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Irradiation damage in nuclear graphite at the atomic scale

A. Chartier, L. van Brutzel, J. Pageot

► **To cite this version:**

A. Chartier, L. van Brutzel, J. Pageot. Irradiation damage in nuclear graphite at the atomic scale. 9th International Conference on Multiscale Materials Modeling, Oct 2018, Osaka, Japan. cea-02338969

HAL Id: cea-02338969

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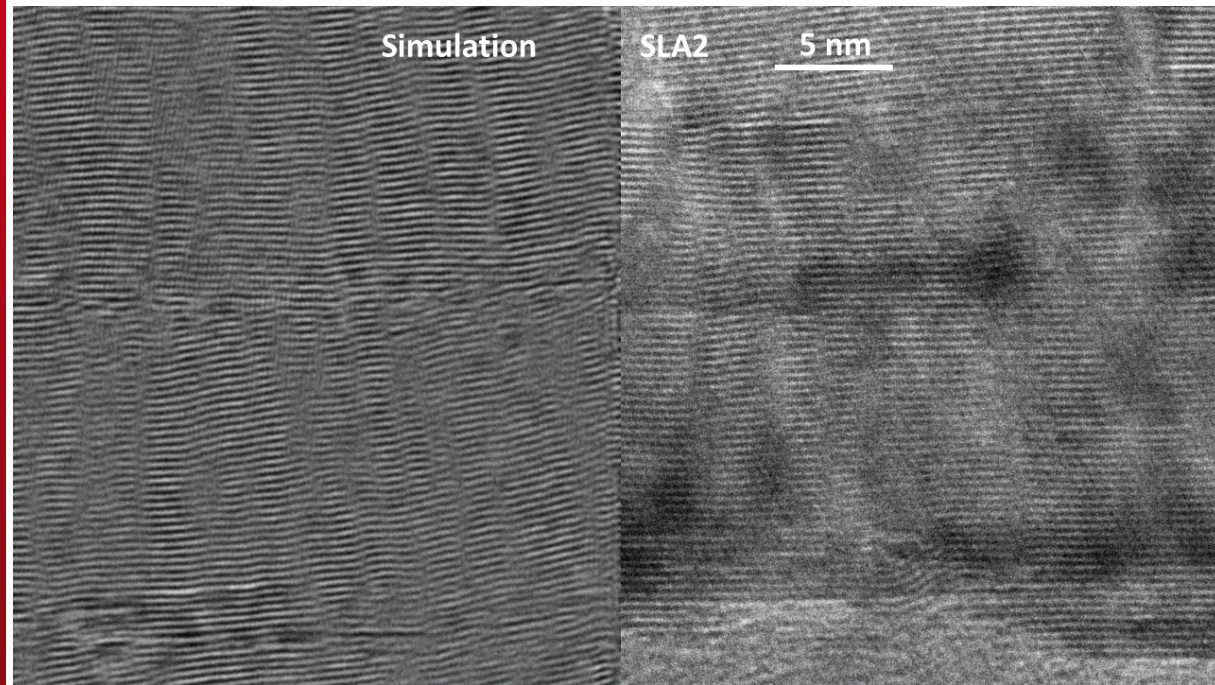
Submitted on 13 Dec 2019

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Irradiation damage in nuclear graphite at the atomic scale

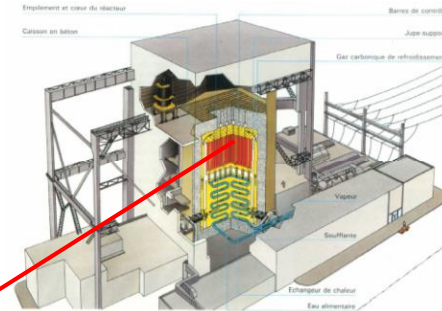
A. Chartier, L. Van Brutzel, and J. Pageot



Framework: decommissioning of graphite from "UNGG"

- Graphite = moderator in "UNGG" → neutrons, temperature
- In 11 years of exploitation **2.6 dpa**
- Activation of some impurities

i-graphite contains ^{14}C & ^{36}Cl ($\frac{1}{2}$ life 5 500 & 3×10^5 years)



Foreseen Solutions:

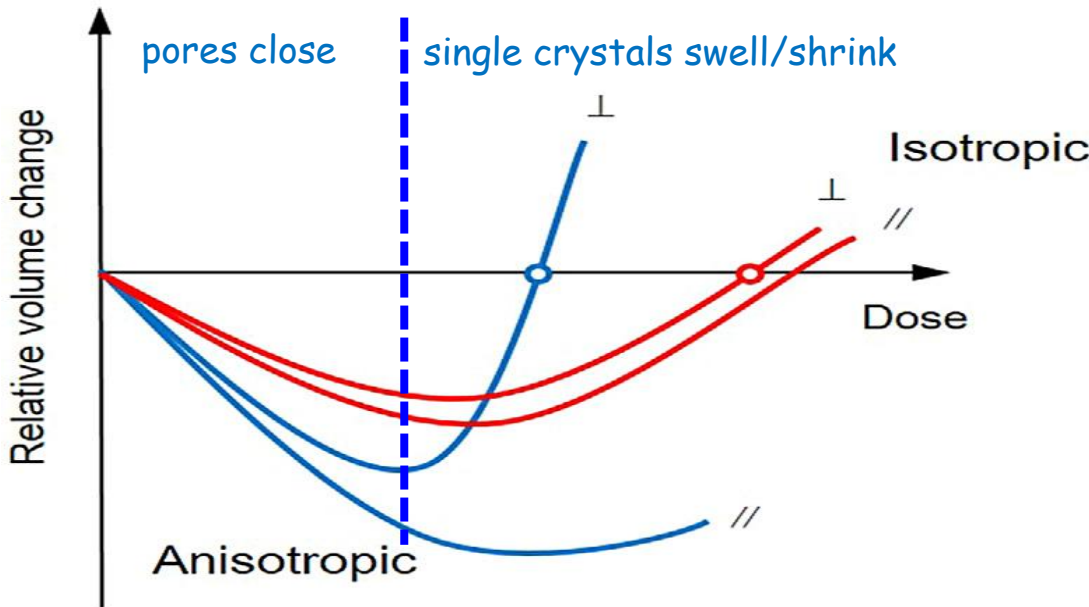
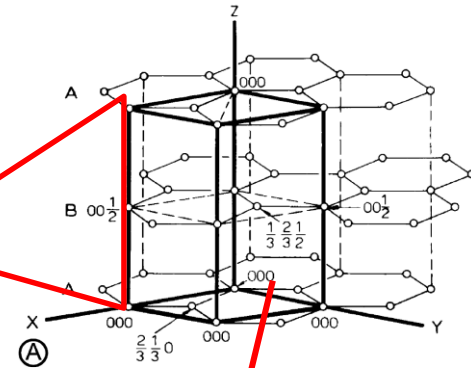
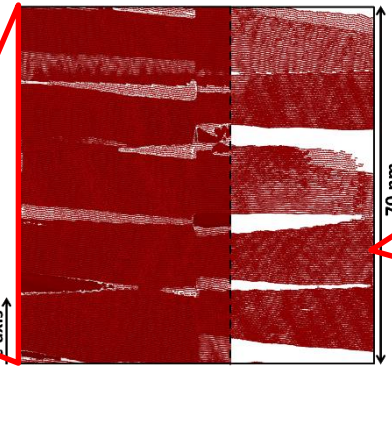
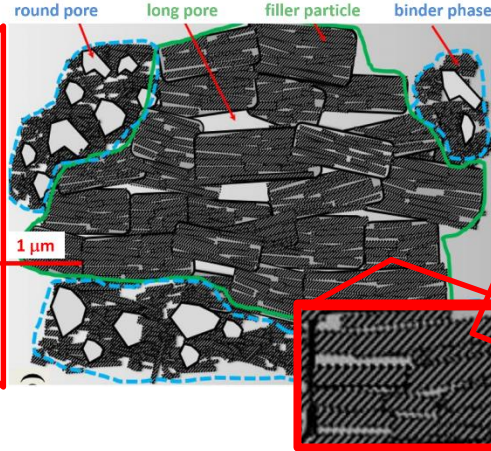
- Decommissioning under water
- Treatment to eliminate ^{14}C & ^{36}Cl , (^3H)

} interaction: graphite \leftrightarrow H_2O , O_2 , CO_2 ...
depends on the **localization of ^{14}C & ^{36}Cl** after irradiation

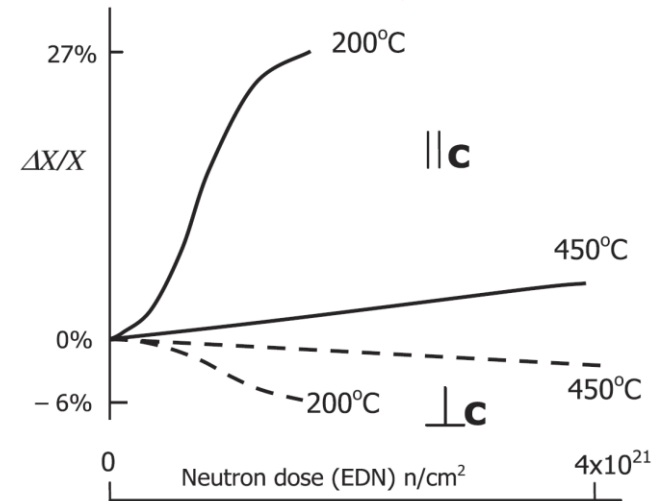
- **Graphite microstructure evolution under irradiation?**
 - radiation damages and swelling
 - link between XRD - HRTEM images and atomic scale structure

cm

nm



Shen et al (2015)



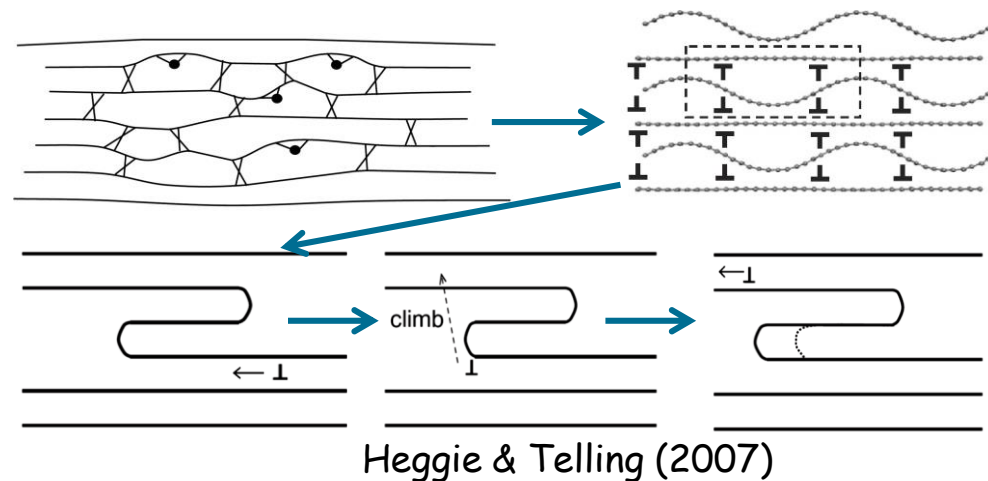
Heggie et al (2011)

Scenario of swelling

1. point defects by irradiation
2. wrinkling
3. sliding
4. formation of dislocations

Based on:

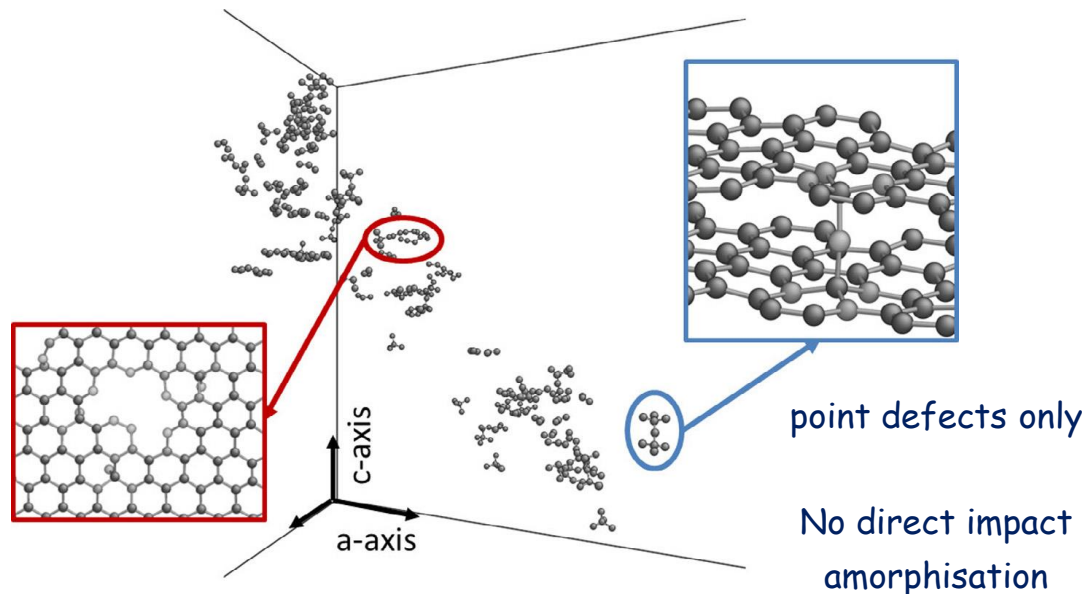
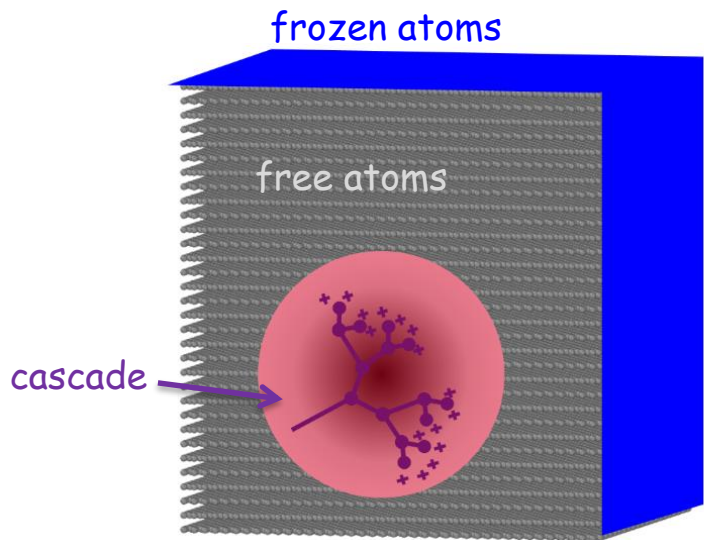
- HRTEM observations
- Atomic scale (DFT - MD) calculations
- Wigner energy estimations



Our goals:

- Link between swelling - atomic configurations for irradiated single crystal
- Link between HRTEM images and atomic configurations for irradiated single crystal for poly-crystal

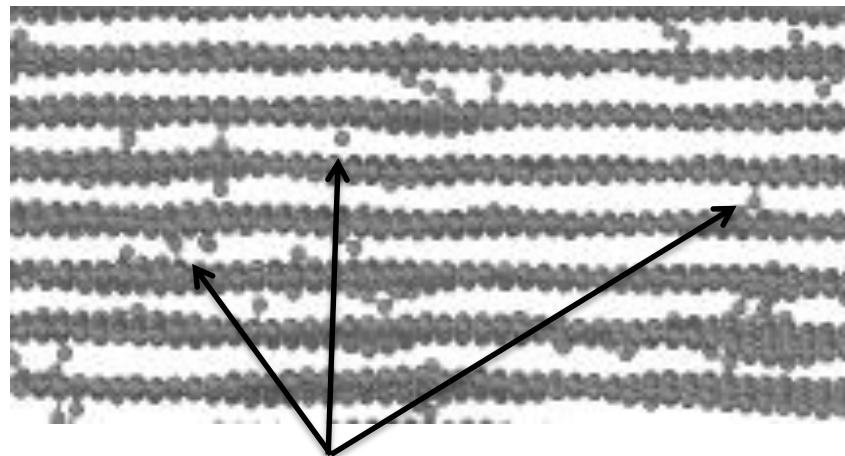
Primary ballistic damage by displacement cascade, with primary knocked on atom with 0.5 to 10 keV



if point defects then

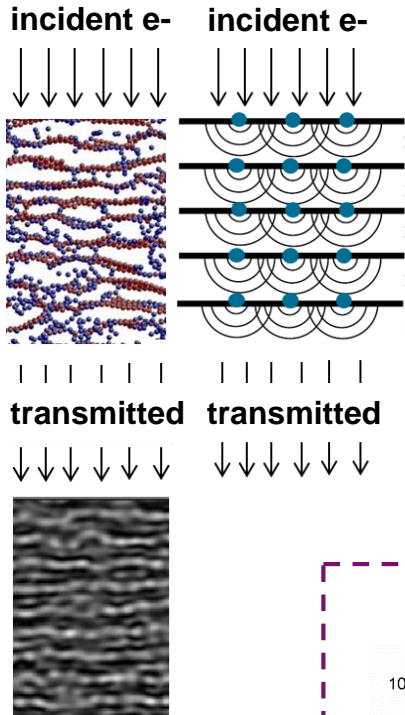
Dose effect by Frenkel pairs accumulation

- Creation of Frenkel pairs every 2 ps
high dose rate
- Dose up to 10 dpa
- constant pressure and constant temperature



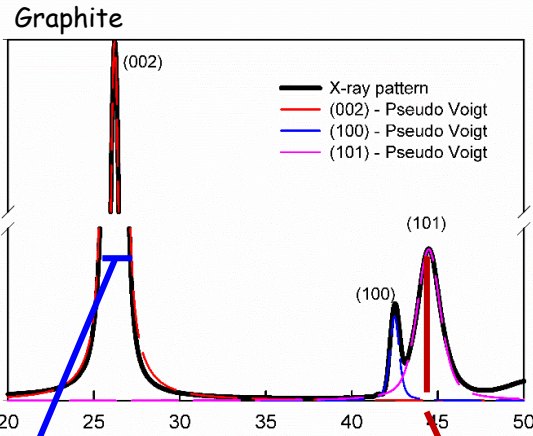
Frenkel pairs

HRTEM simulation (NCEMSS or Multi-slice code)



Graphite

XRD simulation (code Debyer)

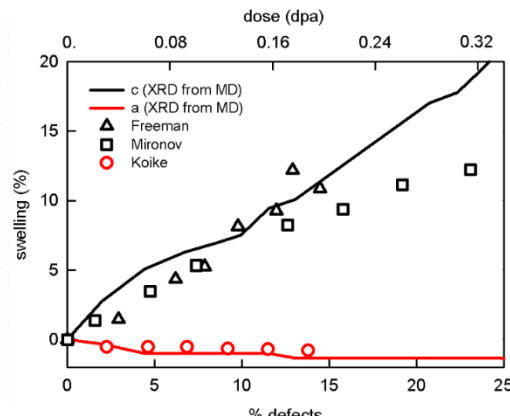
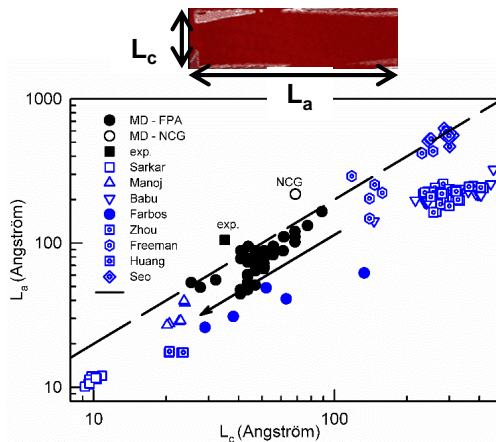


FWHM

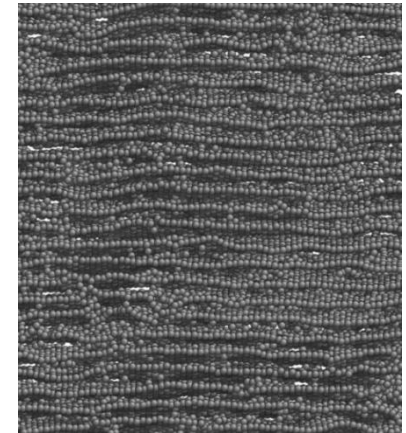
Peak position

Scherrer's formula

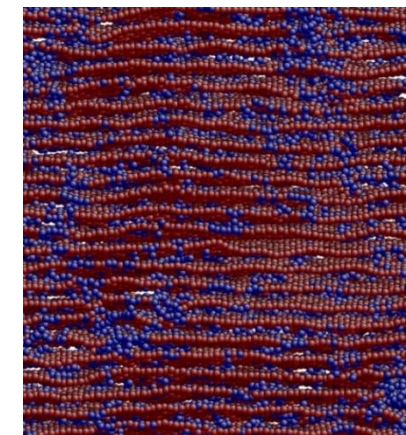
Bragg's law



Topological analysis: local curvature criterion (graphite if curvature > 32°)

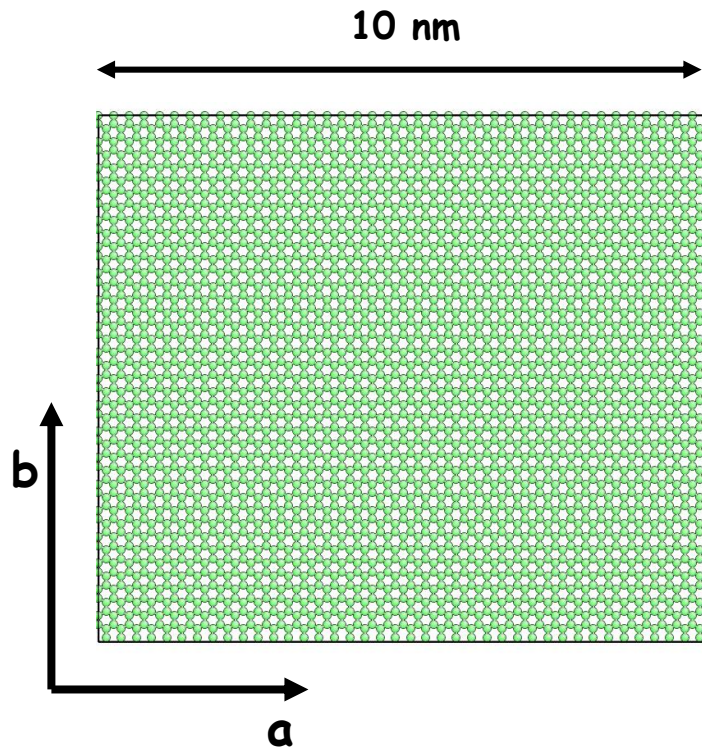


defects in blue



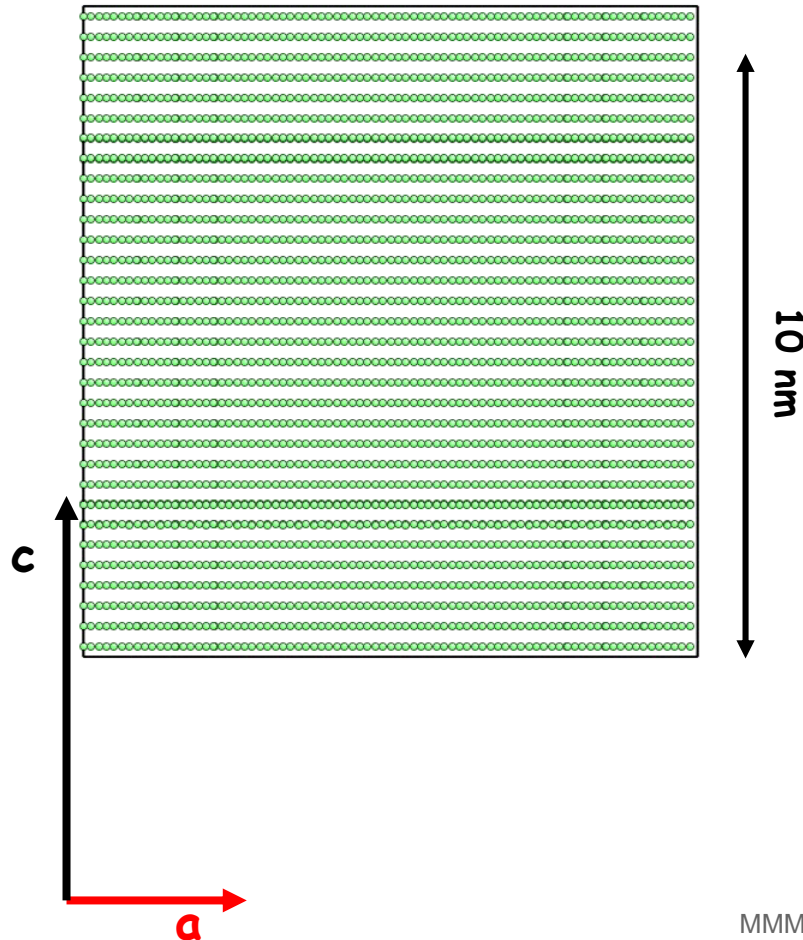
graphene sheets in red

Shrinking in the basal plane

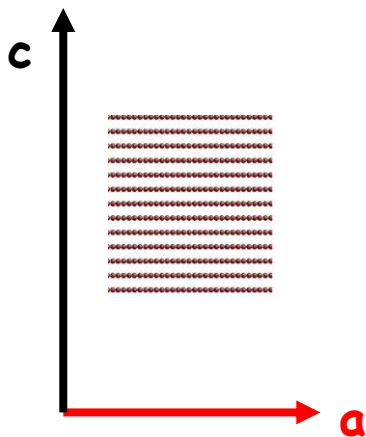
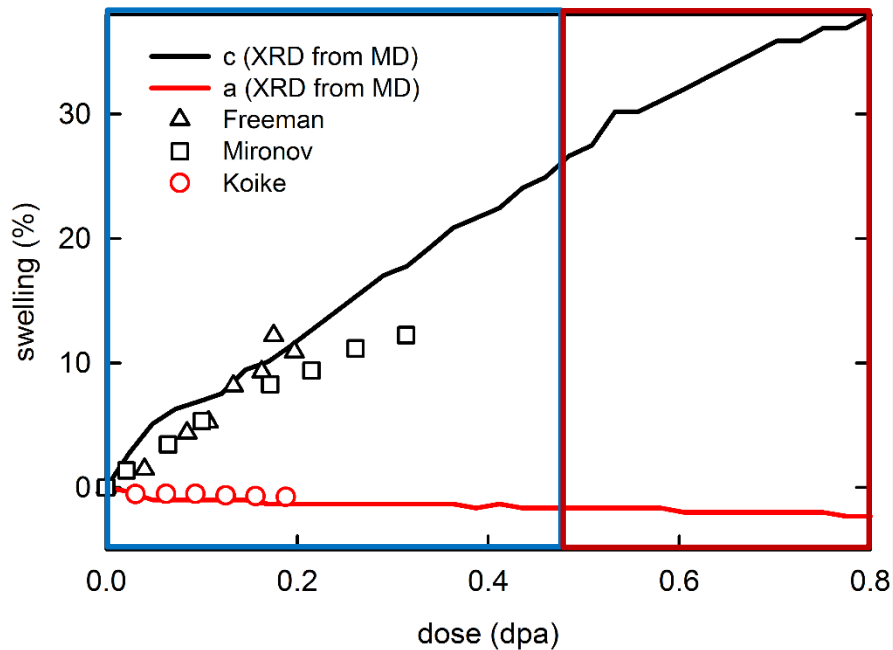


defects in blue
graphene sheets in red

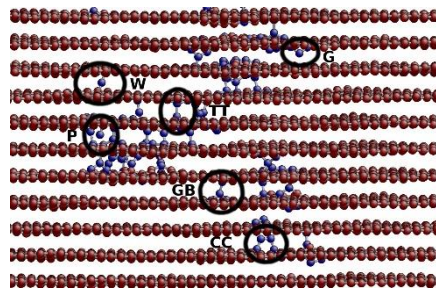
Swelling along c-axis



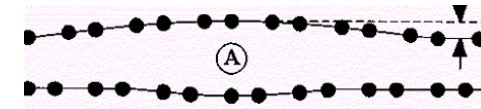
a and **c** from XRD - Bragg's law



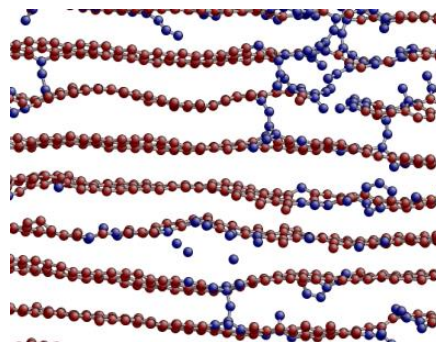
1. Point Defects



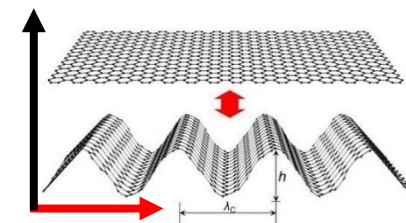
interstitial



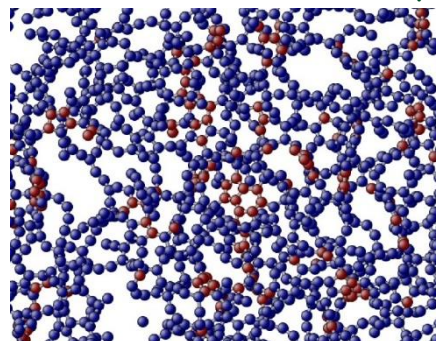
2. Pinning on small disordered cluster and Wrinkling of graphene sheets



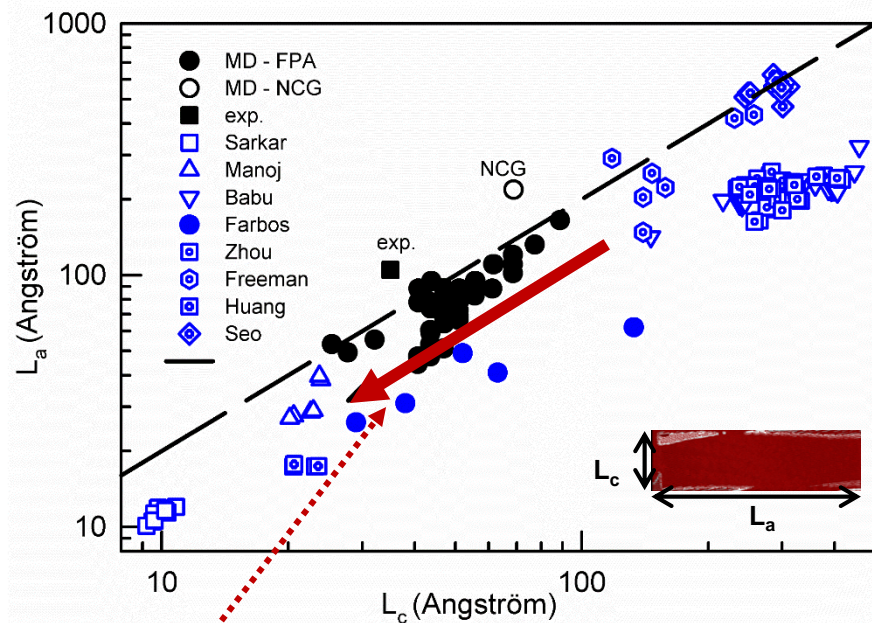
swelling / shrinking



3. Growth of amorphous pockets



L_a and L_c - Scherrer's formula



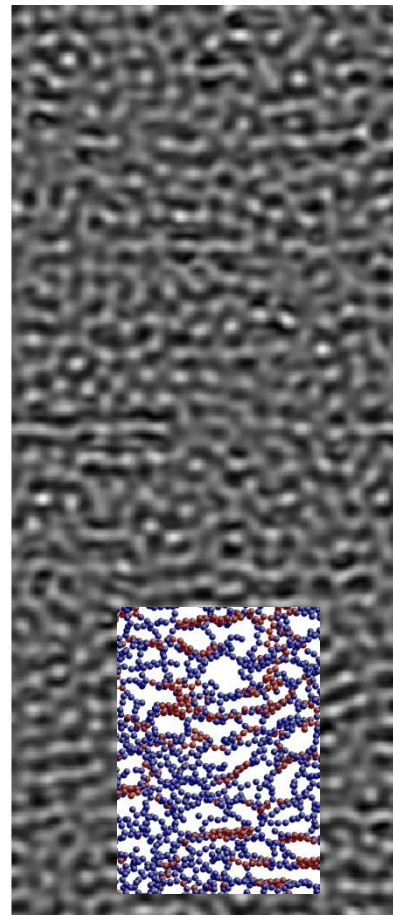
Decrease of graphene size with increasing dose

→ small graphene sheets in "noodles shape"
= BSU (Basic Structural Units)

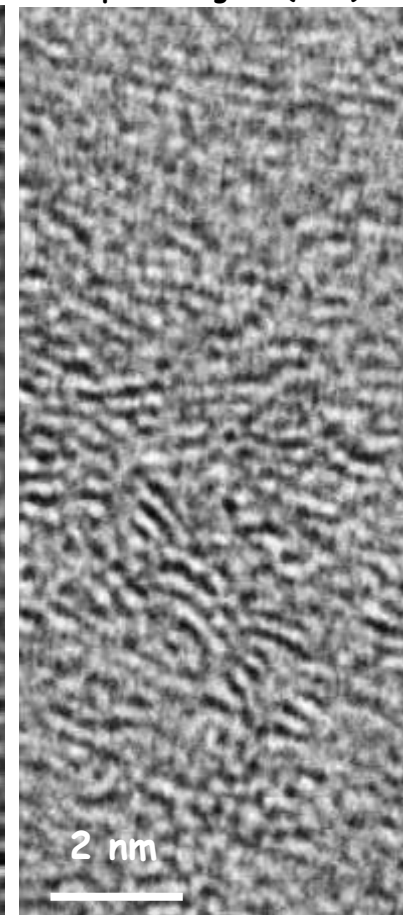
What simulations show:

graphite amorphizes

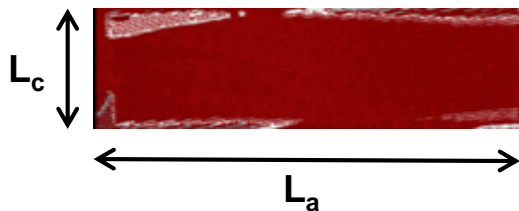
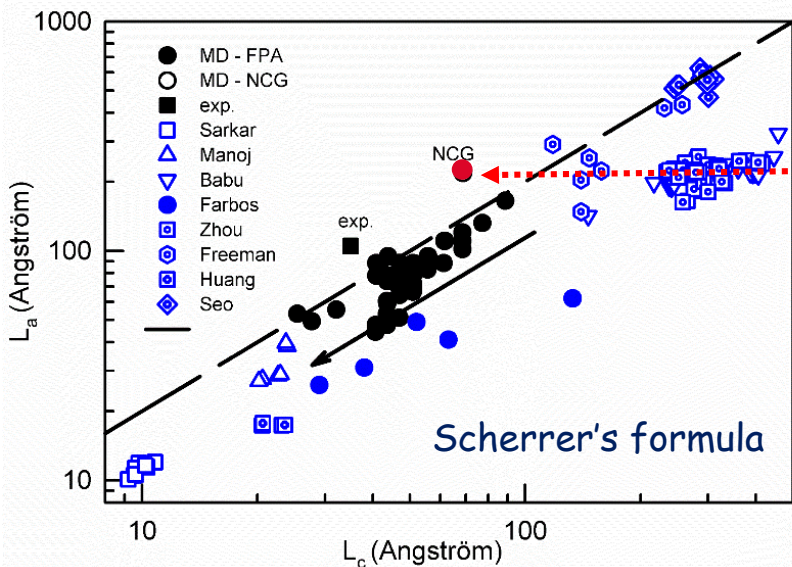
MD simulation



exp. J. Pageot (CEA)

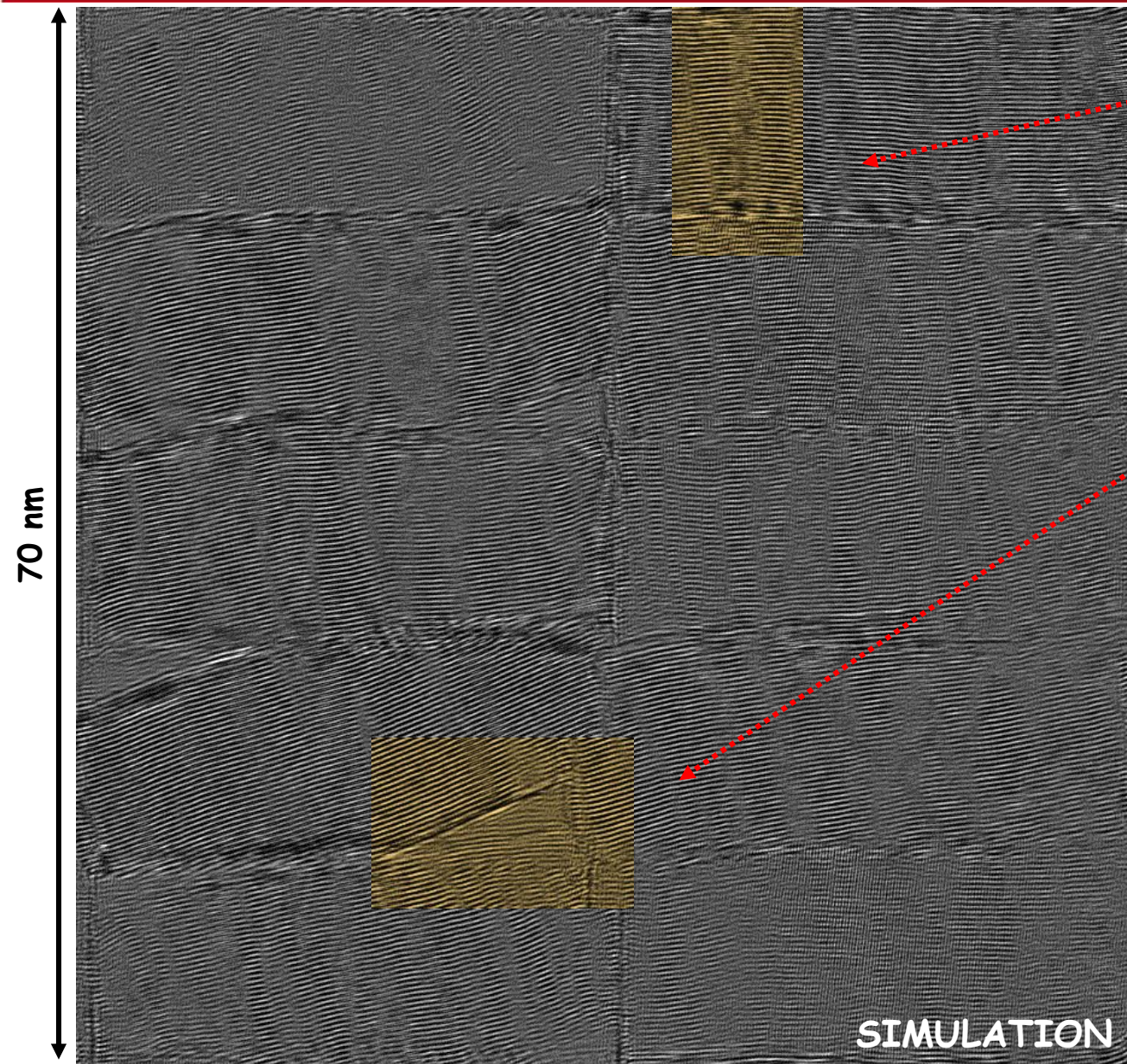


Creation of 20 crystallites, stacked along c-axis, with angles randomly distributed between 0° and 20°



Scherrer → $L_c = 7 \text{ nm}$, $L_a = 22 \text{ nm}$
 initial → $L_c = 15 \text{ nm}$, $L_a = 35 \text{ nm}$

Close to experiments

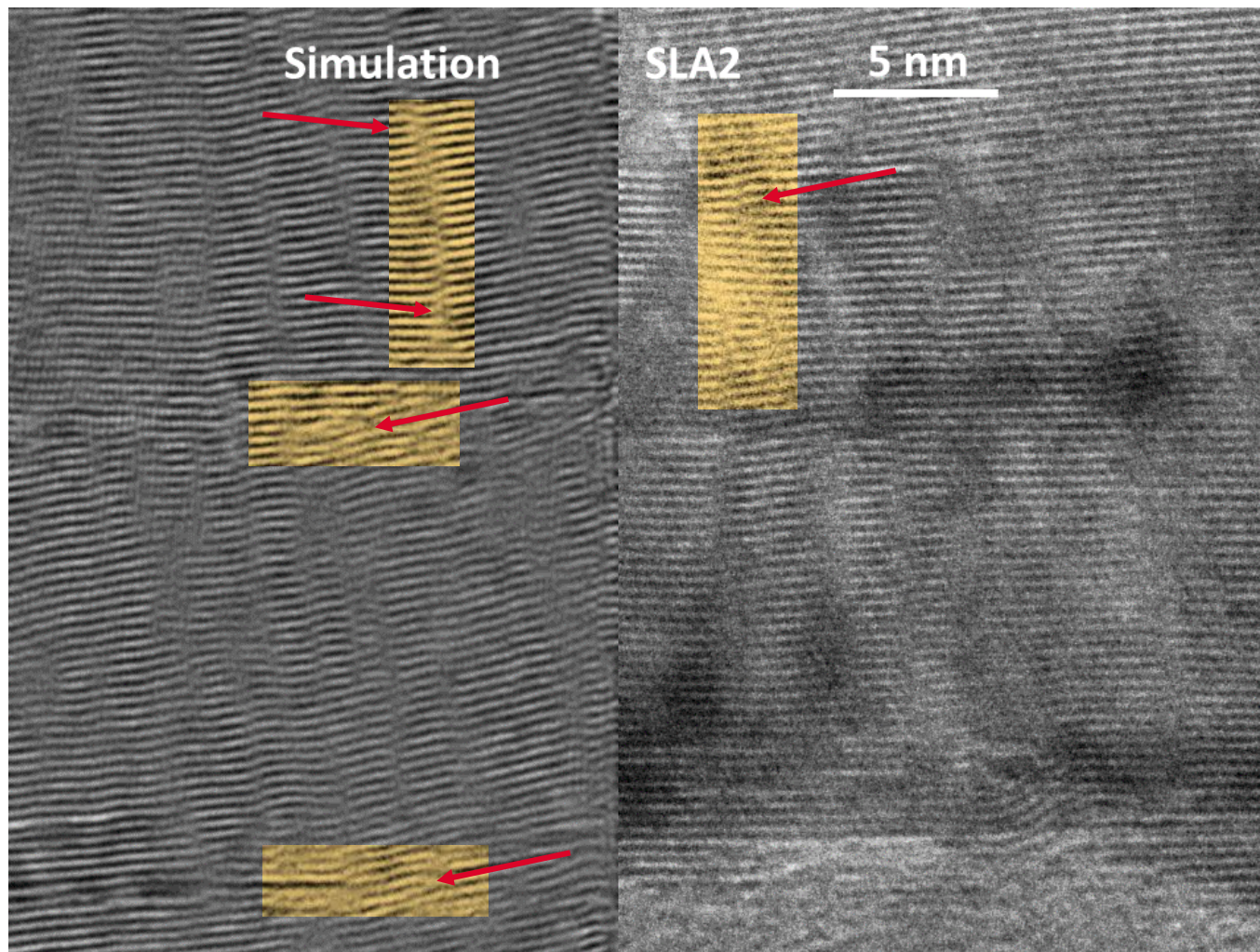


HRTEM image shows dislocation (edge or screw)

MD snapshot shows wrinkles

HRTEM image shows nano-pore

MD snapshot shows nano-pore



HRTEM images
→ dislocations

but

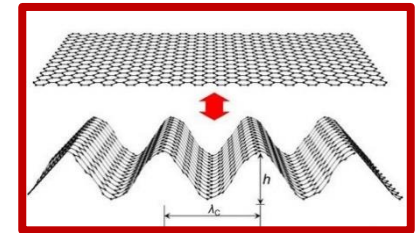
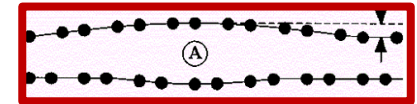
MD snapshots
→ wrinkles

Irradiation damages in graphite single crystal: Frenkel Pair Accumulation method

➤ Scenario in 3 steps:

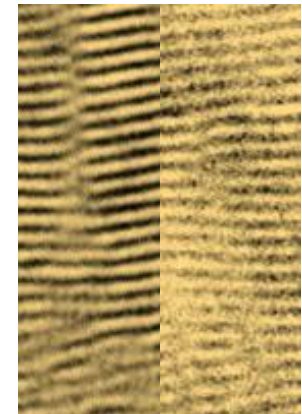
1. Point Defects
2. Pinning + Wrinkling of graphene sheets
3. Growth of amorphous pockets → Amorphisation

➤ Swelling due to both defects + wrinkling of graphene sheets



➤ Microstructure from XRD - HRTEM

1. Link dislocations - wrinkles ??
2. Basic Structural Units or amorphous??



Thanks for listening!

A. Chartier, L. Van Brutzel and J. Pageot, *Carbon* 133 (2018) 224.

A. Chartier, L. Van Brutzel, B. Pannier, and Ph. Baranek, *Carbon* 91 (2015) 395.

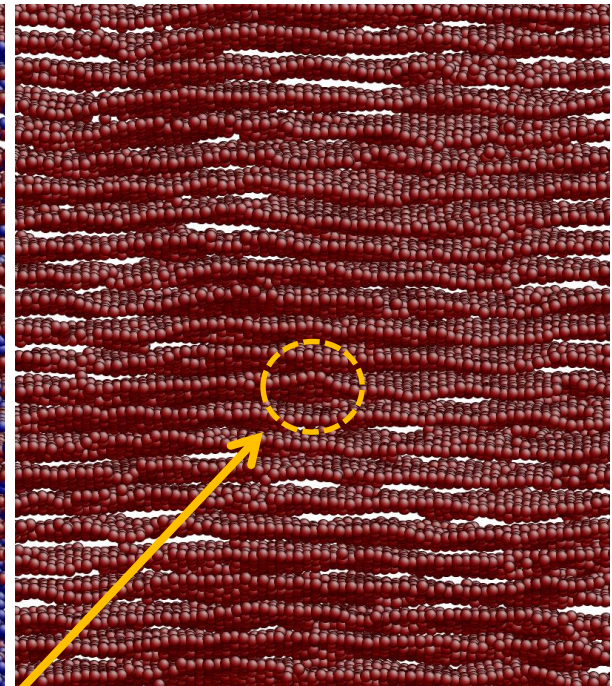
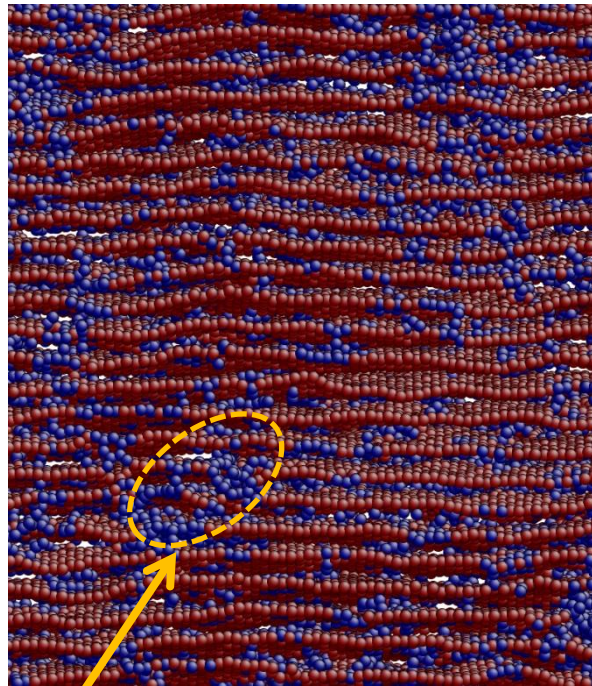
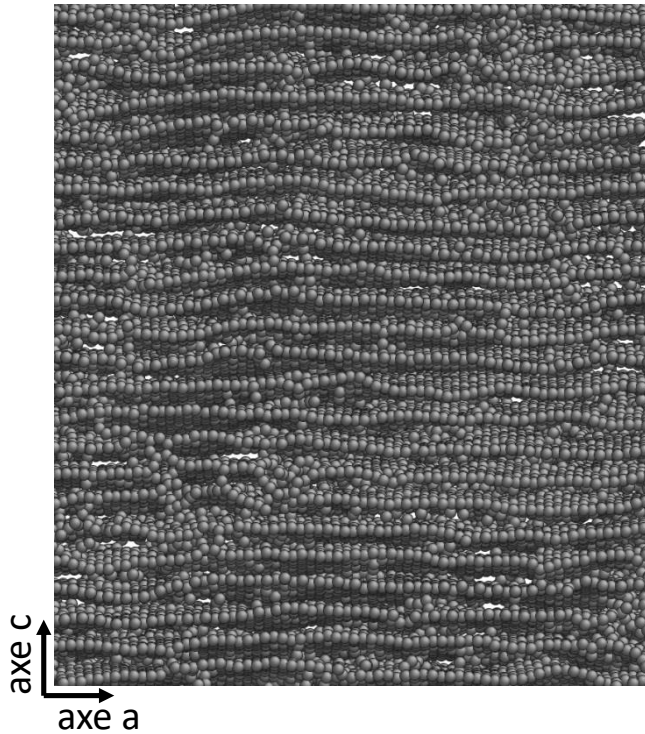
Alain Chartier
Commissariat à l'énergie atomique et aux énergies alternatives
DEN/DANS/DPC/SCCME/LM2T bât. 450SudEsi
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 31 68 | F. +33 (0)1 69 08 92 21

Direction de l'énergie Nucléaire
Département de Physico-Chimie
Service de Corrosion et du
Comportement des Matériaux
dans leur Environnement

0.01 dpa

Local curvature threshold

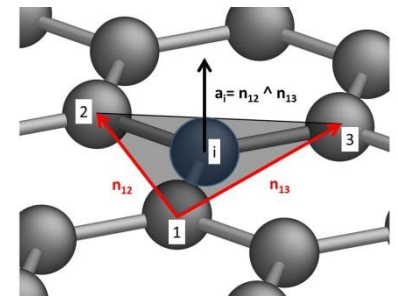
Remaining graphene sheets

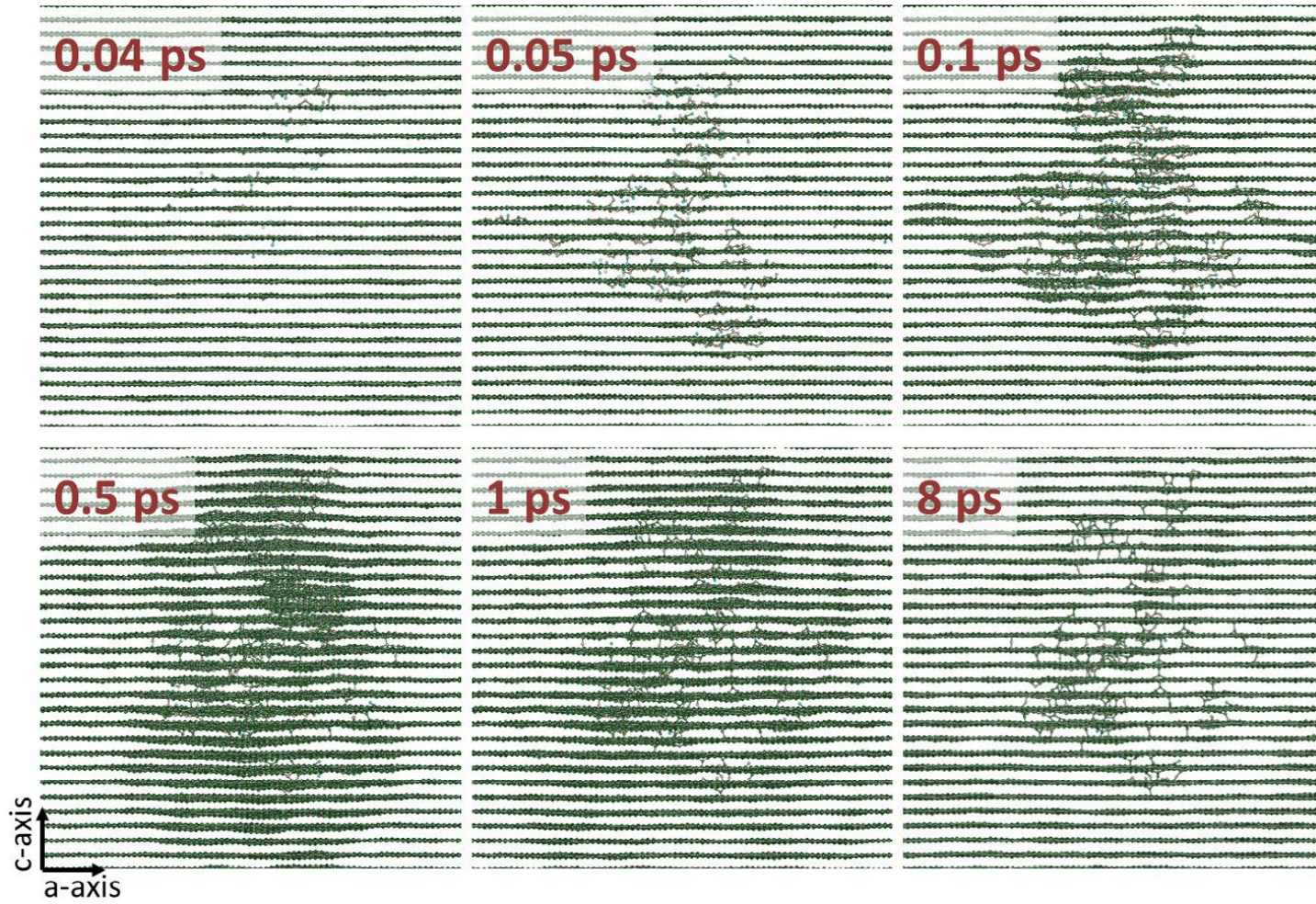


Plans pinned by **disordered amorphous** zones

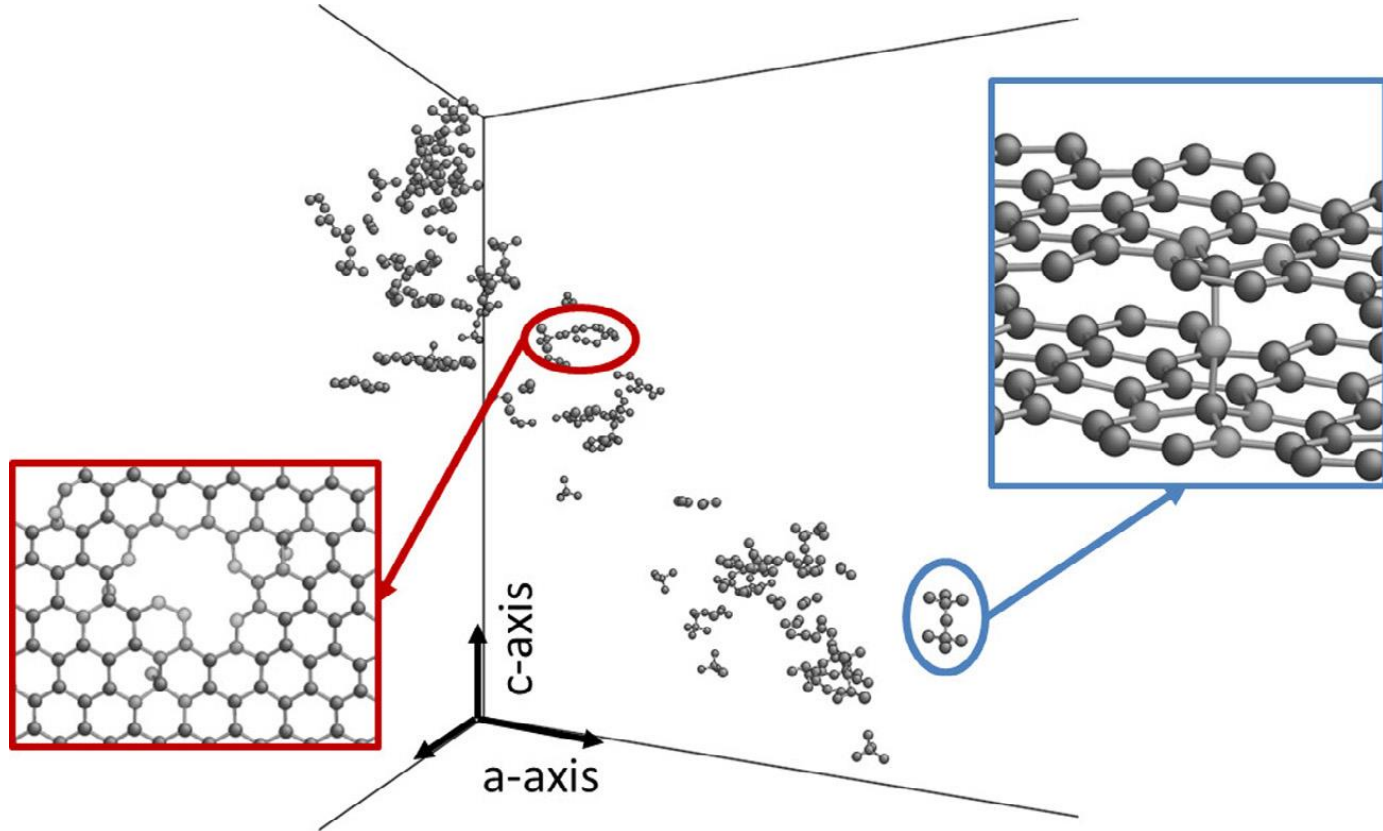
Graphene sheets are discriminated by **local curvature**

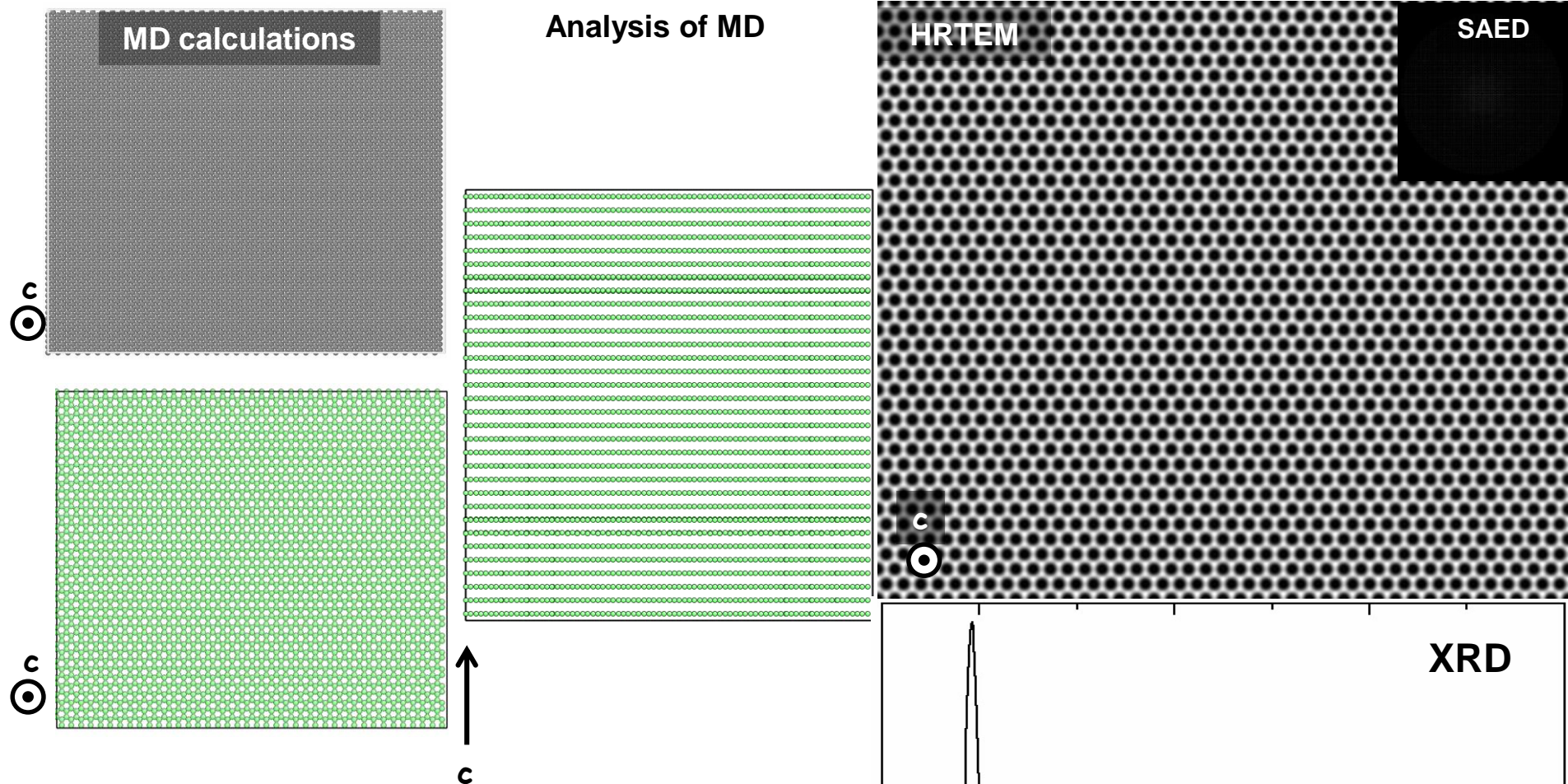
- average curvature radius
- size of graphene clusters



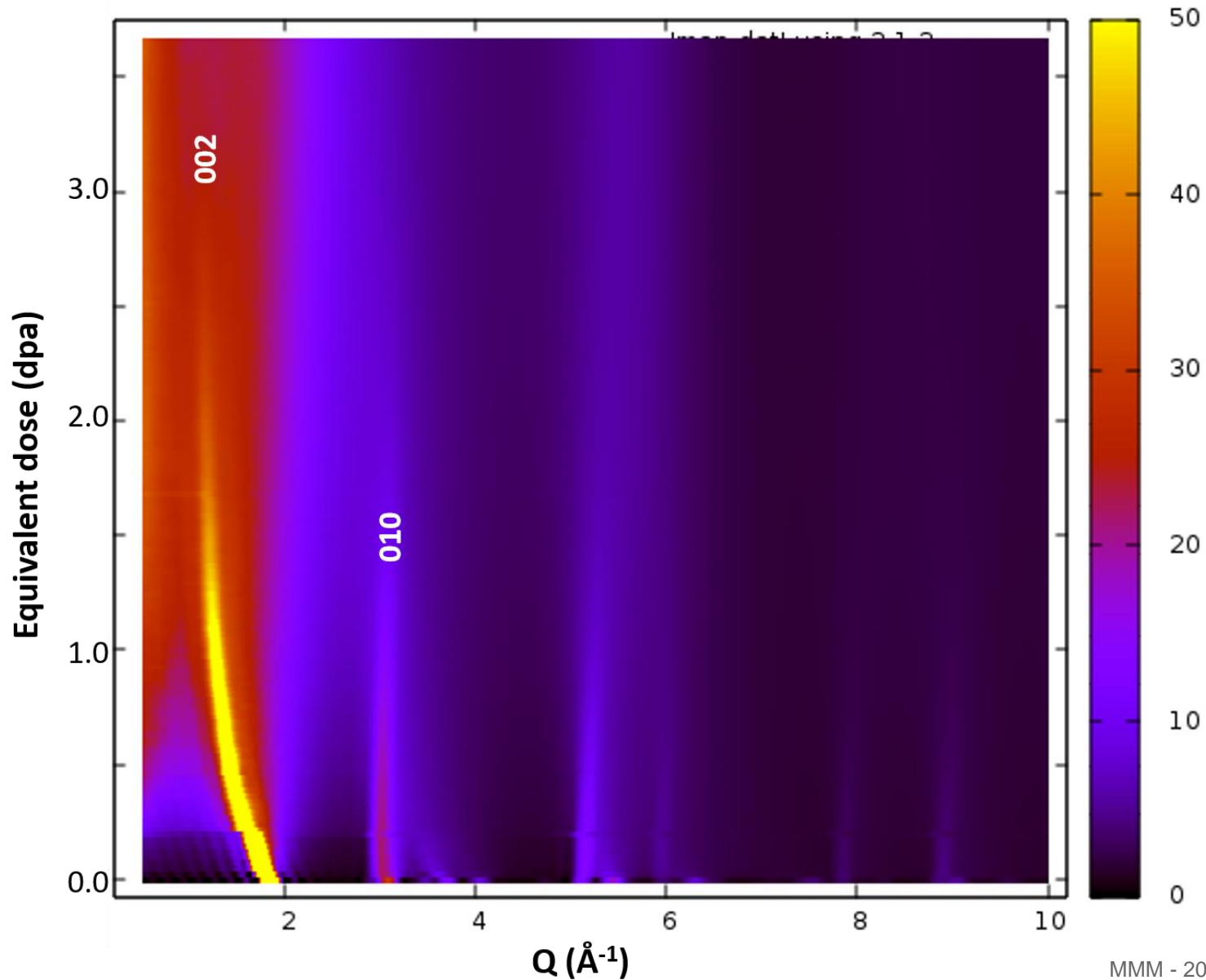


Graphite – displacement cascade

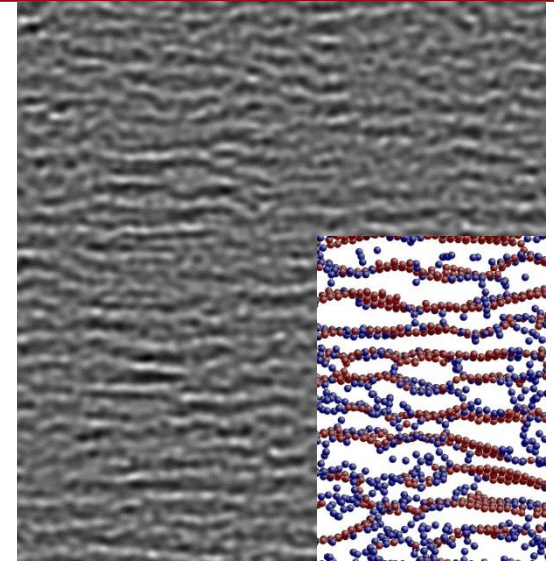
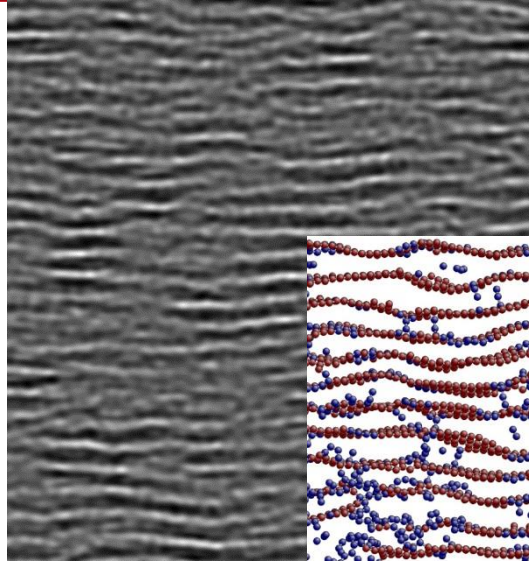
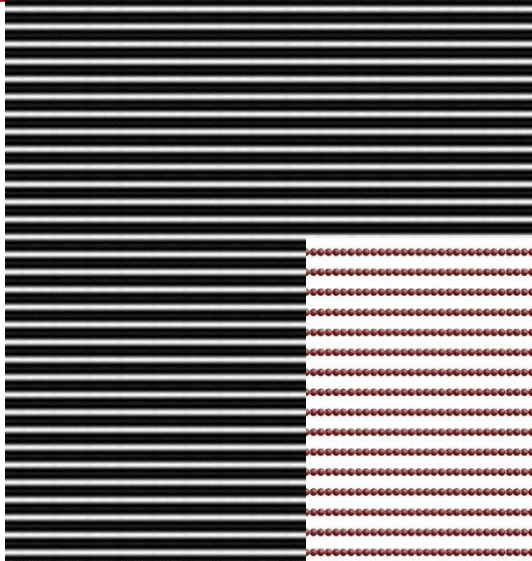




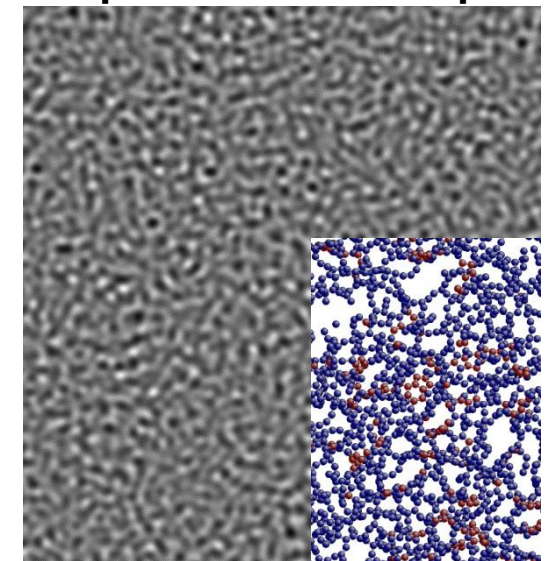
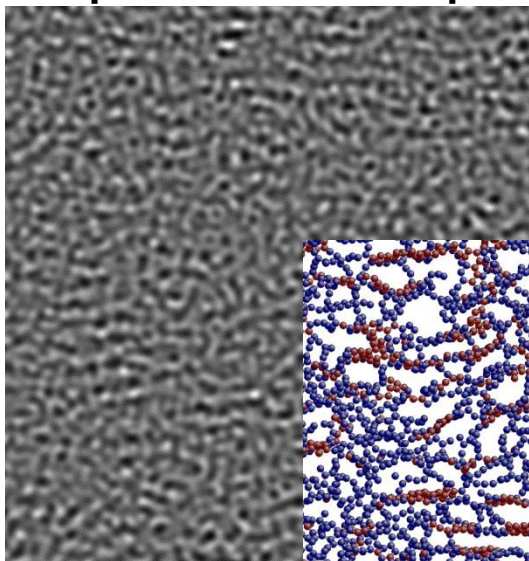
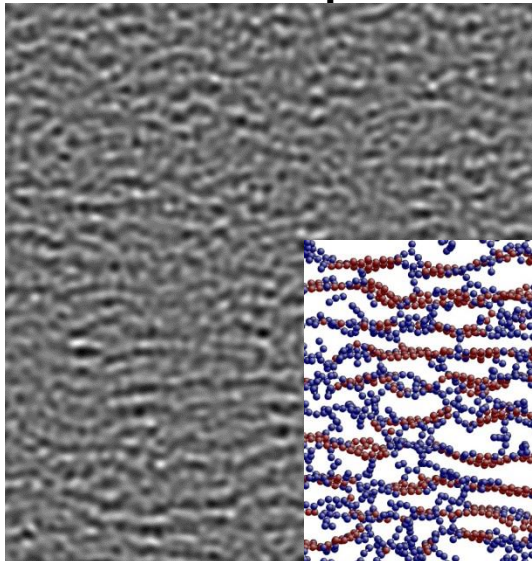
- Visualization of defects in graphene planes (MD and HRTEM)
- Visualization of wrinkles (MD analysis)
- Swelling from size of box (not easy from XRD)
- amorphization at 0.10 dpa (SAED and XRD)



5. nm



5. nm



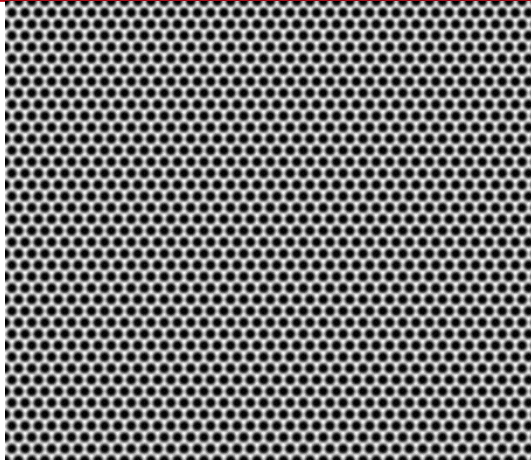
equivalent to 1.2 dpa

equivalent to 2.50 dpa

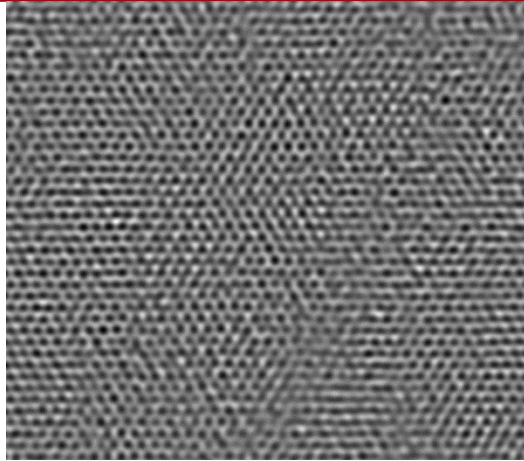
equivalent to 3.50 dpa

Graphite – simulated HRTEM

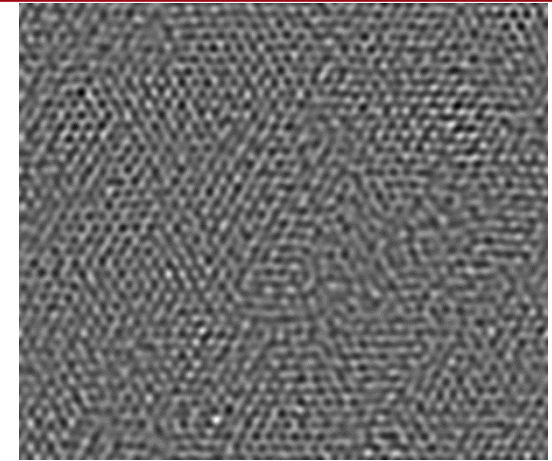
5. nm



0.00 dpa

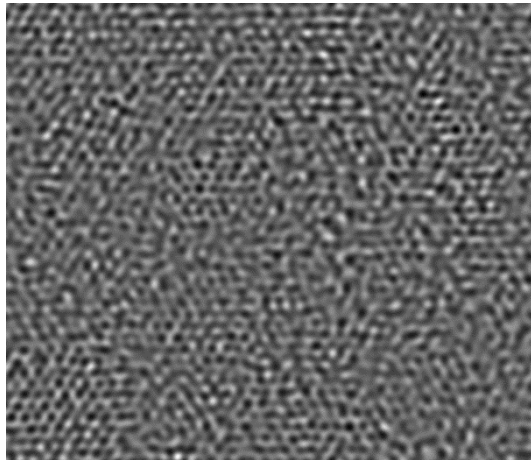


equivalent to 0.50 dpa

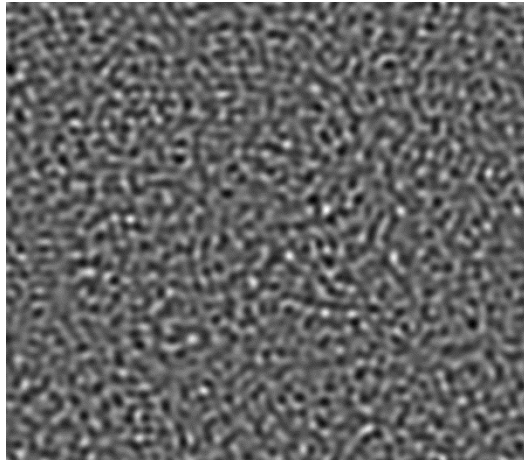


equivalent to 1.00 dpa

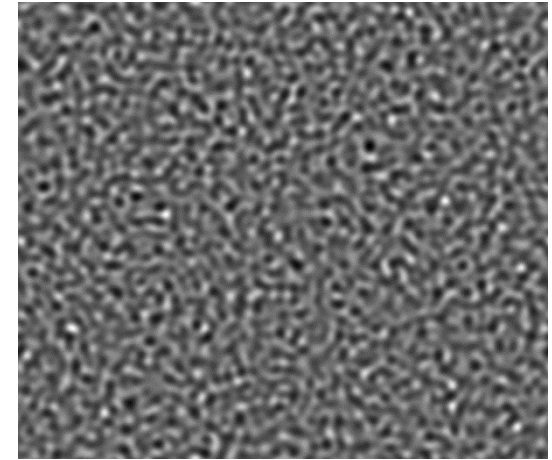
5. nm



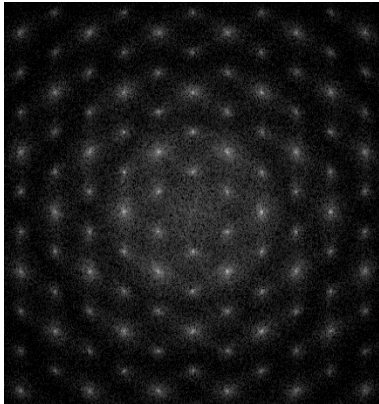
equivalent to 1.2 dpa



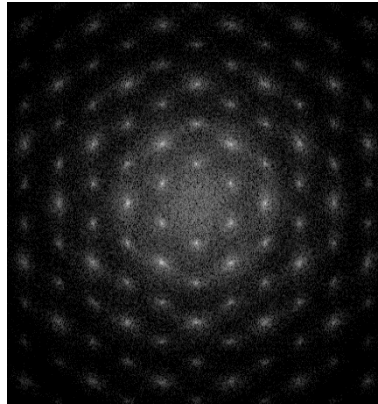
equivalent to 2.50 dpa



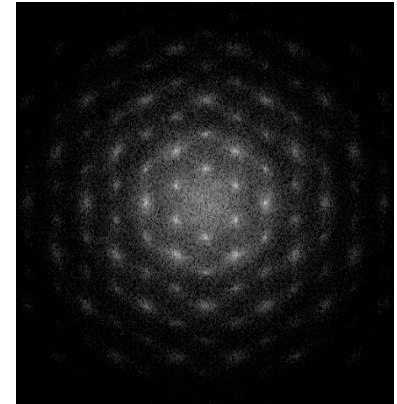
equivalent to 3.50 dpa



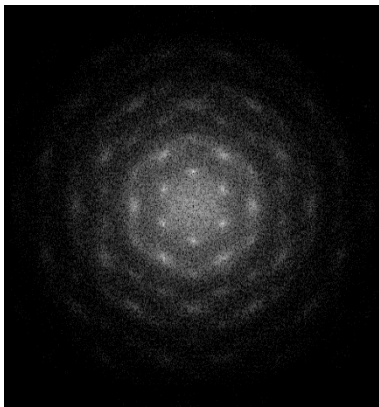
0.00 dpa



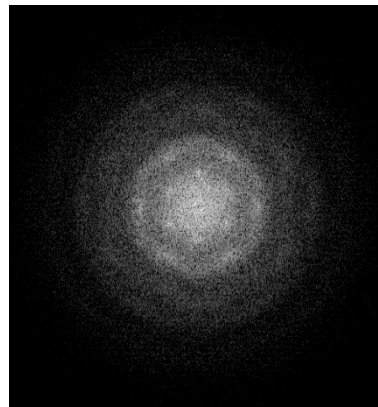
equivalent to 0.50 dpa



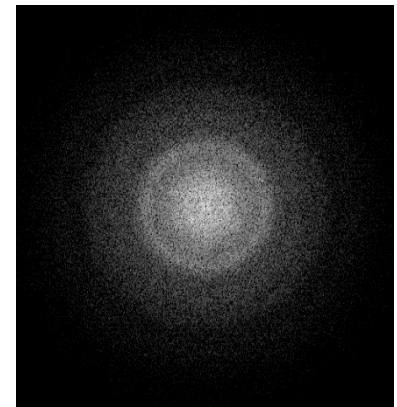
equivalent to 1.00 dpa



equivalent to 1.2 dpa



equivalent to 2.50 dpa



equivalent to 3.50 dpa