



Development of a laser ion source for plutonium and thorium at the IGSOL facility.



Laser Ion Source Workshop

Paris, Oct 23-25, 2016



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Introduction

High resolution laser spectroscopy on actinide elements

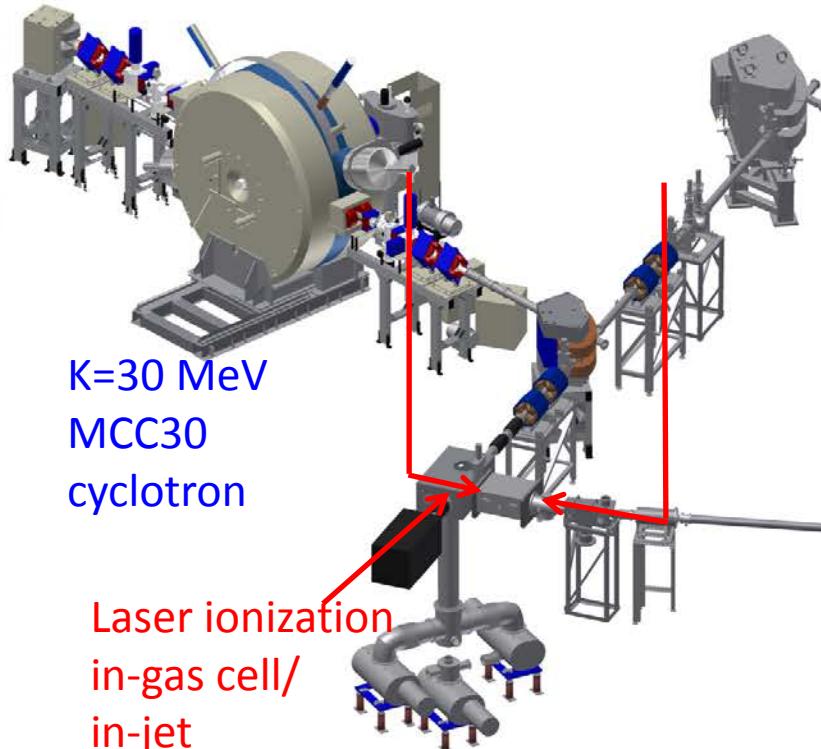
← Long-lived isotopes from nuclear reactors

Outline

- Short overview of the IGISOL facility
- Plutonium
 - Actinide gas cell
 - Laser ion source of Pu isotopes for Collinear laser spectroscopy
- Thorium
 - Studies of ^{232}Th samples
 - U-233 source for $^{229(\text{m})}\text{Th}$
 - Outlook for $^{229\text{m}}\text{Th}$



The IGISOL facility



from K=130 MeV
cyclotron

Off-line ion sources:
(discharge, surface,
carbon cluster...)

$M/\Delta M \approx 500$

Decay spectroscopy
line

Mass spectrometry
& post-trap spectroscopy

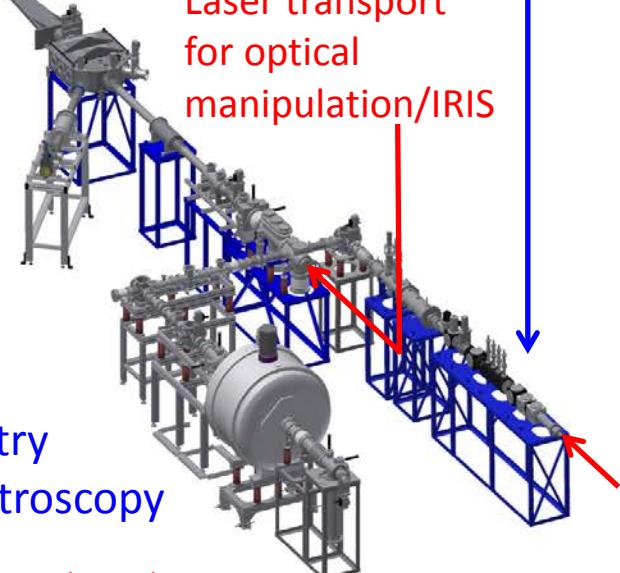
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The University of Manchester

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LIVERPOOL

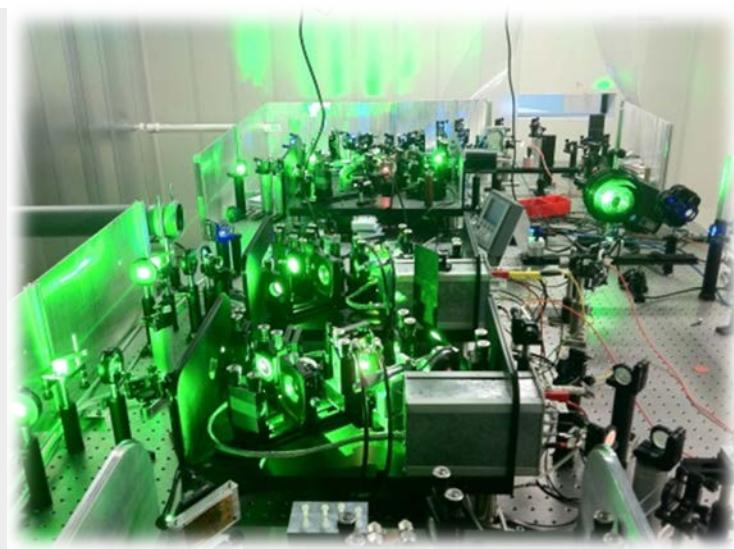
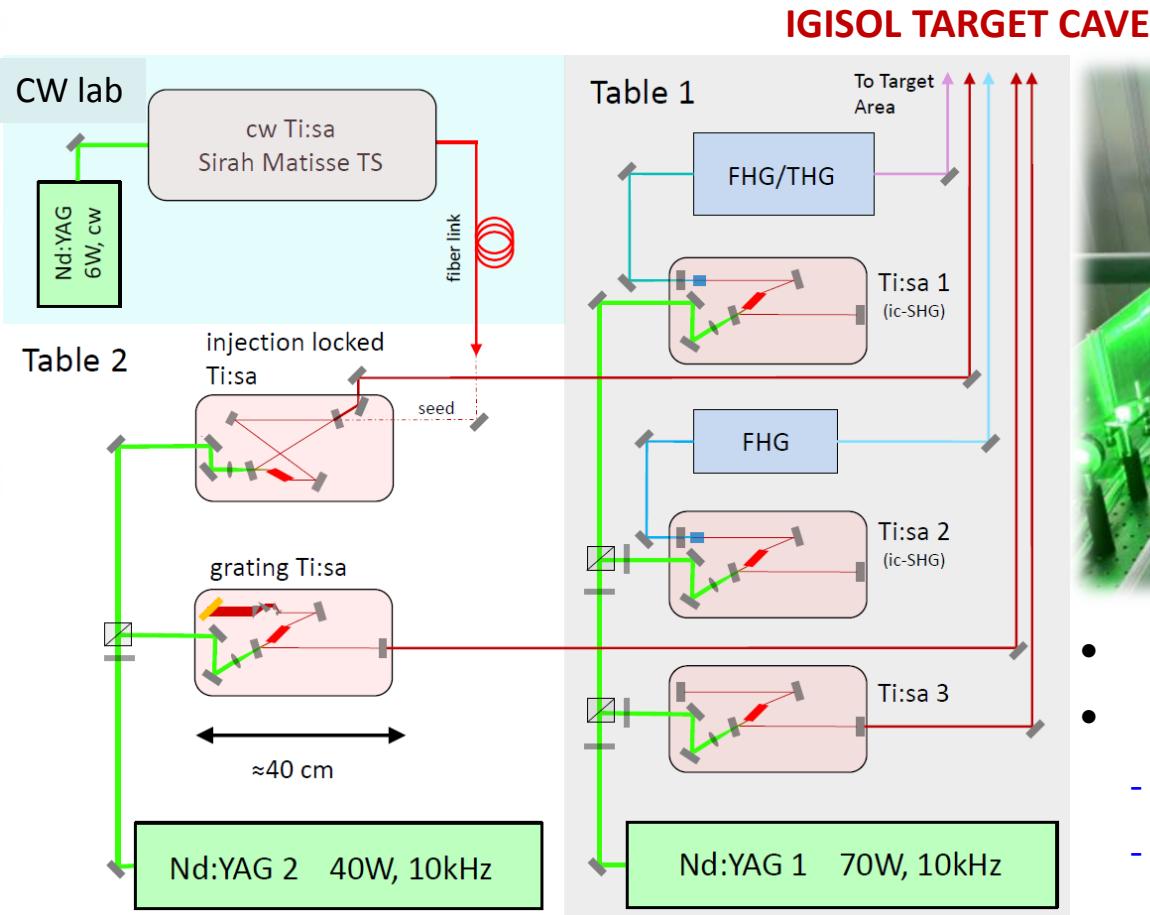
Collinear laser
spectroscopy

Laser transport
for optical
manipulation/IRIS



I.D. Moore et al., NIMB 317 (2013) 208

The FURIOS laser facility

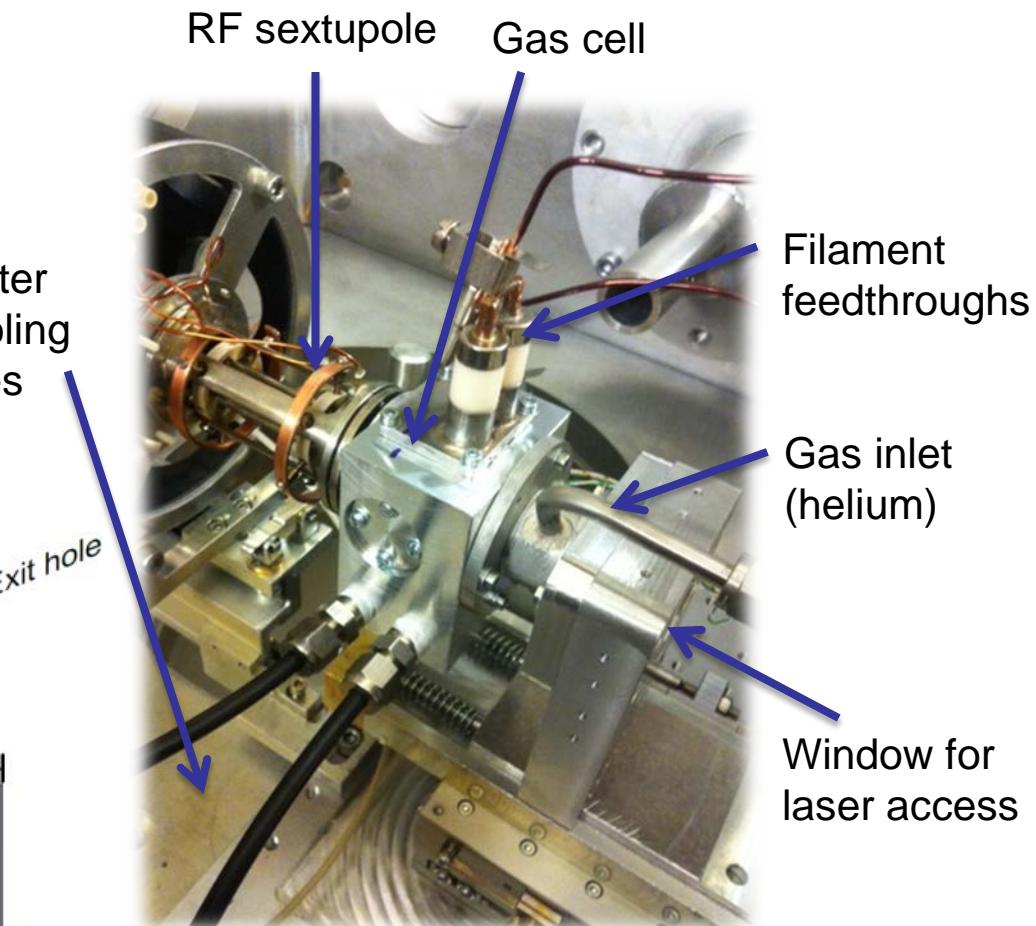
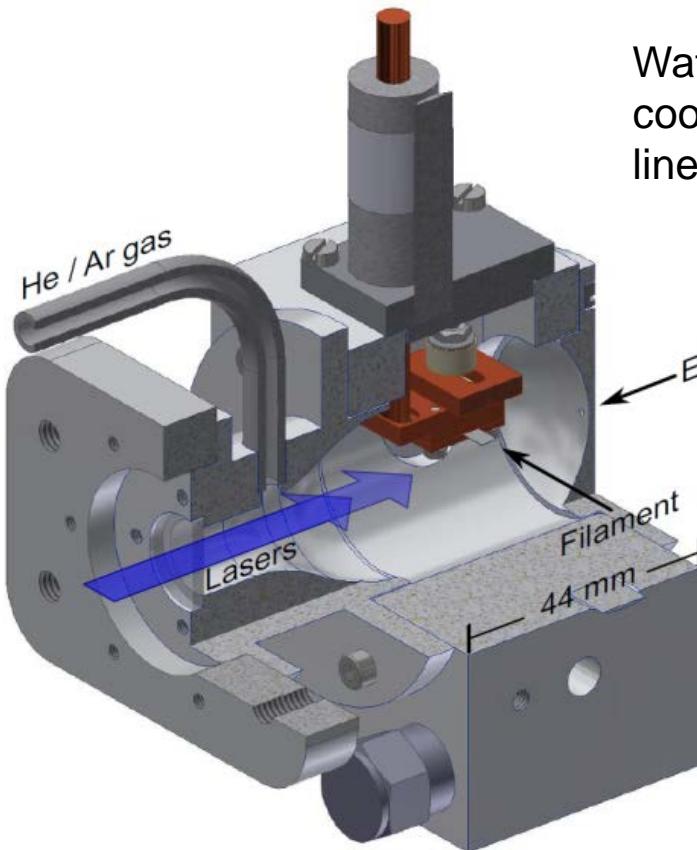


- Continuous wave laser laboratory
(Last talk tomorrow: A. Voss)
- repetition rate: ~10 kHz
- tuning range:
 - fundamental 680 - 1050 nm
 - frequency doubled 340 - 525 nm
 - frequency tripled 250 – 330 nm
 - frequency quad. 205 - 250 nm
- laser linewidth: >5 GHz (broad)
<1 GHz (narrow)

M. Reponen, I.D. Moore et al., Eur. Phys. J. A 48 (2012) 1

A gas cell for (off-line) actinide studies

- Gas cell volume ~30 cm³
- Exit hole $\phi = 1.2$ mm
- $T_{\text{evac}} \sim 50$ ms in He gas



Laser ionization of plutonium at JYFL

- Tantalum filaments from Mainz with $^{238-240,242,244}\text{Pu}$ plutonium isotopes
- **Goal:** Provide sufficient beam of plutonium isotopes ($1+$) to the collinear laser spectroscopy station for IS and HFS measurement

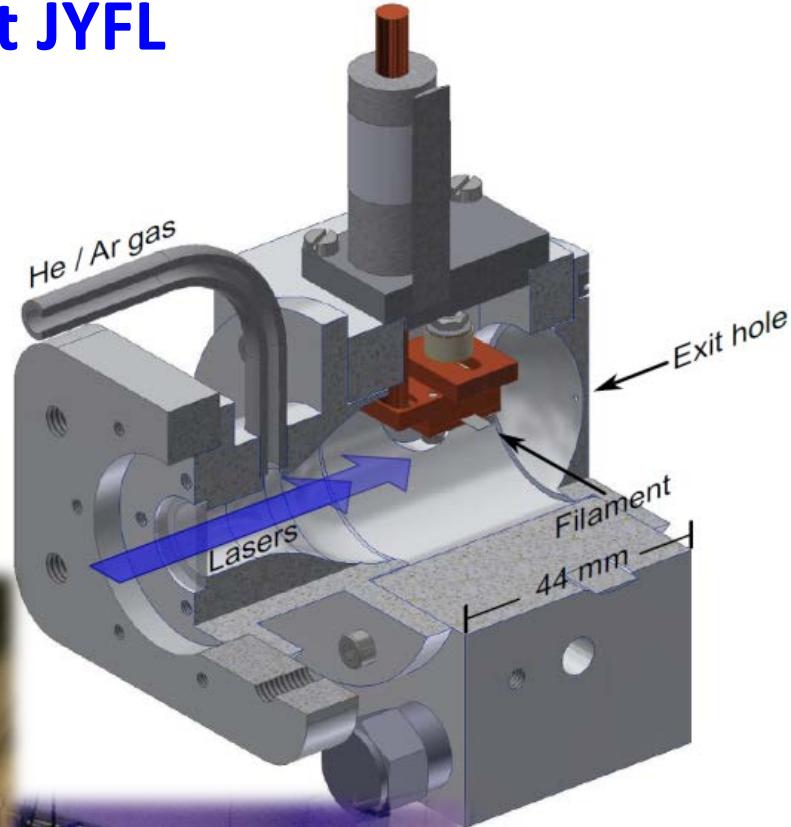
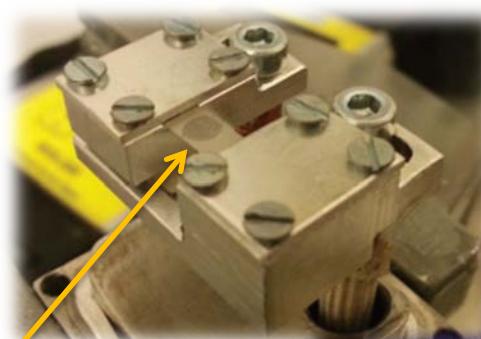


Filaments:
Nuclear Chemistry
department
of the University Mainz

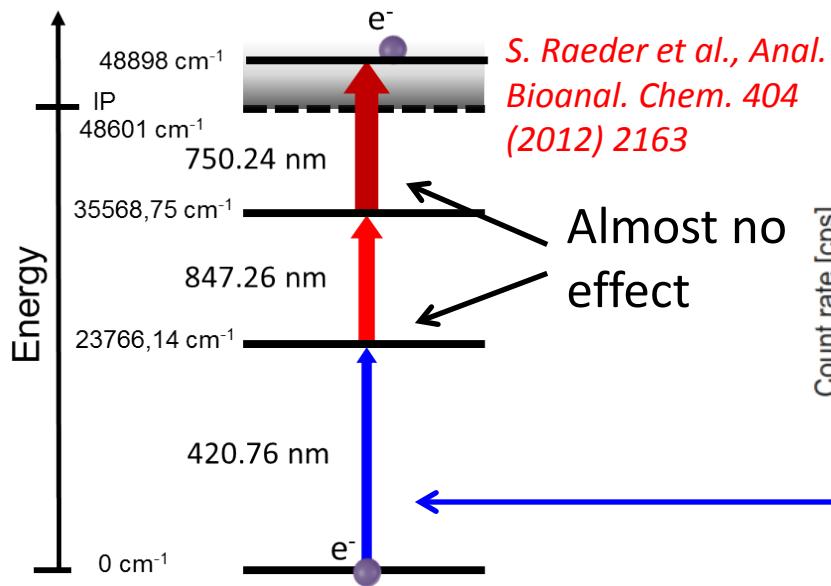
Ti $\sim 1\mu\text{m}$
 $^{238-240,242,244}\text{Pu}$

Ta 50 μm

1000-1200 °C

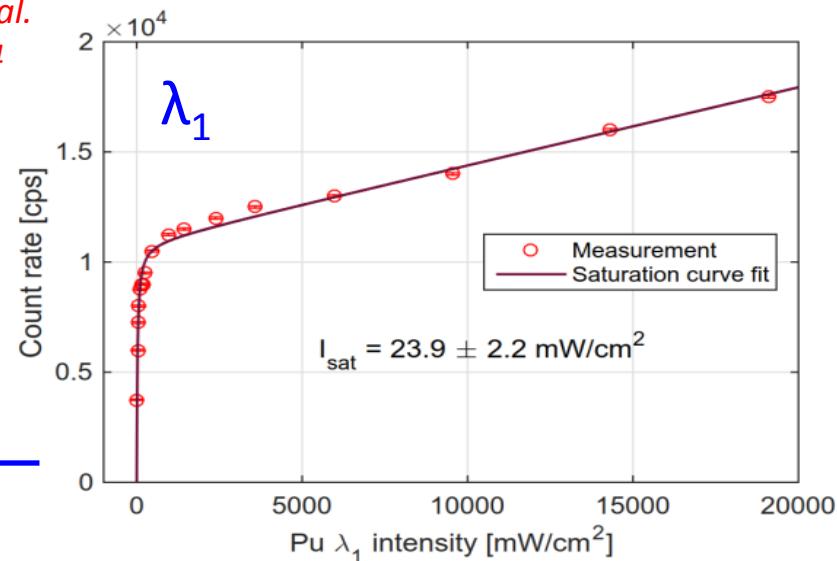


Laser ionization of plutonium at JYFL



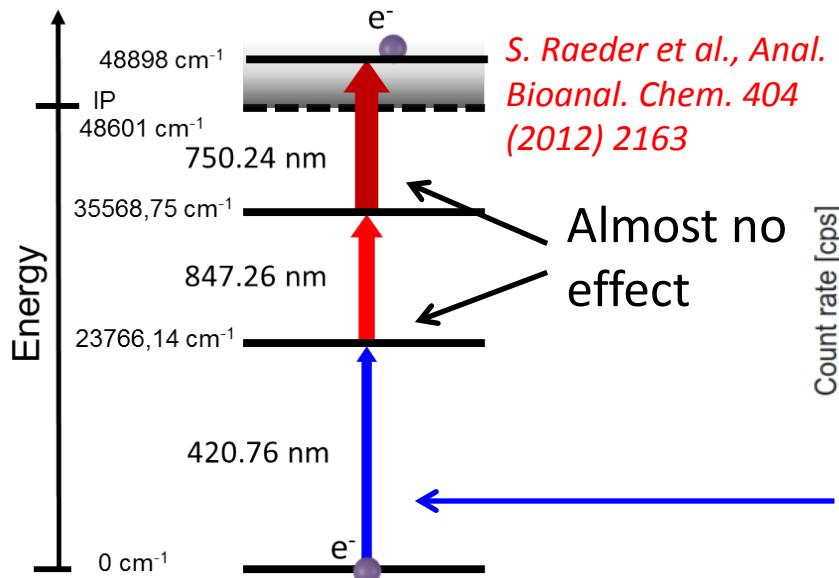
S. Raeder et al., Anal.
Bioanal. Chem. 404
(2012) 2163

Almost no
effect



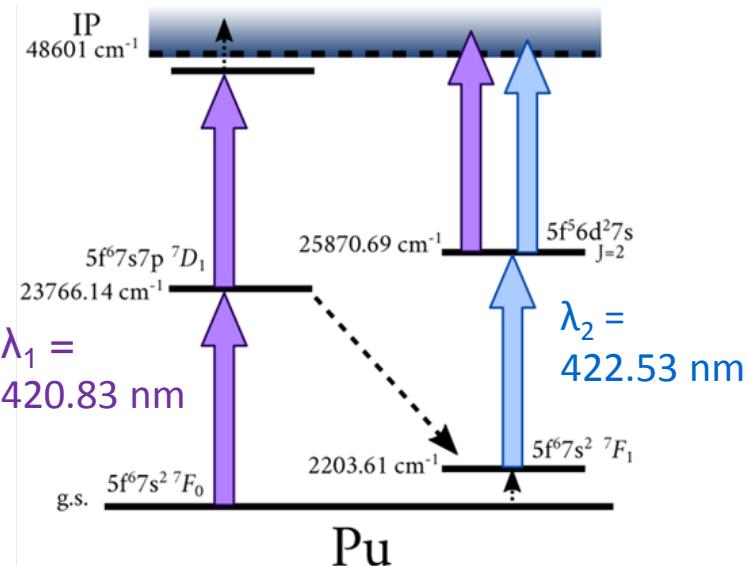
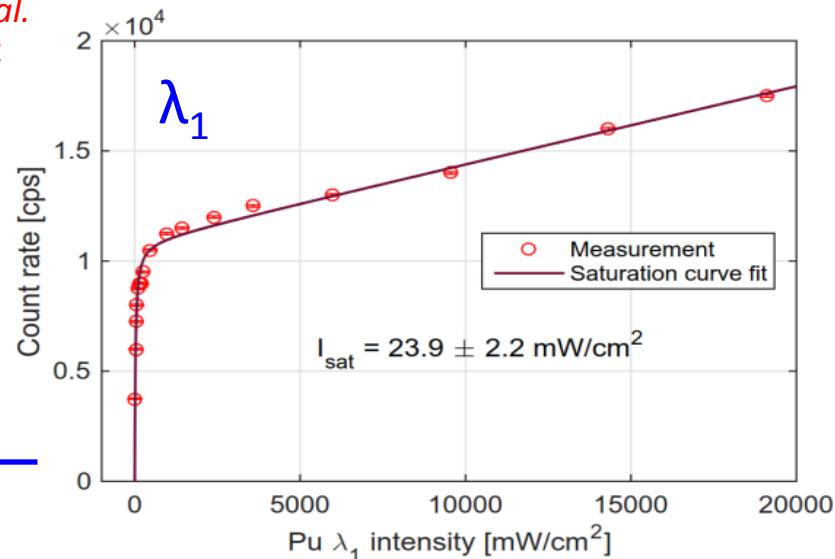
I. Pohjalainen, I.D. Moore, et al., NIMB 376 (2016) 233

Laser ionization of plutonium at JYFL



S. Raeder et al., Anal.
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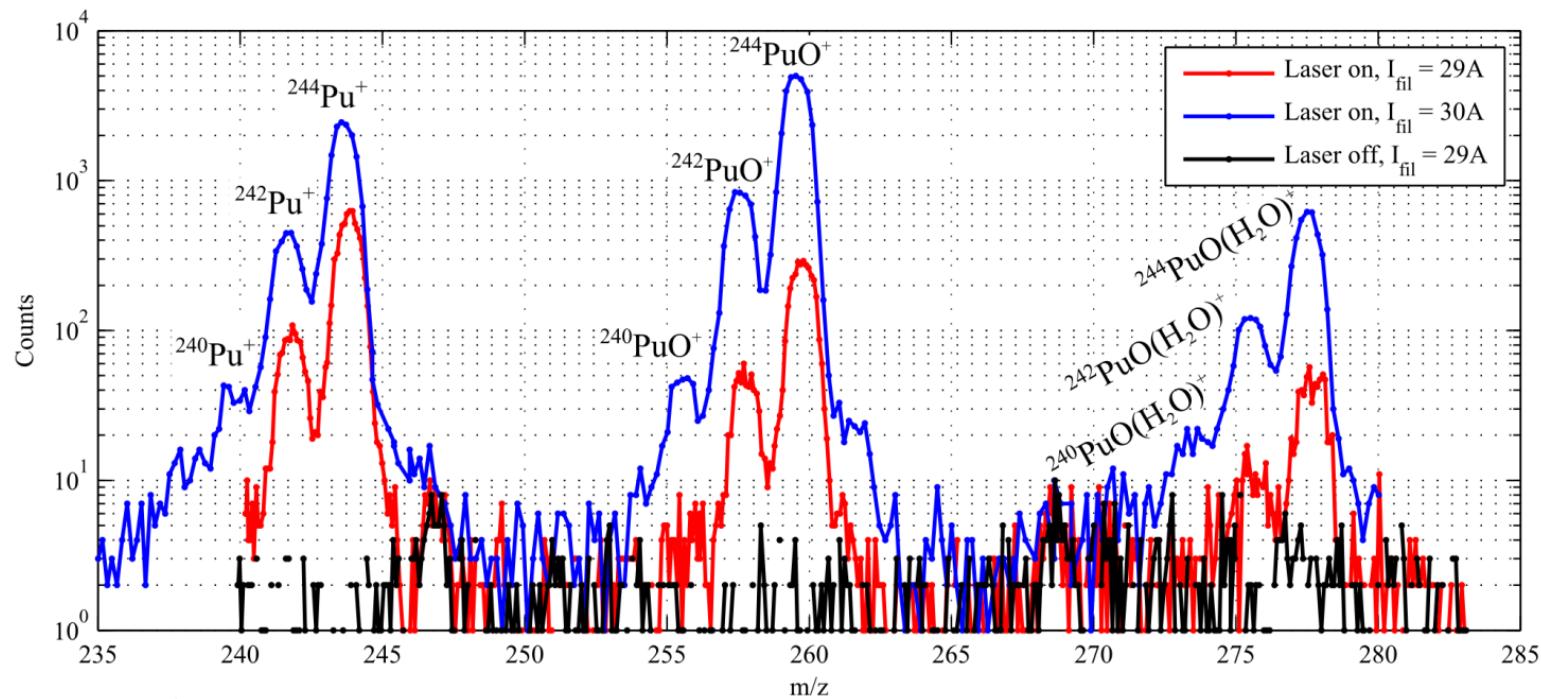


Gas cell environment!

- λ_2 reacts independently from λ_1
- Population mechanism of 2203 cm⁻¹?
- Strongest yields without etalons
→ Laser output very broad
100 GHz ($\sim 0.2 \text{ nm}$)

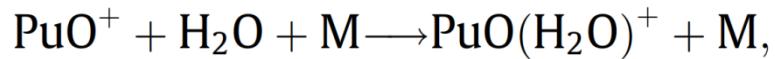
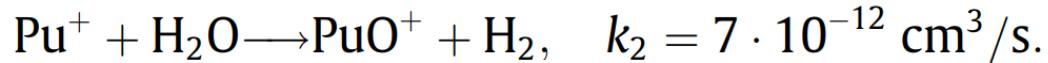
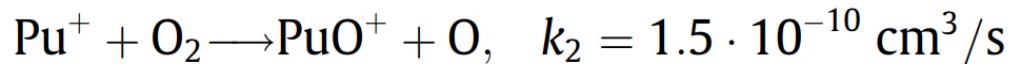
I. Pohjalainen, I.D. Moore, et al., NIMB 376 (2016) 233

Laser ionization of plutonium at JYFL



Successful laser ionization Pu isotopes

- Isotopic pattern consistent with the sample assay
- Clear chemistry with impurities ← Outgassing, dead volume



I. Pohjalainen, I.D. Moore, et al., NIMB 376 (2016) 233

Laser ionization of plutonium at JYFL

- Beams of several Pu isotopes has been successfully provided for the collinear laser station (Nov. 2015)
→ Check to talk tomorrow by A. Voss for the results!

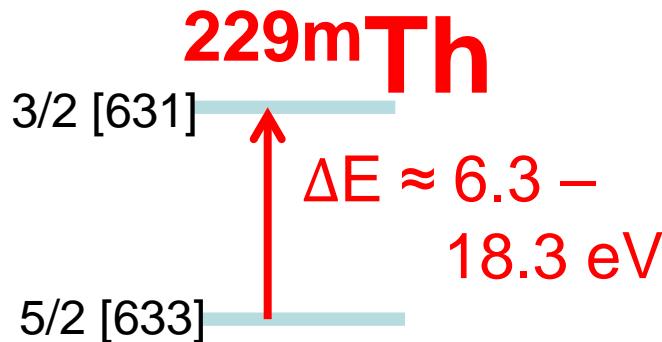


Laser ionization of plutonium at JYFL

- Beams of several Pu isotopes has been successfully provided for the collinear laser station
 - Check to talk tomorrow by A. Voss for the results!
- Details of the blue-blue scheme under investigation
 - More plutonium filaments from University of Mainz
 - Measurement of the gas temperature
 - Blue grating laser from Nagoya



Towards CLS of Th-229 isomer at JYFL



$T_{1/2} \cdot {}^{229(m)}\text{Th} < 1 \text{ s}; {}^{229(m)}\text{Th}^{2+} > 60 \text{ s}$

→ don't try to work with neutral (+1) atom!

- Neutral has “collapsed” HFS: v. Sonnenschein, S. Raeder, IM et al., J. Phys. B 45 (2012) 165005



- a) Identify and characterize the ${}^{229}\text{Th}$ isomer transition
- b) Implement key components to operate a nuclear clock

ARTICLE

doi:10.1038/nature17669

Direct detection of the ${}^{229}\text{Th}$ nuclear clock transition

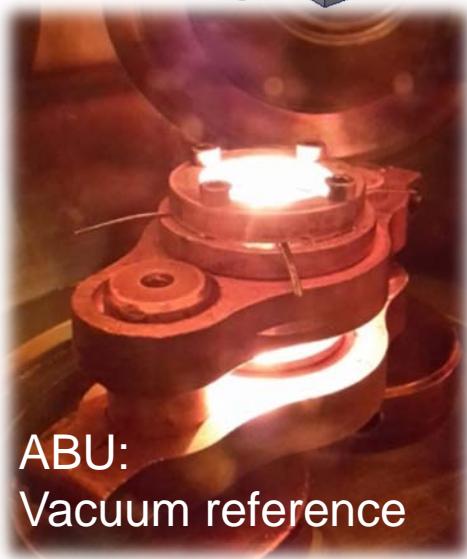
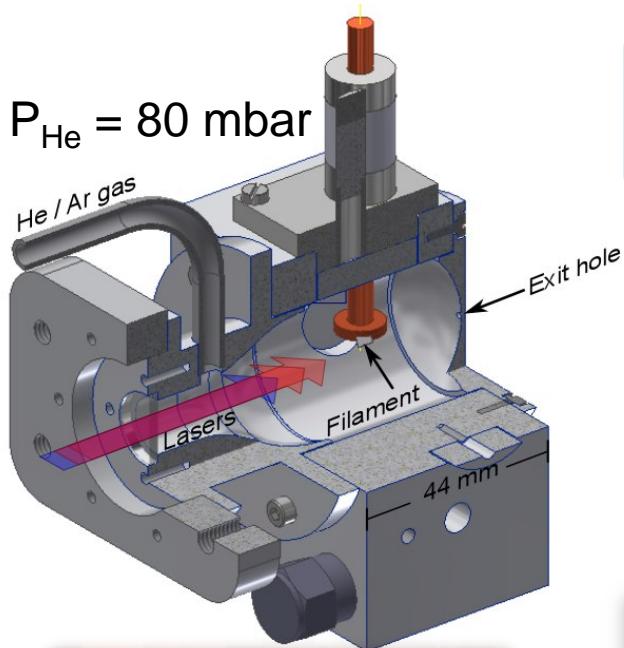
Lars von der Wense¹, Benedict Seiferle¹, Mustapha Laatiaoui^{2,3}, Jürgen B. Neumayr¹, Hans-Jörg Maier¹, Hans-Friedrich Wirth¹, Christoph Mokry^{3,4}, Jörg Runke^{2,4}, Klaus Eberhardt^{3,4}, Christoph E. Düllmann^{2,3,4}, Norbert G. Trautmann⁴ & Peter G. Thirolf¹



Identification of the isomer by measuring the HF structure (CLS)

- Source for ${}^{229}\text{mTh}$
 - U-233 alpha recoil source
 - on-line ${}^{232}\text{Th}(p,p3n){}^{229}\text{mTh}$
- ${}^{229}\text{Th}$ template measurement (ground state only) using filament
 - Resonance laser ionization of ${}^{232}\text{Th}$

Laser ionization of thorium at JYFL



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WIEN

Filaments:

From TU Wien

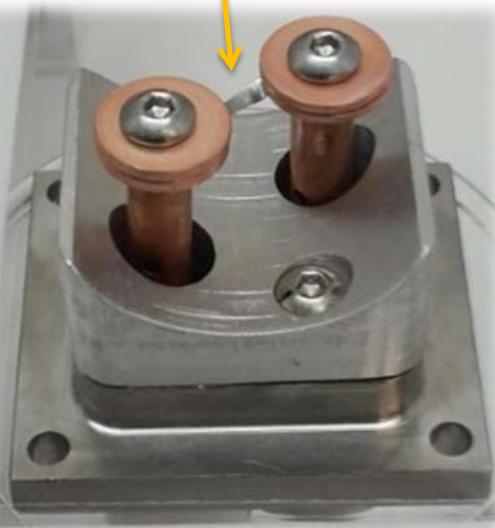
(Ti) / Zr $\sim 1 \mu\text{m}$

$\text{Th}(\text{NO}_3)_4$

Ta $50 \mu\text{m}$

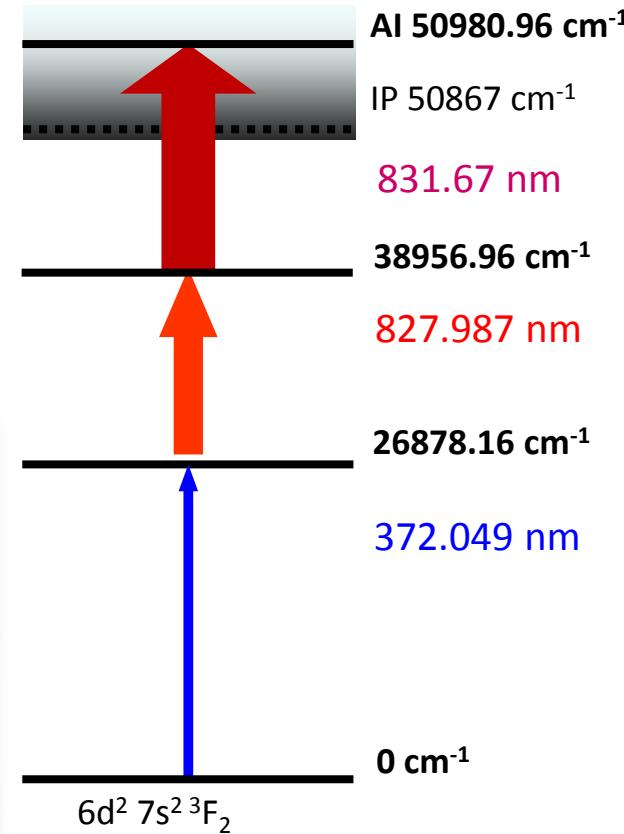
$>2000^\circ\text{C}$

10^{15} Th atoms



filament holder

3-Step ionization Scheme

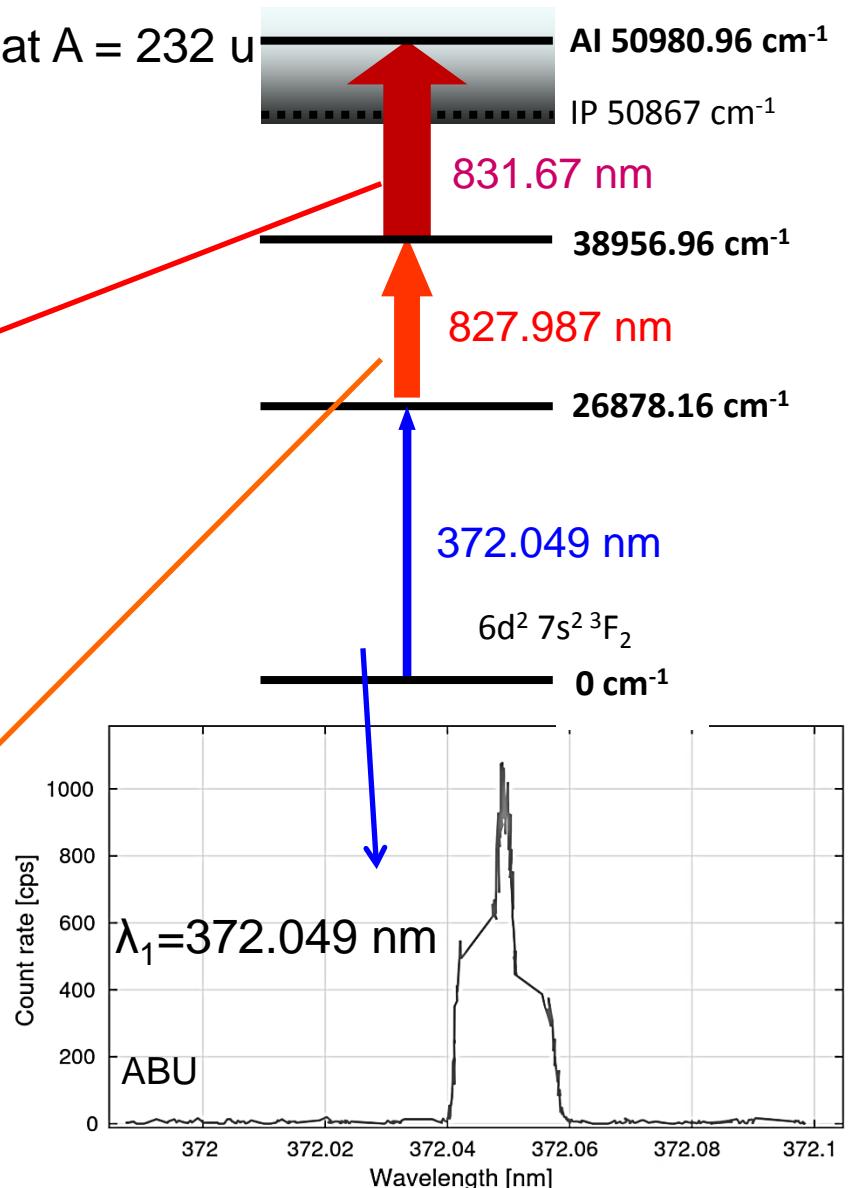
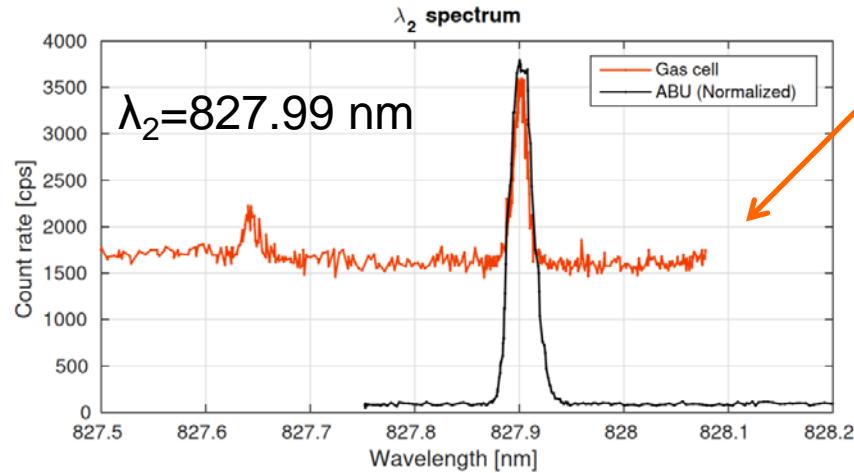
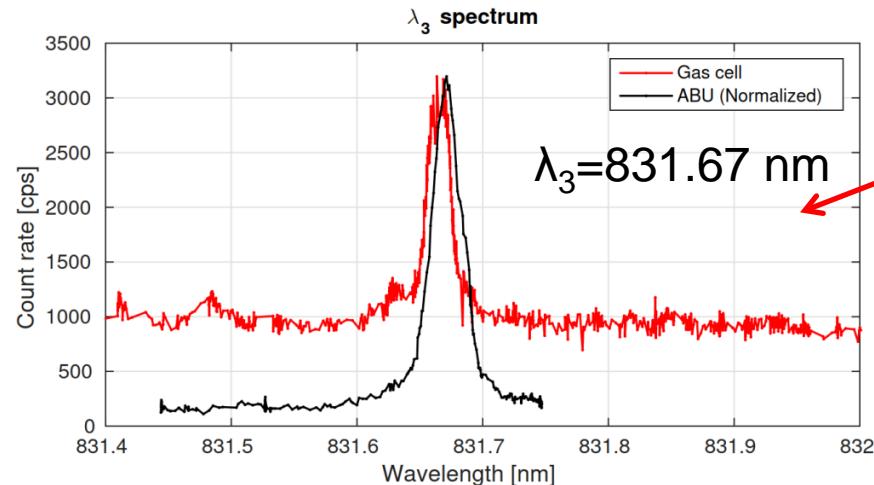


Oak Ridge National Laboratory

Y. Liu and D. Stracener,
NIMB 376 (2016) 233

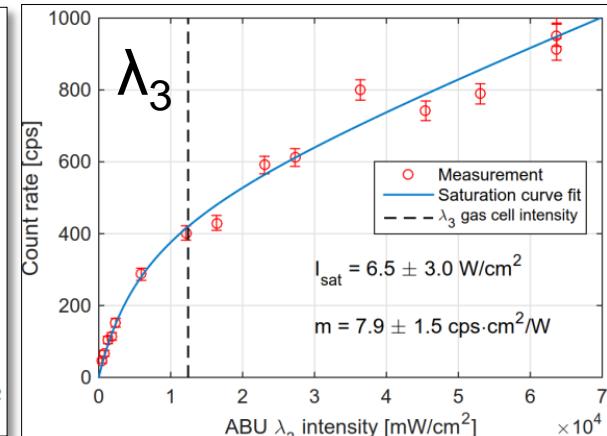
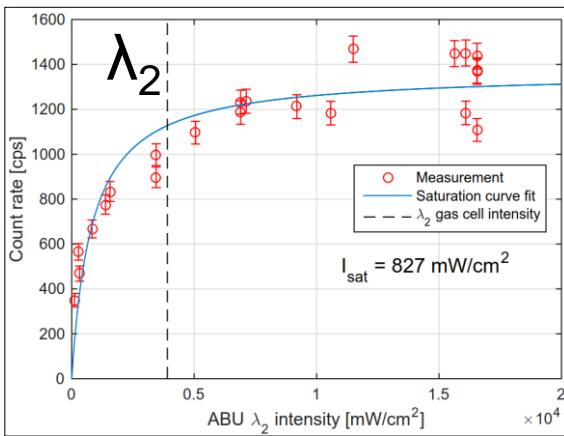
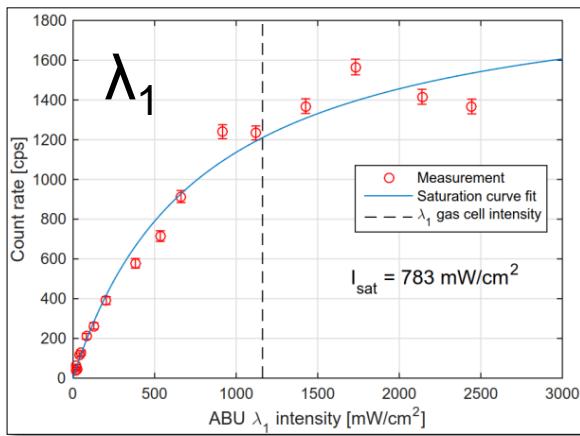
Characterization of the ionization scheme

- From gas cell: mass separated beam at A = 232 u
- ABU confirmation in vacuum

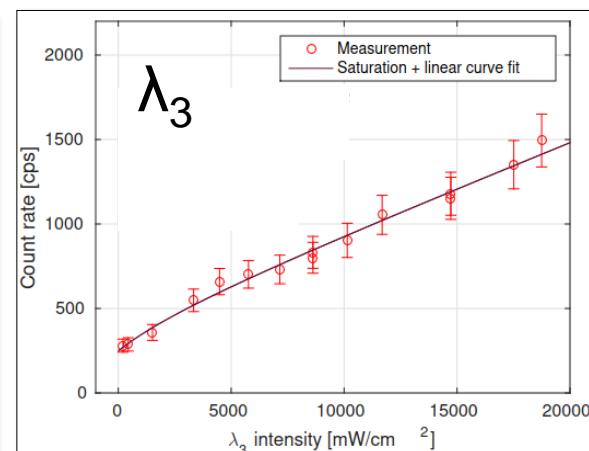
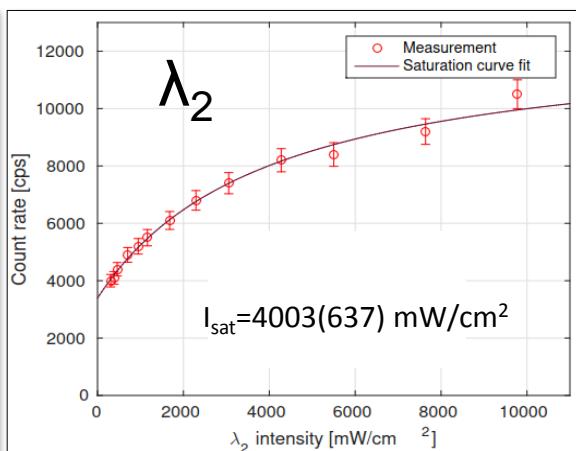
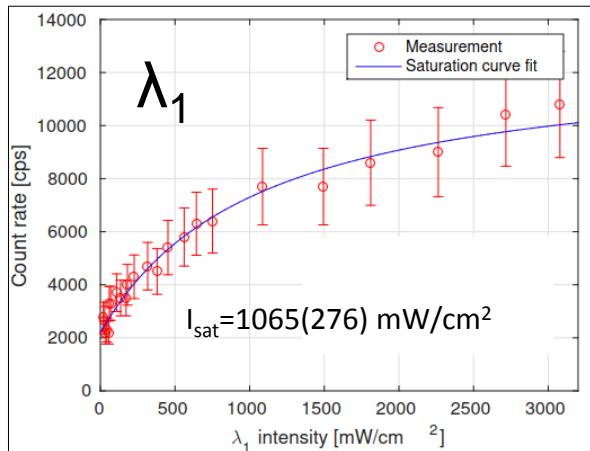


Saturation scans in-vacuum / in-gas cell

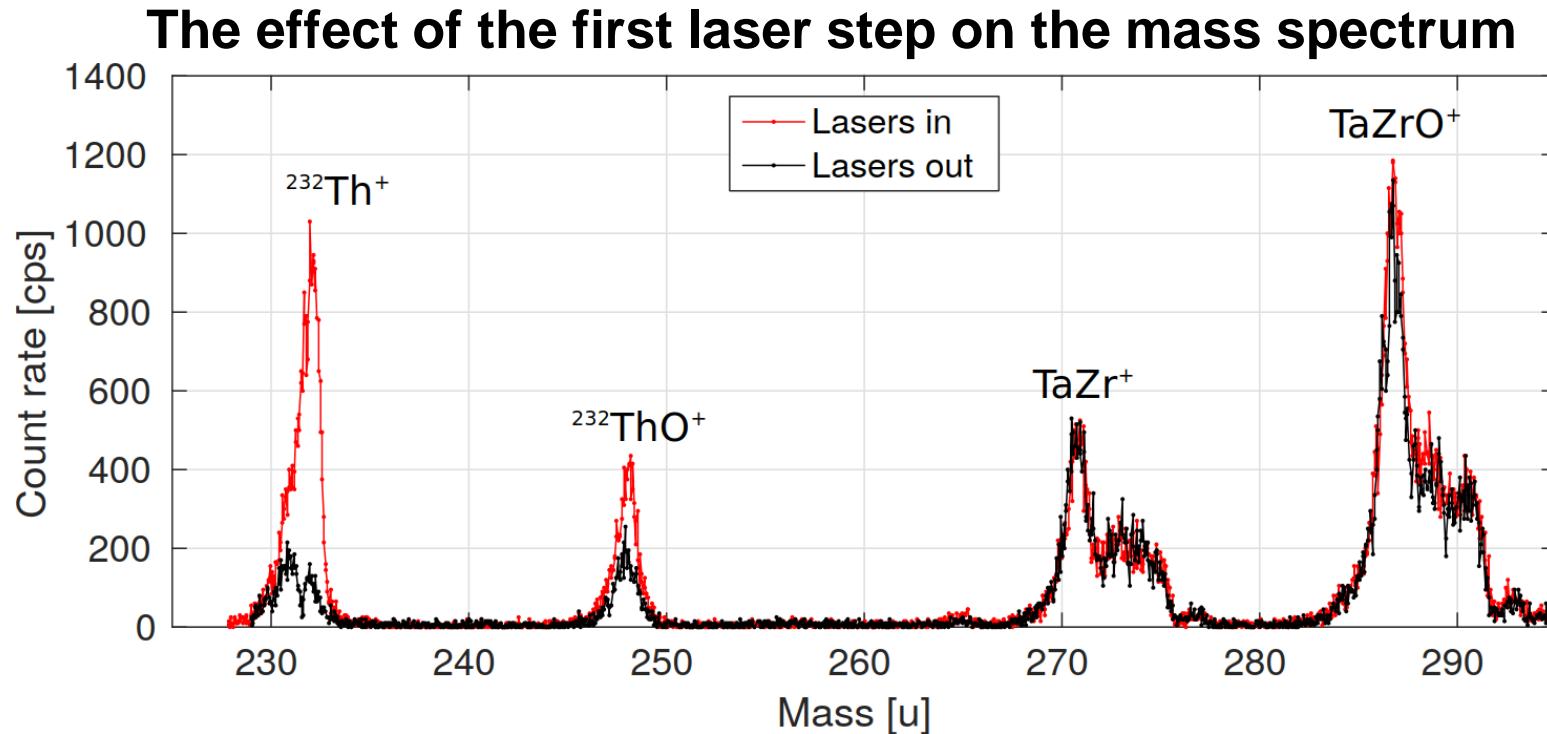
Vacuum (atomic beam unit)



Gas cell



Mass separator scans (Th with Zr coating)

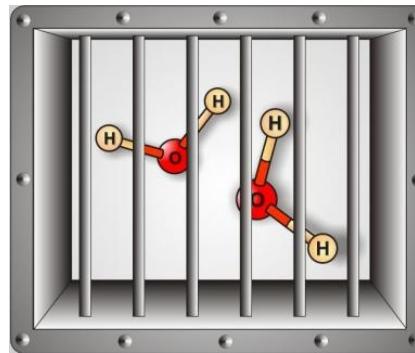


- Yields of FC amounts were possible
→ Plenty of Th ion flux for CLS measurement!
- Strong impurity background at high temperatures
→ High electric potential immediately after gas cell
→ Collision-induced dissociation of molecules

Time profile studies and molecular formation

- Molecular formation; $\tau = 1/k[M]$
- $k = 6 \times 10^{-10} \text{ cm}^3/\text{s}$
- Dynamic processes:

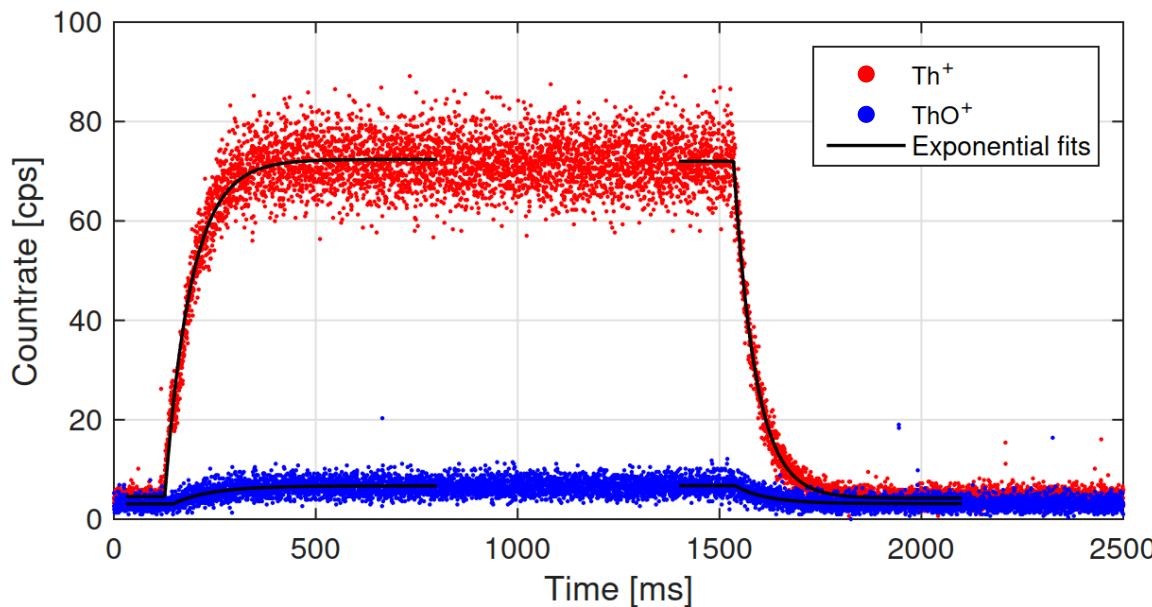
$$y(t) = \begin{cases} A_0 \pm A \left(1 - e^{-\frac{t-t_0}{\tau}}\right) & \text{if } t \geq t_0 \\ A_0 & \text{if } t < t_0 \end{cases}$$



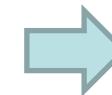
$$dn/dt = -kn[M]$$

$$\tau = 1/k[M]$$

Temporal behavior probed by mechanically chopping laser beam



$$\tau_{fall}(\text{Th}^+) = 55\text{ms}$$

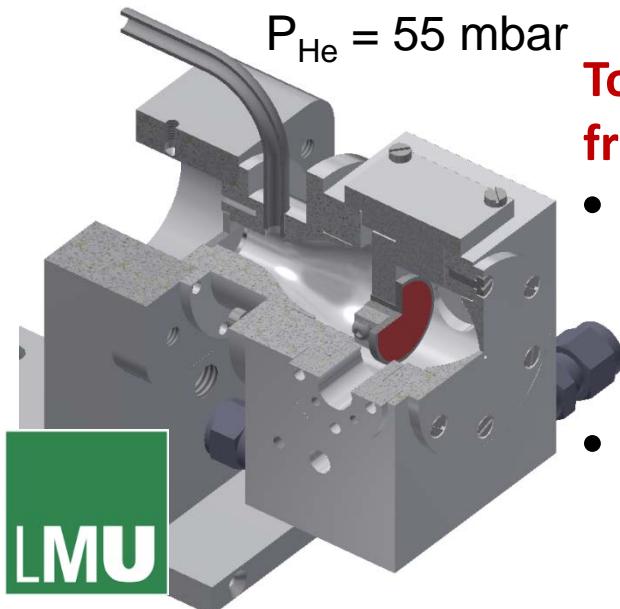


Effective volume:
 $\sim 30 \text{ cm}^3$

Th-229 recoils from U-233 source

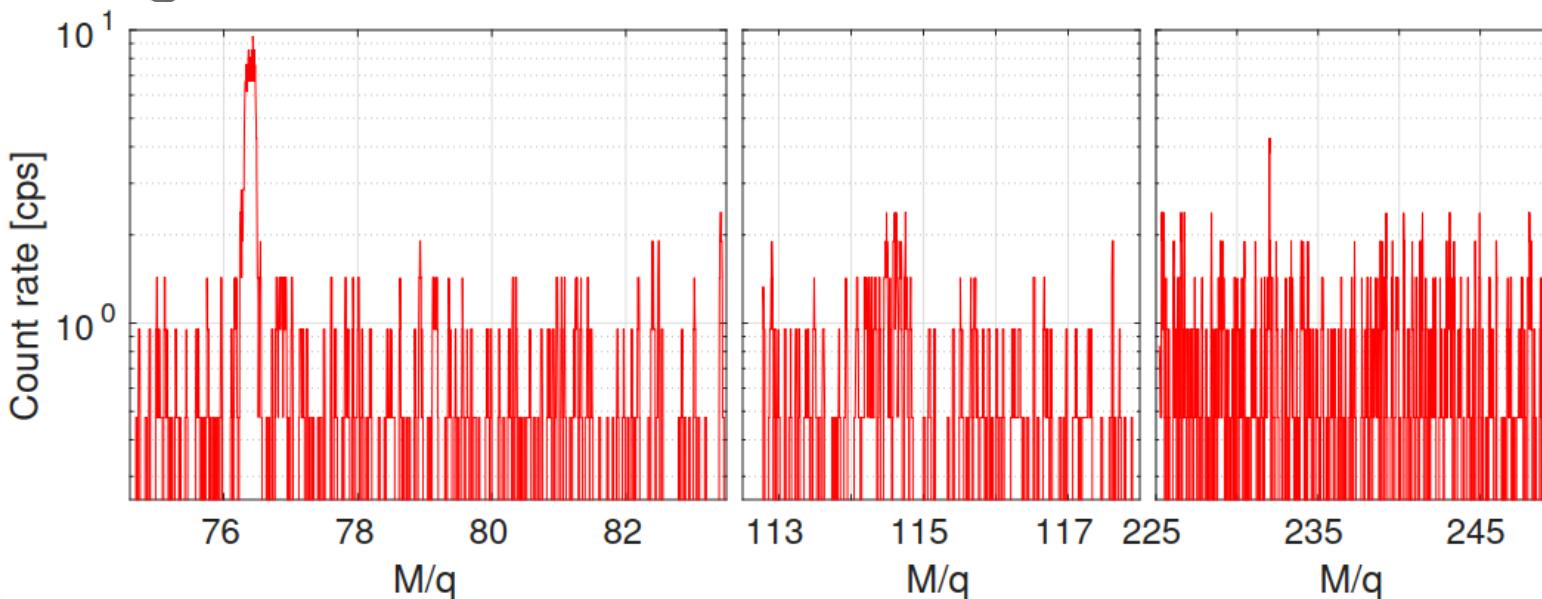
$P_{He} = 55$ mbar

L. v.d.Wense, et. al., Eur. Phys. J. A (2015) 51: 29



To characterize the extraction of Th-229 alpha recoils from a U233 for a CLS measurement

- 230 kBq U-233 source for Th-229 alpha recoils
 - About 10^4 recoils/sec
 - Estimation of 2% fraction to the isomeric state
V. Barci et al., Phys Rev C 68 (2003) 034329.
- Max: ~30 cps of Th³⁺, few cps Th²⁺, no Th¹⁺,
← Identification: mass peak position
(accurate calibration with Xe calibration)



Th-229 recoils from U-233 source

- **Poor efficiency (<1%) !**
 - ← Gas purity – OK
 - ← Recombination with free electrons – very small
 - ← Diffusion losses to the walls or back to the source – likely
 - ← Ion beam transmission (SPIG)
 - to be investigated (Ra-223 Source)

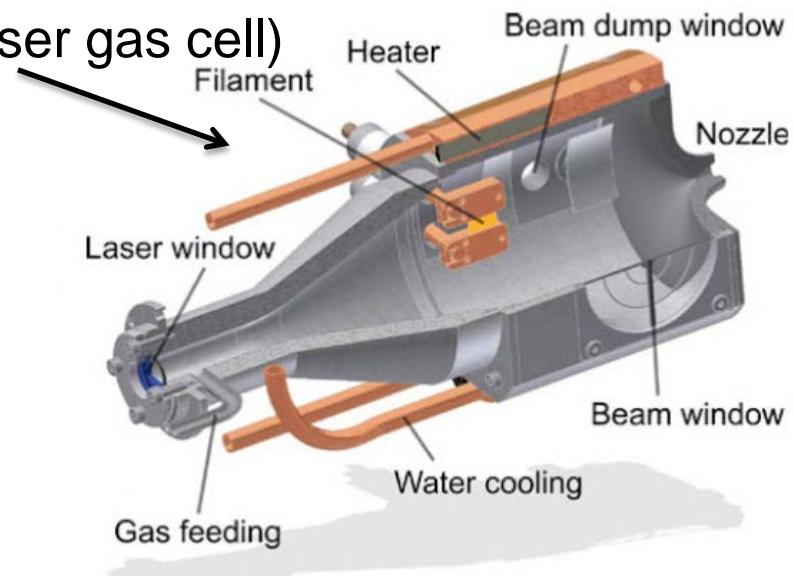


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 - ← Ion beam transmission (SPIG)
 - to be investigated (Ra-223 Source)

- **Solution?**

- Stronger source activity
- Smoother gas flow (HIGISOL laser gas cell)
- Guidance of ions with RF and DC fields in the gas cell

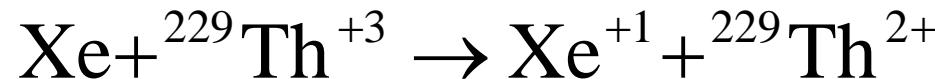


Charge state control of Th

Th IPs (eV)

1+	6.31 eV
2+	11.5 eV
3+	18.3 eV
4+	28.8 eV

Charge state manipulation
by adding Xe/Kr to the helium gas



	He	Ne	Ar	Kr	Xe	O ₂	H ₂ O	N ₂	CO ₂	C ₂ H ₆	C ₃ H ₈	C ₁₀ H ₂₂
IP (eV)	24.6	21.6	15.8	14	12.1	12.1	12.6	15.6	13.8	11.5	10.9	9.65

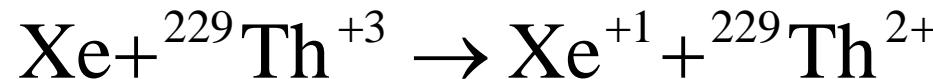


Charge state control of Th

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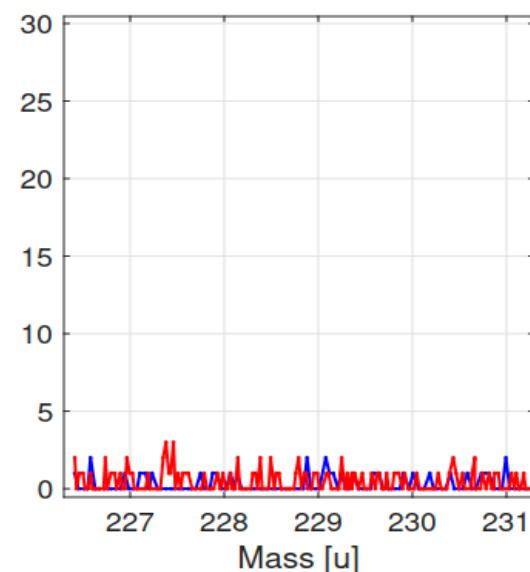
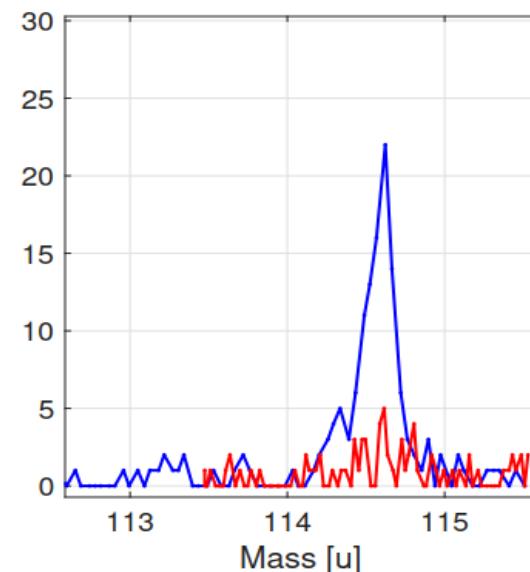
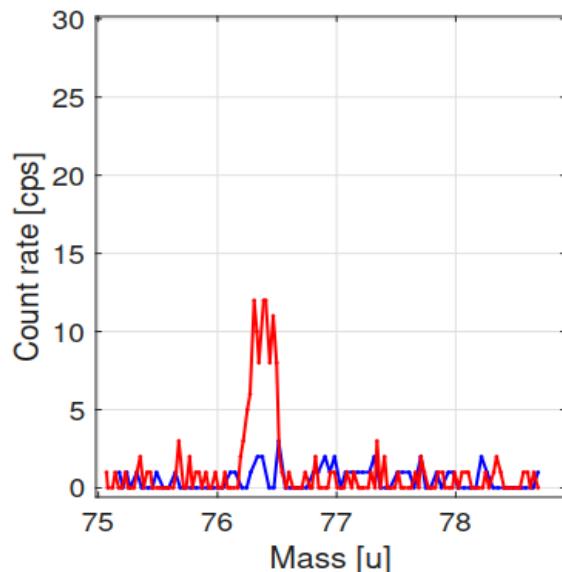
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	He	Ne	Ar	Kr	Xe	O ₂	H ₂ O	N ₂	CO ₂	C ₂ H ₆	C ₃ H ₈	C ₁₀ H ₂₂
IP (eV)	24.6	21.6	15.8	14	12.1	12.1	12.6	15.6	13.8	11.5	10.9	9.65

— With Xe in He — No Xe in He



Summary and Outlook

- New program of high resolution laser spectroscopy on actinide elements
- Sample preparation using ng of material in "actinide sandwiches"
- The gas cell and laser ionization scheme for thorium has been fully characterized using ^{232}Th samples from Vienna.
- ^{233}U from Munich has been characterized the extraction from the gas cell
- ^{229}Th samples from Vienna have arrived. They will be tested and beams delivered to the collinear laser spectroscopy station for HFS and IS measurements.
- Plans to move to on-line production $^{232}\text{Th}(\text{p},\text{p}3\text{n})^{229\text{m}}\text{Th}$ in 2017.
- Collinear laser spectroscopy will be performed on Th^+ , Th^{2+} , (Th^{3+}) for the determination of the ground state and possibly isomeric state HFS when the $^{229}\text{Th}^{n+}$ yield is improved.
- Investigation of 75 eV isomer of U-235 isomer from a ^{239}Pu source as an alpha recoils.



Thanks to all collaborators & for your attention



nuClock

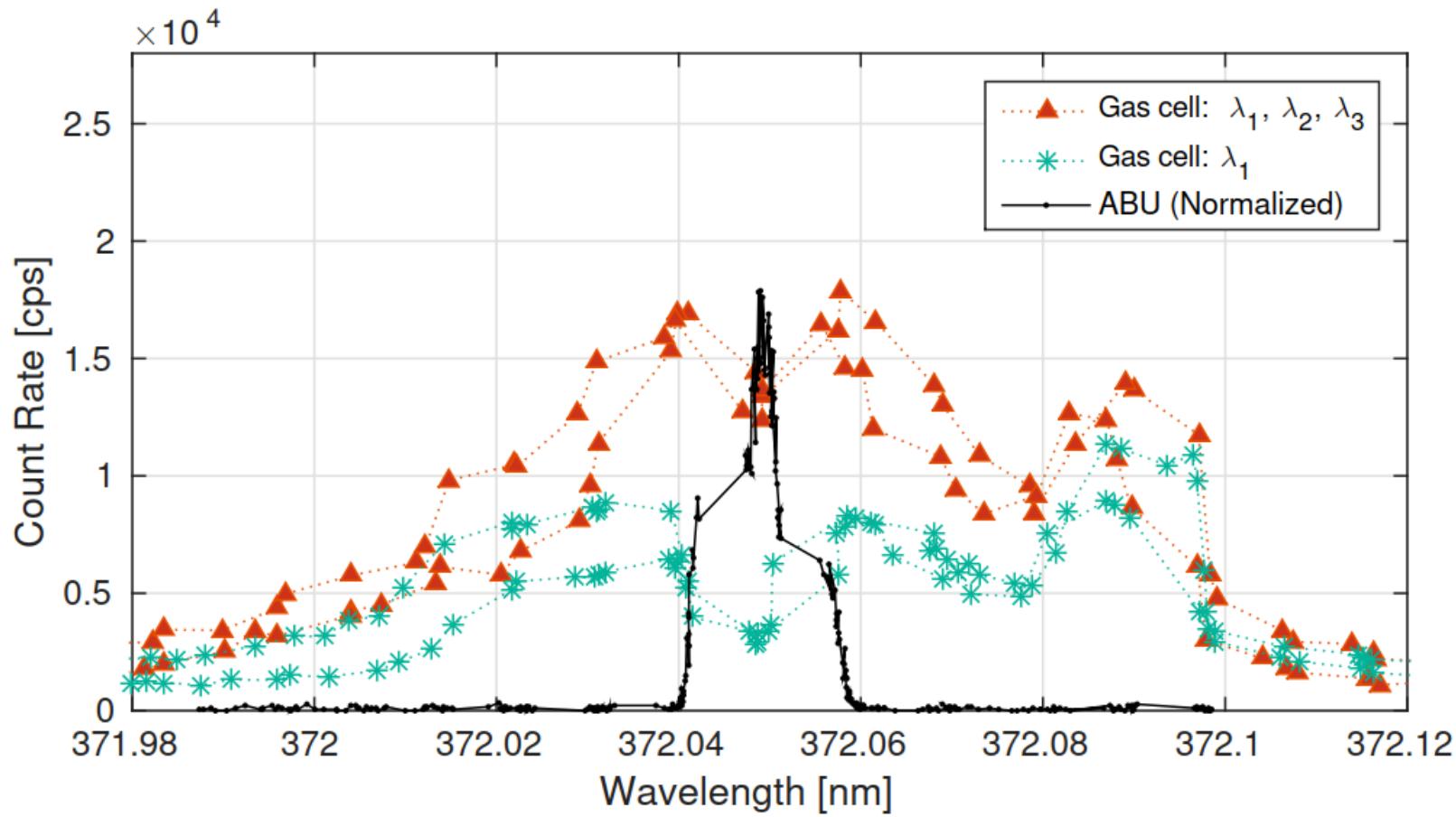


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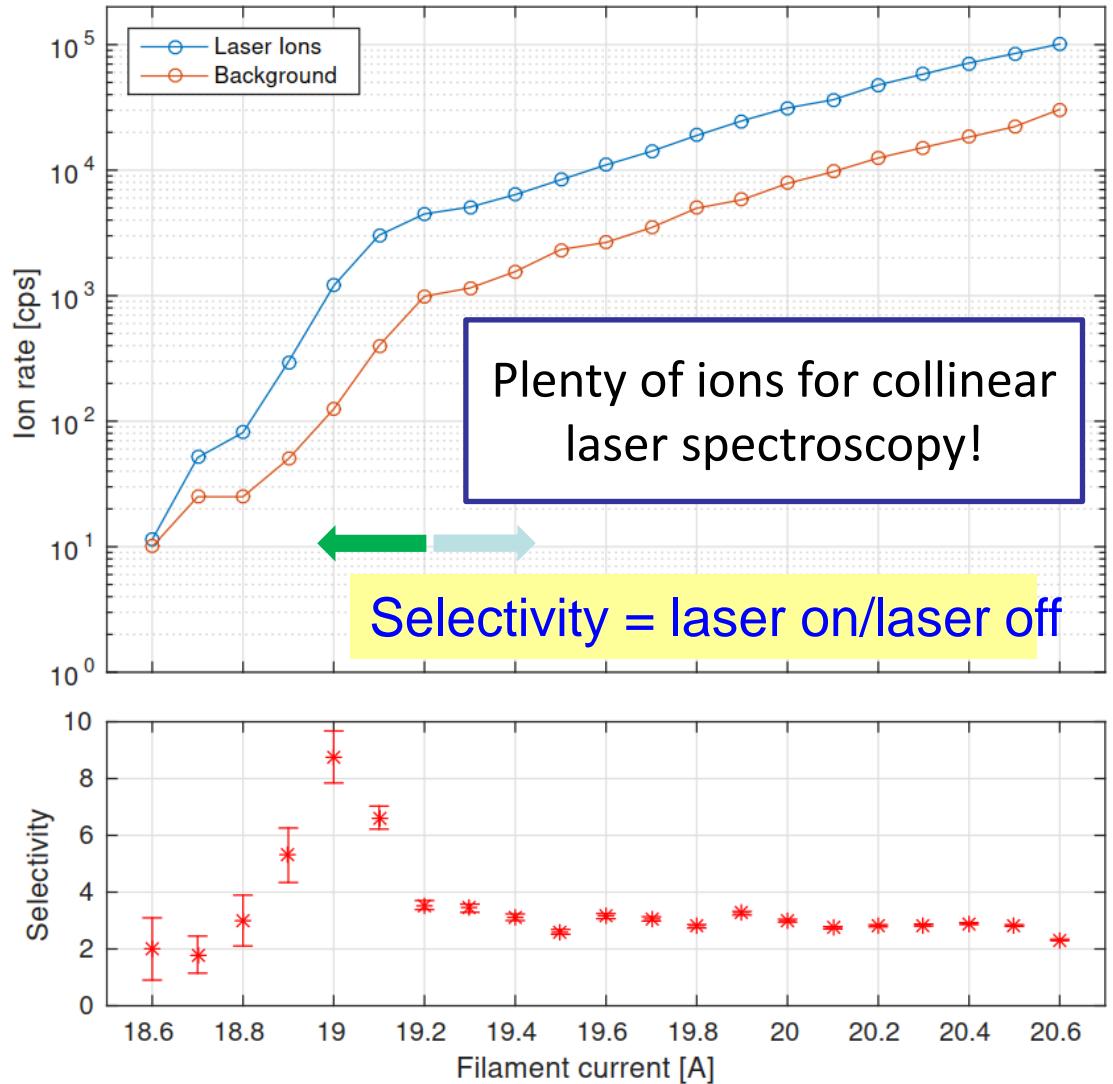
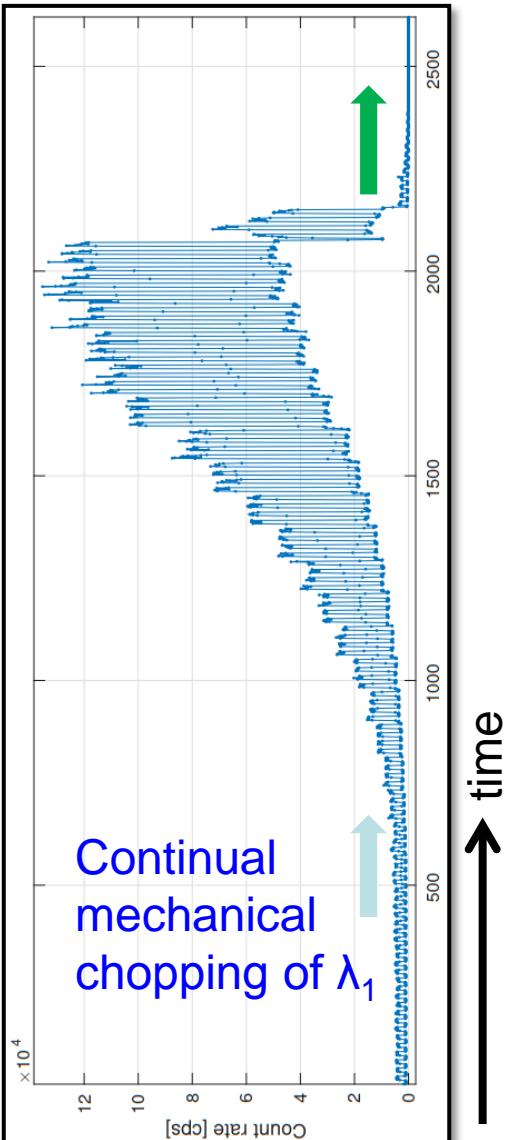
MANCHESTER
1824
The University of Manchester



(Th) First step scan in gas cell



(Th) Selectivity as a function of temperature



(Th) High temperature, chopping first step

