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Quantitative characteristics of H and He interaction with radiation defects in tungsten

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Outline

- Introduction
- Theory of TDS: Detrapping energy determination
- Deuterium
  - Damage by keV ion beam – experiments
  - Comparison to damage by MeV ions and neutrons
- Helium
  - MeV ion beam (with IPP) – preliminary results
- Summary

Introduction

H, He solubility in W is very low. However, the low solubility correlates usually with the high binding energy with radiation defects.

- H&He accumulation in W is very sensitive to presence of defects.
- One need characteristics of H/He-trap interaction to predict the behavior of these species in W correctly.

Progress in determination of quantitative characteristics of H/He-trap interaction using TDS will be discussed.

Potential energy diagram: H in W

H atom in vacancy and in vacancy clusters is placed not in the center, but close to close to the surface.

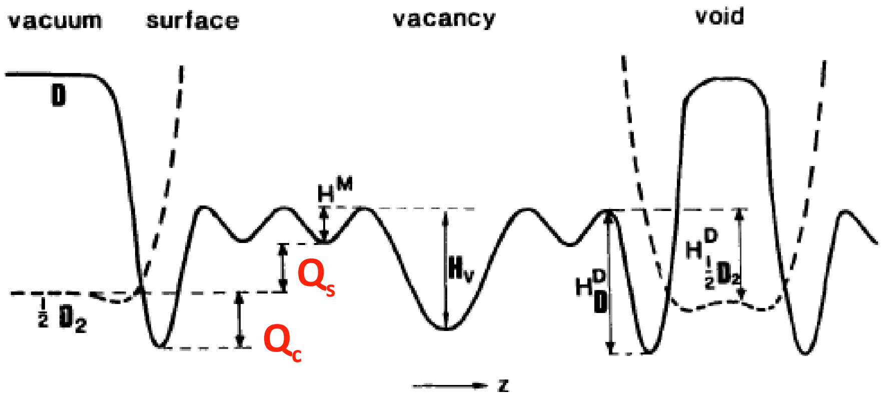
Therefore, simple estimations of binding energies can be made using a simplified “adsorption model”

$$E_b^{VC} = (Q_c + Q_s)$$

$$E_b^{V} = \alpha \cdot (Q_c + Q_s), \quad \alpha \approx 0.5?$$

O.V. Ogorodnikova, 118, 074902 (2015)
A. E. Gorodetsky, et al.,JNM, 93–94, 588 (1980)

H. Eleveld, A. van Veen. JNM, 191-194 (1992) 433



If one takes Q_s = 1.04 eV (Frauenfeder value) and $Q_c \approx$ 1 eV, one will get E^V= 1.02 eV and E^{VC}= 2.04 eV.

Both value are in reasonable agreement with the most of data derived from experiments.

→ Both Q_S and E_{bV} and E_{bVC} are reasonable!

TDS theory

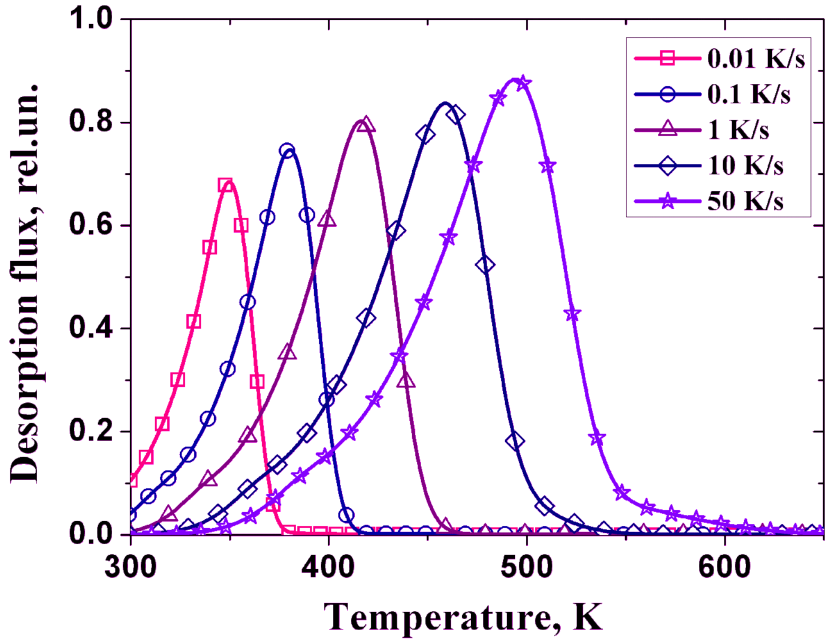
It is difficult to derive precise numbers for the binding energy experimentaly.

•Fitting of TDS spectra is always challenging and often **possible by various sets of parameters.**

•In particular cases, the detrapping energy can be **directly determined from the shift of TDS maximum** in a series of identical experiments performed with different heating rates.

This method was originally used for determination of desorption barriers at the surface.

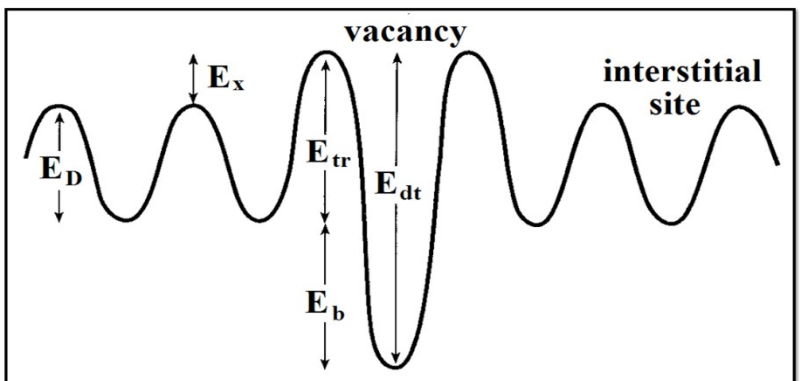
Release of D atoms from the bulk to vacuum is **a multi-step process**, including detrapping, diffusion, re-trapping, desorption. **However, the same procedure can be applied in the case of the high recombination rate!**



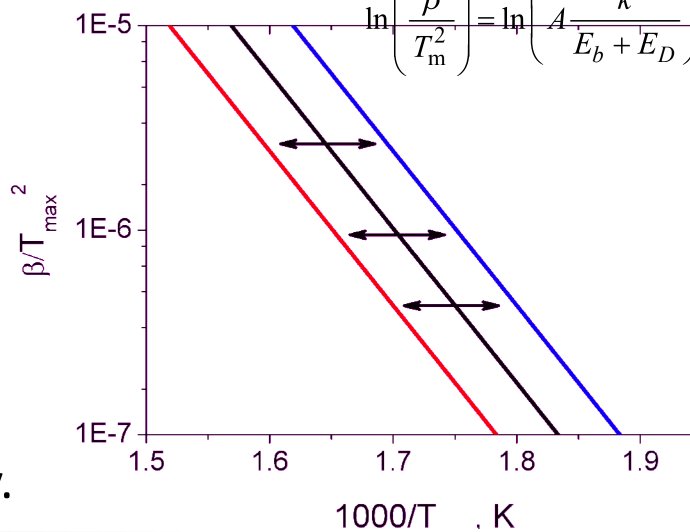
TDS theory: re-trapping effect

- If the **trap concentration is small**, re-trapping is negligble, and situation is close to one-step situation and de-trapping energy ***E_{dt}*** can be determined from experiment.
- If the **trap concentration is high**, than one can show that the shift of the peak corresponds to ***E_b+E_d***. (see M.Zibrov et al., JNM, 477 (2016) 292)

  Here, **the slope doesn't depend on other parameters**, but the peak position depends on diffusivity, characteristic frequencies, and trap distribution.



$$\ln\left(\frac{\beta}{T_m^2}\right)=\ln\left(A\frac{k}{E_b+E_D}\right)-\frac{E_b+E_D}{k}\frac{1}{T_m}$$

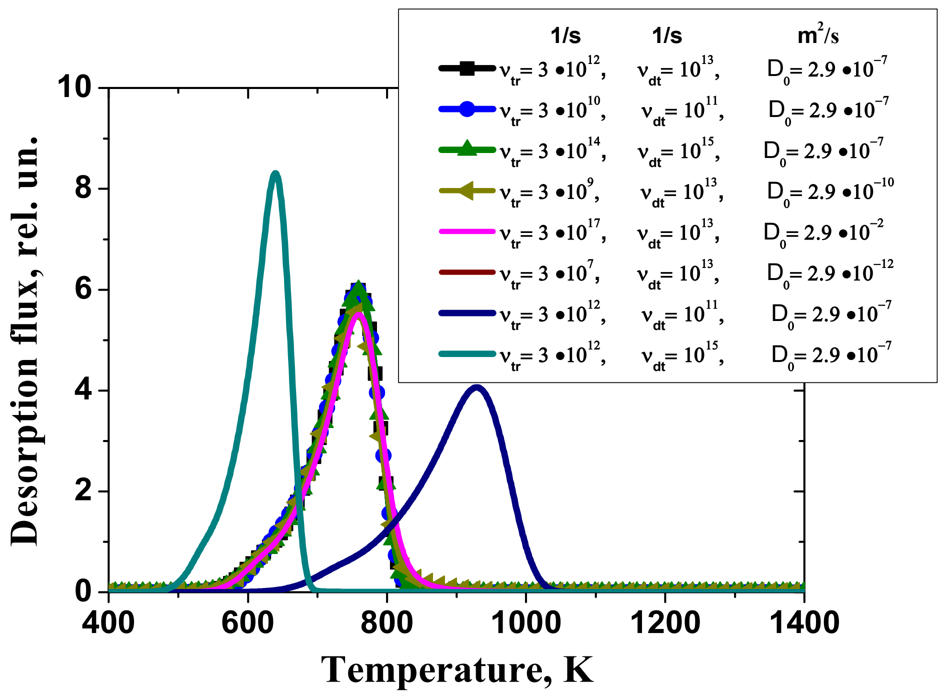


We will assume later ***E_{dt} = E_b+ E_D*** for simplicity.

TDS theory: characteristic frequencies

It is difficult to derive characteristic frequencies by this method.
Even if you know the depth profile of traps and H inside these traps, you still have freedom to choose a combination of D_0, ν_{tr}, ν_{dt}.

There is no shift of the peak, if you keep constant : $$D_0 \frac{\nu_{dt}}{\nu_{tr}} = const$$



Experimental facilities: MEDION

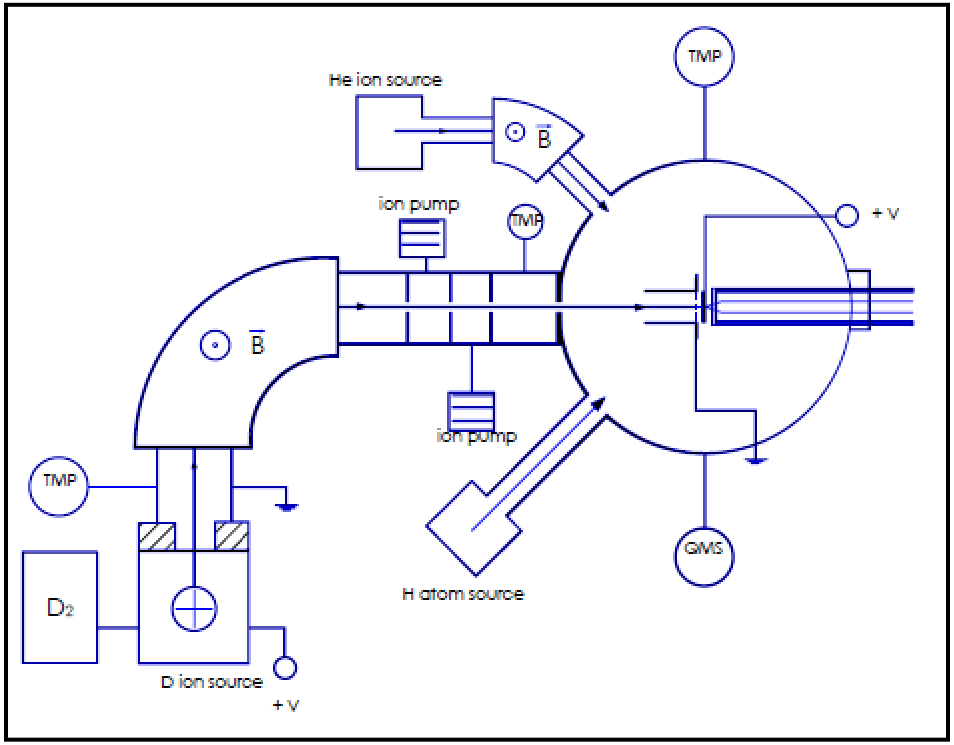
Main advantages:

- **All stages of the expeiment (annealing, irradiation, TDS)** can be done in one chamber **without air exposure.**
- Deuterium desorption from the sample can be **monitored during all stages**.
- Using one sample in a series of experiments, **the shift of the peak position in TDS can be measured precisely**.

Last upgrade: New chamber with water cooled walls designed for multibeam experiment.

The second beam line was tested and will be installed soon.

MEDION



Residual pressure: < 10^{-8} mbar

Energy of ions: 1-20 keV

Incident flux for D: 10^{17}-10^{18} D/m^2s

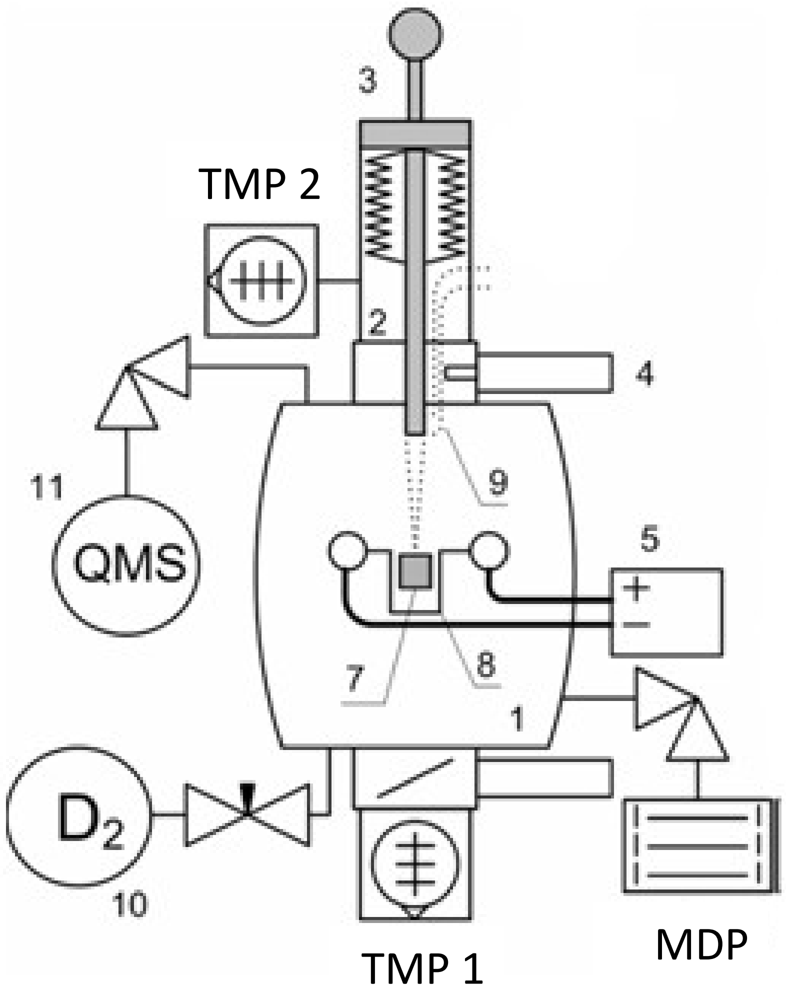
Experimental facilities: TDS stand

The UHV TDS stand is used for analysys of the samples exposed outside the Medion facility.

The exchange chamber allows to perform TDS measurements quickly with a reasonble background.

For experiments with He, the heater in the TDS stand was optimized to **reach 2500 K** for small W samples.

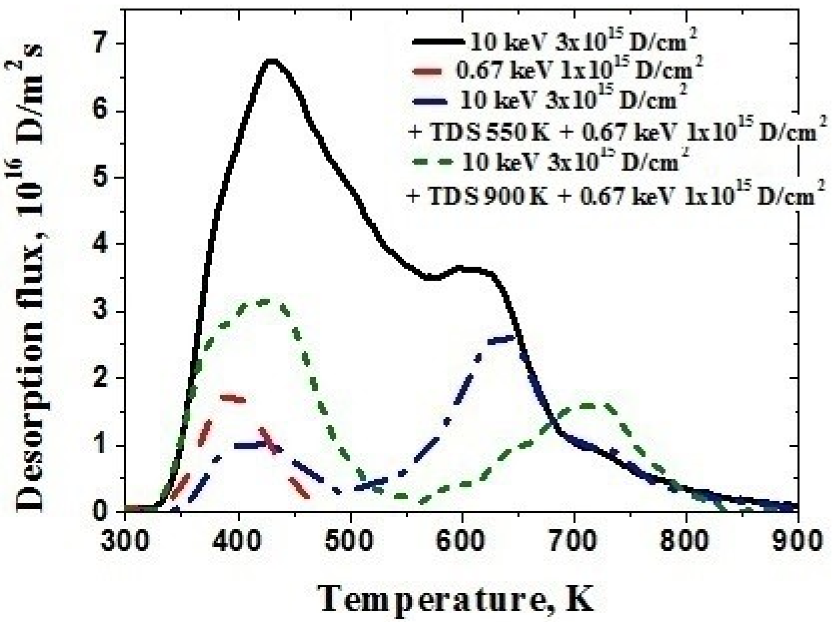
New QMS allows separation of ^{4}He and D_2 signals.



keV ion damage: experimental details

M.Zibrov et al., Physics procedia, 71 (2015) 83

Sample: 25 µm polycrystalline W foil annealed at 1800 K.



Experimental procedure:

1.Damaging the sample by irradiation with **10 keV D^+** ions to the fluence of **3×10^{19} D/m^2**

2.Annealing at 550 K or 900 K for 5 min

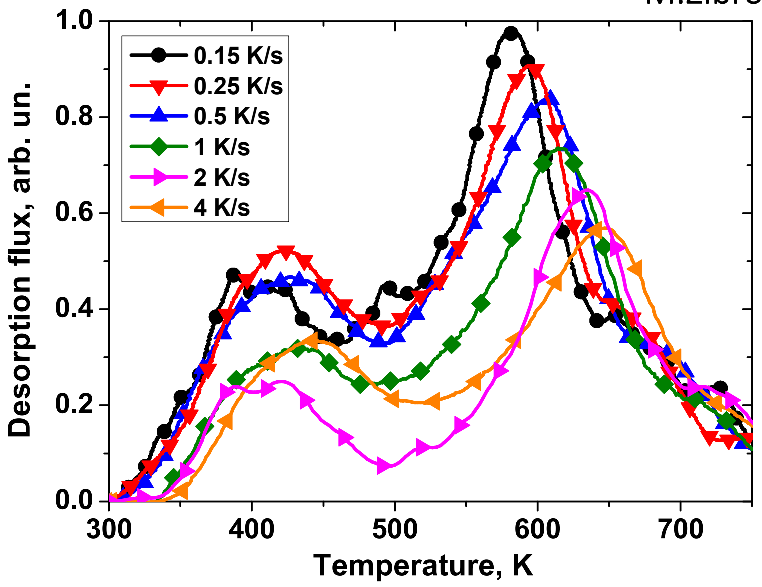
3.Implantation by a **2 keV D_3^+ (0.67 keV/D)** ion beam to the fluence of **1×10^{19} D/m^2**

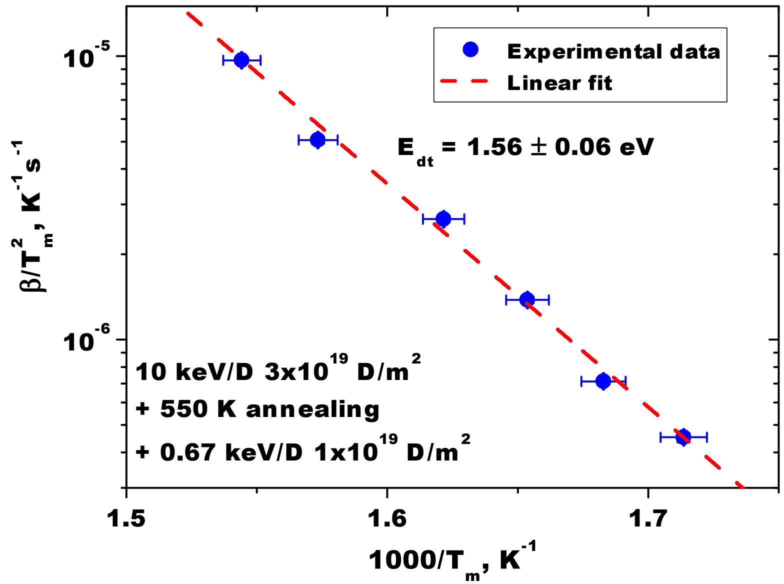
4.TDS measurements were performed with heating rates in the range of **0.15-4 K/s**

Low fluences were used at the step 3 to avoid the effect multiple trapping.

Single vacancies

M.Zibrov et al., JNM, 477 (2016) 292



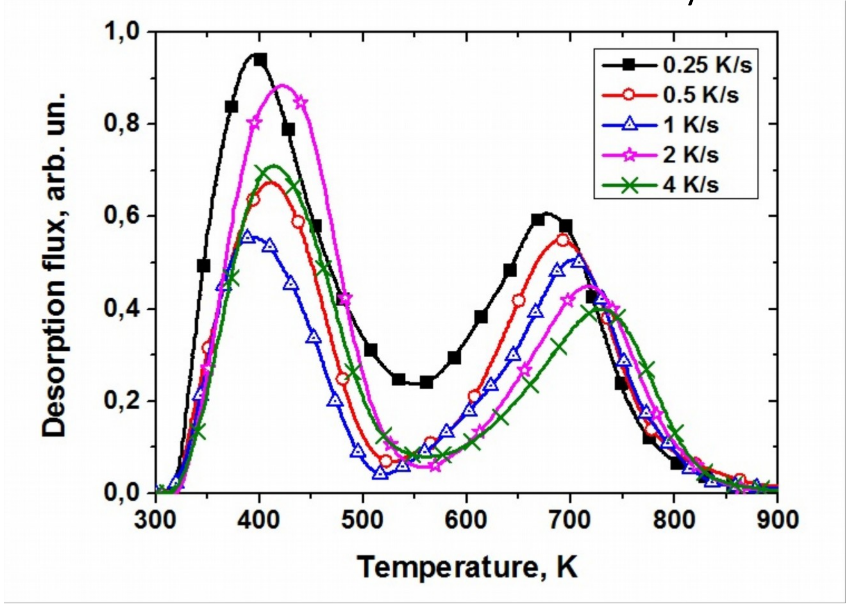


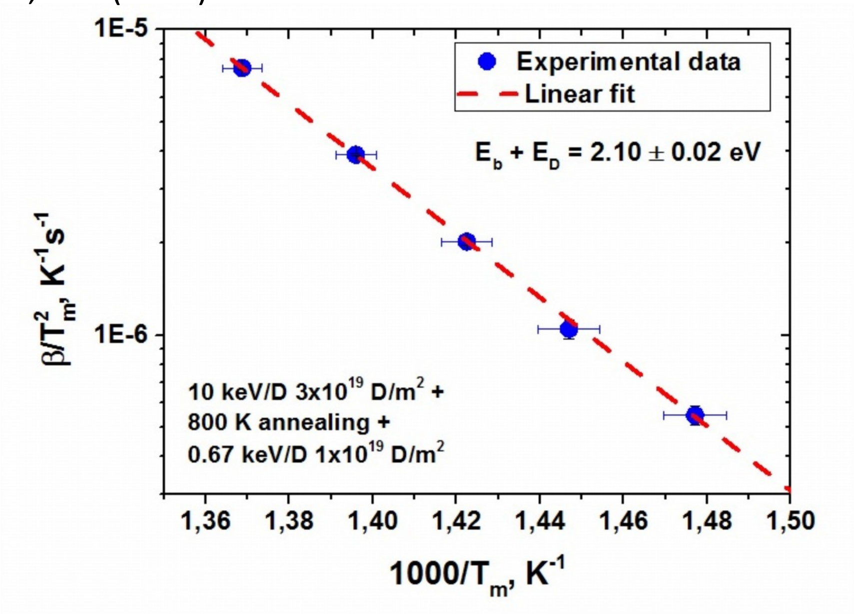
10 keV/D, 3×10^{19} D/m^2 + Annealing at 550 K + 0.67 keV/D, 1×10^{19} D/m^2 (point defects)

The D detrapping energy from vacancies in W: ***E_{dt}*** **= 1.56±0.06 eV**

Vacancy clusters

S.Ryabtsev et al., NIMB, 382 (2016) 101





10 keV/D, 3×10^{19} D/m^2 + Annealing at 900 K + 0.67 keV/D, 1×10^{19} D/m^2 (point defects)

The D detrapping energy from vacancy clusters in W: ***E_{dt}* = 2.10±0.02 eV**

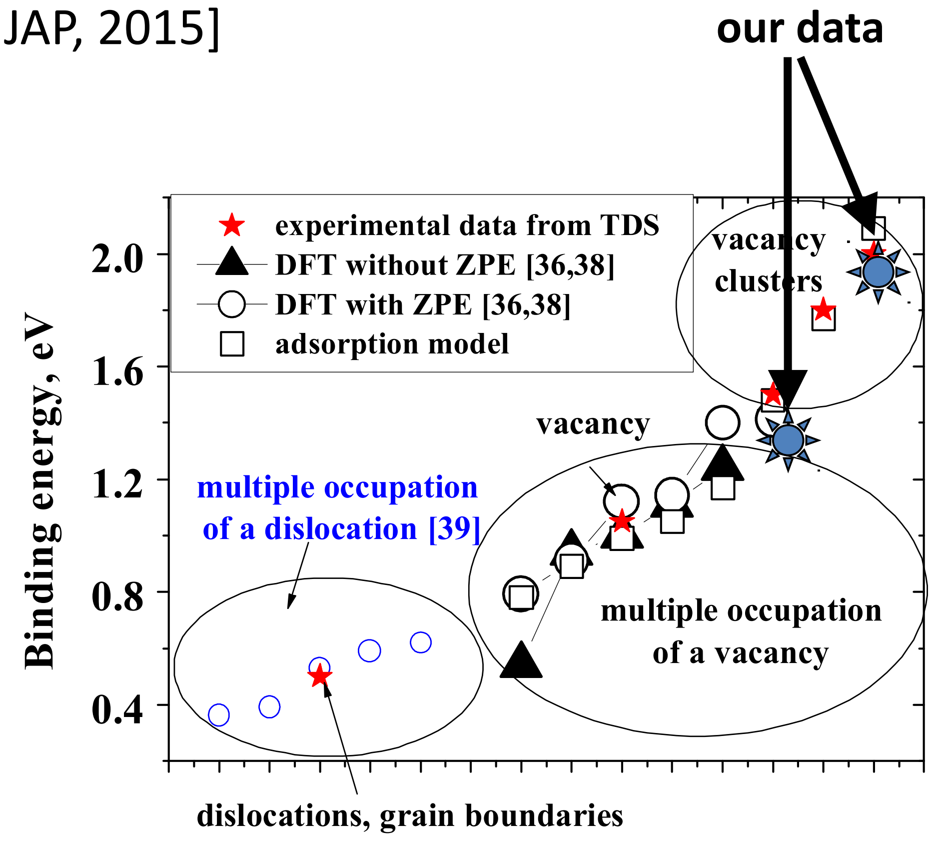
Comparison to other data

The data are compared with DFT calculations and experimental results collected in [O.V. Ogorodnikova, JAP, 2015]

Our data for a single vacnacy are slightly higher than most of other experimental data, but in a good agreement with various DFT calculations (first atom in the trap).

The diffusion barrier of E_D =0.2 eV was used for more correct comparison with DFT.

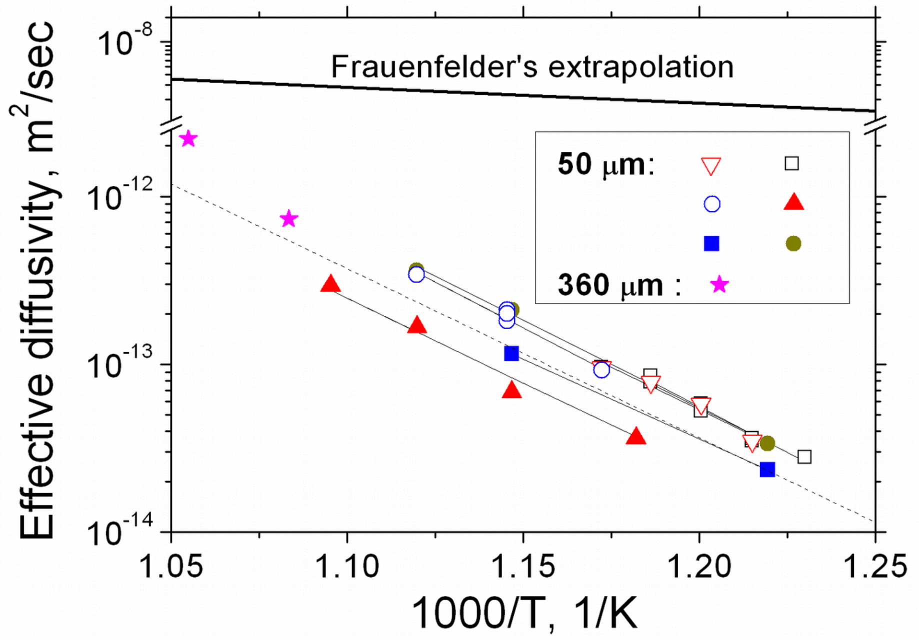
Low fluences were used in our experiments to avoid multiple trapping effect!

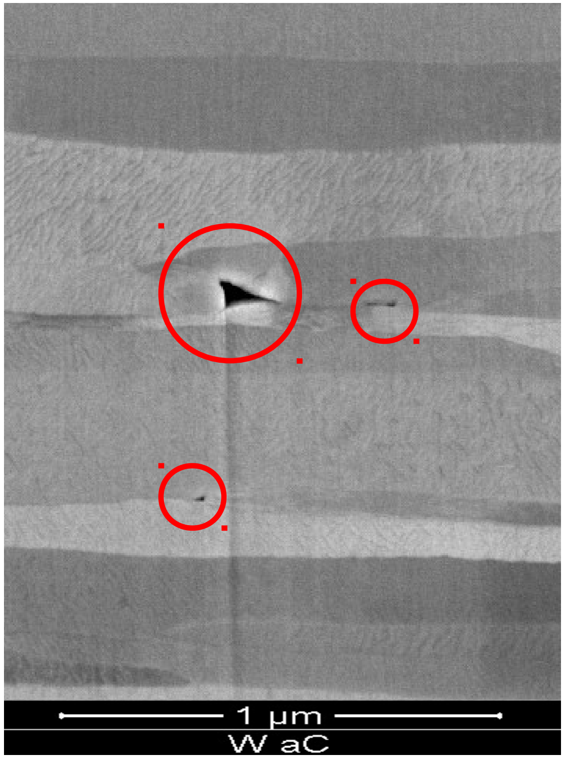


O.V. Ogorodnikova, JAP, 2015

Ion-driven permeation experiments

Yu.Gasparyan et al., JNM, 417 (2011) 54

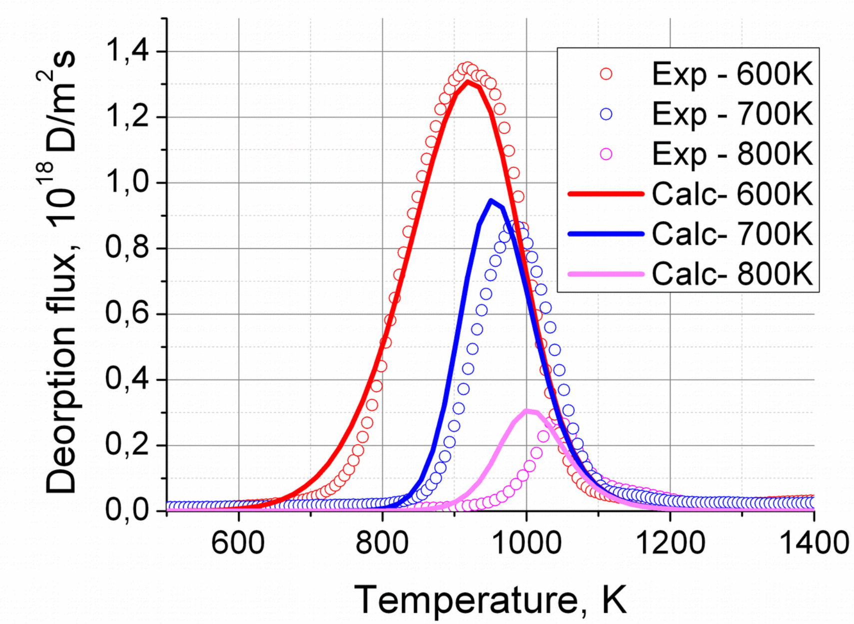




- Experimental points for each single sample can be fitted linearly with the activation energy of (2.05±0.15) eV
- One can conclude that the detrapping energy of traps that have major influence in these conditions **E_{dt} = (2.05±0.15) eV**

20 MeV W: E_{dt} estimations

- A series of D atom exposures at various temperatures were analyzed.
- A significant D retention was observed even after exposure at 800 K.
- The detrapping energy for high energy traps was estimated to be **E_{dt}=1.7–2.0 eV**.
- Similar results was also obtained for n-damaged samples in [Hatano et al.,438 (2013)S114]: **E_{dt} = 1.8 eV**. (New data from Shimada et al. - up to 2.6 eV?)

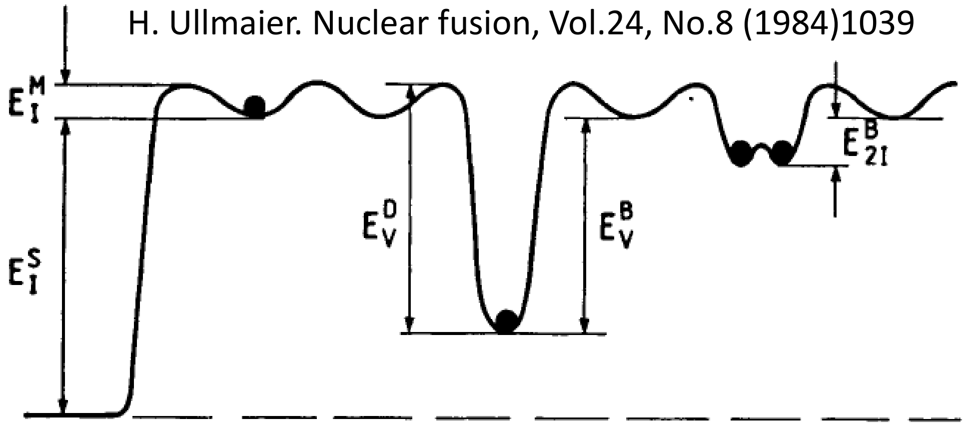


Yu.Gasparyan et al., JNM, 2015.

In spite of possible significant differences in defect structure produced by keV ions, MeV ions and neutrons, vacancy clusters with similar energies seems to play the major role in trapping of hydrogen isotopes at elevated temperatures.

He in W

H. Ullmaier. Nuclear fusion, Vol.24, No.8 (1984)1039



The heat of solution is very high (~5.5 eV ?).

Only energetic particles can enter the bulk (this can explain, for example, the threshold for fuzz formation)

Again, a high binding energy with radiation traps should be expected.

	Peak	Calculated energies relative to the perfect lattice	Theoretical binding energy, eV	Experiment (Kornelsen)
α-band	H	He V → He + V 4.40 → 5.44 + 3.35	4.39	4.05
	$^*I^a$	$He_2V_2{}^I$ → He V_2^{II} + He 8.38 → 7.36 + 5.44	4.42	4.37
	$^*H'^a$	He $(VKr)^I$ → He + $(VKr)^I$ 13.72 → 5.44 + 12.68	4.40	3.84
β-band	G	He_2V → He + He V 6.95 → 5.44 + 4.40	2.89	3.14
	F	He_3V → He + He_2V 9.87 → 5.44 + 6.95	2.52	2.88
	E	He_4V → He + He_3V 12.81 → 5.44 + 9.87	2.50	2.41

He retention in W

Helium retention and release in W was investigated intensively in 70s-80s. He implanted in W with keV energies released at about 1200-1800 K.

There are, however, some results have been observed recently in experiments, which are not well understood:

- He did not release after annealing at 2000 K from tungsten damaged by 20 MeV W ions (E.Markina, Phys. Scr. (2014) 014045)
- Low temperature release from "W fuzz" after exposure at very high temperatures (Yu. Gasparyan, NF, 56 (2016) 054002) - in contrast to hydrogen desorption always above irradiation temperature

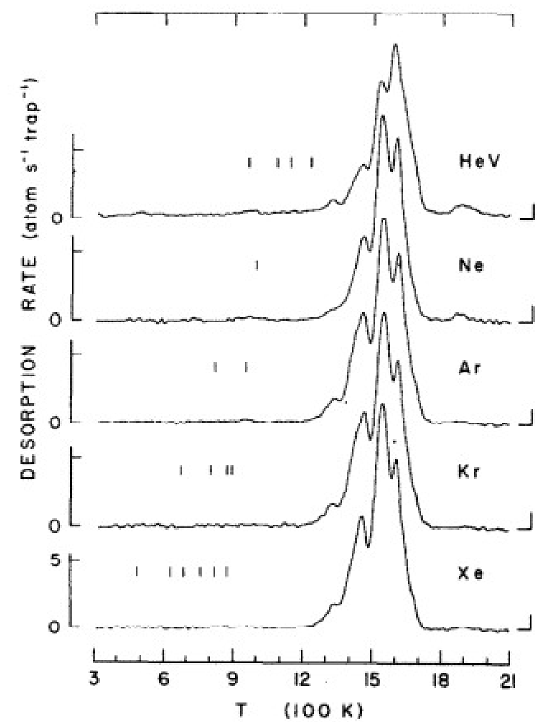


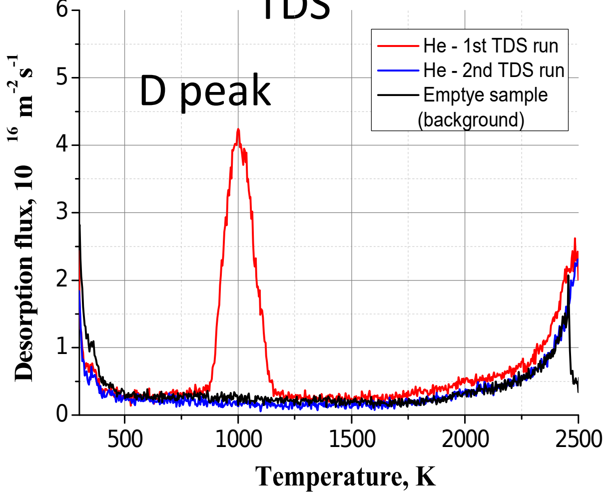
Fig. 6. As for fig. 1, but with $n^+ = 1.2 \times 10^{14}/\mathrm{cm}^2, \bar{i} \approx 62$.

E.V. Kornelsen et al., JNM, 92(1980)79

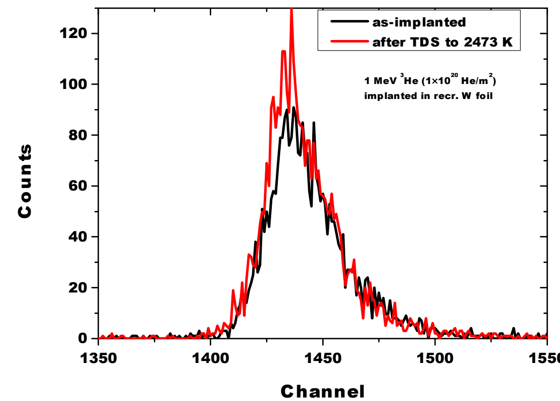
^{3}He release from MeV damage (in collaboration with IPP, Garching)

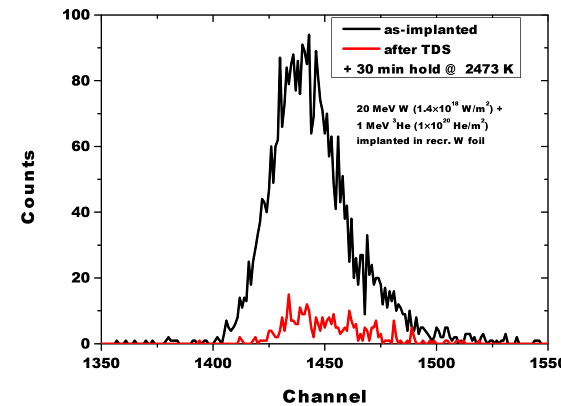
50 µm W foil
annealed at 2000 K
damaged by 20 MeV W^+
1 MeV ^{3}He implantation,
fluence - 10^{20} He/m^2
Preliminary NRA analisys
by D MeV ions

TDS



NRA





- D release from NRA can be perfectly observed and in a perfect agreement with the irradiation fluence.
- **He signal in TDS is small** in spite of 10 times higher fluence. (Upper limit - **3 % of implanted He**)
- 30 minutes annealing at 2470 K led to removal of **~90 % of implanted He** (NRA data).
- **He stay in W well above the re-crystallization temperature.** He can influence strongly annealing of defects and recrystallization process.

^{4}He release from MeV damage (in collaboration with IPP, Garching)

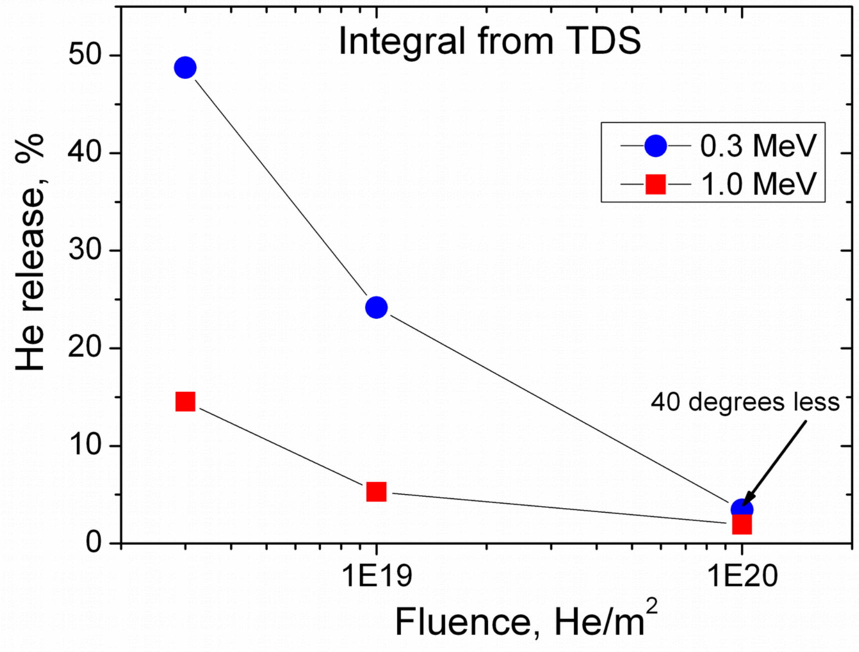
New series of experiments have been done recently:

- 0.3 MeV He irradiation, 10^{18}-10^{20}He/m^2
- 1 MeV He irradiation, 10^{18}-10^{20}He/m^2

TDS was done up to 2470 K, and we did not reach the peak position in all experiments.

Results:

- Clear shift of the He release to lower temperatures, decreasing the incident energy and the fluence
- This is explained by reduction of the trap concentration and the depth.



Summary

- Possible methods for quantitative investigation of H/He behaviour in PFMs were demonstrated.
- Detrapping energies of D from single vacancies and vacancy clusters were deduced from series of TDS experiments.
- Similar detrapping energies were deduced from ion-driven permeation experiments and for D desorption from self-damaged W. One should not expect seriously higher detrapping energy for neutron damage samples.
- He can stay in damaged W above 2500 K. There is no way to remove it by heating of the wall. This influence also annealing of radiation damage.
- A part of He can release at very low temperatures, below irradiation temperature. Mechanisms are not clear.

Thank you for your attention!