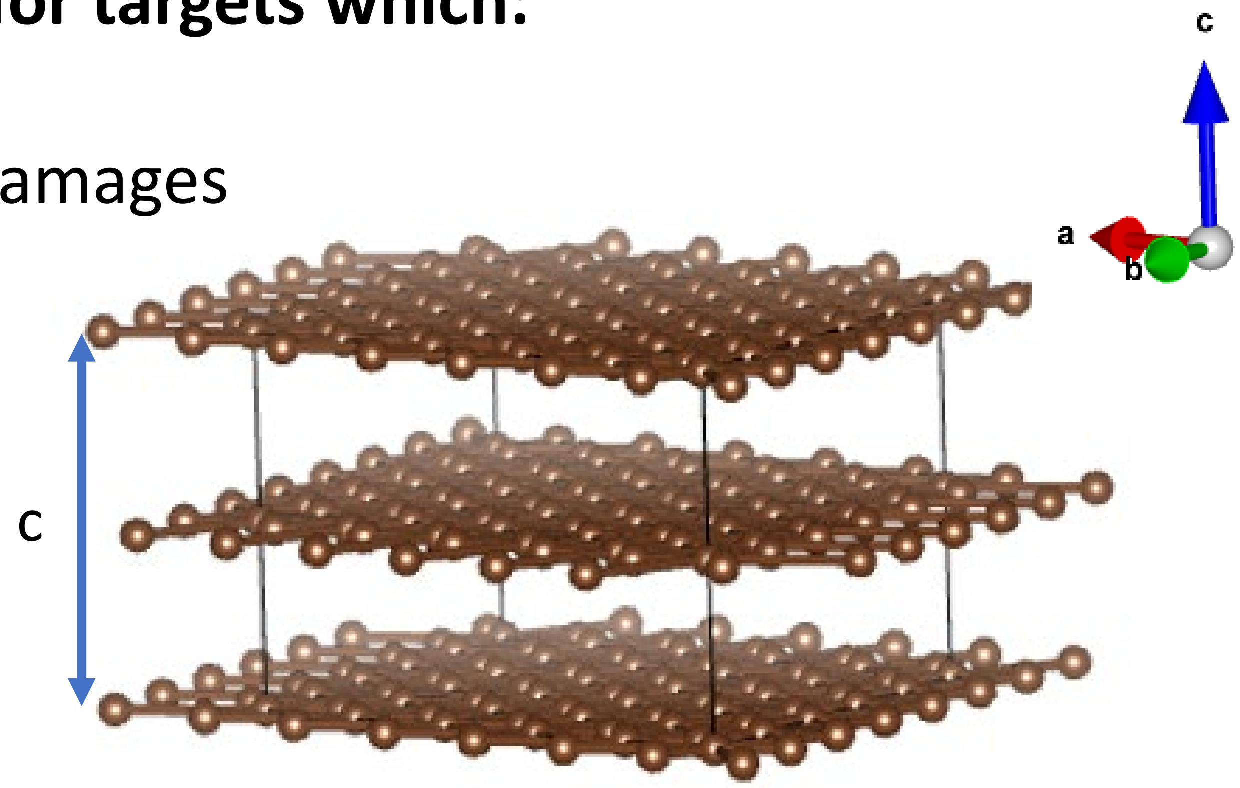


MLG foils

Need for graphitic foils as substrates for targets which:

- ➢ Dissipate heat efficiently
- ➢ Sufficiently resilient to irradiation damages
- ➢ Affect the reaction products energy as little as possible

➡ Proper tickess and uniformity



Interplanar distances d = c/2

Different kind of carbon foils 1-2µm thick

- HOPG (high oriented pyrolitic graphite) from Optigraph, Germany
- MLG (multi-layer graphene) from Kaneka, Japan
- MLG from ACF-metals, Arizona, USA (and Appl. Nanotech, Texas)
- MLG from Micromatter, Canada

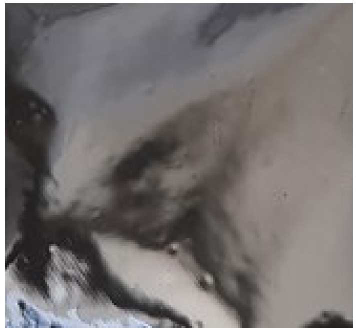
- HOPG-Optigraph

CVD + HT HP annealing

- MLG-Kaneka

Pyrolisis of thin polyimide film

- MLG-ACF/Nanotech



GO + rGO

- MLG-Micromatter

Pulsed Laser Deposition (PLD)

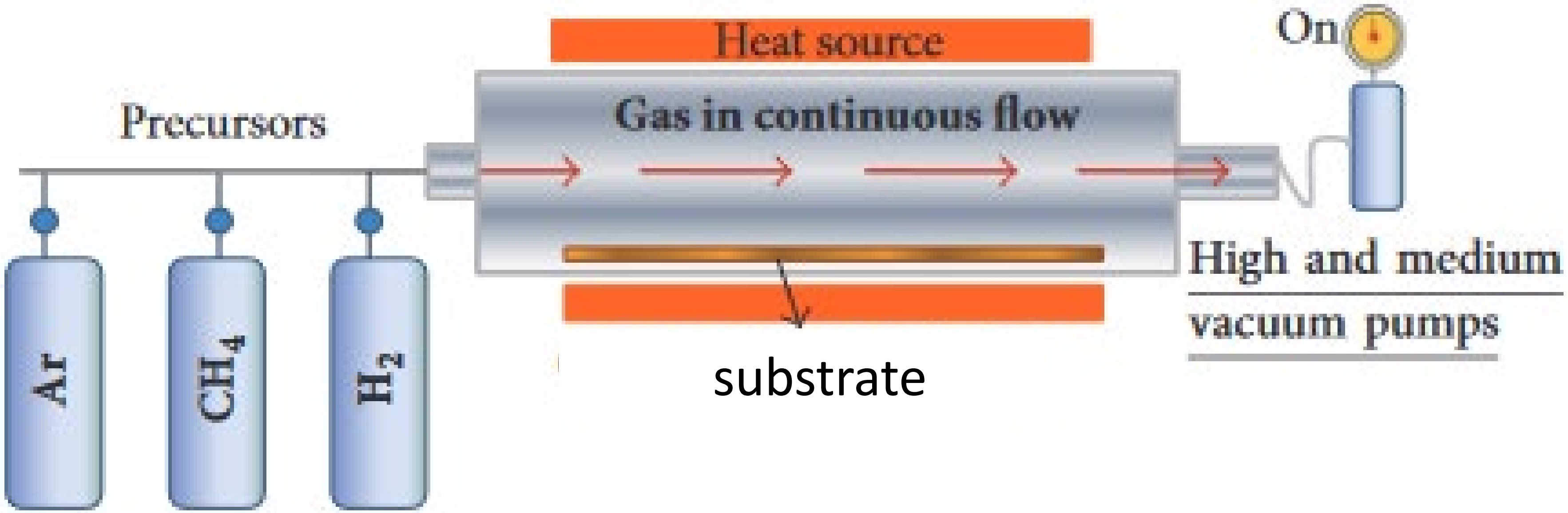
Different appearance (but metallic luster), different preparation processes

HOPG-Optigraph
(a top-down process)

Precursor: hydrocarbon gas (e.g. methane)

Process: pyrolisis in 2 steps

1) CVD with HOPG deposited on a heated graphite substrate
(e.g. **$CH_4 \longrightarrow C(s) + 2\ H_2(g)$**)



L. C. F. Blackman, A. R. Ubbelohde, Proc. R. Soc. A, Math. Phys. Sci. 20–32 (1961).
A. W. Moore, Chemistry and Physics of Carbon 11 Marcel Dekker, Inc. 69–187 (1973).

HOPG-Optigraph

Precursor: hydrocarbon gas (e.g methane)

Process: pyrolisis in 2 steps

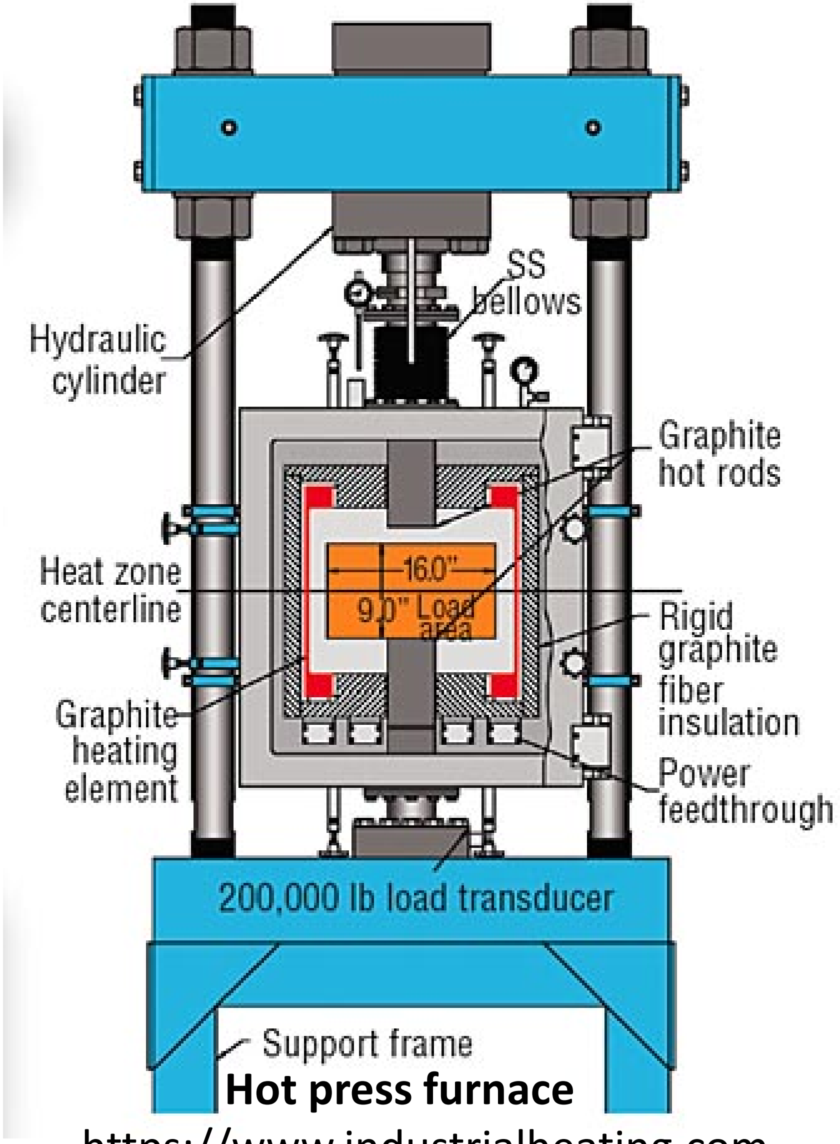
2) Uniaxial pressed during the annealing process at 3000 °C. **Stress recristallization of graphite**. Due to T gradient the quality of material is best at the center of the annealed volume -→ **foils need to be cut (from https://mikromasch.net/)**

Stress recrystallization of graphite

By L. C. F. Blackman* and A. R. Ubbelohde, F.R.S.

Department of Chemical Engineering and Chemical Technology, Imperial College, London, S.W. 7

L.C. F. Blackman and A.R. Ubbelohde, Proc. R. Soc. A. 266 (1962)



https://www.industrialheating.com

MLG-Kaneka

Precursor: solid polyimide

Process: pyrolisis of thin polyimide film in 2 steps

1) **Carbonization**
2) **Graphitization-→ MLG-Kaneka**

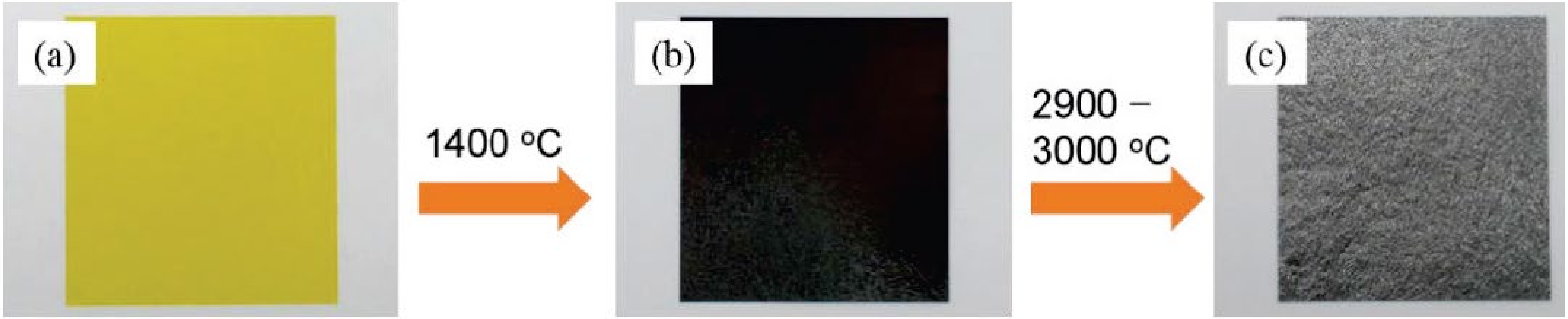
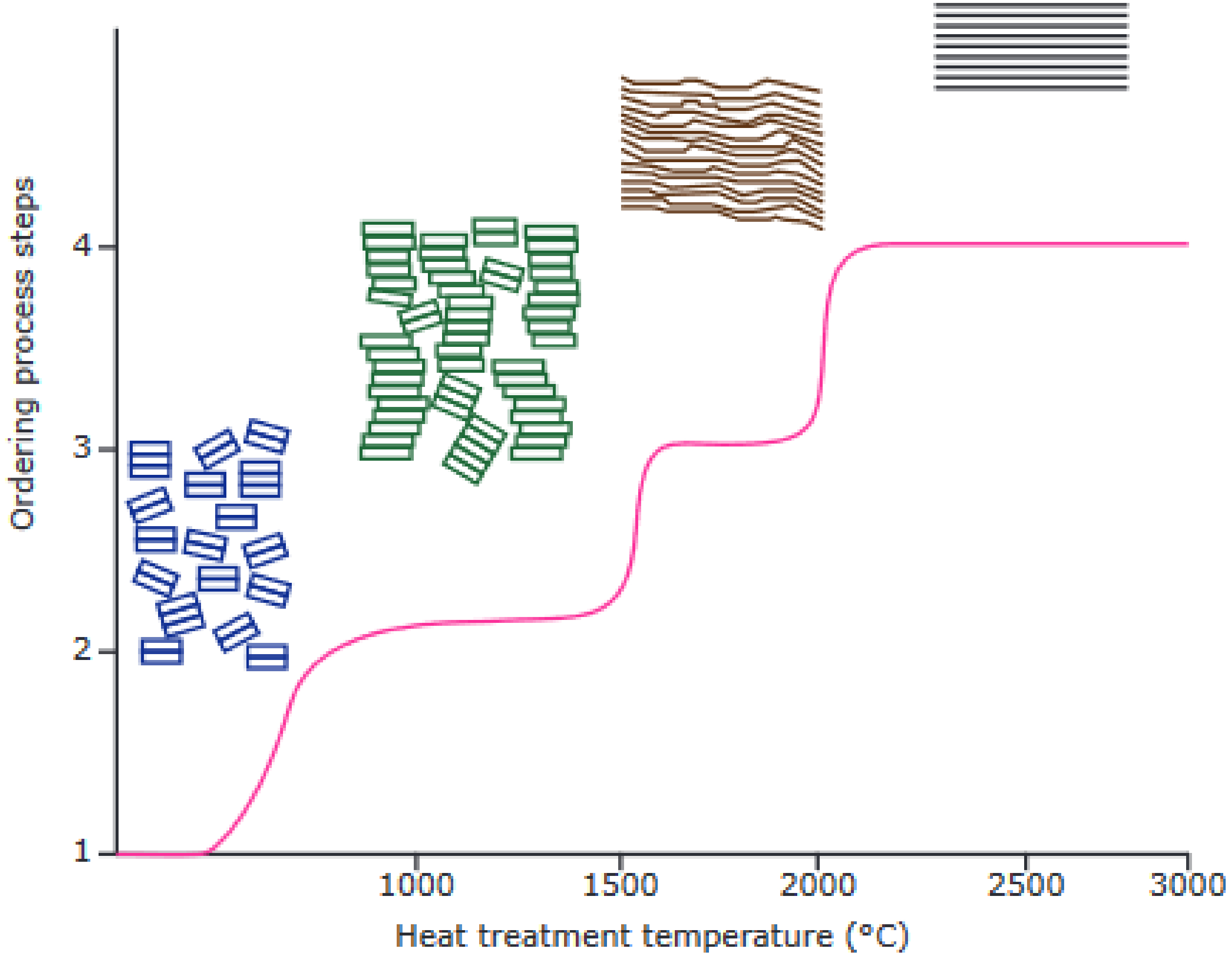


FIGURE 1. Preparation of MLG (a) KANEKA polyimide film; (b) Carbonized film; (c) MLG;

A. Tatami et al., AIP Conference Proceedings **1962**, 030005 (2018)

MLG-Kaneka: Carbonization vs. Graphitization

From carbonization
(chaotic order of small domains)
to graphitization
(large straight layers are formed)



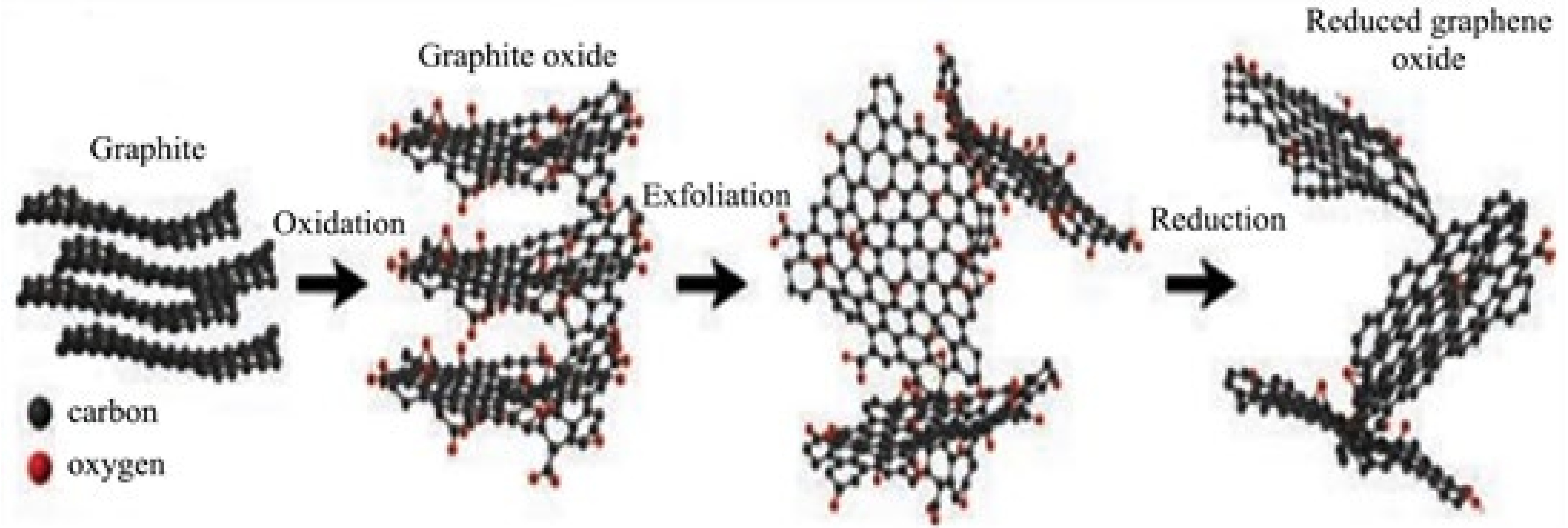
Leading Heat Technology, whitepaper www.carbolite-gero.com

MLG-ACF/Nanotech: a bottom-up process

Precursor: solid graphite

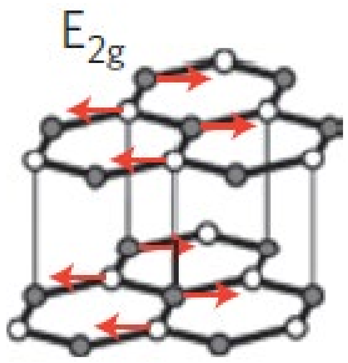
Process: 3 steps

1) **Chemical exfoliation --→ Formation of graphene oxide (GO)**
2) **Dispersions of reduced graphene oxide (rGO)**
3) **Accumulation of rGO up to the desired thickness**

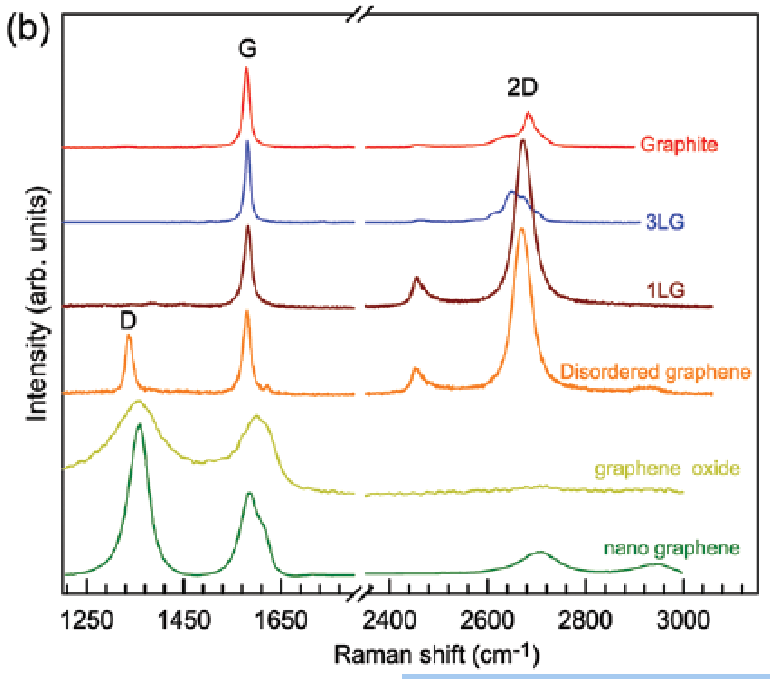


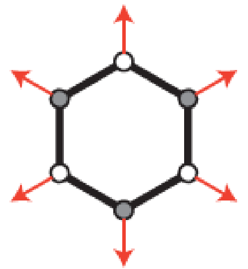
A. Adeniji et al., "Synthesis and Fabrication of Graphene and Graphene Oxide: A Review", Open Journal of Composite Materials Vol. 9 (2019), Scientific Research Publishing

Raman measurements on HOPG-Optigraph



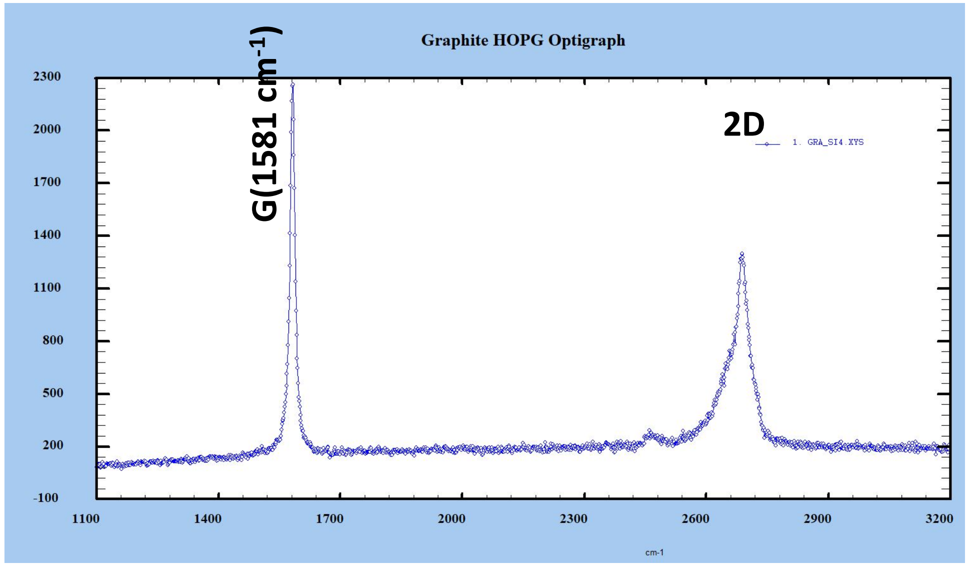
G peak: inplane vibrations of sp^2 rings.
Common in all sp^2 materials

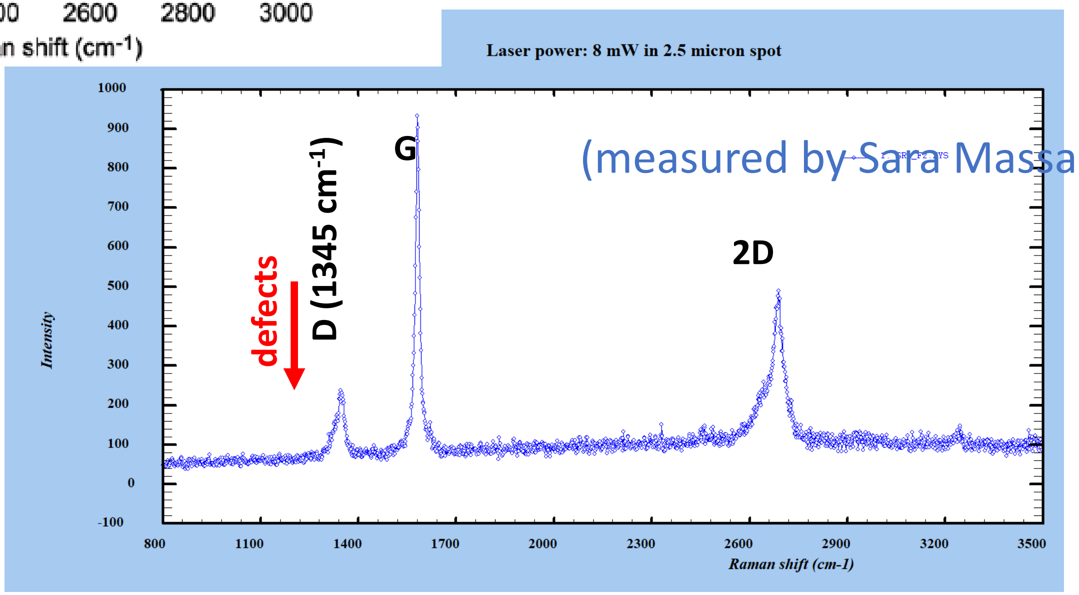




D peak: breathing mode of 6 atom rings (required a **defect** for his activation)
2D peak: D peak overtone, two-phonon process, **no defect is required**

A. Ferrari et al. Nature Nanotech. 8, 2013

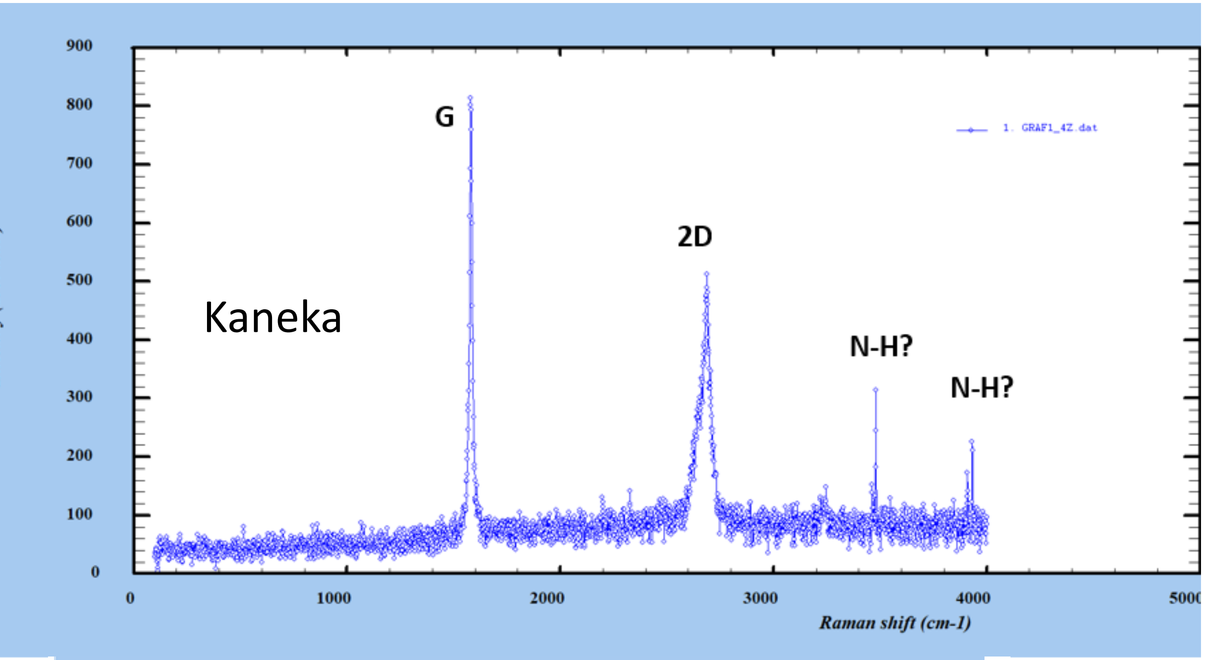


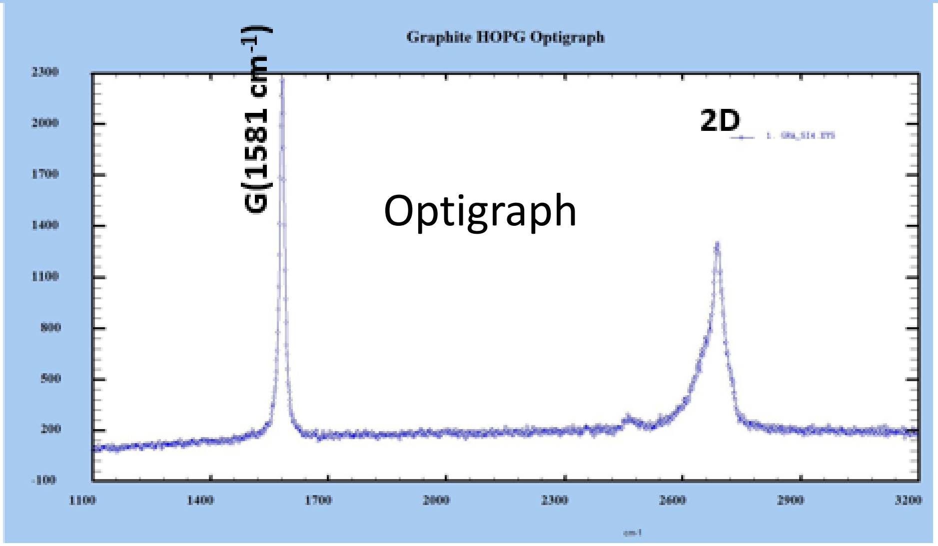


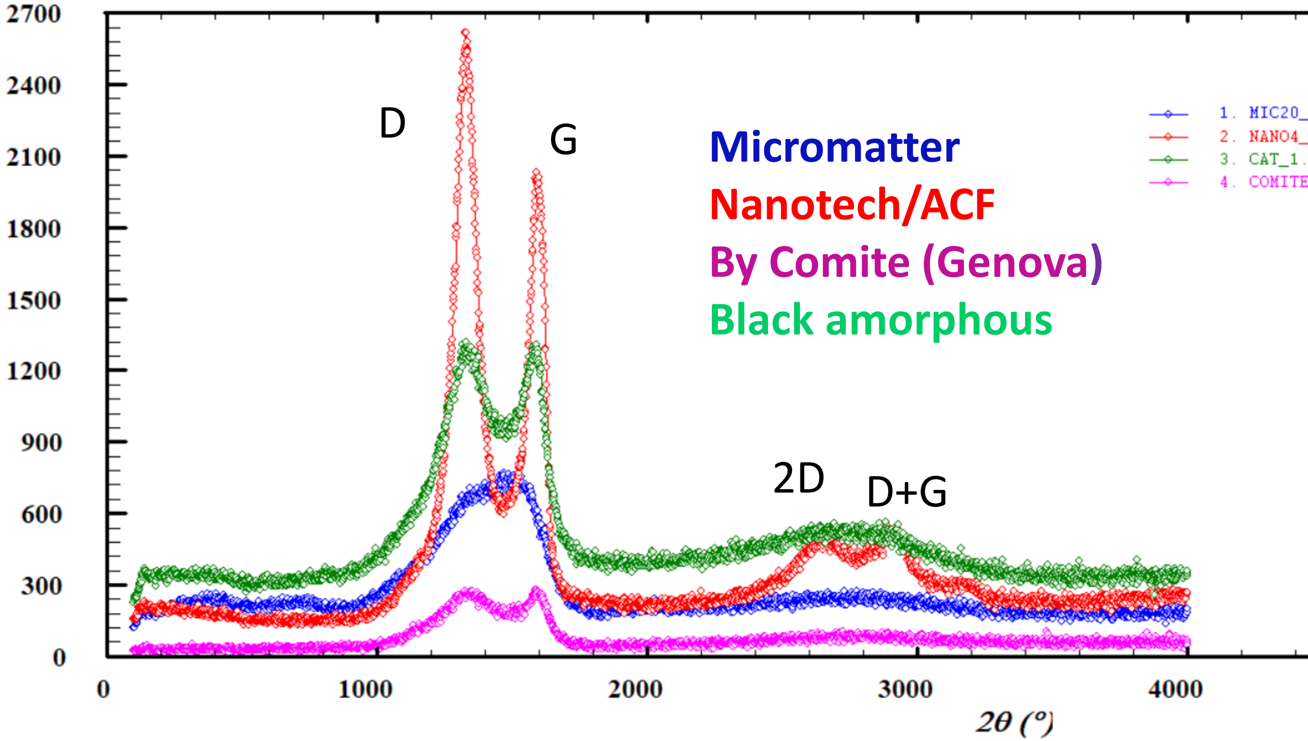
(measured by Sara Massardo, Unige)

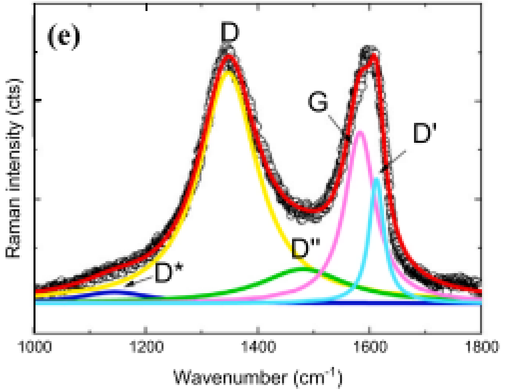
Defect induced by increasing power of laser (8mW)

COMPARISON Between RAMAN MEASUREMENTS









Intensity of D'' inversely related to crystallinity

Evaluating density by XRD

Evaluating thermal diffusivity by laser flash method

$$K = \alpha \ \rho \ C_p$$

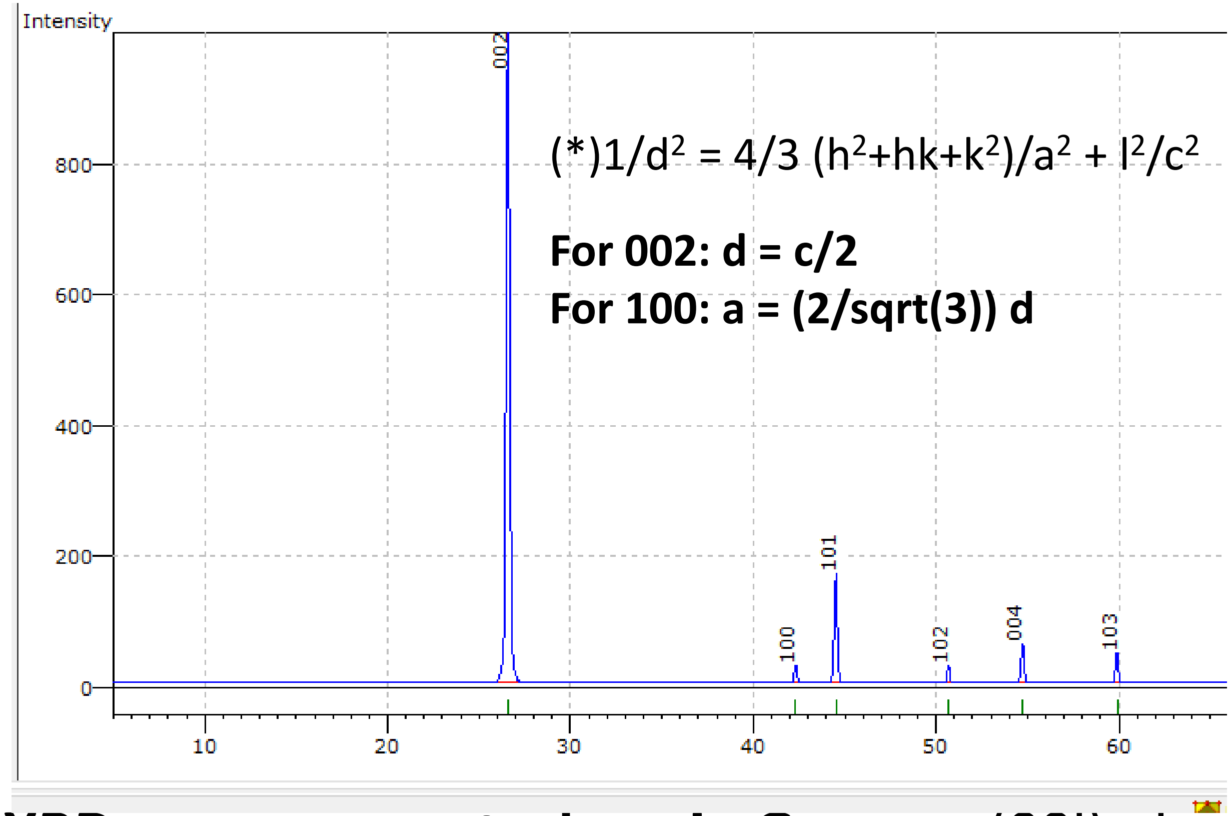
K = thermal conductivity; ρ = density; C_p = specific heat; α = thermal diffusivity

Evaluating density by XRD

$$d = m_{uc}/V_{uc}$$

HOPG: XRD in reflection

Theoretical XRD pattern

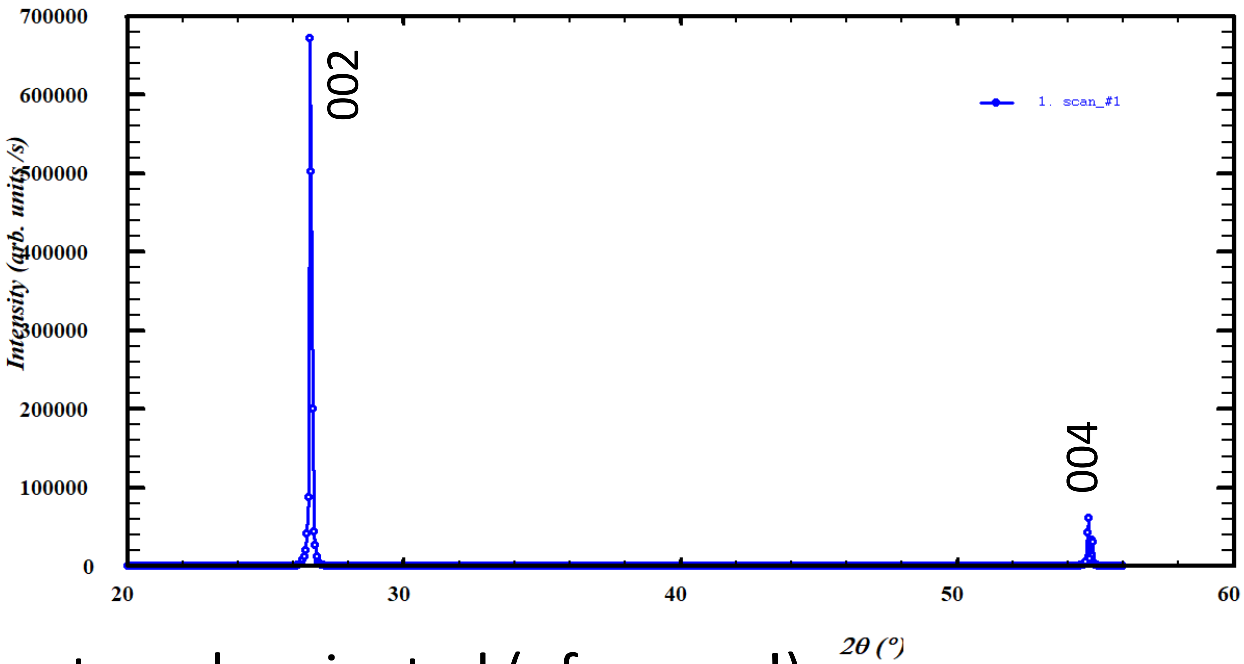


(*)$1/d^2 = 4/3\ (h^2+hk+k^2)/a^2 + l^2/c^2$

For 002: d = c/2

For 100: a = (2/sqrt(3)) d

Experimental XRD pattern



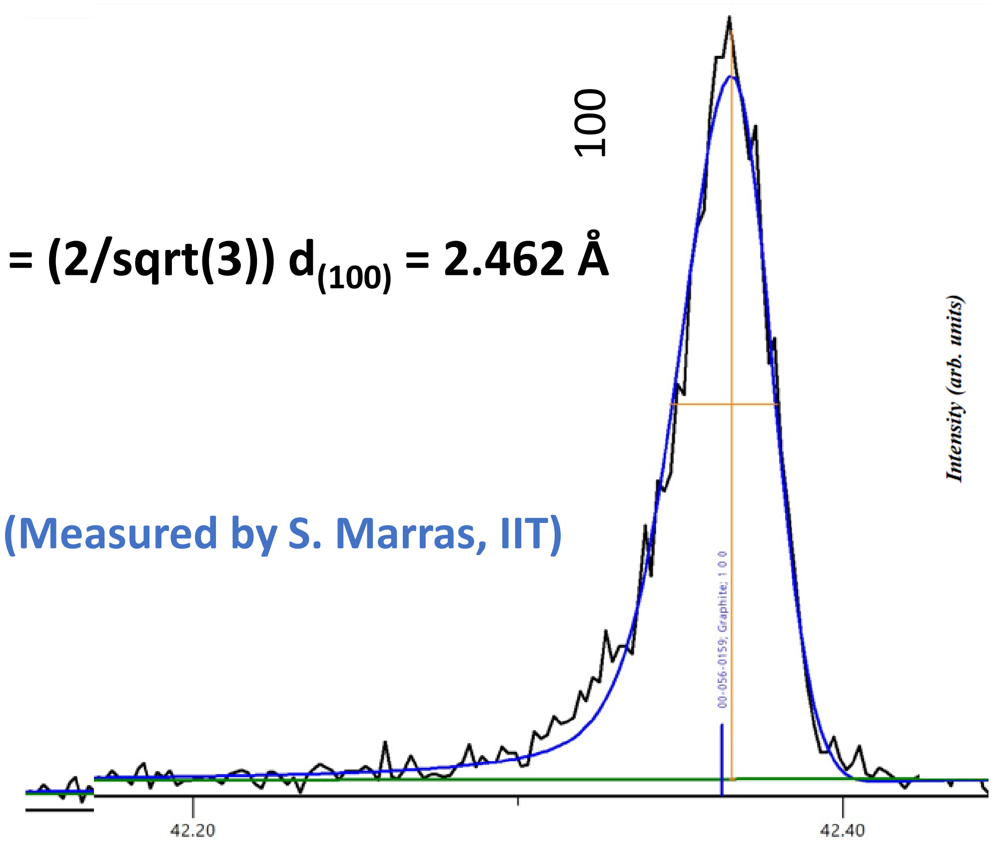
XRD measurements done in Genova: (00l) planes strongly oriented (of course!)

002 peak at 26.61 in 2Θ **d(interlayers) = 3.346 Å**

c = 6.692 Å(*) a = 2.464 Å (literature) **Vuc = a^2 c 0.866 = 35.18 $Å^3$ = 3.518 10^{-23} cm^3**

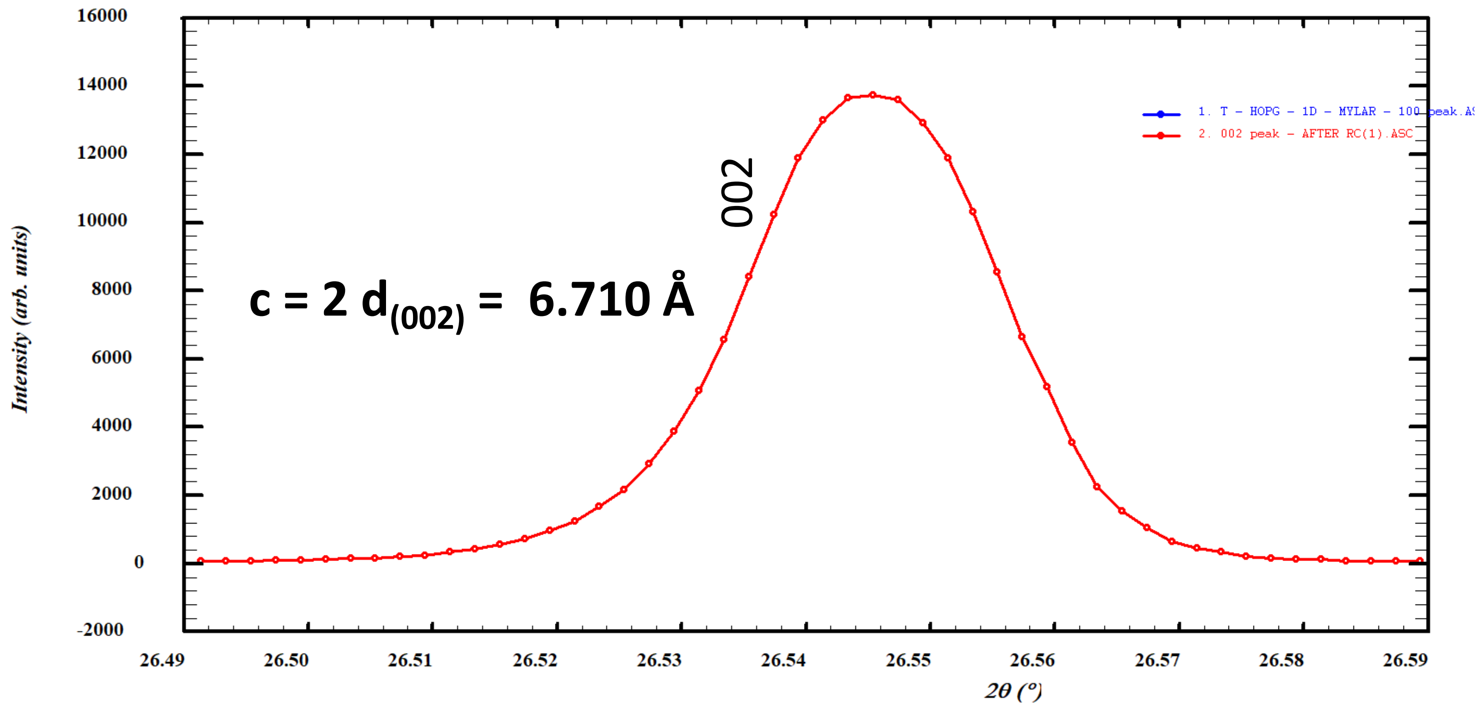
Density = [atomic weight x Natom] /[Vuc x N_A] = [12.0107 x 4] / [3.518 10^{-23} cm^3 x 6.023 10^{23}] = 2.267 g cm^{-3}

HOPG: XRD in trasmission



$a = (2/\mathrm{sqrt}(3))\ d_{(100)} = 2.462$ Å

(Measured by S. Marras, IIT)



$c = 2\ d_{(002)} = 6.710$ Å

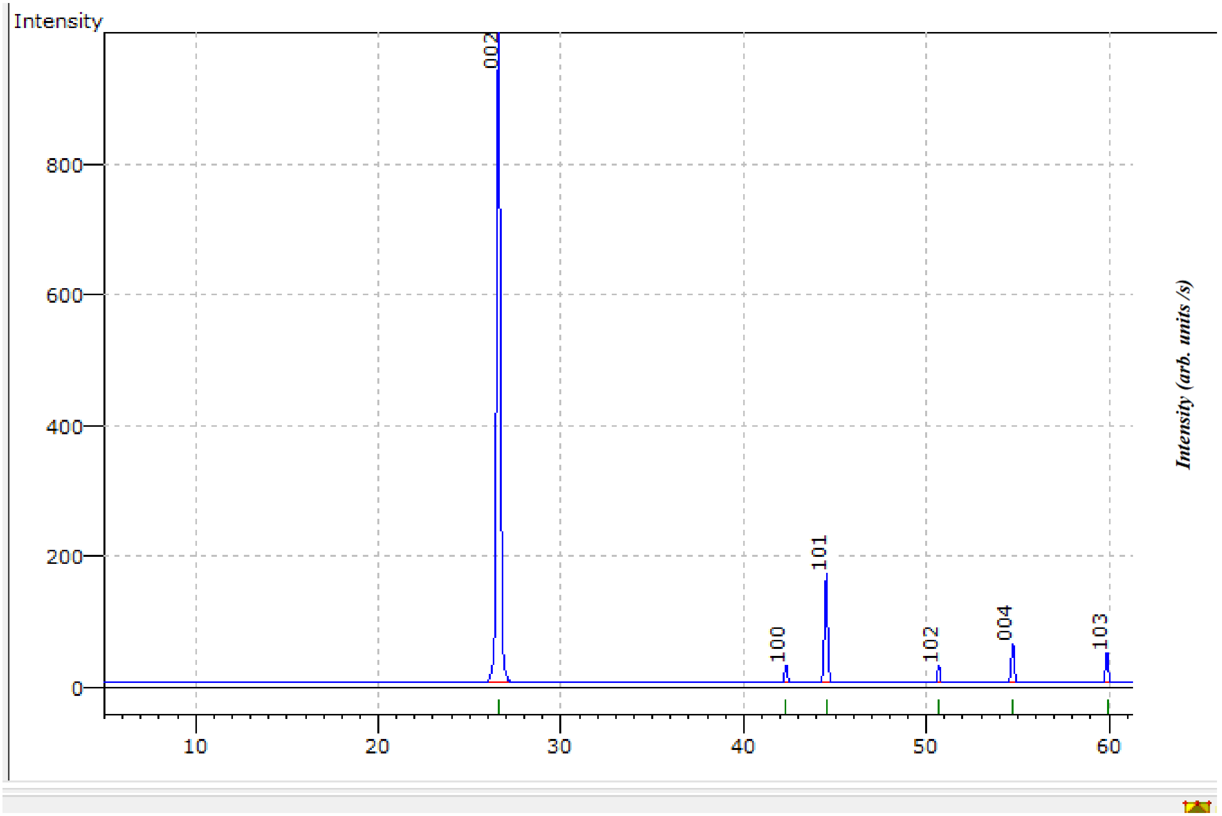
$V_{uc} = a^2 \times c \times 0.866 = 35.18$ Å$^3 = 3.522\ 10^{-23}\ cm^3$

Density = [atomic weight x Natom] /[Vuc x N_A] = [12.0107 x 4] / [3.522 10^{-23} cm^3 x 6.023 10^{23}] =

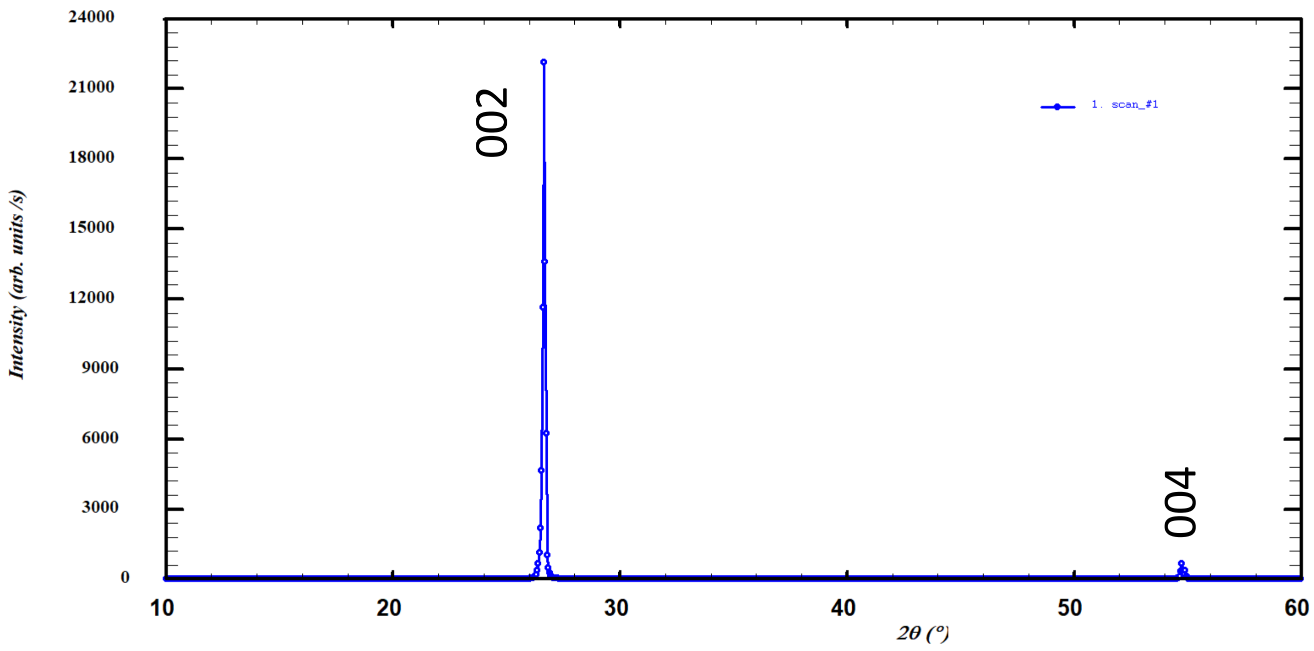
2.265 g cm^{-3}

XRD MLG-Kaneka

Theoretical XRD pattern



Experimental XRD pattern



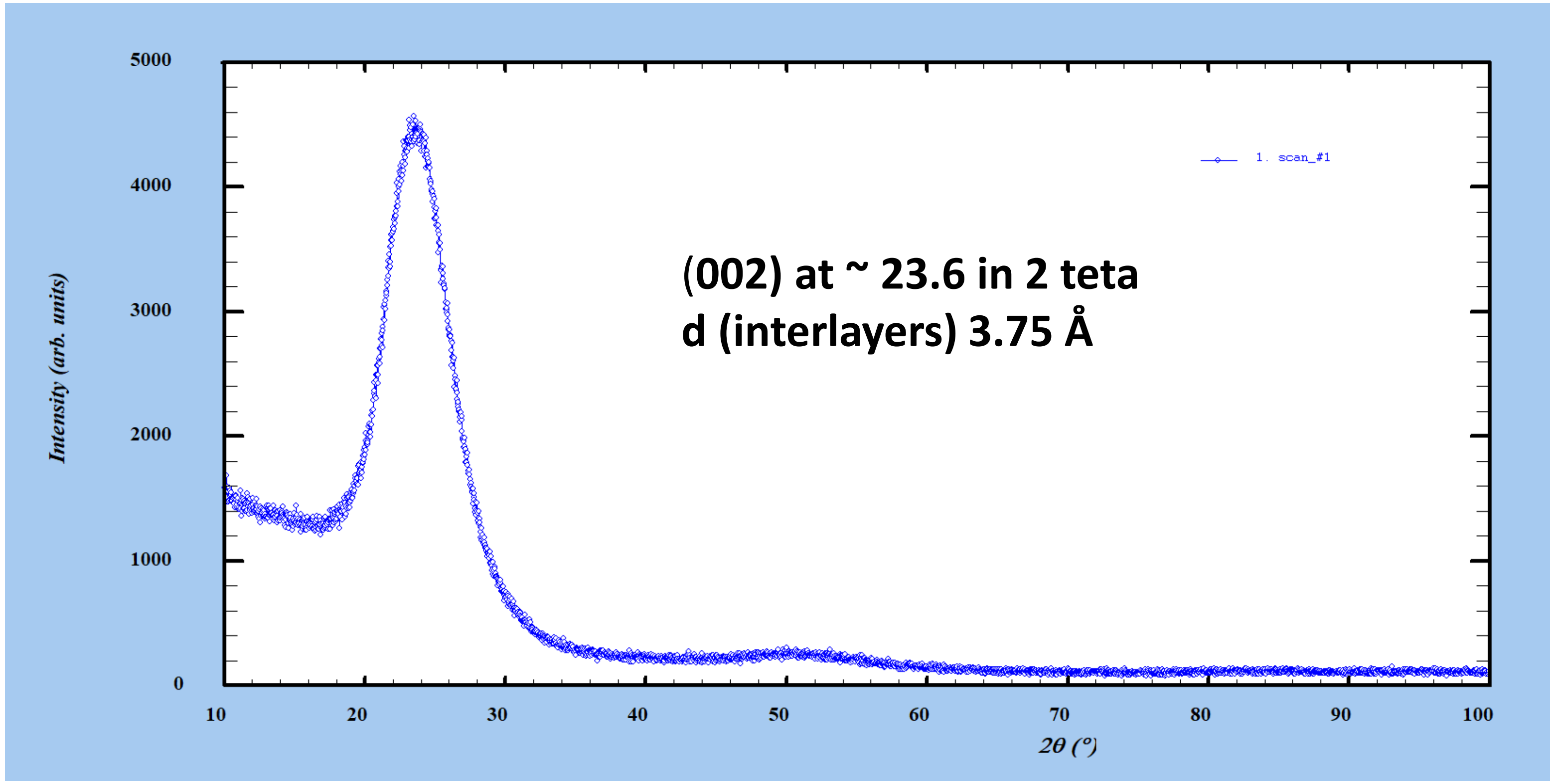
002 peak at 26.68 in 2Θ **d(interlayers) = 3.338 Å**

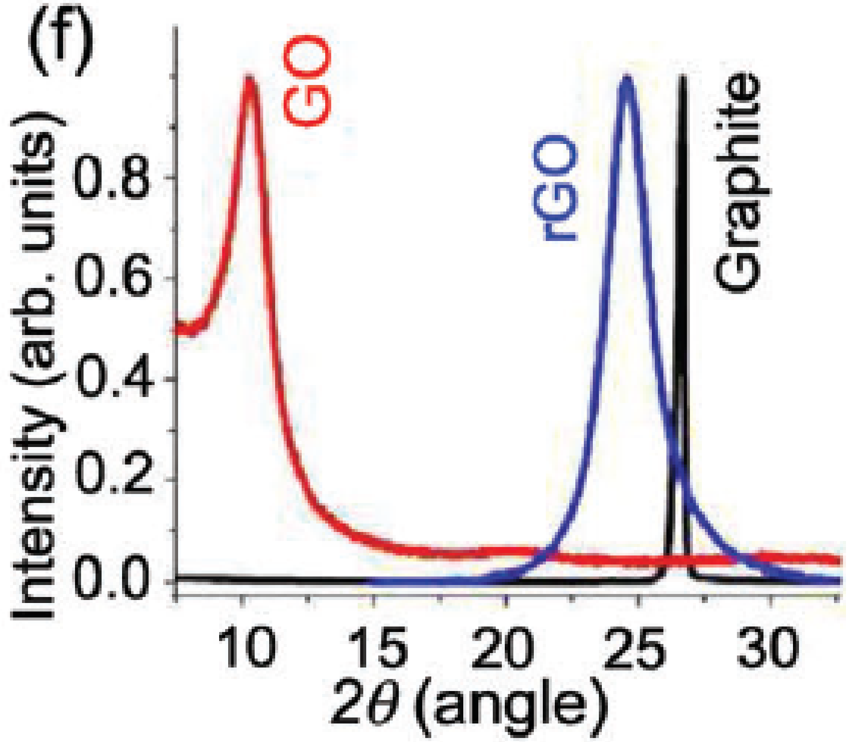
c = 6.676 Å a = 2.464 Å (literature) **V_{uc} = a^2 x c x 0.866 = 35.10 $Å^3$ = 3.510 10^{-23} cm^3**

Density = [atomic weight x Natom] /[V_{uc} x N_A] = [12.0107 x 4] / [3.510 10^{-23} cm^3 x 6.023 10^{23}]

= 2.272 g cm^{-3}

XRD: MLG-ACF/Nanotech





Molecules intercalated between adiacent rGO sheets

J.B. Wu et al. Chem. Soc. Rev. 47 2018, 1822

c = 7.50 Å a = 2.464 Å (literature) **V_{uc} = a^2 c 0.866 = 35.18 $Å^3$ = 3.94 10^{-23} cm^3**

Density = [atomic weight x Natom] /[Vuc x N_A] = [12.0107 x 4] / [3.94 10^{-23} cm^3 x 6.023 10^{23}] = 2.02 g cm^{-3}

Evaluating thermal diffusivity by laser flash method

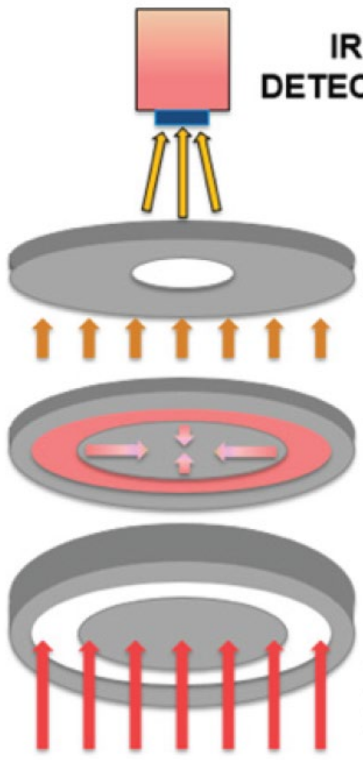
Thermal conductivity of HOPG-Optigraph

The laser flash method measures **thermal diffusivity α**:

$$K = \alpha . \rho . C_p$$

- **Cooperations established:**
  - Prof. Takao Mori, NIMS (National Institute of Materials Science), Tsukuba, Japan)

K = thermal conductivity**; ρ** = density; **C_p** = specific heat; **α** = thermal diffusivity



Flash apparatus LFA467 HyperFlash

Advantages:
- No thermal contact resistance
- Heat losses minimized by short time measurement

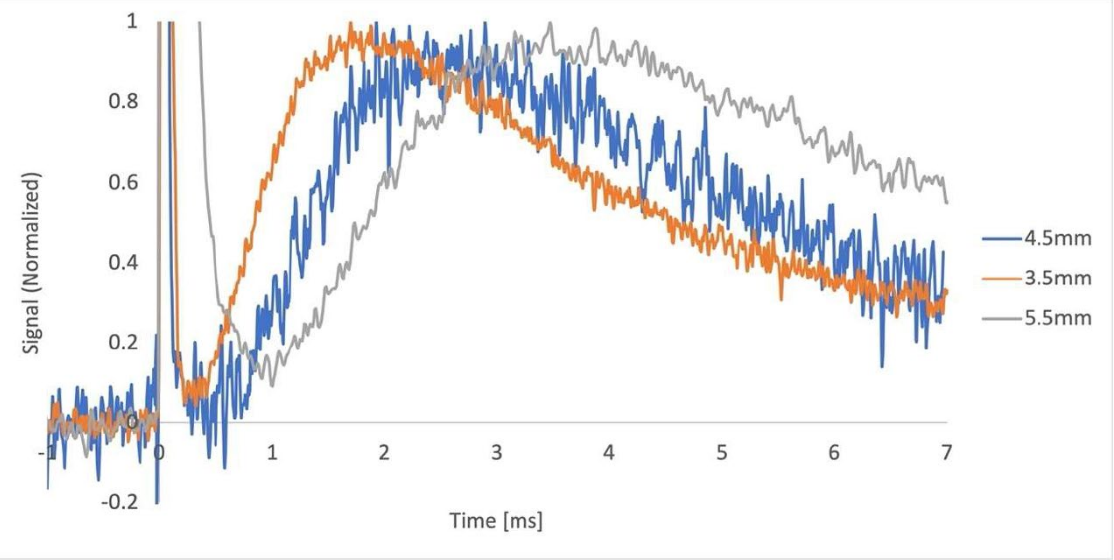


Figure 1: Signals from HOPG film (d = 3.5, 4.5, 5.5[mm])

For a HOPG foil of 2 µm
→ K = 2320 ± 107 [W/(m.K)]

Thermal conductivity of MLG-Kaneka

Film thickness: 1.6 ± 0.2 μm measured using a contact film thickness gauge

$$K = \alpha . \rho . C_p$$

K = thermal conductivity**; ρ** = density; **C_p** = specific heat; **α** = thermal diffusivity

$\alpha = 1.2\ 10^{-3}\ m^2\ s^{-1}$ measured by **Laser Flash method** from 2.1 μm of MLG-Kaneka

A. Tatami et al. Proc. 15° Intern Heat Transer Conf. IHTC-15 Kyoto (2014)

In good agreement with α ($1.0\ 10^{-3}\ m^2\ s^{-1}$) measured **by spot heating Ångström** method

A. Tatami et al. AIP Conference Proceedings 1962, 0300005 (2018)

We can calculate:

K = $1.2\ 10^{-3}\ mm^2\ s^{-1}$ 0.73 J /(g K) 2.272 g / cm^3 **= $2.0\ 10^3\ W\ m^{-1}\ K^{-1}$**

Thermal conductivity of Nanotech foils

performed by NETZSCH for Nanotech

ASTM E1461 Flash Method Thermal Conductivity Results						
Sample	thickness @ 25°C (mm)	bulk density ρ @ 25°C (g/cm^3)	temperature (°C)	specific heat c_p (J/g-K)	diffusivity α (mm^2/s)	conductivity λ (W/m-K)
graphene foil (in-plane)	0.020	1.55	25	0.730	1308	1480

Value of thermal conductivity affected by a value of density too low:

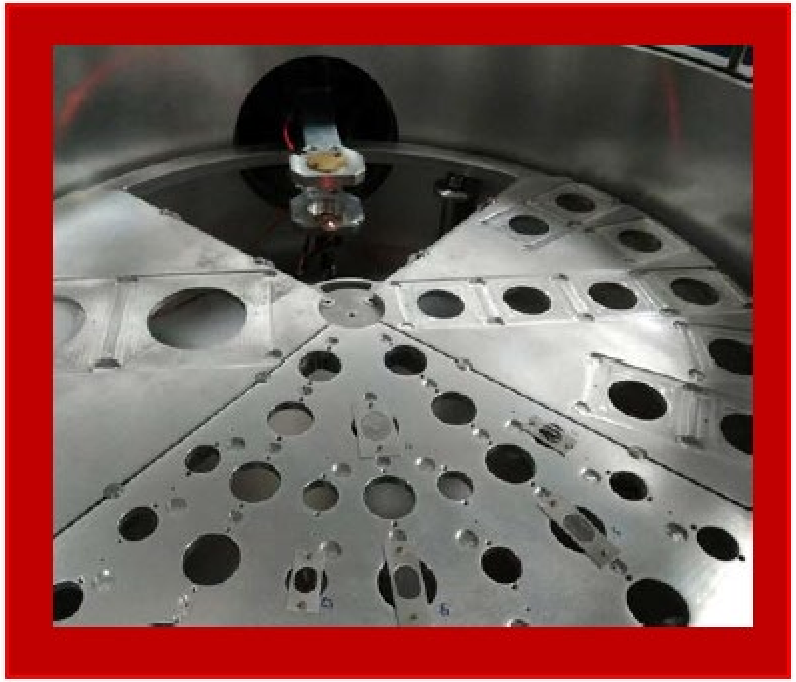
Using a density of 2.02 g/cm^3 -→ **K** = 1480 *2.02/1.55 = **1.93 KW/m K**

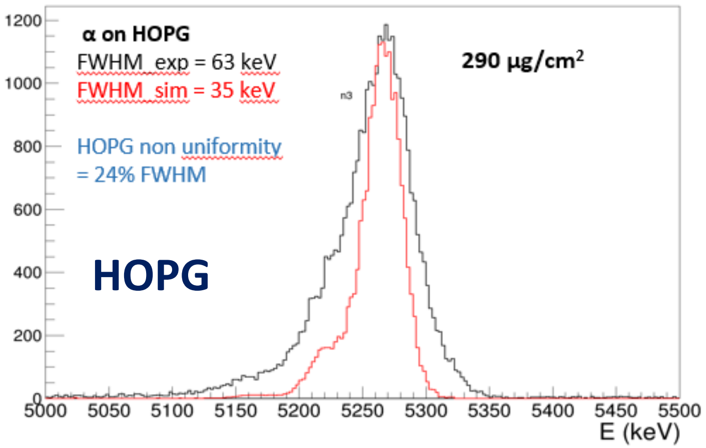
Evaluating tickness uniformity by APT

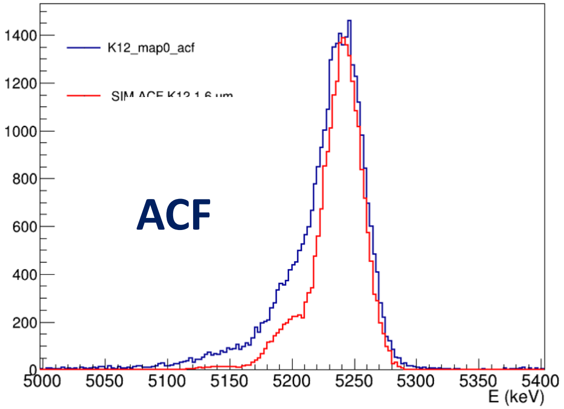
MLG characterization with APT (alpha particle transmission)

Target non-uniformity deduced comparing FWHM of exper. spectrum with FWHM of a simulated spectrum of a uniform substrate (SRIM program)

CACTUS –
Chamber with **A**lpha source to **C**haracterize target **T**hickness and **U**niformity by **S**canning

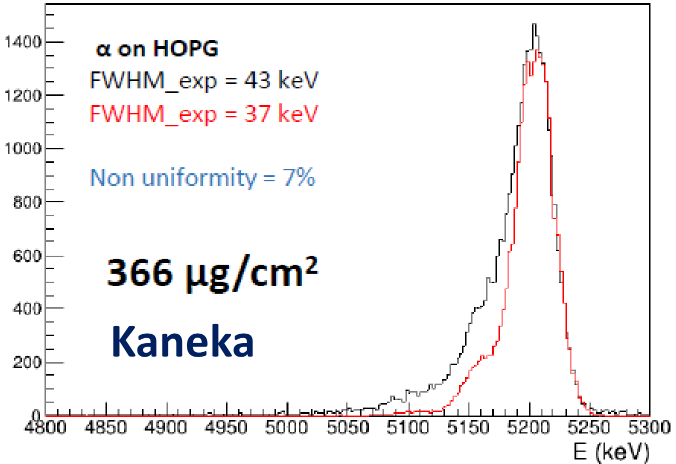






---- K12_map0_acf -------

FWHM_exp = 45 keV
FWHM_sim = 37.5 keV
DE = 244.923 keV
NON-UNIFORMITY = 10%



Summary of results on MLG

- **Synthetic MLG foils with high thermal conductivity can be prepared through different routes**
- **GO + rGO foils (ACF/Nanotech) seems to be a valid alternative as substrates for NUMEN to pyrolitic carbon foils**

Company	Calculated Density [g/cm³]	Purity degree/ crystallinity	Thicknes s [µm]	Thermal diffusivity [mm²/s]	Thermal conductivity [$KWm^{-1}K^{-1}$]	Tickness uniformity by APT
HOPG-Optigraph	2.265	99.9%/HO	2	1550	2.3	poor
MLG-Kaneka	2.272	99%/HO	1.6	1200	2.0 (*)	good
MLG-ACF/Nanotech	2.02	Fair	1-3	1308	1.93 (*)	good
MLG-Micromatter		amorphous			10^{-2}-10^{-1}	

(*) literature values

What's next for MLG?

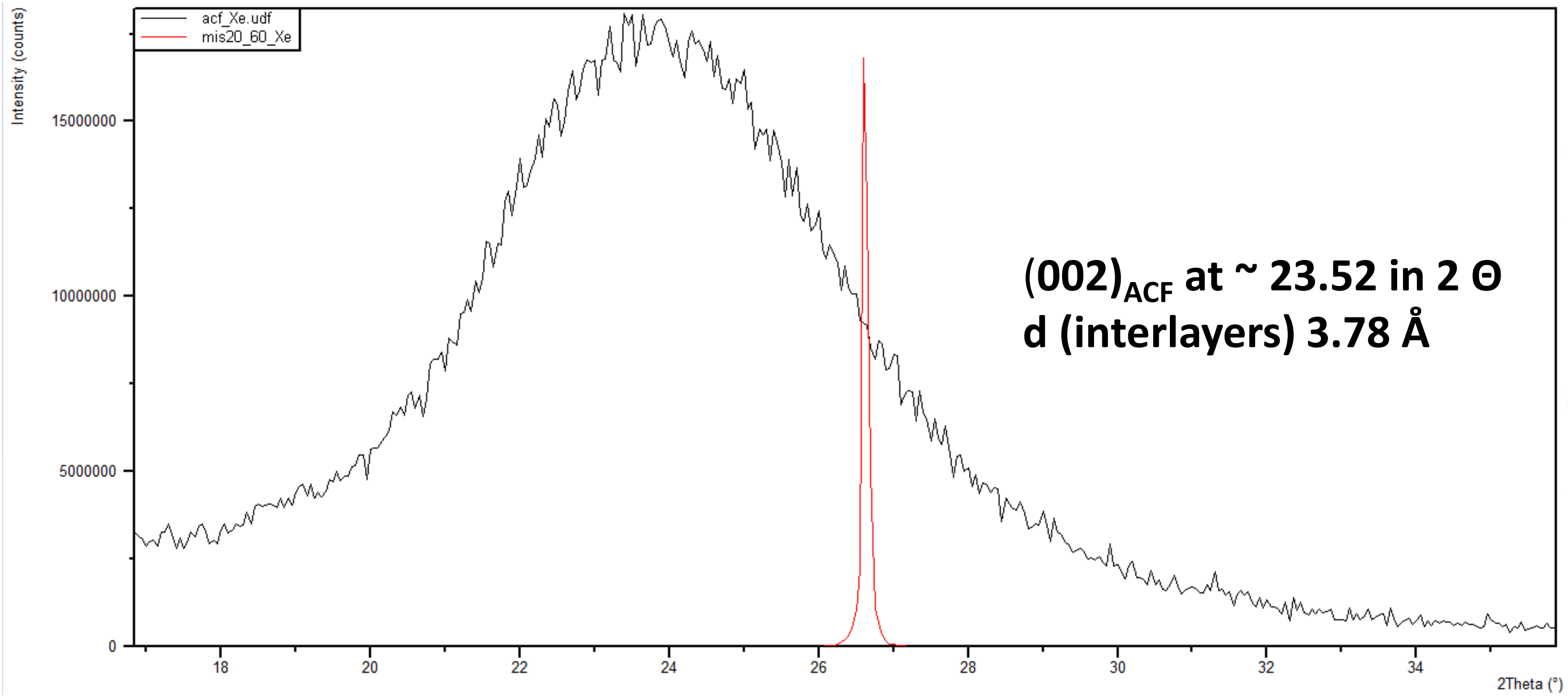
Characterization:

Pristine substrates and substrates analysed after irradiation

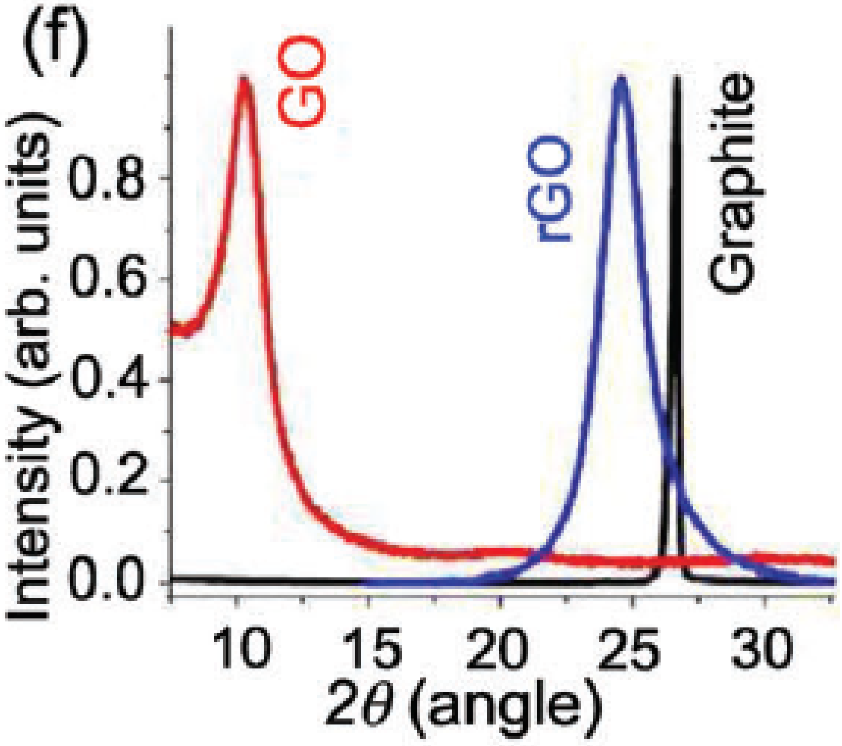
Synthesis: Attempts to prepare MLG using soft chemistry (chimie douce) methods


THANK YOU FOR
YOUR ATTENTION !!!

XRD: MLG-ACF vs MLG-Optigraph



$(002)_{ACF}$ at ~ 23.52 in 2 Θ
d (interlayers) 3.78 Å



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J.B. Wu et al. Chem. Soc. Rev. 47 2018, 1822

Micromatter: an amorphous foil with K = 10-100 $Wm^{-1}K^{-1}$
(Private comunication from Micromatter)