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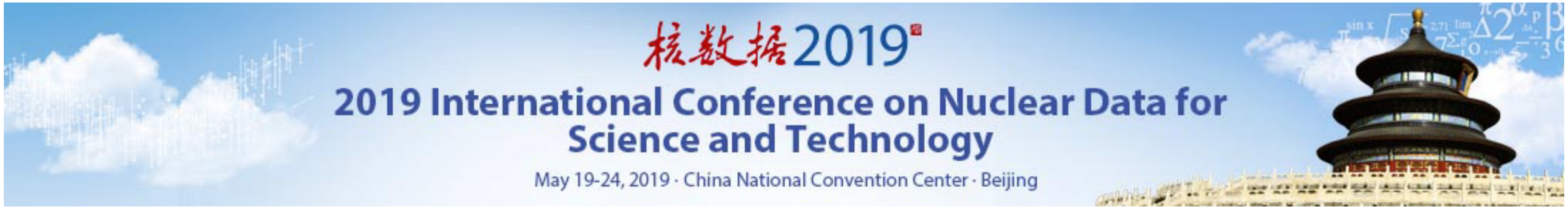
Measurement and analysis of ^{155,157}Gd(n,γ) from thermal energy to 1 keV

Cristian Massimi

Department of Physics and Astronomy



Outline

- **Motivation**

  Nuclear technology, Neutron capture therapy, detectors

- **The (n,γ) measurement at n_TOF (CERN)**

  C_6D_6 with PHWT, 4 enriched and metallic samples

- **Analysis and results**

  Quality checks, assessment of uncertainty, determination of resonance parameters.

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On behalf of the n_TOF Collaboration

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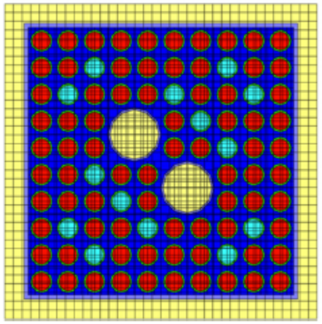
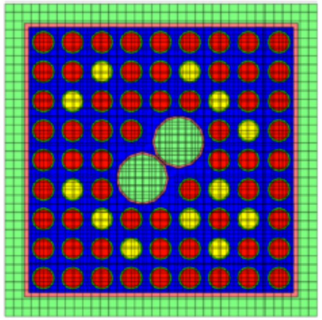
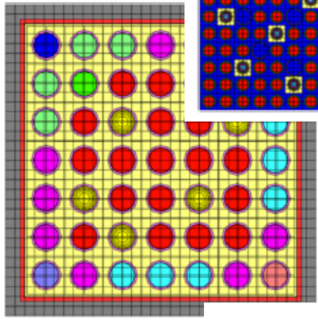
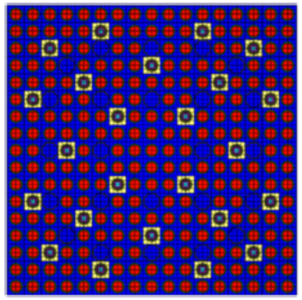
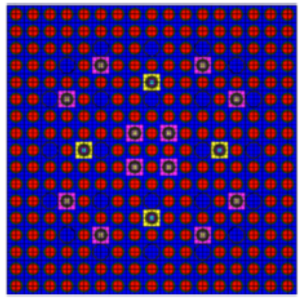
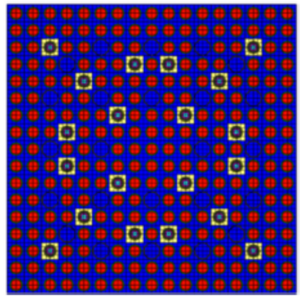
Motivation

Burnable neutron poison

Nuclide-Reaction	Contrib. to Uncertainty in k (% Δk/k)	Rank
^{235}U $\bar{\nu}$ (ave. neut. mult.)	2.70E-01	1.00
^{238}U(n,γ)	1.97E-01	0.81
^{235}U(n,γ)	1.43E-01	0.64
^{235}U(n,f)	1.43E-01	0.56
^{235}U(n,f) vs ^{235}U(n,γ)	1.21E-01	0.54
^{238}U(n,n')	1.20E-01	0.51
^{235}U χ (fiss. neut. spec.)	1.13E-01	0.45
^{238}U $\bar{\nu}$	7.11E-02	0.32
^{157}Gd(n,γ)	6.03E-02	0.26
^{155}Gd(n,γ)	4.48E-02	0.20
^{92}Zr(n,γ)	4.29E-02	0.16
^{1}H(n,γ)	3.67E-02	0.14
^{91}Zr(n,γ)	3.48E-02	0.13
^{1}H(n,n)	3.13E-02	0.12
^{90}Zr(n,γ)	2.82E-02	0.10

EPJ Nuclear Sci. Technol. 3, 21 (2017)

DOI: 10.1051/epjn/2017015

EPJ Nuclear Sciences & Technologies

Available online at:
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REGULAR ARTICLE OPEN ACCESS

Reassessment of gadolinium odd isotopes neutron cross sections: scientific motivations and sensitivity-uncertainty analysis on LWR fuel assembly criticality calculations

Federico Rocchi[1,*], Antonio Guglielmelli[1], Donato Maurizio Castelluccio[1], and Cristian Massimi[2,3]

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[3] INFN, Via Irnerio, 46, 40126 Bologna, Italy

The **uncertainty** on **Gd** cross sections gives the **largest contribution** to the uncertainty on k_{eff} **after ^{235,238}U.**

n_TOF

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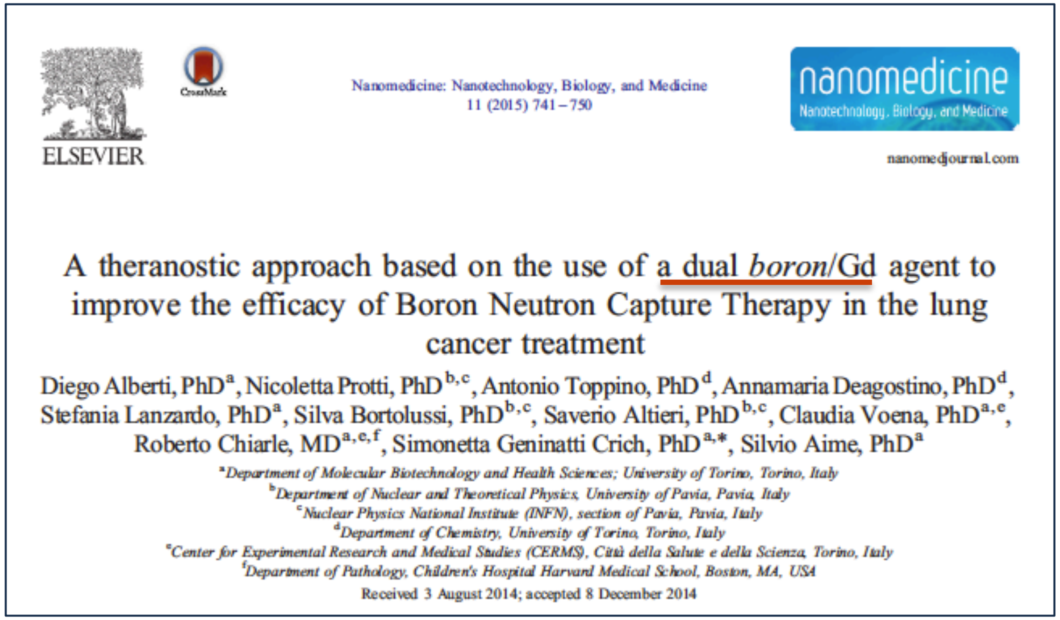
Motivation


Nanomedicine: Nanotechnology, Biology, and Medicine
11 (2015) 741–750
nanomedjournal.com

A theranostic approach based on the use of a dual *boron*/Gd agent to improve the efficacy of Boron Neutron Capture Therapy in the lung cancer treatment

Diego Alberti, PhD[a], Nicoletta Protti, PhD[b,c], Antonio Toppino, PhD[d], Annamaria Deagostino, PhD[d], Stefania Lanzardo, PhD[a], Silva Bortolussi, PhD[b,c], Saverio Altieri, PhD[b,c], Claudia Voena, PhD[a,e], Roberto Chiarle, MD[a,e,f], Simonetta Geninatti Crich, PhD[a,*], Silvio Aime, PhD[a]
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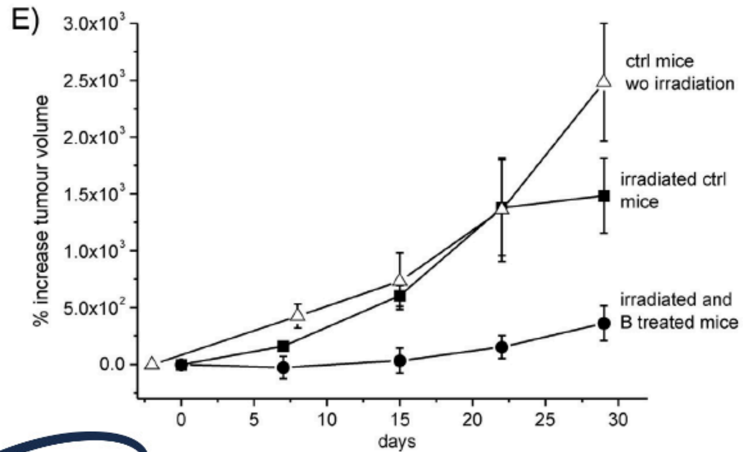
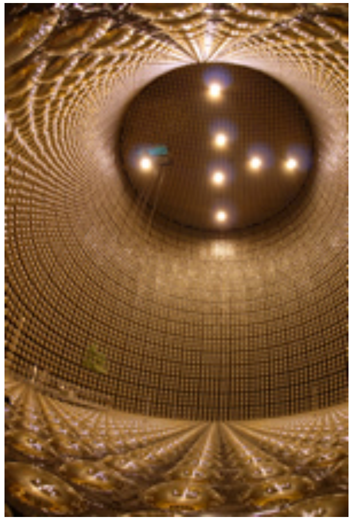
[b]Department of Nuclear and Theoretical Physics, University of Pavia, Pavia, Italy
[c]Nuclear Physics National Institute (INFN), section of Pavia, Pavia, Italy
[d]Department of Chemistry, University of Torino, Torino, Italy
[e]Center for Experimental Research and Medical Studies (CERMS), Città della Salute e della Scienza, Torino, Italy
[f]Department of Pathology, Children's Hospital Harvard Medical School, Boston, MA, USA
Received 3 August 2014; accepted 8 December 2014



Neutron Capture Therapy & Neutrino detectors



Super kamiokande

50 kton water
13 000 PMT

A frontier development of neutrino detectors based on neutron detection

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n_TOF

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Motivation

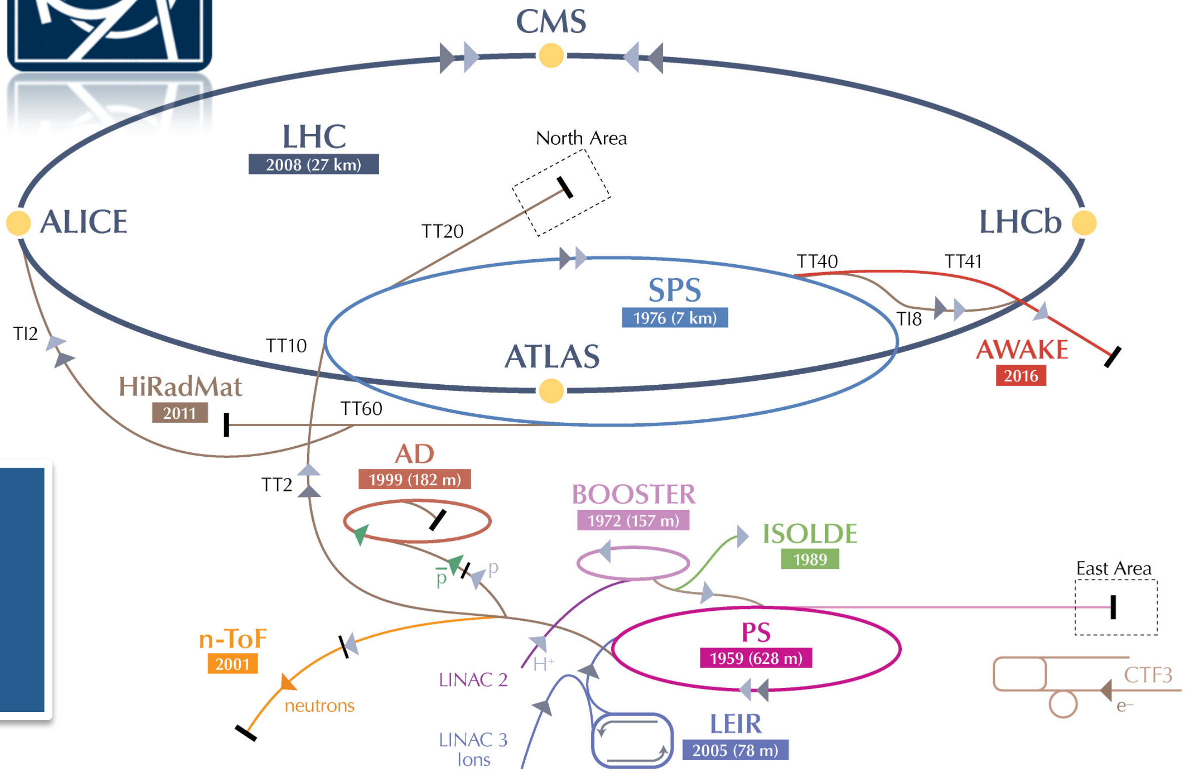
data in literature

Author or evaluation	Year	Thermal XS (kb) Gd-155	Deviation from ENDF	Thermal XS (kb) Gd-157	Deviation from ENDF
Møller (TOF)	1960	58.9 (5) *	- 3.4 %	254 (2) *	0.3 %
Ohno (TOF)	1968	61.9 (6) *	1.6 %	248 (4) *	- 2.1 %
Leinweber (TOF)	2006	60.2	- 1.2 %	226	**- 12 %**
Choi	2014	56.7 (21)	- 7.4 %	239 (6)	**- 6 %**
Mughabghab	2009	60.9 (2)	=	254.0 (8)	0.3 %
JENDL-4.0	2016	60.735	- 0.5 %	253.25	=
JEFF-3.3	2017	60.89	=	254.5	0.5 %
ENDF/B-VIII	2018	60.89	=	253.32	=

* = total cross section

The 155,157Gd(n,γ) measurement at n_TOF

The neutron time-of-flight facility at CERN



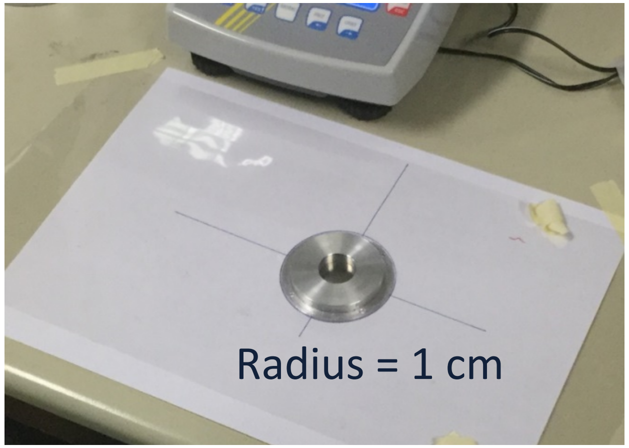
2 Experimental Areas:

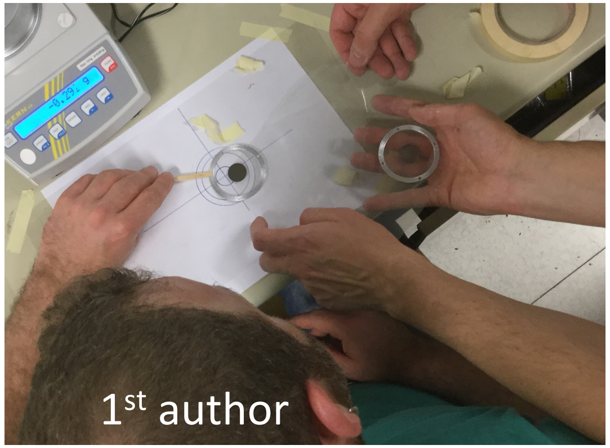
- EAR1 L = 184 m
- EAR2 L = 19 m

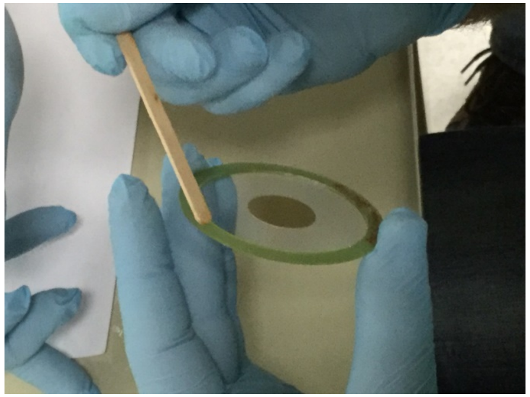
The 155,157Gd(n,γ) measurement at n_TOF

4 metallic samples form National Isotope Development Center (Oak Ridge - USA)

Isotope	Abundance %	% contamination of ^{155} or ^{157}Gd	% main contaminant	Weight mg	Areal density atoms/barn $\times 10^{-8}$
^{155}Gd	91.74 ± 0.18	1.14 ± 0.01	5.12 ± 0.18 ^{156}Gd	100.6 ± 0.1	12440 ± 40
^{155}Gd	91.74 ± 0.18	1.14 ± 0.01	5.12 ± 0.18 ^{156}Gd	10.0 ± 0.1	1236 ± 12
^{157}Gd	88.32 ± 0.01	0.29 ± 0.01	9.10 ± 0.01 ^{158}Gd	191.6 ± 0.1	23390 ± 60
^{157}Gd	88.32 ± 0.01	0.29 ± 0.01	9.10 ± 0.01 ^{158}Gd	4.7 ± 0.1	574 ± 12







Samples were sandwiched between two Mylar foils

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n_TOF

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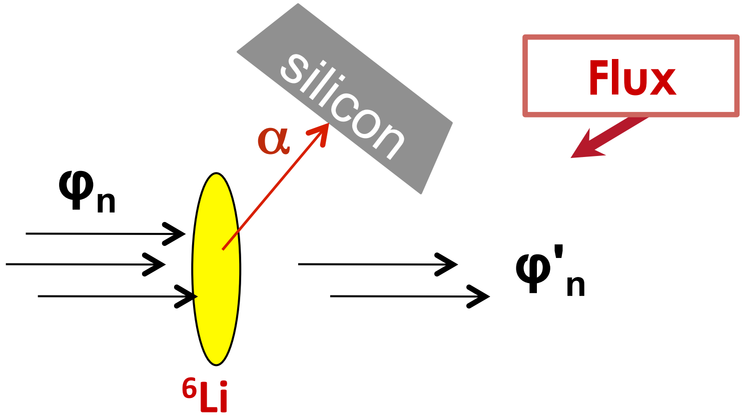
The 155,157Gd(n,γ) measurement at n_TOF

Measurement setup @ EAR1 (184 m): Flux + Capture



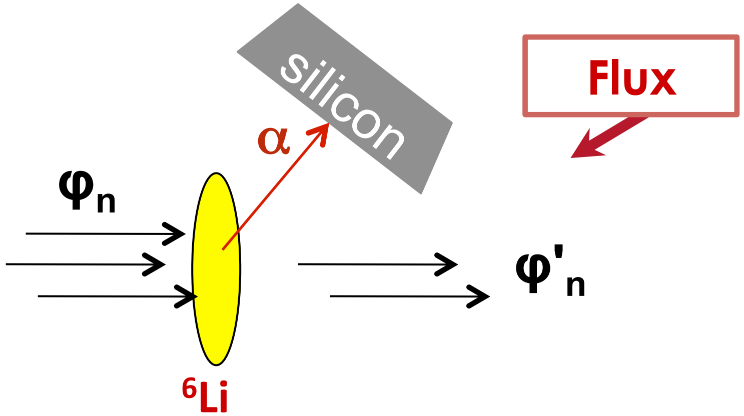
$$Y_n = \frac{C_n}{\varepsilon_n \Omega_n A_n \varphi_n}$$

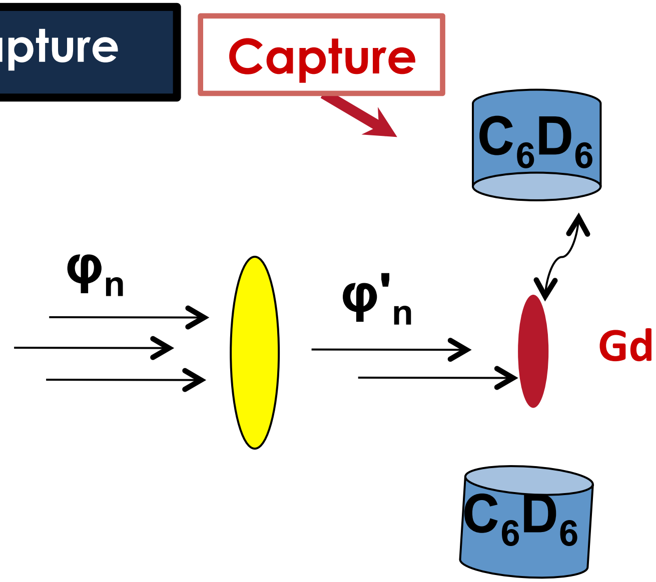
$$Y_n = (1 - e^{-n\sigma_{tot}}) \frac{\sigma_\alpha}{\sigma_{tot}}$$

$$\varphi_n = \frac{C_n}{\varepsilon_n \Omega_n A_n Y_n}$$

The 155,157Gd(n,γ) measurement at n_TOF

Measurement setup @ EAR1 (184 m): Flux + Capture





$$Y_n = \frac{C_n}{\varepsilon_n \Omega_n A_n \varphi_n}$$

$$Y_n = \left(1 - e^{-n\sigma_{tot}}\right)\frac{\sigma_\alpha}{\sigma_{tot}}$$

$$\varphi_n = \frac{C_n}{\varepsilon_n \Omega_n A_n Y_n}$$

$$Y_C = \frac{\varepsilon_n \Omega_n A_n}{\varepsilon_C \Omega_C A_C} \times \frac{C_W}{C_n} \times \frac{\left(1 - e^{-n\sigma_{tot}}\right)\frac{\sigma_\alpha}{\sigma_{tot}}}{e^{-n\sigma_{tot}}}$$

$$Y_C = N \times \frac{C_W}{C_n} \times \frac{Y_n}{e^{-n\sigma_{tot}}}$$

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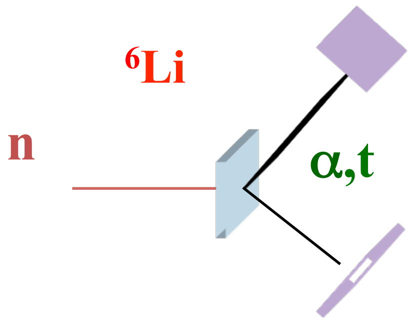
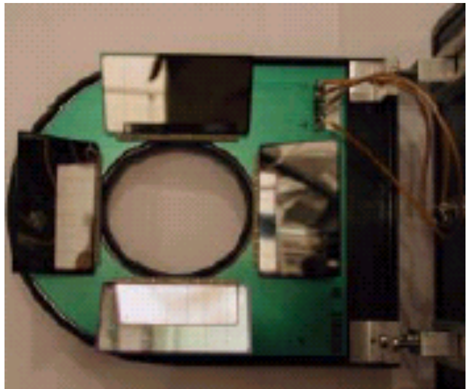
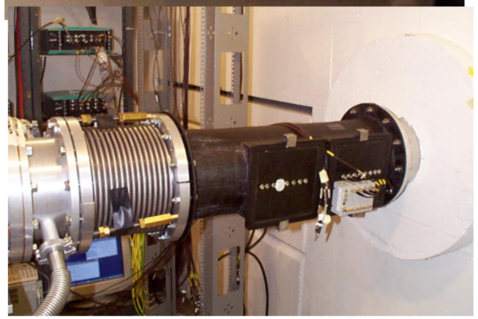
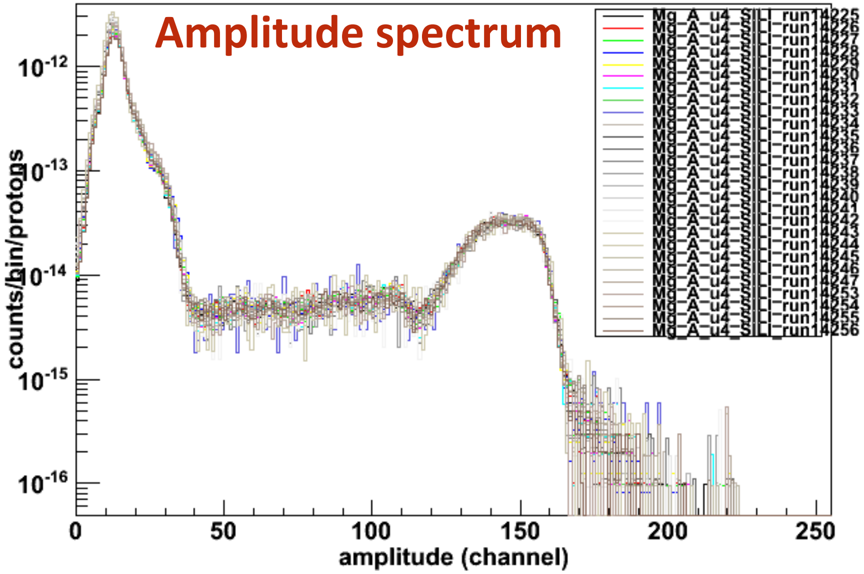
The 155,157Gd(n,γ) measurement at n_TOF

FLUX: Silicon Monitor

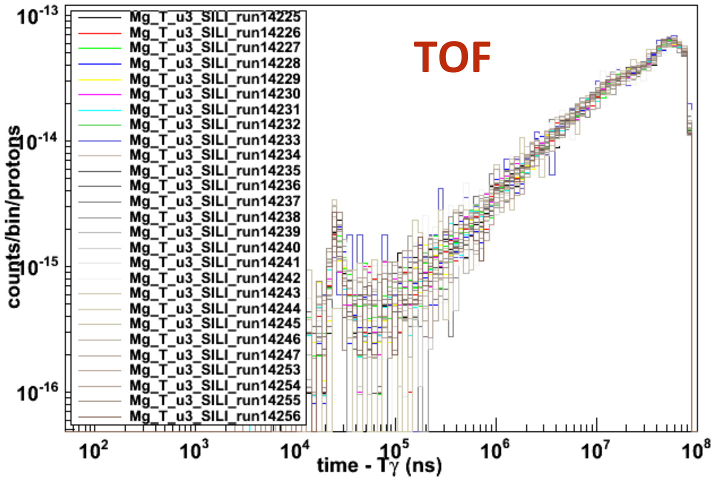








^{6}Li(n,t)α



Runs of about ~ 10 hours
→
detector stability check

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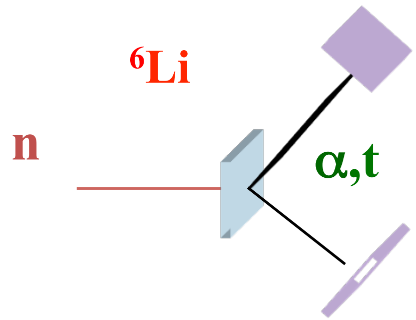
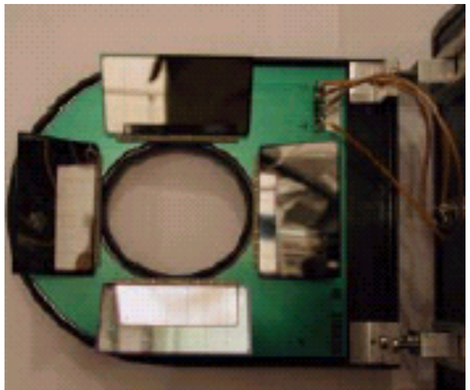
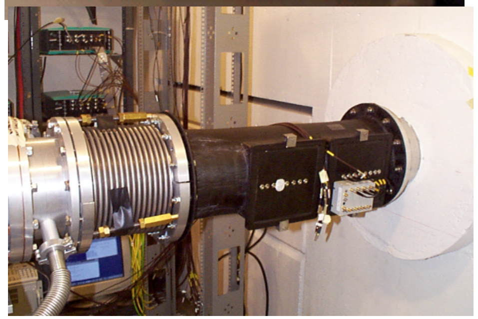
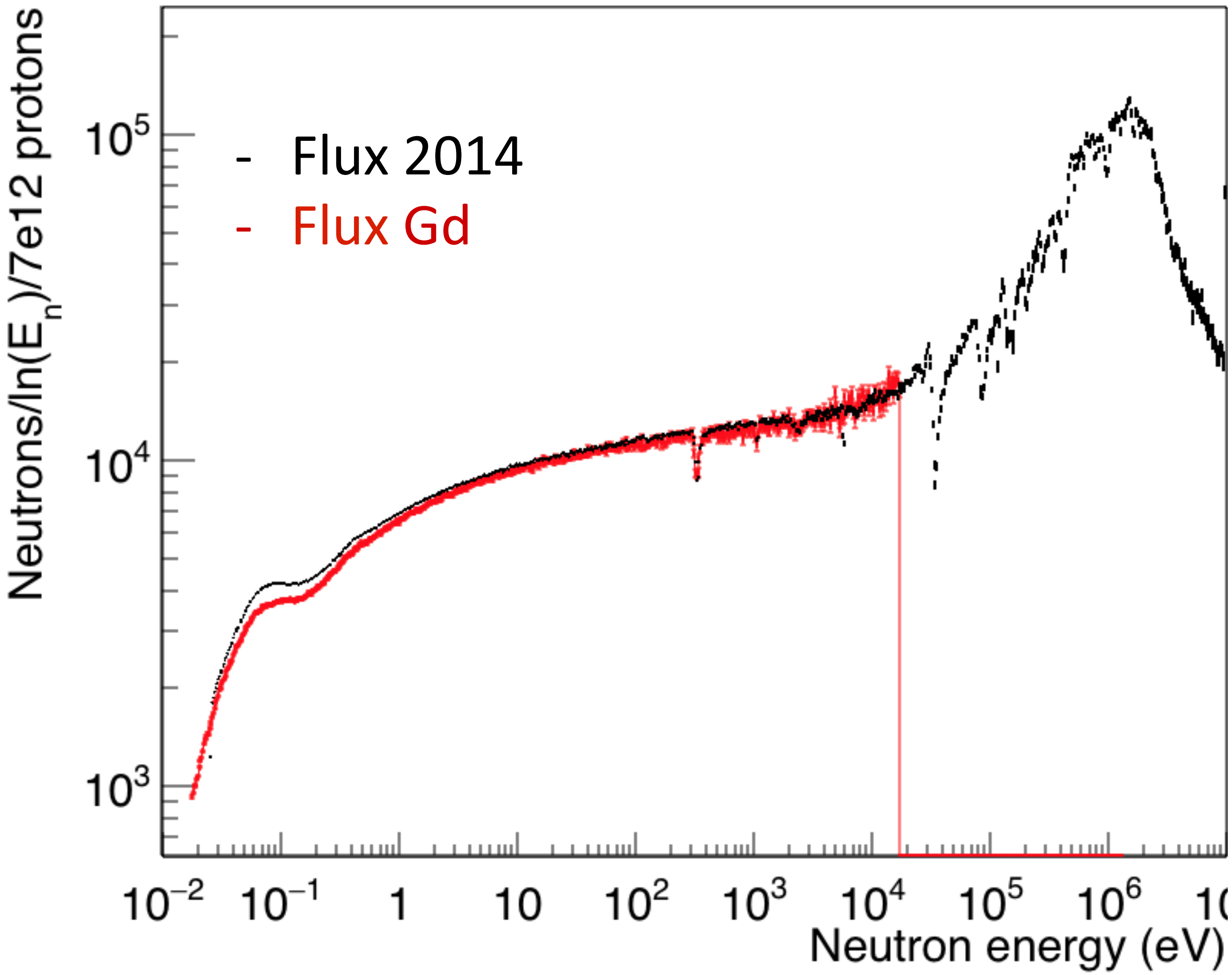
The 155,157Gd(n,γ) measurement at n_TOF

FLUX: Silicon Monitor









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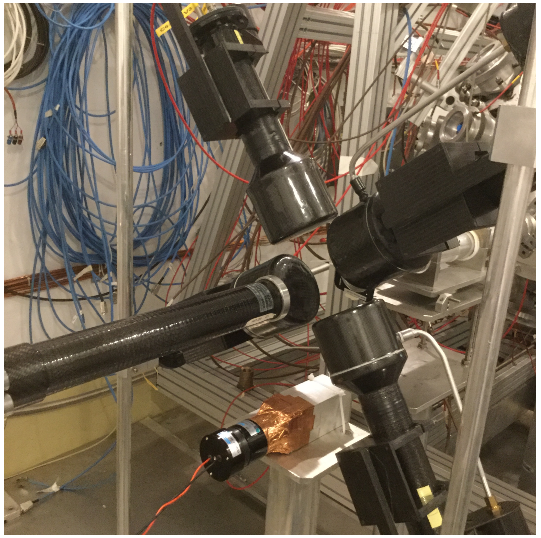
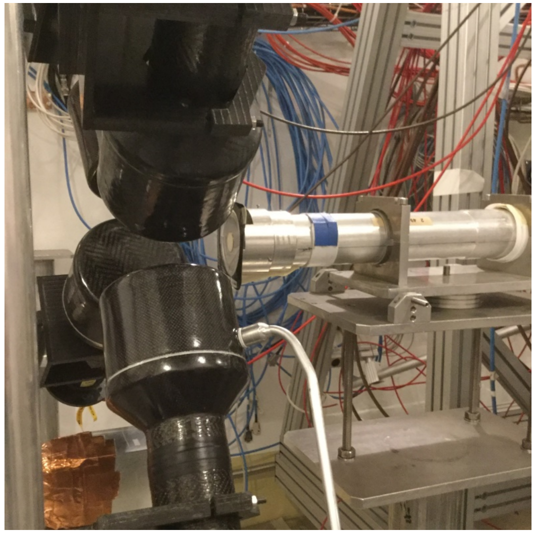
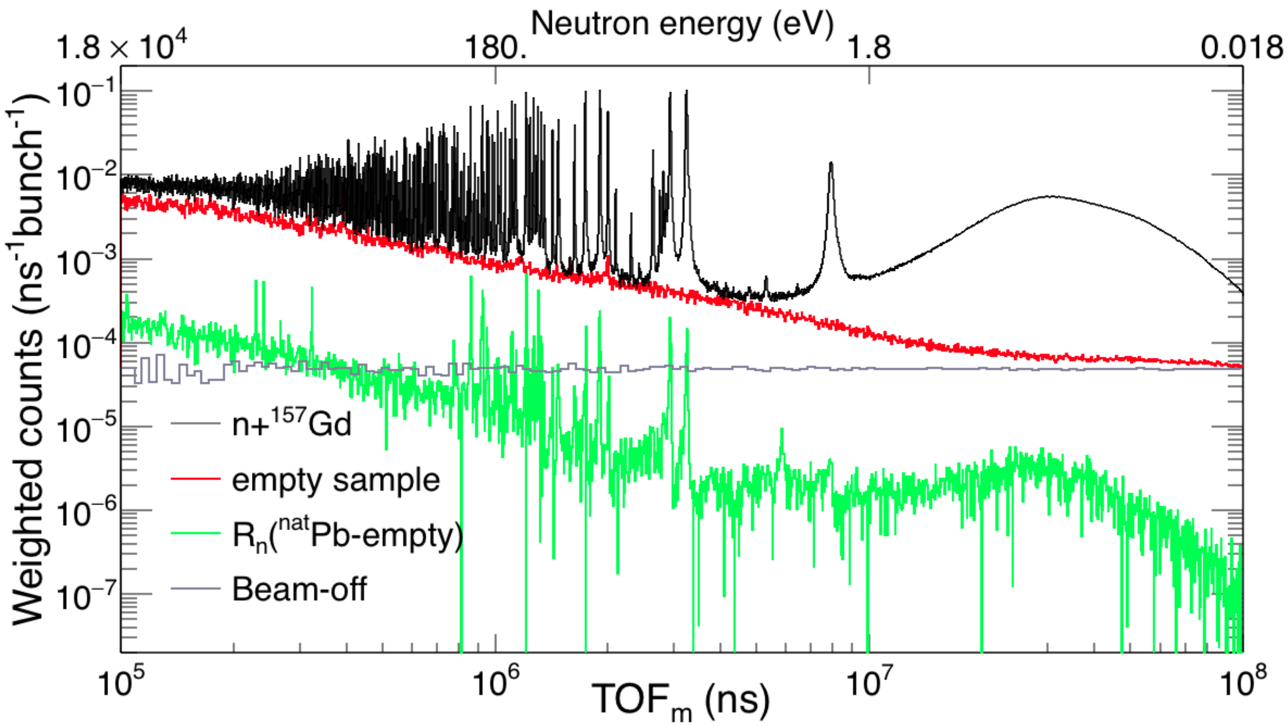
The 155,157Gd(n,γ) measurement at n_TOF

CAPTURE: 4 C_6D_6







RED = empty sample

GREEN = neutron-induced backgrond

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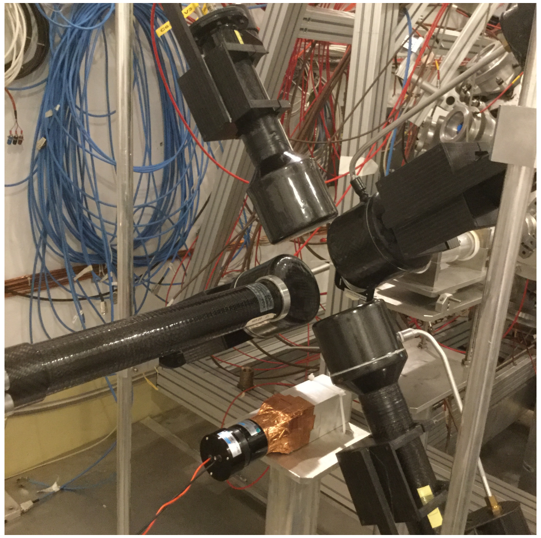
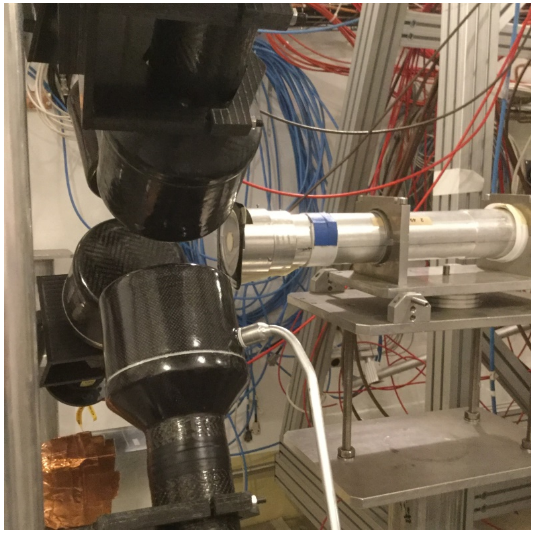
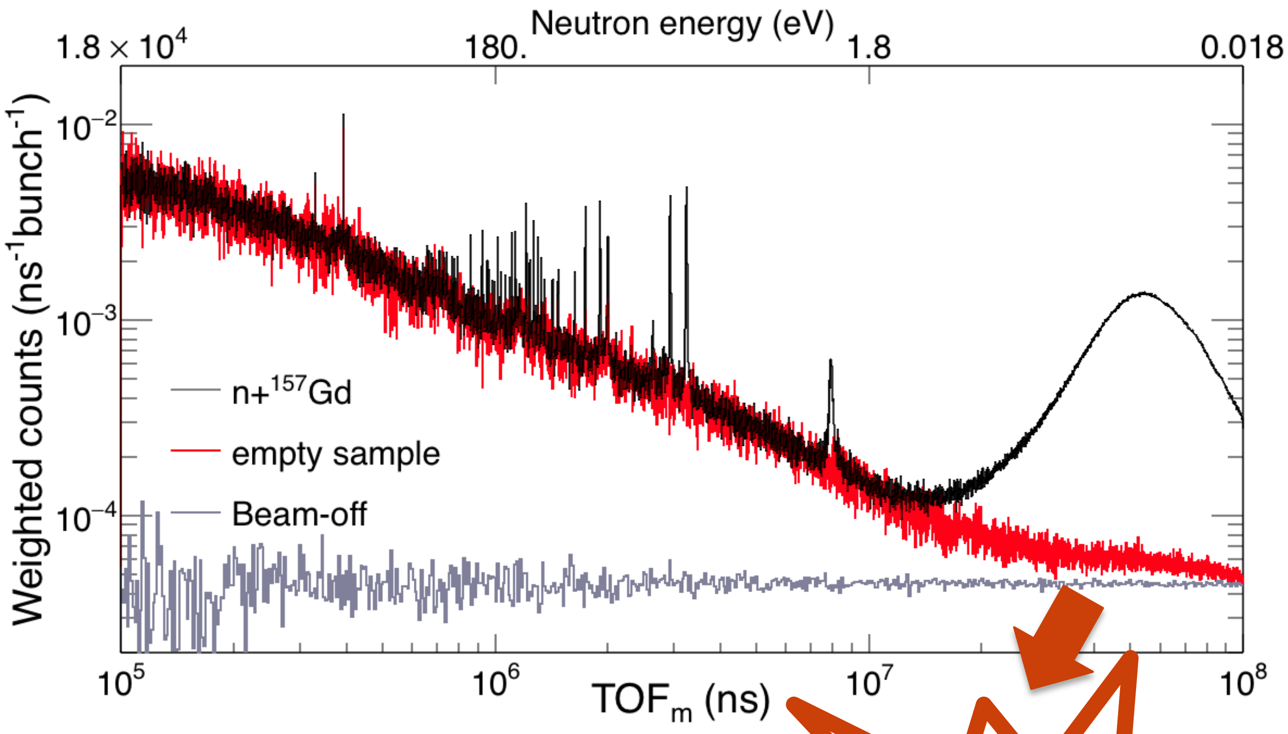
The 155,157Gd(n,γ) measurement at n_TOF

CAPTURE: 4 C_6D_6







1% uncertainty related to background subtraction

Background < 10%

Analysis

Energy calibration & Weighting Function

Results within 1.5%

- **Calibrations**
  - Linear
  - Quadratic

- **Weighting Function (γ-ray transport in the sample)**
  - **Homogeneous** spatial distribution, threshold **150 keV**
  - Homogeneous spatial distribution, threshold **200 keV**
  - **Exponential shape**, threshold 150 keV (**7 different $n^*\sigma_{tot}$**)
  - Exponential shape, threshold 200 keV (**7 different $n^*\sigma_{tot}$**)

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n_TOF

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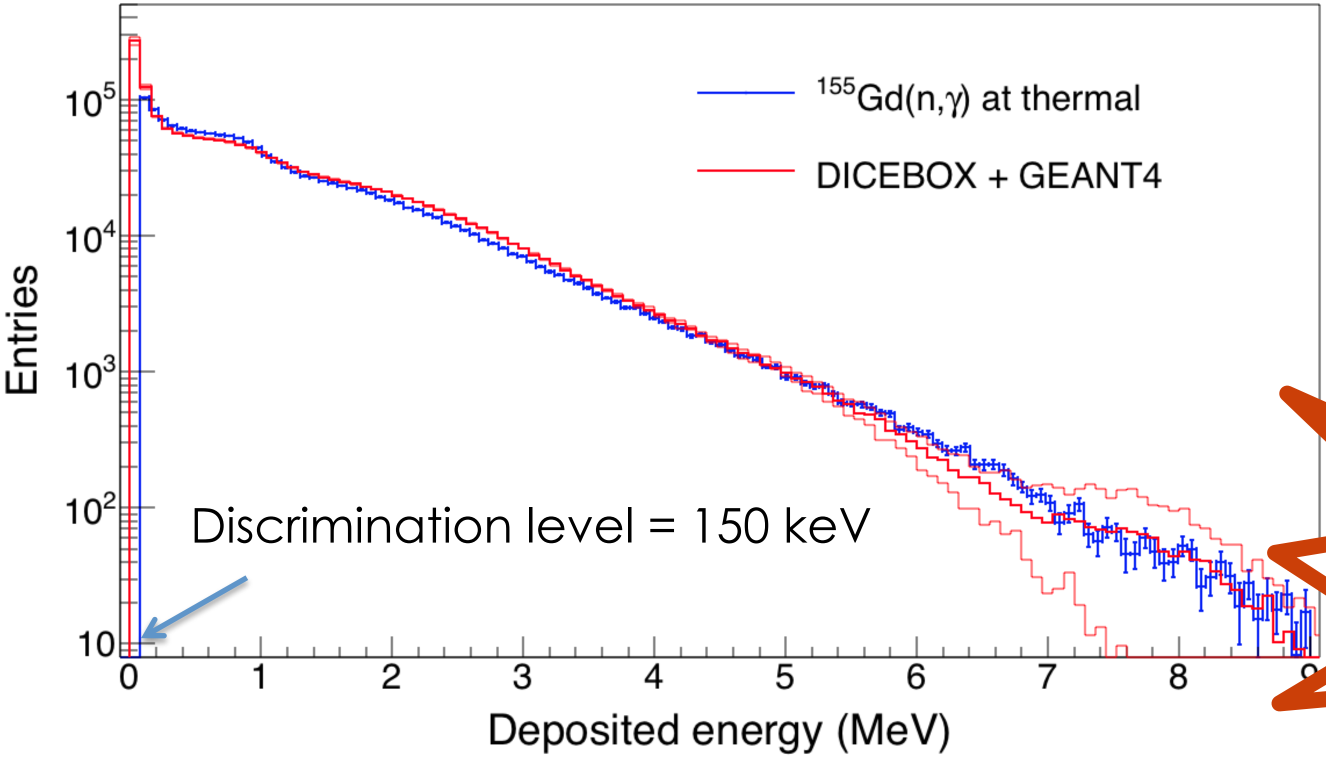
Analysis

Normalization – Au(n,γ) @ 4.9 eV



Missed counts below threshold

	γ-rays	e^-
^{197}Au	1.0 %	0.8 %
^{156}Gd	0.4 %	0.2 %
^{158}Gd	0.4%	0.2%

Normalization Uncertainty 1.5%

^{156}Gd → S_n= 8.54 MeV
^{158}Gd → S_n= 7.94 MeV
^{198}Au → S_n= 6.51 MeV

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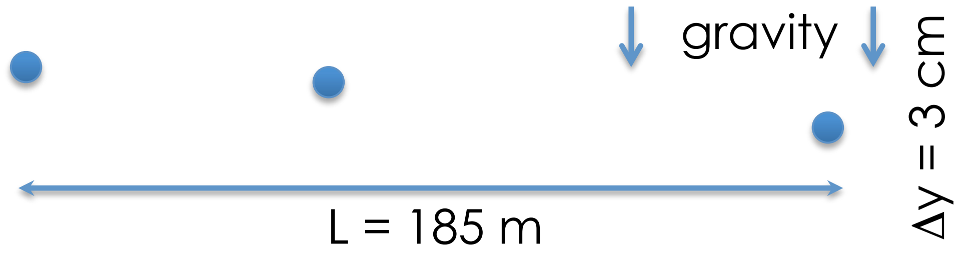
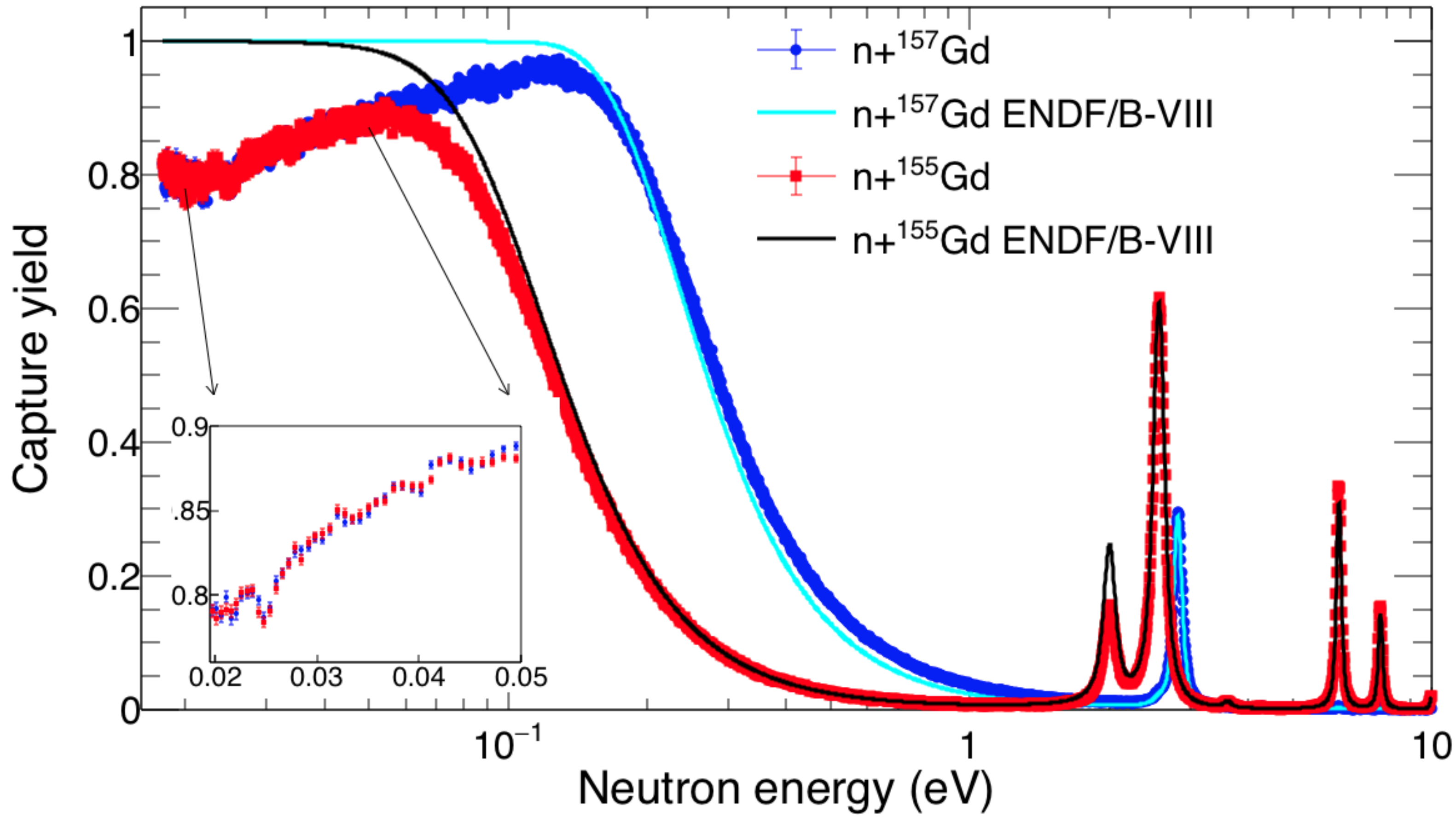
Analysis

Beam Interception factor





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nTOF

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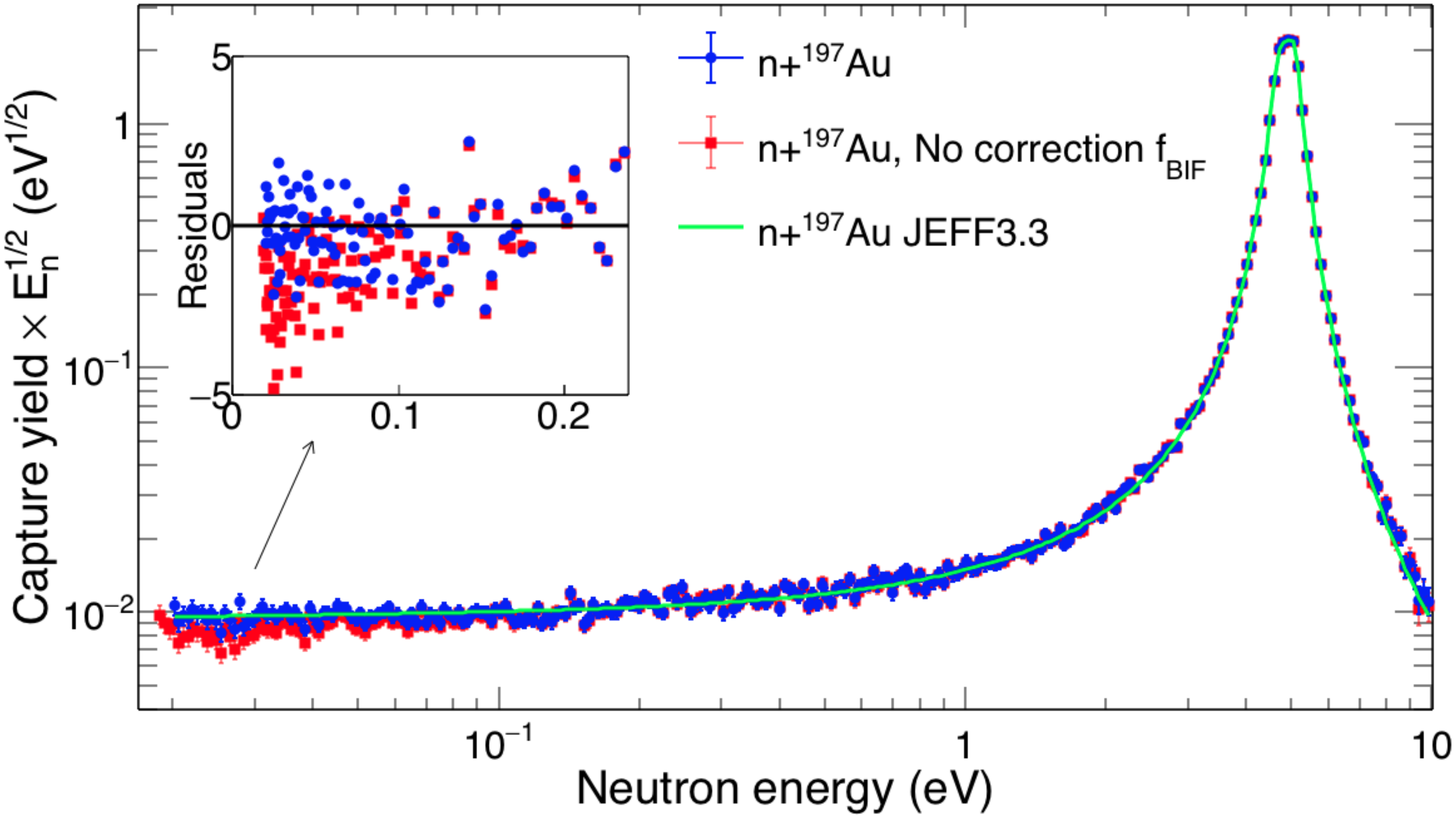
Analysis

Beam Interception factor

Au standard within 1.5%



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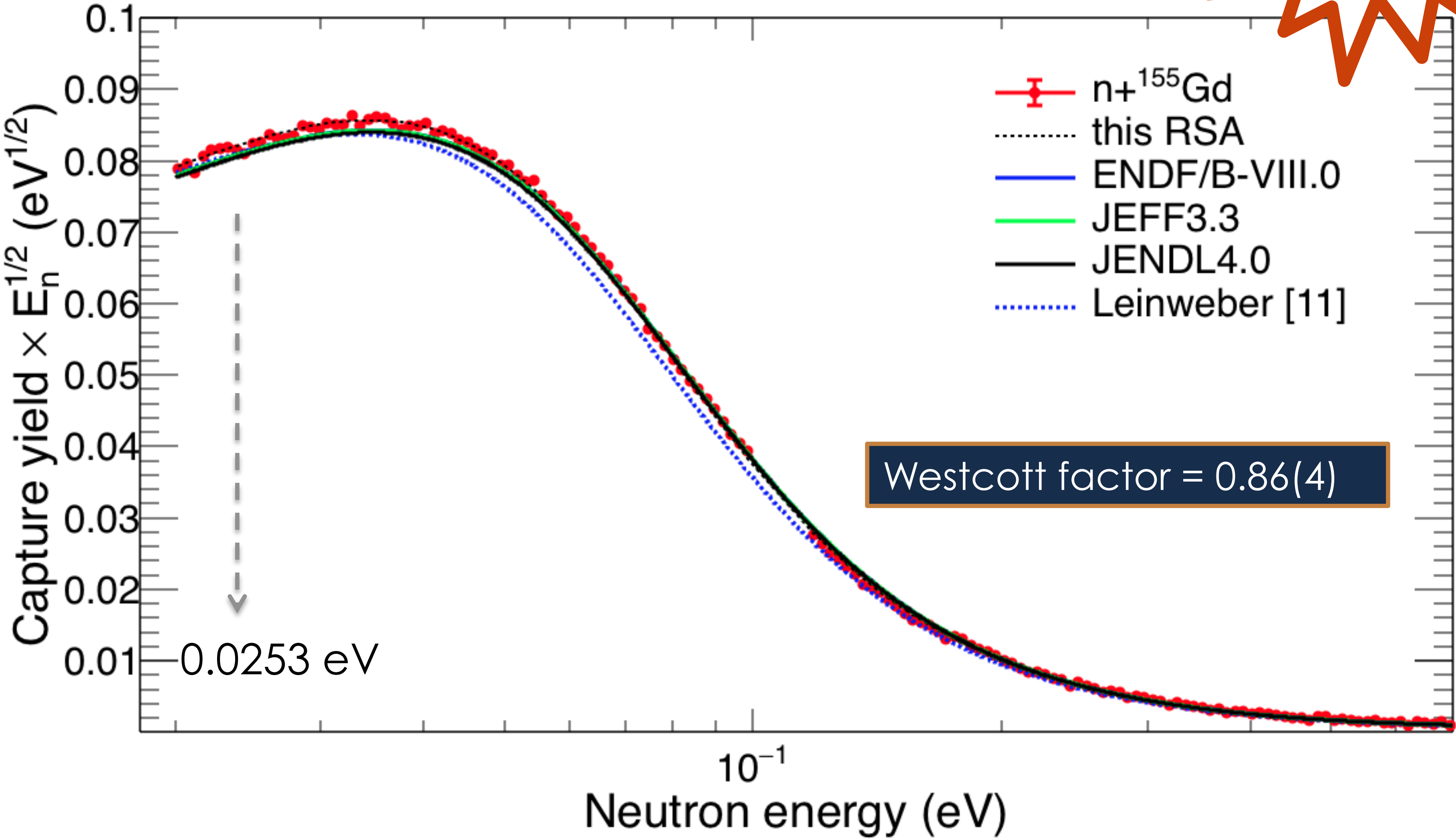
Analysis: capture yield



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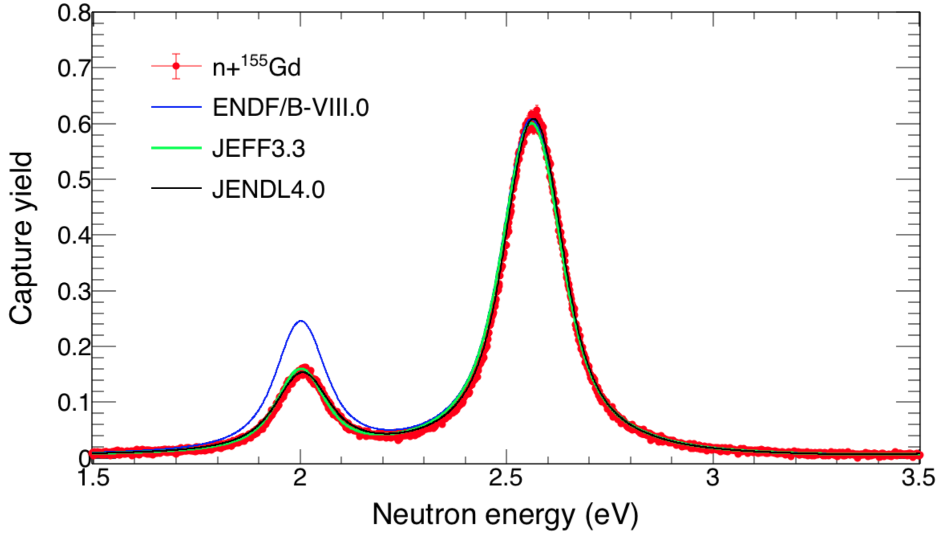
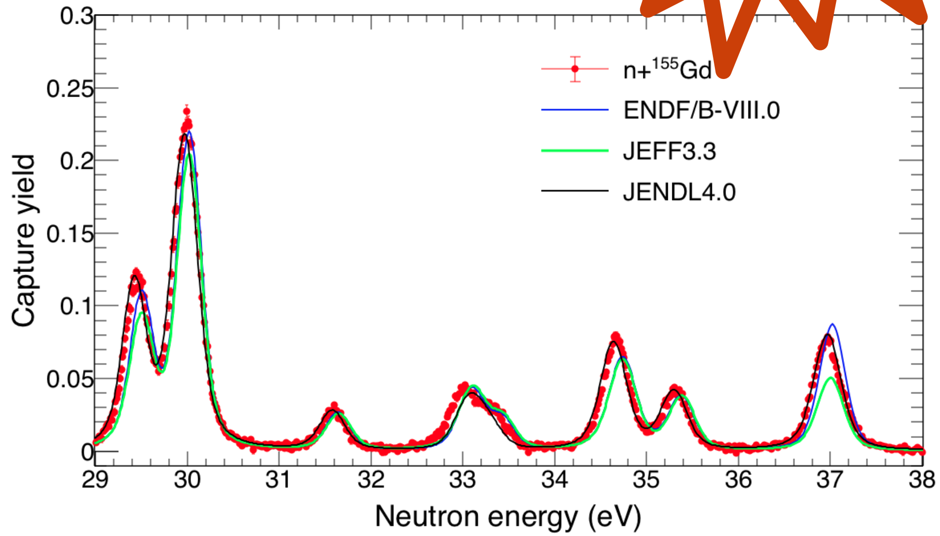
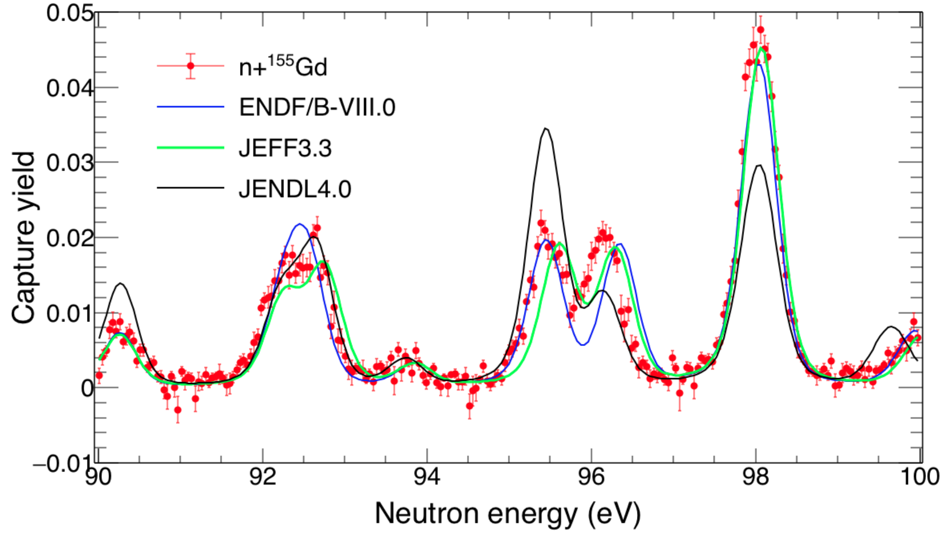
Analysis: capture yield

^{155}Gd







	Resonance kernel ratios
ENDF/B-VIII	0. 98
JEFF-3.3	0.98
Baramsai	1.02
Leinweber	0.92
JENDL-4.0	0.92

n_TOF

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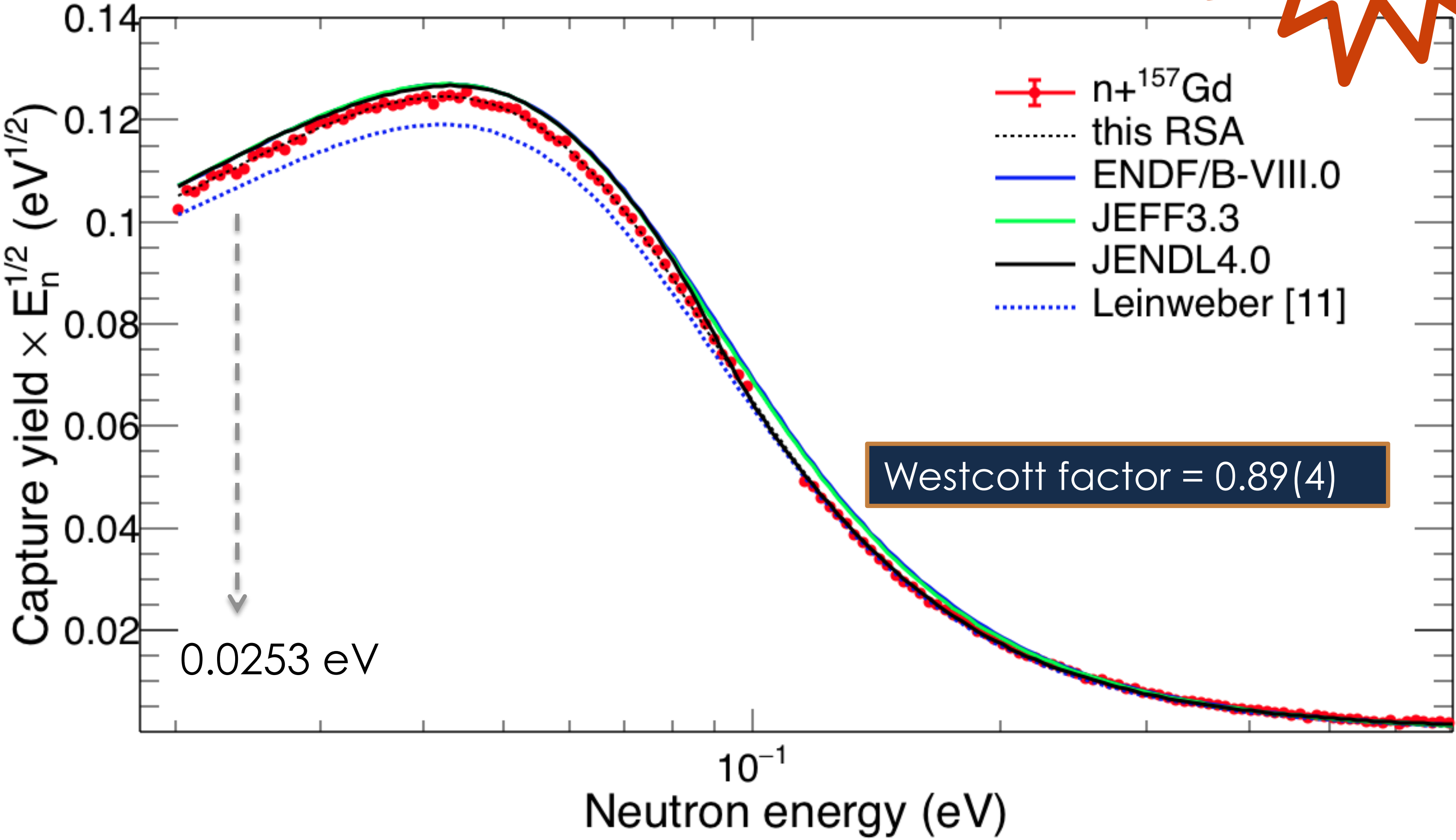
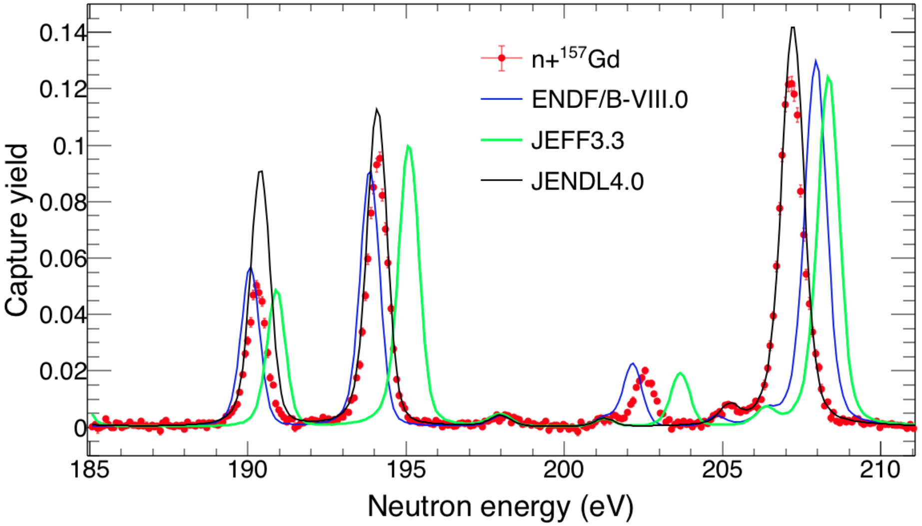
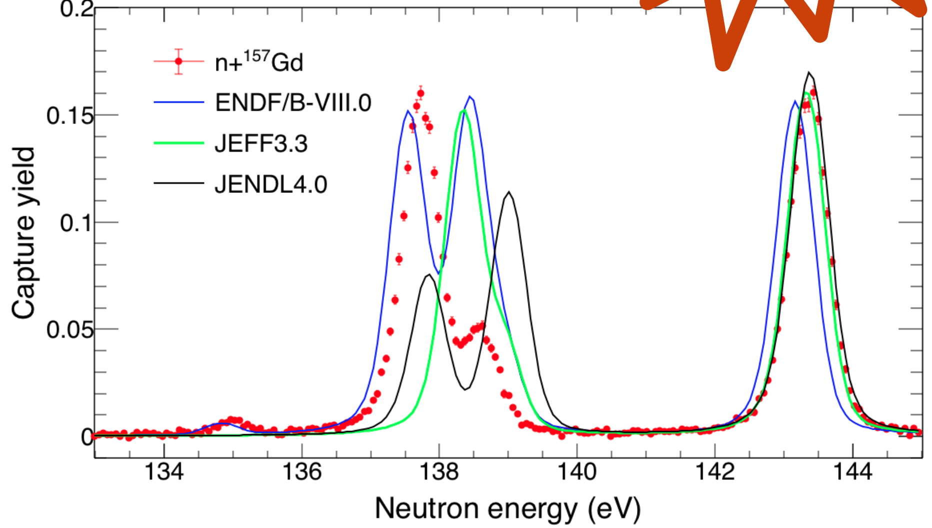
Analysis: capture yield

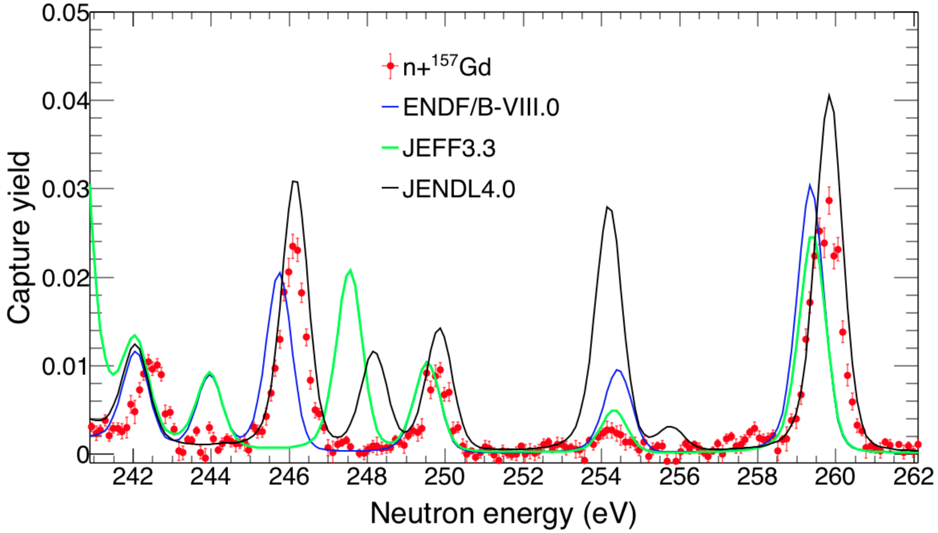


^{157}Gd

Analysis: capture yield







	Resonance kernel ratios
ENDF/B-VIII	0. 98
JEFF-3.3	0.98
JENDL-4.0	0.87
Leinweber	0.87

nTOF

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Conclusions

- ^{155,157}Gd Capture yields in **EXFOR** (entry #23400)

- Improved Resonances parameters

- Possibility to extend evaluation above 180 and 300 eV

- Statistical properties of neutron resonances

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Eur. Phys. J. A (2019) **55**: 9 DOI 10.1140/epja/i2019-12692-7

Cross section measurements of ^{155,157}Gd(n, γ) induced by thermal and epithermal neutrons

n_TOF Collaboration

Conclusions

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Møller (TOF)	1960	58.9 (5) *	- 3.4 %	254 (2) *	0.3 %
Ohno (TOF)	1968	61.9 (6) *	1.6 %	248 (4) *	- 2.1 %
Leinweber (TOF)	2006	60.2	- 1.2 %	226	**- 12 %**
Choi	2014	56.7 (21)	- 7.4 %	239 (6)	**- 6 %**
Mughabghab	2009	60.9 (2)	=	254.0 (8)	0.3 %
JENDL-4.0	2016	60.735	- 0.5 %	253.25	=
JEFF-3.3	2017	60.89	=	254.5	0.5 %
ENDF/B-VIII	2018	60.89	=	253.32	=

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Conclusions

Author or evaluation	Year	Thermal XS (kb) Gd-155	Deviation from ENDF	Thermal XS (kb) Gd-157	Deviation from ENDF
Møller (TOF)	1960	58.9 (5) *	- 3.4 %	254 (2) *	0.3 %
Ohno (TOF)	1968	61.9 (6) *	1.6 %	248 (4) *	- 2.1 %
Leinweber (TOF)	2006	60.2	- 1.2 %	226	**- 12 %**
Choi	2014	56.7 (21)	- 7.4 %	239 (6)	**- 6 %**
Mughabghab	2009	60.9 (2)	=	254.0 (8)	0.3 %
JENDL-4.0	2016	60.735	- 0.5 %	253.25	=
JEFF-3.3	2017	60.89	=	254.5	0.5 %
ENDF/B-VIII	2018	60.89	=	253.32	=
n_TOF	**2019**	**62.2 (22)**	**+ 2.1 %**	**239.8 (84)**	**- 5.6 %**

The n_TOF Collaboration

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Backup

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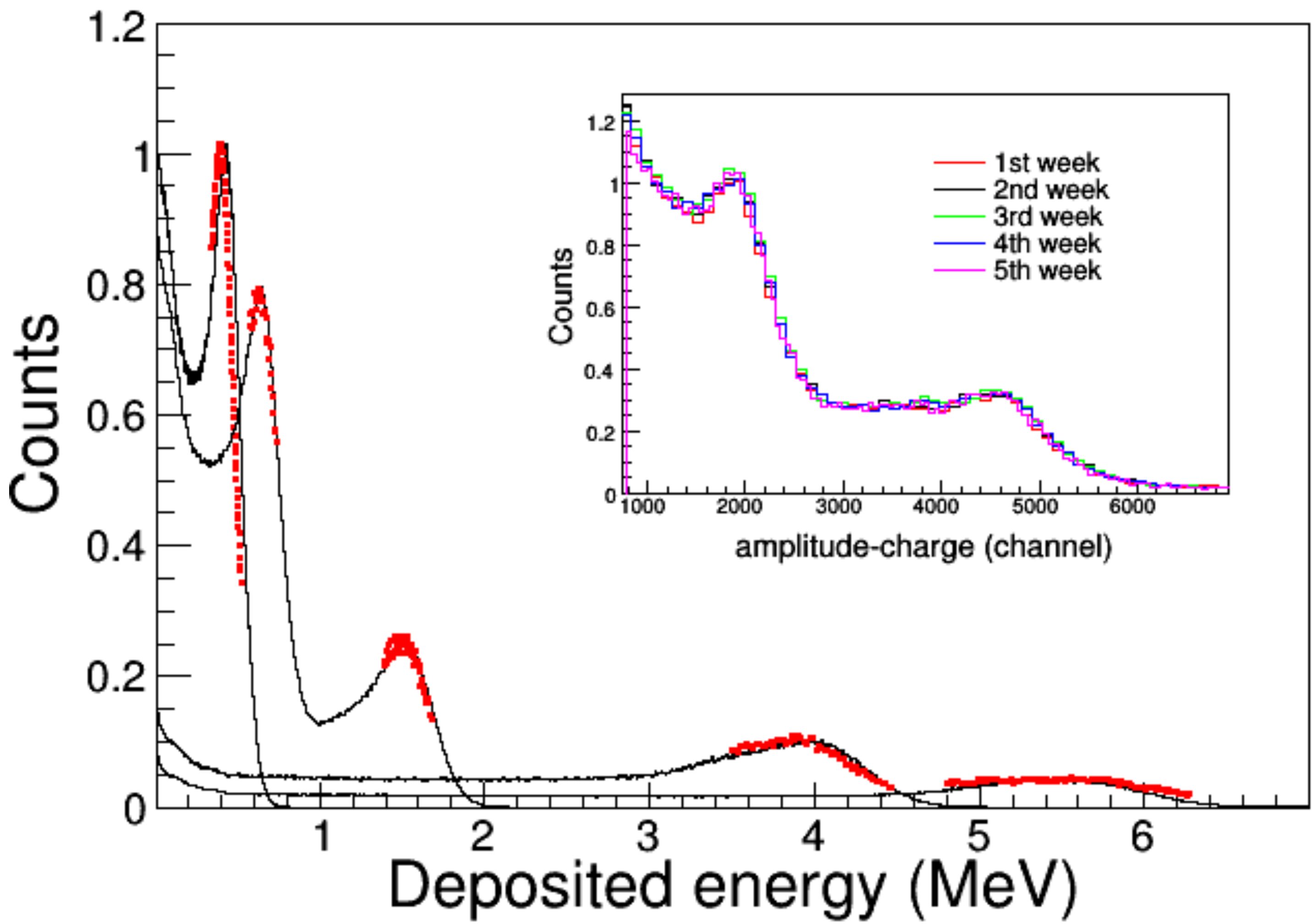
Calibration



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Uncertainties

Source of uncertainty	^{155}Gd(n, γ) near thermal	^{155}Gd(n, γ) resonance region	^{157}Gd(n, γ) near thermal	^{157}Gd(n, γ) resonance region
Normalization	1.2%	1.2%	1.2%	1.2%
PHWT	1.5%	1.5%	1.5%	1.5%
Background	1.4%	$\approx$ 1%	1.0%	$\approx$ 1%
BIF	1.5%		1.5%	
Flux	1.0%	1.0%	1.0%	1.0%
Sample mass	1.0%	$<$ 0.2%	2.1%	$<$ 0.2%
Temperature		1%		1%
Total	3.2%	2.6%	3.5%	2.6%

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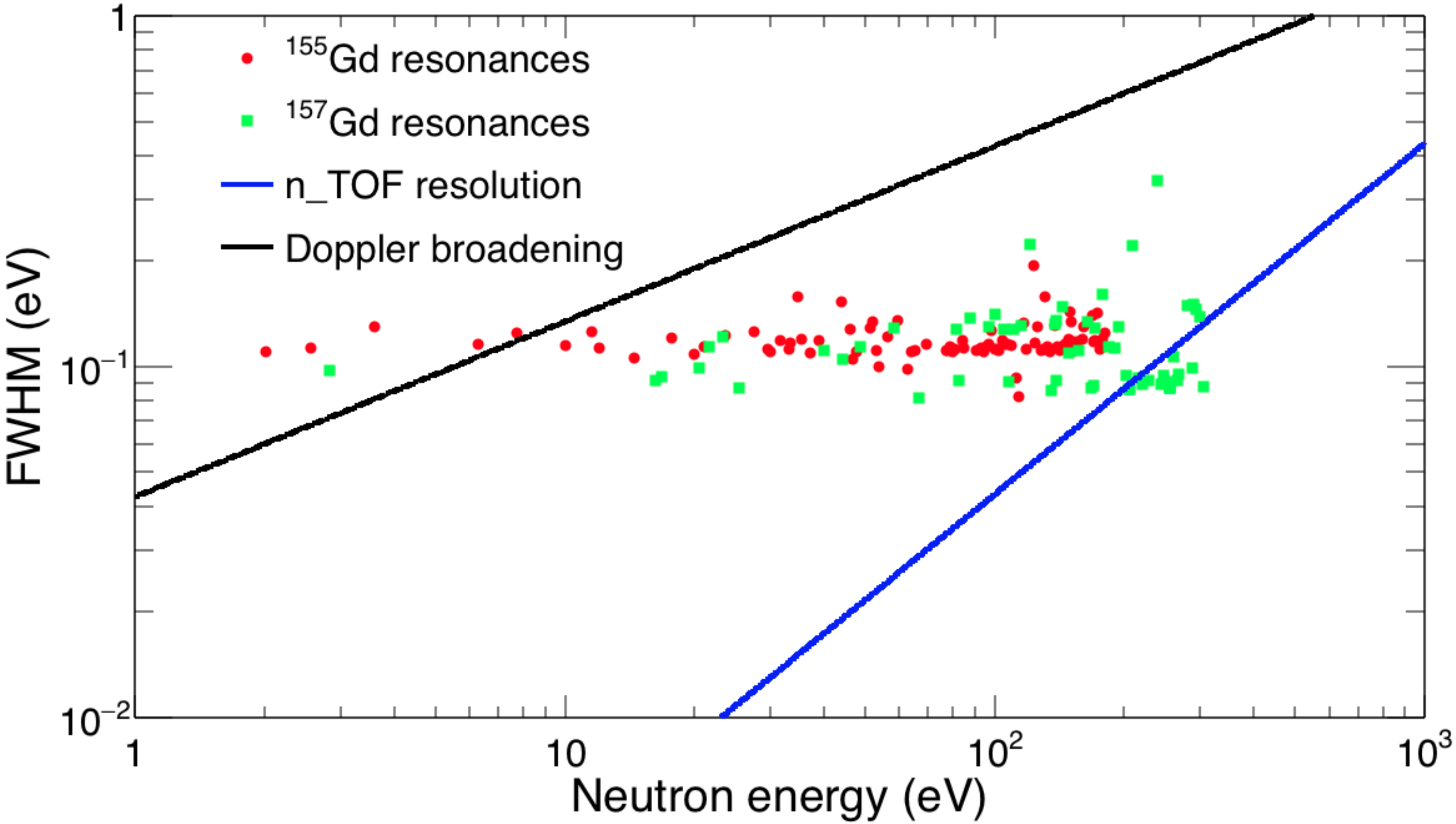
Broadening



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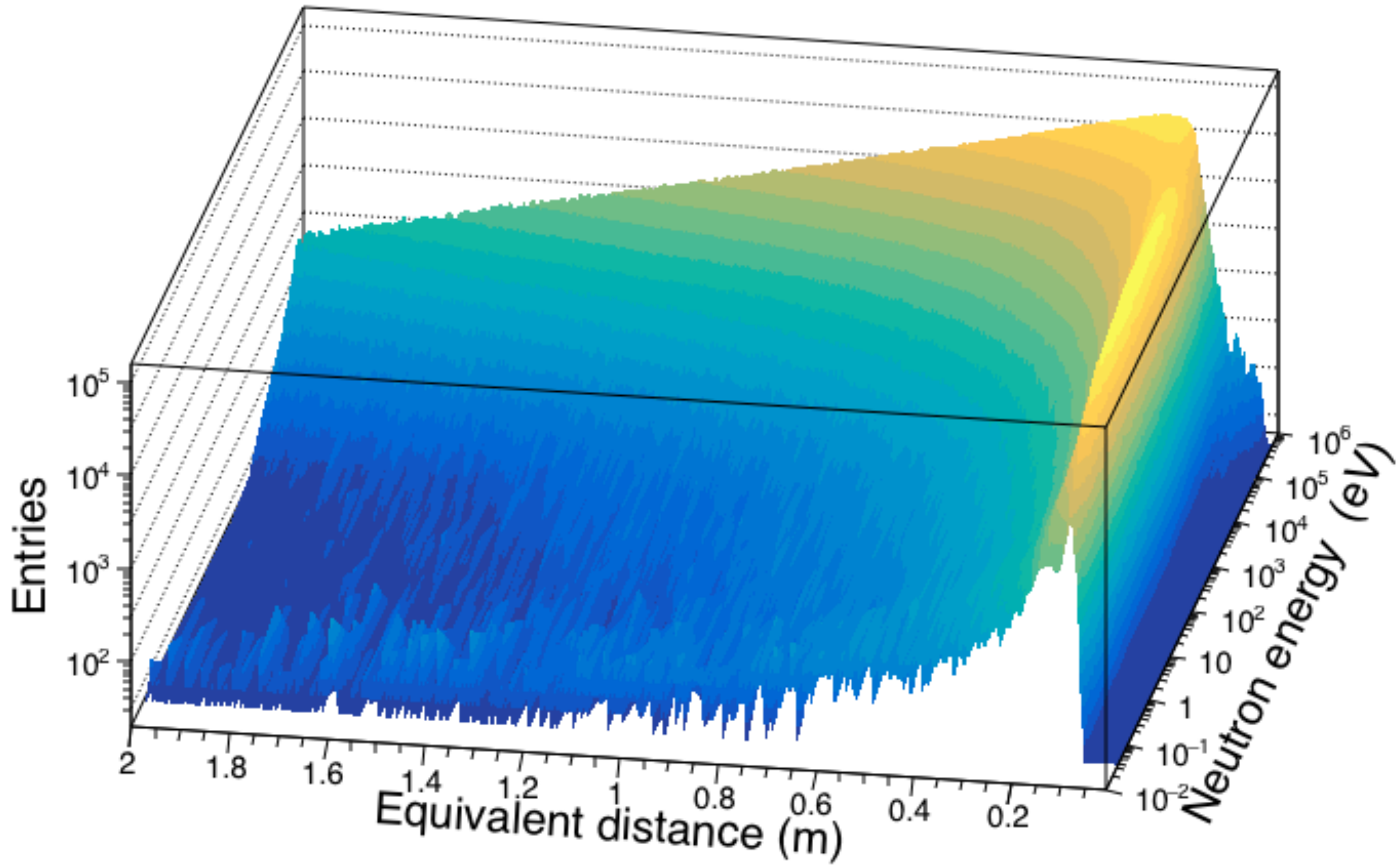
n_TOF Response function



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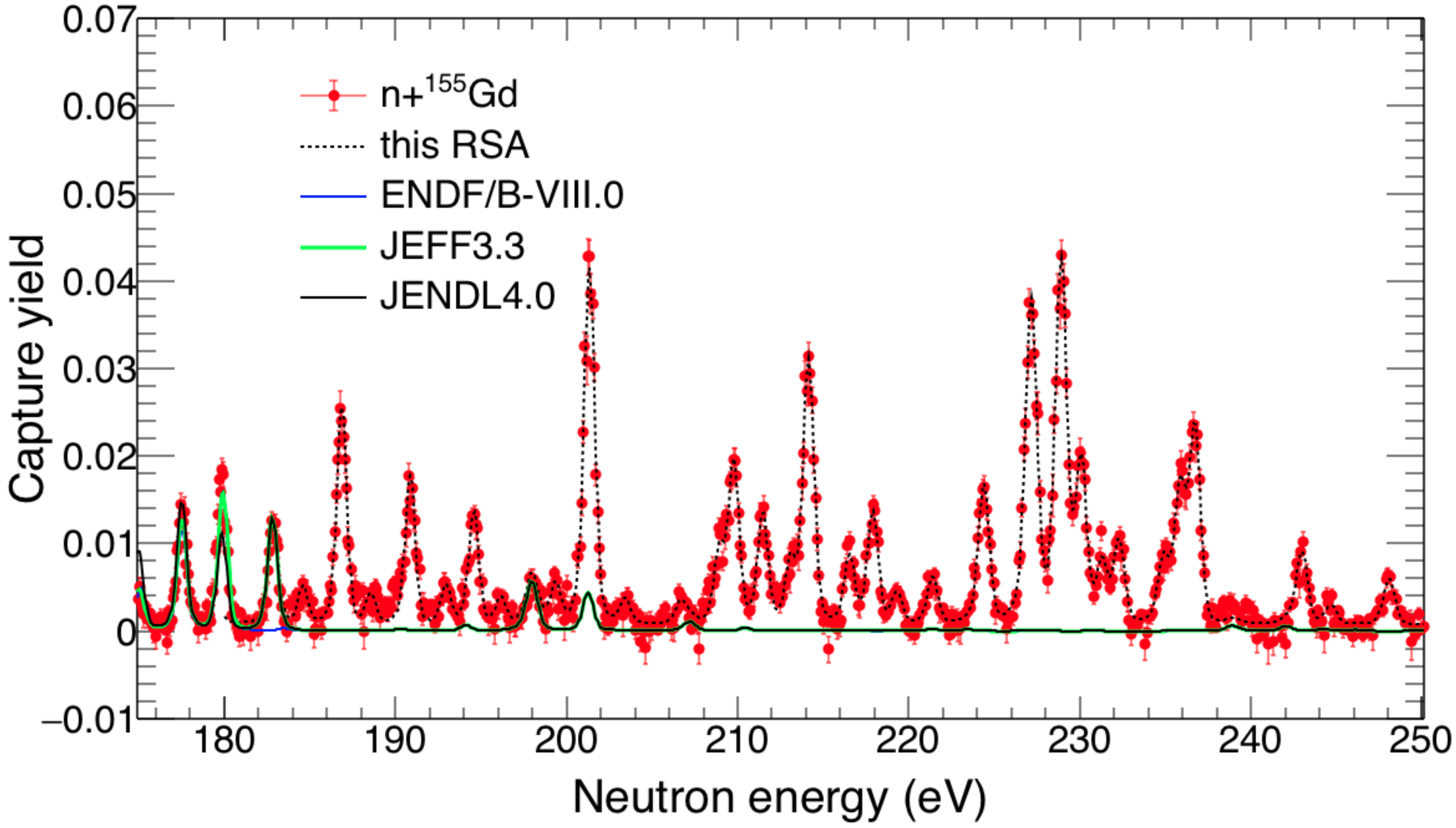
RSA



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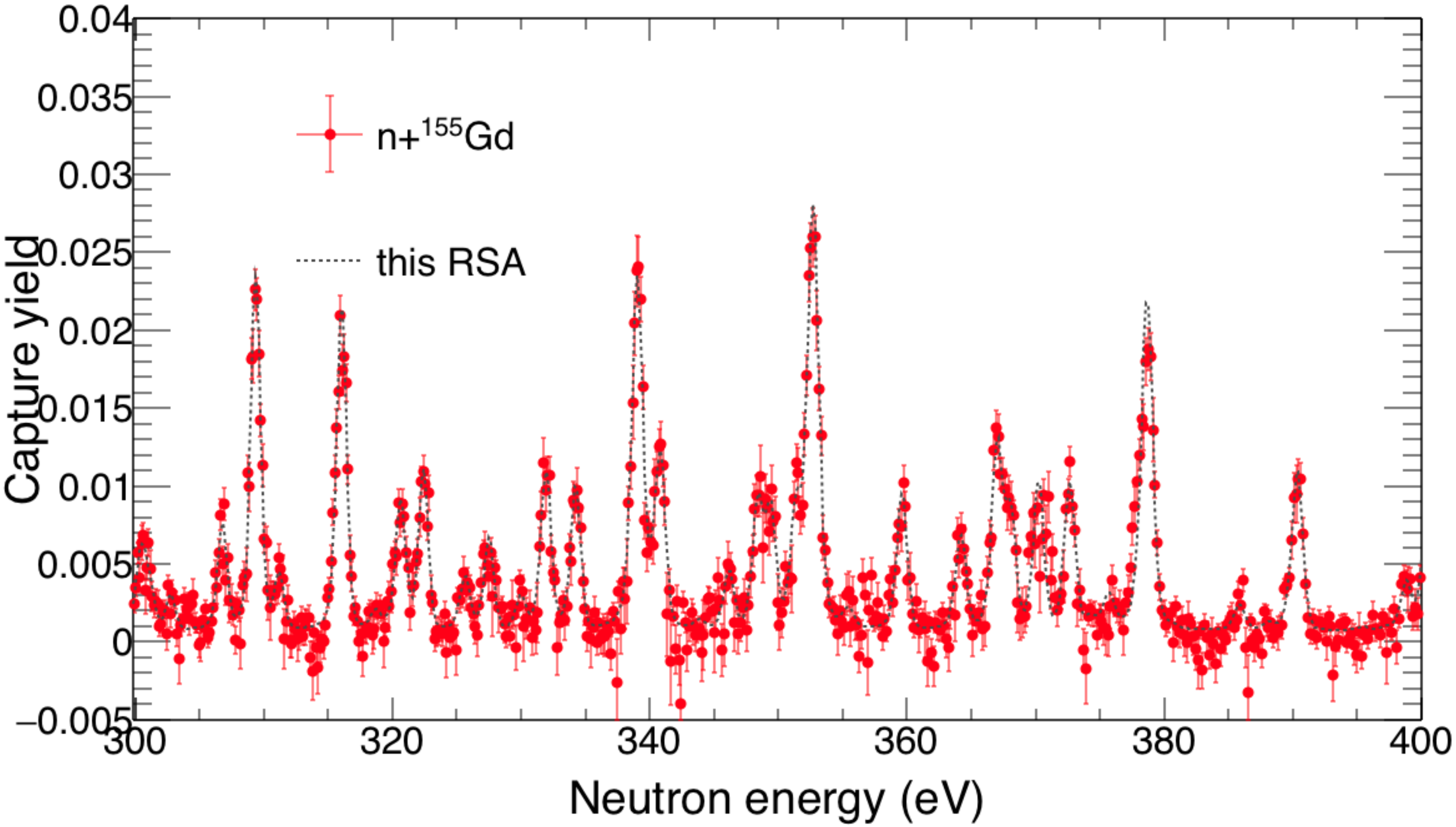
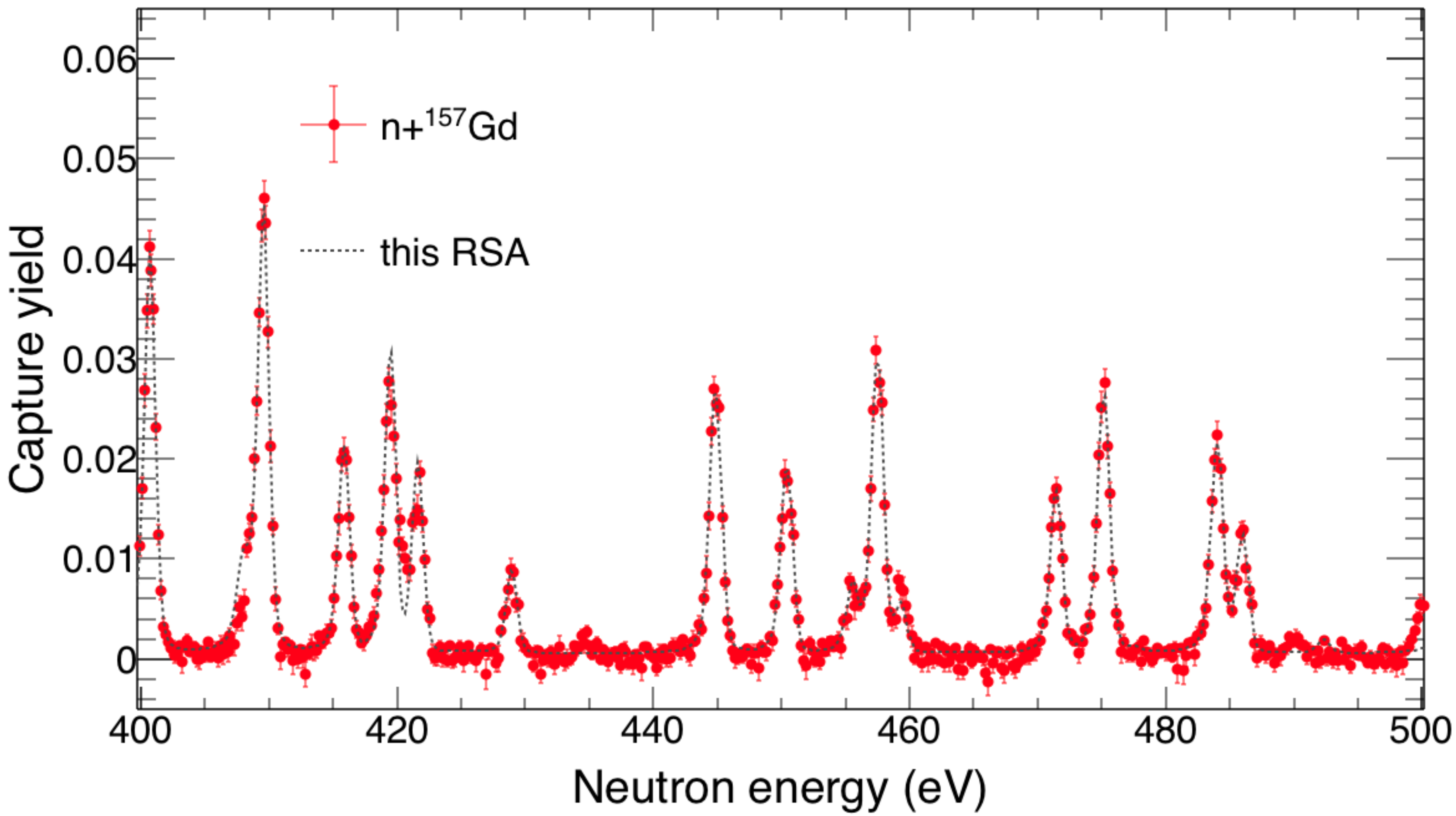
RSA



n+155Gd
this RSA
Capture yield
Neutron energy (eV)
0.04
0.035
0.03
0.025
0.02
0.015
0.01
0.005
0
−0.005
300
320
340
360
380
400

Backup

RSA



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n_TOF

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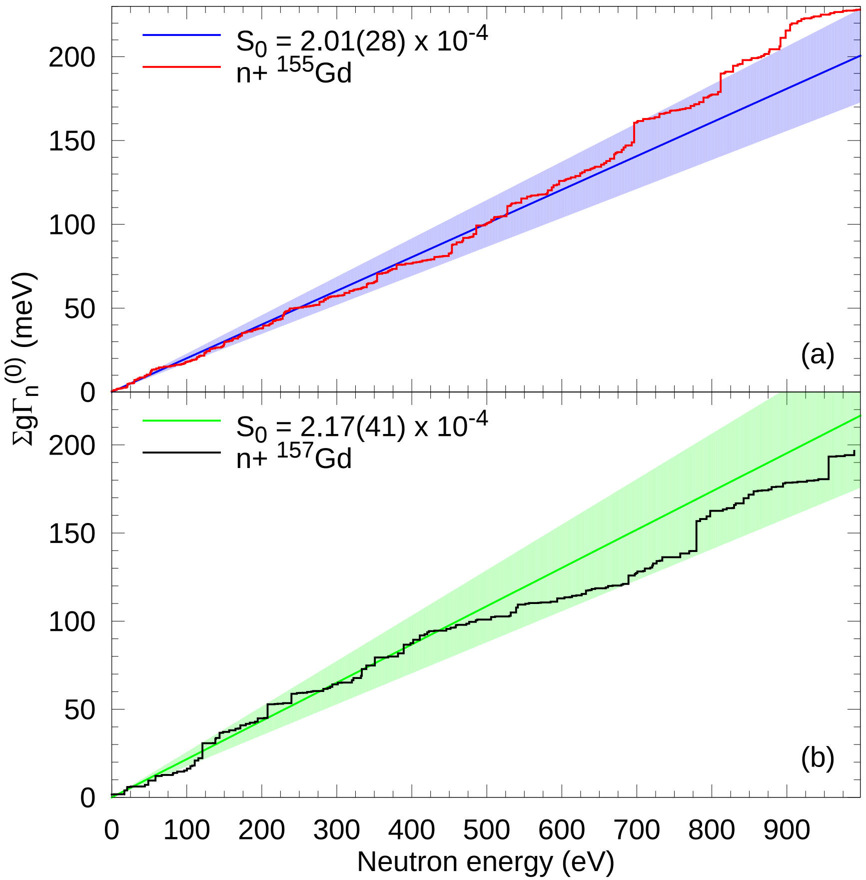
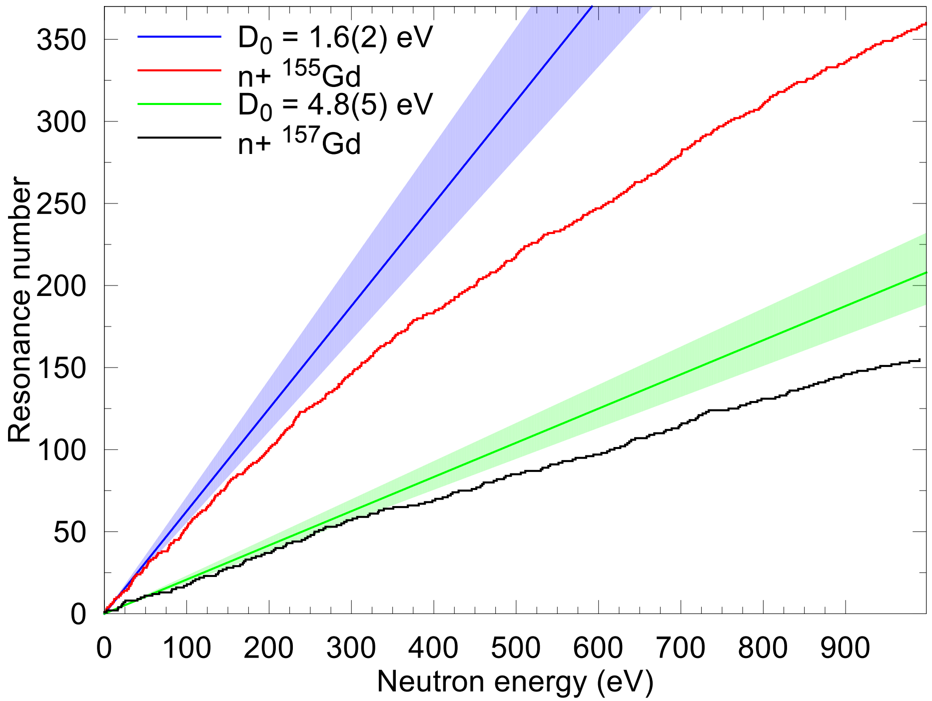
Statistical properties of neutron resonance





	Γ_γ (meV)
^{155}Gd+n	106.8 (14)
^{157}Gd+n	101.2 (20)

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