European
Commission

Actinides on the high priority request list & Experiences with modeling (n,n’g) reactions using TALYS

Arjan Plompen

EC-JRC-IRMM, SN3S Unit

European Commission, Joint Research Centre,

Institute for Reference Materials and Measurements

Standards for Nuclear Safety, Safeguards and Security

www.jrc.ec.europa.eu

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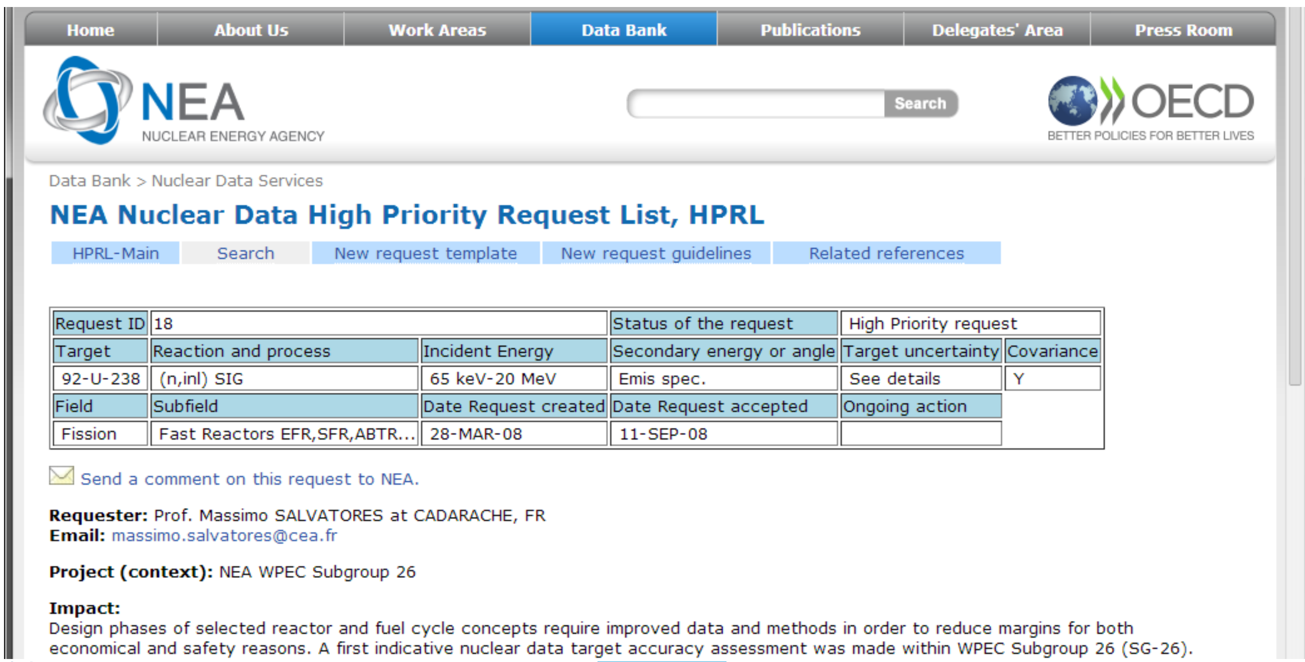
Research
Centre

Actinides on the HPRL

High Priority Request List for nuclear data was renewed in 2004
Current entries 2005-2008
www.oecd-nea.org/dbdata/hprl



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NEA NUCLEAR ENERGY AGENCY — Search — OECD BETTER POLICIES FOR BETTER LIVES

Data Bank > Nuclear Data Services

NEA Nuclear Data High Priority Request List, HPRL

HPRL-Main | Search | New request template | New request guidelines | Related references

Request ID	18		Status of the request	High Priority request	
Target	Reaction and process	Incident Energy	Secondary energy or angle	Target uncertainty	Covariance
92-U-238	(n,inl) SIG	65 keV-20 MeV	Emis spec.	See details	Y
Field	Subfield	Date Request created	Date Request accepted	Ongoing action	
Fission	Fast Reactors EFR,SFR,ABTR...	28-MAR-08	11-SEP-08		

Send a comment on this request to NEA.

Requester: Prof. Massimo SALVATORES at CADARACHE, FR
Email: massimo.salvatores@cea.fr

Project (context): NEA WPEC Subgroup 26

Impact:
Design phases of selected reactor and fuel cycle concepts require improved data and methods in order to reduce margins for both economical and safety reasons. A first indicative nuclear data target accuracy assessment was made within WPEC Subgroup 26 (SG-26).

Actinides on the HPRL

23 of the 36 requests concern actinides

3 of 23 requests are exclusively thermal-resolved resonance region.
Rest cover the unresolved region and the lower fast region (< 6 MeV).
Few cases up to 10 or 20 MeV.

For completeness and consistency, modeling is essential.

All cross sections (total and partial, all energies).
All physics aspects (spectra, ddx).

In most cases the result should be very accurate.

Detailed reproduction of high quality measurements.
Account for important physics aspects that are known.
Implement these without approximation.
Identify what is not done yet.
Account for remaining model defects.

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Actinides on the HPRL

1. H3,H4 239Pu and 235U prompt fission gammas
   a. Deficiencies found in TLD measurements for JHR/RJH at MINERVE and at EOLE (C/E = 0.7, origin request: Rimpault, May 2006)
   b. Required uncertainty PFG data: 7.5%.
   c. Follow-up:
      i. SG-27 (Jacqmin)
      ii. New measurements IRMM, CEA, CNRS, JAEA
      iii. New fission modeling codes (GEF, LANL, CEA 2x)

The γ−heating in a center of a typical fast reactor core [Lut01] comes from several components, roughly:

- 20% from the γ produced in radiative capture
- 40% from the prompt γ emitted by fission fragments
- 30% from the delayed γ produced by fission products
- 10% from the inelastic scattering reactions

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2. H6, H9 U-233(n,g) and nu-bar
    a. G. Noguere (CEA, April 2006, (n,g) 10 keV – 1 MeV)
    b. Underestimation by 9% in Profil & Profil2 experiments Phenix
    c. Required uncertainty 9%.
    d. A. Bidaud (CNRS, April 2007, (n,g) and nu-bar thermal-10 keV
    e. For design studies of Molten Salt Reactors
    f. Desired uncertainty nubar: 0.5%, capture 5%
    g. Follow-up: Measurements are planned at CNRS

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3. H11 Pu-239(n,f), (n,g) SIG, ETA, ALPHA
   a. L. Leal (ORNL, May 2007, jefdoc-1158)
   b. 1 meV to 1 eV
   c. <1% for fission, < 2% for capture
   d. K-eff estimates for thermal MOX bearing reactors
   e. Need for energy dependence
   f. Follow-up
      i. Experimental follow-up would be needed
      ii. Efforts?

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4. H12 U-235(n,g), SIG, Resonance parameters
   a. Y. Nagaya (JAEA, Fast Breeder Reactor test data)
   b. 3-8% uncertainty depending on energy (100 eV – 1 MeV)
   c. Follow-up
      i. WPEC Subgroup 29 (Iwamoto, concluded 2011)
      ii. Recommendations: new alpha measurements in the keV region, new resonance analysis 0.1-2.5 keV region, other sources of overestimation k? (FCA, BFS, ZEUS)
      iii. New experiments n_TOF and CNRS in progress
      iv. Capture measurement LANL was completed

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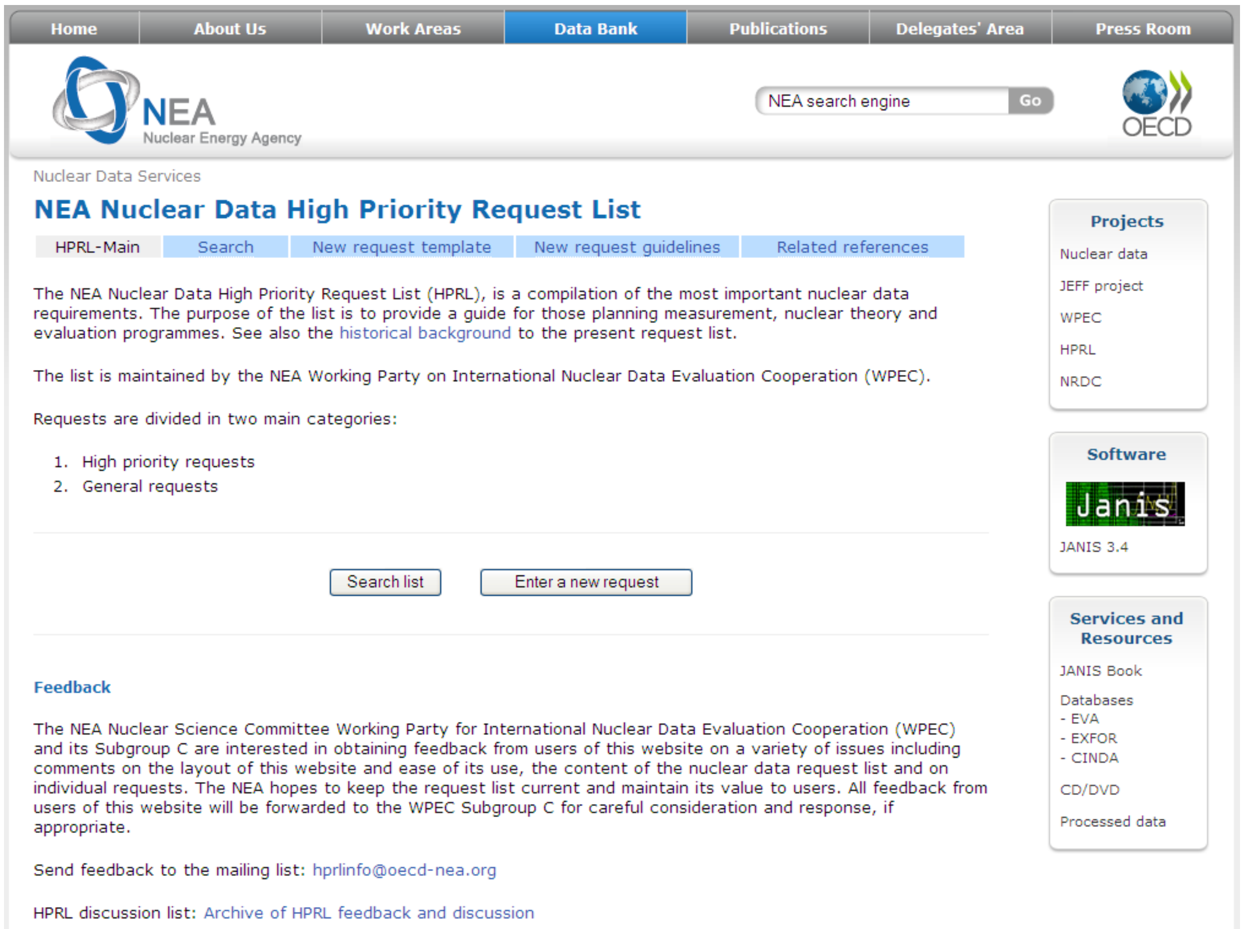
Actinides on the HPRL

6. H14 ^{242}Pu (Noguere, 0.5 eV – 2 keV, SIG tot.& (n,g), PROFIL).
7. H15 ^{241}Am (Nakagawa, ^{241}Am, thermal, SIG tot.& (n,g))
8. H16, H17 (Sasa, ^{243}Am, ^{244}Cm, th-10 MeV, PFNS)

9. H18, 19, 21, 22, 25, 27, 29, 32-39: Salvatores, WPEC SG-26, 2008

High priority request list for nuclear data

Request list coordinated by Action leader www.oecd-nea.fr/dbdata/hprl/



WPEC
Working party on evaluation cooperation

NSC
Nuclear Science Committee

OECD-NEA
Nuclear Energy Agency

Databank

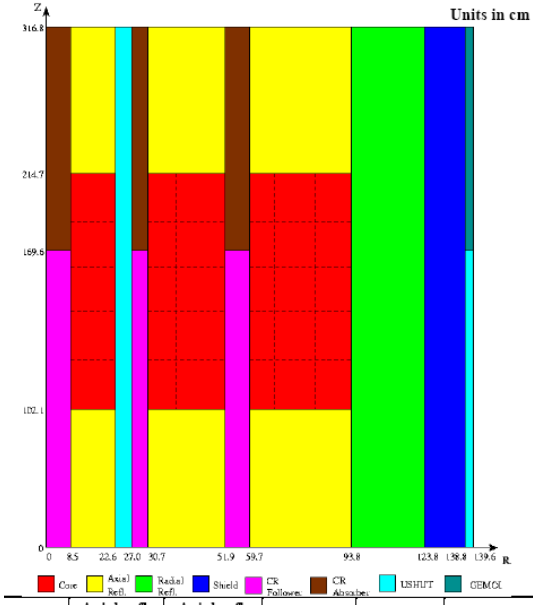
Sensitivity analysis

Quantitative underpinning of requests

System modeling

- **A simple principle…**
- **Conceptual systems**
- **Good understanding**

$$\mathbf{Q} = \mathbf{f}(p)$$



- **Future**
  - Better capabilities
  - More modeling
  - Actual designs (design dependence!)
  - Better feedback from experiments

Multiplication factor (BOL)	300 pcm
Power peak (BOL)	2%
Burn-up reactivity swing	300 pcm
Reactivity coefficients (coolant void and Doppler – BOL)	7%
Major nuclide density at end of irradiation cycle	2%
Other nuclide density at end of irradiation cycle	10%

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Sensitivity analysis

Back propagation method

- **System constraints**
- **Sensitivity coefficients**

- **Leave a domain of acceptable uncertainties**

- **Use the freedom to find the best route to achieve the final goal**

- **Cost function minimization**

System uncertainty limits L_i:

$$r_i(\mathbf{Q}) < L_i \quad \text{for } 1 \leq i \leq k$$

Equivalently for the parameters:

$$\sum_{a,b} S_{ia} S_{ib} C_{ab}(\mathbf{p}) r_a(\mathbf{p}) r_b(\mathbf{p}) < L_i^2, \quad i = 1..k$$

For each Q_i an ellipsoid in $\mathbf{r}(\mathbf{p})$-space

$$\min\left(\sum_a \frac{\lambda_a}{r_a^2(\mathbf{p})}\right)$$

The cost parameters λ_a may be changed according to the relative difficulty of obtaining low uncertainties for the parameter p_a.

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Sensitivity analysis
► Target uncertainties for nuclear data

In many cases very tight requirements

Table 26. SFR: uncertainty reduction requirements needed to meet integral parameter target accuracies

Isotope	Cross-Section	Energy range	Uncertainty (%)			
			Initial	Required		
				λ=1	λ≠1 [(a)]	λ≠1 [(b)]
U238	σ_{capt}	24.8 - 9.12 keV	9	4	3	3
	σ_{inel}	6.07 - 0.498 MeV	20	5	6	10
Pu238	σ_{capt}	183 - 24.8 keV	20	12	12	10
	σ_{fiss}	6.07 – 0.09 MeV	20	3	3	3
	ν	1.35 - 0.067 MeV	7	3	3	2
Pu239	σ_{capt}	498 – 2.03 keV	12	6	4	4
	σ_{inel}	6.07 – 0.498 MeV	25	12	15	22
Pu240	σ_{capt}	498 - 9 keV	12	5	5	4
	σ_{fiss}	6.07 – 0.0045 MeV	10	2	2	2
	ν	2.23 – 0.183 MeV	4	2	2	1
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

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Actinides on the HPRL

10.H18 U-238(n,inl), SIG, Emission spectrum

Request ID	18		Status of the request	High Priority request	
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Energy Range	Initial versus target uncertainties (%)					
�	Initial	ABTR	SFR	EFR	GFR	LFR
6.07-19.6 MeV	29	12			7	
2.23-6.07 MeV	20	3	5	4	2	3
1.35-2.23 MeV	21	4	5	4	2	2
0.498-1.35 MeV	12	7	6	5	2	2
67.4-183 keV	11	7		9	7	4

ABTR: advanced breeder test reactor
SFR: sodium-cooled fast reactor
EFR: European fast reactor
GFR: gas-cooled fast reactor
LFR: lead-cooled fast reactor

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Follow-up:

- Measurements IPHC Strasbourg (next presentation).
- Measurements RPI (Capote, yesterday).
- Improved modeling (Capote, Dupuis, Iwamoto, Kawano, Romain).

Decisive results from experiment alone have been elusive.
Interaction between theory and experiment is essential.

To make best use of available data (avoid approximations).

New experiments are planned

Improve structure data, New data for 2+ to 0+, Neutron spectra.

Actinides on the HPRL

H	19	94-PU-238	(n,f)	SIG	9 keV-6 MeV	See details	Y	Fission	31-MAR-08
H	21	95-AM-241	(n,f)	SIG	180 keV-20 MeV	See details	Y	Fission	31-MAR-08
H	22	95-AM-242	(n,f)	SIG	0.5 keV-6 MeV	See details	Y	Fission	31-MAR-08
H	25	96-CM-244	(n,f)	SIG	65 keV-6 MeV	See details	Y	Fission	04-APR-08
H	27	96-CM-245	(n,f)	SIG	0.5 keV-6 MeV	See details	Y	Fission	04-APR-08
H	32	94-PU-239	(n,g)	SIG	0.1 eV-1.35 MeV	See details	Y	Fission	04-APR-08
H	33	94-PU-241	(n,g)	SIG	0.1 eV-1.35 MeV	See details	Y	Fission	04-APR-08
H	35	94-PU-241	(n,f)	SIG	0.5 eV-1.35 MeV	See details	Y	Fission	04-APR-08
H	36	92-U-238	(n,g)	SIG	20 eV-25 keV	See details	Y	Fission	15-SEP-08
H	37	94-PU-240	(n,f)	SIG	0.5 keV-5 MeV	See details	Y	Fission	15-SEP-08
H	38	94-PU-240	(n,f)	nubar	200 keV-2 MeV	See details	Y	Fission	15-SEP-08
H	39	94-PU-242	(n,f)	SIG	200 keV-20 MeV	See details	Y	Fission	15-SEP-08

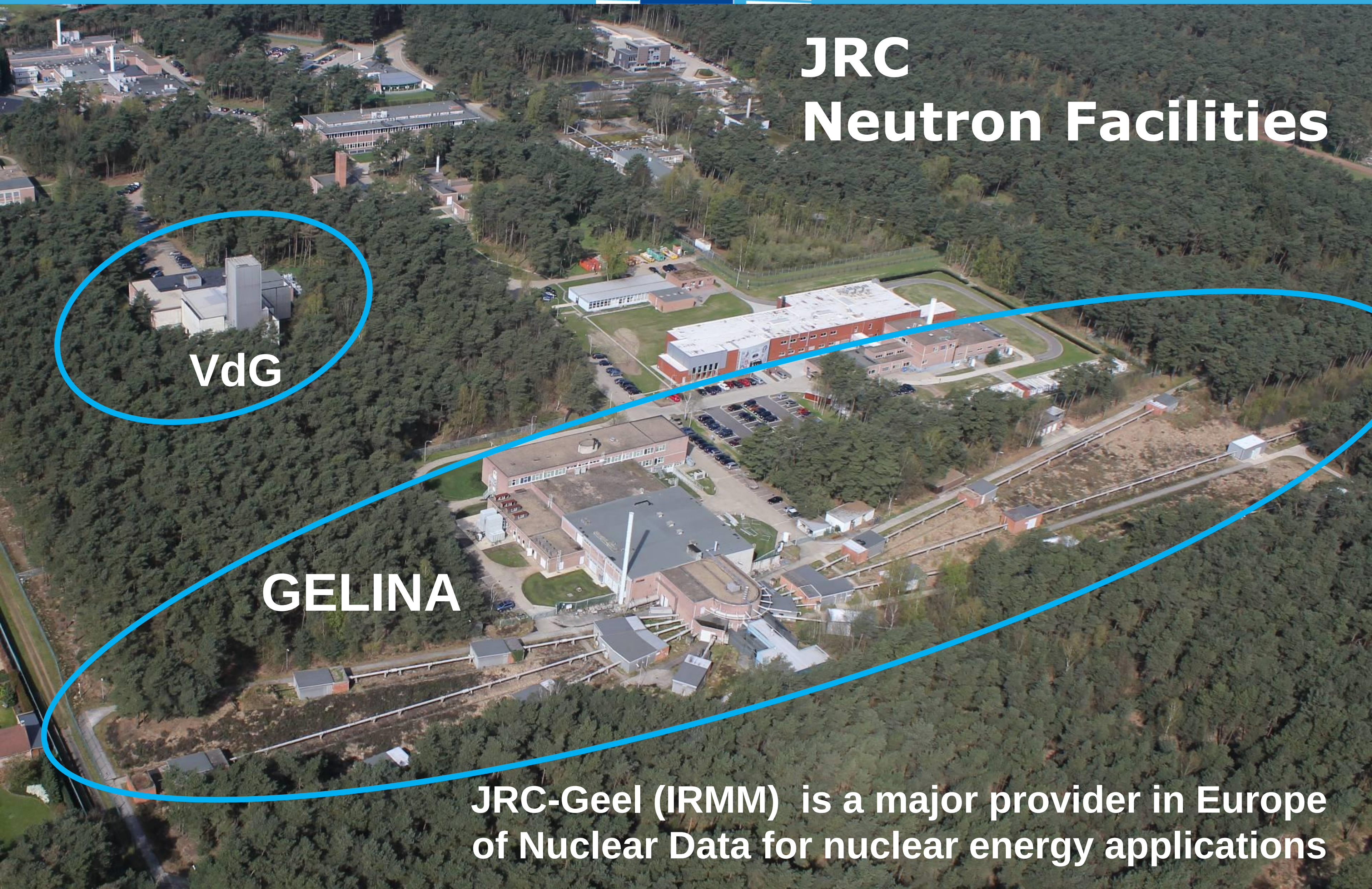
Experiences modeling with TALYS

Measurements with GAINS spectrometer and its predecessors at the GELINA facility of IRMM

(n,n'g) data.
Some activation data.

Start with short overview of projects relevant to HPRL

JRC
Neutron Facilities



JRC-Geel (IRMM) is a major provider in Europe of Nuclear Data for nuclear energy applications

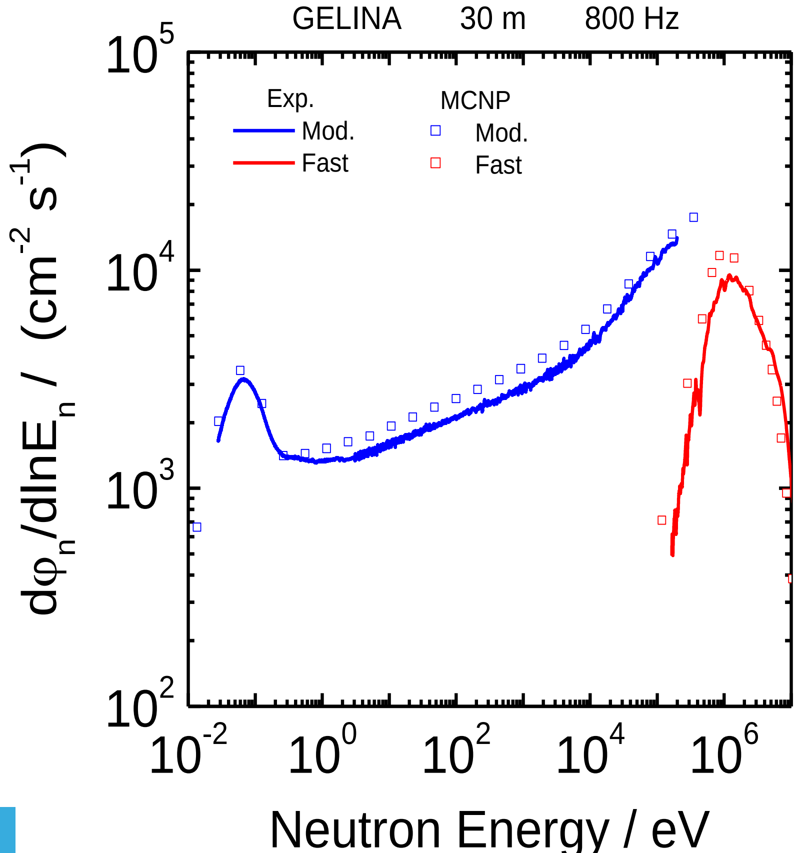
- **Pulsed white neutron source**

  10 meV < E_n < 20 MeV

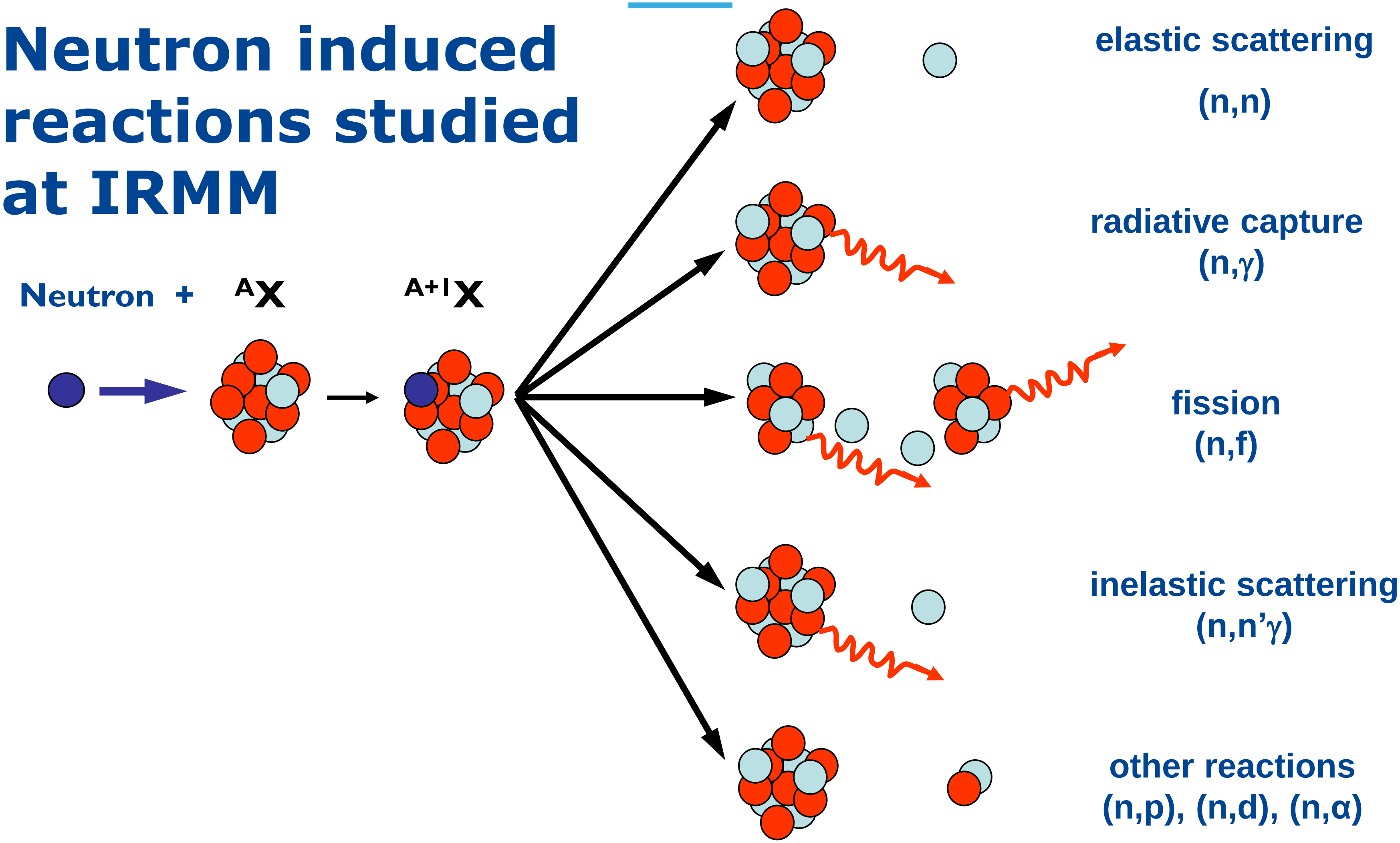
- **Neutron energy : time-of-flight (TOF)**
- **Multi-user facility: 10 flight paths**

  10 m - 400 m

Neutron Production



Neutron induced reactions studied at IRMM



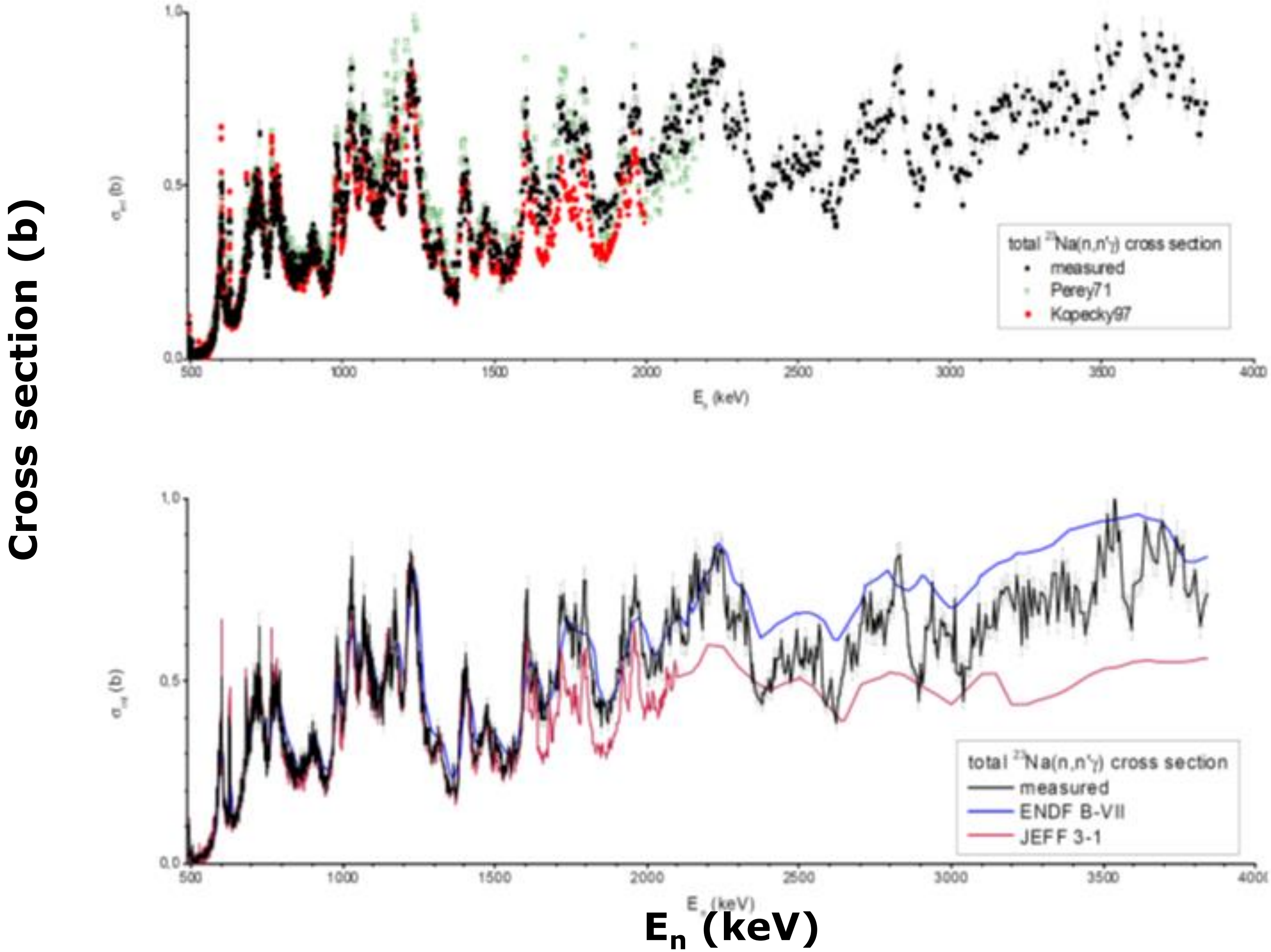
^{23}Na(n,n')

Na inelastic scattering with GAINS

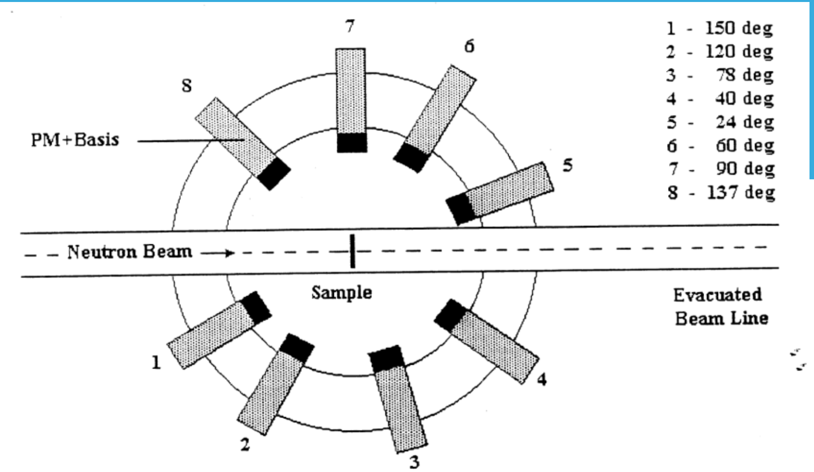
C. Rouki et al., Nucl. Instrum. Meth. A 672 (2012) 82
Na elastic and inelastic scattering with eight liquid scintillators

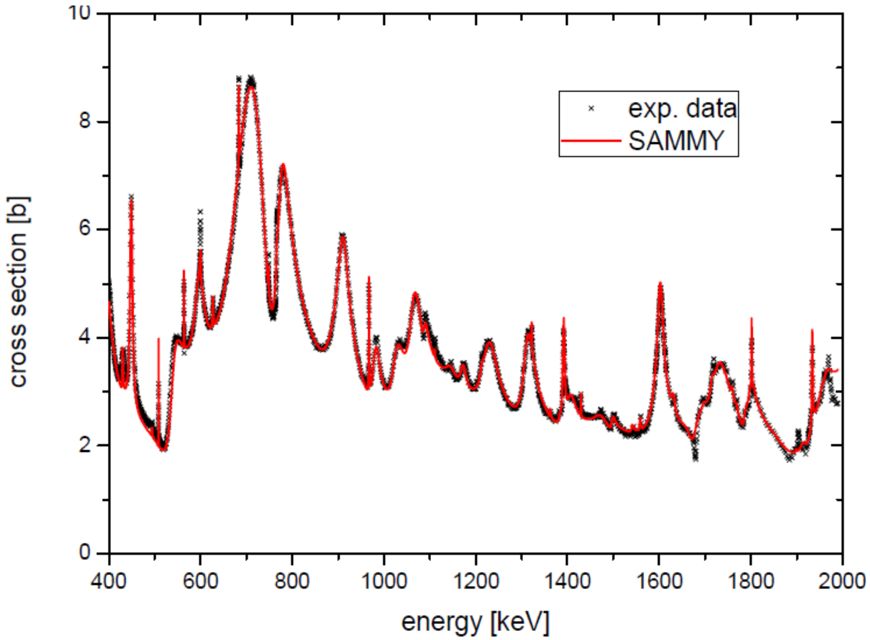
S. Kopecky and A. Plompen "R-matrix analysis of the total and inelastic scattering cross sections" EUR 25067 EN (LANA-25067-EN-N.pdf)

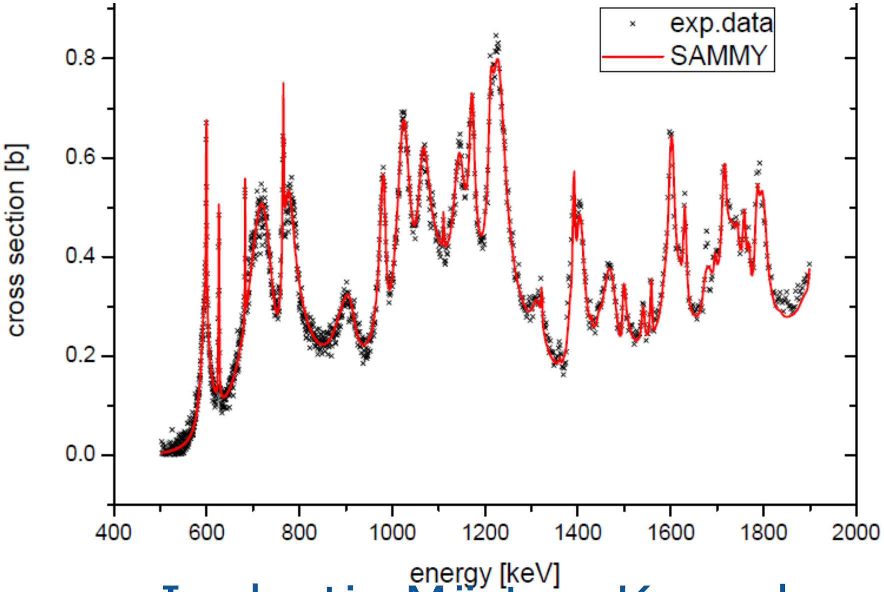
Inelastic versus other data and evaluations



R-matrix fit
total and inelastic





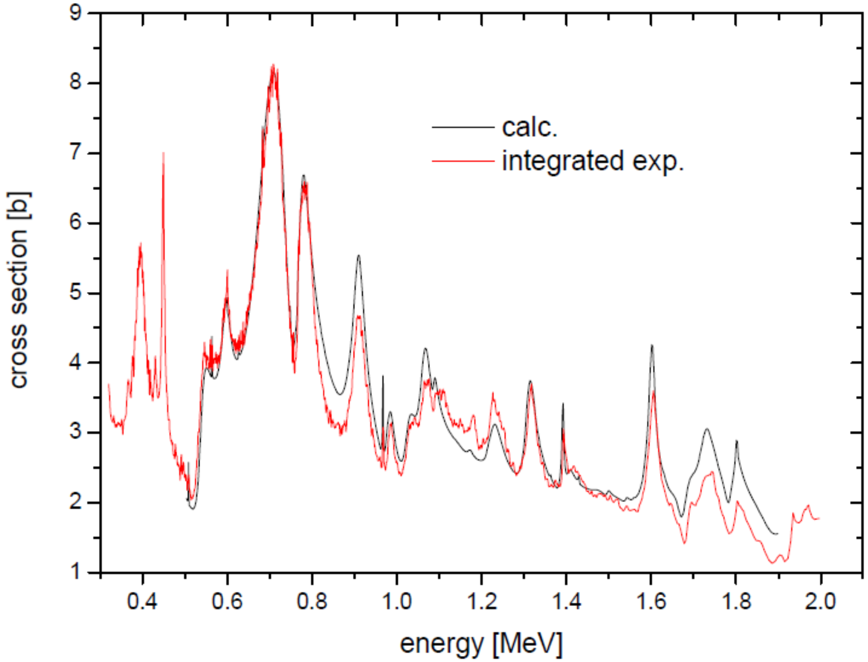


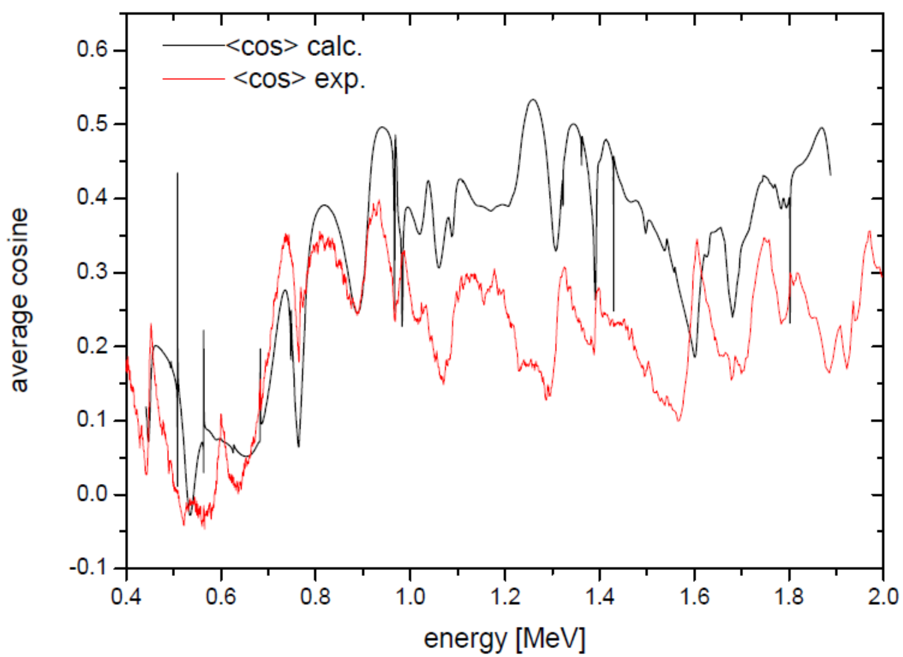
Total xs from Larson ORNL

Inelastic Märten-Kopecky

Good description, 85 resonances, 35 negative parity
Hilaire et al. expect 120 resonances, 20 negative parity

Experimental and calculated σ_{el}, μ



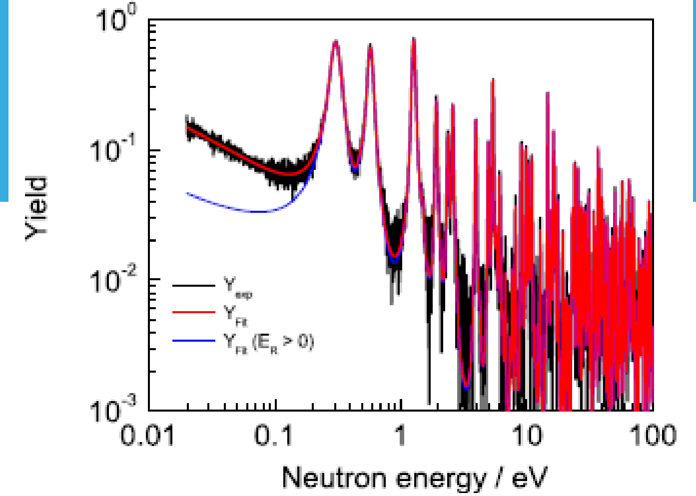


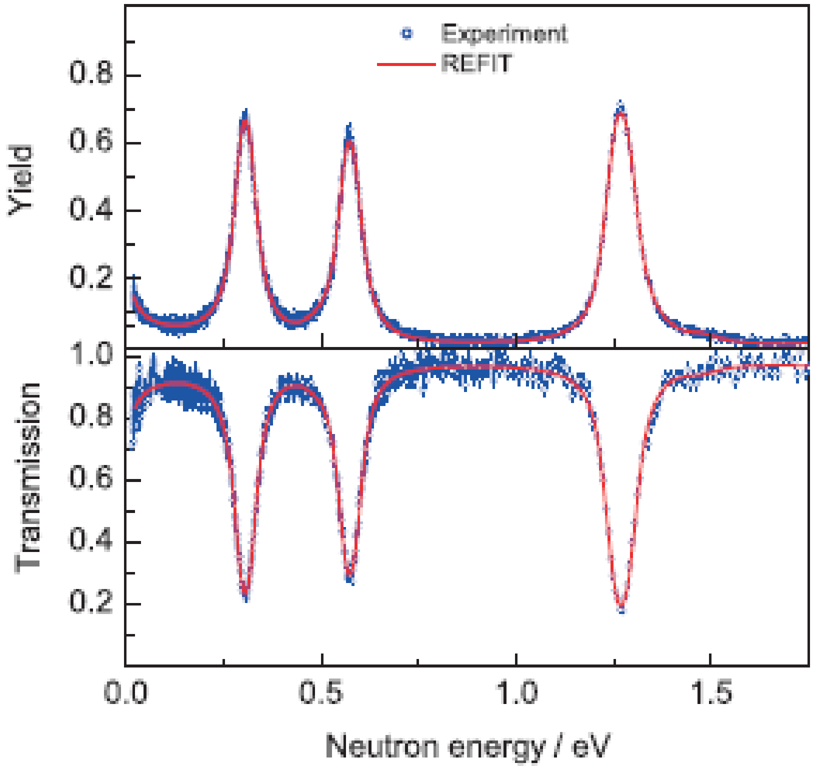
Significant discrepancies

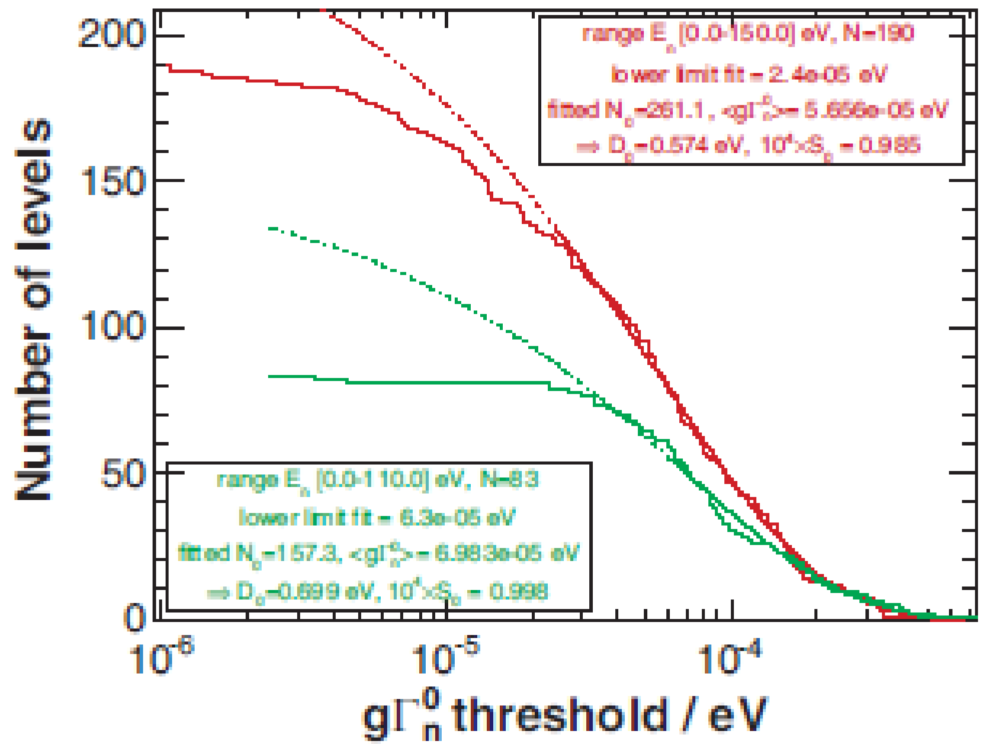
- Experimental difficulties
- R-matrix parameter ambiguities

$^{241}Am(n,\gamma)^{242}Am$

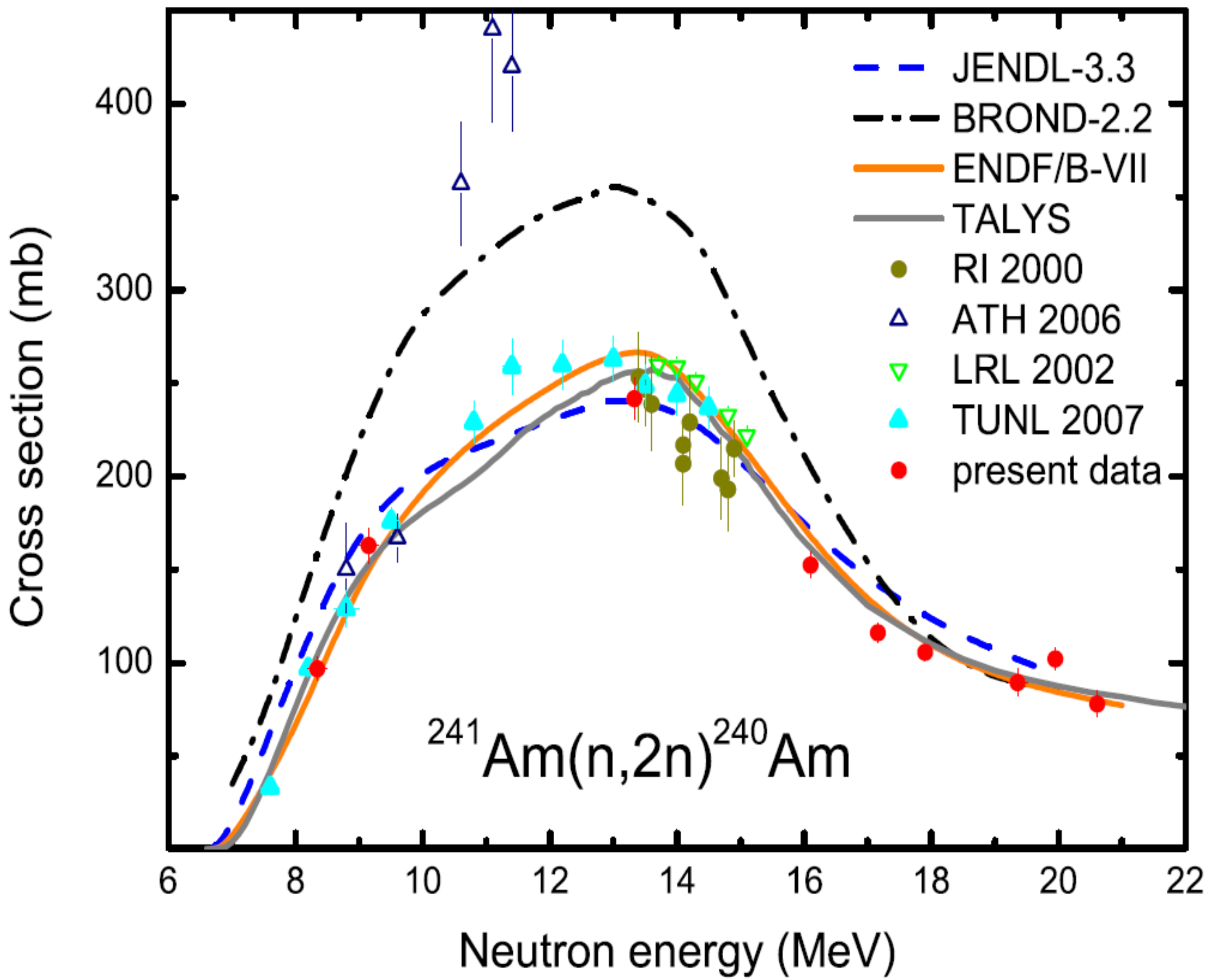
C. Lampoudis et al. EPJ Plus 128(2013)86







Results

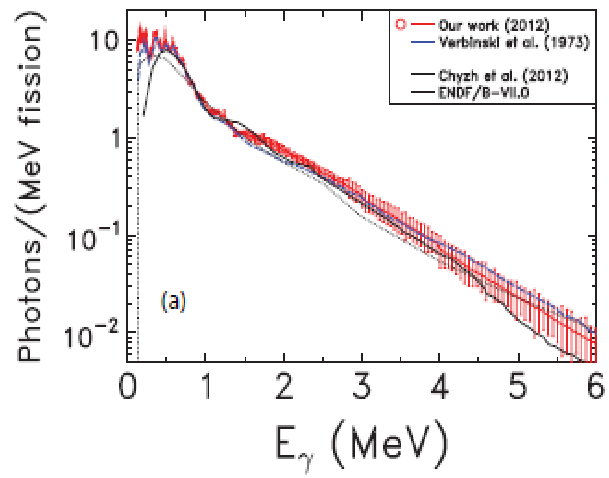


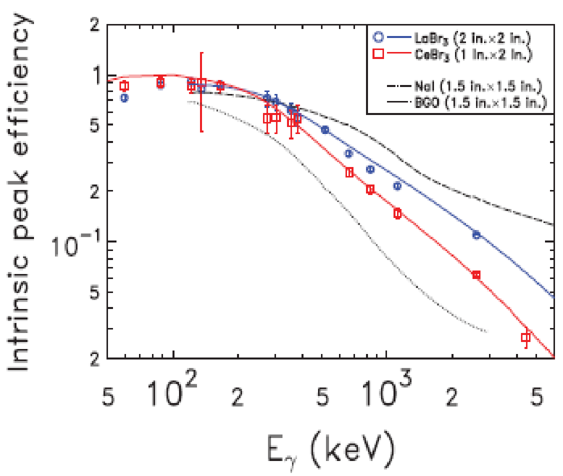
JENDL-3.3
BROND-2.2
ENDF/B-VII
TALYS
RI 2000
ATH 2006
LRL 2002
TUNL 2007
present data
$^{241}Am(n,2n)^{240}Am$
Cross section (mb)
Neutron energy (MeV)
100
200
300
400
6
8
10
12
14
16
18
20
22

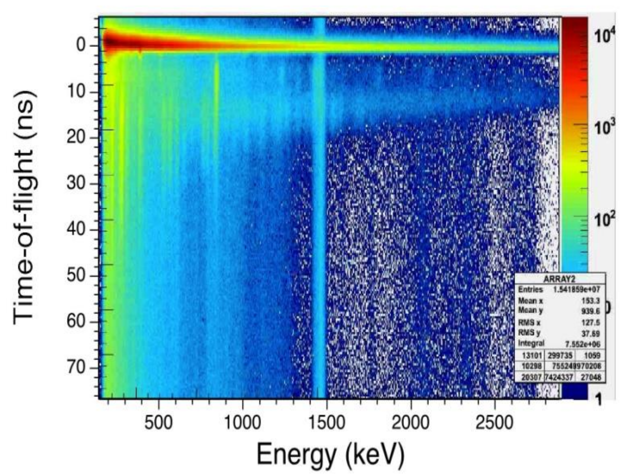
Prompt fission gammas

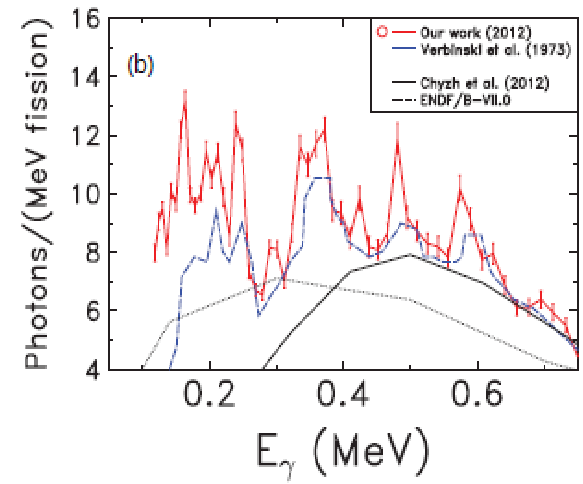
European Commission

- New gamma-ray detectors: $LaBr_3$, LaC
- Testing and characterisation
- First demonstration ^{252}Cf
- Ongoing/nearly completed ^{235}U
- TOF, FIC vs gamma detector
- Neutron-gamma separation







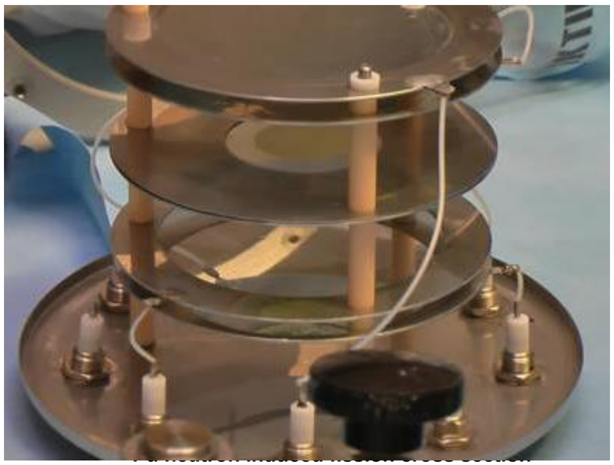


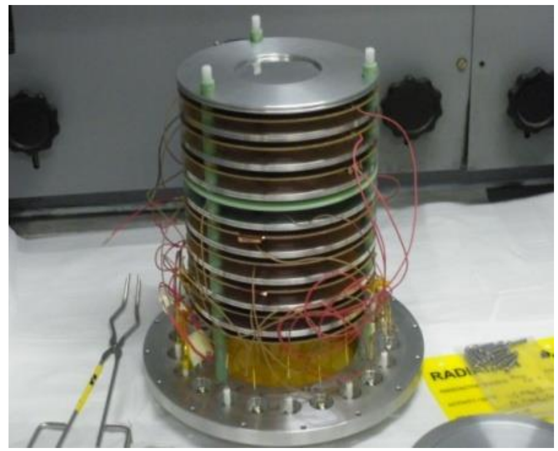
PRC87(2013)024601 Billnert et al.

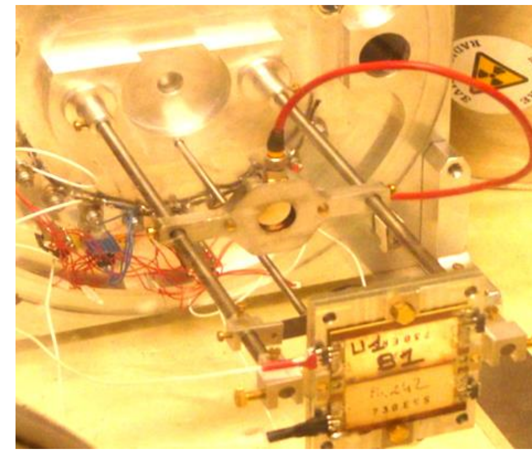
IRMM and EU experiments
^{240,242}Pu(n,f) cross sections

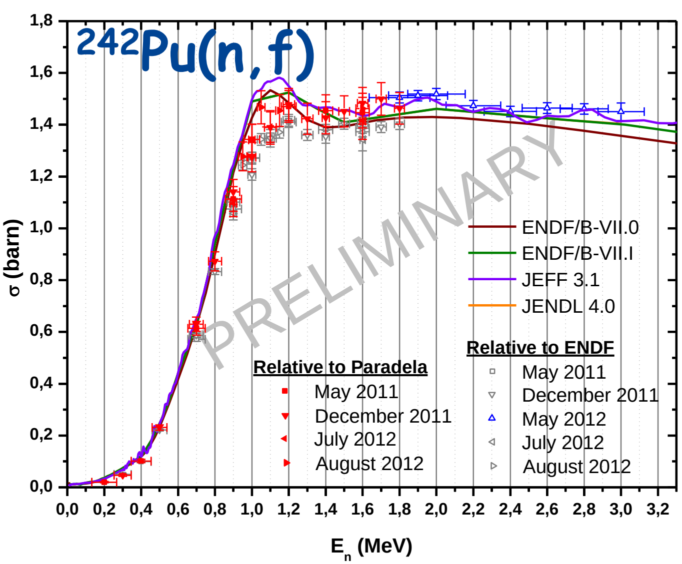
European Commission

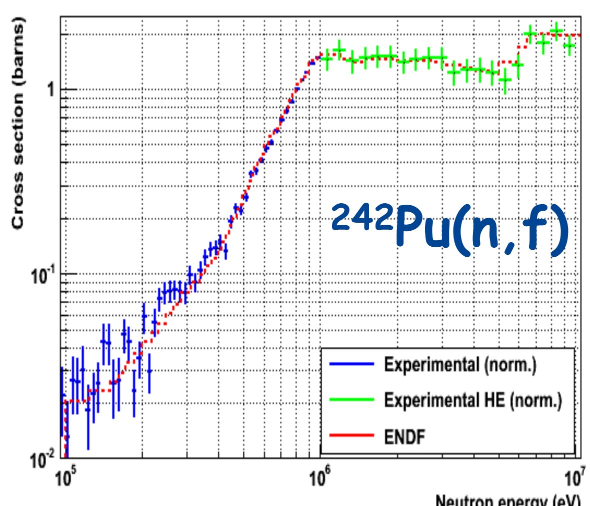
IRMM	CERN	CENBG

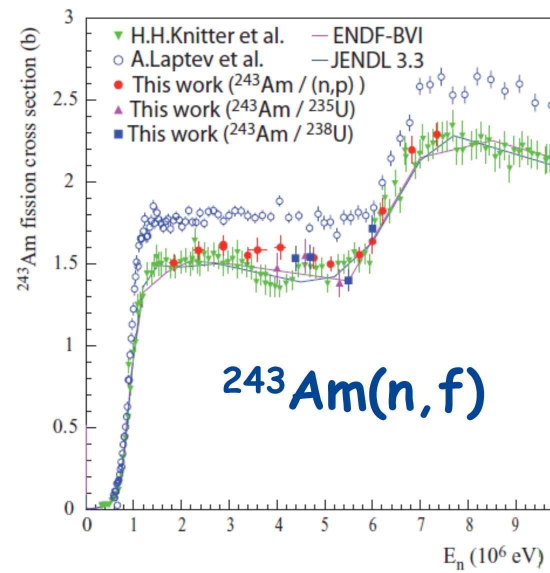












Joint Research Centre

European Commission

Germanium Array for Inelastic Neutron Scattering

GAINS @ FP3/200m

12 HPGe 80 mm ø x 80 mm L
1 keV resolution at 1 MeV (neutrons)
Cross sections 3-5 %

Joint Research Centre

GAINS

Angle integration: $\lambda \leq 3$
Efficiency: calib+MC
Time-response
  12bit 440 MSPS dig.
Flux: U-235(n,f)

L.C. Mihailescu et al. NIMA531(2004)375
L.C. Mihailescu et al. NIMA578(2007)298
D. Deleanu et al. NIMA624(2010)130
A. Plompen et al. KPS59(2011)1581
^{52}Cr: L.C. Mihailescu et al. NPA786(2007)1
^{209}Bi: L.C. Mihailescu et al. NPA799(2008)1
^{208}Pb: L.C. Mihailescu et al. NPA811(2008)1
^{23}Na: C. Rouki et al. NIMA672(2012)82
^{235}U: M. Kerveno et al. PRC87(2013)024609
$0\nu2\beta$ bgs: A. Negret et al. PRC…(2013)…
^{12}C, ^{24}Mg, ^{28}Si, ^{56}Fe, ^{58}Ni, ^{76}Ge, ^{206}Pb, ^{207}Pb, ^{232}Th, ^{238}U: conf.
Ongoing: ^{57}Fe, ^{63}Cu, ^{65}Cu, Mo, Zr

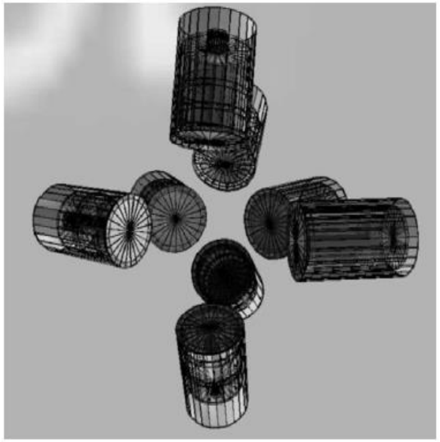
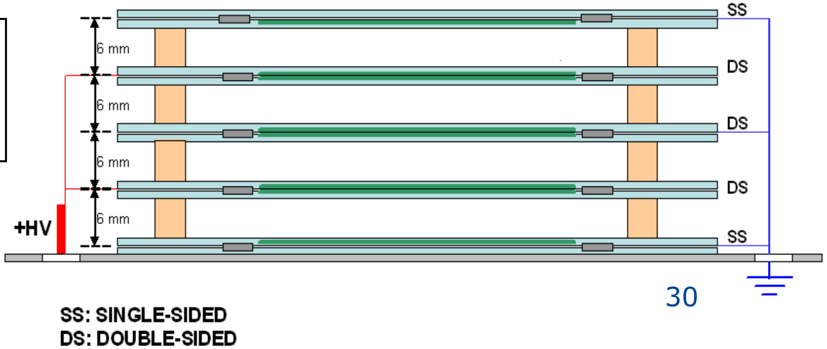


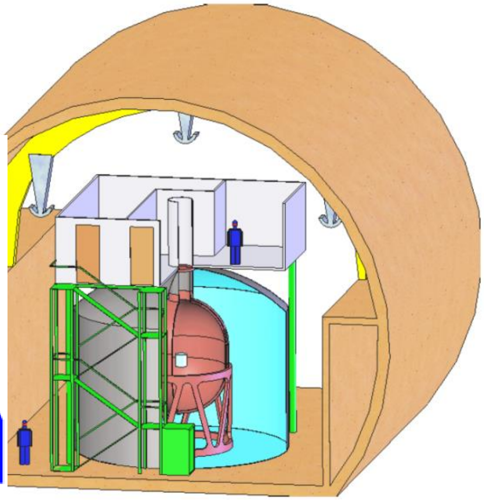
Fig. 1. GAINS. Drawing of the simulated geometry.

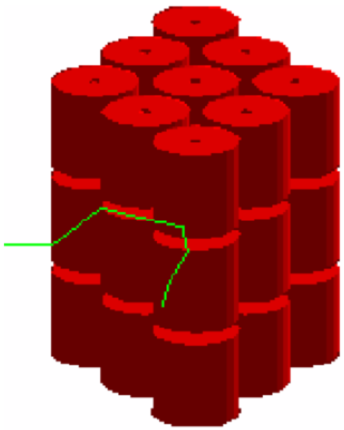
$$\sigma = 2\pi \int_{-1}^{1} \frac{d\sigma}{d\Omega}(x)dx = 2\pi \sum_{i=1}^{2} w_i \frac{d\sigma}{d\Omega}(x_i)$$



European Commission

$^{76}Ge(n,n'g)^{76}Ge$
w. K. Zuber, A. Domula TUD





Motivation: background in $0\nu\beta\beta$-experiments

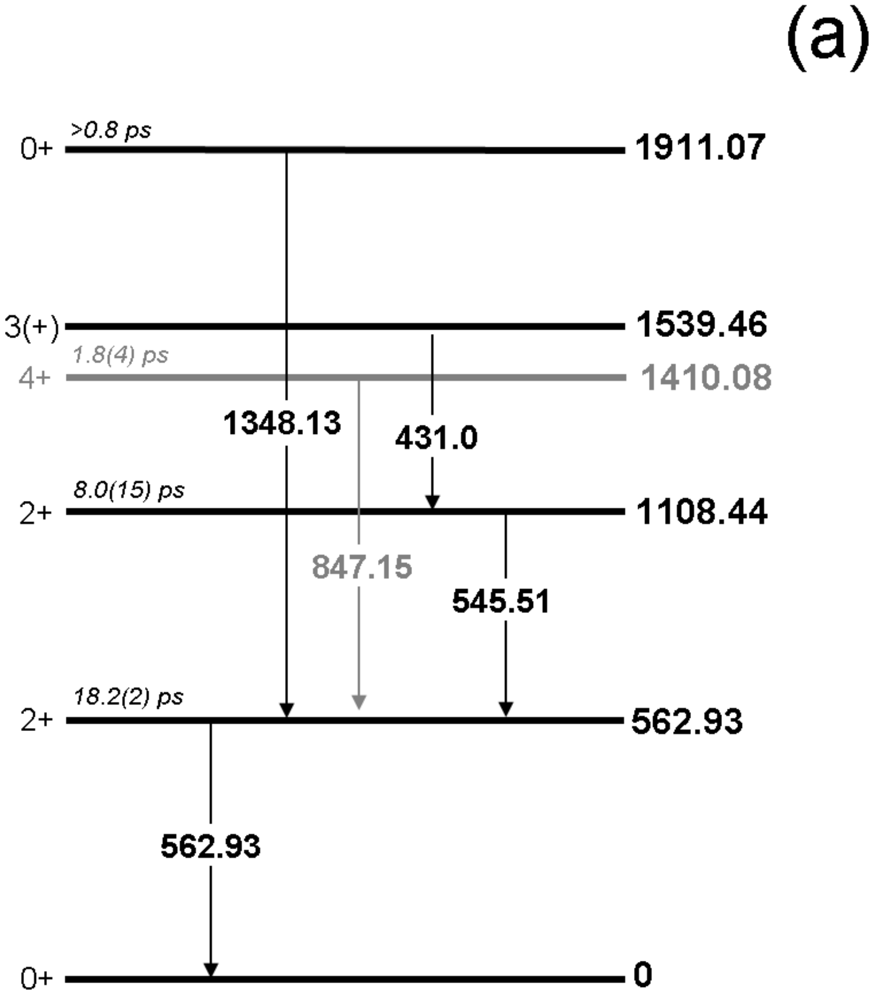
- Is a neutrino its own antiparticle?
- What is the neutrino mass?

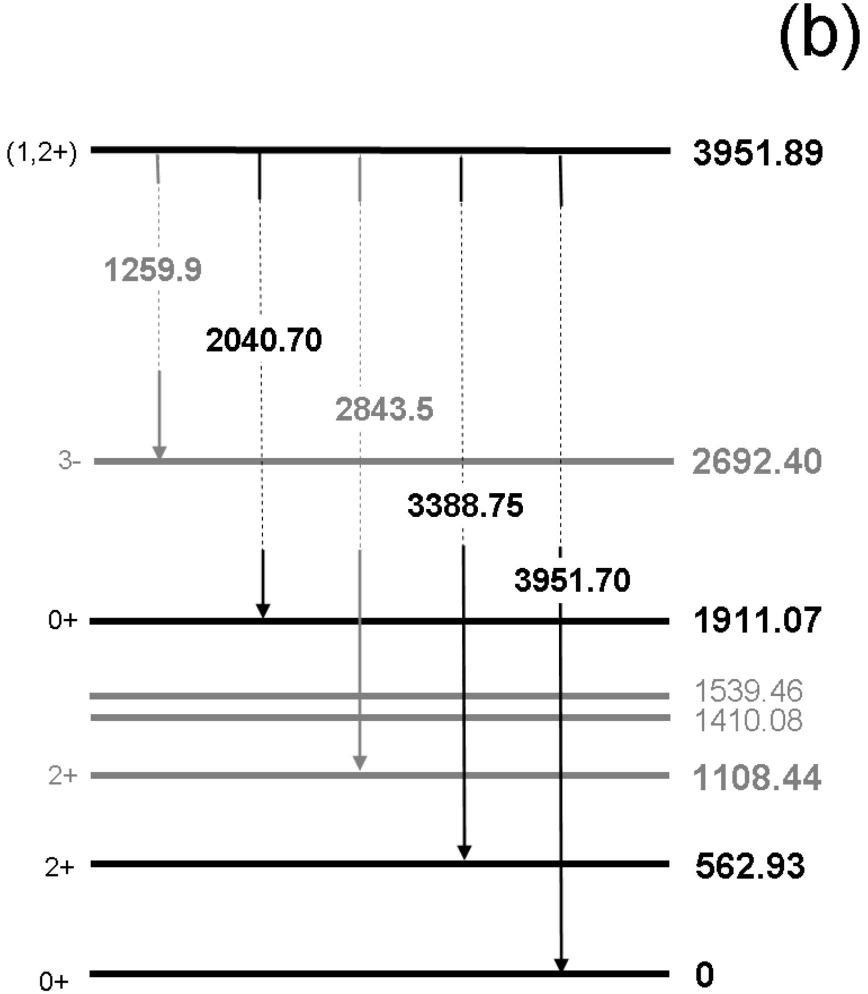
GERDA experiment

- ^{76}Ge, $Q_{\beta\beta}$ = 2039 keV, $T_{1/2} < 2\ 10^{25}$ y
- ^{76}Ge high purity detectors, 9 coaxial 8 x 8cm ø
- Gran Sasso, 3600 mwe
- Background goal 10^{-3} keV^{-1} kg^{-1} y^{-1}
- Components few times 10^{-4} keV^{-1} kg^{-1} y^{-1}

- Two concerns for neutrons
  - Direct production of 2040 keV transition
  - Indirect background due to Eg + Erecoil in inelastic scattering

Relevant portions level scheme ^{76}Ge





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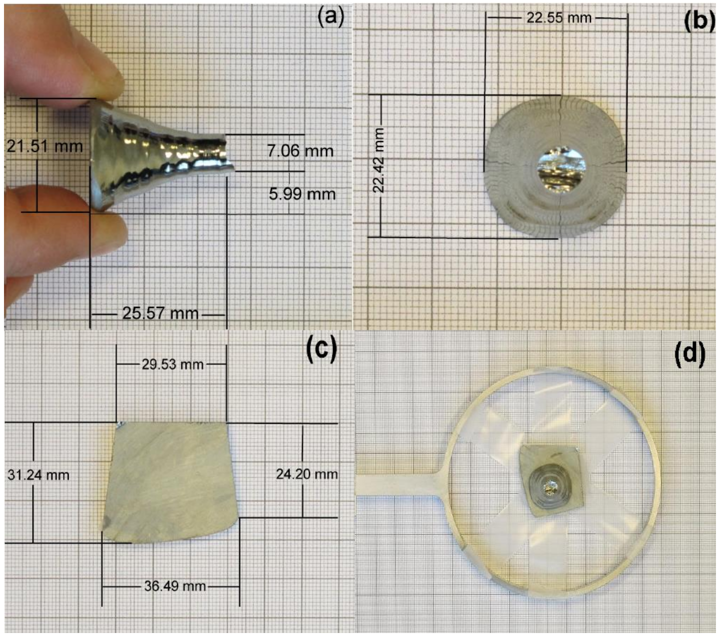
Experiment
$^{76}Ge(n,n'g)^{76}Ge$

European Commission

32 g, 87% enriched in ^{76}Ge
main systematic uncertainty 10%

Cross section of 2039 keV,
L69→L5 **< 3 mb**
Unshielded: 0.43 event/kg/y
(100x above limit)

Shielded: not an issue (3m H2O!)
Future experiments?



Experimental results
- Five gammas, five levels, INL

TALYS model calculations
- Default phenom.
  - KD omp
  - GC LD
  - Kopecky-Uhl γ-strength

-Modified OMP

-Effect of deformation

-DWBA, Rotational, Asymmetric (Toh et al. PRC 2013).

-Microscopic
  - JLM omp, LDA, HF dens.
  - enhanced combinatorial LD
  - HFB γ-strength
  - Here similar to default (not shown)

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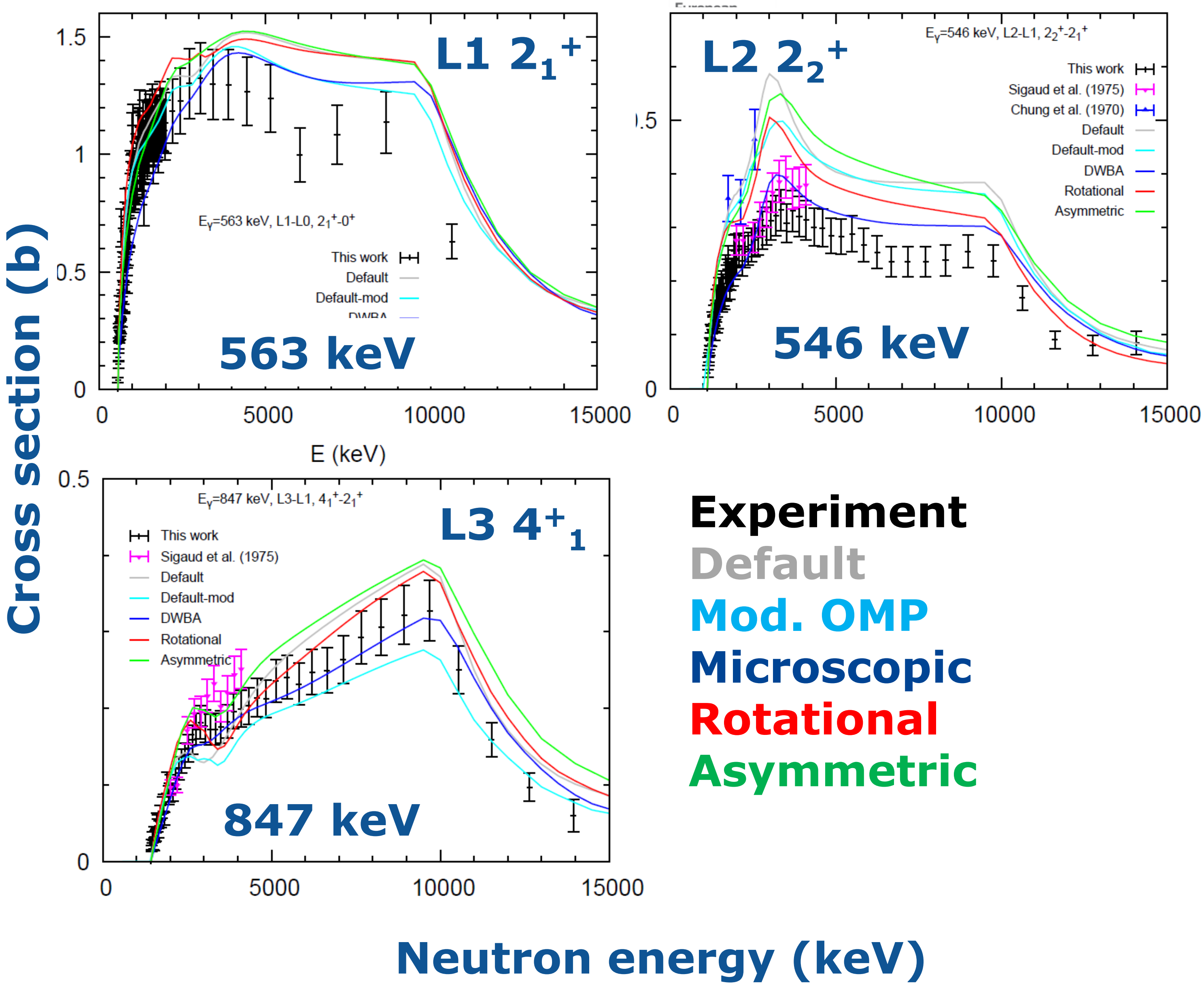
Gamma emission cross sections

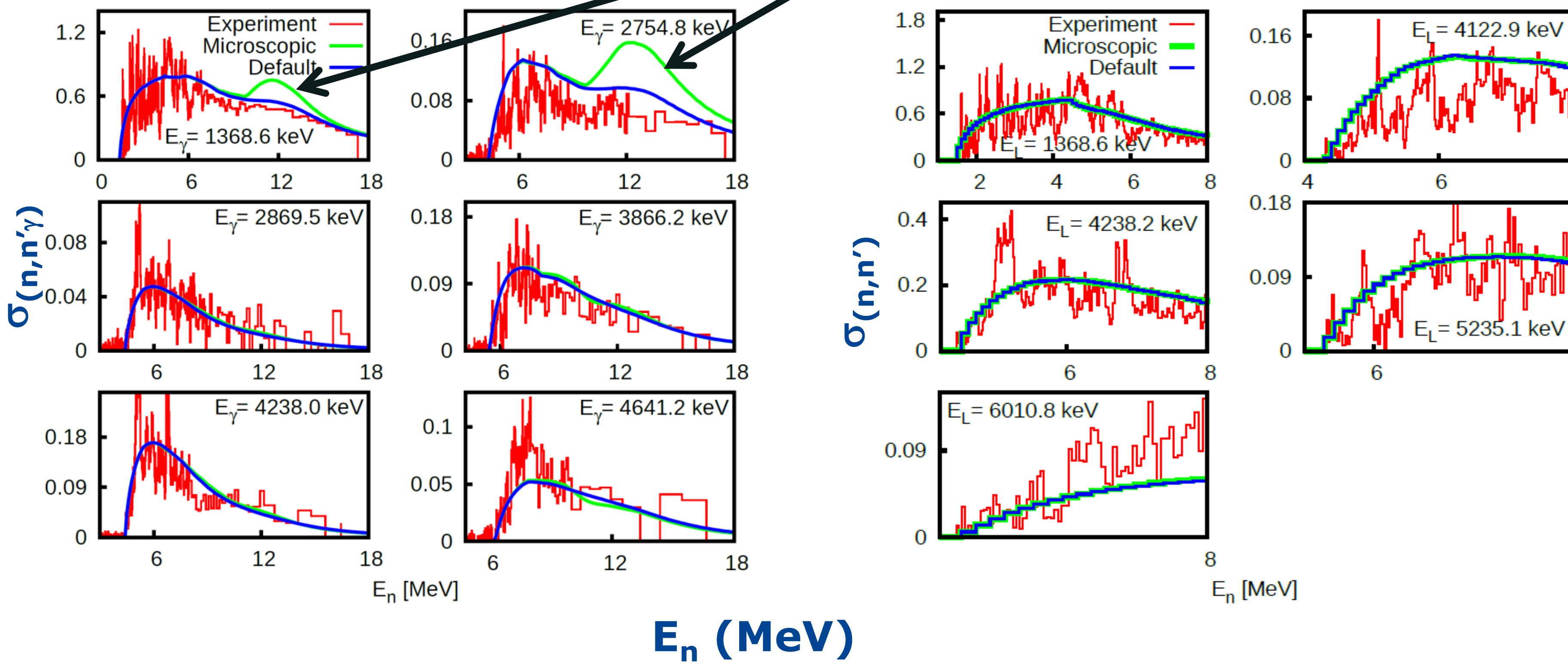


24Mg(n,n'g) modeling

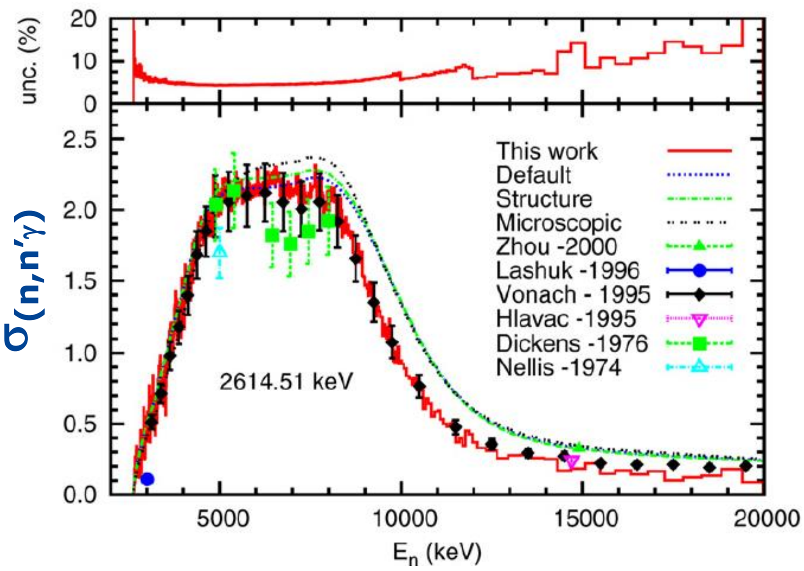
- Microscopic model
  - Level densities Hilaire, Goriely
  - Optical model Bauge
  - Gamma strength functions Goriely
- Compared with default semi-empirical model of TALYS

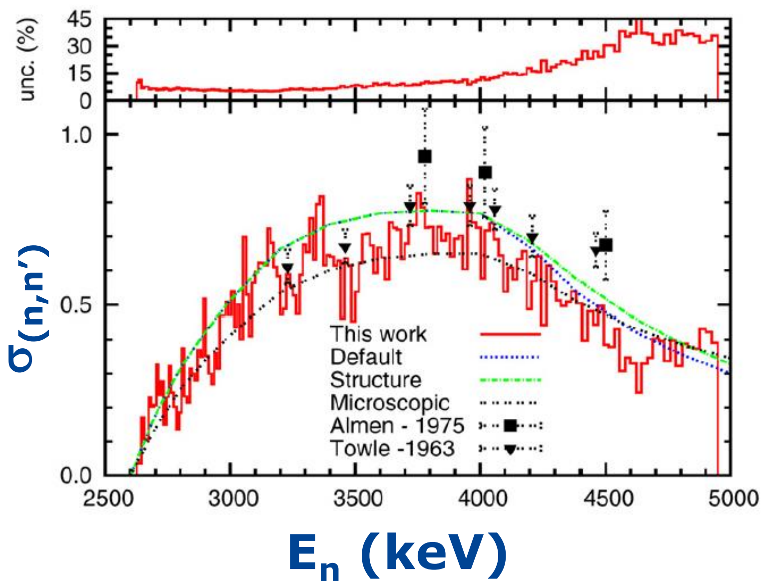
$^{24}Mg(n,n'g)$

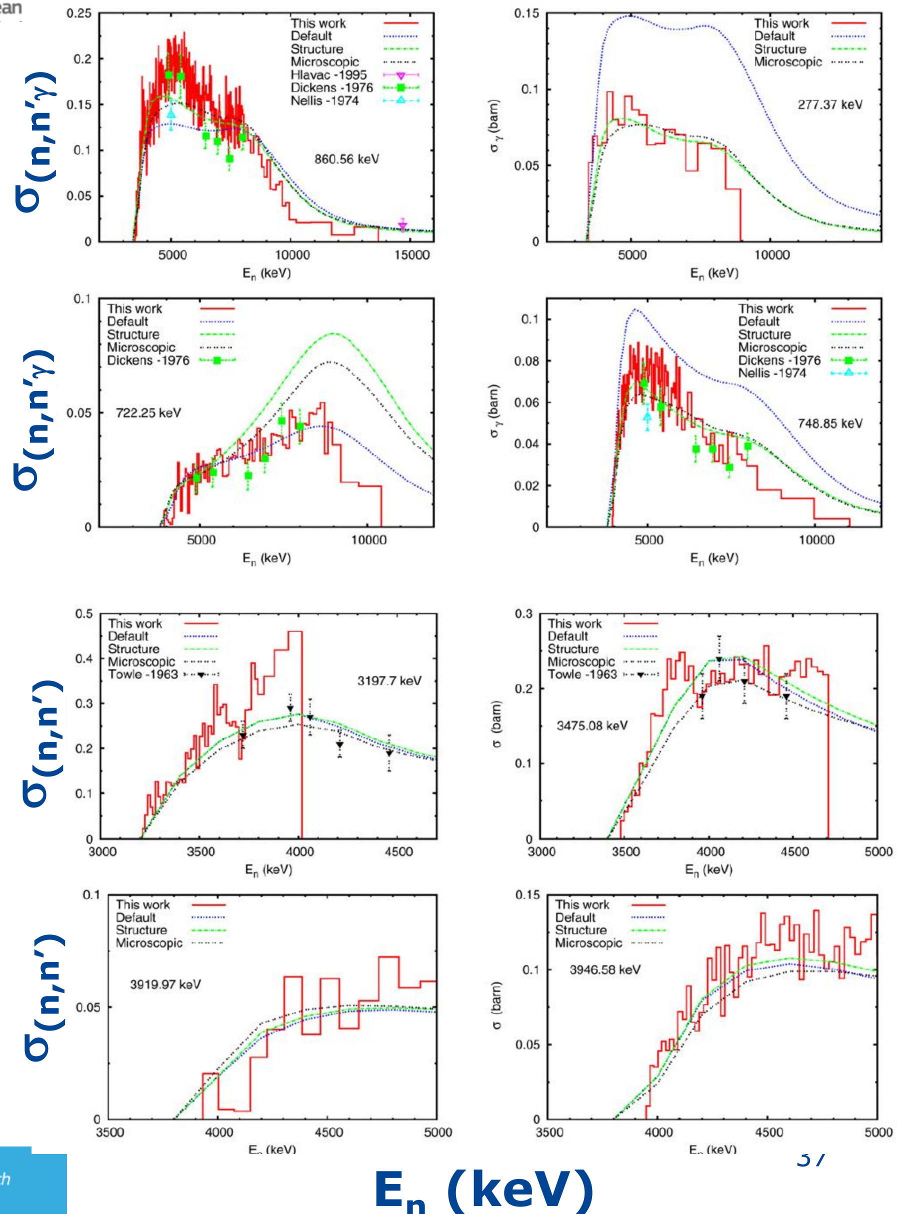
Suppressed competing channel?



$^{208}Pb(n,n'\gamma)$







Angular distributions γs: not isotropic

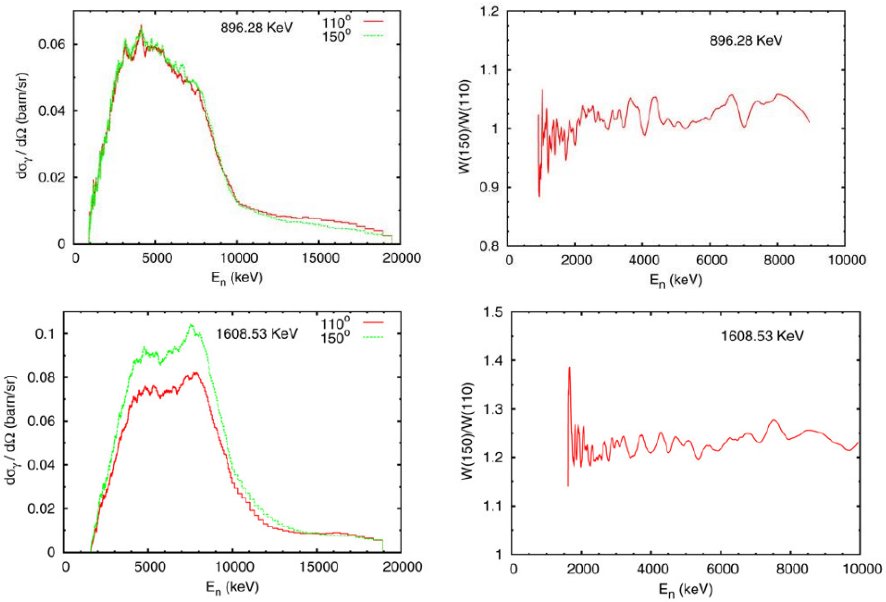


Fig. 5. *Left*: Smoothed differential gamma production cross-section for the 1608.53 keV transition in ^{209}Bi. *Right*: The ratio $W(\theta)$ of the angular distribution at 150 to that at 110 degrees for the same transition.

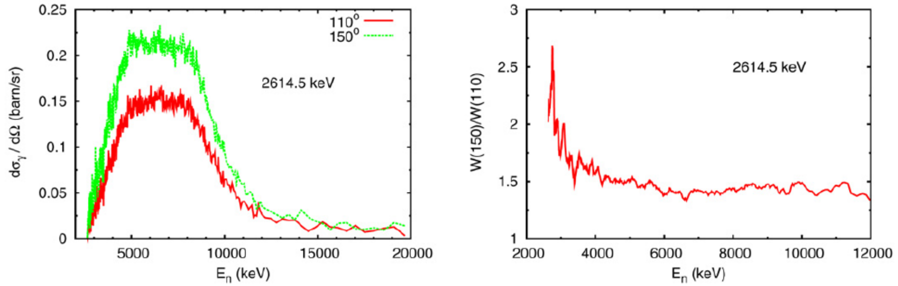


Fig. 3. Differential gamma production cross section for the 2614.51 keV transition in ^{208}Pb. Left: smoothed curves from the threshold up to 20 MeV at two angles, 110° and 150°. Right: the ratio between the angular distribution $W(\theta)$ at 150° and 110°.

^{208}Pb top
^{209}Bi left

Table 1: Low-lying states in ^{168}Er

State	E_x	J	K	π
1	0.0	0	0	+
2	0.08	2	0	+
3	0.264	4	0	+
4	0.549	6	0	+
5	0.862	2	2	+
6	0.895	3	2	+
7	0.93	8	0	+
8	0.99	4	2	+
9	1.12	5	2	+
10	1.13	4	4	-

Level	Conventional HF Cross Section	Deformed HF Cross Section
1	1.0	1.0
2	1.84	0.39
3	0.28	0.04
4	10^{-9}	7×10^{-9}
5	1.66	0.78
6	0.86	0.31
7	8×10^{-10}	5×10^{-9}
8	0.25	0.08
9	0.08	0.02
10	0.25	0.10

2 MeV

Level	Conventional HF Cross Section	Deformed HF Cross Section
1	1.00	1.00
2	3.04	0.63
3	1.89	0.24
4	0.49	0.05
5	2.93	1.35
6	2.64	0.90
7	0.043	0.004
8	1.85	0.52
9	1.04	0.253
10	1.71	0.64

6 MeV

4 Summary

A new Hauser Feshbach code has been developed which includes K as a quantum number in calculating decay of the compound nucleus. Very small effects are found for the continuum. Cross sections for the population of resolved final levels are frequently changed by 40% or more. A systematic tendency is found such that cross sections for levels of large J are reduced and cross sections for low J states are enhanced. Cross sections for $K = 0$ states are reduced relative to those of states of the same J with $K > 0$.

Accounting for K in transmission coefficients Varenna 2012 (PRC)

^{168}Er

Hauser Feshbach Calculations in Deformed Nuclei

S. M. Grimes

Dramatic effect; Should it be accounted for? W data IPHC may help

Conclusions

Interaction theory-experiment is essential to make best use of expensive experiments

- Interpretation of results
- Implications for what is not measured
- Avoid unnecessary approximations in reporting experimental results

(N,n'g) data

- Non-actinide cases Are pretty well modeled semi-empirically and microscopically with certain interesting exceptions (physicist's opinion)
- Are just not modeled well enough (engineer's opinion)

HPRL

- Successful in guiding work to be done
- Needs a review to account for all the follow-up
- New directions