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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

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Submitted on 26 Feb 2020

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MECHANICAL BEHAVIOR OF UO₂ UNDER IRRADIATION: A MOLECULAR DYNAMICS STUDY

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NUFUEL
Research into Nuclear Fuel in Europe

November 16-18, 2015 Karlsruhe, Germany

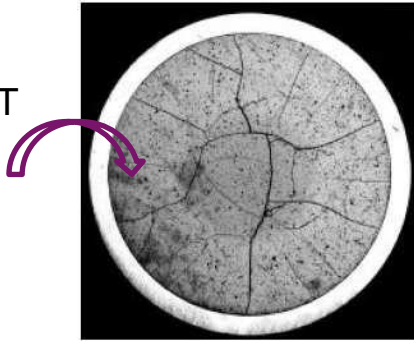
FUEL DESIGN, AN ENGINEERING CHALLENGE

*Macrograph of a fuel pellet
after nuclear reactor*

*SEM image of He
Bubbles in UO_2*

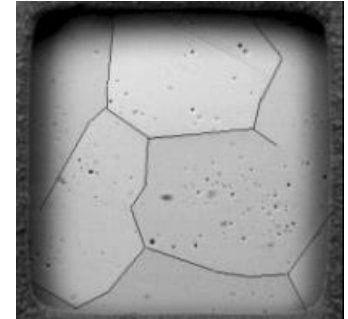
In reactors

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



In interim storage or long-term disposal

- Swelling ← He bubbles



Higher burn-up



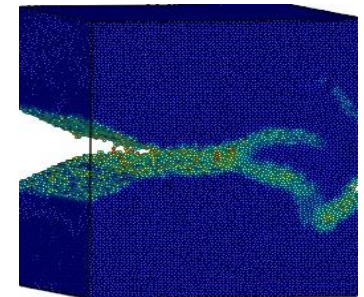
Mechanical properties ?

elasticity, yield stress, hardness, toughness...

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

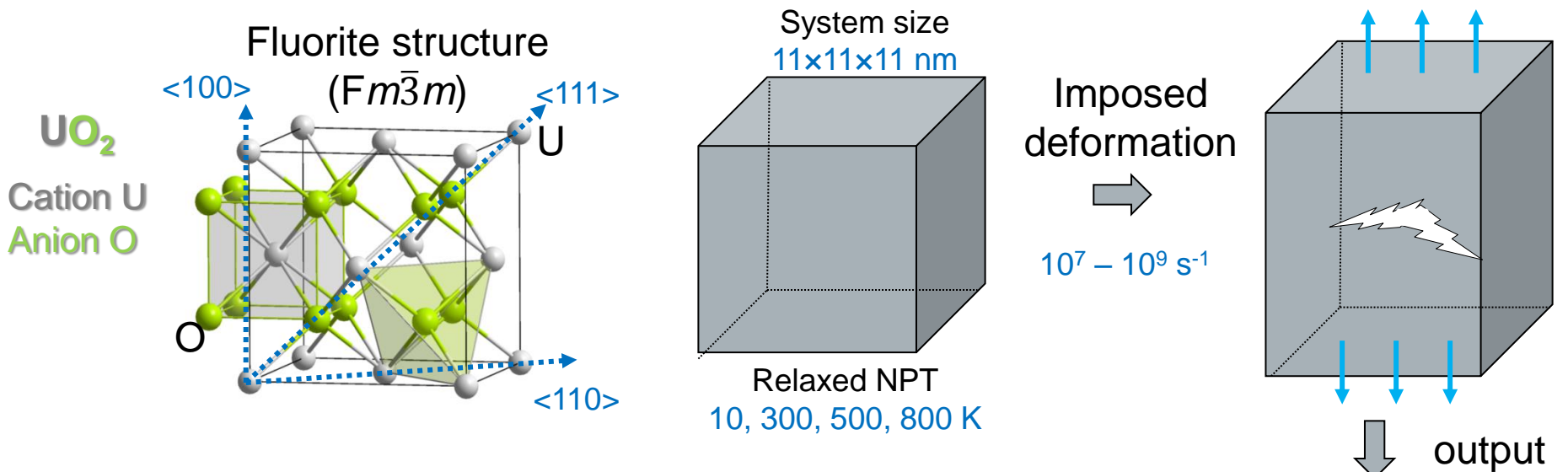
■ **Goal:**

- 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

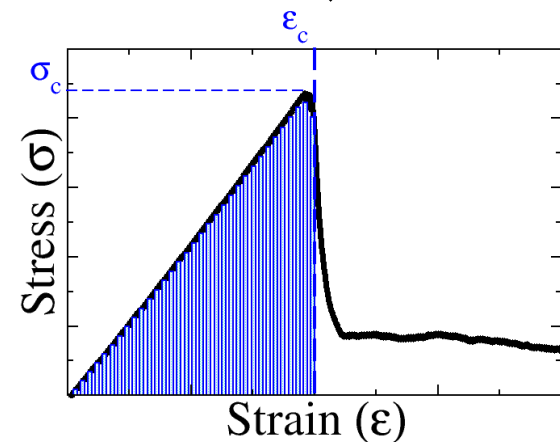


Outputs:

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

Interatomic potentials:

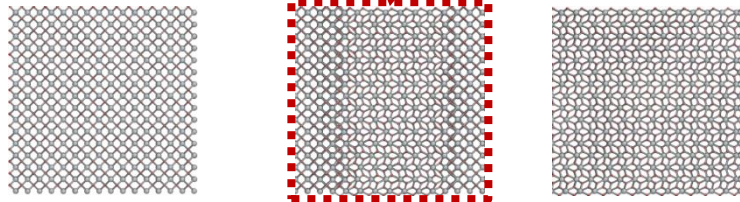
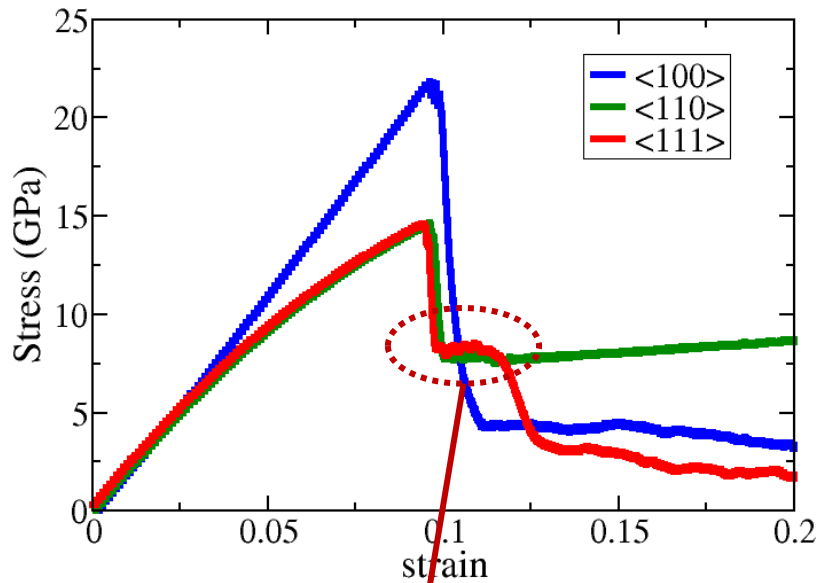
- Yakub: good elastic constants
- Morelon: good phase transitions



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**
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- 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

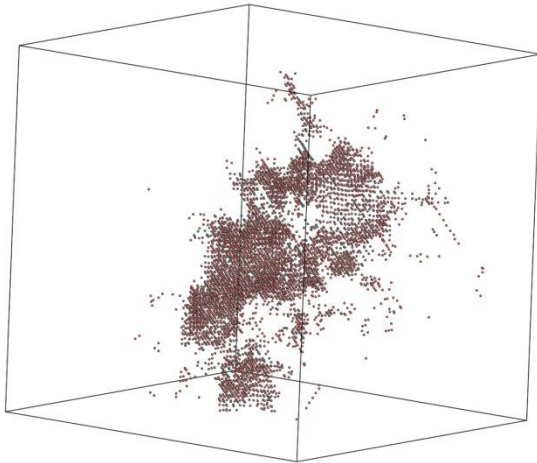


Y. Zhang JNM 430 (2012) 96
P. Fossati PRB 88 (2013) 214112

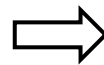
- Weakest crystallographic direction: <111>
- Difference with Griffith criteria \leftrightarrow phase transformation

- I. Mechanical behavior in Bulk
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MD simulation of
80 keV displacement cascade

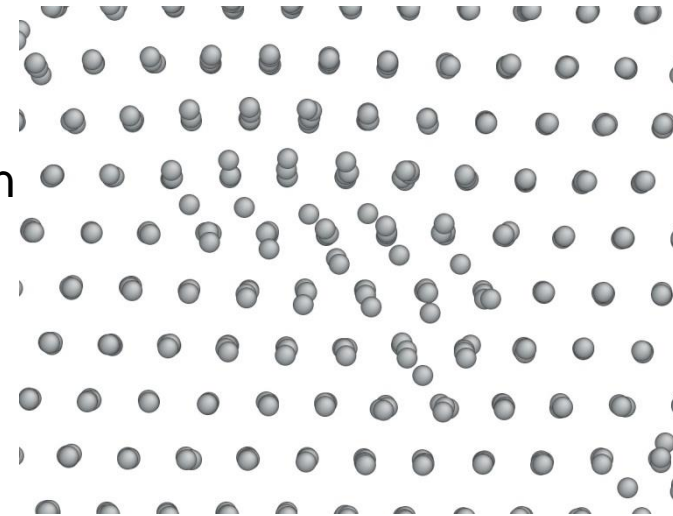


Random
Frenkel pair accumulation



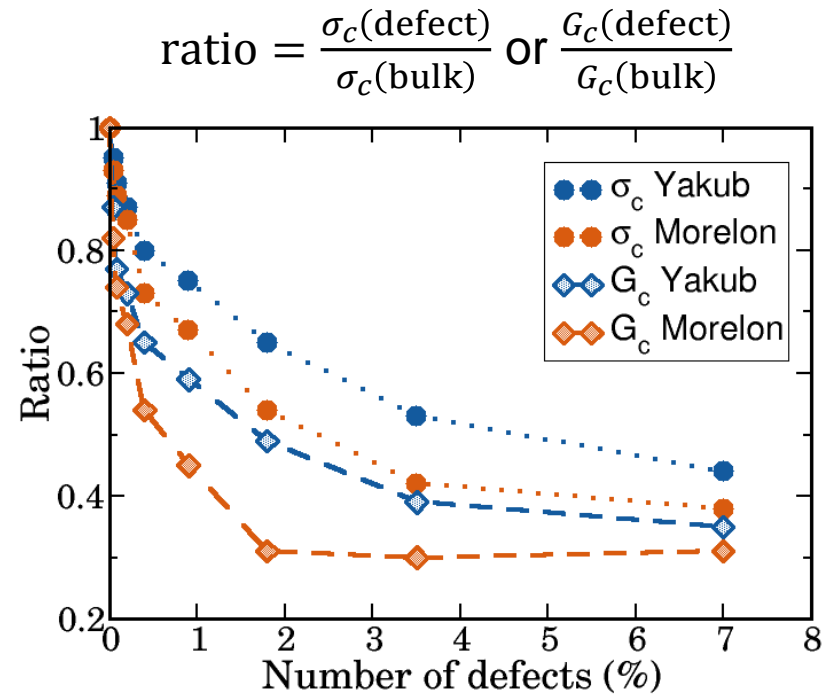
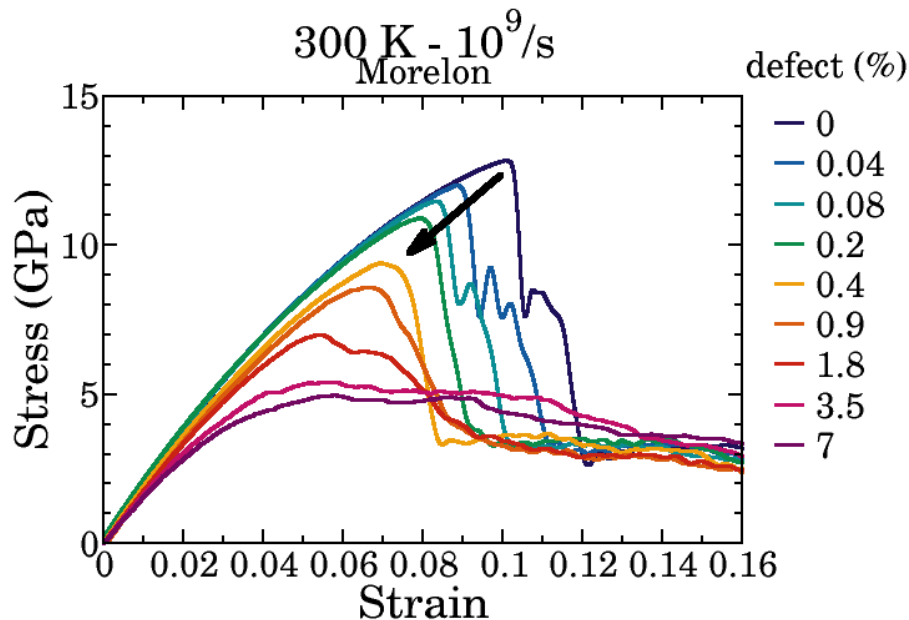
0.04 → 7%

Close up of
small Frank loops



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

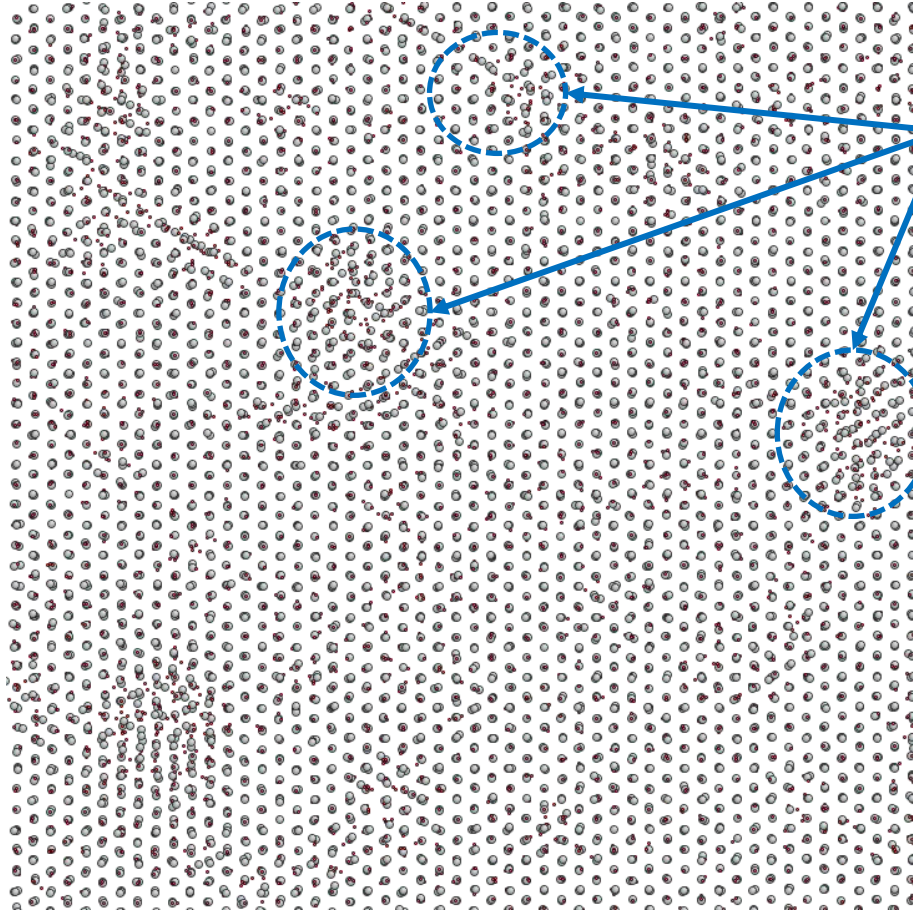
- After NPT relaxation (300 K) → Formation of **small clusters** (mainly int.)
- For systems > 2% of defects, clusters are **small dislocation loops**



- 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



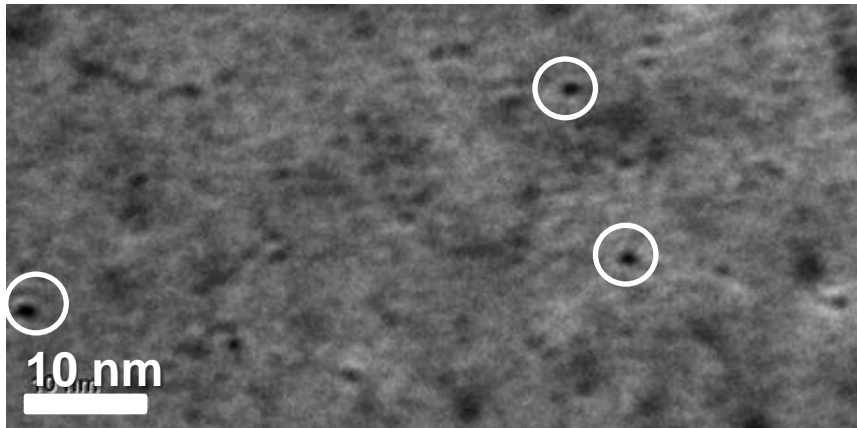
Clusters of defects

- 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

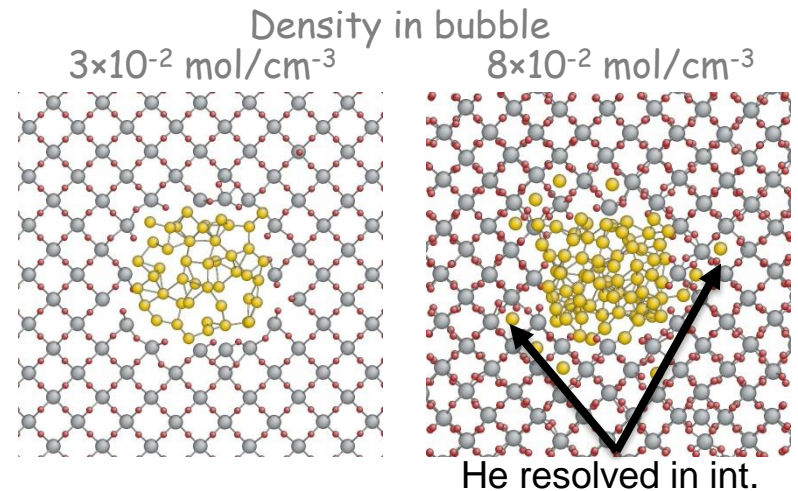
- Bubble density saturates at: $4 \times 10^{23} \text{ m}^{-3}$
- Bubble size: 1 – 2 nm

■ To fit experimental observations:

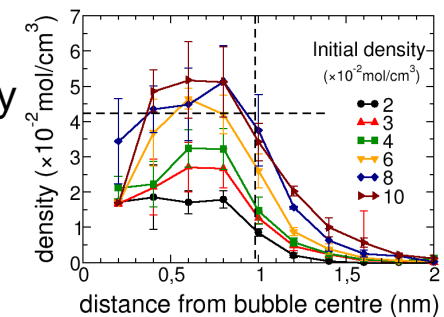
Bubble: Ø 2 nm, density of bubbles = 10^{24} m^{-3} ,

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

MD simulations



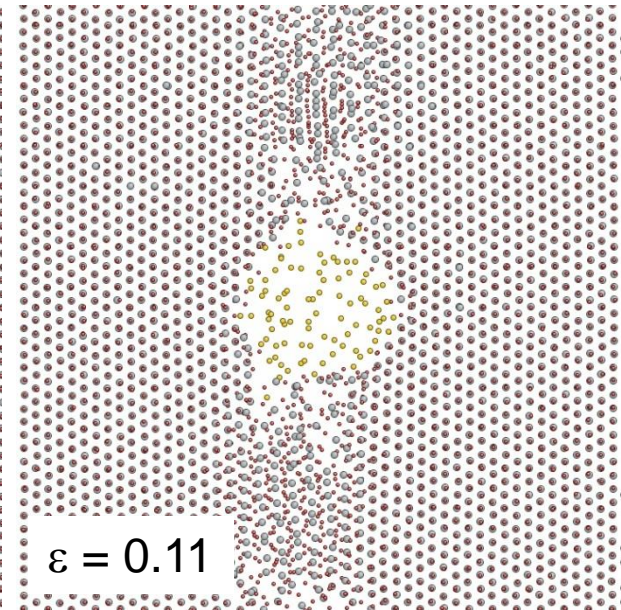
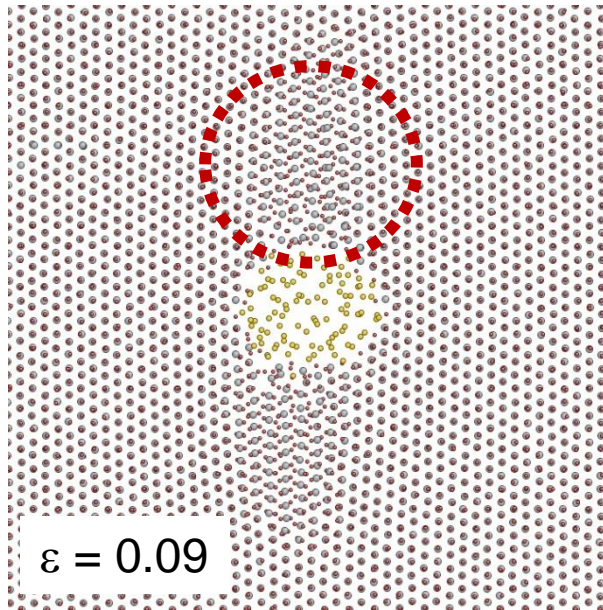
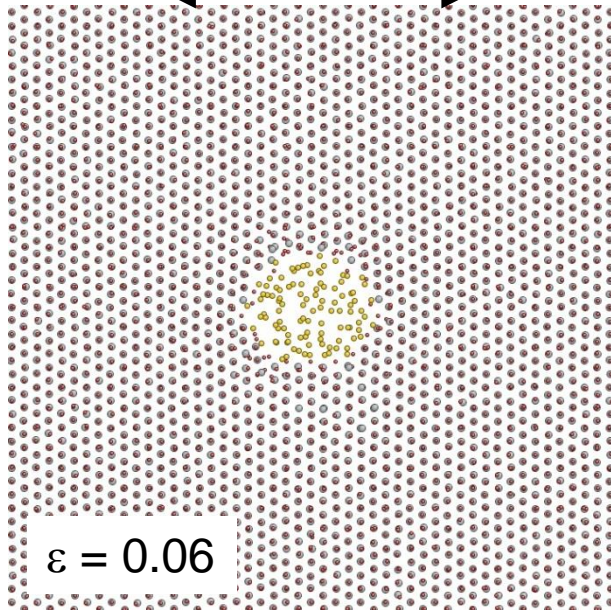
He resolution for density
> $4 \times 10^{-2} \text{ mol/cm}^3$



MECHANICAL BEHAVIOR - WITH HE BUBBLES

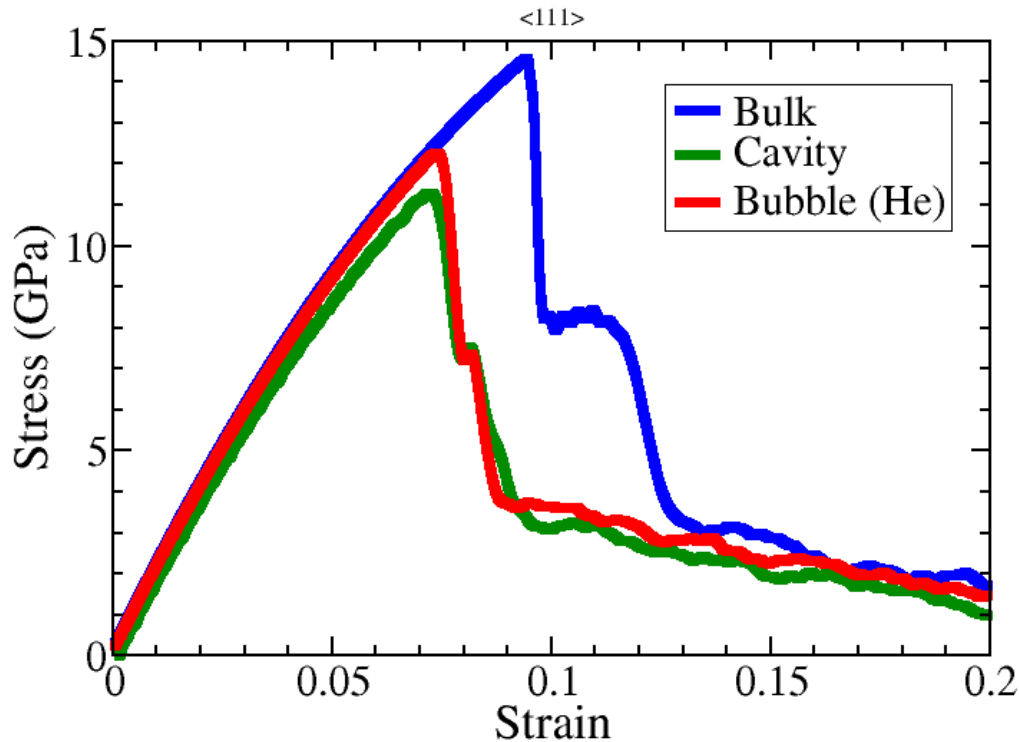
bubble : \varnothing 2 nm, He density in bubble = 4×10^{-2} mol/cm³, Pressure = 500 MPa

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
 $G_C \sim G_C(\text{Griffith})$

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

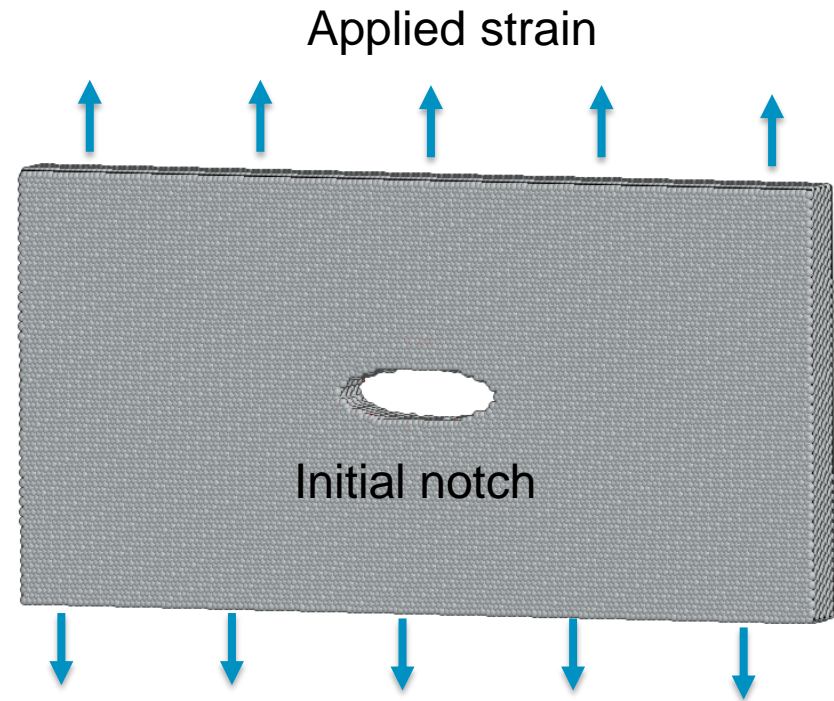
Strain orientation	G_C (J/m ²)		
	Bulk	Cavity	He bubble
<100>	12.73	7.21	7.32
<110>	9.54	5.96	6.76
<111>	9.51	5.62	6.08

- I. **Mechanical behavior in Bulk**
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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



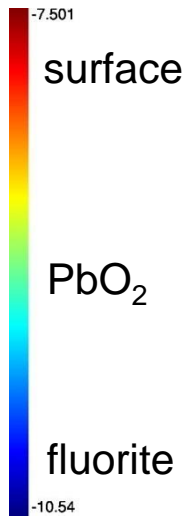
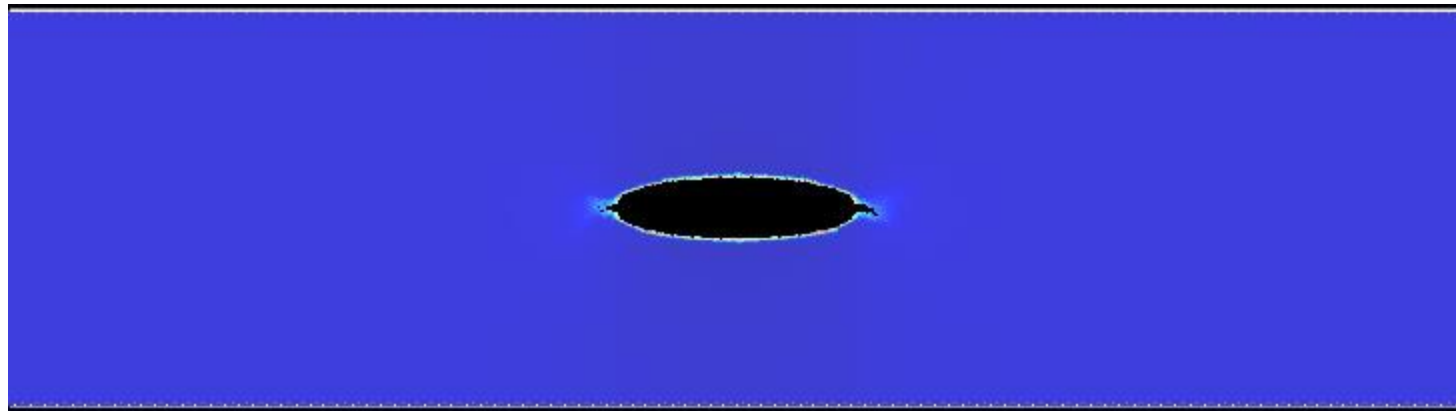
Strip geometry:
 $241 \times 65 \times 4$ nm

Initial notch:
Ellipse 40×8 nm

CRACK PROPAGATION - IN BLUK

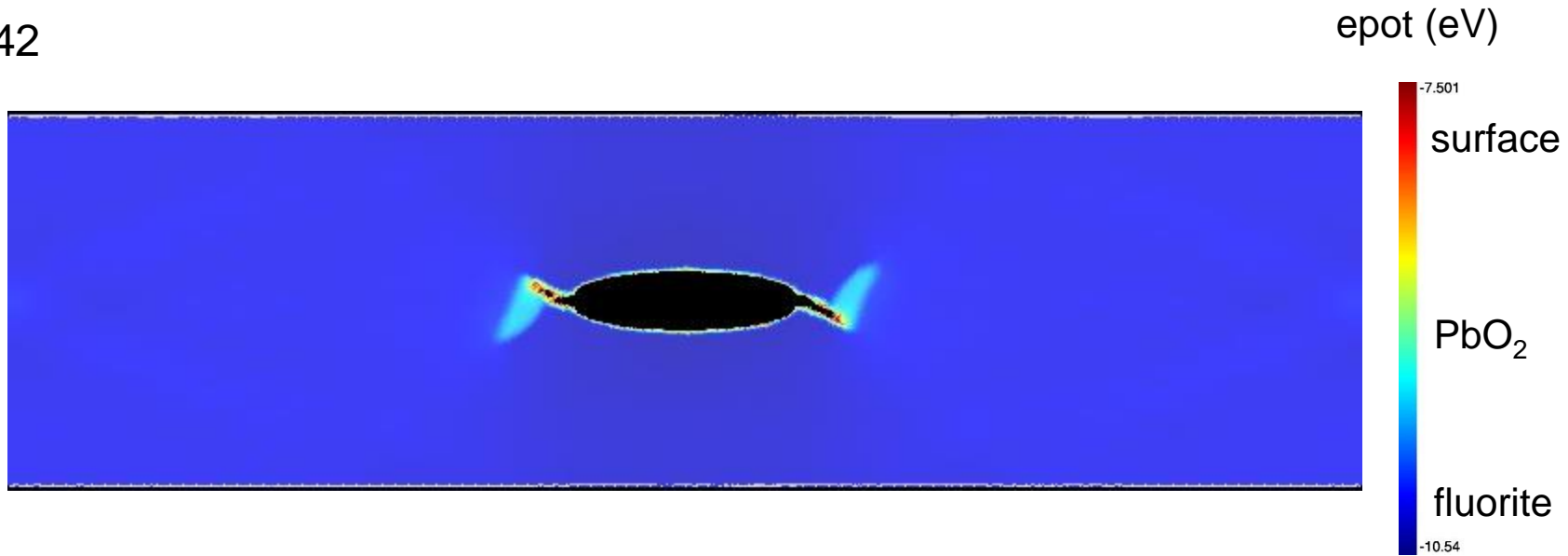
$\varepsilon = 0.036$

epot (eV)

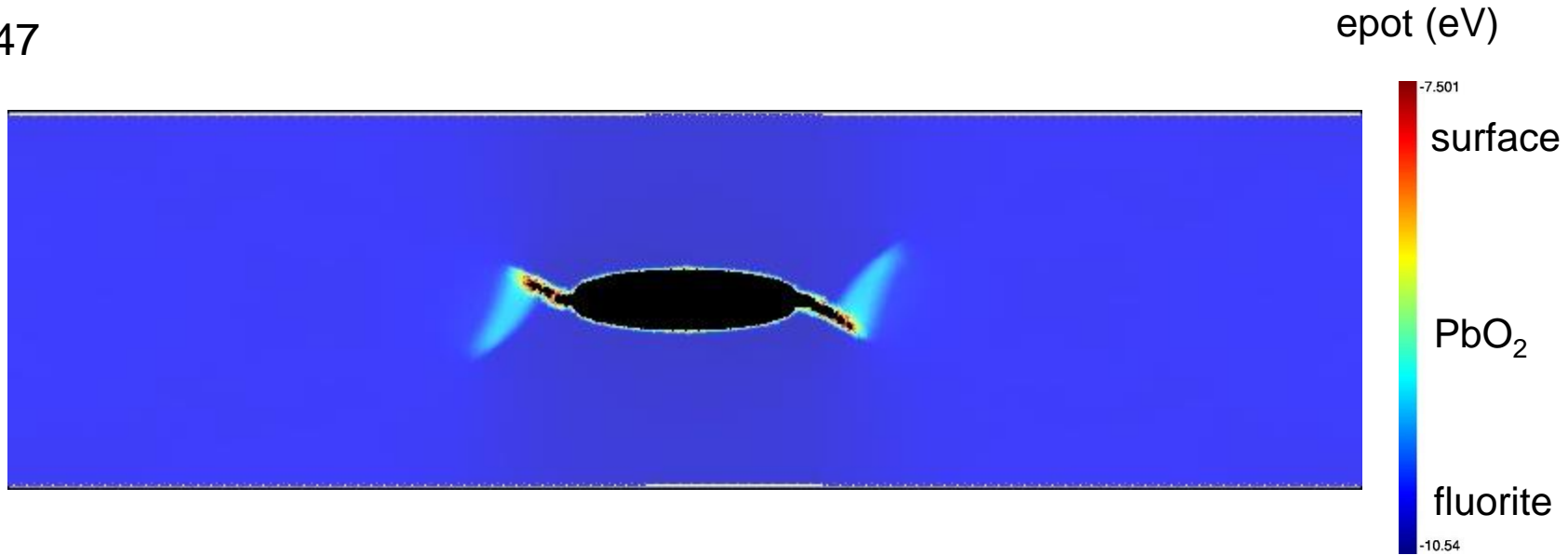


CRACK PROPAGATION - IN BLUK

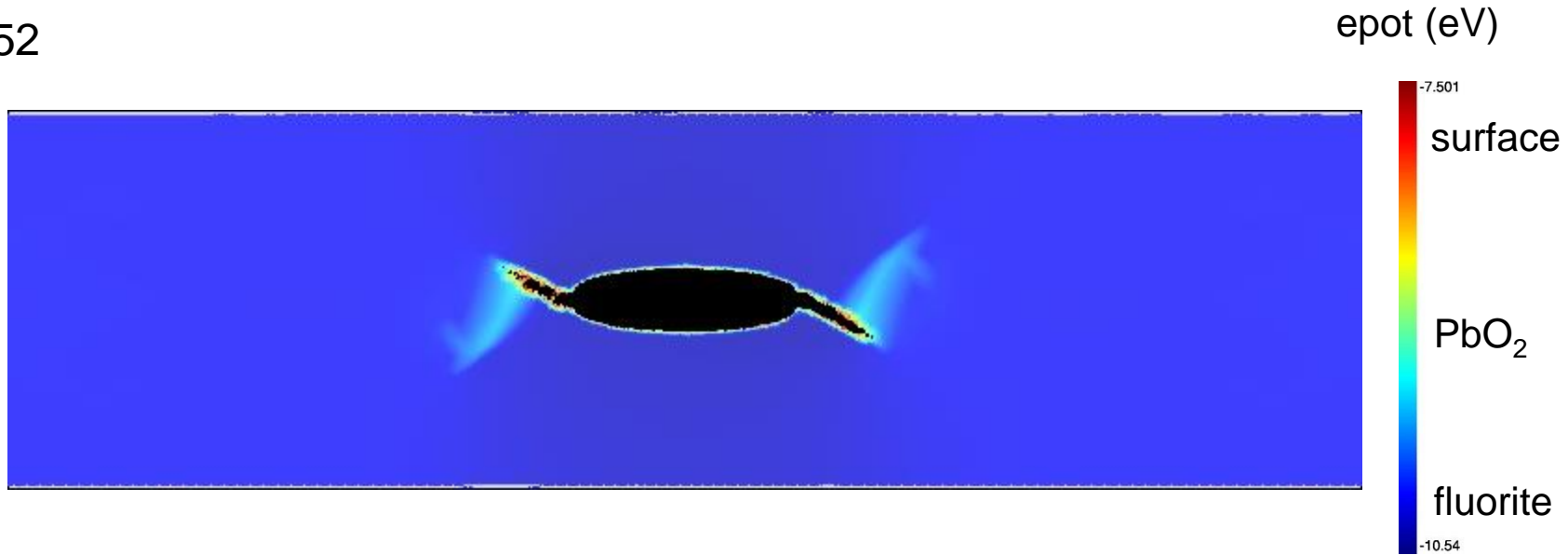
$\varepsilon = 0.042$



$\varepsilon = 0.047$



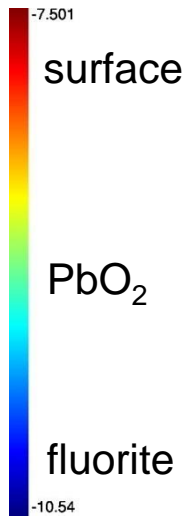
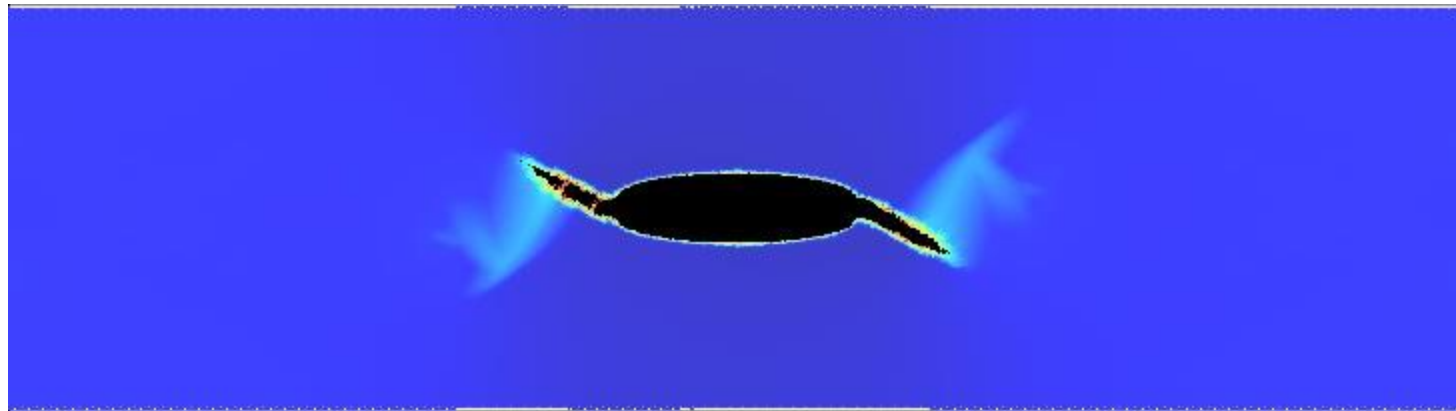
$\varepsilon = 0.052$



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.058$

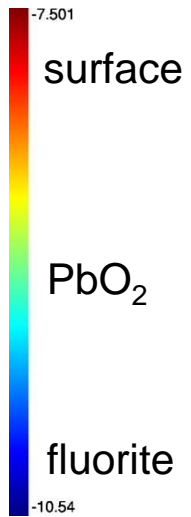
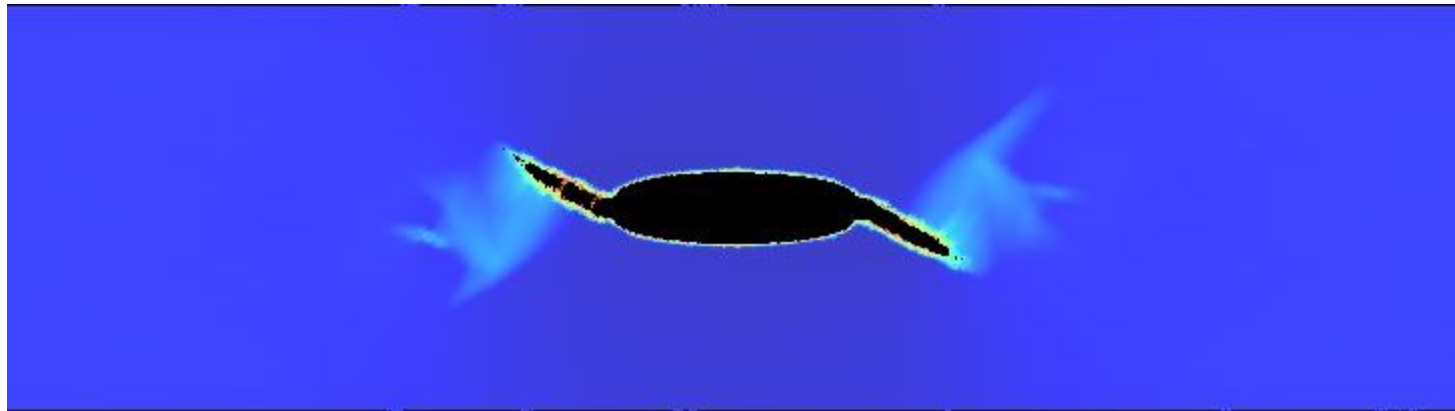
epot (eV)



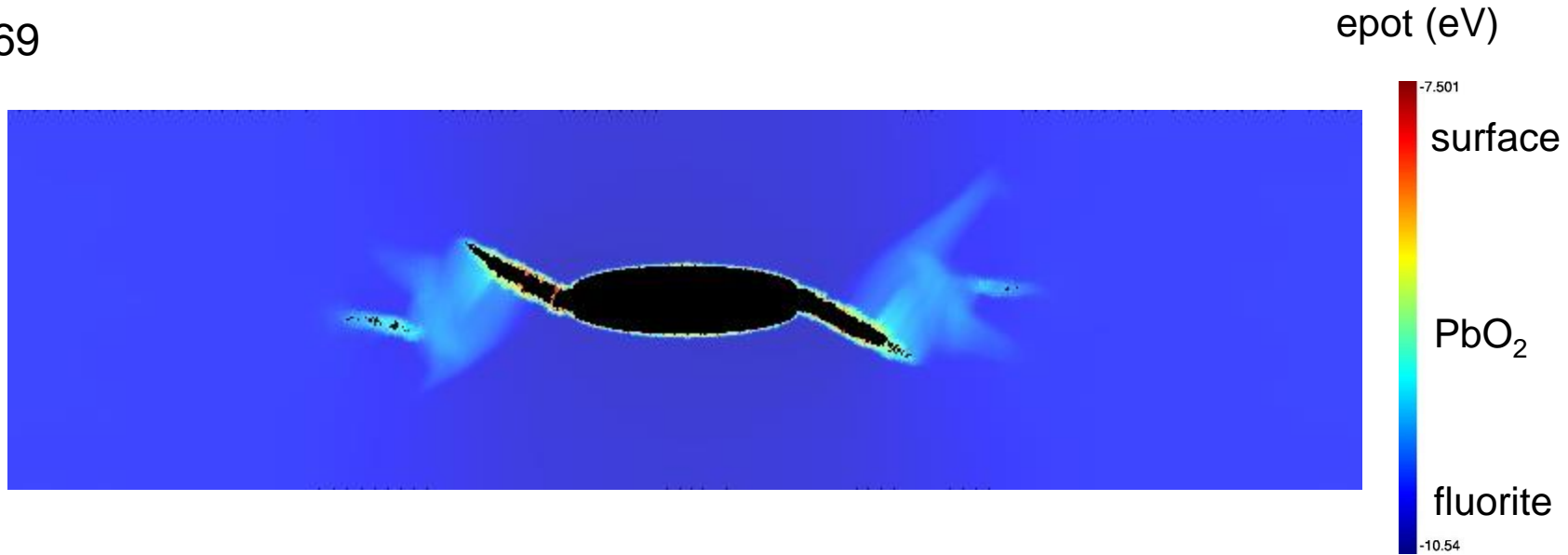
CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.064$

epot (eV)

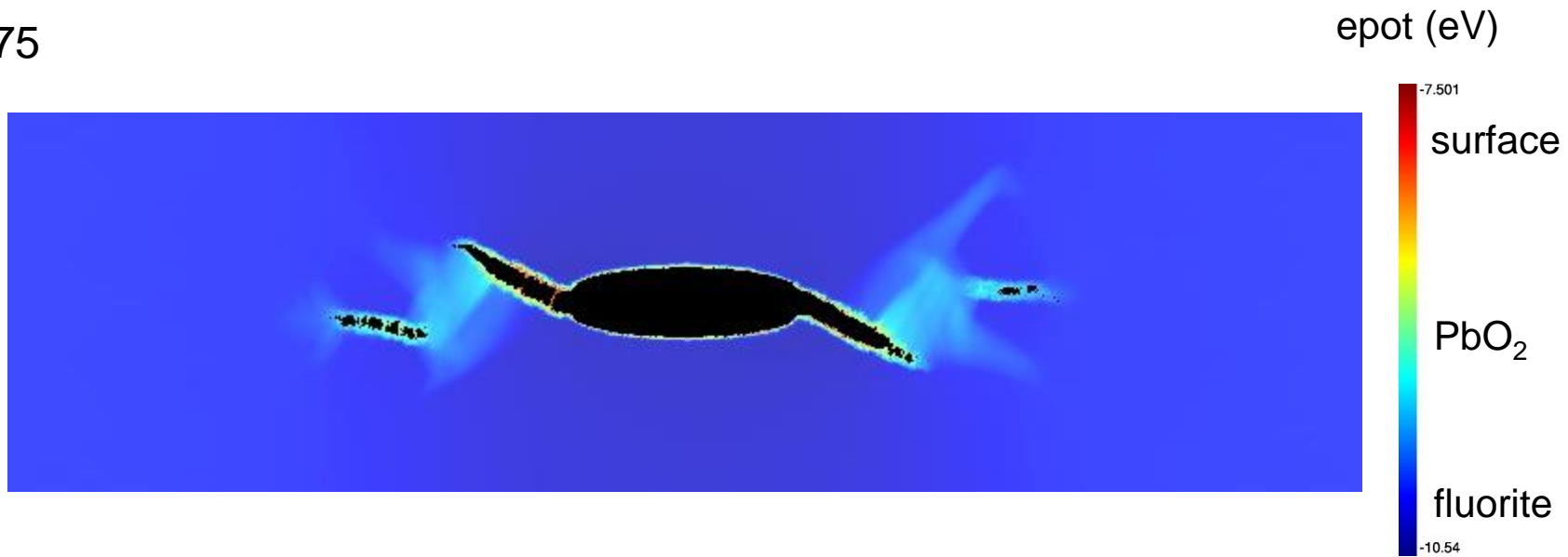


$\varepsilon = 0.069$



CRACK PROPAGATION - IN BLUK

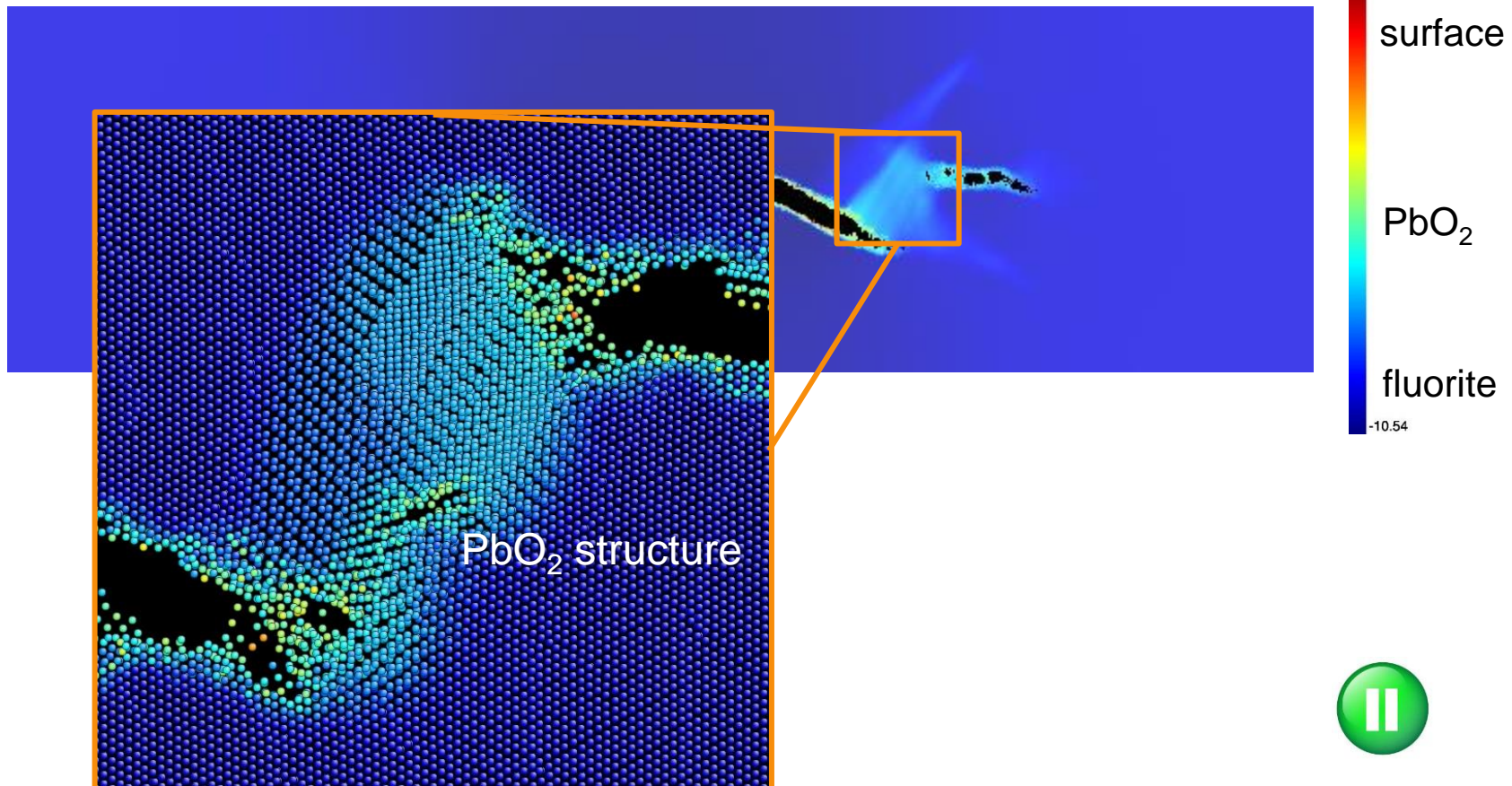
$\varepsilon = 0.075$



CRACK PROPAGATION - IN BLUK

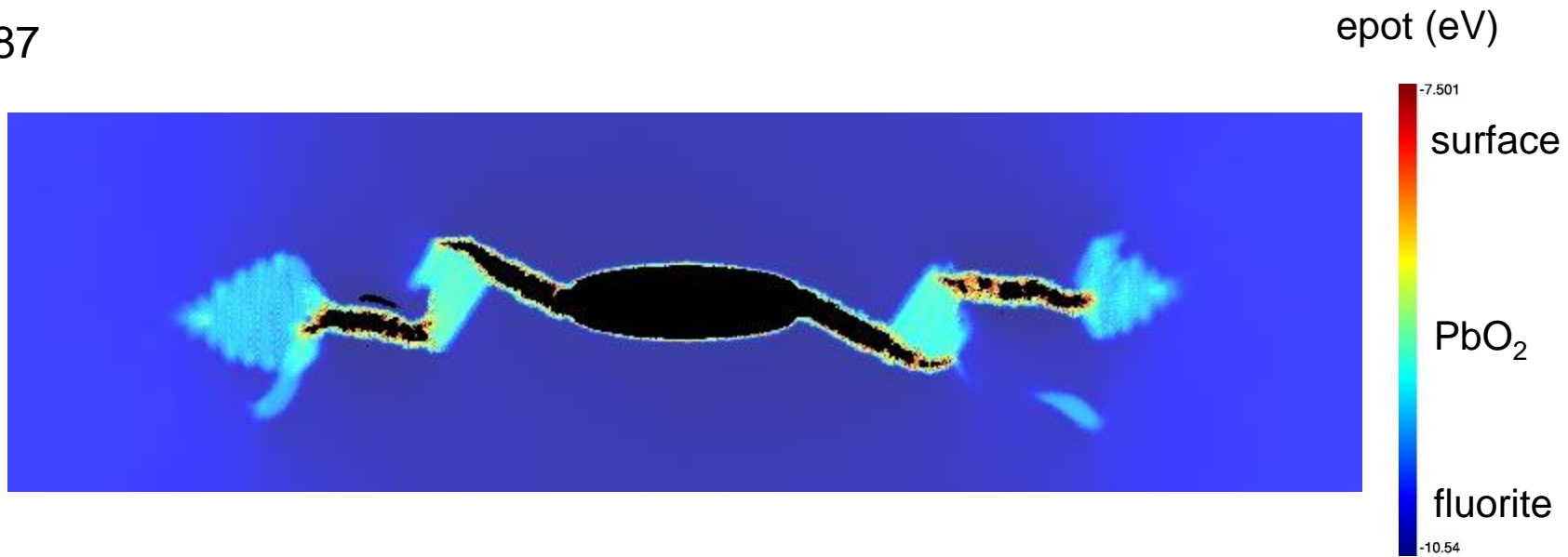
$\varepsilon = 0.081$

epot (eV)



CRACK PROPAGATION - IN BLUK

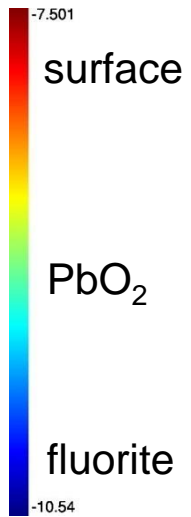
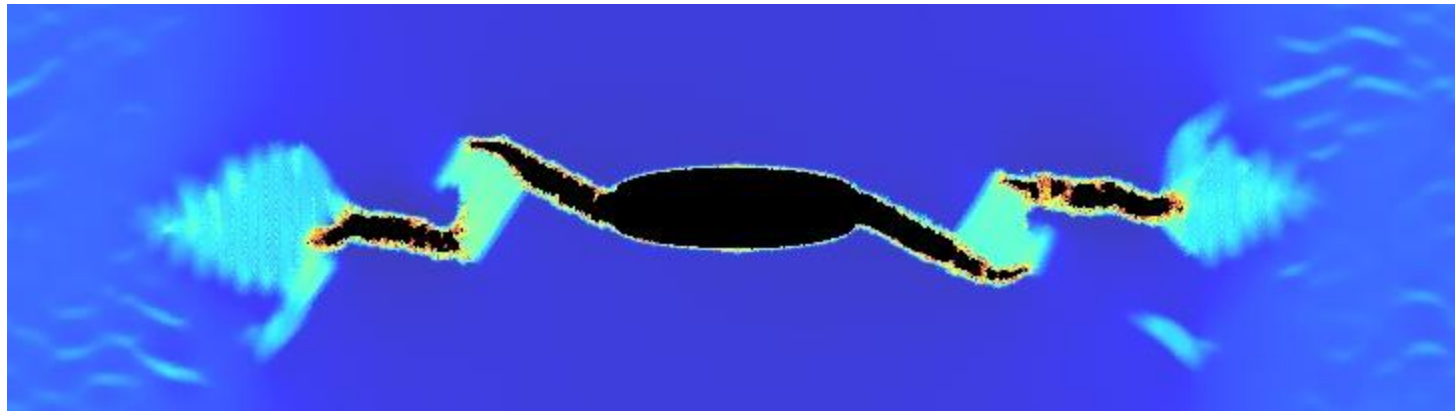
$\varepsilon = 0.087$



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.093$

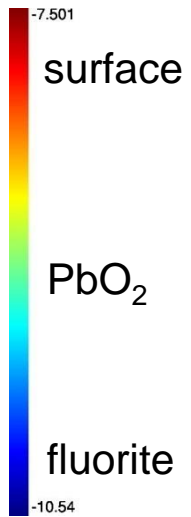
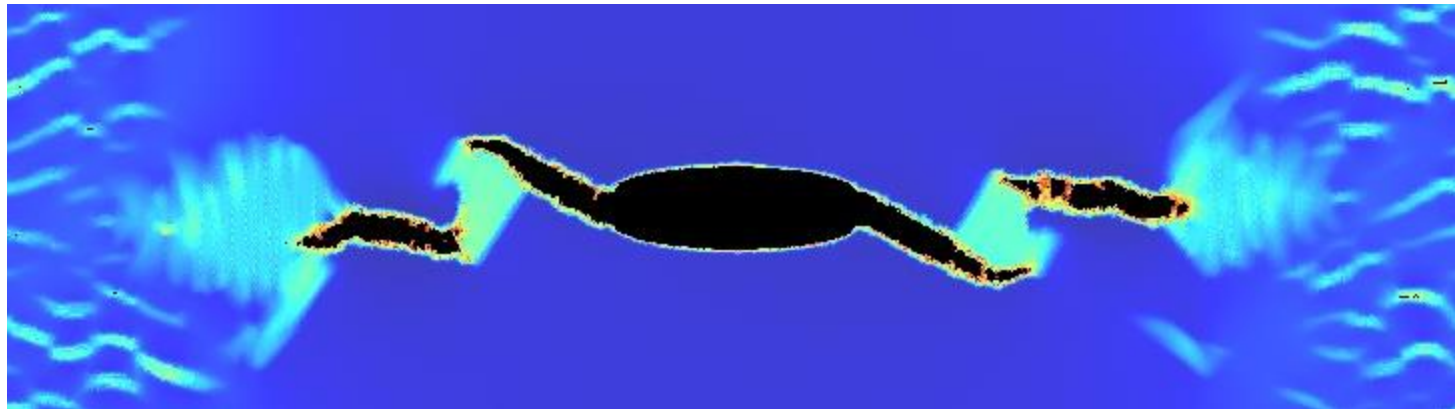
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.098$

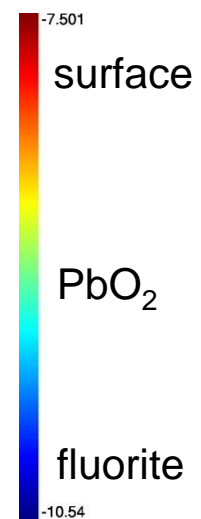
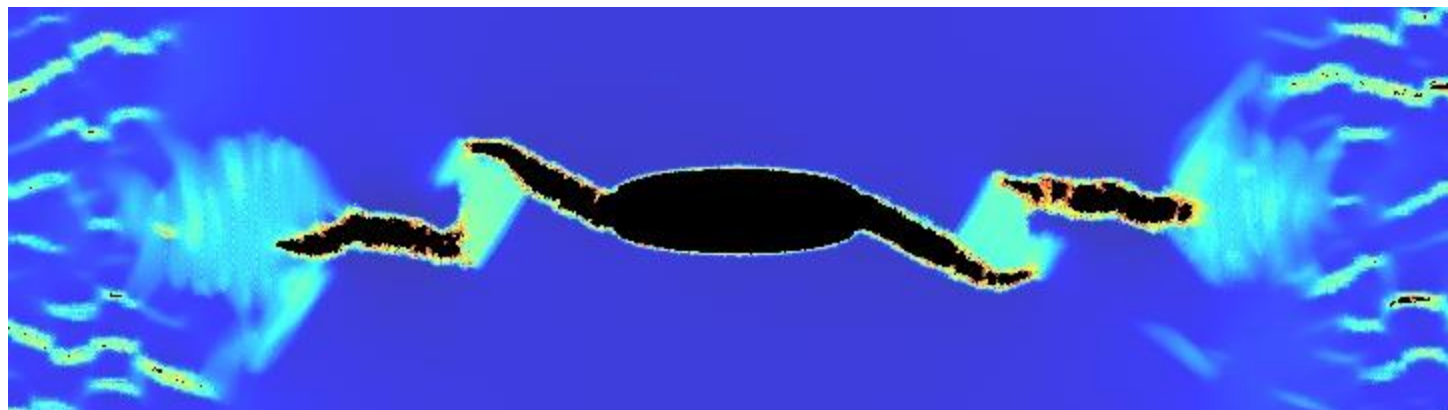
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.104$

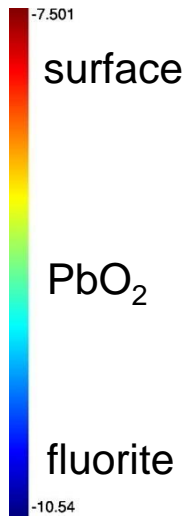
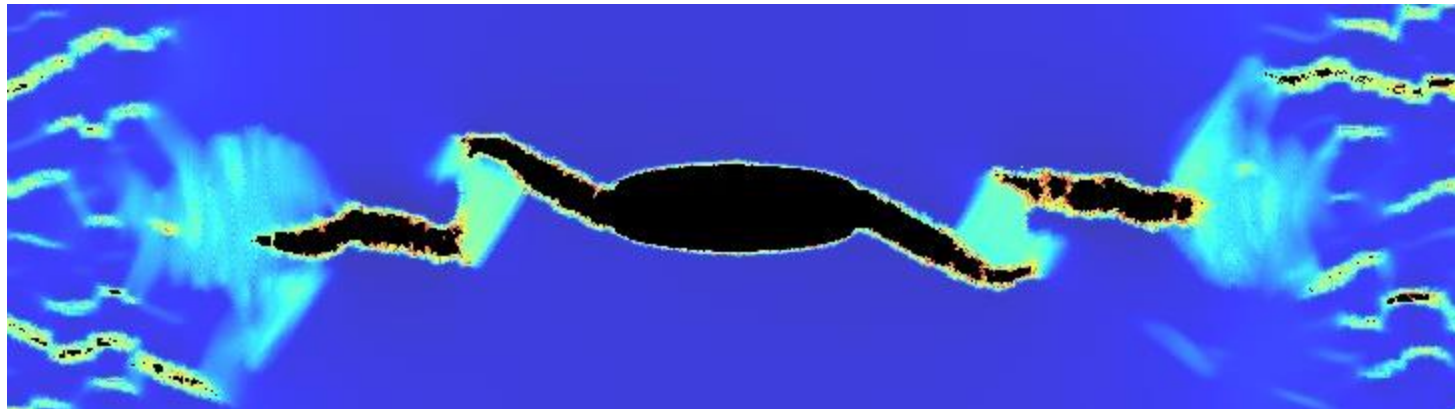
epot (eV)



CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.111$

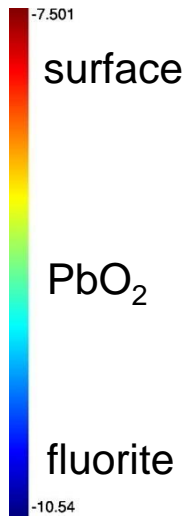
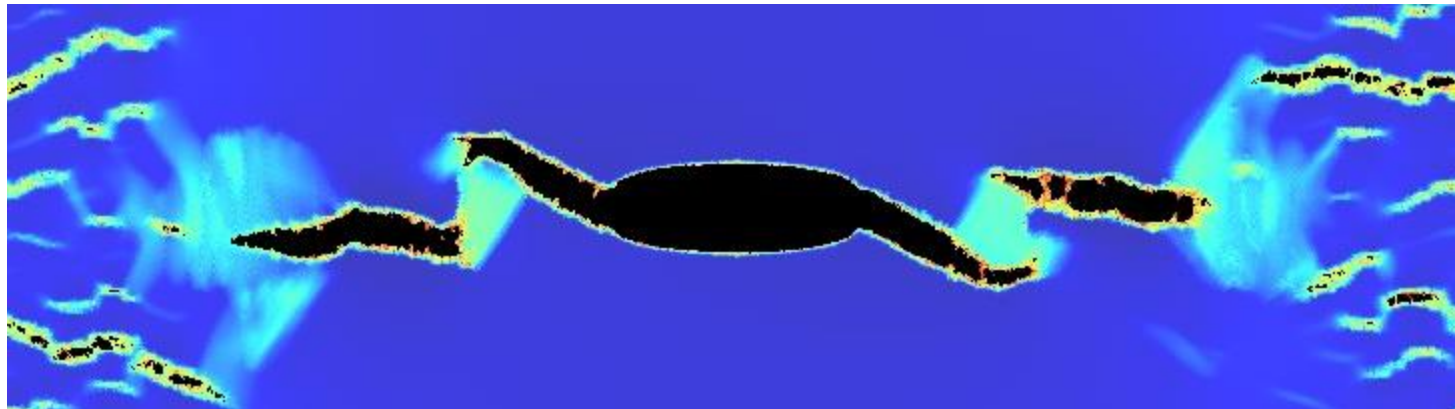
epot (eV)



CRACK PROPAGATION - IN BLUK

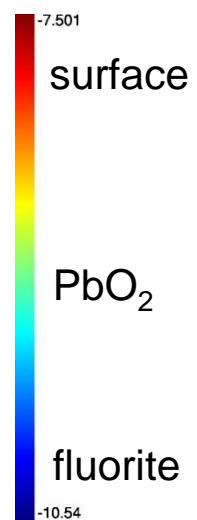
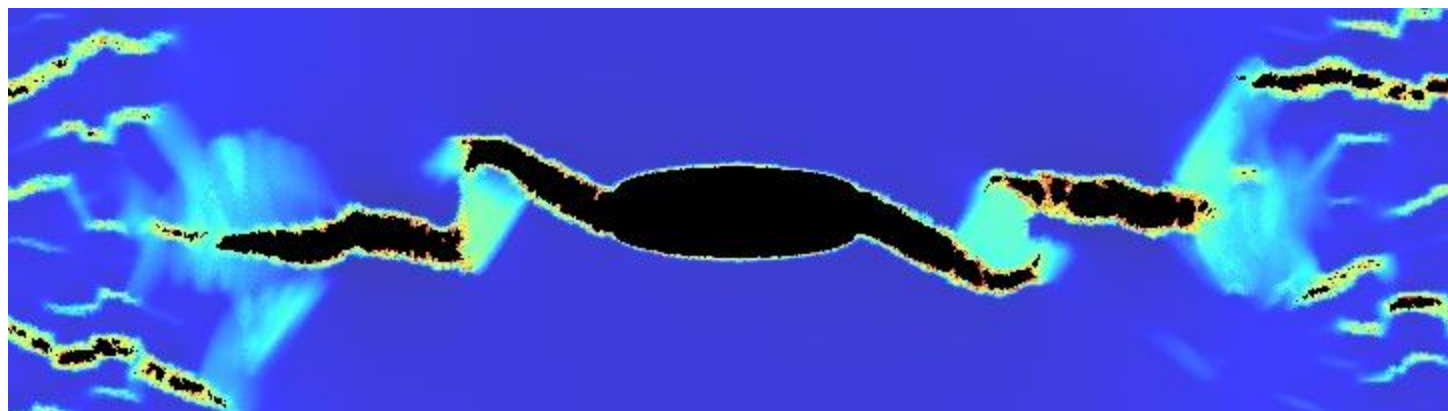
$\varepsilon = 0.117$

epot (eV)



$\varepsilon = 0.123$

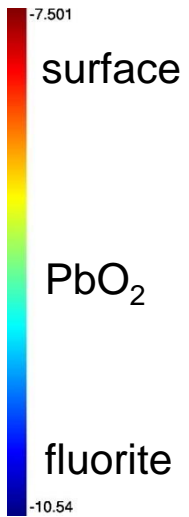
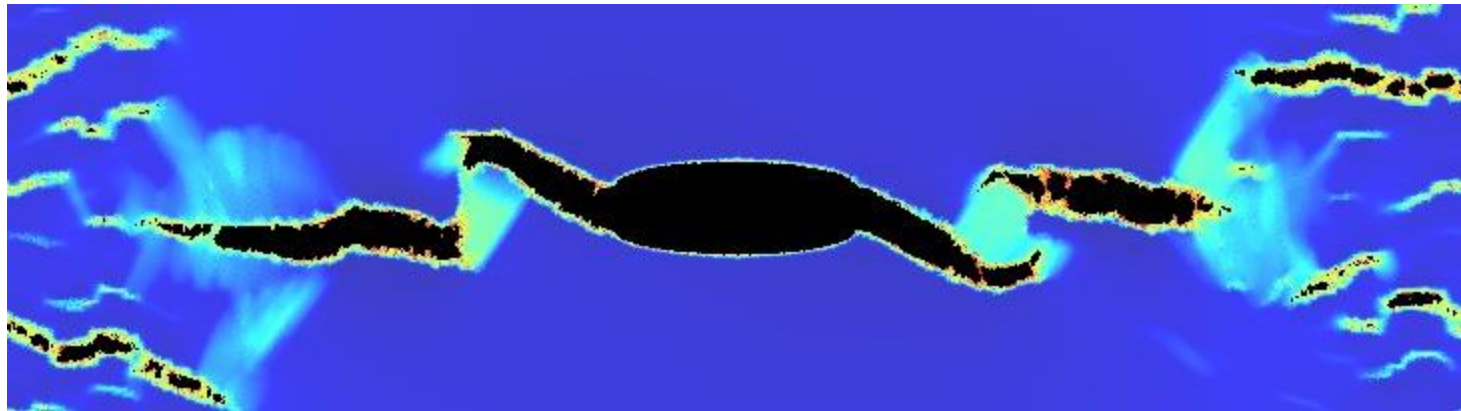
epot (eV)



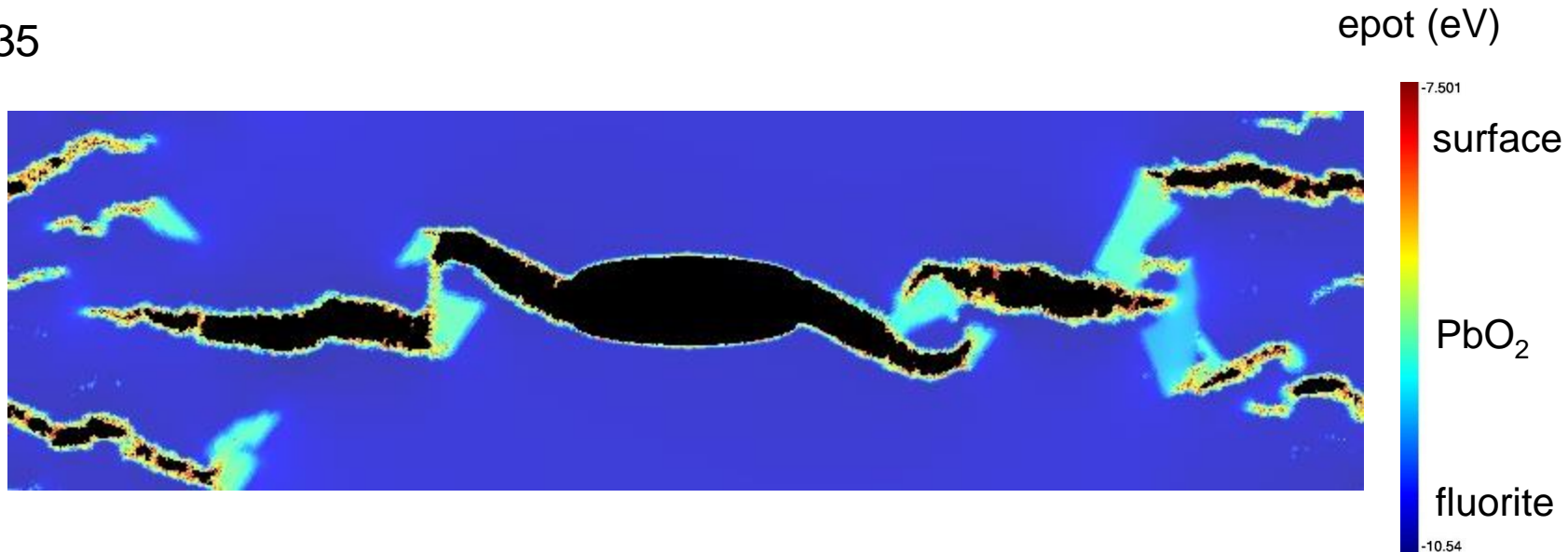
CRACK PROPAGATION - IN BLUK

$\varepsilon = 0.129$

epot (eV)



$\varepsilon = 0.135$



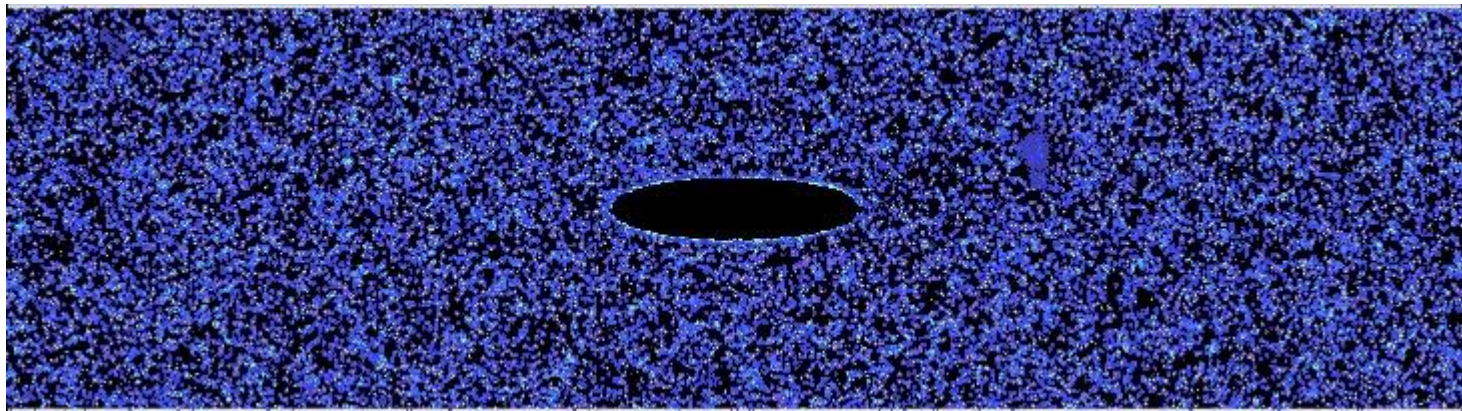
- Crack propagates by **cleavage** in the PbO₂ structure at the crack tip
- Sub-cracks appear, grow and coalesce in secondary phase

CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.036$

1.8% Frenkel pairs randomly distributed

epot (eV)



-6.329
surface

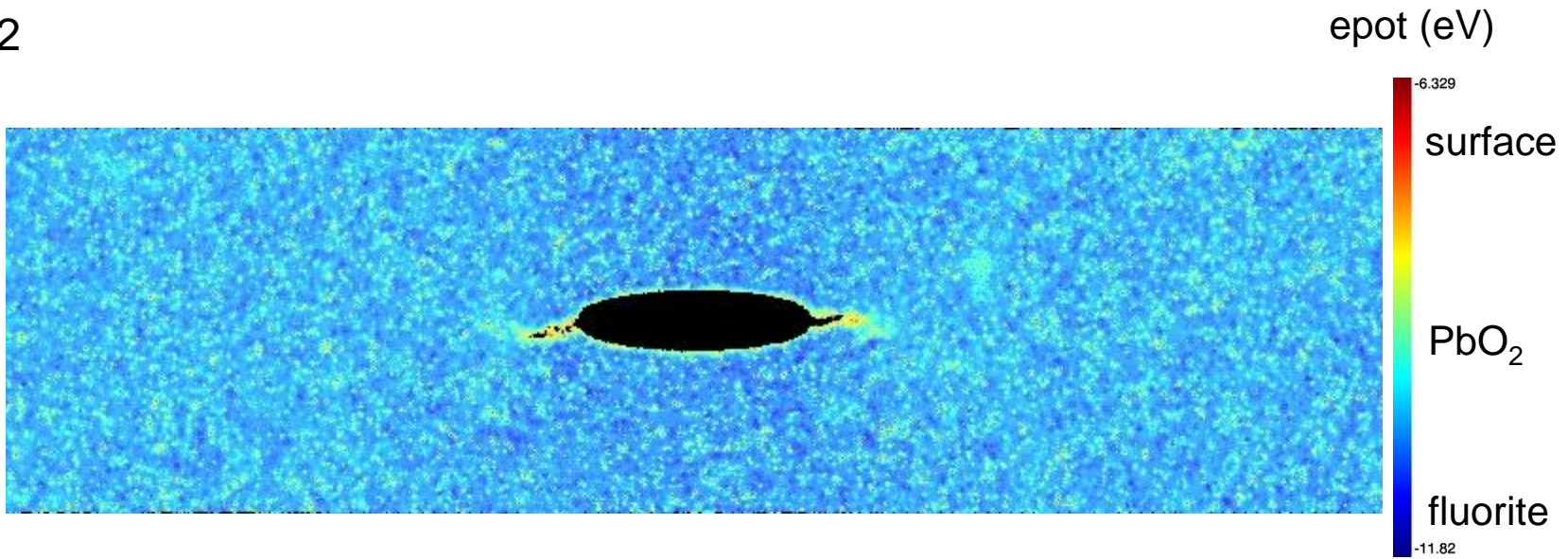
PbO₂

fluorite

-11.82

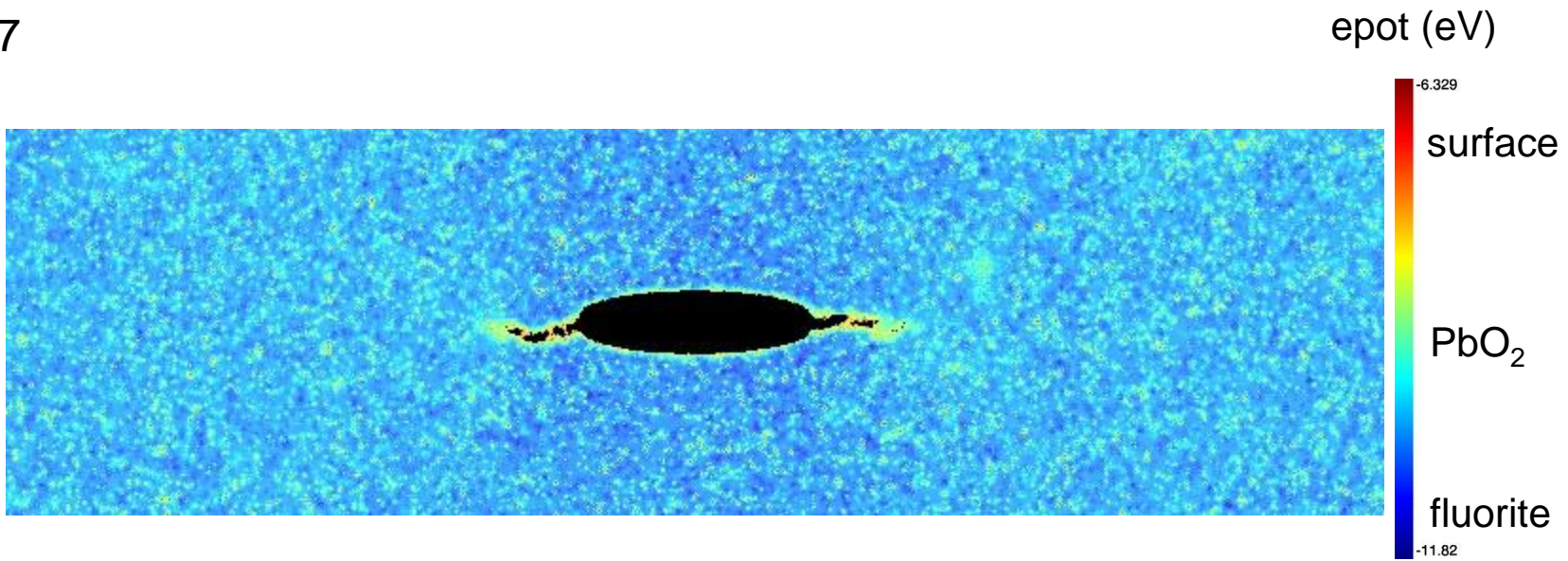
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.042$



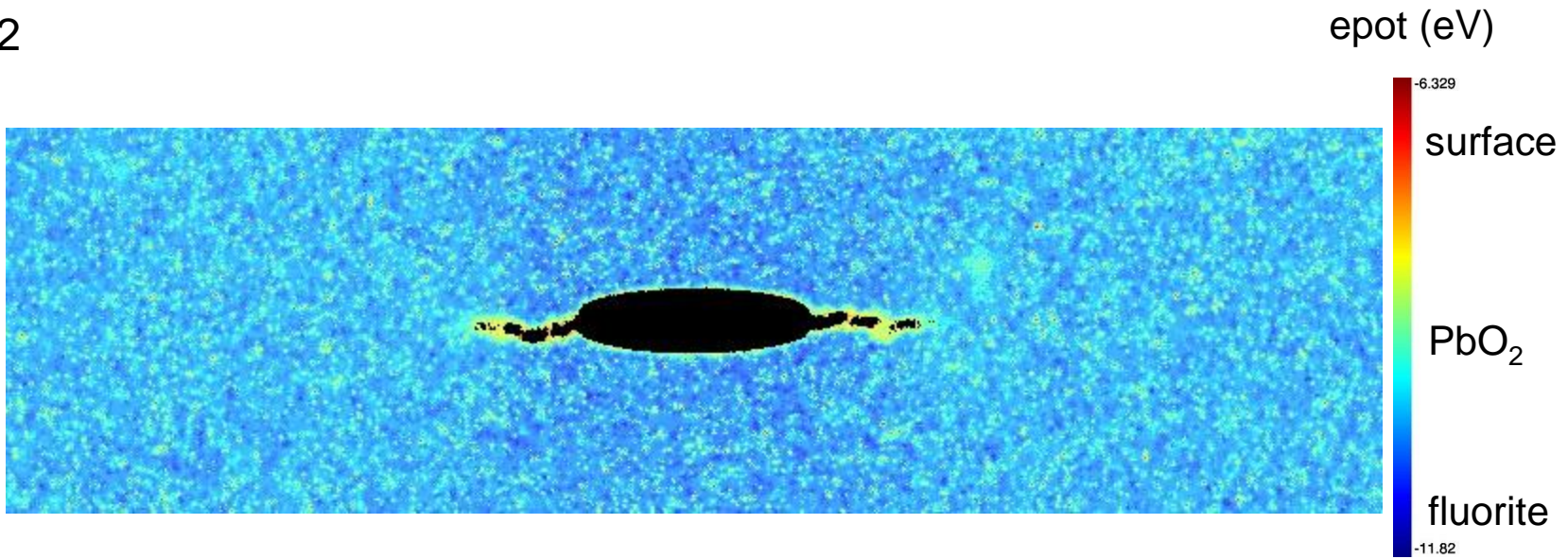
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.047$



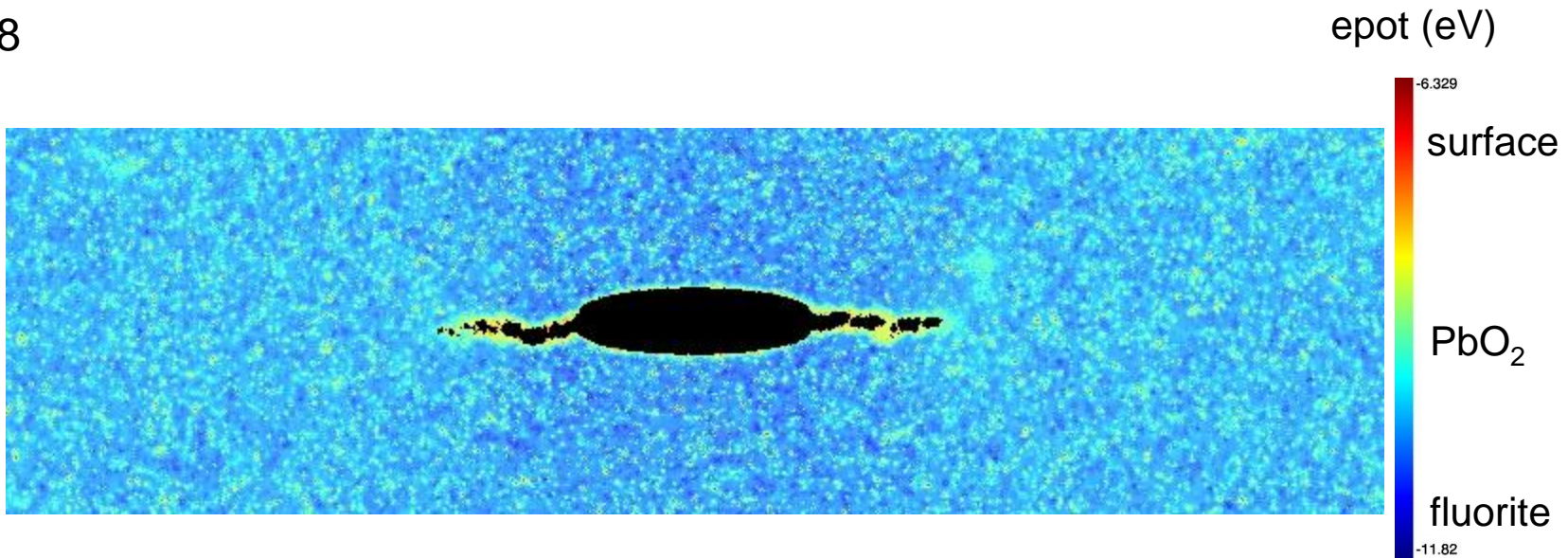
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.052$



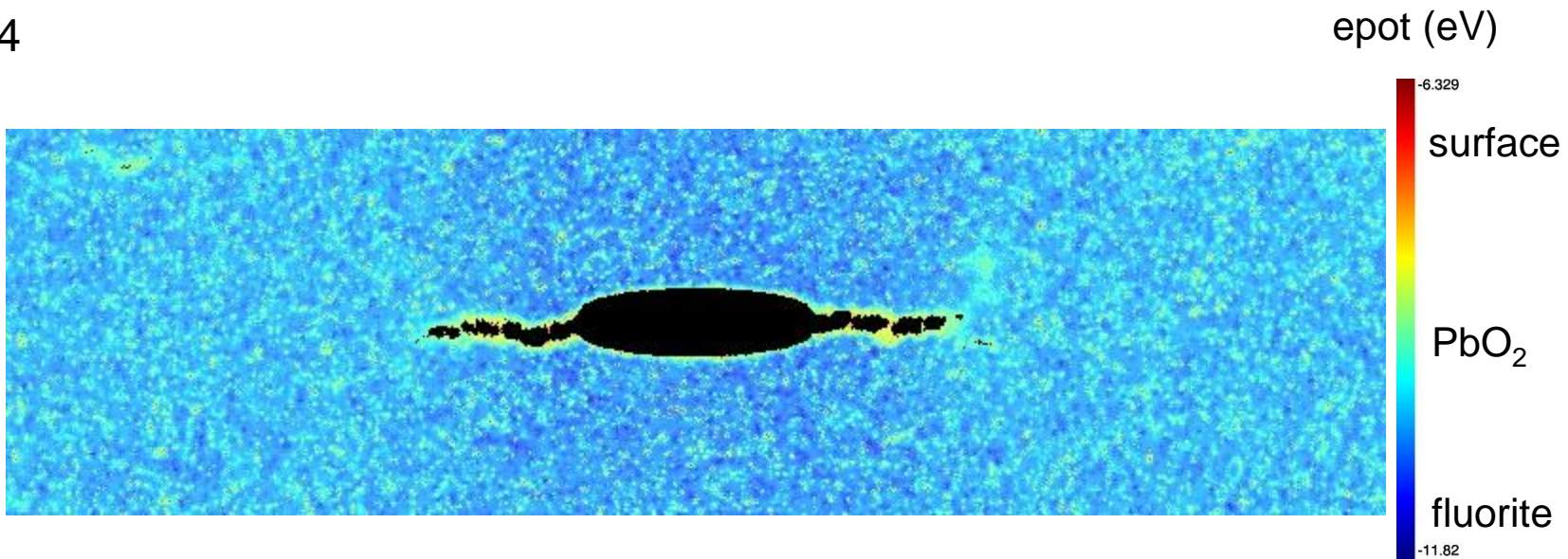
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.058$



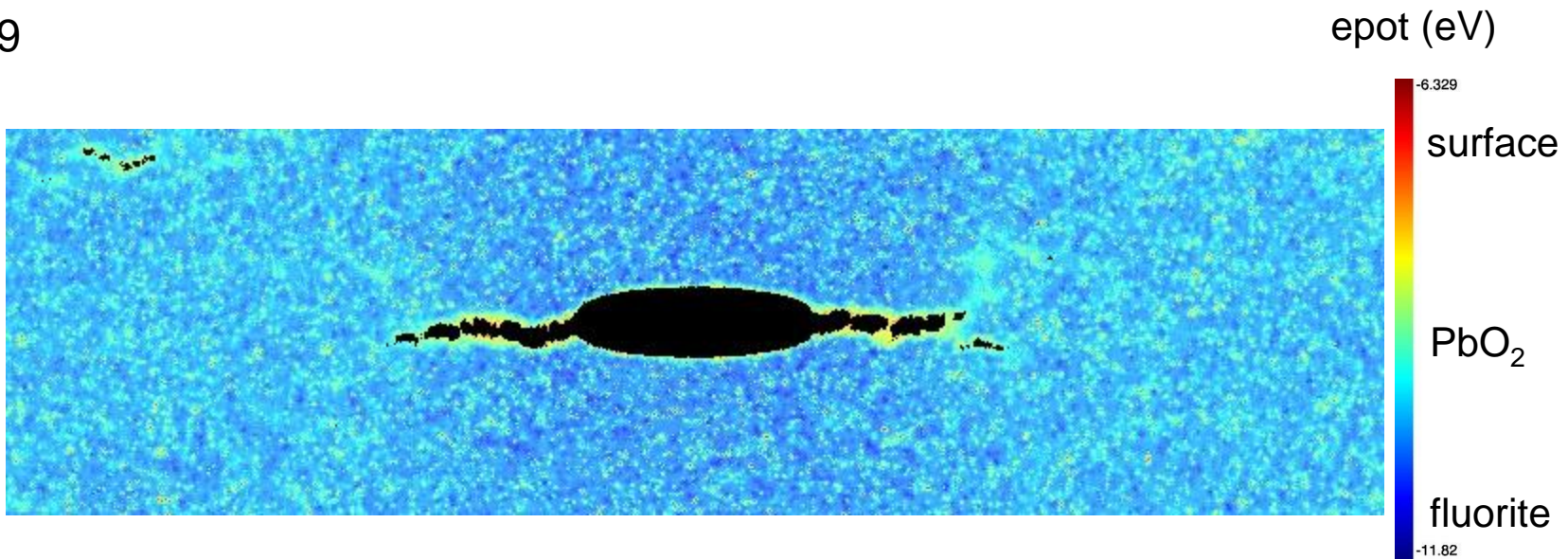
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.064$



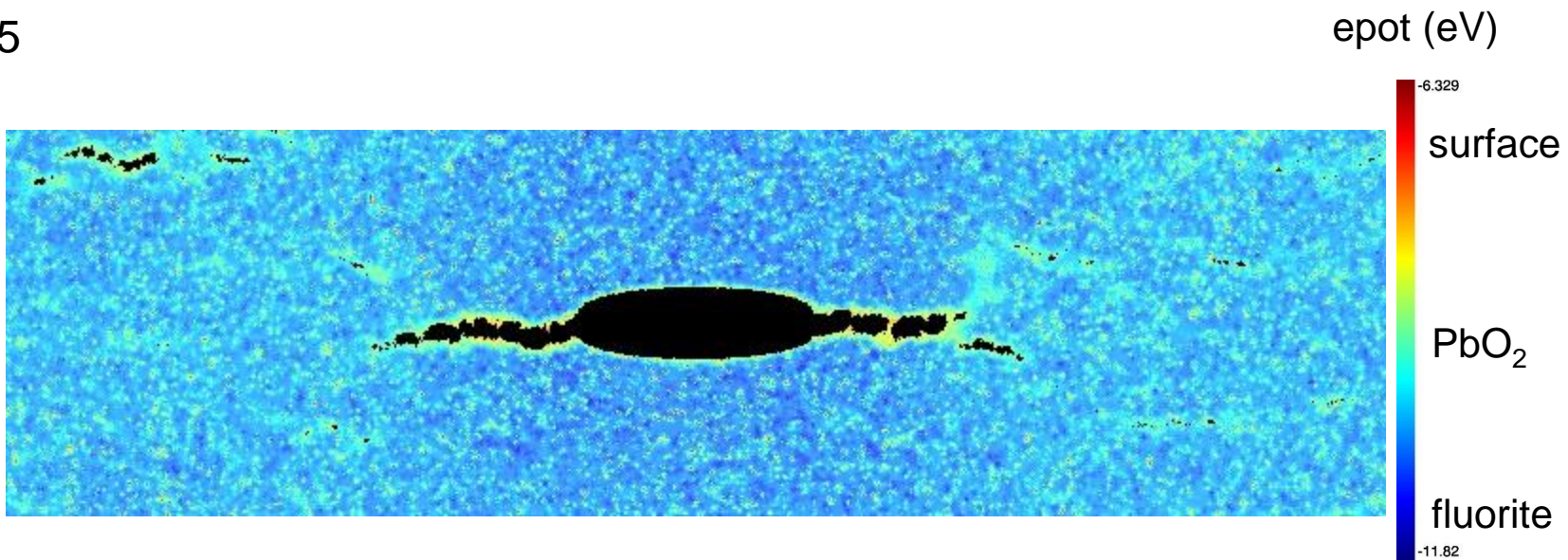
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.069$



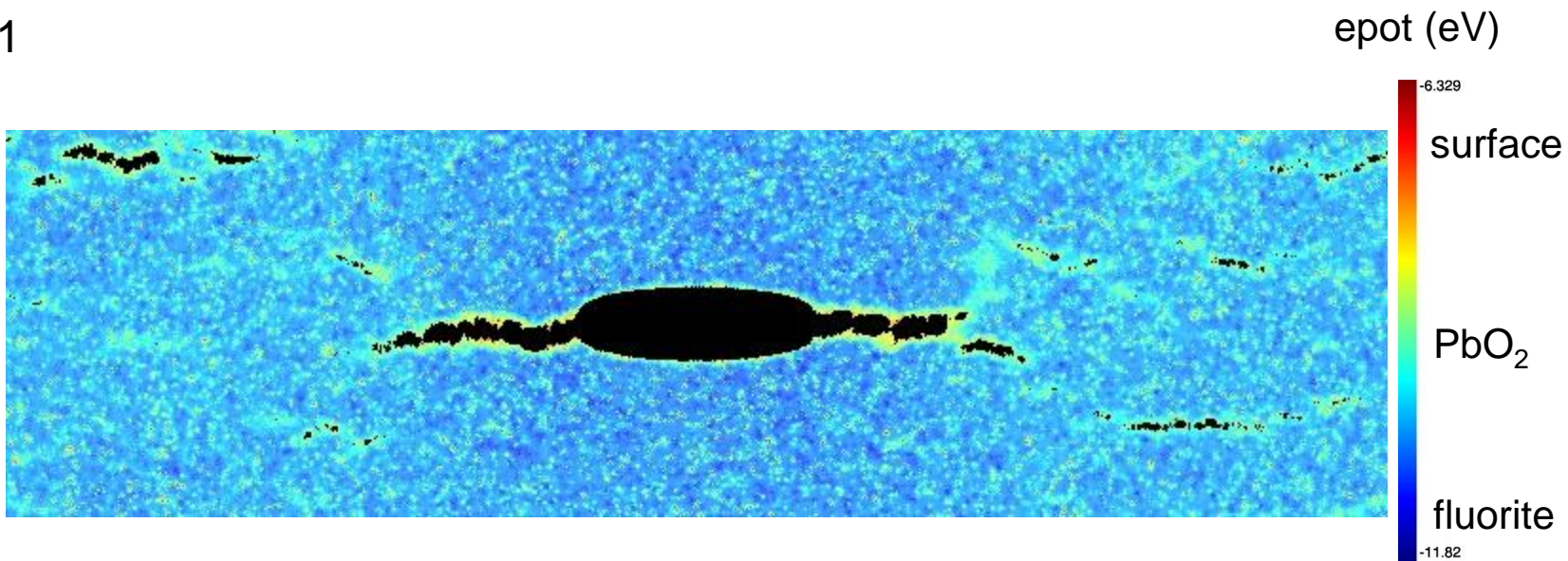
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.075$



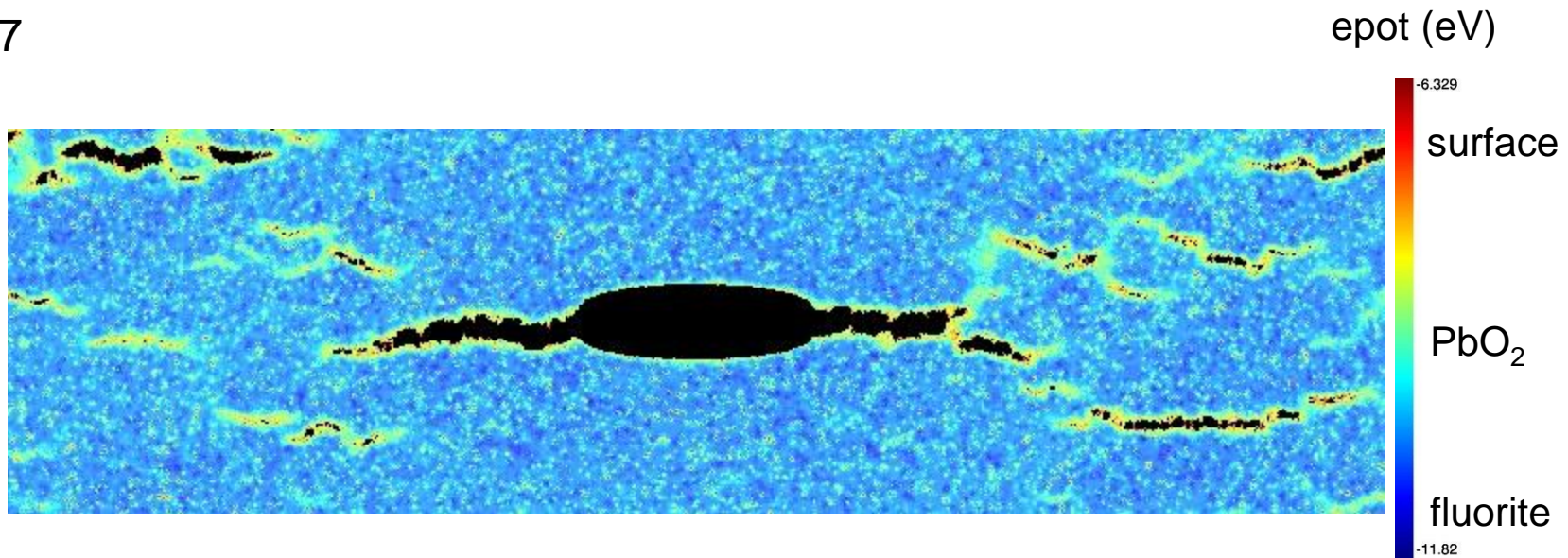
CRACK PROPAGATION - WITH POINT DEFECTS

$$\varepsilon = 0.081$$



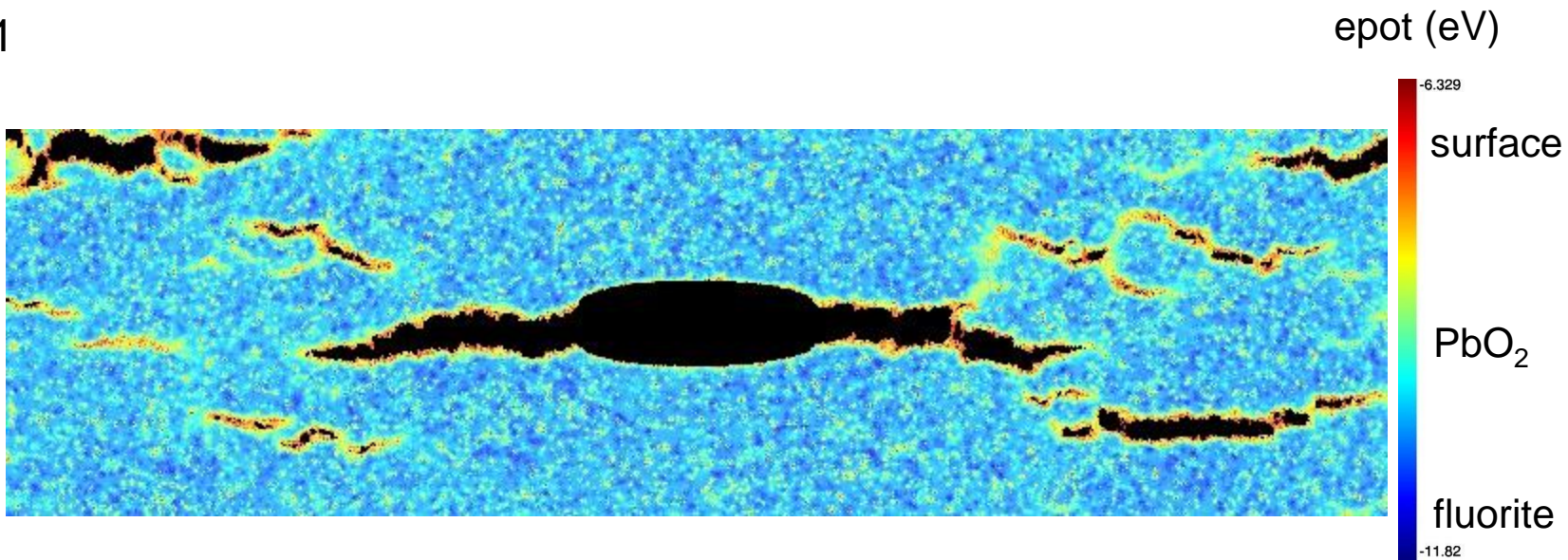
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.087$



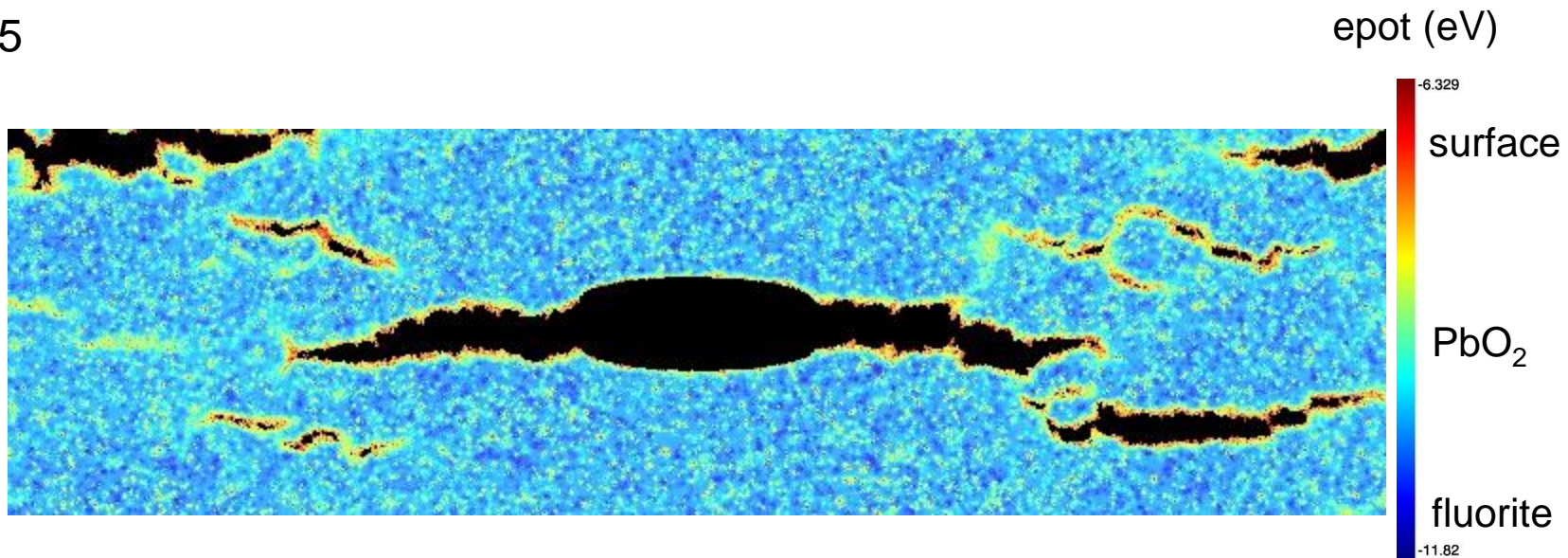
CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.111$



CRACK PROPAGATION - WITH POINT DEFECTS

$\varepsilon = 0.135$



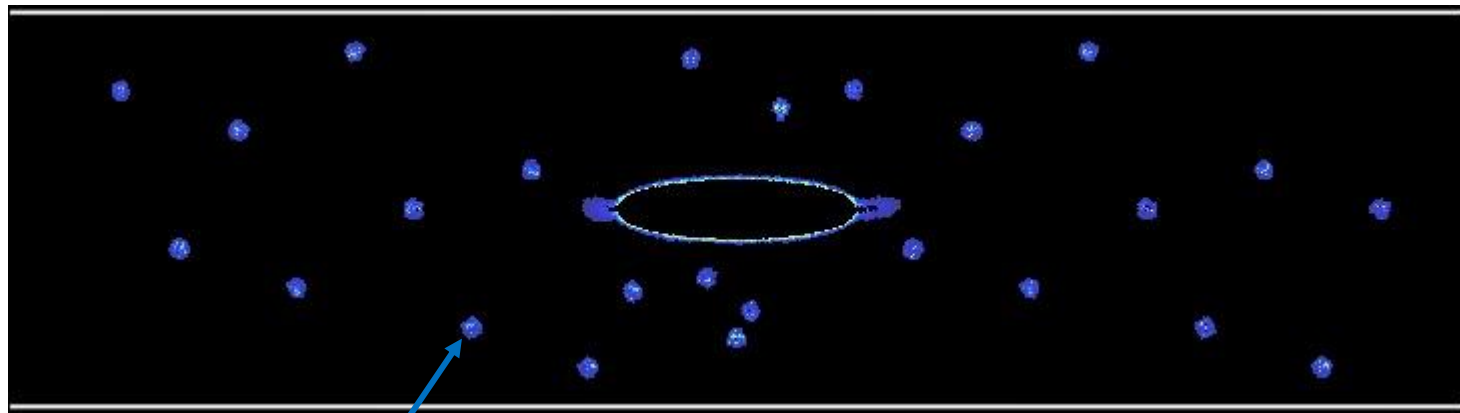
- No phase transition
- Crack propagates by **growth** and **coalescence** of close sub-cracks

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)



surface

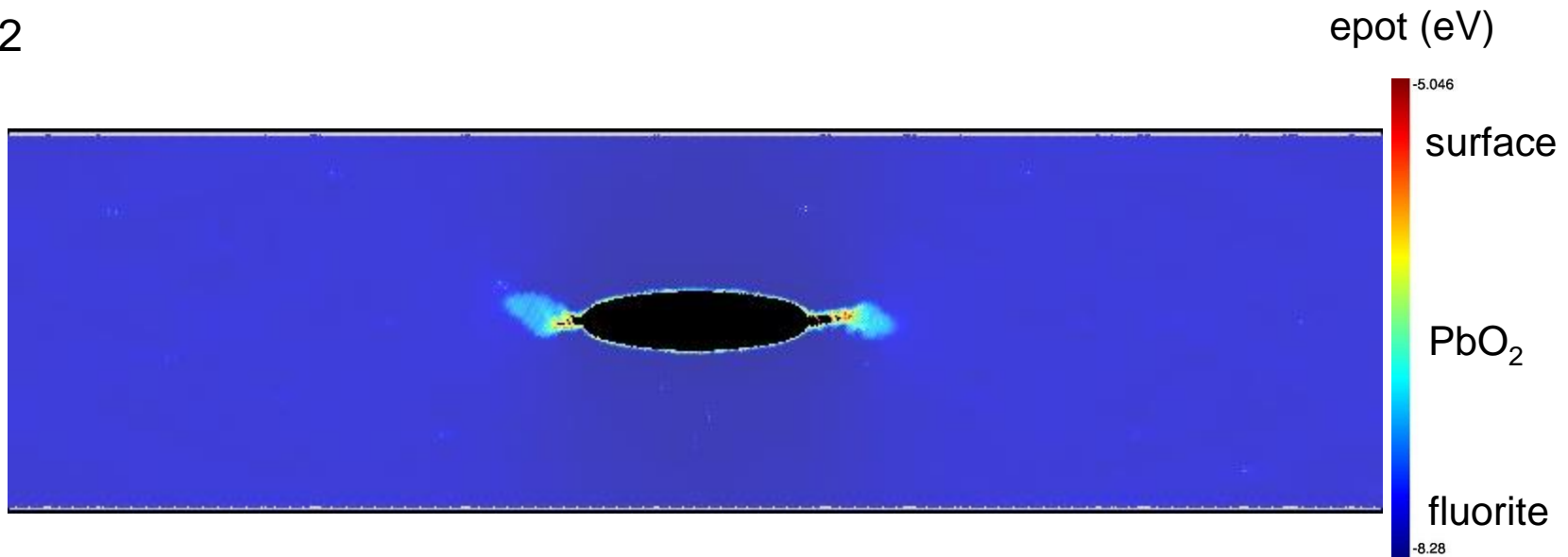
PbO₂

fluorite

He bubble

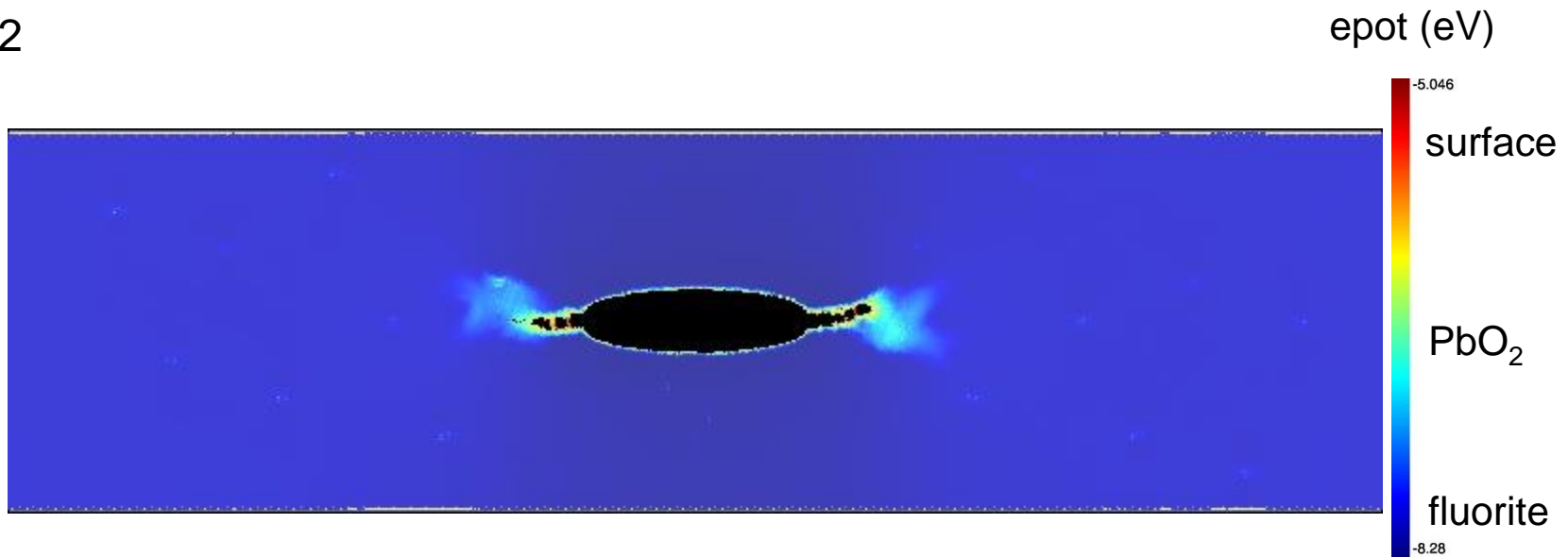
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.042$



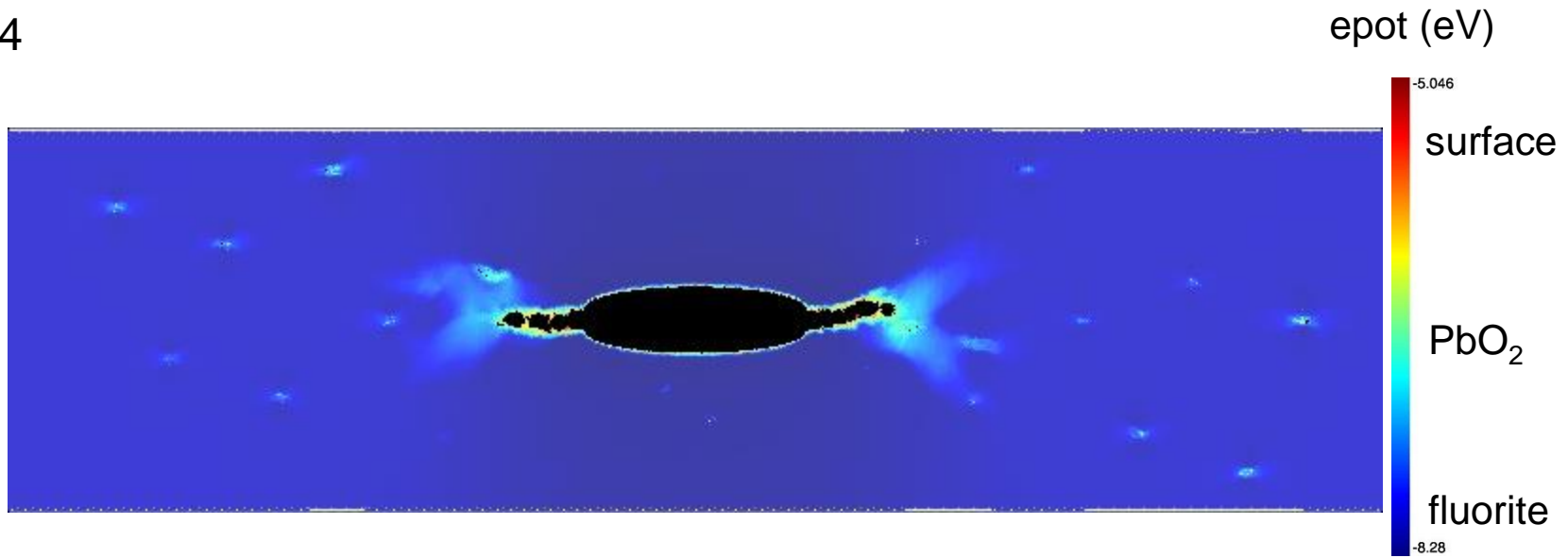
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.052$



CRACK PROPAGATION - WITH HE BUBBLES

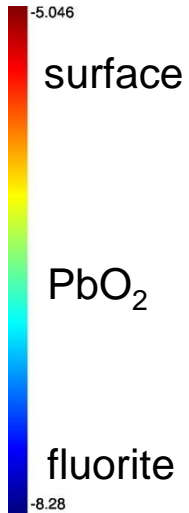
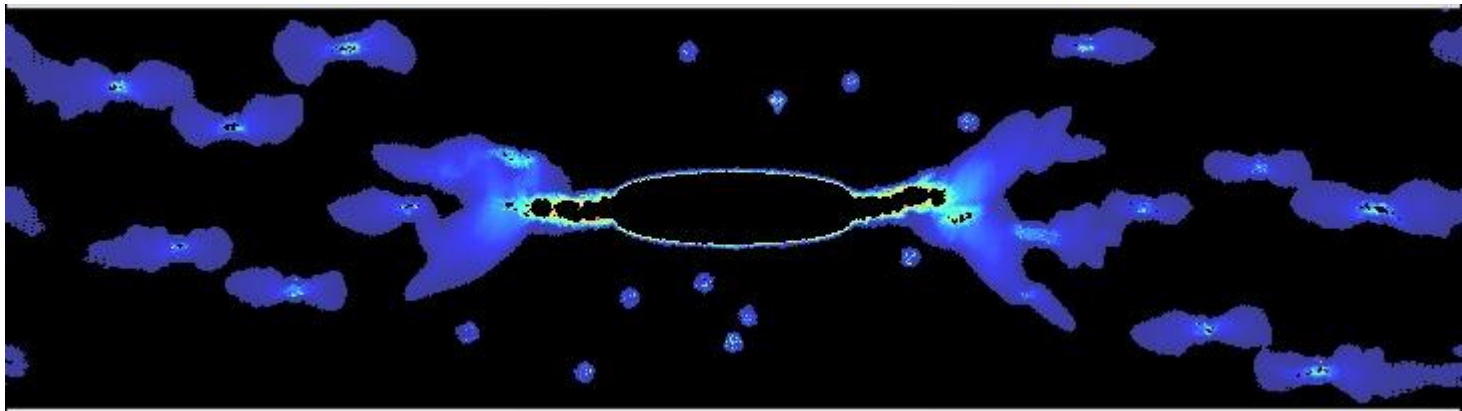
$\varepsilon = 0.064$



CRACK PROPAGATION - WITH HE BUBBLES

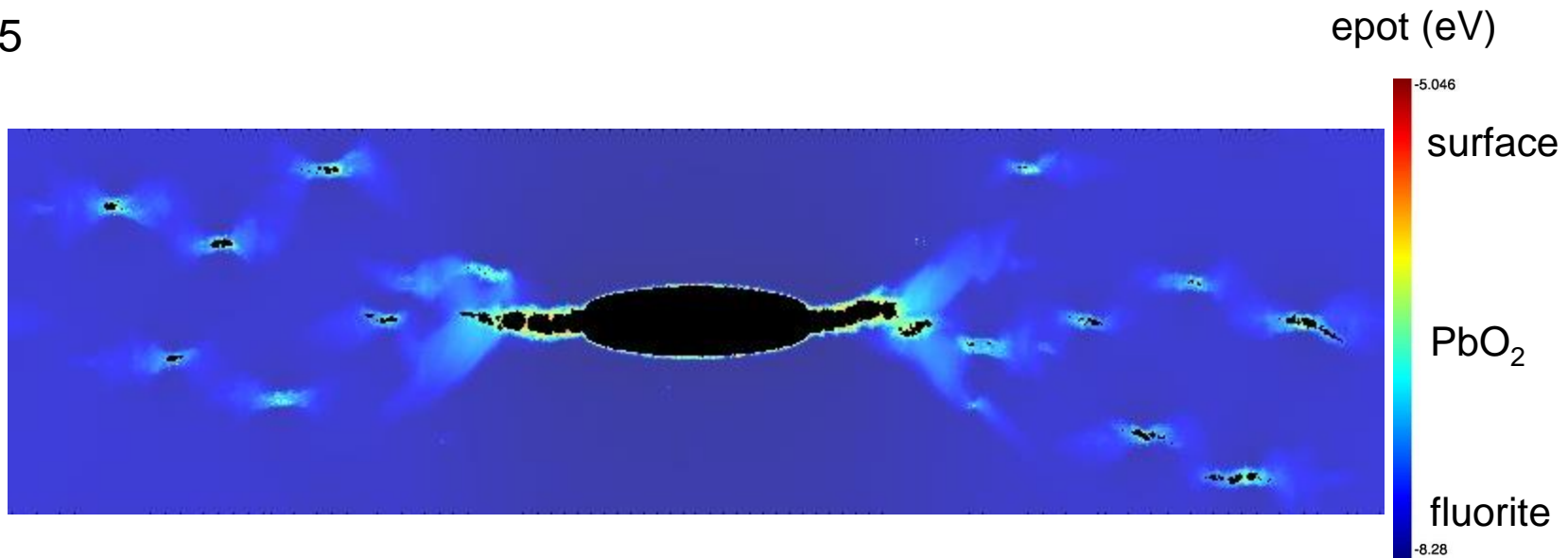
$\varepsilon = 0.069$

epot (eV)



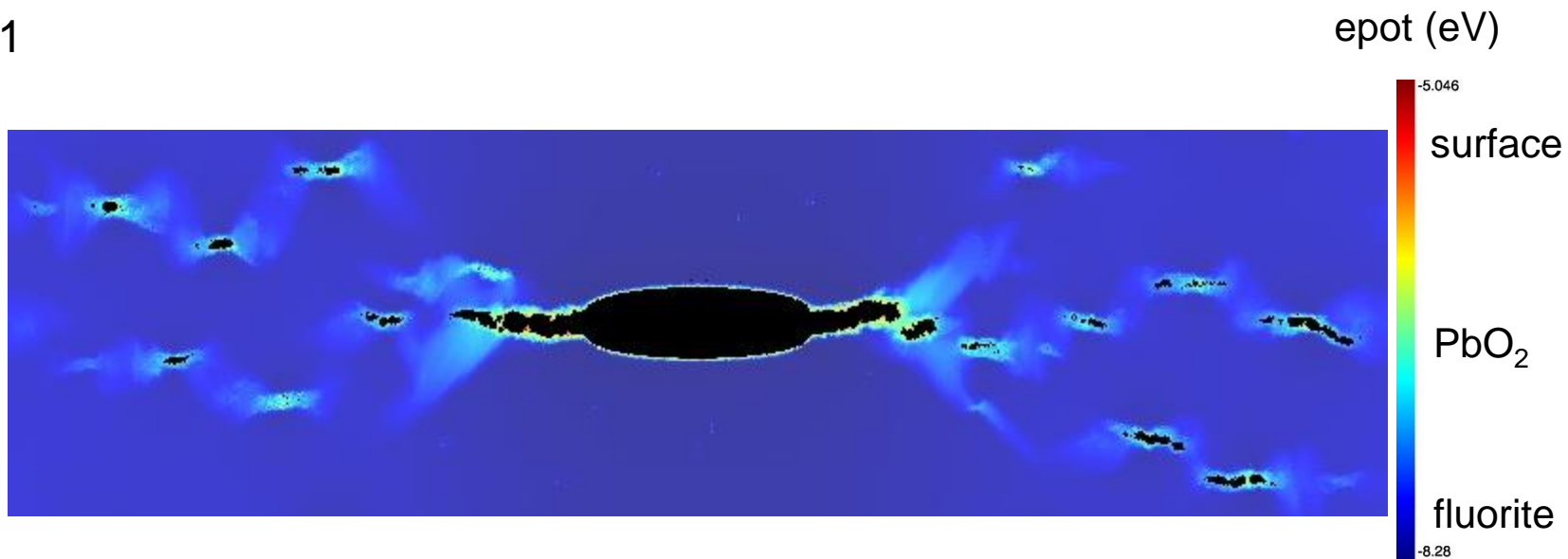
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.075$



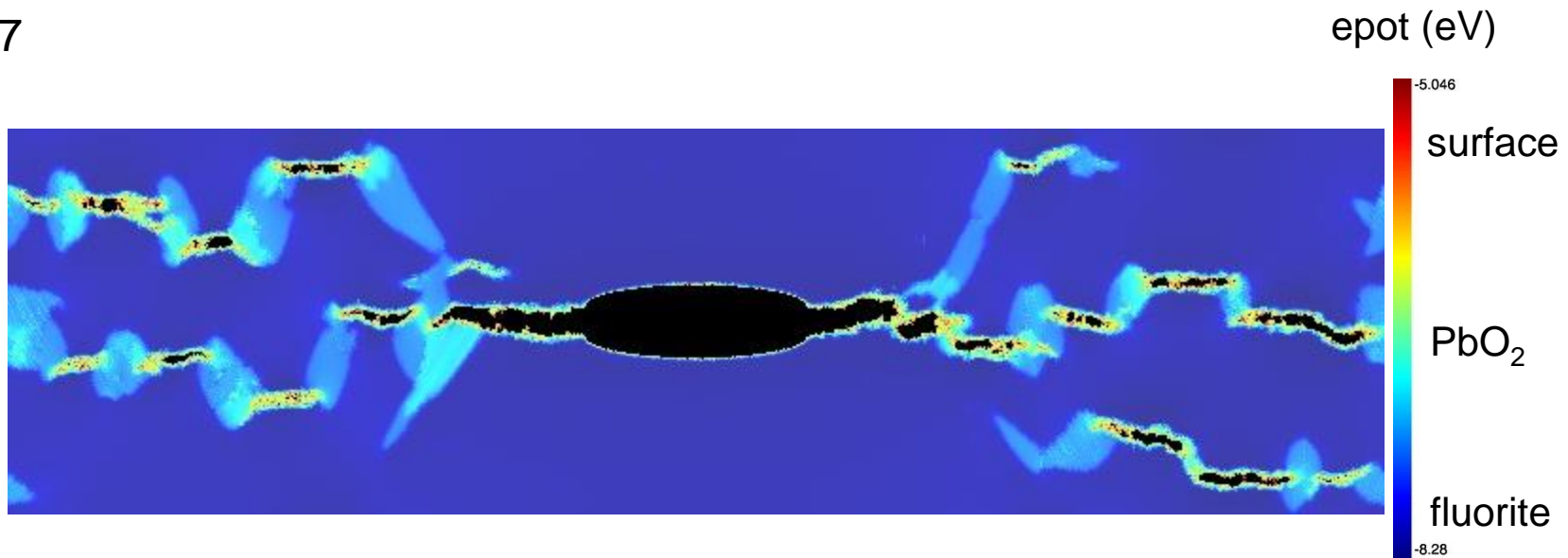
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.081$



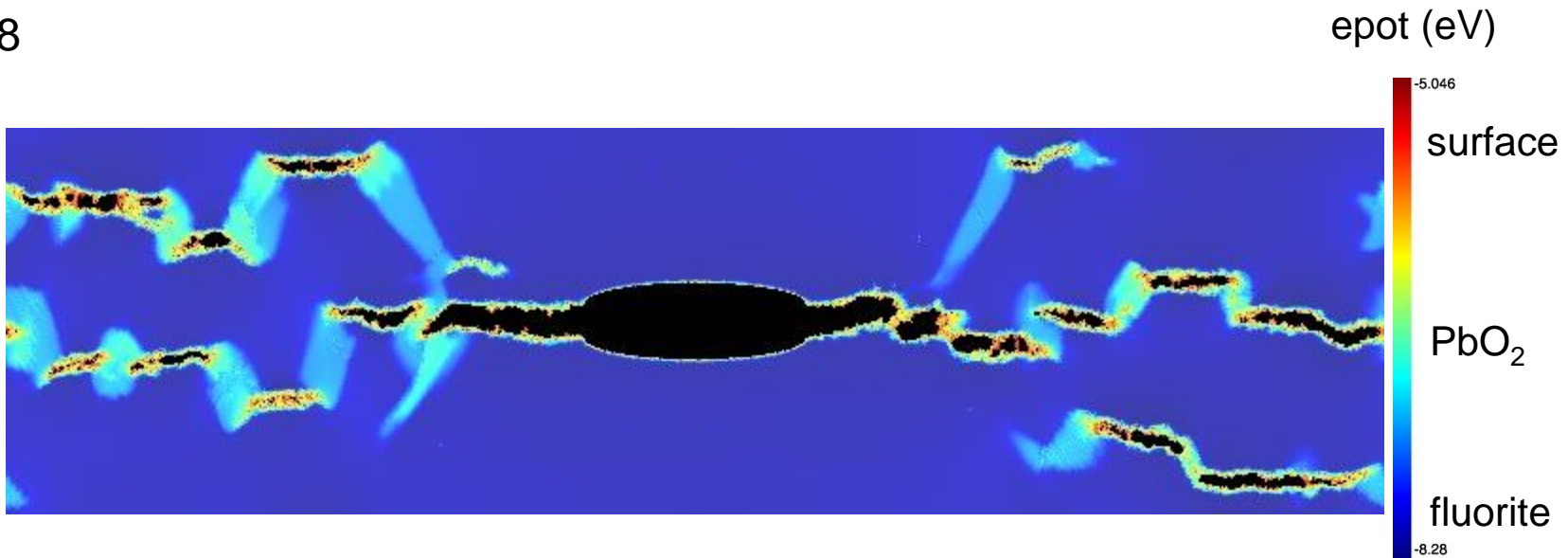
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.087$



CRACK PROPAGATION - WITH HE BUBBLES

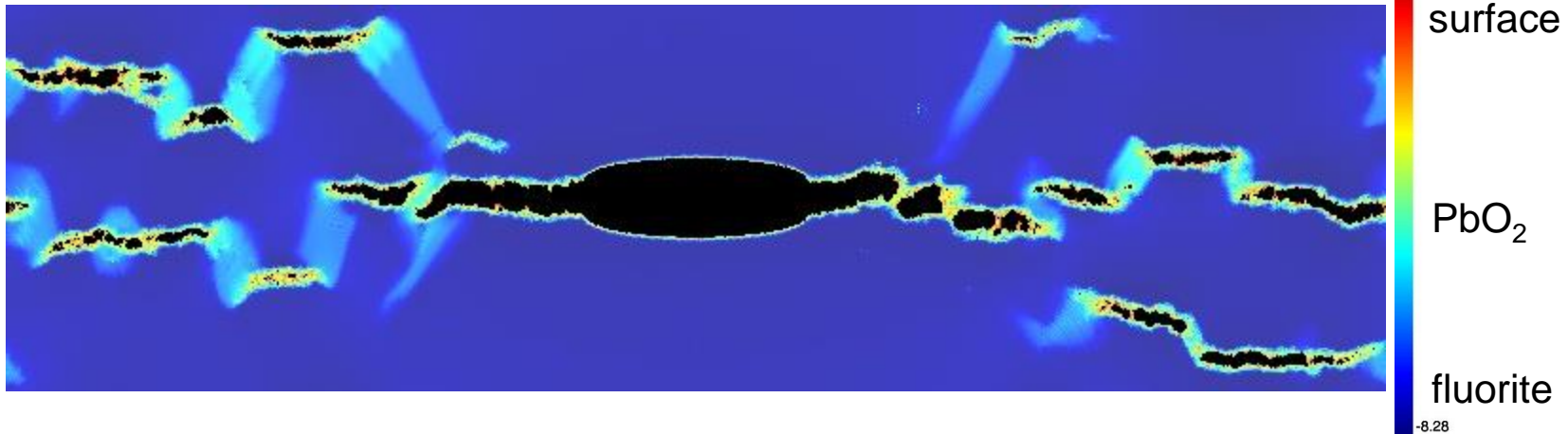
$\varepsilon = 0.098$



CRACK PROPAGATION - WITH HE BUBBLES

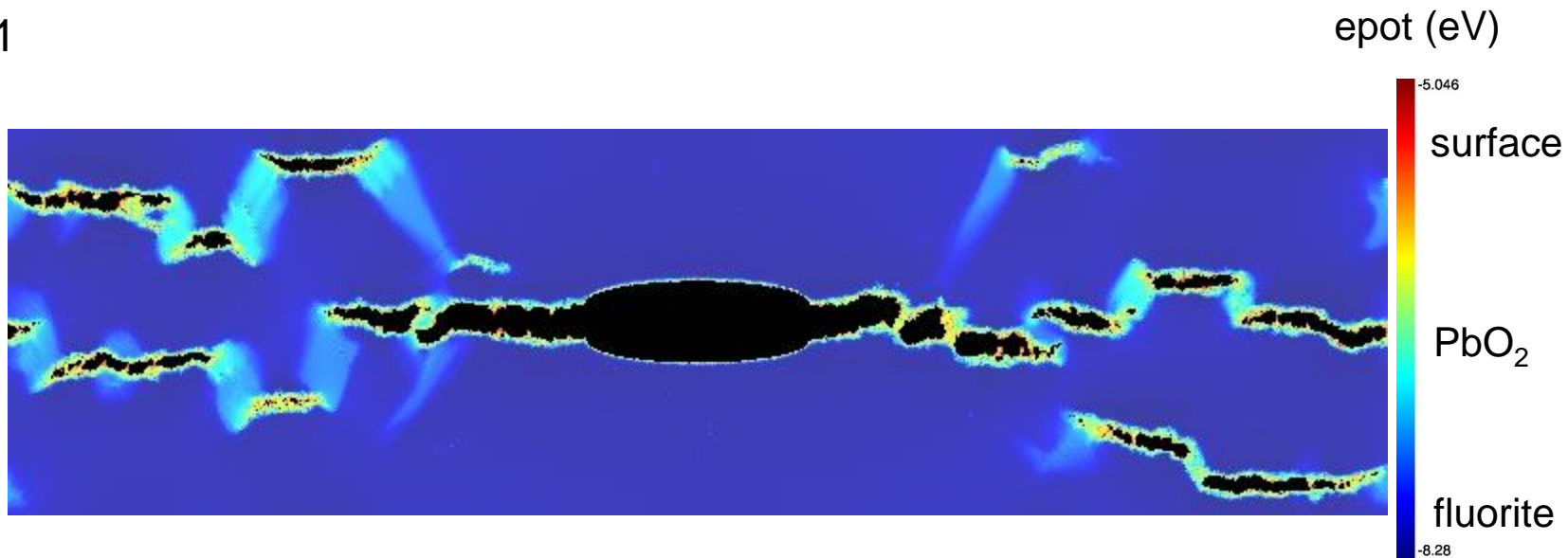
$$\varepsilon = 0.104$$

epot (eV)



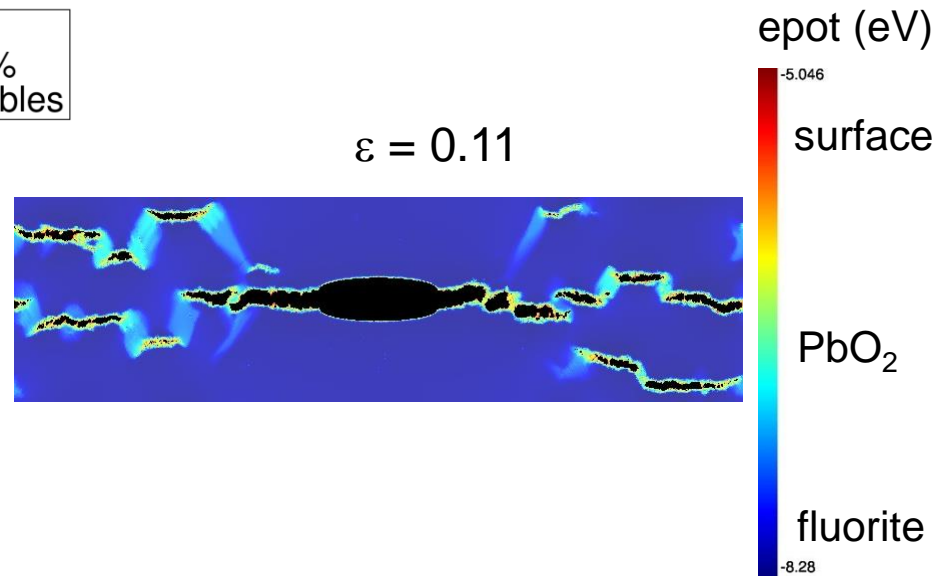
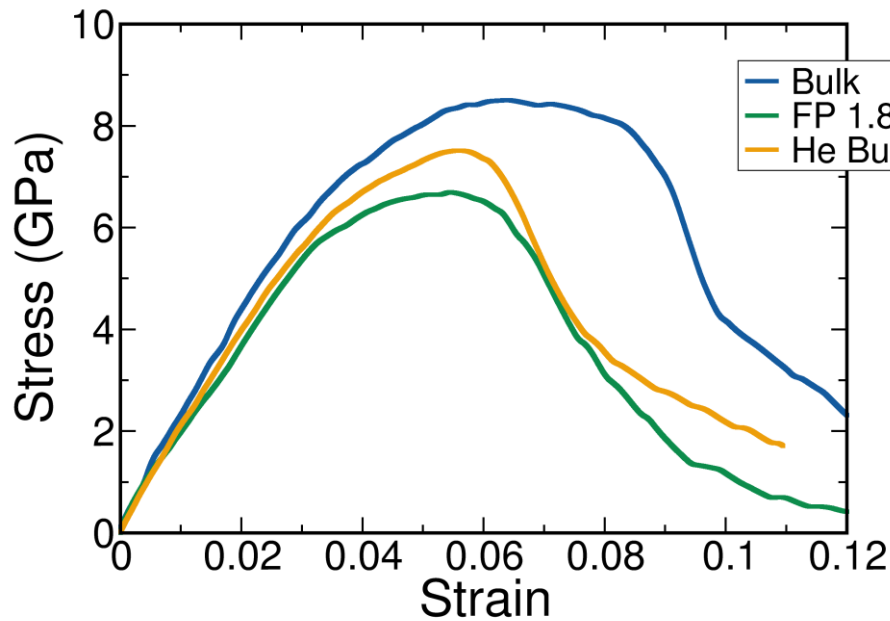
CRACK PROPAGATION - WITH HE BUBBLES

$\varepsilon = 0.111$



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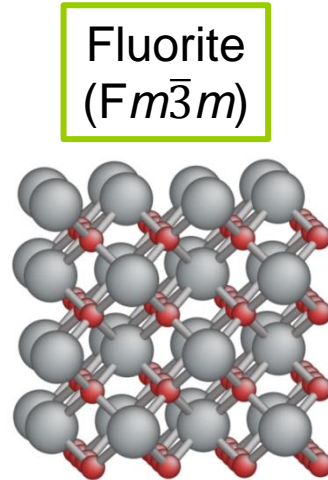
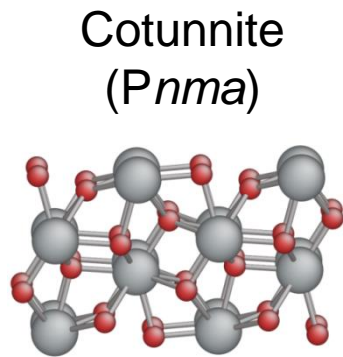
Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

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

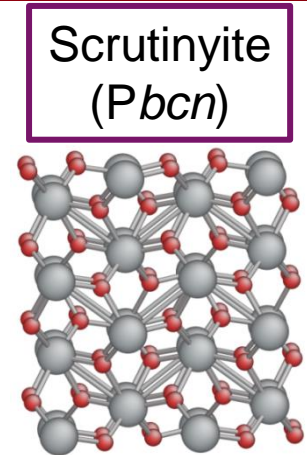
PHASE TRANSITIONS UNDER COMPRESSION

● oxygen

● uranium

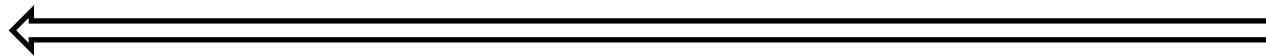
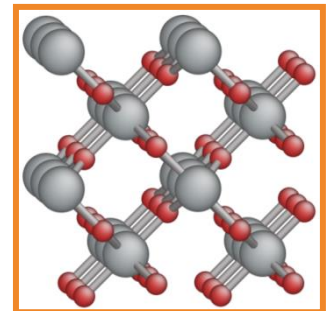


$\langle 100 \rangle$



Rutile
($P4_2/mnm$)

$\langle 110 \rangle$

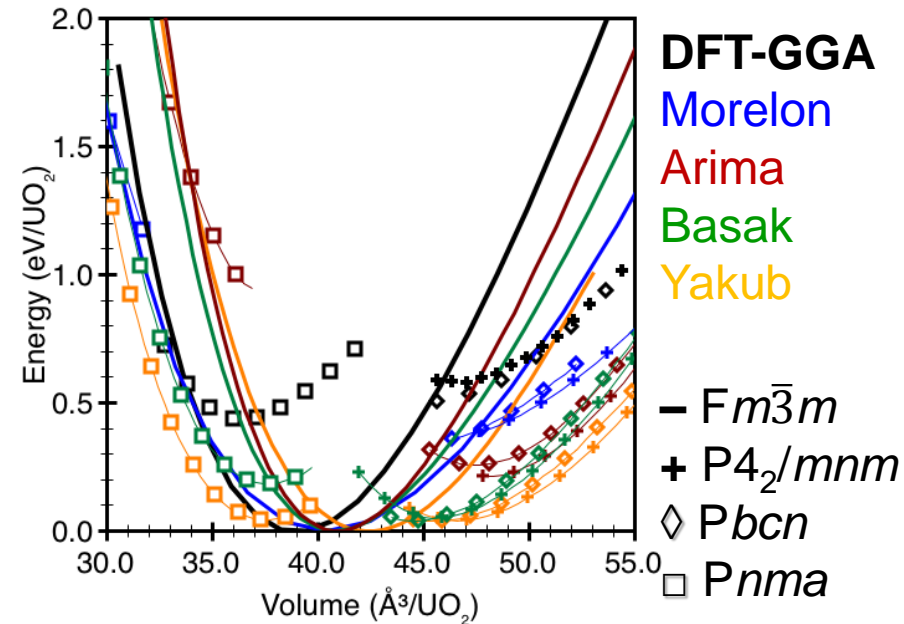


compression

tension



State equations



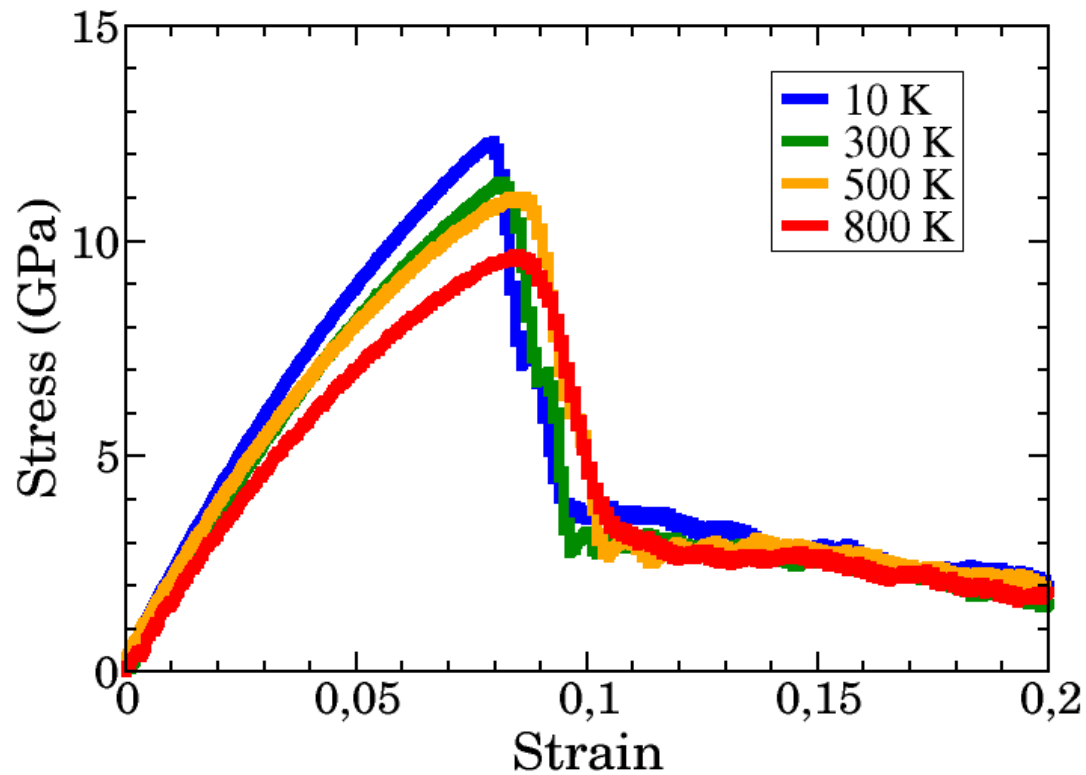
Transition Pressures (GPa)

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

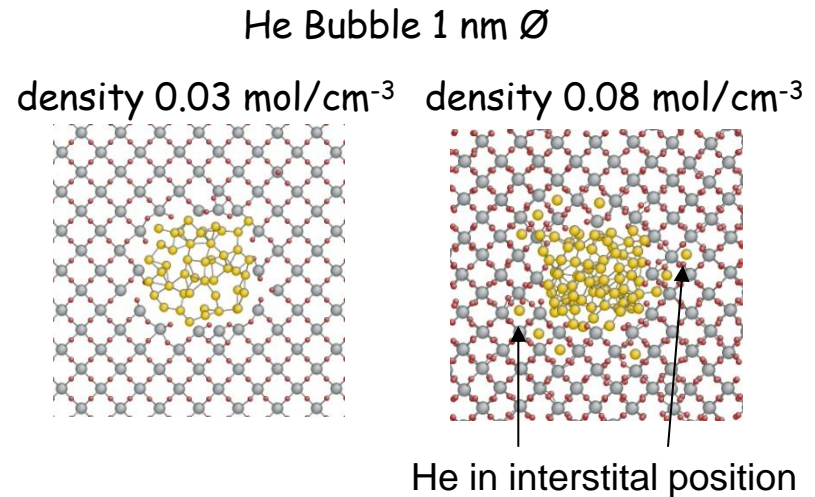
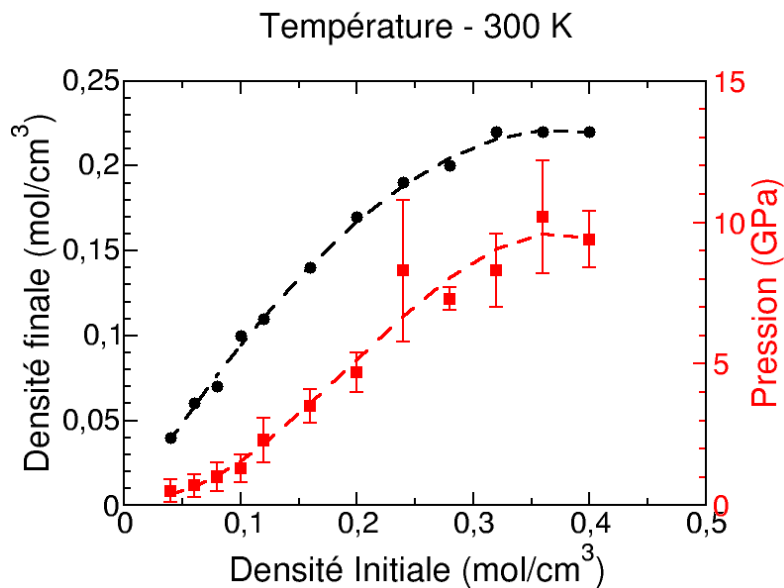


He Bubble - Strain rate $10^9/s$



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

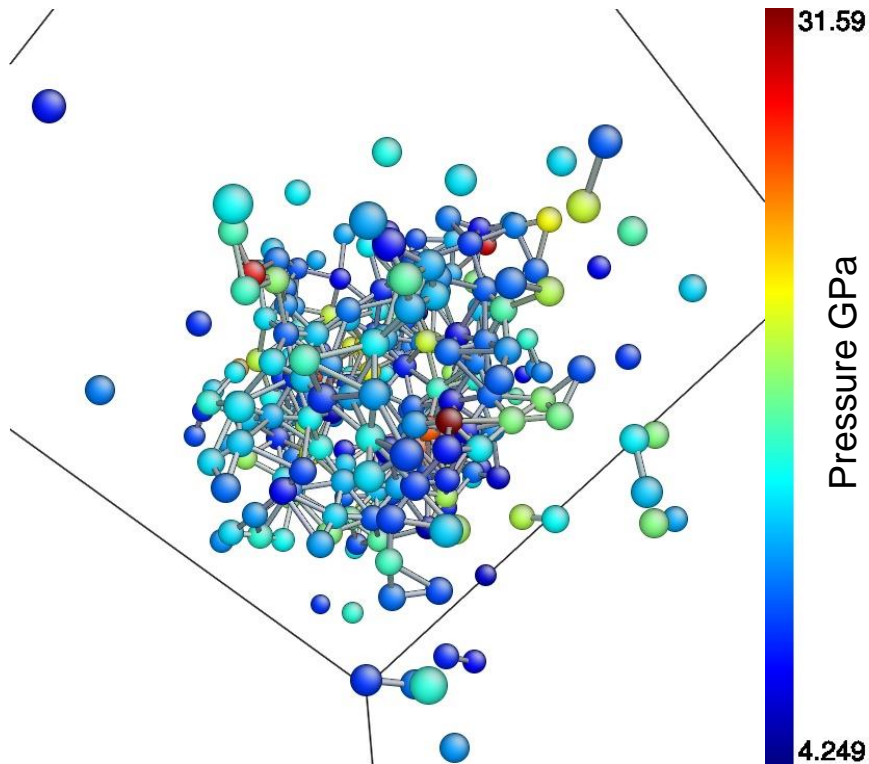
- Goal: study damage created by He bubble in UO_2 (defects, pressure, ...)
- Method: He Bubble in UO_2 (\neq density, size, temperature)
- Main results:



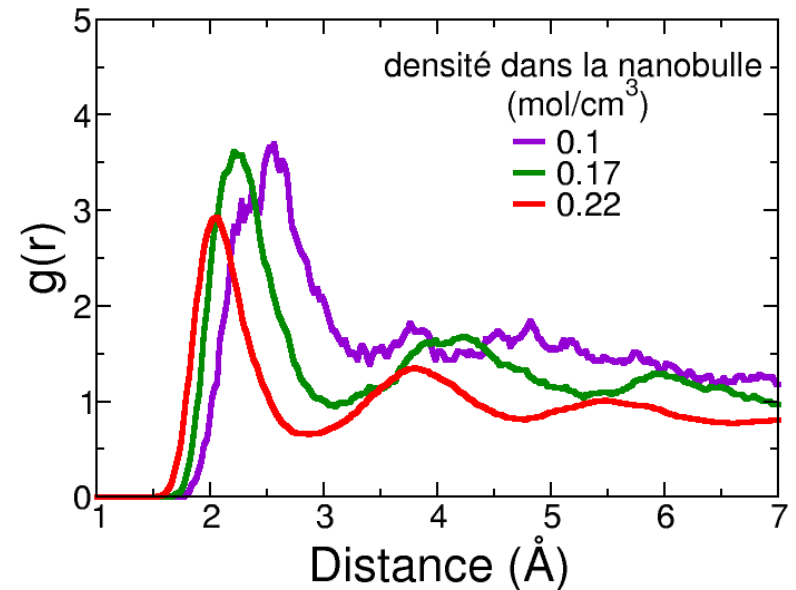
- No influence of the temperature (300 – 1500 K)
- He **partially soluble** in UO_2 (\rightarrow interstitial)
- Density threshold \approx **0.04 mol/cm⁻³** (= 1 He/Schottky)
- Max pressure in bubble **10 GPa**, He state equation \neq VdW
- Pressure at **0.04 mol/cm⁻³** = **500 MPa**

STRUCTURE IN HE BUBBLES

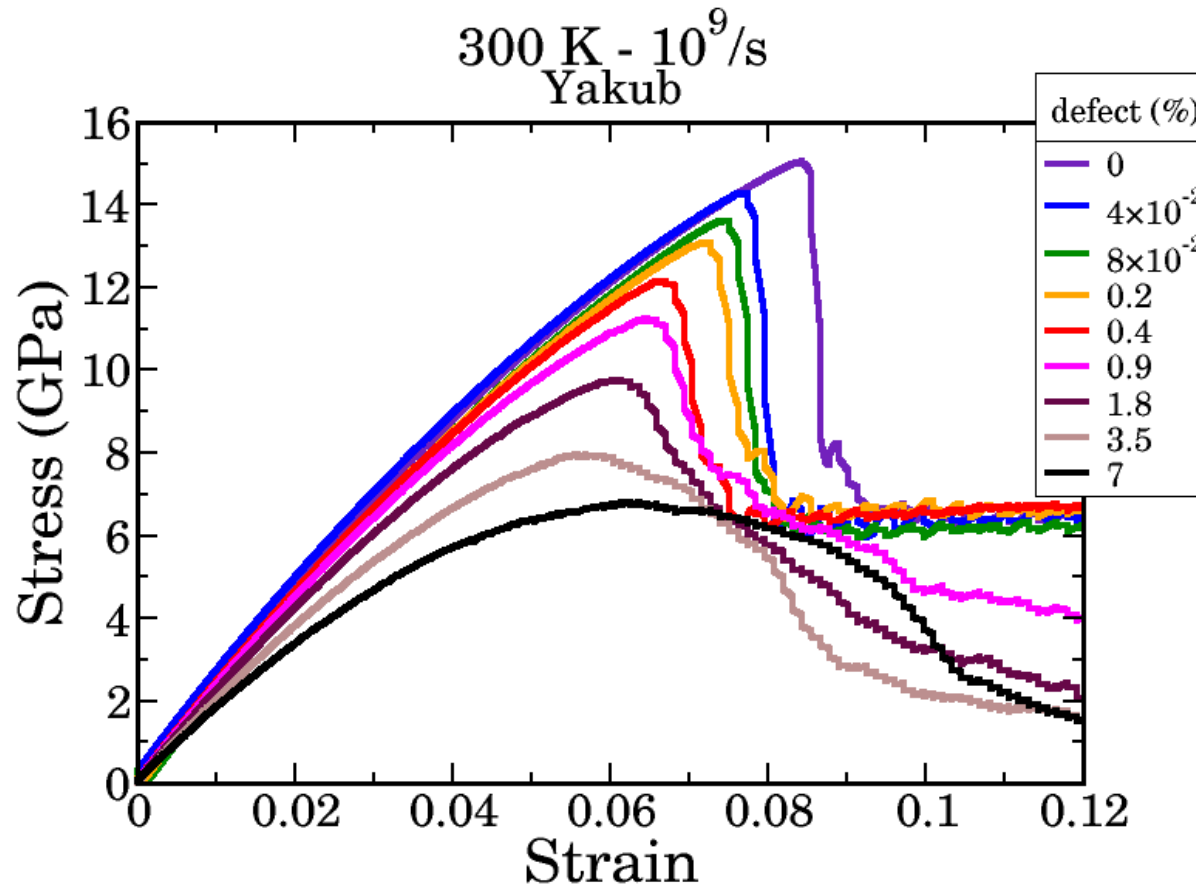
300 K – nanobubble 0.22 mol/cm³



Température - 300 K



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



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