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Preparation and characterization of ^{239}Pu and ^{240}Pu recoil ion sources, providing ^{235m}U and ^{236}U for laser-spectroscopic study

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LISA LASER IONISATION AND SPECTROSCOPY OF ACTINIDES · JG|U · UNIVERSITY OF JYVÄSKYLÄ · GSI · HIM HELMHOLTZ Helmholtz-Institut Mainz

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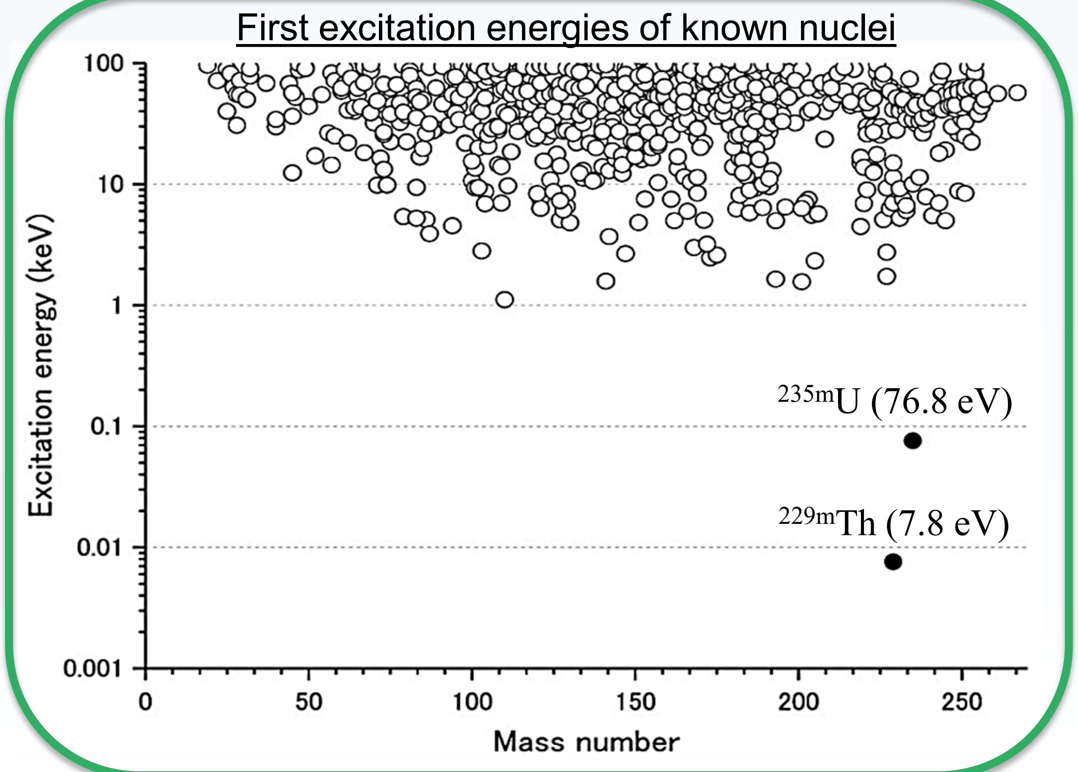
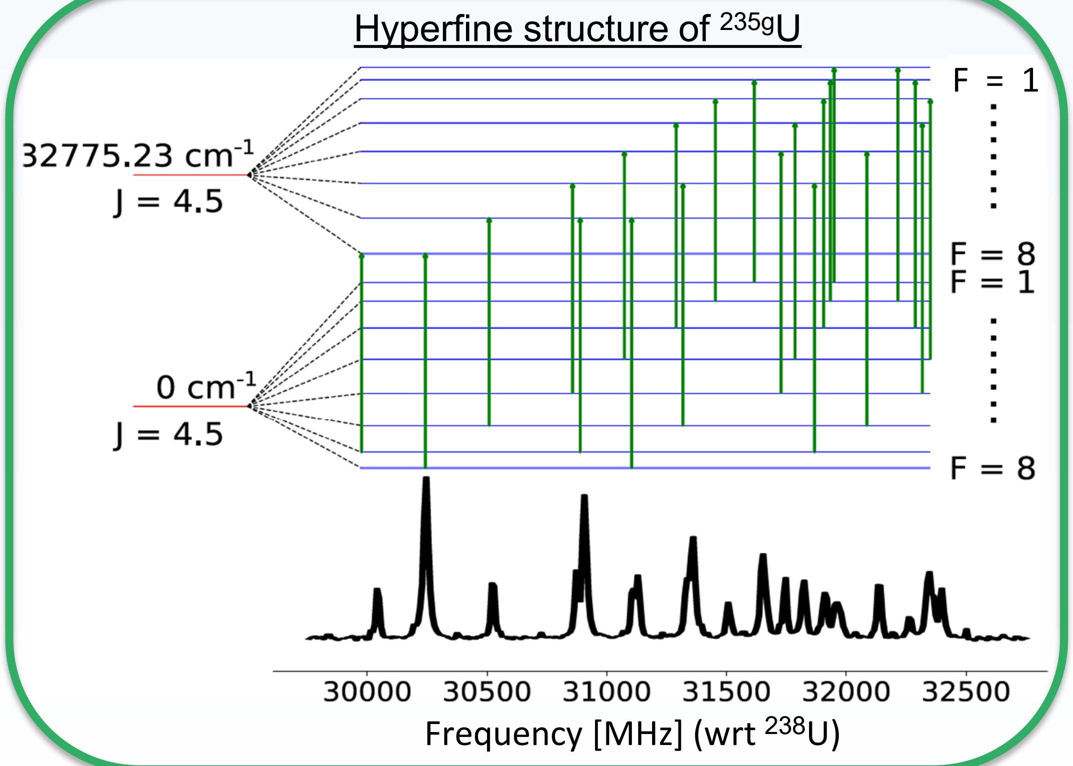
Agenda

- I. Motivations
- II. Molecular plating
- III. Preparation and Characterization of ^{239}Pu and ^{240}Pu recoil ion sources
- V. U recoil yields – working towards laser spectroscopy of ^{235m}U
- VI. DICE - Detection of Internal Conversion Electrons setup

Motivations

- **Ultra-low-lying isomers** → below 100 eV excitation energy



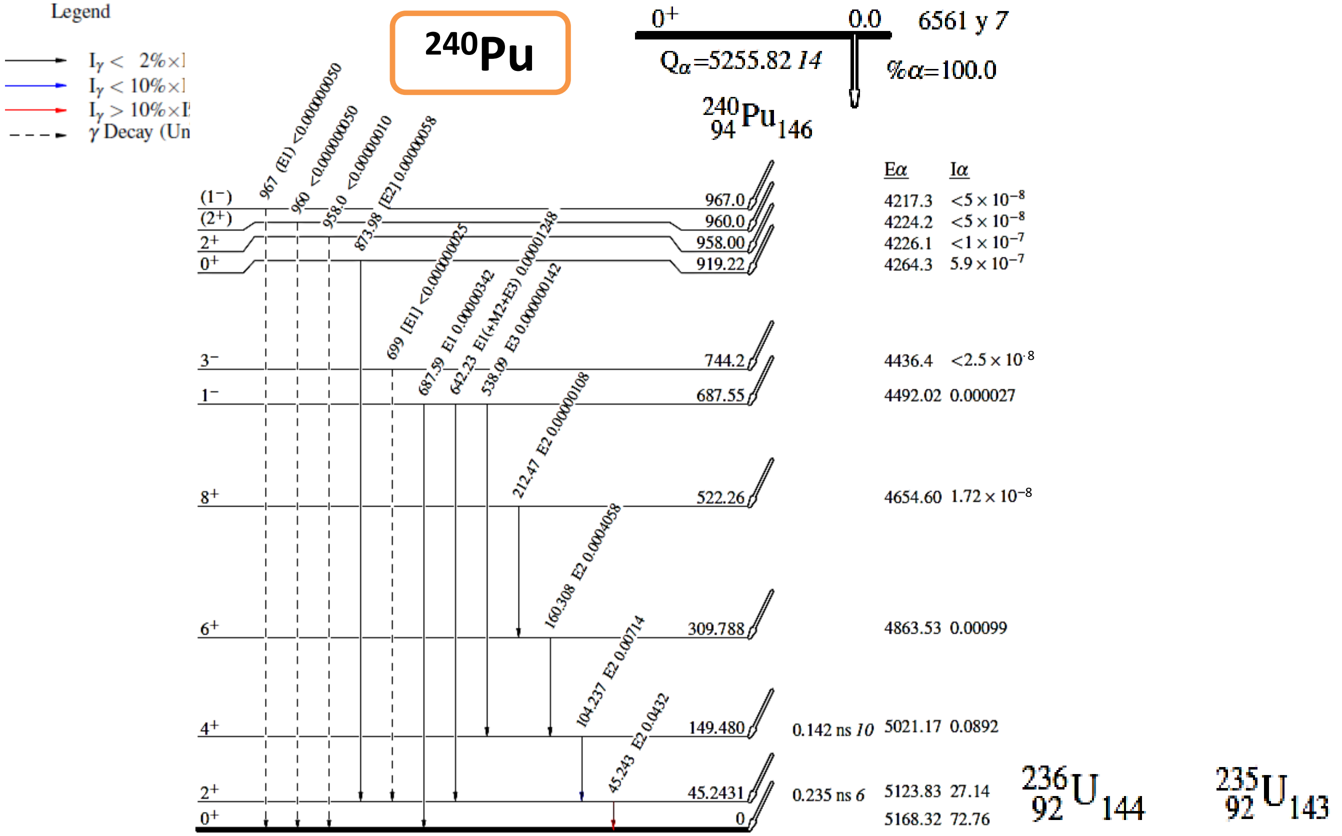


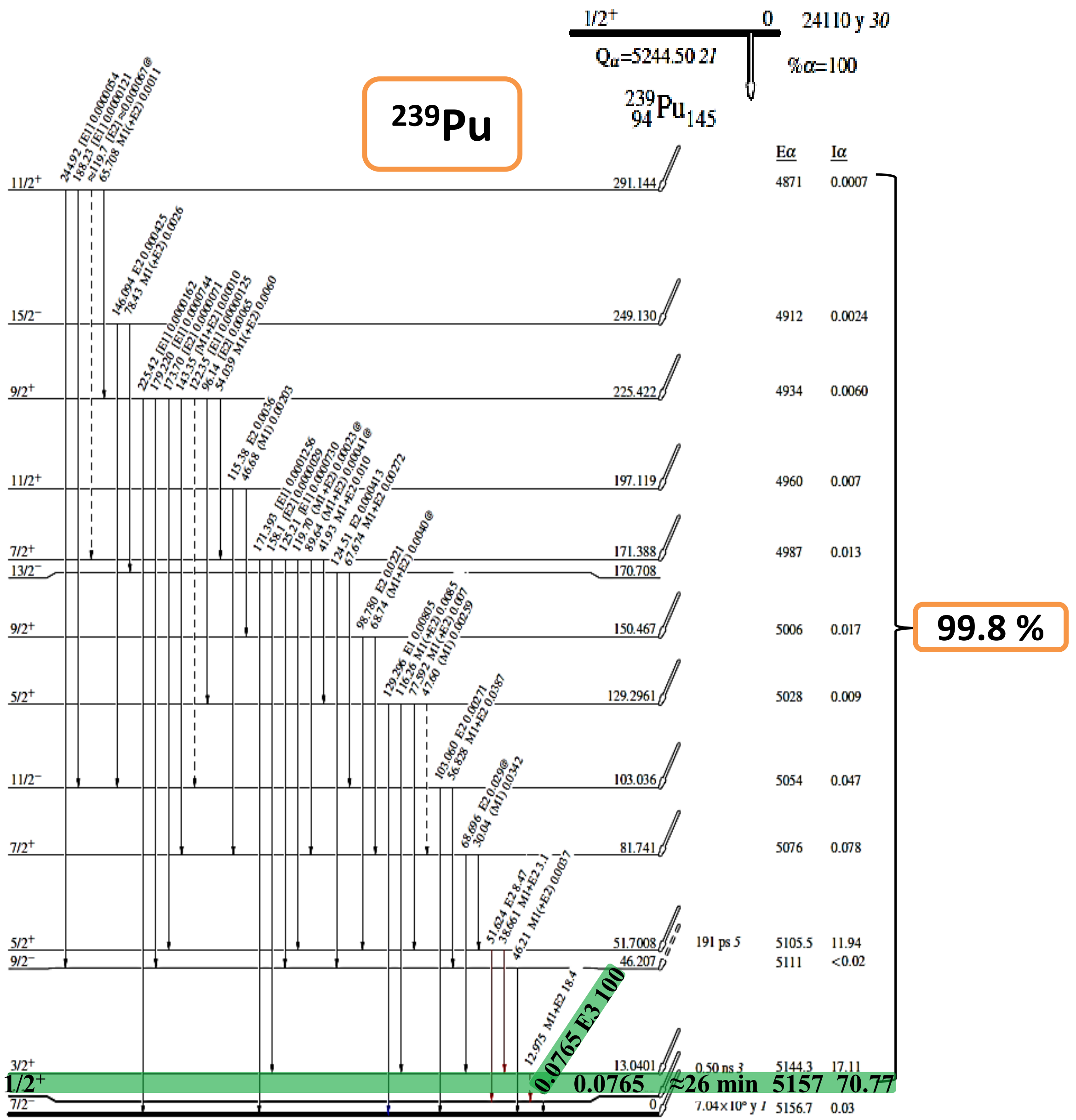
- ^{235m}U hyperfine structure → focus of **laser spectroscopy** study at Jyväskylä University
- Gives access to **nuclear properties** indicative of **nuclear structure**

[1] Y. Kasamatsu and H. Kikunaga, Radioisotopes 67, 471-482, (2018)

Motivations

- Recoil ion sources provide hard-to-obtain nuclides
- ^{239}Pu sources provides **^{235m}U recoil ions**
- ^{240}Pu provides ^{236}U for comparative measurements





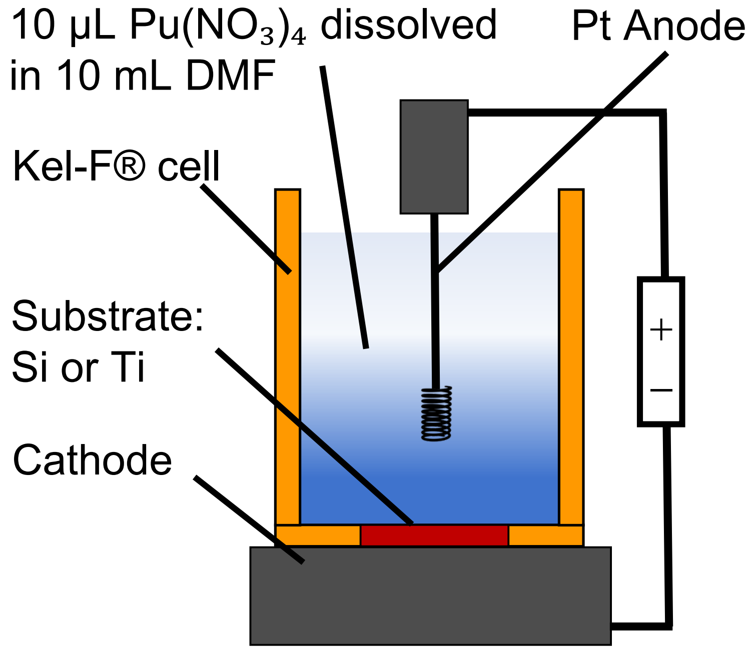
[1] Y. Kasamatsu and H. Kikunaga, Radioisotopes 67, 471-482, (2018)
[2] E. Browne and J. K. Tuli, Nucl. Data Sheets 122, 205 (2014)
[3] S. Zhu, Nucl. Data Sheets 182, 2 (2022)

Molecular plating

+ Reliable technique; long-term experience [4]
+ High yields possible (≥ 90 %) [5]
+ Homogeneous layers [6]

- Impurities from organic solvent
- Deposited chemical form undefined
- Mudcracking at thicker layers

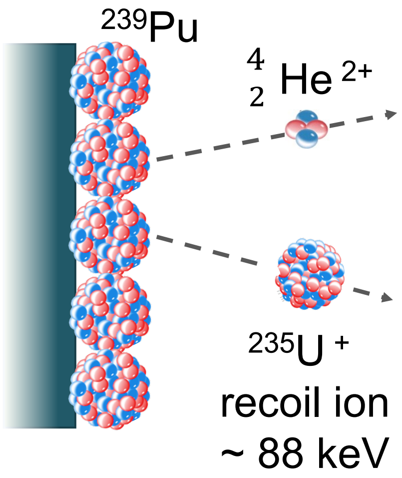
Voltage: 100 V - 800 V (Constant current)
Deposition time: 75 mins



[4] W. Parker and R. Falk, Nucl. Instrum. Meth. 16, 355–357 (1962)
[5] A. Vascon et al., Nucl. Instrum. Meth. in Phys. Research A 696, 180-191 (2012)
[6] A. Vascon et al., Nucl. Instrum. Meth. in Phys. Research A 714, 163-175 (2013)

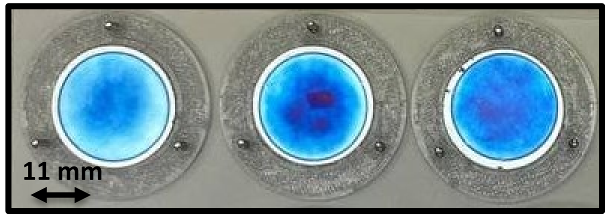
Molecular plating

- **Recoil efficiency** → Source key performance indicator
- Optimizing method gives more efficient recoil ion sources
- Total activity, source homogeneity, substrate roughness and plating solvent influence recoil efficiency [7]
- Optimal source thickness → ^{239}Pu ≈ 16 µg/cm² and ^{240}Pu ≈ 12 µg/cm²



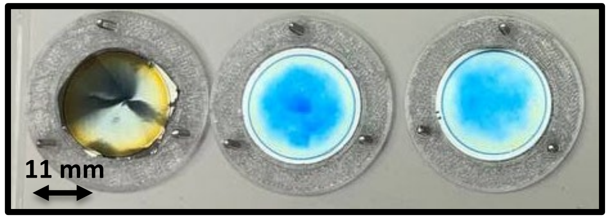
[7] A. Vascon et al., Nucl. Instrum. Meth. in Phys. Research A 721, 35-44 (2013)

Preparation of ^{239}Pu and ^{240}Pu recoil ion sources

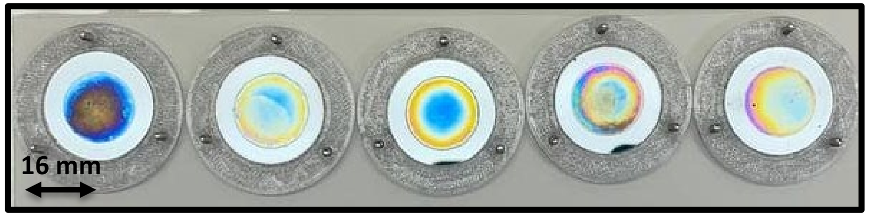


- $3 \times {}^{239}$Pu ~ 135 kBq
- ~ 16 µg/cm² thickness
- 22 mm diameter
- Si substrate

- ^{239}Pu ~ 57 kBq
- ~ 7 µg/cm² thickness
- 22 mm diameter
- 2 x Si, 1 x Ti substrate



- ^{239}Pu ~ 200 kBq
- ~ 23 µg/cm² thickness
- 22 mm diameter
- 2 x Si,1 x Ti substrate



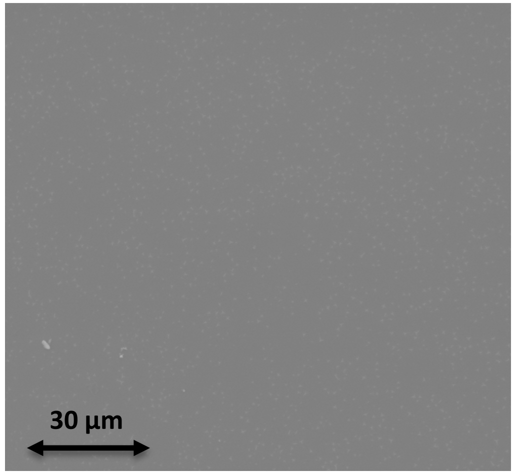
- ^{239}Pu Si substrate
- ~ 16 - 26 µg/cm² thickness
- 16 mm diameter
- 3 x ~ 75 kBq, 2 x ~ 120 kBq

- ^{239}Pu ~ 75 - 200 kBq
- ~ 9 - 23 µg/cm² thickness
- 22 mm diameter
- 3 x Si, 1 x Ti substrate
- Oven dried~ 200˚C for 1.5 hr

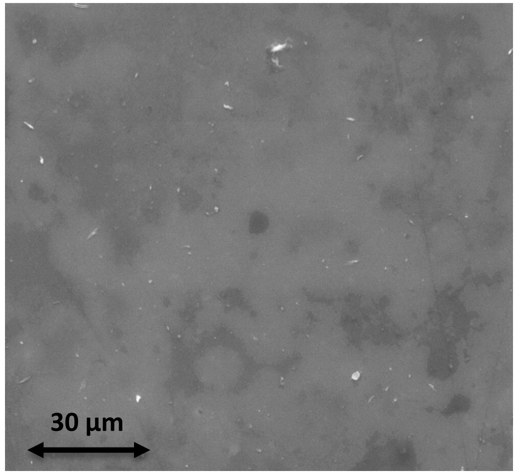
- ^{240}Pu ~ 200 - 400 kBq
- ~ 6 -12 µg/cm² thickness
- 22 mm diameter
- 2 x Si, 1 x Ti substrate

Characterization: Scanning Electron Microscopy (SEM)

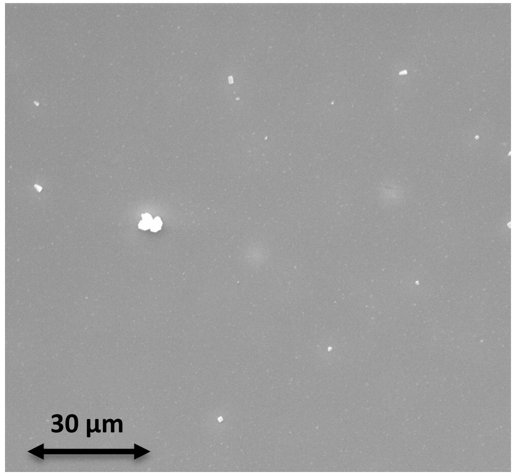
- Sources looked **homogeneous**
- Minimum visible contaminants on the deposition



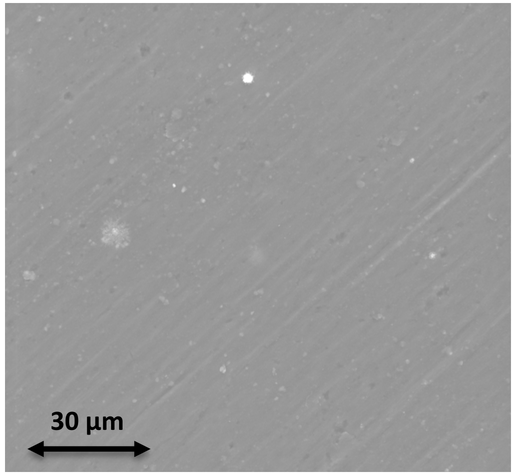
- ^{239}Pu ~ 135 kBq
- 22 mm diameter
- Si substrate
- x 1000 mag. 20 kV



- ^{239}Pu ~ 57 kBq
- 22 mm diameter
- Si substrate
- x 1000 mag. 20 kV



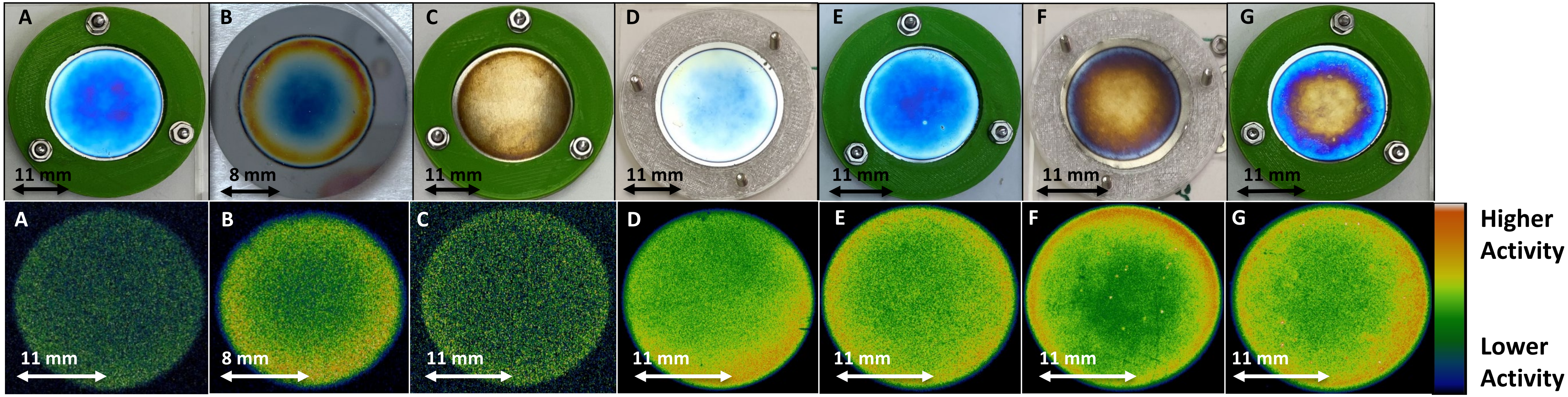
- ^{239}Pu ~ 75 kBq oven dried
- 22 mm diameter
- Si substrate
- x 1000 mag. 20 kV



- ^{239}Pu ~ 200 kBq
- 22 mm diameter
- Ti substrate
- x 1000 mag. 20 kV

Characterization: Radiographic imaging

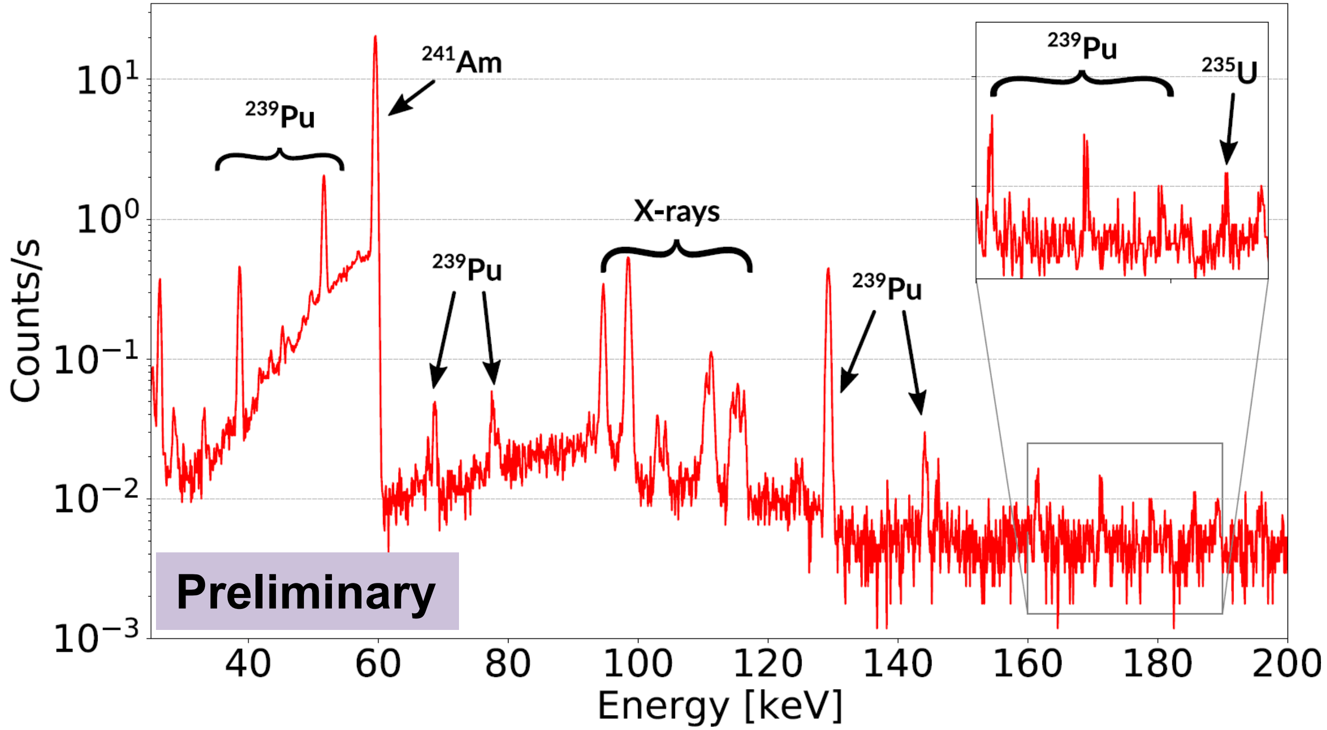
- Deposits had mostly a homogeneous activity distribution
- "Coffee ring" effect was not pronounced and only seen rarely



- Sent to Jyväskylä University for further analysis

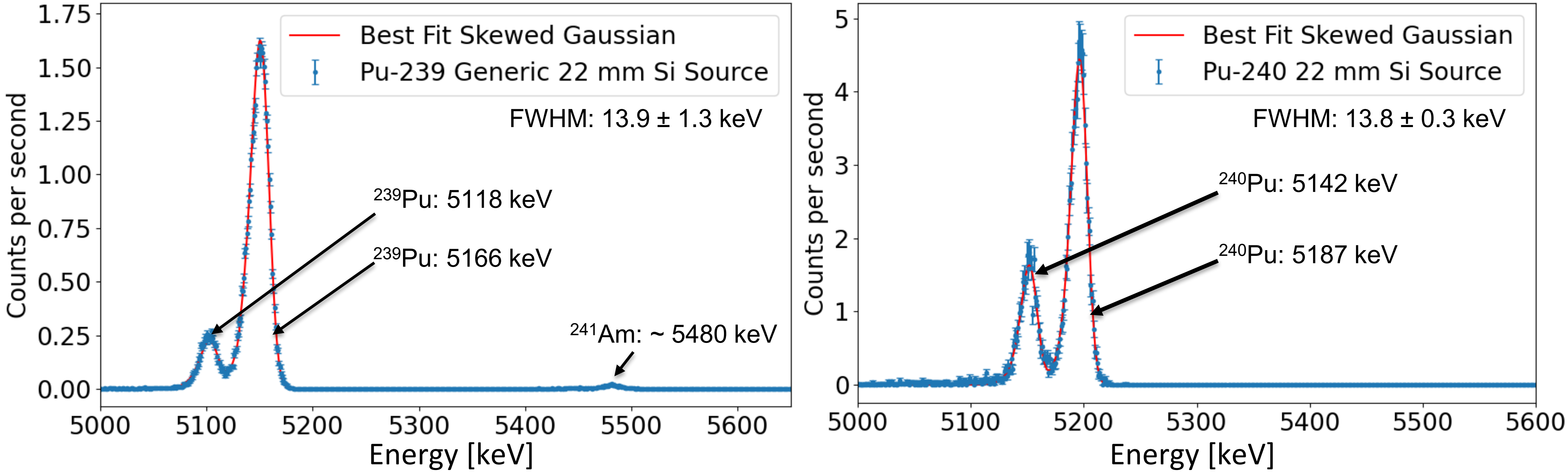
Characterization: γ-spectrometry

- Gamma spectrometry → Deposition yields and elemental composition
- Pu peaks were successfully identified
- Further analysis will quantify activities

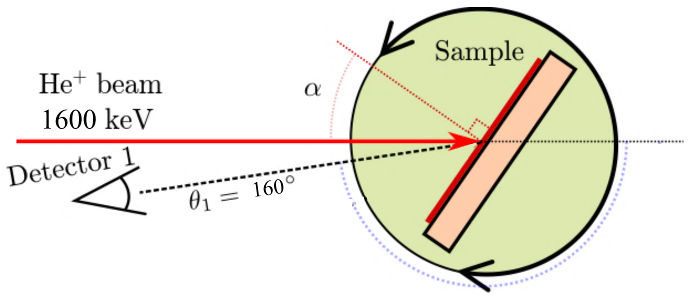


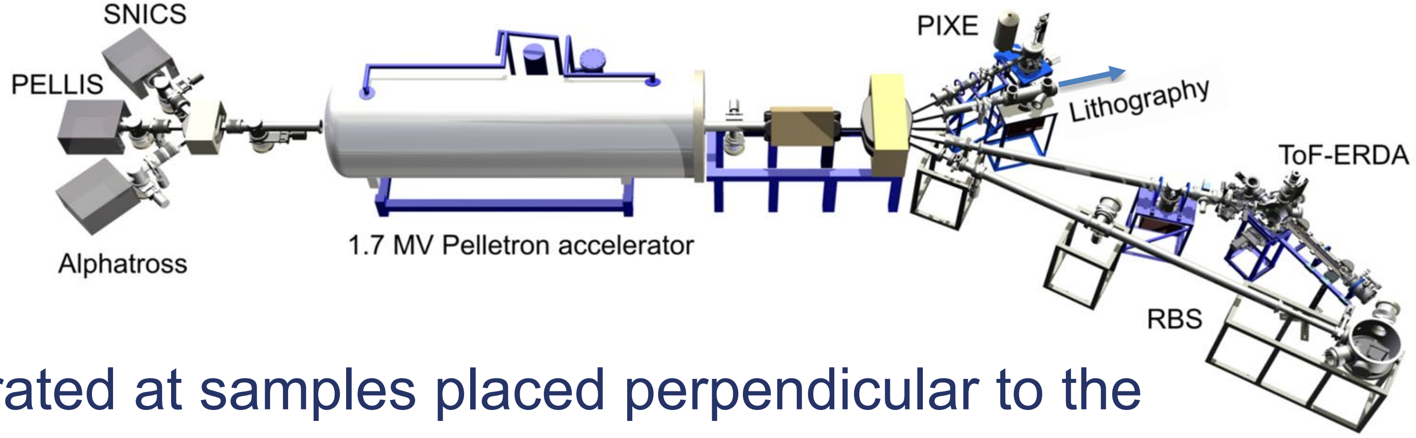
Characterization: α-spectrometry

- < 20 keV resolution achieved
- Minimum energy straggling → minimum organic layer
- **Deposition yields 85 - 95 %** → molecular plating was efficient



Rutherford Backscattering Spectroscopy (RBS) at JYU



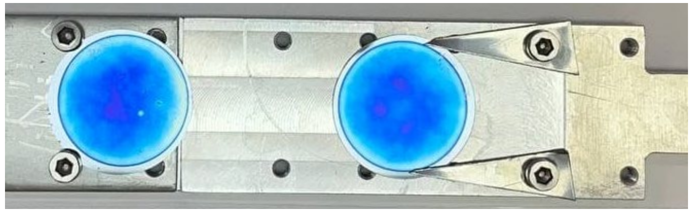


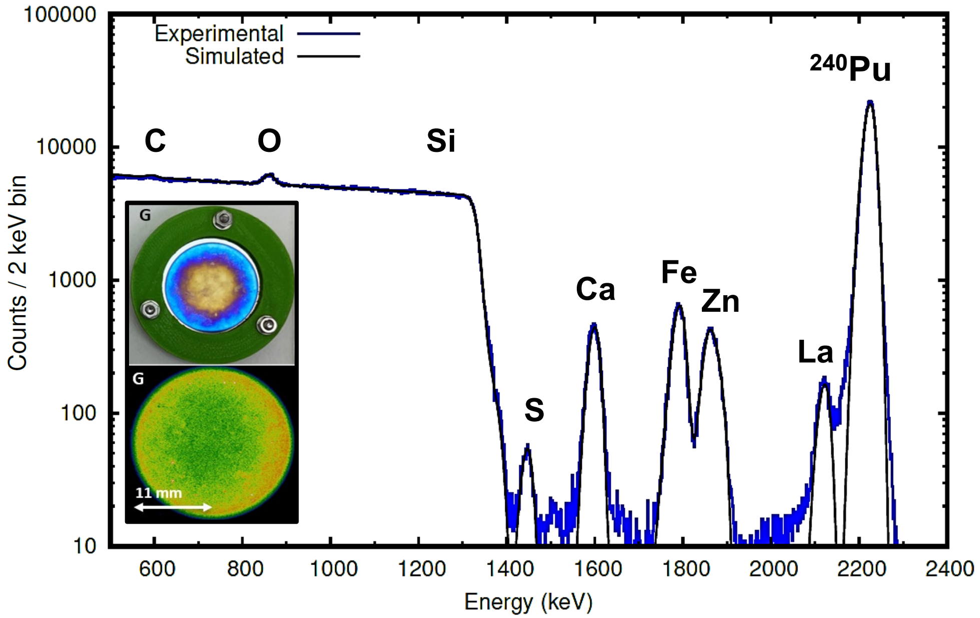
- $^4He^+$ or $^4He^{2+}$ beam accelerated at samples placed perpendicular to the beam
- Ions collide with sample atoms and are elastically backscattered
- Backscattered ions detected by a ring of 14 detectors at a scattering angle of 160°
- RBS spectra obtained by summing data from all detectors; gives information about source **elemental composition**, **depth profile** and **areal density**

[8] I. D. Moore, Pellatrpn Discussions [Presentation], Jyvaskyla, Finland, (2021)

Characterization: (RBS)

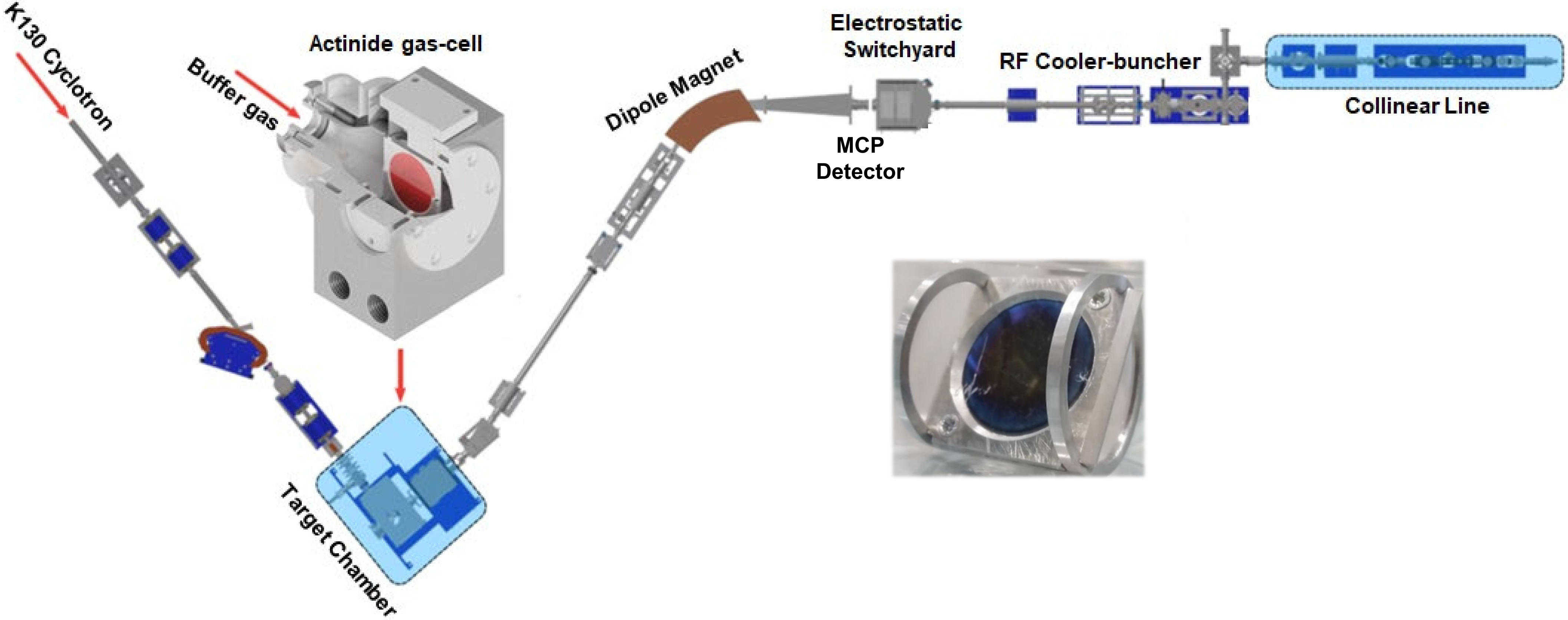
- O:Pu ratio → 6/7:1 and C:Pu ratio → ~ 4:1
- Fit suggests **formate species** compared to the oxide
- Confirms α-spectrometry Pu activities
- 10 % more Pu found at edges of some sources
- Contaminant ratios have high uncertainty due to mass overlaps
- Some measurements difficult to simulate
  - Lateral inhomogeneity (thickness variation)





U recoil yields – working towards laser spectroscopy of ^{235m}U

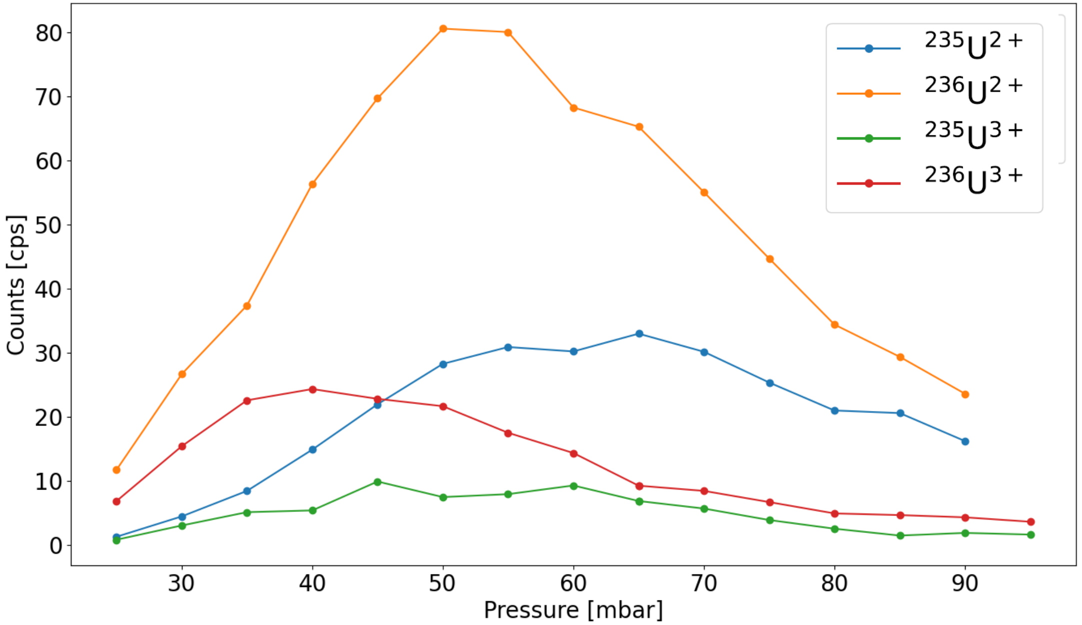
- **U recoil yields**: Recently at JYU U recoils were stopped in He buffer gas, extracted, mass separated and detected via MCP



[9] A. Raggio et al., LISA Summer School [Poster], Normandy, France, (2022)
[10] A. Raggio et al., Physics Day [Poster], Jyvaskyla, Finland, (2021)

U recoil yields – working towards laser spectroscopy of ^{235m}U

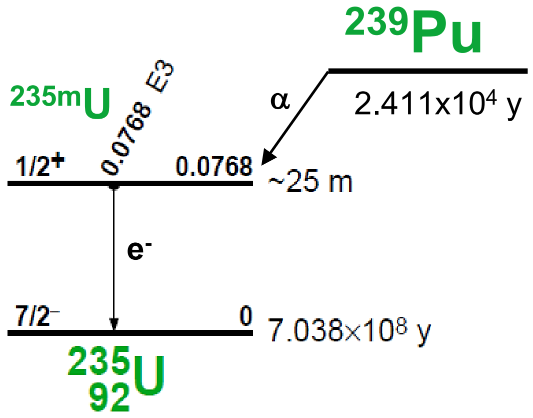
- Count rate insufficient for laser spectroscopy → extraction and transmission efficiency low → Gas cell efficiency optimised for winter.
- 2+ and 3+ states observed; **charge state manipulation to 1+** will be investigated for laser spectroscopy next year by changing buffer gas

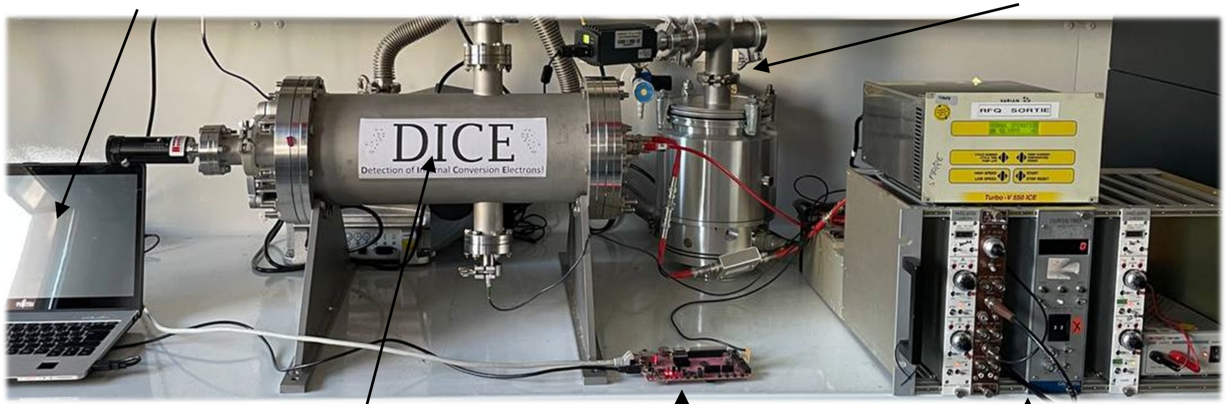


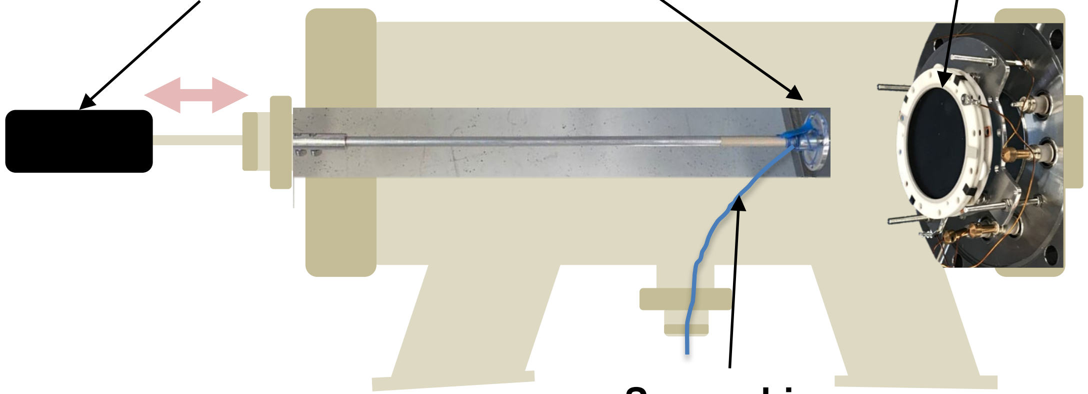
[9] A. Raggio et al., LISA Summer School [Poster], Normandy, France, (2022)

Characterization: DICE setup

- **DICE** – Detection of Internal Conversion Electrons MCP novel setup at JGU aims to provide recoil efficiency data
- Detects ^{235m}U to ^{235}U conversion electrons



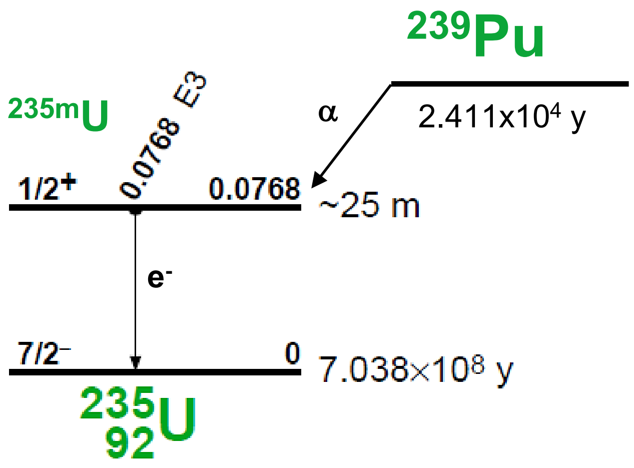


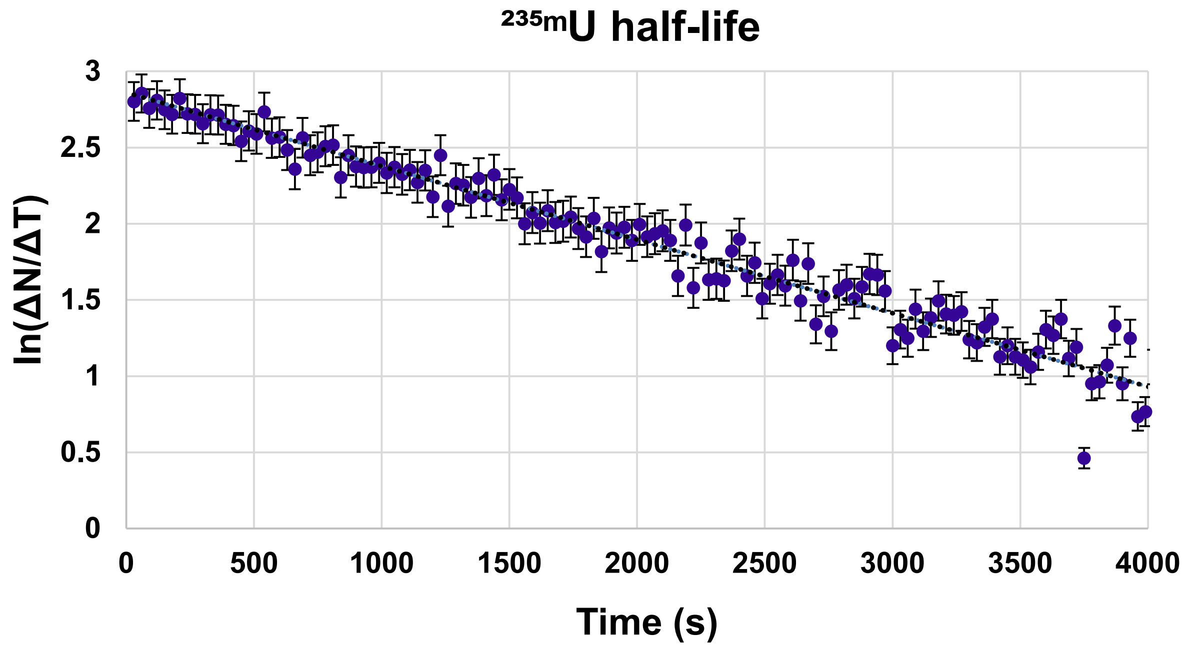


[1] Y. Kasamatsu and H. Kikunaga, Radioisotopes 67, 471-482, (2018)

Characterization: DICE

- Electrons from ^{235m}U to ^{235}U transition have been detected!
- Preliminary results give a **26.00 ± 0.76 minute** half life for ^{235m}U
- Studies are underway to establish the system





[1] Y. Kasamatsu and H. Kikunaga, Radioisotopes 67, 471-482, (2018)
[9] Y. Shigekawa et al., Phys. Rev. C 98, 1-5, (2018)

Acknowledgements

- JGU radiation protection
- JGU mechanical and electrical workshops
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- JYU radiation protection
- JYU mechanical workshop
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- LISA ITN

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Thank you for your attention!

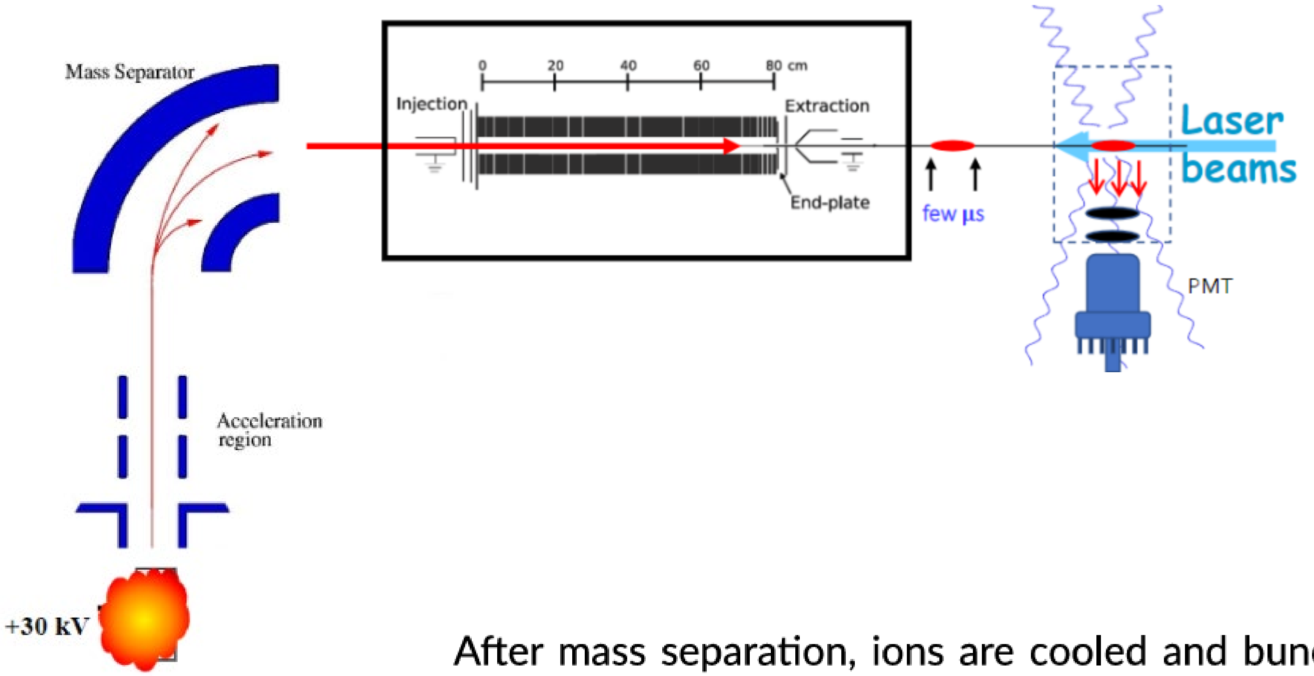
RBS at JYU – Results

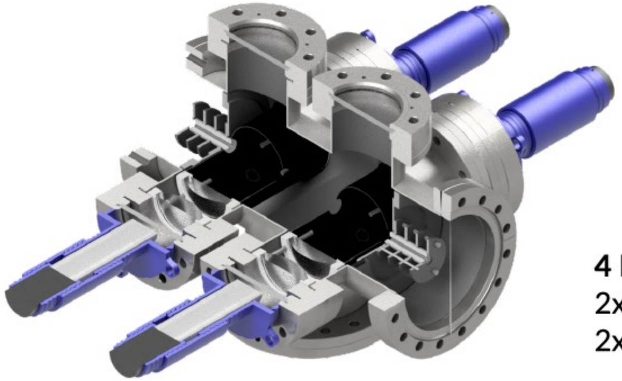
	Areal density (10^{15} at./cm^2)	Ratio to Pu					
Sample	Pu	S	Cl	K	Ca	Fe	Zn
22D dried	40.7	0.09			0.15	0.64	0.17
22B	35.6	0.10			0.48	0.71	0.16
2250B	11.6	0.16	0.26	0.12	0.23	0.67	0.26
22150B	47.7	0.05		0.08	0.08	0.70	0.07
16D	43.6	0.19			0.48	0.48	0.48
223	24.9	0.09			0.47	0.44	0.26

RBS at JYU – Some notes

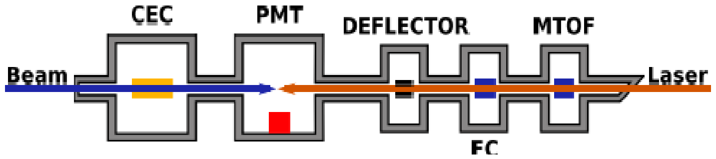
- 1.6 MeV $^{4}He^{1+}$ or 2.4 MeV $^{4}He^{2+}$ beam used.
- RBS has poor sensitivity to light elements
- Beam fluence is normalized based on backscattering signal from a beam chopper
- RBS analysis is based on simulating and fitting spectra to experimental spectra. This was performed using JaBS. Concentrations were fitted by hand, while the software fits e.g. layer thickness and roughness.
- Beam fluence calibration was determined using known samples (SiO2). Statistical uncertainty in fluence normalization of individual measurements is less than 1%.
- Uncertainty in quantification of Pu areal density includes statistical uncertainty, fitting uncertainty and other contributions (e.g. uncertainty in measurement geometry and beam energy) - expected < 2%. Differences between samples can be determined with higher precision. Fitting errors of other elements are much greater and difficult to estimate.

Collinear Laser Spectroscopy explained





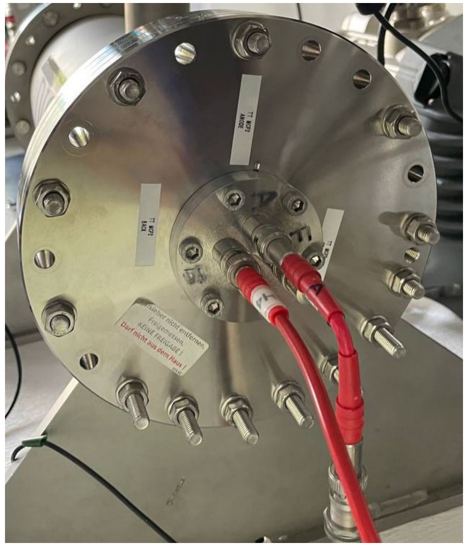
4 PMT tubes:
2x R6427 300nm to 650nm
2x R6834 160nm to 330nm

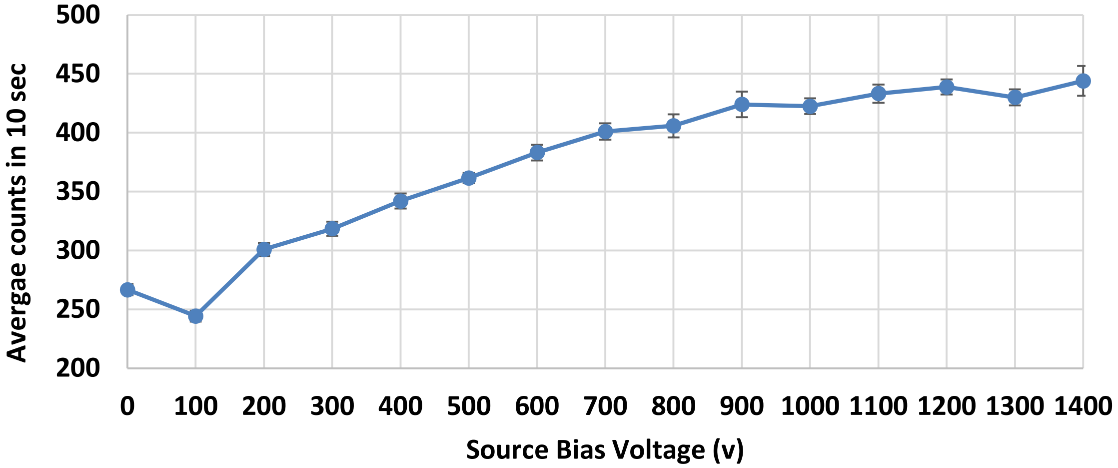


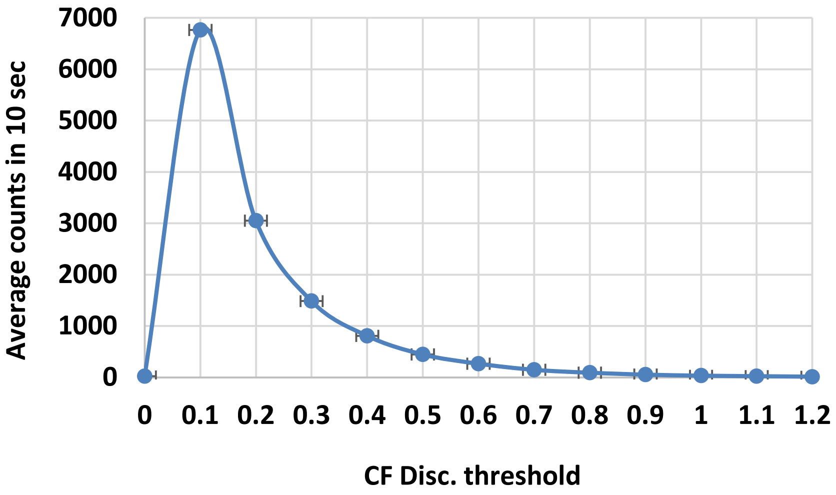
After mass separation, ions are cooled and bunched in a radiofrequency cooler-buncher, before being reaccelerated to 30 keV and delivered to the collinear beamline. There, ions are overlapped in a counter-propagating geometry with a CW laser beam which excites them from either the ground state or low-lying metastable state. Detection of laser-induced fluorescence allows the measurement of IS and HFS from the frequency pattern

[8] I. D. Moore, Pellatrpn Discussions [Presentation], Jyvaskyla, Finland, (2021)
[9] A. Raggio et al., LISA Summer School [Poster], Normandy, France, (2022)
[10] A. Raggio et al., Physics Day [Poster], Jyvaskyla, Finland, (2021)

DICE




Avergae counts in 10 sec
500
450
400
350
300
250
200
0 100 200 300 400 500 600 700 800 900 1000 1100 1200 1300 1400
Source Bias Voltage (v)


Average counts in 10 sec
7000
6000
5000
4000
3000
2000
1000
0
0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1 1.1 1.2
CF Disc. threshold

N,*N*-Dimethylmethanamide (DMF) solvent