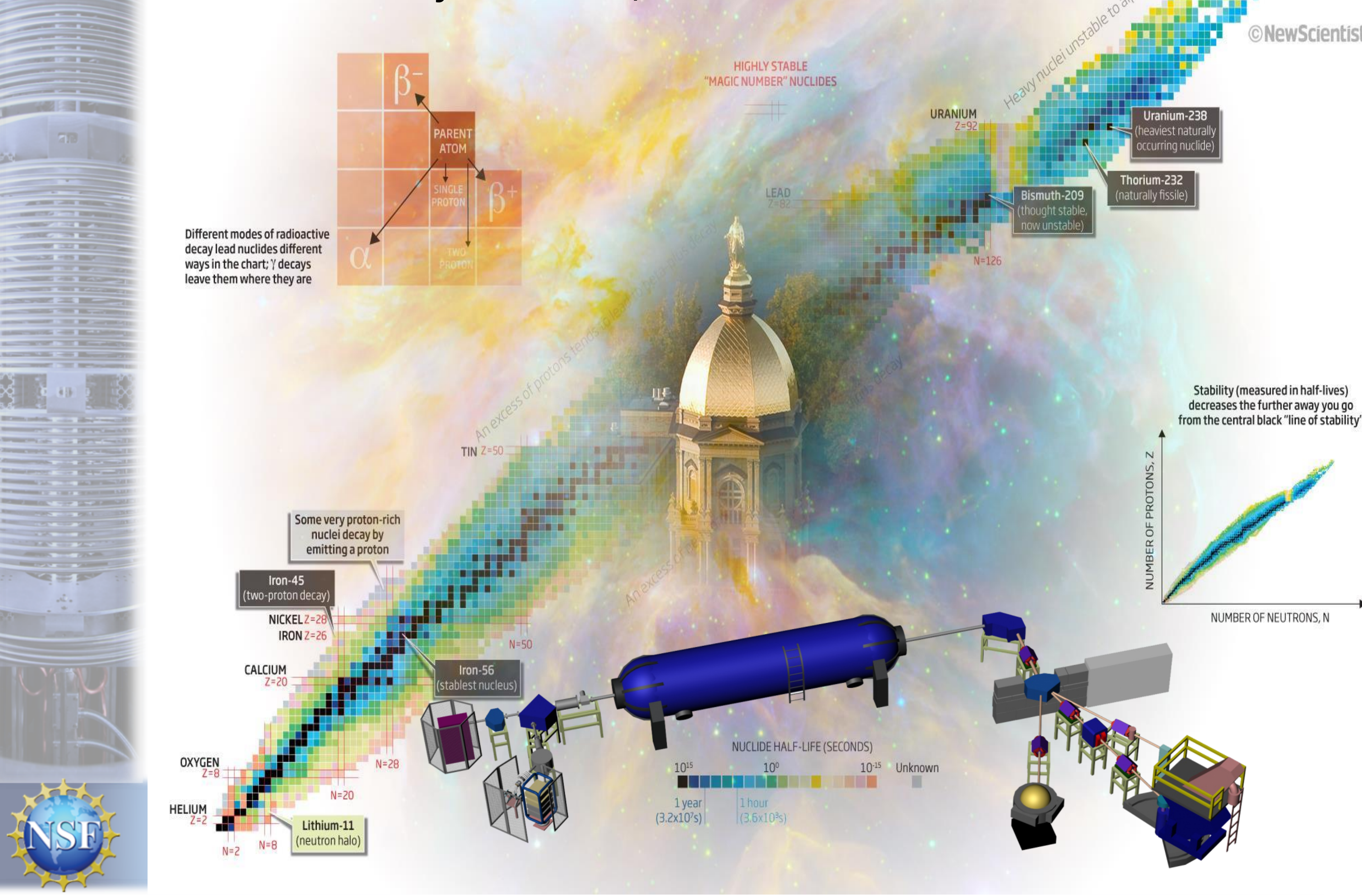
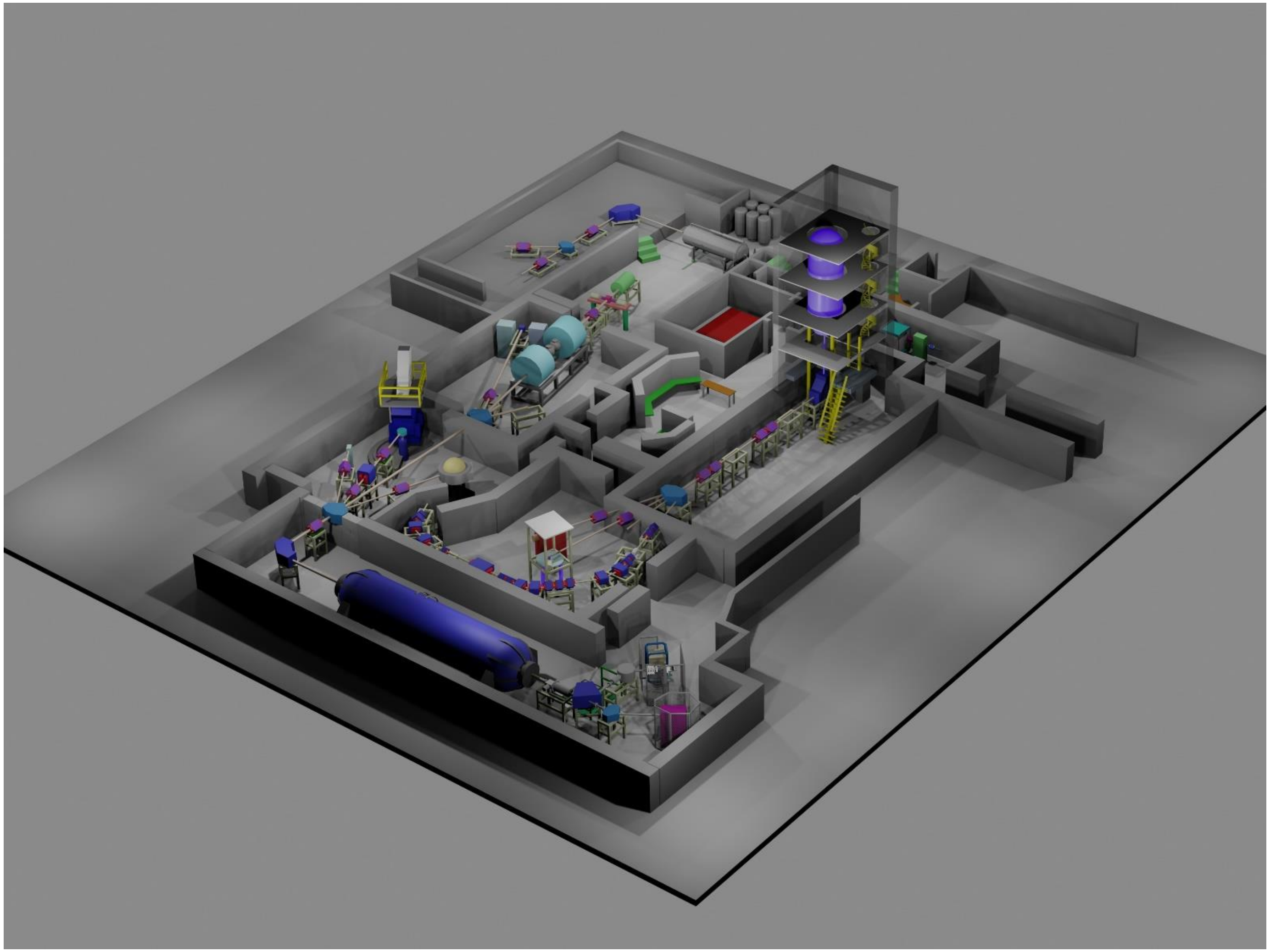
From galactic radioactivity to environmental research and archaeometry at the NSL, AMS and PIXE



Present day NSL layout



AMS orders of magnitude

Isotopic ratios from 10^{-12} to 10^{-17}

Current volume of lake Michigan: 4.9×10^{18} l

Ratio of Olympic pool in lake Michigan: 5.1×10^{-13}

Ratio of a bottle of vodka in lake Michigan: 2.1×10^{-19}
→ Vera bad idea anyway

4.2×10^{-17} corresponds to about 200l of liquid

What is AMS?

What is Accelerator Mass spectrometry (AMS)?

The determination of the concentration of a given radionuclide in a sample can be done in 2 ways:

a) Measure the radiation emitted during the decay

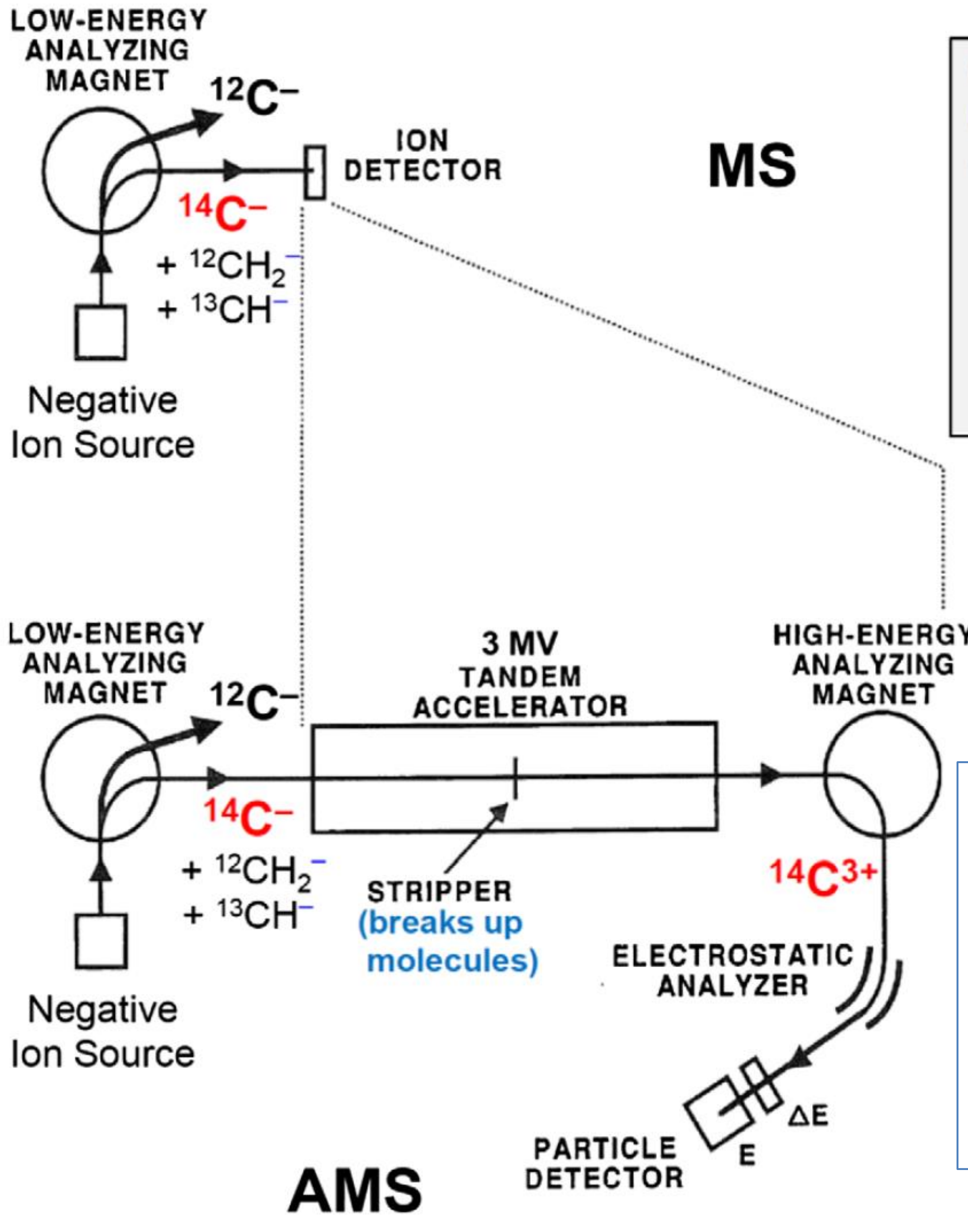
In many cases where concentrations and/or small or long $T_{1/2}$ this becomes impractical

1mg carbon ~ 6 x 10^7 at ^{14}C $\equiv$ ~1 decay/hour

b) Count the number of atoms themselves

In a Mass Spectrometer a sample material is converted to an ion beam that is then magnetically (and electrostatically) analysed

→ MS separates ions by their mass only



Composition of a negative carbon beam

$^{12}C^-$	98.9 %
$^{13}C^-$	1.1 %
$^{14}C^-$	0.000 000 000 1 %
$^{12}CH_2^-$	~0.1%
$^{13}CH^-$	~0.001%
$^{14}N^-$	0 !

An AMS measurement is generally relative:

-Stable isotope (F-cup)

- single atom counting

→ Stability of System

Comparison with traditional technique

AMS is technically more demanding than a radiocarbon dating experiment with LSC, but it is more accurate, and requires smaller samples!

If we consider a 10μg sample:

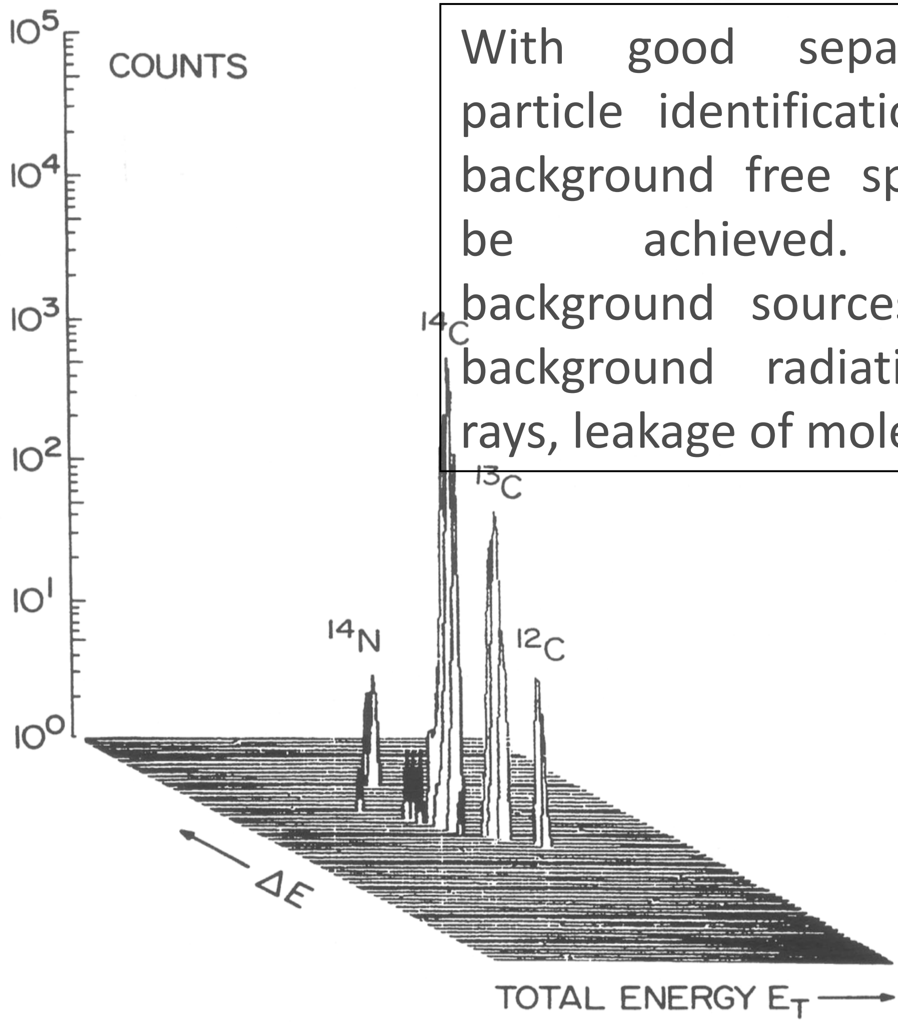
$6 \cdot 10^5$ ^{14}C particles in original sample

AMS	LSC
Assuming 1eμA beam (q=4+)	
~1000 cts/5min	~1000 cts/14y
200 cts/min	$1.4 \cdot 10^{-4}$ cts/min

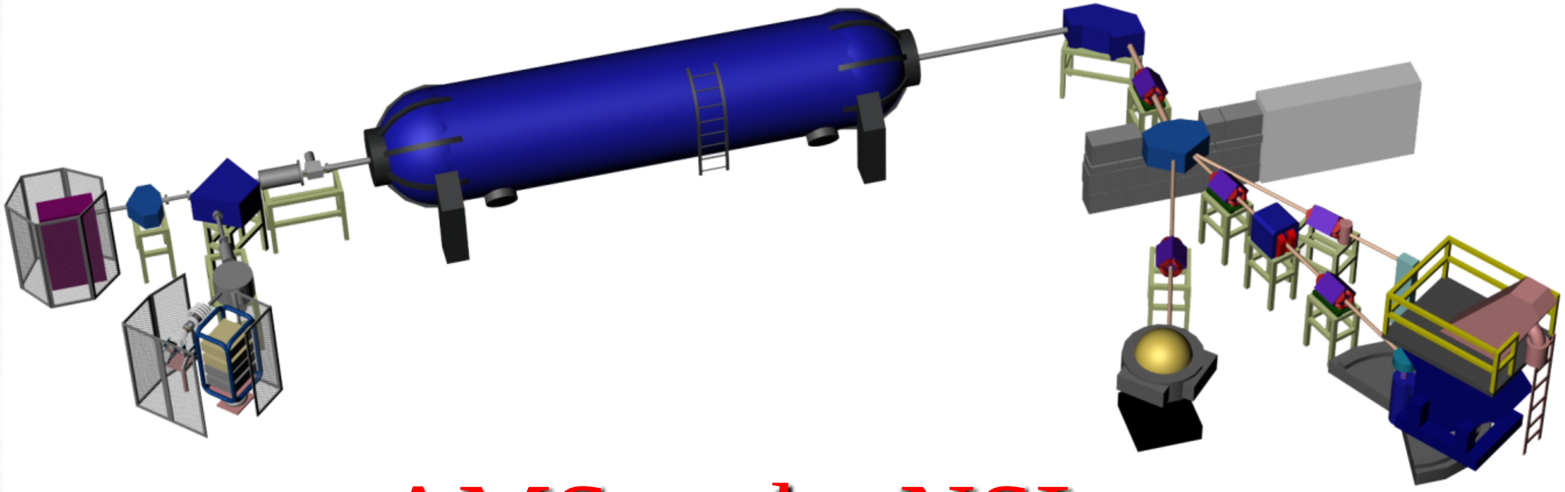
approximately 6-7 orders of magnitude improvement!!!

Separation and identification

Two-dimensional gas counter spectrum for radiocarbon ^{14}C analysis



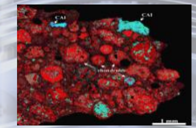
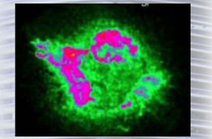
With good separation and particle identification a nearly background free spectrum can be achieved. Potential background sources are room background radiation, cosmic rays, leakage of molecules.

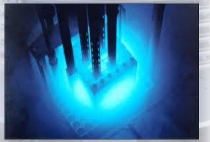


AMS at the NSL

Research focus

I S N A P


- Nuclear processes in early solar system
- Shock-front induced nucleosynthesis (galactic radioactivity)
- Measurement of cosmogenic chronometers for early solar system formation and planetary differentiation
- Measurement of ultra-low isotopic ratios
- Optimizing nuclear fuel use
- Nuclear fuel storage
- Climate research and archaeometry applications

All this would not have been possible without:

Everyone at the NSL over the years, but in particular past graduate students whose hard work made this possible:

Chris Schmitt

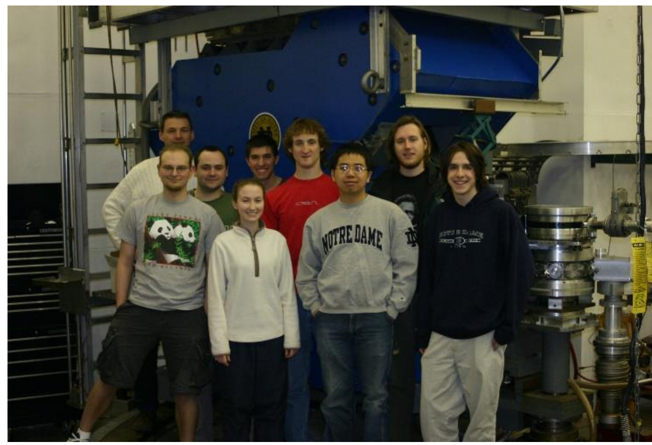
Dan Robertson

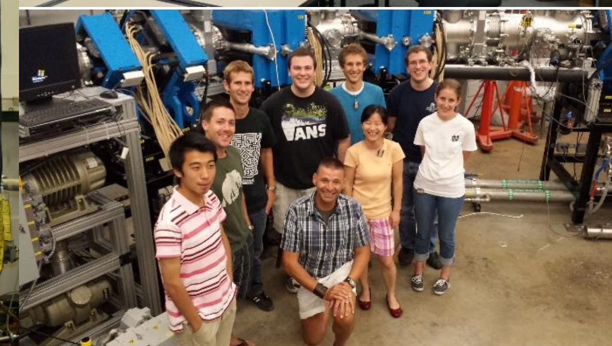
Mat Bowers

Will Bauder

Wenting Lu

Karen Ostdiek

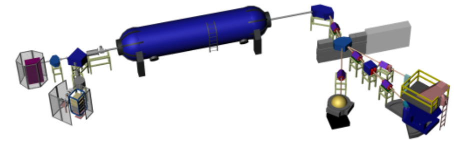


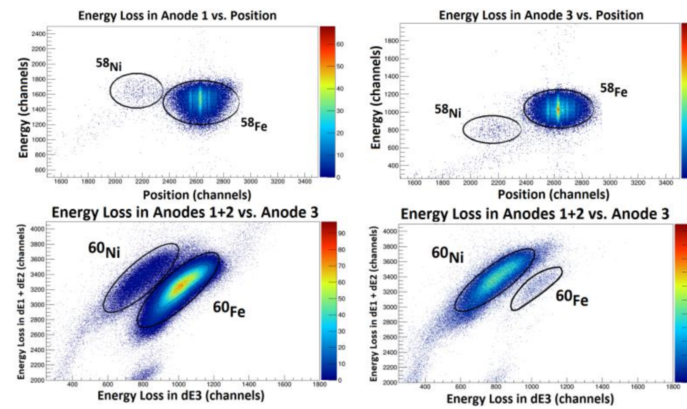


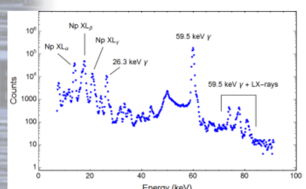
Confirming the half-life of ^{60}Fe: Using AMS and the direct decay

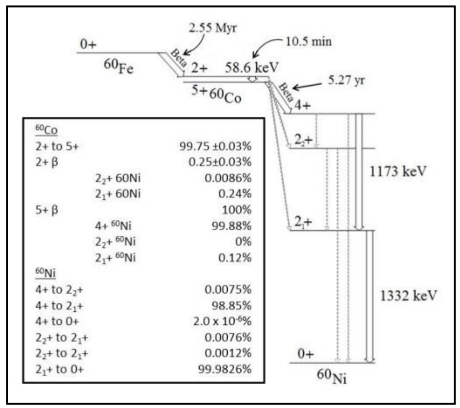
Determining the half-life of ^{60}Fe using activity and AMS

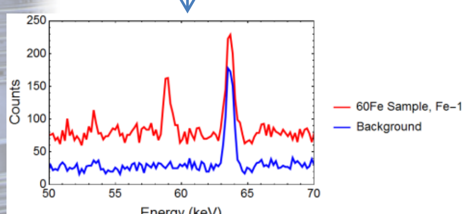
$$\frac{dN}{dt} = -\lambda N$$





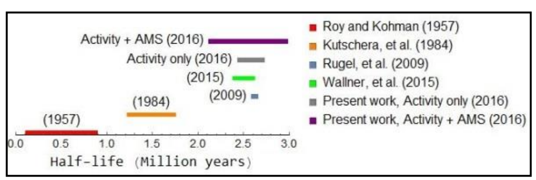






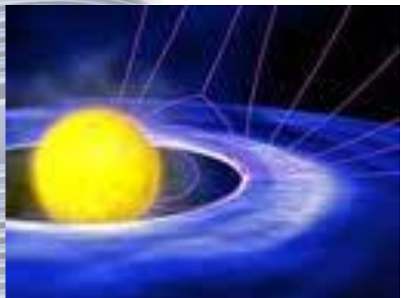
Determined isotopic ratio $^{60}Fe/^{56}Fe$ = $(2.299\pm0.387)\times10^{-6}$

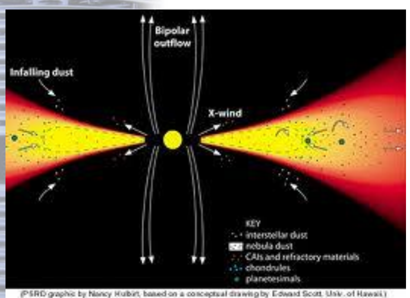
^{60}Fe atoms in sample: $(1.138\pm0.059)\times10^{15}$ atoms

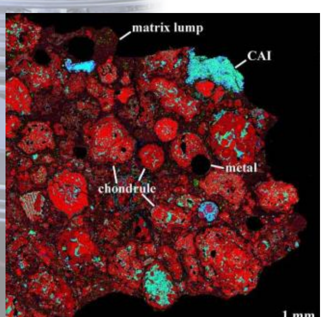


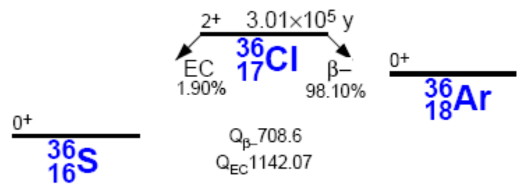
A 13.0016 ± 0.0001g sample has an activity of 9.699 ± 0.377Bq

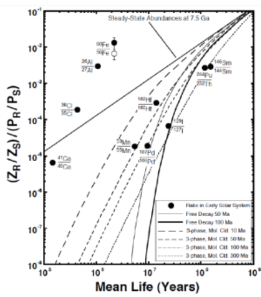
^{36}Cl for X-wind models



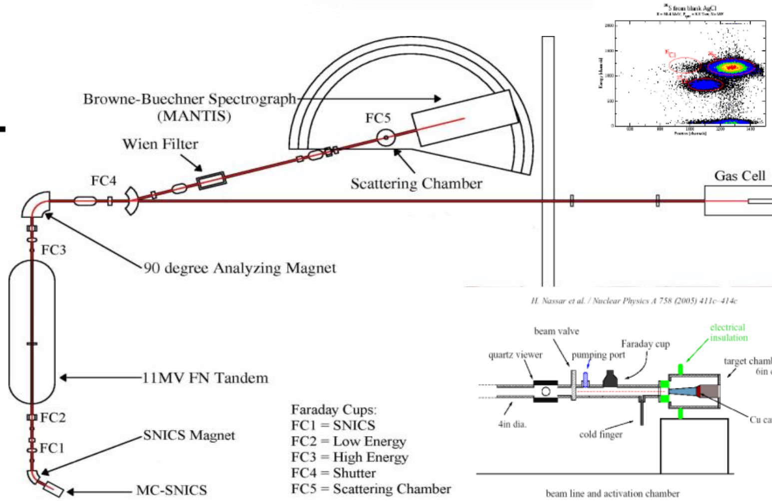


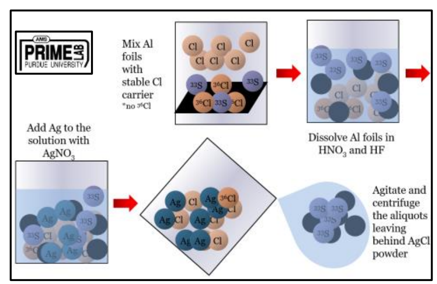


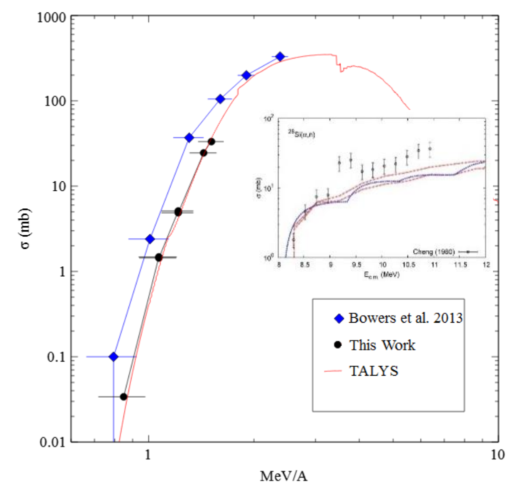




G. R. Huss, B. Meyer, G. Srinivasan, J. N. Goswami, and S. Sahijpal.
Geochim. Cosmochim. Acta, 73:4922, 2009.







Excess ^{36}S in CAIs can indicate either injection from nearby supernova or in situ production in the early solar system by the proto sun

next:: ^{34}S(^{3}He, p)^{36}Cl and ^{34}S(α, pn)^{36}Cl reactions

Helium recovery system (1-10 Torr pressure) 7.5-8MV

MANTRA and ^{39}Ar program

Improved integral 1 and 2 neutron capture cross section data for Actinides essential for GenIV reactor and advanced fuel cycle development

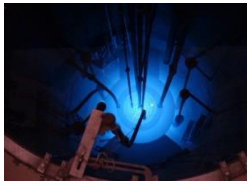
Development work at ATLAS to improve AMS facilities to improve precision and handle large number of samples:

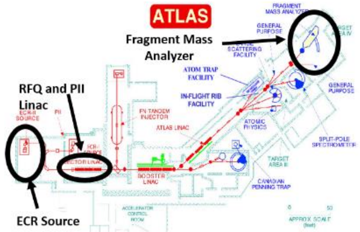
1) Laser Ablation at ECR
2) Multi-Sample Changer
3) Automated accelerator control ("Clock Program")

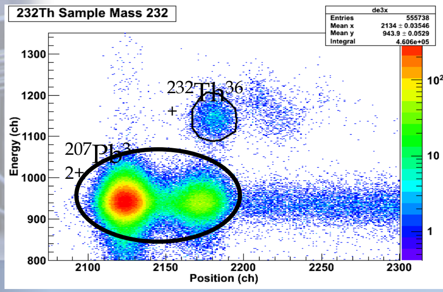
Nucleosynthesis reactions with the High-Intensity SARAF-LILIT Neutron Source:

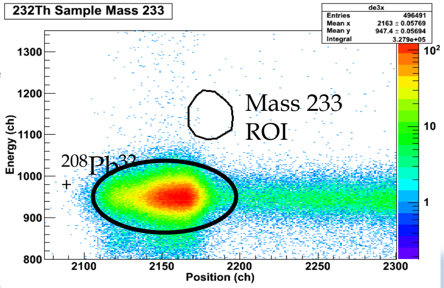
S-process flow through ^{36,38}Ar studied, in particular ^{38}Ar(n,γ)

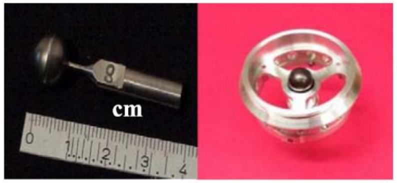
Sample activation at INL navy reactor

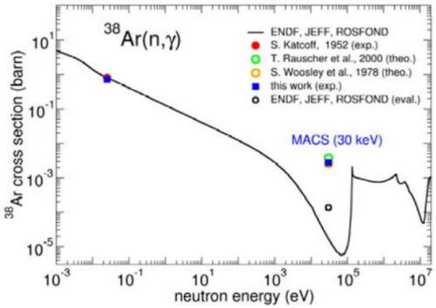


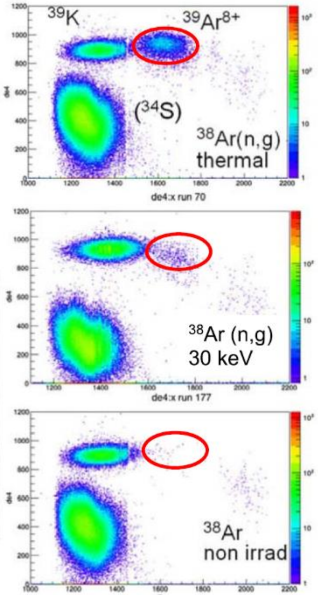






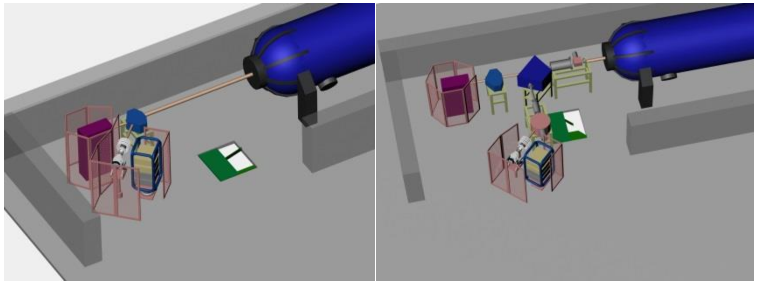




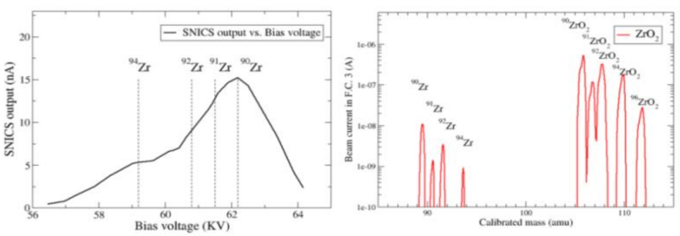


@ NSL, approval for materials, gas and foil stripper tests 3-4 MV

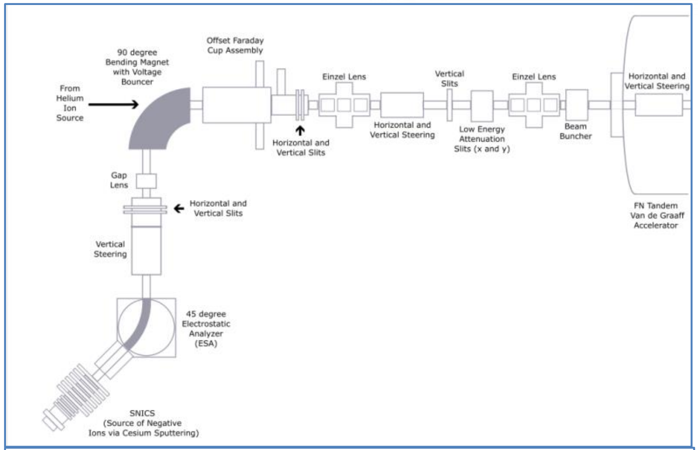
Low energy upgrade

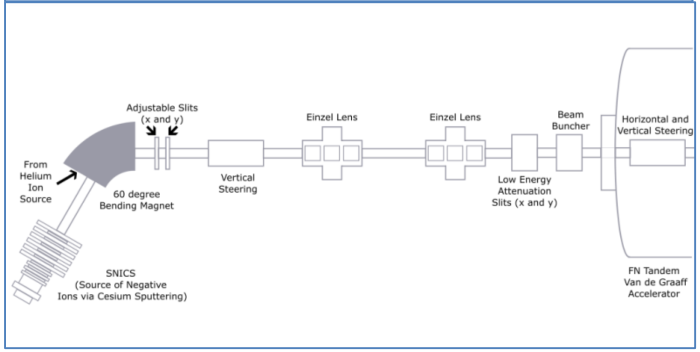


NSL | VERA



The NSL Injection Isotopic Selectivity | Proper AMS Injection Isotopic Selectivity

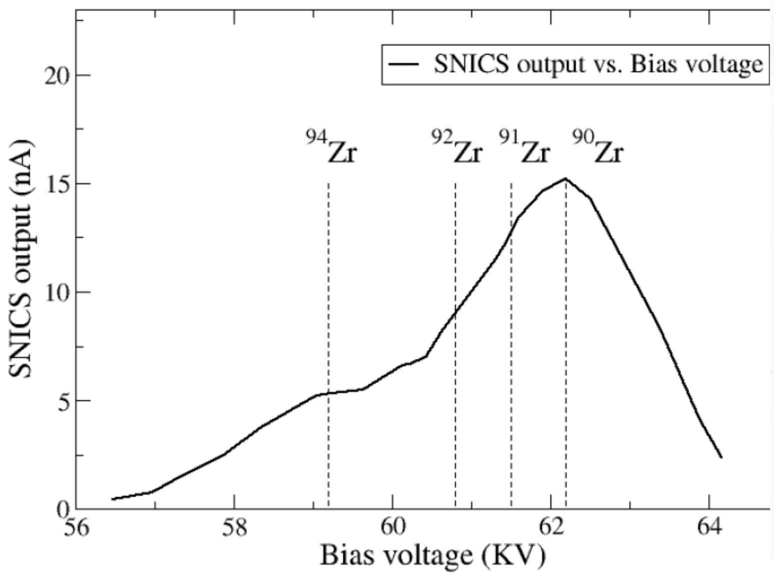




45° electrostatic analyzer, 90° double focusing injection magnet $mE/q^2 = 19$MeV amu, r = 0.475m, sequential injection, offset faraday cups, optimized injection Einzel lense.

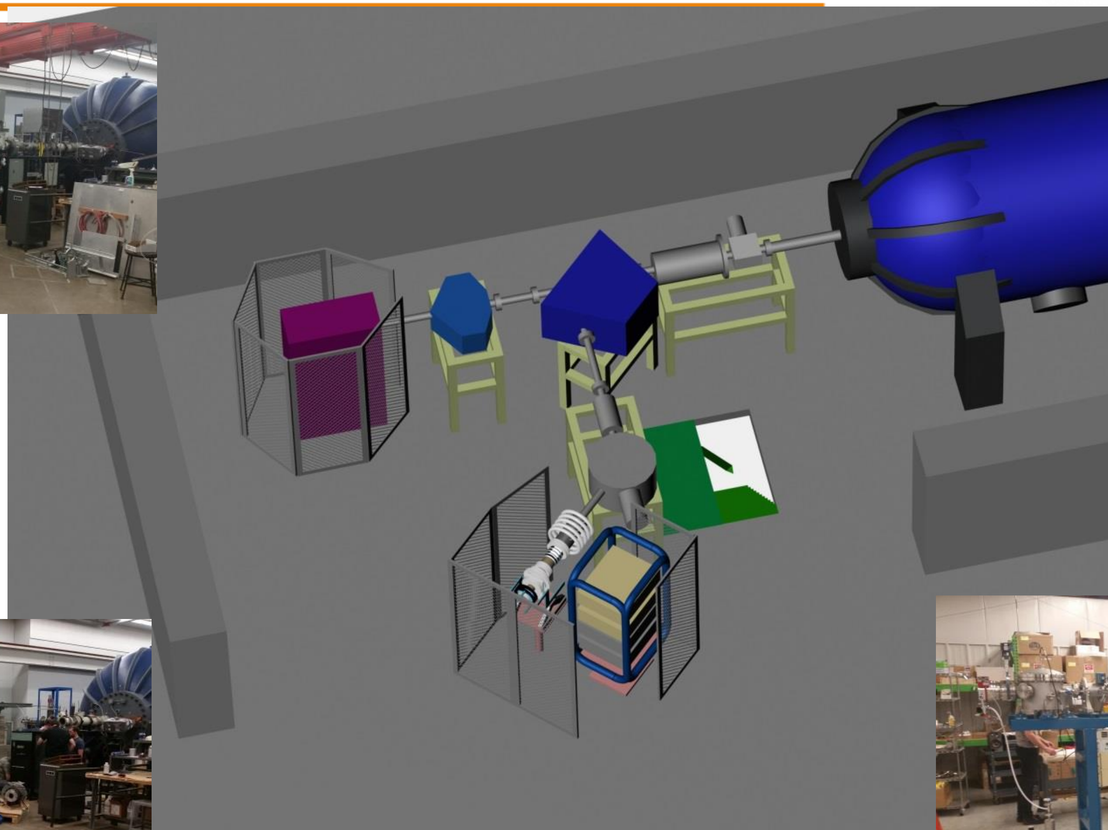
Possibility to later add HE offset cups is built-in

The NSL

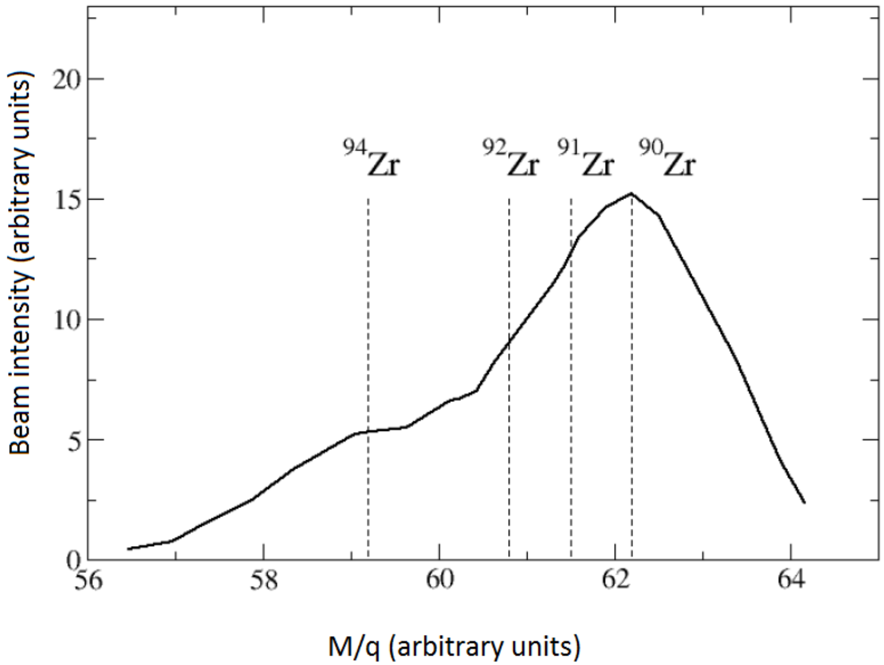


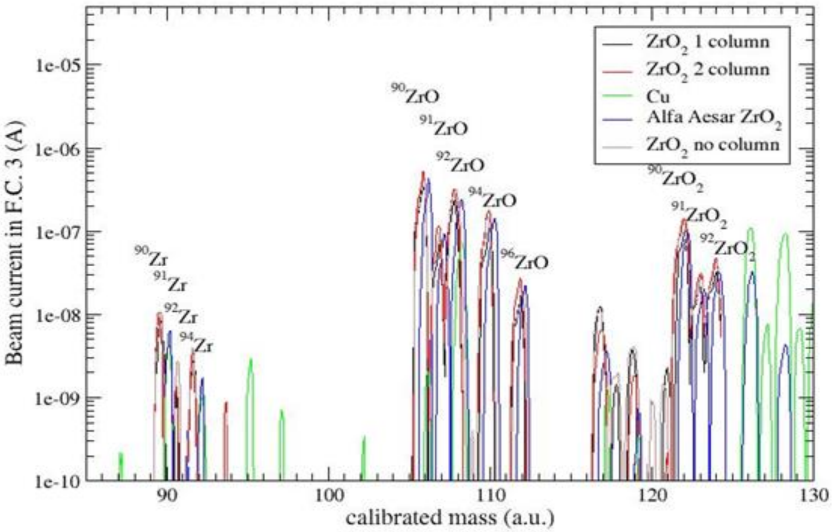
The NSL Injection Isotopic Selectivity

FN low energy injection beamline upgrade



Comparison of analyzed Zr ion beam prior to injection into a tandem accelerator. (left) NSL with low energy injection prior to upgrade. (right) Vienna Environmental Research Accelerator (VERA) with low energy system identical to the one installed as part of NSL upgrade). Both labs use identical NEC MCSNICS ion sources





ISNAP

NSF

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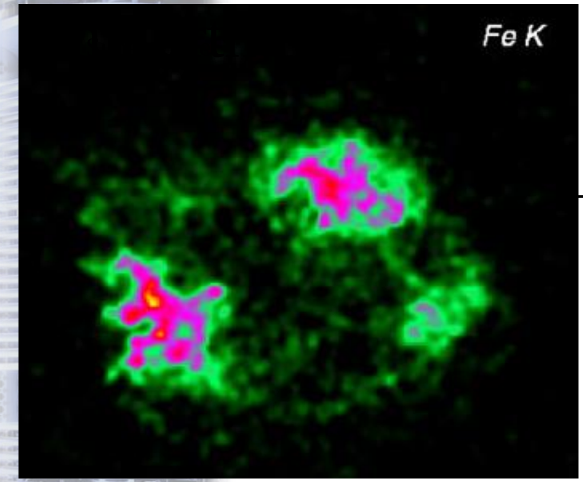
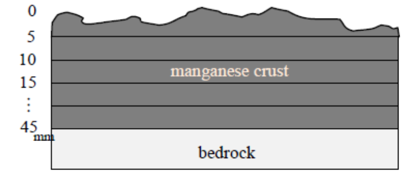
^{60}Fe as a supernova tracer in geological archives
^{93}Zr will be restarted

Nearby supernovae explosion may have influenced certain processes



Injection into the solar system

Deposition in geological formations (ferromaganese crusts)

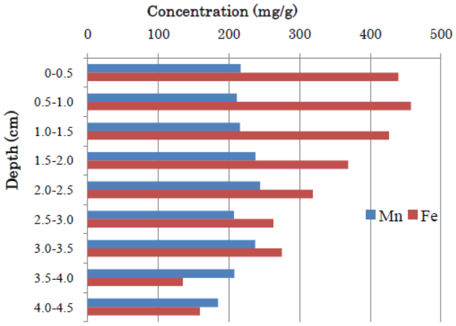


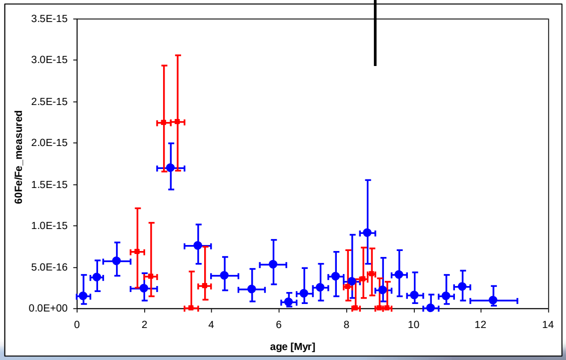
There is evidence that a nearby supernova explosion injected material into the solar system

Detection of ^{60}Fe signal in ferromaganese crusts using AMS

Depth profile of Mn and Fe

Depth (cm)	Concentration (mg/g) Mn	Fe
0-0.5	217	440
0.5-1.0	211	458
1.0-1.5	216	427
1.5-2.0	237	369
2.0-2.5	244	319
2.5-3.0	207	263
3.0-3.5	237	275
3.5-4.0	208	134
4.0-4.5	185	159





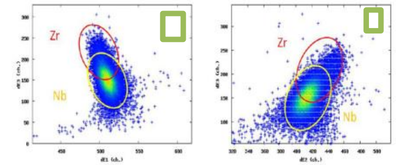
Knie et al 2003

As much Tv as possible

Nuclear forensics and Actinide AMS program

Nuclear Forensics

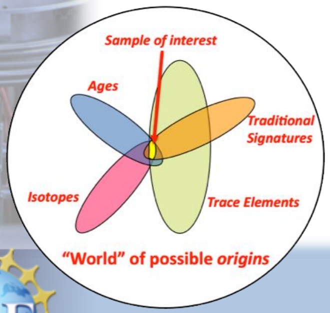
With the upgrade, higher sensitivities can be reached for AMS measurements with the same equipment post-injection, opening the possibility for measurements involving trace radionuclide concentrations, including ones of interest to nuclear forensics such as:

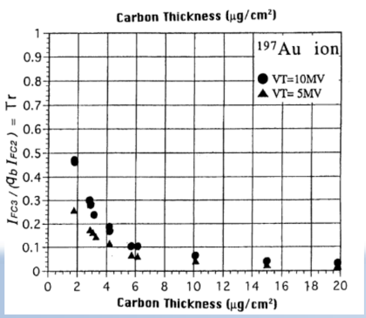


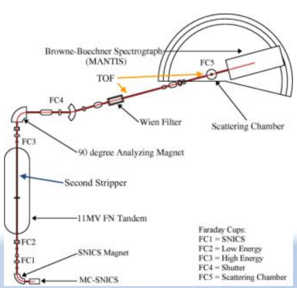
^{36}Cl, ^{41}Ca, ^{129}I (mentioned previously)
^{93}Zr, ^{90}Sr - an isotope that is highly enriched in spent nuclear fuel

AMS with actinides in particular has a number of applications in nuclear forensics, nuclear security, utilizing the nuclear fuel cycle, environmental studies, nuclear astrophysics, etc.
A few actinides of particular interest to AMS studies include but are not limited to:

- ^{230}Th used for the search for superheavy elements in nature
- ^{236}U and ^{238}U which appear in much larger ratios from nuclear reactions compared to weapons fallout
- ^{237}Np which can be an indicator of nuclear reprocessing and used for studying ocean circulation
- ^{240}Pu and ^{239}Pu serve as a fingerprint for the origin of the plutonium and ^{244}Pu is of great interest for Early Solar system models .





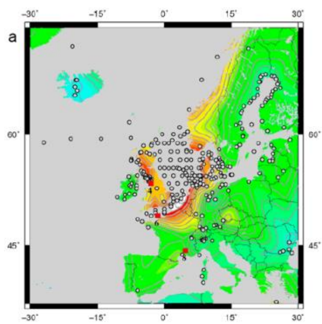


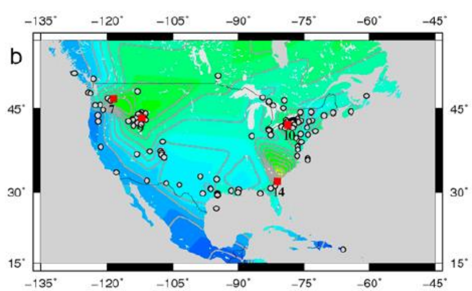
Impact for the NSL:
Beyond the scope of nuclear forensics, there are many studies to be done with the actinides, including nuclear data, nuclear astrophysics, and nuclear structure physics. The success of this work will provide an added capability at the NSL of the University of Notre Dame that others can also benefit from or exploit.

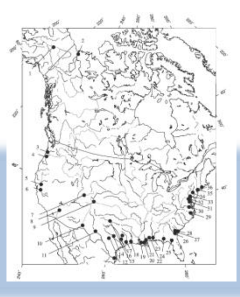
^{129}I for nuclear forensics and environmental science

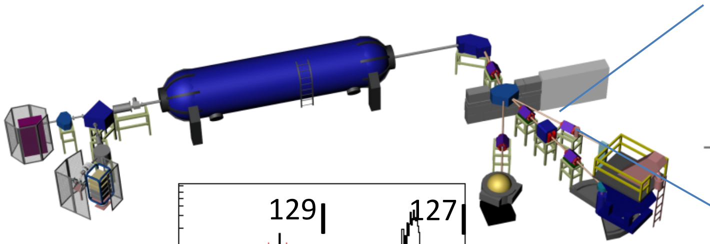
Environmental contamination from ^{129}I highlighting a need for global real-time measurement as a preventative and observational method for detecting nuclear activity

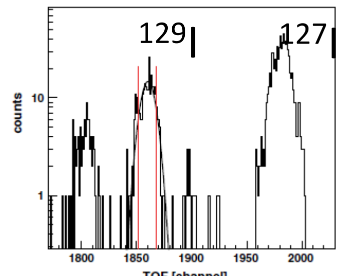
Snyder, G., A. Aldahan, and G. Possnert (2010), Global distribution and long-term fate of anthropogenic ^{129}I in marine and surface water reservoirs, Geochem. Geophys. Geosyst., 11, Q04010,

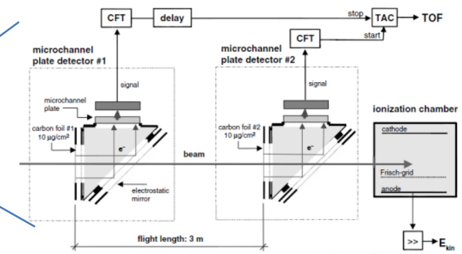


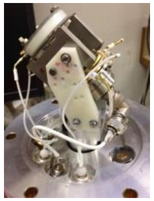


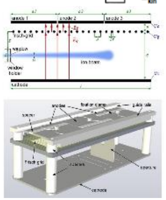












With the upgrade, higher sensitivities can be reached for AMS measurements with the same equipment post-injection, opening the possibility for measurements involving trace radionuclide concentrations, including ones of interest to nuclear forensics such as

^{129}I - an environmental contaminant from fuel reprocessing that can be used as a reference for detecting increased nuclear activity, made possible with the installation of a time-of-flight (TOF) system

Installation of TOF section in AMS beamline

~7MV

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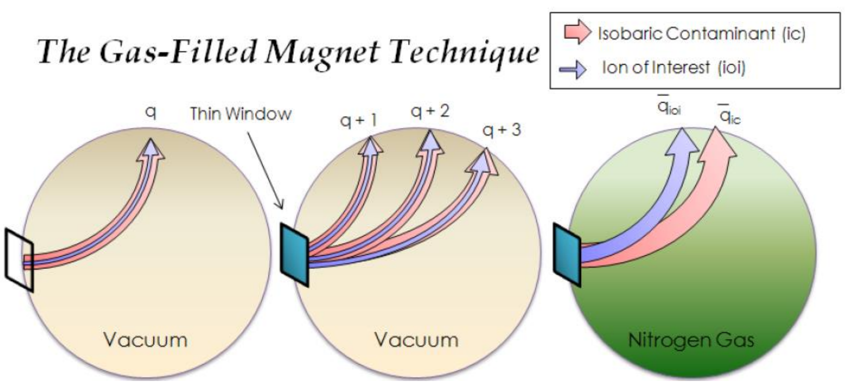
^{41}Ca and ^{53}Mn AMS programs

Motivation

One of the best detection methods for long-lived radioisotopes is with the technique of Accelerator Mass Spectrometry (AMS).

^{41}Ca ($T_{1/2}$ = 1.03x10^{5} years), therefore one of the main viable detection methods is through AMS techniques. ^{41}Ca has shown promise as a chosmochronometer and environmental tracer.

Cosmogenic ^{53}Mn ($T_{1/2}$ = 3.7x10^{6} years) produced on Fe. Currently only 2 facilities provide ^{53}Mn measurements. Applications as geological dating tracer for exposure, erosion and burial dating. Has potential to be proxy to measure long-term Cosmic-ray variations.



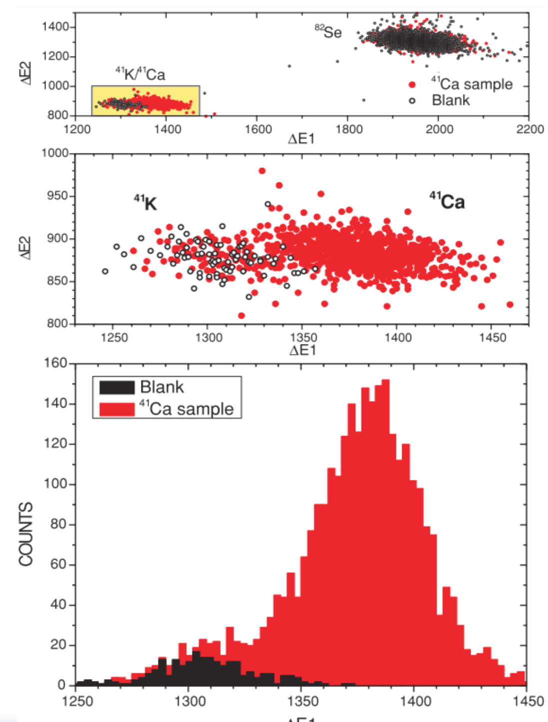
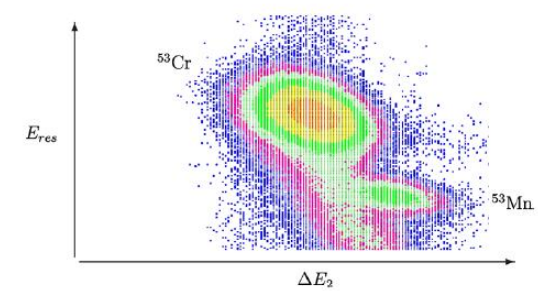


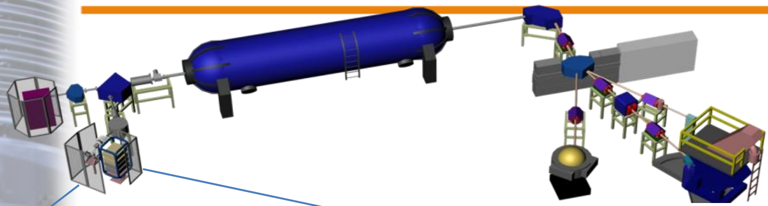
Fig. 2. A plot of E_{res} versus ΔE_2 obtained from the gas-ionization detector for a ANU standard at $\approx 1.8\times10^{-9}$. The ^{53}Mn events, lower right, have a higher ΔE_2 and lower E_{res} compared to ^{53}Cr, middle of plot (z-axis is a log scale).



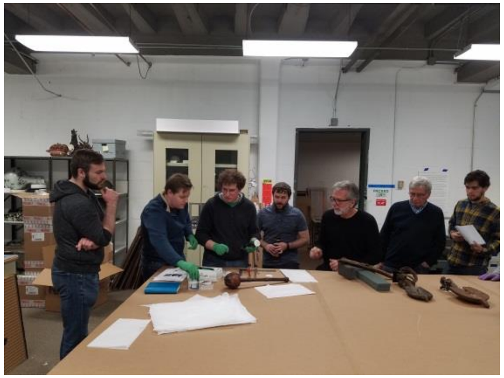
Cathode tests

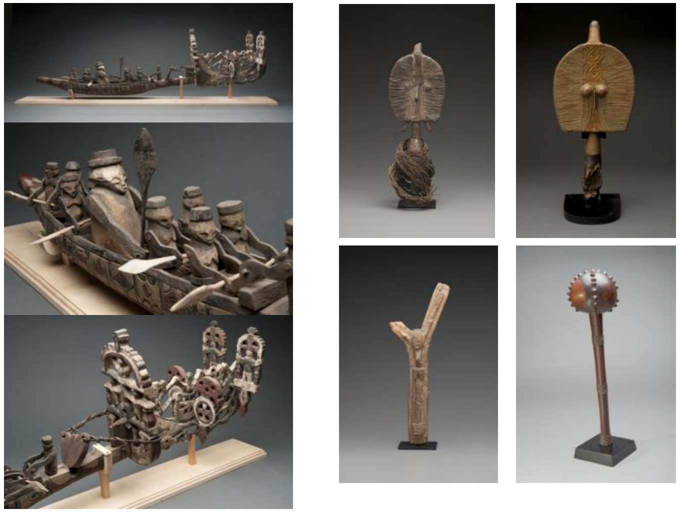
8MV

Radiocarbon AMS program: from tracing glacier runoff to dating African art



African art collection of the Snite museum of art: Determining possible ages.





Wetlands of the Copper River Delta in Alaska: melting glaciers feed the Copper River and introduce ancient carbon into the river delta. Insects feeding off the runoff may provide insight into the effects of the glacial runoff on the local ecosystem.

Graphitization is now running smoothly: up to 30eμA $^{14}C^-$ HE offset cup, improved TV stabilization, cathode centration

Central to AMS

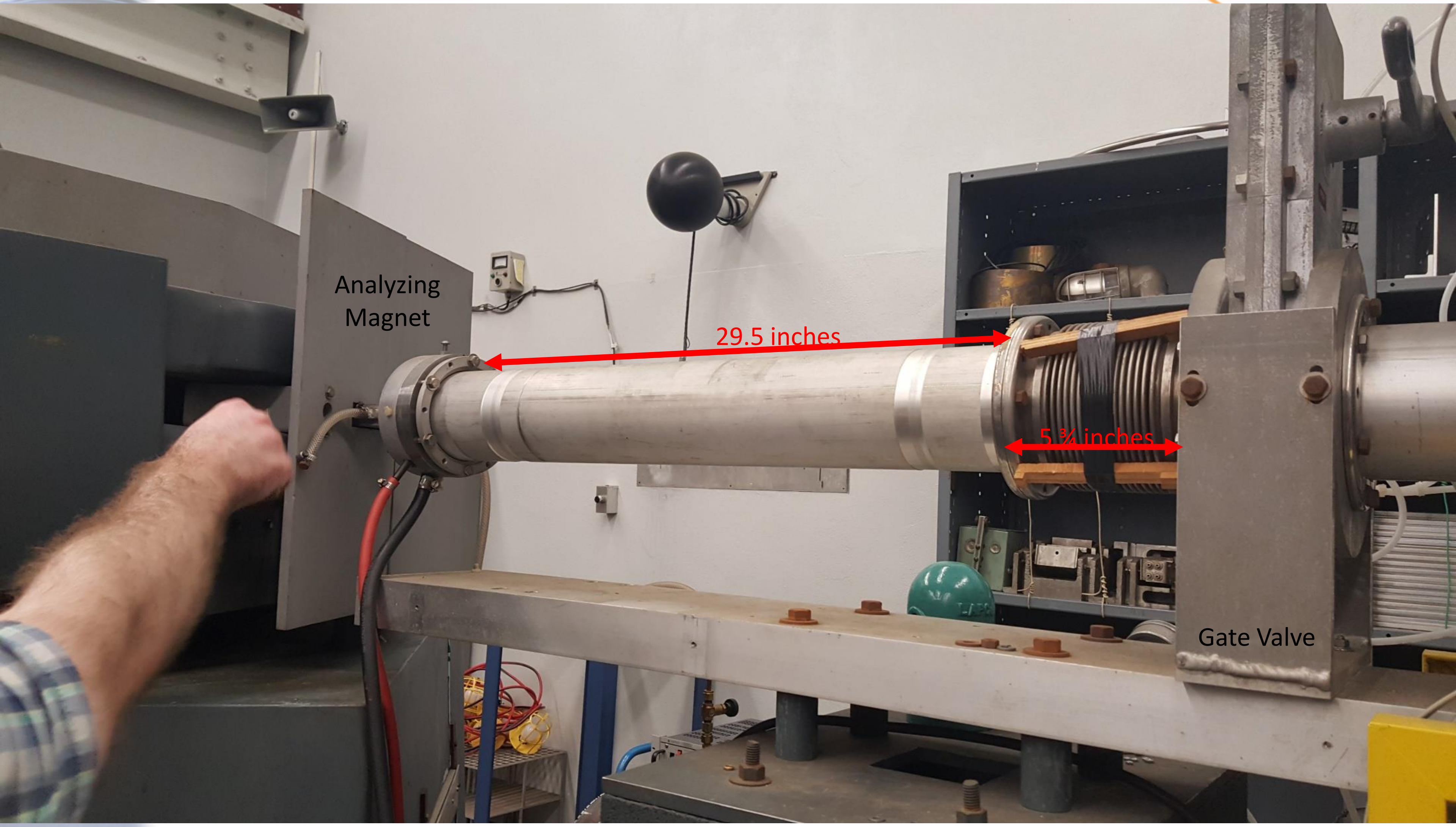
AMS depends on a well controlled accelerator as regular tune switching is essential to any AMS measurement.

- Measurements of radionuclides are done on a BLIND TUNE
- Measurements are RELATIVE in most cases and STABILIY and REPRODUCIBILITY is CENTRAL

AMS measurement:

1 Tune of PILOT beam
2 M/Q adjusted blind tune to radioisotope
3 Repeated measurements of stable isotope(s)
@ offset F-cups
4 Measurements of a radioisotope standard

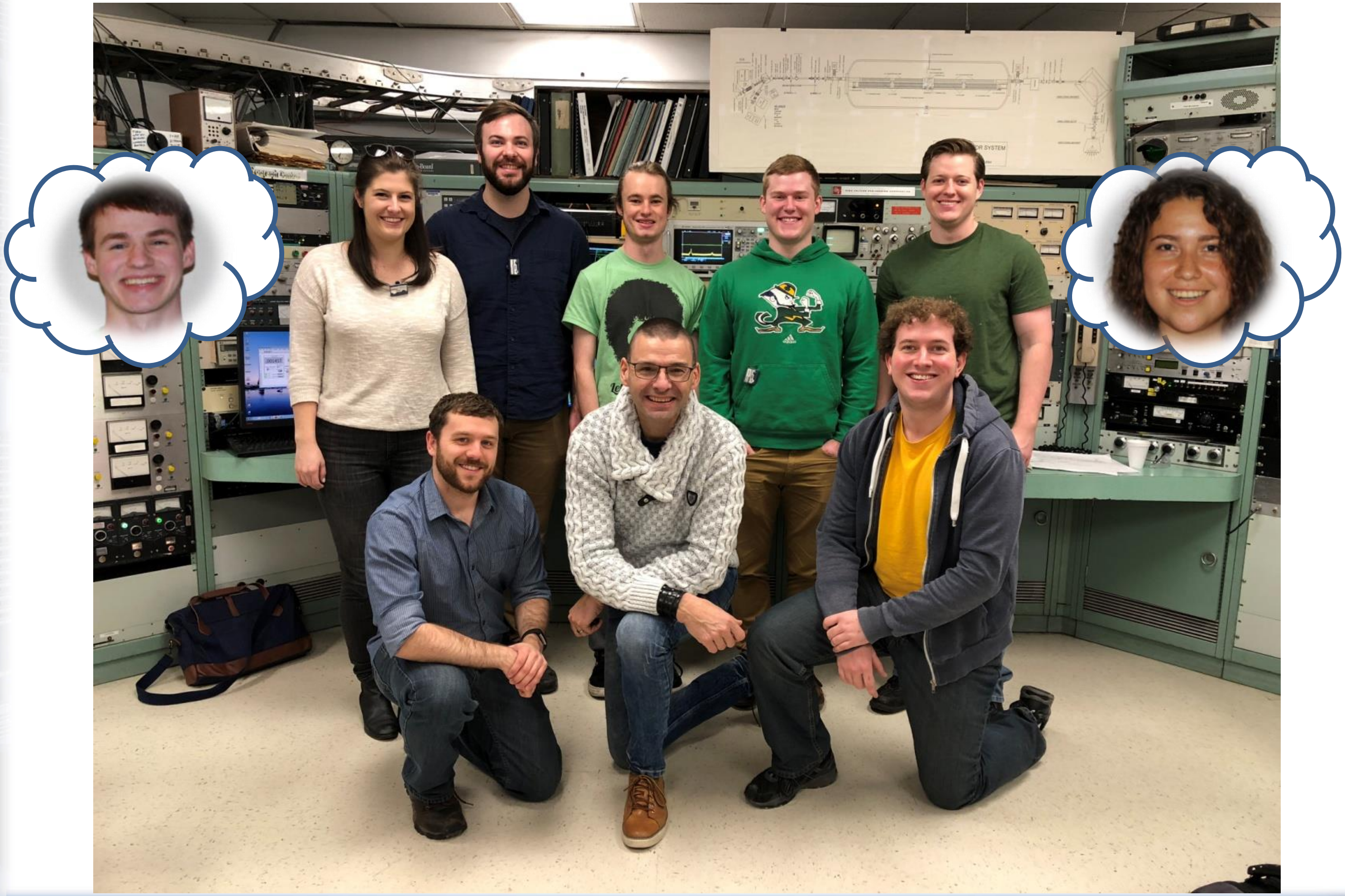
→2 and 3 require multiple switching between tunes
→measurements to a standard requires TV stability


Analyzing
Magnet
29.5 inches
5 ¾ inches
Gate Valve

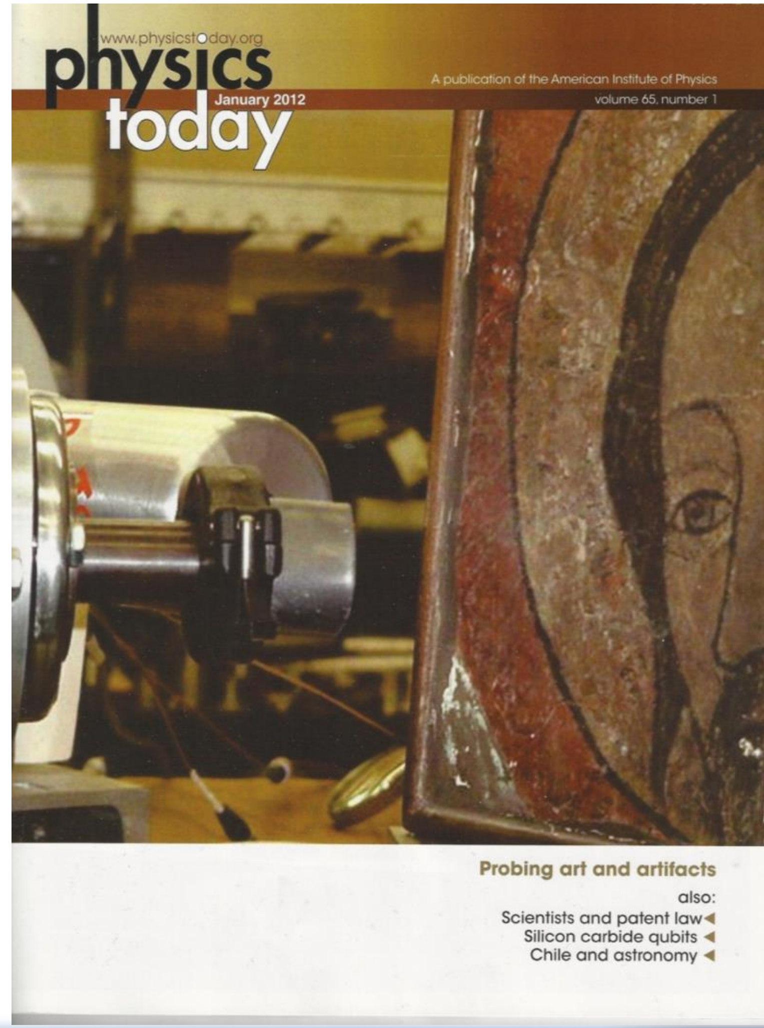
Accelerator "upgrade" and developments

- TOF line in AMS beamline after second quad
- Actinides:
  - Thinking about dedicated ionizer
  - Development of "different" stripper foils
  - Improvements to the gas stripping
    - Using He
    - Laser or optical fibers
    - Pumping to HE > pumping to LE (reduce recombination)
- ^{14}C:
  - Development of a HE offset F-cup (using old ATLAS magnet)
  - Automate cathode centration
- FN:
  - Currently many controls are analog. Digital control would help us
    - Terminal, magnets, AG lenses
  - Improve TV
  - Developing a "fine tuning" program that will refine the tune once a tune is found.

People



How to get Christ on the first page of Physics today?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volume 65, number 1
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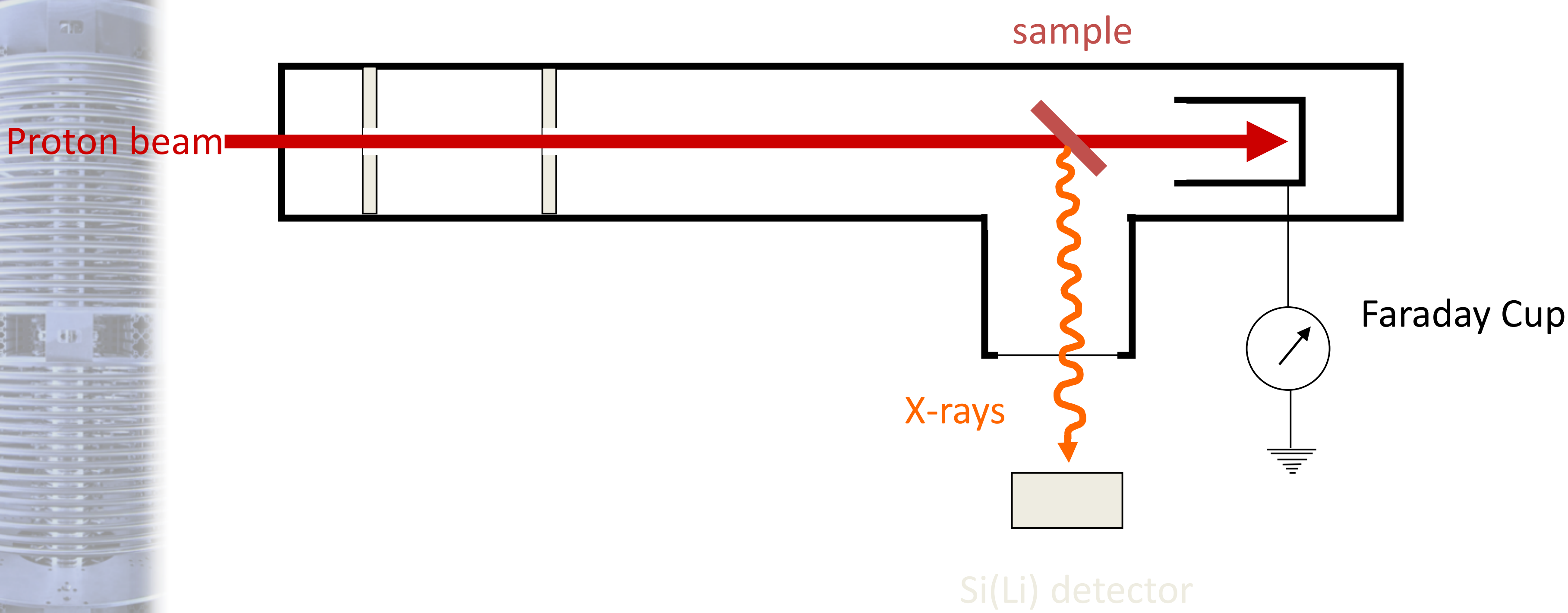
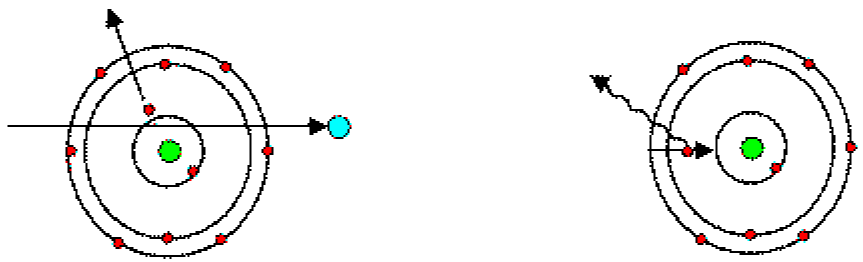
Silicon carbide qubits
Chile and astronomy

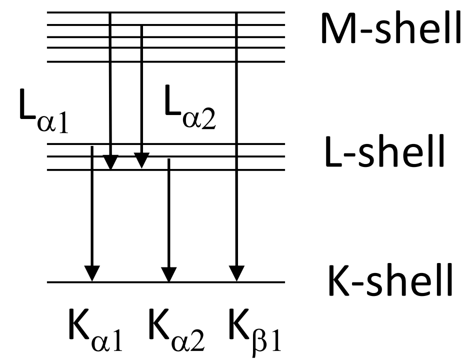
The PIXE arrangements

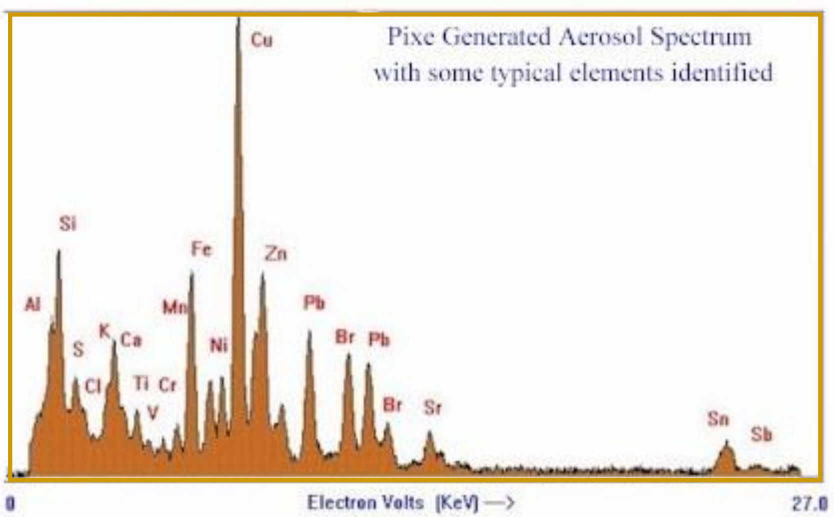


Typically proton beams; protons transmit energy to the inner-shell electrons, ionize atoms with subsequent X-ray de-excitation

PIXE Spectrum







$E_x = E\,(n,l)_i - E\,(n,l)_f$

Same energies as in the previous section, only the electron excitation mechanism is different!

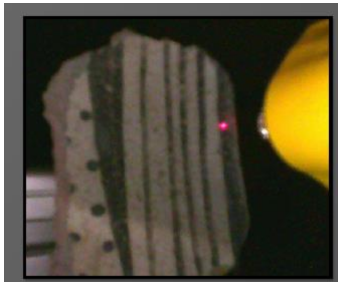
PIXE analysis of Mesa Verde black-on white paints

Compositional analyses of ceramics in the U.S. Southwest have primarily focused on the clays and tempers used to make pots. Less attention has been directed toward determining the elemental composition of the materials used to decorate pots. PIXE was used for a compositional analysis on the black paints used on Mesa Verde Black-on-white bowls from Aztec West in Aztec Ruin National Monument, New Mexico.

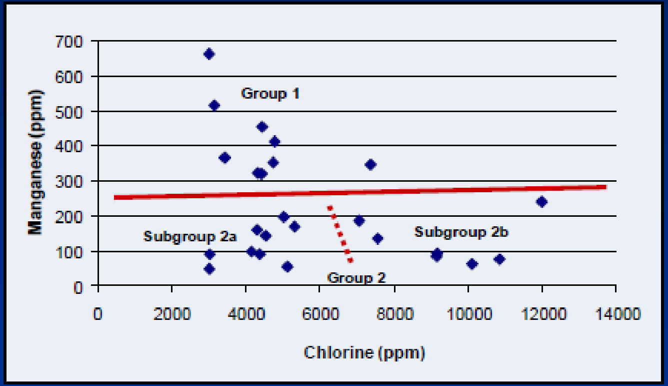
Mesa Verde Black-on-white bowls from Aztec West

Structural Remains at Aztec West

Detector Set-up at ISNAP



First Paint Shot (Sherd 1082)



Bivariate plot of Mn vs Cl showing hierarchical groupings

Andrew R. Steier[1], Donna M. Glowacki[1], Michael Perry[1] and Edward J. Stech[2]

[1] Department of Anthropology, [2] Department of Physics University of Notre Dame

Paint recipe analysis and classification of paints

45 Paints and slips as well as plants were used by ancient pueblo potters from Aztec Ruins National Monument in Mesa Verda, NM

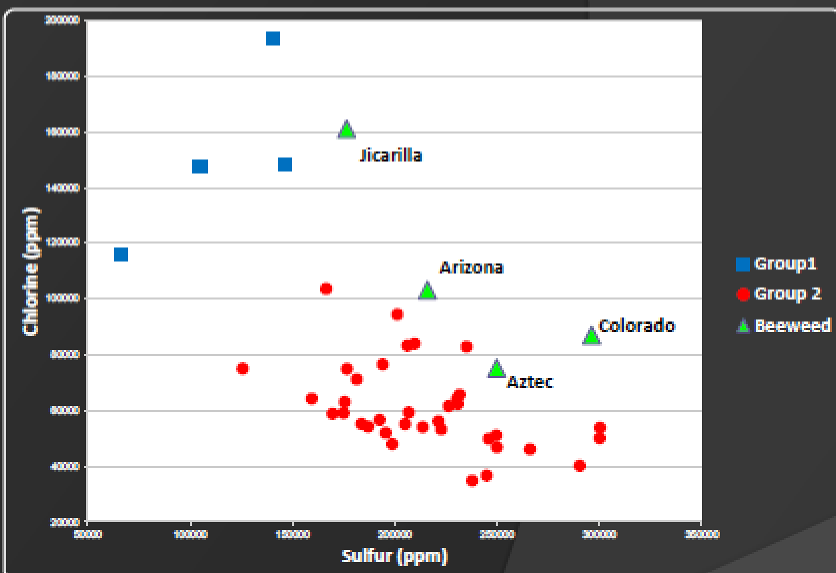
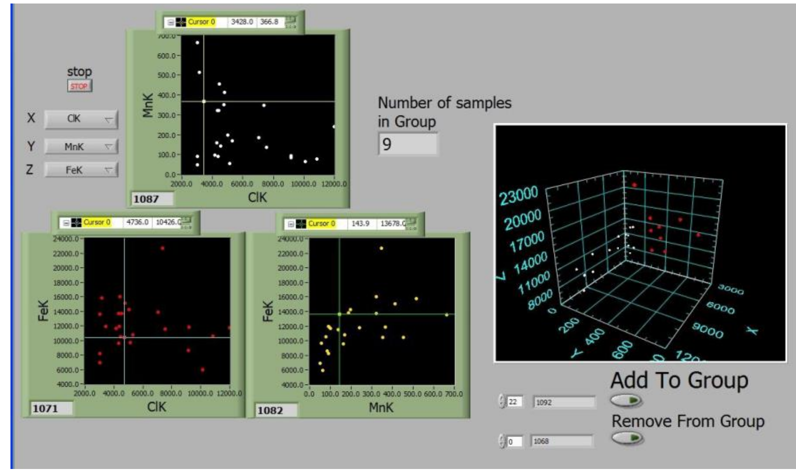


Figure 3: Bivariate plot of Chlorine and Sulfur showing separation of Groups 1 and 2 and the correlation between Group 1 and Jicarilla beeweed sample



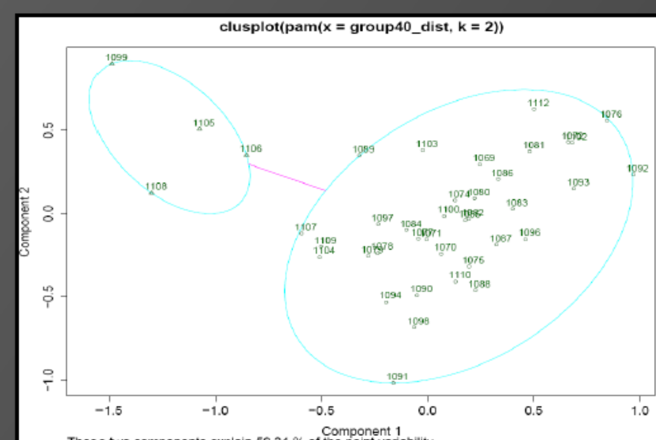


Figure 1: PAM analysis of 40 samples distinguishing Group 1 (n=4) from Group 2 (n=36)

Work done by Anthropology Undergraduate students

Native American and European Copper

Copper made objects have been found in 1400-1500 AD ‘pre-colonization’ settlements along the Ohio River valley. PIXE analysis of the relative Ag, Sb and Pb content revealed origin from native sources (A) or European (trade) sources (B)