ROG Neutron Reflectometer: Ready for Cold Neutrons

March 2023

The ROG Team

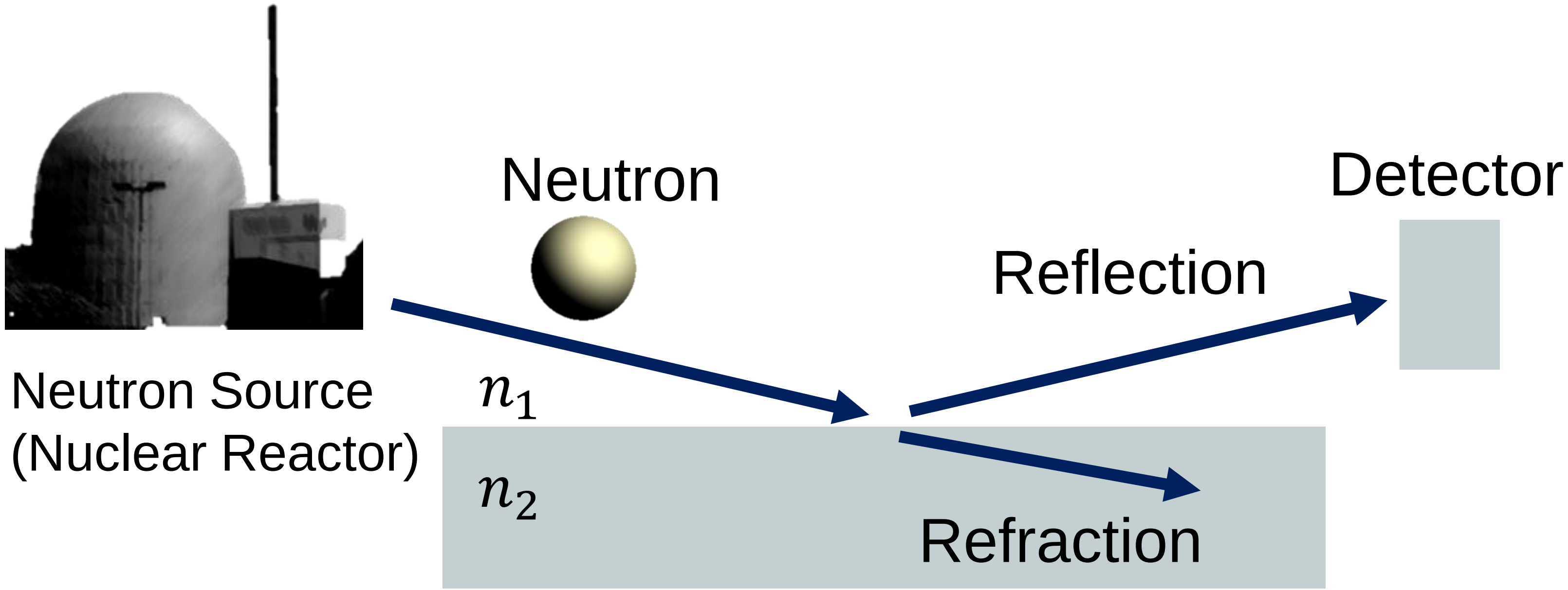
REACTOR INSTITUTE DELFT

TU Delft

Neutron Reflectometry

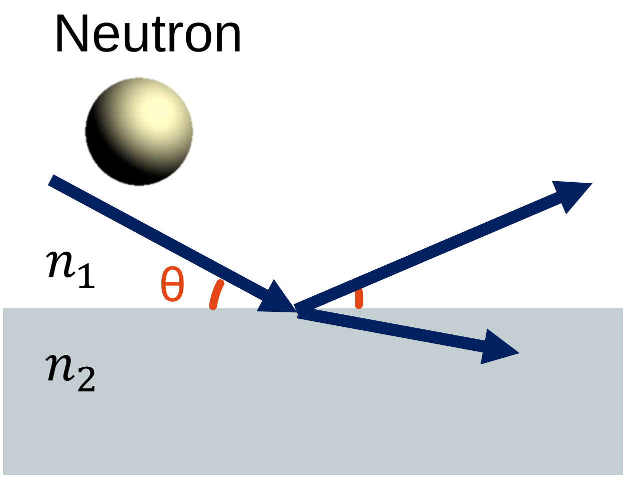
Neutron Reflectometry is a method to obtain structural information (thickness, composition, roughness) of flat samples with length scales between ≈1 – 200 nm (e.g. thin film on a substrate)

A neutron reflectometry experiment involves illuminating a flat sample by a collimated neutron beam and measuring the fraction of neutrons reflected ('Reflectivity')

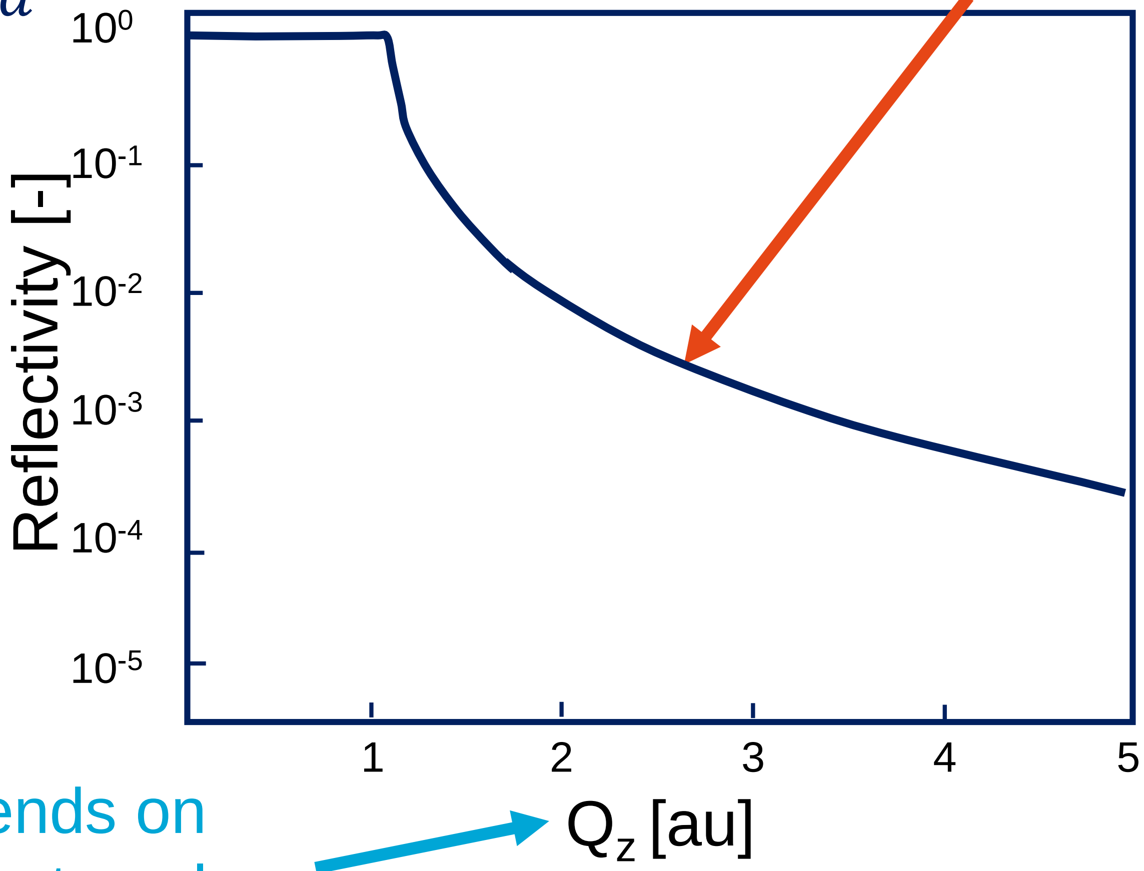


Reflectivity measured

$$Reflectivity \\ = Fraction\ of\ neutrons\ reflected$$



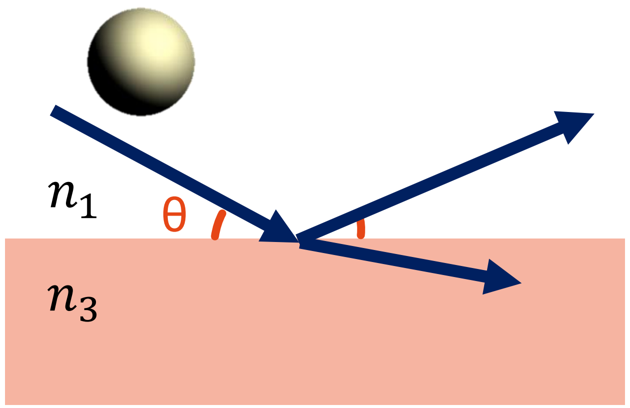
Depends on composition of the material

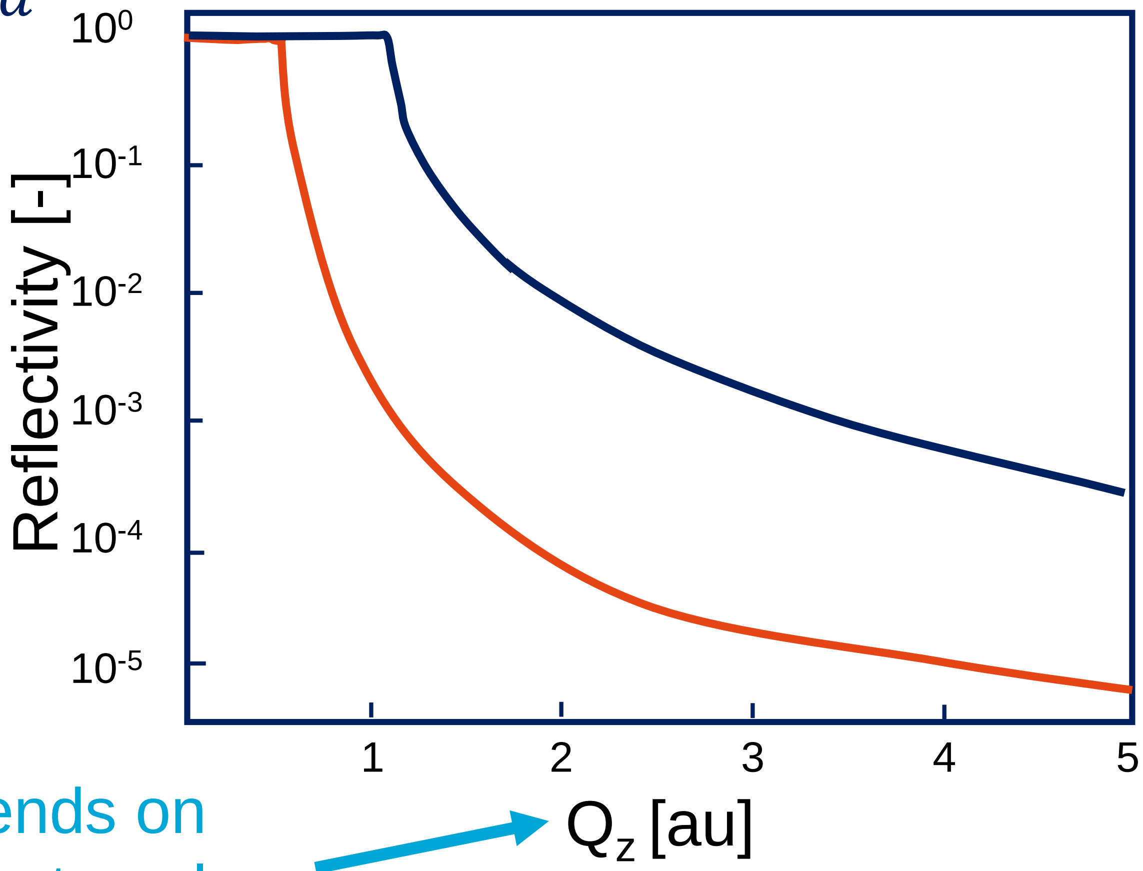


Parameter that depends on wavelength or incident angles

Reflectivity measured – Different material

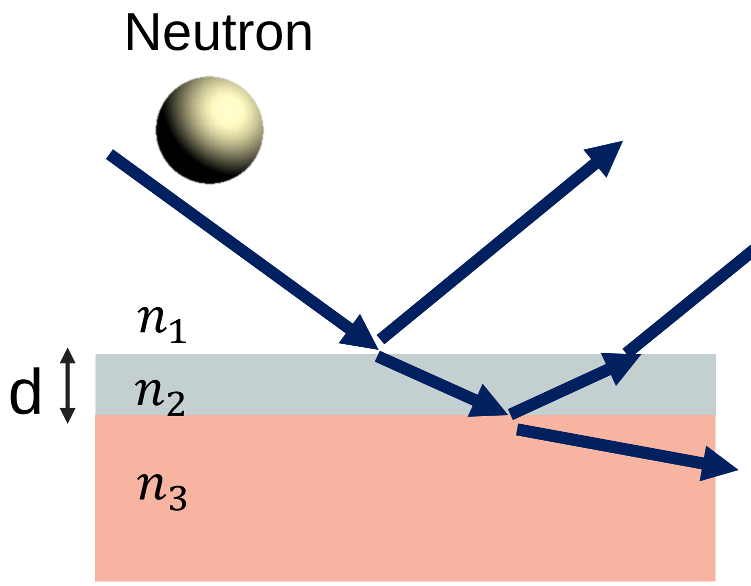
$$Reflectivity$$
$$= Fraction\ of\ neutrons\ reflected$$

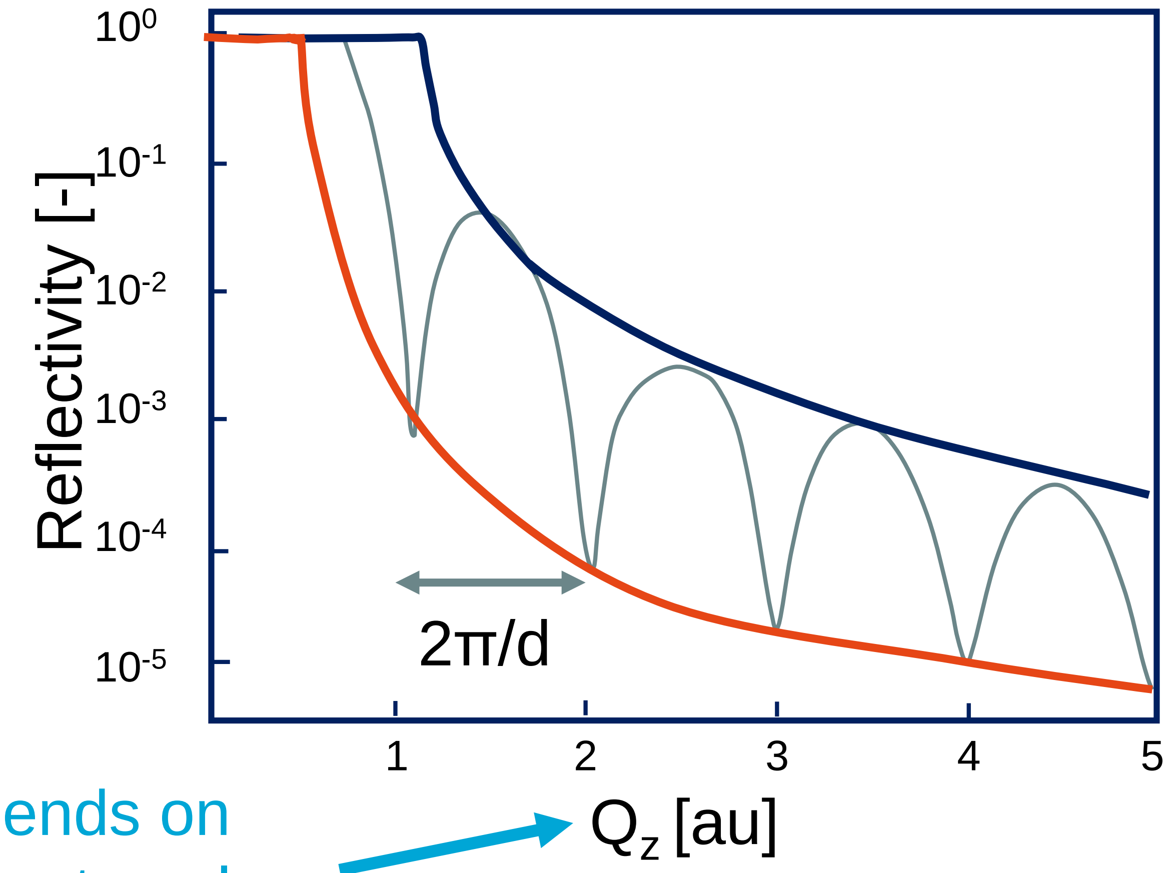




Parameter that depends on wavelength or incident angles

Reflectivity measured – Thin layer on top of a material





Parameter that depends on wavelength or incident angles

Neutron Reflectometry

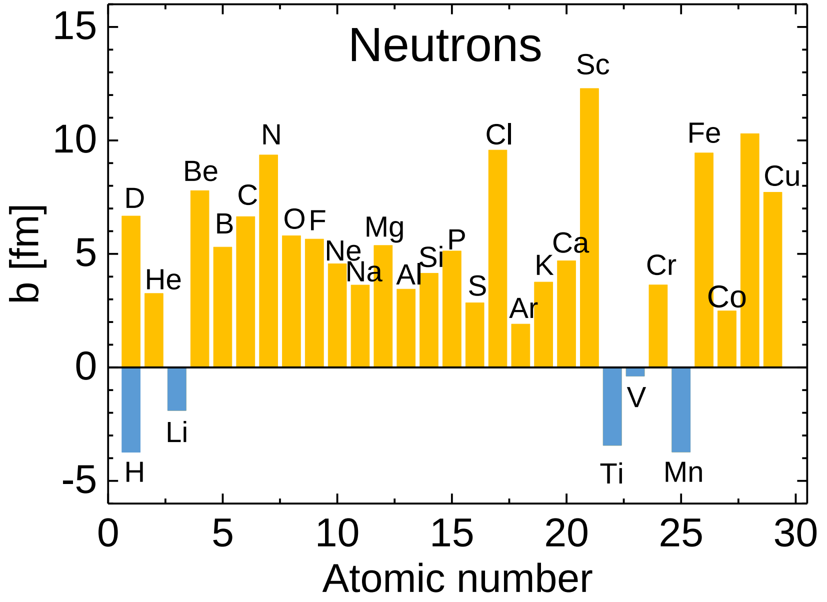
- Neutron Reflectivity (index of refraction) depends on Scattering Length Density (SLD)

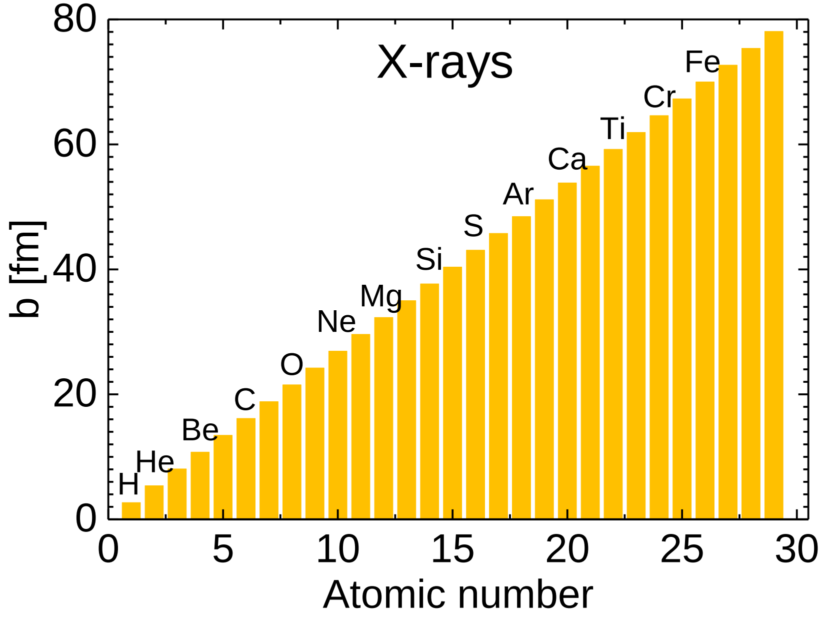
- $SLD \ = \sum_{i=1}^{k} N_i b_i$

Atomic number density

Scattering Length

- Neutrons interact with atomic nuclei
  → Scattering length is isotope dependent
  → Unlike many other techniques, sensitive to light elements such as H and Li





Need a Neutron Reflectometer!

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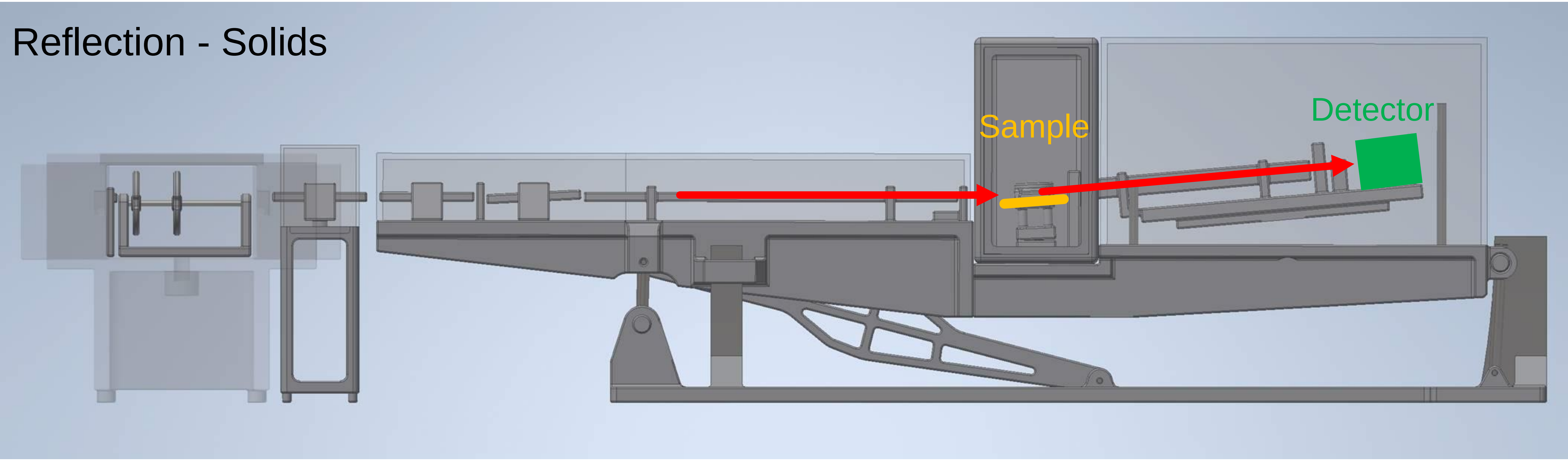
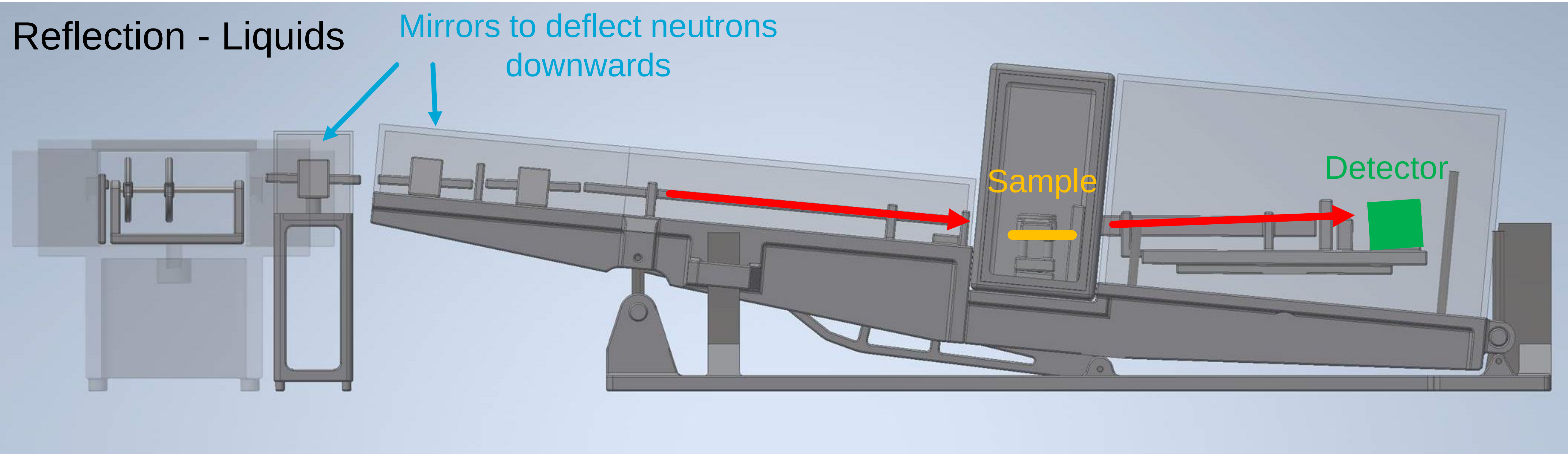
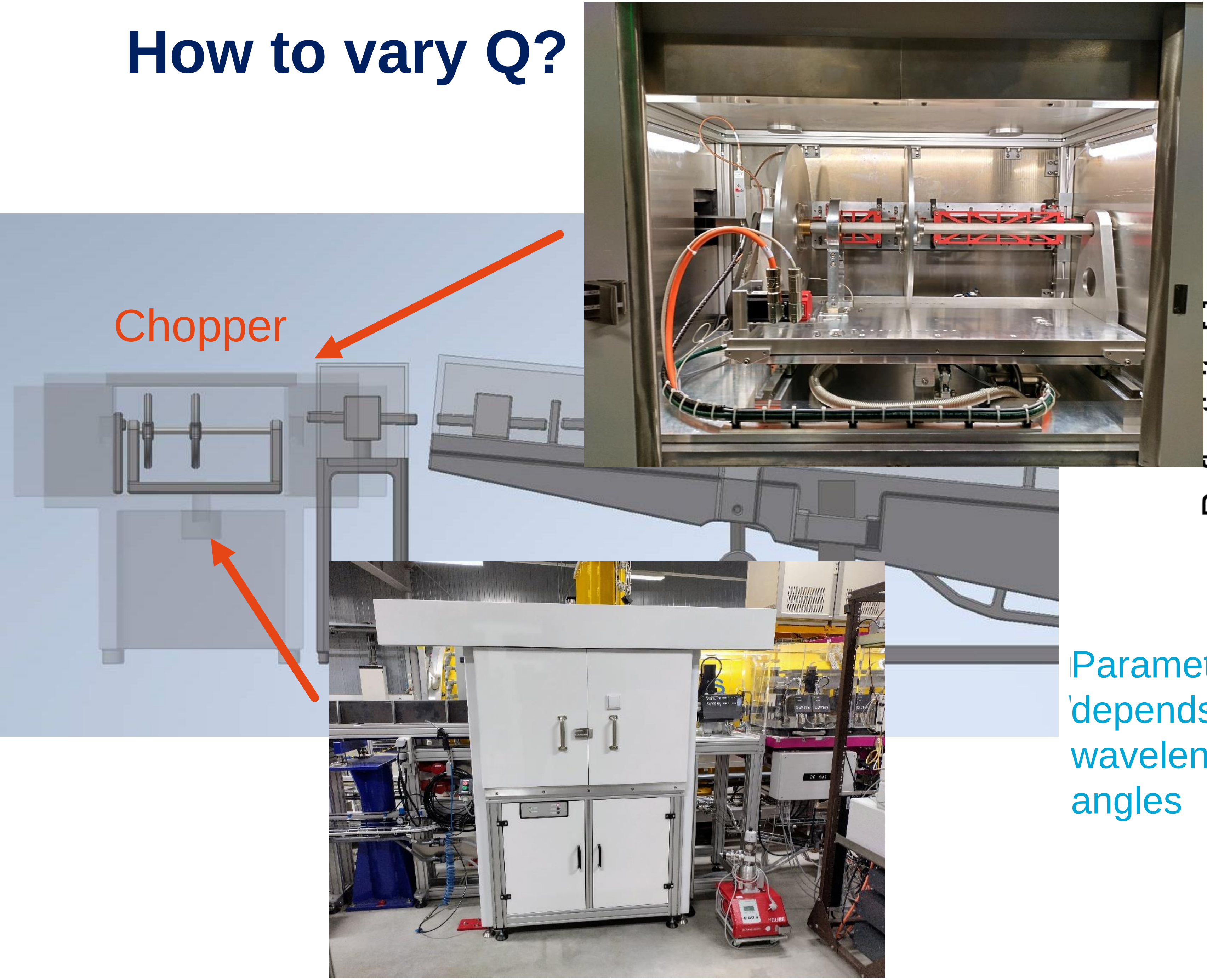
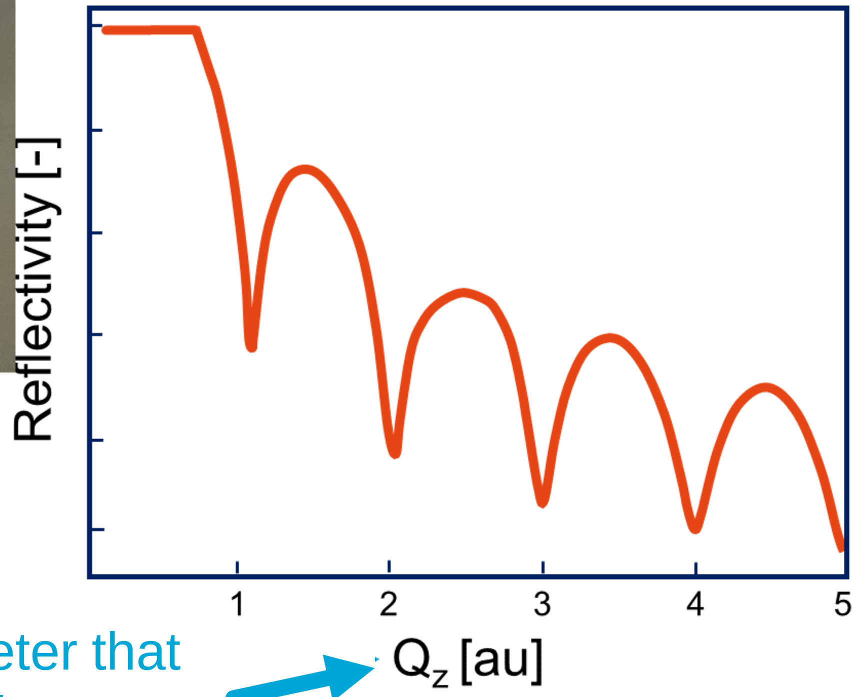


Need a Neutron Reflectometer!



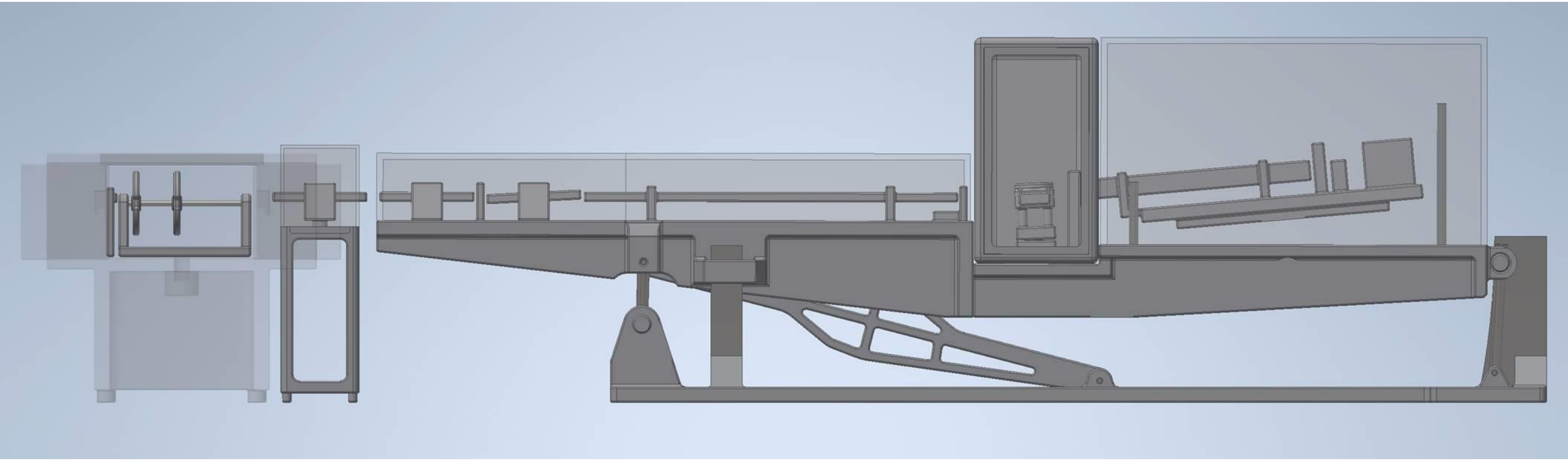
How to vary Q?





Parameter that depends on wavelength or incident angles

How to vary Q?



λ = 0.1 nm

λ = 1 nm

Start

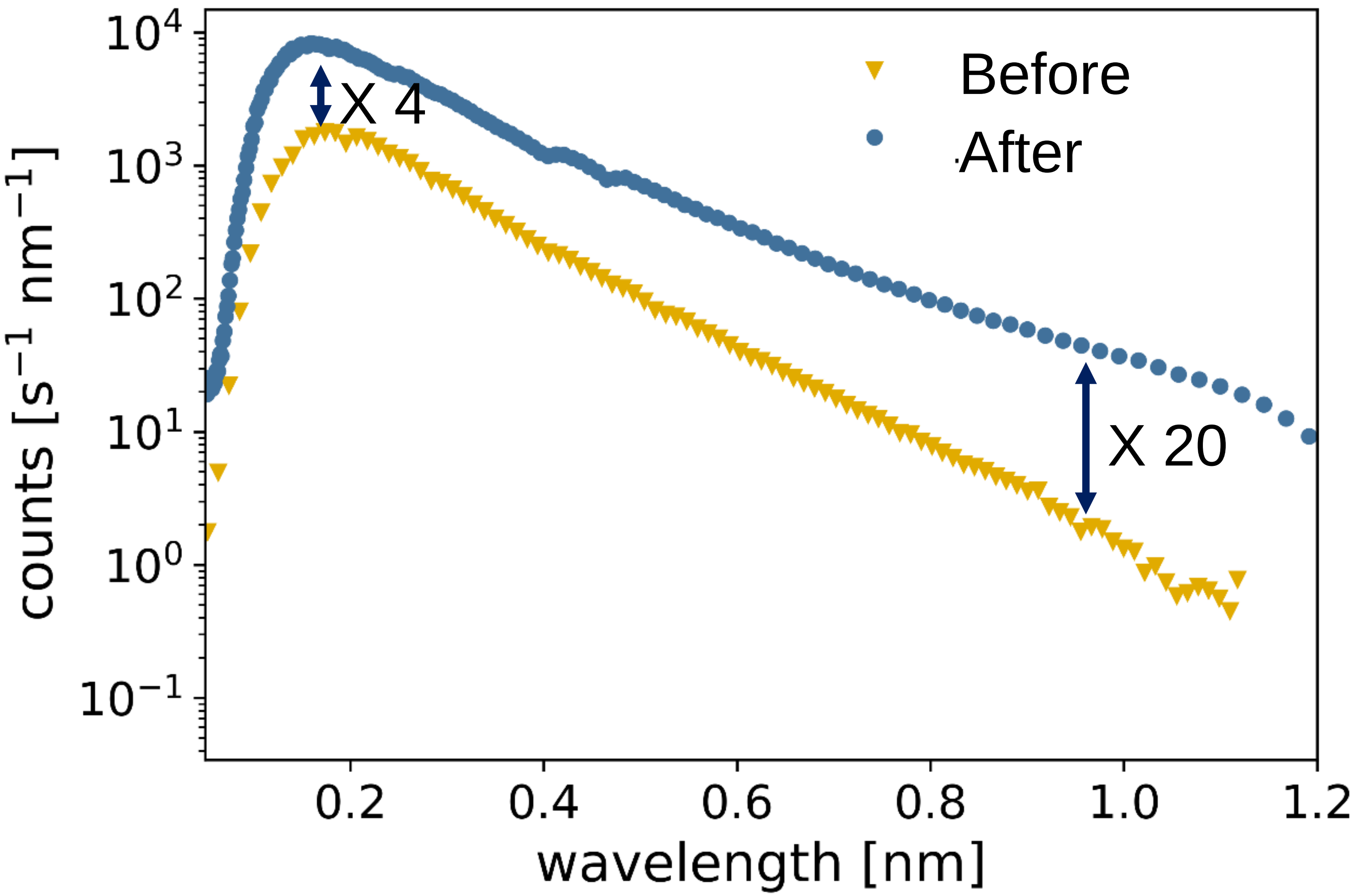
Stop

Recording the time-of-flight allows one to determine the wavelength!

Upgrade and Improvements

- Relocation to guide hall (=lower background).
- New neutron guides.
- Guides in the chopper.
- Guides under vacuum along the entire beamline
- Improved shielding.
- New chopper with variable disc-distance. More flexible when choosing the resolution.

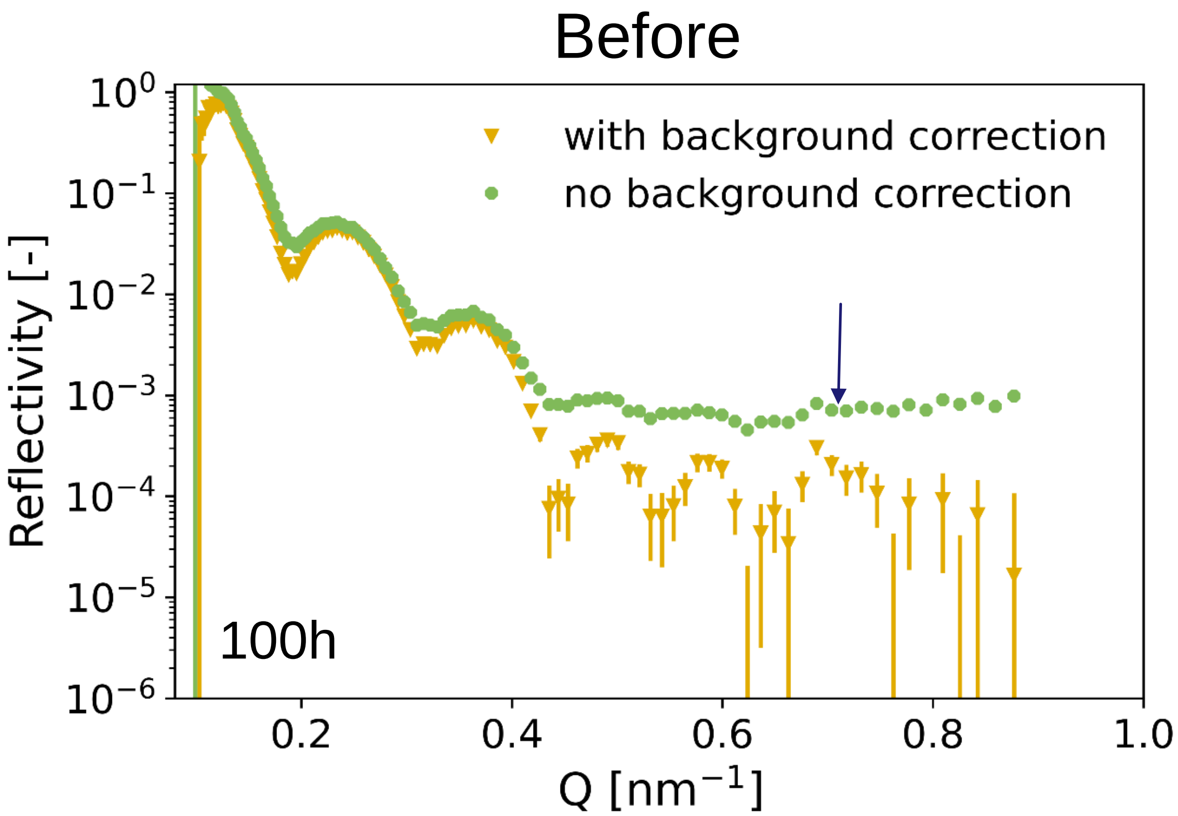
Before vs After

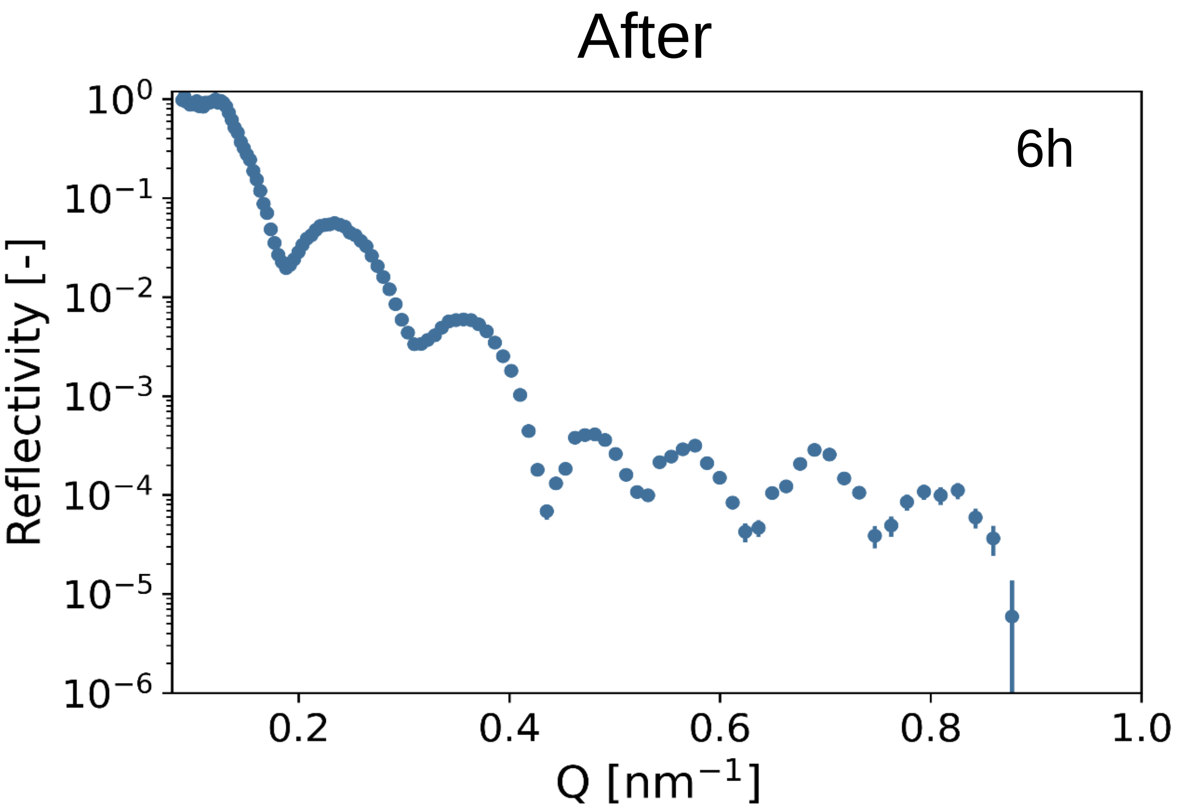


Conclusions

- Much larger intensity, especially at longer wavelengths!

Before vs After



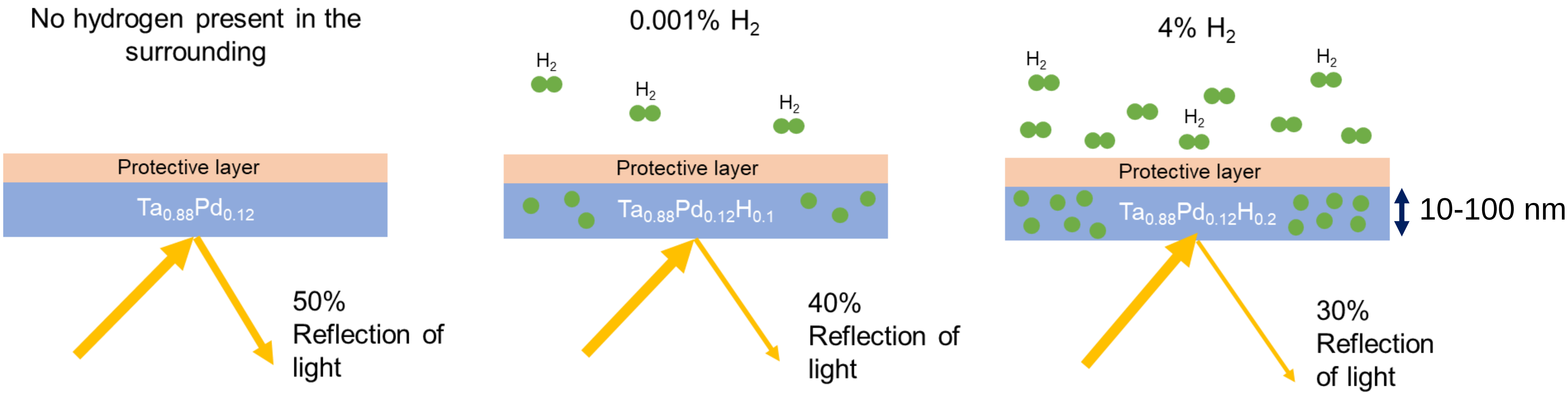


Conclusions

- Much faster measurements
- Lower background allows one to measure lower reflectivity (before: ≈ 10^{-3})

Pd-capped Hf thin film measured at 8.5 mrad.

Example: Hydrogen sensing materials



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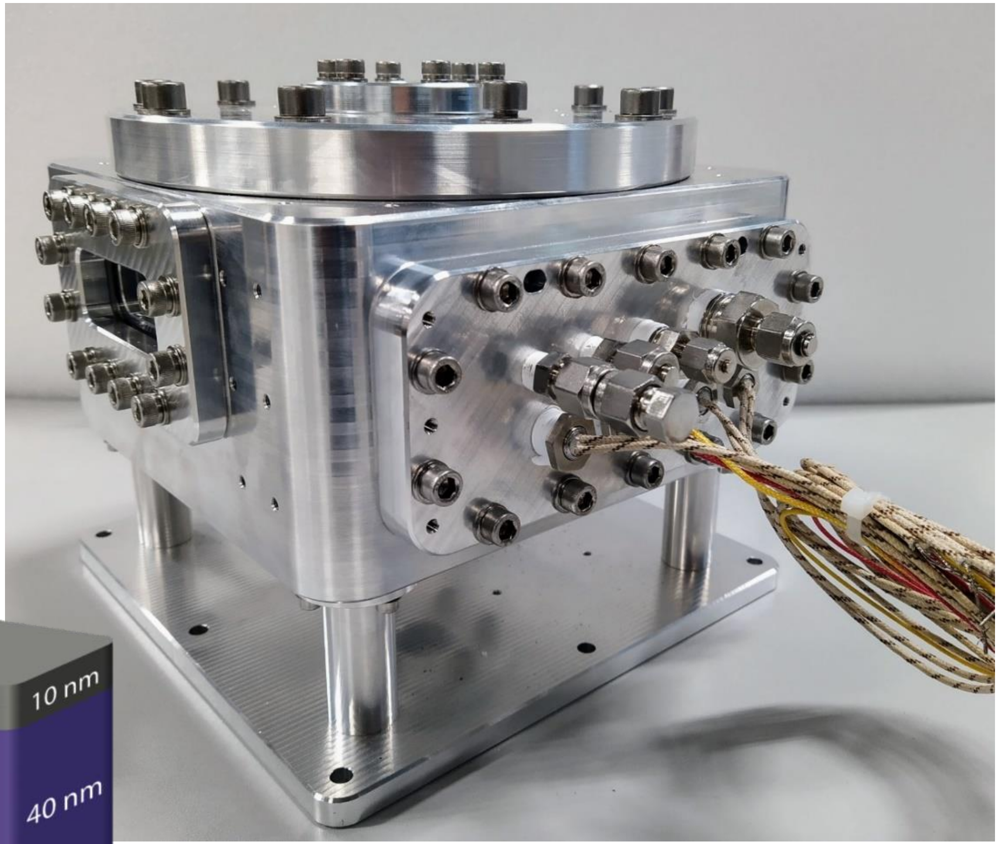
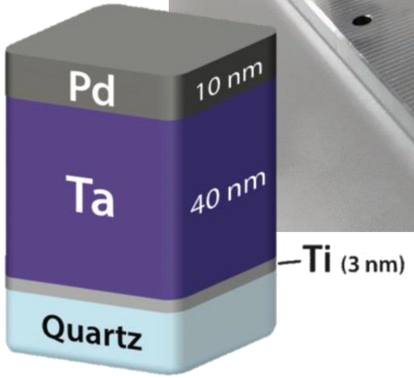
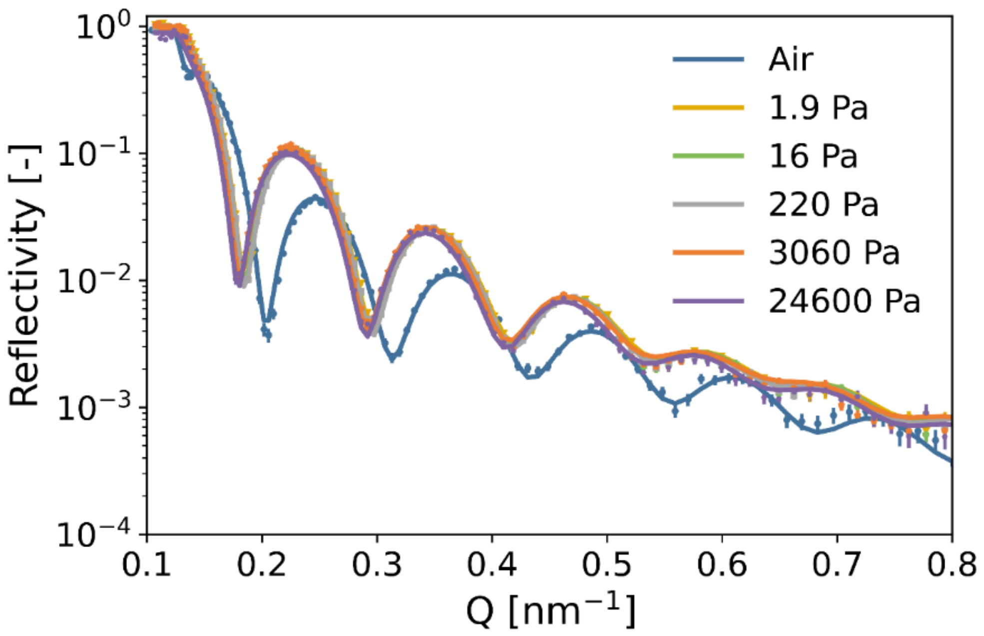
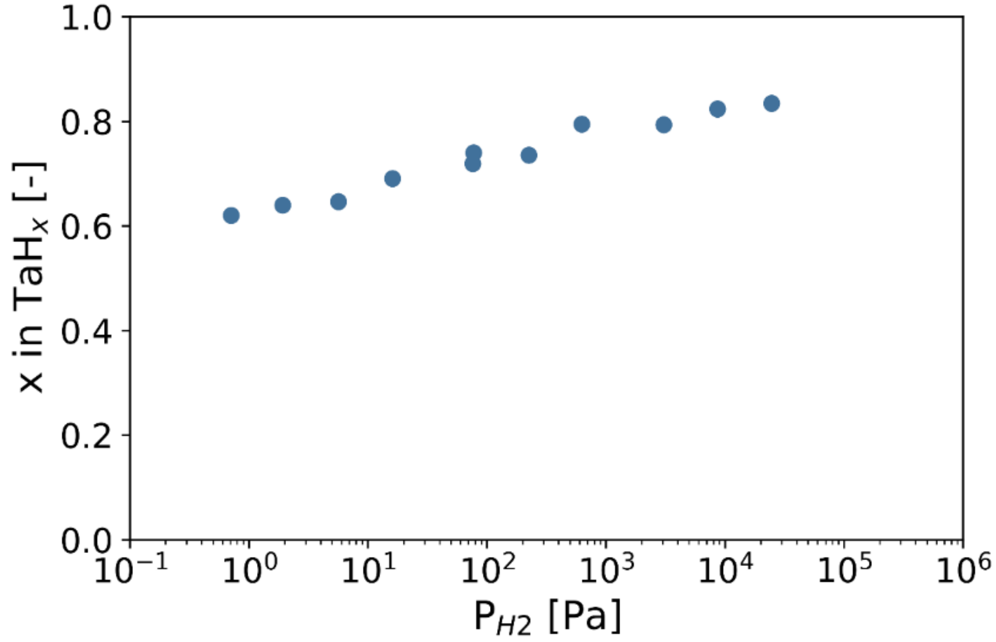








First publication of the ‘New’ ROG

ACS MATERIALS LETTERS

www.acsmaterialsletters.org

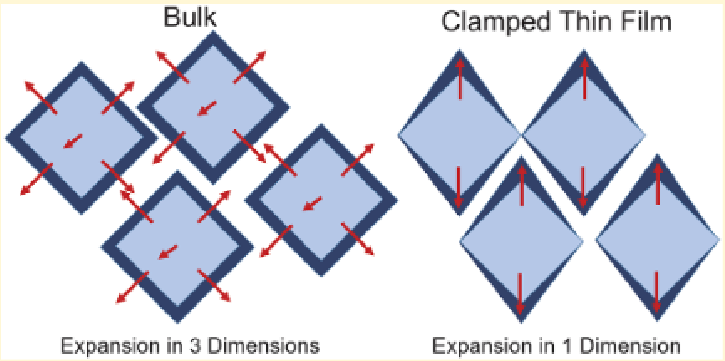
LETTER

Completely Elastic Deformation of Hydrogenated Ta Thin Films

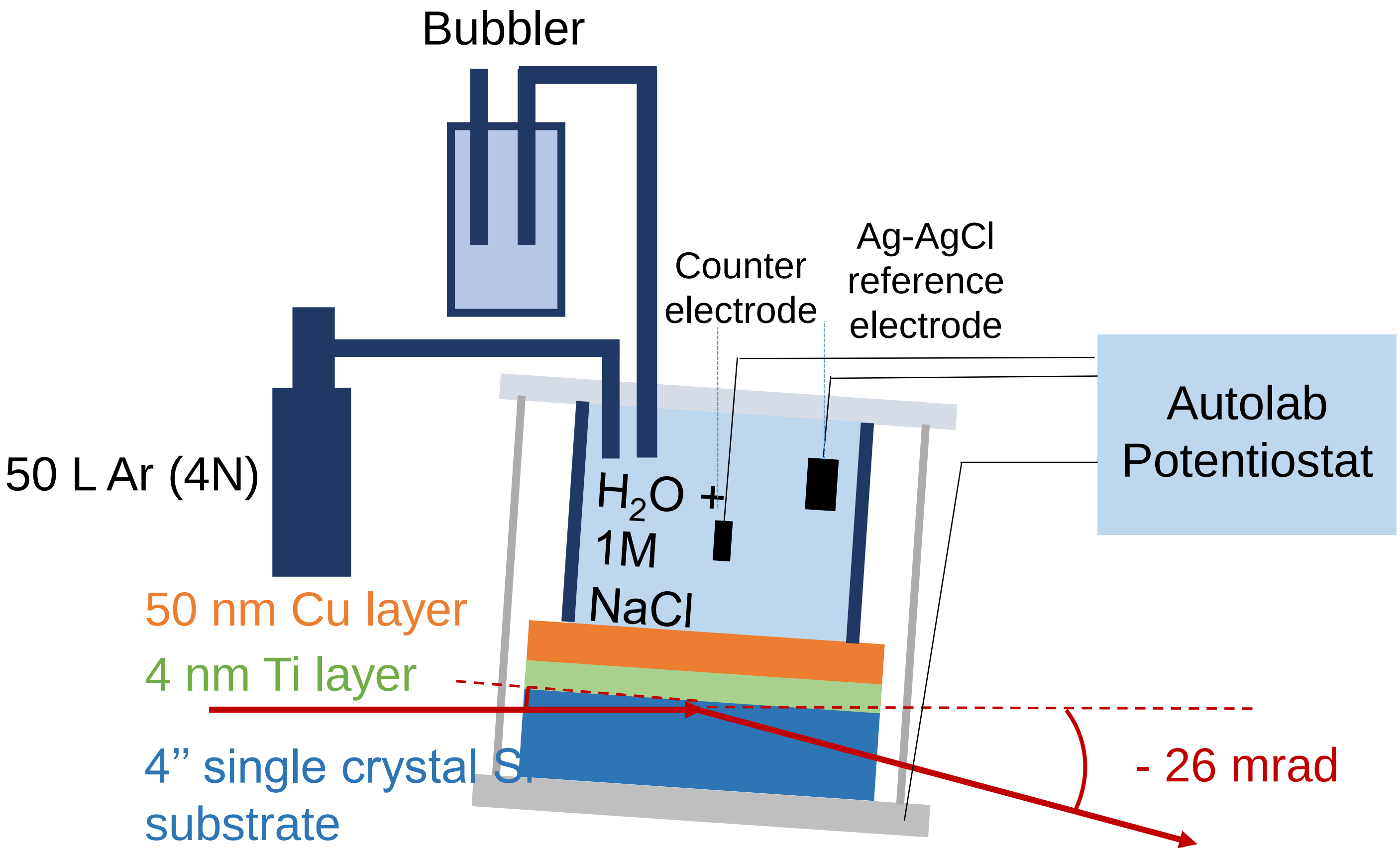
Lars J. Bannenberg,* Larissa Blom, Kouji Sakaki, Kohta Asano, and Herman Schreuders

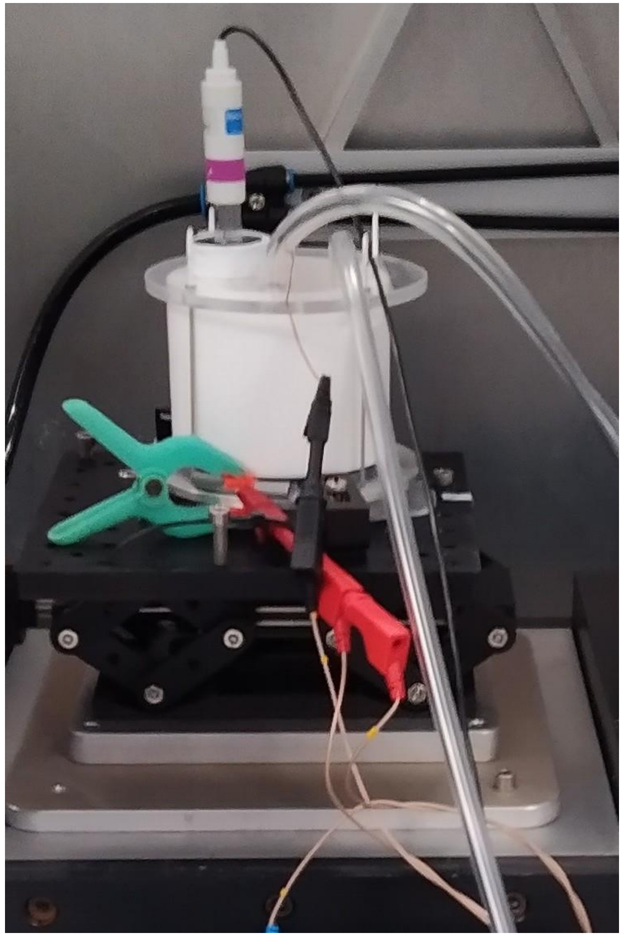
Cite This: *ACS Materials Lett.* 2023, 5, 962–969

ABSTRACT: Nanostructured metal hydrides could play a key role in a hydrogen economy. The nanostructuring or confinement of these materials as, e.g., thin films significantly affects the structural and functional properties. For tantalum hydride, a versatile hydrogen sensing material, we show that the confinement of tantalum as a thin film extends the solubility limit by suppressing the phase transition observed in bulk upon hydrogenation. Different from bulk, the body centered cubic unit cell continuously deforms with unequal lattice constants and angles between lattice vectors. This deformation ensures that the volumetric expansion is realized in the out-of-plane direction, and surprisingly, completely elastic in nature. The first-order phase transition suppression combined with the continuous elastic deformation of the tantalum unit cell over an extraordinary wide solubility range ensures the superb performance of tantalum and its alloys as a hysteresis-free optical hydrogen sensing range over a hydrogen pressure/concentration range of over 7 orders of magnitude.



Example: Corrosion of materials for nuclear waste storage

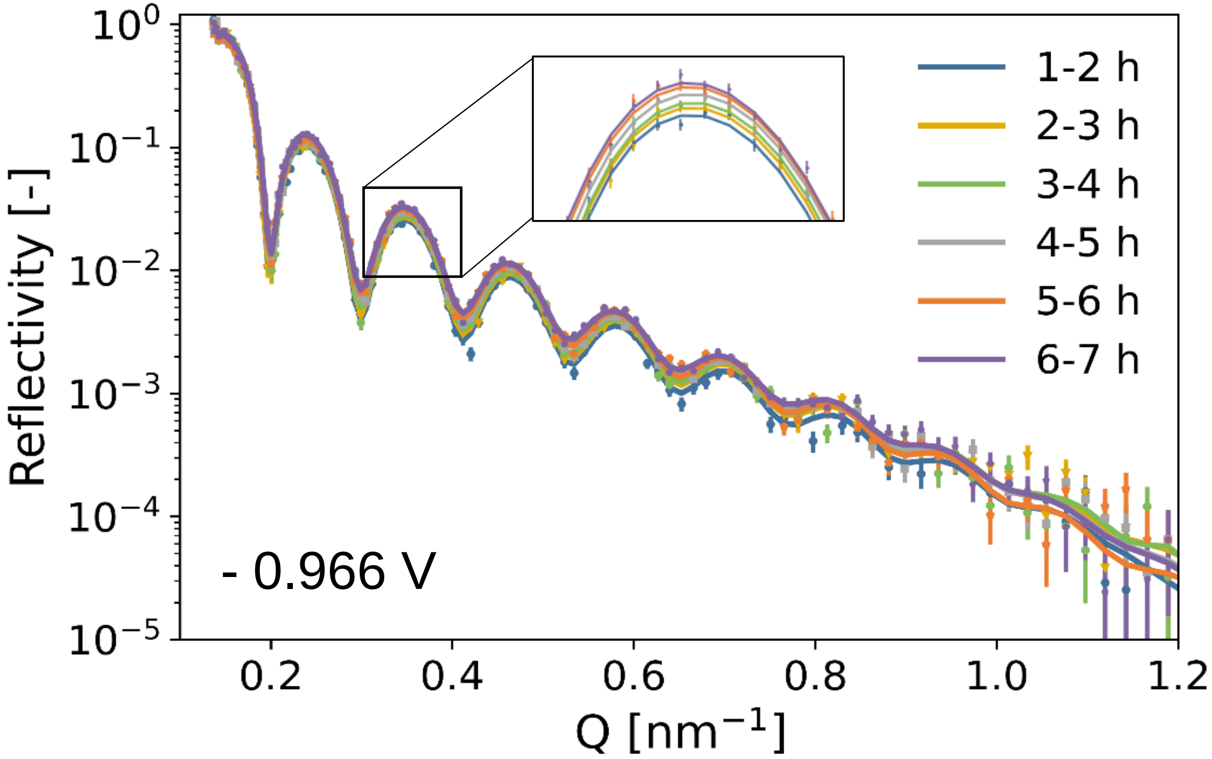


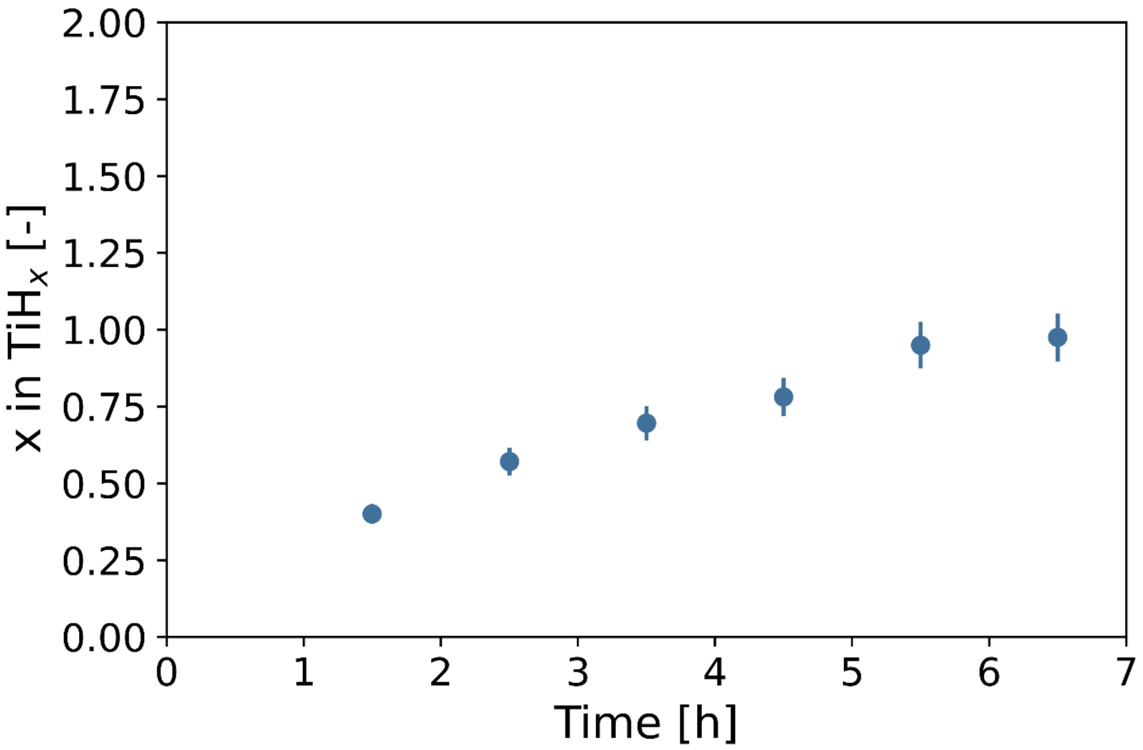


Ti layer serves as an indicator layer to observe H permeation through the Cu layer

Arthur Situm, Behrouz Bahadormanesh, Lars J Bannenberg, Frans Ooms, Hunter A Feltham, German Popov, Lyudmila V Goncharova, James J Noël, Hydrogen absorption into copper-coated titanium measured by in situ neutron reflectometry electrochemical impedance spectroscopy, Journal of The Electrochemical Society, 2023

Example: Corrosion of materials for nuclear waste storage





→ When a potential is applied, hydrogen can permeate the copper layer to the Ti indicator layer

Example: Passivating contacts for Solar cells

Passivating contacts are used in solar cells to prevent early recombination of holes and electrons that decreases the efficiency of solar cells.

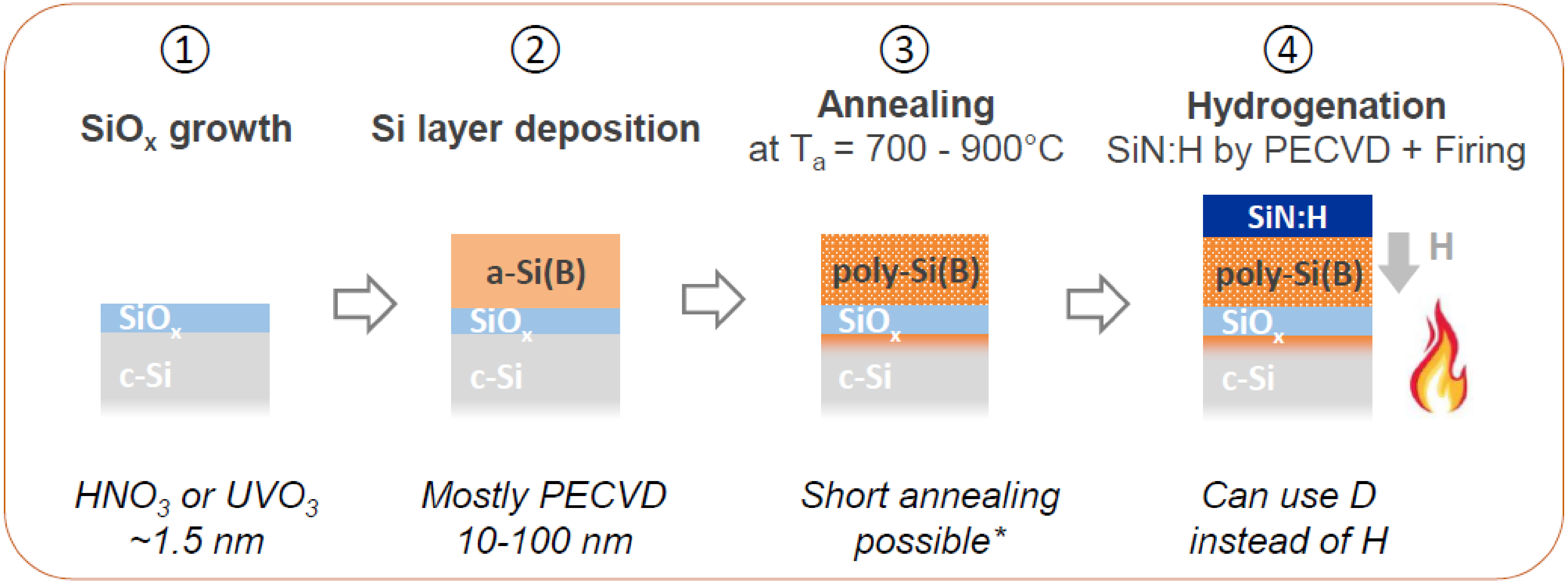


Figure from: Morisset, A., Famprikis, T., Haug, F. J., Ingenito, A., Ballif, C., & Bannenberg, L. J. (2022). In Situ Reflectometry and Diffraction Investigation of the Multiscale Structure of p-Type Polysilicon Passivating Contacts for c-Si Solar Cells. ACS Applied Materials & Interfaces, 14(14), 16413-16423.

Other applications

- Study interfaces in batteries during operation
- Fuel cell materials
- Coatings (for dental implants)
- Polymers
- Proteins

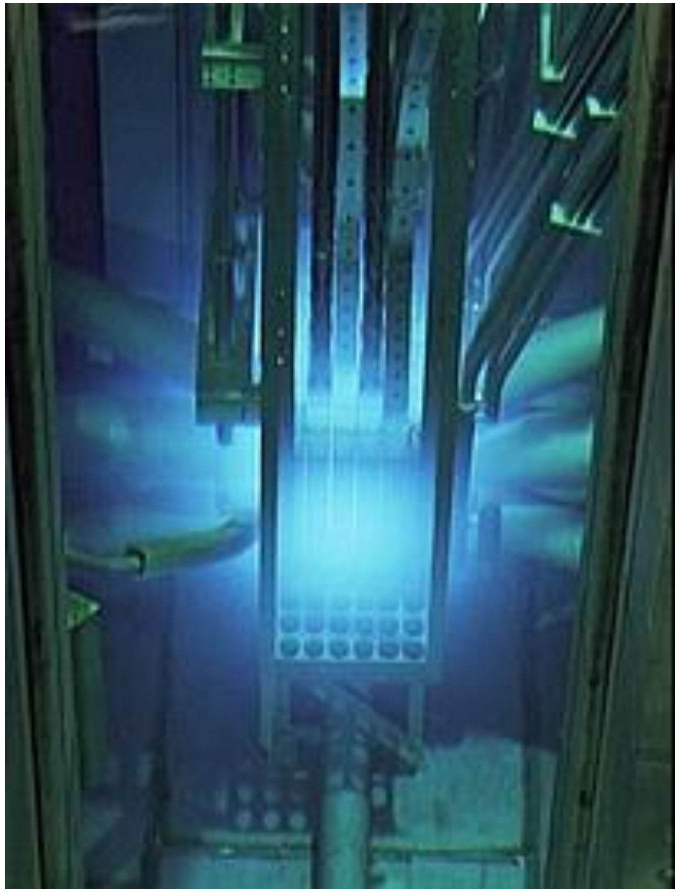
Cold Source

- As part of the Oyster program, a cold neutron ‘source’ will be installed.

- Effectively, it is a freezer that cools neutrons to about -240 °C.
  → Increases wavelength and ensures more ‘usable’ neutrons for the reflectometer and SANS

- ROG is fully ready to receive the ‘cold’ neutrons, only short commissioning is expected after installation.

- Expected increase in intensity by at least a factor of 10



Summary

- Upgrade of the ROG resulted in a factor of 6 larger intensity, reduction of background by a factor of 50 and thus shorter measurement times and higher quality data.
- First three papers published.
- ‘Friendly’ users from Canada, United Kingdom, Germany, the Netherlands and France have measured at the ROG, some visited multiple times.
- 2 Commercial Assignments
- Future is looking bright when the cold source is installed.

Team effort

- Research Engineers
  - Essential for construction, maintenance and measurements

- (Project) Engineers from Demo

Ad	Malte
Ernst	Martin
Esther	Michel
Frans	Piet
Fred	Raymon
Herman	Rien
Jeroen	William
Kees	*All users and collaborators*
	and many more