SAND2020-8909PE

Characterizing hydrogen adsorption sites on a tungsten single crystal

PRESENTED BY

Tim Wong | Aug. 27, 2020 | 8351

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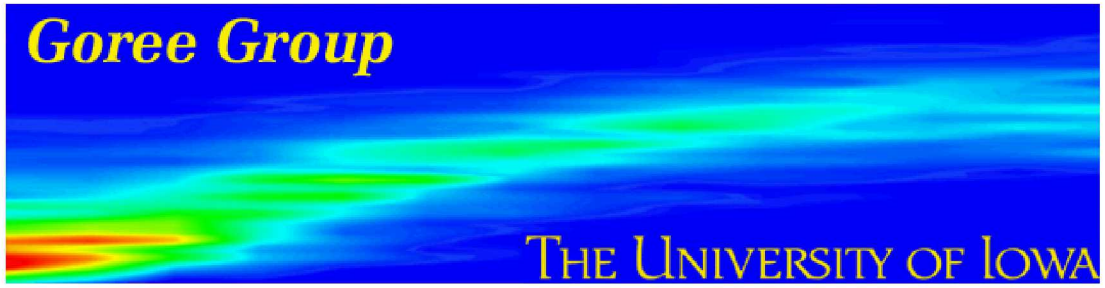
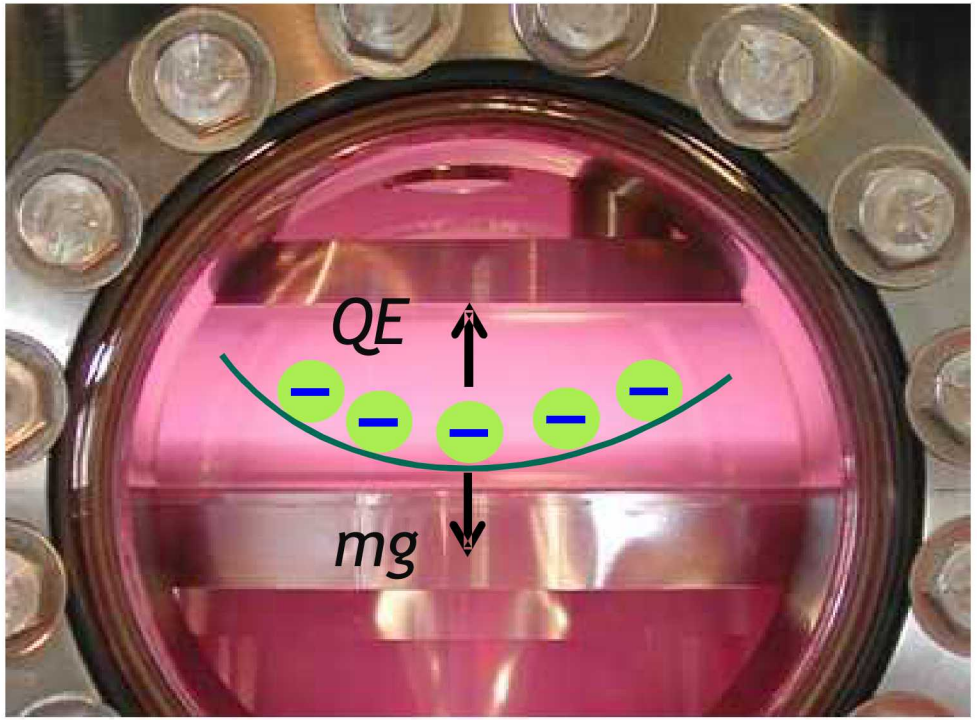
University of Iowa: dusty plasma physics





Extreme conditions in a fusion reactor

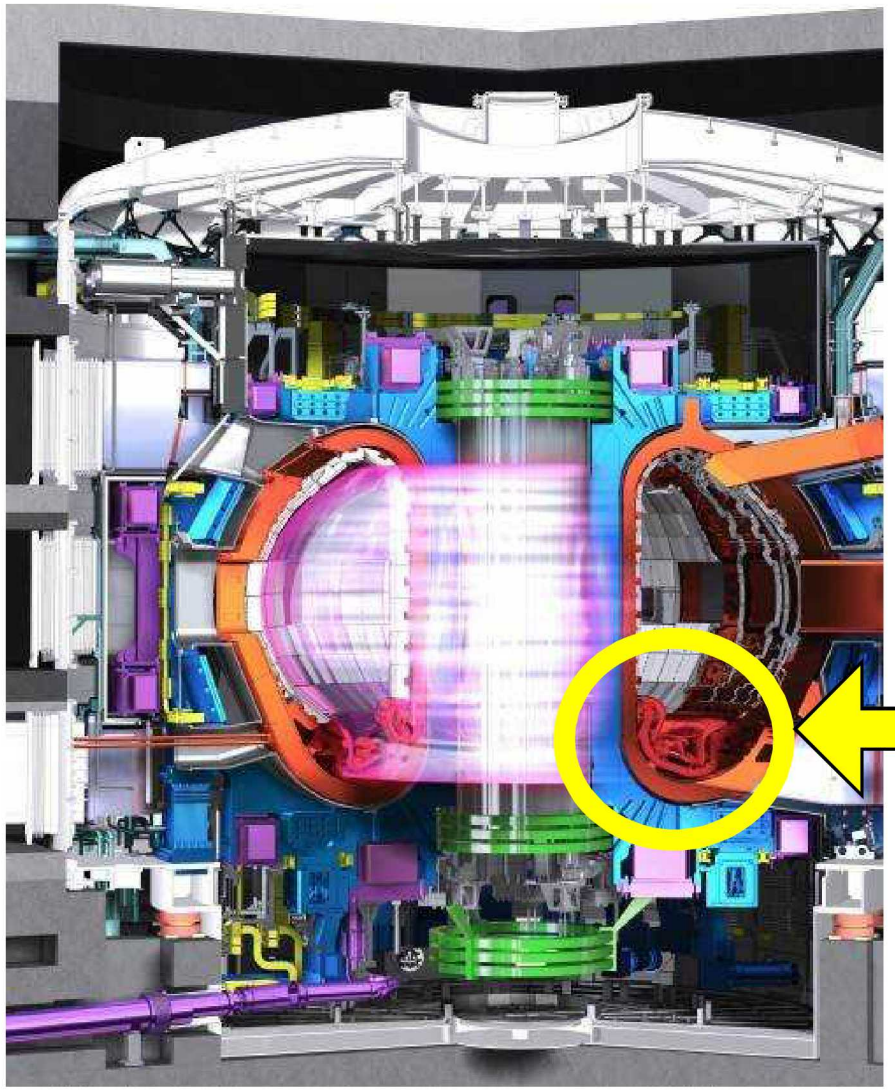


Image: iter.org

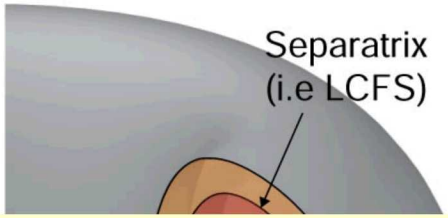
Fusion reaction:

$D + T \rightarrow He + n^0$ (14.1 MeV)

requires confinement of extremely hot (>100,000,000 °C)

and dense plasma

achieve confinement with magnetic fields



Divertor: where heat and particle flux (D, T, He, & impurities) are intentionally dumped

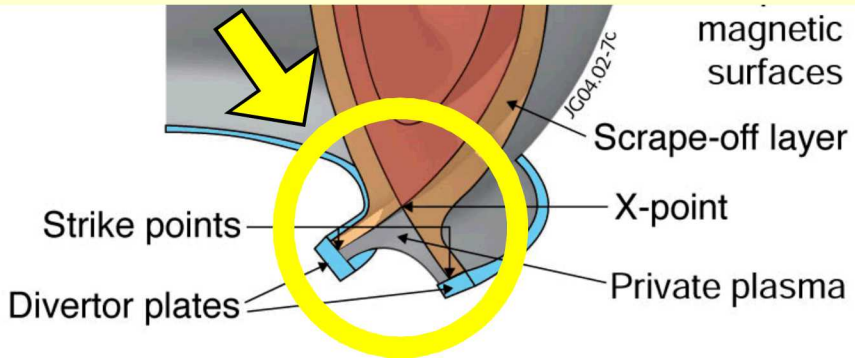


Image: euro-fusion.org

Divertor materials

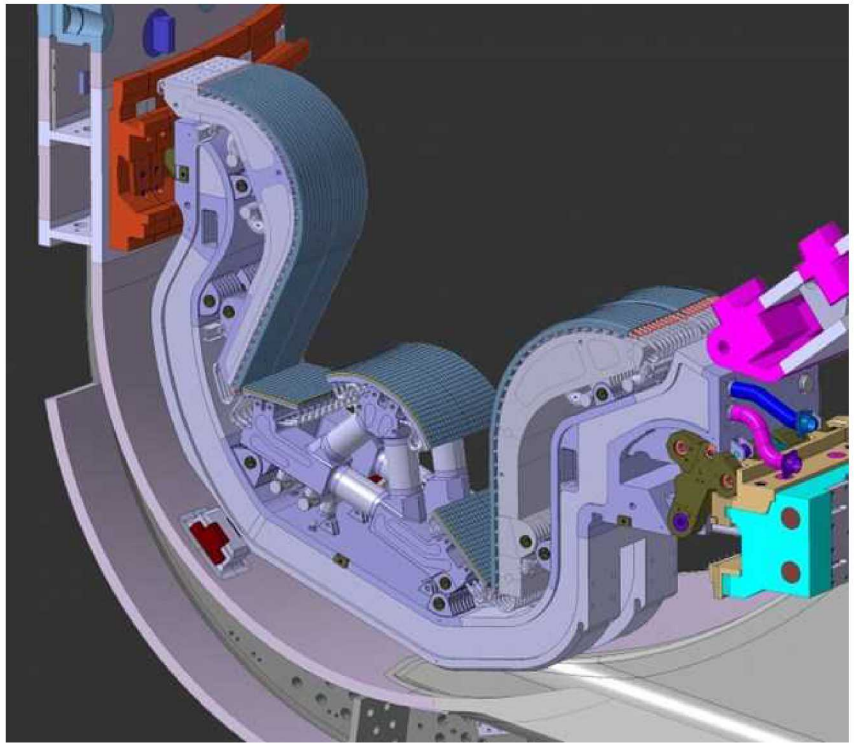


Image: iter.org

tungsten is a leading candidate material

- highest melting point
- low sputter yield
- high thermal conductivity

hydrogen effects on tungsten surfaces

- material degradation
  - embrittlement
  - blistering
- tritium retention and inventory
  - one 5 min shot with ITER ~ 10^5 Ci of T
  - 10 shots in 1 day = 10^6 Ci of T
  - ~10 Ci into environment—track ~10 ppm!
  - 5 μL of T_2O is lethal

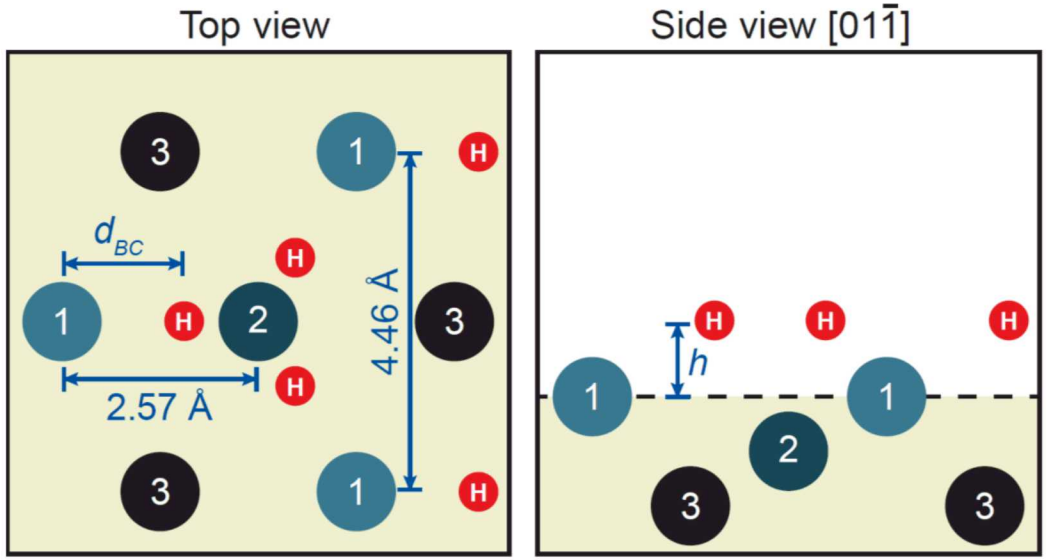
DFT can provide insight into hydrogen-tungsten interactions

Bond centered (BC) site on W(111) predicted using density functional theory (DFT) by our collaborators: Zack Bergstrom and Brian Wirth at University of Tennessee

Experimentally validate DFT results:

DFT provides inputs for larger scale models

- interatomic potentials
- dissociation & recombination of H



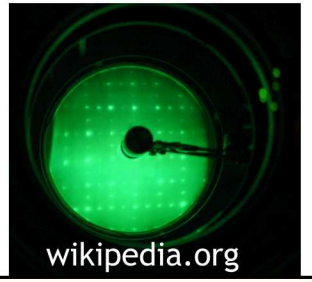
1.5 Å ≤ d_{BC} ≤ 1.7 Å

0.95 Å ≤ h ≤ 1.10 Å

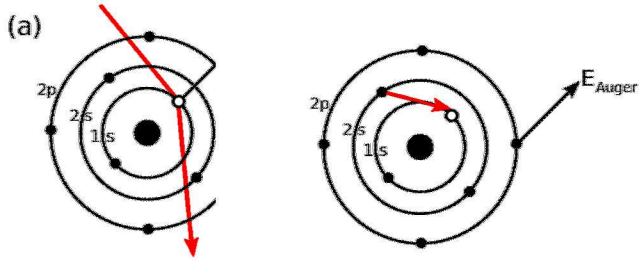
Z. Bergstrom et al., J. Phys. Condens. Matter, (2019).

Challenges for detecting surface H

LEED



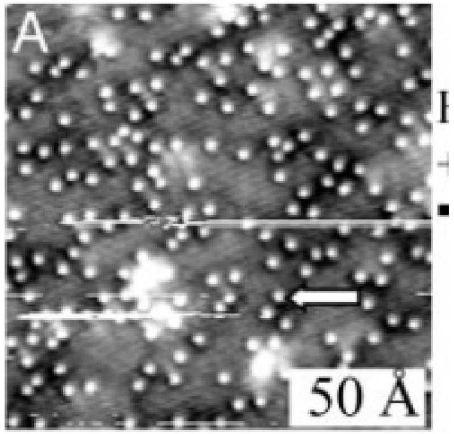
AES



Direct detection of surface H is challenging:

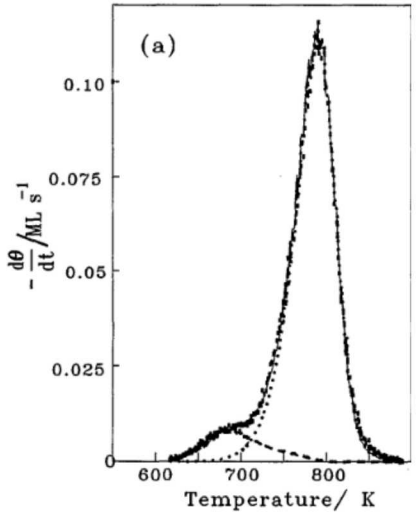
- Direct detection impossible for many techniques (XPS, AES)
- Challenging to disentangle H signal from substrate (LEED, STM, HAS)
- Ambiguous surface/location information (TPD)

STM



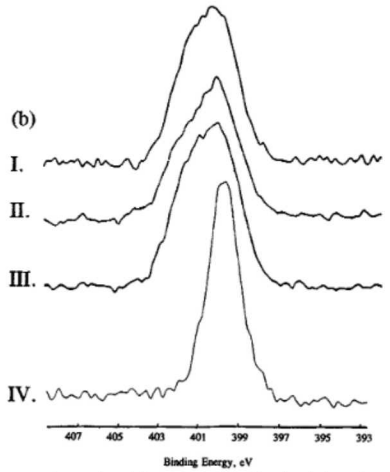
Sykes et al., PNAS (2005).

TPD



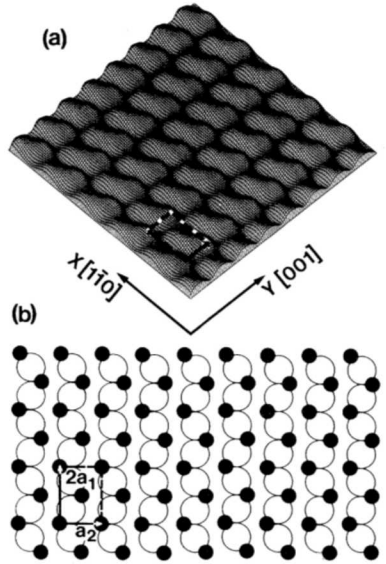
Flowers et al., J. Chem. Phys. (1993).

XPS



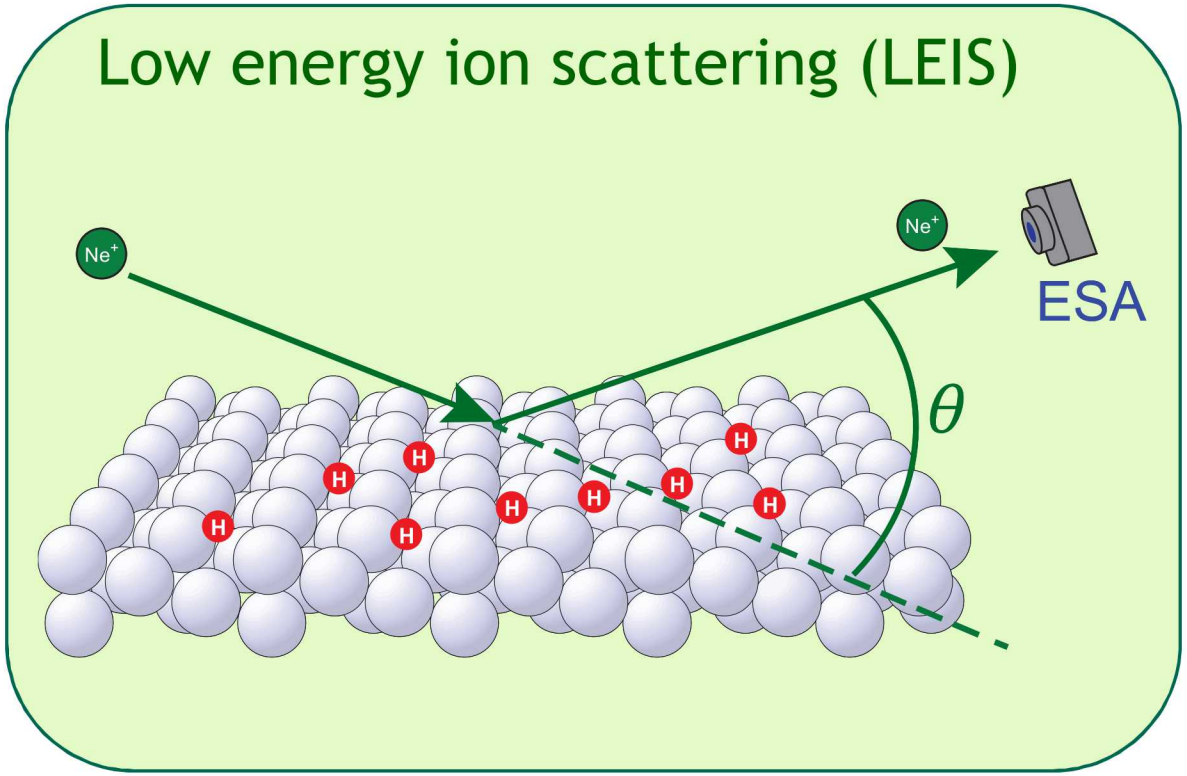
Kerber et al., J. Vacuum Sci. Tech. (1996).

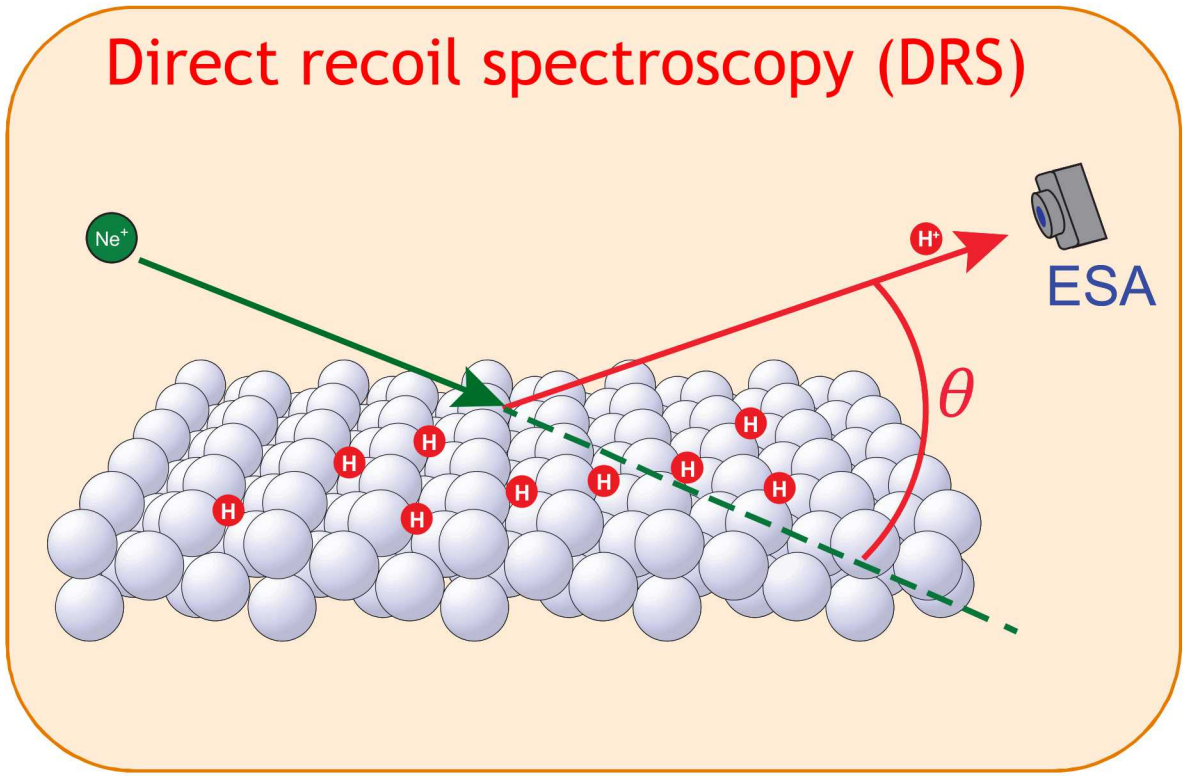
HAS



Rieder and Engel, PRL (1980)

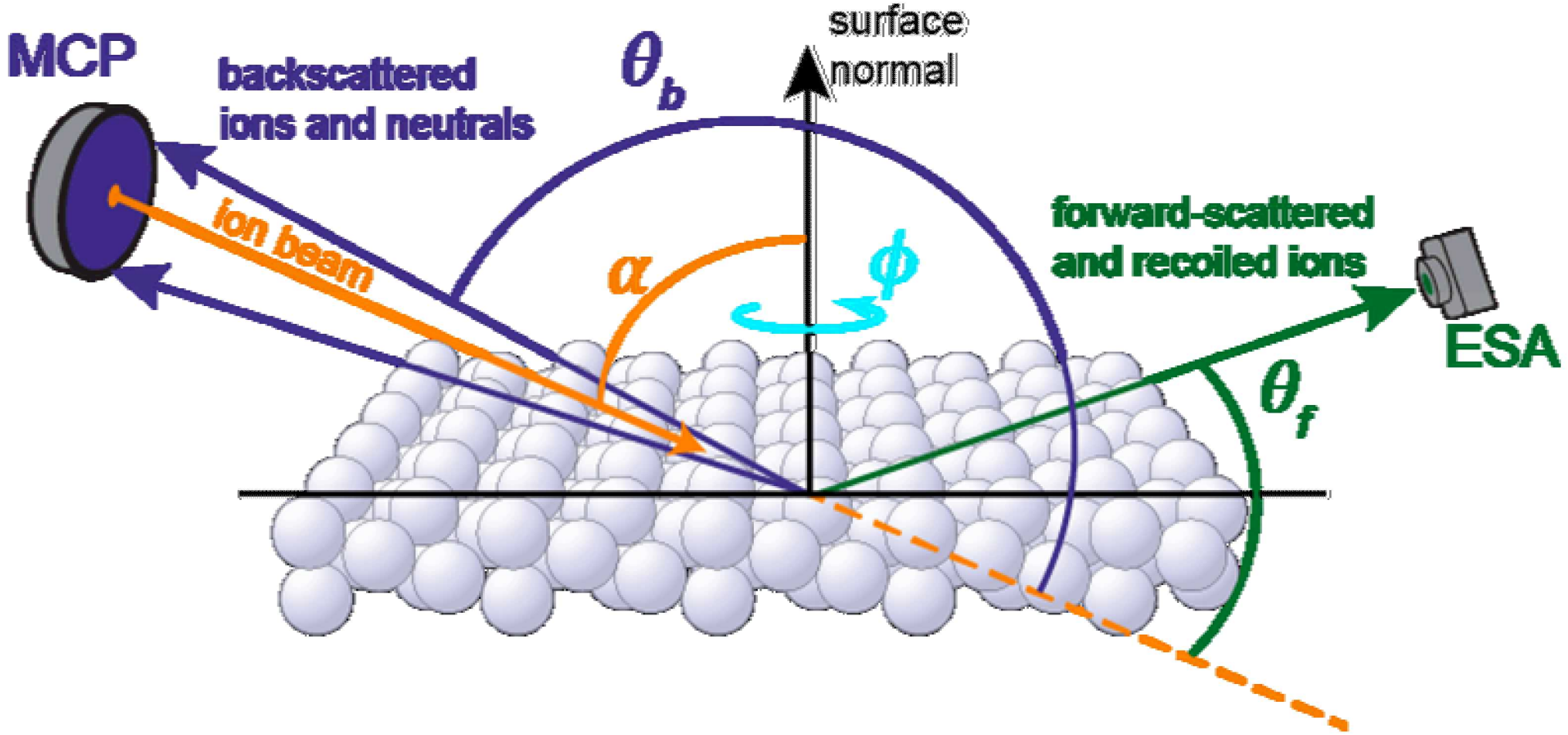
LEIS and DRS for H detection



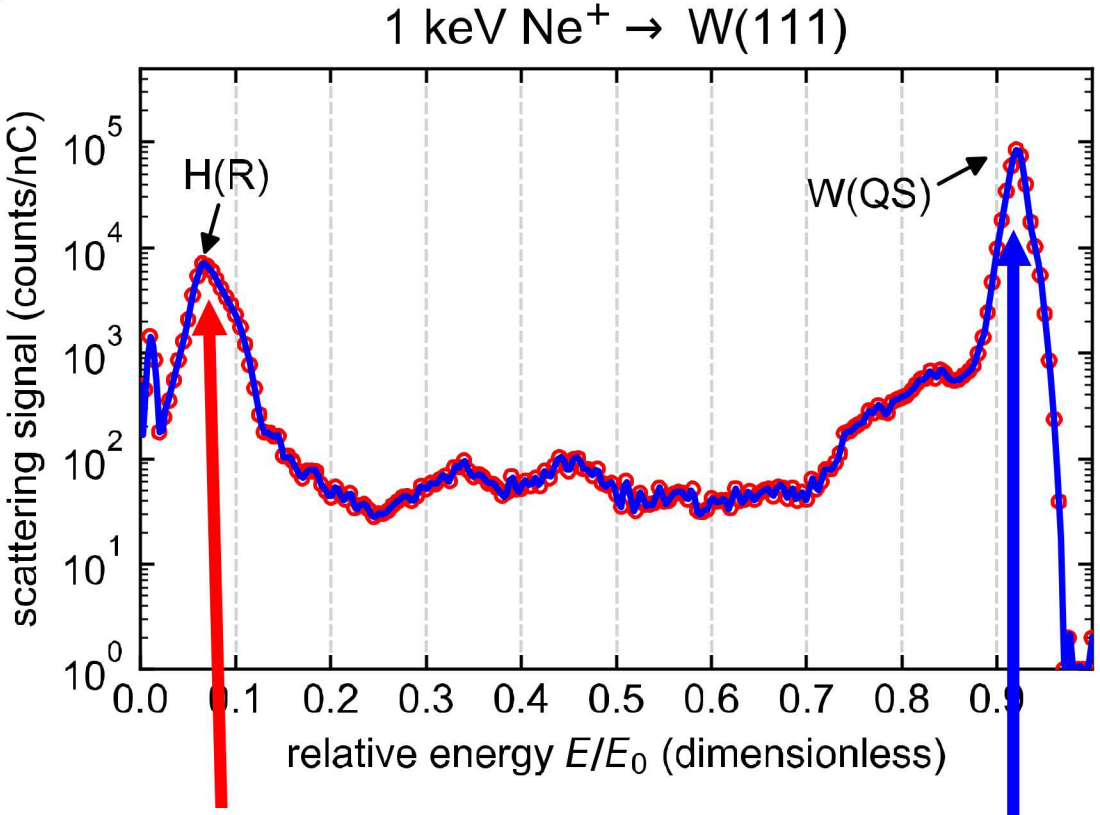


- both techniques are performed simultaneously
- energy of detected ion gives compositional information
- DRS ➔ direct detection of surface hydrogen

ARIES: Angle-Resolved Ion Energy Spectrometer



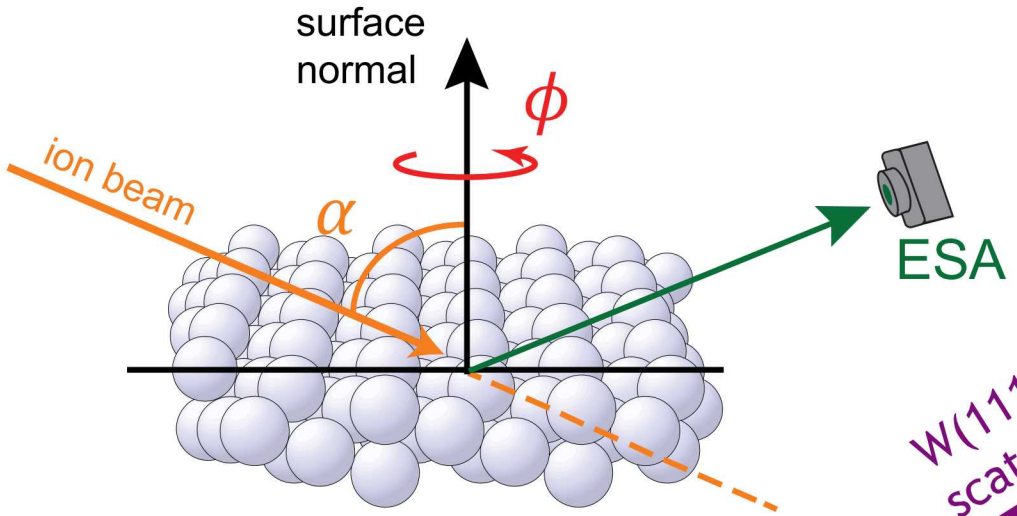
Ion energy spectrum for 1 keV $Ne^+ \rightarrow W(111)$



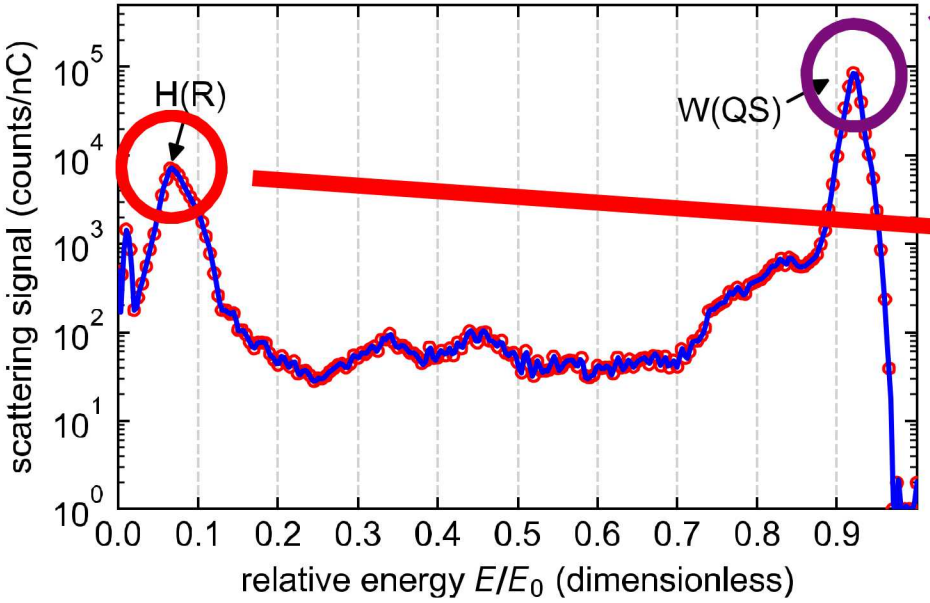
Key advantages of LEIS & DRS

1) direct detection of hydrogen on surface
2) structural information from W(QS) signal
3) surface specificity (monolayer sensitivity)

Constructing multi-angle maps with ARIES

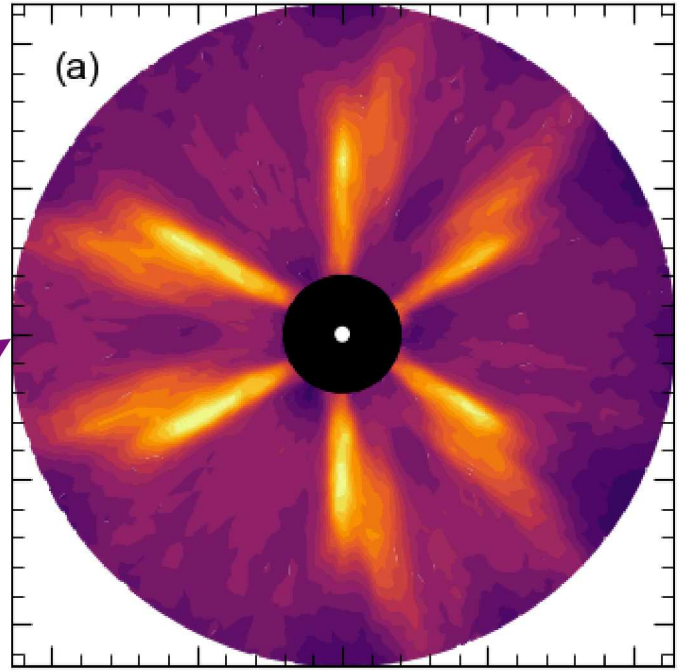


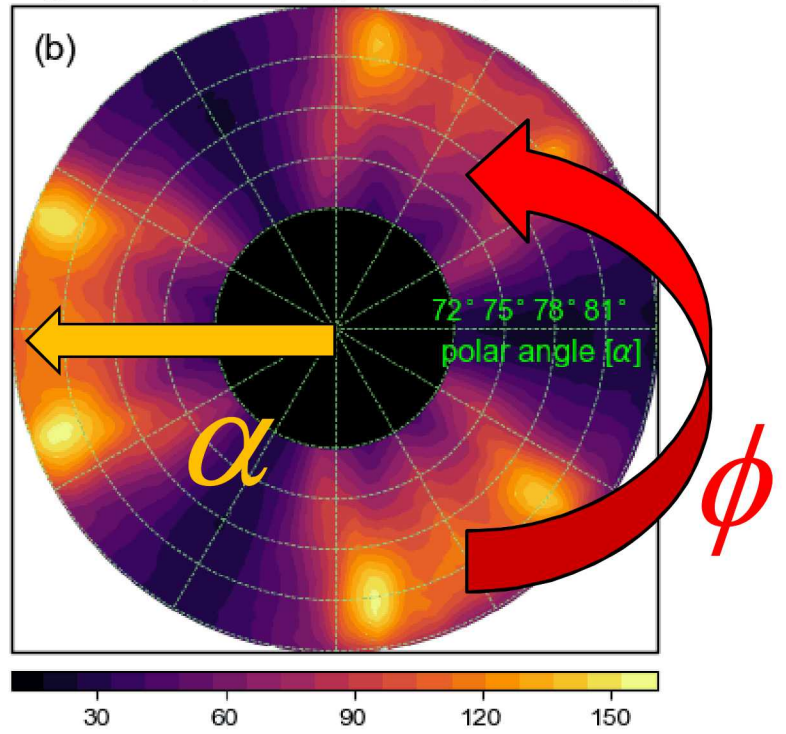
1 keV $Ne^+ \rightarrow W(111)$



W(111) structure & scattering processes

hydrogen location

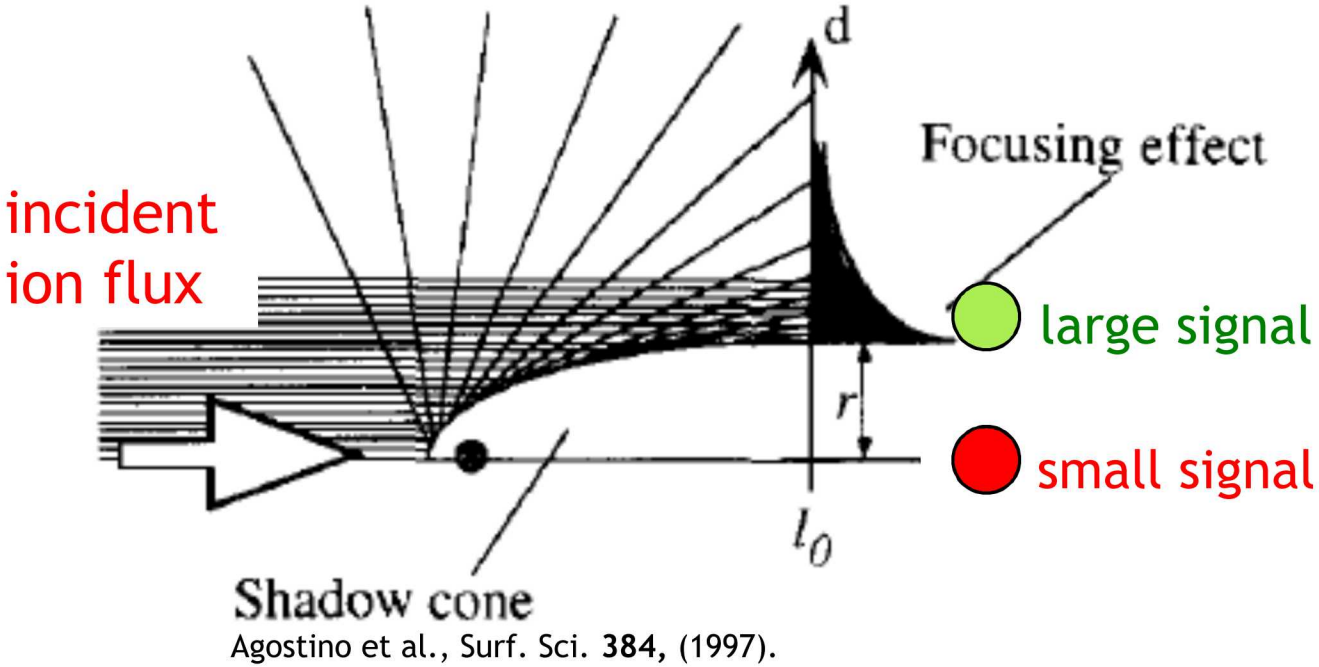




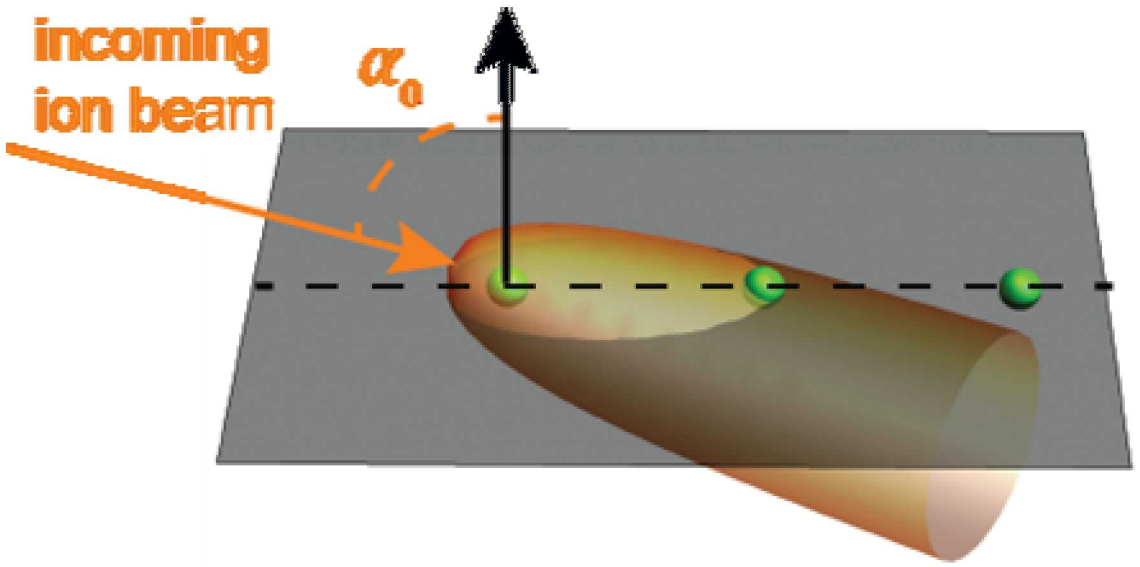
Most of the literature:

limited to single-angle scans

Shadow cones: insight into structure



Agostino et al., Surf. Sci. **384,** (1997).

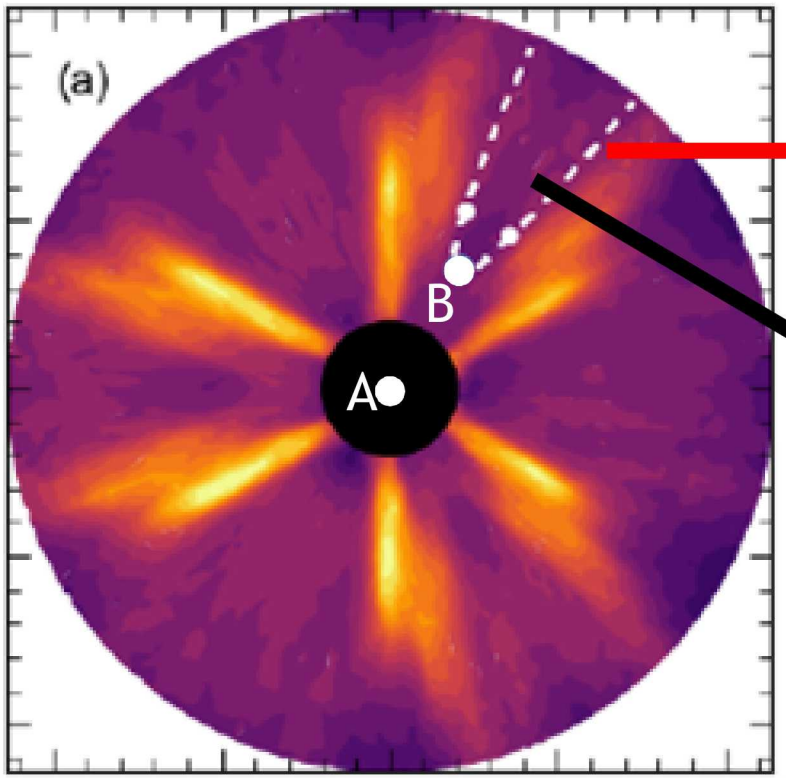


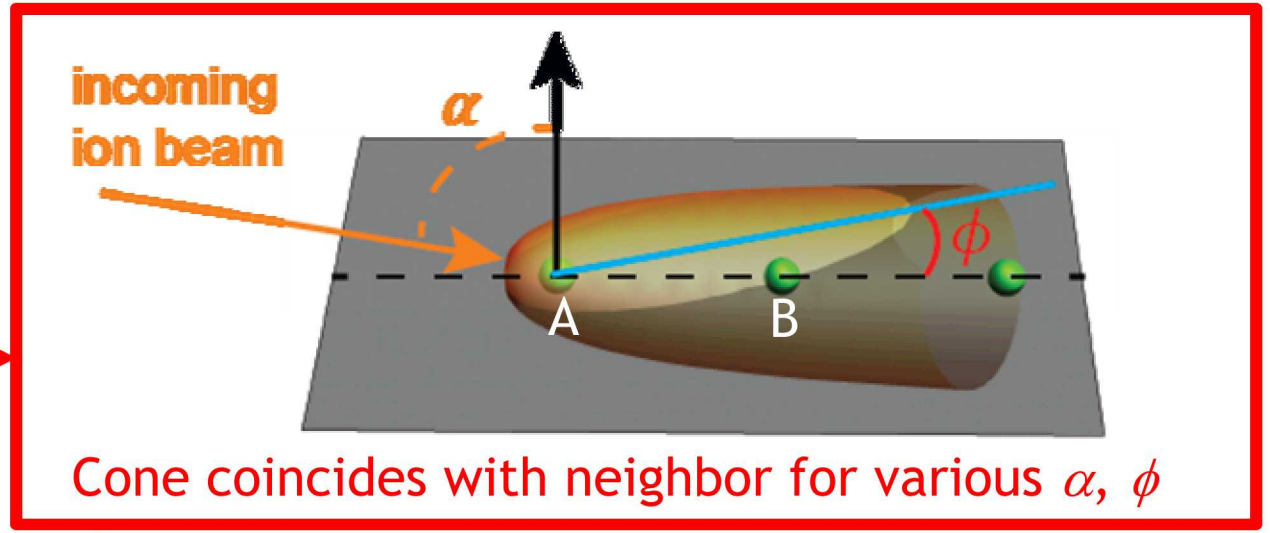
1) Shadow cones arise from ion focusing

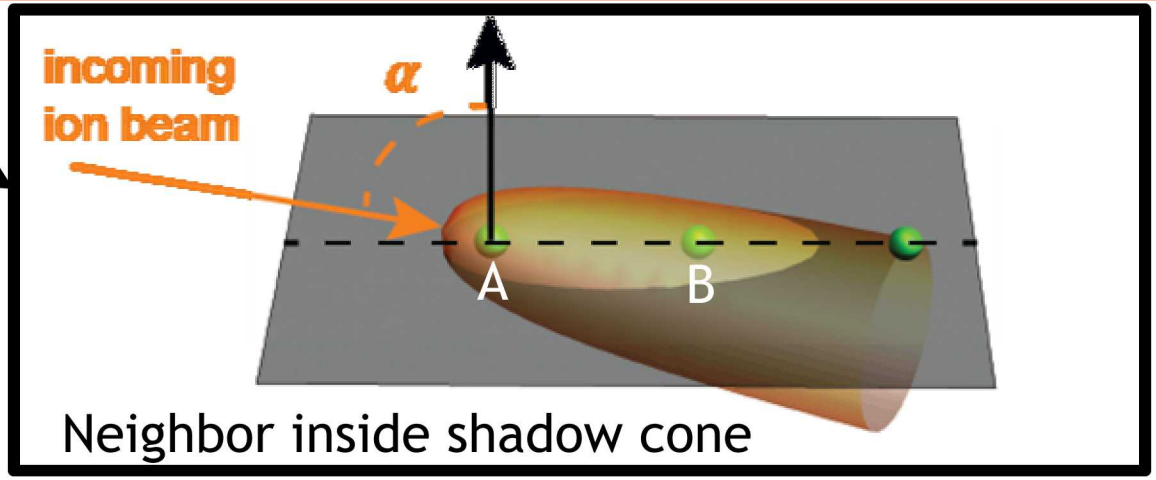
2) Enhanced signal when cone's edge coincides with neighboring atom

3) Structural information obtained with "shadow lines"

Shadow line analysis





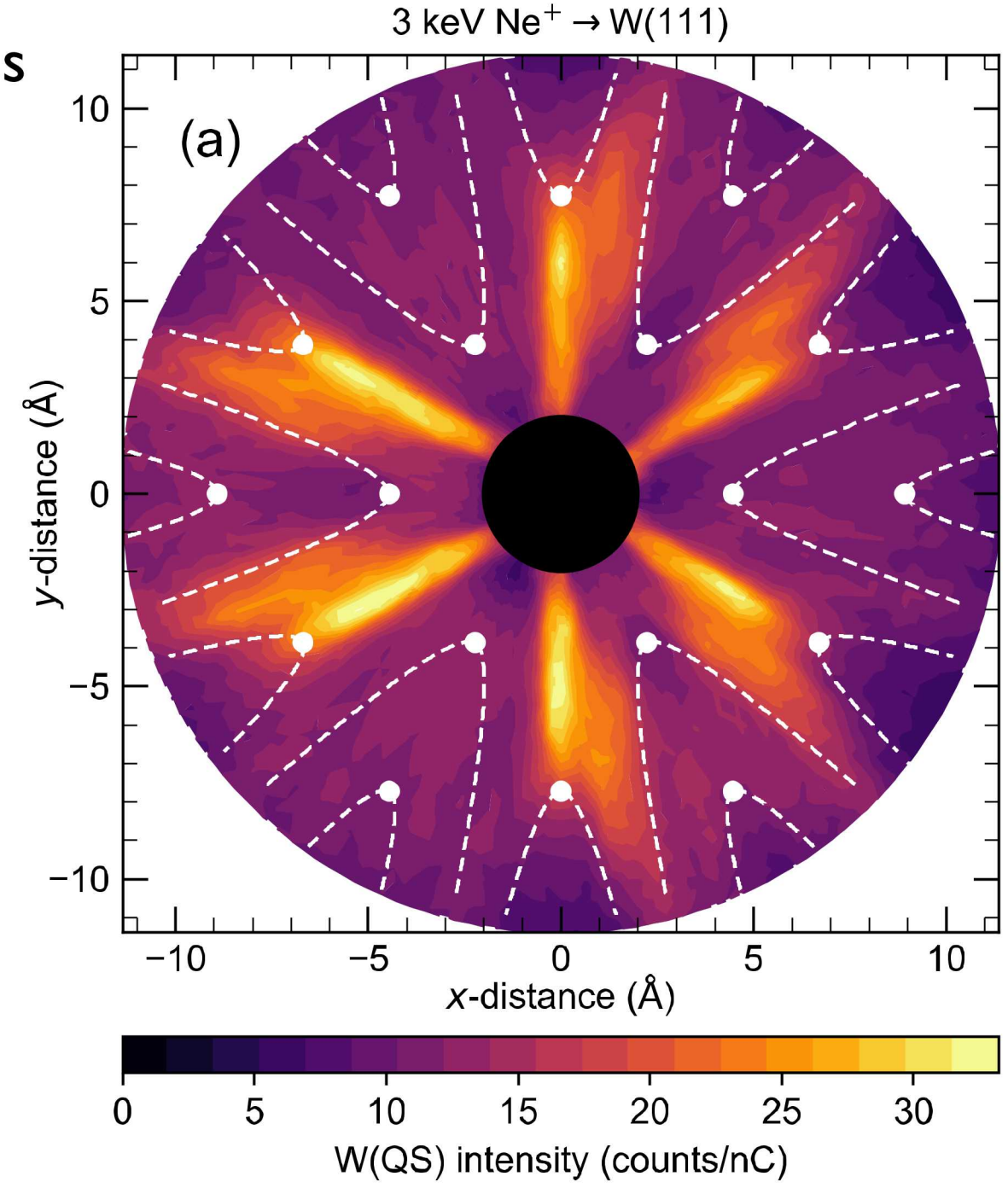


Shadow line:

- traces out α, ϕ that shadow cone coincides with W atom
- delineates region of enhanced scattering from diminished scattering

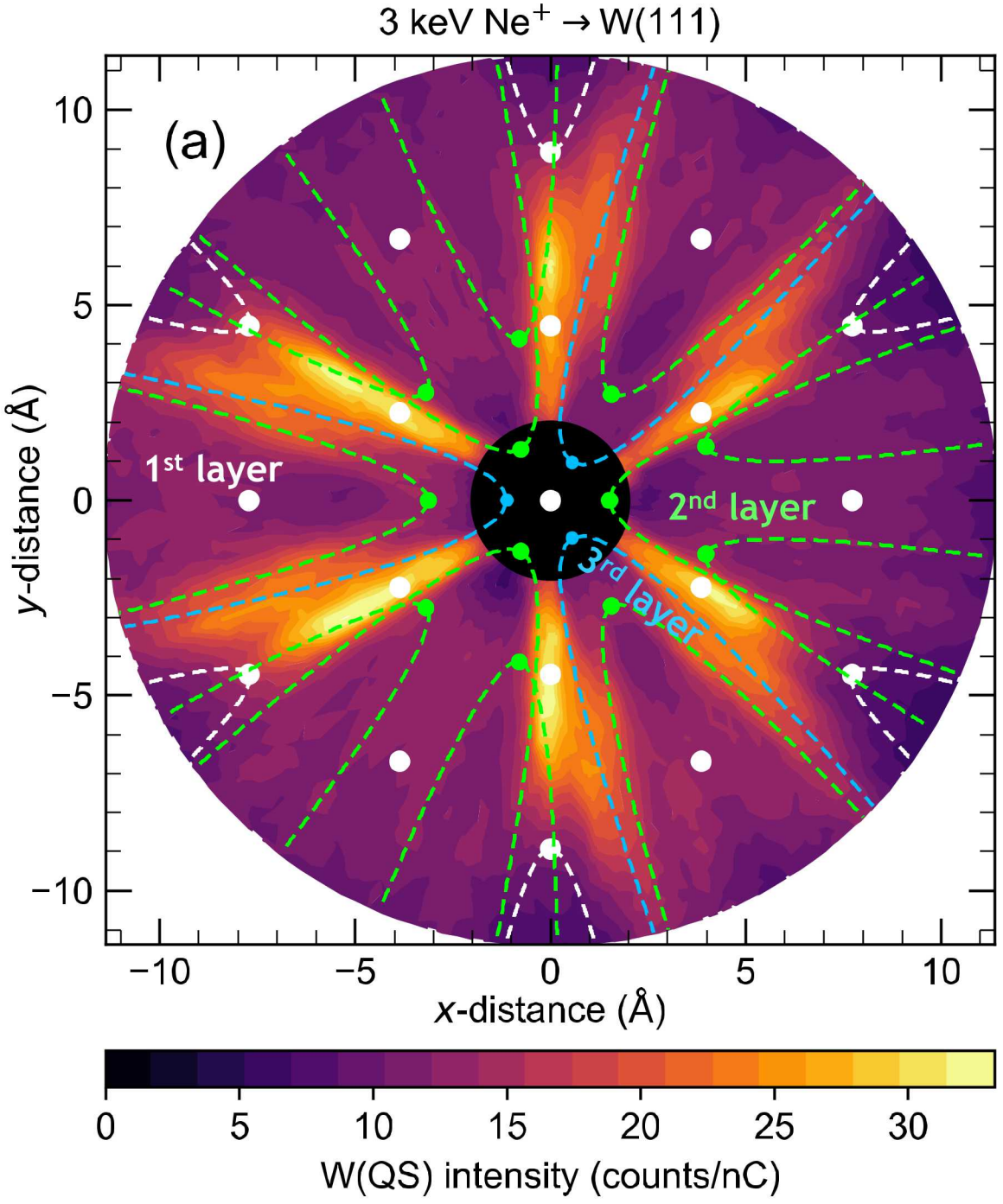
Shadow lines—a natural initial guess

- In the literature: shadow lines applied for only the first monolayer
- Insufficient to describe the signal
- Need to consider multiple layers

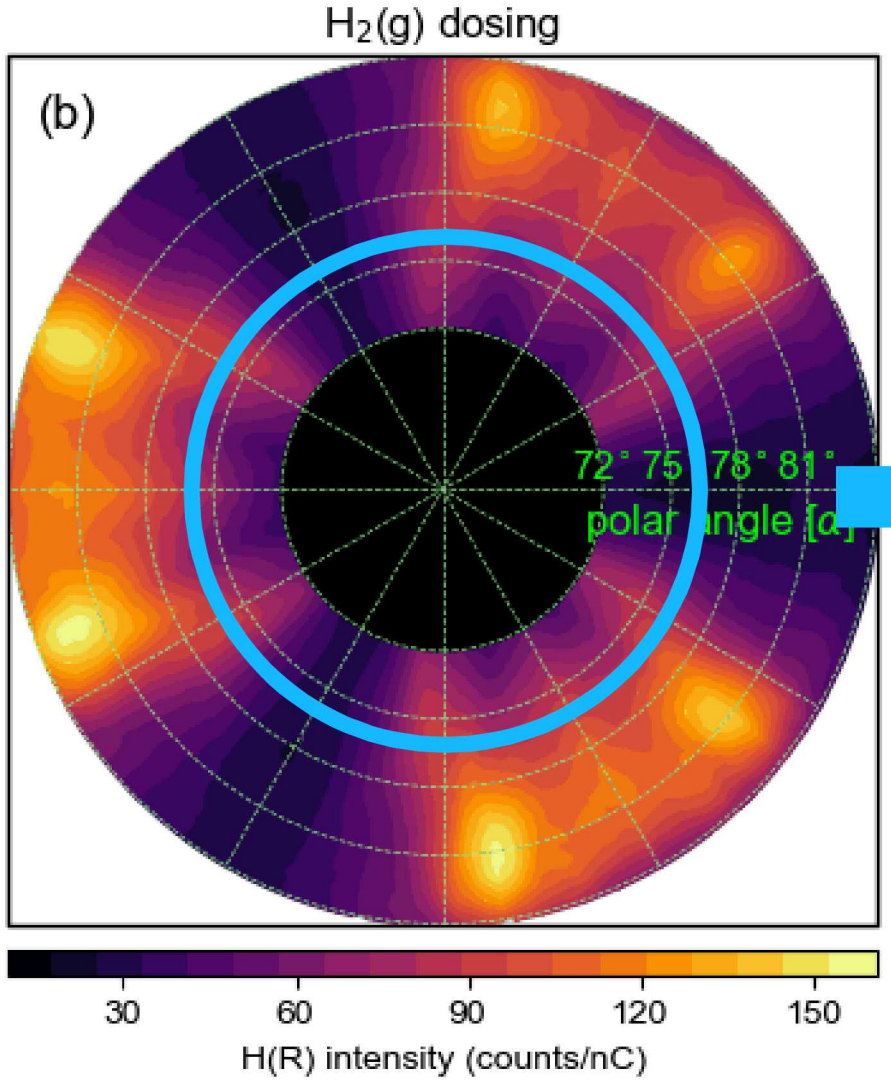


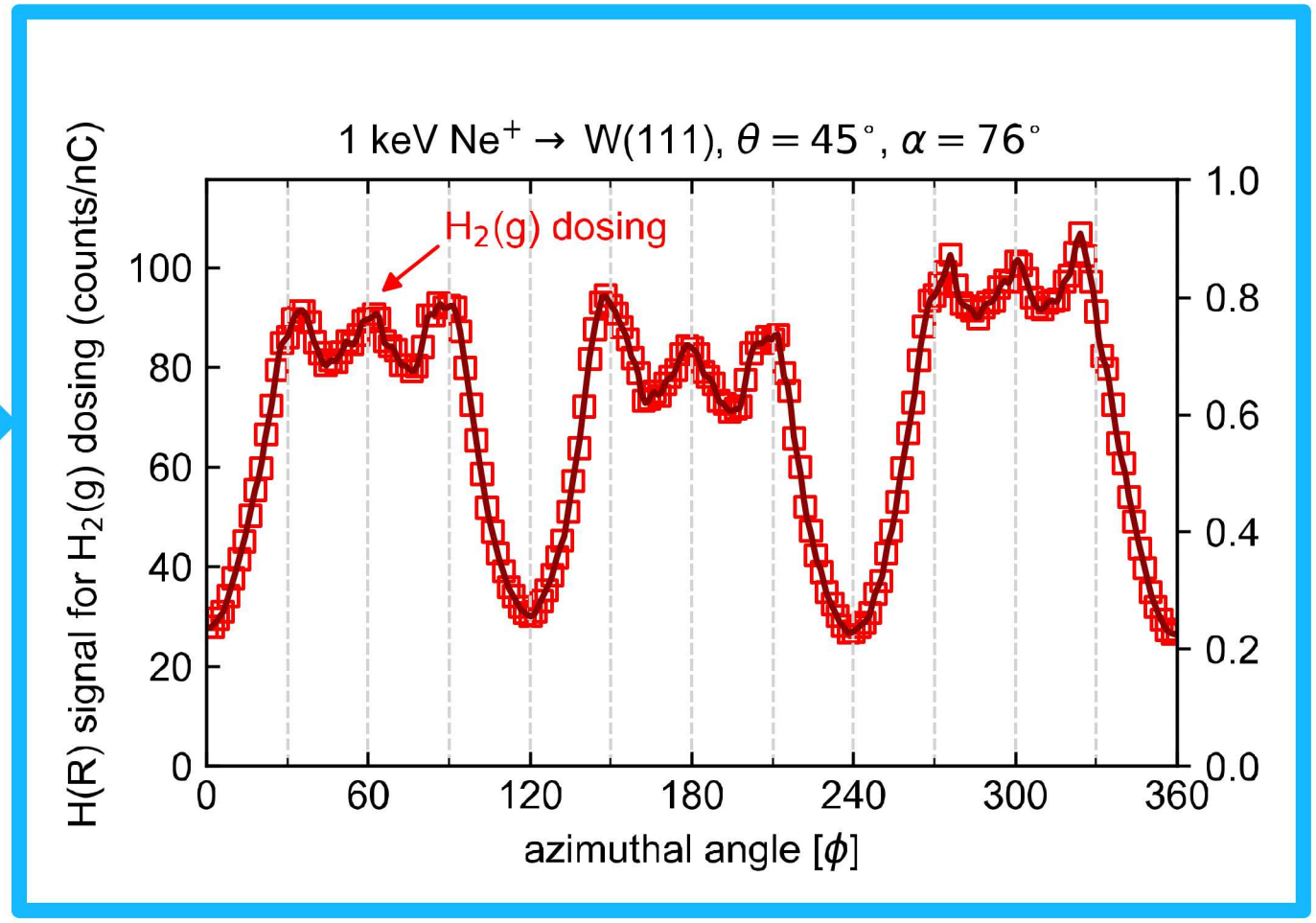
Multi-layer shadow line analysis

- Shadow lines of first 3 layers are required to describe the signal
- Multi-layer effects will play an important role in understanding H recoil maps



Determining H binding sites





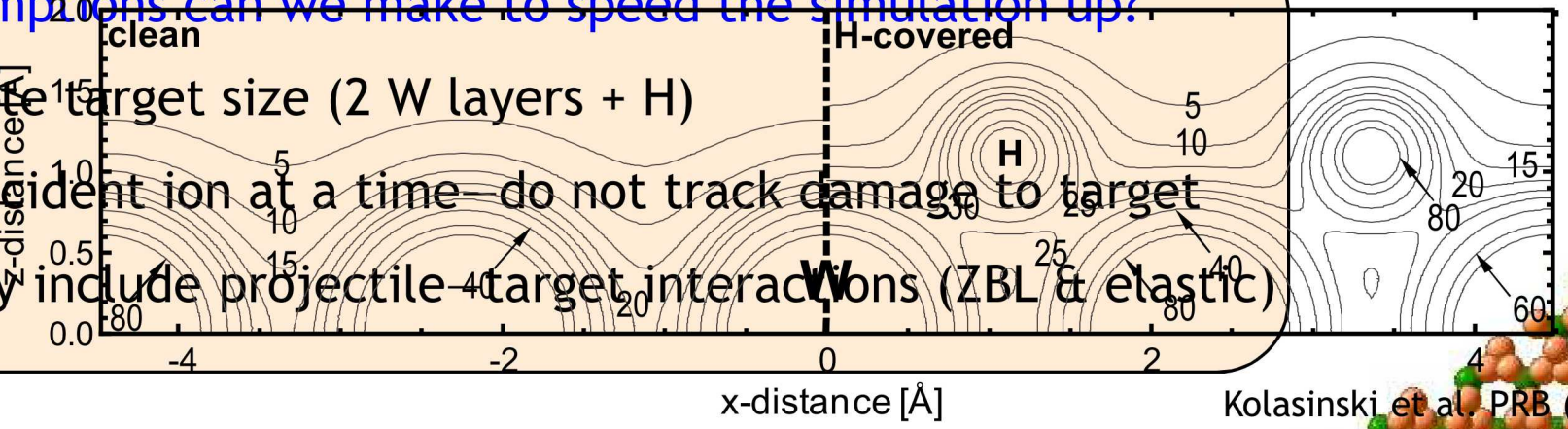
Model this signal to identify H adsorption sites

Modeling DRS measurements

Molecular dynamics (MD): necessary, but slow

What assumptions can we make to speed the simulation up?

- Finite target size (2 W layers + H)
- 1 incident ion at a time– do not track damage to target
- Only include projectile-target interactions (ZBL & elastic)



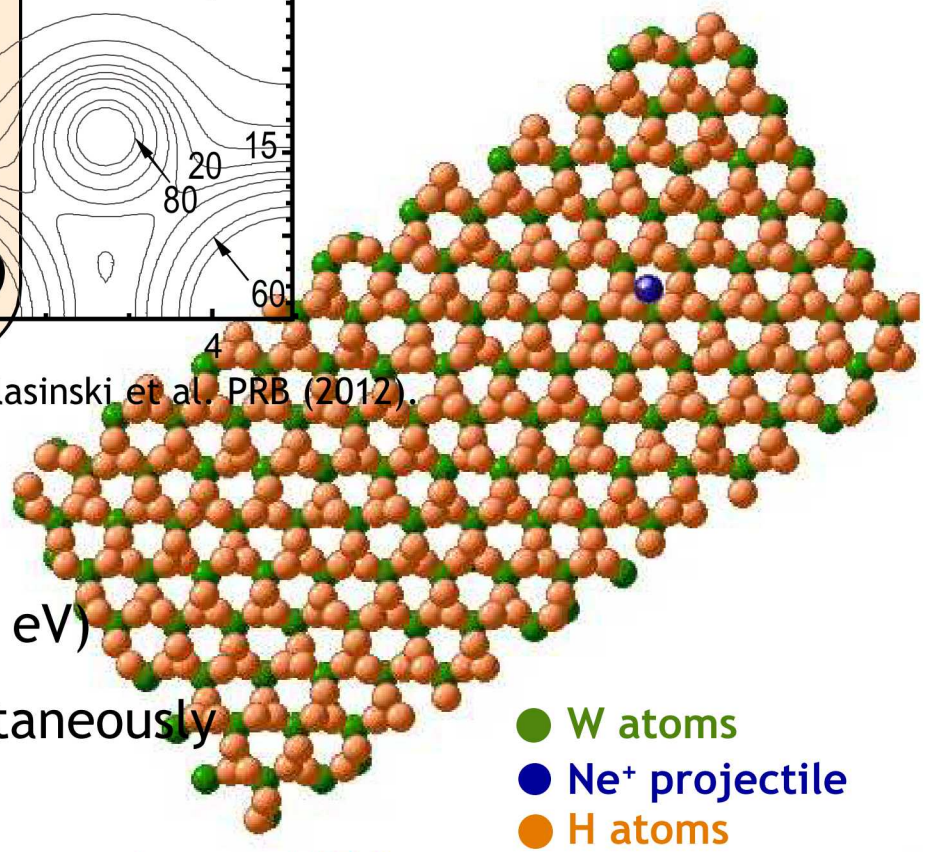
Kolasinski et al. PRB (2012).

At grazing incidences, incident ions:

- have small perpendicular ion energies (<10 eV)
- interact with multiple surface atoms simultaneously

Simulations performed with Kalypso [1], include:

- surface relaxation & vibrational displacements*
- saturated H coverage: H/W = 3
- count H that have correct trajectory to reach detector



Cannot use binary collision approximation…must use MD!

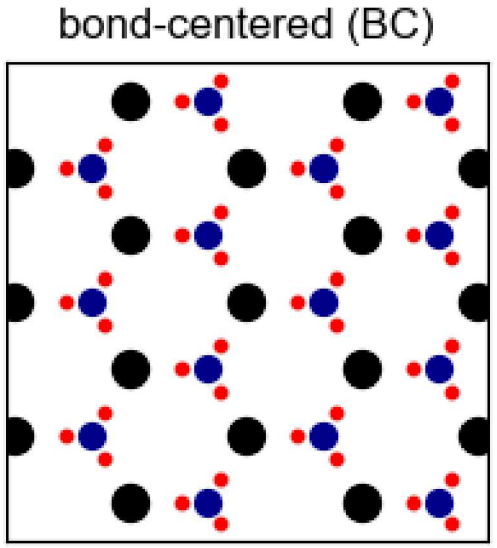
*DFT calculations performed by B. Wirth at U. Tenn

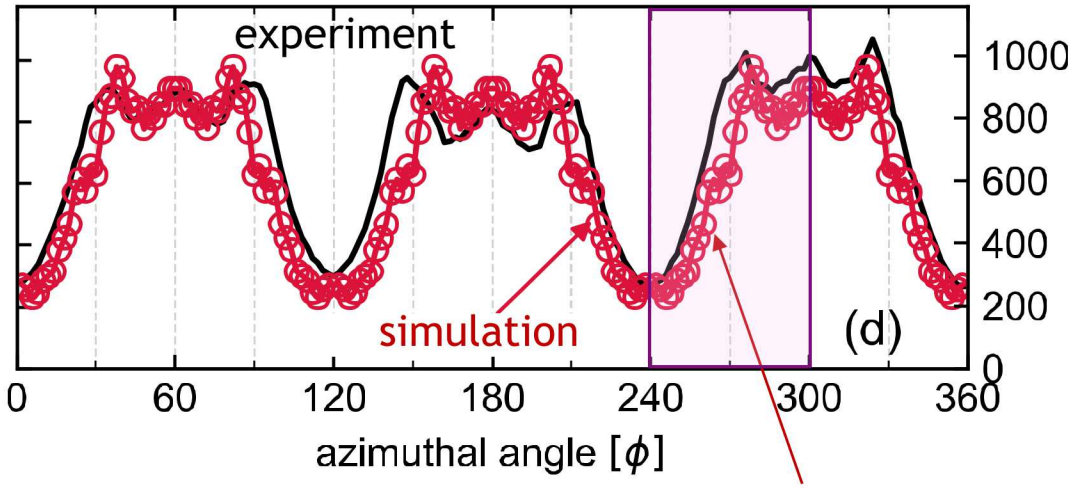
[1] Karolewski, Nucl. Instrum. Methods Phys. Res. B **230,** (2005).

MD simulation compared to ARIES measurement

DFT prediction

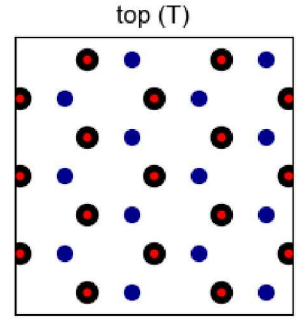
- 1st layer W
- 2nd layer W
- hydrogen

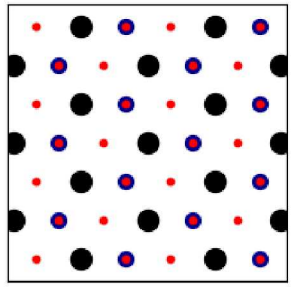


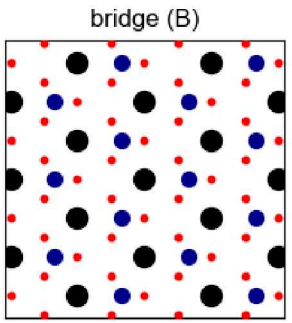


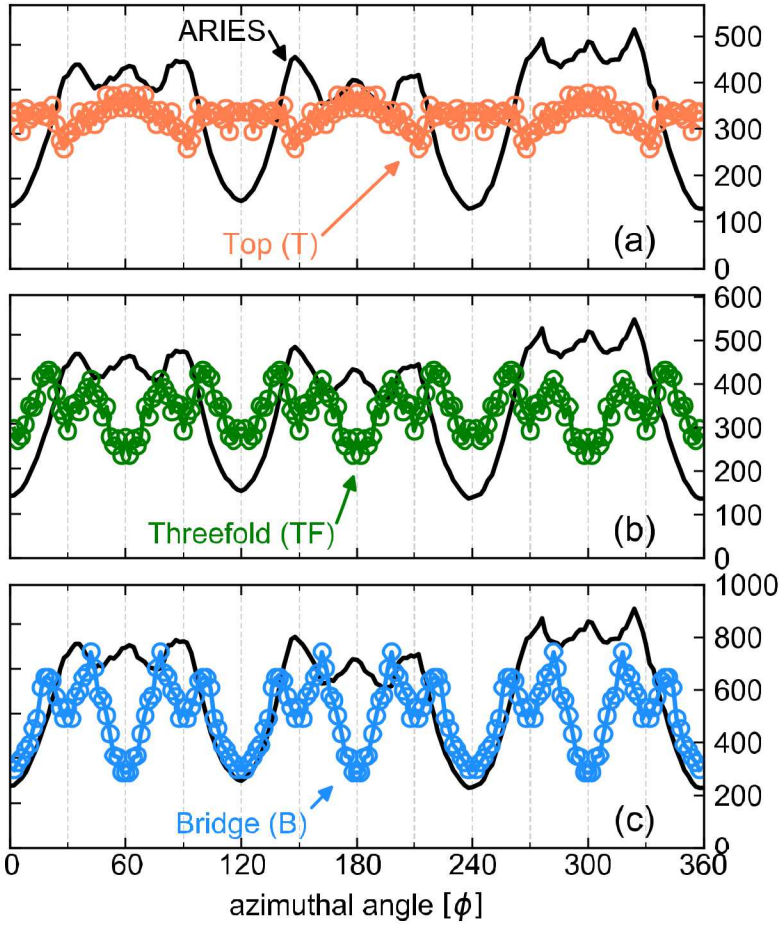
Each data point = 1.5 million simulated ion-target collisions

Other high symmetry sites









Constraining adsorbate height and position

h = 1.0 ± 0.1 Å

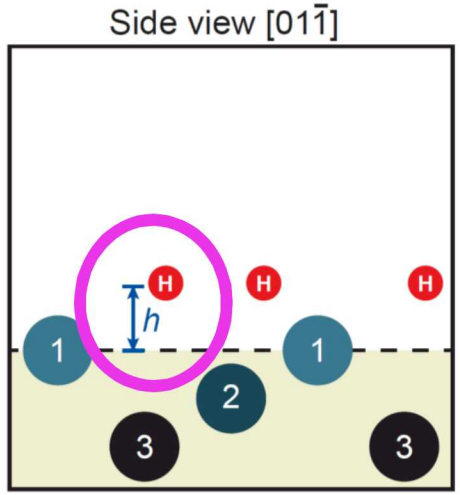
DFT prediction:

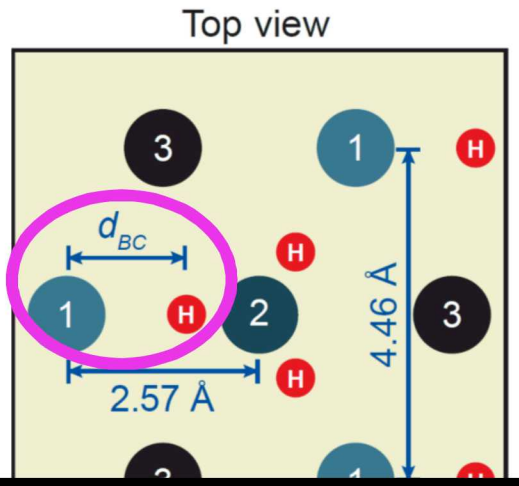
0.95 Å ≤ h ≤ 1.10 Å

d_{BC} = 1.6 ± 0.1 Å

DFT prediction:

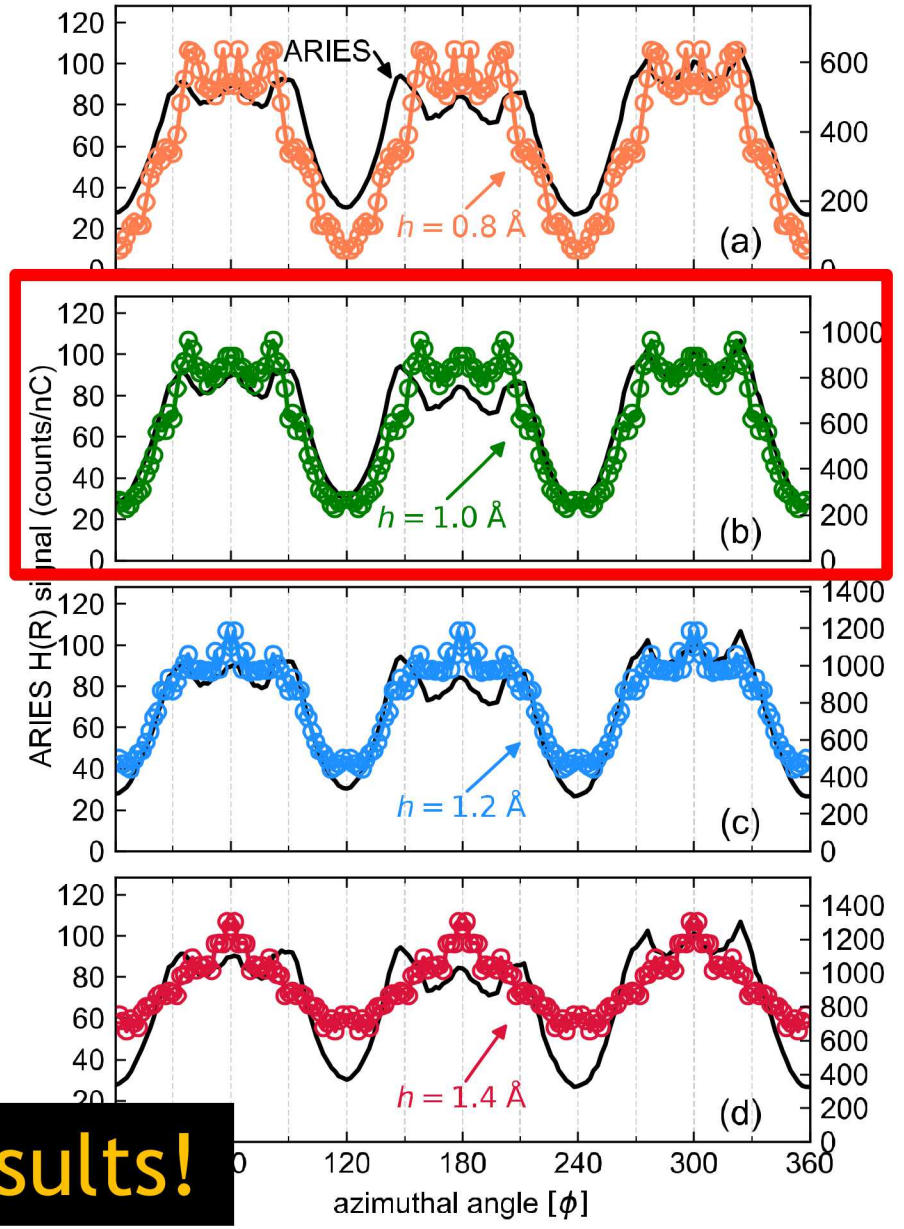
1.5 Å ≤ d_{BC} ≤ 1.7 Å





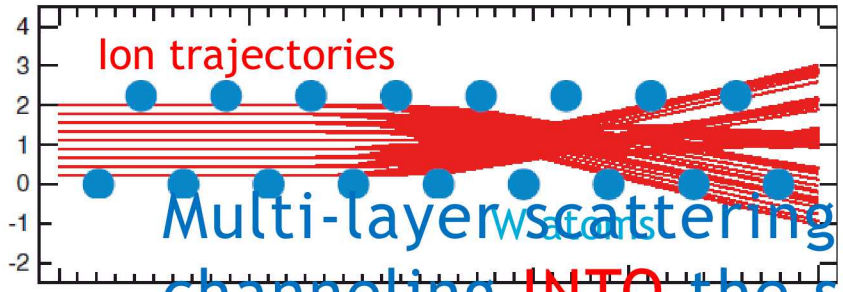
DFT prediction

varying h



Strong validation of DFT results!

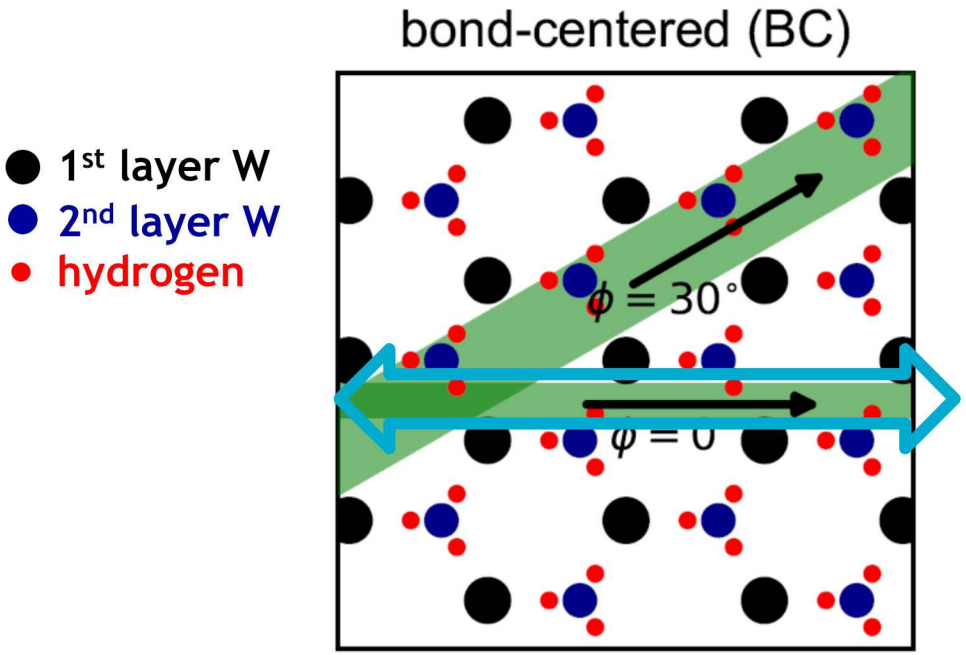
Understanding the H multi-angle map

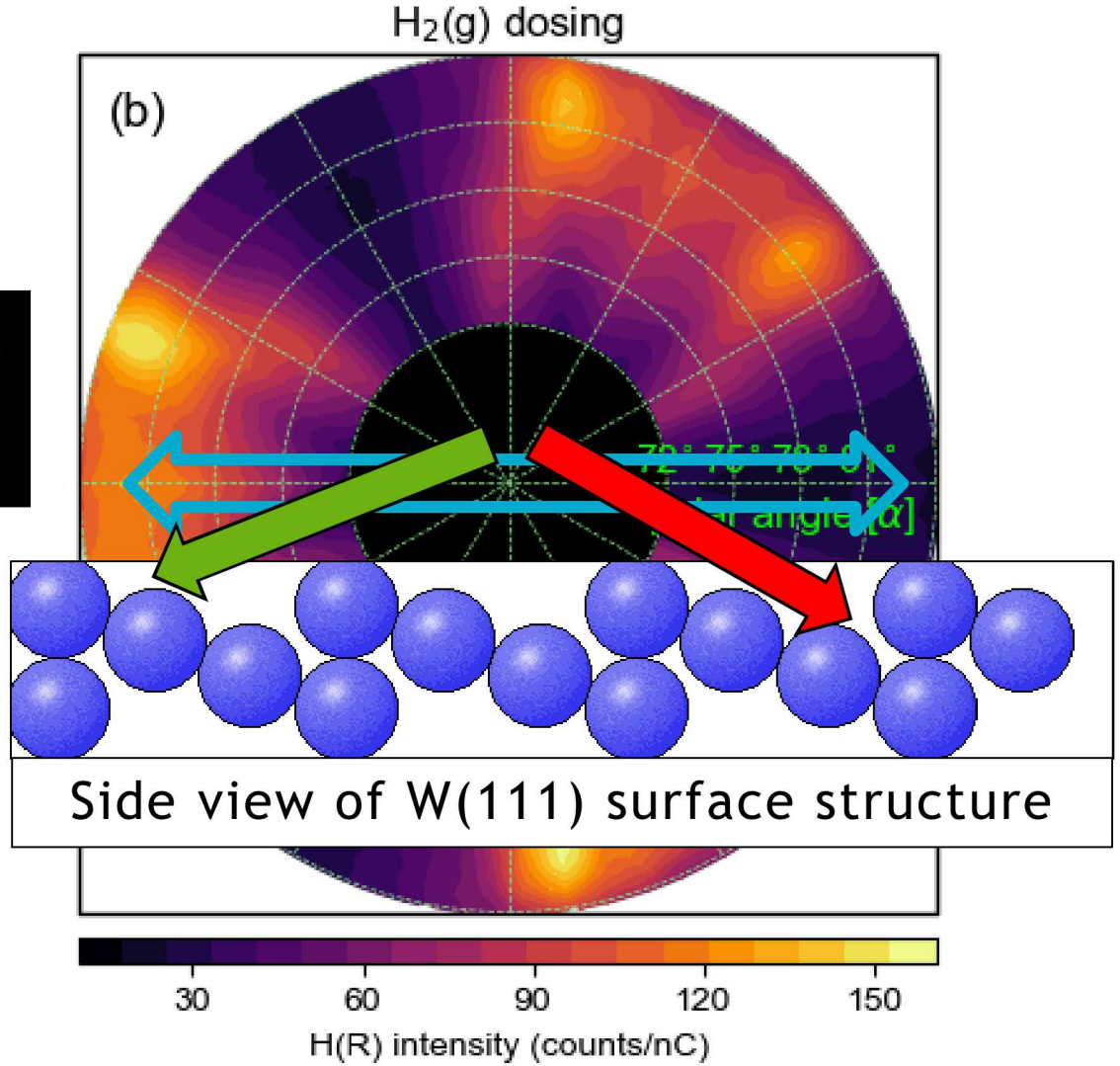


Multi-layer scattering effect:
channeling INTO the surface

Surface channeling of ions at grazing incidence[1]:

H within channel ➔ peak in H(R) signal



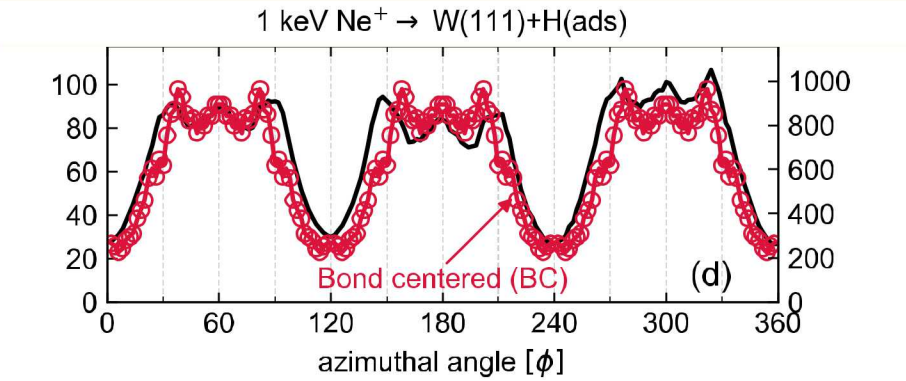


[1] Kolasinski et al., PRB, (2012).

Development of a new MD simulation capability using LAMMPS

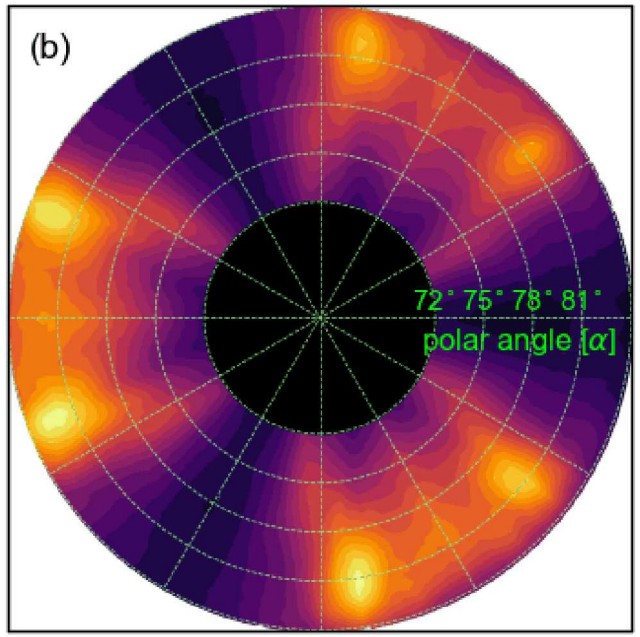
Some limitations of Kalypso:

- Not designed for large-scale parallel simulations
- Lacks flexibility for modeling more complex surfaces
- Poor scaling for larger system sizes

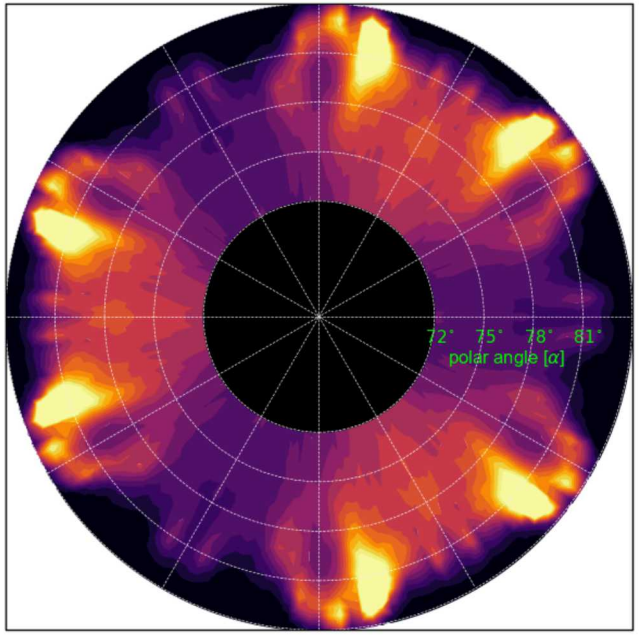


I am developing a Python code that uses LAMMPS [1] to simulate LEIS & DRS

Experiment: ARIES



Simulation: Python—LAMMPS



- ~500 million simulated ion-target collisions
- well beyond existing LEIS MD simulations
- currently tuning choices in parameters

[1] S. Plimpton, J. Comp. Phys. (1995).

Summary of W(111)+H(ads) study

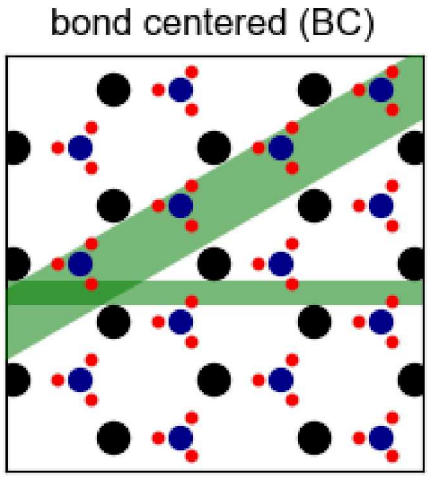
1. Advancement of surface hydrogen characterization:

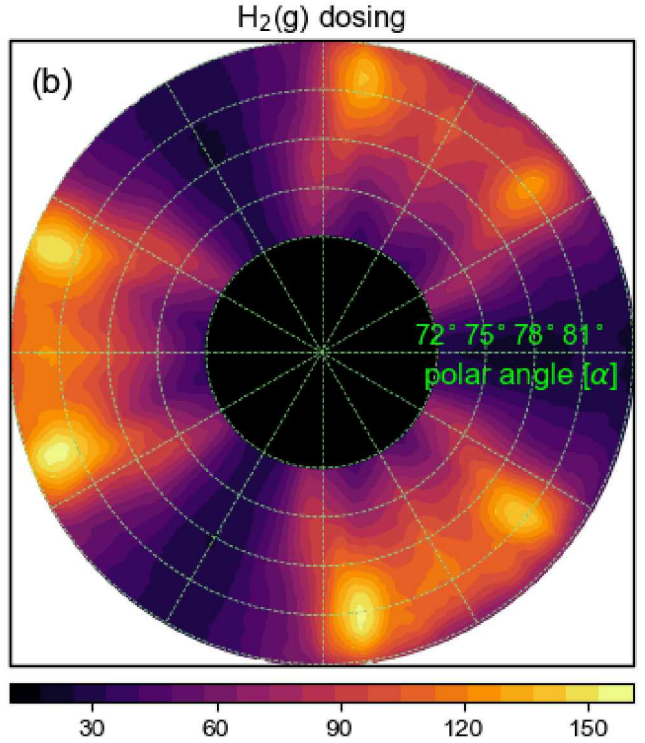
   complex binding geometry and corrugated surface

2. Most extensive MD simulations of LEIS to date:

   constrained H adsorbate height and location

3. Using LAMMPS to improve our existing MD models





C.-S. Wong, J. Whaley, Z. Bergstrom, B. Wirth, and R. Kolasinski, **Phys. Rev. B** 100, 245405 (2019).

Acknowledgements

Sandia National Laboratories:

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- Josh Whaley

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- Brian Wirth
- Zack Bergstrom

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Extra slides

The divertor

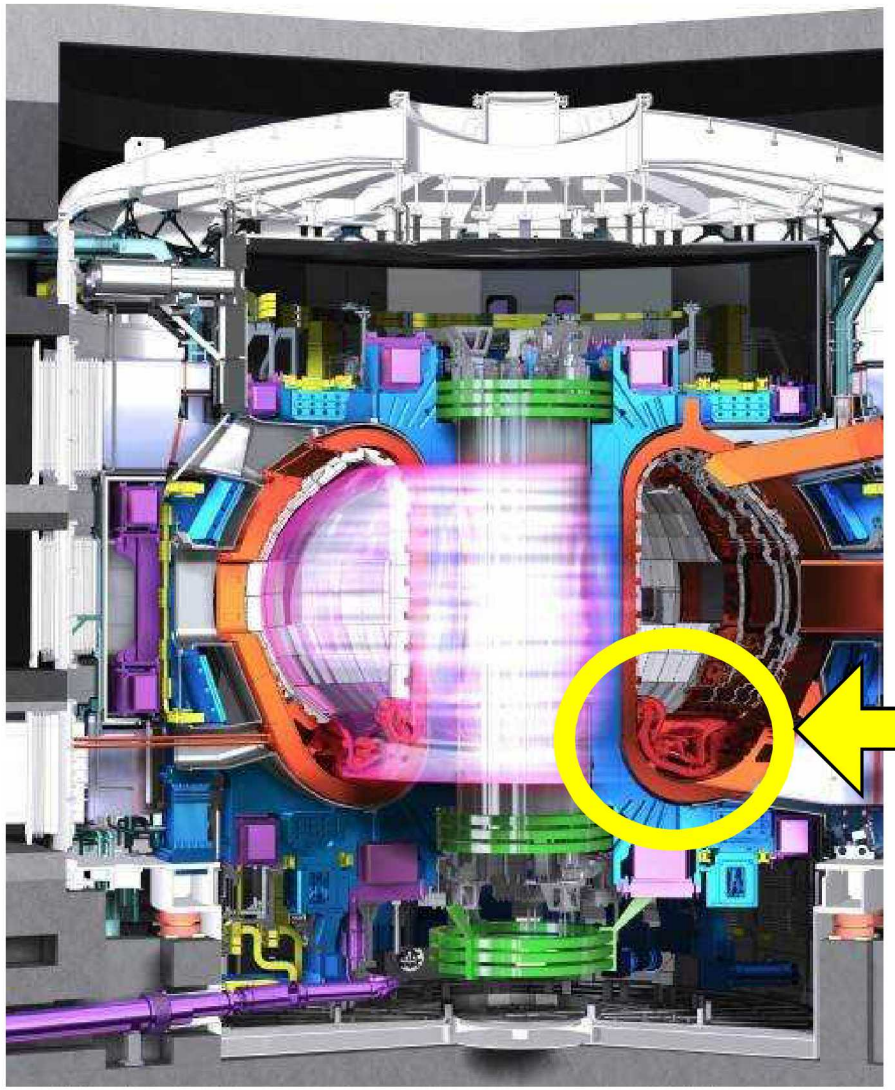


Image: iter.org

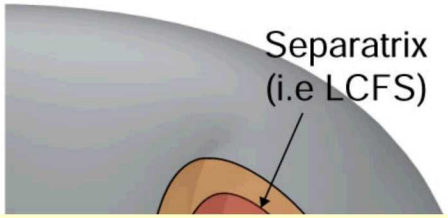
Fusion reaction:

$D + T \rightarrow He + n^0$ (14.1 MeV)

requires confinement of extremely hot (>100,000,000 °C)

and dense plasma

achieve confinement with magnetic fields



Divertor: where heat and particle flux (D, T, He, & impurities) are intentionally dumped

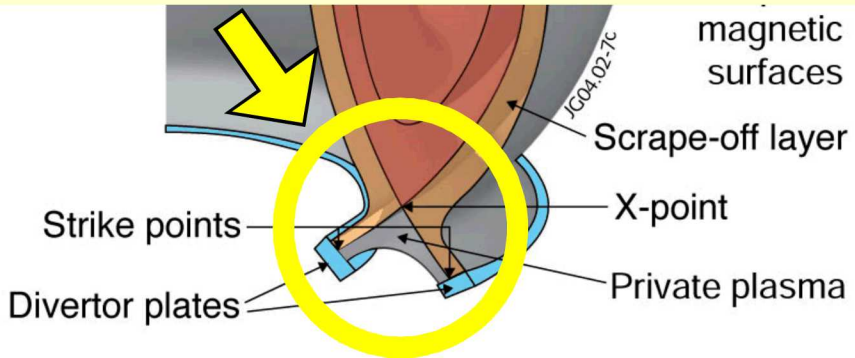
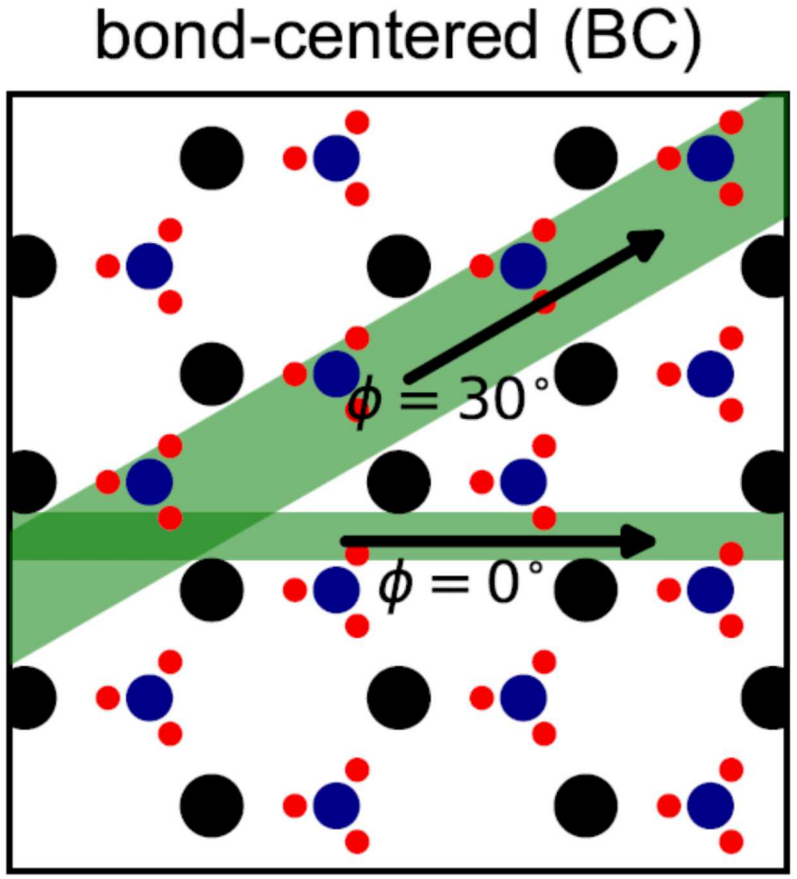
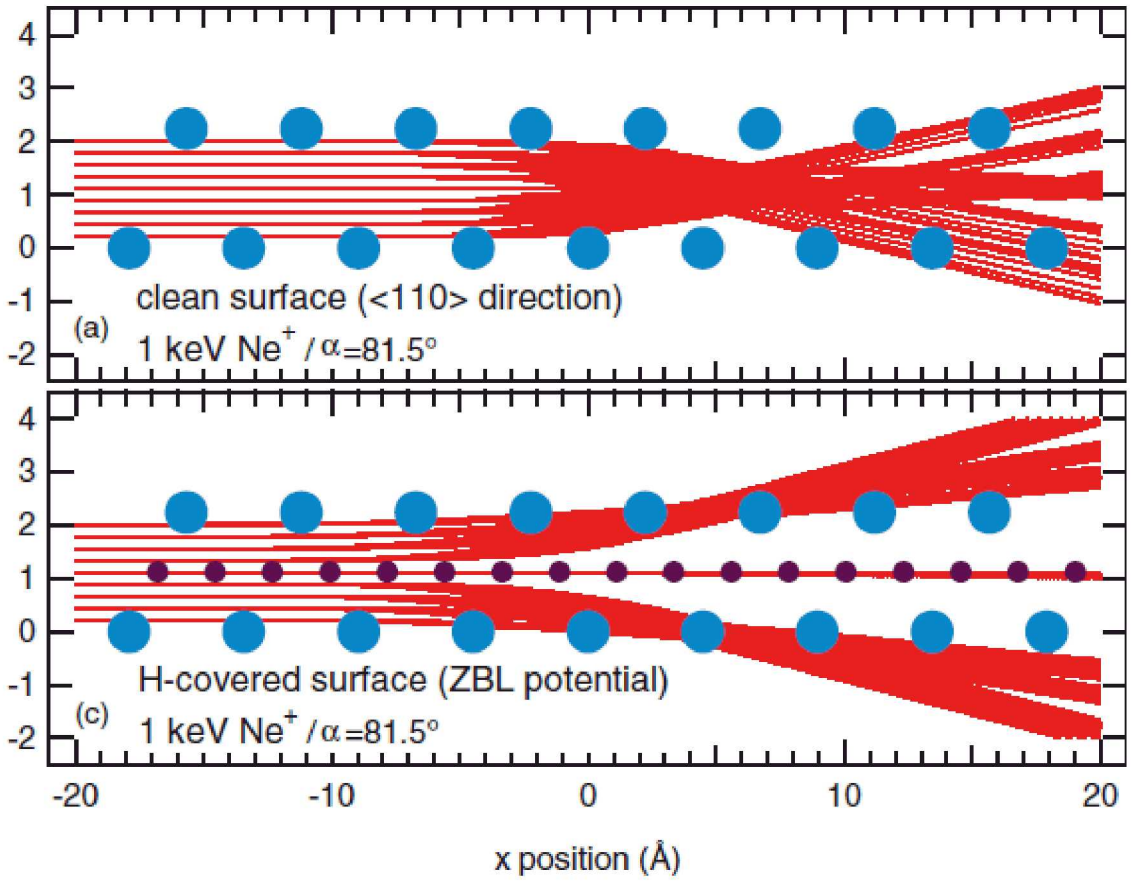


Image: euro-fusion.org

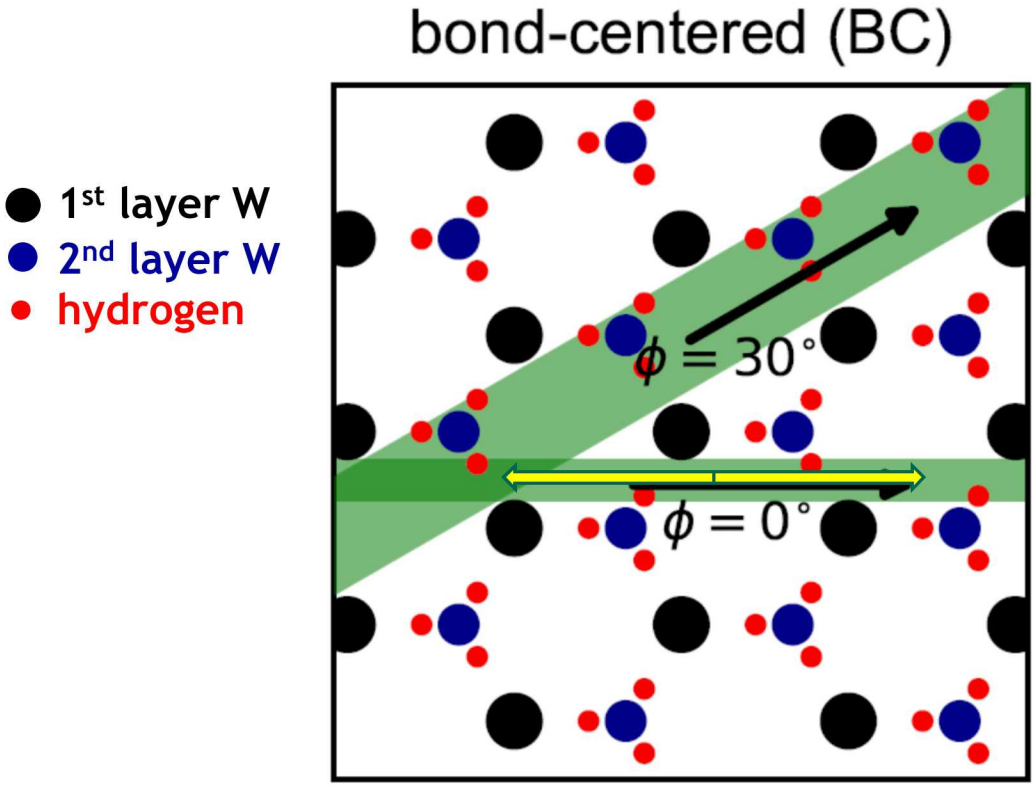
Effect of hydrogen on ion-channeling

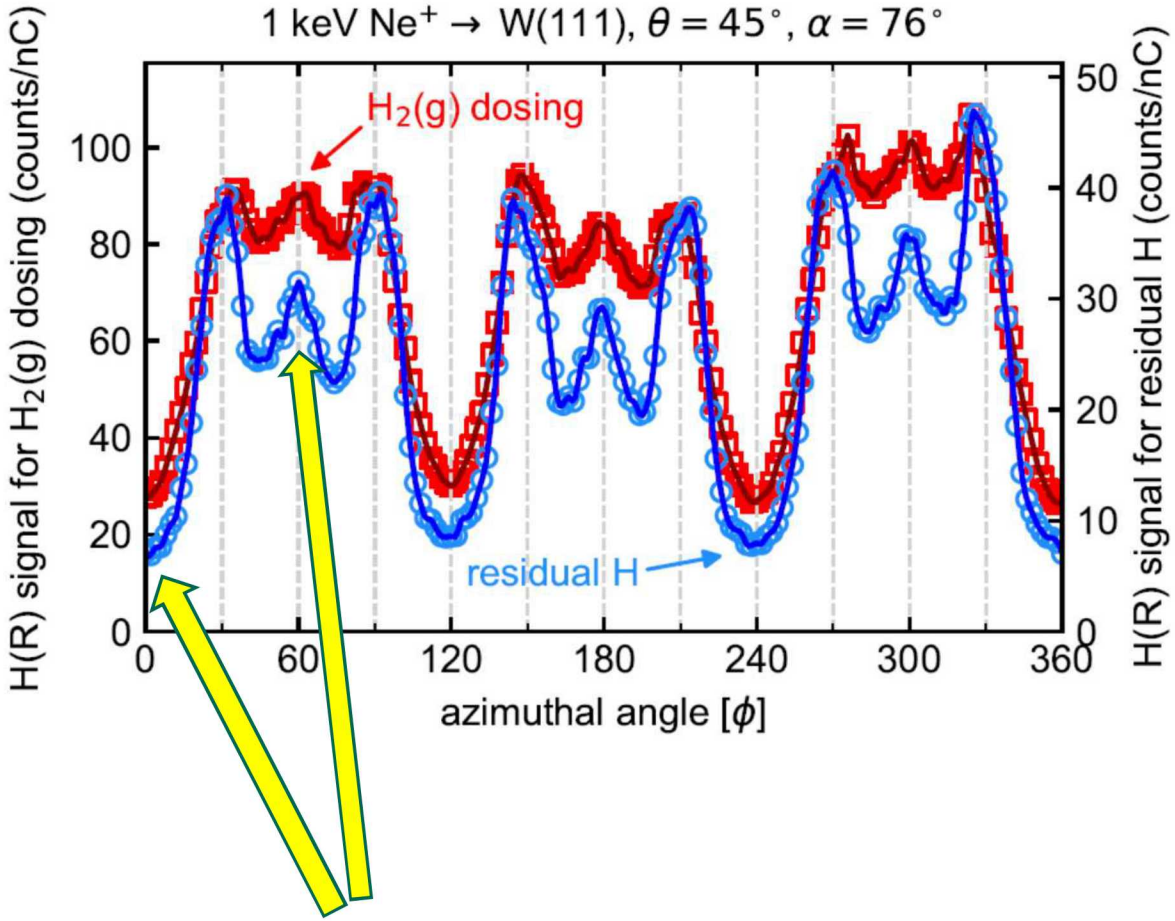




[1] Kolasinski et al., PRB 2012.

Adsorbate enhancement of ion channeling



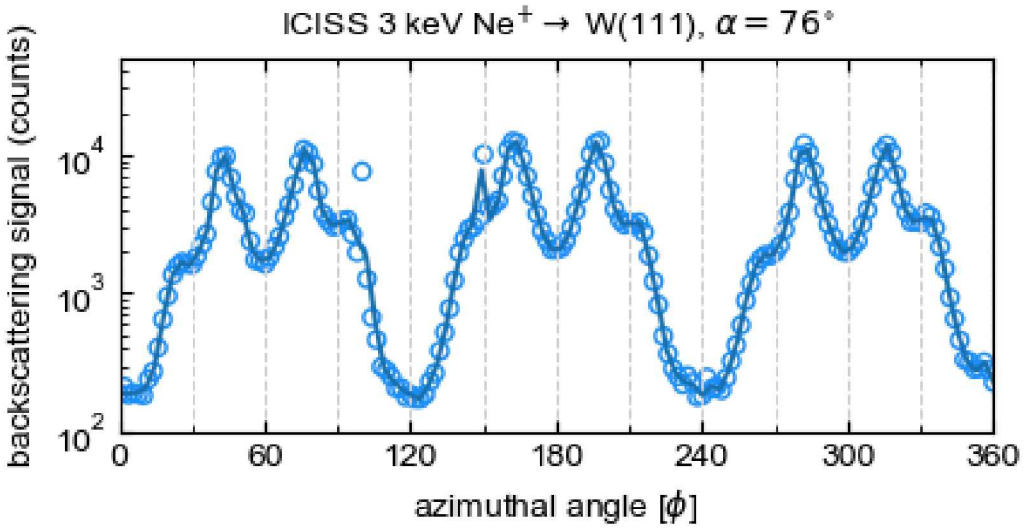


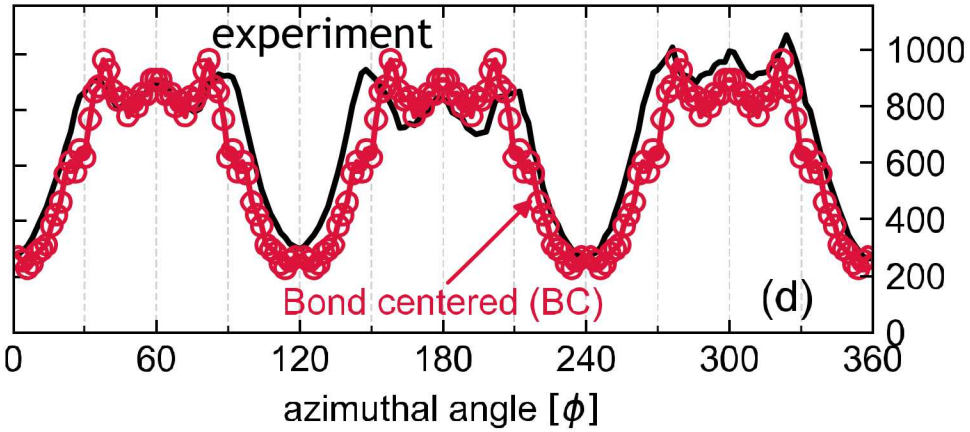
Increased signal with H dosing due to enhanced ion channeling

Channeling into the surface breaks symmetry

backscattering measurements:
minimum ⇔ channeling into surface

DRS hydrogen signal

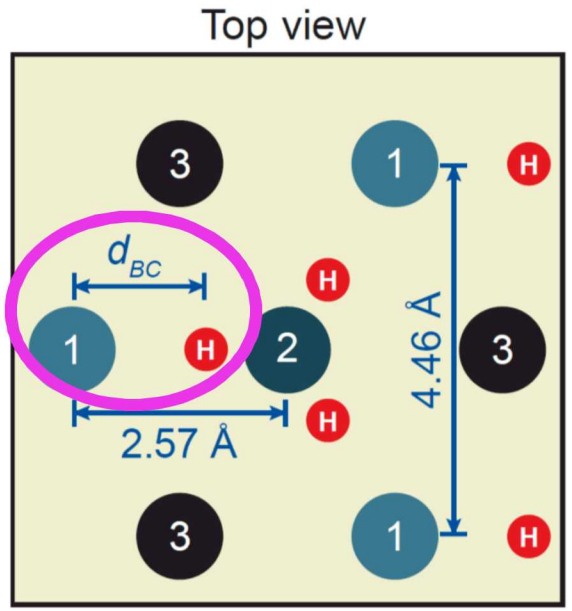


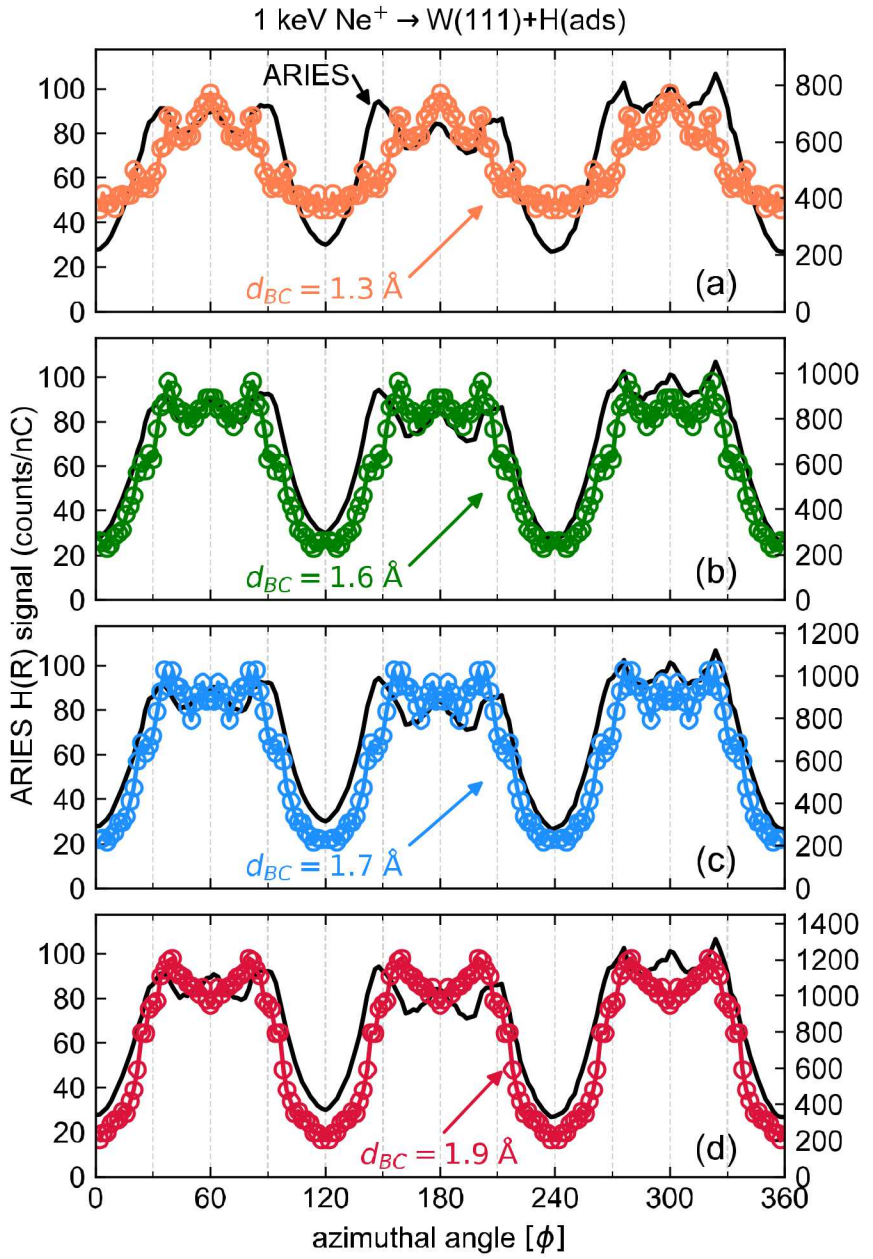


Constraining adsorbate position

We find d_{BC} = 1.6 ± 0.1 Å

DFT prediction:
1.49 Å ≤ d_{BC} ≤ 1.69 Å





MD simulation for multi-layer scattering