New and Improved AMS Facilities

Hans-Arno Synal

ETH

Laboratory of Ion Beam Physics

8093 Zurich

Switzerland

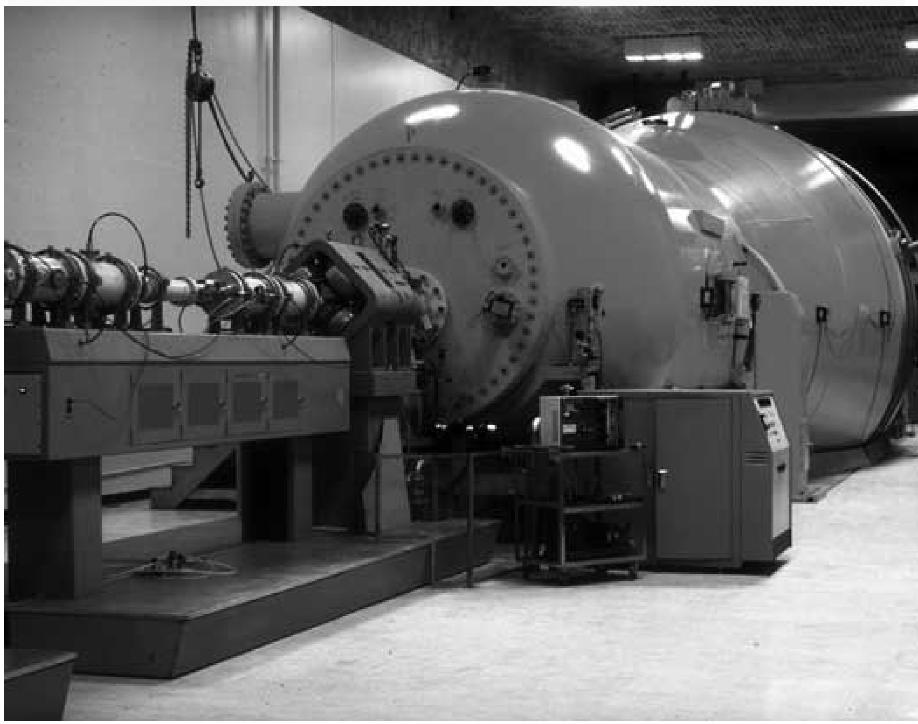
Discovery of AMS in 1977

The New York Times

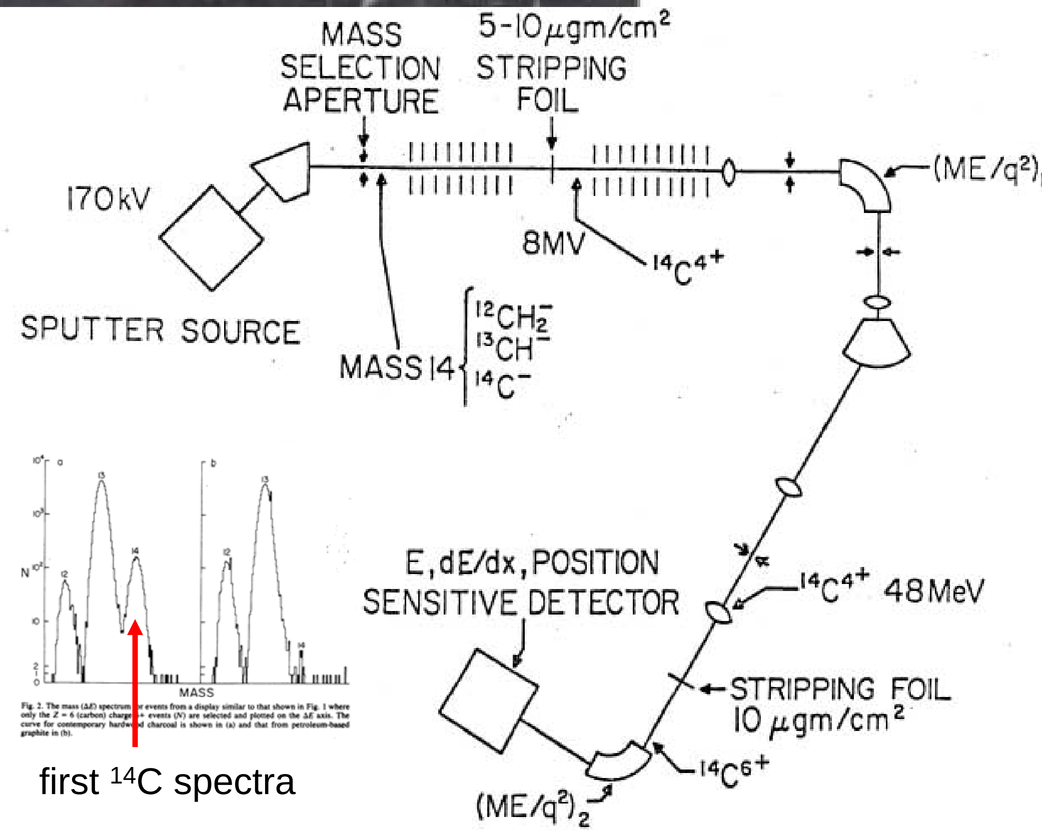
NEW YORK, THURSDAY, JUNE 9, 1977

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

AMS-Heros
A.E. Litherland
K.H. Purser
H.E. Gove
R.P. Beukens
R.P. Clover
W.E. Sondheim
R.B. Liebert
C.L. Bennet

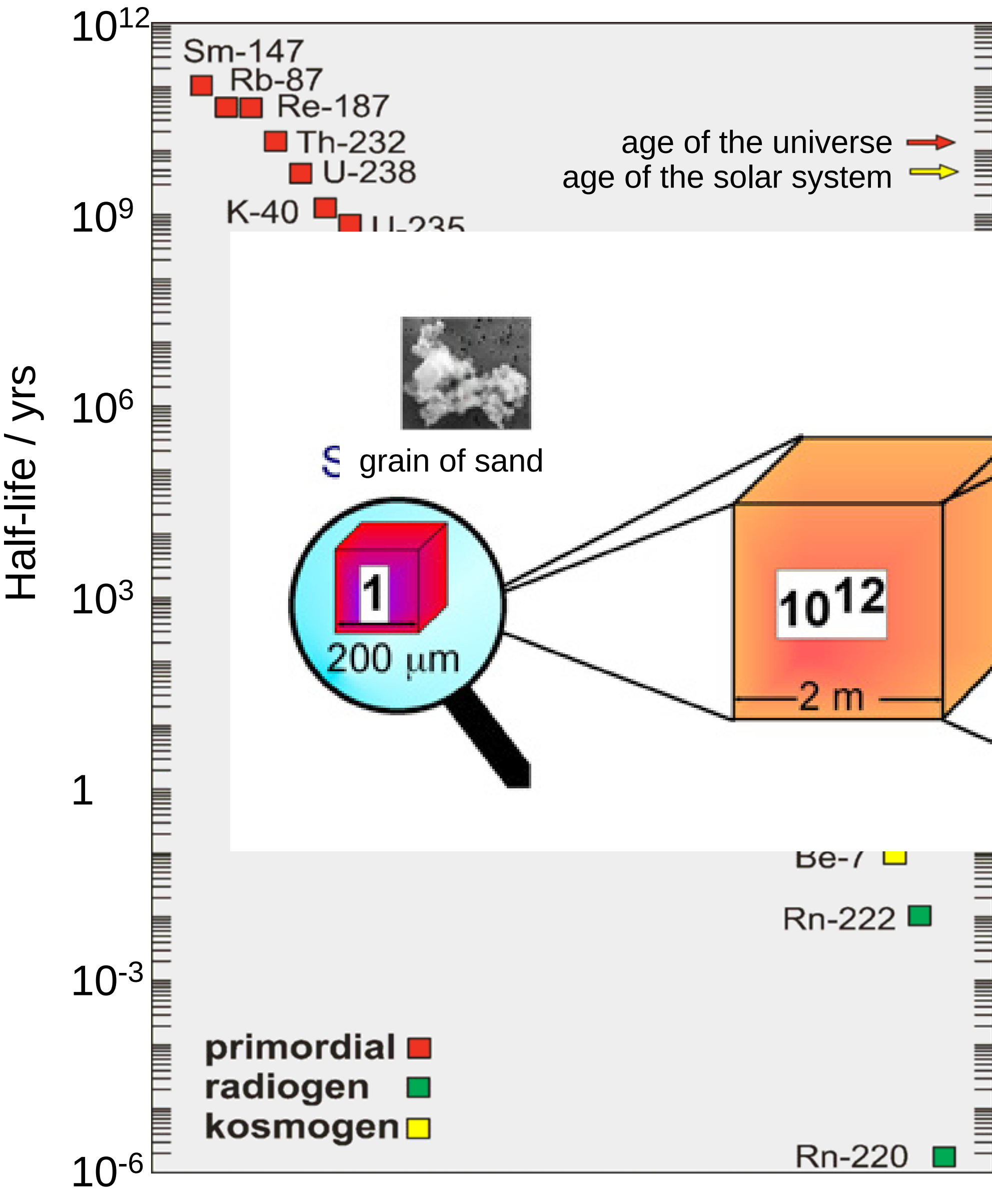


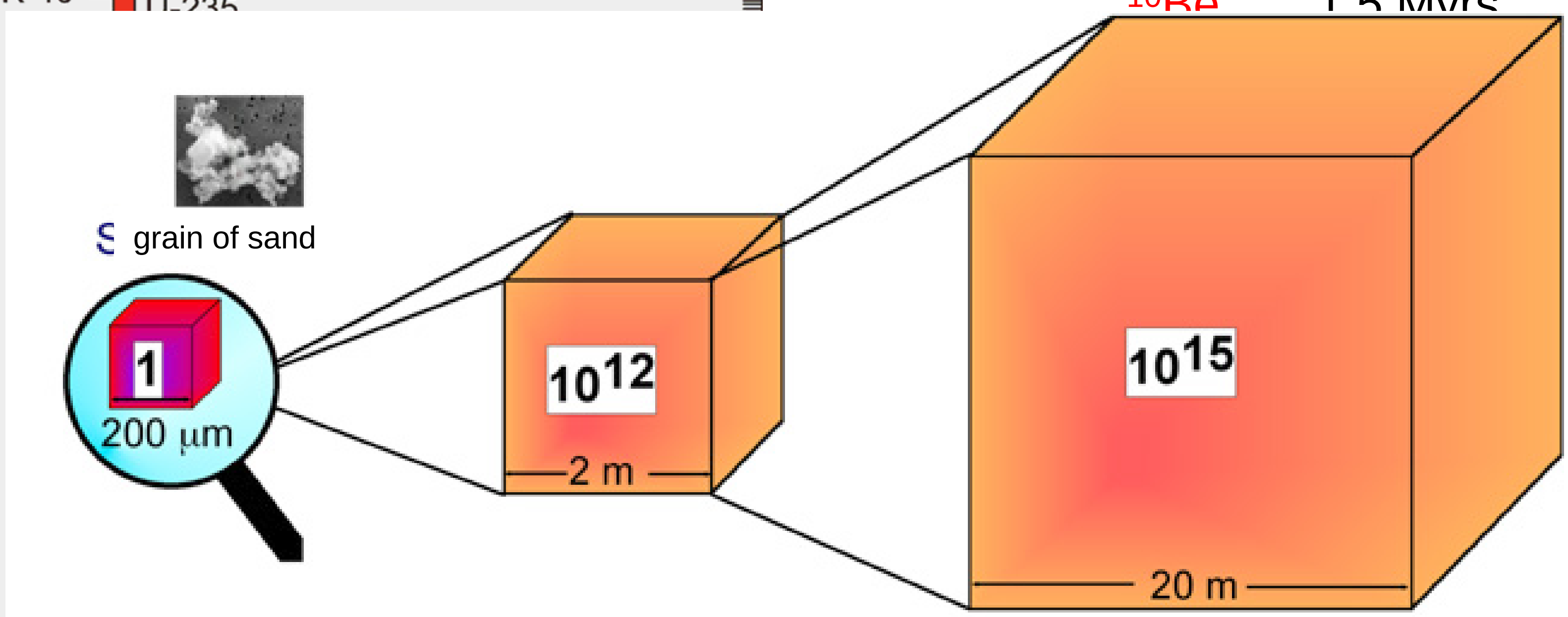
The Rochester MP Tandem accelerator



first ^{14}C spectra

Long-lived / AMS radionuclides





^{14}C 5730 yrs

Archaeology, Art History, Oceanography, Earth & Environmental Sciences

^{10}Be 1.5 Myrs

Earth &

omedical

ctivity,

ons

^{129}I 16 Myrs

Earth & Environmental Sciences, Radioecology, Nuclear safeguards,...

Actinides (Pu, U, Pa)

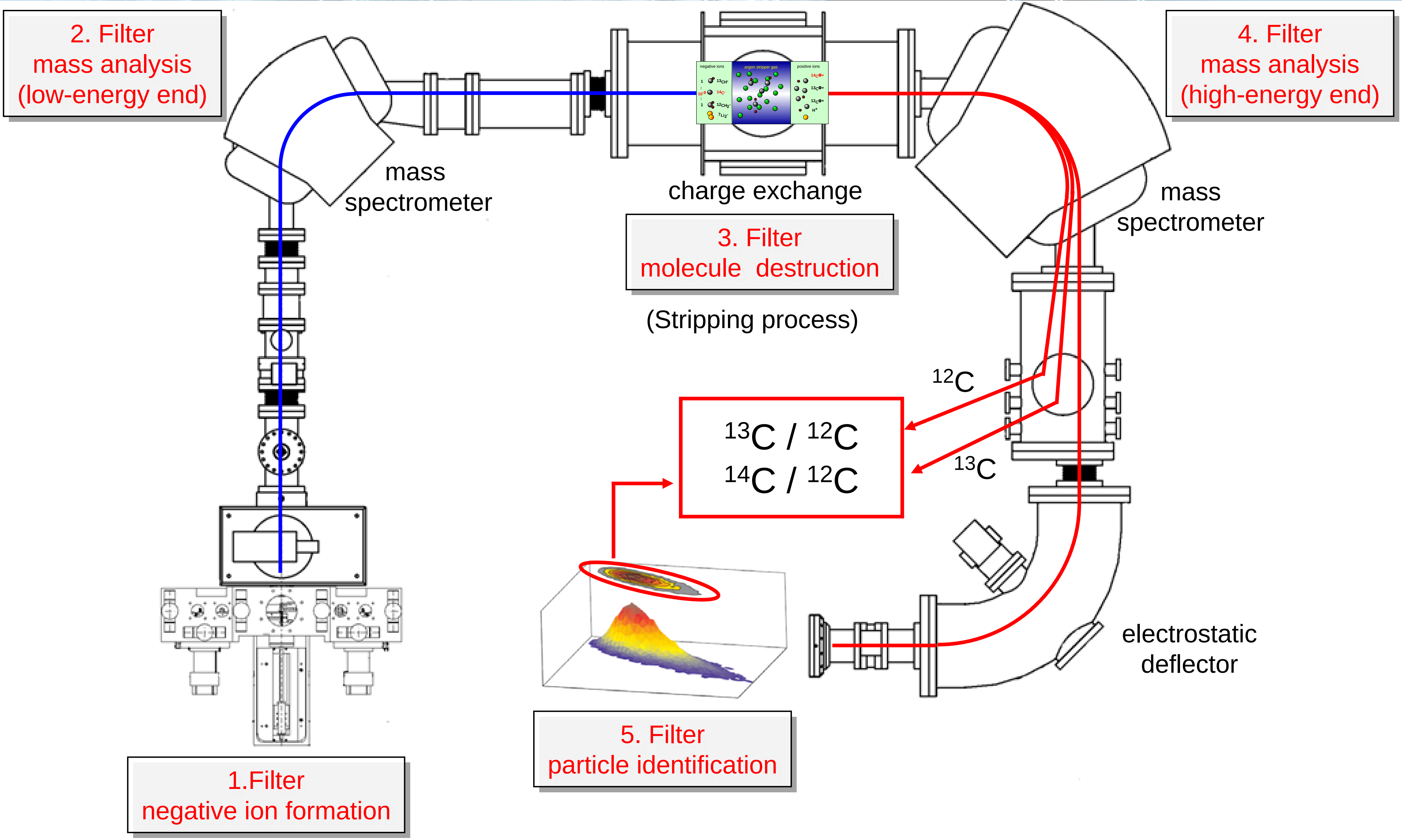
Radioecology, Nuclear Safeguards,Oceanography...

Why does AMS reach unparalleled sensitivity?

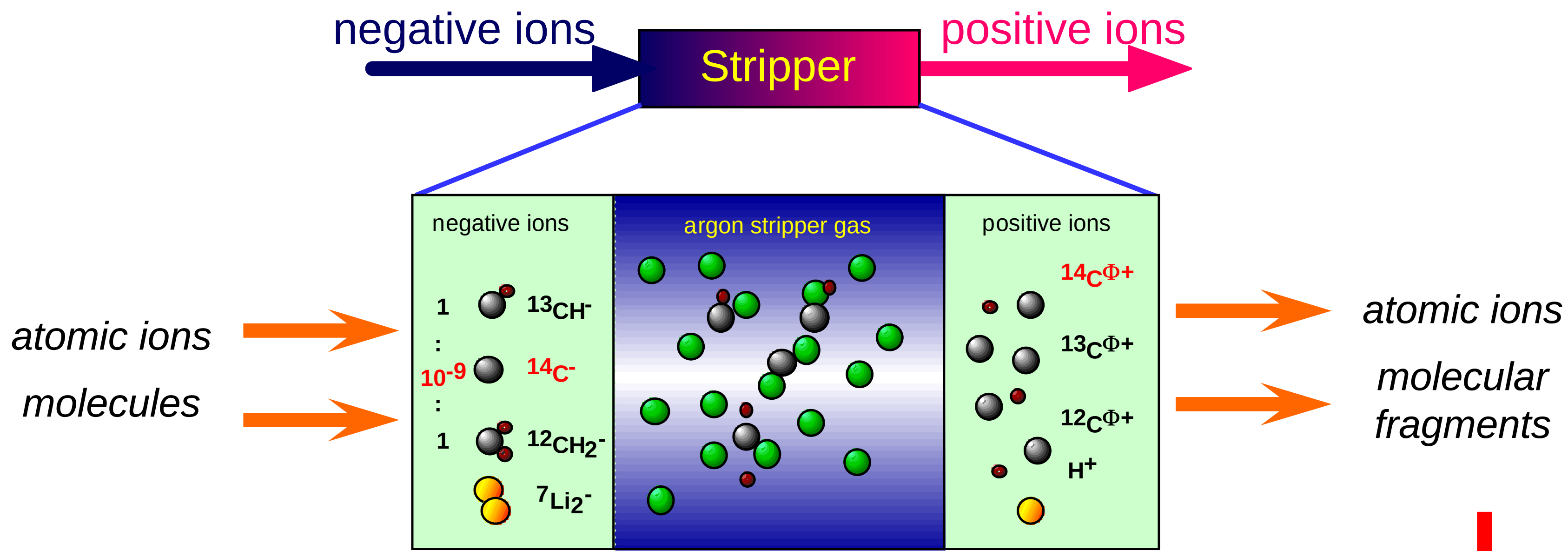
- Isobar separation
  - Negative ion formation
    - $^{14}C(^{14}N)$; $^{26}Al(^{26}Mg)$;
    - $^{10}Be(^{10}B)$; $^{36}Cl(^{36}S)$; $^{41}Ca(^{41}K)$;
    - $^{10}BeF/BeBaF(^{10}BF)$; $^{41}CaH_3(^{41}KH_3)$; $^{41}CaF_3(^{41}KF_3)$
  - Single ion detection
- Abundance sensitivity ($1:10^{15}$)
 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

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

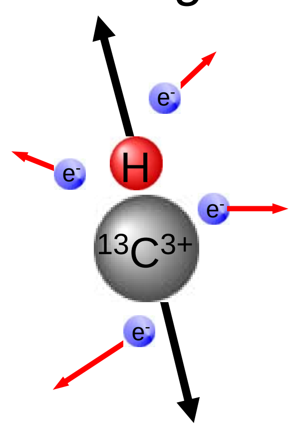
AMS: principles of operation



The Golden AMS Rule: $q > 3^{+}$!!!



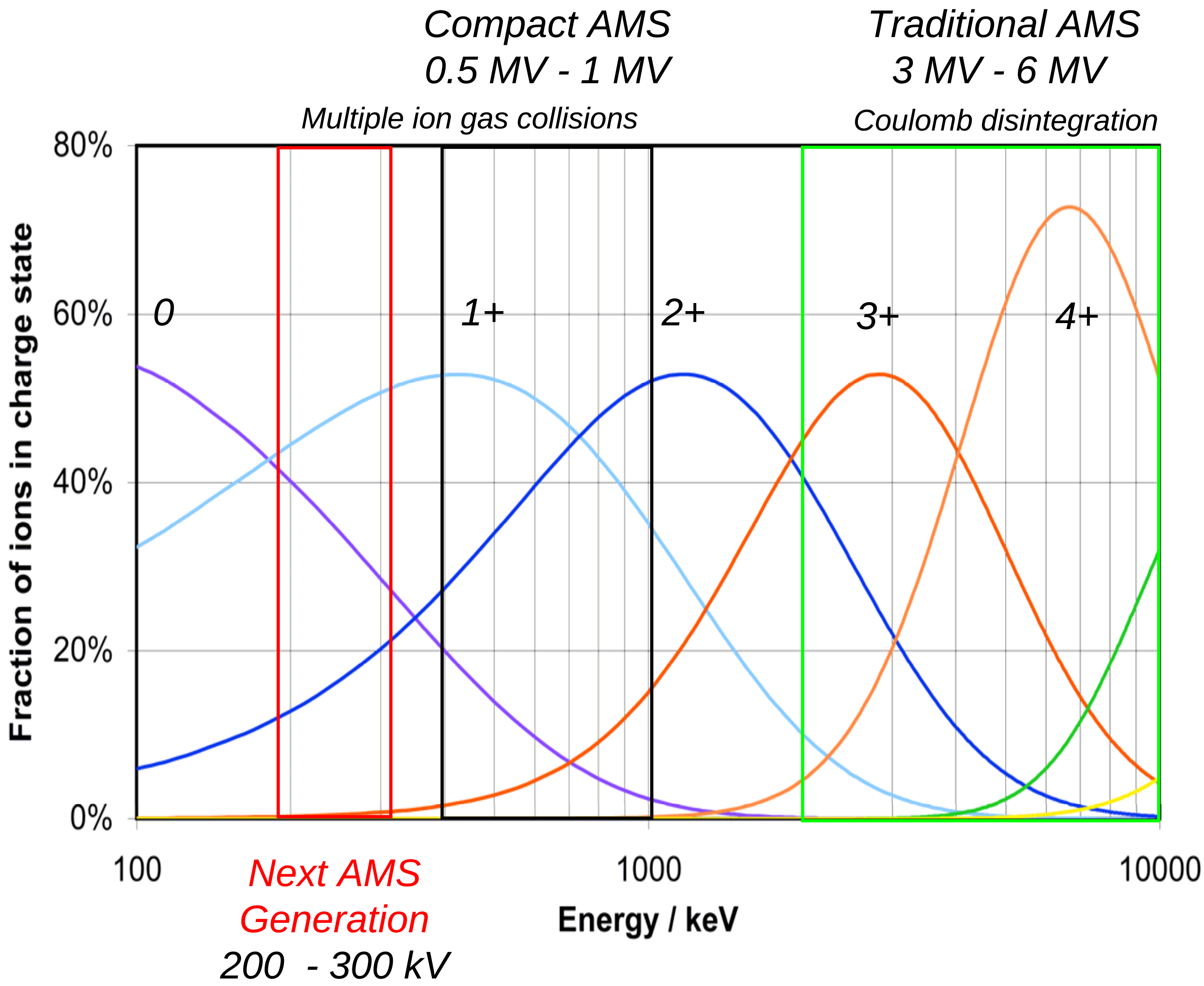
Coulomb disintegration ($q \geq 3^{+}$)



maximum yield (^{14}C):
high energies and
large accelerators

$q=3^{+}$	2.5 MV
$q=4^{+}$	6.5 MV

Charge state yield of ^{14}C ions in Ar gas



Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology

Breaking the Rule: AMS with $q = 1^+, 2^+$ Ions

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

The $^{12}CH_2^{2+}$ molecule has been studied and it was found that the molecule can be effectively eliminated thus allowing detection of $^{14}C^{2+}$ at low terminal voltages of a tandem accelerator. Some implications of this discovery for radiocarbon dating are discussed.

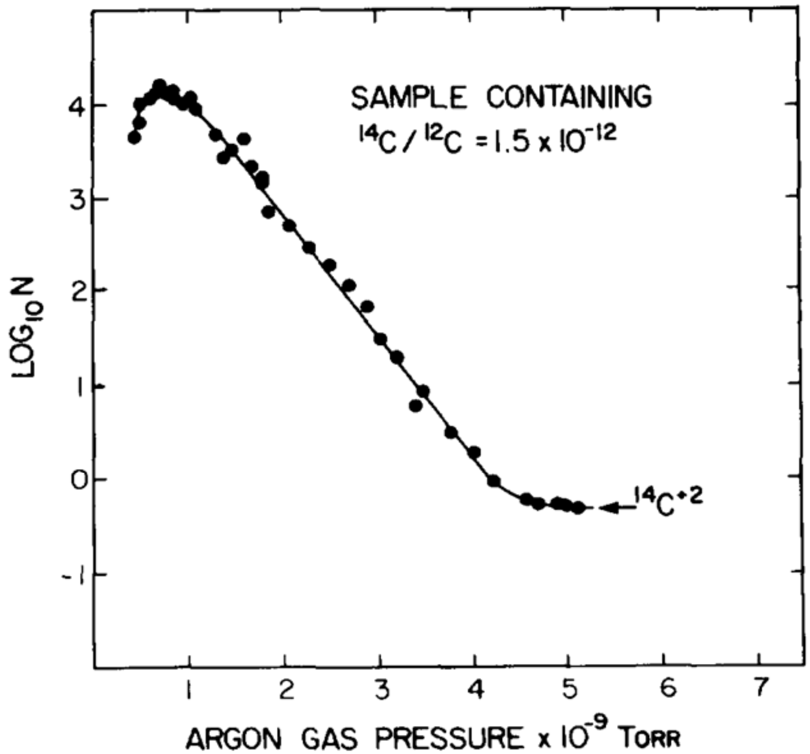
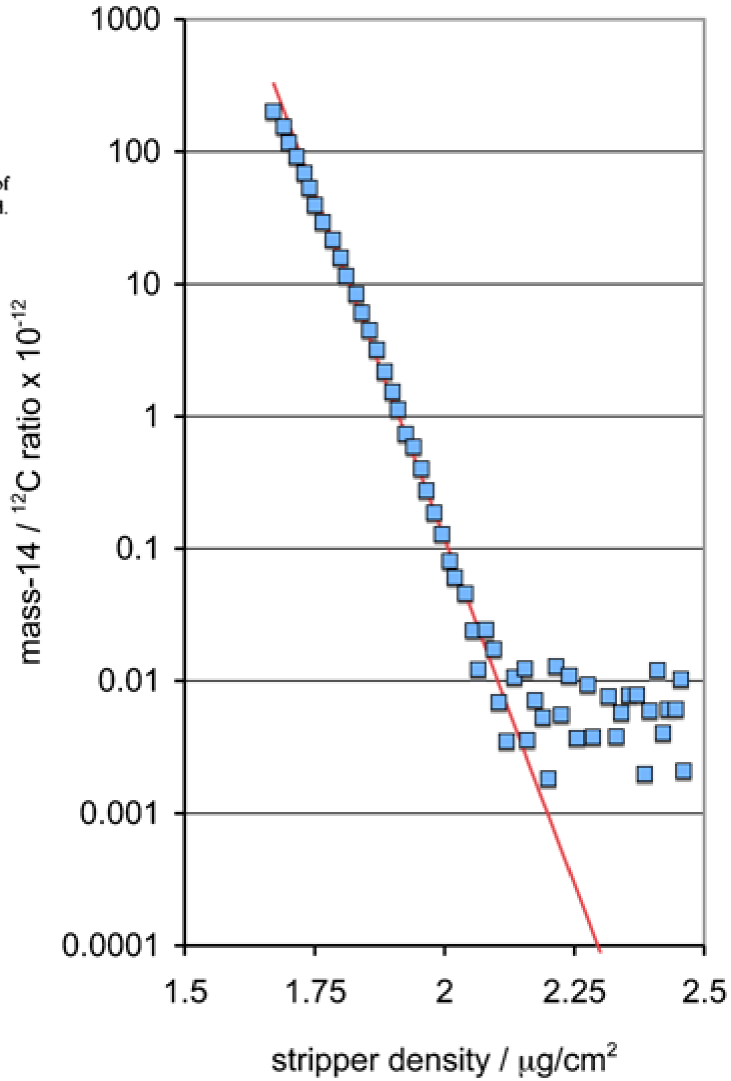


Fig. 2. The $^{12}C^{2+}$ current and number of mass 14 counts as a function of the argon stripper pressure. The plateau in the mass 14 curve is due to $^{14}C^{2+}$ ions along with a very small contribution of $^{14}N^{2+}$ from the modern sample.



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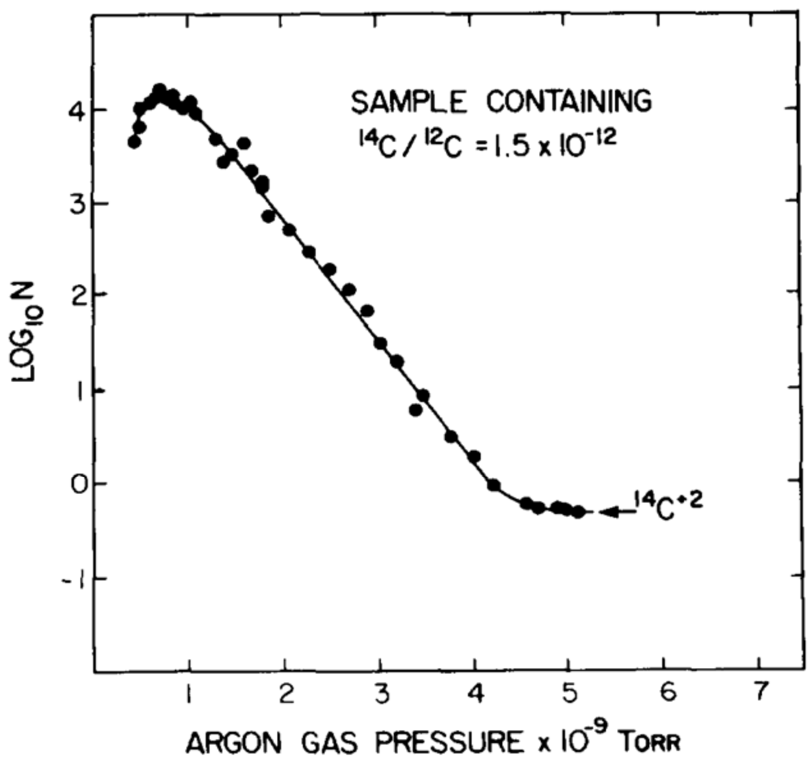

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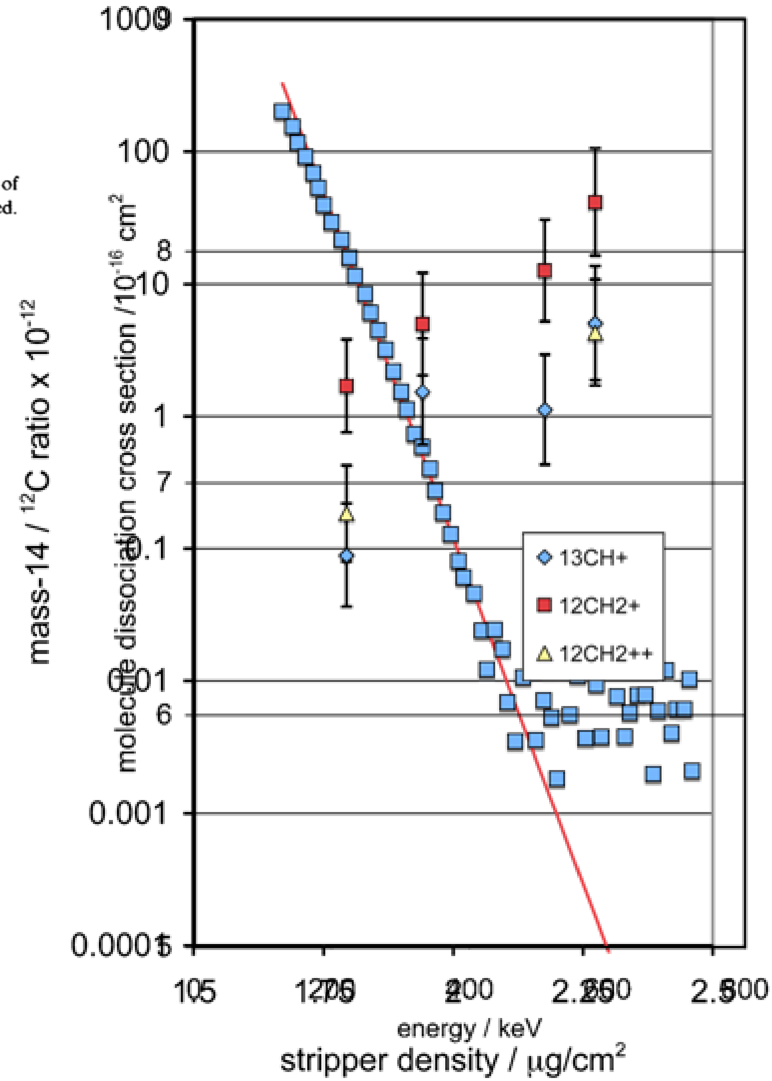
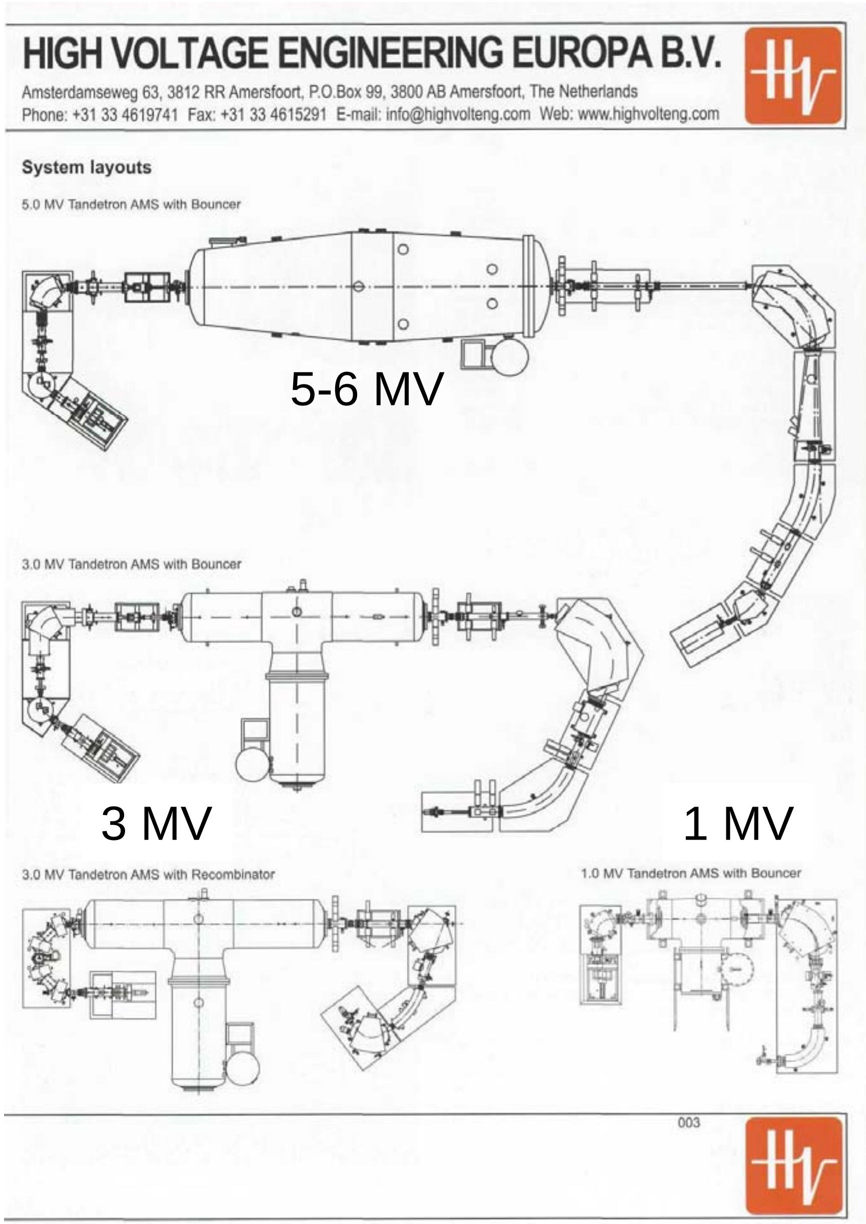


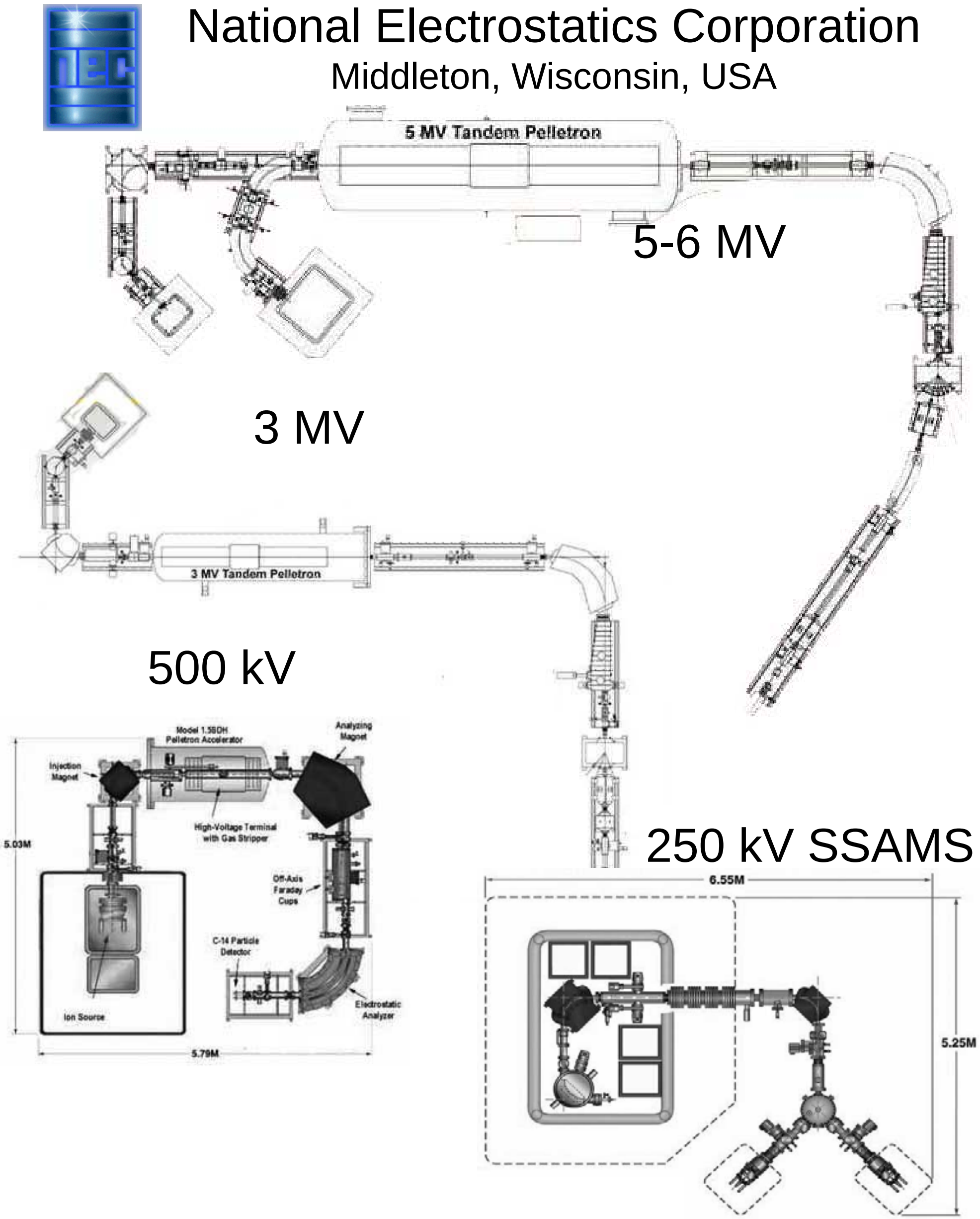
The first compact AMS system (1998) using charge state 1+ (ETH/PSI-NEC Collaboration)

Commercial systems are now on the market from NEC and HVEE

25 out of the 86 world-wide operating AMS spectrometers are based on this principle!

Commercial AMS systems

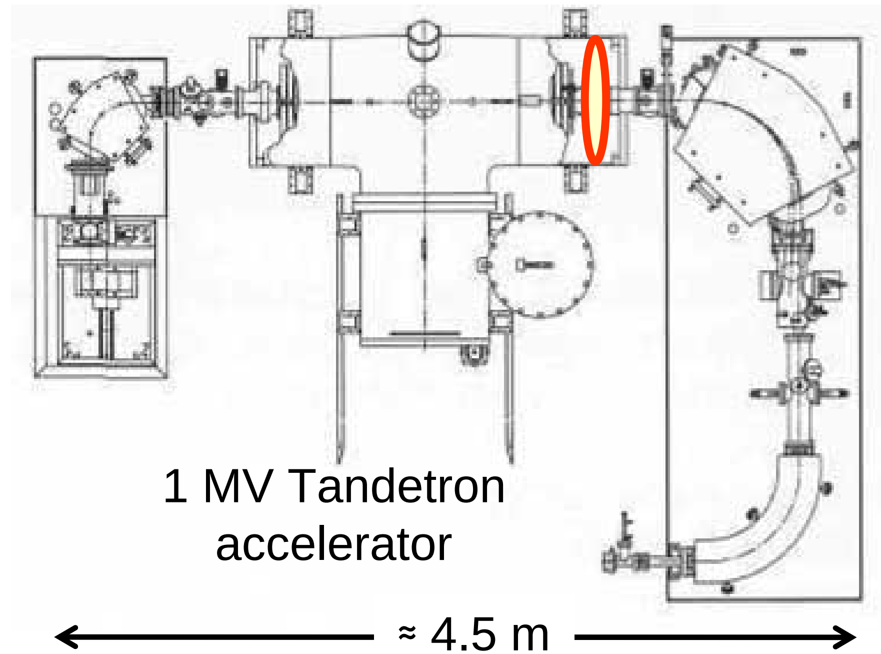




1 MV Multi-nuclide AMS system

Supplier: High Voltage Corp., Amersfoort, The Netherlands

AMS facility, at CNA, Seville, Spain



NEC-Single-Stage-AMS system

- National Electrostatics Corp. (USA)
  - Single-Stage-AMS system (2002)

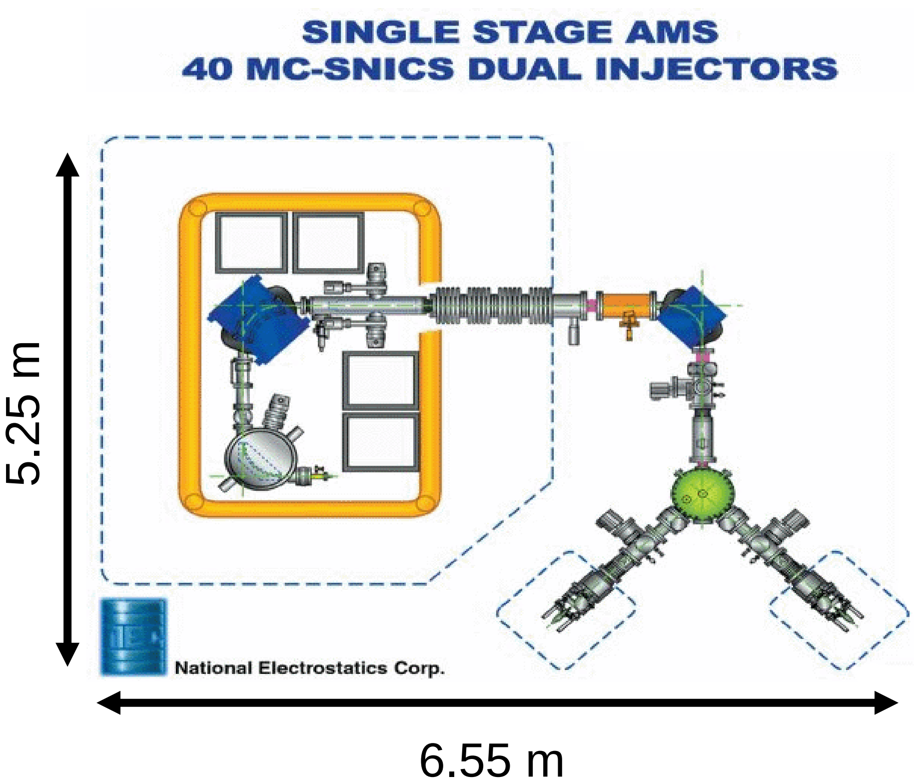
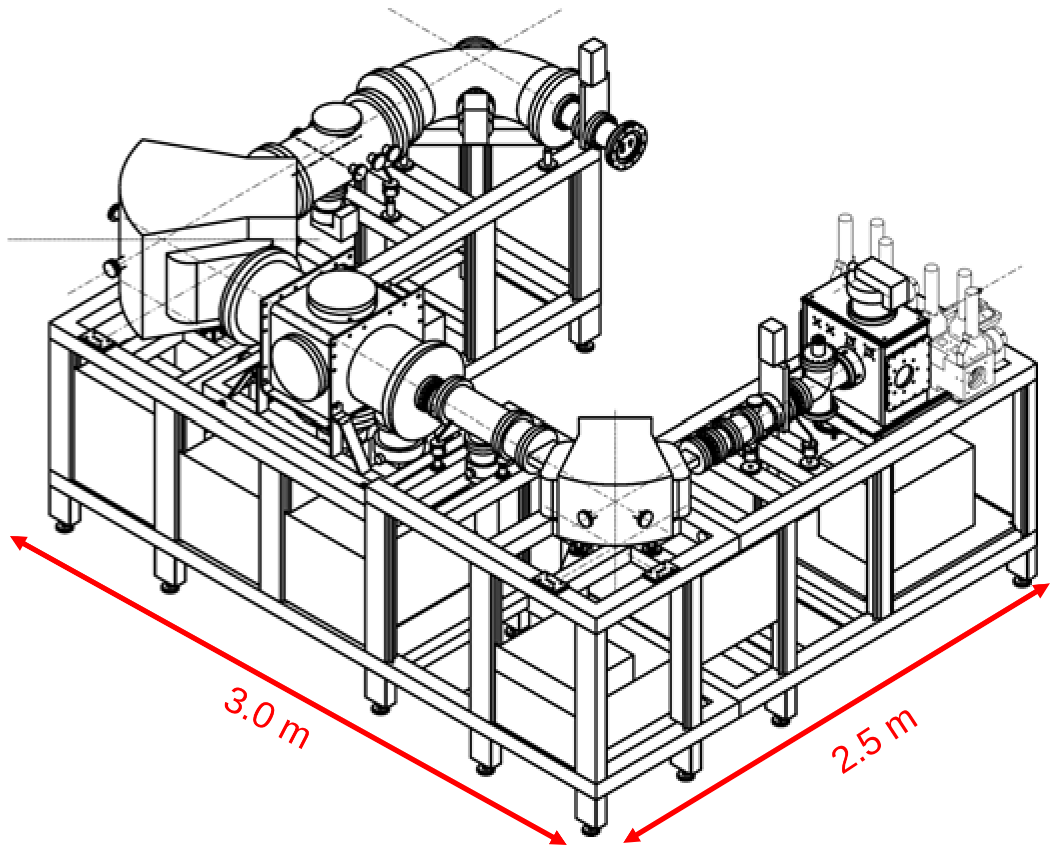


Figure of merit a of next generation AMS systems

Compact lab-sized instrument

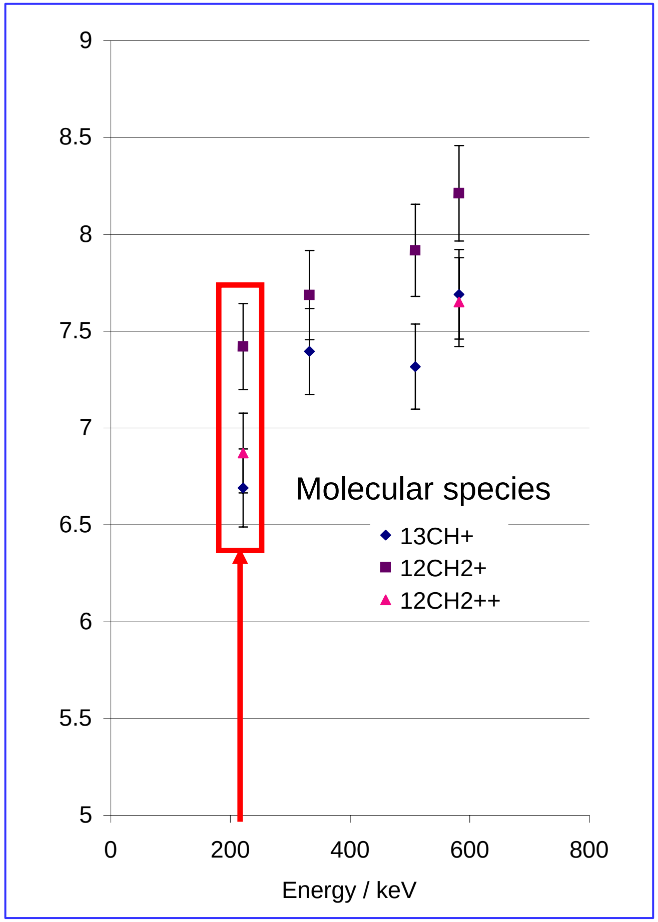
- *Designed for operator safety*
  - *No open high voltages*
    - *Easy to operate*
      - *Easy to tune*
    - *Fully automated*
  - *Web based monitoring*
- *Fail-safe sample handling*
  - *High precision*
  - *High throughput*
- *Reasonable investment costs*

Rather a "mass spectrometer" than an accelerator based system

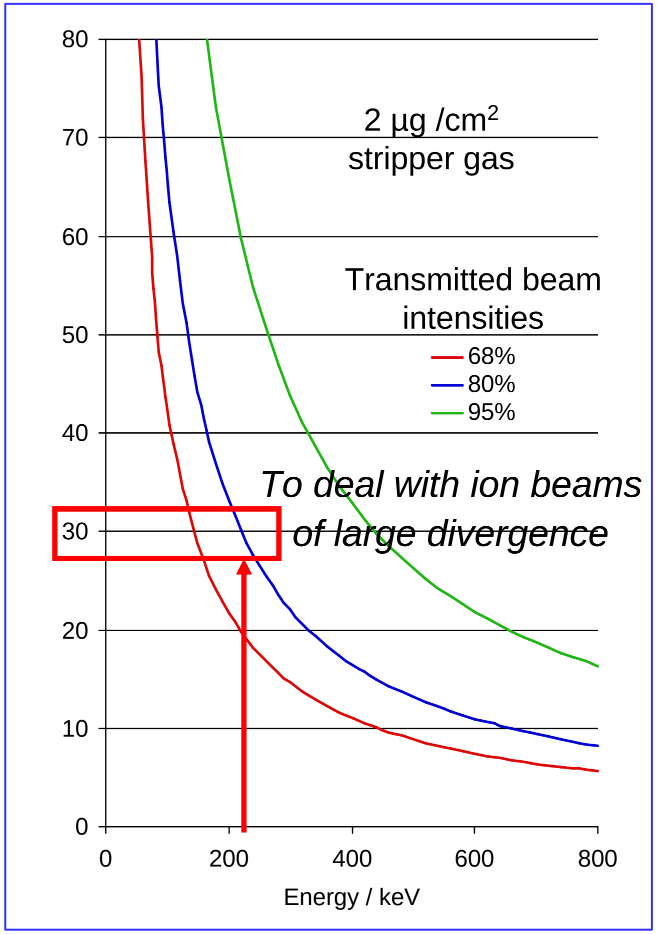


Low energy limits?

Cross sections of molecule destruction processes



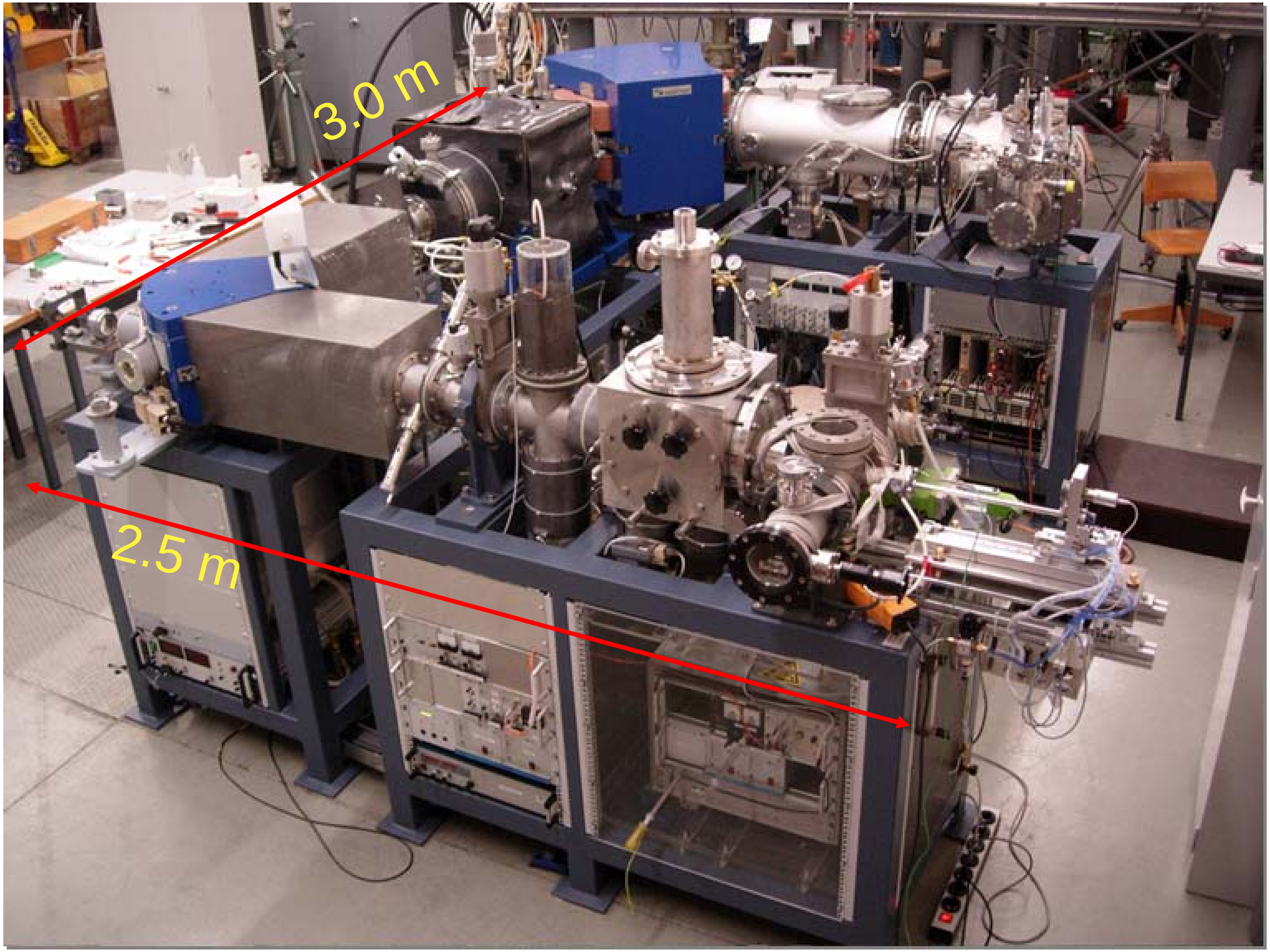
Energy dependence of angular straggling



- *Cross sections are comparable to molecular sizes*
- *Only weak energy dependence*
- *@ 230 keV cross sections are about 10 % lower*

New concepts can be applied at stripping energies below 250 keV!!

MIni CArbon DAting System: Prototype (2004)



Sputter ion source:
spherical ionizer
multi cathode
sample changer

Injection magnet:
fast beam switching
system

200 kV acceleration unit:
vacuum insulated
high voltage platform

High energy end:
achromatic
mass spectrometer

Detector:
high resolution
gas ionization
detector

Acceleration unit details

Stripper housing on insulator

200 kV corona gap
inside acceleration unit

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Routine Performance of ETH-MICADAS AMS system

Routine operation for all radiocarbon measurements at ETH since May 2008

- 3500 graphite targets analyzed
- 700 CO_2 sample (<50 μg)

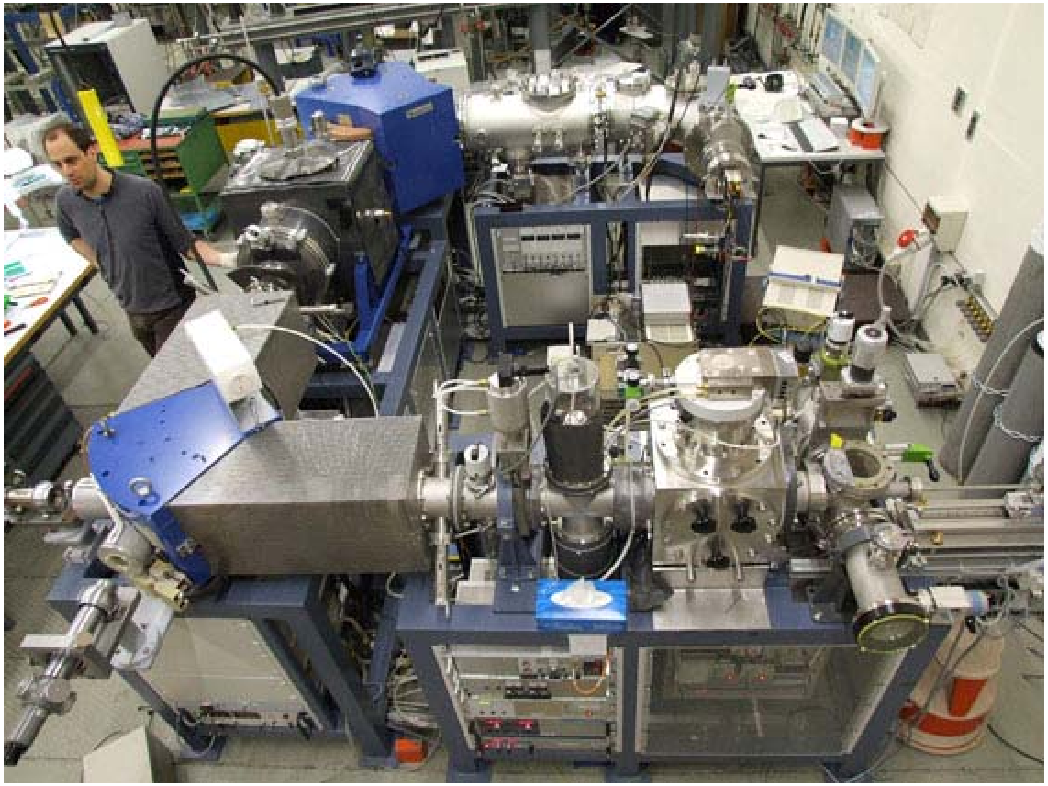
- runs unattended with low maintenance

- dates samples back to 45 000 years

- allows high-precision measurements

Blank variation:	< 0.5 ‰
Statistic uncertainty:	1.0 - 3.0 ‰
Reproducibility of samples:	0.5 - 3.0 ‰
Standard normalization:	0.5 - 1.5 ‰
Over all uncertainty:	**1.5 - 5.0 ‰**

ETH-MICADAS AMS system



PSI Paul Scherrer Institut

VITALEA SCIENCE

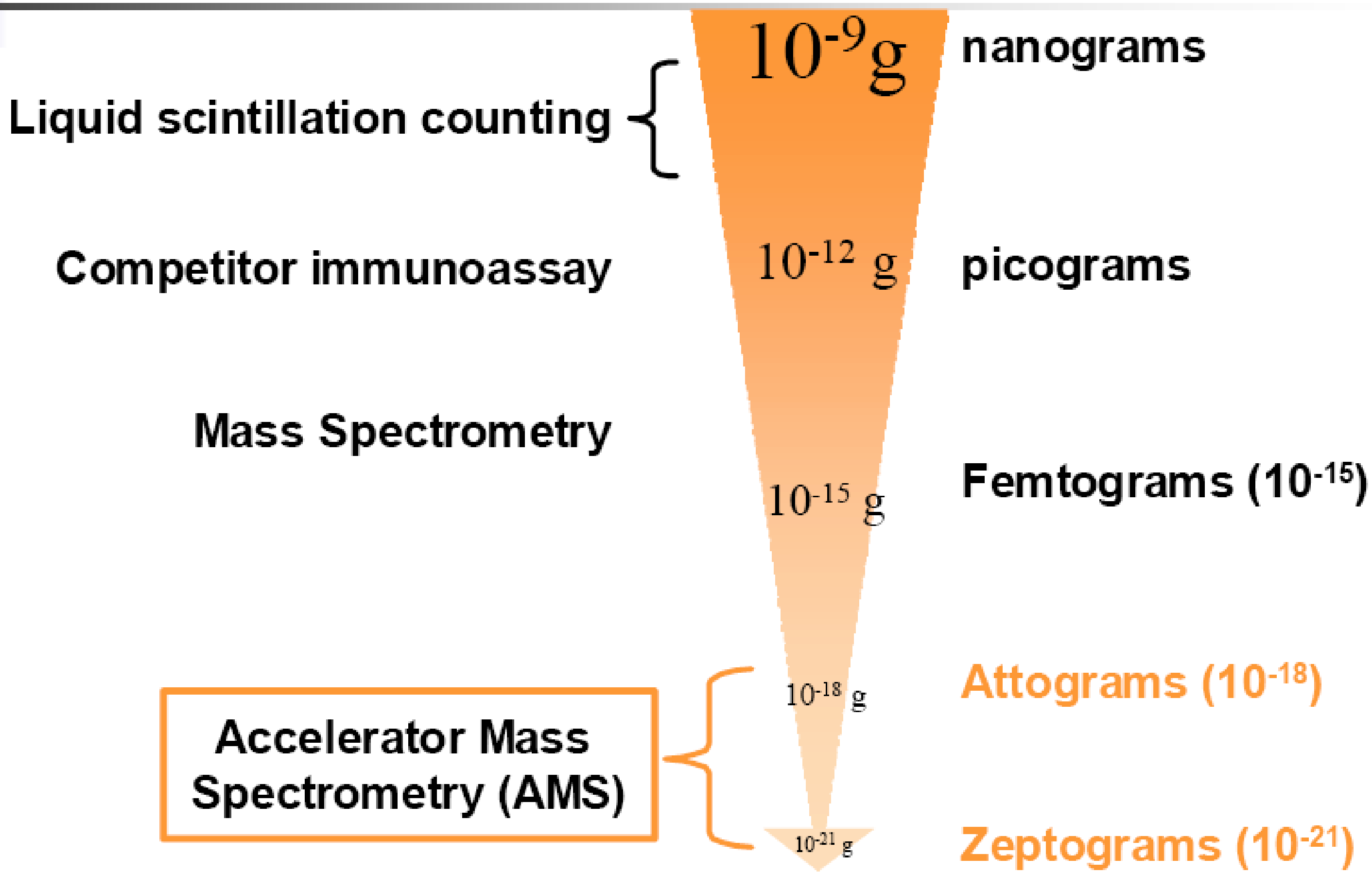
have entered into a collaborative agreement:

To build a novel instrument for biomedical applications using the most advanced AMS technology

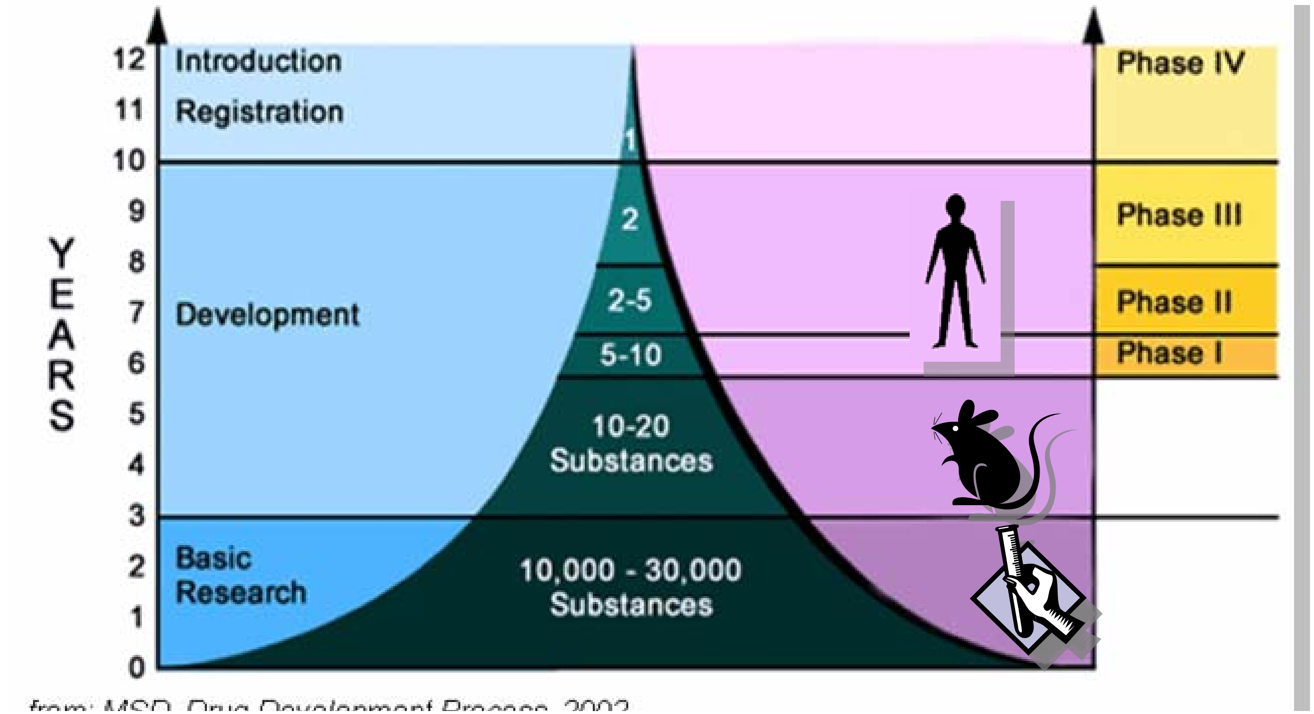
- PSI will provide expertise for design and construction of a BIO-MICADAS machine specifically optimized for biomedical AMS applications
- Vitalea Science will provide expertise and experience as a biomedical AMS service provider.

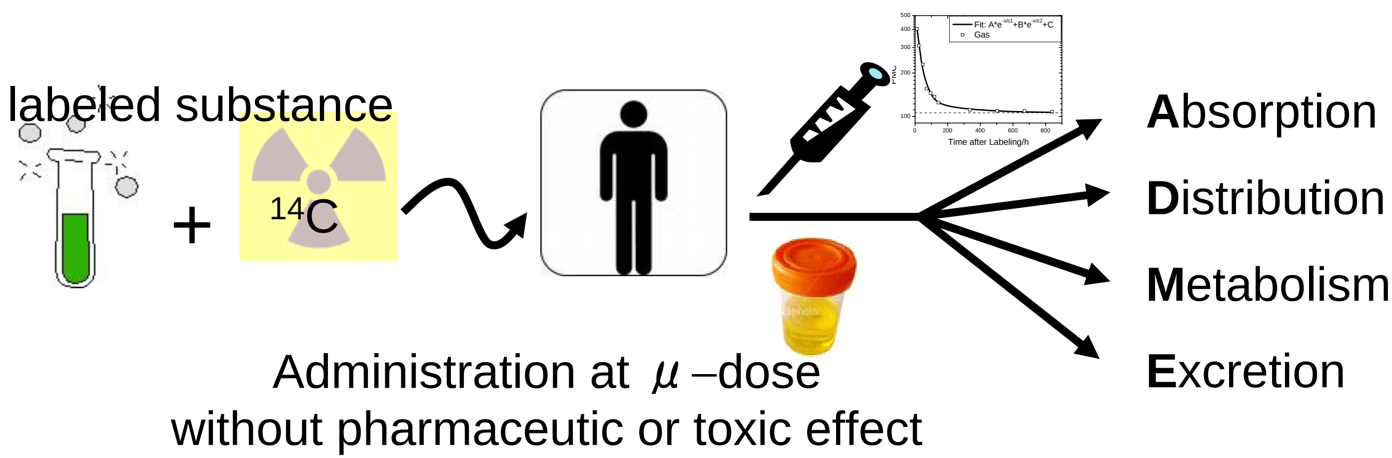
Why AMS for Bio Sciences?

Accelerator Mass Spectrometry (AMS): The Most Sensitive Detection Available

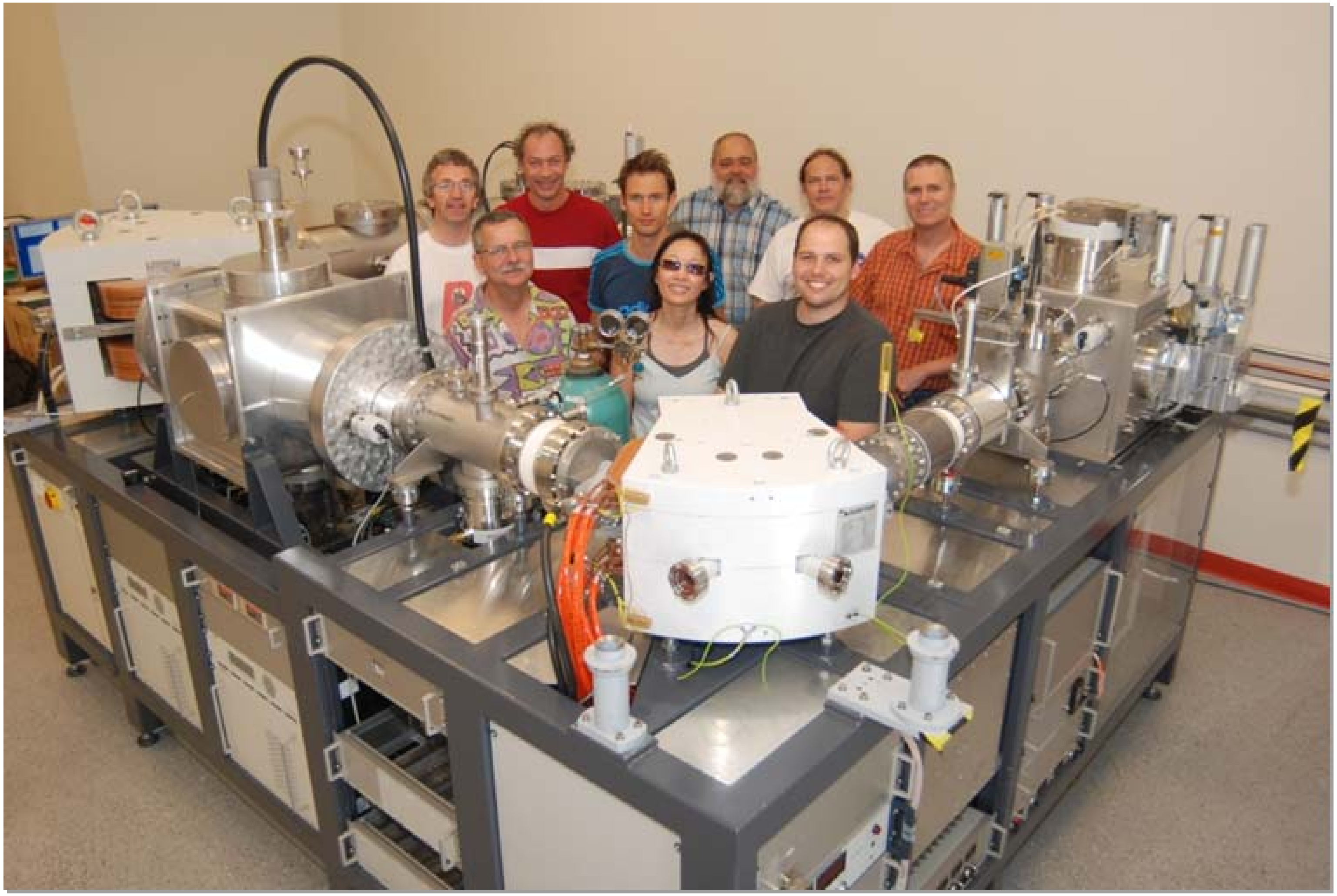


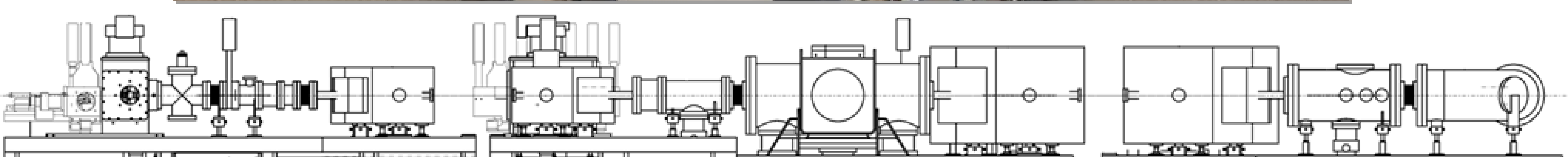
Micro dosing to accelerate drug development





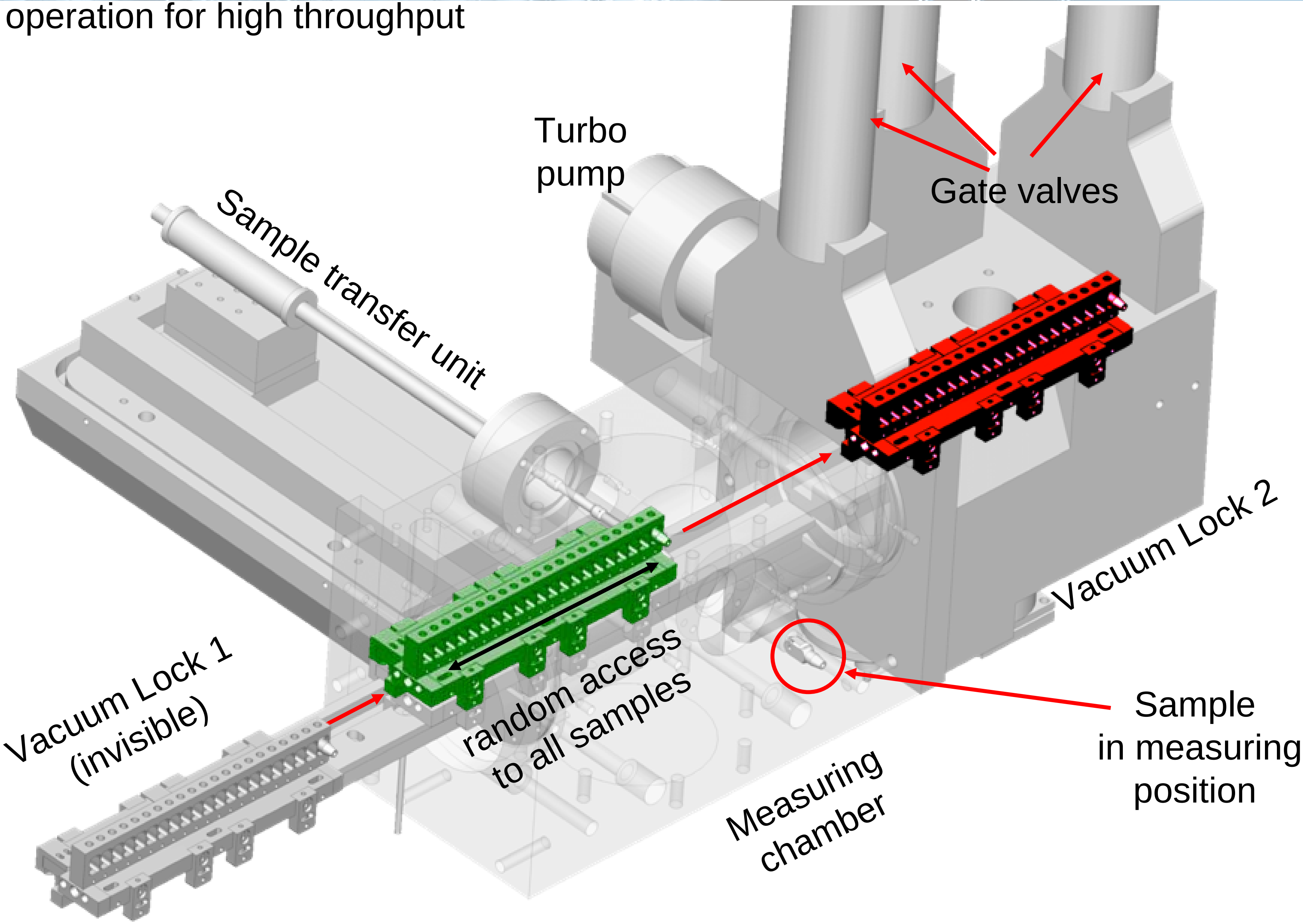
BioMICADAS: Ready to go



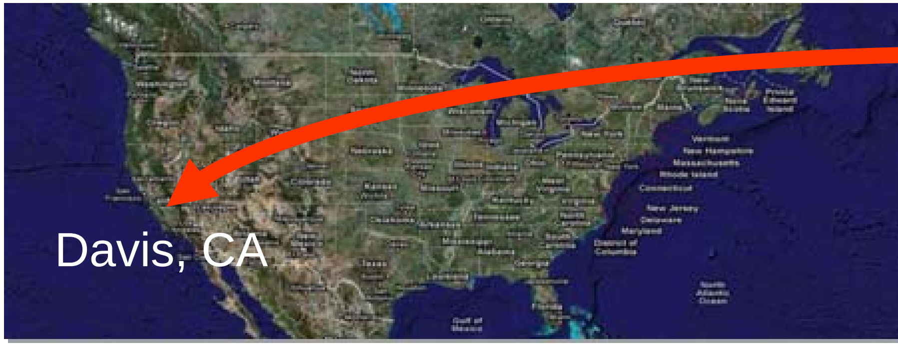


Dual load lock system

continuous operation for high throughput



Installation of BioMICADAS



Shipping:
6260 kg
with air freight

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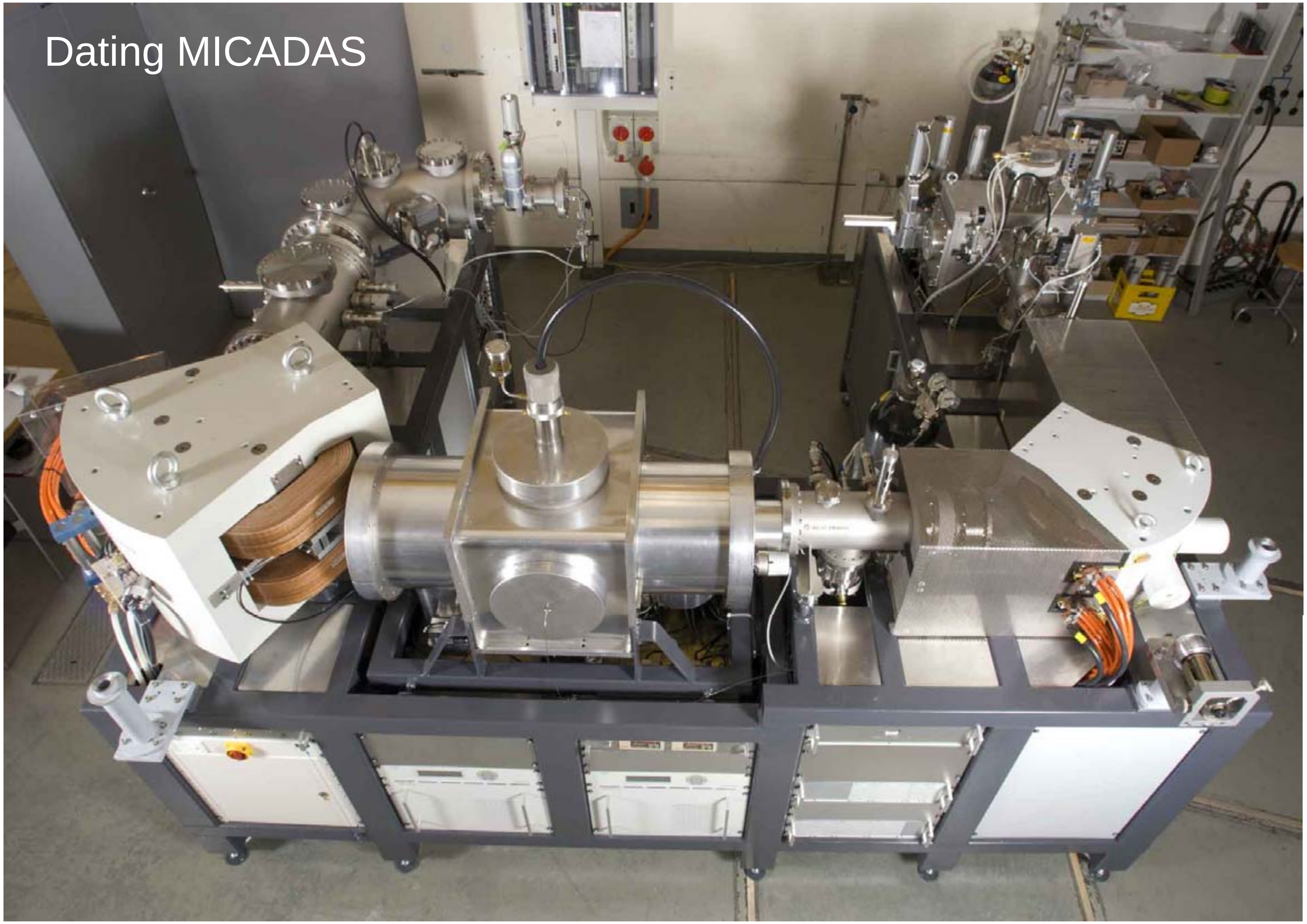
Laboratory of Ion Beam Physics

Characteristic / specifications (BioMICADAS)

	status
• Beam parameter	
– Typical beam ^{12}C currents:	≈ 40-80 μA
– Transmission:	41-42%
– ^{14}C counting rate for modern sample:	≈ 200 cps
• Measuring parameter	
– Typical measurement time per sample:	300 s
– Repeatability:	≈ 2 ‰
– Cover the full range of Biomed AMS	10^{-9}- 3 x 10^{-15}
• Operation parameter	
– Continuous measurements of samples	
– Throughput per running day (24h):	more than 200 samples / day
– 20’000 - 50’000 annual measurement capacity	
• Shipping the instrument to Vitalea, Davis USA	
– Shipping June 19-26, 2008	
– Installation and commissioning June 28 - July 8, 2008	
– Final acceptance tests July 17-21, 2008	

High precision radiocarbon dating



Developed and built at ETH

Operated in Mannheim (spring 2010)

Curt-Engelhorn-Centre for Archaeometry
associated with the University of Tübingen

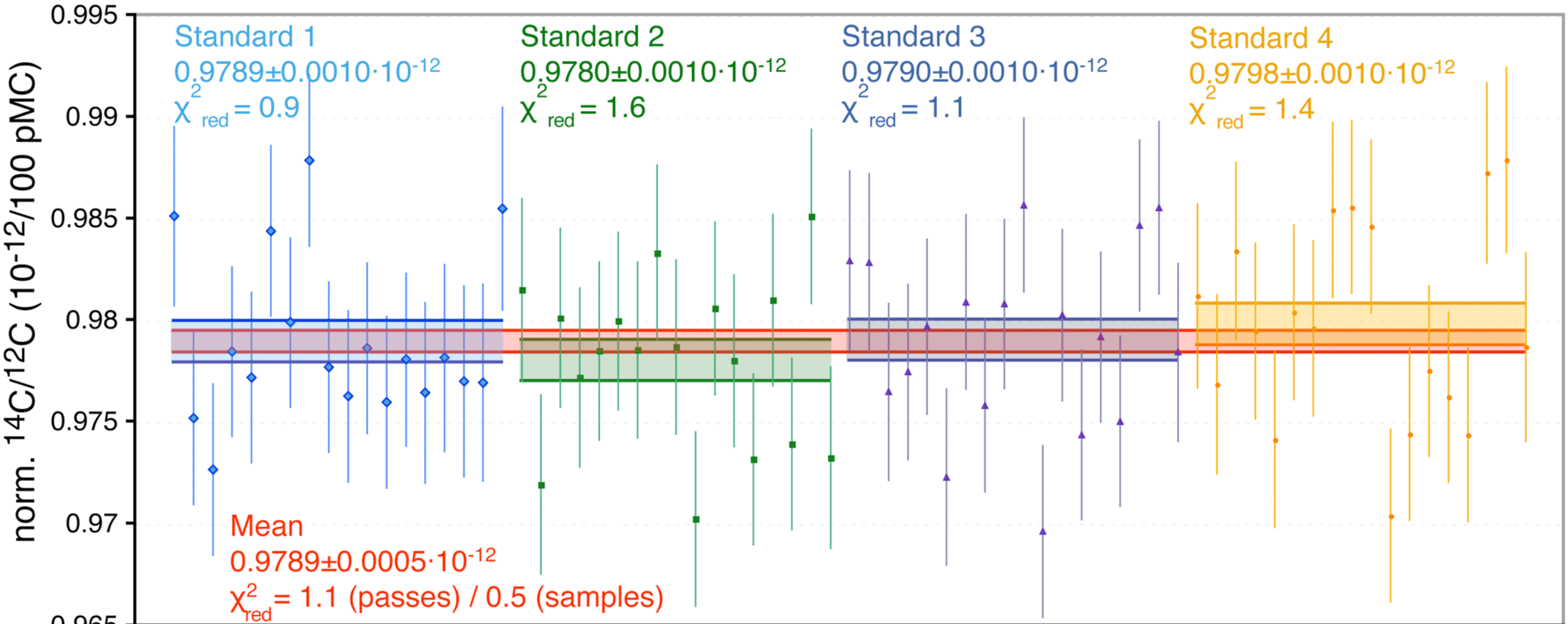
The latest generation of radiocarbon AMS systems
(MICADAS and DatingMICADAS)

- rather mass spectrometer than accelerator based systems
- improved radiocarbon measurement capabilities

What can we do with this?

Test of high precision capabilities

4 individually processed Oxa II standards during high precision measurements



Each standard was analysed 3 hours, acquiring a total counting statistic of ≈ 1‰

Sampling the historic document

Friedrich II
(Emperor 1194–1250

Berne: an Imperial City of the Holy Roman Empire of the German Nation

Signed: 15. April 1218
Aproved by
Rudolf v. Habsburg 1274

Cutting sample material from the parchment document

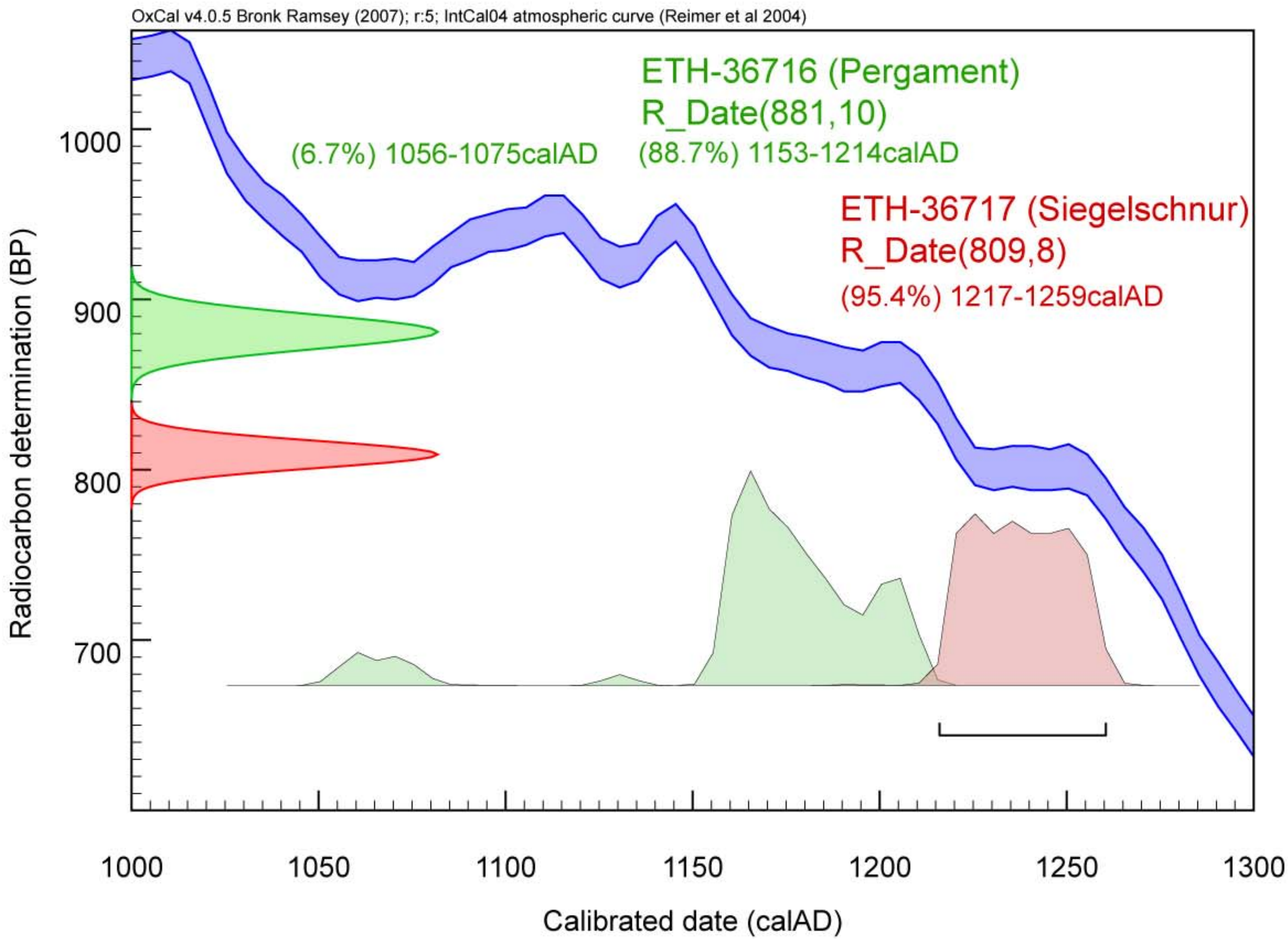
Taking fibers of the seal cord

"Goldene Handfeste“ results

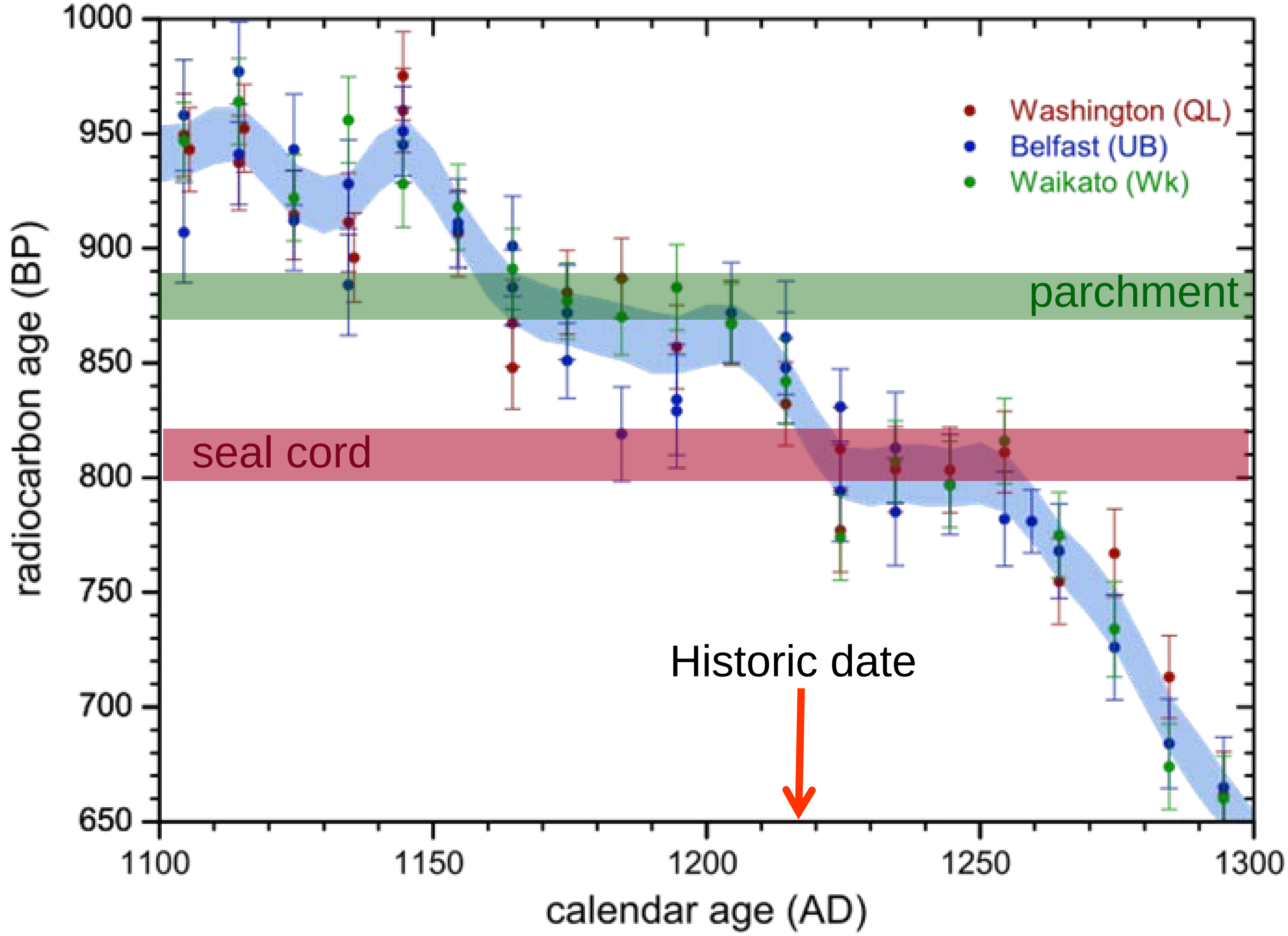
Sample number	Type	Radiocarbon age Years (BP)	$\delta\,^{13}C$ (‰)	Date of measurement
ETH-36716.1	Parchment	888 ± 20	-22.6 ± 1.1	18. Dec. 08
ETH-36716.2	Parchment	878 ± 19	-20.7 ± 1.1	18. Dec. 08
ETH-36716.3	Parchment	882 ± 19	-23.9 ± 1.1	27. Jan. 09
ETH-36716.4	Parchment	875 ± 19	-22.3 ± 1.1	27. Jan. 09
ETH-36716	**Parchment**	**881 ± 10**	**-22.4 ± 0.6**	**mean**

Sample number	Type	Radiocarbon age Years (BP)	$\delta\,^{13}C$ (‰)	Date of measurement
ETH-36717.1	Seal cord	800 ± 20	-24.0 ± 1.1	18. Dec. 08
ETH-36717.2	Seal cord	808 ± 19	-29.1 ± 1.1	27. Jan. 09
ETH-36717.3	Seal cord	833 ± 18	-25.5 ± 1.1	27. Jan. 09
ETH-36717.4	Seal cord	808 ± 18	-27.1 ± 1.1	27. Jan. 09
ETH-36717.5	Seal cord	800 ± 17	-27.7 ± 1.1	27. Jan. 09
ETH-36717	**Seal cord**	**809 ± 8**	**-26.7 ± 0.9**	**mean**

Absolute age calibration



Refining the calibration curve



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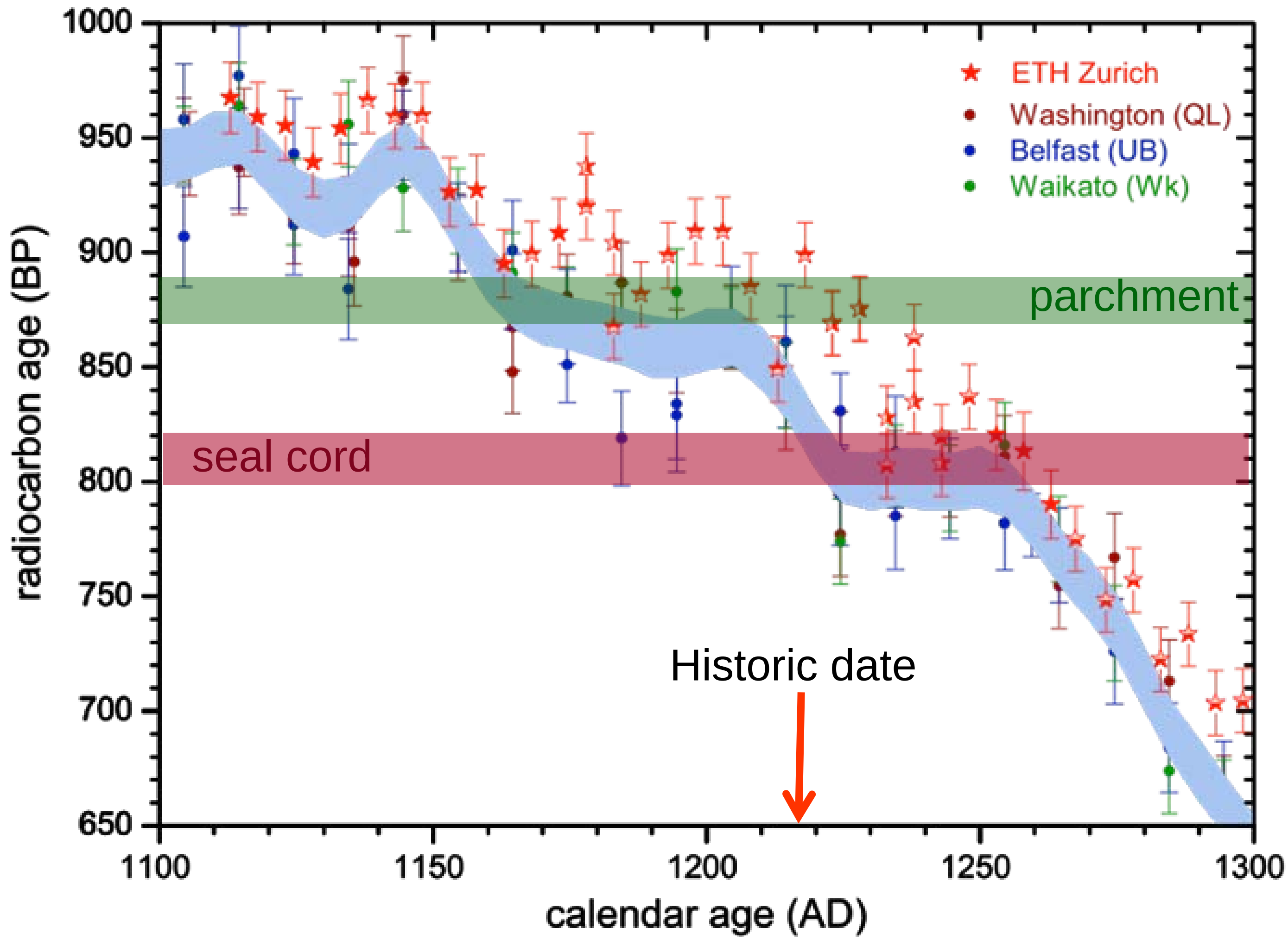


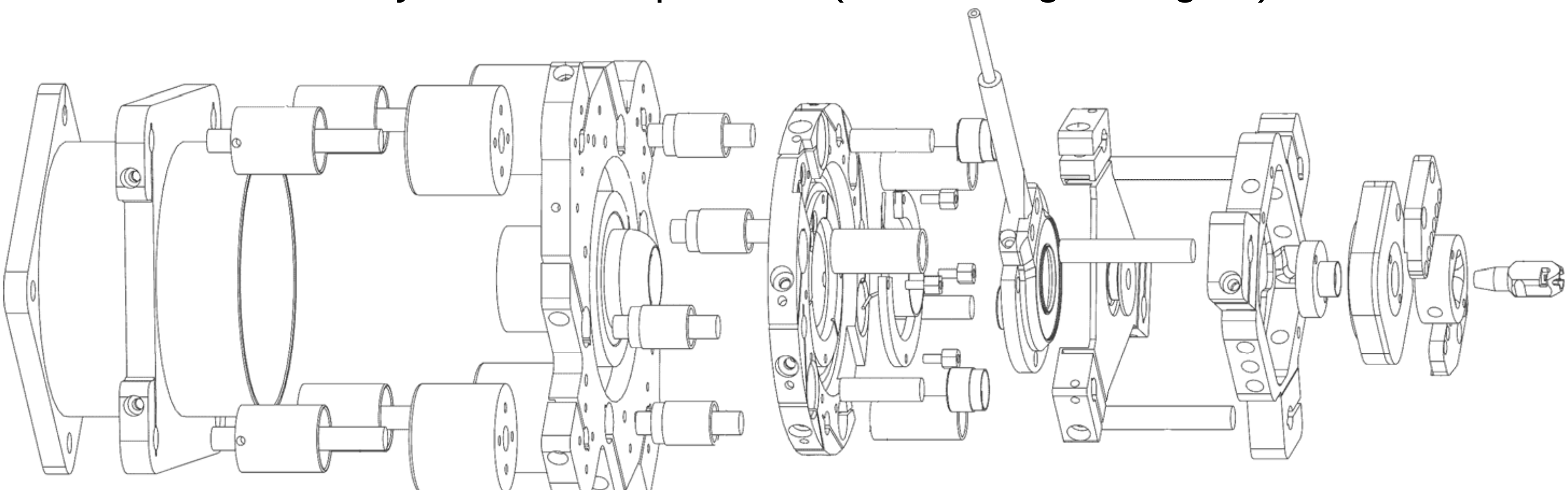
Ion source developments

Physical constrains:

- beam emittance
- ionization efficiency
- isotope fractionation

Practical constrains:

- reliability of operation
- sample handling
- maintenance and performance
- hybrid mode operation (solid and gas targets)

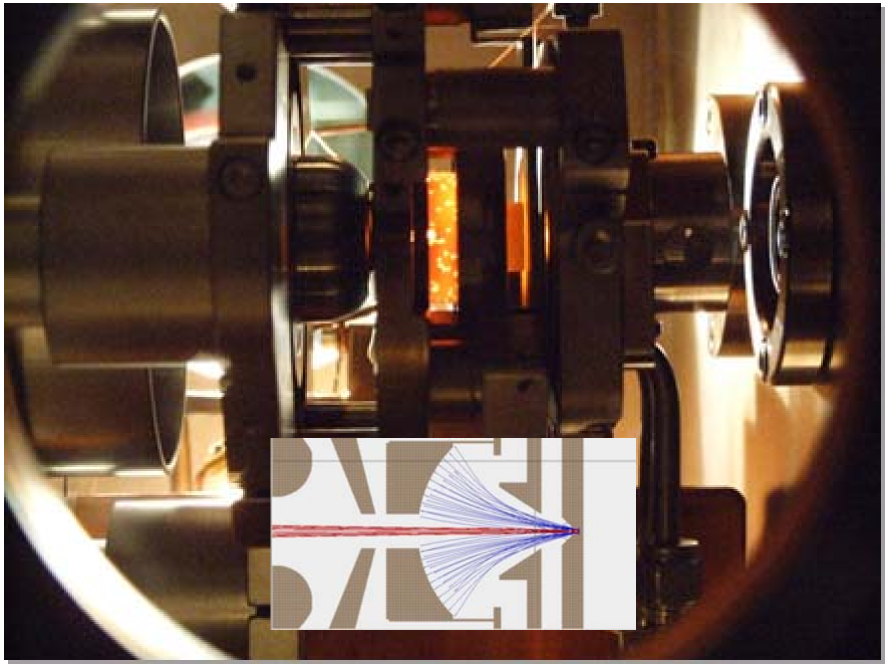


The ion source is the most critical element of an AMS system!

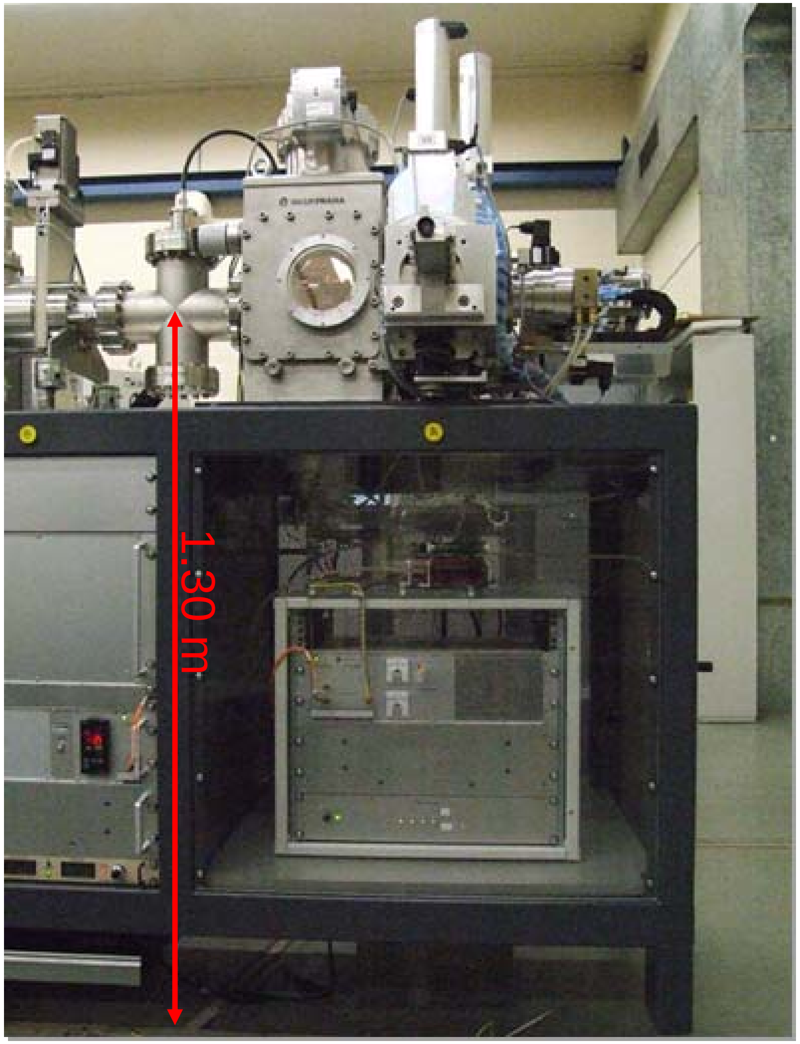
New ion source concepts

Primary design goals

- *No open high voltage potentials*
- *Precise/reproducible positioning of targets*
- *Good vacuum conditions*
- *Hybrid operation graphite / gaseous samples*
- *Continuous operation during exchange of magazines*



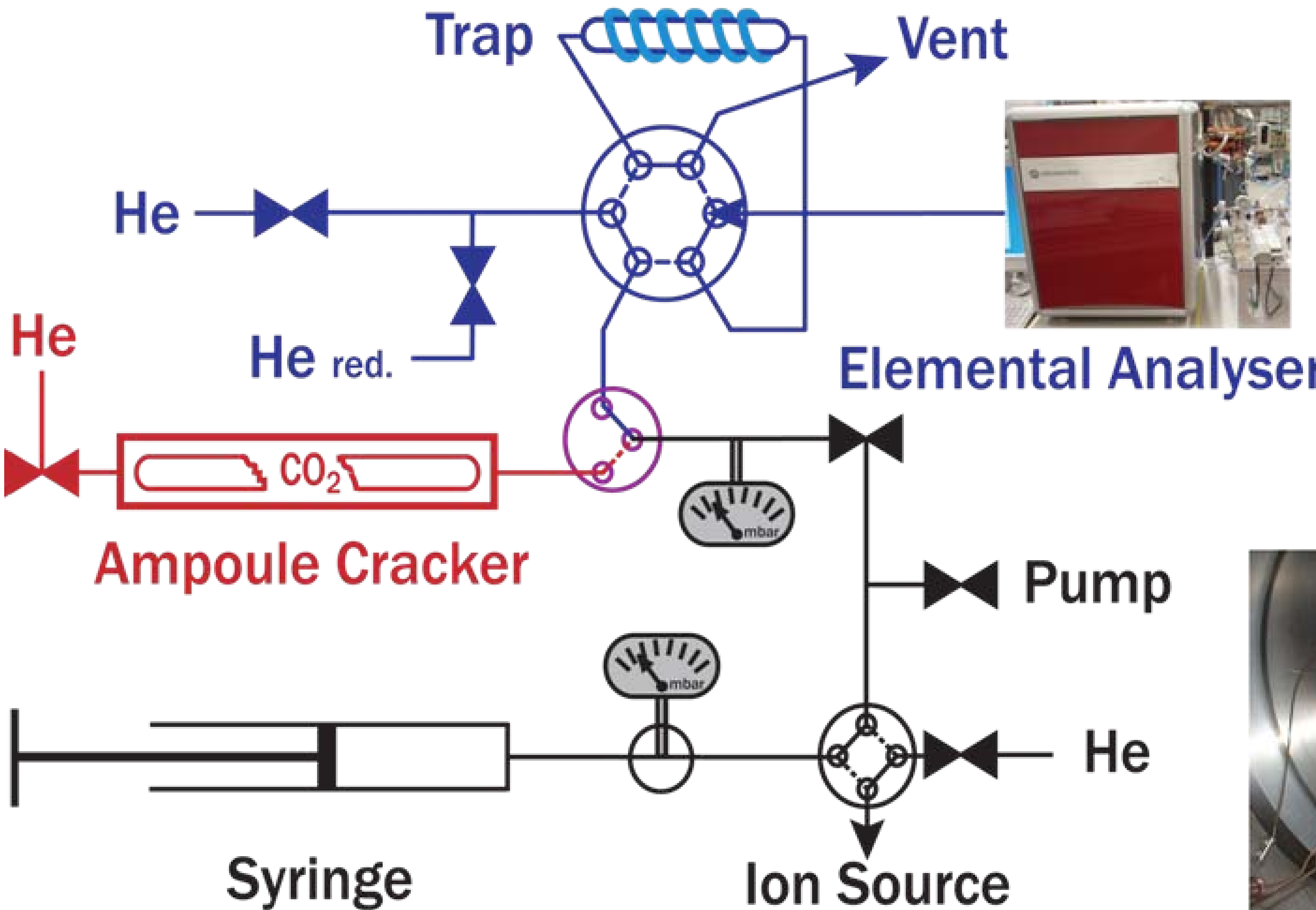
Ionizer assembly in operation: view through ion source window



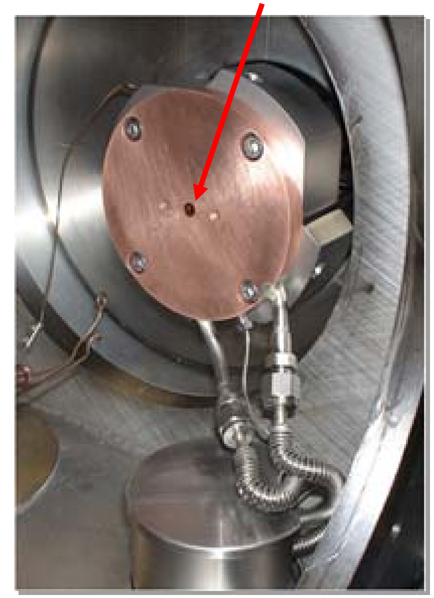
ETH ion source

Hybrid source for direct CO_2 analysis

Gas feed operation (Ultra small samples <50 μg)



Target position
In source



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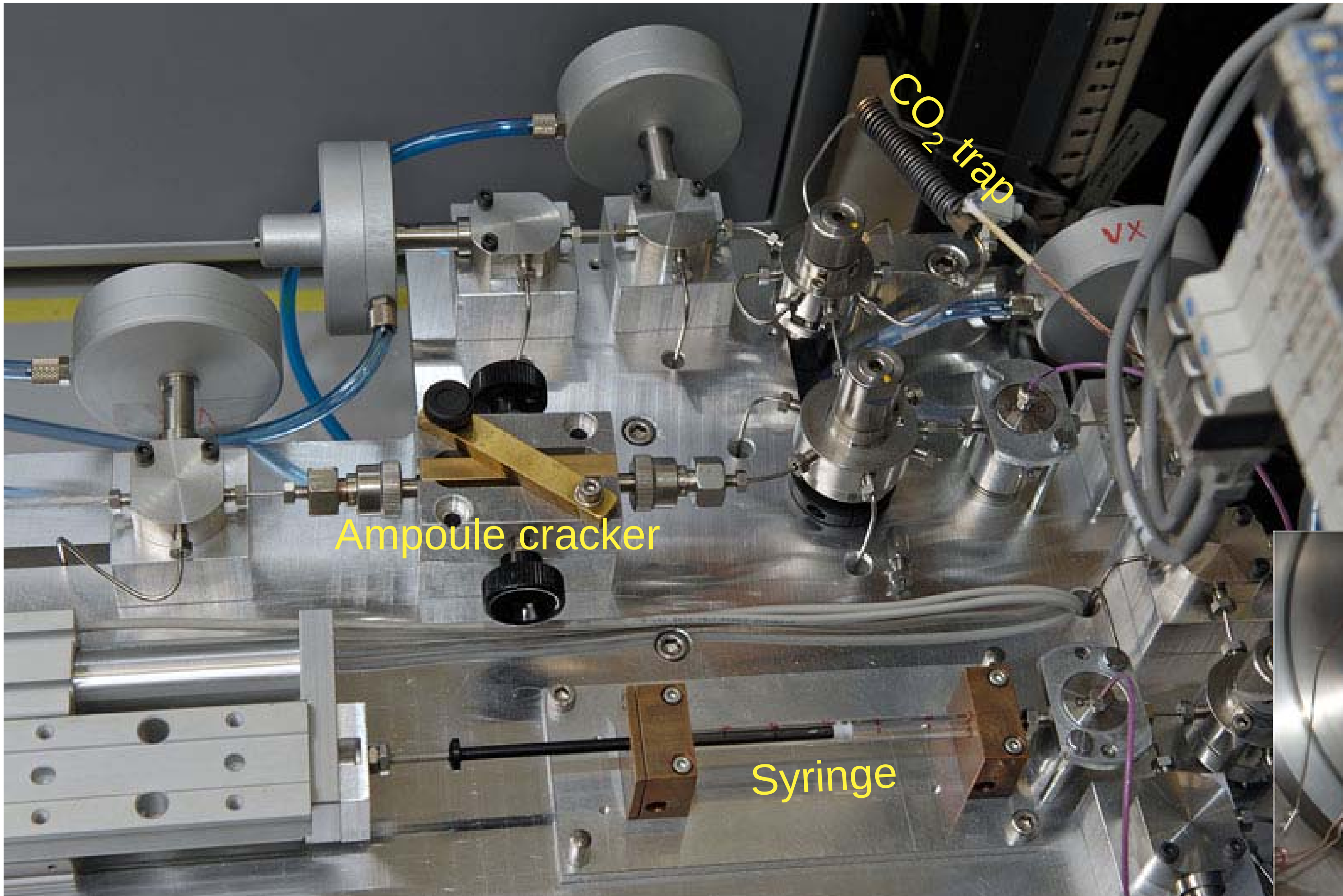
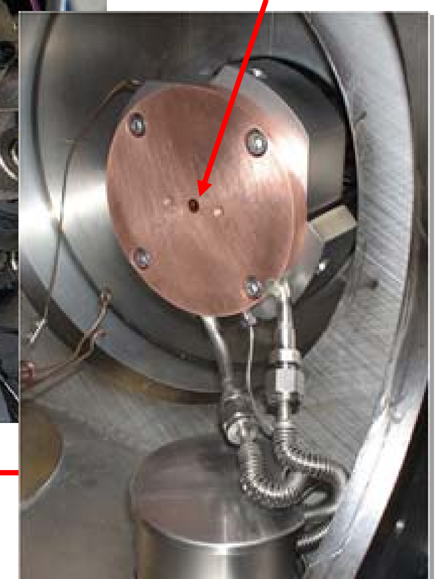
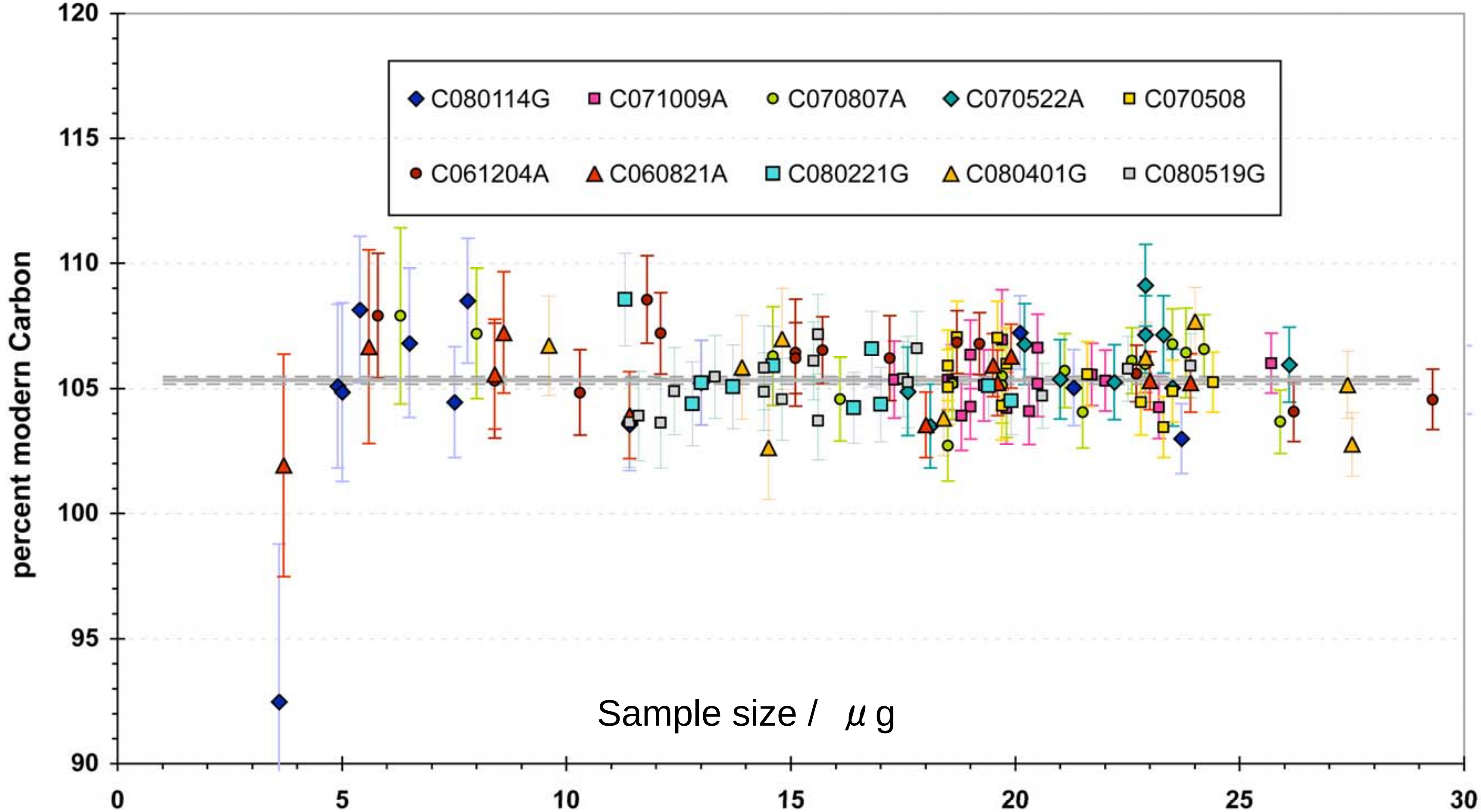


Target position
In source



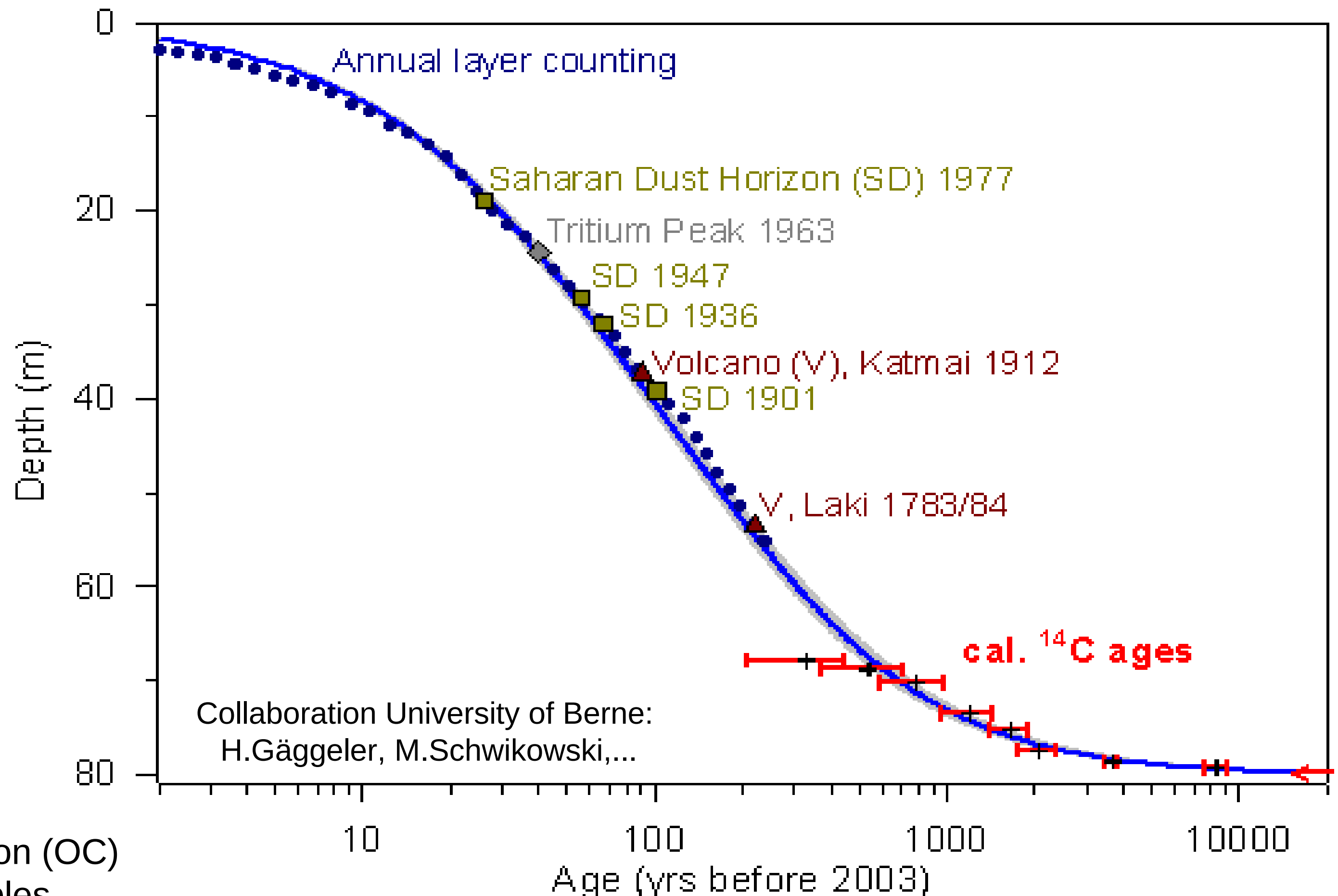
└capillary to ion source

Performance in routine operation



Measured samples (12 month):	700 direct CO_2
Limit of quantification:	35'000 yrs
Ion source currents:	3-5 μA
Typical uncertainty:	8-15 ‰
Sample size:	5-50 μg

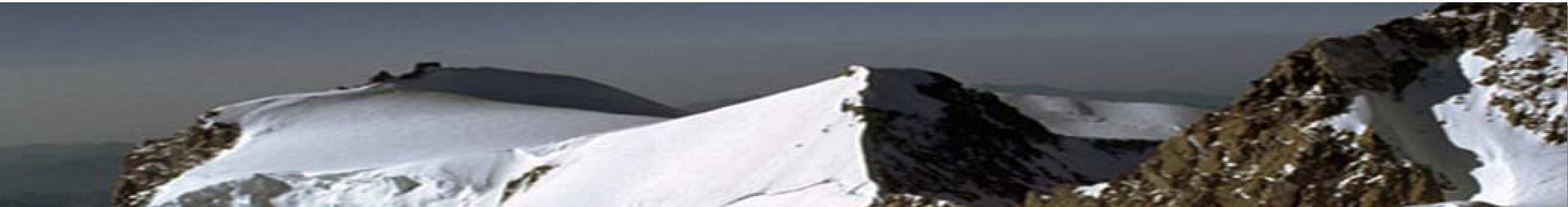
Dating Alpine ice cores (Collé Gnifetti)



Method:

Dating of organic carbon (OC)

- 5-20 μg samples



The other nuclides?

^{10}Be, ^{26}Al, ^{41}Ca, ^{129}I, Pu,..

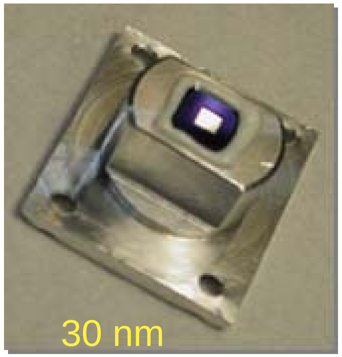
Has to be explored at very low beam energies.

Accelerators of about 500 kV,
are interesting alternatives!

^{36}Cl remains a "pièce de resistance"
But there might be alternatives at eV energies!

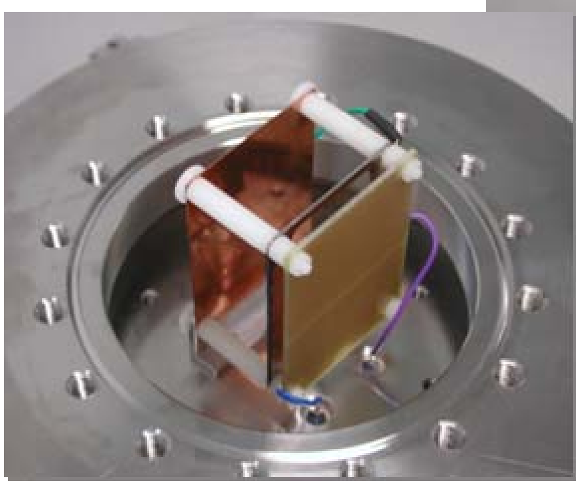
Revolution in low energy ion detection

Si_3N_4 detector window

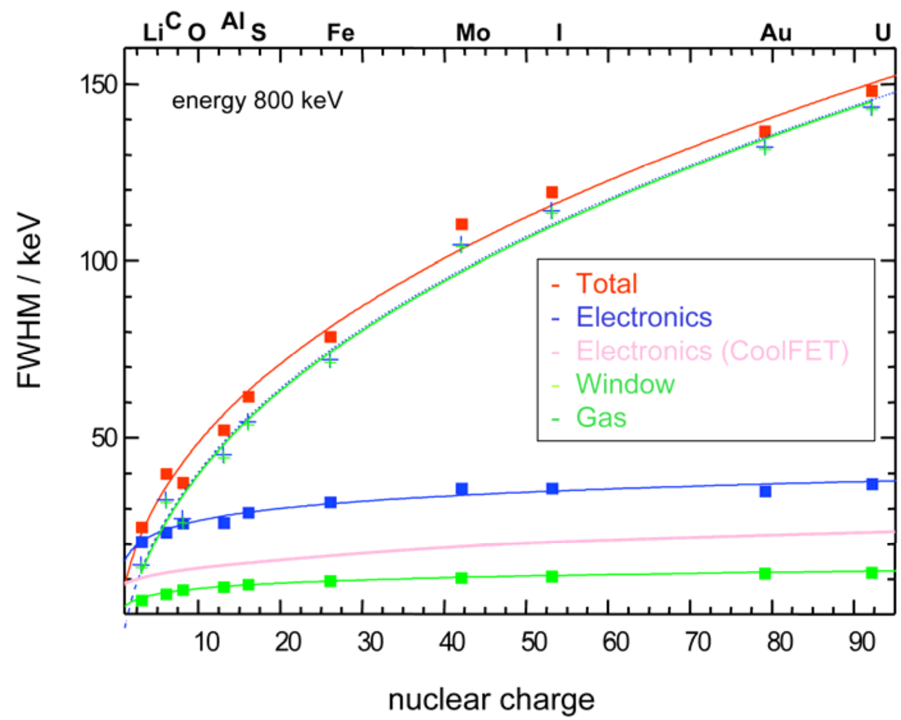


Detector with CoolFET pre-amplifiers

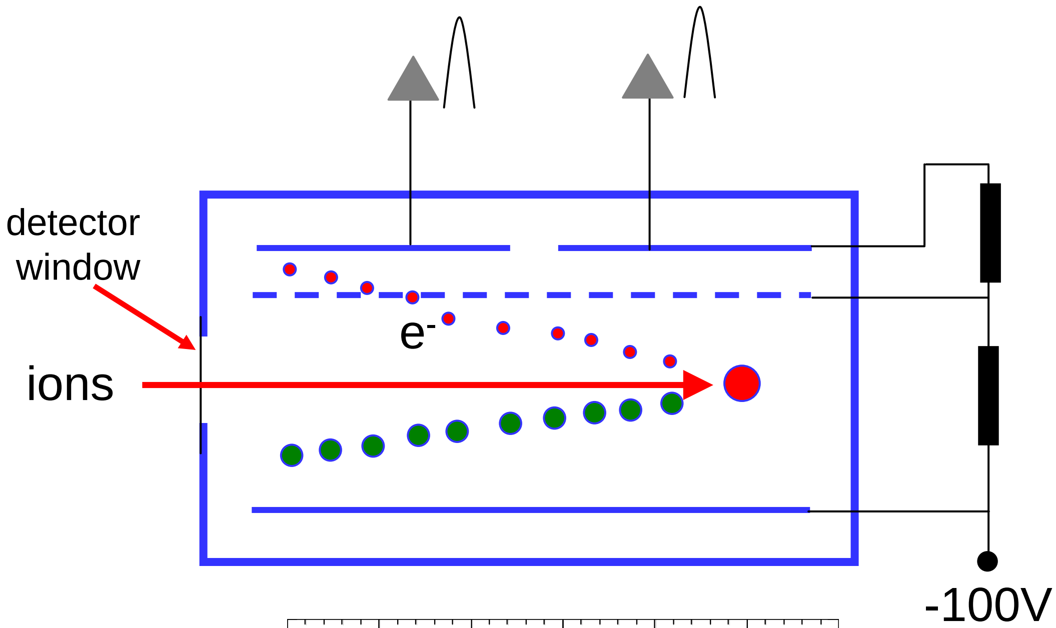
Anodes arrangement

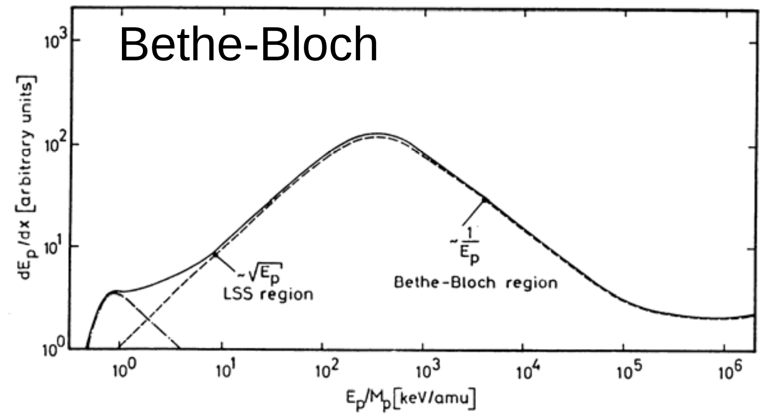


Resolution of total energy measurements

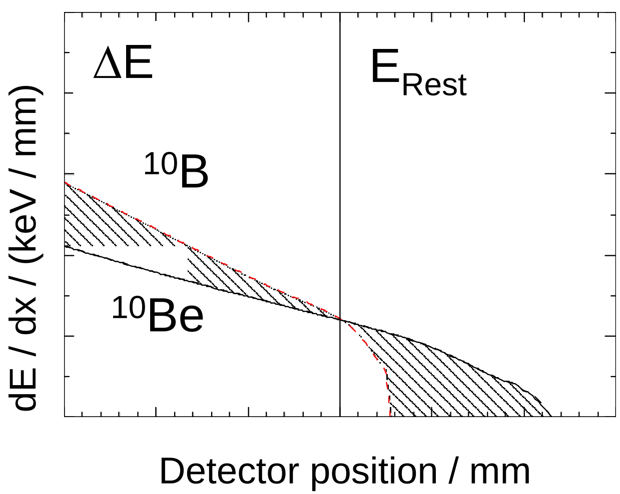


Gas ionization detector





- Ionization
- Charge collection
- Charge sensitive Pre-Amplifier
- Energy loss measurements



Nuclide separation: specific Stopping power characteristics

$$\delta = \int_{\Delta E + E_{rest}} \left| \left(\frac{dE_1}{dx}\right) - \left(\frac{dE_2}{dx}\right) \right| dx$$

Energy-loss straggling

Bohr:

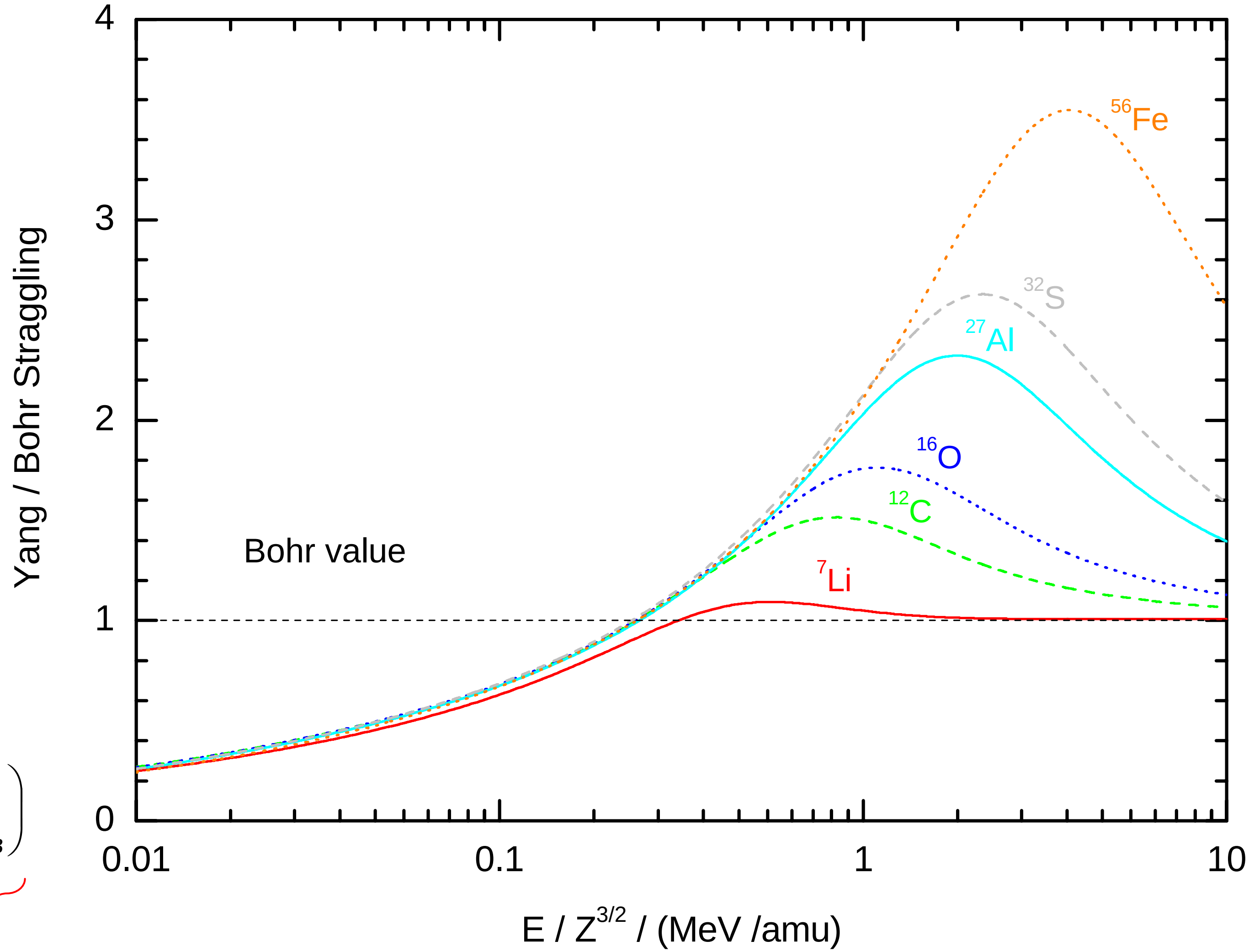
$$\Omega^2 \propto \cdot Z_1^2 \cdot Z_2 \cdot N$$

Chu/Yang:

$$\left(\Omega^2 / \Omega_B^2\right)_{Ion} = \gamma^2 \left(\Omega_{Chu}^2 / \Omega_B^2\right) + \left(\Delta\Omega^2 / \Omega_B^2\right)$$

Energy straggling semi empirical model form Chu, γ : mean charge of ion

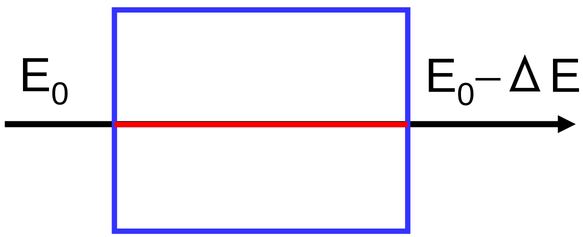
Charge state fluctuations $Z_1 > 2$



Focusing of energy straggling

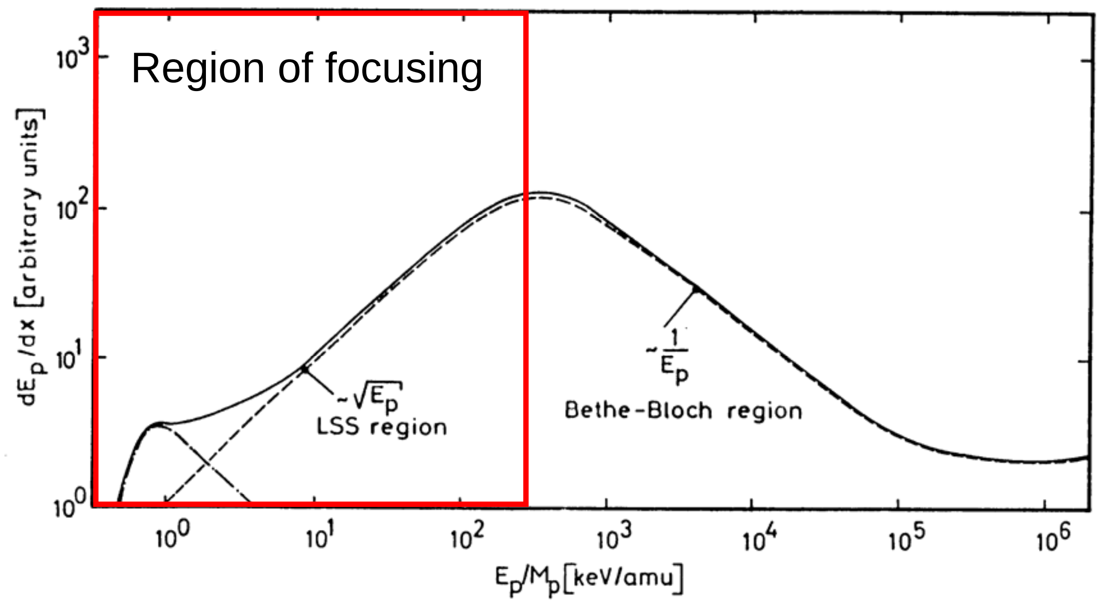
- Energy straggling is less than expected
- Pulse height defect helps for isobar suppression
- Focusing of straggling at left side of Bragg peak (Tschalär und Schmidt-Böcking)

Scaling for energy loss straggling



$$F_T = \frac{S(E_0 - \Delta E)}{S(E_0)}$$

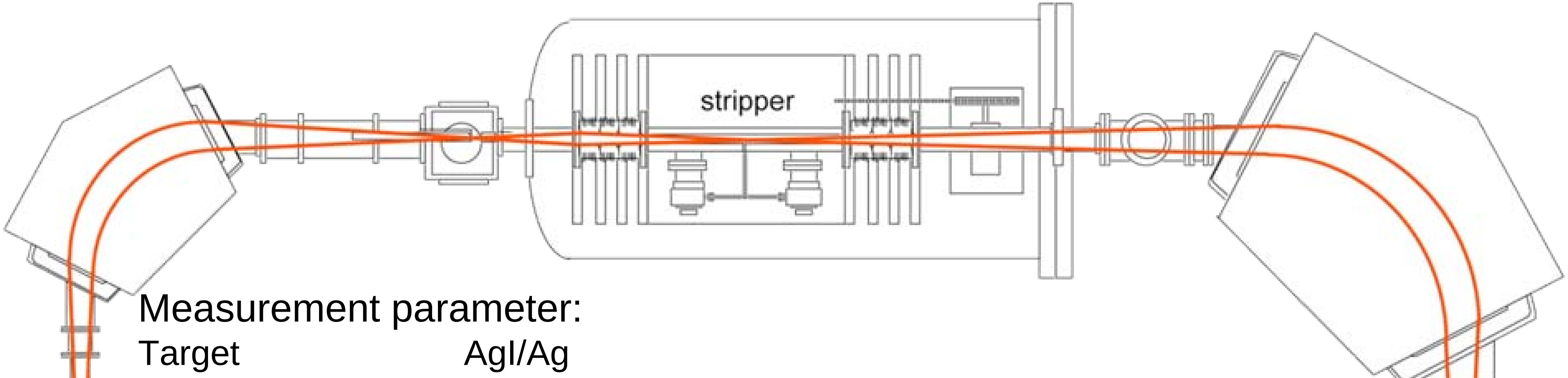
S(E) : Stopping power at energy E
E : initial ion energy
ΔE : energy loss



ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology

Laboratory of Ion Beam Physics

Routine ^{129}I analysis @ 2 MeV



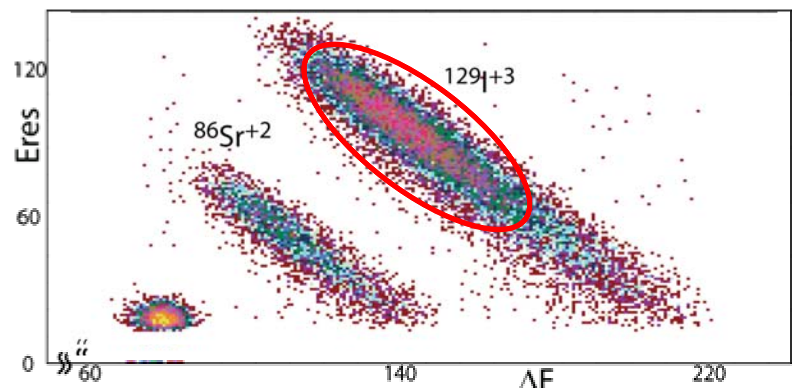
Measurement parameter:

Target	AgI/Ag
Current	5-10 μA
Charge state	3+
Stripper	Ar (0.22 $\mu g/cm^2$)
Terminal	0.5 MV -> 2 MeV
Transmisson	8%
Machine blank	$9 \cdot 10^{-14}$
$20 \cdot 10^{-12}$ sample	1.5% in 2 min

Detector parameter:

2 Anodes	3 cm each
Gas	Isobutane
SiN window	4x4 mm^2, 50 nm

Standard: $^{129}I/I \approx 10^{-11}$



≈ 700 ^{129}I samples analysed per year (unattended operation)

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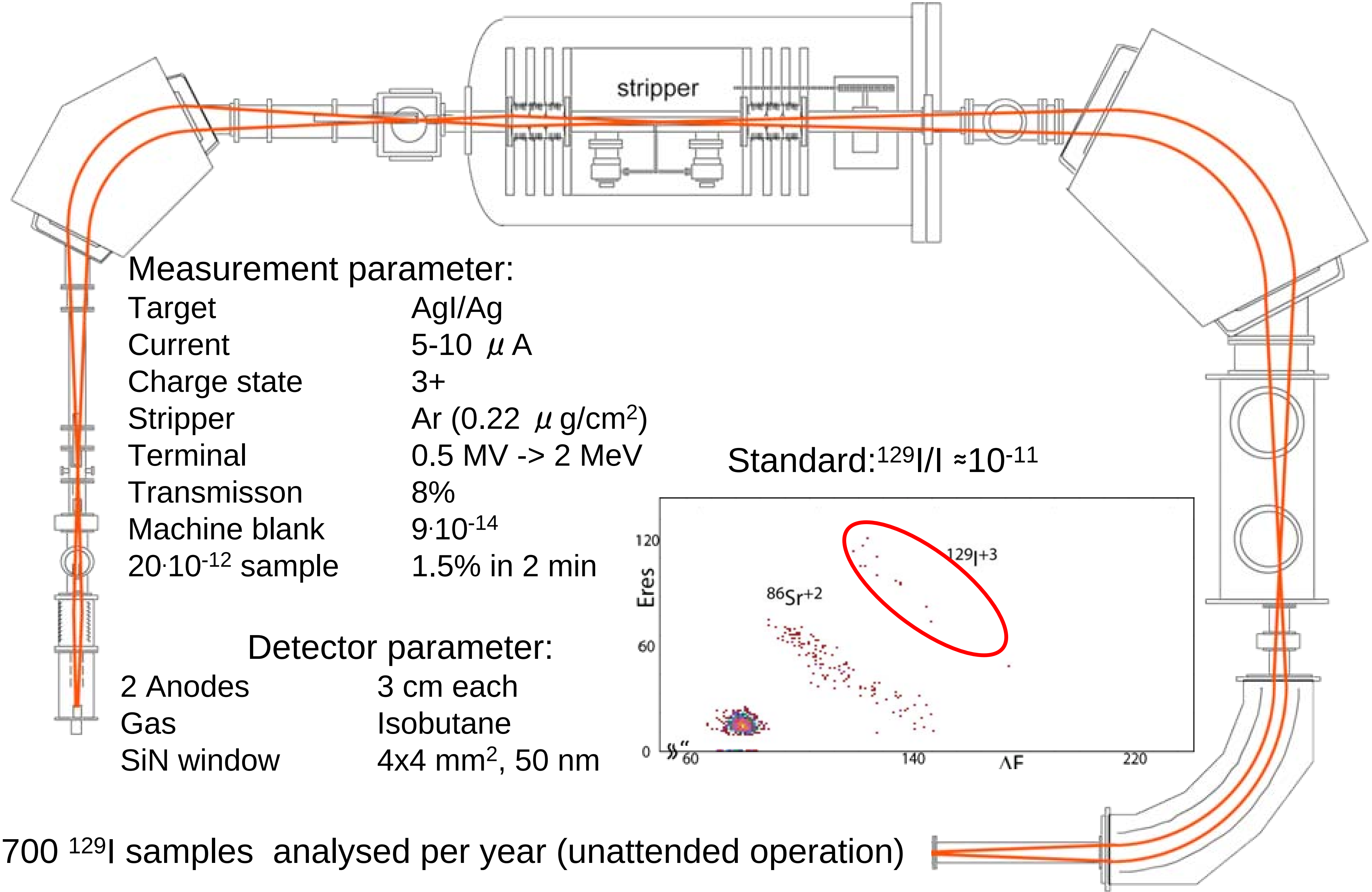

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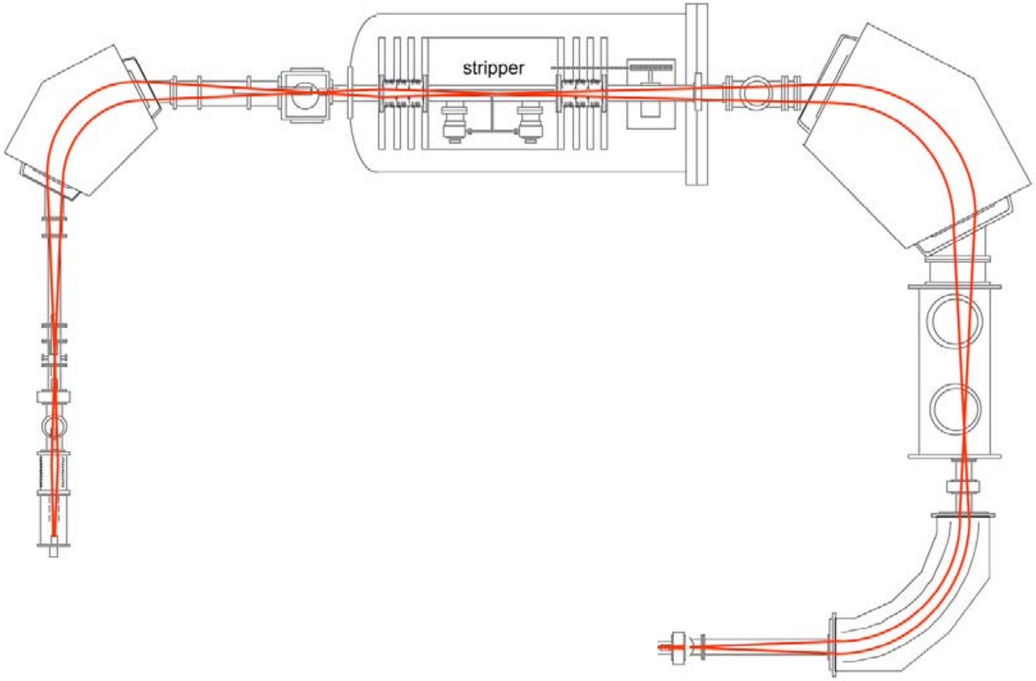
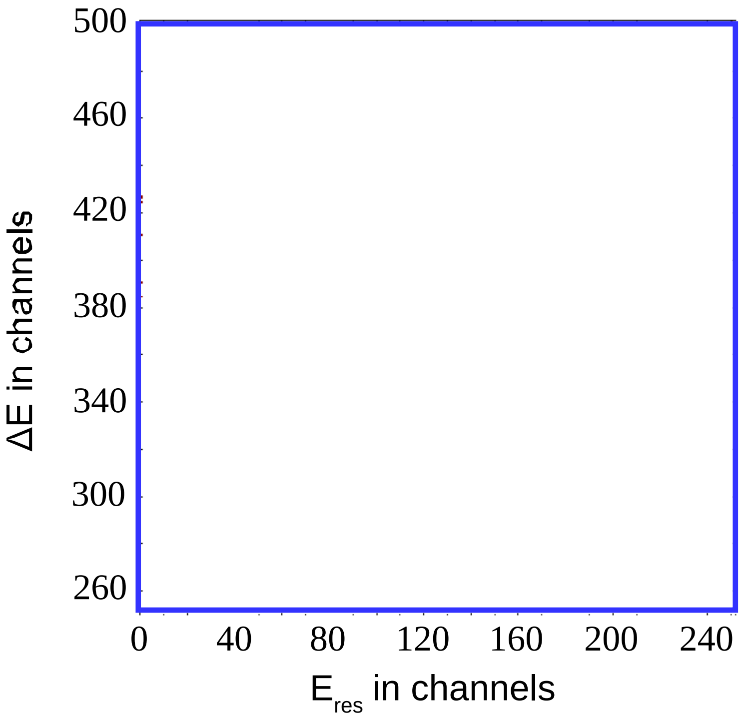
$^{10}BeO^{-}$ and SiN degrader

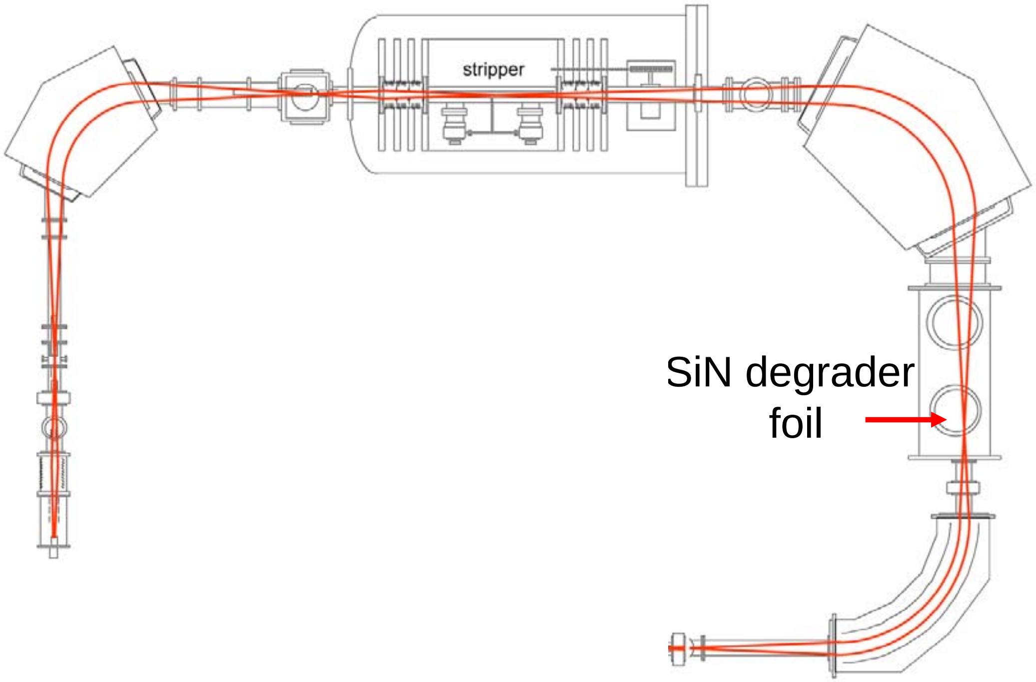
ETH- 600 kV Pelletron System

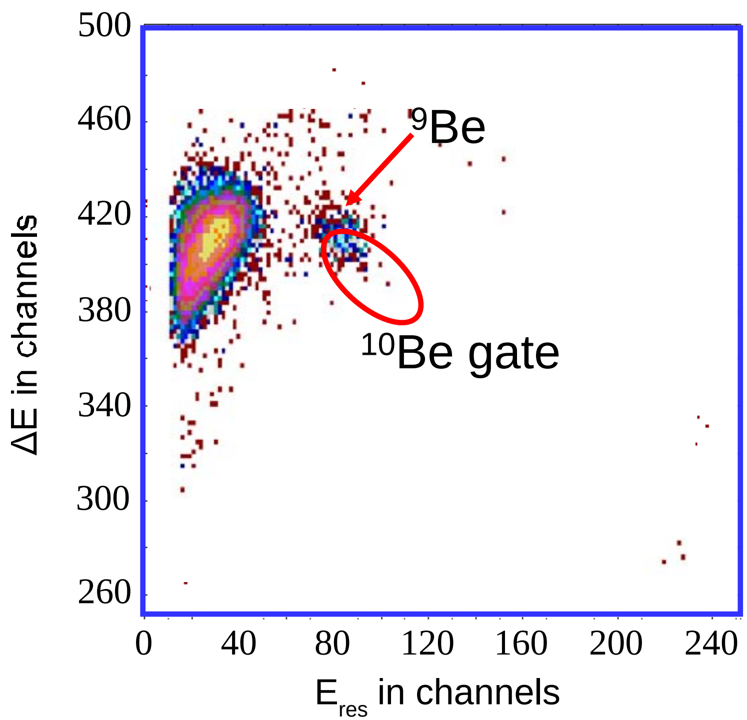




$^{10}BeO^-$ and SiN degrader

ETH- 600 kV Pelletron System





$^{10}BeO^-$ and SiN degrader

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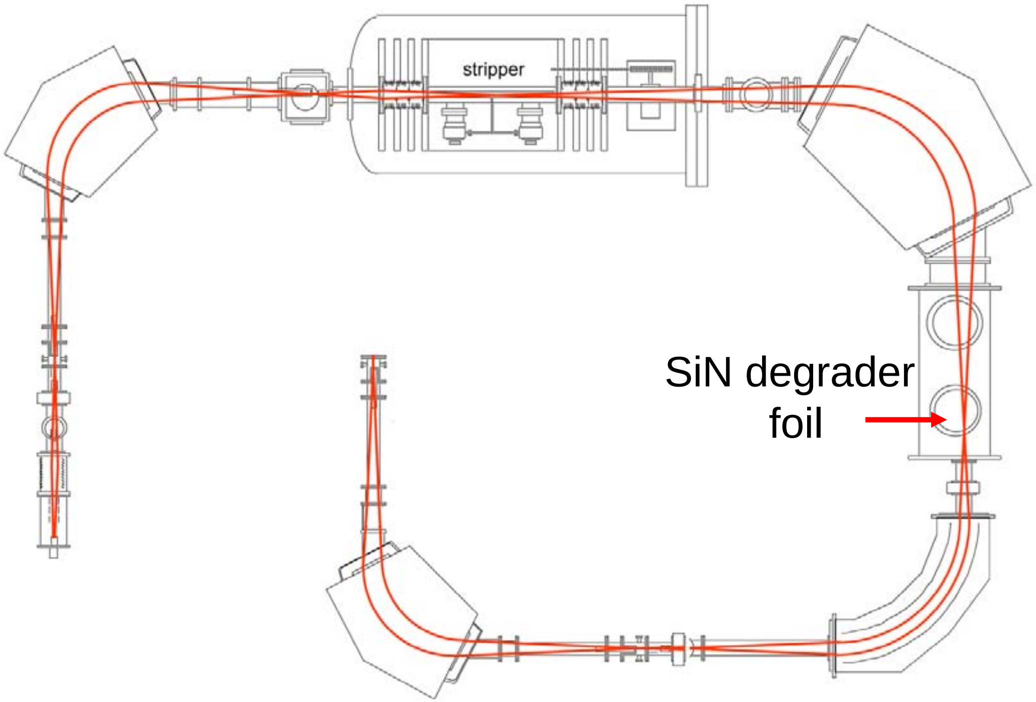

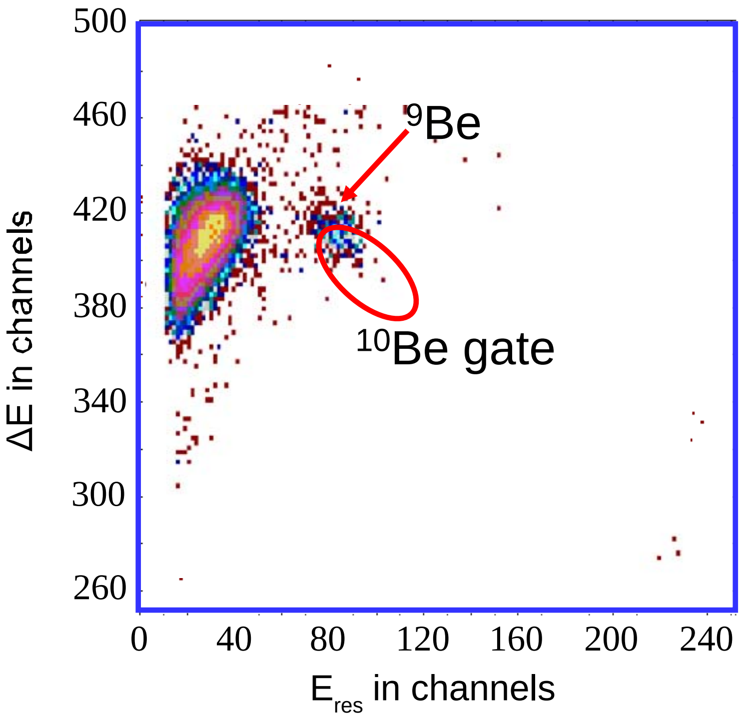

$^{10}BeO^{-}$ and SiN degrader

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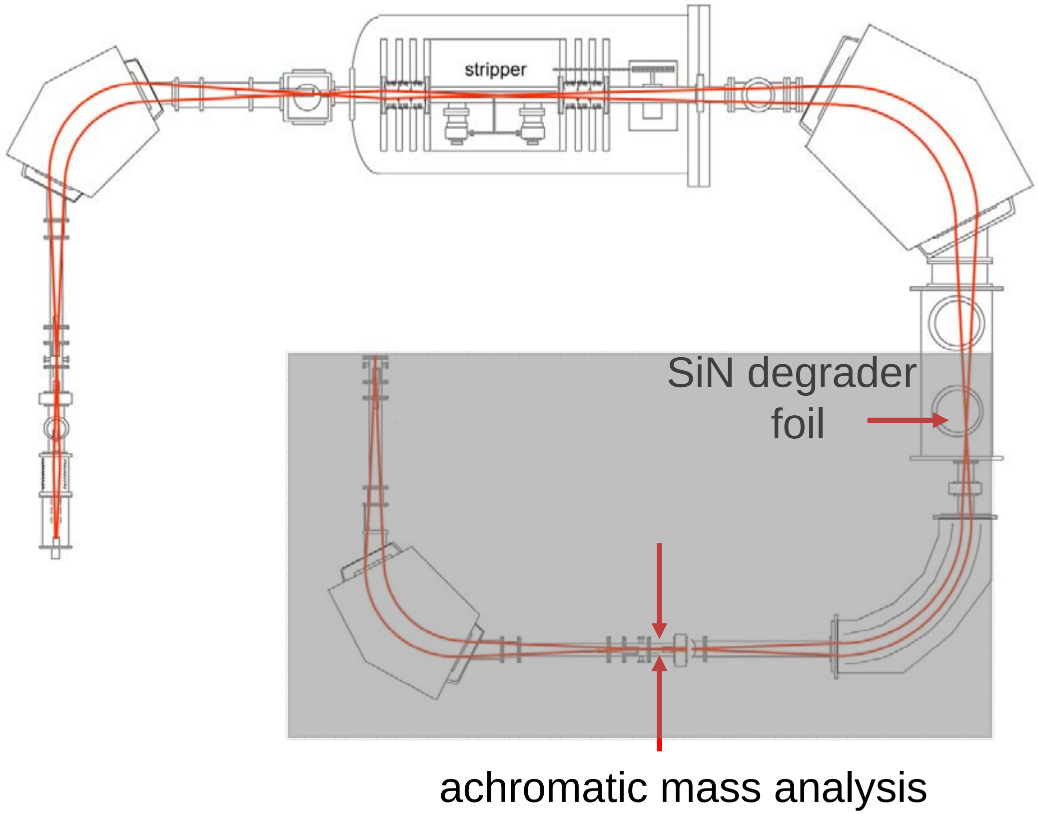
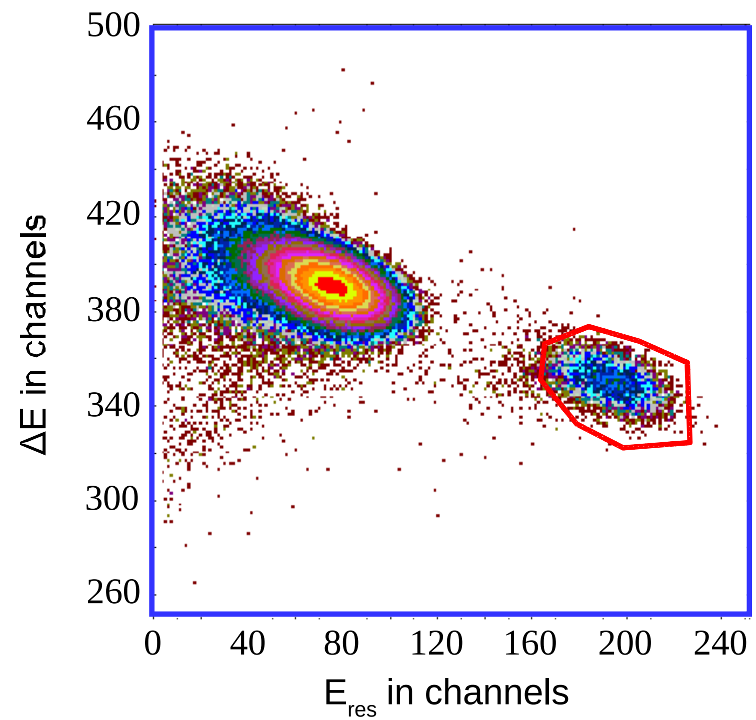
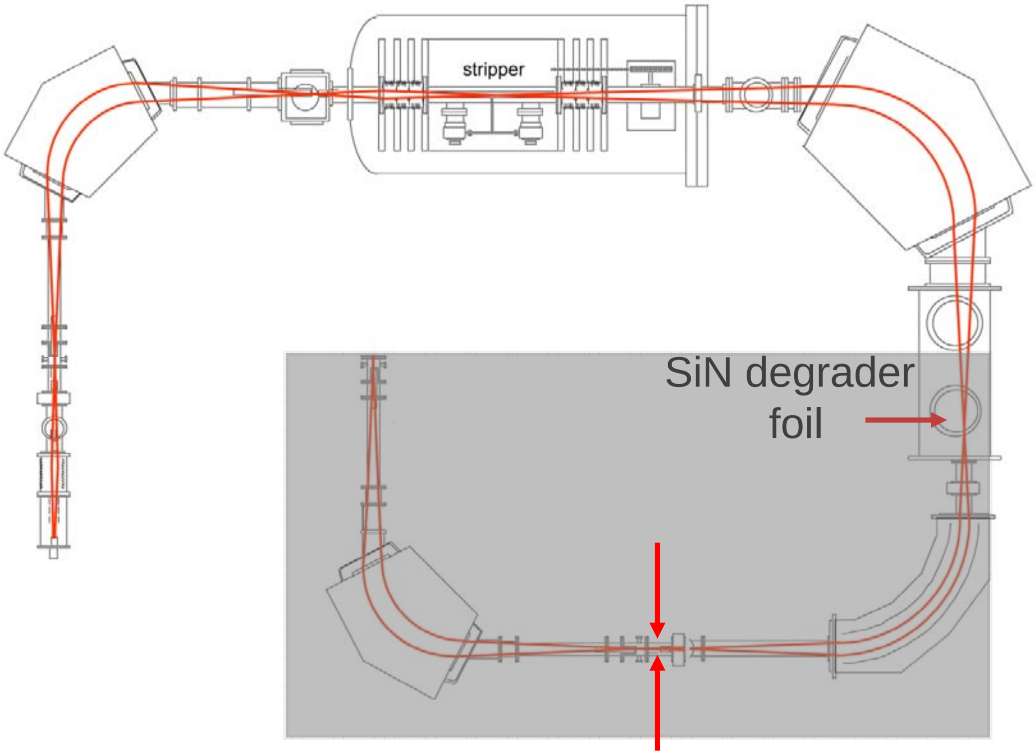


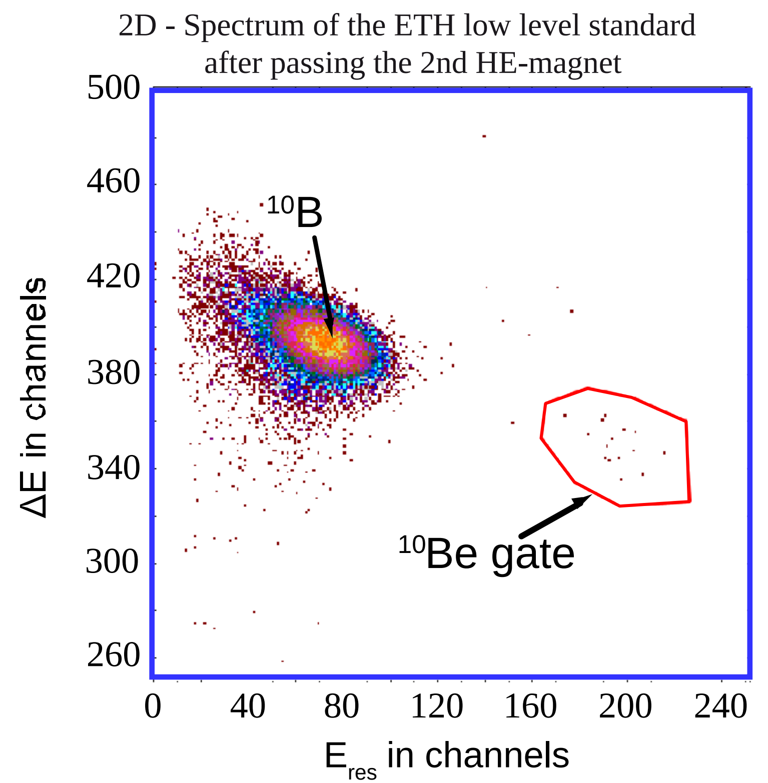


$^{10}BeO^-$ and SiN degrader

ETH- 600 kV Pelletron System



achromatic mass analysis



Transmission:

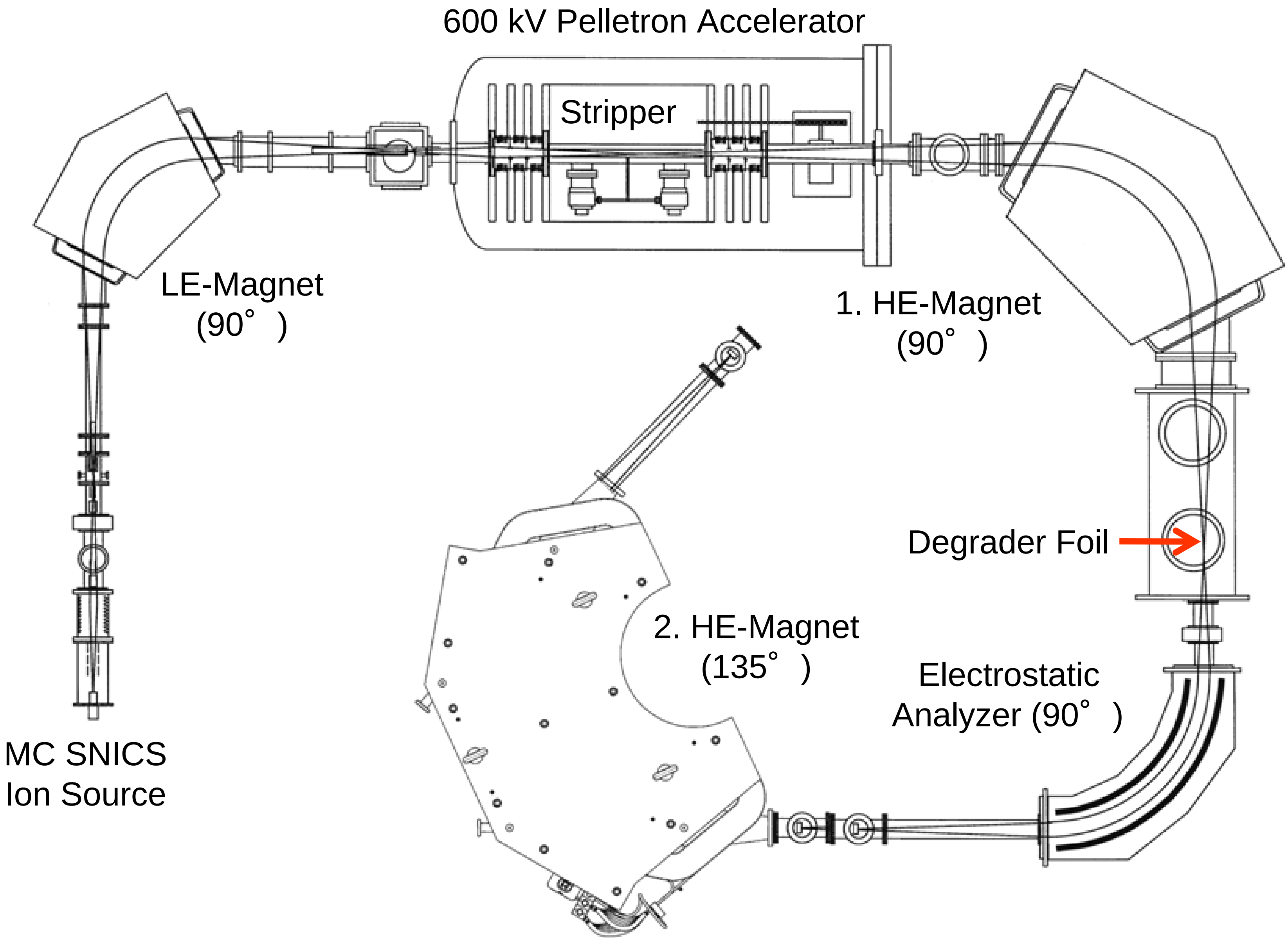
Degrader foil to detector: **15 %**

Stripper (charge state 1+): **50 - 55 %**

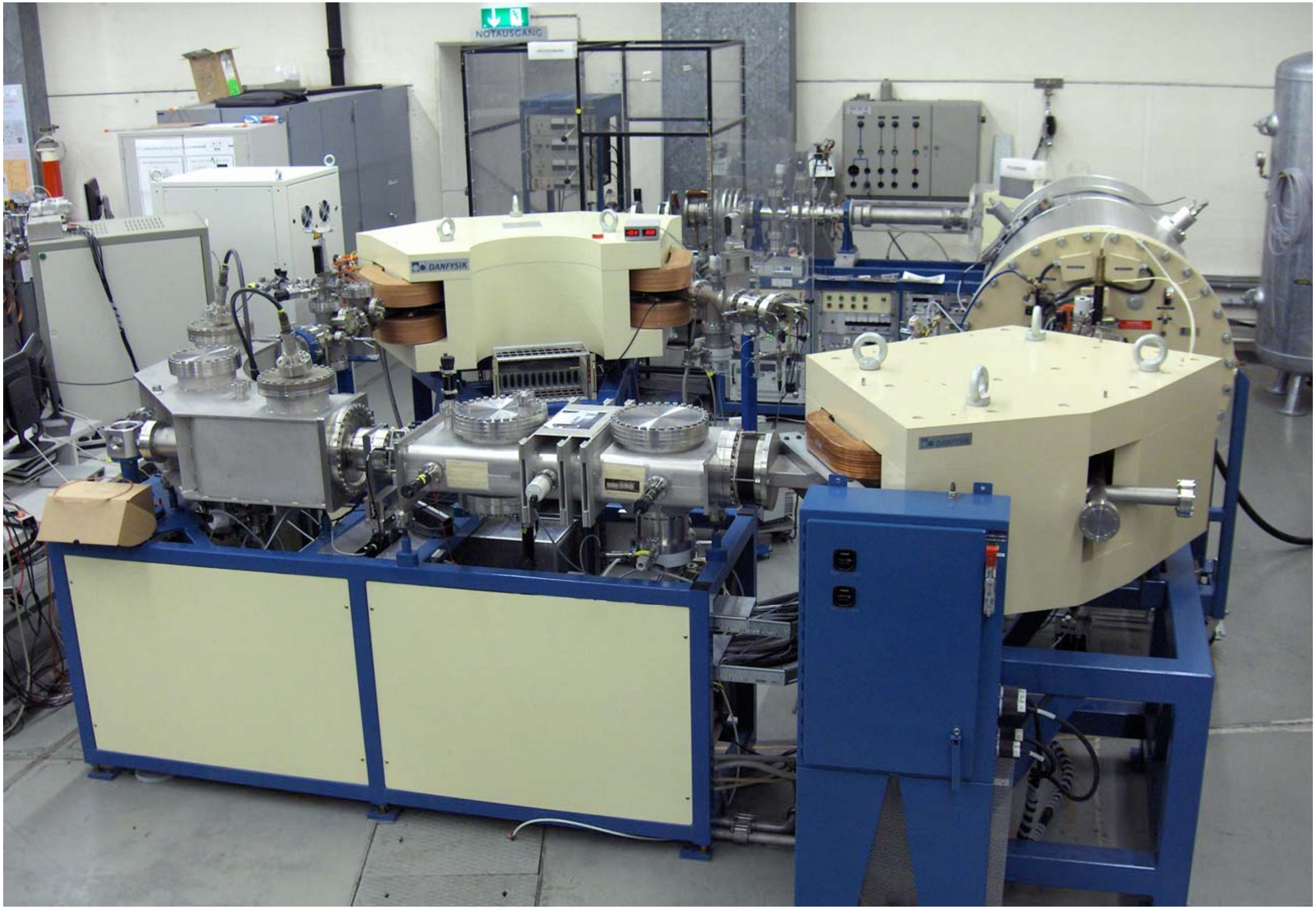
→ LE-side to detector: 7 - 8 %

Achieved $^{10}Be/^{9}Be$ background level [3]: **<5·10^-15**

Extension of HE-mass spectrometer

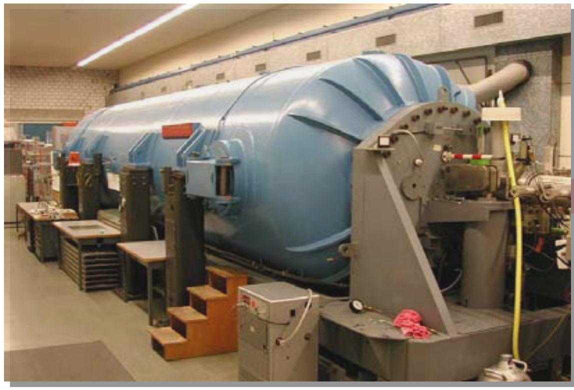


Improved HE-spectrometer setup for 600kV system



Evolution of AMS over the last 10 years

6 MV EN-Tandem

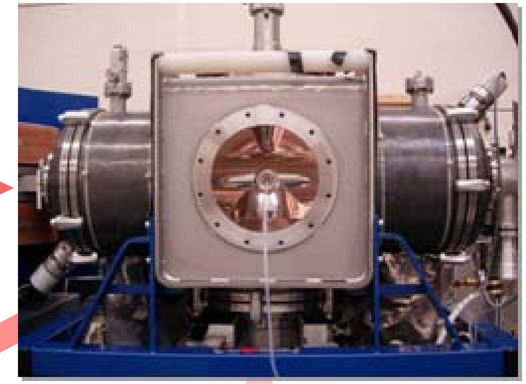


15 m

600 kV PSI/ETH system

2.5 m

200 kV system for ^{14}C

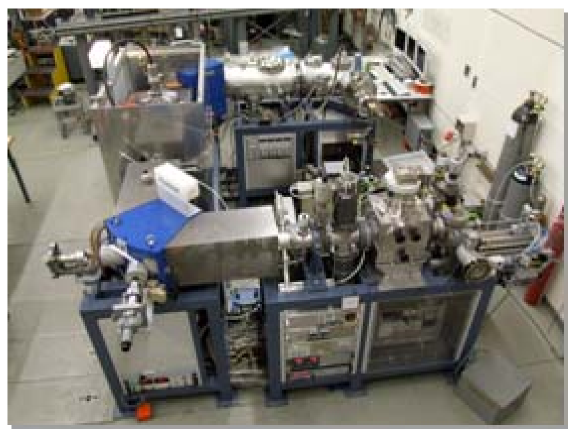


1 m

Multi isotope system

TANDY

High precision ^{14}C dating



MICADAS/DatingMICADAS

Biomedical ^{14}C applications

BioMICADAS