

Nuclear Astrophysics with Accelerator Mass Spectrometry (AMS)

Georg Rugel
working (now) at

Helmholtz-Zentrum Dresden-Rossendorf (HZDR)

@ FWIR: Accelerator Mass Spectrometry & Isotope Research
measuring (now) **@DREAMS (DREsden AMS)**

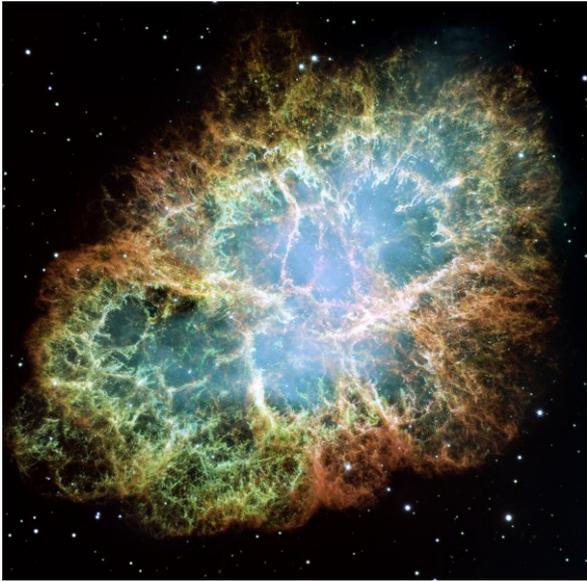
17th Rußbach School on Nuclear Astrophysics, 2022 March 13th-19th



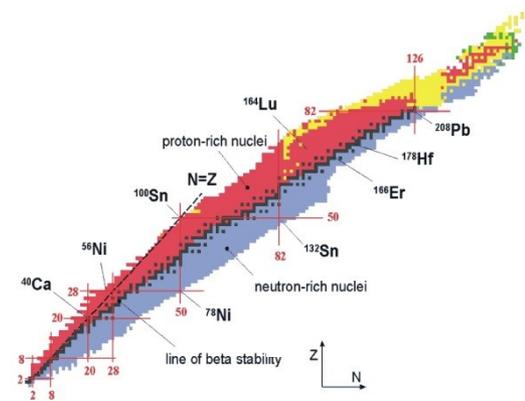
Overview

- **Accelerator Mass Spectrometry
DREsden AMS**
- Applications of AMS to Astrophysics
some examples (only very brief)

Application Astrophysics



HST Crab Nebula



Messengers: Radionuclides

Crab nebula ~ 2000 pc

Near-Earth Supernova
~ 100 pc 1 pc ~ 3 Lyr

Detectives:

Accelerator Mass Spectrometry
(measure rare isotopes)



DREAMS
(DREsden AMS)



MLL
AMS
at TU Munich

Accelerator Mass Spectrometry

Counting individual atoms (like conventional **MS**)

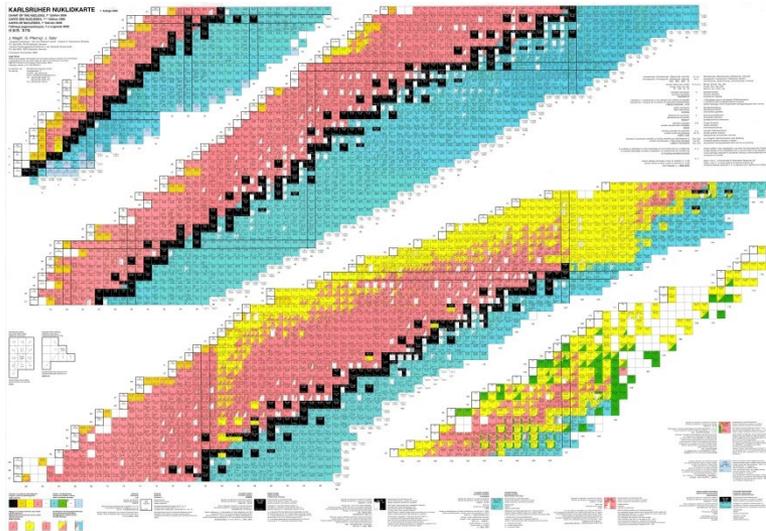
But

- accelerator (mostly a tandem – but new developments)
- molecules destroyed by stripping
- challenge: isobar separation
techniques: e.g. gas-filled magnet (GFM),
degrader foil, + detector (ionisation chamber)
- mostly for radionuclides
- extremely sensitive (isotopic ratio $\sim 10^{-16}$)



Historical

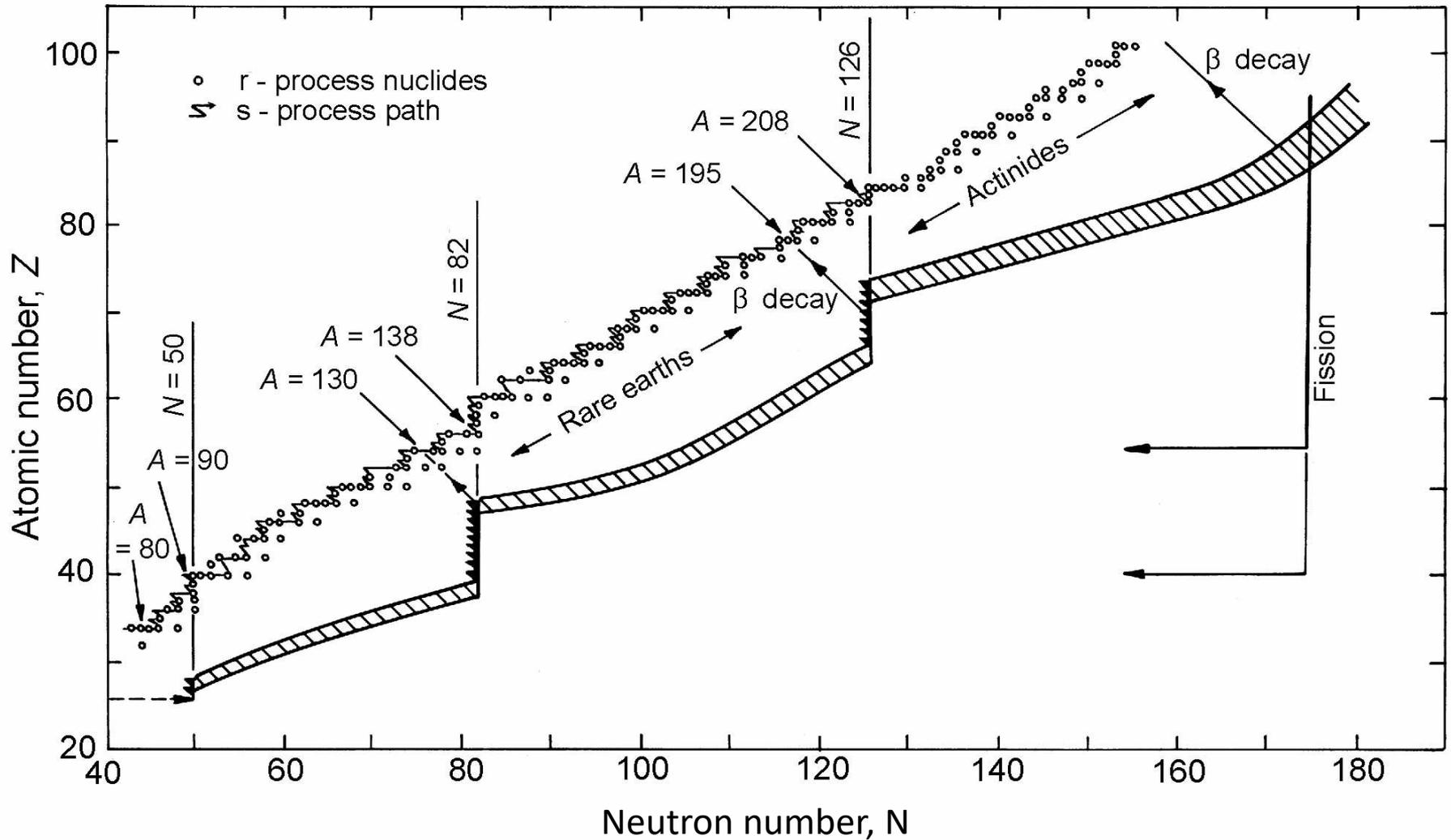
- 1939** Alvarez and Cornog discovered accidentally ^3He (cyclotron).
From 1939 until 1977, for around 40 years, dark and silent!
- 1977** Muller proposed to measure ^{14}C or ^{10}Be by means of a cyclotron.
Independently, Gove, Litherland, and Purser suggested the use of a Tandem accelerator for ^{14}C measurements.
- 2020** Much more than 100 AMS laboratories worldwide.
From small table top accelerators until large facilities with more than 10 MV terminal voltage.
Isotopes of interest from ^3H until the heaviest nuclides like ^{244}Pu .



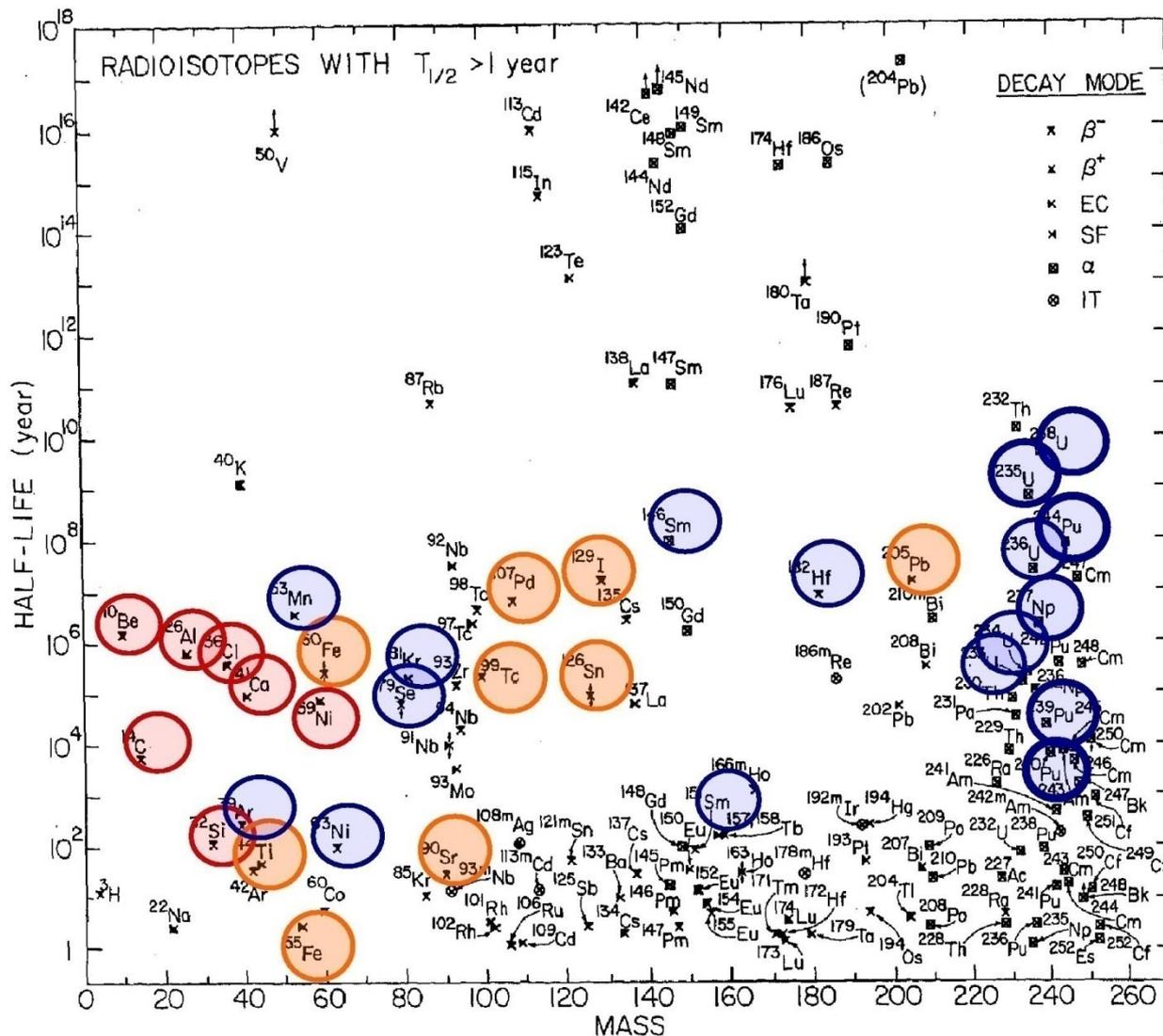
The majority of AMS laboratories is dedicated to ^{14}C and ^{10}Be .

Every 3 years AMS conference:
2021 online (Sydney) –
A lot of new ideas, developments!

Nucleosynthesis



Radionuclides detected by AMS



**Status
1981**

^{10}Be , ^{14}C , ^{26}Al ,
 ^{32}Si , ^{36}Cl ,
 ^{41}Ca , ^{59}Ni

**Status
1996**

+9 new
isotopes

**Status
2008**

+17 more
new isotopes

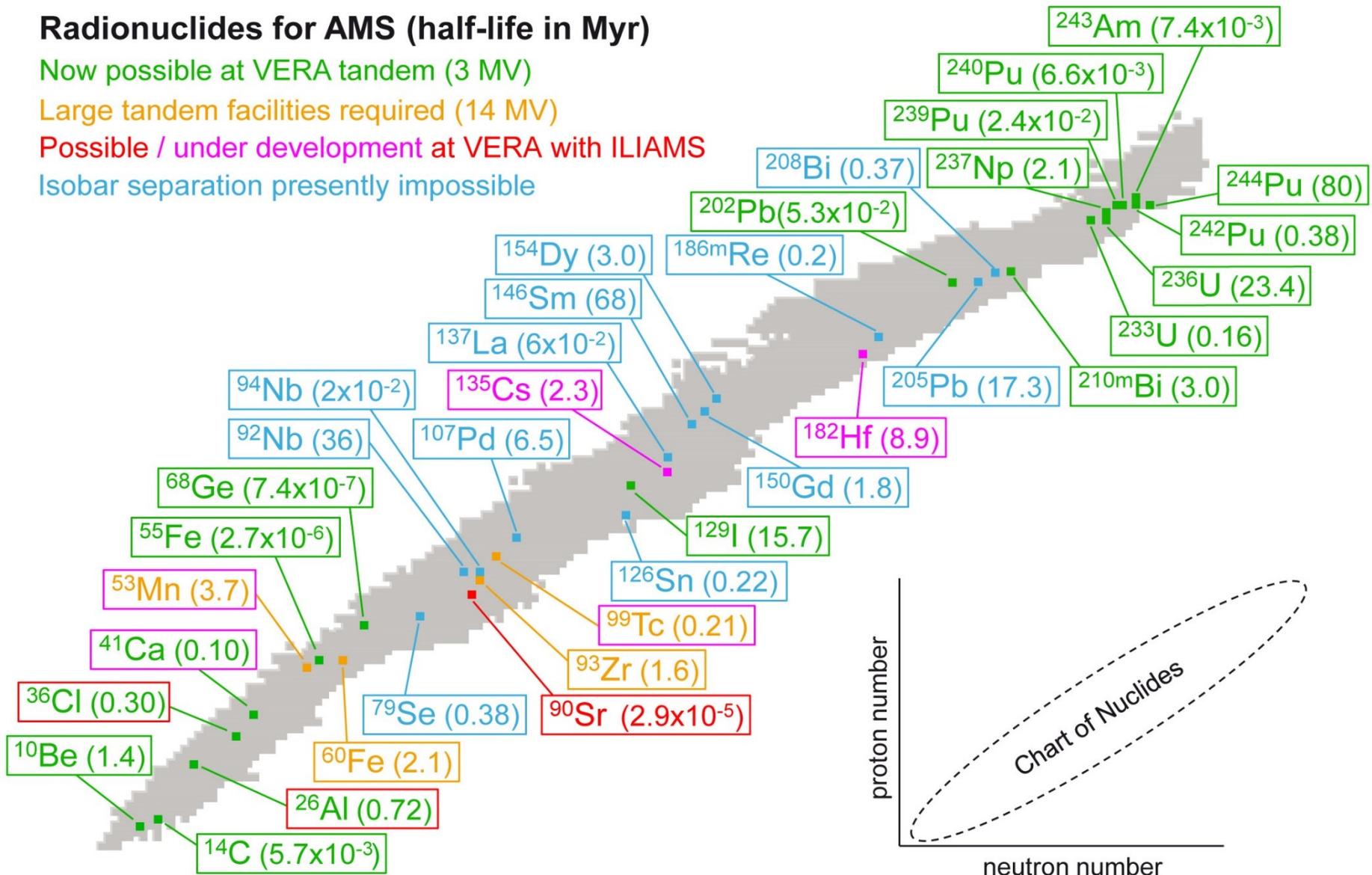
Radionuclides for AMS (half-life in Myr)

Now possible at VERA tandem (3 MV)

Large tandem facilities required (14 MV)

Possible / under development at VERA with ILIAMS

Isobar separation presently impossible



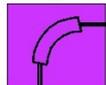
From VERA

AMS Setup at Munich



Negative ion source

2nd mass spectrometer



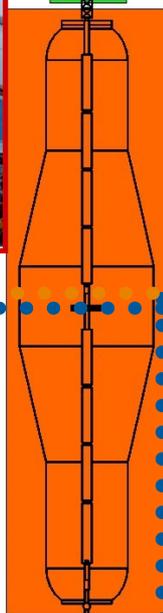
Wien filter 2

90 deg analyzing magnet



Wien filter 1

high-energy MS
(+ positive ions)



Stripper foil

14MV MP tandem

10m

150 kV preacceleration

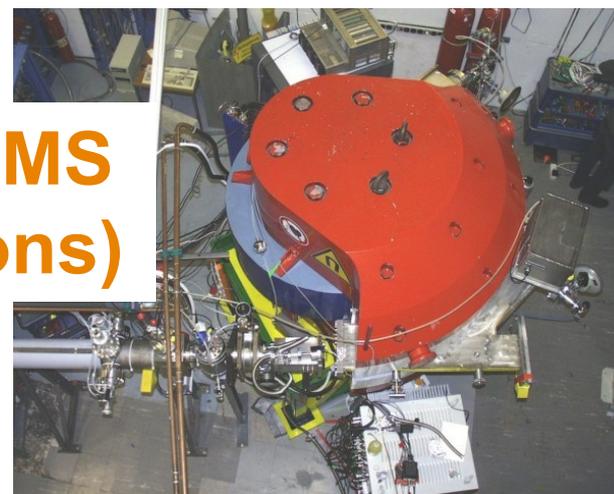
18 deg. electrostatic deflection

1st mass spectrometer

time-of-flight

gas-filled magnet

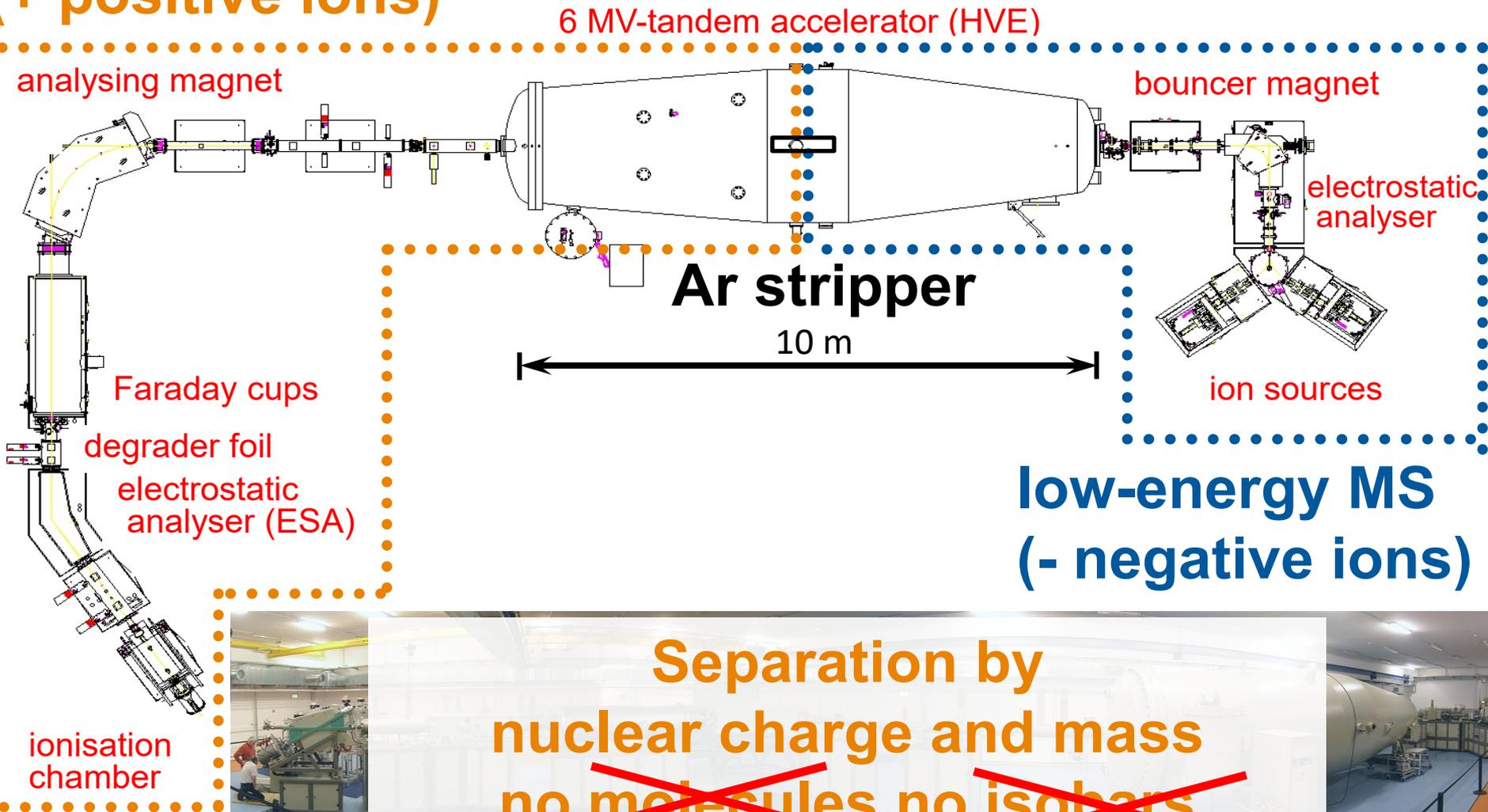
ionization chamber



low-energy MS
(- negative ions)

DREAMS - DREsden AMS

high-energy MS
(+ positive ions)



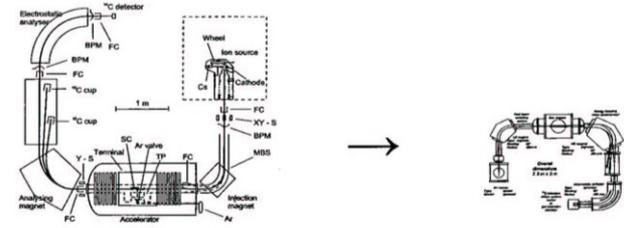
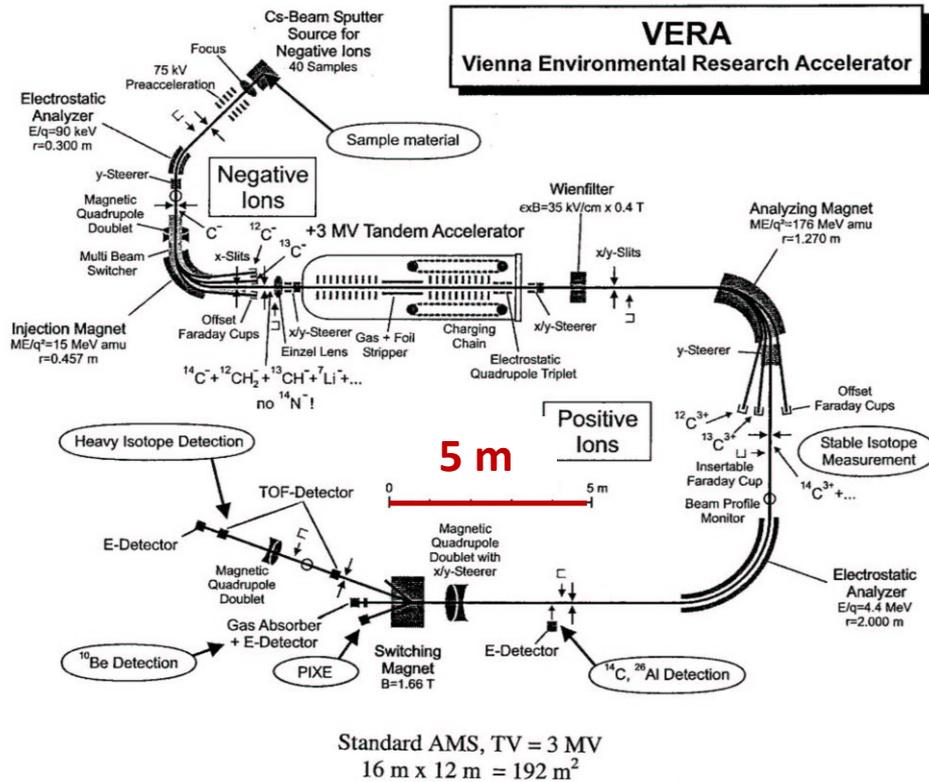
low-energy MS
(- negative ions)

Separation by
nuclear charge and mass
~~no molecules~~ ~~no isobars~~

Akhmadaliev et al., NIMB, 2013.; Rugel et al., NIMB, 2016.

AMS Facilities

max. voltages: 3 MV; 0.5 MV; 0.2 MV



Compact AMS, TV = 0.5 MV
6 m x 5 m = 30 m²

Poznan

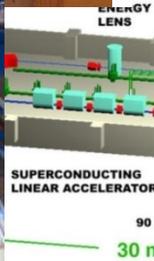
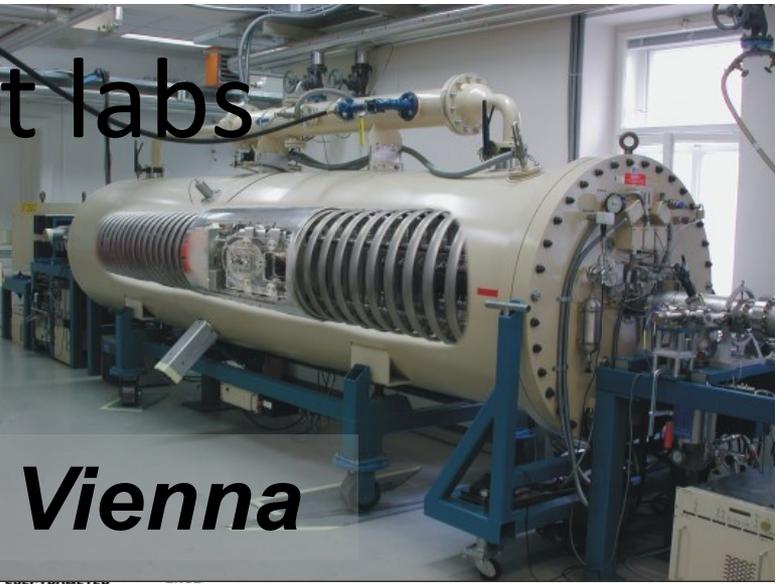
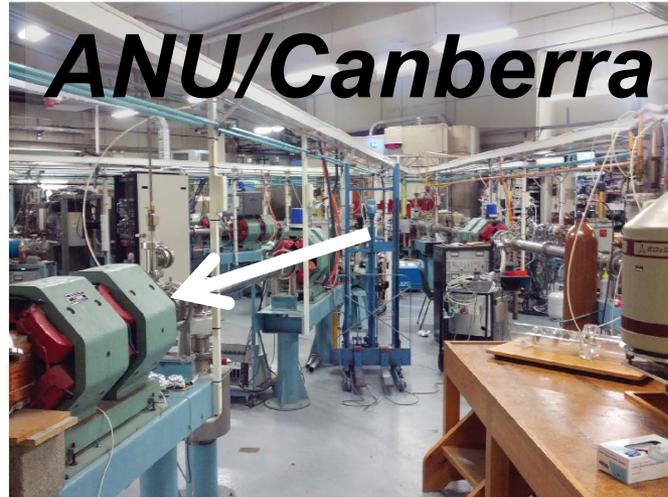
Mini AMS, TV = 0.2 MV
3.0 m x 2.3 m = 6.9 m²

Zurich

Vienna

AMS at different labs

ANU/Canberra



ETH Zurich

Slide from Anton Wallner

New dedicated AMS @ HZDR

Building Construction started 2022

max. voltage: 1 MV

Novel projects – like:

Isobar suppression at low energy side
“Laser”

contact:

Anton Wallner, Johannes Lachner



NEW Street

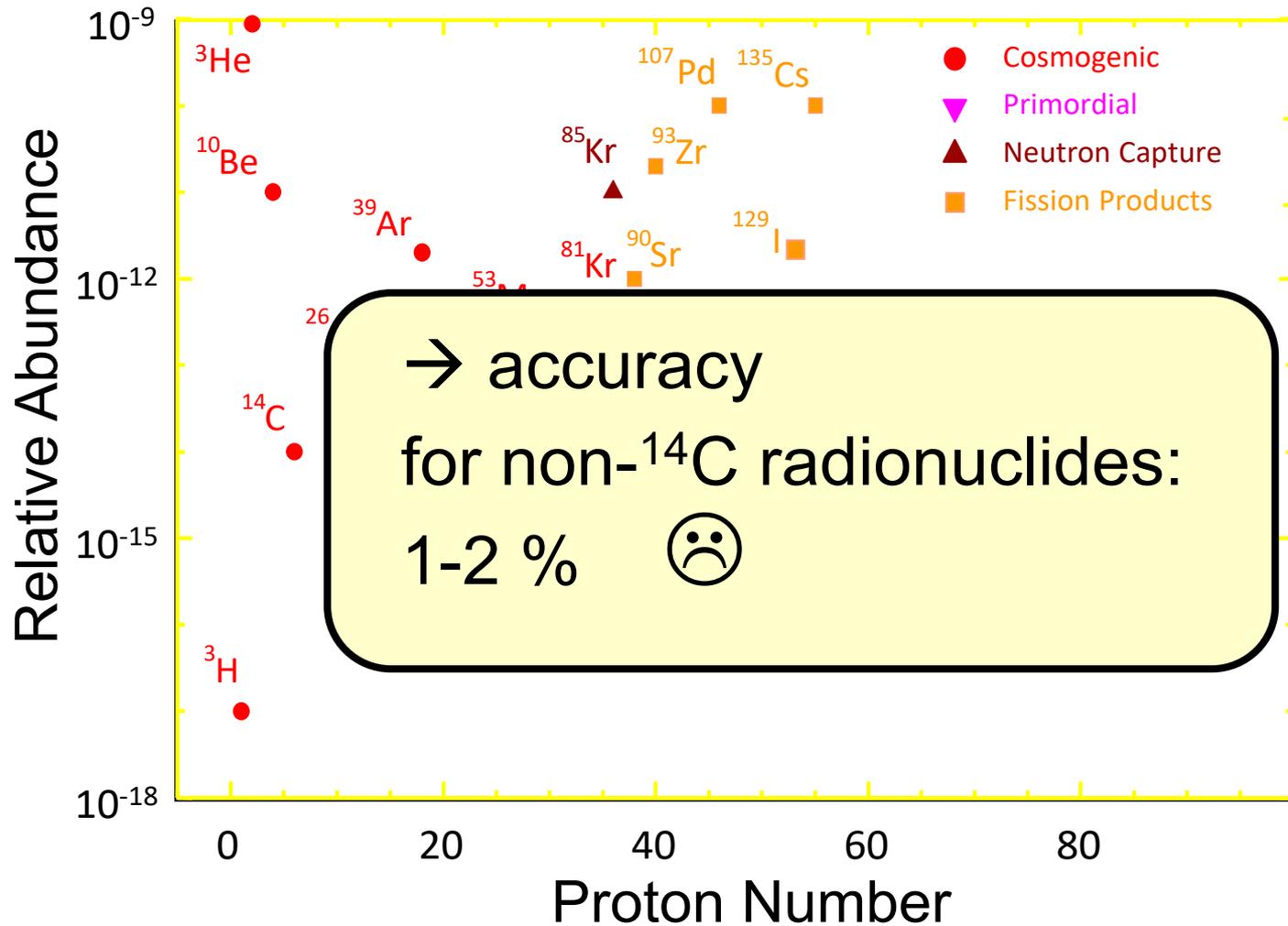
NEW Building



NEW Street

NEW Building

Typical isotopic ratios



Conventional Mass Spec.
(TIMS, ICP-MS, ...)

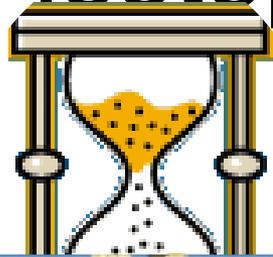
Laser Mass Spectrometry
(RIMS @ UMz)

AMS

adapted from Klaus Wendt



Isotopic ratios @ 10^{-15} ?



e.g. sand grain \varnothing 0.44 mm



One black grain : $\sim 10^{-15}$



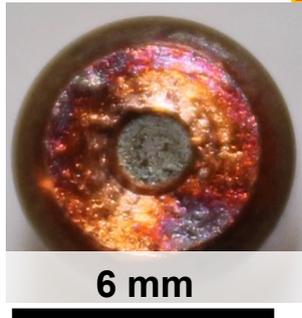
Frauenkirche Dresden, Germany

AMS needs chemistry (sample material into an ion source!)

Careful extraction of RN
(quantitative, no contamination,
1st isobar separation)



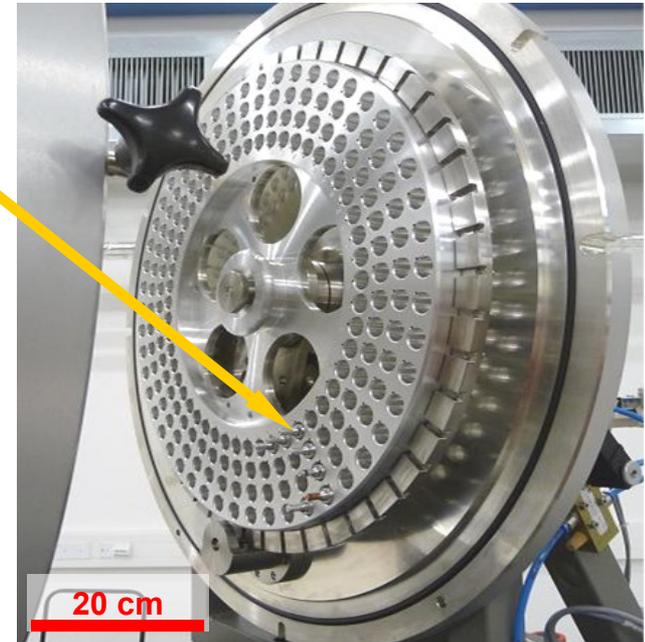
1 mg



AMS needs a high performance
negative ion source



AMS target holder



Wheel for 200 samples (4 x 50)

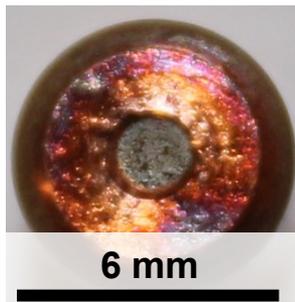
Sometimes possible to avoid chemistry;
e.g. cross section measurements

Chemical AMS sample preparation @ DREAMS

(operational since September 2009)
many users; two laboratories



Volatile elements:
 ^{36}Cl , ^{129}I



Other elements:
e.g. ^{10}Be , ^{26}Al , ^{41}Ca
(^{53}Mn , ^{60}Fe , ...)

- [Acknowledgements](#)
- [Introduction](#)
- [Negative Ion Source](#)
- [Operating Conditions & Procedures](#)
- [Cathode Preparation](#)
- [Gas Cathodes](#)
- [Ionization Efficiency](#)
- [Source Maintenance & Cleaning](#)
- [Toxicity of Cathode Materials](#)
- [Ionization Potentials & Electron Affinities](#)

A Negative-Ion Cookbook

Roy Middleton

Department Of Physics, University of Pennsylvania
 Philadelphia, PA 19104
 October 1989 (Revised February 1990)

1H Hydrogen							2He Helium
3Li Lithium	4Be Beryllium	5B Boron	6C Carbon	7N Nitrogen	8O Oxygen	9F Fluorine	10Ne Neon
11Na Sodium	12Mg Magnesium	13Al Aluminum	14Si Silicon	15P Phosphorus	16S Sulfur	17Cl Chlorine	18Ar Argon
19K Potassium	20Ca Calcium	31Ga Gallium	32Ge Germanium	33As Arsenic	34Se Selenium	35Br Bromine	36Kr Krypton
37Rb Rubidium	38Sr Strontium	49In Indium	50Sn Tin	51Sb Antimony	52Te Tellurium	53I Iodine	54Xe Xenon
55Cs Cesium	56Ba Barium	81Tl Thallium	82Pb Lead	83Bi Bismuth	84Po Polonium	85At Astatine	86Rn Radon

Transition Elements									
21Sc Scandium	22Ti Titanium	23V Vanadium	24Cr Chromium	25Mn Manganese	26Fe Iron	27Co Cobalt	28Ni Nickel	29Cu Copper	30Zn Zinc
39Y Yttrium	40Zr Zirconium	41Nb Niobium	42Mo Molybdenum	43Tc Technetium	44Ru Ruthenium	45Rh Rhodium	46Pd Palladium	47Ag Silver	48Cd Cadmium
57La Lanthanum	72Hf Hafnium	73Ta Tantalum	74W Tungsten	75Re Rhenium	76Os Osmium	77Ir Iridium	78Pt Platinum	79Au Gold	80Hg Mercury

Lanthanides						
58Ce Cerium	59Pr Praseodymium	60Nd Neodymium	61Pm Promethium	62Sm Samarium	63Eu Europium	64Gd Gadolinium
65Tb Terbium	66Dy Dysprosium	67Ho Holmium	68Er Erbium	69Tm Thulium	70Yb Ytterbium	71Lu Lutetium

Actinides		
90Th Thorium	91Pa Protactinium	92U Uranium

Michael Wiplich at the Tandem Van de Graaff Accelerator located at the Brookhaven National Laboratory prepared the electronic version of the Negative-Ion Cookbook. The original paper version was converted to electronic form using an H.P. Scanjet 6200C scanner and H.P. Precision-Scan Pro OCR software followed by manual fix up in MS Word 2000 Pro which was also used to convert the document to HTML.

Please address any comments, suggestions or corrections to mwiplich@bnl.gov.

The Home Page of the Brookhaven National Laboratory Tandem Van de Graaff Accelerator Is Located At <http://tvdg10.phy.bnl.gov/>.

Working horse:
Sputter ion sources
Middleton type

No double or triple charged negative ions!

Scanned version available under:

<http://tvdg10.phy.bnl.gov/COOKBOOK/>



Ionisation

Ionization Potentials and Electron Affinities of the Elements

IA	IIA	IIIA	IVA	VA	VIA	VIIA	VIIIA
1H 13.59 0.754	Ionization Potential Electron Affinity						2He 24.48 0.078*
3Li 5.39 0.618	4Be 9.32 0.195*	5B 8.30 0.277	6C 11.26 1.263	7N 14.53 -0.07	8O 13.61 1.461	9F 17.42 3.399	10Ne 21.56 < 0
11Na 5.14 0.548	12Mg 7.64 < 0	13Al 5.98 0.441	14Si 8.15 1.385	15P 10.48 0.747	16S 10.36 2.077	17Cl 13.01 3.617	18Ar 15.76 < 0
19K 4.34 0.501	20Ca 6.11 0.043	31Ga 6.00 0.30	32Ge 7.90 1.2	33As 9.81 0.81	34Se 9.75 2.021	35Br 11.81 3.365	36Kr 14.00 < 0
37Rb 4.18 0.486	39Sr 5.70 < 0	49In 5.79 0.3	50Sn 7.34 1.2	51Sb 8.64 1.07	52Te 9.01 1.971	53I 10.45 3.059	54Xe 12.13 < 0
55Cs 3.89 0.472	56Ba 5.21 < 0	81Tl 6.11 0.2	82Pb 7.42 0.364	83Bi 7.29 0.946	84Po 8.42 1.9	85At 9.5 2.8	86Rn 10.75 < 0

*Metastable

[eV]



Ionisation
Potential
Electron
Affinity

11.26 14.53
1.263 -0.07

Complete suppression
in the ion source

Transition Elements									
21Sc 6.54 0.188	22Ti 6.82 0.079	23V 6.74 0.525	24Cr 6.77 0.666	26Mn 7.44 < 0	26Fe 7.87 0.163	27Co 7.86 0.661	28Ni 7.64 1.156	29Cu 7.73 1.228	30Zn 9.39 < 0
39Y 6.38 0.307	40Zr 6.84 0.426	41Nb 6.88 0.893	42Mo 7.10 0.746	43Tc 7.28 0.55	44Ru 7.37 1.05	46Rh 7.46 1.137	46Pd 8.34 0.557	47Ag 7.58 1.302	49Cd 8.99 < 0
57La 5.58 0.5	72Hf 7.0 ~0	73Ta 7.89 0.322	74W 7.98 0.815	75Re 7.88 0.15	76Os 8.7 1.1	77Ir 9.1 1.565	79Pt 9.0 2.128	79Au 9.22 2.309	80Hg 10.44 < 0

Lanthanides †						
58Ce 5.47 0.5	59Pr 5.42 0.0	60Nd 5.49 -0.3	61Pm 5.55 -0.3	62Sm 5.63 0.3	63Eu 5.67 -0.3	64Gd 6.16 0.5
65Tb 5.85 0.5	66Dy 5.93 -0.3	67Ho 6.02 -0.3	68Er 6.10 -0.3	69Tm 5.18 0.3	70Yb 6.25 -0.3	71Lu 5.43 0.5

Actinides ‡		
90Th 6.95 0.5	91Pa ? 0.3	92U 6.08 0.3

† Theoretical values of electron affinities from: Electron Affinities of the Lanthanides S.G. Bratsch, Chem. Phys. Lett., **98**(2) 113-117 (1983).

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IONIZATION POTENTIALS AND ELECTRON AFFINITIES OF THE ELEMENTS - Mozilla

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Actinides ‡

90Th 6.95 0.5	91Pa ? 0.3	92U 6.08 0.3
----------------------------	-------------------------	---------------------------



Ionisation

[eV]

⁶⁰Fe

⁶⁰Ni

Ionization
Potential

7.87

7.64

Electron
Affinity

0.163

1.156

Ni makes much more likely neg. ions!

Trick:

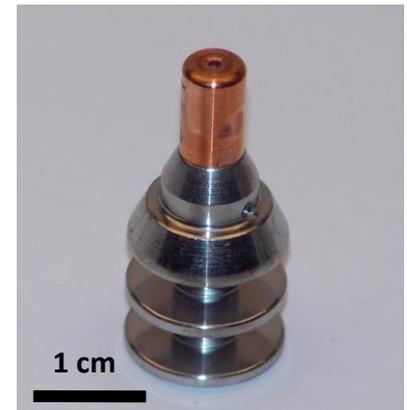
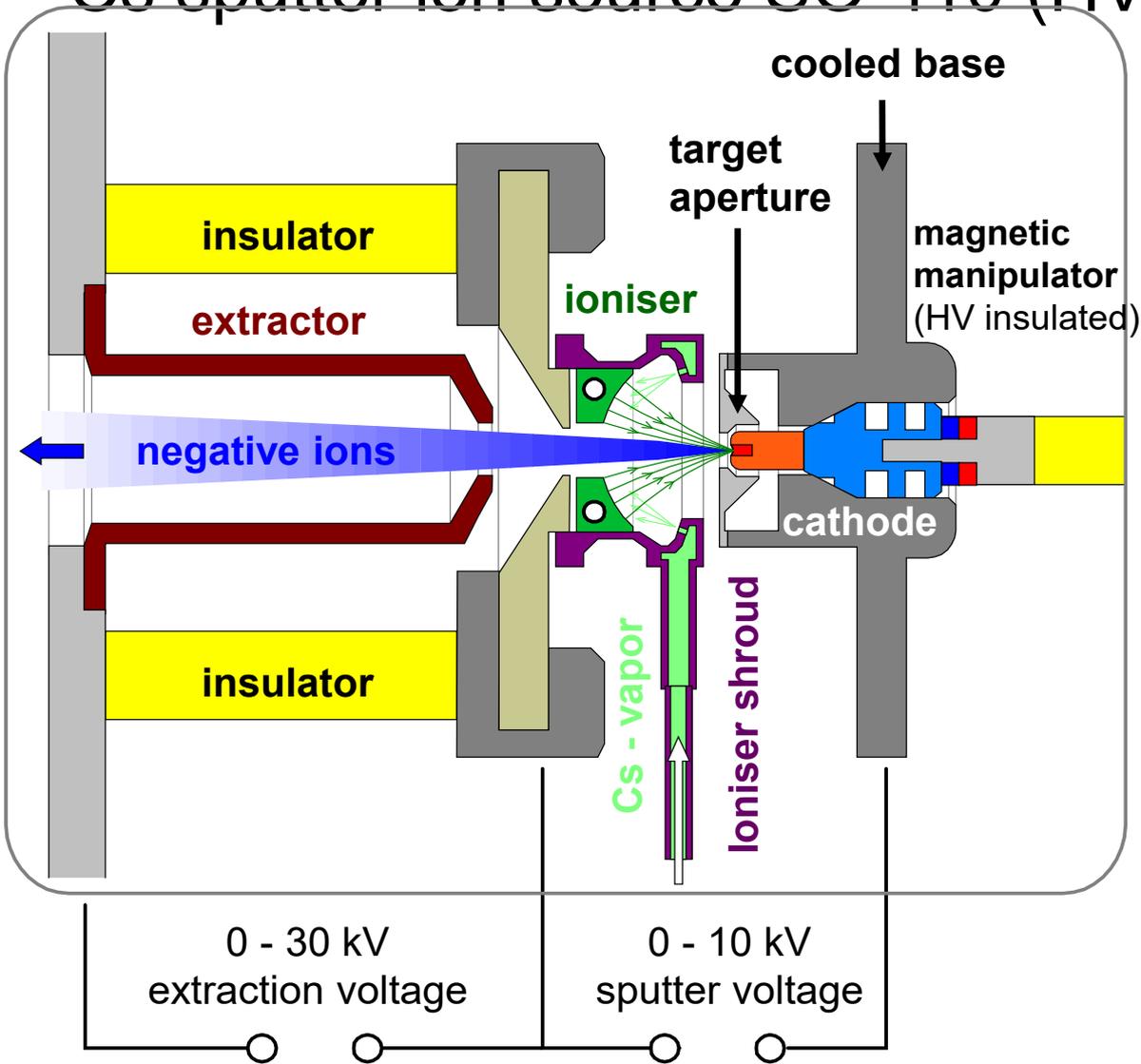
FeO⁻ and NiO⁻ have about the same (high) electron affinity

† Theoretical values of electron affinities from: Electron Affinities of the Lanthanides S.G. Bratsch, Chem. Phys. Lett., **98**(2) 113-117 (1983).

‡ Theoretical values of electron affinities from: Electron Affinities of the Actinides, S.G. Bratsch and J.J. Lagowski, Chem. Phys. Lett., **107**(2) 136-140 (1984).

AMS ion source:

Cs sputter ion source SO-110 (HVEE)



AMS target holder

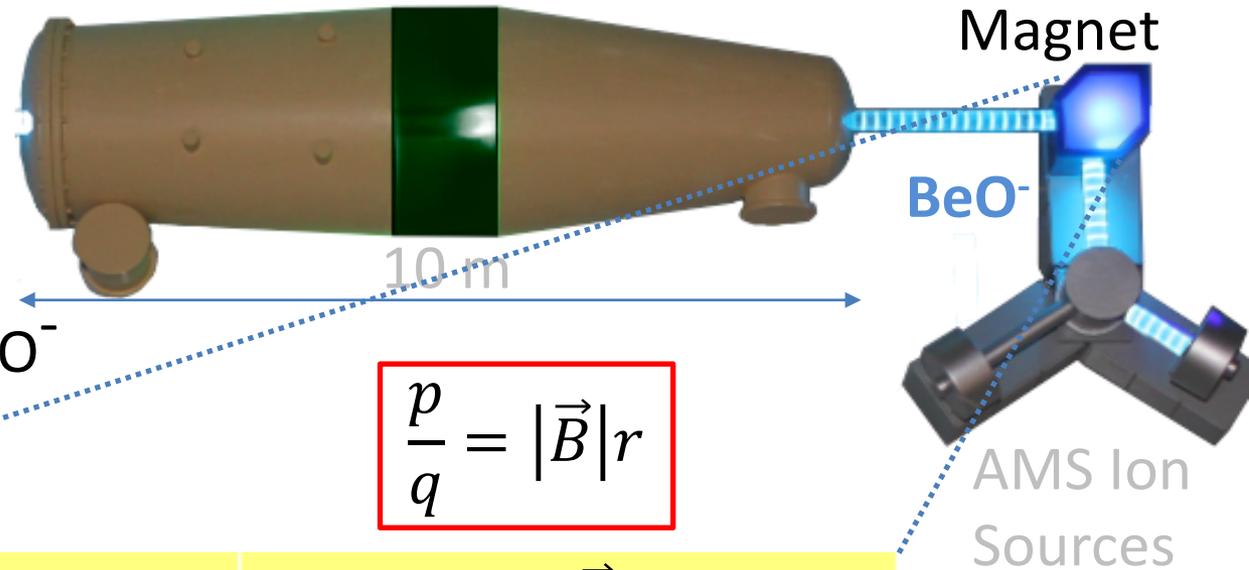


Wheel for 200 samples

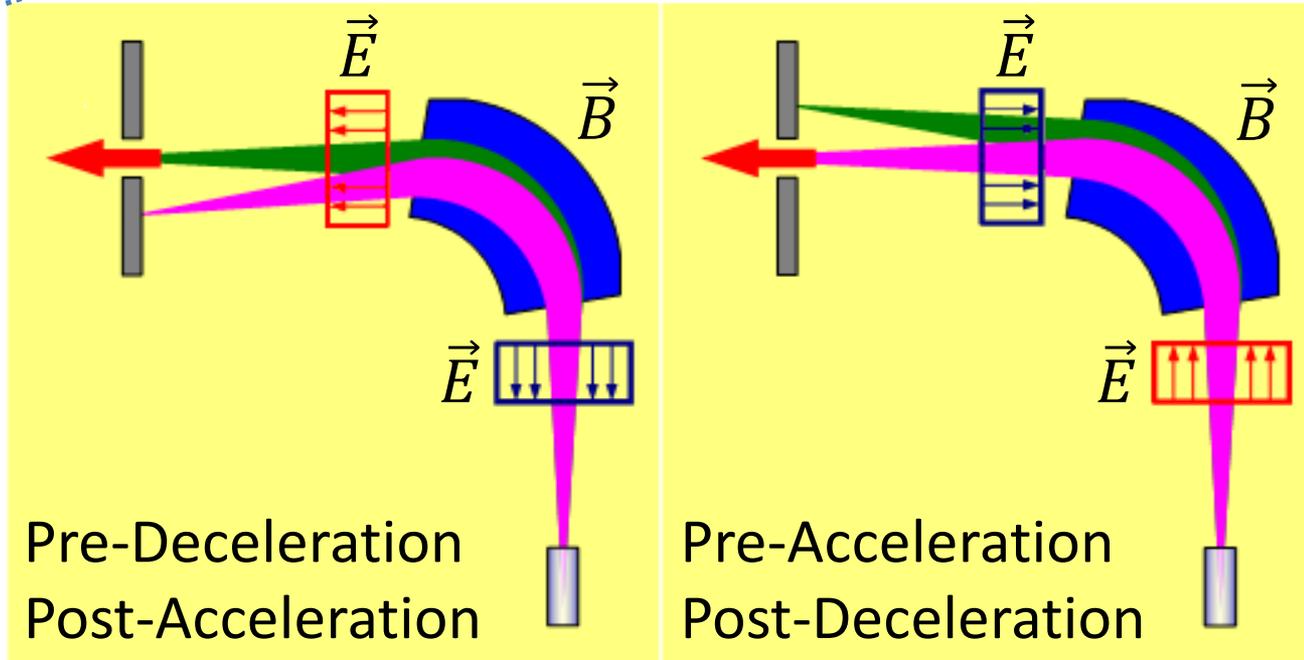
Quasi-Simultaneous Measurements

Fast Sequential Injection

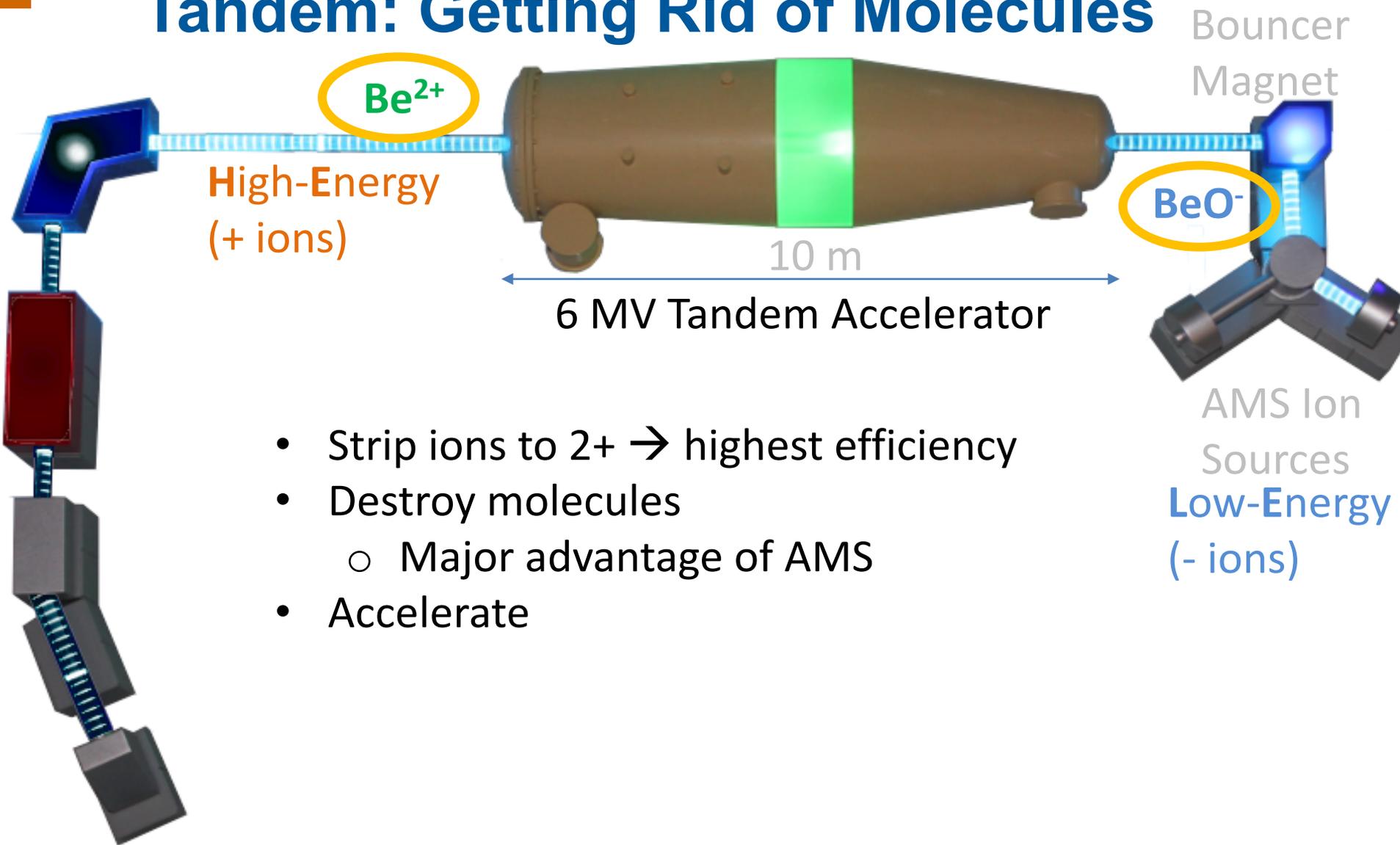
- Quasi-simultaneous stable and radio-nuclides
- E.g. ${}^9\text{Be}^{16}\text{O}^-$ and ${}^{10}\text{Be}^{16}\text{O}^-$
- Change \vec{E} not \vec{B}



$$\frac{p}{q} = |\vec{B}|r$$



Tandem: Getting Rid of Molecules



- Strip ions to 2+ → highest efficiency
- Destroy molecules
 - Major advantage of AMS
- Accelerate

HE 90°
Magnet

Measure Stable ^9Be

Bouncer
Magnet

Be^{2+}

10 m

6 MV Tandem Accelerator

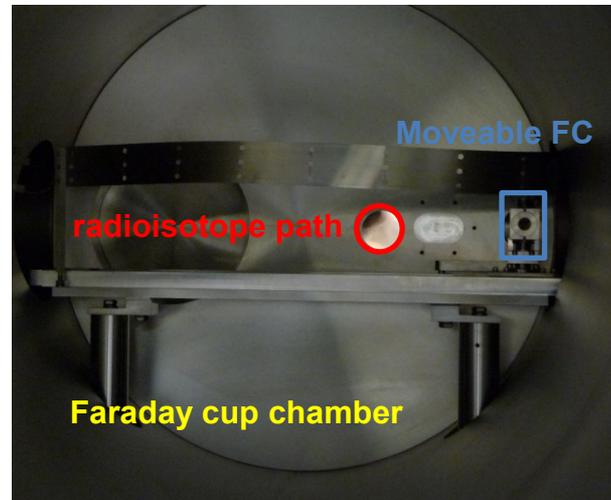
BeO^-

AMS Ion
Sources

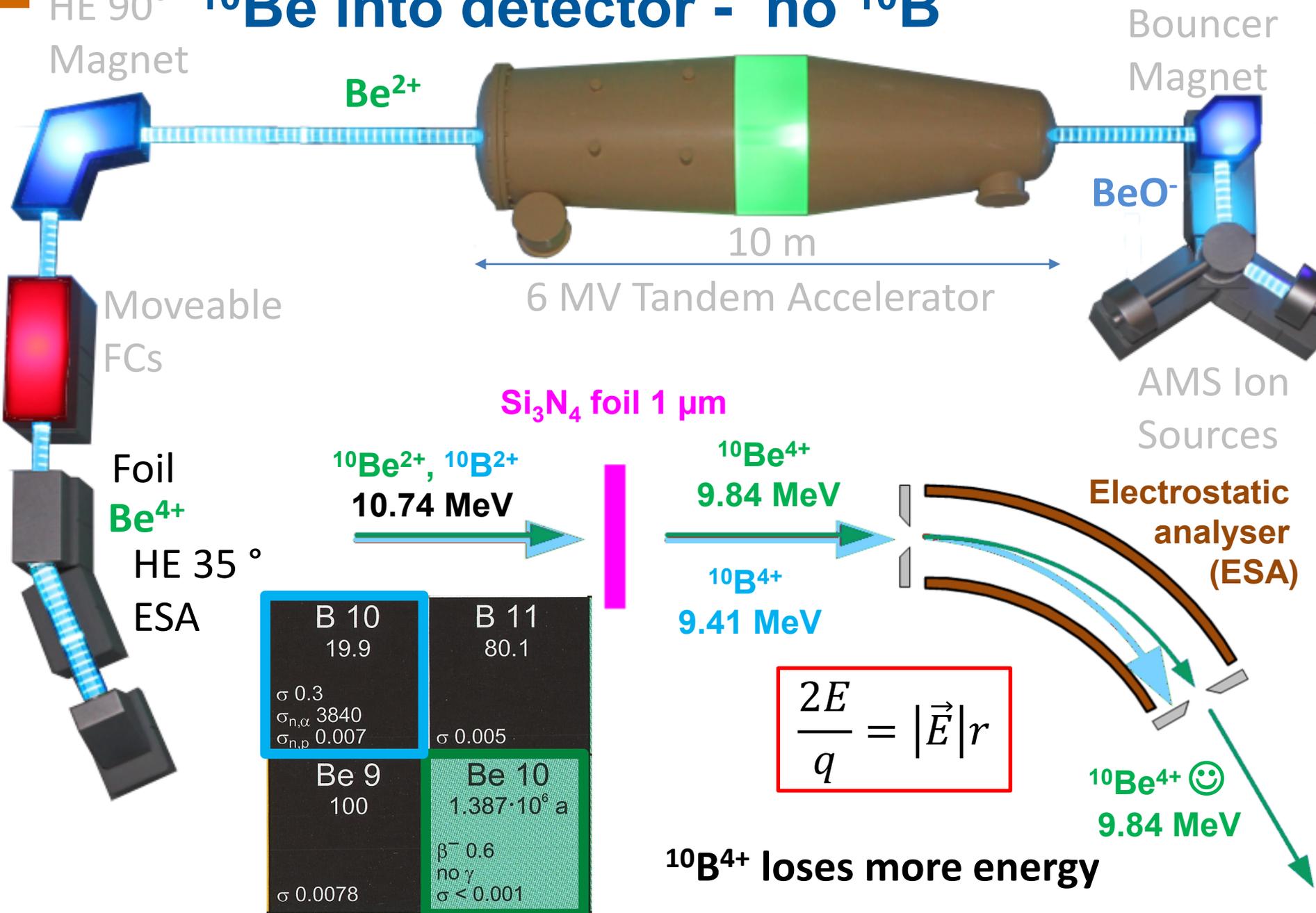
- ^9Be measured at Faraday Cup
- ^{10}Be go to radioisotope path



radioisotope path



HE 90° ¹⁰Be into detector - no ¹⁰B



B 10 19.9	B 11 80.1
σ 0.3 σ _{n,α} 3840 σ _{n,p} 0.007	σ 0.005
Be 9 100	Be 10 1.387 · 10 ⁶ a
σ 0.0078	β ⁻ 0.6 no γ σ < 0.001

$$\frac{2E}{q} = |\vec{E}|r$$

Typical performance data DREAMS

B 10 19.9 σ 0.3 $\sigma_{n, \alpha}$ 3840 $\sigma_{n, p}$ 0.007	B 11 80.1 σ 0.005
Be 9 100 σ 0.0088	Be 10 1.387 · 10 ⁶ a β^- 0.6 no γ $\sigma < 0.001$

0.5 mg BeO

$^{10}\text{Be}/^{9}\text{Be}$: 10^{-12} at/at

→ $1.2 \cdot 10^7$ at ^{10}Be

Total efficiency: 1:1400

→ $6.6 \cdot 10^3$ cts ^{10}Be at detector

Blank: $5 \cdot 10^{-16}$ ($^{10}\text{Be}/^{9}\text{Be}$)

Al 26 6.35 s 7.16 · 10 ⁵ a β^+ 1.2 γ 1809; 1130... $\sigma_{n, \alpha}$ 0.34 $\sigma_{n, p}$ 1.97 β^+ 3.2	Al 27 100 σ 0.230
Mg 25 10.00 σ 0.20	Mg 26 11.01 σ 0.038

0.3 mg Al₂O₃

$^{26}\text{Al}/^{27}\text{Al}$: 10^{-12} at/at

→ $7 \cdot 10^5$ at ^{26}Al

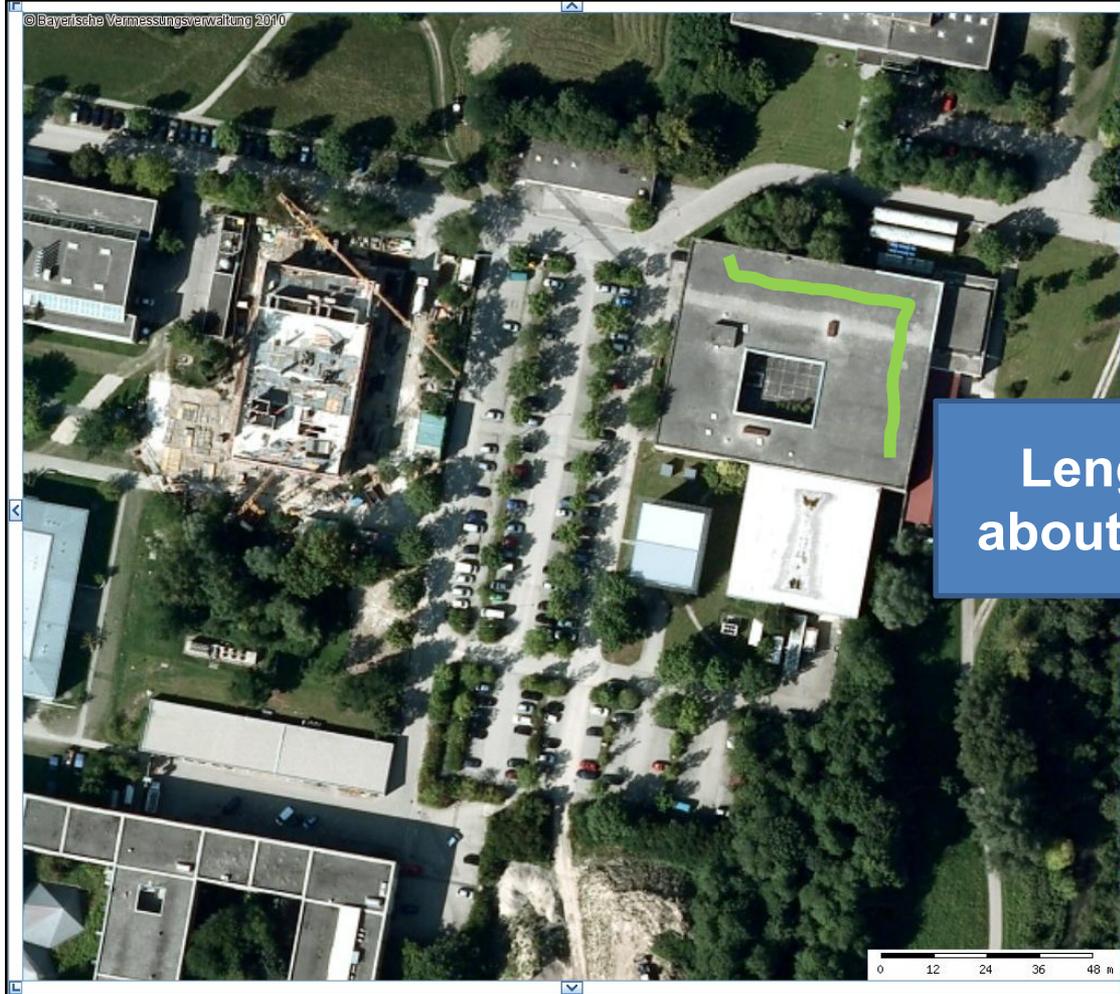
Total efficiency: 1:2000

→ 350 cts ^{26}Al at detector

Blank: $6 \cdot 10^{-16}$ ($^{26}\text{Al}/^{27}\text{Al}$)

Tandem Accelerator Laboratory Garching Part of the MLL (Maier Leibnitz Laboratory)

Ludwig Maximilians Universität and Technische Universität München



Length:
about 76 m

Closed end 2019

Low-energy side of the Tandem

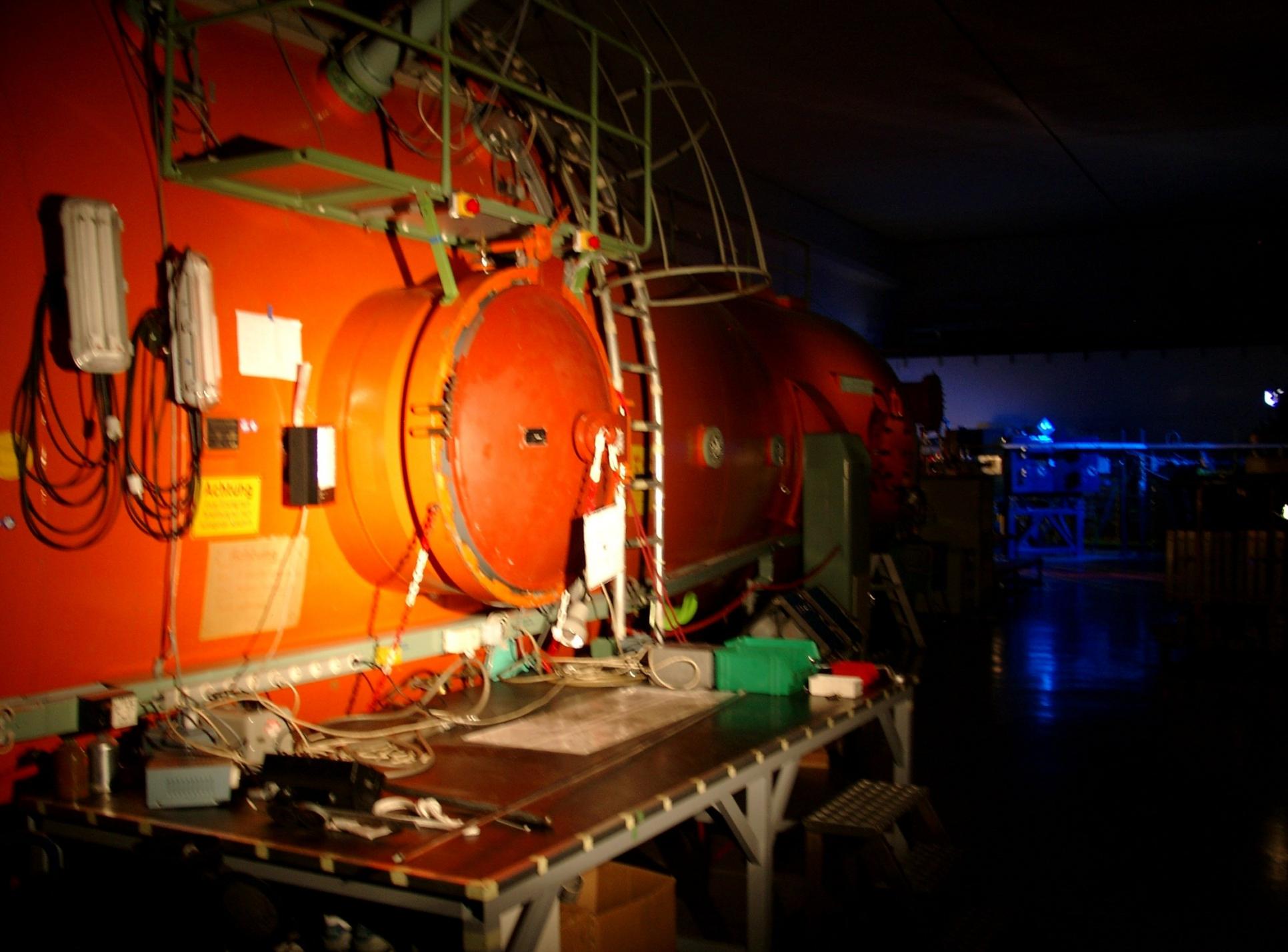


Unbefugten
Zutritt verboten



Hochspannung
Lebensgefahr







High-energy side of the Tandem

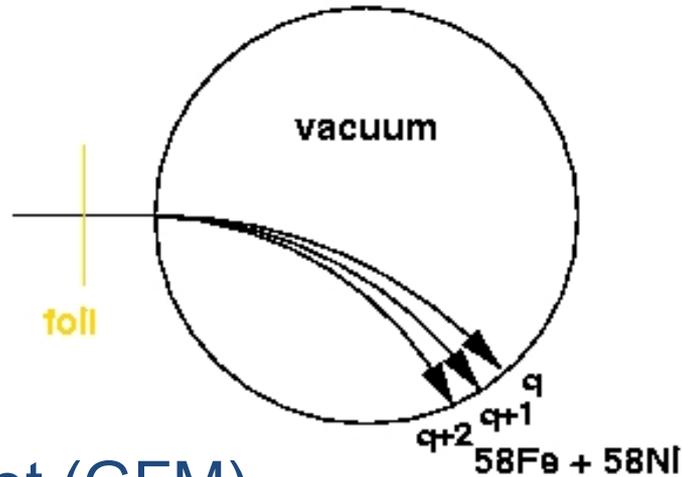
e.g.: $^{60}\text{Ni}^{11+} + ^{60}\text{Fe}^{11+}$
isobars not yet
suppressed

Isobar Separation in a Gas-filled Magnet



magnetic rigidity

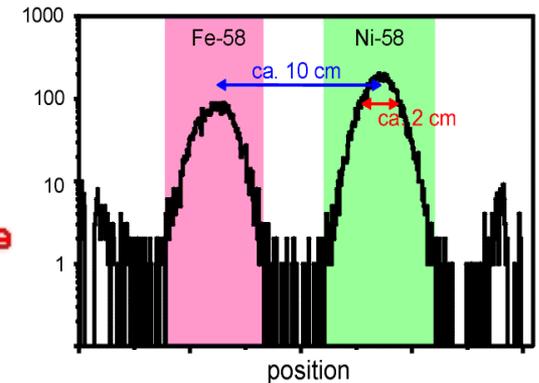
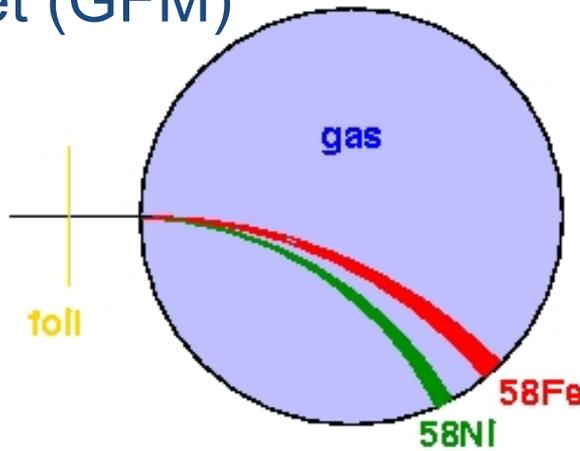
$$B\rho = \frac{Mv}{qe_0}$$



in a gas-filled magnet (GFM)

$$\langle q \rangle \sim vZ^{0.4}$$

$$B\rho \sim \frac{M}{Z^{0.4}}$$



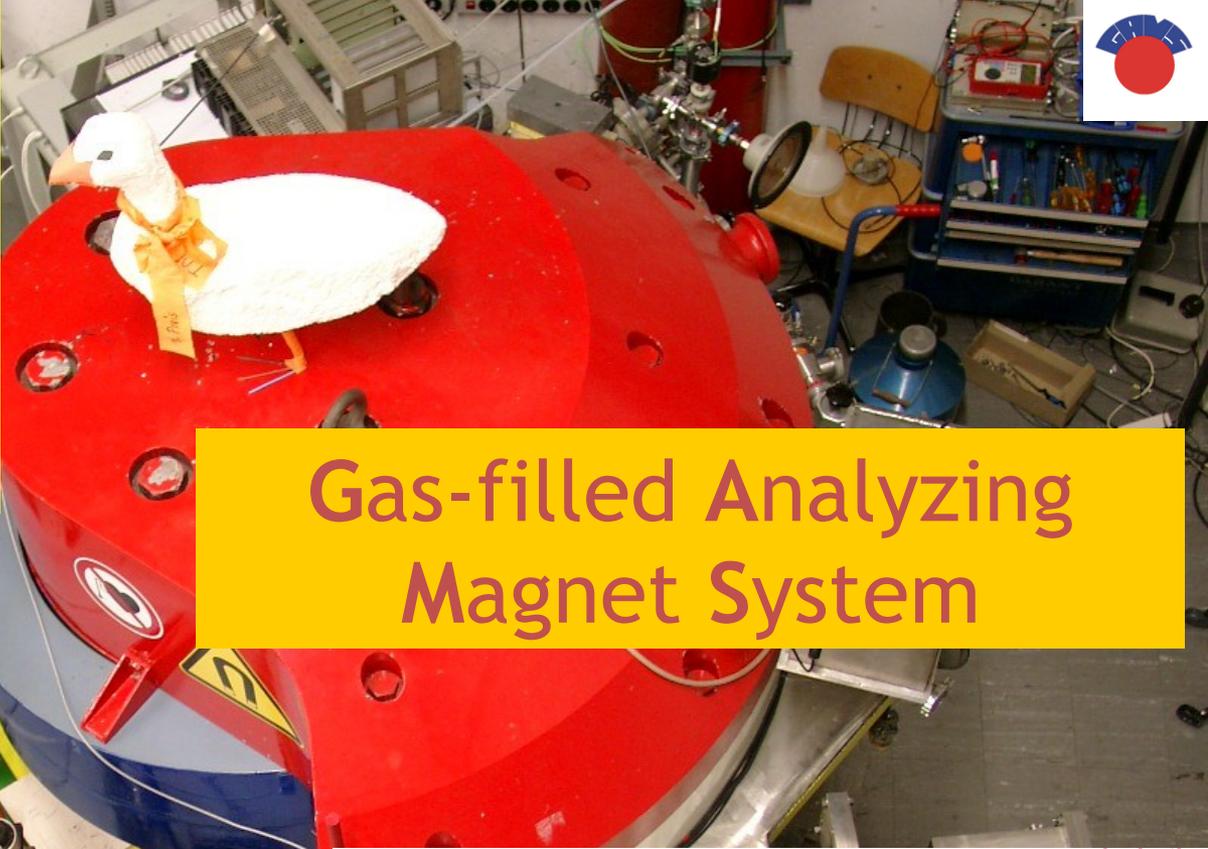


Isobaric suppression with GAMS-setup:

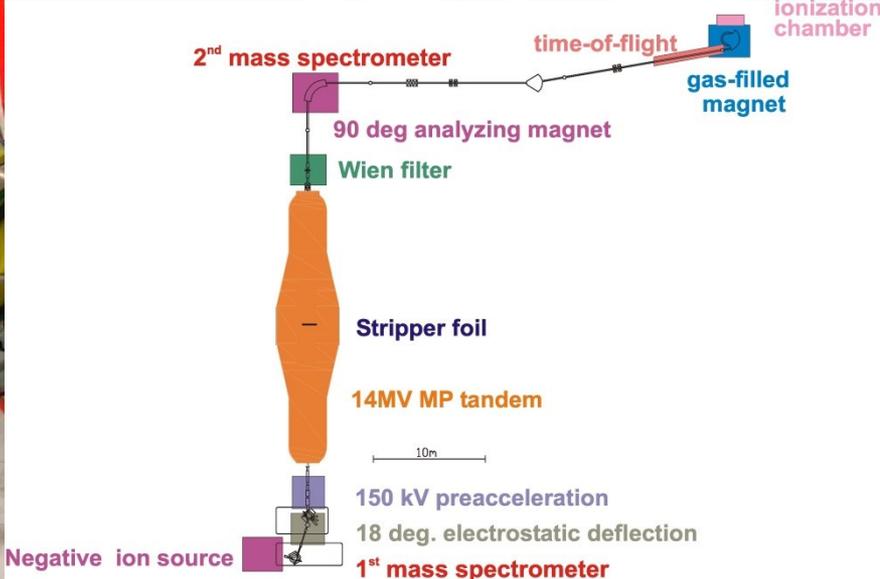
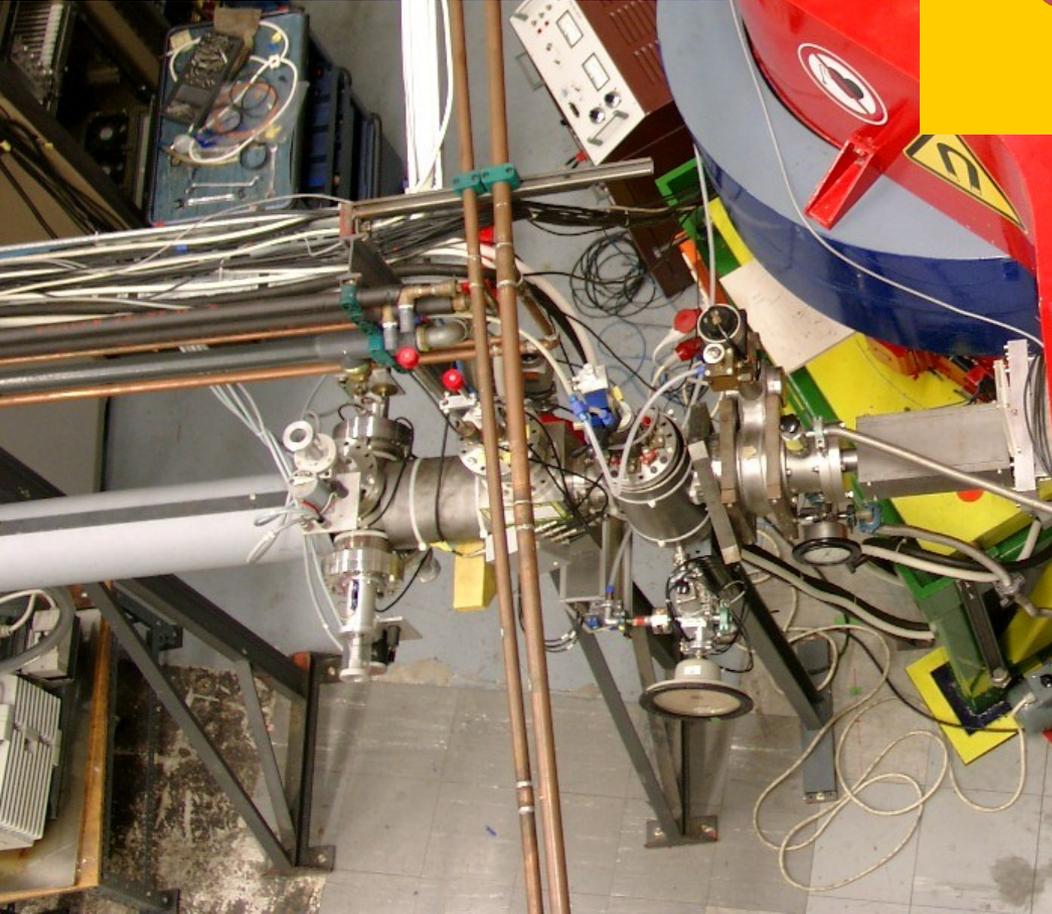
10^6 : ^{79}Se

10^8 : ^{53}Mn , ^{59}Ni , ^{63}Ni

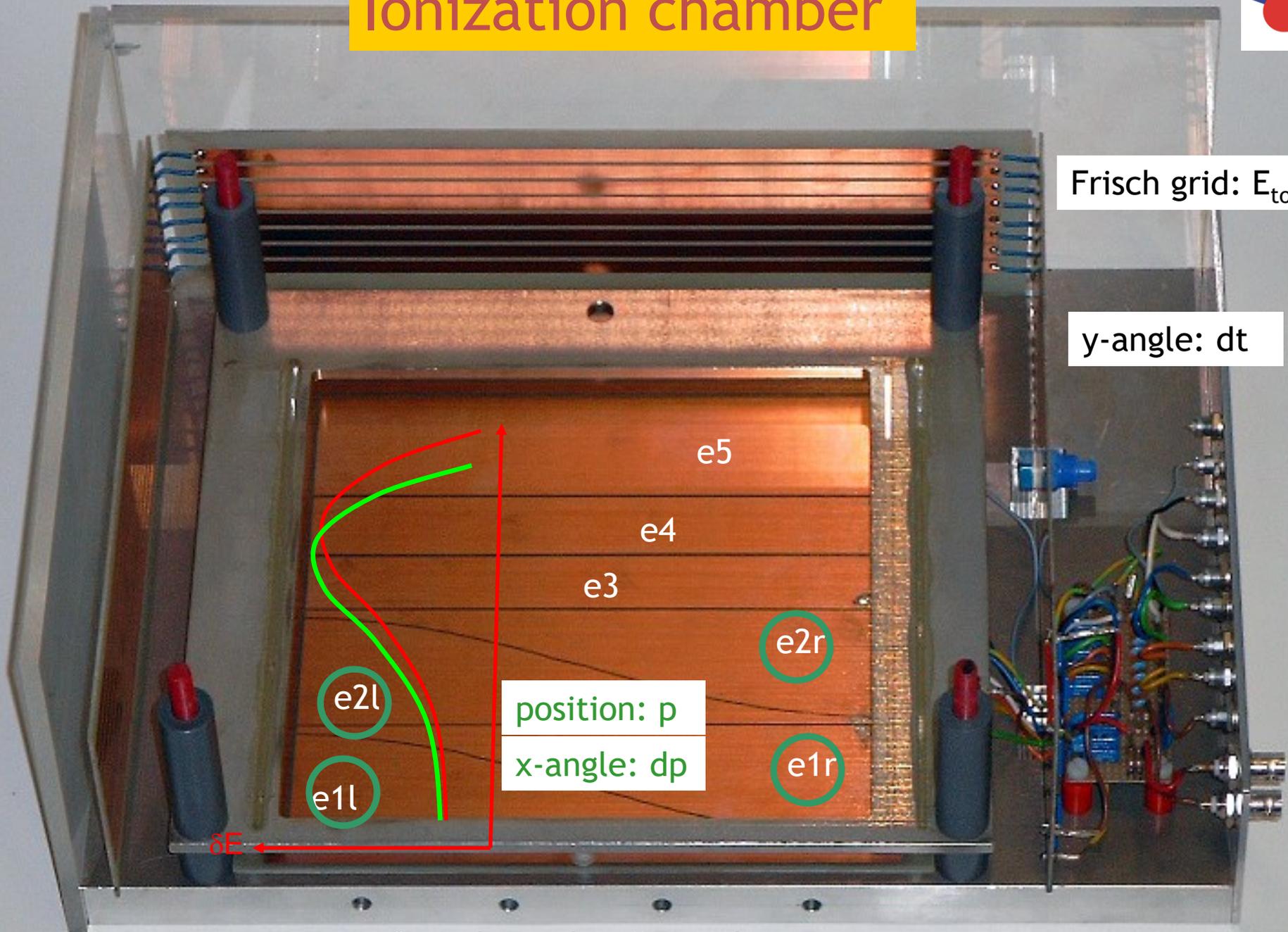
10^{11} : ^{60}Fe ($\Delta Z = 2$)



Gas-filled Analyzing Magnet System



Ionization chamber



Frisch grid: E_{tot}

y-angle: dt

e5

e4

e3

e2r

e2l

e1l

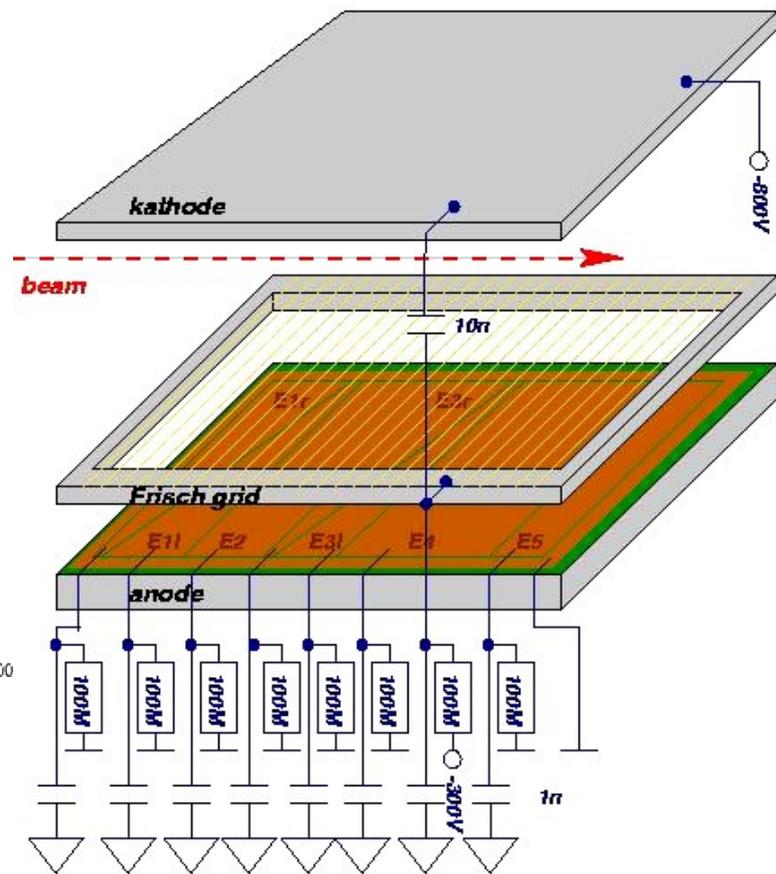
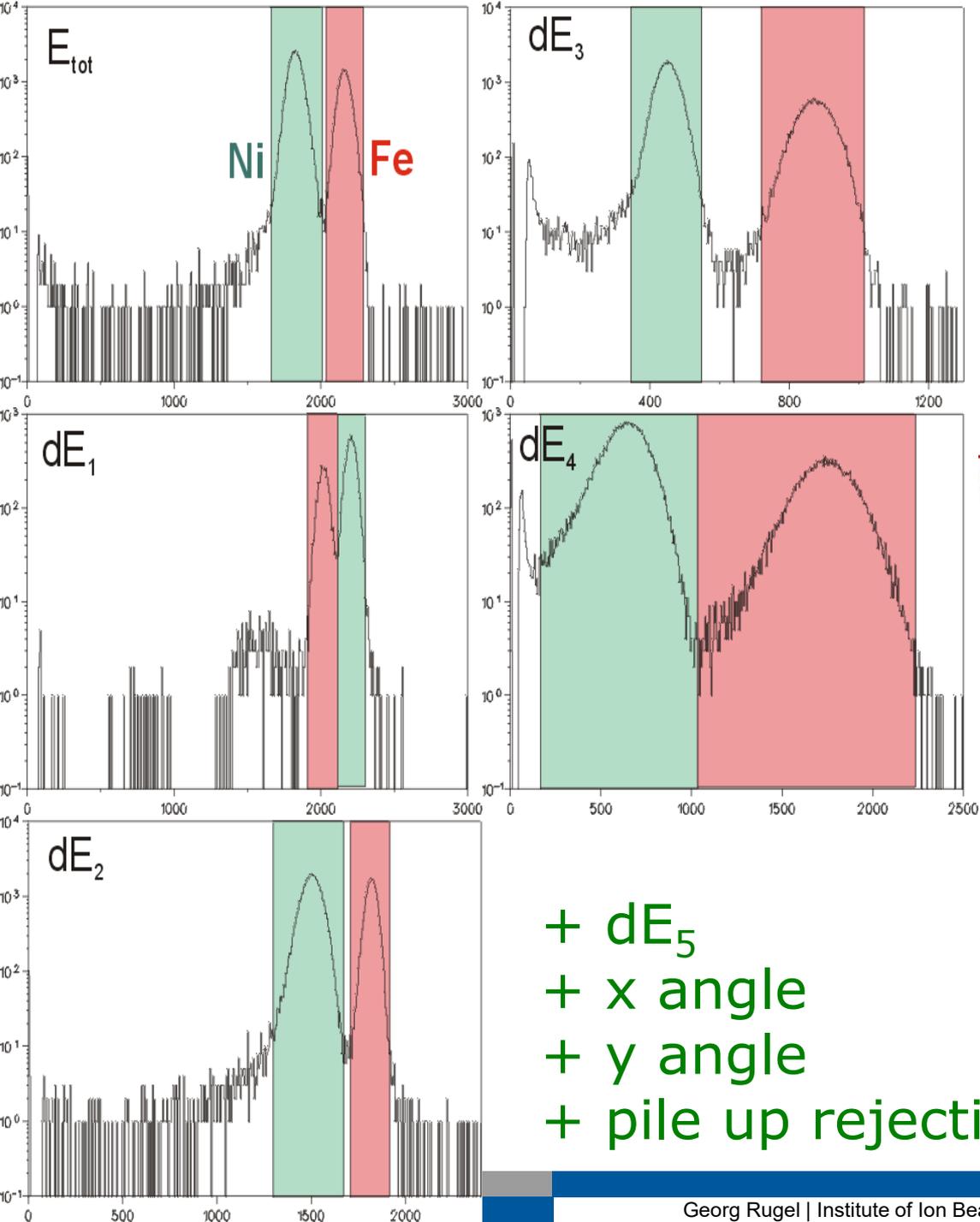
position: p

x-angle: dp

e1r

δE

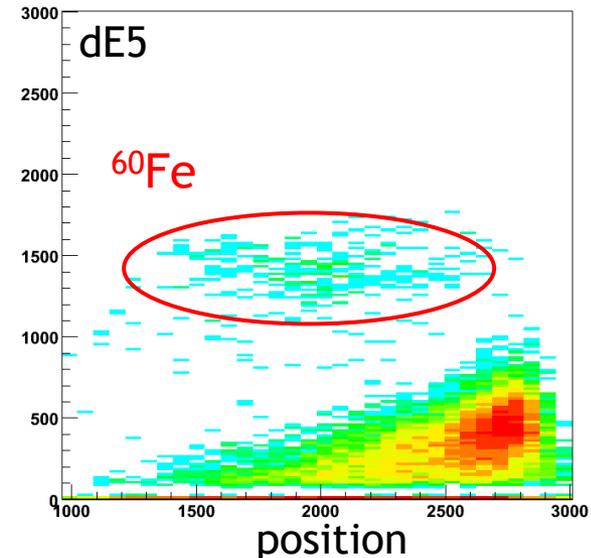
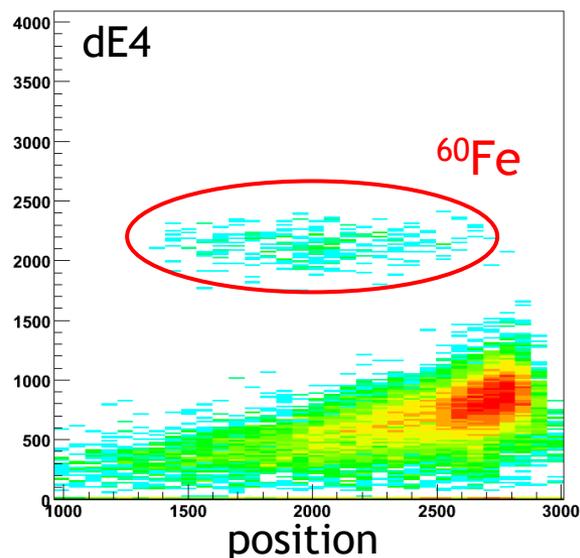
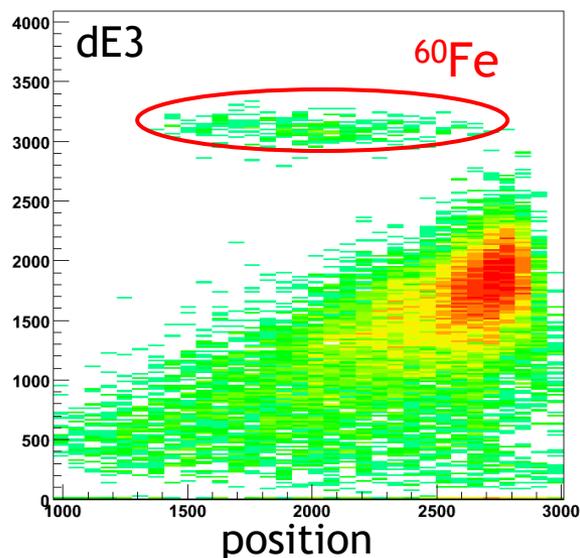
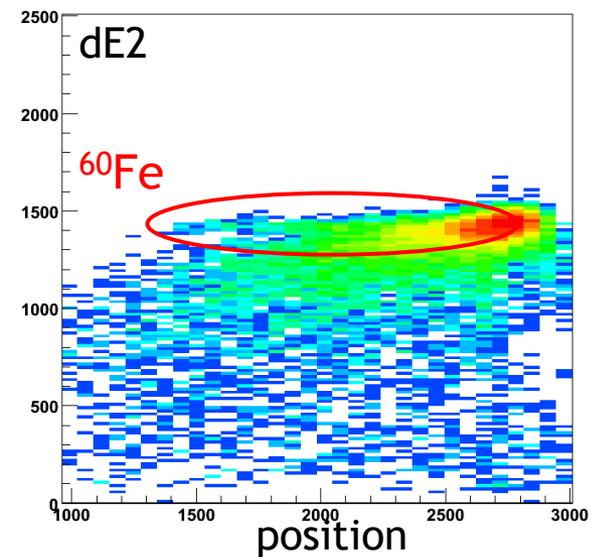
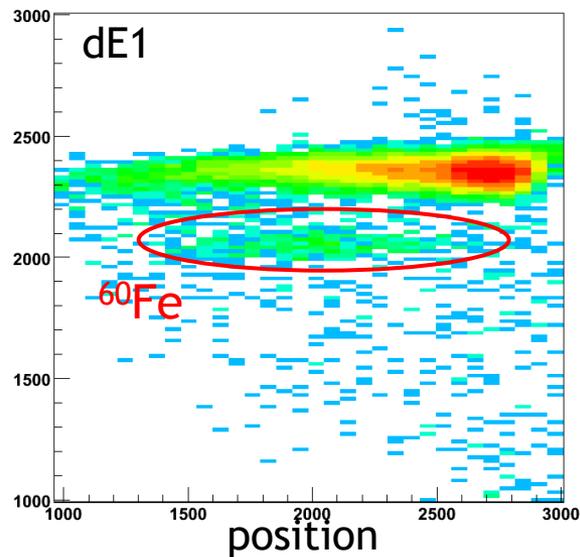
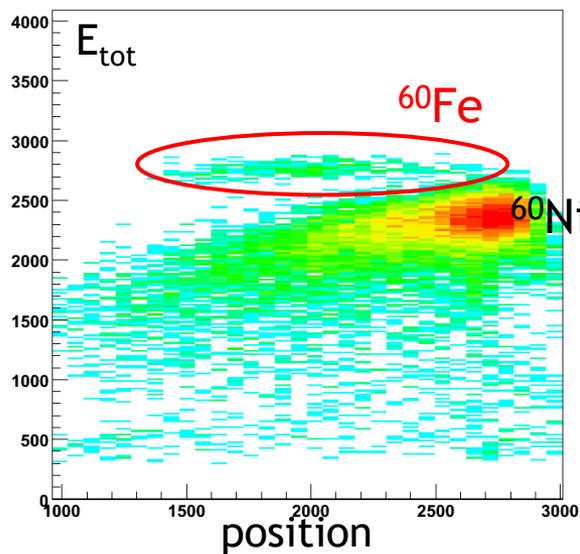
9 „independent“ signals



- + dE_5
- + x angle
- + y angle
- + pile up rejection

spectra for calibration; sample: $^{60}\text{Fe}/\text{Fe} = 5 \cdot 10^{-12}$

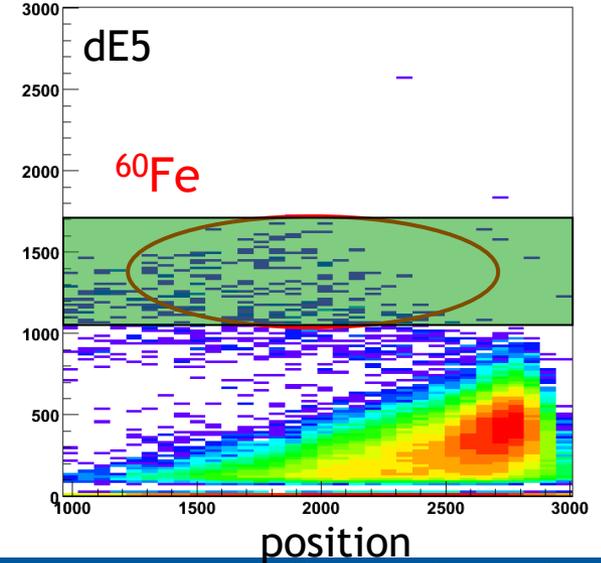
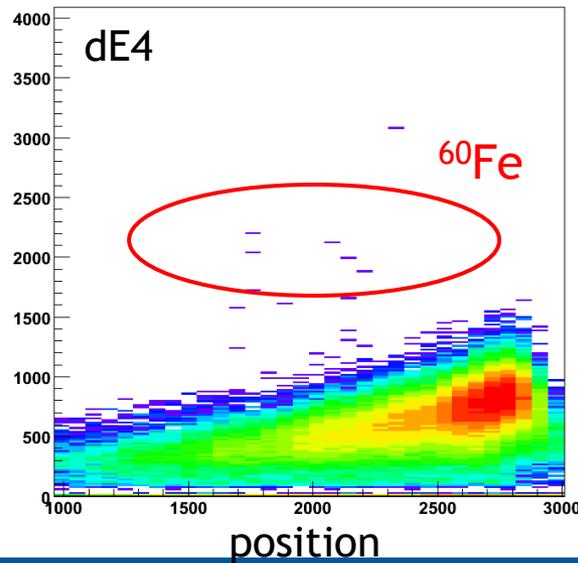
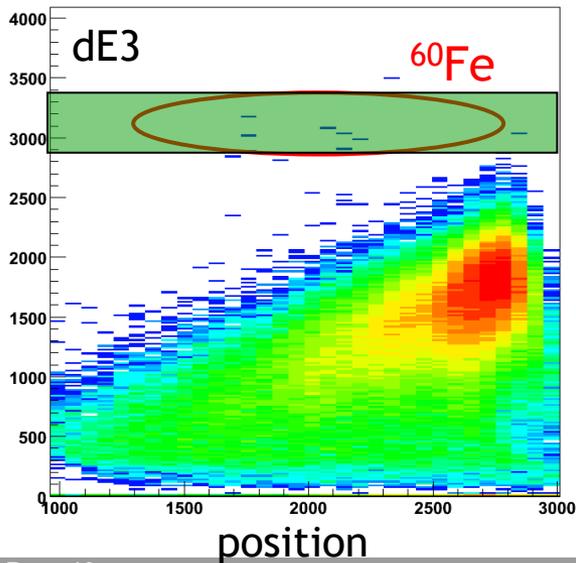
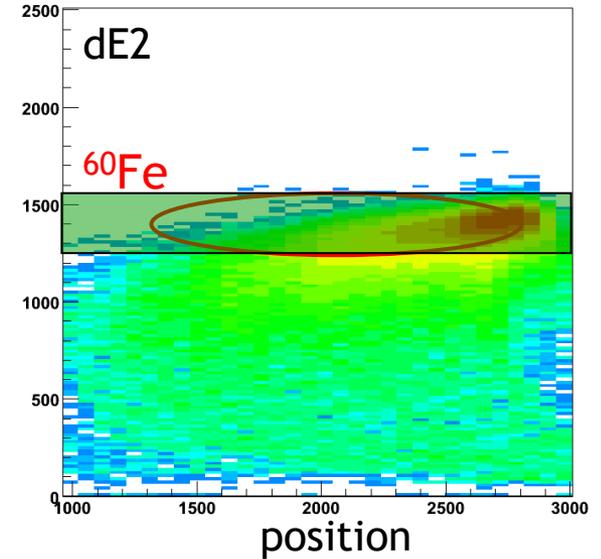
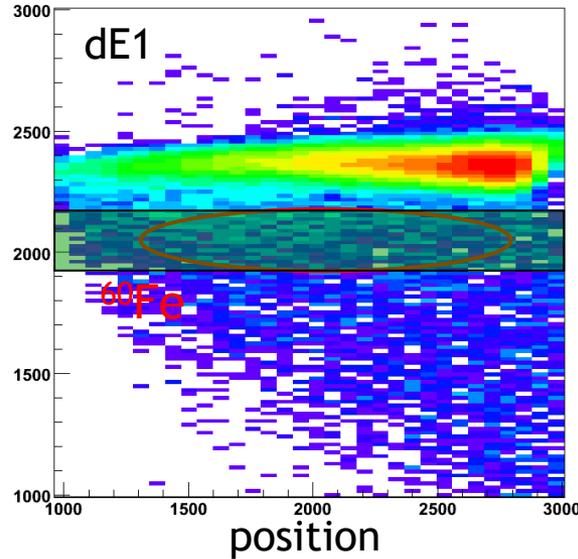
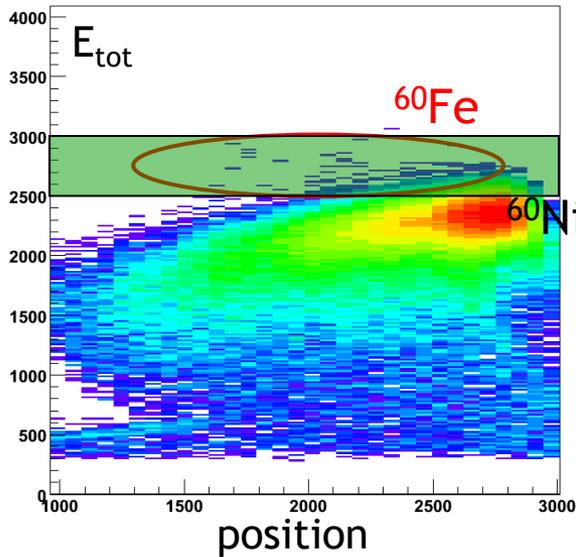
$$\text{Ni}/\text{Fe} \approx 10^{-5} \Rightarrow ^{60}\text{Ni}/^{60}\text{Fe} \approx 10^6$$



spectra for crust sample: $^{60}\text{Fe}/\text{Fe} \sim 2 \cdot 10^{-15}$

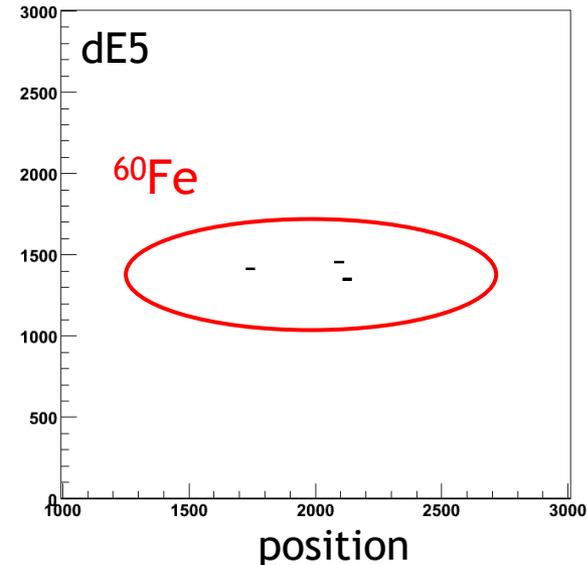
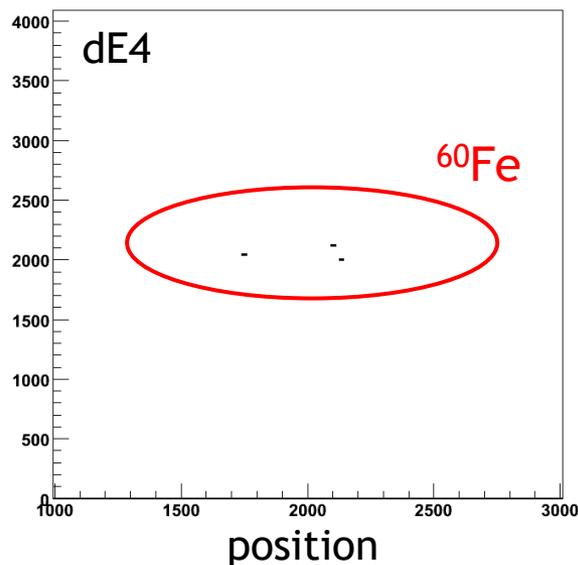
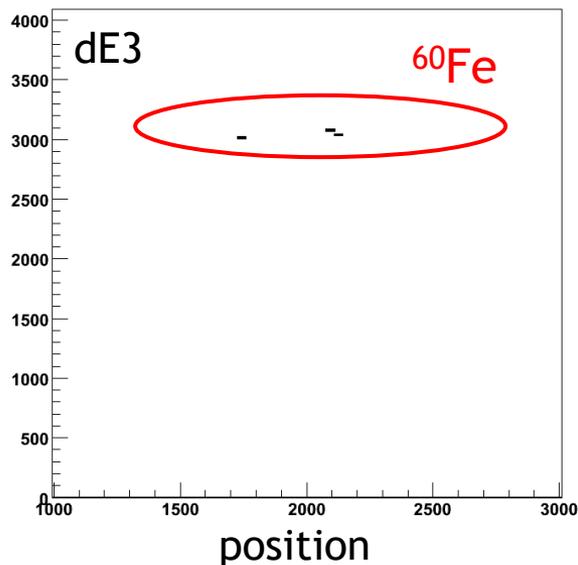
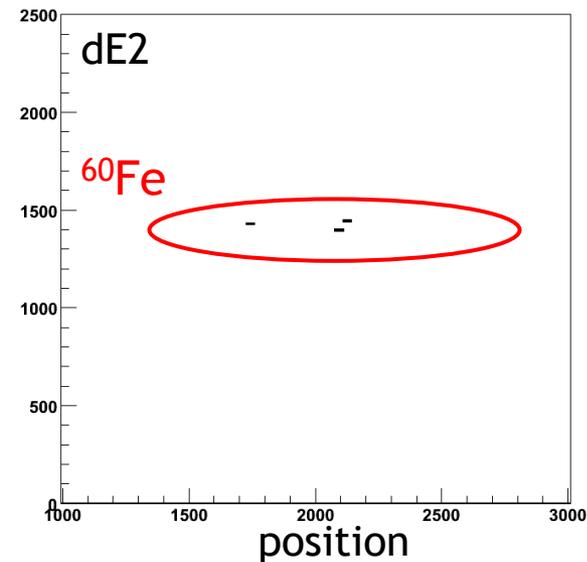
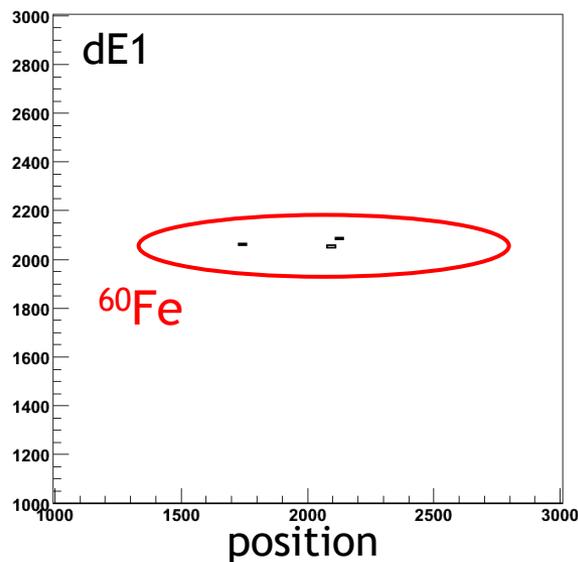
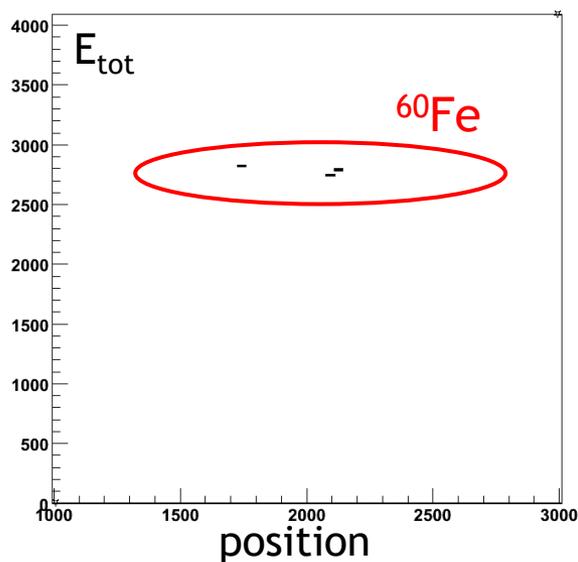


with windows



spectra for crust sample: $^{60}\text{Fe}/\text{Fe} = 1.4 \cdot 10^{-15}$

with windows



some numbers



1 $\mu\text{A } ^{56}\text{FeO}^-$ \Rightarrow $6 \cdot 10^{12} / \text{sec}$
transmission 5% \Rightarrow $3 \cdot 10^{11} / \text{sec}$
for $^{60}\text{Fe} / ^{56}\text{Fe} = 10^{-15}$ \Rightarrow $3 \cdot 10^{-4} / \text{sec} = 1 \text{ event/hour}$

10 mg ^{56}Fe \Rightarrow 10^{20} atoms
1 $\mu\text{A } ^{56}\text{FeO}^-$ \Rightarrow $6 \cdot 10^{12}$ atoms/sec
neg. ion yield 10^{-3} \Rightarrow $6 \cdot 10^{15}$ atoms/sec used
sample lasts for \Rightarrow $1.5 \cdot 10^4 \text{ sec} = 4 \text{ hours}$

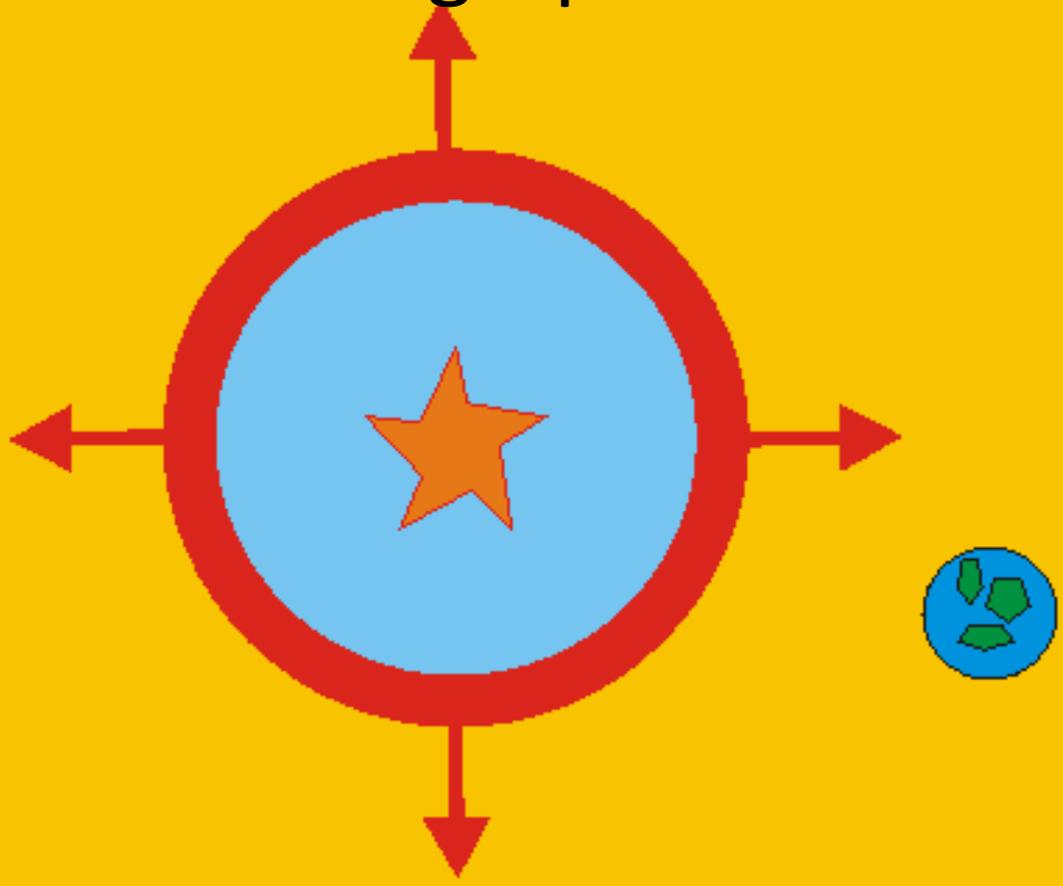
Slide from Thomas Faestermann

Supernova fingerprints on Earth?

Supernova fingerprints on Earth?



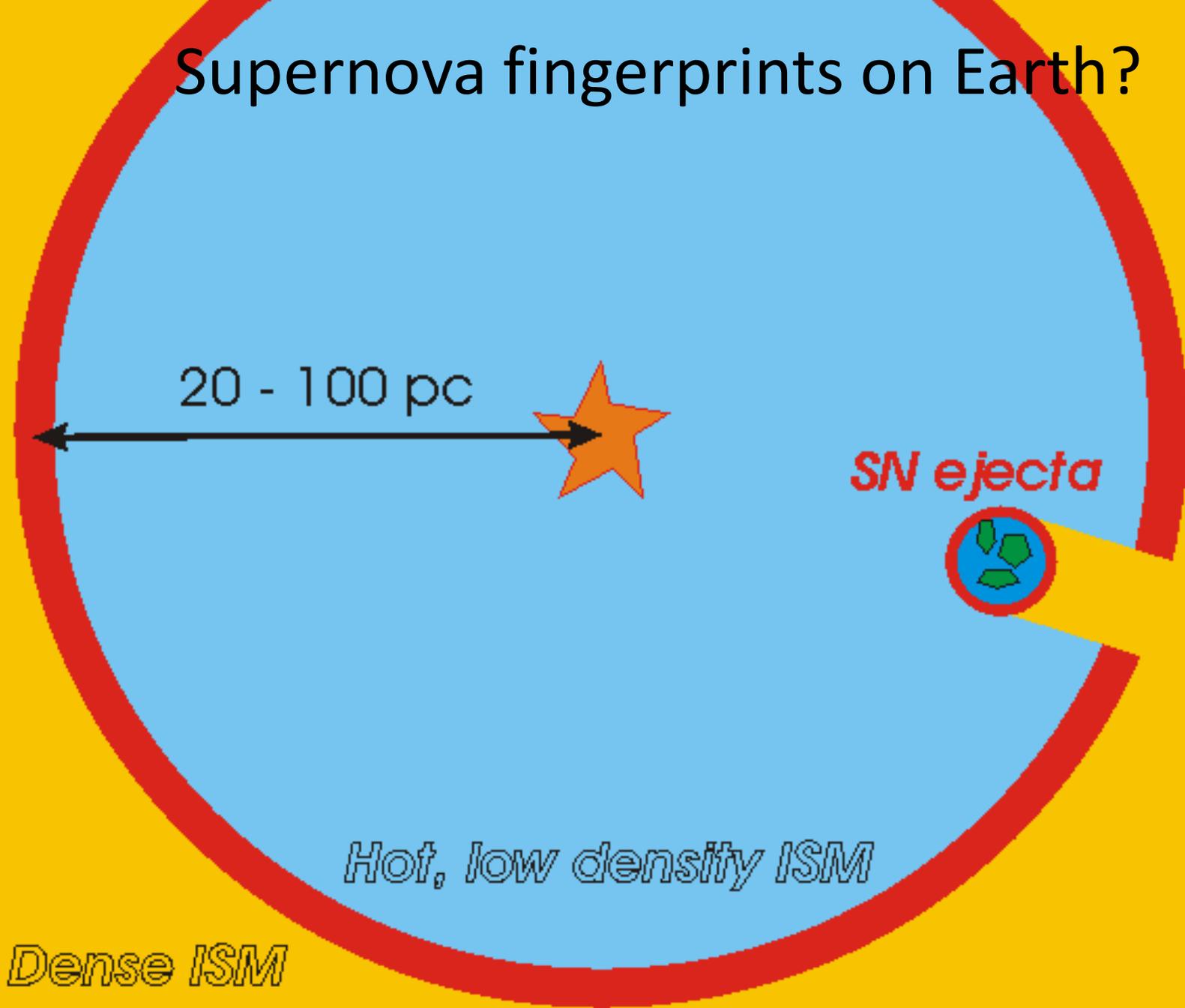
Supernova fingerprints on Earth?



Supernova fingerprints on Earth?

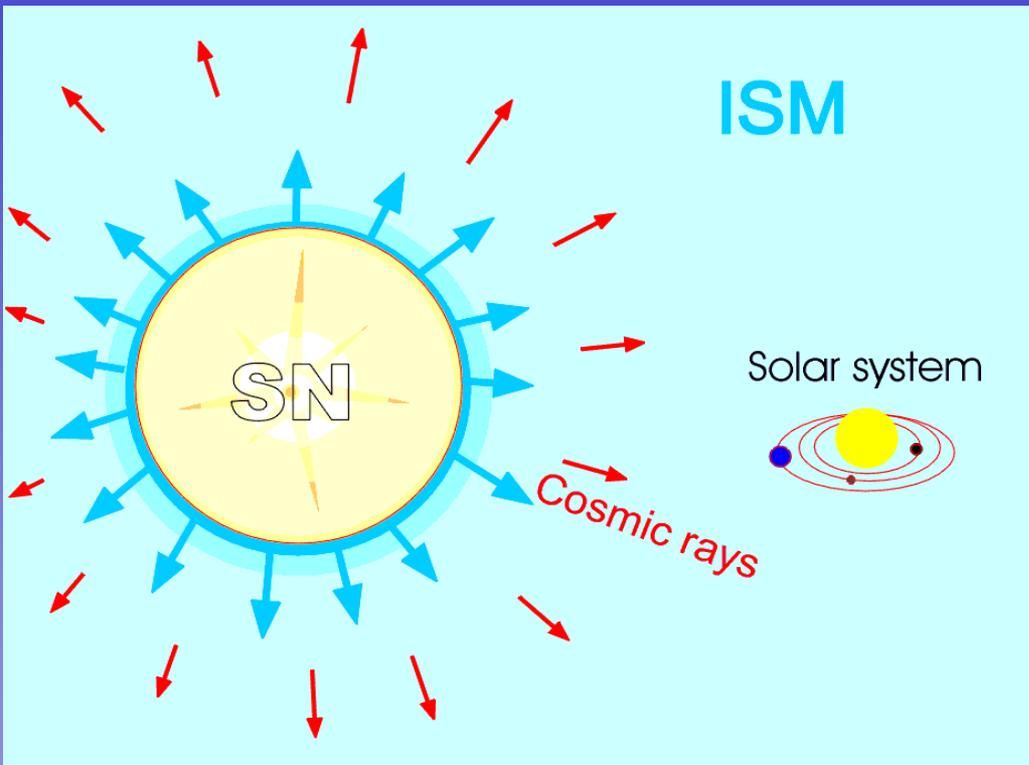


Supernova fingerprints on Earth?





Live radioactivity as signature of nearby SNe



Search for long-lived, SN produced radionuclides with negligible abundance inside the solar system:

^{10}Be , ^{26}Al , ^{36}Cl , ^{53}Mn , ^{59}Ni ,
 ^{60}Fe , ^{129}I , ^{146}Sm , ^{244}Pu

Ellis et al. ApJ 470(1996)1227,
Korschinek et al., Radiocarbon 38(1996)68

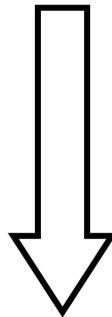
Some radionuclides measured with AMS

^{55}Fe	2.7 years
^3H	12.3
^{44}Ti	60
^{63}Ni	100
^{32}Si	140

Independent of half-life

^{14}C	5 / 30
^{59}Ni	75 000

fundamental physics Independent of sample mass



applied sciences

^{81}Kr	230 000
^{79}Se	280 000
^{36}Cl	301 000
^{26}Al	720 000
^{10}Be	1 388 000
^{60}Fe	2 600 000
^{53}Mn	3 600 000
^{182}Hf	8 900 000
^{129}Xe	15 000 000

atom counting of radionuclides via isotope ratio measurements

$^{239-244}\text{Pu}, ^{247}\text{Cm}$ - 81 000 000

new: $^{55}\text{Fe}, ^{68}\text{Ge}, ^{93}\text{Zr}, ^{106}\text{Pd}, ^{135}\text{Cs}, ^{146}\text{Sm},$
 $^{202}\text{Pb}, ^{210}\text{Bi}, ^{226}\text{Ra}, ^{229}\text{Th},$ stable, ...

Slide from Anton Wallner

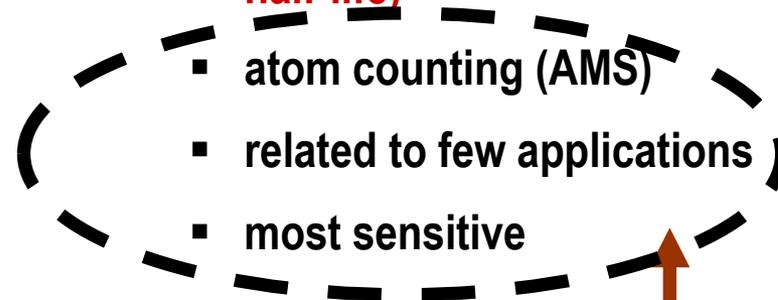
Measurement of Cross Sections

online method

- detection of prompt γ -rays of reaction product (characteristic)
- or: recoil separator technique
- if continuous E-distribution: + TOF
- all reaction products accessible
- radioactive beams

offline method

- activation technique:
 - Sample irradiation
 - Determination of reaction product
- quasi-stellar spectrum at once
- sensitive technique
 - detects decay products
 - needs radioactive samples (not too long half-life)



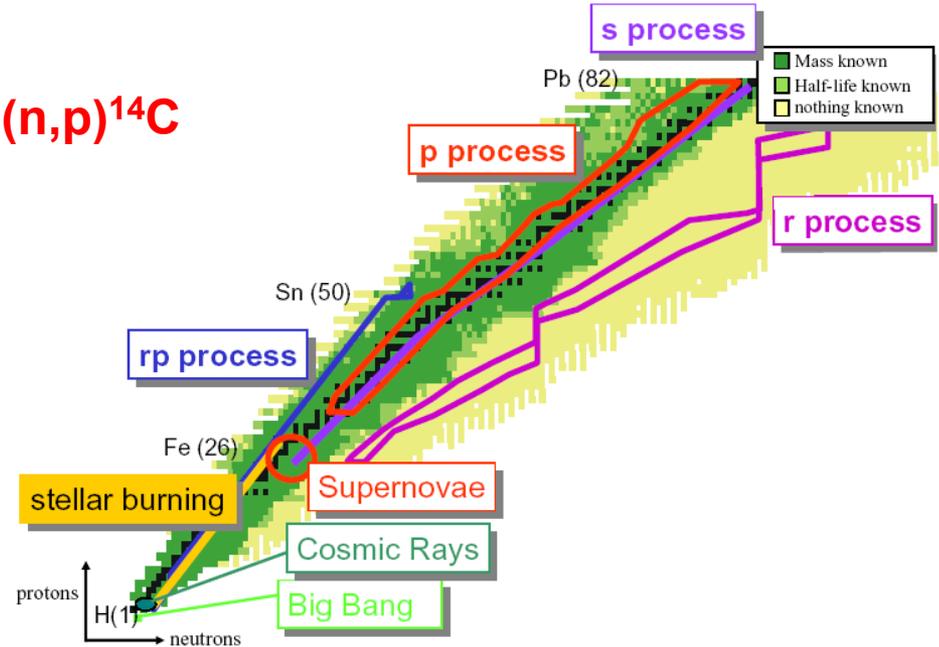
Different systematic uncertainties involved

Slide from Anton Wallner

Applications of AMS to Astrophysics

Nuclear reaction data - cross-section measurements

- ${}^9\text{Be}(n, \gamma){}^{10}\text{Be}$, ${}^{13}\text{C}(n, \gamma){}^{14}\text{C}$, ${}^{14}\text{N}(n, p){}^{14}\text{C}$
- ${}^{35}\text{Cl}(n, \gamma){}^{36}\text{Cl}$, ${}^{40}\text{Ca}(n, \gamma){}^{41}\text{Ca}$
- ${}^{54}\text{Fe}(n, \gamma){}^{55}\text{Fe}$, ${}^{58}\text{Ni}(n, \gamma){}^{59}\text{Ni}$,
 ${}^{62}\text{Ni}(n, \gamma){}^{63}\text{Ni}$, ${}^{78}\text{Se}(n, \gamma){}^{79}\text{Se}$,
- ${}^{209}\text{Bi}(n, \gamma){}^{210}\text{Bi}$,
-



Nucleosynthesis of elements in stars

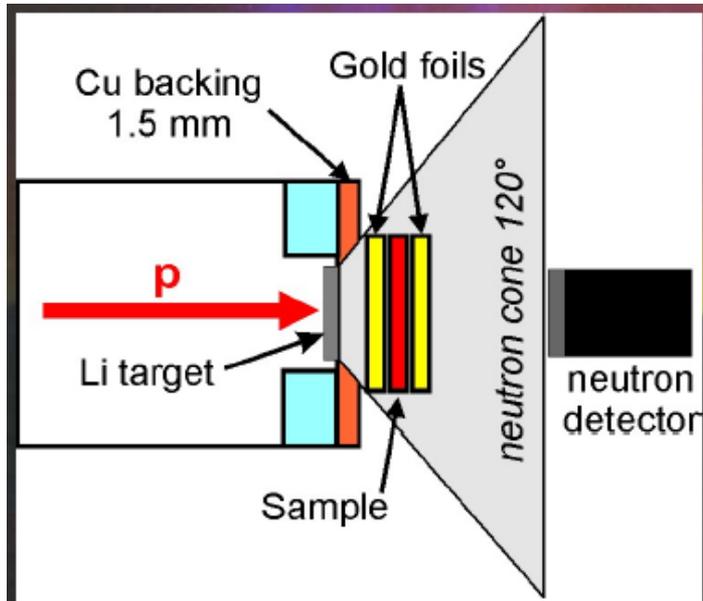
all reactions: identical irradiation geometry

Slide from Anton Wallner

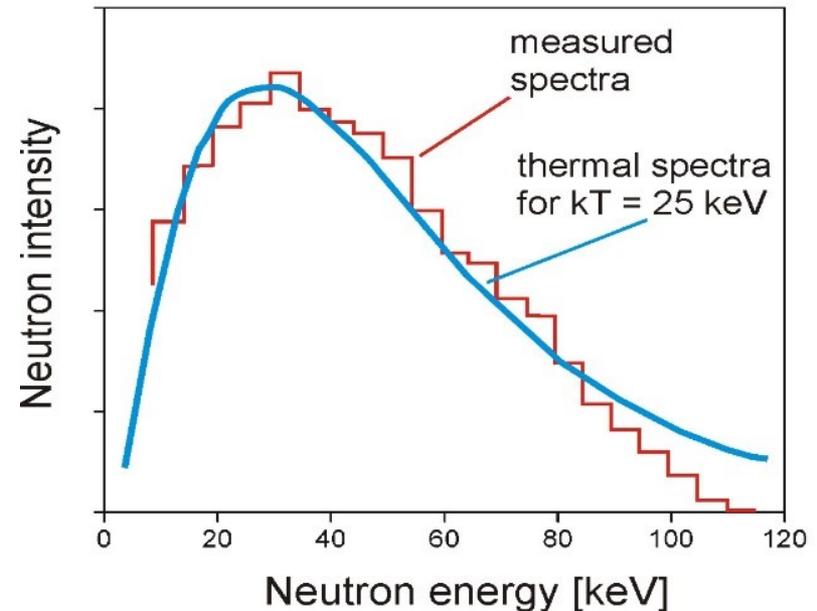
Maxwellian averaged cross sections

Experiment KIT

- (1) Activation
- (2) Offline – AMS measurement



same conditions for all experiments

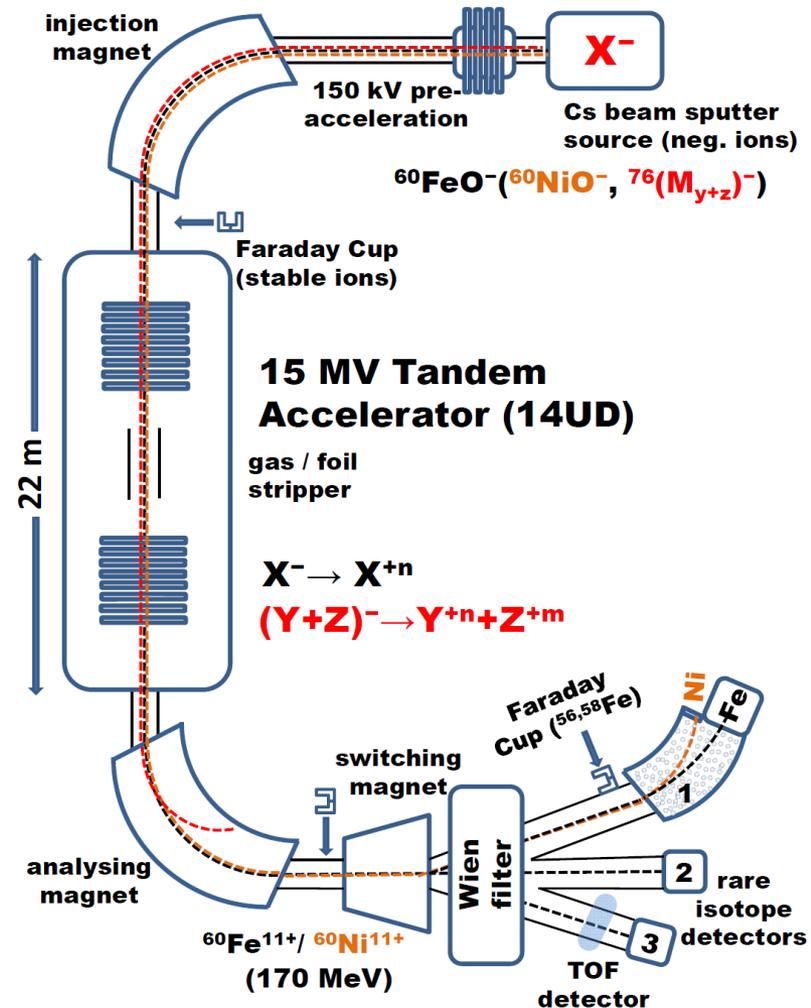


Slide from Anton Wallner

AMS basics

- **AMS determines isotope ratios – atom counting technique**
- **$^{14}\text{C}/^{12}\text{C}$ – radiocarbon dating**
- **highest sensitivity: 10^{-12} – 10^{-16}**
- **no isobaric background (<-> ICPMS) (molecules are completely destroyed)**
- **isotopic background clearly identified**

...radionuclides



Slide from Anton Wallner

New group
Prof. Anton Wallner at TU Dresden
with focus on
AMS
and astrophysics

Anton Wallner's group at HZDR



2022-03-01