EUROfusion

Influence of the presence of deuterium on displacement damage in tungsten

T. Schwarz-Selinger[1], J. Bauer[1], S. Elgeti[1]
M. Pečovnik[2], S. Markelj[2]

1 Max-Planck-Institut für Plasmaphysik

2 Jožef Stefan Institute

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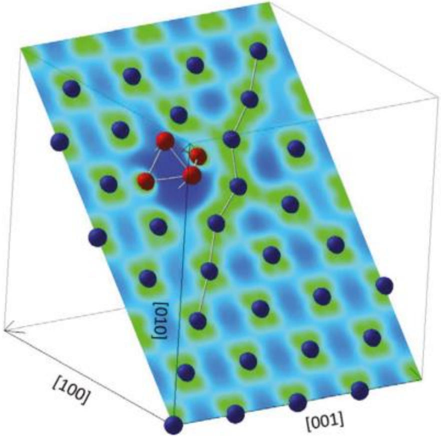
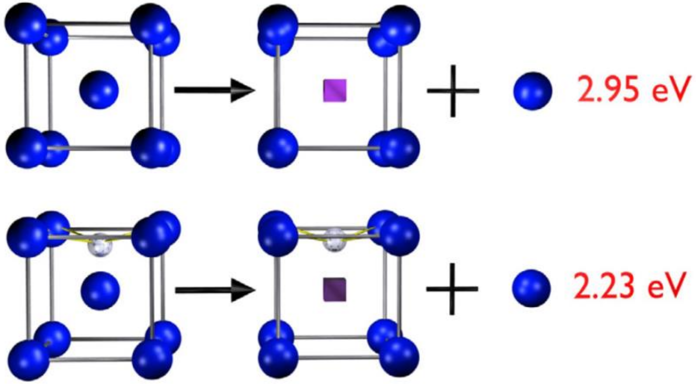
Theoretical predictions

- ‘DFT molecular dynamics revealed that **hydrogen clusters can prevent a vacancy from recombining** with the neighboring crowdion-type self-interstitial-atom.’
  D. Kato et al., Nucl. Fusion 55 (2015) 083019

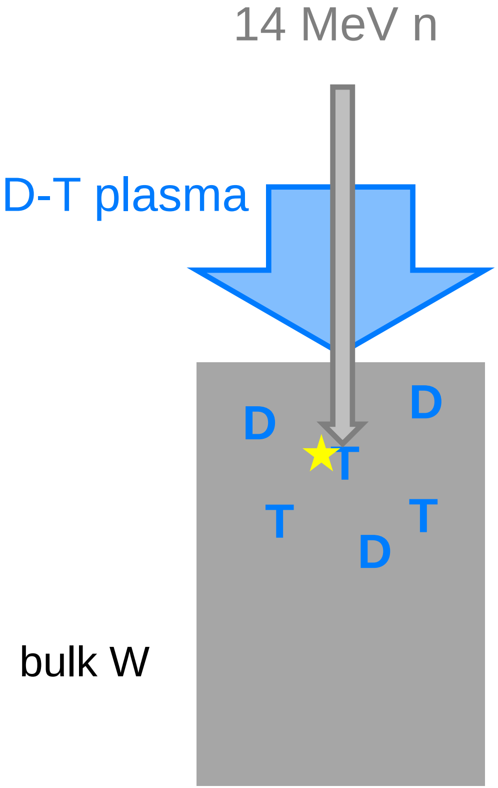
- ‘Atomic scale computer simulations have predicted a **decrease in the W vacancy formation energy in the presence of H …** Findings of this work suggest that H not only promotes vacancy formation in W, but once formed the **vacancy will also initiate further H clustering**’
  S.C. Middleburgh, J. Nucl. Mater. 448 (2014) 270



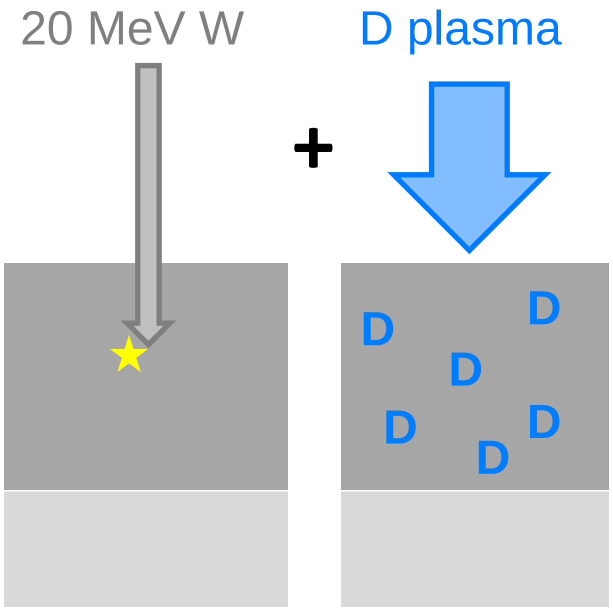


Motivation

In a future fusion reactor:



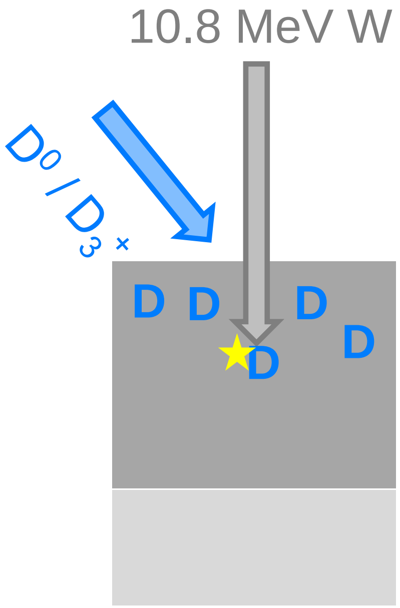
In present day lab experiments:



⇒ **mutual influence of D on damage creation/evolution?**

Experimental strategy

Shown before by Sabina:

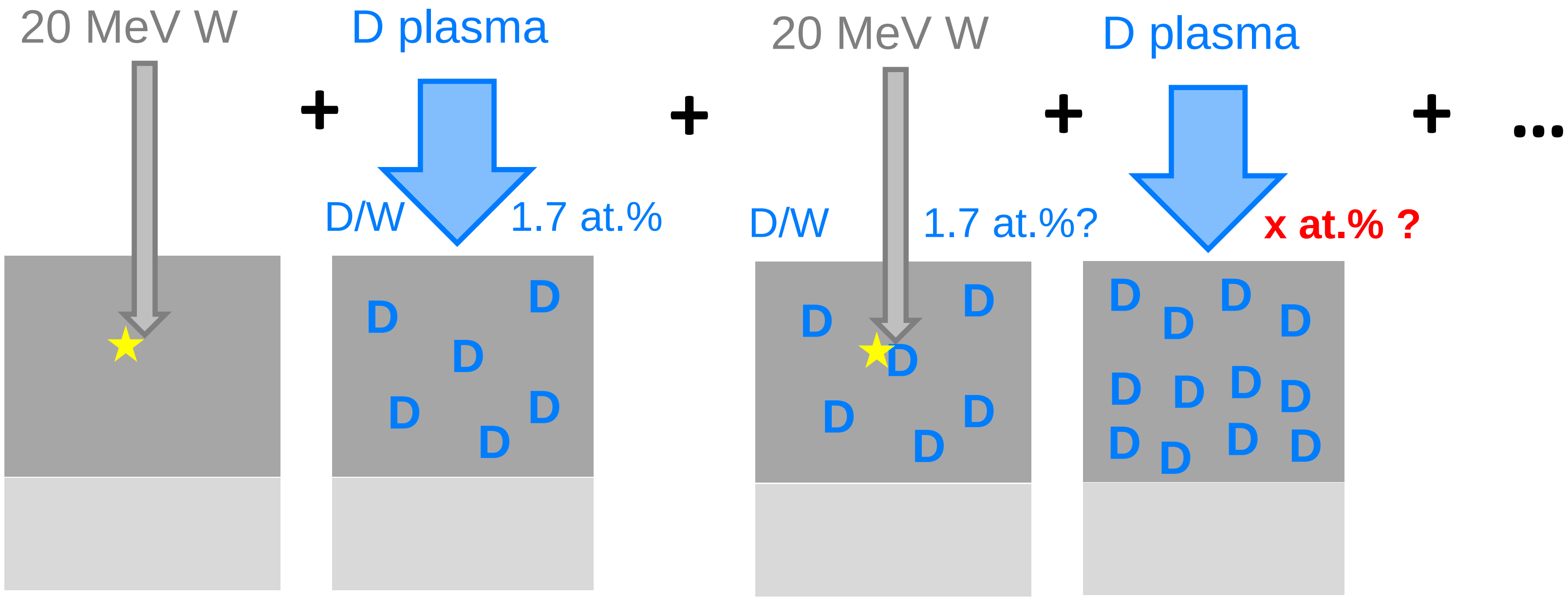


sequentially or simultaneously

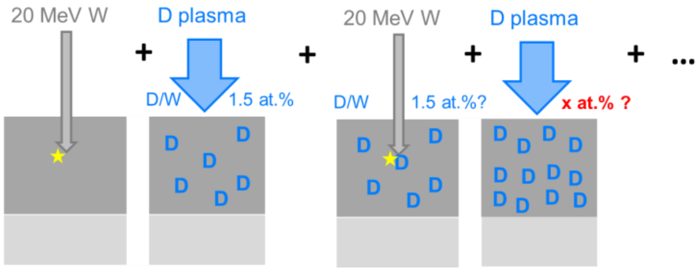
+ additional D decoration

Experimental strategy

Approach here: sequential treatment multiple times



Experimental strategy



- Compare D retention in
  - tungsten free of D
  - tungsten ‘saturated with D’

after 20 MeV W bombardment and D decoration of defects

⇒ Questions to address beforehand:

  - ➢ D uptake as function of W damaging fluence (Does damage saturate?)
  - ➢ D uptake as function of D fluence (How to decorate defects without creating new ones?)

Outline

- Motivation

- D retention in self-damaged tungsten

- Multiple sequence experiments: Damage creation → D loading

  D depth profiles and thermal desorption data

- Present rate equation modelling approaches

A comment before I start

- High energy and/or high flux D (plasma) exposure leads to

  - H oversaturation
    [L.Gao et al., Nucl. Fusion 2017 https://doi.org/10.1088/0029-5515/57/1/016026]

  - damage creation (point defects … blisters)

  which we want to avoid in this study (not trivial, see e.g.
  S. Kapser et al., Nucl. Fusion, 2018 http://dx.doi.org/10.1088/1741-4326/aab571)

- The strategy here is to investigate the effect of displacement damage, hence D loading needs to be done <u>without creating new damage</u>

D decoration: gentle plasma exposure

- known flux and energy

  - energy: „<5 eV/D“ (floating targets)

  - ion flux: 6 ×10^{19} D/(m^2s)

    (97% as D_3^+, 2% as D_2^+, 1% as D^+)

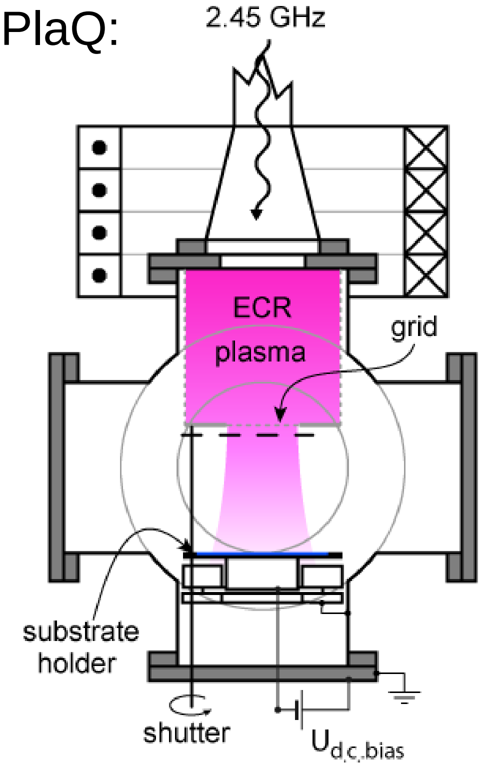
  - atom flux > 10^{21} D^0/(m^2s)

  - ion fluence: up to 5·10^{24} D/m^2 per day

- ’gentle’ loading = ‘decoration’:
  T = 370 K
  - no additional defect creation
  - no defect evolution/annealing)

- six samples simultaneously

PlaQ:

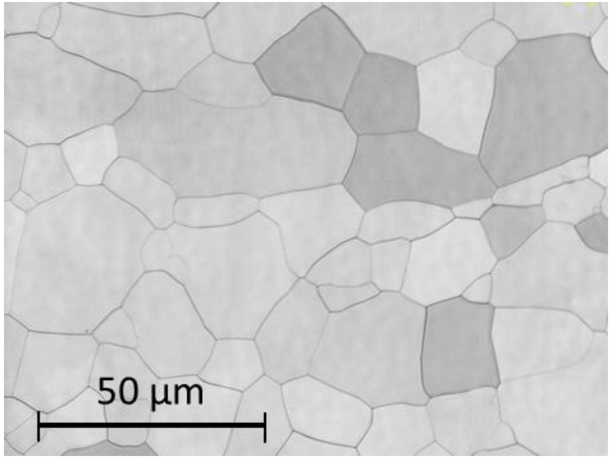


A. Manhard, Plasma Sources Sci. Technol. ***20*** *(2011) 015010*

The tungsten substrate material

- Plansee AG hot-rolled tungsten, purity 99.97 wt.-%
- chemo-mechanically polished to mirror finish [1]
- annealed at 2000 K for 2 min at $p < 5 \times 10^{-8}$ mbar to reduce initial defect density
- to 2×10^{12} m/m^3 [2]

confocal scanning laser microscopy



[1] A. Manhard et al., Pract. Metallogr. 50 (1) (2013) 6–15.

[2] A. Manhard et al., Pract. Metallorg. 52 (2015) 437.

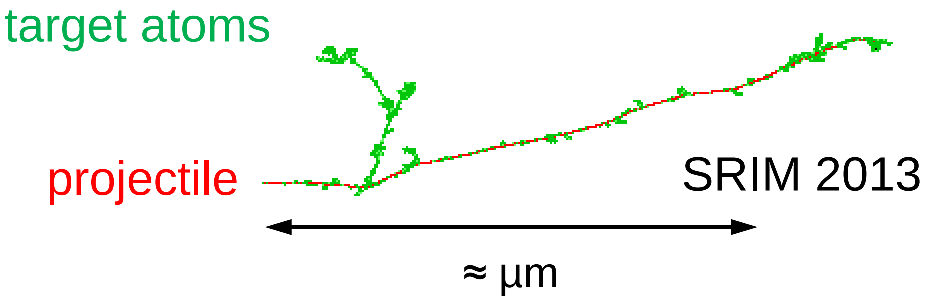
Creating displacement damage: W self-implantation

- 14 MeV fusion neutrons will cause
  - transmutation
  - gas production
  - displacement damage (E_{pka} < 200 keV)
- Here: only displacement damage aspect is studied with W self-implantation

Why tungsten ions?
+ no chemical effects
+ dense cascades
+ fast: 1 dpa in 1 hour

- vacancies, vacancy clusters, voids, ….
- too high E_{pka}

20 MeV W in W



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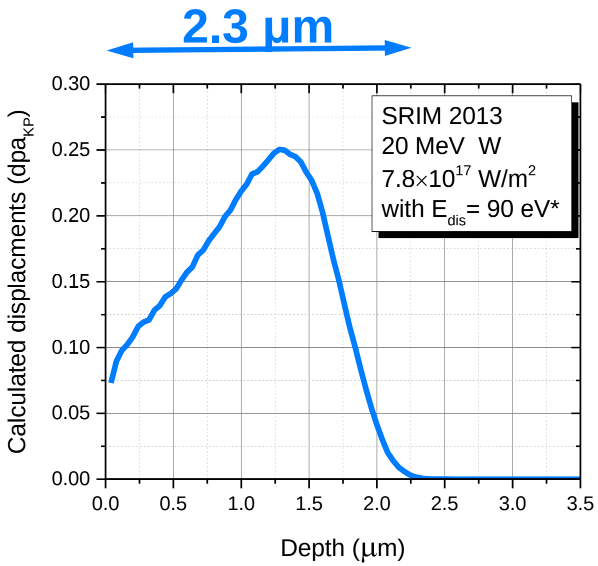
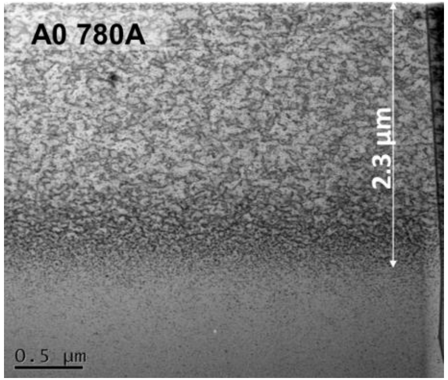


J. Grzonka et al., NIMB Vol 340, p. 27 (2014)

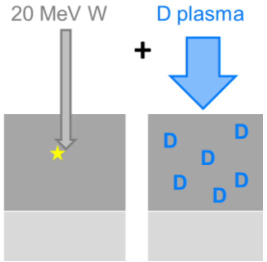


STEM micrograph

- 300 keV W would reduce information depth to 30 nm : Too little material for diagnostics (nuclear reaction analysis, thermal desorption spectroscopy)
- Cascade splitting makes it still relevant (?)

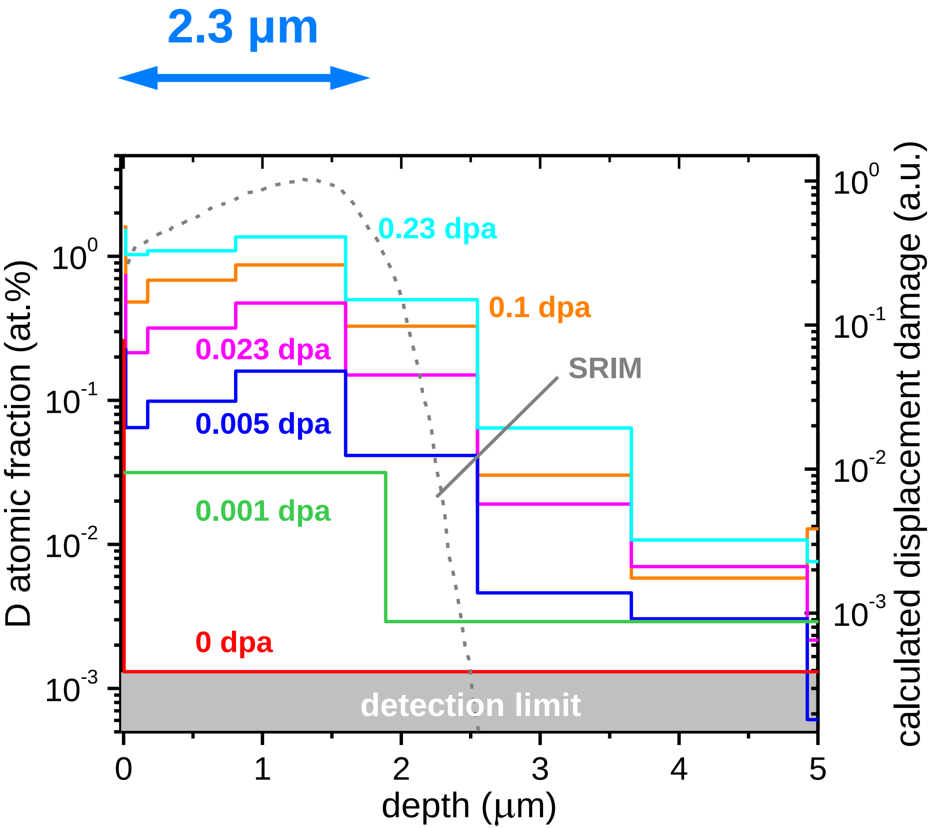
[A. Sand et al. *Mater. Res. Lett.* 5 (5), 357–63 (2017)]

D retention in self-damaged W

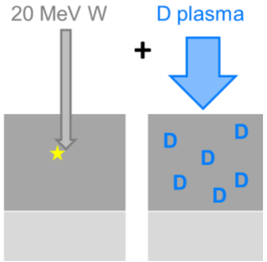


Previous investigation:

- fluence series 20 MeV W^{6+} @ 290 K
- D decoration with < 5 eV/D
  for 72 h (1.5×10^{25} D/m^2) @ 450 K
- D/W > 1 at.% @ 0.23 dpa



D retention in self-damaged W



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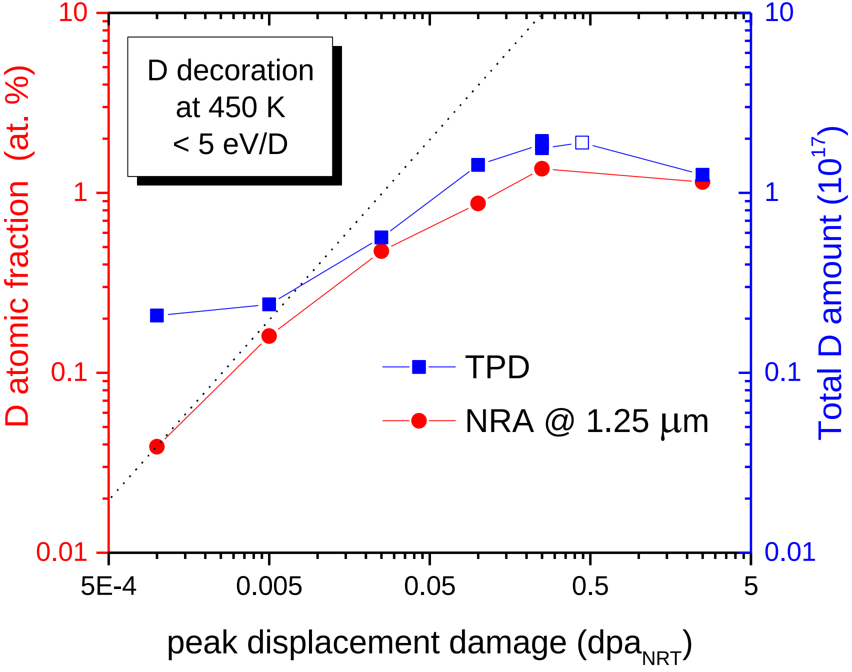
- saturation in D for > 0.23 dpa



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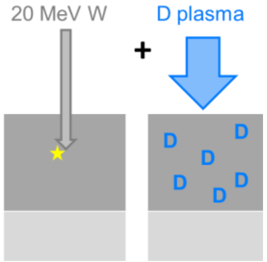

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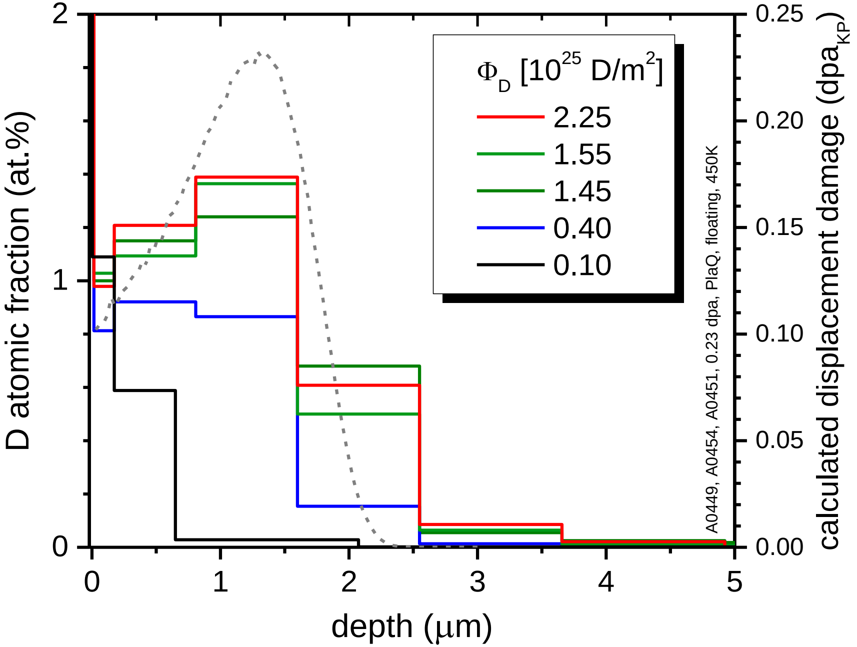
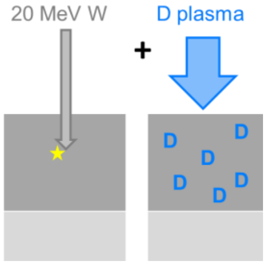


D retention in self-damaged W



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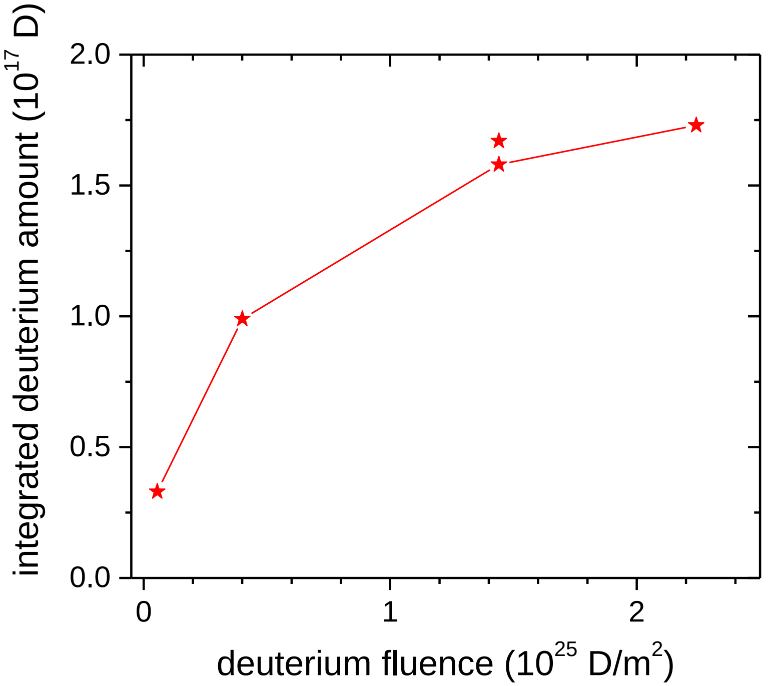
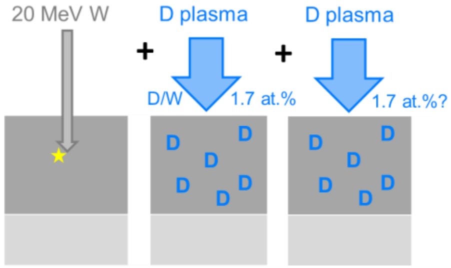


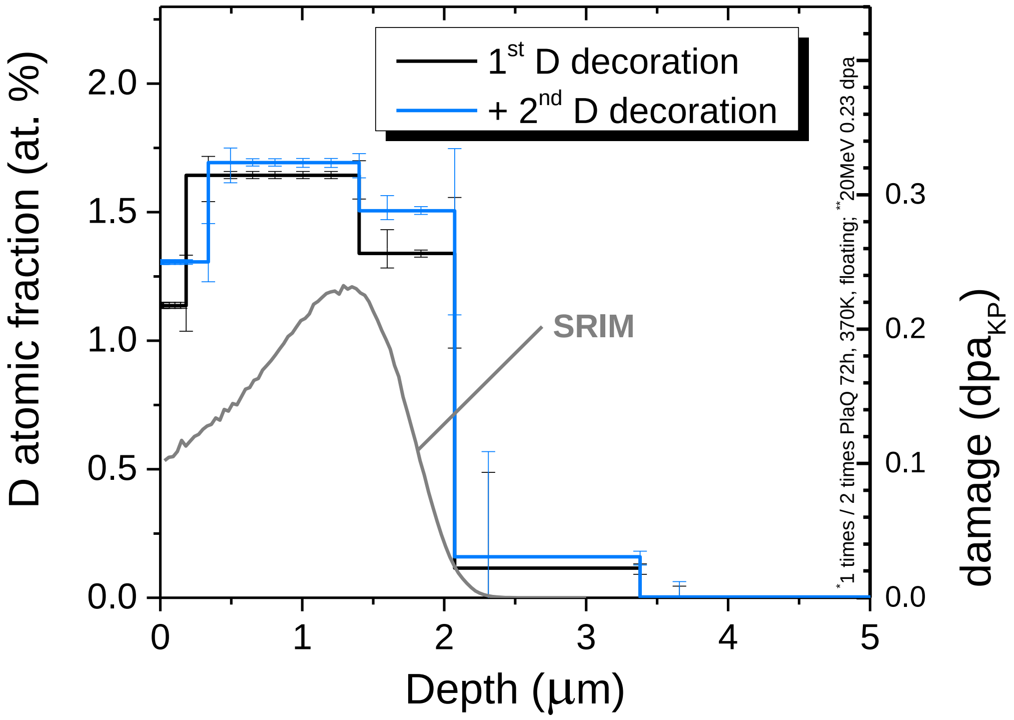
Saturating displacement damage with D



This study:

- D decoration @ 370 K
- 2 times $1.5 \cdot 10^{25}$ D/m² (2 x 72 h)
- check if damaged zone is completely filled with D

⇒ It is, up to **1.7 at.%**



Doubling the D fluence does not increase D amount

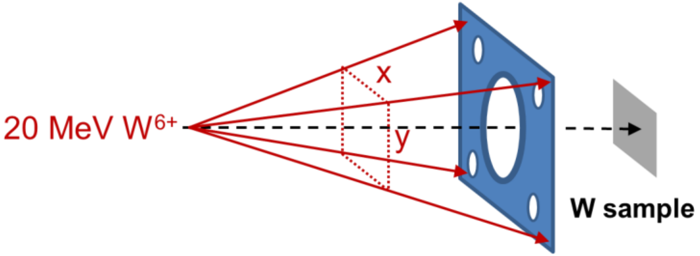
D retention in self-damaged W

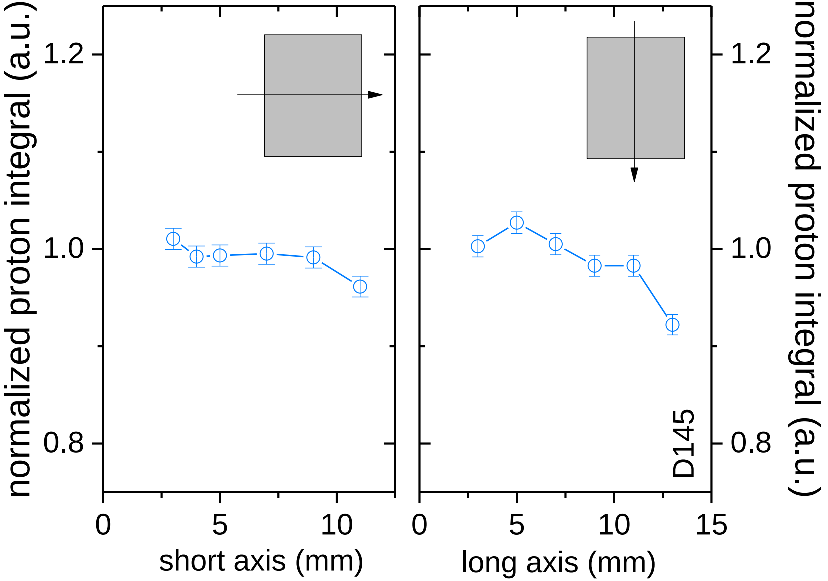
- beam sweep for laterally homogenous damage

- accuracy, reproducibility:

  better than 5%

⇒ box like D reservoir





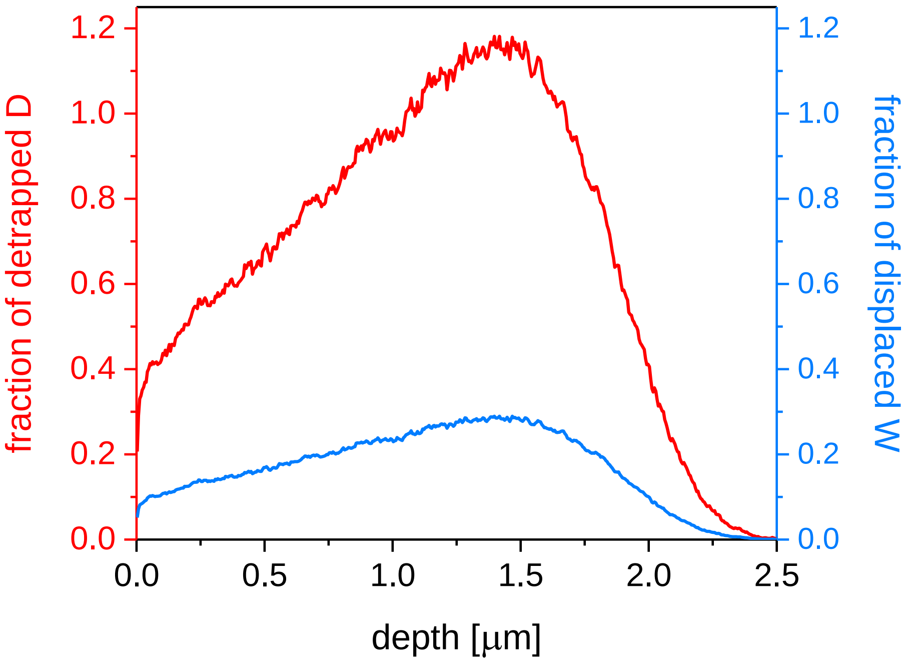
Displacements during 20 MeV W

SDTrimSP calculation:

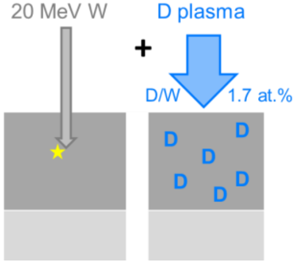
- 20 MeV W on W, containing 2 % D
- $\Phi = 7.87 \times 10^{17}$ W^{6+}/m^2
- displacement energy
  - $E_{displ.\ W} = 90$ eV, $E_{cutoff,\ W} = 2.2$ eV
  - $E_{displ,\ D} = 1$ eV, $E_{cutoff,\ D} = 0.25$ eV

⇒ tungsten atoms are displaced and defects are generated (0.23 dpa)

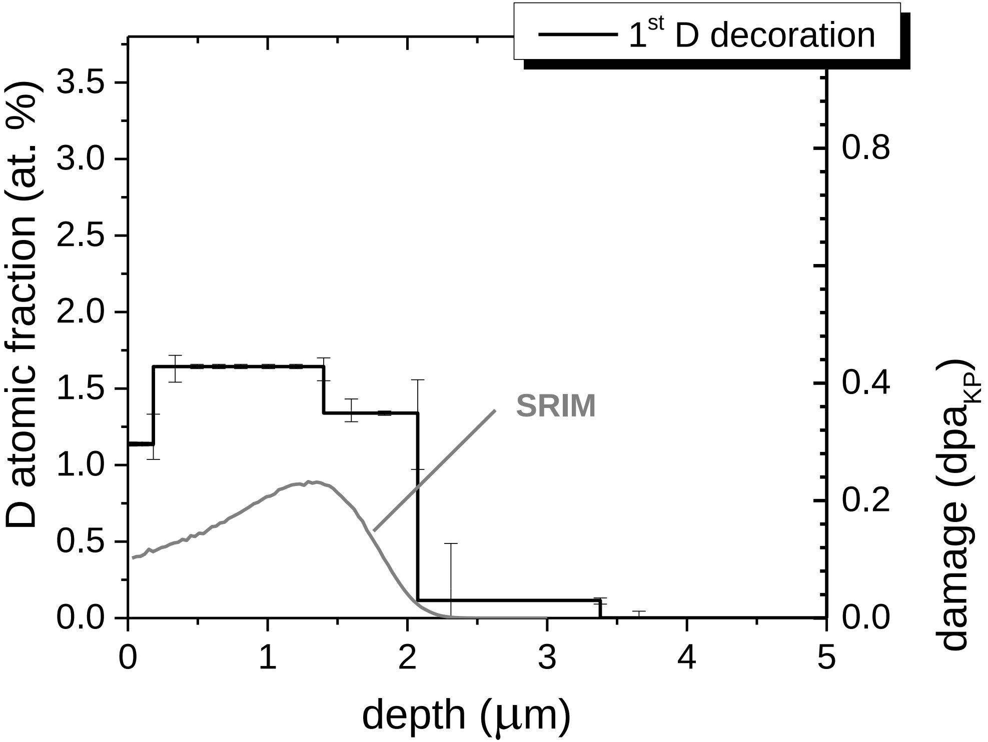
⇒ simultaneously, retained deuterium atoms (1.7%!) are de-trapped in the vicinity of the displacement damage: kinetic detrapping



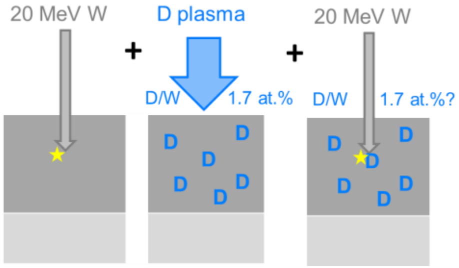
D depth profiles



What happens to the initially retained D?



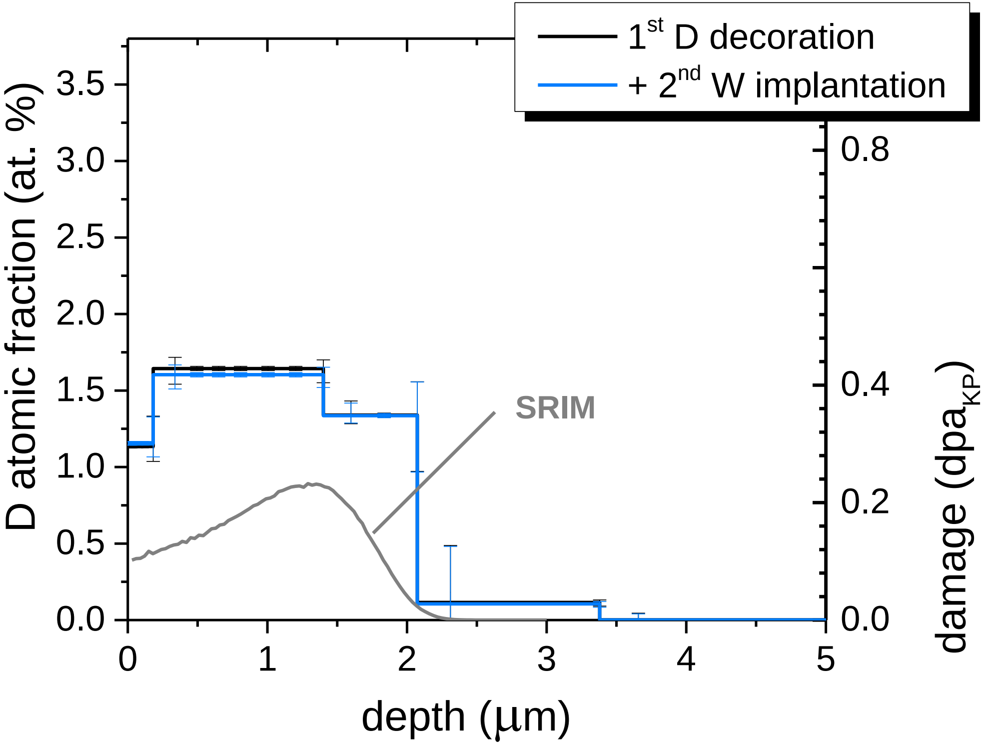
D depth profiles



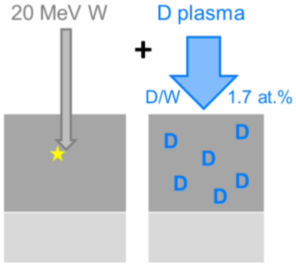
What happens to the initially retained D?

⇒ no change in depth profile

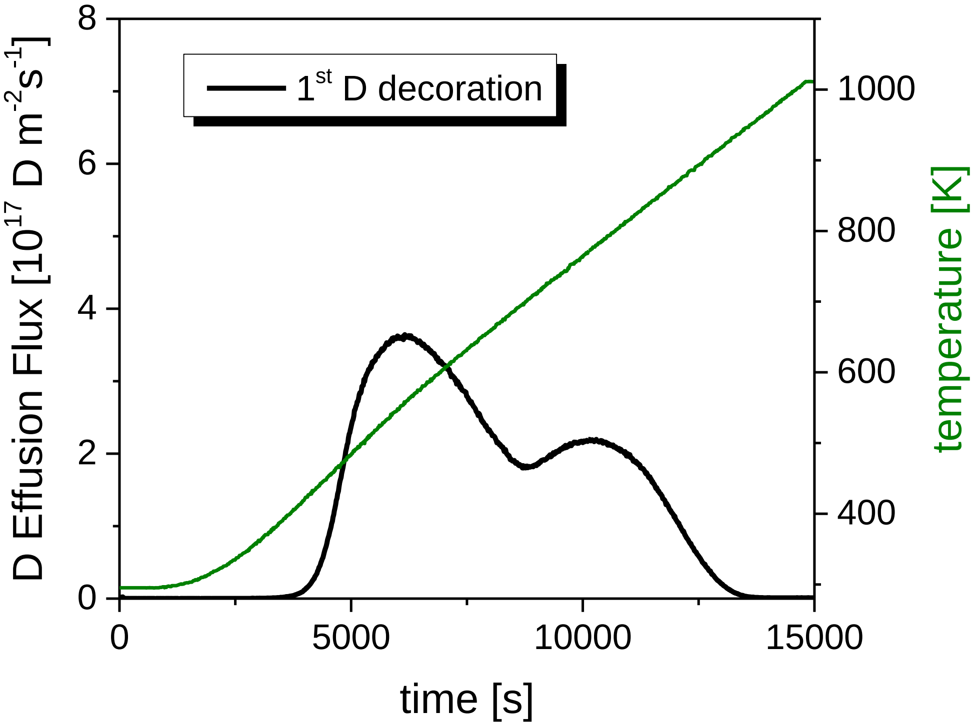
⇒ D gets efficiently re-trapped during W implantation



D effusion during thermal desorption



What happens to the D binding?



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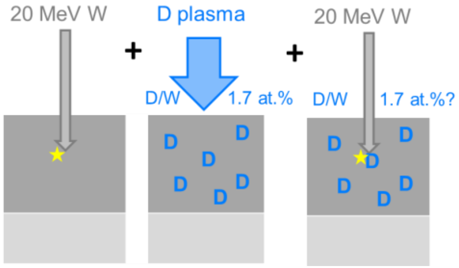
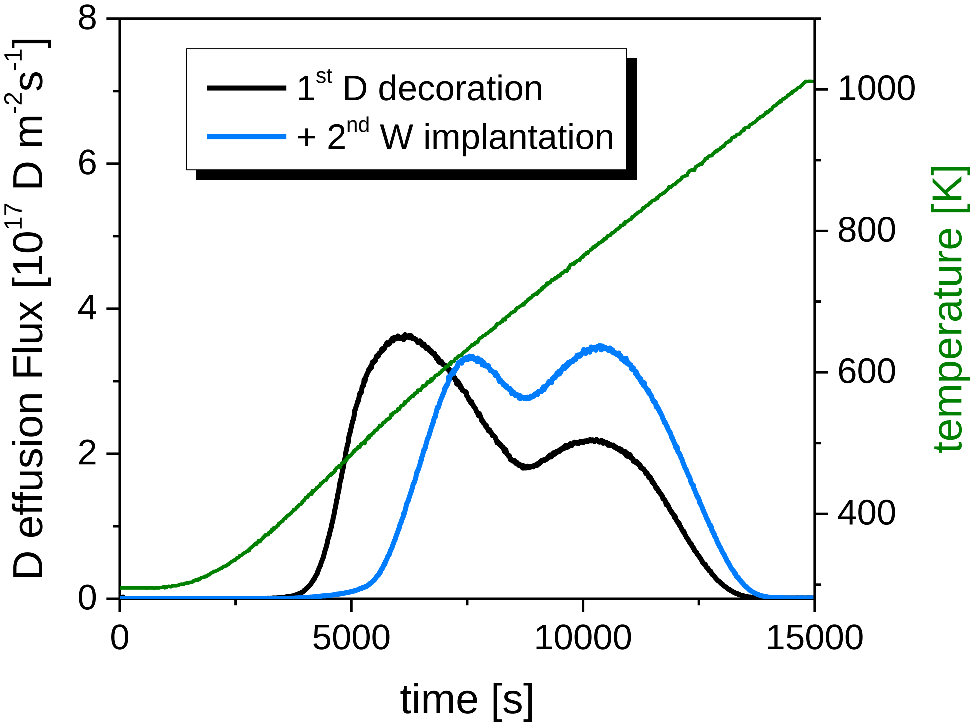


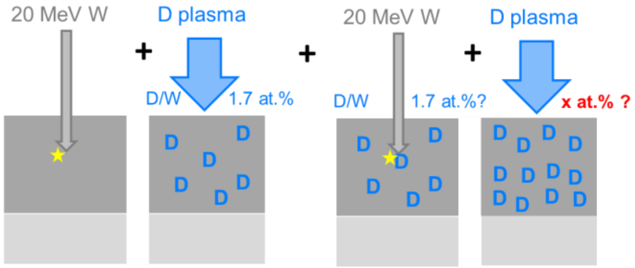
What happens to the D binding?

⇒ shift of desorption to
larger desorption energies!

⇒ new trap types?



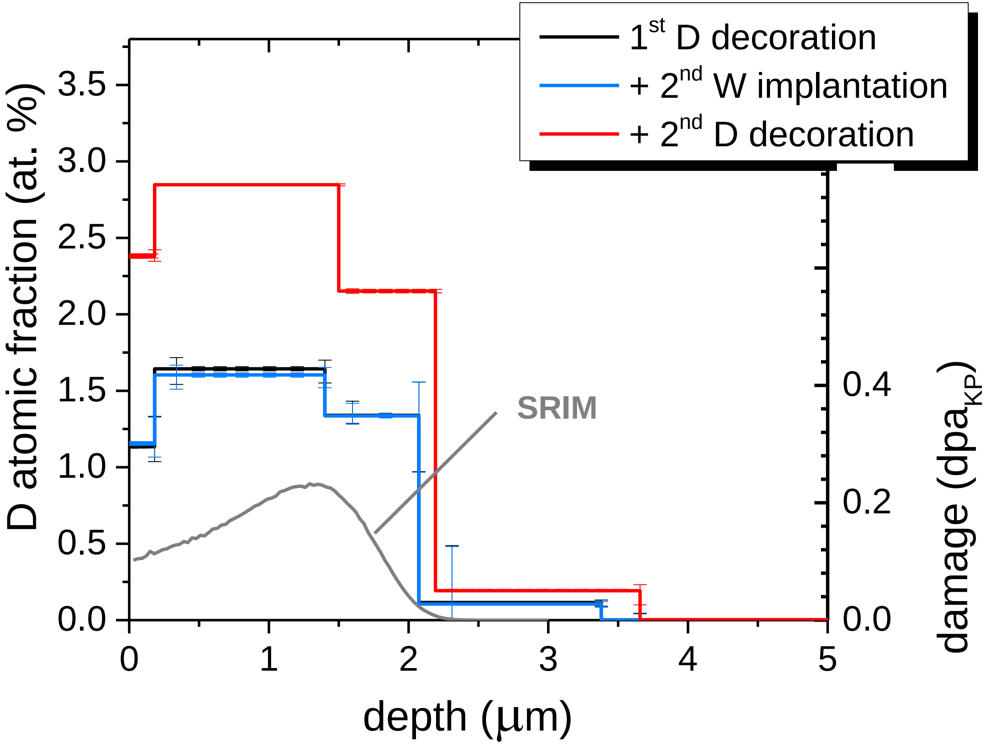
D depth profiles



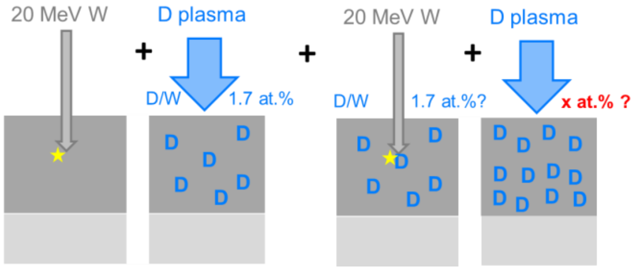
Decoration after 2^{nd} damaging:

⇒ D retention increases to 2.8 at.% (!) exceeding D at. fraction by ≈ factor 2 beyond previous ‘saturation value’!

⇒ new trap types now filled?



Thermal desorption spectroscopy

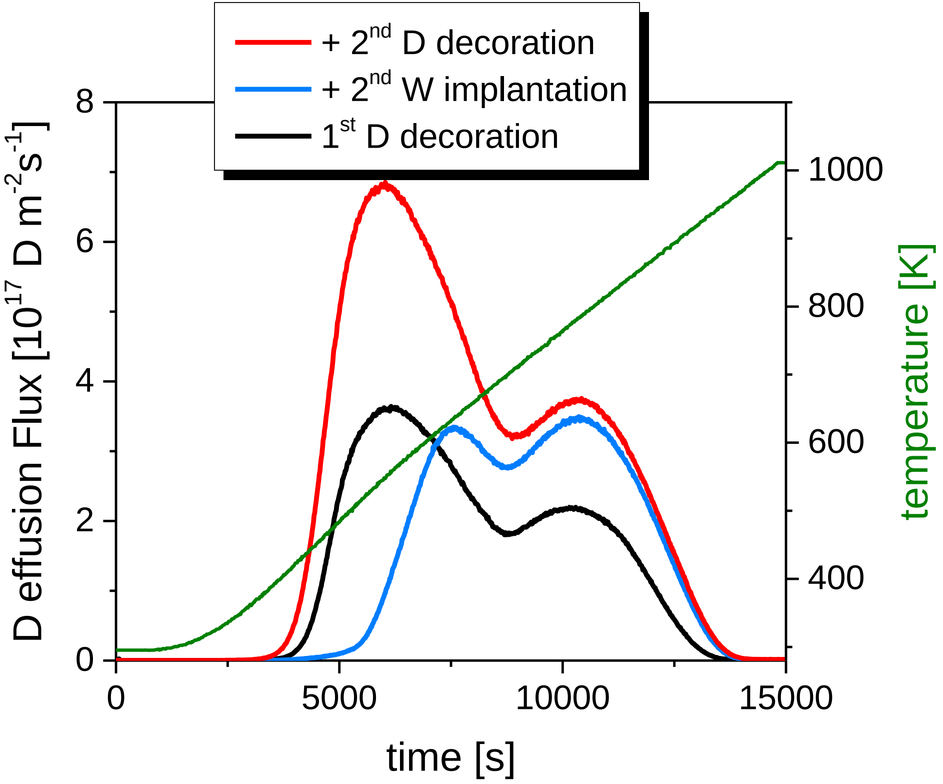


Decoration after 2nd damaging:

⇒ TDS spectra resembles again the spectrum of the initially decorated, singly damaged W!

⇒ only larger intensity

⇒ same trap types?

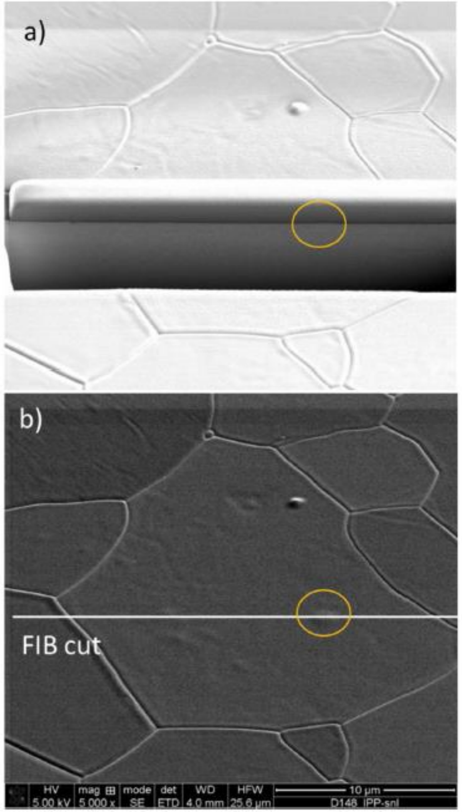


Artefact from surface blisters?

Suspicion:

- increased retention due to surface blisters?
  (unlikely giving the same depth profile)
- 30 SEM micrographs (30 µm in size) show no indication on gas filled cavities
- in line with dedicated studies such as a
  S. Kapser et al., Nucl. Fusion
- in line with lack of D_2 bursts in TDS spectra

SE images

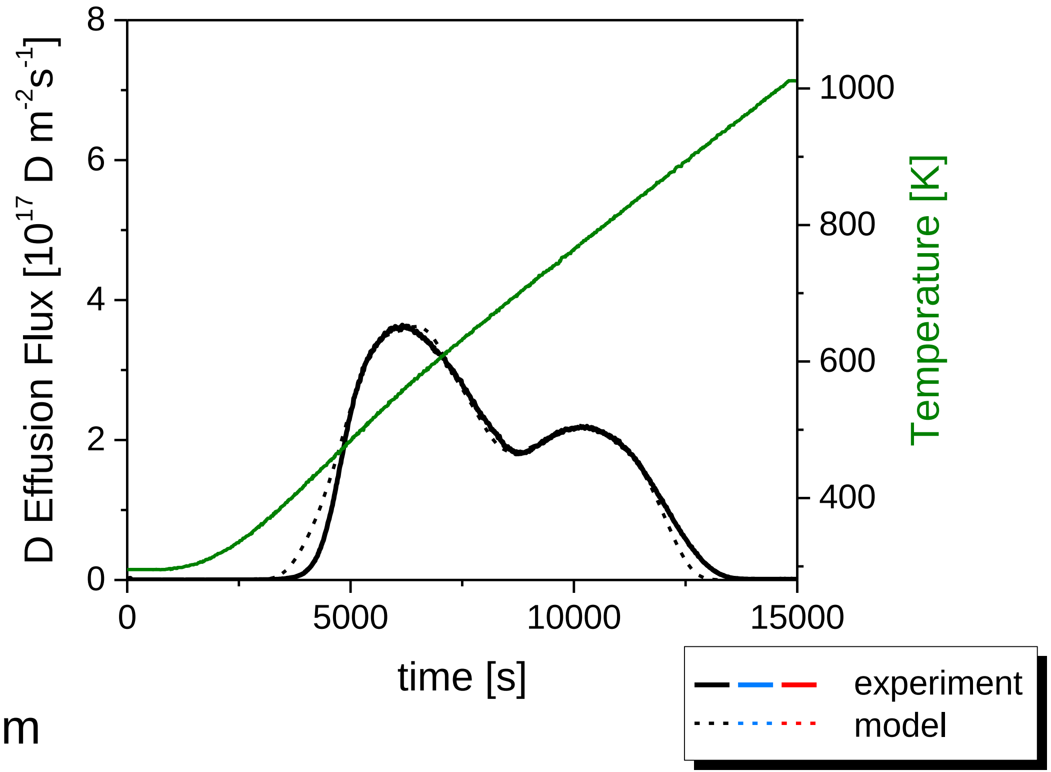


Rate equation modelling

TESSIM-X code with fill-level-dependent trapping [K. Schmid et al. JAP 116, 134901 (2014)]

Parameters from previous isotope exchange study:

- diffusion coefficient
  $D_0 = 1.58\times10^{-7}/\sqrt{2}$,
  $E_{diff} = 0.25$ eV
- one trap type with five fill-levels, (E_{detrap} = 1.18 eV, 1.32 eV, 1.46 eV, 1.7 eV, 1.84 eV, $\nu_0 = 1\ \text{x}10^{13}\ \text{s}^{-1}$),
- trap profile constant down to 2.2 µm



Rate equation modelling

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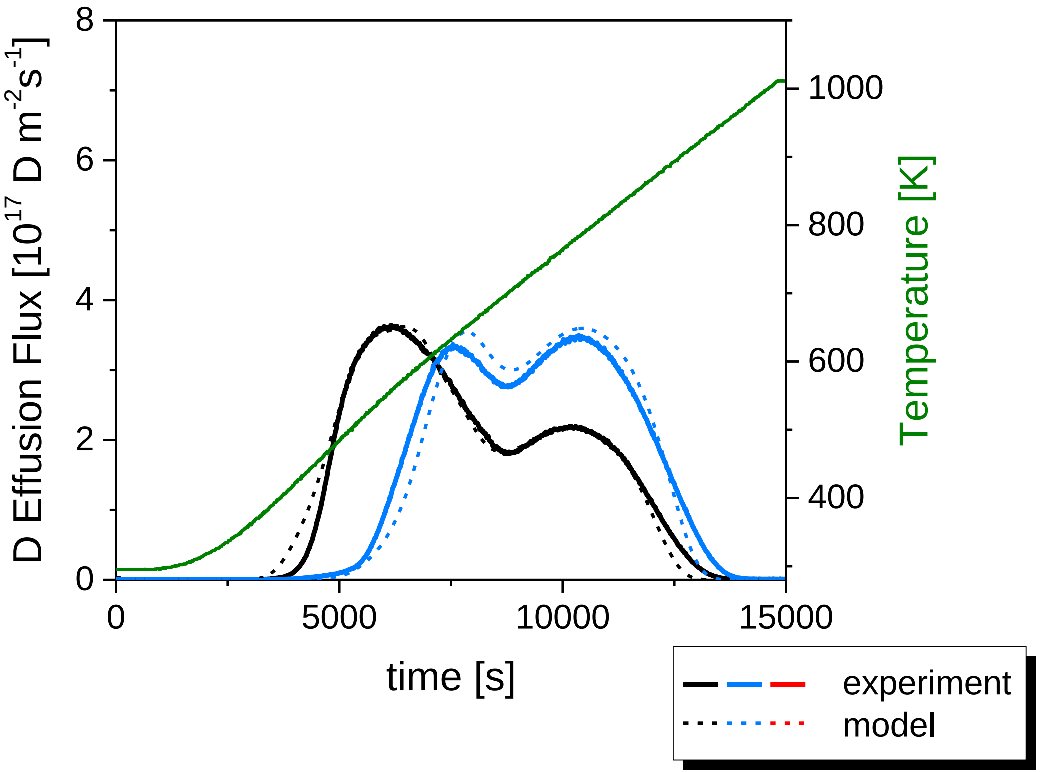
- creation of additional empty traps (by a factor of 1.7 during 2nd W implantation) explains temperature shift!
- TDS spectra with and without kinetic de-trapping indistinguishable



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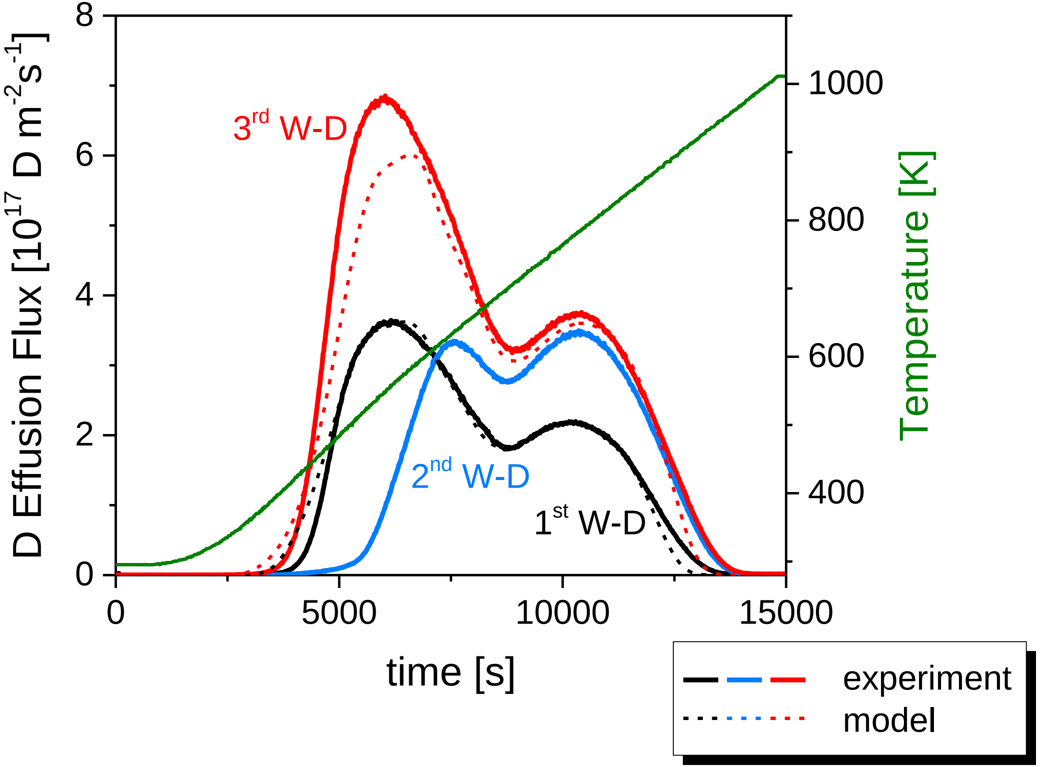
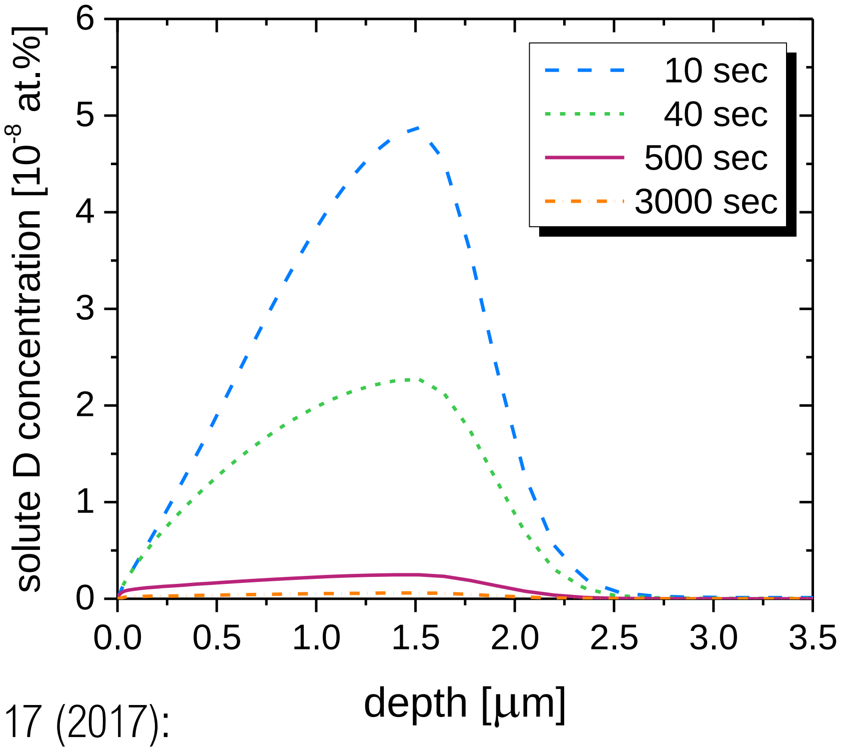


Rate equation modelling

- evolution of D solute concentration during 50 minutes W damaging
- differentiation between solute and retained D meaningful on timescale of damage cascade?
- D is static on ps timescale (1 Å in 1 ps @ 2000 K)

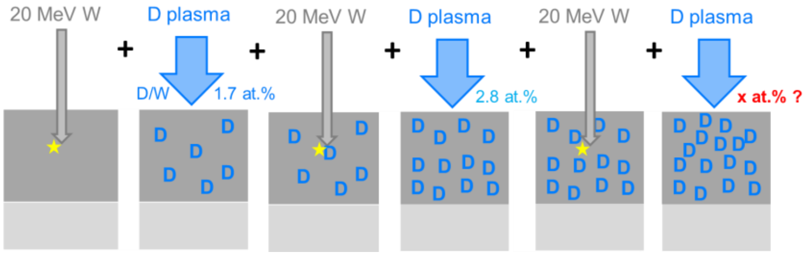


See: T. Schwarz-Selinger et al. *Nucl. Mater. Energy* 17 (2017): 228–34. https://doi.org/10.1016/j.nme.2018.10.005.

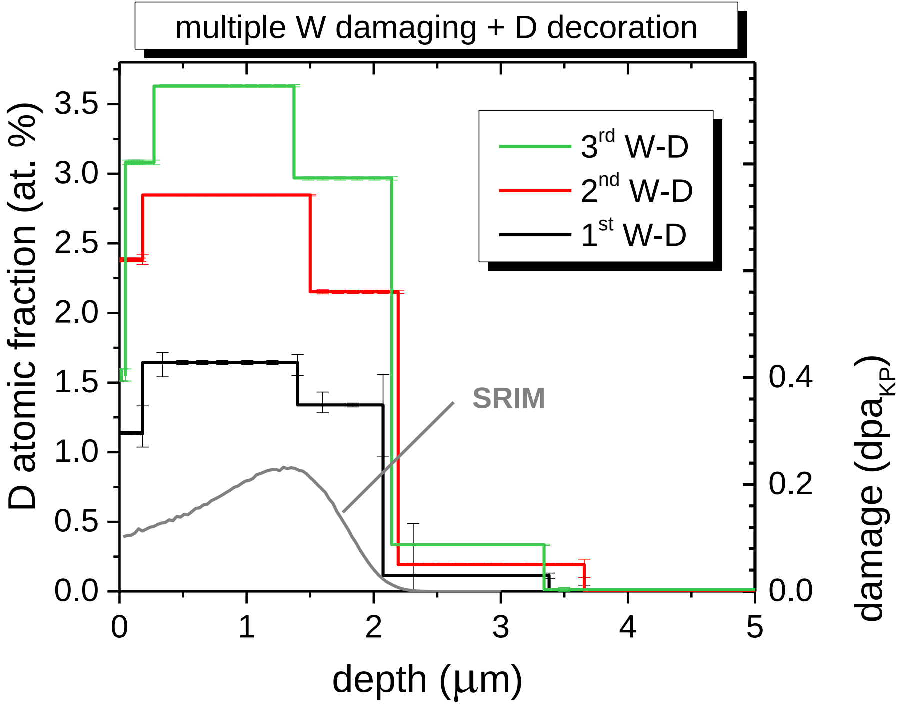
Triple damaging

- Did we reach with 2.8 at. % the maximum D concentration?

- Triple damaging



- **⇒ 3.6 at. %**



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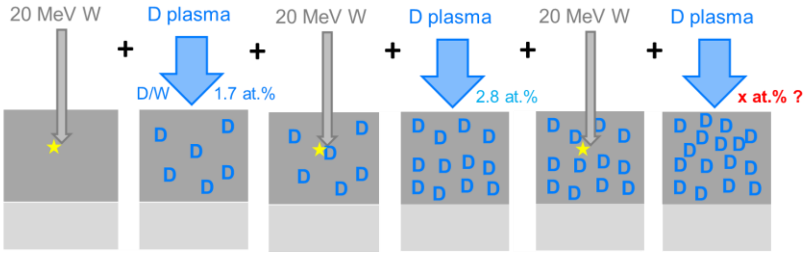

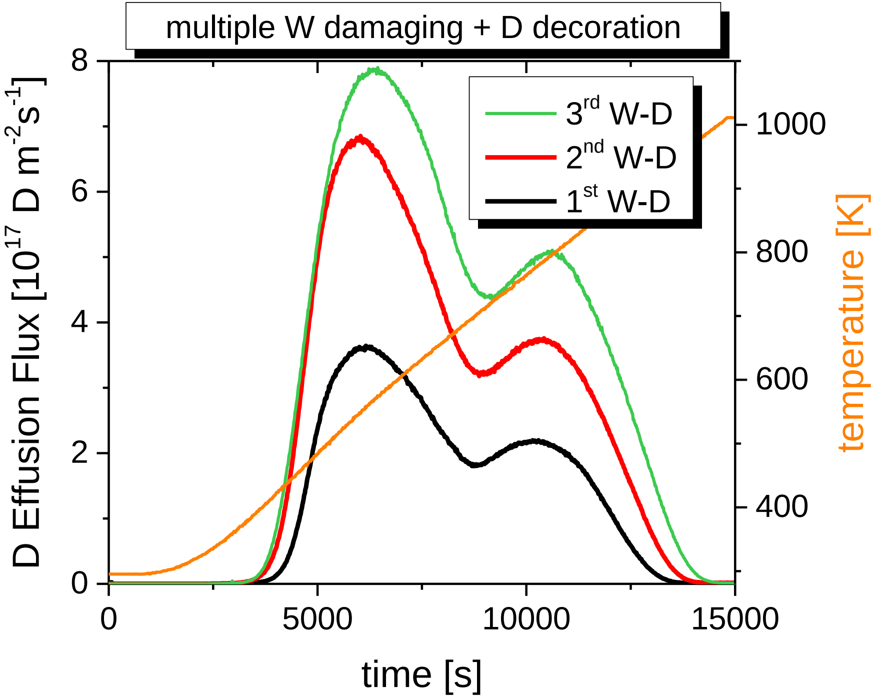

Damage stabilization modelling

Damage stabilization model: $$\frac{dn_i(x,t)}{dt} = \frac{\Gamma_W\ \eta\ \Theta(x)}{\rho}\left[1 - \frac{\mathrm{n_i}(x,t)}{\mathrm{n_{i,max}}}\left(1 - \alpha_i \underbrace{\frac{\mathrm{n_i}(x,t) - n_i^0(x,t)}{\mathrm{n_i}(x,t)}}\right)\right]$$

ratio of D occupied defects of type *i*

$n_i(x, t)$: density of defect type *i*.

Γ_W: flux of damaging W particles,

$\Theta(x)$: SRIM calculated primary damage profile

η: probability of an impinging W particle to create a defect per unit length

ρ: density of tungsten

n_i^0: density of empty defects of type *i*

Free parameters of the model

$n_{i,max}$: saturation density of defect type *i*,

α_i: stabilization parameter for defect type *i*.

[M. Pečovnik et al. submitted to Nucl. Fusion]

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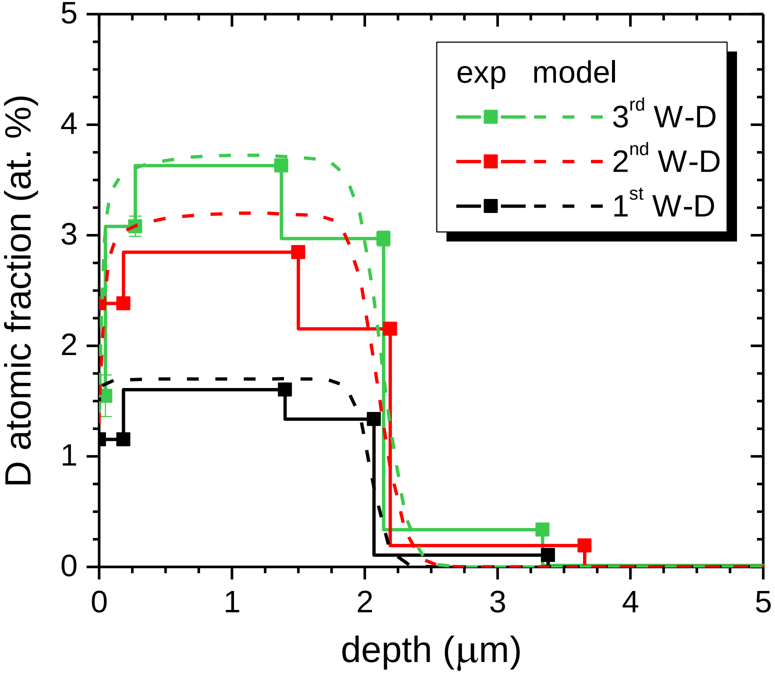
$$\frac{dn_i(x,t)}{dt} = \frac{\Gamma_W\ \eta\ \theta(x)}{\rho}\left[1 - \frac{\mathrm{n_i}(x,t)}{\mathrm{n_{i,max}}}\left(1\ -\ \alpha_i\,\frac{\mathrm{n_i}(x,t) - n_i^0(x,t)}{\mathrm{n_i}(x,t)}\right)\right]$$

- defect type I: n_{max} = 0.21 at.%
  fill level energies: 1.07 eV, 1.15 eV,
  1.23 eV, 1.33 eV, 1.43 eV,
- defect type II: n_{max} = 0.29 at.%
  fill level energies: 1.66 eV, 1.84 eV,
- defect type III: n_{max} = 0.04 at.%
  fill level energy: 2.06 eV.

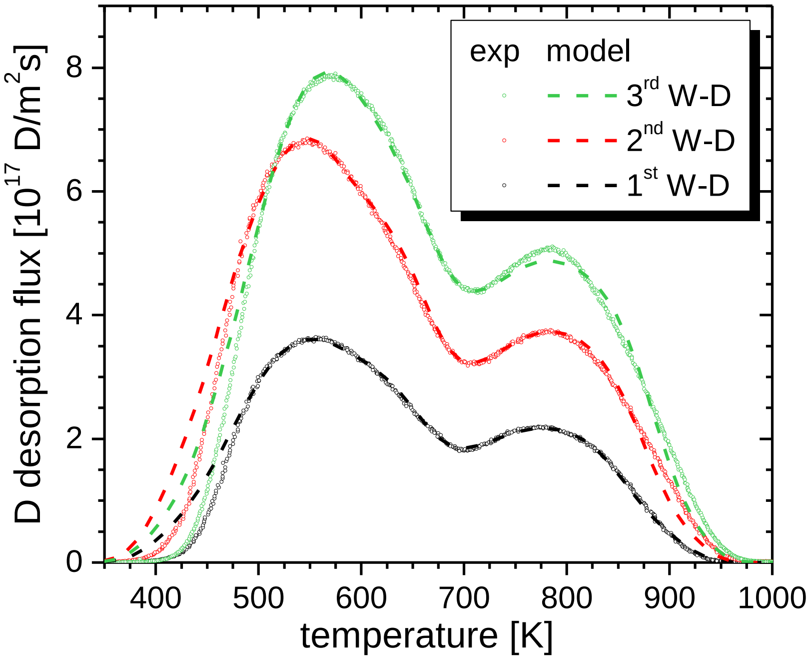


Damage stabilization modelling

Damage stabilization model:

$$\frac{dn_i(x,t)}{dt} = \frac{\Gamma_W\ \eta\ \theta(x)}{\rho}\left[1 - \frac{\mathrm{n_i}(x,t)}{\mathrm{n_{i,max}}}\left(1\ -\ \alpha_i\,\frac{\mathrm{n_i}(x,t) - n_i^0(x,t)}{\mathrm{n_i}(x,t)}\right)\right]$$

- defect type I: n_{max} = 0.21 at.%
  fill level energies: 1.07 eV, 1.15 eV,
  1.23 eV, 1.33 eV, 1.43 eV,
- defect type II: n_{max} = 0.29 at.%
  fill level energies: 1.66 eV, 1.84 eV,
- defect type III: n_{max} = 0.04 at.%
  fill level energy: 2.06 eV.



Damage stabilization modelling

Damage stabilization model: $$\frac{dn_i(x,t)}{dt} = \frac{\Gamma_W \eta\, \theta(x)}{\rho}\left[1 - \frac{\mathrm{n_i}(x,t)}{\mathrm{n_{i,max}}}\left(1 - \alpha_i \frac{\mathrm{n_i}(x,t) - n_i^0(x,t)}{\mathrm{n_i}(x,t)}\right)\right]$$

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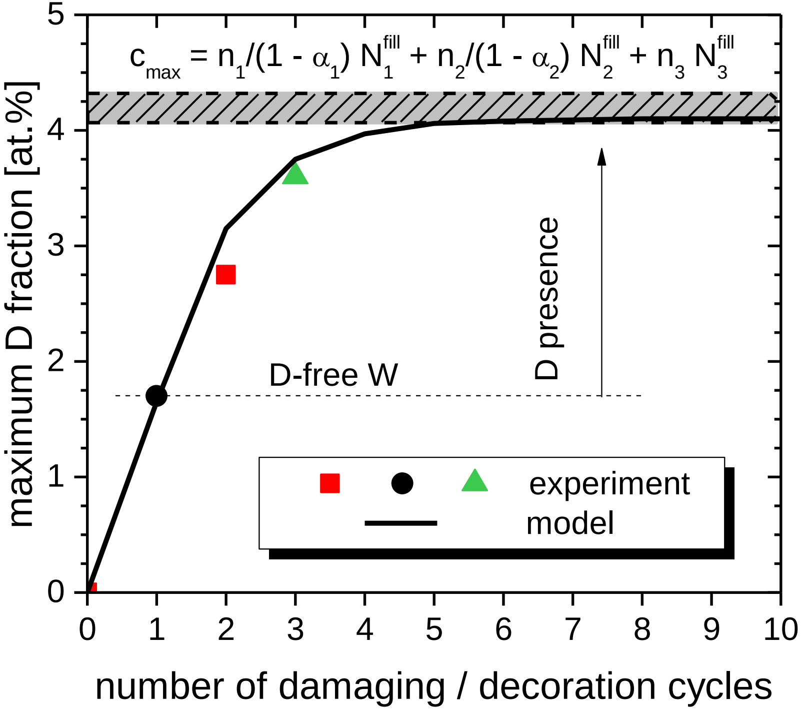
  fill level energies: 1.66 eV, 1.84 eV,
- defect type III: n_{max} = 0.04 at.%
  fill level energy: 2.06 eV.
- Fill levels: $N_1^{fill} = 5$, $N_2^{fill} = 2$

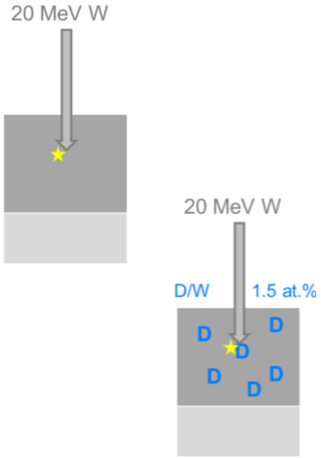


Present interpretation

Different saturation/defect levels:

- D free W: saturation in n_{Dmax} = 1.7 at.% above 0.2 dpa
  because
  Frenkel pairs can annihilate with existing ones

- D filled W: n_{Dmax} = 4.2 at.%
  because
  newly created defects cannot annihilate with existing ones
  when they are occupied by D: stabilization



Summary

Influence of the presence of D on displacement damage

- Multiple sequences of creating displacement damage and decorating defects with D allows to study the influence of D on damage creation/stabilization (even at low temperatures) without the need for a dual beam in-situ setup
- **D retention exceeds the initial 'saturation value' by more than a factor of two (at 290 K damaging) n_D = 1.7 at.% ⇒ 2.8 at.% ⇒ 3.6 at.% ⇒ … 4.x at.%**
- No D is lost during consecutive W implantations / D is de-trapped but is effectively re-trapped
- D is redistributed from the low temperature de-trapping peak to the high temperature de-trapping peak (during W irradiation or during TDS)
- TDS shows no indication for new defect nature but only increased density
- Rate equation modelling successful with increased defect density only
- Damage stabilization model describes observation successfully

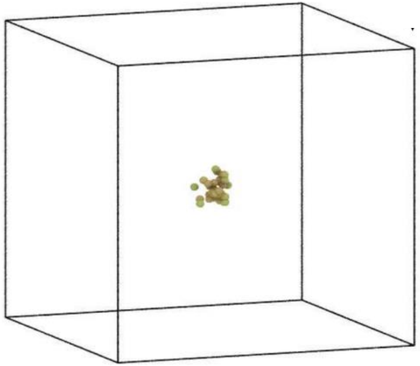
Backup slides

Related MD modelling

F. J. Domınguez-Gutierrez and U. von Toussaint

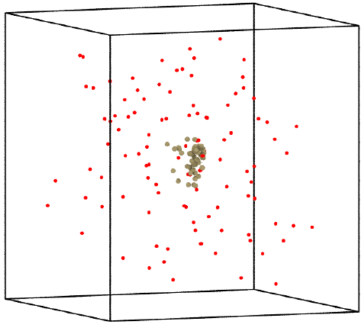
- Simulating the cascade core by heating to 10000 K for 5 ps (using a Langevin thermostat) to emulate the core region of a collision cascade with and without D present (work in progress)
- Using descriptor based method [*F. J. Domınguez-Gutierrez and U. von Toussaint, submitted to J. Nucl. Mater*] to identify defects created (IAEA challenge winner)

D free



32 point defects

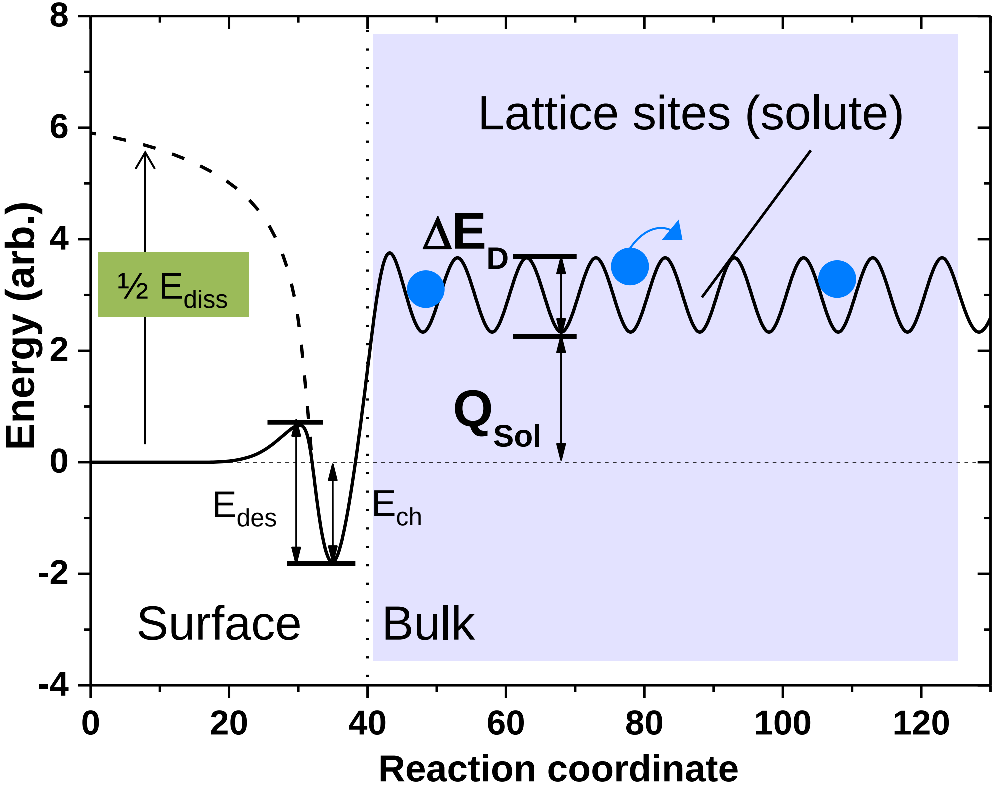
2% D



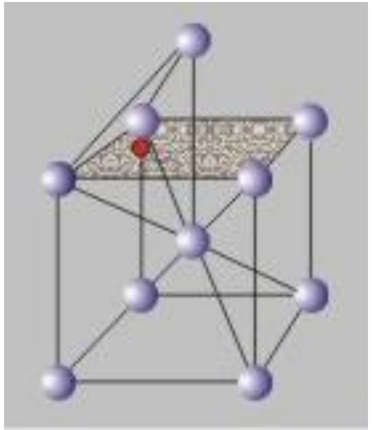
41 point defects

PFMC 2019

Potential diagram

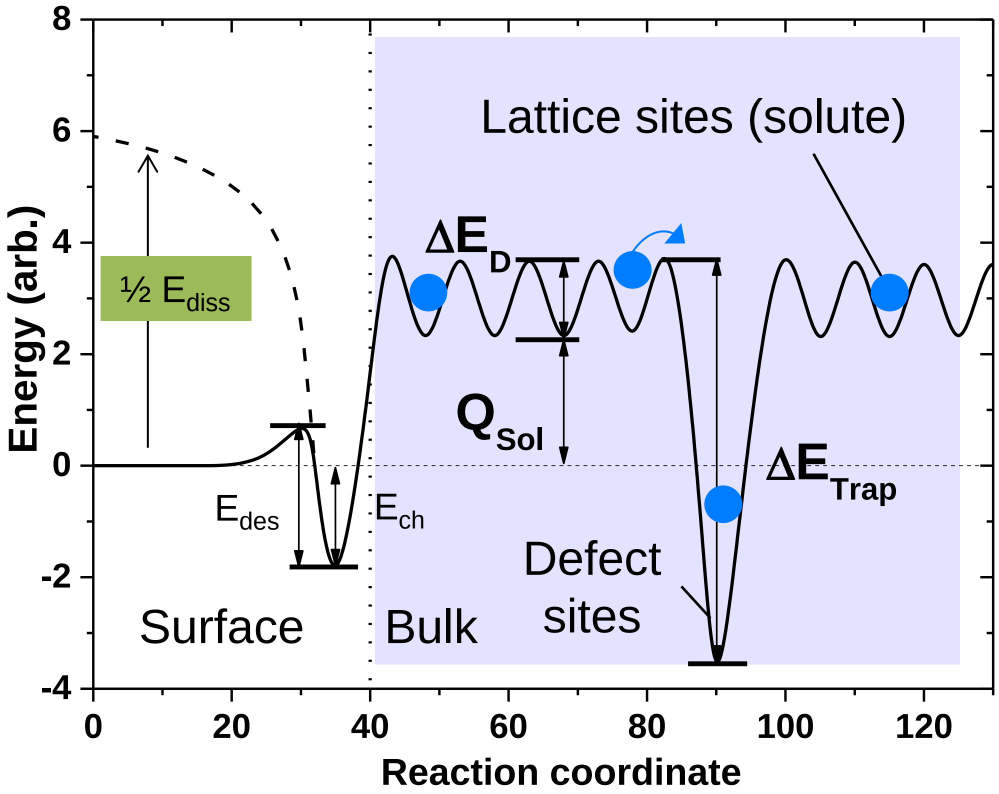


Tetrahedral sites in



bcc lattice

Potential diagram



Parameters for W

½ E_{diss}= 2.25 eV
E_{ch}=0.5 – 0.8 eV
Q_{Sol}=1.04 eV

Two populations:

ΔE_D = 0.39 eV (better 0.25 eV?)

$\Delta E_{trap} \approx 0.8 – 2$ eV

Defect evolution during gentle loading

- D retention in W foils after 192 h plasma exposure (fluence ≈ 4·10^{25} D/m²)

S. Kapser et al., Nucl. Fusion, 2018 http://dx.doi.org/10.1088/1741-4326/aab571)