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Materials analysis by ion beams - advanced complementary methods in materials characterization

Daniel Primetzhofer – Uppsala University

Tandem Laboratory
Uppsala University

The Tandem Laboratory @ Uppsala University

A national infrastructure for world-leading ion beam-based research in:

- Accelerator Mass Spectrometry - AMS
- Ion Beam Modification of Materials - IBMM
- Ion Beam Analysis - IBA

Supported by:

Swedish Foundation for Strategic Research

The Swedish Research Council

EU - Eurofusion

Consortium: Uppsala University, Linköping Universitet & KTH

The Tandem Laboratory @ Uppsala University

Recent investments in infrastructure

4 accelerators, 9 ion sources, 11 beam lines:

Operational time 2018: ~2000h

2018: ~2000h

2018: ~3000h

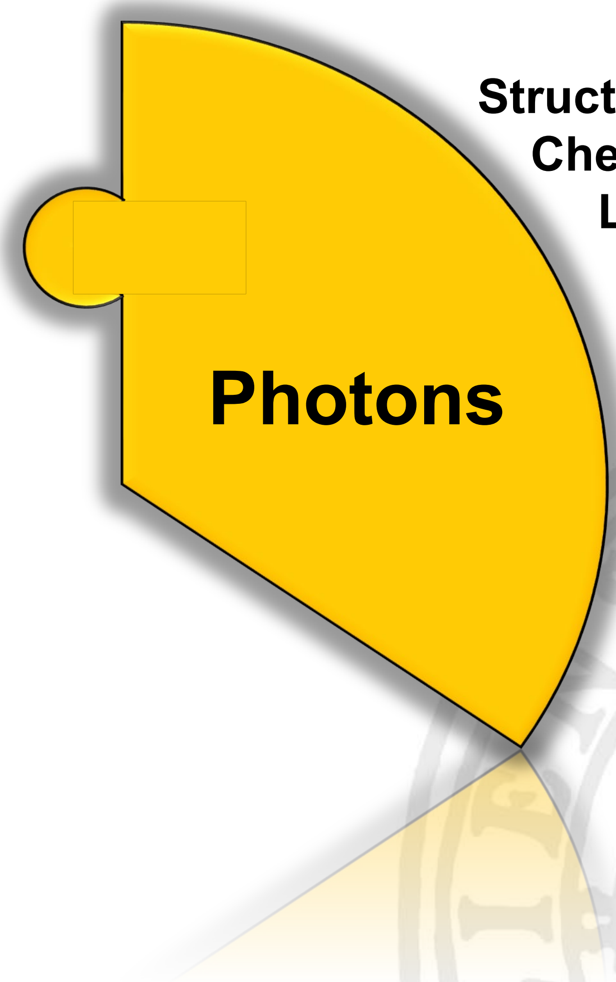
Under start-up

Tandem accelerator NEC 5 MV Pelletron ('01)	**IB**
Linear 350 kV Danfysik	**IBA, IBMM**
MICADAS - 170 kV, ETH ('14)	**AMS**
Dedic ple preparation	**AMS, IBA, IBMM**
Low-Energy Ion Scattering set-up ('18)	**Under start-up**

Continual investments in experimental end-stations

The Tandem Laboratory @ Uppsala University

What is our complementary offer?



Structural Analysis
Chemical information
Layer thickness
….

The Tandem Laboratory @ Uppsala University

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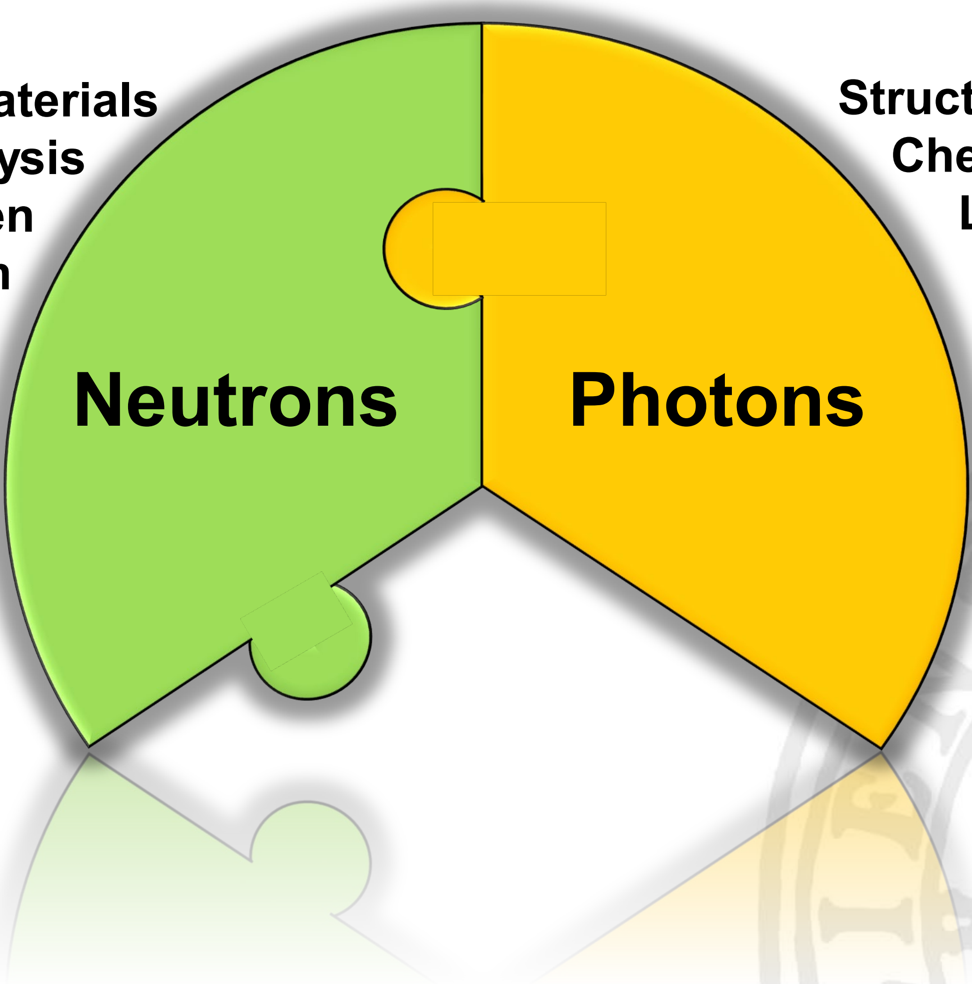

Amorphous materials
Structural Analysis
Hydrogen
Magnetism
Gases & liquids
….

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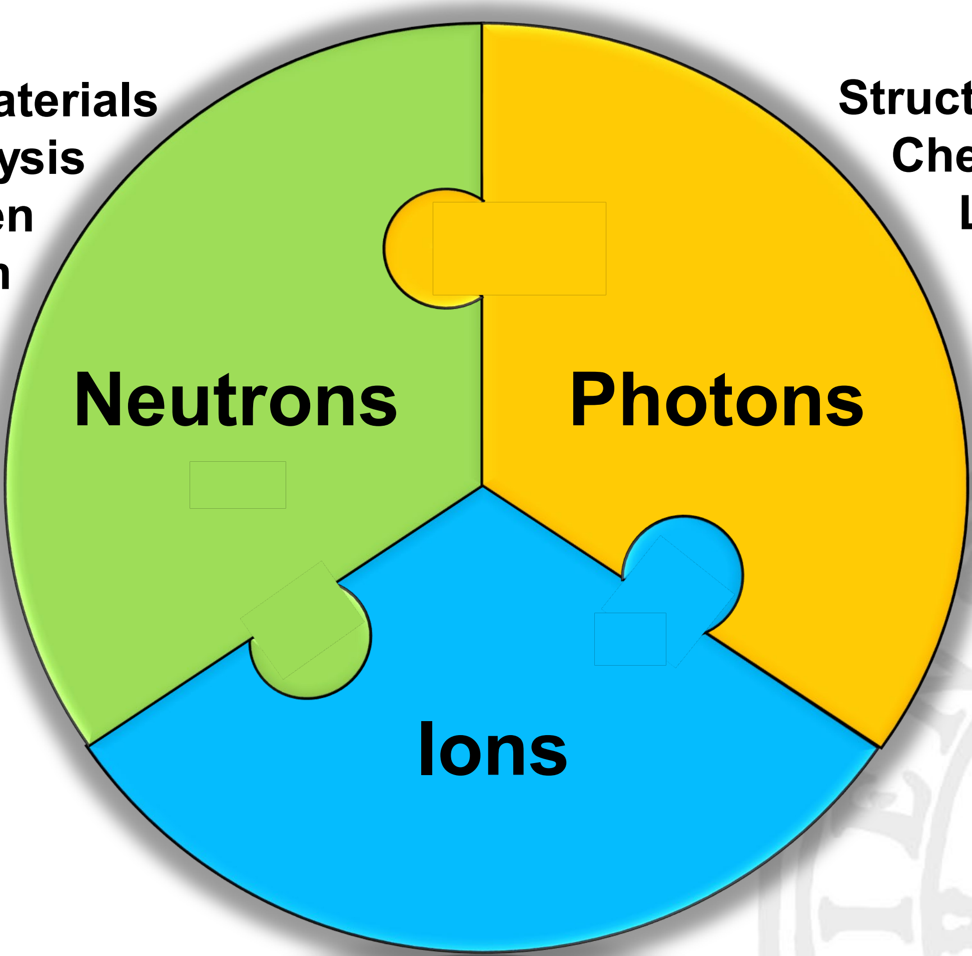
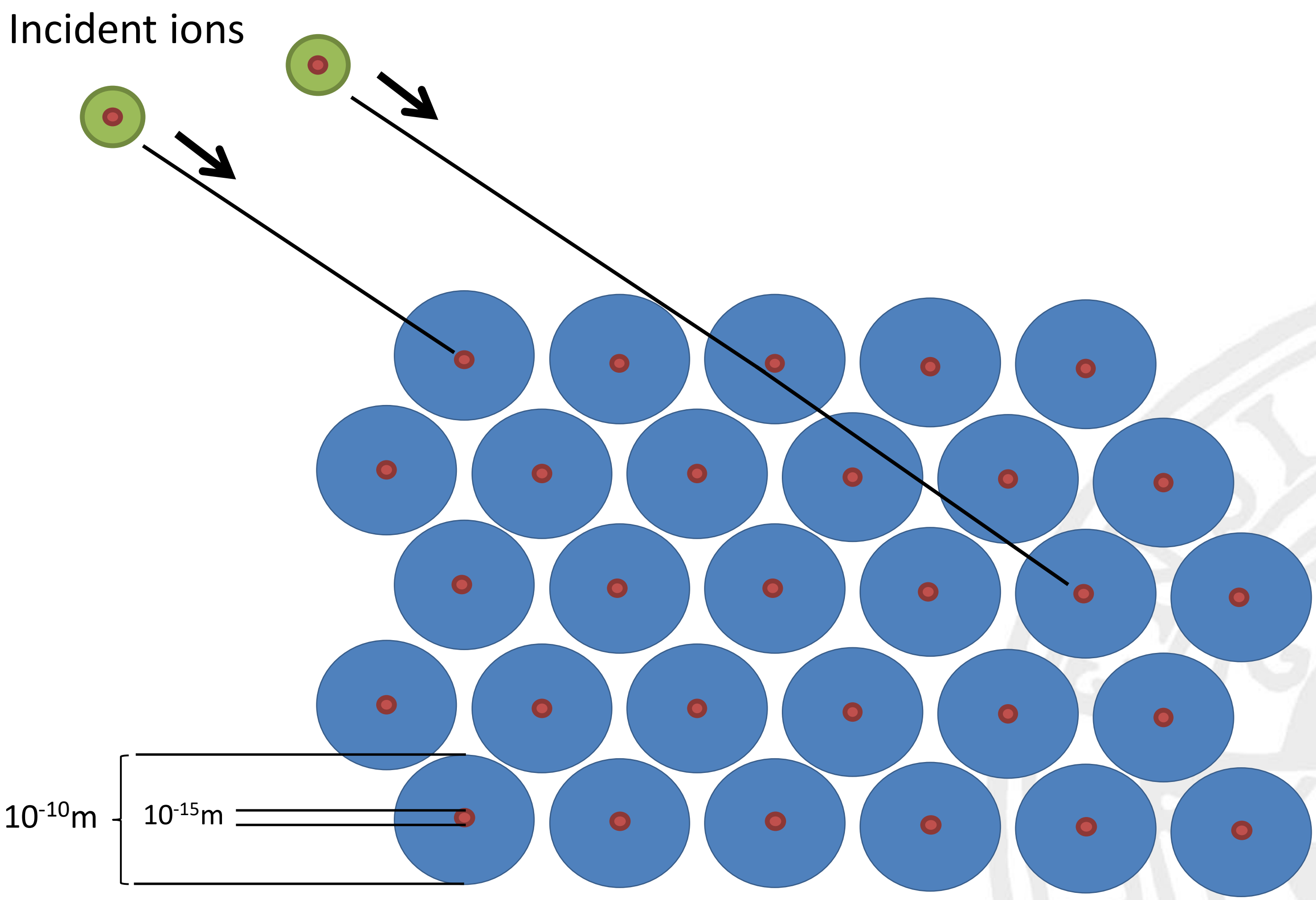


***Depth-resolved*, quantitative & non-destructive composition analysis**

Ion-solid interaction in a nutshell



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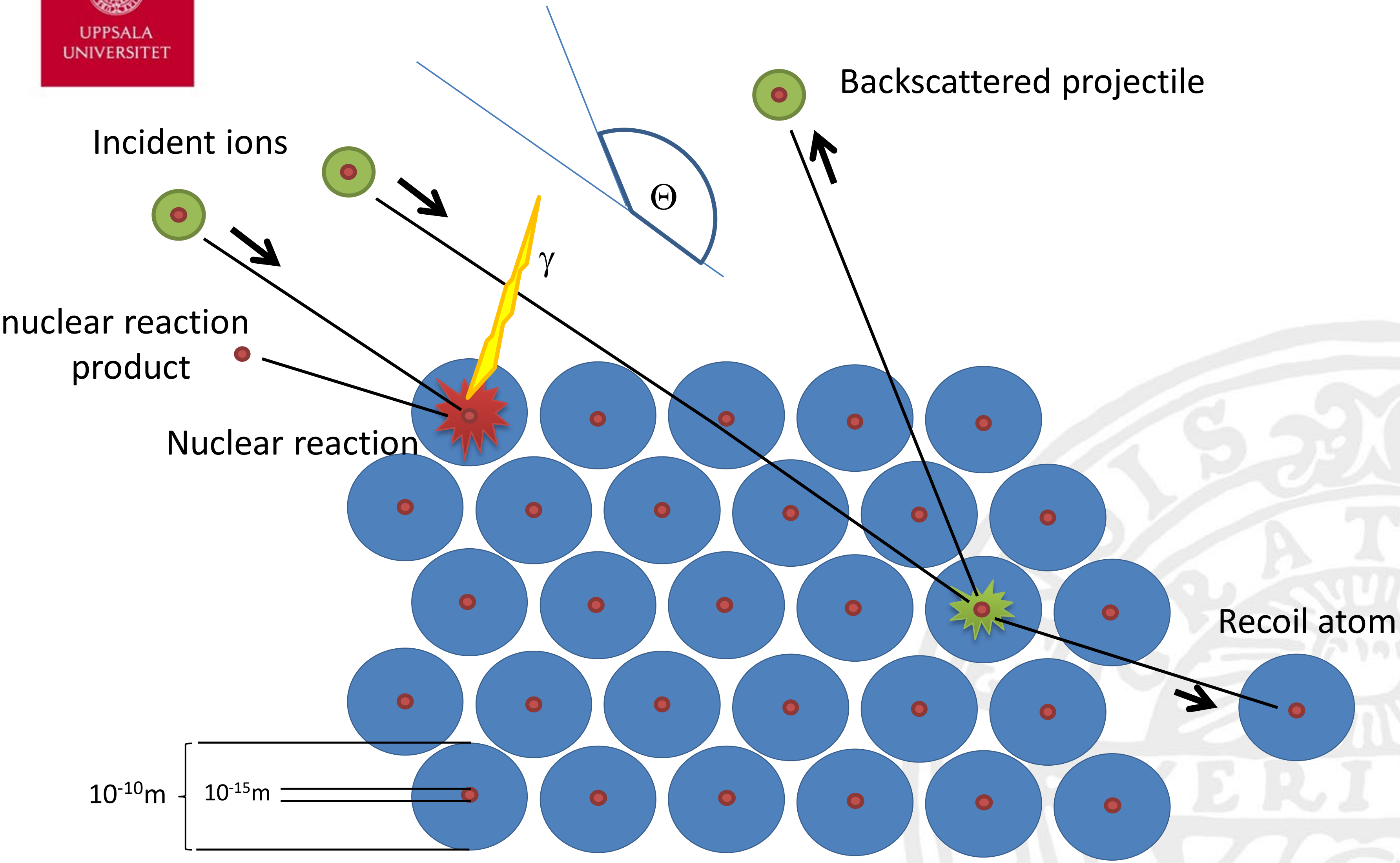

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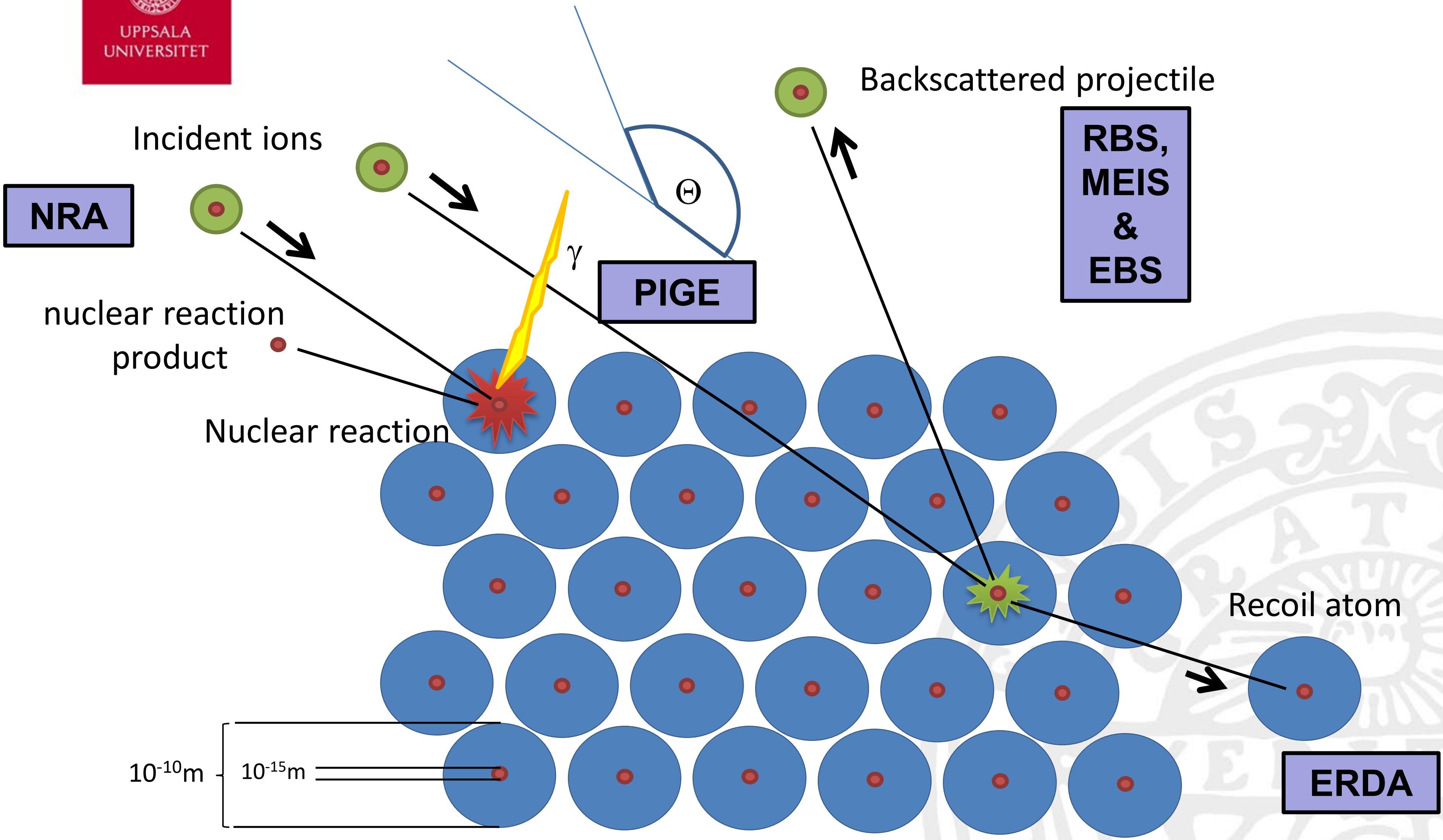

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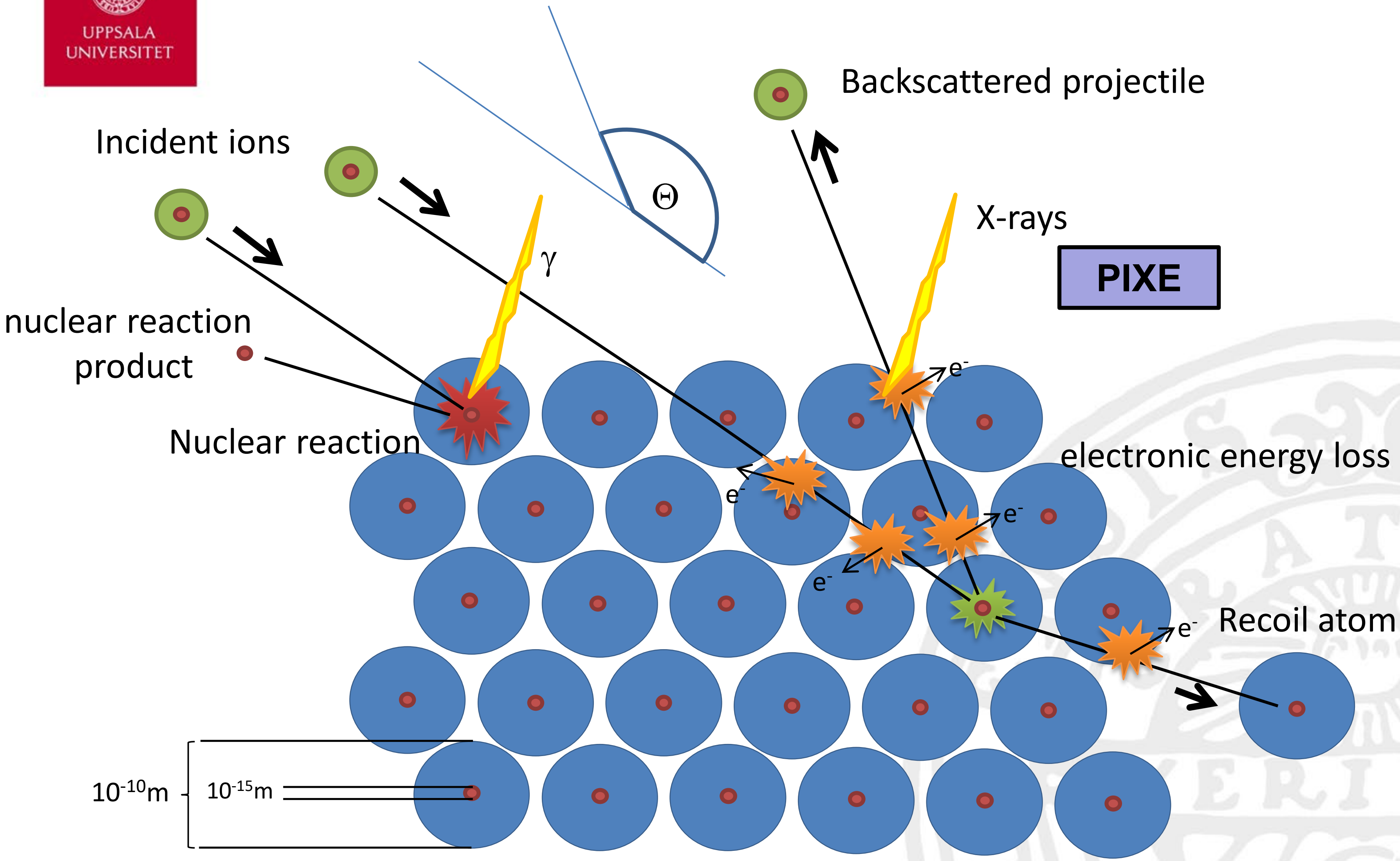

Composition depth profiling by ion beams

Some common properties

- Interaction with nuclei yields information on composition
  - **No matrix effects** – electronic system is not affecting quantification
  - Dependent on specific methods **all elements/isotopes** can be traced
- Interaction with electrons yields depth perception
  - Large number of minor interactions – **global description possible**
  - Straight trajectories of particles – **single scattering models applicable**

Highly quantitative tools

Composition depth profiling by ion beams

Some common properties

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- Interaction with electrons yields depth perception
  - Large number of minor interactions – **global description possible**
  - Straight trajectories of particles – **single scattering models applicable**

- Interaction with electrons can also yield information on composition
  - Via X-rays – limited depth information
  - **Extreme sensitivity** due to absence of Bremsstrahlung (up to 200x higher than EDX)

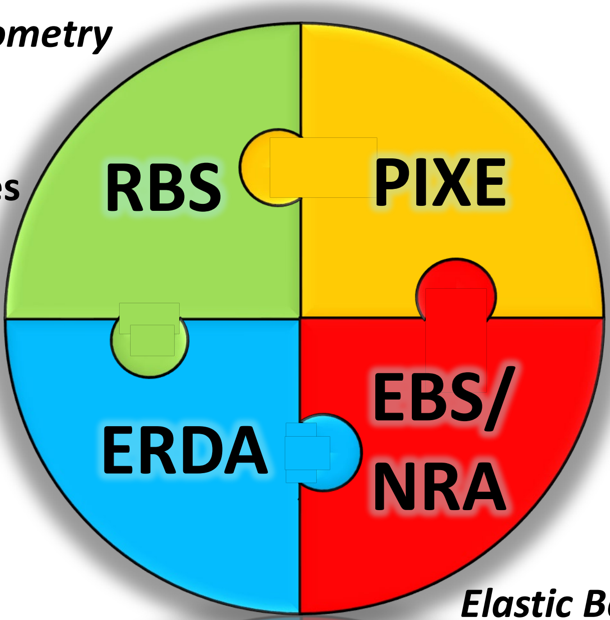
- Interaction in large volume with small number of probing species
  - 10^{13} probing particles interact with 10^{19} atoms in probing volume
  - **Non-destructive analysis** is possible

Composition depth profiling by ion beams

Different methods – different capabilities

Rutherford Backscattering spectrometry

- Elastic scattering of light ions
- High accuracy for heavy species



Particle Induced X-ray Emission

- Characteristic X-rays from sample
- High sensitivity for heavy species

Elastic Recoil Detection Analysis

- Ions recoiled by primary particles
- Sensitive for light & heavy species
- High accuracy

Elastic Backscattering Spectrometry/
Nuclear Reaction Analysis

- Light ions & nuclear potentials
- High sensitivity for light species (H, O, or C)
- Typically highest depth-resolution

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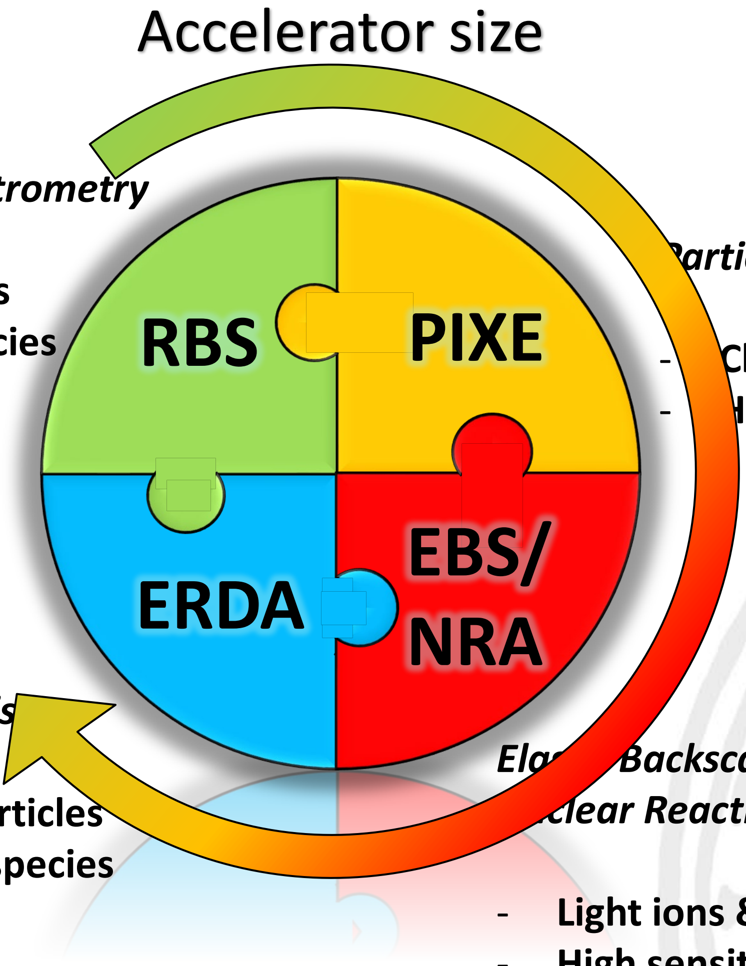
Composition depth profiling by ion beams

Different methods – different capabilities



Rutherford Backscattering spectrometry

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Ela Backscattering Spectrometry/
clear Reaction Analysis

- Light ions & nuclear potentials
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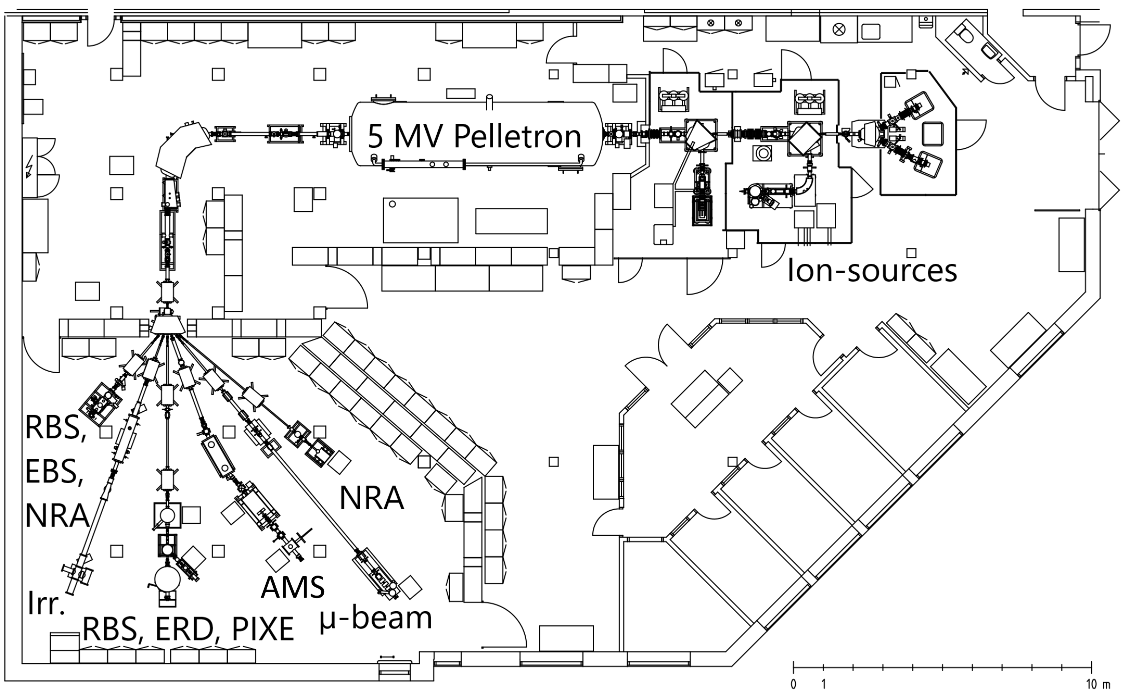
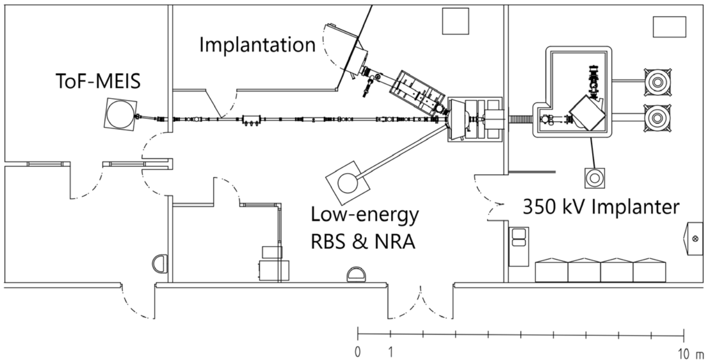
Equipment for IBA & IBMM – accessible for external users

5 MV 15-SDH2 pelletron accelerator



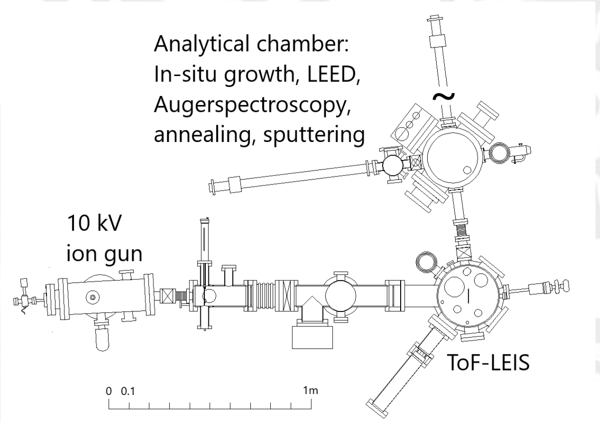
2 gas & 2 sputter ion sources – beams of H, D, ^{3}He, ^{4}He, C, N, O, Cu, Br, I, Au, …

350 kV Danfysik implanter



3 interchangeable sources: Gas, oven, sputter

ToF-LEIS system



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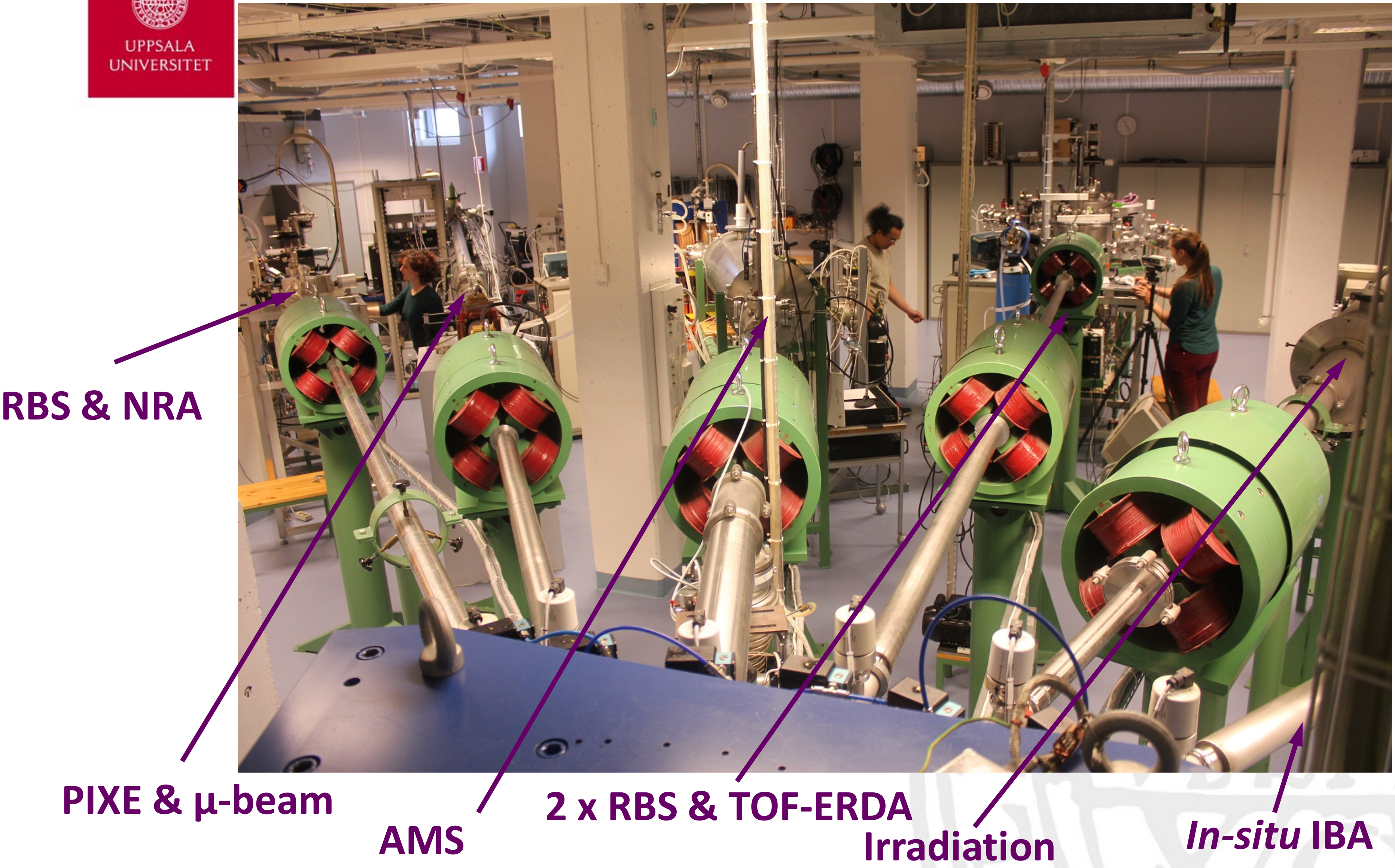


RBS & NRA

PIXE & µ-beam

AMS

2 x RBS & TOF-ERDA

Irradiation

***In-situ* IBA**

The Tandem Laboratory @ Uppsala University

5 MV pelletron accelerator

I. Applications

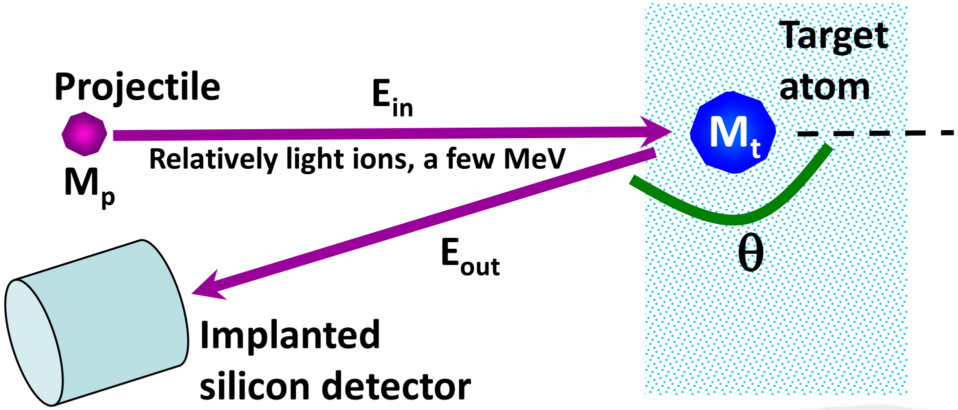
Rutherford Backscattering Spectrometry (RBS)

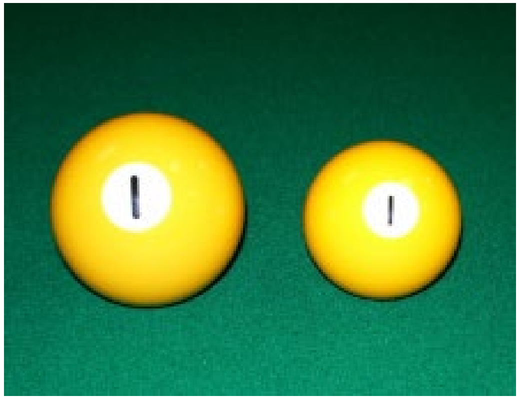
Method and applications

Measure energy of backscattered particles

⇩

Data on sample composition
+ depth information





Billard with unequal sized balls…

Rutherford Backscattering Spectrometry (RBS)

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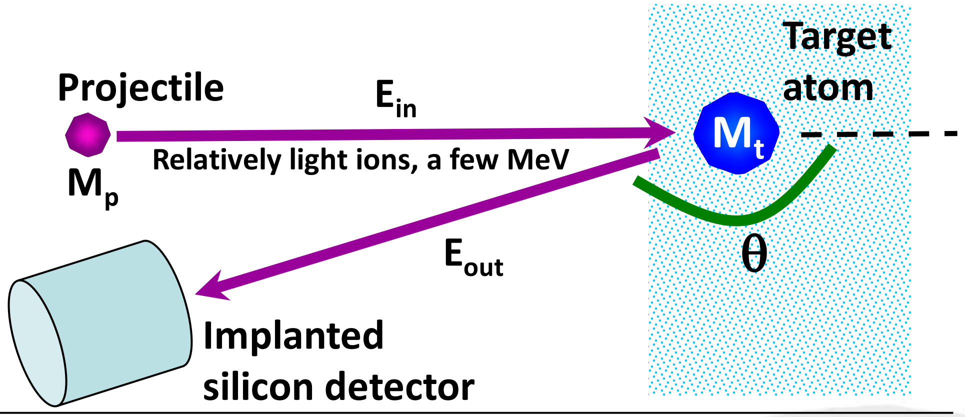


Projectile

M_p

E_{in}

Relatively light ions, a few MeV

Target atom

M_t

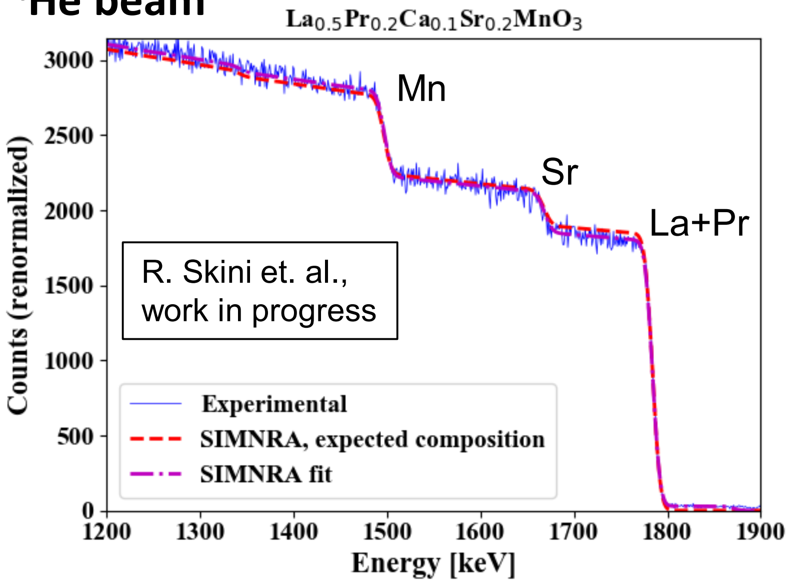
θ

E_{out}

Implanted silicon detector

---

^{4}He beam



Magnetocaloric material,
Composition of bulk sample

Bulk materials:

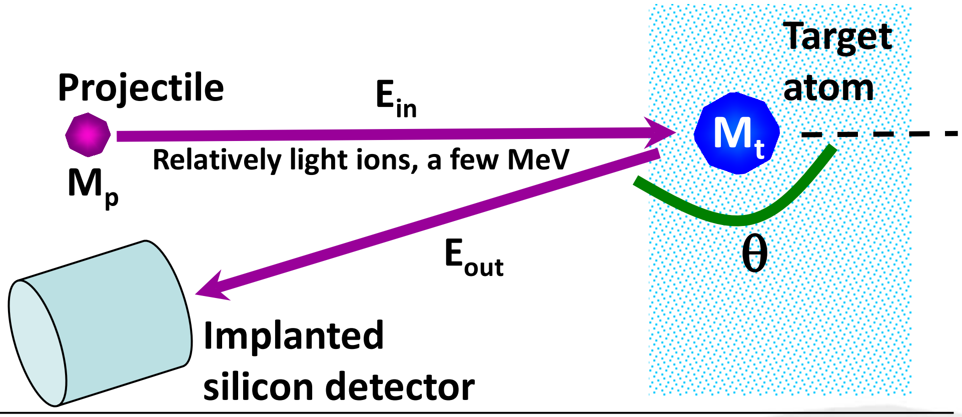
- **step position: mass/chemical element**
- **step height: atomic concentration**

Rutherford Backscattering Spectrometry (RBS)

Method and applications

Measure energy of backscattered particles

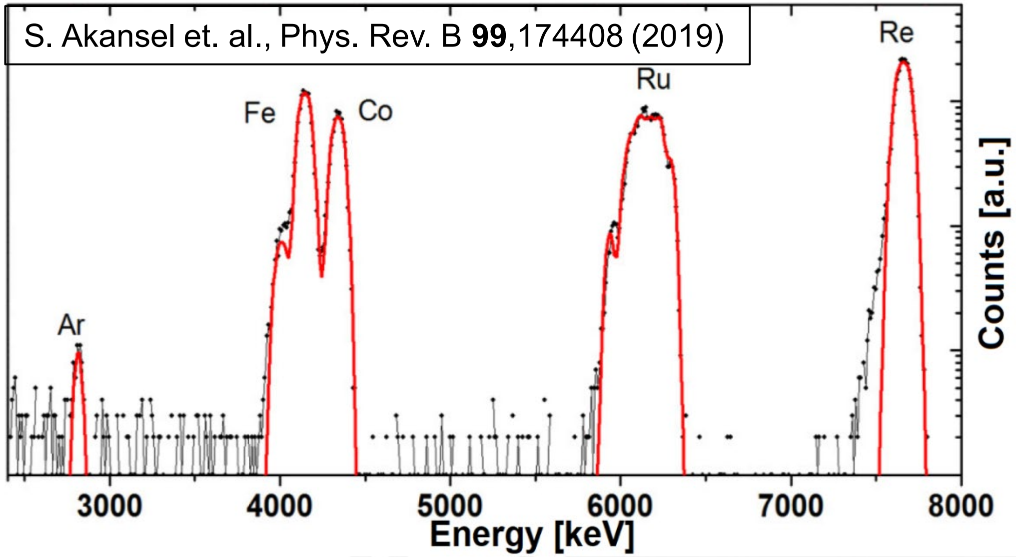
Data on sample composition
+ depth information



Thin films:

- peak position: mass/chemical element
- peak height: atomic concentration
- Proper beam: separation of e.g. Fe/Co

^{12}C beam


S. Akansel et. al., Phys. Rev. B **99**,174408 (2019)

Re-doped Fe_xCo_y thin films
on Ru on Si/SiO_2

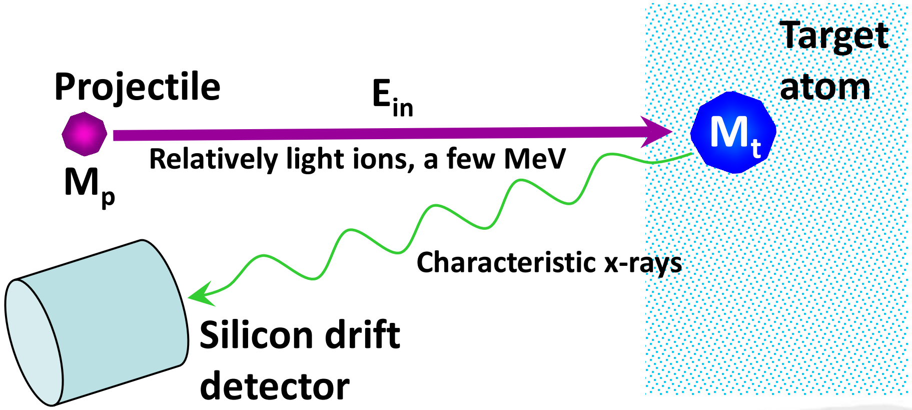
Particle Induced X-ray Emission (PIXE)

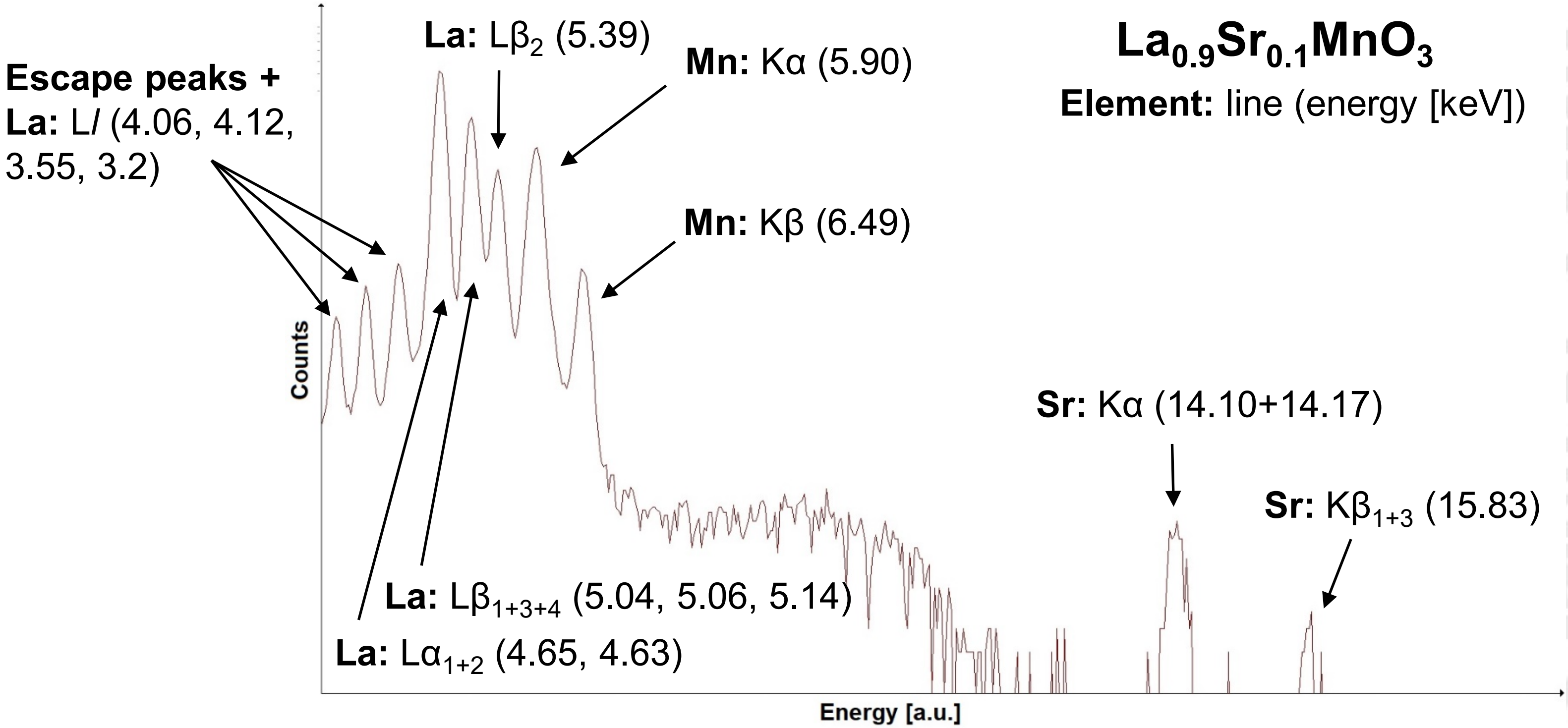
Method and applications

Characteristic X-rays

⇩

Quantification of impurities and overall composition without depth information



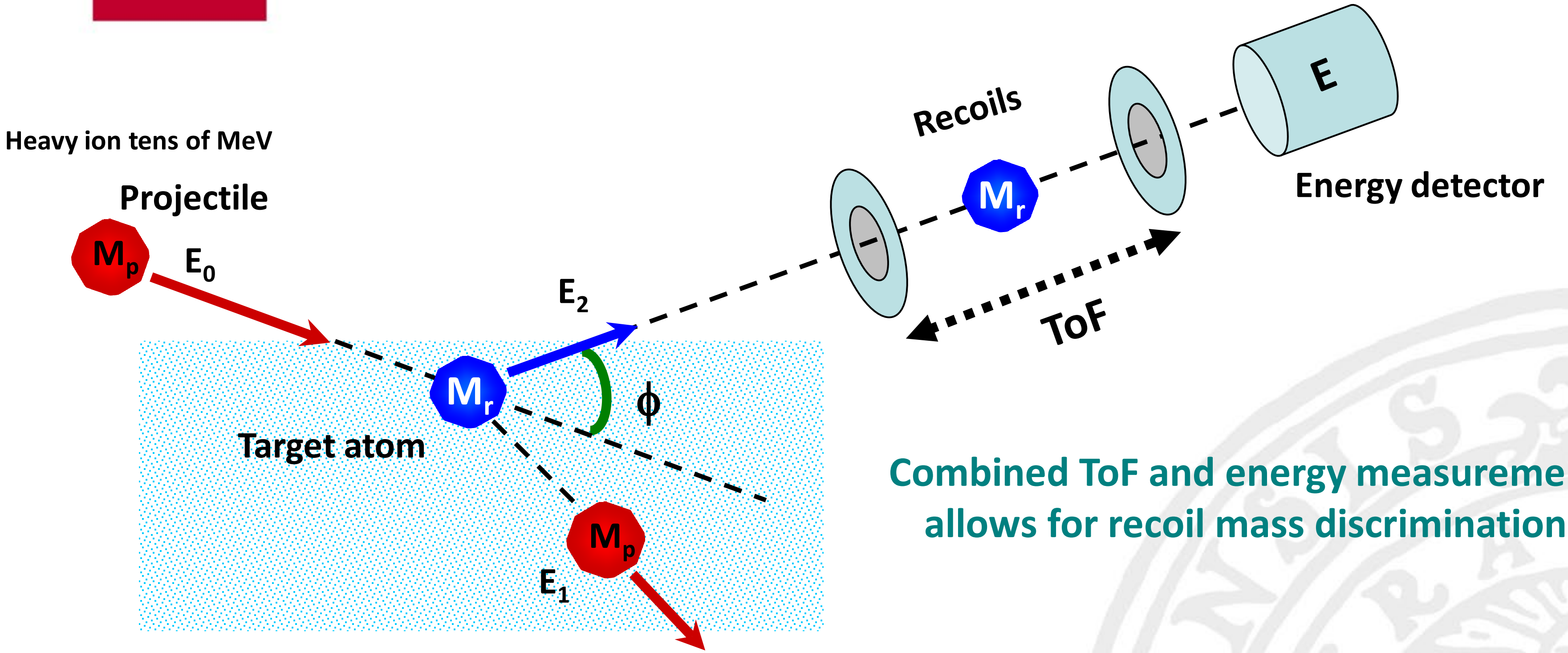


$La_{0.9}Sr_{0.1}MnO_3$

Element: line (energy [keV])

Elastic Recoil Detection Analysis (ERDA)

Principles of TOF-HIERDA



Combined ToF and energy measurement allows for recoil mass discrimination

- **Coincidence measurement of energy and time-of-flight for recoiled atoms**
- $E = \frac{m \cdot v^2}{2}$ **- mass identification and separation in a two-dimensional plot time vs. energy**

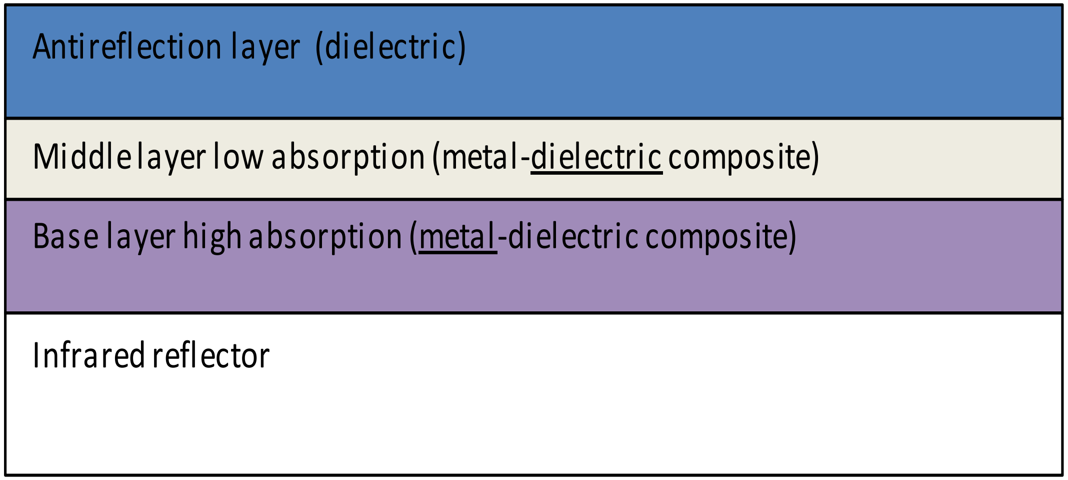
ToF-ERDA – from hydrogen to tungsten

Profiling solar absorbers

Solar absorbers for photo-thermal conversion

Aim: High absorptance: > 0.9 / low thermal emittance < 0.1 @ 350 deg. C

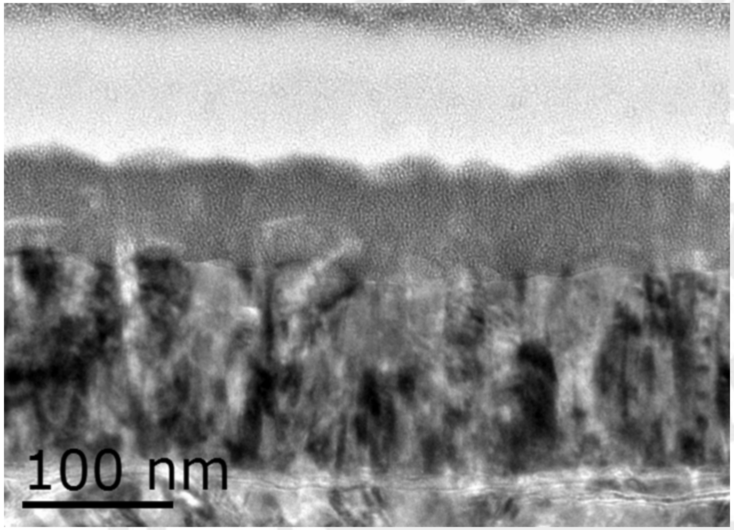
Questions: composition and thermal stability


Antireflection layer (dielectric)

Middle layer low absorption (metal-dielectric composite)

Base layer high absorption (metal-dielectric composite)

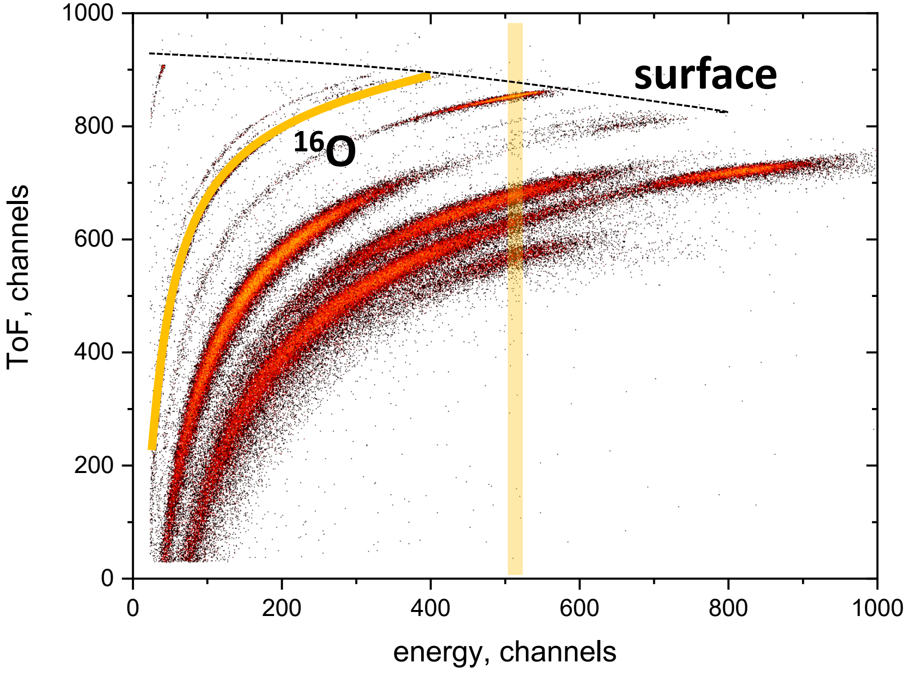
Infrared reflector

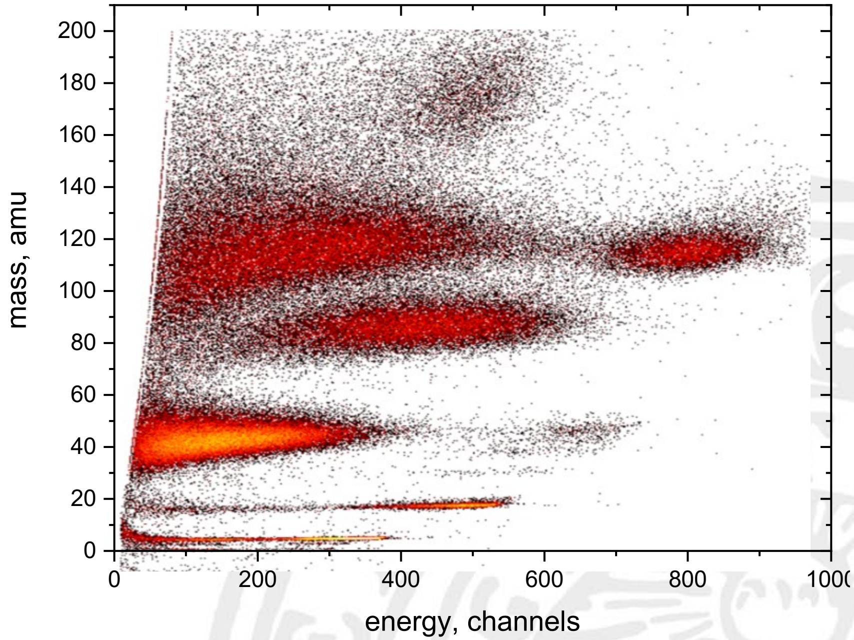


E. Wäckelgård, et al. Sol. Ener. Mat. & Sol. Cells (2015)

ToF-ERDA – from hydrogen to tungsten

Profiling solar absorbers

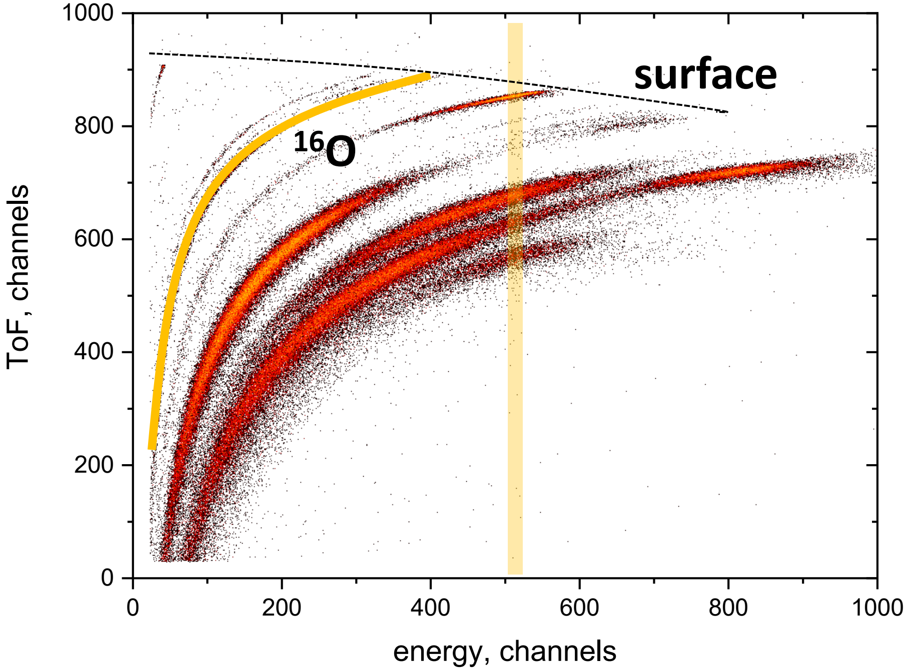


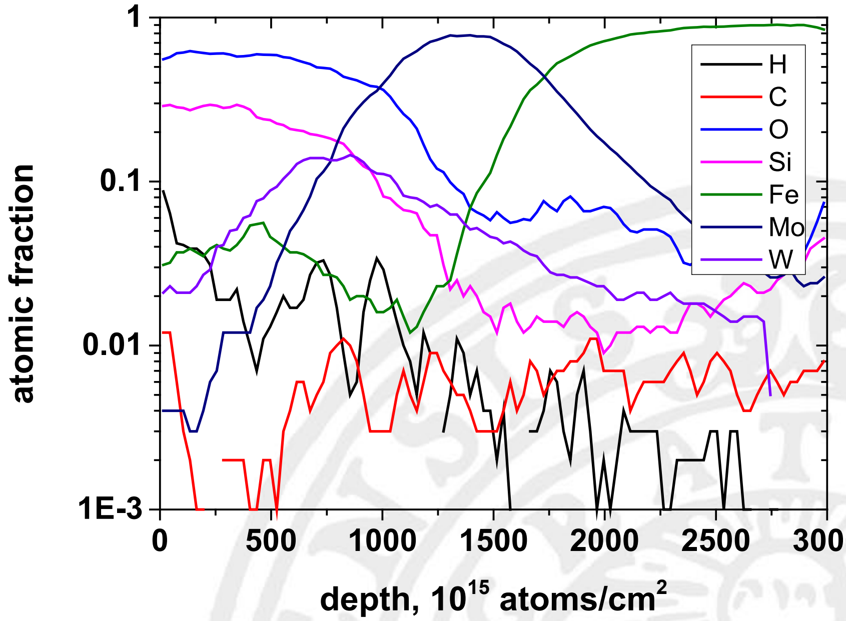


- For given energy, lighter particles are faster – unique discrimination and identification
- One track – one species
- Virtual surface from collision kinematics – depth profile along every single track

ToF-ERDA – from hydrogen to tungsten

Profiling solar absorbers





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ToF-ERDA – from hydrogen to tungsten

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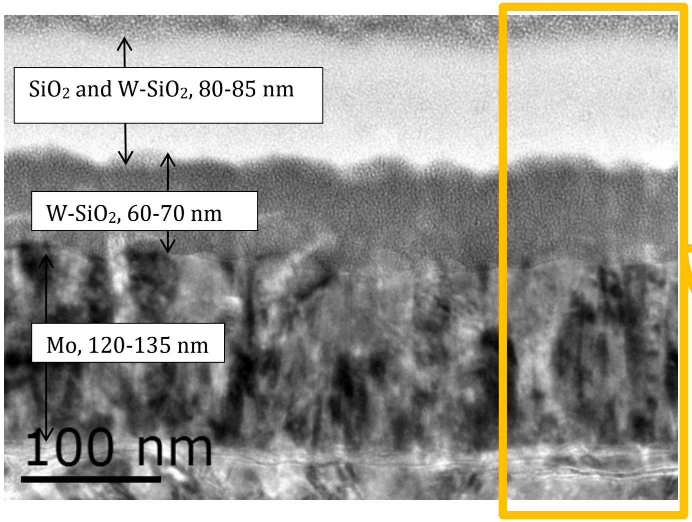
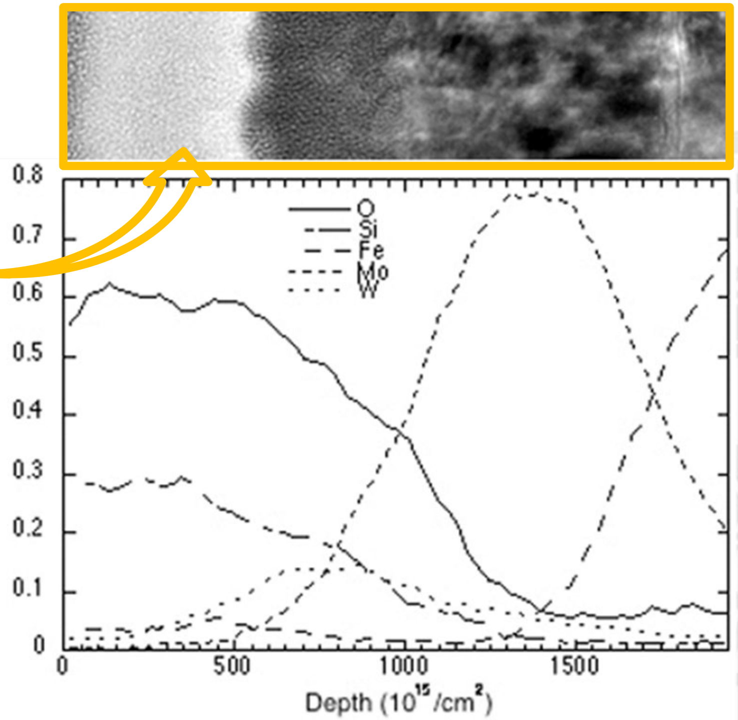




Simultaneous non-destructive detection of a series of chemical elements in a multi-layer system

Depth resolution ca. 20-40 nm

E. Wäckelgård, et al. Sol. Ener. Mat. & Sol. Cells (2015)

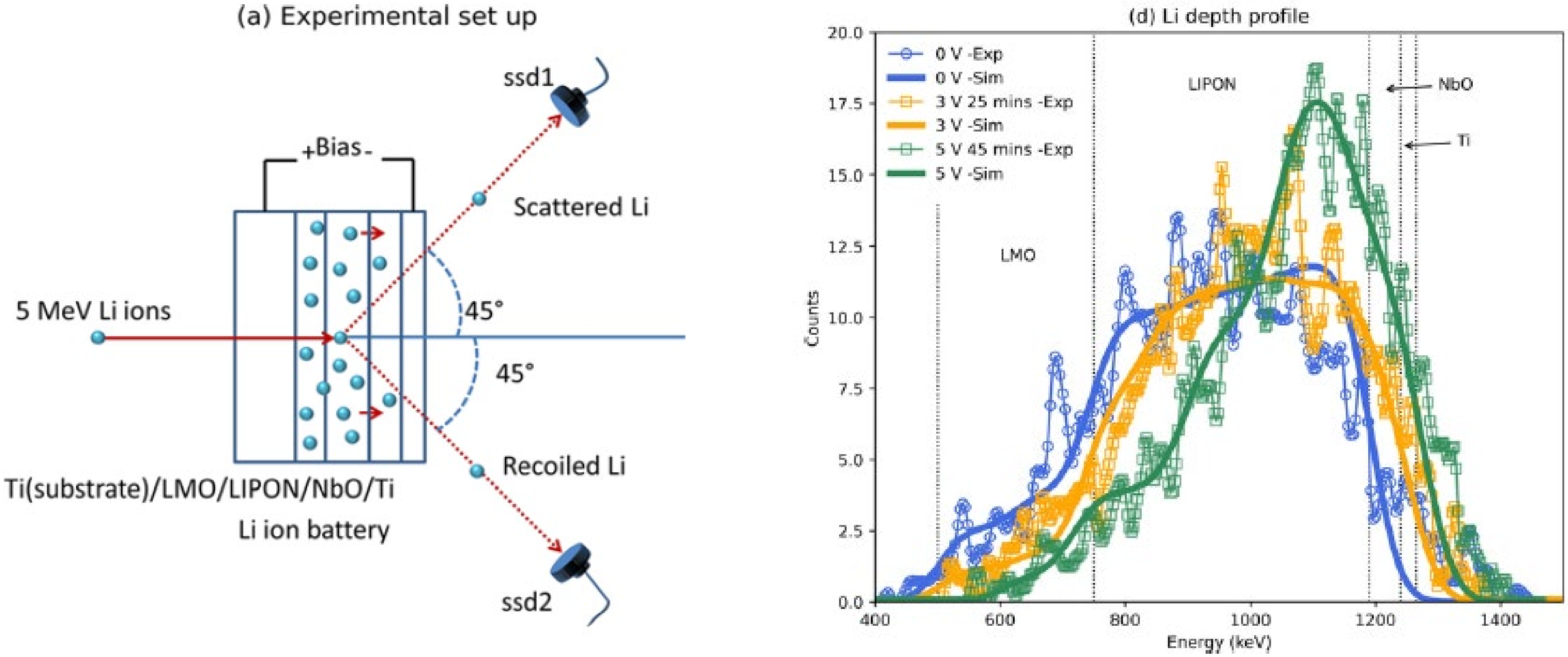
The Tandem Laboratory @ Uppsala University

5 MV pelletron accelerator

II. Developments

5 MV Pelletron, Uppsala University

In-operando studies of material transport

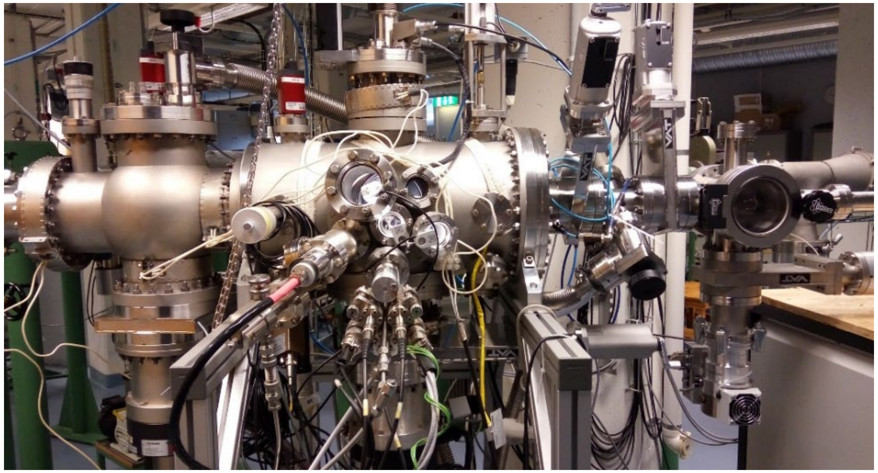


V. Mathayan et al., Appl. Phys. Lett. (2020)

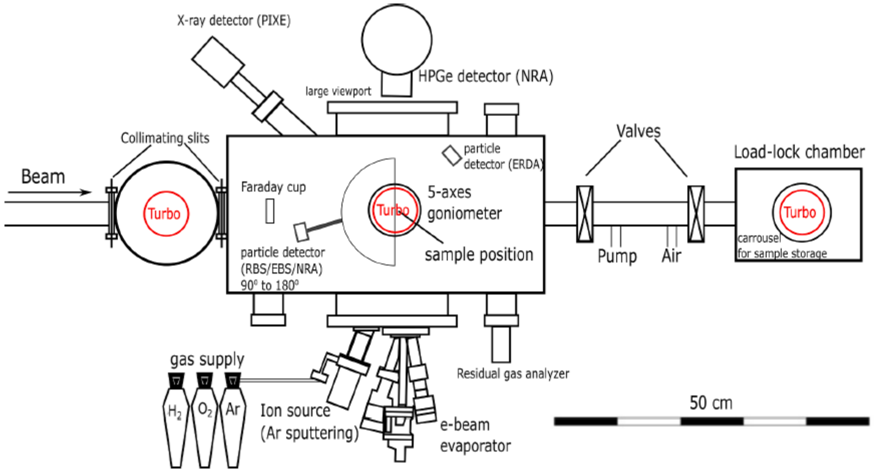
- **Depth-resolved profiling of Li in lithium thin film batteries**

5 MV Pelletron, Uppsala University

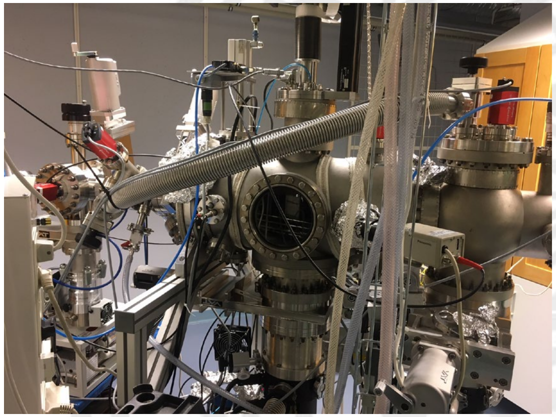
In-situ target modification & IBA characterization



- UHV-chamber at T6 @ 5MV tandem
- Accessible for light and heavy ions
- Beam energies from 2 – 50 MeV
- Large viewport (for e.g. optical characterization or similar)

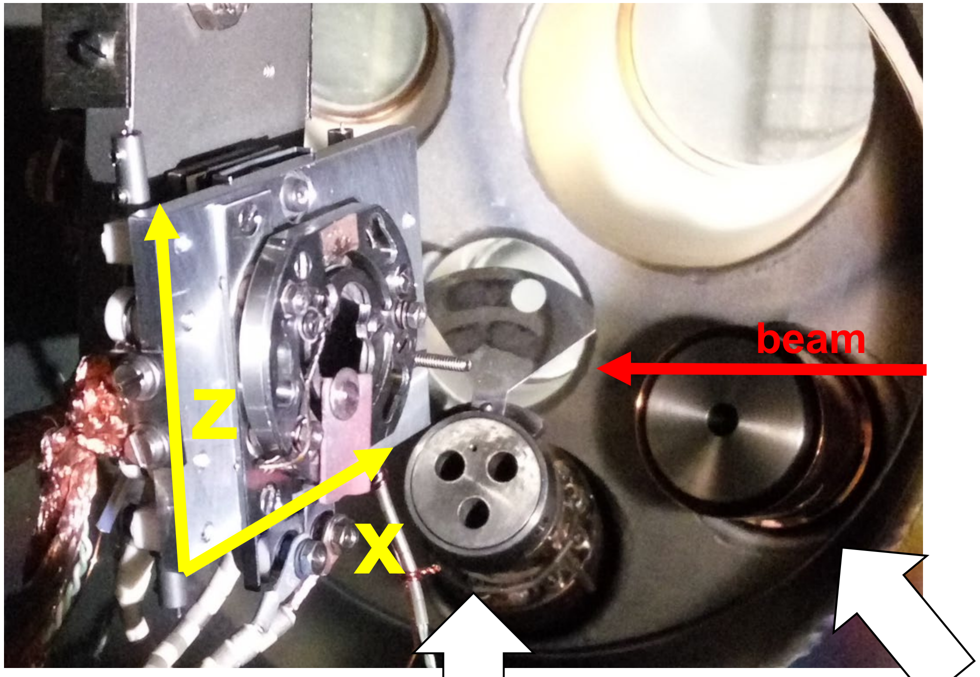


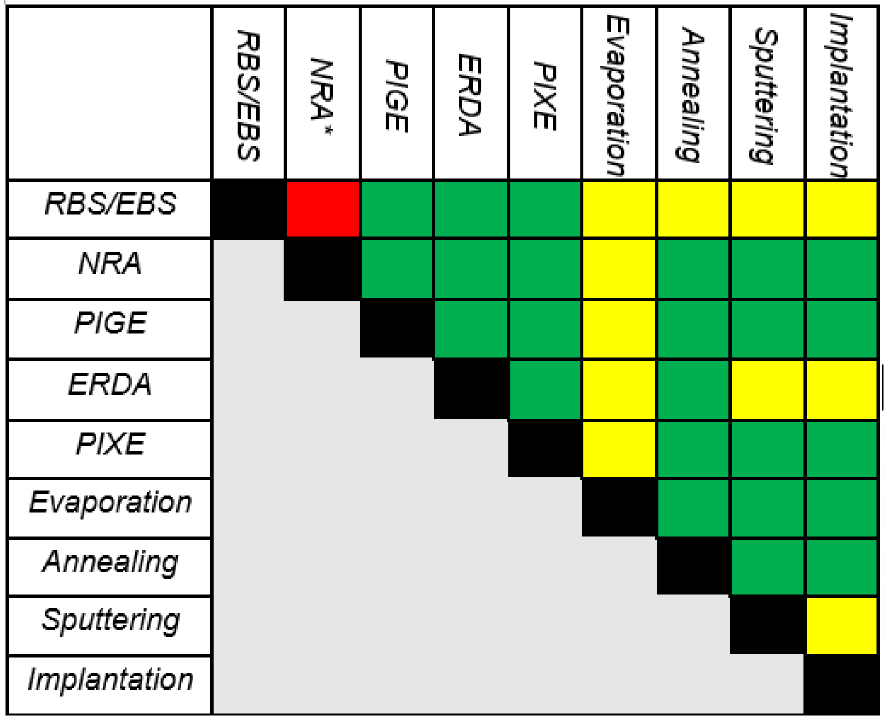
K. Kantre et al., Nucl. Instr. Meth. B (2020)



5 MV Pelletron, Uppsala University

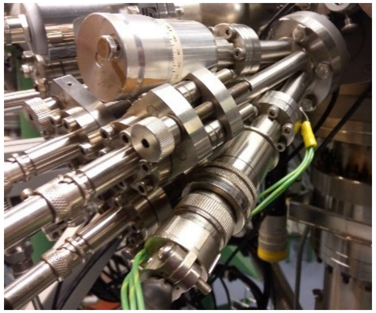
Multi-method capabilities



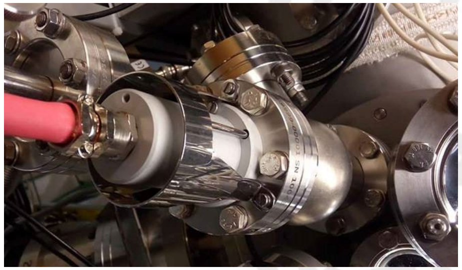


	RBS/EBS	NRA*	PIGE	ERDA	PIXE	Evaporation	Annealing	Sputtering	Implantation
RBS/EBS	⬛	🟥	🟩	🟩	🟩	🟨	🟨	🟨	🟨
NRA		⬛	🟩	🟩	🟩	🟨	🟩	🟩	🟩
PIGE			⬛	🟩	🟩	🟨	🟩	🟩	🟩
ERDA				⬛	🟩	🟨	🟩	🟨	🟨
PIXE					⬛	🟨	🟩	🟩	🟩
Evaporation						⬛	🟩	🟩	🟩
Annealing							⬛	🟩	🟩
Sputtering								⬛	🟨
Implantation									⬛

K. Kantre et al., Nucl. Instr. Meth. B (2020)



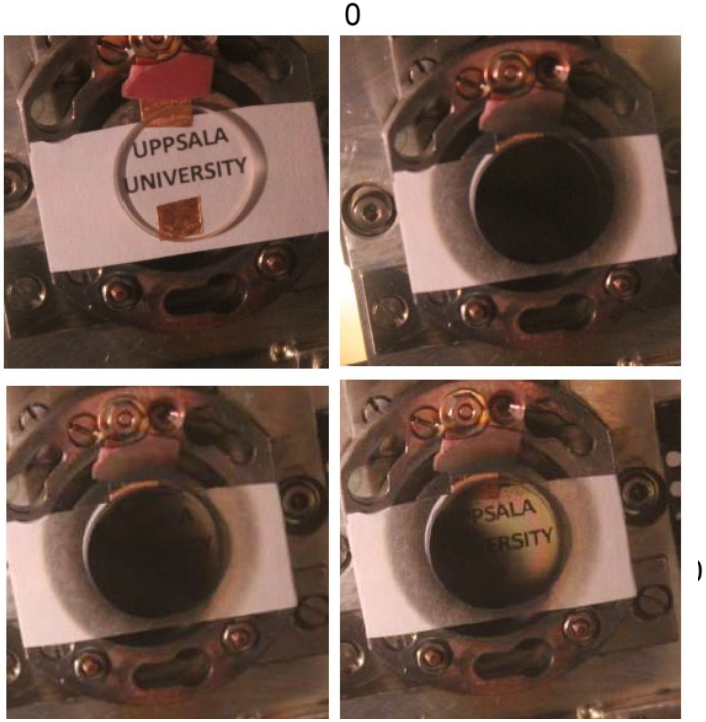
3 evaporation cells:
Co-deposition possible



1-5 keV ion
sputter gun

In-situ IBA of target modification

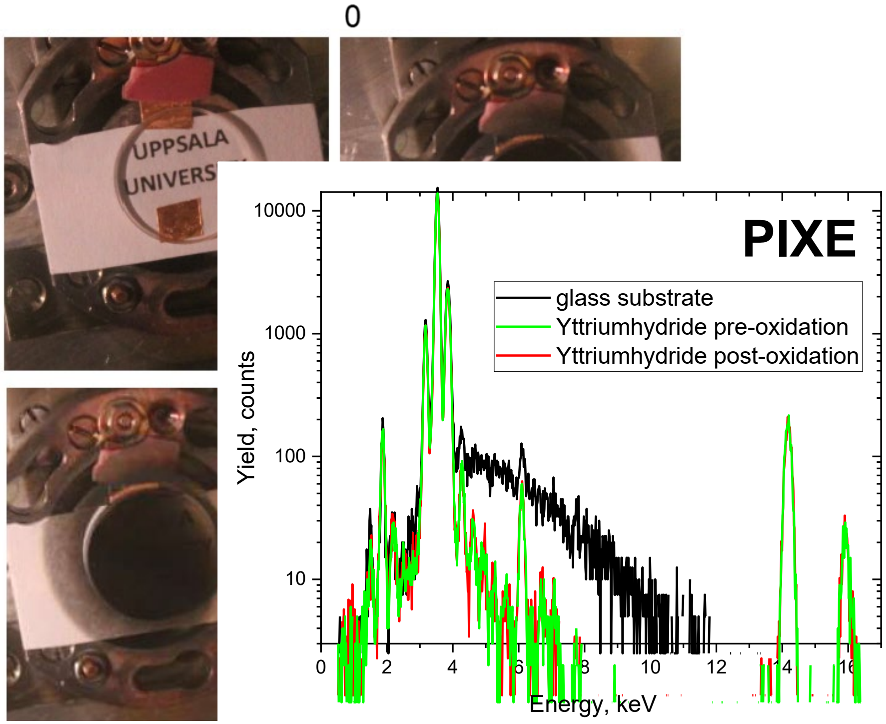
Research on photochromic materials



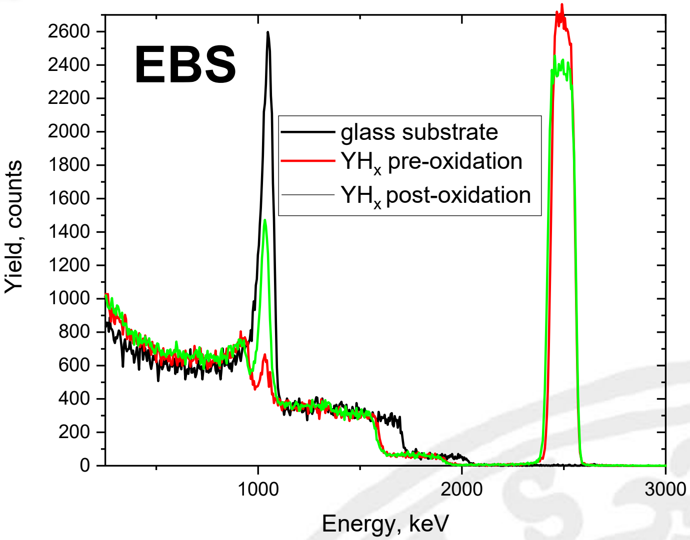
- Y growth in hydrogen atmosphere
- O-exposure under controlled pressures
- RBS/EBS, PIXE & ERD with He-beam
- Post-mortem TOF-HIERDA analysis

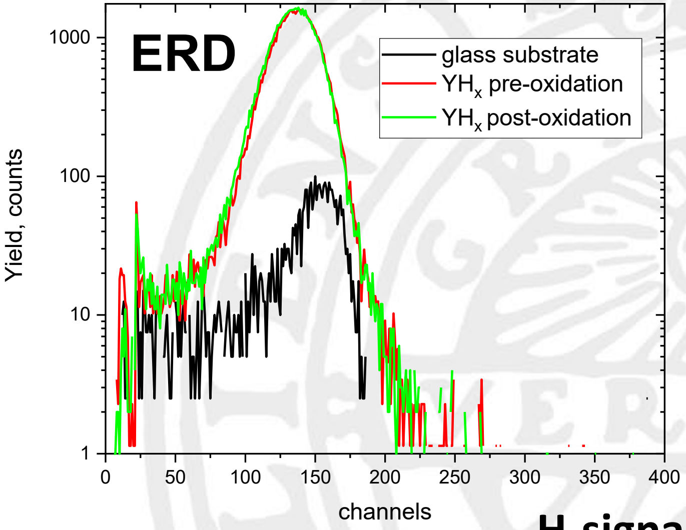
In-situ IBA of target modification

Research on photochromic materials



O-signal



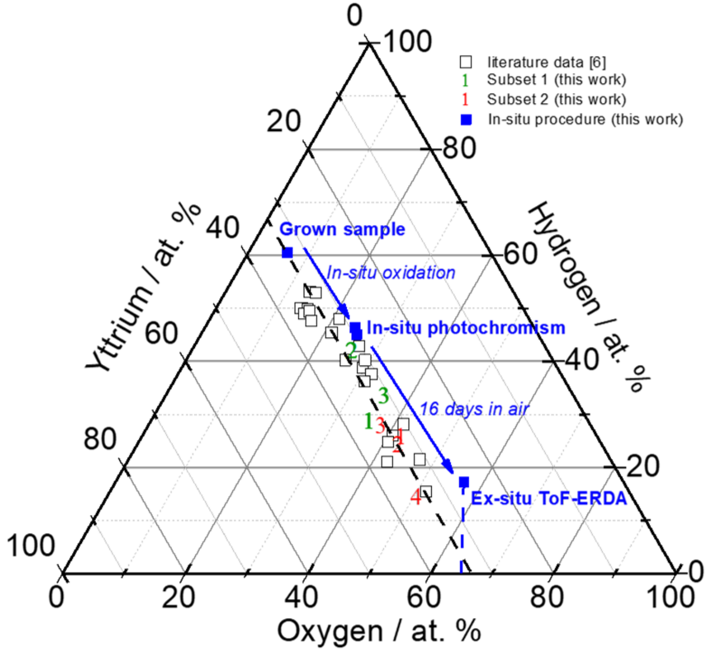


H-signal

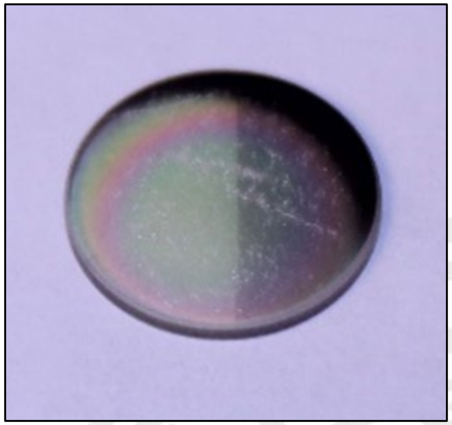
- Y growth in hydrogen atmosphere
- O-exposure under controlled pressures
- RBS/EBS, PIXE & ERD with He-beam
- Post-mortem TOF-HIERDA analysis

In-situ IBA of target modification

Research on photochromic materials



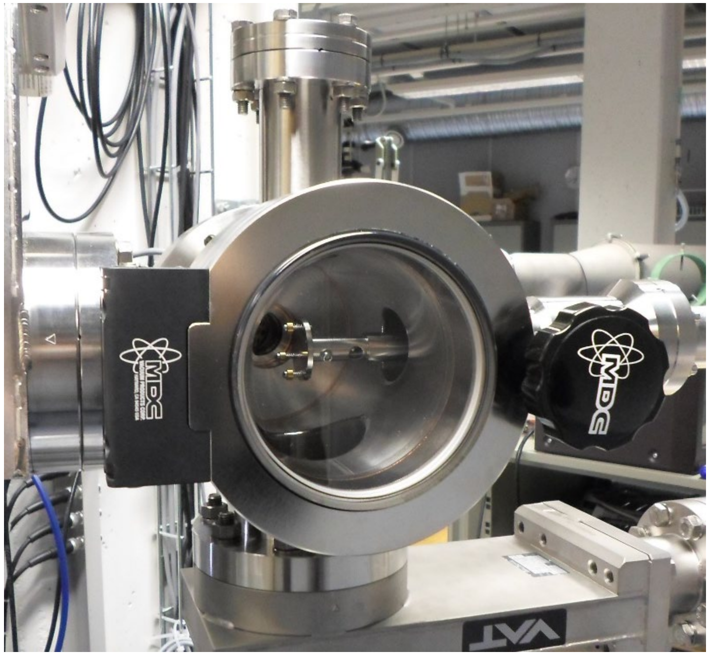
modified from Moldarev et al. Phys. Rev. Mat. (2018)
Kantre et al., Scripta Materialia (2020)



- **Correlating composition & photochromic performance**
- **Tayloring properties in a controlled approach**

In-situ IBA of target modification

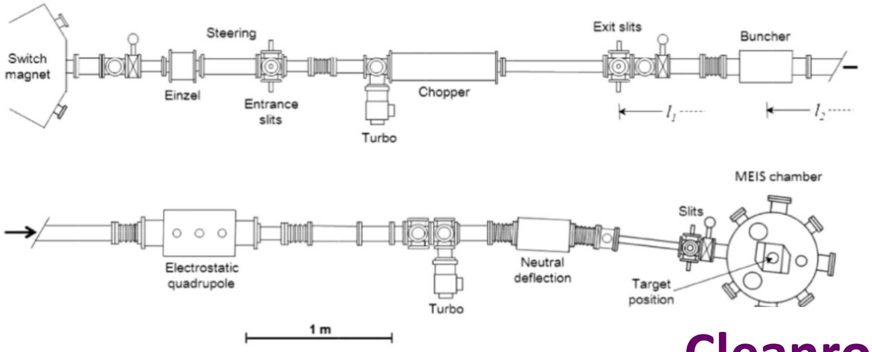
Sample transfer with mobile high-vacuum chamber



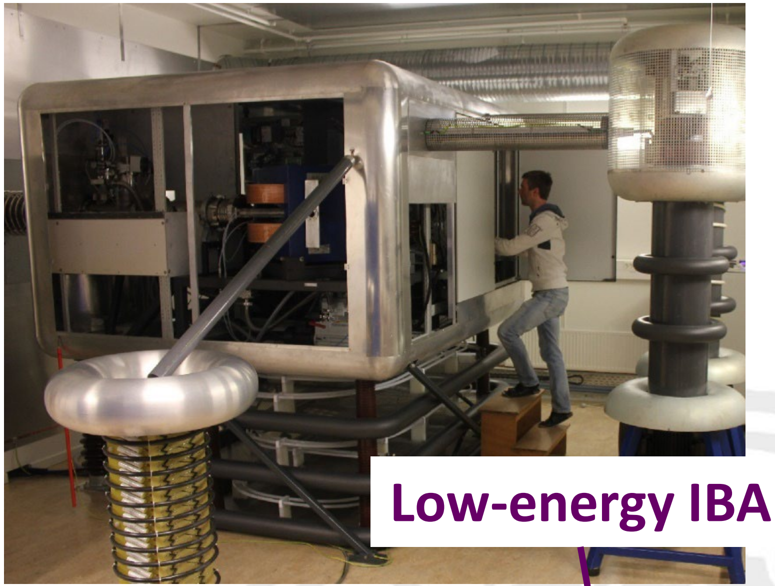
- Sample transfer under high vacuum (actively pumped)
- Carroussel holding 4 samples
- Transfer from Tandemaccelerator to MEIS system

The Uppsala ToF-MEIS system

Basic information



M. Linnarsson et al., Rev. Sci. Instr. (2



Cleanroom Implantations

Low-energy IBA

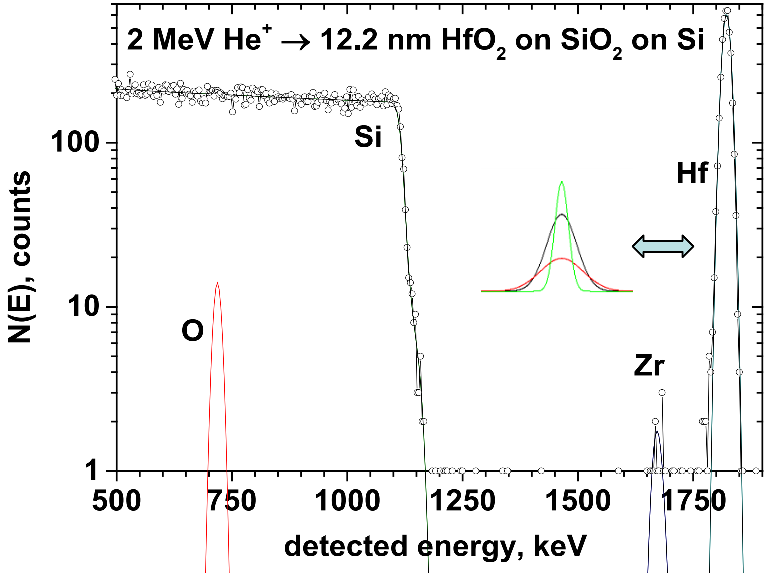
MEIS

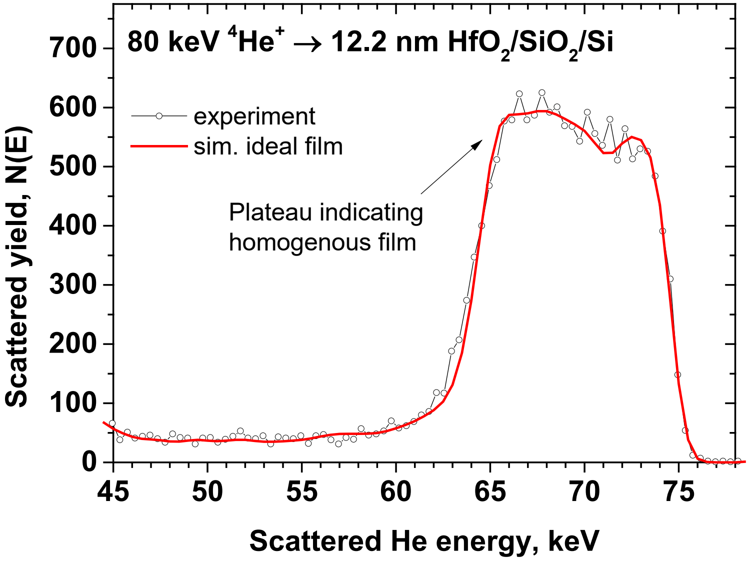
DanFysik implanter as ion source:

- **350 kV voltage at the platform**
- **beam energies from 20 to 300 keV for MEIS**
- **gas/sputter/oven: beams of all elements**
- **In MEIS: H, D, He, B, Ne, Ar**

The Uppsala ToF-MEIS system

Comparison to Standard-RBS





RBS: $\Delta E < \delta E$

MEIS: $\Delta E > \delta E$

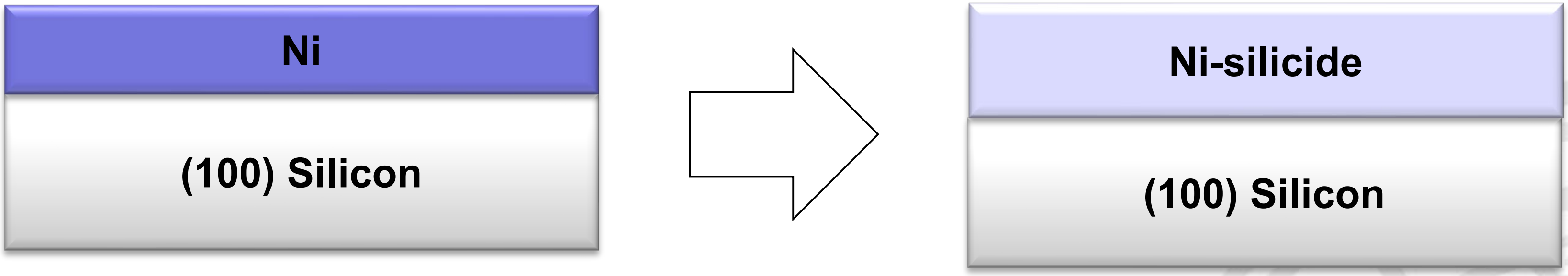
The Uppsala ToF-MEIS system

What is it good for?

Applications

In-situ annealing in TOF-MEIS

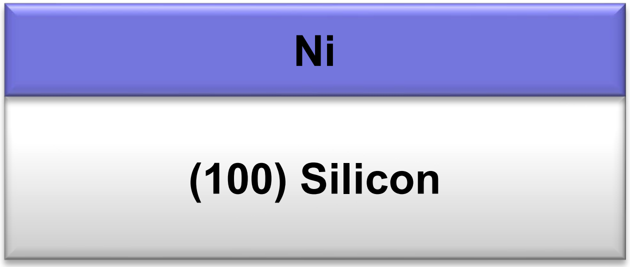
CMOS-processing – poly & single crystalline materials



- **Ni-silicide formation due to annealing of amorphous Ni on Si: standard CMOS step**

In-situ annealing in TOF-MEIS

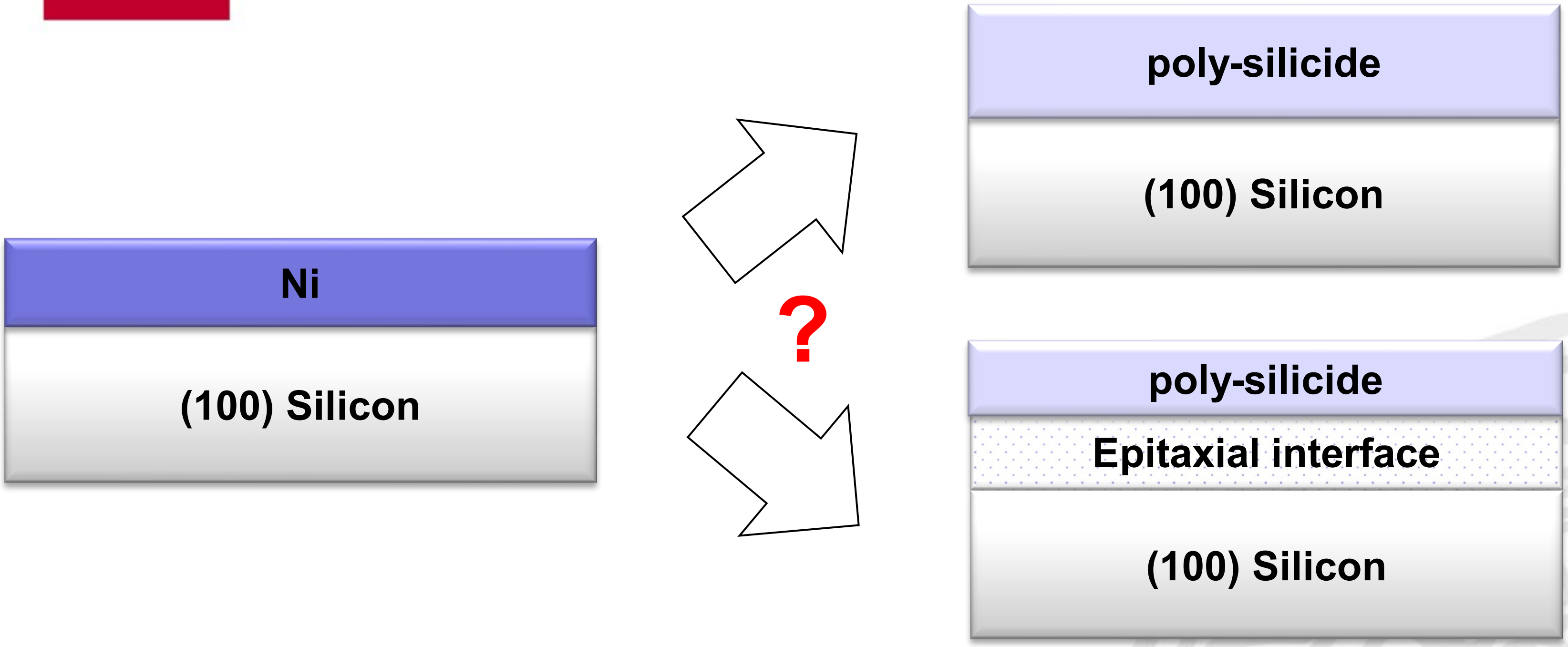
CMOS-processing – poly & single crystalline materials



- Ni-silicide formation due to annealing of amorphous Ni on Si: standard CMOS step
- Step 1: Ni-deposition on Si-substrate

In-situ annealing in TOF-MEIS

CMOS-processing – poly & single crystalline materials



Ni

(100) Silicon

?

poly-silicide

(100) Silicon

poly-silicide

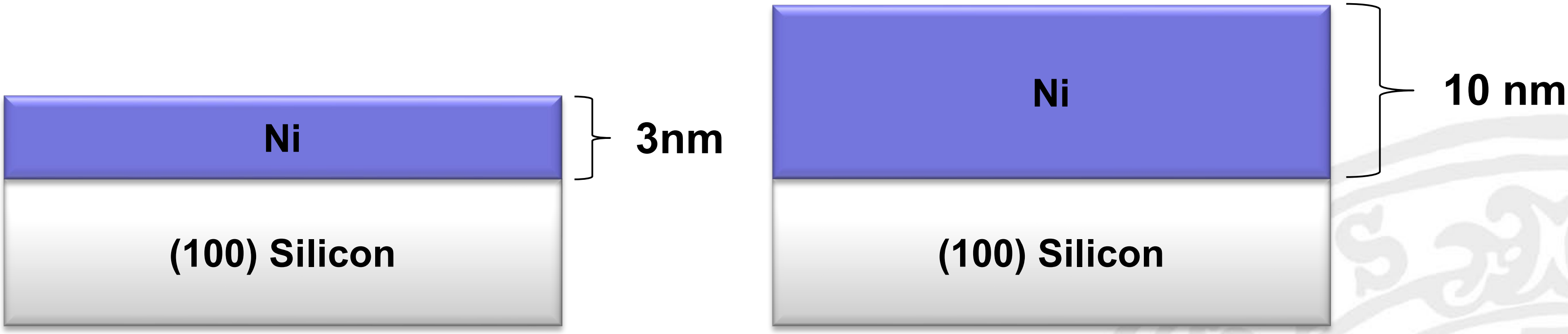
Epitaxial interface

(100) Silicon

- Ni-silicide formation due to annealing of amorphous Ni on Si: standard CMOS step
- Step 1: Ni-deposition on Si-substrate
- Step 2: Thermal anneal forming Ni_2Si, NiSi, or $NiSi_2$
- Synthesis pathways may depend on thickness of Ni-layer and temperature

In-situ annealing in TOF-MEIS

CMOS-processing – poly & single crystalline materials



Ni

3nm

(100) Silicon

Ni

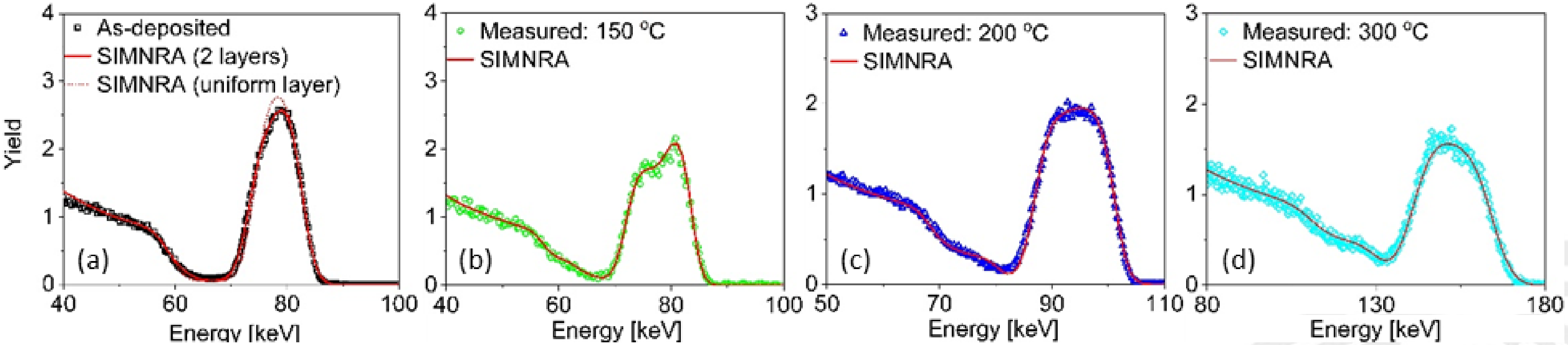
10 nm

(100) Silicon

- 2 different sample systems investigated
- Using ToF-MEIS for *in-situ* depth profiling during silicidation process
- 2D-detector enables to study depth resolved real-space crystallographic order

In-situ annealing in TOF-MEIS

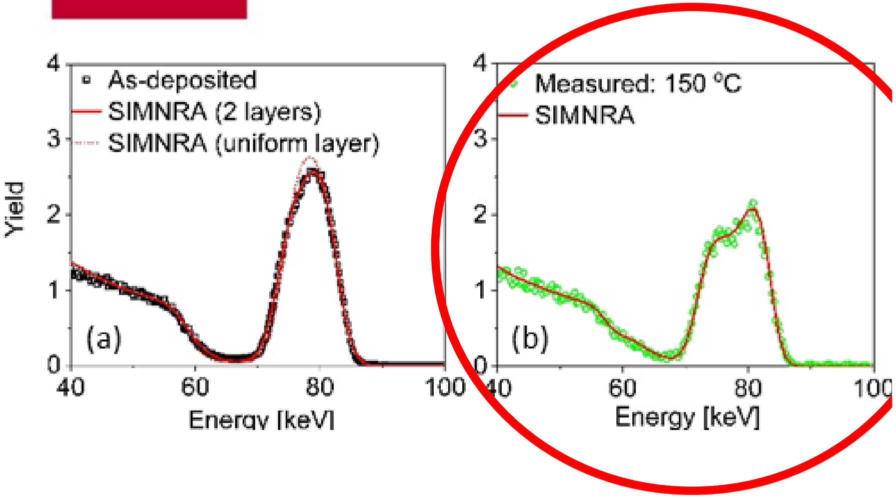
in-situ silicidation: results

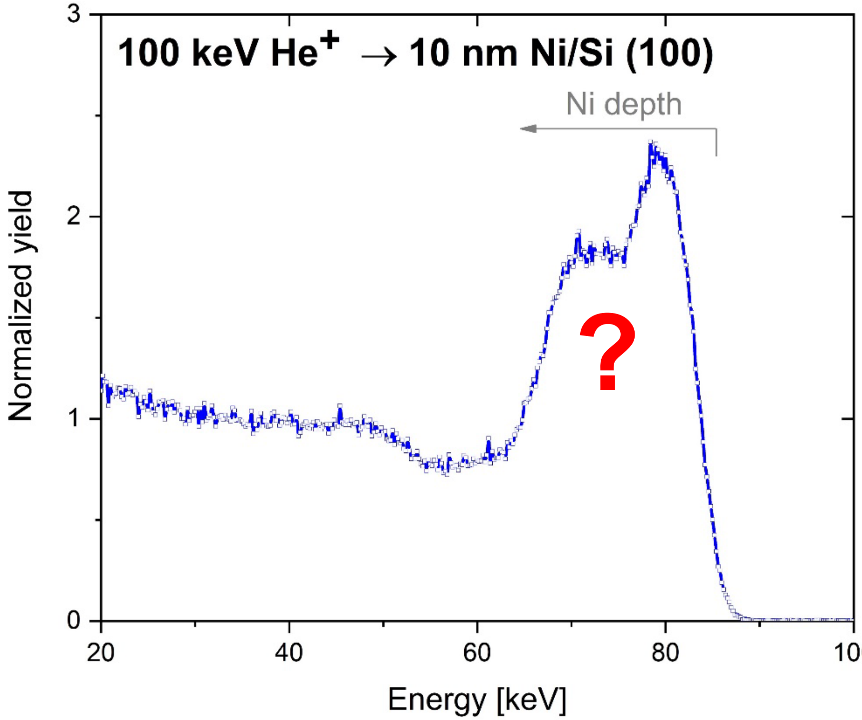


- **ToF-MEIS profiles of silicidation process**
- **Clear transformation visible**
- **Process starting from interface to Si**

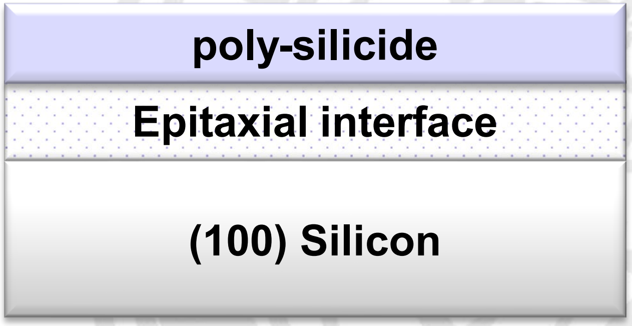
In-situ annealing in TOF-MEIS

in-situ silicidation: results





- **ToF-MEIS profiles of silicidation process**
- **Clear transformation visible**
- **Process starting from interface to Si**
- **Epitaxial transformation at interface?**



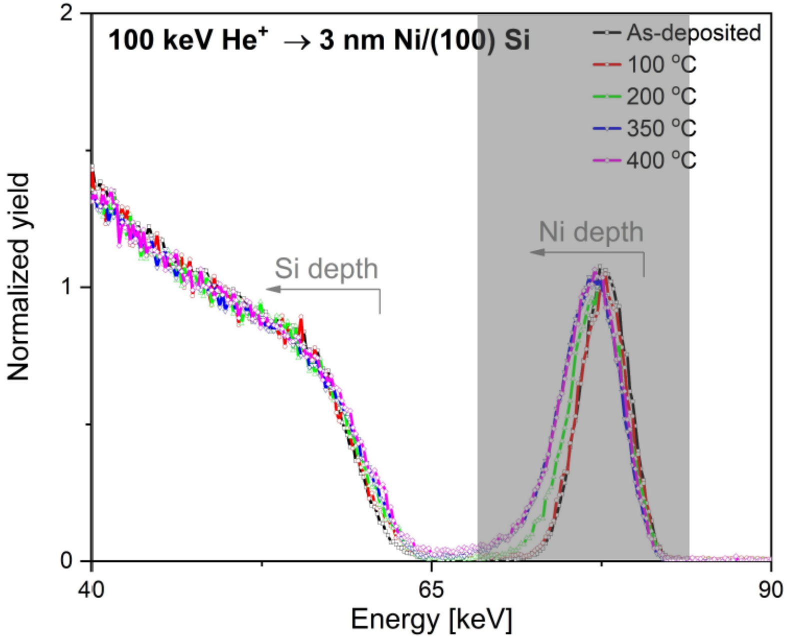
poly-silicide

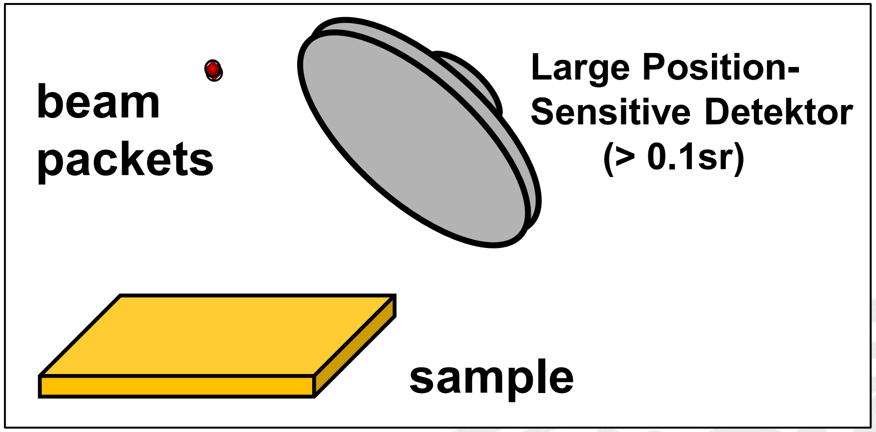
Epitaxial interface

(100) Silicon

In-situ annealing in TOF-MEIS

Single crystalline materials

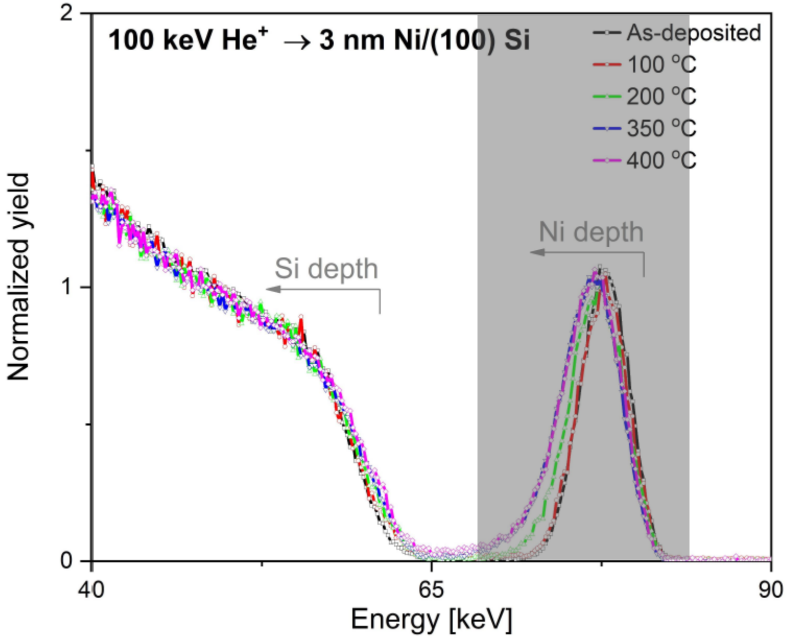


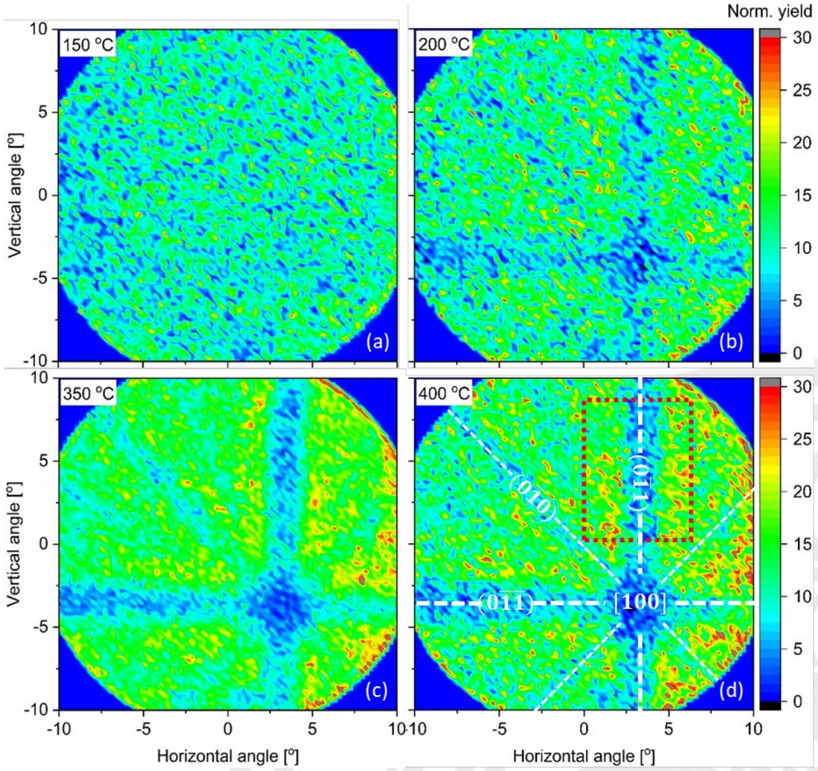


- **Width of signal associated with Ni changes – indication of transformation**

In-situ annealing in TOF-MEIS

Single crystalline materials



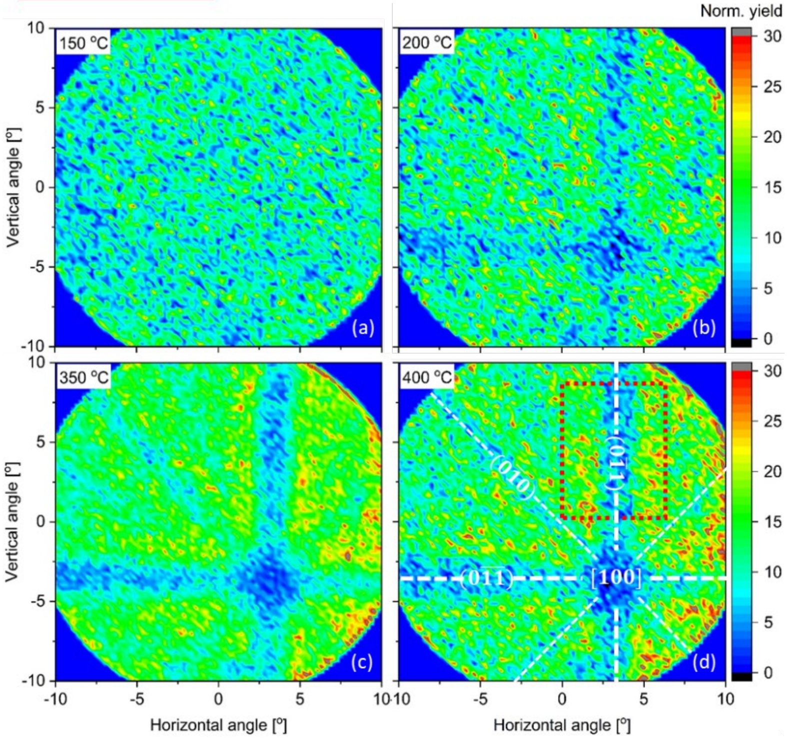


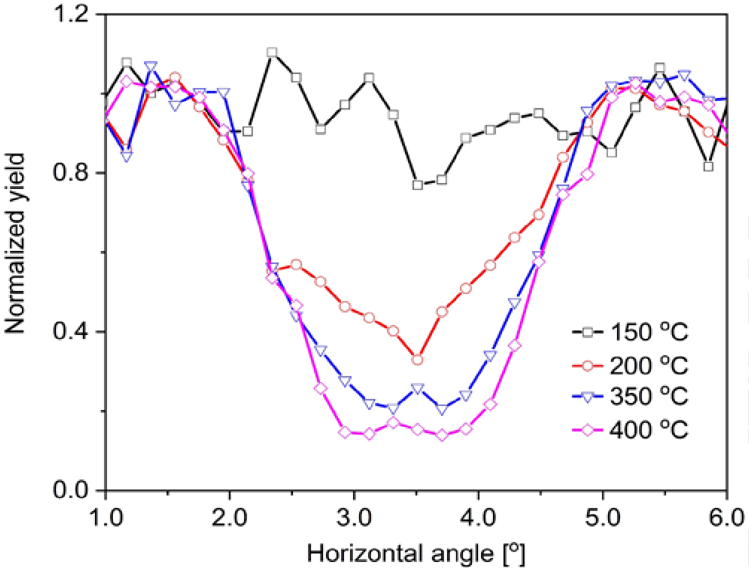
Tuan Tran et al., Scientific Reports (2020)

- **Width of signal associated with Ni changes – indication of transformation**
- **Annealing of ultrathin amorphous layers leads to formation of epitaxial layer**

In-situ annealing in TOF-MEIS

Single crystalline materials

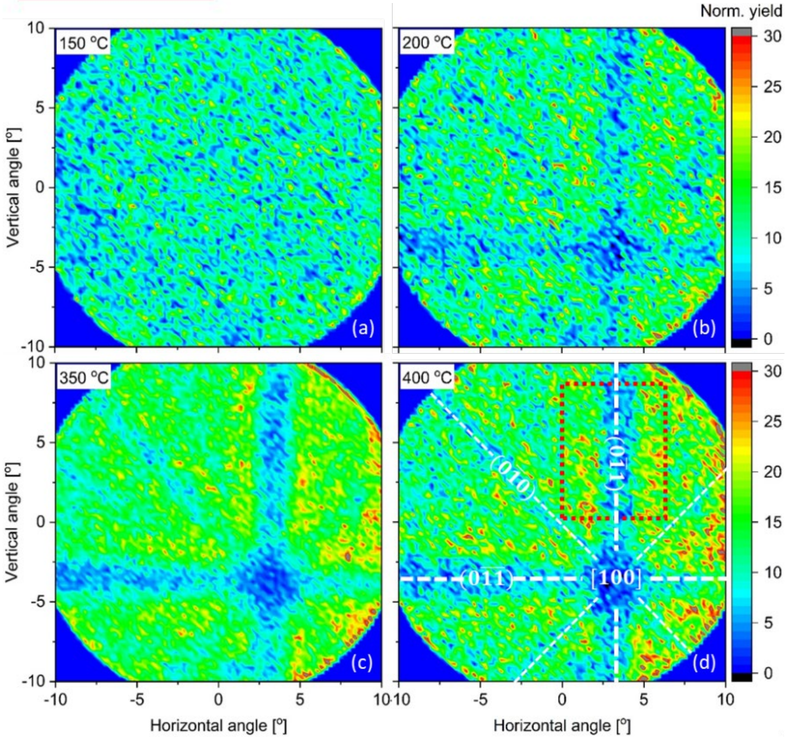


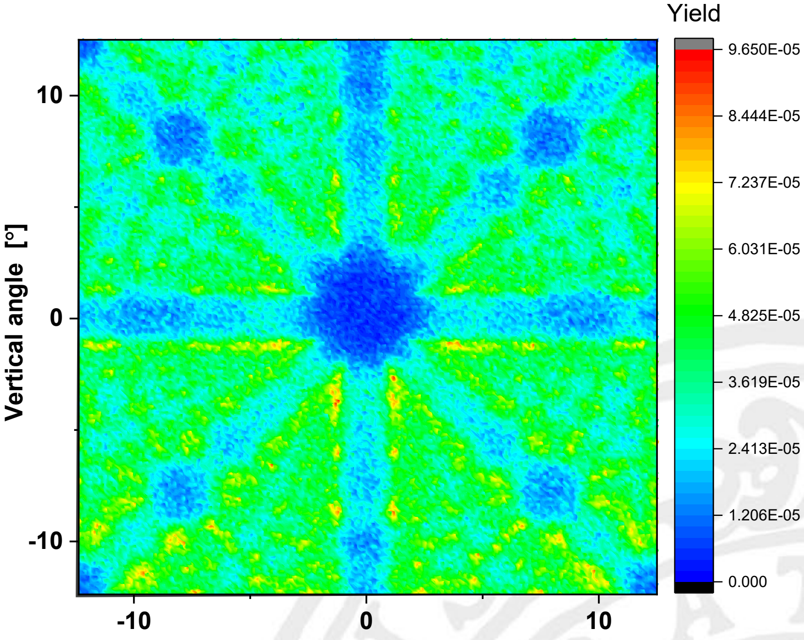


- **Annealing of ultrathin amorphous layers leads to formation of epitaxial layer**
- **Depth-resolved analysis of strain & layer relaxation becomes possible *in-situ***

In-situ annealing in TOF-MEIS

Single crystalline materials



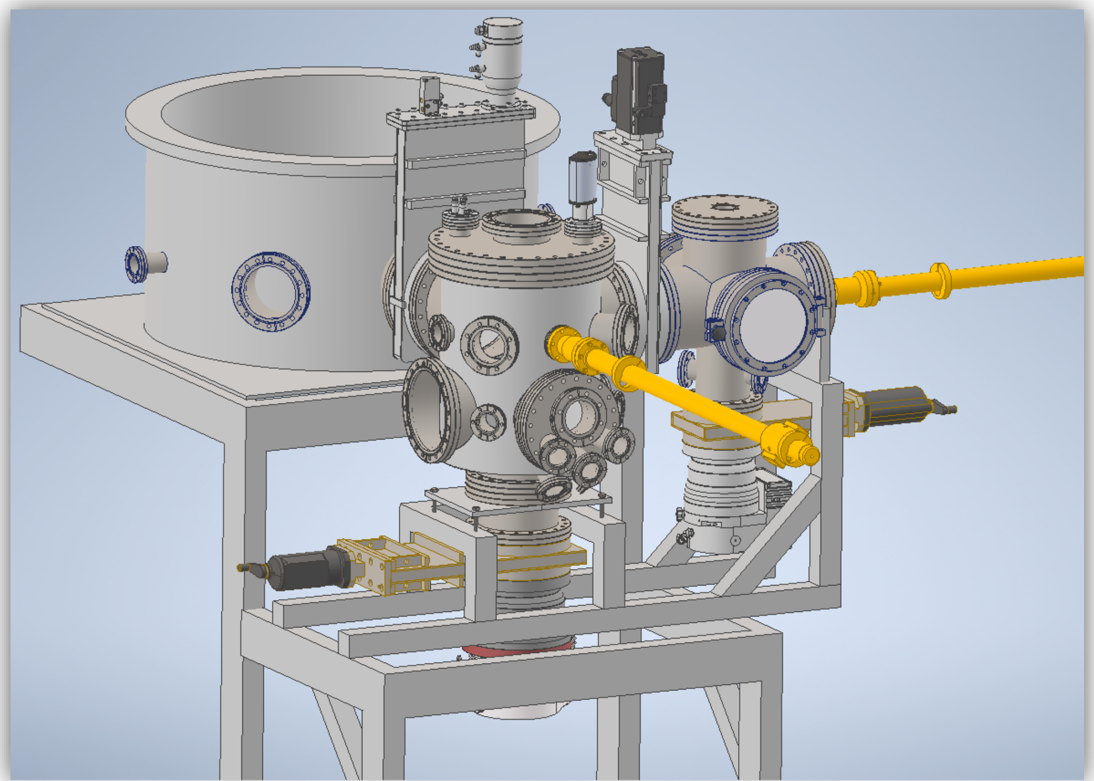


Simulation using TRIC, *Khodyrev et al. PRE*

- **Annealing of ultrathin amorphous layers leads to formation of epitaxial layer**
- **Depth-resolved analysis of strain & layer relaxation becomes possible *in-situ***
- **Simulation of patterns possible using Monte-Carlo tools**

The Uppsala ToF-MEIS system

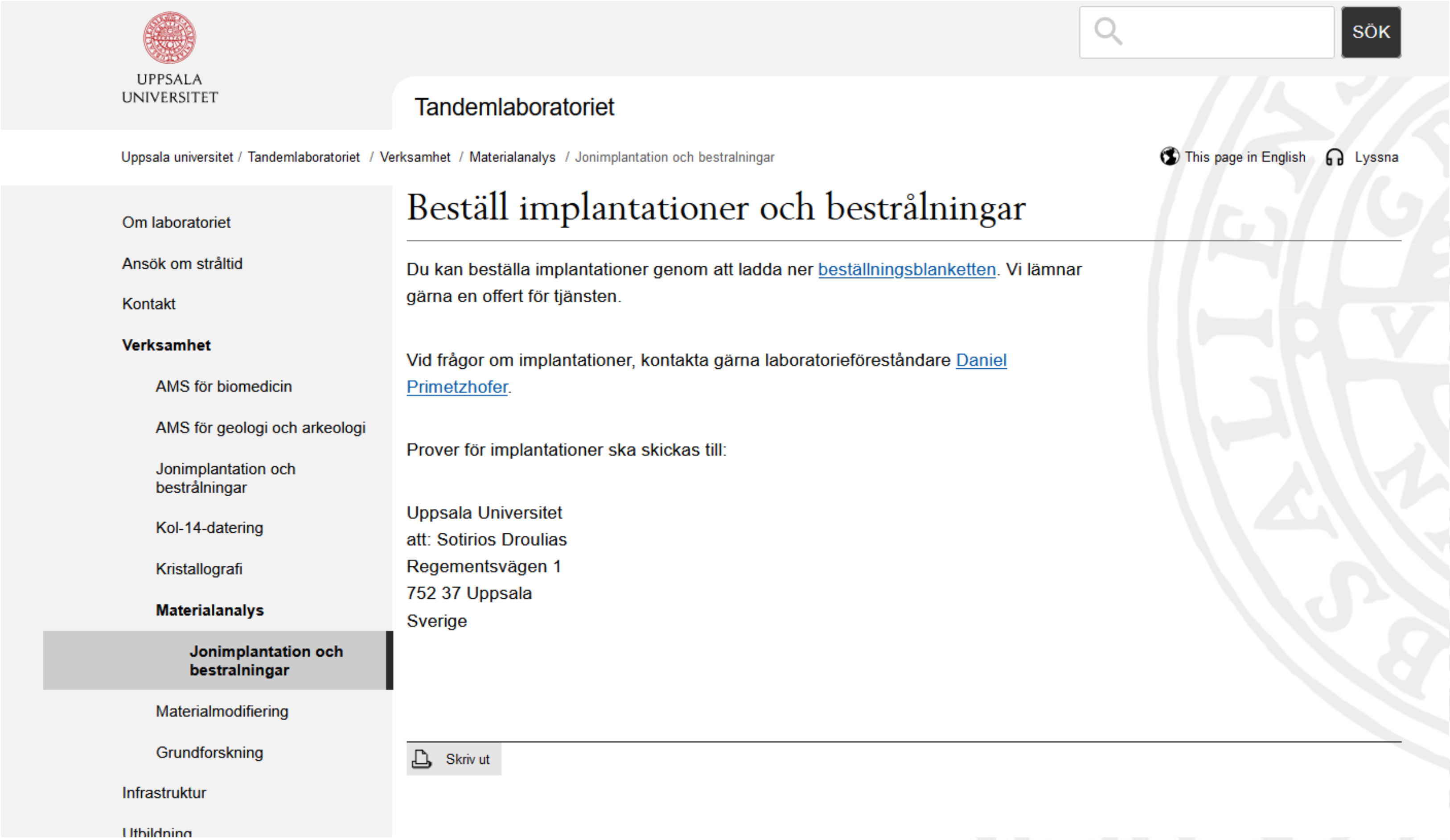
Present upgrades: target manufacturing & modification



- *in-situ* growth (evaporation & magnetron)
- gas exposure
- sputtering
- annealing
- …

Access

How to get beam time?



Beställ implantationer och bestrålningar

Du kan beställa implantationer genom att ladda ner beställningsblanketten. Vi lämnar gärna en offert för tjänsten.

Vid frågor om implantationer, kontakta gärna laboratorieföreståndare Daniel Primetzhofer.

Prover för implantationer ska skickas till:

Uppsala Universitet
att: Sotirios Droulias
Regementsvägen 1
752 37 Uppsala
Sverige

Access

How to get beam time?

Standard ion beam analysis:	5 week schedule upon beam time application
Irradiations:	5 week schedule upon beam time application
Implantations:	3 week schedule upon beam time application

Regular users receive ”calls for application”

Contact: go to www.tandemlab.uu.se or

mail: tandemlaboratoriet@physics.uu.se

Access

How to get beam time?

Standard runs:

Work for hire by typ. industrial customers

You request a measurement

We send an offer for beamtime and (if requested) data analysis

Upon agreement the measurement is scheduled

Non-standard runs:

(Industrial customers & others)

You request a measurement

Joint grant application

Test study

Joint publication