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Title: Study of 149Sm capture and total cross sections for burnup credit applications*

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Study of ^{149}Sm capture and total cross sections for burnup credit applications*

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LA-UR-23-xxxxx

Introduction: Motivation

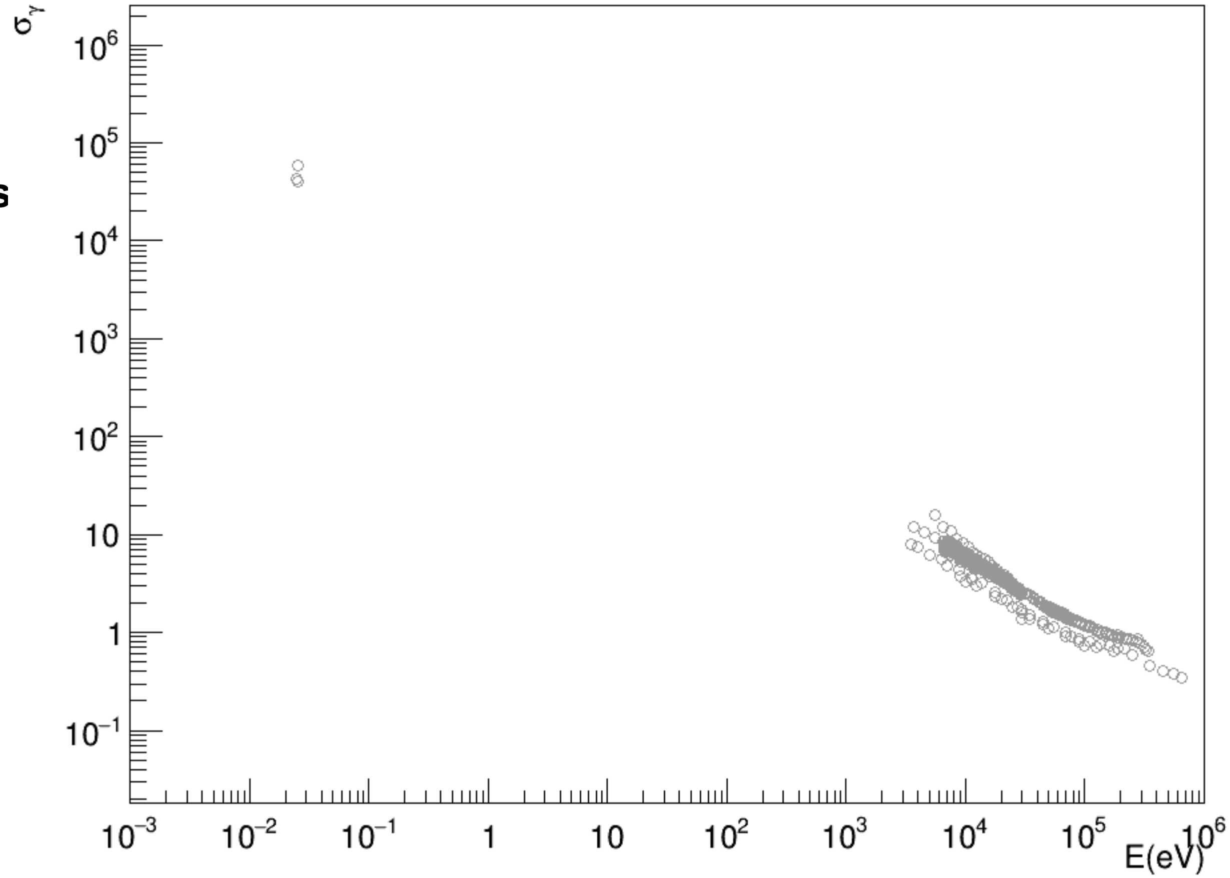
- **According to a report by L. Leal (ORNL/TM-2005/065)**

  ^{149}Sm capture rate underestimates measured capture rates for PWR experiments by 4.8%

- **And**

  evaluated data files do not provide cross-section covariance information for ^{149}Sm

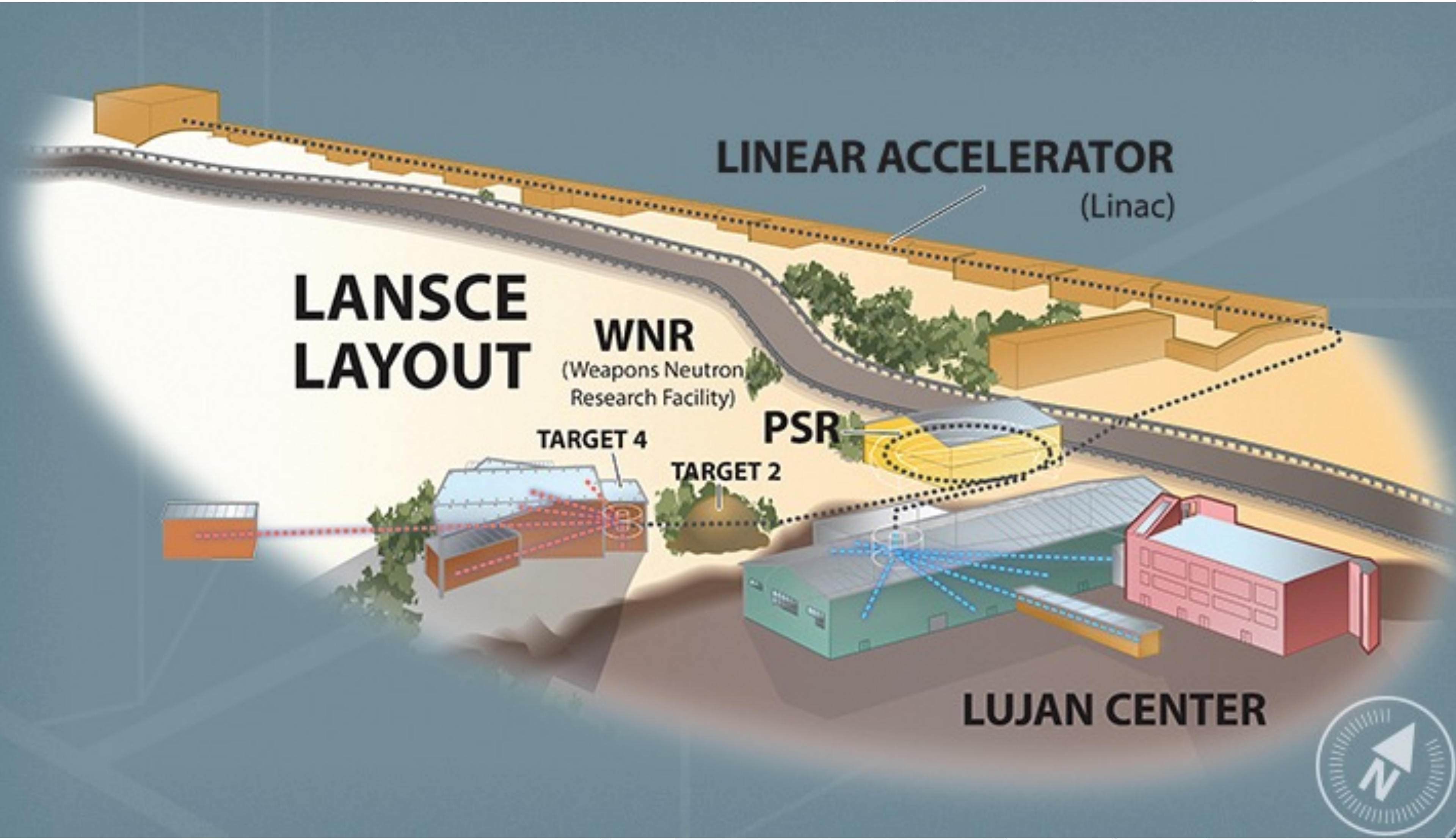
- **A few data sets in EXFOR**
- **Resonance parameters do exist but better uncertainties and covariances are needed**



Introduction: Summary

- We performed capture and transmission measurements with:
  - the DANCE (Detector for Advanced Neutron Capture Experiments) instrument
  - Capture data from 8 eV – 1 keV
  - and DICER (Device for Indirect Capture Experiments on Radionuclides) instruments
  - Transmission data from 1meV – 1 keV
- Additional measurements of ^{147}Sm
  - Contaminant in the samples
  - 3.4 eV strong resonance
  - Interesting abnormalities
- Data analysis is complete
- R-Matrix analysis almost (90%) complete

The Los Alamos Neutron Science Center - LANSCE



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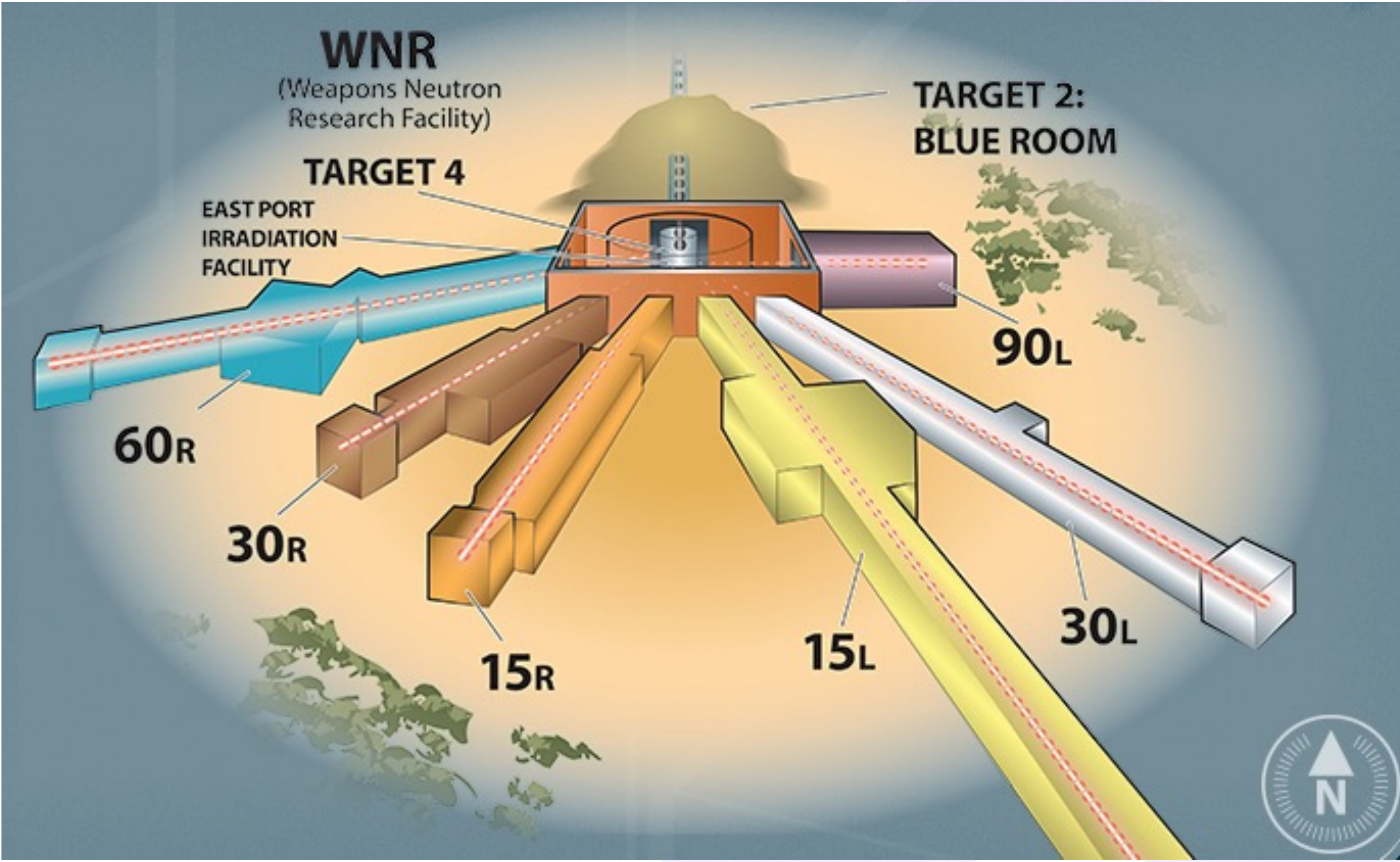

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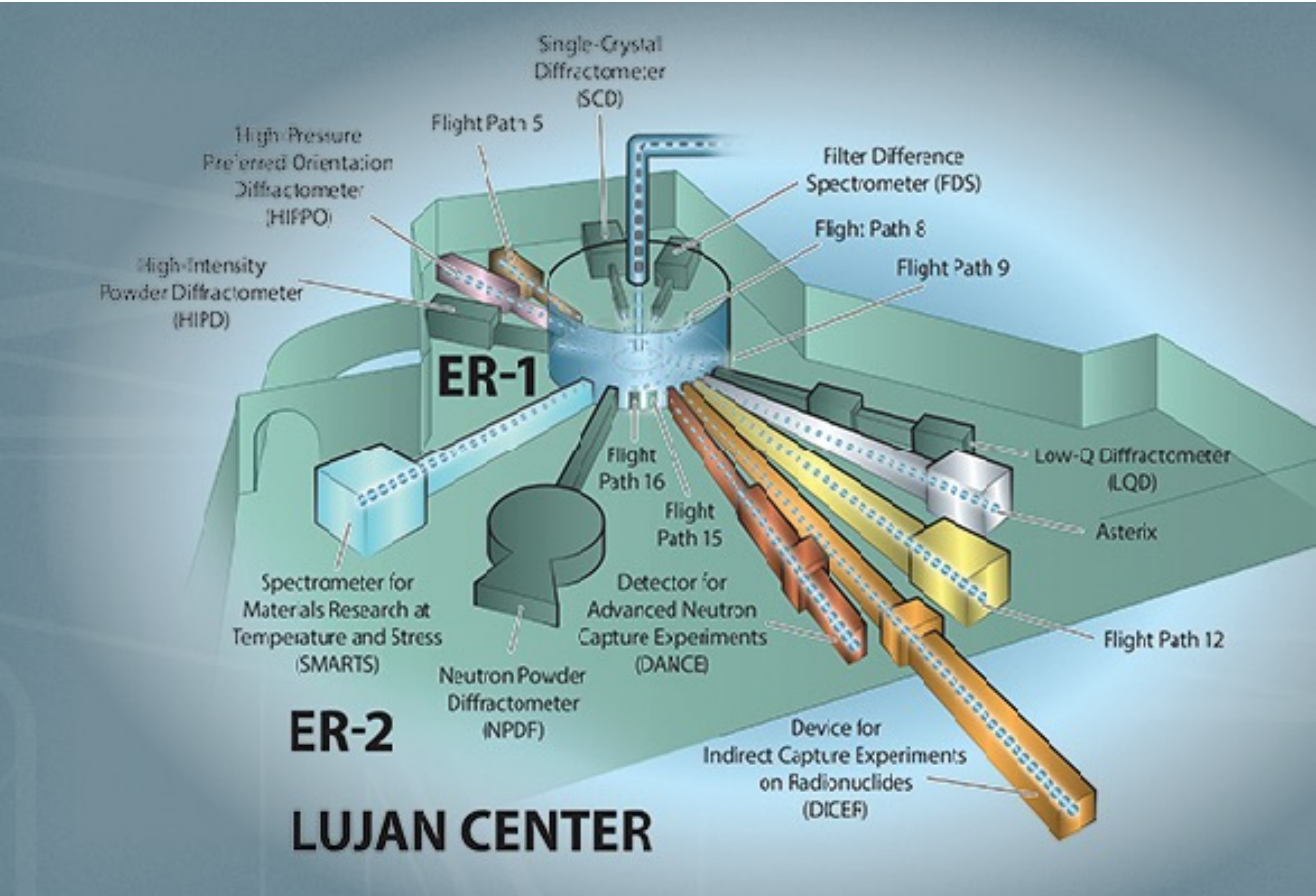
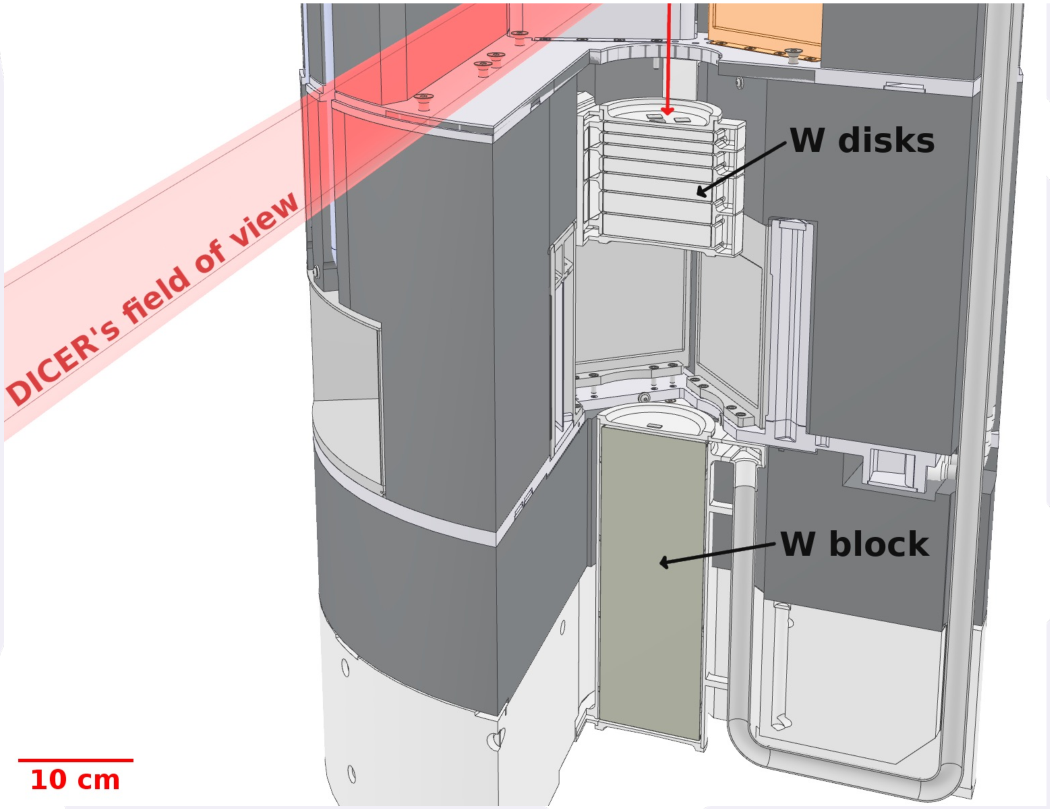


Description of the apparatus: Lujan@LANSCE@LANL

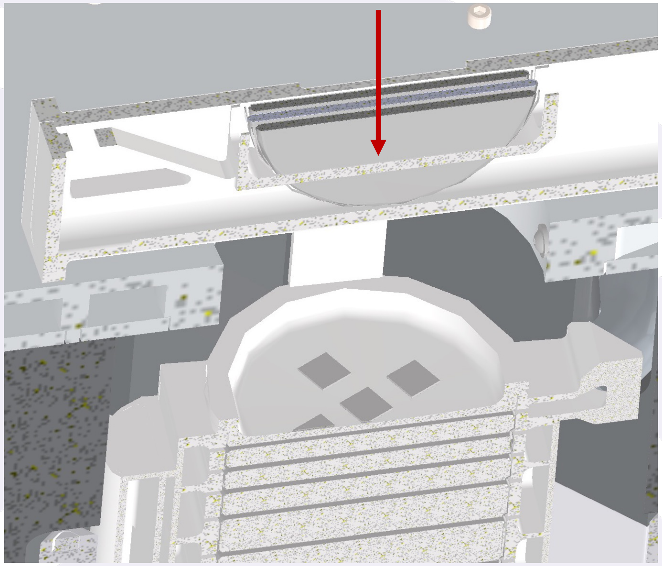
- The Los Alamos Neutron Science Center (LANSCE) is LANL's accelerator complex
- DICER belongs to the Lujan Center instruments which serve material and nuclear science studies.
- Neutrons are spallation products
- 800MeV protons impinge on a split W target at a 20 Hz duty cycle.
- DICER points to the liquid hydrogen moderator
- DICER flux spans from 0.2 meV – 100 keV
- Total: ~6x10^{7} n/s/cm^{2}

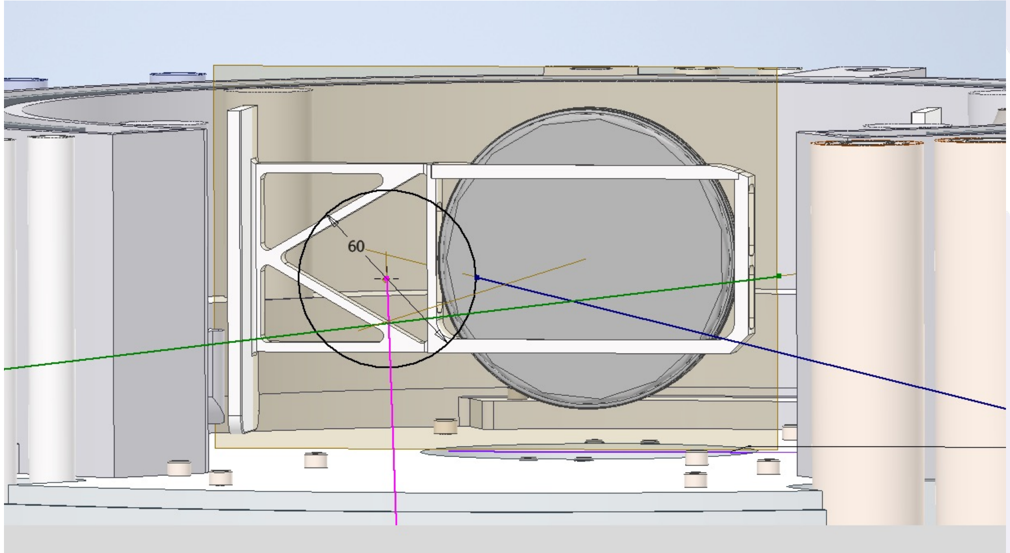
Mark-III

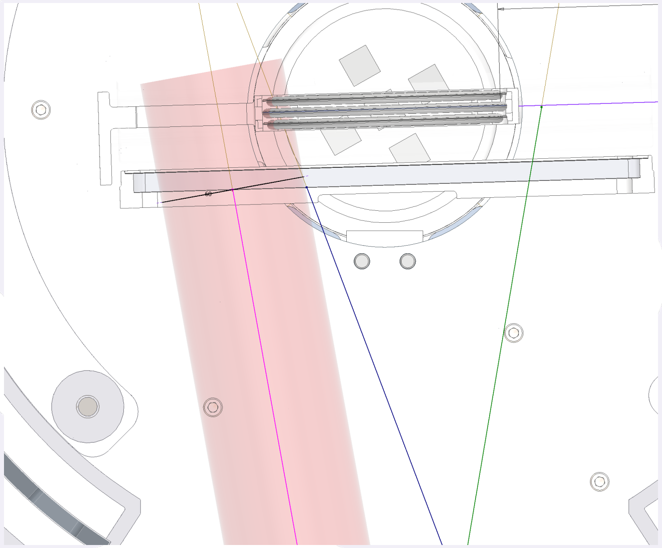


New neutron target at Lujan (Mark-IV)

- 800MeV protons impinge on a split W target at a 20 Hz duty cycle.
- DICER points to the liquid hydrogen moderator
- DICER flux spans from 0.2 meV – 100 keV
- Total: ~$6x10^7$ n/s/cm^2







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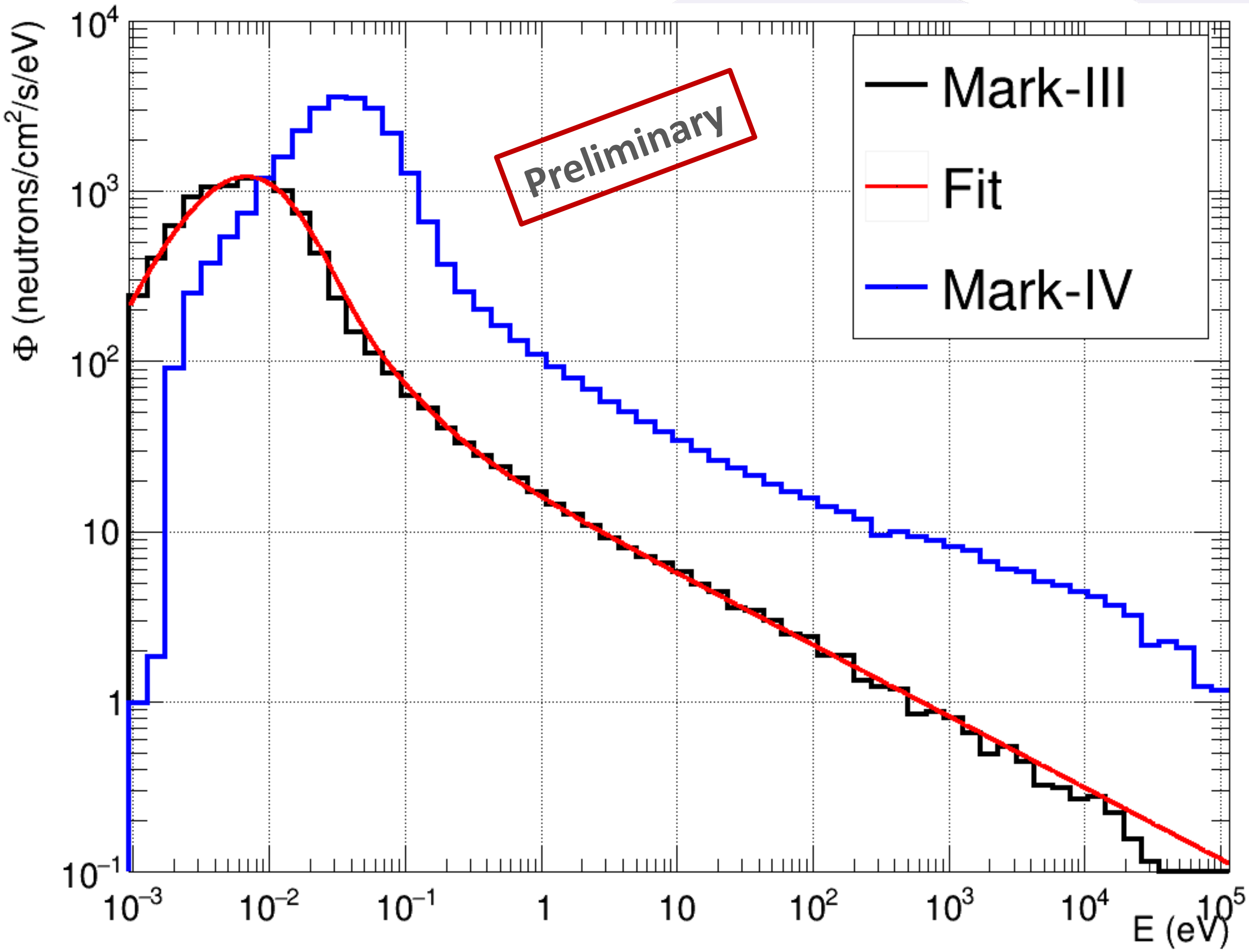
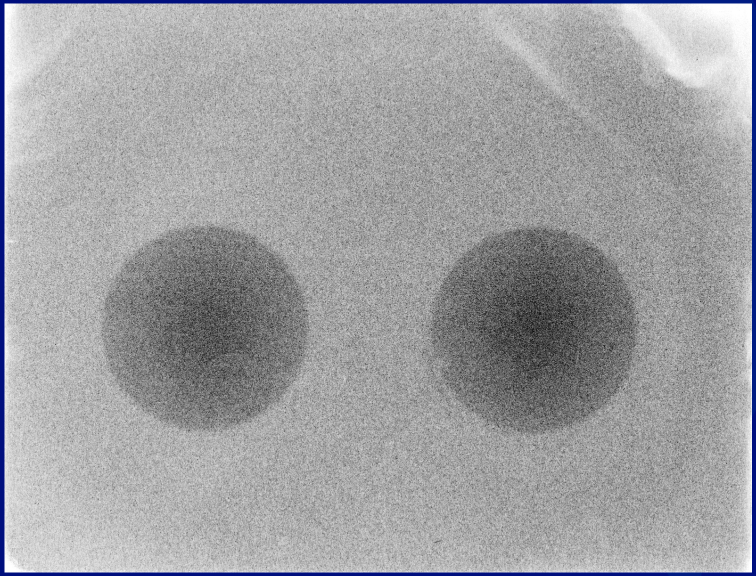
Description of the apparatus: Neutron spectrum

- Mark-IV provides an enhanced keV spectrum compared to Mark-III
- Mark-IV is expected to improve the neutron energy resolution

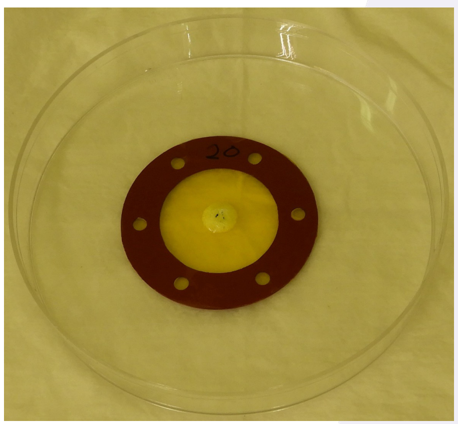


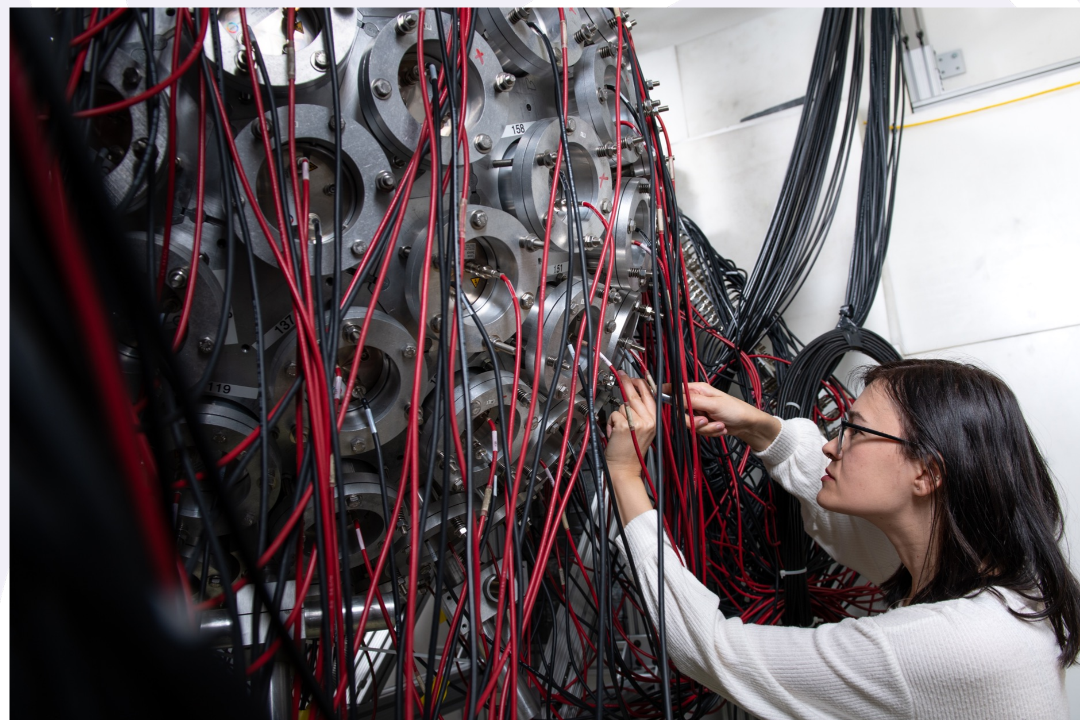
Neutron capture measurements @ DANCE



Detector for Advanced Neutron Capture Experiments: DANCE

- **DANCE is a 4π spherical detector array (Heil et al. NIMA (2001))**
- **Gamma-ray calorimeter (E^{γ}_{sum}, Multiplicity)**
- **Spinometer**
- **160 BaF_2 detectors**
- **Designed to perform capture measurements on small radioactive samples 4 mm in diameter, mg – 10s mg**





Contacts

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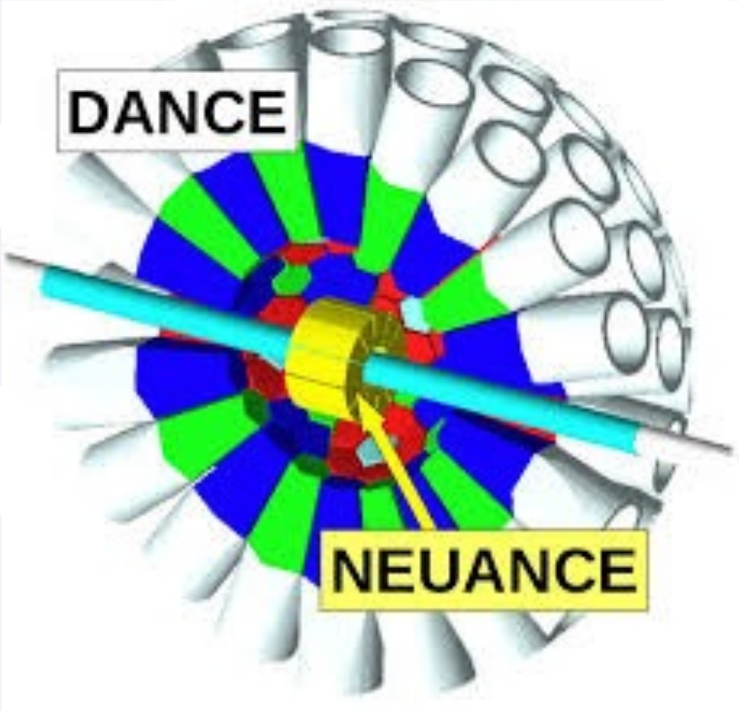
Detector for Advanced Neutron Capture Experiments: DANCE

- **DANCE has an inner cavity of 17 cm**
- **NEUANCE (Neutron detector array at dANCE) (M. Jandel et al. NIM 00 (2017) 1-27).**
- **Works in (anti)coincidence with DANCE to tag fission events and perform (n,γ) studies on fissile nuclei (i.e. 233U E. Leal-Cidoncha et al., Submitted to Phys. Rev. C)**



Contacts

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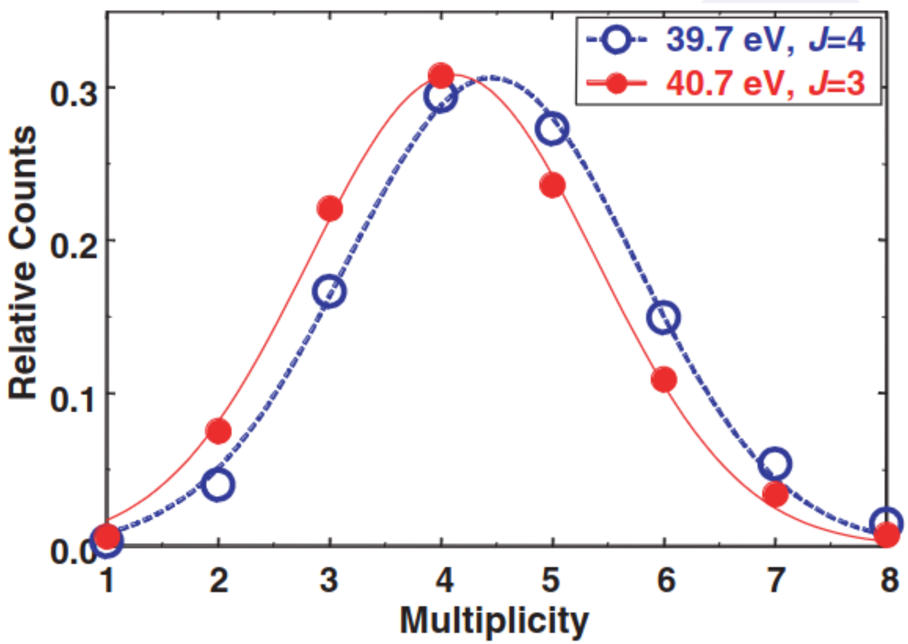
Samples

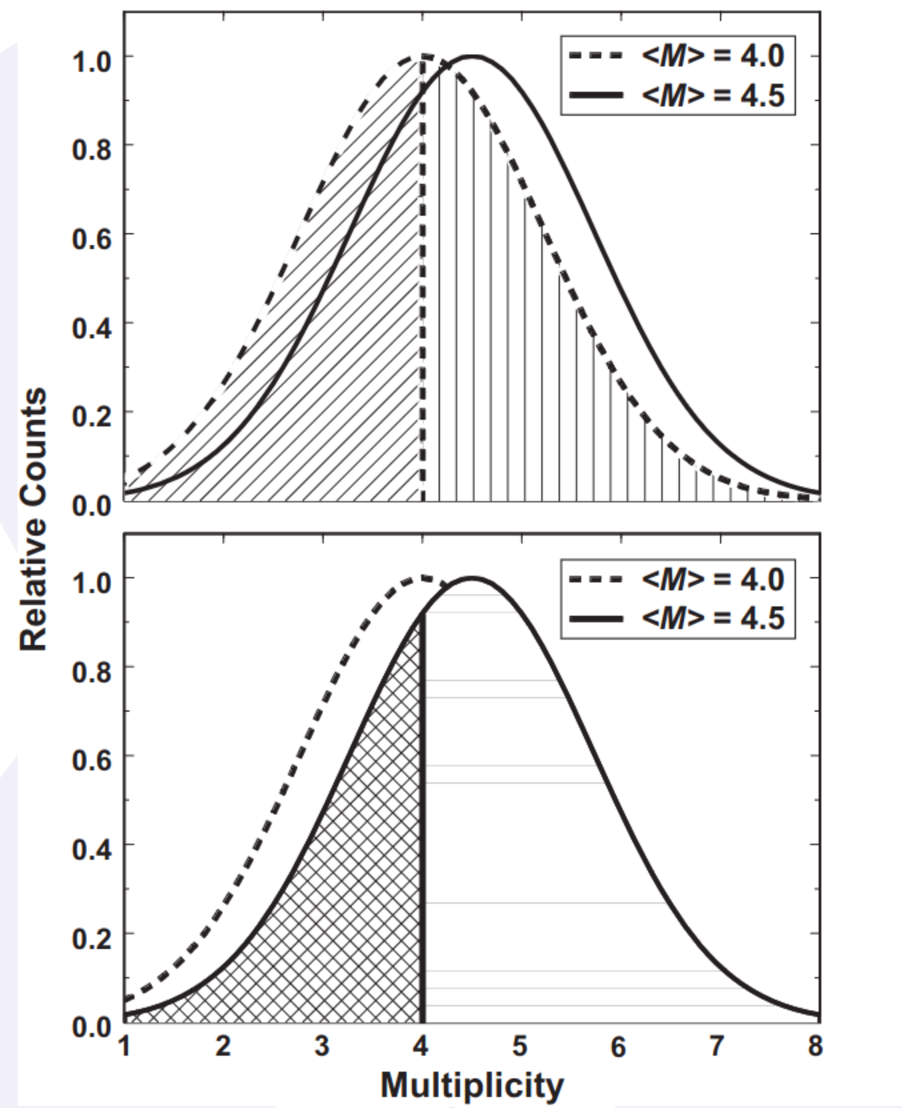
- 10 mg $^{149,147}Sm$ sample
- 4 mm diameter
- Procured from ORNL

Analysis - Multiplicity

- **DANCE provides multiplicities**
- **Different multiplicity distributions for different Js**
- **DANCE is used as a spinometer**

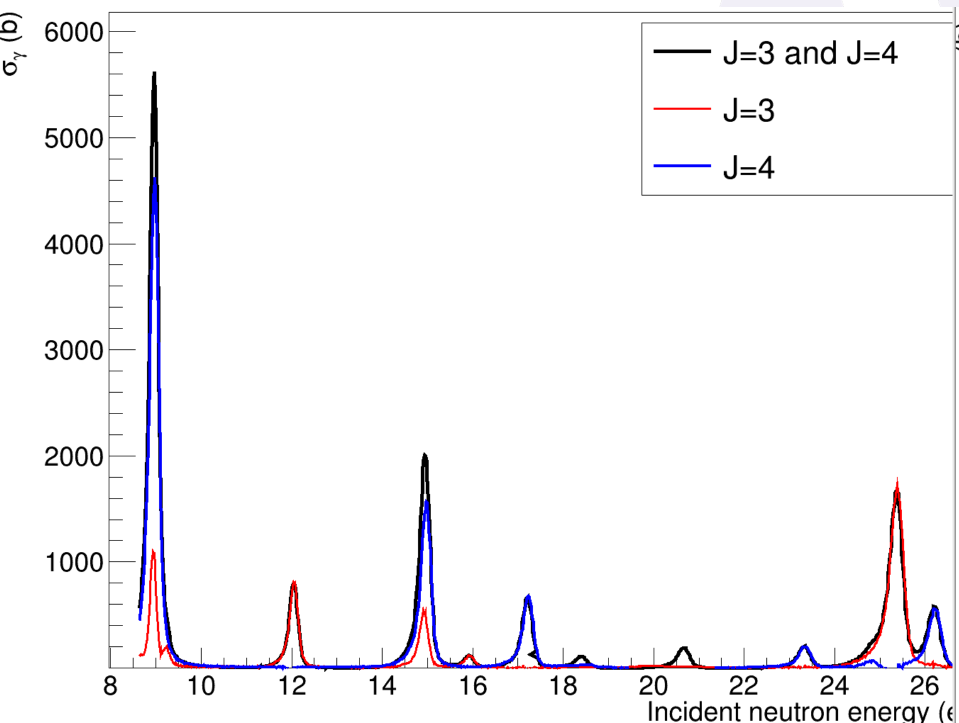
PHYSICAL REVIEW C **76**, 025804 (2007)

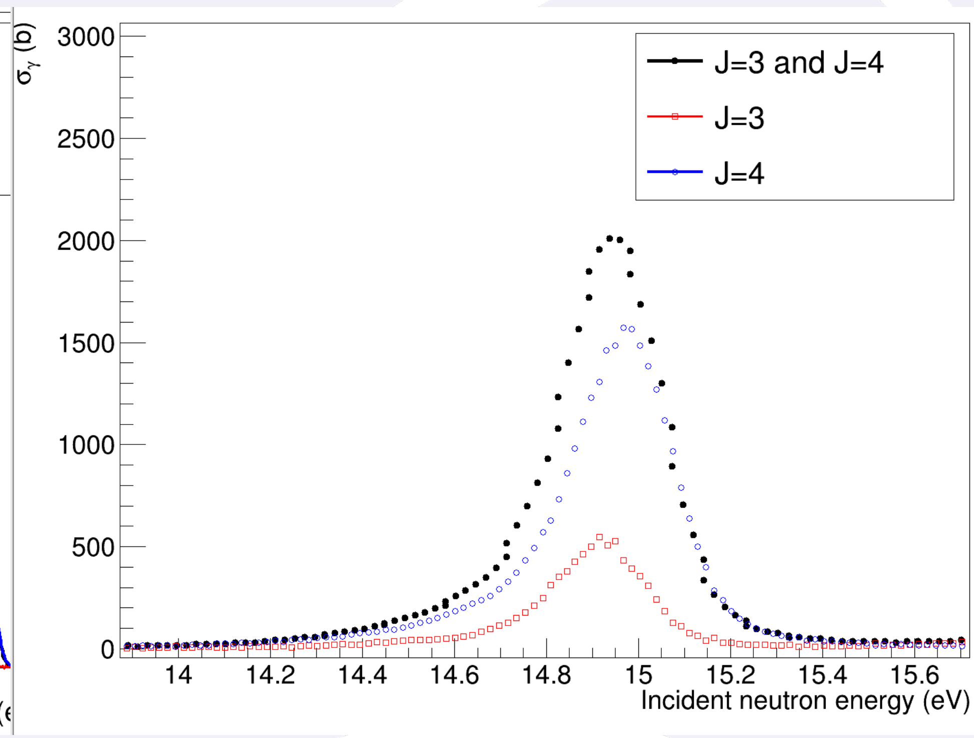




Analysis – J-dependent cross sections

- **Doublets can be resolved**





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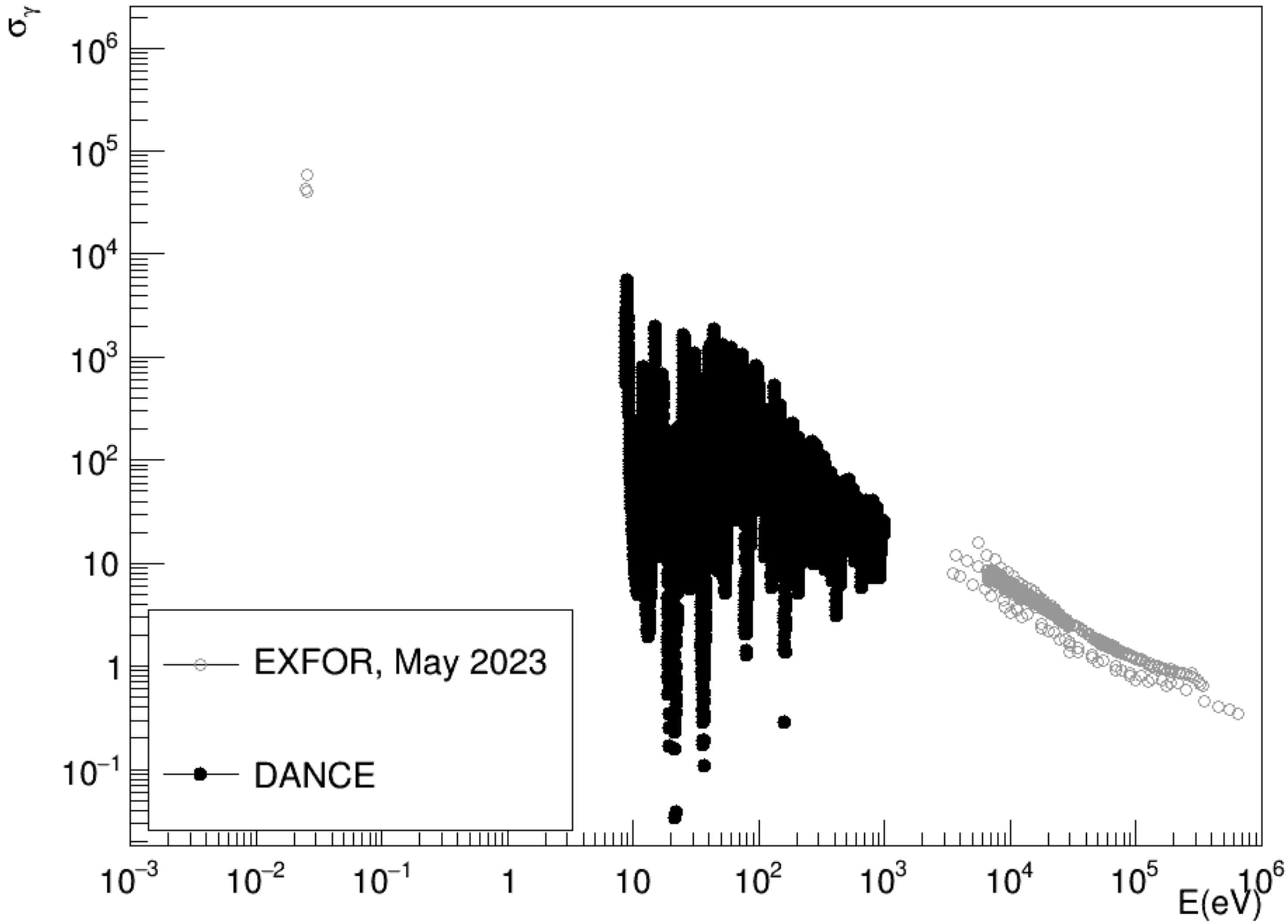
Analysis

- **Cross section from 8 eV – 1 keV**
- **R-Matrix analysis almost complete**



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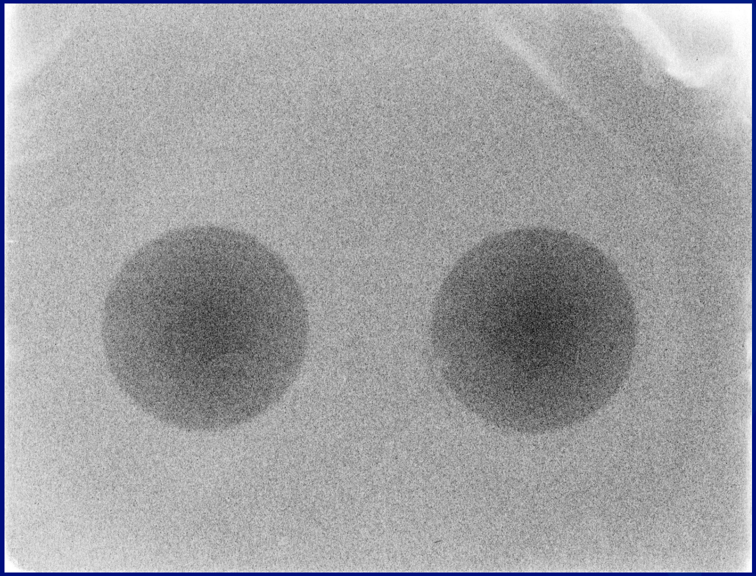
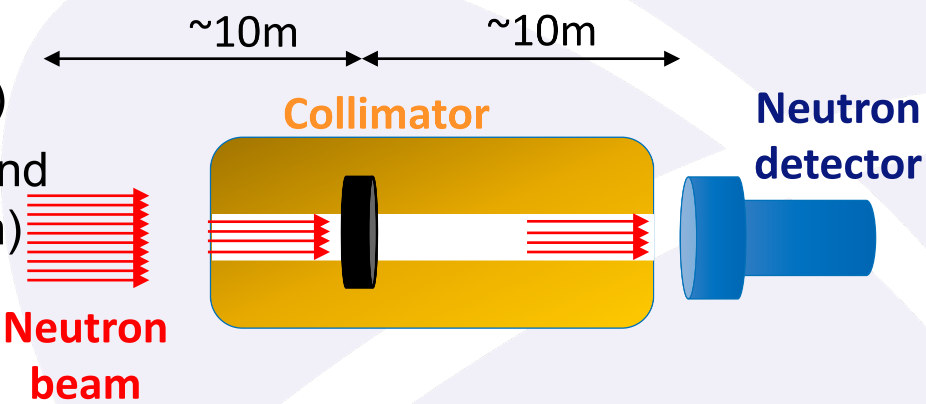
Neutron transmission measurements @ DICER



Traditional transmission measurements: How to

- The neutron spectrum (C_{out}) and backgrounds (B_{out}) are recorded by a neutron detector (**sample out**)
- A sample, usually big, is installed and absorption dips appear (**sample in**)

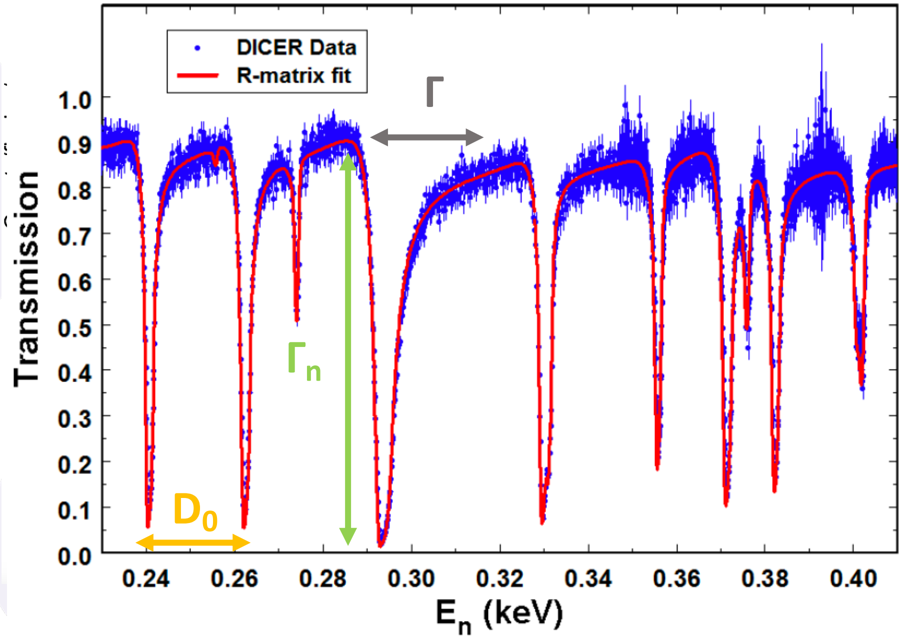


- The transmission is the ratio sample in/out

$$T = \frac{C_{in} - B_{in}}{C_{out} - B_{out}}$$

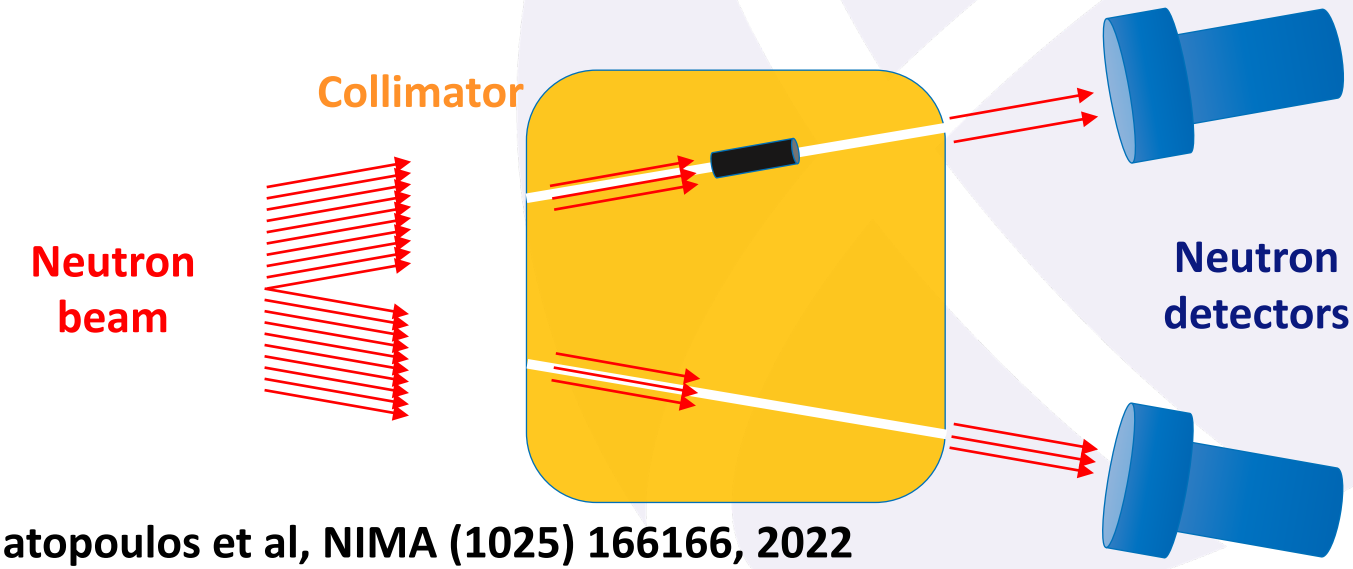
- Transmission measurements provide information on the total (Γ) and neutron (Γ_n) widths and the level spacing (D_0)

Γ : **Resonance width**
Γ_n : **Resonance depth**
D_0: **Level spacing**



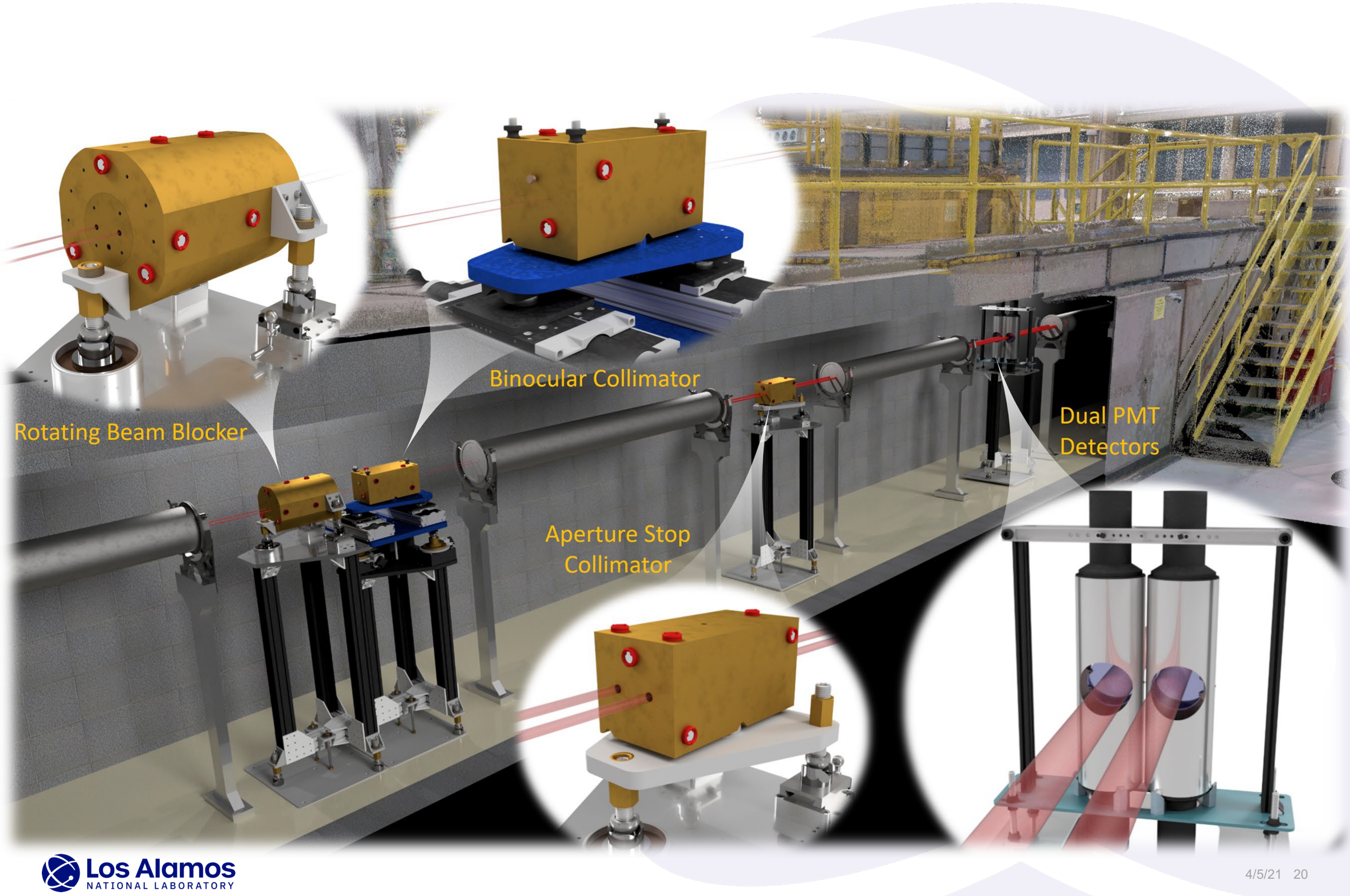
Non-traditional measurements: Binocular approach

- Binocular mode of operation: Simultaneous measurement of sample in/out
- Binocular collimator is a unique concept conceived, designed and executed at DICER.
- No precise repositioning concerns, as long as the sample is precisely positioned beforehand: metrology network ~10μm and ~10 mdeg accuracy
- Added bonus: measurements will be completed 50% faster!
- High flux facility → small samples (~10,000 smaller than typical)
- ~50 radionuclides are within reach ($t_{1/2}$ > 30days, D_0<50eV)



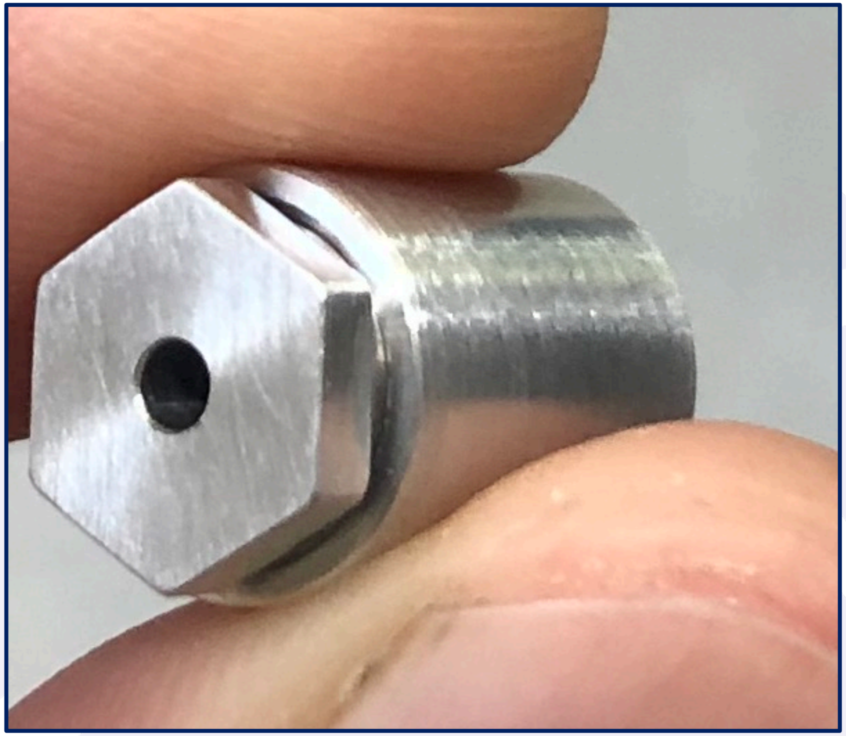
A. Stamatopoulos et al, NIMA (1025) 166166, 2022

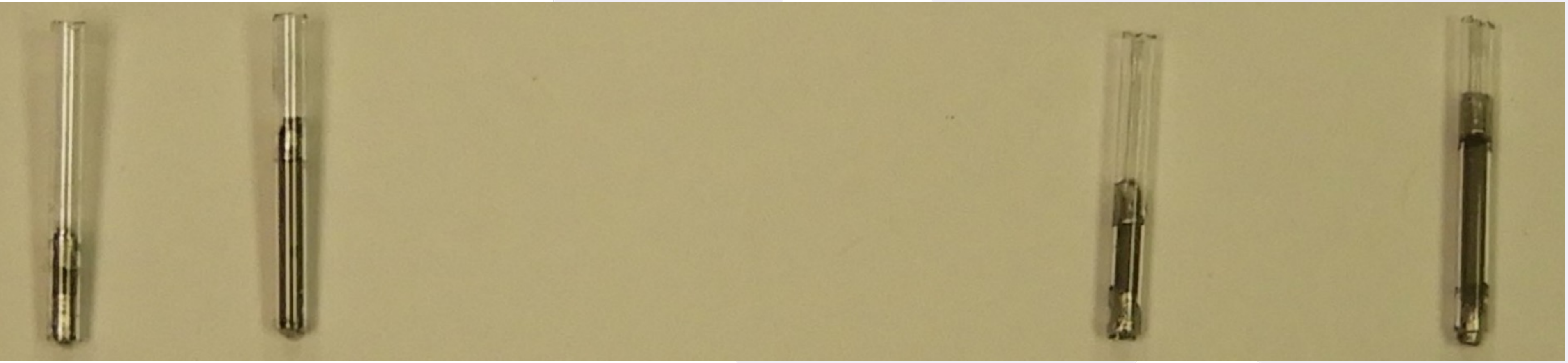
Description of the apparatus:
Device for Indirect Capture Experiments on Radionuclides



Samples

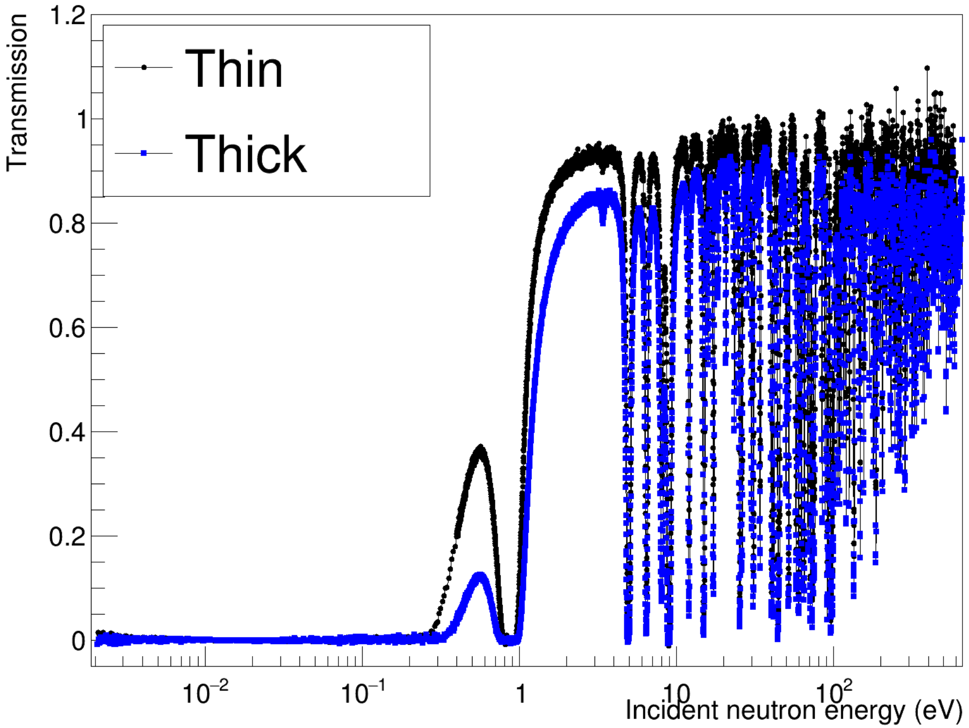
- ~5mg $^{149,147}Sm$ thin sample
- ~12mg $^{149,147}Sm$ thick sample
- 1 mm diameter
- Powder stuffed in capillary tubes
- Capillary tubes inserted in Al canisters

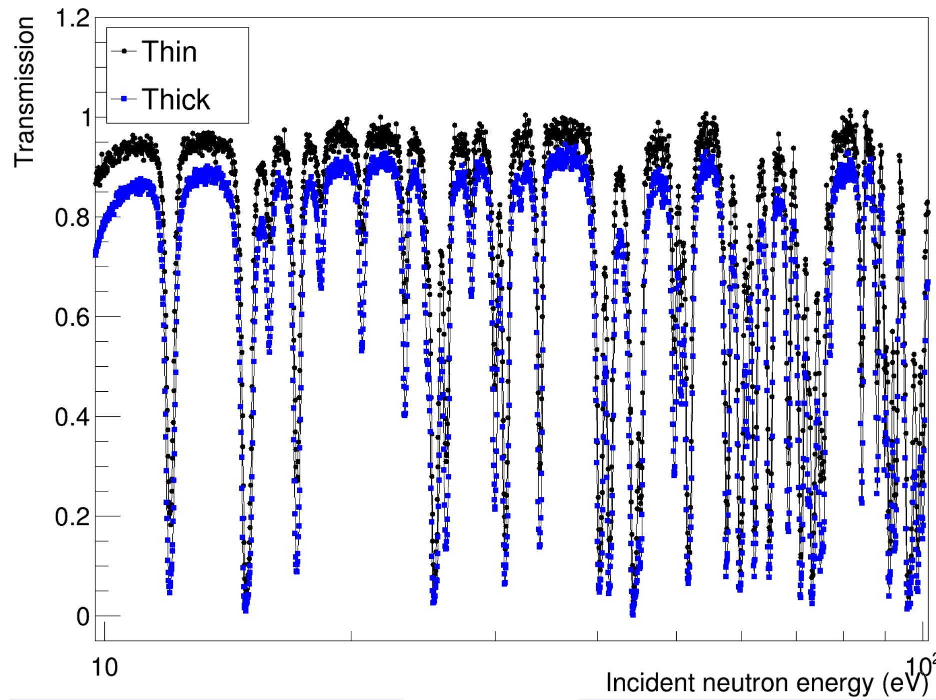




Analysis

- **163 resonances were resolved**
- **R-Matrix is almost complete**





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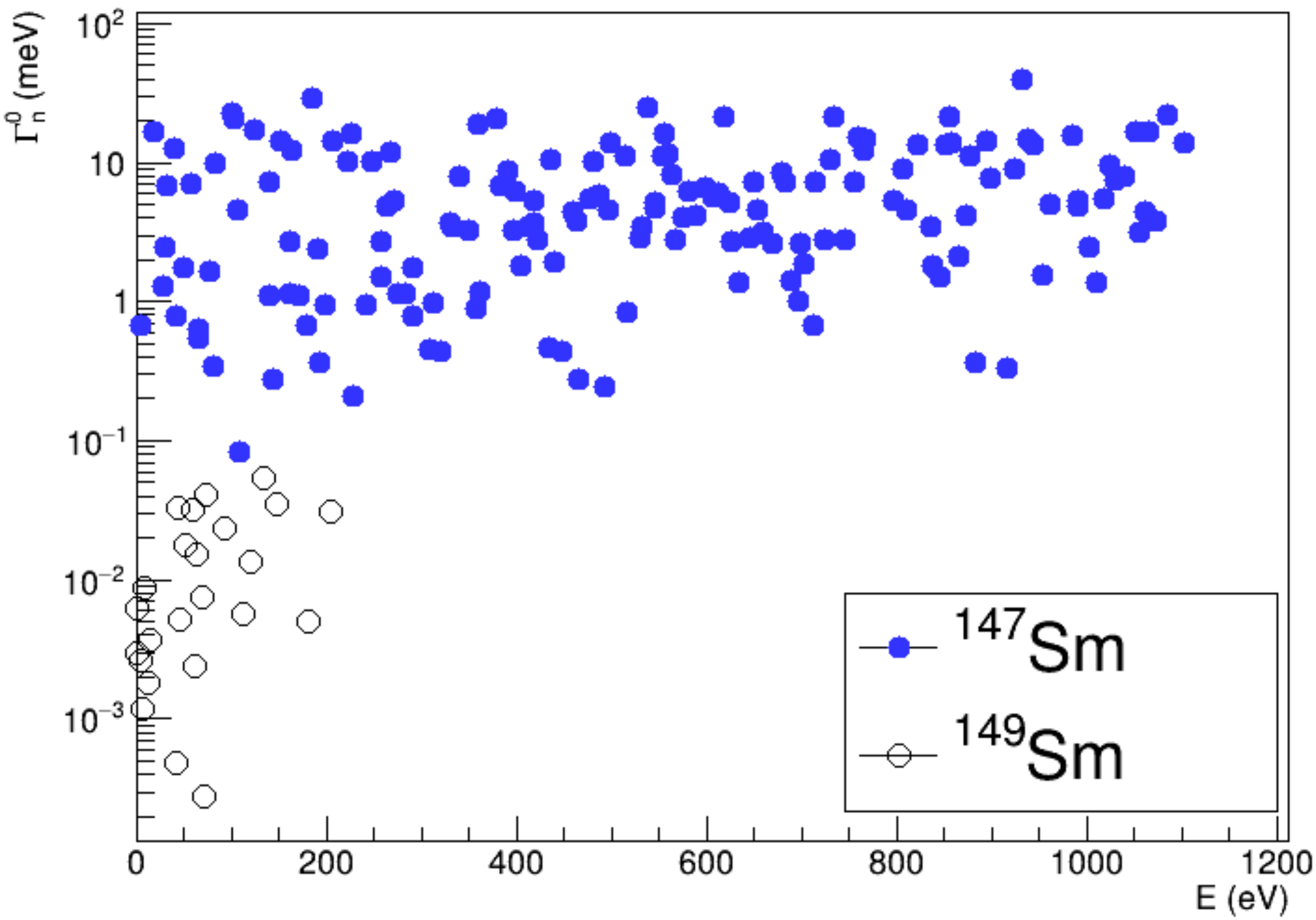
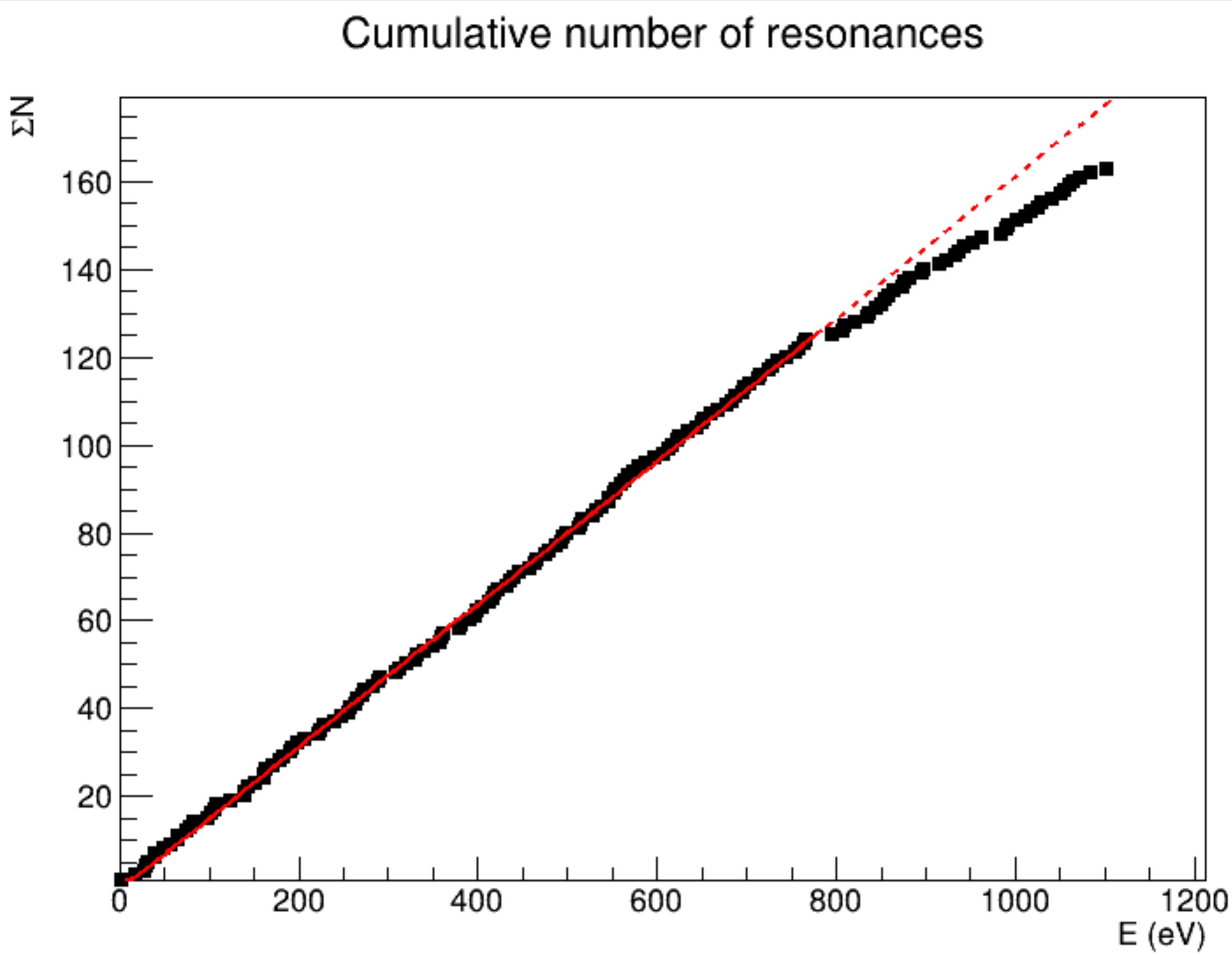
Analysis

- **Able to detect small neutron widths**
- **From the ^{147}Sm data: ^{147}Sm + ^{149}Sm (scaled to abundance) reduced widths**



Analysis

- 163 resonances were resolved
- R-Matrix is almost complete
- Simultaneous fits on capture (J=3, 4) and transmission (thick and thin samples)



Analysis

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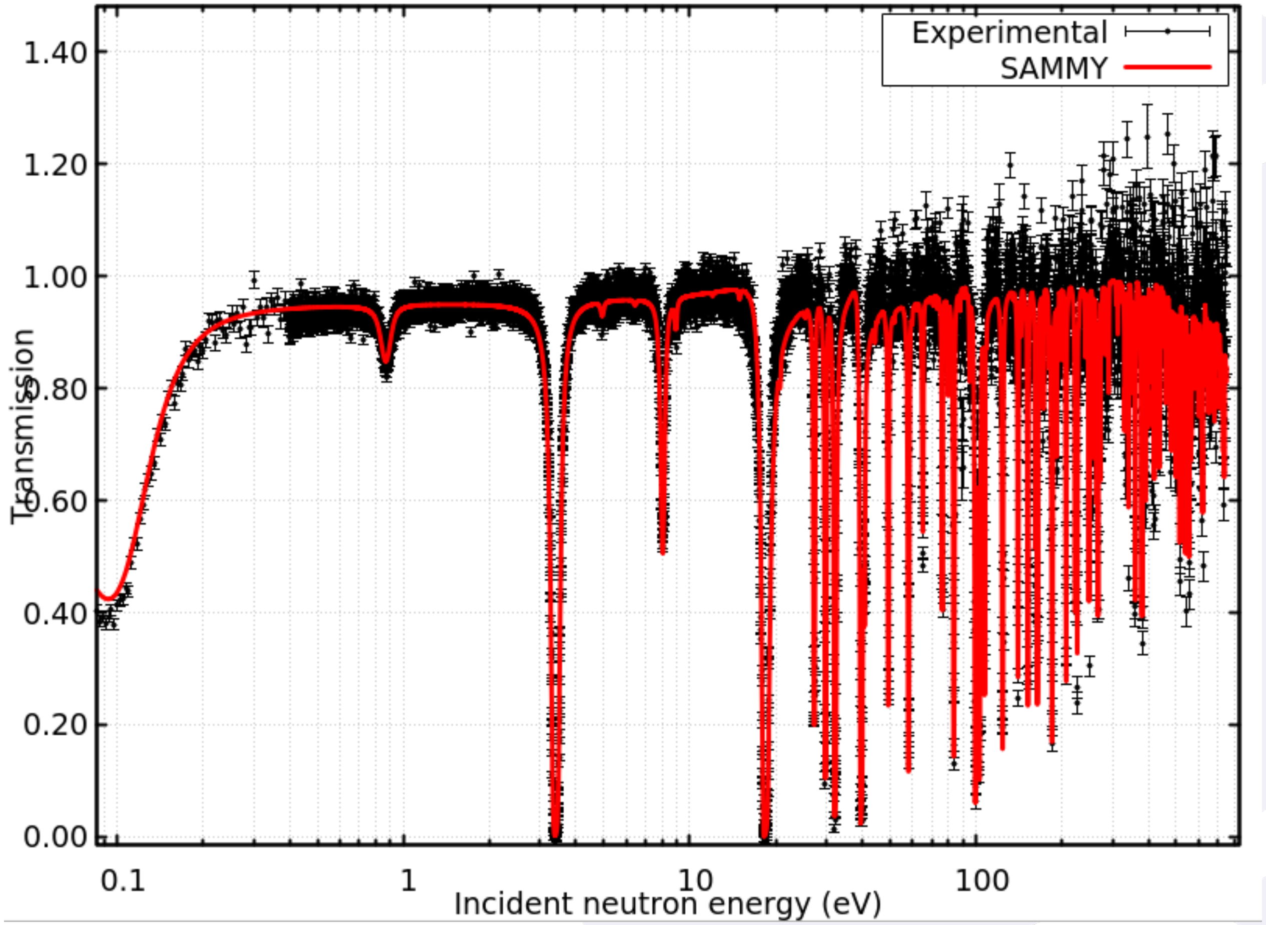

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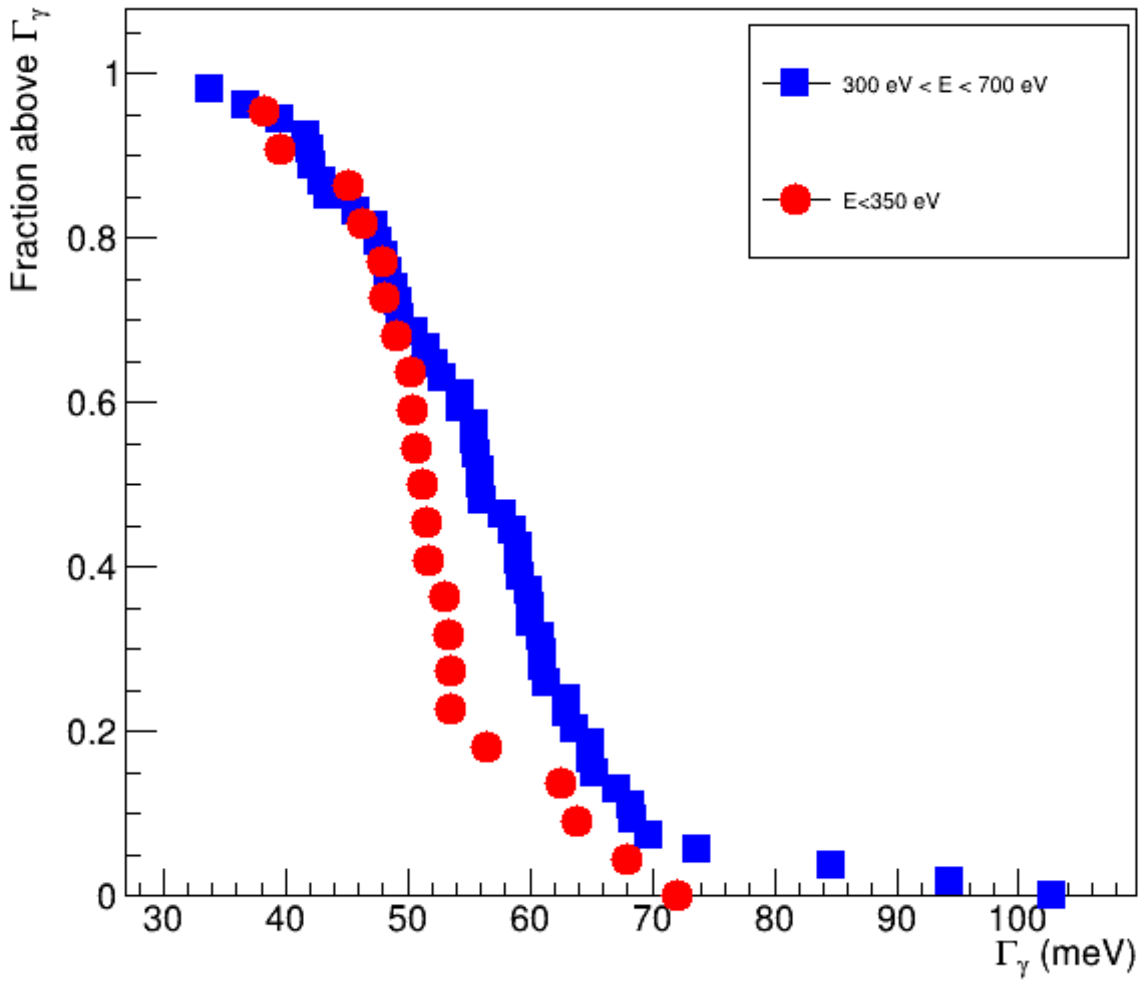
Interesting findings on ^{147}Sm

- **Capture widths change abruptly around 300 eV**




Thank you for
your attention!

Back-up slides

Introduction: The technique

- Resonances are used to calculate **level spacing** (D_0), **strength function**(S_0) and the **average radiation width** ($<\Gamma_\gamma>$)

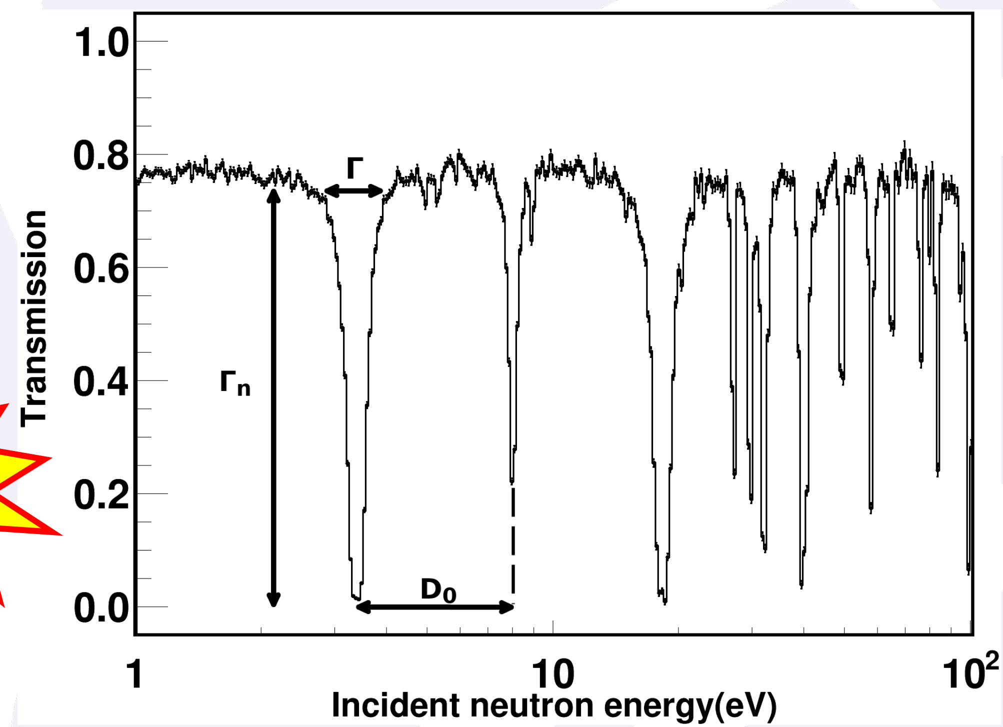
$$\Gamma = \Gamma_n + \Gamma_\gamma \qquad S_0 = \frac{< g\Gamma_n^0 >}{D_0}$$

Total   Scattering   Capture

$$\langle \sigma_\gamma \rangle = \frac{2\pi^2}{k^2} \frac{S_0 \langle \Gamma_\gamma \rangle / D_0}{S_0 + \langle \Gamma_\gamma \rangle / D_0} W$$

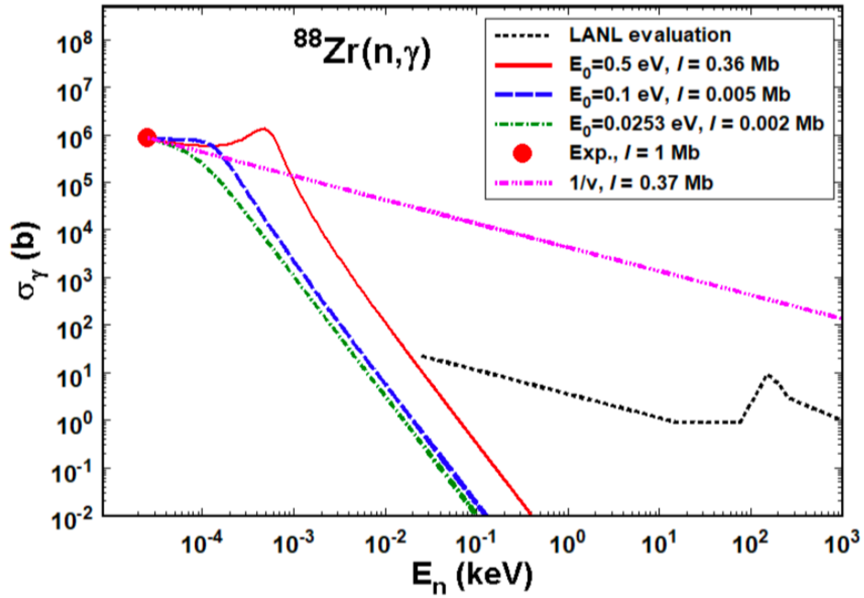
$= f(\Gamma_n, \Gamma, D_0)$

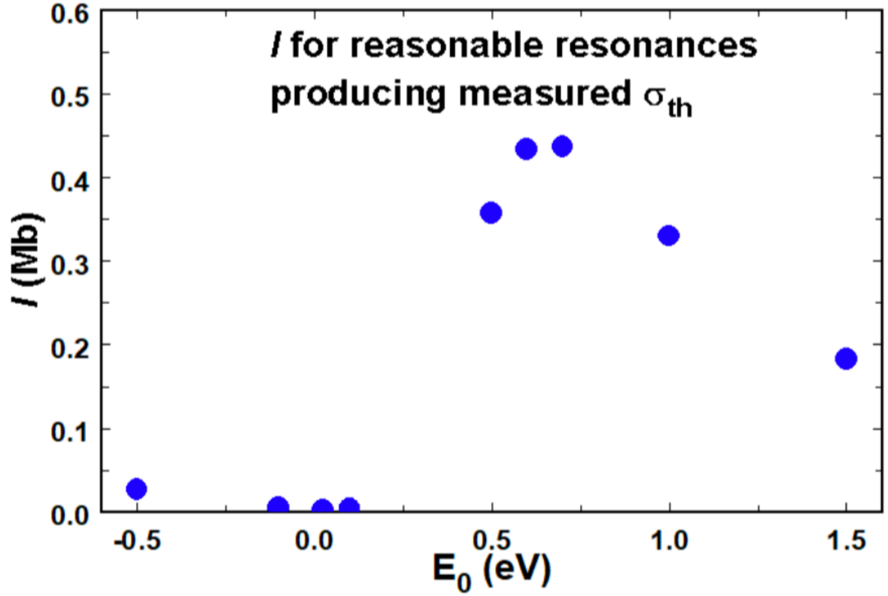
- k : Wave number
- S_0 : Strength function
- D_0 : Level spacing
- $<\Gamma_\gamma>$: Average radiation width
- W : Width fluctuation factor



^{88}Zr

- Original motivation; quantify the resonance responsible for the huge, $\sigma_{\gamma,th}$=861±69 kb, ^{88}Zr(n_{th},γ) cross section (Shusterman *et al*. Nature 2019)
- Latest report (Nick Scielzo 12/4/20); Resonance integral, I =(2.53±0.28) Mb.
  Have not found any resonance parameters which yield reported $\sigma_{\gamma,th}$ and *I*
- Appears even more likely that the LANL ^{88}Zr(n,γ) cross section is much too small
  Even extreme assumption of 1/v cross section yields only ~1/7 of reported *I*

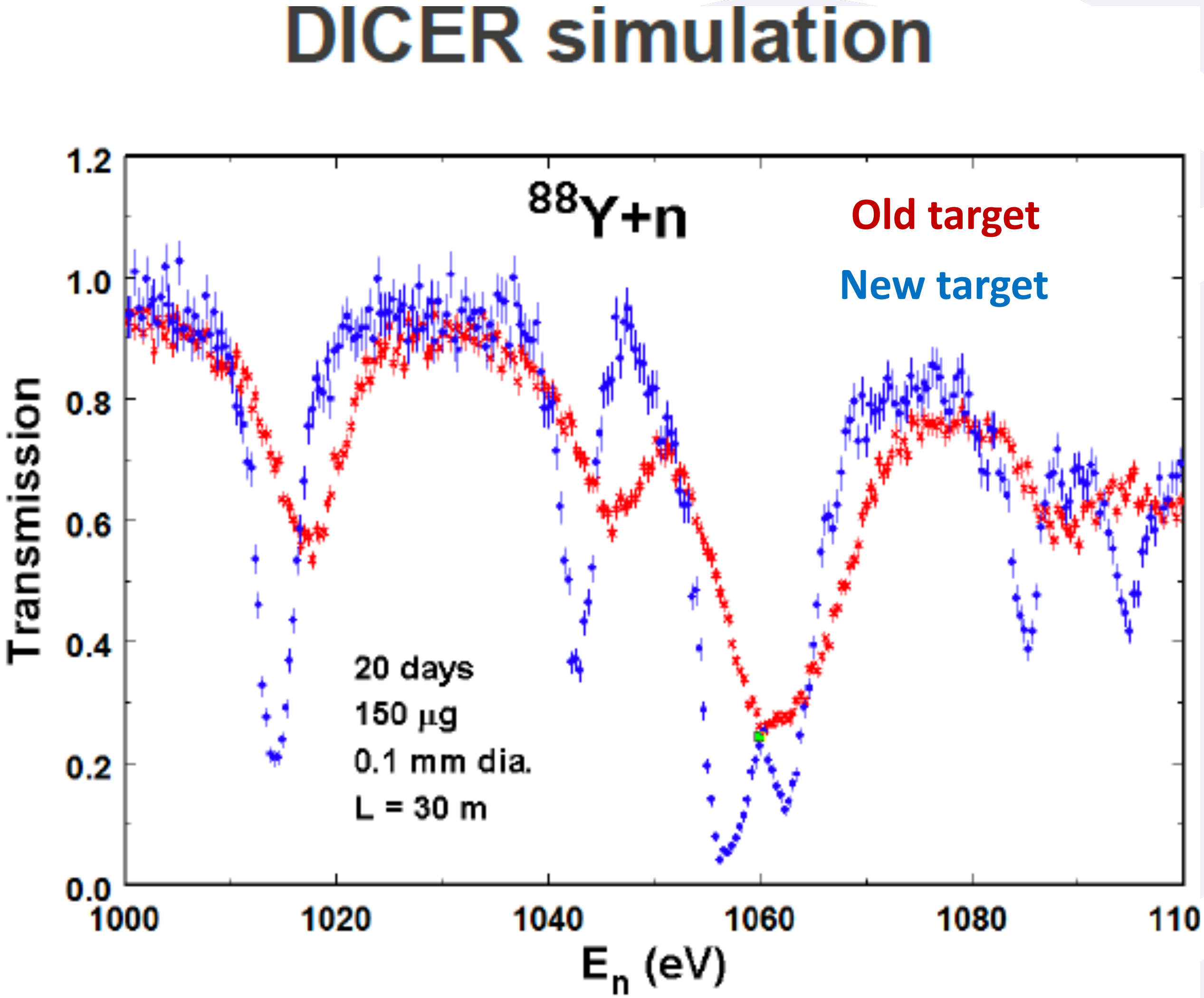




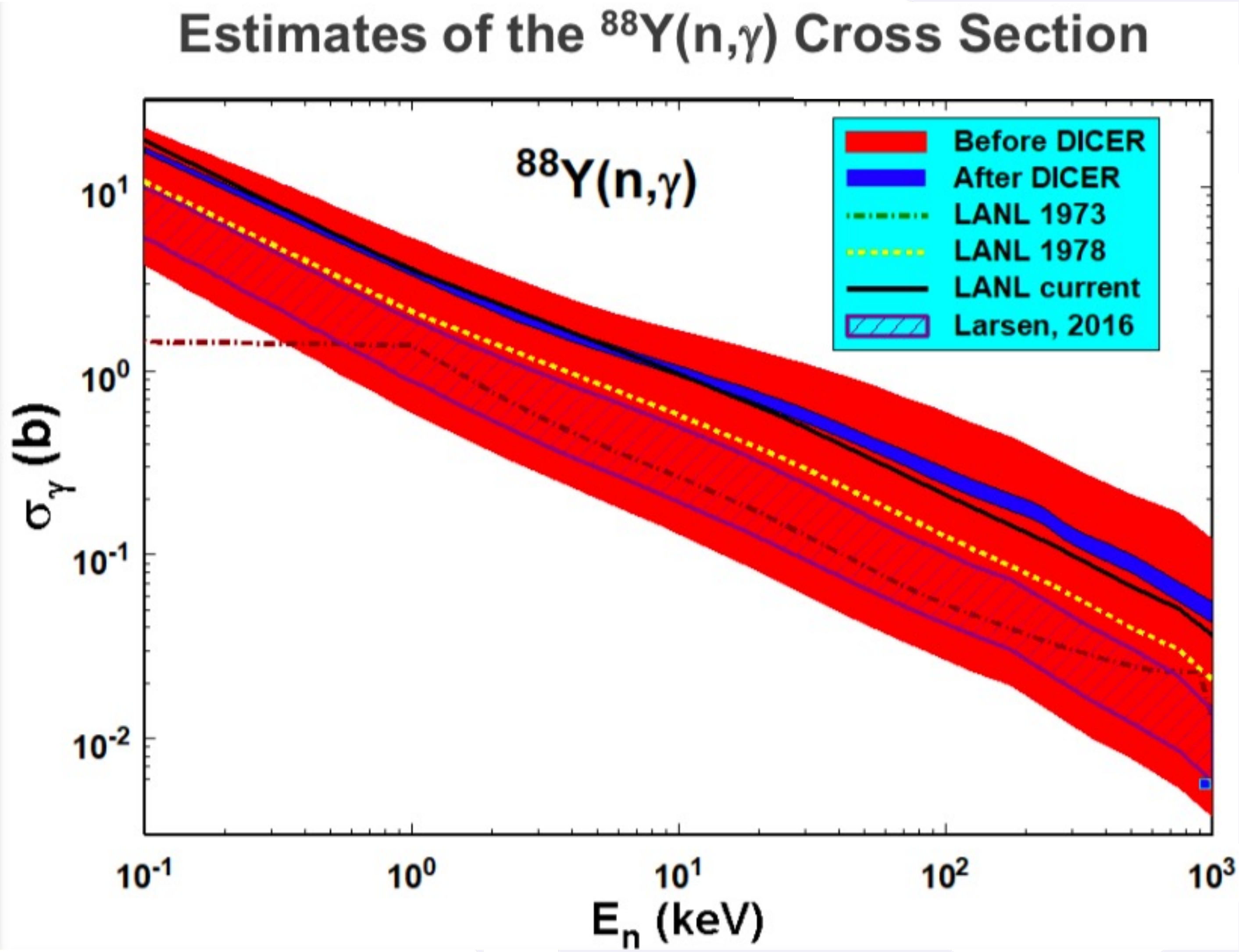
Signal to background: B-Poly wall

Installation of Mark-IV spallation target@LANSCE

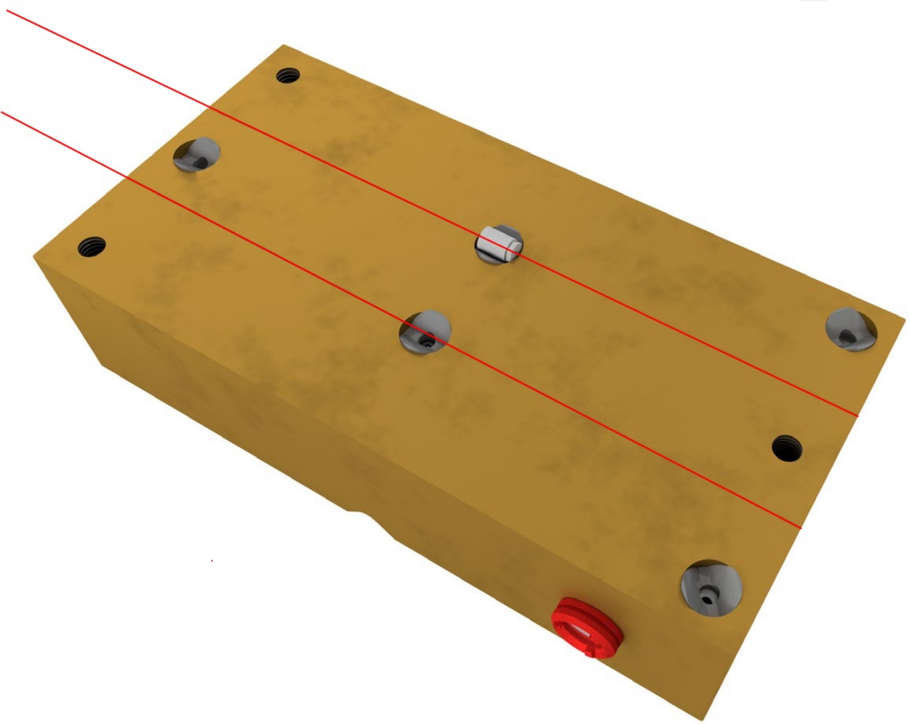
- Underway the upgrade of Lujan spallation target
- Improved flux and resolution

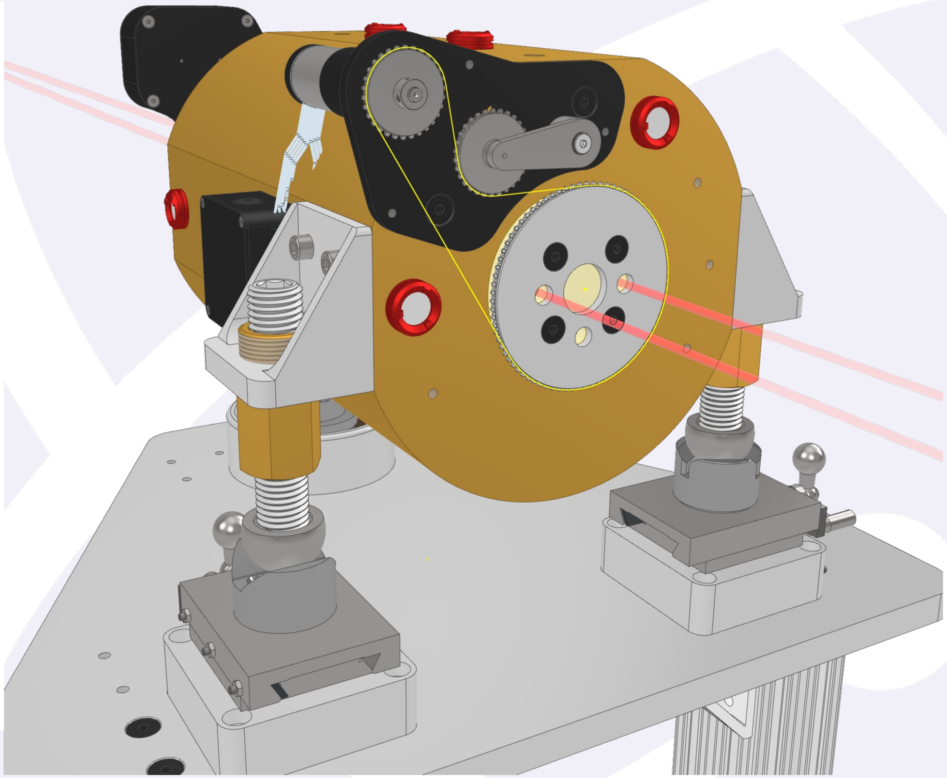


^{88}Y



Estimates of the ^{88}Y(n,γ) Cross Section
^{88}Y(n,γ)
Before DICER
After DICER
LANL 1973
LANL 1978
LANL current
Larsen, 2016
10^{1}
10^{0}
10^{-1}
10^{-2}
σ_γ (b)
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
E_n (keV)





Two Main Technical Challenges

1. Producing enough ^{88}Zr and ^{88}Y and fabricating suitable samples

Nuclide	Mass (µg)	Diameter (mm)	Activity (Ci)	Area Ratio (Bk/sample)	Activity Ratio (sample/larger Bk)
^{88}Zr	1.1	1.0	0.020	2.56	0.022
^{88}Y	138	0.1	1.9	256	2.0
^{249}Bk	500	1.6	0.082	1	0.088
^{249}Bk	5650	1.6	0.93	1	1

LANSCE flux is >250 higher than facility (ORELA) where ^{249}Bk experiment was made
^{88}Y sample activity is only 2 times higher than ^{249}Bk sample

2. Obtaining acceptable transmission data with these samples

Sample must be thick enough for resonances to cause transmission observable dips

Diameter determined by thickness and amount of material available

The smaller the collimator, the harder the measurement


SKY HOOK
SKY HOOK


SKY HOOK
4
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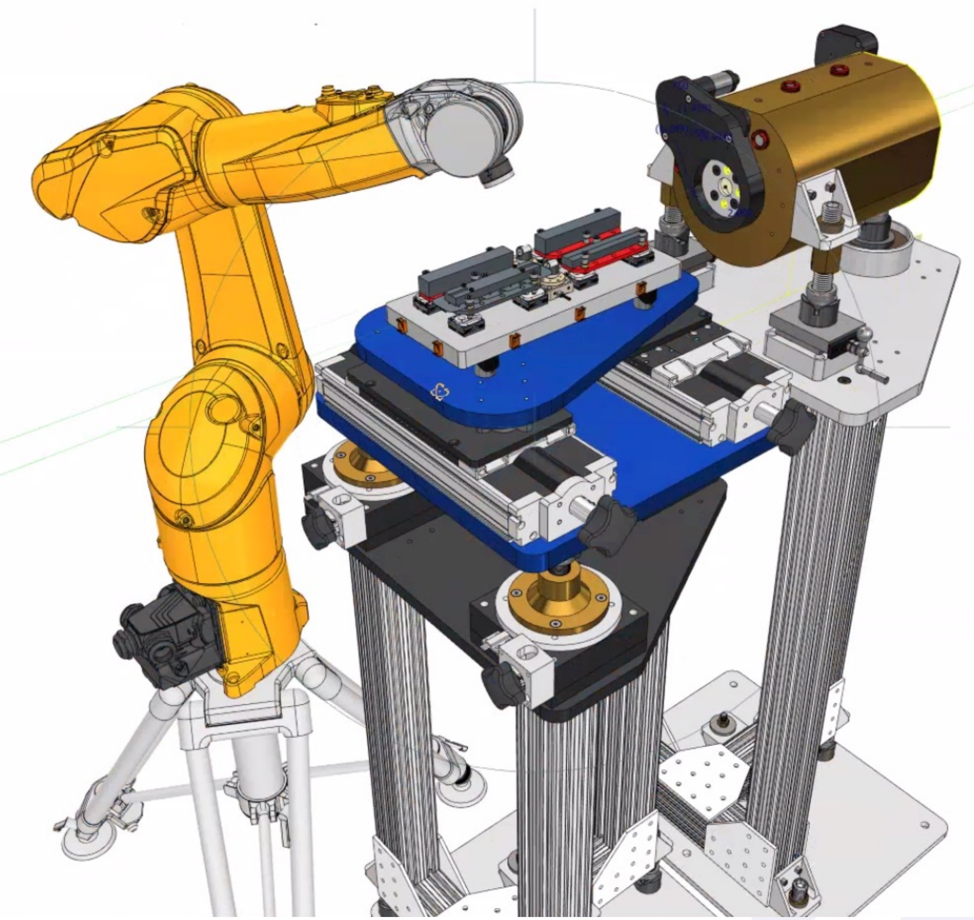

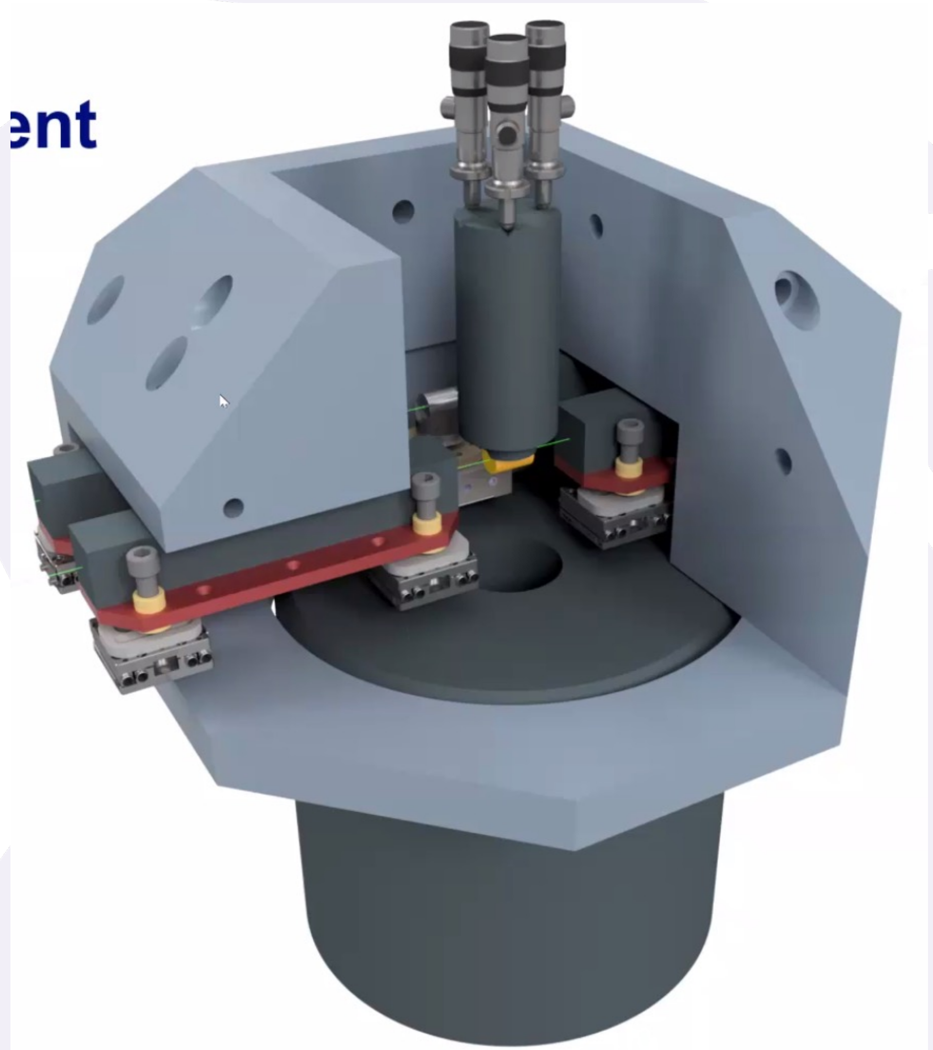
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PHYSICAL REVIEW C **82**, 064610 (2010)

γ-ray strength function method and its application to ^{107}Pd

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(Received 27 September 2010; published 27 December 2010)

The γ-ray strength function method is devised to indirectly determine radiative neutron capture cross sections for radioactive nuclei. This method is applied here to the ^{107}Pd ($T_{1/2} = 6.5 \times 10^6$ yr) case. Photoneutron cross sections were measured for ^{105,106,108}Pd near neutron threshold with quasimonochromatic laser-Compton-scattering γ-ray beams. These photoneutron cross sections as well as the reverse radiative neutron capture cross sections for ^{104,105}Pd are used to provide constraints on the ^{107}Pd$(n,\gamma)^{108}$Pd cross section.

DOI: 10.1103/PhysRevC.82.064610 PACS number(s): 25.20.Lj, 28.20.Fc, 28.20.Np

I. INTRODUCTION

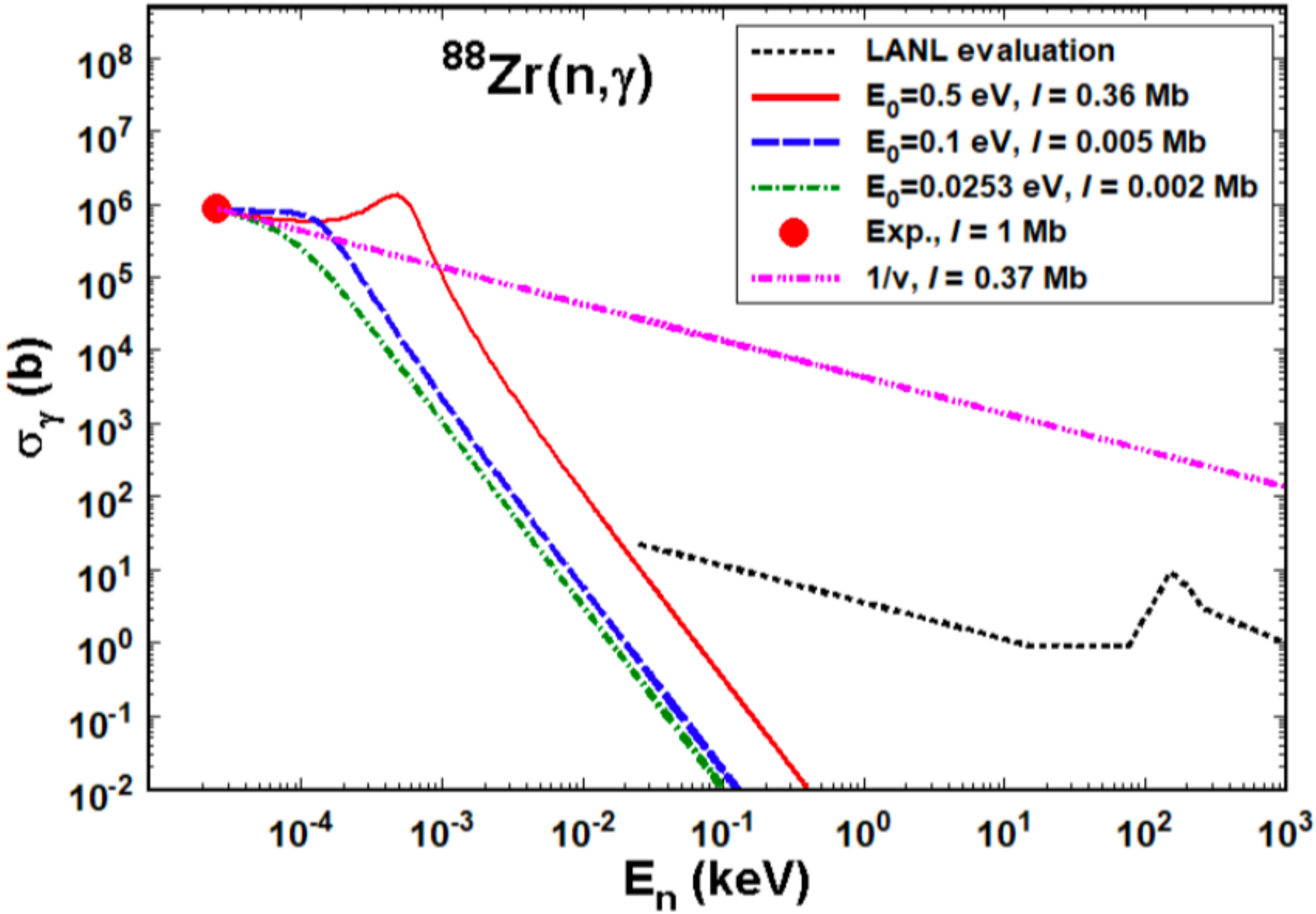
Radiative neutron capture cross sections are of direct relevance for the synthesis of heavy elements referred to as the *s*-process and the *r*-process in nuclear astrophysics and constitute basic data in the field of nuclear engineering. The surrogate reaction technique is in active use in the latter field to indirectly determine radiative neutron capture cross sections for unstable nuclei [1]. We have devised an alternative method based on the γ-ray strength function (γSF), a nuclear statistical quantity that interconnects photoneutron emission and radiative neutron capture in the Hauser-Feshbach model calculation. In this article, we outline the γSF method and apply it to nuclear waste ^{107}Pd ($T_{1/2} = 6.5 \times 10^6$ yr), known as one of long-lived fission products that can be transmuted to a stable ^{108}Pd by neutron capture. The methodology of the γSF method has been developed via a series of photoneutron cross-section measurements made in the past and applied to a long-lived ^{93}Zr (1.53×10^6 yr) and a short-lived ^{95}Zr (64 d) [2]. By comparative observations one finds that the surrogate reaction technique provides (n,γ) cross sections for ^{93,95}Zr that are larger by a factor of ~3 than those of the γSF method. The method was previously introduced in two recent conferences [3,4].

respectively, and $T_{\rm tot} = T_\gamma + T_n$; and g_J is the statistical factor given by $g_J = (2J+1)/[2(2J_A+1)]$ with the spin J_A of the target nucleus. The γ transmission coefficient can be sorted according to the multipolarity $X\lambda$ of the γ transition as

$$T_\gamma(E, J, \pi) = \sum_{\nu,X,\lambda} T^{\nu}_{X\lambda}(\epsilon_\gamma) + \sum_{X,\lambda} \int T_{X\lambda}(\epsilon_\gamma)\rho(E-\epsilon_\gamma)d\epsilon_\gamma. \quad (2)$$

Here $T_{X\lambda}(\epsilon_\gamma)$ is the γ-transmission coefficient for energy ϵ_γ with the multipolarity $X\lambda$ and ρ is the nuclear level density. The first term is a sum of $T_{X\lambda}$ over low-lying discrete states ν relevant to a given multipolarity $(X\lambda)$ and the second term is an integration of $T_{X\lambda}$ over nuclear states $\rho(E-\epsilon_\gamma)d\epsilon_\gamma$ in the energy interval $d\epsilon_\gamma$ at the excitation energy $E-\epsilon_\gamma$. Note that in the second term of Eq. (2), the integration over the relevant spin-parity distribution of the level density is performed according to the γ-decay selection rule.

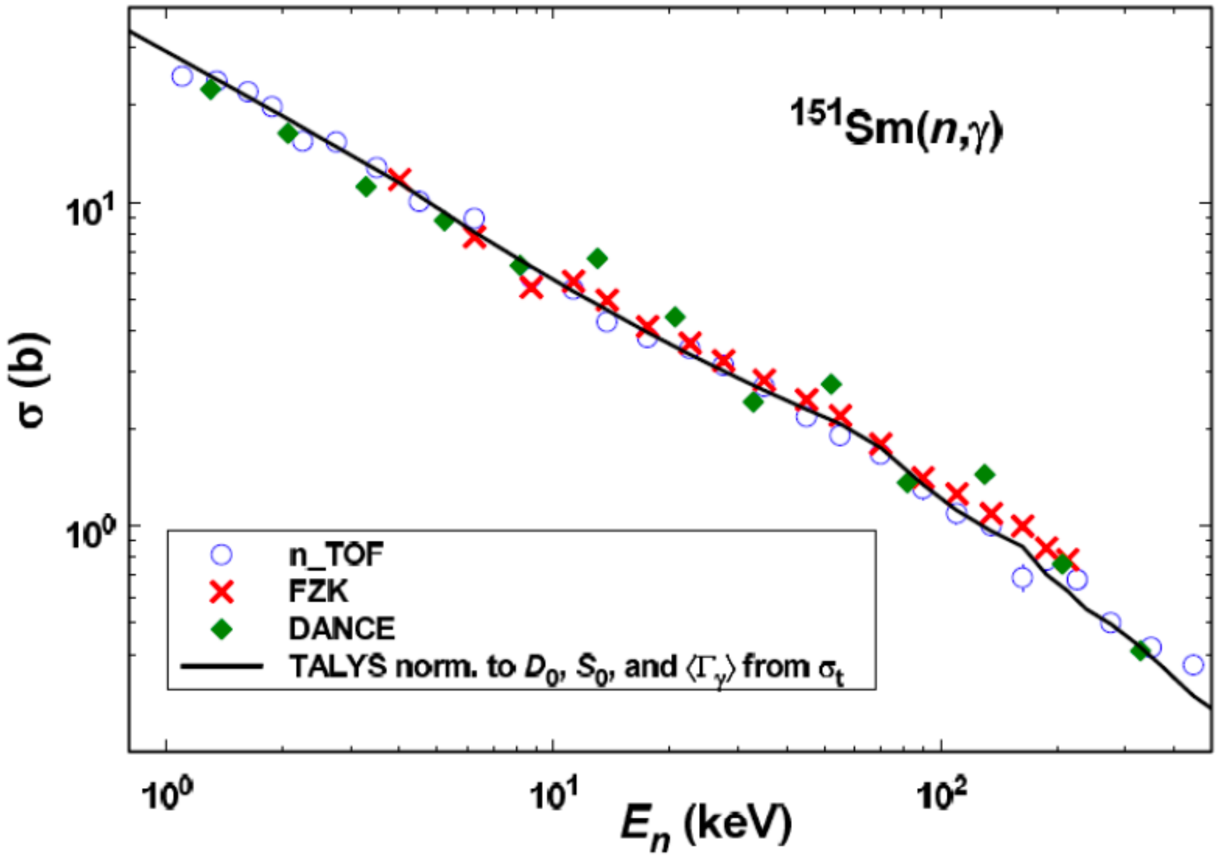
The γ transmission coefficient is uniquely related to the γ-ray strength function $f_{X\lambda}(\epsilon_\gamma)\downarrow$ by $T_{X\lambda}(\epsilon_\gamma) = 2\pi\epsilon_\gamma^{(2\lambda+1)} f_{X\lambda}(\epsilon_\gamma)\downarrow$. Because the radiative decay of a neutron capture state proceeds via cascade transitions, the downward γ strength function for ϵ_γ lower than the neutron separation energy governs the process. Given the γSF, the main source of uncertainties in the HF calculation of the radiative neutron



$^{88}Zr(n,\gamma)$
LANL evaluation
E_0=0.5 eV, I = 0.36 Mb
E_0=0.1 eV, I = 0.005 Mb
E_0=0.0253 eV, I = 0.002 Mb
Exp., I = 1 Mb
1/v, I = 0.37 Mb
σ_γ (b)
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^{-1}
10^{-2}
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^0
10^1
10^2
10^3
E_n (keV)

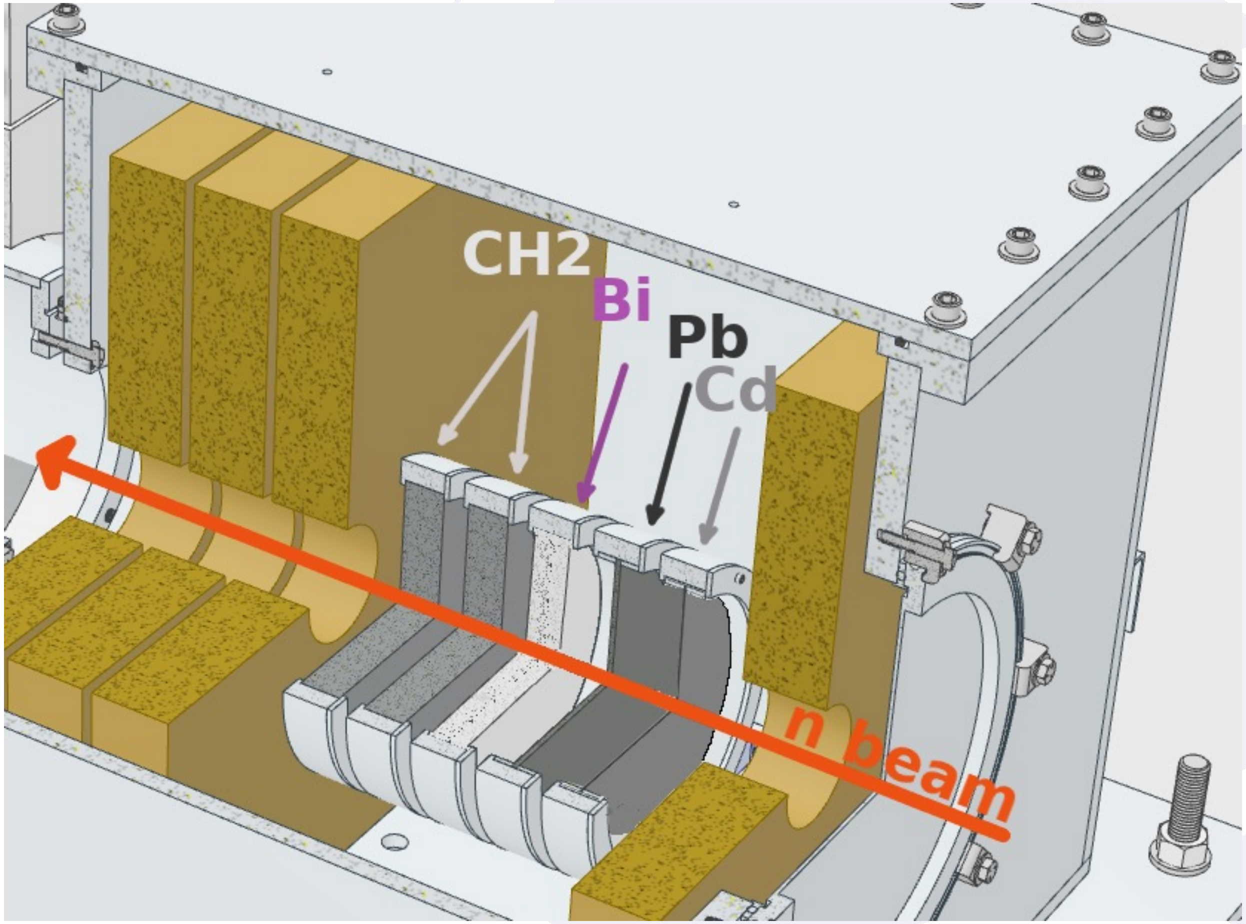
Introduction: Proof of Principle

- Resonance analysis on ^{151}Sm(n,tot) data from RPI
- Normalize TALYS calculations and compare to ^{151}Sm(n,γ) EXFOR data
  P. Koehler, LA-UR-14-21466



Description of the apparatus: Neutron spectrum

- A set of paddles can be used to filter the neutron spectrum
  - Cd, 0.08 cm : Neutron absorption below 0.5 eV
  - Pb, 0.3cm : Photon attenuation
  - Bi, 2.5cm : Background calculations
  - CH_2, 2x2.5cm : Neutron scatterer, photon background



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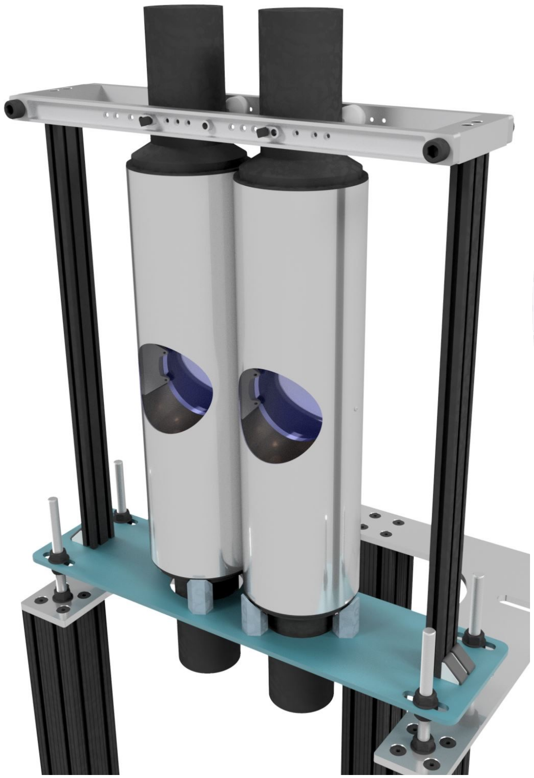
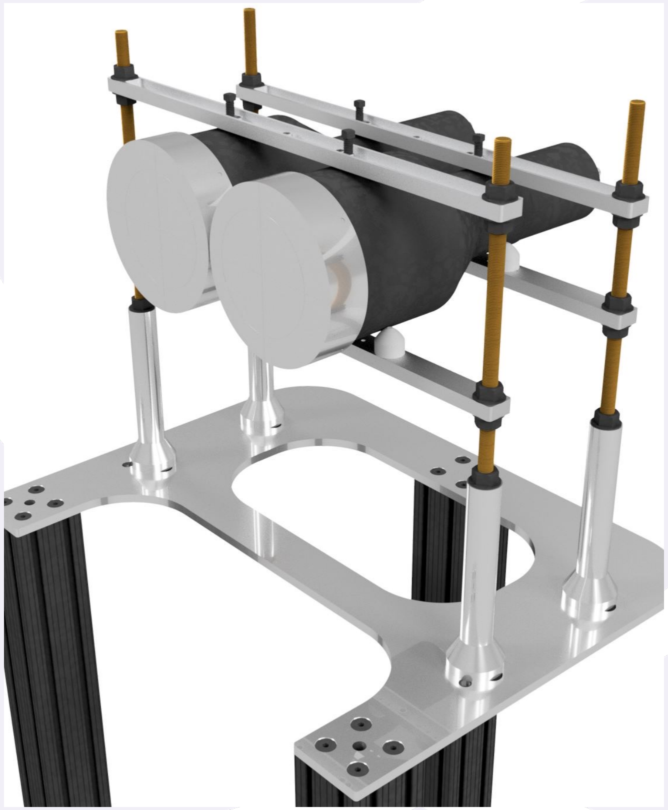
Description of the apparatus: Single pixel detectors

- A commercial ^{235}U loaded fission chamber is installed at ~7.8 m from the neutron source, to monitor the flux.

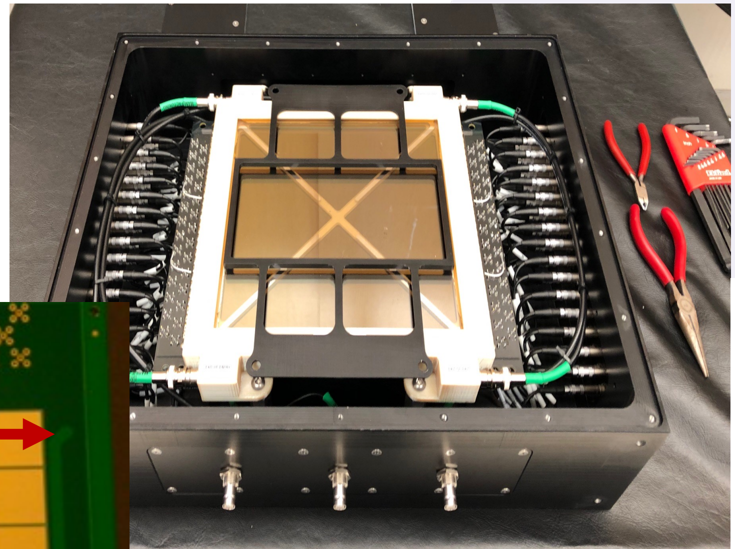
- Two types of single pixel ^{6}Li-glass neutron detectors are available
  - ORELA type: Dual PMT detector.
    Less material in beam
    Poorer light collection
  - Chi-nu type : Single PMT detector.
    PMT in contact with glass
    More material in beam

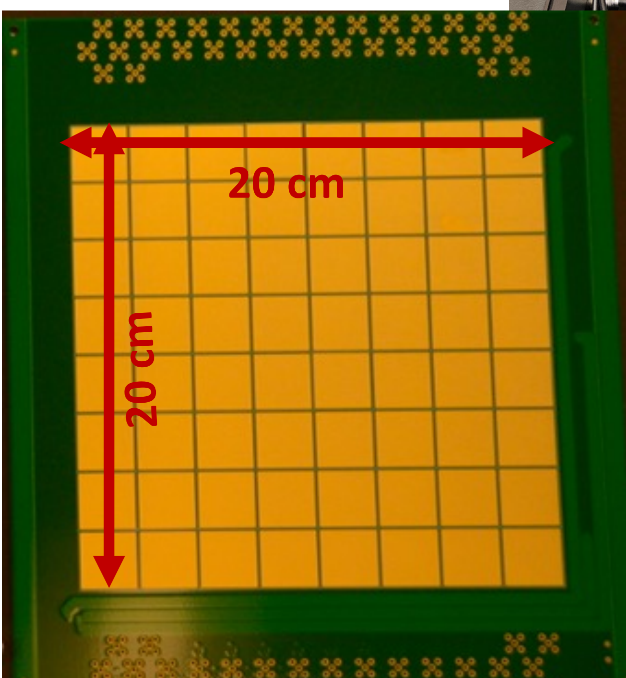


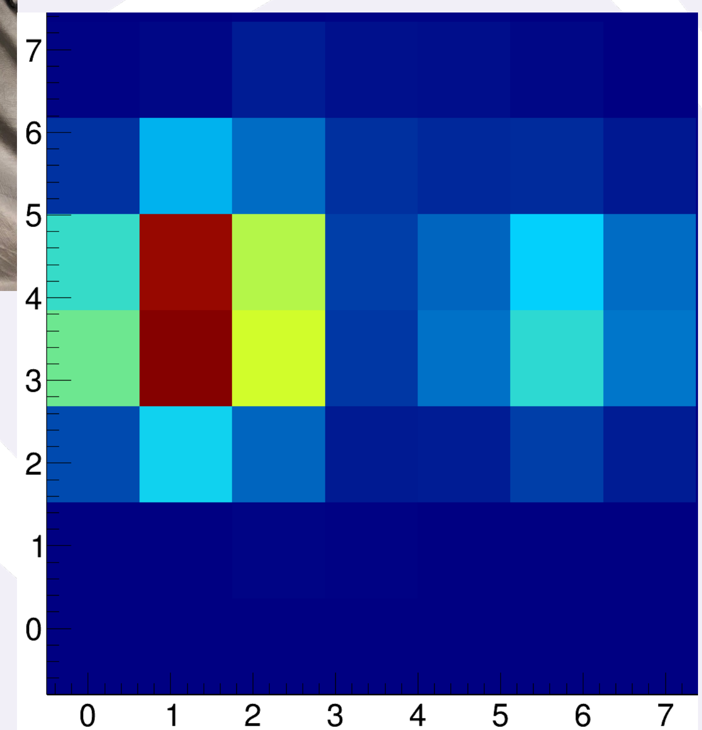


Description of the apparatus: Multipixel detector

- The Large Area Picosecond Photo-detector (LAPPD) is a multi-channel plate (MCP) based detector, able to provide spatial information (nominally ~mm)
- 8 x 8 pads, 20 x 20 cm total coverage area

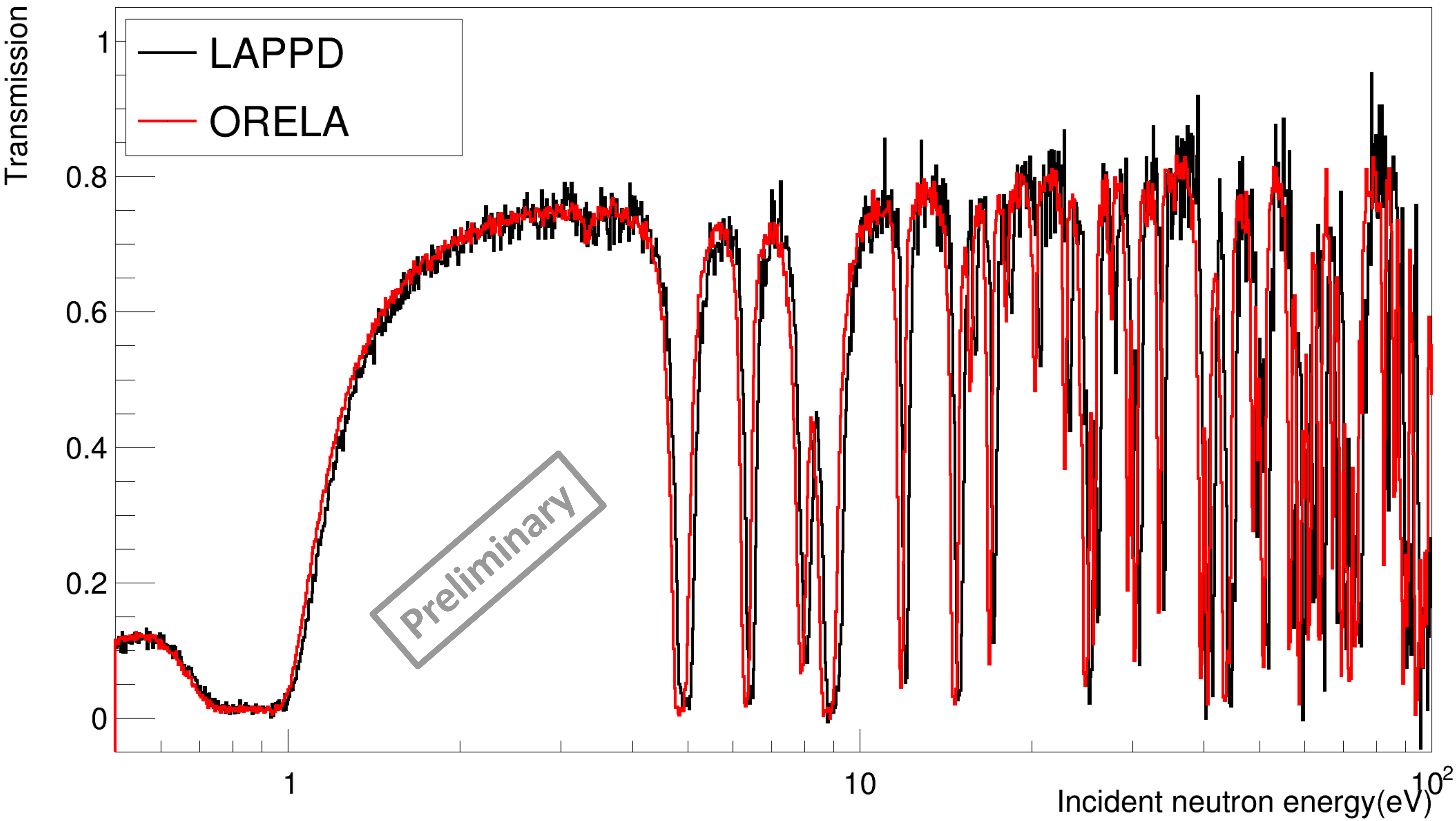






Description of the apparatus: Multipixel detector

- Single pad transmissions agree well with previous DICER data.



The technique in a nutshell

- Cases where the most probable reactions are capture and scattering

$$\sigma_{tot} \approx \sigma_\gamma + \sigma_n$$

Total — Capture — Scattering

- Each resonance can be analytically described by its **Energy** and **Width**

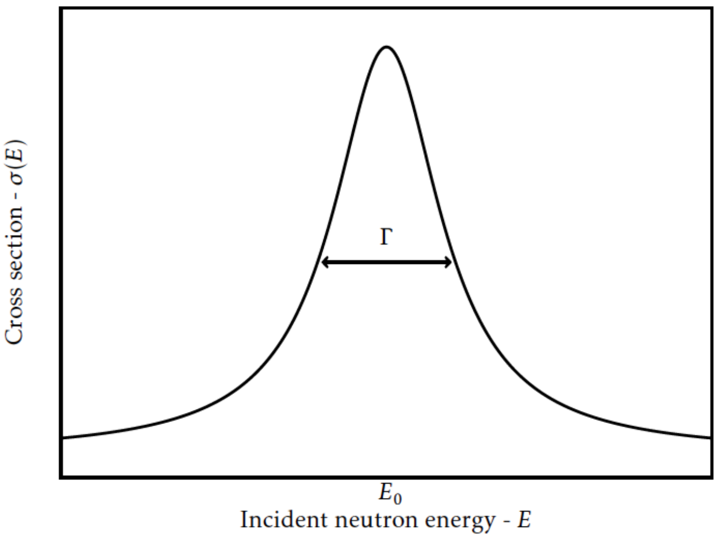
- Similar to cross sections, resonance widths are:

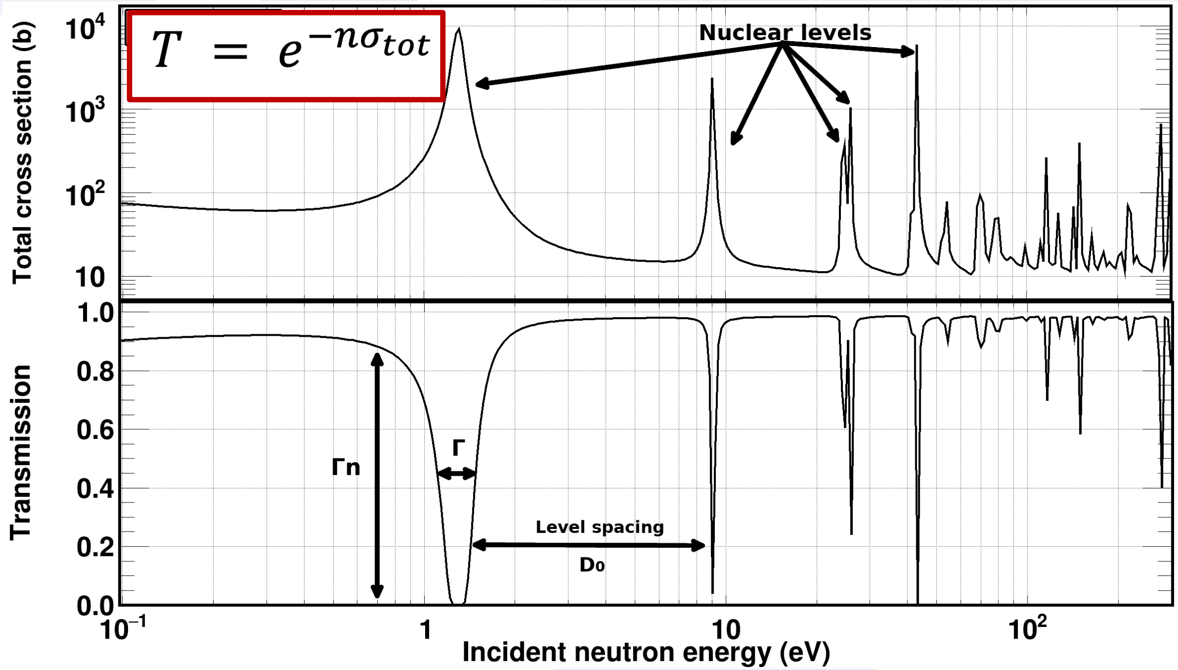
$$\Gamma_{tot} = \Gamma_\gamma + \Gamma_n$$

Total — Capture — Scattering

- The average (n,γ) cross section will be:

$$< \sigma_\gamma > = f(< \Gamma_{tot} >, < \Gamma_n >, < D0 >)$$





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Description of the apparatus: Metrology and alignment

- Universal Spatial Metrology Network (USNM)
- Monuments are installed in the flight path cave
- Laser trackers are used to locate the position of the collimators in the flight path cave
- Stations have 5&6 DOF precision alignment systems
- Collimators are fiducialized
- Alignment over the 30 m length must be held to very tight tolerances.
- Alignment to better than 20um and 0.010^0