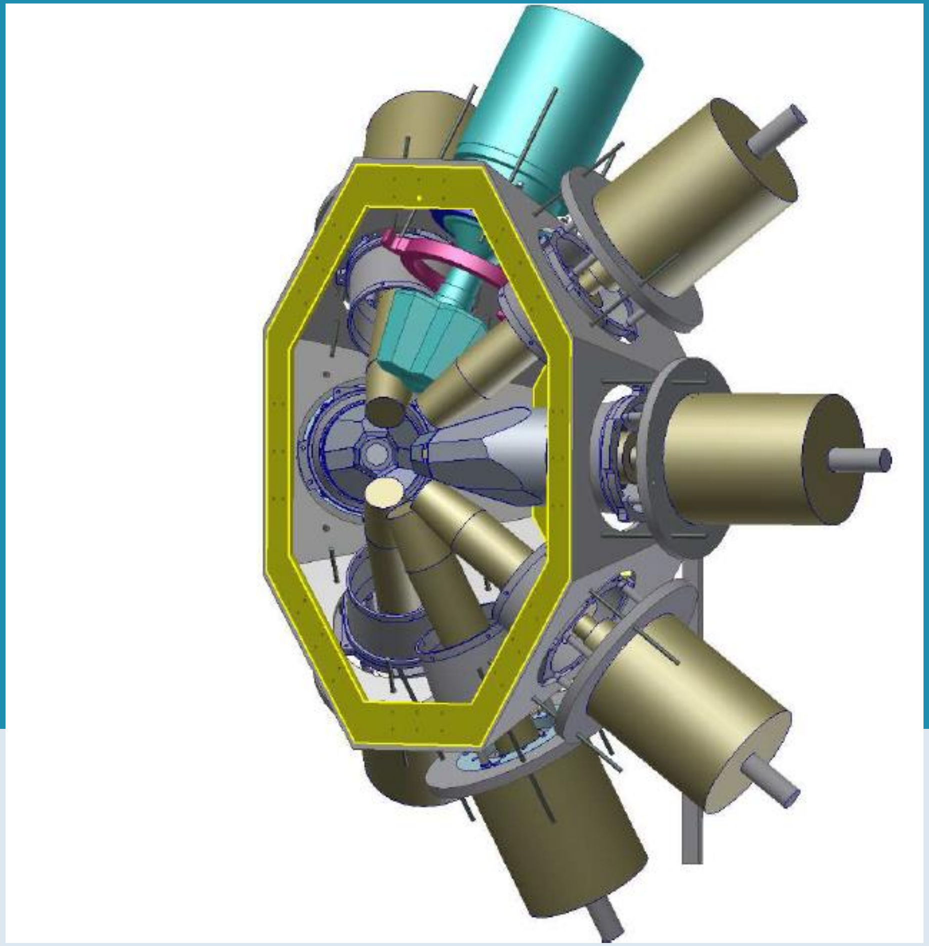
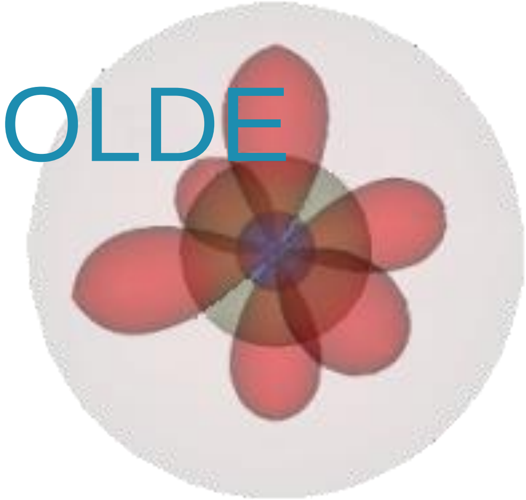
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

Absolute charge radii of radioactive isotopes measured by muonic x-ray spectroscopy at PSI



Proposal INTC-P-552 – 24 June 2020
Spokesperson: T.E. Cocolios – thomas.elias.cocolios@cern.ch
Contact person: C. Duchemin – charlotte.duchemin@cern.ch

Atomic isotope shifts as measured in ISOLDE



∂v, A and v are experimental observables,
while $\partial\langle r^2\rangle$ is the nuclear parameter of interest.

$$\delta\nu^{AA'} = \frac{A' - A}{AA'}\left(m_e\nu + M_{SMS}\right) + F\delta\langle r^2\rangle^{AA'}$$

Mass shift

From one isotope to the next, the reduced mass of the nucleus + electron system varies, giving rise to a mass shift, scaling with A^{-2}

Field shift

From one nuclear state to the next, the charge distribution within the nucleus may vary, perturbing electron orbitals with a non-vanishing overlap with the nucleus

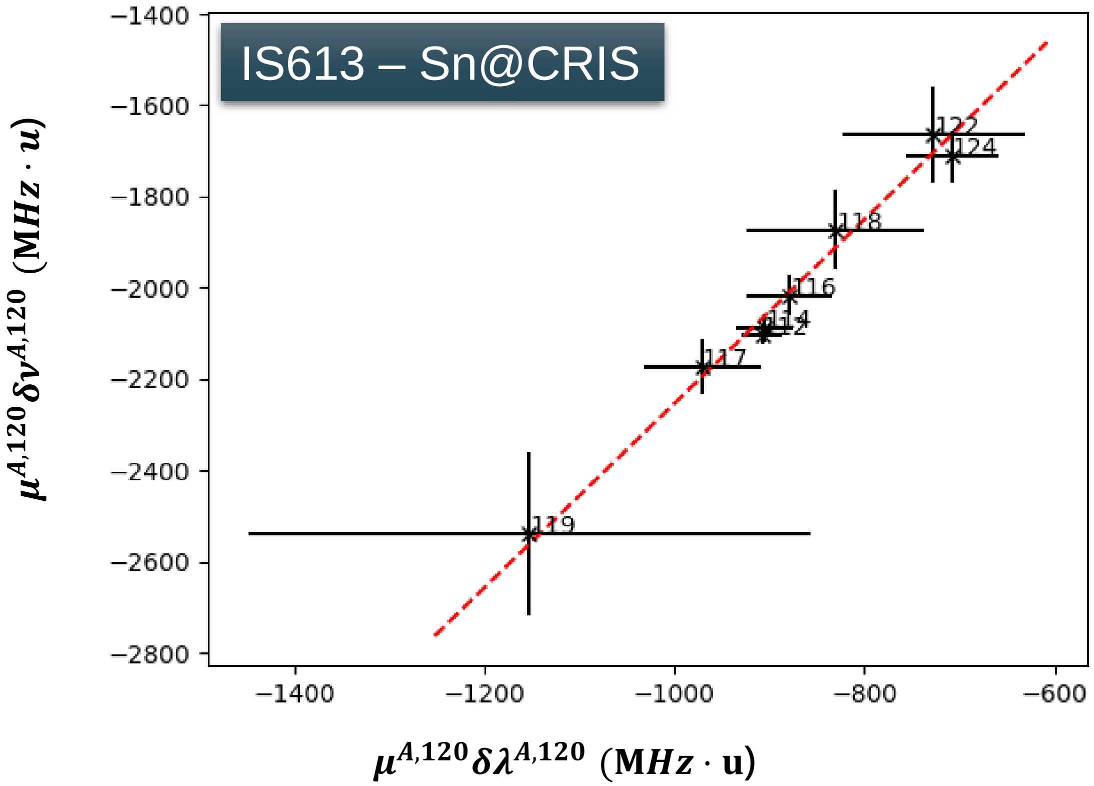
M & *F* are empirical parameters dependent on the transition. Their exact determination from first principles is only possible for systems up to 3 electrons.

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

Experimental approach: King plot

$$\mu_{AA'} = \frac{AA'}{A'-A}$$

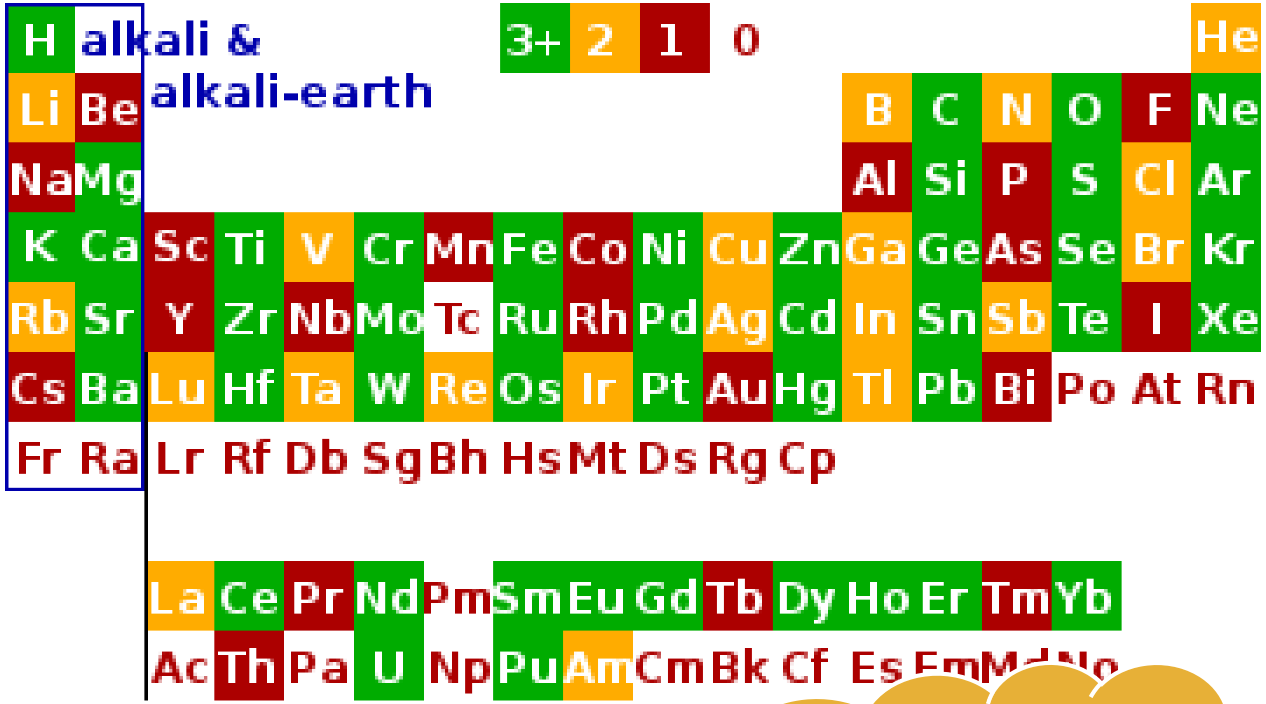
$$\mu_{AA'}\delta\nu^{AA'} = M + F\mu_{AA'}\delta\langle r^2\rangle^{AA'}$$



- The modified isotope shifts are linearly proportional to the modified changes in the charge radii.
- The y-intercept and the slope give direct access to the atomic parameters.
- You need 2 points to draw a line, and a reference to determine a 'change' ➔ 3 absolute radii

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

Measuring 3 absolute charge radii



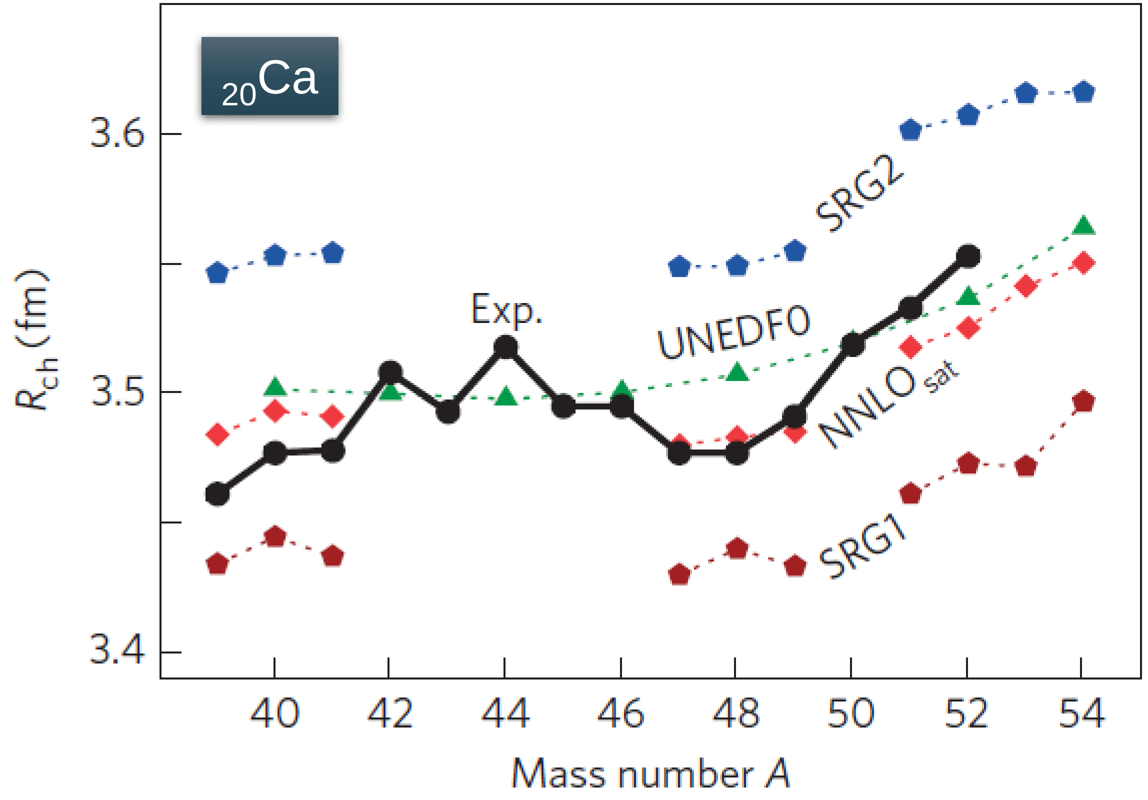
- There are **no odd-*Z* element with 3 stable isotopes** for the standard techniques (e.g. muonic x rays, electron scattering)
- Applying those techniques to **radioactive isotopes** is currently extremely limited:
  - ❑ **SCRIT@RIKEN**: electron scattering from photo-fission products;
  - ✓ **muX@PSI**: muonic x-ray spectroscopy of long-lived radioisotopes;
  - ❑ Absolute radii are best extracted from the combined analysis of both techniques.

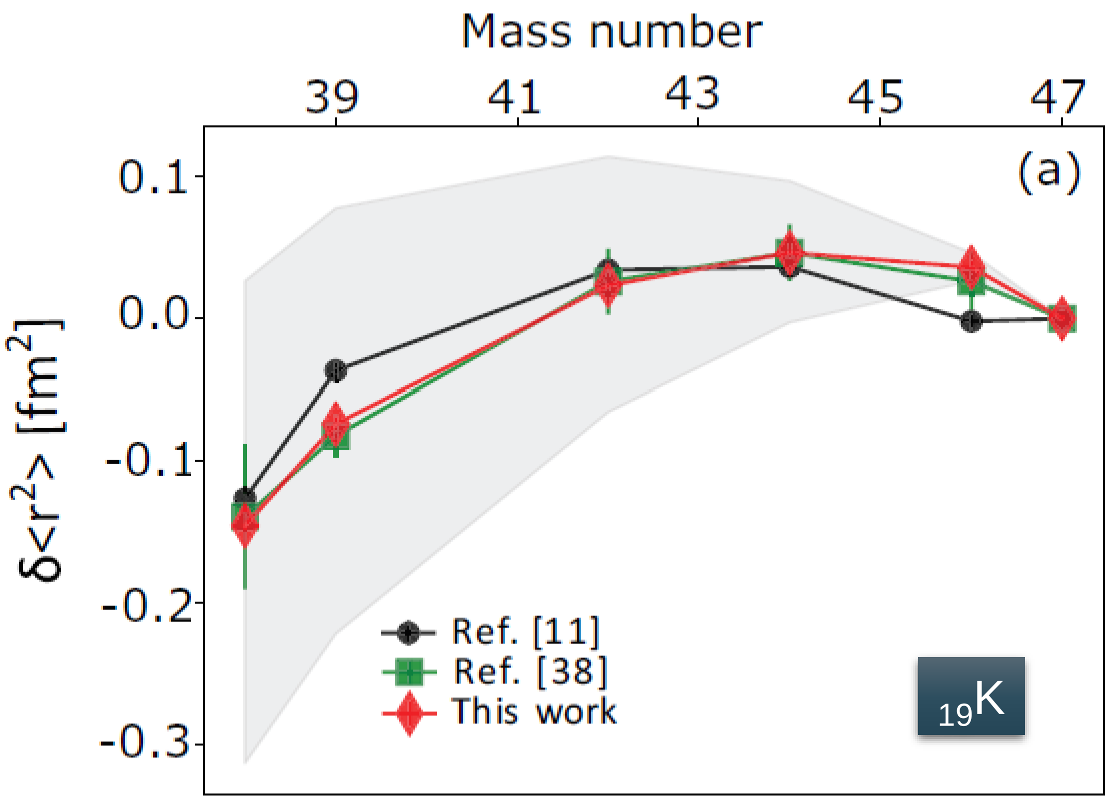
Combining the available ISOLDE isotopes with electron scattering would open many new possibilities

K. Tsukada et al, First elastic electron scattering fr ... *PRL* ***118*** *(2017) 262501.*
E. Rapisarda et al, Measurement of the quadrupole moment o ... *^{187}Re from the hyperfine structure of muonic X rays, Physical Review C* ***101*** *(2020) 054313.*

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

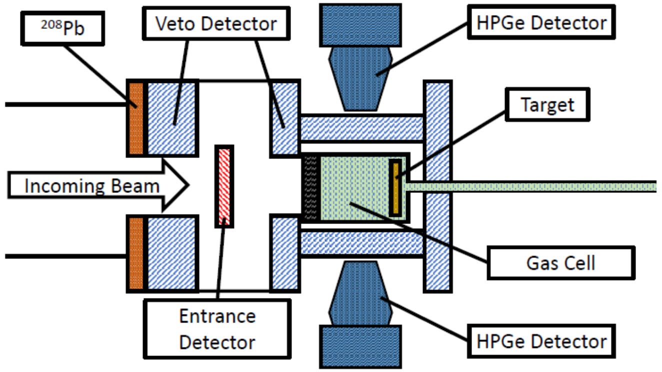
Impact on the nuclear physics interpretation

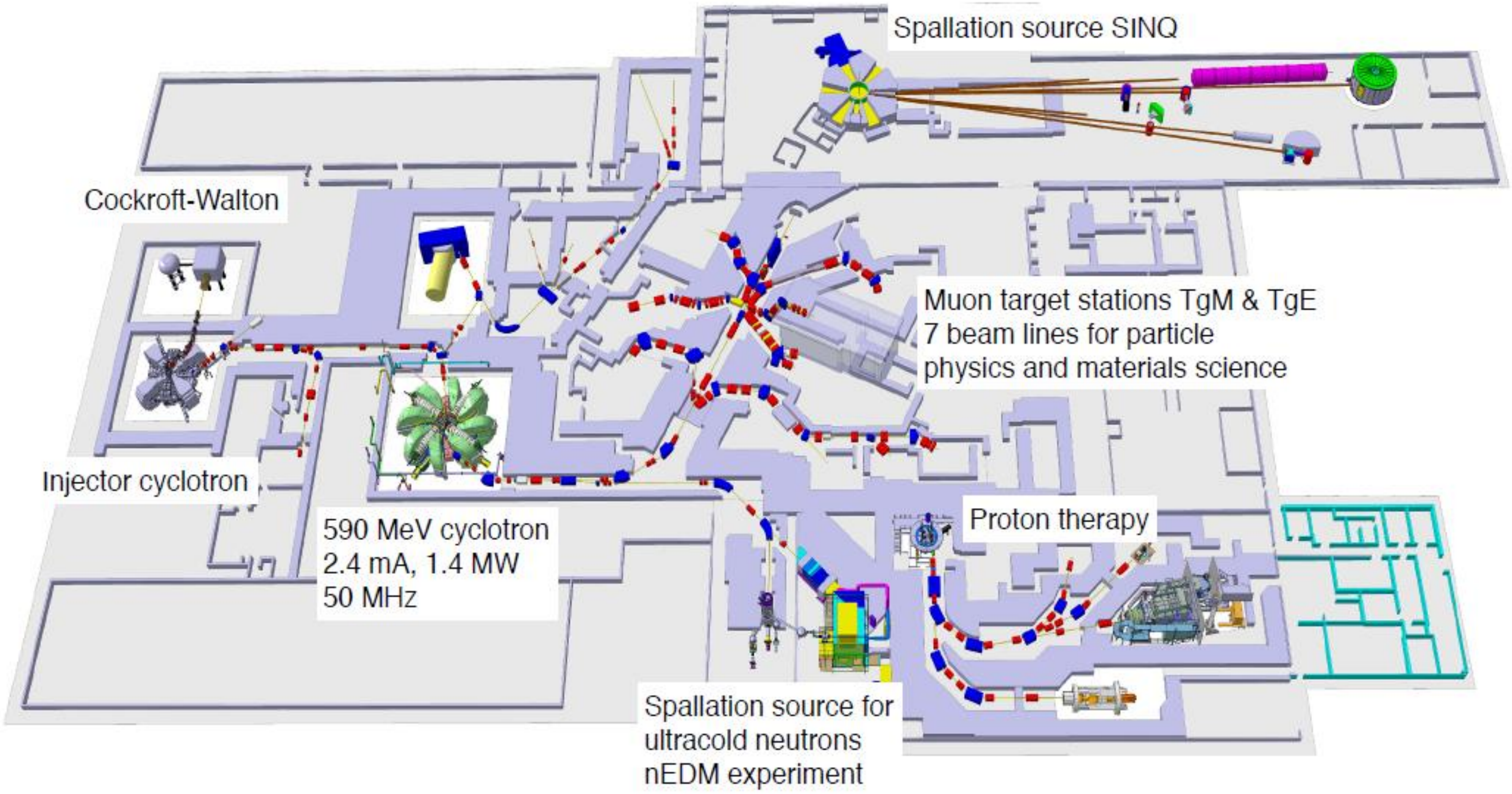




*R.F. Garcia Ruiz et al, …radii of neutron-rich calcium isotopes, Nature Physics **12** (2016) 594-598.*
*A. Koszorus et al, … radii of potassium isotopes, PRC **100** (2019) 034304 & work in preparation (IS620)*

μ-atomic spectroscopy with muX@PSI

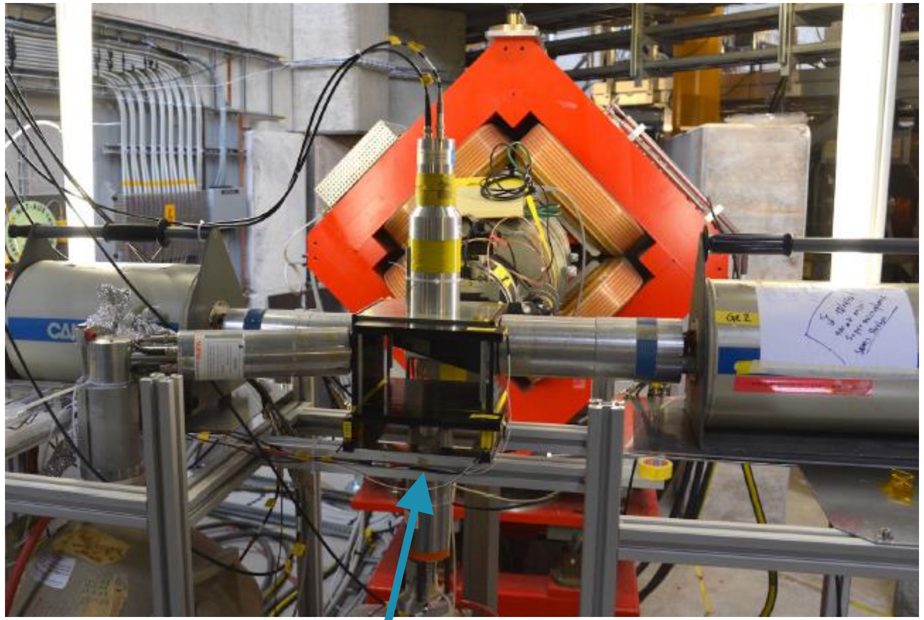


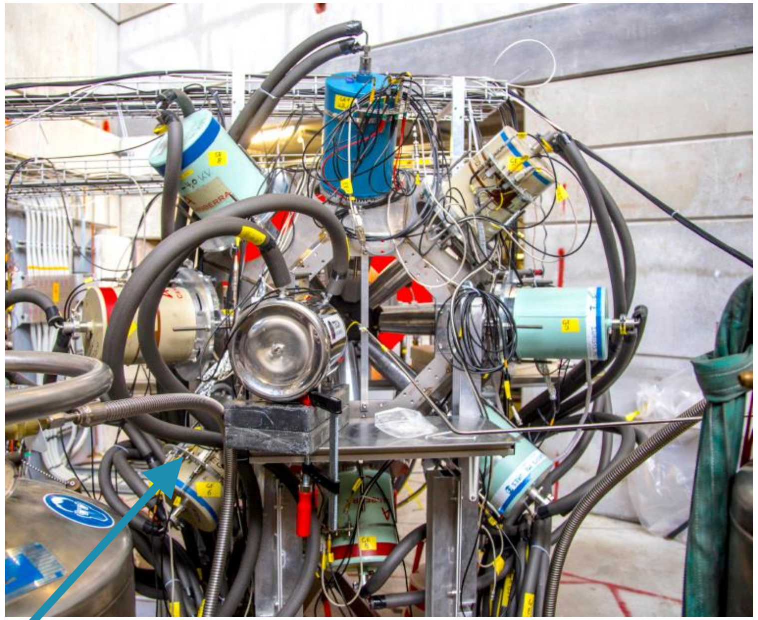


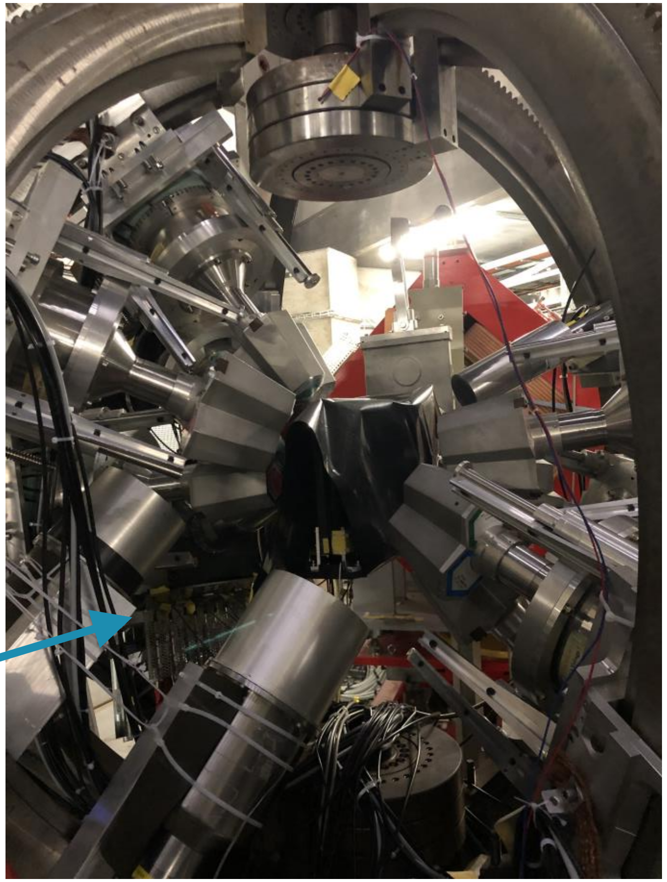
- A muon beam is delivered from the HIPA facility at PSI to the πE1 experimental area.
- The muon enters the experimental setup and is captured in a hydrogen gas cell, transferred to deuteron and eventually to the isotope of interest.
- The captured muon cascades down towards the ground state and the emitted x rays are monitored with the Ge detector array

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

µ-atomic spectroscopy with muX@PSI





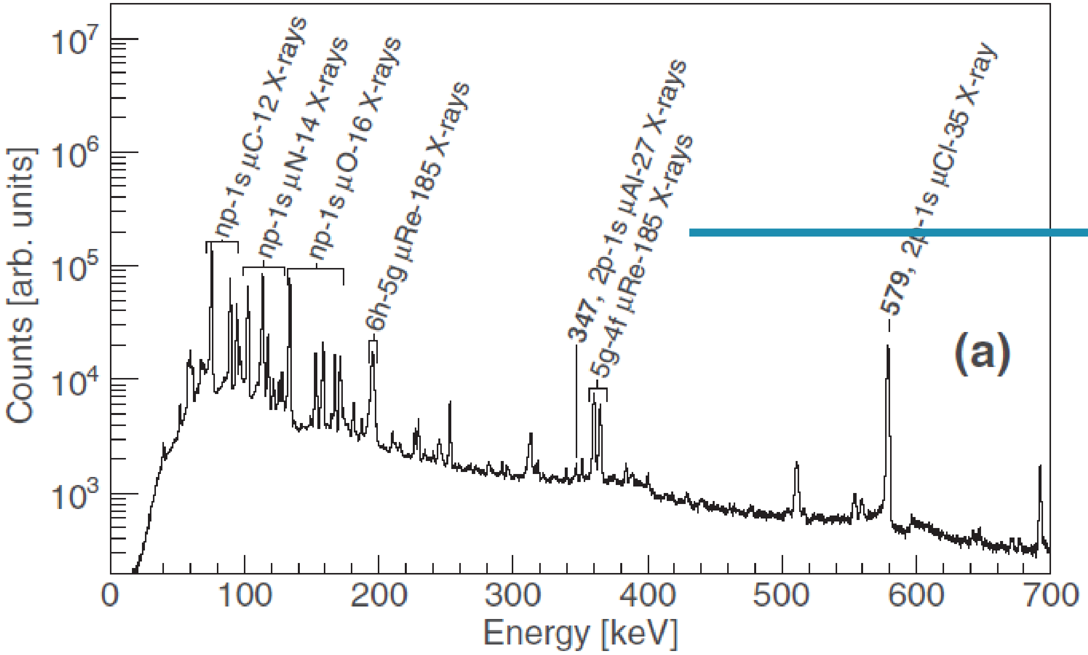


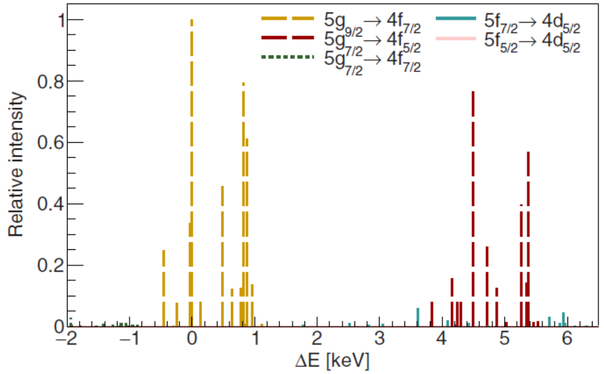
- 2016: 2 HPGe (Leuven & PSI)
- 2017/18: the Death Star Array: 1 MiniBall Cluster (Leuven)
- + detectors from PSI and the gamma loan pool (Orsay)
- 2019: MiniBall@PSI campaign
- 2021 and beyond: back to the Death Star Array but seeking better detectors

A. Adamczak et al, Nuclear structure with radioactive muonic atoms, EPJ WC ***193*** *(2018) 04014.*
A. Skawran et al, Towards nuclear structure with radioactive muonic atoms, Il Nuovo Cimento ***42 C*** *(2019) 125.*

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

First measurement: ^{185,187}Re





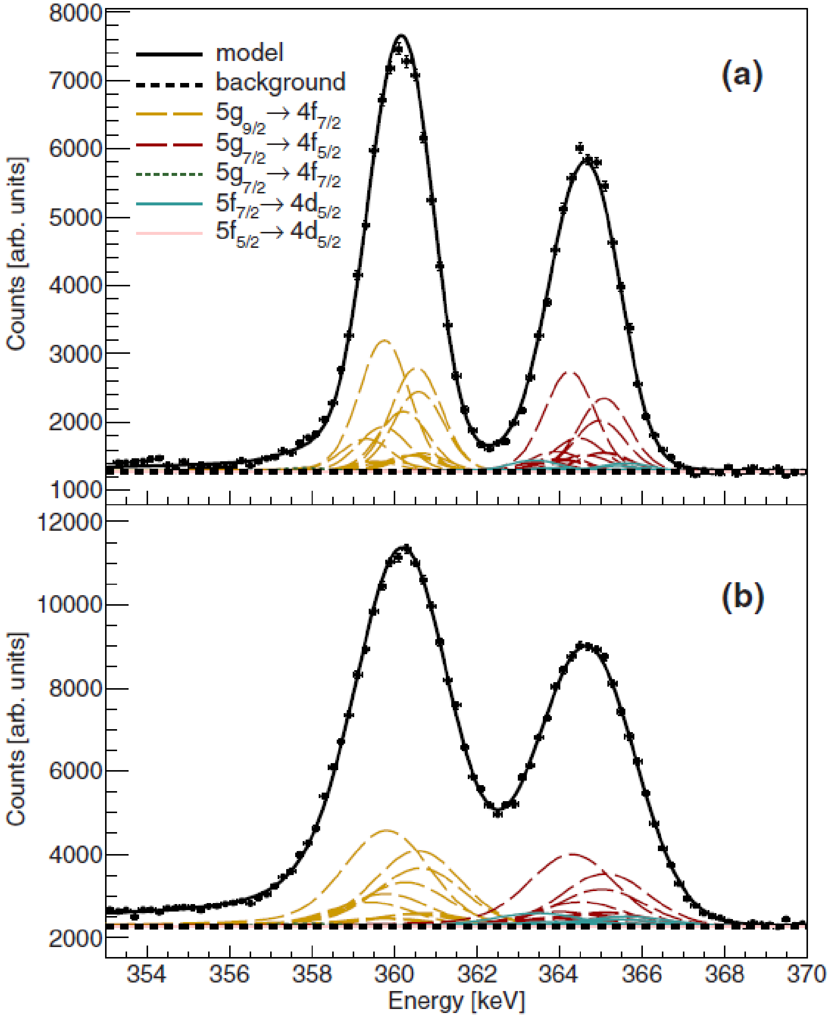


FIG. 7. Prompt γ-ray spectrum of ^{185}Re obtained by GeR (a) and GeL (b) showing the $5g \rightarrow 4f$ hf complex. The full black line shows the best fit to the data. The lines predicted by the hfs formalism are shown below the spectra.

*E. Rapisarda et al, Measurement of the quadrupole moment of ^{185}Re and ^{187}Re from the hyperfine structure of muonic X rays, Physical Review C **101** (2020) 054313.*

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

Where to with muX?

- Research in support of laser spectroscopy activities across the nuclear landscape: hoping to reach from $_{19}K$ to $_{89}Ac$ and beyond…
- Producing and isolating a pure target is one of the challenges that we face.
- Choosing where to begin is another!

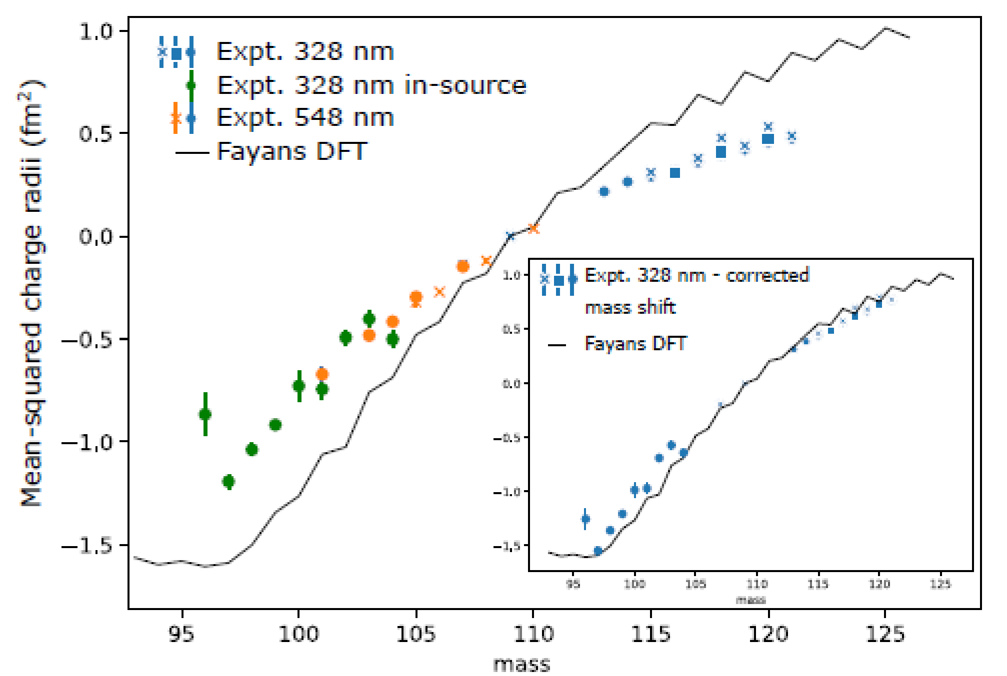
- $_{47}Ag$
- $_{57}La$
- $_{65}Tb$

- TAC: purity of the collected sample?
  - 90% elemental purity
  - 99% isotopic purity

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

$_{47}$Ag

- In the vicinity of Z=50 and spanning from N=50 to N=82;
- Previous investigations already reached down to N=50 (LISOL);
- New work under analysis from Jyvaskyla from N=49 to N=74;
- New proposal at ISOLDE to push to N=82;
- At term, many groups aim to study the elusive case of the high-spin isomers in the self-conjugate ^{94}Ag.



R. Ferrer et al, … radii of N=50-54Ag, PLB **728** (2014) 191-197.
R.P. de Groote et al, [CRIS] of silver…, INTC-P-551.
I. Mukha et al, … two-proton radioactivity of ^{94}Ag, Nature **439** (2006) 298-302.

$_{47}$Ag

- $_{47}$Ag features 2 stable isotopes: ^{107,109}Ag with spin ½
- It also has a long-lived isotope: ^{108m}Ag with spin 6 and $T_{1/2}$=438 years
- Production: ^{107}Ag(n,γ) followed by mass separation
- ISOLDE has experience in the production of $_{47}$Ag

^{107}Cd β+	^{108}Cd Stable	^{109}Cd e- capture	^{110}Cd Stable	^{111}Cd Stable
^{106}Ag β+	^{107}Ag Stable	^{108}Ag β-	^{109}Ag Stable	^{110}Ag β-
^{105}Pd Stable	^{106}Pd Stable	^{107}Pd β-	^{108}Pd Stable	^{109}Pd β-

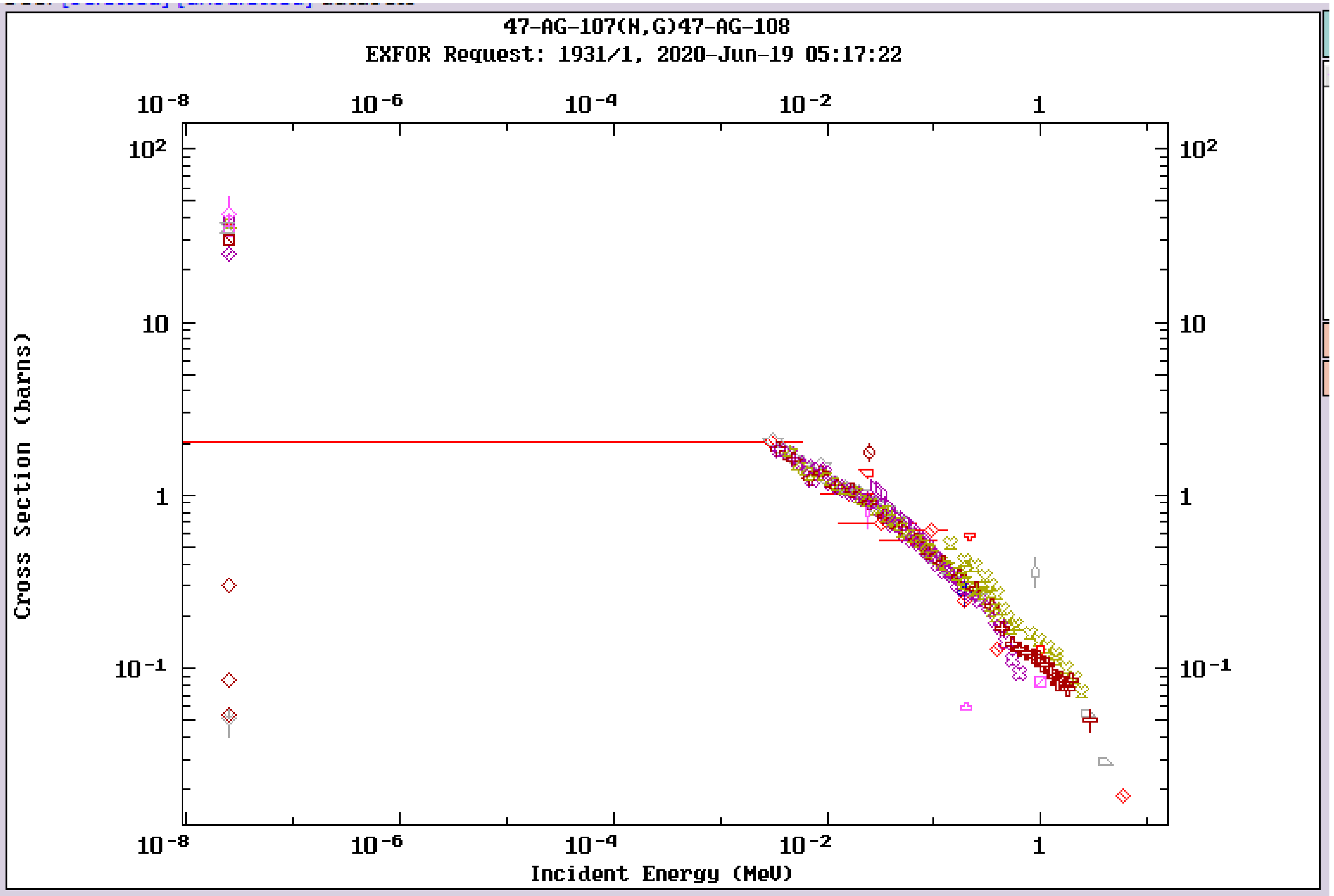
- TAC: Is the isomer produced?
- TAC: Ratio of ^{107}Ag to ^{108m}Ag?

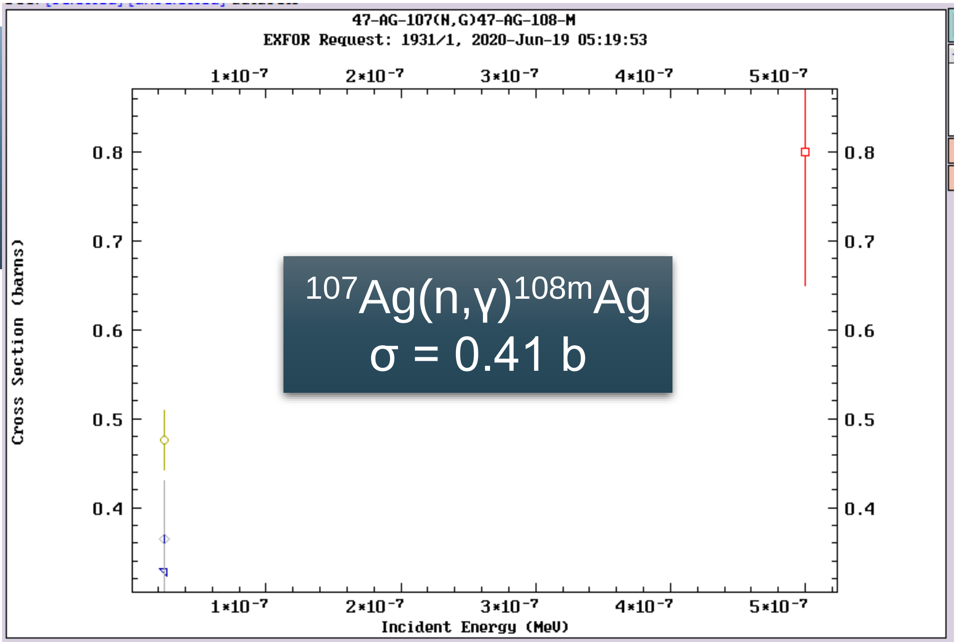
*R. Ferrer et al, … radii of N=50-54Ag, PLB **728** (2014) 191-197.*
R.P. de Groote et al, [CRIS] of silver…, INTC-P-551.
*I. Mukha et al, … two-proton radioactivity of ^{94}Ag, Nature **439** (2006) 298-302.*

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

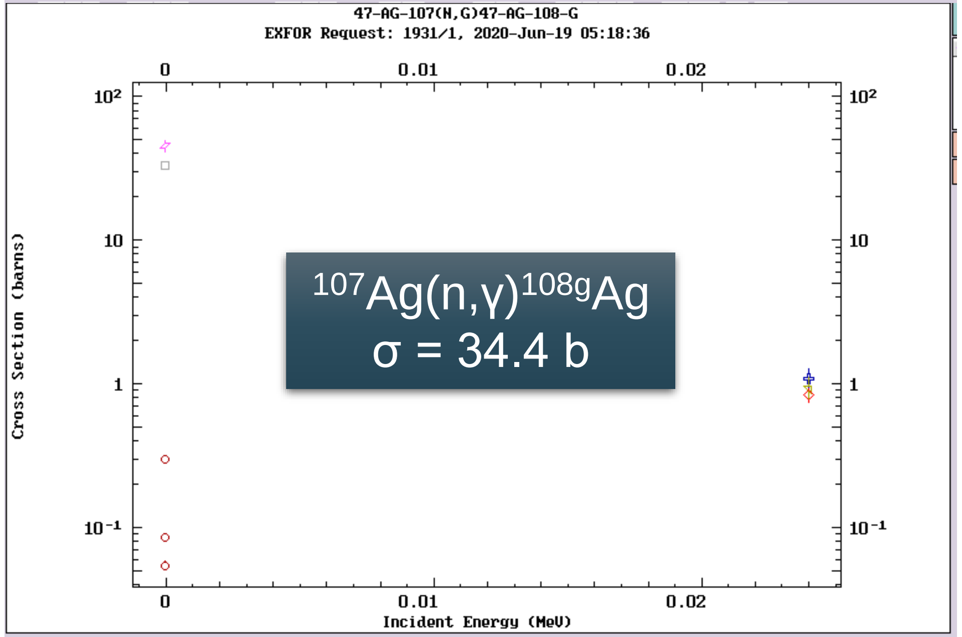
$_{47}$Ag

- TAC: Is the isomer produced?
  - ✓ Yes





^{107}Ag(n,γ)^{108m}Ag
σ = 0.41 b



^{107}Ag(n,γ)^{108g}Ag
σ = 34.4 b

EXFOR database from NNDC: https://www.nndc.bnl.gov/

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

$_{47}Ag$

- TAC: Ratio of ^{107}Ag to ^{108m}Ag?
- TAC: Can the ion source handle the throughput?

- Situation similar to the $^{168}Er(n,\gamma)^{169}Er$ case at MEDICIS
  - Same order of magnitude cross section
  - Enriched material as starting point
  - 2% radioisotope content at the end of irradiation
  - Demonstrated a collection of 17 MBq = 2×10^{13} atoms without RILIS (ε_{ion}~0.2%)

✓1mg of ^{107}Ag can yield 12MBq of ^{108m}Ag, i.e. 2.3 x 10^{17} atoms, after 50 days irradiation + 10 days cooling

✓This represents ~4% fraction of the total sample size

✓RILIS reports a 14% ionization efficiency, resulting in 3 x 10^{16} collected atoms.

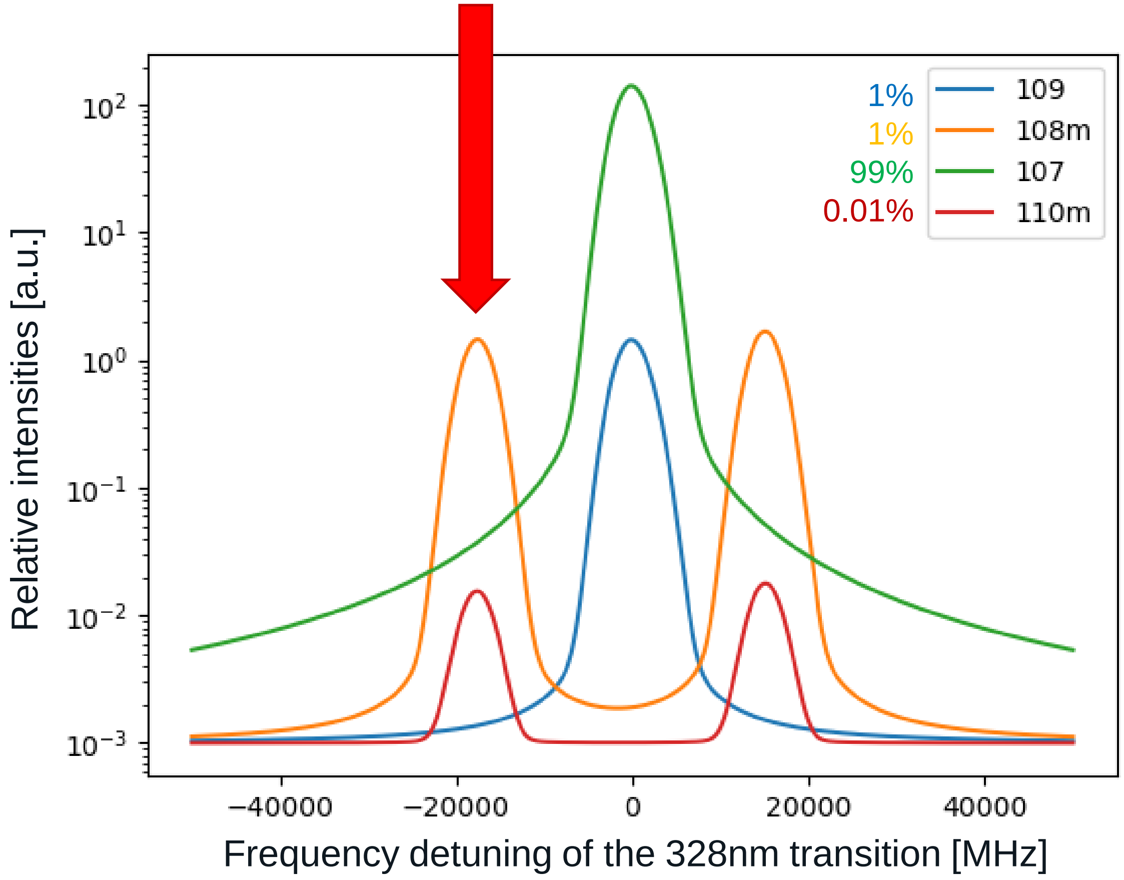
*R. Formento-Cavaier et al, … ^{169}Er production…, NIMB **463** (2020) 468-471.*

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

$_{47}$Ag

✓ TAC: Ratio of ^{107}Ag to ^{108m}Ag?

✓ TAC: Can the ion source handle the throughput?



✓RILIS reports a 14% ionization efficiency, resulting in 3 x 10^{16} collected atoms.

- RILIS in normal mode can already separate Ag isotopes with widely different spin/nuclear structure:
  - If tuned on ^{108m}Ag, both ^{107,109}Ag are naturally suppressed from the source

✓No throughput difficulty

HFS simulation courtesy of R.P. de Groote.

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

$_{57}$La

- Neutron-deficient lanthanides are gaining interest with new facilities such as MARA in Jyvaskyla and S3 at GANIL
- Limited laser spectroscopy work so far to $^{131,135,137\text{-}139}$La, suggesting triaxiality in the neutron-deficient isotopes
- Most-neutron-deficient isotope exhibits proton emission and could reveal new features in charge radii

*H. Iimura et al, … isotope shifts of ^{135}La, ^{137}La, and ^{138}La…, PRC **68** (2003) 054328.*
T.E. Cocolios, Master Thesis, McGill University 2005.
H.A. Schuessler et al, Nuclear moments of … ^{131}La…, Proceedings to LAP2006, p95-96 (2006).

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

$_{57}$La

- TAC: La scheme?
- TAC: What can Mainz do?

^{136}Ce Stable	^{137}Ce β+	^{138}Ce Stable	^{139}Ce e- capture	^{140}Ce Stable
^{135}La β+	^{136}La β+	^{137}La e- capture	^{138}La Stable	^{139}La Stable
^{134}Ba Stable	^{135}Ba Stable	^{136}Ba Stable	^{137}Ba Stable	^{138}Ba Stable

- $_{57}$La contains 2 stable isotopes: ^{138}La and ^{139}La
  - ❖Natural abundance of ^{138}La: 0.00088
  - ➢Has not been studied with μx rays or e^- scattering
  - ❑Commercially available with enrichment to 5%
  - ➢Requires further enrichment to be suitable at muX
- An additional long-lived isotopes is available: ^{137}La with $T_{1/2}$=60,000 years
  - Can be produced in ^{136}Ce(n,γ)^{137}Ce(β^+/EC)^{137}La with the intermediate isotope having $T_{1/2}$=9h, followed with radiochemistry to isolate La from Ce
  - ❑Risks to produce isotopic contamination from ^{138}Ce(n,γ)^{139}Ce(EC)^{139}La where the intermediate isotope has $T_{1/2}$=137.6d
  - ➢Sample purity requires mass measurement for quality control (no γ/stable)

*H. Iimura et al, … isotope shifts of ^{135}La, ^{137}La, and ^{138}La…, PRC **68** (2003) 054328.*
T.E. Cocolios, Master Thesis, McGill University 2005.
H.A. Schuessler et al, Nuclear moments of … ^{131}La…, Proceedings to LAP2006, p95-96 (2006).

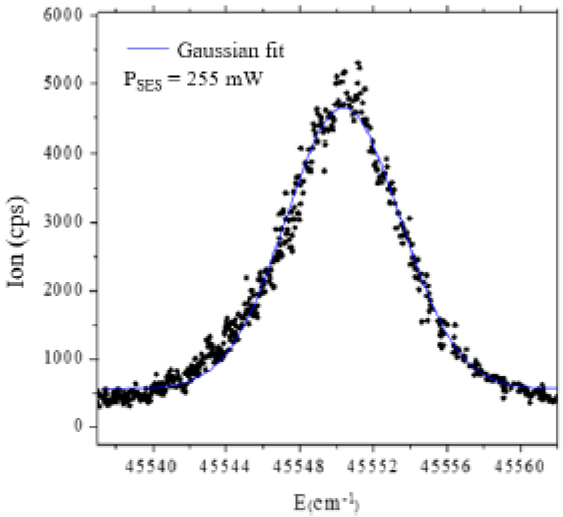
Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN
NUCLEAR AND RADIATION PHYSICS

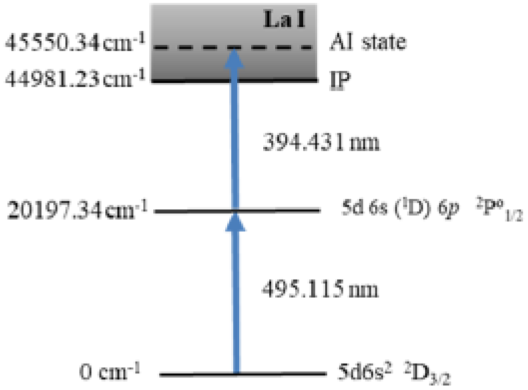
$_{57}$La

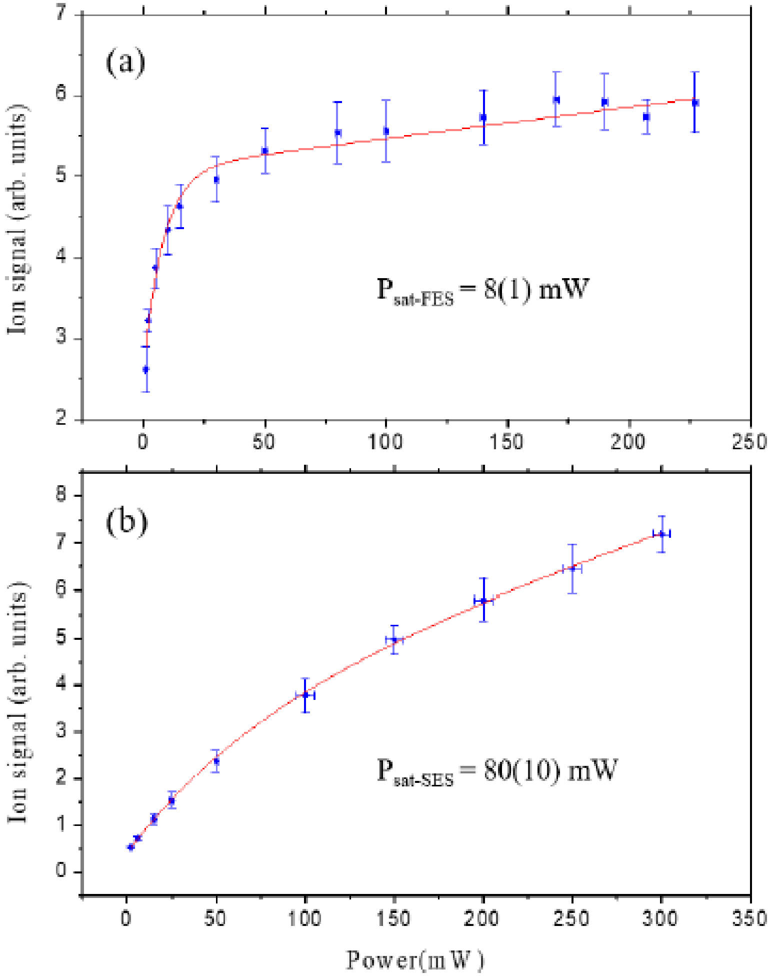
- ✓ TAC: La scheme?
- ✓ TAC: What can Mainz do?

✓ $_{57}$La has been successfully laser ionized at TRIUMF and the scheme is available.

✓ Mainz can perform the characterization of the isotope ratio in the case of ^{137}La.







Maryam Mostamand, Laser developments and study of Rydberg and autoionizing Rydeberg states in Tm, La and At using resonant ionization laser spectroscopy, PhD Thesis, University of Manitoba (2020). *https://mspace.lib.umanitoba.ca/handle/1993/34548*

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy
KU LEUVEN NUCLEAR AND RADIATION PHYSICS

$_{65}$Tb

- $_{65}$Tb is in the middle of the lanthanide series
- It is gaining interest beyond its medical applications
- Neutron-rich isotopes are mid-shell in both protons and neutrons, where many correlations are possible and leading to a rich nuclear structure
- Most-neutron-deficient isotope exhibits proton emission and could reveal new features in charge radii

M. Mougeot et al, … neutron-rich Tb … approaching the doubly mid-shell in Rare Earth nuclei…, INTC-I-217.

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

$_{65}$Tb

✓ TAC: What can Mainz do?

- $_{65}$Tb has only 1 stable isotope: ^{159}Tb
- It however also has 2 long-lived isotopes:
  - ➢ ^{158}Tb, $T_{1/2}$=180 years
  - ✓ Production via ^{158}Gd(d,2n)^{158}Tb
  - ❖ Risks of isotopic contamination with ^{nat}Gd(d,xn)^{157,159}Tb
  - ➢ ^{157}Tb, $T_{1/2}$=99 years
  - ✓ Production via ^{156}Dy(n,γ)^{157}Dy(EC)^{157}Tb
  - ❖ Risks of isotopic contamination with ^{158}Dy(n,γ)^{159}Dy(EC)^{159}Tb

✓ Requires characterization in Mainz

^{156}Dy Stable	^{157}Dy β+	^{158}Dy Stable	^{159}Dy e- capture	^{160}Dy Stable
^{155}Tb e- capture	^{156}Tb β+	^{157}Tb e- capture	^{158}Tb β+	^{159}Tb Stable
^{154}Gd Stable	^{155}Gd Stable	^{156}Gd Stable	^{157}Gd Stable	^{158}Gd Stable
^{153}Eu Stable	^{154}Eu β-	^{155}Eu β-	^{156}Eu β-	^{157}Eu β-

M. Mougeot et al, … neutron-rich Tb … approaching the doubly mid-shell in Rare Earth nuclei…, INTC-I-217.

Interdisciplinary Research Group
Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

Updated request and remaining questions

Separation of ^{108m}Ag

- Can be produced at ILL
- Should contain 1-5% ^{108m}Ag
- Should take 1 to 2 weeks of operation
- Ampoule handling should be done by CERN
  - ✓ Ampoule preparation by Charlotte Duchemin (KU Leuven/CERN EN-STI-RBS)
  - ✓ New protocol by EN-STI-RBS to reduce time & exposure after irradiation; demonstrated & approved by RP

Separation of ^{138}La

- Enriched material with 5% abundance commercially available
- Laser ionization scheme developed at TRIUMF and compatible with RILIS operation

- Other activities are moved to Mainz: quality control of ^{137}La, $^{157,158}Tb$
- No need for new target units, provided that they do not contain isobaric impurities at A=108,138

Instituut voor Kern- en Stralingsfysica
Department of Physics & Astronomy

Collaboration

… for the interest in absolute radii for laser spectroscopy

… for the muX collaboration

… for the production & purification