P2-15

Ion - induced erosion of tungsten surfaces studied by a sensitive quartz-crystal-microbalance technique

A. Golczewski[a*], A. Kuzucan[a], K. Schmid[b], J. Roth[b], M. Schmid[a], and F. Aumayr[a]

[a] *Institut für Allgemeine Physik, TU Wien, Association EURATOM-ÖAW, Vienna, Austria*
[b] *Max Planck Institute for Plasma Physics, EURATOM Association, Garching, Germany*

ABSTRACT

A highly sensitive quartz crystal microbalance technique was used to study erosion of polycrystalline tungsten films due to impact of deuterium, carbon and argon ions, as well as retention of deuterium in these films. Polycrystalline tungsten films coated onto a SC-cut quartz crystal were bombarded by ions with impact energies from 100 eV up to a few keV and the frequency change due to mass loss (sputtering, desorption) or mass gain (implantation, adsorption) during bombardment was determined. Our setup was capable of detecting mass-changes as small as 10^{-5} μg/s, which corresponds to a removal (or deposition) of only 10^{-4} W monolayers/s. While our total sputtering yields for deuterium and argon projectiles compare well with the results of previous work, we derive new data on sputtering of tungsten by carbon ions. In addition we demonstrate that our setup is well suited for determining deuterium retention rates in tungsten.

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PSI 18 Keywords: Sputtering, Erosion & Deposition, High-Z material, Retention, Tungsten

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Corresponding author:
A. Golczewski, Institut f. Allgemeine Physik, TU Wien
Wiedner Hauptstr. 8-10/134, A-1040 Wien
golczewski@iap.tuwien.ac.at

Presenting author:
A. Golczewski, Institut f. Allgemeine Physik, TU Wien
Wiedner Hauptstr. 8-10/134, A-1040 Wien
golczewski@iap.tuwien.ac.at

1. INTRODUCTION

Quartz-crystal-microbalances (QCM) are widely used as sensors for monitoring of thin film growth in deposition processes [1-2]. In the past years, a variety of QCM applications in gaseous and liquid environments such as gas chromatography detector, gas sensor, biosensor and diagnostic tool for plasma-wall interaction [3] have been reported. At TU Wien, we have developed a sensitive QCM in order to measure total sputter yields (including both neutral and charged secondary particles) in ion - surface collisions [4-6]. In this experiment the target material is first deposited on the quartz crystal as a thin polycrystalline film and then bombarded by slow (projectile range smaller than film thickness) singly or multiply charged ions, with the sputter yield being determined from the mass loss of the target film, measured as a change of the QCM frequency.

Whereas quartz crystals are commonly used for determination of the area mass and hence the thickness of deposited material, the rate of material removal has mainly been studied with other techniques such as the conventional microbalance, secondary neutral mass spectrometry or catcher foils subsequently analyzed by Rutherford back-scattering. This is not astonishing because the use of quartz crystals for direct sputter-yield measurements encounters severe problems. The rates of material removal and hence the frequency changes are rather low compared to most deposition applications, requiring a rather high frequency stability of the crystal and of the oscillator circuit as well as high accuracy and resolution in determining the resonance frequency. Furthermore, a substantial amount of energy is deposited by the primary particles on the sputtered surface, causing problems due to thermal drift. In many deposition applications, the energy deposition per incident atom is only a few eV (sublimation energy plus heat radiation from the evaporation source), while in the case of sputtering the energy deposited per impinging projectile is rather in the range of a few hundred eV up to several keV. Other problems arise from the sensitivity of the resonance frequency to surface stress induced by, e.g., sputter induced defects, implantation of projectile ions and non-uniformal mass removal across the ion beam cross section [6, 7].

The QCM apparatus developed at TU Vienna has been optimized with respect to stability and accuracy [6] and is therefore well suited to determine total sputtering yields in ion-surface collisions relevant for plasma-wall-interaction in present day fusion devices. In this contribution we will present recent measurements of the erosion of polycrystalline tungsten films due to impact of deuterium, carbon and argon ions, as well as retention of deuterium in these films.

2. EXPERIMENTAL METHOD

Polycrystalline tungsten films (typical thicknesses between 500 and 1000 nm) were deposited onto cylindrical quartz crystals, which themselves are coated with thin Au-electrodes on both sides. For the deposition of the polycrystalline tungsten films either an e-beam evaporation system at TU Wien or a magnetron-sputter system at IPP Garching was used. Since the sputter yield for both deposition methods did not differ substantially no further investigations regarding the differences in surface structure of the samples were conducted. The crystals were SC-cut (stress-compensated) to ensure a minimum influence of the radial stress due to the mounting of the crystal in a sample-holder. The sample holder was designed to minimize all electronic influences by possible inductive and capacitive coupling and the wiring within the UHV-chamber was realized towards the same aim. To exclude possible electronic disturbances such as the stray capacitance, all in-vacuum wiring was doubled, so that a dummy branch was created, which was subject to the same electronic disturbances as the branch connected to the QCM itself [6]. This background can then be subtracted from the signal of the QCM. To minimize all out-of-vacuum influences the first stage of the analyzing electronics was located as close as possible to the UHV feedthroughs of our chamber. The QCM was temperature stabilized and usually operated at the minimum of the frequency vs. temperature curve (around 190 – 200 °C) to minimize effects of thermal drift. The heating up of the crystal by the incident ion beam was estimated to be around 3-5°C in the case of the low current irradiations (typically around 10^{11} particles/cm^{2}s) at the facility at the TU Wien and could be compensated by the heating control unit. In the case of the high current irradiations (typically around 10^{14} particles/cm^{2}s) at the IPP Garching however, the temperature increase was estimated to be around 40-50°C, but since the mass change resulted in a drastically bigger frequency change than the effect of the thermal drift, this influence could be neglected. The nominal Eigenfrequency of the crystal is very close to 6 Mhz and the high stability of our setup allows the detection of changes as small as several mHz [6].

With these precautions our setup is capable of detecting mass-changes as small as 10^{-5} μg/s, which corresponds to a removal (or deposition) of only about 10^{-4} monolayers of tungsten per second. In cases where the temperature dependence of sputtering processes is of interest, our heating system allows to perform such measurements from room temperature up to 250°C, though with considerably reduced sensitivity.

All experiments have been conducted in high vacuum (typ. pressure of $5 \cdot 10^{-8}$ mbar). The carbon and argon ions used for the tungsten erosion investigations in this work were extracted from the 14.5 GHz ECR-ion source at TU Wien [8] with energies between 1 and 5 keV. A deceleration lens just in front of our QCM setup allowed to extend the impact energy range down to 100 eV. Deuterium ions of 100 eV - 8 keV from the Dua-Pigatron type ion source at IPP-Garching were used for the tungsten erosion and deuterium retention measurements [15]. In the IPP-Garching measurements the final beam energy was reached both through deceleration in front of the target and by reducing the ion extraction voltage.

A typical response curve of the QCM is presented in Fig. 1. Before irradiation a stable Eigen-frequency of the crystal is observed, which drastically changes when the ion bombardment sets in. Mass removal (i.e. sputtering) leads to a frequency increase until the ion-beam is switched-off again and a constant frequency is re-established. A non-linear increase of the frequency response despite irradiation under stable ion beam conditions usually hints at a change of surface conditions, like the removal of an initial oxide or carbide layer. After several minutes of irradiation, such nonlinearities disappear, and a steady state surface composition is reached.

According to Sauerbrey [9] the frequency change of a cylindrical quartz crystal induced by a mass change Δm can be expressed through the relationship:

$$\frac{\Delta f}{f} = -\frac{\Delta m}{m} \tag{1}$$

where m is the total mass per area of the coated crystal and f is the nominal Eigenfrequency of the quartz crystal. From this starting point a simple relation between the frequency-change per unit time $\Delta f/\Delta t$ and the total mass change per unit time $\Delta m/\Delta t$ and therefore the amount of removed (or implanted) material mass per impinging projectile y (in amu/ion) can be determined:

$$y = \frac{mqe_0}{m_u f}\frac{1}{I}\frac{\Delta f}{\Delta t} \tag{2}$$

where qe_0 is the charge of the projectile, m_u the conversion factor between atomic mass units and SI units, I the total ion beam current and $\Delta f/\Delta t$ the slope of the mass change due to bombardment. The total sputtering yield Y (i.e. the number of sputtered target atoms per incident ion) is then given by

$$Y = \frac{y}{m_T} \tag{3}$$

where m_T is the atomic mass of the target (m_T= 184 amu in the case of W).

3. SPUTTERING MEASUREMENTS

To check the accuracy of our setup, sputtering yields for Au films during Ar ion bombardment was re-measured, because for this case sufficiently reliable data are available. Our results agree very well with previous experimental data for this projectile target combination [10,11]. Fig. 2 shows measured sputtering yields obtained by bombardment of polycrystalline tungsten with Ar^+ and C^+ ions in the energy range from 180 eV to 4 keV. Our results for Ar^+ projectiles compare well with the results of other groups [12-16] using different experimental methods. Additionally all data are well described by TRIM.SP calculations [12]. We also report new data on the energy dependence of sputtering of tungsten by carbon ions (shown in fig. 2), since existing literature presents general investigations of tungsten erosion by carbon impact, but provides no systematic examination of the energy dependence of the sputtering process [17]. Again predictions by TRIM.SP calculations agree well with our experimental data at least for impact energies above 300 eV. The deviation from the theoretical predictions for lower energies is likely to be the effect of a yet unknown portion of fast neutral projectiles within the beamline. This error occurs only for the two lowest energy-points, since those were the only measurements, where a deceleration was used. Error-bars (omitted in fig. 2 for clarity) for our sputtering yields were estimated and did not exceed ± 10%.

We also looked for possible charge-state dependent sputtering effects in tungsten ("potential sputtering", see e.g. [4, 5]) by using carbon ions in different charge states (1+ and 2+). Within our experimental limits, however, we could not find any significant difference and therefore our conclusion is that (as expected) tungsten is not subject to potential sputtering.

Sputtering yields measured with our QCM setup for impact of deuterium ions (impact energies well above the sputter threshold) on tungsten are shown in Fig. 3. Results for both e-beam evaporated and magnetron sputtered tungsten surfaces show no significant difference. A comparison with existing experimental data and recent TRIM.SP calculations yields good agreement in the low impact energy range, while above 500eV our sputtering yields are significantly higher. We note, however, that earlier TRIM.SP calculations [18] have already yielded sputtering yields corresponding very well to our measured data, but were corrected to better fit the then existing experimental data. Both theoretical predictions are shown in Fig. 3.

4. D^+ RETENTION IN TUNGSTEN

The high sensitivity of our QCM triggered interest of the PWI community to use it for studying other plasma-wall interaction processes, in particular deuterium (= fuel) retention in tungsten. In fig. 4 we present first results obtained during a recent experimental campaign at IPP Garching. In this experiment the tungsten film was bombarded with 100 eV deuterium ions, an impact energy that can be regarded as sufficiently below the sputter threshold (see fig. 3). This ensures that the QCM only registers the mass increase due to deuterium implantation in W. The retained D^+ fluence as measured by the QCM shows the right order of magnitude (10^{-2}) as seen in earlier retention experiments [21].

The retention in our case, however, indicates the onset of saturation behavior not observed in the earlier work. Whether this is due to the use of tungsten films of limited thickness (in our work, a film thickness of 500nm was used) as compared to bulk tungsten (previous work) has still to be investigated. Although very suitable for the real-time measurement of the mass increase due to deuterium retention in plasma wall materials in a laboratory environment, the QCM apparatus shows considerable limitations regarding the applicability in non-laboratory conditions. Mainly, since the total mass change is determined, any portion of fast projectiles within the ion-beam proves problematic, because the sputtering by those fast particles results in a mass removal camouflaging the mass intake due to retention. Also for surroundings with strongly varying temperatures the accuracy of the QCM is lowered (due to thermal drift) to a point, where retention measurements might prove impossible.

5. OUTLOOK

We have demonstrated the capability of our QCM method to measure mass removal from fusion relevant materials like tungsten during ion bombardment with high sensitivity and accuracy. In addition we have also presented the first evidence that the method is also able to study deuterium retention in relevant divertor and wall materials. The prospect to investigate these processes as a function of surface temperature (although linked to an intrinsic decrease in sensitivity) will open up an even wider field of possible plasma-wall-interaction studies.

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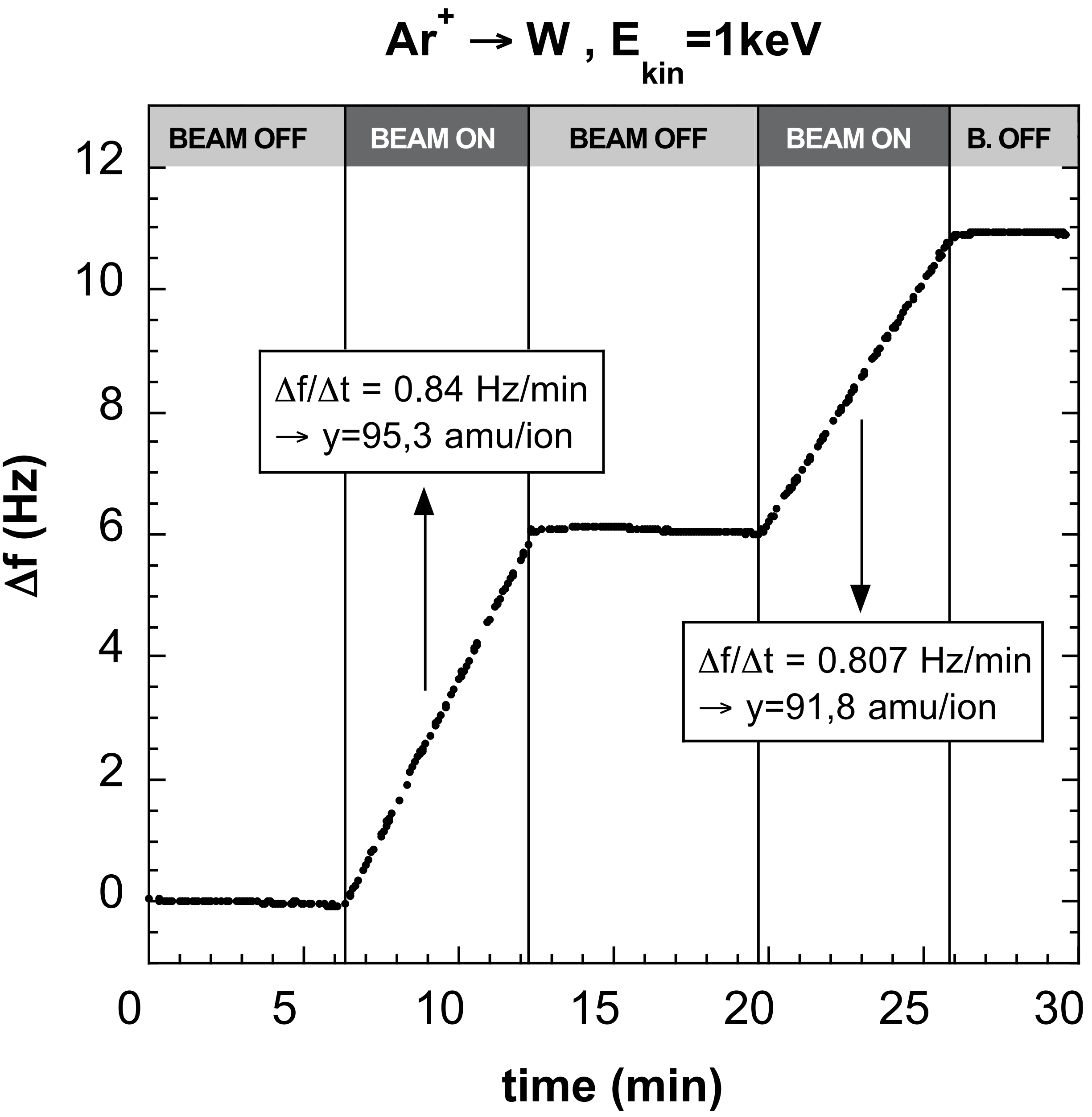
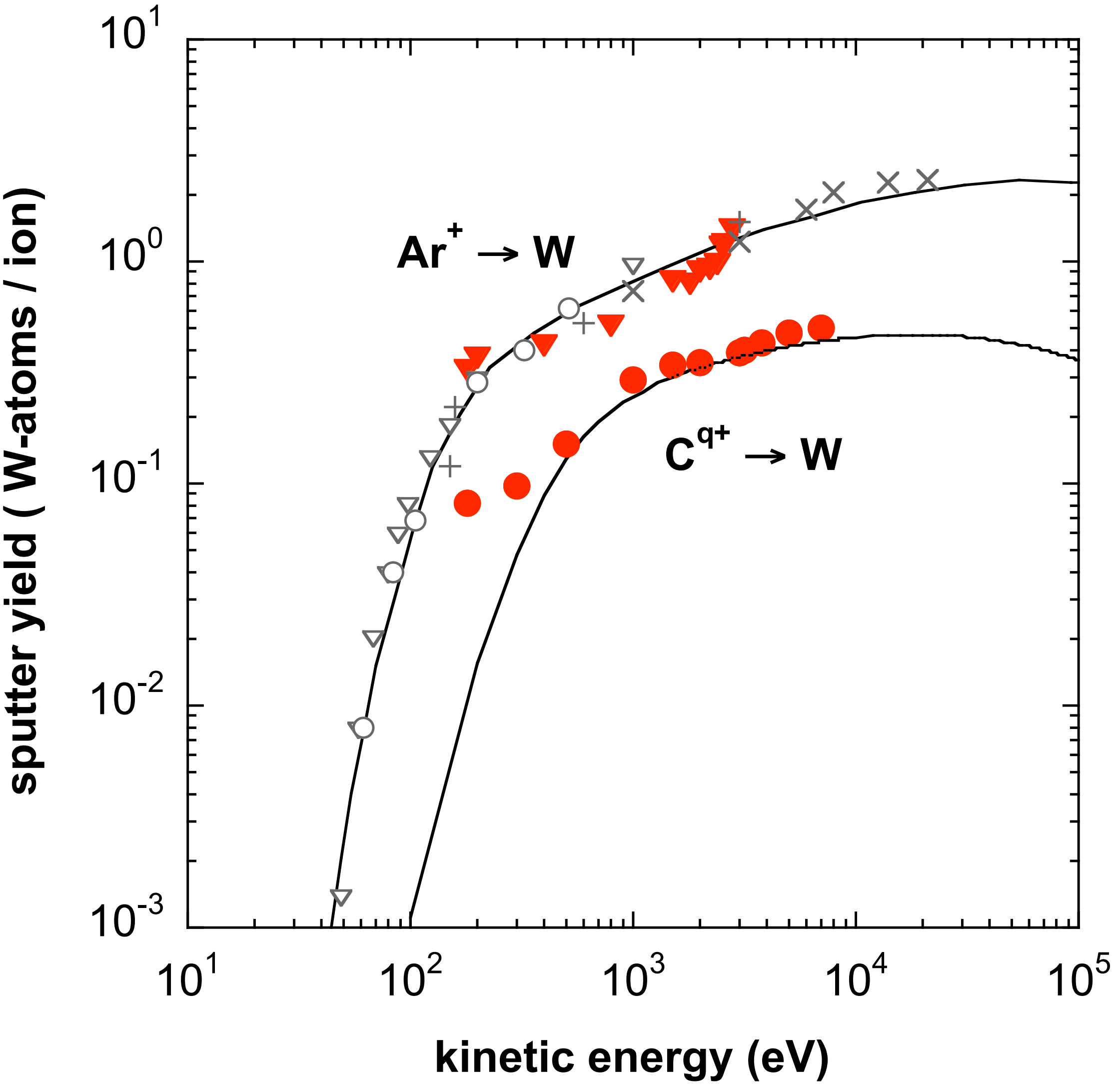
Fig. 1: Typical frequency response of the QCM to irradiation of the W film by Ar^+ ions. Two subsequent measurements are shown.

Fig. 2: Total sputter yields of polycrystalline tungsten bombarded by Ar^+ and C^+ ions. Full (red) symbols have been obtained within this work; solid lines represent TRIM.SP calculations [12]. Previously published data are included for comparison: Smith et al. 1975 (∇) [13], Laegreid et al. 1961 (o) [14], Roth et al 1979 (+) [15] and Koshkin et al. 1969 (x) [16].

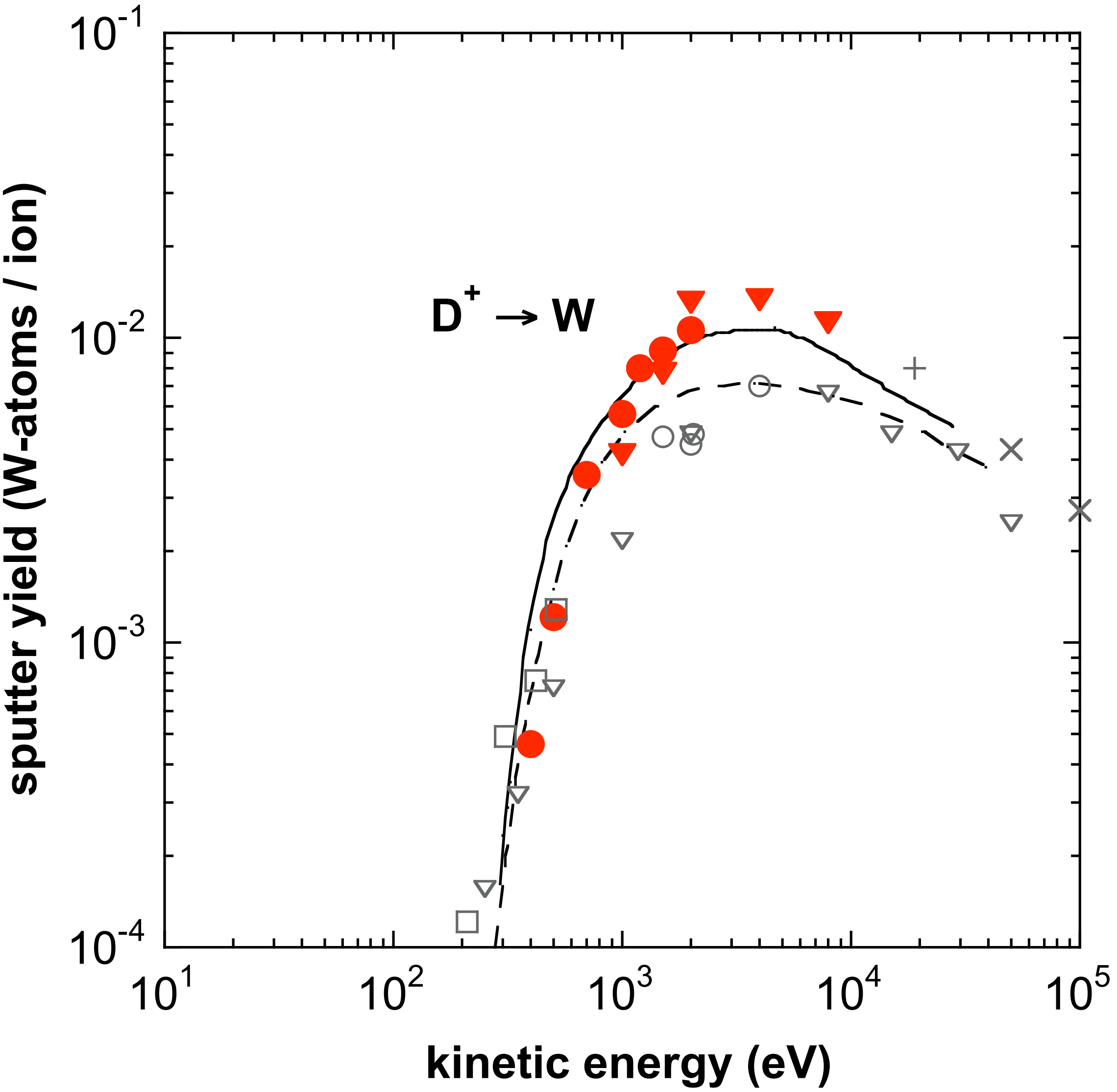
Fig. 3: Total sputter yields of polycrystalline tungsten bombarded by D^+ ions. Full (red) symbols have been obtained within this work; (circles represent bombardment on e-beam evaporated and triangles on magnetron sputter deposited W films); the dashed line represents recent TRIM.SP calculations [12], while the solid line shows earlier TRIM.SP predictions [18]. Previously published data are included for comparison: Roth et al 1979 (∇) [15], Eckstein 1993 (o) [18], Guseva et al 1999 (squares [19]), Yonts et al. 1969 (+) [20] and Bohdansky et al. 1982 (x) [18].

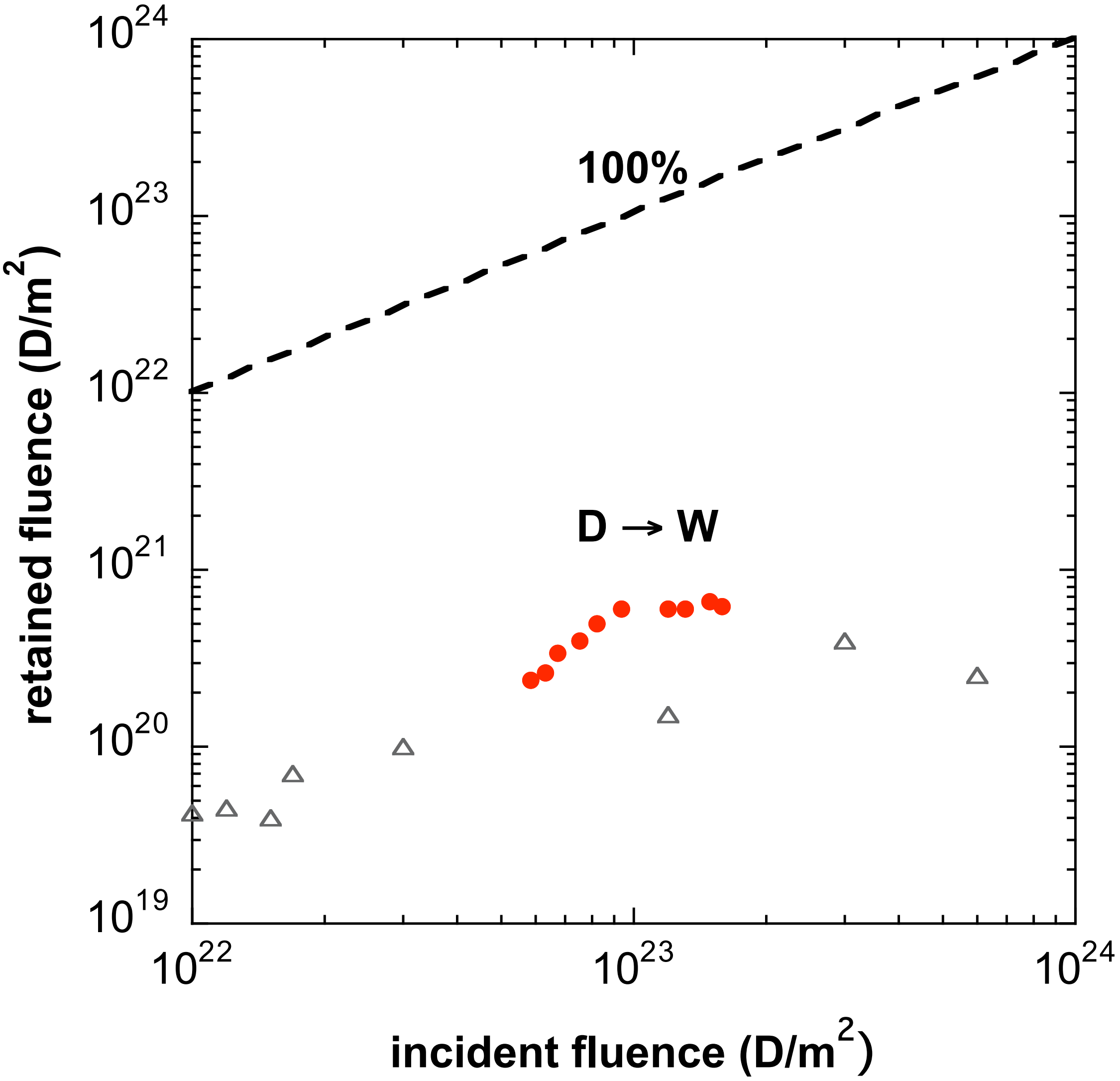
Fig. 4: Deuterium retention in polycrystalline tungsten under bombardment by 100eV D^+. The dashed line represents a hypothetical absolute retention of all incoming flux. Full (red) symbols have been obtained within this work, while open triangles are implantation data reported in [21].


$Ar^+ \rightarrow W$, E_{kin}=1keV
BEAM OFF
BEAM ON
BEAM OFF
BEAM ON
B. OFF
Δf/Δt = 0.84 Hz/min
→ y=95,3 amu/ion
Δf/Δt = 0.807 Hz/min
→ y=91,8 amu/ion
Δf (Hz)
0
2
4
6
8
10
12
time (min)
0
5
10
15
20
25
30



sputter yield (W-atoms / ion)
kinetic energy (eV)
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^1
10^2
10^3
10^4
10^5
$Ar^+ \rightarrow W$
$C^{q+} \rightarrow W$


D$^+$ → W
sputter yield (W-atoms / ion)
kinetic energy (eV)
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^1
10^2
10^3
10^4
10^5


100%
D → W
retained fluence (D/m^2)
incident fluence (D/m^2)
10^{24}
10^{23}
10^{22}
10^{21}
10^{20}
10^{19}
10^{22}
10^{23}
10^{24}