Searches for the ^{229}Th isomer half-life and studies of the ^{235m}U isomer

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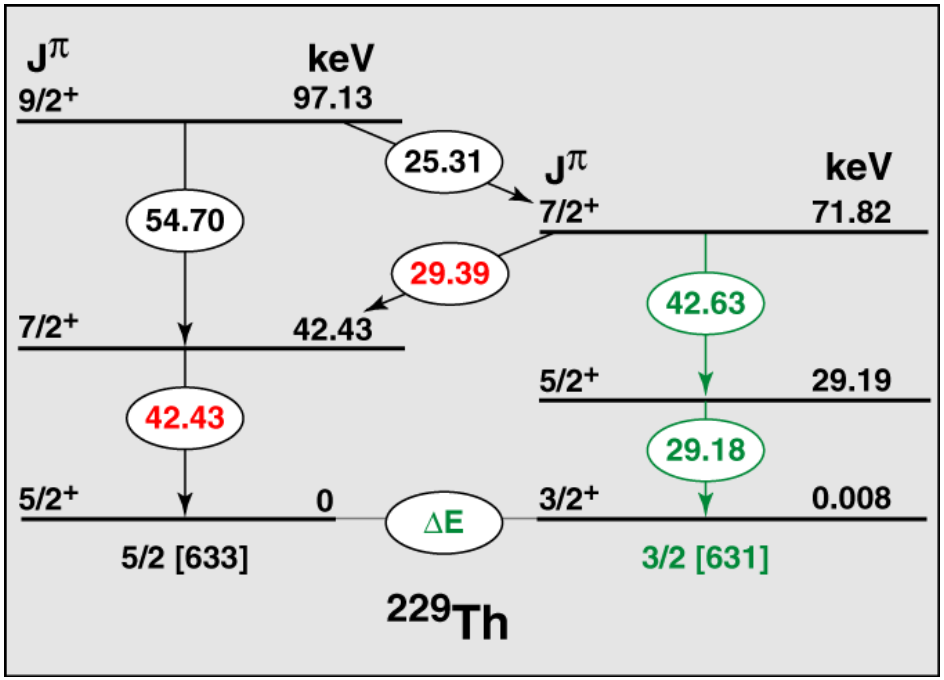
Lawrence Livermore National Laboratory

Overview

- Background and History of ^{229m}Th
- Theoretical half life predictions
- Experimental efforts and complications
- Results
- Future efforts (very near future)

Low-lying ^{229}Th partial level scheme after ^{233}U decay – characterized by 2 non-interacting rotational bands*

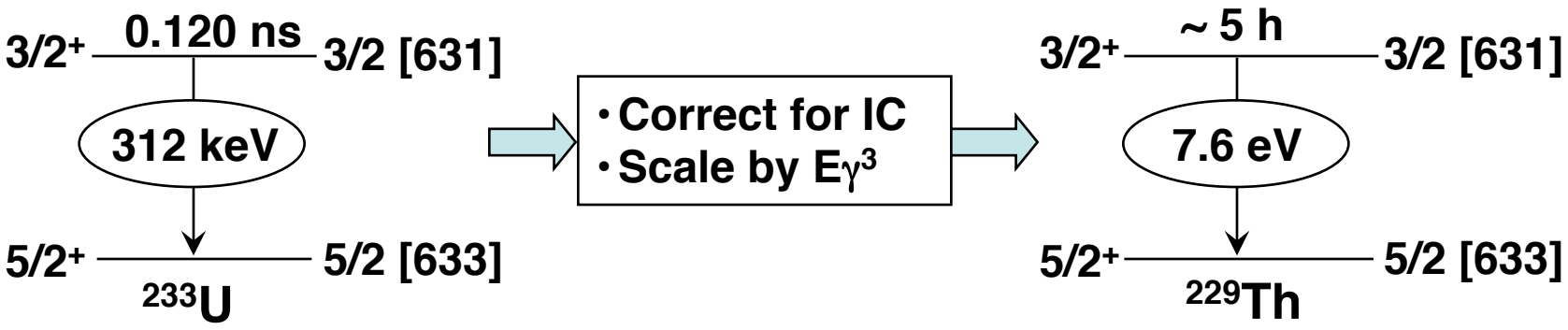


Two closely spaced with bandheads within ~ 5 - 7 eV
- **How small is ΔE ?**
- **Can you excite upper level with laser?**

*** Pioneered by Helmer, Reich, *et al.***

What is lifetime of $3/2^+$ state?

- Scale from analogous transition in ^{233}U with known lifetime
  - Following Helmer and Reich*



Estimate T(1/2) ~ 5 h
—Assume no IC

- Ionization = 6.6 eV

* R. G. Helmer and C. W. Reich, Phys. Rev. C 49, 1845 (1994)

Calculated rates for internal conversion (IC) and bound internal conversion (BIC) from Mau Chen - LLNL

- Nuclear radiative decay rate from Weisskopf estimate 0.013 s-1 or 76.9 second lifetime

-Internal conversion rate 2.58 x 10^{7} gives a lifetime of 0.039 x 10^{-6} seconds

-Th^{2+} BIC rate ~2.5 x 10^{6} gives a lifetime of 0.4 x 10^{-6} seconds

-Th^{3+} BIC rate ~4 x 10^{6} gives a lifetime of 0.25 x 10^{-7} seconds

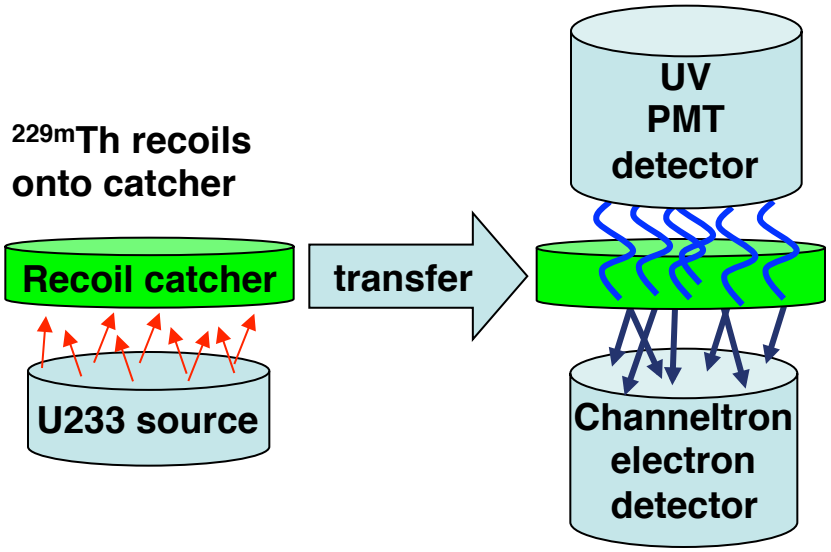
-Literature quotes Weisskopf estimate in this high mass region not very accurate and hindrance factor needs to be applied. We multiply by ~300.

Th atom	12 x 10^{-6} seconds	1 by definition
Th2+ ion	120 x 10^{-6} seconds	10 times
Th3+ ion	75 x 10^{-6} seconds	6.25 times

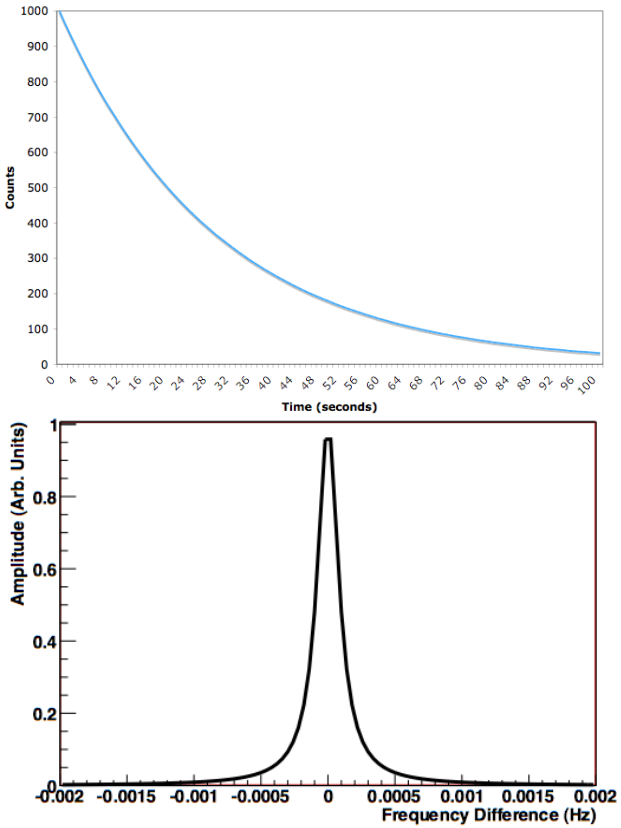
Half-life estimates: 5 hours ~ 18000 seconds to 10^{-6} seconds. We only need to cover 10 orders of magnitude with our search….

Measuring the half-life and decay mode of ^{229m}Th

- Goal: Measure the half-life of the first excited state of ^{229}Th nucleus to determine its linewidth for future direct laser manipulation.
- Use “hot-atom” technique to collect ^{229m}Th recoils from ^{233}U alpha decay and transfer in front of UV-Vis sensitive PMT and Multi-Channel Plate electron detector.
- ^{233}U background essentially gone



Half-life decay curve of ^{229m}Th

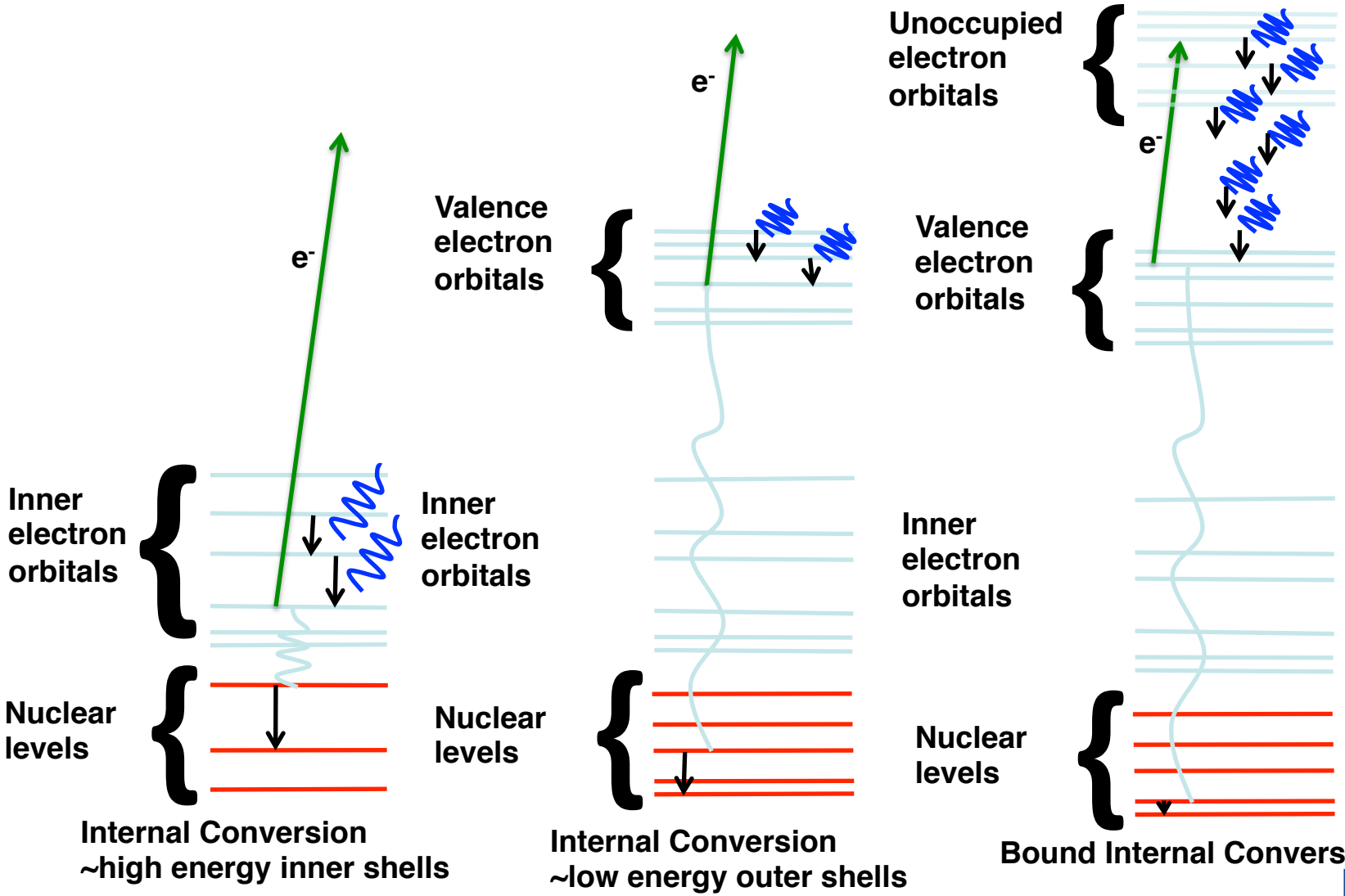


In addition, we learn the ratio of the gamma emission to the electron internal conversion rates.

Detectors used to date

- Multi-Channel Plate (MCP) detector
  - Detect Internal Conversion
  - Sensitive to individual electrons and ions
- Photo-Multiplier Tube (PMT)
  - Detect gamma decay or other light
  - Sensitive to individual photons
  - 120 nm to 600 nm
  - Can be chilled to -30 degrees Celsius to reduce background from 3000 cps to 150 cps
- Silicon alpha detector
  - Detects alpha particles and their energies and direction
- Use the Alpha Recoil Technique (ART) to obtain ^{229m}Th from ^{233}U
- Use different arrangements to look at different time scales
- Used ^{235m}U to prove the system works
- Custom Root/C++ codes were used for data analysis

Internal conversion and bound internal conversion



Use two reactions: control and unknown

- Control study is $^{239}Pu \Rightarrow ^{235m}U + \alpha$
  - Half-life is well known at 26.4 minutes
  - Energy of level isomer is 77 eV
  - Populated approximately 100% of the time
  - Recoil energy of ^{235m}U nucleus is approximately 80 keV

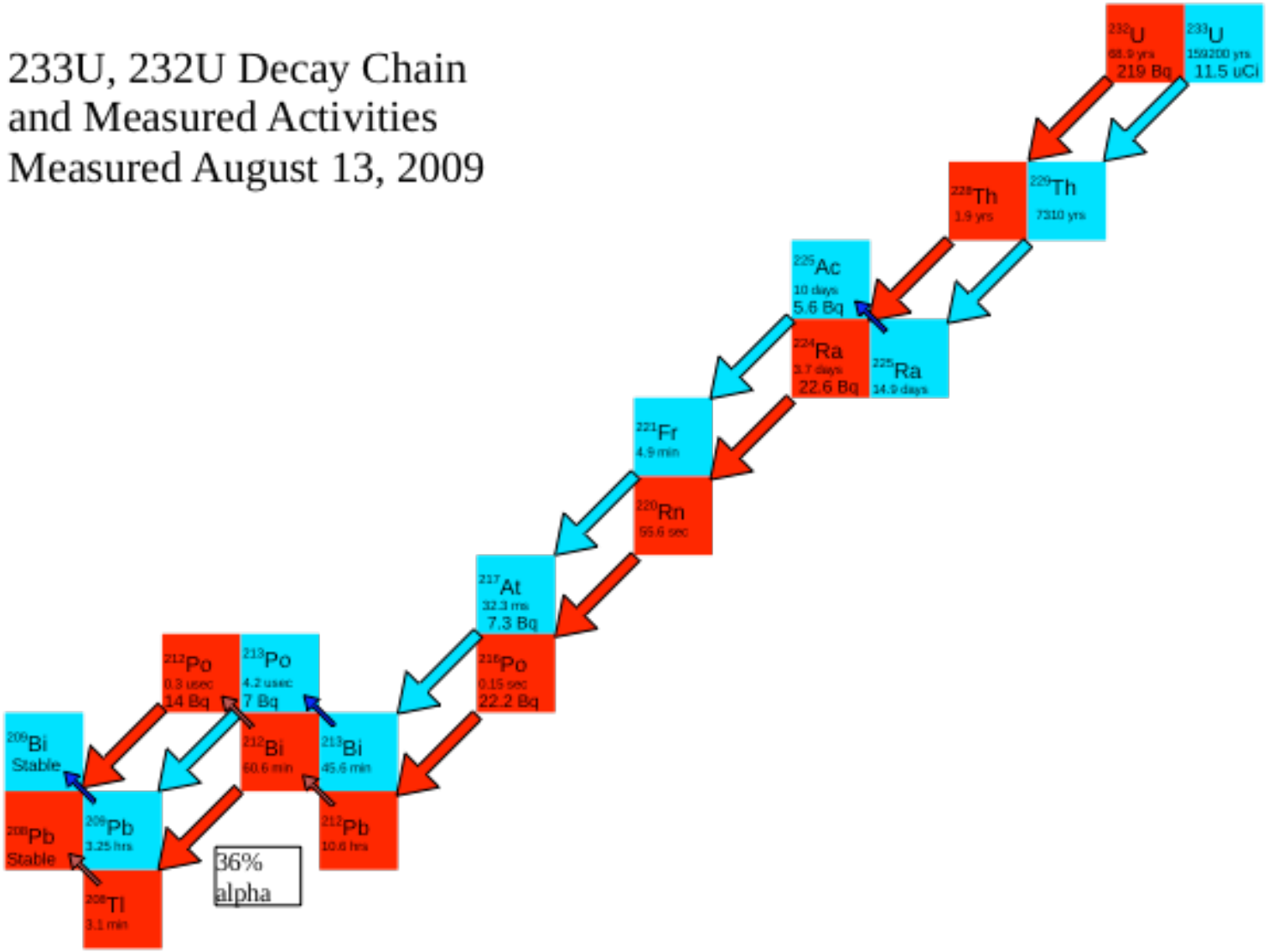
- Unknown study is $^{233}U \Rightarrow ^{229m}Th + \alpha$
  - Half-life is unknown at ??? seconds-minutes-hours
  - Energy of level isomer is 7.6 eV
  - Populated approximately 2-3% of the time
  - Recoil energy of ^{229}Th nucleus is approximately 80 keV

^{233}U production scheme (~1960 MIT reactor)
Material and α-source fab (2007, LLNL)

A	Irradiate ^{232}Th in reactor
B	^{232}Th + n → ^{233}Th(22.3 m) → ^{233}Pa(27.0 d)
C	Chemically isolate the ^{233}Pa [T(1/2) = 27 d]
D	^{233}Pa → ^{233}U
E	Clean up and deposit onto Al planchet (x 5), 1.9-cm D x 254-μm thick

^{233}U free of ^{232}U contamination
— Verified by γ-spectrum analysis

^{232}U impurity and daughters in the ^{233}U Source
1ppm ^{232}U in ^{233}U or 1/1000 Bq



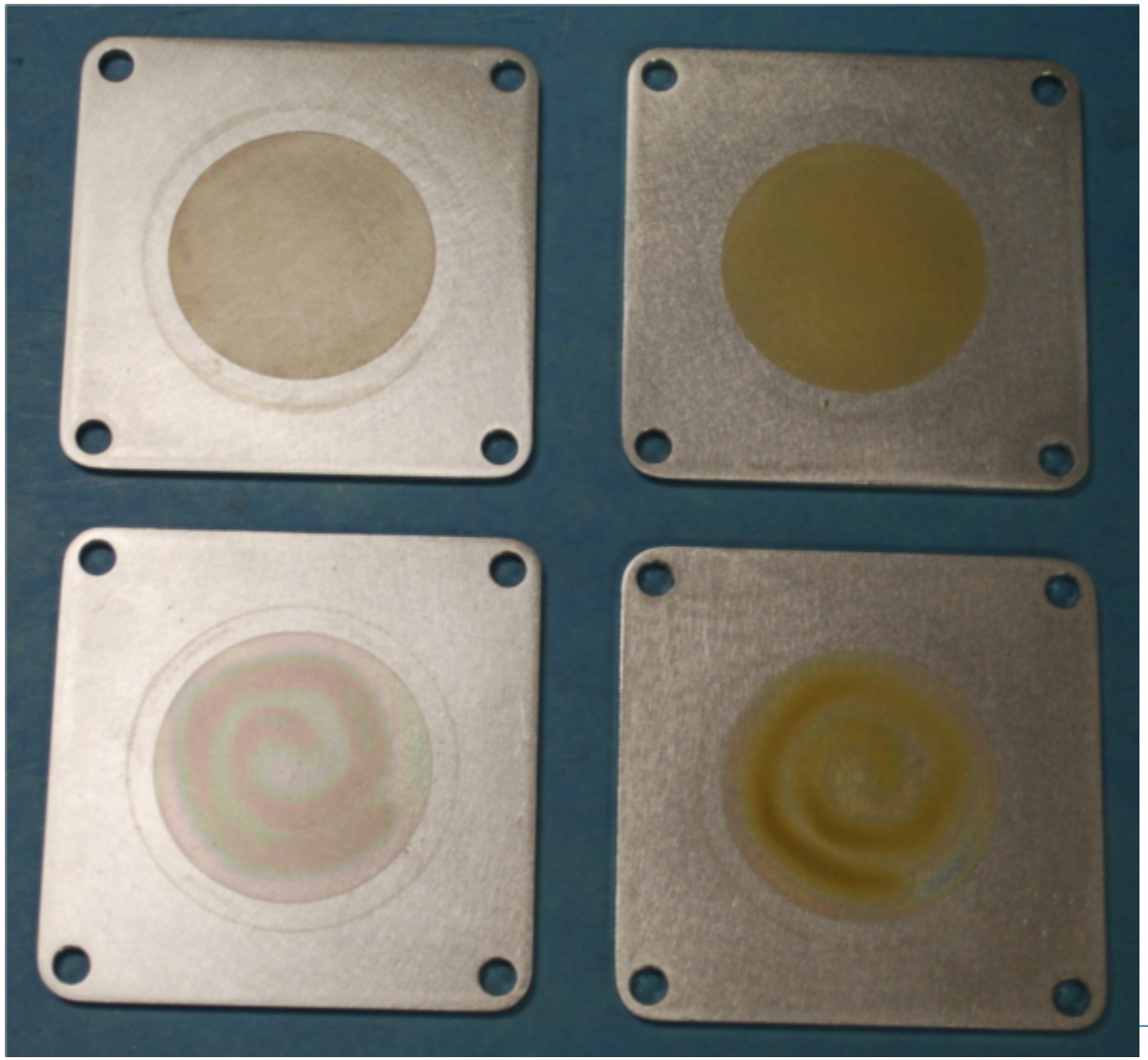
Range of Recoiling Th Nuclei: Need a thin source!

- Range is 100-250 Å, depending on chemical form
- Our ^{233}U source is 3.53 μCi, chemical composition not known
  - U metal would be 93 Å thick
  - $UO_2(NO_3)_2$ would be 1070 Å thick
- SRIM predicts recoil collection between 31% (U) and 12% ($UO_2(NO_3)_2$)
- Measured recoil collection efficiency is 38 ± 5% into 4Pi solid angle. So 74% if we measure the alpha direction.

Sources of Pu239 and U233

2 µCi ^{239}Pu

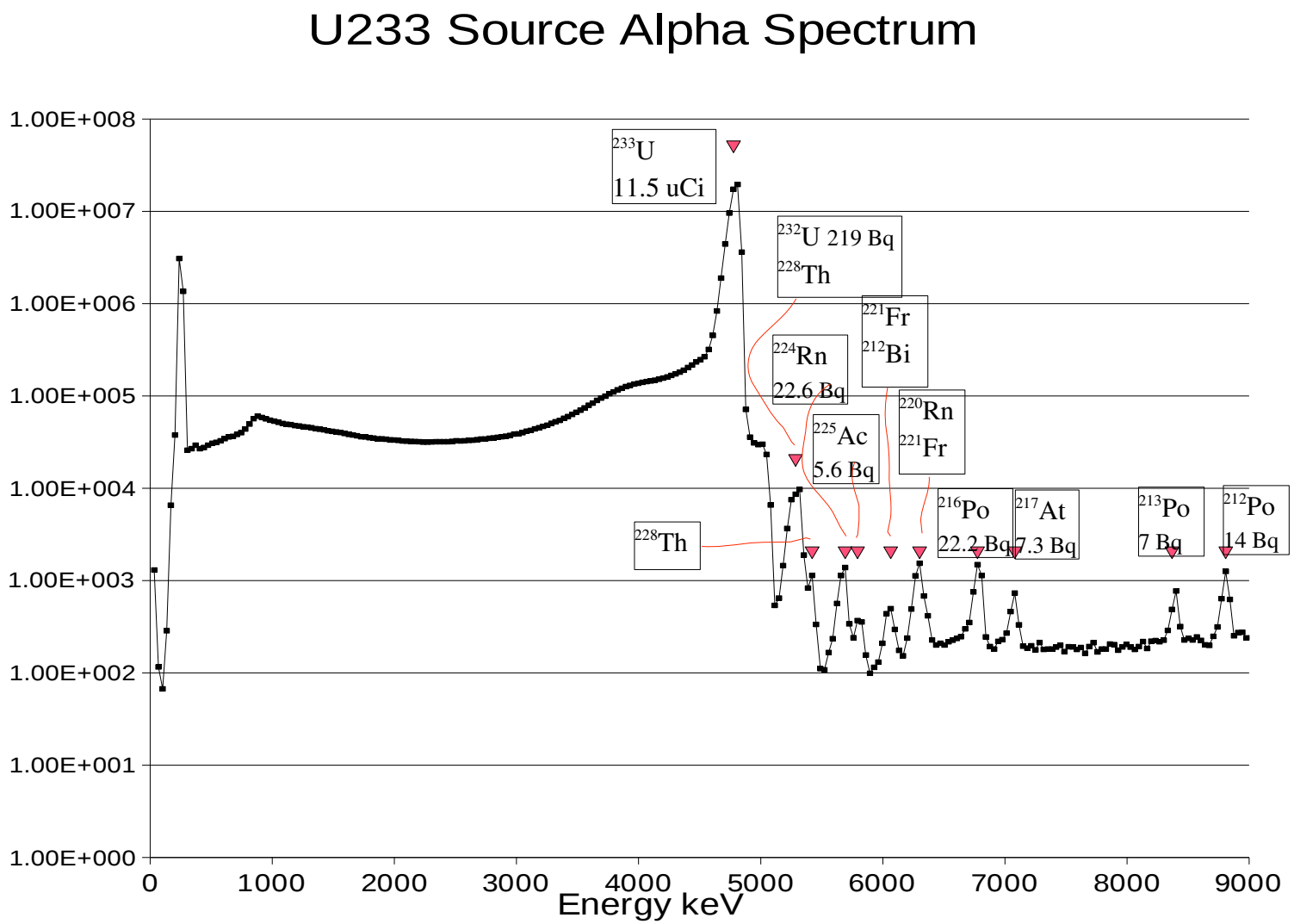
10 µCi ^{239}Pu



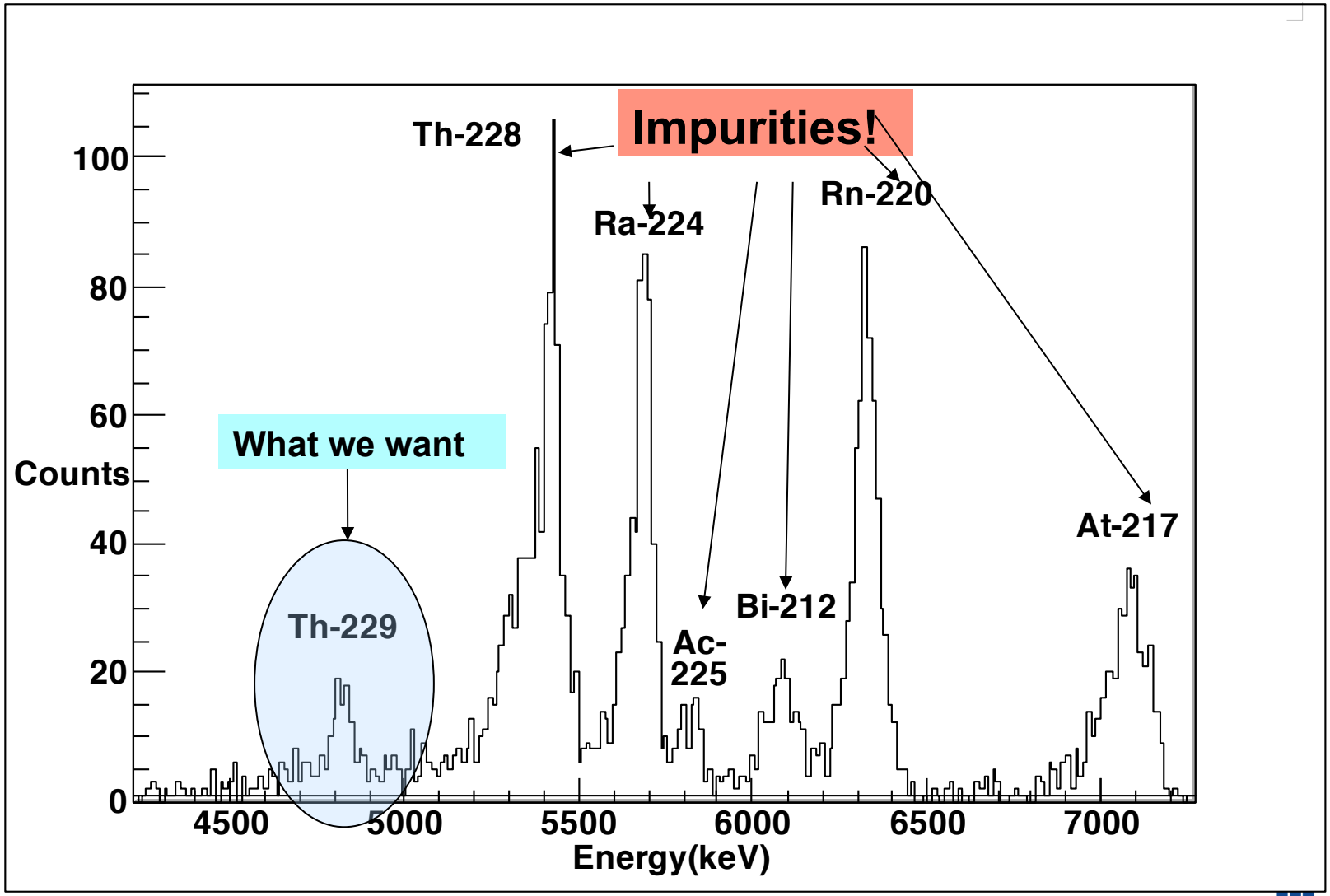
10 µCi ^{233}U

10 µCi ^{233}U

We have approximately 1 ppm contamination of ^{232}U in the sample by number of atoms or parts per thousand by decays per second.



Alpha spectrum of catcher foil after a long exposure (> 1 week). Source and catcher mm's apart.



Early days at LLNL – Burke Lab ~2007

Different setups for large range of half lives

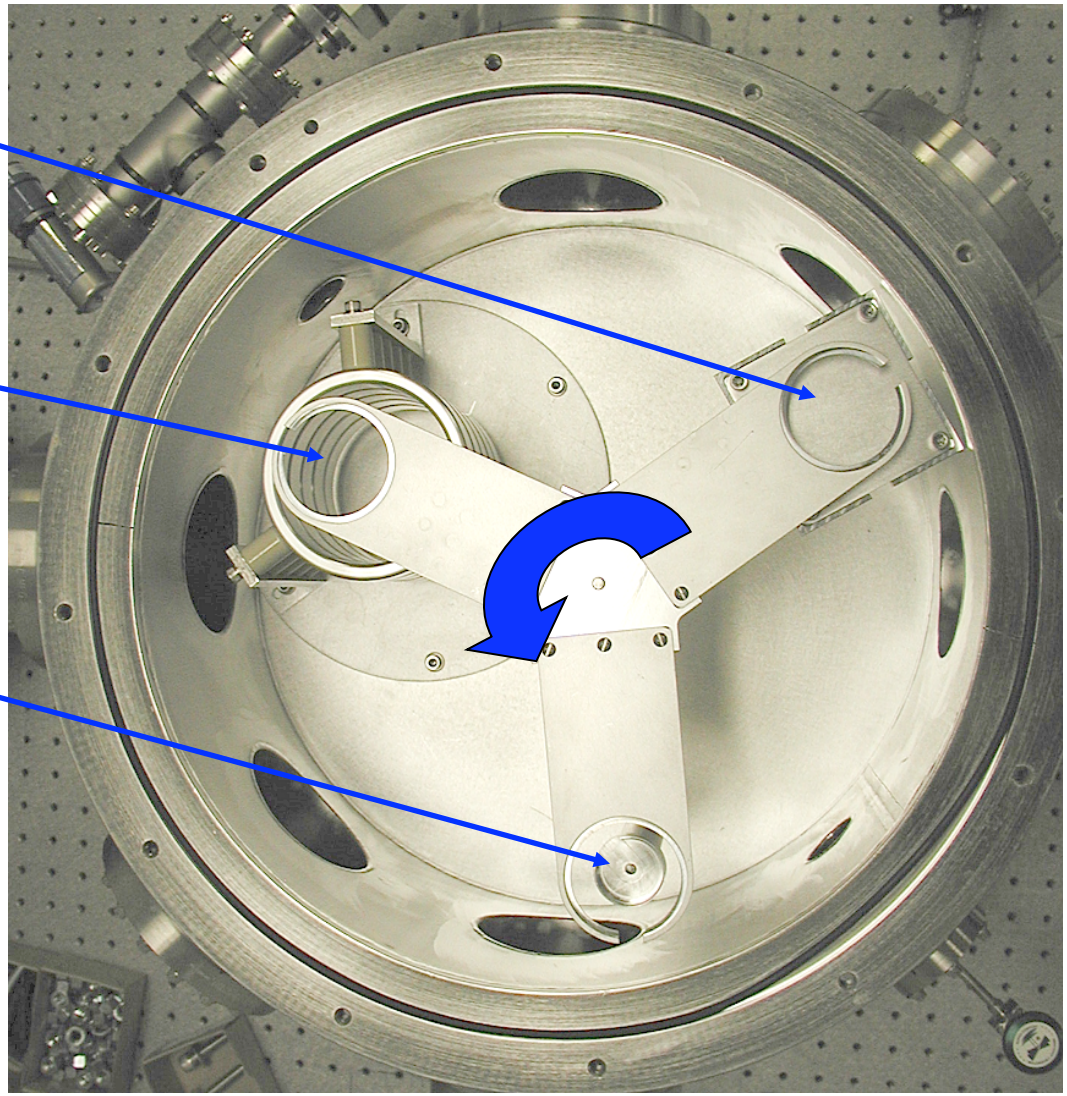
- The possible range of half lives is from 46 nanoseconds to 46,000 seconds. Need more than one experiment.
  - Manually rotate catcher foils
    - From 1 second to days
  - Use a fast mechanical shutter
    - From 2 milliseconds to hours
  - Alpha Coincidence
    - From 50 nanoseconds to 2 milliseconds

Internal design of experimental chamber.

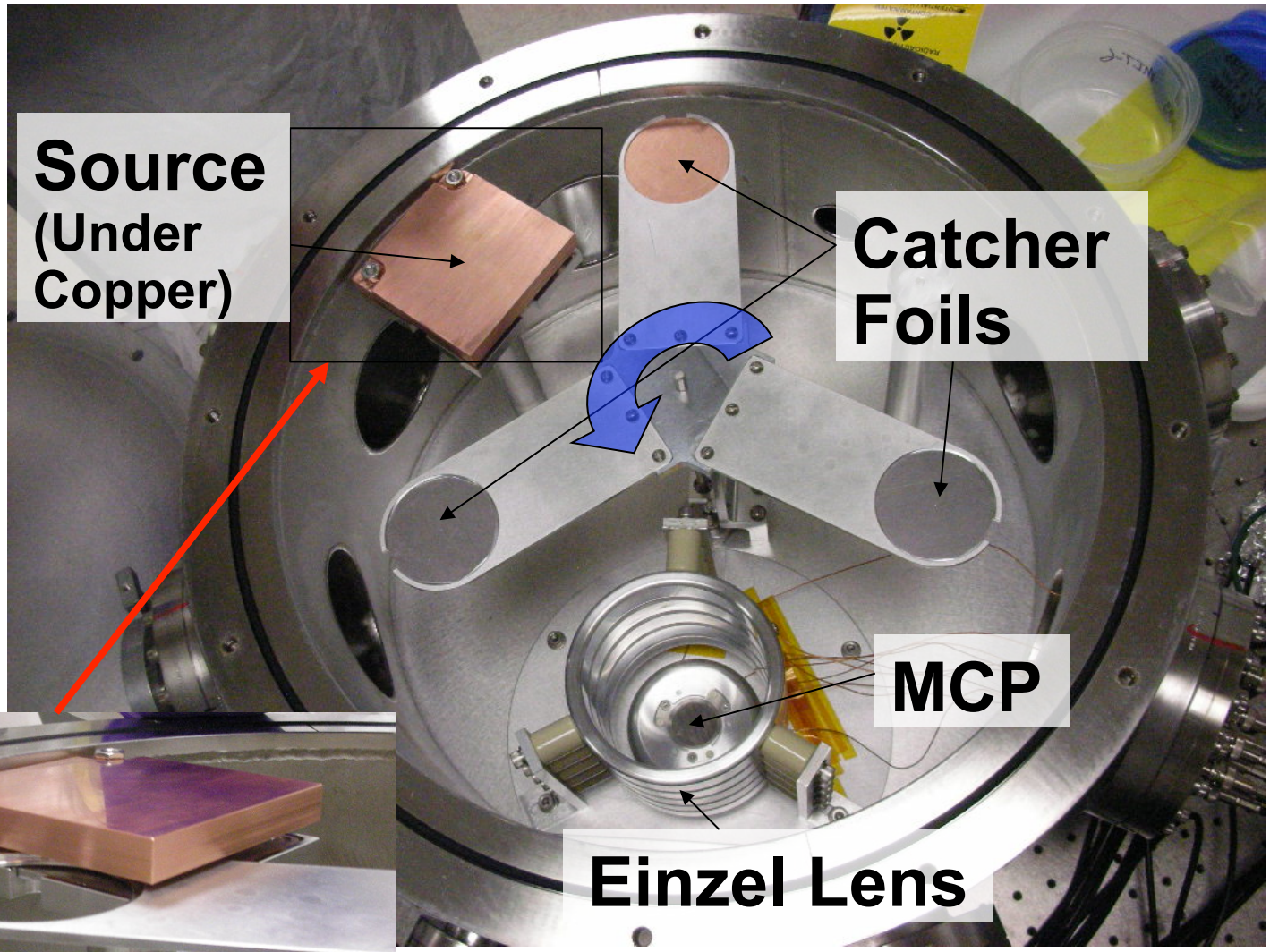
Loading stage (Pu-239 or U-233 sample) 2 inch diameter.

Electron lens and MCP detector.

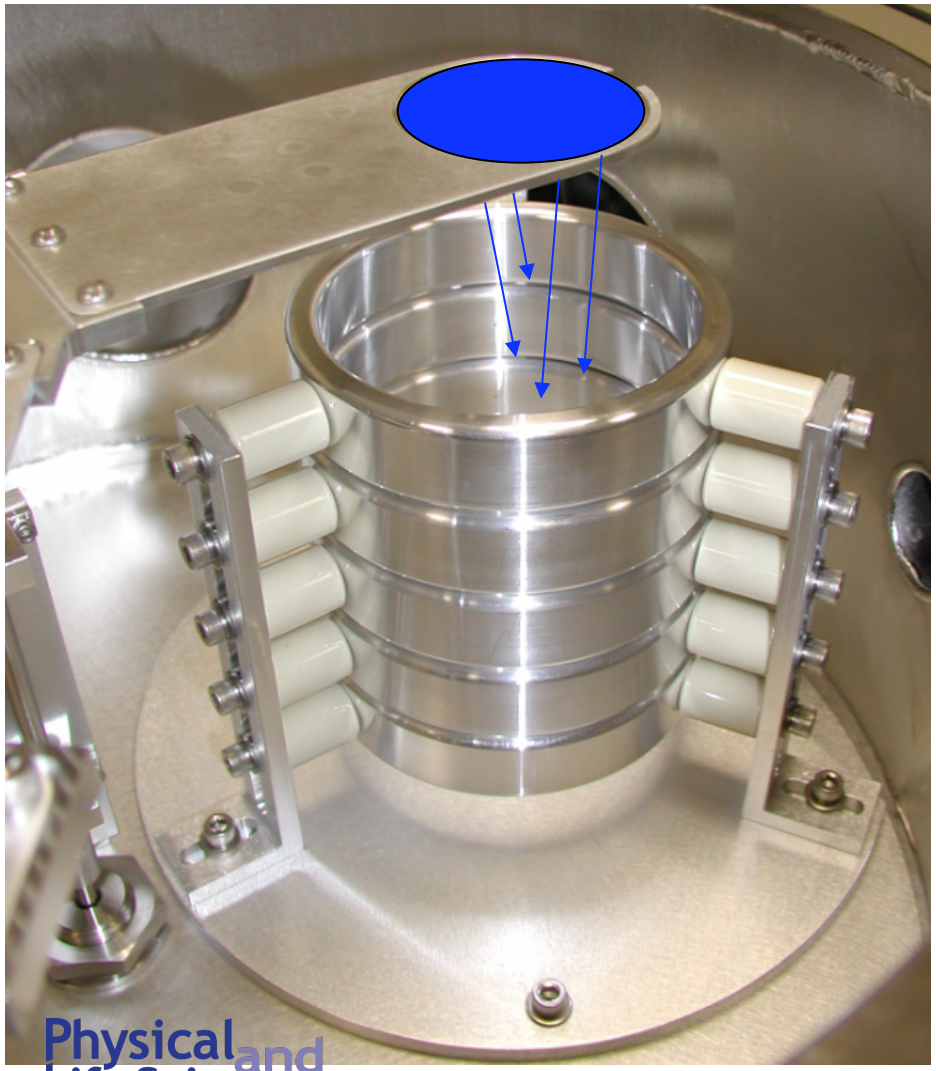
Silicon detector / contaminant background detector

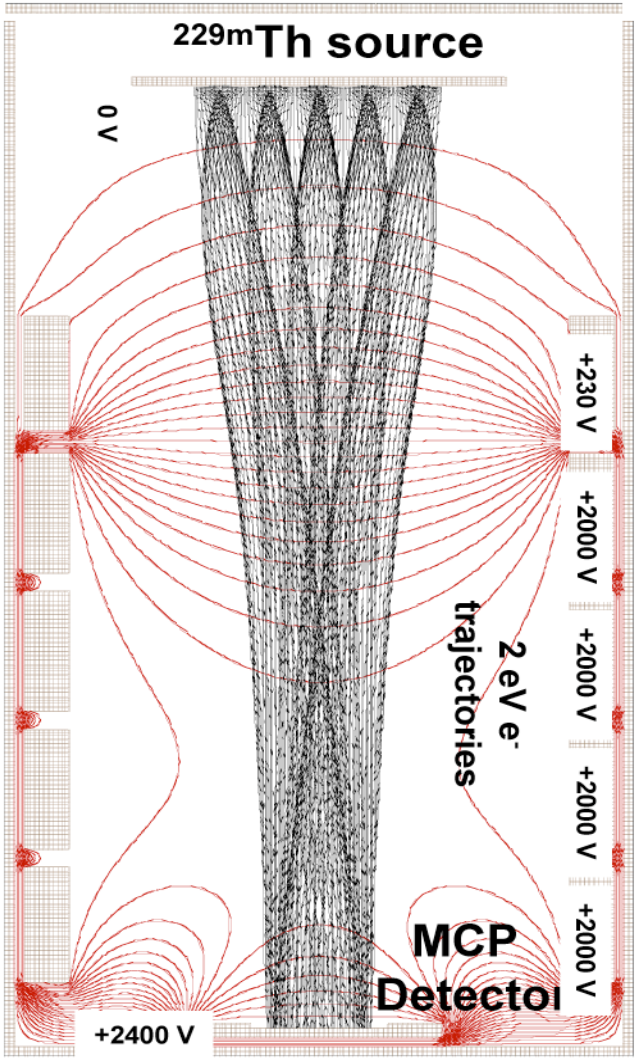


Approach 1) Slow half life experiment 1 second to 30 hours



Electron lens, MCP detector and SimIon simulation



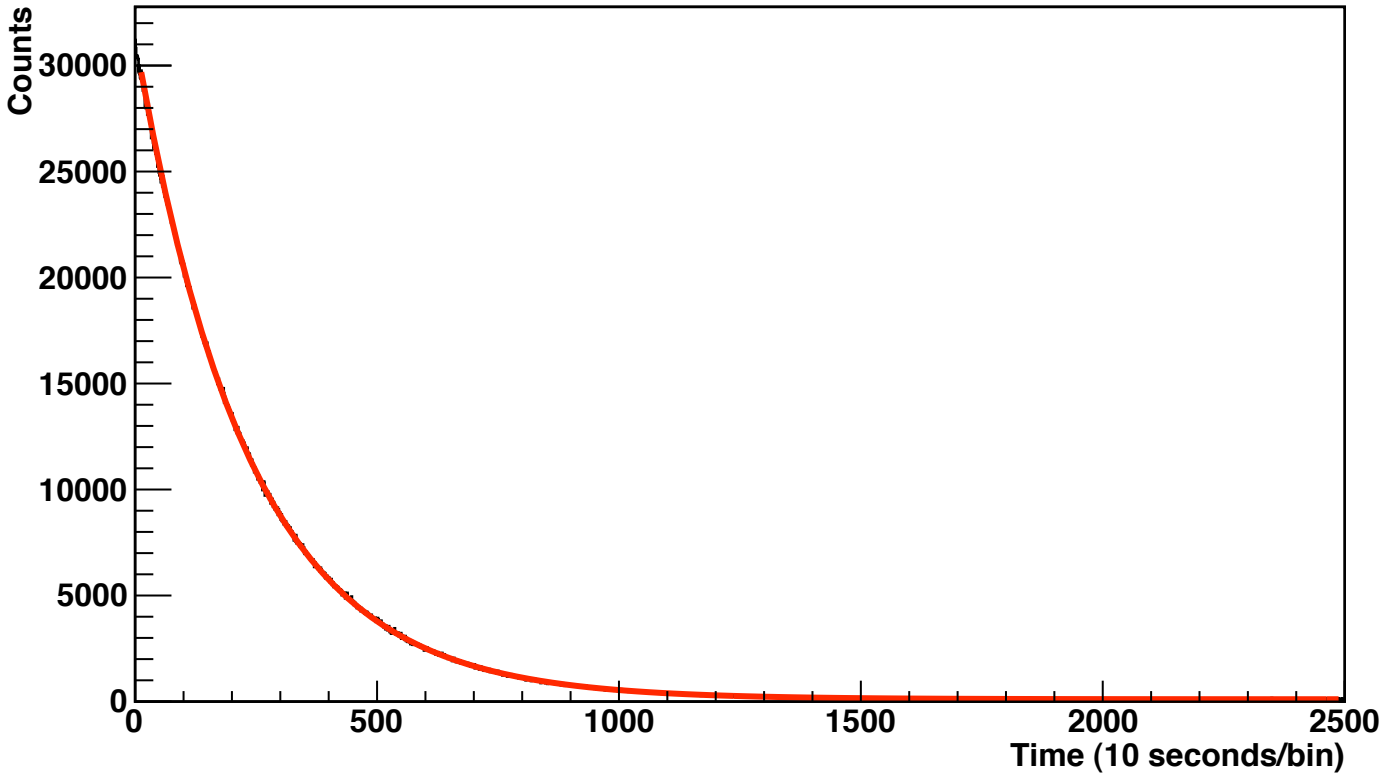


Physical and Life Sciences

Measurement of ^{235m}U decay from internal conversion electrons using MCP detector

$$R = N_o e^{\frac{(-Ln(2)T)}{\tau_{1/2}}} + B_o$$

N_o = 3116.0 ± 1.7 cps
$\tau_{1/2}$ = 27.012 ± 0.012 minutes
B_o = 10.64 ± 0.033 cps



Previous work from 1960's on ^{235m}U looked very promising.

CHEMICAL EFFECT ON THE HALF-LIFE OF U^{235m}

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Centre d'Etude de l'Energie Nucléaire–Studie Centrum voor Kernenergie, Mo–Donk, Belgium
(Received 30 June 1969)

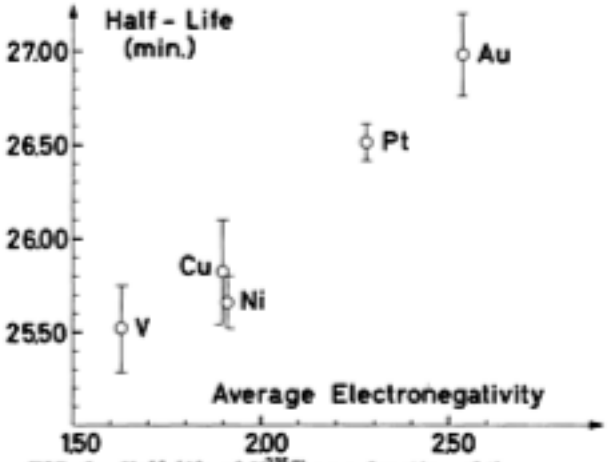


FIG. 1. Half-life of U^{235m} as a function of the average electronegativity of the metal into which the U atoms are implanted.

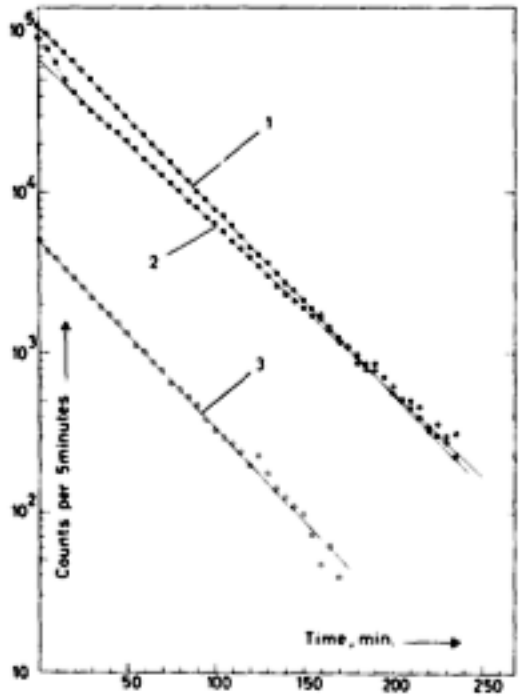
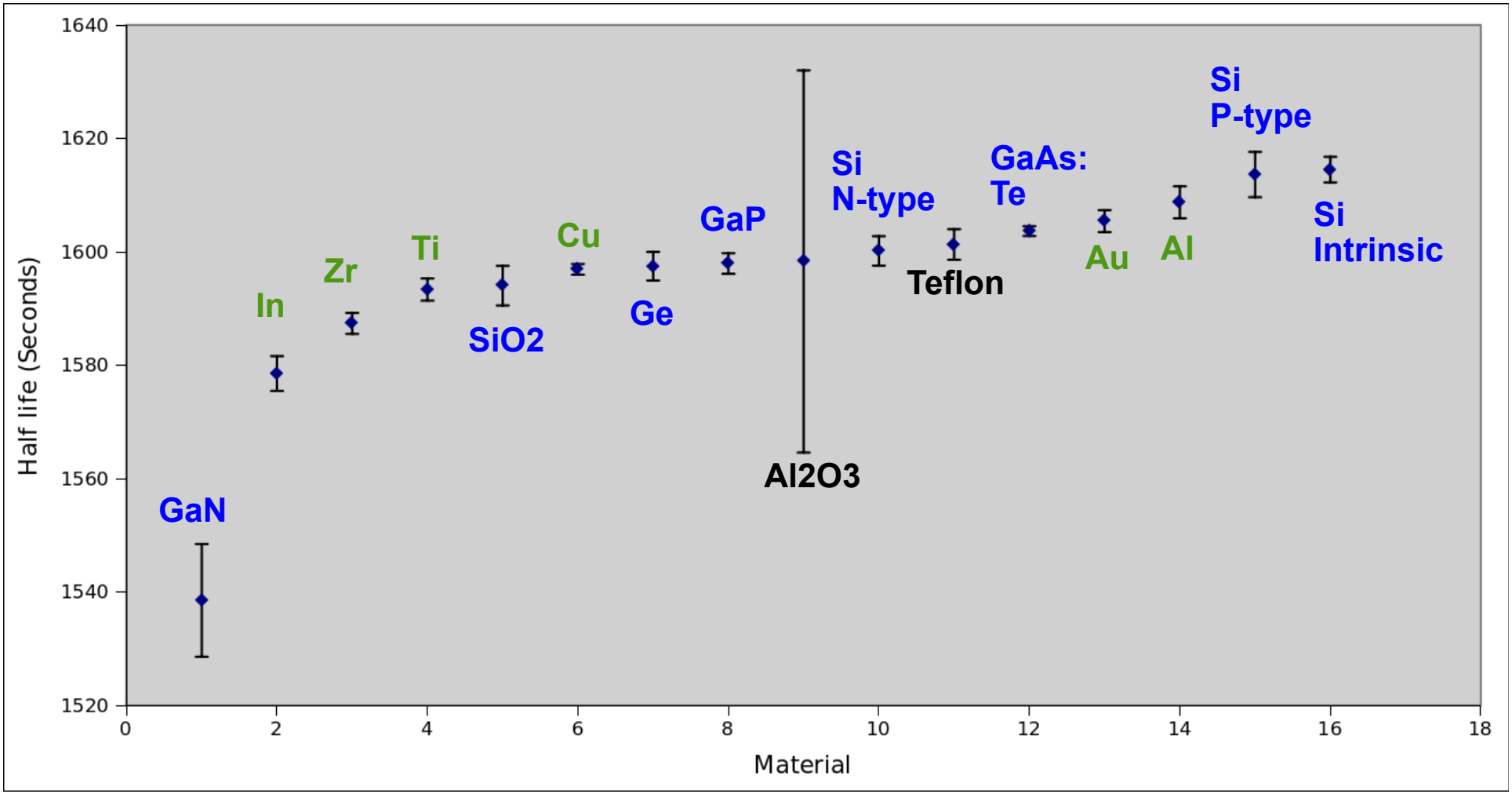


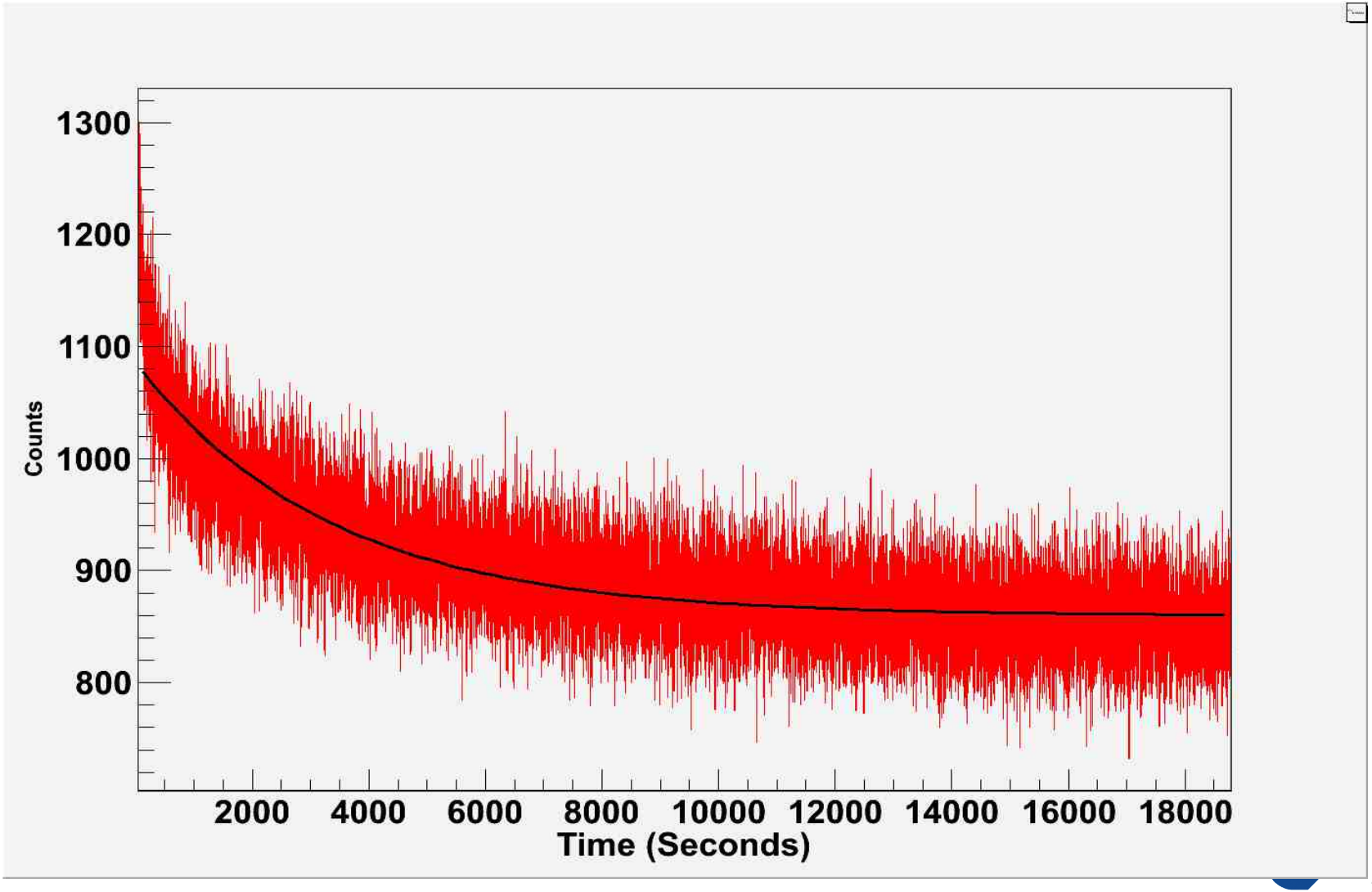
Fig. 1. Typical decay curves of $^{235}U^{m}$ collected on Cu.
Curve 1: argon (1 atm.) between ^{239}Pu layer and collector; 1 h collection time; single-channel analyser selecting the range 15-25 V from the 0 - 100 V output of the amplifier; $T_{\frac{1}{2}}$ = 26.21 ± 0.05 min.
Curve 2: same as 1, except for the single-channel analyser adjusted to 50 - 90 V; $T_{\frac{1}{2}}$ = 29.1 ± 0.1 min.
Curve 3: vacuum (~ 5×10^{-6} mm Hg) between ^{239}Pu layer and collector; 2 h collection time; single-channel analyser adjusted to 15-25 V; $T_{\frac{1}{2}}$ = 25.75 ± 0.11 min.
For all measurements, the decay was followed for at least two decades, a single constant background was subtracted from the data, and the half-life was deduced from a least-squares fit.

^{235m}U Half Life Variation

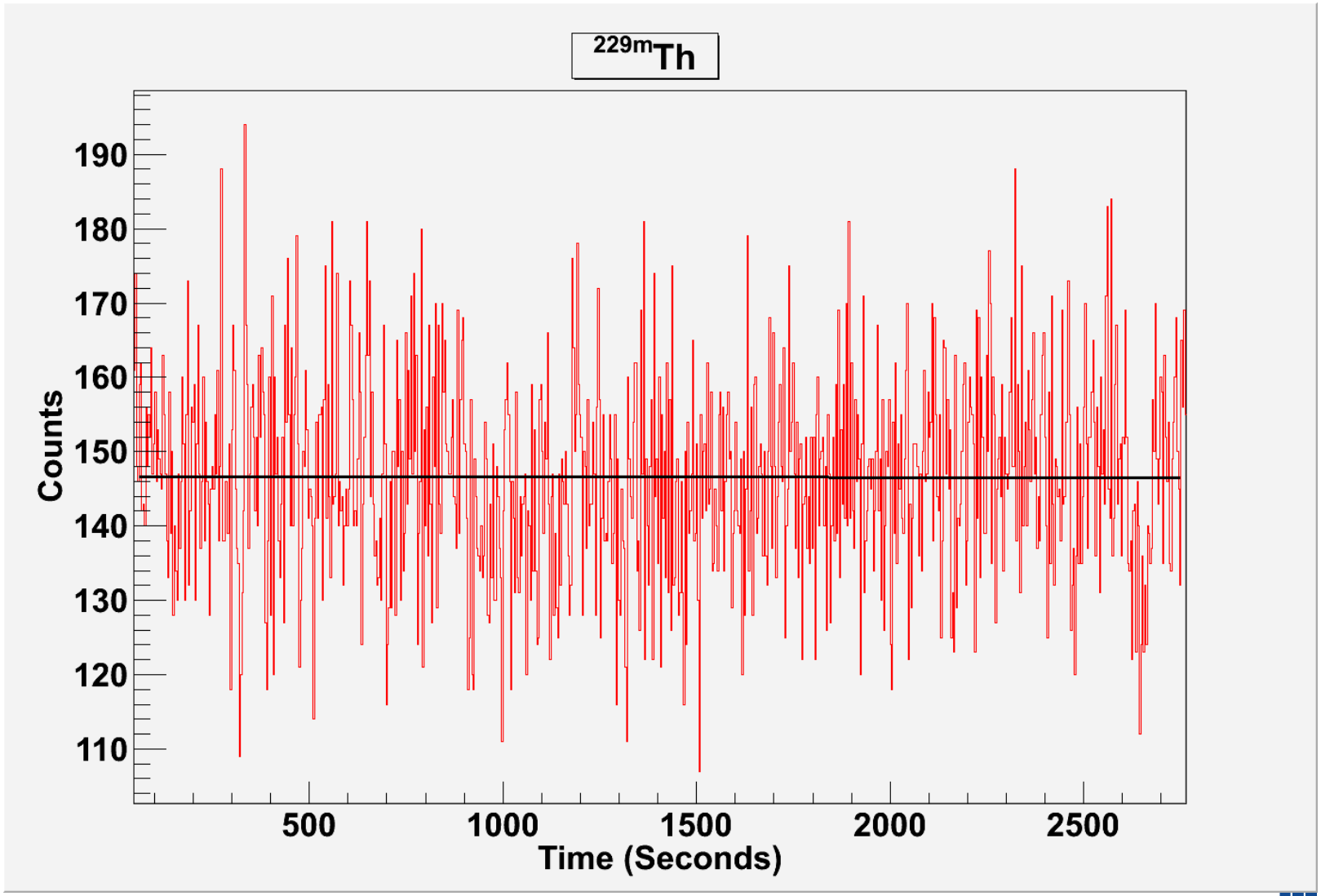
Half life changes based on electronegativity, oxidation state and material due to availability of electrons for internal conversion.



False decay curve from impurities in U233 source



Purified ^{233}U Source Results: No decay between 1 second and 30 hours

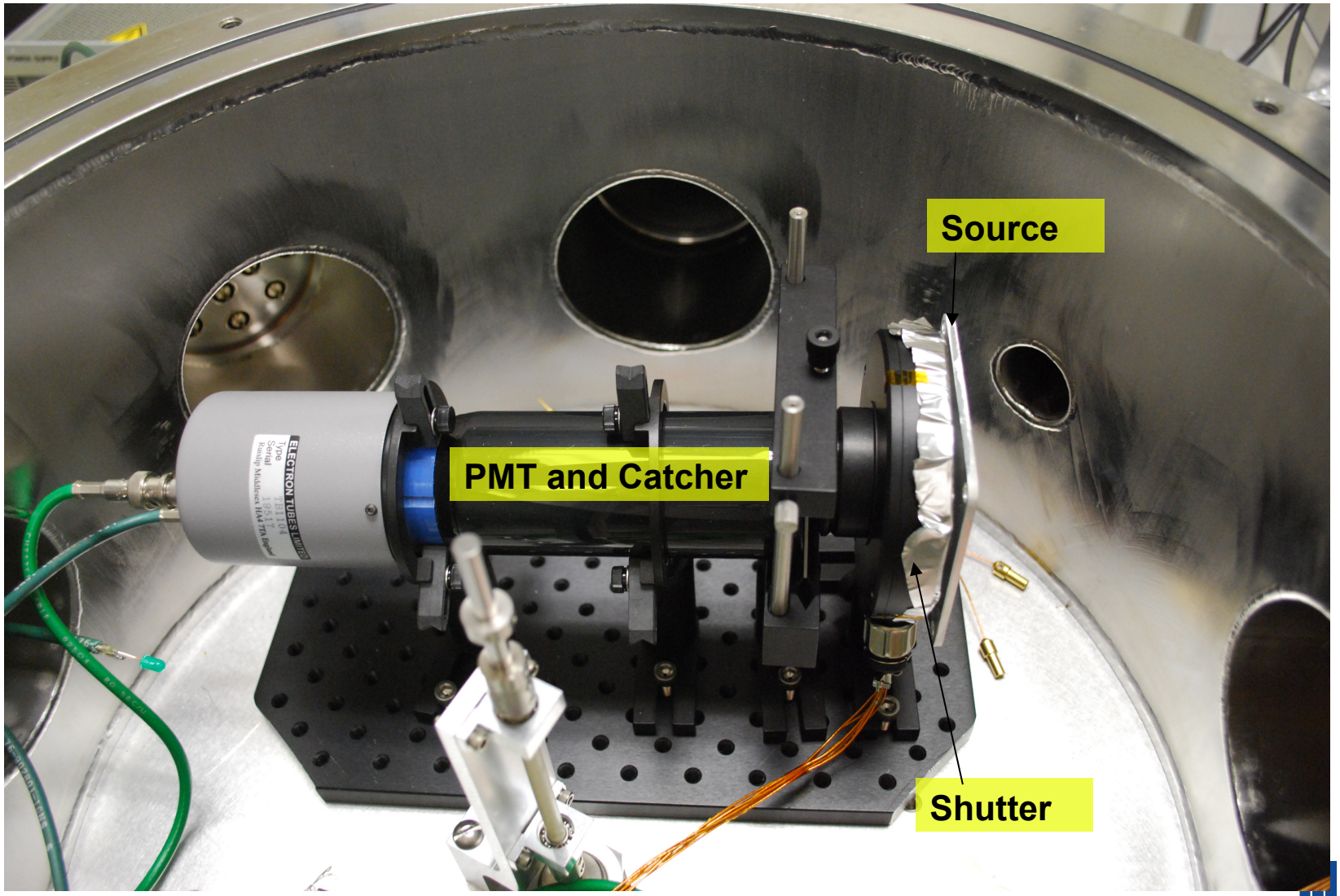


Be careful: The work function of the catcher material must be overcome

- Low work function helps an electron escape
- Electronegativity effects electron energy and half life
- Band Gap effects electron escape
- Direct band gap produces more light that we could detect
- We applied many different bias fields to “encourage” the electrons to migrate out of the bulk to no avail.

Material	Work Function (eV)	Electro-negativity	Band Gap (eV)	direct or indirect
Al	4.1	1.61		
Ti	4.3	1.62		
Zr	4.1	1.33		
Au	5.2	2.54		
Cu	4.6	1.9		
Teflon				
Indium	4.1	1.78		
Ge		2.01	0.67	indirect
Intrinsic Silicon	4.6	1.9	1.12	indirect
P-type Silicon	4.6	1.9	1.12	indirect
N-type Silicon	4.6	1.9	1.12	indirect
GaAs:Te		2	1.43	
GaP			2.26	indirect
GaN (on Al2O3)			3.4	direct
Al2O3			8.8	
Quartz			9	

Approach 2: Shorter time scales using mechanical shutter closes in 5 ms ~2 ms to 10 seconds

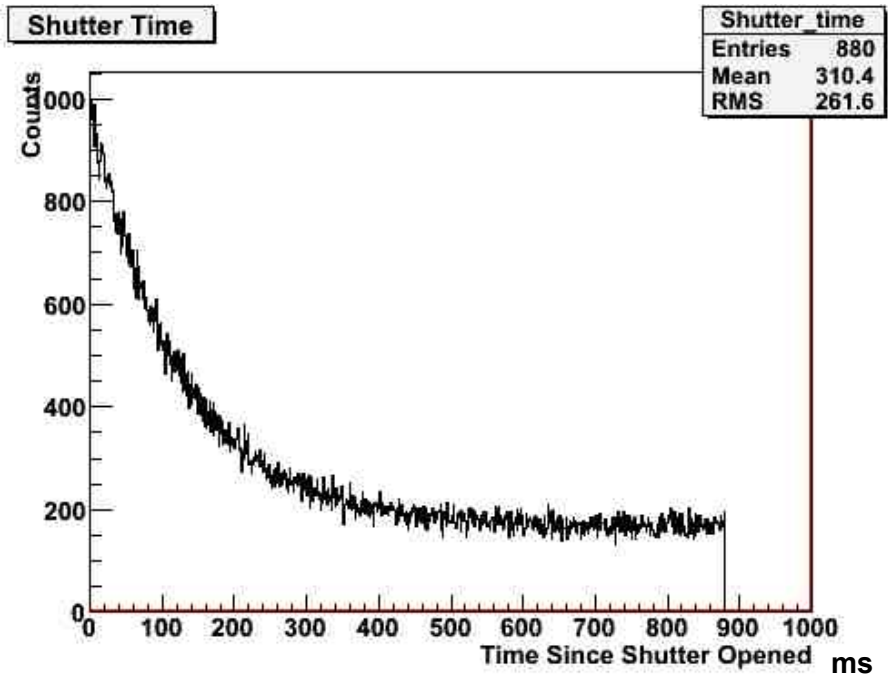


Mechanical Shutter

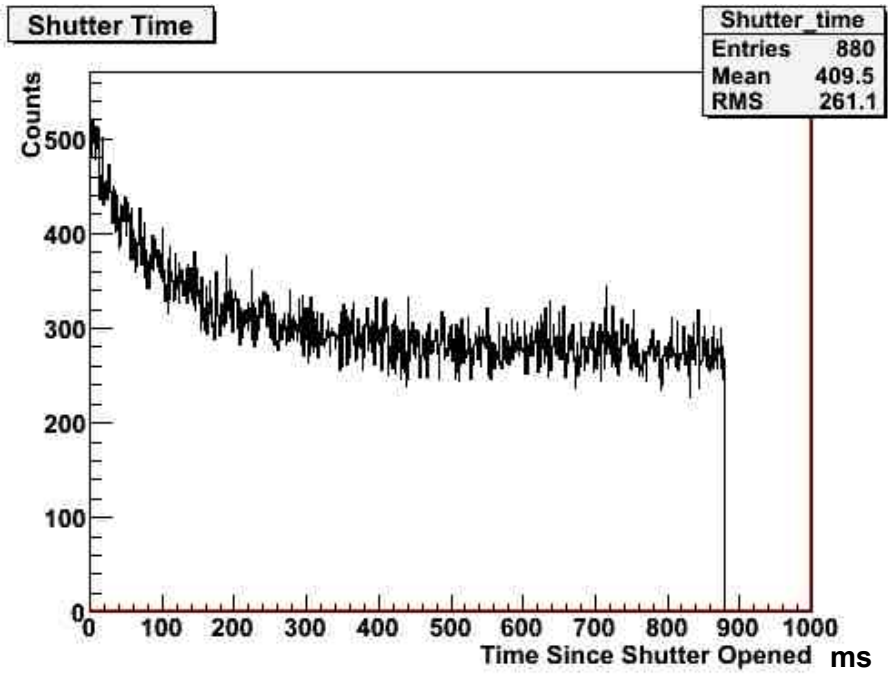
- Opens and closes in 5 ms
- Used with PMT and MCP
- Ran automated and repeated multiple times
- Plotted time of signal arrival after shutter close
- Used ^{239}Pu source as control
- Made measurements from 2 ms to 10 seconds

False Signals:
Mechanical Shutter MgF_2 Catcher with PMT

- Decay is 86 ms for both! Comes from MgF_2 excited by the alpha particles striking it.
- Saran wrap used to range out Th recoils, but allow alpha particles through.

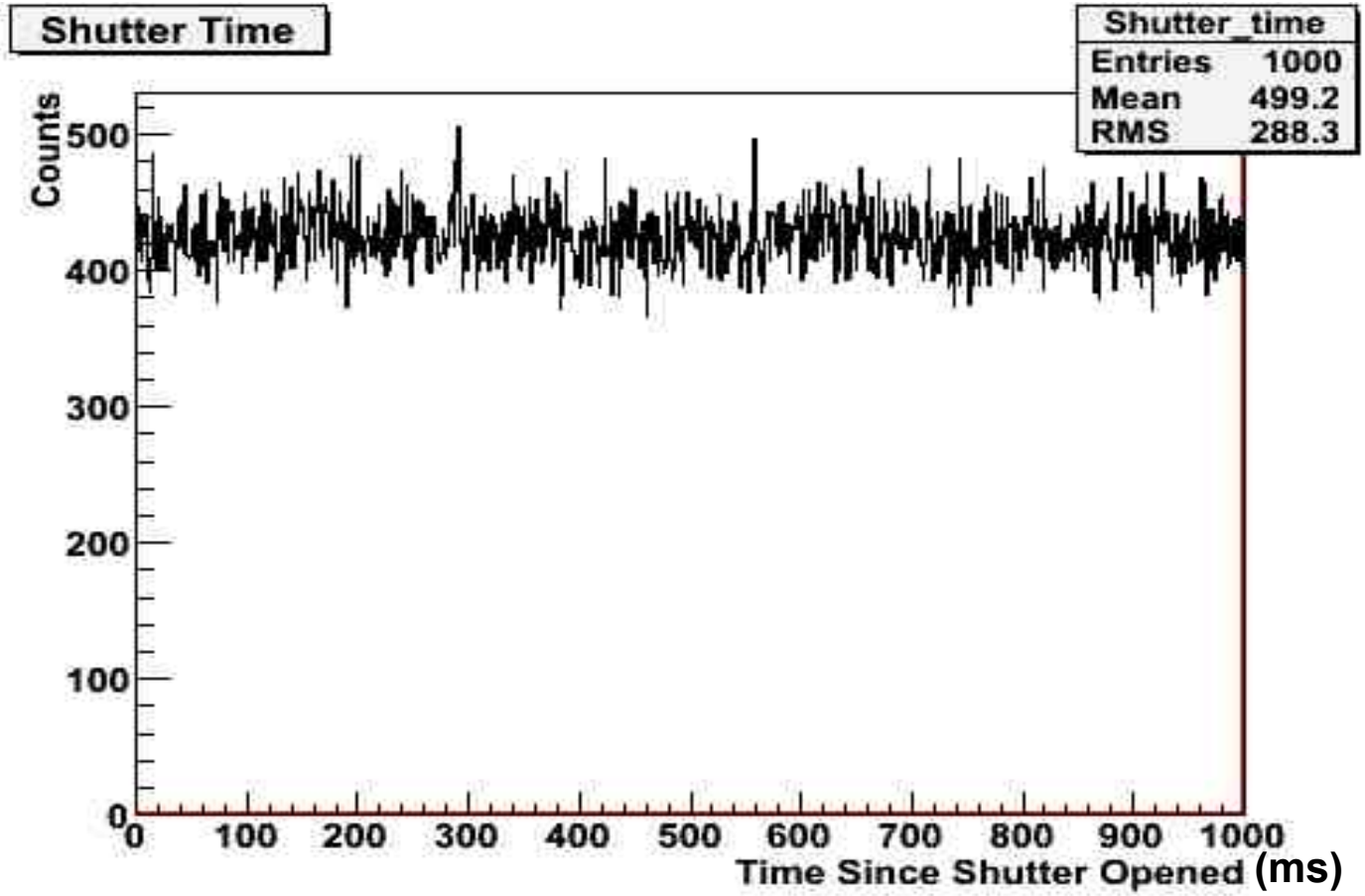


^{239}Pu Source



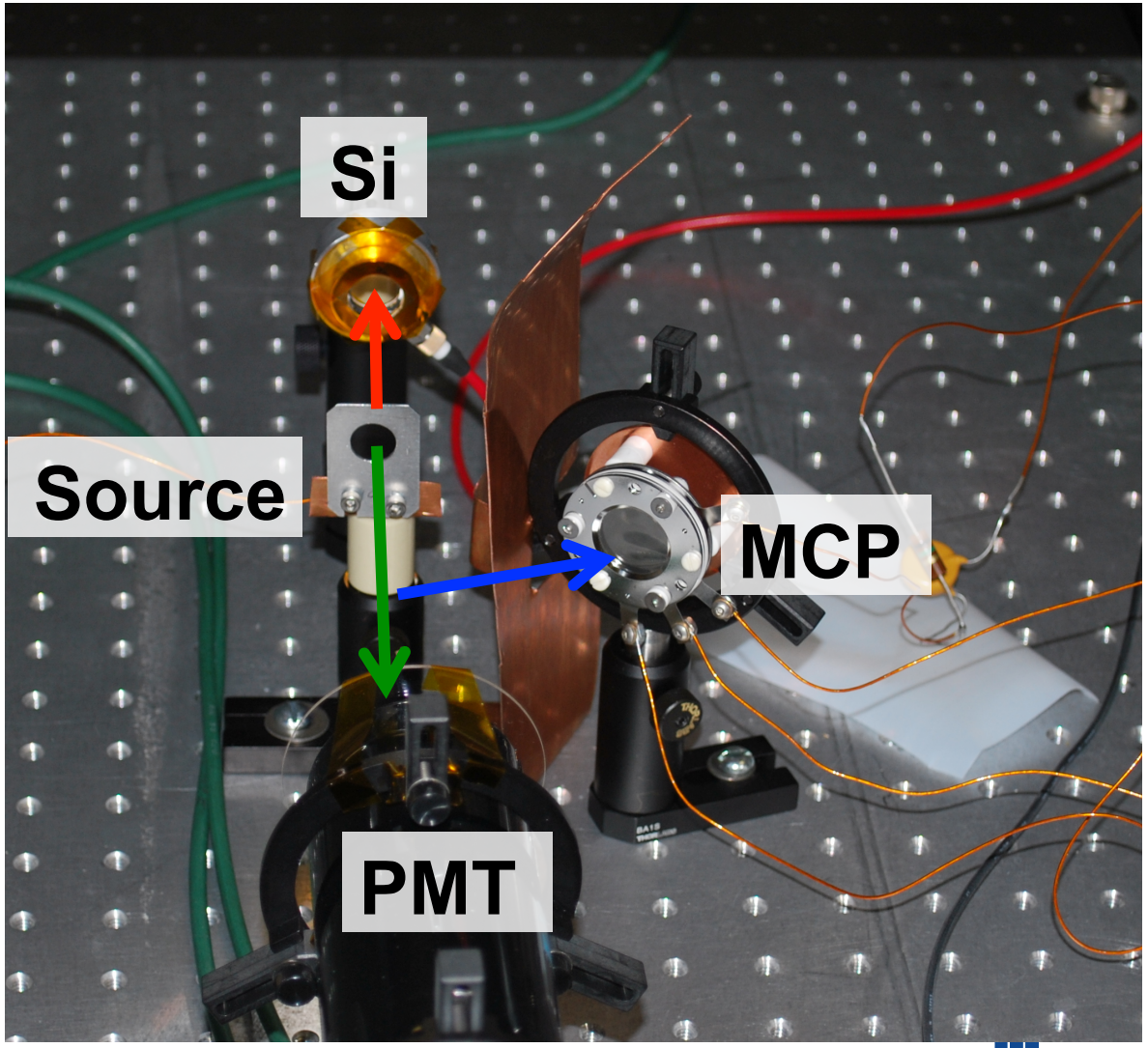
^{233}U Source

Mechanical Shutter MgF_2 Catcher with MCP
No Decay from 2 ms to 10 seconds



Approach 3: Alpha Coincidence
Fastest 50 nanoseconds to 2 milliseconds

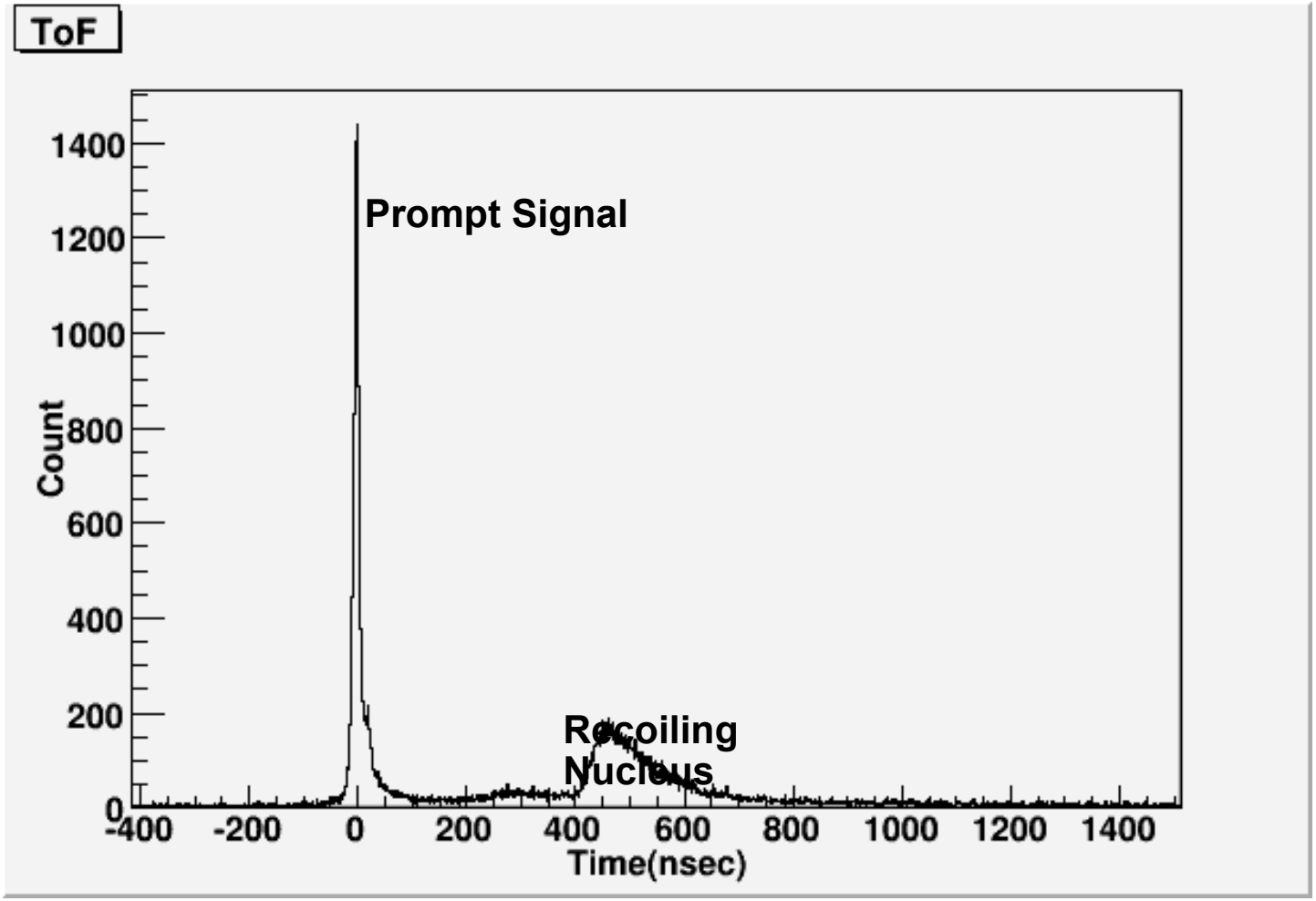
- Detecting an alpha means a ^{229}Th was just created.
- Only 1.6% to 2% of the time a ^{229m}Th.
- Look for subsequent Th229m decay.



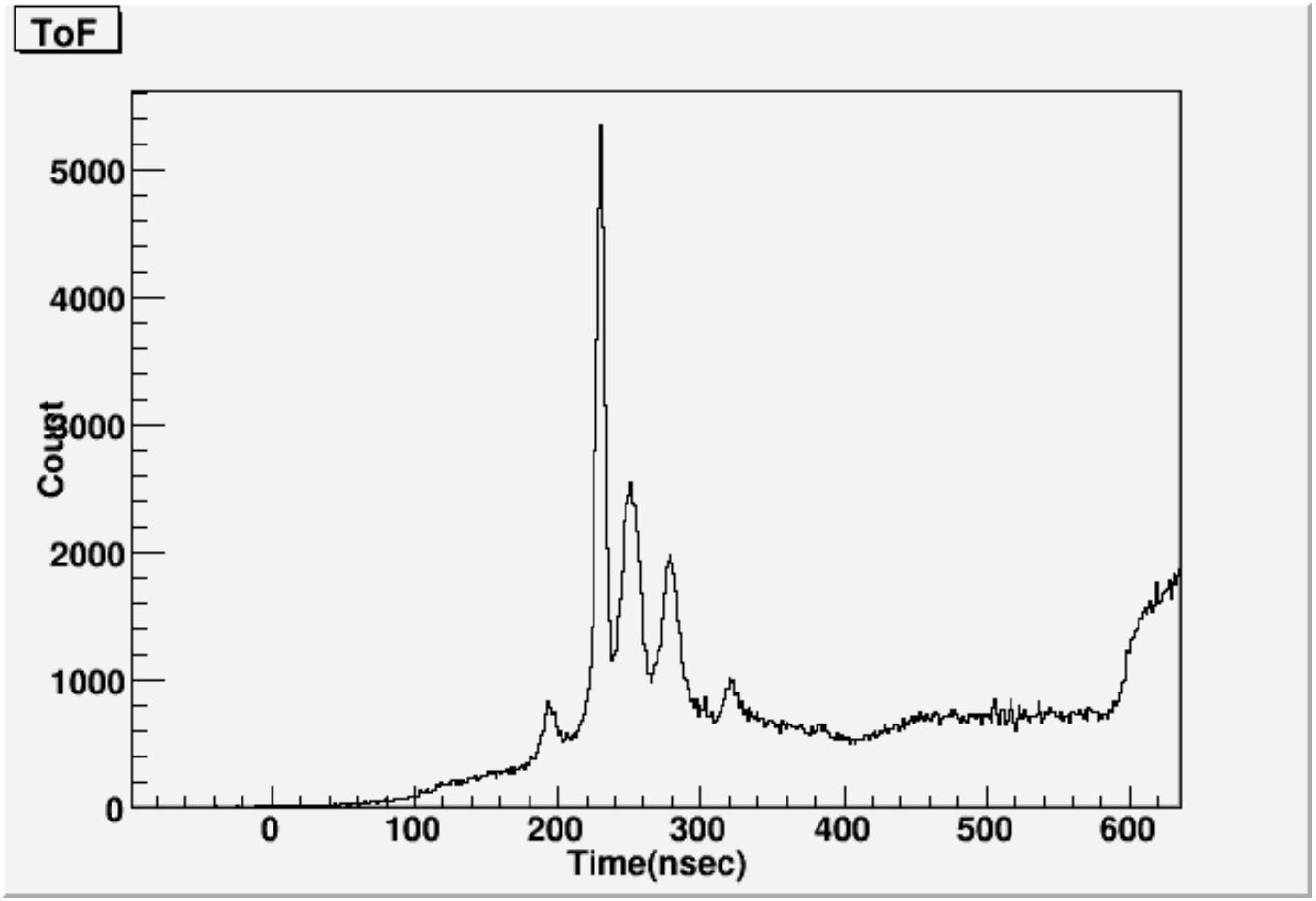
Alpha Coincidence: Shortest time period 50 nanoseconds to 2 milliseconds

- Measure time between events
  - Alpha-signal
  - Alpha-signal with time window
  - Alpha-recoil-signal
  - Also measured alpha energy
- Used with PMT and MCP
- Ran for several days at a time
- Used ^{234}U source as control
- Made measurements from 50 ns to 2 ms

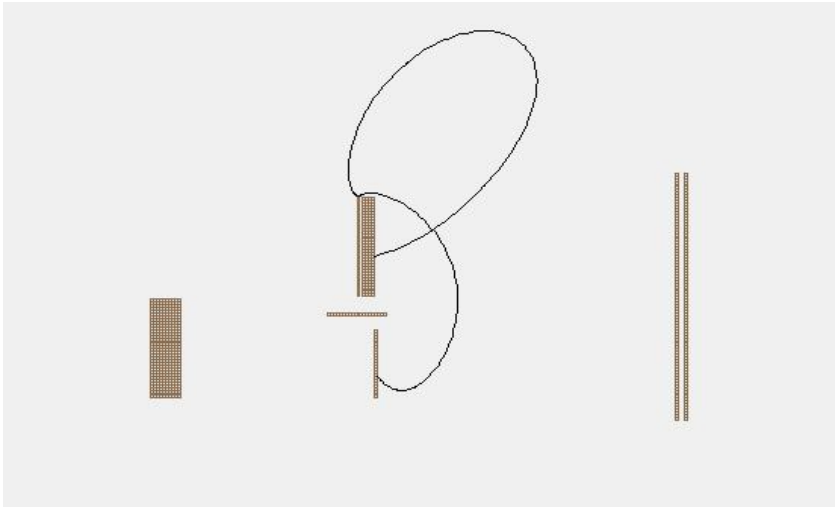
Alpha Coincidence with a PMT

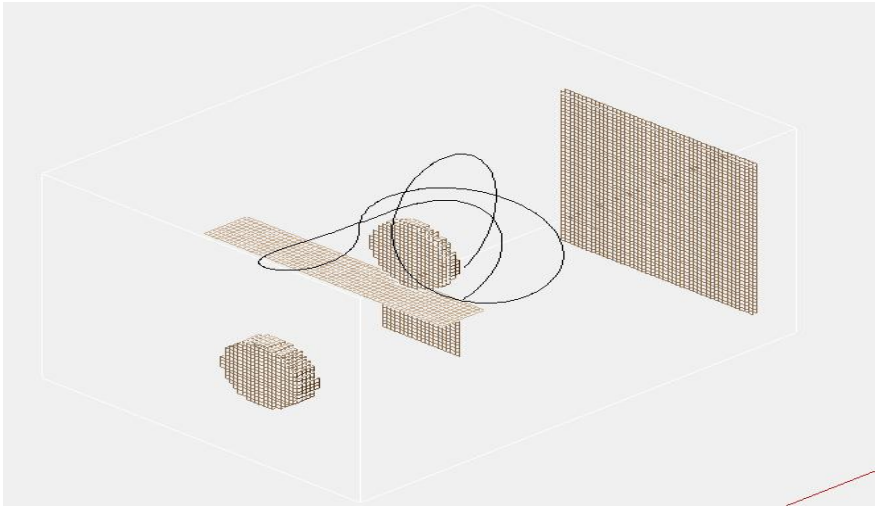


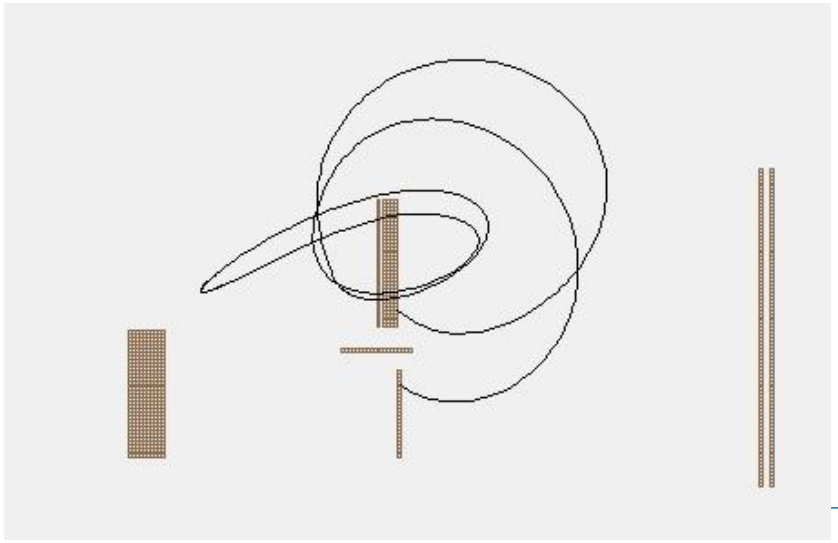
Alpha Coincidence MCP seeing peaks in ToF, which is incorrect

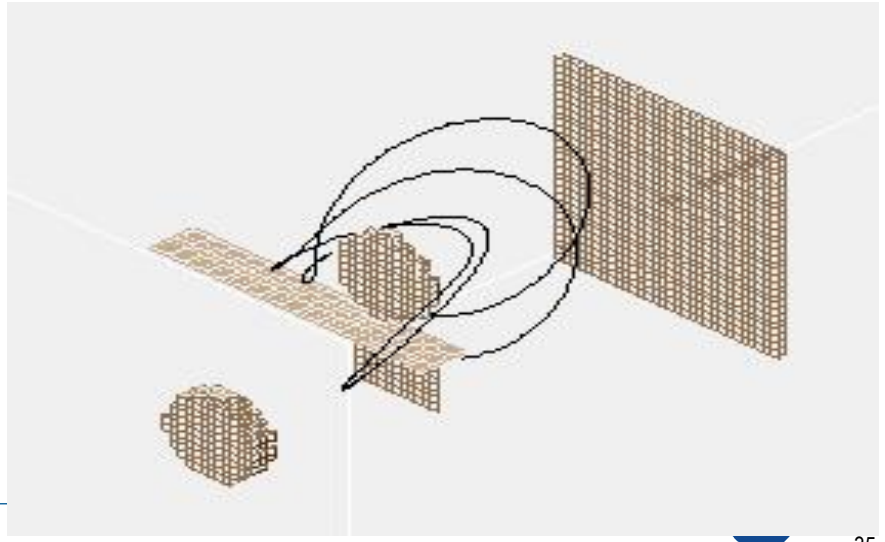


Solution: Multiple electron orbits caused by fringe fields
SimION: Electrons Orbit MCP, Causing One Peak per Orbit









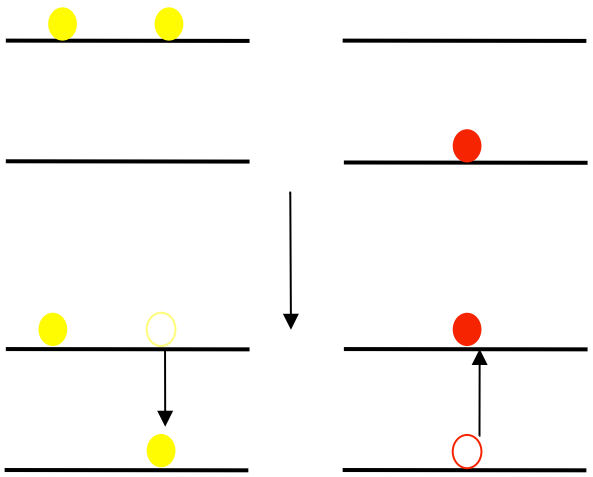
The elusive ^{229m}Th decay current conclusions

- Less than 50 nsec or greater than 30 hours
- The signal could be below the detection limit
- IC electrons never escaped – work function
- BIC need to be transparent to optical wavelengths
- Non-radiative decay in a material
- A different energy….
- Fundamental range problem: Recoil goes 20-30 nm deep. Electron range 2 nm, light not much further….

Note: we think 233U -> Th229 + α is auto-ionizing. The 29 keV decay to the isomer by internal conversion causes a cascade of Auger electron emission which leaves the Th229 in a ~<8+> charged state.

Nuclear Excitation by Electronic Transition (NEET)

- Rare nuclear excitation predicted to occur in certain isotopes
- The inverse of bound internal conversion
- NEET Process
  1) Electrons are in an excited state
  2) Nucleus is in its ground state
  3) An excited electron makes an atomic transition to a lower state
  4) Due to atomic-nuclear coupling, the energy from the atomic transition excites the nucleus to an excited state
- NEET can occur when an isotope has atomic and nuclear transitions that have the same energy and similar multipolarity



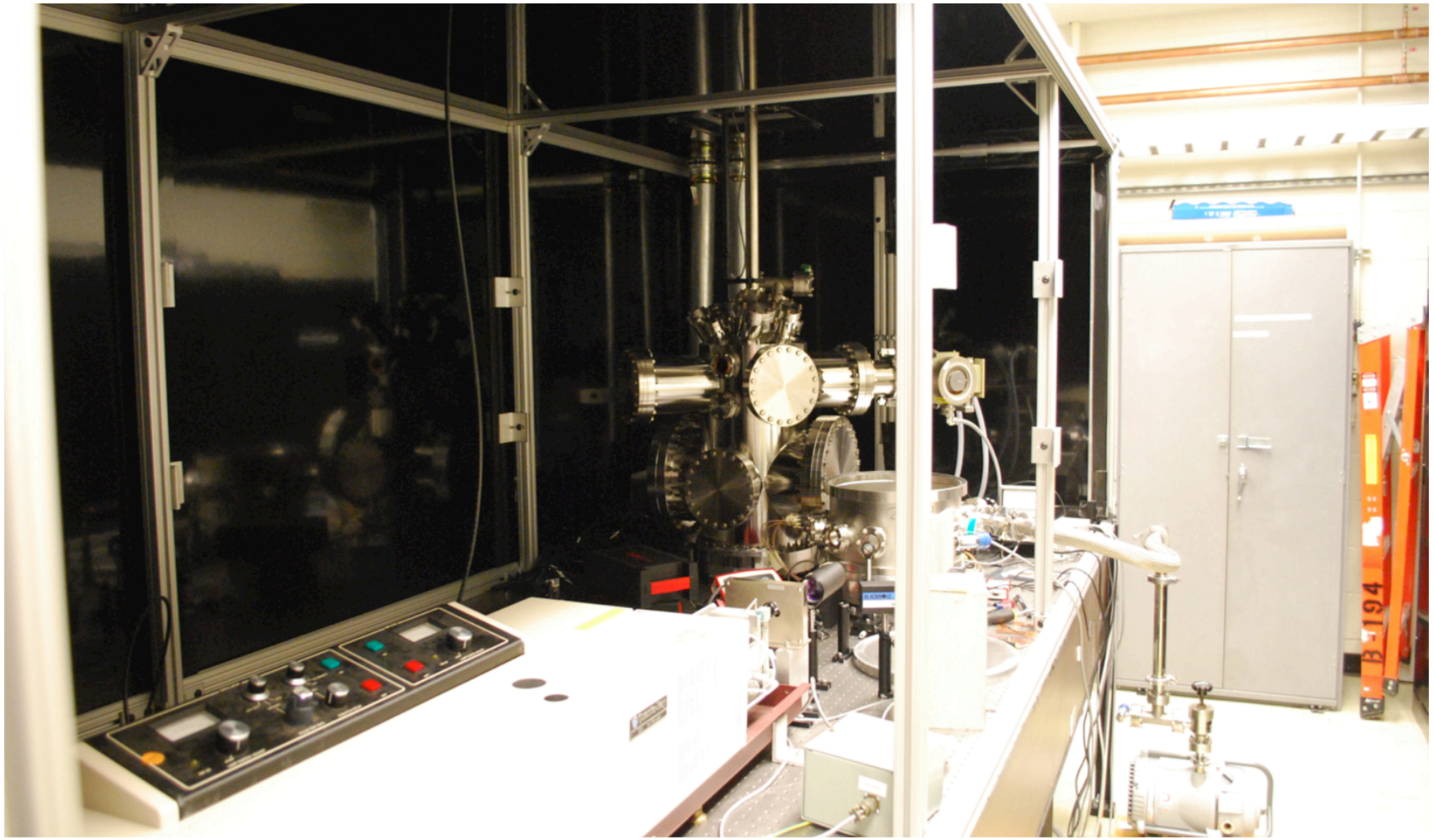
- **Possible electronic transitions in ^{235}U**
- **6p1/2 → 5d5/2 (M2)**
- **6d5/2 → 6p1/2 (M2)**

- **Nuclear transition in ^{235}U**
- **Ground state: 0 eV Jπ=7/2-**
- **Excited state: 77 eV Jπ=1/2+**
- **7/2- → 1/2+ (E3)**

Experimental Setup for U235 NEET

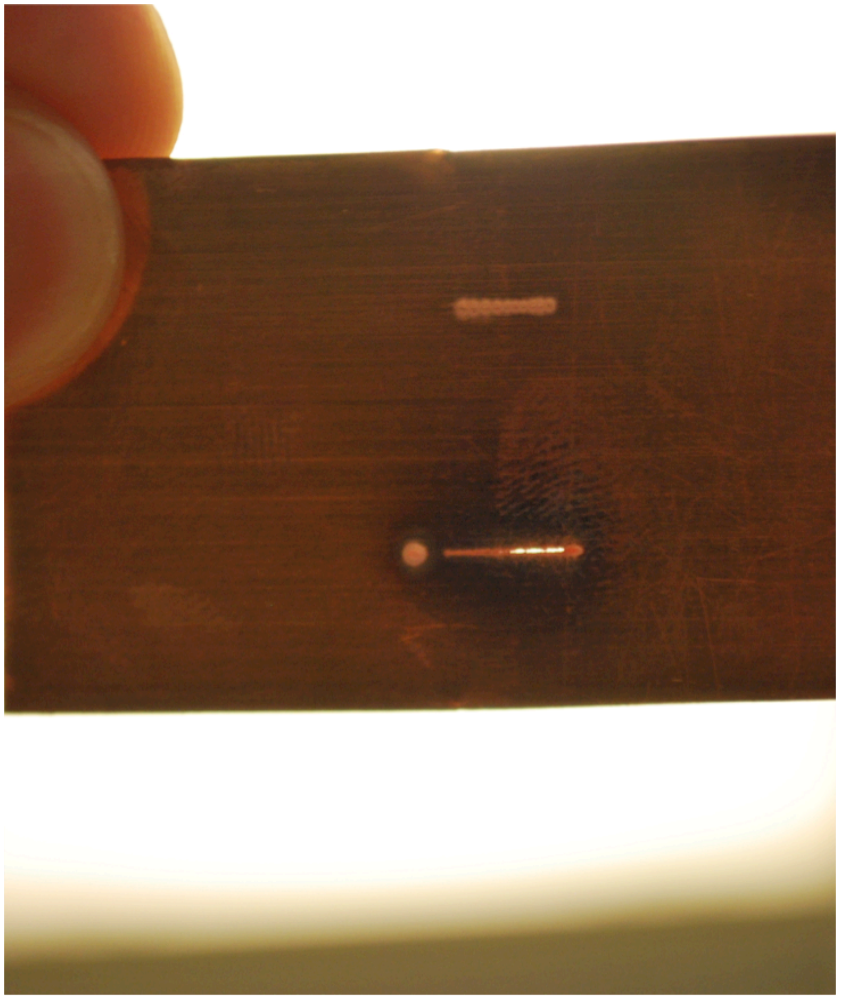
- For NEET to occur, the uranium atom has to be in an excited state
- It is predicted the highest NEET rate will occur when the uranium is in a charge state of 23+
- A high powered Q-switched Nd:YAG laser will be used to turn the uranium into a plasma
  - 650 mJ per pulse, 1064 nm, 10 Hz
  - 8 ns pulse width (normal), 2.5 ns pulse width (fast)
  - 10^{11}-10^{14} W/cm^2
- In order to have large signal to background, the uranium target will be highly enriched uranium (HEU)
- A charged plate will catch the uranium plasma
- The electrons emitted by the decay of the isomeric state will be focused onto a microchannel plate (MCP) detector by an Einzel lens

Current setup in new laboratory

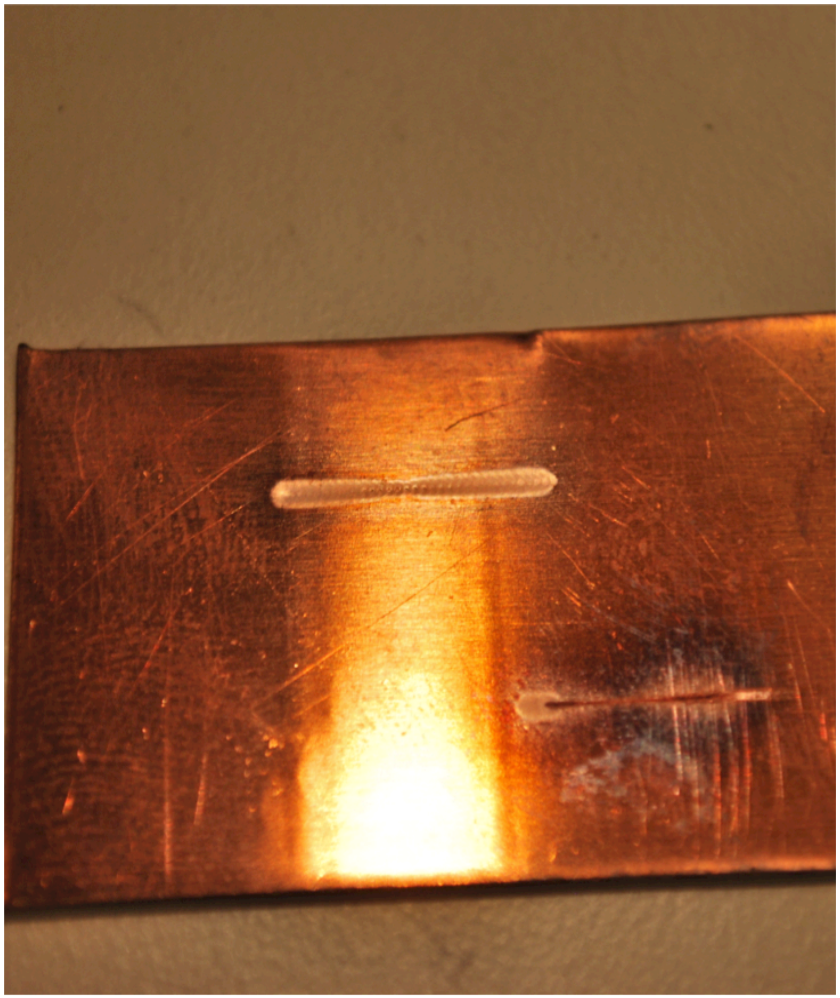


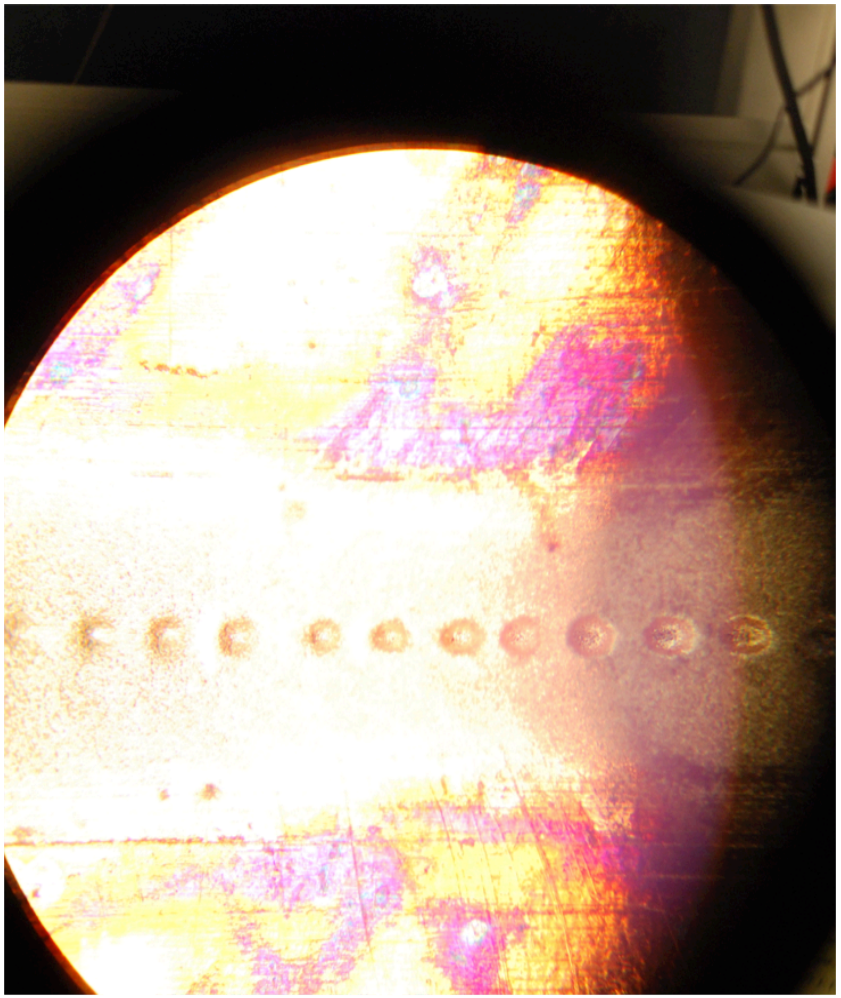
Targets post irradiation

- Spot sizes down to about 50 microns
- At 10 Hz laser induced plasma cuts through the target in less than a second
- 1st laser shot looks different than other craters



Surrogate targets: copper, aluminum….





$^{229m}Th/^{235m}U$ Studies Acknowledgements

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