SCIENCE AND APPLICATION OF NUCLEAR RESONANCE FLUORESCENCE: MEASUREMENT OF ^{237}NP

C.T. Angell, R. Yee, T. Joshi, E.B. Norman

UC Berkeley

W.D. Kulp

GeorgiaTech

G. Warren

PNNL

C. Hicks, S. Korbly, A. Klimenko, C. Wilson

Passport Systems

R. Copping, D. K. Shuh

LBNL

Advanced Photons and Science Evolution 2010

June 16, 2010, Osaka, Japan

^{237}Np NRF

- Analog in atomic systems
  - Surface plasmon resonance
- **Nuclear Resonance Fluorescence**
  - Detection and Assay
  - ^{237}Np
- Experiment
  - Setup
  - Results
  - Comparison to other actinides

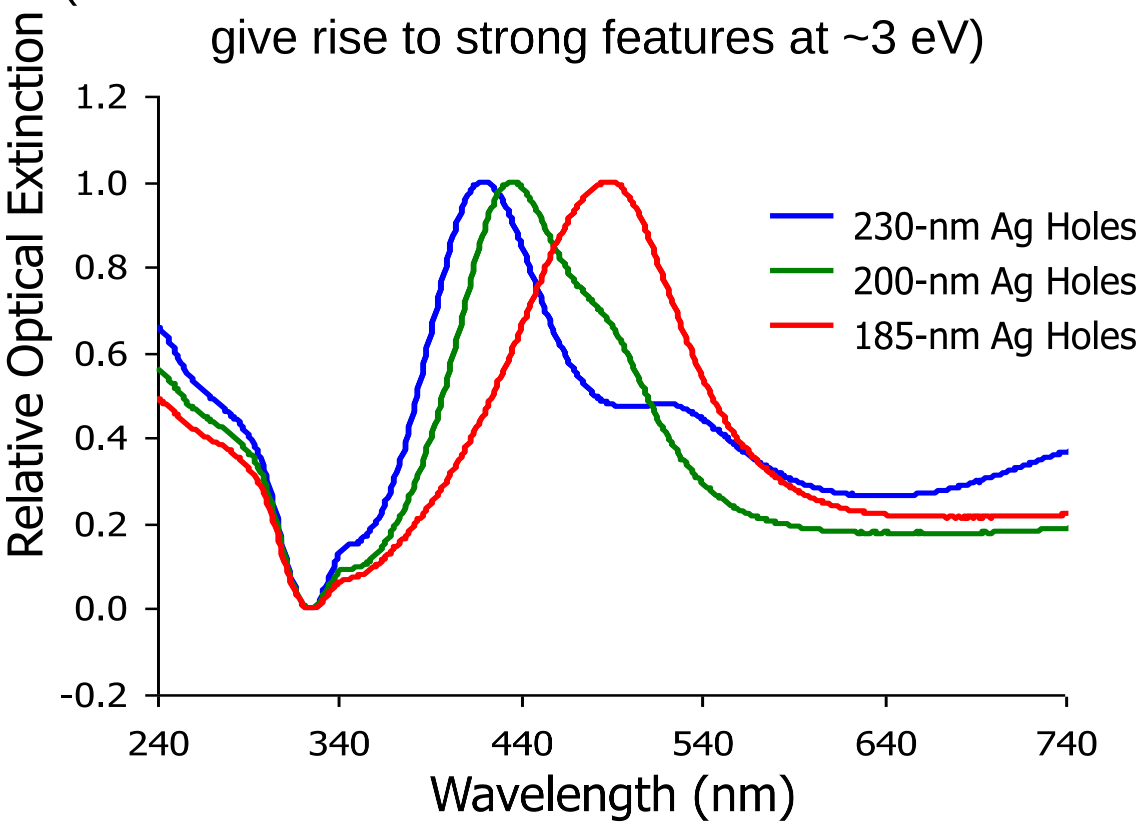
Surface Plasmon Resonance

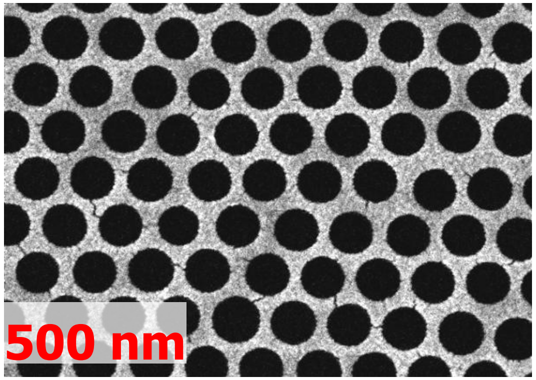
Courtesy of Robert Baker, UC Berkeley

- Nanoscale features in metal show resonant oscillation of electrons at a fixed frequency.
- Width ~ 100 nm (1-3 eV).
- Analogous to nuclear excitation with photons

Optical Spectra of Nano-Holes in Ag Films

(Resonant oscillation of conduction band electrons give rise to strong features at ~3 eV)

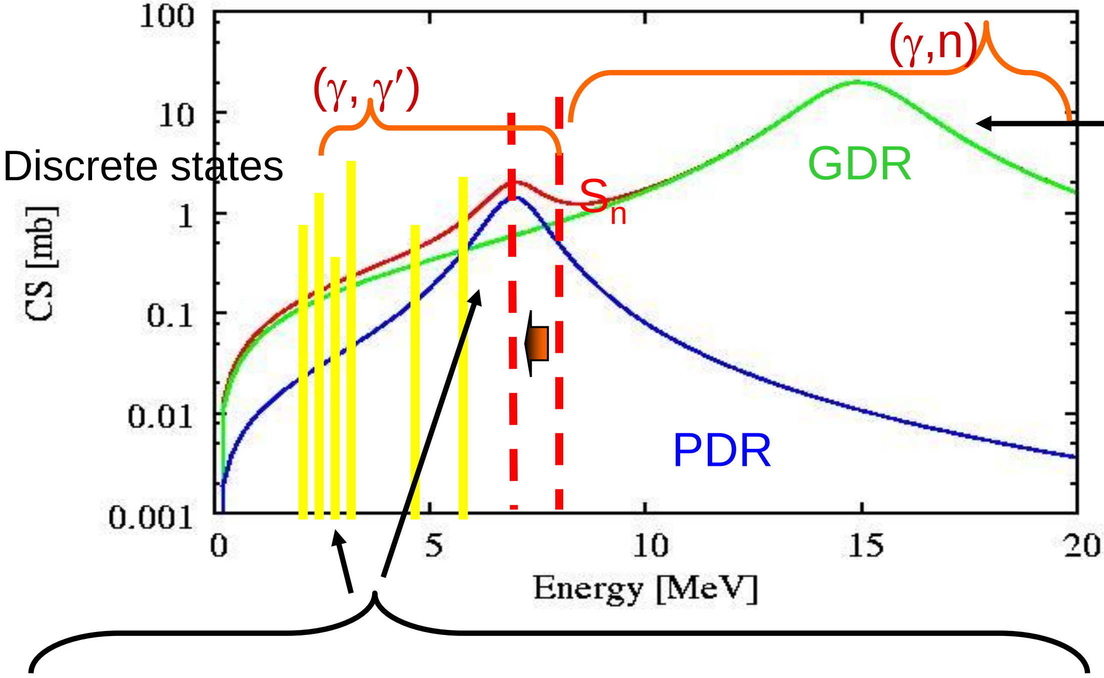




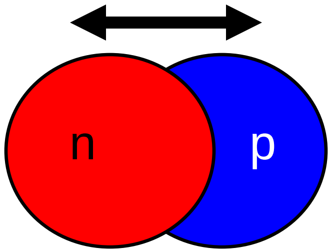
Array of nano-holes in a Ag film

Dipole Resonances: Giant and Pygmy

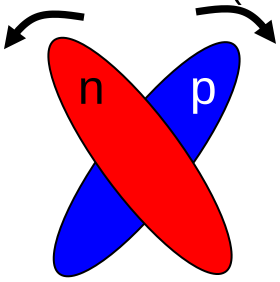
Total photoabsorption cross section



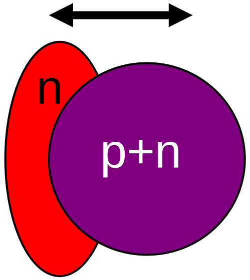
Giant E1 Resonance



Scissors Mode (M1)



Neutron Skin Oscillation (E1)

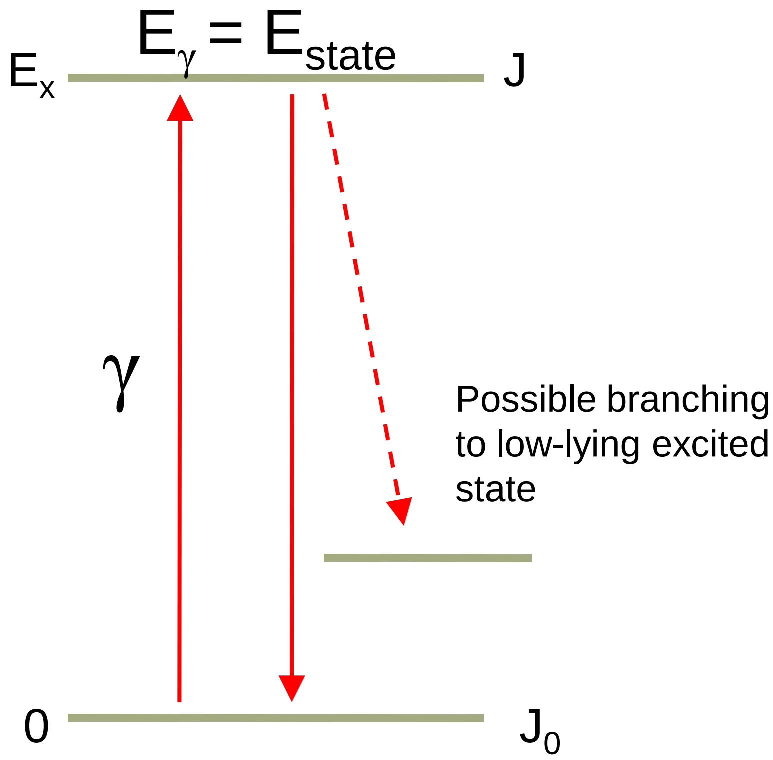


- E1 photoabsorption cross section is dominated by the Giant Dipole Resonance (GDR)
- The low energy extrapolation of the GDR determines the photo-absorption cross section
- Lorentzian shape used normally
- Can measure discrete states at low energy (1 – 3 MeV)

Nuclear Resonance Fluorescence

- Resonantly excite state with γ rays
  - States are narrow:

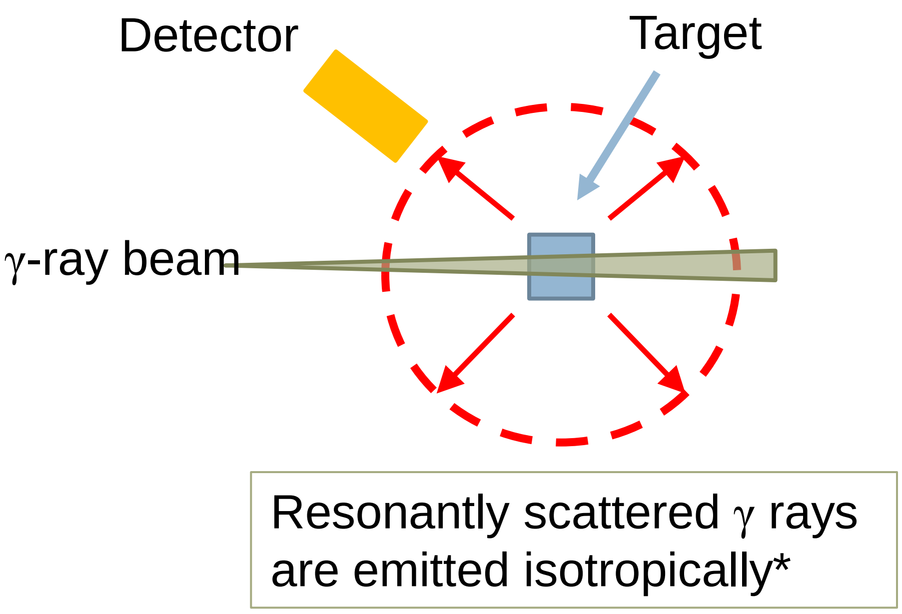
    $\Gamma \sim 10 - 100$ meV
- De-excites emitting characteristic γ ray
- Unique identifier of nucleus
  - "Thumbprint" for each isotope of interest
- Only *E1, M1, E2* transitions excited
  - $\Delta J \leq 2$



$$I_{cs} = \frac{2J+1}{2J_0+1}\left(\frac{\pi\hbar c}{E_\gamma}\right)^2 \frac{\Gamma_0 \Gamma_f}{\Gamma}$$

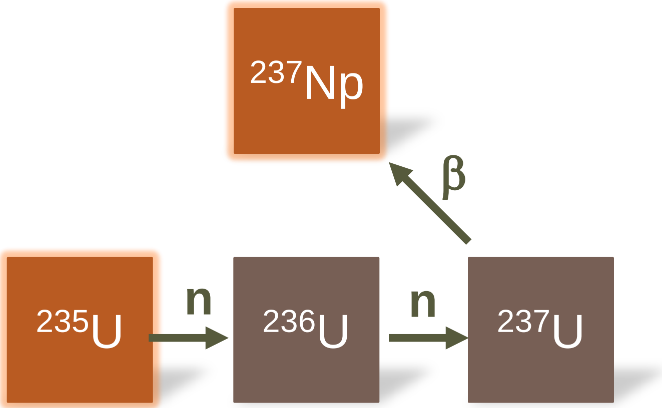
NRF Applications

- Unique identifier of nucleus
  - Can distinguish between fissile materials
- Actively interrogate material
- γ rays are highly penetrating
- Non-destructive
- Applications:
  - Non-invasive assay
    - Spent fuel rods
  - Active Interrogation
    - Cargo containers



Neptunium-237

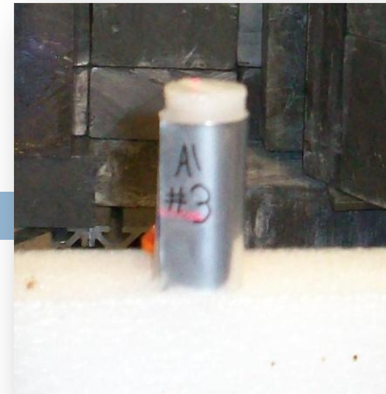
- Made in nuclear reactors from ^{235}U
  - $$^{235}_{92}\mathrm{U} + ^{1}_{0}\mathrm{n} \longrightarrow ^{236}_{92}\mathrm{U}_{\mathrm{m}} \xrightarrow[120\ \mathrm{ns}]{} ^{236}_{92}\mathrm{U} + \gamma$$
    $$^{236}_{92}\mathrm{U} + ^{1}_{0}\mathrm{n} \longrightarrow ^{237}_{92}\mathrm{U} \xrightarrow[6.75\ \mathrm{d}]{\beta^-} ^{237}_{93}\mathrm{Np}$$



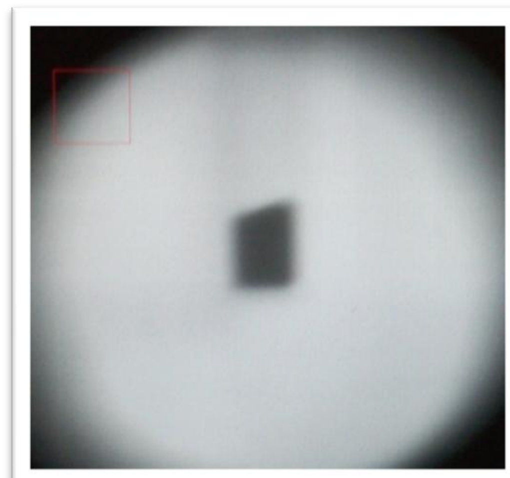
- One of the longest lived components of spent nuclear fuel
  - $T_{1/2}$ = 2.1 million years
- Fissile – yet not under stringent IAEA safeguards
  - Techniques needed to detect and assay it

^{237}Np criticality experiment at LANL

Target



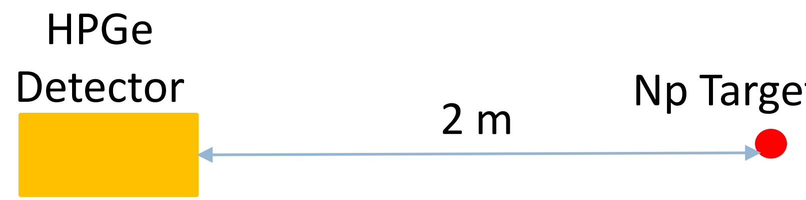
- Target: **^{237}Np** – (NpO_2)
  - 9.6 g total (from assay)
- Powder doubly encapsulated in plastic cylinders
- 8.8 g ^{181}Ta - in beam calibration
  - Foil wrapped around target
  - Flux normalizer



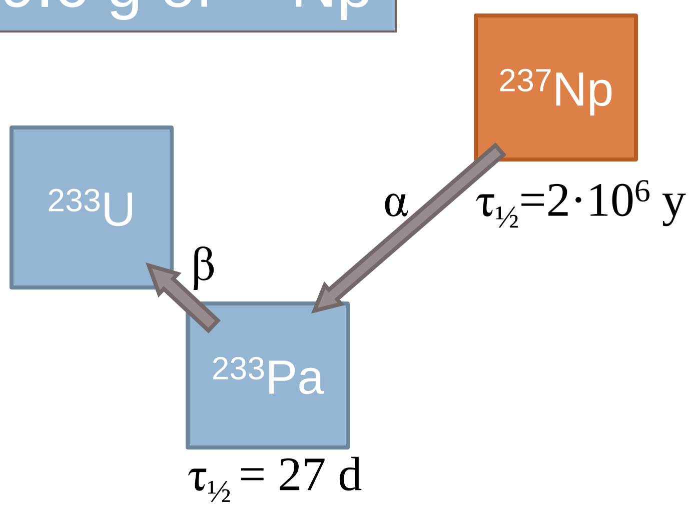
In-beam Radiograph of ^{237}Np Target

Assay of Np Target

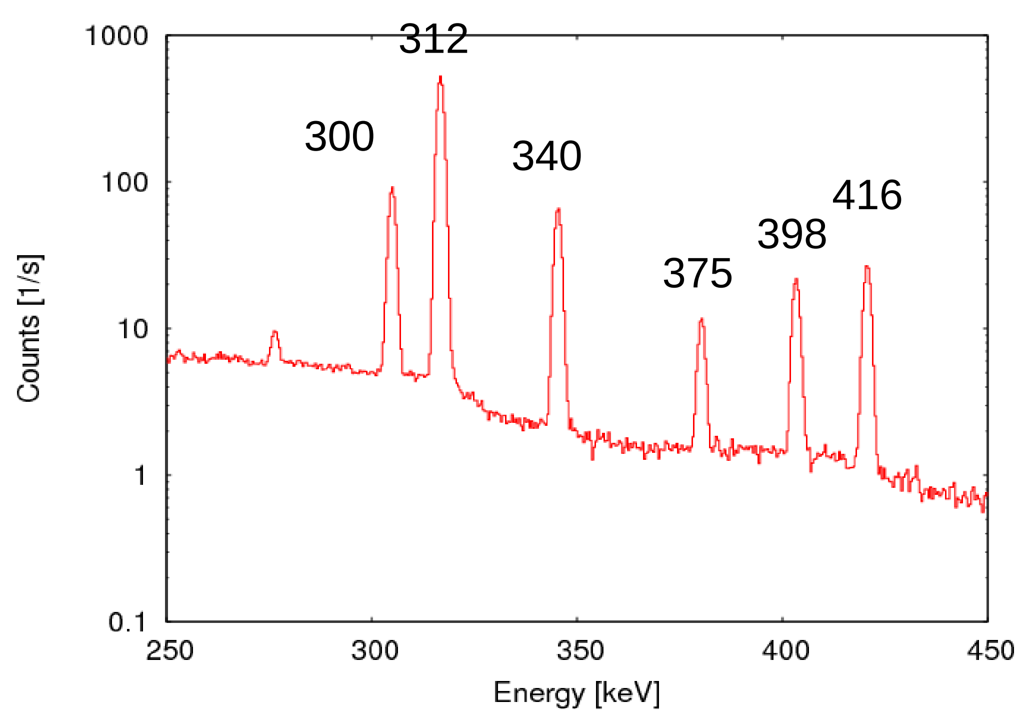
- Target composition initially unknown
- Activity: 7 mCi
  - Detector rate <u>over 1 kHz</u> at 2 m
- Used ^{233}Pa - secular equilibrium



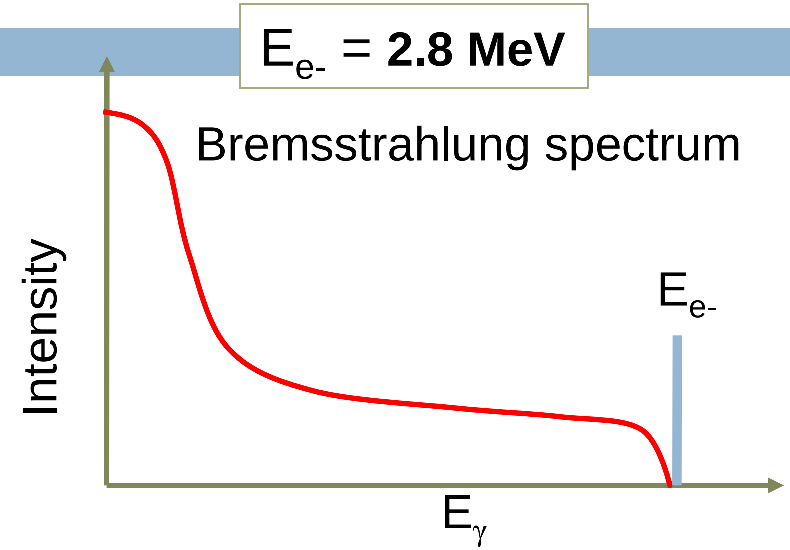
9.6 g of ^{237}Np



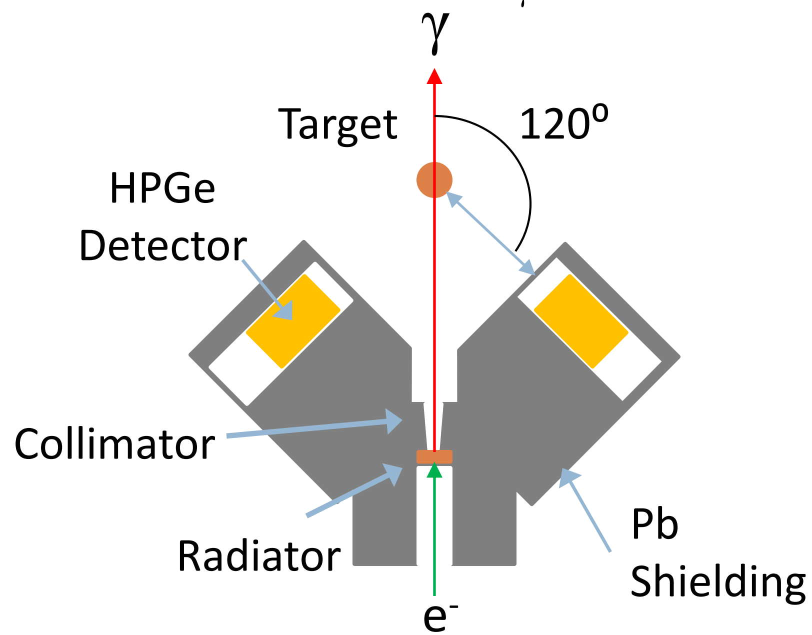
^{233}Pa γ rays



Setup at the High Voltage Research Lab



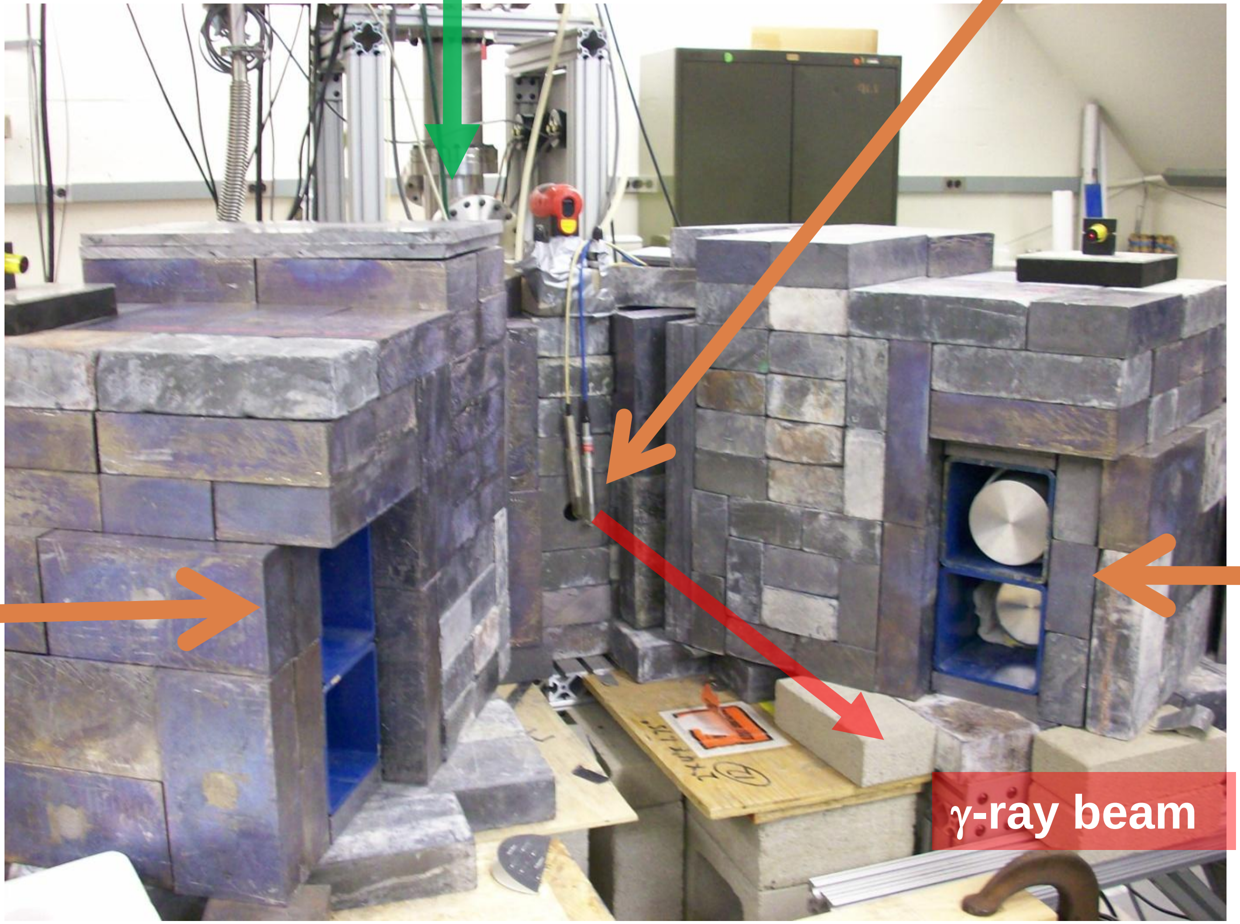
- γ ray source : Bremsstrahlung
  - Radiation from stopping electrons
- Electron beam -Van De Graff accelerator at the HVRL at MIT
- Radiator
  - thin layer of gold on copper
- 4 HPGe detectors
  - Two detectors on each side
- Pb Shielding
  - 60 cm Pb between radiator and detectors
  - 1.3 cm Pb in front of detectors



Experimental Setup

Electrons (to radiator after 90° bend)

Collimator



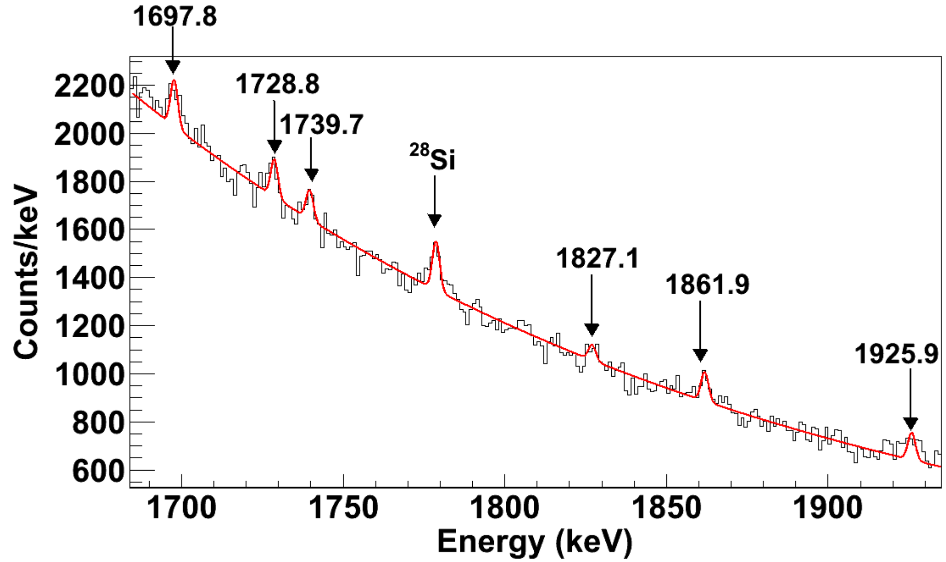
Detectors

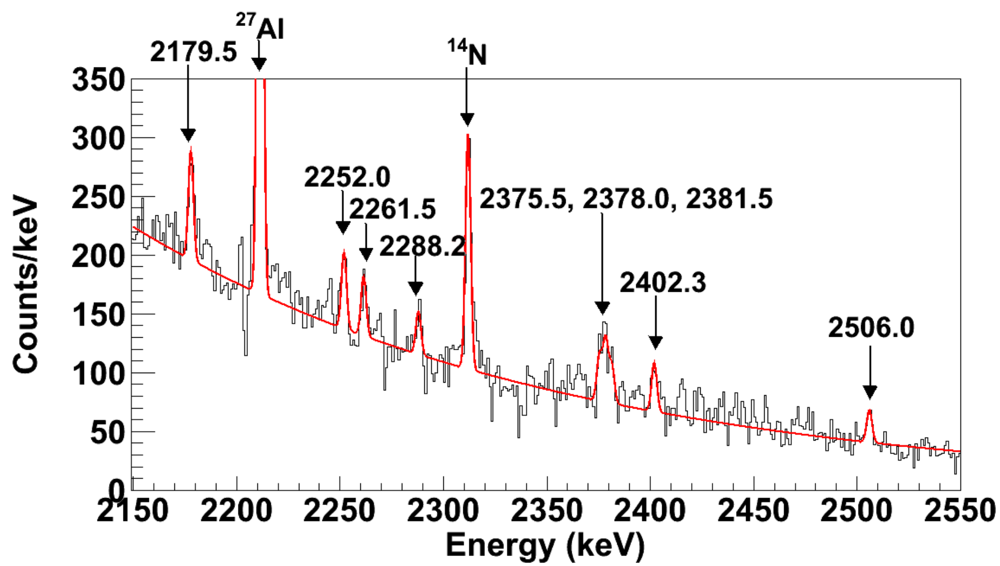
Detectors

^{237}Np Measured Spectra and States

Energy [keV]	Energy [keV]
1697	2261
1728	2288
1739	2376
1828	2378
1862	2381
1926	2403
2180	2506
2252	

Run length (Np): 13 Hours
Run length (Np+Ta): 10 Hours
Run length (Ta): 6 Hours



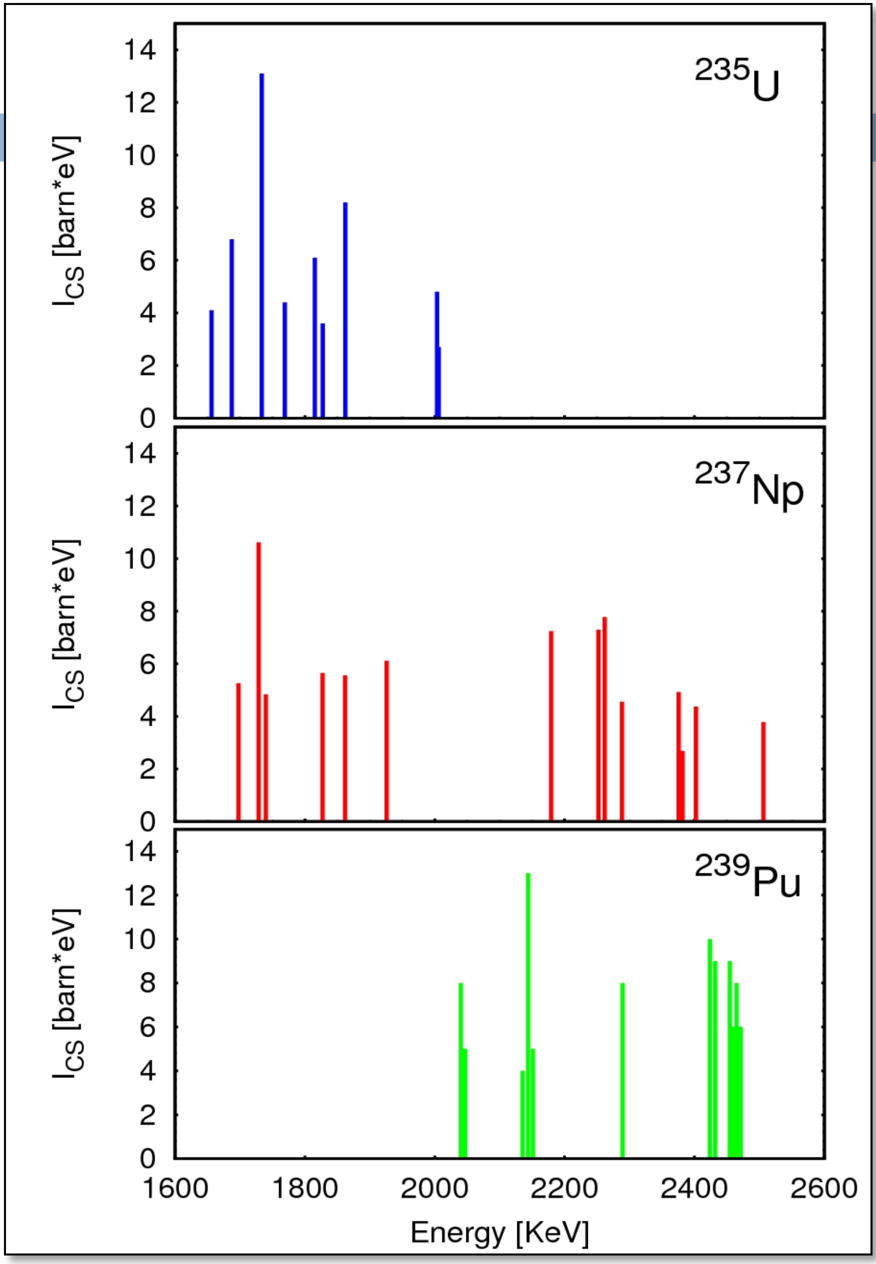


^{237}Np NRF: Comparison to other fissile actinides

- Measured 15 new states in ^{237}Np
- Integrated cross section:

$$I_{cs} = \frac{2J+1}{2J_0+1}\left(\frac{\pi\hbar c}{E_\gamma}\right)^2 \frac{\Gamma_0 \Gamma_f}{\Gamma}$$

- Comparison to ^{235}U / ^{239}Pu:
  - States are distributed wider in energy
  - Similar I_{cs}
- States discovered can be used to assay and detect ^{237}Np



Possible Scissors Mode?

- Assume M1 excitations only
- Calculate ∑B(M1↑)
  - For each state:

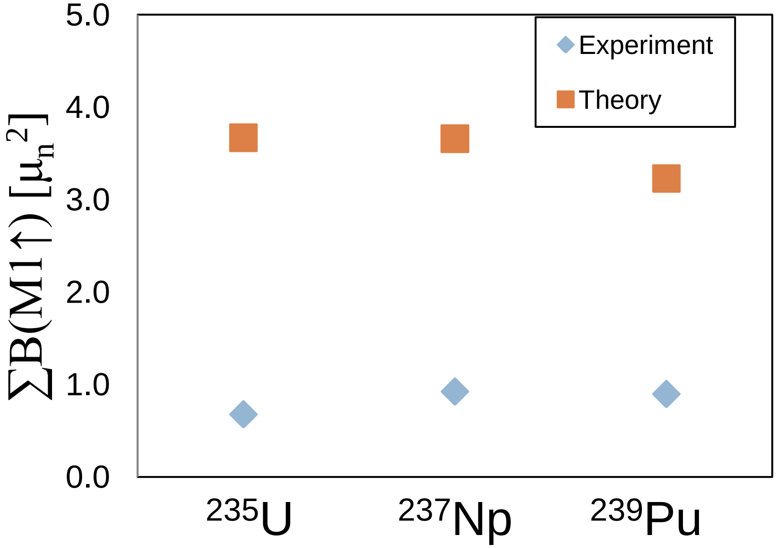
$$B(M1\uparrow) = 2.253\times10^{-4}\frac{I_s}{E_\gamma}$$

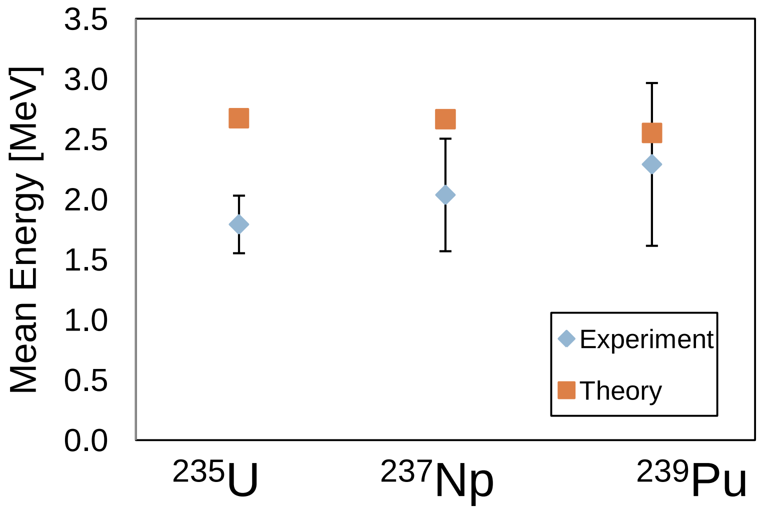
  - From theory use Lo Iodice & Richter scissors sum rule
    - Used HFB caluclated deformation, δ, from RIPL-II
    - Mean excitation energies taken from systematics
- Calculate mean energy
  - Mean weighted by B(M1↑) strength for each state
  - Scissors mode systematics (from rare earth elements):

$$E_x = 66\cdot\delta\cdot A^{-1/3}$$

Possible Scissors Mode?

- $\sum B(M1\uparrow)$ and theory
  - Assuming all the measured strength is M1:
  - Most strength missing, possibly from high fragmentation
  - Missing states at higher energies and/or below limit of detectability
- Mean energy and systematics
  - Opposite trend seen – experimental excitation energies increase while those from systematics drops
  - Possibly not scissors mode?





Conclusions

- First measurement of NRF on ^{237}Np
- 15 new peaks found
- Can be used to identify and assay ^{237}Np
- Similar in structure and strength to those states identified in ^{235}U and ^{239}Pu

- Funded by the US DHS – Domestic Nuclear Detection Office

Integrated Cross Sections I

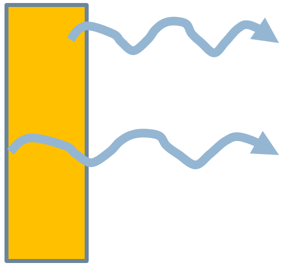
$$I_{\mathrm{CS}} = \frac{N_{\mathrm{peak}}}{\varepsilon_{ff}\,\varphi_{flux}\,f_{\mathrm{target}}\,f_{\mathrm{pb}}N_{t}}$$

- $\varepsilon(E_\gamma)$ - efficiency
- $\sigma_{\mathrm{Schiff}}(E_\gamma, E_e)$– Schiff cross section for Bremmstrahlung
- $f(E_\gamma)$ – attenuation factor for beam through target (in and out), and through Pb attenuators
- N^{target} – number of target nuclei
- $\alpha(E_e)$ – Flux normalization constant

Target Mass

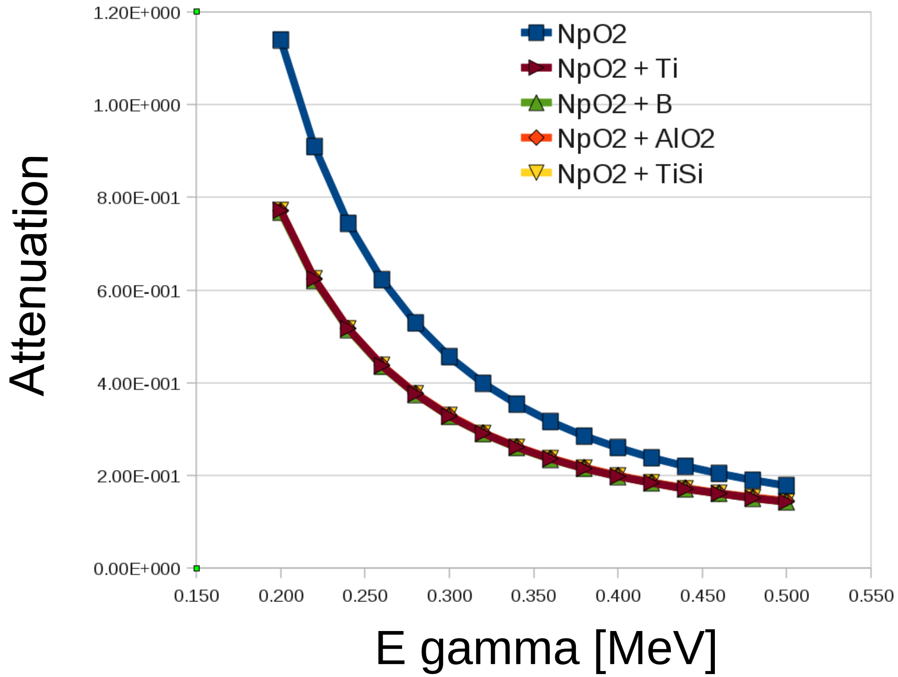
- Target 1 had unknown component
- Must determine mass from assay
- Unknown composition
- Unknown density
- Use iterative method based on differences in attenuation in 5 gamma rays from ^{233}Pa

Different depth,
Different attenuation

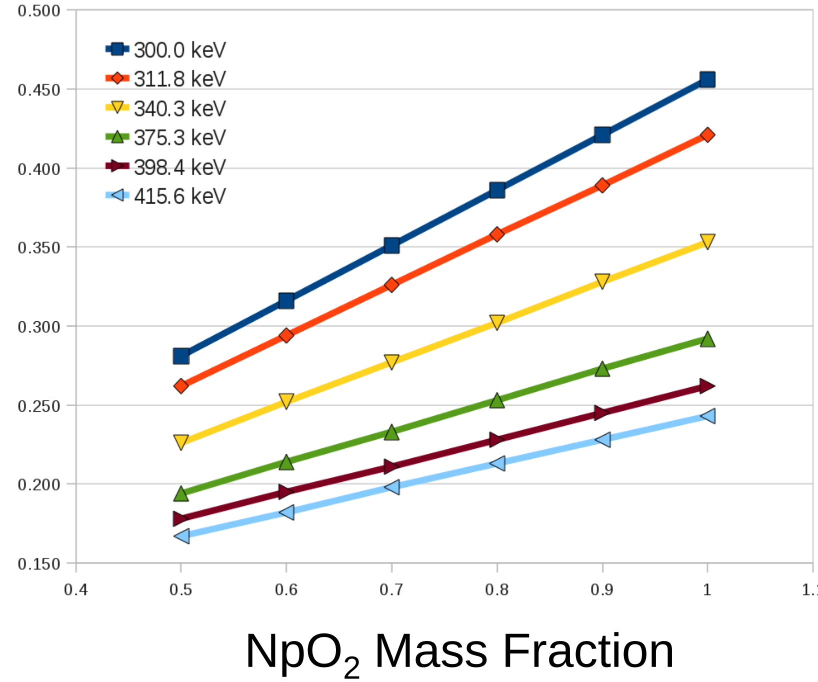


Attenuation Factor for NpO_2

Attenuation factor doesn't depend on light element constituent

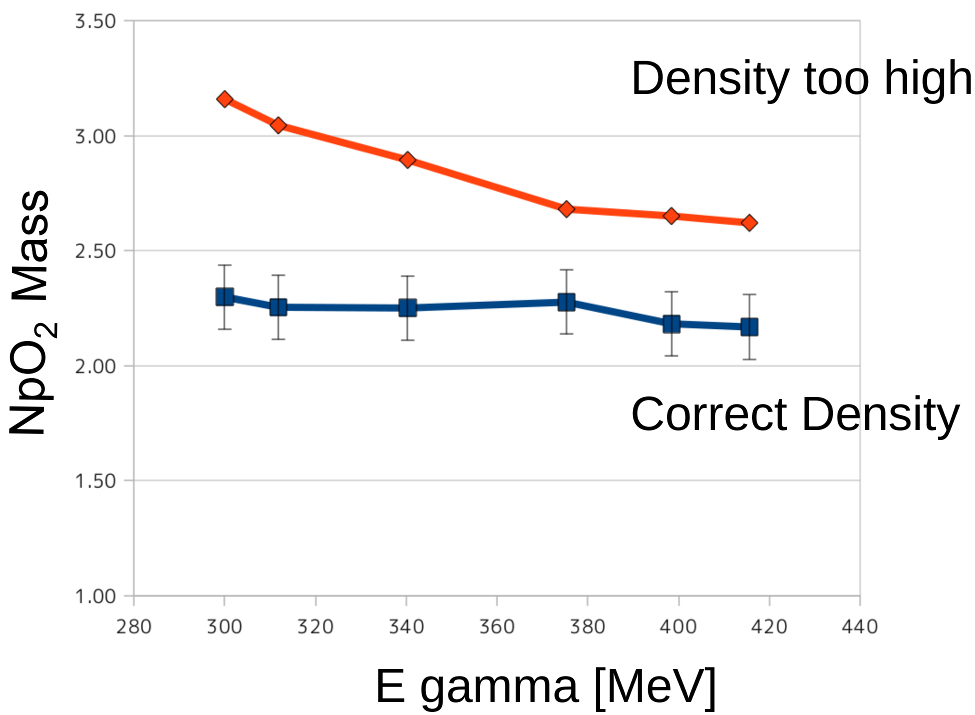


Attenuation factor changes differently for each line with mass fraction



Determining target mass

Small target mass from assay



Sample	Total Mass [g]	NpO_2 Mass [g]	Mass Frac [%]
1	10.37	8.97	.87±.06
2	2.24	2.23	.99±.05