

Irradiation damage in nuclear graphite at the atomic scale

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

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DE LA RECHERCHE À L'INDUSTRIE

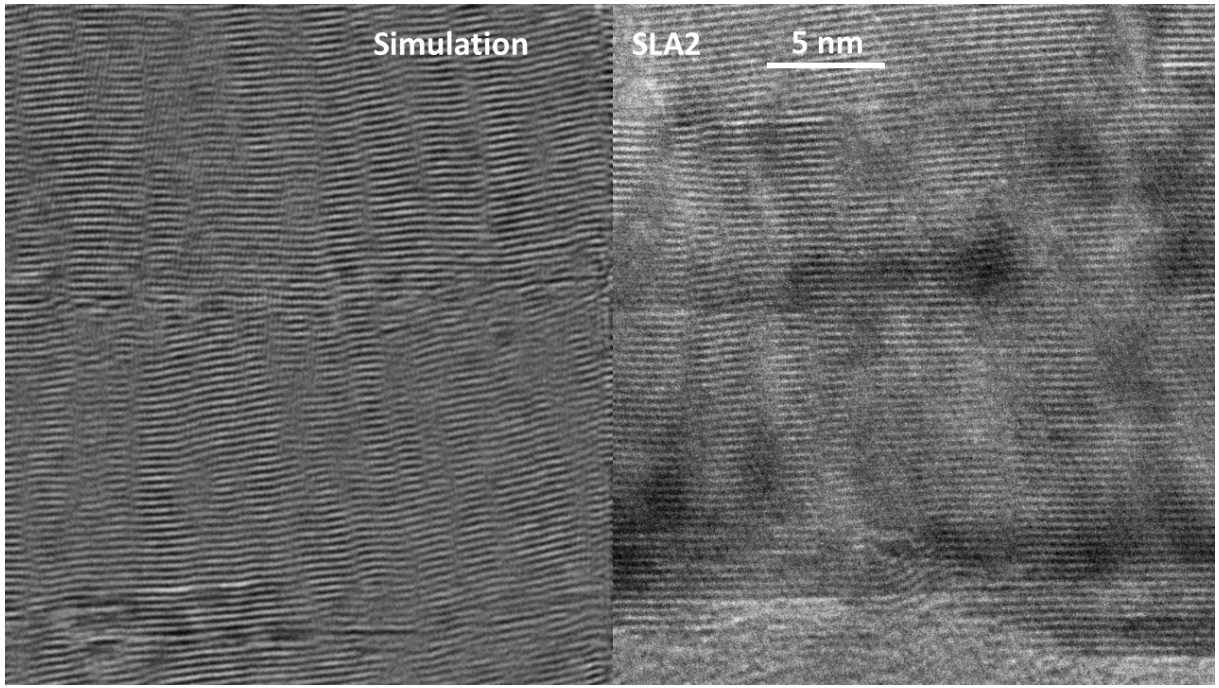
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2018 Multiscale Materials Modeling

Irradiation damage in nuclear graphite at the atomic scale

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

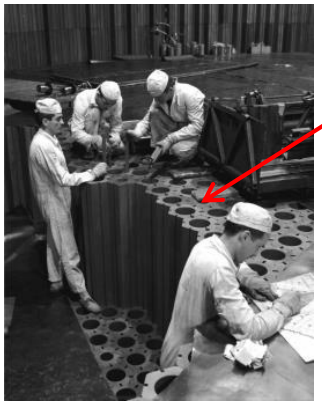


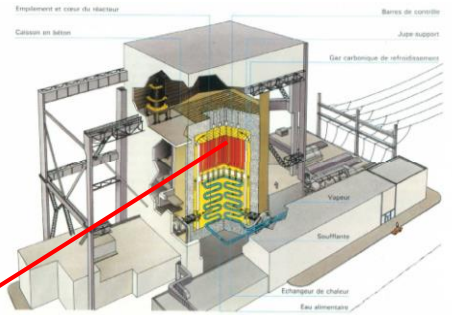
Irradiated graphite in graphite-gas nuclear reactors

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:

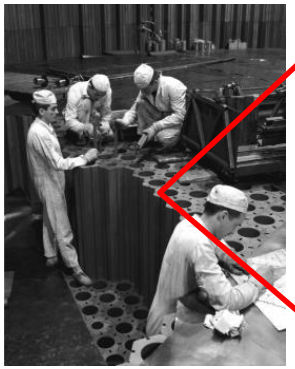
- Decommissioning under water
- Treatment to eliminate ^{14}C & ^{36}Cl, (^{3}H)

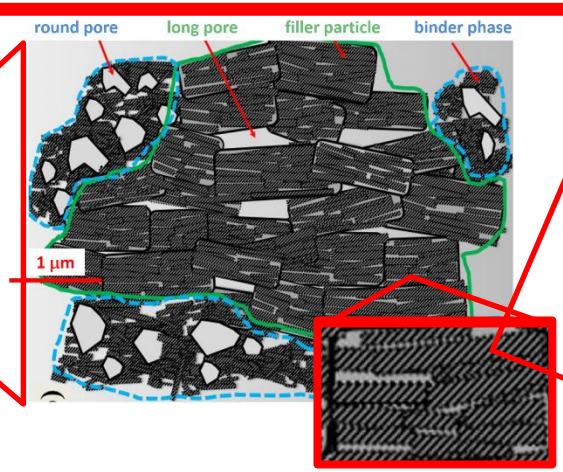
} interaction: graphite ↔ H_2O, O_2, CO_2…
depends on the localization of ^{14}C & ^{36}Cl after irradiation

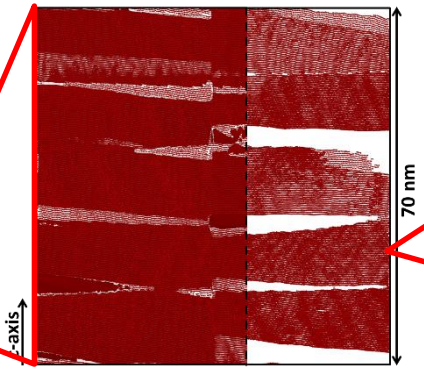
- **Graphite microstructure evolution under irradiation?**
  - - radiation damages and swelling
  - - link between XRD – HRTEM images and atomic scale structure

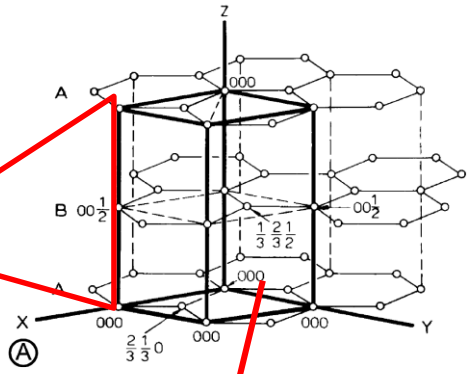
Microstructure / swelling by irradiation

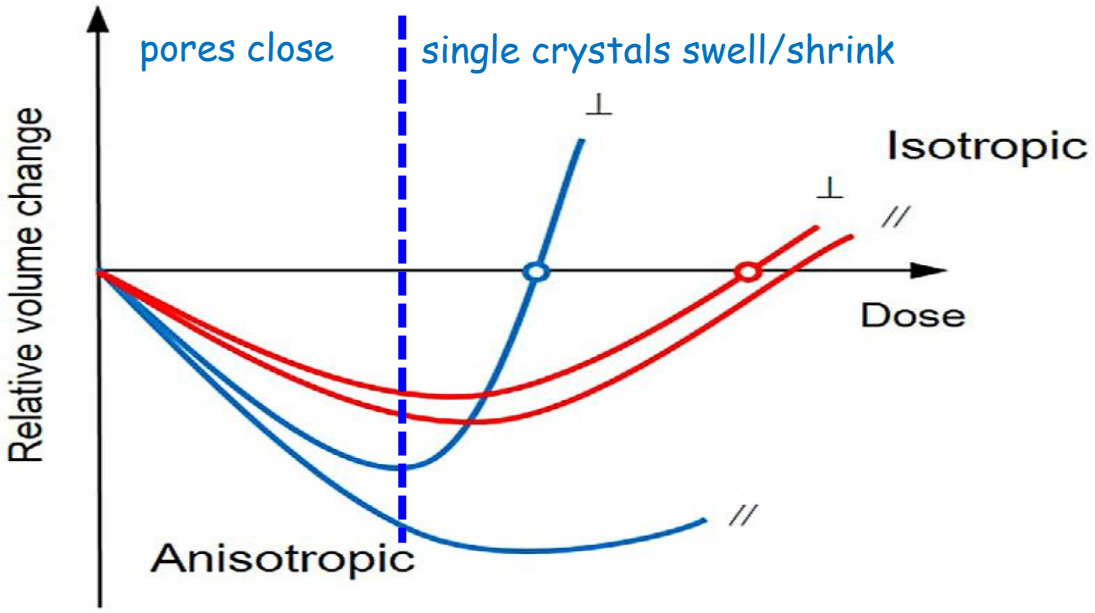
cm → nm



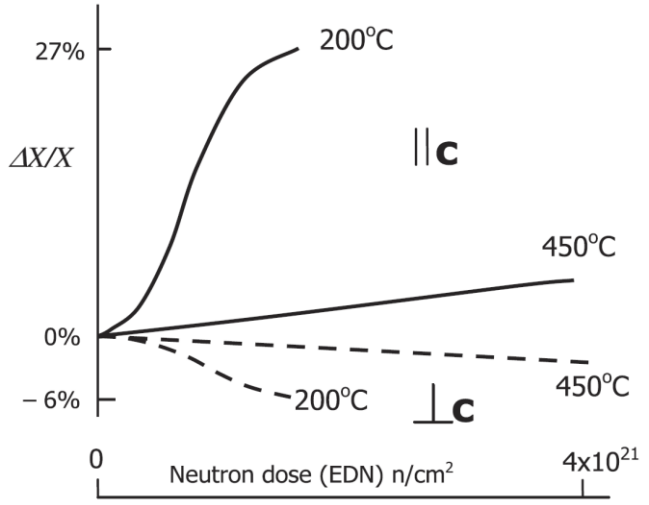








Shen et al (2015)

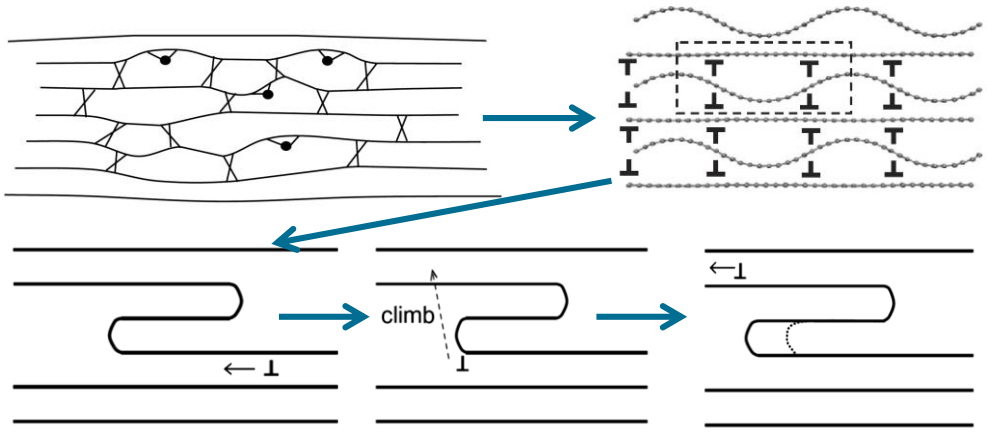


Heggie et al (2011)

Scenario of swelling under irradiation

Scenario of swelling

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



Heggie & Telling (2007)

Based on:

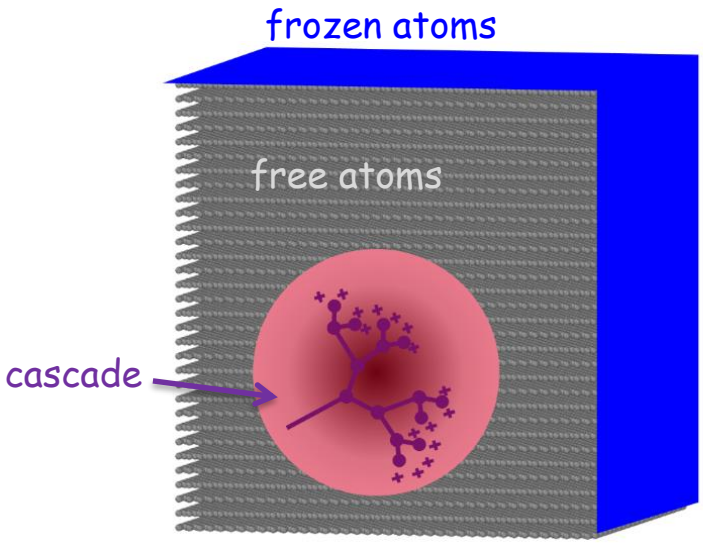
- HRTEM observations
- Atomic scale (DFT – MD) calculations
- Wigner energy estimations

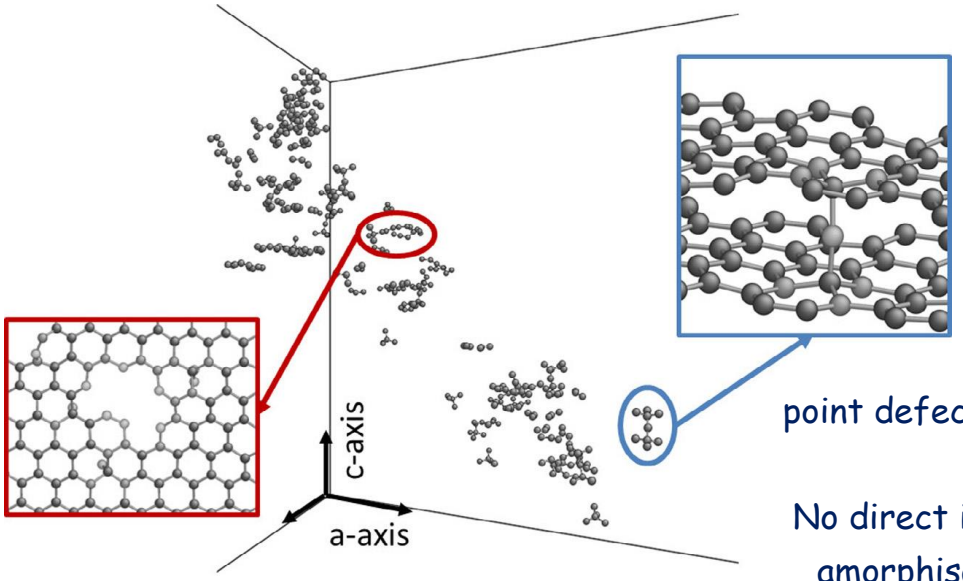
Our goals:

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

Modelling radiation damages

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





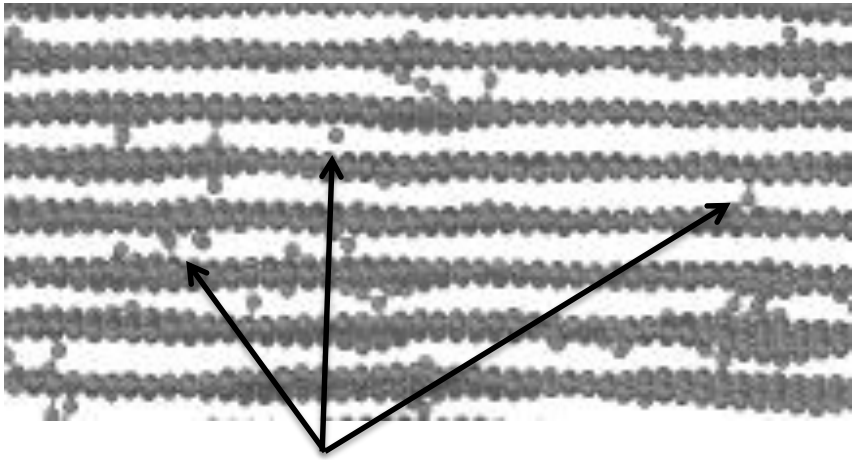
point defects only

No direct impact
amorphisation

if point defects then

Dose effect by Frenkel pairs accumulation

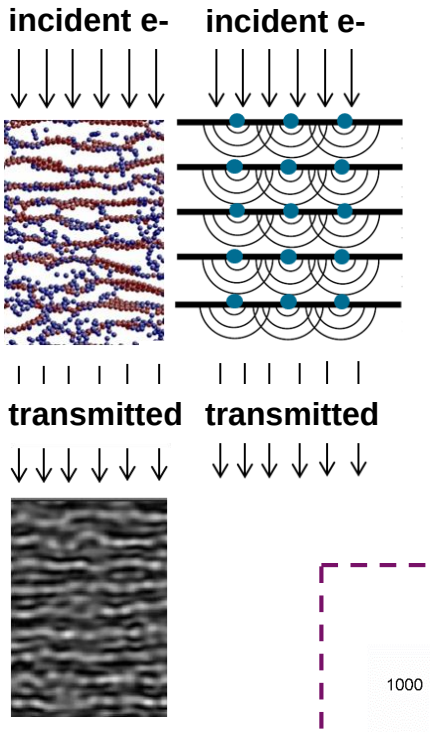
- Creation of Frenkel pairs every 2 ps
  high dose rate
- Dose up to 10 dpa
- constant pressure and constant temperature



Frenkel pairs

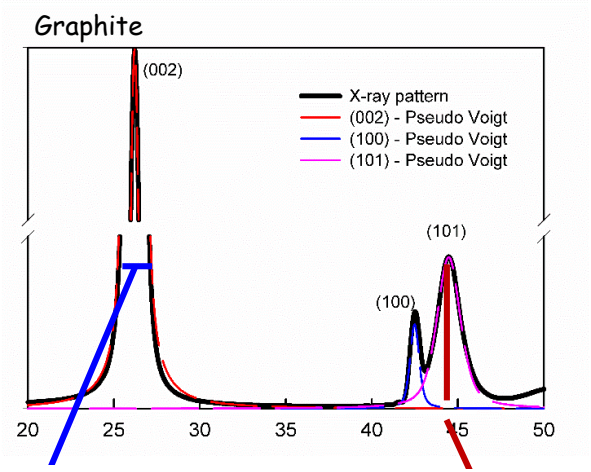
Analysis of MD snapshots

HRTEM simulation
(NCEMSS or Multi-slice code)



Graphite

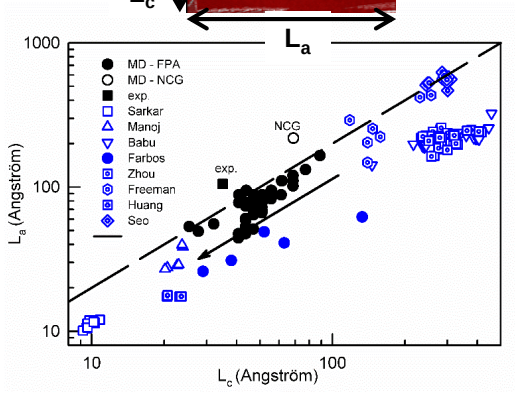
XRD simulation
(code Debyer)

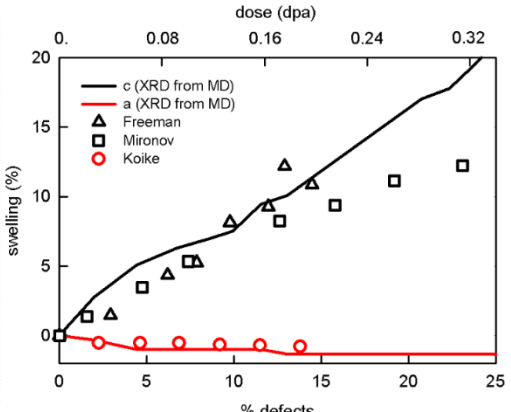


FWHM → Scherrer's formula

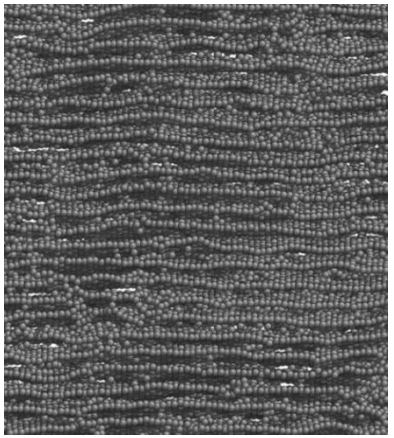
Peak position → Bragg's law



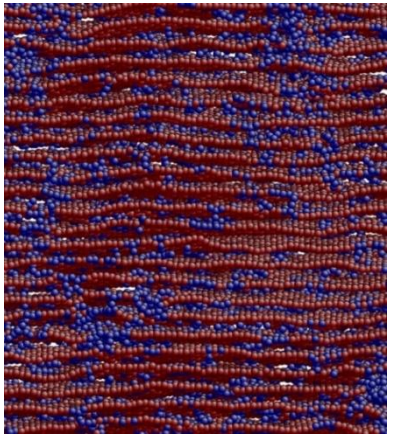




Topological analysis:
local curvature criterion
(graphite if curvature > 32°)



defects in blue

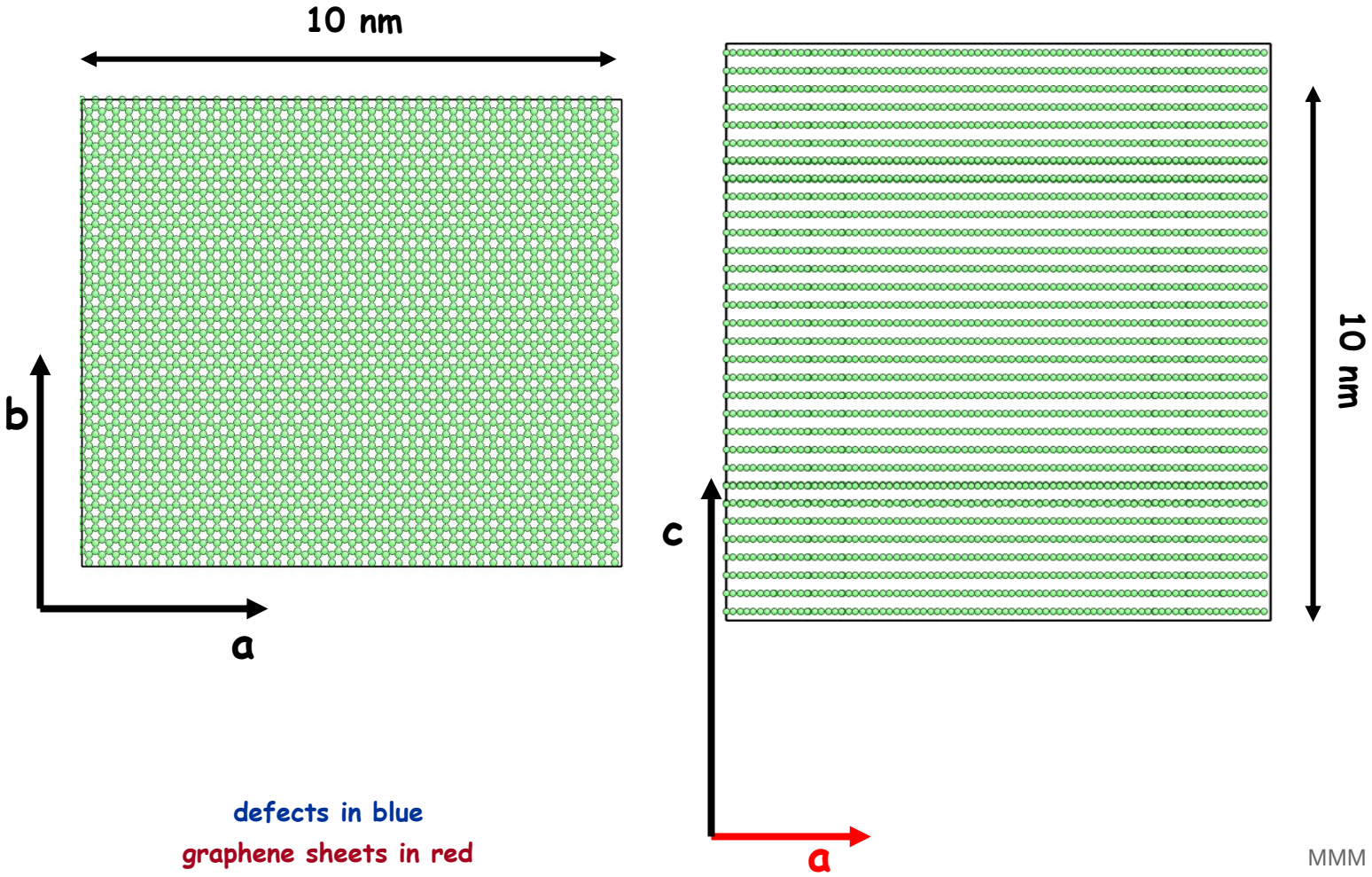


graphene sheets in red

Frenkel pairs accumulation

Shrinking in the basal plane

Swelling along c-axis

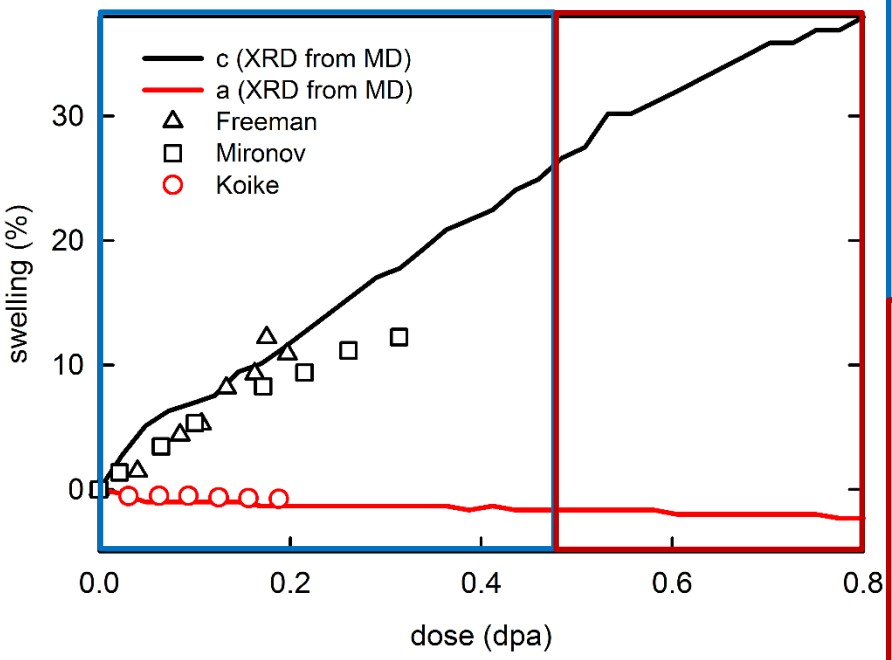


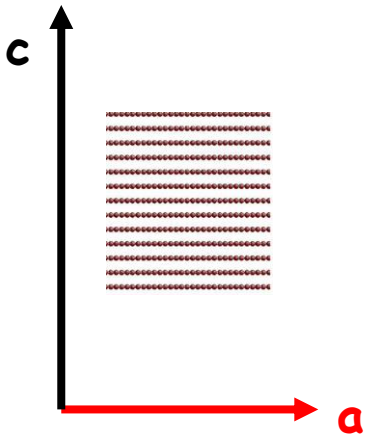
defects in blue

graphene sheets in red

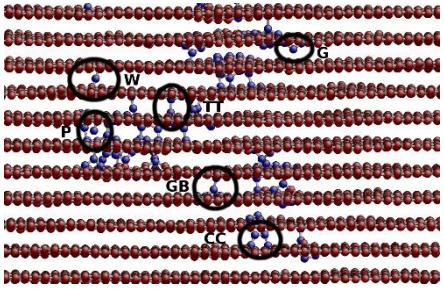
Link between point defects and swelling

a and c from XRD - Bragg's law

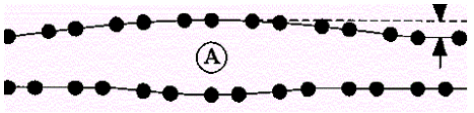




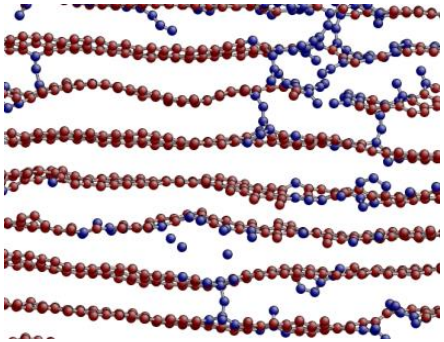
1. Point Defects



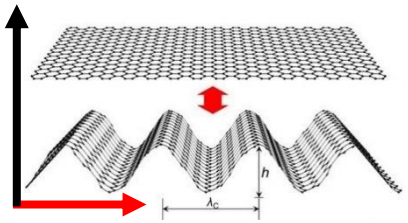
interstitial



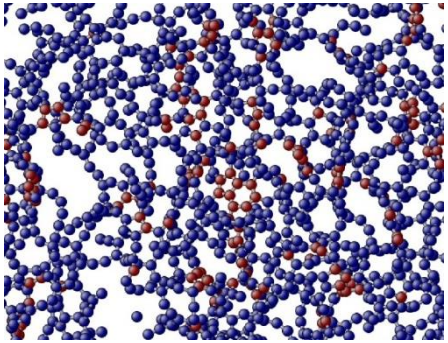
2. Pinning on small disordered cluster and Wrinkling of graphene sheets



swelling / shrinking

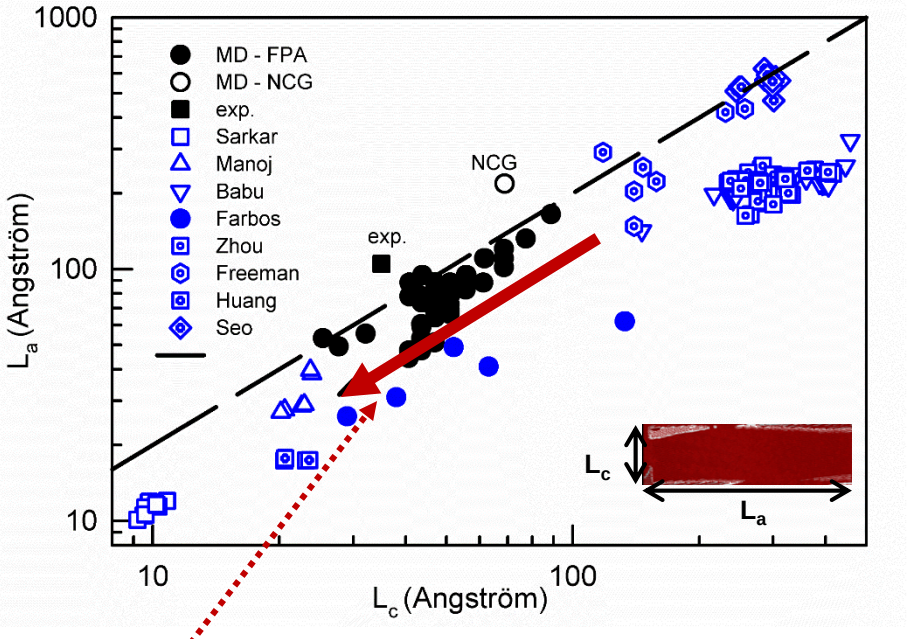


3. Growth of amorphous pockets



Microstructure and HRTEM images

La and Lc – Scherrer's formula



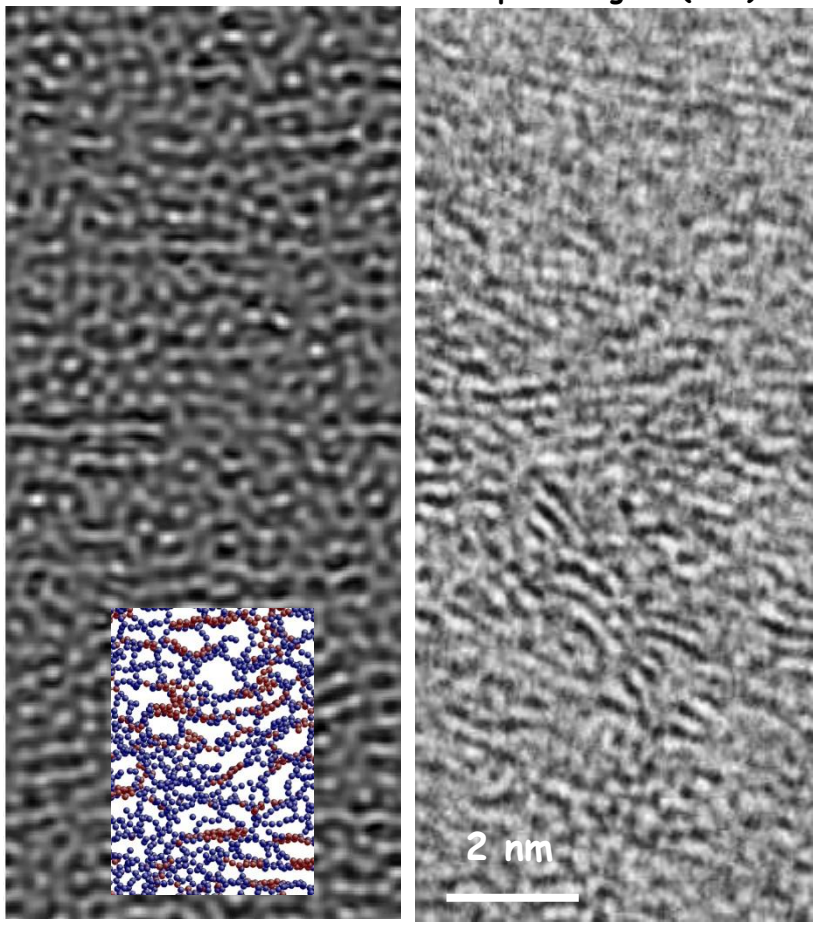
Decrease of graphene size with increasing dose

→ small graphene sheets in "noodles shape"

= BSU (Basic Structural Units)

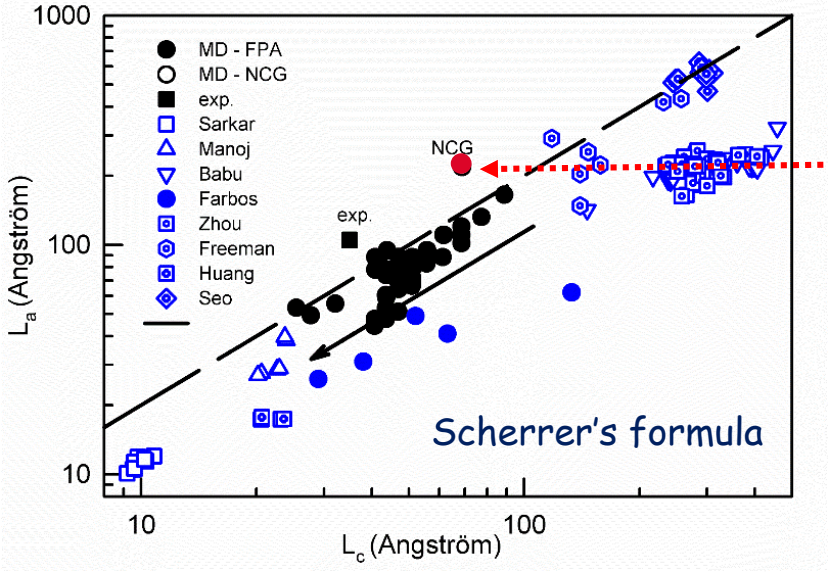
What simulations show:

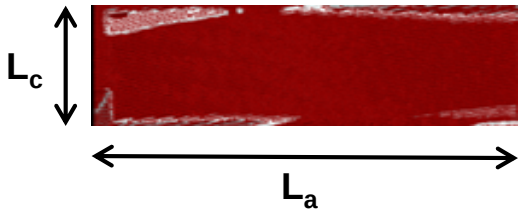
graphite amorphizes

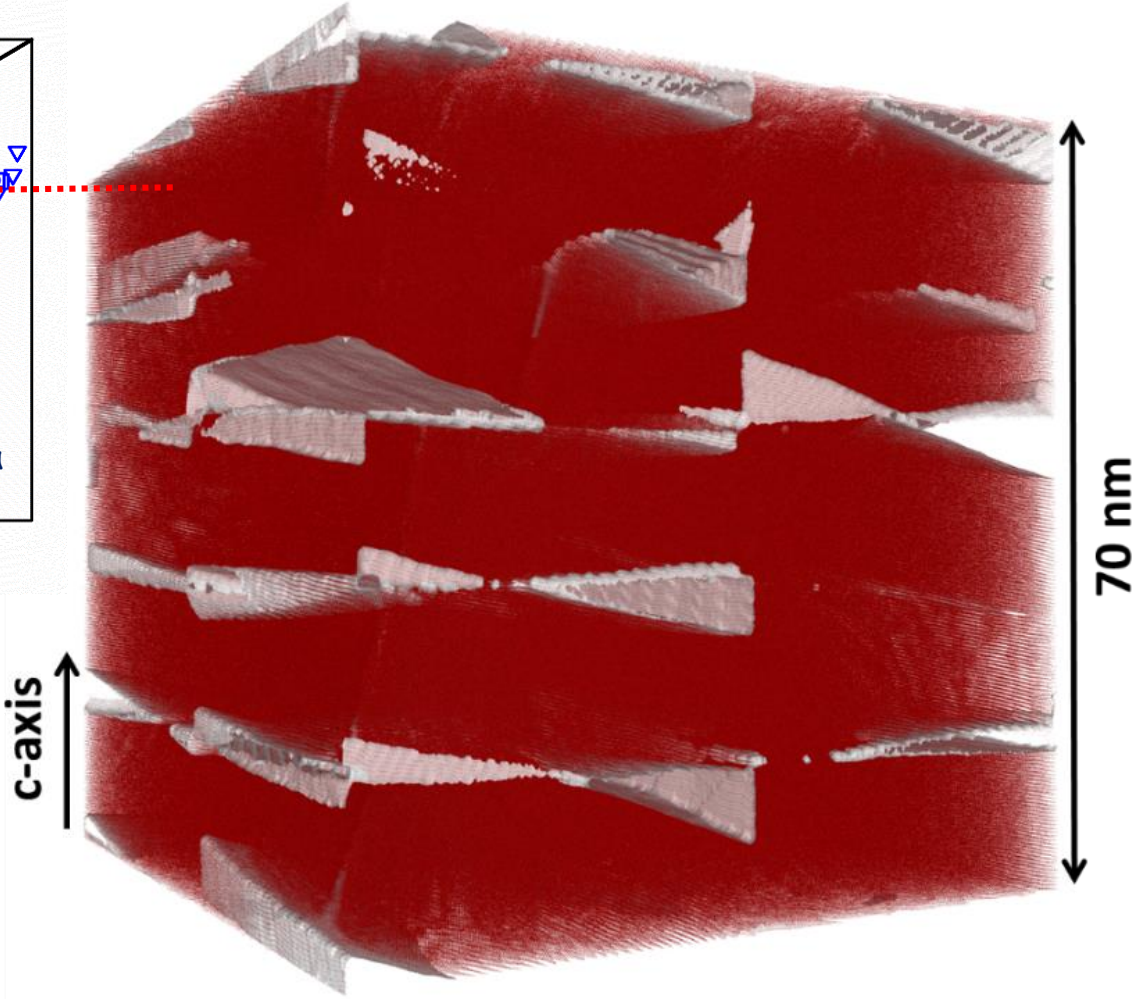


Nano-crystalline graphite

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





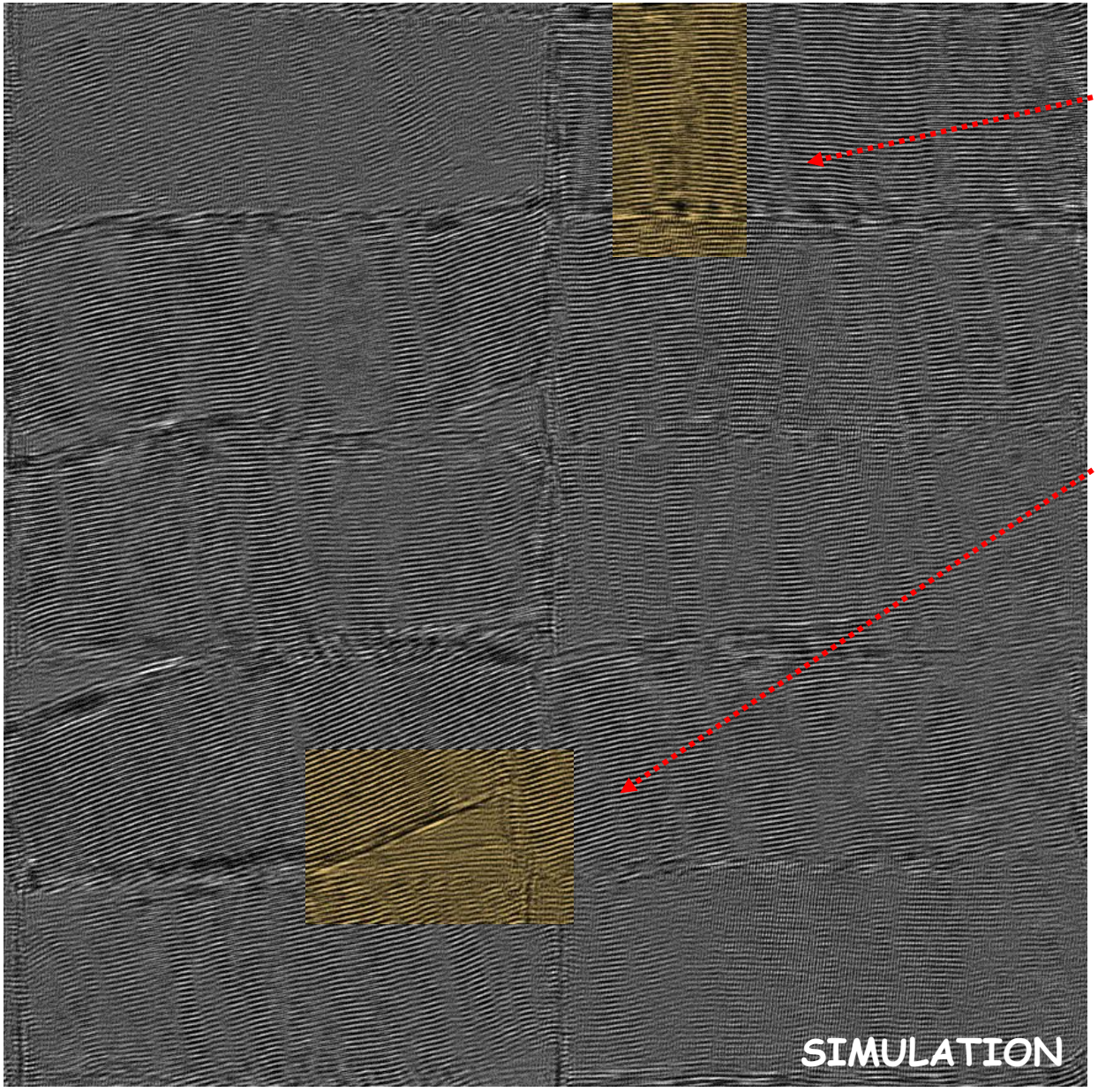


Scherrer → Lc = 7 nm, La = 22 nm

initial → Lc = 15 nm, La = 35 nm

Close to experiments

HRTEM images of nano-crystalline graphite



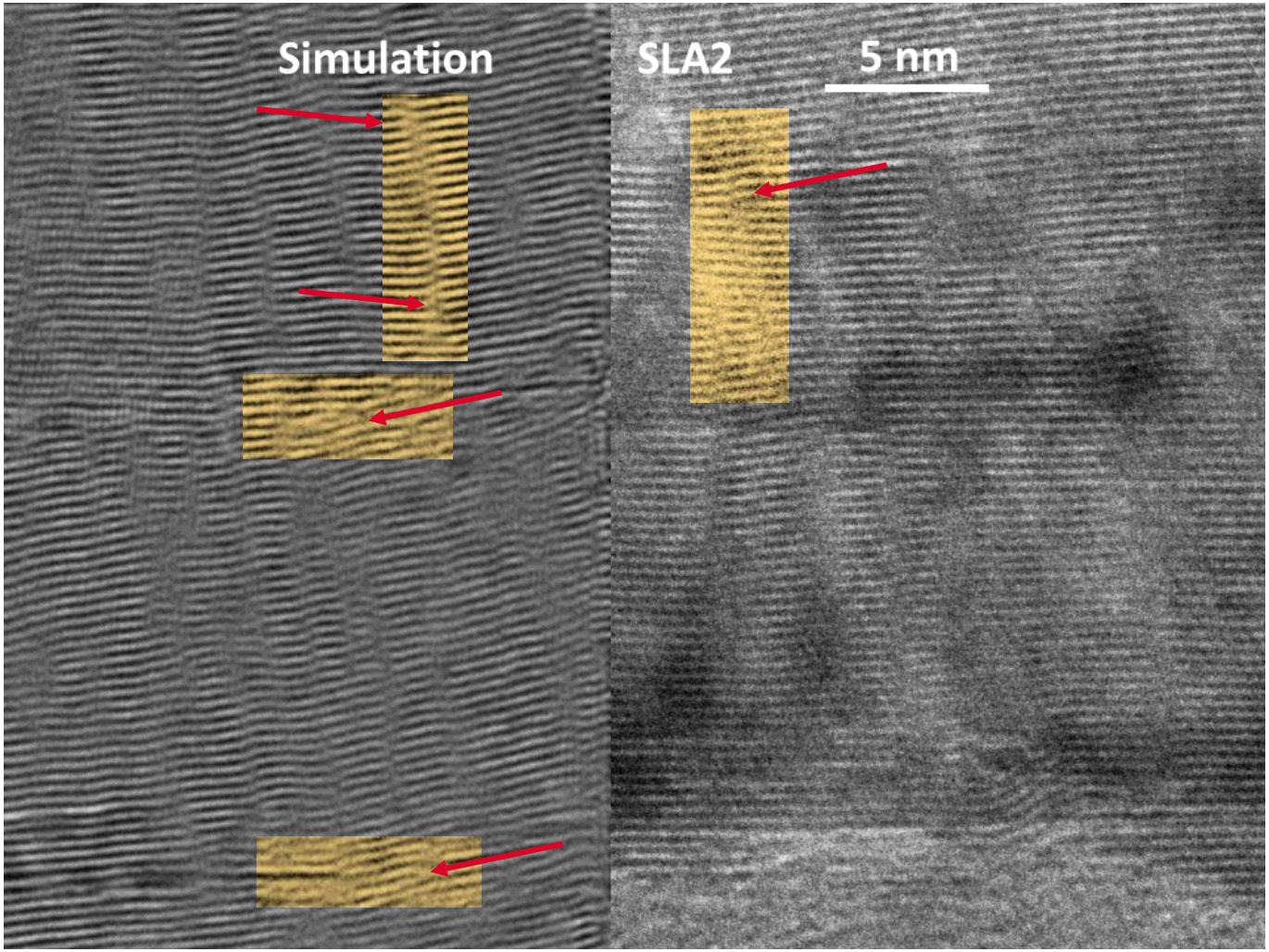
HRTEM image shows dislocation (edge or screw)

MD snapshot shows wrinkles

HRTEM image shows nano-pore

MD snapshot shows nano-pore

HRTEM images of nano-crystalline graphite



HRTEM images
→ dislocations

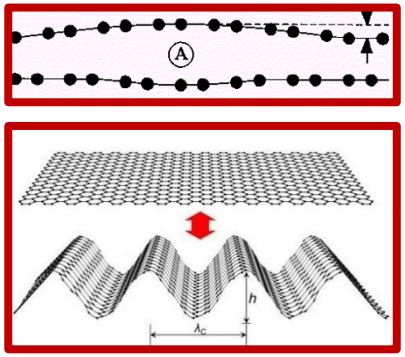
but

MD snapshots
→ wrinkles

Conclusion - summary

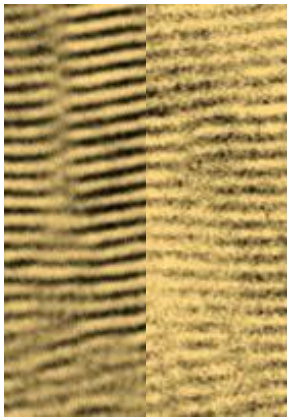
Irradiation damages in graphite single crystal: Frenkel Pair Accumulation method

- 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.

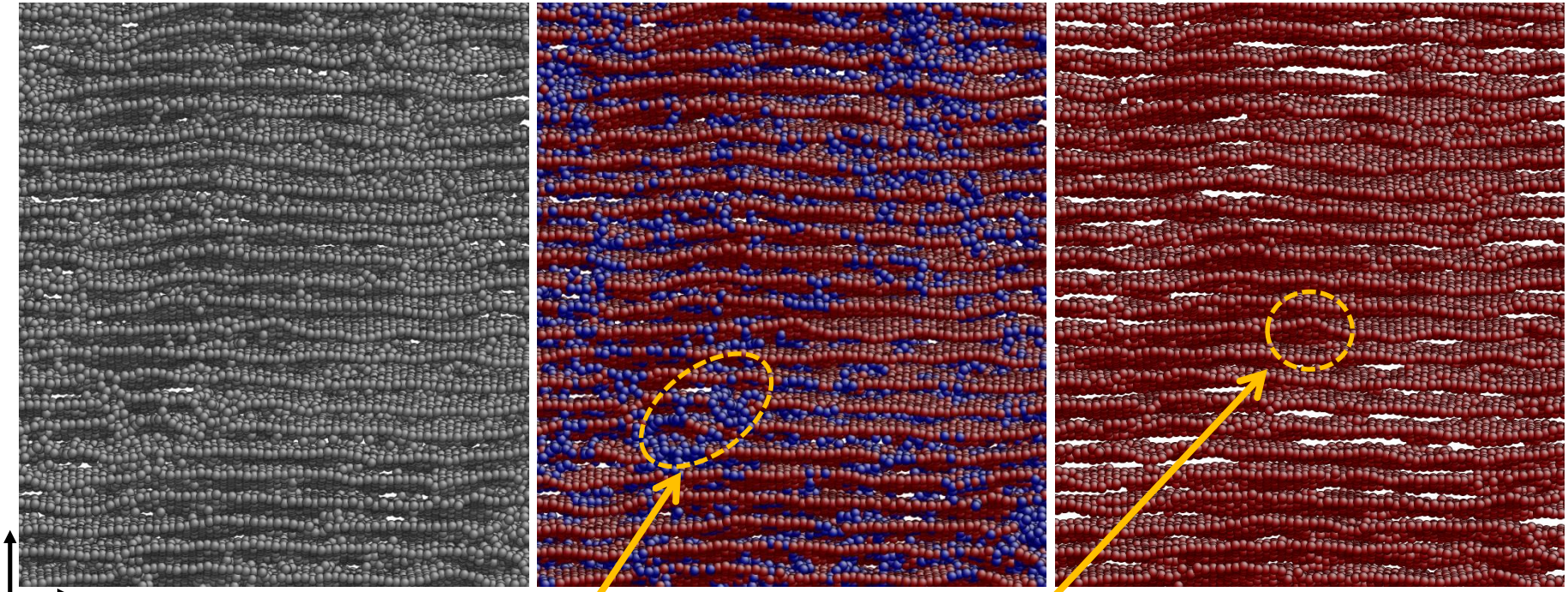
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Direction de l'énergie Nucléaire

Département de Physico-Chimie
Service de Corrosion et du Comportement des Matériaux dans leur Environnement

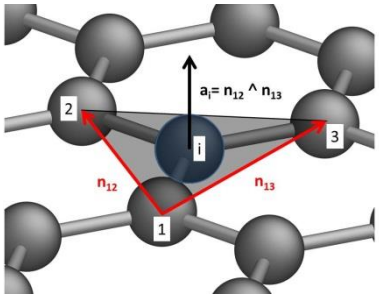
Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

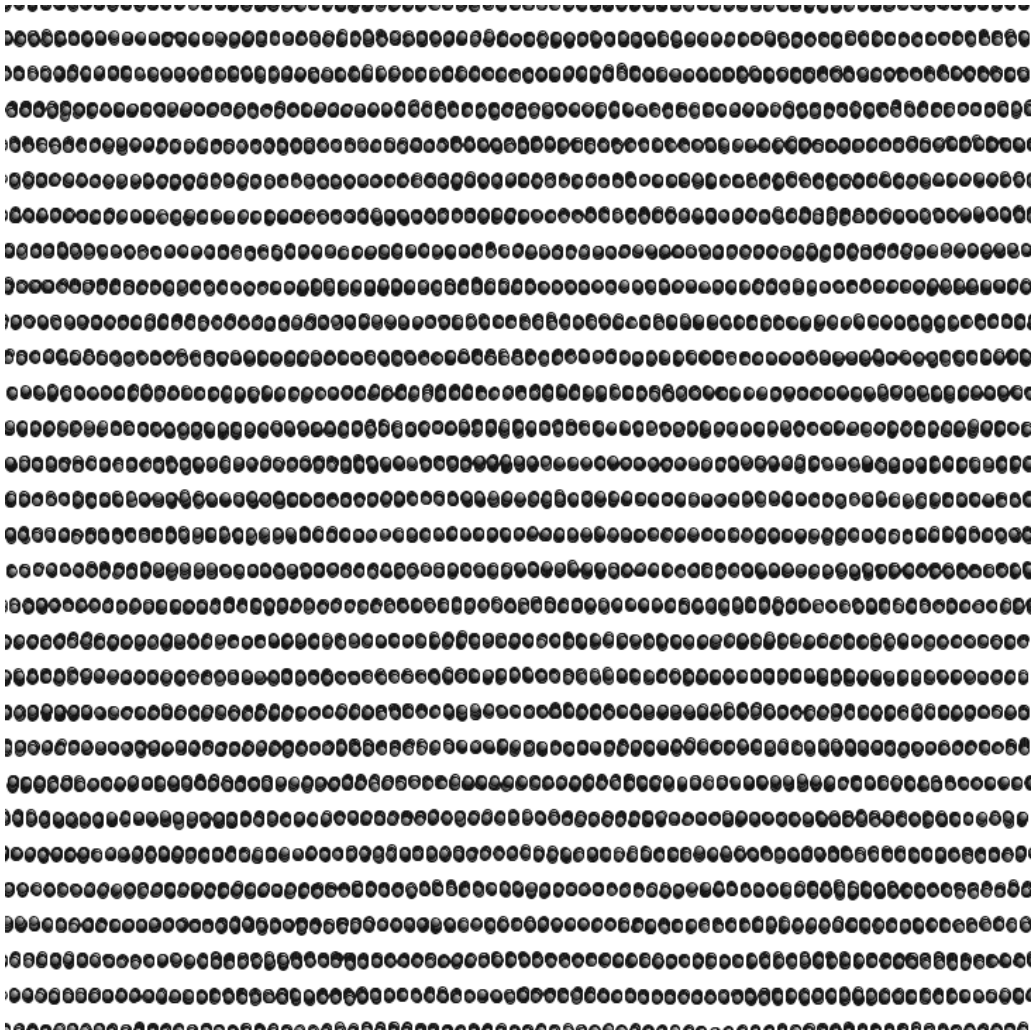
0.01 dpa | Local curvature threshold | Remaining graphene sheets

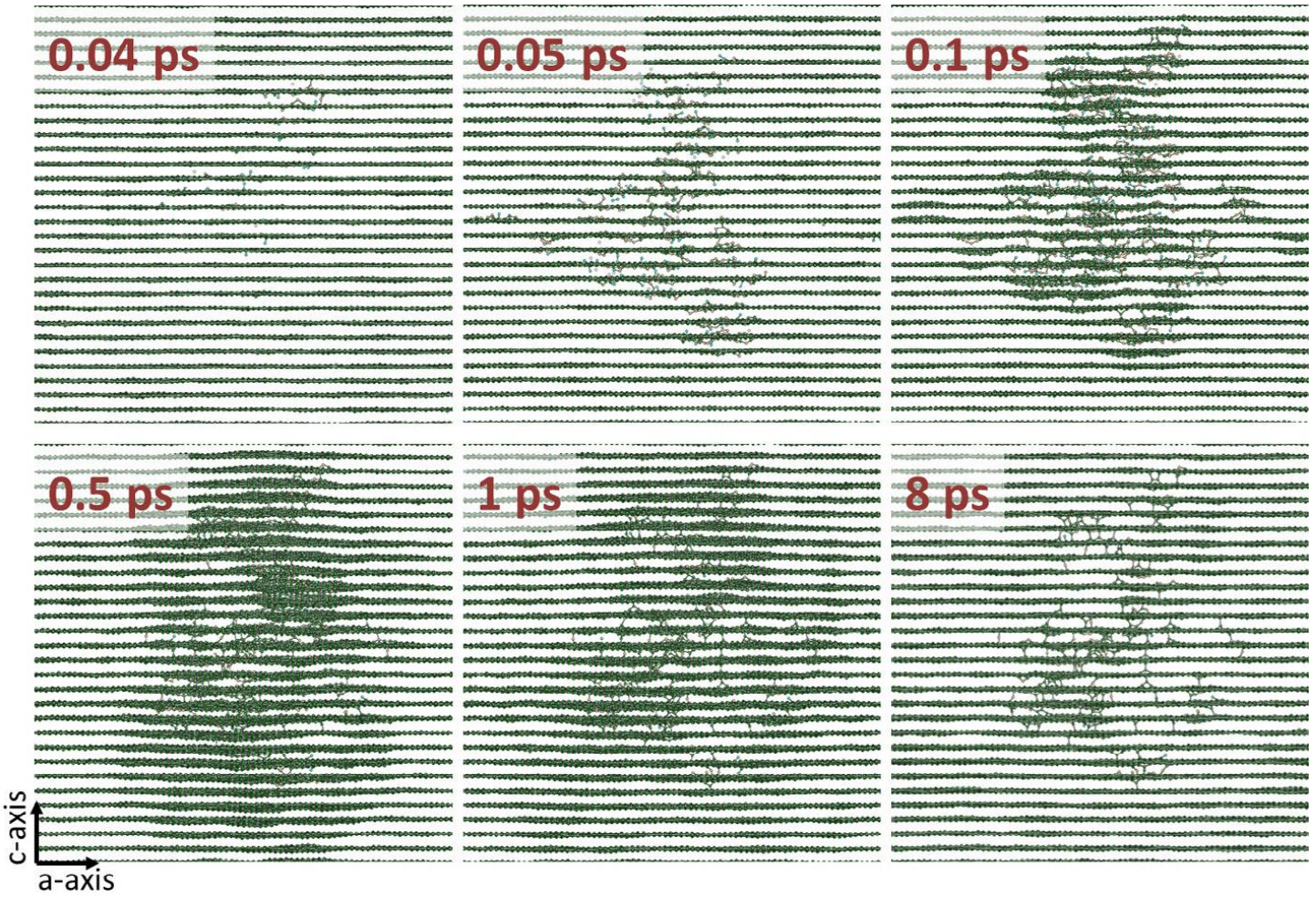


Plans pinned by disordered amorphous zones

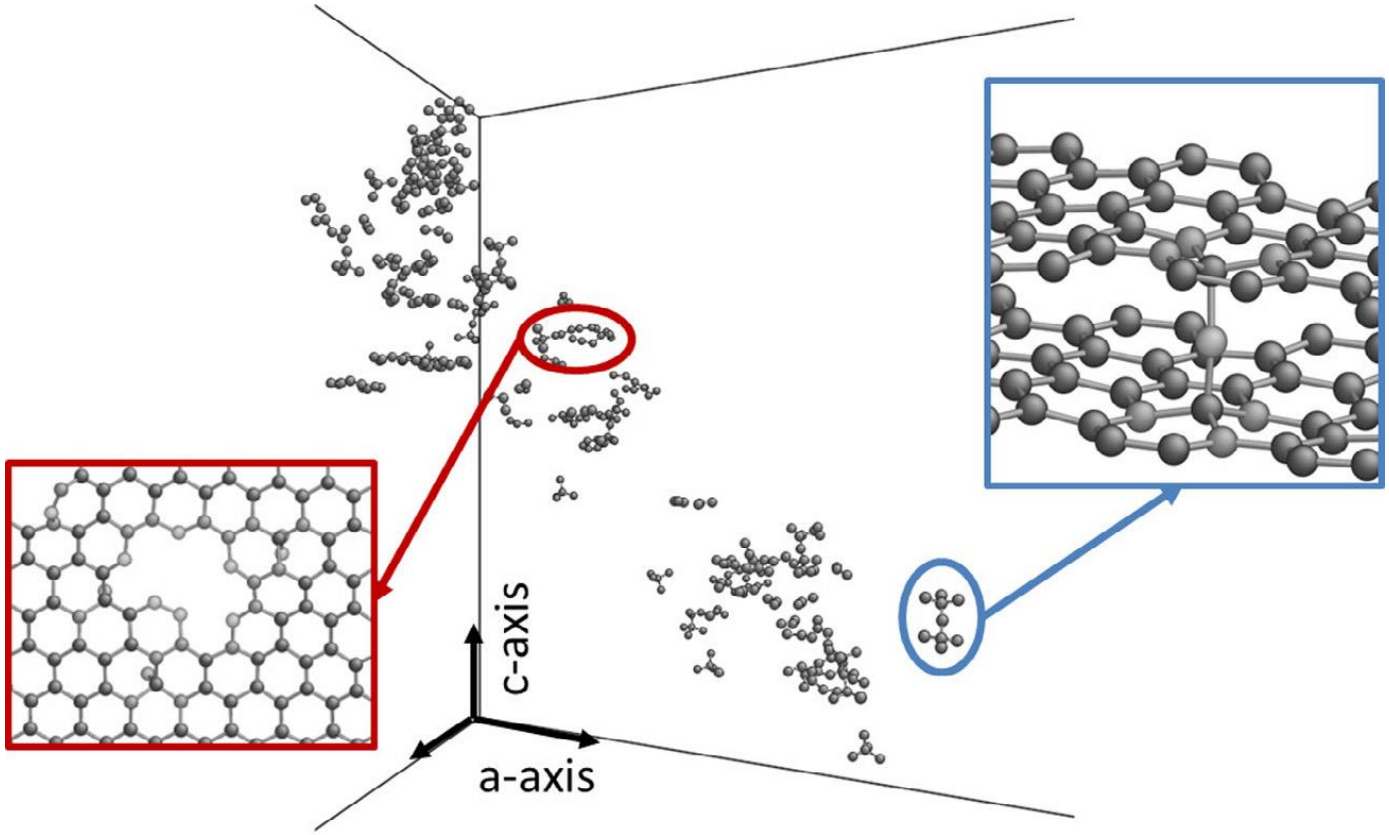
Graphene sheets are discriminated by local curvature
→ average curvature radius
→ size of graphene clusters



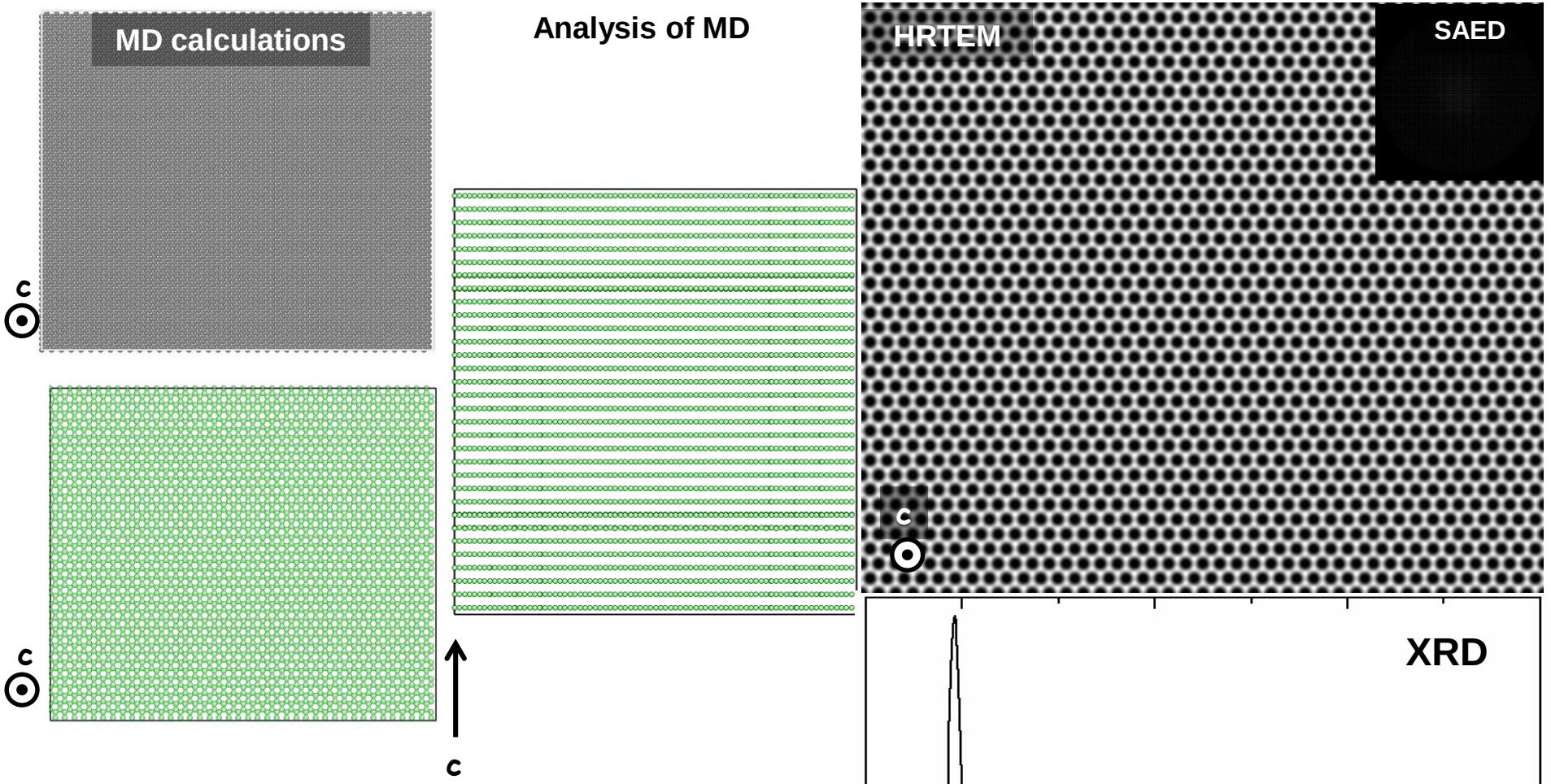



0.04 ps
0.05 ps
0.1 ps
0.5 ps
1 ps
8 ps
c-axis
a-axis

Graphite – displacement cascade



Graphite - Analysis with DXA, HRTEM and XRD



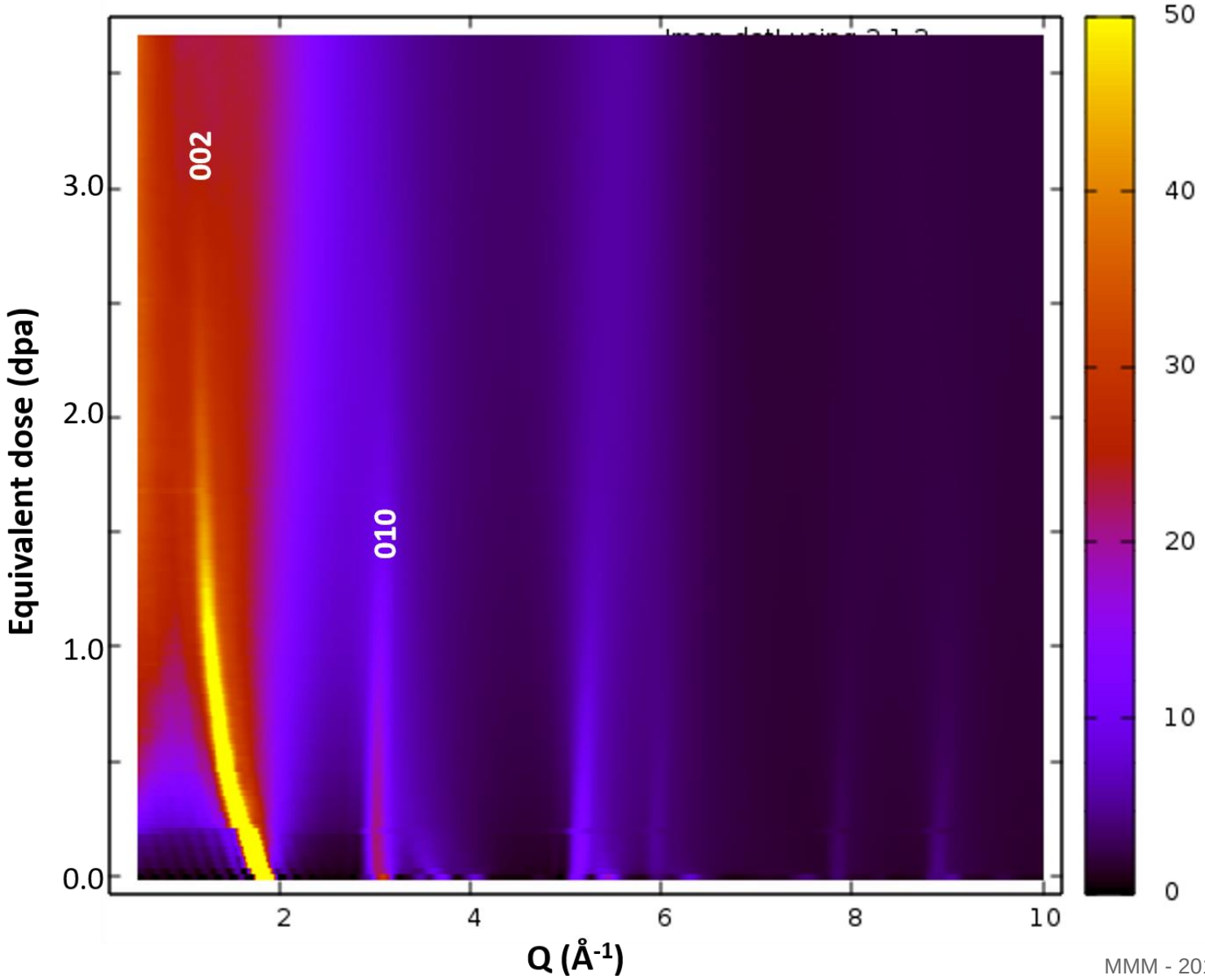
→ Vizualization of defects in graphene planes (MD and HRTEM)

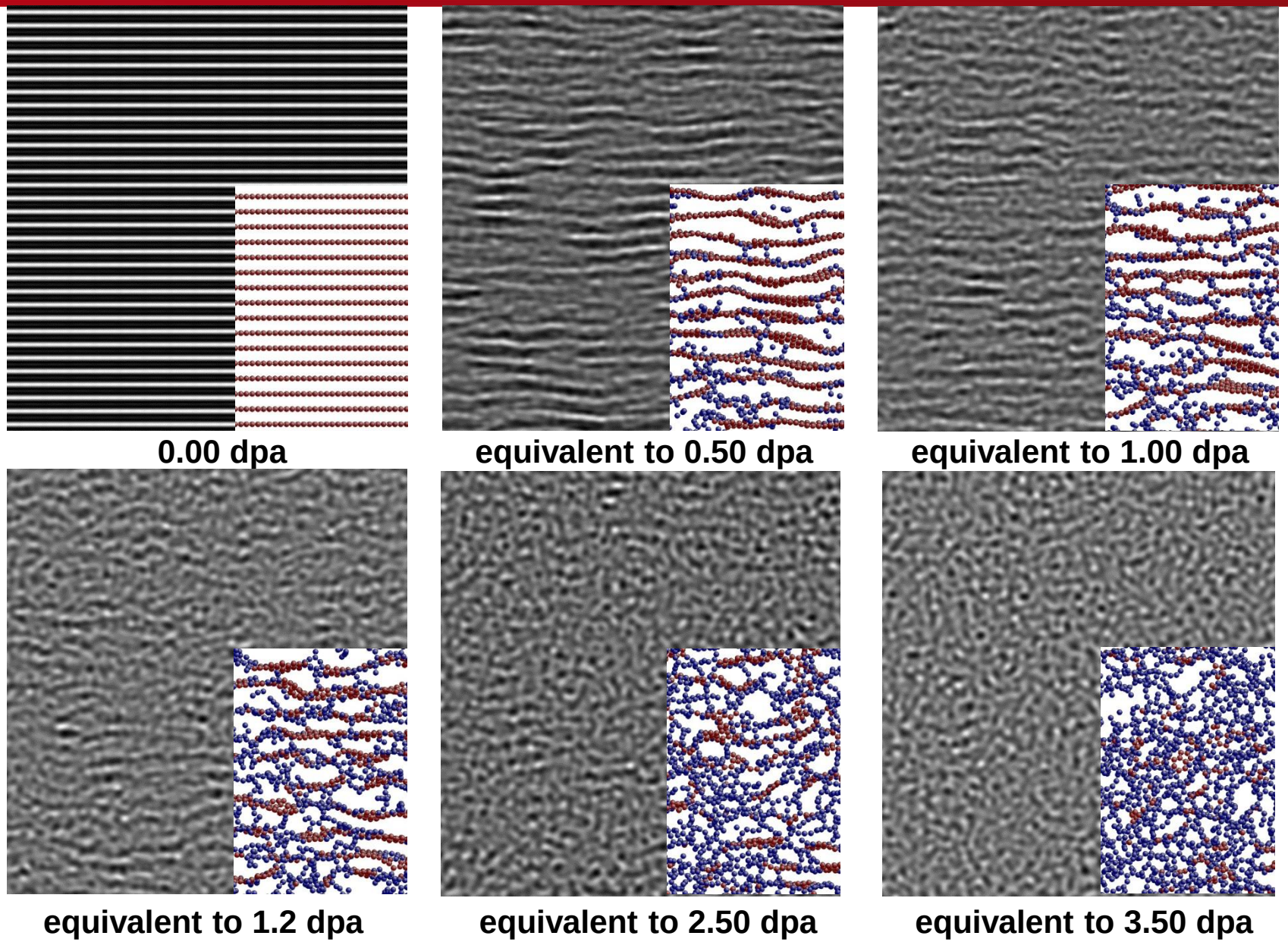
→ Vizualization of wrinkles (MD analysis)

→ Swelling from size of box (not easy from XRD)

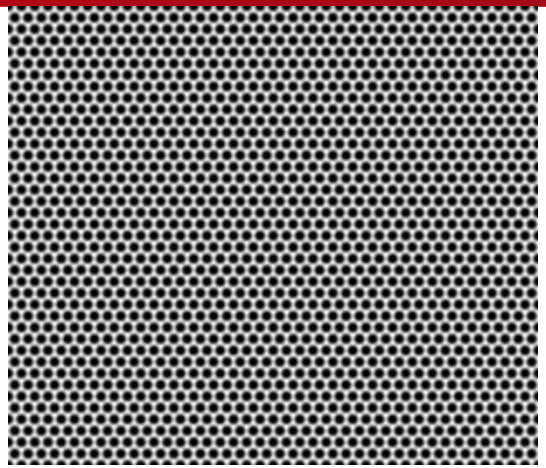
→ amorphization at 0.10 dpa (SAED and XRD)

Graphite – simulated XRD

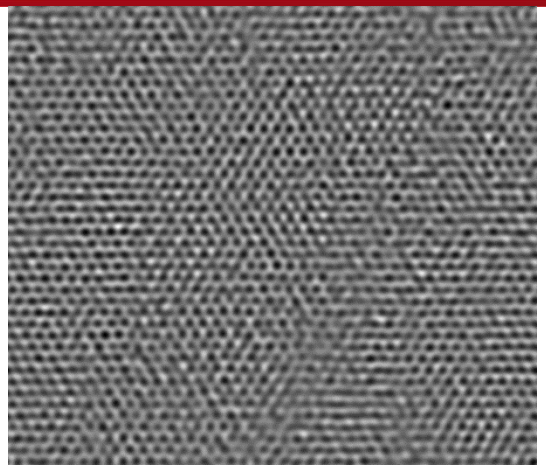



5. nm
0.00 dpa
equivalent to 0.50 dpa
equivalent to 1.00 dpa
5. nm
equivalent to 1.2 dpa
equivalent to 2.50 dpa
equivalent to 3.50 dpa

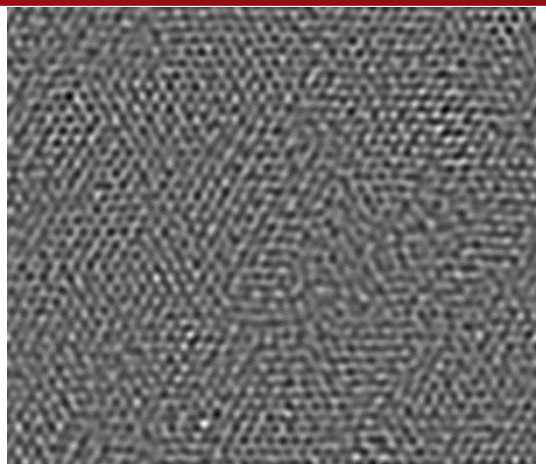
Graphite – simulated HRTEM



0.00 dpa

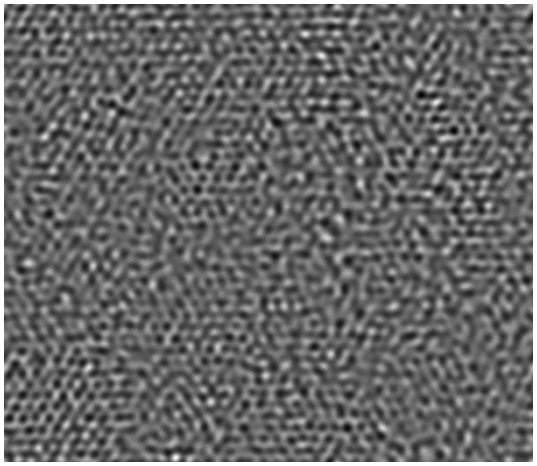


equivalent to 0.50 dpa

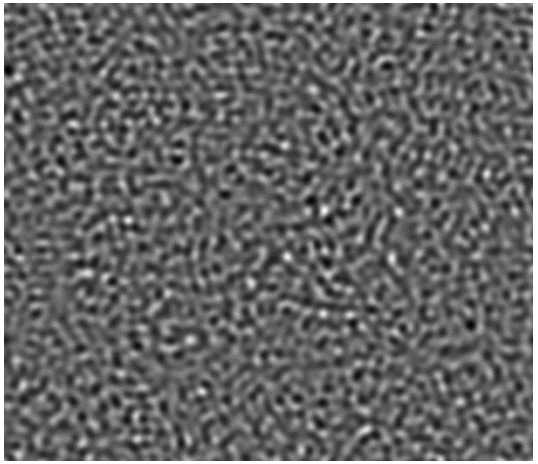


equivalent to 1.00 dpa

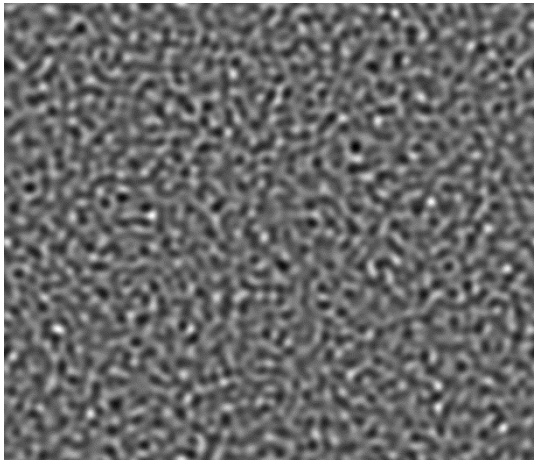
5. nm



equivalent to 1.2 dpa



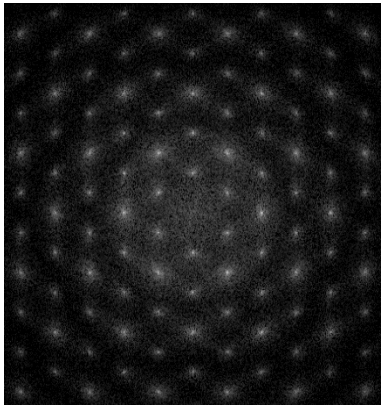
equivalent to 2.50 dpa



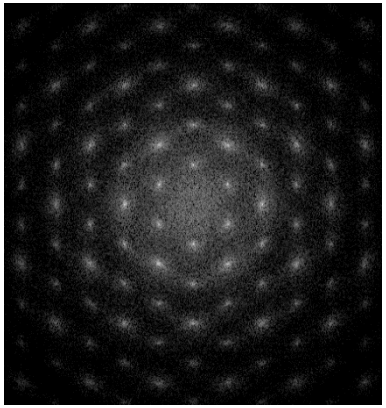
equivalent to 3.50 dpa

5. nm

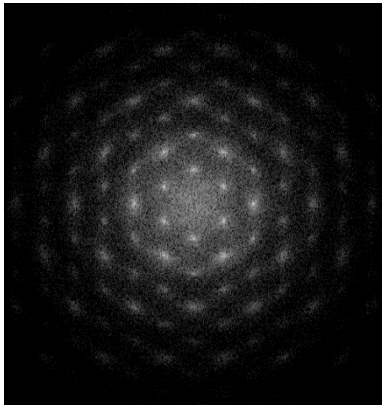
Graphite – simulated SAED



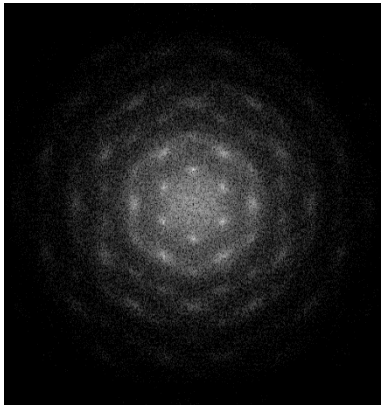
0.00 dpa



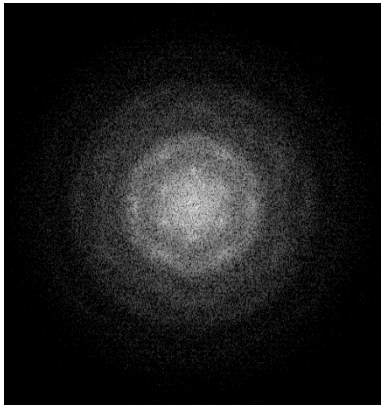
equivalent to 0.50 dpa



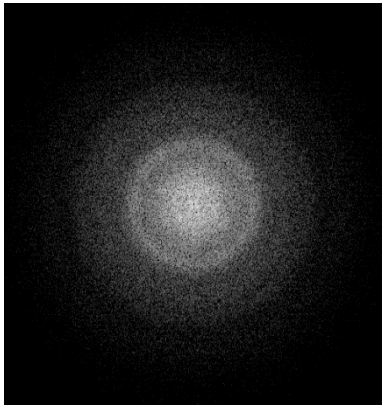
equivalent to 1.00 dpa



equivalent to 1.2 dpa



equivalent to 2.50 dpa



equivalent to 3.50 dpa