



HAL
open science

Mechanical behaviour of UO₂ under irradiation a molecular dynamics study

L. van Brutzel, A. Chartier

► **To cite this version:**

L. van Brutzel, A. Chartier. Mechanical behaviour of UO₂ under irradiation a molecular dynamics study. NuFuel and MMSNF 2015, Nov 2015, Karlsruhe, Germany. cea-02491637

HAL Id: cea-02491637

<https://cea.hal.science/cea-02491637>

Submitted on 26 Feb 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

DE LA RECHERCHE À L'INDUSTRIE



université
PARIS-SACLAY

www.cea.fr

MECHANICAL BEHAVIOR OF UO₂ UNDER IRRADIATION: A MOLECULAR DYNAMICS STUDY

L. Van Brutzel, A. Chartier

DEN, DPC, SCCME
CEA 91191 Gif-sur-Yvette, FRANCE

NUFUEL
Research into Nuclear Fuel in Europe

November 16-18, 2015 Karlsruhe, Germany

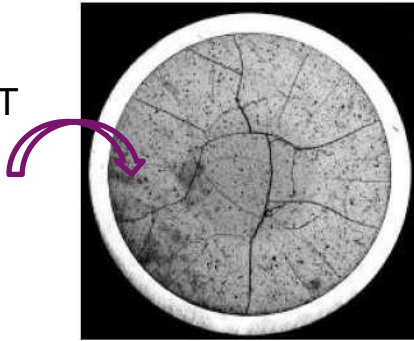
FUEL DESIGN, AN ENGINEERING CHALLENGE

*Macrograph of a fuel pellet
after nuclear reactor*

*SEM image of He
Bubbles in UO_2*

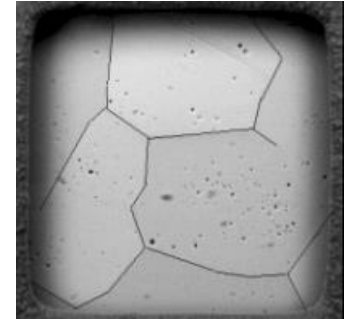
In reactors

- Temperature, ∇T
- Stress, $\nabla \sigma$
- Burn-up



In interim storage or long-term disposal

- Swelling ← He bubbles



Higher burn-up



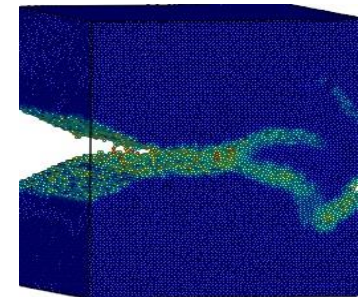
Mechanical properties ?

elasticity, yield stress, hardness, toughness...

■ **Method:** atomistic simulations with molecular dynamics

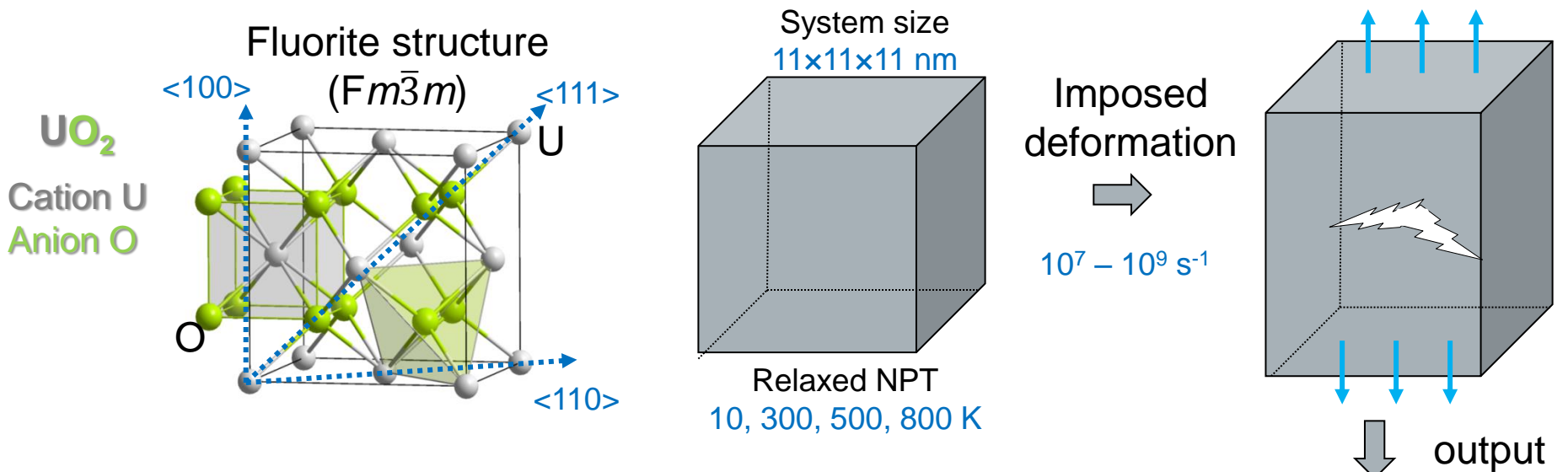
■ **Goal:**

- Understanding phenomena
- Input parameters for mesoscale models



- I. Mechanical behavior in Bulk**
- II. Mechanical behavior with Point Defects**
- III. Mechanical behavior with He Bubbles**
- IV. Crack propagation**
- V. Summary**

MECHANICAL PROPERTIES CALCULATION

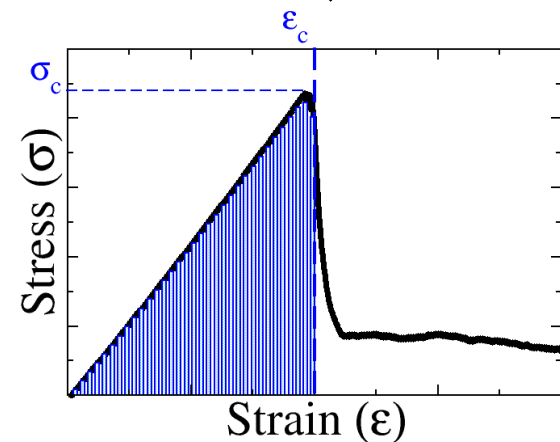


Outputs:

- Onset of crack localization
- Yield stress & strain
- Energy release rate, Toughness
- Crack propagation

Interatomic potentials:

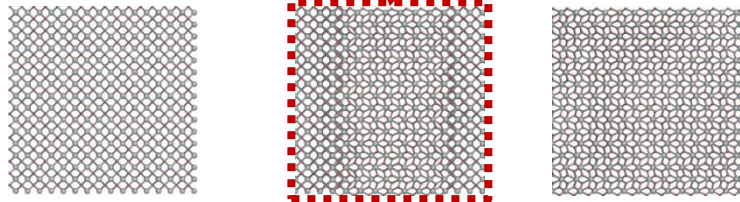
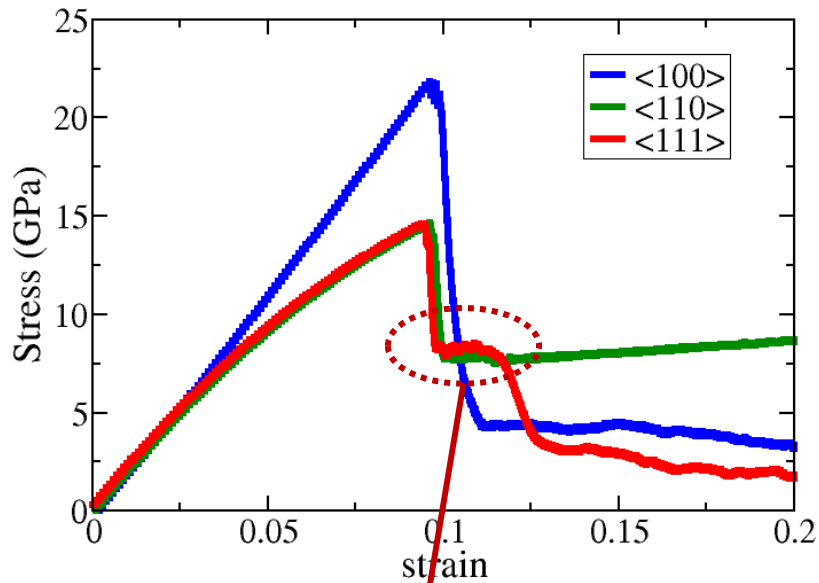
- Yakub: good elastic constants
- Morelon: good phase transitions



Energy release rate:

$$G_c = L_Z \int_0^{\epsilon_c} d\epsilon \sigma(\epsilon)$$

- I. Mechanical behavior in Bulk**
- II. Mechanical behavior with Point Defects**
- III. Mechanical behavior with He Bubbles**
- IV. Crack propagation**
- V. Summary**



Strain orientation	G_C (J/m ²)
<100>	12.73
<110>	9.54
<111>	9.51
Griffith	3.4 – 7.5

$$G_C(\text{Griffith}) = 2\gamma_S$$

γ_S : surface energy

Phase transition: fluorite \rightarrow PbO₂

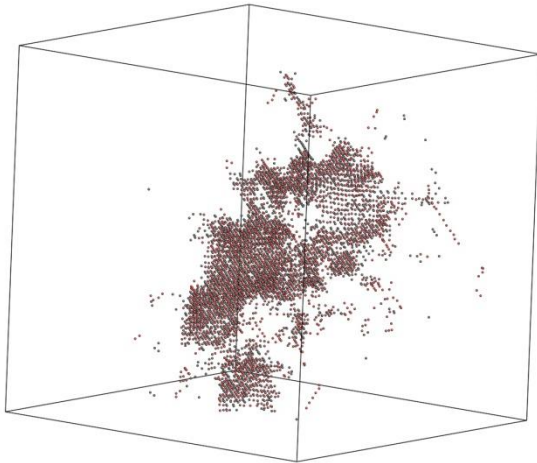


Y. Zhang JNM 430 (2012) 96
P. Fossati PRB 88 (2013) 214112

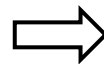
- Weakest crystallographic direction: <111>
- Difference with Griffith criteria \leftrightarrow phase transformation

- I. Mechanical behavior in Bulk
- II. Mechanical behavior with Point Defects**
- III. Mechanical behavior with He Bubbles
- IV. Crack propagation
- V. Summary

MD simulation of
80 keV displacement cascade

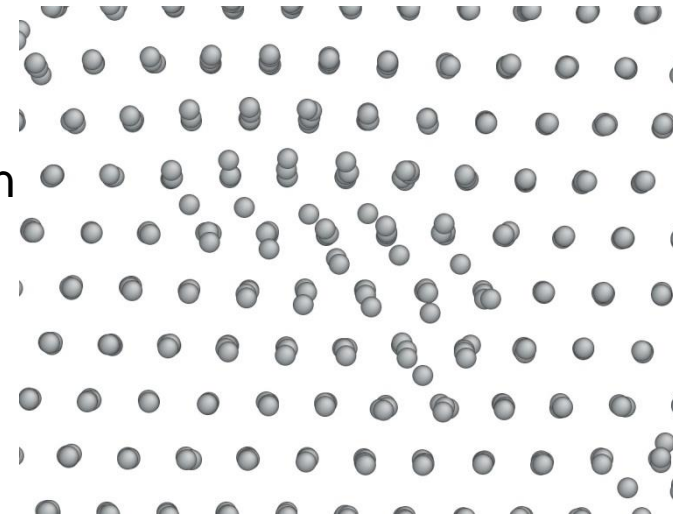


Random
Frenkel pair accumulation



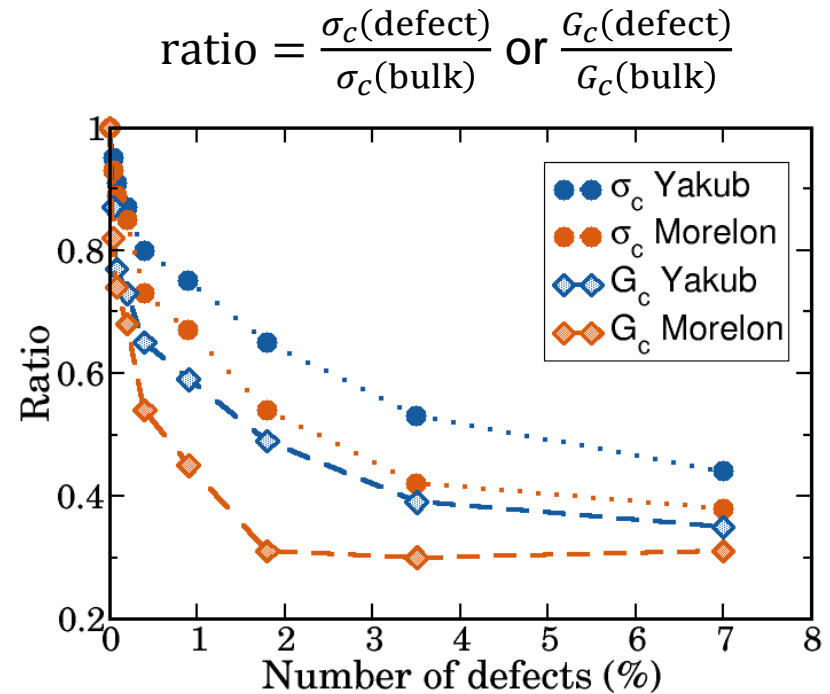
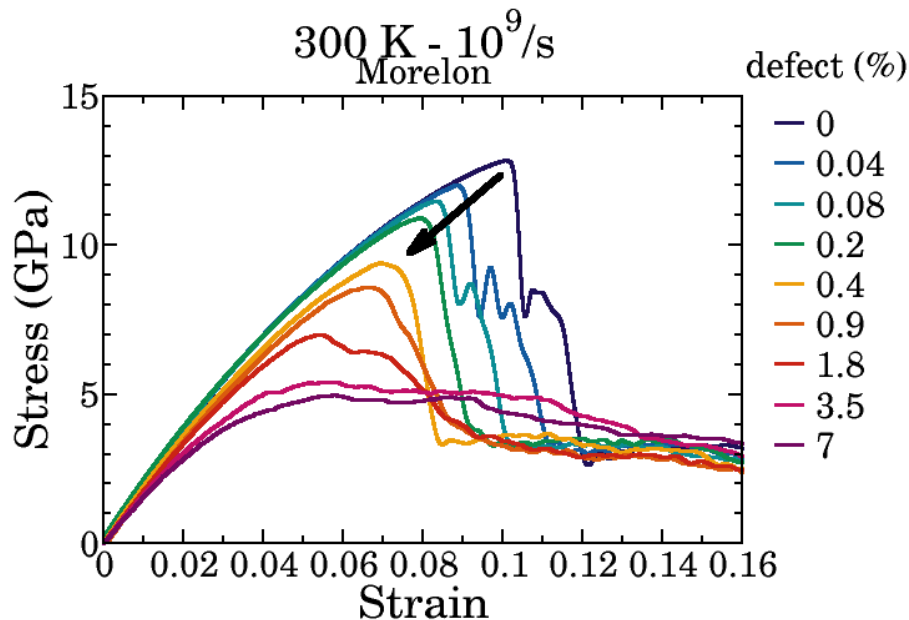
0.04 → 7%

Close up of
small Frank loops



1 cascade 80 keV → 0.04% point defects
(Frenkel pairs)

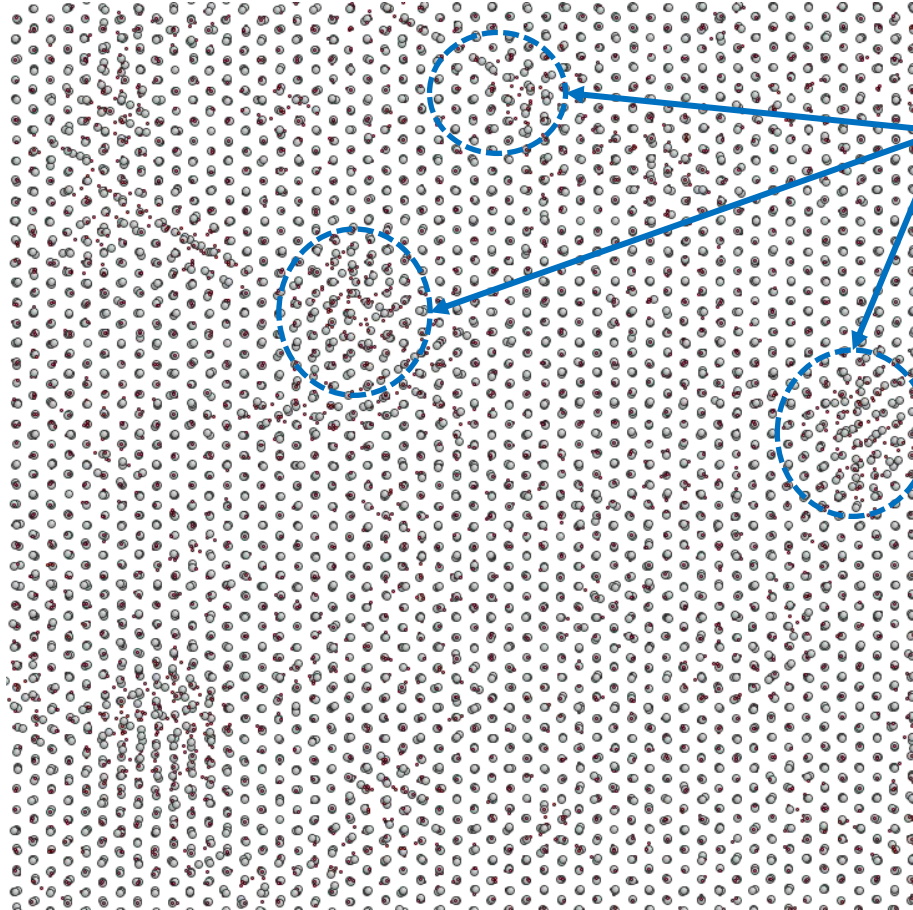
- After NPT relaxation (300 K) → Formation of **small clusters** (mainly int.)
- For systems > 2% of defects, clusters are **small dislocation loops**



- General decrease of the toughness with increase of defects
- Threshold at $\sim 1.8\%$ defects: plasticity, no phase transformation

MECHANICAL BEHAVIOR - WITH POINT DEFECTS

Slab of UO_2 ,
after 3% FP accumulation



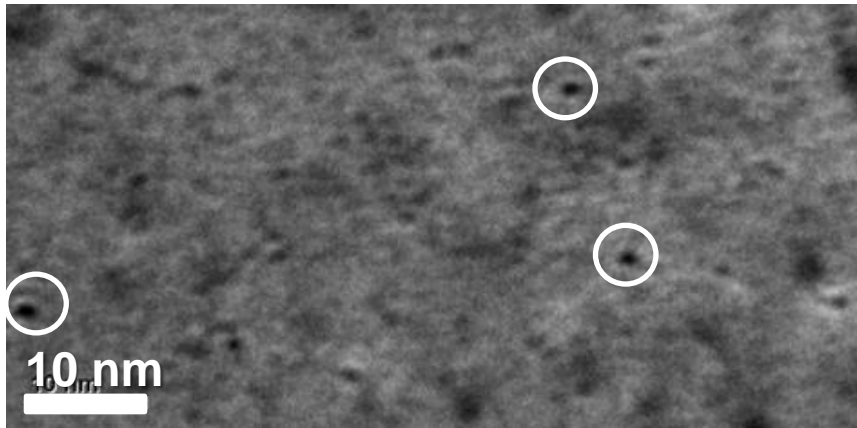
Clusters of defects

- Crack initiation: at cluster of point defects
- “Plasticity” → due to nanodomain creation

- I. Mechanical behavior in Bulk
- II. Mechanical behavior with Point Defects
- III. Mechanical behavior with He Bubbles**
- IV. Crack propagation
- V. Summary

Experiments

UO₂ irradiated 6×10¹² Au.cm⁻², 600°C



HRTEM: C. Sabathier NIMB 266 (2008) 3027

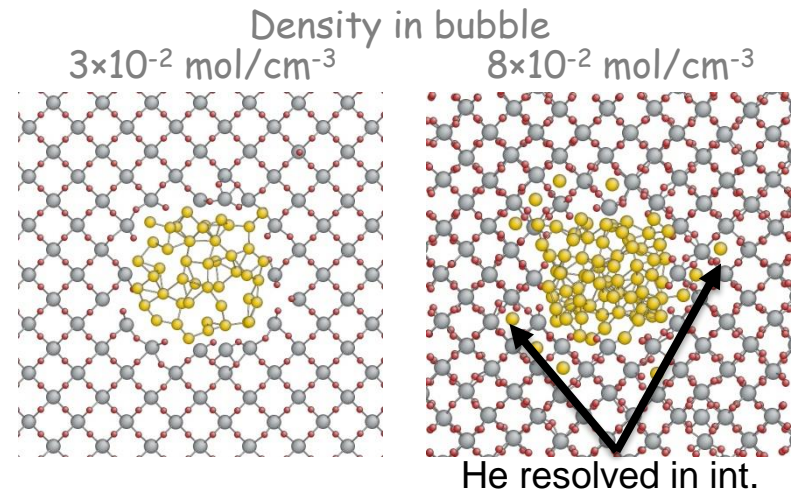
- Bubble density saturates at: $4 \times 10^{23} \text{ m}^{-3}$
- Bubble size: 1 – 2 nm

■ To fit experimental observations:

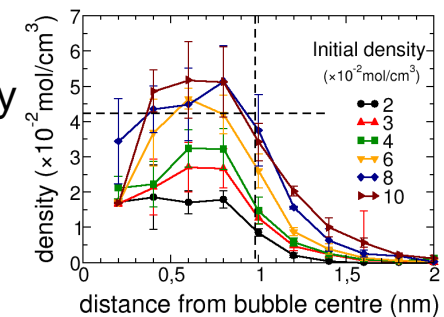
Bubble: Ø 2 nm, density of bubbles = 10^{24} m^{-3} ,

He density in bubble = $4 \times 10^{-2} \text{ mol/cm}^3$, → Pressure inside bubble = 500 MPa

MD simulations



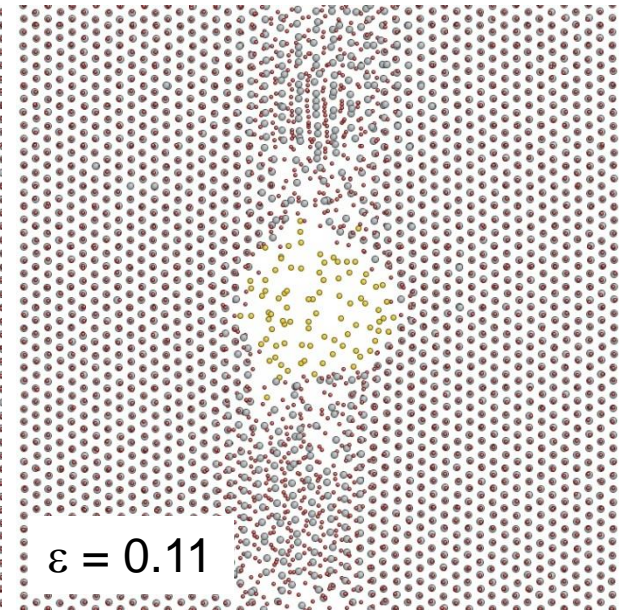
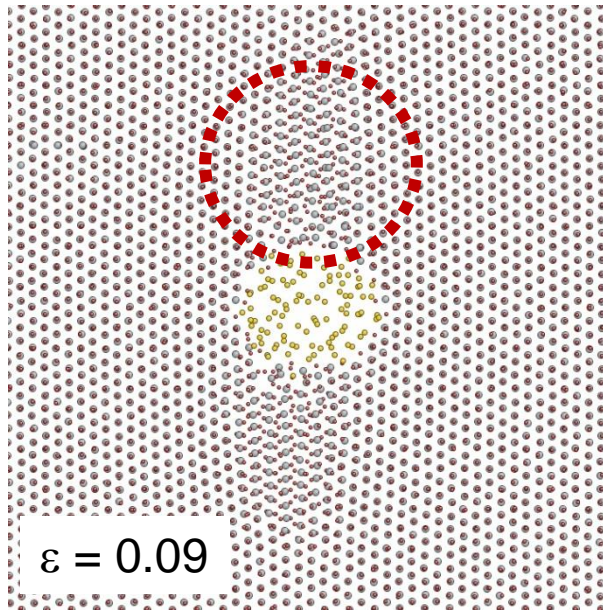
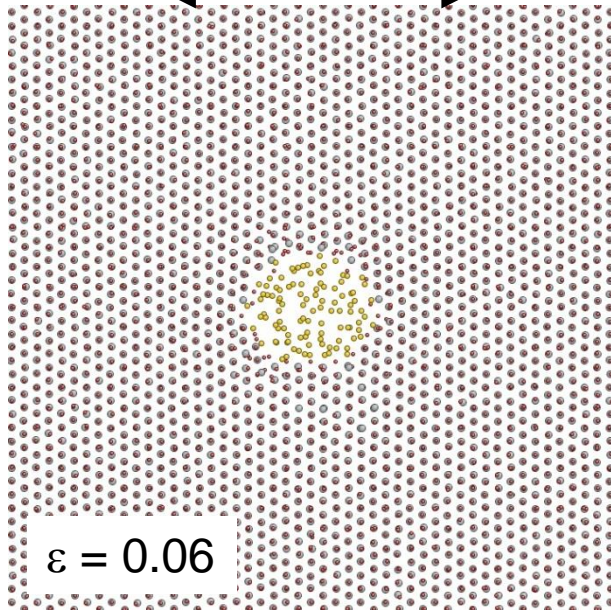
He resolution for density
> $4 \times 10^{-2} \text{ mol/cm}^3$



MECHANICAL BEHAVIOR - WITH HE BUBBLES

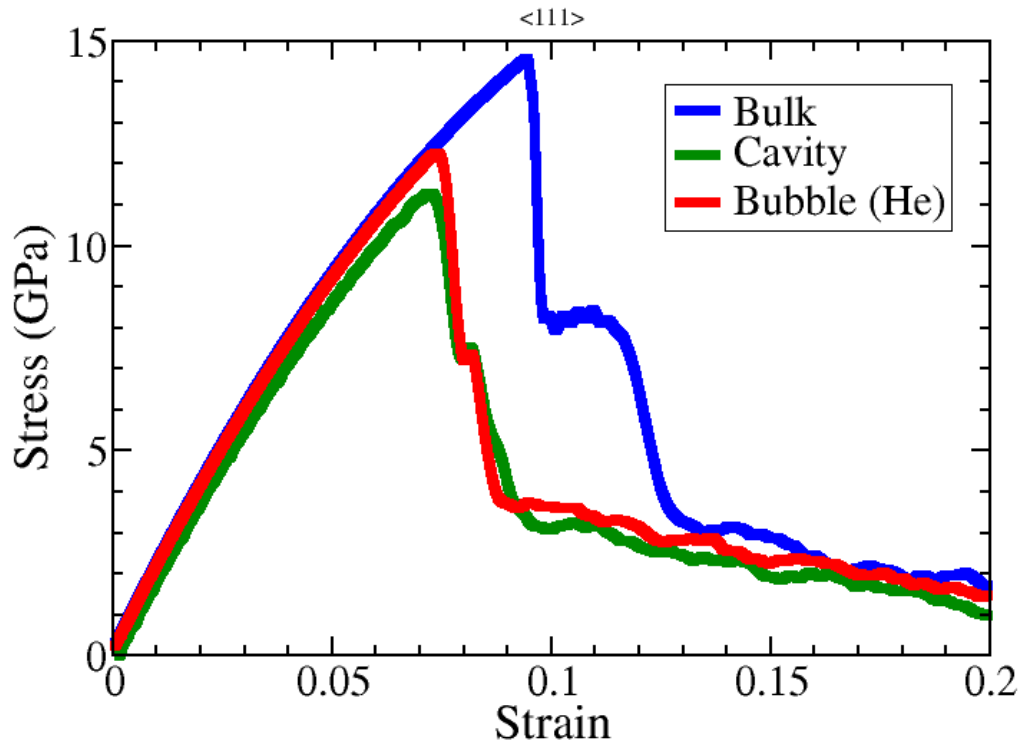
bubble : \varnothing 2 nm, He density in bubble = 4×10^{-2} mol/cm³, Pressure = 500 MPa

strain



- Phase transition at the onset of the crack
- Crack initiates systematically at the bubble surface

MECHANICAL BEHAVIOR - WITH HE BUBBLES



■ ϵ_C , σ_C , and G_C decrease with cavity and He bubble
 ⇒ Easy fracture initiation

■ Decrease of phase transformation
 $G_C \sim G_C(\text{Griffith})$

■ $G_C \text{ bubble} \geq G_C \text{ cavity}$

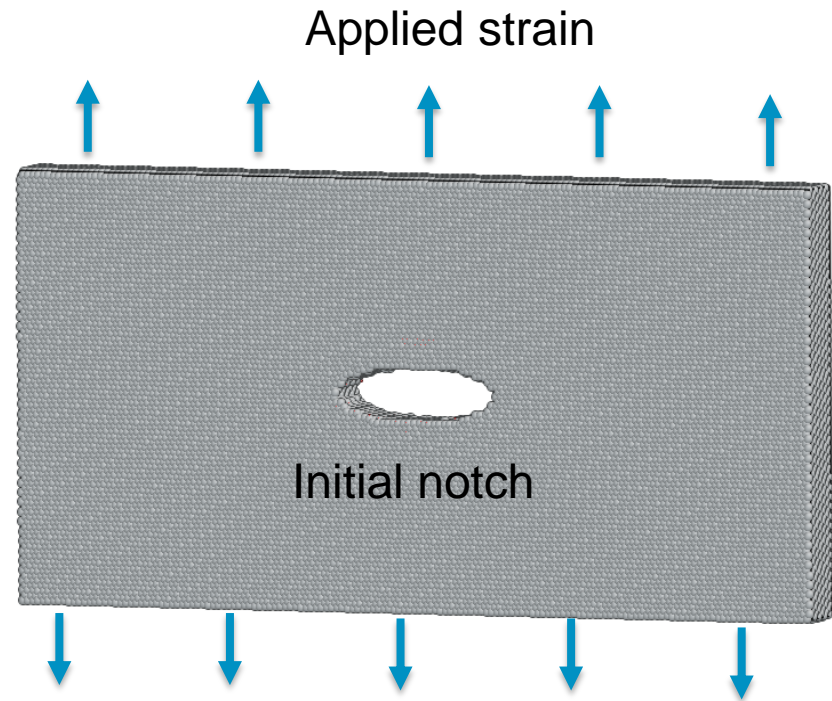
Strain orientation	G_C (J/m ²)		
	Bulk	Cavity	He bubble
<100>	12.73	7.21	7.32
<110>	9.54	5.96	6.76
<111>	9.51	5.62	6.08

- I. **Mechanical behavior in Bulk**
- II. **Mechanical behavior with Point Defects**
- III. **Mechanical behavior with He Bubbles**
- IV. **Crack propagation**
- V. **Summary**

CRACK PROPAGATION - METHOD

- Strip geometry
- Create initial notch
- Relaxation NPT 300 K, 20 ps
- Impose deformation along $\langle 111 \rangle$ direction
- Strain-rate $10^9/s$

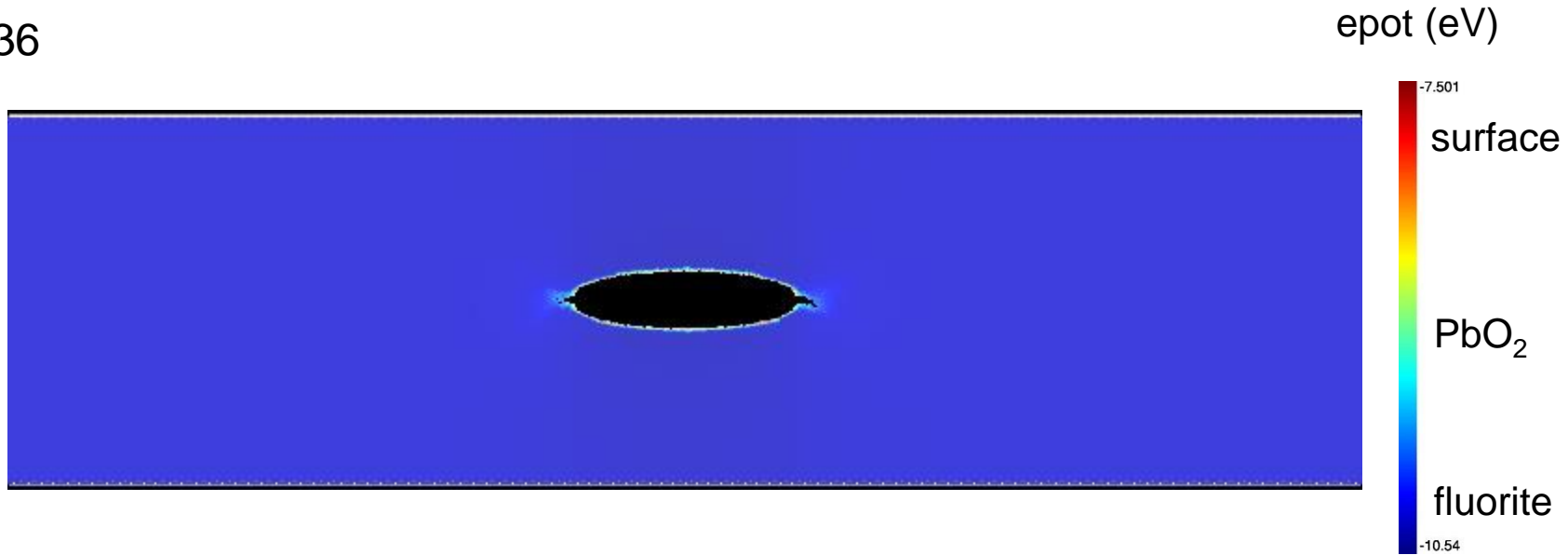
- Outputs: microstructure evolution, local energies, local stresses



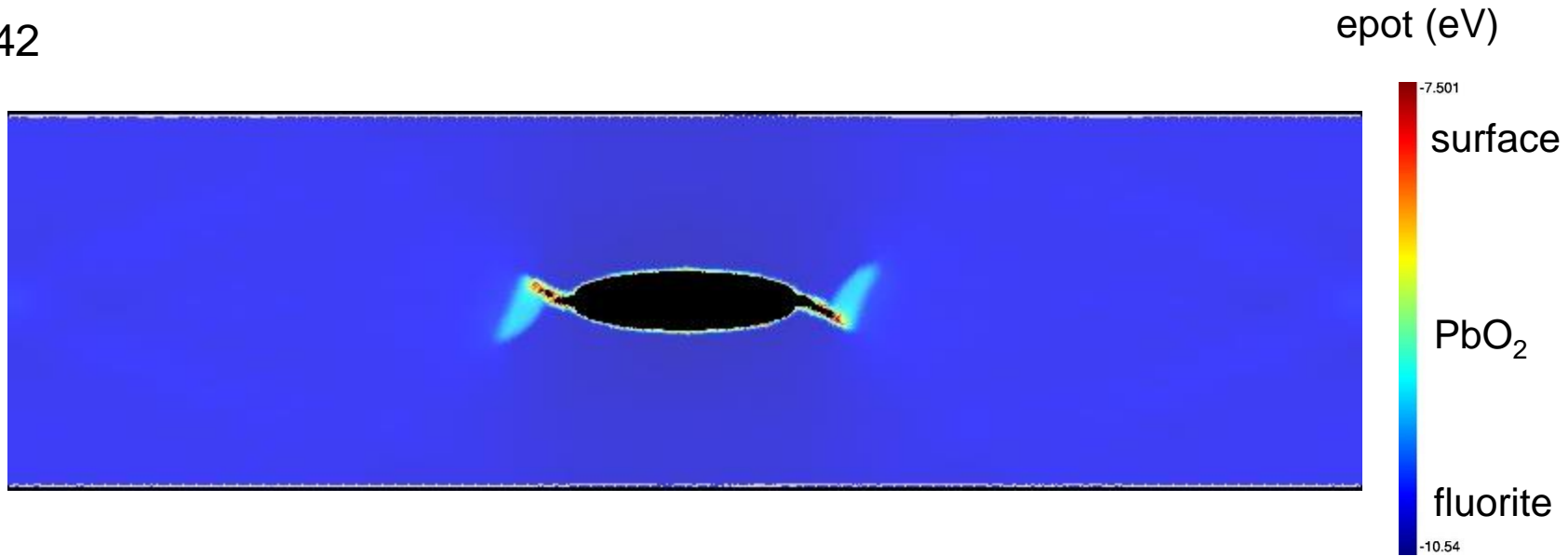
Strip geometry:
 $241 \times 65 \times 4$ nm

Initial notch:
Ellipse 40×8 nm

$\varepsilon = 0.036$

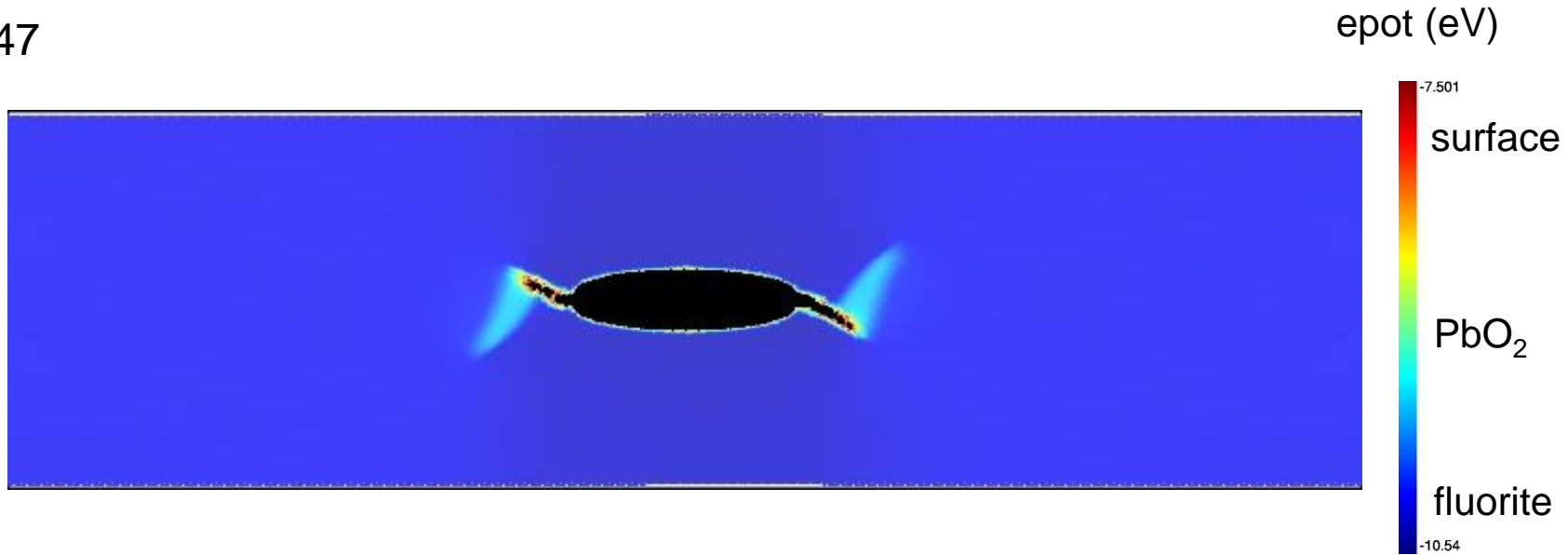


$\varepsilon = 0.042$



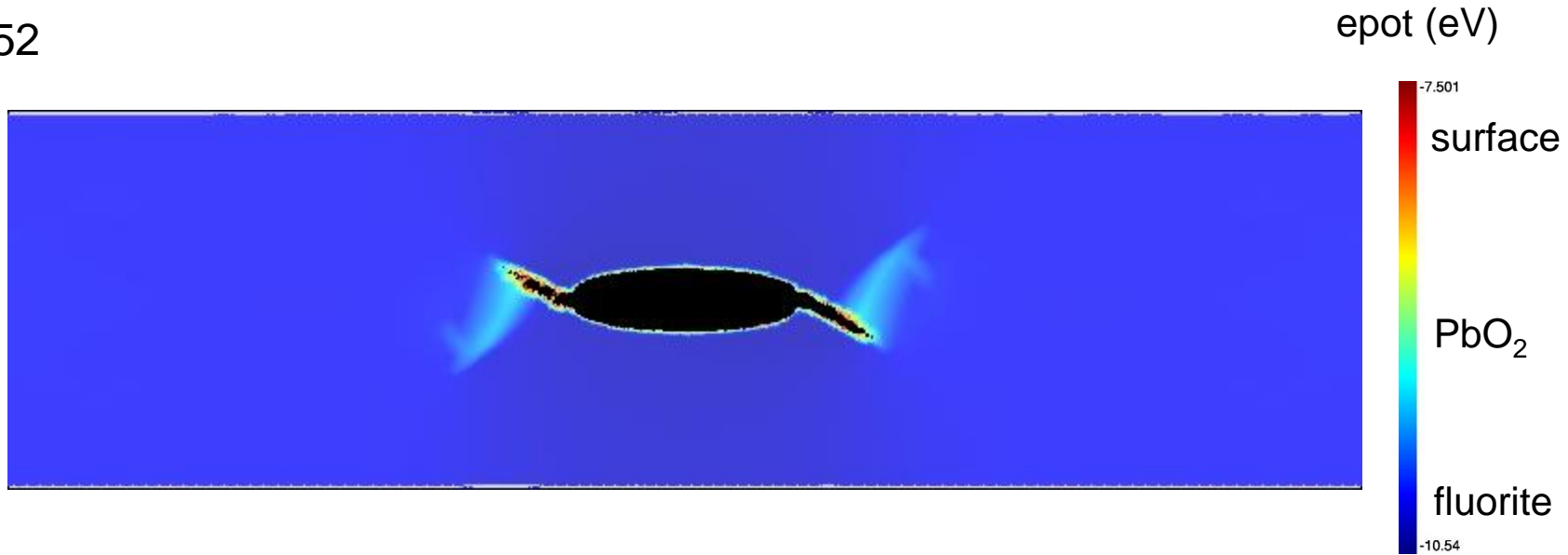
CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.047$



CRACK PROPAGATION - IN BLUK

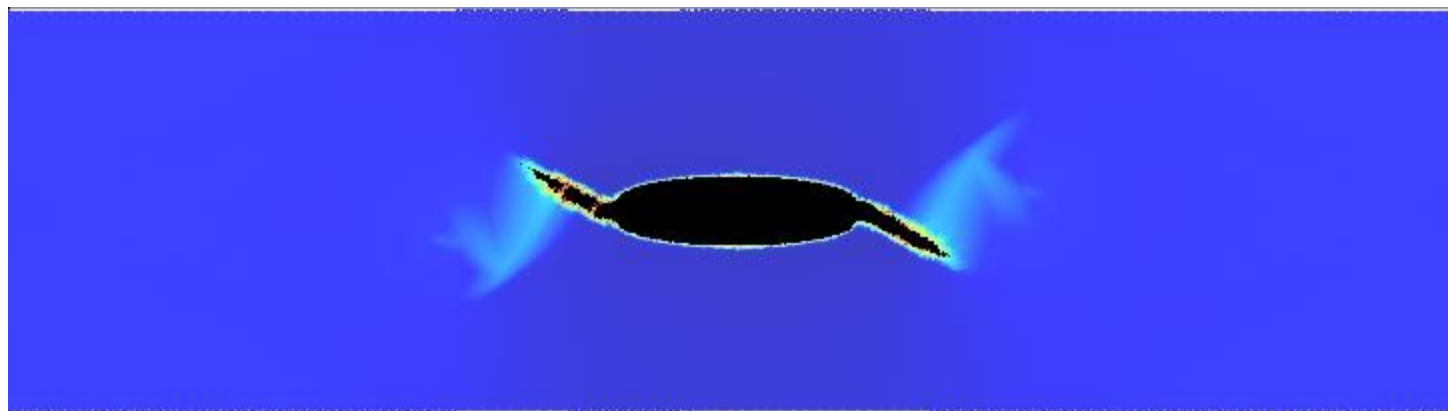
$\varepsilon = 0.052$



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.058$

epot (eV)



-7.501

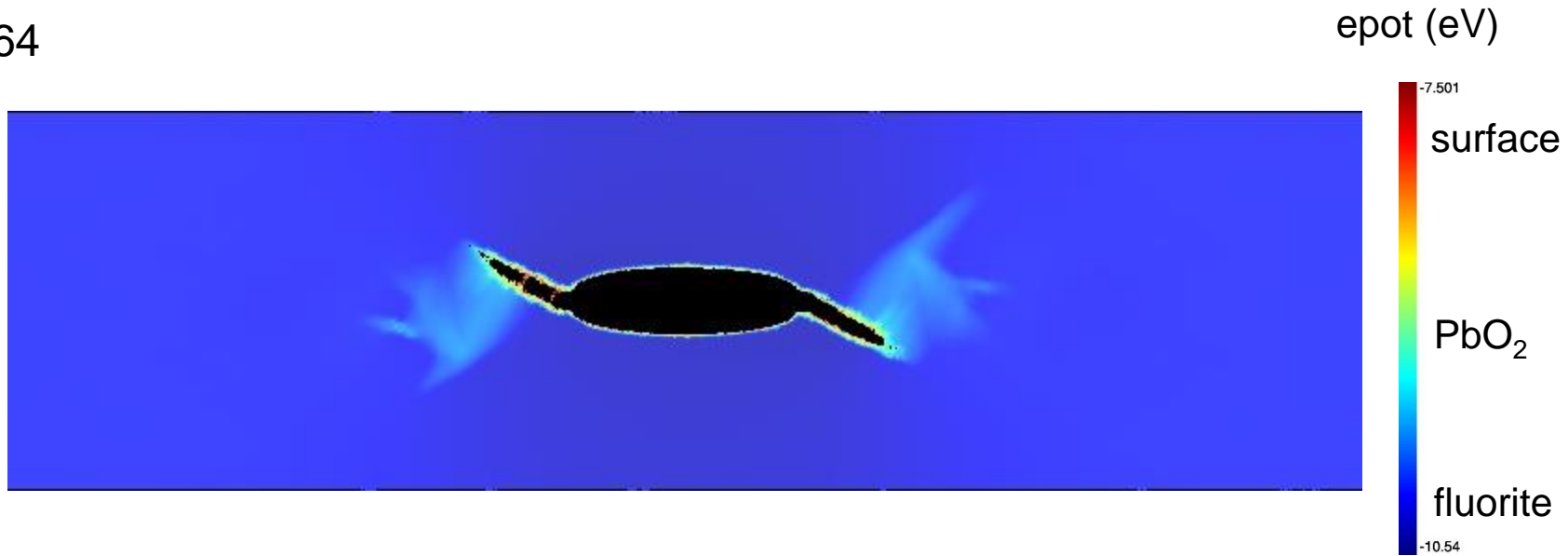
surface

PbO₂

fluorite

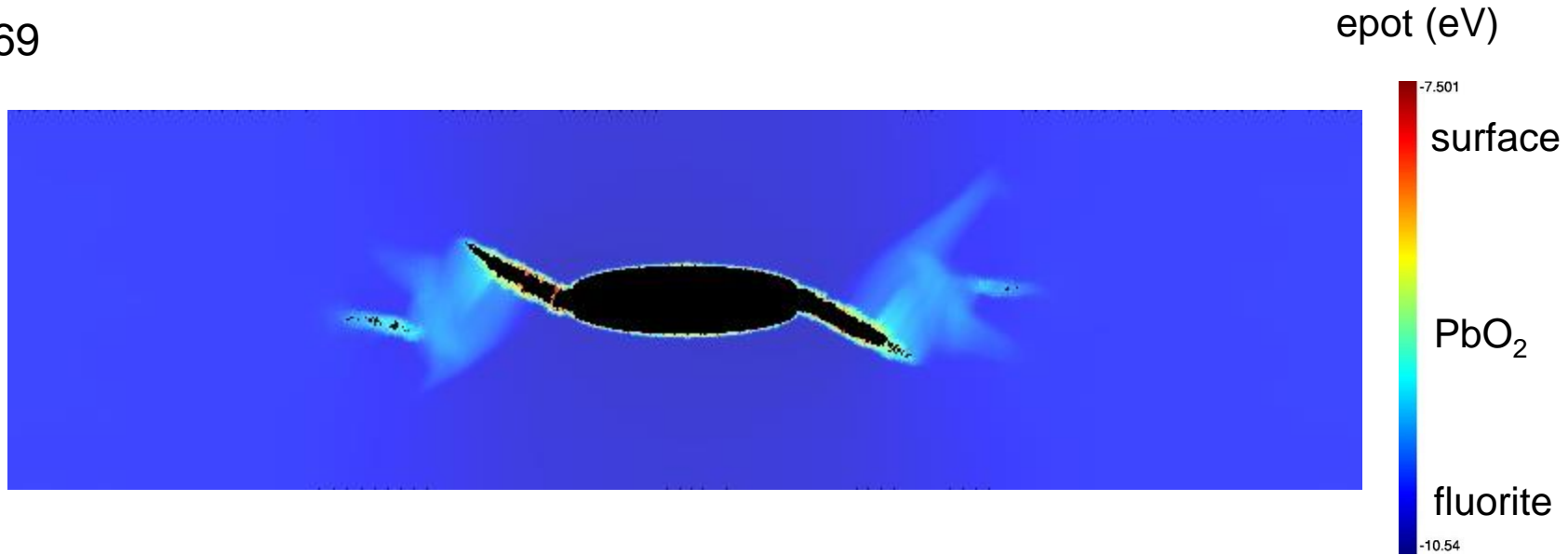
-10.54

$\varepsilon = 0.064$



CRACK PROPAGATION - IN BLUK

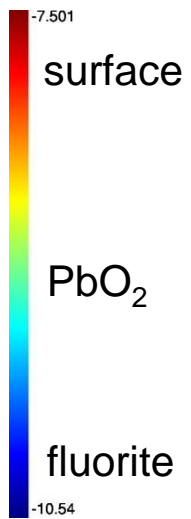
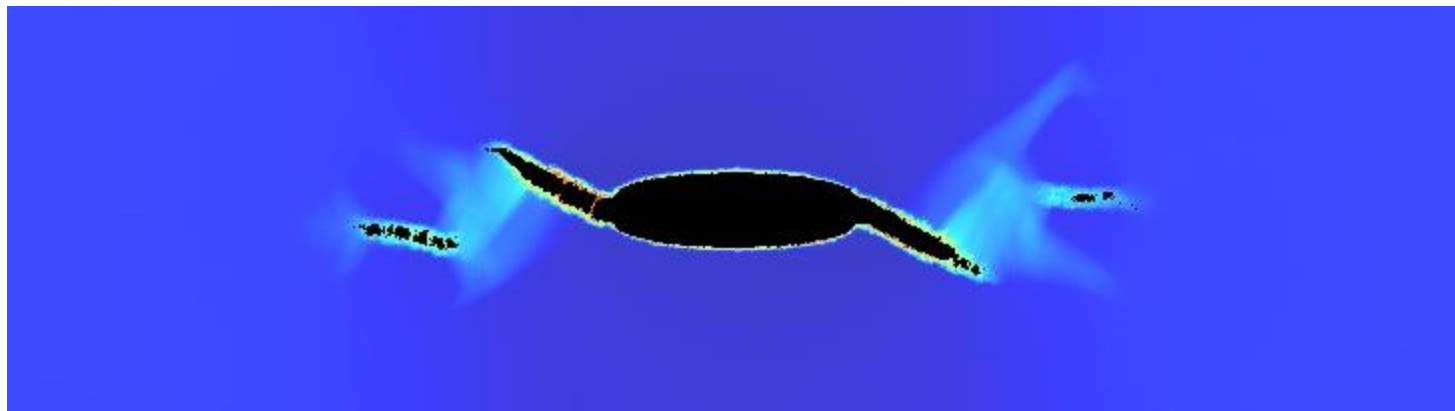
$\varepsilon = 0.069$



CRACK PROPAGATION - IN BLUK

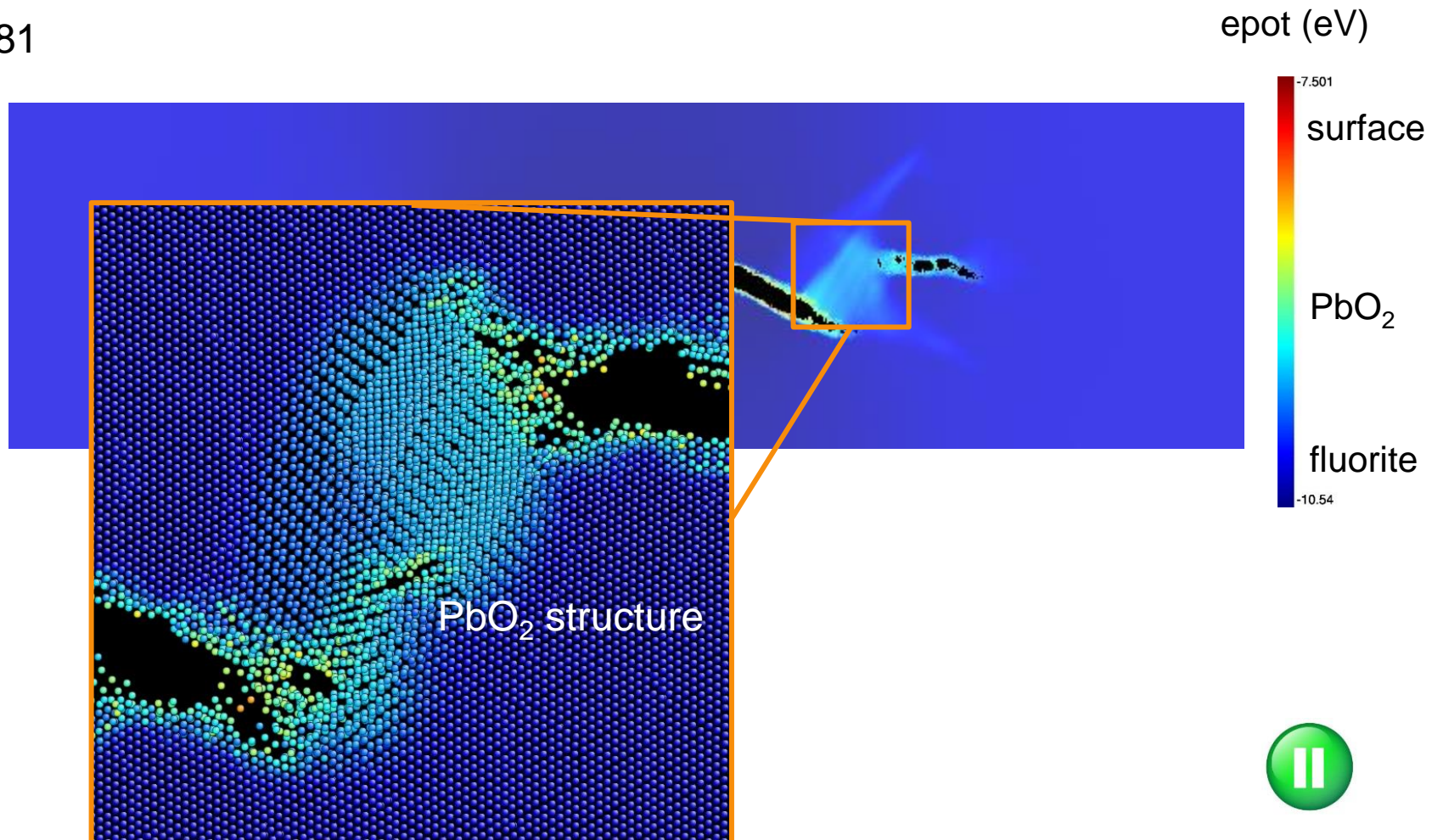
$\varepsilon = 0.075$

epot (eV)



CRACK PROPAGATION - IN BLUK

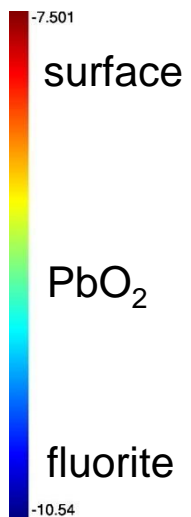
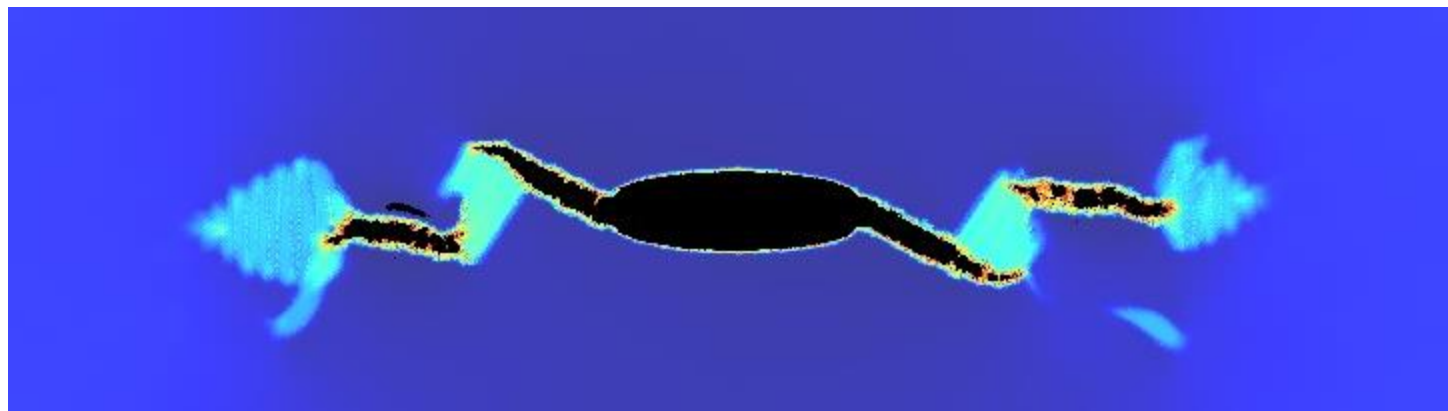
$\varepsilon = 0.081$



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.087$

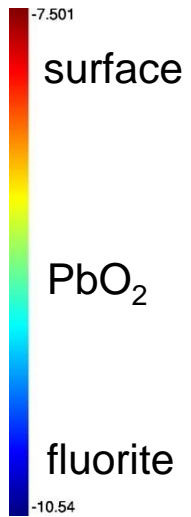
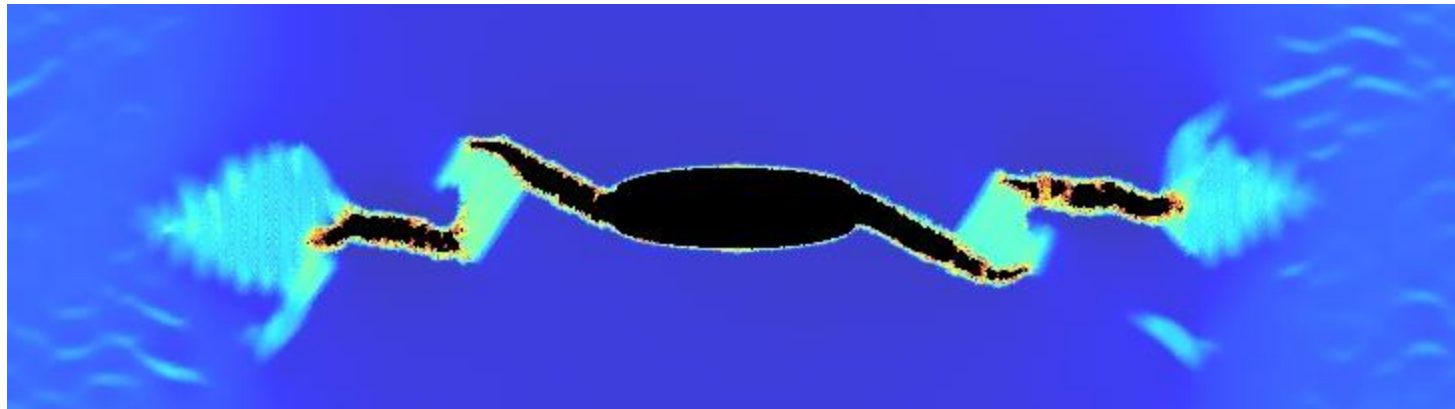
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.093$

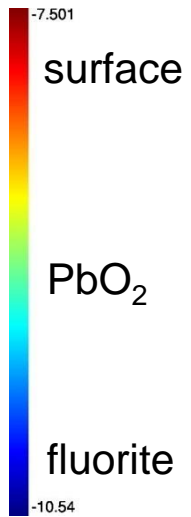
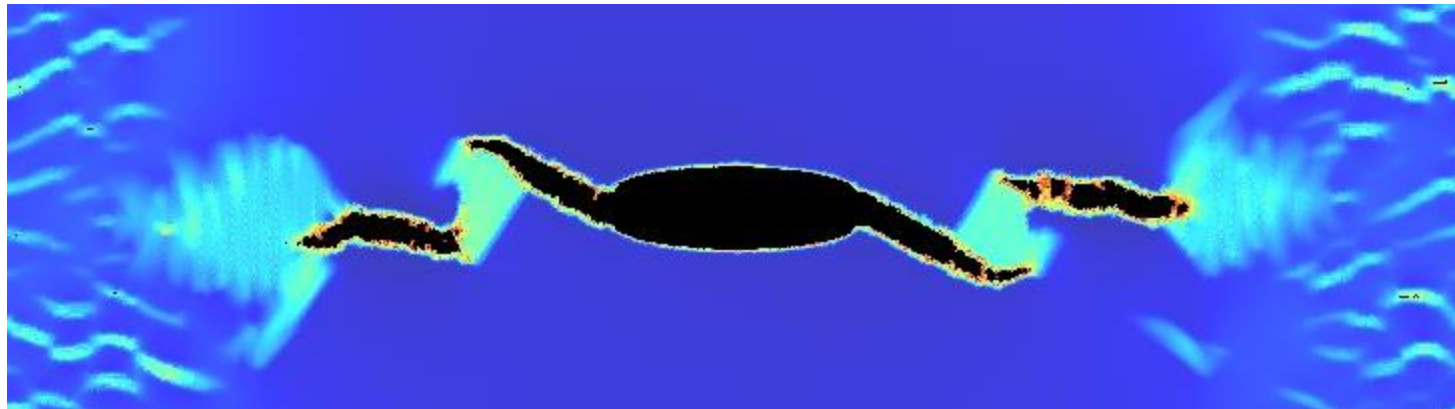
epot (eV)



CRACK PROPAGATION - IN BLUK

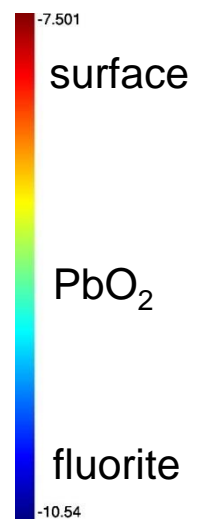
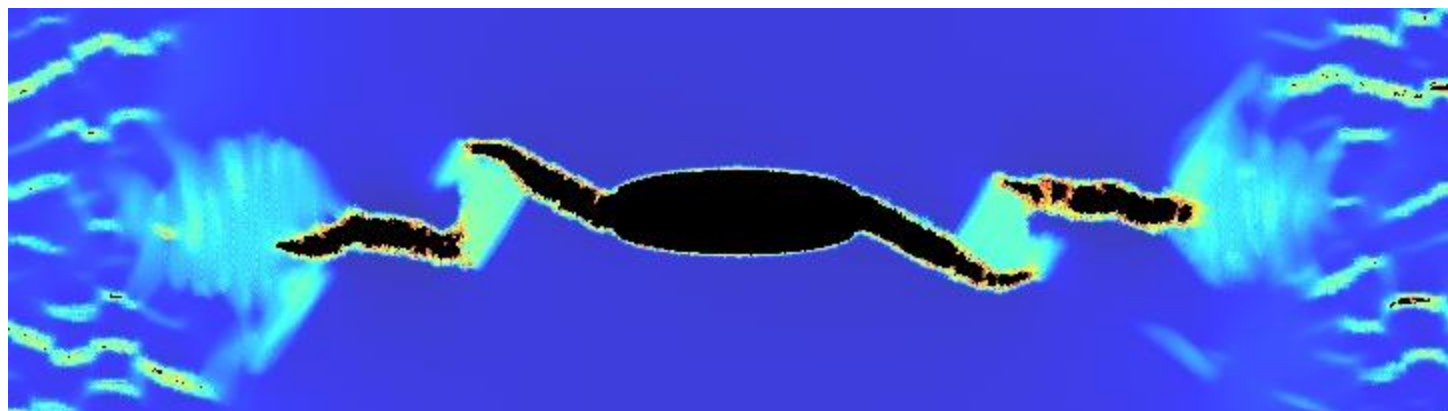
$\varepsilon = 0.098$

epot (eV)



$\varepsilon = 0.104$

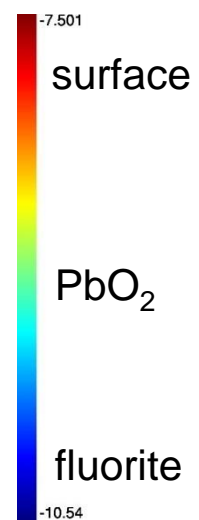
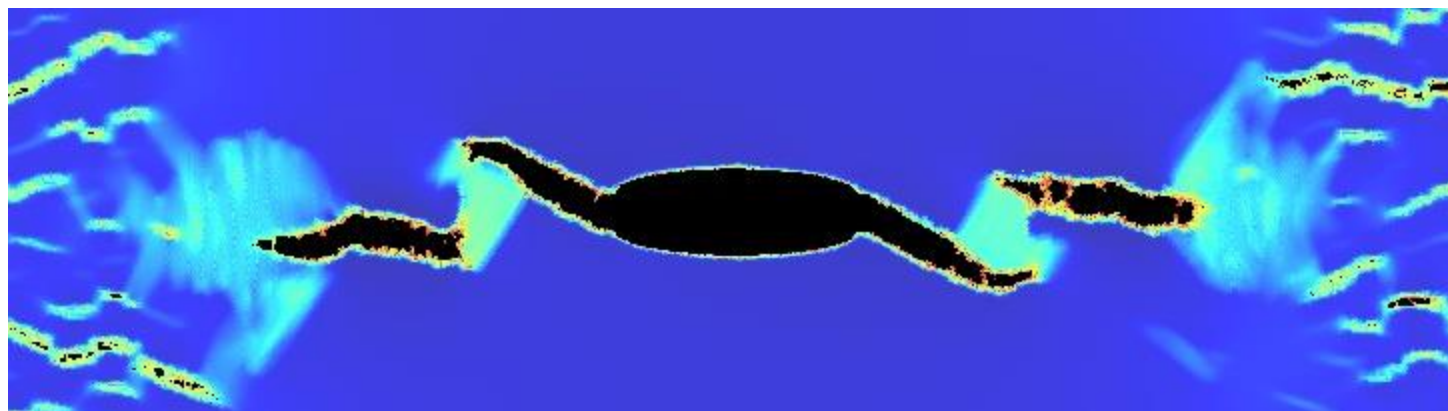
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.111$

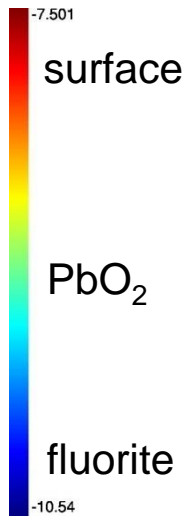
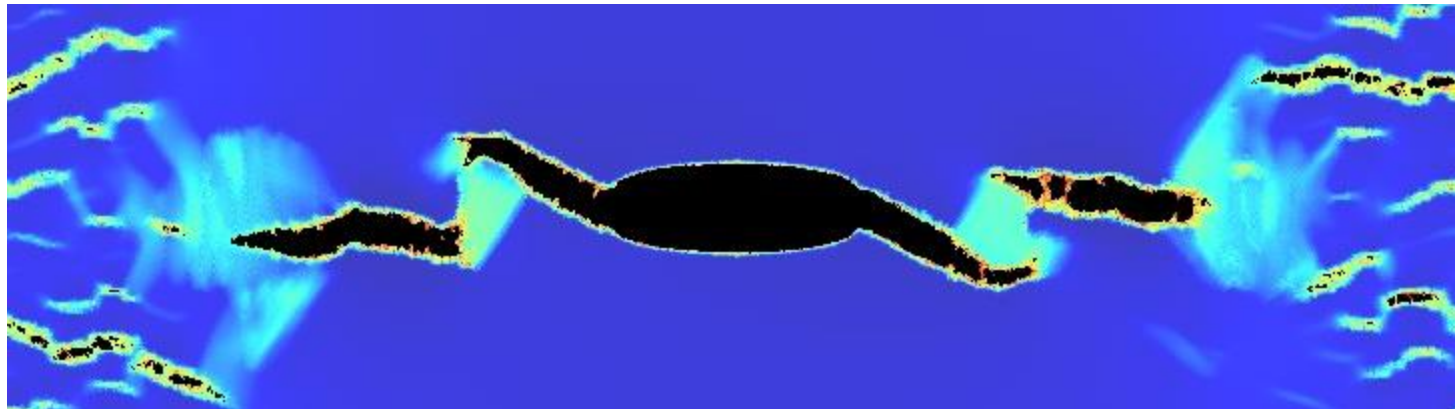
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.117$

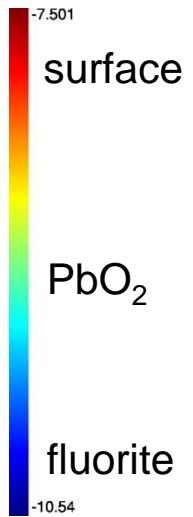
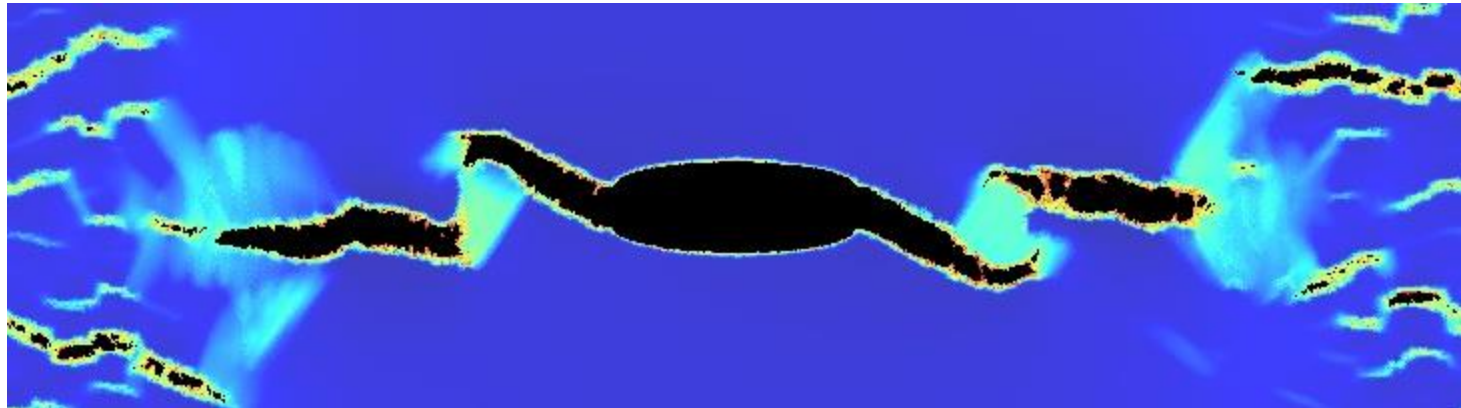
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.123$

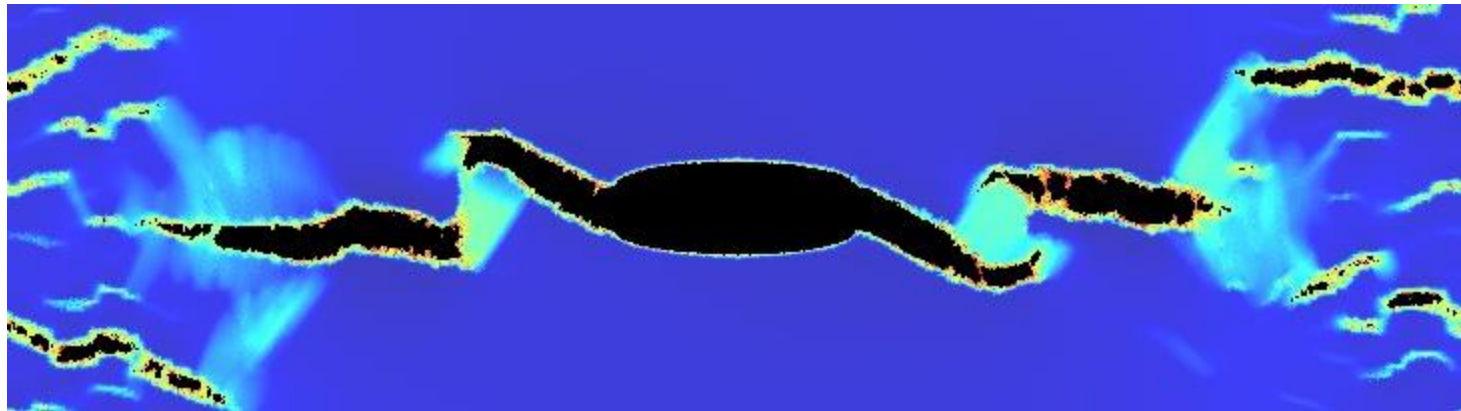
epot (eV)



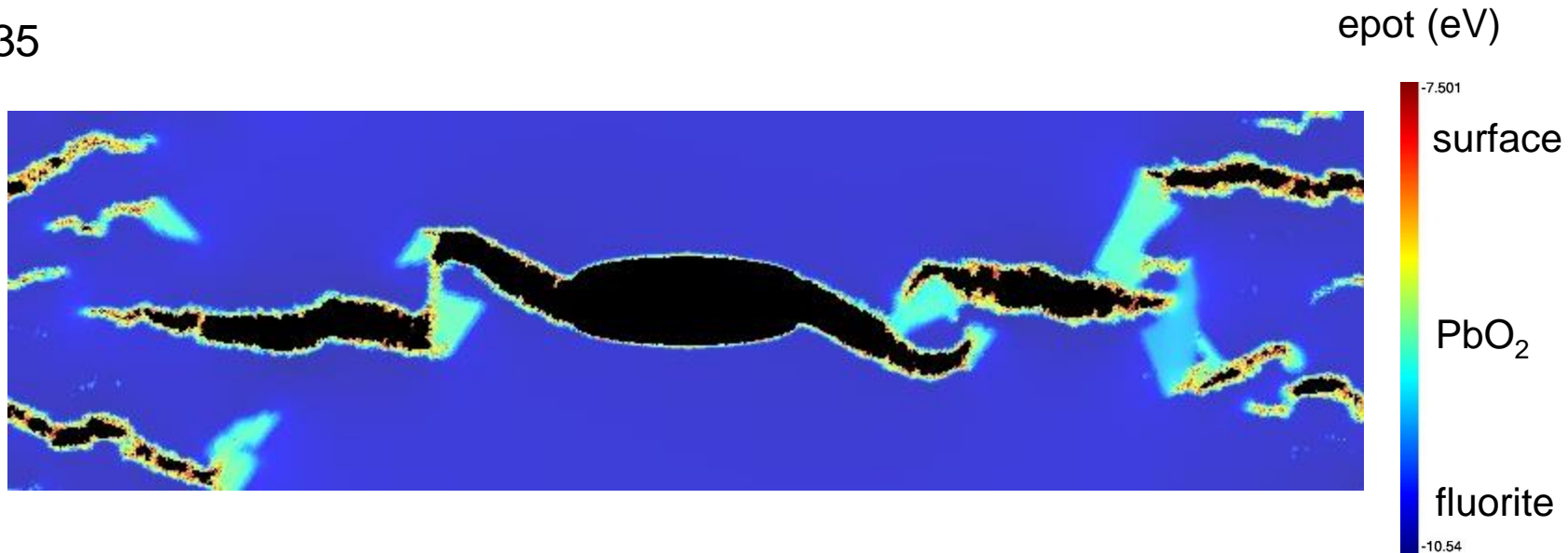
CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.129$

epot (eV)



$\varepsilon = 0.135$



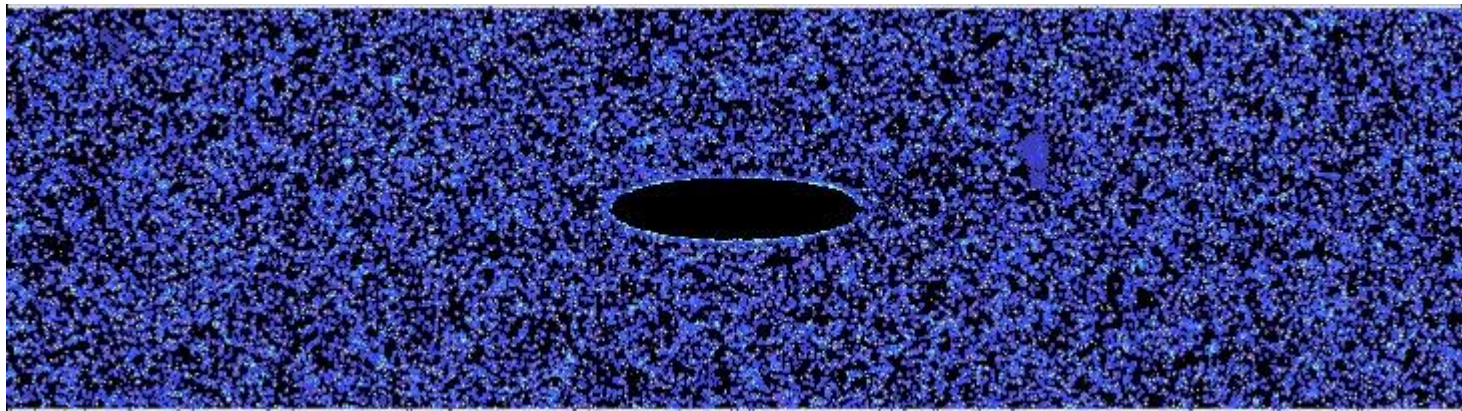
- Crack propagates by **cleavage** in the PbO_2 structure at the crack tip
- Sub-cracks appear, grow and coalesce in secondary phase

CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.036$

1.8% Frenkel pairs randomly distributed

epot (eV)



surface

PbO₂

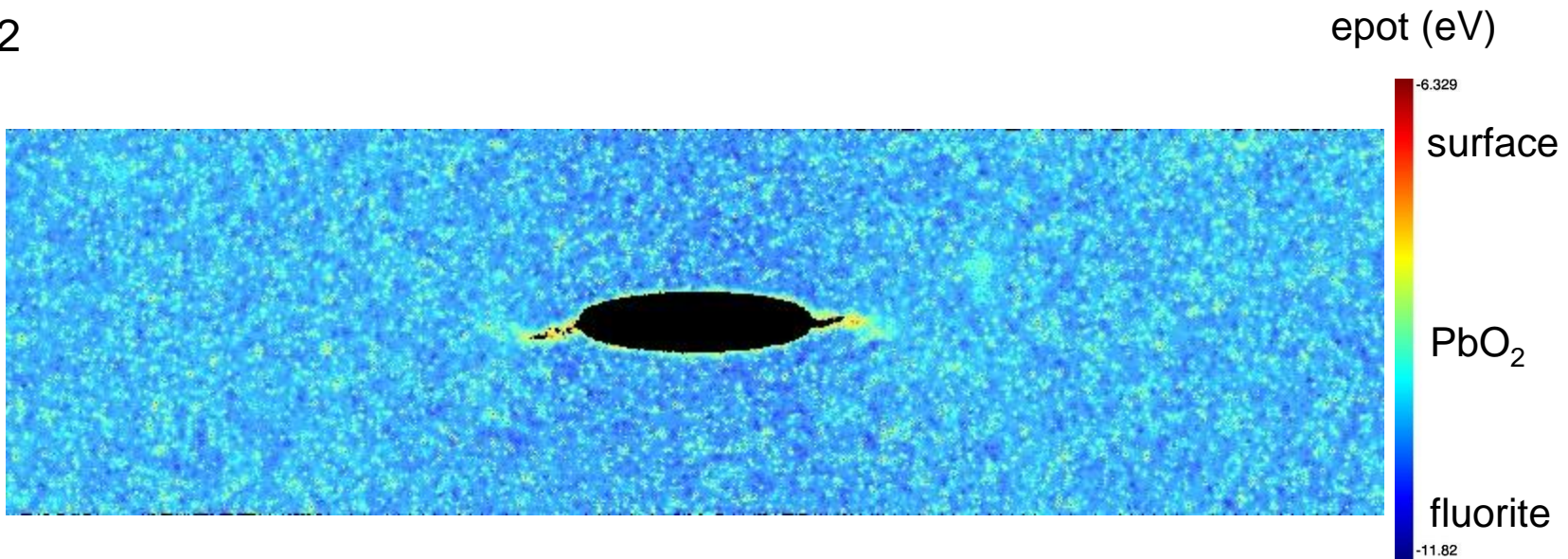
fluorite

-11.82

-6.329

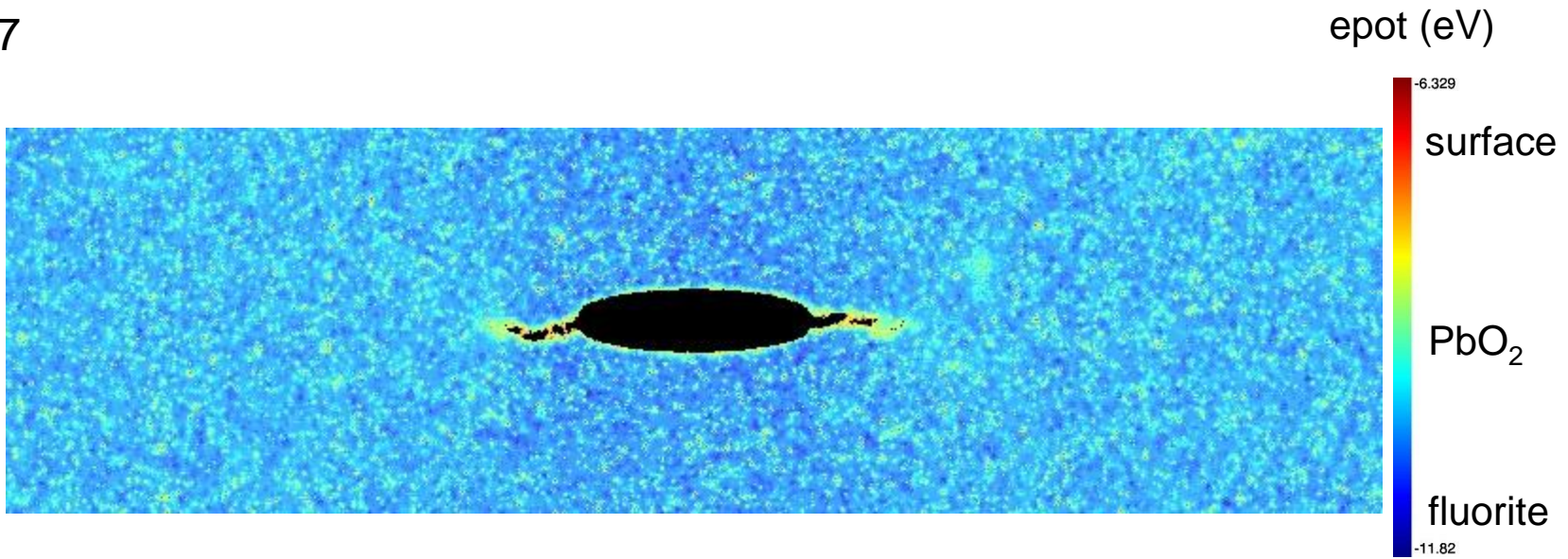
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.042$



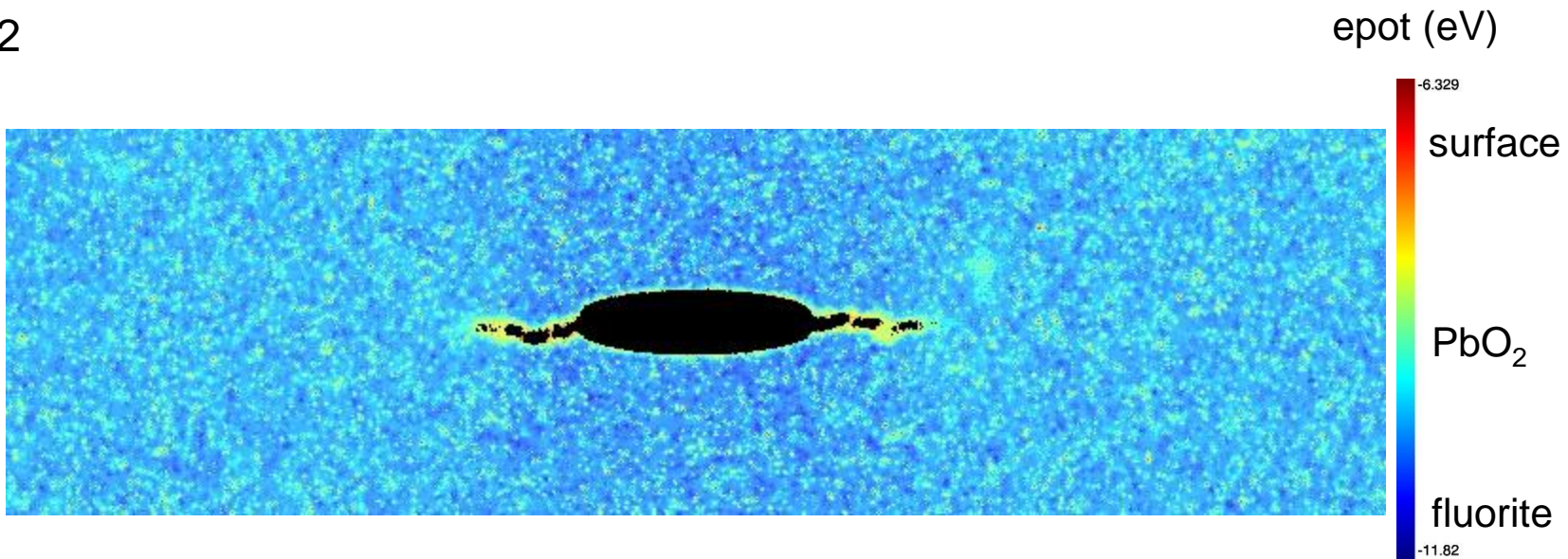
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.047$



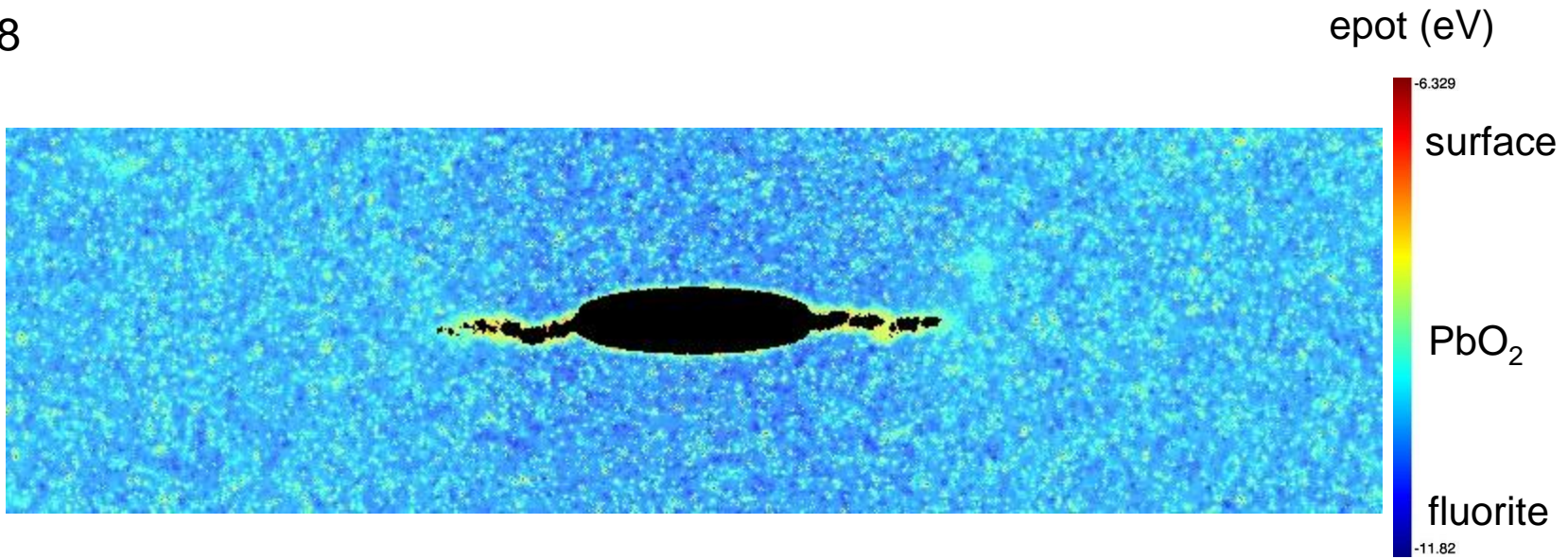
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.052$



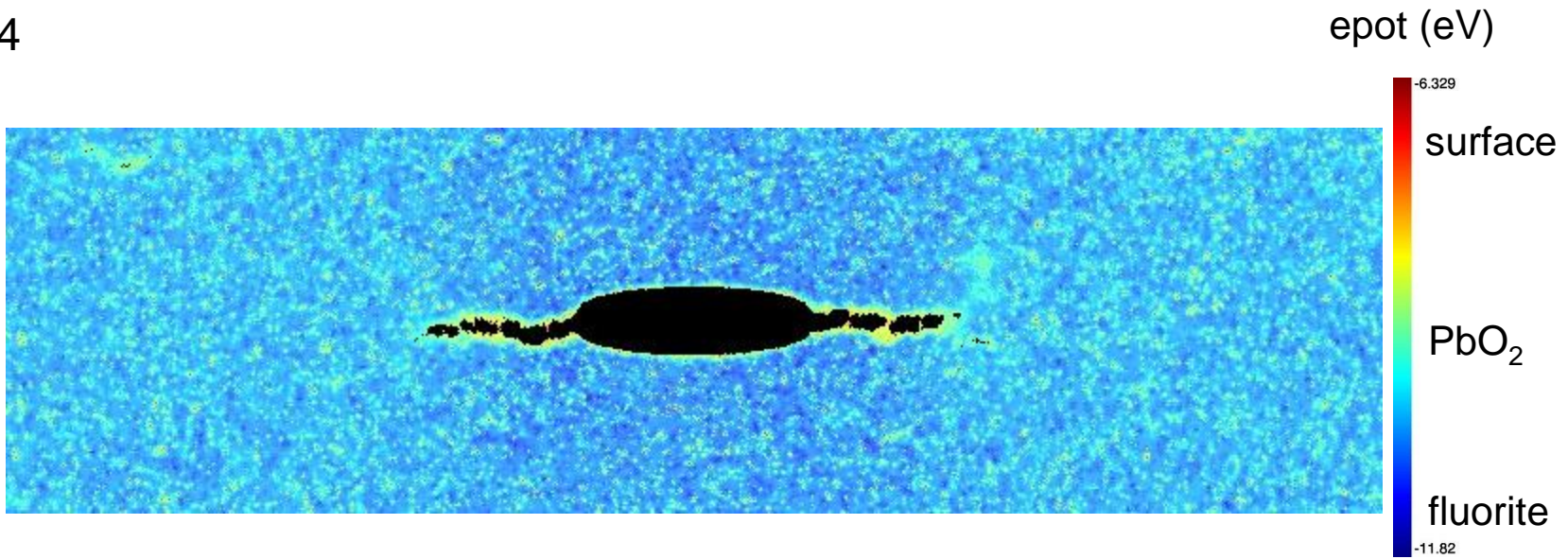
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.058$



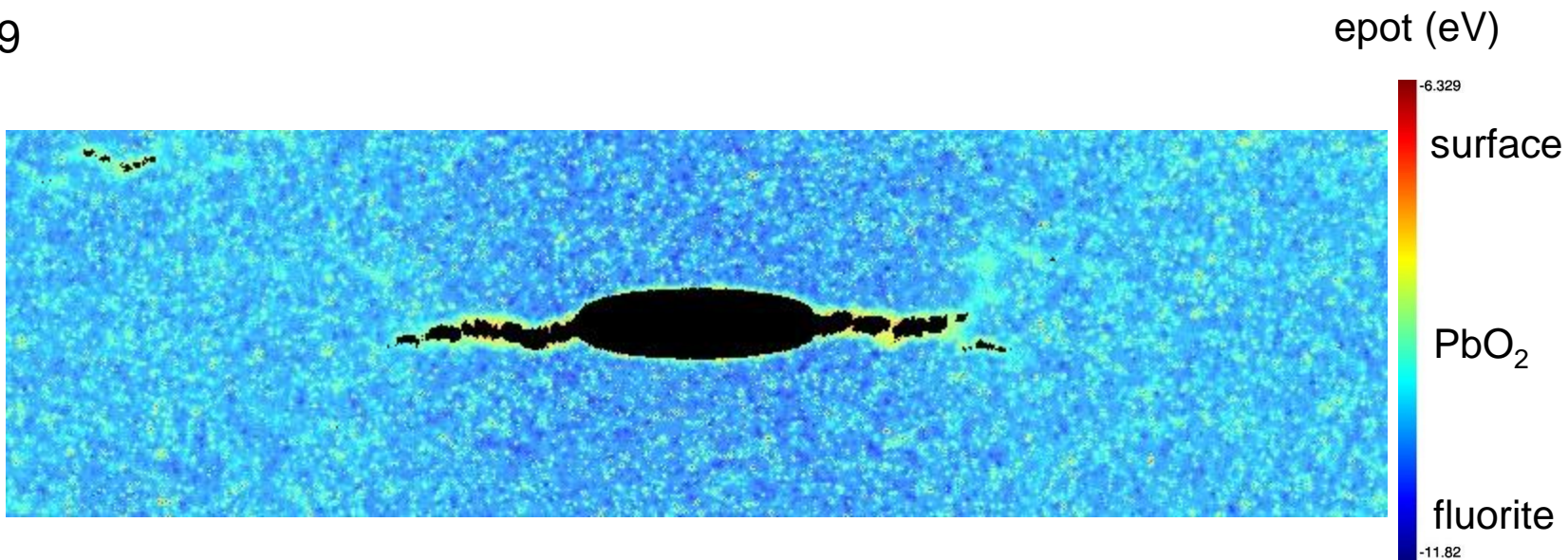
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.064$



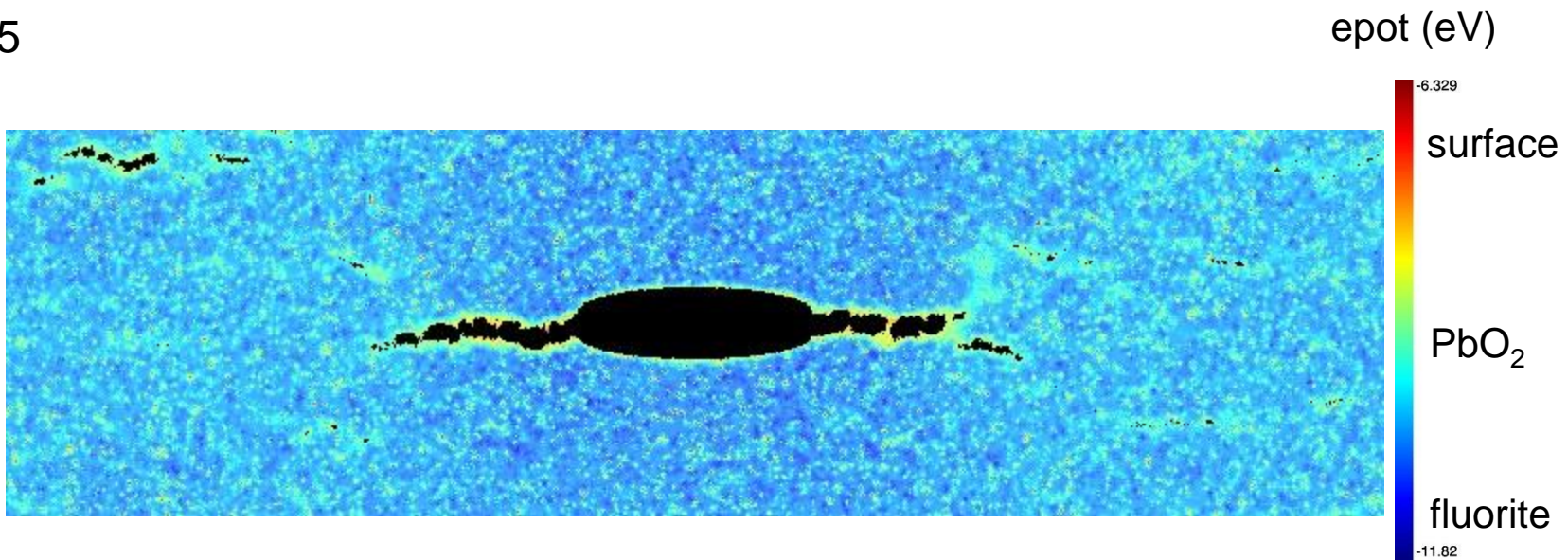
CRACK PROPAGATION - WITH POINT DEFECTS

$$\varepsilon = 0.069$$



CRACK PROPAGATION - WITH POINT DEFECTS

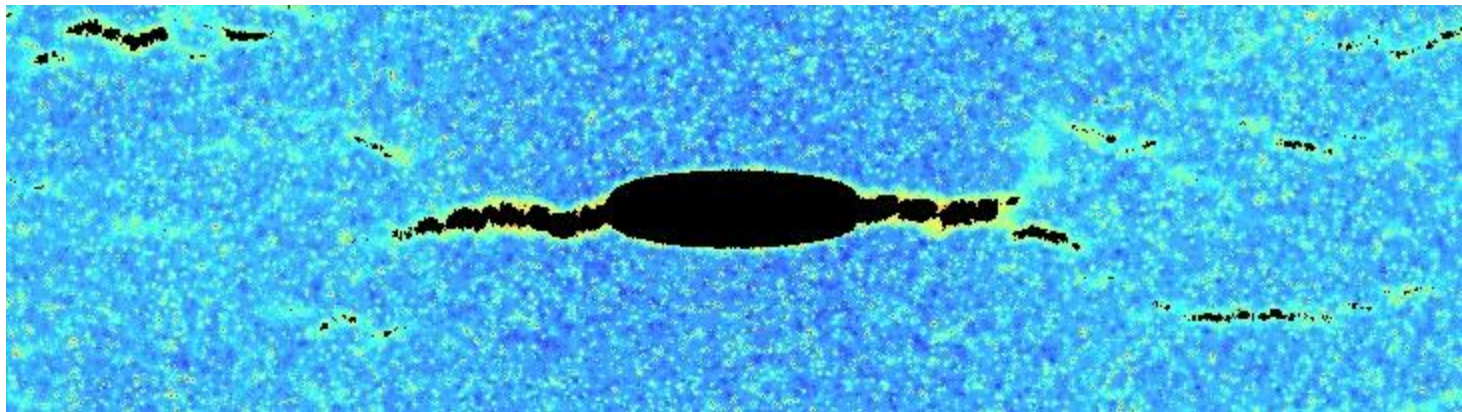
$\varepsilon = 0.075$



CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.081$

epot (eV)



-6.329

surface

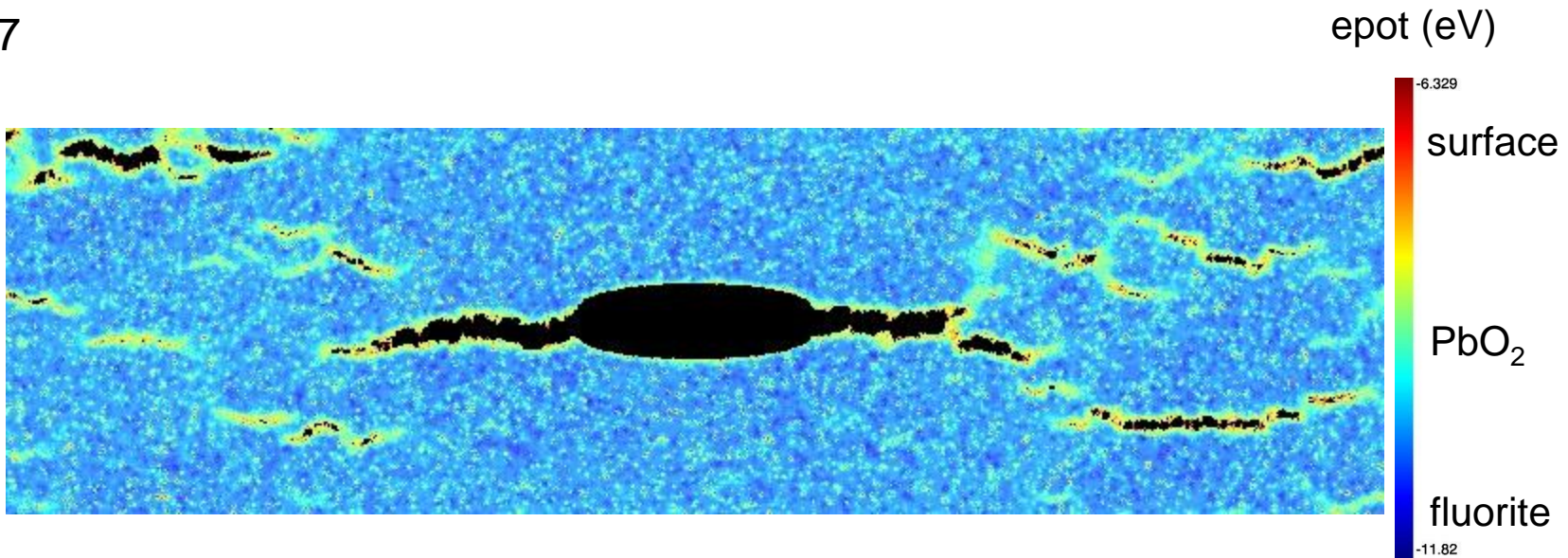
PbO₂

fluorite

-11.82

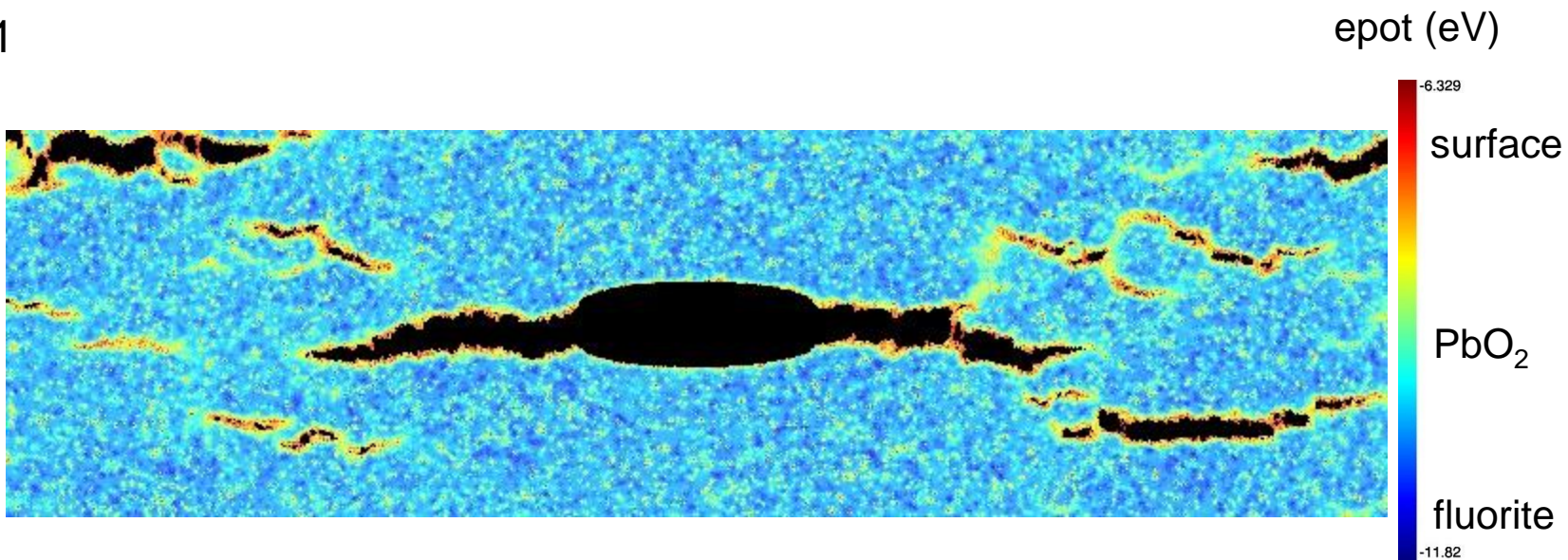
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.087$



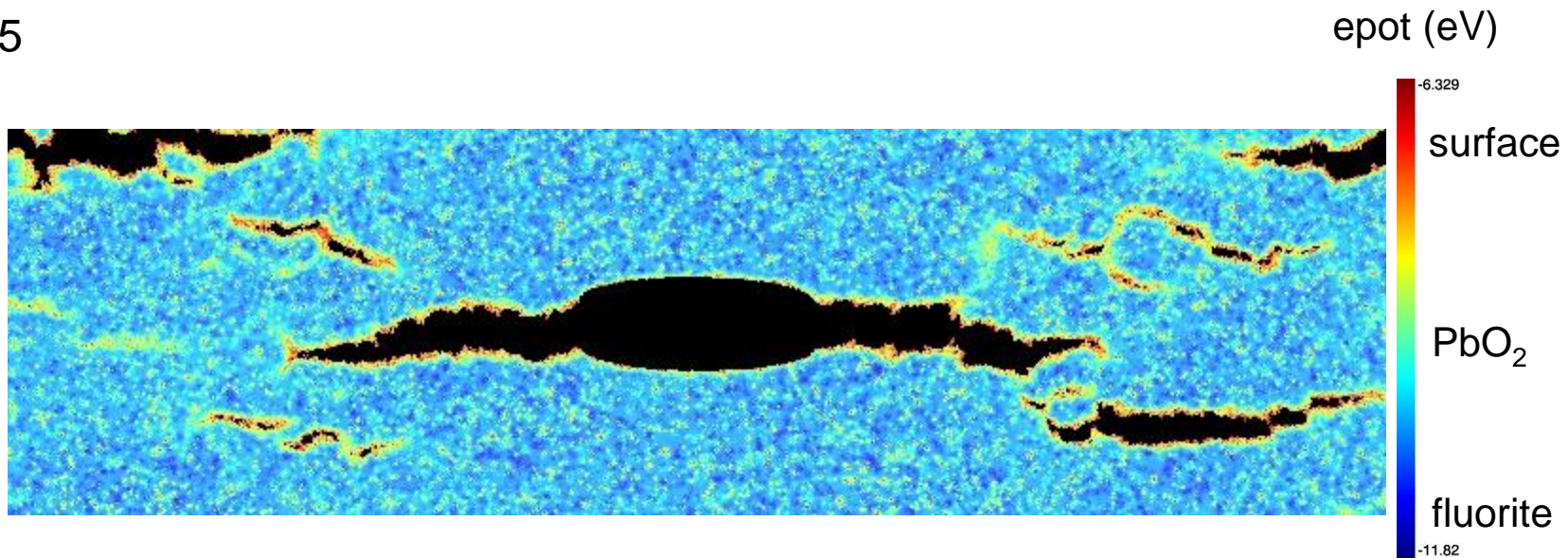
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.111$



CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.135$



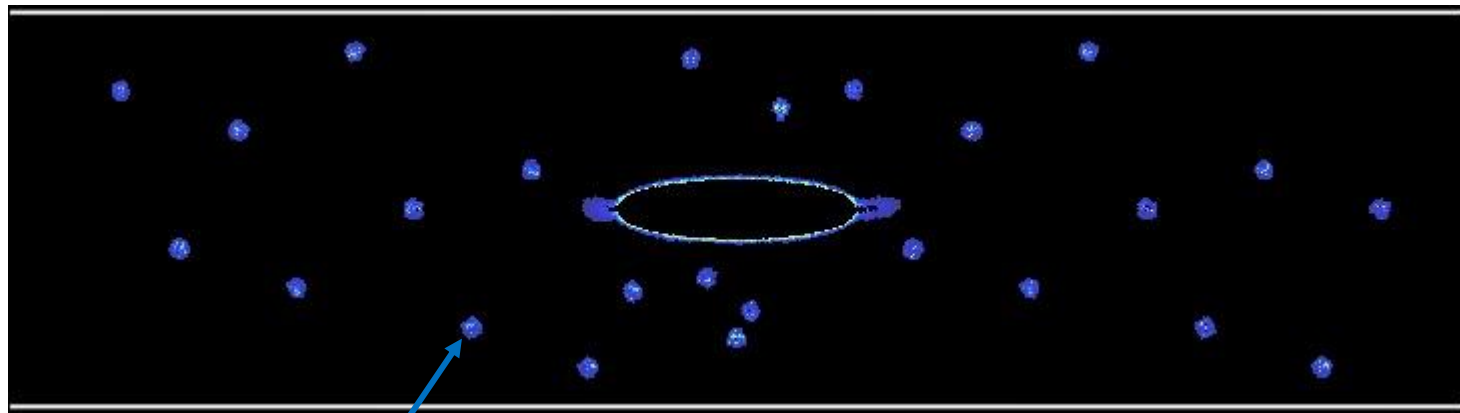
- No phase transition
- Crack propagates by **growth** and **coalescence** of close sub-cracks

CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.036$

$4 \times 10^{23}/\text{m}^3$ He bubbles, 2 nm, with $4 \times 10^{-2} \text{ mol}/\text{cm}^3$ He

epot (eV)



-5.046

surface

PbO₂

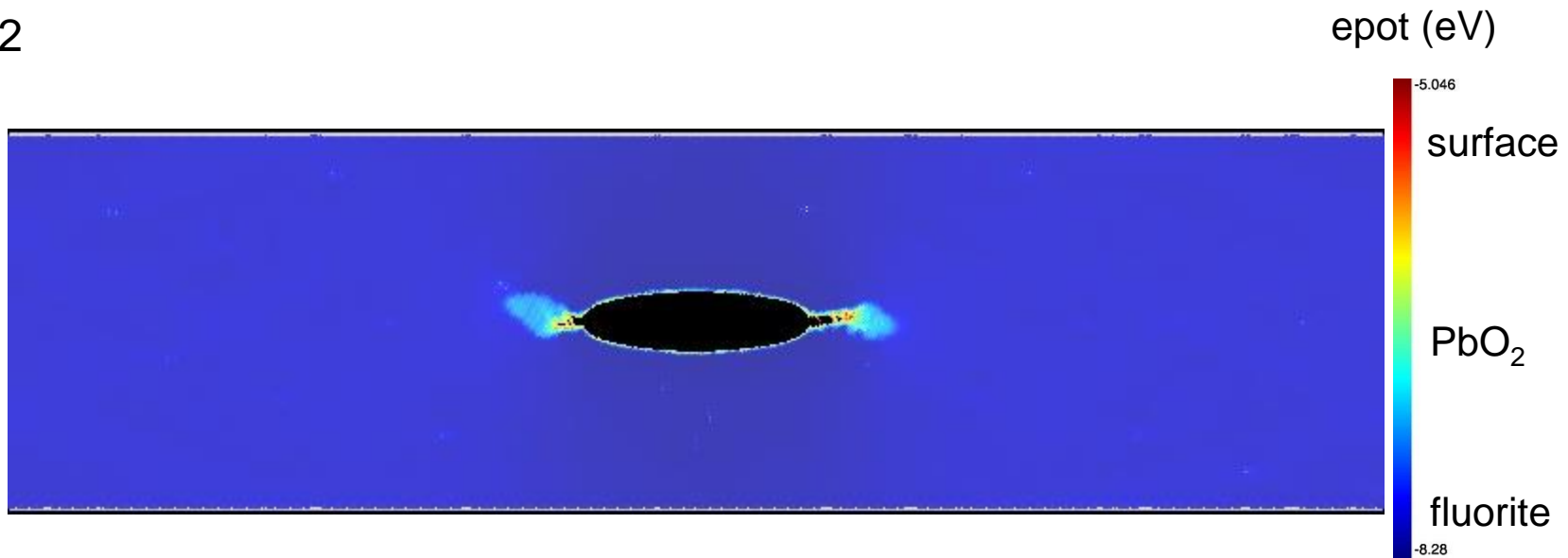
fluorite

-8.28

He bubble

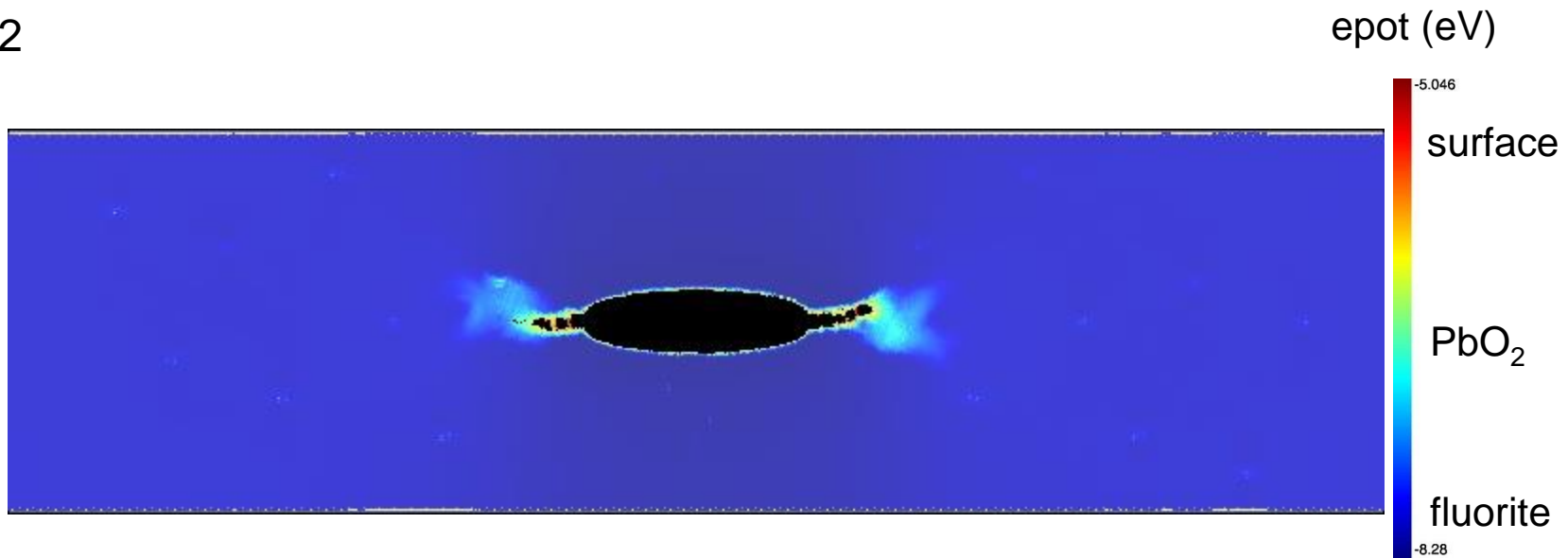
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.042$



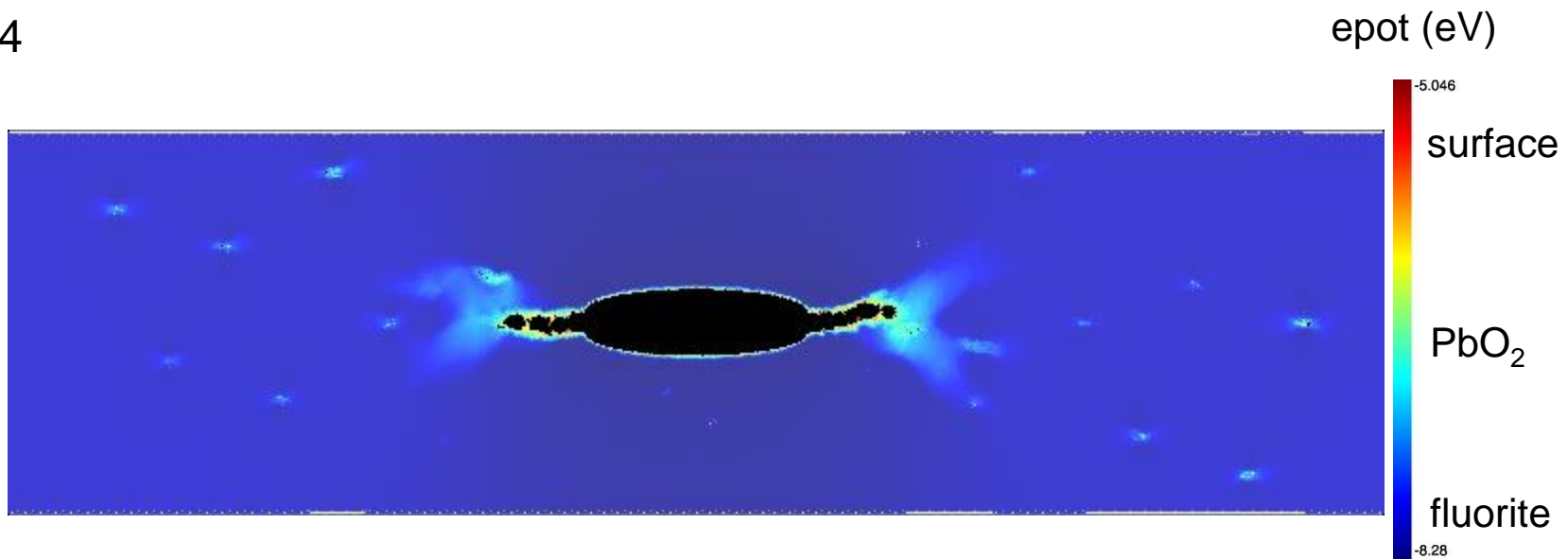
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.052$



CRACK PROPAGATION - WITH HE BUBBLES

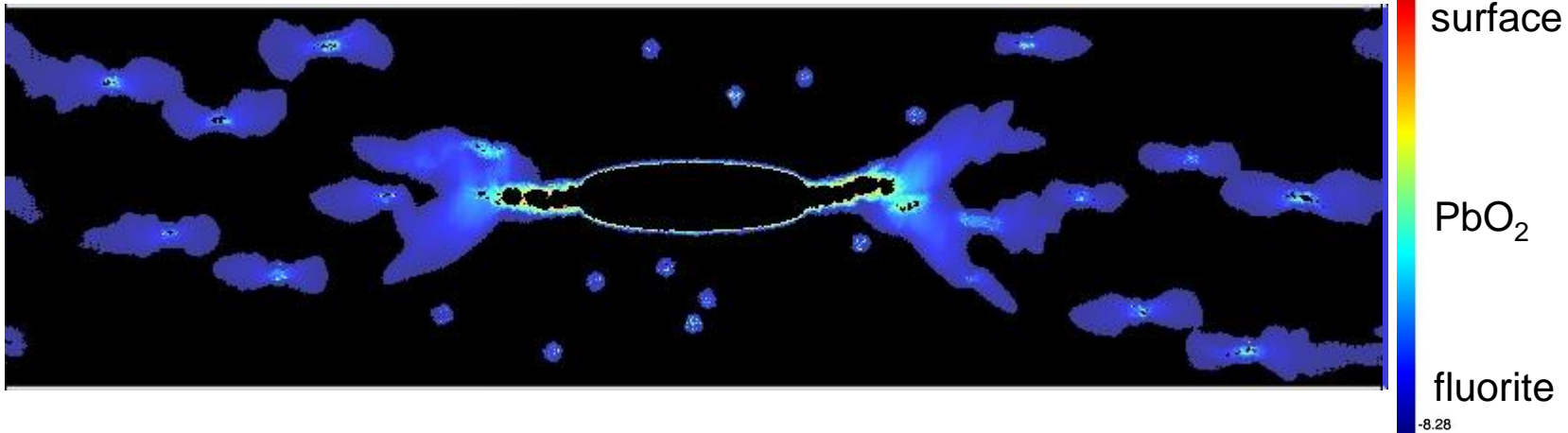
$\varepsilon = 0.064$



CRACK PROPAGATION - WITH HE BUBBLES

$$\varepsilon = 0.069$$

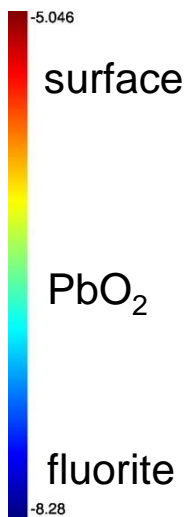
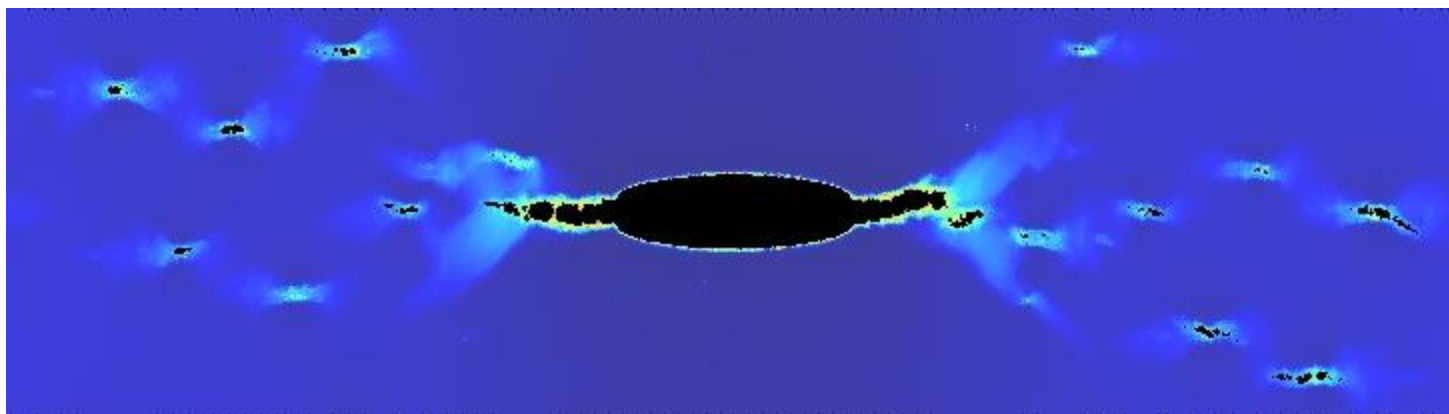
epot (eV)



CRACK PROPAGATION - WITH HE BUBBLES

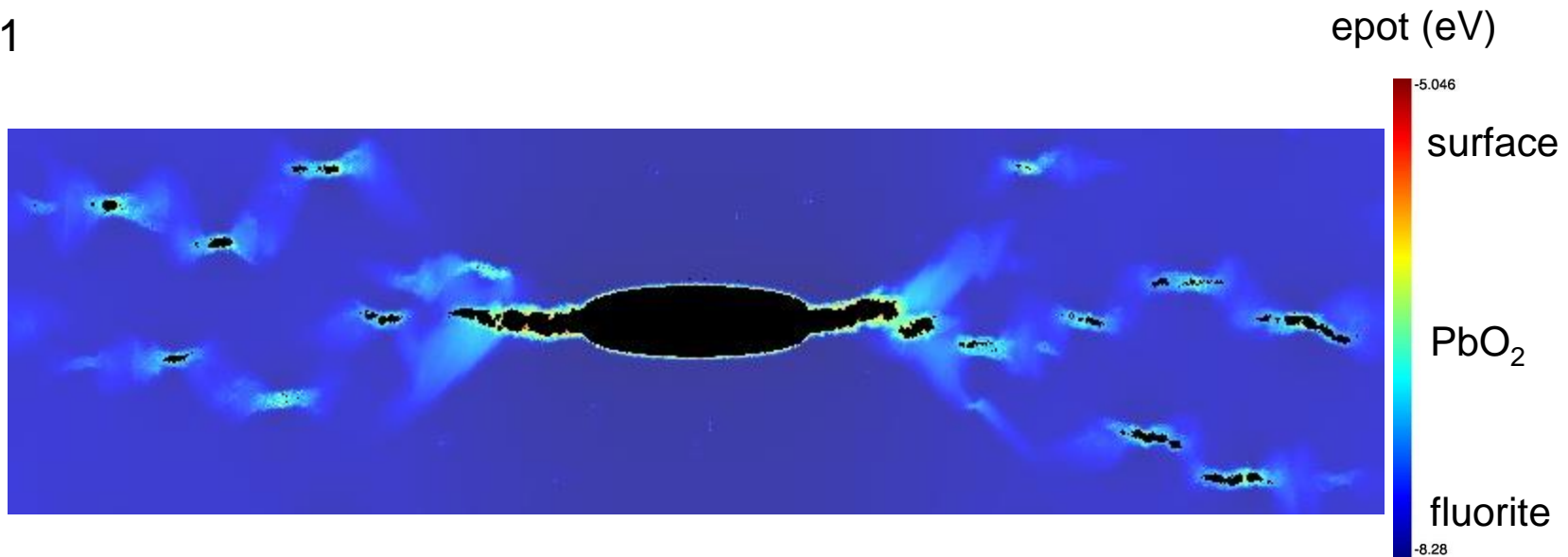
$\varepsilon = 0.075$

epot (eV)



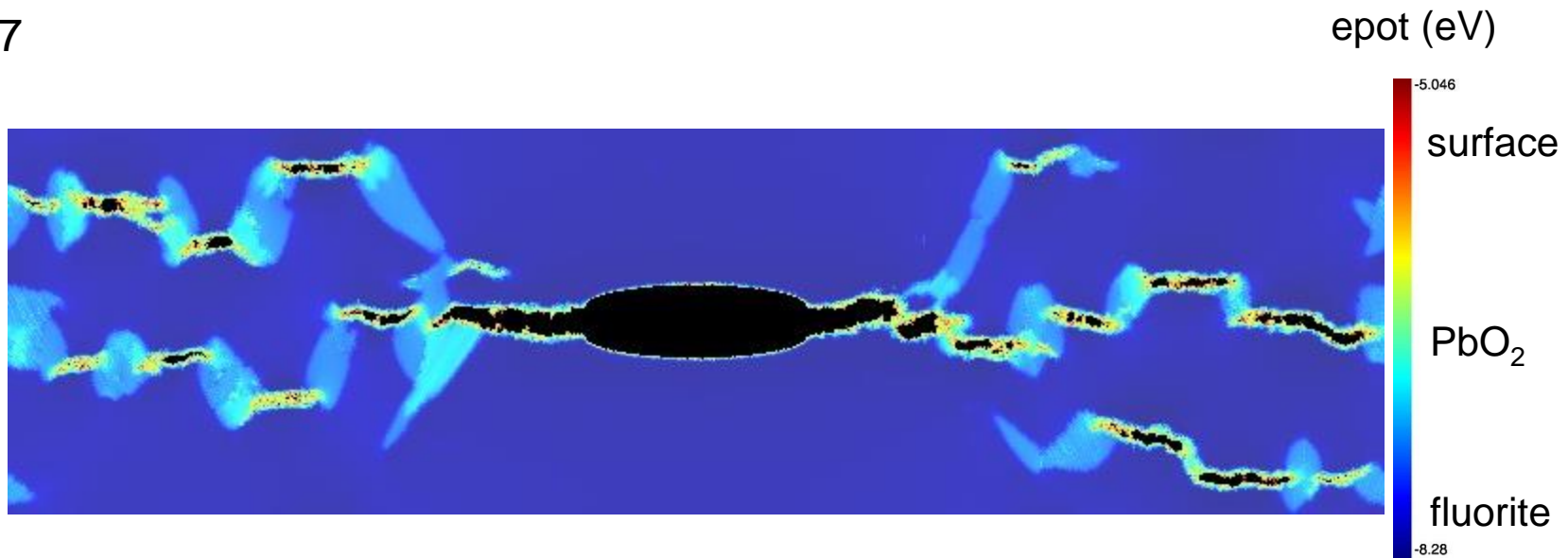
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.081$



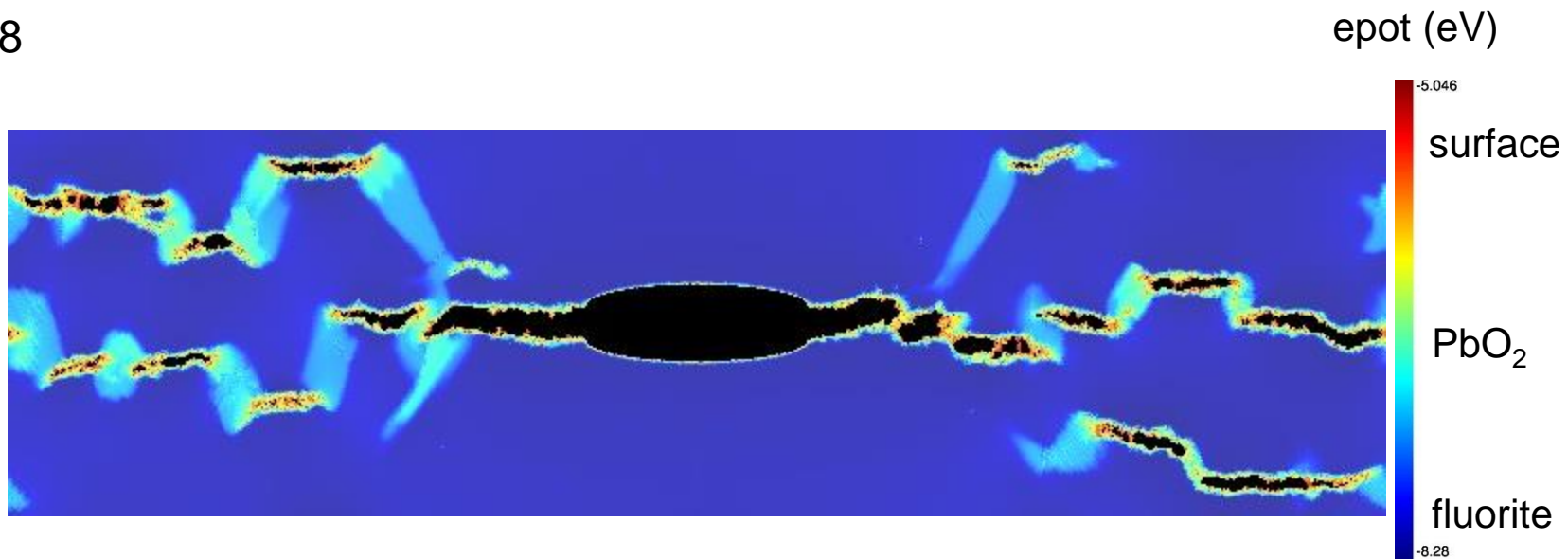
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.087$



CRACK PROPAGATION - WITH HE BUBBLES

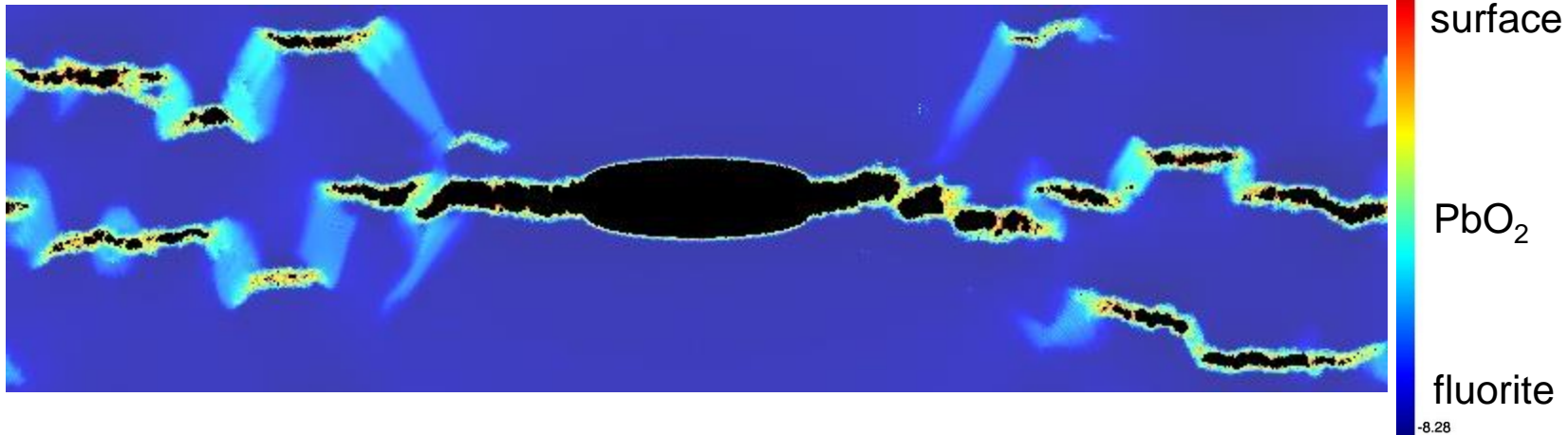
$\varepsilon = 0.098$



CRACK PROPAGATION - WITH HE BUBBLES

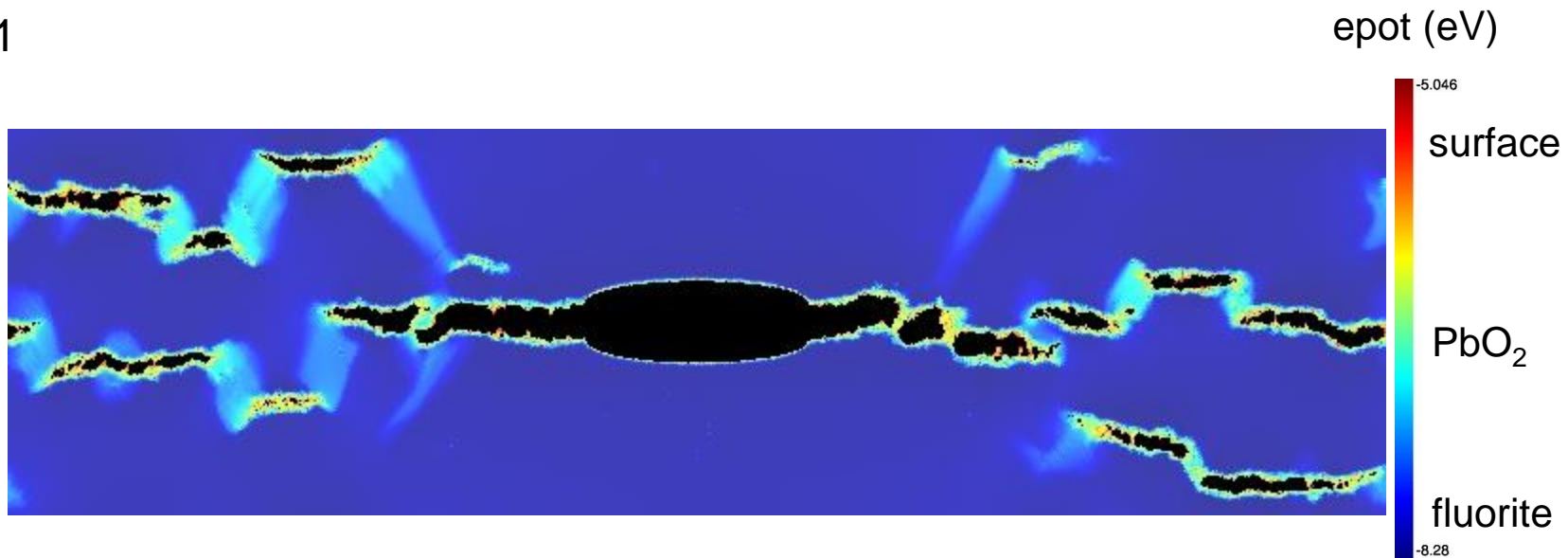
$\varepsilon = 0.104$

epot (eV)



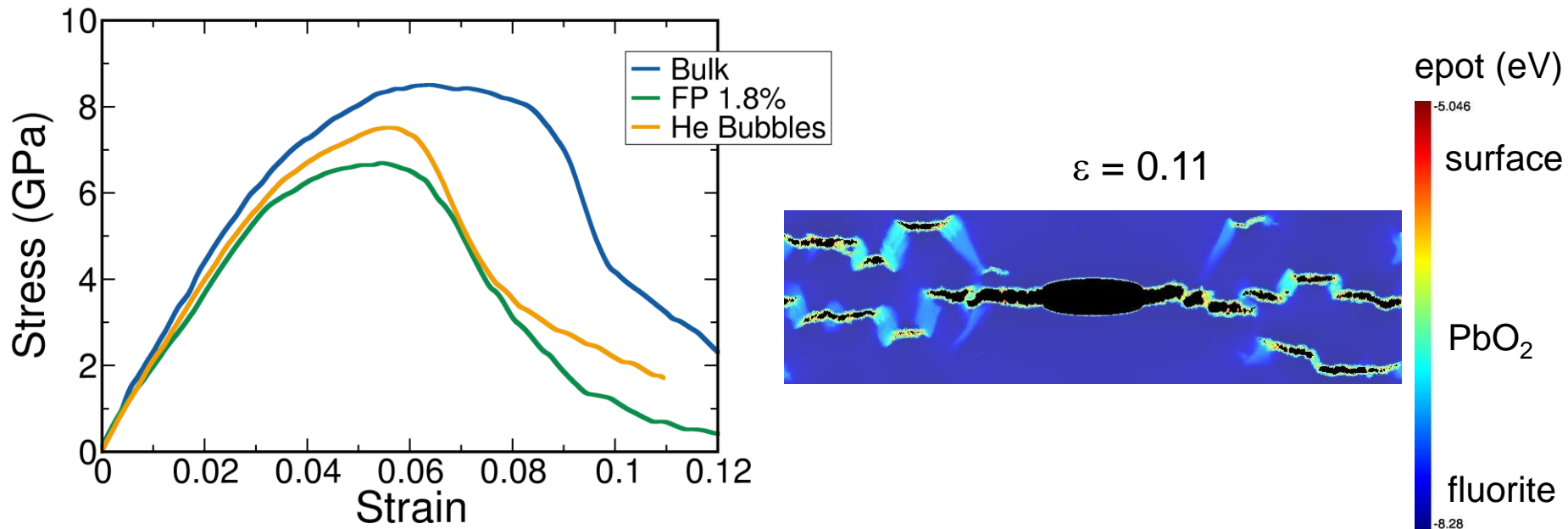
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.111$



- Same crack propagation than in bulk
- Sub-cracks initiated on the He bubbles

Crack propagation



- Crack propagation via **coalescence** of **sub-cracks**
- **Decrease** of the **toughness** with the presence of defects
- **Sub-cracks** initiated at the surfaces of the **He bubbles**

- Mechanical behavior of UO_2 with MD simulations
- **In Bulk**
 - Most fragile crystallographic orientation: $\langle 111 \rangle$
 - Difference with Griffith: due to **phase transition**
- **With point defects** – 0.04 to 7%
 - **Decrease** of yield stress and toughness
 - Threshold at **1.8%** point defects → **plasticity**
- **With He bubbles** – 2 nm, $10^{24}/\text{m}^3$
 - **Decrease** of yield stress and toughness
 - **Initiation** of crack **at the bubble**
- **Crack propagation**
 - Propagation with **coalescence** of **sub-crack**
 - Sub-cracks initiated at defects

Acknowledgments

JP Crocombette

P Fossati



Challenge project – C097073



Thank you for listening

Questions ?

Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 79 15 | F. +33 (0)1 69 08 92 21

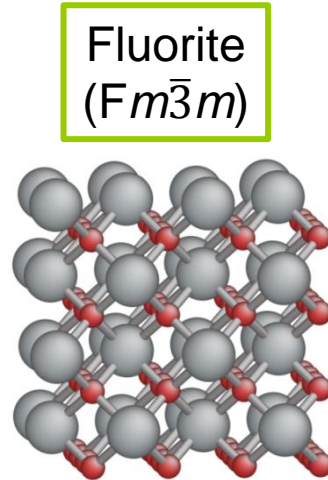
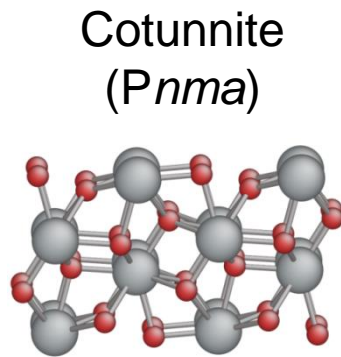
Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

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

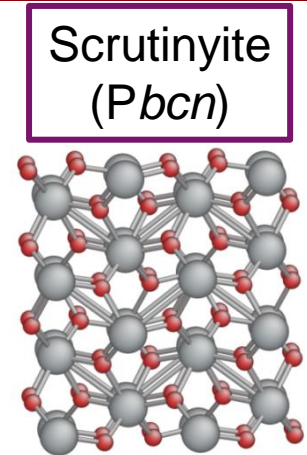
PHASE TRANSITIONS UNDER COMPRESSION

● oxygen

● uranium

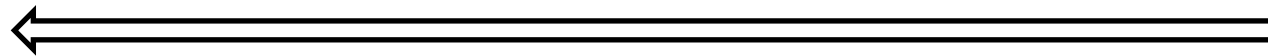
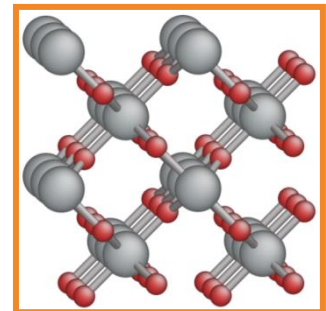


$\langle 100 \rangle$



Rutile
($P4_2/mnm$)

$\langle 110 \rangle$

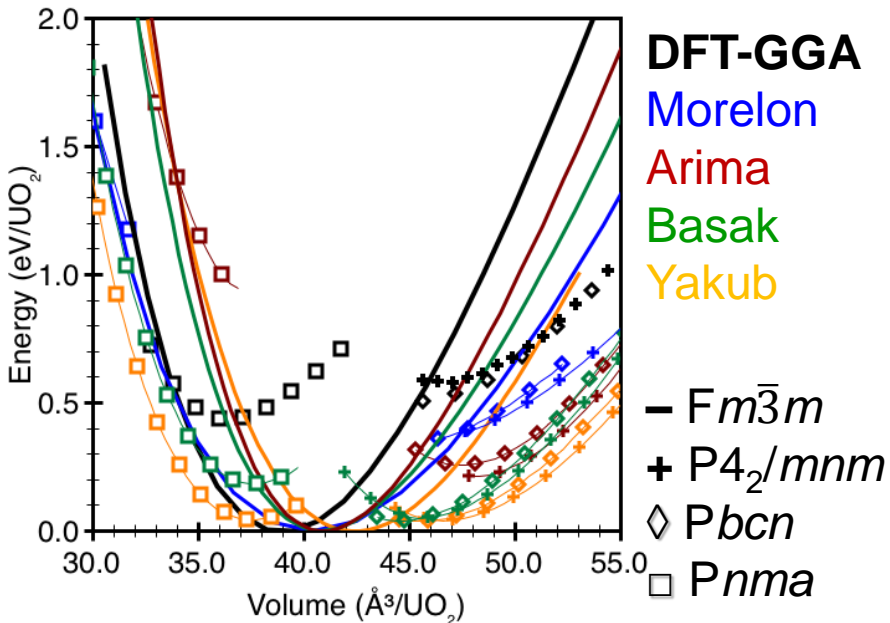


compression

tension



State equations



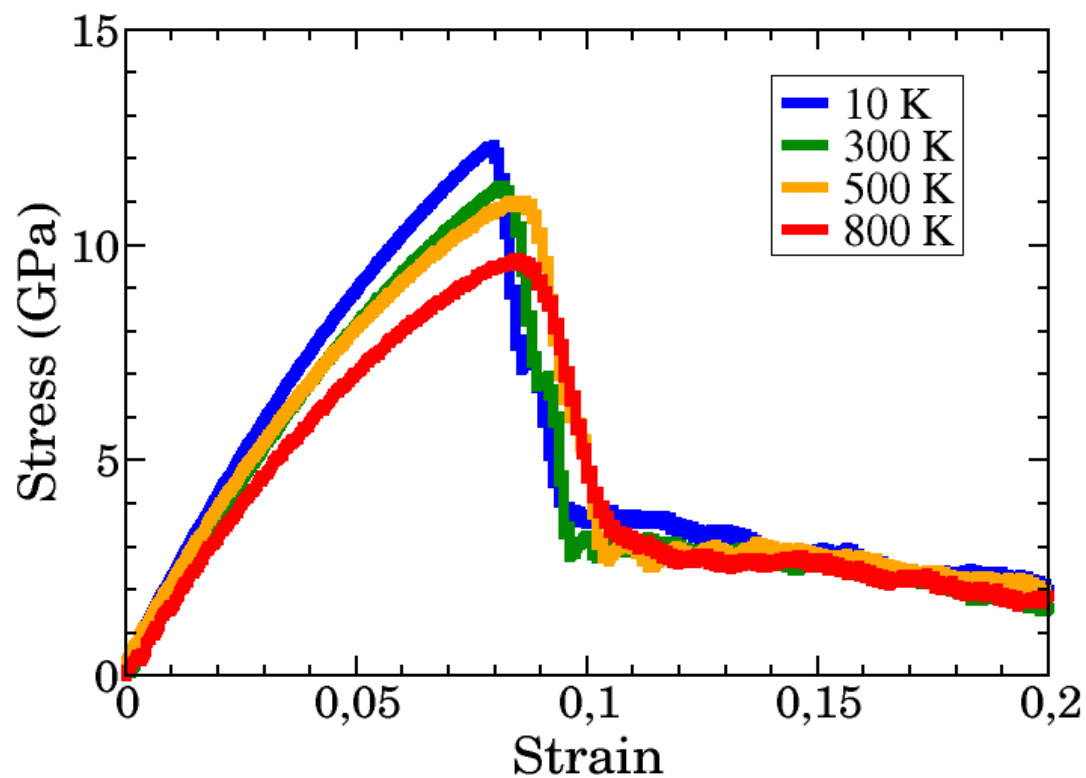
Transition Pressures (GPa)

Potentiel	cotunnite	scrutinyite	rutile
Arima	57	- 6	- 5
Basak	10	- 1,7	- 1,7
Morelon	45	-10	- 9
Yakub	1,3	- 1,4	- 1,9
Reference	42 (exp.)	- 8 (DFT)	-

- All potentials yield to the same phase transitions
- Morelon potential is the closest to the reference values

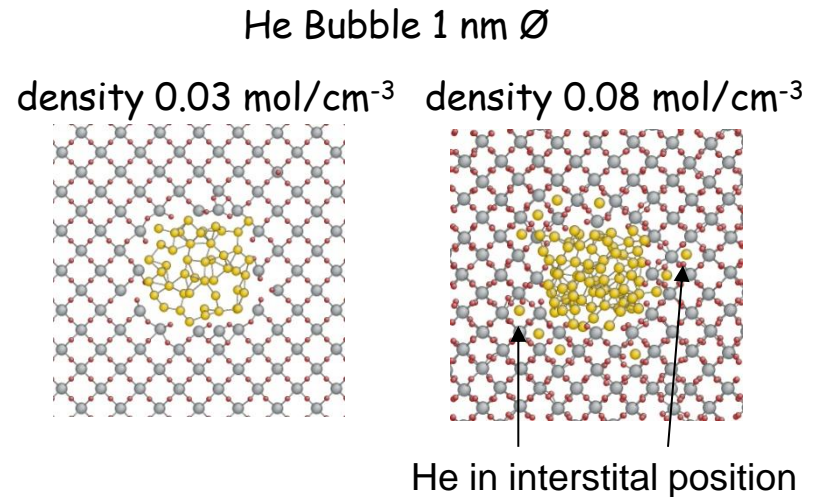
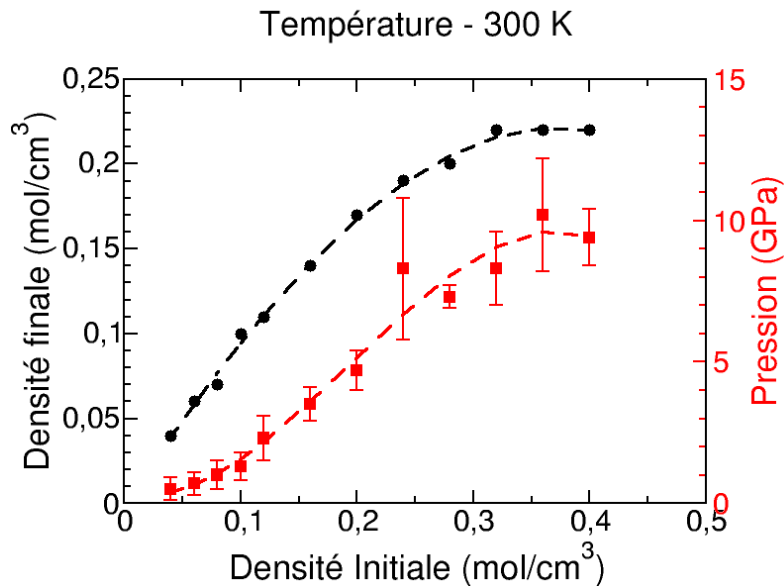


He Bubble - Strain rate $10^9/s$



- Temperature increase → **elasticity loss**
- **Disappearance of phase transition** with the increase of the temperature

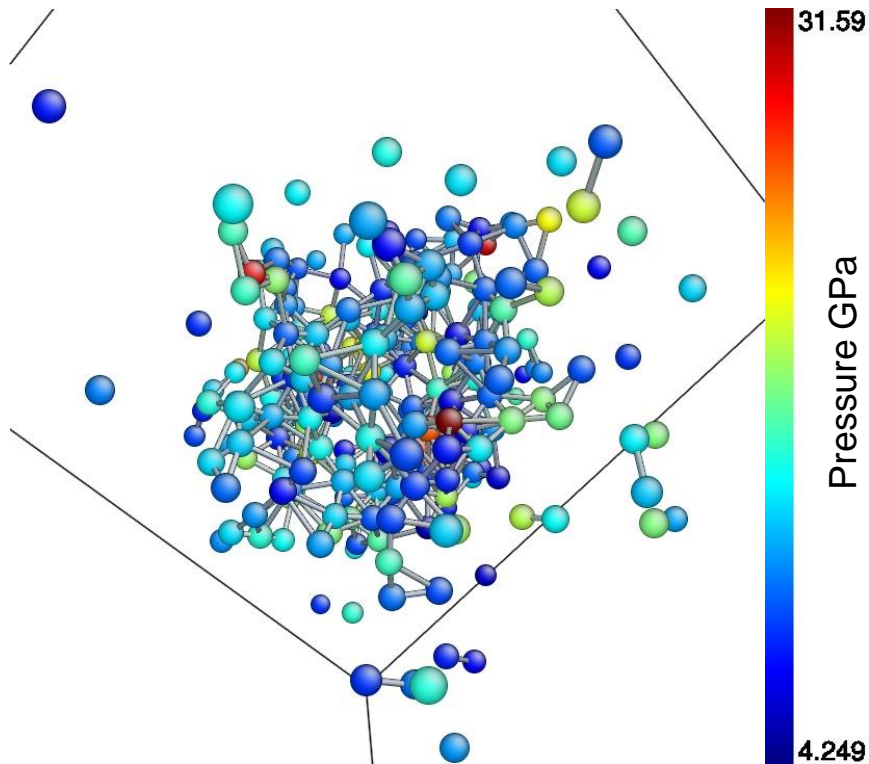
- Goal: study damage created by He bubble in UO_2 (defects, pressure, ...)
- Method: He Bubble in UO_2 (\neq density, size, temperature)
- Main results:



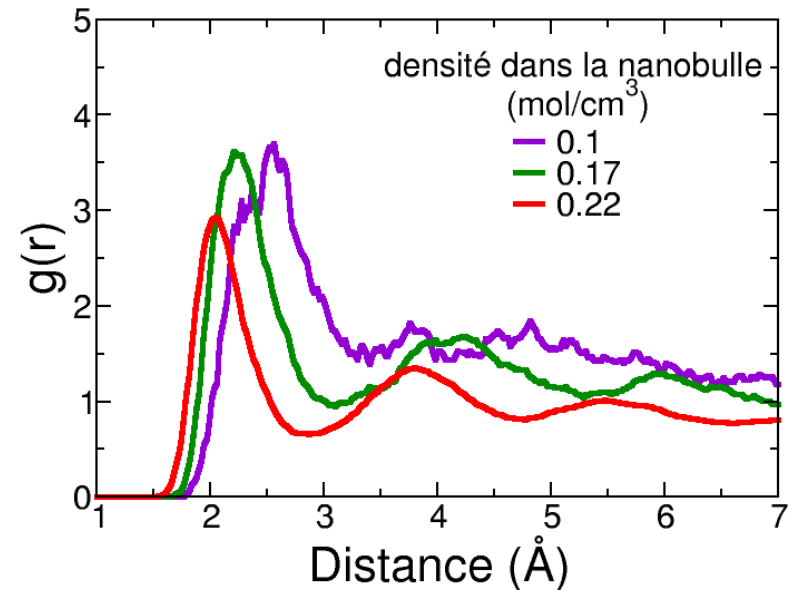
- No influence of the temperature (300 – 1500 K)
- He **partially soluble** in UO_2 (\rightarrow interstitial)
- Density threshold \approx **0.04 mol/cm⁻³** (= 1 He/Schottky)
- Max pressure in bubble **10 GPa**, He state equation \neq VdW
- Pressure at **0.04 mol/cm⁻³** = **500 MPa**

STRUCTURE IN HE BUBBLES

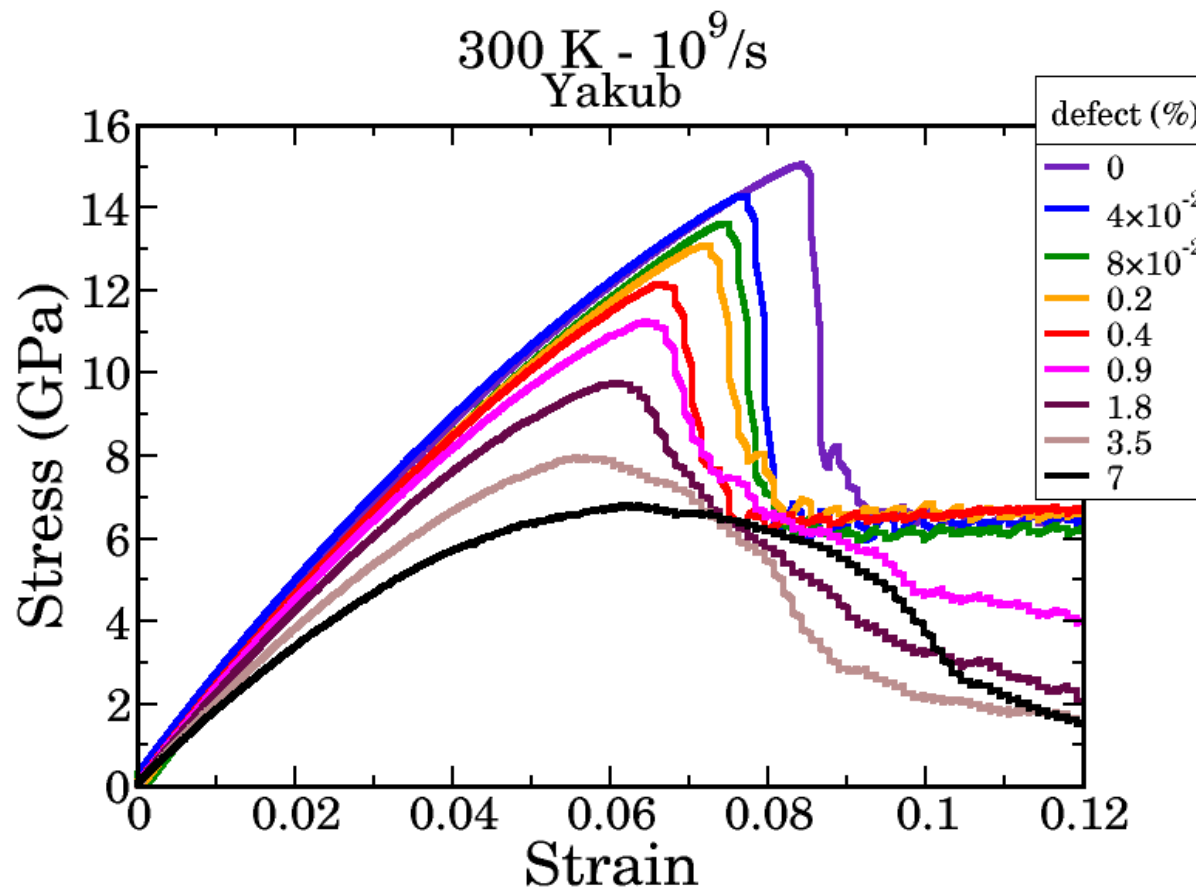
300 K – nanobubble 0.22 mol/cm³



Température - 300 K



- Gaz structure → amorphous with increasing density
- ≠ Xénon (fcc structure for high density)



- General no phase transformation
- Threshold at ~ 1.8 % defects