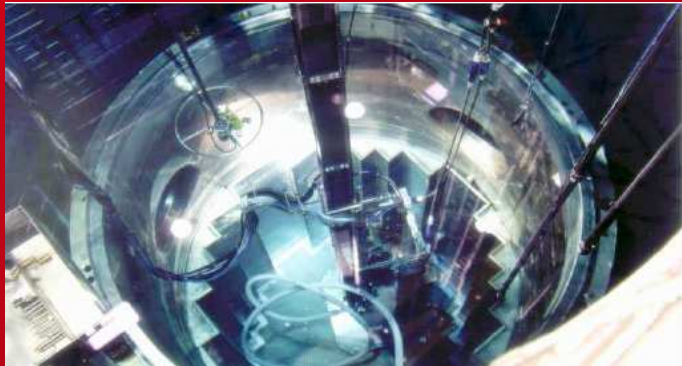


DE LA RECHERCHE À L'INDUSTRIE

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Irradiation embrittlement of austenitic stainless steels in PWR vessel's internals

-
Experiments and modelling from micro to mesoscale

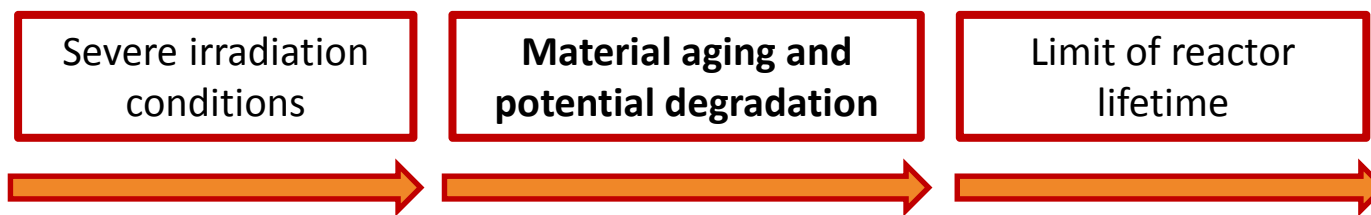
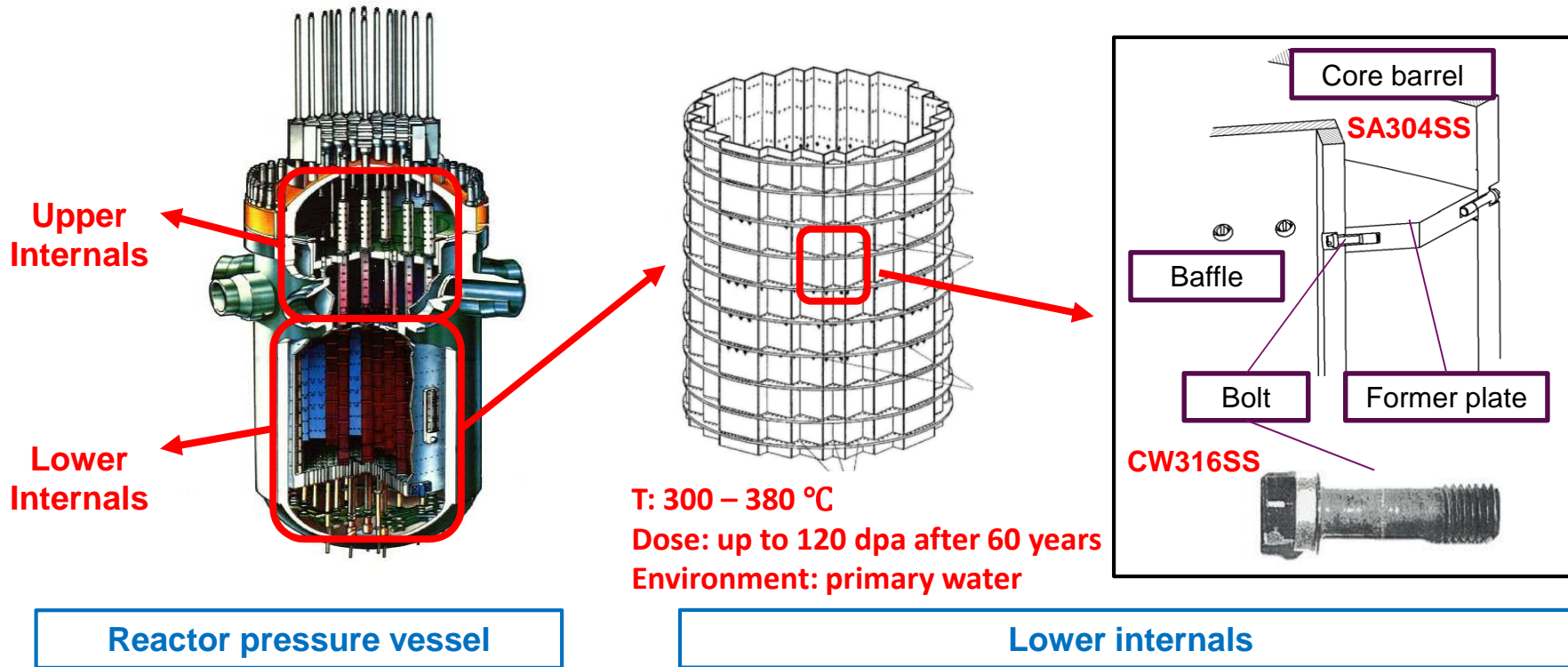
Benoit Tanguy, J. Hure, X. Han, C. Ling, P-O Barrioz,

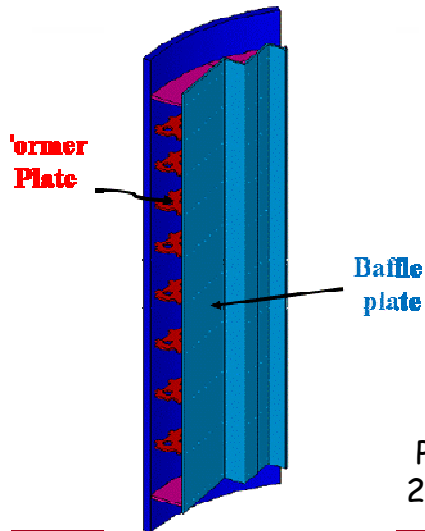
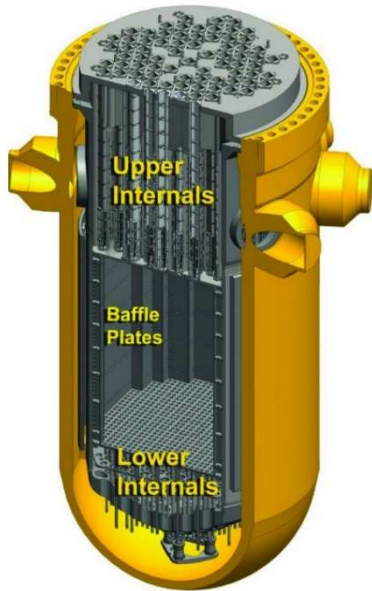
in collaboration with: J. Besson, S. Forest, Félix Latourte

Department of Materials for Nuclear Applications,
CEA, France



Materials resistant to extreme conditions for future energy systems, EC International Workshop, Kyiev, Ukraine, June 12-14 2017





➤ Ageing issues related to lower Internals

❑ Hardening, uniform elongation, and fracture toughness decreases

❑ Creep under irradiation

❑ Swelling (risk)

❑ Radiation induced segregation, precipitation

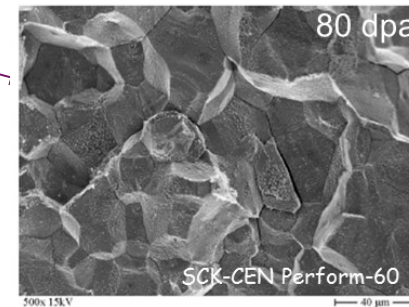
under irradiation

Environment

❑ Irradiation assisted stress corrosion cracking (IASCC)

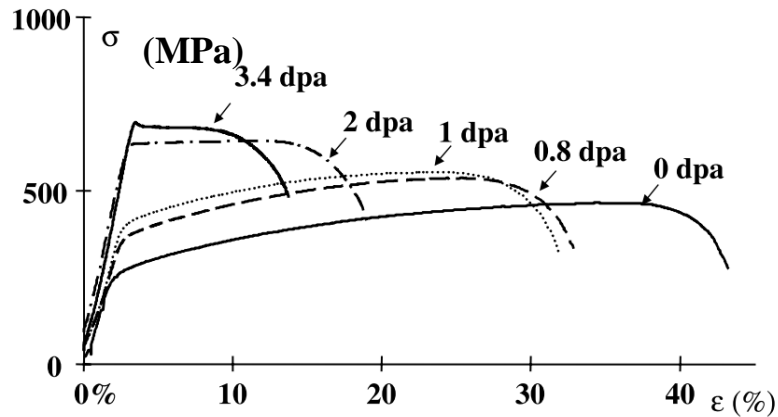
❑ Wear

PWR water 155 bars, 288°C, 30ccH₂/kgH₂O, 2 ppm Li, 1000ppm B, O₂ < 5 ppb, pH_{300°C} ≈ 7



Irradiation induced mechanical and microstructural modification

Macroscopic properties



[Pokor et al. (2004a)]

Dose dependent mechanical behavior:

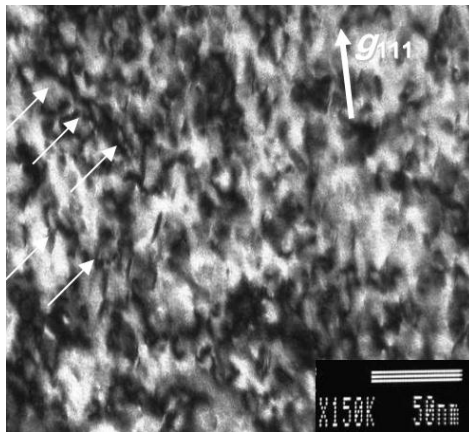
- ✓ Increasing yield strength
- ✓ Decreasing ductility
- ✓ Decreasing strain hardening capacity

In the literature:

- ✓ Single crystals: Patra and McDowell 2012 (BCC), etc.
- ✓ Polycrystals: Barton et al. 2013 (BCC) ...

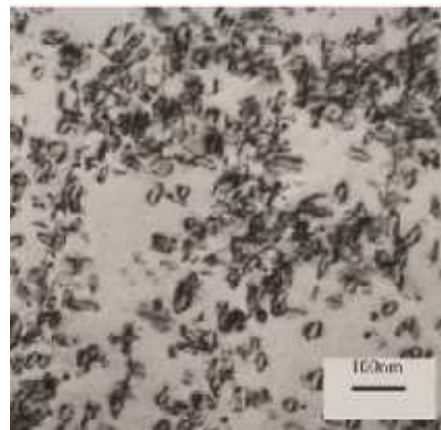
Physically based Modeling

Dislocations



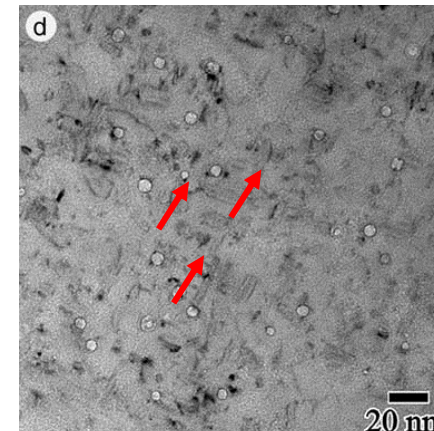
[Renault et al. (2010)]

Frank loops



[Garner et al. (2004)]

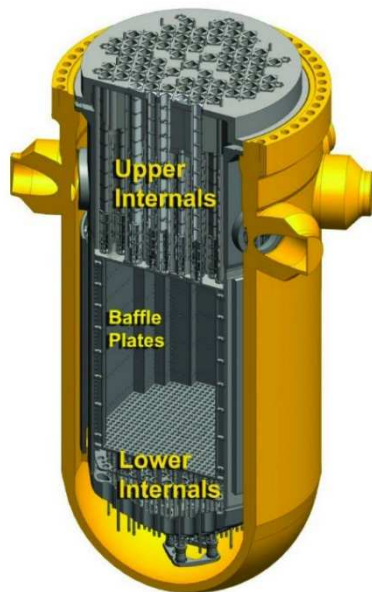
Cavities



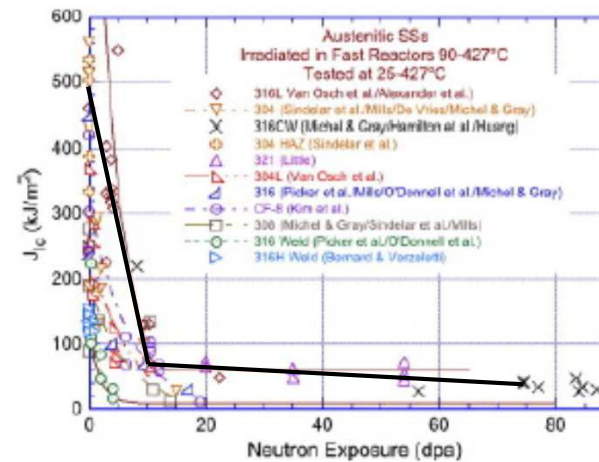
[Edwards et al. (2003)]

Microstructure

PWR's internals structures made of austenitic stainless steel (300 series)



Fracture toughness

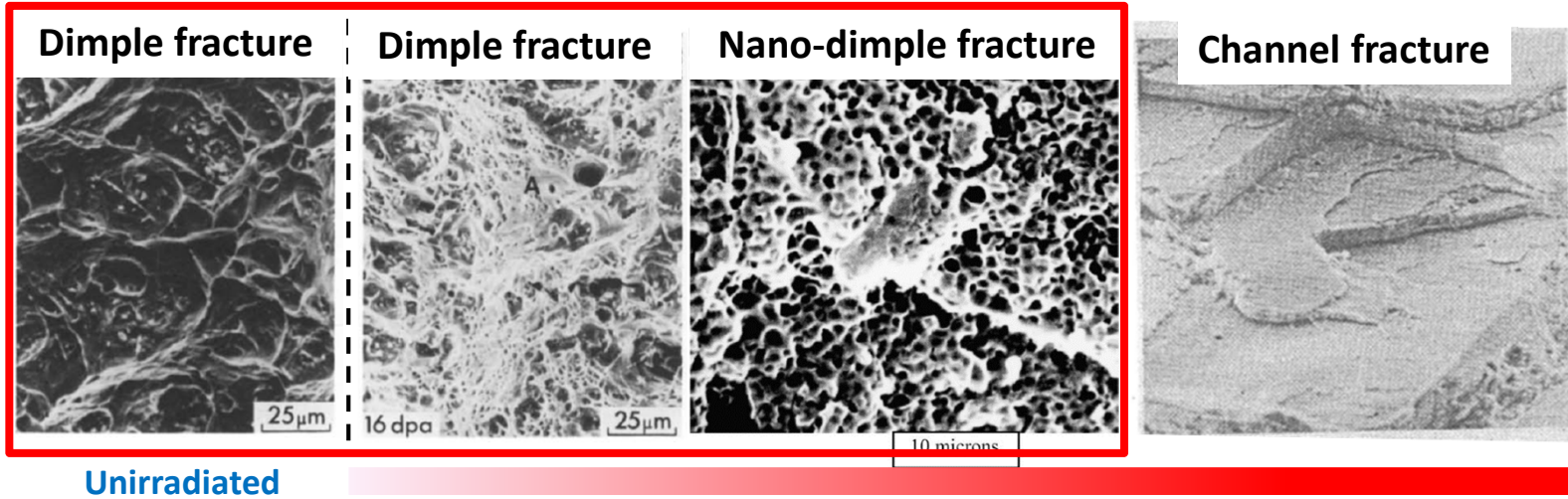


[Chopra & Rao (2011)]

Strong decrease of fracture toughness with irradiation related (How?)
to evolution of mechanical properties (due to irradiation defects).

Physical mechanisms ? -> Fractographics observations

Fractography



[Little (1986), Neustroev and Garner (2009), Fish (1973)]

Dimple-type Transgranular Fracture

Chanelling Fracture

➤ At high doses

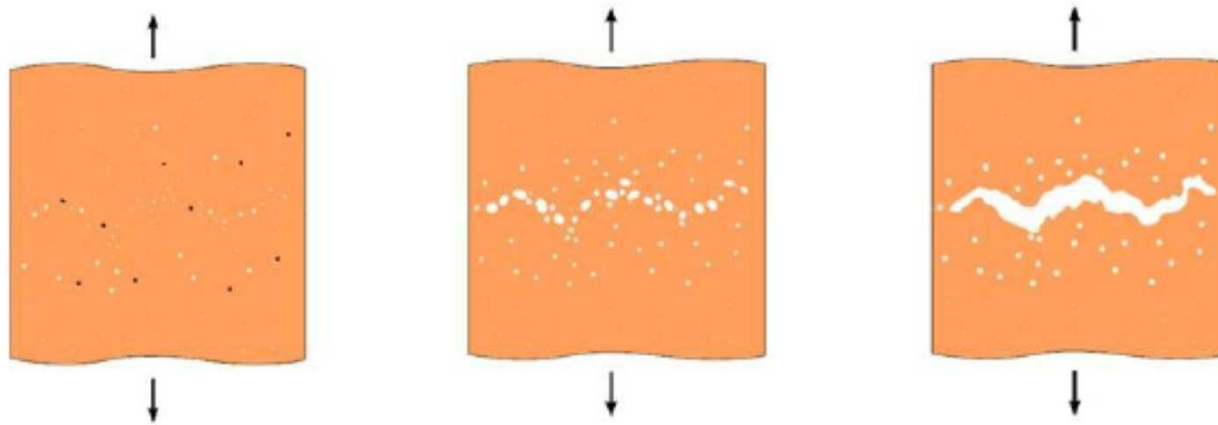
Some characteristics of the ductile fracture

- ❑ Intragranular voids
- ❑ Decreasing dimple size with irradiation
- ❑ Potential nano-dimple fracture at high irradiation levels

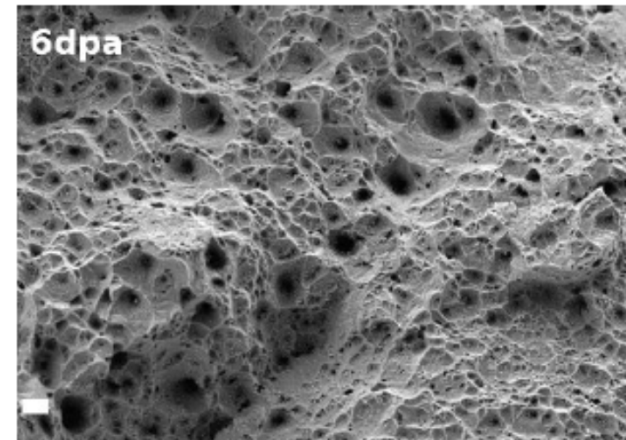
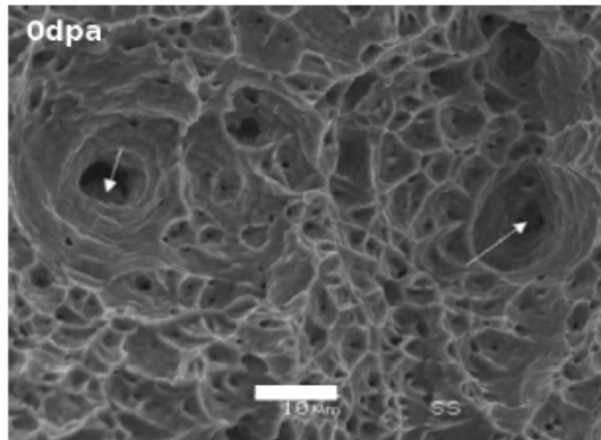
- Long-term objective:
 - ✓ Develop theoretical and numerical tools to predict the evolution of fracture toughness with irradiation

- Content of the present study:
 - ✓ Experimental study of ductile fracture in FCC steels
 - ✓ Modeling and simulation of ductile fracture of irradiated austenitic stainless steels involving intragranular voids
 - ✓ Understanding how fracture mechanisms influence fracture toughness by FE simulations at micro-scale

Dimple-type transgranular fracture : void growth and coalescence



- **Initiation:** Creation of voids in the material
- **Growth:** Enlargement of (non-interacting) voids
- **Coalescence:** Linkage of interacting adjacent voids



For low to medium irradiation doses

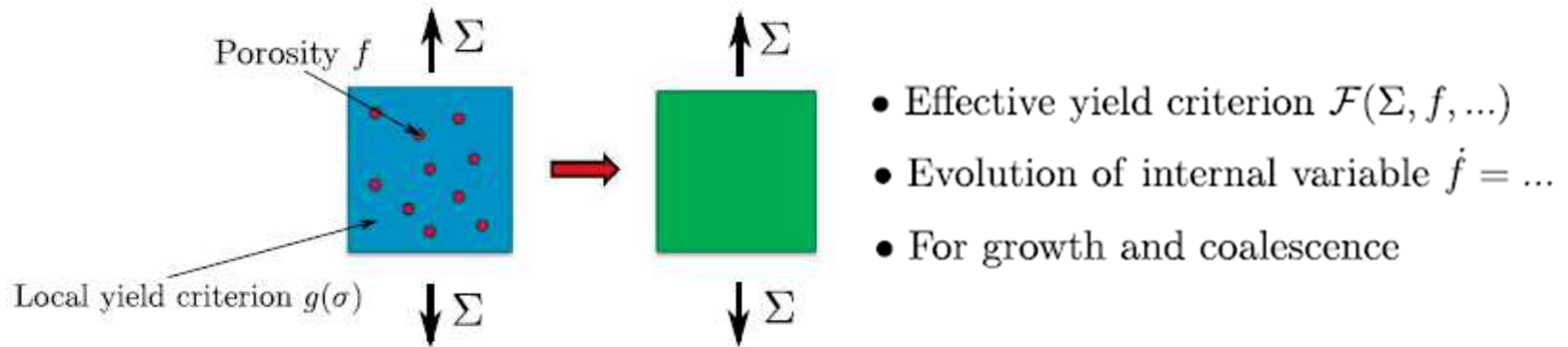
Ductile Fracture <-> Void growth and coalescence

Old research topic (since the 60's)-> **Ductile fracture modeling**

Pioneering modeling: McClintock (1968), Rice and Tracey (1969), Gurson (1977)

What may be different with irradiated stainless steel ? -> **Open questions**

From a porous material (of porosity f) to an effective (equivalent) material:

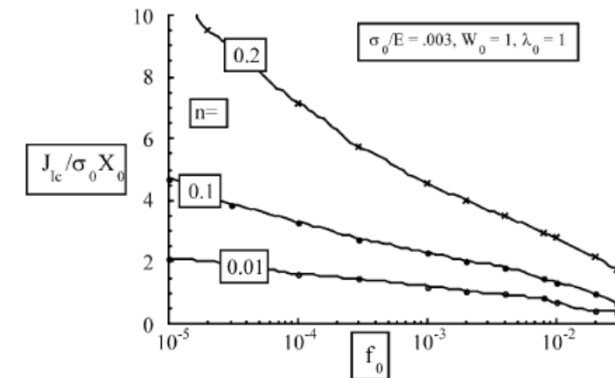


To obtain the **effective constitutive equations** requires:

- **Experimental data**
for void growth and coalescence
- **Theoretical approach:**
homogeneisation, limit analysis
- **Numerical simulations**

for different void lengthscales (μm , nm)

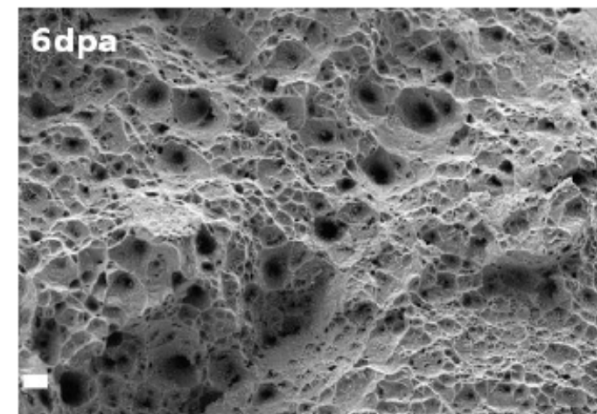
- Decrease of toughness with irradiation seems stronger than expected
 - ✓ Hardening $\rightarrow J_c \times (2-4)$ [$J_c \sim \alpha \sigma_y \times \lambda$]
 - ✓ Loss of strain hardening $\rightarrow J_c / (5-10)$



[Pardoen, Acta Mater. 2003]

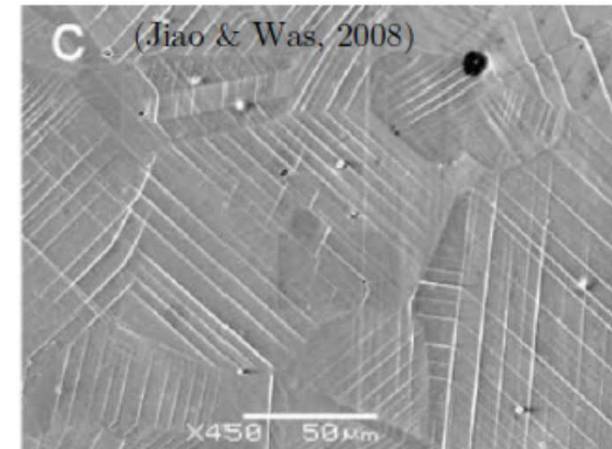
Physical mechanisms of voids growth in irradiated materials?

- Dimples (thus voids) are small !
 - ✓ Grain-scale modeling
 - ✓ Nano-voids \rightarrow size effects ?



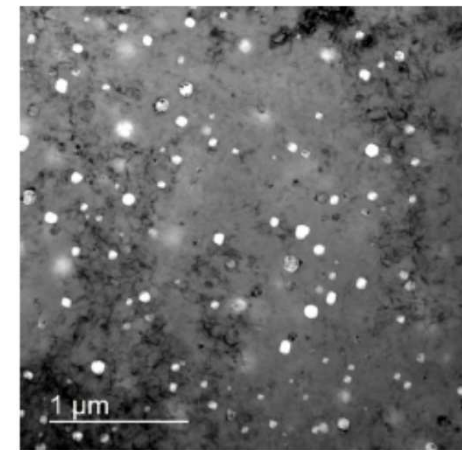
Effect of void size on physical mechanisms?

- Decrease of toughness with irradiation seems stronger than expected
 - ✓ Hardening $\rightarrow J_c \times (2-4)$ [$J_c \sim \alpha \sigma_y \times \lambda$]
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Physical mechanisms of voids growth in irradiated materials?

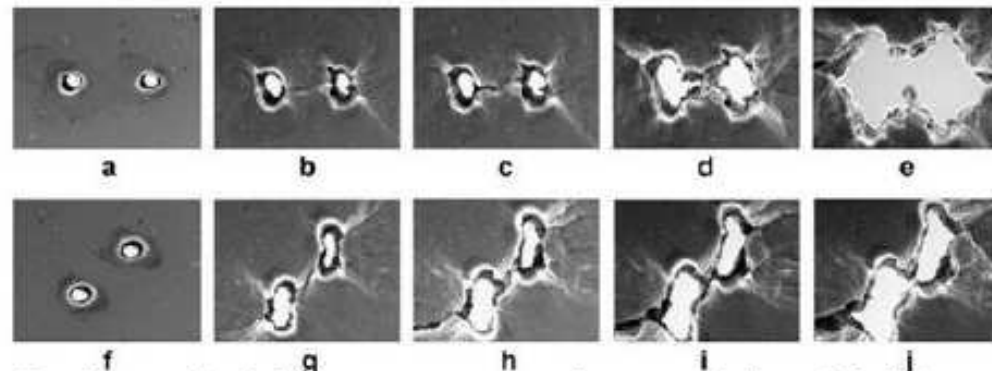
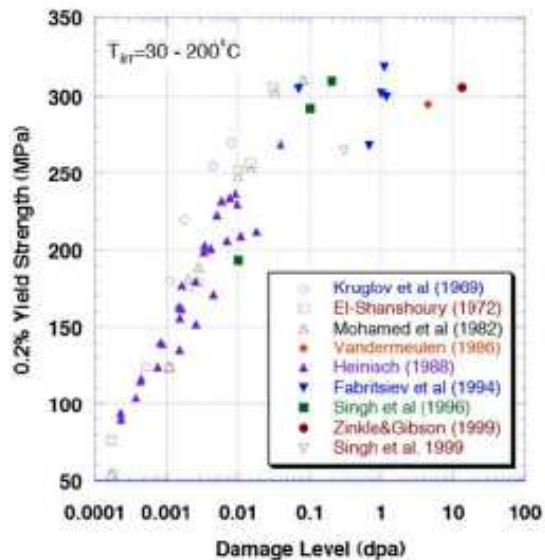
- Dimples (thus voids) are small !
 - ✓ Grain-scale modeling
 - ✓ Nano-voids \rightarrow size effects ?



Effect of void size on physical mechanisms?

Experimental methodology: Micro-void growth and coalescence

- **Irradiated material: polycrystalline pure copper**
 - ✓ Pure Cu → FCC, Significant hardening with low dose
 - ✓ Protons irradiation → no residual radioactivity
- **Model voids under uniaxial tension**
 - ✓ Focused-ion Beam (FIB) drilling of cylindrical holes...
 - ✓ ...in a tensile sample



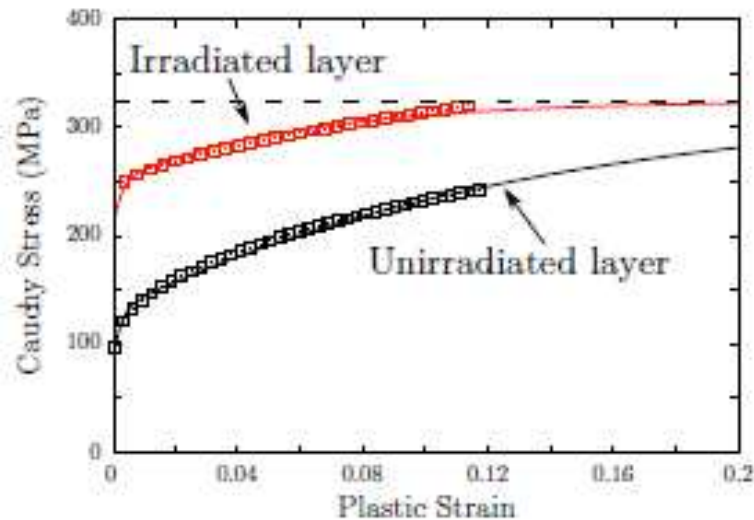
(Similar methodology on unirradiated material, from Weck & Wilkinson, 2008)

Proton-irradiation of pure Cu plate

- Thin plate: 75 μm thickness
- 2MeV H^+ , low temperature
- Irradiation depth 20 μm , 0.02 dpa (surface)



Ion beam at Jannus Saclay

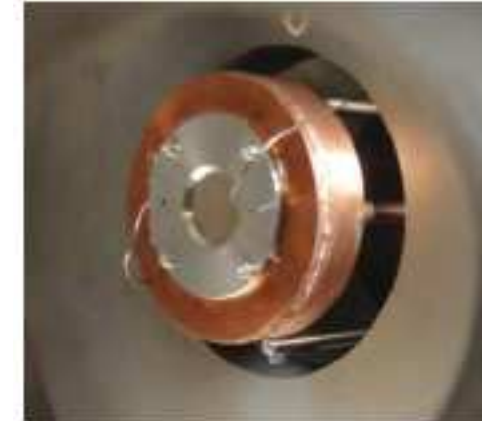


Mechanical properties after irradiation

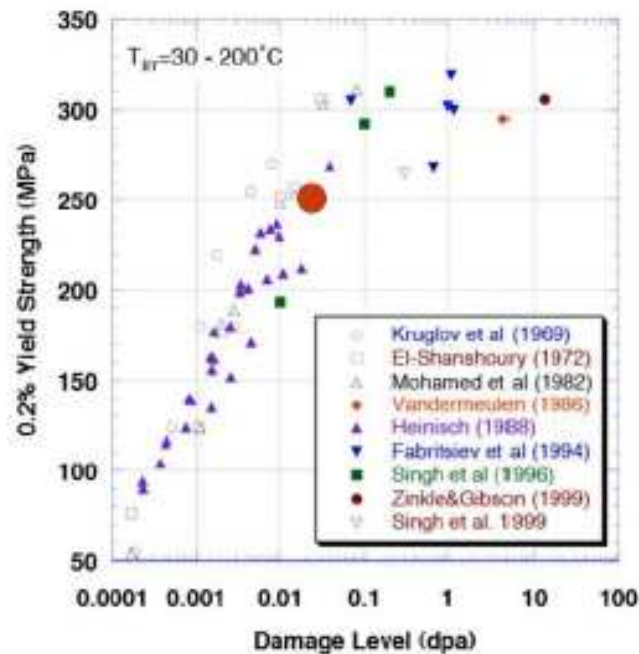
- Tensile test on partly-irradiated material
 - ✓ Stress-strain curve of irradiated layer
- $\Delta\sigma_{ys} = 130\text{MPa}$

Proton-irradiation of pure Cu plate

- Thin plate: 75 μm thickness
- 2MeV H^+ , low temperature
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Experimental set-up

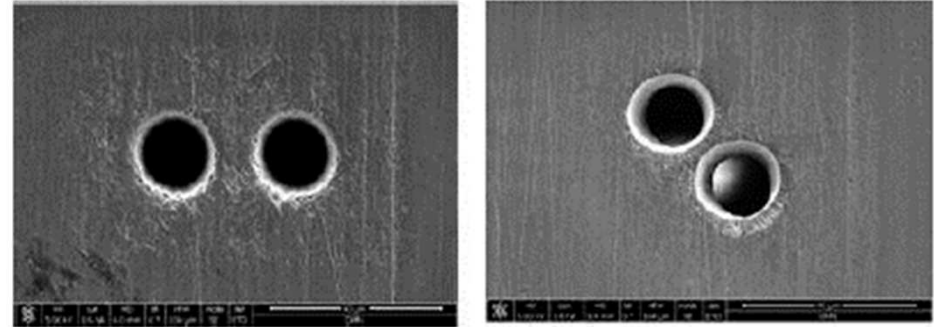


Mechanical properties after irradiation

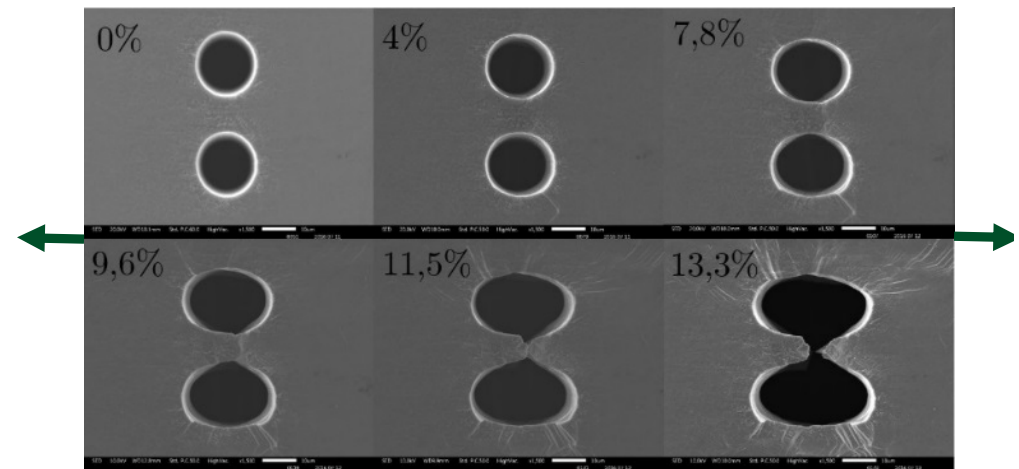
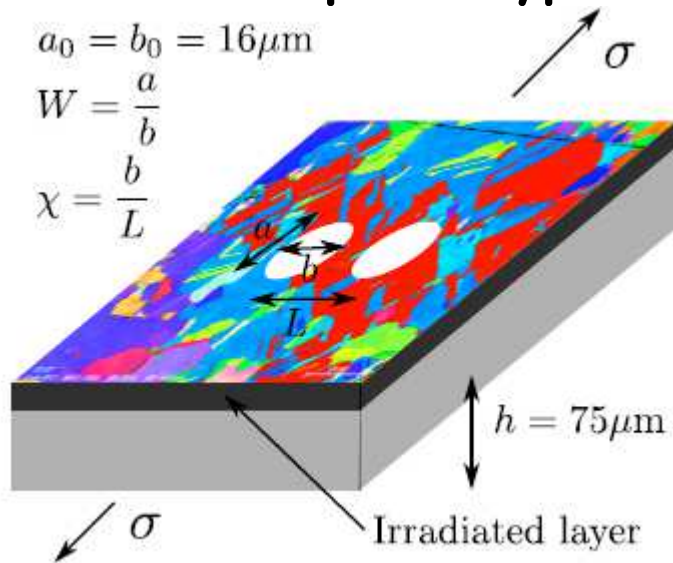
- Tensile test on partly-irradiated material
 - ✓ Stress-strain curve of irradiated layer
- $\Delta\sigma_{ys} = 130\text{MPa}$

Models voids

- FIB drilling of cylindrical holes
- 16 μm radius
- Two geometries
- ... through tensile samples

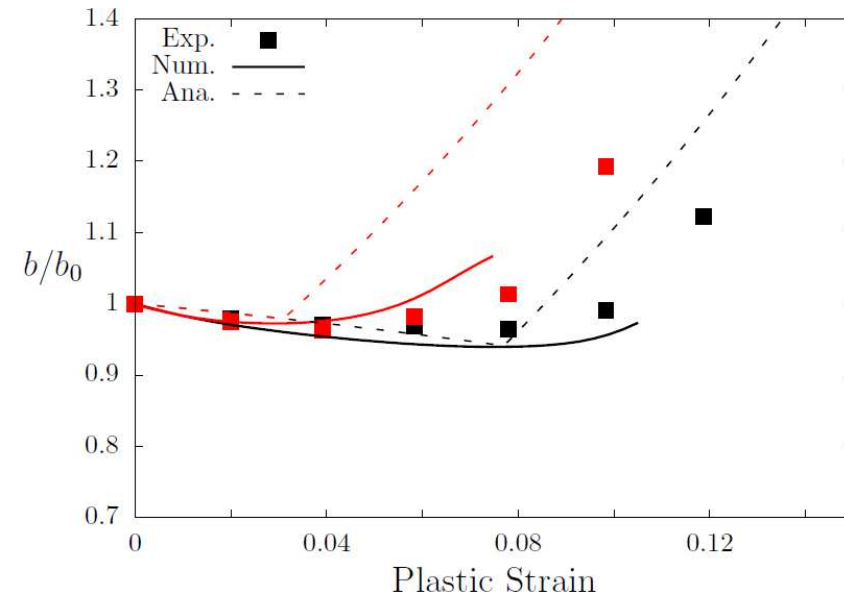
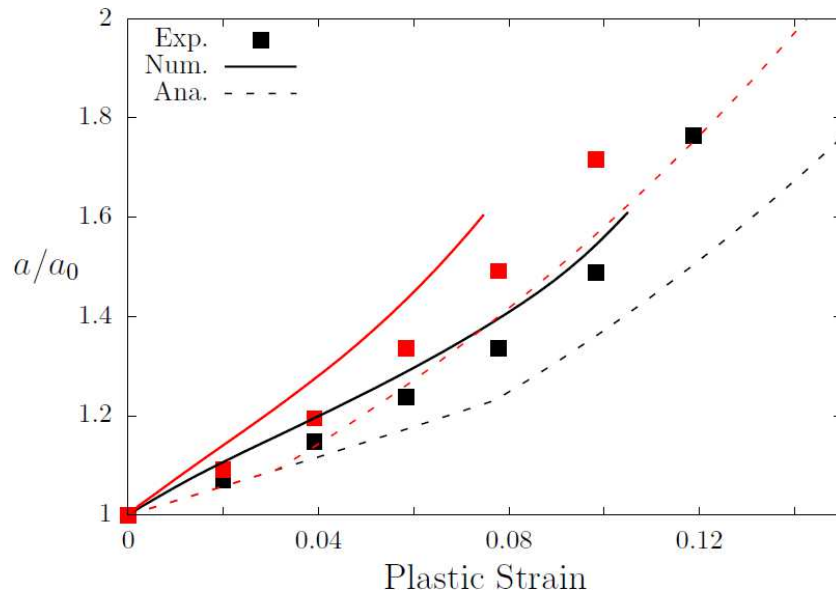


Experimental setup and typical observations



- SEM measurements of void dimensions with applied strain

Experimental results



- Voids in irradiated material grow faster (and coalesce earlier)
- Experimental data in good agreement with:
 - ✓ Finite-element simulations
 - ✓ Analytical model (McClintock growth model)
- That account only for hardening (and lower strain hardening)

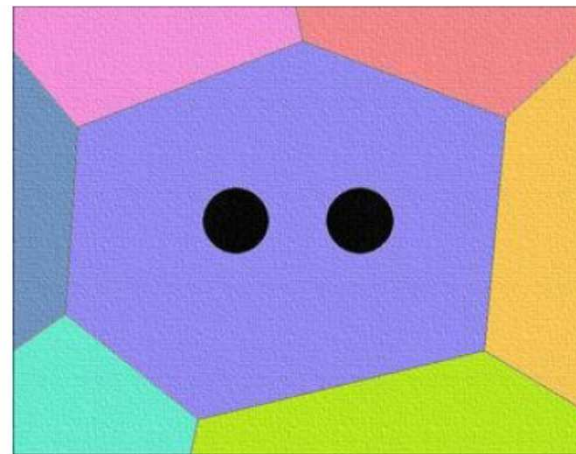
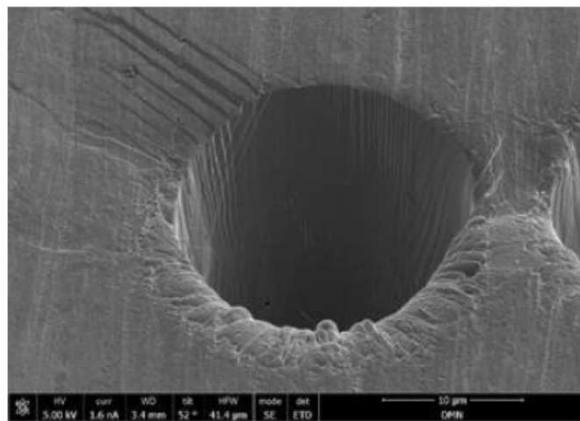
Experimental data on **micro-void** growth and coalescence indicates:

- Accelerated growth and coalescence on irradiated material...
- ...well captured accounting only for macroscopic hardening
 - ✓ No significant effect of strain localization (for low dose)

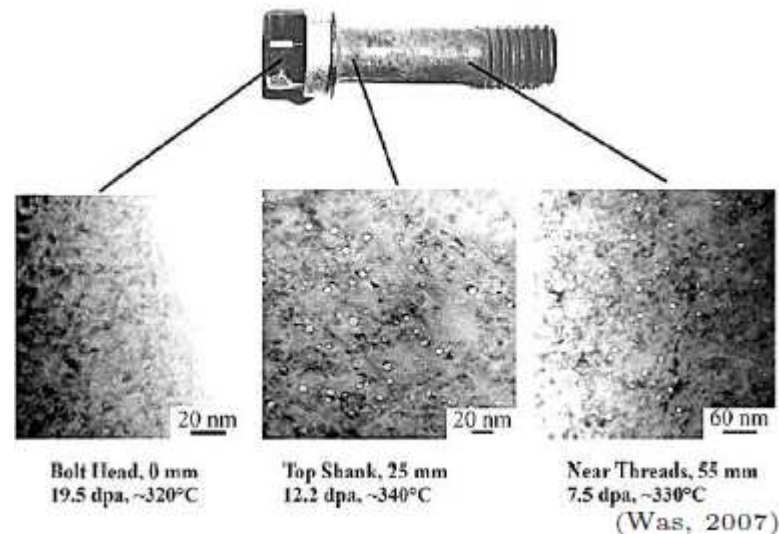
for voids size **larger** than the grain size

On-going study: voids size **lower than** the grain size

- 304L stainless steel
- Unirradiated and Proton irradiated



- Study of μm voids growth and coalescence in irradiated materials is relevant for ductile fracture modelling, but
- Nano-voids might also be present as irradiation defects:
 - ✓ e.g. in austenitic stainless steels PWR bolts

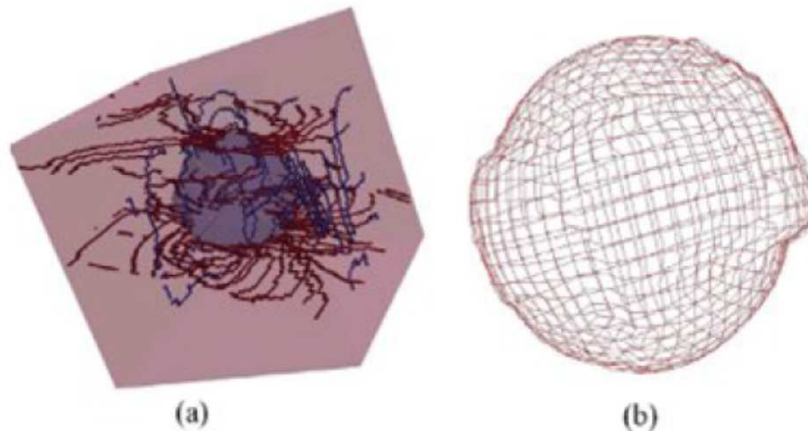


- What is the behavior of nanovoids under mechanical loading?
Small voids -> Hardening / Large voids -> Softening ?

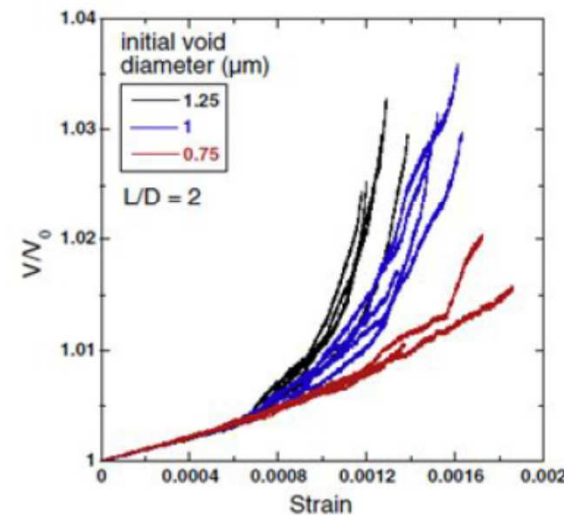
Why a nano-void might behave differently than larger voids ?

- Size effect predicted by MD and DD simulations ...
- ... due to intrinsic additional lengthscales:
 - ✓ Typical distance between dislocations: $\frac{1}{\sqrt{\rho}}$
 - ✓ Surface energy (void interface) *vs.* matrix yield stress: $\frac{\gamma}{\sigma_{ys}}$

If the size of the void $R \lesssim \left[\frac{1}{\sqrt{\rho}}, \frac{\gamma}{\sigma_{ys}} \right]$, size effects are expected

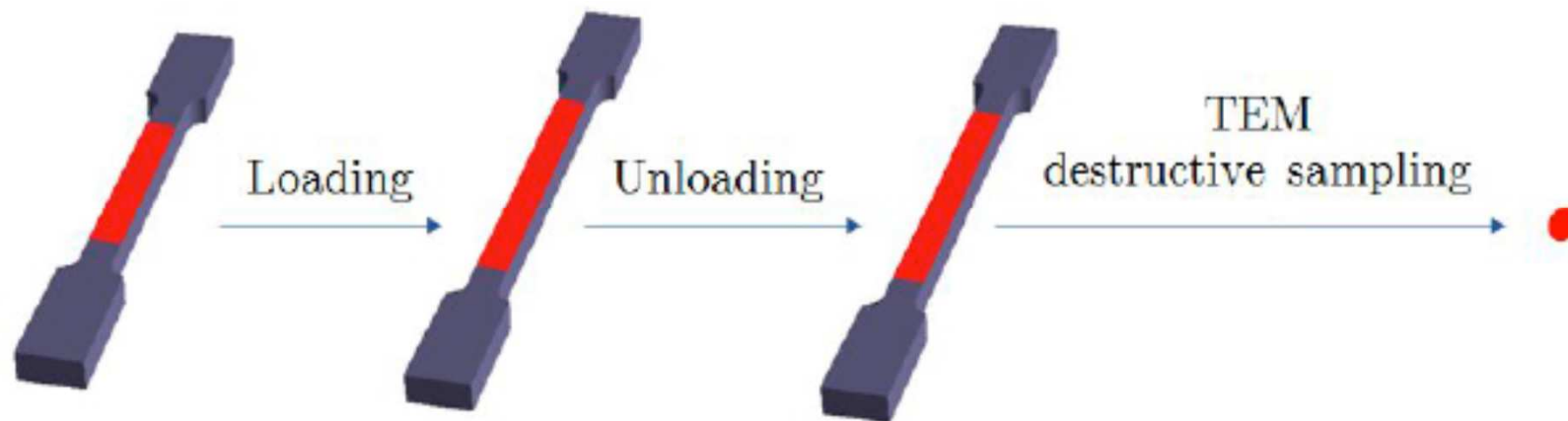


(Chang *et al.*, 2015)



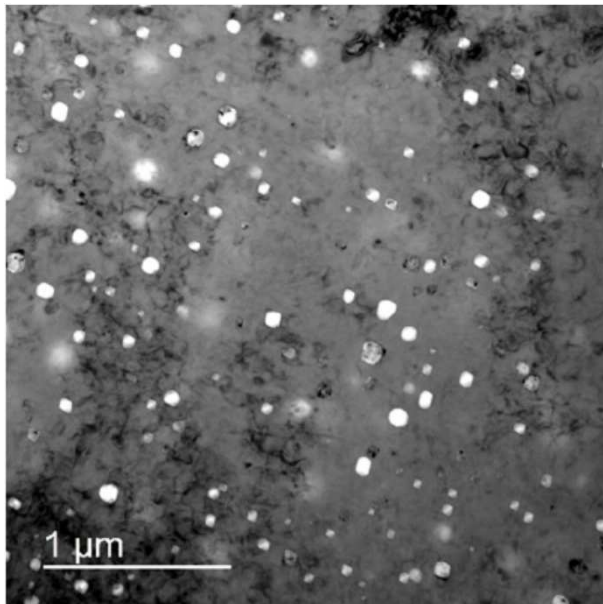
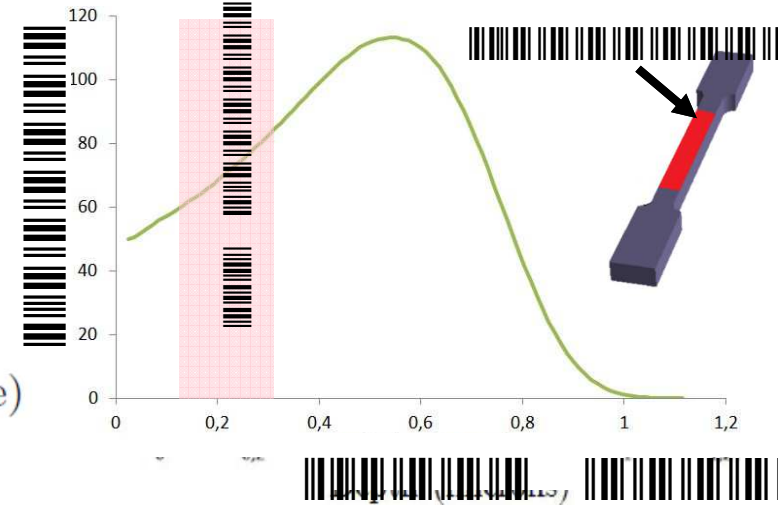
Experimental methodology: Nano-void growth (and coalescence)

- Irradiated material: SA 304L austenitic stainless steel
 - ✓ Fe irradiation → high dose → swelling: Model nanoporous materials
 - ✓ On tensile samples
 - ✓ Nano-voids characterization before mechanical loading
- Nano-voids under uniaxial tension
 - ✓ Tensile test on irradiated sample
 - ✓ 300°C, ~30% strain
 - ✓ Nano-voids characterization post-mechanical loading



Fe-irradiation of SA304L

- Thick sample: 2mm thickness
- Irradiation performed at JANNuS Saclay
 - ✓ 2MeV Fe²⁺, 600°C
 - ✓ **Irradiation depth** 1μm, 50dpa (surface)

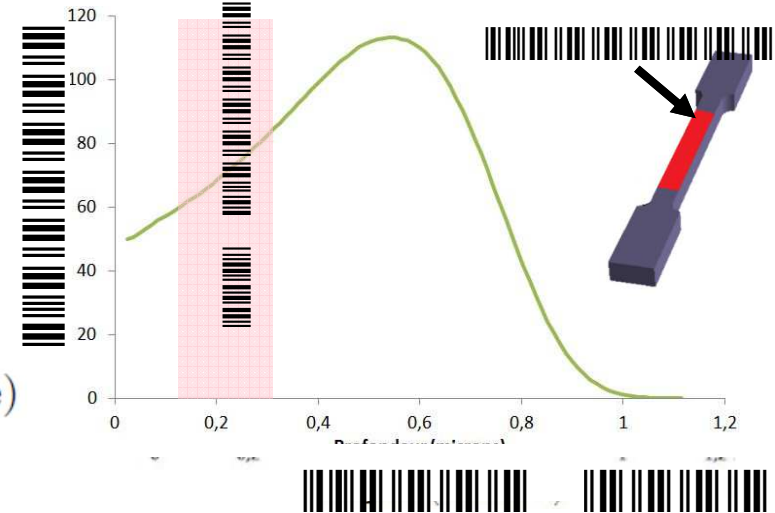


Microstructure after irradiation

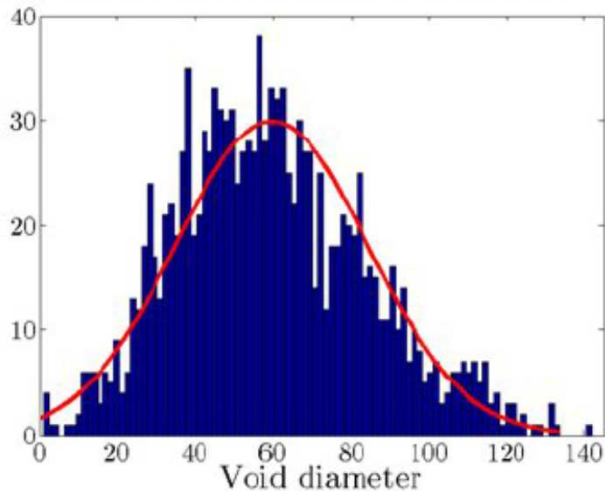
- Spherical nano-voids: 60nm ± 25nm
- Void density: 4%
- Dislocation density: $5 \cdot 10^{14} \cdot \text{m}^{-2}$

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Diameter distribution of the reference state



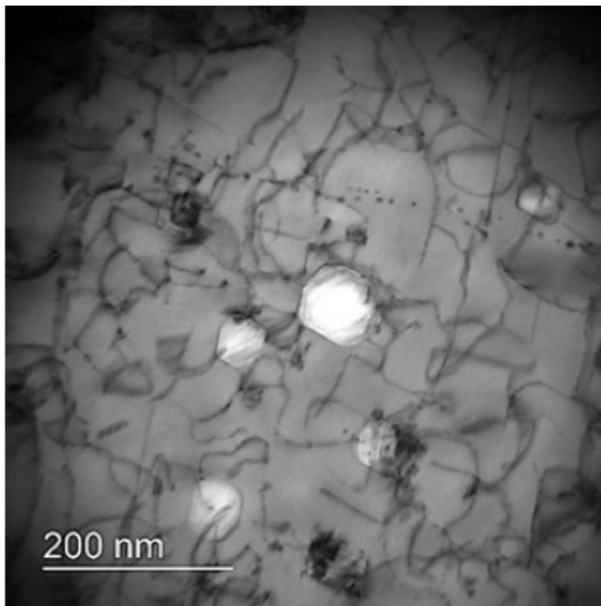
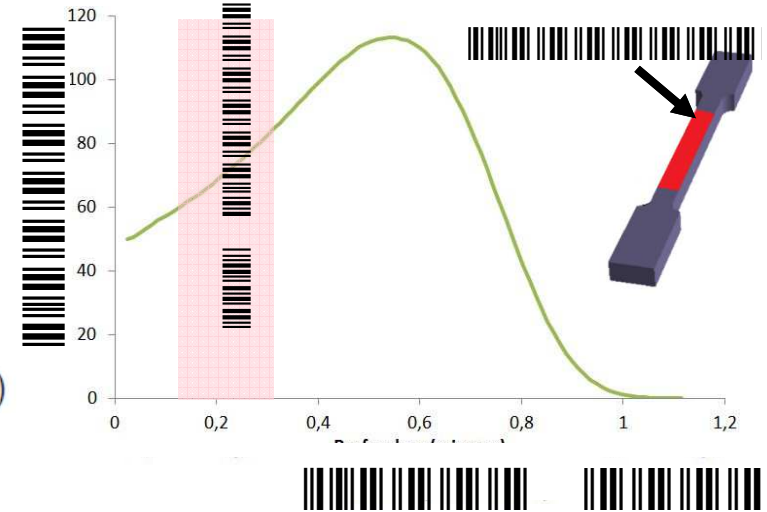
Microstructure after irradiation

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Nano-void growth and coalescence in irradiated materials

Fe-irradiation of SA304L

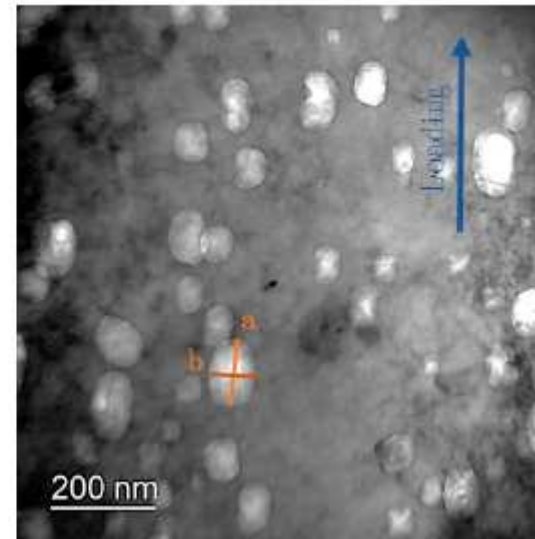
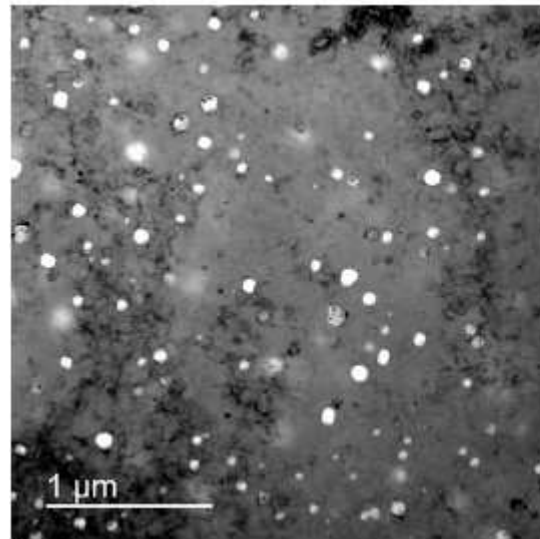
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Microstructure after irradiation

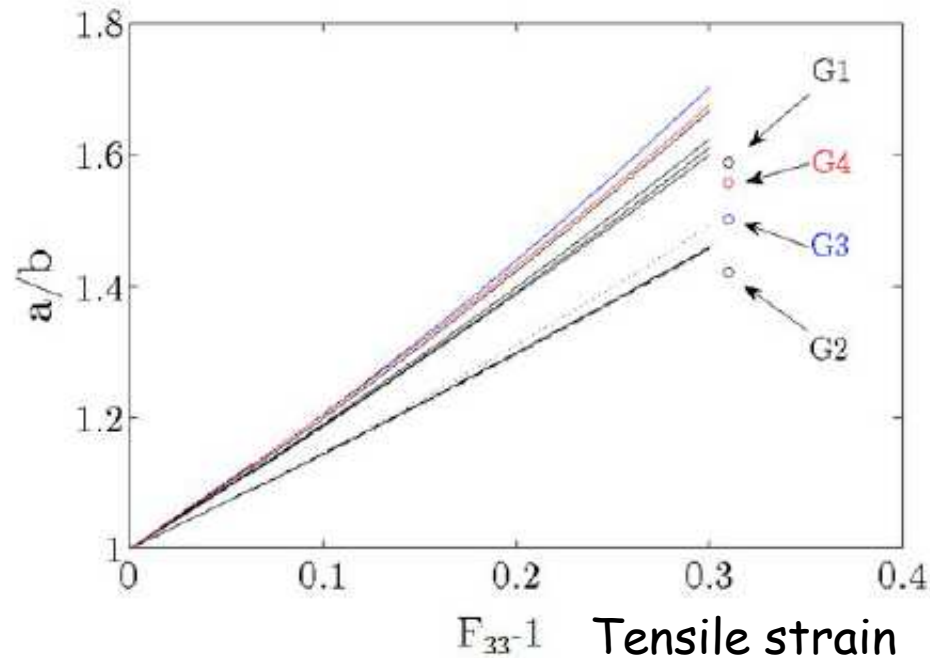
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Typical experimental observations after mechanical loading



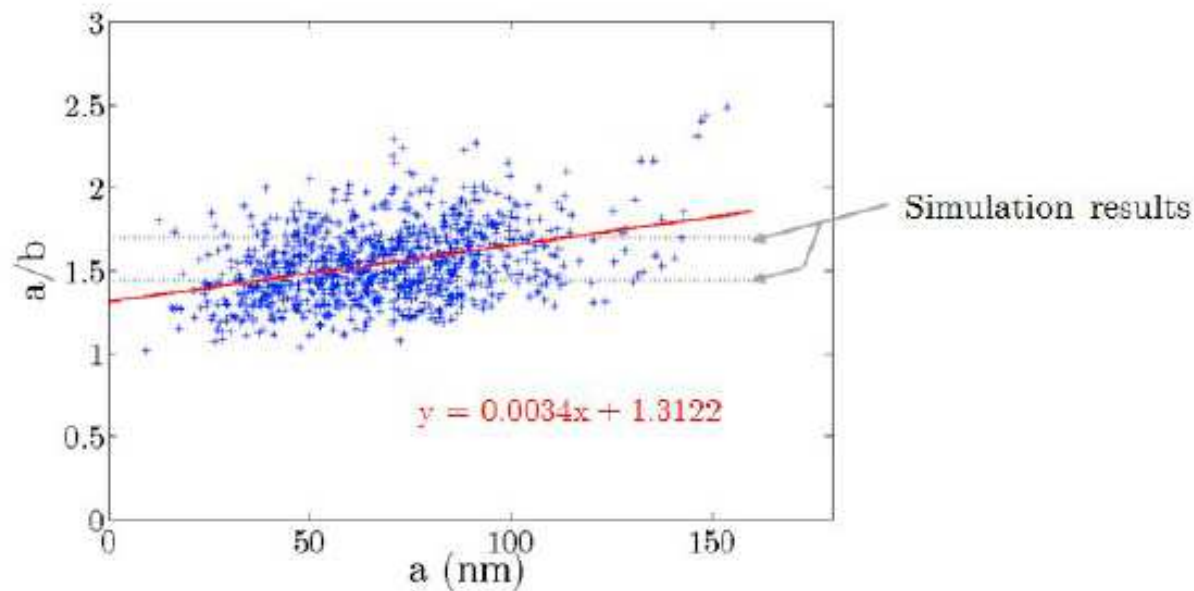
- Elongation along tensile axis: ellipsoidal shapes
- TEM measurements (up to now...):
 - ✓ Ratio a/b of the semi-axis of the plane projection of the ellipsoid...
 - ✓ ...in different grains (\neq crystallographic orientations)

Experimental and numerical (crystal plasticity) results for the mean ratio a/b



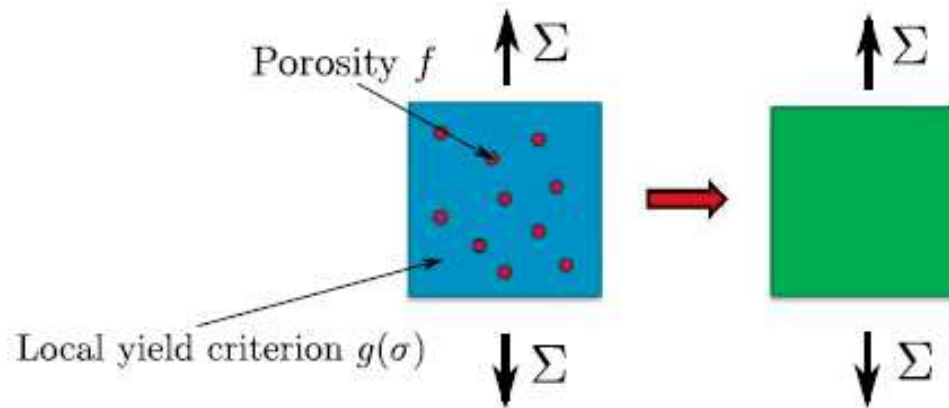
- Differences (slight) between different crystallographic orientations
- Good agreement with numerical simulations! Why?

Statistics of nano-voids aspect ratio after mechanical loading



- Strong variability of void deformation → Modelling ?
 - Slight effect: The smaller the void, the less the aspect ratio
- ✓ But $R > \left[\frac{1}{\sqrt{\rho}}, \frac{\gamma}{\sigma_{ys}} \right]$ → weak (or no) size effect: consistent with data

From a porous material (of porosity f) to an effective (equivalent) material:



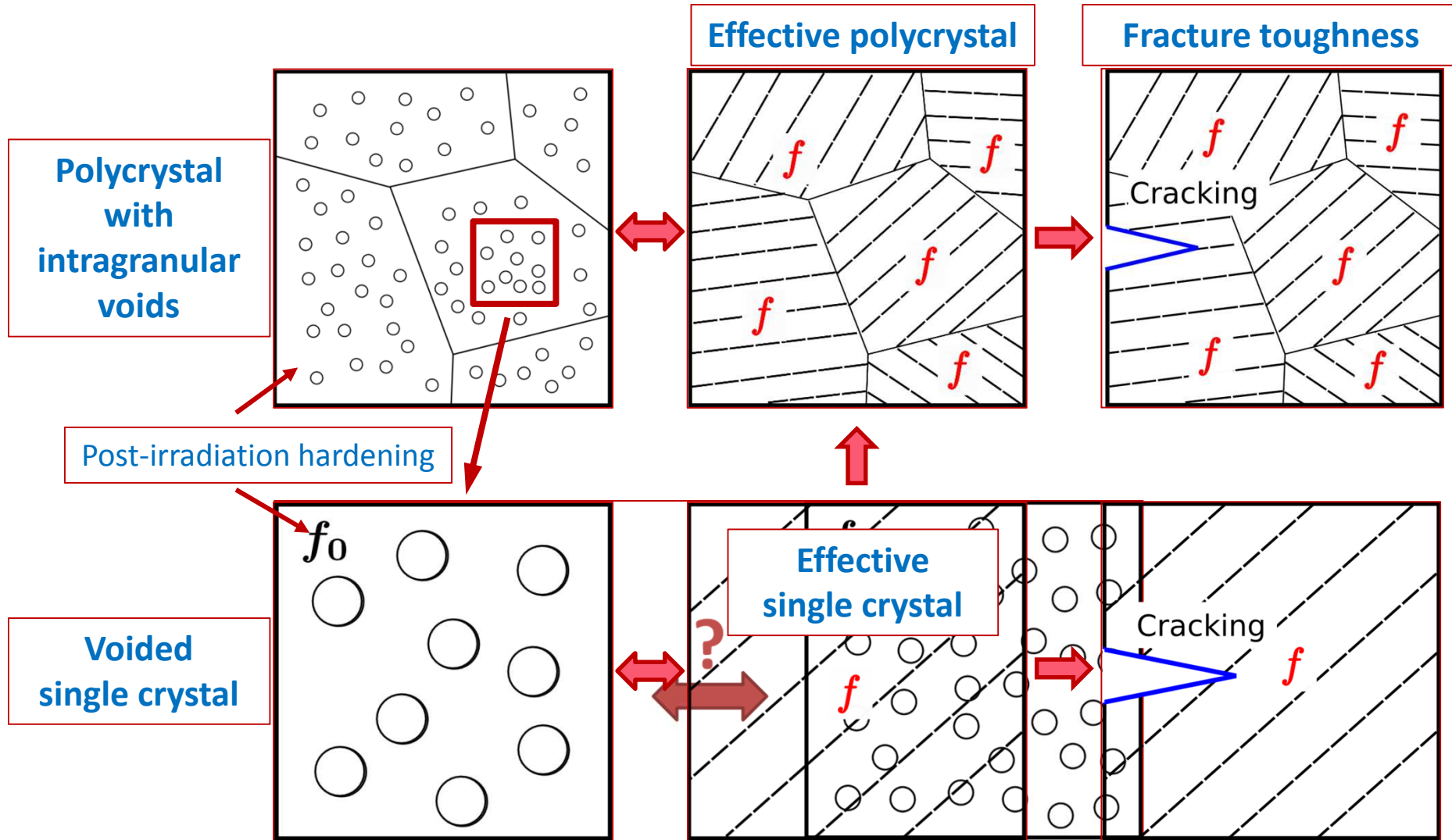
- Effective yield criterion $\mathcal{F}(\Sigma, f, \dots)$
- Evolution of internal variable $\dot{f} = \dots$
- For growth and coalescence

To obtain the **effective constitutive equations** requires:

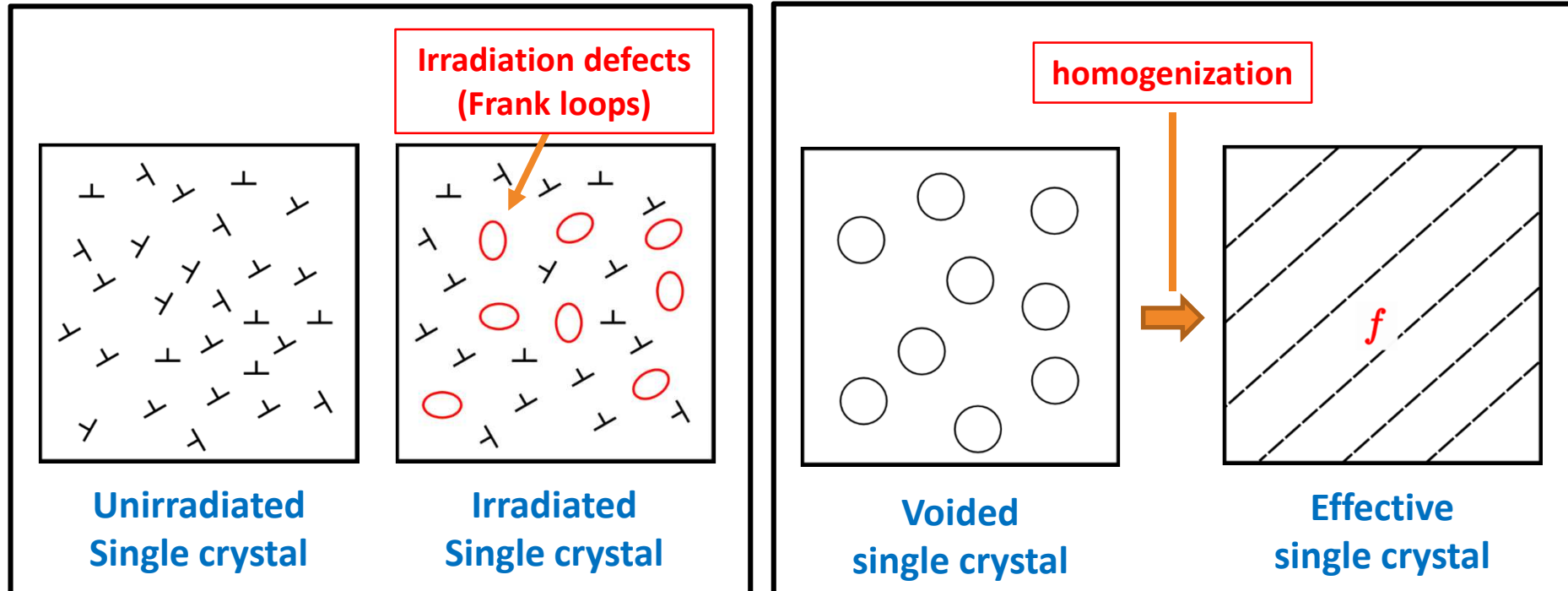
- **Experimental data** for void growth and coalescence
- **Theoretical approach:** homogenisation, limit analysis

- **Numerical simulations**

for different void lengthscales (μm , nm)



▣ Needed tools



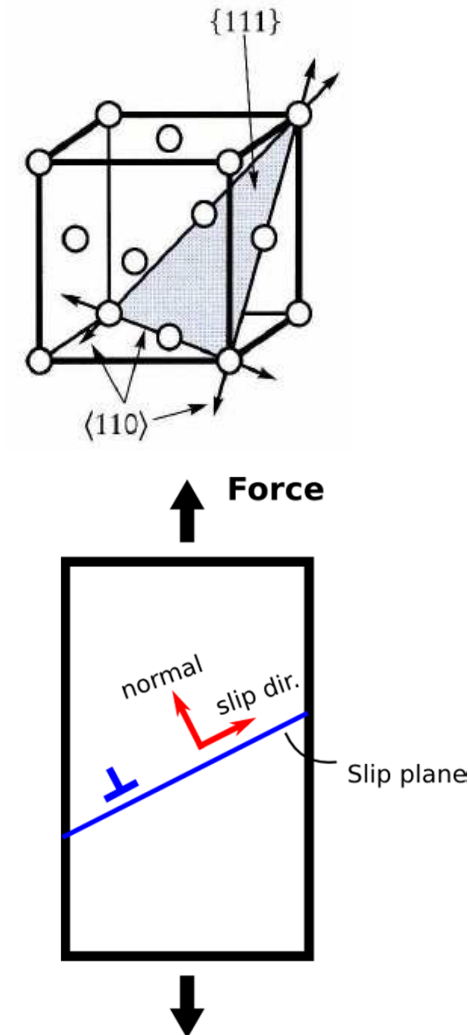
- ✓ Constitutive model for irradiated FCC single crystals accounting for Frank loops
- ✓ Yield function for single crystals containing voids including void growth and coalescence

□ FCC crystal

- ✓ Plasticity: dislocation motion
- ✓ Slip planes $\{111\}$
- ✓ Slip directions $\langle \bar{1}10 \rangle$
- ✓ 12 slip systems
- ✓ Schmid tensor $\underline{N}^s = \underline{m}^s \otimes \underline{n}^s$

□ Schmid's law:

Plastic slip is initiated when the resolved shear stress τ^s on a slip plane reaches a critical value τ_c^s



Void growth and coalescence at micro-scale ->theoretical background: crystal plasticity

✓ **Kinematics:**

Deformation gradient: $\tilde{\mathbf{F}} = \tilde{\mathbf{E}} \cdot \tilde{\mathbf{P}}$

✓ **Flow rule:**

Yield function: $\phi^s = \tau^{*s} - \tau_c^s \geq 0$

$$\tau^{*s} = |\tau^s| = |\tilde{\mathbf{M}} : \tilde{\mathbf{N}}^s| \quad \tilde{\mathbf{M}} = J_e \tilde{\mathbf{E}}^T \cdot \tilde{\boldsymbol{\sigma}} \cdot \tilde{\mathbf{E}}^{-T}$$

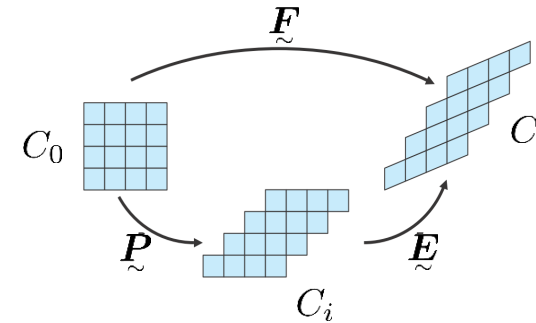
Plastic strain rate: $\dot{\tilde{\mathbf{P}}} \cdot \tilde{\mathbf{P}}^{-1} = \sum_{s=1}^{12} \dot{\gamma}^s \frac{\partial \phi^s}{\partial \tilde{\mathbf{M}}} = \sum_{s=1}^{12} \dot{\gamma}^s \text{sign}(\tau^s) \tilde{\mathbf{N}}^s$

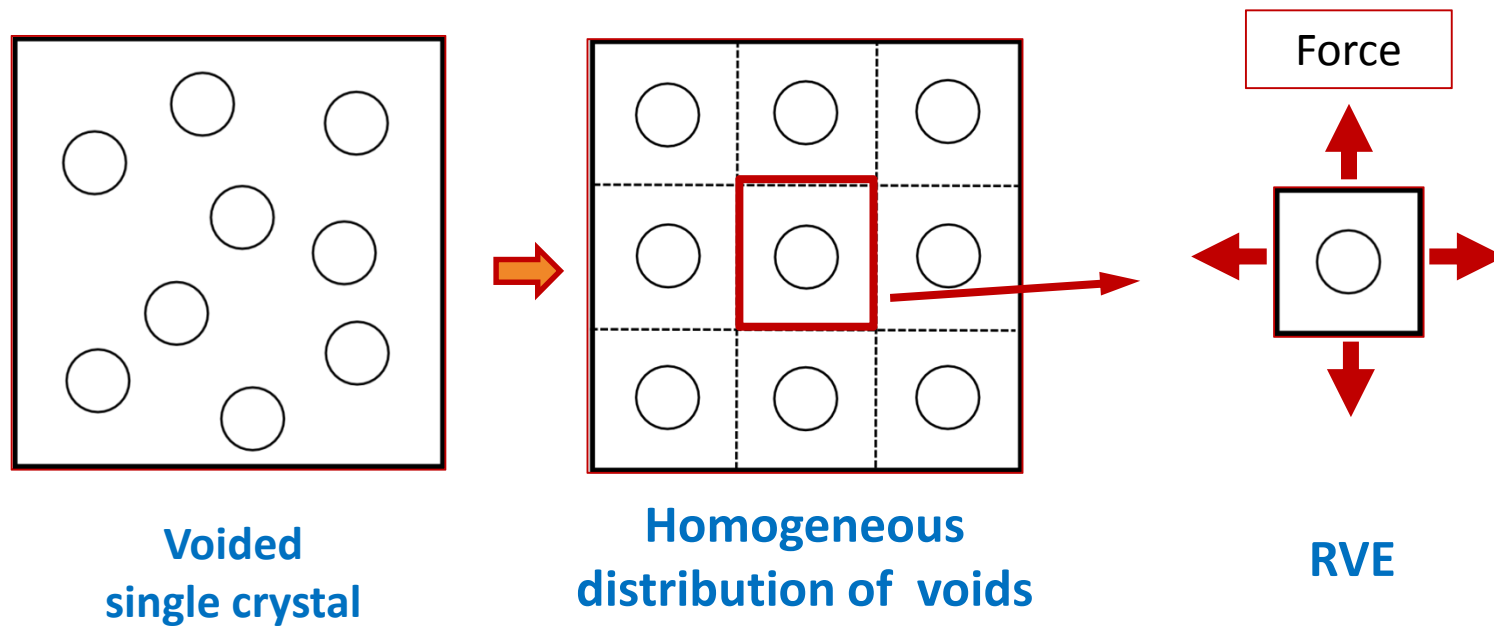
Plastic slip rate: $\dot{\gamma}^s = \dot{\gamma}_{\text{ref}} \left\langle \frac{\phi^s}{\tau_{\text{ref}}} \right\rangle^n$

✓ **Hardening rule** $\tau_c^s = \tau_T^s + \mu b_D \sqrt{\sum_{u=1}^{12} a^{su} \rho_D^u}$ [Kubin (2008)]

✓ **Dislocation density** $\dot{\rho}_D^s = \frac{1}{b_D} \left(\frac{1}{L^s} - g_c \rho_D^s \right) \dot{\gamma}^s$ with $L^s = \kappa \left(\sum_{u=1}^{12} b^{su} \rho_D^u \right)^{-1/2}$

↙
↘
 Multiplication Annihilation





RVE: Representative Volume Element

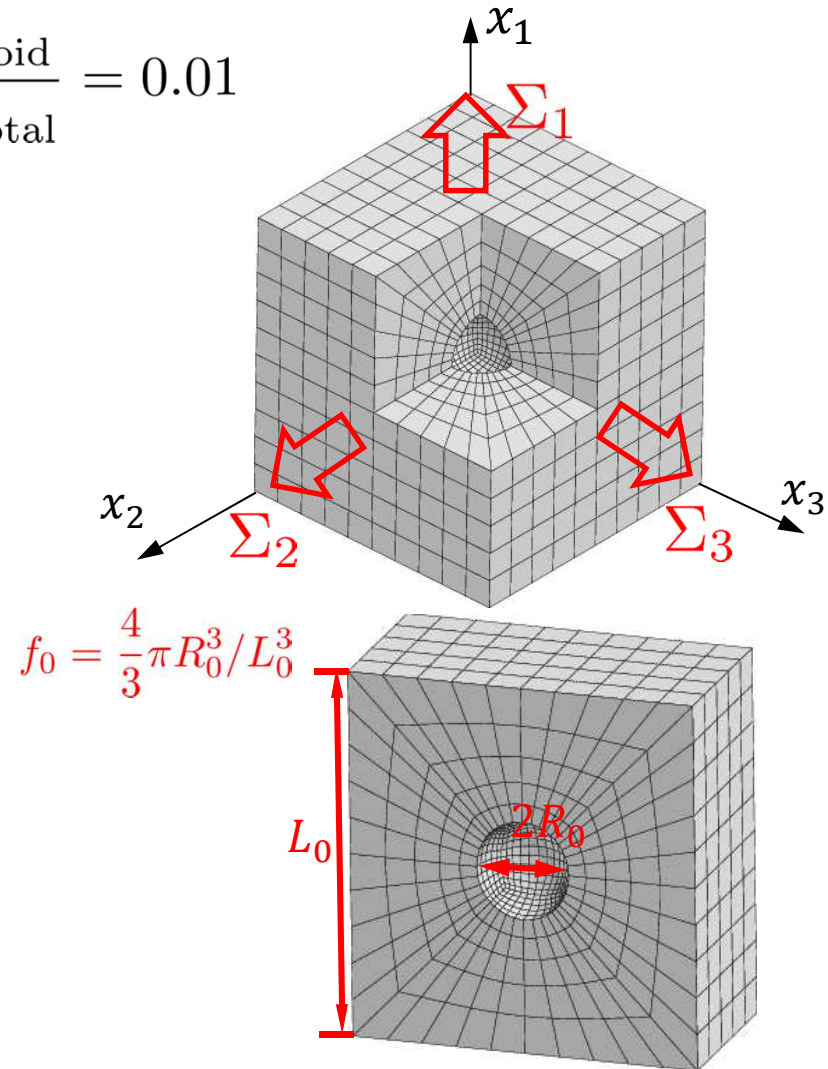
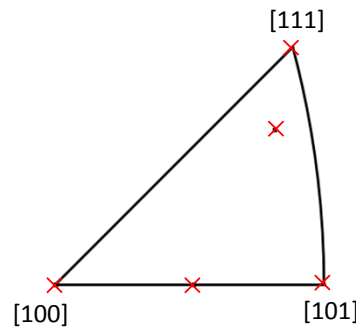
- Initial void volume fraction $f = \frac{V_{\text{void}}}{V_{\text{total}}} = 0.01$
- Periodic boundary conditions
- Axisymmetric loading $\Sigma_2 = \Sigma_3$
- Constant stress triaxiality

$$T = \frac{\Sigma_m}{\Sigma_{eq}} = 1 - 3$$

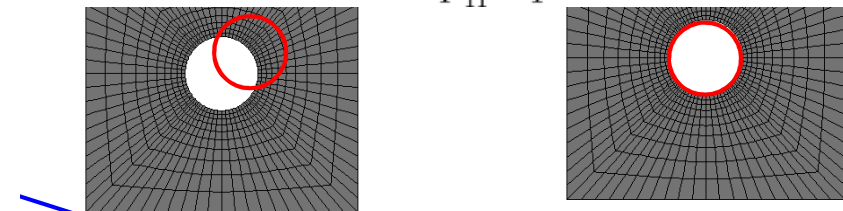
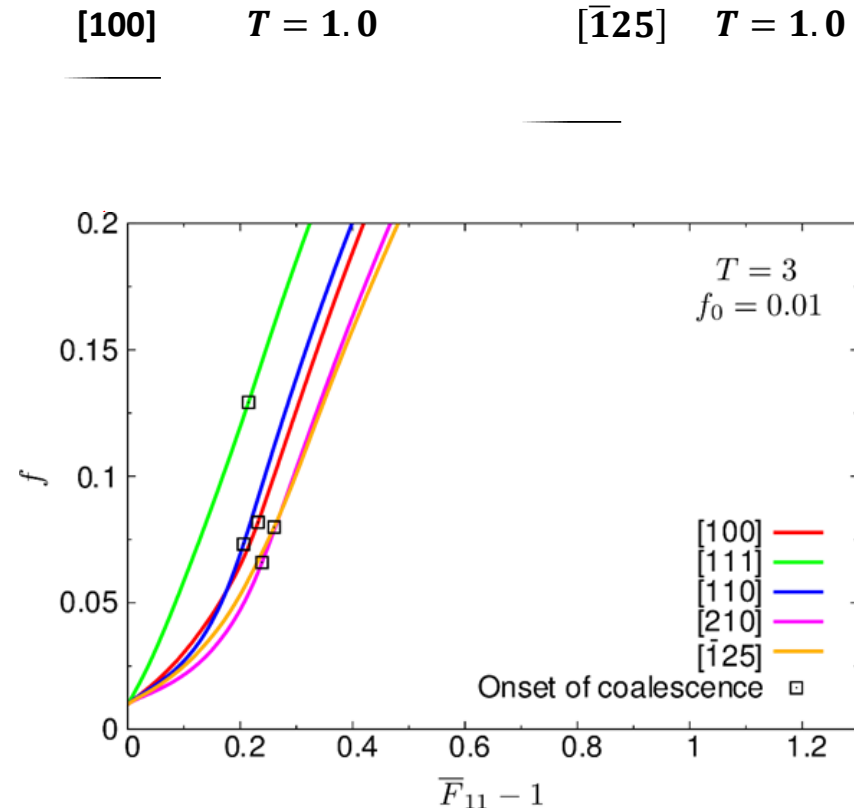
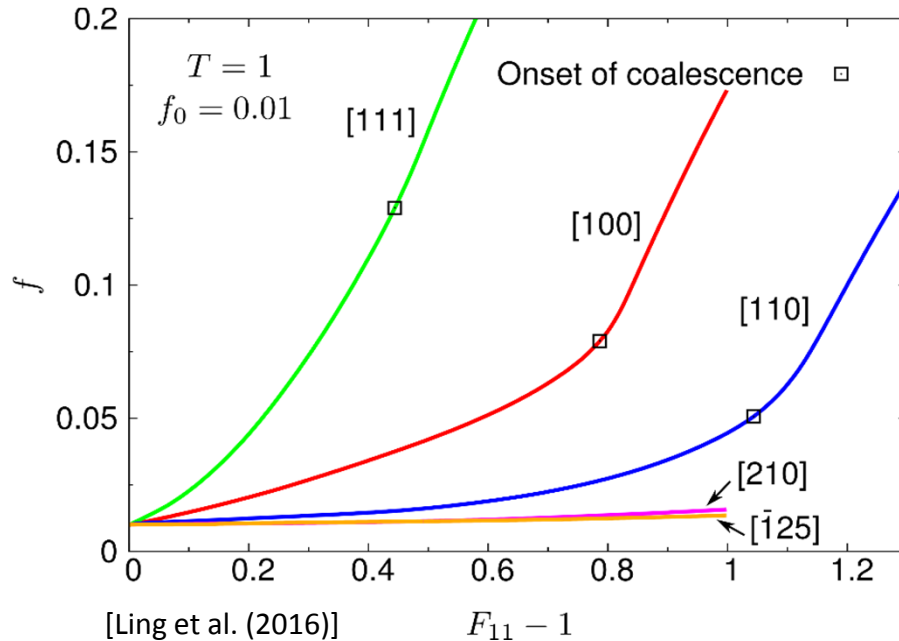
- Different crystal orientations

$x_1 - x_2 - x_3$

[100]-[010]-[001]
 [110]-[1̄10]-[001]
 [111]-[2̄11]-[01̄1]
 [210]-[1̄20]-[001]
 [1̄25]-[12̄1]-[210]



Void growth and coalescence at micro-scale ->Effect of crystal orientation



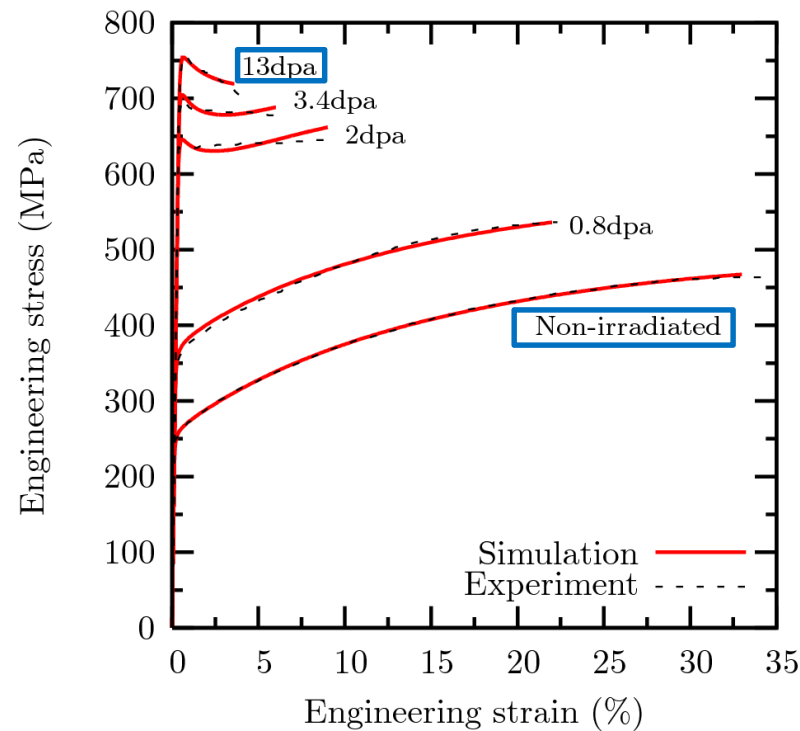
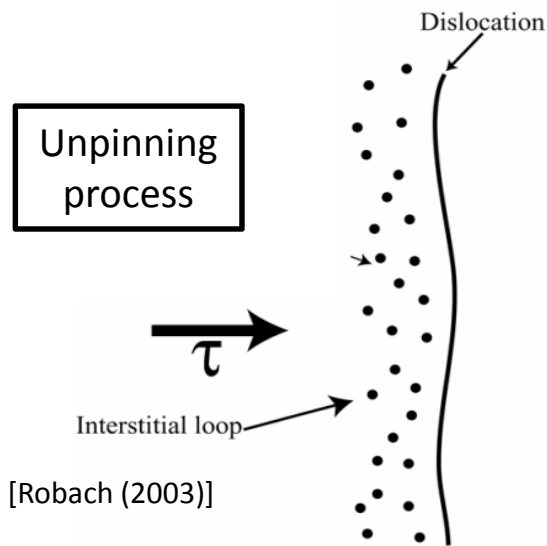
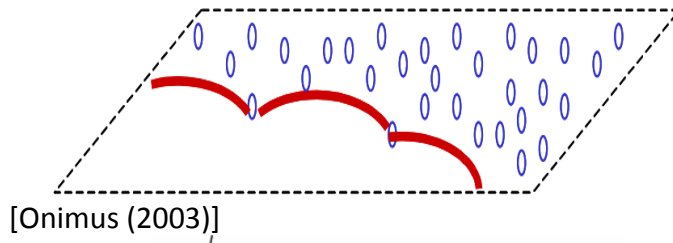
- ✓ Effect of crystal orientation on the evolution of void shape
- ✓ Two stages: growth and coalescence
- ✓ Significant effect of the crystal orientation on void growth rate at $T=1$

Void growth and coalescence at micro-scale ->Effect of post-irradiation hardening

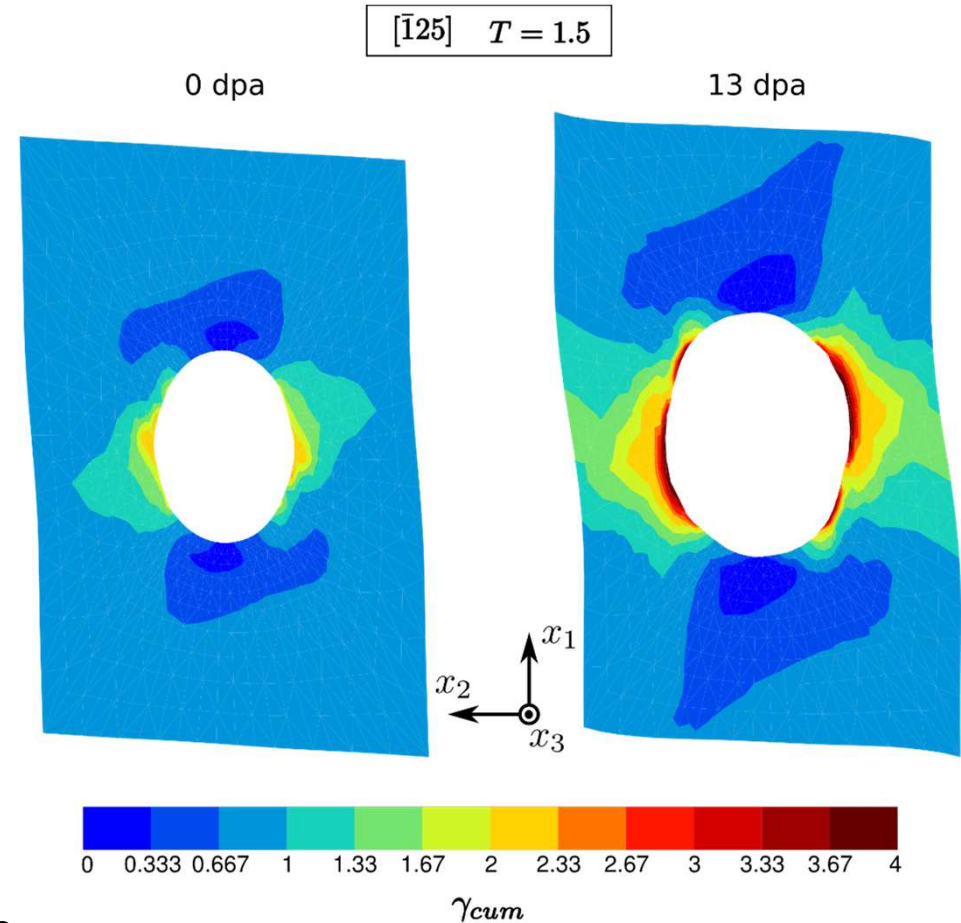
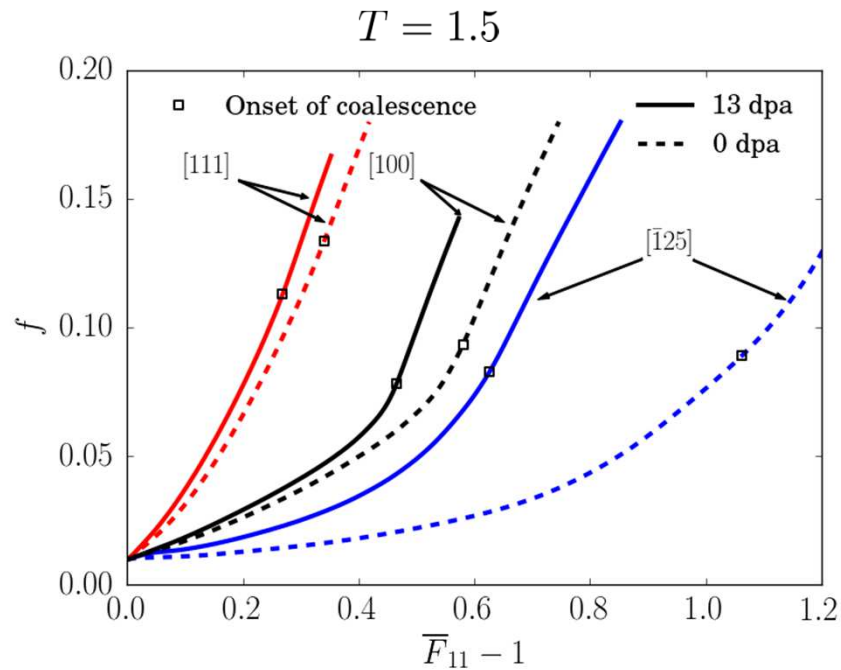
Hardening law

$$\tau_c^s = \tau_T^s + \underbrace{\mu b_D \sqrt{\sum_{u=1}^{12} a^{su} \rho_D^u}}_{\text{Dislocations}} + \underbrace{\alpha_L \mu b_L \sqrt{\sum_{p=1}^4 \phi_L \rho_L^p}}_{\text{Frank loops}} + \underbrace{\tau_a \exp\left(-\frac{\gamma^s}{\gamma_0}\right)}_{\text{Unpinning term}}$$

[Tanguy et al., Plasticity Conf., 2013]



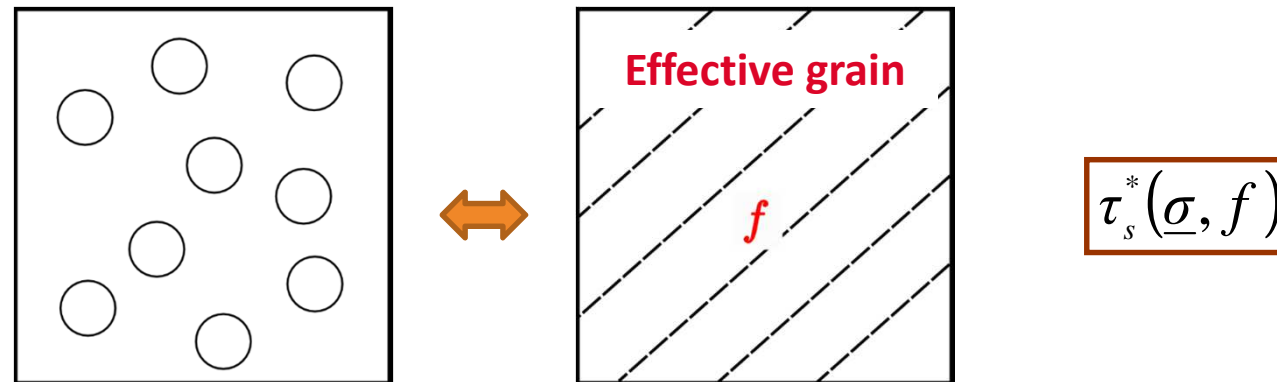
Void growth and coalescence at micro-scale ->Effect of post-irradiation hardening



- ✓ Void growth is accelerated after the irradiation.
- ✓ Higher void growth rate induced by more significant localization of plastic slip.

$$F_{11} - 1 = 0.5$$

- ❑ Void growth rate depends on crystal orientation and the effect is more significant at lower stress triaxialities.
 - ✓ This justifies the proposed approach for modeling ductile fracture at the scale of grain
- ❑ Void growth is accelerated after irradiation:
 - ✓ This implies a decrease in fracture toughness after irradiation



Yield function for single crystals containing voids [Han et al., 2013, IJSS 50]

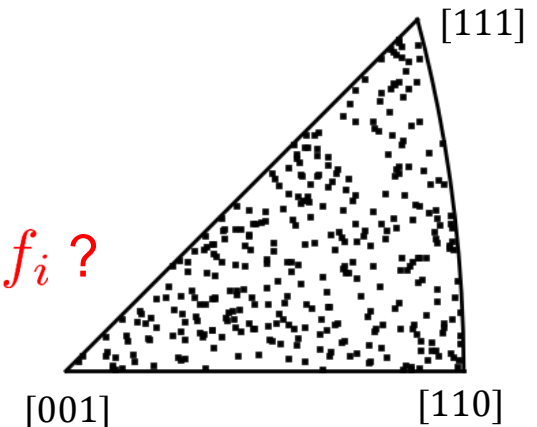
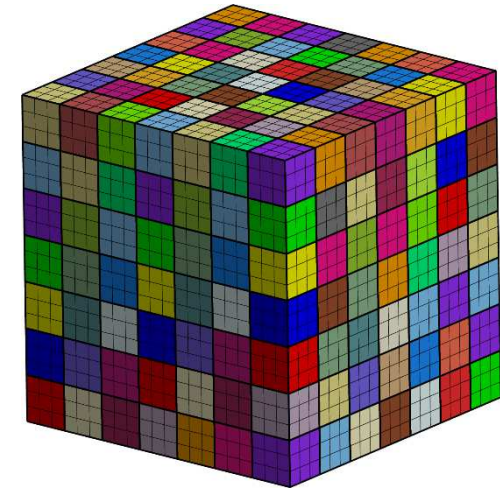
$$\left(\frac{\tau^{s2}}{\tau_c^{s2}} + \alpha \frac{2}{45} f \frac{\sigma_{eq}^2}{\tau_c^{s2}} \right) + 2q_1 f \cosh \left(q_2 \sqrt{\frac{3}{20}} \frac{\sigma_m}{\tau_c^s} \right) - 1 - q_1^2 f^2 = 0$$

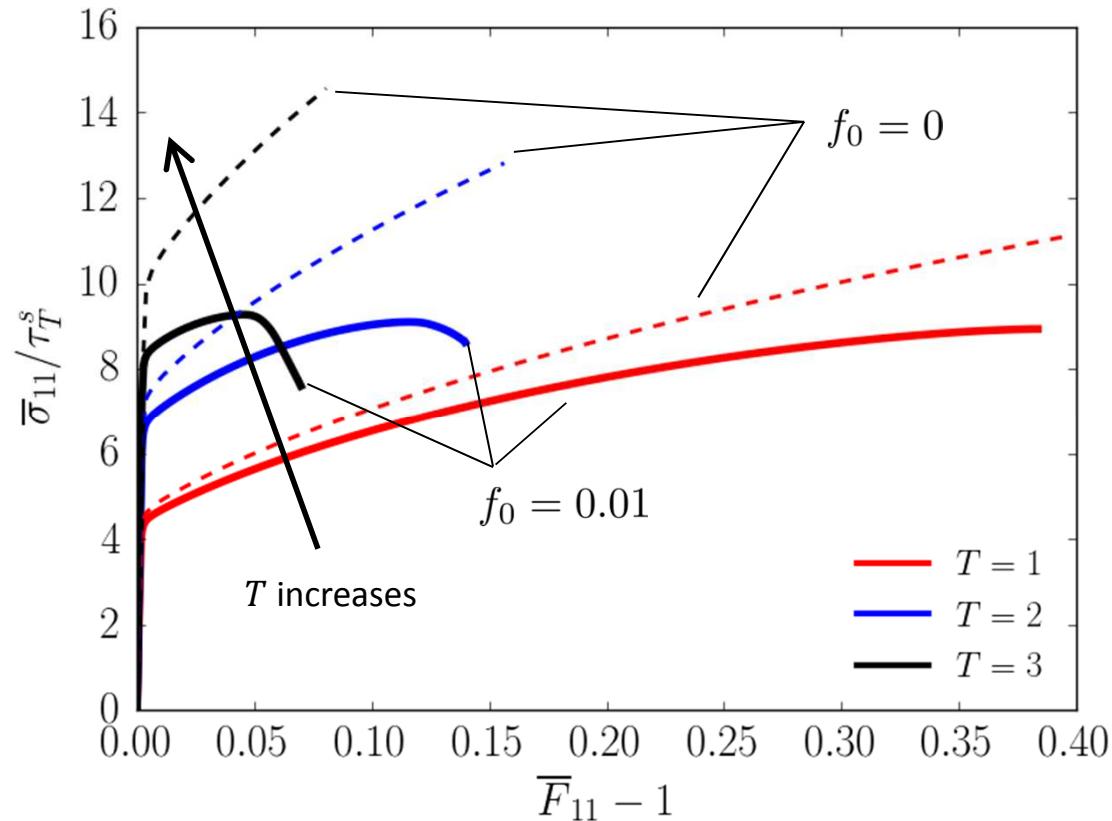
Definition of the **effective scalar resolved shear stress** (for each slip system s)

- ✓ f : void volume fraction
- ✓ α, q_1, q_2 : heuristic parameters used to better represent the result of unit cell simulations

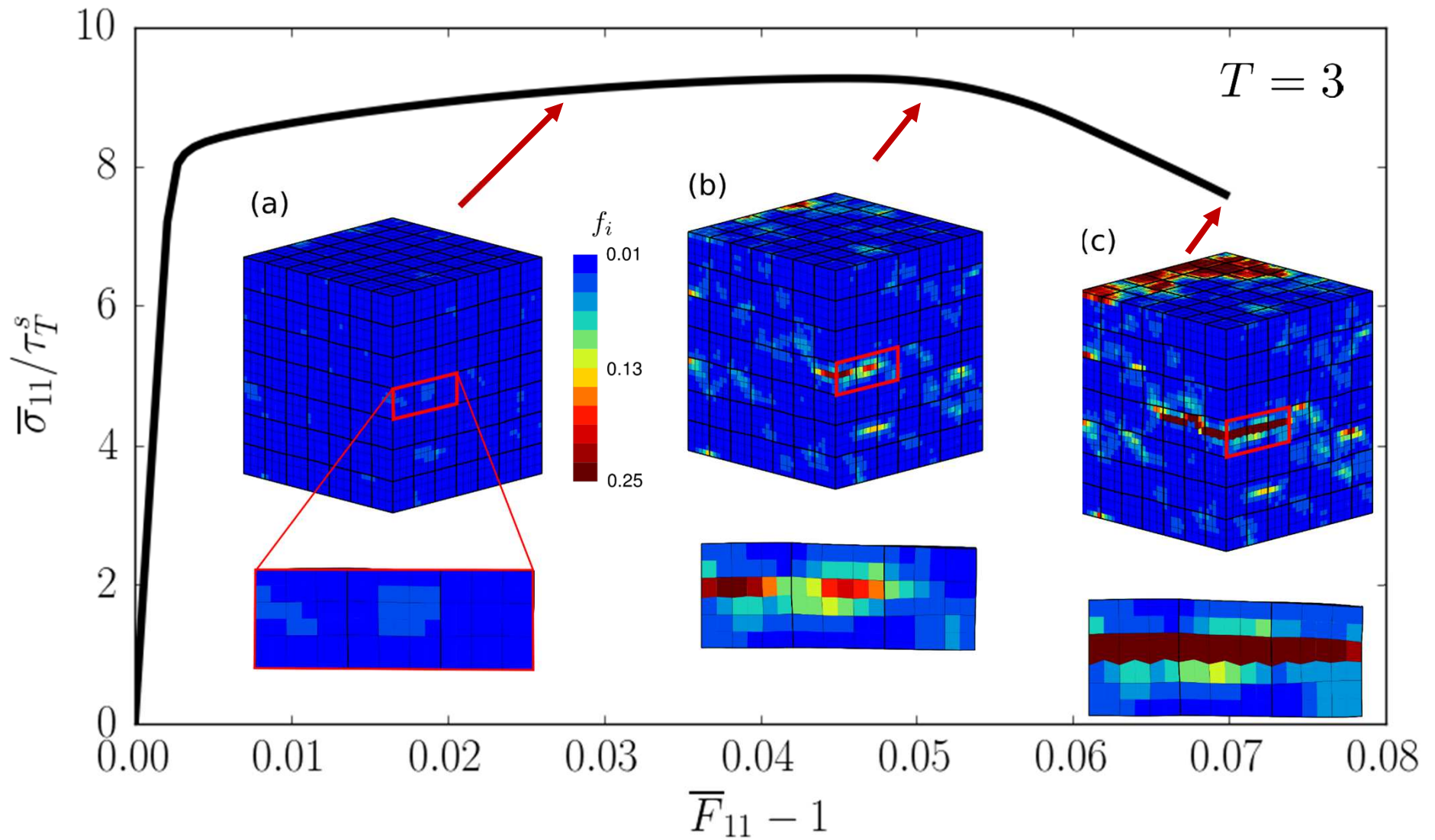
- ✓ Extended to void growth and Finite strain in [Ling et al., IJP, 2016]

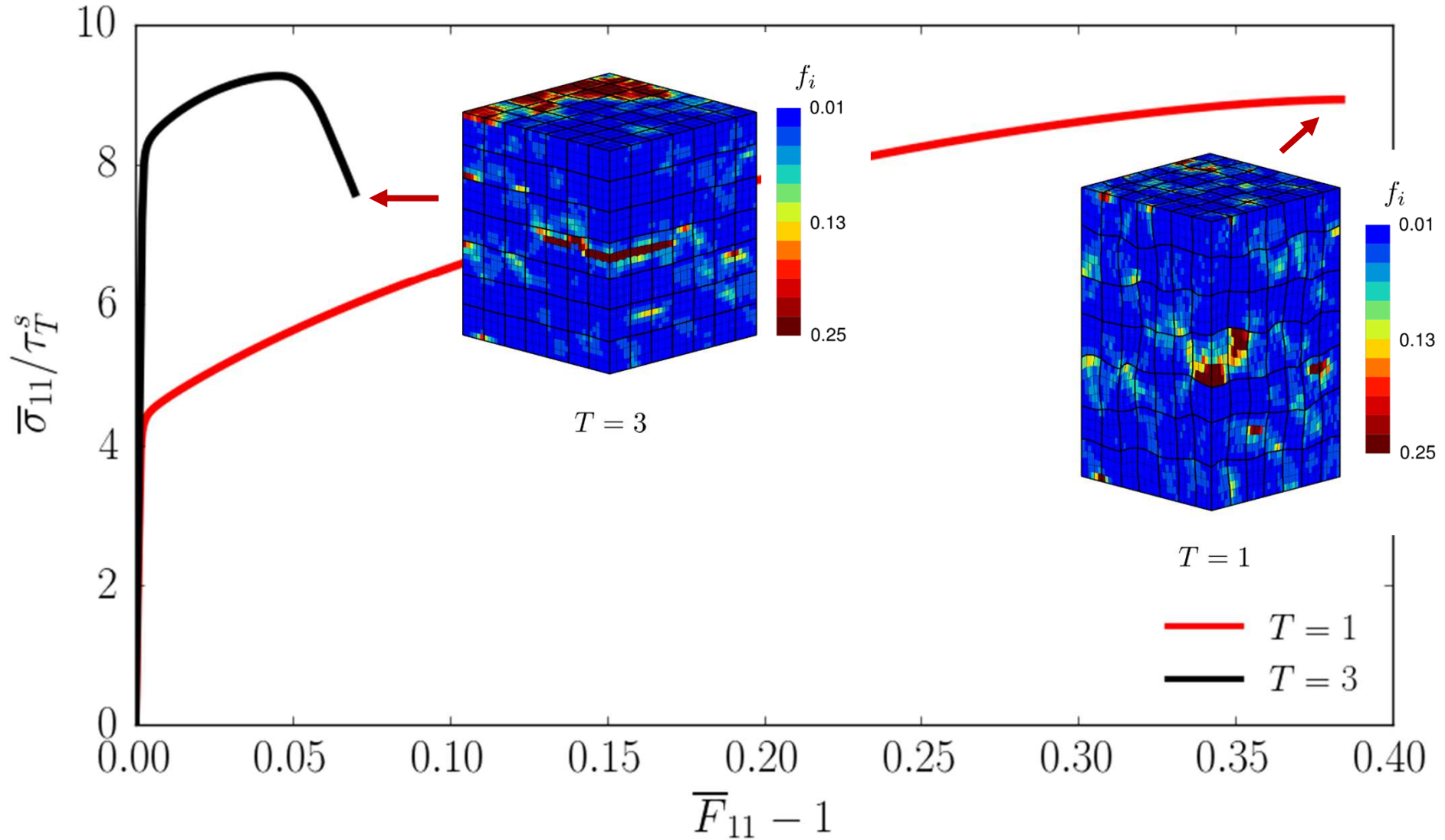
- ❑ 343 grains, 27 quadratic elements/grain
- ❑ Random distribution of grain orientation
- ❑ Initial void volume fraction 0.01
- ❑ Hardening law for the unirradiated steel
- ❑ Constant overall stress triaxiality
- ❑ Local evolution of damage variable (porosity) f_i ?

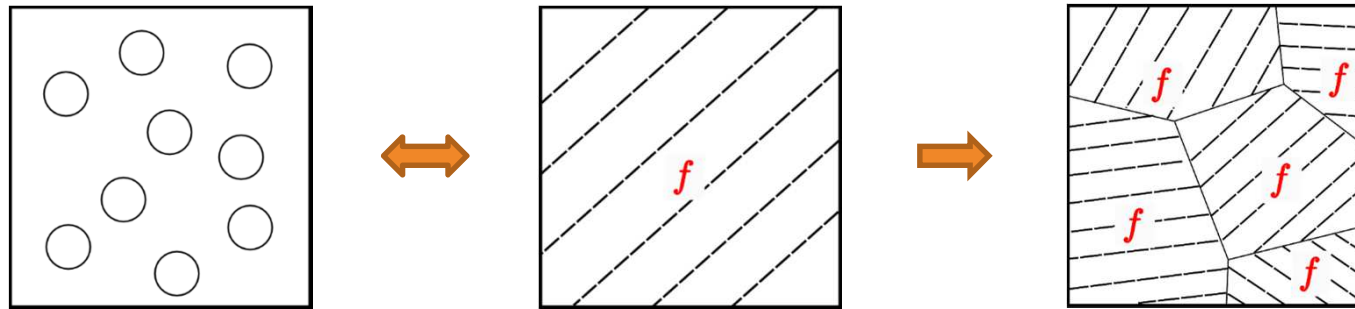




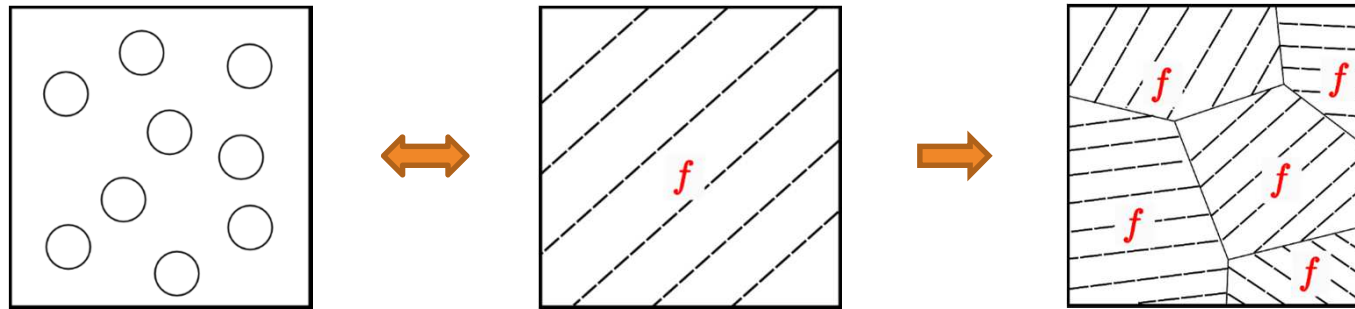
- Higher stress triaxiality increase void growth rate, leading to earlier softening.
- The basic effect of triaxiality on ductile damage is captured.







- A multi-scale approach for modeling intragranular ductile fracture of irradiated stainless steels.
- Unit cell simulations for studying void growth in single crystals:
 - ✓ Capture well the effect of irradiation on growth rate
- The first porous single crystal plasticity model at finite strains incorporating hardening.
- The first simulations of ductile damage initiation and propagation in a polycrystal aggregate.



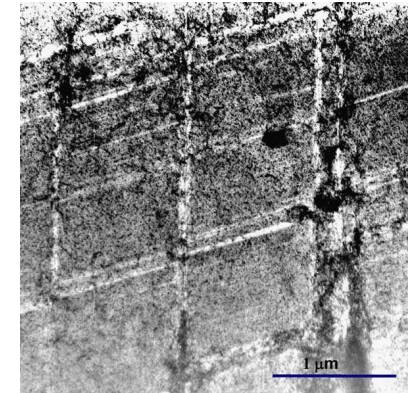
□ Developed tools can be applied to describe ductile damage of others materials as far as it is driven by growth and coalescence

□ Potential applications: void growth in Zirconium alloy for fuel cladding, micro-crack growth in Ni based single crystal superalloys



turbine blades in jet turbo-engines

- Enhancement of the crystal plasticity model to describe size effect (On-going work)
- Investigate the effect of deformation channels
 - ✓ growth and coalescence of micro and nanovoids
 - ✓ Nucleation of voids
- Refined the yield criterion for porous crystal in the coalescence regime
- Prediction of the evolution of the fracture toughness of irradiated austenitic stainless steels



[Byun et al. 2006]

- "Void growth and coalescence in triaxial stress fields in irradiated FCC single crystals" C. Ling, B. Tanguy, J. Besson, S. Forest, F. Latourte. *Journal of Nuclear Materials*, 492 (2017) 157-170
- "An elastoviscoplastic model for porous single crystals at finite strains and its assessment based on unit cell simulations" C. Ling, J. Besson, S. Forest, B. Tanguy, F. Latourte, E. Bosso, *International Journal of Plasticity*, 84 (2016), 58-87
- "A yield function for single crystals containing voids" Xu Han; Jacques Besson; Samuel Forest; Benoit Tanguy; Stéphane Bugat, *Int. Journal of Solids and Structure* 50 (2013) 2215-2131
- "Void growth and coalescence in irradiated materials " P.O. Barrioz, J. Hure and B. Tanguy, 14th International Conference on Fracture (ICF 14) June 18-23, 2017, Rhodes, Greece
- Simulations of polycrystalline aggregate under triaxial loading accounting for intragranular cavities by a homogenization model" C. Ling, B. Tanguy, S. Forest, J. Besson, F. Latourte, 15th European Mechanics of Materials Conference, 7-9 September 2016 - Brussels, Belgium
- "Experimental assessment of nanovoids growth", P.O. Barrioz, J. Hure and B. Tanguy, XXIV ICTAM, 21-26 August 2016, Montreal, Canada
- "Void size effect on its growth and coalescence in single crystals", C. Ling, B. Tanguy, S. Forest, J. Besson, F. Latourte, E. Bosso, XXIV ICTAM, 21-26 August 2016, Montreal, Canada
- "Void Growth in FCC Single Crystal - Comparison Between Gurson-type Model and Unit Cell Simulations" C. Ling, J. Besson, S. Forest, B. Tanguy, E. Bosso, F. Latourte, 9th European Solid Mechanics Conference, July 6-10, Madrid, Spain, 2015
- "Ductile damage behavior modelling of irradiated austenitic steels based on a Gurson-type model for porous single crystals" B. Tanguy, X. Han, S. Forest, J. Besson, C. Ling, J. Hure, M. Callahan, F. Latourte 14Th European Mechanics of Materials Conference, EMMC14 Gothenburg, Sweden, August 27-29, 2014
- "A Gurson-Type Model to Describe the Behavior of Porous Single Crystals" B. Tanguy, X. Han, J. Besson, S. Forest, Symposium Materials Fundamentals of Fatigue and Fracture, MRS Fall Meeting, Boston, MA, December 2-5, 2013
- "Dislocations and Irradiation Defects-Based Micromechanical Modelling For Neutron Irradiated Austenitic Stainless Steels" B. Tanguy, X. Han, J. Besson, S. Forest, C. Robertson, N. Rupin, International Symposium on Plasticity 2013 and its current applications, Nassau, Bahamas, 3-8 January 2013

Thank you for your attention!



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