

BVR 50: Progress Report R-16-01.1

Measurement of the charge radius of radium

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Ch.E. Düllmann^{4,6,7}, R. Eichler², P. Indelicato⁸, K. Jungmann⁹,
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and L. Willmann⁹

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⁷Helmholtz Institute Mainz, Germany

⁸LKB Paris, France

⁹University of Groningen, The Netherlands

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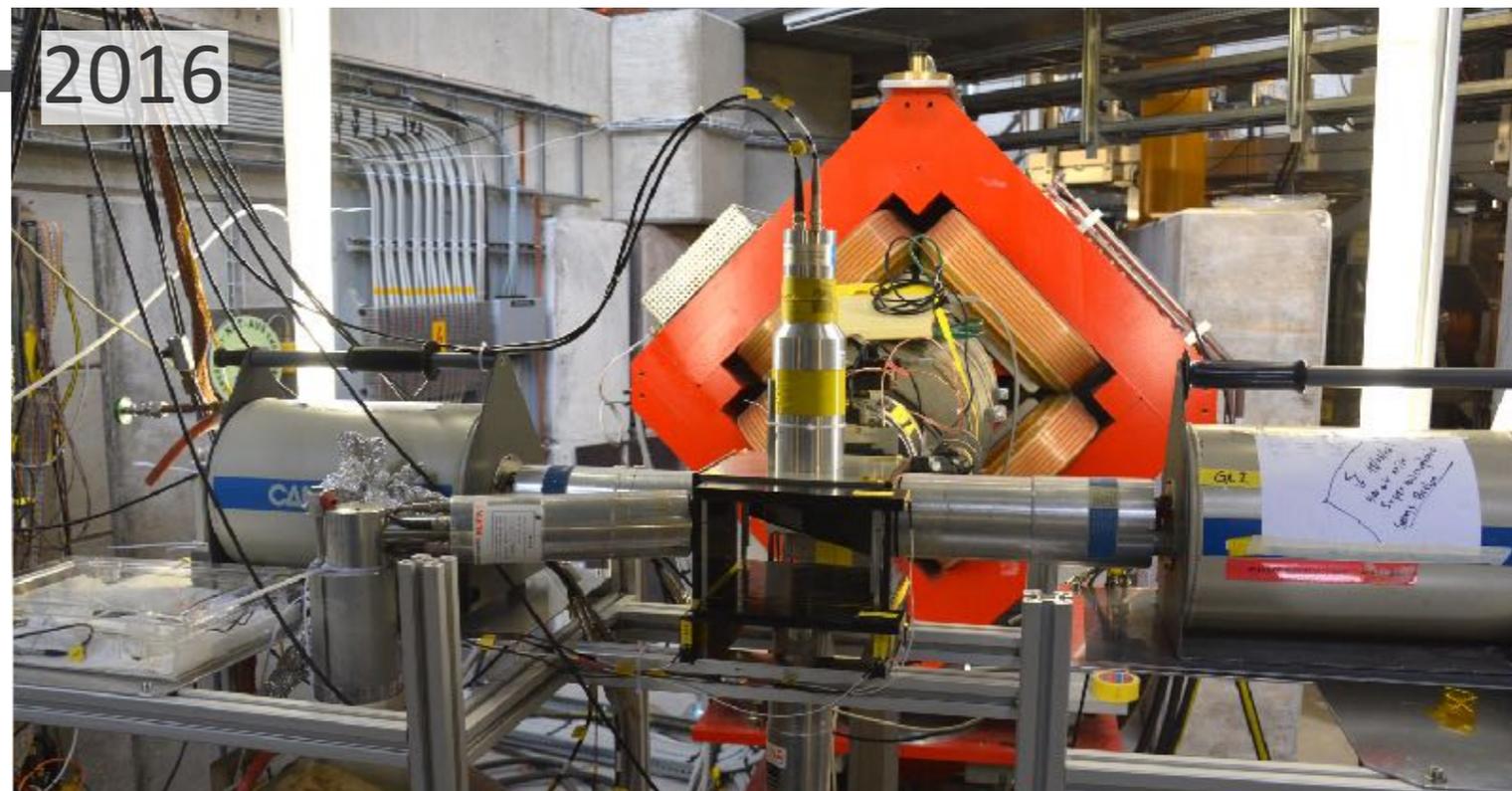
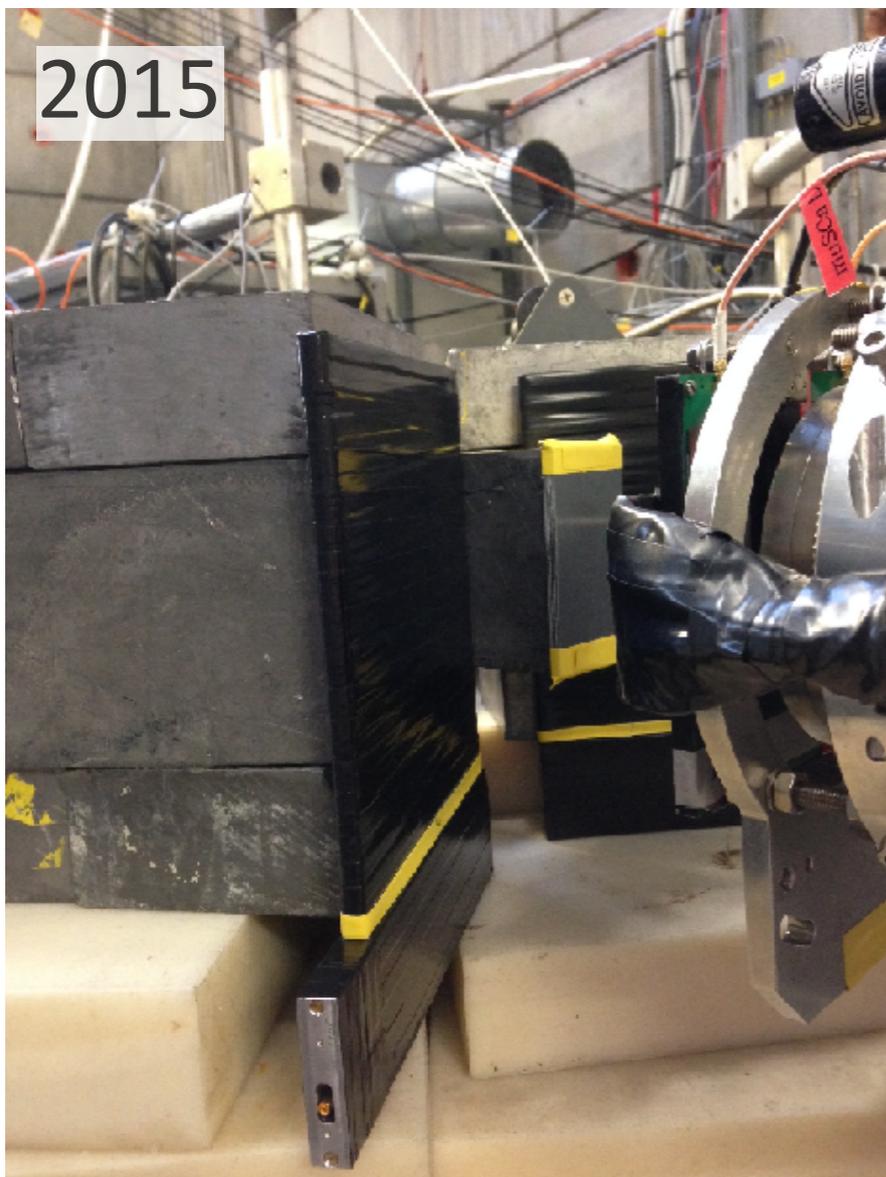
¹¹University of Victoria, Canada

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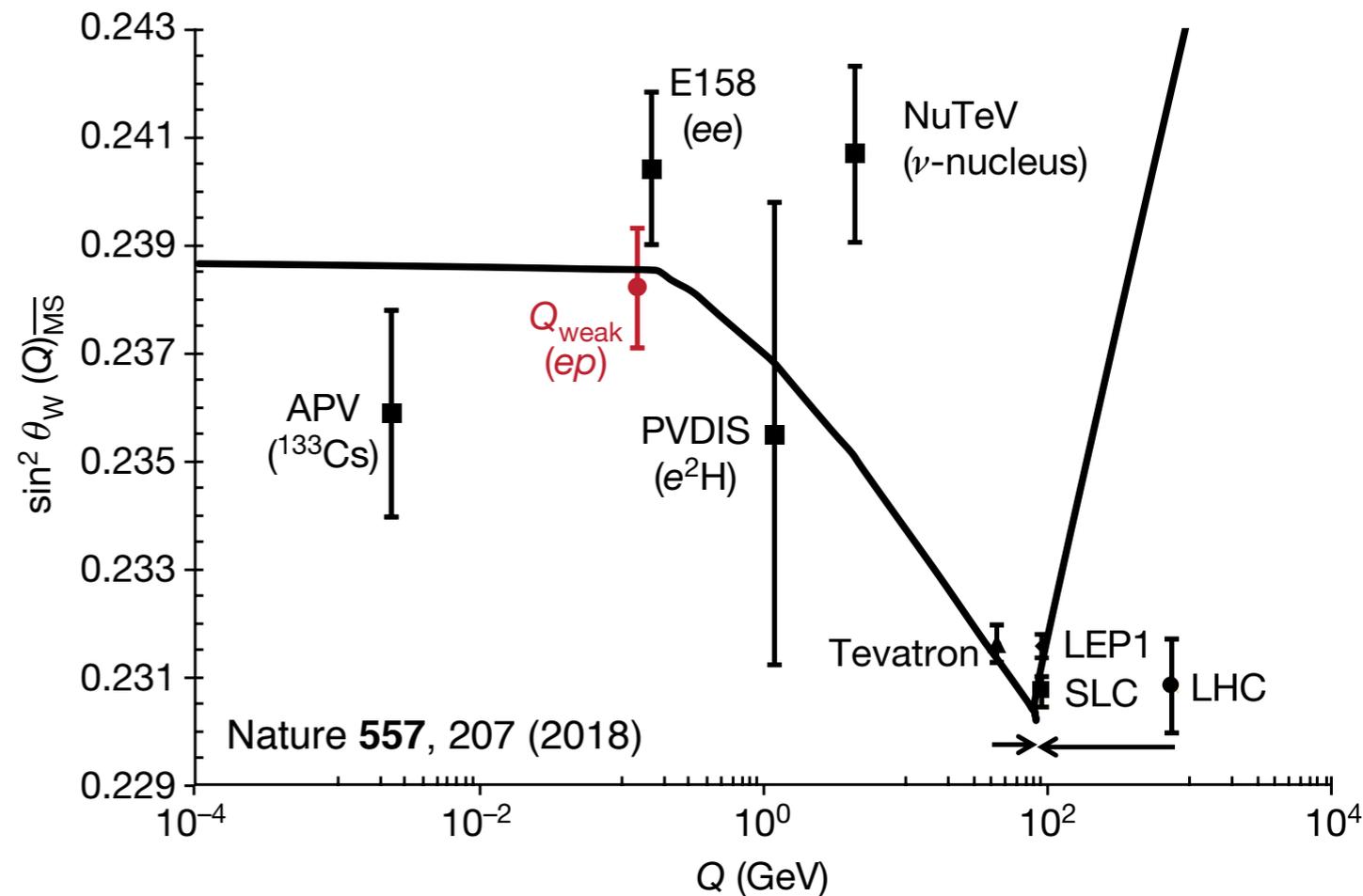
¹⁴CSNSM, Université Paris Sud, CNRS/IN2P3, Université Paris Saclay,
Orsay Campus, France

History



Atomic parity violation in radium

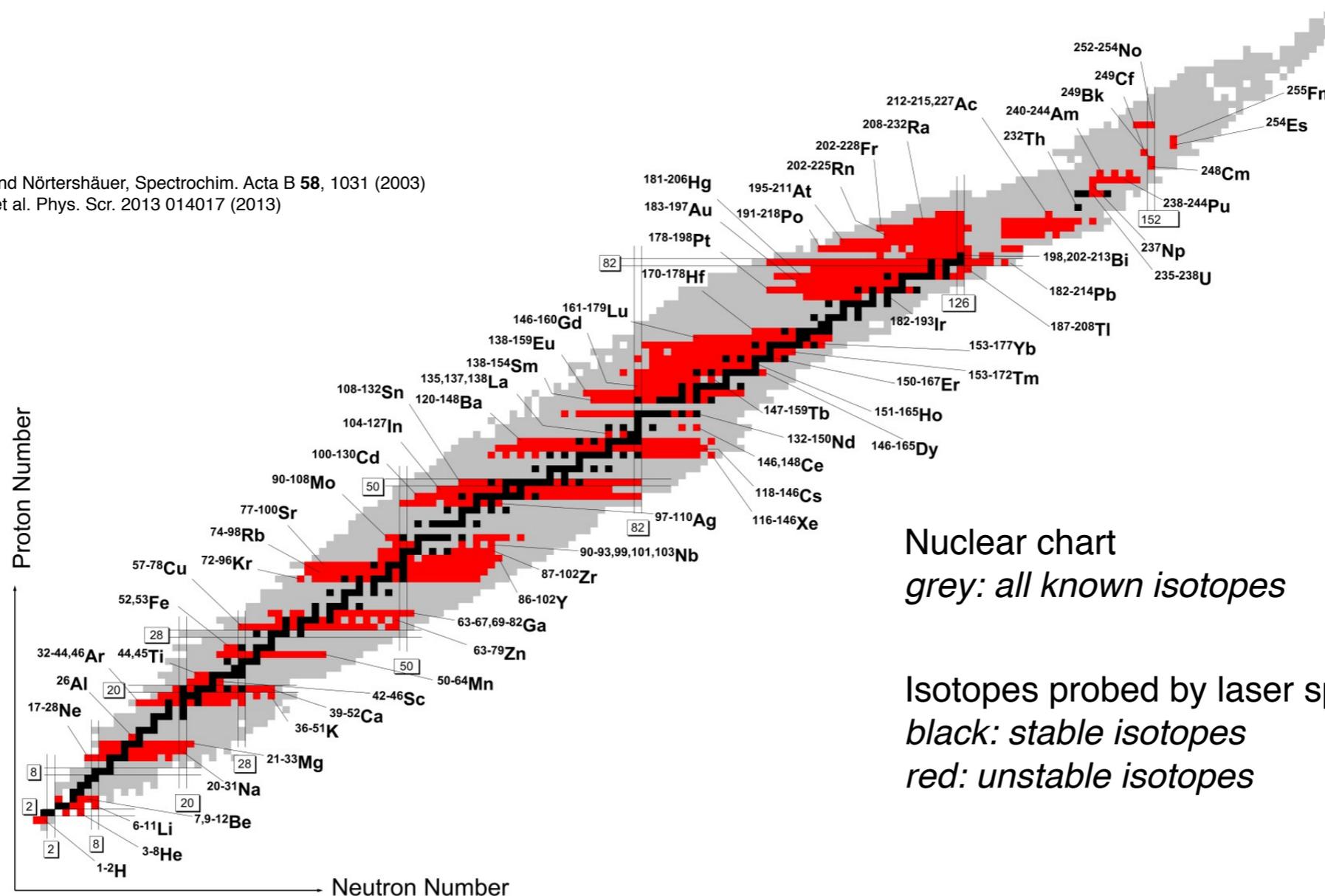
- ▶ Weak interaction leads to parity violating effects in atomic transitions
→ enhanced in heavy atoms ($\propto Z^3$) due to large overlap with nucleus
- ▶ Extract Weinberg angle using precision atomic calculations
→ Needs knowledge of the radium charge radius with 0.2% accuracy



Atomic parity violation fixes weak interaction properties at low momentum

Charge radii in nuclear physics

Kluge and Nörtershäuer, Spectrochim. Acta B 58, 1031 (2003)
Blaum et al. Phys. Scr. 2013 014017 (2013)



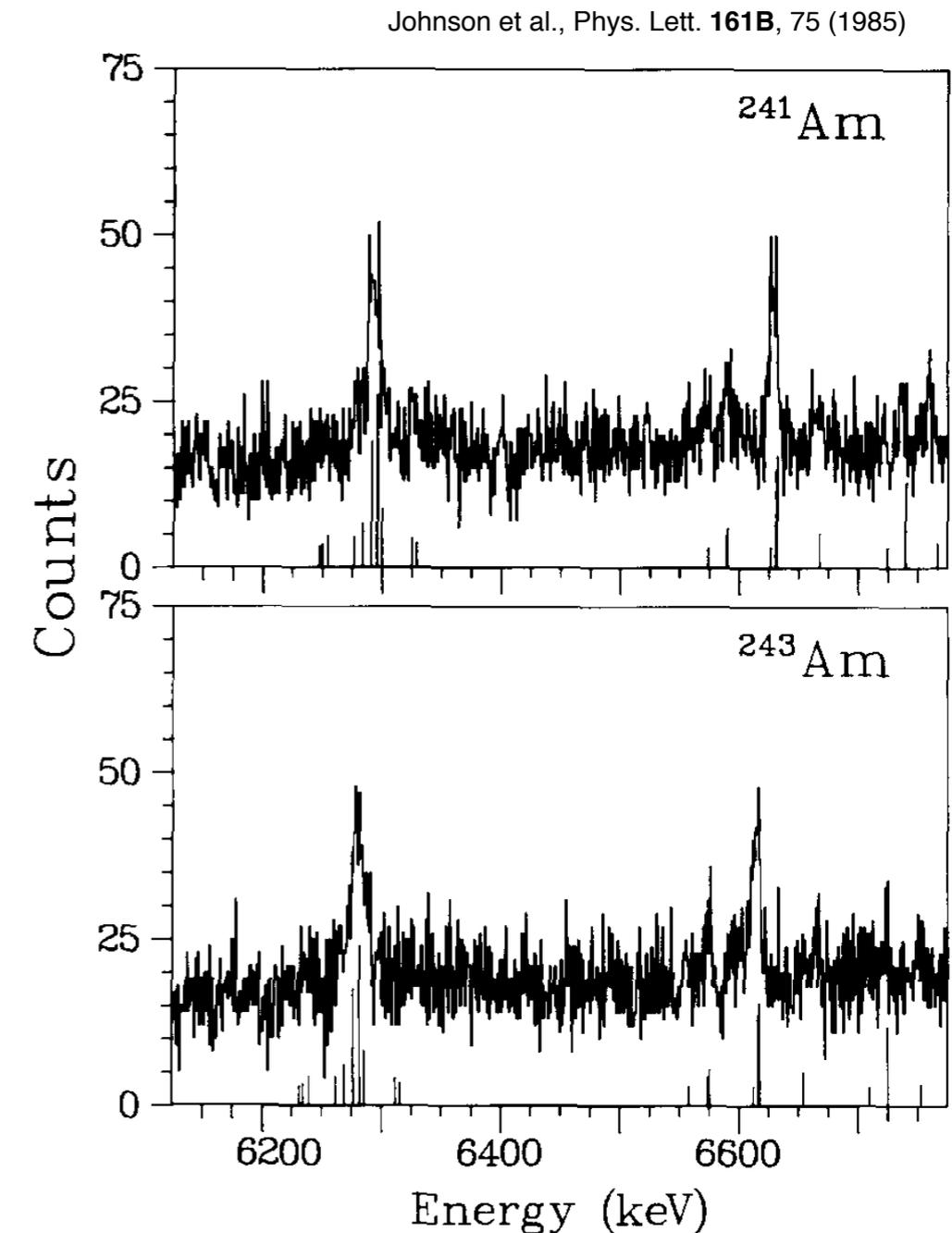
Nuclear chart
grey: all known isotopes

Isotopes probed by laser spectroscopy
black: stable isotopes
red: unstable isotopes

- ▶ Large efforts at ion beam facilities to determine charge radii
- ▶ Wealth of information on nuclear properties from laser spectroscopy
- ▶ Need electron scattering or muonic atom spectroscopy for absolute radii

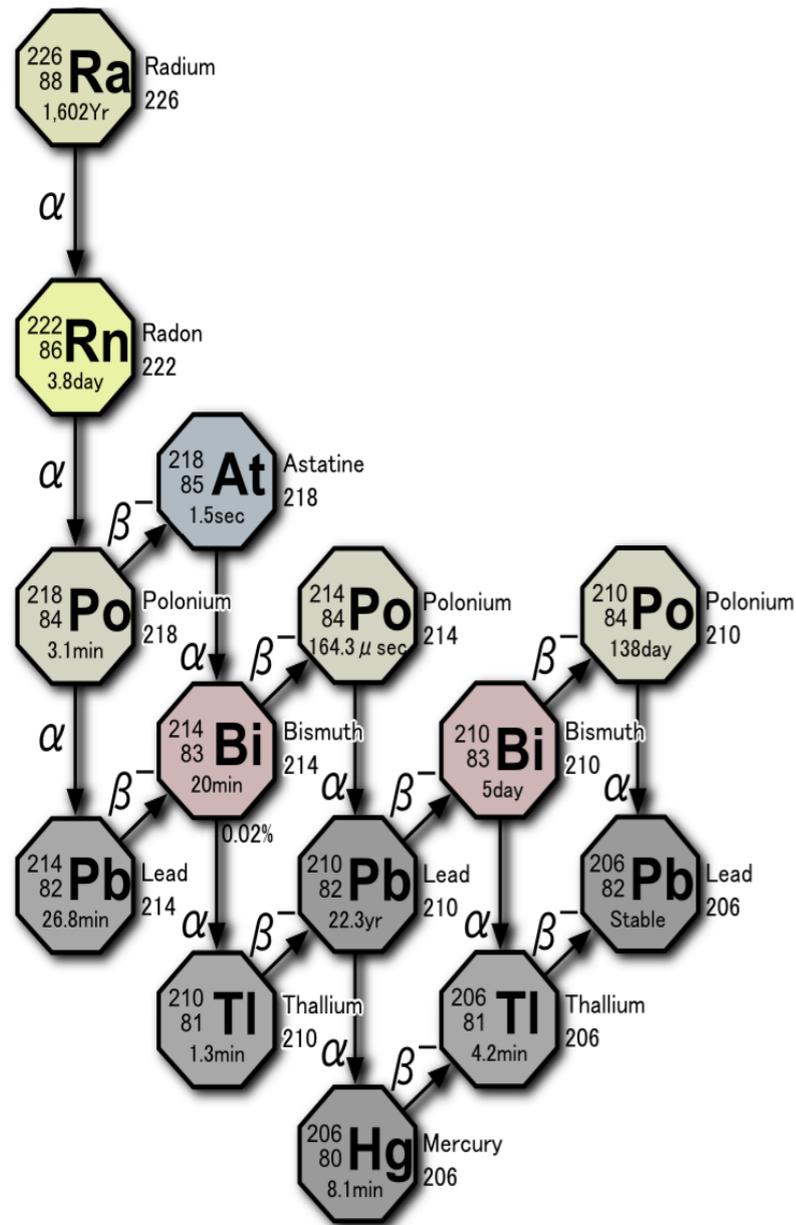
What about radioactive atoms?

- ▶ Most of the stable isotopes have been measured with muonic atom spectroscopy
- ▶ In a few special cases also radioactive isotopes, e.g. americium
 - ▶ The paper describes the americium target as “modest weight of 1 gram”
- ▶ Nowadays: 0.2 μg of ^{241}Am allowed in experimental hall...



Cannot stop muons directly in microgram targets
Need new method!

Our radioactive targets



^{248}Cm , 3×10^5 y

SF: 8%

α : 92%

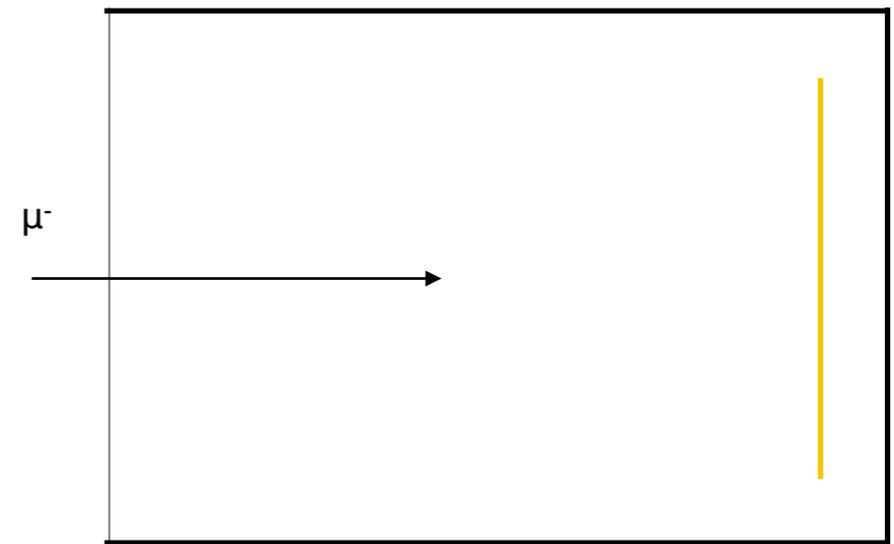
^{244}Pu , 8×10^7 y

- ▶ 5.5 μg target material allowed
- ▶ Gamma rate of ~ 400 kHz from all daughters
- ▶ Interest from atomic parity violation

- ▶ 32.6 μg target material allowed
- ▶ Heaviest nucleus accessible

Transfer reactions

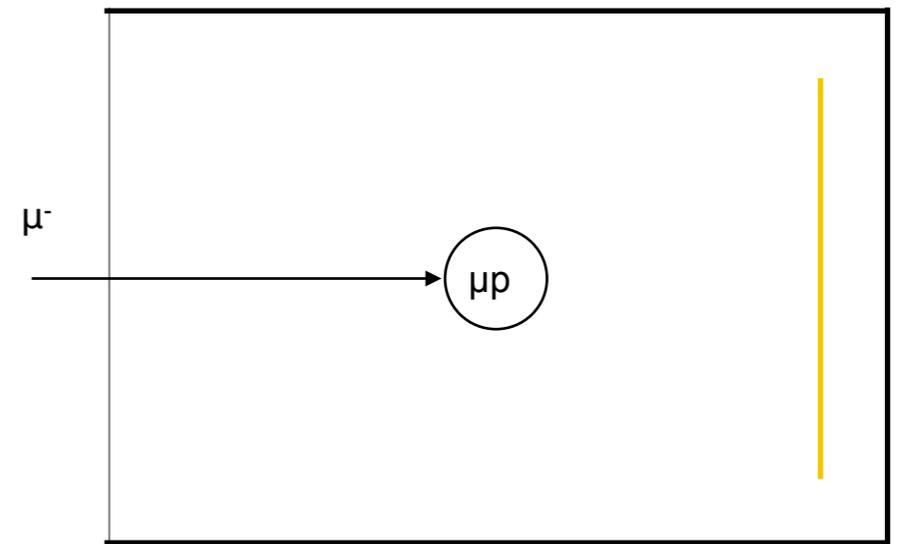
- ▶ Stop in 100 bar hydrogen target with 0.25% deuterium admixture
- ▶ Form muonic hydrogen μp
- ▶ Transfer to deuterium forming μd , gain binding energy of 45 eV
- ▶ Hydrogen gas quasi transparent for μd at ~ 5 eV (Ramsauer-Townsend effect)
- ▶ μd reaches target and transfers to μRa
- ▶ Measure emitted X-rays from cascade



Inspired by work of Strasser et al.
and Kraiman et al.

Transfer reactions

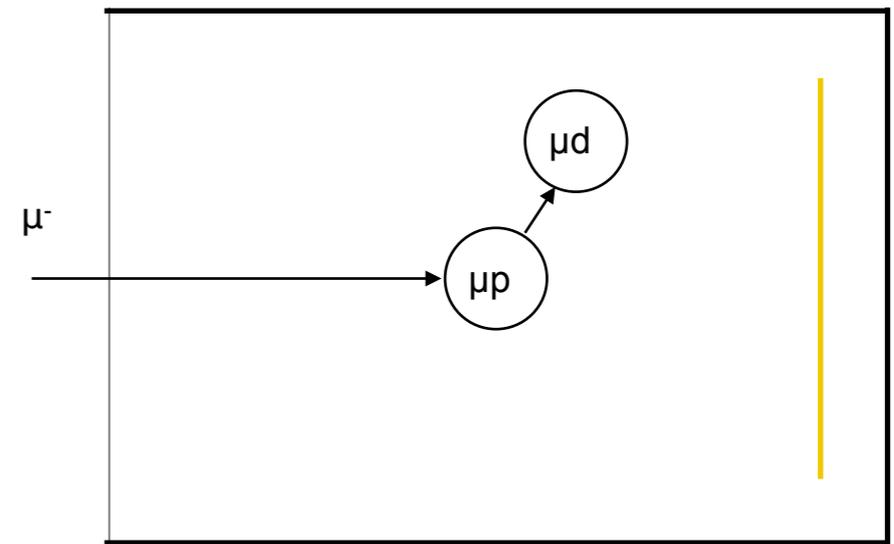
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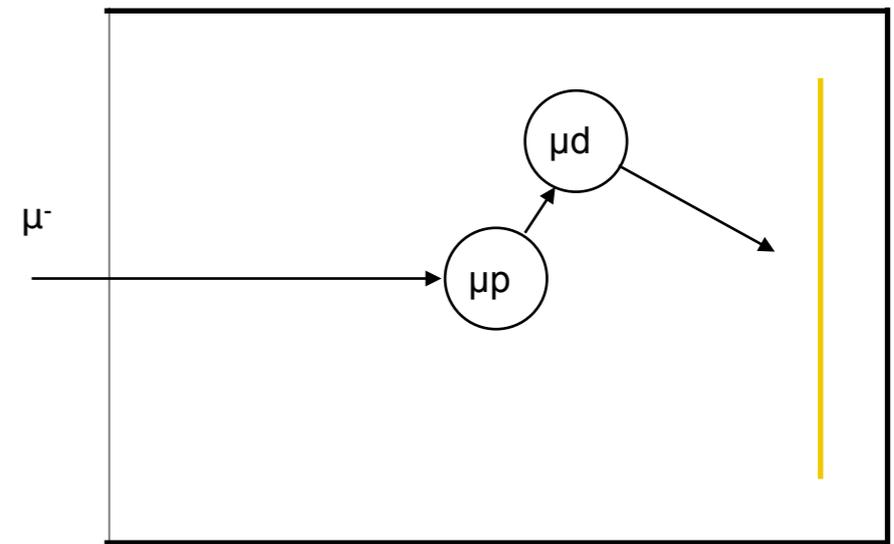
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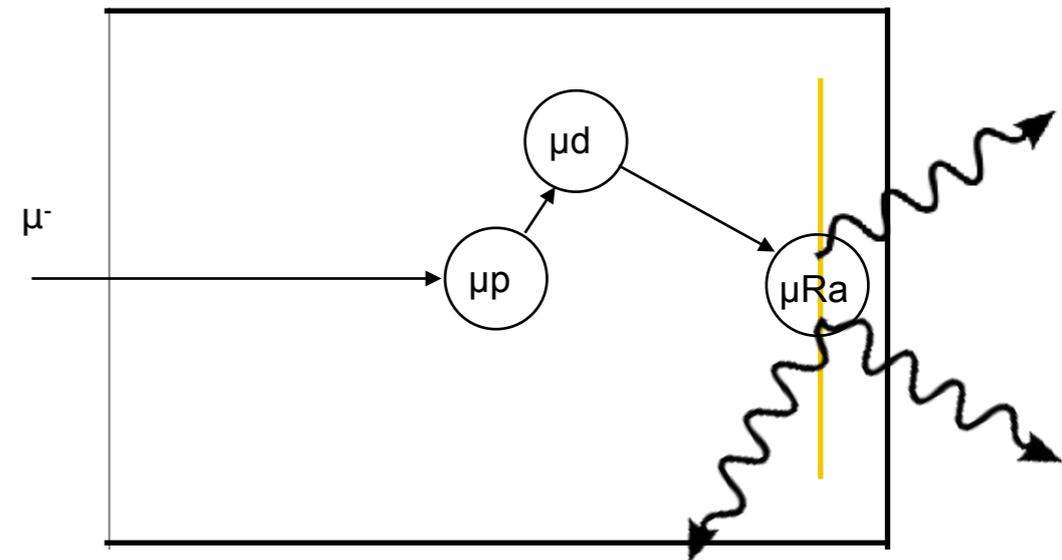
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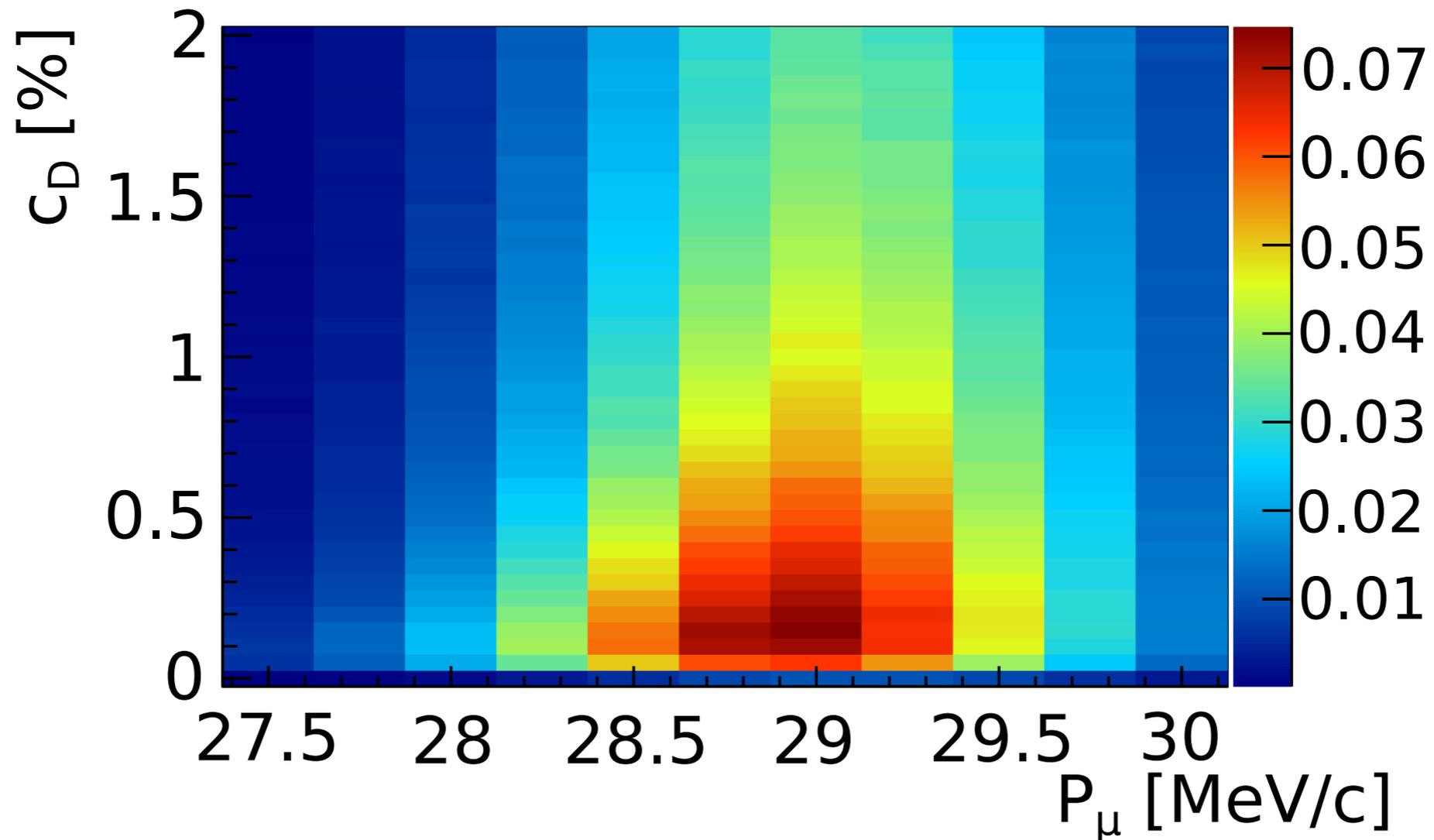
Transfer reactions

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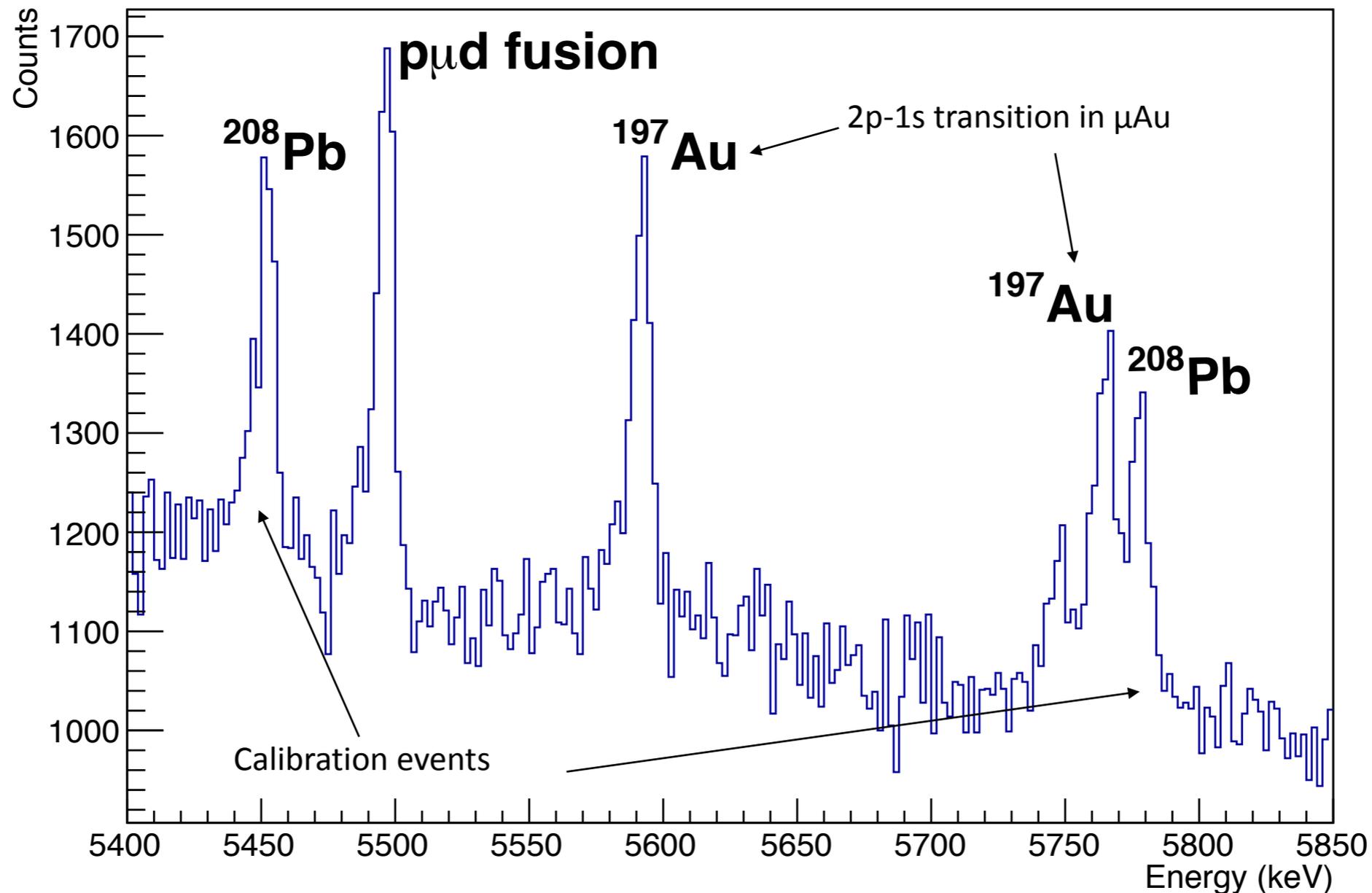


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Simulation of transfer



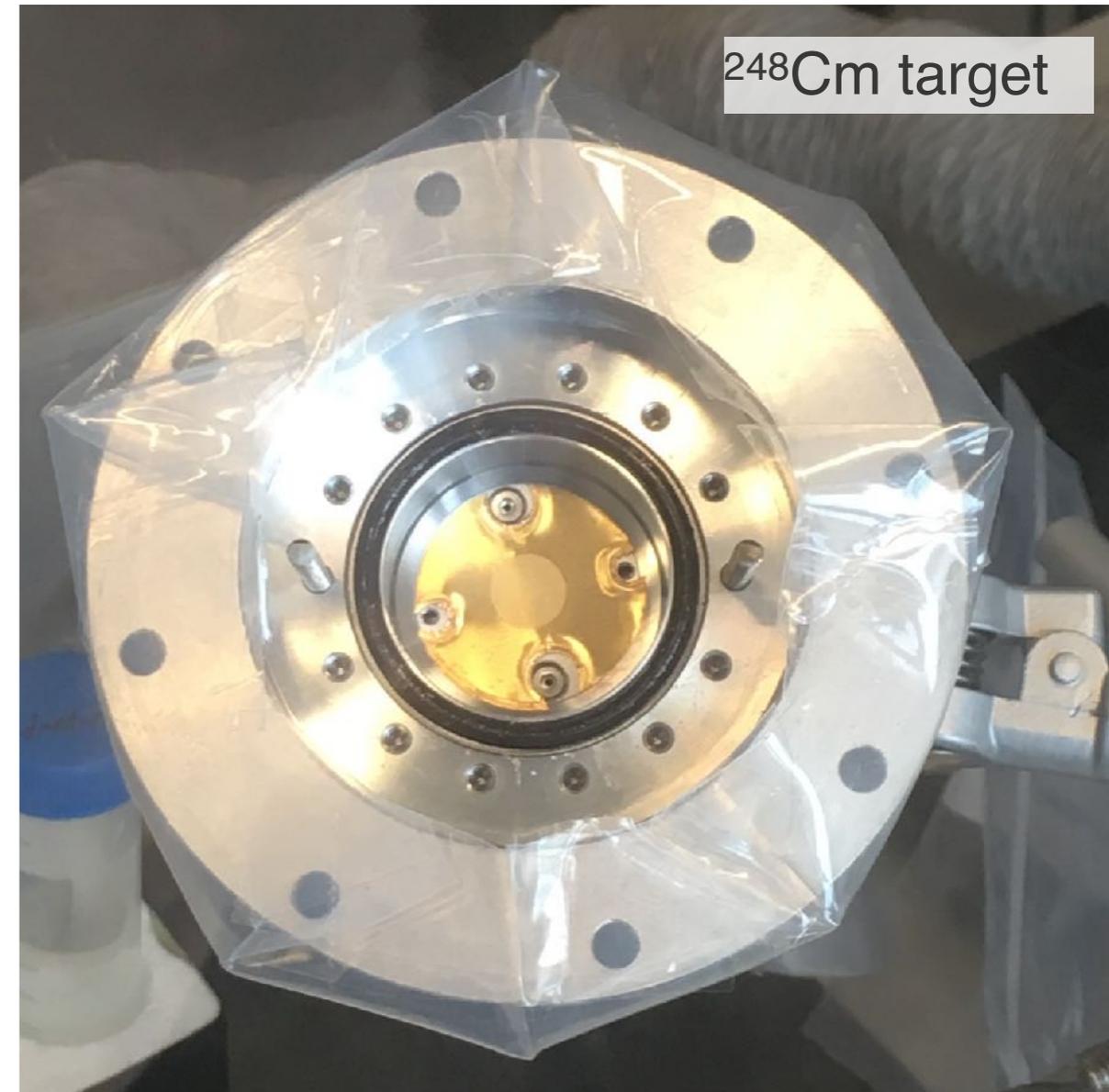
- ▶ Developed simulation to predict efficiency of transfer
- ▶ Momentum of beam determines stopping distribution with respect to the target
- ▶ Deuterium concentration determines speed of transfer but limits range due to $\mu d + D_2$ scattering
- ▶ ~1% efficiency for 5 μg radium target expected



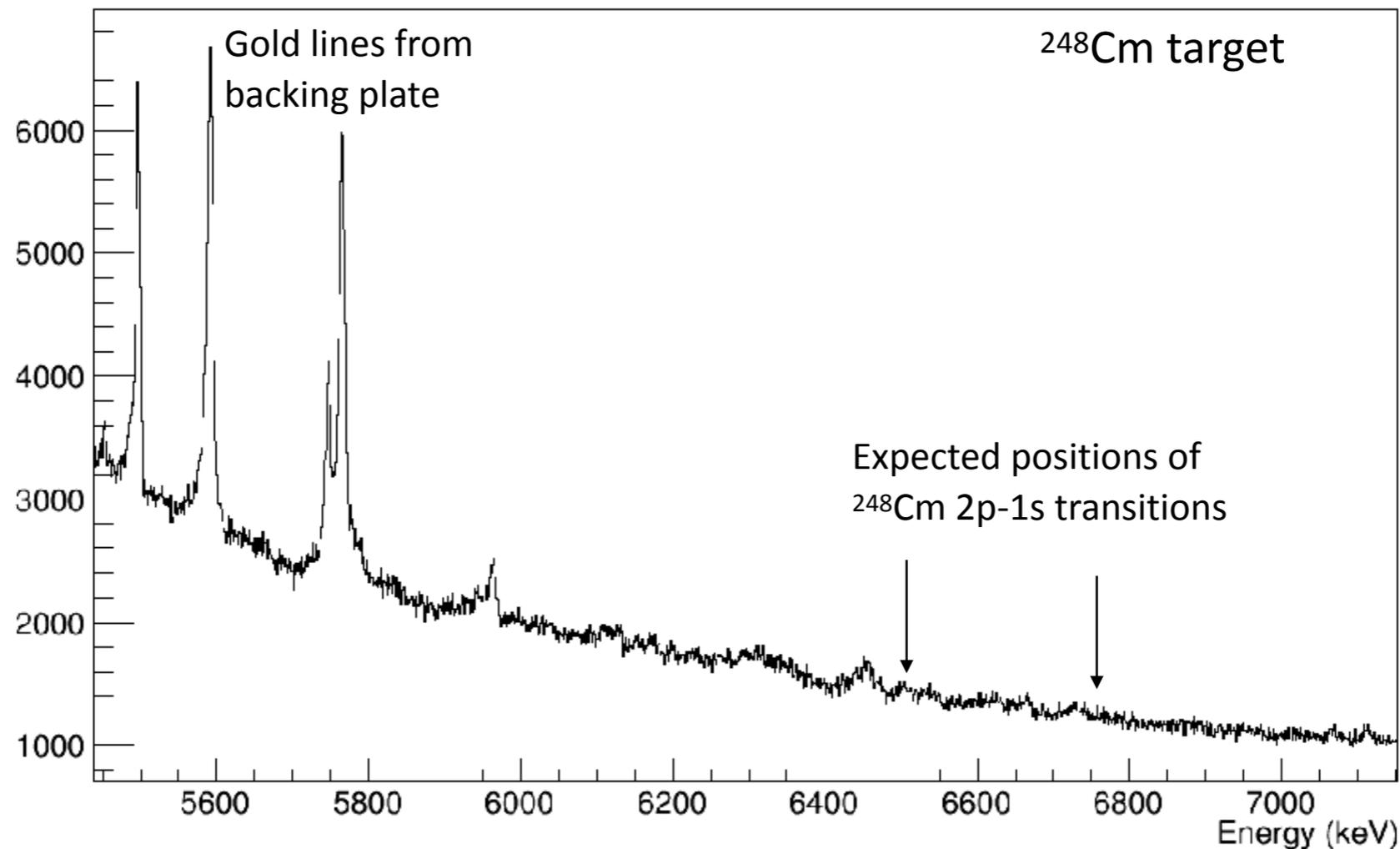
- ▶ Measurement with 5 μg gold target as proof-of-principle
- ▶ Spectrum taken over 18.5 h
- ▶ Setup tested with high-rate gamma sources and uranium targets of a few mg

Radioactive targets 2018

- ▶ Curium-248 target was made in Mainz by molecular plating method
- ▶ Radium-226 target was made at PSI also by molecular plating methods
- ▶ Handling and installation of target foils into gas cells was done in a glove box in the laboratories of the radiochemistry at PSI



Measuring radioactive targets



- ▶ In the end we did not see any sign of curium x-rays
- ▶ Electroplating inherently leads to organic layers on the target
- ▶ The fact that we see the outline of the target clearly indicates a reasonably thick layer
- ▶ Tried several times to burn away organic layer on curium target, but without success
- ▶ For radium there were unexpected issues with the plating
→ only 1% of required target mass on target foil

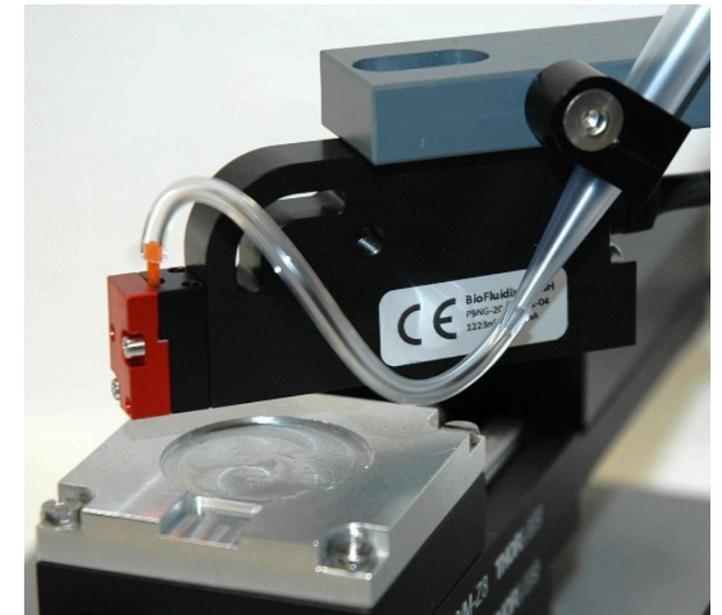
Carbon coatings on gold

- ▶ In order to understand the influence of the organic layer on our measurements prepared gold coatings with 100 and 500 nm carbon coating on top.
- ▶ Results:
 - ▶ 100 nm: 27% of gold x-rays left
 - ▶ 500 nm: no gold x-rays seen
- ▶ We are much more sensitive to organic layers than expected!

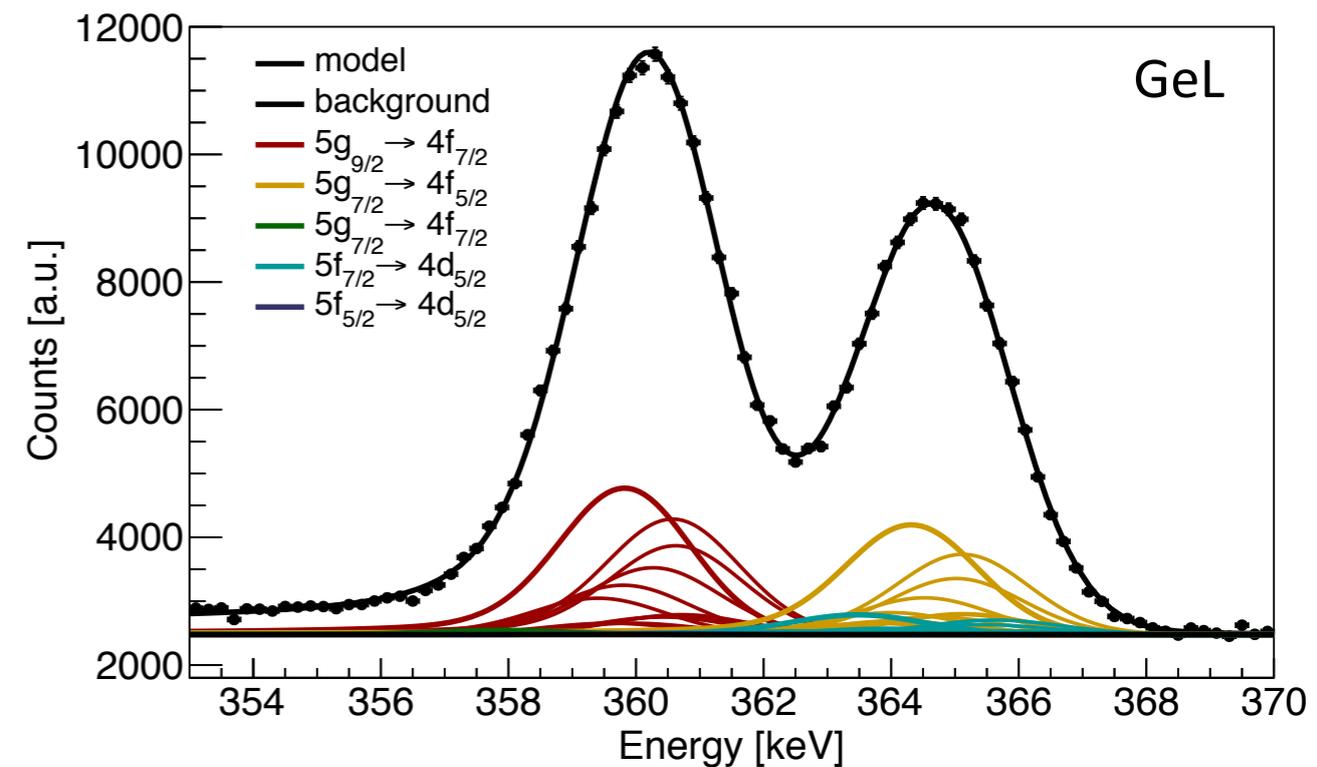
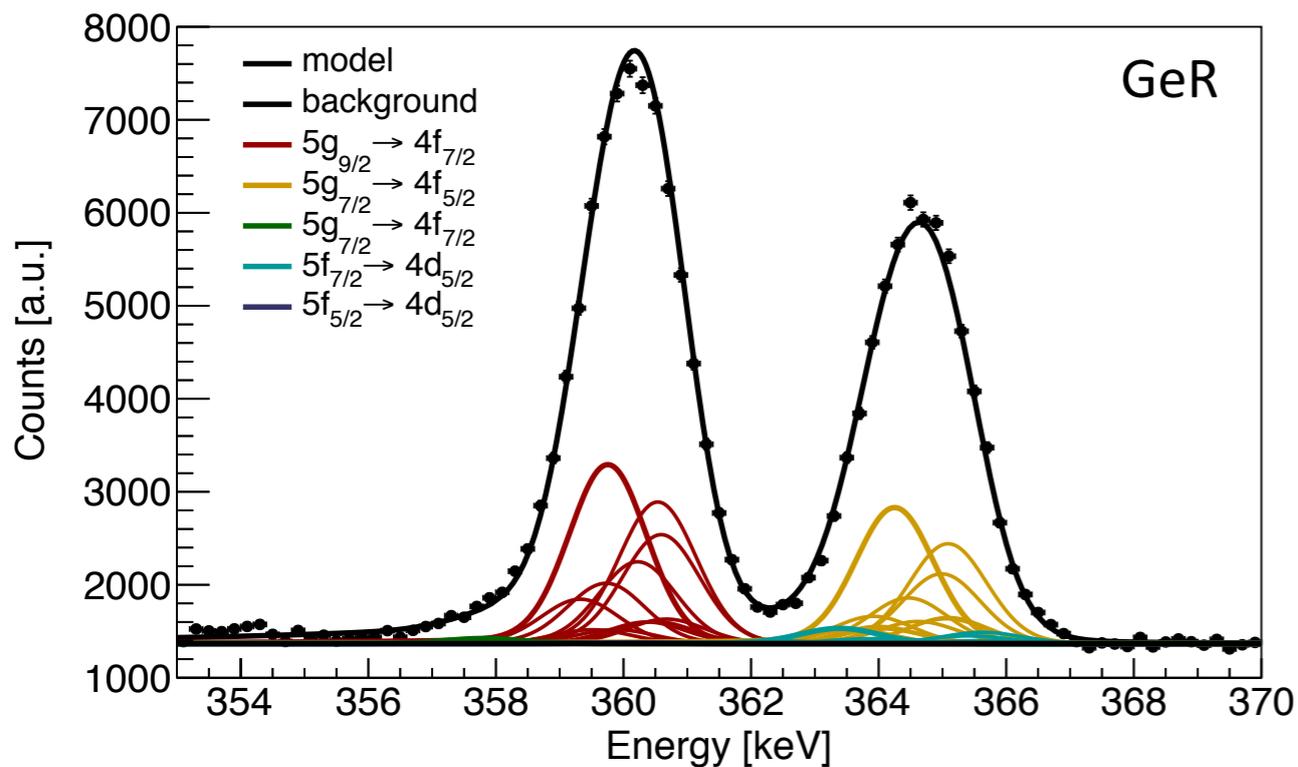


Developments for 2019 campaign

- ▶ Radioactive target developments:
 - ▶ Drop-on-demand technique in Mainz (for curium & radium)
 - ▶ Intermetallic targets at PSI (for radium)
 - ▶ Offline methods to measure $O(10\text{ nm})$ thick layers of organic contamination
- ▶ Low-Z target cell to reduce background
- ▶ Improved gas handling system to allow pre-mixing of D_2/H_2
- ▶ Use of Miniball germanium array (more in Frederik's talk)



Progress in Theory: Analysis of $^{185,187}\text{Re}$

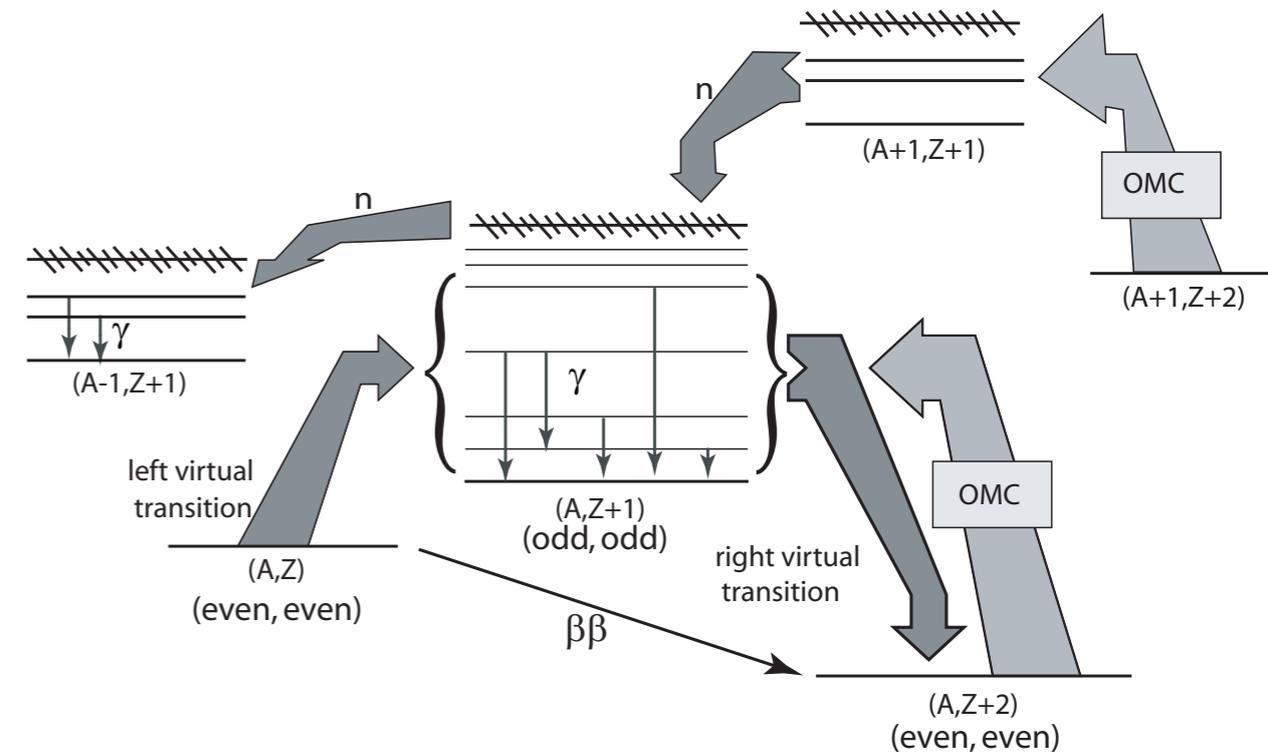


- Analysis of 5-4 transitions in rhenium
- Full calculation of all transition energies and strengths as a function of quadrupole moment Q in collaboration with MPI Heidelberg
- Fit of all 76 transitions to measured spectrum allows to extract Q
- Publication in preparation
- Moving on to lower transitions with sensitivity to charge radius

Addendum to Proposal

- ▶ Muon capture measurement on nuclei relevant for double beta decay (^{82}Kr , ^{130}Xe)
- ▶ Compare measurements to nuclear shell model predictions for ^{24}Mg as a benchmark for NSM to calculate higher mass nuclei
- ▶ Measure muon capture rates and branches to the different daughter isotopes

Zinatulina et al., arXiv:1803.10960 (2018)



target	enr-ment	composition	element mass	thickness mg/cm ²
^{82}Kr	99.9%	Kr gas	1.0 l (1 atm.)	37.3
$^{\text{nat}}\text{Kr}$	—	Kr gas	1.0 l (1 atm.)	37.3
^{130}Xe	99.9%	Xe gas	1.0 l (1 atm.)	58.1
$^{\text{nat}}\text{Xe}$	—	Xe gas	1.0 l (1 atm.)	58.1
^{24}Mg	99.89%	MgO powder	1.0 g	250

Beam request 2019

- ▶ 3.5 weeks of beam time for measurement of charge radius of ^{248}Cm and ^{226}Ra
 - ▶ 1.5 weeks of setup
 - ▶ 2 weeks of data taking

- ▶ Beam time for muon capture measurements: 2 weeks of data taking

- ▶ Additional test requests using the muX setup:
 - ▶ 2s-1s measurements: 1 week (see Frederik's talk)
 - ▶ Elemental analysis (μSR): 1 week

muX collaboration

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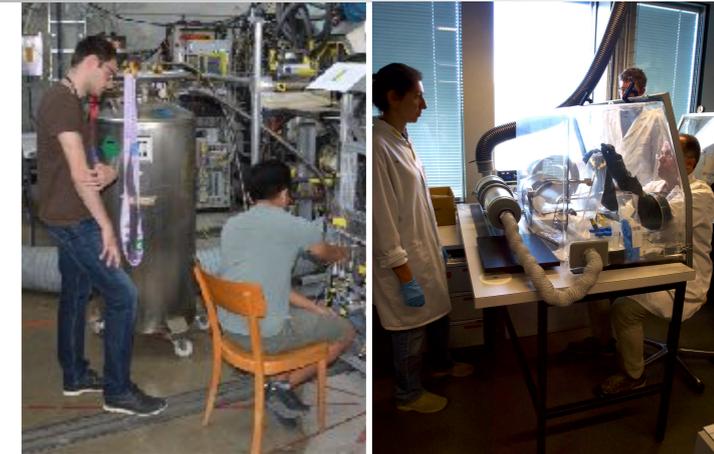
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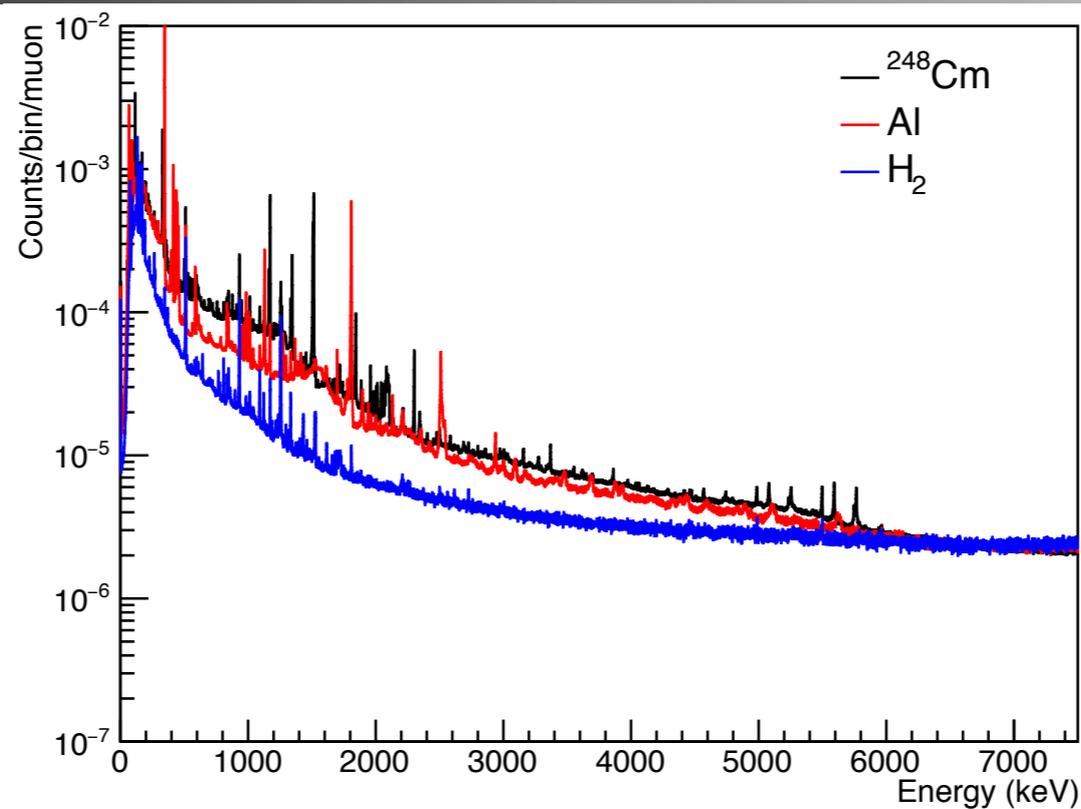
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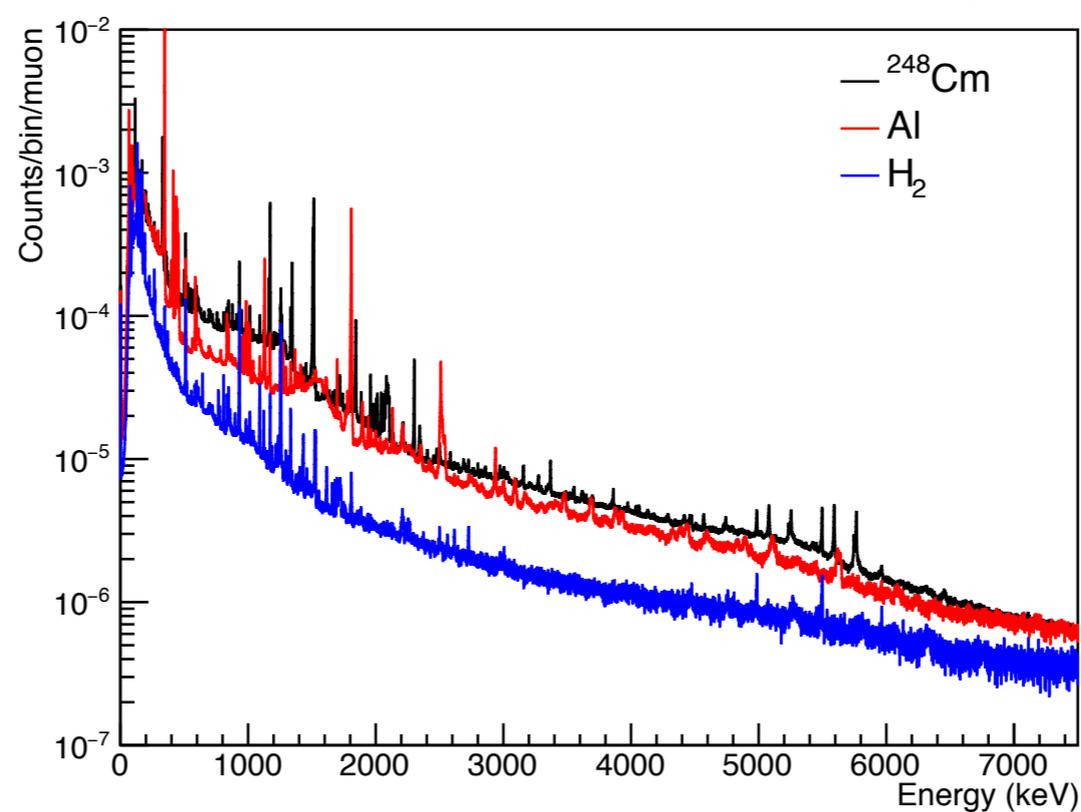


Backup

Background Measurements

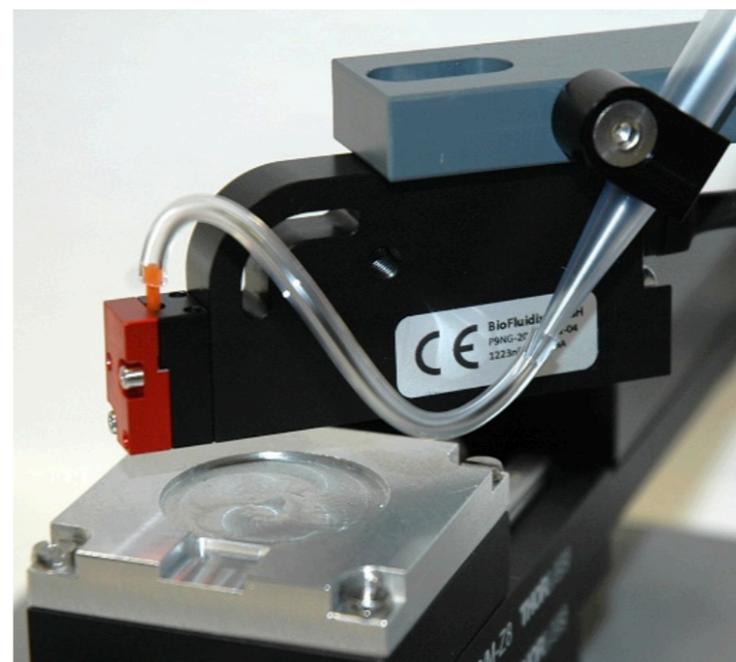
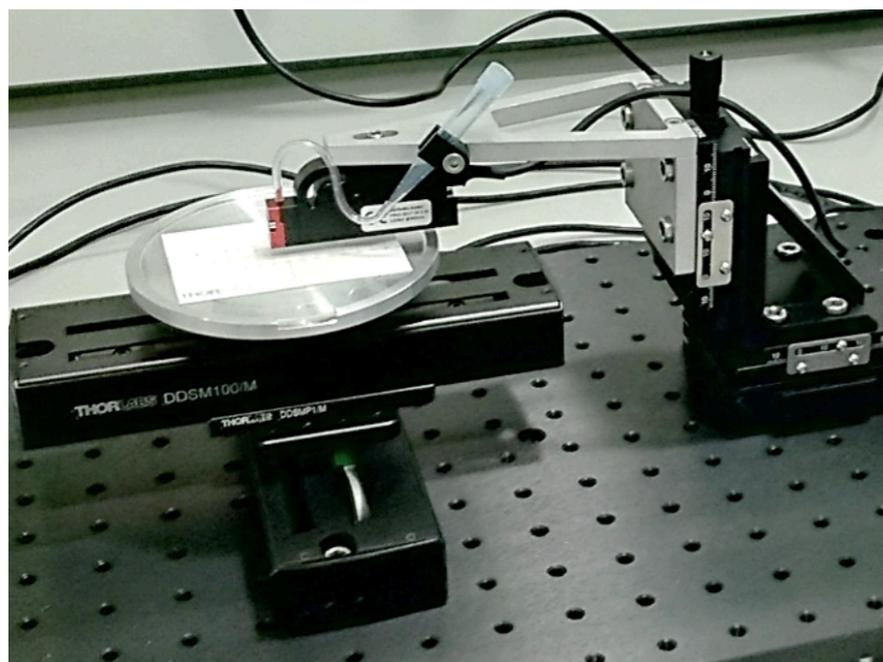


No electron veto

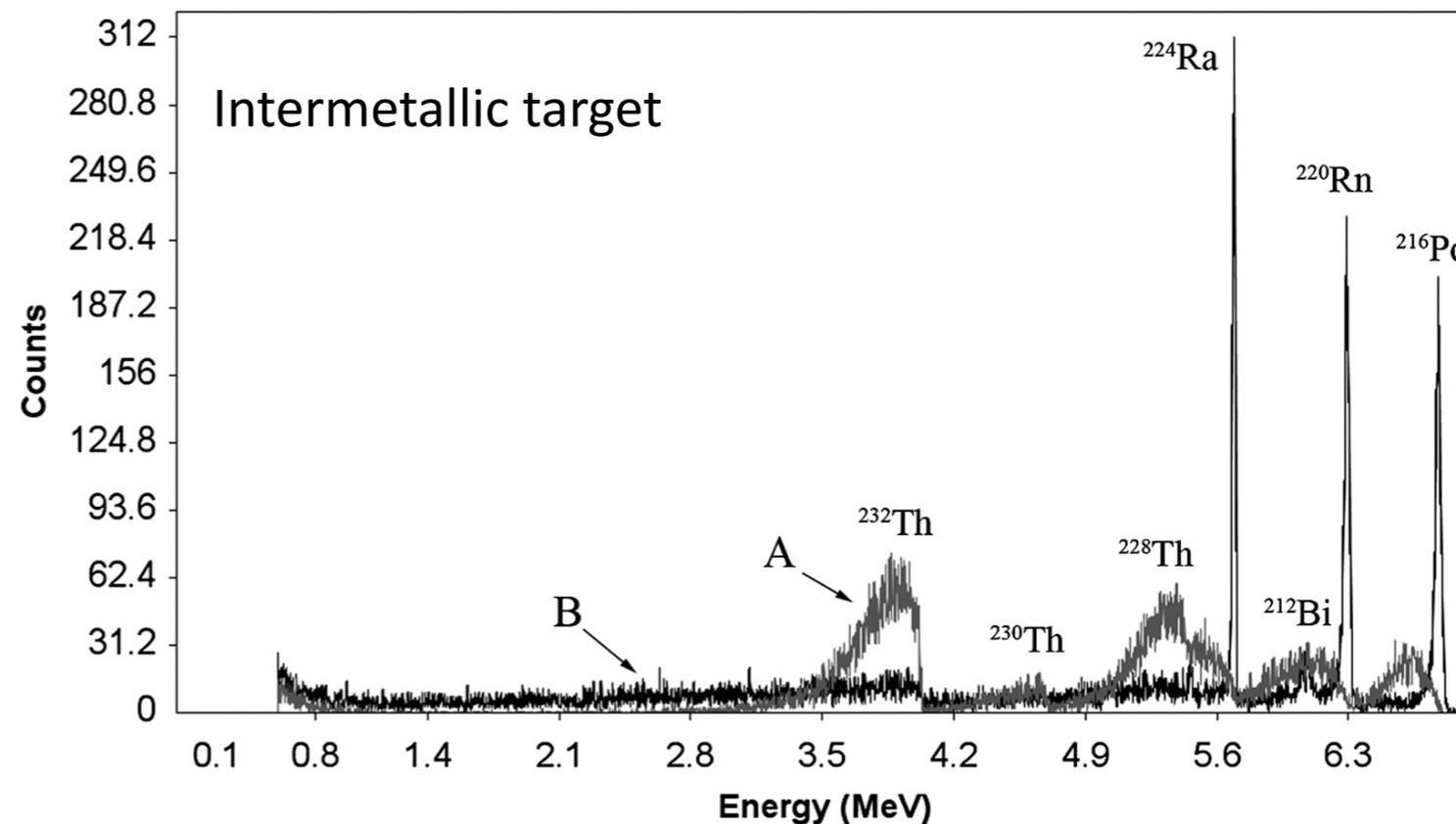


With electron veto

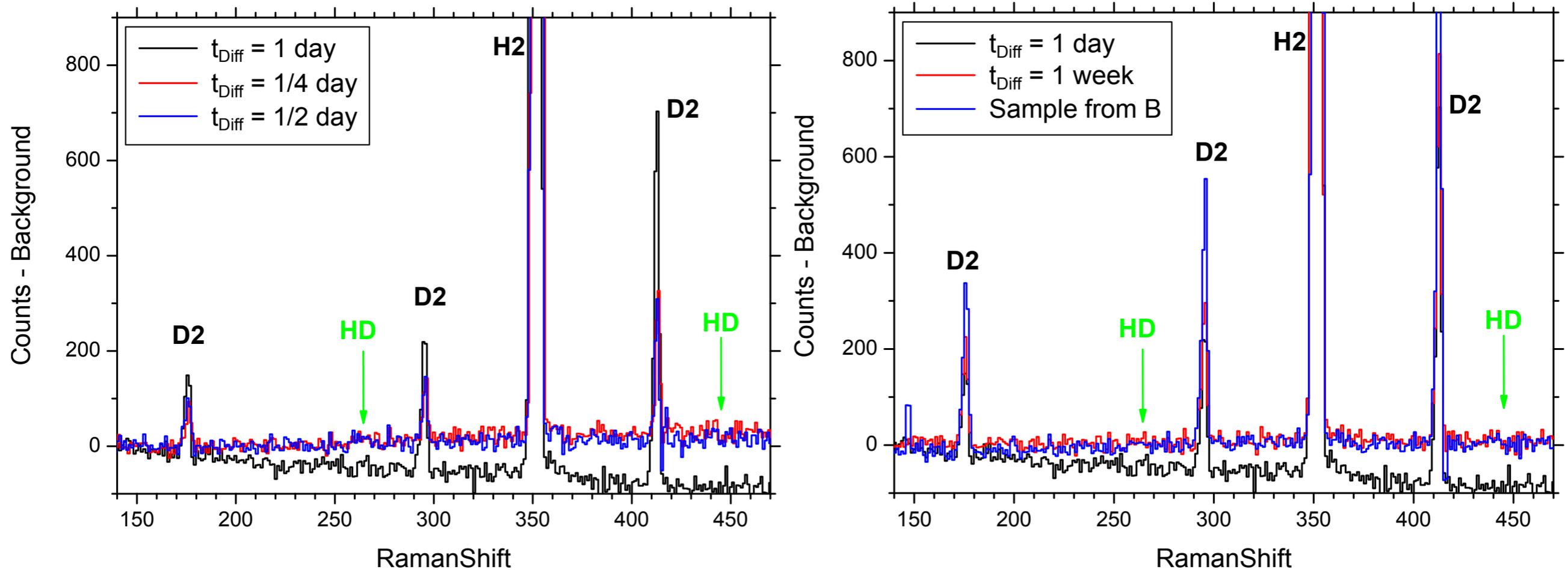
Developments for radioactive targets



Printer for drop-on-demand

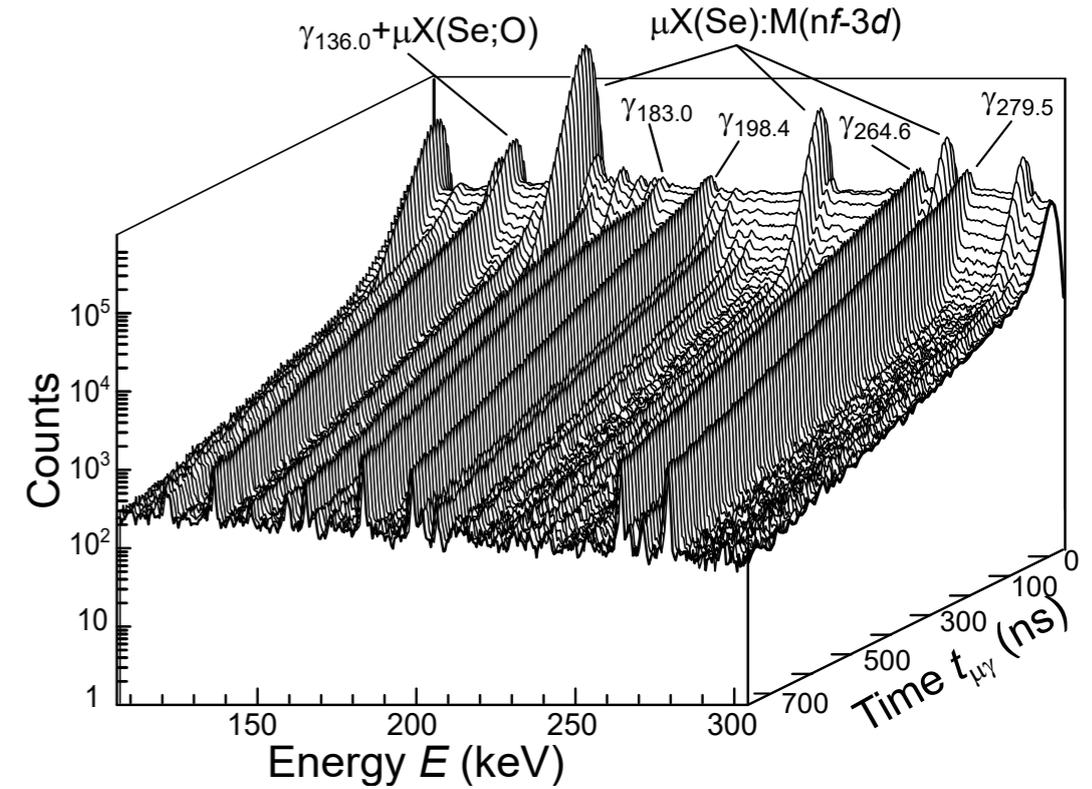
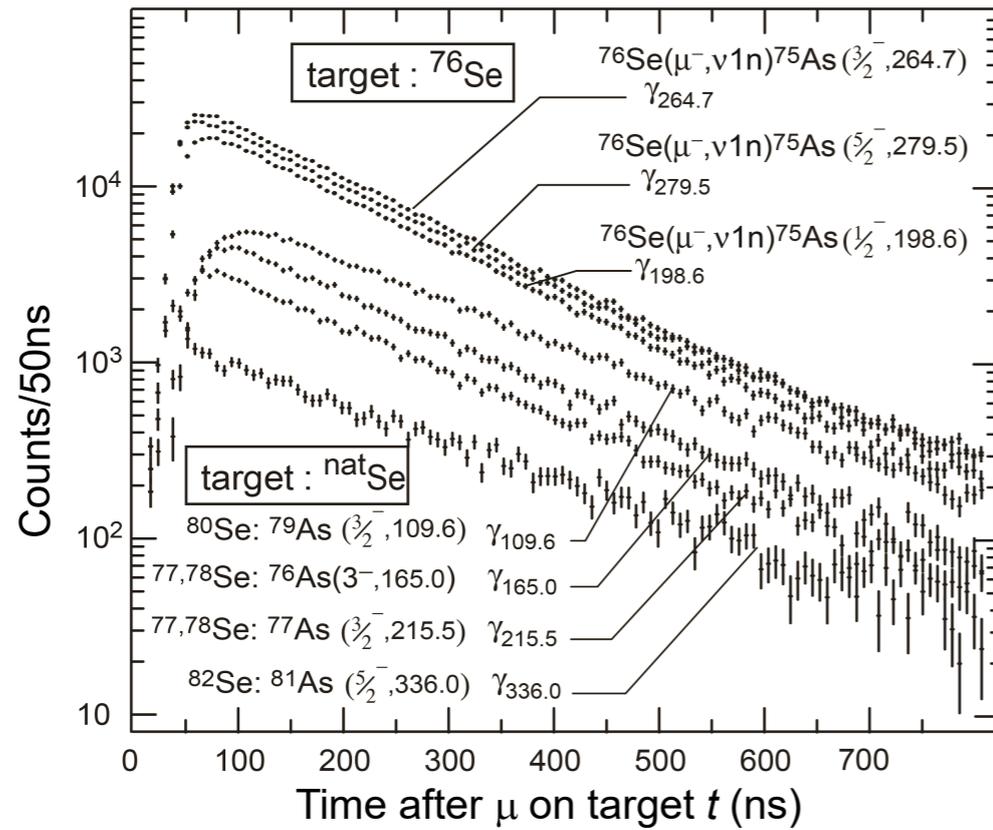


Raman spectroscopy



- Equilibration of D₂ in gas systems takes days/weeks!

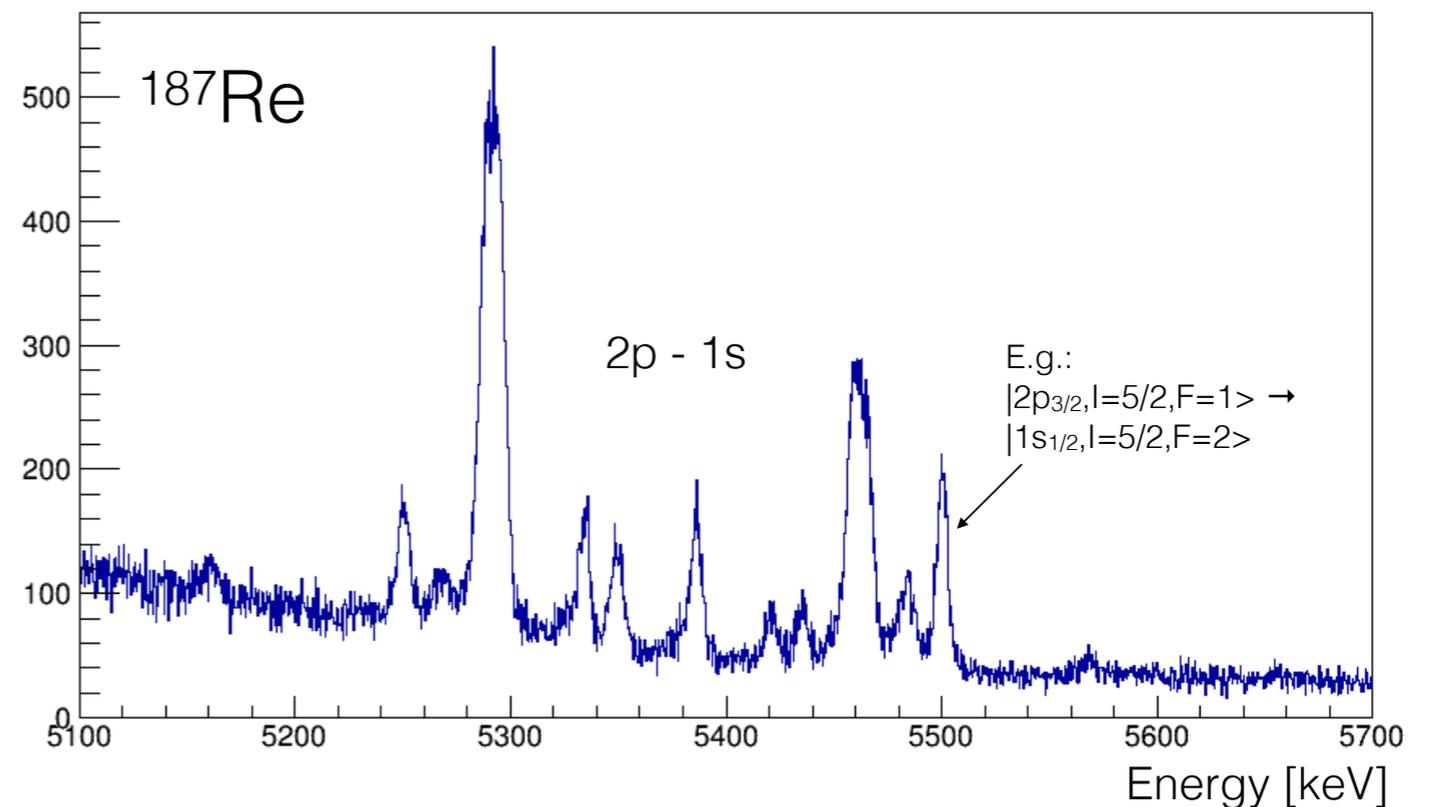
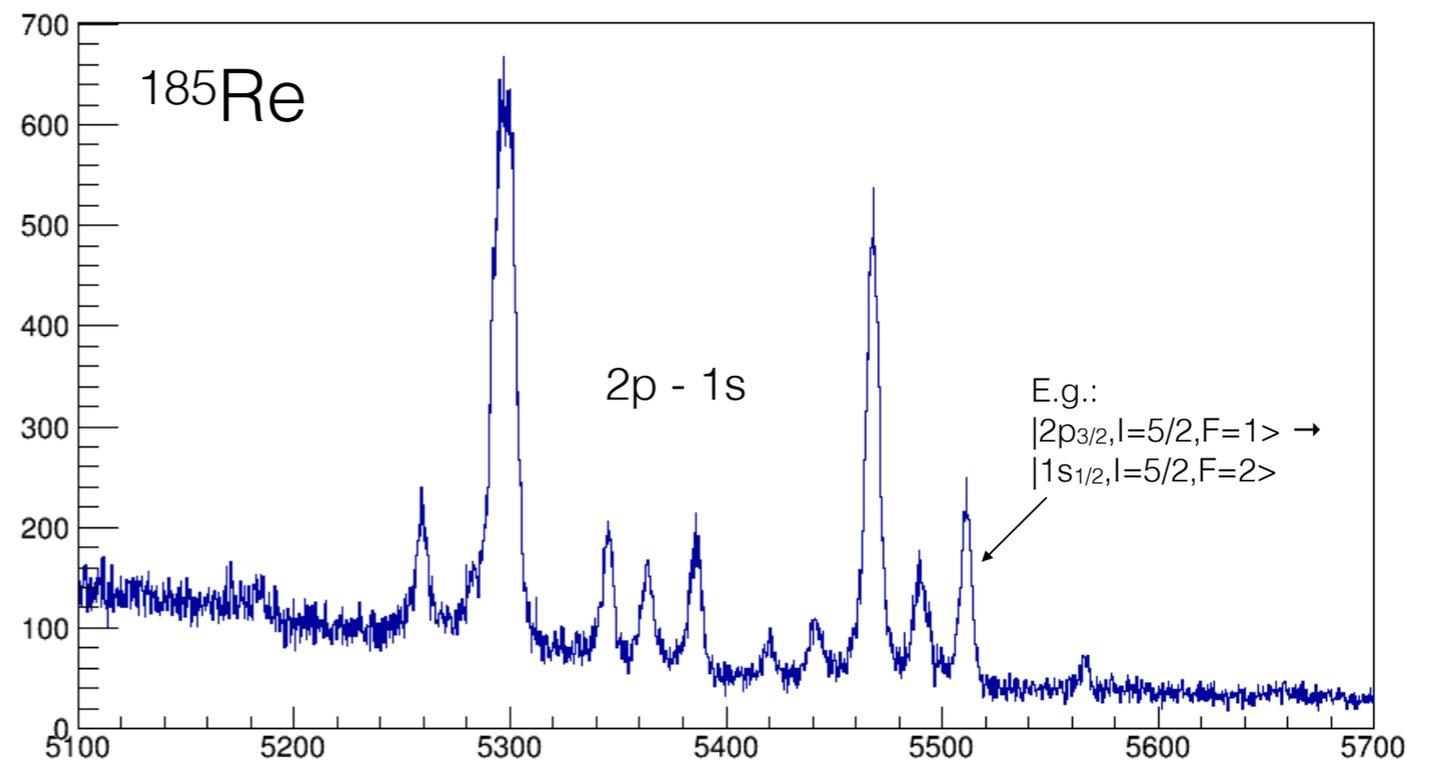
Muon Capture Measurements


 D. Zinatulina et. al *arXiv:1801.06969v2* [nucl-ex]

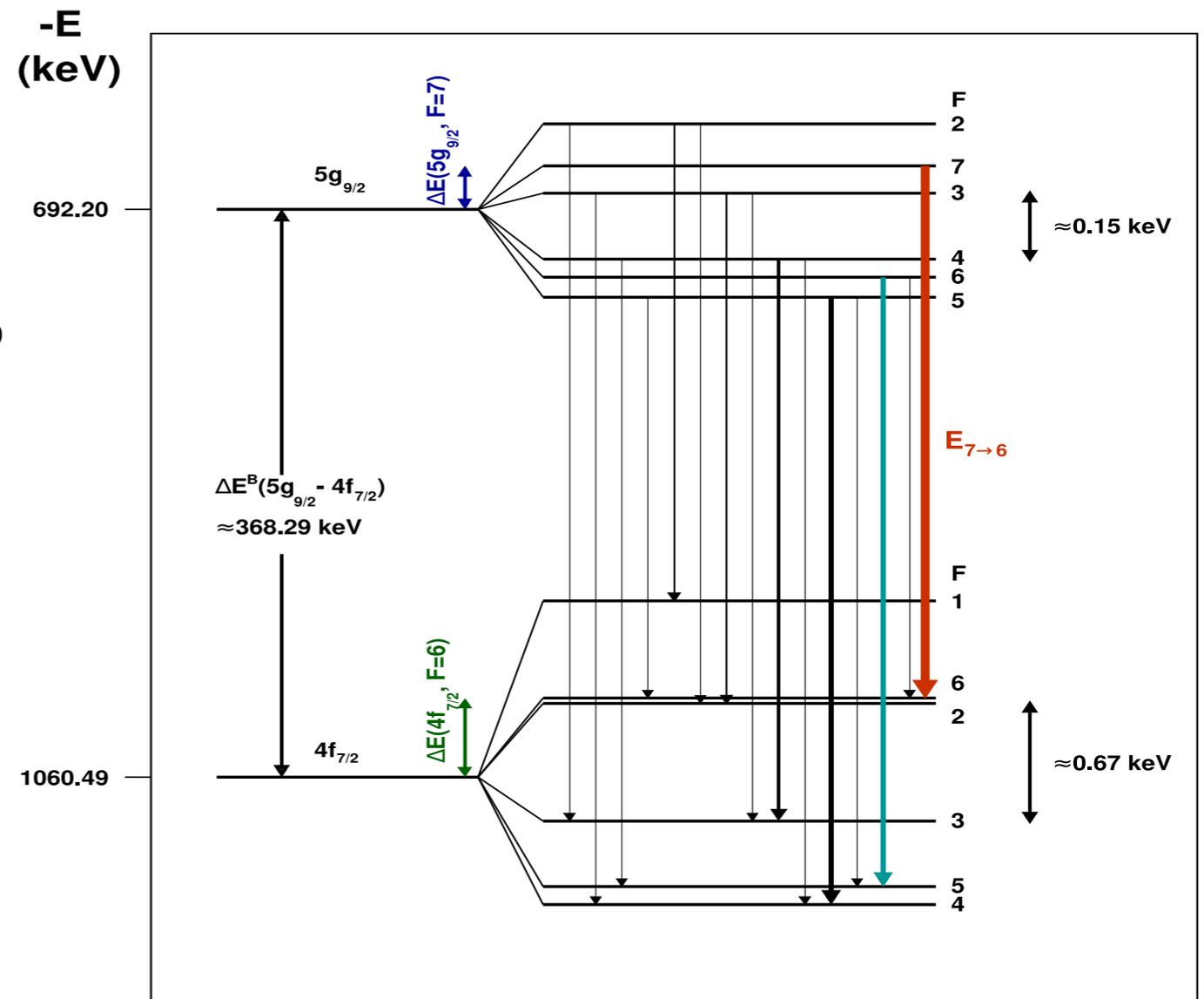
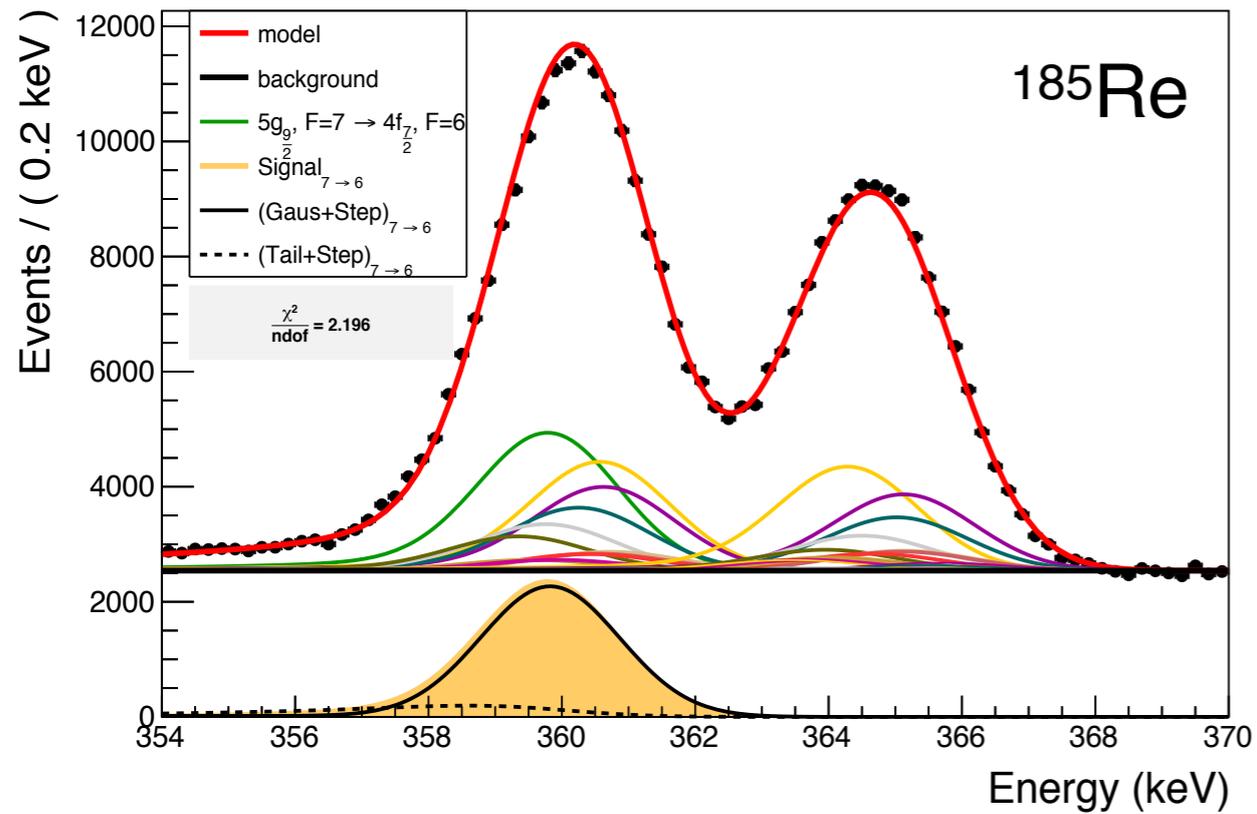
E_j [keV]	J^π	P_j^μ [%]	E_j [keV]	J^π	P_j^μ [%]	E_j [keV]	J^π	P_j^μ [%]
0.0	2^-	g.s.	499.6	$[1^+, 2^-]$	0.99(36)	802.4	$(1^-, 2^-, 3^+)$	0.17(10)
120.3	1^+	0.32(12)	505.2	$(2, 3)^+$	0.25(6)	863.3	1^+	0.27(20)
122.2	$(1)^-$	0.21(11)	517.6	$(1, 2)^+$	0.24(11)	893.2	$(1^-, 2^-, 3^+)$	0.23(10)
165.0	$(3)^-$	0.54(31)	544.0	$(2, 3)^-$	0.39(24)	924.7	$(\leq 3)^-$	0.24(10)
203.5	$(0, 1)^+$	0.08(4)	610.0	$(1, 2, 3)^-$	0.68(20)	939.7	$(1, 2, 3)$	0.33(25)
280.3	$(1, 2)^+$	0.11(5)	640.1	$(1^-, 2^-)$	0.18(9)	958.4	≤ 3	0.13(8)
292.6	$(2, 3, 4)^-$	0.05(1)	669.1	$(1^+, 2^+)$	0.64(20)	985.5	$(1, 2, 3)^+$	0.21(12)
328.5	$(3, 4)^-$	0.09(4)	681.1	$(1 - 4)$	0.33(10)	1026.2	$[1^+, 3^+]$	0.96(24)
352.4	$(3)^-$	0.05(2)	734.4	$(\leq 4)^-$	0.08(4)	1034.2	$(1, 2, 3)^+$	0.13(8)
401.8	$(1, 2)^+$	0.41(26)	751.8	$(0^-, 1, 2)$	0.37(19)	1064.5	1^+	0.23(15)
436.8	$(1, 2, 3)^-$	0.28(13)	756.6	$(0^+, 3^+)$	0.26(10)			
447.2	$(1, 2)^+$	0.46(23)	774.4	$[1^+, 3^+]$	0.23(11)			
471.0	$(2)^-$	0.05(4)	793.6	$(1, 2, 3)^+$	0.20(15)			
								$\Sigma = 11.99(105)$

^{185}Re & ^{187}Re spectra

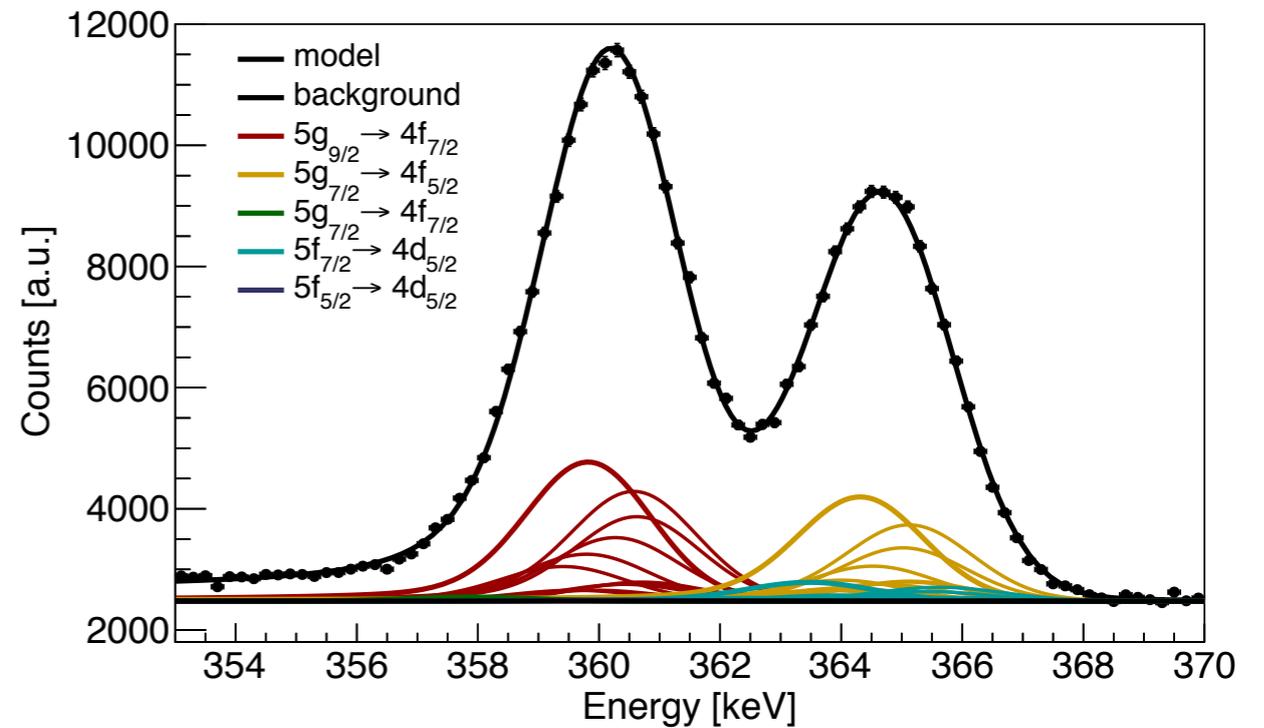
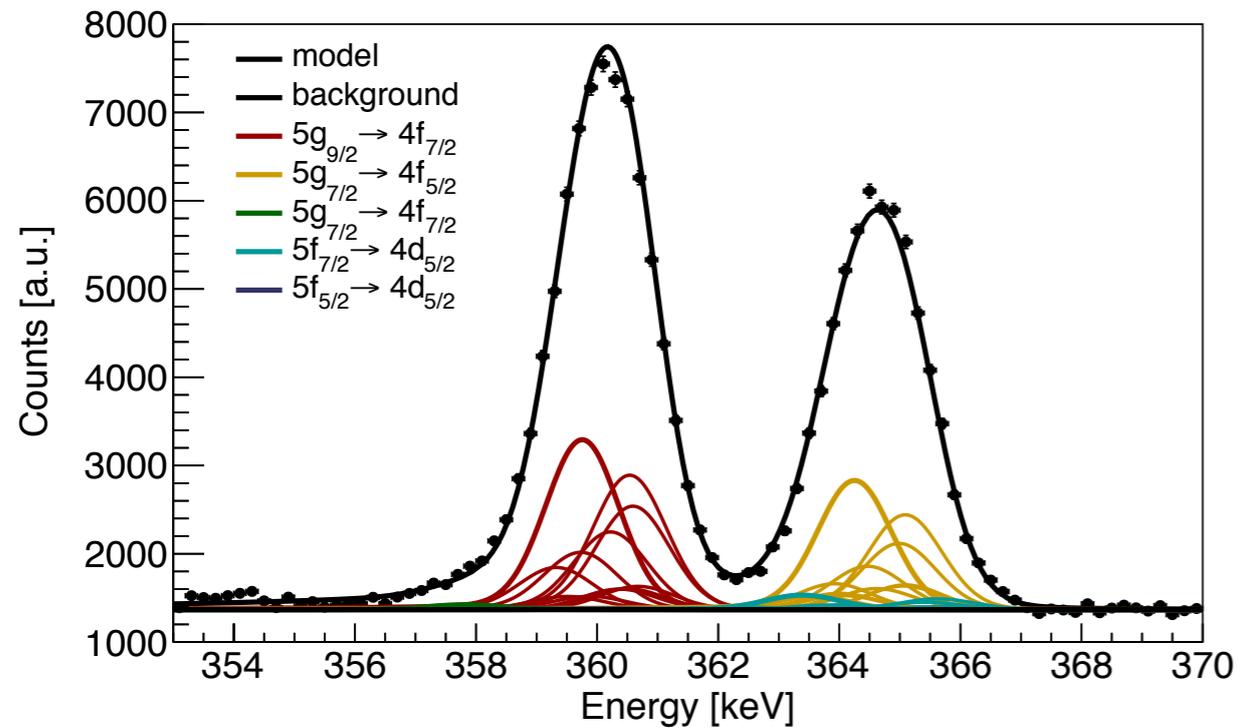
- ▶ Hyperfine structure + low-lying nuclear levels
- ▶ Highly complicated spectra
- ▶ Need very detailed theoretical calculations to extract nuclear properties



Extraction of quadrupole moments



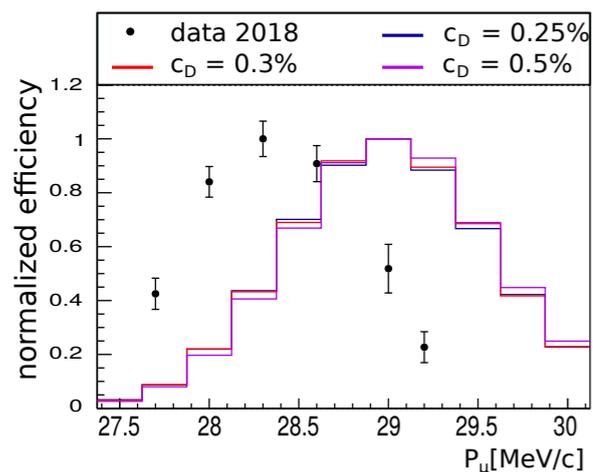
Quadrupole Moments of $^{185,187}\text{Re}$



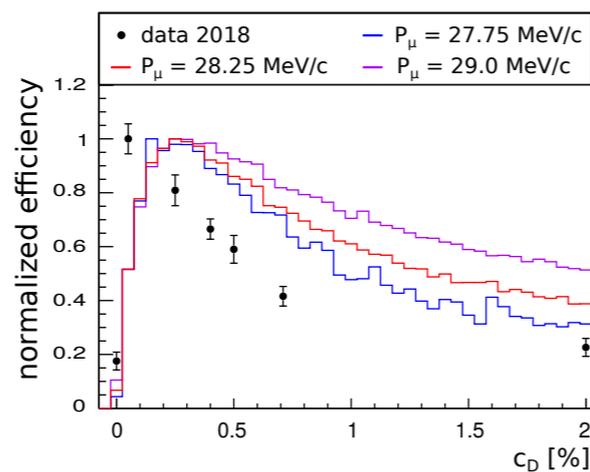
► Preliminary result on quadrupole moments of $^{185,187}\text{Re}$

Nucleus	detector	Q (barn)	χ_{red}^2	Relative intensity (%)
^{185}Re	GeR	2.12(2)	2.45	9.0(8)
	GeL	2.03(4)	1.50	14.1(7)
^{187}Re	GeR	1.97(2)	1.83	12(1)
	GeL	1.93(4)	1.14	17.0(7)

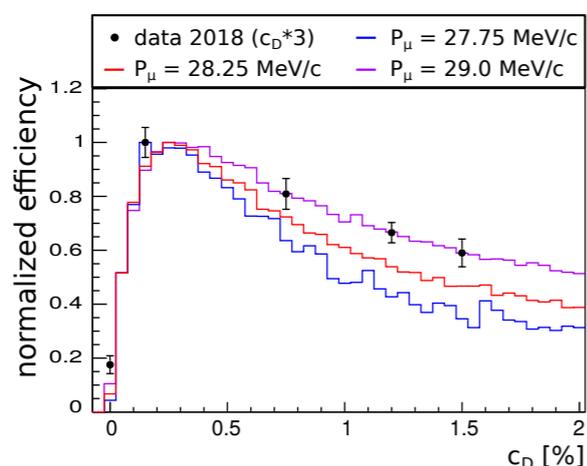
Simulations



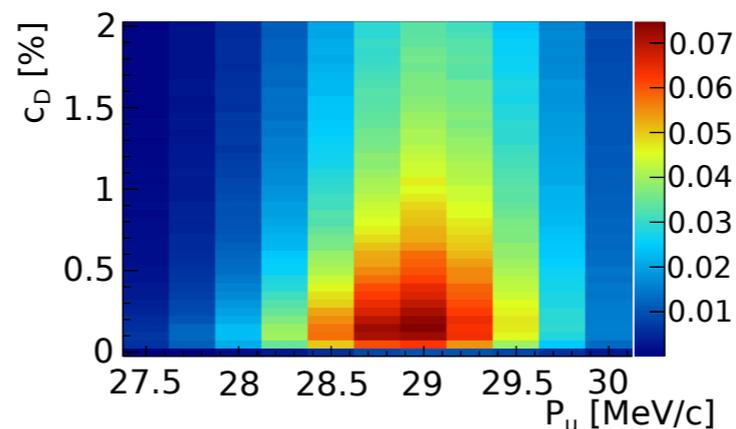
a) P_μ scan (nominal $c_D=0.25\%$)



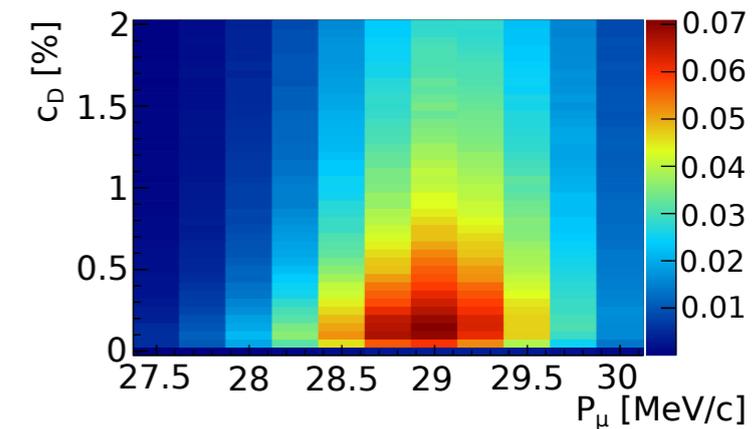
b) c_D scan ($P_\mu=28.6$ MeV/c)



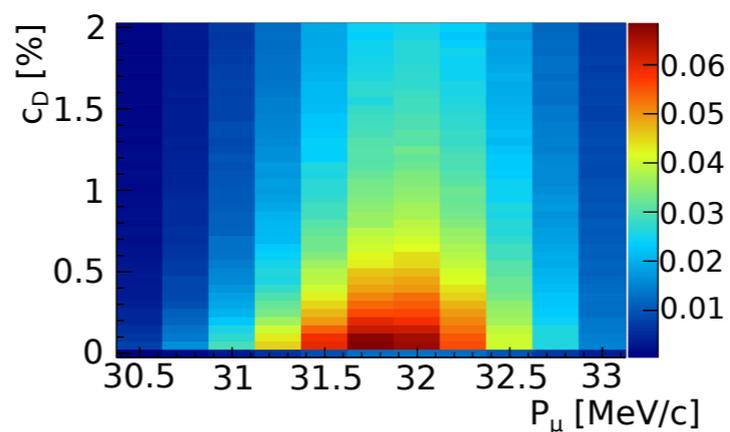
c) c_D scan, modified data



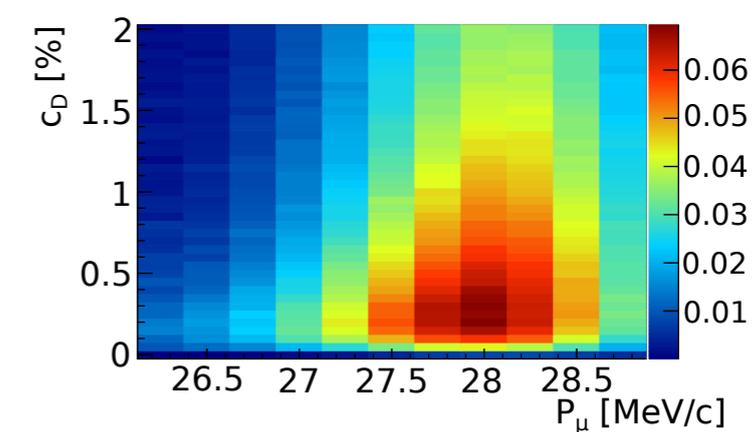
a) original muX 2018



b) $T = 50$ K, $p = 17$ bar



c) higher pressure $p=300$ bar



d) lower pressure $p=50$ bar

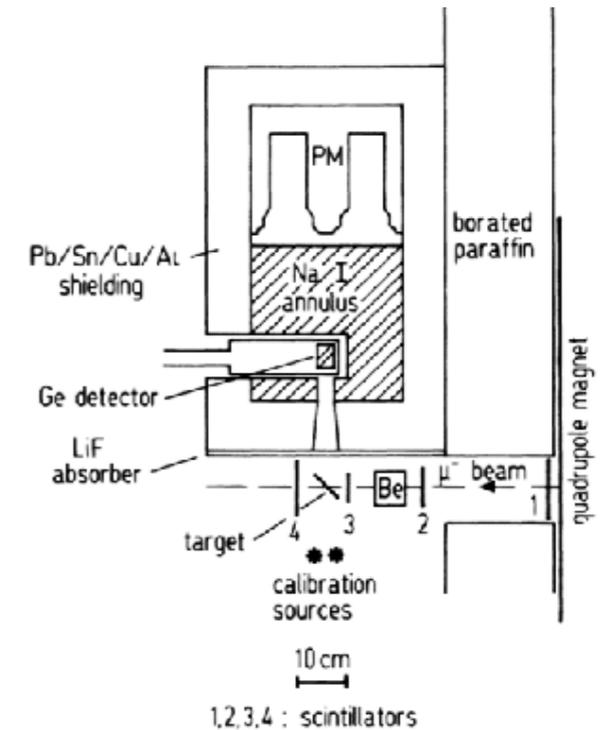
Muonic atom spectroscopy

- ▶ Impressive precision in the extracted charge radius can be achieved
- ▶ For ^{208}Pb : $\langle r^2 \rangle^{1/2} = 5.5031(11)$ fm
 2×10^{-4} relative precision

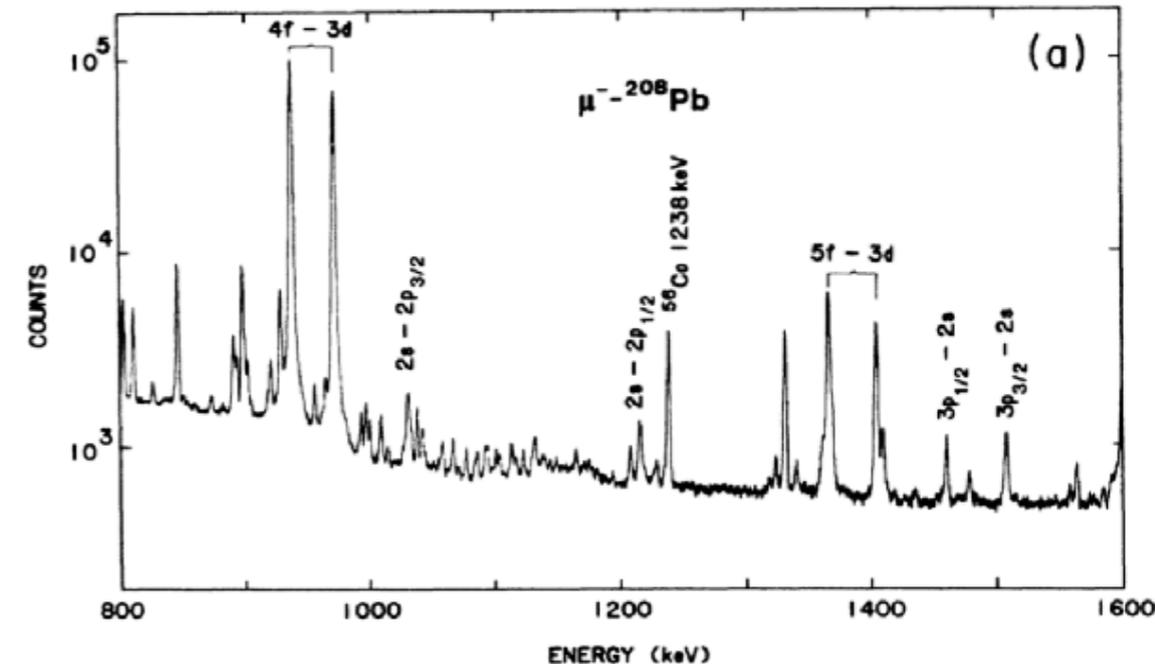
TABLE V. Experimental muonic transition energies (keV) in ^{208}Pb (recoil corrected).

Transition	Kessler (Ref. 9)	Hoehn (Ref. 27)	This experiment
$2p_{3/2}-1s_{1/2}$	5 962.770(420)		5 962.854(90)
$2p_{1/2}-1s_{1/2}$	5 777.910(400)		5 778.058(100)
$3d_{3/2}-2p_{1/2}$	2 642.110(60)	2642.292(23)	2 642.332(30)
$3d_{5/2}-2p_{3/2}$	2 500.330(60)	2500.580(28)	2 500.590(30)
$3d_{3/2}-2p_{3/2}$	2 457.200(200)		2 457.569(70)
$3p_{3/2}-2s_{1/2}$	1 507.480(260)		1 507.754(50)
$3p_{1/2}-2s_{1/2}$			1 460.558(32)
$2s_{1/2}-2p_{1/2}$	1 215.430(260)		1 215.330(30)
$2s_{1/2}-2p_{3/2}$	1 030.440(170)		1 030.543(27)
$5f_{5/2}-3d_{3/2}$	1 404.740(80)		1 404.659(20)
$5f_{7/2}-3d_{5/2}$	1 366.520(80)		1 366.347(19)
$5f_{5/2}-3d_{5/2}$			1 361.748(250)
$4f_{5/2}-3d_{3/2}$	971.850(60)	971.971(16)	971.974(17)
$4f_{7/2}-3d_{5/2}$	937.980(60)	938.113(13)	938.096(18)
$4f_{5/2}-3d_{5/2}$			928.883(14)
$4d_{3/2}-3p_{1/2}$			920.959(28)
$4d_{5/2}-3p_{3/2}$			891.383(22)
$4d_{3/2}-3p_{3/2}$			873.761(63)

Bergem et al., PRC 37, 2821 (1988)

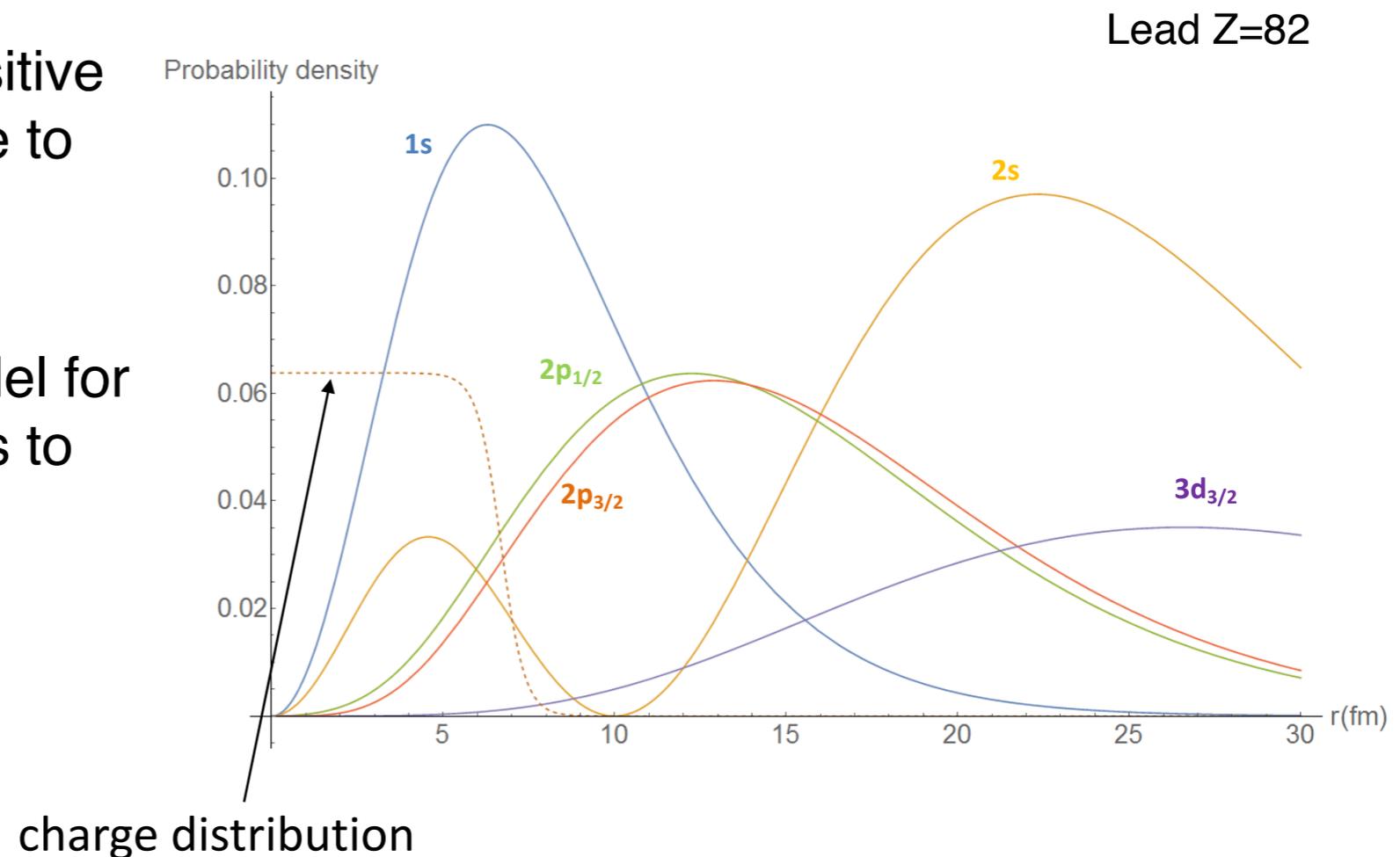


μE1 channel at PSI
 $5 \times 10^6 \mu/\text{s}$ at 125 MeV/c



Muonic atom spectroscopy

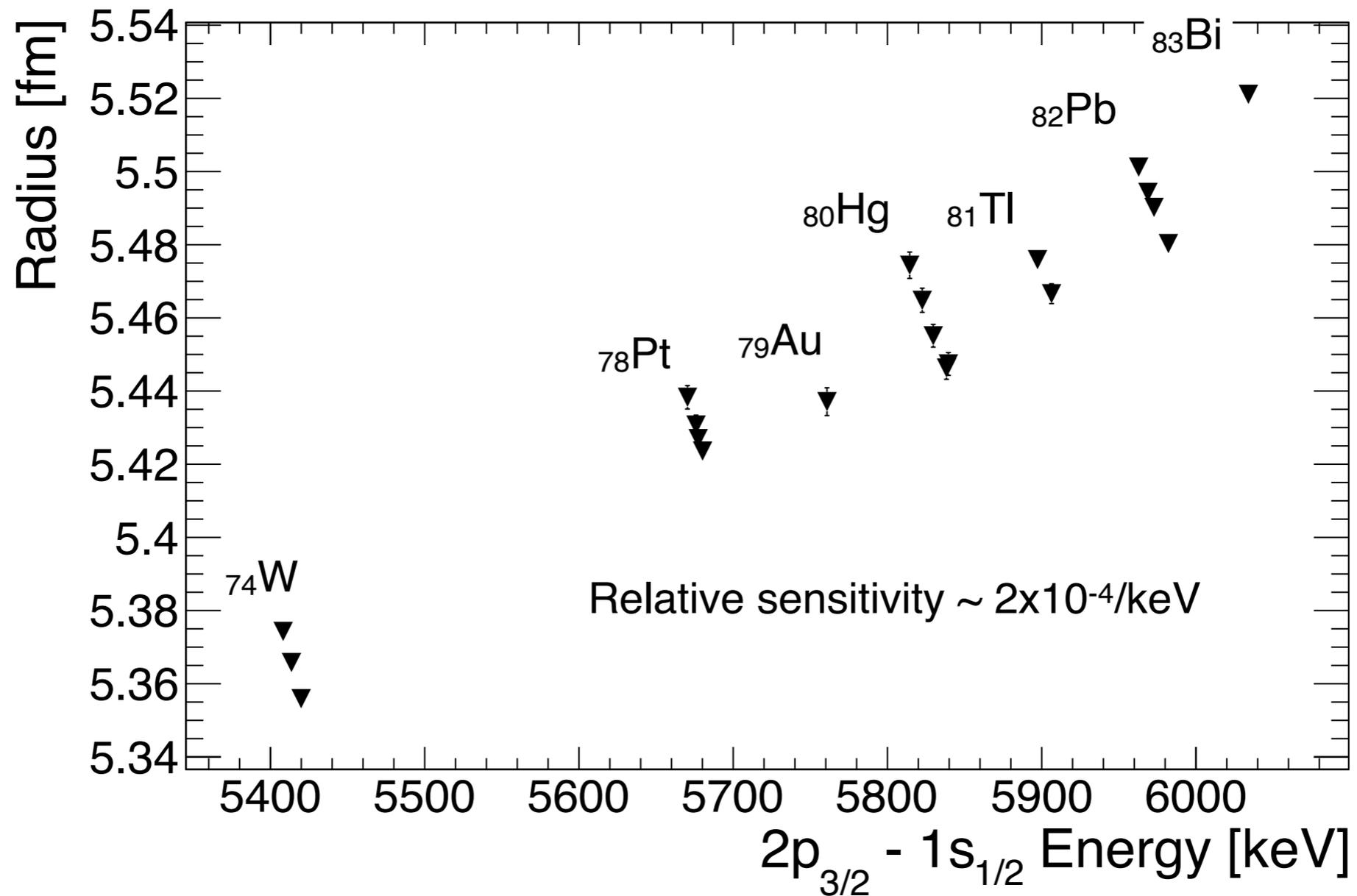
- ▶ Muonic energy levels highly sensitive to nuclear charge distribution due to large overlap
- ▶ Using QED calculations and model for nuclear charge distribution allows to extract charge radius



Large effect:

E_{1s} (Z=82) \sim 19 MeV (point nucleus)
 \rightarrow 10.6 MeV (finite size)

Muonic atom spectroscopy



- ▶ $2p - 1s$ energy is highly sensitive to charge radius
- ▶ What is the limiting factor?

Muonic atom spectroscopy

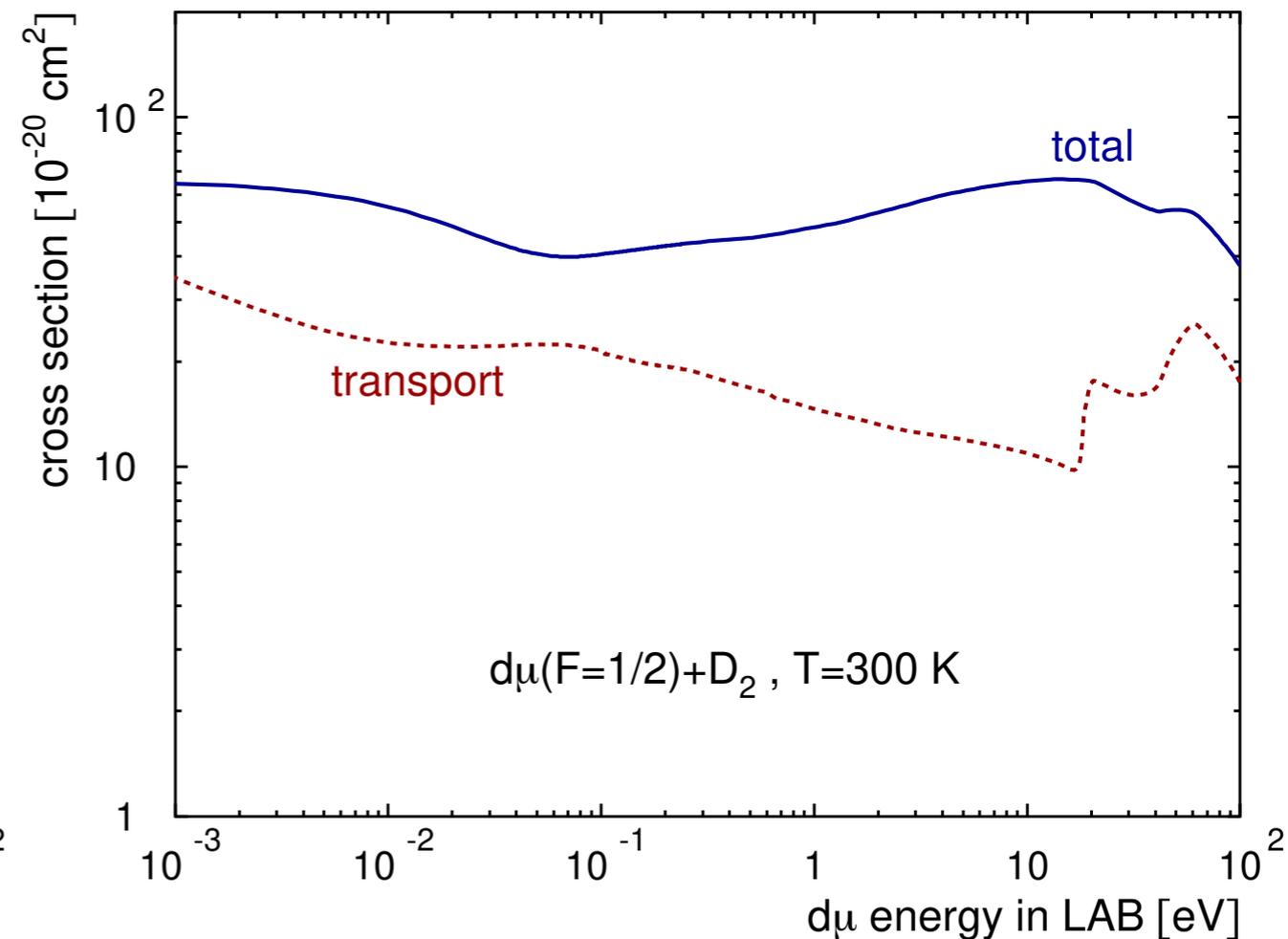
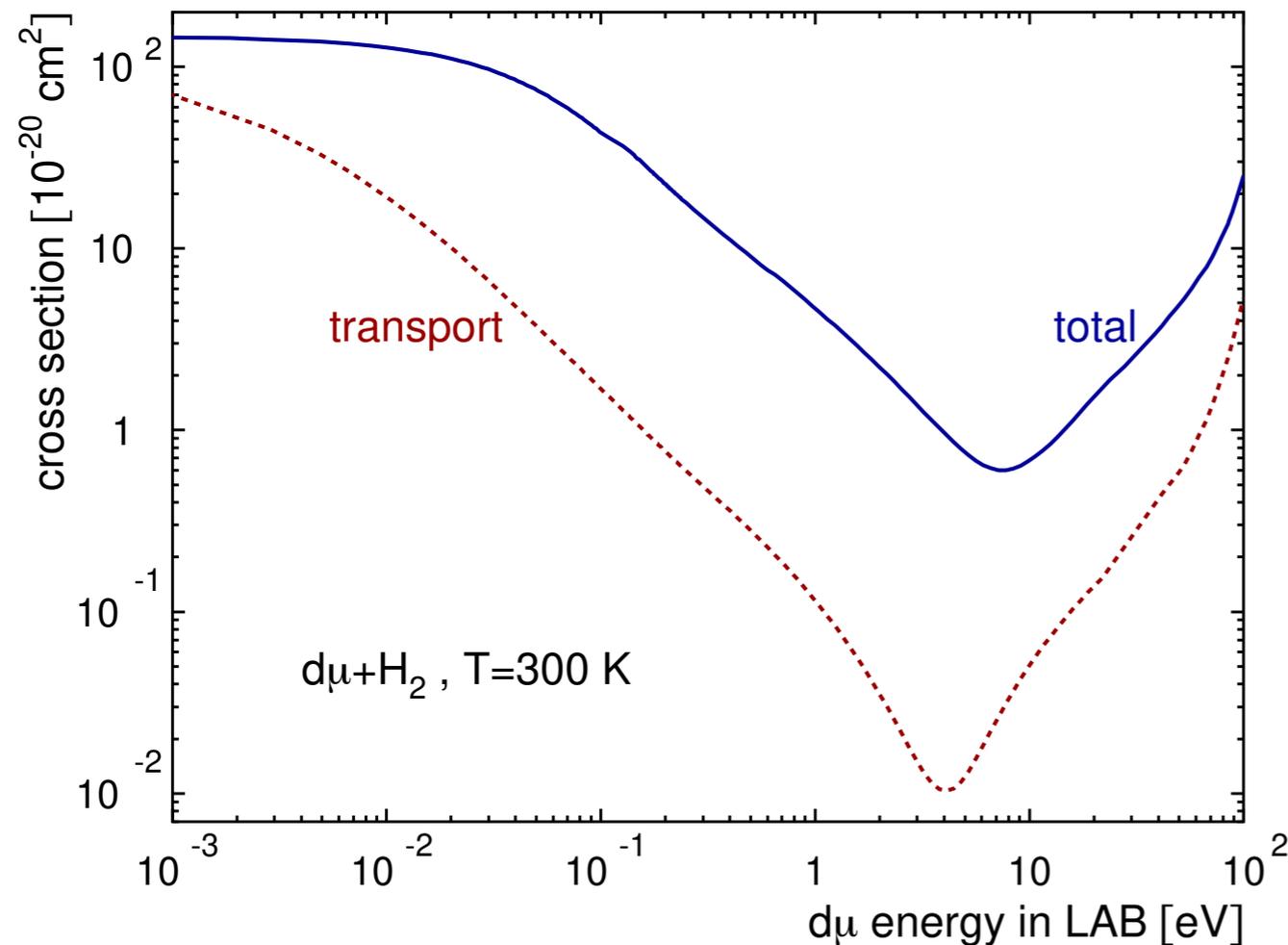
- ▶ Nuclear polarisation is the dominating factor that in the end determines the accuracy of the extracted charge radius
- ▶ Typically assumed uncertainty: 10 - 30%
- ▶ Nuclear excitation spectra important
- ▶ Looking for theorists that want to tackle these calculations with modern methods

TABLE II. Theoretical nuclear polarization corrections in ^{208}Pb .

Energy (MeV)	I^π	$B(E\lambda)^\dagger$ ($e^2b^{2\lambda}$)	$1s_{1/2}$ (eV)	$2s_{1/2}$ (eV)	$2p_{1/2}$ (eV)	$2p_{3/2}$ (eV)	$3p_{1/2}$ (eV)	$3p_{3/2}$ (eV)	$3d_{3/2}$ (eV)	$3d_{5/2}$ (eV)
2.615	3^-	0.612	135	12	90	84	26	26	111	-63
4.085	2^+	0.318	198	20	182	180	76	84	6	4
4.324	4^+	0.155	14	1	8	7	2	2	1	1
4.842	1^-	0.001 56	7	1	-9	-8	0	0	1	1
5.240	3^-	0.130	27	2	16	15	5	5	2	2
5.293	1^-	0.002 04	9	2	-27	-19	0	-1	1	1
5.512	1^-	0.003 80	16	3	-90	-53	-1	-1	1	1
5.946	1^-	0.000 07	0	0	3	-30	0	0	0	0
6.193	2^+	0.050 5	29	3	22	21	7	7	0	0
6.262	1^-	0.000 24	1	0	3	5	0	0	0	0
6.312	1^-	0.000 22	1	0	3	4	0	0	0	0
6.363	1^-	0.000 14	1	0	2	2	0	0	0	0
6.721	1^-	0.000 75	3	1	6	7	0	-1	0	0
7.064	1^-	0.001 56	6	1	9	11	-1	-1	0	0
7.083	1^-	0.000 75	3	1	4	5	-1	-1	0	0
7.332	1^-	0.002 04	8	1	10	11	-2	-2	0	0
Total low-lying states			458	48	233	242	111	117	123	-53
13.5	0^+	0.047 872	906	315	64	38	24	15	1	0
22.8	0^+	0.043 658	546	147	43	26	15	10	0	0
13.7	1^-	0.537 672	1454	221	786	738	255	258	66	54
10.6	2^+	0.761 038	375	37	237	222	67	68	33	30
21.9	2^+	0.566 709	207	21	108	99	29	29	8	7
18.6	3^-	0.497 596	77	7	40	36	11	11	3	2
33.1	3^-	0.429 112	53	5	25	23	7	7	2	1
	$> 3^a$		176	15	80	71	21	21	4	4
Total high-lying states			3794	768	1383	1253	429	419	117	98
Total			4252	816	1616	1495	540	536	240	45

^aValues from Ref. 7. Positive NP values mean that the respective binding energies are increased.

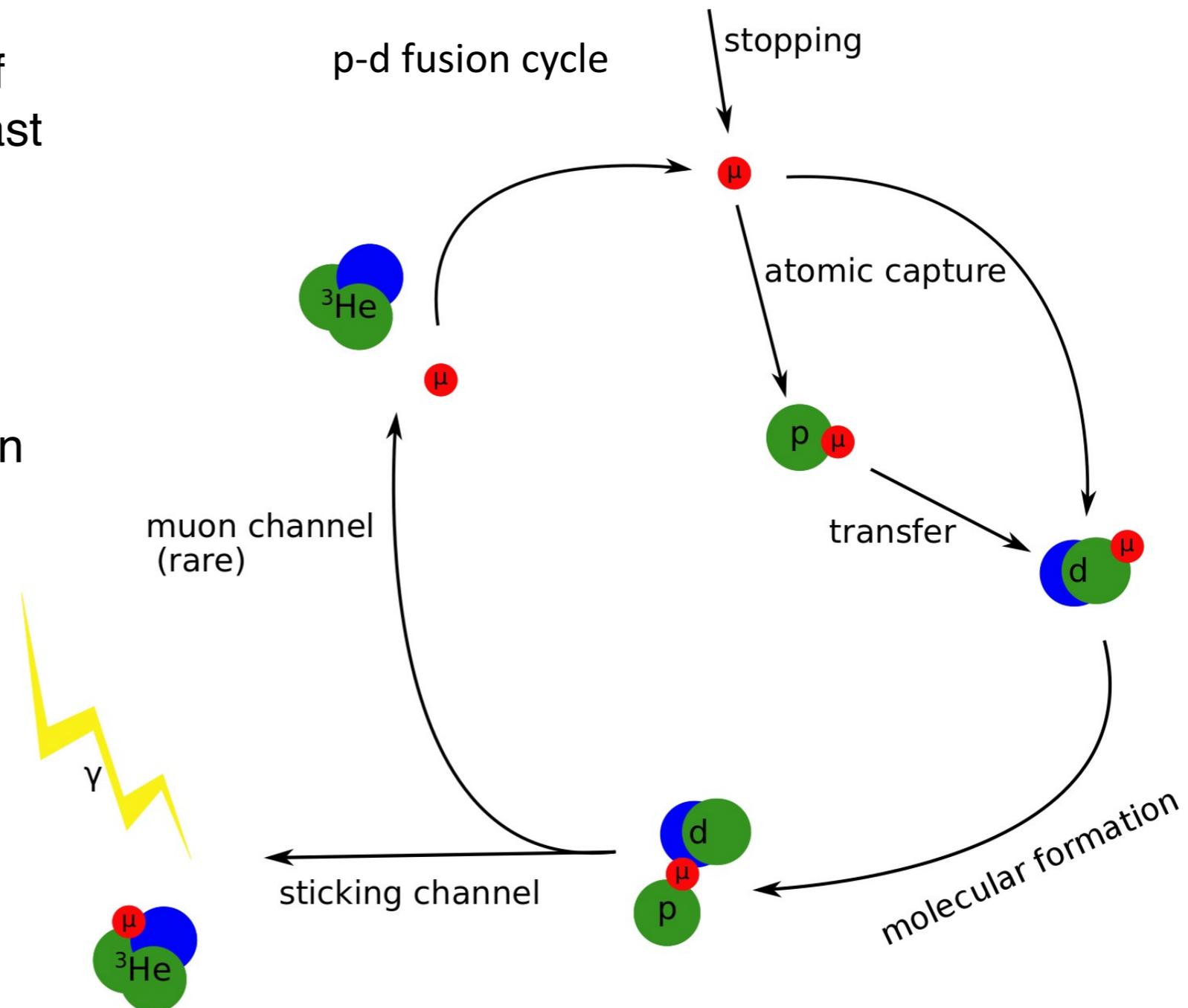
Scattering cross sections



- Scattering on deuterium does not show a Ramsauer-Townsend minimum
- Need to be careful to not have too much deuterium in the gas mixture

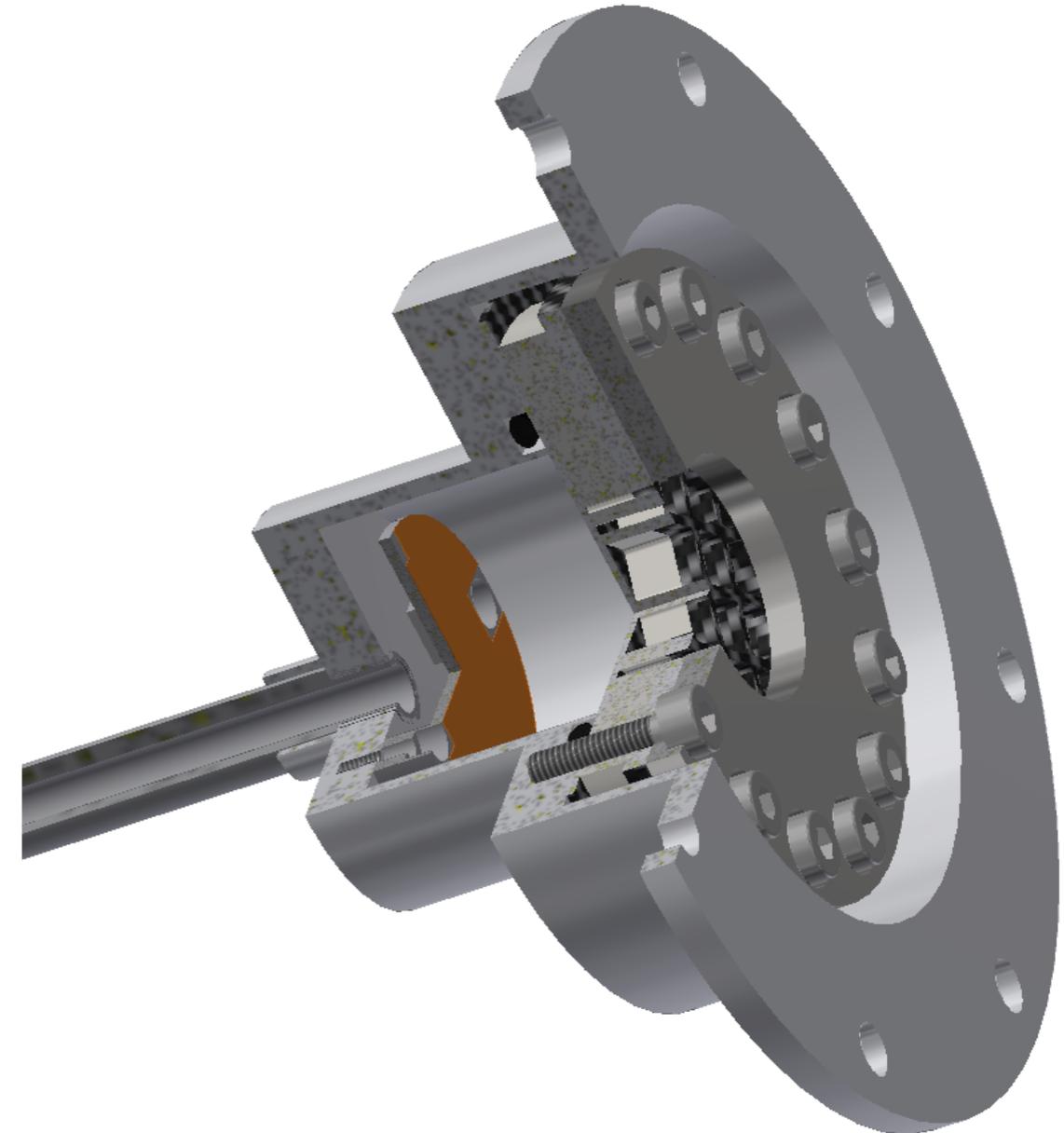
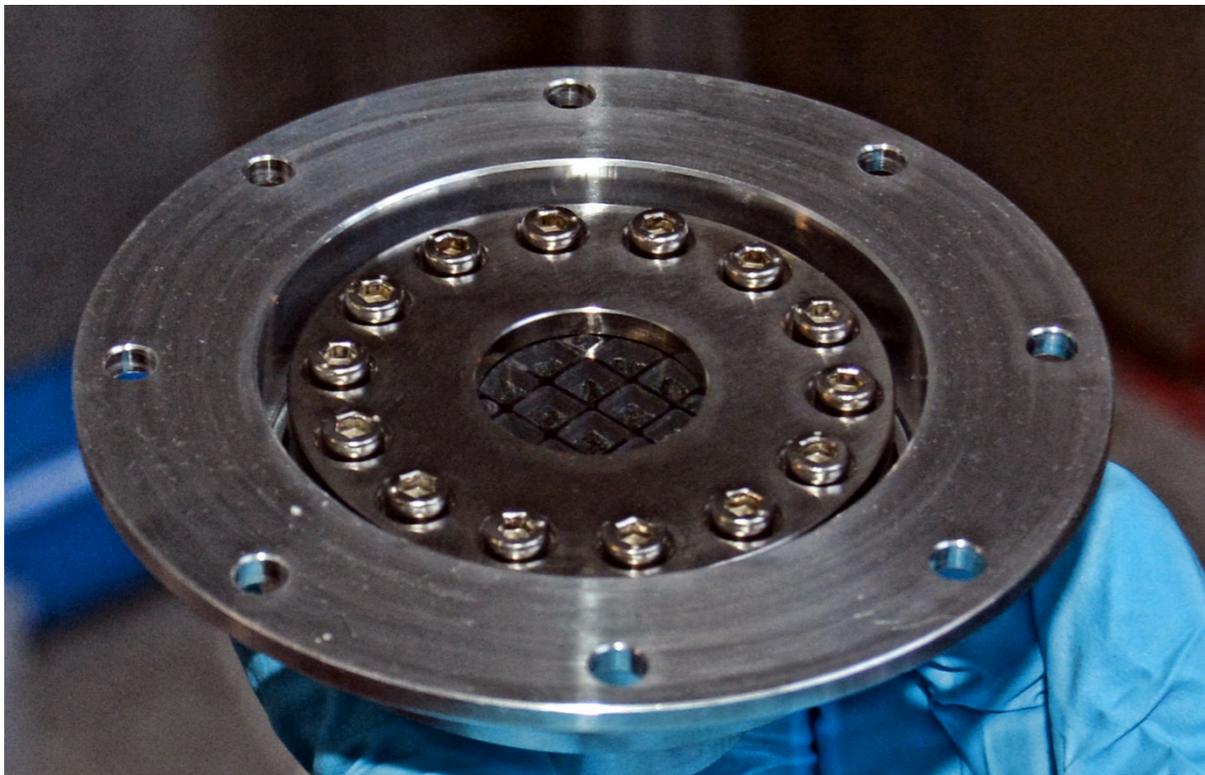
Muon catalysed fusion

- ▶ Lot of experience on behaviour of muons in hydrogen gas due to past work on muon catalysed fusion
- ▶ Most efficient cycle: d-t fusion, up to 150 fusions per muon
- ▶ Not enough for energy break-even



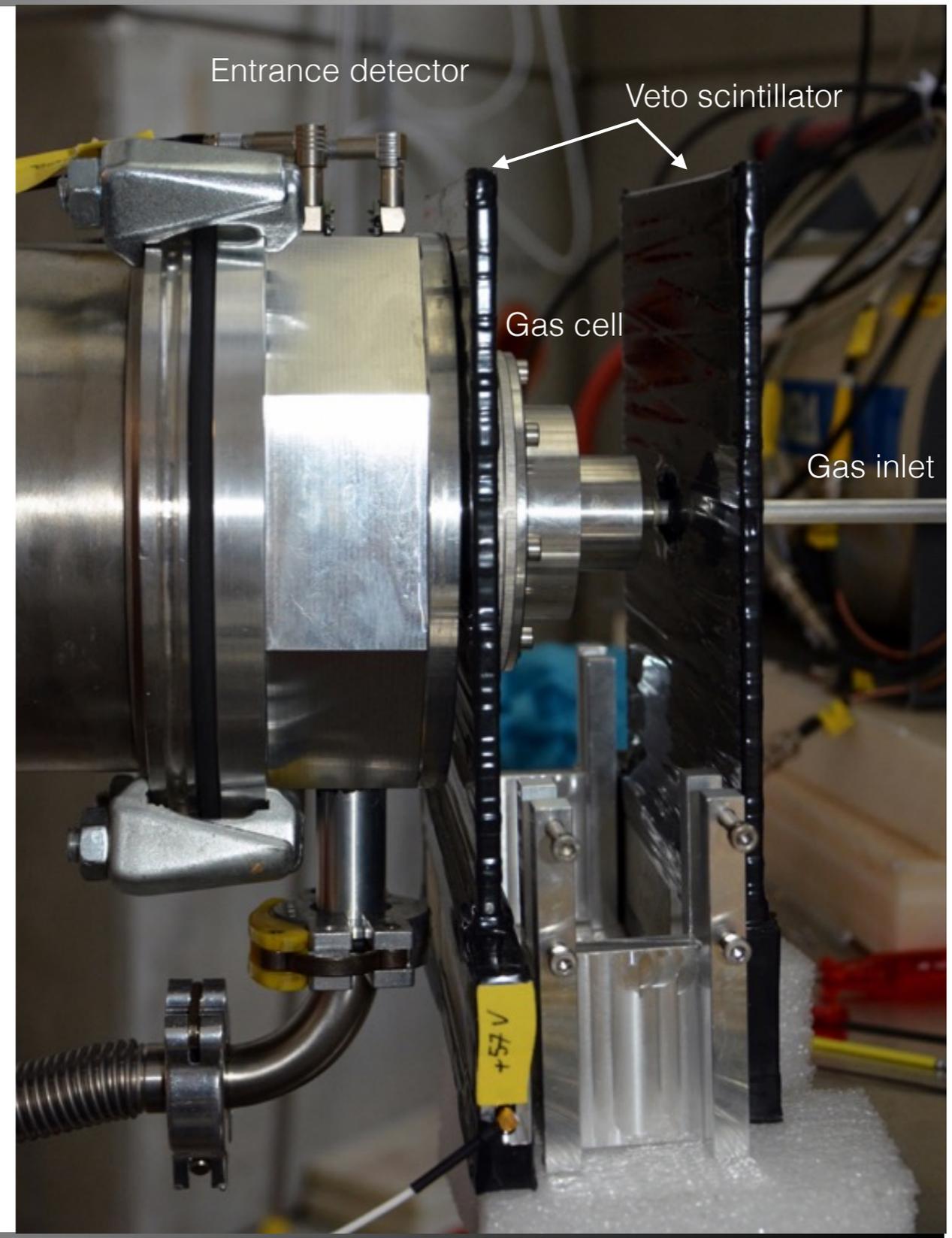
100 bar hydrogen target

- ▶ Target sealed with 0.6 mm carbon fibre window plus carbon fibre/titanium support grid
- ▶ Target holds up to 350 bar
- ▶ 10 mm stopping distribution (FWHM) inside 15 mm gas volume
- ▶ Target disks mounted onto the back of the cell



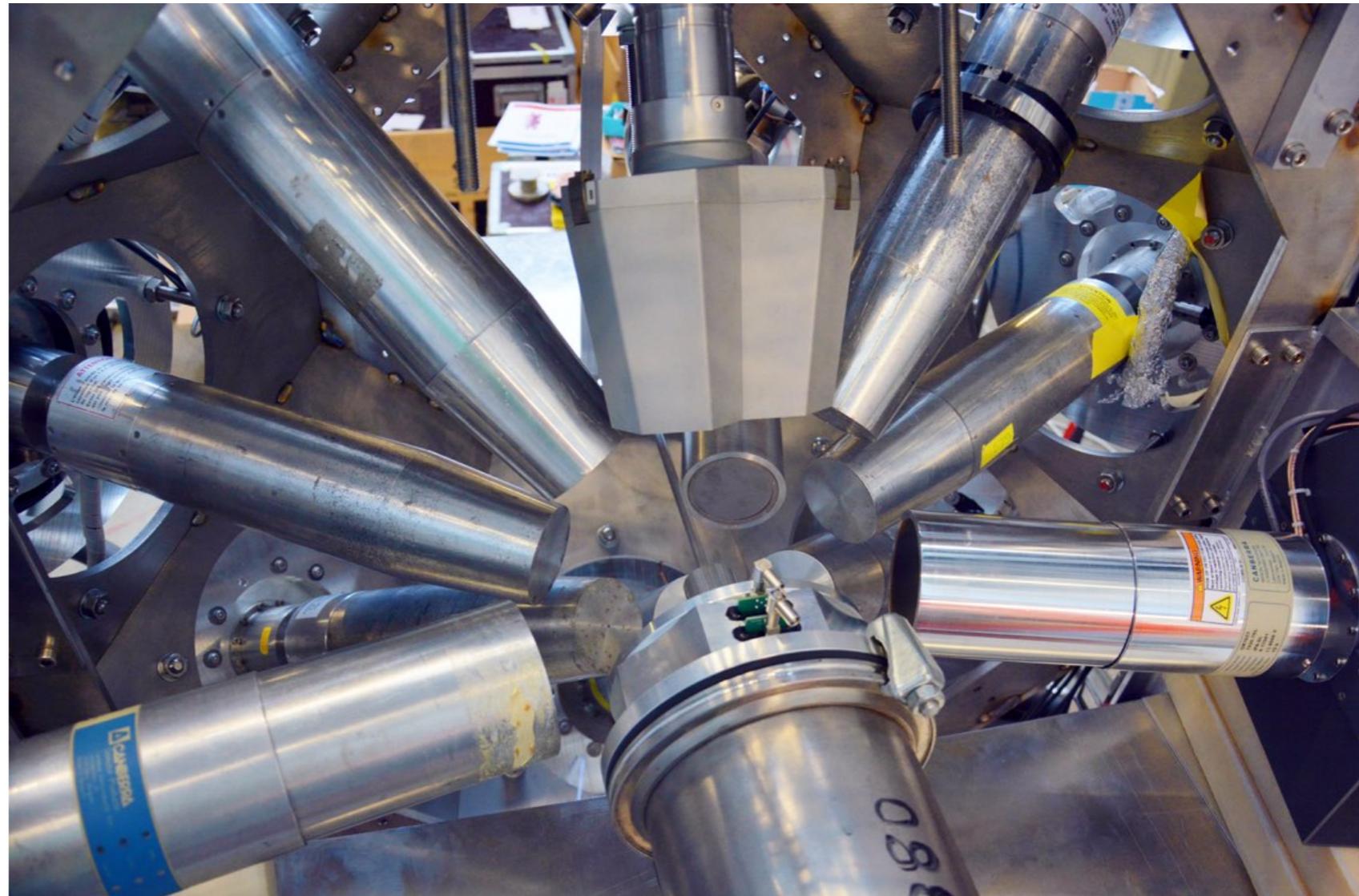
Entrance & veto detectors

- ▶ Entrance detector to see incoming muon
- ▶ Veto scintillators to form anti-coincidence with decay electron

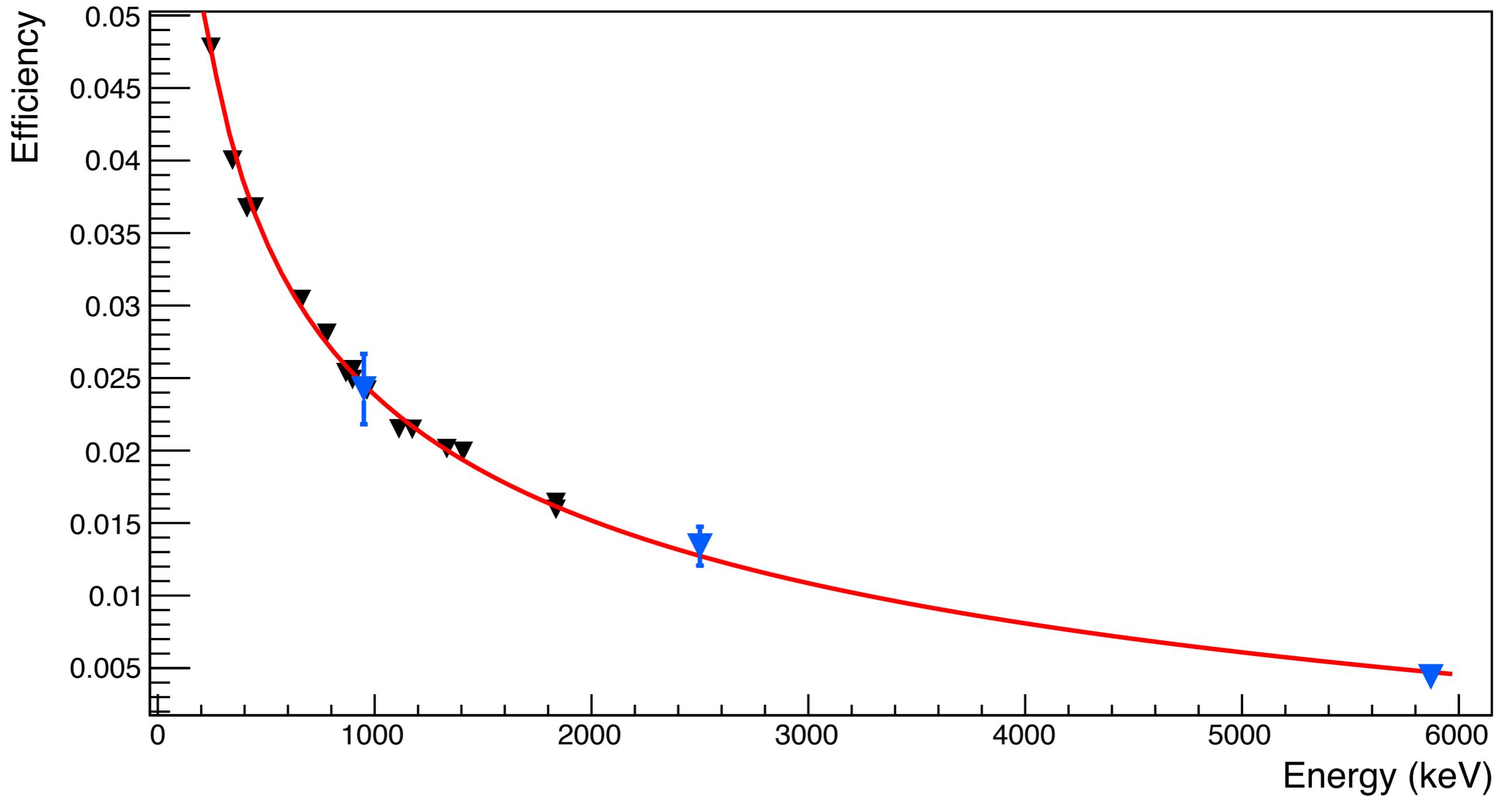


Germanium array

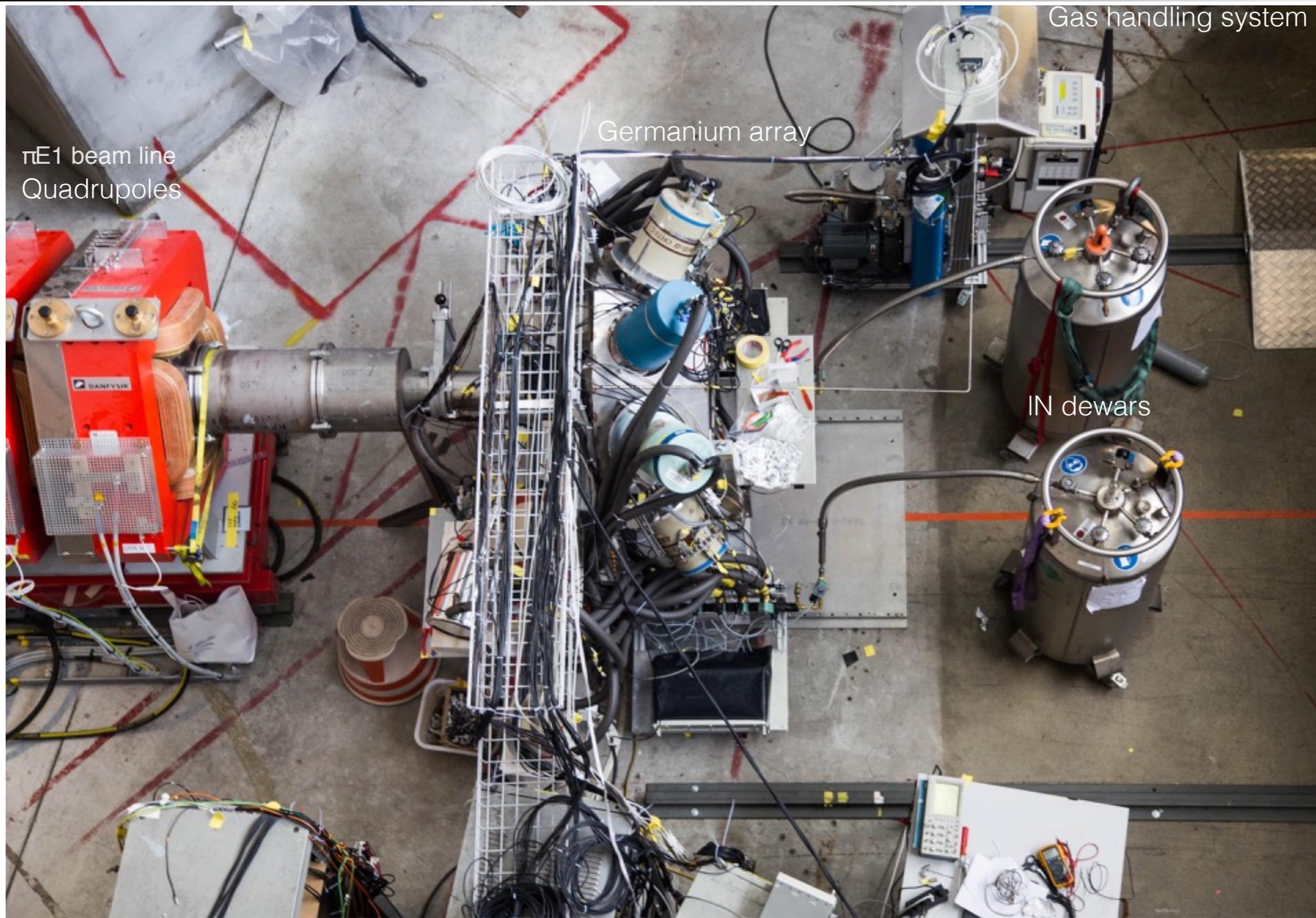
- ▶ 11 germanium detectors in an array from French/UK loan pool, Leuven, PSI
- ▶ First time a large array is used for muonic atom spectroscopy



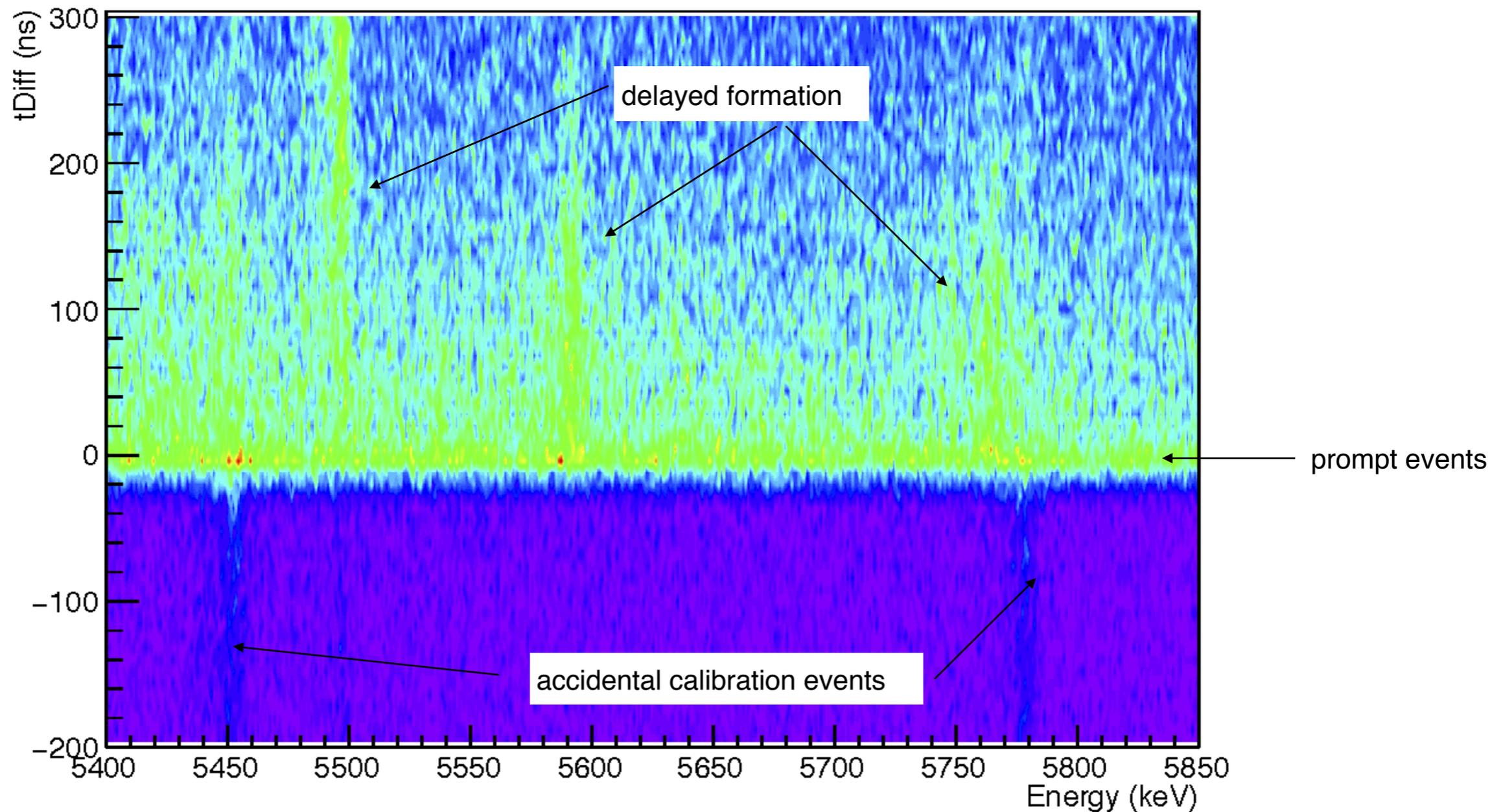
Array Detection Efficiency



Experimental setup 2017/2018

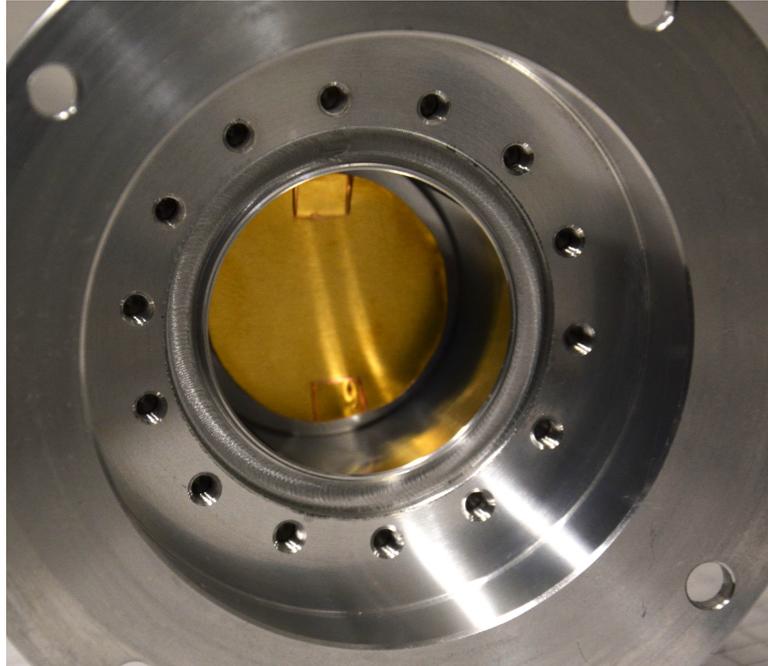


Energy vs. time spectra



- ▶ DAQ is free-running and recording every detector with a timestamp
- ▶ Sorting germanium detector hits in time after muon entrance hit

Understanding target conditions

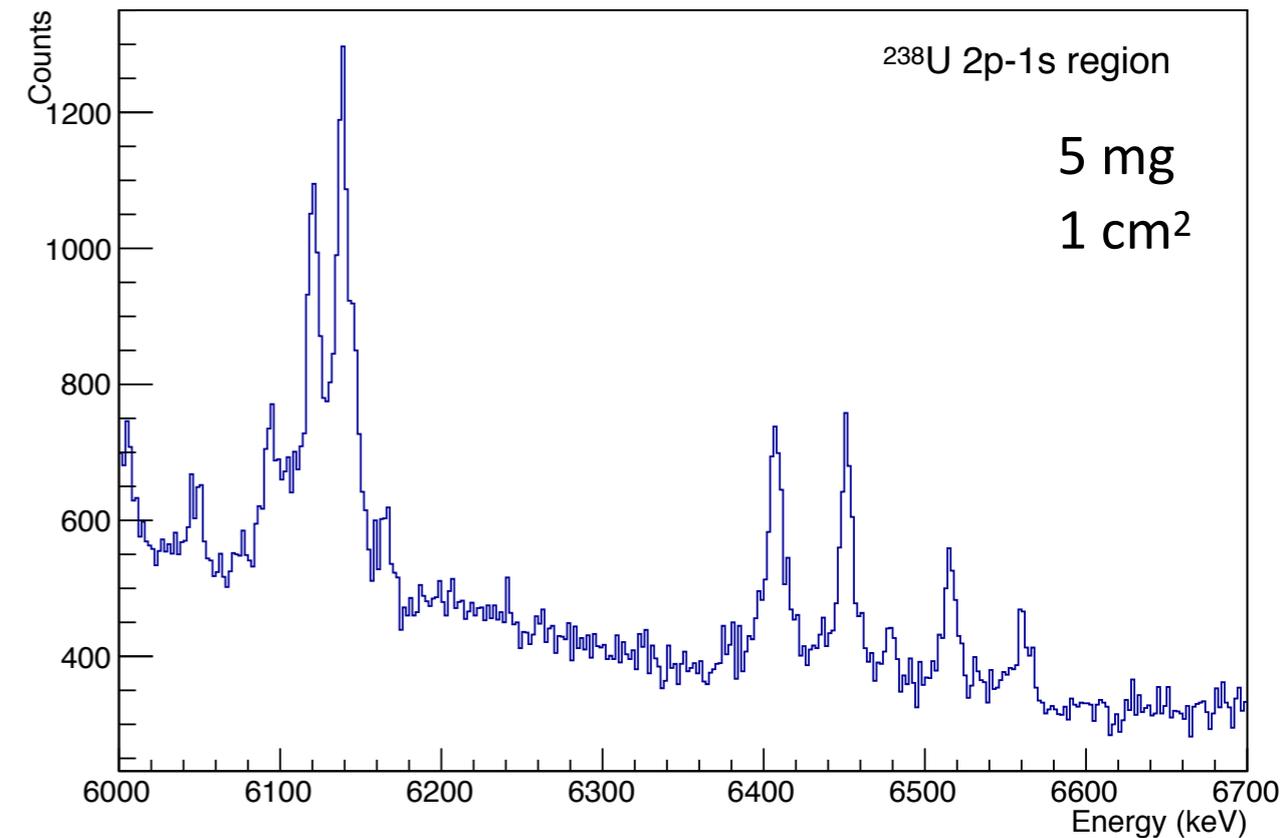


Target	Size	Backing	N_γ / N_μ	ϵ
50 nm Au	4.9 cm ²	Cu	$(10.9 \pm 0.3) \times 10^{-5}$	10.0%
10 nm Au	4.9 cm ²	Cu	$(6.9 \pm 0.2) \times 10^{-5}$	6.3%
3 nm Au	4.9 cm ²	Cu	$(3.6 \pm 0.1) \times 10^{-5}$	3.3%
3 nm Au	4.9 cm ²	kapton	$(3.2 \pm 0.1) \times 10^{-5}$	2.9%
3 nm Au	1 cm ²	Cu	$(1.3 \pm 0.1) \times 10^{-5}$	1.2%

- ▶ Detected 2p-1s gammas per incoming muon for various targets
- ▶ Not all μd converted in thin targets
- ▶ Impact of backing material small
- ▶ Can still reliably see gammas from 5 μg gold target (1 cm², 3 nm)

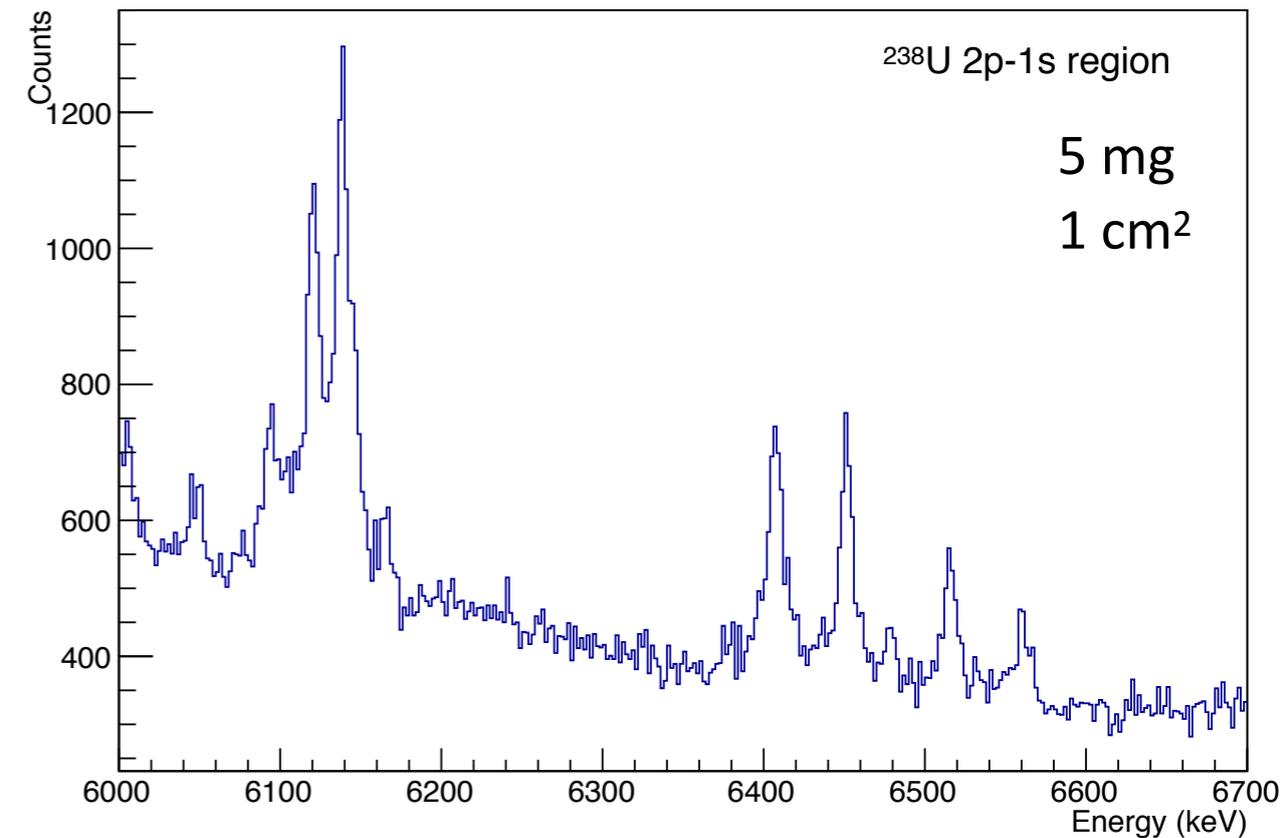
Measurement with uranium

- ▶ Measurement with ~5 mg uranium as a test for handling radioactive materials in our setup
- ▶ Complicated spectrum due to hyperfine splitting plus low-lying nuclear excitations
- ▶ ^{226}Ra will look very similar

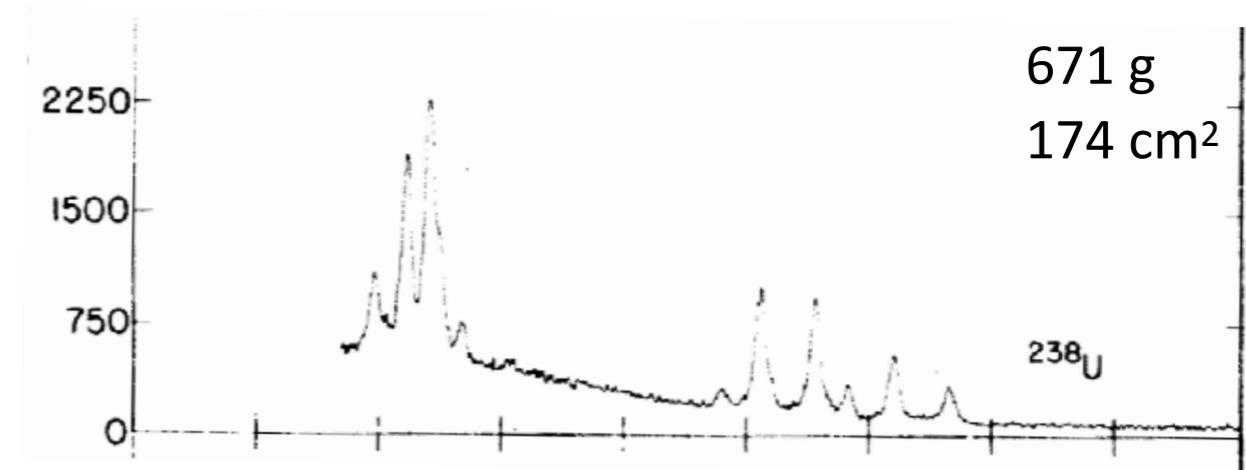


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Close et al., Phys. Rev. C 17, 1433 (1978)

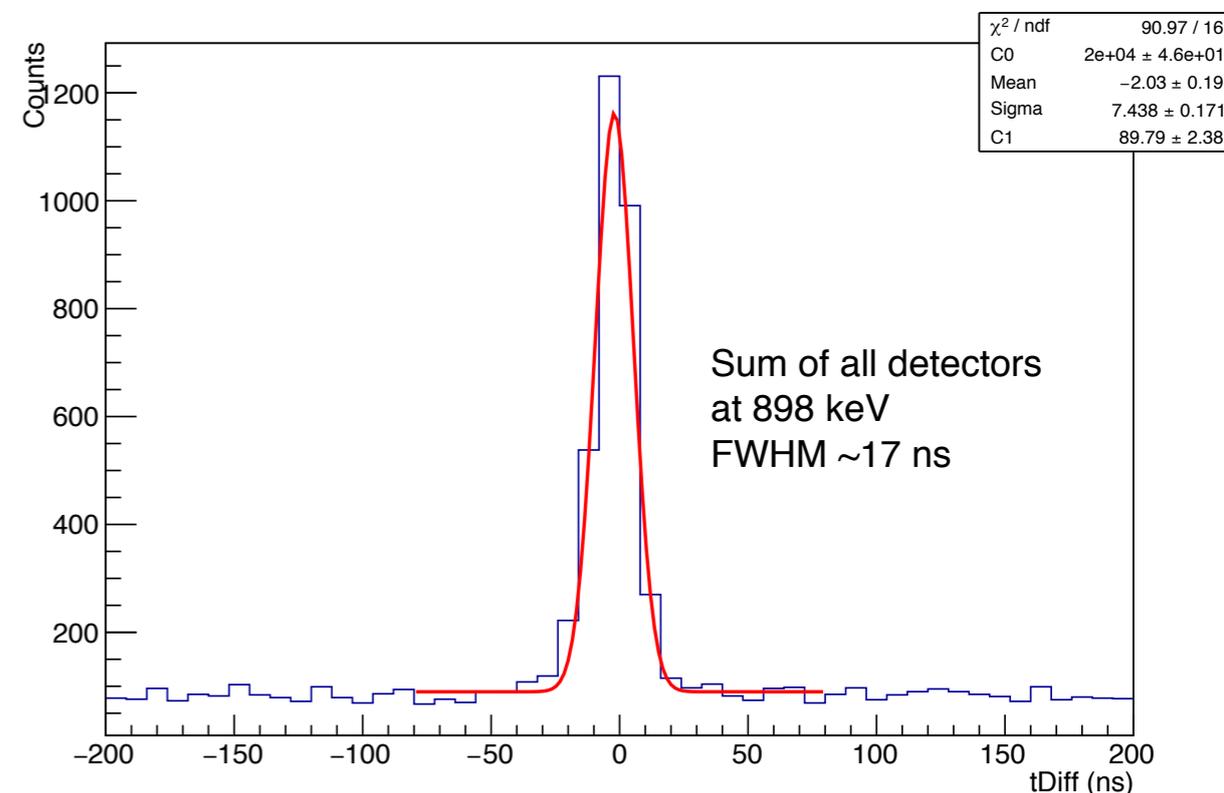
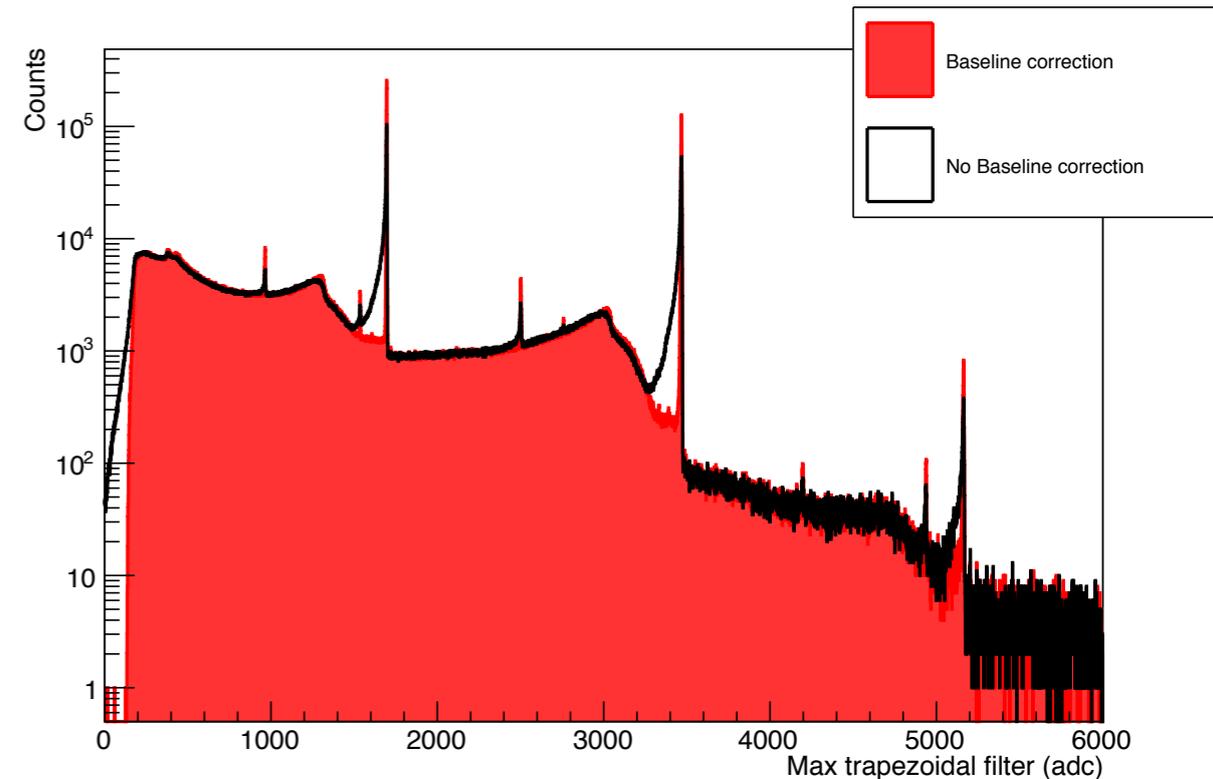


Similar performance as in the past
but a factor 10^5 less target material



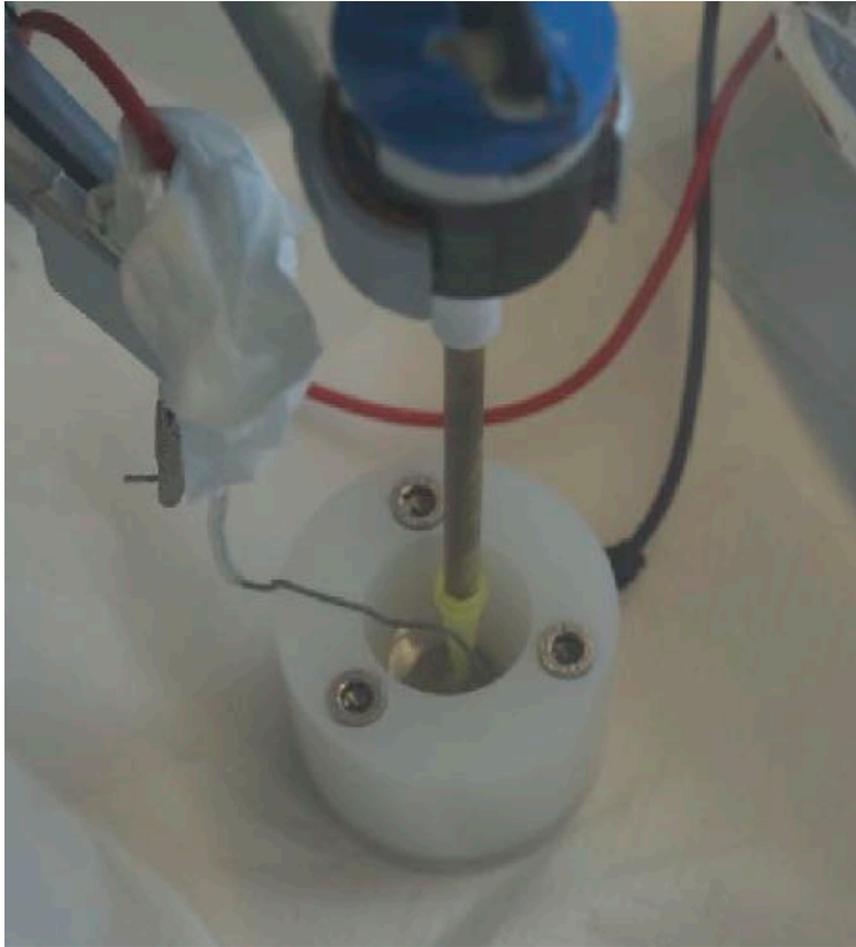
Measurement with high rates

- ▶ Performed measurements with strong ^{88}Y source producing 420 kHz gammas comparable to ^{226}Ra target
- ▶ Able through offline analysis to improve energy and time resolution
- ▶ DAQ able to cope with data rate



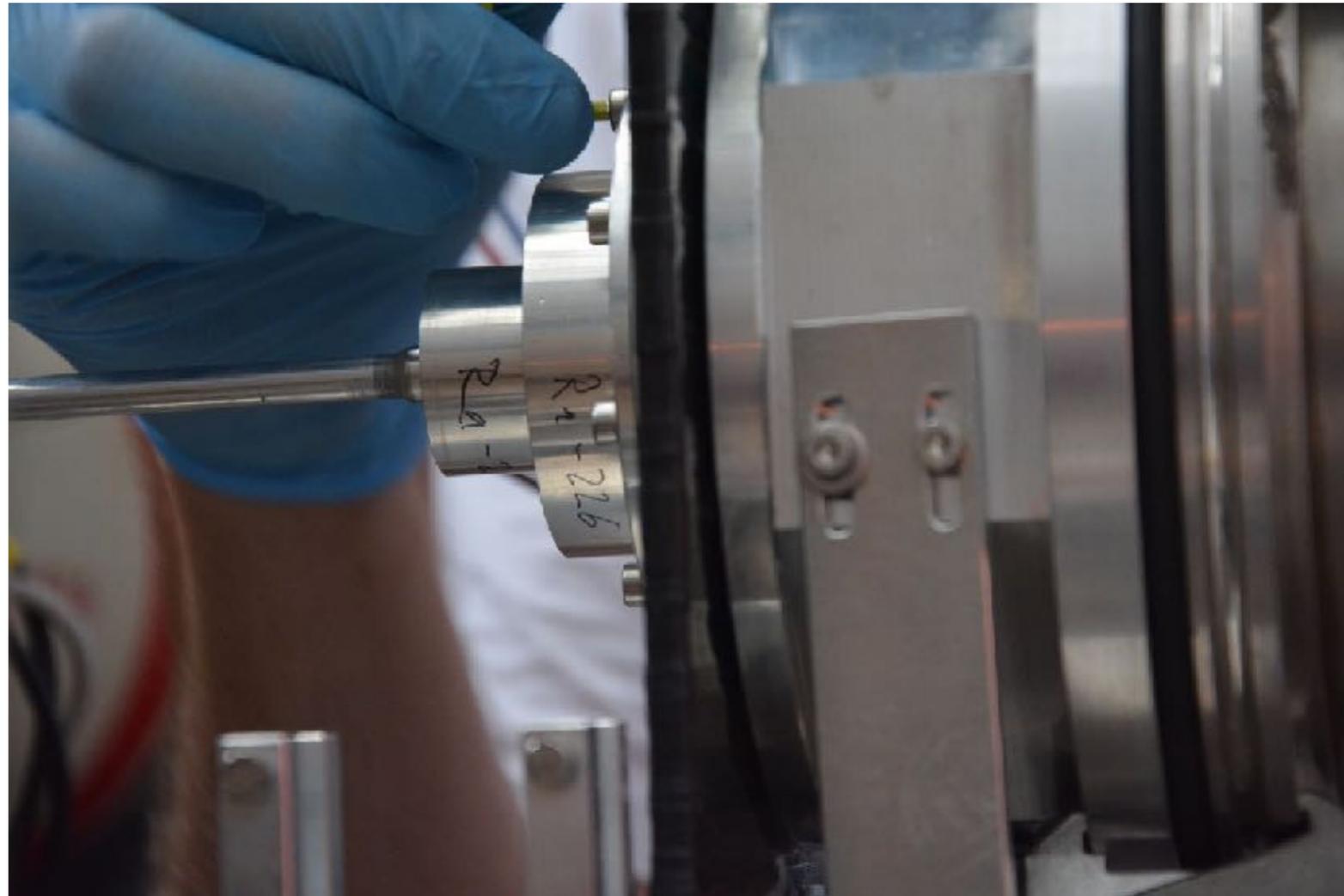
Experiment is ready for measurements with radioactive targets!

Making radium target



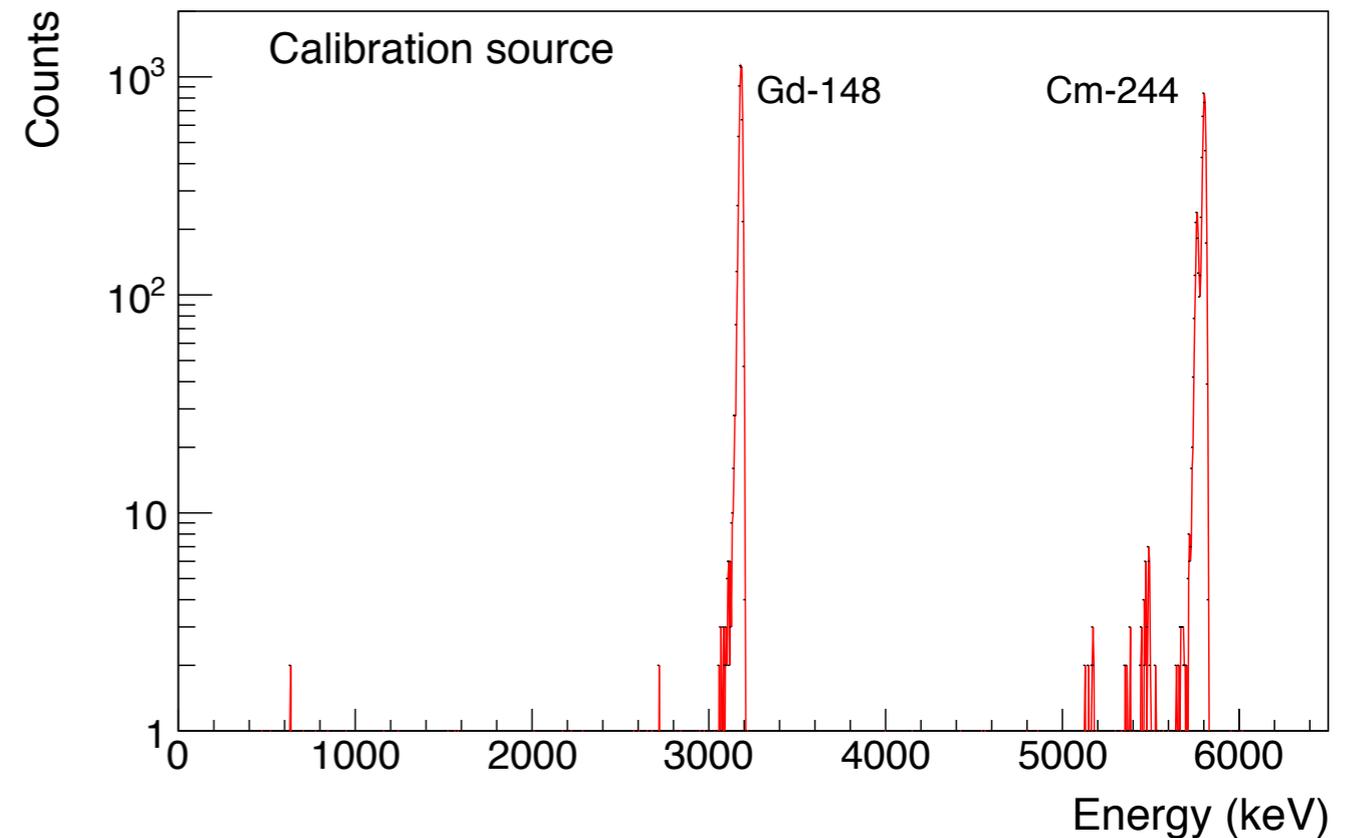
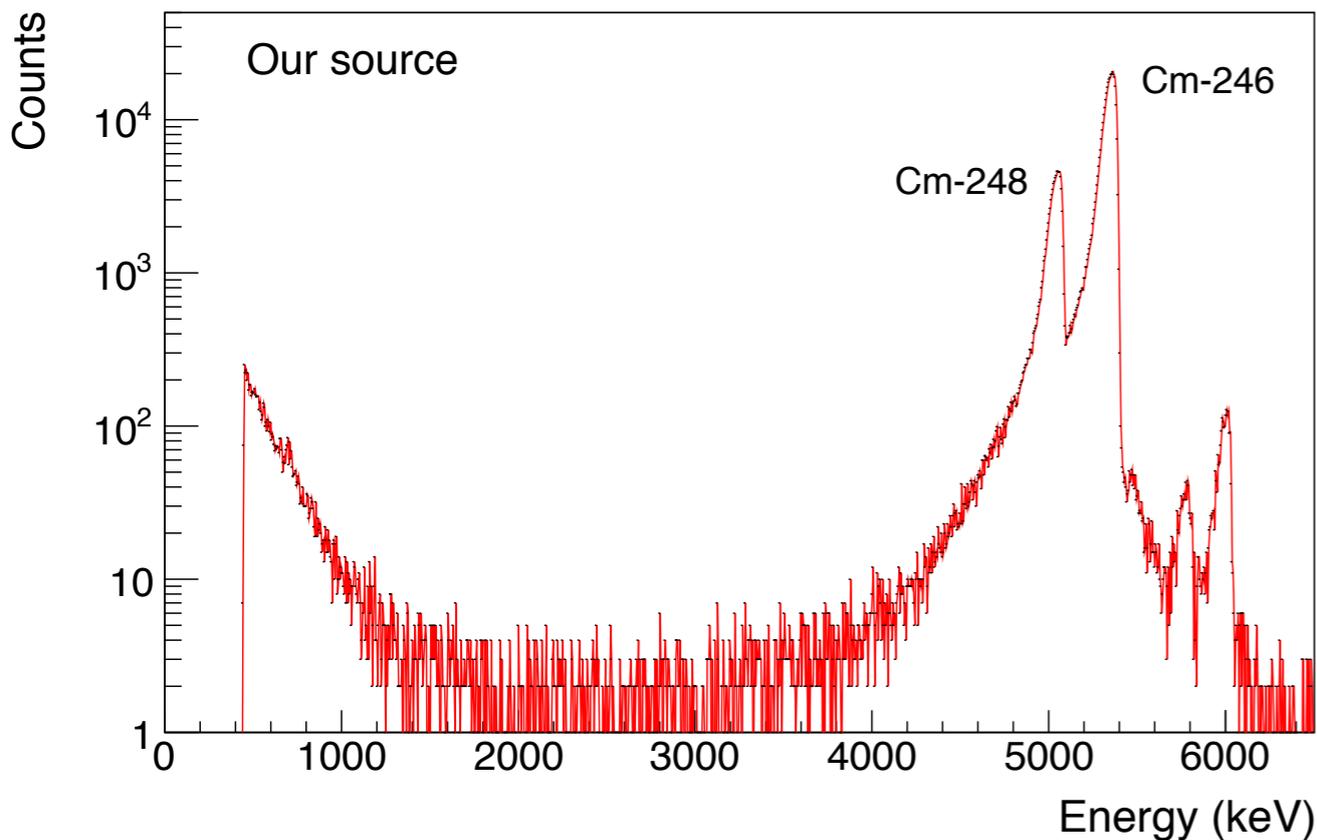
- ▶ Attempted a measurement of ^{226}Ra and ^{248}Cm this year
- ▶ Electroplating the ^{226}Ra out of the isopropanol solution onto gold plated copper foil

Measuring radium target



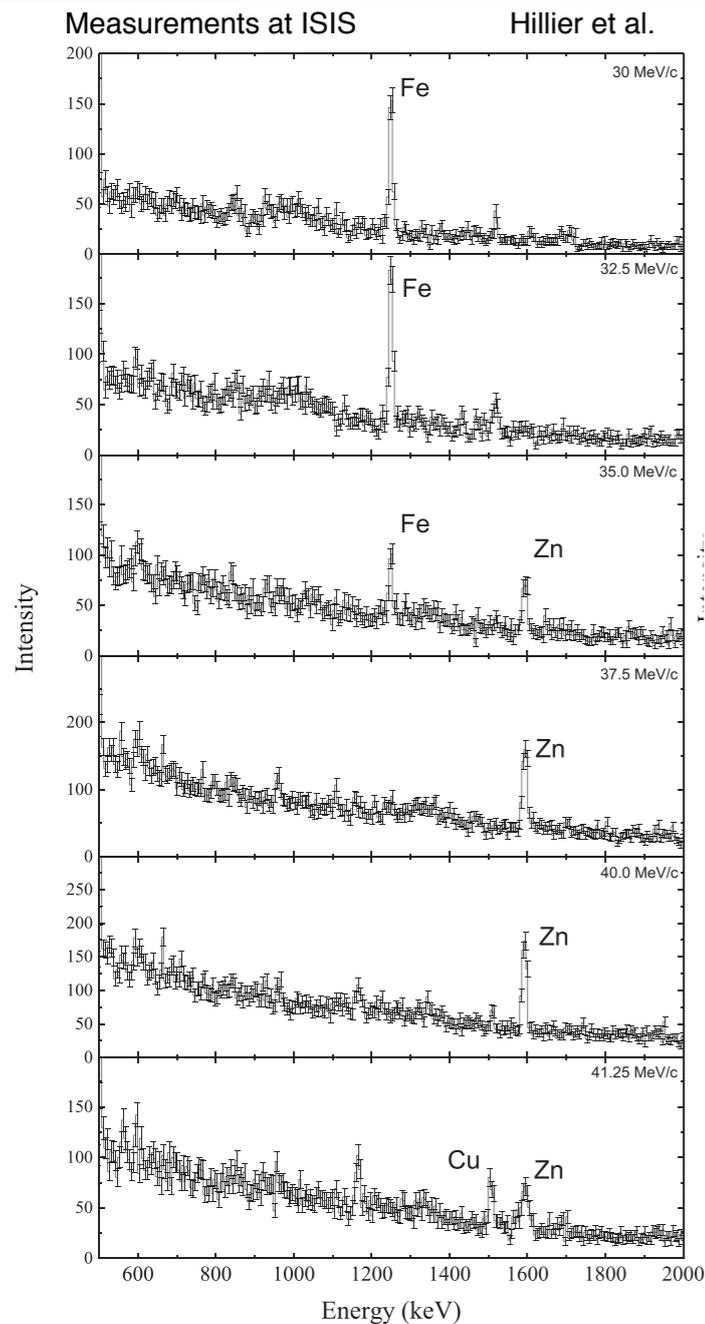
- ▶ We knew that we had lost a lot of radium in the target making process plus target had a large organic contamination
- ▶ Mounted target anyway but immediately saw that we had only 1% of the required target mass...
- ▶ Measured for a while, but clearly saw nothing

Alpha Spectrum

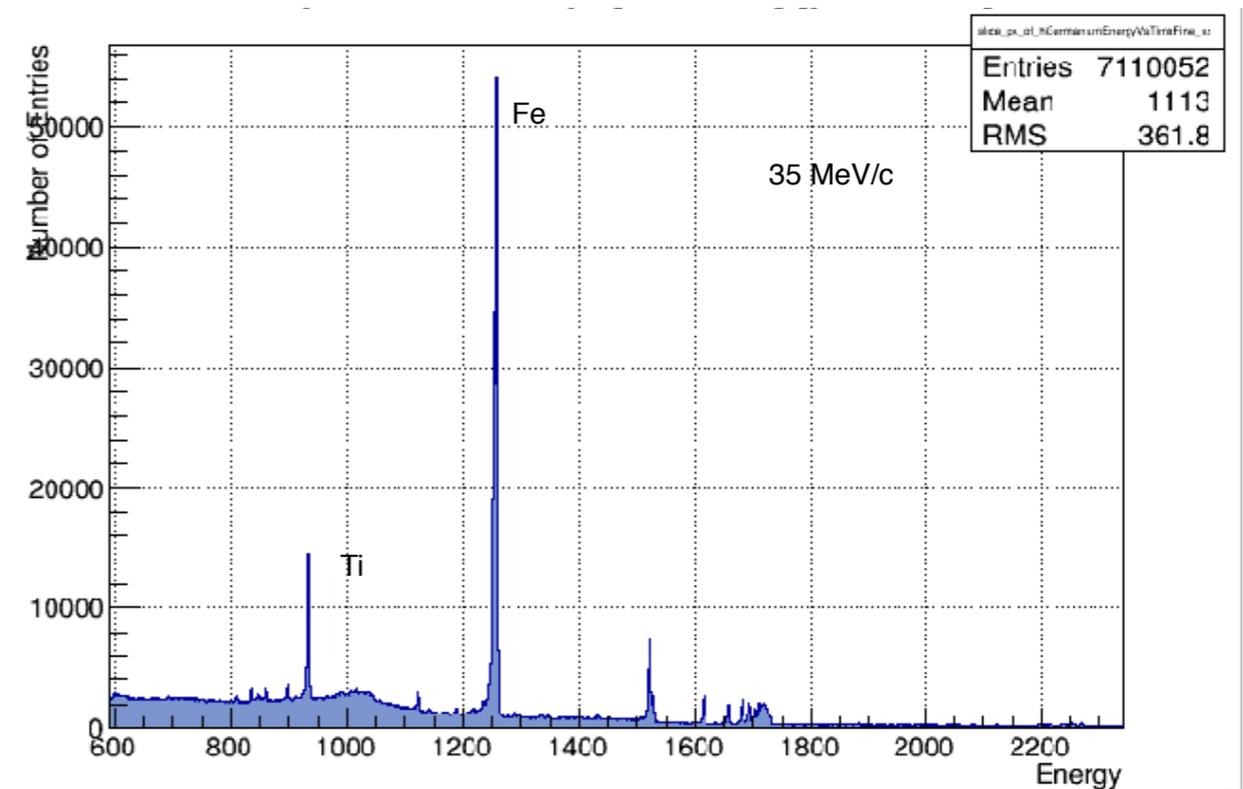
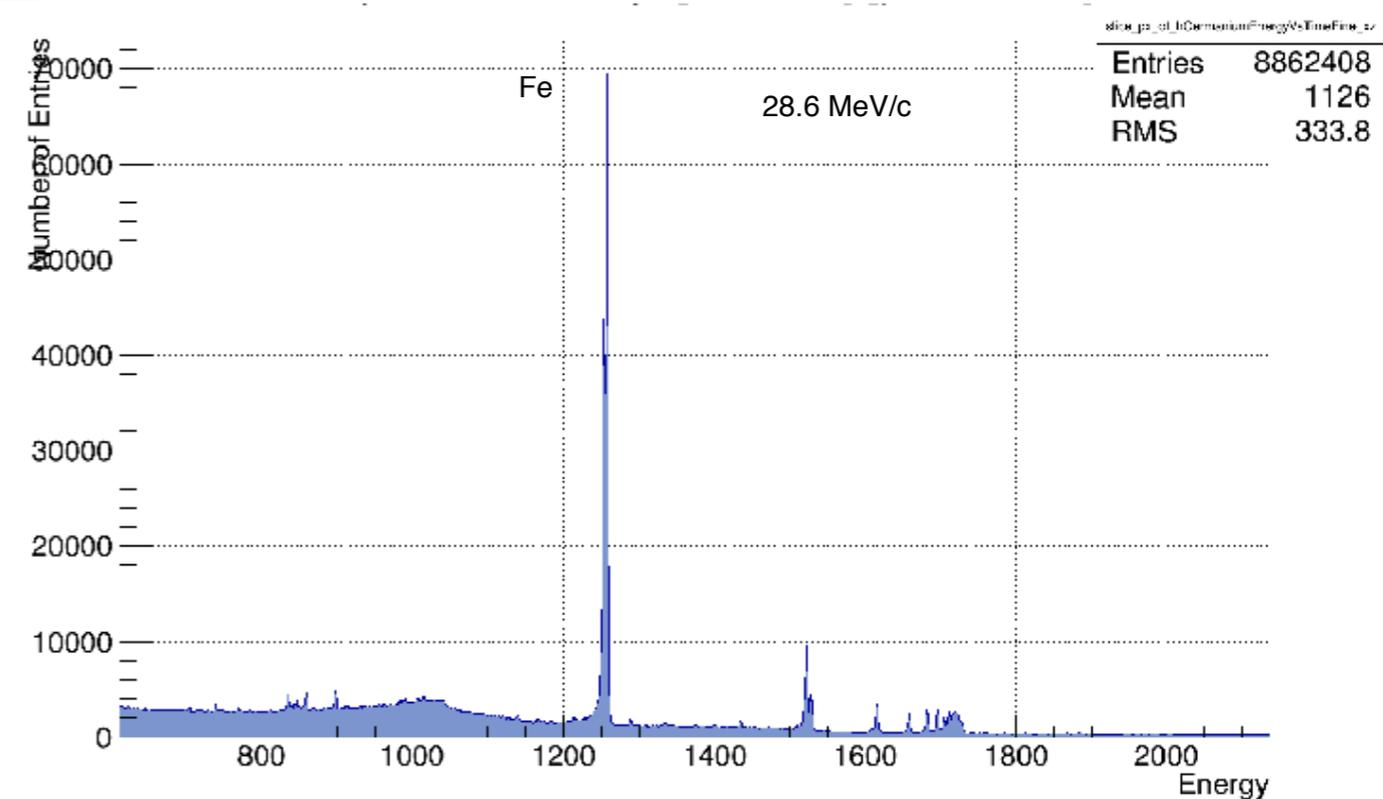


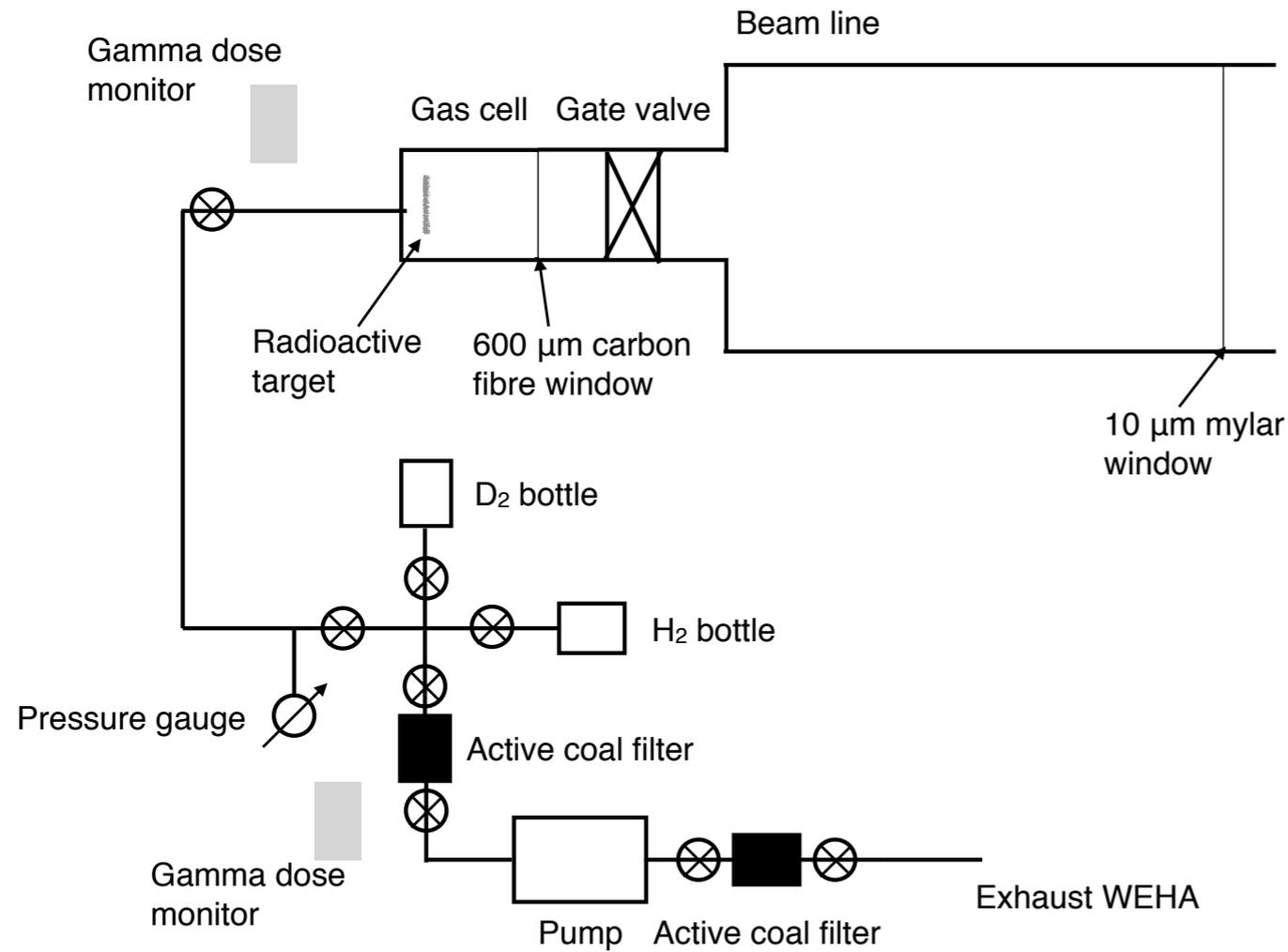
- ▶ Alpha spectrum measurements can reveal some hints on source thickness
- ▶ Tails and unresolved double peak clearly show that we have a “thick” source
- ▶ Performed some alpha spectrum simulations but quite a lot of free parameters
- ▶ Simulations tend to point towards organic layer of order 500 nm

Elemental analysis with negative muons



- ▶ Elemental analysis with muonic x-rays
- ▶ Depth profiling as a function of momentum
- ▶ Proof-of-principle with stacks of foils





- Implemented full safety features for handling radioactive targets