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# Irradiation damage in nuclear graphite at the atomic scale

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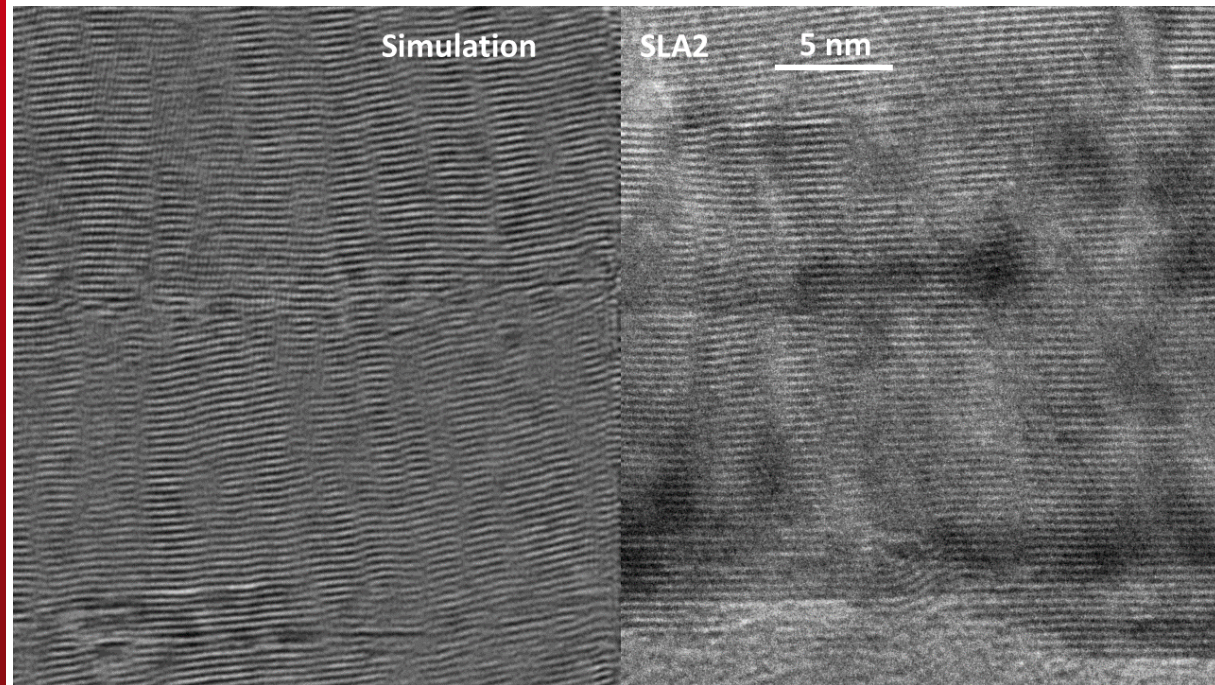
Submitted on 13 Dec 2019

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# Irradiation damage in nuclear graphite at the atomic scale

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



DE LA RECHERCHE À L'INDUSTRIE  
**cea den**

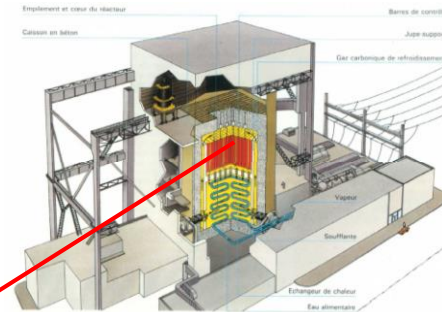
**université**  
PARIS-SACLAY



## 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  $^{14}\text{C}$  &  $^{36}\text{Cl}$  ( $\frac{1}{2}$  life 5 500 &  $3 \times 10^5$  years)



## Foreseen Solutions:

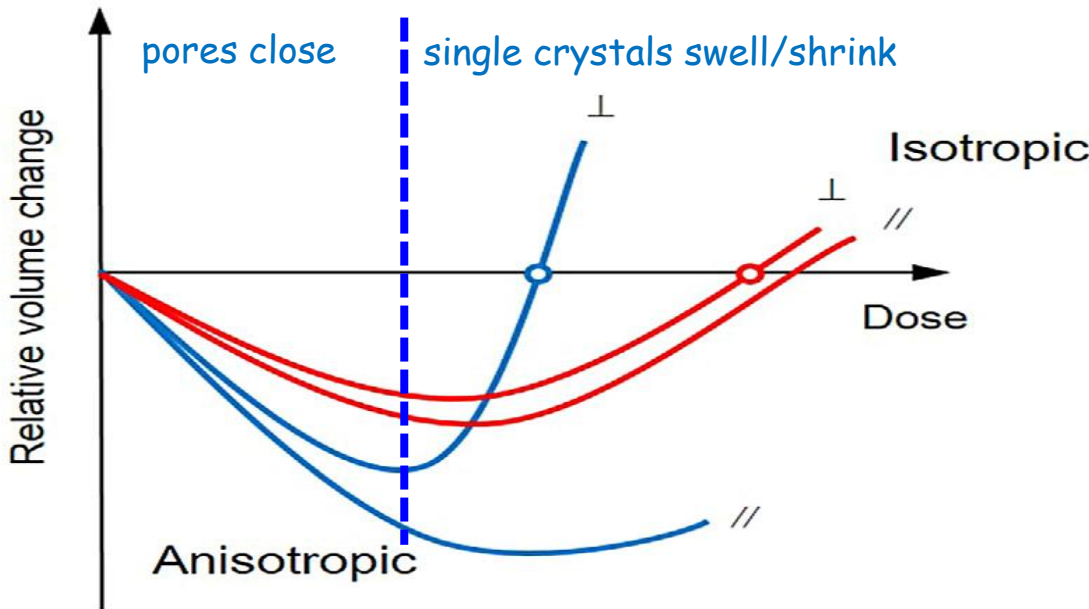
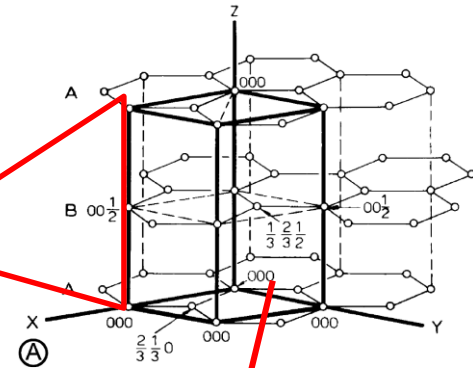
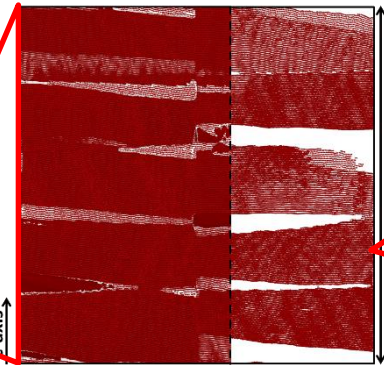
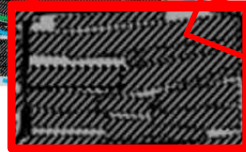
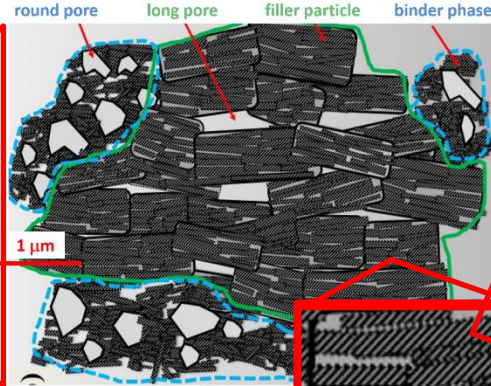
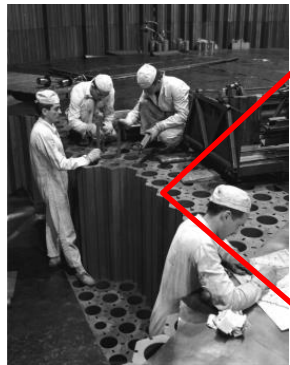
- Decommissioning under water
- Treatment to eliminate  $^{14}\text{C}$  &  $^{36}\text{Cl}$ , ( $^3\text{H}$ )

} interaction: graphite  $\leftrightarrow$   $\text{H}_2\text{O}$ ,  $\text{O}_2$ ,  $\text{CO}_2$ ...  
depends on the **localization of  $^{14}\text{C}$  &  $^{36}\text{Cl}$**  after irradiation

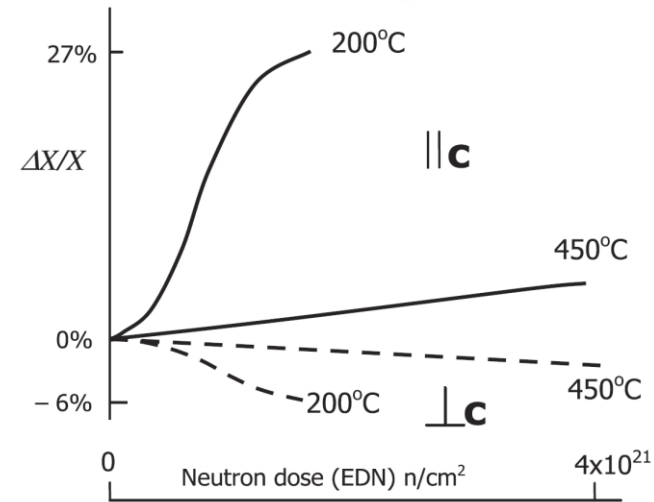
- **Graphite microstructure evolution under irradiation?**
  - - radiation damages and swelling
  - - link between XRD - HRTEM images and atomic scale structure

cm

nm



Shen et al (2015)



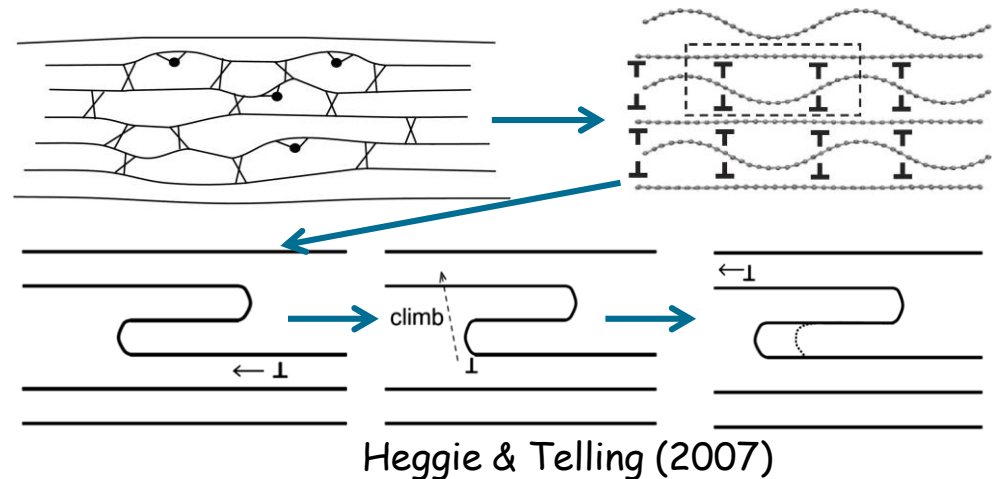
Heggie et al (2011)

## Scenario of swelling

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

### Based on:

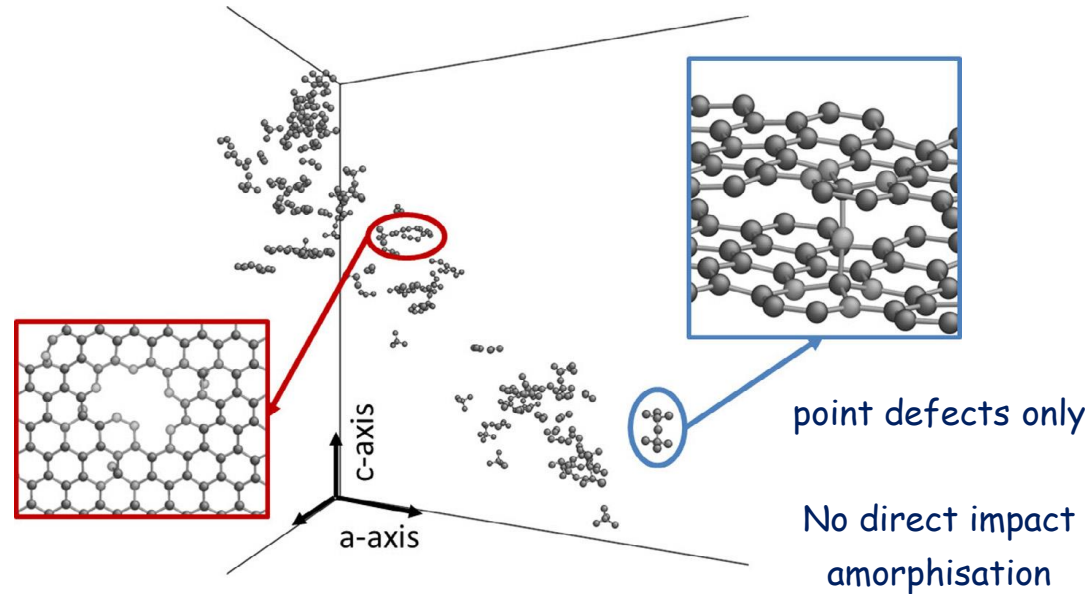
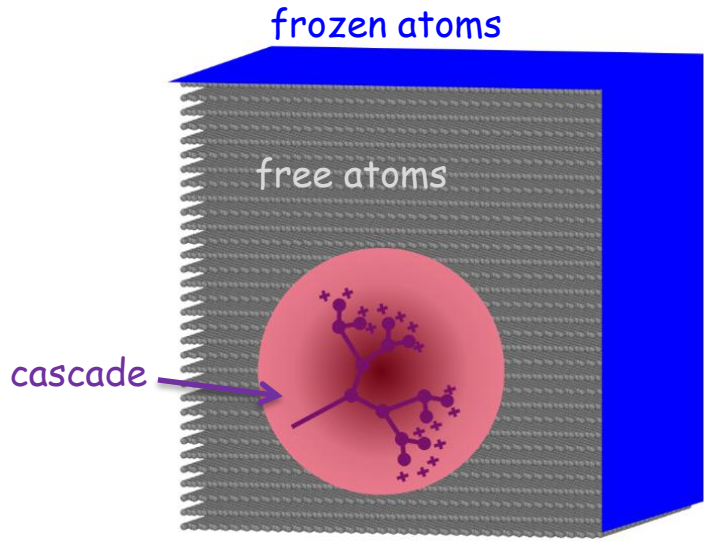
- HRTEM observations
- Atomic scale (DFT - MD) calculations
- Wigner energy estimations



### 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

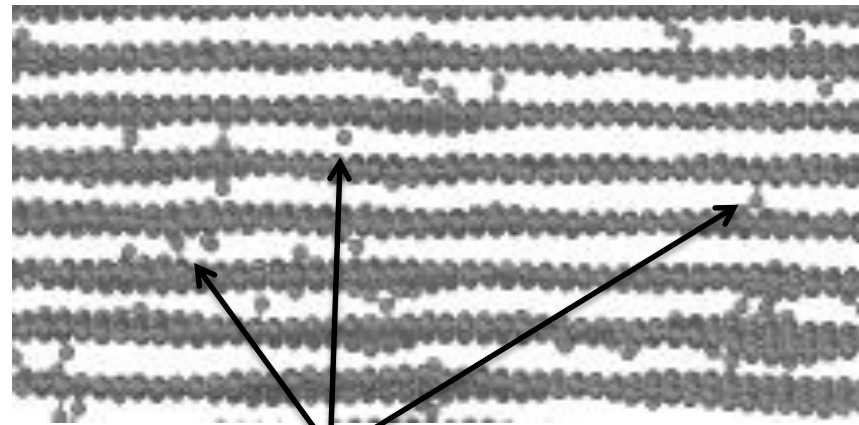
## Primary ballistic damage by displacement cascade, with primary knocked on atom with 0.5 to 10 keV



if point defects then

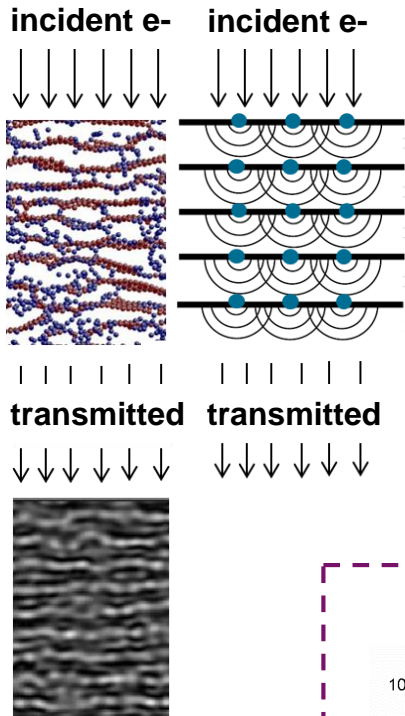
## Dose effect by Frenkel pairs accumulation

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



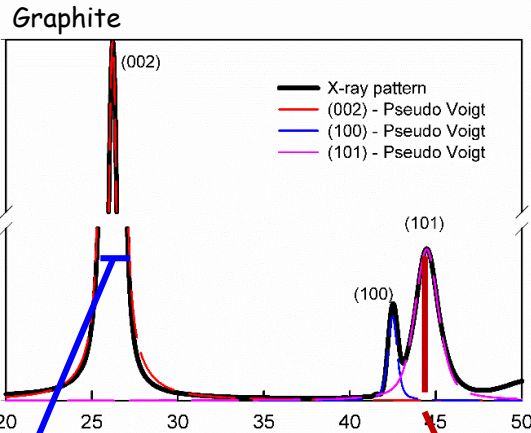
Frenkel pairs

## HRTEM simulation (NCEMSS or Multi-slice code)



Graphite

## XRD simulation (code Debyer)

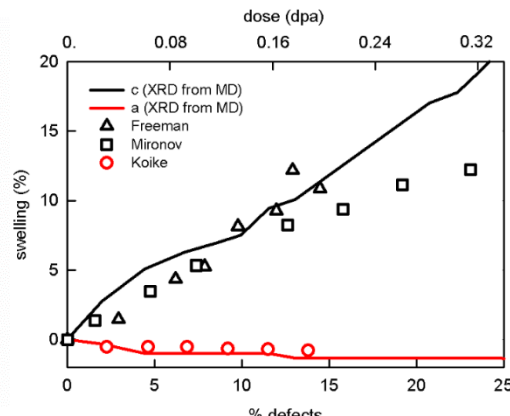
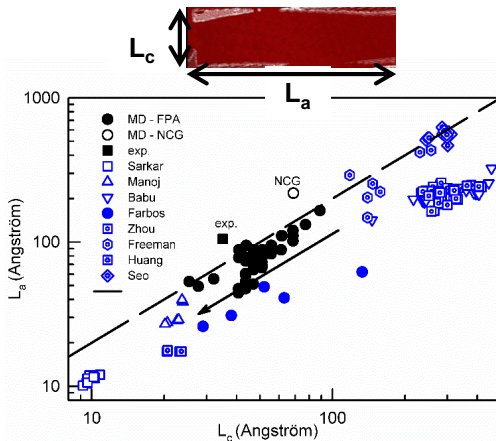


FWHM

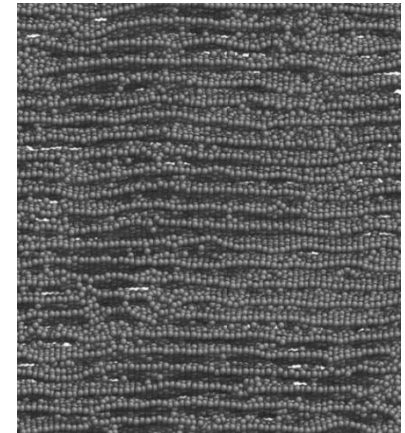
Peak position

Scherrer's formula

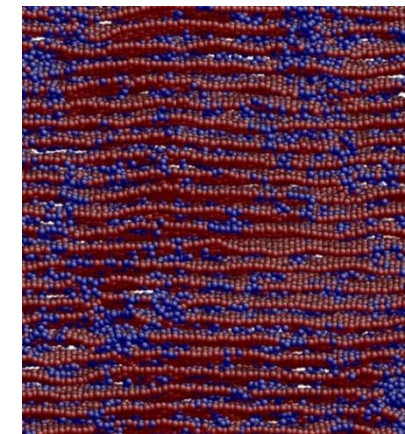
Bragg's law



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

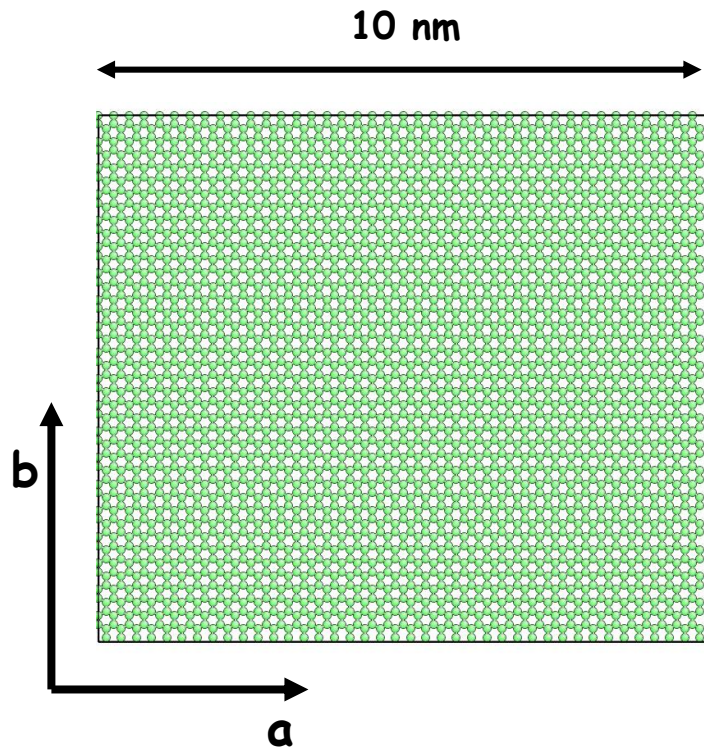


defects in blue



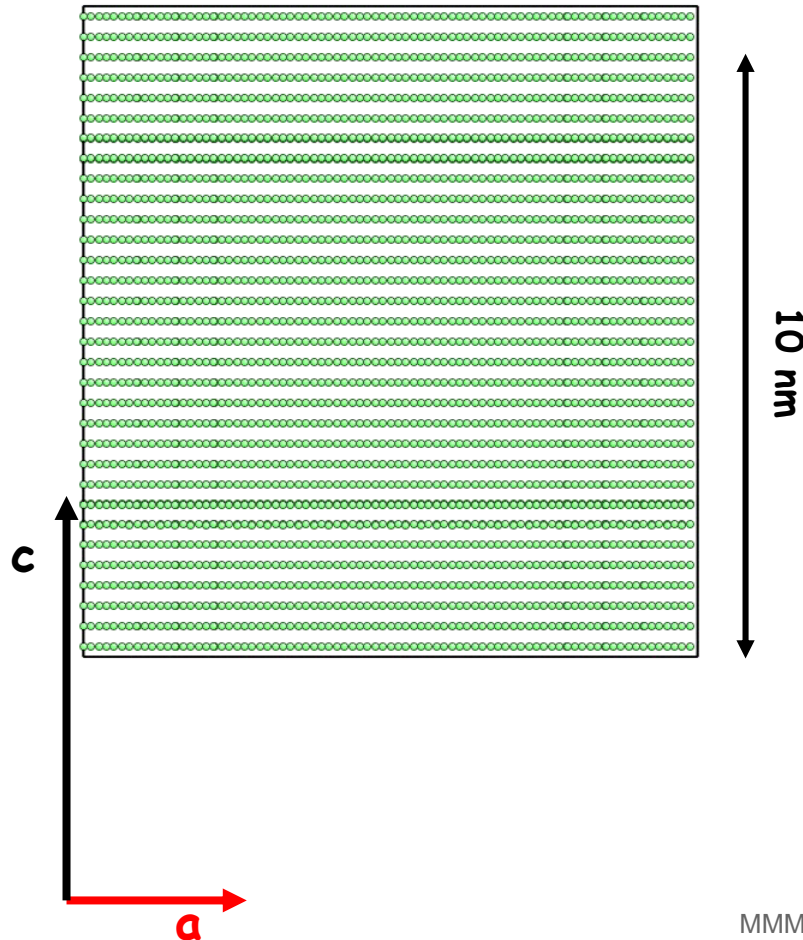
graphene sheets in red

Shrinking in the basal plane



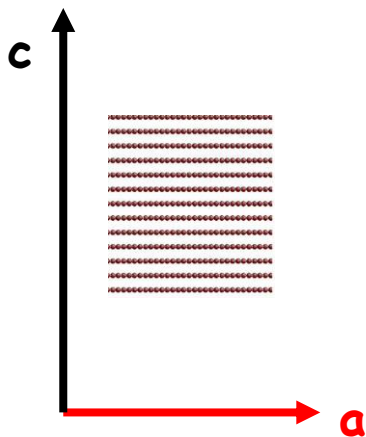
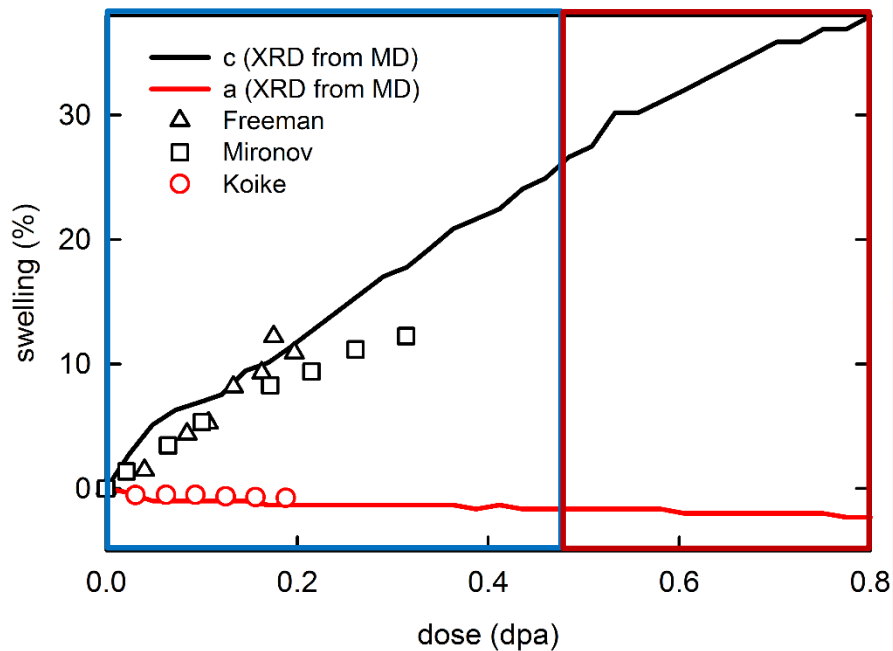
defects in blue  
graphene sheets in red

Swelling along c-axis

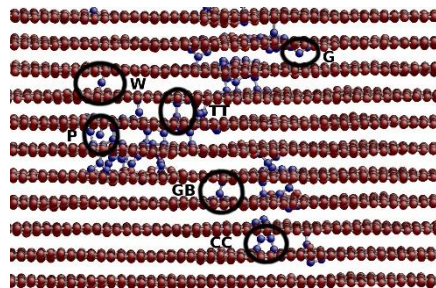




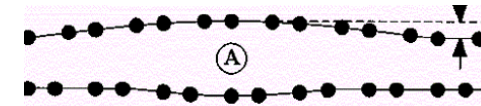
**a** and **c** from XRD - Bragg's law



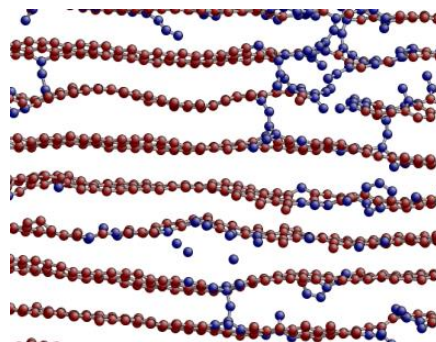
## 1. Point Defects



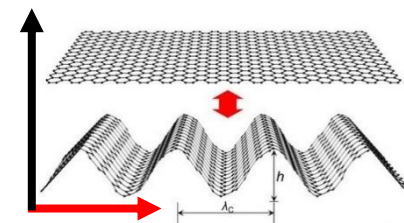
interstitial



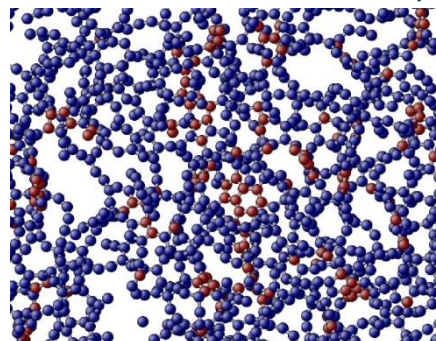
## 2. Pinning on small disordered cluster and Wrinkling of graphene sheets



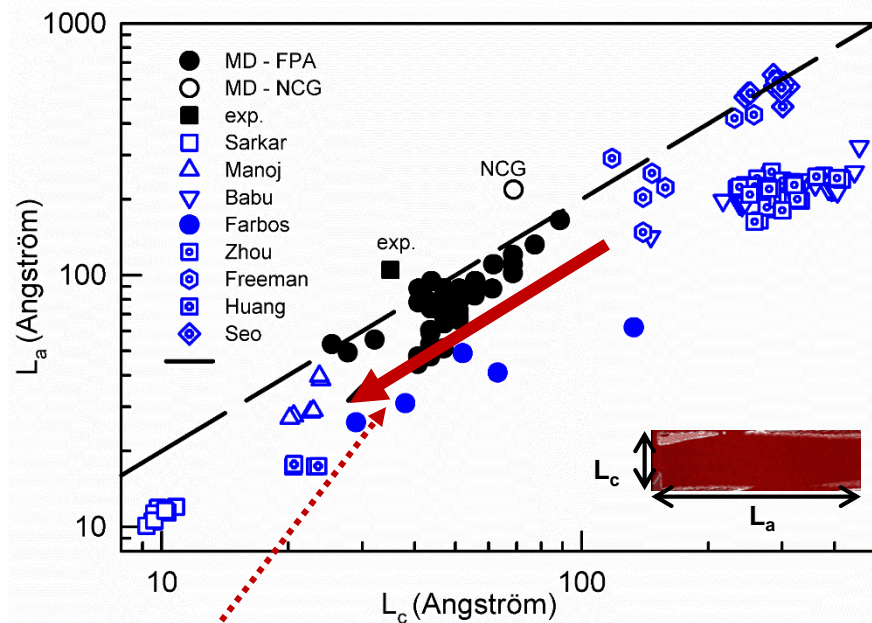
swelling / shrinking



## 3. Growth of amorphous pockets



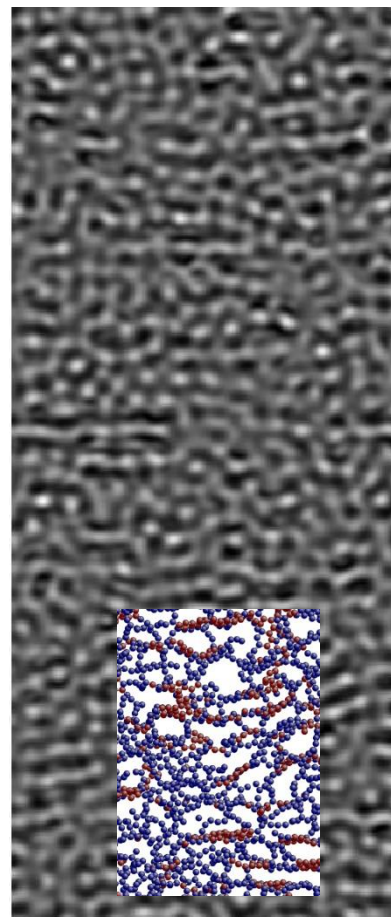
## La and Lc - Scherrer's formula



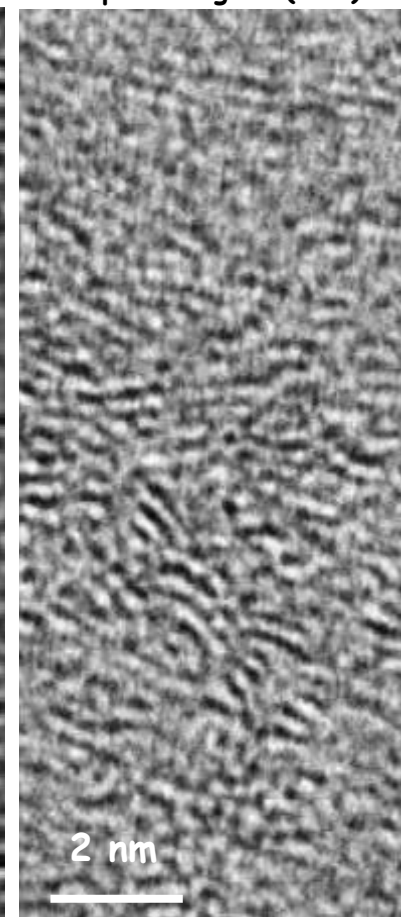
Decrease of graphene size with increasing dose

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

MD simulation



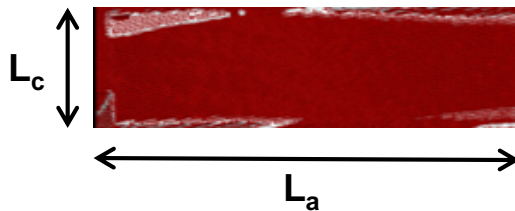
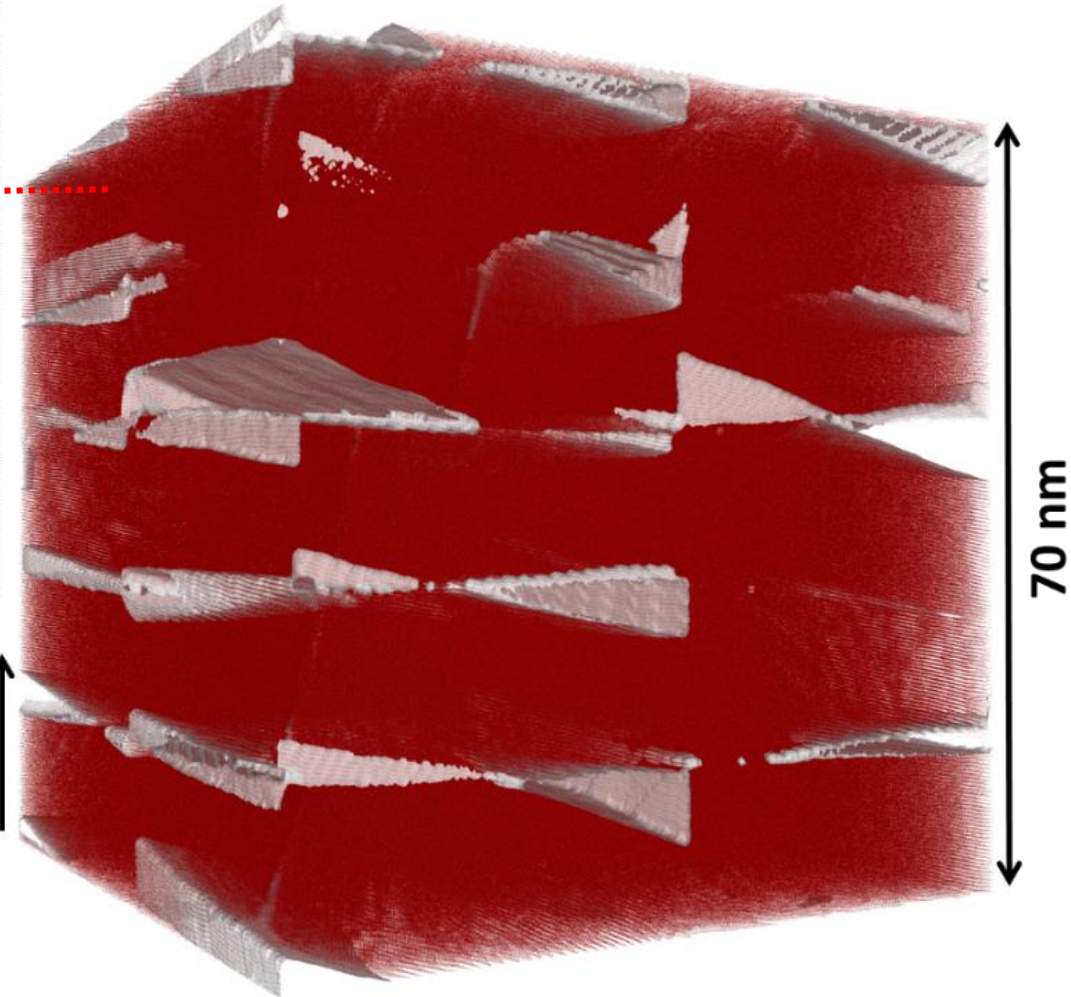
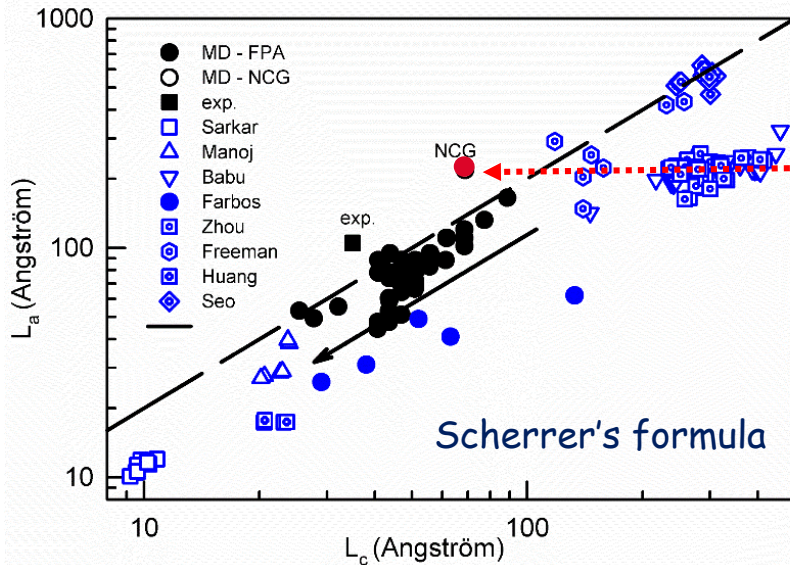
exp. J. Pageot (CEA)



What simulations show:

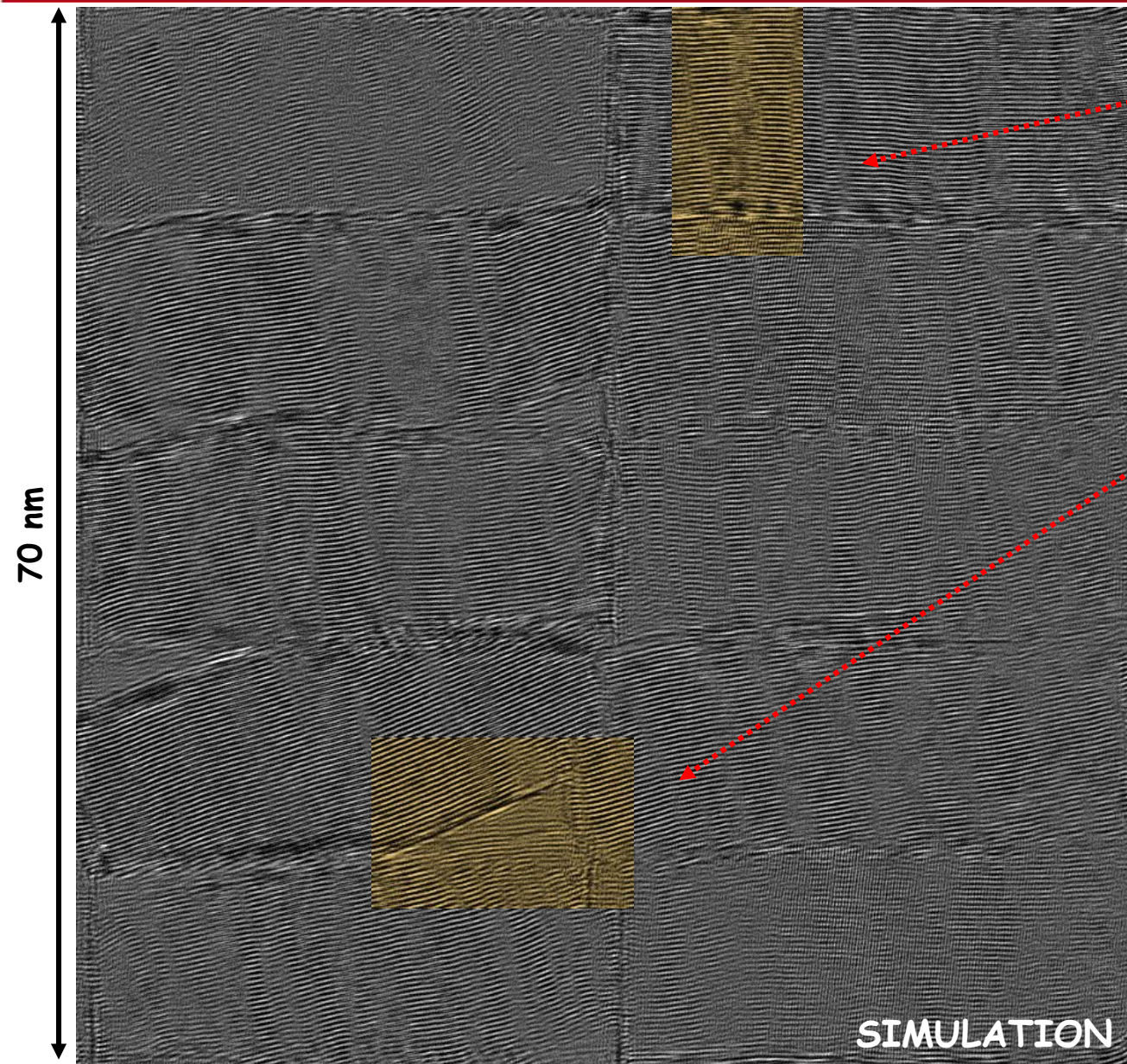
graphite amorphizes

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

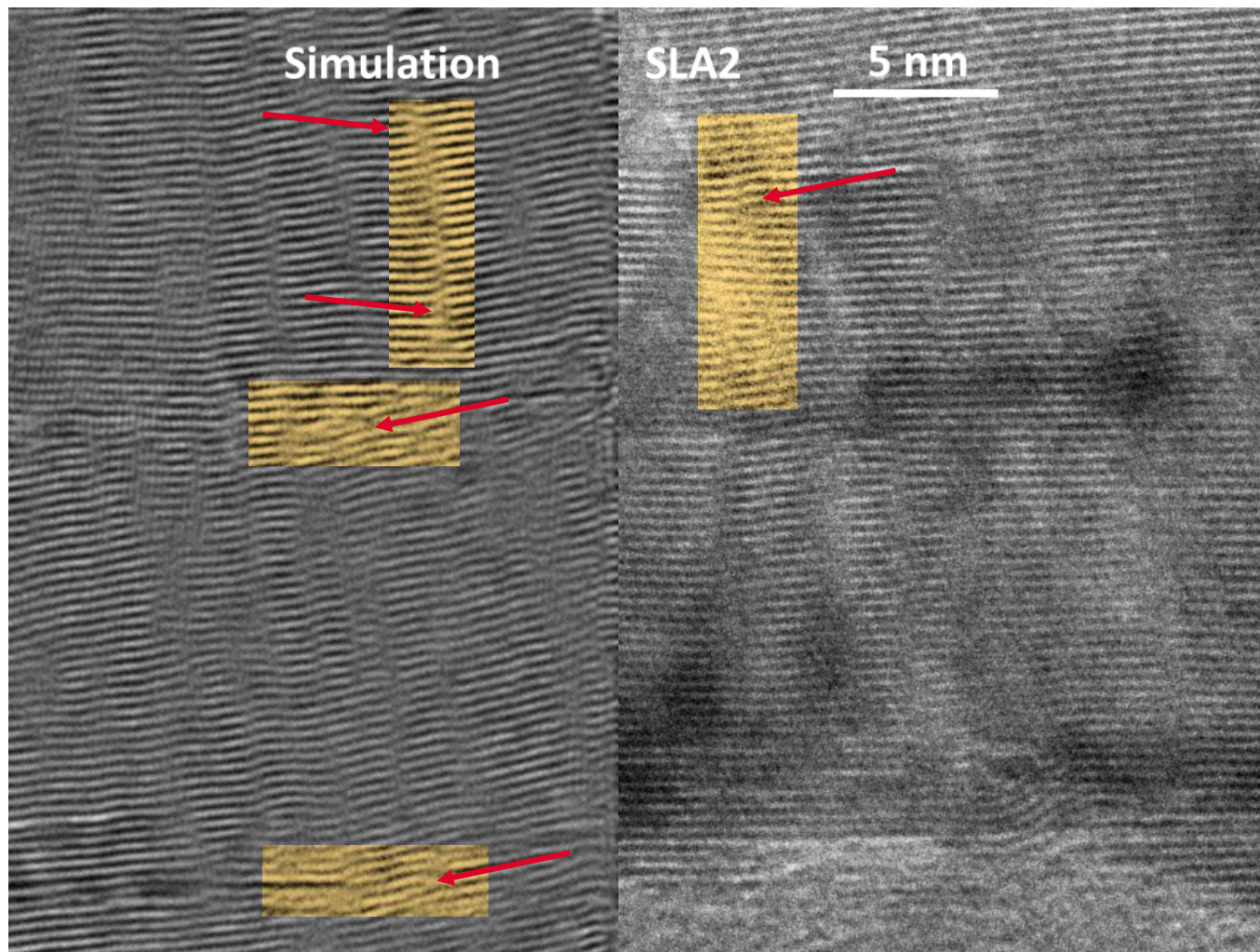


HRTEM image shows dislocation (edge or screw)

MD snapshot shows wrinkles

HRTEM image shows nano-pore

MD snapshot shows nano-pore



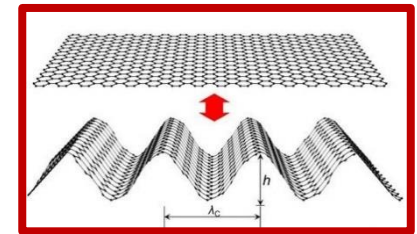
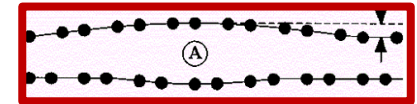
HRTEM images  
→ dislocations

**but**

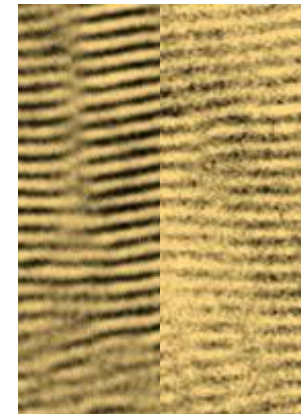
MD snapshots  
→ wrinkles

## 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.

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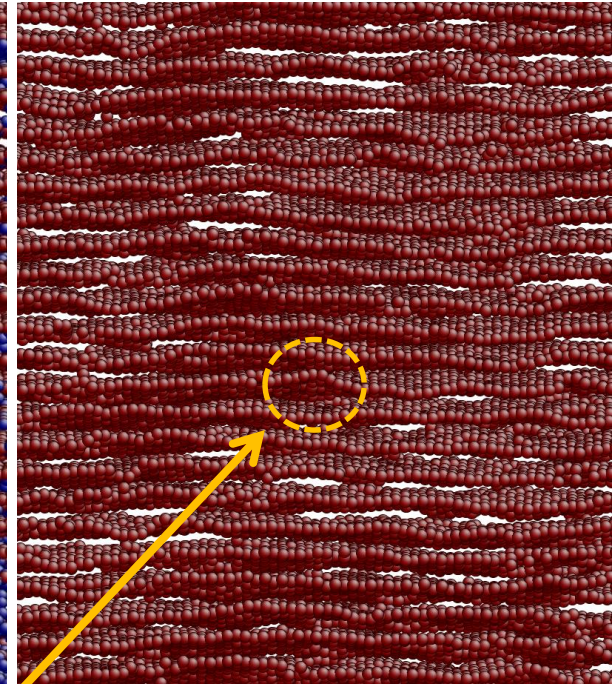
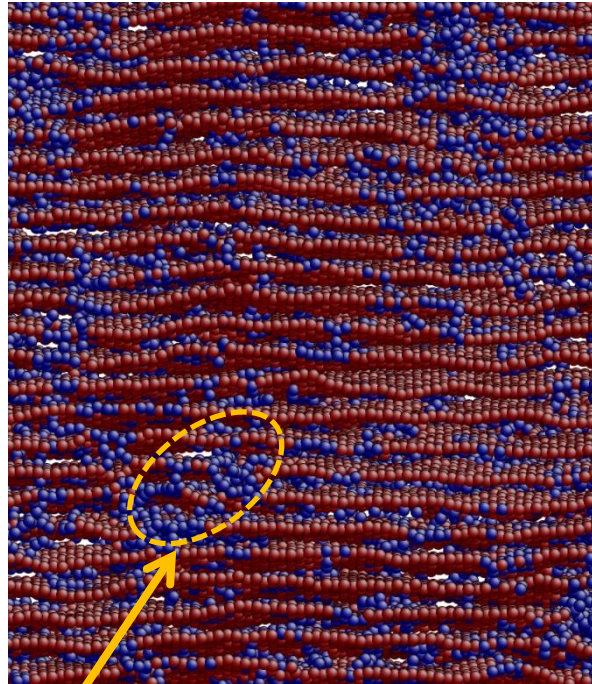
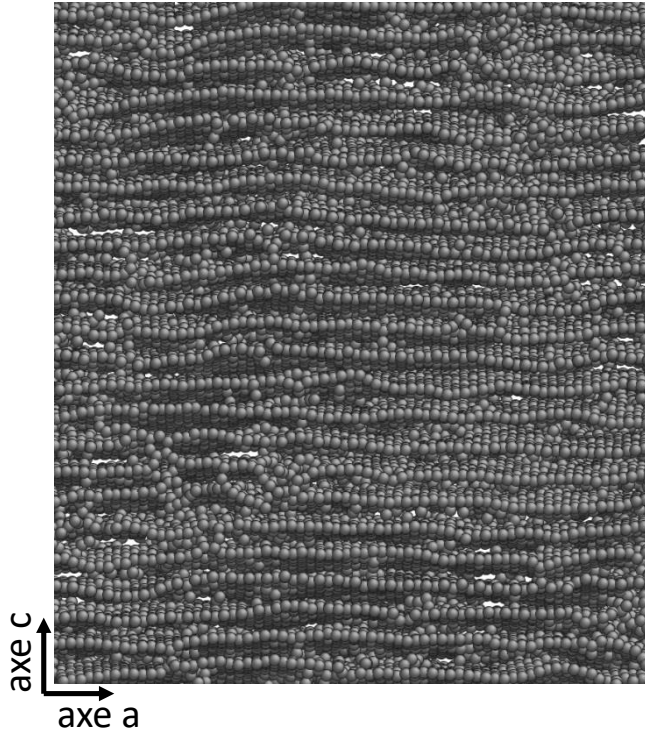
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Direction de l'énergie Nucléaire  
Département de Physico-Chimie  
Service de Corrosion et du  
Comportement des Matériaux  
dans leur Environnement

0.01 dpa

Local curvature threshold

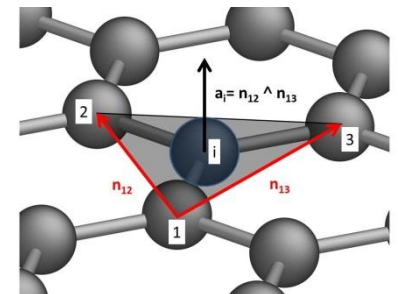
Remaining graphene sheets



Plans pinned by **disordered amorphous** zones

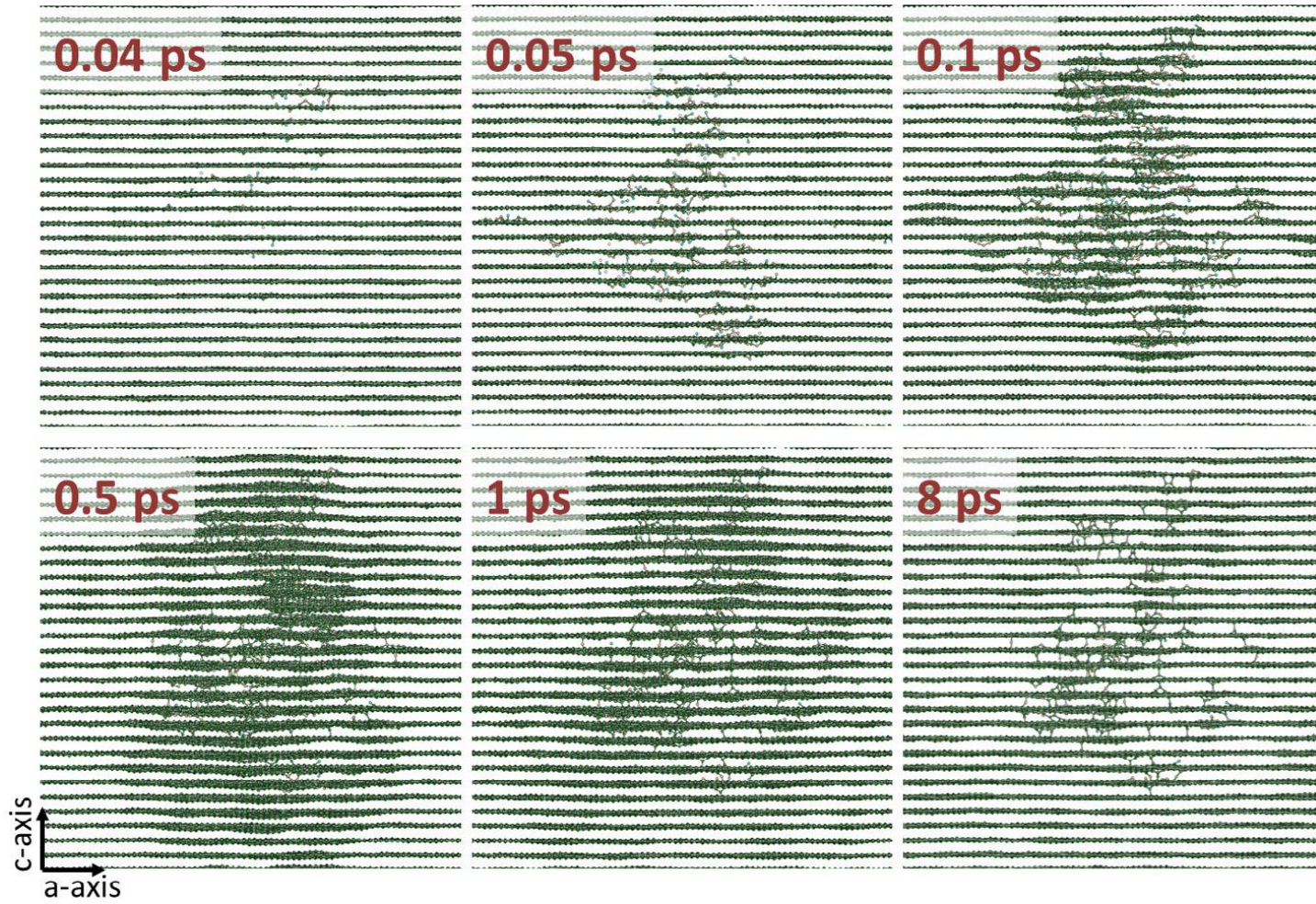
**Graphene sheets** are discriminated by **local curvature**

- average curvature radius
- size of graphene clusters

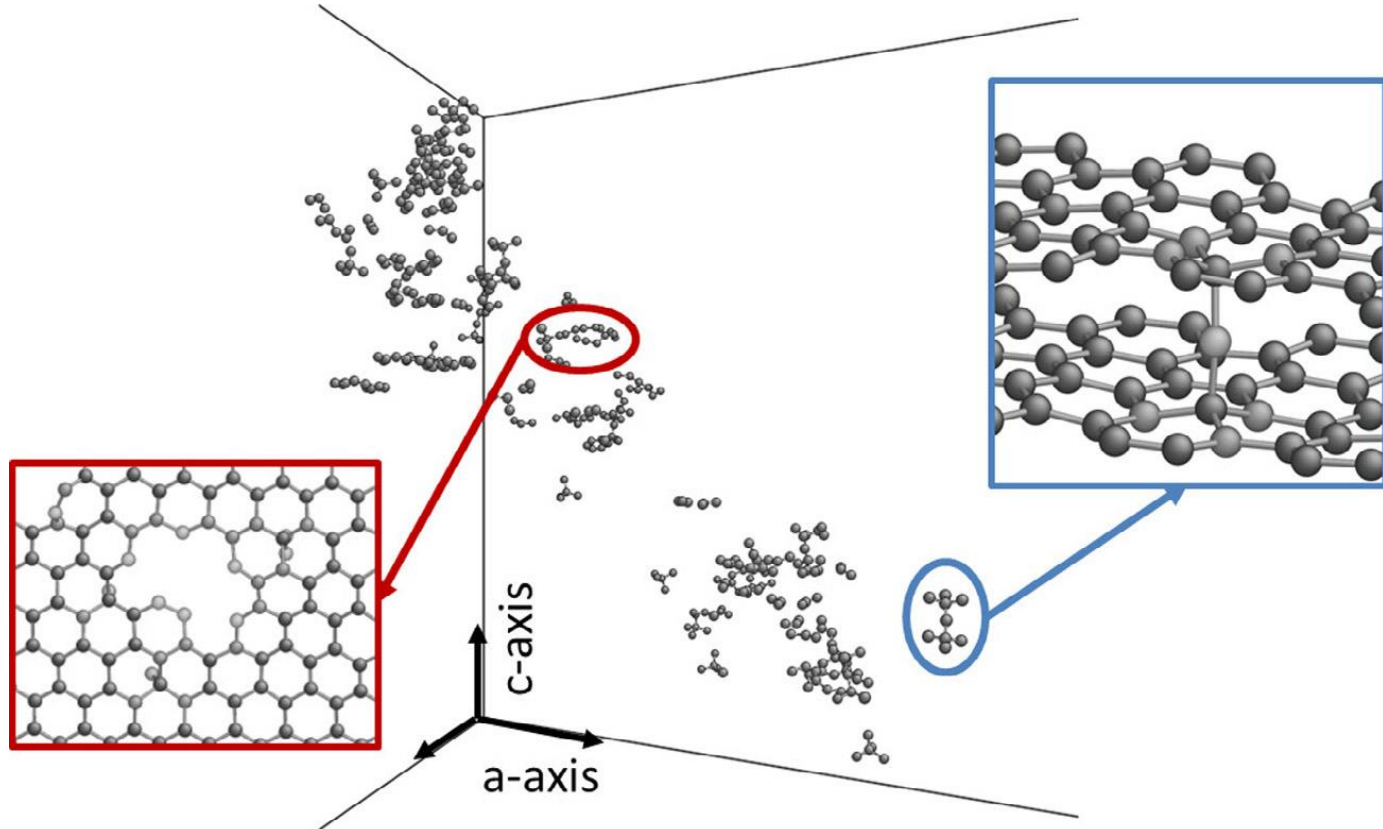




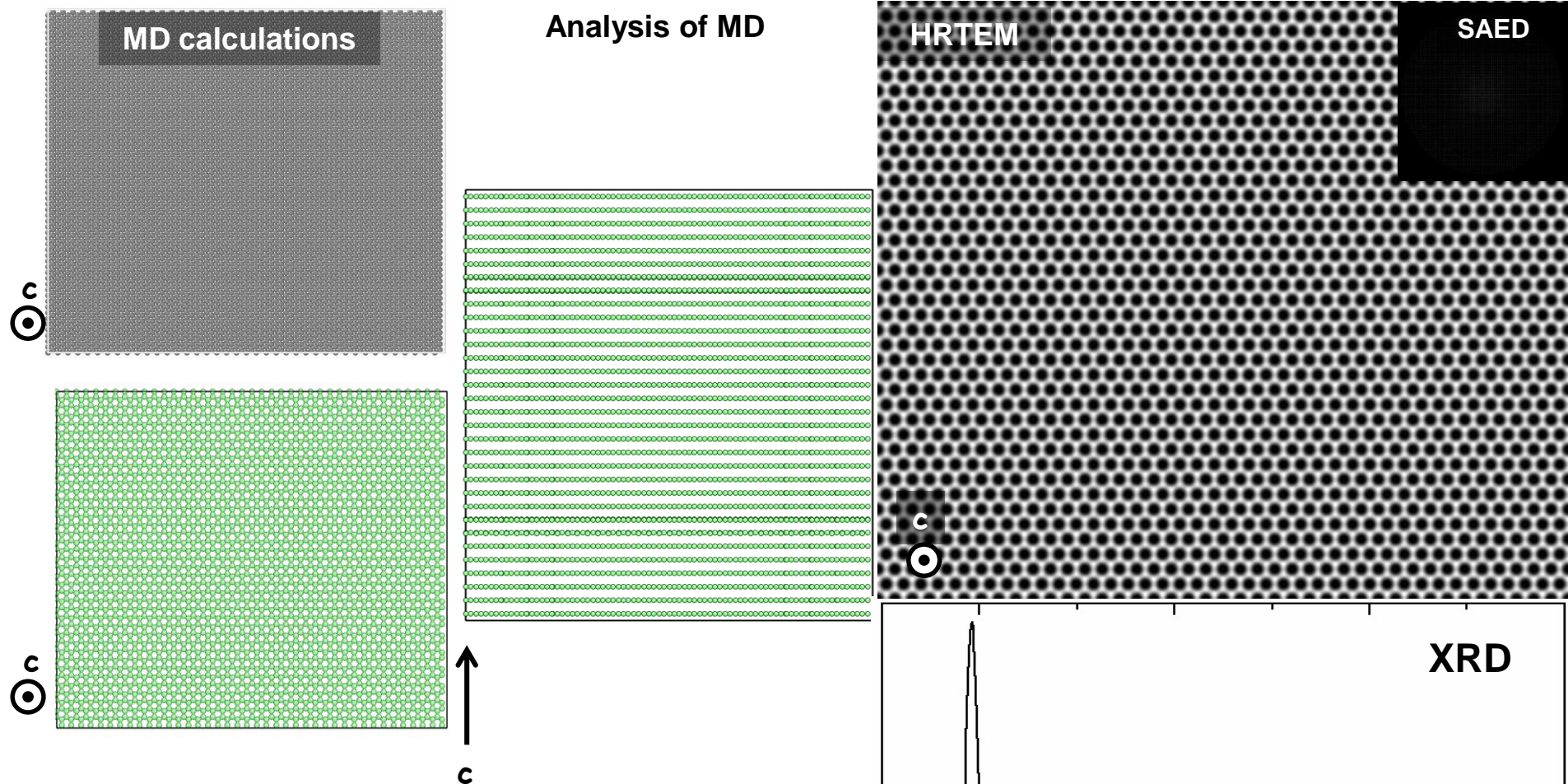




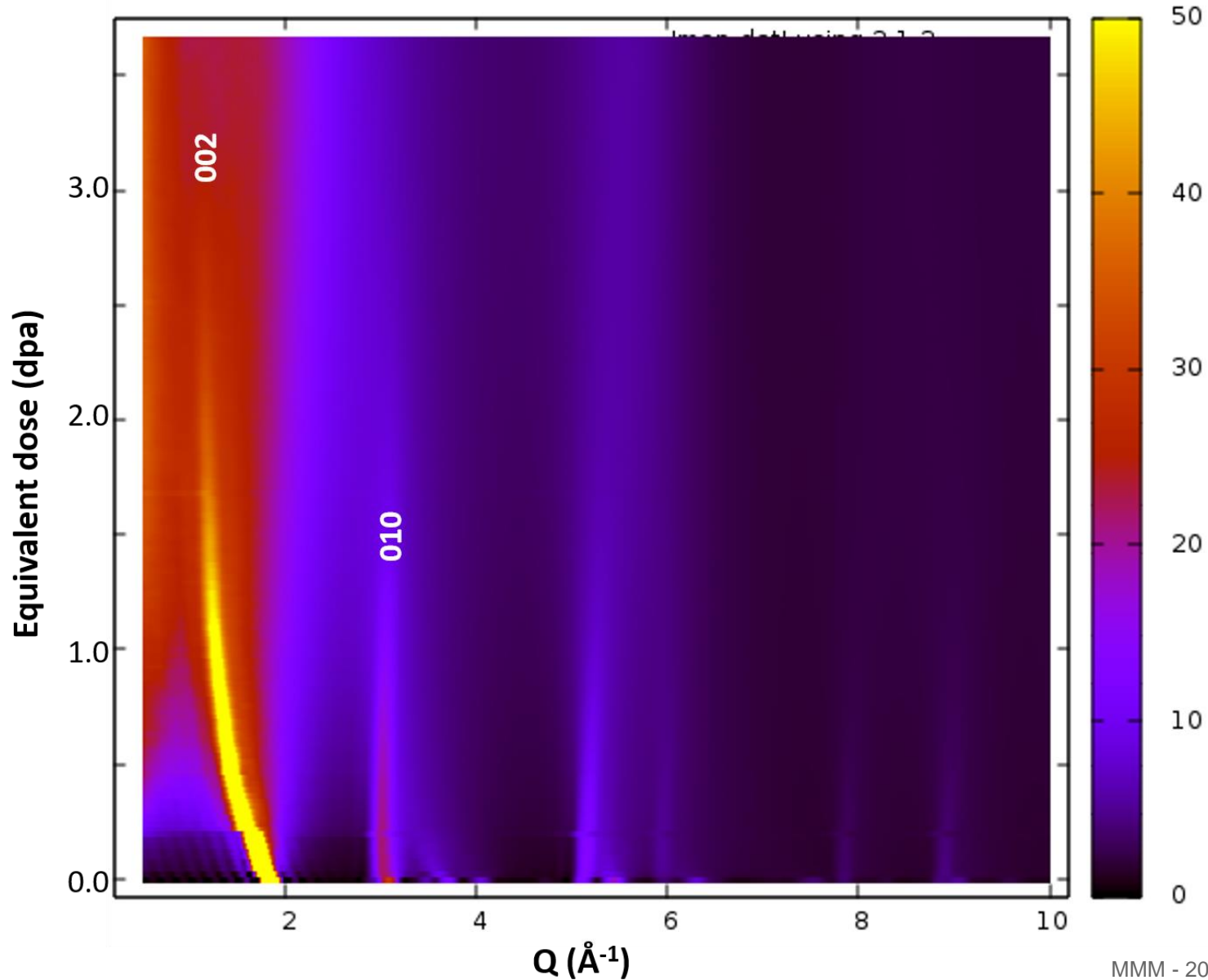
# Graphite – displacement cascade



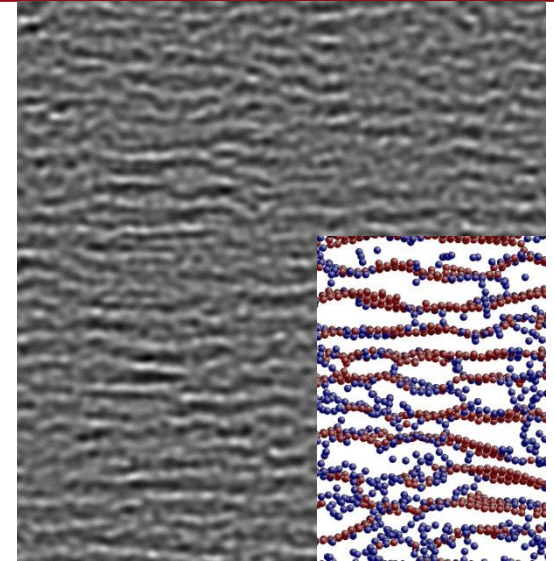
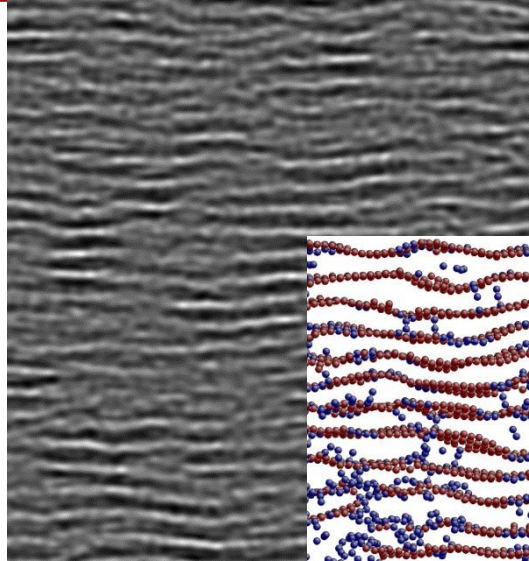
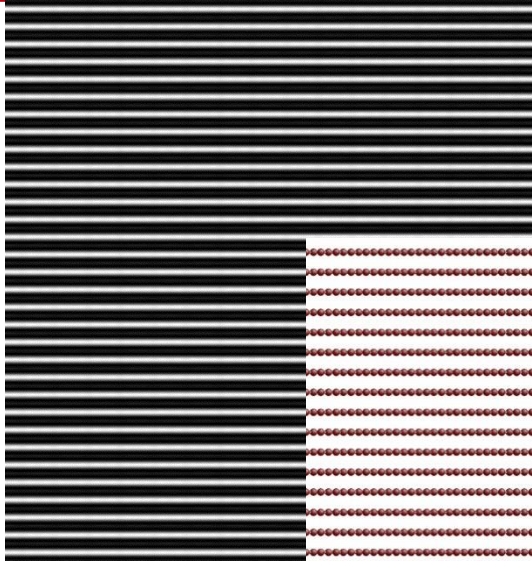
## Graphite - Analysis with DXA, HRTEM and XRD



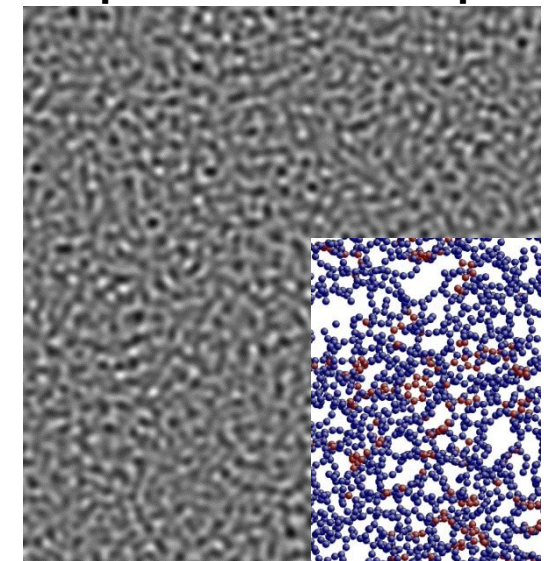
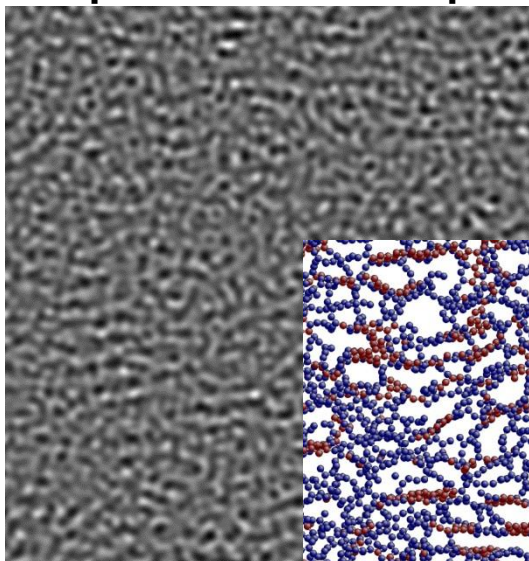
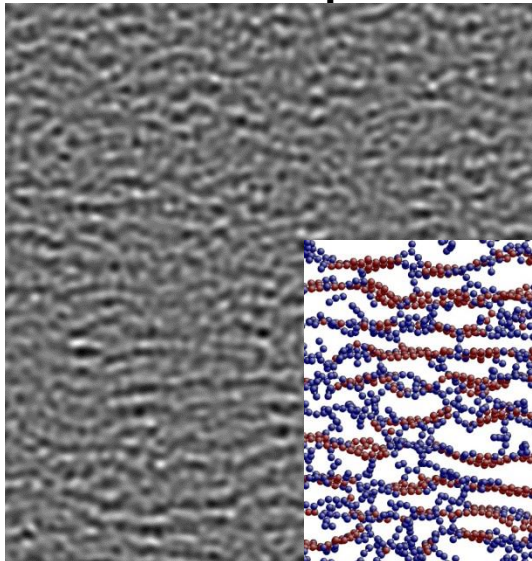
- Visualization of defects in graphene planes (MD and HRTEM)
- Visualization of wrinkles (MD analysis)
- Swelling from size of box (not easy from XRD)
- amorphization at 0.10 dpa (SAED and XRD)



5. nm



5. nm



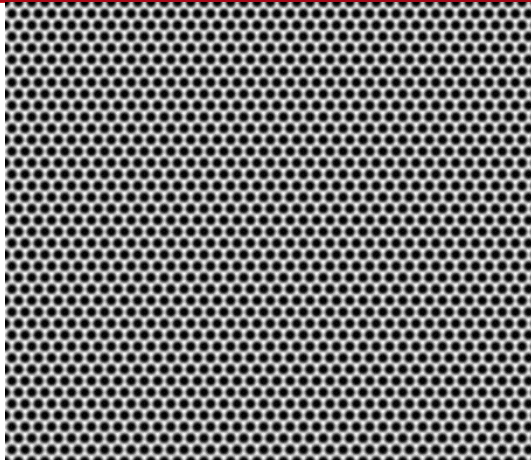
equivalent to 1.2 dpa

equivalent to 2.50 dpa

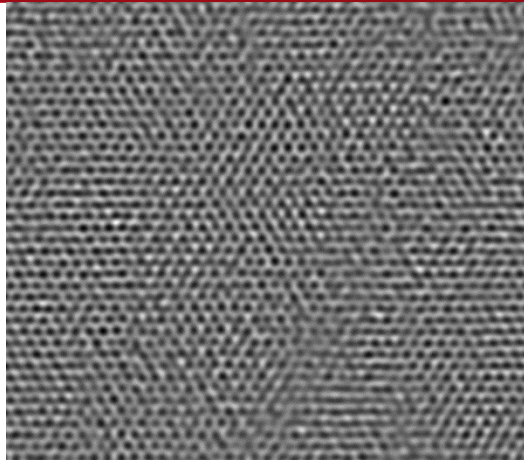
equivalent to 3.50 dpa

# Graphite – simulated HRTEM

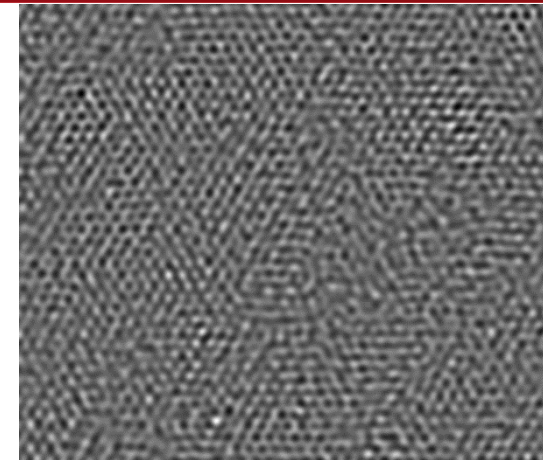
5. nm



0.00 dpa

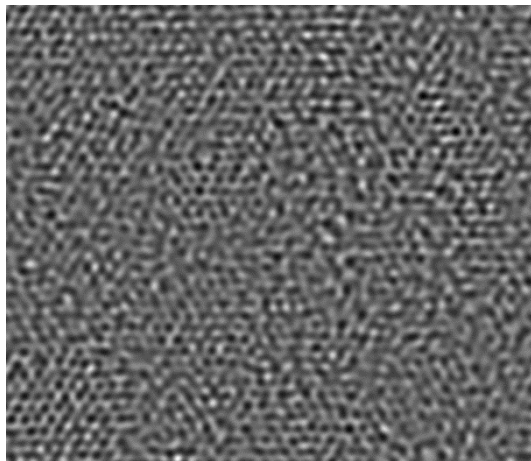


equivalent to 0.50 dpa

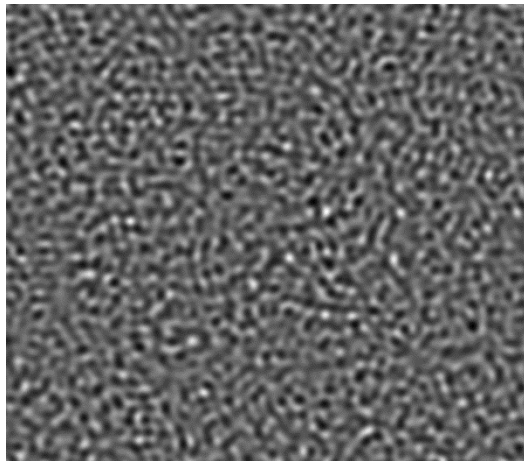


equivalent to 1.00 dpa

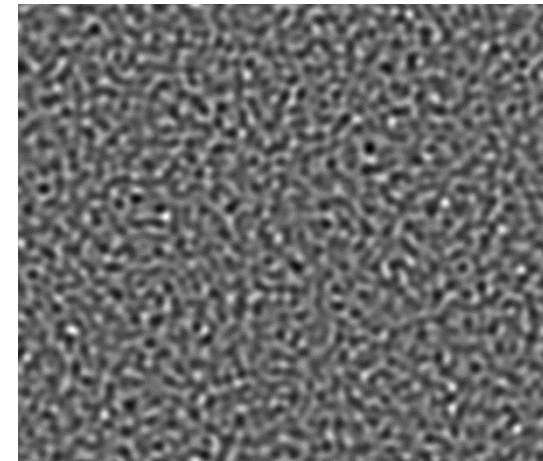
5. nm



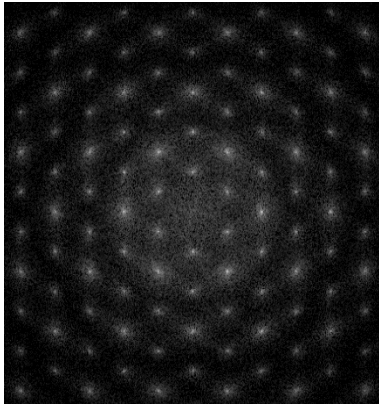
equivalent to 1.2 dpa



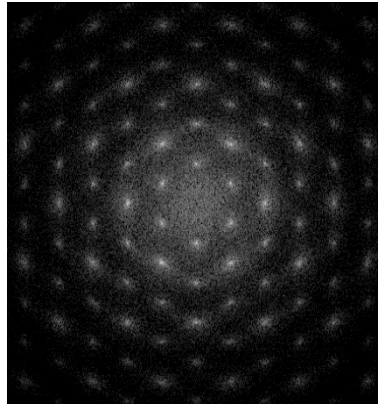
equivalent to 2.50 dpa



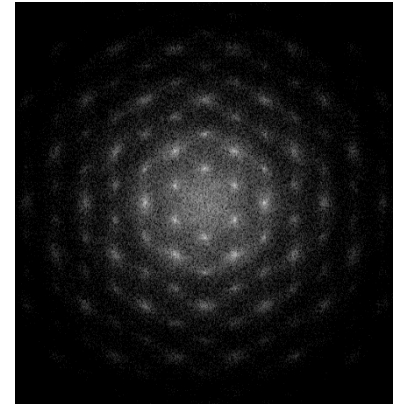
equivalent to 3.50 dpa



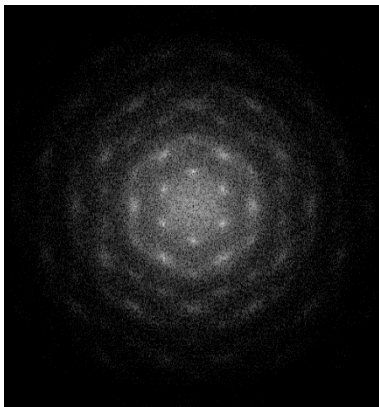
**0.00 dpa**



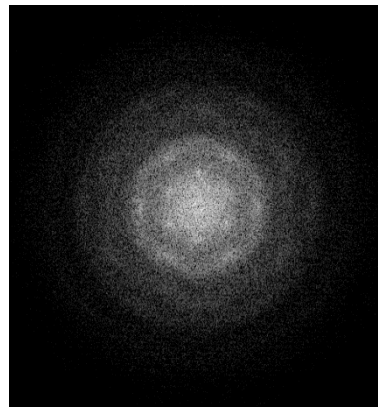
**equivalent to 0.50 dpa**



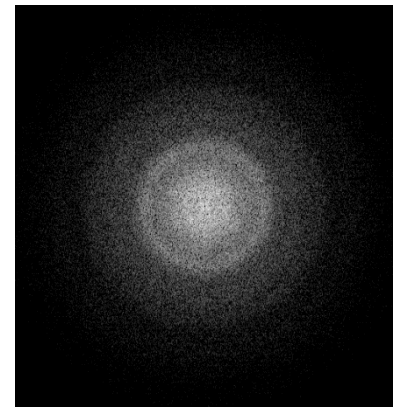
**equivalent to 1.00 dpa**



**equivalent to 1.2 dpa**



**equivalent to 2.50 dpa**



**equivalent to 3.50 dpa**