Accelerator Mass Spectrometry at VERA – Current status and new developments

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Overview

- **Basic Introduction of Radiocarbon dating**
- **What is AMS (Accelerator Mass Spectrometry)**
- **Description of VERA (Vienna Environmental Research Accelerator)**
- **Search for beta-delayed proton emission in ^{11}Be**
- **Application of AMS to Astrophysics**
- **New developments at VERA:**
  - Laser Photodetachment as a possible solutions to isobar problem?

Radiocarbon dating

3 important Carbon Isotopes:

Stable Isotopes: ^{12}C, ^{13}C

Radioisotope: ^{14}C, $t_{1/2} = 5730$ a

Abundances: $^{13}C/^{12}C \sim 10^{-2}$
$^{14}C/^{12}C \sim 1.2 \times 10^{-12}$

^{14}C is produced in the upper atmosphere by cosmic radiation. Production rate is not constant -> Measurement has to be calibrated!

During lifetime of a species carbon is exchanged. After the death no more exchange -> $^{14}C/^{12}C$ ratio changes by decay of ^{14}C atoms to ^{14}N.

What is AMS

AMS measures long-lived radioisotopes by counting atoms rather than waiting for their infrequent decay.

$$dN/dt = N\left(\ln 2 / t_{1/2}\right)$$

Example for ^{14}C dating ($^{14}C/^{12}C = 1.2 \times 10^{-12}$, $t_{1/2}$ = 5730 years):

1 mg of modern organic carbon contains:	**6×10^{7}**	**^{14}C atoms**
Decay counting with LSC or GPC:	**0.5**	**^{14}C atoms/h**
Atom counting with AMS:	**5×10^{5}**	**^{14}C atoms/h**

AMS is one million times more sensitive than decay counting!

What is AMS (contd.)

Radioisotopes are measured with AMS through isotope ratios

Typical isotope ratios in AMS:

radioisotope/stable isotope = 10^{-12} to 10^{-16}

The main challenge in measuring such minute isotope ratios is the separation of the radioisotope from interfering background of stable isobars and molecules.

Molecules can be removed effectively by stripping and subsequent Coulomb breakup in the terminal of the tandem accelerator

Principles of AMS

- **Sample material into an ion source: (neg. ion beam)**
- **Low energy mass separation**
- **(Tandem) accelerator**
  - stripping to positive ions (molecule destruction)
  - high particle energies for identification
- **High energy mass separation**
- **Particle identification (detector + Faraday cups)**

Principles of AMS (contd.)

Three good reasons for the use of negative ions:

$^{14}C^{-}$ – ~~$^{14}N^{-}$~~

$^{26}Al^{-}$ – ~~$^{26}Mg^{-}$~~

$^{129}I^{-}$ – ~~$^{129}Xe^{-}$~~

Principles of AMS (contd.)

Sometimes special negative molecules help:

$$^{41}\mathrm{CaH}_3^- \quad – \quad \xcancel{^{41}\mathrm{KH}}_3^-$$

$$^{10}\mathrm{BeF}^- \quad – \quad \xcancel{^{10}\mathrm{BF}}^-$$

$$^{182}\mathrm{HfF}_5^- \quad – \quad \xcancel{^{182}\mathrm{WF}}_5^-$$

All other radioisotopes are more difficult to be cleaned up from stable isobar interference.

Principles of AMS (contd.)

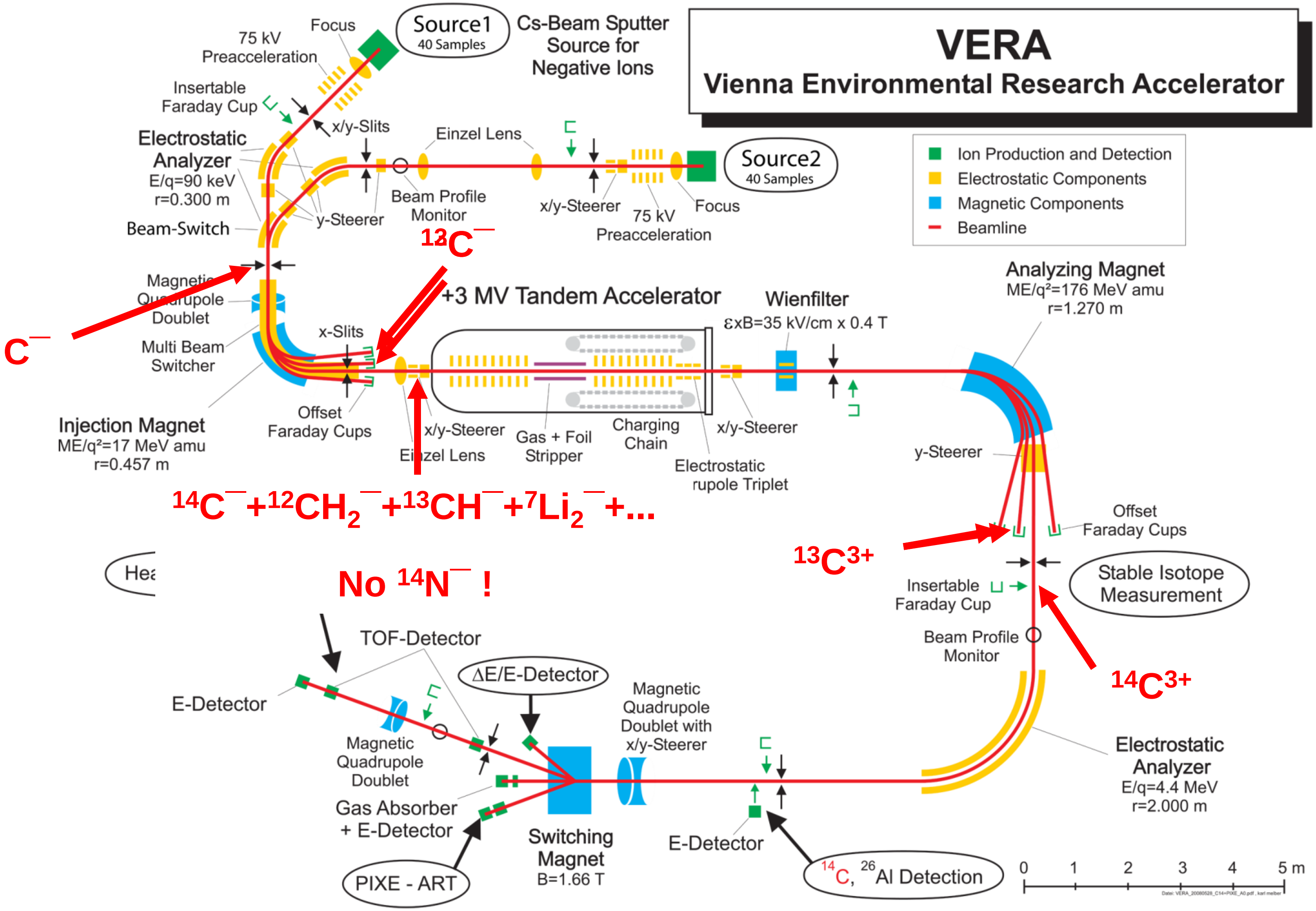
The remaining molecular background after low energy mass separation is destroyed in the stripping process:

$$^{14}\mathrm{C}^{-} \rightarrow {}^{14}\mathrm{C}^{3+}$$

$$^{13}\mathrm{CH}^{-} \rightarrow \xcancel{^{13}\mathrm{CH}^{3+}}$$

$$^{12}\mathrm{CH}_2^{-} \rightarrow \xcancel{^{12}\mathrm{CH}_2^{3+}}$$

VERA



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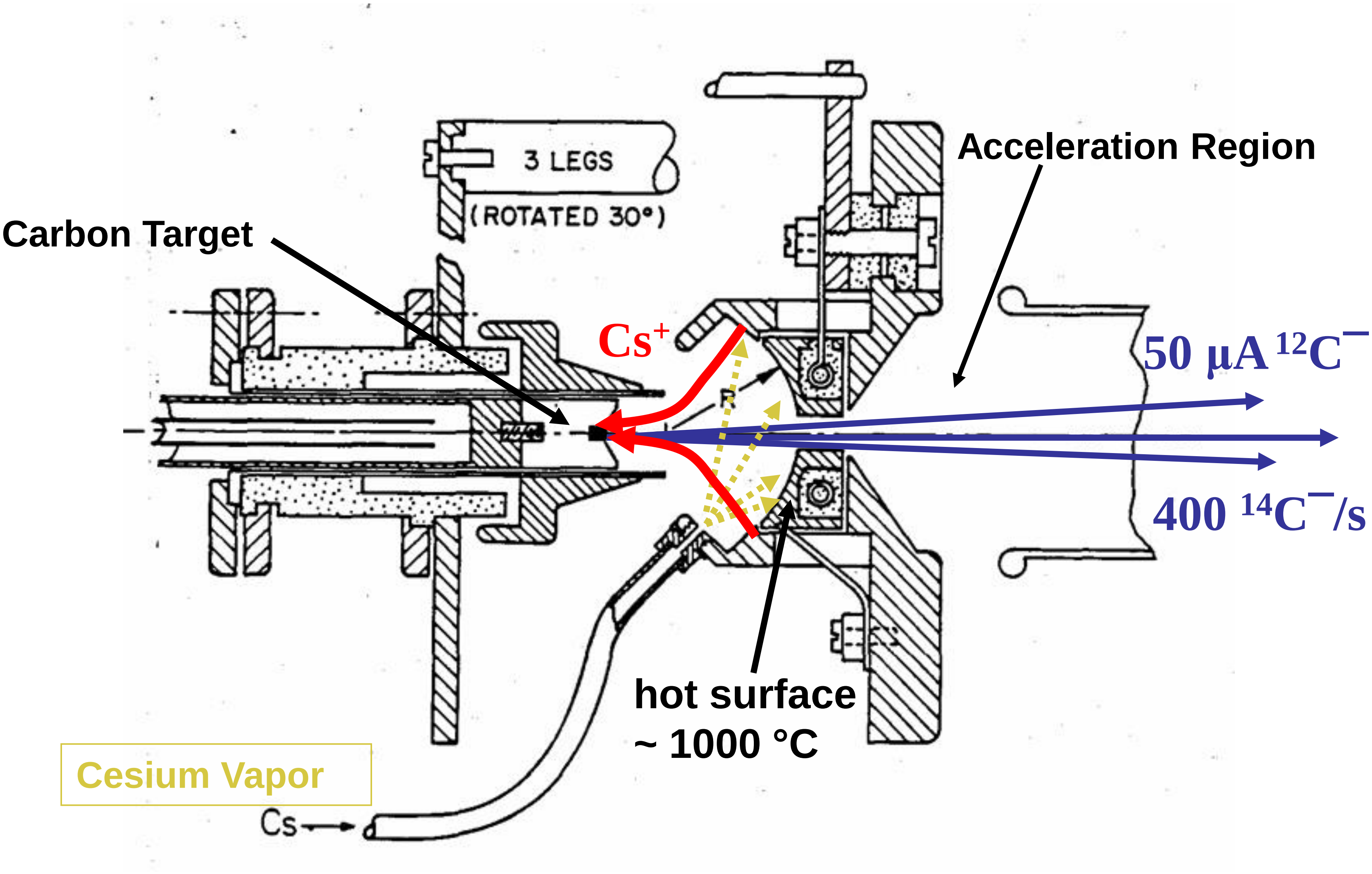
The Cesium-Beam Sputter Source for Negative Ions



Carbon Target

Acceleration Region

Cs^+

$50\ \mu A\ {}^{12}C^-$

$400\ {}^{14}C^-/s$

hot surface
~ 1000 °C

Cesium Vapor

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Cs-Beam Sputter Source for Negative Ions

40 Samples

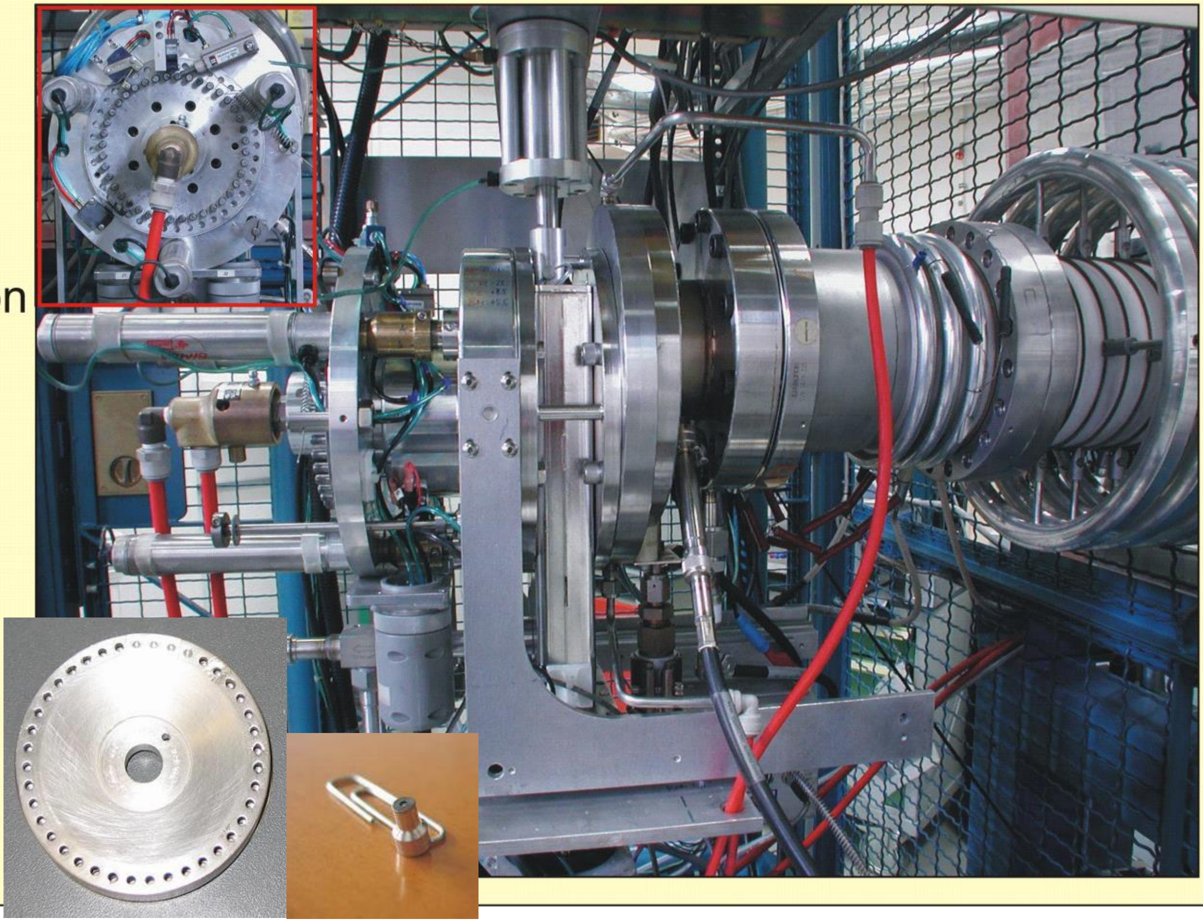
max. 75 keV
Preacceleration

Ion Currents:

C^-: 60 µA

BeO^-: 3 µA

UO^-: 100 nA



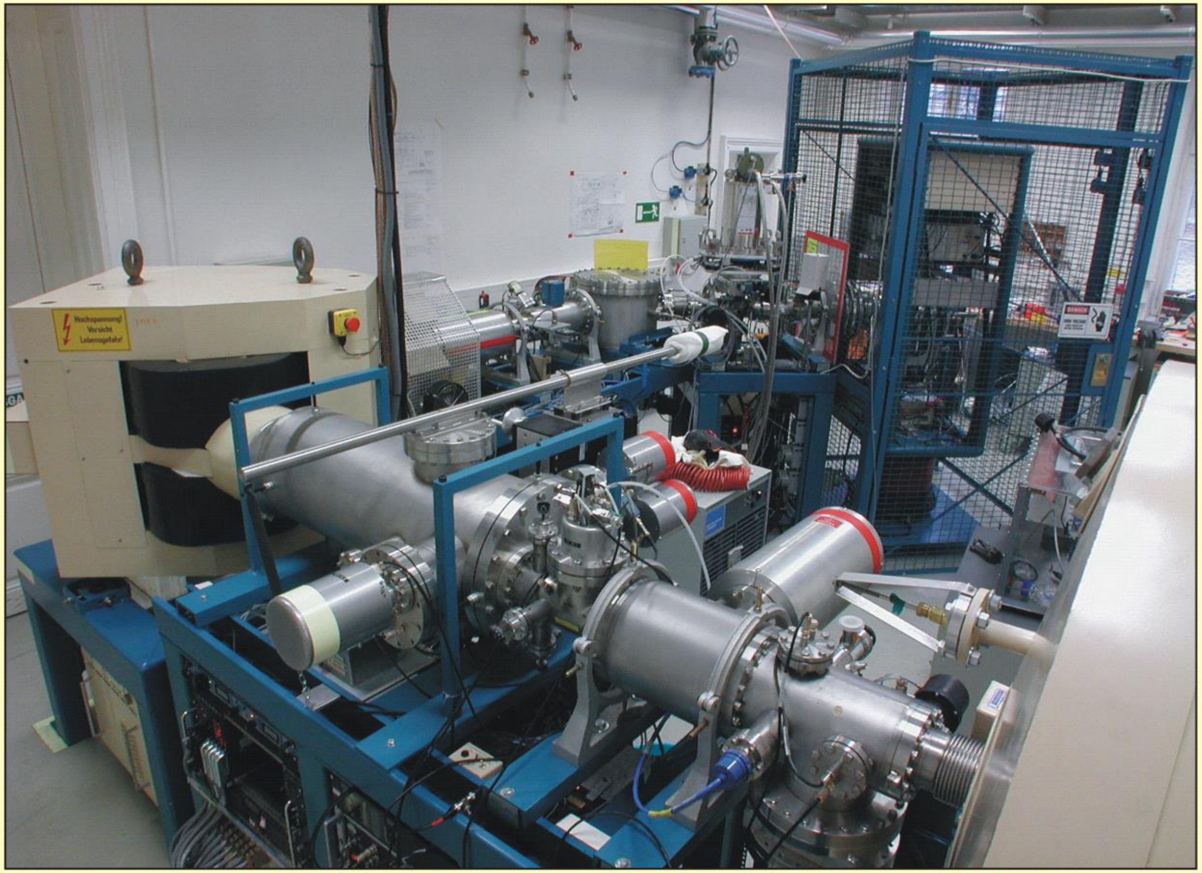
Injection Side of VERA

45° ESA:
r = 300 mm
E/q = 92 kV

Quadrupole Doublett

90° Injection Magnet
r = 457.2 mm
ME/q^2 =
15 MeV amu

Multi Beam Switcher



+3 MV Tandem Accelerator

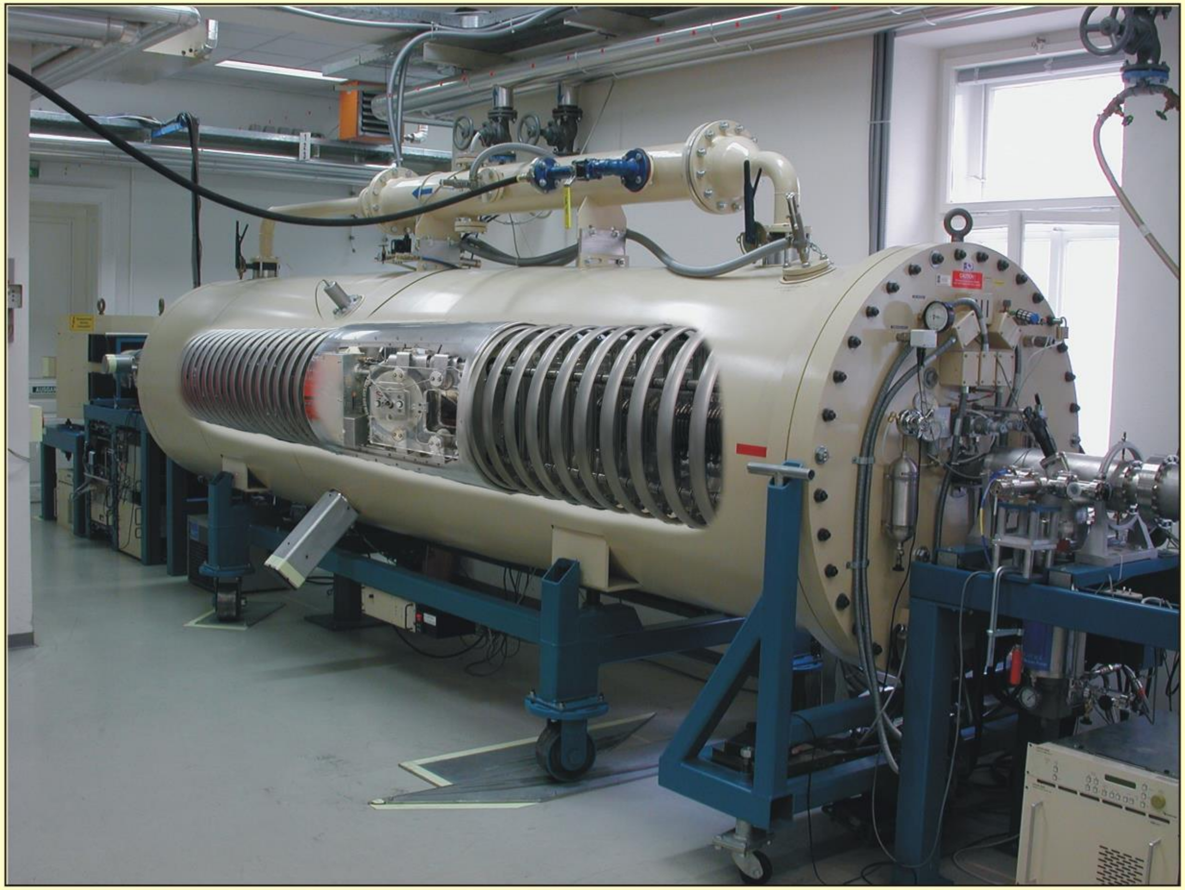
Pelletron type

2 charging chains

maximal charging current: 230 μA

Ar gas stripper/ foil stipper

insulating gas: SF_6

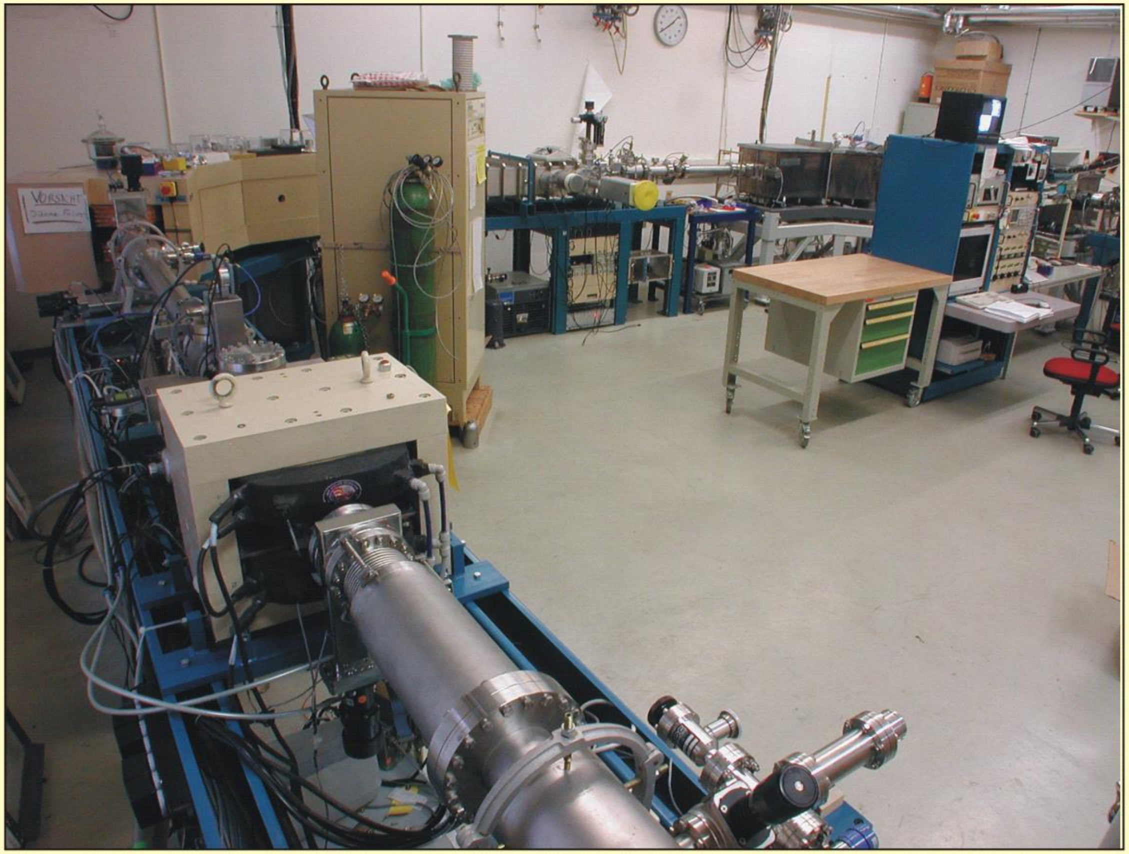


Analyzing Side of VERA

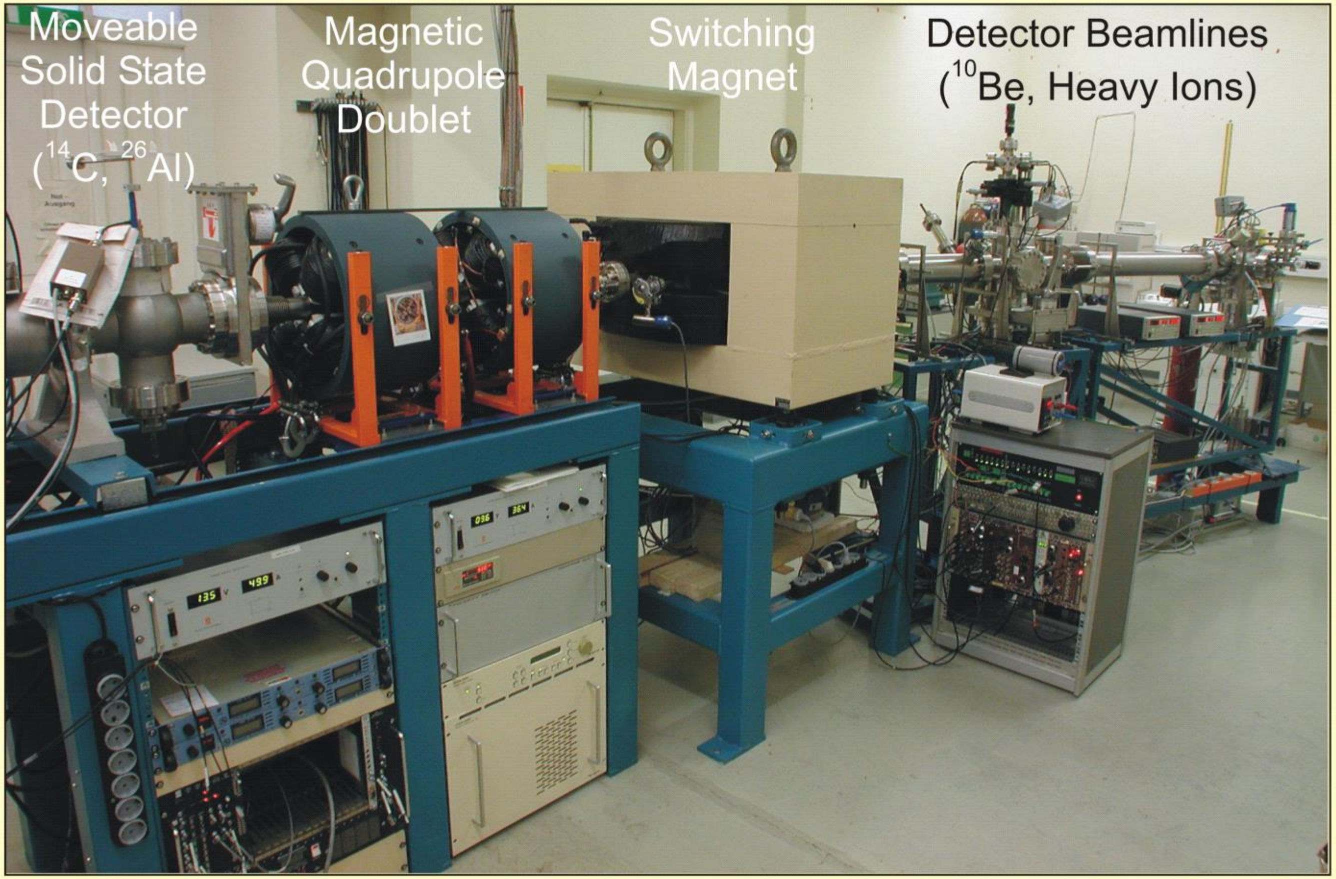
Wienfilter
ExB =
35 kV/cm x 0.4 T

Analyzing Magnet
r = 1270 mm
ME/q^2 =
176 MeV amu

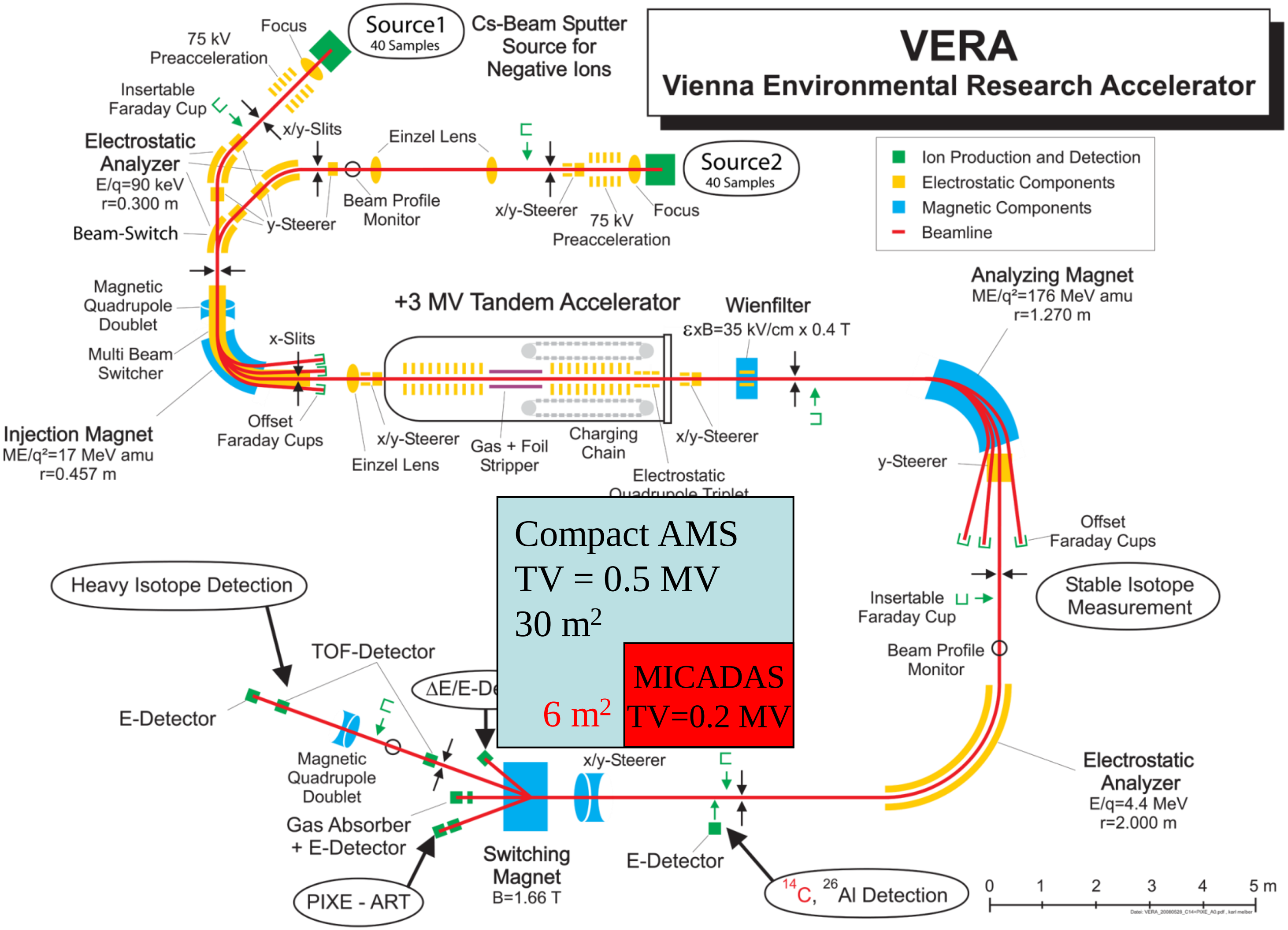
Electrostatic Analyzer



Detector Beamlines



Size of VERA compared to Compact- and Mini-AMS



Compact AMS

TV = 0.5 MV

30 m^2

6 m^2

MICADAS

TV=0.2 MV

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Latest Developments at the ETH Zurich:

Radiocarbon AMS System with 200 kV Terminal Voltage
Ions are stripped to 1^+ charge state

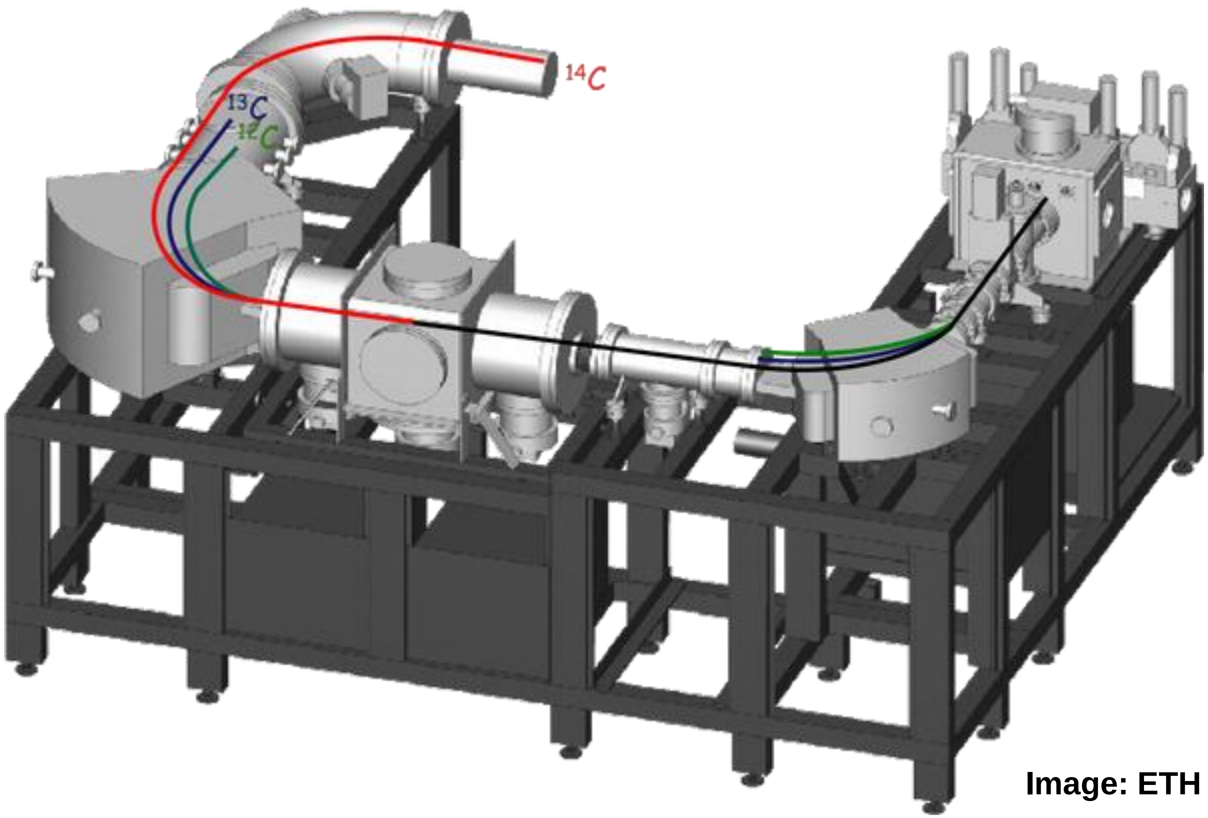


Image: ETH Zurich

Some AMS-nuclides measured at VERA (3 MV)

Radio-nuclide	Half-life	Overall Efficiency	Detection Limit	Precision
^{10}Be	1.39 Ma	1×10^{-4}	$< 5\times10^{-16}$	< 3%
^{14}C	5730 a	2×10^{-2}	$< 3\times10^{-16}$	< 0.5 %
^{26}Al	0.7 Ma	5×10^{-4}	$< 6\times10^{-16}$	< 1.0 %
^{129}I	15.7 Ma	1×10^{-2}	2×10^{-14}	2 %
^{182}Hf	8.9 Ma	1×10^{-4}	1×10^{-11}	5 %
^{236}U	23.4 Ma	--	$< 6\times10^{-11}$	5 %
^{244}Pu	80.0 Ma	$> 4\times10^{-5}$	--	5 %
^{36}Cl	0.3 Ma	--	1×10^{-13}	
^{41}Ca	0.1 Ma	--	1×10^{-13}	
^{55}Fe	2.73 a	--	$< 1\times10^{-15}$	

Beta-p in ^{11}Be

Why look for beta-delayed proton emission in ^{11}Be?

- **Only possible due to the low neutron separation energy:**

$$Q_{\beta p} = m_n - m_H - S_n(Z,N) = 0.782 - S_n = 0.280 \text{ MeV}$$

- **Similar to beta-delayed deuteron emission in ^{6}He and ^{11}Li**
- **If occurring: coupled to the neutron halo in ^{11}Be**
- **According to theory the decay mechanism is also dynamically coupled to the halo structure: decay directly to continuum**

→ This needs experimental proof!

Collection point

Theoret. calculations predict

βp branching ratio ~ 10^{-8}

Aim for collection: 10^{12} ^{11}Be

Estimated collection time with

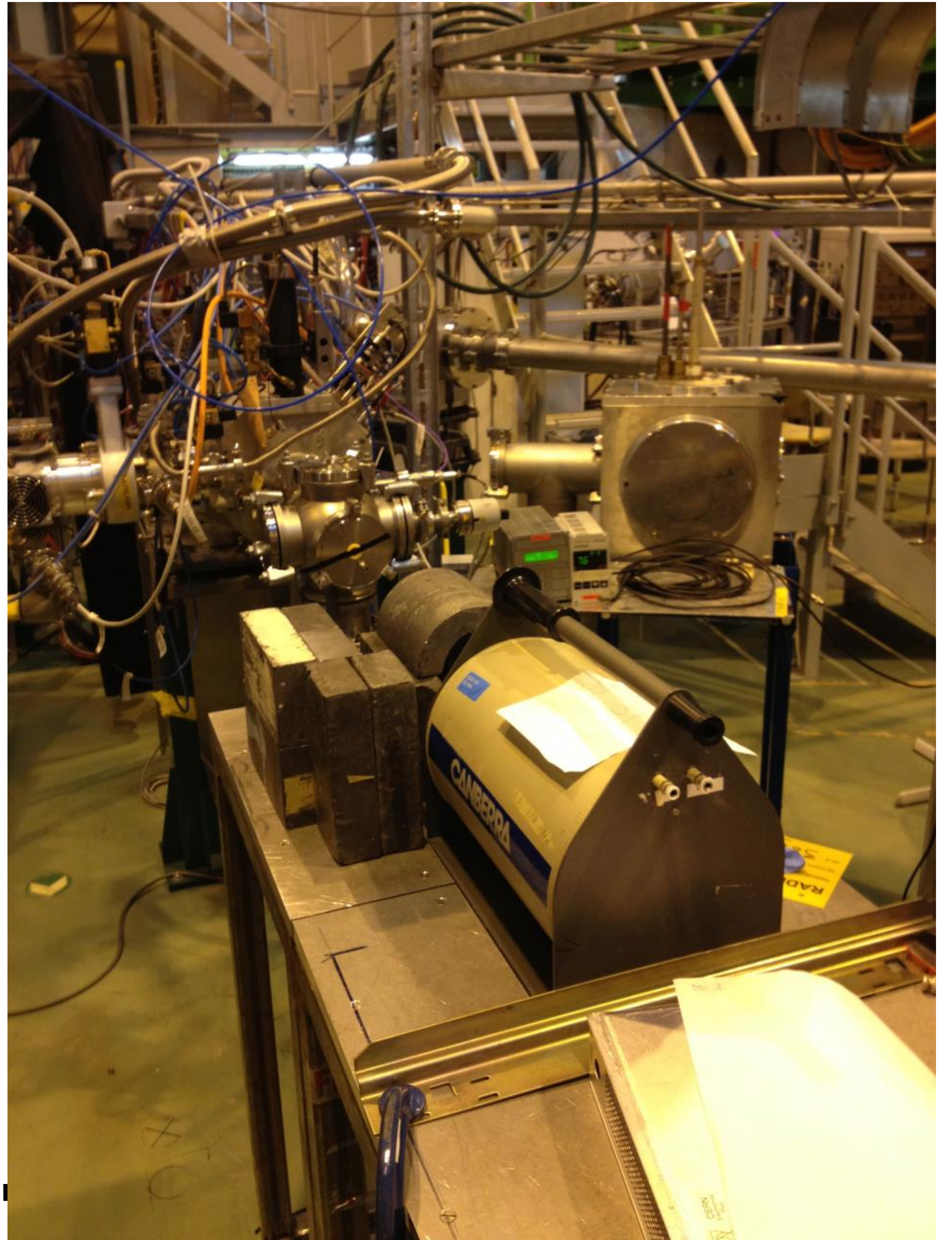
actual ISOLDE yield: 16 hrs

Collection was continuously

monitored with a HPGE

detector. (2124.7 keV gamma

line from ^{11}Be decay.



Sample preparation

After collection samples where transferred to Vienna and chemically treated to get AMS samples.

Implantation depth of 60 keV beryllium into copper is below 1 µm.

→ only surface was leached with nitric acid

359 µg ^{9}Be carrier was added to the solution

Solution treated with ammonium hydroxide to precipitate $Be(OH)_2$

Copper stays in the solution

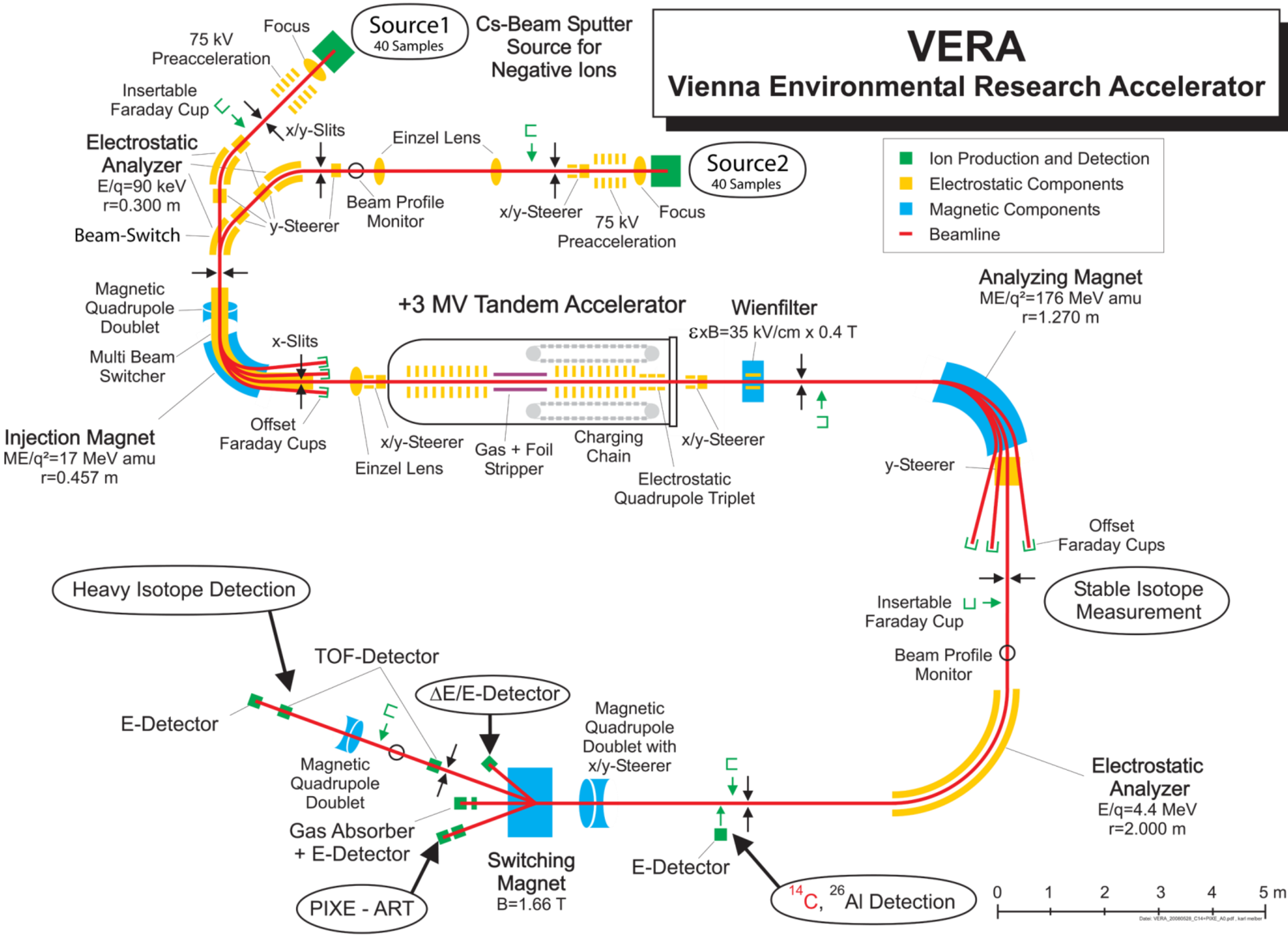
Beryllium hydroxide was dried at 900℃ for at least 8 hrs forming BeO

Mixed 1:1 with high purity copper and pressed into sample holder

^{10}Be/^{9}Be measurement at VERA

- ^{10}Be/^{9}Be ratio determined with AMS.
- Extract BeO$^-$ from ion source
- Accelerate and strip to Be^{2+}
- Used dual anode gas ionization chamber with passive absorber (SiN) to get rid of isobar ^{10}B

- Overall efficiency: 5×10^{-4}



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Two beam times at ISOLDE: July ’12, Dec ’12

July 2012:

Due to problems with Booster transfer line and with the high voltage of ISCOOL ^{11}Be yields were not sufficient. However beam time was used to collect ^{10}Be samples to check sample preparation and efficiency of VERA → successful

December 2012:

36 hrs collection of ^{11}Be: $(1.447 \pm 0.055)\cdot 10^{12}$ atoms (Sample S1)

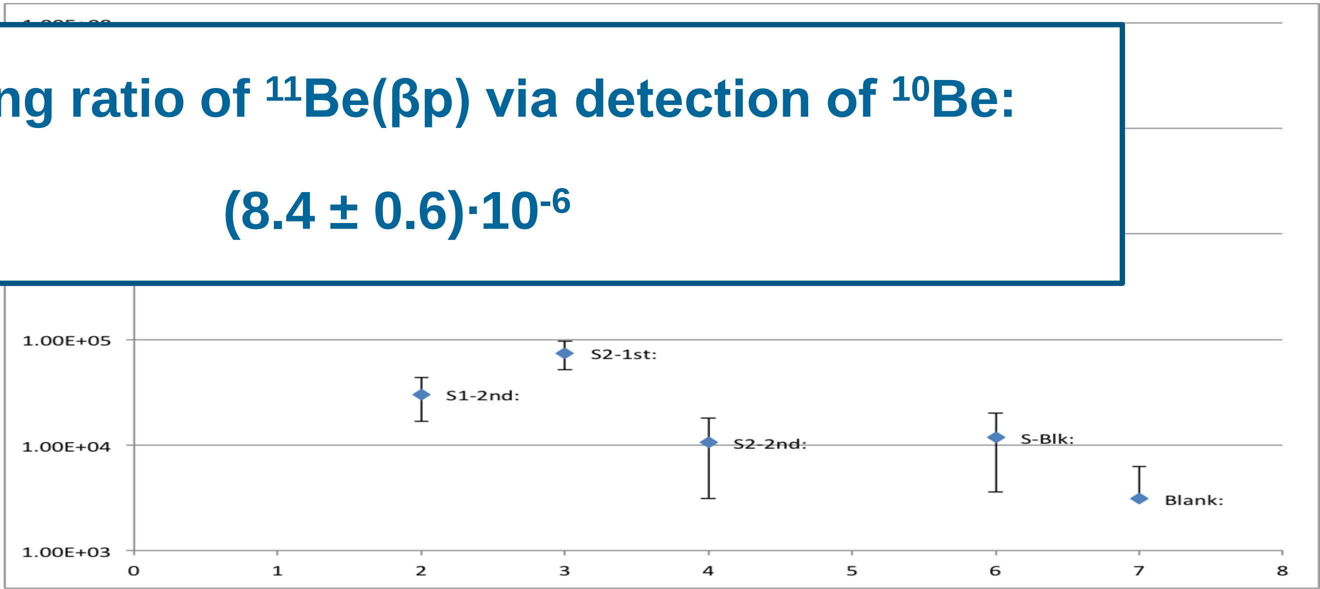
Additional collection of two calibration samples:

- S2: ^{11}Li (0.02 mass units above ^{11}Be), decays to ^{10}Be via βn (83%)
- S3: ^{10}Be, 3.5 pA, 1 s collection → $2.2\cdot 10^{7}$ atoms

Results

AMS Results of the December Run:

Sample	^{10}Be/ ^{9}Be ratio	^{10}Be atoms
S1-1st	$(4.87 \pm 0.13) \cdot 10^{-13}$	$(1.17 \pm 0.05) \cdot 10^{7}$
S1-2nd	$(1.26 \pm 0.56) \cdot 10^{-15}$	$(3.03 \pm 1.35) \cdot 10^{4}$
S2-1st	$(3.10 \pm 0.94) \cdot 10^{-15}$	$(7.45 \pm 2.27) \cdot 10^{4}$
S2-2nd	$(4.4 \pm 3.1) \cdot 10^{-16}$	$(1.06 \pm 0.75) \cdot 10^{4}$
S3-1st	$(1.54 \pm 0.03) \cdot 10^{-12}$	$(3.70 \pm 0.13) \cdot 10^{7}$
S-blank	$(4.9 \pm 3.4) \cdot 10^{-16}$	$(1.18 \pm 0.82) \cdot 10^{4}$
blank	$(1.3 \pm 1.3) \cdot 10^{-16}$	$(3.12 \pm 3.12) \cdot 10^{3}$



Branching ratio of ^{11}Be(βp) via detection of ^{10}Be:

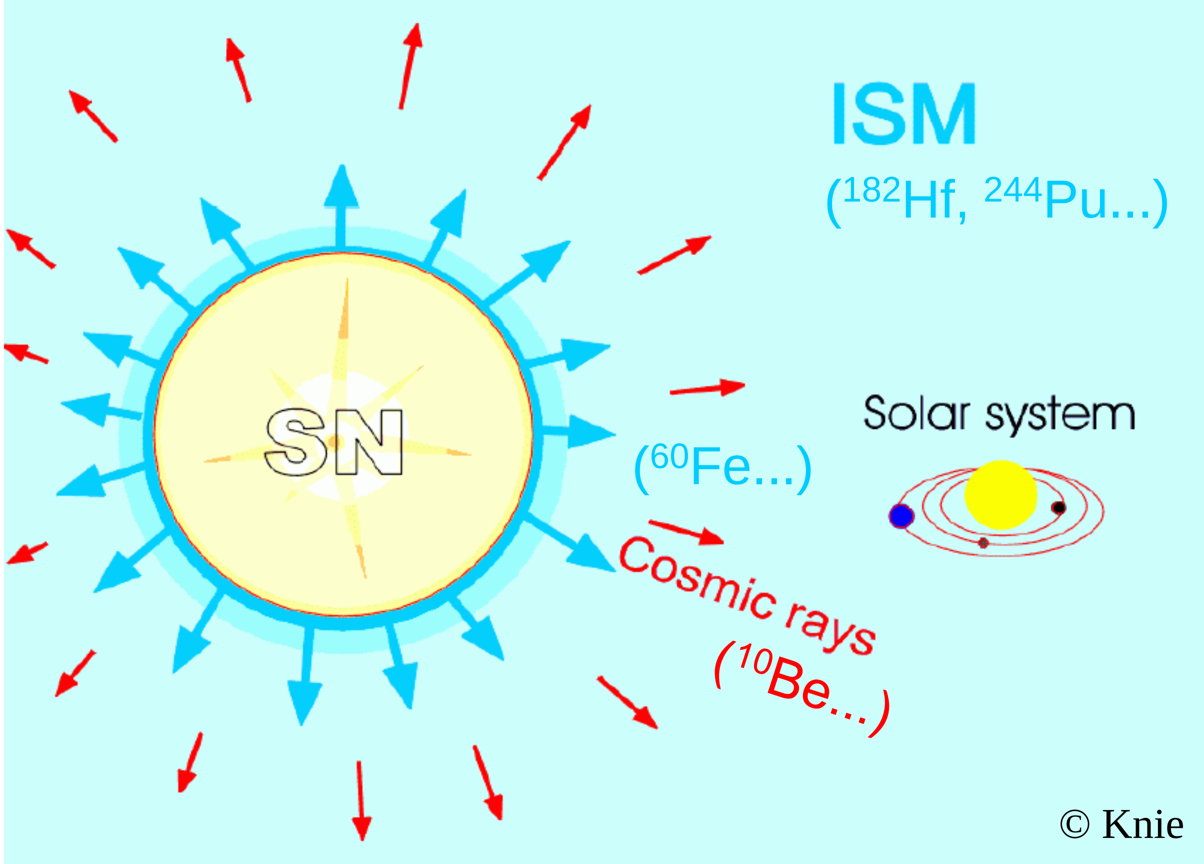
$(8.4 \pm 0.6) \cdot 10^{-6}$

Applications of AMS to Astrophysics

- **Search for live radioisotopes as signatures of a nearby SN (^{60}Fe, ^{244}Pu, ...)**
- **Meteorites**
  - cosmic ray exposure, ...
- **Nucleosynthesis models**
  - measure neutron capture cross sections
- **Solar system abundance**
  - early solar system (presolar, extinct radionuclides)
  - solar system abundance now
- **Search for superheavy elements in nature**
  - search long lived supernova produced SHEs ores

Live long-lived radionuclides on Earth

nearby supernova (SN II): < 100 pc, rate ~ 0.3 − 10 $(\mathrm{Ma})^{-1}$



G. Korschinek, K. Knie et al. TU Munich

Life ^{60}Fe as Signature of a Nearby Supernovae

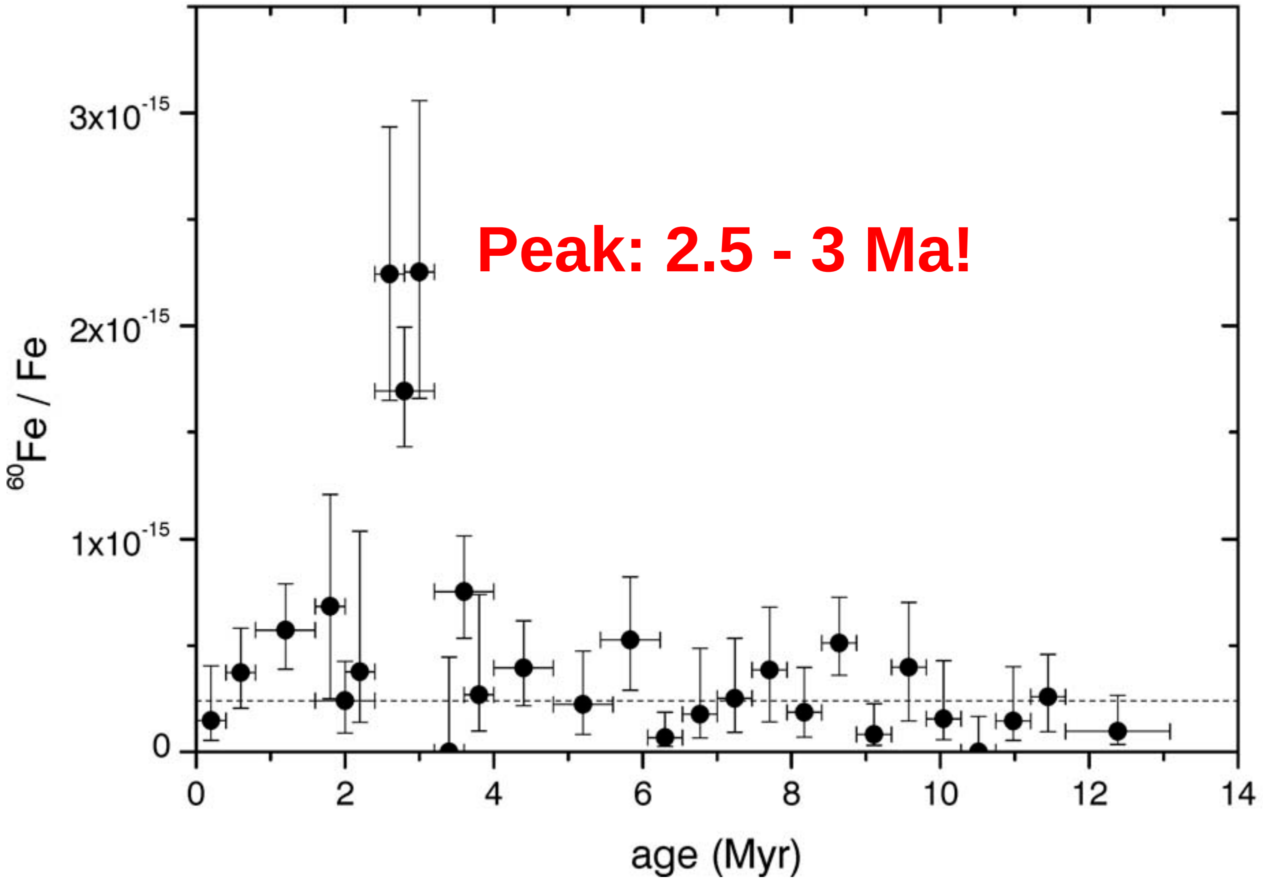
- Deep-sea manganese crust
- Growth: 2.5 mm / Ma
- 28 layers (1−2 mm) were measured for ^{60}Fe-content ($T_{1/2}$=1.5 Ma)
- ^{60}Fe: no significant terrestrial production
- GAMS-setup Munich (14−MV tandem)

Life ^{60}Fe as Signature of a Nearby Supernovae (contd.)

VOLUME 93, NUMBER 17 PHYSICAL REVIEW LETTERS week ending 22 OCTOBER 2004

^{60}Fe Anomaly in a Deep-Sea Manganese Crust and Implications for a Nearby Supernova Source

K. Knie,[1] G. Korschinek,[1,*] T. Faestermann,[1] E. A. Dorfi,[2] G. Rugel,[1,3] and A. Wallner[1,3]



Peak: 2.5 - 3 Ma!

Past and Ongoing Measurements at VERA:

Deep-Sea Manganese Crust

- **^{244}Pu**

  No natural source for ^{244}Pu on earth. Only recently produced by atmospheric nuclear weapon tests or in nuclear power plants. Can the ^{60}Fe peak also be seen in ^{244}Pu?

- **^{10}Be, ^{26}Al**

  ^{10}Be and ^{26}Al are produced by cosmic radiation. The increased cosmic radiation from a SN should result in an increase of both.

- **^{182}Hf**

  ^{182}Hf only produced in nuclear reactors. Is there a peak in deep sea crusts or sediments?

Isobaric background

How to measure isotopes under the influence of stable isobaric background?

→ only counting atoms no longer sufficient, particle identification is needed.

Solutions:

- **Measure ion velocity with TOF detector**
- **Use the Z-dependence of energy loss in matter**

  e.g. gas-filled magnet, split anode gas ionization chamber

  → required particle energy increases with mass

  highest mass so far: ^{60}Fe at the Munich 14-MV tandem.

→ Other solutions have to be found !!

Non-Resonant Laser-Photodetachment

- **Negative Ions are hit by an incident Laser beam**
- **Electron can be detached if photon energy > EA (electron affinity)**

  → non-resonant process !!!
- **Cross-section around threshold described by Wigner-Law:**

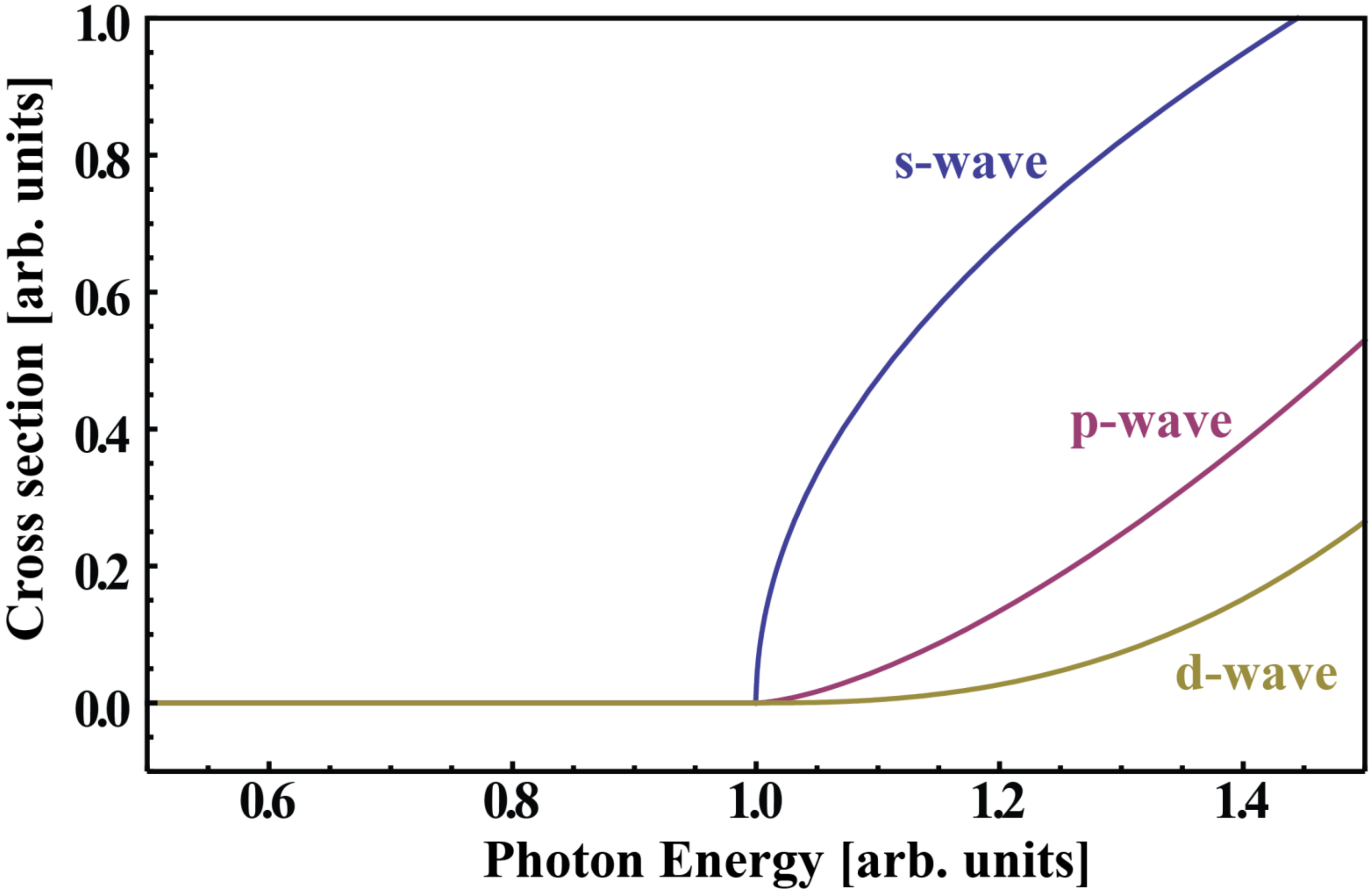
$$\sigma \propto \left(E - E_{EA}\right)^{l+1/2}$$

- **Using Laser Photodetachment for suppression of isobars was applied by Berkovits et al. for the case of S/Cl and Co/Ni**
  - Berkovits et al., NIM A281 (1989) 663-666
  - Berkovits et al., NIM B52 (1990) 378-383
- **Drawback: low cross-sections for the Photodetachment process and low duty cycles of the pulsed Lasers**

  → Significant Improvement of the Method is necessary

Wigner-Law

Cross Section behavior around the threshold for s-, p- and d-wave detachment (Wigner-Law).



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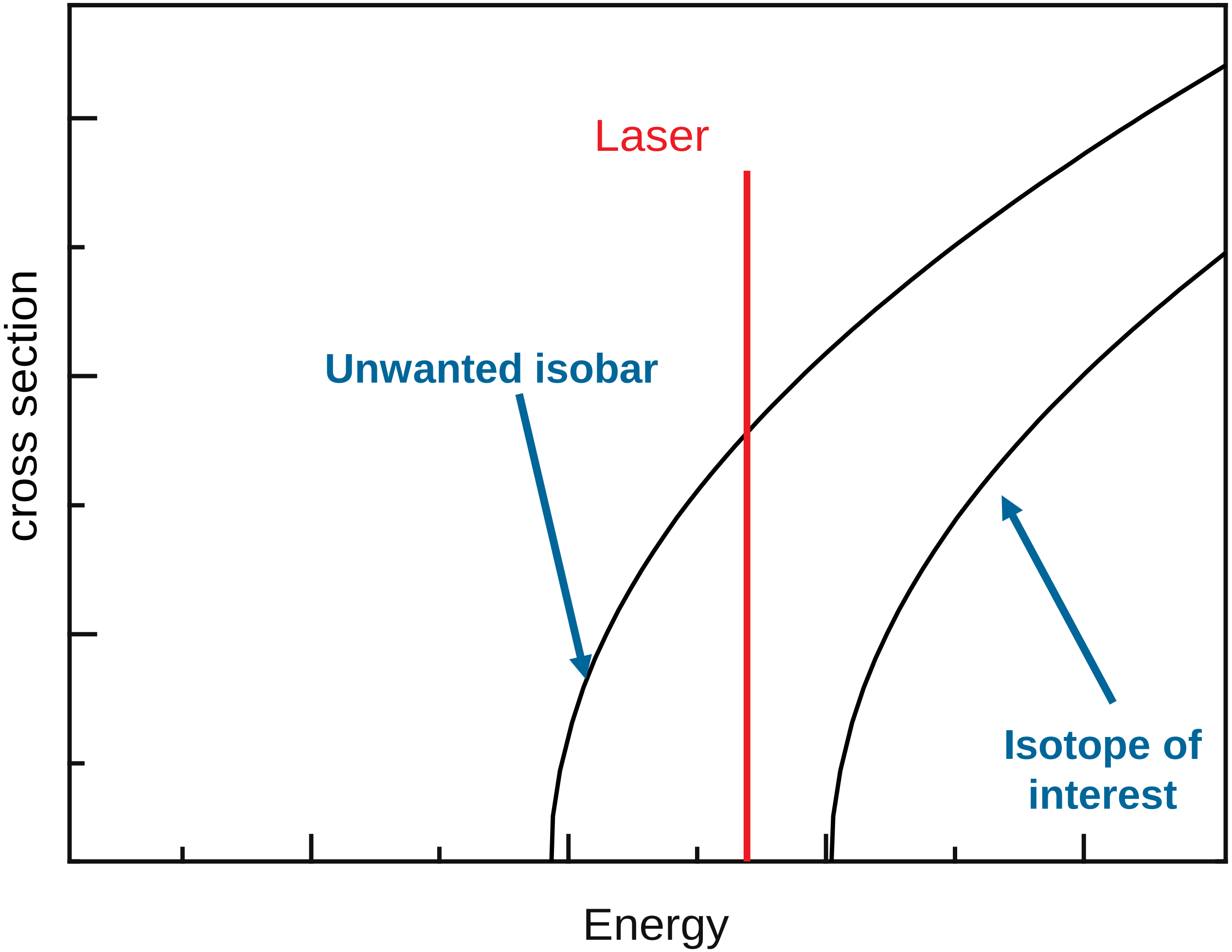
Selective Detachment

Laser will neutralize isobar while isotope of interest remains unchanged.



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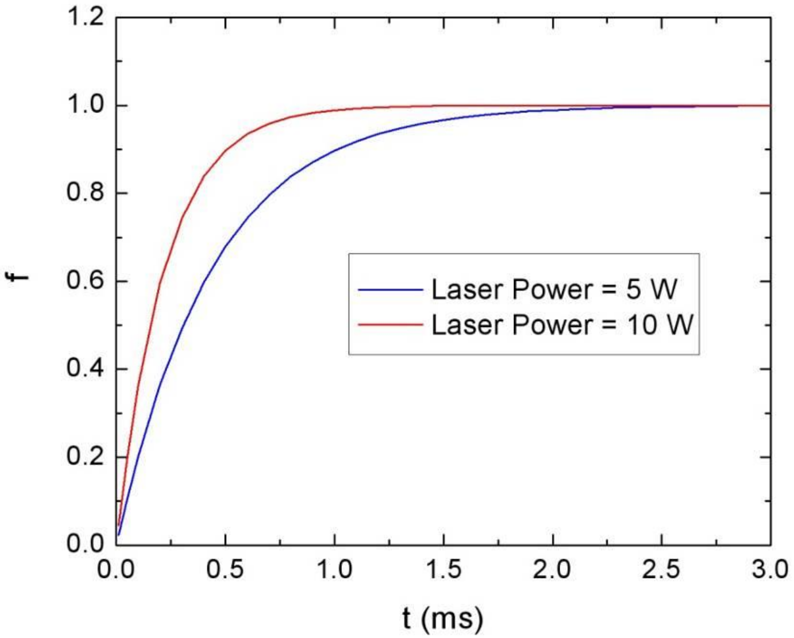
Non-Resonant Laser-Photodetachment (II)

Fraction of negative ions removed by photodetachment is given by:

$$f = 1 - e^{-\sigma\phi t}$$

ϕ = photon flux (photons/ cm^2 s)
σ = photodetachment cross section (cm^2)
t = laser-ion interaction time (s)



λ = 1064 nm, $\sigma(Co^-) = 6 \times 10^{-18}$ cm^2
Laser beam size = Φ3 mm

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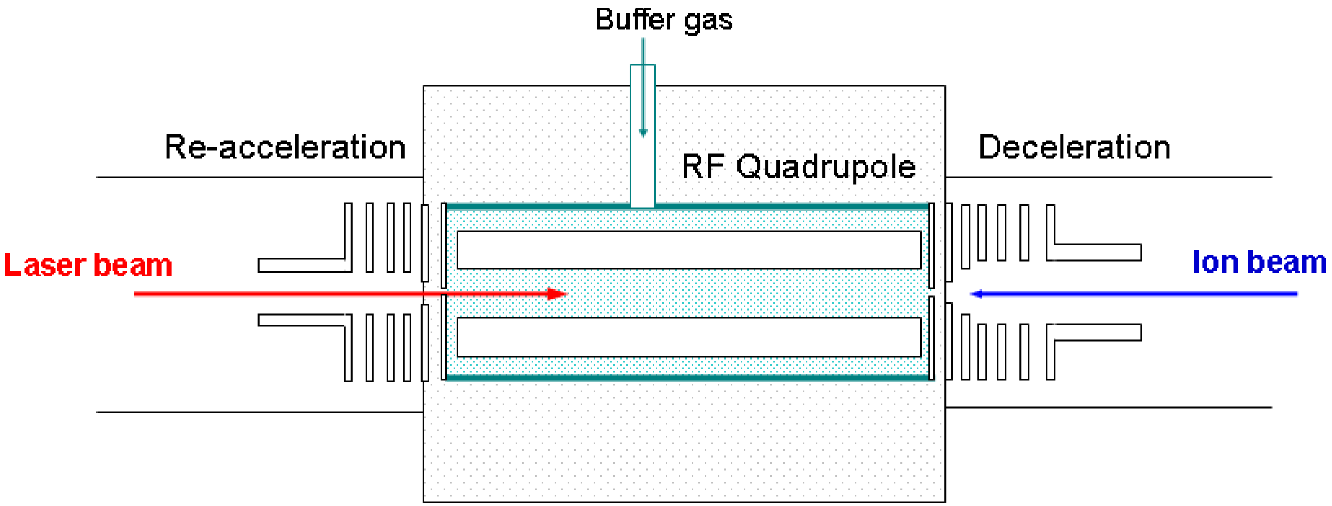
RF Quadrupole

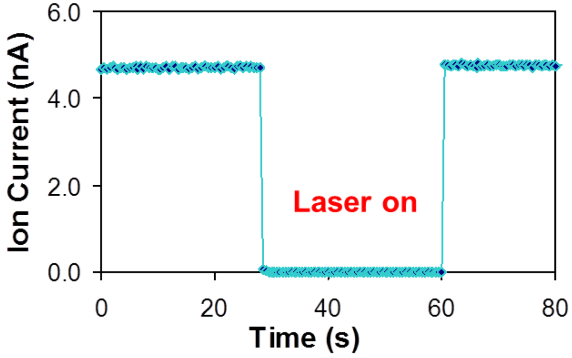
APPLIED PHYSICS LETTERS 87, 113504 (2005)

Isobar suppression by photodetachment in a gas-filled rf quadrupole ion guide

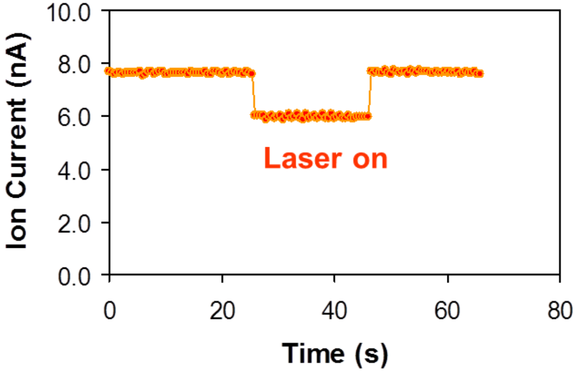
Y. Liu, J. R. Beene, C. C. Havener, and J. F. Liang
Physics Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6368



^{59}Co: 99.99% detached



^{58}Ni: 20% detached



Nd:YAG Laser at 1064 nm
Power ~ 3 W
He pressure ~ 6x10^{-2} mbar

P. Andersson et al., J. Appl. Phys. 107, 026102 (2010)

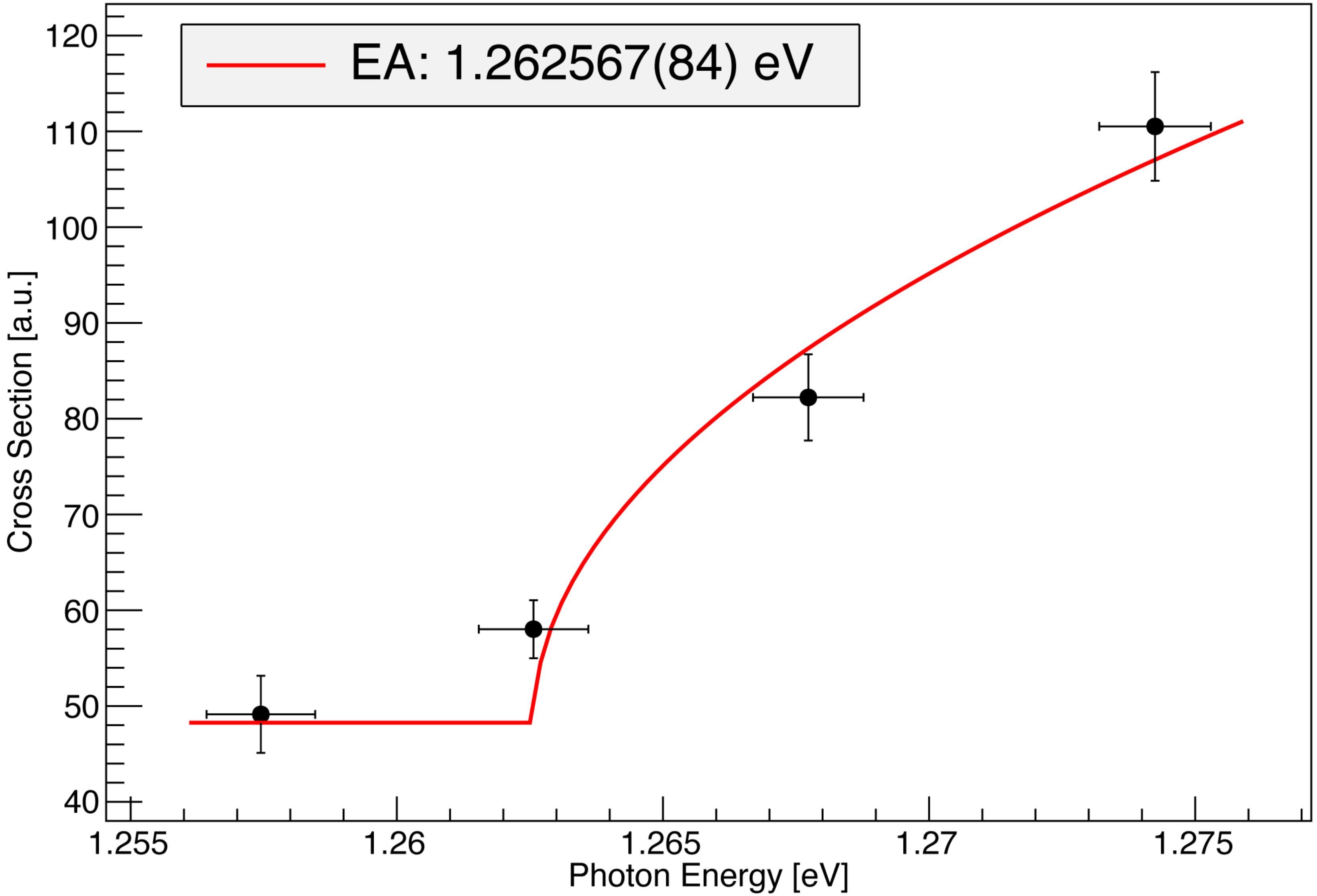
First photodetachment studies

Construction of the cooler is in progress. First results expected for spring 2014.

First isobar systems to be studied:

- **Selective suppression of molecular isobars CH^- and CH_2^-**
  Electron Affinities:
  C^-: 1.262119(20) eV, 982.3494(16) nm
  CH^-: 1.238(8) eV, 1001(6) nm
  CH_2^-: 0.652(6) eV, 1901(17)nm
- **Suppression of NiH^- from FeH^-**
  Electron Affinities:
  FeH^-: 0.934(11) eV, 1327(16) nm
  NiH^-: 0.481(7) eV, 2578(38) nm
- **Suppression of WF_5^- from HfF_5^-**
  Electron Affinities currently unknown, however theoretical calculations suggest 8.8 eV HfF_5^- for and 3.9 eV for WF_5^-.
  H. Chen, P. Andersson, A.O. Lindahl, D. Hanstorp, Chem. Phys. Lett. Vol. 511 (2011) 196-200.

Photodetachment of C^-



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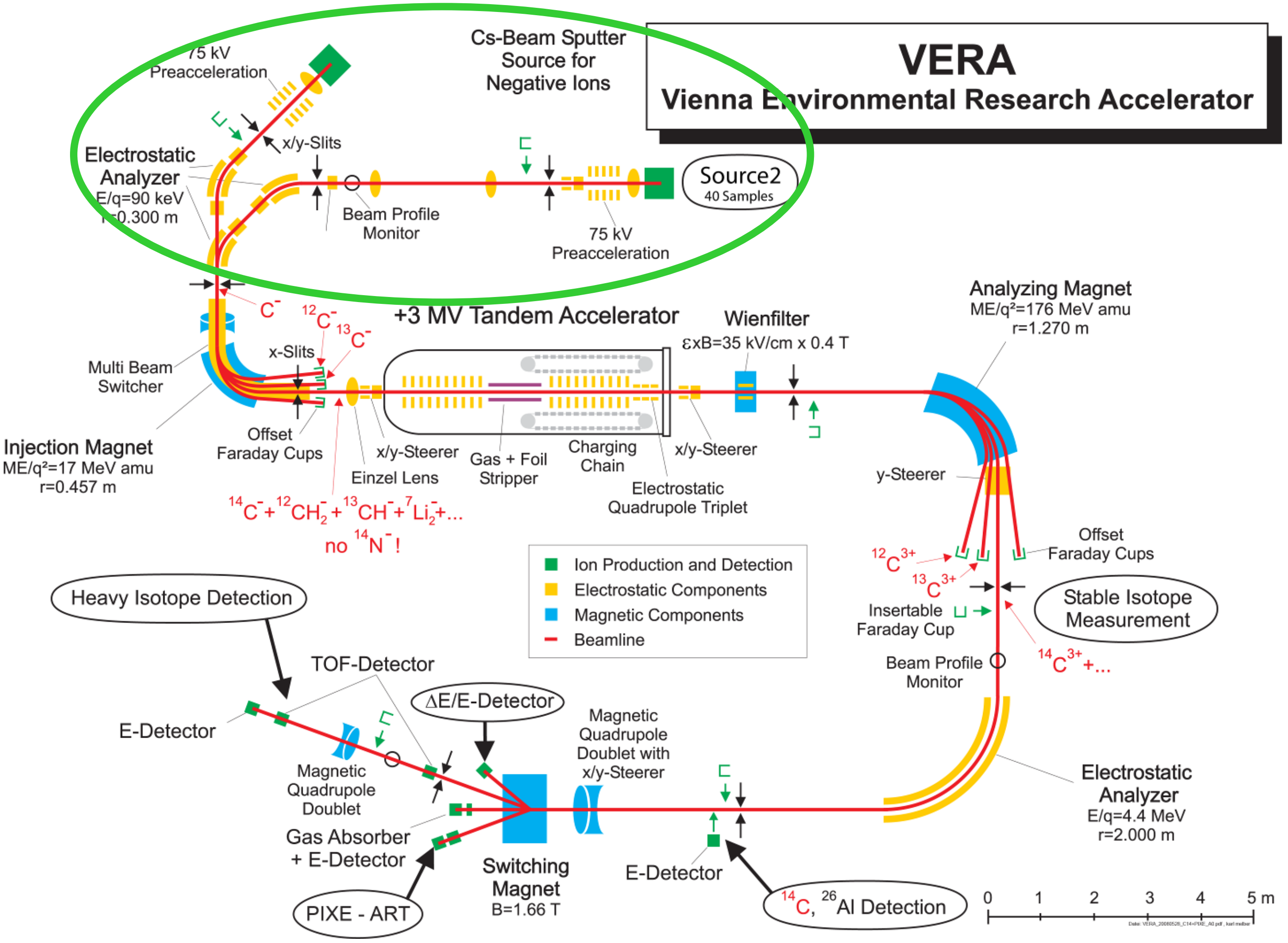
VERA Schematic

- **Actual VERA Setup including second injector**



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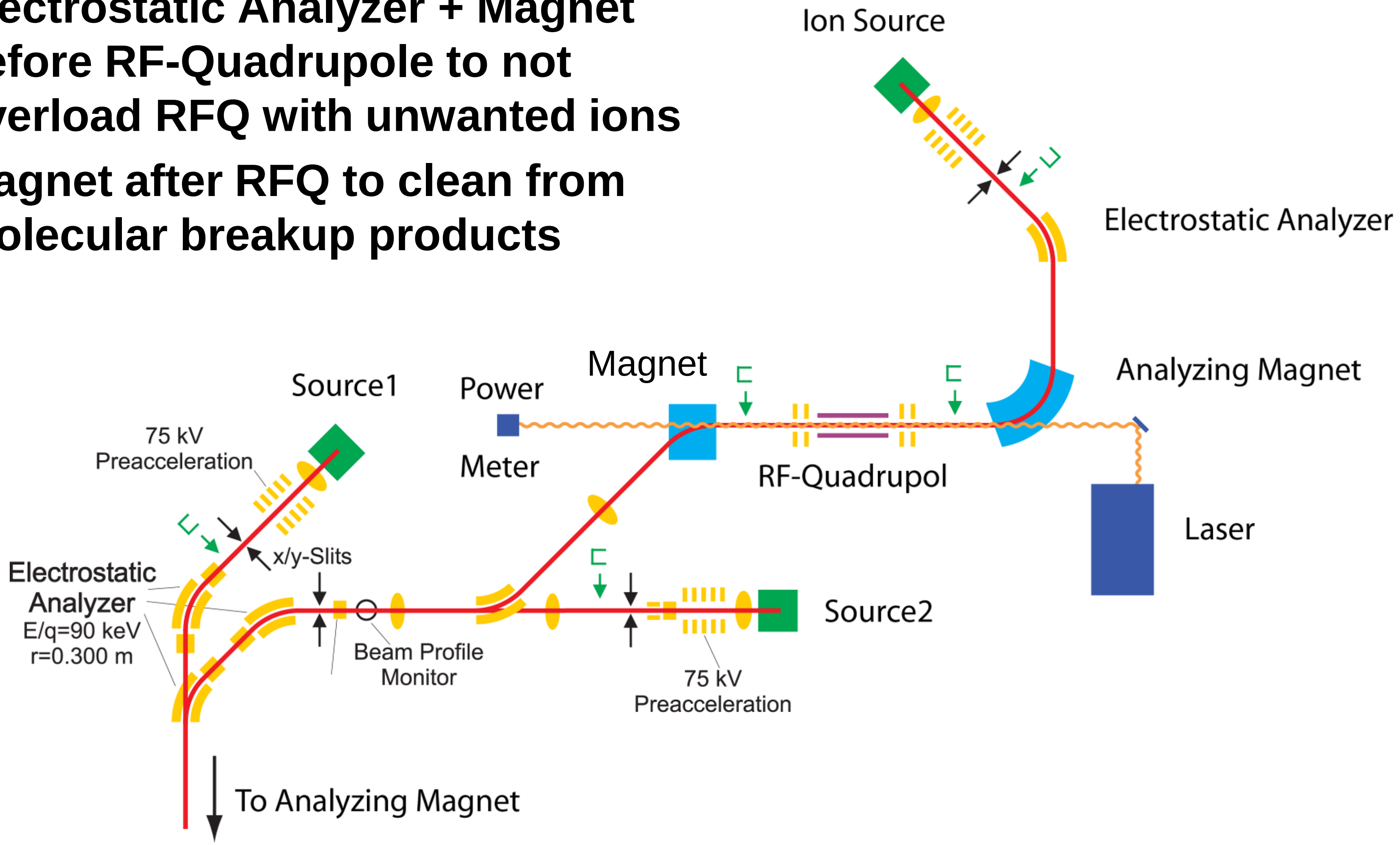
Design Study for a possible Extension of VERA

- **Electrostatic Analyzer + Magnet before RF-Quadrupole to not overload RFQ with unwanted ions**
- **Magnet after RFQ to clean from molecular breakup products**



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