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Deuterium trapping and release from high-temperature ion irradiated tungsten: experiments and reaction-diffusion simulations

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EUROfusion

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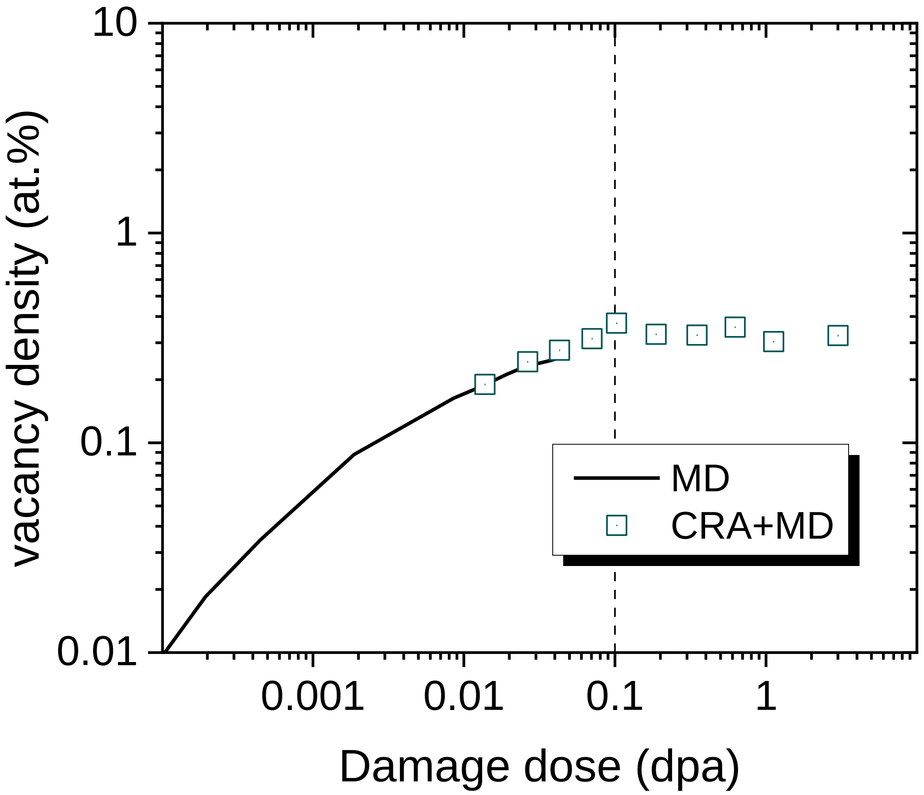
Motivation: trapped D concentration in W vs. damage dose
irradiation near room temperature

- Irradiation at <u>low temperatures</u>: vacancies are immobile, density of radiation defects <u>saturates</u> at damage doses > 0.1 dpa
- Damage is dominated by single vacancies and small vacancy clusters



MD: Molecular Dynamics
CRA: Creation Relaxation Algorithm

D.R. Mason et al., Phys. Rev. Mater. 5 (2021) 095403
M. Boleininger et al., Sci Rep 13 (2023) 1684

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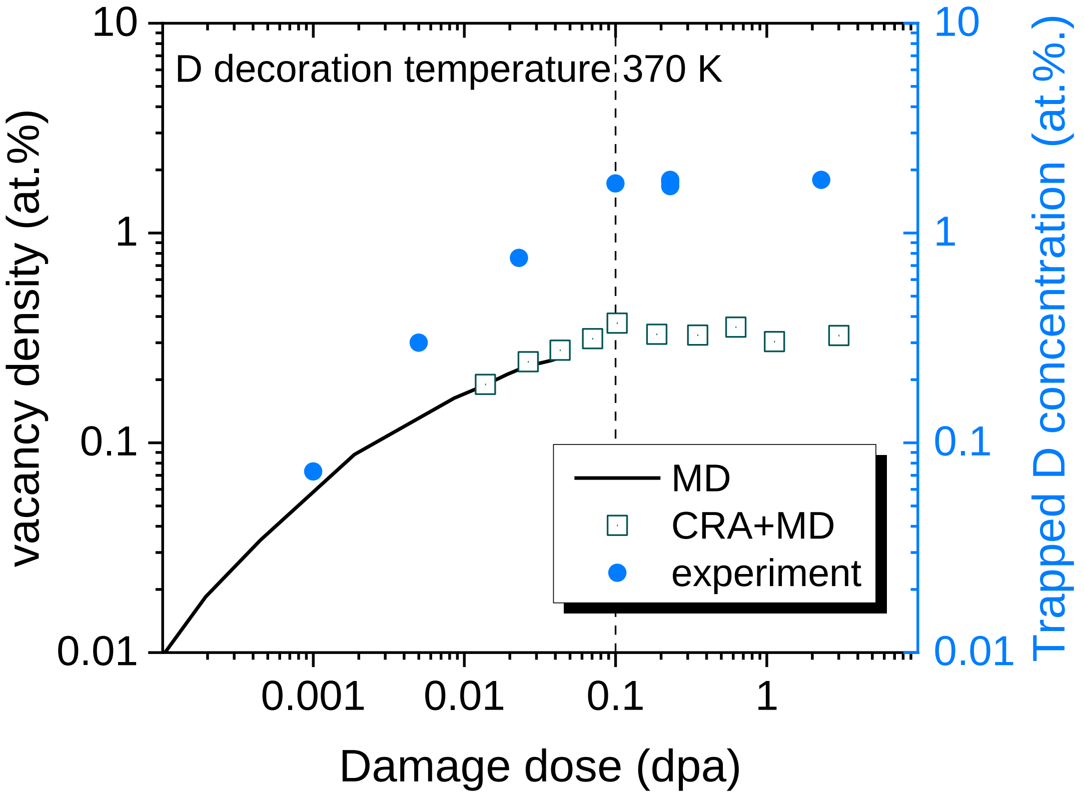
- Irradiation at low temperatures:
  vacancies are immobile,
  density of radiation defects saturates at damage doses > 0.1 dpa
- Damage is dominated by single vacancies and small vacancy clusters
- Corresponds to saturation of trapped D concentration in the defects



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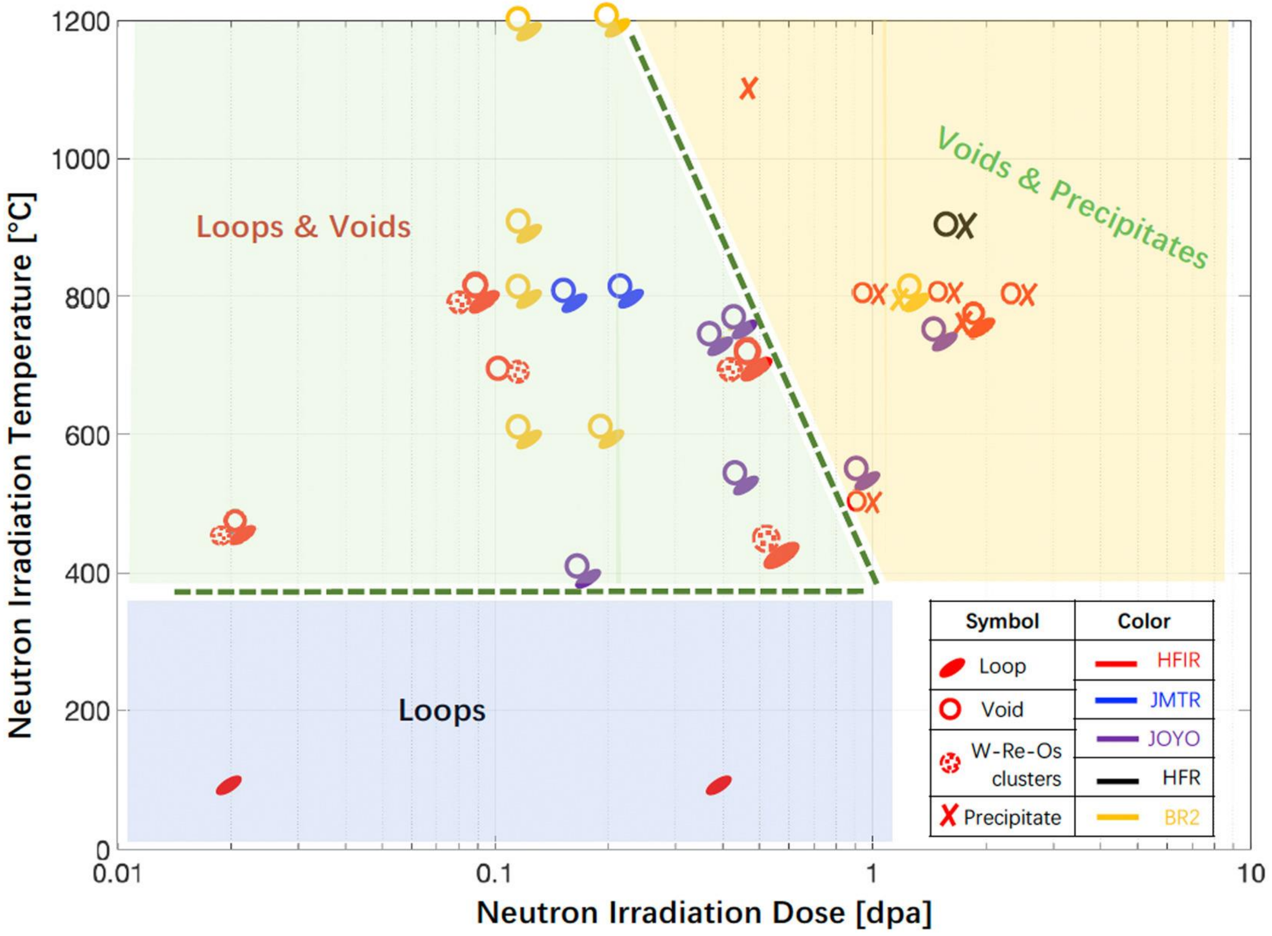
Motivation: microstructure of W vs. irradiation temperature

- Vacancies in W are mobile at temperatures > 600 K and can agglomerate into clusters
- Microstructure of W at high temperatures is dominated by nm-sized **voids**
- Limited data at doses > 1 dpa
- **Dose dependence of defect density and trapped D concentration at high temperatures?**

Fission neutron irradiation of tungsten



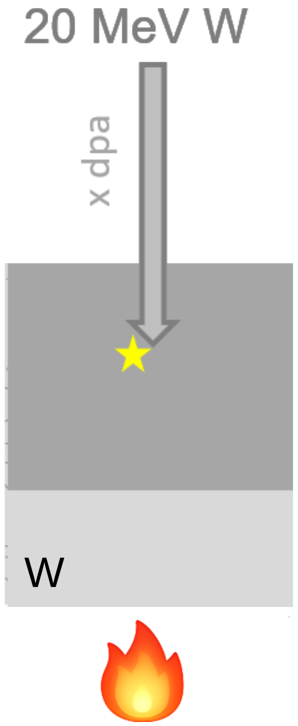
X. Hu, Journal of Nuclear Materials 568 (2022) 153856

MeV self-ion irradiation
to simulate displacement damage produced by fusion neutrons

1. Creating displacement damage

20 MeV W-ion irradiation to different damage doses (dpa) at 1350 K

raster-scanned (1 kHz) focused beam (2 mm), average dose rate: ~3×10^{-5} dpa/s



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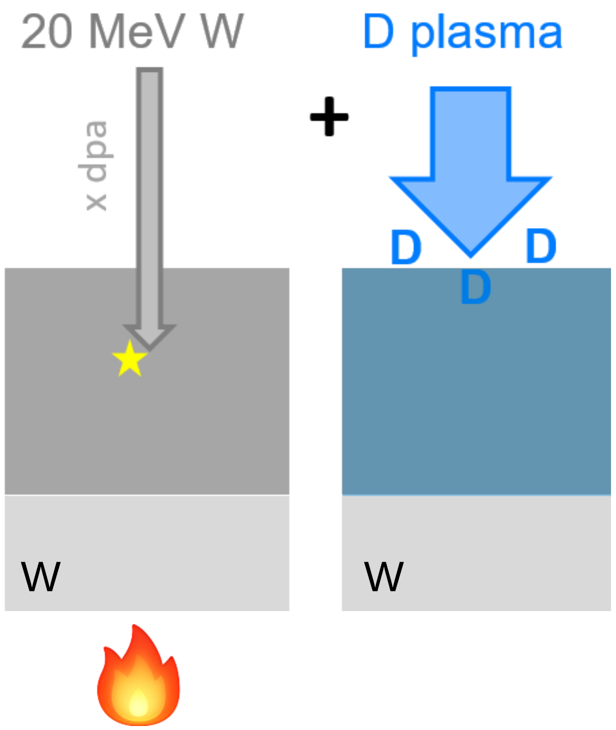

2. Decorating damage with deuterium

T_{sample} = 370 K, E_{ion} < 5 eV/D,

$\Gamma_{ion} < 10^{20}$ D/(m^{2}s), $\Phi_{ion} > 10^{25}$ D/m^2

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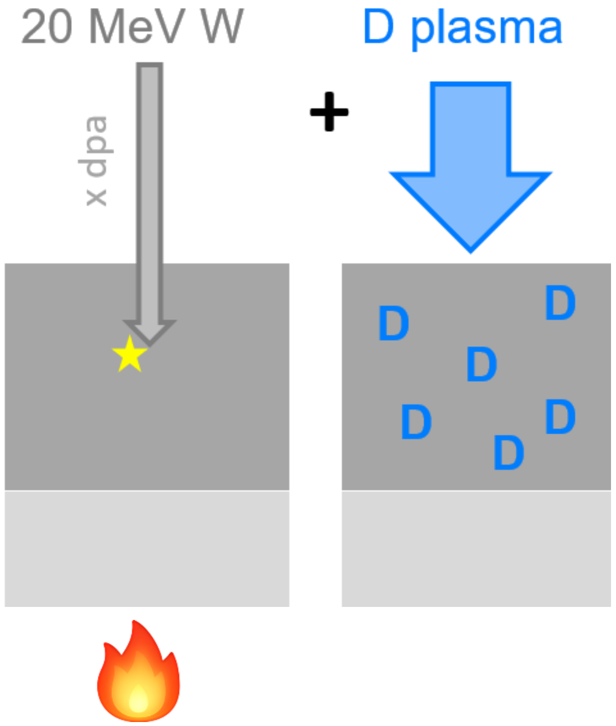

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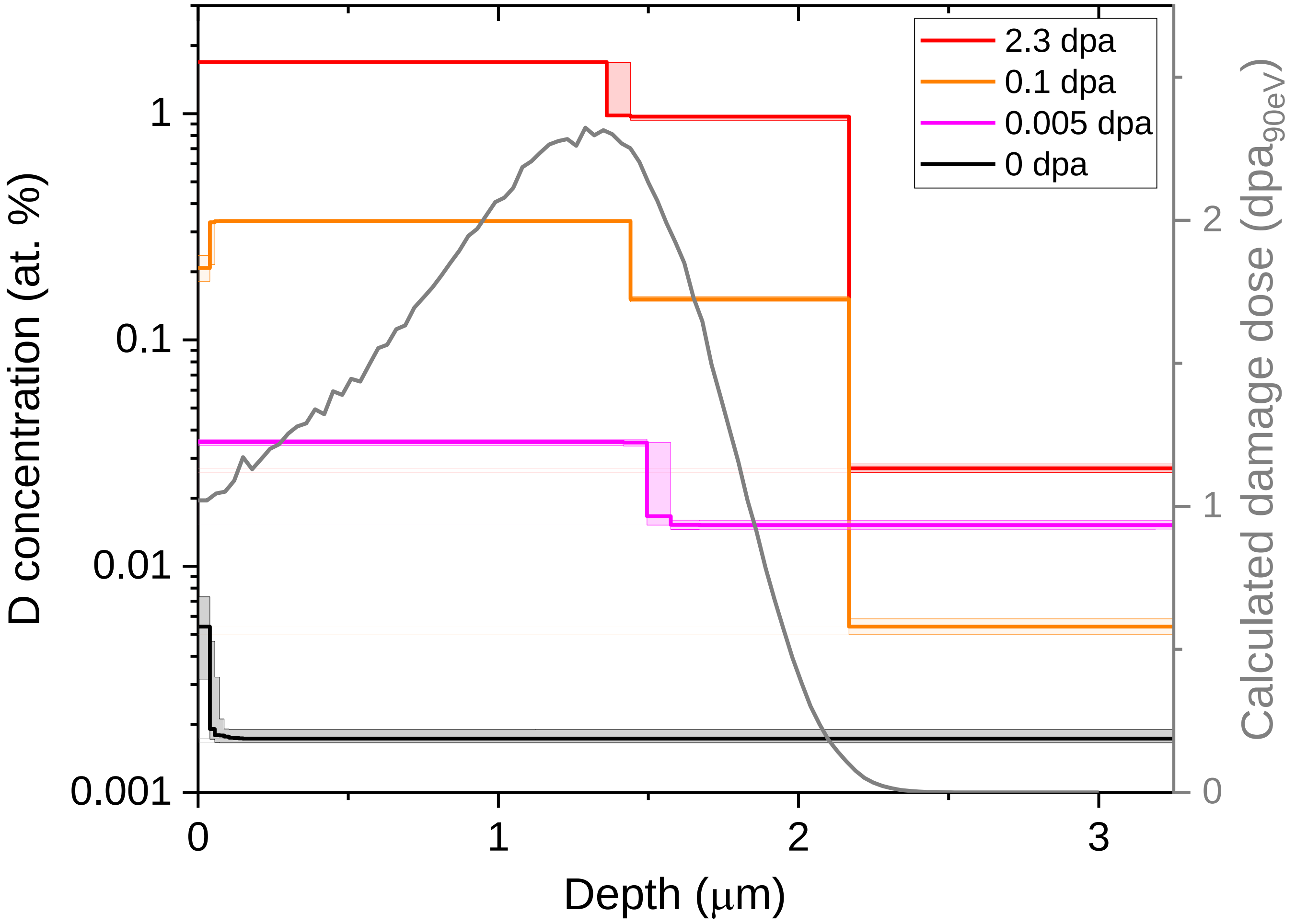
3. Quantitative analyses

- $D(^3He,p)\alpha$ nuclear reaction analysis
  $\Rightarrow$ Trapped D concentration
- Thermal desorption spectroscopy
  $\Rightarrow$ D trapping mechanisms

D retention in W irradiated at 1350 K

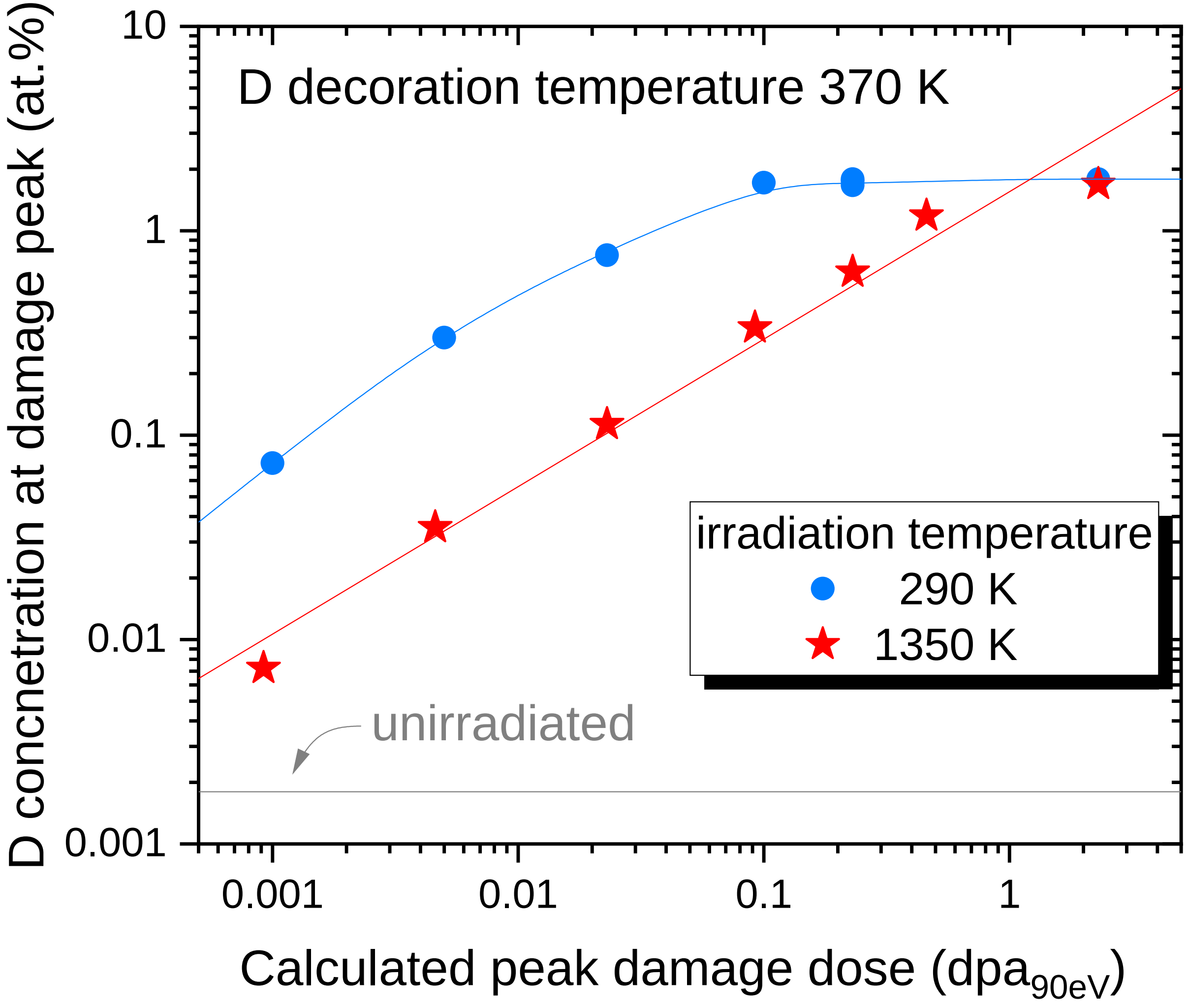
- Enhanced D concentration within the irradiated region



T. Schwarz-Selinger et al., unpublished

D retention in W irradiated at 1350 K

- No clear saturation of trapped D concentration up to 2.3 dpa

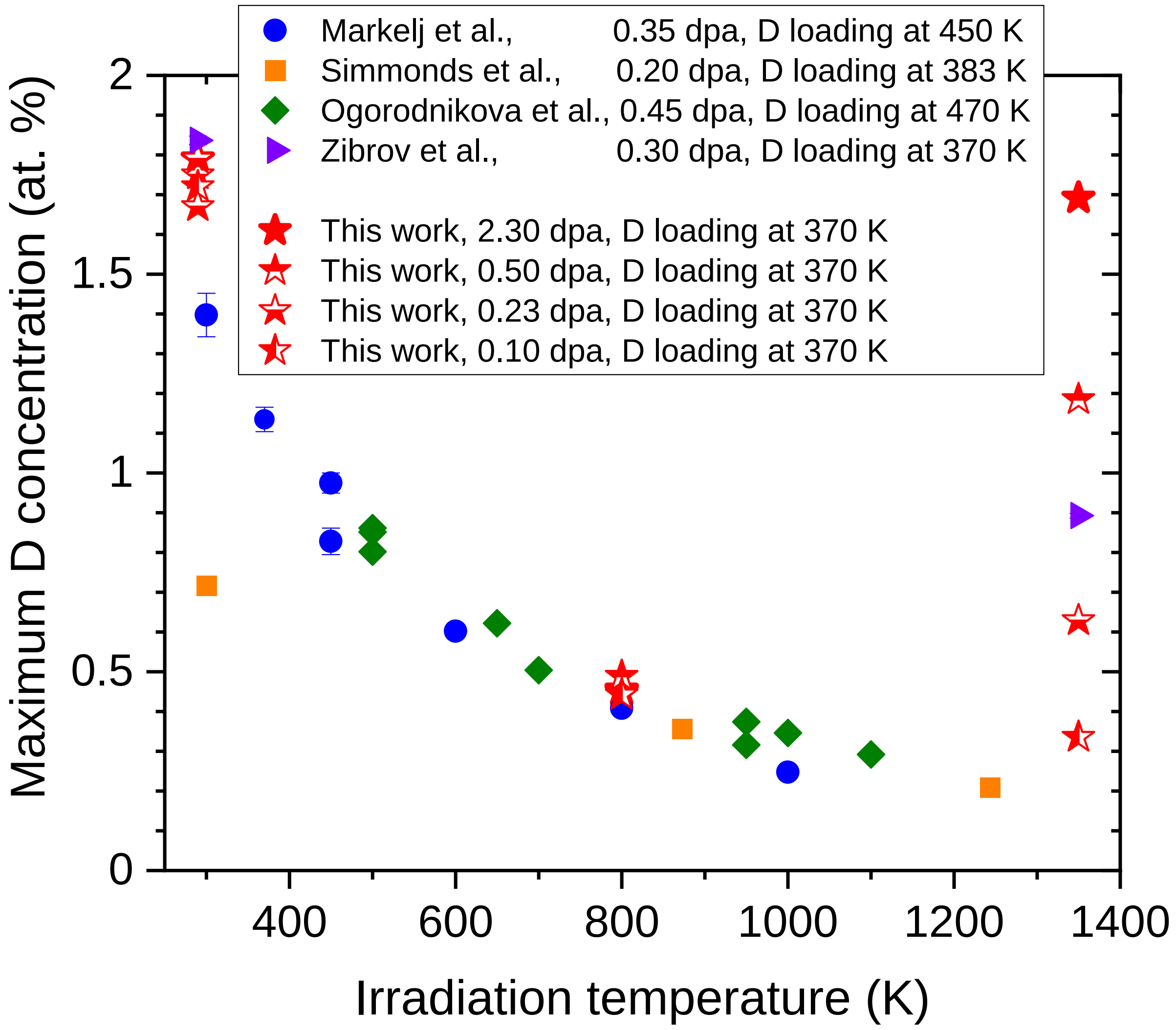


T. Schwarz-Selinger et al., unpublished

D retention in W irradiated at 1350 K

- Different behavior compared with irradiations at lower temperatures

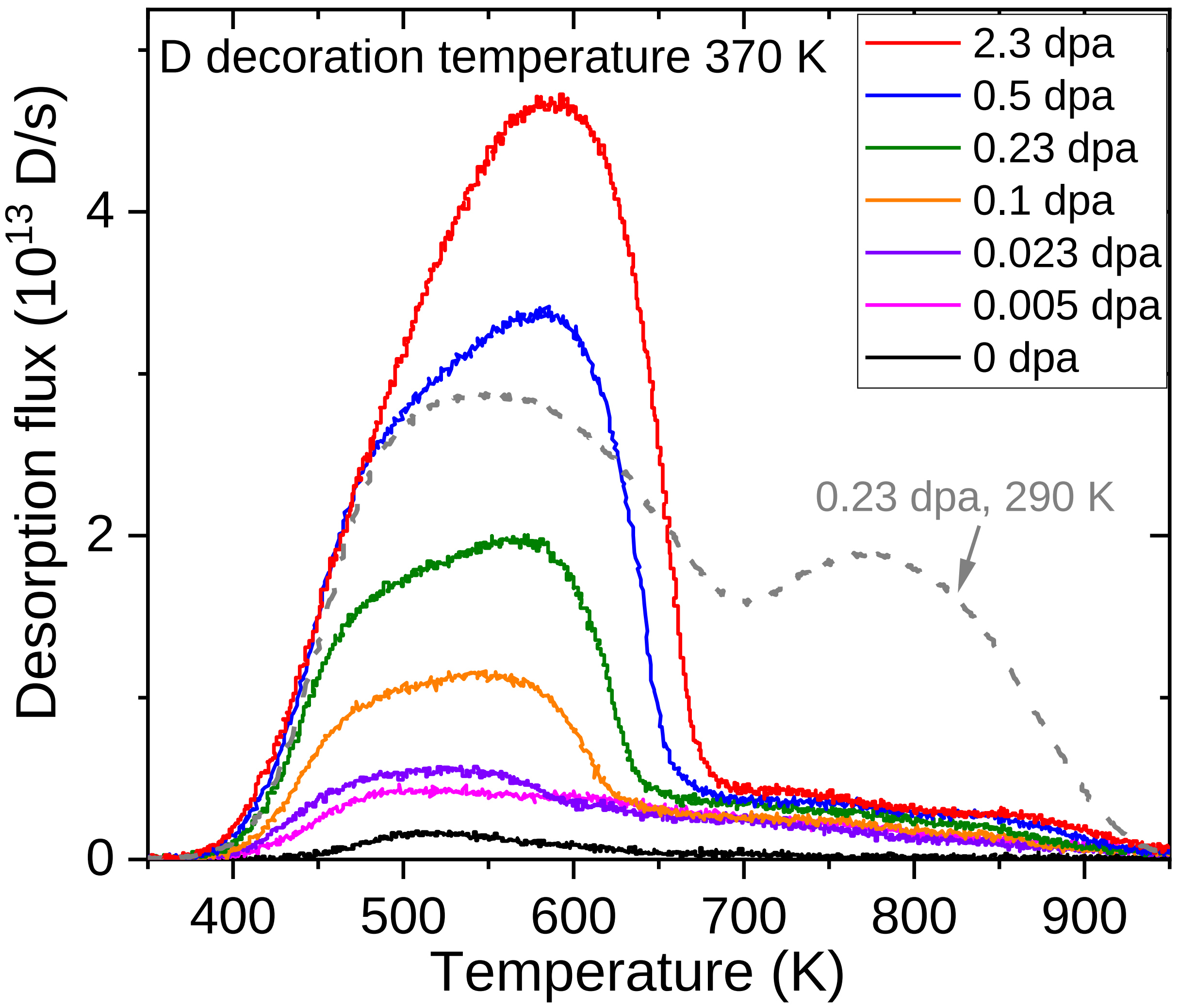
Literature: 0.2–0.45 dpa, D loading at 370–470 K



T. Schwarz-Selinger et al., unpublished

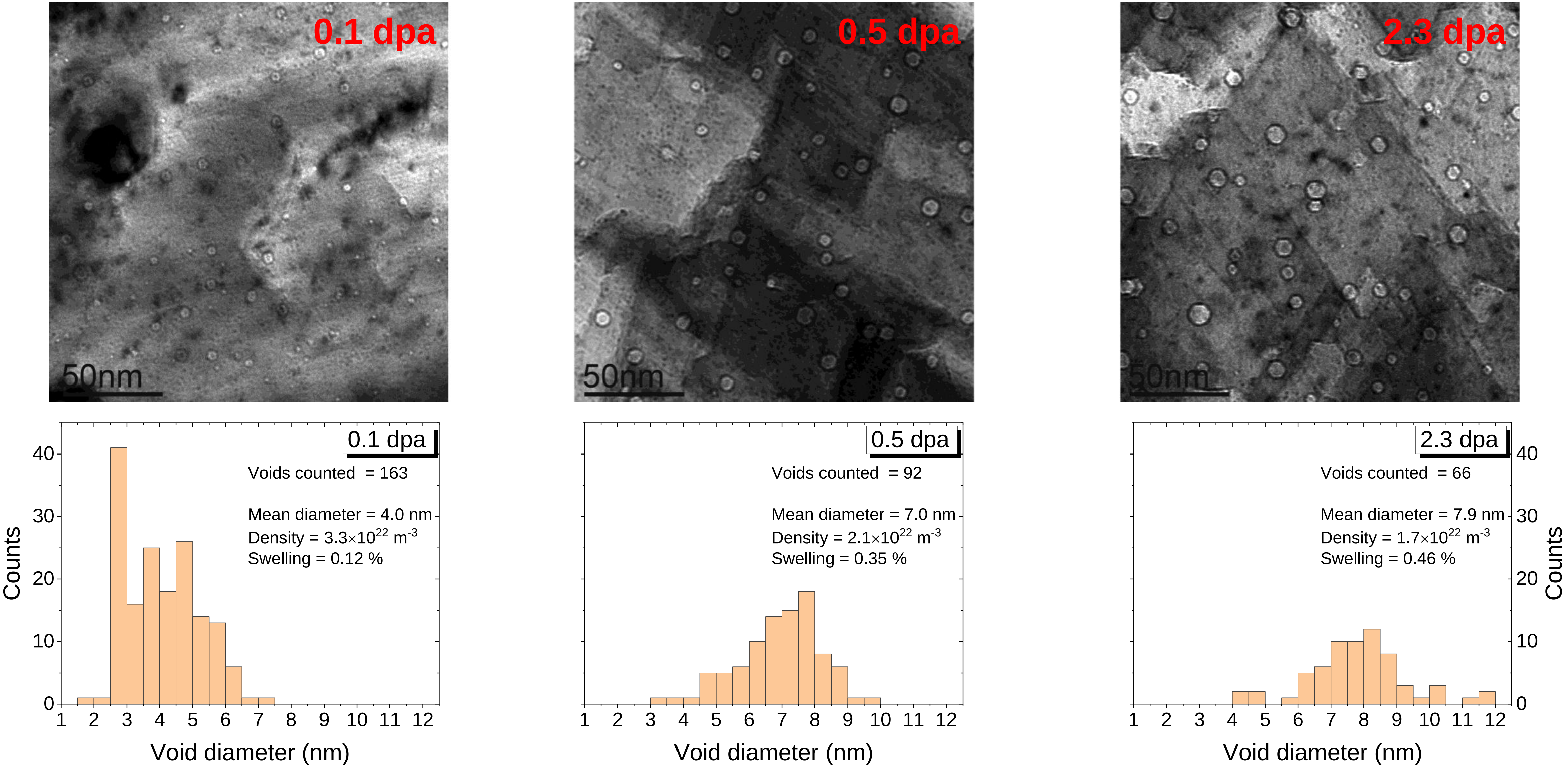
D retention in W irradiated at 1350 K

- Different shape of TDS spectra compared with irradiation at 290 K
  ⇒ Different D trapping mechanism



T. Schwarz-Selinger et al., unpublished
Z. Shen et al., unpublished

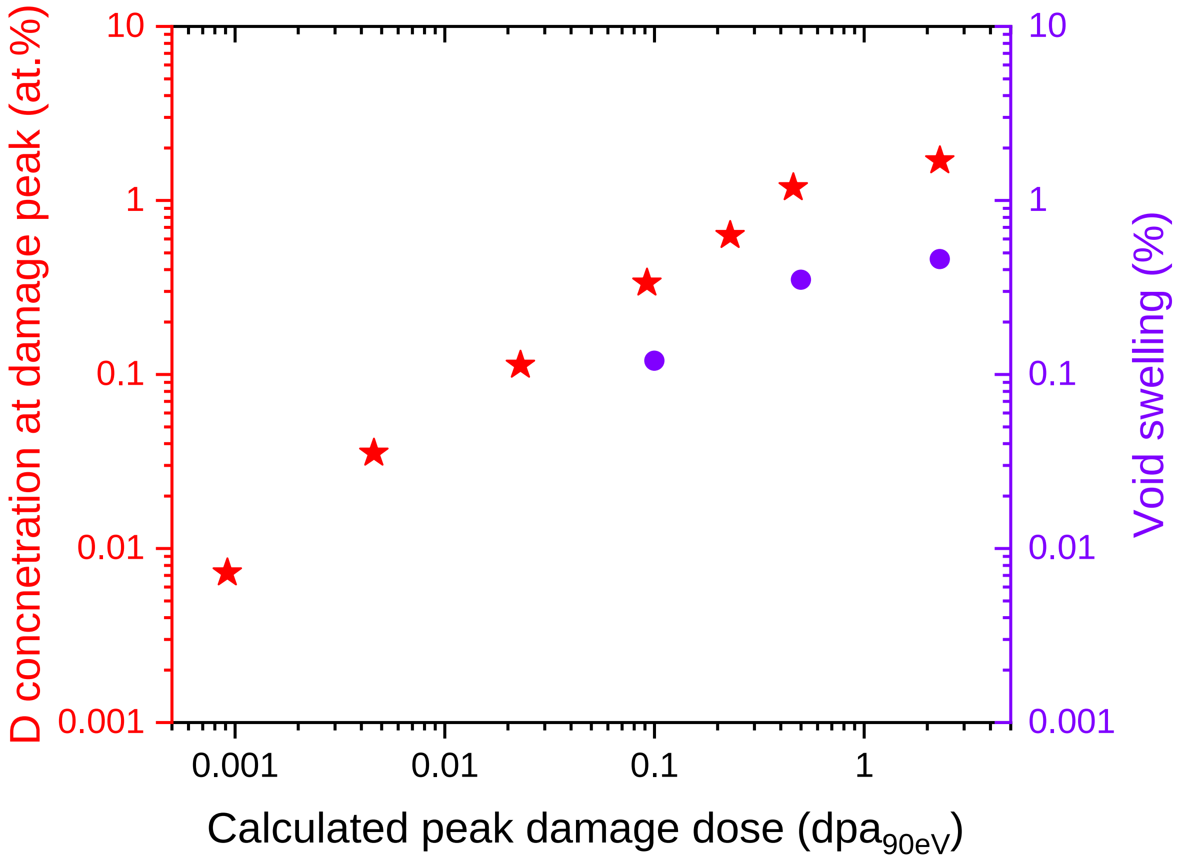
Microstructure of W irradiated at 1350 K



M. Klimenkov, KIT

Microstructure of W irradiated at 1350 K

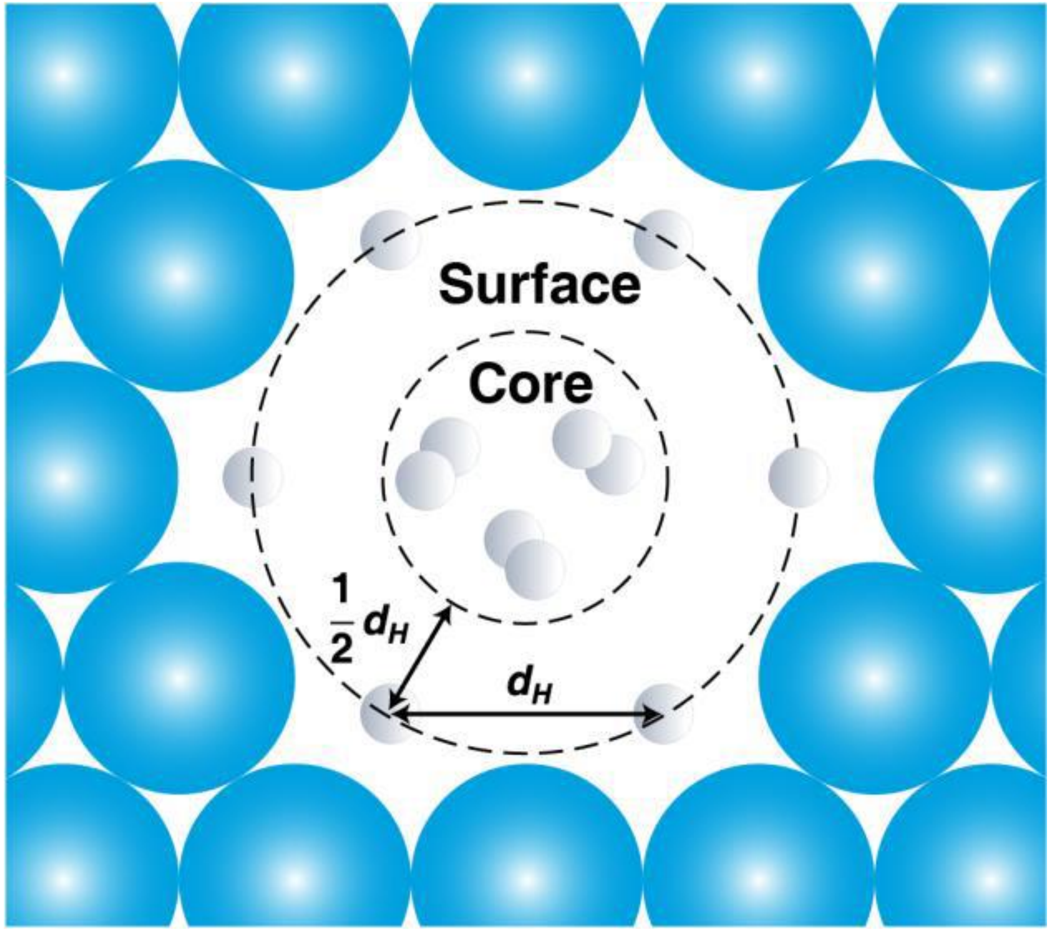
- Observed void sizes/densities are comparable with those in neutron-irradiated W, despite much higher dose rate, no transmutation elements, H, He
- No voids in samples irradiated at 290 K
- Void swelling (volume fraction of voids in the material) increases with dpa
- **Is D retained in voids?**



T. Schwarz-Selinger et al., unpublished

Results from DFT and ab-initio MD simulations

- D atoms adsorbed at a void surface
- D_2 molecules in a void volume
- **Different D retention mechanism compared with single vacancies (irradiation at low temperatures), where only D atoms are trapped!**



J. Hou et al., Nature Materials 2019 (18), 833

Reaction-diffusion model of D trapping and release from voids

- Diffusion equation for interstitial D including trapping:

$$\frac{\partial C}{\partial t} = \mathcal{D}(T)\frac{\partial^2 C}{\partial x^2} - \Lambda_{void}(x,t)$$

- D trapping/release from voids:

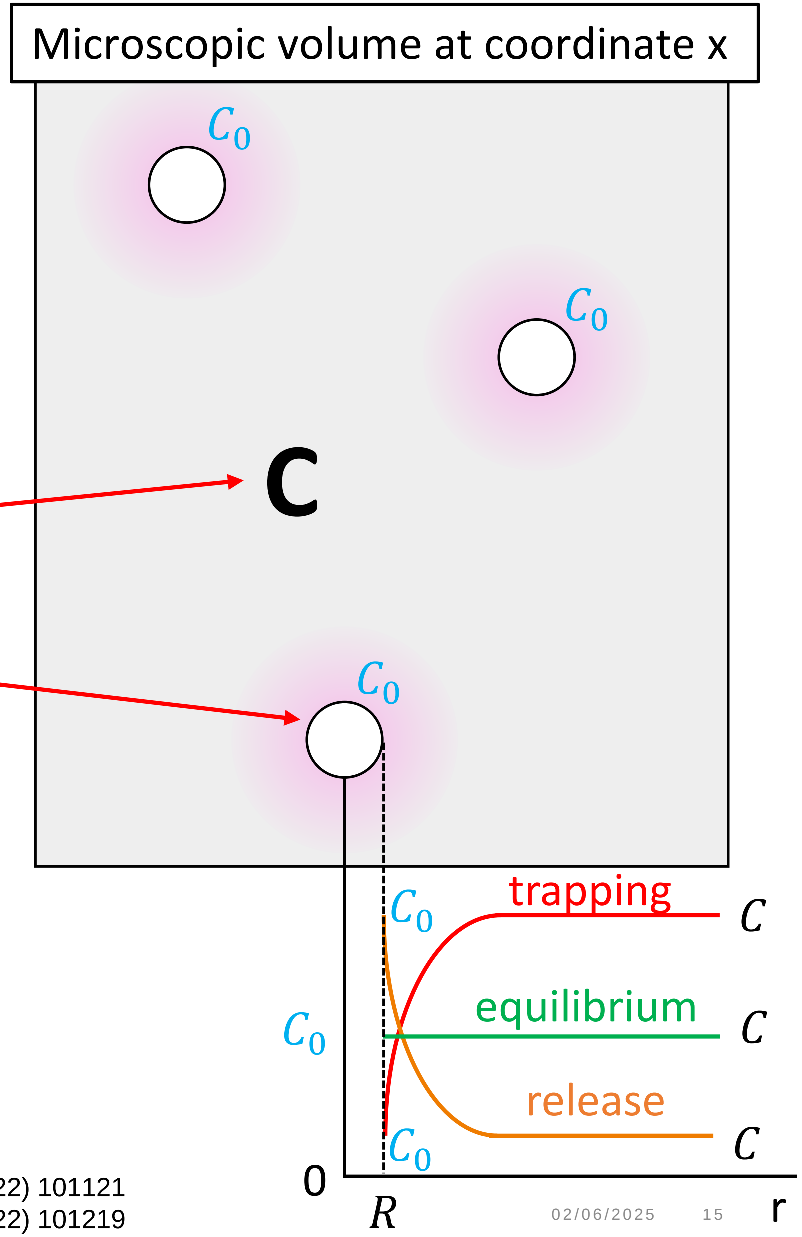
$$\Lambda_{void}(x,t) = 4\pi R\mathcal{D}(T)N_{\text{void}}(x)[C(x,t) - C_0(x,t)]$$

$C > C_0$: D trapping

$C < C_0$: D release

$C = C_0$: Equilibrium

- **Need to relate C_0 with D content in a void**

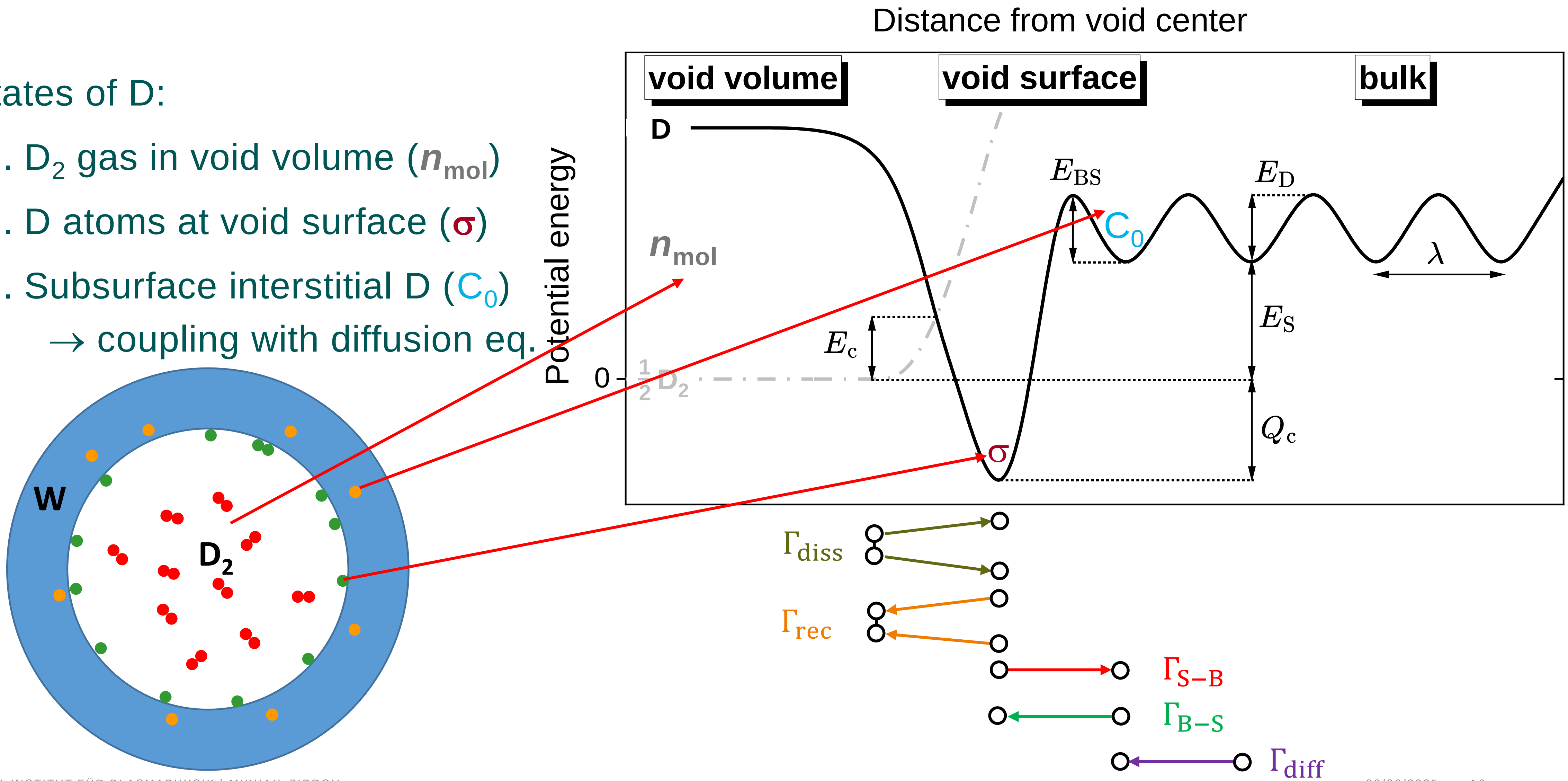


M. Zibrov, K. Schmid, Nucl. Mater.Energy 30 (2022) 101121
M. Zibrov, K. Schmid, Nucl. Mater.Energy 32 (2022) 101219

D potential energy landscape near a void surface

- States of D:
  1. D_2 gas in void volume ($\boldsymbol{n}_{\text{mol}}$)
  2. D atoms at void surface (σ)
  3. Subsurface interstitial D (C_0)

     $\rightarrow$ coupling with diffusion eq.



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  1. D_2 gas in void volume (***n***$_{\text{mol}}$)
  2. D atoms at void surface (**σ**)
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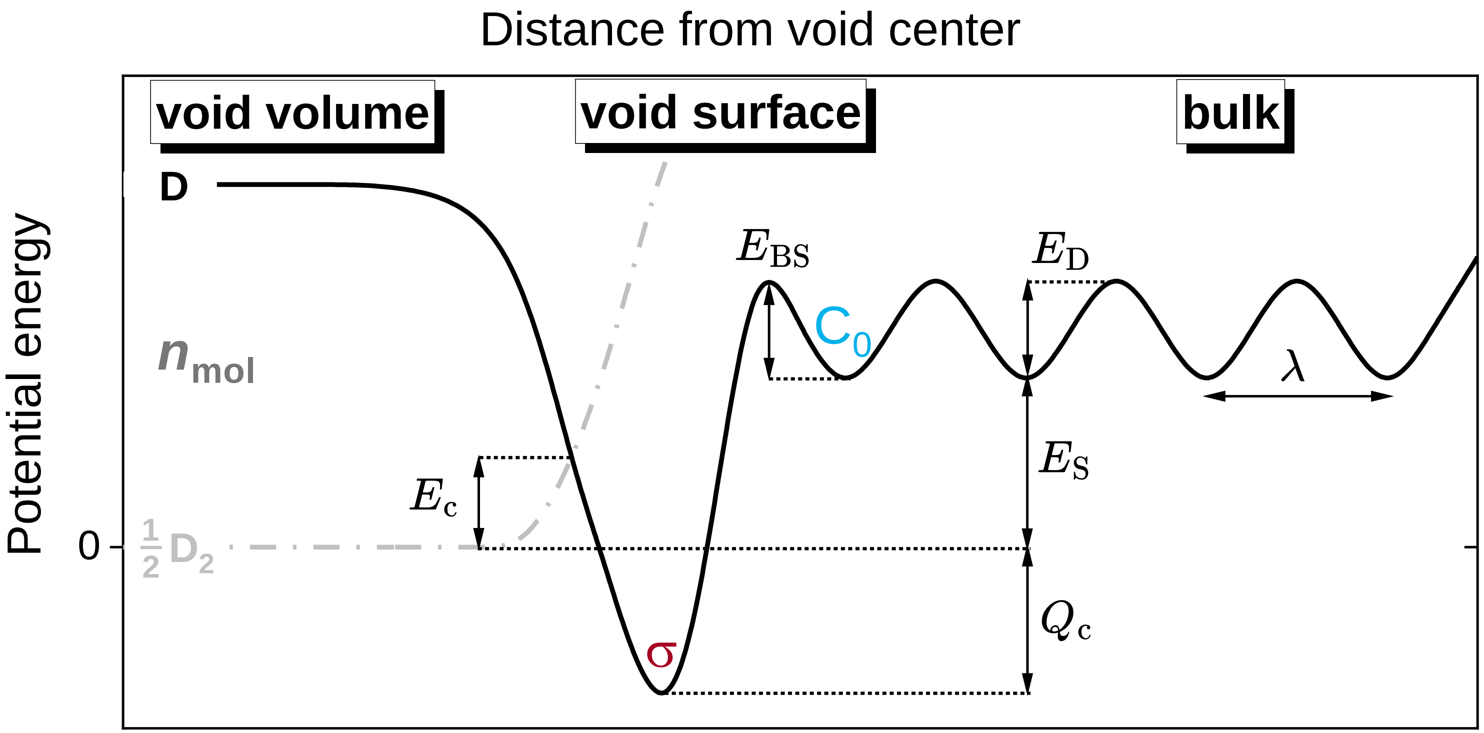
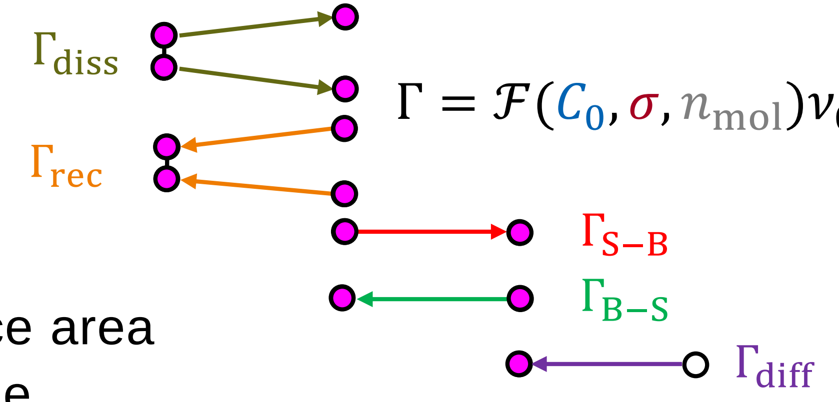
     $\rightarrow$ coupling with diffusion eq.
- Balance of particles:

$$\lambda \frac{\partial C_0}{\partial t} = \Gamma_{\text{S-B}} - \Gamma_{\text{B-S}} + \Gamma_{\text{diff}}$$

$$\frac{\partial \sigma}{\partial t} = \Gamma_{\text{diss}} - \Gamma_{\text{rec}} - \Gamma_{\text{S-B}} + \Gamma_{\text{B-S}}$$

$$2V \frac{\partial n_{\text{mol}}}{\partial t} = A(\Gamma_{\text{rec}} - \Gamma_{\text{diss}})$$





$$\Gamma = \mathcal{F}(C_0, \sigma, n_{\text{mol}})\nu_0 \exp\left(-\frac{E_{\text{act}}}{k_B T}\right)$$

A – void surface area
V – void volume

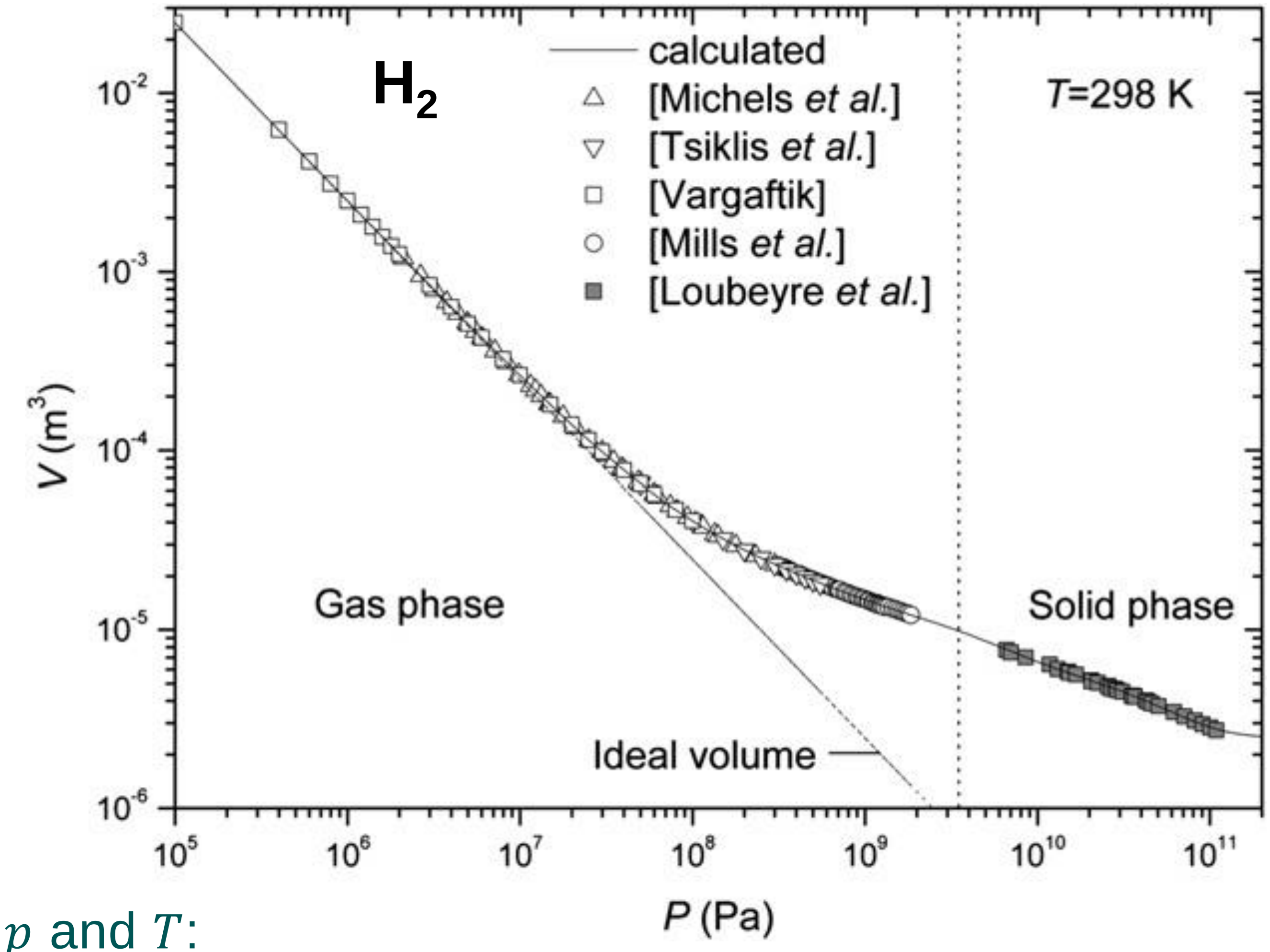
D_2 equation of state

- Deviations from ideal gas behavior at $p > 10^7$ Pa
- D_2 equation of state (EOS):

$$V_m(p,T) = \underbrace{\frac{RT}{p}}_{\text{Ideal gas}} + \underbrace{c + \sum_{i=1}^{5} a_i \exp\left(-\frac{p}{b_i}\right)}_{\text{Corrections for non-ideal behavior}}$$

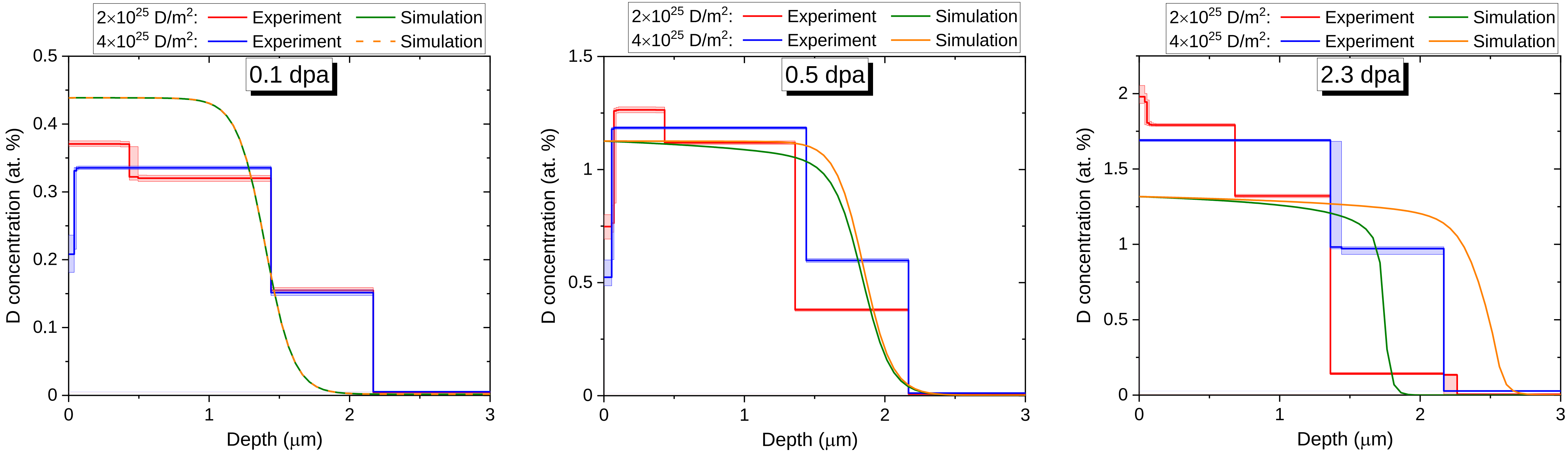
- Applicable for $p \leq 10^{11}$ Pa and $298\ \text{K} \leq T \leq 1000\ \text{K}$
- Density of D_2 molecules as function of p and T:

$$n_{\text{mol}} = \frac{N_A}{V_m(p,T)}$$



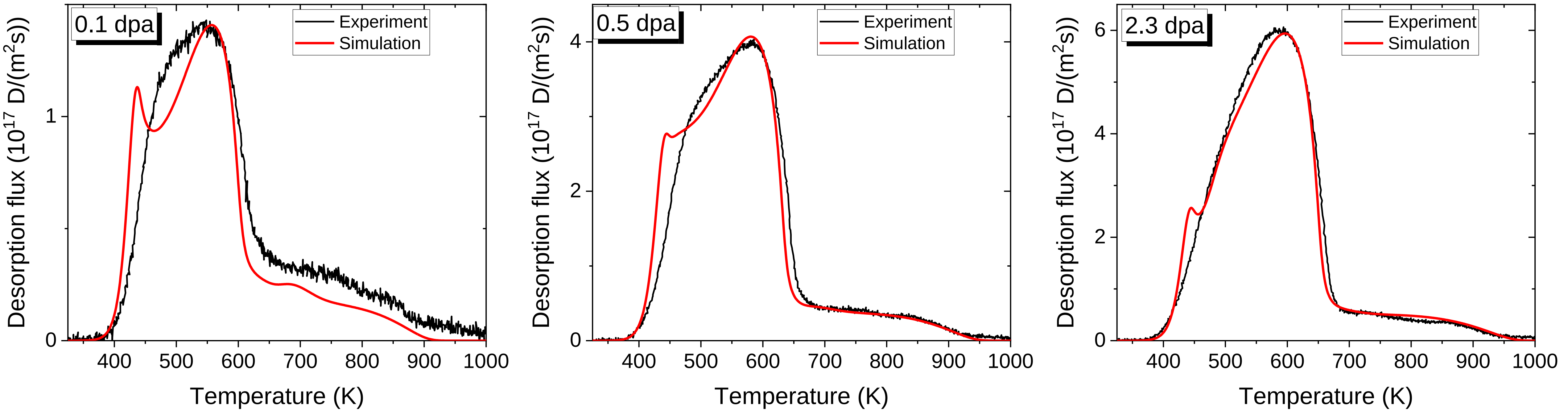
J.-M. Joubert, Int. J. Hydrogen Energ. 35 (2010) 2104.
J.-M. Joubert, S. Thiébaut, Acta Mater. 59 (2011) 1680.

Simulation results: D depth profiles



- Assume presence of only voids in the damaged zone + one intrinsic bulk trap
- Use void density and average size measured by TEM
- Simulate D plasma exposure and TDS
- Reasonable agreement with measured D depth profiles at two different D fluences

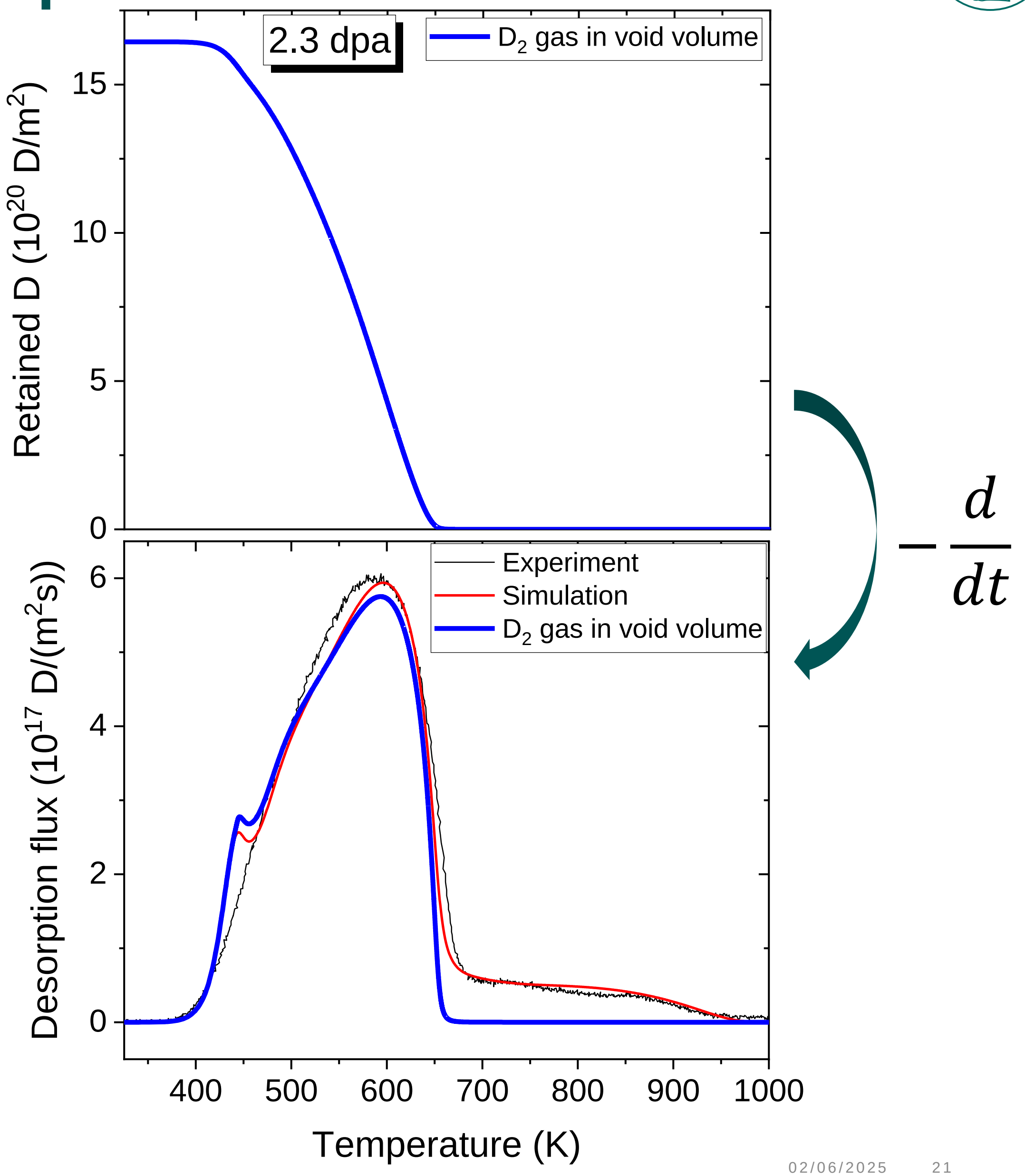
Simulation results: TDS spectra



- Reasonable agreement with experimental TDS spectra

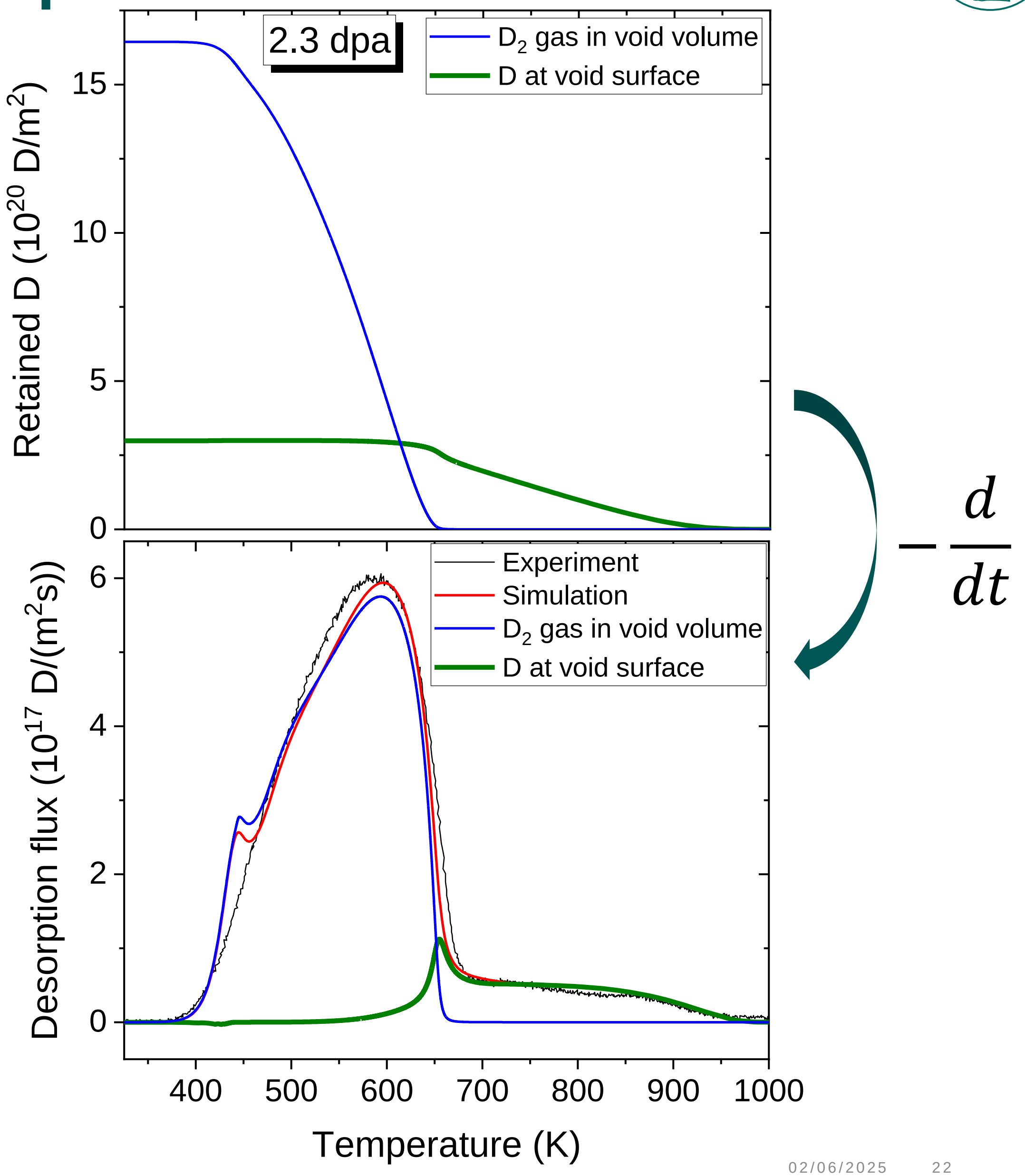
Simulation results: composition of TDS spectra

- Main TDS peak corresponds to depletion of D_2 gas in the void volume



Simulation results: composition of TDS spectra

- Main TDS peak corresponds to depletion of D_2 gas in the void volume
  $\Rightarrow$ concentration of D atoms at the void surface stays close to saturation value due to supply from D_2 gas in the volume
- High-temperature shoulder corresponds to depletion of chemisorbed D at the void surface (after no D_2 gas left in void volume)



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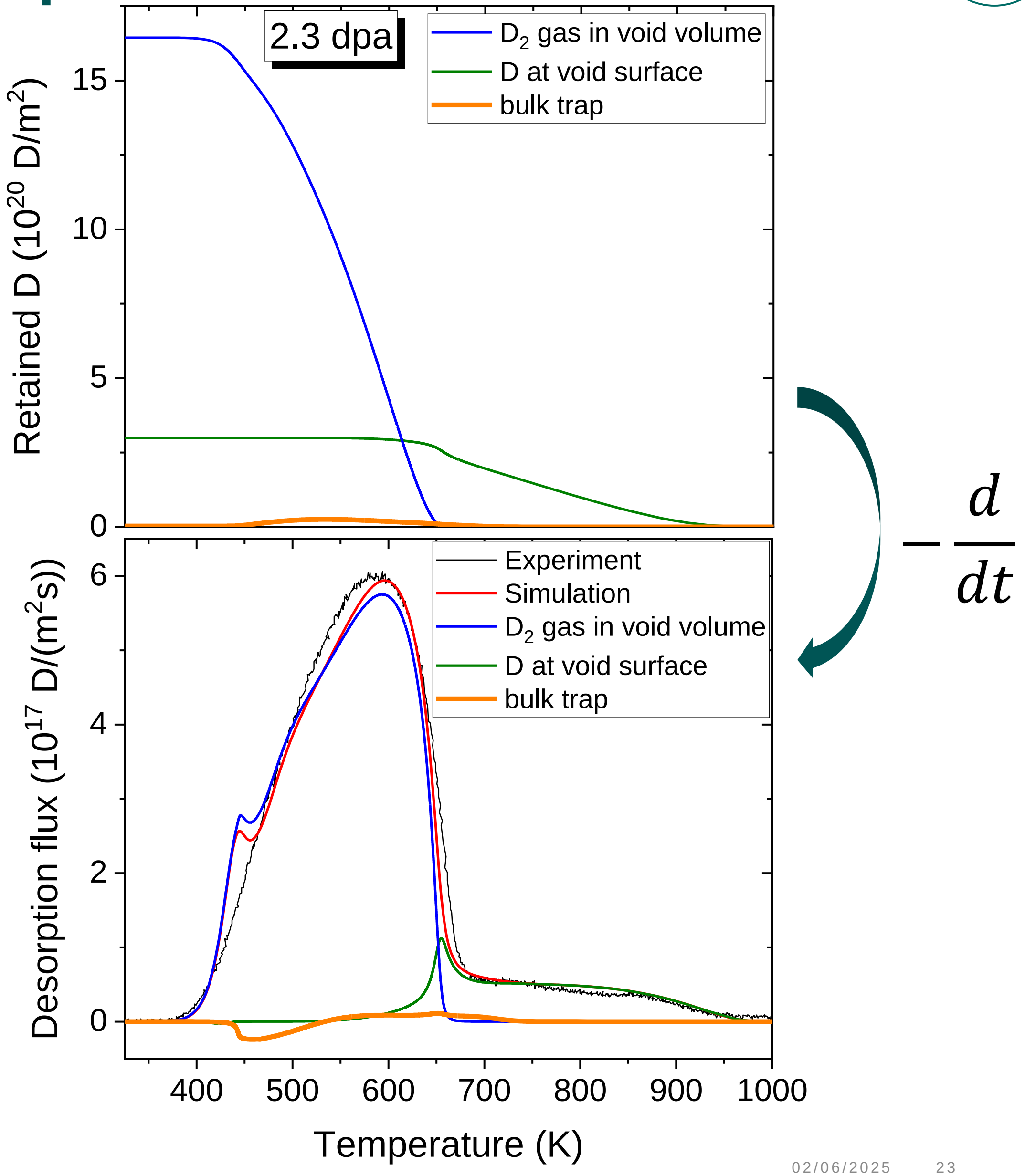
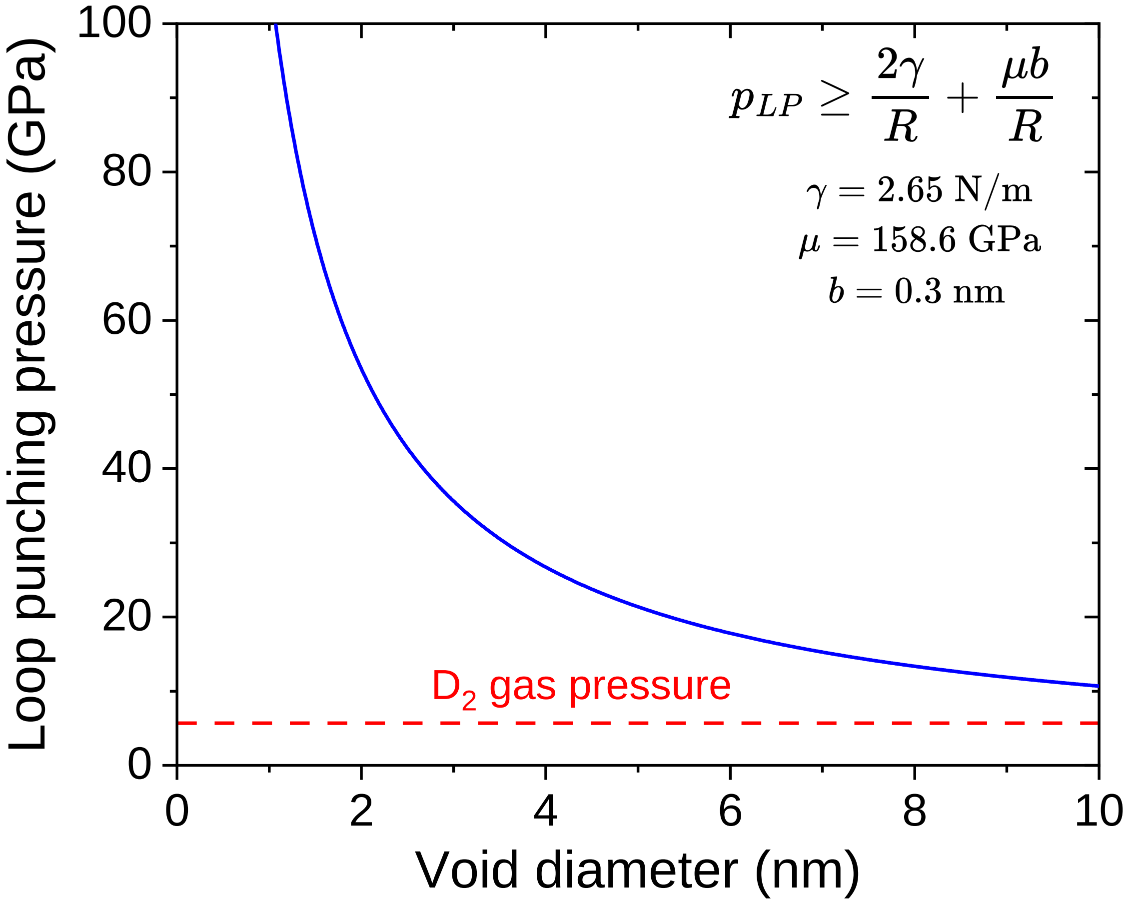


Simulation results: discussion

- Best fit is obtained using
  heat of D solution: E_S = 1.0 eV;
  experiment (W): E_S = 1.14 eV G. Holzner, PhD thesis.
- Equilibrium D_2 pressure in voids: 5.68 GPa
- Below the critical pressure to cause void volume increase by dislocation loop punching



G.W. Greenwood et al., Journal of Nuclear Materials 4 (1959) 305
R.D. Kolasinski et al., Journal of Nuclear Materials 415 (2011) S676

Summary

- Formation of nm-sized voids after self-ion irradiation of W at 1350 K
- No clear saturation of void swelling up to 2.3 dpa
- No clear saturation of trapped D concentration
- TDS indicates different D trapping mechanism compared with irradiation at 290 K
- Can be explained by assuming that D is trapped as D_2 gas in void volume and as D atoms at void surface