Studies of Fission-Induced Surface Damage in Actinides Using Ultracold Neutrons

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Fission-Induced Damage

**Sputtering**

- Fission event: 2 fragments, $E \sim 100$ MeV, $A \sim 100$
- Fast, heavy charged particles $\rightarrow$ ejection of atoms
- Damage to material surface

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**Not well understood**

- Underlying mechanism?
- Sputtered atoms per fission event?
- Damage to the material surface?
- Competing models?
- Quality of surface (oxide layer)?
- Sputtering from “deep” fissions ($\sim 10 \ \mu m$)?

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**Aging of Nuclear Materials**

- Reactor fuel rods
- Satellites: thin film on batteries
- Stockpile stewardship
Ultracold Neutrons

<table>
<thead>
<tr>
<th>Class</th>
<th>Energy</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fast</td>
<td>$&gt; 1$ MeV</td>
<td>Fission reactions / Spallation</td>
</tr>
<tr>
<td>Slow</td>
<td>eV – keV</td>
<td>Moderation</td>
</tr>
<tr>
<td>Thermal</td>
<td>0.025 ev</td>
<td>Thermal equilibrium</td>
</tr>
<tr>
<td>Cold</td>
<td>$\mu$eV – meV</td>
<td>Cold moderation</td>
</tr>
<tr>
<td>Ultracold</td>
<td>$\leq 300$ neV</td>
<td>Downscattering</td>
</tr>
</tbody>
</table>

How cold is Ultracold?

- Temperature $< 4$ mK
- Velocity $< 8$ m/s
- Usain Bolt $\sim 12$ m/s

UCN can be bottled

- Gravitational ($V = mgh$): 100 neV / meter
- Magnetic ($V = -\vec{\mu} \cdot \vec{B}$): 60 neV / Tesla

Material ($V = \frac{2\pi \hbar^2 N_b}{m}$)

- $^{58}$Ni : 335 neV
- DLC : 250 neV
- BeO : 250 neV
- Cu : 170 neV
UCN-Induced Fission

Very high cross section: \( \sigma \sim \frac{1}{V} \)

Cross Section (barns)

<table>
<thead>
<tr>
<th>UCN Energy</th>
<th>200 neV</th>
<th>300 neV</th>
<th>400 neV</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{235}\text{U}(\text{n,tot}) )</td>
<td>( 2.64 \times 10^5 )</td>
<td>( 2.16 \times 10^5 )</td>
<td>( 1.87 \times 10^5 )</td>
</tr>
<tr>
<td>( ^{238}\text{U}(\text{n,tot}) )</td>
<td>( 1.17 \times 10^3 )</td>
<td>( 9.57 \times 10^2 )</td>
<td>( 8.29 \times 10^2 )</td>
</tr>
</tbody>
</table>

Finely tune depth into material

<table>
<thead>
<tr>
<th>Comp.</th>
<th>% ( ^{235}\text{U} )</th>
<th>200 neV</th>
<th>300 neV</th>
<th>400 neV</th>
</tr>
</thead>
<tbody>
<tr>
<td>DU</td>
<td>0.2%</td>
<td>118</td>
<td>144</td>
<td>191</td>
</tr>
<tr>
<td>NatU</td>
<td>0.7%</td>
<td>66</td>
<td>81</td>
<td>101</td>
</tr>
<tr>
<td>SEU</td>
<td>2%</td>
<td>31</td>
<td>38</td>
<td>45</td>
</tr>
<tr>
<td>LEU</td>
<td>5%</td>
<td>13</td>
<td>17</td>
<td>20</td>
</tr>
<tr>
<td>HEU</td>
<td>20%</td>
<td>4</td>
<td>4.5</td>
<td>5</td>
</tr>
<tr>
<td>100%</td>
<td></td>
<td>0.8</td>
<td>0.9</td>
<td>1</td>
</tr>
</tbody>
</table>
**UCN Source at LANSCE**

**UCN Source\(^1\)**

- UCN Source: 800 MeV proton beam + Tungsten target = CN
- CN downscatter in SD\(_2\) crystal = UCN
- UCN Monitor = Normalize for fluctuations in UCN production

**Detection**

- Gate valve permits UCN entry to experiment
- 6 T magnet = near 100% polarization
- UCN drop through Al window into ion chamber

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UCN Rate Normalization

UCN Beam Monitor\(^2\)

- \(^3\)He filled multi-wire proportional chamber
- \(^3\)He + n → p (573 keV) + t (191 keV)
- 50% transmission through window into detector; 80% efficient

Baseline UCN Rates\(^3\)

- Boron-coated cylindrical ion chamber, 1 barr argon
- \(^{10}\)B + n → α + \(^7\)Li
- Near 100% efficient for UCN entering chamber
- Rate: 4.5kHz (for 125 Hz beam monitor rate)


Proof of Concept: Fission Rate

Experiment

- Identical experimental setup
- Cylindrical ion chamber with boron coating removed
- Effect of UCN bottling?
- 200 mbarr argon: α’s range out

$^{235}\text{U}$

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

$^{238}\text{U}$

- 2.25 cm diameter, 1 mm thick disk of Depleted Uranium ($\sim 0.2\%^{235}\text{U}$)
- Rate: $(1.3 \pm 0.8) \times 10^{-4}$ fission/UCN
Neutron Capture Gammas

HPGe detector

- Calibration: $^{60}\text{Co}$ and $^{137}\text{Cs}$ gamma sources
- Goal: tag gamma, look for fission

Observed Spectra

- Empty chamber with/without UCN: additional 480 keV line from residual Boron coating
- Decay gammas from $^{235}\text{U}/^{238}\text{U}$ observed; some additional lines
- No additional gamma lines with UCN?
Neutron Spin Dependence

Neutron Polarization

- 6 T Magnet: near 100% UCN polarization
- Neutron spin aligned with field

Experiment

- Neodymium magnets installed on chamber: $\vec{B}$ field normal and parallel to surface
- $\sim 200$G field normal to surface: $(1.92 \pm 0.02) \times 10^{-2}$ fission/UCN
- $\sim 50$G field parallel to surface: $(1.94 \pm 0.02) \times 10^{-2}$ fission/UCN
- No magnets: $(1.90 \pm 0.02) \times 10^{-2}$ fission/UCN
Sputtering

Evidence of UCN-induced sputtering?

- Installed $^{235}$U for $\sim$20 minutes
- Exposed to UCN for $\sim$10 minutes
- Removed sample: small signal still observed!
- $\alpha$ rate = $2.63\pm0.07$ Hz ($\sim 10^{17}$ atoms)

Check: No UCN exposure

- $^{235}$U installed on removable copper plate: reduce chance of contamination
- Installed for $\sim$15 minutes, not exposed to UCN
- Removed copper plate with sample
- $\alpha$ rate = $0.78\pm0.04$ Hz ($\sim 10^{16}$ atoms)
- Inconclusive: contamination? $\alpha$-induced sputtering? chamber pumping/pressurizing?
Characterize Ejected Material

Important questions:

- How much comes off?
- Size distribution vs. depth/surface quality?
- Kinetics vs. depth?
Summary

First observation of UCN-induced fission

- Previously no fission data at these energies
- Determine relative cross-sections (e.g. vanadium sample)

Next: Confirm UCN-induced sputtering

- Sputtered rate as function of exposure time
- Better sample mounting: eliminate possibility of contamination
- Electropolish sample: clean, well-understood surface

Program Goals

- Characterize sputtered ejecta from various actinides
- Control fission depth via UCN energy: gravity/magnetic potentials
- Understand effect of depth and surface quality
- Examine different alloys, material layers