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Member of the US Nuclear Data Program

Experimental Nuclear Data Activities at ANL

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U.S. Department of Energy

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Office of Science U.S. DEPARTMENT OF ENERGY

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CSEWG-USNDP Meetings, BNL, November 4-7, 2008

Highlights

❑ **Decay studies of selected actinide nuclei** (with I. Ahmad & J. Greene, ANL-PHY & A.L. Nichols & M.A. Kellett, IAEA **-** part of the ANL commitment to the IAEA-CRP on *"Updated Decay Data Library for Actinides"*

✓studies of **^{233}Pa, ^{237}Np, ^{240}Pu, ^{242m}Am, ^{243,244,245,246}Cm & ^{249,250}Cf** using α–decay and γ–ray spectroscopy techniques and mass separated sources

✓during FY08 work focused on priority nuclides identified by the IAEA-CRP where large data discrepancies exist: **^{233}Pa, ^{243}Cm & ^{237}Np**

❑ **Development of ANL TAGS** (ANL LDRD project – ANL-NE and PHY & ANSTO/ANU, Australia) – **AFC** (decay heat at short cooling times), **Homeland Security** (cargo inspections) & **astrophysics** applications – in conjunction with **CARIBU** RIB facility at ANL, based on 1Ci - ^{252}Cf source (will be operational in FY09)

❑ **Studies of ^{236}Np with GS and CHICO** (ANL, LLNL,UR, Kolkata) – data relevant to ^{237}Np(n,2n)^{236}Np CS measurements

❑ **Studies at the accelerator driven sub-critical facility YALINA** (NNSA sponsored: **Y. Gohar**, G. Aliberti, A. Talamo, Z. Zhong, FGK & colleagues from JIPNR-Sosny)

✓ successfully converted from 90% HEU to 36% LEU – full characterization of the assembly at different fuel loadings – reactivity, activation & spectra unfolding measurements – detailed analysis at ANL using various ND libraries

^{233}Pa γ–ray emission probabilities

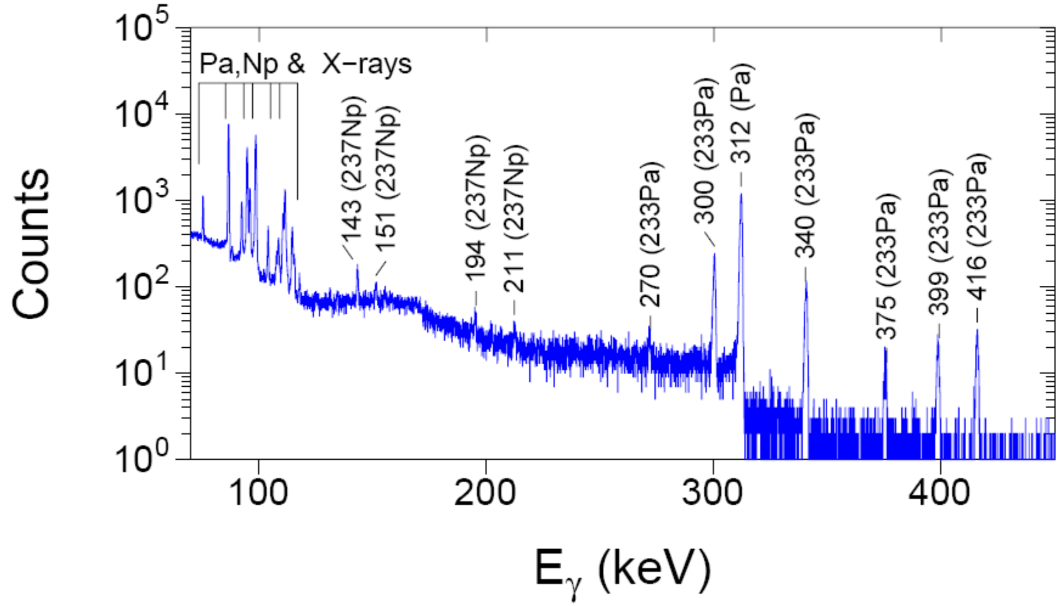
- it has been of a special interest since the first IAEA-CRP (1977-1984)
  - ✓ very high-precision measurement on P_γ (312 keV), e.g. 38.6 (5) % (Gehrke et al.), 38.6 (15) % (Smith et al.), 38.5 (4)% (Schotzig et al.), 38.65 (39) % (Vaninbroukx et al.), 38.7 (4) % (Woods et al.), 37.80 (23) % (Luca et al.), BUT …
    41.6 (9) % (Harada et al. J. Nucl. Sci. and Techn. 43 (2006) 1289)
- at the last two meetings inconsistencies for P_γ(28.6 keV) were pointed out

E_γ/keV	P_γ (%)									
	Albridge et al. (1961)	Valkeapaa et al. (1973)[a]	Gehrke et al. (1979)	Vaninbroukx et al. (1984)	Kouassi et al. (1990)[a]	Luca et al. (2000)	Schotzig et al. (2000)	Woods et al. (2000)	Luca et al. (2002)	Shchukin et al. (2004)
28.559(10)		0.070(8)		0.15(1)	0.075(8)	0.034(10)			0.034(10)	0.019(2)

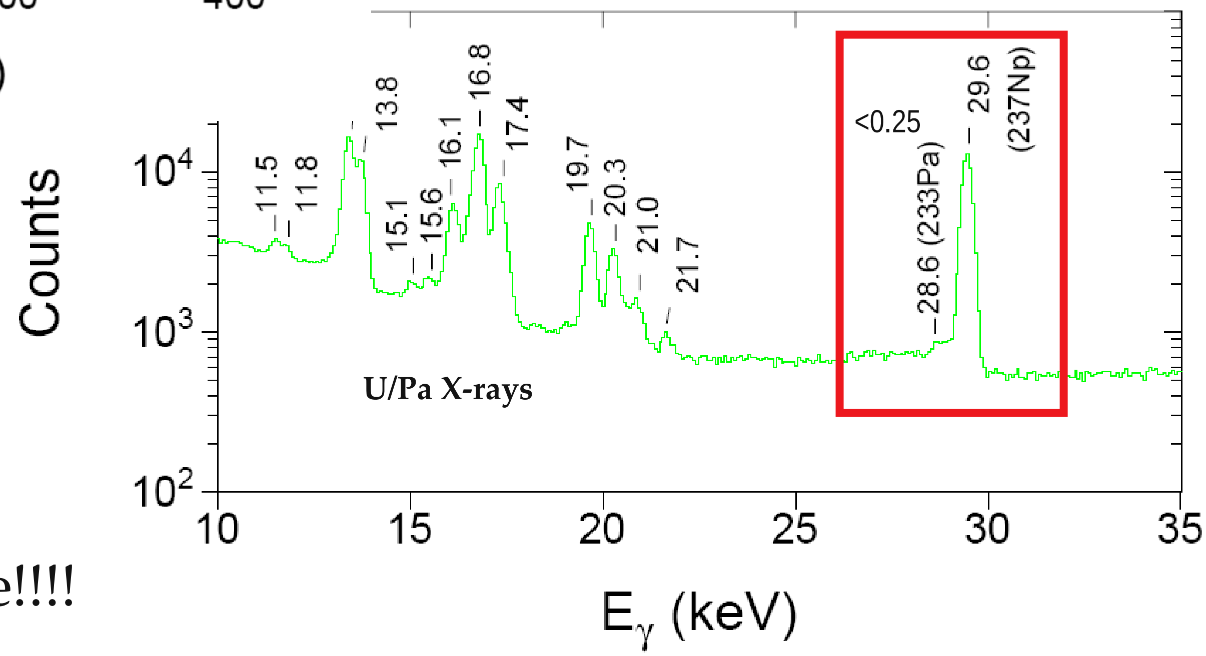
> Why is important? – 28.6 keV (M1+E2) transition determines the β– feeding to the 5/2+, 340 keV level - strongly fed in β– decay of ^{233}Pa:
>
> $P_{\gamma,tot}(28.6\ keV)=P_\gamma(28.6\ keV)*(1+\alpha_T)=P_\gamma*\mathbf{311!}$

- there are differences between various measurements
- there are differences between various evaluations
- there has been a lot of effort in the past, but the decay scheme is still "discrepant"

^{233}Pa γ–ray emission probabilities - cont



- initial measurements (FY07) using a mass separated ^{237}Np source (4 nCi) & γ–ray counting – ^{237}Np/^{233}Pa in equilibrium
- high isotopic purity



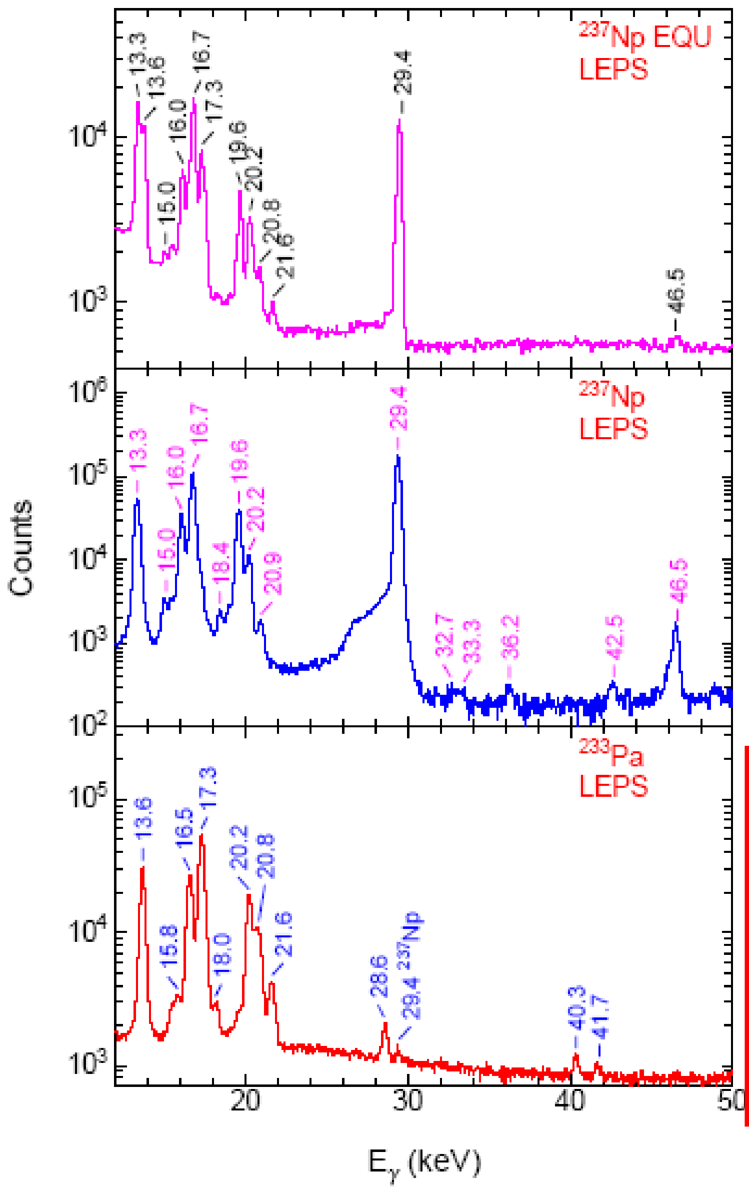
- … but, it is inconclusive!!!!

^{233}Pa γ–ray emission probabilities - cont

New measurements at ANL – (August/September 2008)

- using a chemical separation to extract ^{233}Pa from ^{237}Np
  - ✓ procedure was similar to that used by Gehrke et al. – dissolved ^{237}Np material (in equilibrium with ^{233}Pa) in HNO_3 – transferred the solution to a beaker and dried it – use 4 M of HNO_3 to dissolve ^{237}Np (but not ^{233}Pa) – repeat the procedure several times to achieve the desired purity
  - ✓ several sources were produced and measured with 3 cm^3 LEPS & 25% Ge detectors – efficiency calibration determined using a calibrated mixed source containing ^{57,60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{139}Ce, ^{203}Hg and ^{241}Am nuclides, and isotopically pure ^{243}Am source – accuracy ~1% for low- and high-energy photons

^{233}Pa γ–ray emission probabilities - cont



- ✓ 29.4 keV ^{237}Np line dominates – high Compton tail that masks 28.6 keV (^{233}Pa)
- ✓ Compton background associated with much stronger high-energy γ–rays of ^{233}Pa
- ✓ $P_\gamma(29.4/^{237}Np)/P_\gamma(75.3/^{233}Pa) = 10.6\ (1)$

- ✓ no ^{233}Pa – 75.3 keV line (^{233}Pa) is gone – pure Pa X-rays (from decay of ^{237}Np)
- ✓ Compton background associated with the high-energy γ–rays of ^{233}Pa is reduced

- ✓ 29.4 keV ^{237}Np line is significantly reduced, e.g. $P_\gamma(29.4/^{237}Np)/P_\gamma(75.3/^{233}Pa) = 0.060\ (17)$
- ✓ pure U X-rays
- ✓ S(28.6 keV)=9000 counts – statistical uncertainty of about 1%

^{233}Pa γ–ray emission probabilities - cont

E_γ/keV	P_γ (%)									
	Albridge et al. (1961)	Valkeapaa et al. (1973)[a]	Gehrke et al. (1979)	Vaninbroukx et al. (1984)	Kouassi et al. (1990)[a]	Luca et al. (2000)	Schotzig et al. (2000)	Woods et al. (2000)	Luca et al. (2002)	Shchukin et al. (2004)
28.559(10)		0.070(8)		0.15(1)	0.075(8)	0.034(10)			0.034(10)	0.019(2)

Eγ, keV	present	Valkeapaa73	Kouassi90	Gehrke79	Vanin.84	Woods88
28.57	0.076 (3)	0.068(8)	0.074 (8)		0.15 (1)	0.068 (9)
29.37	0.0169 (15)					
39.77	0.0034 (9)					
40.33	0.0228 (14)	0.039 (8)	0.024 (4)			
41.65	0.0121 (10)	0.013 (4)	0.014 (3)			
75.26	1.27 (3)	1.25 (8)	1.25 (9)	1.39 (8)	1.30 (4)	1.25 (9)
86.57	2.00 (4)	1.87 (23)	1.93 (11)	1.97 (12)		1.87 (25)
94.64	8.51 (17)					
98.42	13.70 (27)					
103.84	0.85 (2)	0.73 (8)	0.847 (6)	0.87 (3)	0.87 (3)	0.73 (9)
110.41	1.64 (3)					
111.30	3.23 (7)					
114.48	1.31 (3)					
115.38	0.423 (9)					
271.57	0.361 (12)	0.30 (3)	0.334 (17)	0.33 (1)	0.32 (1)	
300.16	6.41 (13)	6.57 (31)	6.76 (6)	6.62 (10)	6.64 (6)	6.57 (46)
311.94	38.6 (5)			38.6 (5)	38.65 (39)	

ANL TAGS - introduction

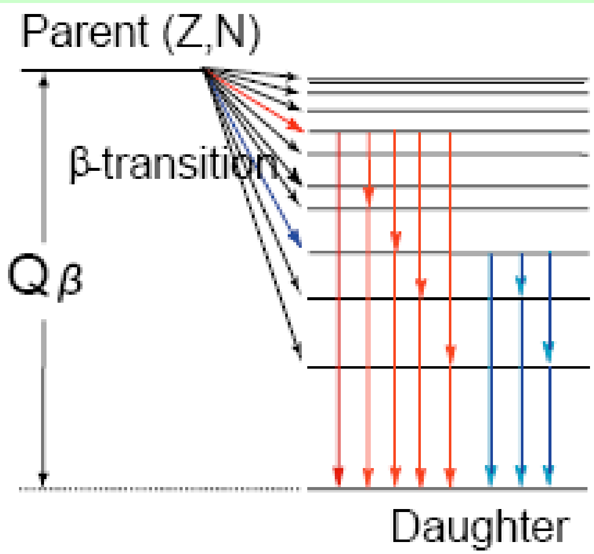
- required decay data are incomplete

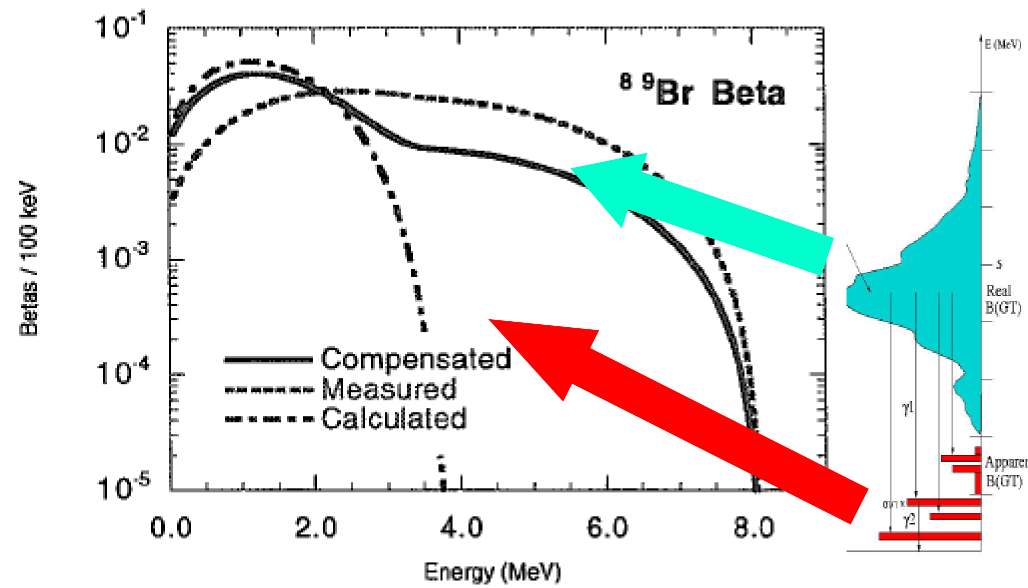
$$Q_{ls} \neq Q_m$$

  - ✓ lack of pure, intense FP sources
  - ✓ lack of modern detector systems in past β⁻ decay studies

"Pandemonium" effect

J. C. Hardy *et al.*, Phys. Lett. **71B** (1977) 307





J. Katakura *et al.*, JNST, Suppl. 2 (2002) 444

- JENDL FP (based on ENSDF) - "contaminated" by Gross Beta-decay Theory for ~500 FP (almost half of all FP)!
- there are significant differences between various libraries, e.g. JEFF vs. JENDL vs. ENDF

"large" $Q_{\beta-}$ – **large density of levels** and more complicated decay schemes – usually **low γ–ray multiplicities**, but isomers!

- about 50 cases studied using TAGS, but there are also drawbacks
- only a handful of cases studied with modern γ–ray arrays (e.g. GSI, ORNL)

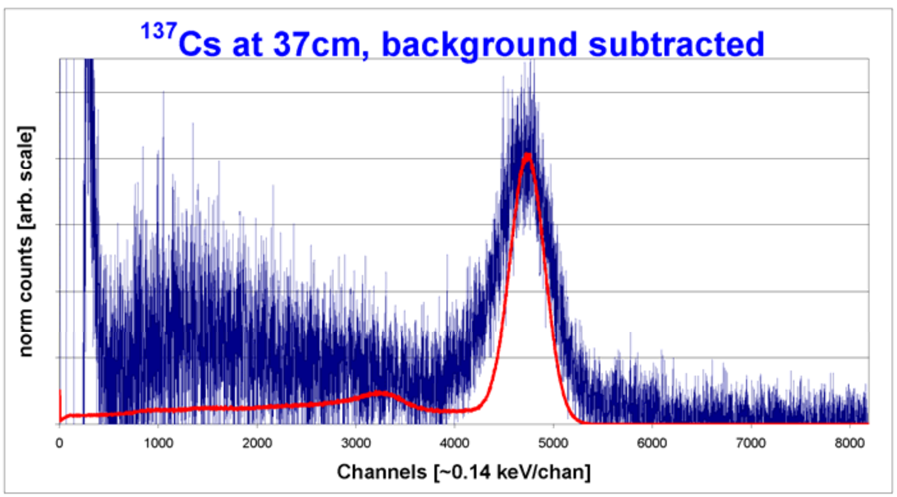
ANL TAGS – cont.

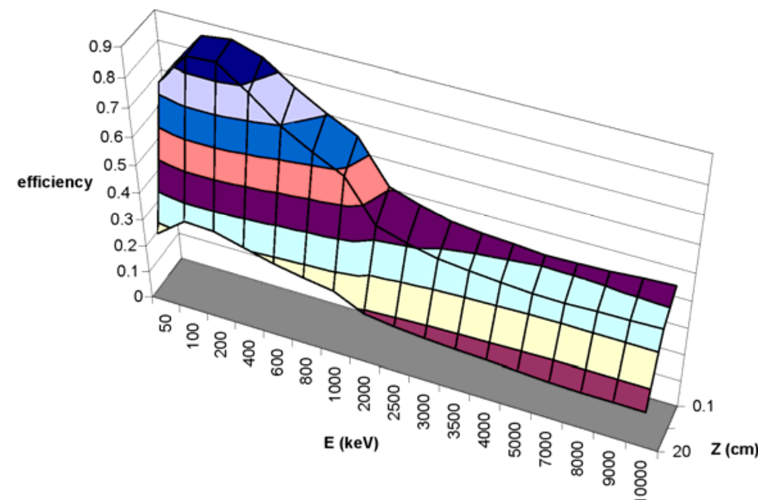
<u>ANL LDRD/DCG funding – C.J. Chiara, F.G. Kondev (NE), K. Lister (PHY), M. Smith (ANSTO/ANU)</u>

based on the INEL NaI(Tl) detector

developing necessary infrastructure – electronics, tape-moving system, shielding, etc.; tests using RA sources

possibility to use other state-of-the art equipment at ANL – GS & FMA





ANL TAGS – cont.

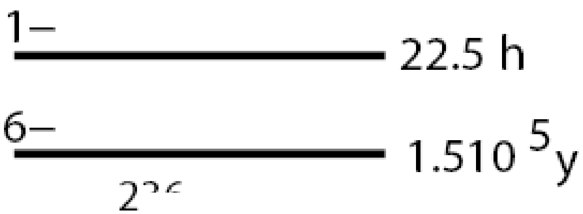
- Development of data analysis tools in conjunction with GEANT simulations (ENSDF data are incorporated so that helps, but will need human intervention, as well …)
  - ✓ TAGS data analysis is **not straight-forward** – uncertainties?

$$d_i = \sum_{j=0}^{j_{max}} R_{ij} f_j, \; i = 1, i_{max}$$

  - – ***d*** = observed spectrum; ***f*** = level feeding distribution (decay scheme); ***R*** = response function of detector folded with the decay scheme
  - ✓ we are developing a new unfolding method (faster) that would allow to use MC procedure to determine uncertainties – the idea is similar to that of A. Koning and others in CS studies – the modeling work on FP at LANL is also of very high value!

- approved ATLAS experiment on "Beta-delayed fission studies in the Pb region" (in collaboration with LANL) – opportunity to test the equipment and analysis tools
- will participate early next year in an IAEA consultants' meeting (in collaboration with NEA-OECD) to coordinate effort and enhance collaborations with groups from EU & India – a lot of new data expected in the foreseen future – good news for DE!

Data relevant to $^{237}Np(n,2n)^{236}Np$ CS



very long-lived ground state – activation measurements difficult – similar to $^{239}Pu(n,2n)$

RAPID COMMUNICATIONS

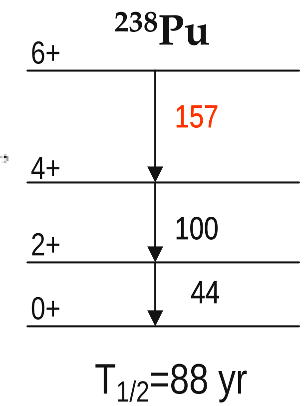
PHYSICAL REVIEW C, VOLUME 65, 021601(R)

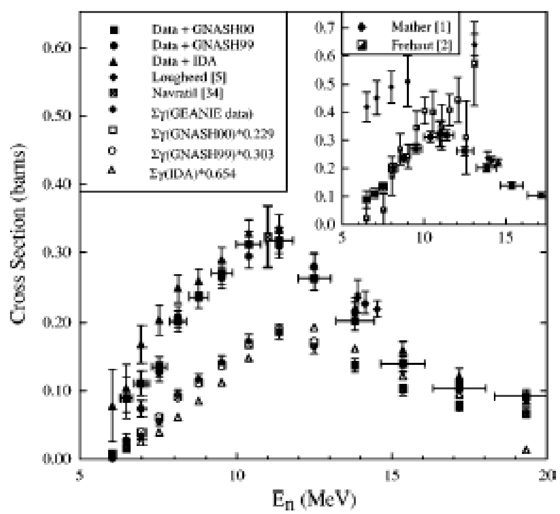
$^{239}Pu(n,2n)^{238}Pu$ cross section deduced using a combination of experiment and theory

L. A. Bernstein, J. A. Becker, P. E. Garrett, W. Younes, D. P. McNabb, D. E. Archer, C. A. McGrath,* H. Chen, W. E. Ormand, and M. A. Stoyer
Lawrence Livermore National Laboratory, Livermore, California 94551

R. O. Nelson, M. B. Chadwick, G. D. Johns, W. S. Wilburn, M. Devlin, D. M. Drake, and P. G. Young
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 23 July 2001; published 9 January 2002)





measured partial σ(n,2nγ) CS → deduced total σ(n,2n) (with the magic of theory!)

BUT…

in the case of ^{236}Np little is known about excited structures and higher energy γ–rays …

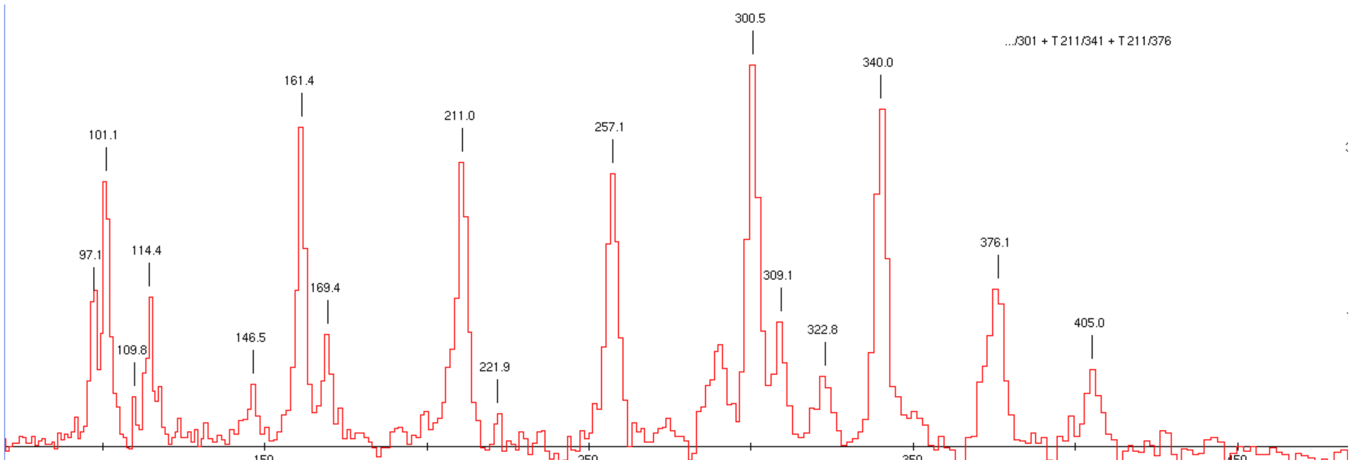
Data relevant to ^{237}Np(n,2n) CS –cont.

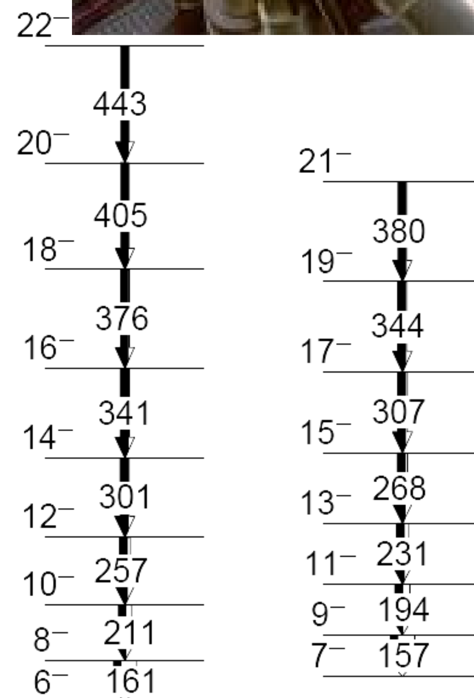
ANL, LLNL, University of Rochester, Kolkata collaboration

Experiment: **deep inelastic reactions** in conjunction with **GS & CHICO**: **^{116}Sn + ^{237}Np @ 800 MeV**/~20% above CB

✓information on excited structures, deformation, level structures & densities + interesting physics!

✓Doppler correction for projectile like and target like recoils with their velocities determined by CHICO





G. Mukherjee et al.

Identification

✓elemental: coincidences with characteristic X-rays

✓isotopic: cross-correlations with beam-like products (^{117}Sn)