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Actinide Studies with Ultracold Neutrons

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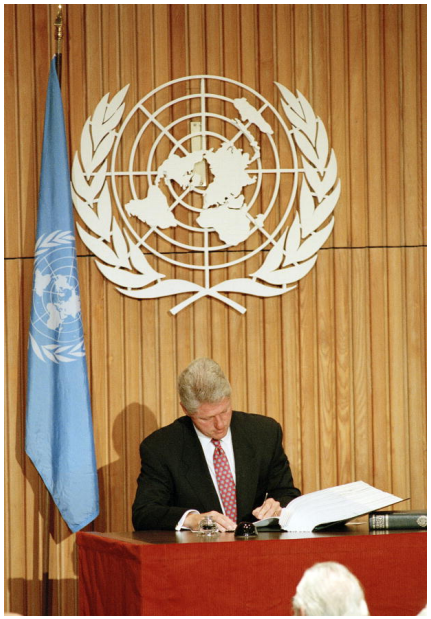
June 5, 2014

Nuclear Safety in the Modern Era

Comprehensive Nuclear Test-Ban Treaty

- United States signed on September 24, 1996
- Ratification dependent on **Science Based Stockpile Stewardship Program** to ensure safety and reliability of nuclear weapons in active stockpile



Stockpile stewardship

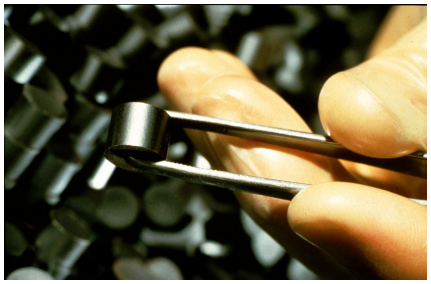
- No full-scale nuclear weapons testing since 1992
- How does aging of nuclear material affect reliability of nuclear weapons?
- NNSA mission: *Maintain the safety, security, and effectiveness of the nuclear deterrent without nuclear testing*

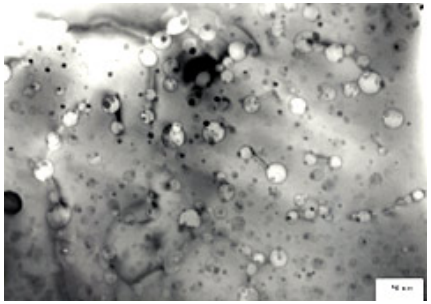
Major Effort at LANL

- DARHT: X-ray radiography for non-nuclear tests
- LANSCE: spallation neutrons
- pRAD: proton radiography
- TA-55: Pu operations

Aging of Nuclear Material

Nuclear energy





- Stopping of fission fragments in UO_2 = primary source of heating
- Irradiations causes "fission spikes" $\rightarrow$ atomic displacements
- Physical and chemical properties of fuel change over time: reduced thermal conductivity, enhanced diffusion, creep, and gas release
- Formation/resolution of fission gas bubbles
- UO_2 pellets in fuel rods: porosity gradually destroyed by fission tracks

Damage from Material Sputtering

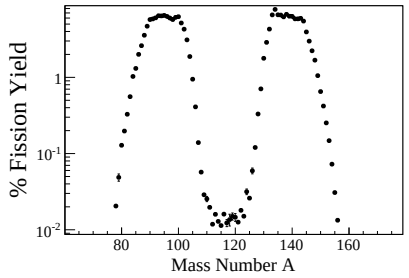
- Insulators vs. conducting material
- Semiconductors and metals in microelectronic components
- Space science: Predicting lifetime of unshielded detectors in space

Question: Effects of Fission on Surrounding Material?

Fission well studied

- Typically 2 fragments emitted
- A$\sim$100, E$\sim$100 MeV, $\frac{v}{c} \sim 10\%$, Range$\sim$10 μm

^{235}U fission fragments

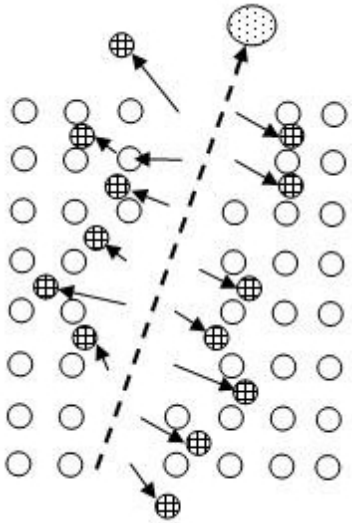


Effect of fission fragments: aging

- Damage to the material
- Sputtering of matter near surface

Description of energy transfer?

- $>$ 90% energy $\rightarrow$ electrons, rest nuclear collisions
- No complete QM description
- Competing models: Binary Collision Approximation, "Thermal Spike", "Coulomb Explosion", shock waves?
- Different predictions: yield, angular distribution of sputtered atoms
- Depth of fission fragments $\rightarrow$ sputtering



Inducing Fission with Ultracold Neutrons

Experimental evidence

- Many previous measurements of sputtered atoms per fission
- **Significant disagreement in yield, distribution!**
- Key to differentiating models

New Technique for understanding sputtering

- Induce fission using Ultracold Neutrons
- Excellent control of neutron energy
- Very sensitive probe of fission as function of depth

LANL: Unique Position for work

- LANSCE: one of world's brightest sources of UCN
- Expertise in fabrication and analysis of actinide targets

Ultracold Neutrons

Class	Energy	Source
Fast	> 1 MeV	Fission reactions / Spallation
Slow	eV – keV	Moderation
Thermal	0.025 ev	Thermal equilibrium
Cold	μeV – meV	Cold moderation
Ultracold	≤ 300 neV	Downscattering

How cold is Ultracold?

- Temperature < 4 mK
- Velocity < 8 m/s
- Usain Bolt ~ 12 m/s

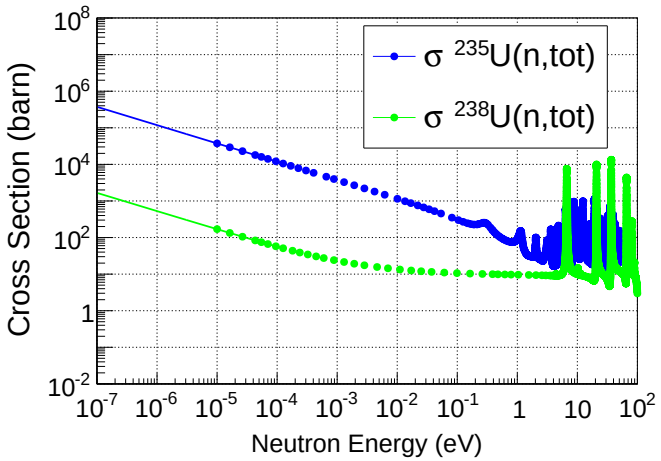
UCN can be bottled

- Gravitational ($V = mgh$): 100 neV / meter
- Magnetic ($V = -\vec{\mu} \cdot \vec{B}$): 60 neV / Tesla
- Material $\left(V = \frac{2\pi\hbar^2 Nb}{m}\right)$ $\begin{cases} {}^{58}\text{Ni}: & 335 \text{ neV} \\ \text{DLC}: & 250 \text{ neV} \\ \text{BeO}: & 250 \text{ neV} \\ \text{Cu}: & 170 \text{ neV} \end{cases}$

UCN-induced Fission in Uranium

Uncharted energy regime

- 3 orders of magnitude lower than ever explored
- Very high theoretical cross section $\sigma \sim \frac{1}{v}$
- 300 neV UCN: 2.16×10^5 barn



Control depth of fission event

Range of UCN in uranium (μm)						
	DU	NatU	SEU	LEU	HEU	VHEU
$\%^{235}$U	0.2	0.7	2	5	20	100
200 neV	118	66	312	13	4	0.8
300 neV	144	81	38	17	4.5	0.9
400 neV	191	101	45	20	5	1

Program Goals

Characterize sputtered material...

- Yield per fission
- Mass distribution
- Energy distribution
- Angular distribution

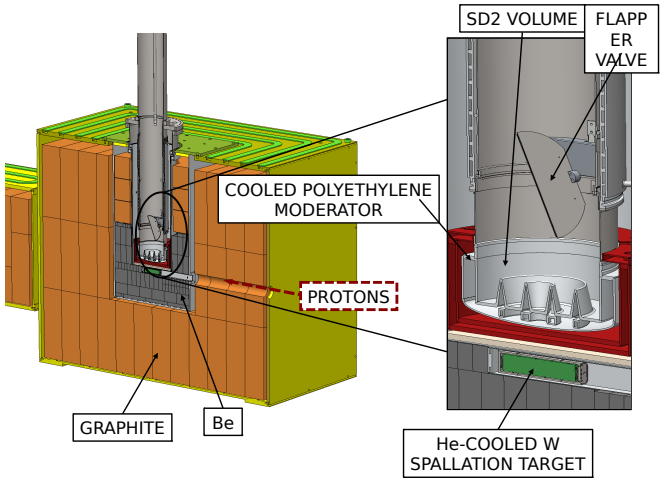
as a function of...

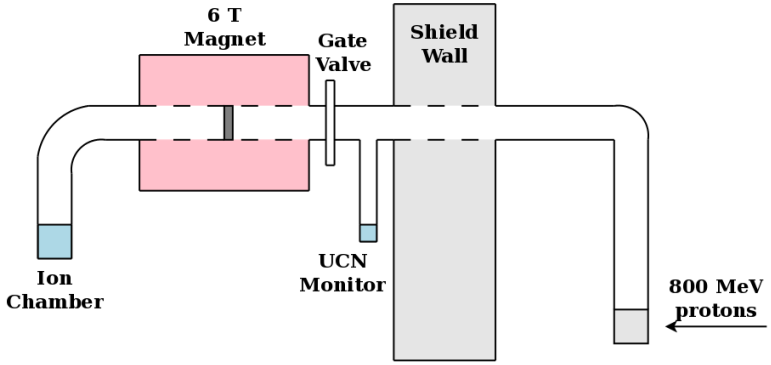
- UCN-energy
- Material thickness
- Material surface preparation
- Alloys, material layers
- Expand to more actinides

Important Preliminary Questions

1. Can we induce fission with UCN?
2. Can we detect sputtered material?
3. Do we see a dependence on UCN energy?

Ultracold Neutron Facility at LANSCE





UCN Source

- 800 MeV proton beam + W target → spallation neutrons
- single scatter in SD_2: CN→UCN + phonon
- High density at shield wall: 50 UCN/cc
- Pulsed beam: Low background

Experimental Area

- UCN bounce along guides, through Al window, into detection chamber
- 6 T magnet = near 100% polarization

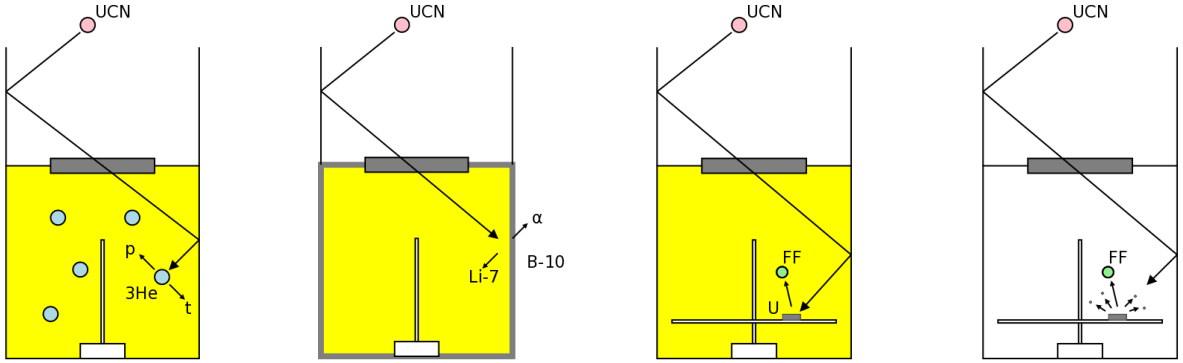
Detection system

Ion chamber

- Same detector used for normalization and fission studies
- UCN $\sim$ 50% transmission into detector

Modes of operation

- Mode 1: Counting ultracold neutrons
  - $^{3}\mathrm{He} + \mathrm{n} \rightarrow \mathrm{p} + \mathrm{t}$
  - $^{10}\mathrm{B} + \mathrm{n} \rightarrow \alpha + {}^{7}\mathrm{Li}$
- Mode 2: Counting fission fragments
- Mode 3: Exposure (sputtering)



Mode 1a Mode 1b Mode 2 Mode 3

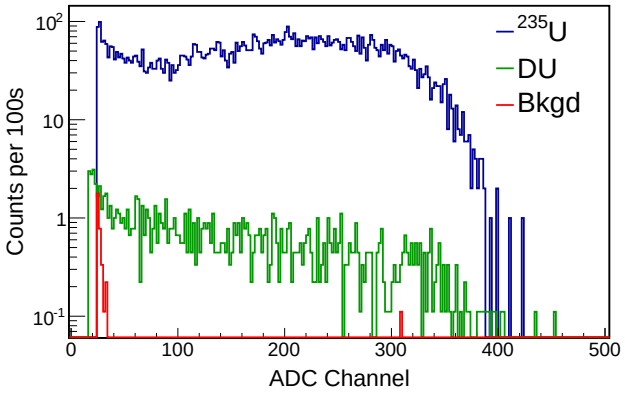
1. Can we induce fission with UCN?

^{238}U

- 2.25 cm diameter, 1 mm thick disk of DU ($\sim 0.2\%$ ^{235}U)
- Rate: 0.5 ± 0.3 fission/s
- $= (1.3 \pm 0.8) \times 10^{-4}$ fission/UCN

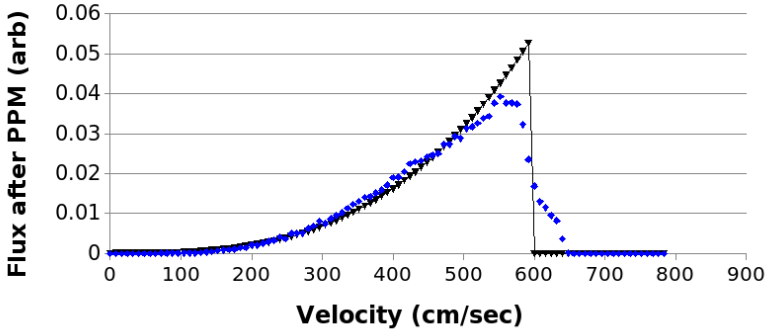
^{235}U

- 2.2 cm diameter, 1 mm thick disk of HEU ($> 80\%$ ^{235}U)
- Rate: 70.9 ± 0.8 fission/s
- $= (1.90 \pm 0.02) \times 10^{-2}$ fission/UCN



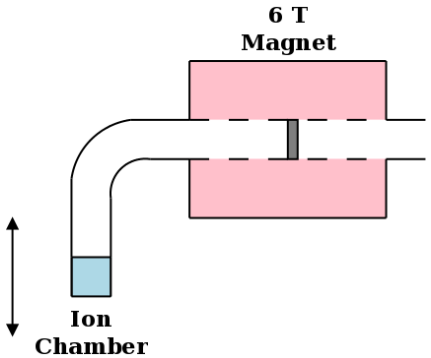
Varying UCN Energy

Simulated UCN energy spectrum after magnet



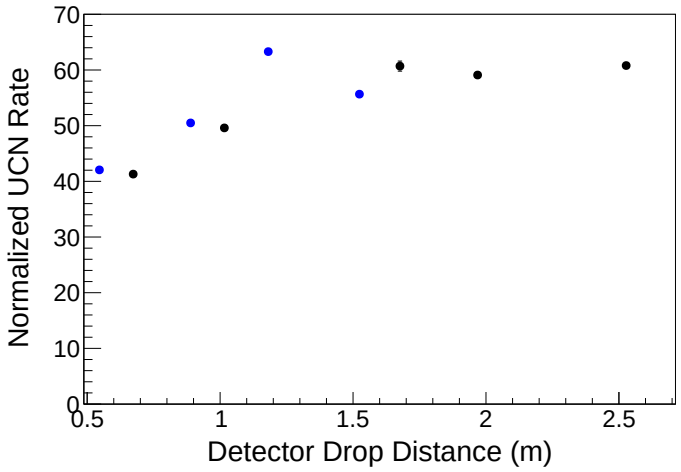
Two primary methods for control

- Magnetic field (60 neV/T)
- Gravity (100 neV/m)



Gravity scan

- Adjust height of ion chamber
- Sensitive to geometry, guide quality
- Al window: 50 neV barrier
- $^3He + n \rightarrow p + t$



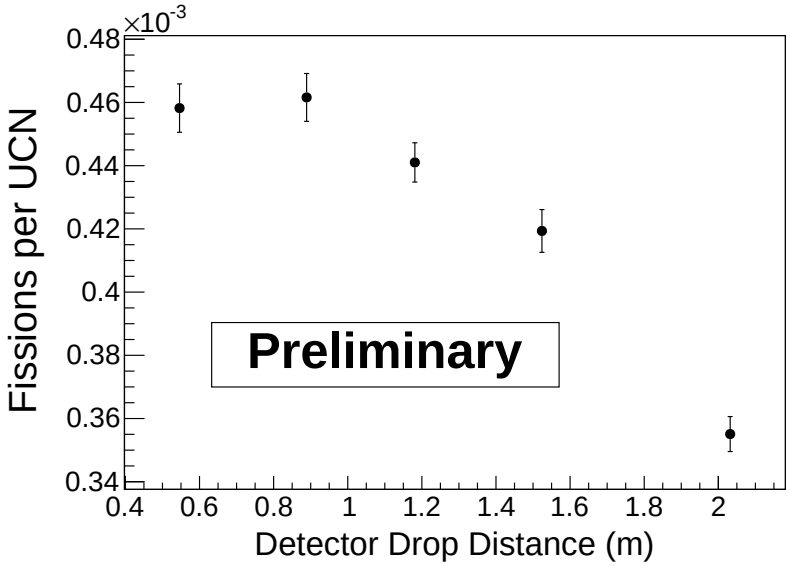
2. Do we see a dependence on UCN energy?

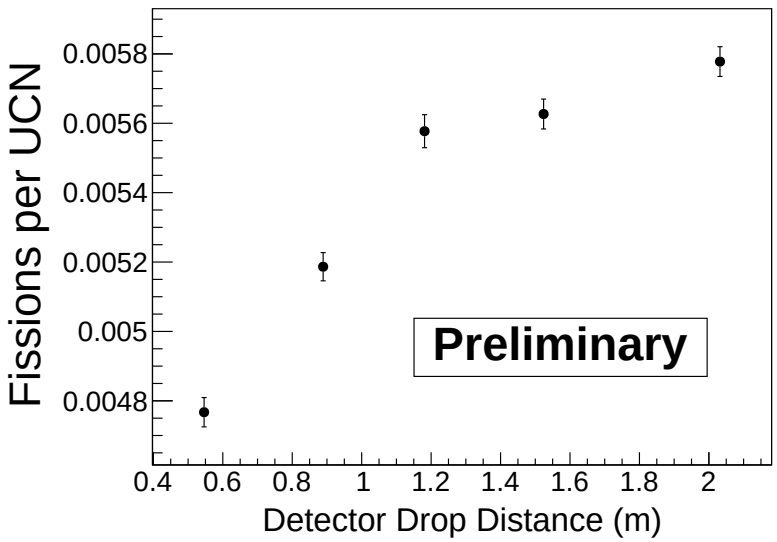
DU sample

- Electropolished, 2.25 cm diameter, 1 mm thick disk
- Measured fission rate decreases with UCN energy

^{235}U sample

- Thin sample: 30 μg heavily oxidized ^{235}U on tape
- Measured fission rate increases as UCN energy increases

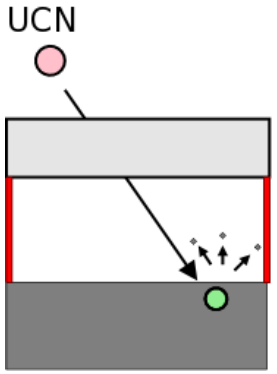




Sputtering from UCN-induced fission

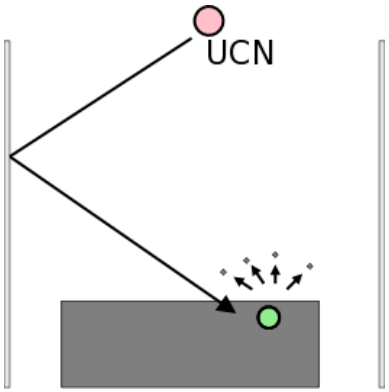
Exposure to Silicon wafer

- 1" diameter, 475 μm thick, polished wafer
- Exposed to 2.2 cm diameter, 1 mm thick electropolished DU disk
- 3×10^7 total UCN in chamber
- 0.18 μg ^{238}U collected on wafer
- Analyze with scanning probe microscopy



Exposure to Ni cylindrical foil

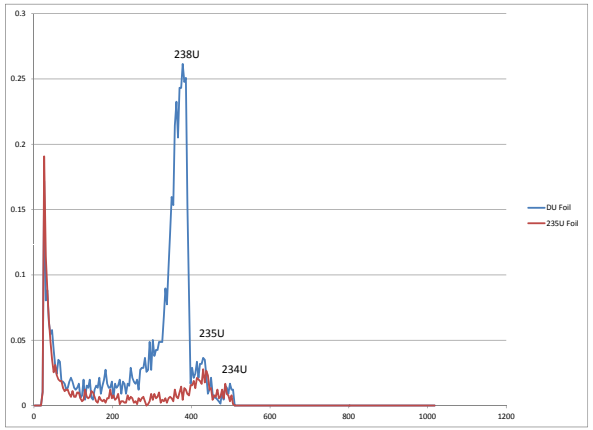
- 0.005" Ni foil, 1.15" diameter, 2.835" height
- UCN bottle: Ni material potential $\sim$ 300 neV
- Exposed to DU disk and 30 μg ^{235}U thin sample



Can we detect sputtered material?

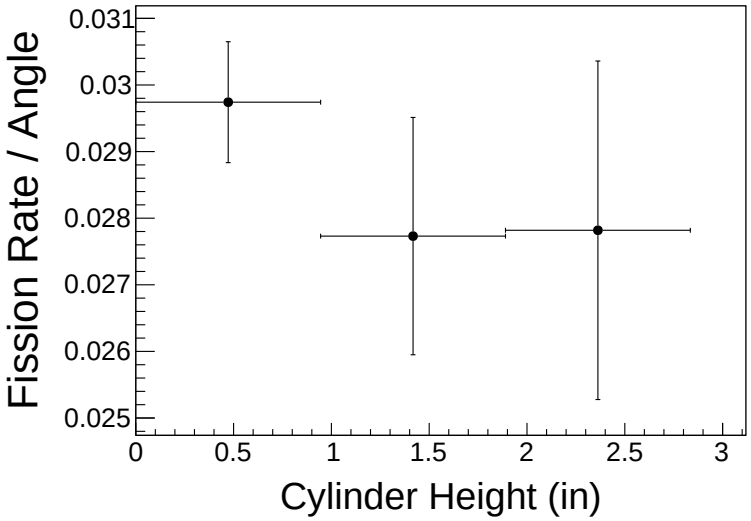
Expose to DU disk and HEU film

- Each sample exposed to $\sim 10^8$ UCN
- Determine yield from α decay rate in ion chamber
- Can distinguish α's from ^{238}U, ^{235}U decay

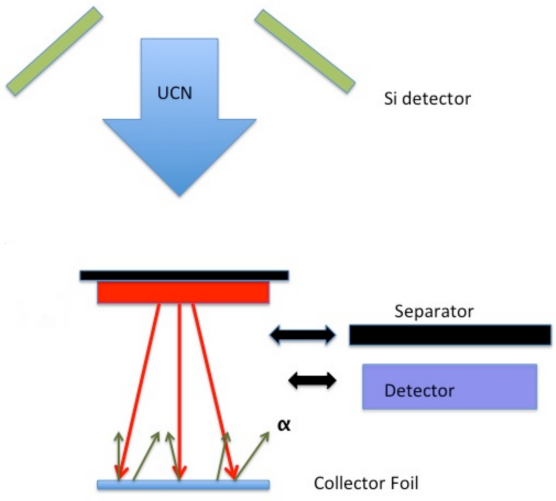


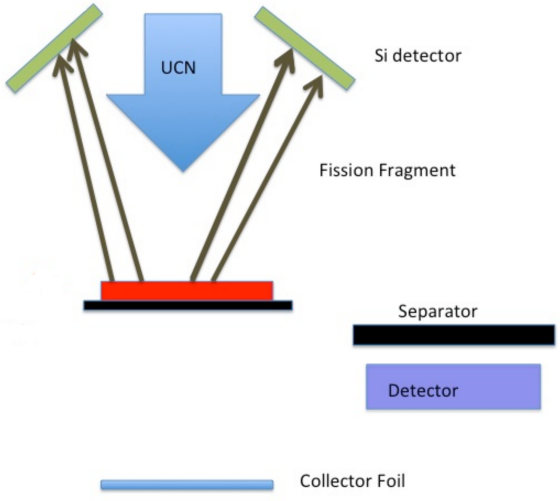
Angular distribution

- Mask vertical sections of foil
- Results from DU foil
- Distribution $\sim$ isotropic



Next: Characterize Ejected Material In-Situ





Key questions:

- How much comes off?
- Size distribution vs. depth/surface quality?
- Kinetics vs. depth?
- Effect of surface preparation?

Summary

First characterization of UCN-induced fission

- Previously no fission data at these energies
- Initial characterizations of UCN energy dependence, material thickness

First observation of sputtering from UCN-induced fission

- Proof of principle demonstrated
- Analysis of sputtered rates, angular and size distribution underway

Future plans:

- Determine absolute cross sections
- Develop scheme for characterizing ejected material
- Explore effect of material thickness, surface preparation in more detail
- Develop path forward for plutonium and other actinides