ENABLING RESEARCH PROJECT ENR-MAT.01.VR

Electronic interactions of slow ions and their influence on defect formation & sputter yields for plasma facing components

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24 October 2024, EUROfusion Science Meeting on results of Enabling Research Projects 2021-2024

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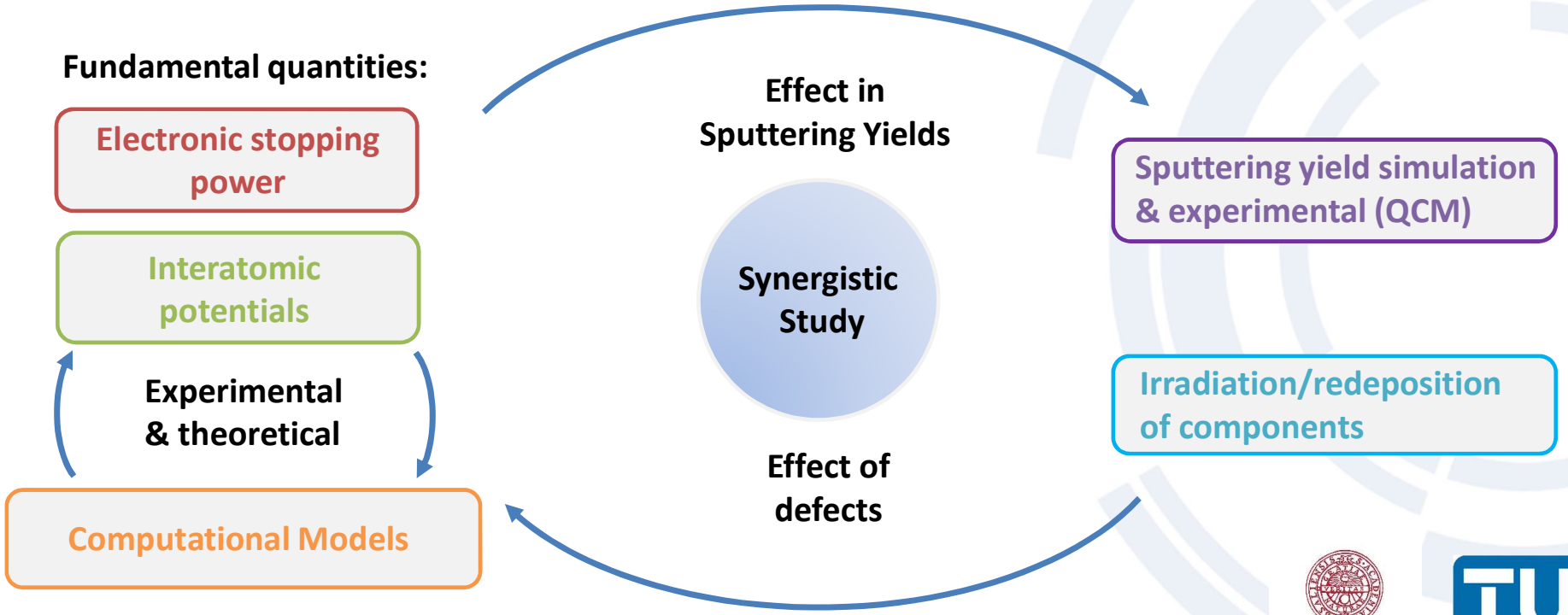
Aim

To investigate underlying **quantities fundamental** for **sputtering** and **defect formation** from plasma-wall interaction:

- Important input variables in modelling of **erosion** and **implantation** in plasma facing components.
- Essential for precise **material characterization** using **ion beam analyses**.



UPPSALA UNIVERSITET

TU WIEN

Aalto University

Team: 2021-2024

VR	ÖAW	VTT
Eduardo Pitthan (PI)	Martina Fellinger	Andrea Sand
Jila Shams-Latifi	Johannes Brötzner	Evgeniia Ponomareva
Philipp M. Wolf	Christian Cupak	Ludovico Caveglia Curtil
Petter Ström	Richard Wilhelm	Tetiana Malykhina
Per Petersson	Friedrich Aumayr	Akseli Aro

Start: May of 2021.

VR main tasks: Sample preparation and characterization, electronic loss measurements, interatomic potential measurements, and ion irradiation experiments.

ÖAW main tasks: Sputtering yield measurements, and BCA-based simulations.

VTT main tasks: Computational modelling (simulations of electronic stopping power & MD sputtering yields).

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This presentation

Energy losses

Background and motivation.
Experimental procedure.
Experimental results.
Theoretical results (TD-DFT).

Interatomic potentials

Background and motivation.
Experimental/modelling procedure.
Experimental results.
Comparison with current models.

Sputtering yields

Experimental procedure.
Experimental results.
Comparison with BCA simulations.
Effect of fundamental quantities in BCA simulations.

Aim: Energy losses

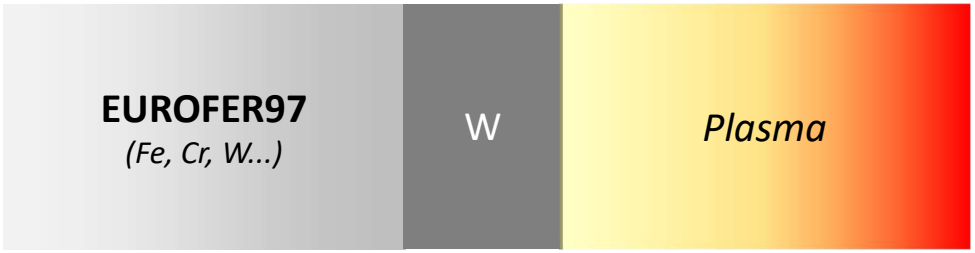
Important fundamental quantity for simulation of PWI:

$-dE/dx$ ➡ **Stopping Power** (energy loss per unity path).

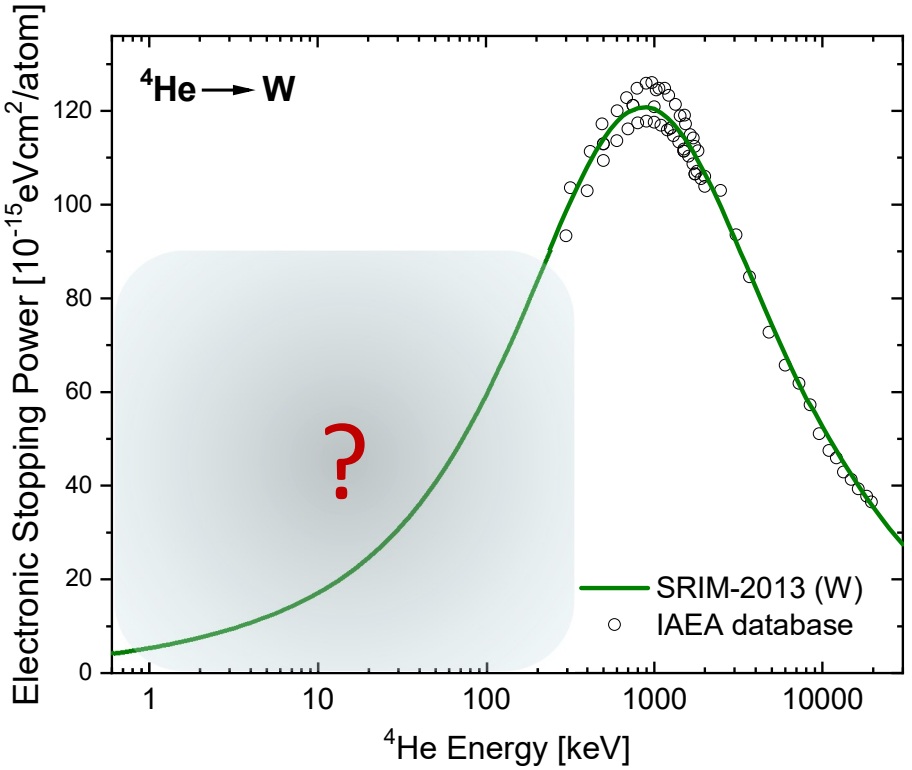
$\varepsilon = -\frac{1}{N}\frac{dE}{dx}$ ➡ **Stopping cross section** (independent of density).

Semi-empirical models (such as used in SRIM code [1])
use ***available experimental data…***

First wall in DEMO:



→ Only one dataset below 300 keV (M. Moro-UU, 2022).
→ Limited experimental information at low energy ranges.
→ For Fe-Cr alloys: No experimental data at any energy.



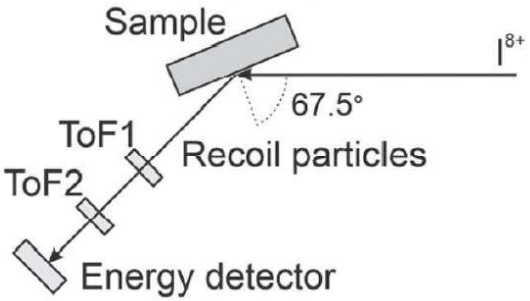
Electronic Stopping Power of Matter for Ions: available data from International Atomic Energy Agency (IAEA) [2] (Mor21 database removed – this work)

[1] J. F. Ziegler *et al. Nucl. Instr. Meth. Phys. Res. B, 268, 1818-1823 (2010).*
[2] *https://www-nds.iaea.org/stopping/stopping_hydr.html.*

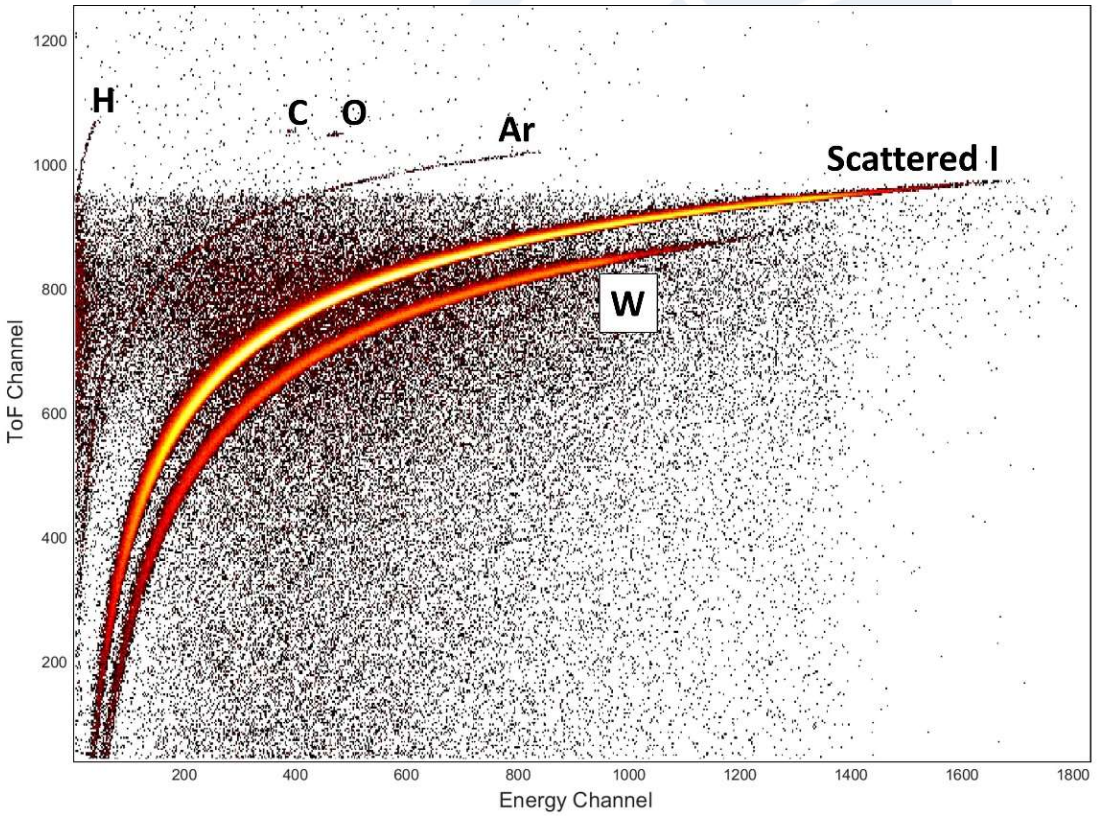
Sample preparation/characterization

Characterization of the chemical composition of the pristine samples (Fe, W, EUROFER) by combined ion beam based techniques (UU), as a protocol for the standard quality control.

→ Atomic concentration depth profiles by ToF-ERDA:



Example: ToF-ERDA spectrum from sputter-deposited W film.



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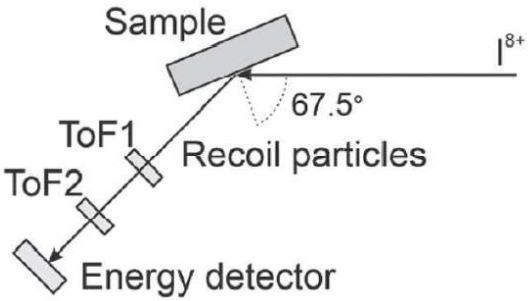
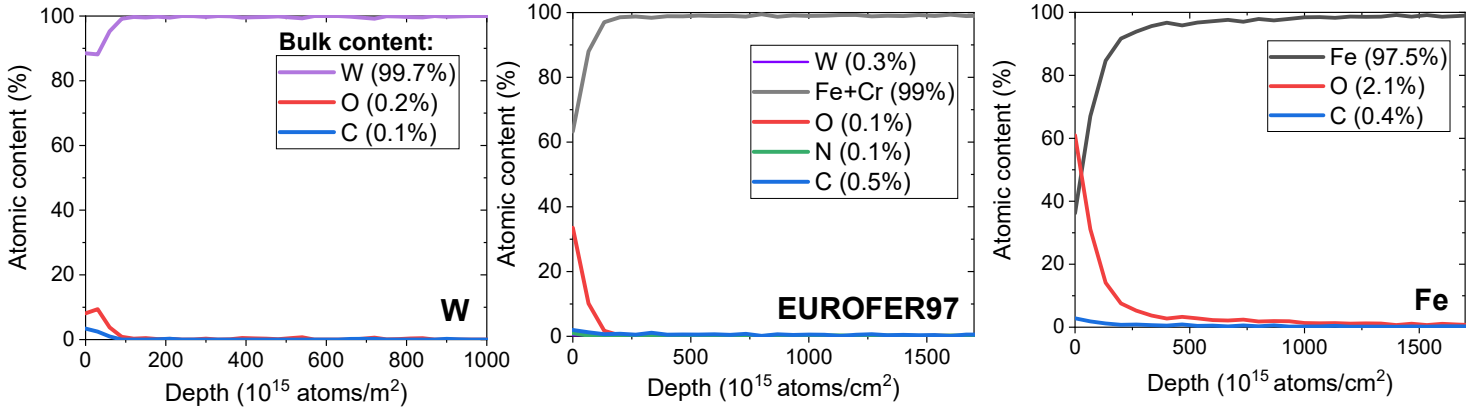
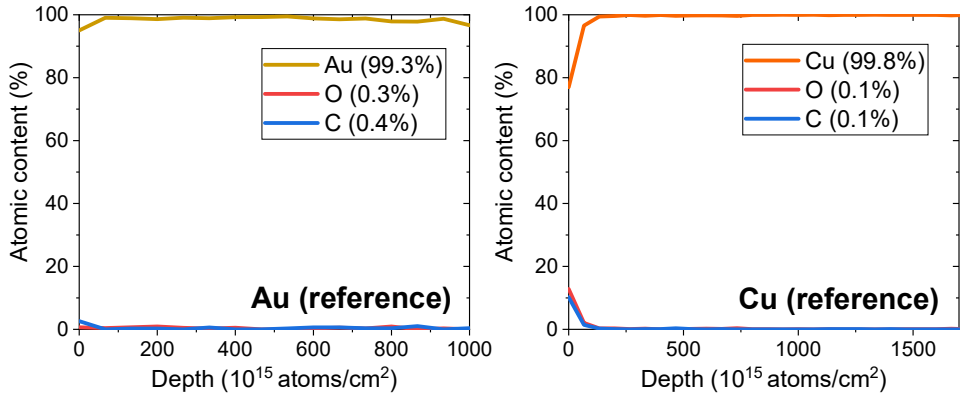




→Low impurity concentrations in bulk.
Evaluation window 500-1000×10^{15} atoms/cm^2

Final stopping data corrected by Bragg's rule:

$$\epsilon^{A_m B_n \cdots} = m\epsilon^A + n\epsilon^B + \cdots$$



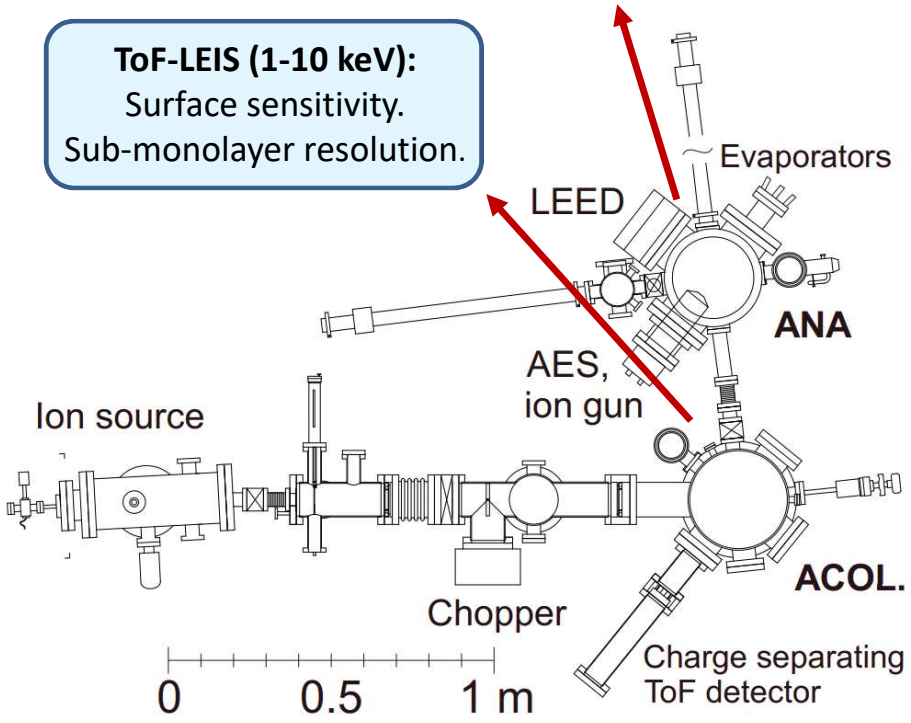
Electronic energy loss measurements

Experimental procedure for low energy regime

ACOLISSA experimental set-up:

Analytical Chamber: Sputtering cleaning, annealing, AES, e⁻beam evaporation, and LEED.

ToF-LEIS (1-10 keV):
Surface sensitivity.
Sub-monolayer resolution.



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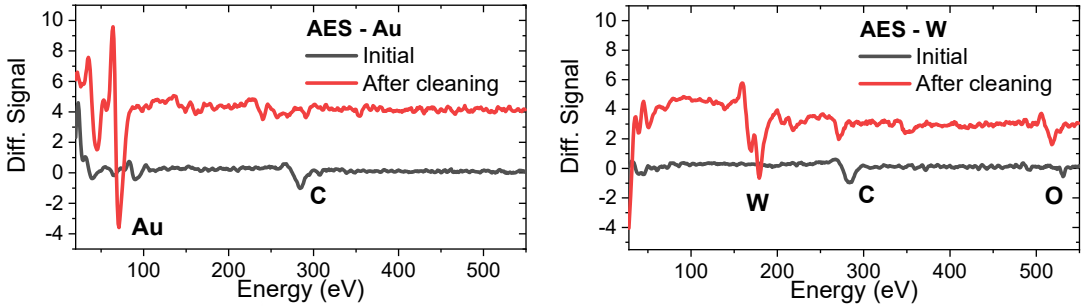
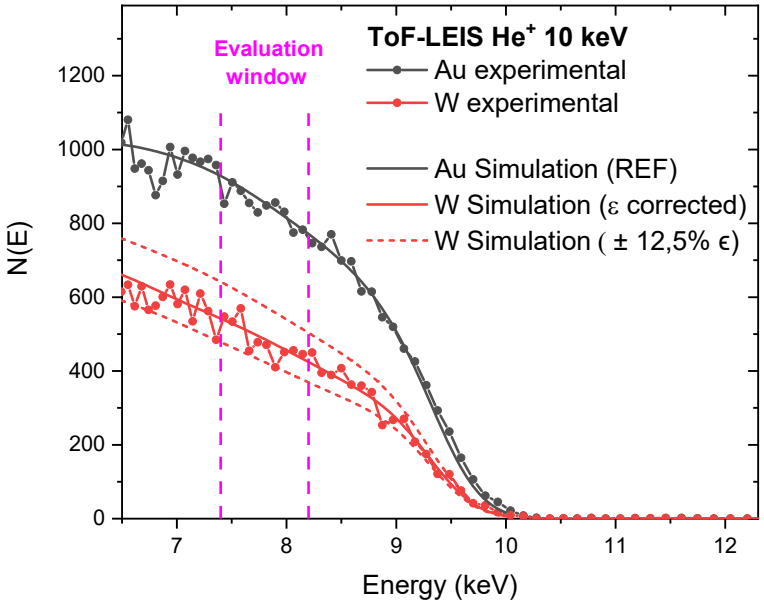
Sample cleaning: *cycles of Ar^+ sputtering 3 keV 30˚.*



SCS relative measurements in comparison with MC simulations:



→Cu/Au as reference under same experimental condition.

→SCS extraction from height ratio.

→ Similar approach for MEIS and MeV energies our laboratory.

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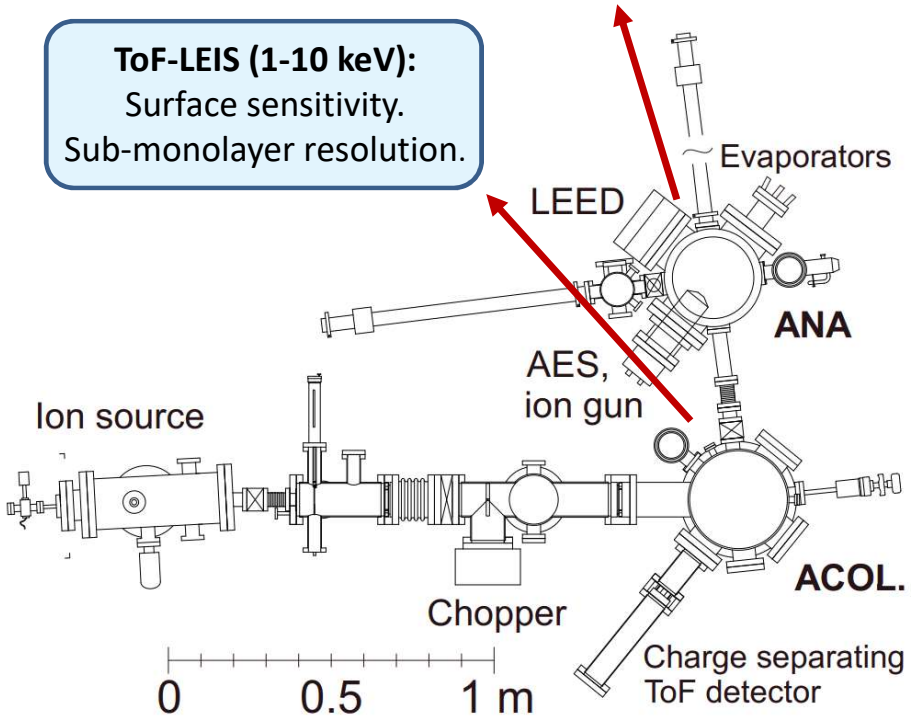

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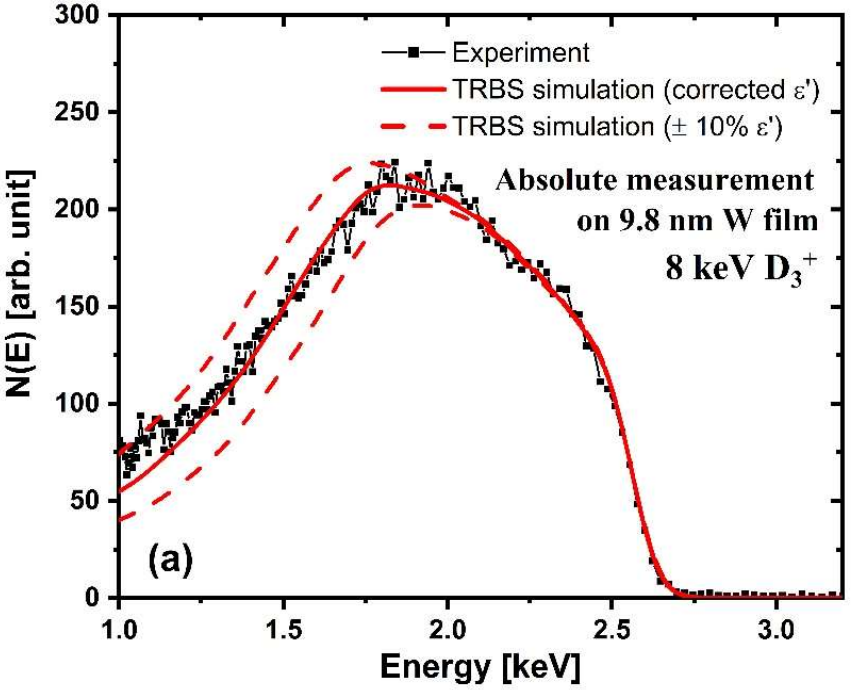
Complementary approach (absolute approach):
Sputter-deposited thin films of PFC on light (C and Si) substrates.

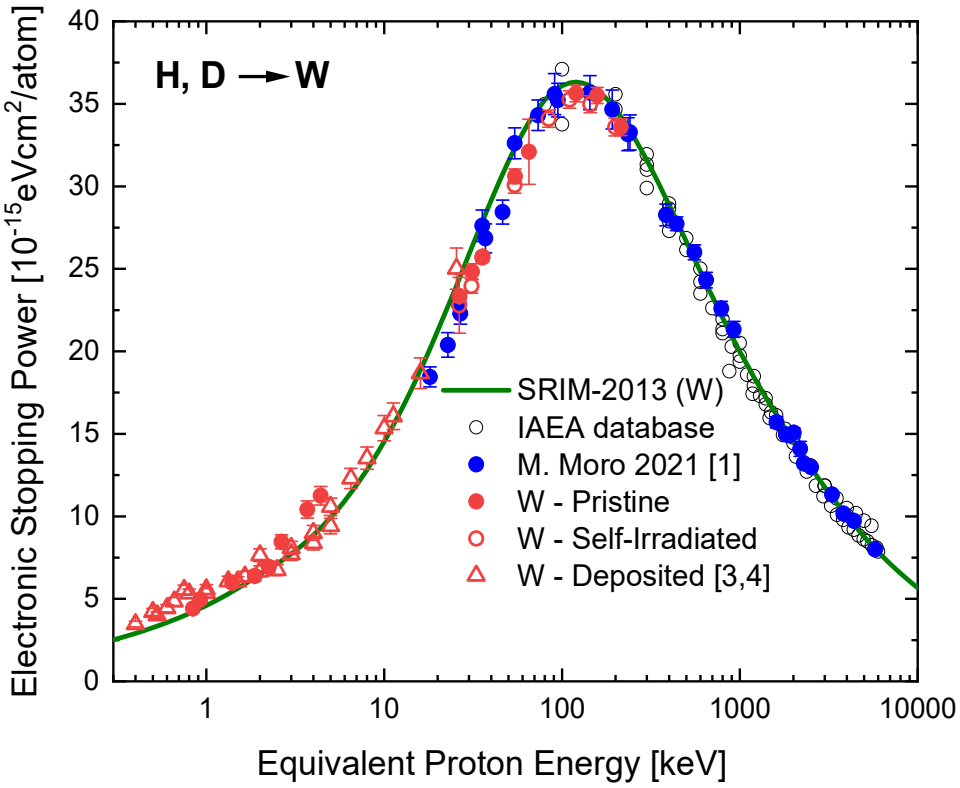
Film thickness was measured by RBS and simulated using SIMNRA to be employed in TRBS simulations.

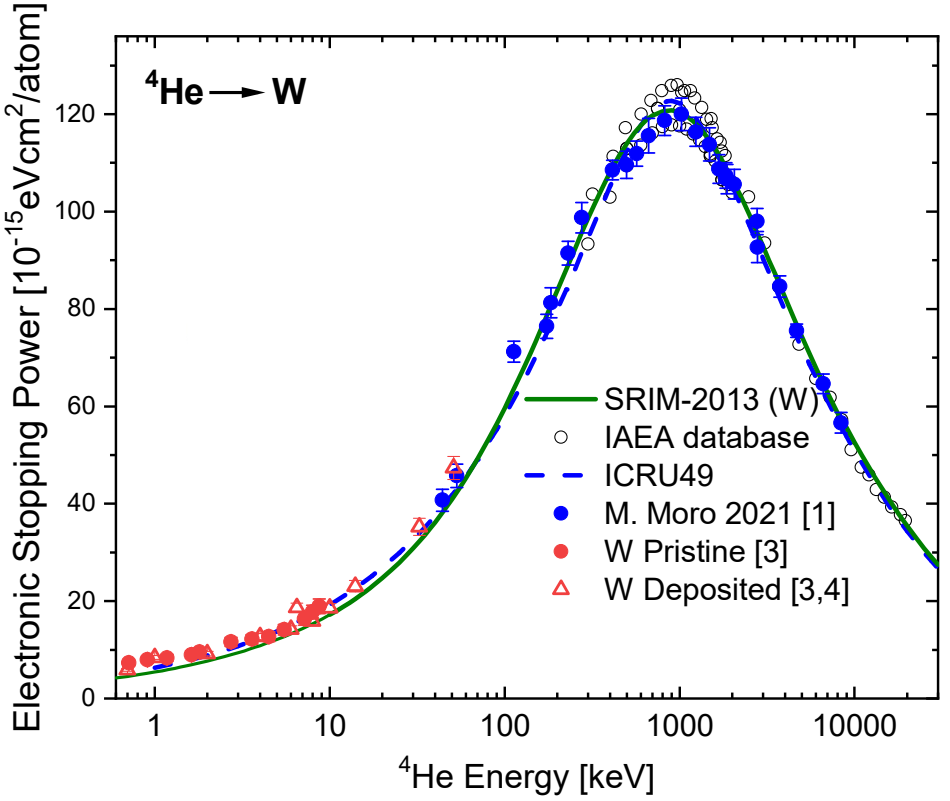
SCS from width from spectrum signal.



Electronic energy loss measurements

Experimental stopping cross-section of pristine Fe, W, and EUROFER97





[1] M. V. Moro et al., Nucl. Instrum. Meth B **498** (2021).
[2] M. J. Berger, et al., Report 49, Oxford Academic (1993).
[3] J. Shams-Latifi, et al., Nucl. Mater. and Energy **36** (2023).
[4] E. Ponomareva, et al., Phys. Rev. B **109** (2024).

Damage of PFC by Self-irradiation:
W^{2+} 2.84 MeV → W (1.6×10^{15} W/cm^2);
Fe^{+} 300 keV → Fe/EUROFER97 (42.2×10^{15} Fe/cm^2).

No significant effect of self-irradiation observed in this work.

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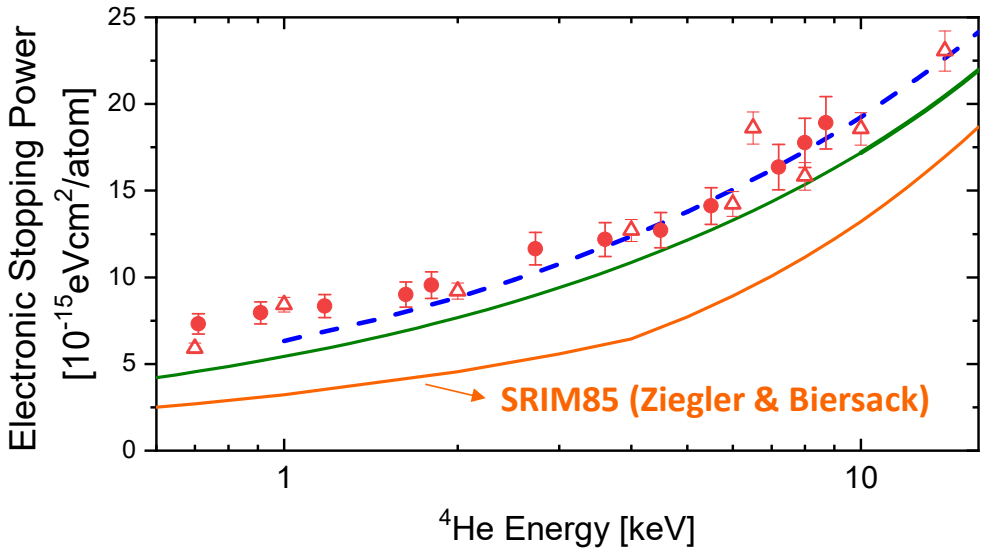
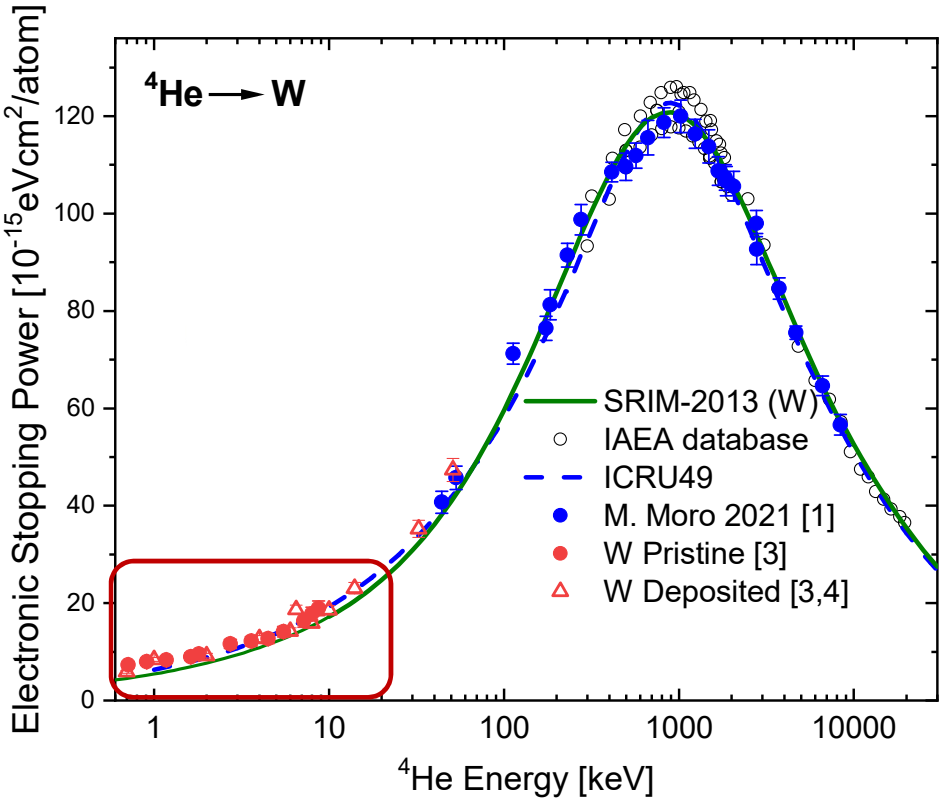


SCS at LEIS published [3,4]:
LEIS (<15 keV): Discrepancies from SRIM-2013 up to 20% for protons and 60% for He.

Discrepancies up to 210% from SRIM-1985.



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Electronic energy loss measurements

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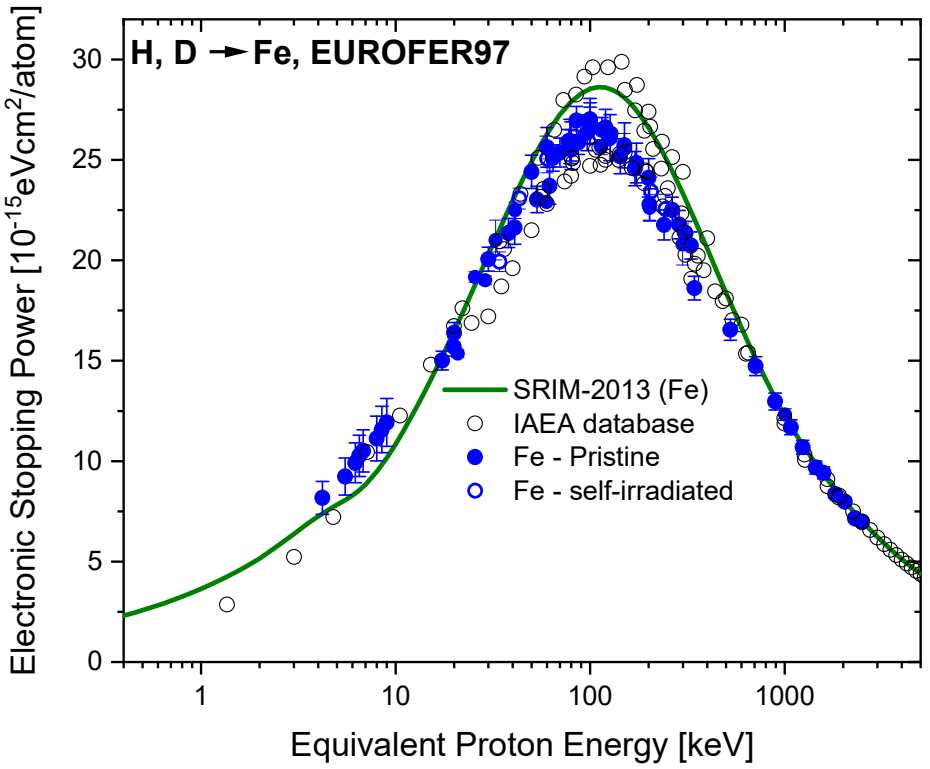
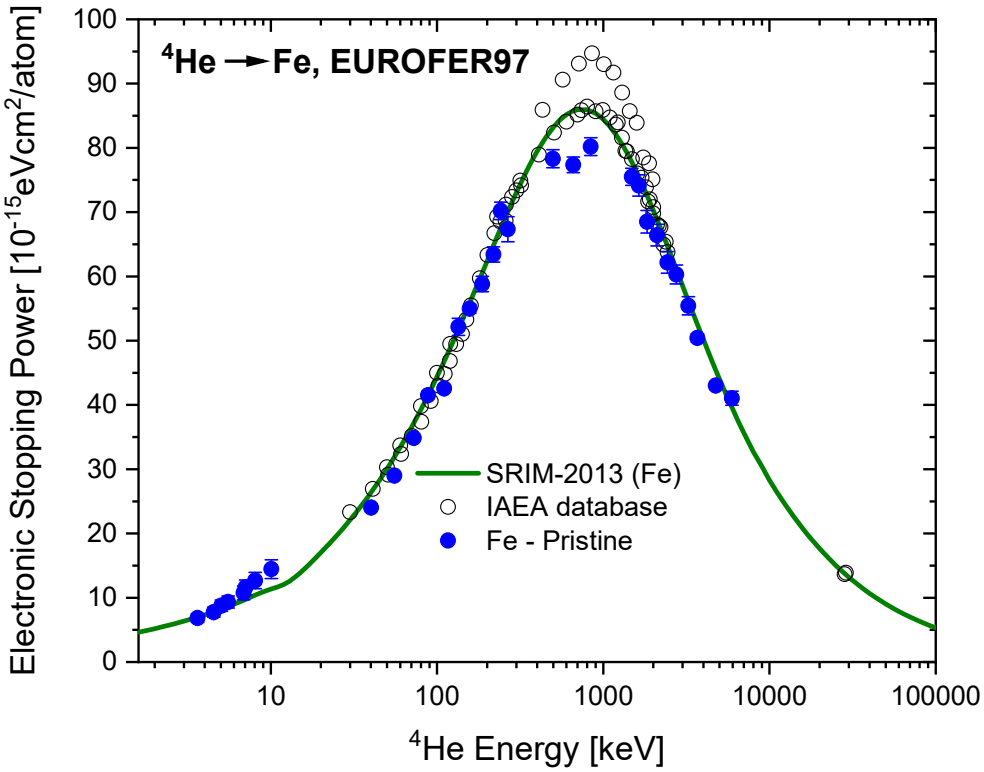




H^+ → Fe

→ Good agreement at MeV range.

→ Large discrepancy with SRIM around maximum (up to 11%).

→ Large discrepancy with SRIM at low energy range (up to 20%).

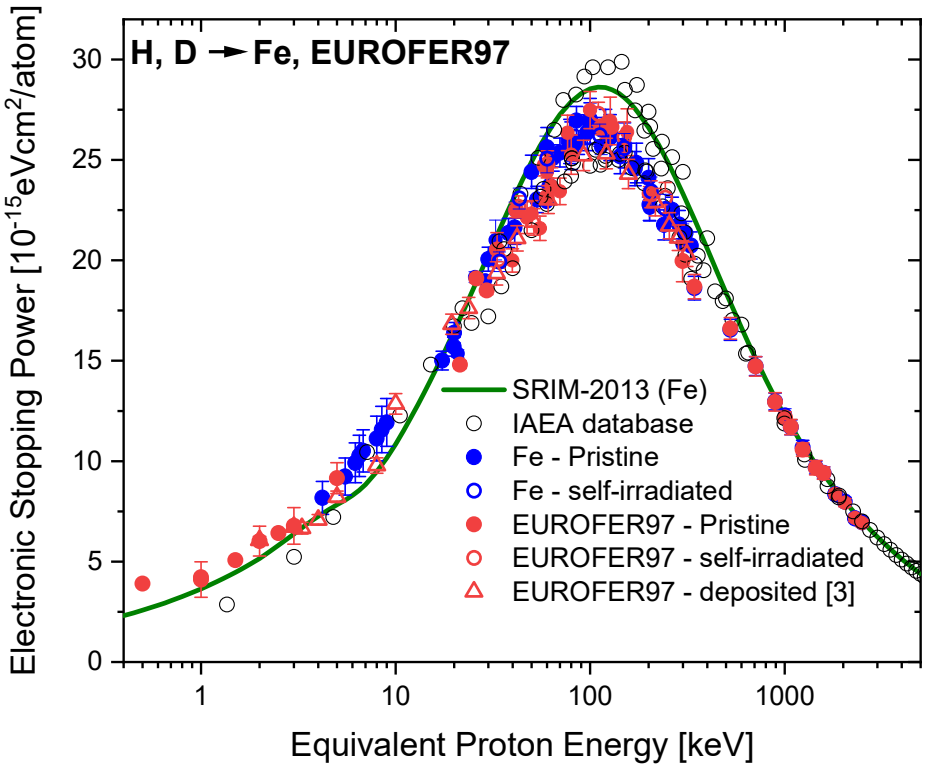
He^+ → Fe

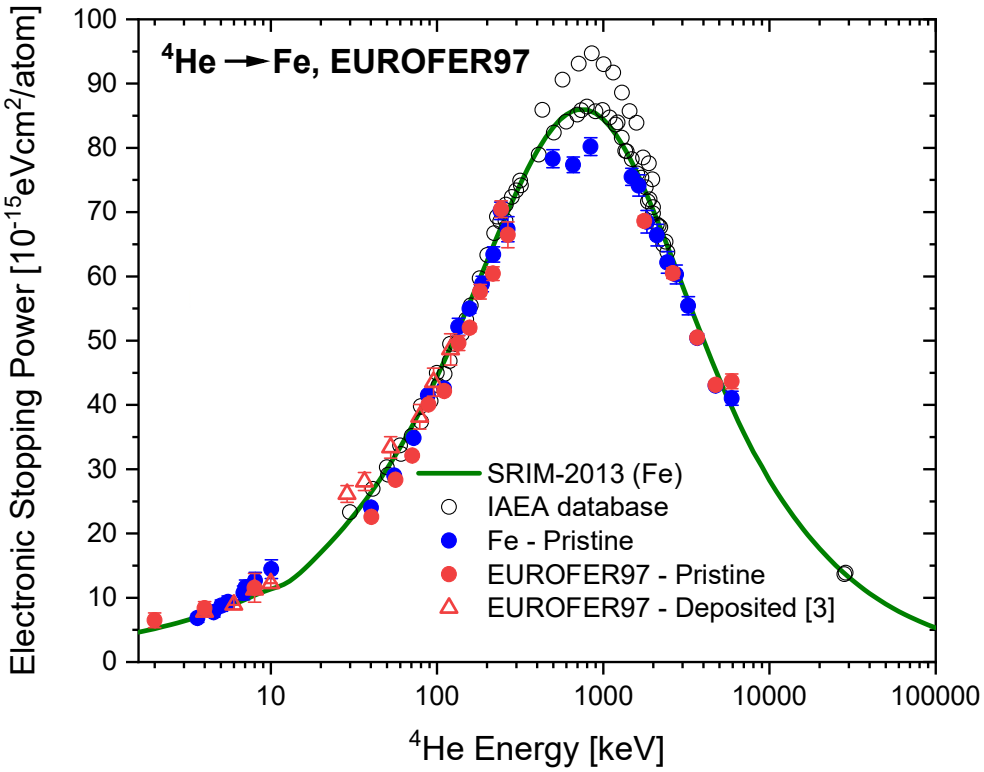
→ Good agreement at keV/MeV range within 6%.

→ LEIS: discrepancy up to 26%.

Electronic energy loss measurements

Experimental stopping cross-section of pristine Fe, W, and EUROFER97





H^+ → Fe, EUROFER97

Similar behaviour between Fe and EUROFER97

No significant discrepancy from Bragg's rule.

Electronic energy loss: TD-DFT simulations

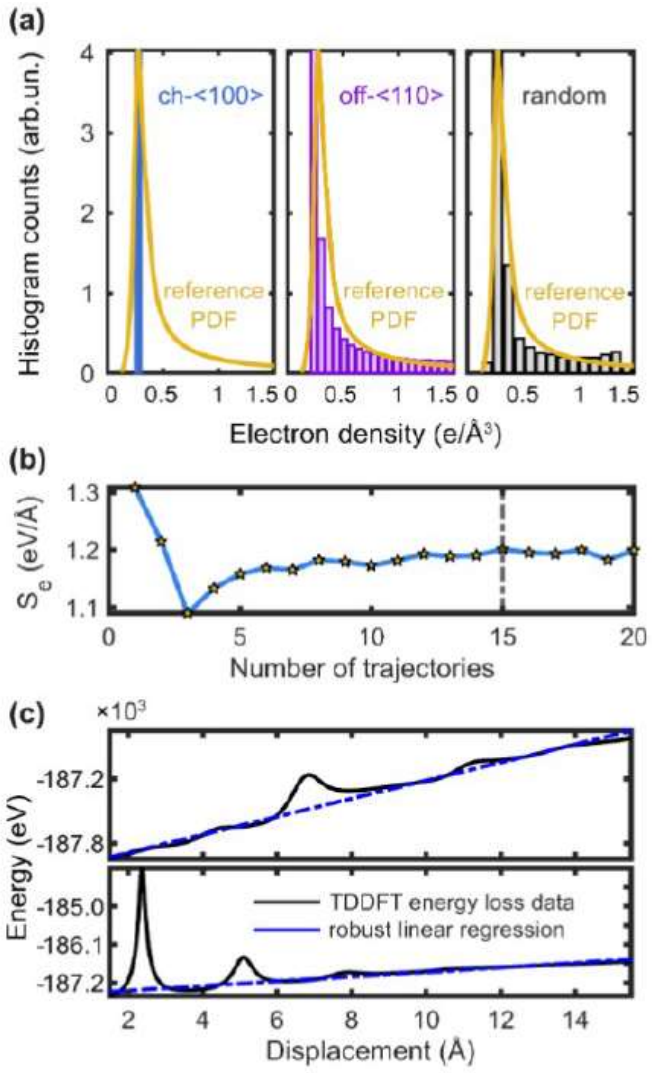
Theoretical stopping power of pristine W, Fe, and Fe-Cr

Theoretical calculation of electronic stopping power for random trajectories of light ions in pristine W, Fe and Fe-alloys using TD-DFT calculations.

Electronic stopping power from rate of change of total energy of the system, with ion travelling at constant velocity.

Simulation cell: 6x3x3 supercell (i.e. 108 W + 1 H atoms).
Qb@ll TDDFT code + LDA norm-conserving pseudopotentials.
lattice constant *a* with impact parameter *b*.

Method for selecting **random trajectories** sampling of different impact parameters aiming to capture all possible energy loss pathways between the ion and target atoms.



E. Ponomareva et al., Phys. Rev. B **109** (2024).

Electronic energy loss: TD-DFT simulations

Theoretical stopping power of pristine W, Fe, and Fe-Cr

Systematic investigation of the influence of **ion-target geometry** (channelling, off-centre, and random geometries) and **core-states description** (W12 and W 20) of the target in the electronic energy losses.

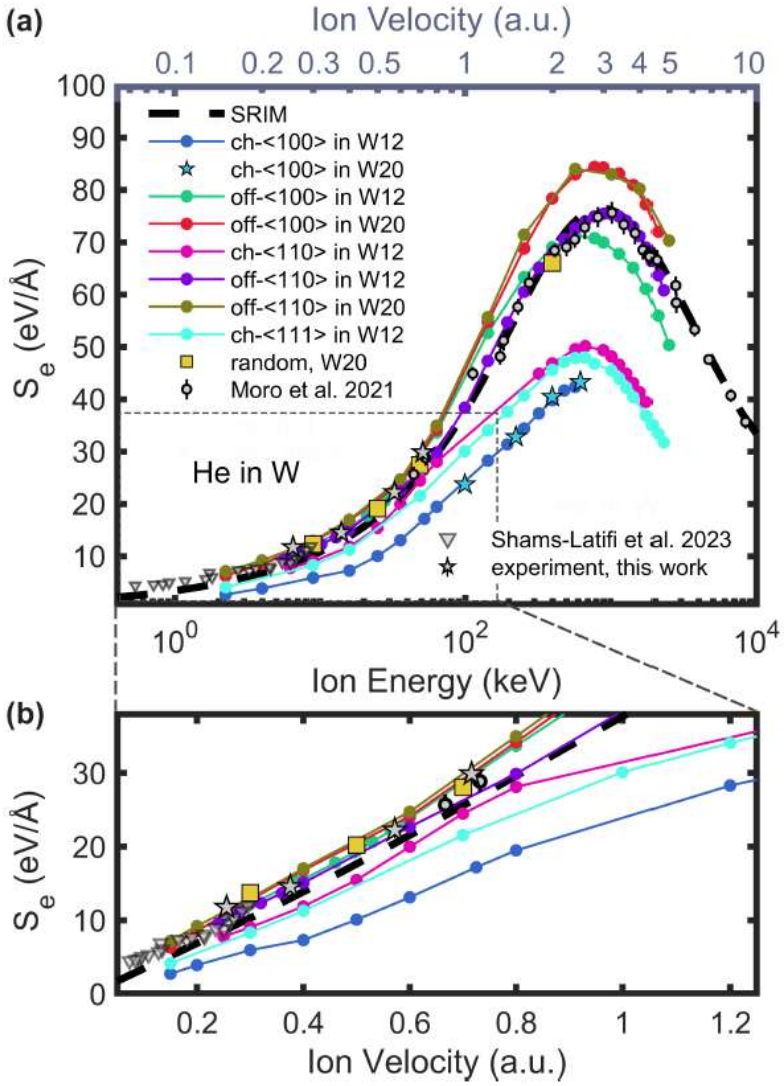
Hyperchanneling trajectories are significantly lower than the experimental data.

Good agreement with experimental data at lower energies.

Deviation from velocity proportionality for both projectiles traversing the hyperchanneling directions can be explained through the existence of a threshold velocity leading to the activation of semicore states.

Additional observations:
Vacancies and Cr in Fe-Cr found to have negligible effect on stopping power → in agreement with experimental observations.

E. Ponomareva et al., Phys. Rev. B **109** (2024).



Interatomic potentials

Motivation and Theoretical Background

At low energies, interactions with electrons cannot be neglected.

The screened Coulomb potential:

$$V(r) = \underbrace{\frac{Z_p Z_t e^2}{r}}_{\text{Coulomb potential}} \times \underbrace{\phi\left(\frac{r}{a}\right)}_{\text{Screening function}}$$

Correcting the screening length:

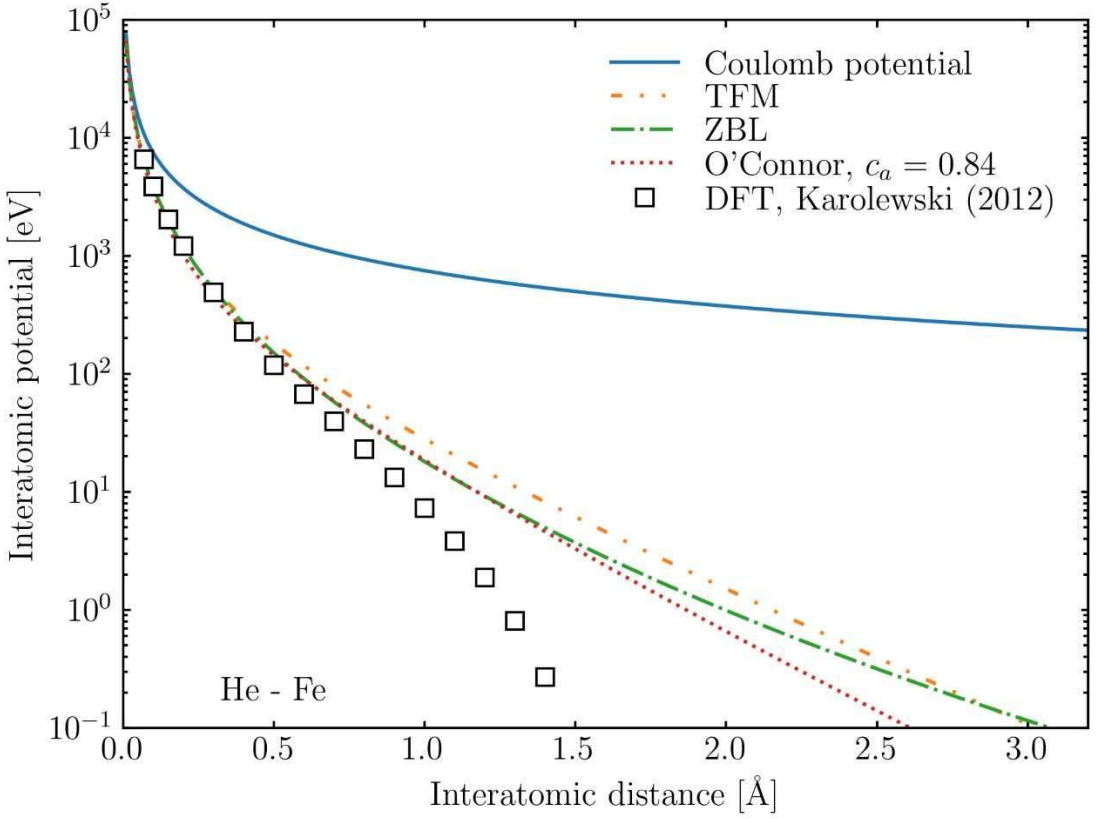
$$a = a_f \times C_a$$

a_f = Firsov screening length

C_a = Empirical correction factor

Common models (TFM and ZBL) are known to present inaccuracy.

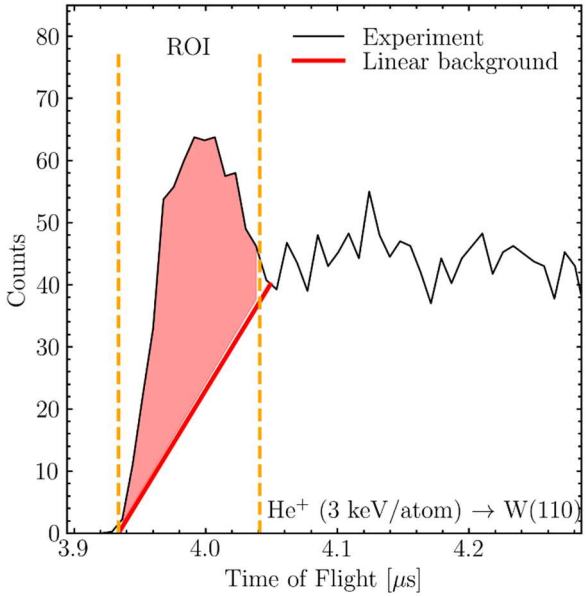
Lack of experimental reference data for many ion-solid combinations, to assess the quality of models for the interatomic potentials difficult.



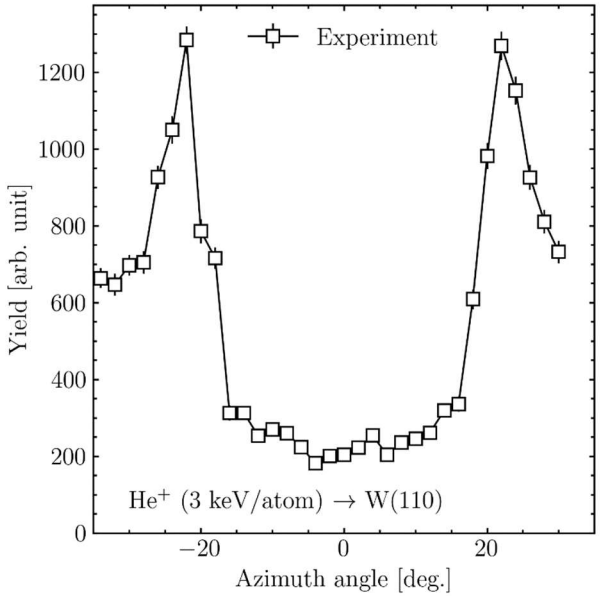
Interatomic potentials

Experimental approach

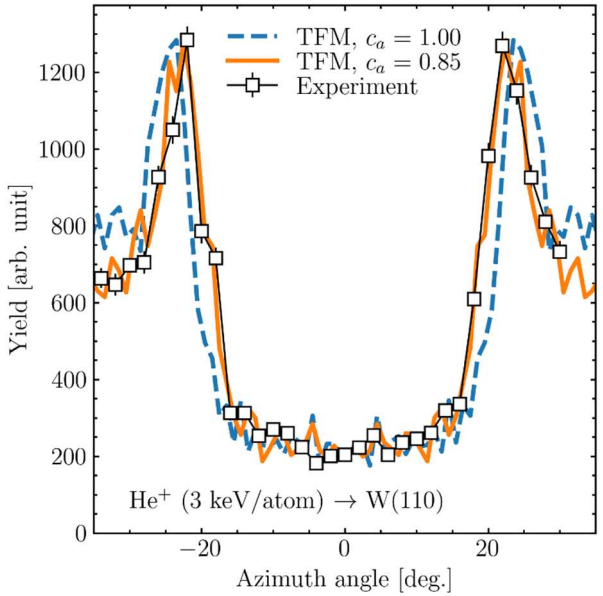
ToF-LEIS spectrum



Experimental angular scan

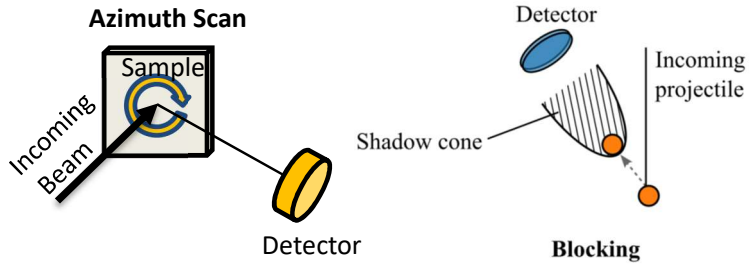


Fit with MD simulation



After sequential steps of surface cleaning (Ar sputtering and annealing) monitored with AES/LEED.

Background subtraction: multiple scattering from deeper layers.



Ions:
- Helium
- Deuterium

Single crystal surfaces:
- W(110)
- Fe(100)

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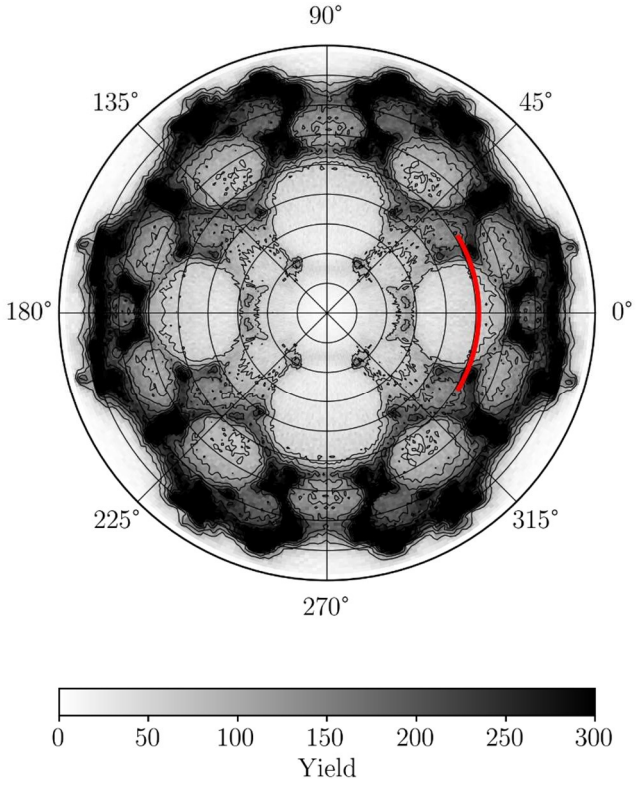
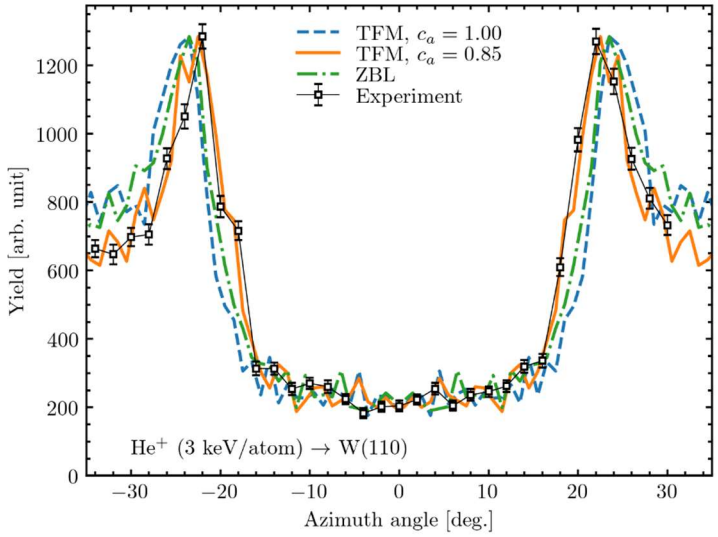
Interatomic potentials

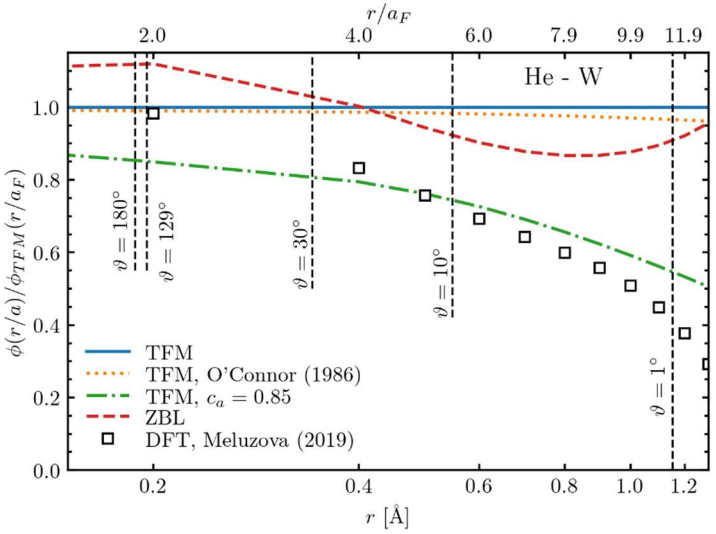
Results

Helium - Tungsten



Backscattering map
3 keV He from W(110)





- Scan crossing [010] crystal axis of the (110) surface
- Best agreement for TFM with c_a=*0.85*
- TFM, ZBL, and O'Connor predict higher values
- Good agreement with DFT for relevant angles

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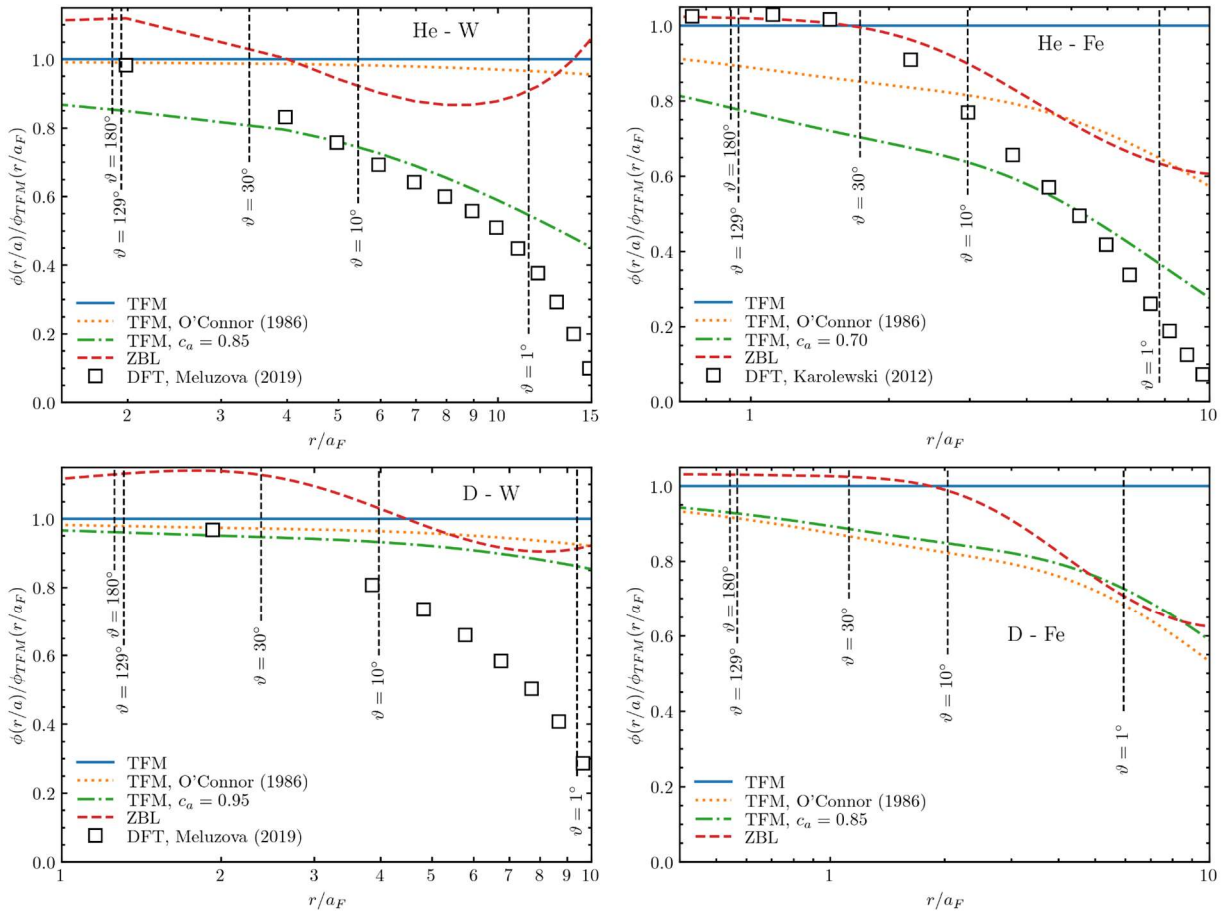
Interatomic potentials

Overview of main Results

Ion	c_a (TFM)	
	W(110)	Fe(100)
He	0.85	0.7
D_2	0.95	≥0.85

- TFM, ZBL and O'Connor deliver better results for cases involving D than He.
- TFM consistently overestimates the interatomic potential.
- O'Connor correction improves the situation but still overestimates the potential.
- ZBL model is better than TFM but still overestimates the potential.
- DFT data agrees well with the experiment for He cases.



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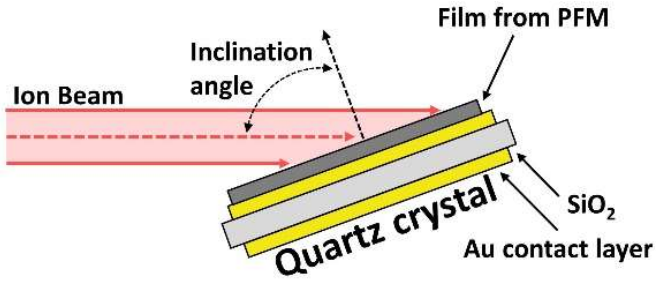
Sputtering Yields

Experimental appraoch using high-sensitivity quartz crystal microbalance QCM

Experimental configuration

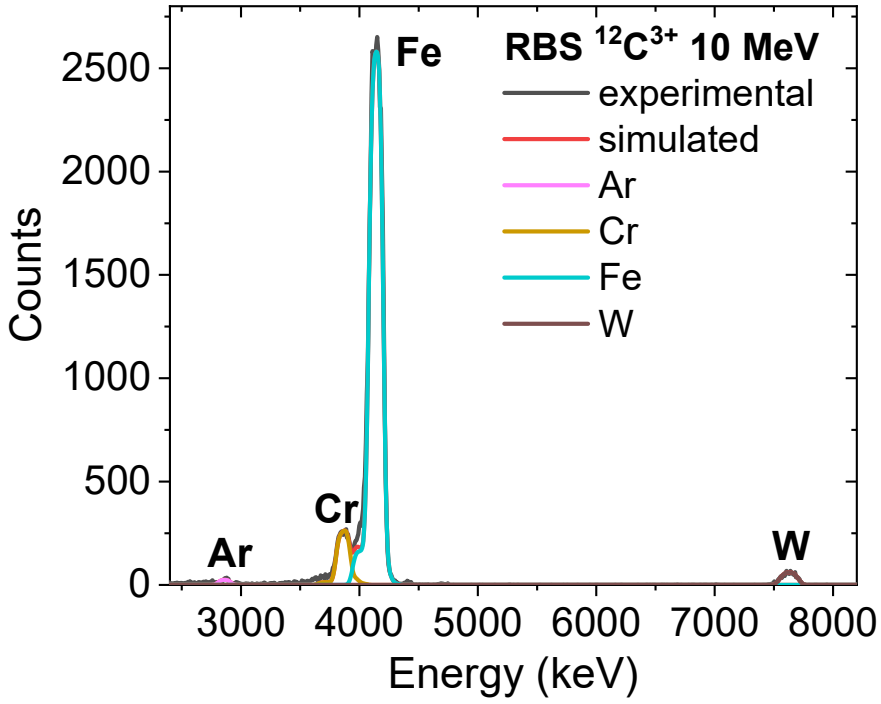


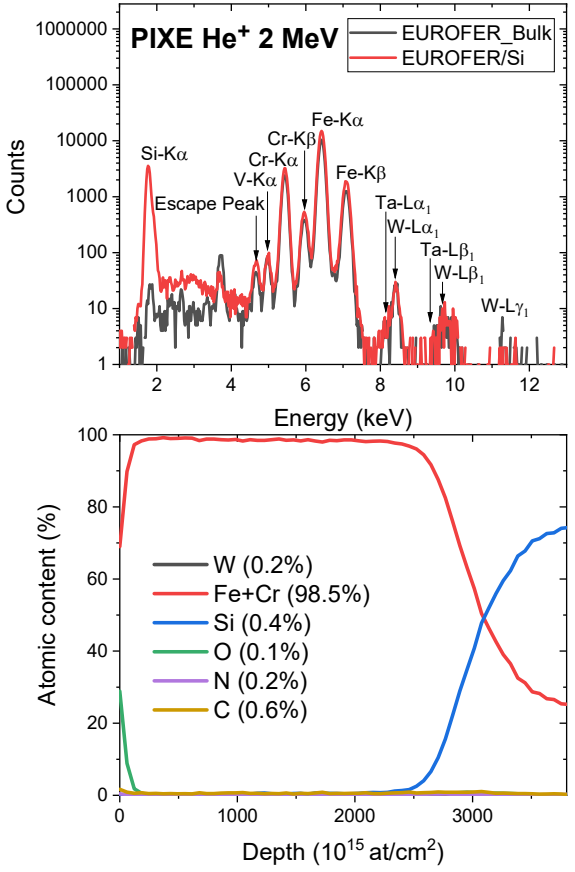
From SIMNRA: Similar composition to bulk.

	At. content (%)	
	Sputtering	Bulk (nominal)
Fe	88.7	88.9
Cr	11.0	9.5
W	0.3	0.3

[1] E. Pitthan, P. Petersson et al. Nucl. Mater. Energy 34 (2023) 101375.

Formation and characterization of thin films from EUROFER97 target [1]:



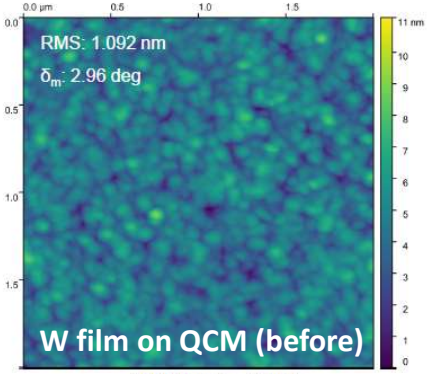


Sputtering Yields

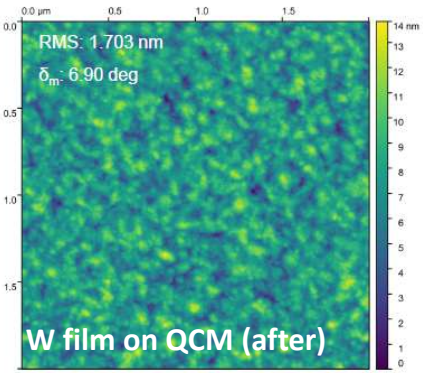
Results

→ **AFM measurements before/after measurements:**

- Typical RMS<1.8 nm for films;
- homogeneous film growth;
- no significant roughening/smoothing of surface morphology observed after measurements.



AFM images - Center

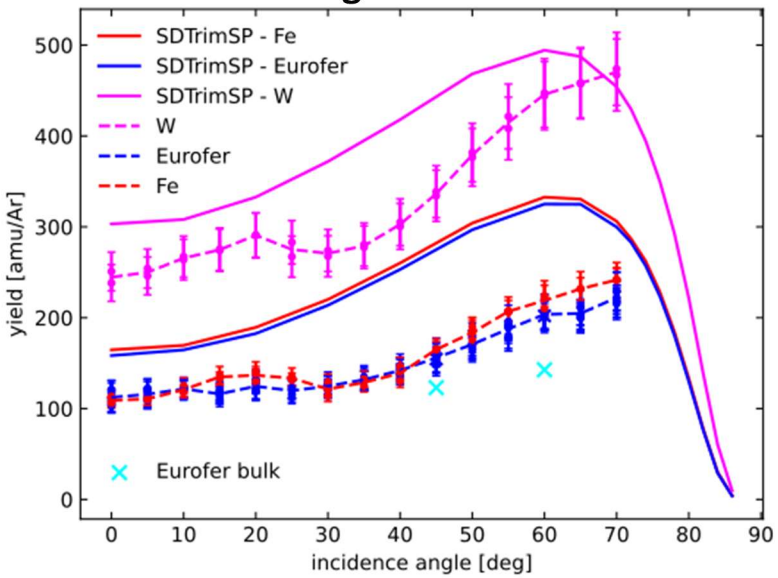


AFM images - Center

→ **Sputtering yield measurements Ar^+, D_2^+, He^+ bombardment of PFC thin films in different incident angles.**

→ **Bulk (catcher mode) and self-irradiated layers also measured.**

Irradiation using 2 keV Ar^+ ions



- Poor agreement with default SDTrimSP in all investigated cases (Ar/D on W, Fe and Eurofer).
- **Investigate effect of different parameter adjustments in BCA code simulations.**

Additional observations:

- No significant change in self-irradiated samples.
- Smaller SY from EUROFER bulk attributed to roughness.

Sputtering Yields

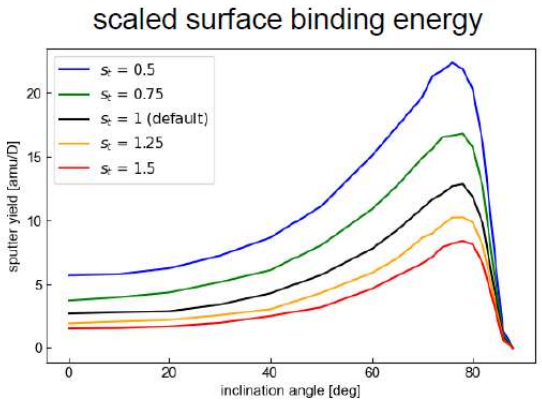
Effect of nuclear data corrections in sputtering yield simulations.

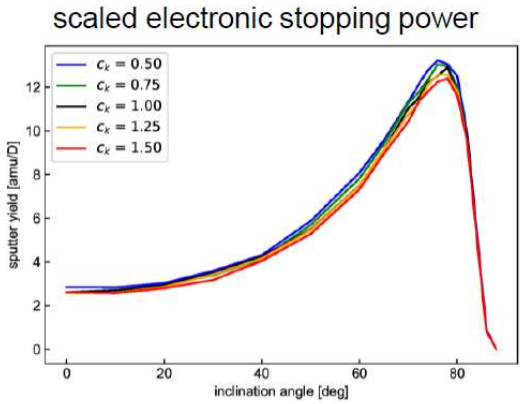
Systematic investigation of effect of nuclear data corrections in sputtering yield simulations.

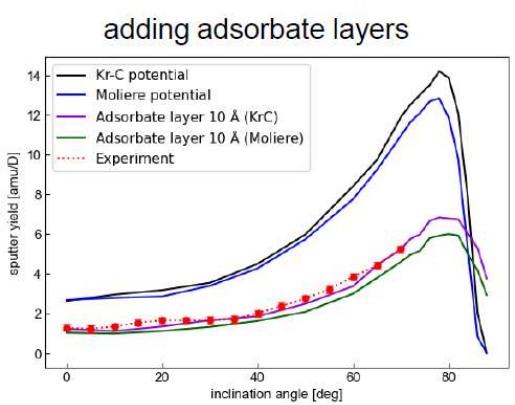
SDTrimSP (BCA-based) with Graphical User Interface [1]: 1 keV D on Eurofer, Lindhard-Scharff stopping model, Moliere interaction potential.

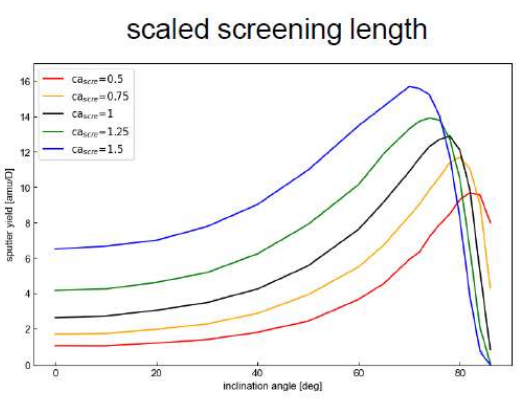
Scaling electronic stopping power;
Surface binding energy;
Adsorbate layer thickness;
→ global offset in sputtering yields

Screening length in interatomic potential.
→ offset in sputtering yields
→ Shifted maximum in yields over angle curve









[1] P. S. Szabo et al. Nucl. Instrum. Methods Phys. Res. B 522 (2022) 47-53.

Sputtering Yields

Effect of nuclear data corrections in sputtering yield simulations.

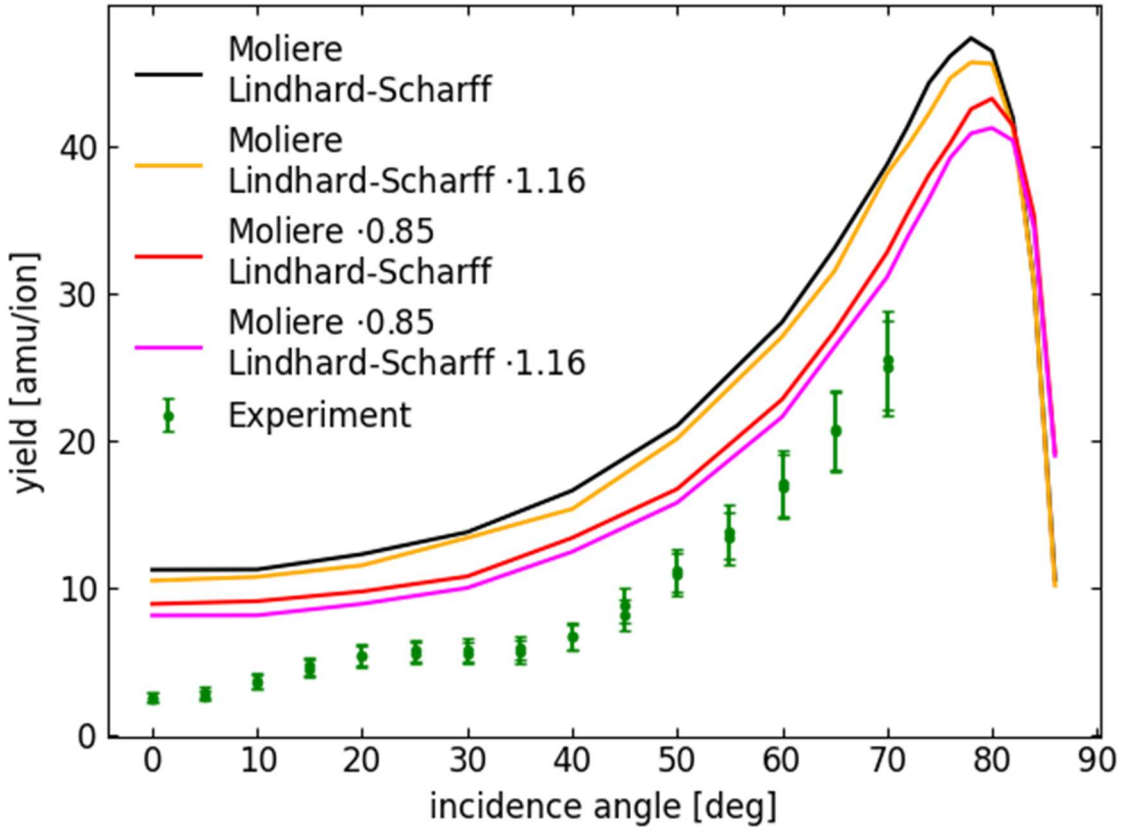
Applying energy loss and screening length experimental corrections obtained experimentally in BCA simulations and comparison with experimental sputtering yields.

He 3 keV → W/QCM

Electronic energy loss model:
Lindhard-Scharff (equivalent to SRIM-13)
Empirical correction × 1.13
J. Shams-Latifi, et al., Nucl. Mater. and Energy 36 (2023).

Interatomic potential model:
Thomas-Fermi-Moliere potential (TFM)
Screening length correction × 0.85
P. M. Wolf, E. Pitthan, D. Primetzhofer, submitted for publication (Pinboard 38569)

Both corrections lead to a reduction of the sputter yield, going into the direction of the experimental values.



Sputtering Yields

Effect of surface orientation on sputtering yields

Experimental observations (TU Wien):

- local minimum in yield over angle curve at around

35 deg incidence angle observed for different measurements for W and Fe layers.

- Possibility of texture and crystallinity effects

from preferential grain orientation.

- Detailed theoretical investigation of crystal orientation effect.

Sputtering with MD simulations (Aalto):

- Effect of surface orientation.
- Effect of surface roughness (step defect).
- Fitting of ion-electron coupling for two temperature

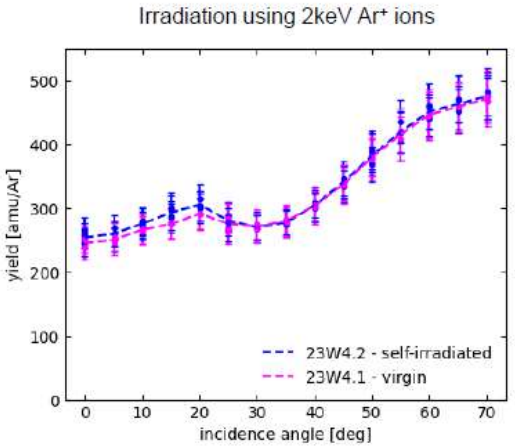
model simulations with environmentally dependent stopping power.

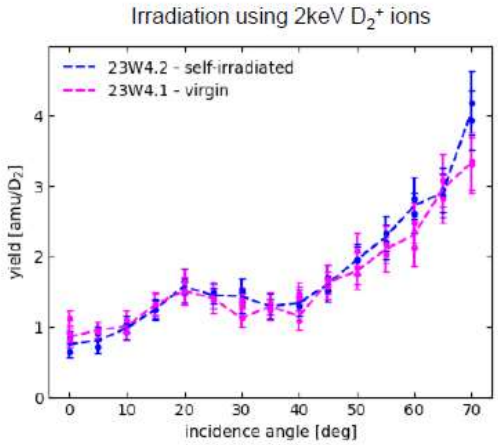
Main observations:

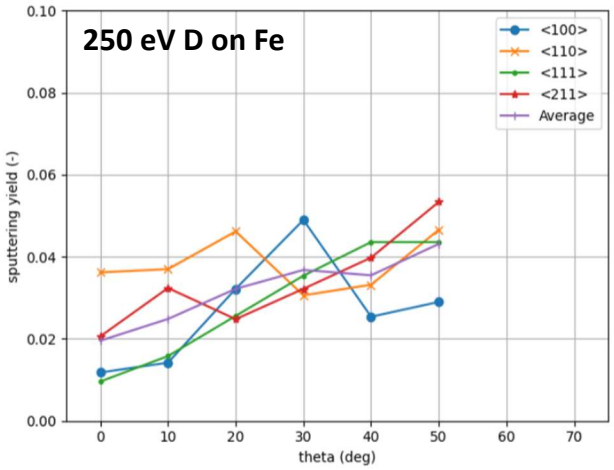
→Dependence on surface orientation (publication under preparation with TUWien).

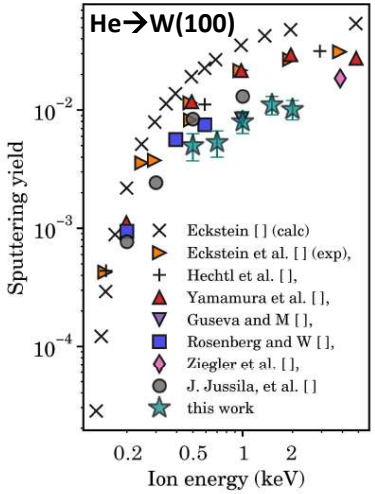
→ Negligible effect of unevenness of surface (step-defect).

→ Electronic stopping removes the energy available for sputtering process; all MD-EPH simulations reveal lower yields (under preparation for publication).





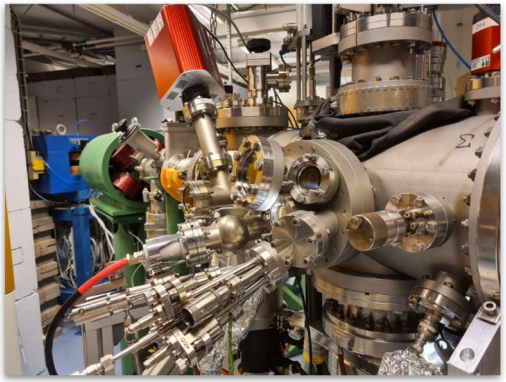


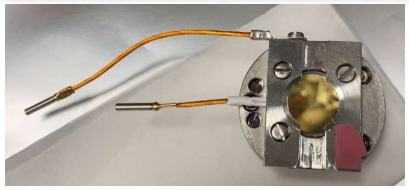


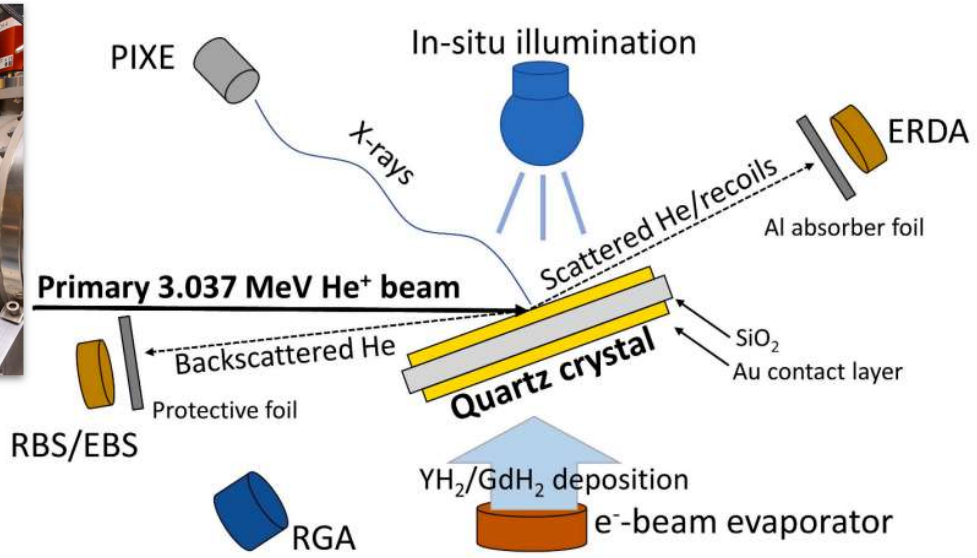
Sputtering Yields

Implementation of new experimental set-up for in-situ studies

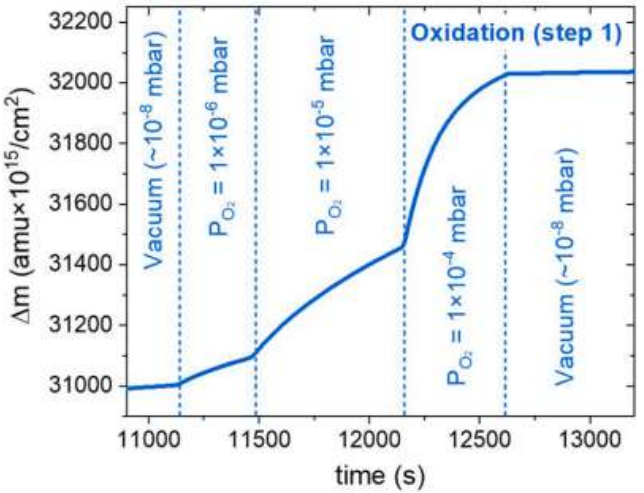
In-situ and real-time mass-change measurements (QCM) and subsequent IBA (RBS, PIXE, ERDA).



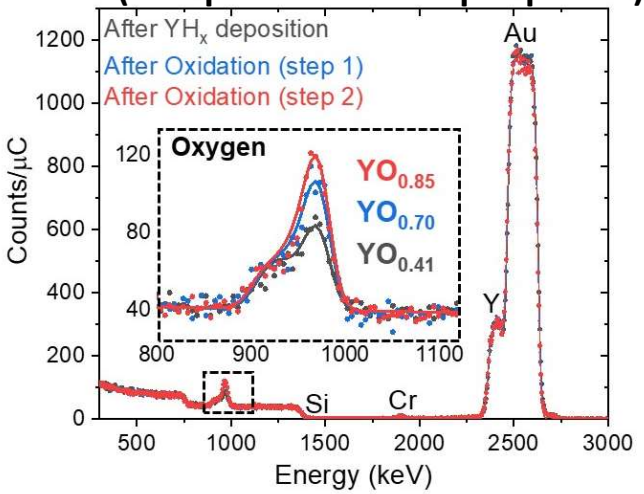




QCM (mass changes monitor)



IBA (Composition and depth profile)



Allows in-situ implantation/erosion/deposition/oxidation investigations simultaneous to material characterization by IBA.

Benchmarking: investigation of formation and oxidation of oxygen-containing rare-earth hydrides

E. Pitthan et al. Materialia 27 (2023) 101675.

Overview

Main scientific achievements

Energy losses

- First experimental data for energy losses of light ions (H, D, and He) in PFC (W, Fe & EUROFER97) at low energies:
- Pronounced discrepancies between semi-empirical models (SRIM-2013) were identified: up to 60% for He in W.
- Detailed TD-DFT calculations (crystal orientation and core-states description) of electronic energy losses for W, Fe, and Fe-Cr: strong crystalline influence and good agreement with previous experimental observations for random geometries.
- No significant effect of defects in experiments and simulations.

Interatomic potentials

- First experimental measurements of screening length correction for W and Fe and compared to available models:
- Allow assess the quality of models for the interatomic potentials (overall good agreement with recent DFT models).

Sputtering yields

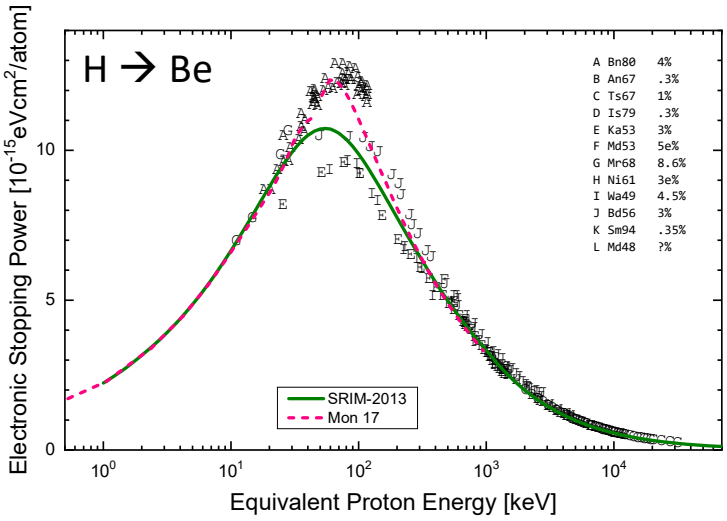
- New experimental sputtering yields angular dependence for PFC (W, Fe, and re-deposited EUROFER97).
- Detailed investigation of effect of fundamental nuclear data uncertainties in sputtering yield simulations.
- Molecular Dynamic (MD) simulations dependence of crystalline orientation, defects, and electronic effects in SY.
- Integration of real-time mass-change measurements (QCM) in in-situ IBA system.

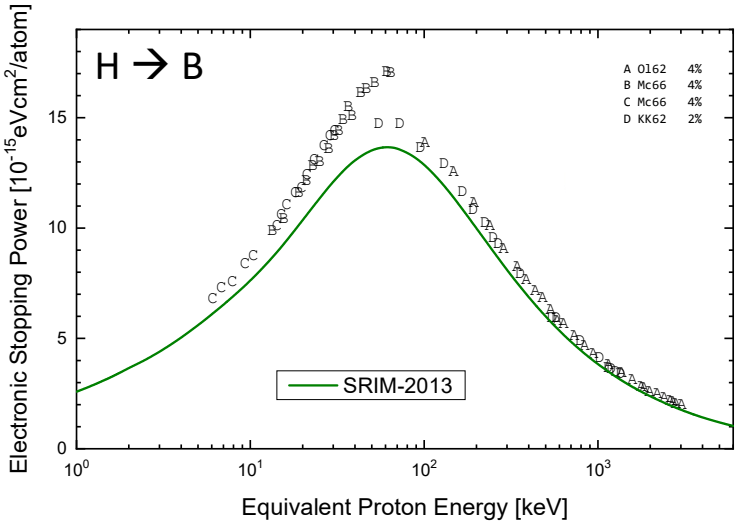
Overview

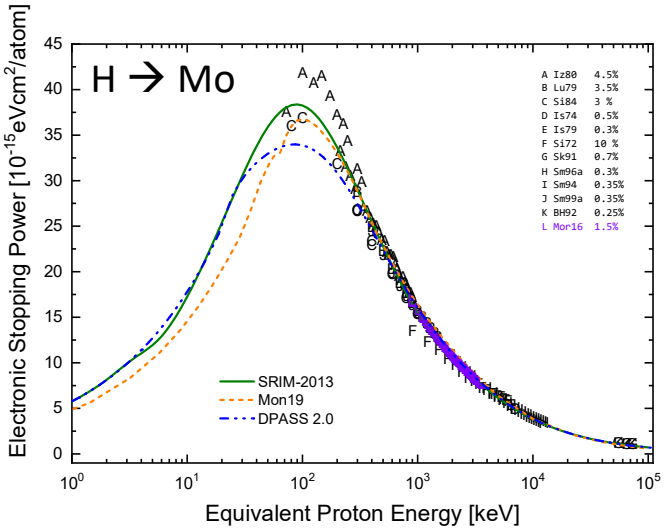
Recommendations:

- To use updated data-sets for fundamental quantities (data is already available in IAEA-most cases).
- Awareness of uncertainties in different combination of ion-matter and potential impact in sputtering yields.

Other examples related to fusion materials







Be, B, and Mo: Limited data at low energies and high discrepancy at higher energies.

Formation of oxides can lead to larger discrepancies from brags rule: specific studies for compounds are needed.

More work is needed on the determination and evaluation of nuclear data for elements and isotopes with relevance for fusion.

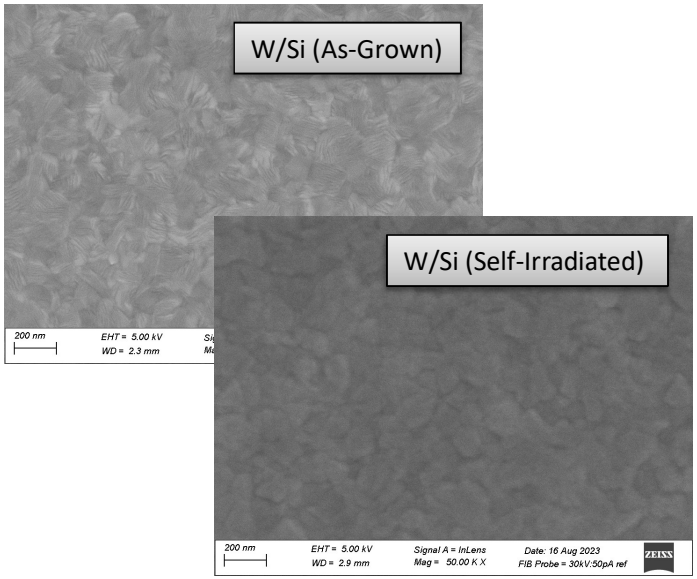
Thank you!

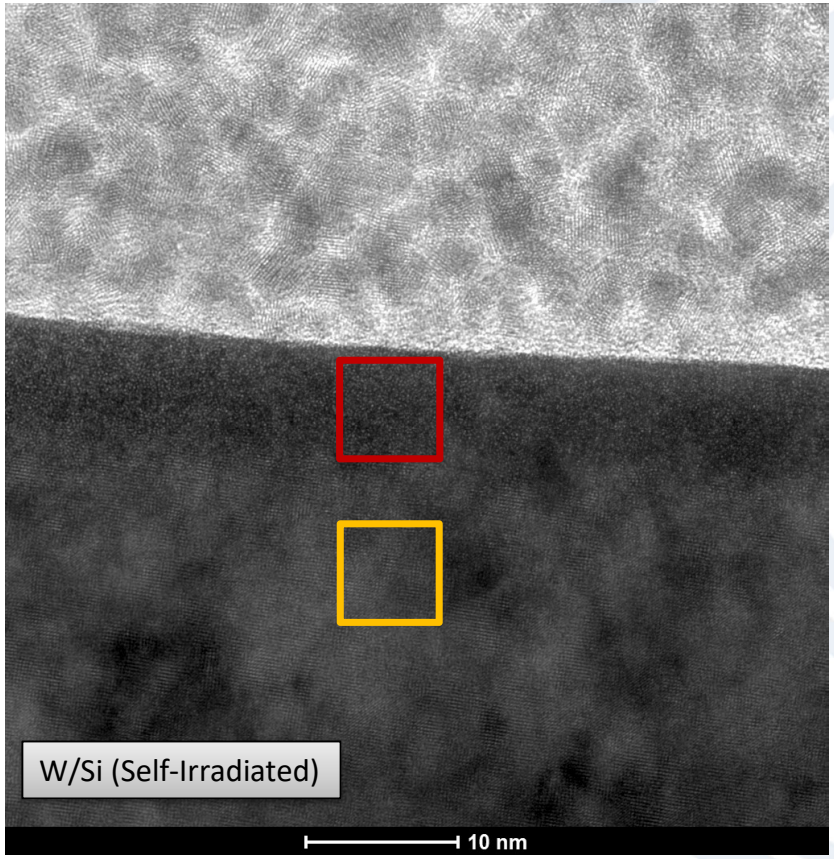
This work has been carried out within the framework of the EUROfusion Consortium, funded by the European Union via the Euratom Research and Training Programme (Grant Agreement No 101052200 — EUROfusion). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

Extras

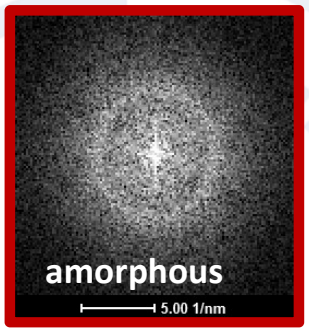
Ion-irradiation experiments

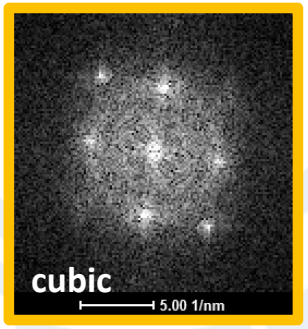
Experimental (preparation for WP-3 and 5)
Sample: W(300 nm)/(Si, C, QCM)
Implantation: W^{2+} 600 keV
Fluence: 1.5×10^{15} atoms/cm^2
SEM: *Smoothening of surface after Implantation.*
TEM: *amorphization of surface.*





FFT:





Electronic energy loss measurements

Experimental procedure for low energy regime

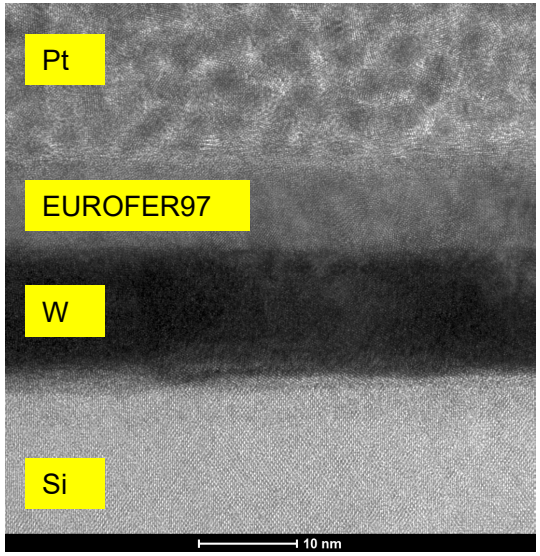
Complementary approach (marker layer):

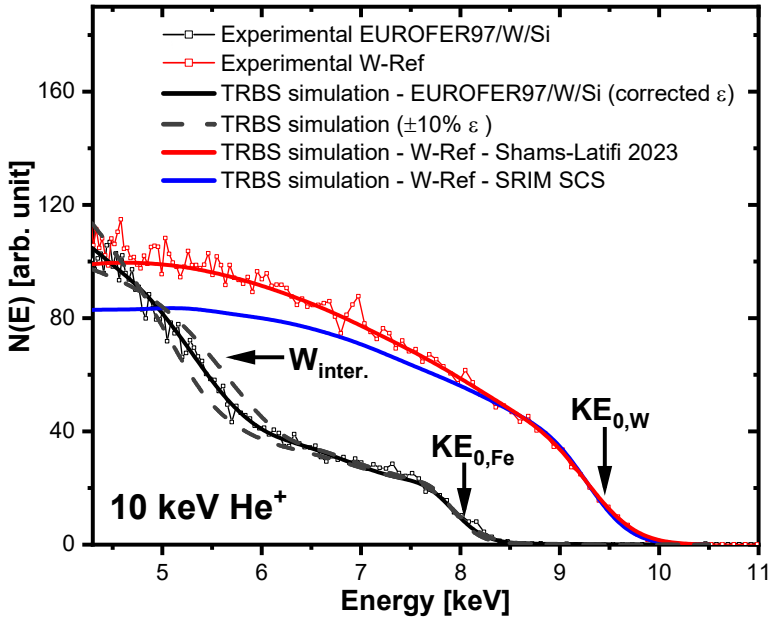
Experimental stopping cross-section of sputter-deposited EUROFER97 for LEIS

→ Analysis from spectrum height or spectrum width unfavorable for compound systems and lighter species.

→ Surface interface segregation is a risk – best to look on the signal indirectly – via a marker layer.

Marker layer:





Characterization of re-deposited PFM structures (pinboard 36144):
J. Shams-Latifi, E. Pitthan, et. al Mater. Res Express. 11 (2024) 016518.