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Determination of binding energies for hydrogen with radiation defects in tungsten by means of TDS: theory and experimental data

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Outline

- Introduction
- Theory of TDS: Binding energy determination
- Experiments
  - Damage by keV ion beam – experiments
  - Damage by MeV electrons
  - Damage by MeV ion beam (with IPP)
- Multiple site trapping
- Final remarks and summary

Introduction

- H solubility in W is very low. Therefore, H accumulation in the bulk of W is very sensitive to presence of defects.
- A lot of TDS experiments have been performed for W, but there are much less data on characteristics of traps in W.
- Experimental data even for detrapping energy from point defects scatter in the range of 1.3-1.6 eV [1-3].

[1] H. Eleveld, A. van Veen, J. Nucl. Mater. 191–194, 433 (1992)
[2] O.V. Ogorodnikova, J. Roth, M. Mayer, J. Appl. Phys. 103, 034902 (2008)
[3] M. Poon, A.A. Haasz, J.W. Davis, J. Nucl. Mater 374, 390 (2008)

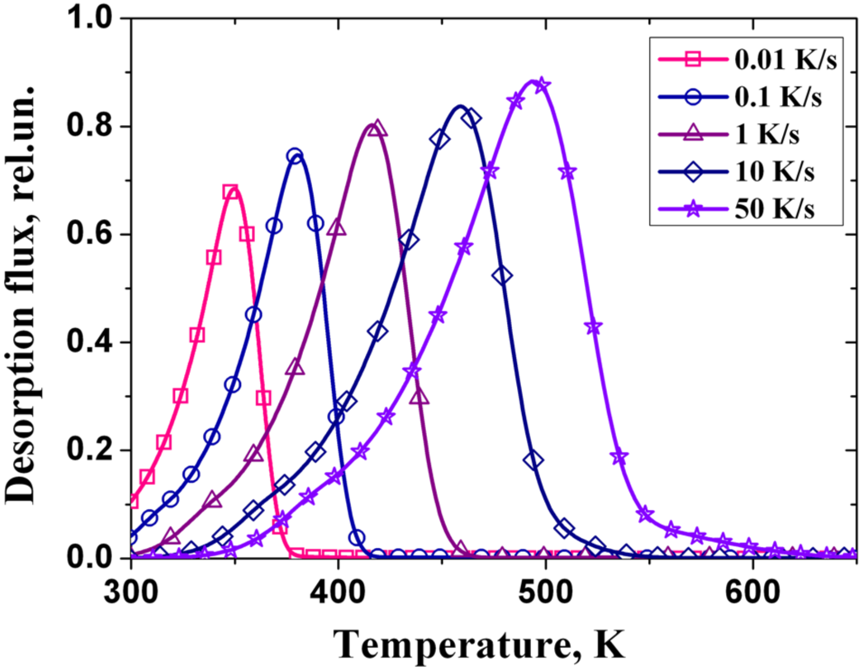
Analysis of TDS applicability for determination of trap characteristics has been performed.
Experimental data on D retention in various radiation defects and status of ongoing works are also presented.

TDS theory

- 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 measurements performed with different heating rates.

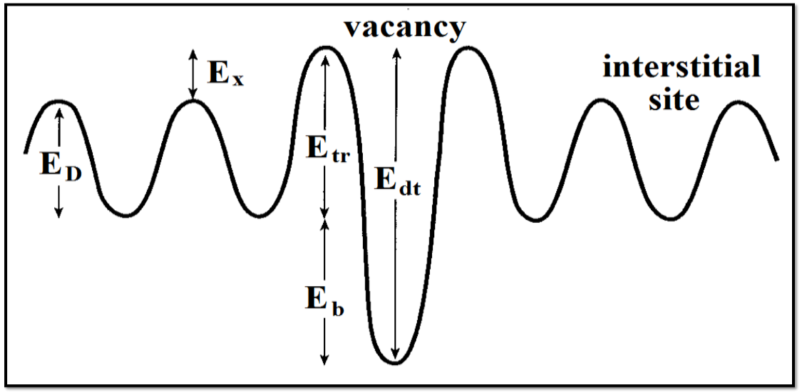
This method was originally used for determination of desorption barriers at the surface [1].

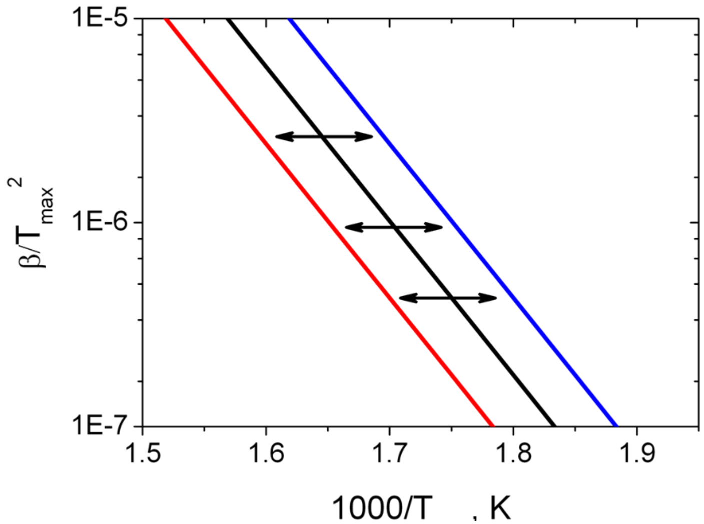
Release of D atoms from the bulk to vacuum is **a multi-step process**, including detrapping, difusion, re-trapping, desorption.



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 $\boldsymbol{E_{dt}}$ can be determined from experiment.
- If the **trap concentration is high**, than one can show that the shift of the peak corresponds to $\boldsymbol{E_b+E_d}$.
- In the last case the slope do not depend on many other parameters, but the peak position depends on diffusivity, characteristic frequencies, and trap distribution and parameters.





Is there theoretical data on E_x?

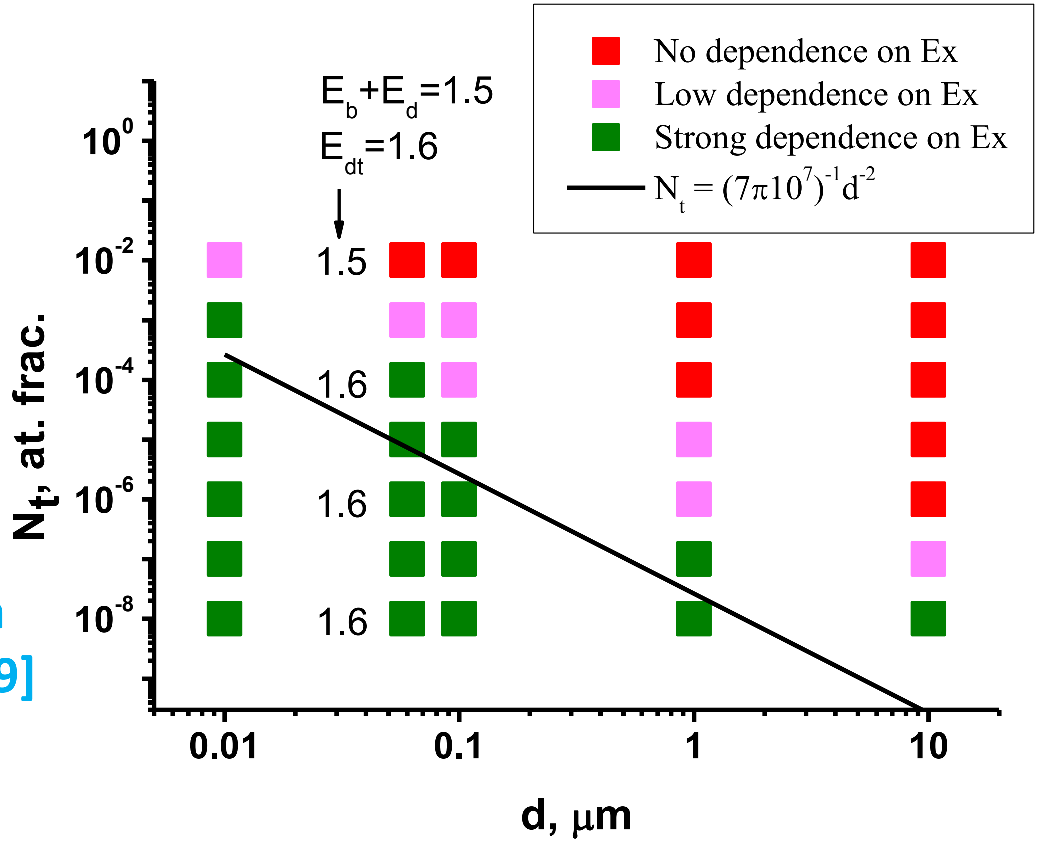
Re-trapping: TMAP calculations

In the wide range of parameters the energy related to traps can be determined from experiment, but this can be **different for various concentration of traps**!

For small trap concentration (no re-trapping)
$A \times N_t \times d^2 << 1 \Rightarrow E_{dt}$

For high trap concentration,
$A \times N_t \times d^2 >> 1 \Rightarrow E_b + E_d$

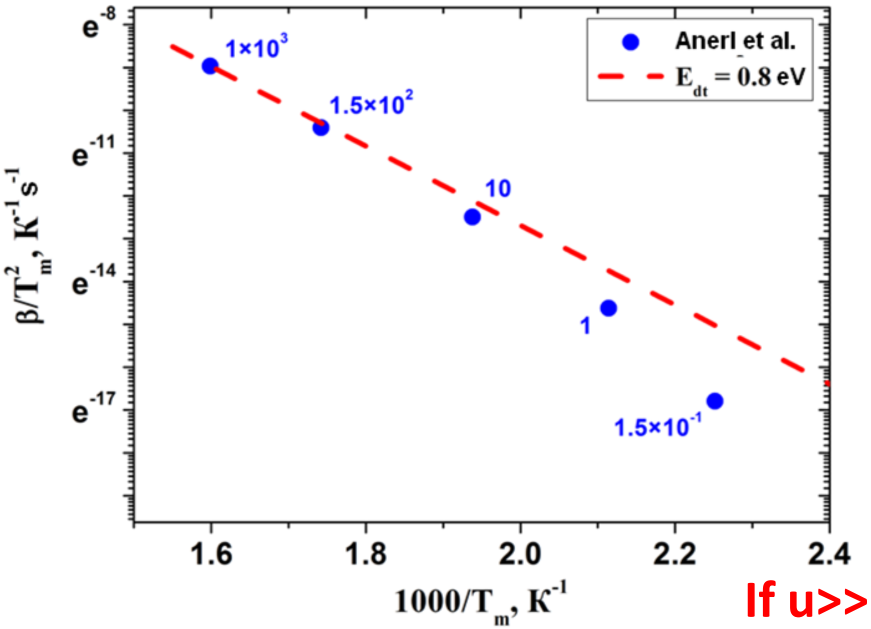
d – width of damaged zone
A = const = $(7\pi 10^7)^{-1}$ in [A.A. Van Gorkum, Vacuum, 1981,v.31,p.89]



TDS theory: Influence of surface

All formulas above are correct only in the case of fast recombination at the surface!

M.Zibrov et al., PAS&T, ser. Thermonuclear Fusion, 38 No. 1 (2015) 32-41 (in Russian).



The criteria of method's applicability is proposed:

$$u = \frac{K_r(T_m)C(T_m)l}{D(T_m)}$$

$$u = \frac{\rho M l}{2D_0} K_r(T_m)\exp\left(-\frac{(E_{dt} - 2E_D)}{kT_m}\right)$$

ρ – concentration of atoms, K_r- recombination coefficient, T_m – temperature of the maximal desorption, l – implantation depth, M – population of traps

If u>>1 than the method can be applied successfully!

- The influence of the surface is highest for low binding energies.
- One should pay attention to surface conditions in this kind of experiments!

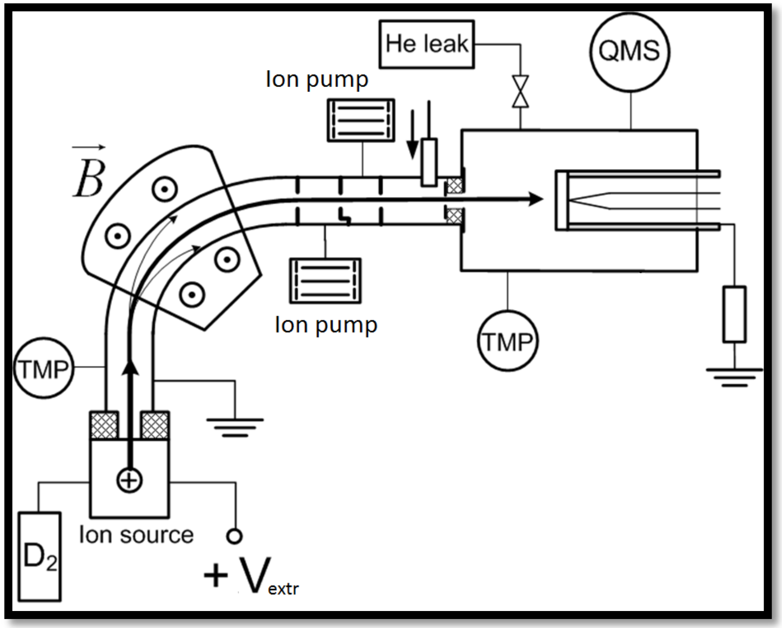
keV ion damage: experimental details

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

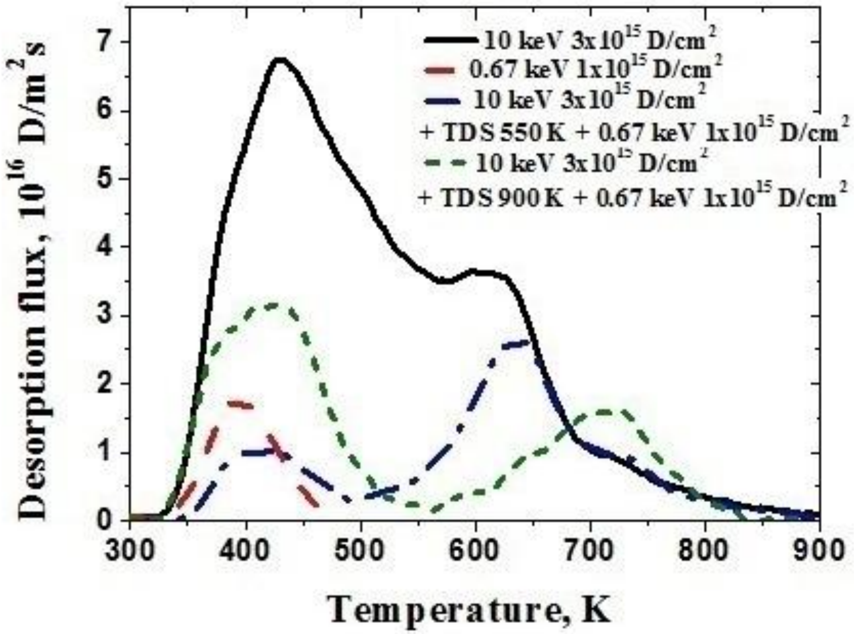
Experimental procedure:

1. **Damaging** the sample by irradiation with **10 keV D^+** ions to the fluence of **3×10^{19} D/m²**
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/m²**
4. **TDS measurements** were performed with heating rates in the range of **0.15-4 K/s**

MEDION Facility



keV ion damage: peak isolation

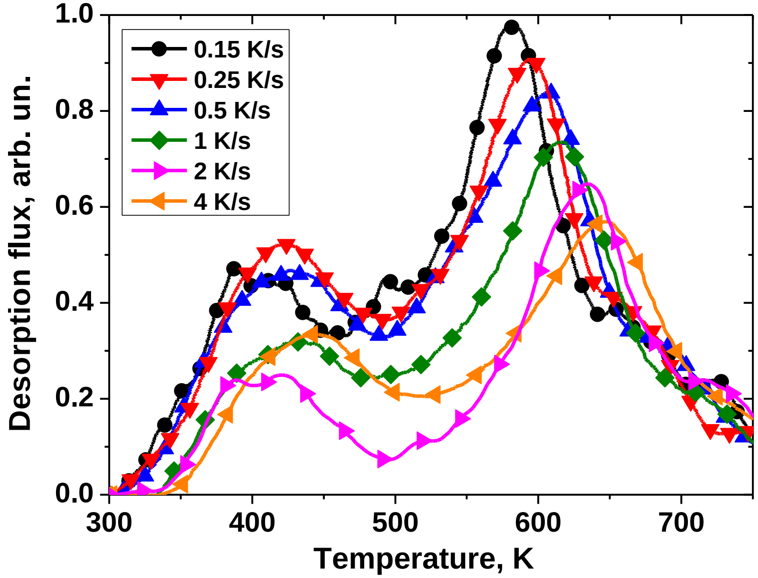


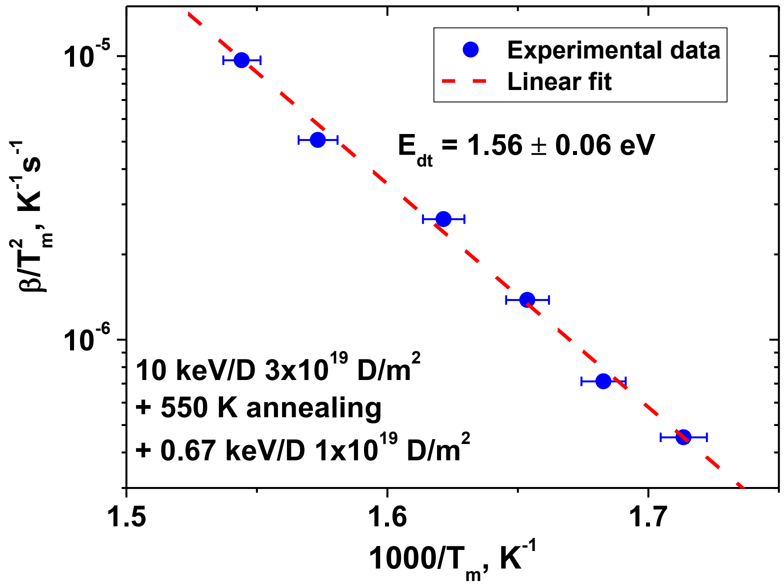
M.Zibrov et al., Physics procedia, in print.

1. Implantation without damaging: a single peak at 400 K
2. Damaging without annealing: not well resolved peak at 650 K
3. Annealing at 550 K allows to achieve a good resolution of peak at 650 K, attributed to desorption from single vacancies
4. Annealing at 900 K: well resolved peak at 720 K, corresponding to release from vacancy clusters (vacancies start to agglomerate at ~650 K [Eleveld, 1994; Debelle, 2008]

Experiment at MEDION (kev ions)

M.Zibrov et al., JNM, to be submitted.





10 keV/D, 3×10^{19} D/m^2 + TDS 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

The D detrapping energy from vacancy clusters in W: E_{dt} = 1,96±0,09 eV

These values are close or lie within the range obtained in other experiments [1-3]

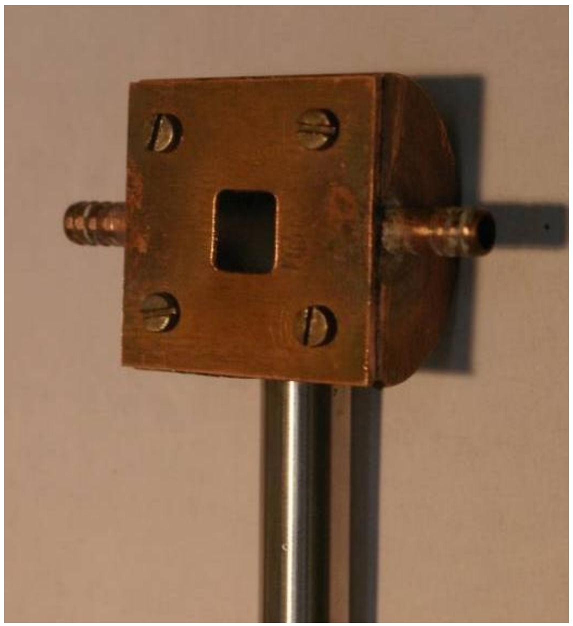
[1] J.R. Fransens et. al. Journal of Physics: Condensed Matter, 3 (1991) 9871.

[2] K. Heinola et. al. Physical Review B, 82 (2010) 094102.

[3] Yu. Gasparyan et. al. Journal of Nuclear Materials 463 (2015) 1013-1016.

e-damage

The main feature of e-damdge is small energy transfer in collisions with atoms of the lattice. Therefore, point defects are formed mainly in this case.



Experimental facility:
Energy: 3.5 MeV
Current: 40 μA/cm^2

The sample is placed at the atmosphere. Water cooling allows to keep the sample temperature below 100 °C.

Irradiated samples are under analysis by PALS in Munich TU and SCN CEN (Belgium).
The next step will be D plasma or atomic loading.
Then, NRA and TDS analysis.

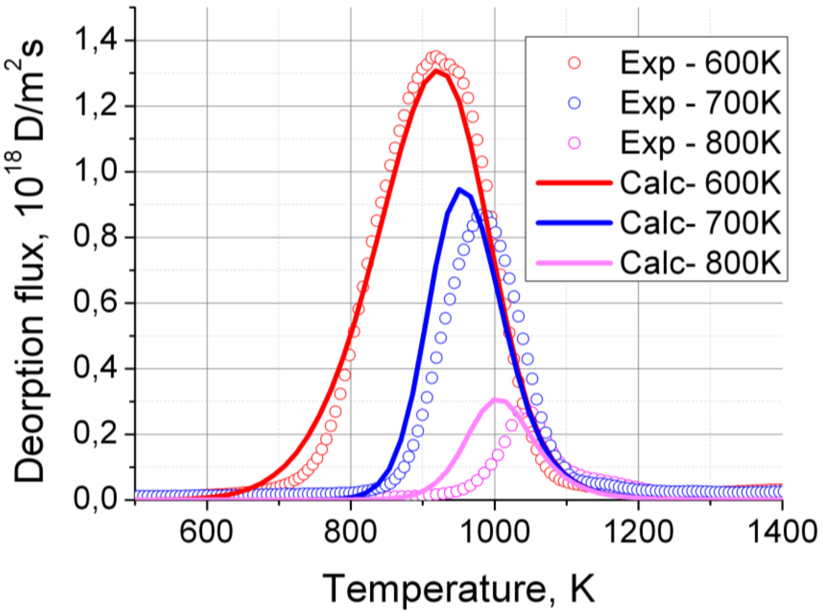
20 MeV W ion damage

A number of experiments have been done already at IPP (Garching) with damaging by 20 MeV W ions.
The damaged zone is much larger (about 2 μm) in this case.

A part of TDS for these experiments has been done in MEPhI.

A significant D retention was observed even after exposure at 800 K.

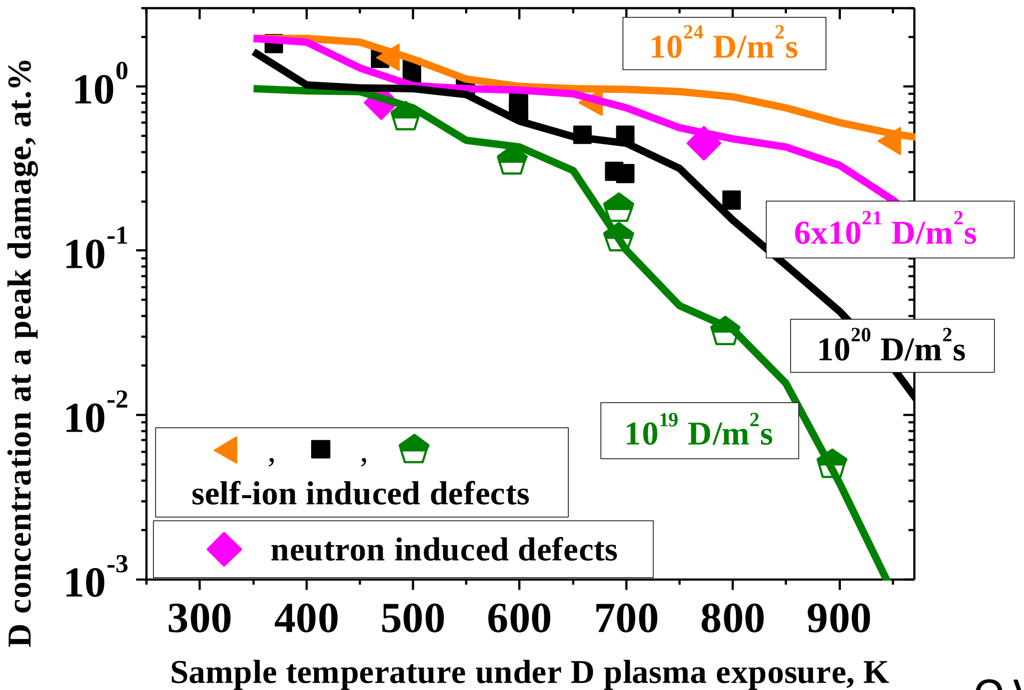
The detrapping energy for high energy traps was estimated from these experiments to be E =1.7 – 2.0 eV



Yu.Gasparyan et al., JNM, 2015.

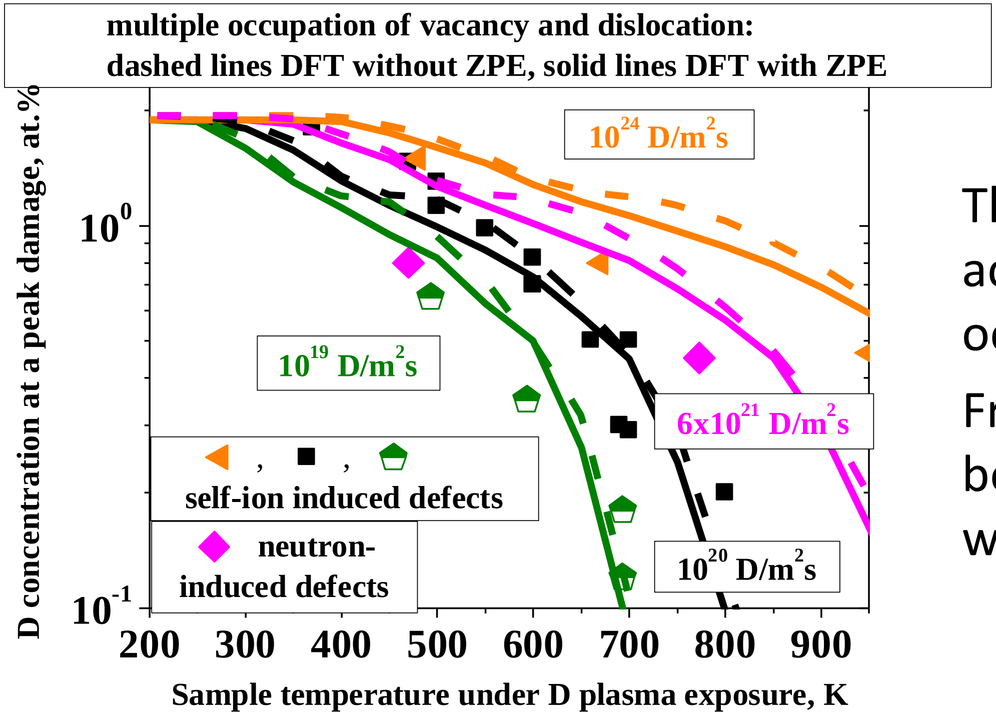
20 MeV W ion damage

A number of experiments with self-damaged W was done by O. Ogorodnikova. This plot presents experimental data and calculations for D concentration at the peak damage. Data for n-damaged samples by [Hatano et al., Nuclear fusion, 2013] are also given for comparison.



O.V. Ogorodnikova, JAP, 2015

Multiple site trapping



O.V. Ogorodnikova, JAP, 2015

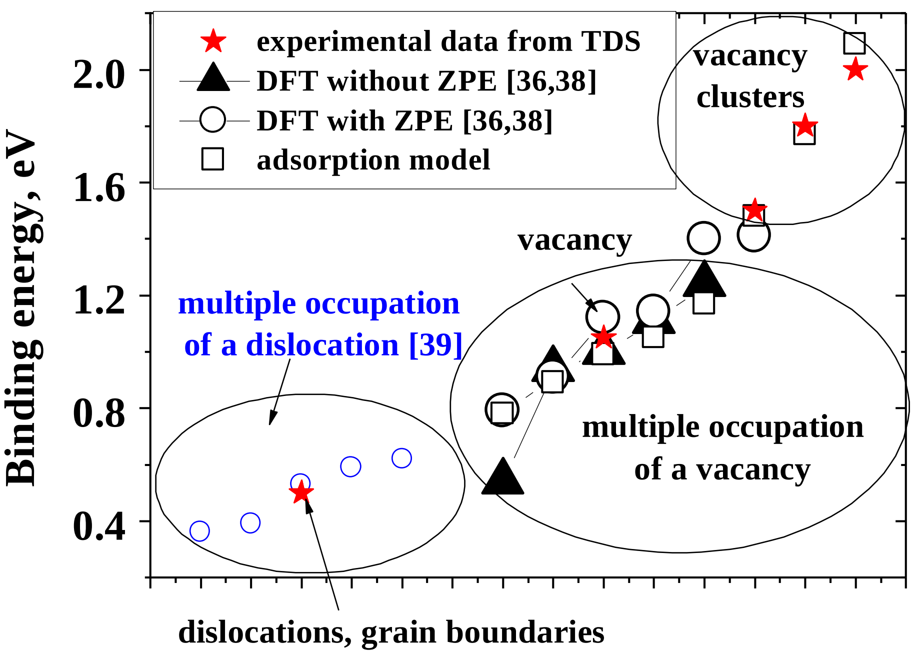
The best agreement with experiment was achieved, if one assume multiple sites occupation of traps.

From the other side, the similar result can be achieved if one use a range of defects with similar binding energy.

To see the difference between multiple trapping sites and several kinds of traps one needs a very delicate experiment with low concentration of defects.
At higher concentration the effect can not be observed due to re-trapping.
(analyzed in E. Marenkov et al., submitted to JNM)

Final remarks

O.V. Ogorodnikova, JAP, 2015



Vacancies and vacancy clusters have considerably higher binding energy than other types of defects.
One can see often well separated peaks corresponding to these two types of defects.
Often these peaks are very broad, likely, due to multiple sites trapping.

There are a number of defects (dislocations, grain boundaries, et al.) with low binding energy, which form low temperature peak. It is often difficult to separate peaks in this region.

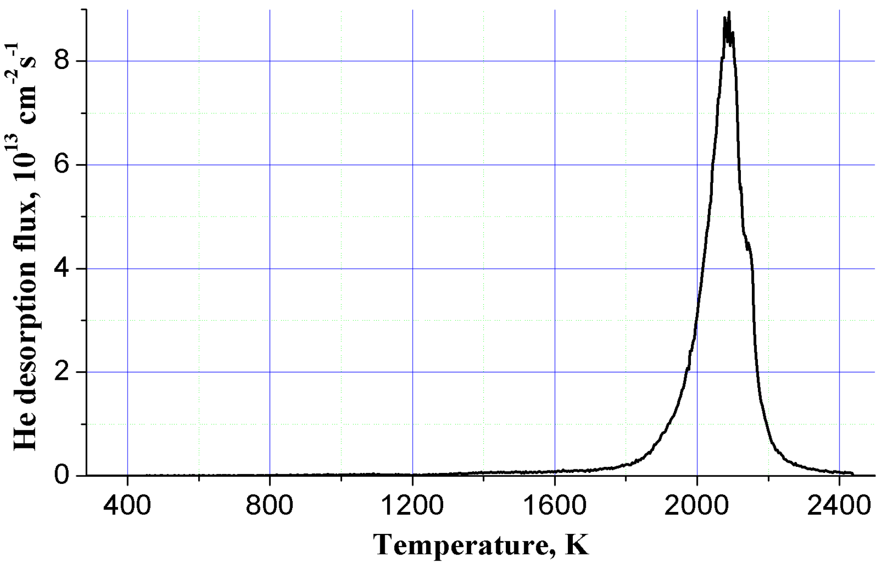
Summary

- The list of binding energies for radiation defects, formed by various irradiation types is similar. The highest binding energies should be attributed to vacancies, vacancy clusters and voids.
- To derive a precise numbers one should perform delicate experiments with well controlled conditions.
- E_{dt} or (E_b+E_d) can be derived from the series of experiments with different heating rates, depending on trap concentration. One should pay attention also to surface conditions.
- Steady state D concentration in radiation defects depends on the incident flux. High total retention is possible even at 800 K.

Thank you for your attention!

Helium desorption

3 keV He⁺, RT,
recrystalized sample



100 eV plasma, ~1200 K,
unannealed sample