



# Evolution of graphite structure under irradiation : A new scenario

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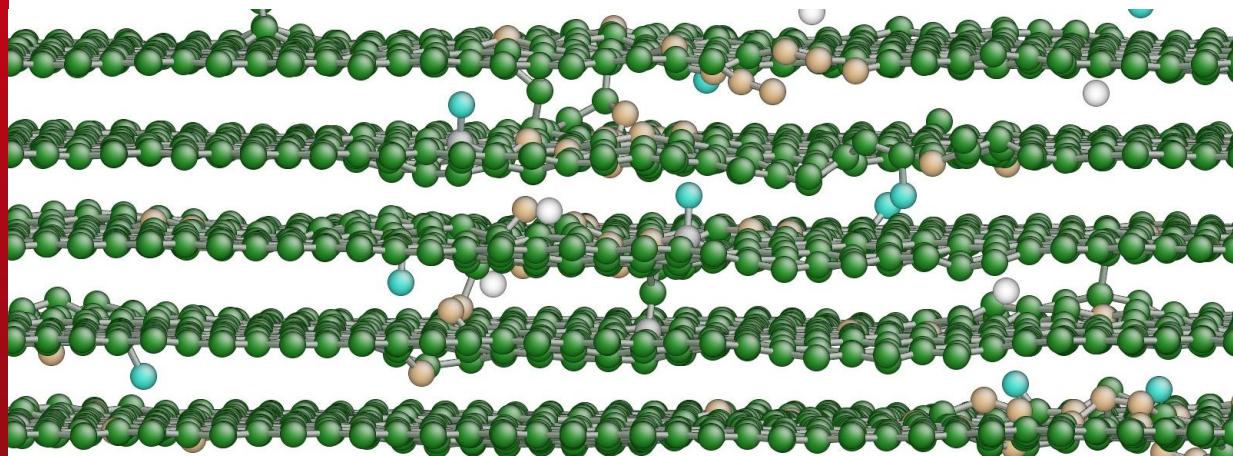
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## EVOLUTION OF GRAPHITE STRUCTURE UNDER IRRADIATION: A NEW SCENARIO

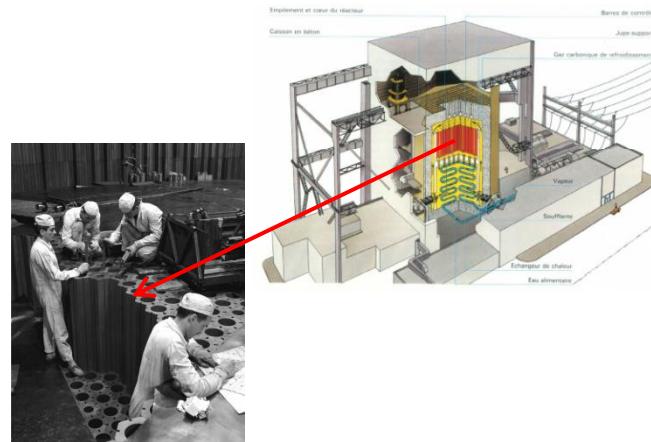


Alain Chartier, Laurent Van Brutzel

# Irradiated graphite in graphite-gas nuclear reactors

**Framework:** decommissioning of graphite from "UNGG"

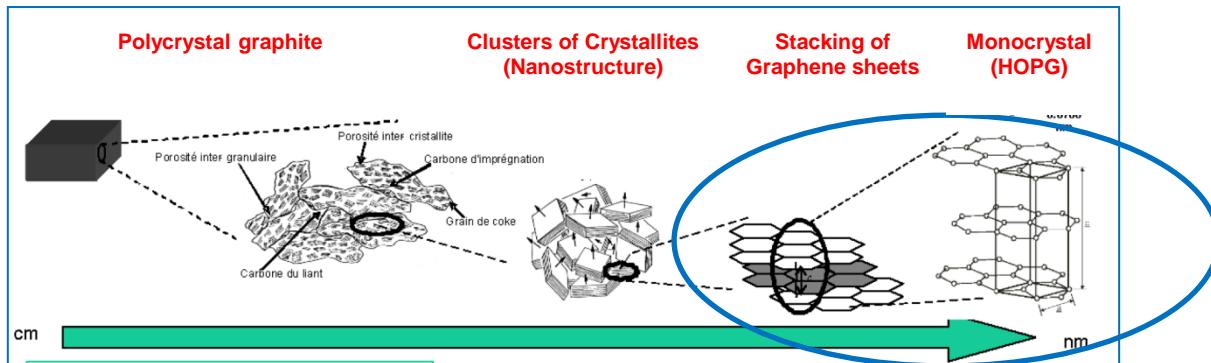
- Graphite = moderator in "UNGG" → neutrons, temperature
  - In 11 years of exploitation **2.6 dpa**
  - Activation of some impurities
- i-graphite contains  $^{14}\text{C}$  &  $^{36}\text{Cl}$  ( $\frac{1}{2}$  life 5 500 &  $3 \times 10^5$  years)



**Foreseen Solutions:**

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

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



Simulation of the primary damage  
with molecular dynamics  
simulations

# Scenarii of radiation damages of graphite

## Question:

Neutron damages in nuclear graphite?

## Answers:

- Some review articles

Heggie (2007)

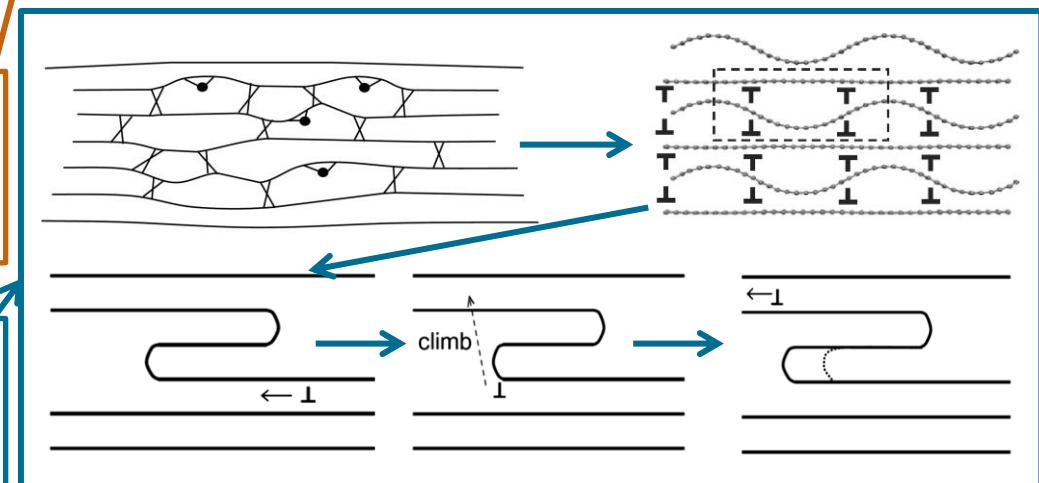
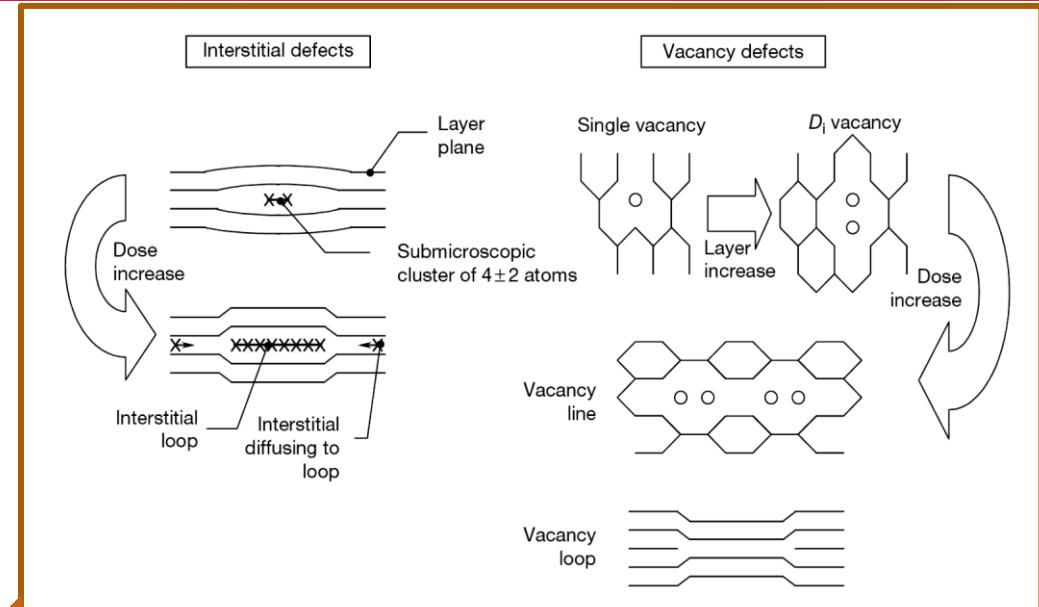
Marsden, Burchell and Fachinger (2012)

However, few data on  
Primary Damage State

- Two main scenarii proposed to describe radiation damages:

1. Creation of point defects, then clustering via diffusion and dislocation formation

2. Creation of point defects, then wrinkling, sliding, and dislocation formation  
(Heggie & Telling)



# Modeling of irradiations

## Study of Primary Damage with Molecular Dynamics

Code: LAMMPS

Empirical potential: LCBOP (bond-order)



### Primary Damage: Displacement Cascades

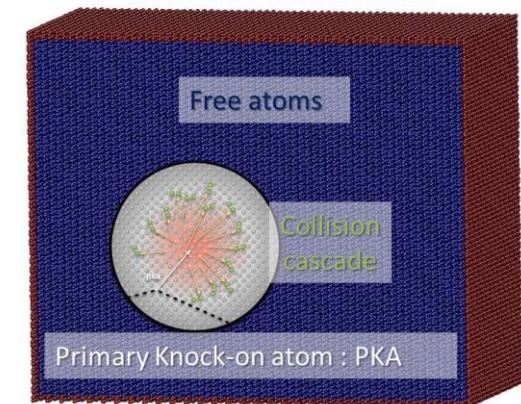
Primary Knock-on Atom: C

$E_{PKA} = 0.5 \rightarrow 10 \text{ keV}$

$T = 300 - 1000 \text{ K}$

Statistics: 16 cascades /  $E_{PKA}$

Thermostat : frozen atoms



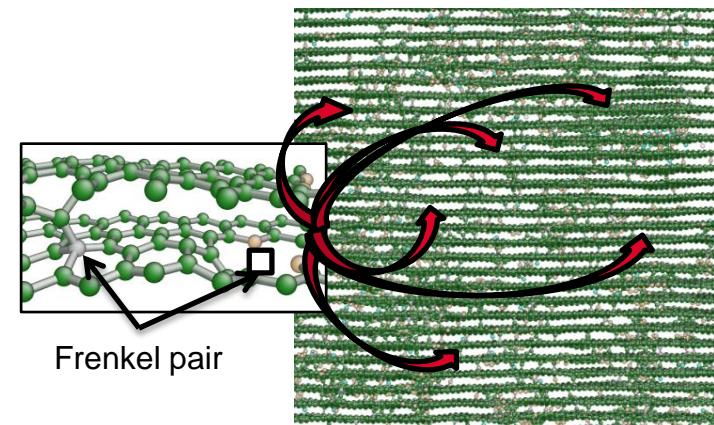
### Dose Effects: Point Defects Accumulation

Creation of random Frenkel pairs

Dose rate:  $10^{10} \text{ dpa/s}$

Dose:  $0.1 \rightarrow 1 \text{ dpa}$

$300 - 1000 \text{ K}$  (relaxation NPT)  $\rightarrow$  Swelling



# Primary damage in graphite

Example: Cascade with 5 keV PKA

Projectile C - direction [100] – 300 K

## Three Stages:

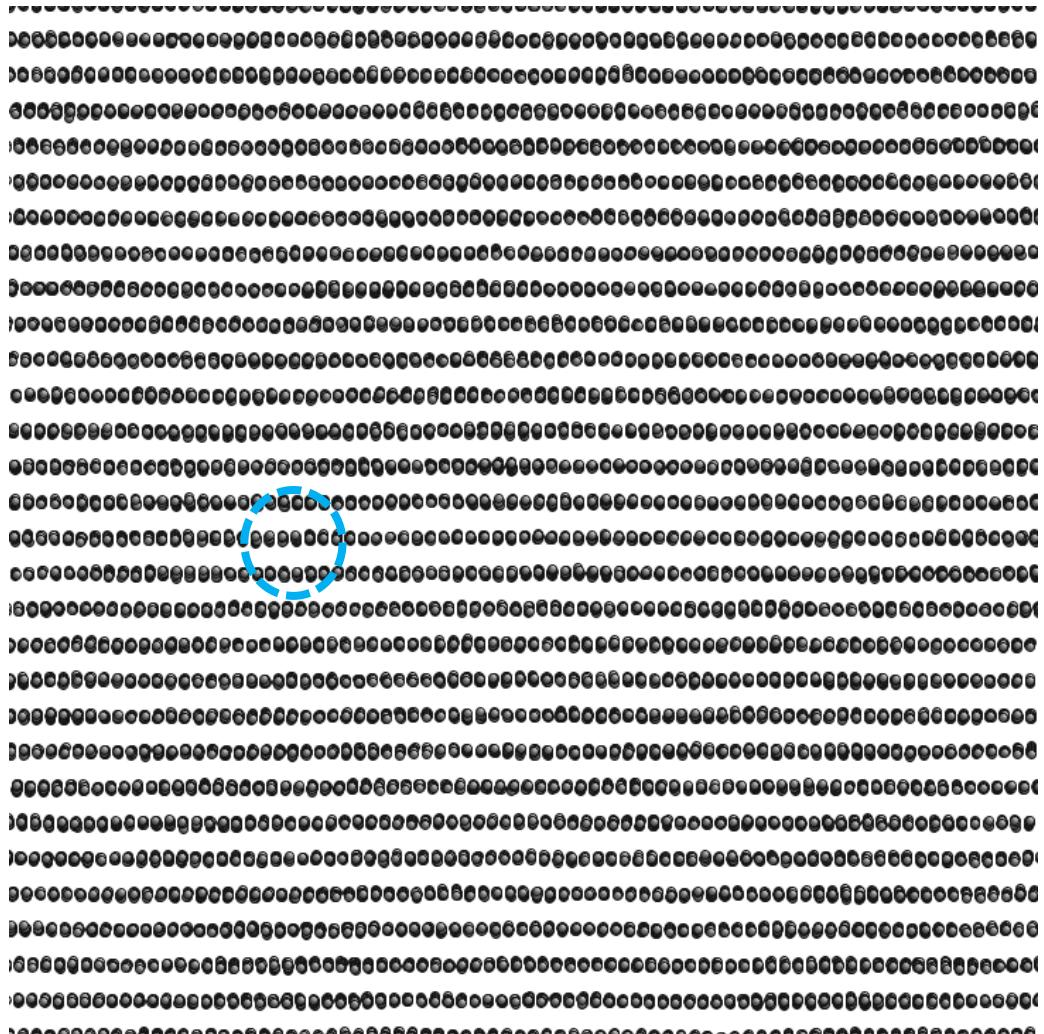
1. Collision Stage : 2 ps
2. Thermal Spike : 5 ps
3. Relaxation : 10 ps

## Damage:

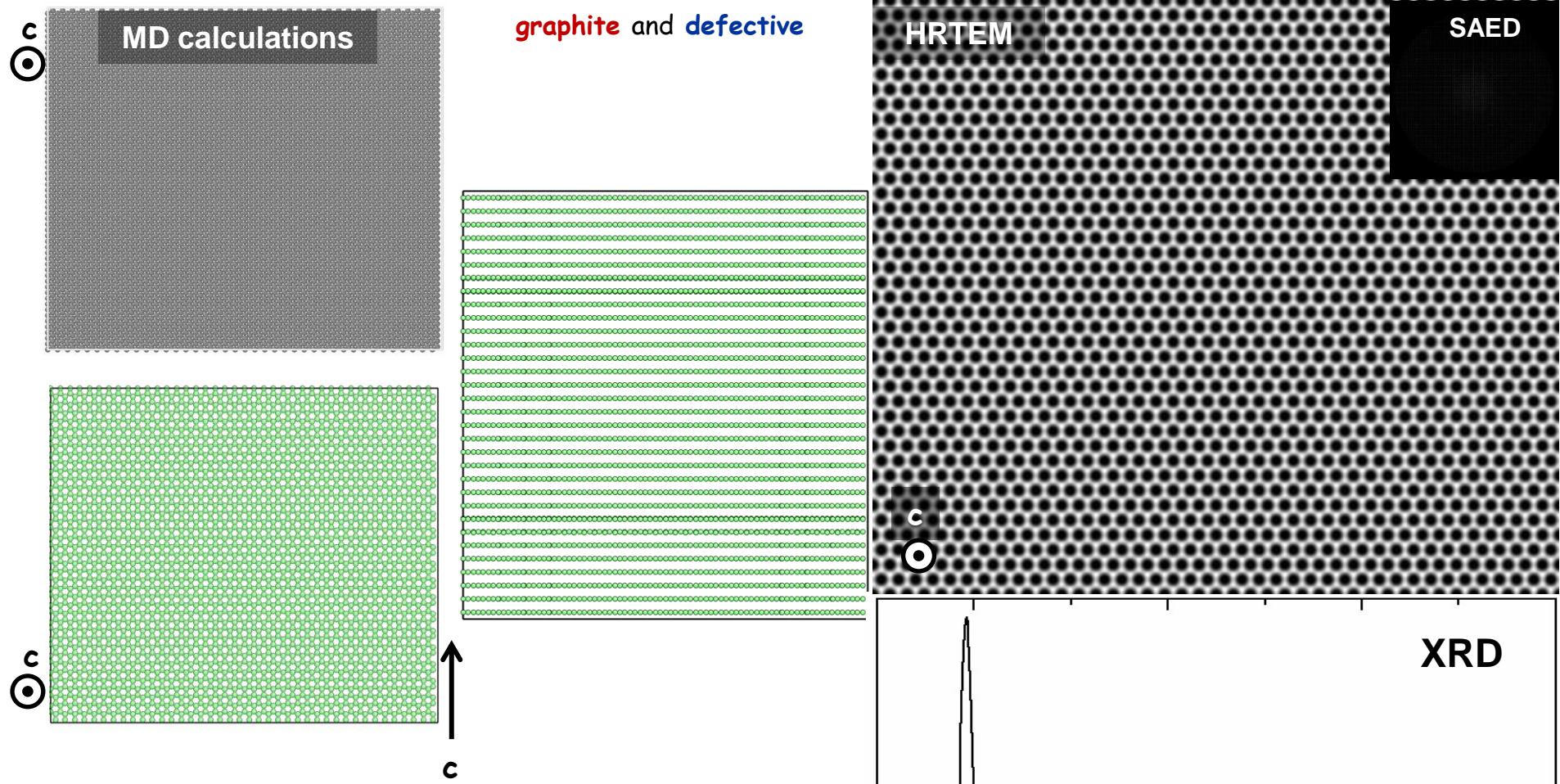
- No Amorphisation
- Numerous Point defects
- Few Recombination
- No influence of the initial PKA orientation
- Number of vacancies  $\approx$  interstitials



→ Point Defect Accumulation  
method



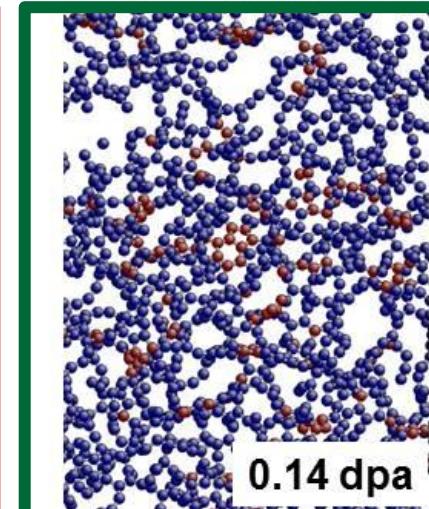
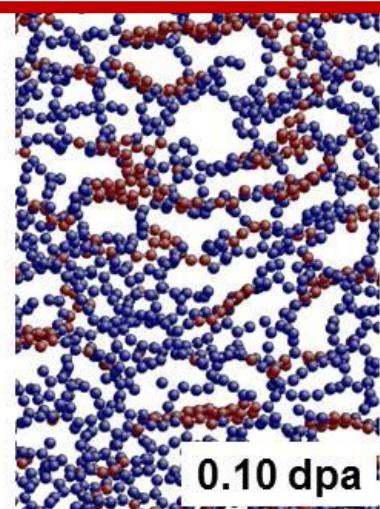
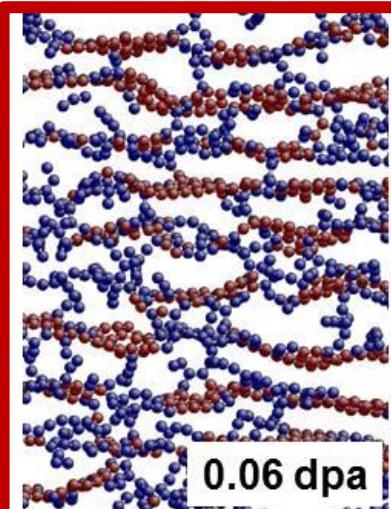
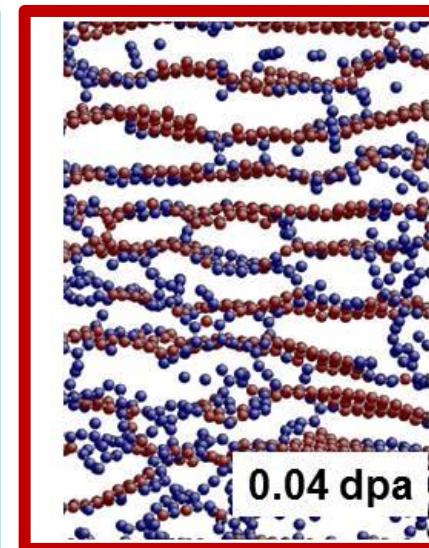
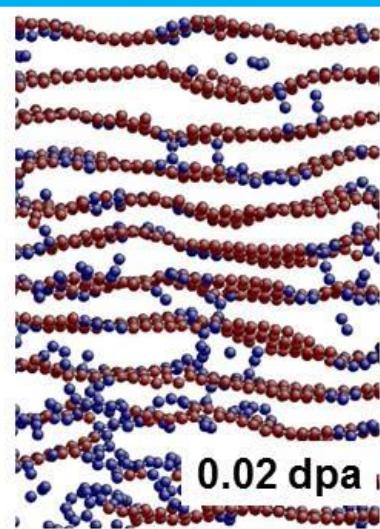
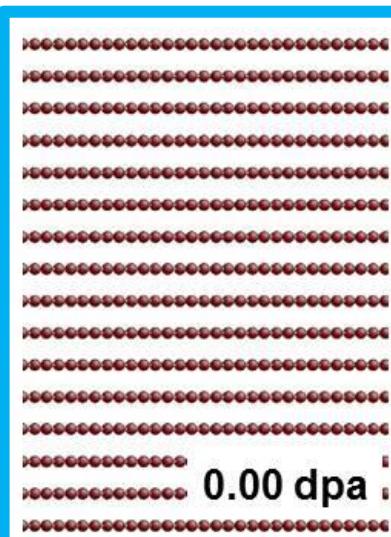
# Dose effect from MD – Analysis from curvature criterion, HRTEM, SAED and XRD calculations



- 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)

# Dose effect modeling

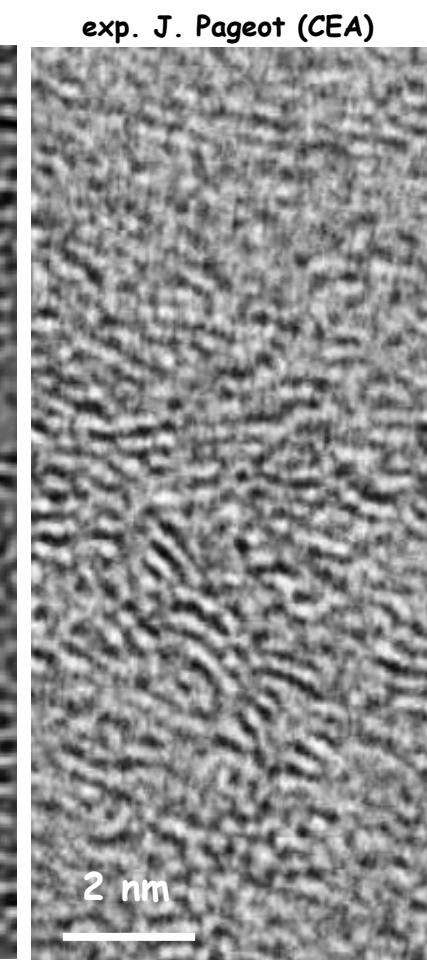
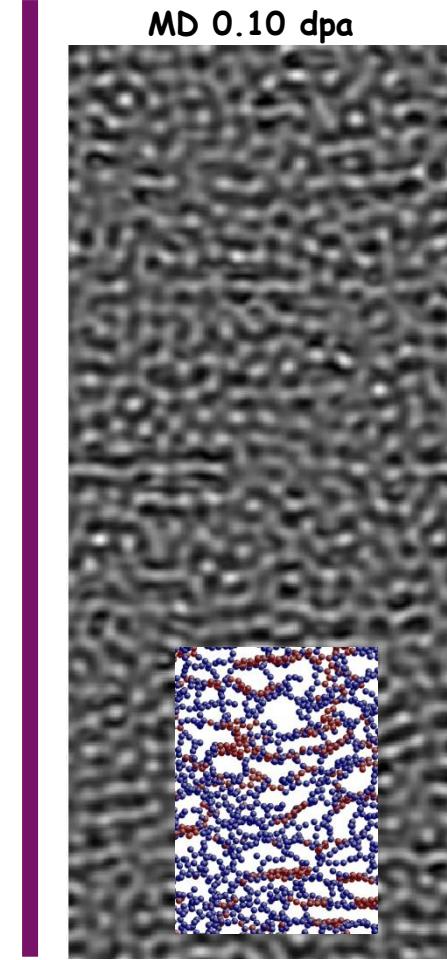
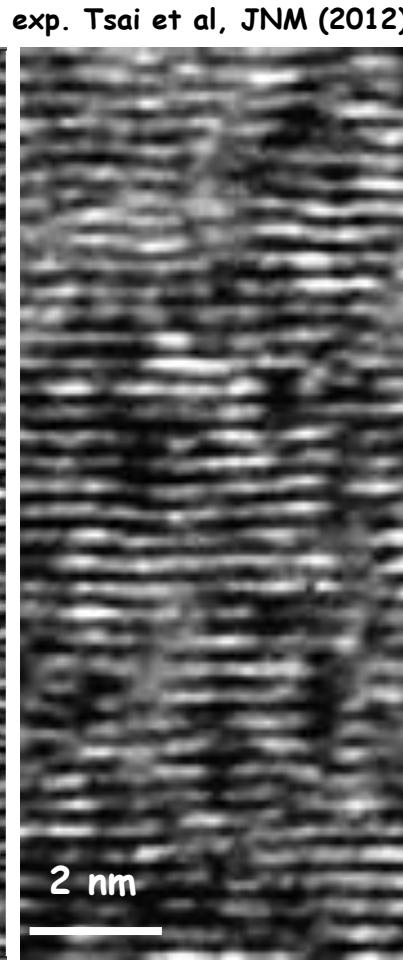
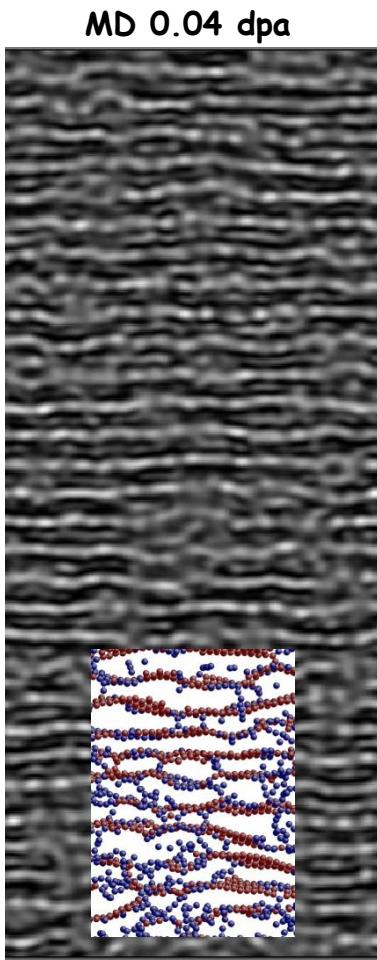
Perfect graphene sheets in red and point defects in blue



Three Steps:

1. Point Defects
2. Pinning on small disordered cluster and Wrinkling of graphene sheets
3. Growth of amorphous pockets  
→ Amorphisation

## HRTEM from atomic simulations



Graphene sheets in red, defects in blue

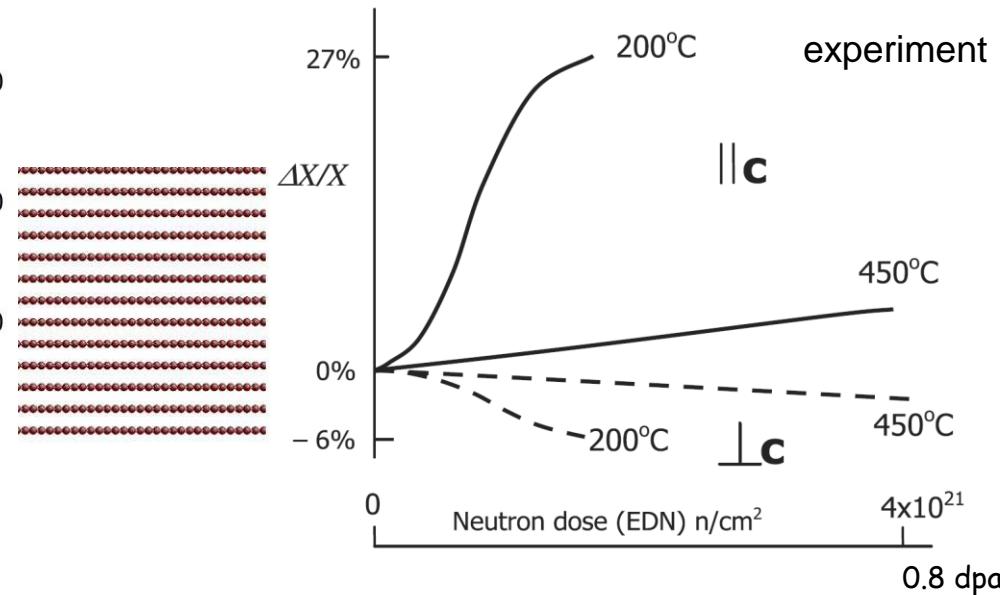
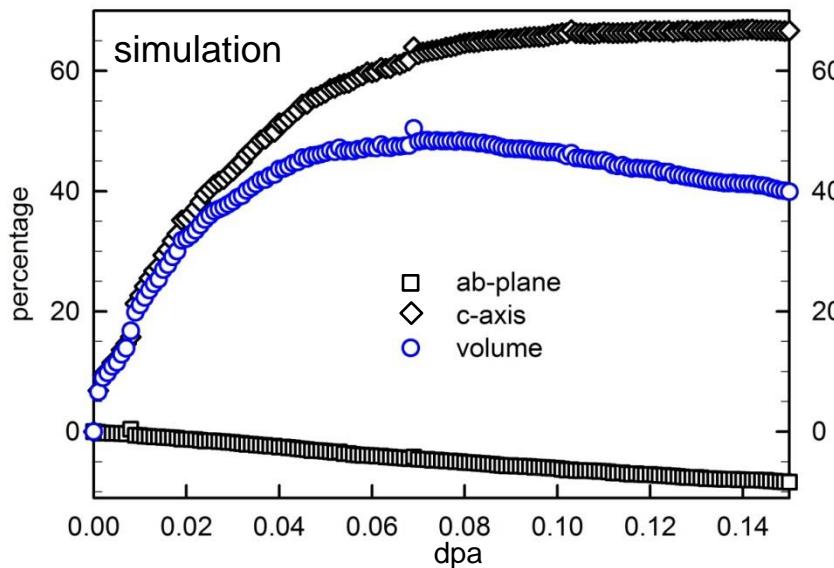
**Common explanation:** Decrease of graphene sheet' size with increasing dose

→ small graphene sheets in "noodles shape"

**What simulations show:** graphite amorphizes at 0.10 dpa

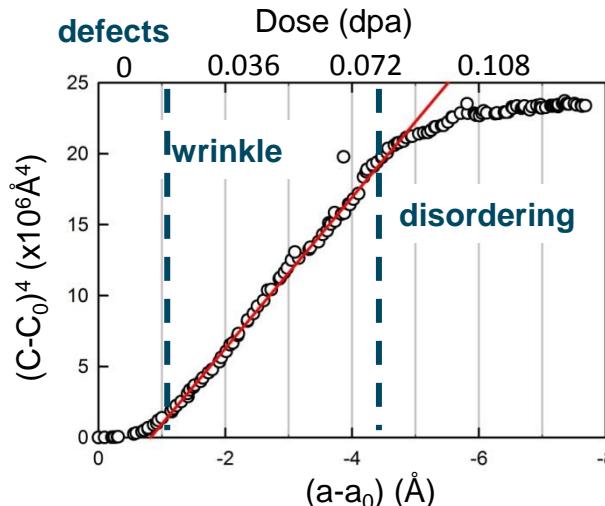
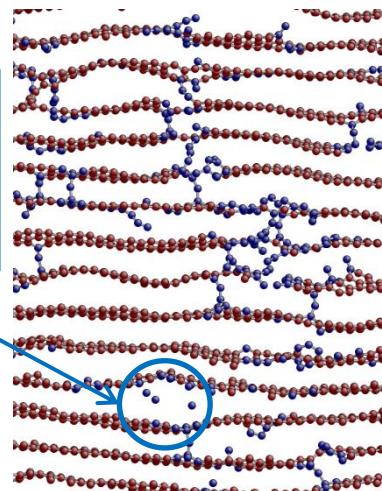


# Swelling under radiation



Expansion along the c-axis and shrinking of the graphene sheets

Interstitials pushing graphene sheets



Wrinkling of  
graphene sheets:  
 $c^4 \propto a$



Tapaszto 2012 Nature  
Physics Letters1

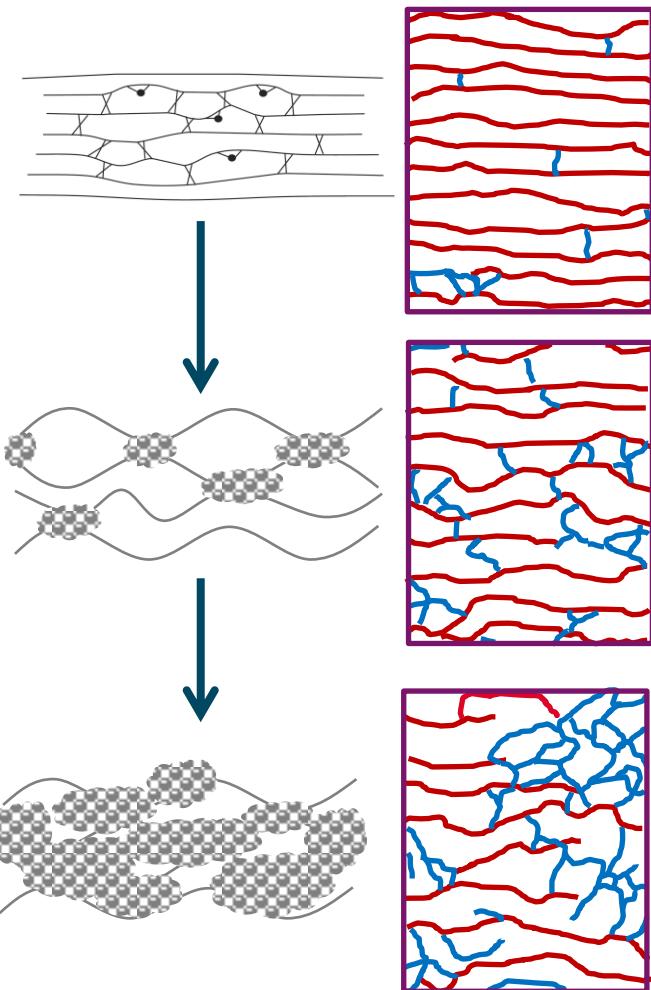
# Summary - Conclusion

## Primary Damage: Displacement Cascades

- Point Defects (large variety)
- Localized along PKA track
- # Vacancies  $\approx$  # Interstitials  $\rightarrow$  Frenkel pairs

## Dose effects: Frenkel Pair Accumulation method

- New scenario for Amorphisation in 3 steps:
  1. Point Defects
  2. Pinning + Wrinkling of graphene sheets
  3. Growth of amorphous pockets  $\rightarrow$  Amorphisation
- Swelling due to both defects + wrinkling of graphene sheets





Challenge project – C097073



# Thank you for listening

## Questions ?

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