Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Progress in Accelerator Mass Spectrometry

How far have we travelled?
How far can we get?

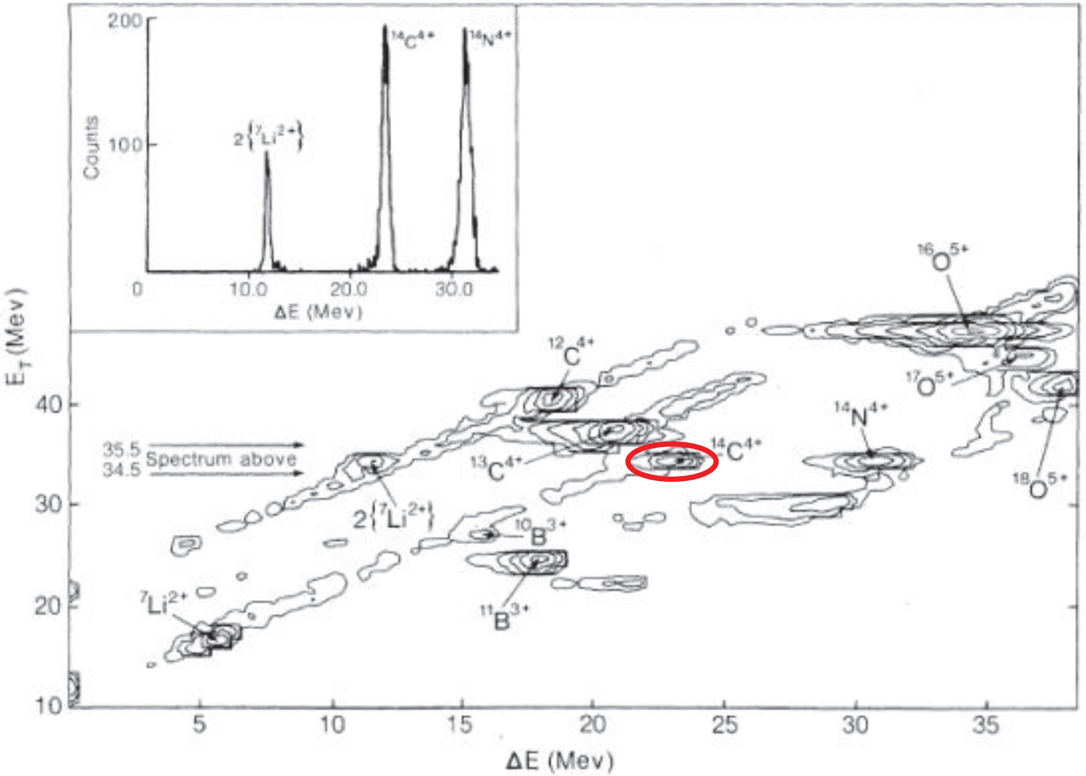
Hans-Arno Synal
Laboratory of Ion Beam Physics
ETH Zurich

synal@phys.ethz.ch

40 years ago: Science vol.198 p. 507-508

Carbon-14: Direct Detection at Natural Concentrations

Abstract. *The ^{14}C atoms naturally present in a piece of 19th-century wood have been detected directly by means of a tandem Van de Graaff accelerator used as a high-energy mass spectrometer. The ^{14}C ions were easily resolved from interfering ions with the use of a ΔE-E detector telescope (this telescope consists of a pair of detectors; one of them measures the specific ionization, ΔE, and the sum of the signals from both detectors gives the total energy for each ion, E_T). The technique offers a number of practical advantages.*

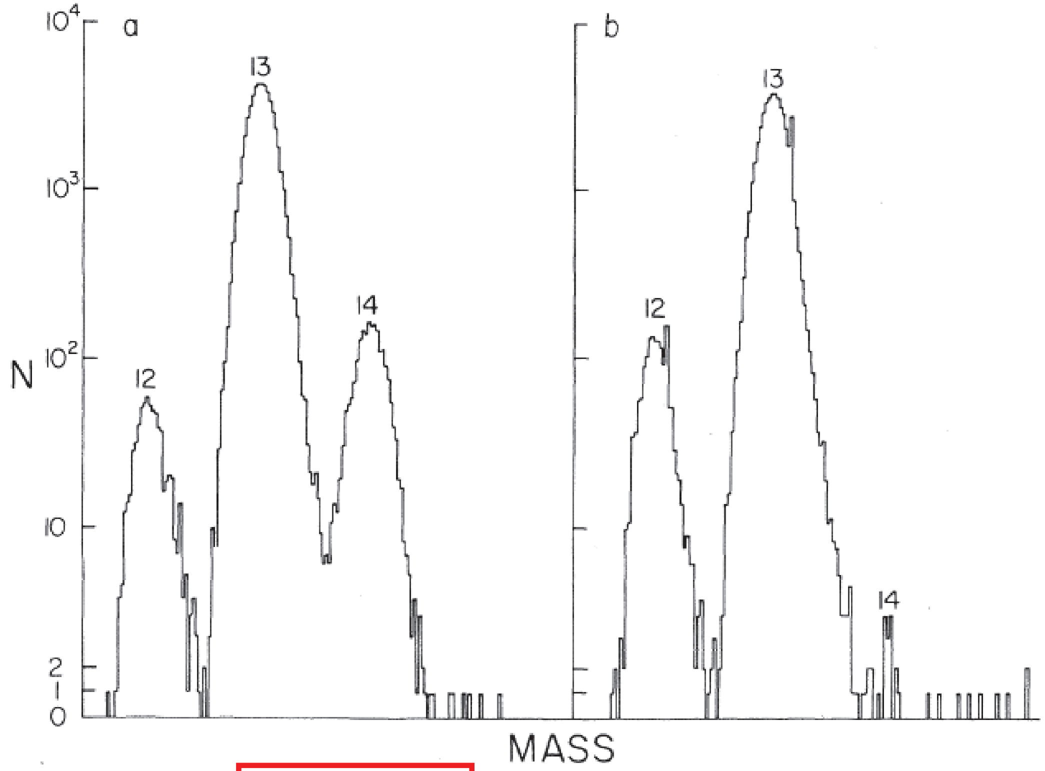


29 June 1977; revised 15 September 1977

Simon Fraser/ MacMaster University
D. E. Nelson; R. G. Korteling; W. R. Stott

Radiocarbon Dating Using Electrostatic Accelerators: Negative Ions Provide the Key

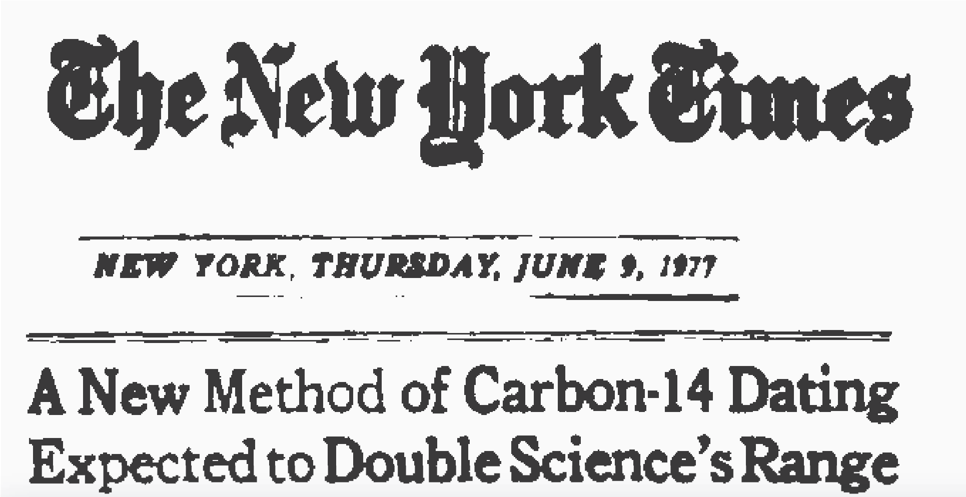
Abstract. *Mass spectrometric methods have long been suggested as ways of measuring $^{14}C/^{12}C$ ratios for carbon dating. One problem has been to distinguish between ^{14}N and ^{14}C. With negative ions and a tandem electrostatic accelerator, the ^{14}N background is virtually absent and fewer than three ^{14}C atoms in 10^{16} atoms of ^{12}C have been easily measured.*



30 June 1977; revised 2 September 1977

University of Rochester
C. L. Bennett; R. P. Beukens; M. R. Clover; H. E. Gove;
R. B. Liebert; A. E. Litherland; K. H. Purser; W. E. Sondheim

Discoverers of Accelerator Mass Spectrometry (1977)


The New York Times

NEW YORK, THURSDAY, JUNE 9, 1977

A New Method of Carbon-14 Dating Expected to Double Science's Range

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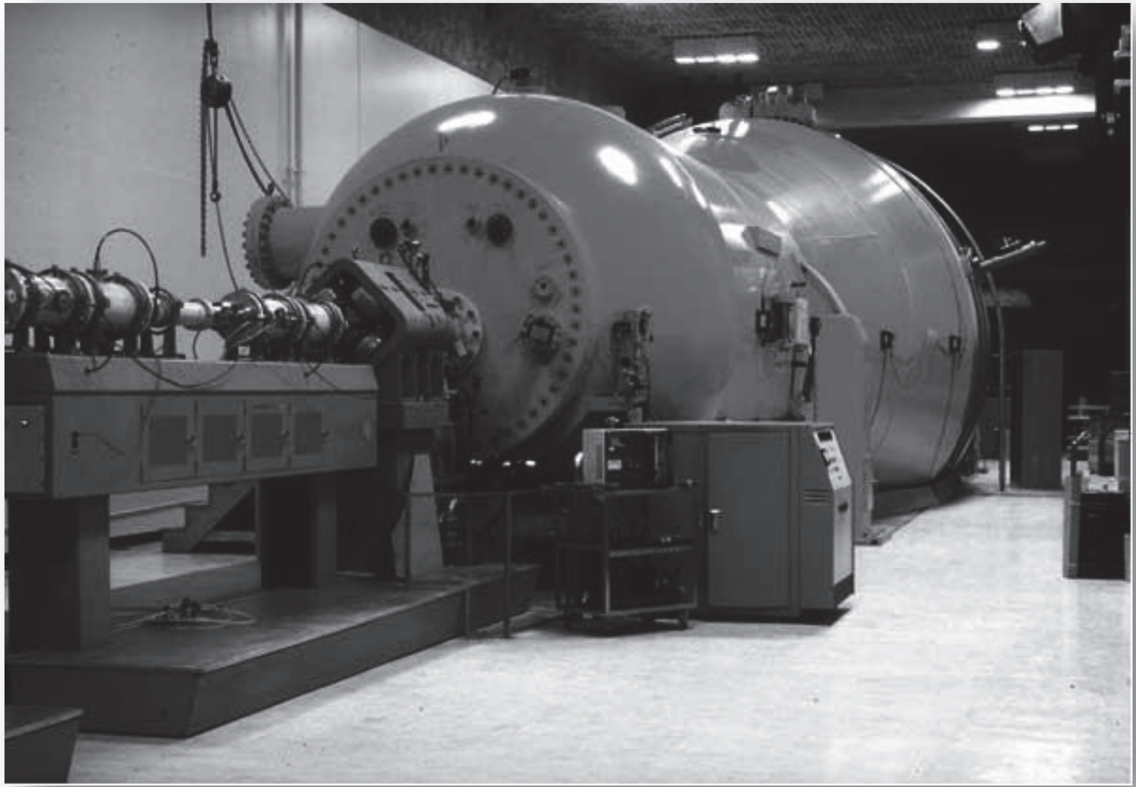
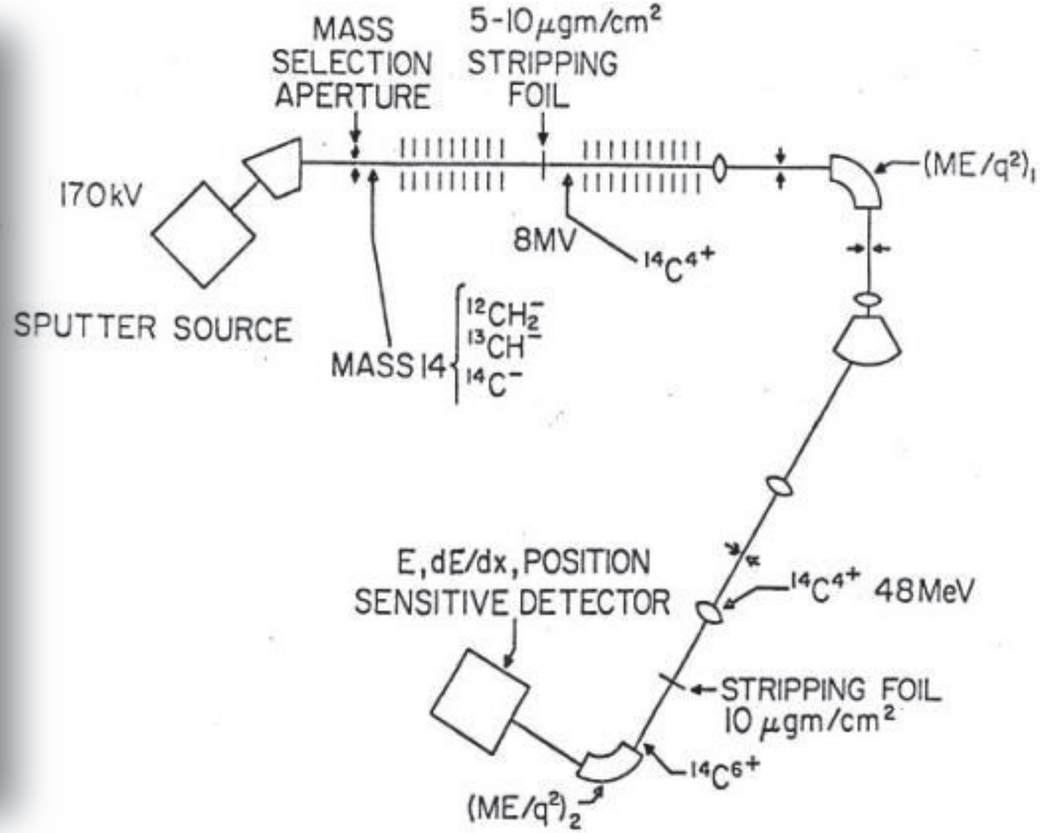


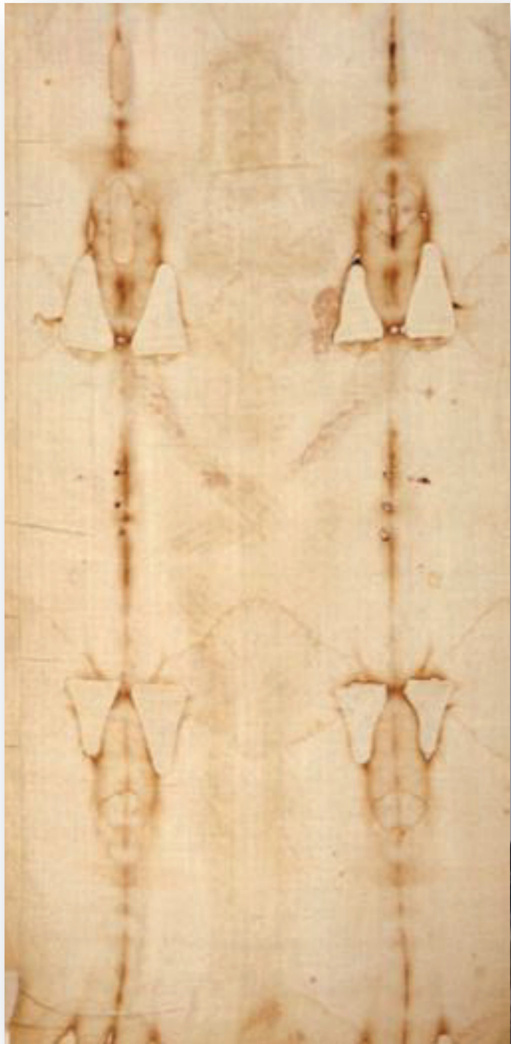
Rochester, NY(USA) MP tandem accelerator



Famous examples of Radiocarbon dating

Buddha's of Bamiyan Afghanistan

The Shroud of Turin

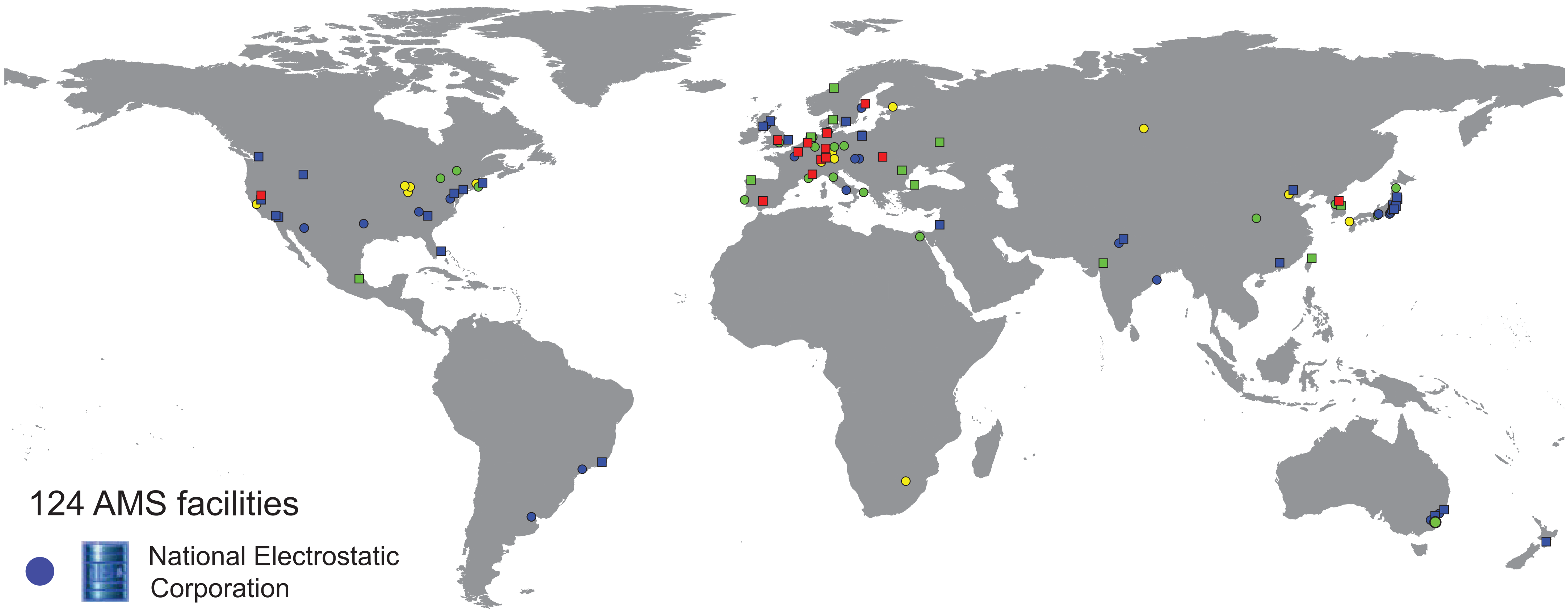


"Özi" the Ice Man

Art historic object

The Minoan Eruption (Santorini)

The World of AMS facilities in 2016



124 AMS facilities

- National Electrostatic Corporation
- High Voltage Engineering Europe
- ETH Zurich Ionplus engineering scientific instruments
- Large Tandem / exotic systems

Commercial AMS systems

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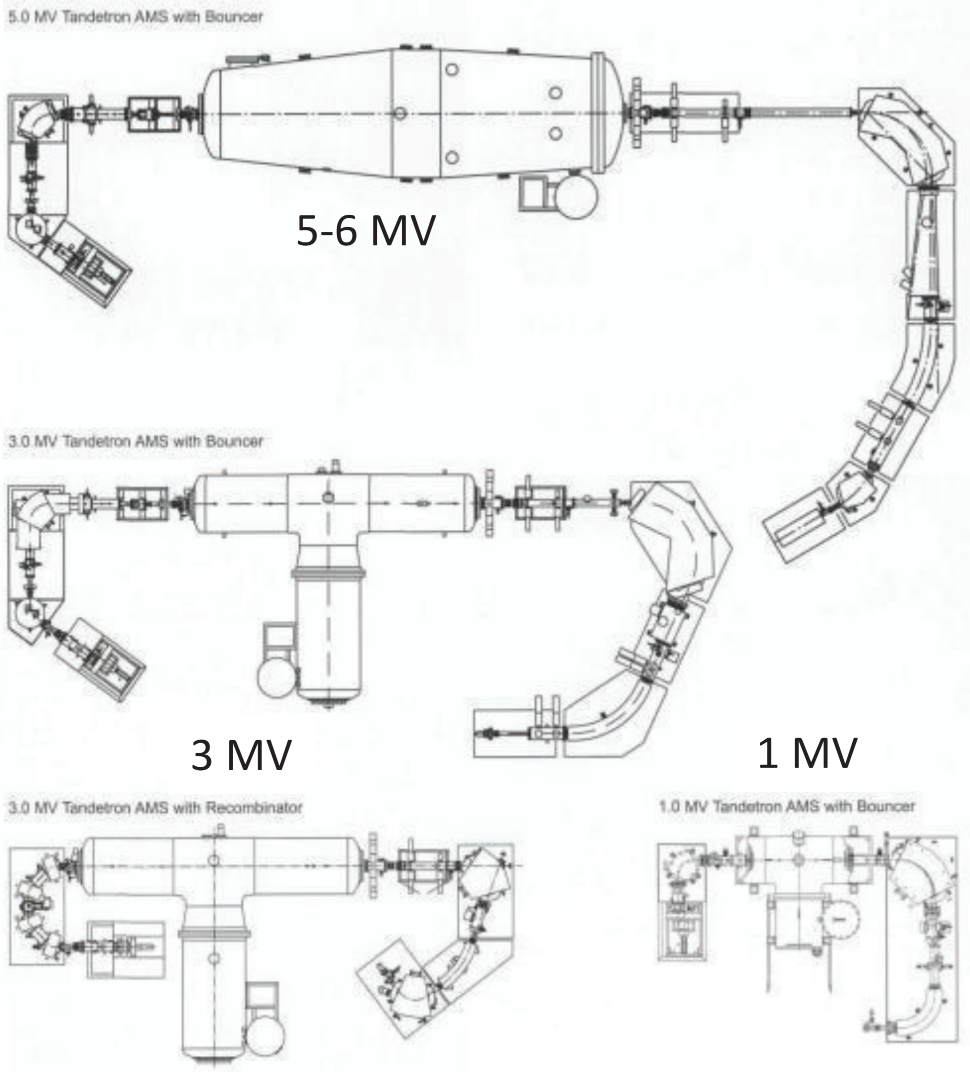
Swiss Federal Institute of Technology Zurich

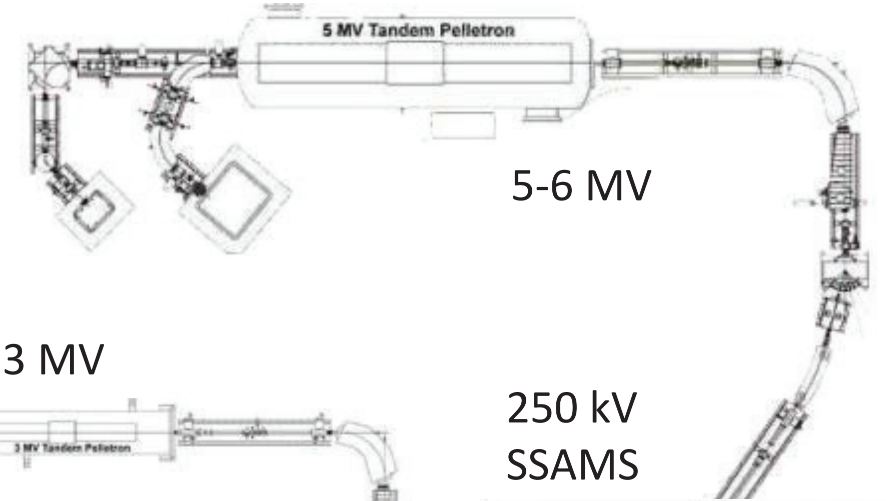
HIGH VOLTAGE ENGINEERING EUROPA B.V.

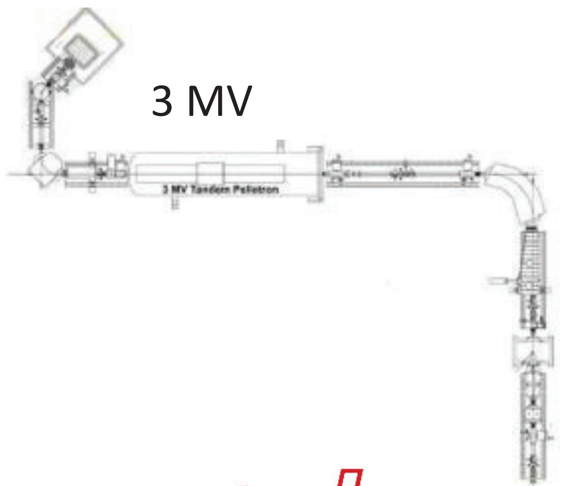
Amsterdamseweg 63, 3812 RR Amersfoort, P.O.Box 99, 3800 AB Amersfoort, The Netherlands
Phone: +31 33 4619741 Fax: +31 33 4615291 E-mail: info@highvolteng.com Web: www.highvolteng.com

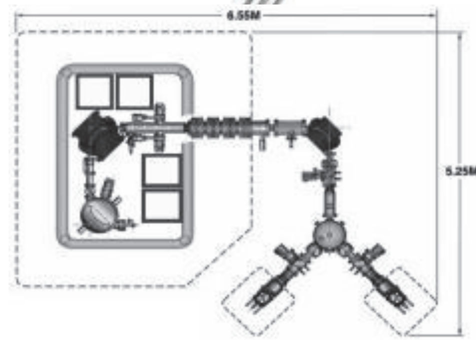
System layouts



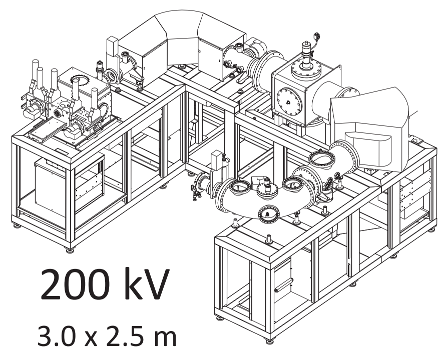
National Electrostatics Corporation
Middleton, Wisconsin, USA

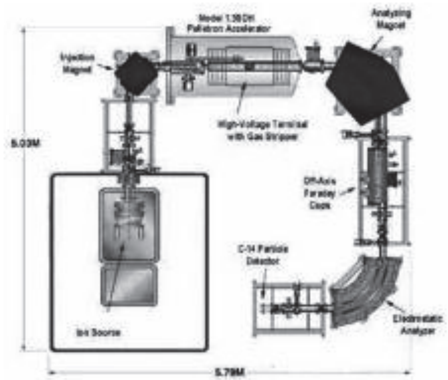




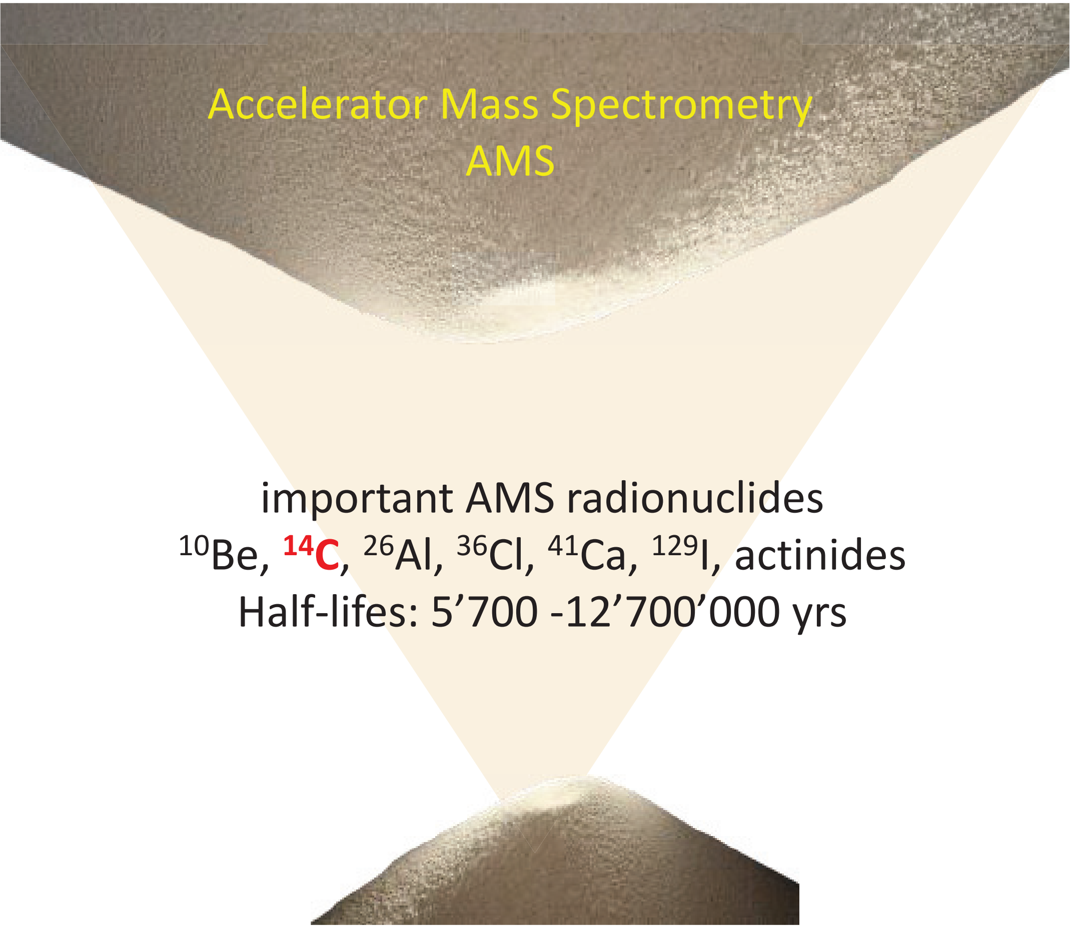


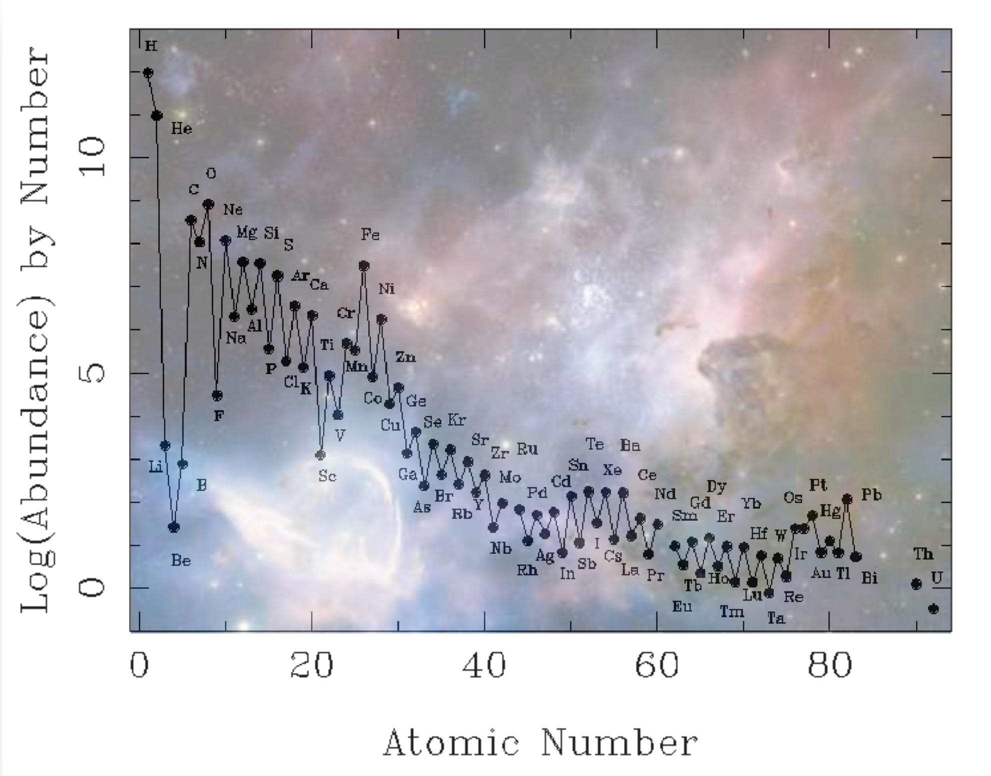
Ionplus
engineering scientific instruments





Accelerator Mass Spectrometry a unique detection technique





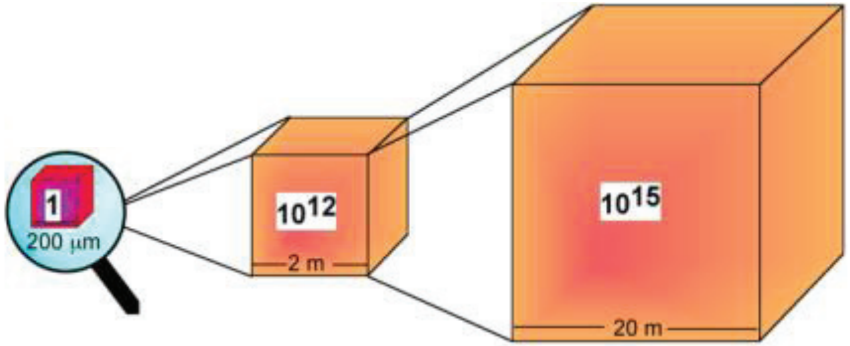
- Single atom detection capabilities
- High accuracy (‰)
- Huge dynamic range (10^4)
- Extreme sensitivity

  Isotope ratios: 10^{-6} - 10^{-15}

Sample:

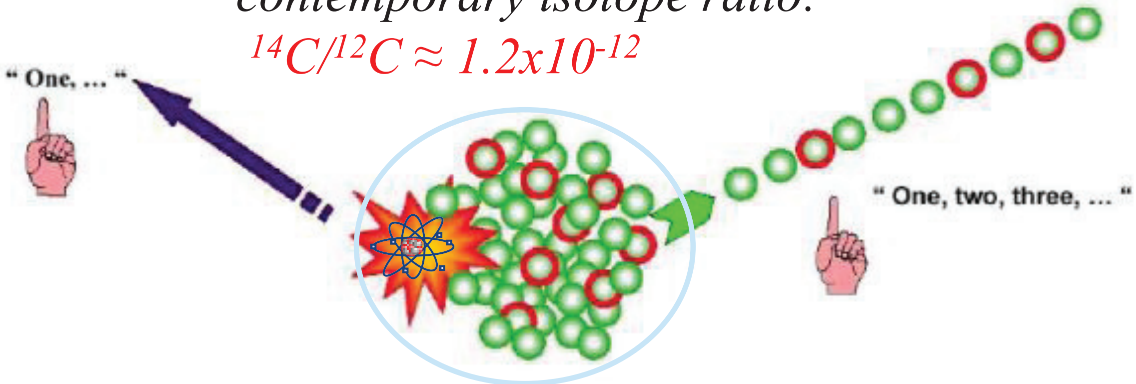
Typical size: mg → µg

10^4 → million atoms



Counting Atoms

carbon sample
contemporary isotope ratio:
$^{14}C/^{12}C \approx 1.2x10^{-12}$



Decay counting
(Libby)

$$N(t) = N_0 \times e^{-\lambda t}; \quad \lambda = \frac{\ln(2)}{T_{1/2}}$$

$$A(t) = \frac{dN(t)}{dt} \quad \Rightarrow \quad A(t) = N(t) \times \lambda$$

$$T_{1/2} = 5730\ a$$

$$\left.\begin{array}{l} \lambda = 3.84 \times 10^{-12}/s \\ t_{count} \approx 10^6 s \end{array}\right\} \Rightarrow \varepsilon_{tot} \approx 4 \times 10^{-6}$$

Counting atoms
(AMS)

$$N_{AMS} = N \times \varepsilon_{tot}$$

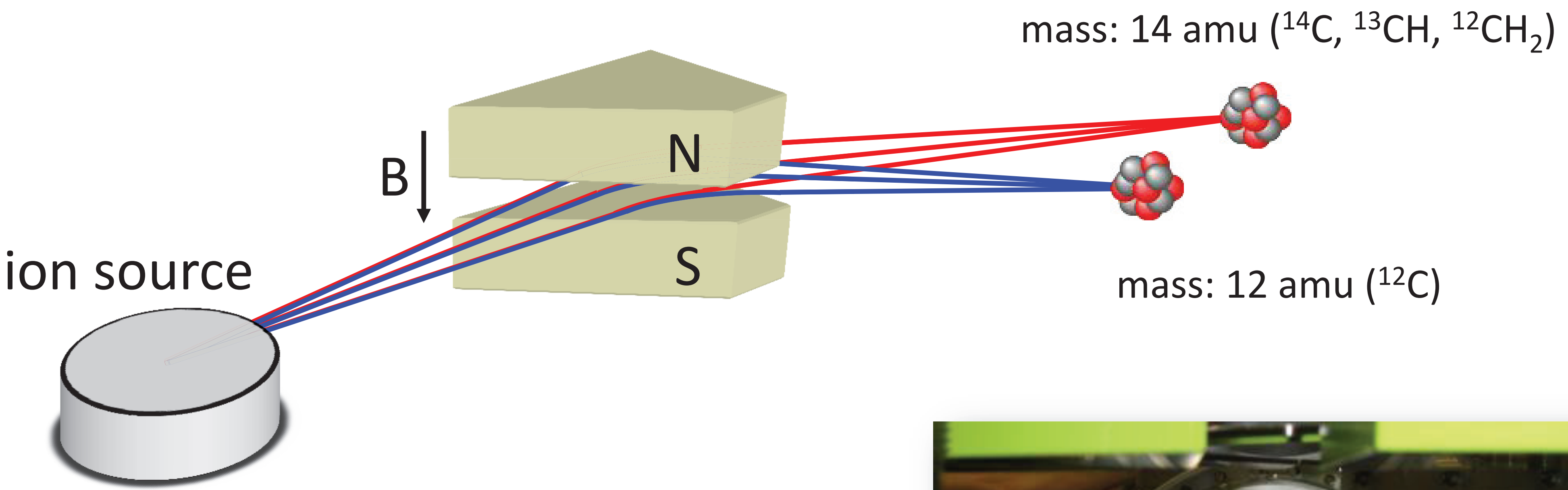
$$\varepsilon_{tot} = \varepsilon_{ion} \times T \times \varepsilon_{det}$$

$$\left.\begin{array}{l} \varepsilon_{ion} \approx 10\% \\ \varepsilon_{det} \approx 100\% \\ T \approx 40-50\% \end{array}\right\} \Rightarrow \varepsilon_{tot} \approx 4-5\%$$

At least 4 orders of magnitude better!!!

(g) ← ***Sample size*** → (mg → µg)

mass spectrometer



isobar/ molecule	mass (MeV/c²)	Δ mass (MeV/c²)	m/Δm
^{14}C	13044.0422		
^{14}N	13043.8861	0.1561	83562
$^{12}CH_2$	13055.6004	11.5582	1129
^{13}CH	13051.4364	7.3942	1766

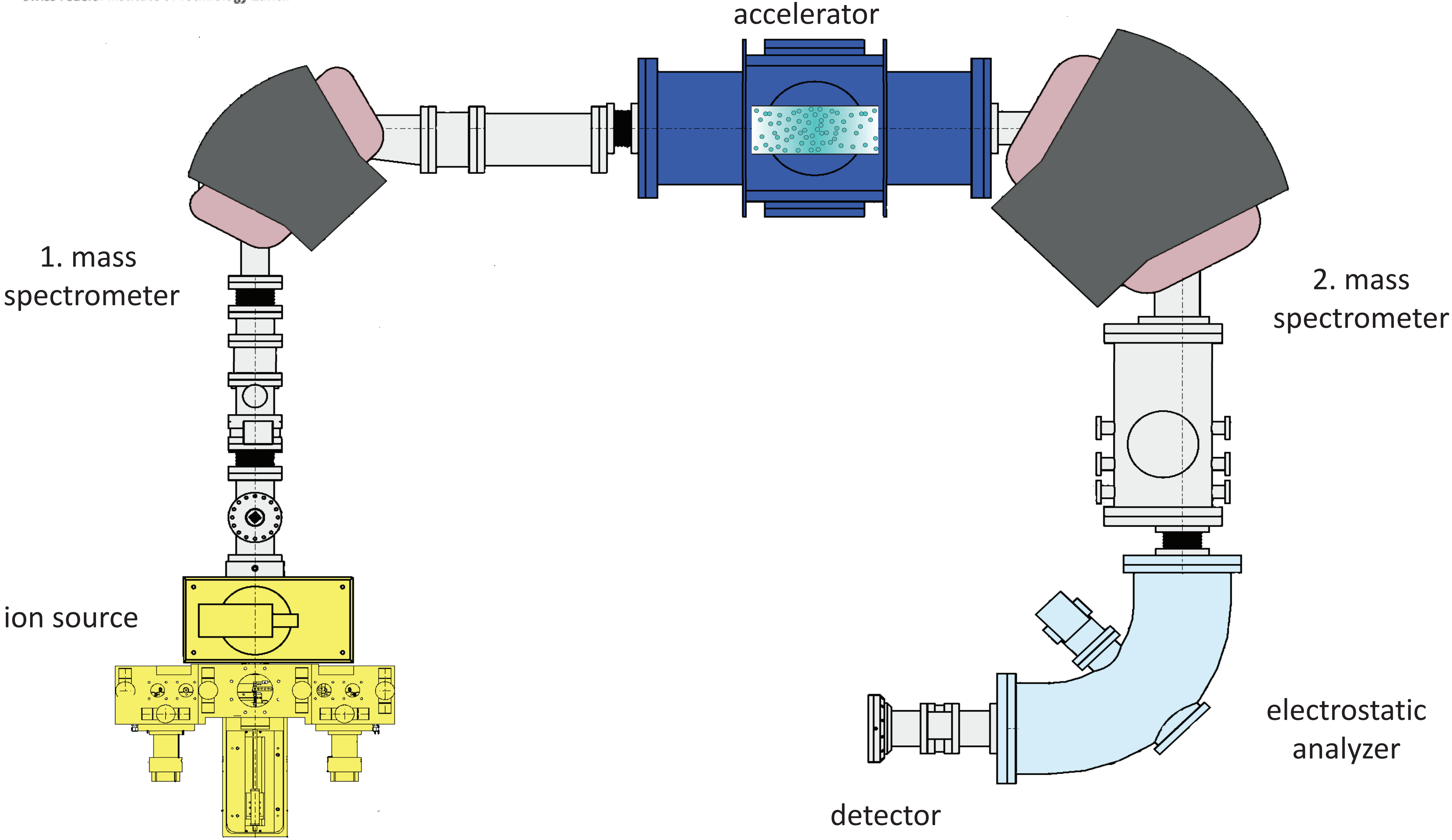
Key features for radionuclide detection

"golden AMS rules"

- Isobar separation
  - Negative ion formation
    - $^{14}C(^{14}N)$; $^{26}Al(^{26}Mg)$; $^{10}Be(^{10}B)$; $^{36}Cl(^{36}S)$; $^{41}Ca(^{41}K)$;
    - $^{10}BeF/BeBaF(^{10}BF)$; $^{41}CaH_3(^{41}KH_3)$; $^{41}CaF_3(^{41}KF_3)$;
  - Ion detection
- Abundance sensitivity
  - Suppression of neighboring isotopes (1:10^{15})
  - Multi-step mass filtering process
- Reproducible isotope ratio measurements
  - High ion optical transmission
  - reliable normalization procedure
- Eliminate mass interferences
  - Molecule destruction

  - Charge state >3^+ molecule dissociation by coulomb force
  - Charge state 1^+ in multiple ion gas collisions

How does AMS work?



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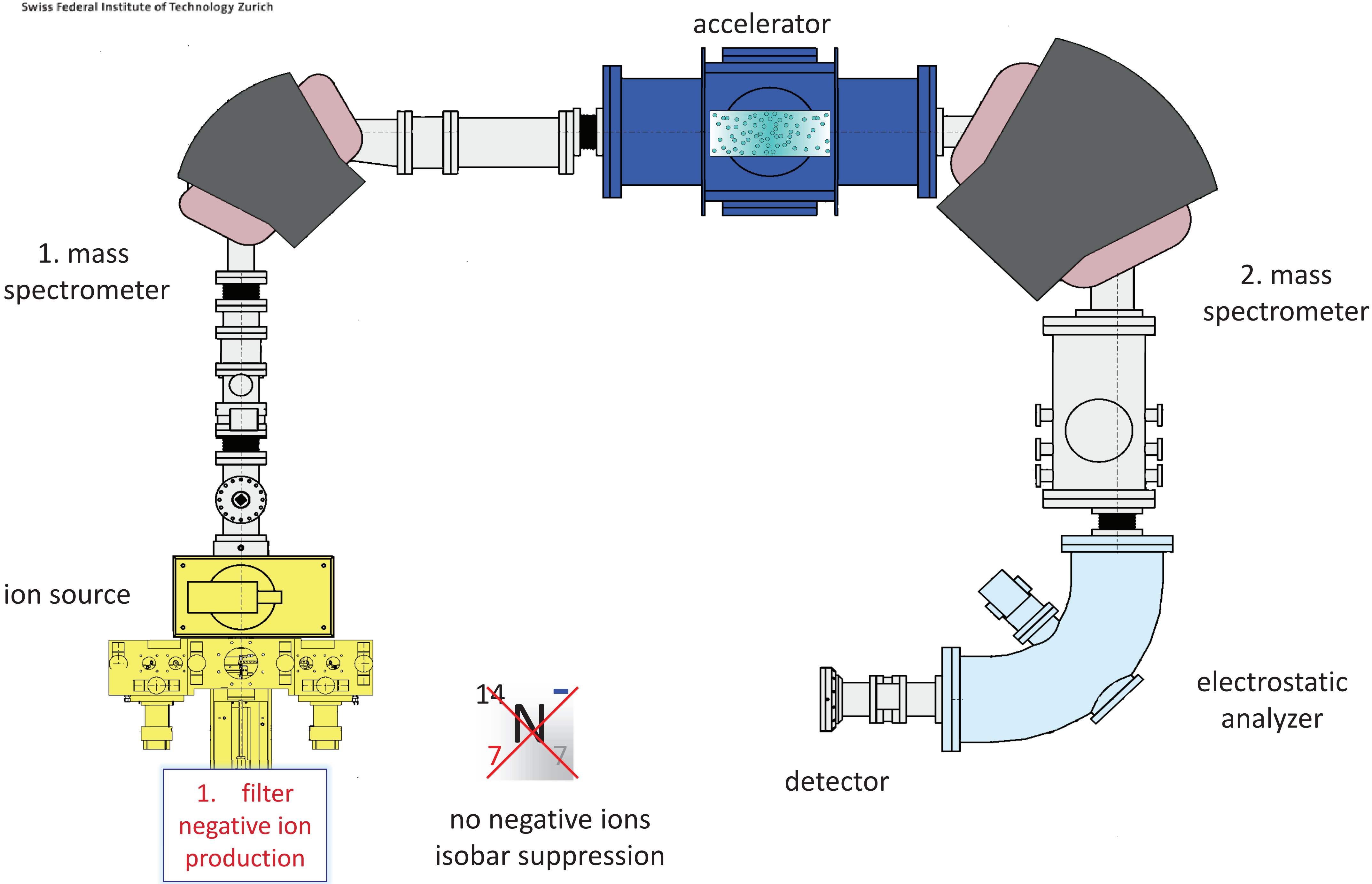
How does AMS work?



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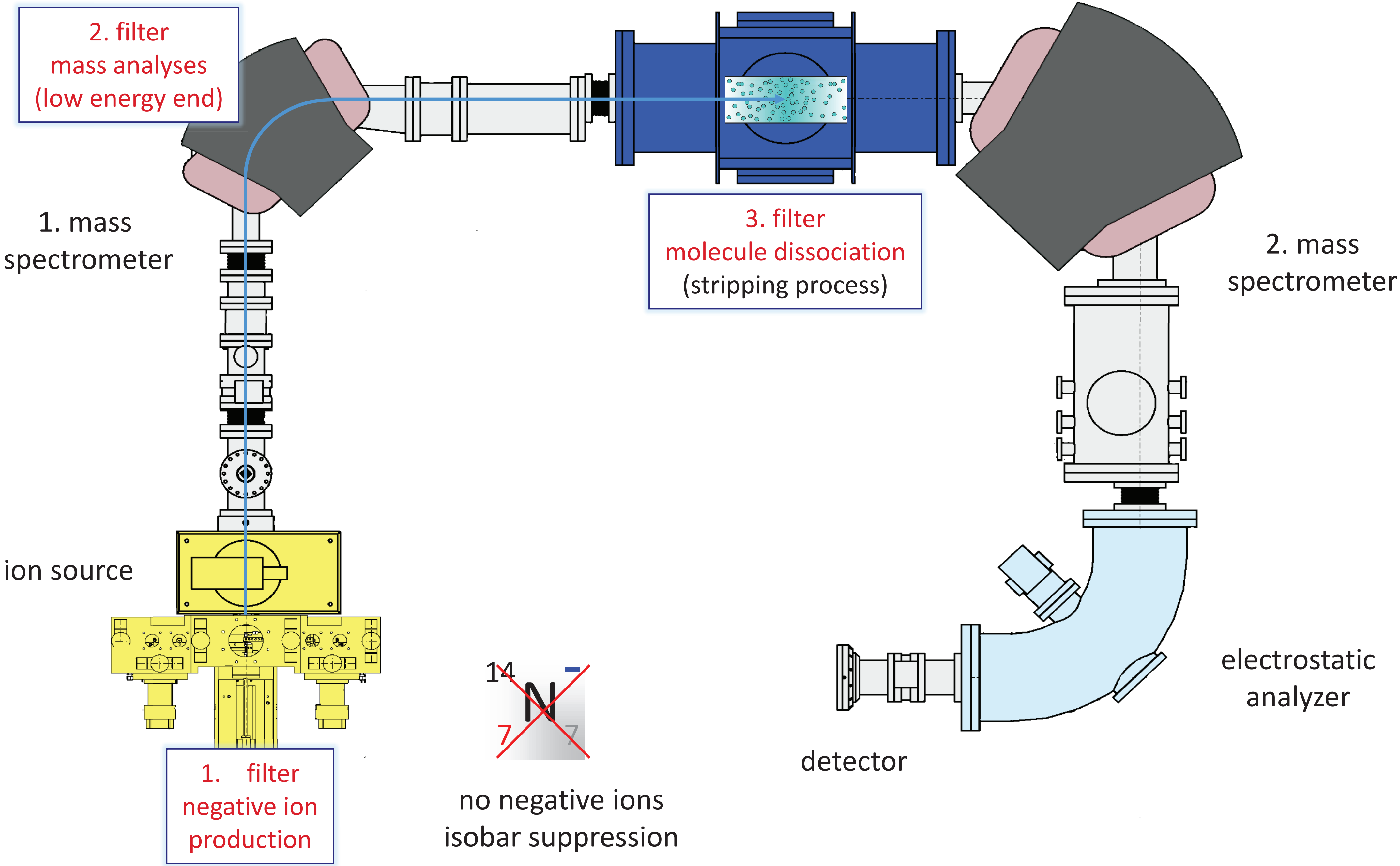
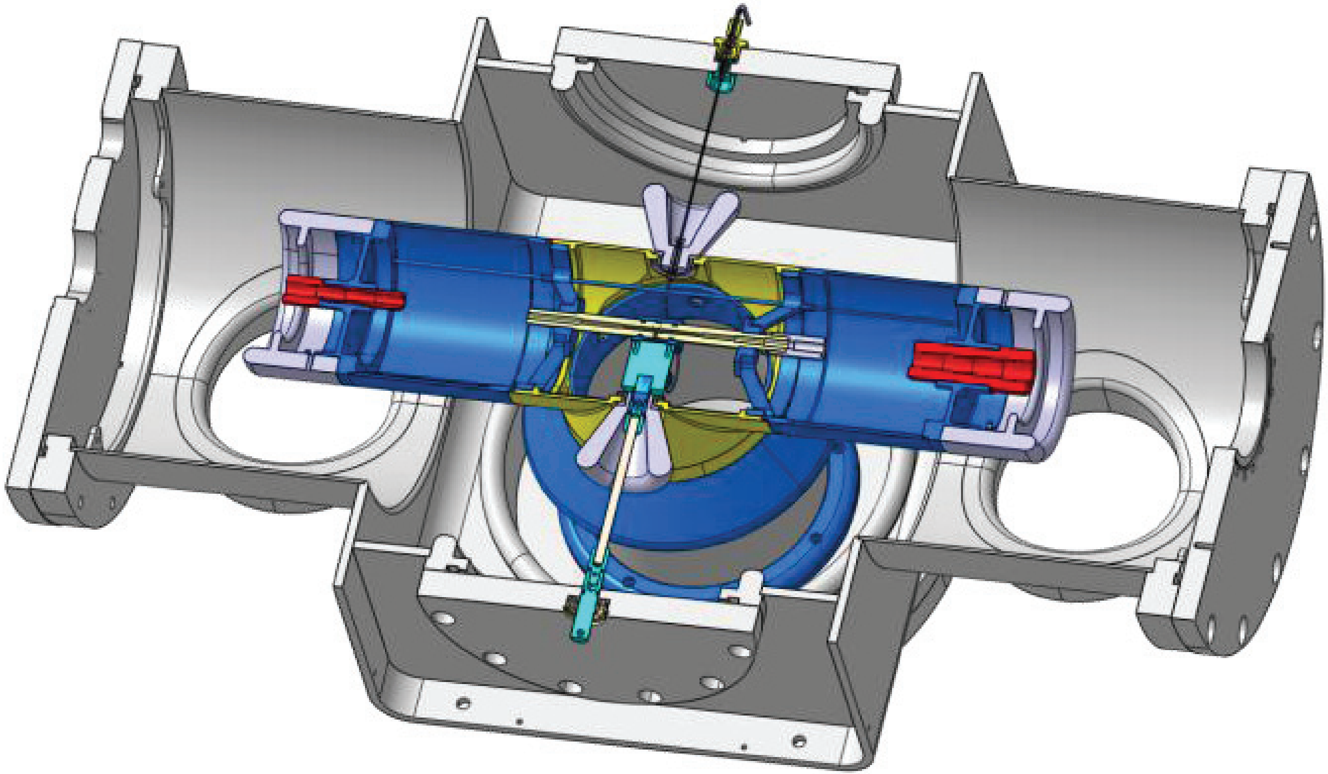


Molecule Destruction

atomic and
molecular ions:
^{14}C, ^{13}CH, $^{12}CH_2$

mass 14 amu



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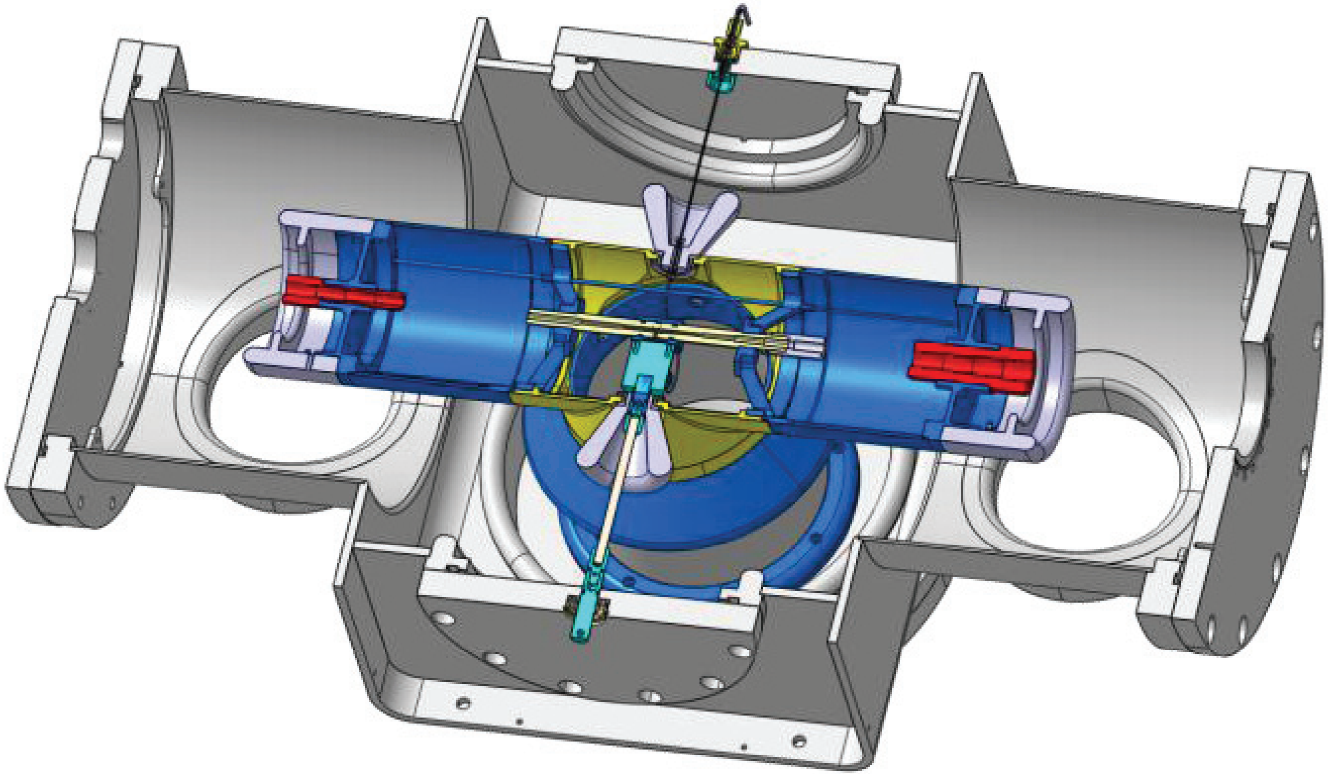
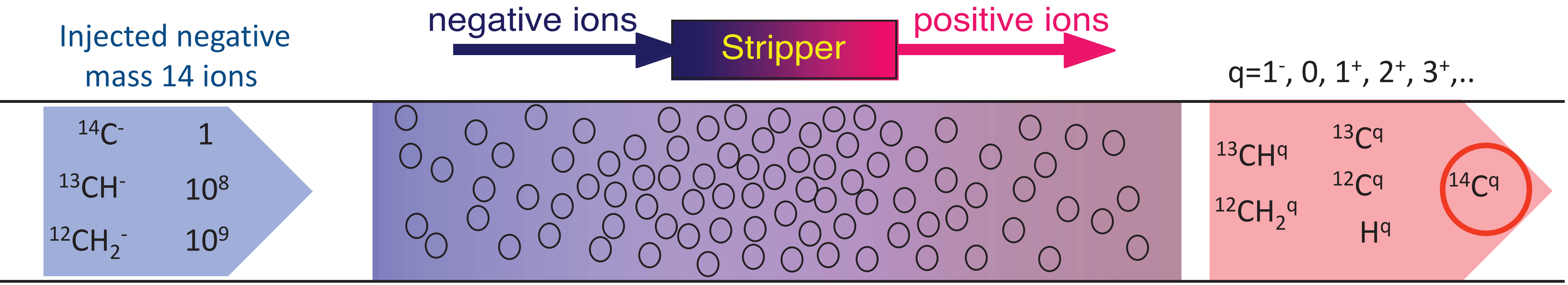


only atomic ions
$^{14}C, ^{13}C, ^{12}C$, H

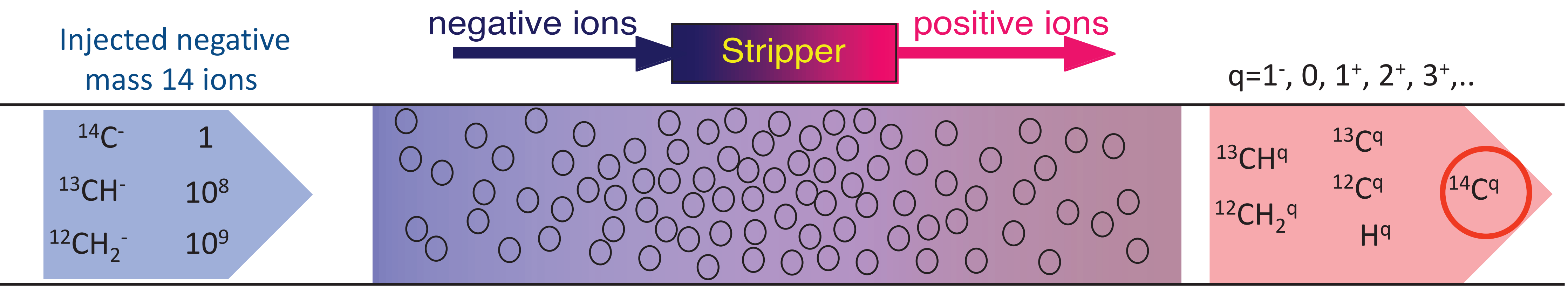
mass: 14 amu
13 amu
12 amu
1 amu

Stripping Process



- Electron-loss
- Electron capture
- Break-up of molecules
- Energy straggling
- Angular straggling

Stripping Process



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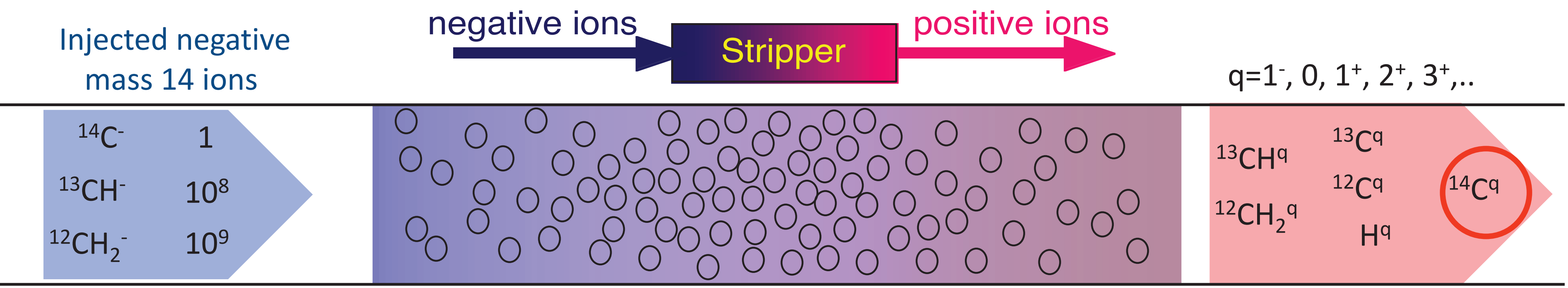

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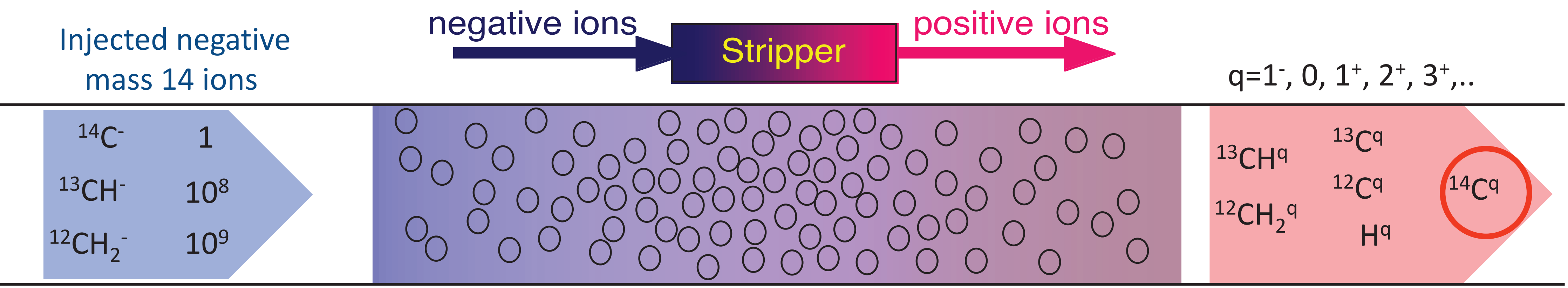

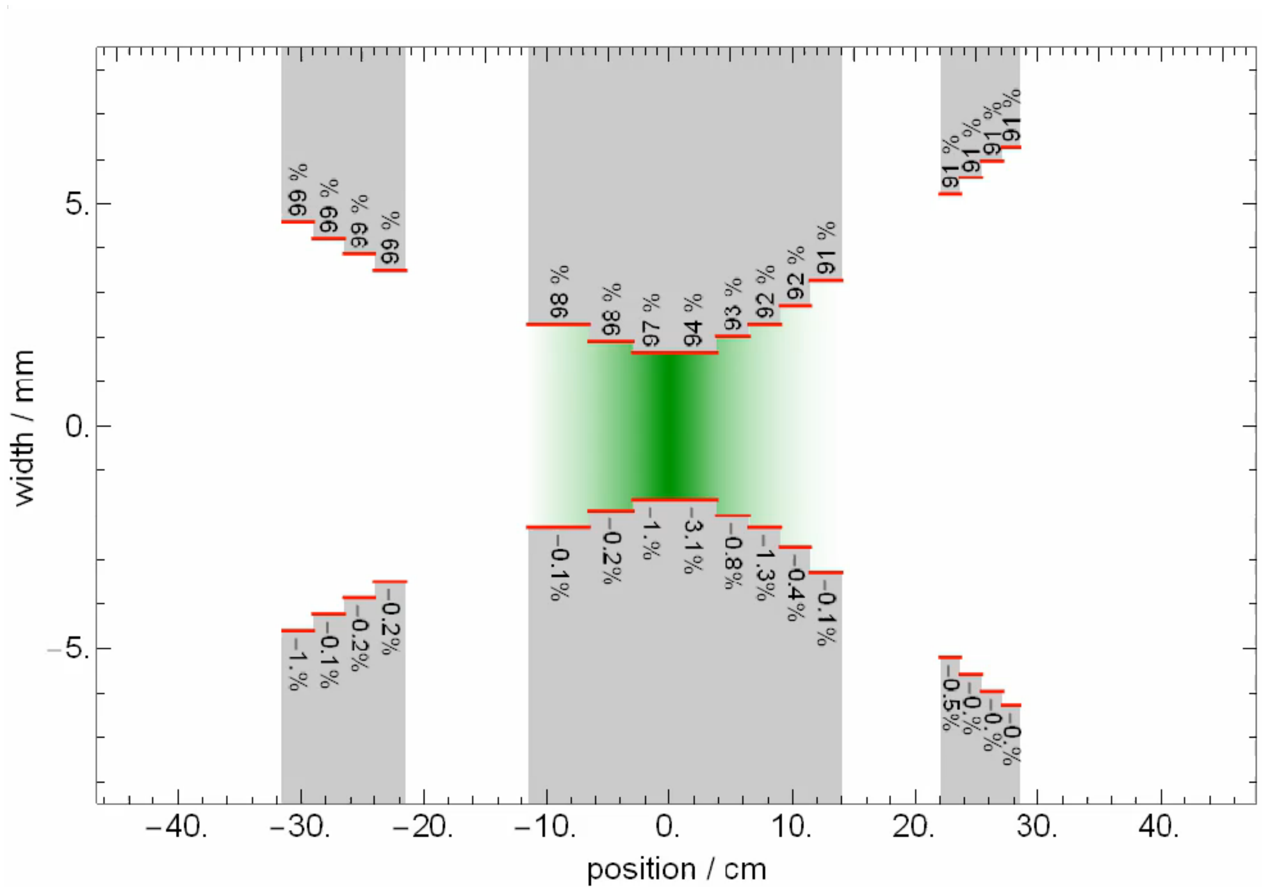

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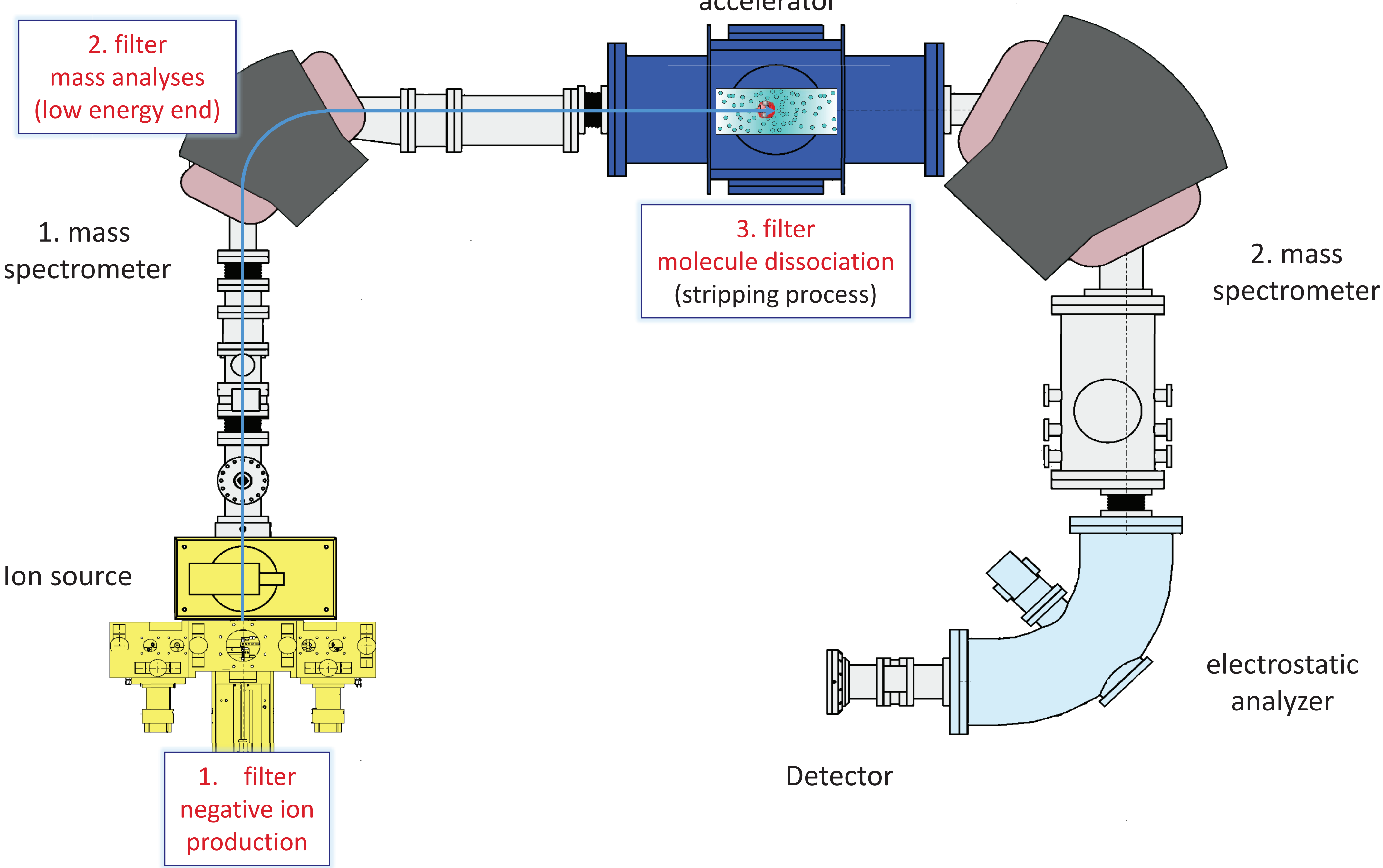

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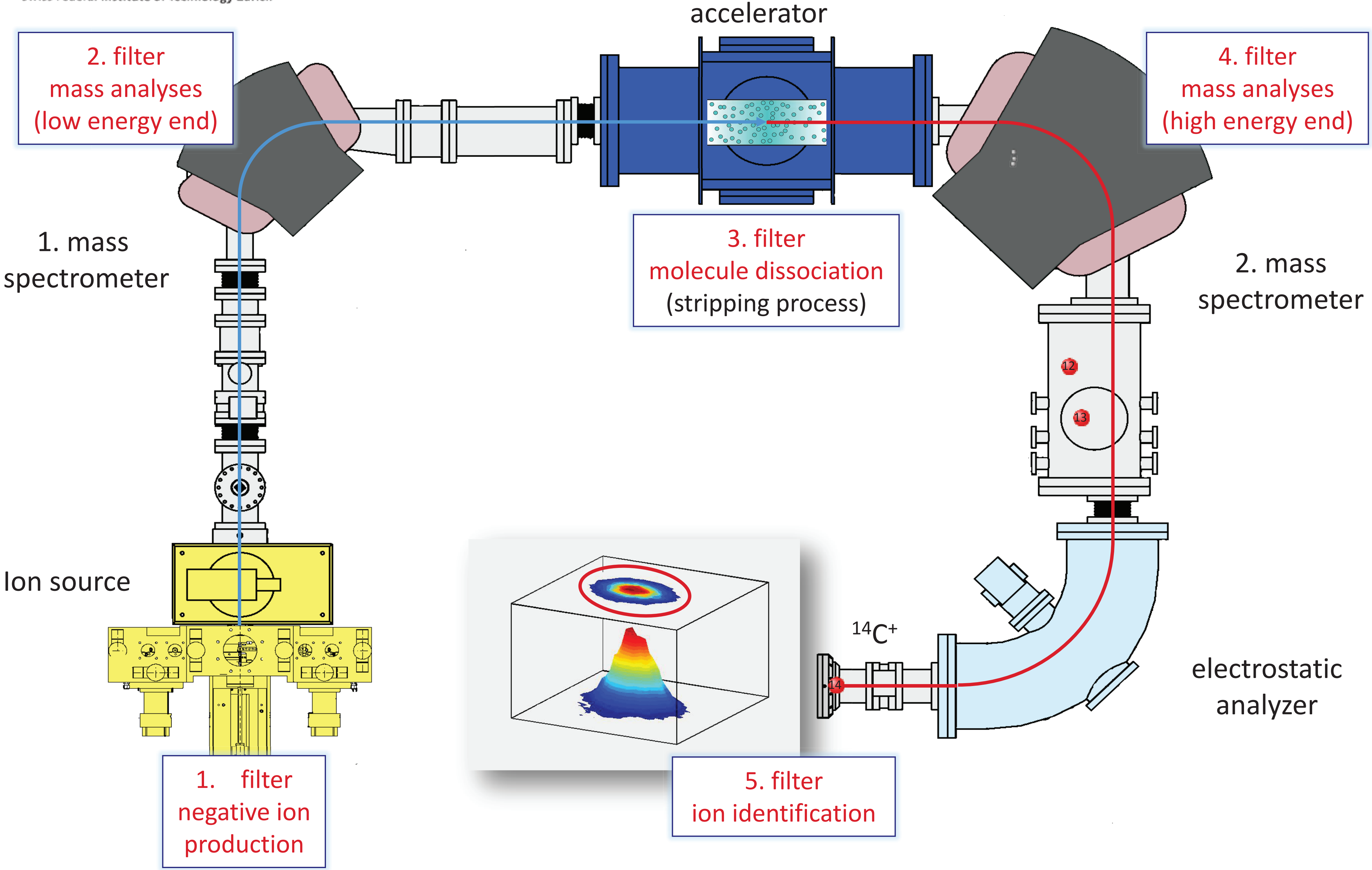

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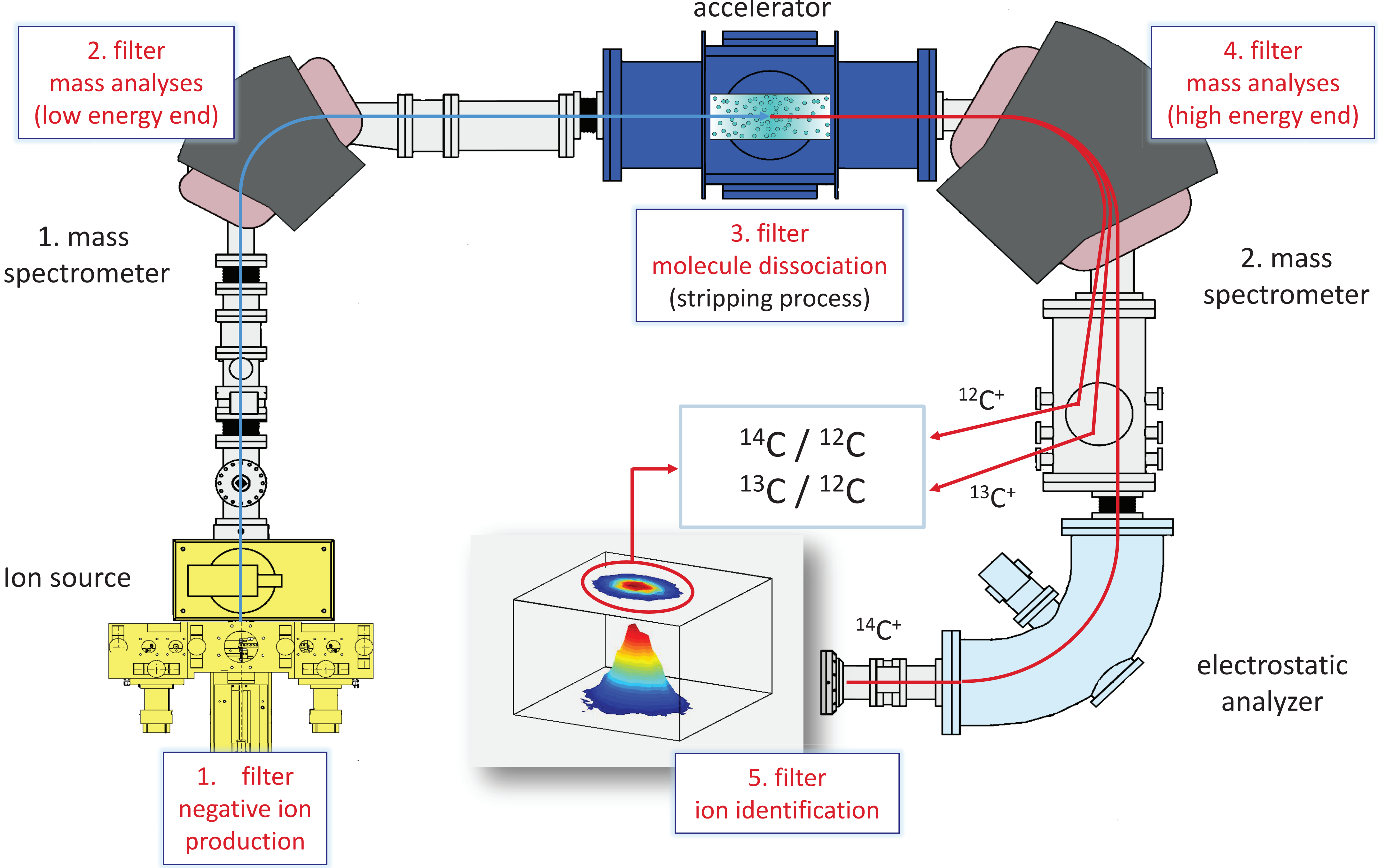

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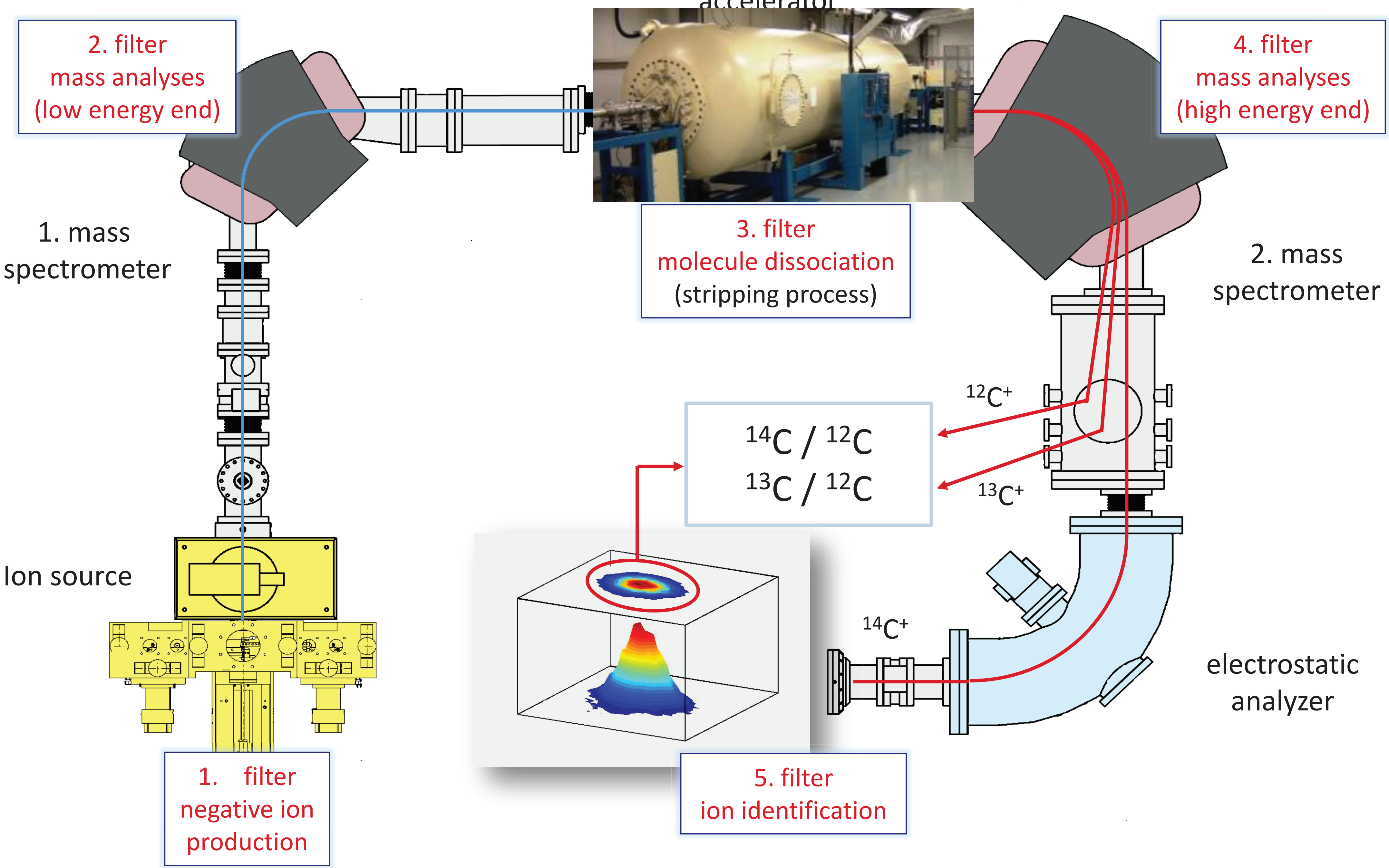
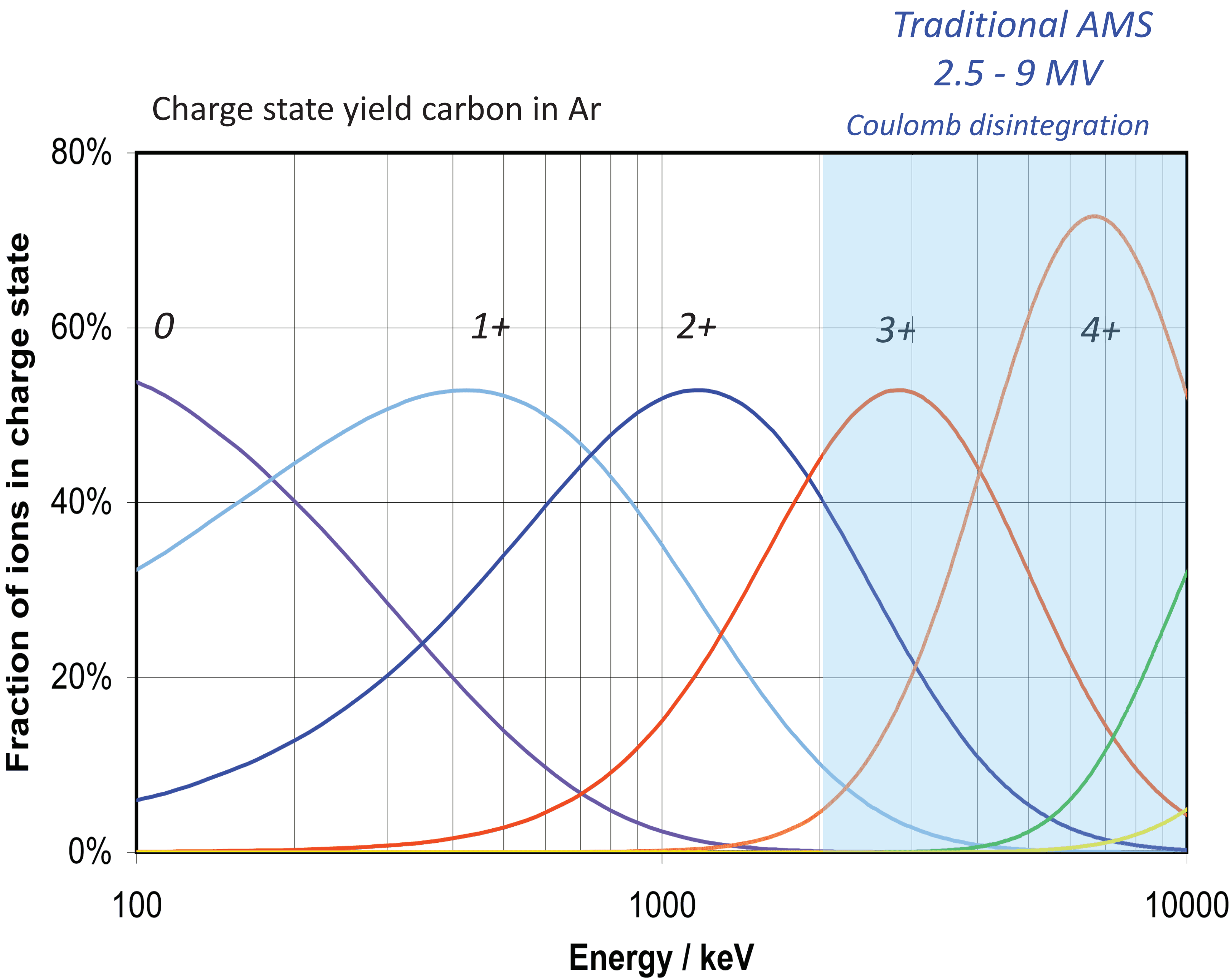


Charge state yield & accelerator size



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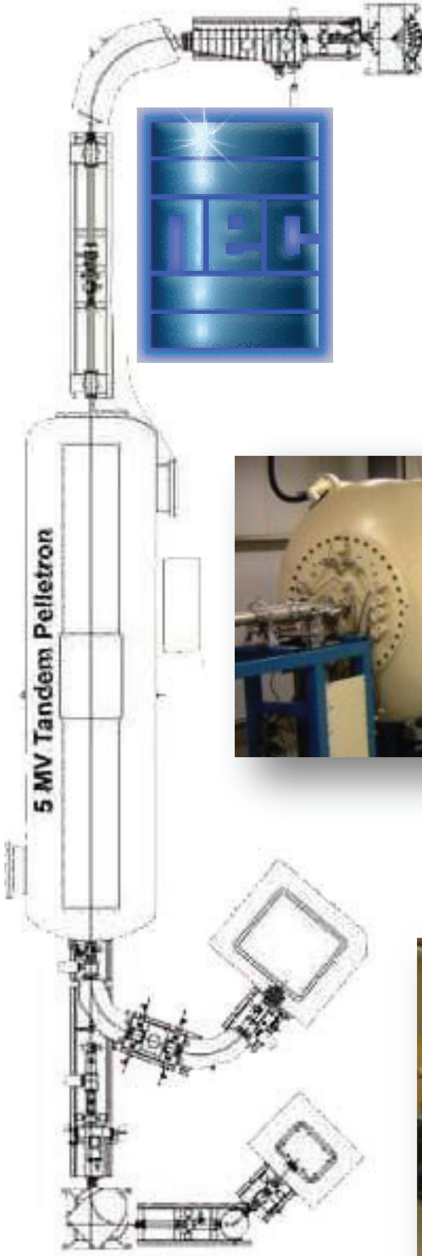
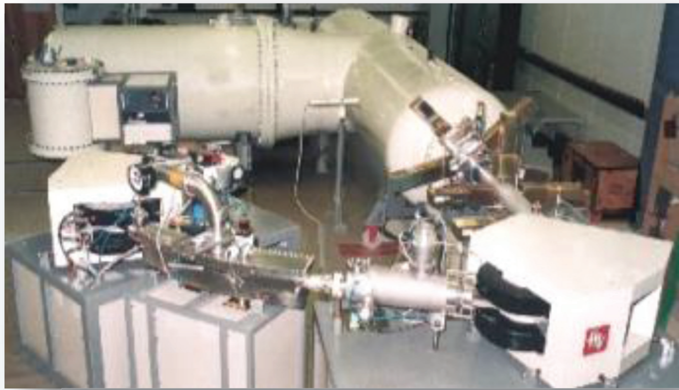
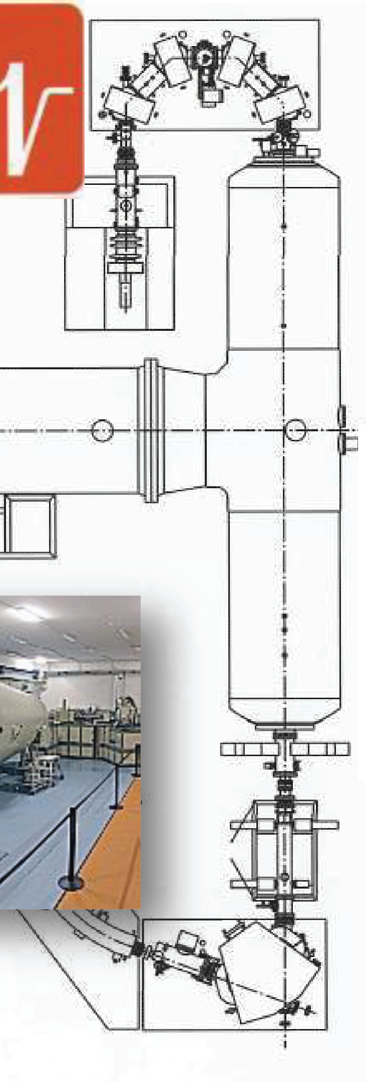
Traditional 3-6 MV AMS systems

20 -25 m



Leibniz AMS 3 MV facility, Kiel, GER





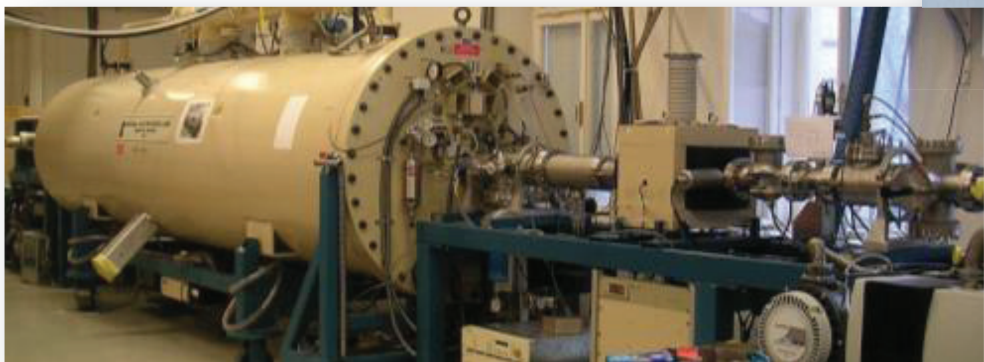
≈ 10 -15 m

Glasgow, UK

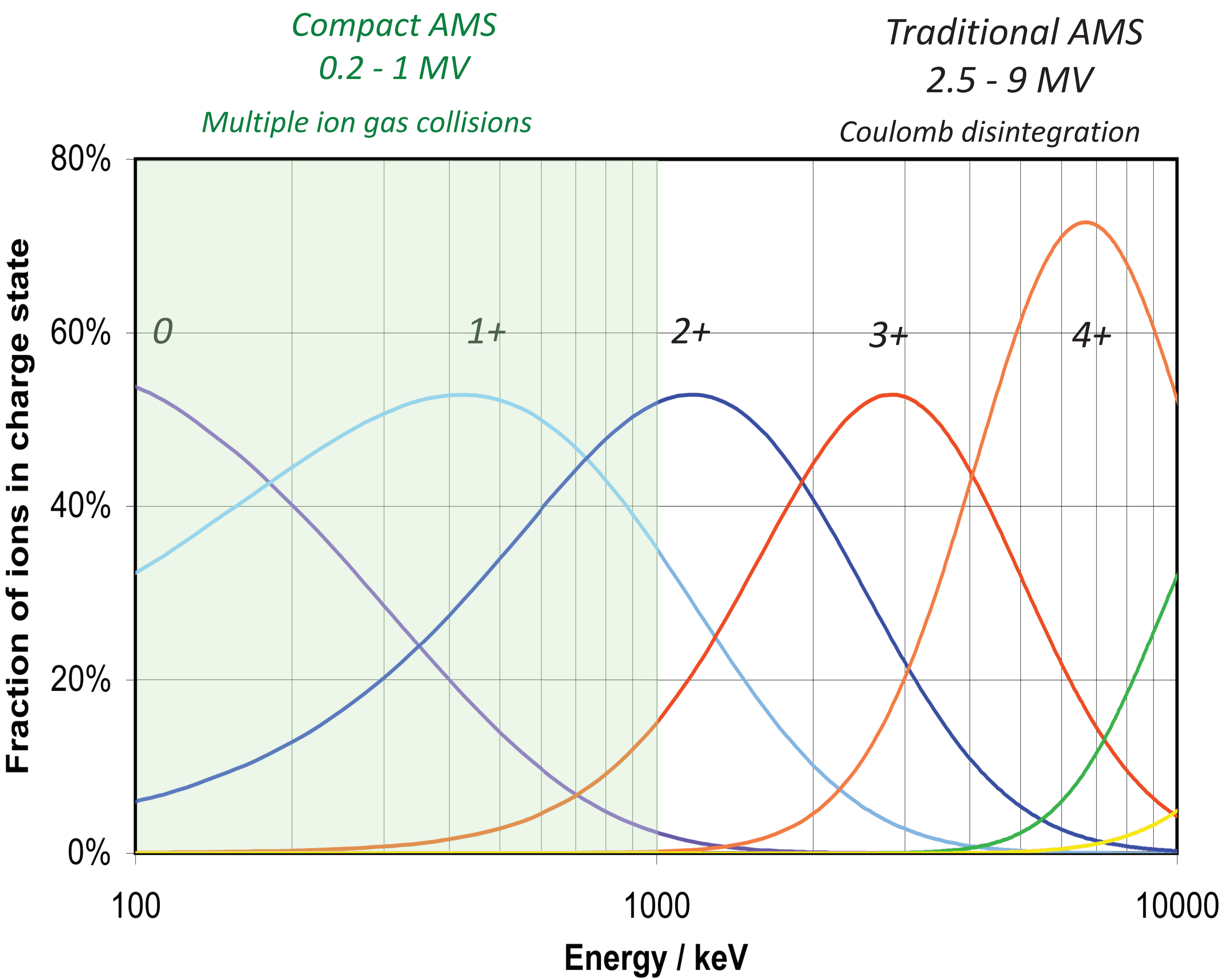
Sydney, AUS

Rossendorf, GER

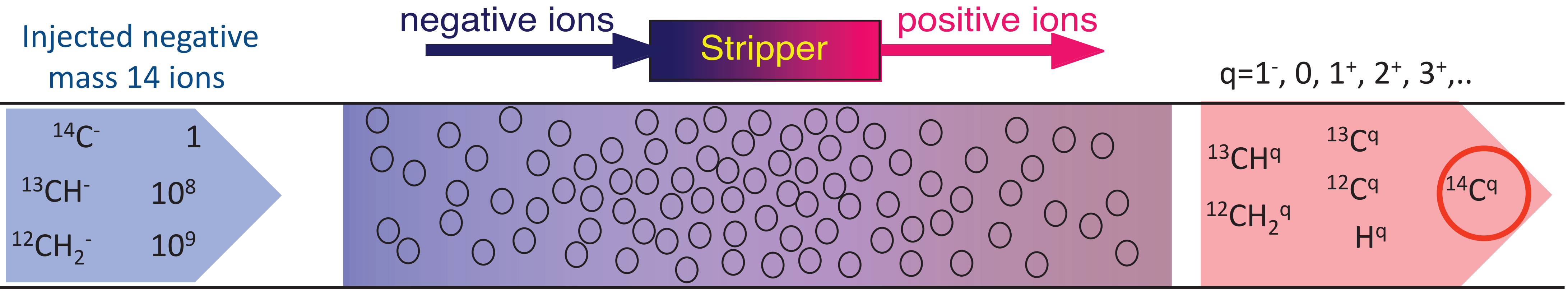
VERA AMS 3 MV facility, Vienna, Austria



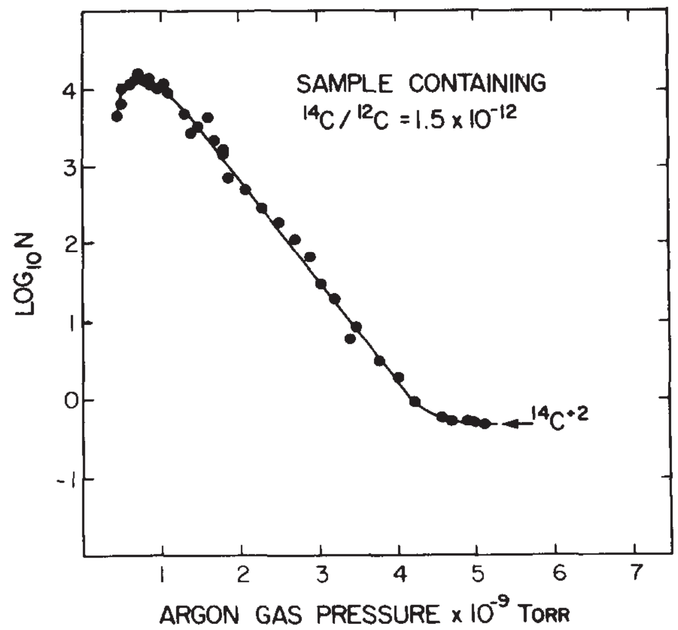
^{14}C charge state yield & accelerator size



Stripping Process



- Electron-loss
- Electron capture
- Break-up of molecules
- Energy straggling
- Angular straggling

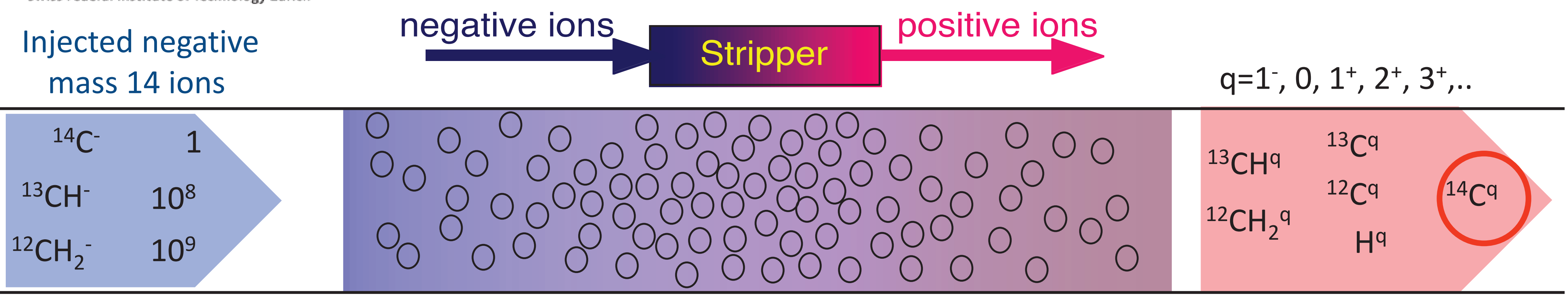


THE $^{12}CH_2^{2+}$ MOLECULE AND RADIOCARBON DATING BY ACCELERATOR MASS SPECTROMETRY

H.W. LEE, A. GALINDO-URIBARRI *, K.H. CHANG, L.R. KILIUS and A.E. LITHERLAND

ISOTRACE Laboratory, University of Toronto, Toronto, Ontario M5S 1A7, Canada

Stripping Process



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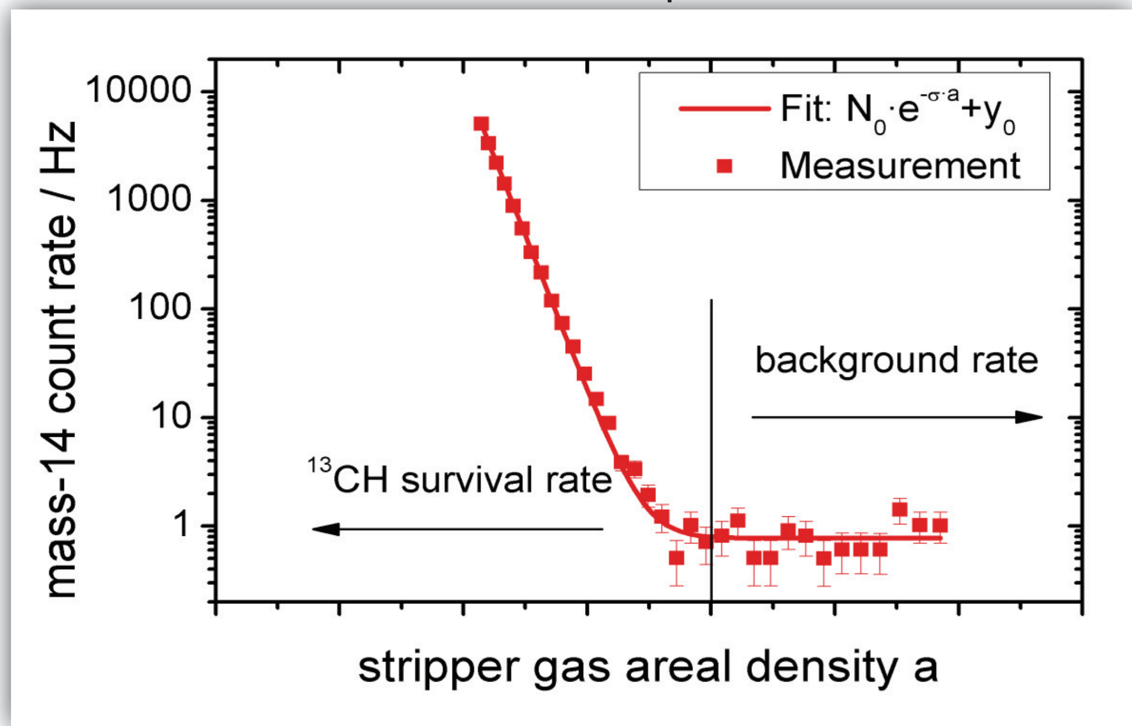
Destruction of molecular ions in q=1⁺

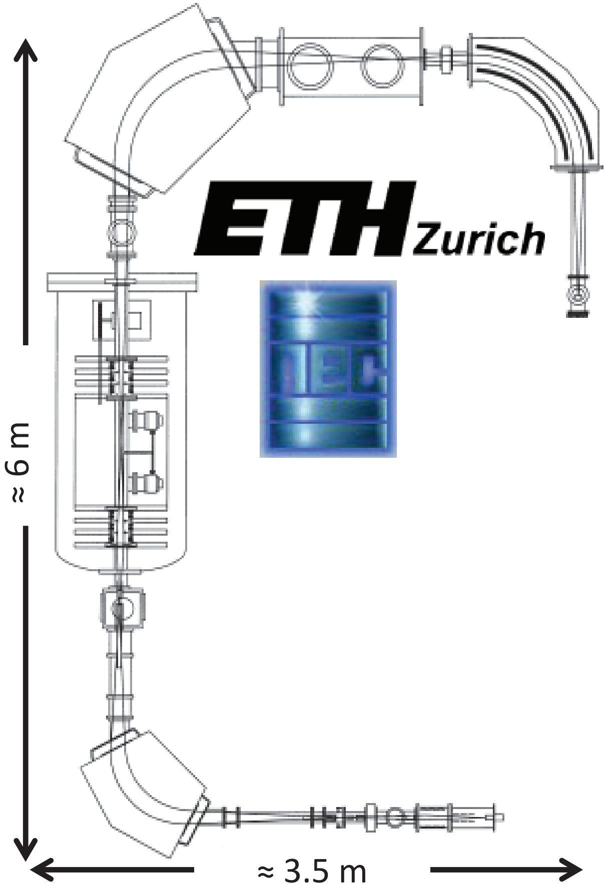


The Generation of Compact AMS Systems

The first compact AMS system (1998) using charge state 1^+

Commercial systems are on the market from NEC and HVEE

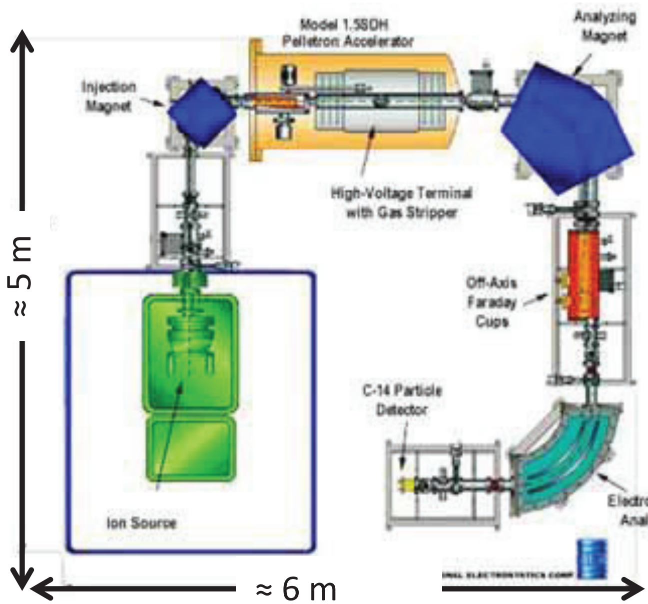
Compact AMS Systems (1 MV- 500 kV)

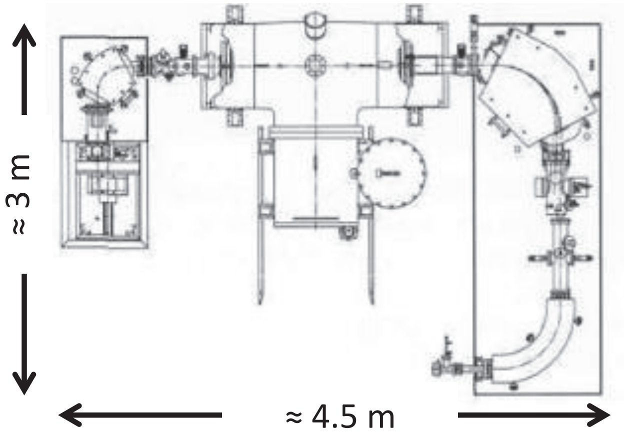


KECK AMS facility, Irvine, USA

AMS facility, Seville, Spain

Tandy AMS facility, Zurich, CH





1 MV Tandetron accelerator

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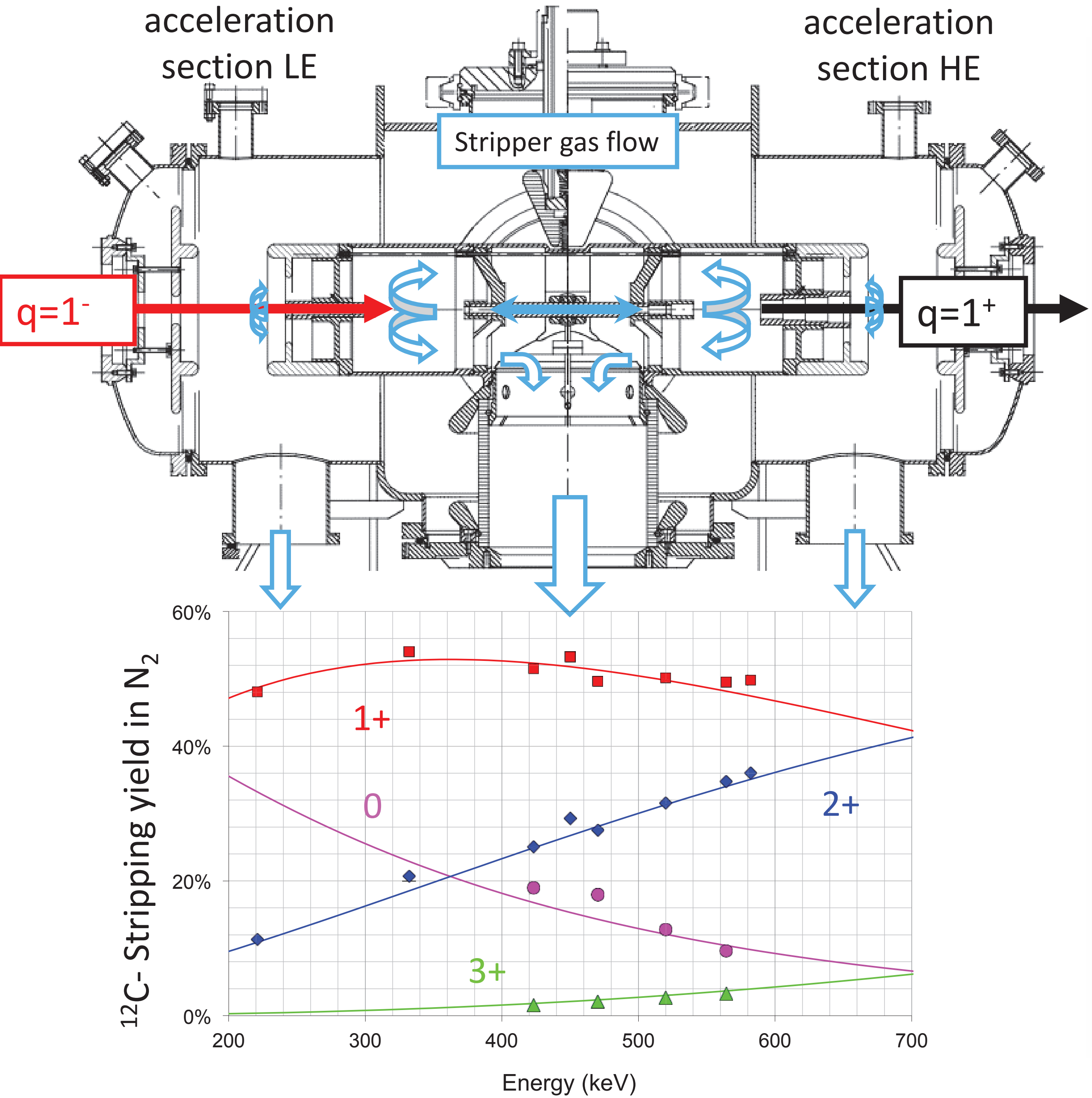
Replacing the traditional Accelerator (MICADAS system)



He-stripping to enhance performance: AixMICADAS (2014)

in collaboration with: Aix Marseille université

Vendredi 11 juillet 2014

performance:

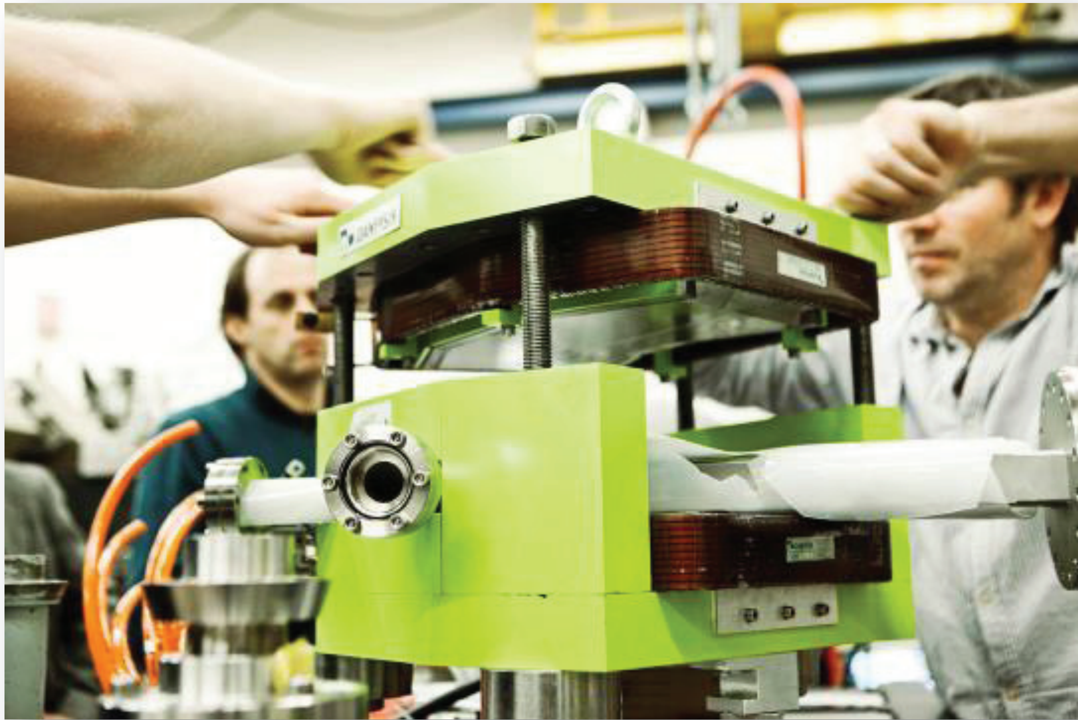
- 48 % transmission
- > 90% ion optical transmission
- stable operation conditions

DANFYSIK

Arnd Braurichter
Franz Boedker
Leif Baandrup

UPPSALA UNIVERSITET

Göran Possnert
Mehran Salehpour



Introducing permanent magnets in AMS instruments

- Simplified installation
- No cooling water required
- significant reduction in operating costs

Why shall we get away from a well established concept?

energy consumption of magnets:
10 kW @ 3000 h/year
energy costs: ~ 0.3 $/kWh → 9000 $/yr

GreenMICADAS developed at ETHZ

Wall plug 3 x 400 V/16 A

compact lab-sized instrument (low power consumption)

- automatic operation
- low maintenance costs
- no open high voltages
- simple operation

Discoverers of Accelerator Mass Spectrometry (1977)

The New York Times

NEW YORK, THURSDAY, JUNE 9, 1977

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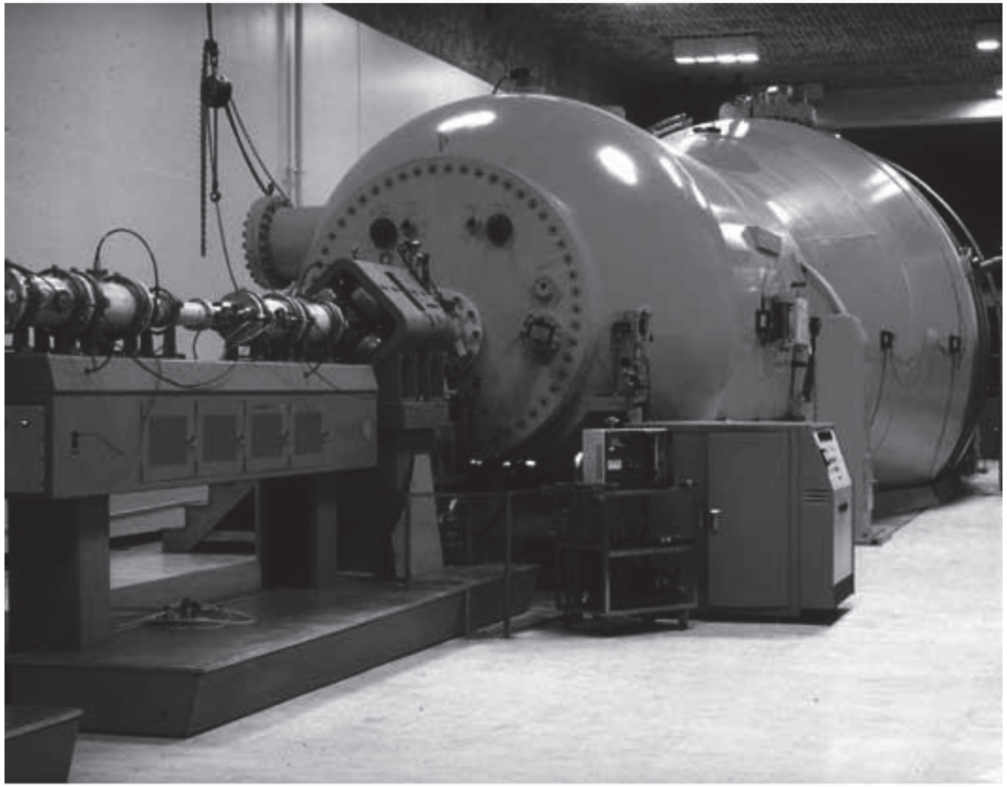
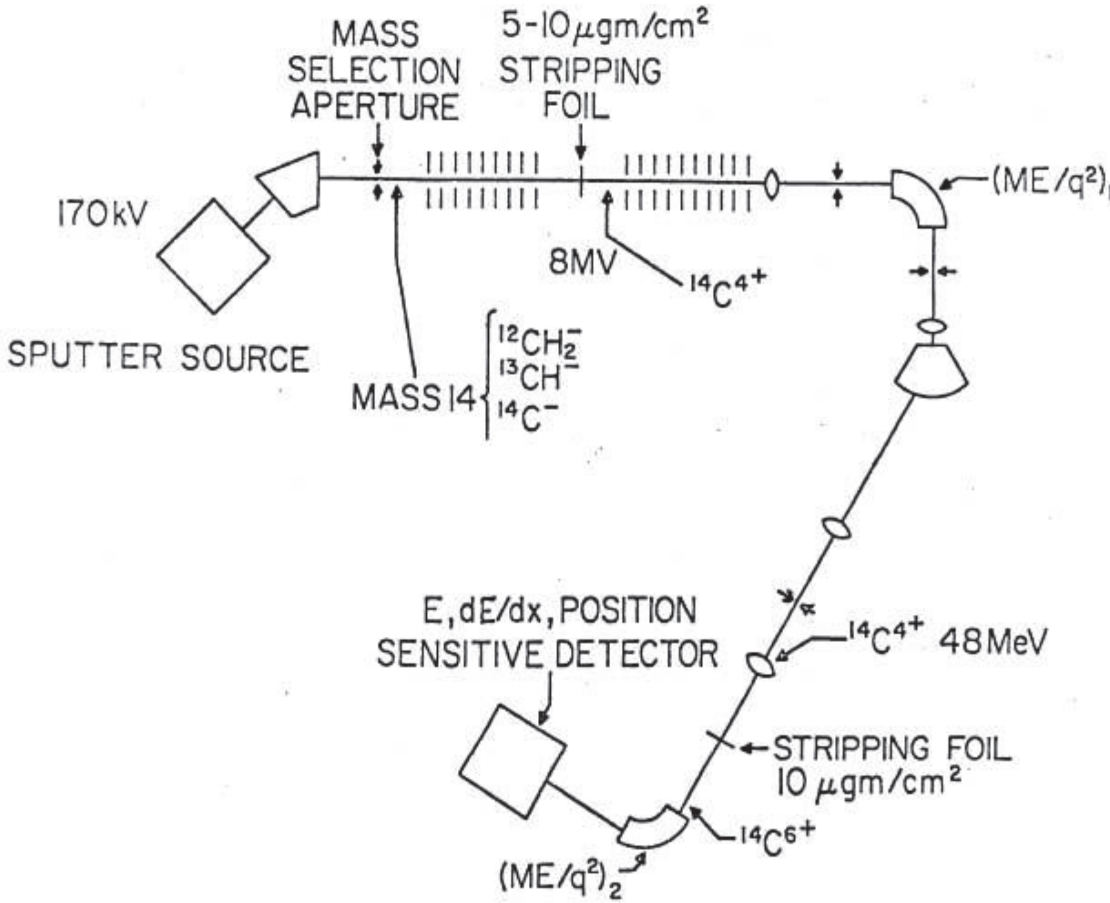


Rochester, NY(USA) MP tandem accelerator



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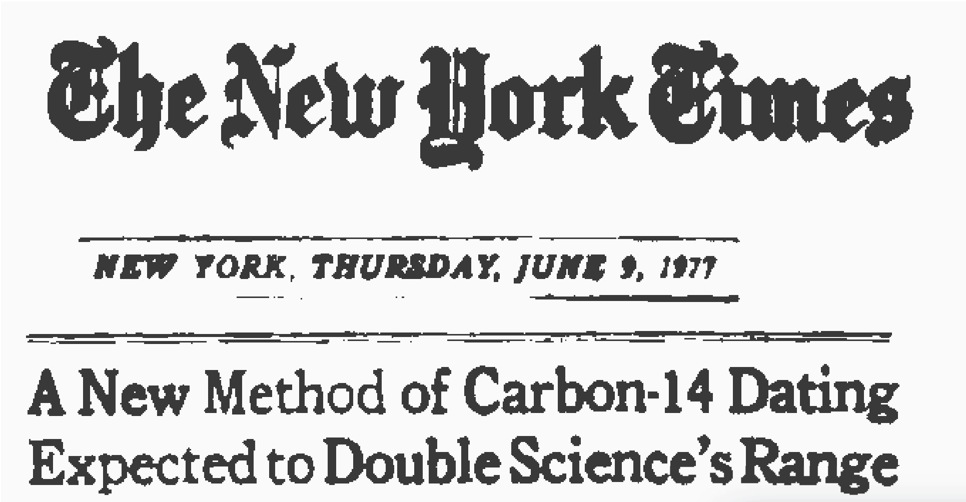
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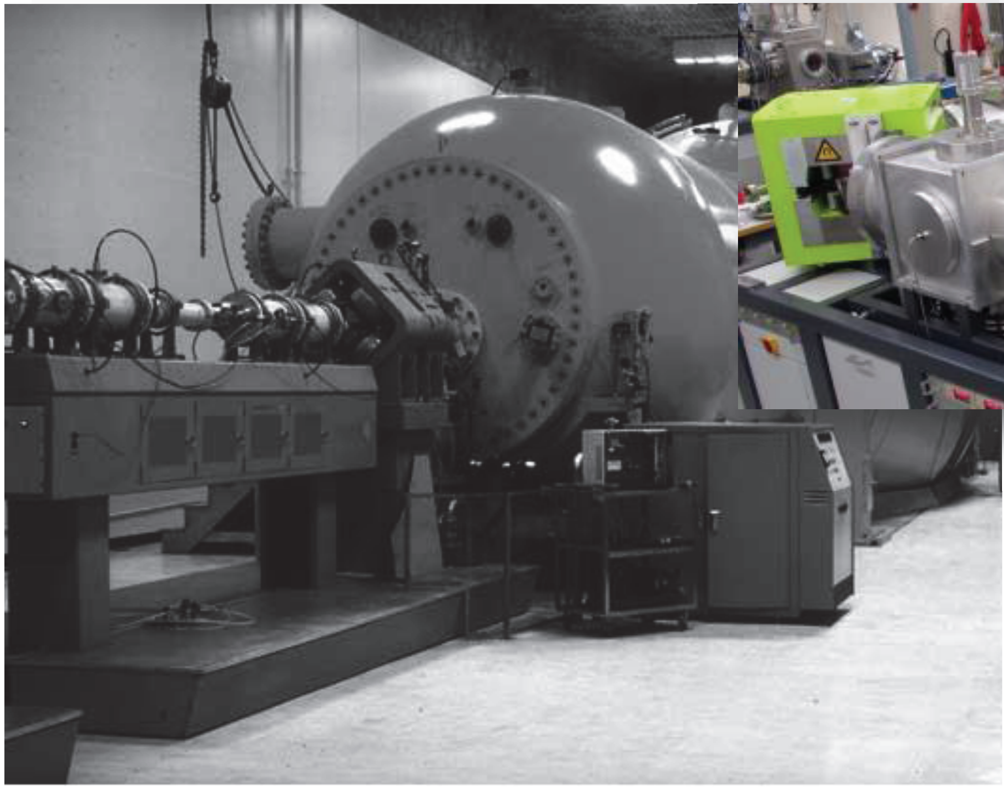
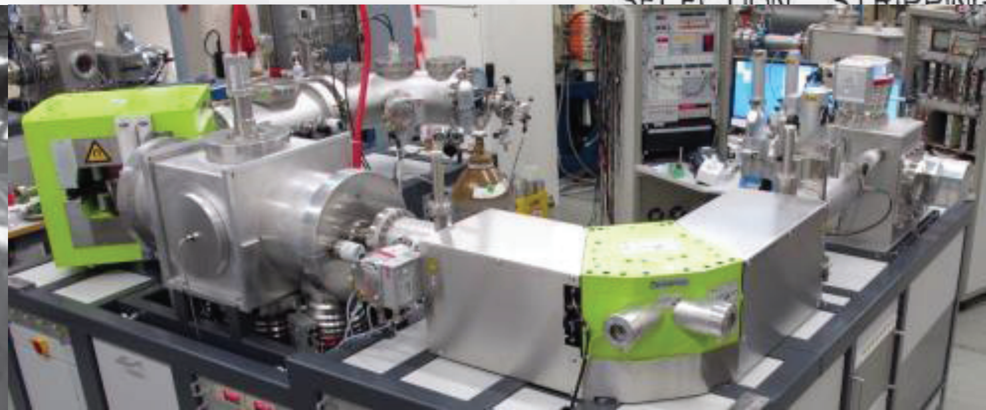
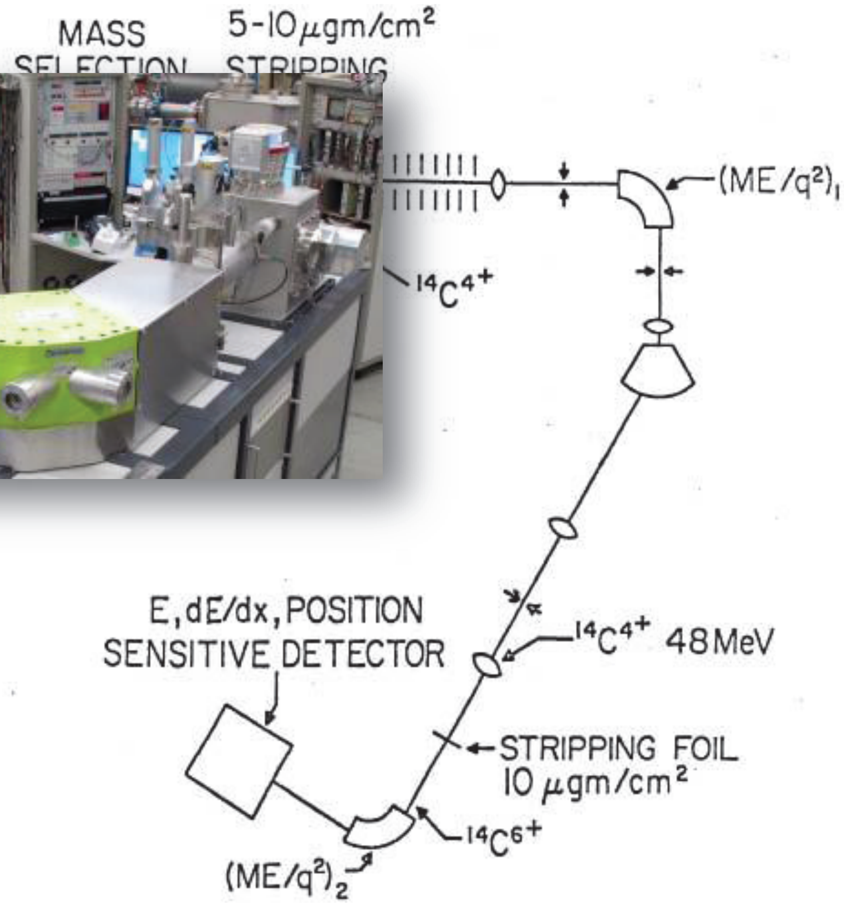
G. Raisbeck
R. Muller

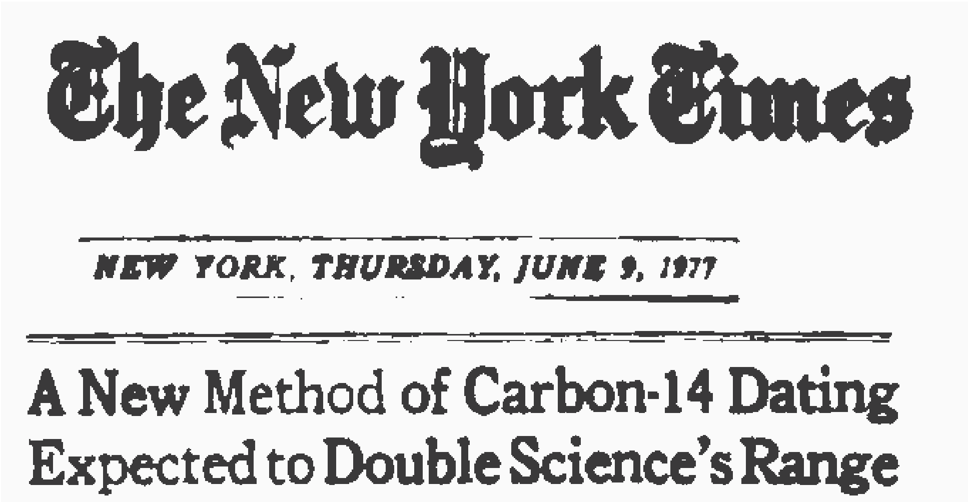






Rochester, NY(USA) MP tandem accelerator

Discoverers of Accelerator Mass Spectrometry (1977)

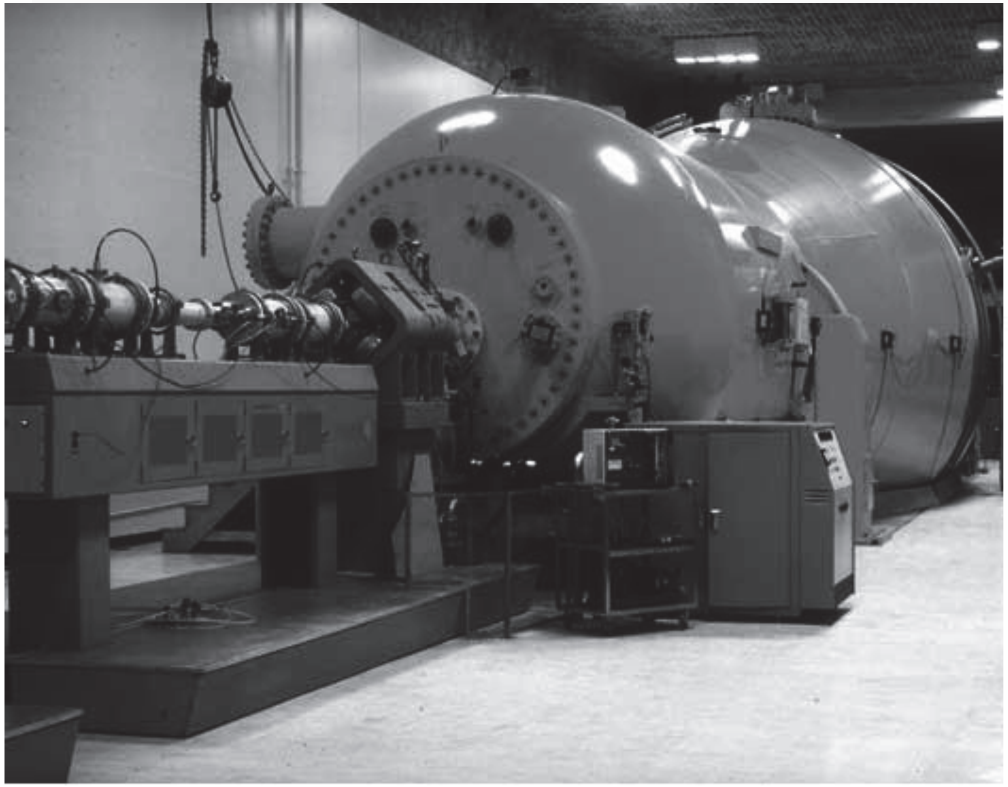

The New York Times

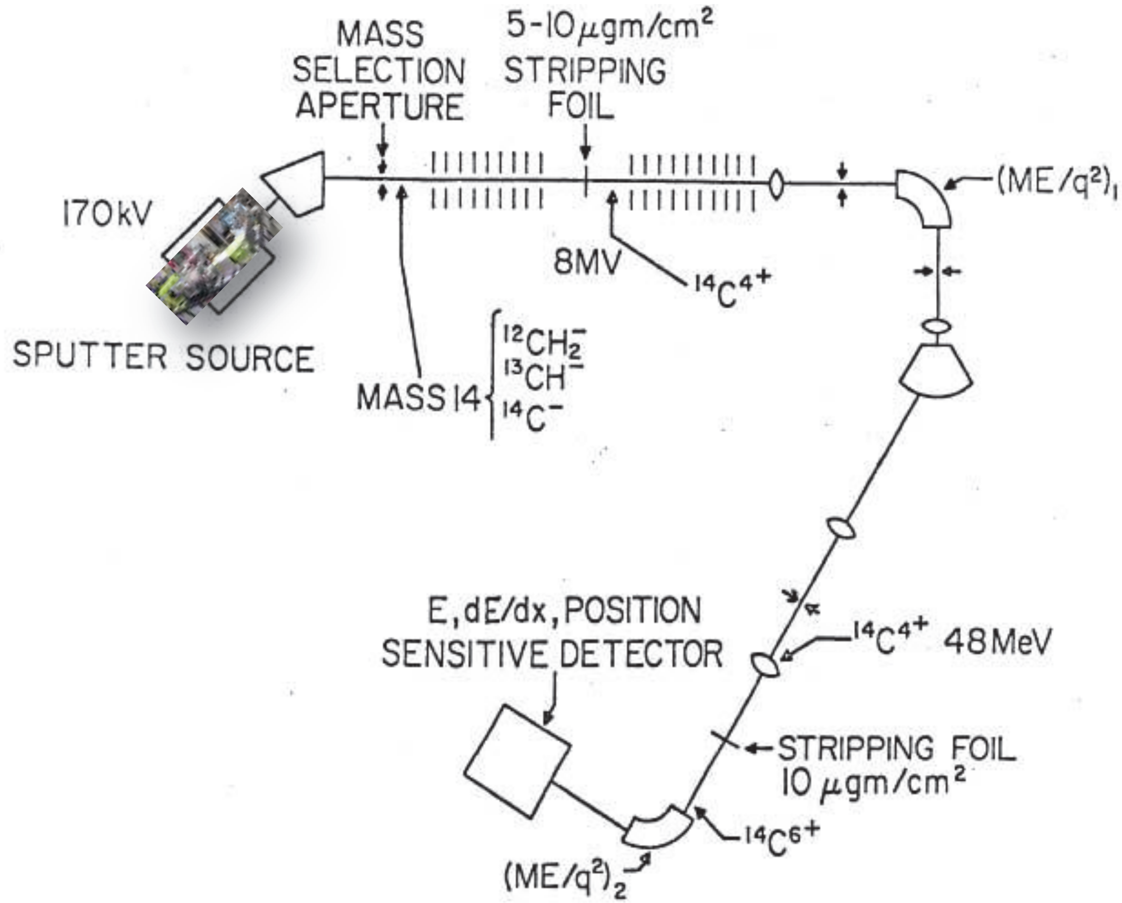
NEW YORK, THURSDAY, JUNE 9, 1977

A New Method of Carbon-14 Dating Expected to Double Science's Range

AMS-Heros
A.E. Litherland
K.H. Purser
H.E. Gove

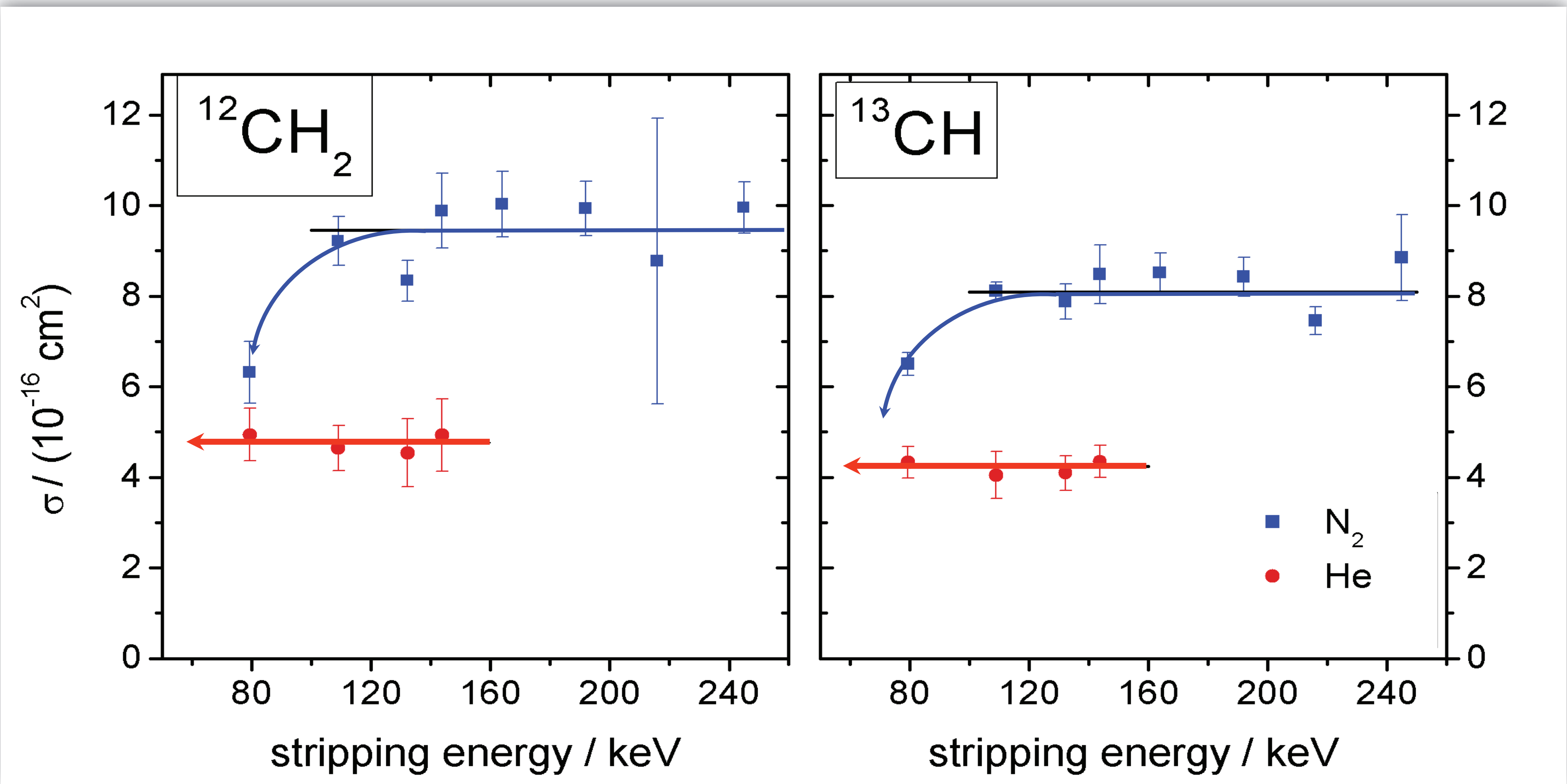
not in picture
E. Nelson
G. Raisbeck
R. Muller


Rochester, NY(USA) MP tandem accelerator



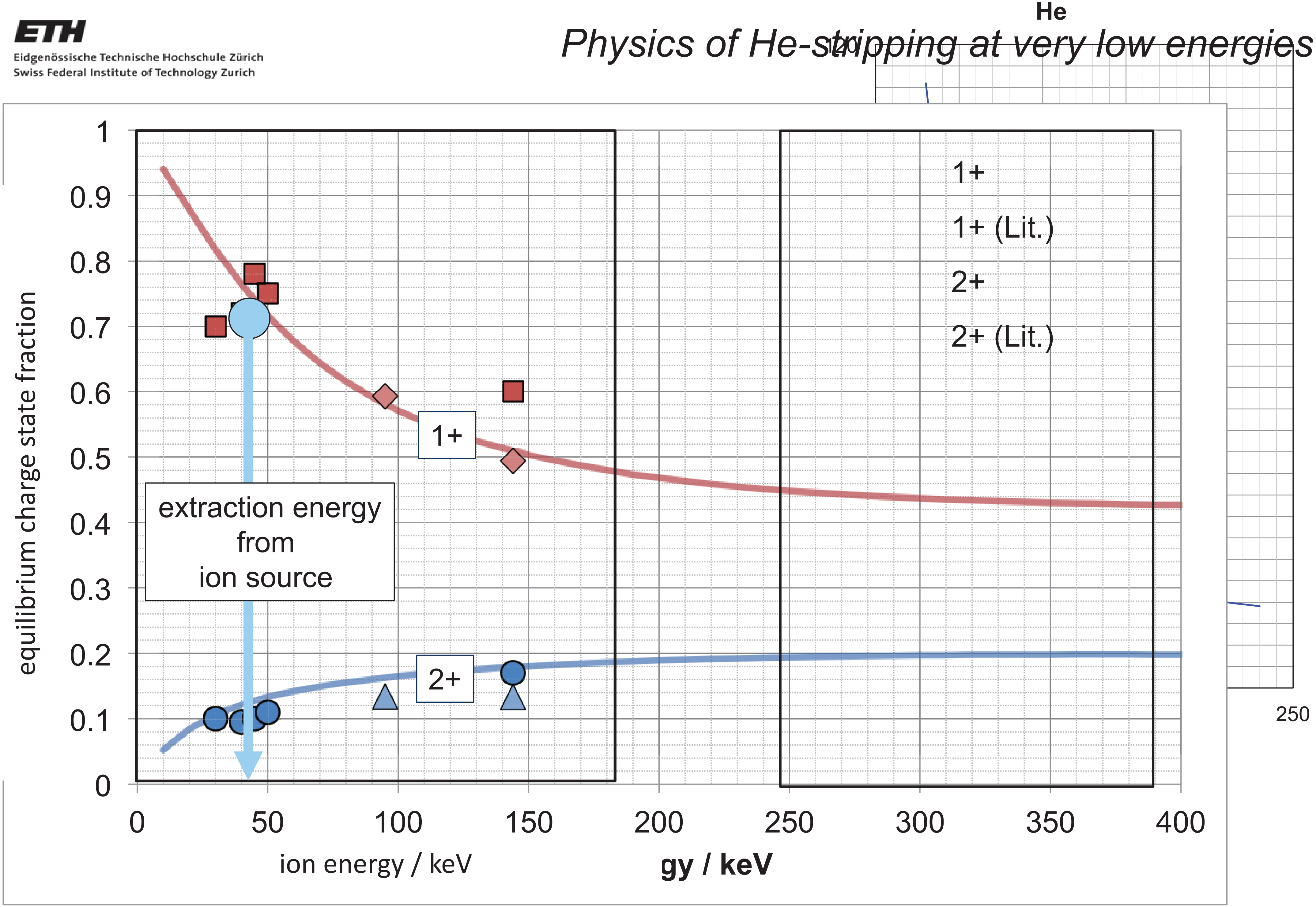
What else??

Cross section of molecule dissociation



He areal density of ≈ 0.5µg / cm^2 should be sufficient to get rid of molecules

Physics of He-stripping at very low energies

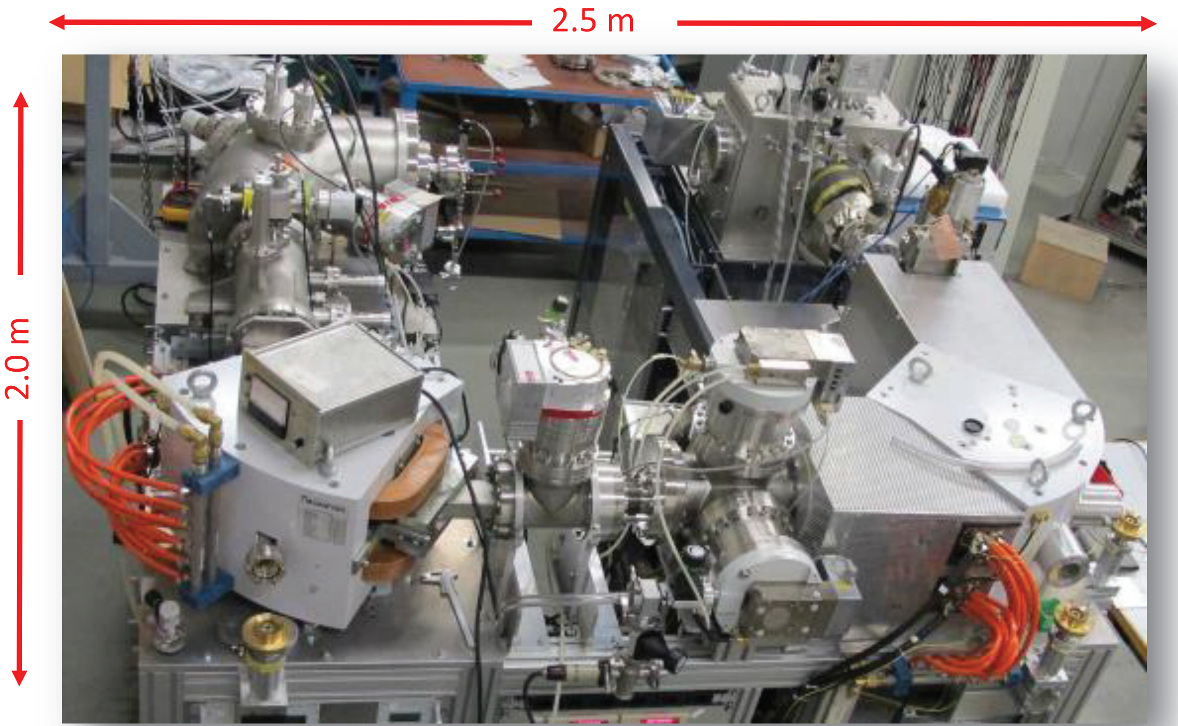


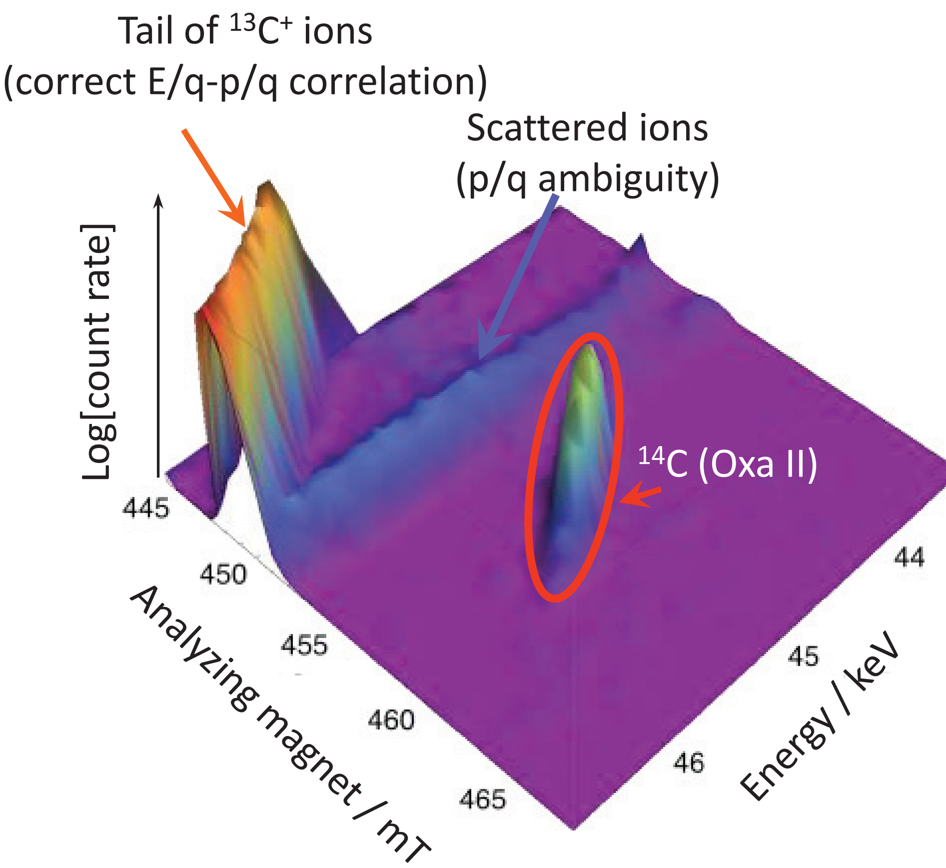
ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

A pure mass spectrometer for ^{14}C detection

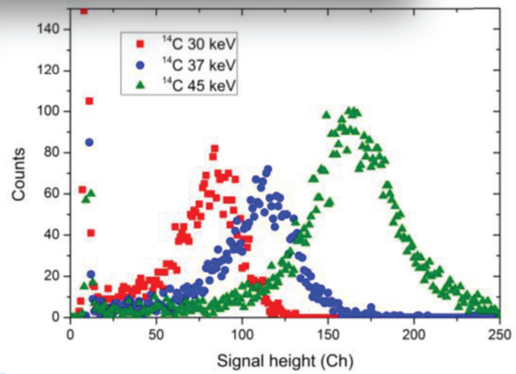
Experimental platform (prototype instrument)

- Investigate physical processes
- find best suited ion optical conditions
- test designs for a dedicated ^{14}C mass spectrometer

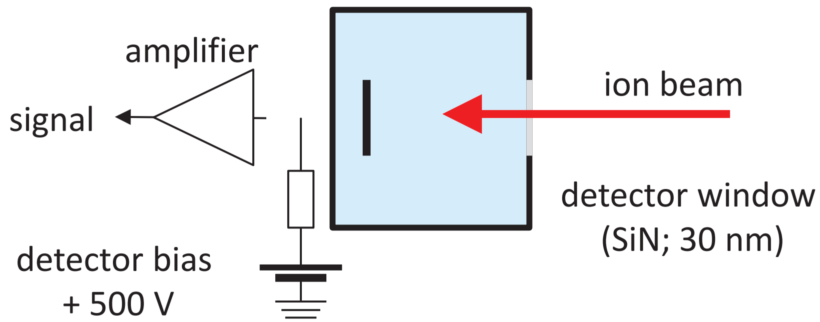




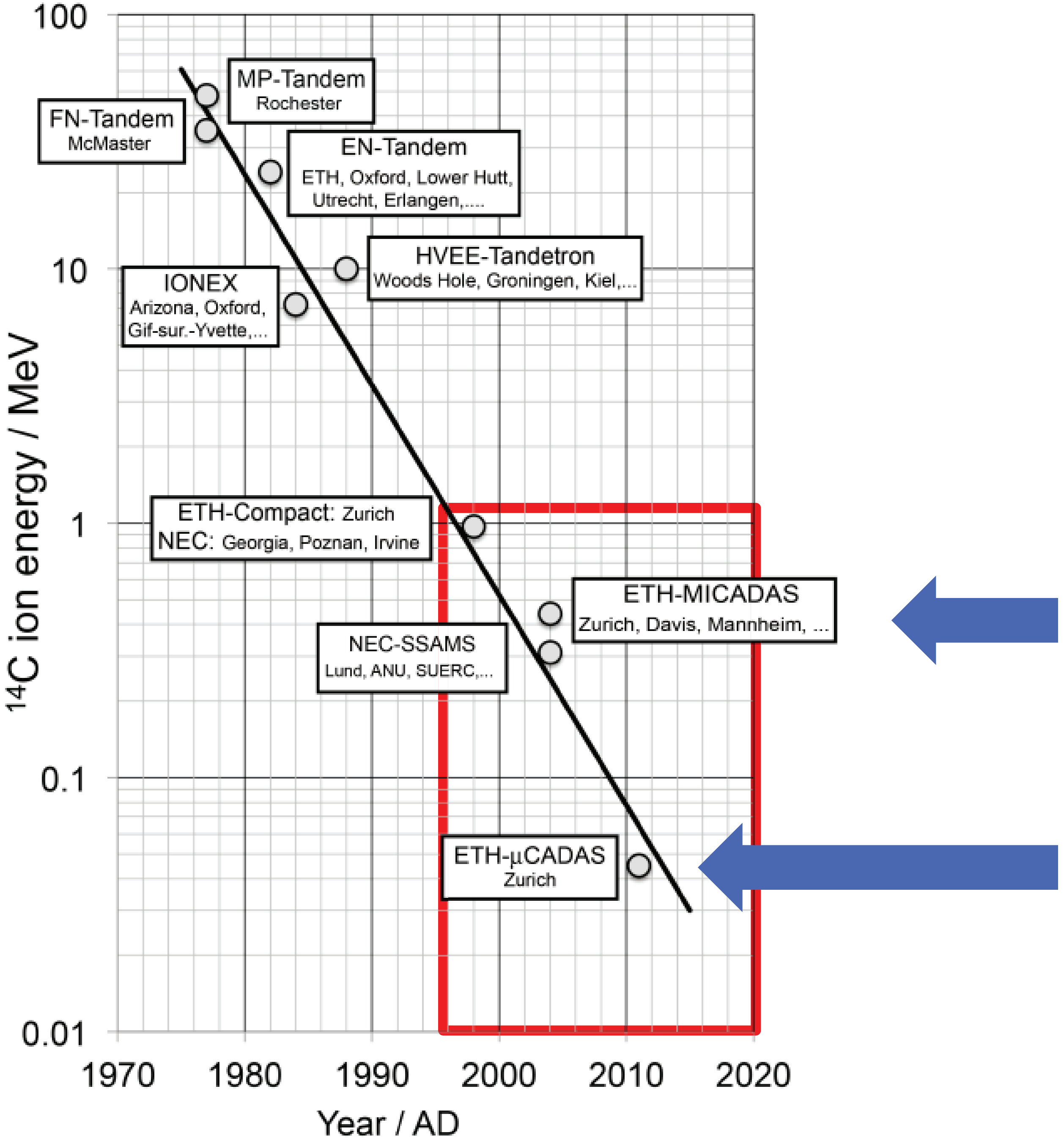
Performance not comparable
to state-of-the-art AMS systems
but
suitable for many applications !!!



Schematic of Bragg detector



Moor's Law in AMS

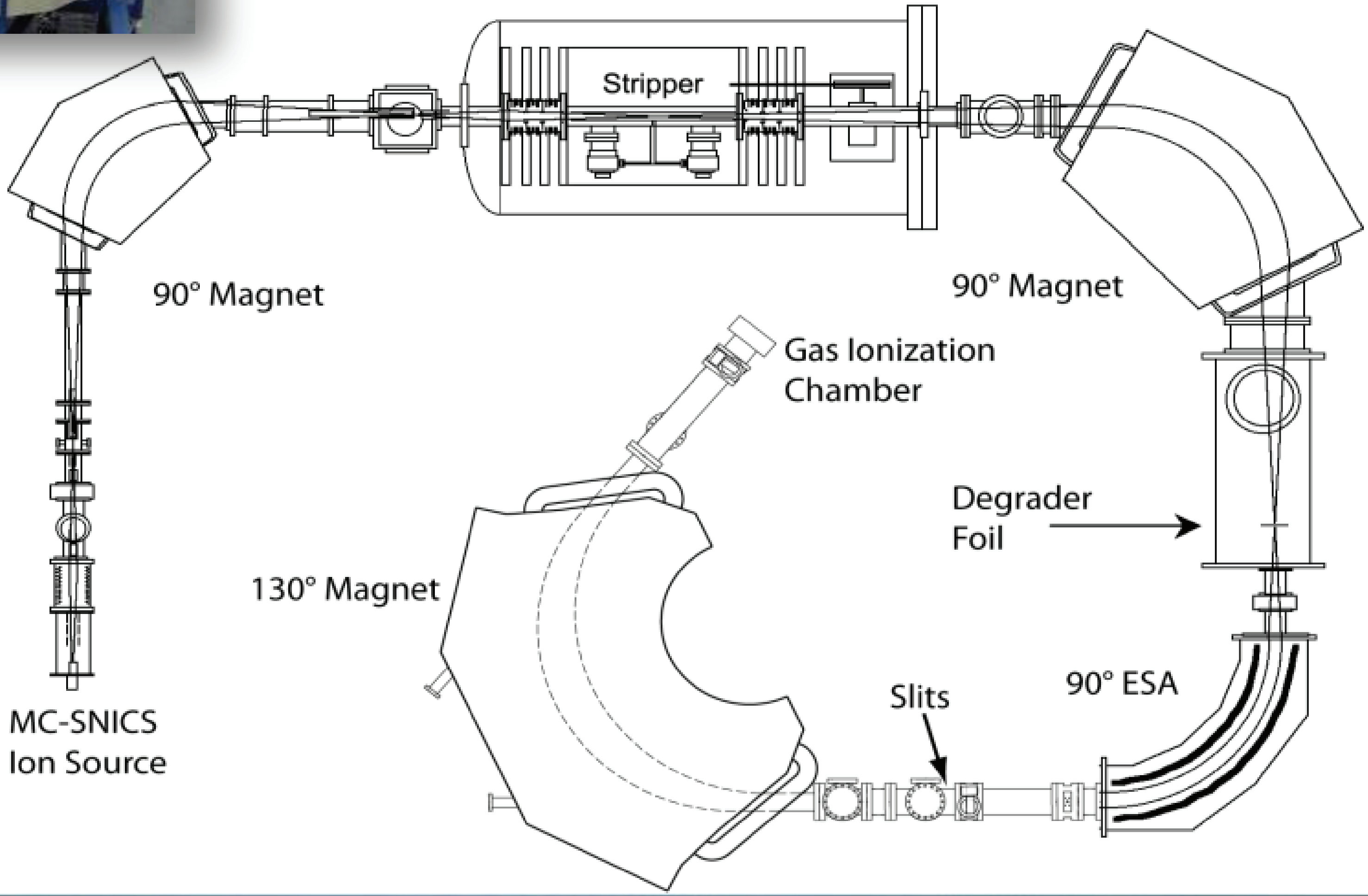


well established technology

further developments for routine operation of specific applications

Universal compact AMS instrument

- ^{10}Be – ^{10}B isobar suppression
- ^{14}C
- ^{26}Al
- ^{41}Ca
- ^{129}I
- Actinides (U, Pu, Pa, Np)

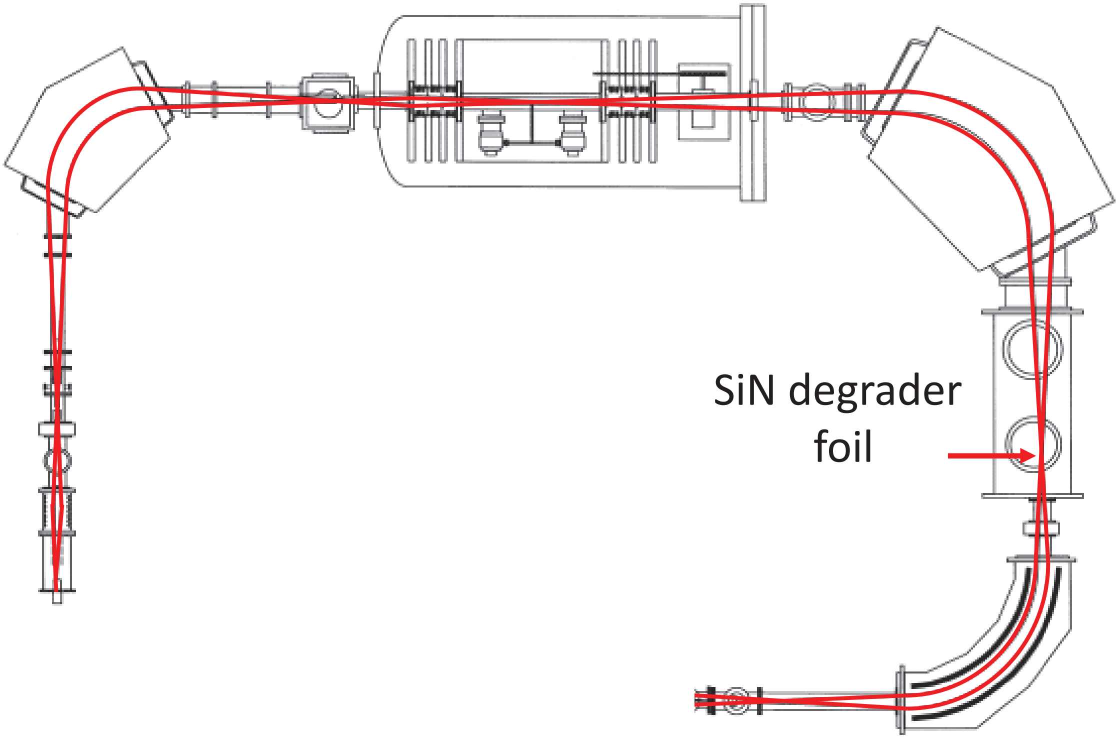


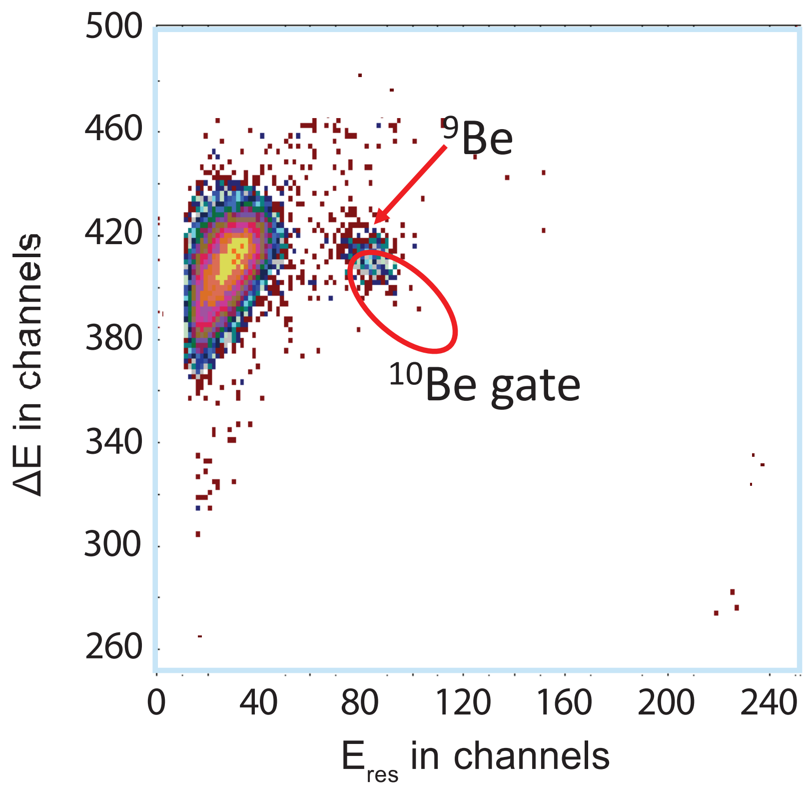
- *Progress*
  in detection technology

- *Better understanding of Stripping processes*

^{10}B isobar separation using SiN degrader

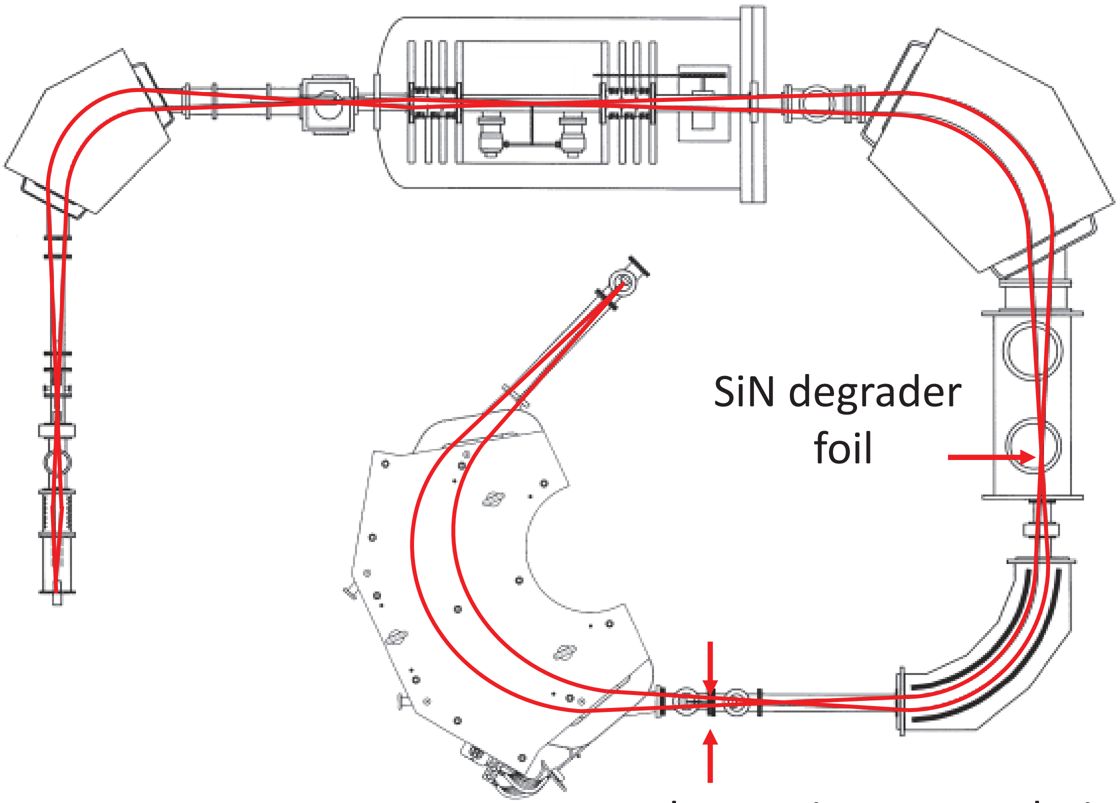
ETH- 500 kV Pelletron System

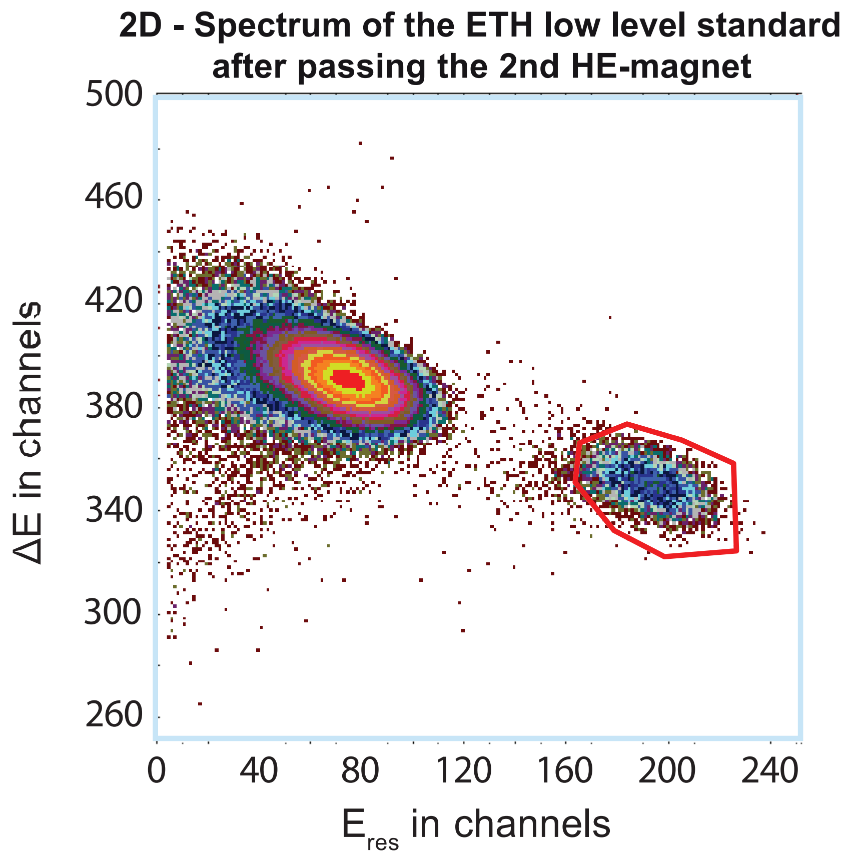




^{10}B isobar separation using SiN degrader

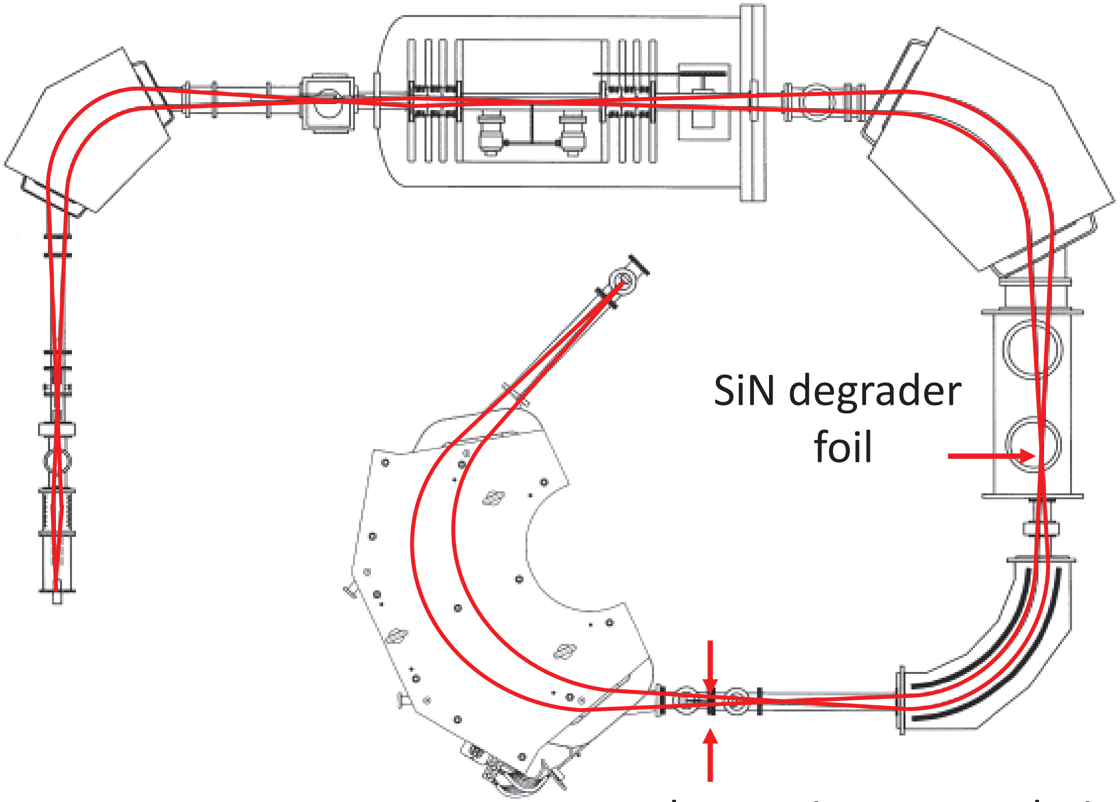
ETH- 500 kV Pelletron System



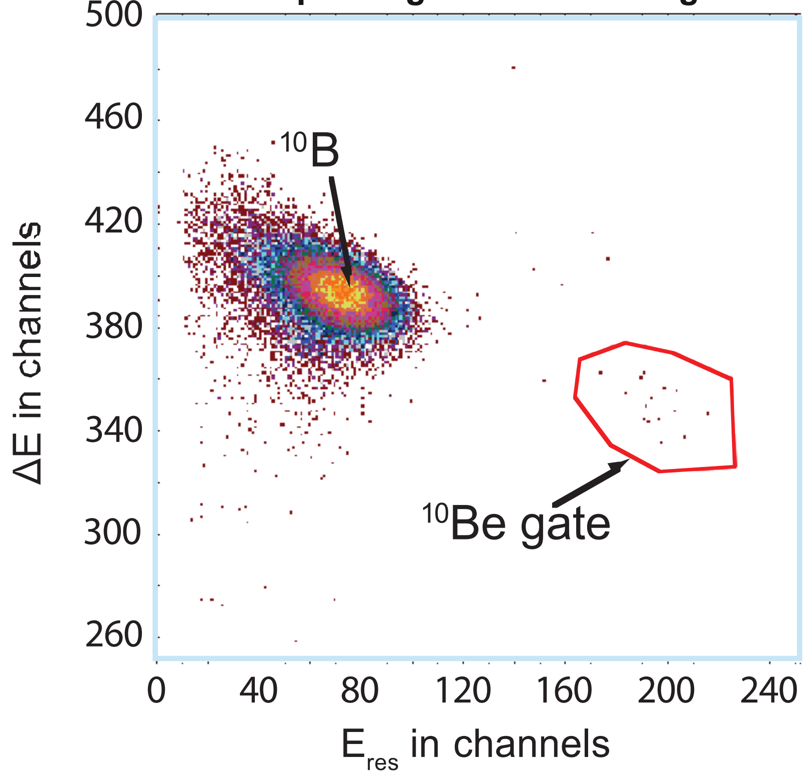


^{10}B isobar separation using SiN degrader

ETH- 500 kV Pelletron System



2D - Spectrum of the ETH low level standard after passing the 2nd HE-magnet



For comparison:
overall Transmission at large AMS facilities (6-8 MV)
(LLNL~ 35%, Aster ~25%)

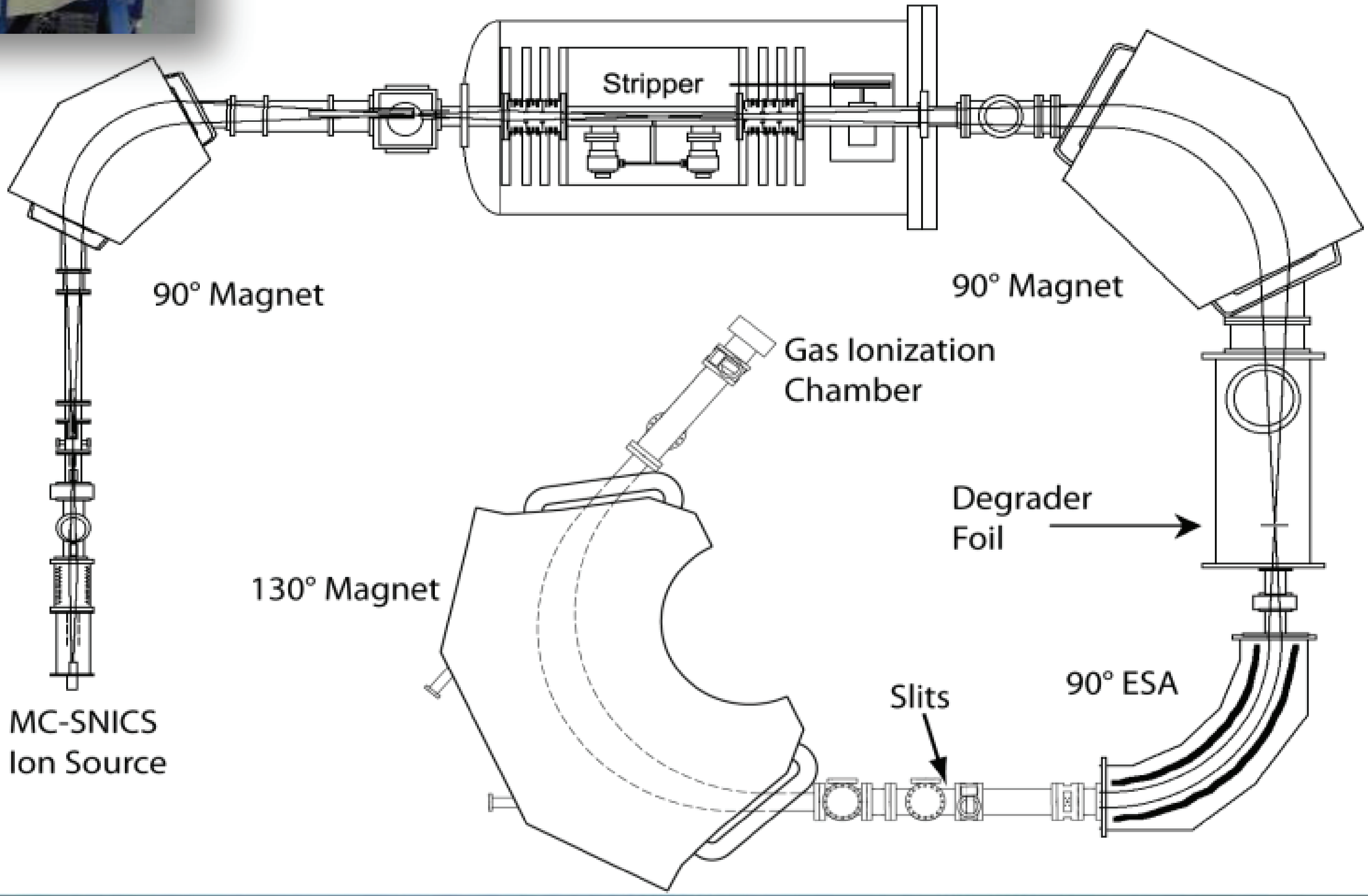
Transmission:

Degrader foil to detector:	20%
Stripper (charge state 1^+):	55%
→ LE-end to detector:	10-12%
$^{10}Be/^{9}Be$ background level:	10^{-15}

Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Universal compact AMS instrument

- ^{10}Be
- ^{14}C
- ^{26}Al
- ^{41}Ca
- ^{129}I
- Actinides (U, Pu, Pa, Np)



Improving ^{129}I measurements conditions

3+ interferences from ions in 1+ and 2+:

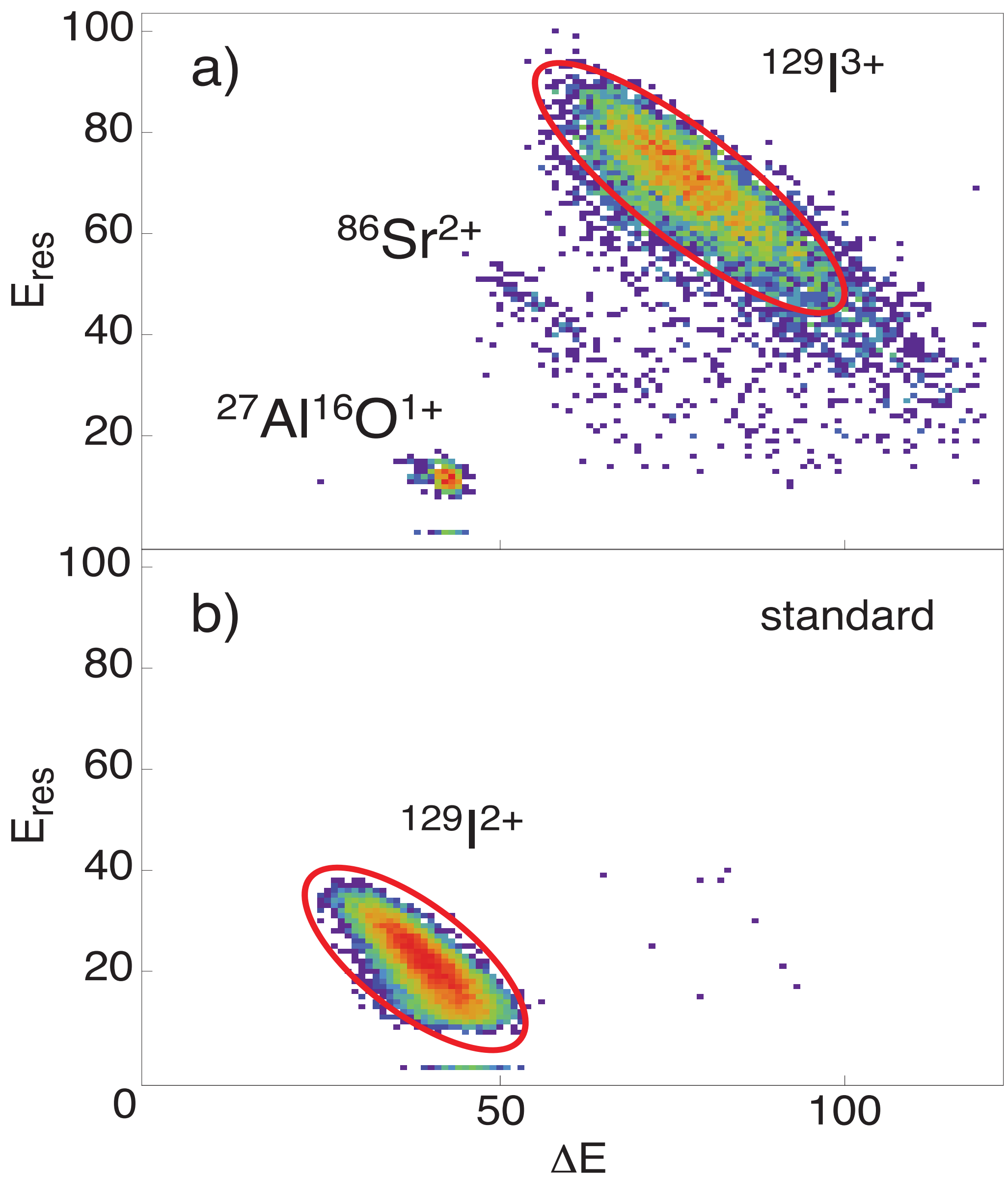
$129 \times 1/3 = 43$

$129 \times 2/3 = 86$

2+ no interference from ions in 1+:

$129 \times 1/2 = 64.5$

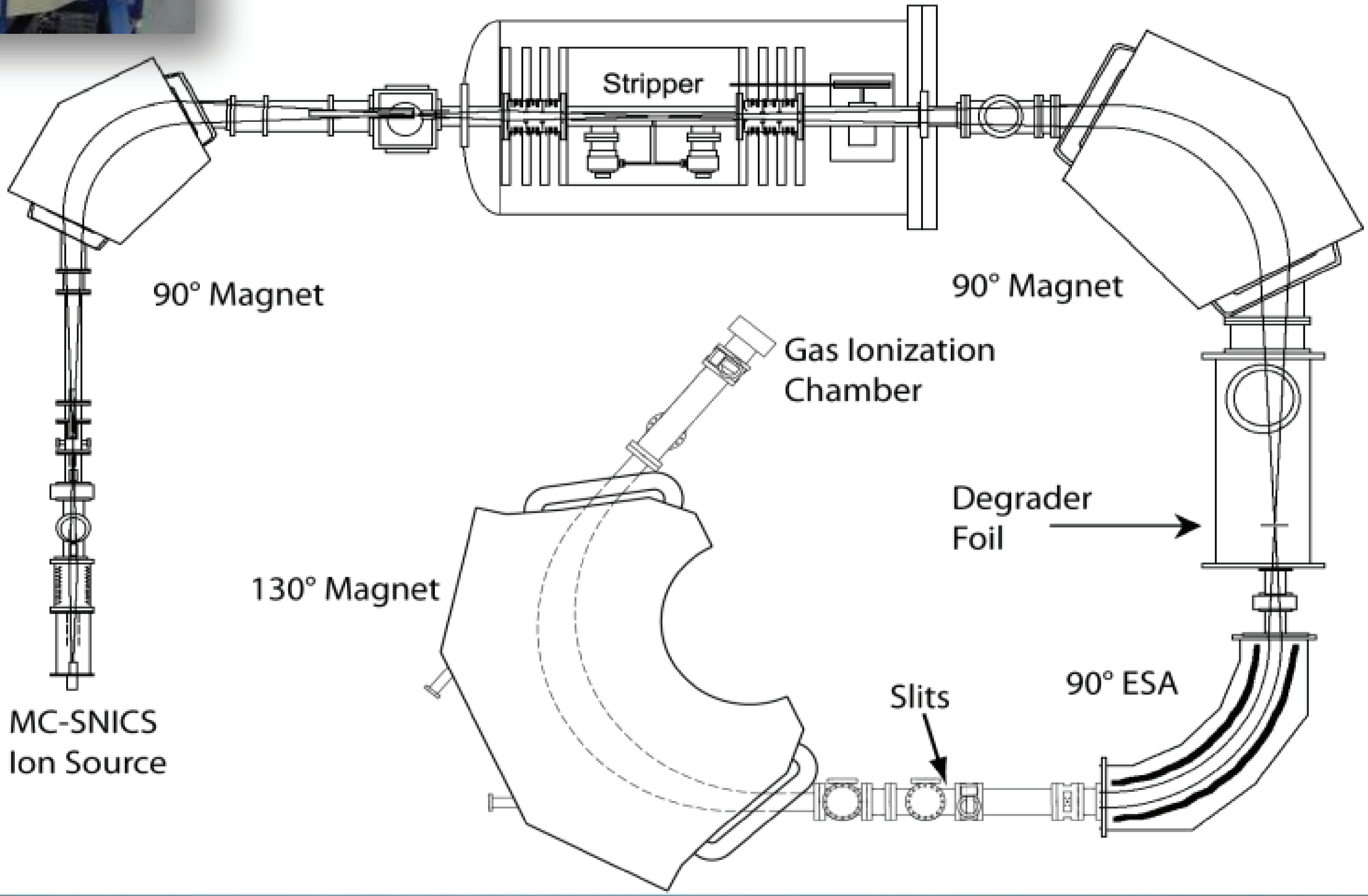
performance data compact AMS	
acceleration voltage	300 kV
charge state	2+
transmission	≈ 53 %
HE transmission	≈ 90 %
overall transmission	**≈ 48 %**
blank (cross contamination)	$\approx 1 \cdot 10^{-13}$



Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

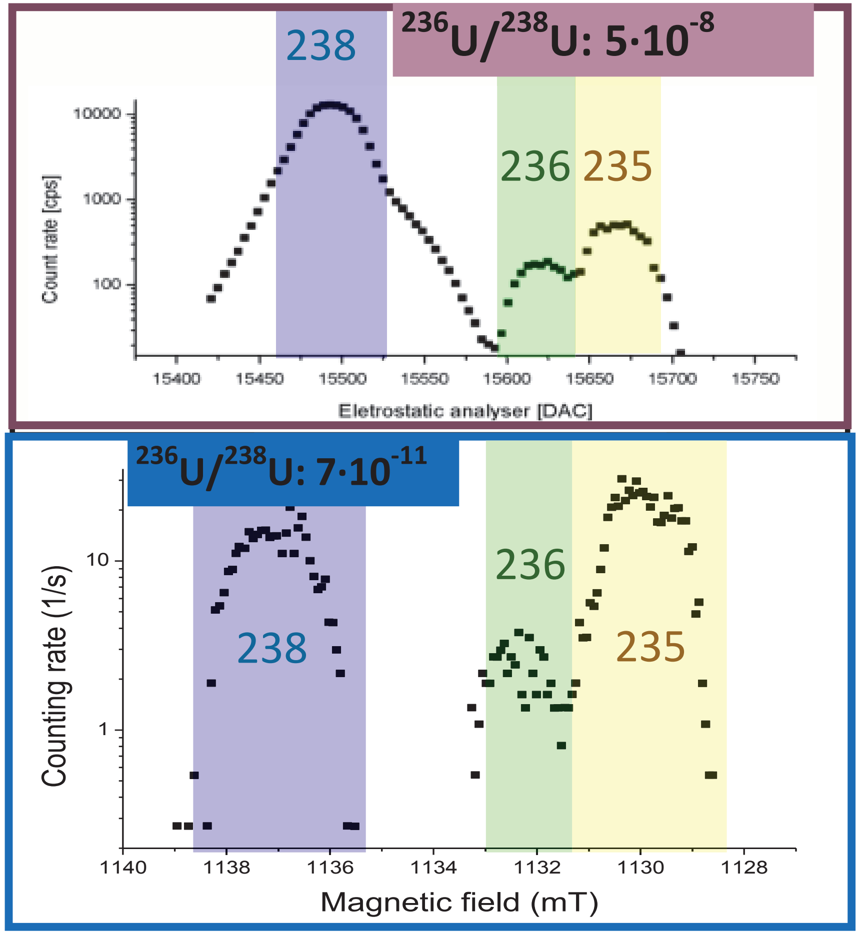
Universal compact AMS instrument

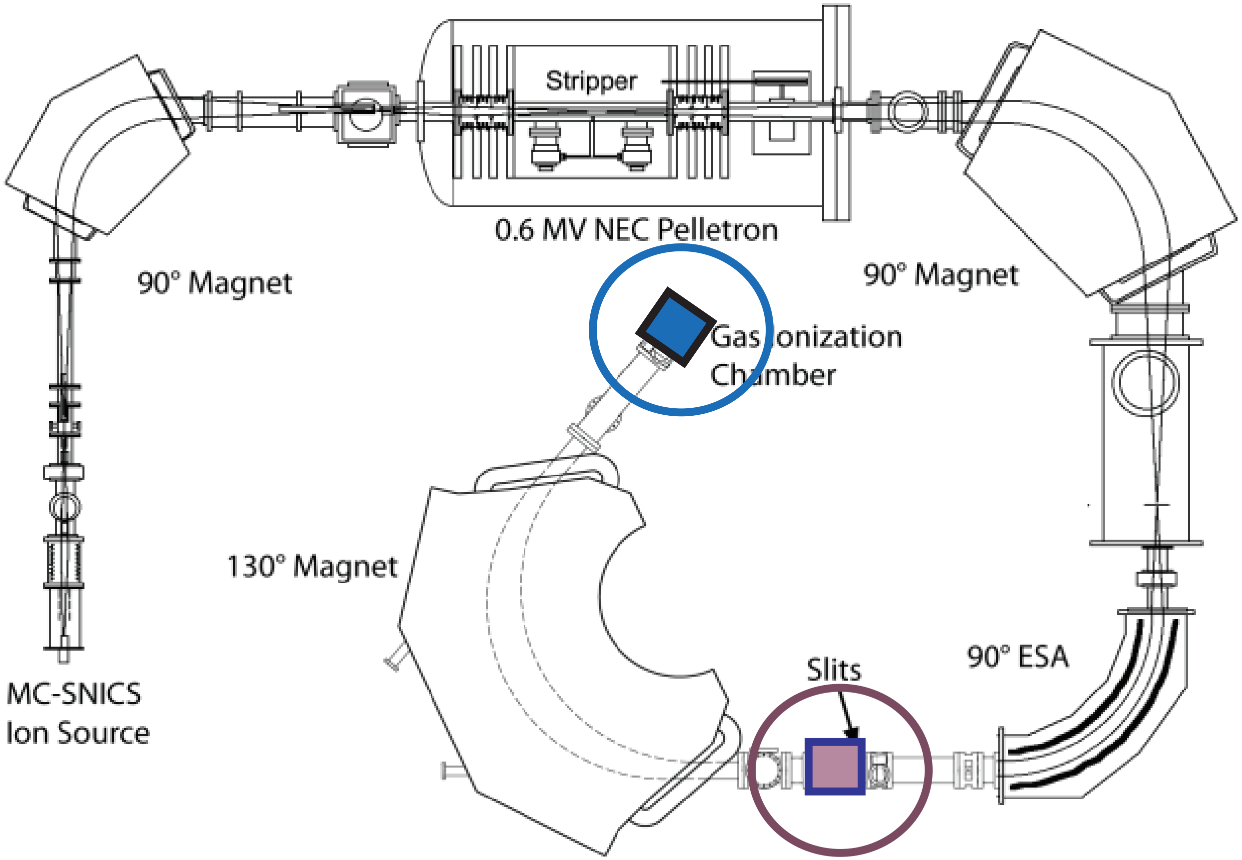
- ^{10}Be
- ^{14}C
- ^{26}Al
- ^{41}Ca
- ^{129}I
- Actinides (U, Pu, Pa, Np) – "no" isobars



Suppression of heavy isotopes ($^{235}U/^{236}U$)

- tailing and straggling of ^{235}U produces background @ m=236
- after the ESA: $^{236}U/^{238}U>10^{-9}$
- with additional magnet: $^{236}U/^{238}U <10^{-12}$



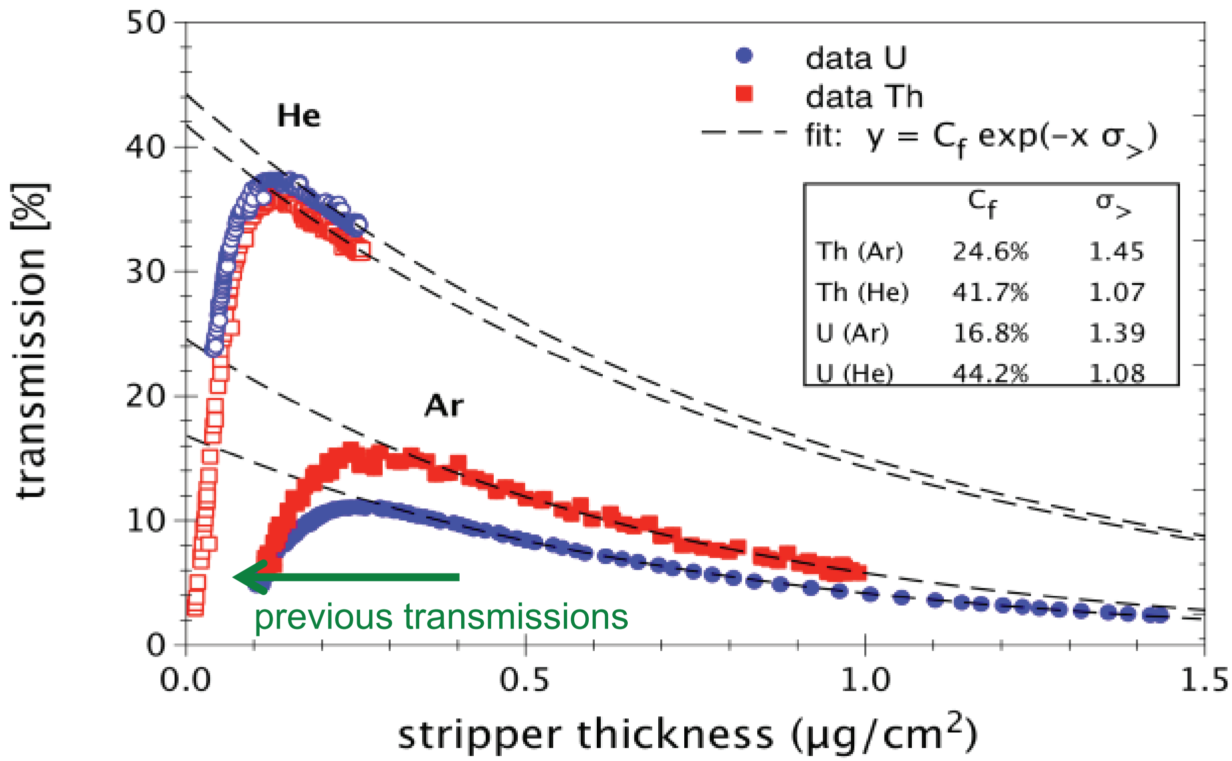


Improving stripping yield

He instead of Ar:

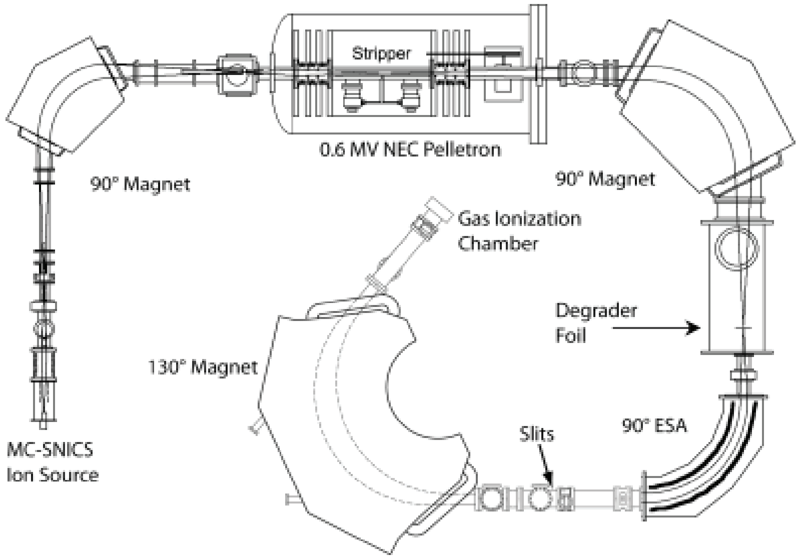
factor 3-4 more > 35% transmission

previous transmissions at other facilities:
≤ 5% transmission



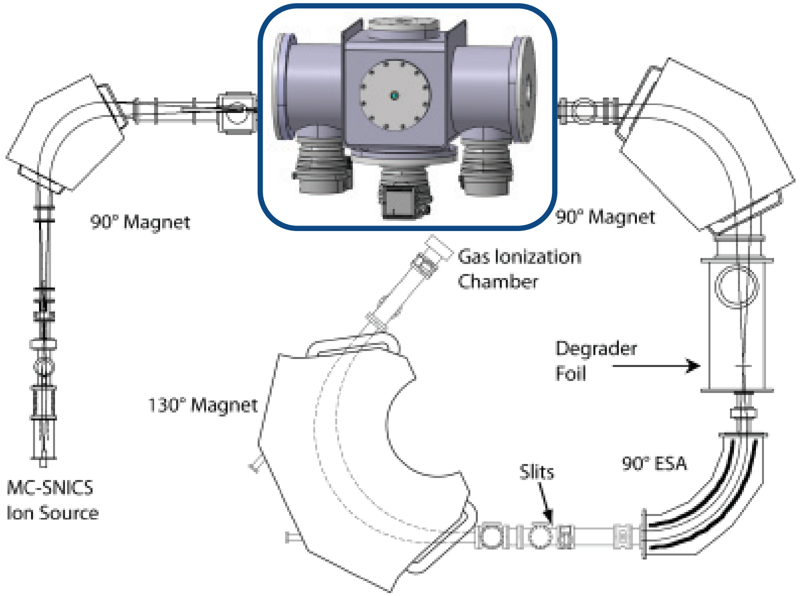
element	suppression neighboring masses	detection limit: 1 count equivalent	detection efficiency #counts/atoms
Thorium	$233/^{232}Th \approx 10^{-9}$ $231/^{232}Th \approx 4*10^{-12}$	≈ 10 ag	$4*10^{-5}$
Uran	$236/^{235}U \approx <10^{-10}$ $236/^{238}U \approx <10^{-12}$	≈ 5 ag	10^{-4}
Plutonium	$239/^{238}U \approx 3*10^{-10}$ $237/^{238}U \approx 10^{-12}$	≈ 10 ag	$5*10^{-5}$

Compact AMS: a versatile Instruments!! But, can we go on?



Compact AMS: a versatile Instruments!! But, can we go on?

MICADAS accelerator @ 300 kV



Measurement performance
equivalent or better
compared to Pelletron accelerator

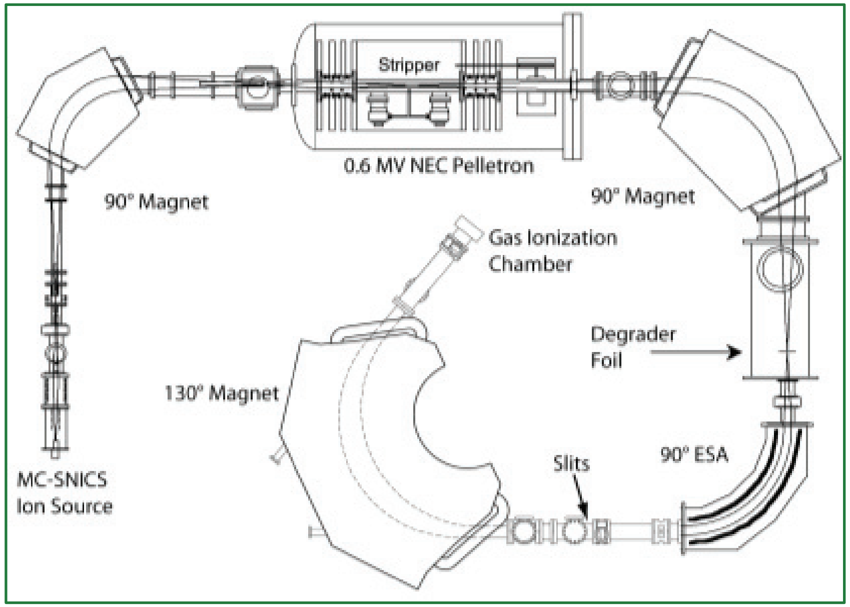
design of a new
Multi-isotope AMS spectrometer
driven by a
vacuum insulated HV platform

proof-of-principle experiment

- 300 kV technically feasible
- AMS test measurements performed
  - ^{236}U
  - ^{129}I
  - ^{41}Ca
  - ^{26}Al

Universal AMS system for ^{10}Be, ^{14}C, ^{26}Al, ^{41}Ca, ^{129}I, and actinides

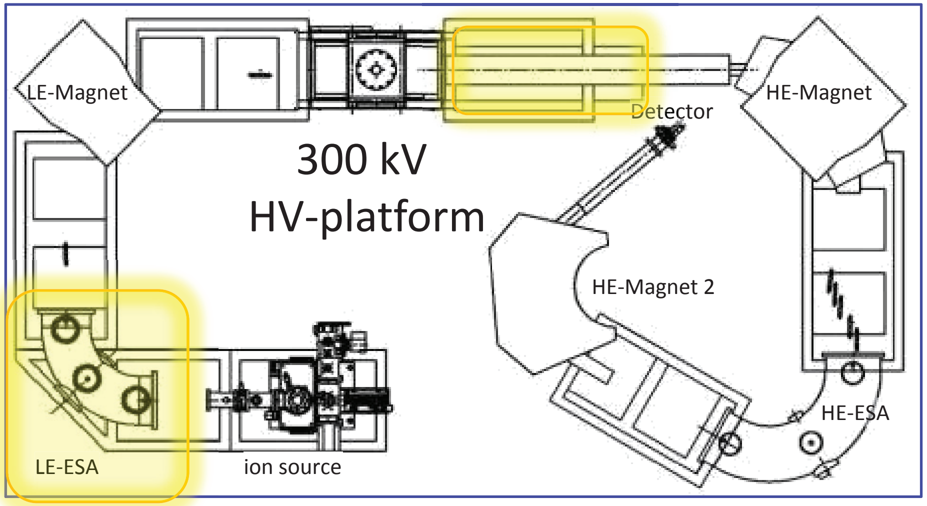
Compact universal AMS



4.3 m

6 m

New system (preview)



3.6 m

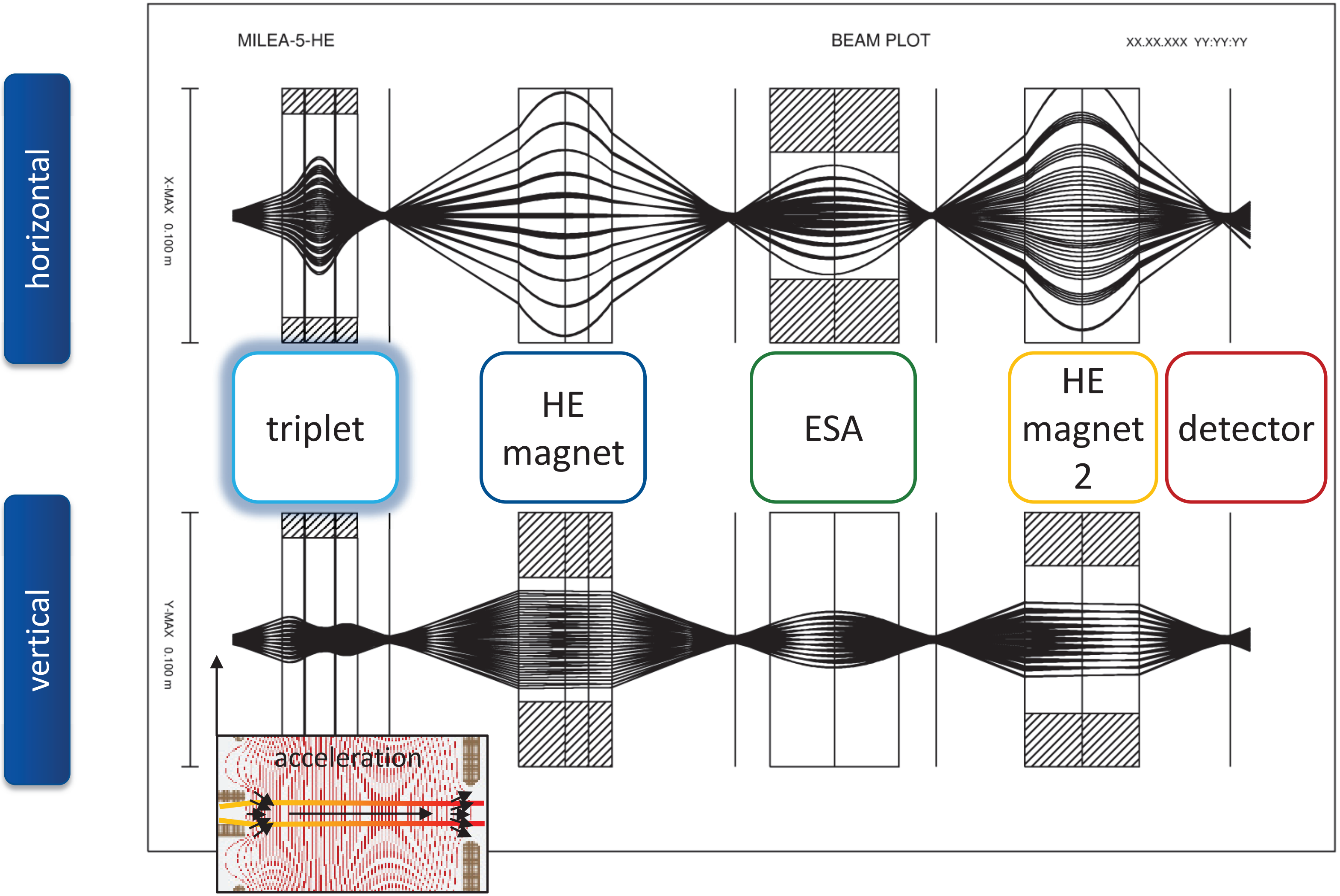
6.7 m

- Sputter ion source
- Additional ESA on injection side
- Compact acceleration & optimized stripper
- Quadrupole lens triplet after acceleration
- Increased angular acceptance on HE side
- …

Improved Ion optic beam transport

Because focusing effect of acceleration depends on particle charge state

✓ Can be corrected by using a quadrupole lens system

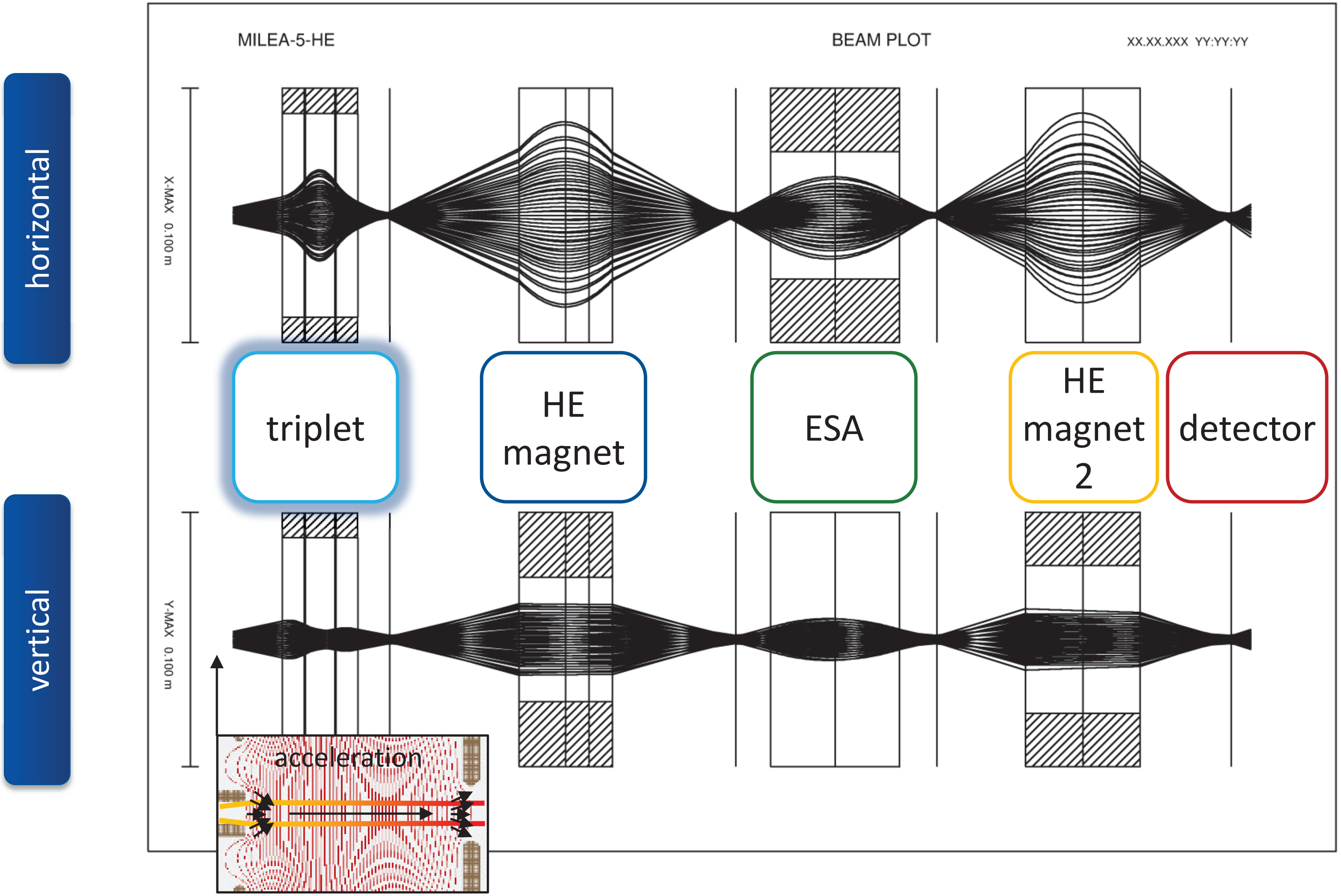


1+ (^{14}C)

Improved Ion optic beam transport

Because focusing effect of acceleration depends on particle charge state

✓ Can be corrected by using a quadrupole lens system

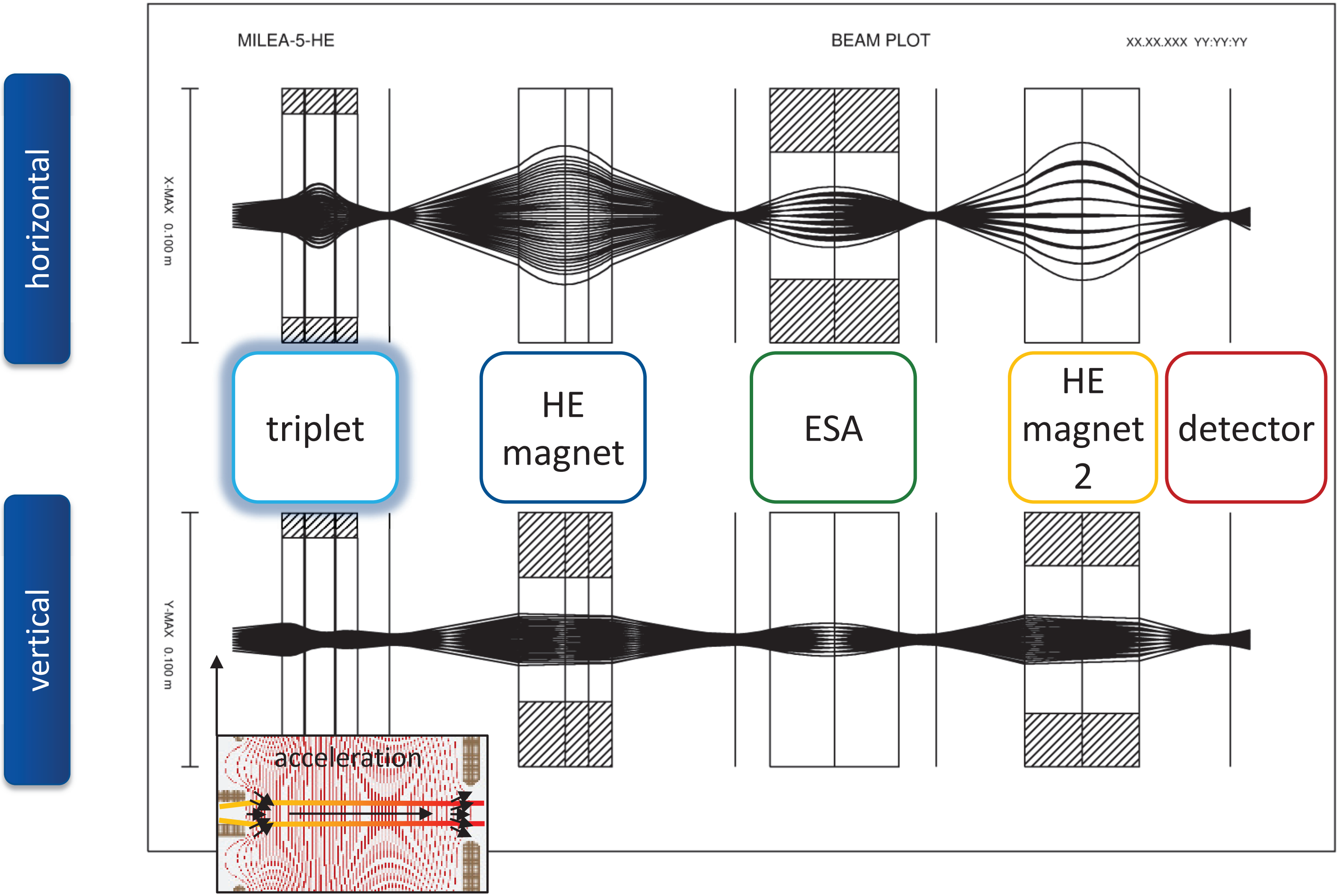


2+ (^{129}I)

Improved Ion optic beam transport

Because focusing effect of acceleration depends on particle charge state

✓ Can be corrected by using a quadrupole lens system

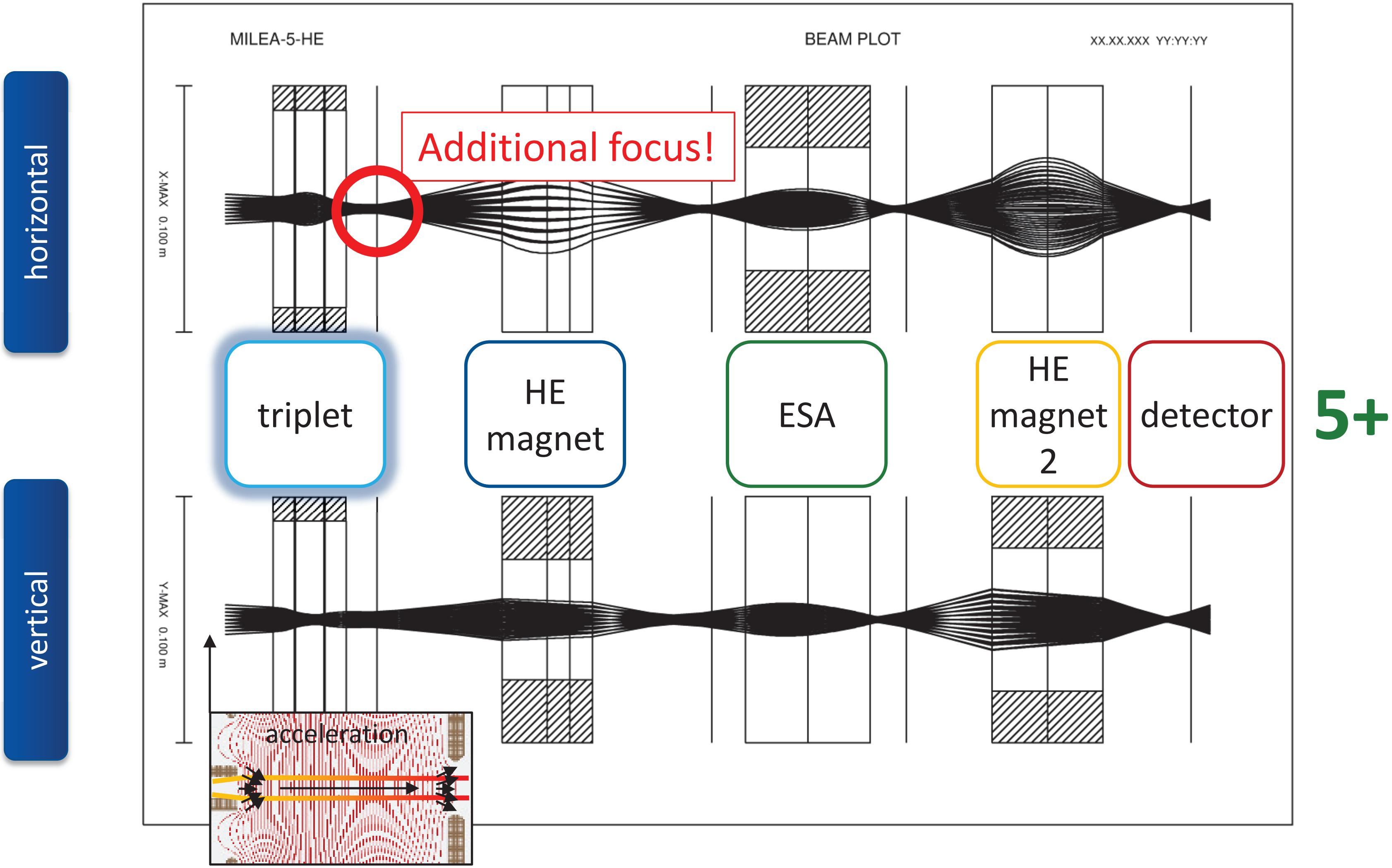


3+ (^{236}U)

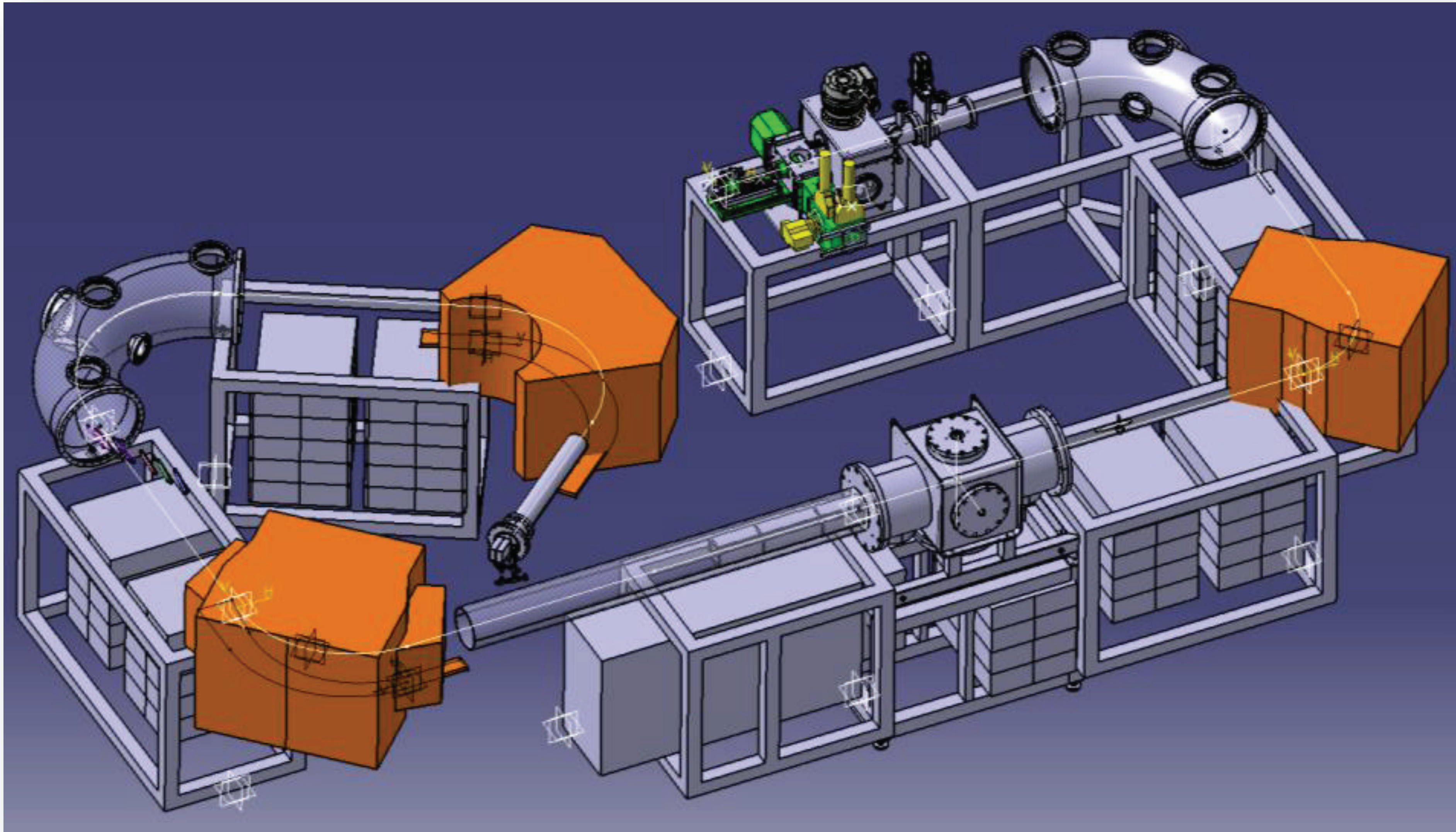
Improved Ion optic beam transport

Because focusing effect of acceleration depends on particle charge state

✓ Can be corrected by using a quadrupole lens system

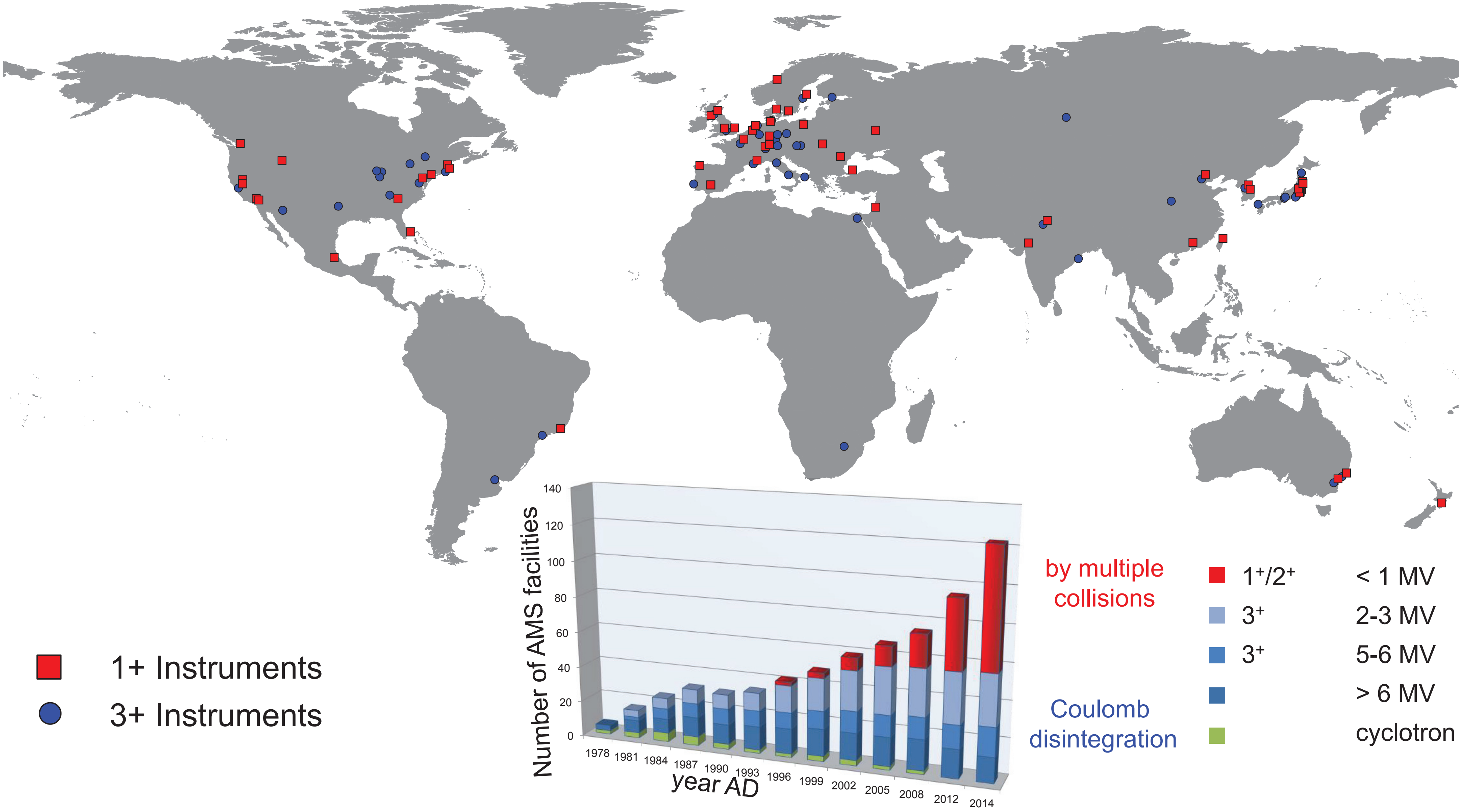


Prototype development ongoing



AMS measurements of ^{14}C, ^{10}Be, ^{26}Al, ^{41}Ca, ^{129}I, actinides, …. 2017??

AMS-World today



Conclusions

- AMS has matured over the past 40 years
  - High performance instruments are available from various commercial suppliers
  - Instruments can be tailored to the specific needs of the users

- Major breakthrough has been reached by introducing stripping to 1+
  - molecule interferences can be destroyed efficiently
  - Compact AMS systems are now most favored type of instruments

- Tradition accelerator technology can be replaced (200 kV HV power supplies)
- He stripping is the key to enhanced performance of instruments
- Permanent magnets reduce complexity and operation cost of dedicated ^{14}C Instruments
  - High ion optical transmission for ^{14}C
  - Stable an reproducible measurement conditions
  - Enable measurement performance similar to tradition MS

- First steps towards a true mass spectrometer for ^{14}C detection has been made
  - present day performance not jet on a competitive basis to i.e. 200 kV MICADAS system
  - but potential for high throughput / lower precision ^{14}C (e.g. bio/environmental science)

- HV power supply driven system are suitable for detection of all major AMS nuclides
- He stripping provides unique transmission efficiencies
  - for very heavy nuclides such as ^{129}I and actinides the will outperform any other approach
  - dedicated instruments will be available soon

Conclusions

- Progress became possible because:
  - systematic research of the physics of the fundamental processes behind the AMS method
  - charge exchange processes, angular straggling, molecule dissociation cross sections

- Simulation and mathematical modelling tools have been developed
  - optimize ion beam transport
  - Simulate ion interaction with e.g. stripper gas or within the particle detector

- Tradition accelerator technology can be replaced (200 kV HV power supplies)
- He stripping is the key to enhanced performance of instruments
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