IAEA-CN301-159

THE USE OF IN-SITU TRANSMISSION ELECTRON MICROSCOPY TO INVESTIGATE MICROSTRUCTURE EVOLUTION UNDER ION IRRADIATION

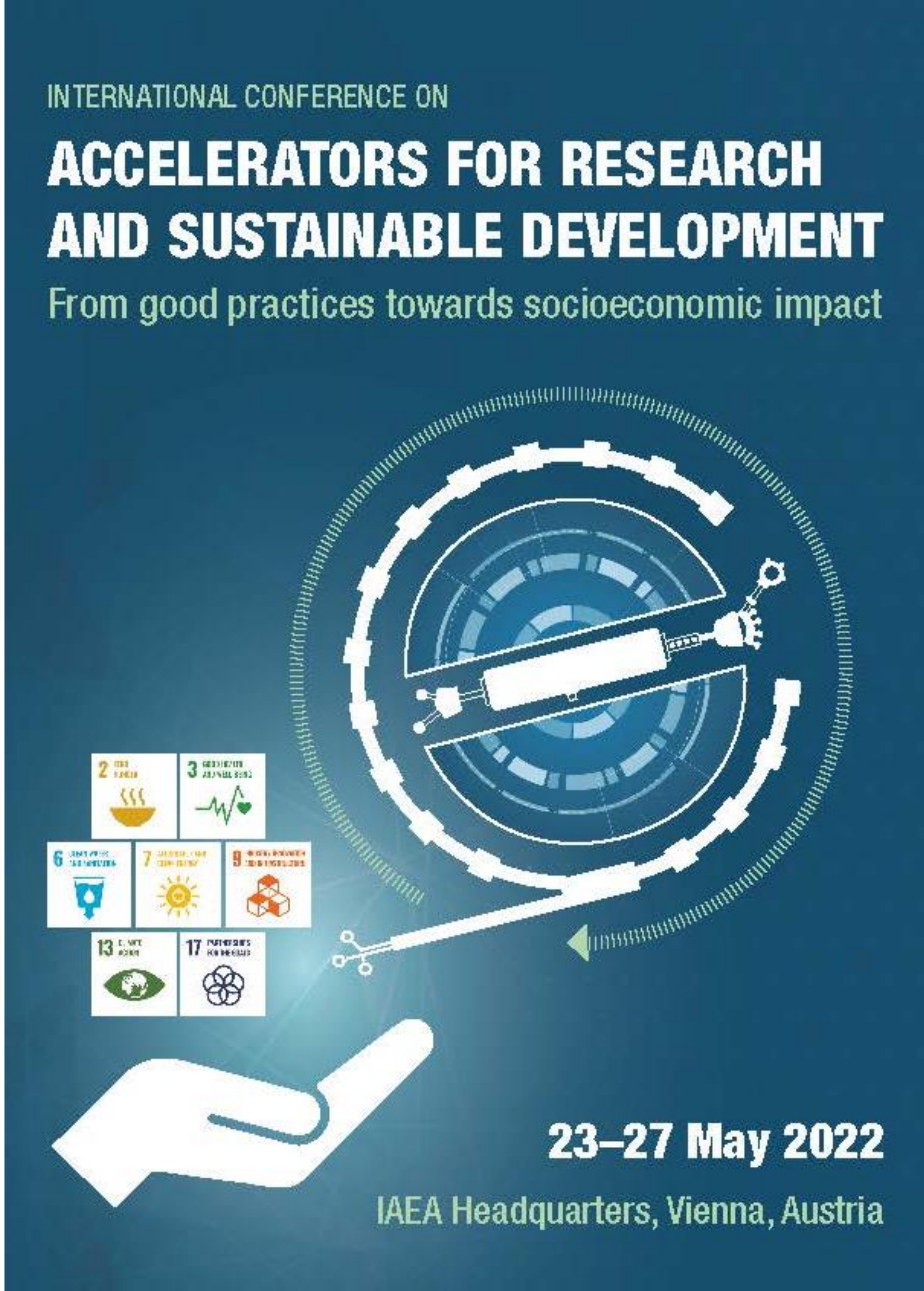
Djamel Kaoumi

.....

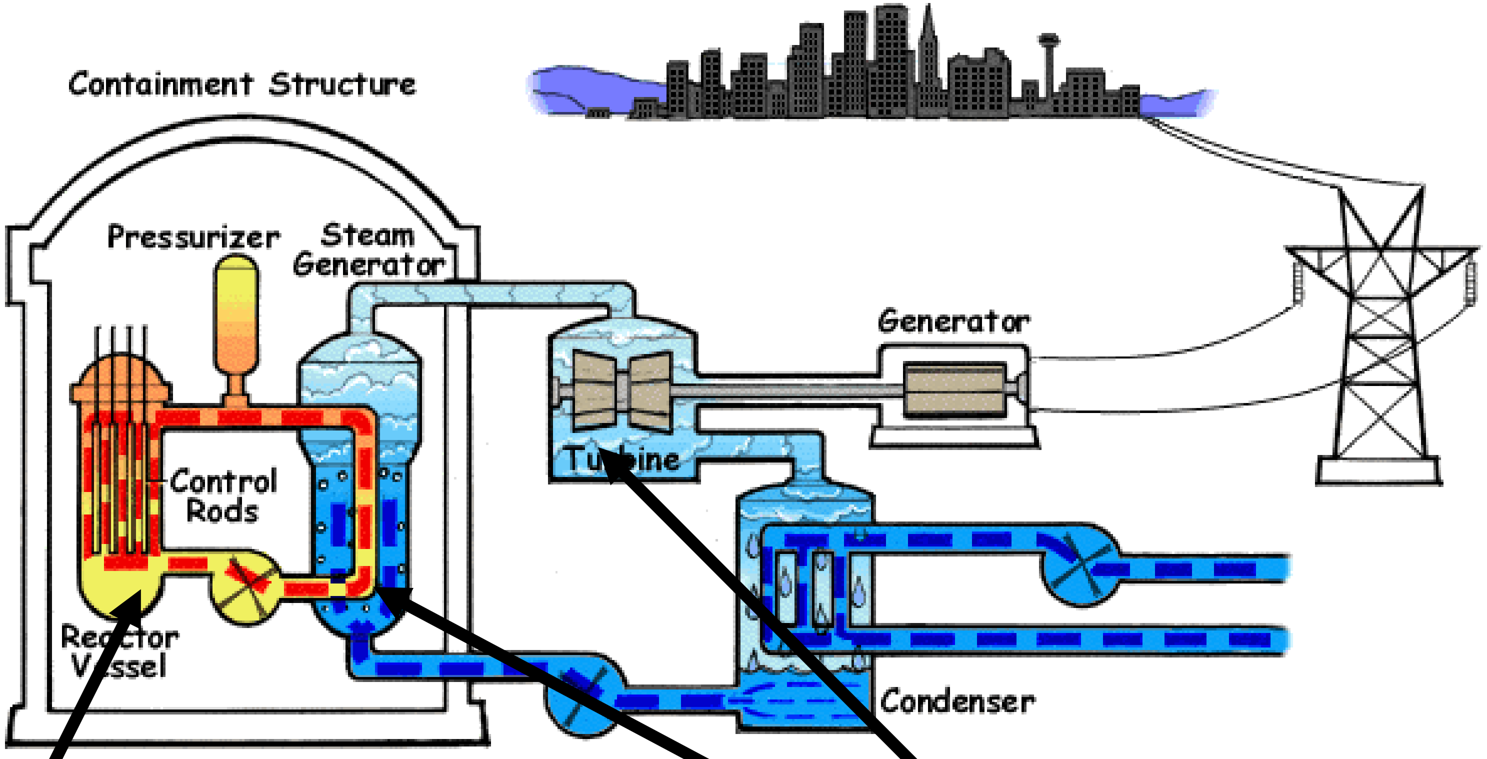
Associate Professor of Nuclear Engineering,

North Carolina State University

dkaoumi@ncsu.edu



Use of materials in Nuclear Power Plants



Irradiation,
Mechanical stress,
High temperature,
Corrosive medium

High temperature,
Corrosive medium,
Mechanical stress,

Introduction

Microstructure: average grain size, grain size distribution, alloying elements, second-phase precipitates

Material properties: strength, ductility, corrosion resistance...

Microstructure governs properties in materials.

Microstructure changes under irradiation/temperature: atoms are displaced, vacancies and interstitials, clusters, loops, phase transformations, precipitation or dissolution of second phases.

=>As a result, the materials properties are altered which ultimately can cause failure.

Introduction

Microstructure: average grain size, grain size distribution, alloying elements, second-phase precipitates

Material properties: strength, ductility, corrosion resistance...

Microstructure governs properties in materials.

=> Understanding microstructure evolution under irradiation and temperature which is at the root of macroscopic effects => to predict the materials performance and to design better alloys for high-dose cladding (high burn-ups), and structural applications in advanced reactor designs (e.g. Gen-IV fission, fusion).

In-situ experiments: source of kinetics information

- One of the difficulties of studying the microstructure evolution under external solicitation (irradiation/temperature) in materials is the lack of kinetics information. Samples are usually examined *ex-situ* after the sample was cooled down.

- Given the dynamical nature of the microstructure evolution, direct in situ observation is useful for better understanding the mechanisms and driving forces of the processes involved.

- In-situ irradiation Transmission Electron Microscopy (TEM) can be useful for that matter. The spatial resolution of the TEM makes it an ideal environment in which one can continuously track the real-time response of a system to an external stimulus, which can help discover and quantify the rate-limiting *micro*scopic processes and mechanisms governing the *macro*scopic properties.

In-Situ Experiments:

In-situ irradiations:

- **The IVEM: Transmission Electron Microscope** (Hitachi 9000; operating voltage 100-300 kV; maximum resolution of 0.25 nm) **interfaced with an ion accelerator.**

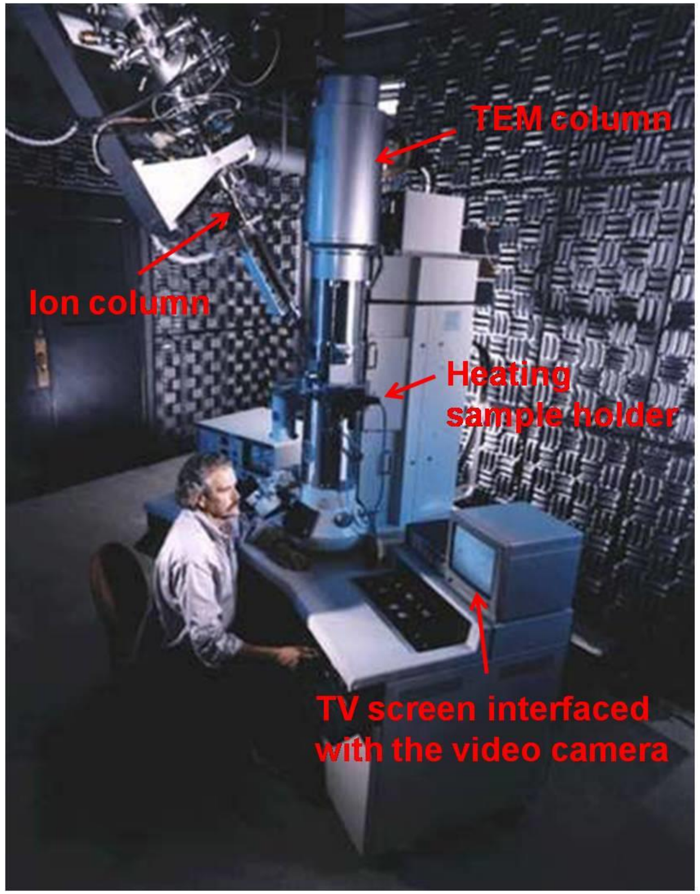
- Choice of:
  - the ion type, (ex: Ar, Kr, Fe..), energy
  - the ion flux (or dose rate),
  - the irradiation temperature (20-1200 K)

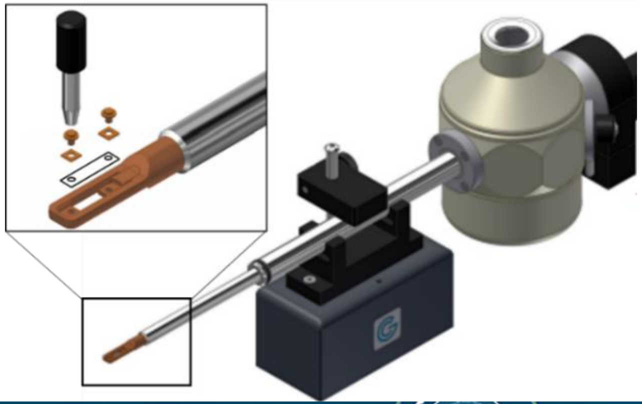
- **Damage observed directly as it proceeds in the material**. Thus, it provides information on the **kinetics** of the phenomena and the **synergy** of the processes involved.

- **Ion irradiation enables to reach high doses much quicker than neutron irradiation**.

In-situ straining:

The sample is strained while the microstructure is followed under the TEM; it provides information on the **dislocation dynamics**.



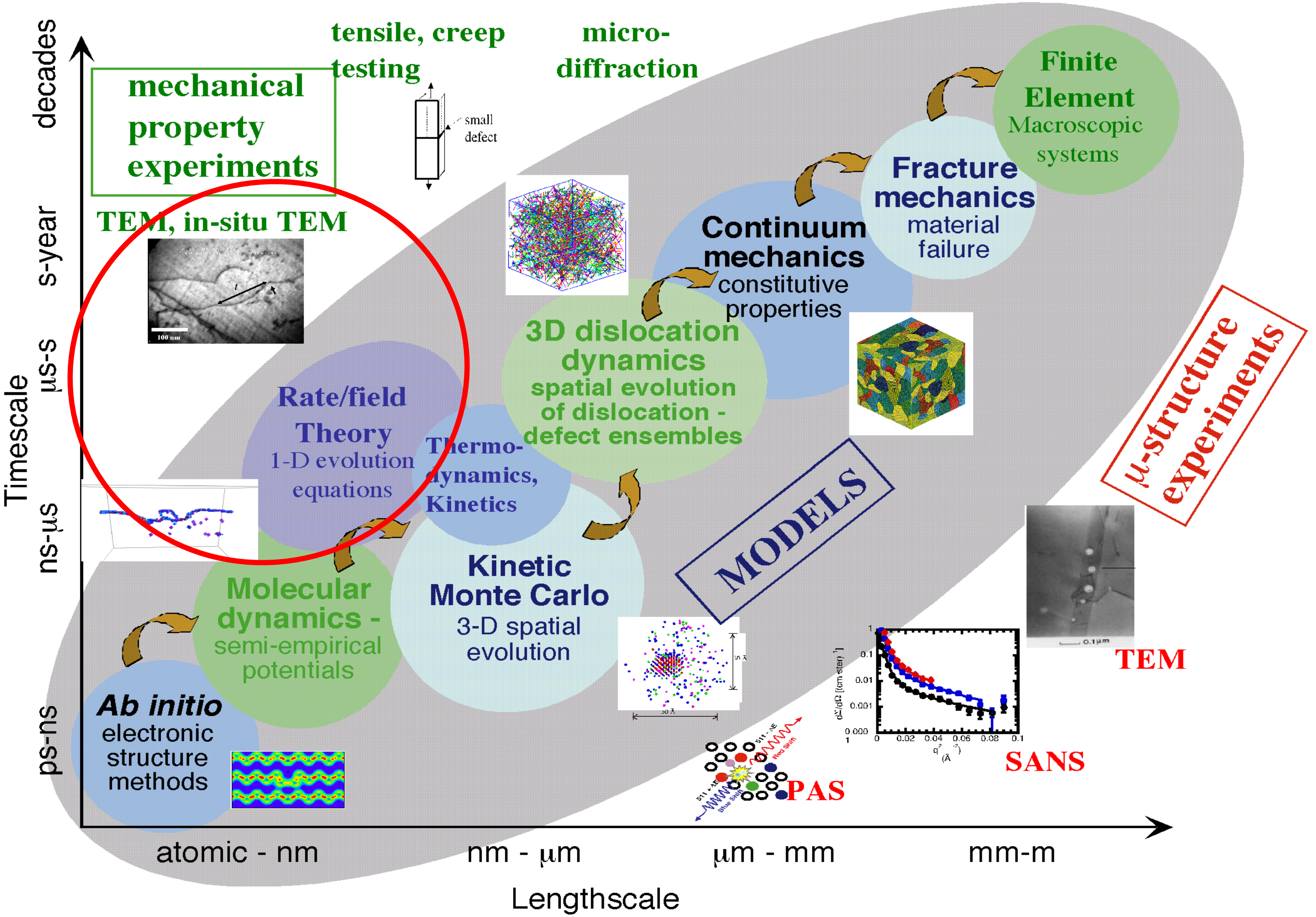


Examples of IN-SITU ION IRRADIATION in a TEM

- *Example 1:* Use of **in-situ irradiation** to study radiation damage in structural alloys

- *Example 2:* Use of **in-situ irradiation** to study and model irradiation induced grain growth in nanocrystalline metals

Radiation damage: a multiscale phenomenon



Different modeling approaches depending on the scale

Bridges between different time/space scale approaches is often lacking

Radiation damage kinetics data

Systematic Irradiation experiments:

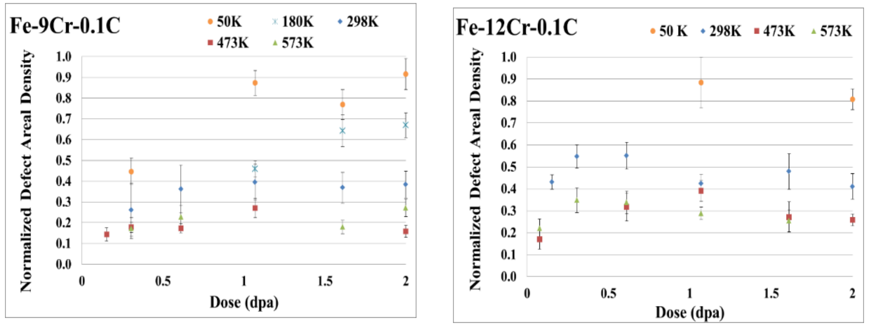
By following the same area we can measure kinetic data:

– Defect areal density vs. dose

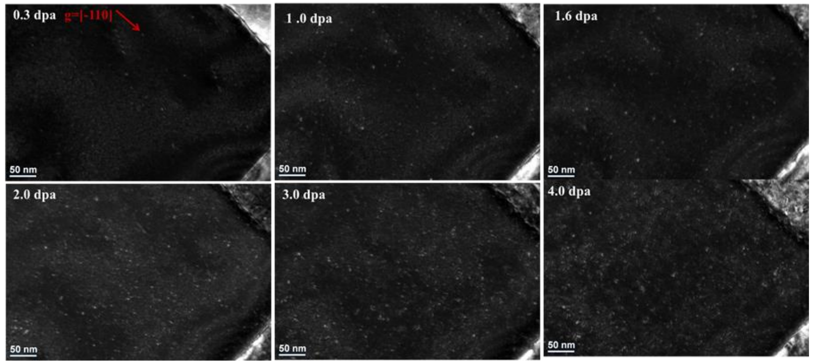
– Loop size distribution vs. dose

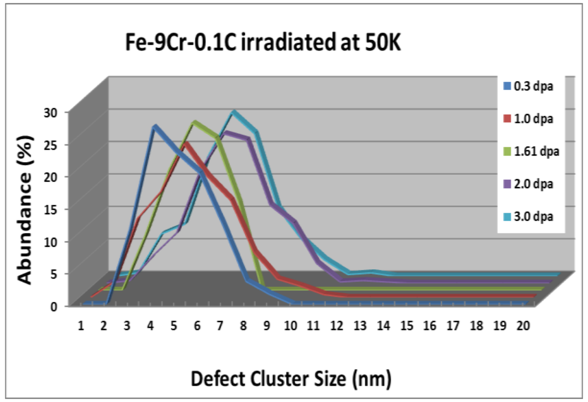
– Temperature effect

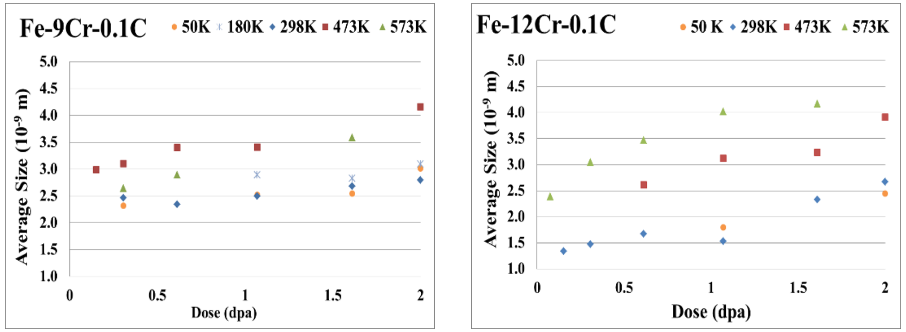
– Spatial correlation of defects with pre-existing microstructure / Mechanisms of interaction / fate of the pre-existing microstructure



Defect areal density vs. dose vs. irradiation temperature for the 9Cr model alloy and 12Cr model alloy







Average visible defect size vs. dose vs. irraditation temperature for the 9Cr model steel and 12Cr model steel.

Dark Field TEM micrographs of the 9Cr model alloy irradiated at 50K to doses ranging from 0.3 to 4 dpa with 1MeV Kr^{2+} ion (z=[001] g=[-110]); (bottom) Corresponding cluster size distributions.

1. *Journal of Materials Research*, v 30, n 9, p 1246-1274, 2015
2. *Journal of Nuclear Materials*, v 466, p 179-186, 2015
3. *Journal of Nuclear Materials*, 448: p 233–238, 2014
4. *Journal of Nuclear Materials*, 445 (1–3): p. 12–19, 2014

Spatially dependent cluster dynamics model equations

Use spatially dependent cluster dynamics $C_i(x,t)$

Diffusion term

Production Term

Loss due to Sinks

$$\frac{\partial C_i}{\partial t} = \nabla(D_i \nabla(C_i)) + \dot{\varepsilon} f(i)\frac{1}{\Omega} - D_i C_i S_i +$$

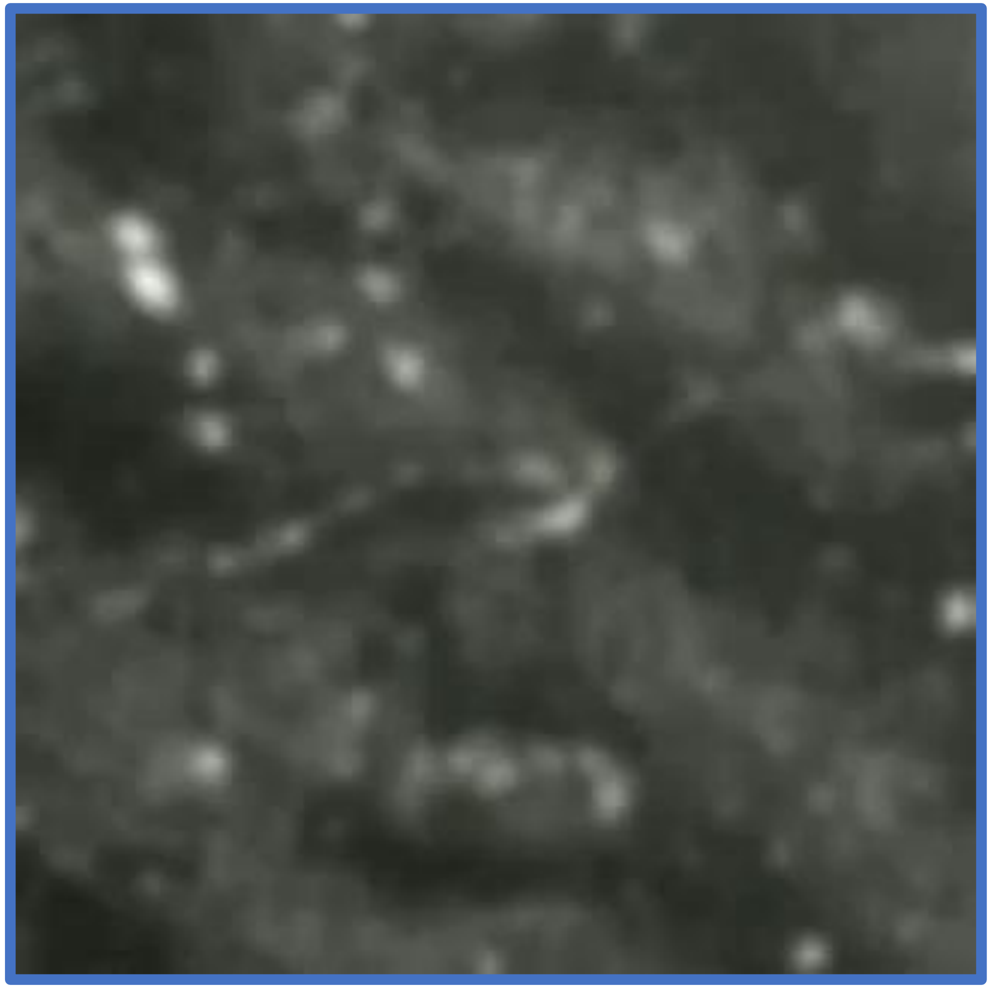
$$\underbrace{\sum_{\substack{n+p=i \\ (n,p)\text{ in the range}}} k^+_{n,p} C_n C_p - \sum_{\substack{n+i=p \\ n\neq i \\ (n,p)\text{ in the range}}} k^+_{n,i} C_i C_n - \underset{\text{if } 2i \text{ in the range}}{2k^+_{i,i} C_i^2} +}_{\text{Recombination terms}}$$

$$\underbrace{\underset{(\text{if } i+1 \text{ in the range})}{k^-_{i+1}\frac{1}{\Omega} C_{i+1}} - k^-_i \frac{1}{\Omega} C_i}_{\text{Dissociation terms}}$$

- The capture constants are calculated using the rate theory model:

$$k^+_{i,j} = 4\pi(r_i + r_j)(D_i + D_j) \quad \text{and} \quad k^-_i = k^+_{i-1,1} e^{-\frac{Eb_i}{kT}}.$$

Dynamic picture of radiation damage formation and evolution



Fe-9Cr-0.1C model alloy irradiated at RT

In-situ Experiments offer a more accurate picture of radiation damage in terms of its inherent dynamic nature:

- Defects form, move, annihilate,

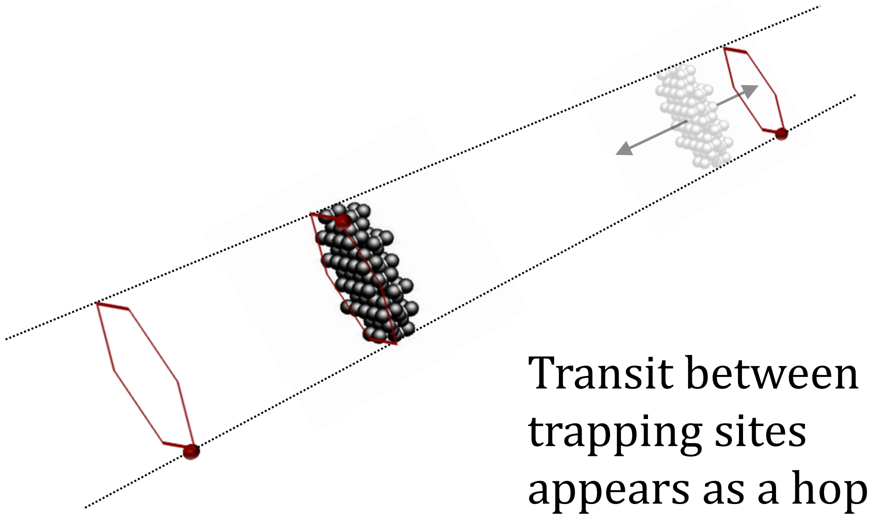
- Loops are sessile at trapping sites but then can glide is certain crystallographic directions

=> More accurate depiction of defect motion helps improve predictive models or diation damage kinetics

Trapping / Detrapping of loops and improvement of predictive models

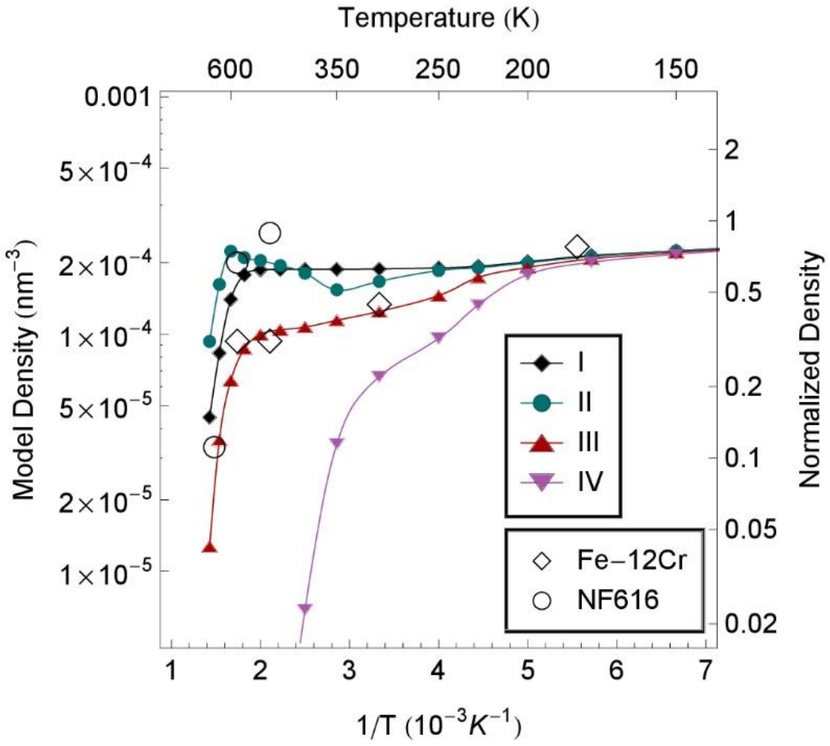
Following observations of discrete hops during *in situ* experiments, Trap mediated diffusion was introduced to the model:

- Loops are sessile at trapping sites
- Occasional detrapping in cascade events
- Low activation glide between traps



Transit between trapping sites appears as a hop

Improving and Benchmarking predictive models

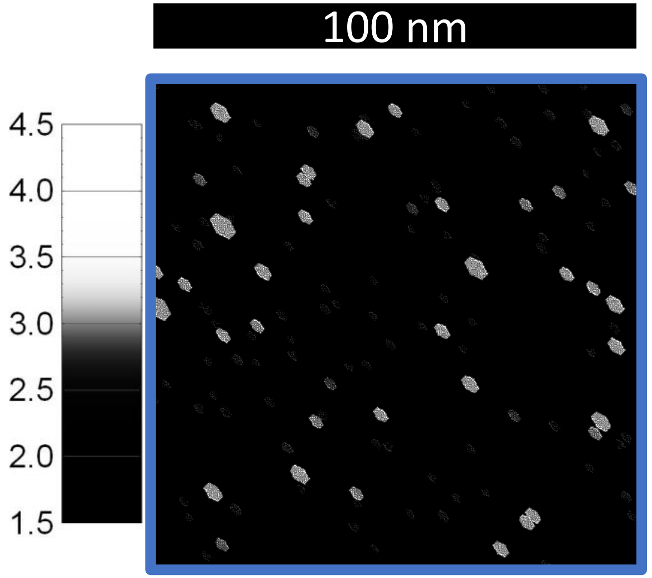


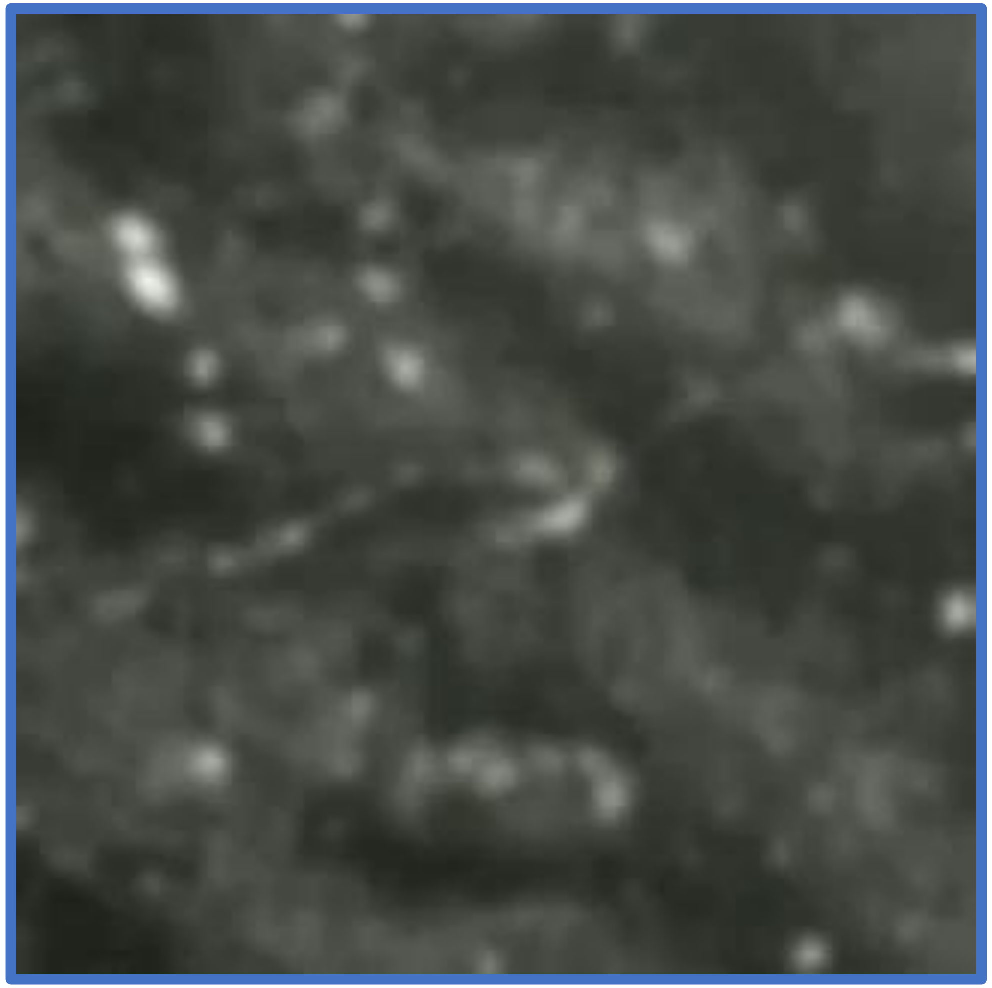
Fe-12Cr - D. Kaoumi et al J. Nucl. Mater. 445 (2014) 12–19
NF616 - C. Topbasi et al J. Nucl. Mater. 425 (2012) 48–53

Object Kinetic Monte Carlo

- Rate theory cross compared with OKMC
- OKMC Dynamic behavior can mimic in situ experiments

  (A. Kohnert, LANL, B. Wirth, UTK)

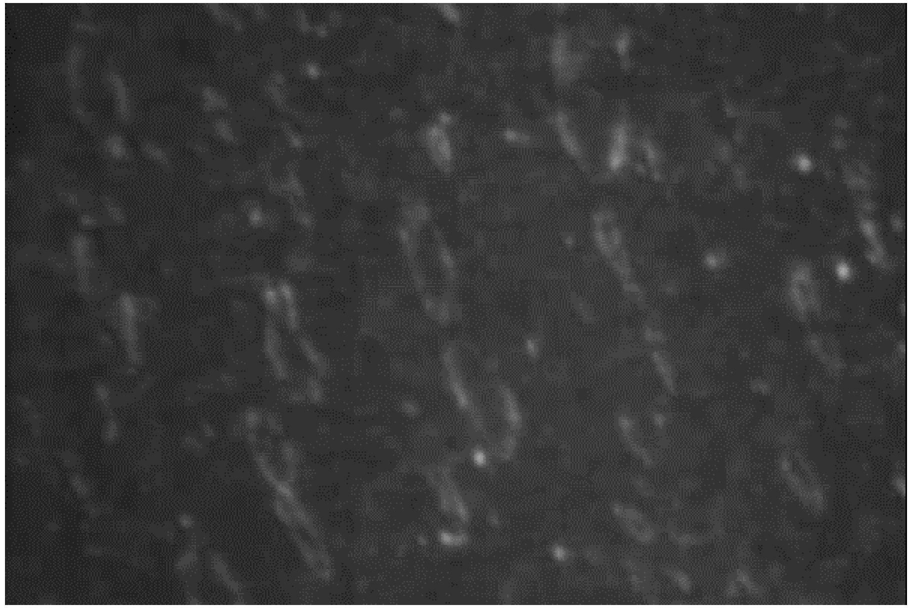




Loop growth kinetics and mechanisms

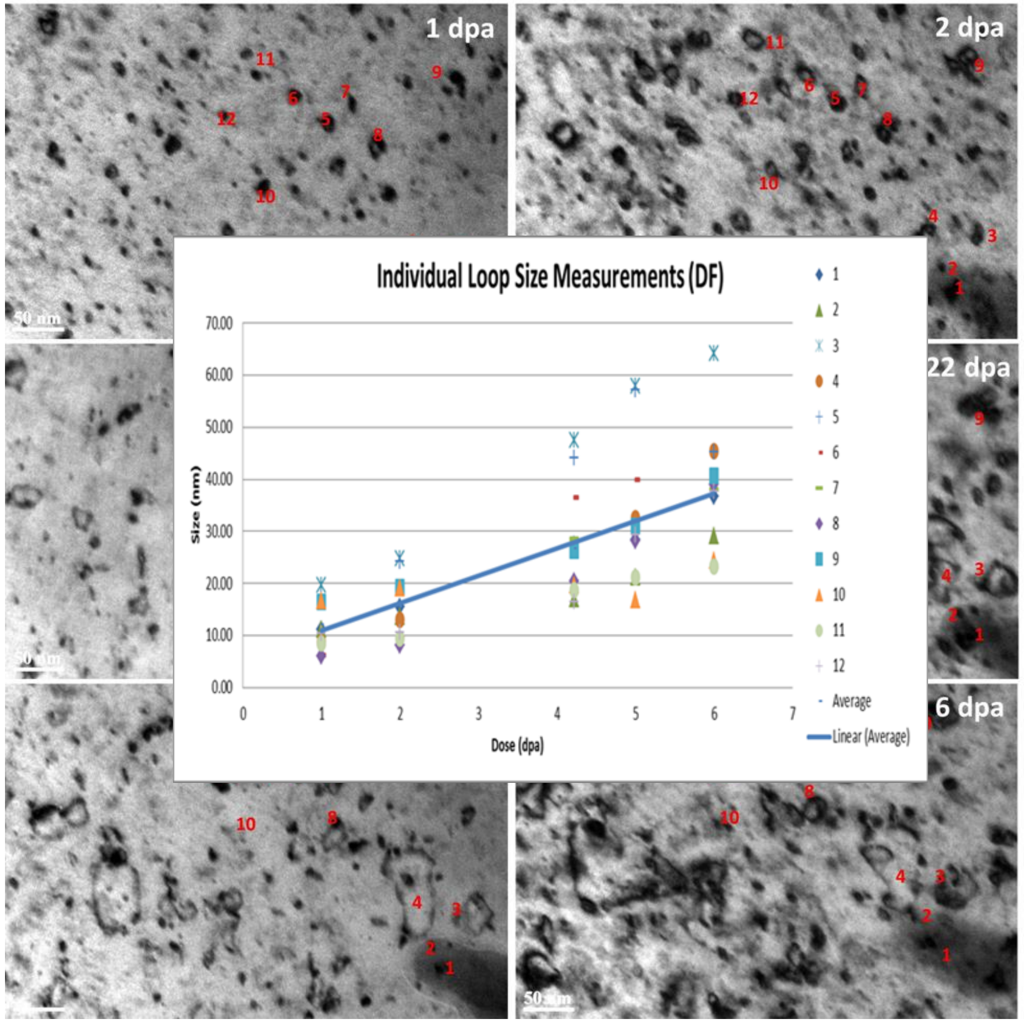
Loop motion and growth by coalescence is observed In Ultra-high purity Fe

high doses, 300C



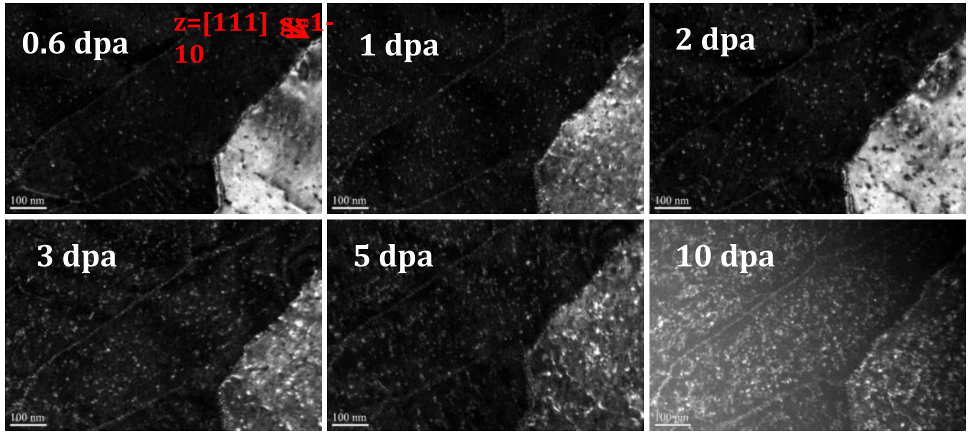
Jenkins, oxford

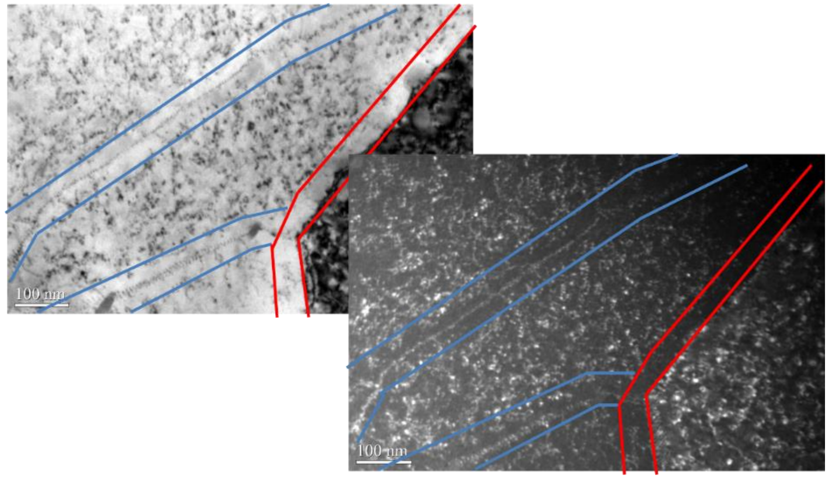
12Cr-model alloy, 200°C, 1 MeV Kr ions



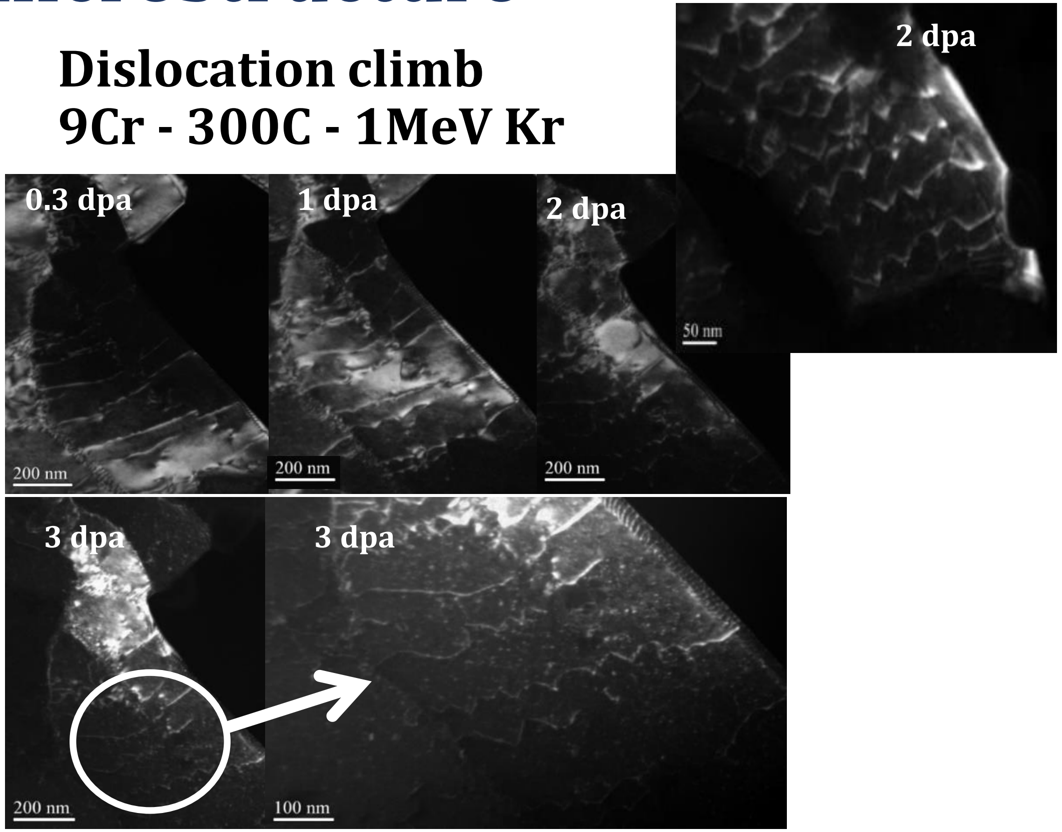
Illustrations of interactions between irradiation induced defects and microstructure

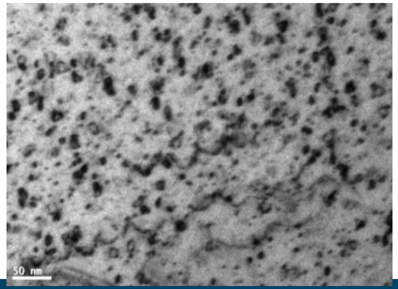
Denuded zones near grain-boundaries:
9Cr - 200C - 1MeV Kr





Dislocation climb
9Cr - 300C - 1MeV Kr





Helicoidal configuration

12Cr model alloy, 200°C,
1 MeV Kr ions, 1.6 dpa

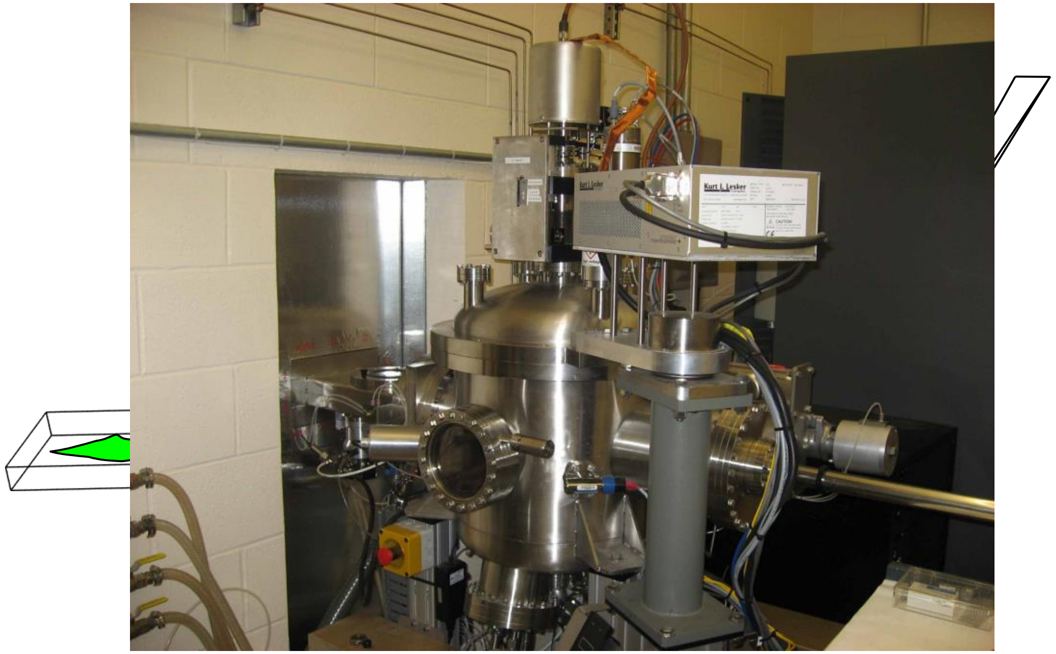
Examples of IN-SITU ION IRRADIATION in a TEM

- *Example 1:* Use of **in-situ irradiation** to study radiation damage in structural alloys

- *Example 2:* Use of **in-situ irradiation** to study and model irradiation induced grain growth in nanocrystalline metals

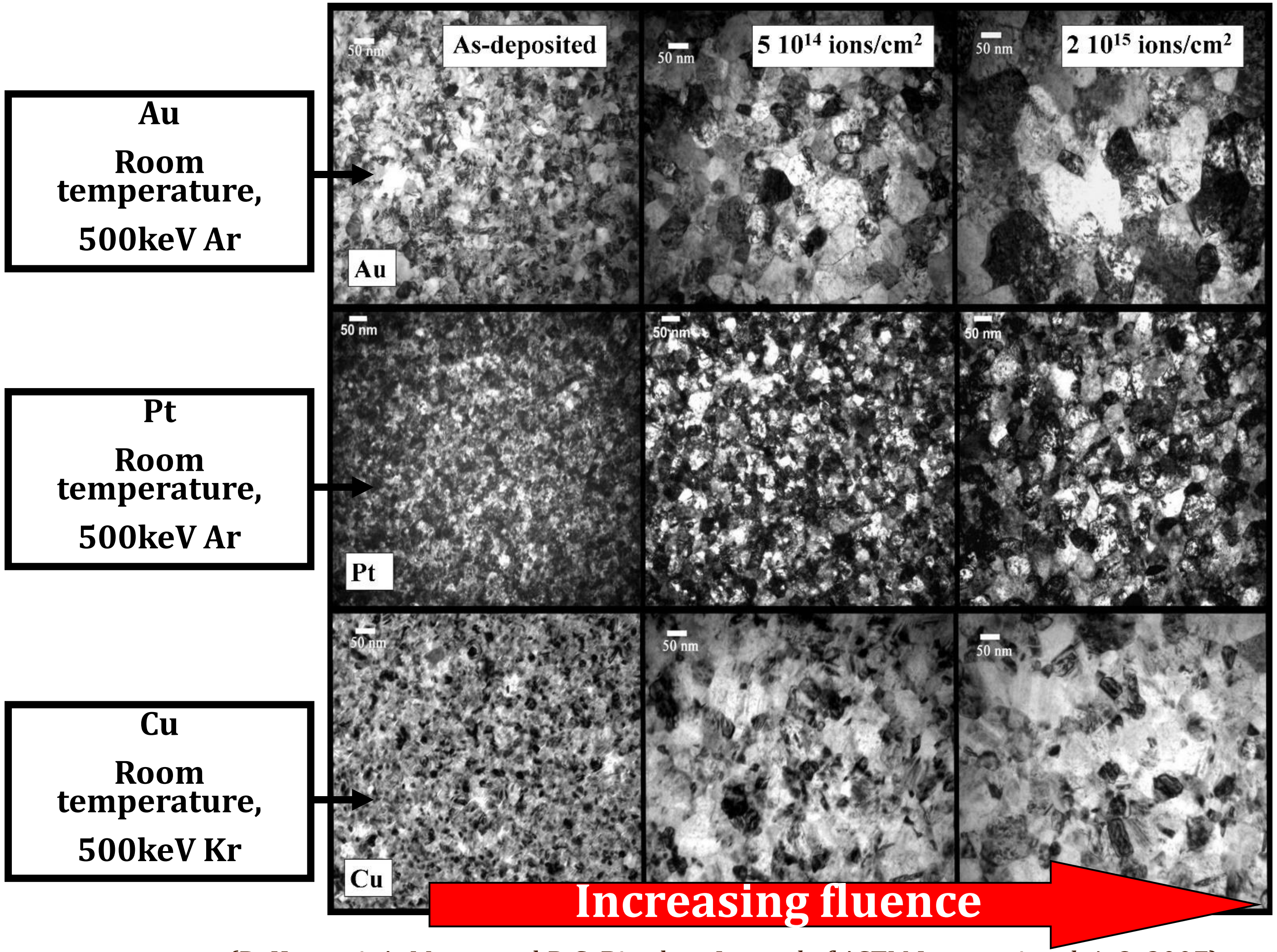
Sample preparation

Free standing thin-films of Au, Pt, Cu, Zr, Zr-Fe were prepared by sputter-deposition of a 80-90 nm layer onto NaCl substrates. Initial grain size ~10-15 nms.



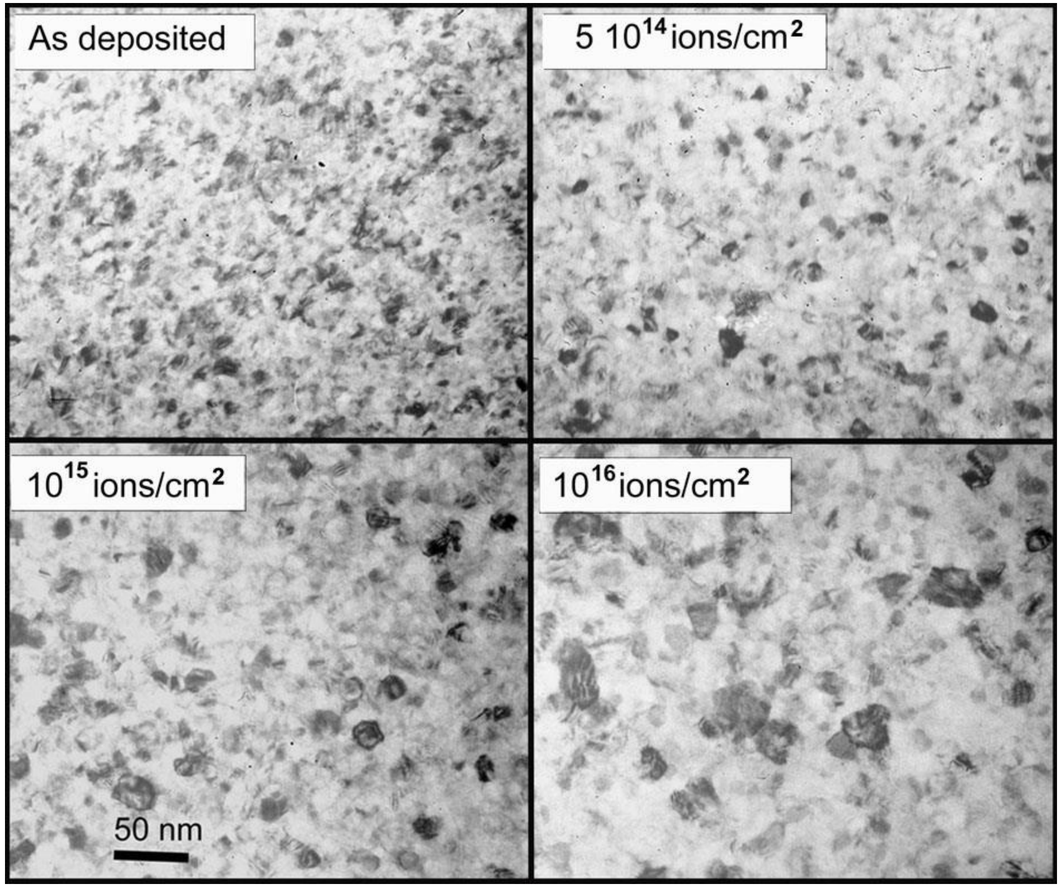
The samples were then floated on a liquid solution (80% de-ionized water and 20% ethanol) onto TEM copper grids, cleaned, and dried.

Grain Growth Under Ion irradiation



(D. Kaoumi, A. Motta, and R.C. Birtcher, *Journal of ASTM International*, 4, 8, 2007)

Ion-Beam Induced Grain Growth at 20K

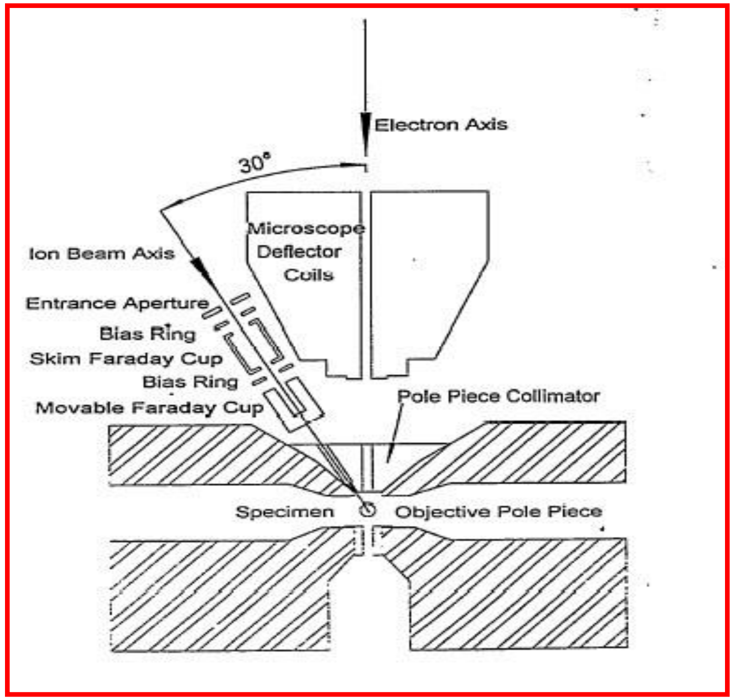


=> Is ion beam induced grain growth happening due to ion beam heating?

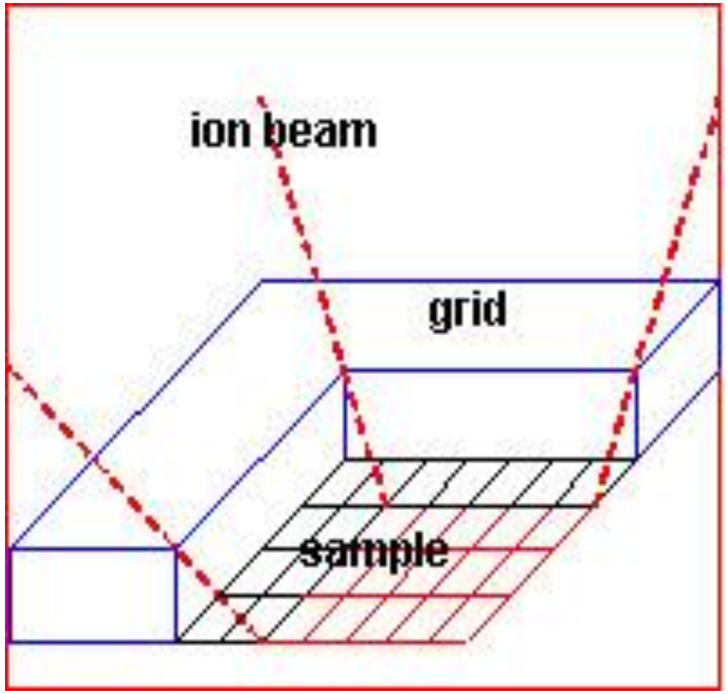
Pure Zr thin-film irradiated with 500 keV Kr ions at 20K.

Is ion-beam induced grain growth due to ion beam heating?

Geometry of the system



IVEM configuration



Sample configuration: irradiated sample area (red) and unirradiated area (black)

Shadowing of the sample by the grid creates a region not exposed to the beam which allows to separate the effects of irradiation and temperature.

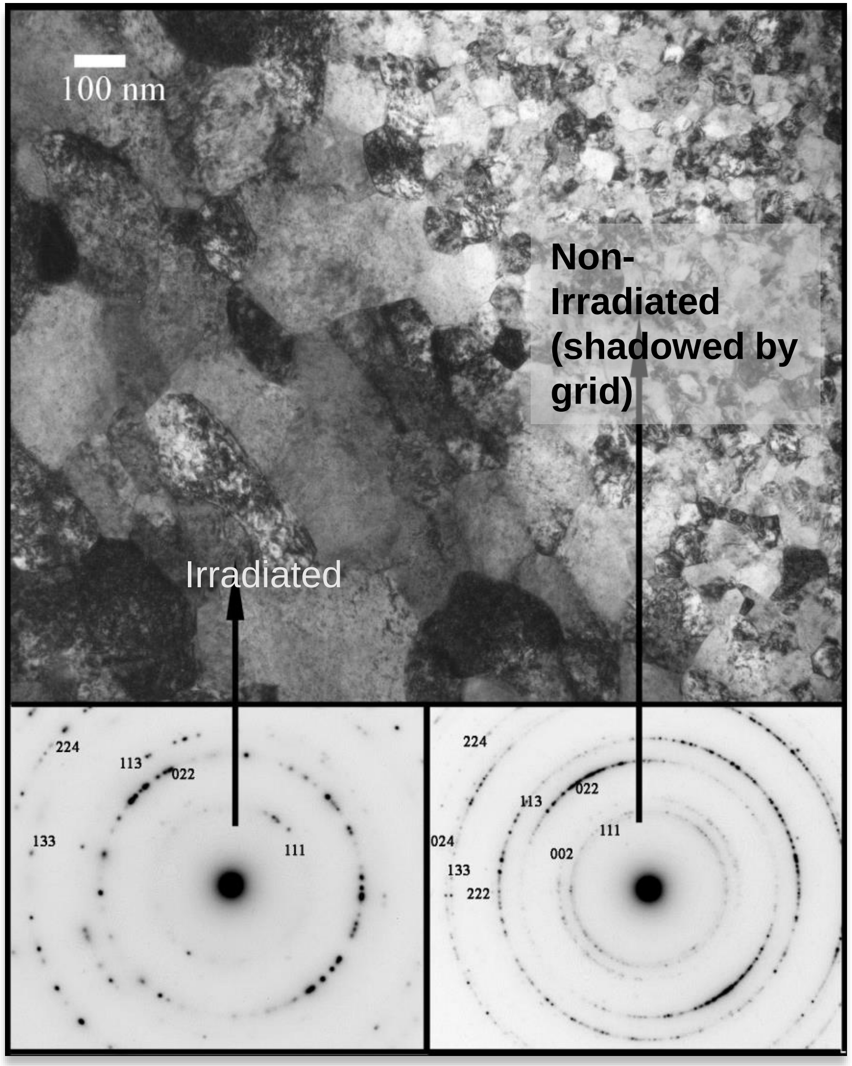
Ion-beam induced grain growth is NOT due to ion beam heating

Au film irradiated with 500keV Ar ions at room temperature to a fluence of 10^{16} ions/cm^2.

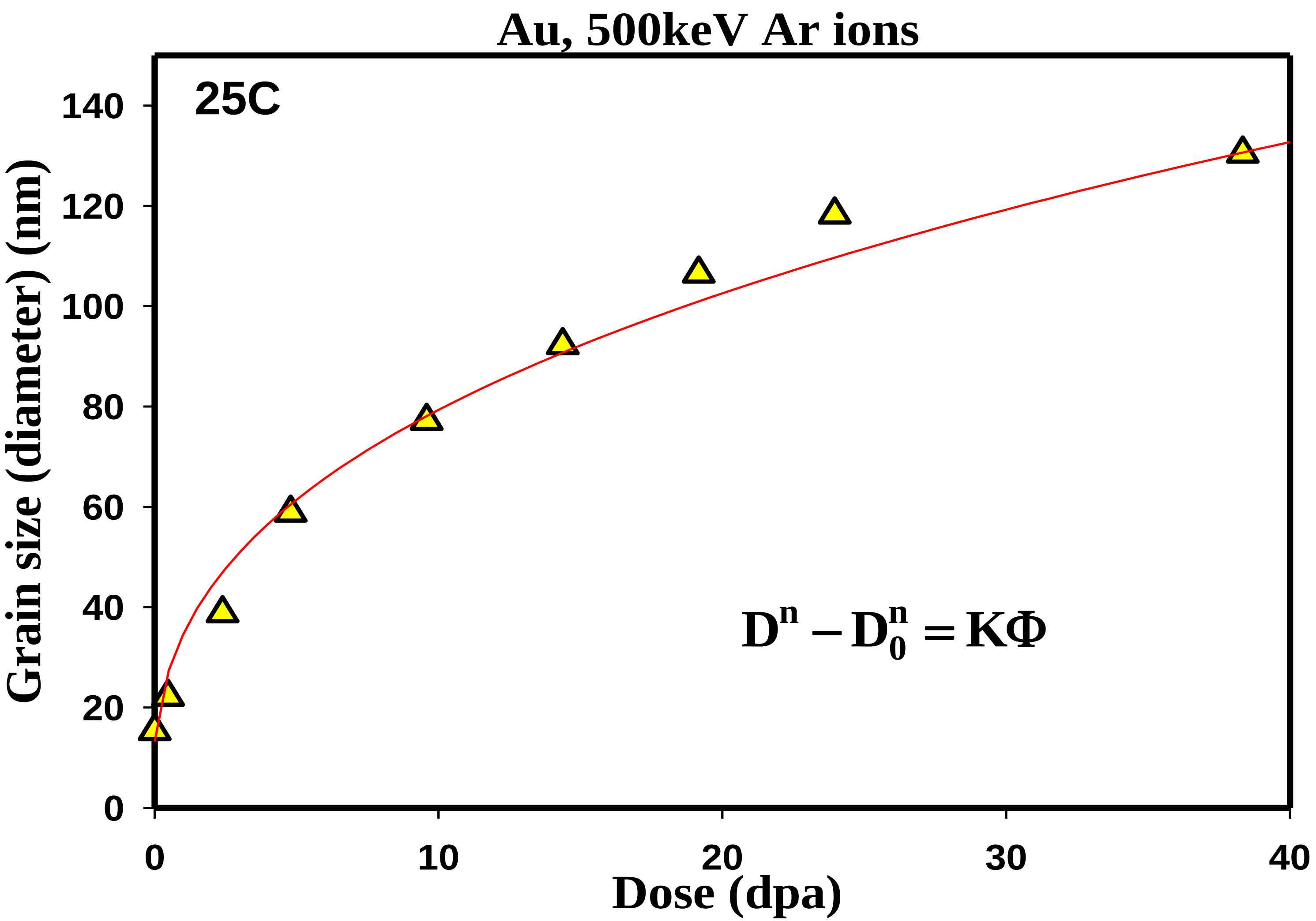
Top: bright field picture of the irradiatied/non-irradiated interface;

Bottom: diffraction patterns associated with the irradiated (left) and non-irradiated (right) areas respectively.

=> Ion-beam induced grain growth is NOT due to ion beam heating



Irradiation Induced grain growth kinetics



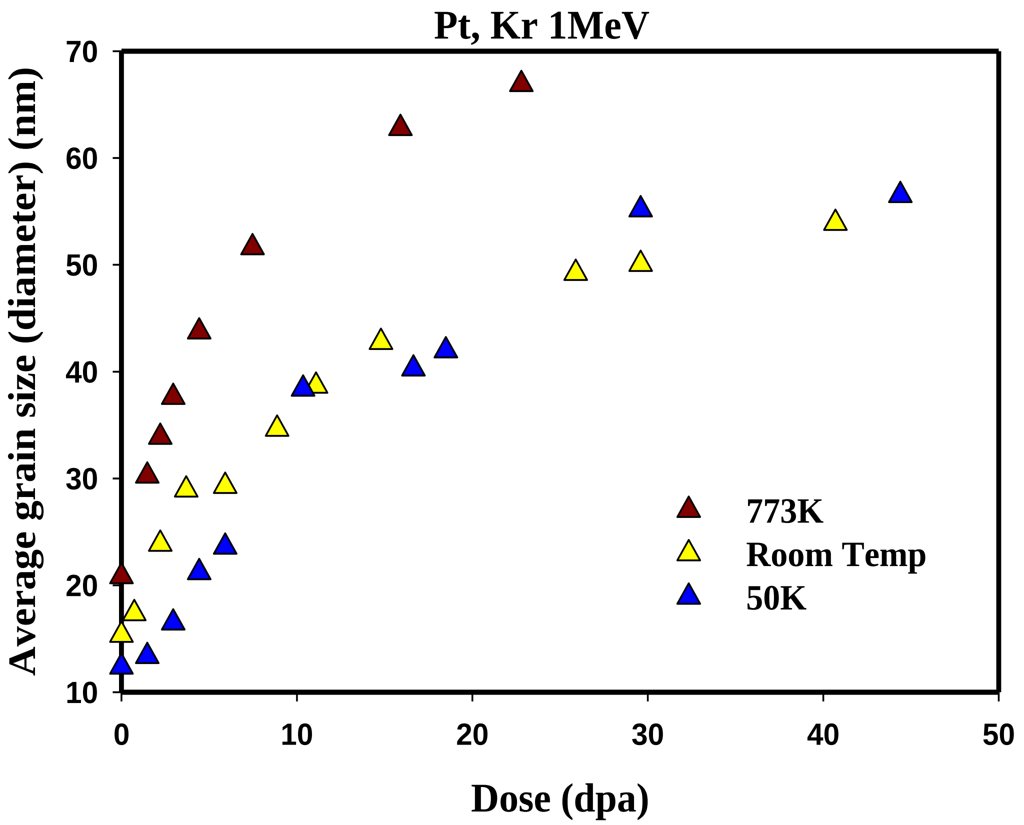
Experimental fitting parameters

The measured grain size D and ion dose Φ were fit to $\mathbf{D^n - D_0^n = K\Phi}$ **in the dose range from 0 to 50 dpa.**

Element	Ion type / Energy	Temperature (K)	Fit parameters			
			D_0 (nm)	n	K (nm^n/dpa)	Coefficient of determination R^2
Zr	Kr/500keV	20	14.17	2.74	0.41×10^3	99.7%
Au	Ar/500keV	50	9.24	2.91	16.6×10^3	91.3%
		298	13.2	2.68	12.2×10^3	97.3%
		473	25.53	3.00	$1.86\ 10^5$	91.3%
Cu	Kr/500keV	50	12.30	3.30	2.91×10^3	99.1%
		298	13.6	3.17	4.15×10^3	99.6%
		573	73.69	3.51	$1.53\ 10^{10}$	98.0%
Pt	Kr/1MeV	50	12.36	3.12	7.36×10^3	97.5%
	Kr/1MeV	298	14.5	3.27	12.4×10^3	99.0%
	Ar/500 keV	298	12.0	2.97	4.15×10^3	99.0%
	Kr/1MeV	773	20.22	3.25	$4.10\ 10^4$	99.1%

Experimental values of n are close to the average value of 3.

Effect of irradiation temperature



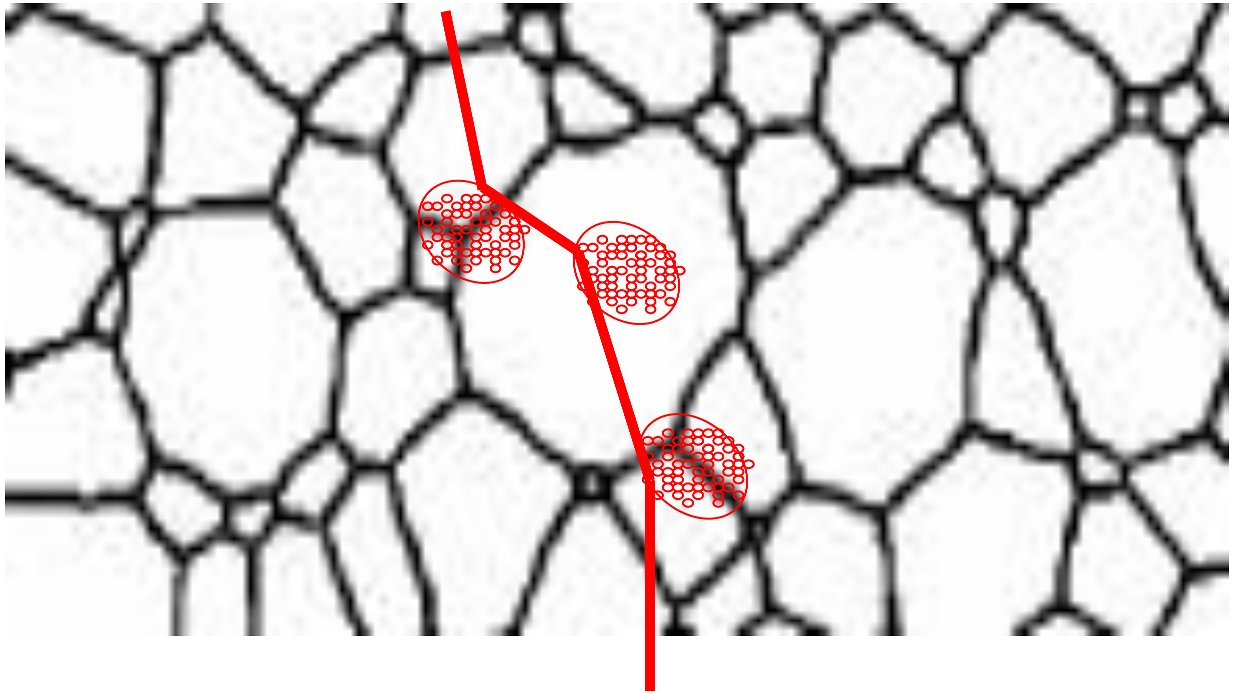
Similarly to Ion-Beam Mixing and several other irradiation effects, there are 2 regimes with respect to the ambiant temperature:

-> Low-temperature regime: grain-growth kinetics are independent of the substrate temperature: irradiation effects control the process of grain boundary migration.

-> Thermally-assisted regime: rate of grain-growth and final size increase with temperature: grain boundary migration is the result of the combined effects of irradiation and substrate temperature.

Transition temperature between the two regimes is: [0.14-0.22]T_{melt}.

Proposed mechanism for grain-growth under irradiation in the low temperature regime



Proposed mechanism:

- Ion creates primary knock-on atoms (PKAs) along its path. Some have enough energy to develop into displacement cascades and result in thermal spikes; some do not.
- Only the spikes/cascades occurring at grain-boundaries contribute to grain-growth: need to take into account the probability of spike/cascade occurring at a grain-boundary, and the size of average cascade.
- The atomic jumps within the cascade occurring at the grain boundary are biased by the local driving force

Derivation of the Irradiation induced grain-growth kinetics law based on the proposed mechanism

$$\boxed{\frac{d\overline{D}}{dt} \approx \dot{\Phi}\,\chi\, f_{GB}\,\eta\;V_{at}\delta}$$

$\rightarrow$

$$\boxed{\frac{d\overline{D}}{dt} = \dot{\Phi}\,\chi\,\delta\,V_{at}\,\frac{3d_{spike}}{\overline{D}}\left[\frac{4\gamma}{\overline{D}k_B}\,\frac{\sqrt{\frac{3}{5}}\,\Gamma\!\left(\frac{8}{3}\right)k_B^{8/3}}{10\pi\, c_0^{2/3}\kappa_0}\,\frac{Q^{5/3}}{E_a^{8/3}}\right]}$$

$$\boxed{\overline{D}^3 - \overline{D}_0^3 = \underbrace{\left[36\,\gamma\, d_{spike}\,\chi\,\delta\,\frac{V_{at}\,\nu\sqrt{\frac{3}{5}}\,\Gamma\!\left(\frac{8}{3}\right)k_B^{5/3}}{10\pi\, c_0^{2/3}\kappa_0}\,\frac{Q^{5/3}}{E_a^{8/3}}\right]}_{\mathbf{K}}\Phi}$$

(D. Kaoumi, A. Motta, R. Birtcher, *Journal of Applied Physics*, 104, 7, 2008).

Conclusions

Overall, we illustrate here how the use of accelerator coupled with TEM is an effective and invaluable tool for investigating irradiation induced microstructure evolution

The spatial resolution of the TEM makes it an invaluable tool to track the real-time response of microstructure under irradiation, shedding light on the mechanisms and kinetics of radiation damage formation and evolution in materials

The kinetics information captured through in-situ experiments inform modellers to improve predictive models

Thank you

Acknowledgements

US Department Of Energy Sponsor

Nuclear Science User Facility program which provides access to the IVEM facility at Argonne National Laboratory