Comparison of a Quantitative Diffusion-Trapping Model With Experiments on D Uptake in Damaged W

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- Introduction
- Model and Parameters
- Fluence and Temperature Variations
- Possible Mechanisms for Temperature-Dependent D Uptake
- Summary

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Introduction

Introduction: Filling of (damaged) W with D

- **Model system** for **benchmarking** of **diffusion-trapping** code **TESSIM** [1]
  - ➢ Polycrystalline **tungsten damaged** by **MeV W** ions
  - ➢ **“Gentle” plasma loading**
    - ➷ **Decoration** of traps with D **without further modification** of sample
  - ➢ **Analyse trapped D**
    - ➷ Nuclear reaction analysis (NRA): **depth profile**
    - ➷ Temperature programmed desorption (TPD): **binding energies**

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[1] K. Schmid et al., J. Nucl. Mater. 426, 247–253 (2012)

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- Take as many **input parameters** for simulation from **literature**
- Determine the remaining parameters **experimentally**
- Tune the model to **one reference case**
- Try to **predict other experiments** without further parameter adjustments

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- **Motivation:** Huge **discrepancy** of modelling and experiments at **Pilot-PSI** [2]
  - ➢ **Uptake** of **low-energy D** (at high plasma flux) surprisingly **slow**
  - ➢ Can only be reproduced by model if **incident flux** is **drastically reduced**
  - ➢ **Strong temperature dependence** of effective D uptake

[1] K. Schmid et al., J. Nucl. Mater. 426, 247–253 (2012)
[2] M.J.H. t'Hoen et al., Phys. Rev. Lett. **111** (2013) 225001

Model and Parameters

Diffusion-Trapping Model TESSIM

- **TESSIM** code: implemented in Mathematica by K. Schmid [1]
- Same basic principle as, e.g., TMAP7 [3]
  - ➢ Rate equations for **diffusion, trapping, implantation** and **surface loss**
- Highly **flexible** and **adaptive** code
  - ➢ Allows **arbitrary resolution** of **calculation grid**
  - ➢ Allows introducing any process that can be mathematically formulated (and is physically sensible), e.g.,
    - ↳ **Parametrised,** spatially distributed implantation source term
    - ↳ Trap densities evolving with time/fluence (e.g., blistering!)
- Good parallelisation by using Mathematica‘s NDsolve package
  - ➢ Currently using Method of Lines to solve PDE system

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[1] K. Schmid et al., J. Nucl. Mater. 426, 247–253 (2012)
[3] G. Longhurst (2008) INEEL/EXT-04-02352Rev.2

Reference Case and Input Parameters

- Sample: **recrystallized W**, damaged by **20 MeV W^{6+}** ions **(~0.5 dpa)**
  - ➢ Very **low background trap density**
  - ➢ W fluence sufficient to approximately **saturate radiation damage**
  - ➢ **No blistering** or strong **“surface peak”** observed
    - ⇘ **Trap density** in damaged region appears **unaffected** by plasma exposure

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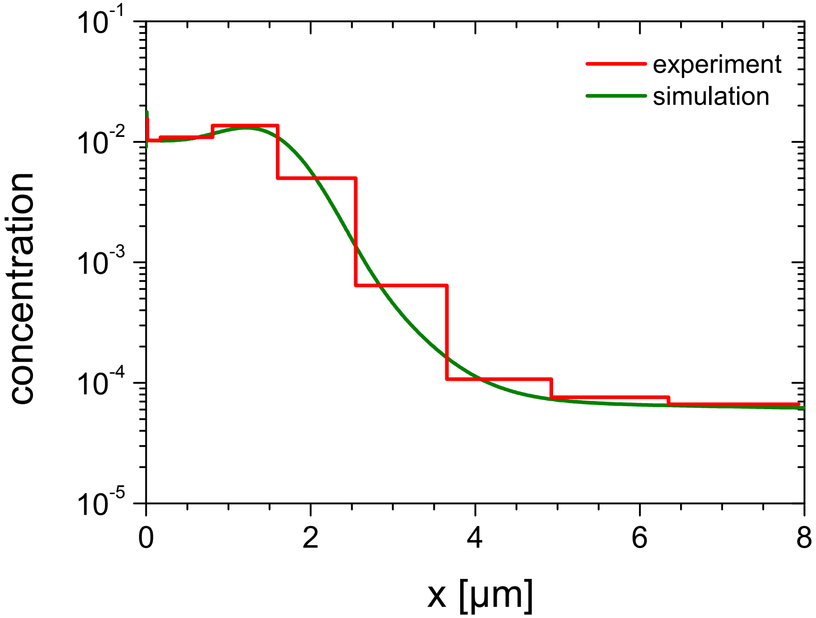
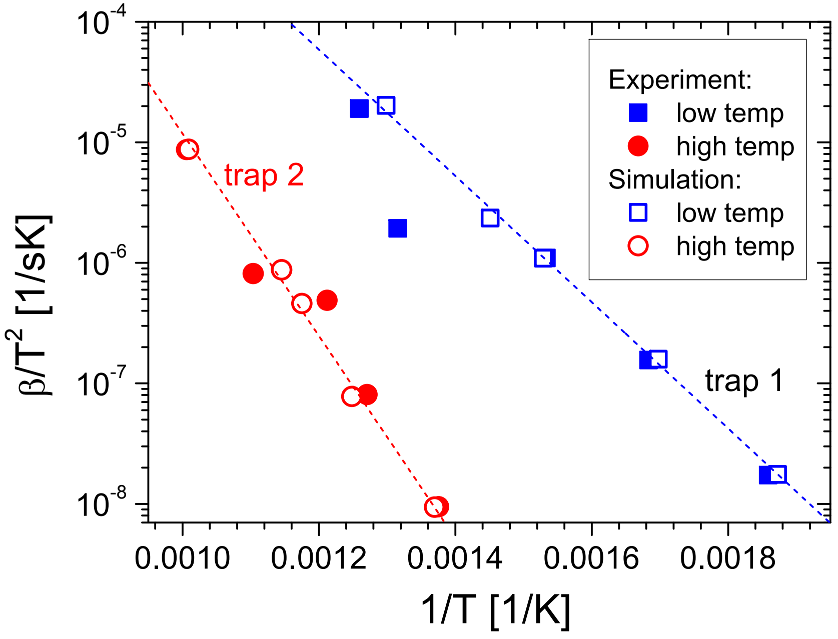
    - ➭ **Trap density** in damaged region appears **unaffected** by plasma exposure
- **Reference** for simulations:
  - ➢ **3 days (72 h)** of plasma exposure at **floating potential** and **450 K**
- Total **trap density** replicates **NRA depth profile** of **D-saturated sample**



Reference Case and Input Parameters

- Trap **binding energies** and **pre-factors** from **TPD** with **ramp variations**
  - Low energy trap: **1.0 eV, 1×10^{10} s^{-1}** (damage peak and bulk)
  - High energy trap: **1.5 eV, 6×10^{10} s^{-1}** (damage peak only)



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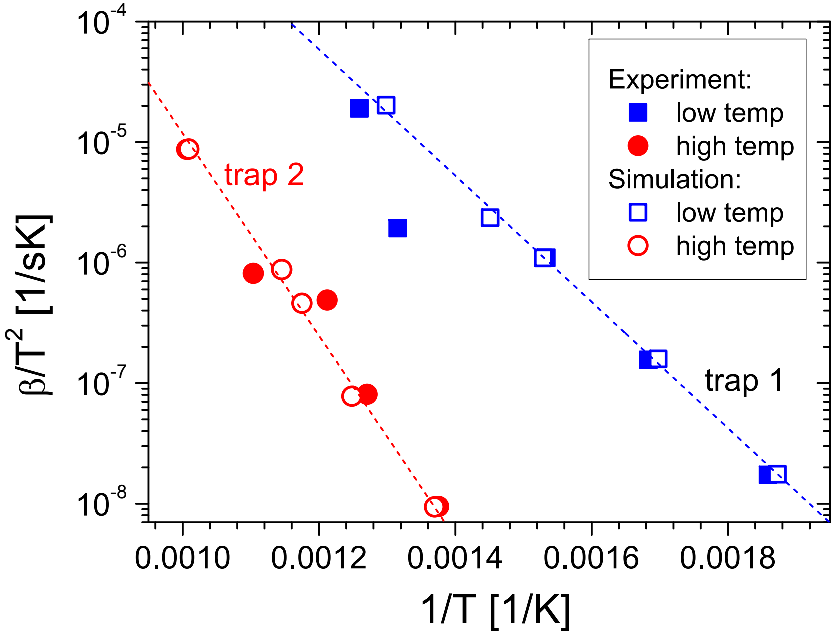
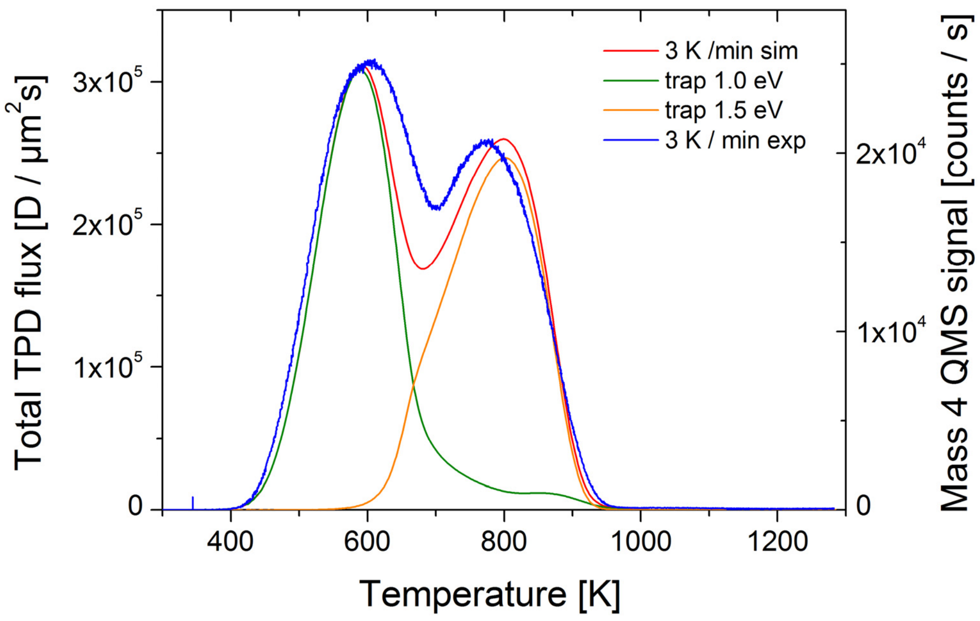
- **Density ratio** of traps replicates **TPD peak ratio**
  - ➢ **High energy** trap only in **damaged zone**
  - ➢ **Low energy** trap in **damaged zone** as well as in **bulk**





Reference Case and Input Parameters

- **Incident flux: 5.6 – 9×10^{19} D/m^2s** (97% from D_3^+, 2% from D_2^+, 1% from D^+)
  - ➢ Values from **characterisation** of plasma source **PlaQ** [4]
  - ➢ **Implantation profile**: parametrisation of **SDTrim.SP** calculations
    - ➯ For low ion energies as used here: **Gaussian fit** to profile is adequate
  - ➢ **Reflection yield: Eckstein's fit** for light projectiles [5]
  - ➢ Implantation profile and reflection yield **for each ion species**
  - ➢ Implemented as **functions of sample bias** voltage
- **Plasma potential 15 eV,** (negative) **bias** voltage **0 V (floating)** or **100 V**

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- **Plasma potential 15 eV,** (negative) **bias** voltage **0 V (floating)** or **100 V**
- **Diffusion coefficient: Frauenfelder's** expression [6] **divided by √2** (for D!)
- **Assumption: Boundary condition** at surface: **c = 0**
  - ➢ **Surface loss of D is "infinitely" fast**
    - ⇘ **Surface desorption** of D_2 (or D) is **not** the **rate-determining** step
    - ⇘ Instead: determined by **transport** of D within W **to the surface**

[4] A. Manhard et al., Plasma Sources Sci. Technol. **20** (2011) 015010

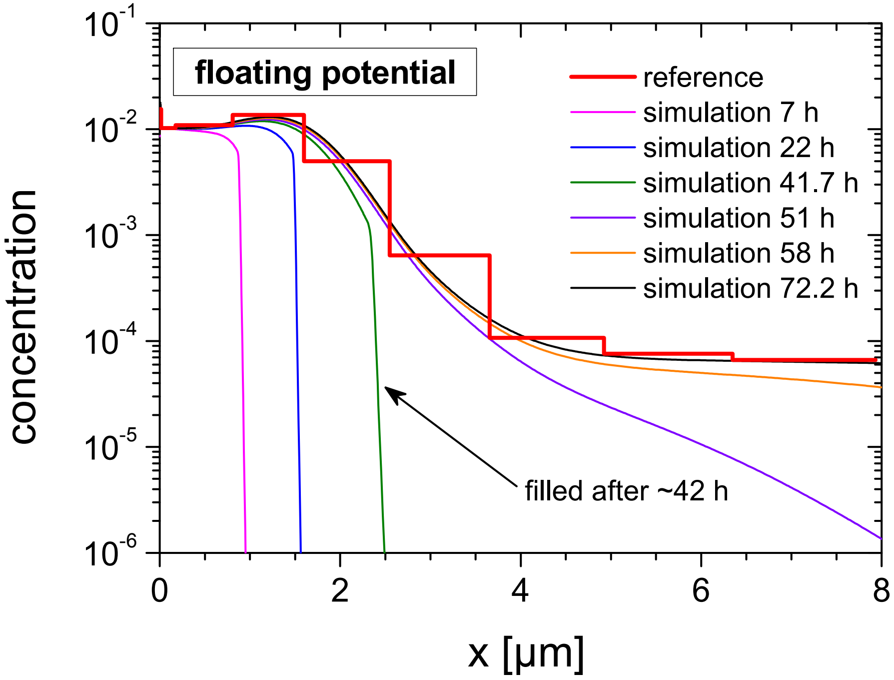
[5] W. Eckstein, IPP Report **17/12** (2009)

[6] R. Frauenfelder, J. Vac. Sci. Technol. **6(3)** (1969) 388-397

Fluence Variation

Filling of damaged W with D

- Comparison: **filling of damaged W** (**20 MeV** W beam, **0.5 dpa**) in **PlaQ**
  - ➢ without bias (**~5 eV/D**) at **450 K**



- Damaged zone filled after **42 hours** without bias

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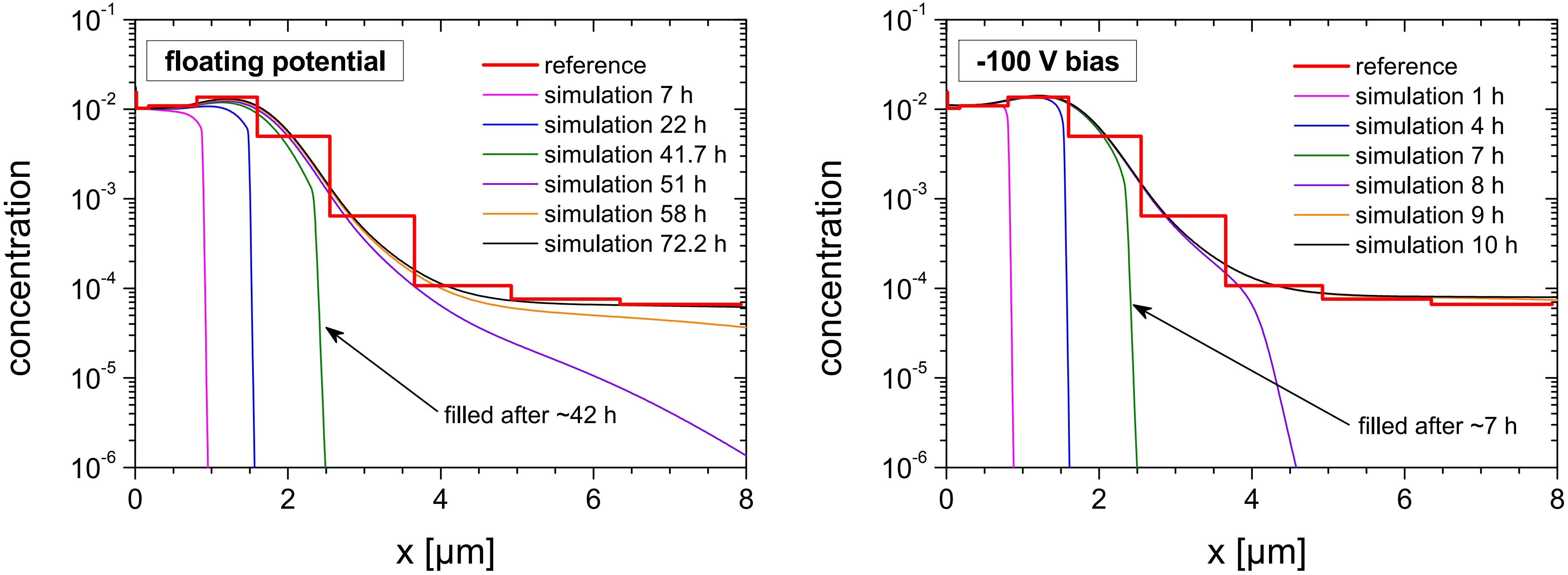
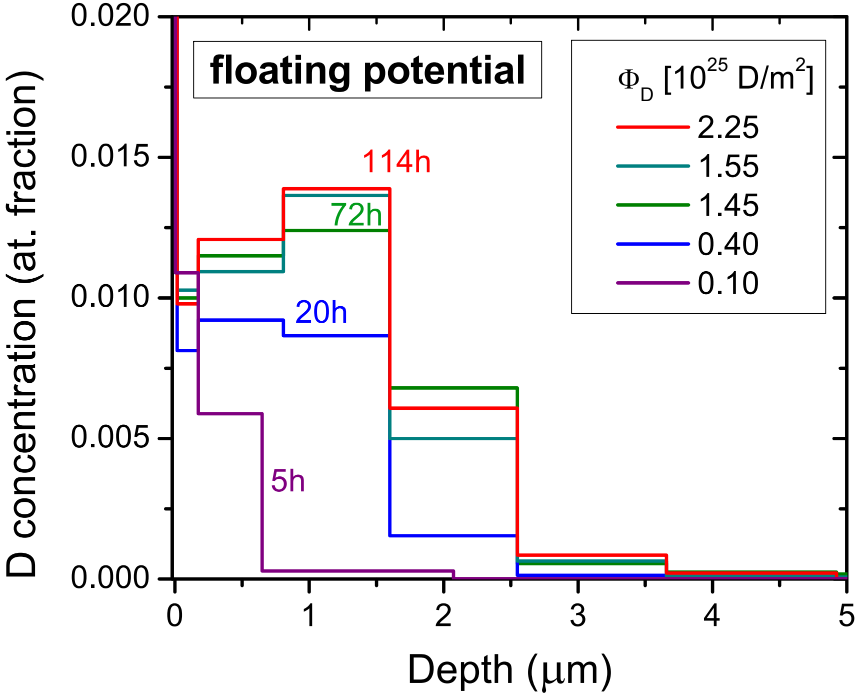
- Comparison: **filling of damaged W** (**20 MeV** W beam, **0.5 dpa**) in **PlaQ**
  - ➢ without bias (**~5 eV/D**) at **450 K**
  - ➢ with -100 V bias **(~38 eV/D**) at **450 K**



- Damaged zone filled after **42 hours** without bias, after **7 hours** with 100 V bias
  - ➢ Approximately **6x faster** filling **with bias**

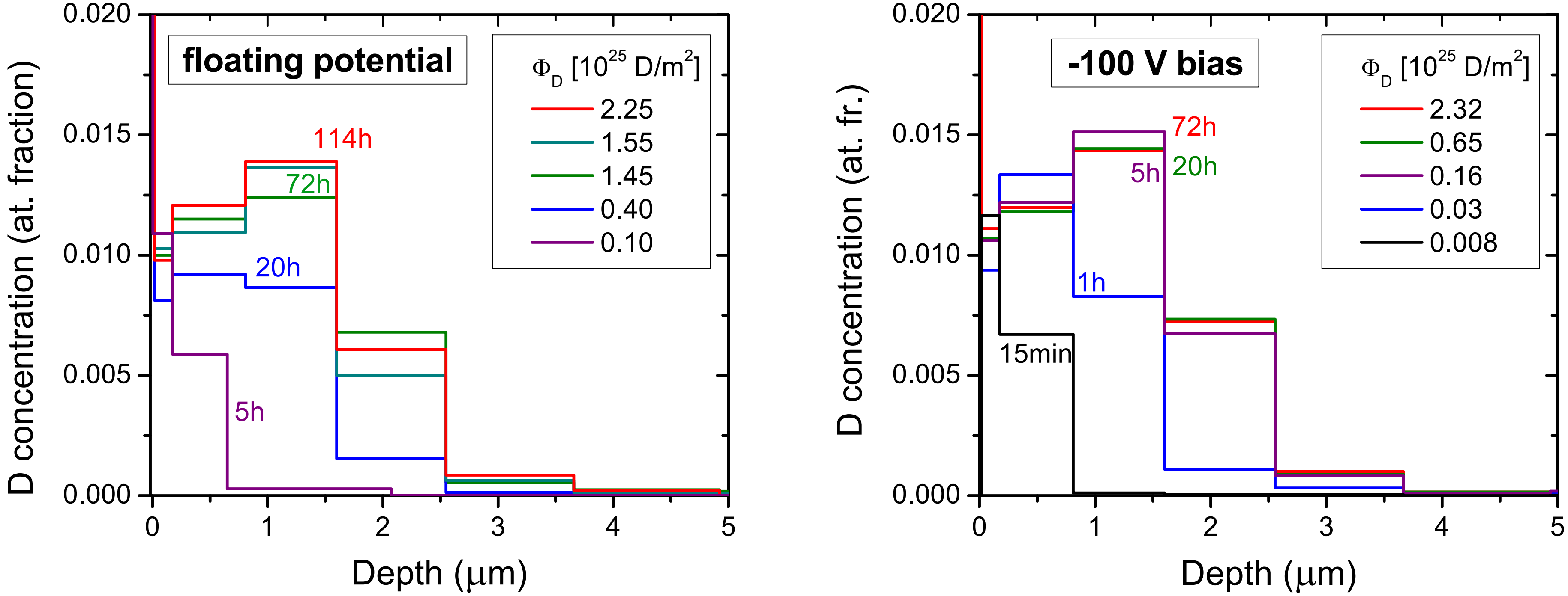
Fluence series in PlaQ for damaged W

- **Experiment:** fluence variation at **450 K** sample temperature
  - ➢ **More than 20 h, less than 72 h** filling time at **floating potential**



Fluence series in PlaQ for damaged W

- **Experiment:** fluence variation at **450 K** sample temperature
  - **More than 20 h, less than 72 h** filling time at **floating potential**
  - **More than 1 h, less than 5 h** at **-100 V bias**
  - **Closely matched by simulations**



Temperature Variation

Temperature dependence of filling: floating potential

- Influence of **temperature** on filling of damaged W **(290 – 650 K)**
- Samples loaded with D at **floating potential** for **7 hours** each **(partially filled)**
  - ➢ Compare **total retention** in samples

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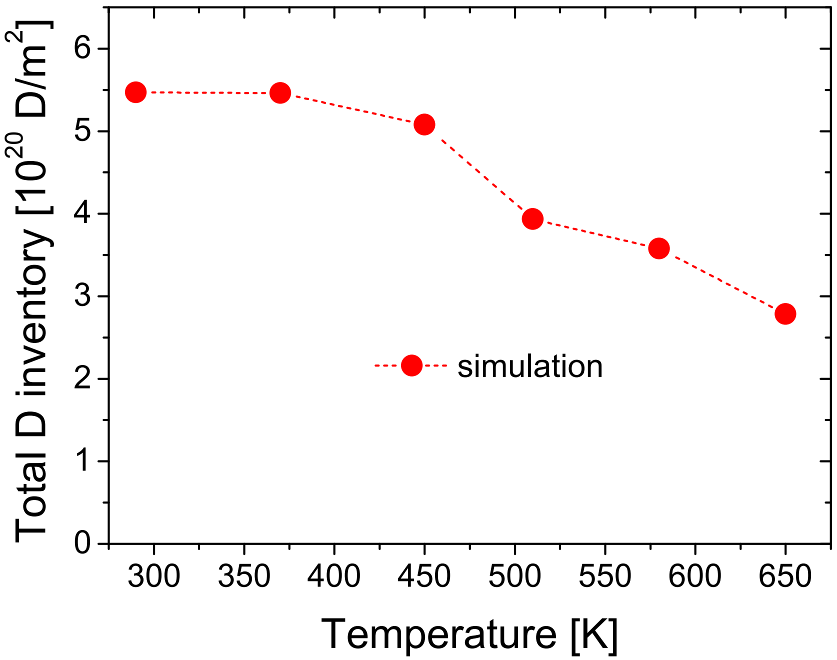

- Prediction: Inventory after 7 h nearly **independent** from **temperature** for ≤ **450 K!**
  - ➢ **Rate-limiting step** is the **filling of traps** (not diffusion to traps)
  - ➢ **Higher** temperatures: **traps not fully populated** (detrapping!)

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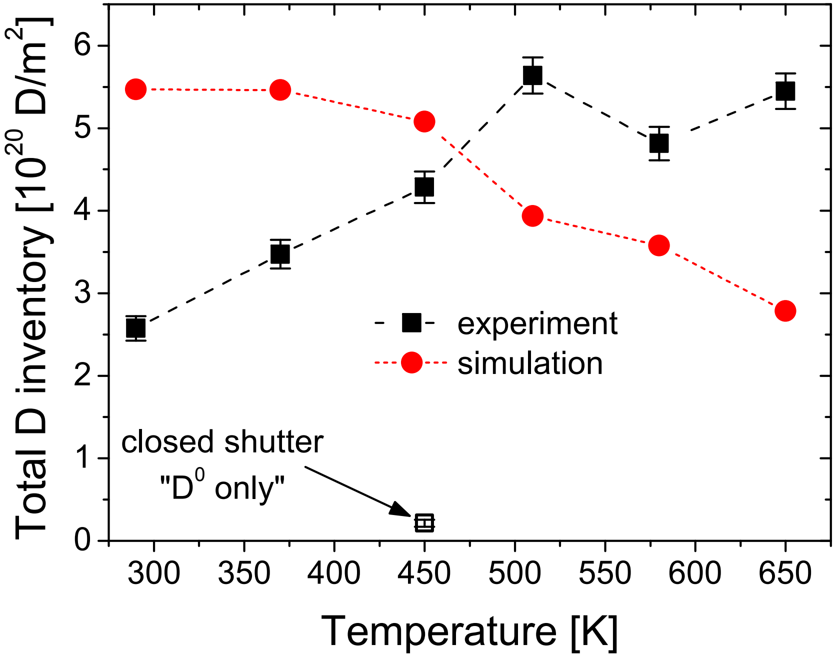

- **Experimental data:** total inventory **steadily increases** with temperature
  - ➢ **Close match at 450 K** → crossover point of trends
- **Trap densities** and **parameters fixed** by other experiments (450 K, TPD)
- Model results **insensitive** to **diffusion coefficient** (within 100x more or less)

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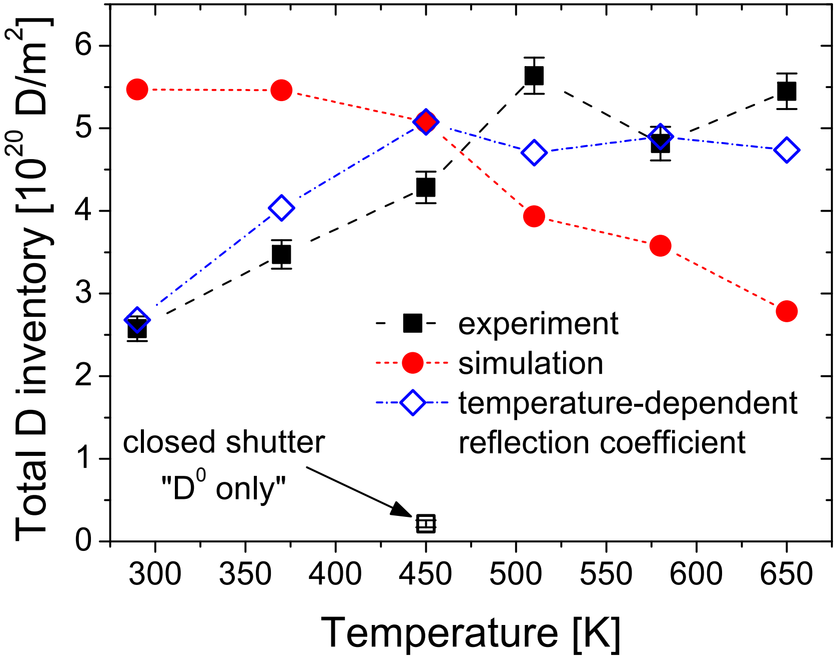

- Only parameter available to change results: **effective implantation flux**
  - ➢ Temperature-dependent D uptake with activation energy **~0.1 eV → match**
  - ➢ Same direction, but much **weaker dependence** than **t'Hoen et al. (1-2 eV**) [2]

[2] M.J.H. t'Hoen et al., Phys. Rev. Lett. **111** (2013) 225001

Temperature dependence of filling: with bias

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- Samples loaded with D at **-100 V bias** for **45 minutes** each **(partially filled)**
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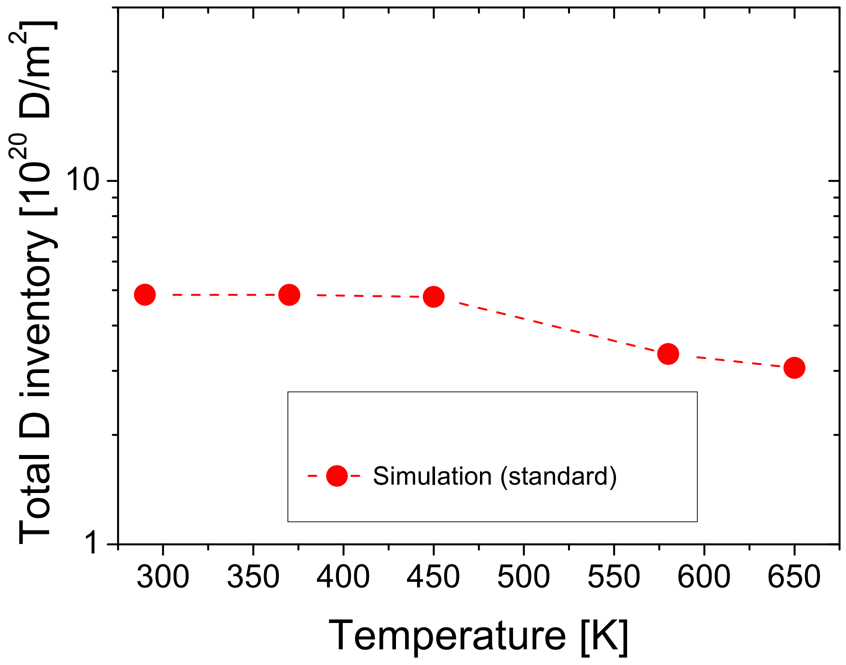

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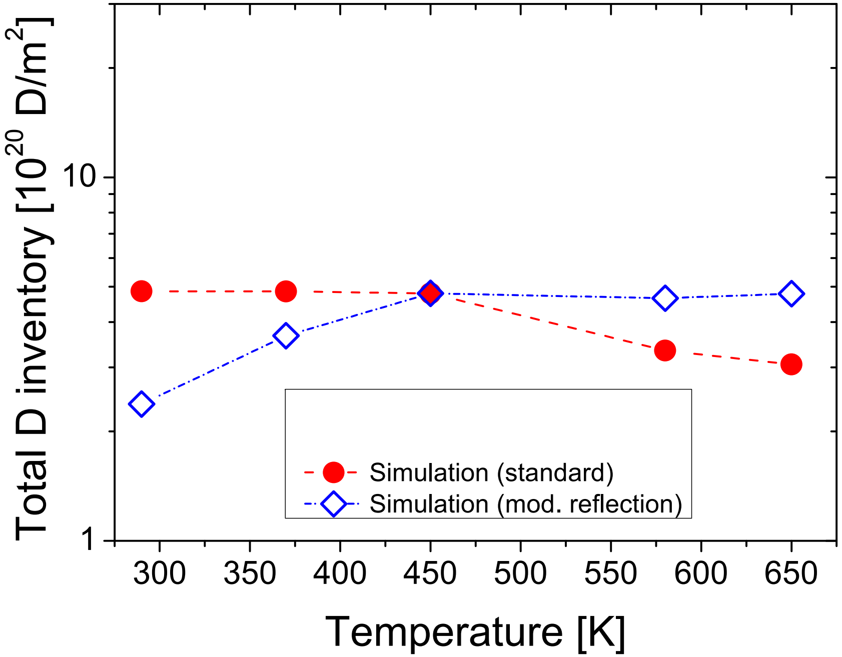

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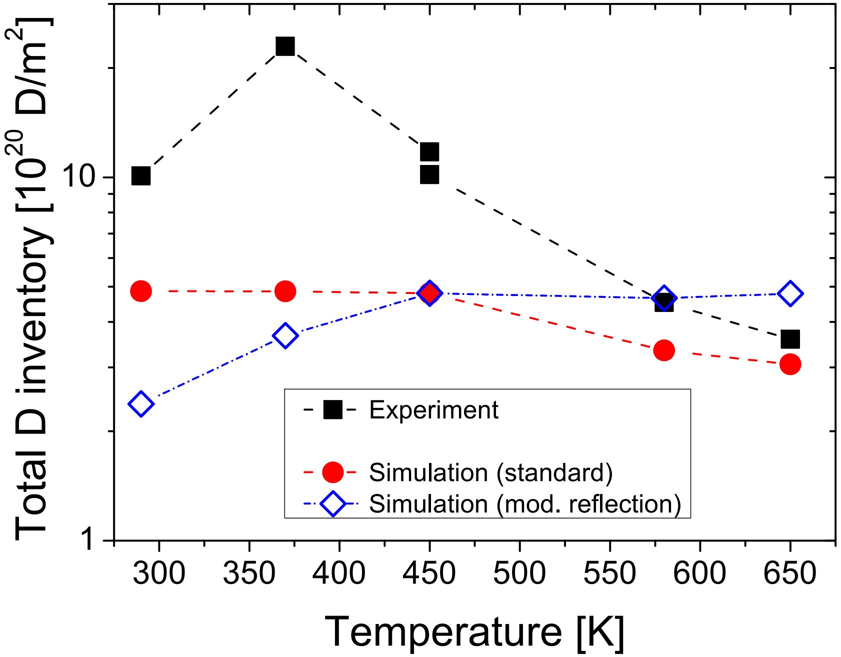

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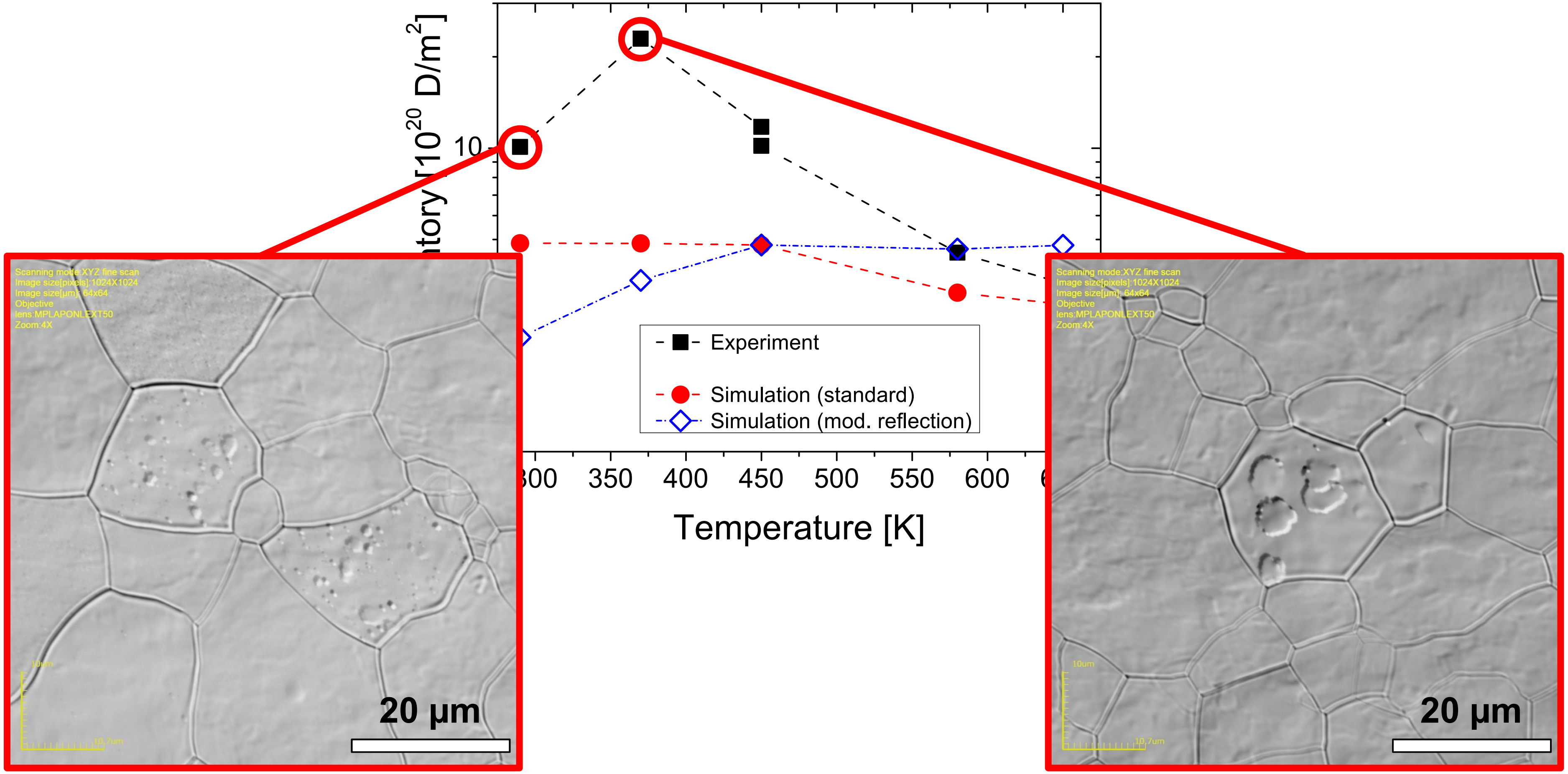

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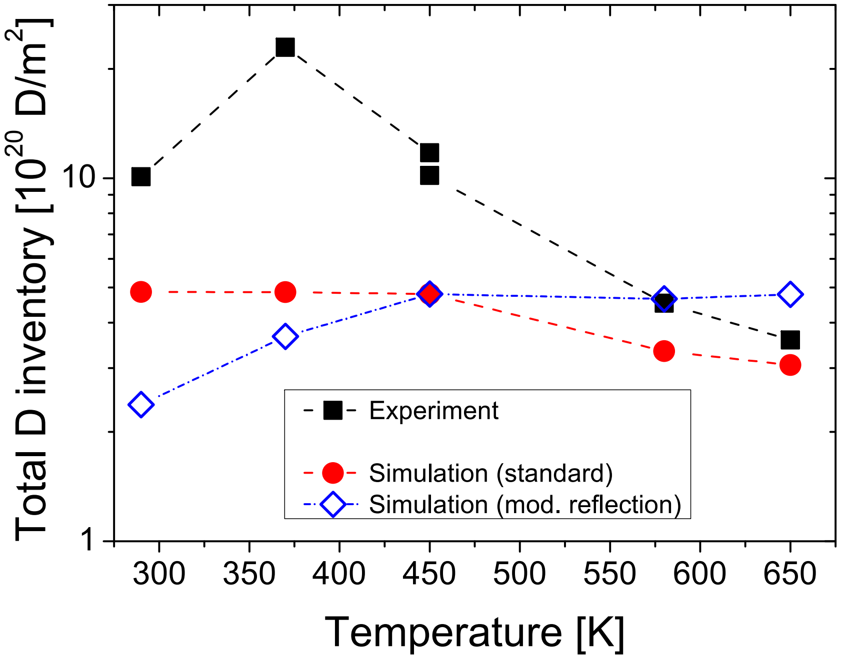

- Samples **modified during plasma** exposure
  - ➢ Clearly visible **protrusions (“blisters”)** at 290 and 370 K
  - ➢ **No self-consistent model** for blistering currently available
  - ➢ **Not suitable** for elucidation of temperature dependence at floating potential

Mechanisms for Temperature Dependence

Possible Mechanisms

- **Temperature dependence** of **D uptake** observed
  - E.g., **here** and at **Pilot-PSI**
  - Effective D uptake of implanted D **smaller at lower temperatures**

[7] P.W. Tamm, L.D. Schmid, J. Chem. Phys. **54** (1971) 4775

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  - ➢ Absorption of **atomic D** particularly at **higher temperatures**
  - ➢ **Enhanced desorption** at **high surface coverage** (i.e., low temperature):
    - ↳ But: **surface** already assumed to be **perfect sink** for H/D (c = 0 at x = 0!)
    - ↳ Relevant if actual reflection coefficient is much higher than assumed here

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- Idea: **transport effect?**
  - Expected: $\Gamma_{\mathbf{surface}}$ **/** $\Gamma_{\mathbf{bulk}}$ **=** $\mathbf{r}_{\mathbf{implantation}}$ **/** $\mathbf{r}_{\mathbf{diffusion}}$ (independent of temperature)
  - How can the **branching ratio** of **transport** towards surface and bulk **change?**
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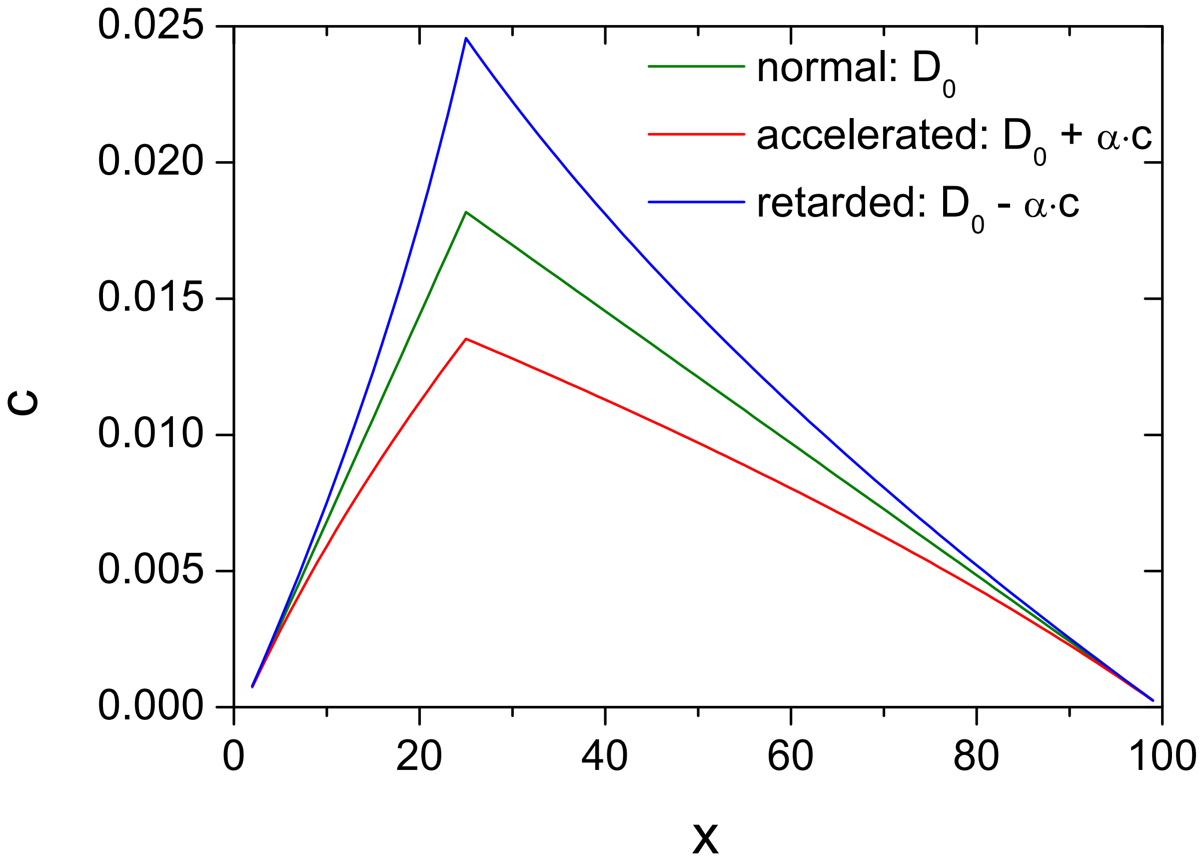

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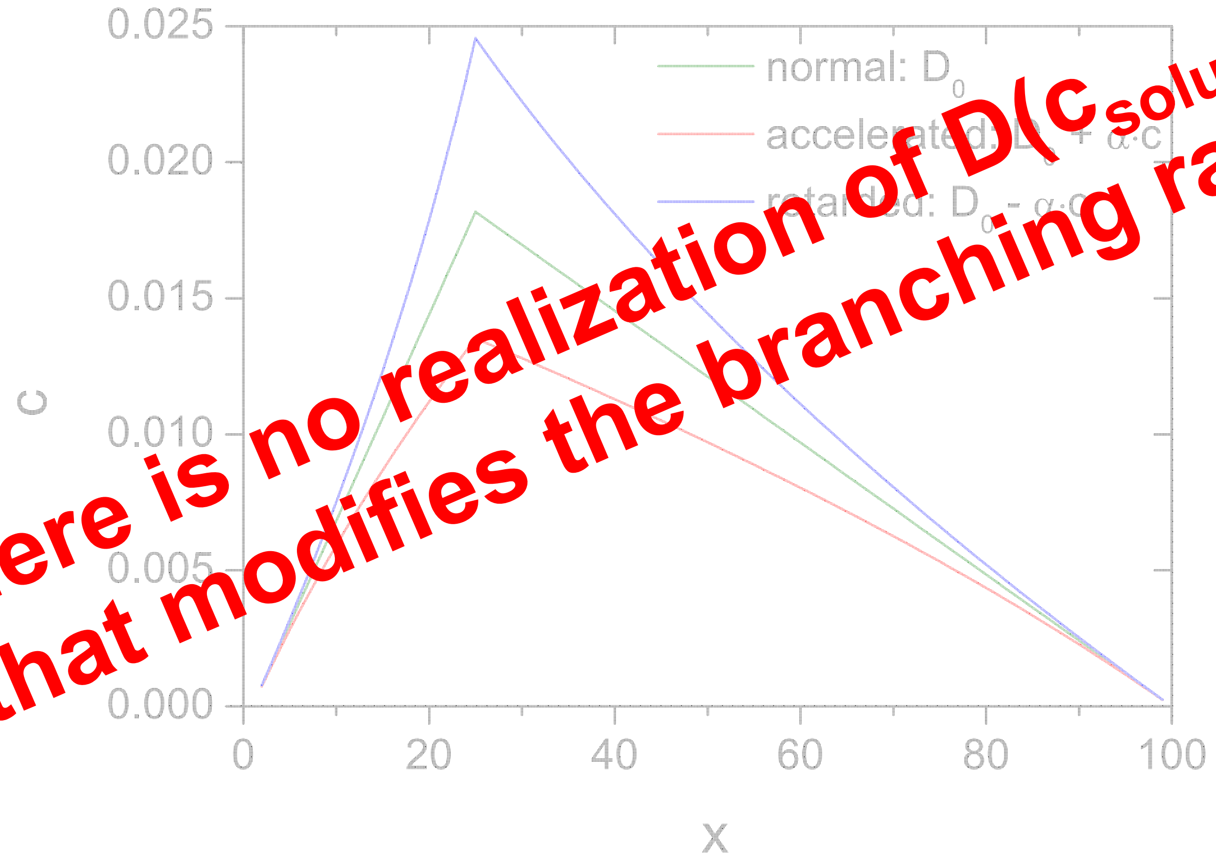

There is no realization of D(c_{solute},T) that modifies the branching ratio!

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[8] C. J. Penington *et al.*, Phys. Rev. E **84** (2011) 041120

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- If diffusion equation becomes **non-local**, e.g., $D(T) \rightarrow D(c_{solute},\nabla c_{solute},T)$
  - Able to **change branching ratio**
  - **H-H interaction** (exception: not simple volume exclusion)
  - Example: **transport** depends on **stress** induced by **H concentration** (e.g., [9])

[8] C. J. Penington *et al.*, Phys. Rev. E **84** (2011) 041120
[9] O.V. Ogorodnikova *et al.*, J. Appl. Phys. **103** (2008) 034902

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- **Temperature variation** (290 – 650 K) at **floating potential**
  - ➢ Experiment and simulation show **similar range of values**, but **opposite trend**
  - ➢ Reconciled by artificially introducing **temperature-dependent effective D flux**
  - ➢ Activation energy of **~0.1 eV**
  - ➢ Results for **-100 V** bias **difficult to interpret** because of **blistering**

Summary

- **Diffusion-trapping model** with **all parameters fixed** by experiments or literature
- Compared to **experiments** in **low-flux** ECR plasma
- **Fluence variation at 450 K**
  - ➢ Close **agreement** of **simulation** and **experiment** at **floating potential**
  - ➢ **Agreement** also for sample **bias of -100 V**
    - ⇘ **Biasing reduces filling time** of damaged zone by **factor ~6**
- **Temperature variation** (290 – 650 K) at **floating potential**
  - ➢ Experiment and simulation show **similar range of values**, but **opposite trend**
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➢ **Possible mechanisms** for observed temperature dependence of D uptake
- ➢ **Surface** effect
  - ⇘ E.g., enhanced **reflection** or **re-emission,** or **D^0 atom uptake**
- ➢ **Non-local diffusion law:** e.g., **H-H interaction** leading to **$D(c_{solute},\nabla c_{solute},T)$**