Direct Measurement of the $^{114}Cd(n,\gamma)$ Cross Section in the 1 eV to 300 keV Energy Range

Ben Crider

Ph.D. Dissertation work of Kofi T. A. Assumin-Gyimah
(defended May 2023)

Acknowledgements: Thank You!

Mississippi State University
- K. Assumin-Gyimah
- B. Crider
- D. Dutta
- D. Araya
- S. Vajdic

LANSCE
- A. Couture
- J. Ullmann
- C. Prokop
- C. Fry
- E. Leal Cidoncha
- G. Rusev

University of Kentucky Group
- S. Yates
- E. Peters

Charles University
- M. Krtička
- S. Valenta

University of Dallas
- S. Hicks

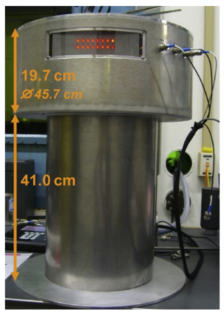
United States Naval Academy
- J. Vanhoy

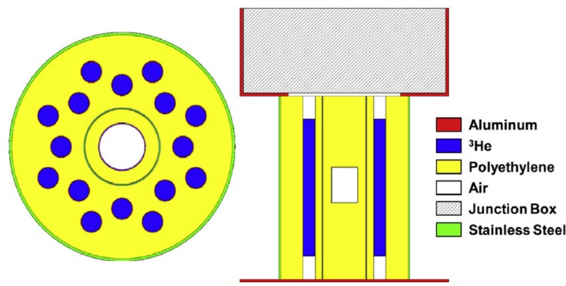
Supported by U.S. DoE FY20/21/22/23 awards SC0021424, SC0021243, SC0021175, SSC000056

Motivation for studying Cd capture: applied physics

- Cadmium data are relevant to safeguards instruments that use Cd in non-destructive assay techniques [1, 2]
  - Passive Neutron Albedo Reactivity (PNAR)
  - Self-Interrogation Neutron Resonance Dosimetry (SINRD)

- PNAR [3]





  - Measure the neutron flux from a fissile material twice and find ratio
  - Fuel is surrounded by a material that maximizes neutron multiplication
  - The low multiplying case created by placing a Cd liner around the the assembly

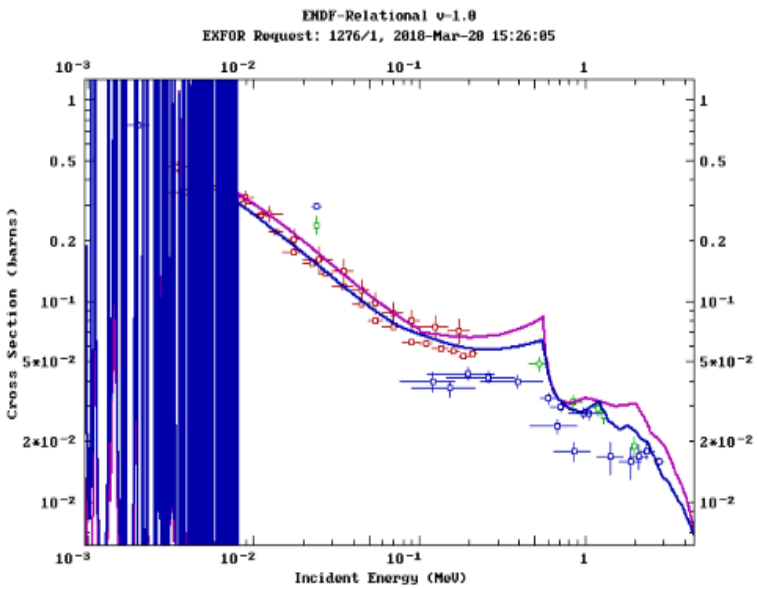
Abundances of "Stable" Cadmium Isotopes

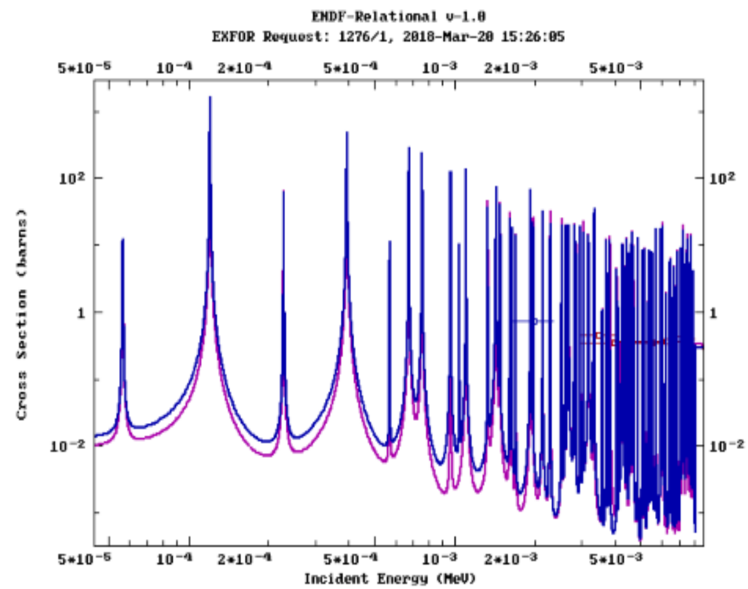
^{106}Cd (1.25%)	stable
^{108}Cd (0.89%)	stable
^{110}Cd (12.47%)	stable
^{111}Cd (12.80%)	stable
^{112}Cd (24.11%)	stable
^{113}Cd (12.23%)	stable
^{114}Cd (28.75%)	stable
^{116}Cd (7.51%)	3.1×10^{19} yrs.

[1] Nuclear Data Needs and Capabilities for Applications, 2015 Workshop at LBNL (2015)
[2] Bahran *et al.* from 55th annual Institute of Nuclear Materials Management meeting
[3] Radiation Measurements **61**, 83 (2014).

Motivation from a nuclear data perspective

- Overall, there is a lack of data on ^{114}Cd capture cross sections
  - Most data come from either Musgrove et al. [1] or Wisshak et al. [2] and often they don't agree within uncertainty



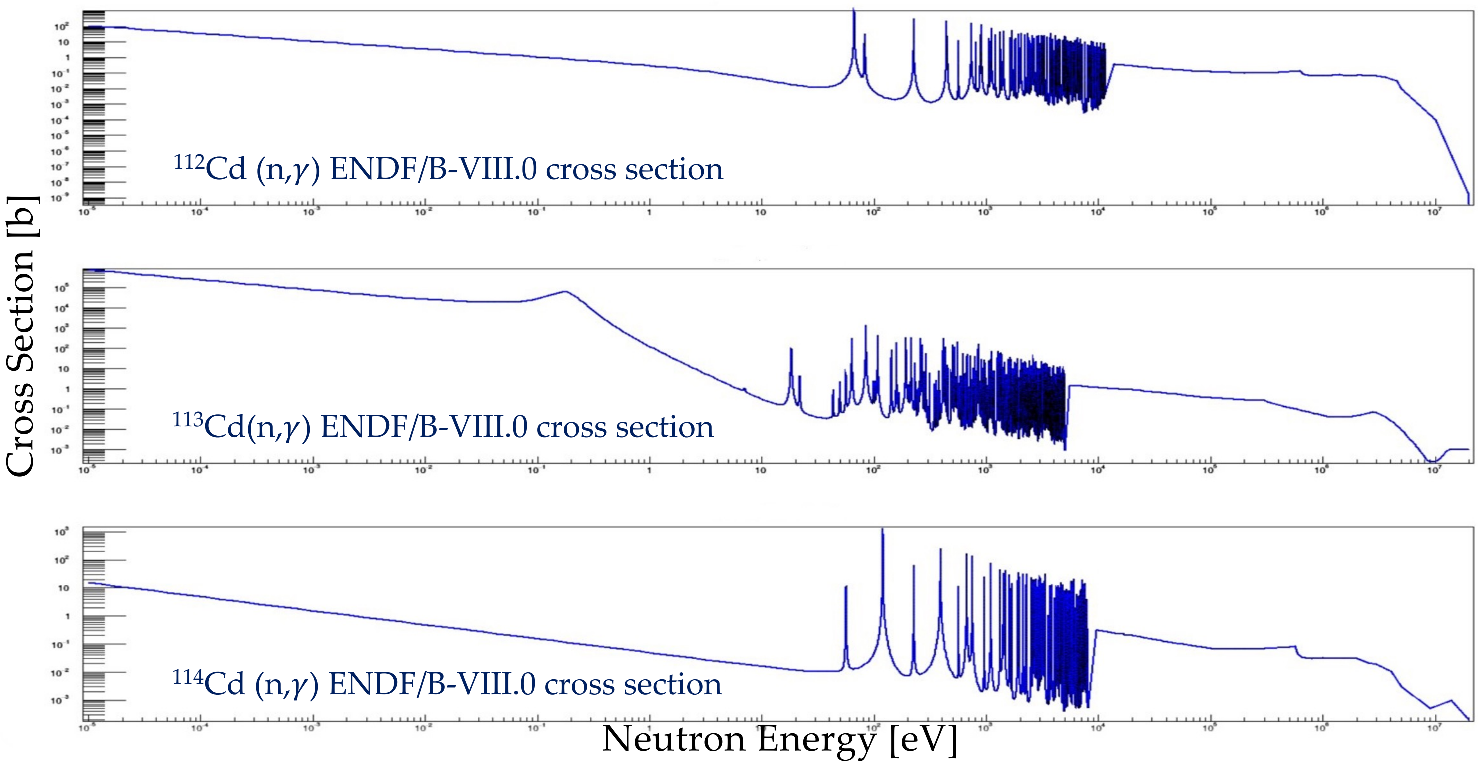


- No direct measurement of resonance region

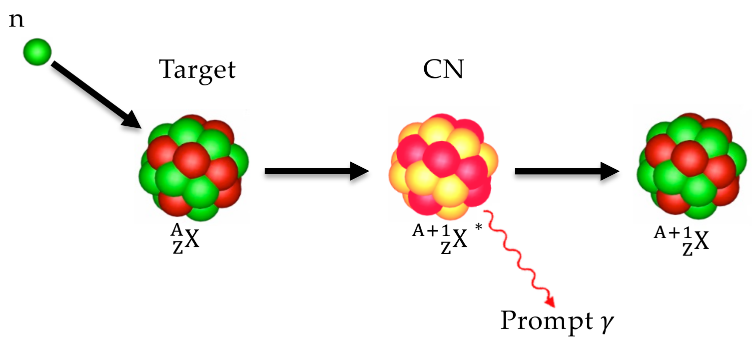
[1] A R de L Musgrove, *et al.* J. Phys. G: Nucl. Phys. **4** 771 (1978)
[2] K. Wisshak, *et al.* Phys. Rev. C **66**, 025801 (2002)

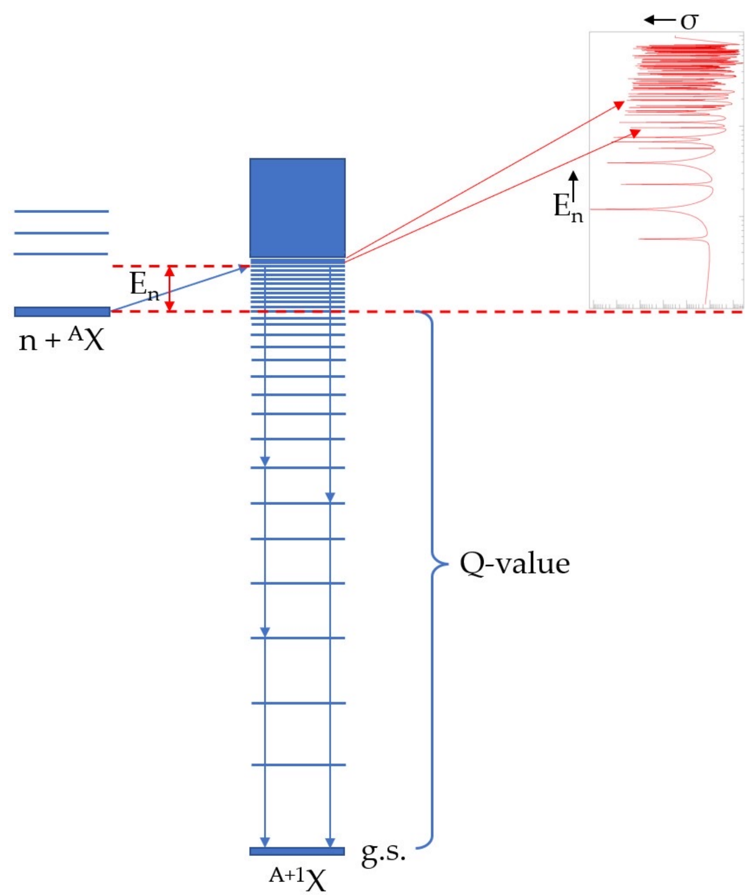
Cd nuclei challenges

- CS different for each nuclide… quite different for ^{113}Cd at low energies



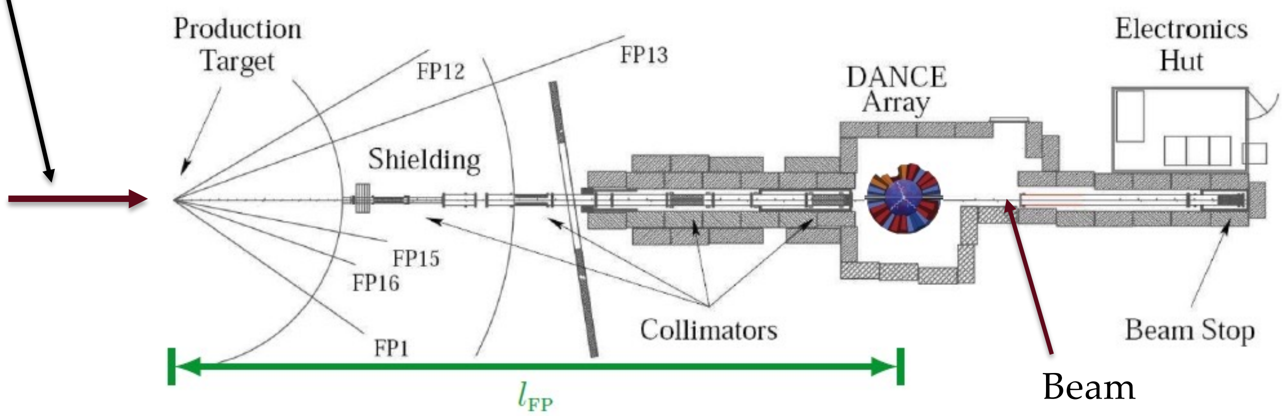
Neutron capture reaction





DANCE Flight Path and Neutron Production

- Proton Beam, E_p = 800 MeV, provided by LANSCE (Los Alamos Neutron Science Center)



J. Ullmann, *Progress of Theoretical Physics*, 92(275), (1996)

*Pulsed, white neutron source

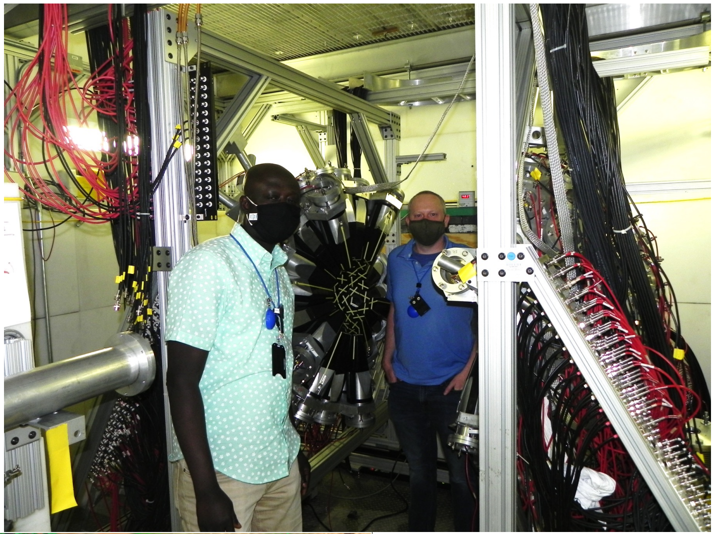
- Use TOF method to determine neutron energy: $E_n = \frac{1}{2} m_n \frac{l_{Fp}^2}{t_{TOF}^2}$

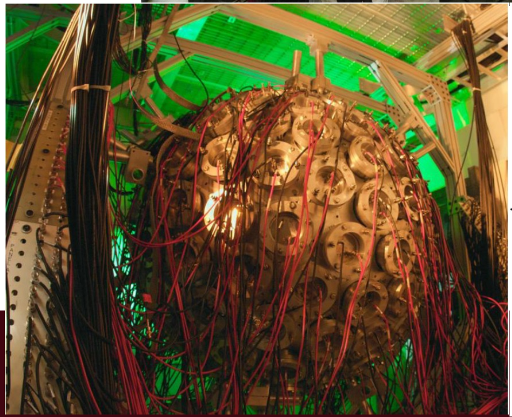
m_n-mass of neutron, l_{FP}-flight path, t_{TOF}-time-of-flight

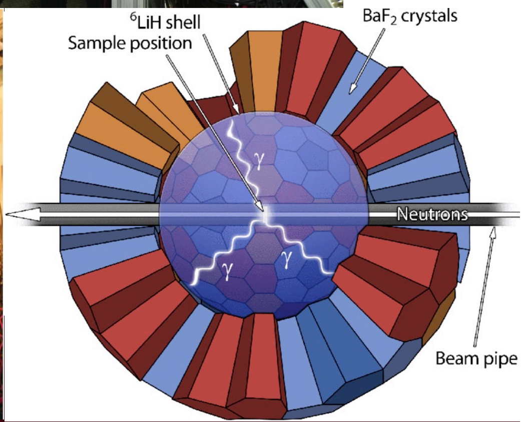
- Total energy measured in DANCE: $E_{sum} = E(\gamma_1) + E(\gamma_2) + E(\gamma_3) + \cdots + E(\gamma_n)$

The Detector for Advanced Neutron Capture Experiments (DANCE)

- 160 BaF_2 detectors in a close packed ~4π geometry (≈3.5 π)
- For a 1 MeV γ DANCE is ~85% efficient
- Relatively flat efficiency curve (70% efficient at ~10 MeV)
- Epithermal neutron beam allowing measurement across wide neutron energy range from 0.01 eV to
  ~500 keV
- 20.28 m flight path







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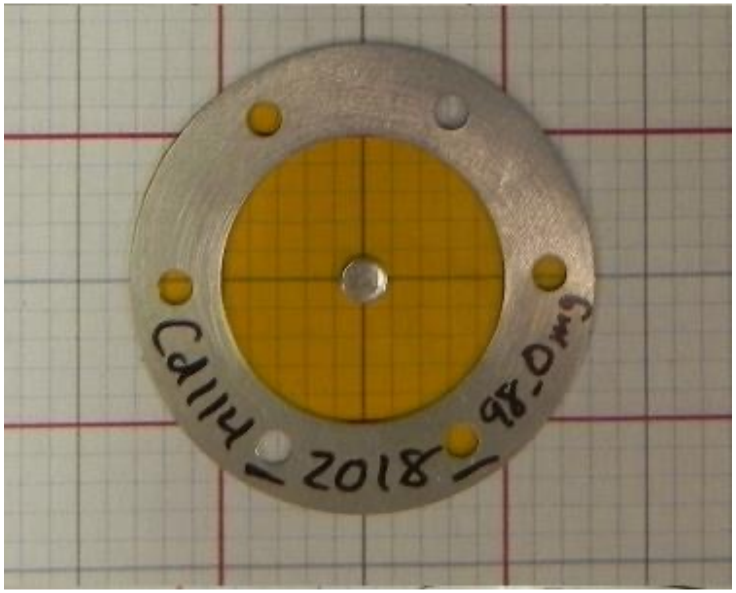
http://lansce.lanl.gov/facilities/lujan/instruments/fp-14/index.php

Samples

- Our sample: enriched ^{114}Cd sample was obtained from NIDC at Oak Ridge
  - Mass of 98.0 mg
  - 98.69% ^{114}Cd with 0.178% ^{116}Cd, 0.48% ^{113}Cd, 0.37% ^{112}Cd, 0.183% ^{111}Cd, 0.086% ^{110}Cd
  - 4-mm diameter
- Other Cd samples
  - 95.10% enriched ^{113}Cd (Mass of 10.7 mg)
  - 98.27% enriched ^{112}Cd (Mass of 95.7 mg)
  - Also have ^{111}Cd and ^{110}Cd
- ^{208}Pb sample
  - Used for scattered neutron background
  - 4-mm diameter
- ^{197}Au sample
  - For beam flux normalization
  - 5-kA thickness
  - 4-mm diameter
- ^{22}Na, ^{88}Y, PuBe
  - For calibrations

All targets seeing neutrons, including ^{197}Au CS normalization and ^{208}Pb scatter subtraction had the same 4-mm diameter!



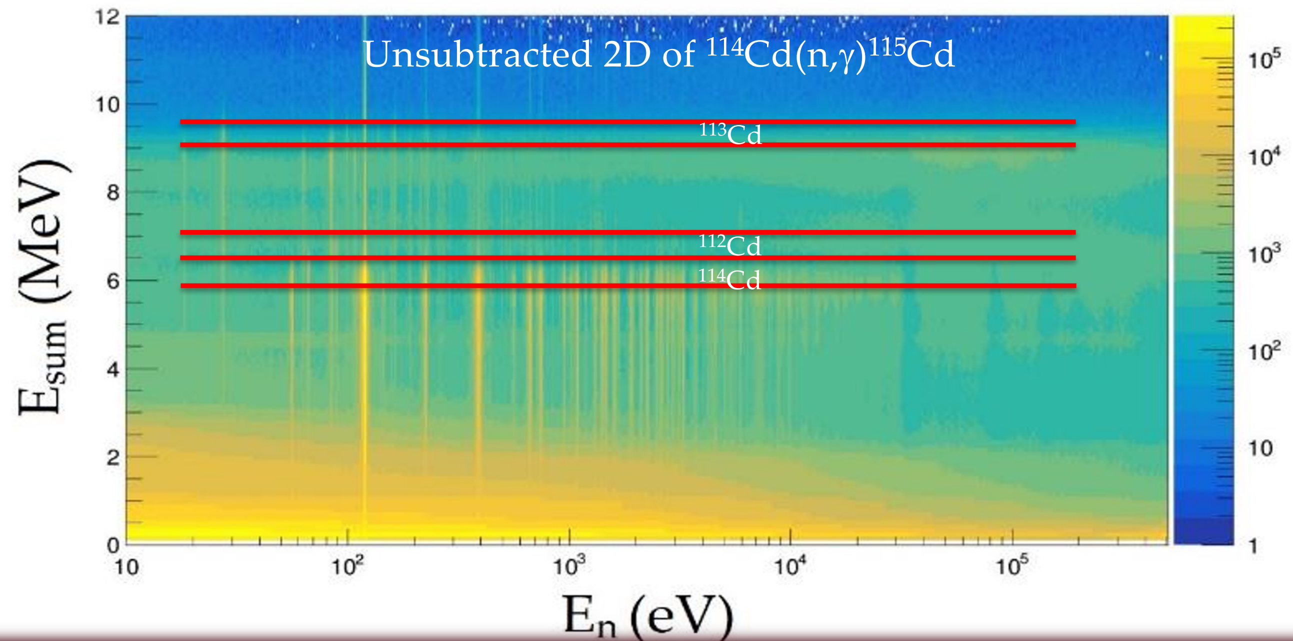
Cross section of (n,γ) reaction using DANCE

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y_{n,\gamma}^{114}(E_n)}{N_{114}} \underbrace{\left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right)}_{\propto \Phi_{BM}^{-1}}$$

- Yield ($Y_{n,\gamma}^{114}(E_n)$) in DANCE
- Neutron flux at beam monitor (BM) locations ($\Phi_{BM}(E_n)$)
- Target flux normalization constant (κ)
- Transmission Correction for Neutron Monitors ($f_a(E_n)$)
- Efficiency of DANCE array (ε_{cut}) for detecting gamma-ray cascades
- Number of atoms (N_{114})

DANCE Data Analysis

- **E_{sum}** – summed energy of all gamma rays detected in a given event
- **E_n** – neutron energy derived from Time-Of-Flight
- Dominating features are resonances in ^{114}Cd(n,γ), contaminants from ^{113}Cd(n,γ) and ^{112}Cd(n,γ) contaminants and scattered neutron background



Targets of Cd Isotopes we've Acquired and their (n,γ) Q-Values

^{110}Cd(n,γ) – 6.89 MeV

^{111}Cd(n,γ) – 9.39 MeV

^{112}Cd(n,γ) – 6.54 MeV

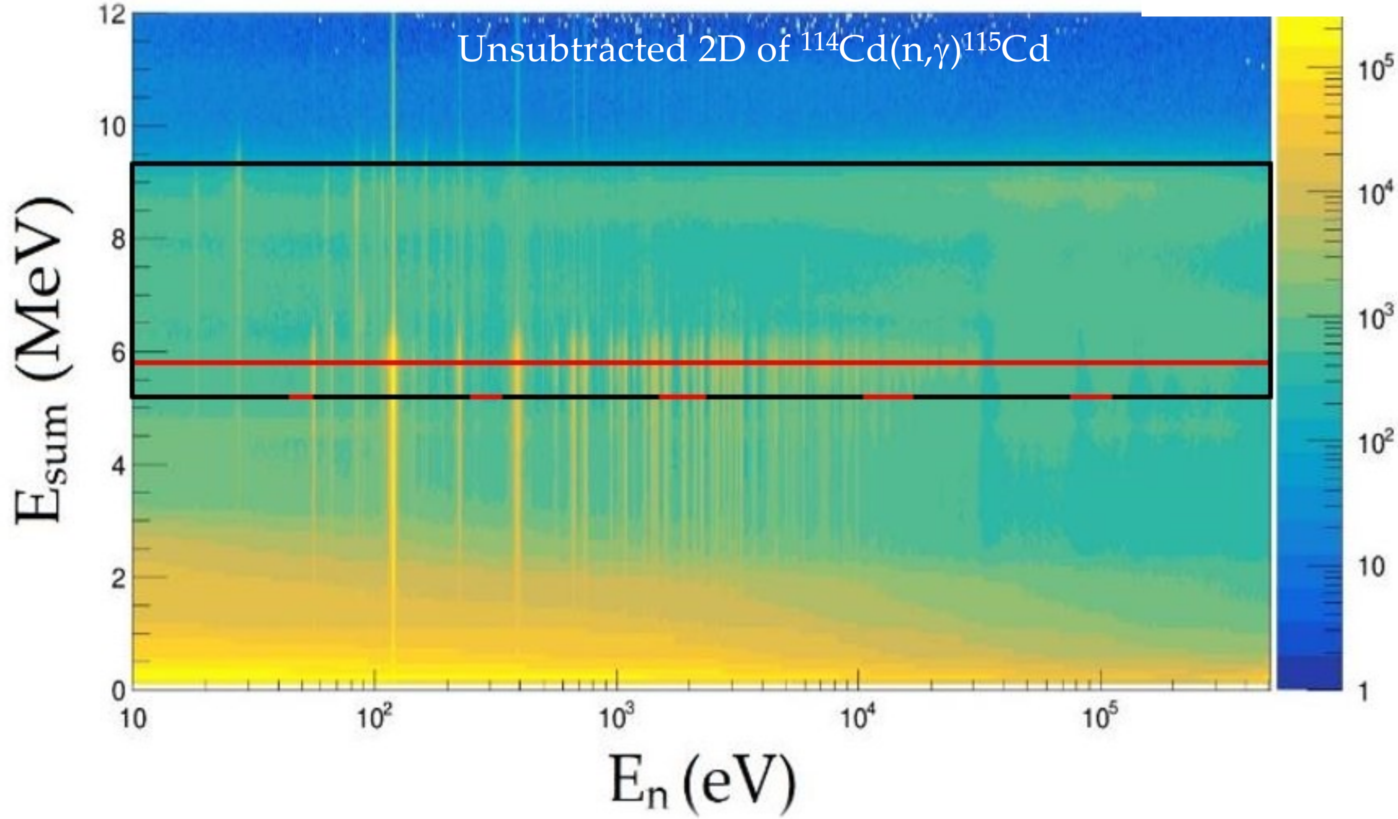
^{113}Cd(n,γ) – 9.04 MeV

^{114}Cd(n,γ) – 6.14 MeV

DANCE Data Analysis: Yields

- Need to characterize contaminants and background

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y_{n,\gamma}^{114}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right.$$



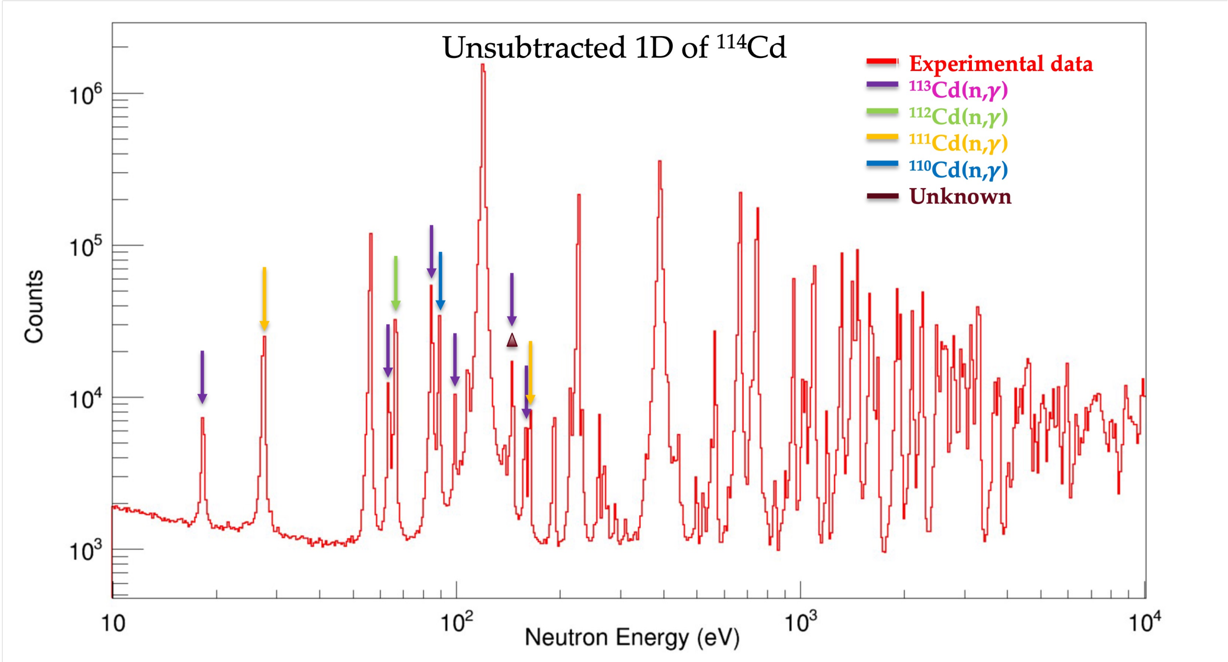
DANCE Data Analysis: Yields

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{\boxed{Y^{114}_{n,\gamma}(E_n)}}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right.$$

- Characterize contaminants and background

- For Cd contaminants:
  - Use data taken from enriched targets (here, ^{112}Cd and ^{113}Cd)
  - Normalize to assay, integrated beam current
  - Subtract from yield

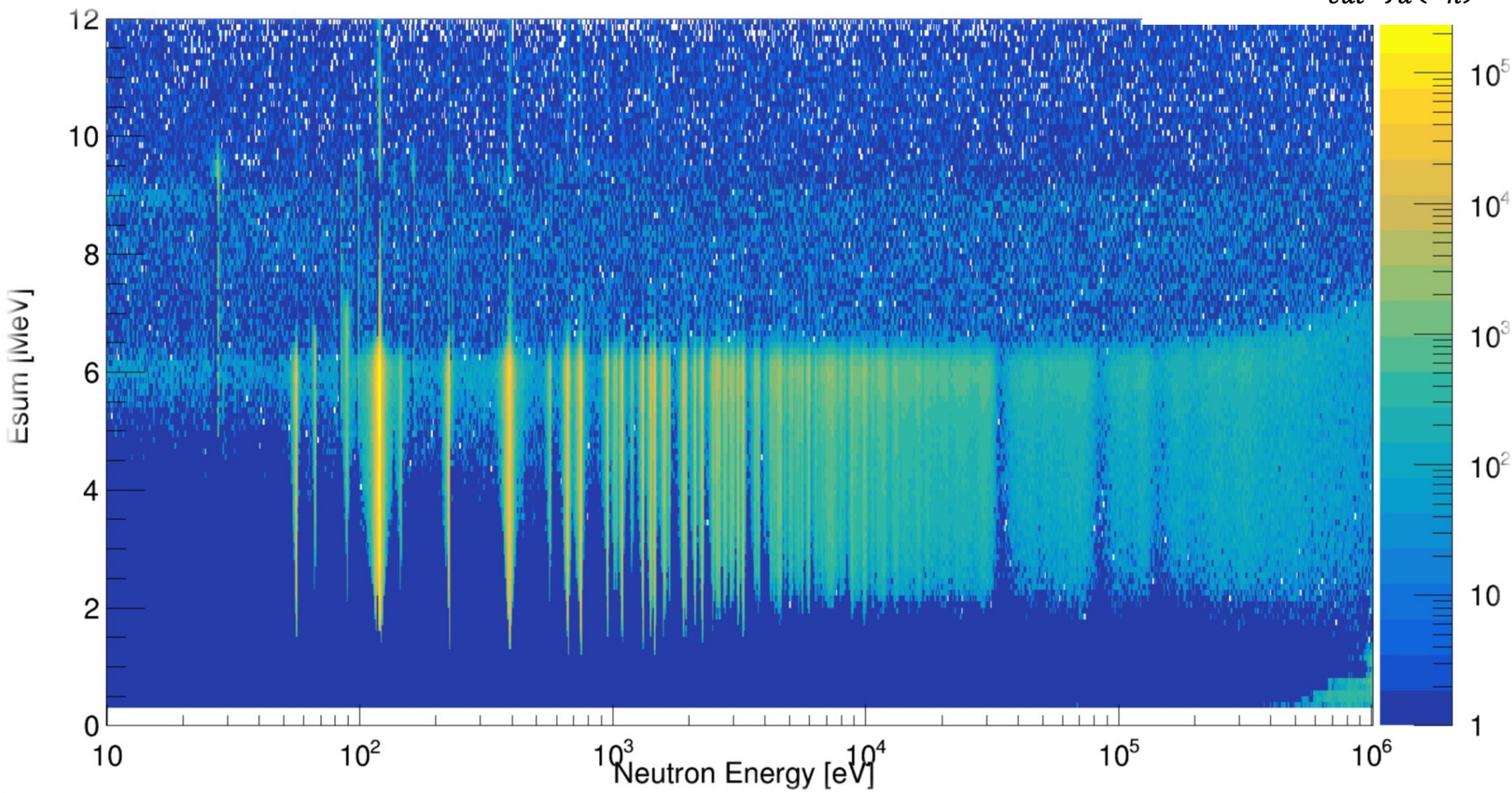
- For scatter background:
  - Use ^{208}Pb target data
  - Normalize Ba capture events from ^{208}Pb to ^{114}Cd data
  - Subtract from yield



DANCE Data Analysis: Yields

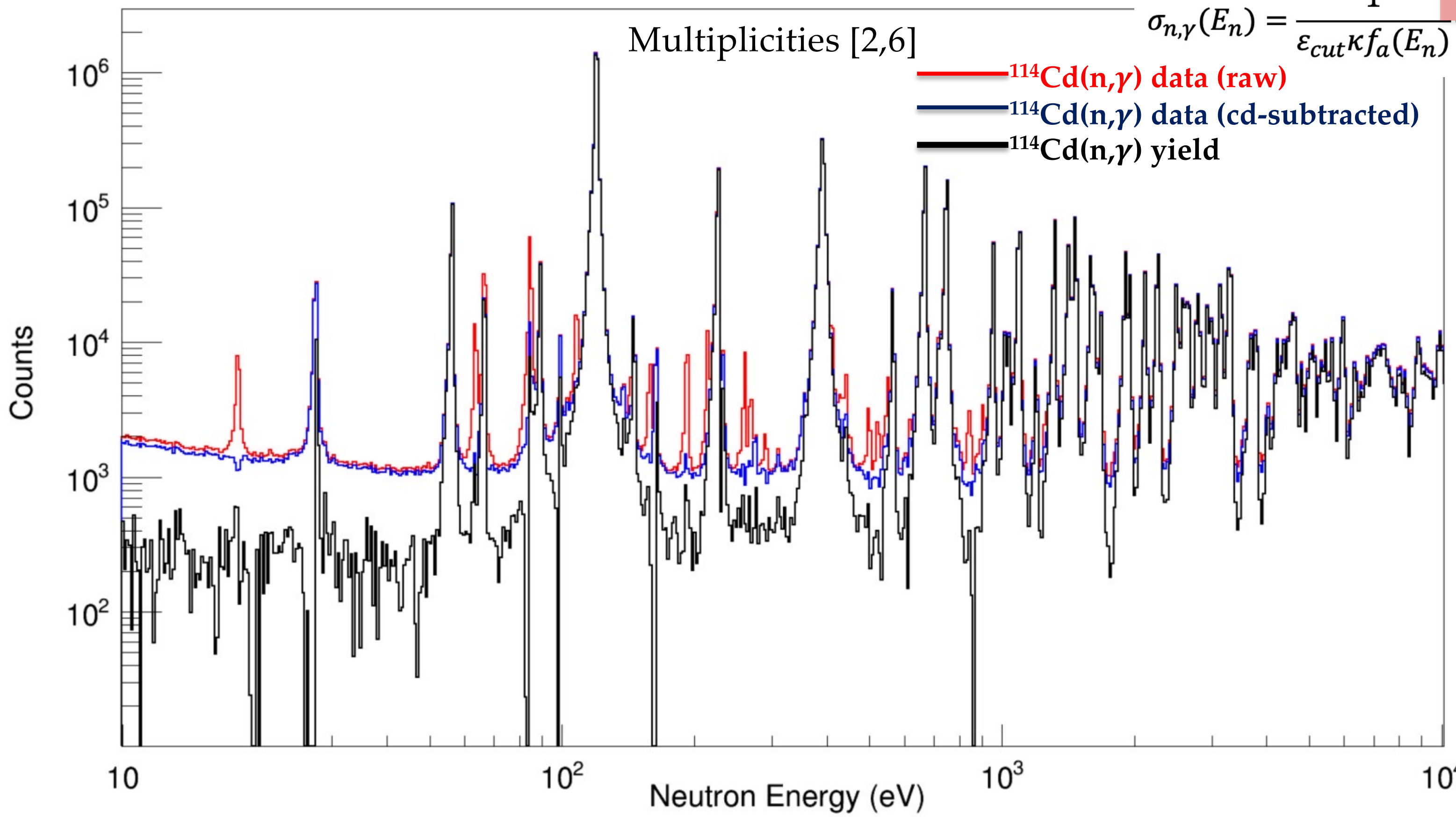
$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)}\frac{\boxed{Y_{n,\gamma}^{114}(E_n)}}{N_{114}}\left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right.$$

Esum vs En, Multiplicities [2,6]



DANCE Data Analysis: Yields

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y^{114}_{n,\gamma}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right)$$



DANCE Beam Monitors and Neutron Fluence

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y_{n,\gamma}^{114}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right)$$

- We used two beam monitors for this experiment:
  - ^{6}Li Beam Monitor: ^{6}LiF converter foil that utilizes the ^{6}Li*(n,t)* reaction
  - ^{235}U Beam Monitor: Gas filled ^{235}U fission chamber that utilized the ^{235}U*(n,f)* reaction

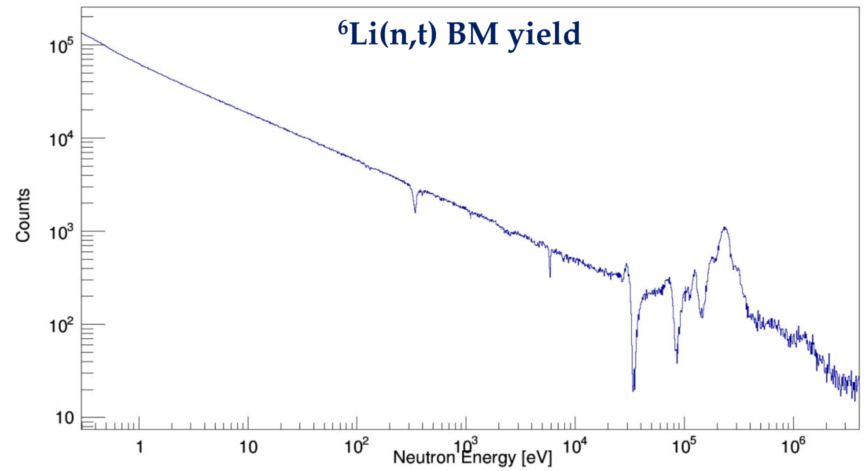
- Neutron flux is proportional to the monitor yields divided by the salient cross section (Y/σ)

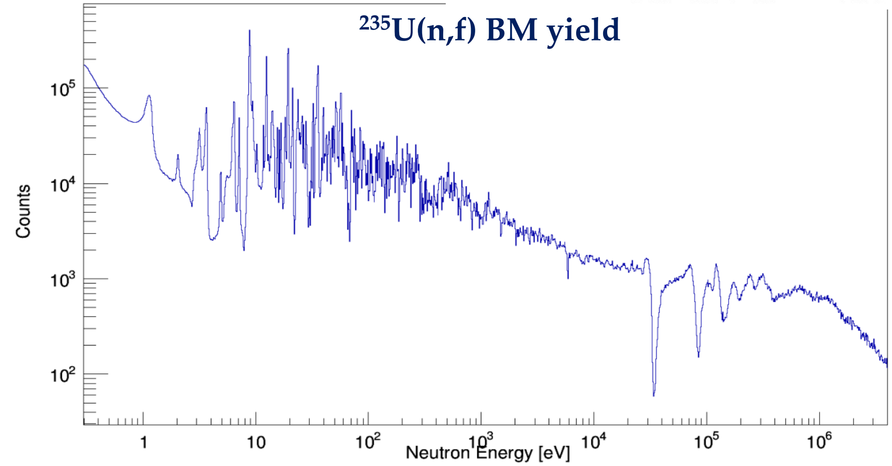
$$\Phi_{BM}(E_n) \propto \frac{Y_{BM}(E_n)}{\sigma_{BM}(E_n)} \qquad (1)$$

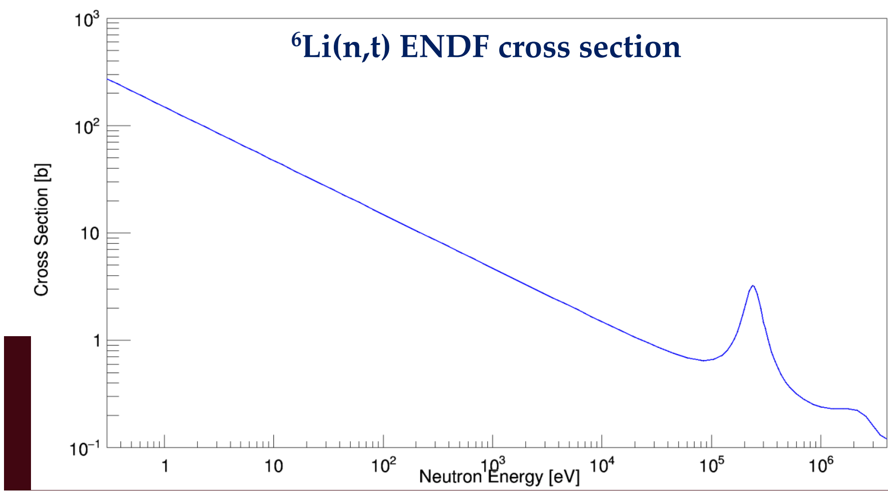
- ^{6}Li monitor used from 1 eV to 5 keV, ^{235}U beam monitor (BM) from 3 keV to 1 MeV, ^{235}U monitor normalized to ^{6}Li BM between 3-5 keV

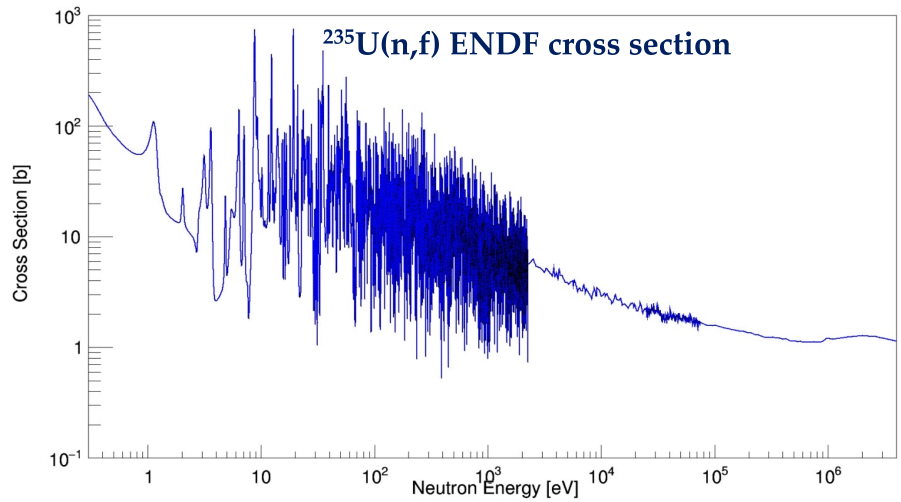
DANCE Beam Monitors and Neutron Fluence

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y^{114}_{n,\gamma}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right)$$









Neutron flux at sample position: normalization to ^{197}Au

- Flux at sample position

$$\Phi(E_n) = \kappa \frac{Y_{BM}(E_n)}{\sigma_{BM}(E_n)} \qquad (1)$$

- The yield is related to cross section by

$$Y_{n,\gamma}^{Au}(E_n) = \Phi(E_n) N_{Au} \sigma_{n,\gamma}^{Au}(E_n) \qquad (2)$$

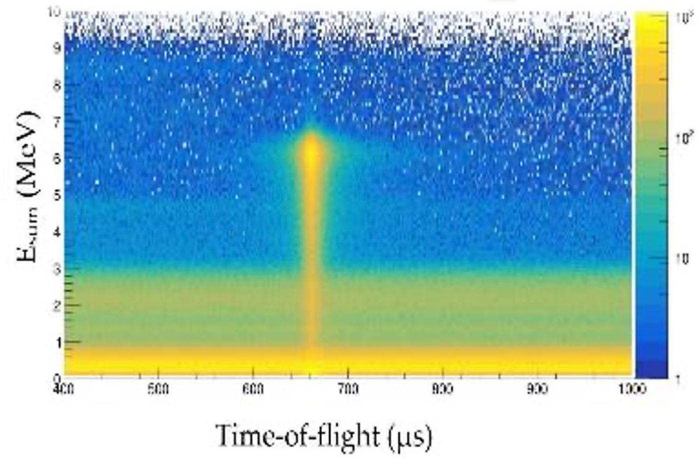
- Substituting (1) into (2):

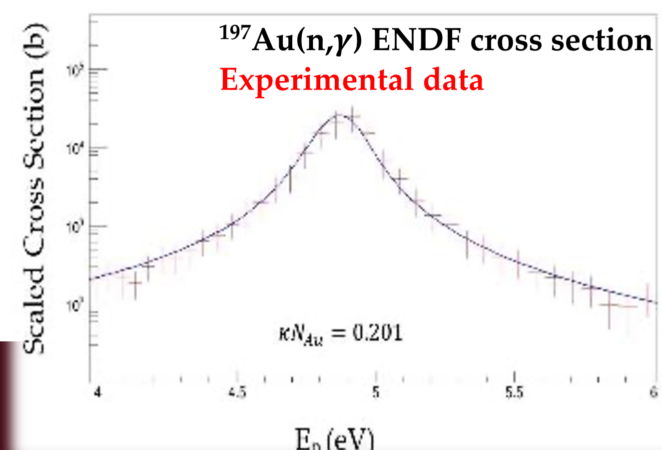
$$Y_{n,\gamma}^{Au} = \kappa \frac{Y_{BM}(E_n)}{\sigma_{BM}(E_n)} N_{Au} \sigma_{n,\gamma}^{Au}(E_n)$$

$$\sigma_{n,\gamma}^{Au}(E_n) = \frac{1}{\kappa} \frac{Y_{n,\gamma}^{Au}(E_n)}{N_{Au}} \frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)} \qquad (3)$$

- Fitting 4.89 eV resonance in ENDF/B-VIII.0 ^{197}Au(n,γ) cross section (27 kbarns) to ^{197}Au yield gives κN_{Au}.

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut} \kappa f_a(E_n)} \frac{Y_{n,\gamma}^{114}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)} \right)$$



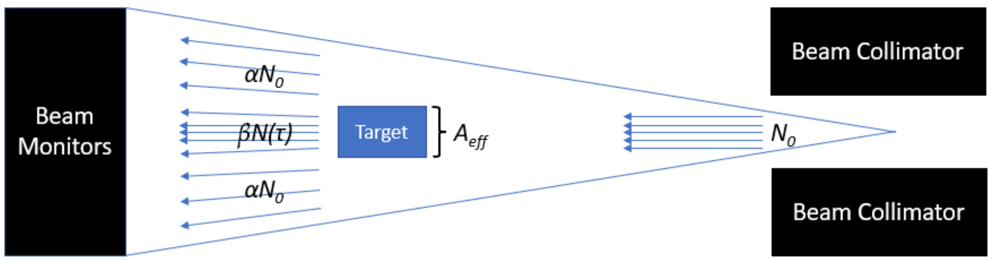


Transmission Correction for neutrons shadowed by target

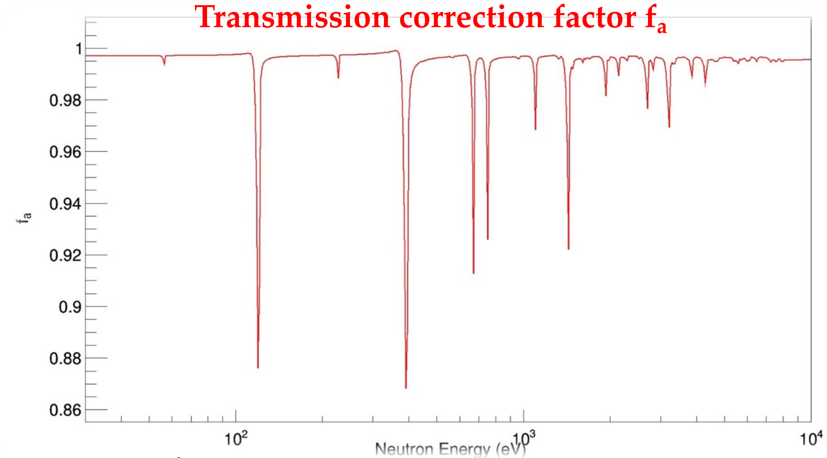
- If beam of N_0 neutrons is incident on a target of thickness dx, and cross section σ_{tot}, the number of attenuated neutrons $-dN$ is given by

$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y_{n,\gamma}^{114}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right)$$

$$dN = -N_0\sigma_{tot}dx,\ N(x) = N_0 e^{-\sigma_{tot}x}$$



Transmission correction factor f_a



- N_{BM} = neutrons that don't interact with target + fraction transmitted through target

$$N_{BM} = \alpha N_0 + \beta N(\tau),\quad \alpha + \beta = 1,\quad f_a = \frac{N_{BM}}{N_0} = 1 - \beta + \beta e^{-\sigma_{tot}\tau}$$

- In BM data, scale region of maximal transition (375 eV, just below largest resonance at 392 eV) to 1, use fact that largest resonance essentially absorbs all neutrons in ^{114}Cd sample, and find transmission factor

Cascade Efficiency Determination

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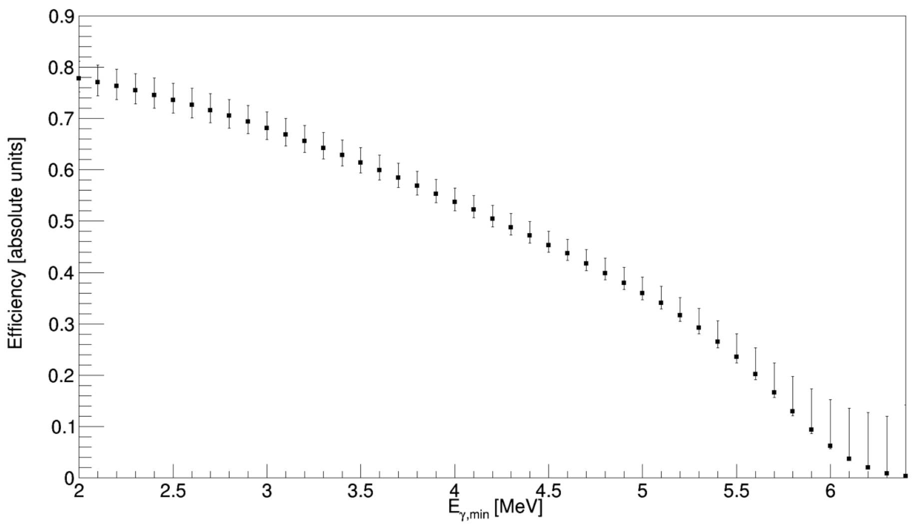
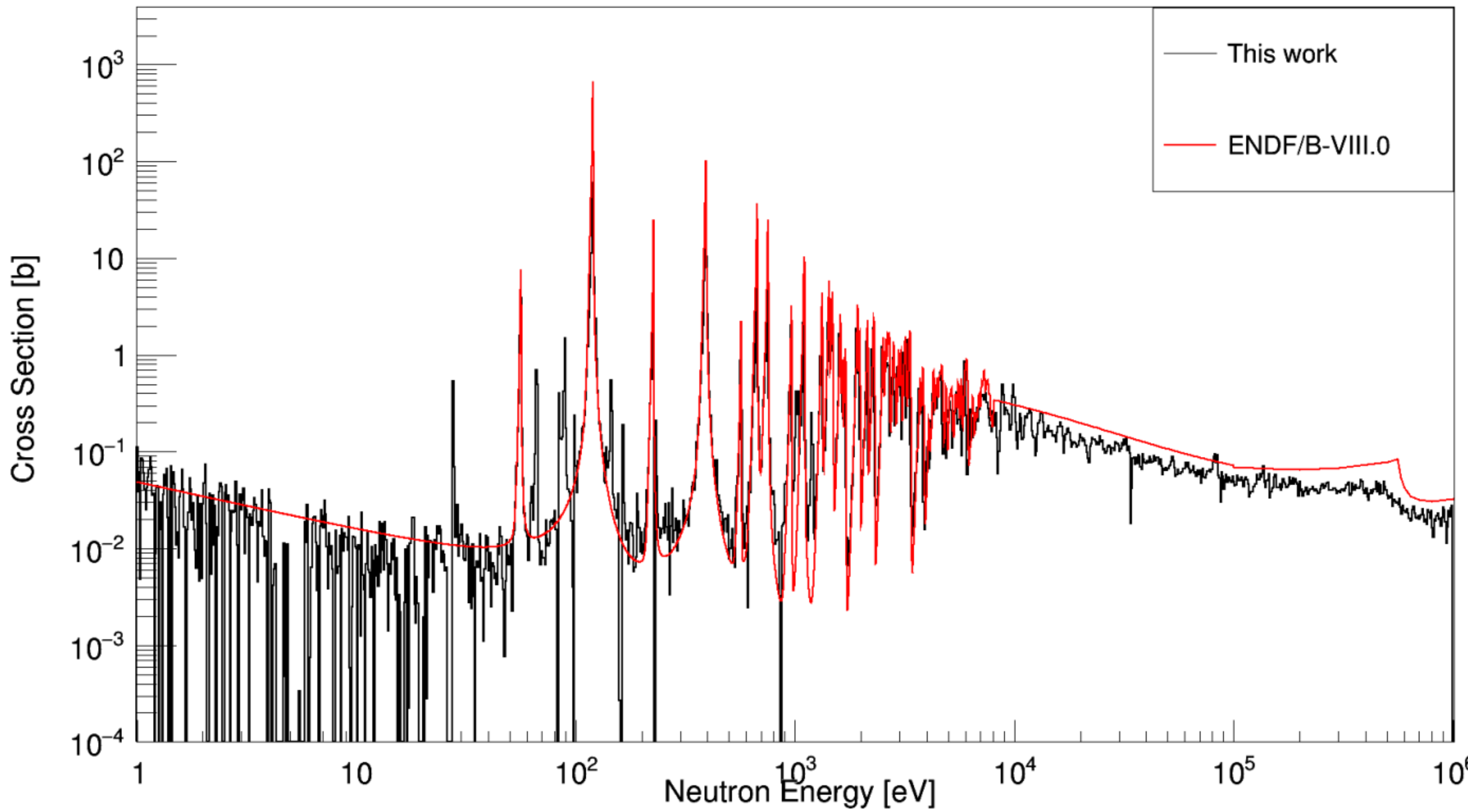
$$\sigma_{n,\gamma}(E_n) = \frac{1}{\varepsilon_{cut}\kappa f_a(E_n)} \frac{Y^{114}_{n,\gamma}(E_n)}{N_{114}} \left(\frac{\sigma_{BM}(E_n)}{Y_{BM}(E_n)}\right)$$

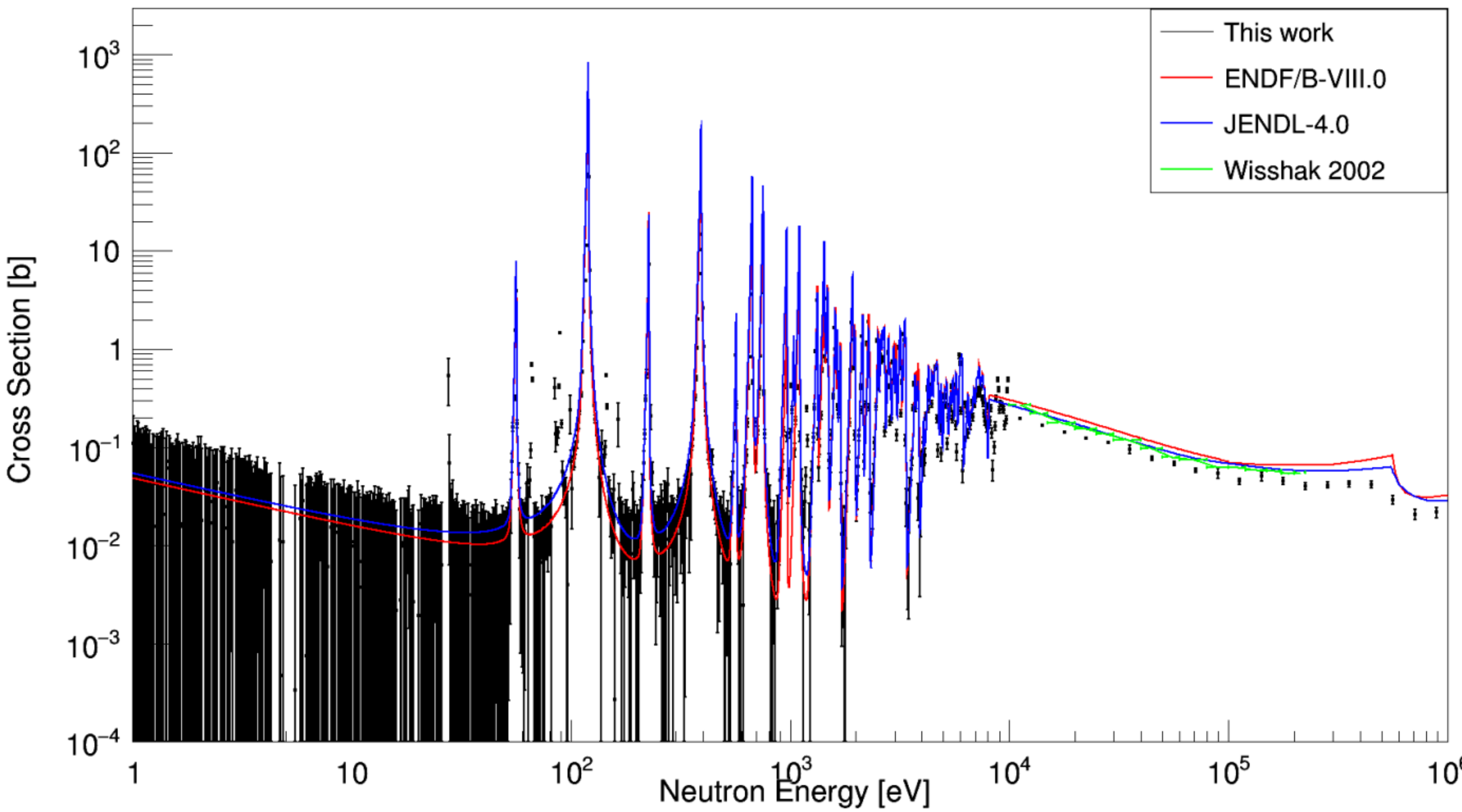
- Efficiency of DANCE for our measurement was determined using DICEBOX and GEANT4
  - DICEBOX is used to simulate γ cascades from neutron capture
  - It uses a statistical model produced from a combination of various nuclear level density models (NLDs) and photon strength functions (PSFs)
- GEANT4 model of DANCE was used to process the γ-cascade capture events
- GEANT4 simulation of the DANCE array response was done with target, detector, and beamline components
- Validated DICEBOX/GEANT4 simulation models by comparing them to the experimental spectra



Preliminary Differential Cross Section



Preliminary Differential Cross Section contd.



Conclusion

- We performed a direct measurement of the ^{114}Cd(n, γ)^{115}Cd reaction cross section using DANCE
- We report new cross section for this reaction from 1 eV to 300 keV
- The measured ^{114}Cd(n, γ)^{115}Cd cross section may have impact on NDA techniques and s-process nucleosynthesis models that use these cross sections as input
  - Currently working on assessing impacts of new data
  - The cross section have been converted to new MACS from 5 keV to 100 keV
  - We have been in communications with safeguards group at LANL to discuss impact on PNAR simulations

Acknowledgements: Thank You!

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Supported by U.S. DoE FY20/21/22/23 awards SC0021424, SC0021243, SC0021175, SSC000056