Precision Cross-Section Measurements using Monoenergetic Photon and Neutron Beams at TUNL

Anton P. Tonchev, Calvin R. Howell, Elaine Kwan, Rajarshi Raut,

Gencho Rusev, and Werner Tornow

Duke University and TUNL, Durham, NC

John H. Kelley

North Carolina State University and TUNL, Raleigh, NC

In collaboration with:

Los Alamos National Laboratory, Lawrence Livermore National Laboratory

Research Objectives

- Perform new precision (*n,xnp*) cross-section measurements on actinides at E_n = (4 to 18) MeV to uncertainties smaller than 5 %.

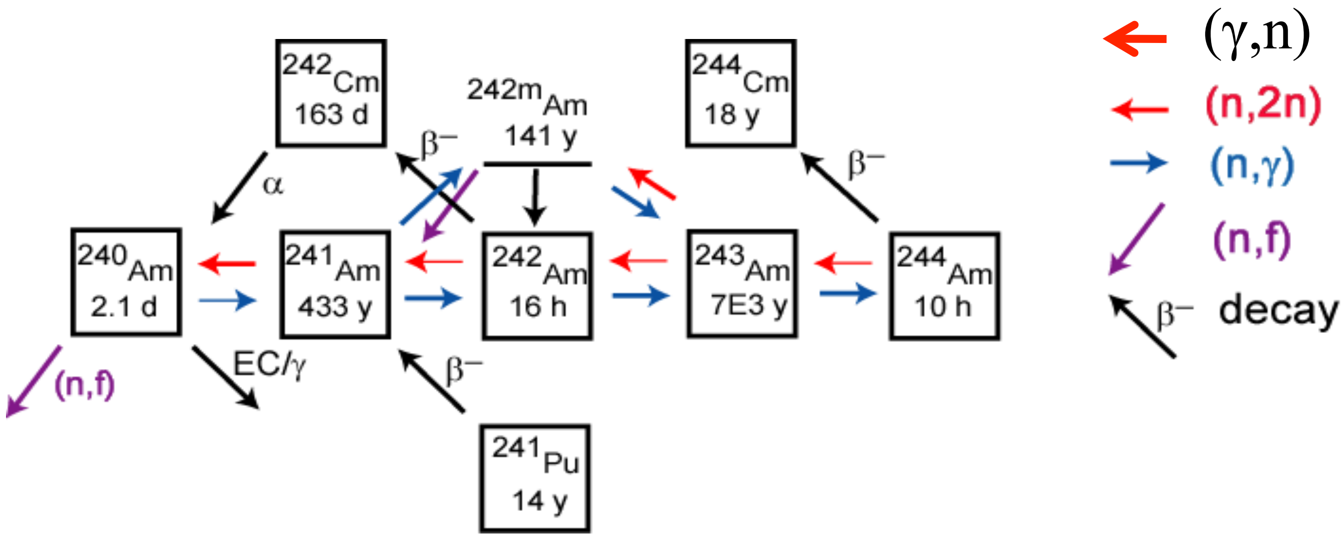
  Important for:

  - Basic science (testing nuclear models)
  - Nuclear astrophysics (*s*- and *p*-processes)
  - Nuclear energy and nuclear transmutation
  - Nuclear forensics
  - Accelerator application (radiation shielding)
  - Medical applications (isotope production)

- Improving our understanding of the actinide nuclear data through complementary (γ,*xn*) cross-section measurements.
- Techniques: direct measurements using monoenergetic beams
  - Photo- and Neutron Activation Analysis
  - In-beam (n,n') and (γ,γ') measurements

Americium Campaign

Nuclear reaction chain on Am isotopes



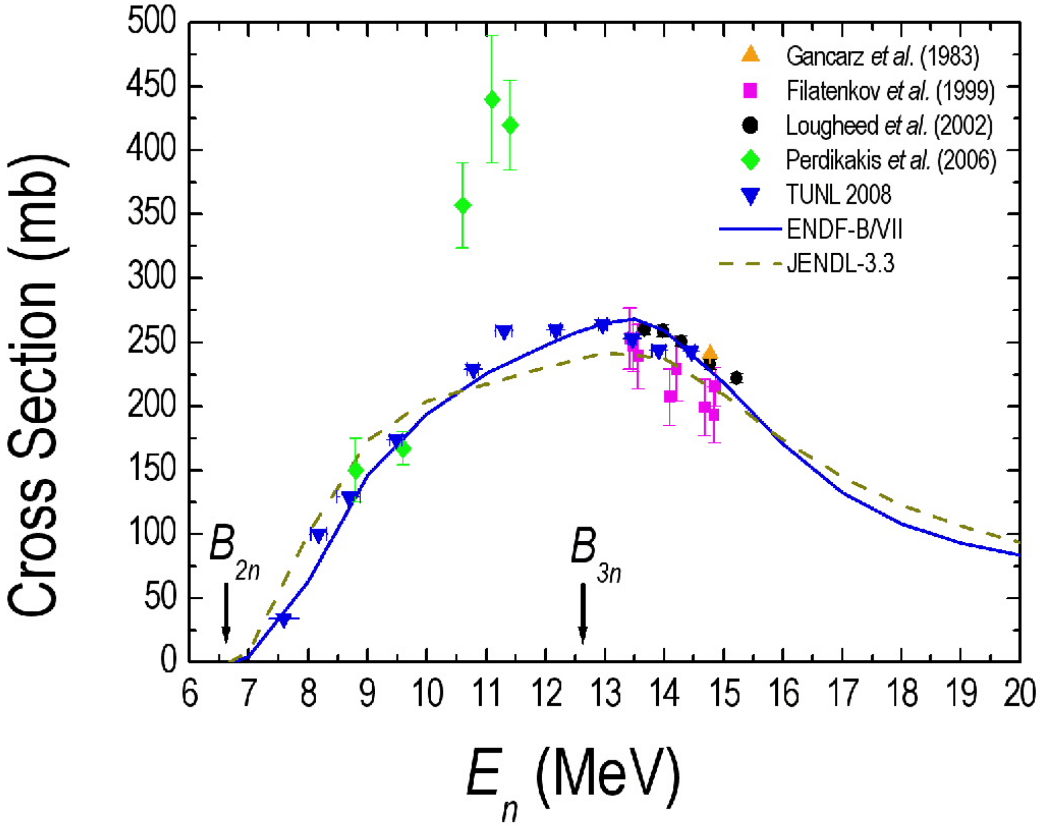
$\Delta A = {}^{240}Am/{}^{241}Am$

- **Sensitive to high-energy neutrons**
  $^{241}Am(n,2n)$ cross section needed

$^{242}Cm/^{241}Am$

- **Sensitive to low-energy n’s**
- **$^{241,243}Am(n,\gamma)$ cross section needed**

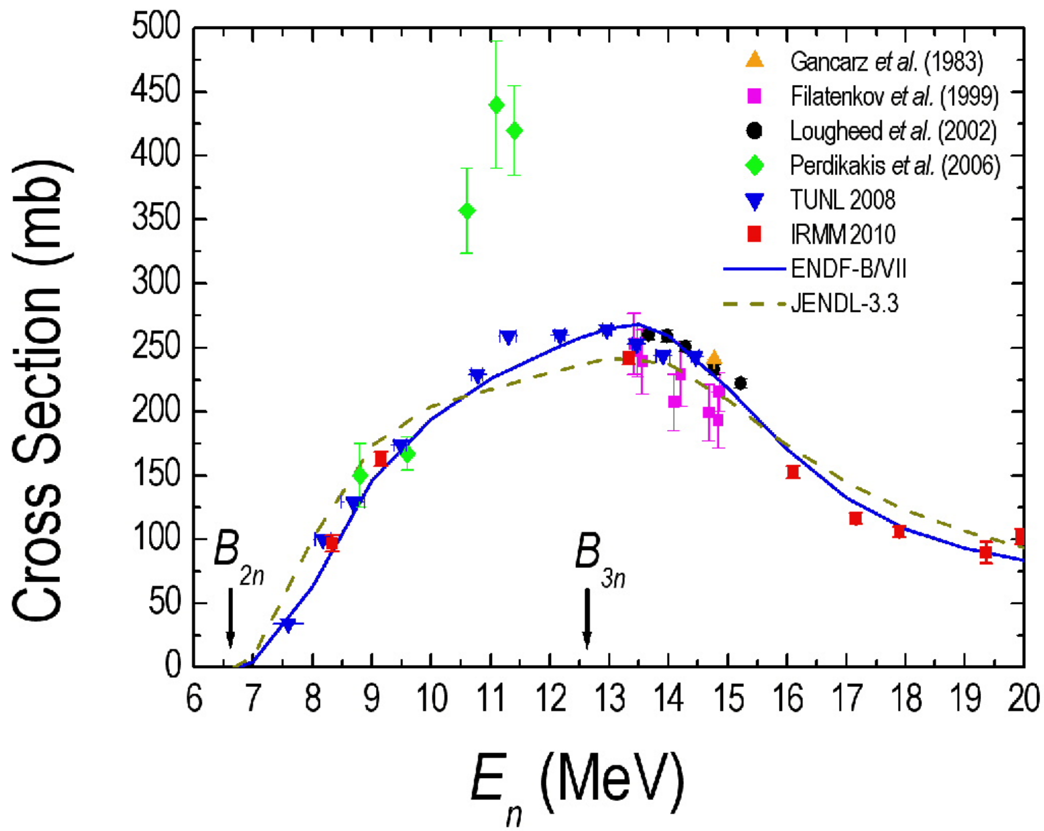
Experimental Results: $^{241}Am(n,2n)^{240}Am$



- CS saturation at E_n=12.5 MeV
- σ_{max} = 264 mb
- Rule out the CS data at E_n=11 MeV
- ENDF-B/VII agrees well with the TUNL data

A. P. Tonchev et al., PRC **77**, 054610 (2008)

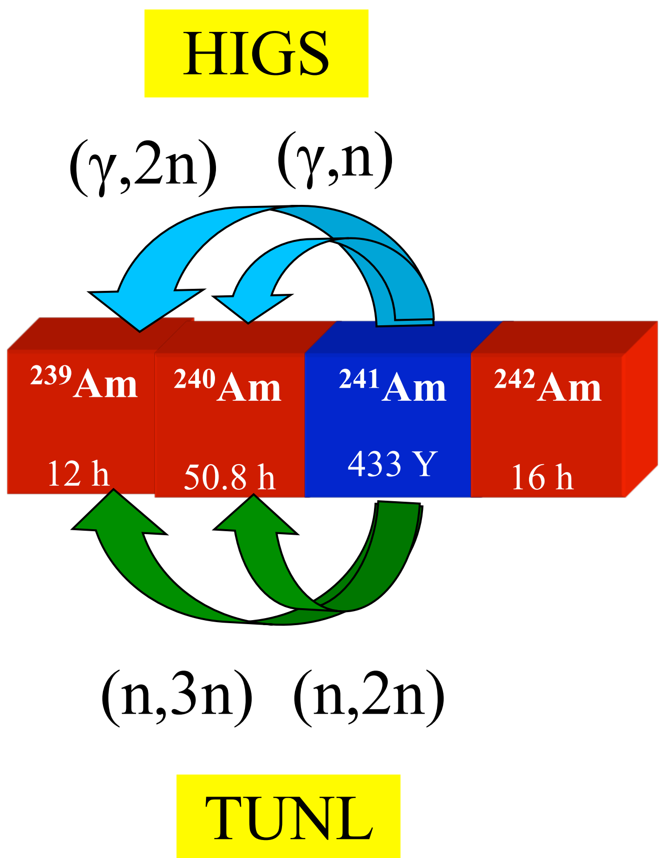
Experimental Results: $^{241}Am(n,2n)^{240}Am$



- CS saturation at E_n=12.5 MeV
- σ_{max} = 264 mb
- Rule out the CS data at E_n=11 MeV
- ENDF-B/VII agrees well with the TUNL data
- IRMM 2010 extended the $^{241}Am(n,2n)$ cross-section measurements up to E_n = 21 MeV
- IRMM data confirmed the earlier TUNL measurements

A. P. Tonchev et al., PRC **77**, 054610 (2008)
C. Sage et al., PRC **81**, 064604 (2010)

$^{241}Am(\gamma,n)^{240}Am$ Cross Section Measurements



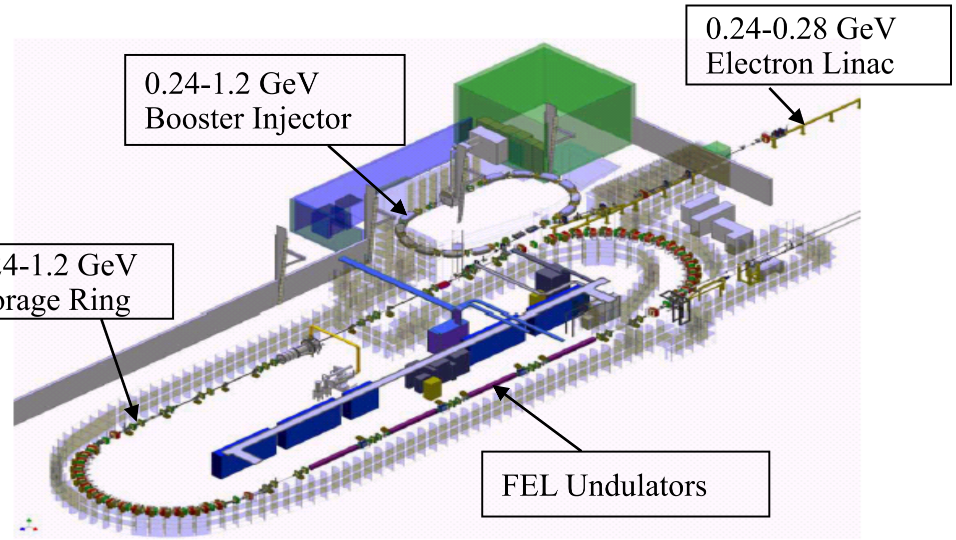
Motivation:

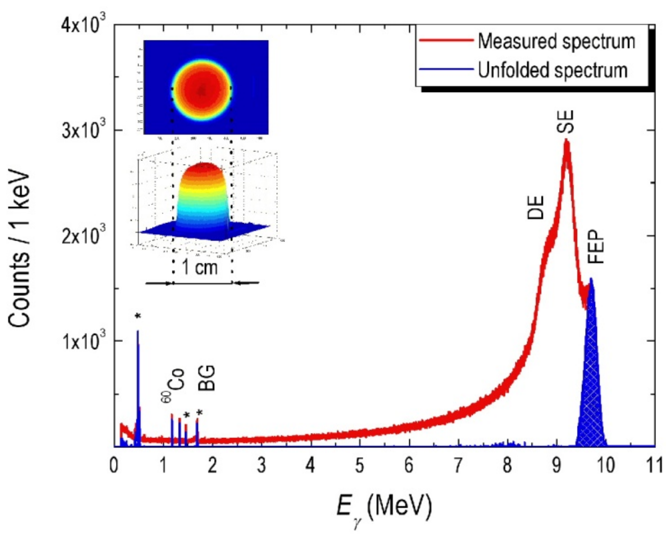
- $^{241}Am(\gamma,n)^{240}Am$ reaction is complementary to the $^{241}Am(n,2n)^{240}Am$
- ^{241}Am is the most important minor actinide
- It is one of the most harmful radioisotopes
- Data needed for advanced fuel cycle, new fast reactors, and transmutation studies

Goal: To measure the $^{241}Am(\gamma,n)$ cross section within (5 to10) % uncertainty for E_γ = 6.7 MeV to 20 MeV

High Intensity Gamma-Ray Source (HIGS)

Duke Free Electron Laser Laboratory





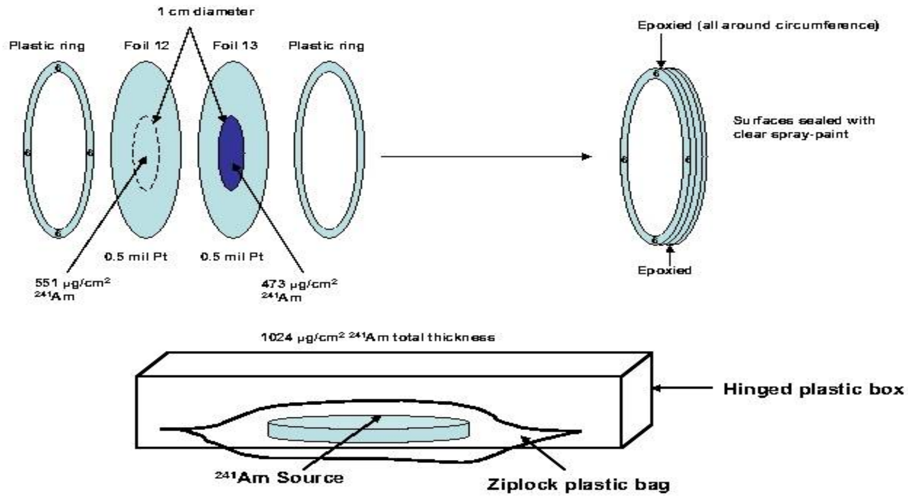
What makes HIGS unique?

1. Nearly monoenergetic beam $\Delta E/E = 0.01 – 0.03$
2. Tunable from (1 to100) MeV
3. Photon flux > 10^7 s^{-1} cm^{-2} on target

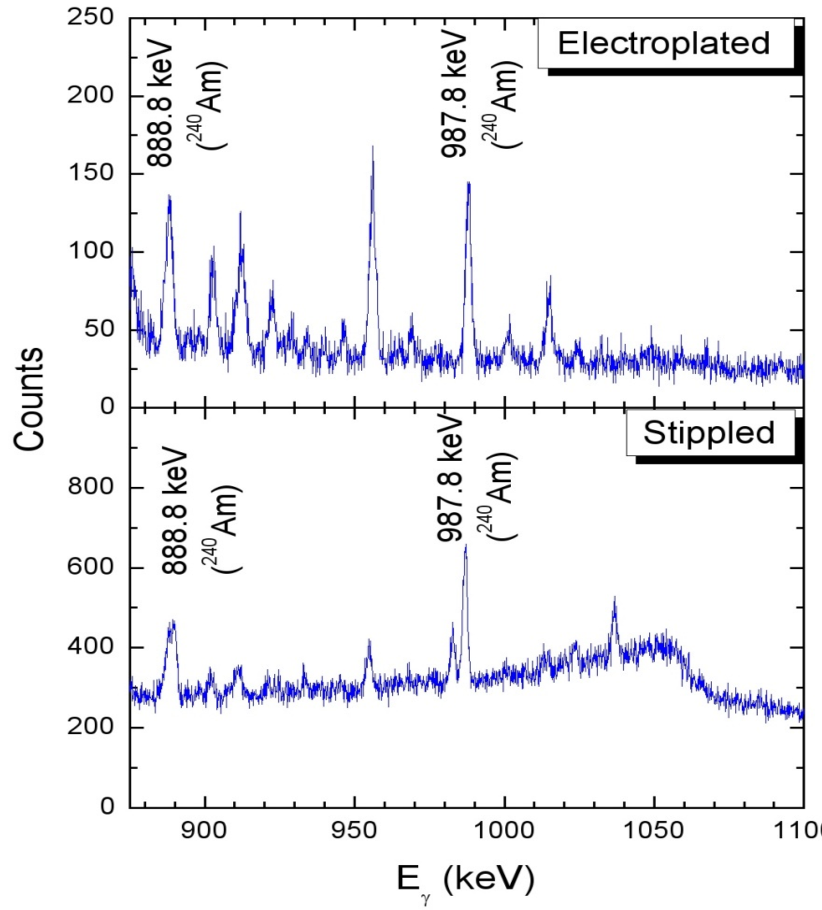
Advantages:

1. Intense peak in narrow energy window
2. Excite only at energy and levels of interest

^{241}Am Target Production

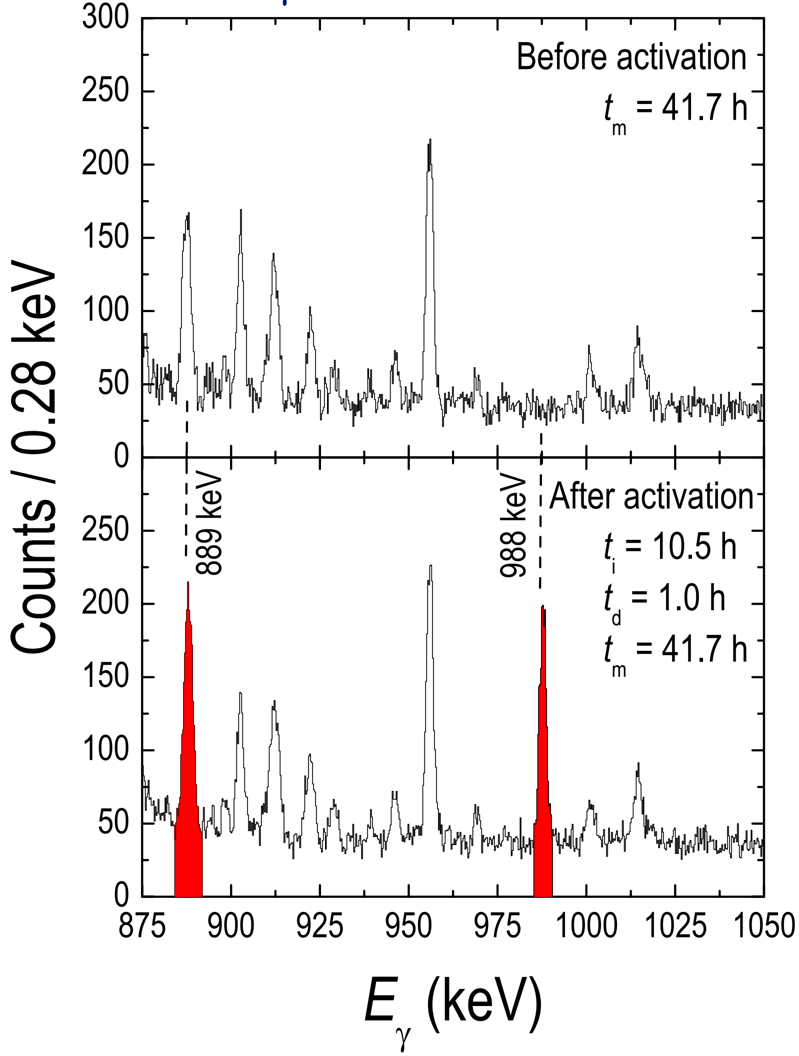


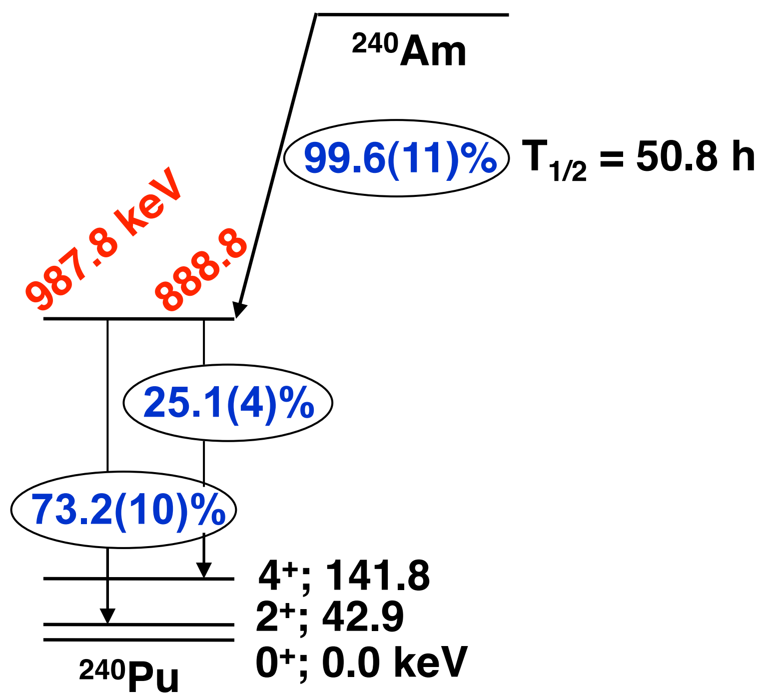
- Fabrication of ^{241}Am target (LANL and LLNL)
  - 3 stippled targets (Evelyn Bond/LANL)
  - 6 electroplated targets after additional chemical purification (Mark Stoyer/LLNL)
- ~ 2% statistical uncertainties
- Pk/bg ratio improved from 2:1 to 6:1



Americium Signature

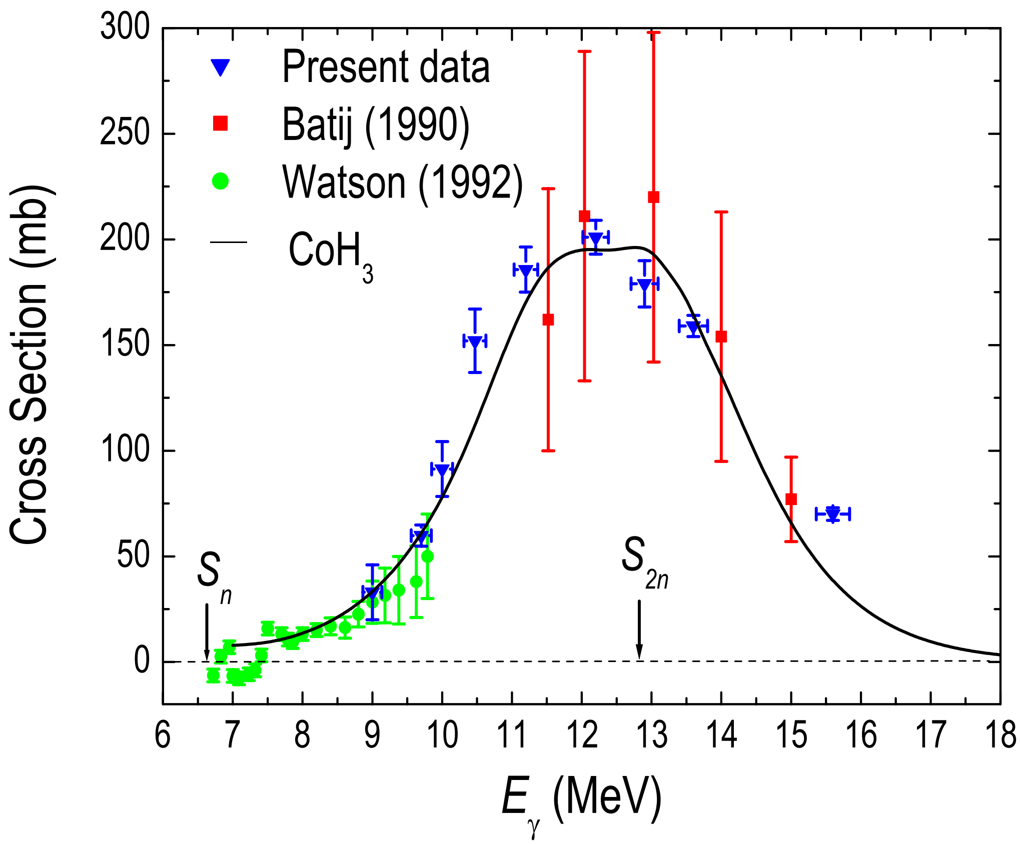
$E_\gamma = 13.6$ MeV





- Measured 500-1500 γ events after 12 h of irradiation and two days of counting time
- Measured nine energies in 2009

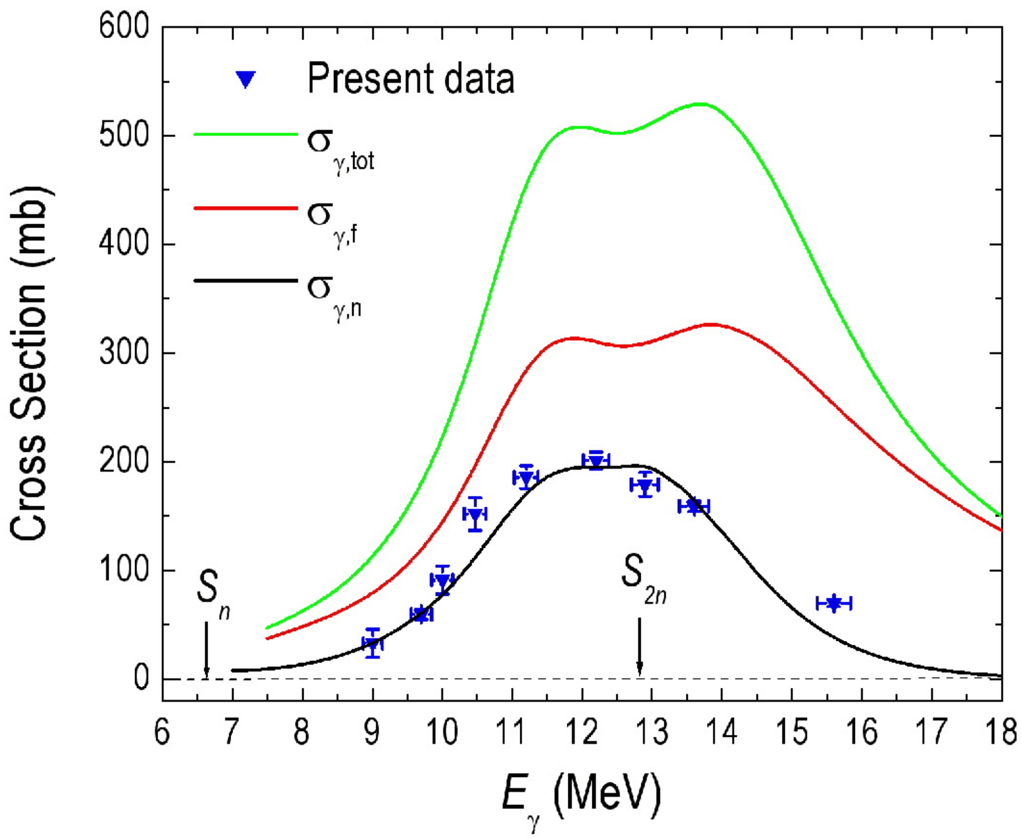
Measurements of the $^{241}Am(\gamma,n)$ Reaction Cross Section at HIGS



- The cross section saturates at $E_n = 12.5$ MeV
- $\sigma_{max} = 200$ mb at E_n=12.5 MeV
- Present data supports a single Lorentzian curve

A.P. Tonchev *et al*. AIP **1099**, 820 (2009)
A.P. Tonchev *et al.* Accepted to PRC (2010)

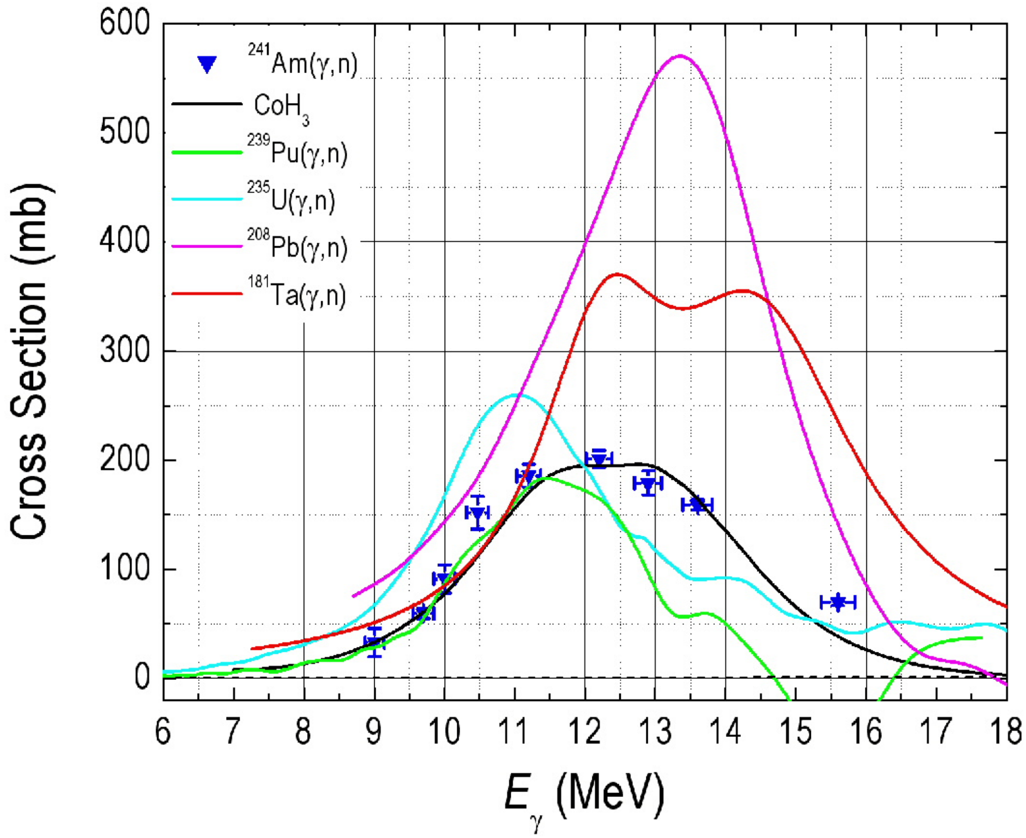
Measurements of the ^{241}Am(γ,n) Reaction Cross Section at HIGS



- The cross section saturates at $E_n = 12.5$ MeV
- $\sigma_{max} = 200$ mb at $E_n=12.5$ MeV
- Present data supports a single Lorentzian curve

A.P. Tonchev *et al*. AIP **1099**, 820 (2009)
A.P. Tonchev *et al.* Accepted to PRC (2010)

Measurements of the ^{241}Am(γ,n) Reaction Cross Section at HIGS



- The cross section saturates at $E_n = 12.5$ MeV
- $\sigma_{max} = 200$ mb at E_n=12.5 MeV
- Present data supports a single Lorentzian curve
- The ^{241}Am(γ,n) cross section is consistent with other fissile nuclei like ^{235}U and ^{239}Pu
- HIGS make possible cross section measurements using radioactive targets with μg quantities.

A.P. Tonchev *et al*. AIP **1099**, 820 (2009)
A.P. Tonchev *et al.* Accepted to PRC (2010)

Cross-Section Measurements on Ga and As

Importance: semiconductor materials; nuclear reaction mechanism studies; testing the statistical model.

Astrophysical relevance: ^{73,75}As are important *p*-process nuclei

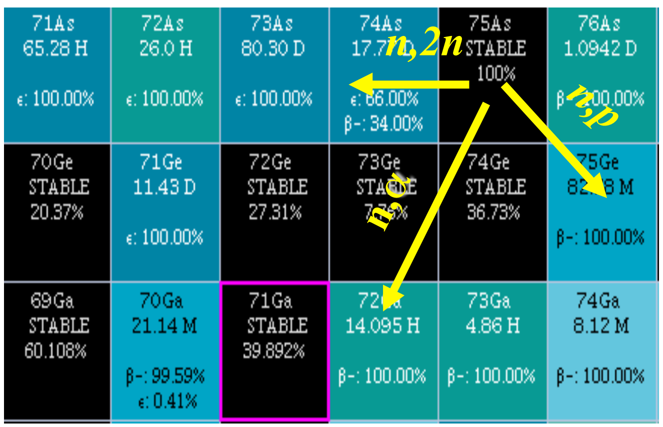


TABLE I: Neutron induced reactions on GaAs and monitor foils measured in the present work.

Reaction Channel	Product Half-life	Q-value (keV)	E_γ (keV)	I_γ (%)
		GaAs Reactions		
^{69}Ga$(n,2n)^{68}$Ga	67.71 m	-10312.95	1077.34	3.22
^{69}Ga$(n,p)^{69m}$Zn	13.76 h	-127.44	438.634	94.77
^{71}Ga$(n,p)^{71m}$Zn	3.96 h	-2031.0	386.28	93
^{75}As$(n,2n)^{74}$As	17.77 d	-10243.76	634.78	15.4
^{75}As$(n,p)^{75}$Ge	82.78 m	-393.63	264.6	11.4
		Monitor Reactions		
^{197}Au$(n,2n)^{196}$Au	6.1669 d	-8072.39	355.73	0.87
^{27}Al$(n,\alpha)^{24}$Na	14.997 h	-3132.14	1368.626	99.9936

These cross section measurements on stable Ga and As targets are important steps to understand the (n,Xnγ) measurements on radioactive ^{73}As target.

^{73}As target produced at LANL

^{nat}Ge(p,xn) production reaction at IPF (LANSCE)

E_p = 100 MeV, I_p = 250 μA

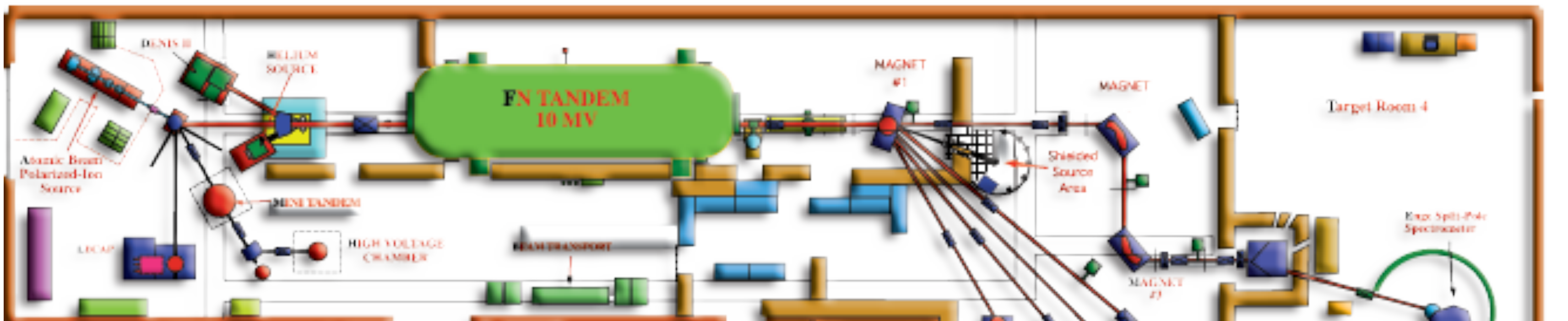
500 mCi of ^{73}As

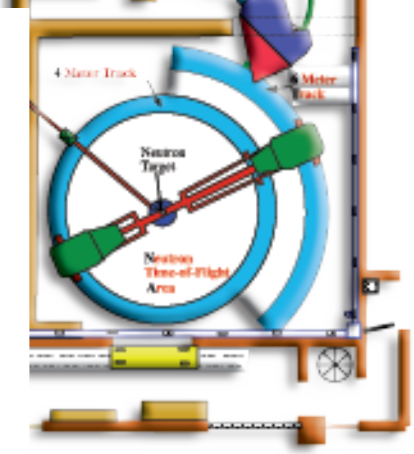
Activation Measurements at TUNL

DENIS

FN TANDEM 10MV

Shielded neutron source area





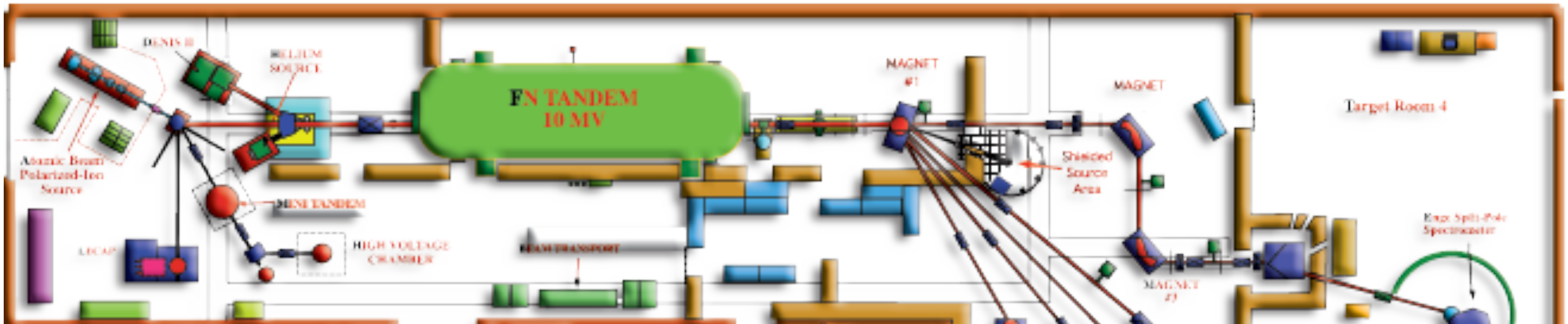
- $^{2}H(d,n)^{3}He$ neutron source
- $I_d = (1\text{-}3)\ \mu A$
- Tunable from 4 to 18 MeV
- Gas pressure 3 atm
- Flux on target $(10^7 - 10^8)\ cm^{-2}\ s^{-1}$
- Energy spread $dE/E = 0.02$ to 0.04

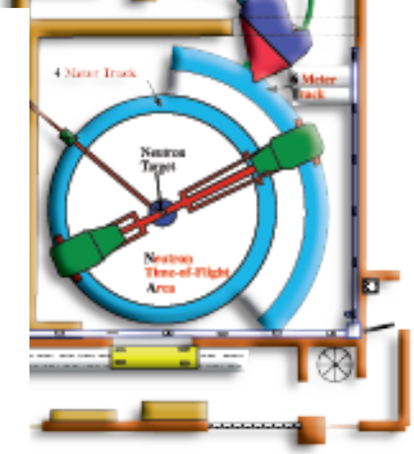
Activation Measurements at TUNL

DENIS

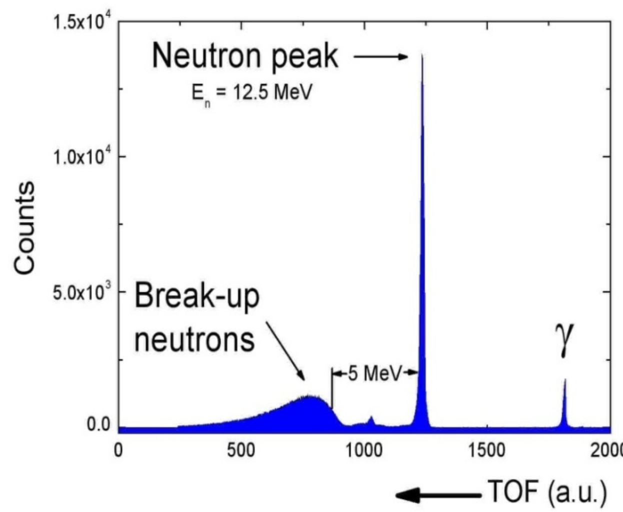
FN TANDEM 10MV

Shielded neutron source area

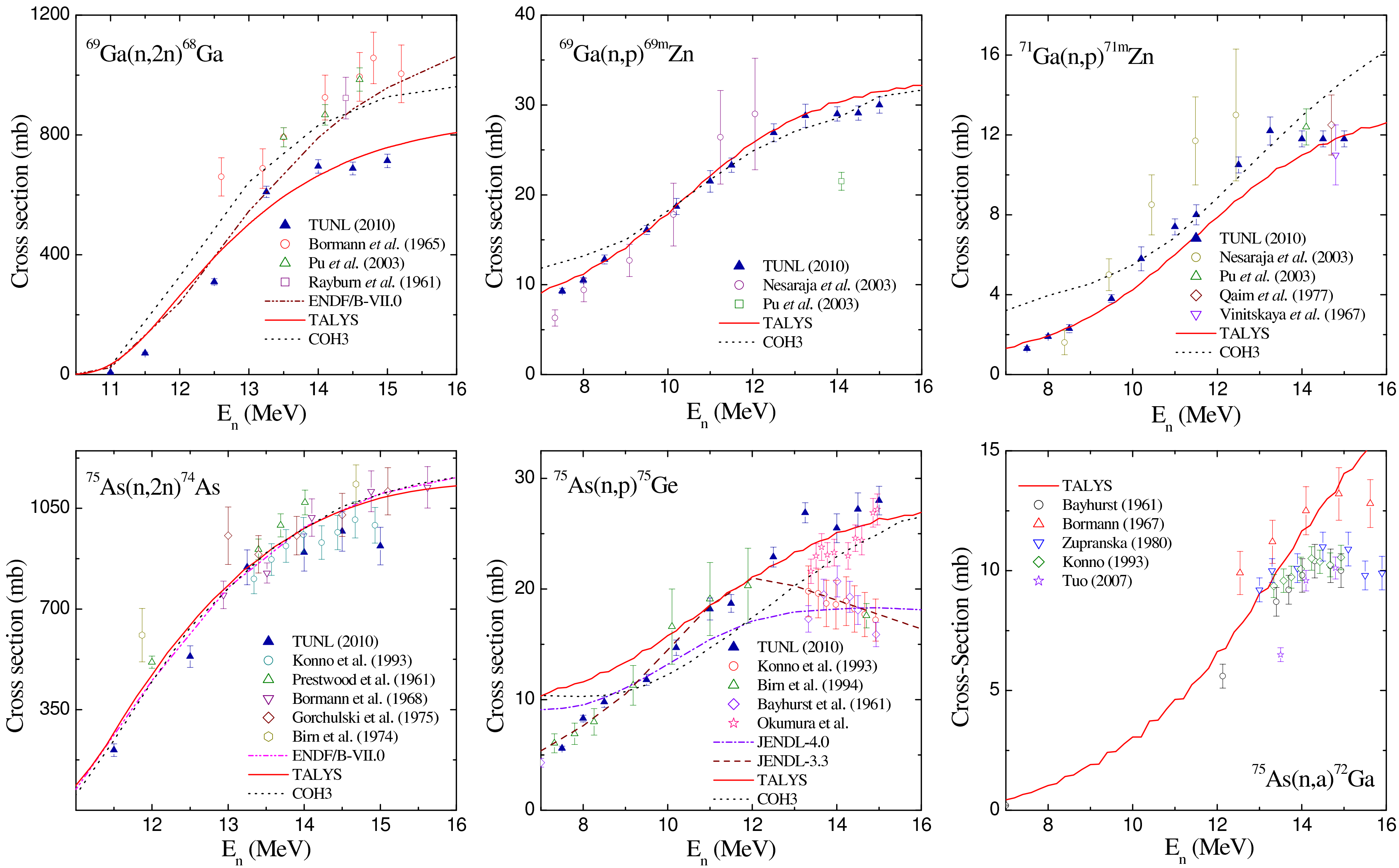




- ❑ $^{2}H(d,n)^{3}He$ neutron source
- ❑ I_d = (1-3) μA
- ❑ Tunable from 4 to 18 MeV
- ❑ Gas pressure 3 atm
- ❑ Flux on target (10^7 – 10^8) $cm^{-2}\ s^{-1}$
- ❑ Energy spread dE/E = 0.02 to 0.04

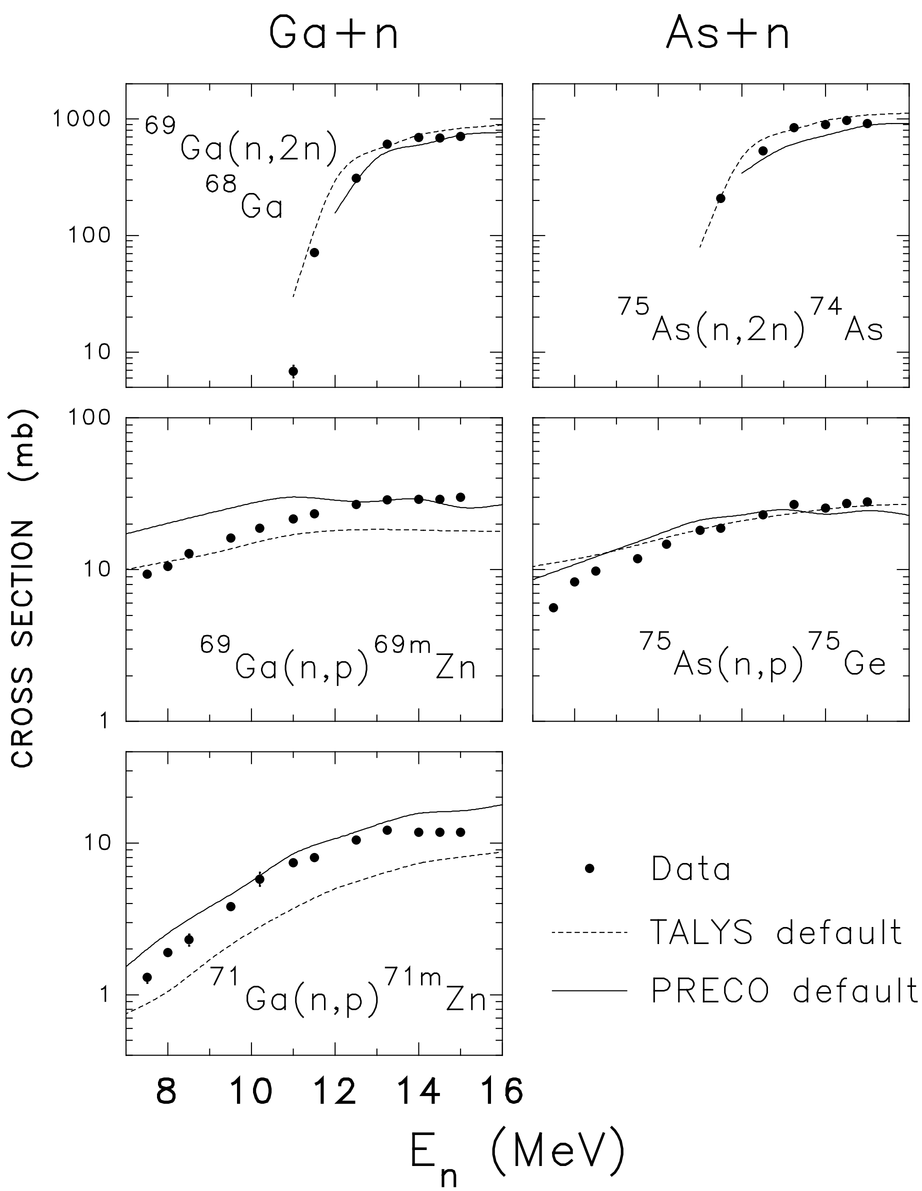


Neutron Induced Reactions on Ga and As



R. Raut et al. Prepared for publication in PRC

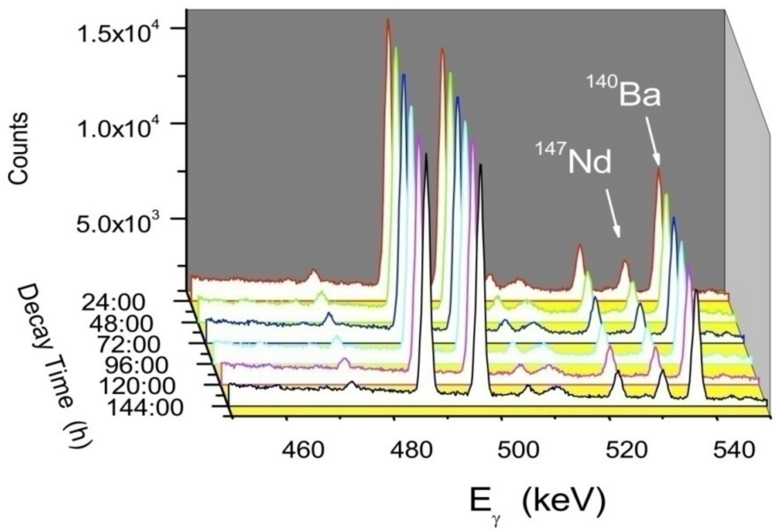
Neutron Induced Reactions on Ga and As

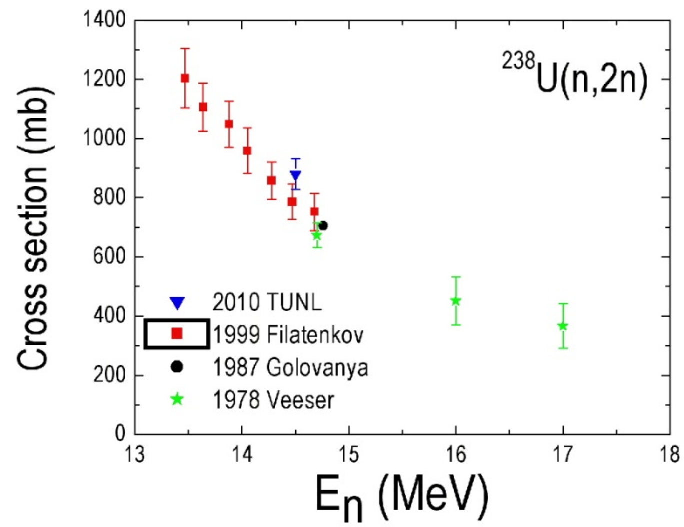


Shape of the ^{147}Nd FPY at low incident neutron energy

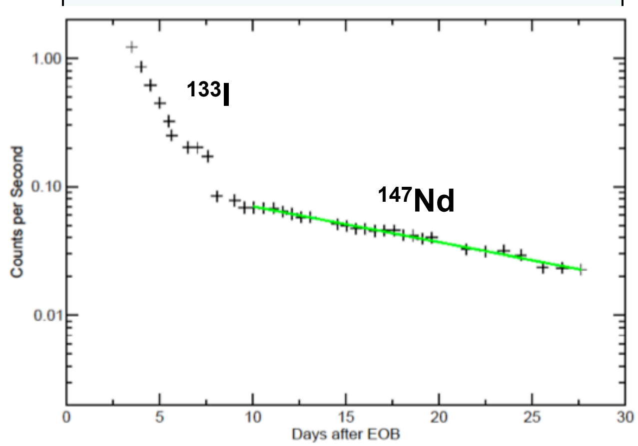
- A slope to the ^{147}Nd FPY has been reported at low incident neutron energy
- (3.7% per MeV at 1.5 MeV, Chadwick, et al.)
- The slope to the ^{147}Nd FPY at low energies is determined from
  - Critical assembly measurements
  - Fast reactor measurements
- Critical assembly measurements require detailed knowledge of the neutron flux to develop an "average" neutron energy
  - Measurements with monoenergetic neutrons avoid this issue
- Proposal: Measure FP gamma-ray yields relative to ^{140}Ba reference
- Report:
  - FP Yield Ratios (< 1%), consistency, Thermal to 14 MeV E_n
  - FP Energy Dependence ^{147}Nd (< 1.5%), Thermal to 8 MeV E_n
  - FP Yields (8 – 10%)
  - Focus on ^{147}Nd, ^{99}Mo, ^{95}Zr

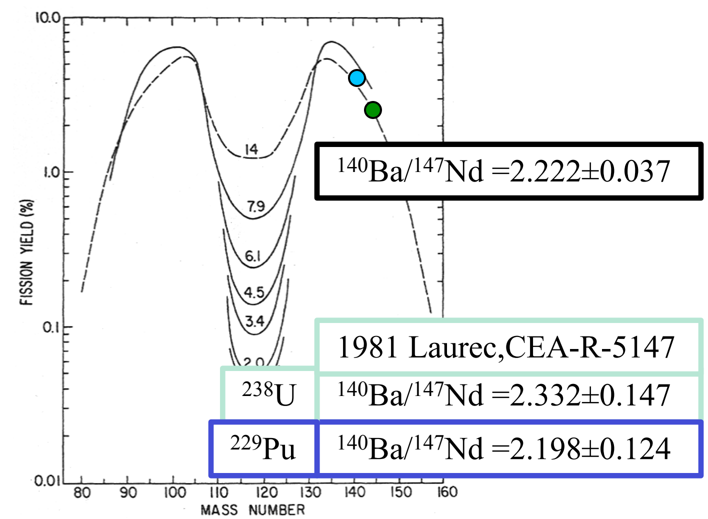
Energy dependence of ^{147}Nd (and other) products in the ^{239}Pu (n,f) reaction - Thermal to 16 MeV





^{147}Nd + ^{133}I 531-keV Complex f(t)





A.P. Tonchev et al., 2010 TUNL Progress Report.

Summary

- A total of nine measurements have been performed at HIGS for the $^{241}Am(\gamma,n)^{240}Am$ reaction from E_γ= 9.6 MeV to 16.0 MeV.
- Precision cross-section measurements on Ga and As targets from 7 MeV to 15 MeV.
- First measurements of the neutron induced fission fragment yields at TUNL.

Summary

TUNL

Duke:

A.S. Crowell
B. Fallin
C.R. Howell
A. Hutcheson
E. Kwan
R. Raut
G. Rusev
A.P. Tonchev
W. Tornow

Univ. of N. Carolina:

S. Hammond

N. Carolina State:

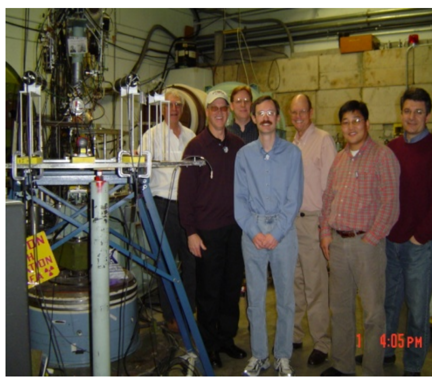
J.H. Kelley
D. Dashdorj*
C. Huibregtse

Livermore

J.A. Becker
D. Dashdorj*
J. Kenneally
R.A. Macri
D. Shaughnessy
M.A. Stoyer
Ch.Y. Wu

Los Alamos

E. Bond
J. FitzPatrick
R.S. Rundberg
A. Slemmons
W. Taylor
D.J. Vieira
J.B. Wilhelmy



This research was sponsored in part by the Office of Science of the US Department of Energy (DOE): DE-FG02-97ER41033 and National Nuclear Security Administration grants: DE-PS52-08NA28920 and DE-FG52-09NA29448.

Sources of Uncertainties

Source of uncertainty	Magnitude (%)	
	Am	Monitors
Relative uncertainties		
Count rate (statistics and background)	2-5	<1
γ-ray absorption in sample	<1	<0.1
Sample mass (γ +α counting)	2	<1
Total relative uncertainties	5.3	1.7
Absolute uncertainties		
Detector efficiency	2	2
γ-ray emission probability	1.1	<1
Half-life	<1	<1
Coincidence summing	<2	<1
Absolute source activity	1	
Gamma flux fluctuation	<1.0	
Reference monitor cross section		3-5*
Total uncertainty	4.6	5.7

* NNDC web side

Experimental Results

Neutron Energy (MeV)	Cross Section (mb)				
	$^{69}Ga(n,2n)^{68}Ga$	$^{69}Ga(n,p)^{69m}Zn$	$^{71}Ga(n,p)^{71m}Zn$	$^{75}As(n,2n)^{74}As$	$^{75}As(n,p)^{75}Ge$
7.5±0.2		9.3±0.3	1.3±0.1		5.6±0.2
8.0±0.1		10.5±0.3	1.9±0.1		8.3±0.3
8.5±0.2		12.8±0.5	2.3±0.2		9.8±0.5
9.5±0.1		16.1±0.5	3.8±0.2		11.8±0.5
10.2±0.1		18.7±0.9	5.8±0.6		14.7±0.7
11.0±0.1	6.9±0.8	21.5±1.2	7.4±0.4		18.2±1.0
11.5±0.1	71.6±3.5	23.3±0.8	8.0±0.5	209.3±21.9	18.7±0.8
12.5±0.1	309.7±10.5	26.9±1.0	10.5±0.4	534.6±37.6	22.9±0.9
13.25±0.1	716.1±21.2	28.8±1.3	12.2±0.7	845.0±60.7	26.9±0.9
14.0±0.1	694.9±22.3	29.0±0.8	11.8±0.4	896.5±64.4	25.5±1.3
14.5±0.1	688.1±21.6	29.1±0.8	11.8±0.4	969.7±68.9	27.2±1.5
15.0±0.1	713.5±22.4	30.0±0.9	11.8±0.4	918.6±65.5	28.0±1.3

TABLE II: Sources and approximate magnitudes (in %) of the uncertainties in the present cross section measurements.

Uncertainty	Magnitude (%)
Statistics	1-2[a]
Sample mass	< 1
Detector efficiency	2-3
Branching ratio	≤ 1
Product half-life	≤ 1
Monitor cross section	1-4 (Al) 1-4 (Au)[b]
Low energy neutrons	< 1
Total[c]	3-5

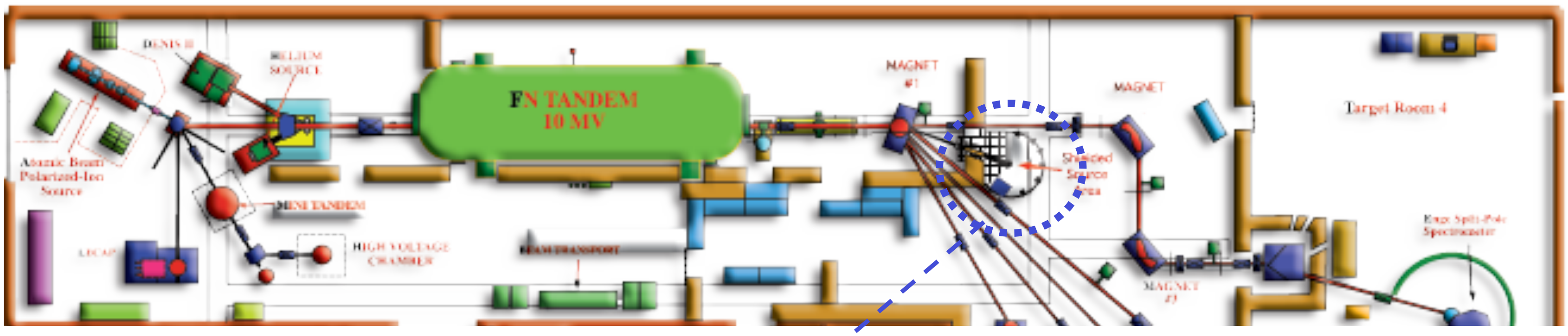
R. Raut *et. al.* Prepared for PRC (2010)

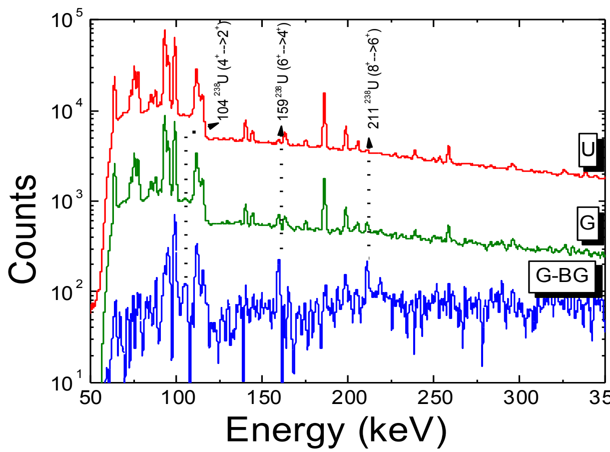
In-Beam Measurements at TUNL

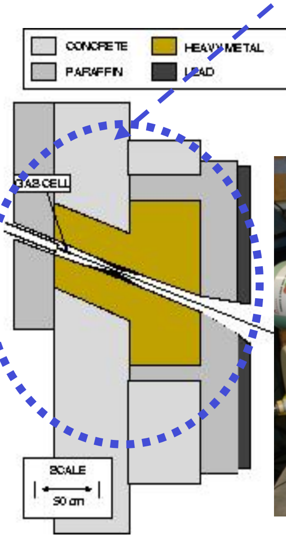
DENIS

FN TANDEM 10MV

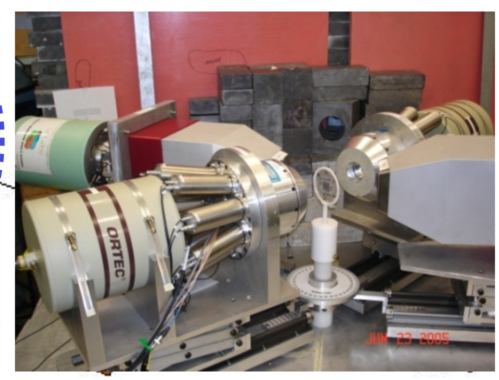
Shielded neutron source area







Experimental set-up



- $^{2}H(d,n)^{3}He$ neutron source
- I_d = (1-3) μA
- Tunable from 4 to 18 MeV
- Pulsed at 2.5 MHz
- Flux on target 5×10^{4} s^{-1}

A. Hutcheson et al., PRC **80**, 014603 (2009)

Cross Section Measurements using the Monoenergetic HIGS beams

Activation technique: separate irradiation from measurement time

⇒ low background, high sensitivities, highly accurate residual identification

Relative measurements: ^{197}Au(γ,n) and ^{58}Ni(γ,n) as monitor reactions

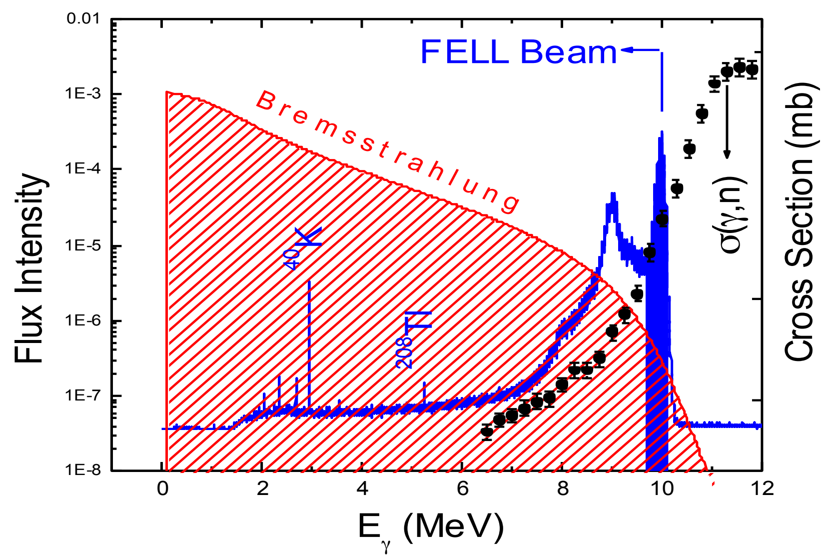
$$S = \frac{m\theta I_\gamma \varepsilon_\gamma}{\lambda}(1-e^{-\lambda t_i})e^{-\lambda t_d}(1-e^{-\lambda t_m})\prod_k c_k \int_{E_{th}}^{E\max}\Phi(E,E_{\max})\sigma(E)dE$$

Fluctuations in the gamma flux

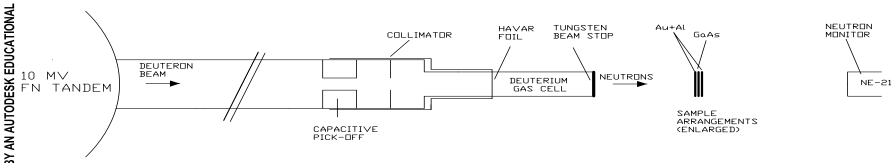
$$C_{flux} = \frac{\overline{\Phi}(1-e^{-\lambda t_i})}{\sum_{i=1}^{n}\Phi_i(1-e^{-\lambda\Delta t})e^{-\lambda(n-i)\Delta t}}$$

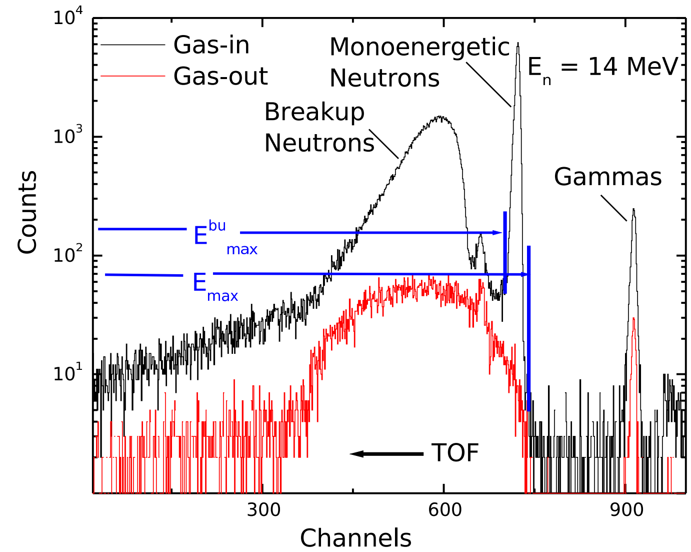
Mean gamma energy

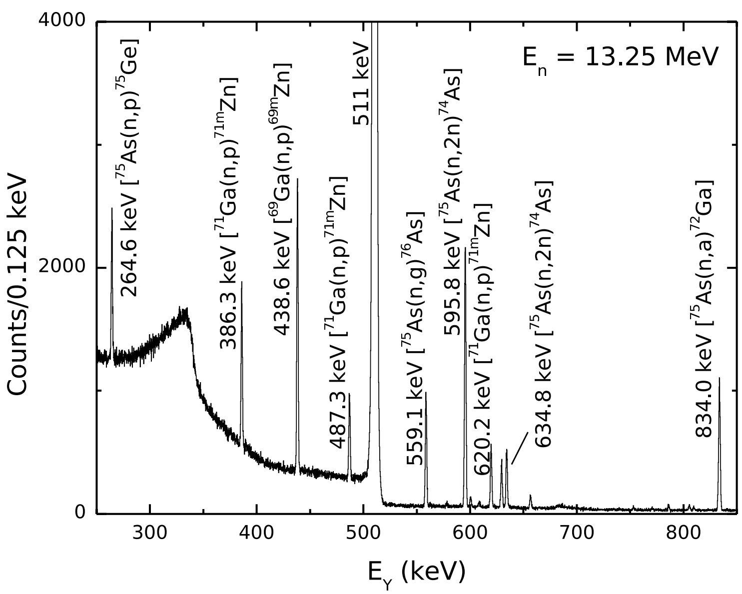
$$\overline{E} = \frac{\int_0^{\theta_{\max}} E(\theta)\sigma(E,\theta)\theta d\theta}{\int_0^{\theta_{\max}}\sigma(E,\theta)\theta d\theta}$$



Summary


10 MV
FN TANDEM
DEUTERON BEAM
COLLIMATOR
CAPACITIVE PICK-OFF
HAVAR FOIL
TUNGSTEN BEAM STOP
DEUTERIUM GAS CELL
NEUTRONS
Au+Al
GaAs
SAMPLE ARRANGEMENTS (ENLARGED)
NEUTRON MONITOR
NE-213


Gas-in
Gas-out
Monoenergetic Neutrons
E_n = 14 MeV
Breakup Neutrons
Gammas
E^bu_max
E_max
TOF
Counts
Channels


E_n = 13.25 MeV
264.6 keV [75As(n,p)75Ge]
386.3 keV [71Ga(n,p)71mZn]
438.6 keV [69Ga(n,p)69mZn]
487.3 keV [71Ga(n,p)71mZn]
511 keV
559.1 keV [75As(n,g)76As]
595.8 keV [75As(n,2n)74As]
620.2 keV [71Ga(n,p)71mZn]
634.8 keV [75As(n,2n)74As]
834.0 keV [75As(n,a)72Ga]
Counts/0.125 keV
E_Y (keV)