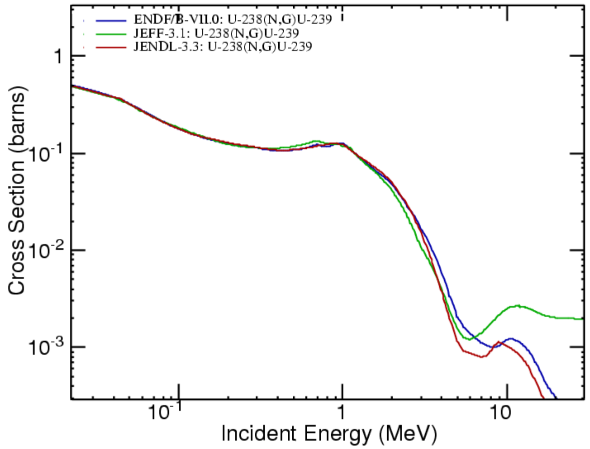
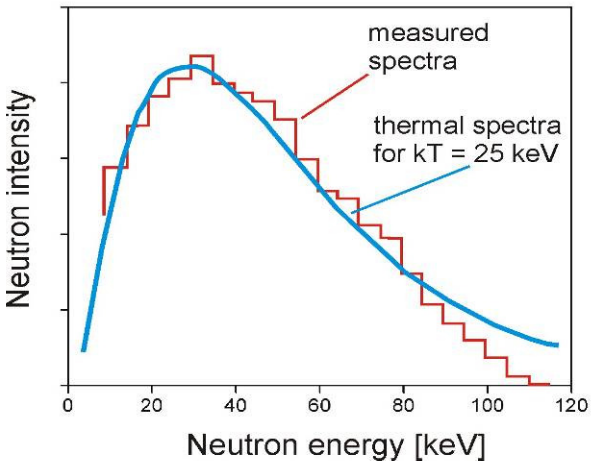
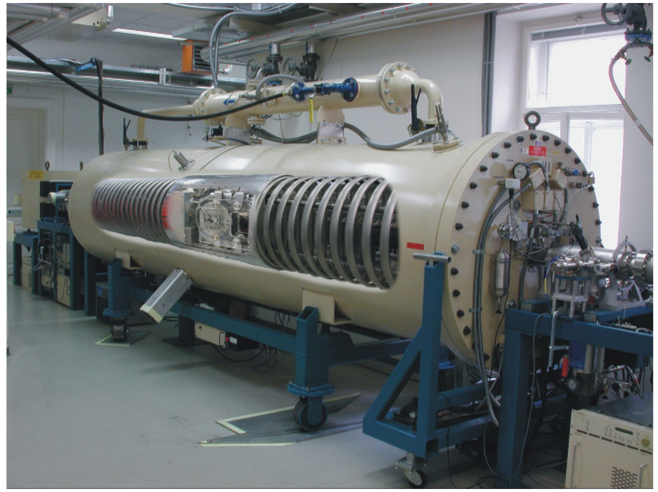
Neutron Capture Studies of ^{235}U and ^{238}U via AMS



^{238}U(n,γ)^{239}U – eval.

FZK- 25 keV





VERA

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Content

- ***Method: combination of activation and AMS***
- ***neutron capture cross section of $^{235,238}U$ in the keV region – a prime example for applying AMS***
- ***AMS at VERA – overview***
- ***first results***
- ***other nuclides for neutron reaction data via AMS - outlook***

Activation (I) → AMS-Measurement (II) fusion / fission / ADS products

(1) Recent neutron irradiations:

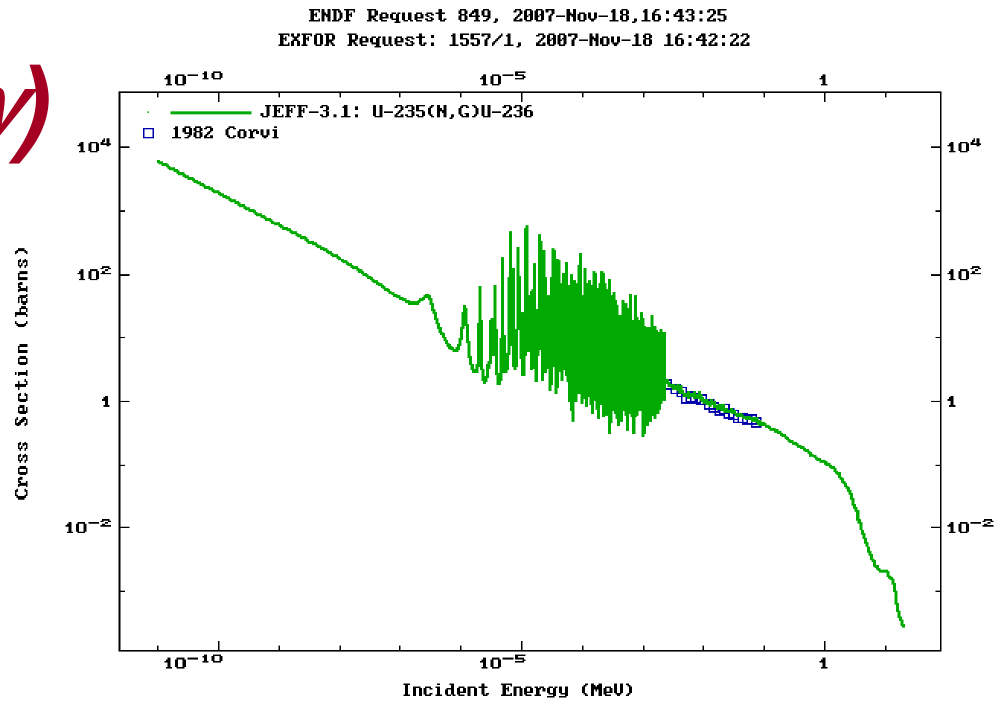
- **FZ Karlsruhe:** 3.7 MV Van de Graaff: $^{7}Li(p,n)^{7}Be$: 25 keV Maxwell-Boltzmann, 120 keV, 180 keV, 500 keV, ...)
- **IRMM Geel:** 7 MV Van de Graaff: ($T(d,n)^{4}He$: E_n = 14 – 20 MeV)
- **FZ Dresden/Rossendorf:** 300 kV ($T(d,n)^{4}He$: E_n = 13.3 – 14.9 MeV)
- **Atominstitut Vienna:** research reactor, thermal neutrons
- **IKI Budapest:** research reactor, thermal neutrons; cold neutrons

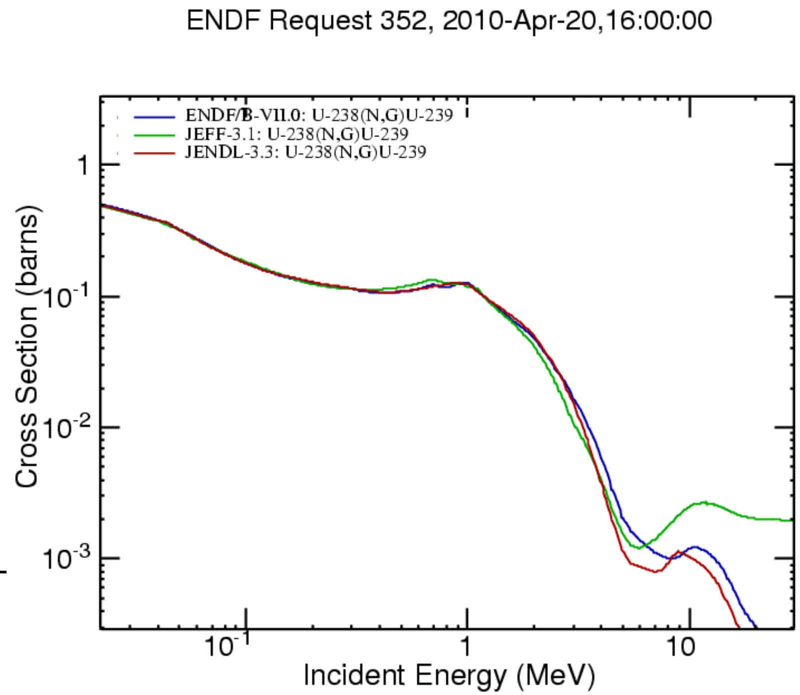
(2) AMS measurements e.g. at VERA: from Be to ^{238}U, Pu

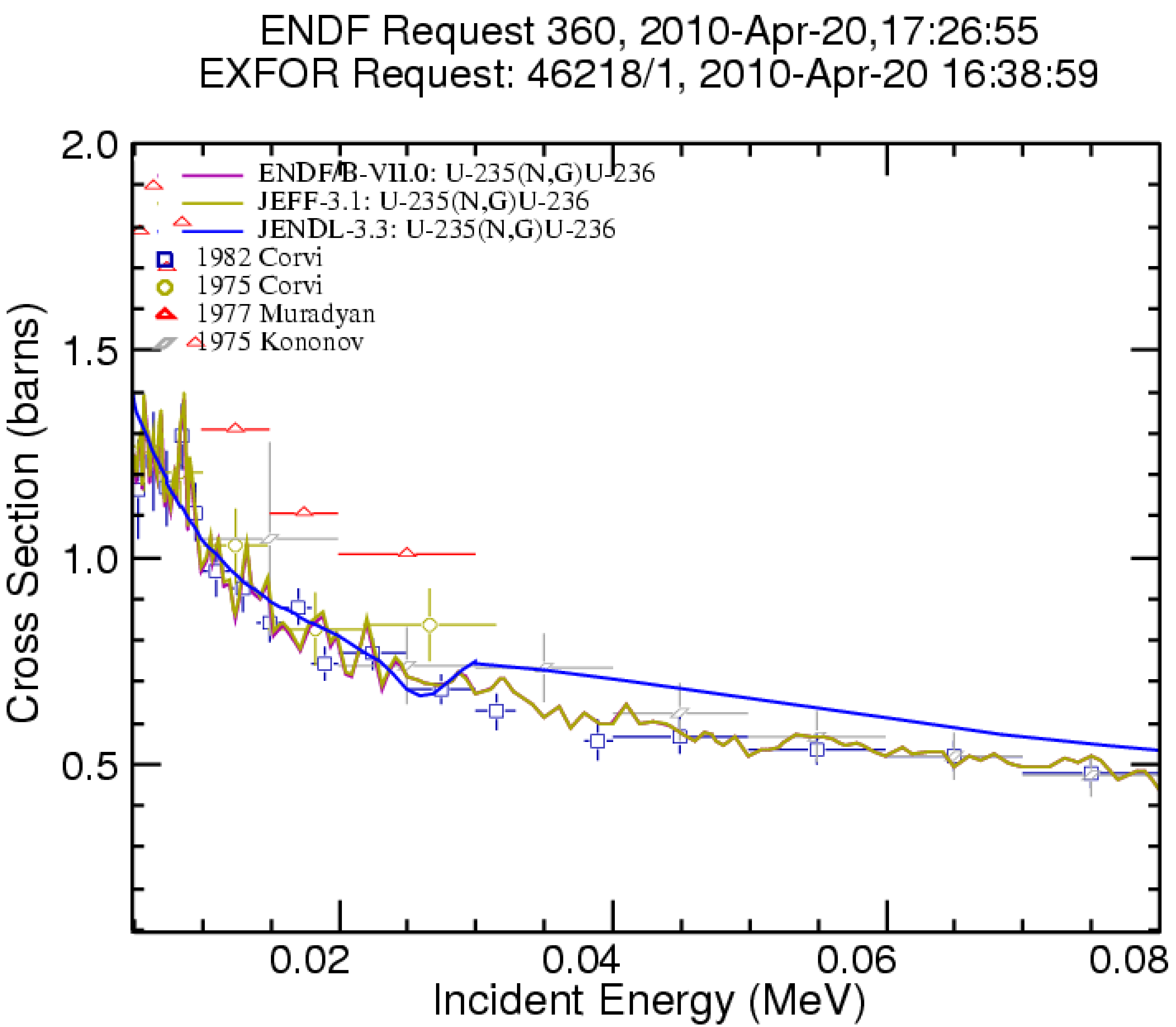
Motivation: $^{235/8}$U(n,γ)

- ratio of capture to fission of the fissile isotopes: capture on ^{235}U
- ^{235}U: existing data via TOF and detections of prompt γ-rays;
- ^{238}U also decay of ^{239}Np
- since 1980 only 1 measurement in the keV region for ^{235}U (1982 Corvi et al.)
- Activation + AMS is an independent method; no influence from fission channel
- may serve as proof-of-principle measurement - to be extended to other reactions

! discrepancy of evaluations !





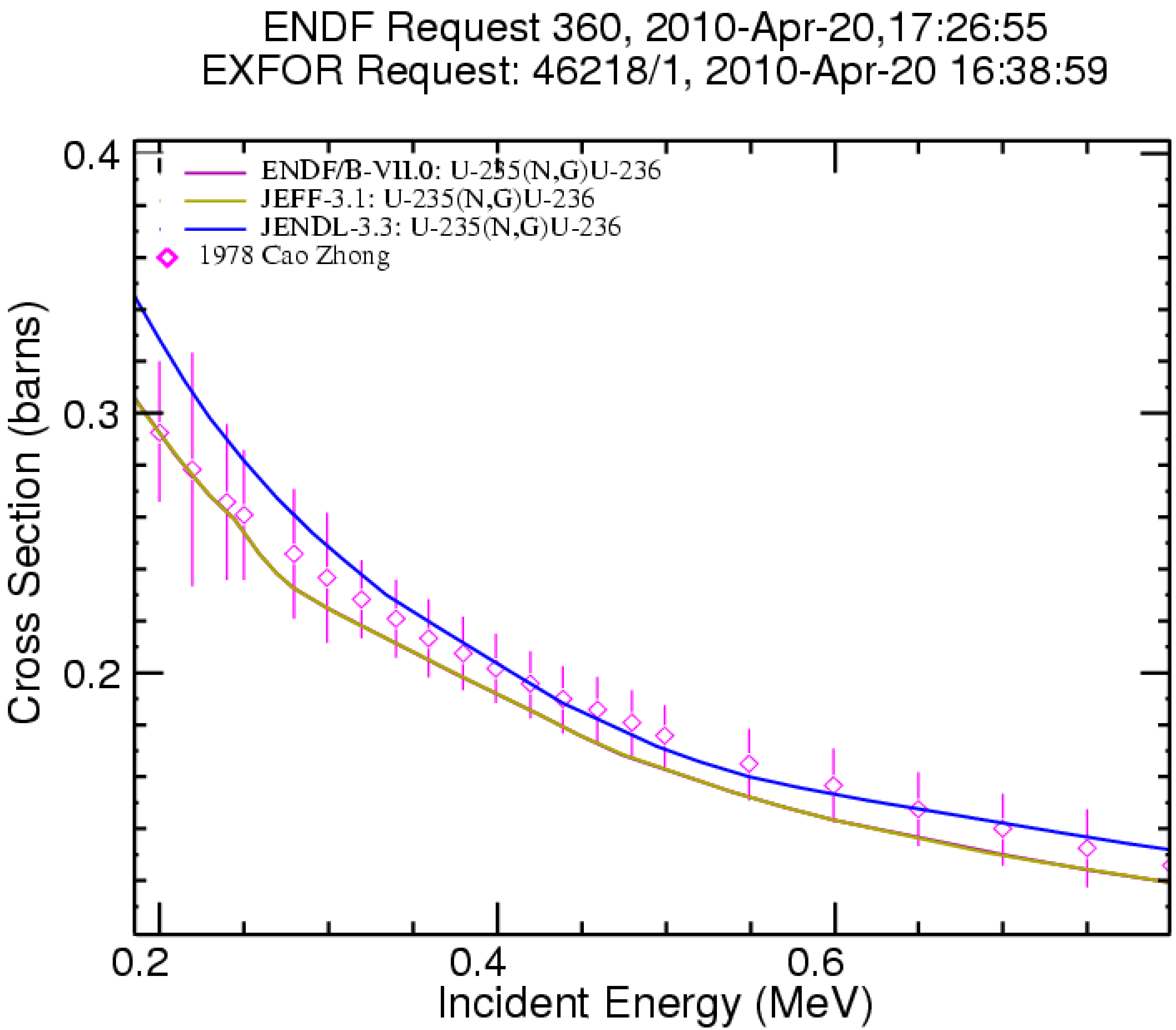


$^{235}U(n,\gamma)^{236}U$

EXFOR database

ENDF = JEFF

5 – 80 keV



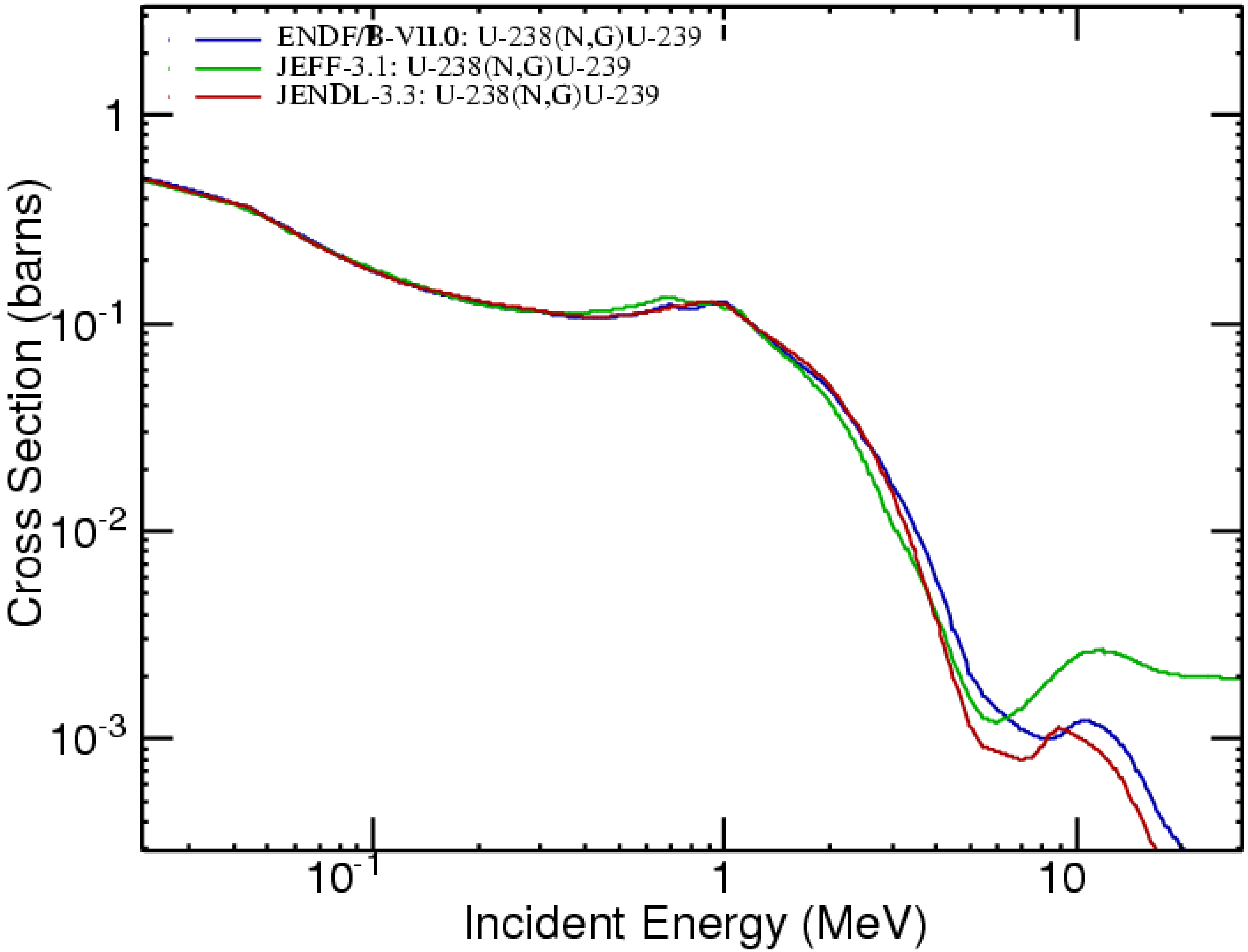
$^{235}U(n,\gamma)^{236}U$

EXFOR database

ENDF = JEFF

180 – 800 keV

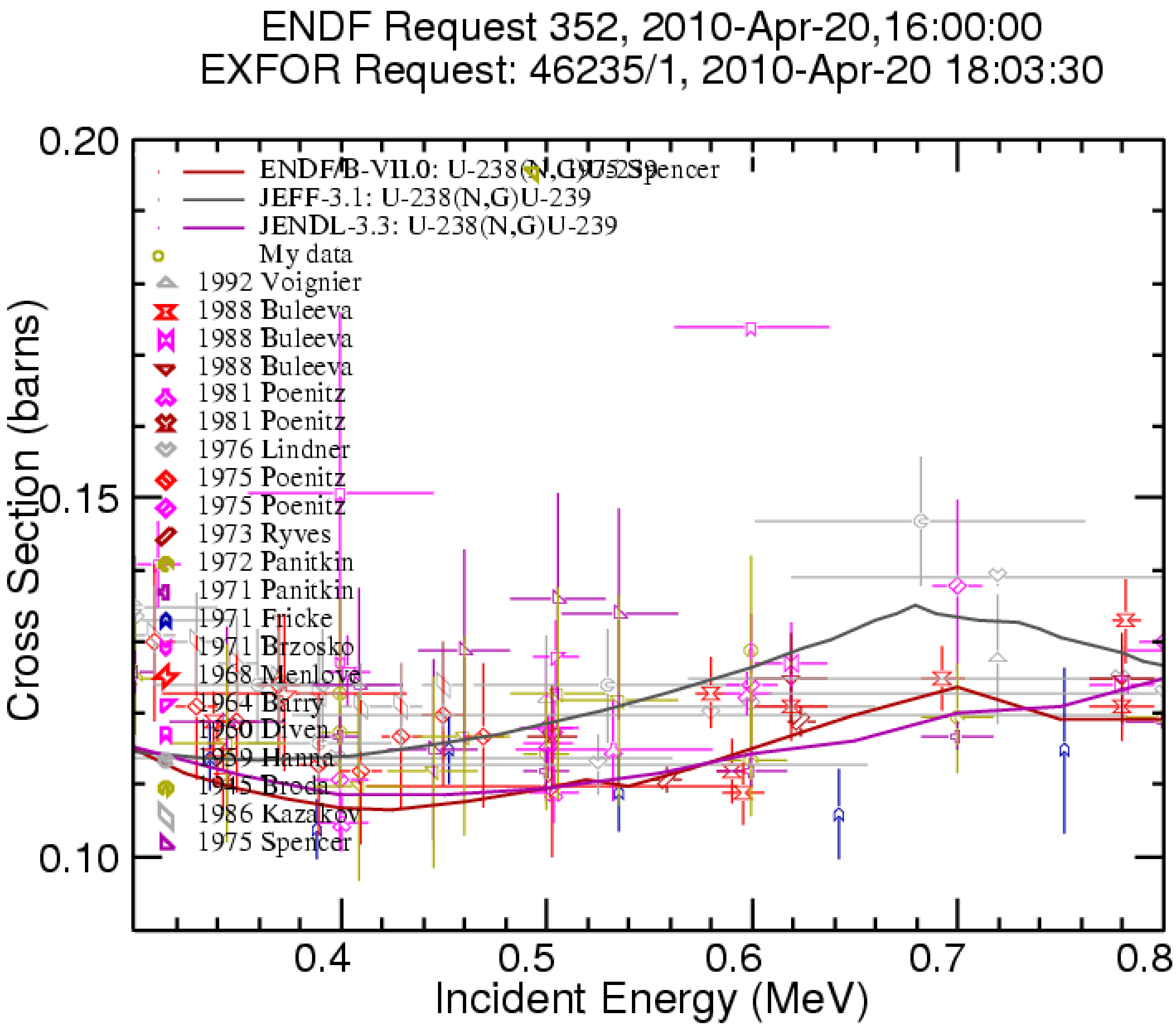
ENDF Request 352, 2010-Apr-20,16:00:00



$^{238}U(n,\gamma)^{239}U$

IAEA database

20 keV– 20 MeV

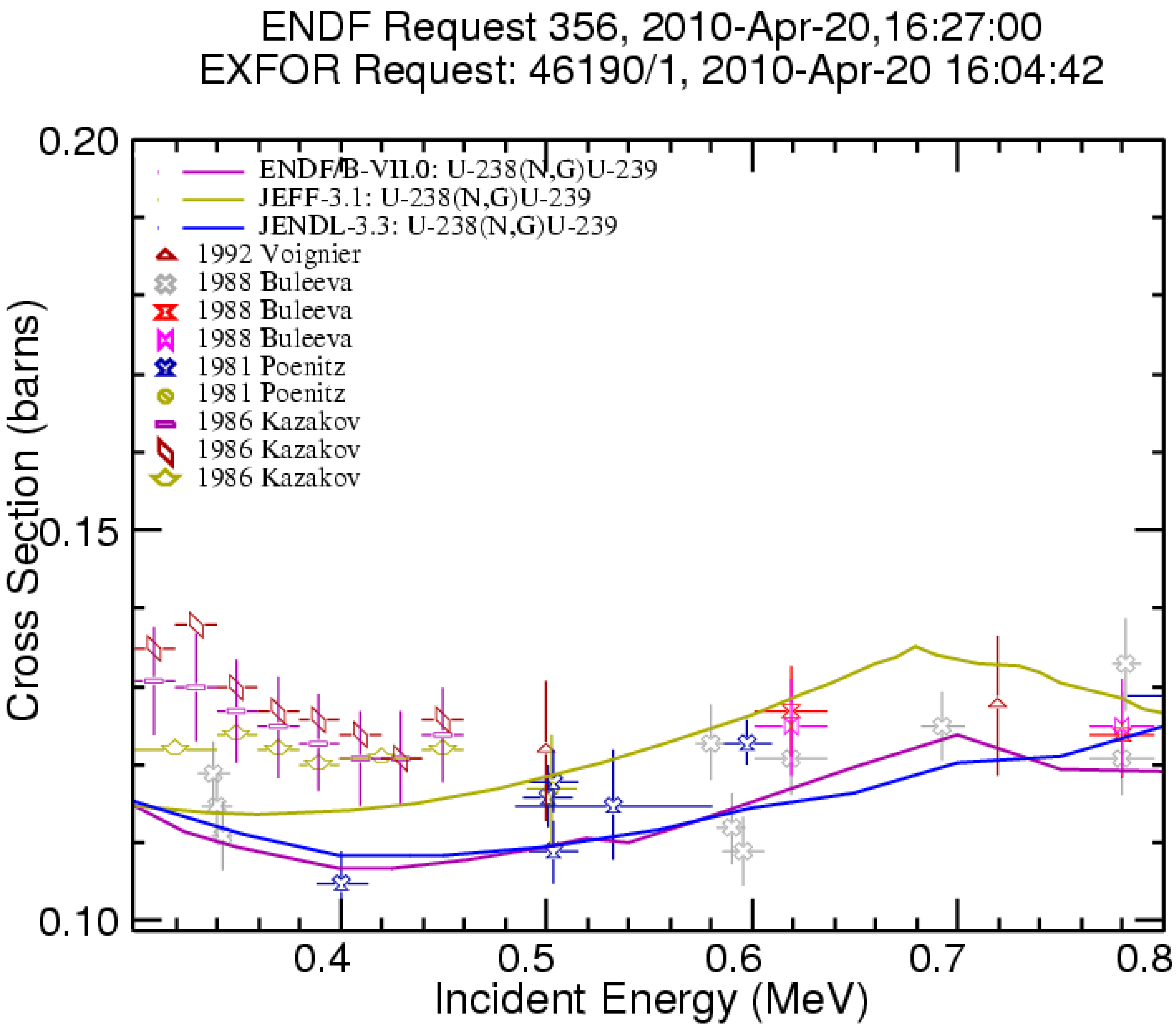


$^{238}U(n,\gamma)^{239}U$

EXFOR database

300 – 800 keV

exp. data



$^{238}U(n,\gamma)^{239}U$

EXFOR database

300 – 800 keV

exp. data > 1980

^{236}U & ^{239}U: Production & Decay

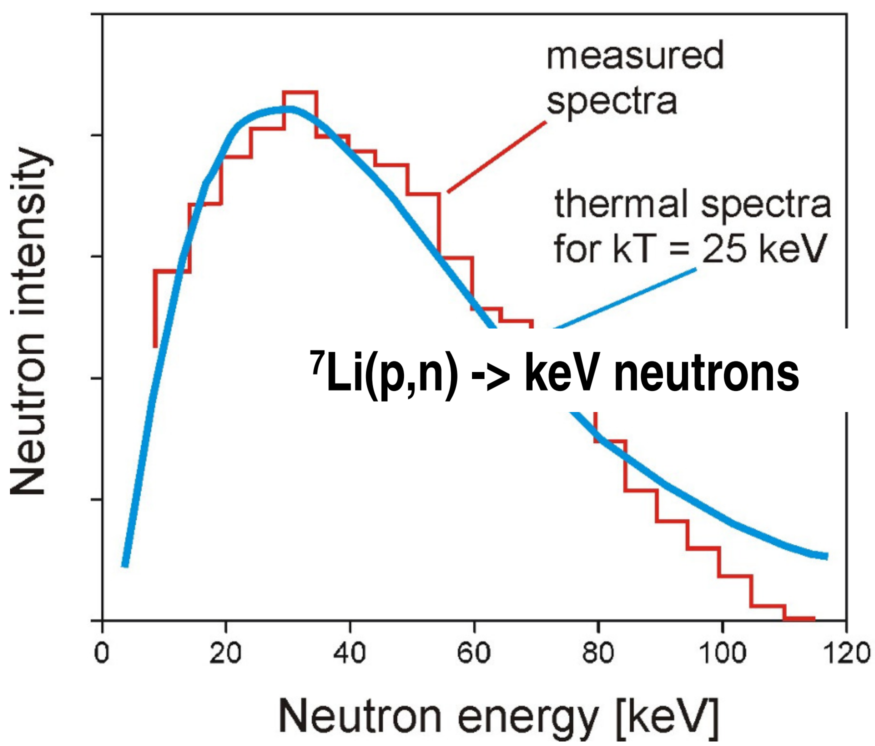
^{236}U: $t_{1/2}$ = 23.4 Myr

^{239}U: $t_{1/2}$ = 23.5 min

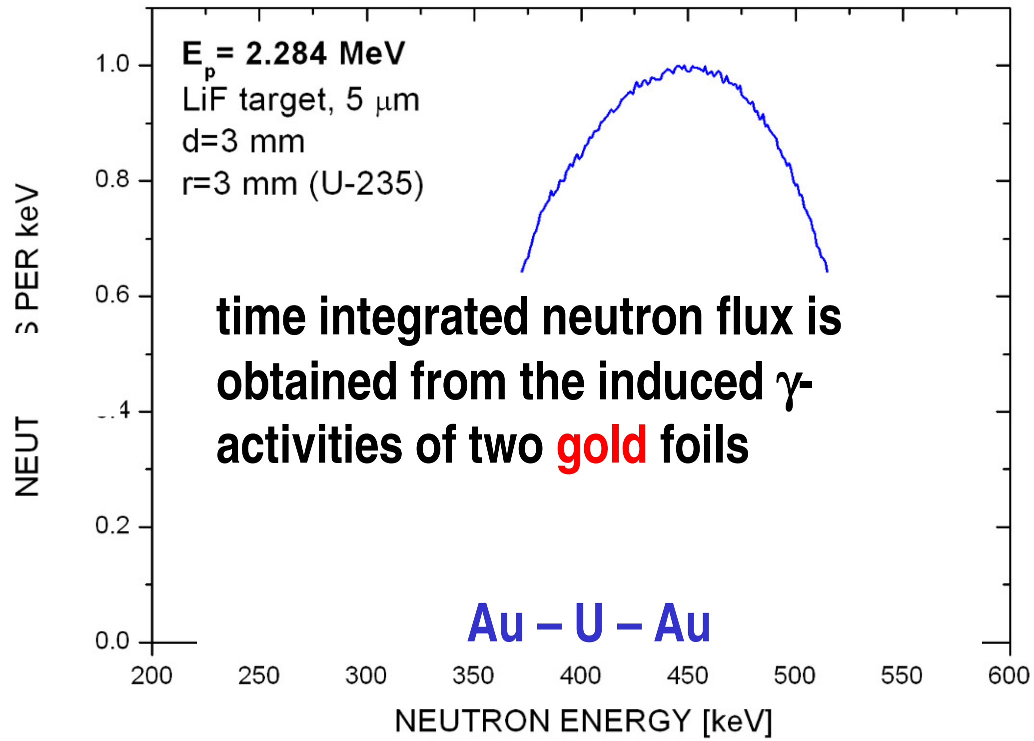
^{239}Np: $t_{1/2}$ = 2.35 days

^{239}Pu: $t_{1/2}$ = 24.1 kyr

Ist-irradiation FZK: 25 / 500 keV neutrons (EFNUDAT)



^{7}Li(p,n) -> keV neutrons



time integrated neutron flux is obtained from the induced γ-activities of two gold foils

Au – U – Au

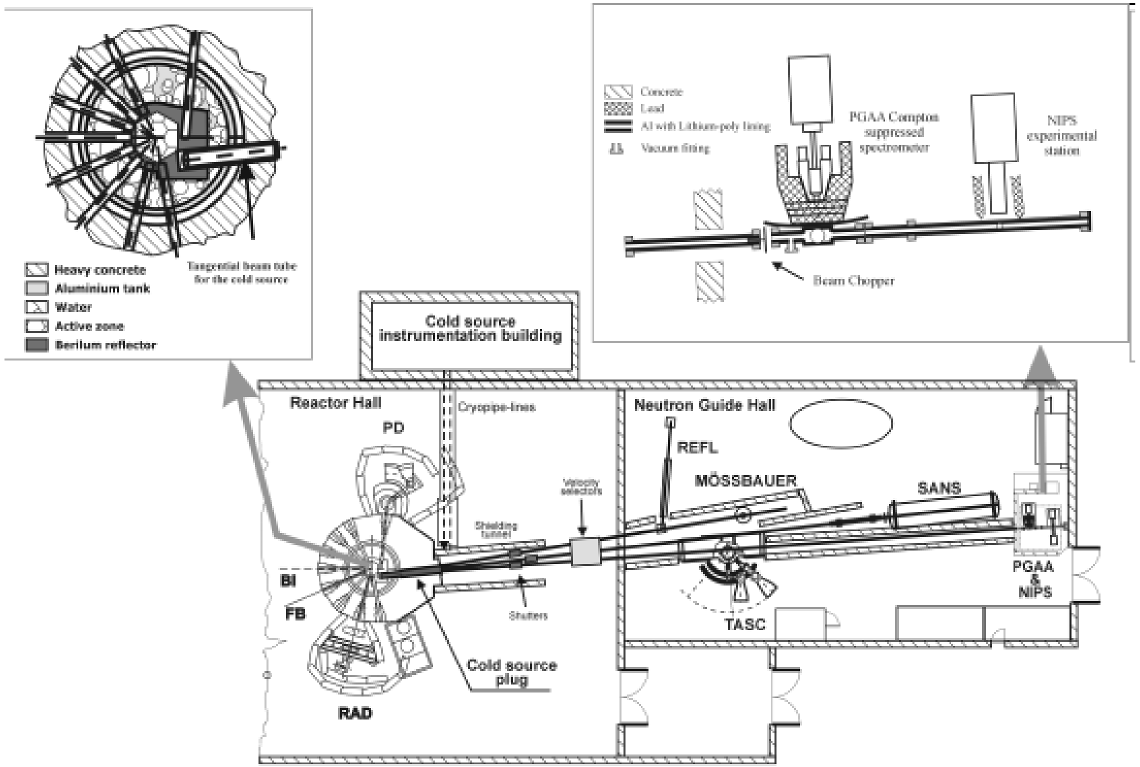
© Rene Reifarth et al.: PINO

neutron energy:	~ 25 keV	~ 450 keV
fluence:	~ $1.7 \cdot 10^{15}$ ncm^{-2}	~ $4 \cdot 10^{15}$ ncm^{-2}
cross section:	~ 690 / 450 mb (U-5 / U-8)	~ 163 / 109 mb

irradiation at Budapest (IKI): cold neutrons $^{235/8}U(n,\gamma)^{236/9}U$ – EFNUDAT project

- Neutron irradiations at Budapest
- activations with cold neutrons
- time integrated neutron flux from
- γ-activities of gold foils and of
- Au powder mixed into the U sample
- ^{nat}U: ^{239}Np activity as additional internal monitor

Au – (U+Au) – Au



$^{235/8}U(n,\gamma)^{236/9}U$ – sample summary

- U samples with natural isotopic composition (0.72 % ^{235}U & 99.28 % ^{238}U)
- IRMM-BC0206: certified for stoichiometry - U_3O_8 & certified $^{235}U/^{238}U$ isotope ratio
- no enrichment; no reactor history
- selected sample: prior to neutron irradiation was found to be low ($^{236}U/^{238}U$ few*10^{-12})

- FZK: 25 keV irradiation finished (1.8*10^{15} n cm^{-2}) 1 pellet 55 mg Au- U_3O_8 - Au

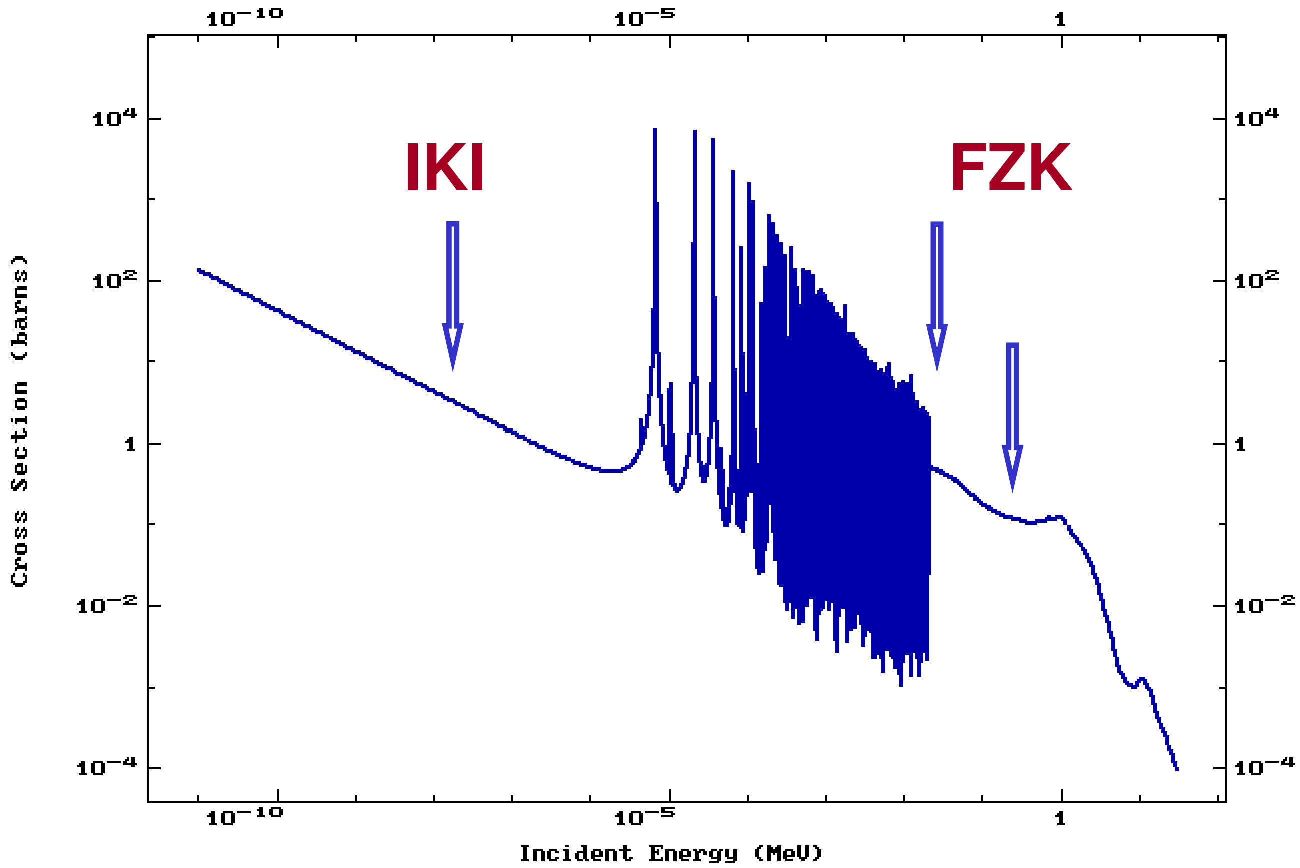
  500 keV irradiation finished (4.3*10^{15} n cm^{-2}) 1 pellet 55 mg Au- U_3O_8 - Au
- IKI: cold neutrons finished (0.7 & 3*10^{14} n cm^{-2}) 2 pellets 70 mg (Au- U_3O_8+Au) - Au

- VERA: AMS measurements 2009-2010

expected isotope ratios: $^{236}U/^{238}U$ ca. 5-8*10^{-12} and 2*10^{-11}

$^{239}Pu/^{242}Pu$: no background interference

$^{238}U(n,\gamma)^{239}U$ -> ^{239}Np -> ^{239}Pu (24 kyr)



$^{235}U(n,\gamma)^{236}U$ (23 Myr) - simultaneously

Analytical Strategy

Au for fluence → **Sample** ← **^{233}U and ^{242}Pu spike**

↓

Chemical separation of U / Pu

↓

AMS

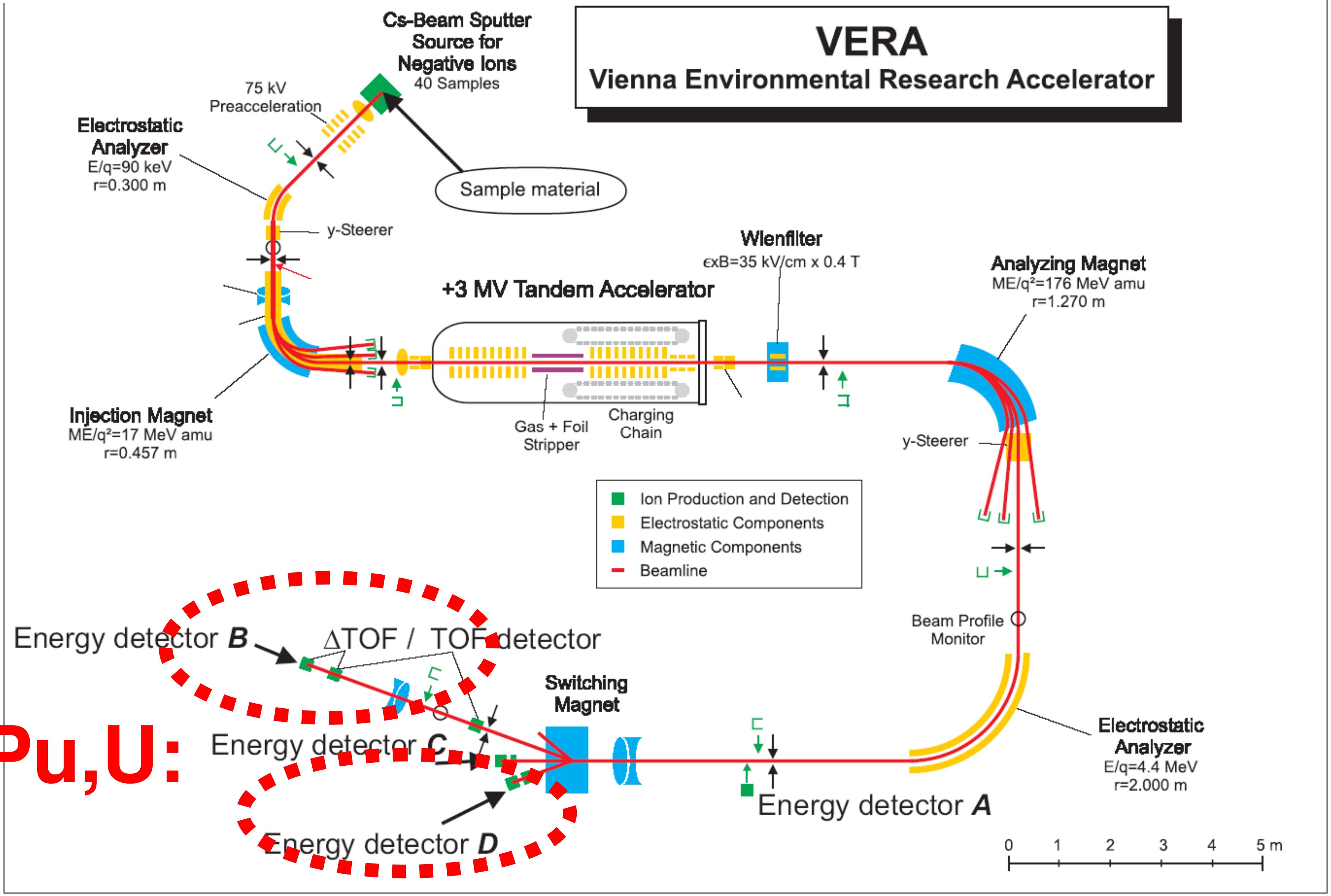
↙ ↘

^{239}Pu / ^{242}Pu — **^{233}U / ^{236}U / ^{238}U**

↓ ↓

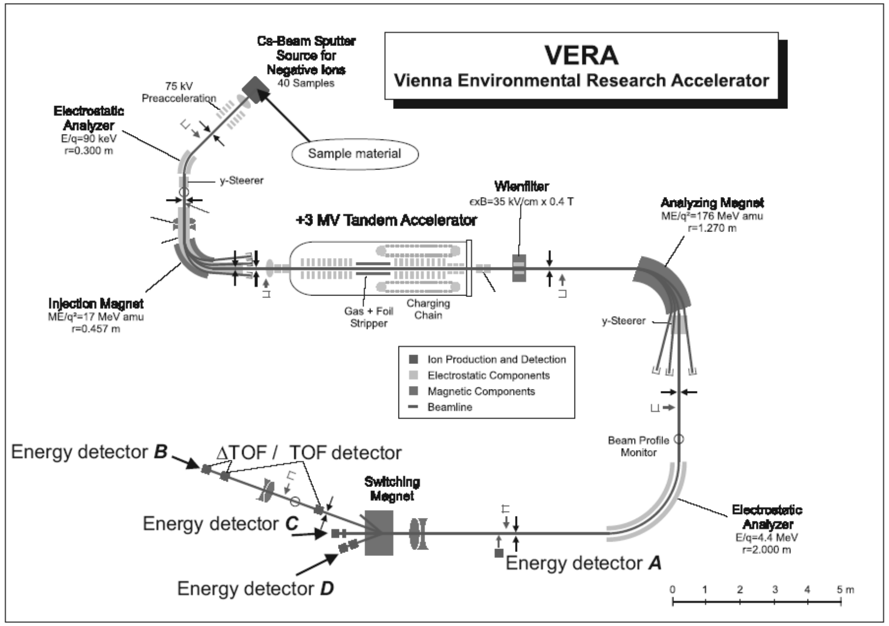
$^{238}U(n,\gamma)^{239}U$ — **$^{235}U(n,\gamma)^{236}U$**

2^{nd}: AMS measurement



Pu,U:

2nd: $^{235/8}U(n,\gamma)^{236/9}U$ via AMS



- AMS determines isotope ratios
- no interfering isobaric background (<-> ICPMS)

  (molecules are completely destroyed)
- isotopic background strongly reduced

$^{236}U/^{235}U$ isotope ratio at VERA

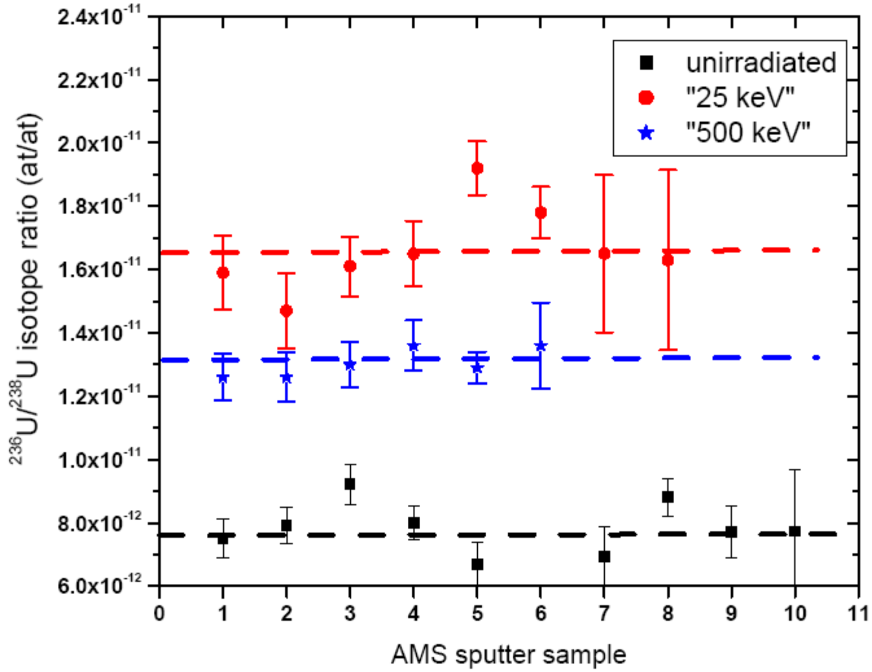
- isotope ratio via fast switching between "stable" and radioisotope (5 s^{-1})
- *precision limited* due to beam losses along detection beamline + background
- ^{236}U blank background of unirradiated sample

„$^{239}Pu/^{238}U$" isotope ratio at VERA

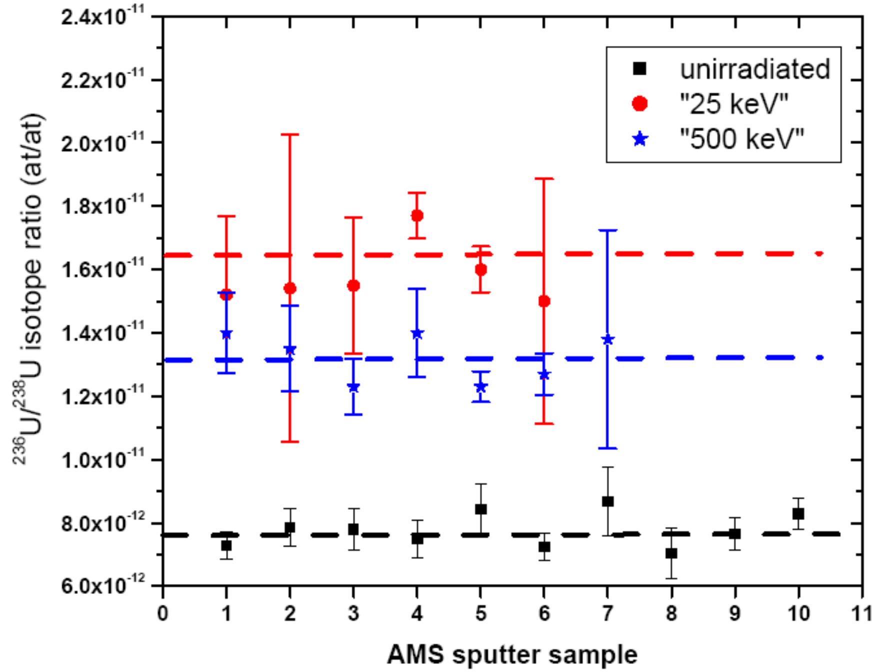
- isotope ratio via switching between reference isotope ^{242}Pu and ^{239}Pu
- *precision limited* due to beam losses along detection beamline + background
- no significant background for ^{239}Pu

$^{235}U(n,\gamma)^{236}U$: AMS results - 25 and 500keV

beamtime I



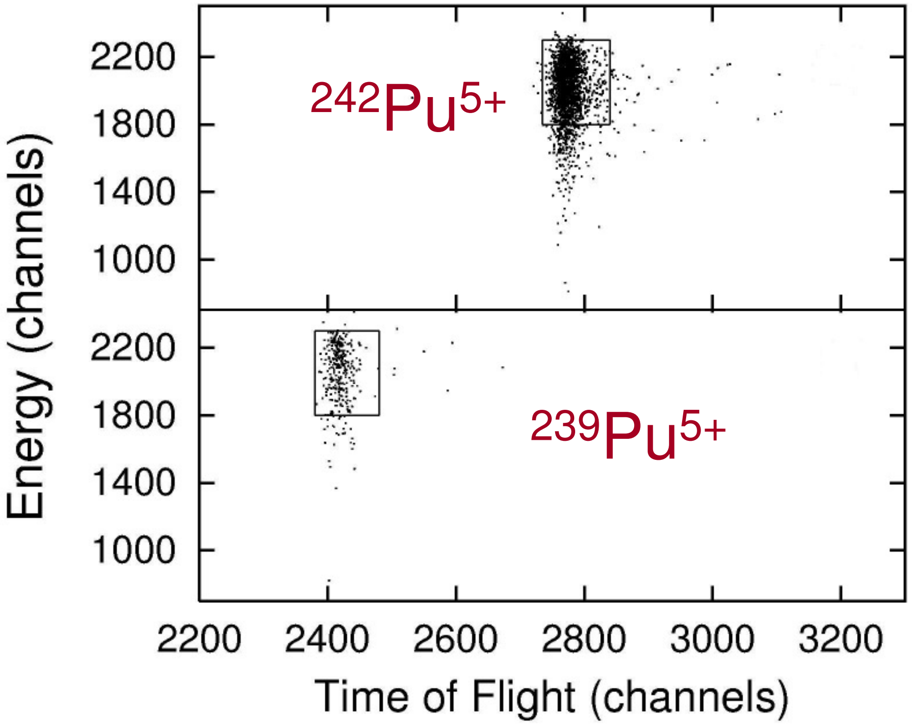
beamtime II



all data normalized to our in-house standard: +- 4 %

later to ^{233}U-spike (+- 1%)

typical AMS-spectrum for Pu measurements

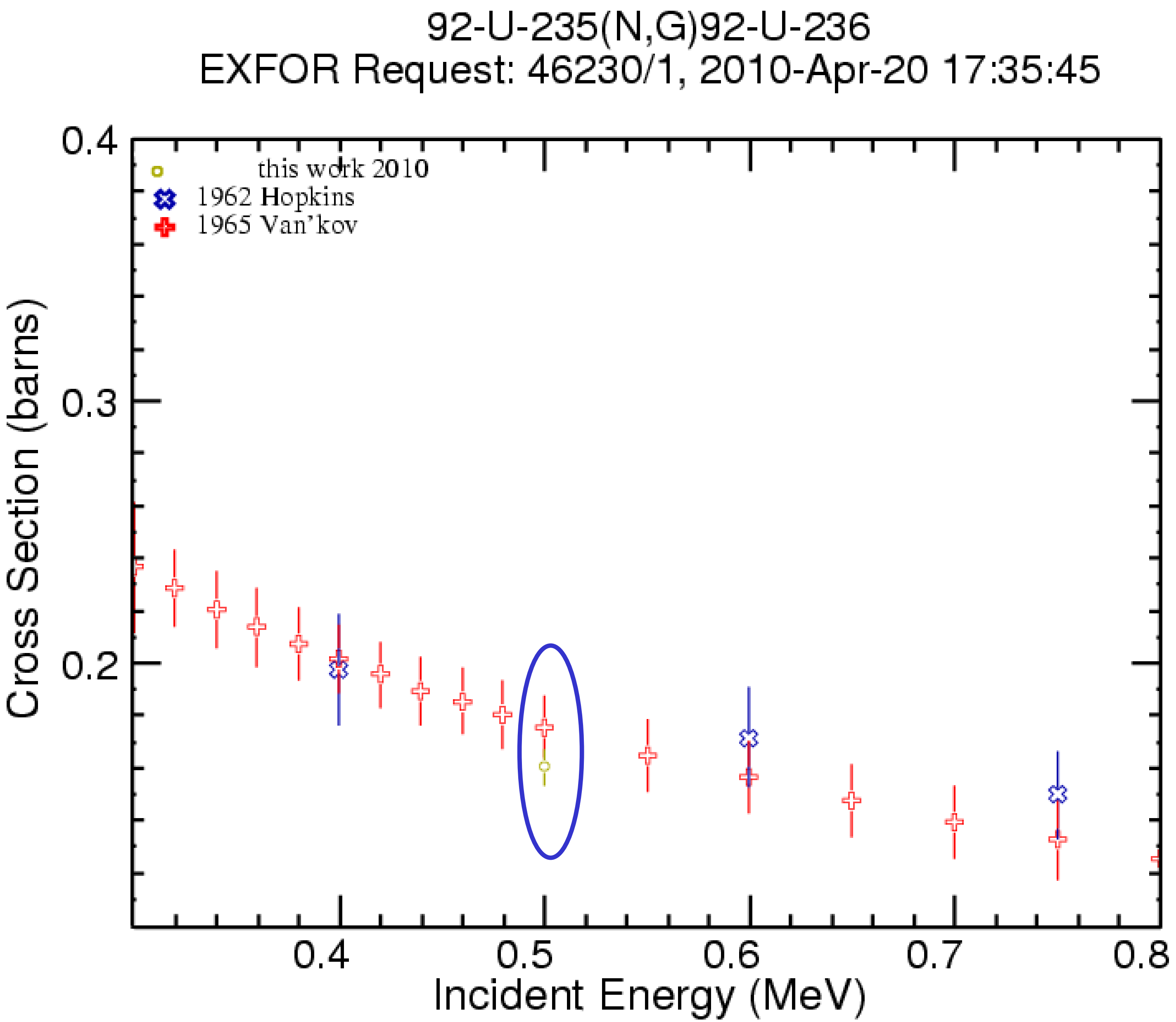


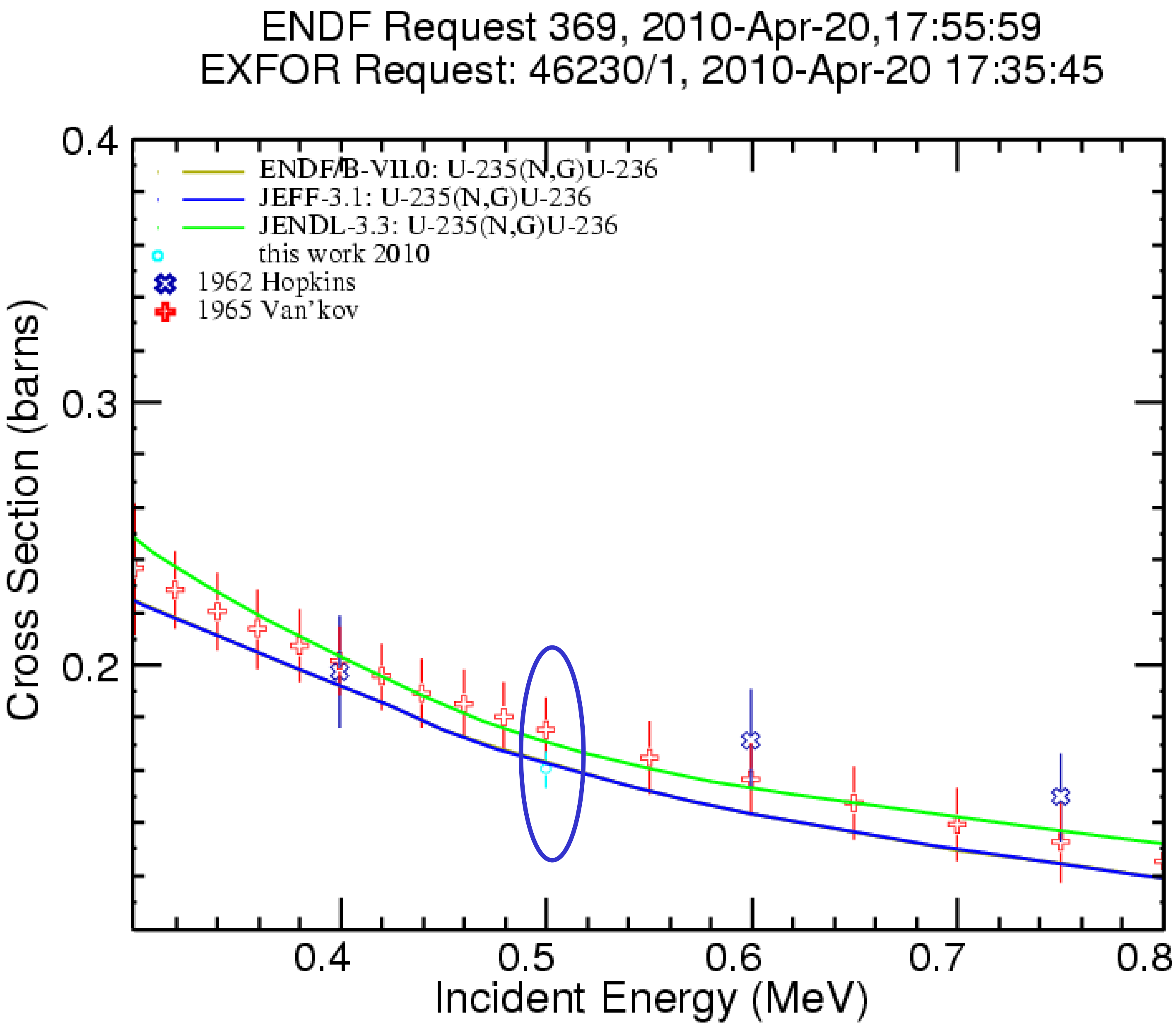
Results

$^{235}U(n,\gamma)^{236}U$

EXFOR database

300 – 800 keV





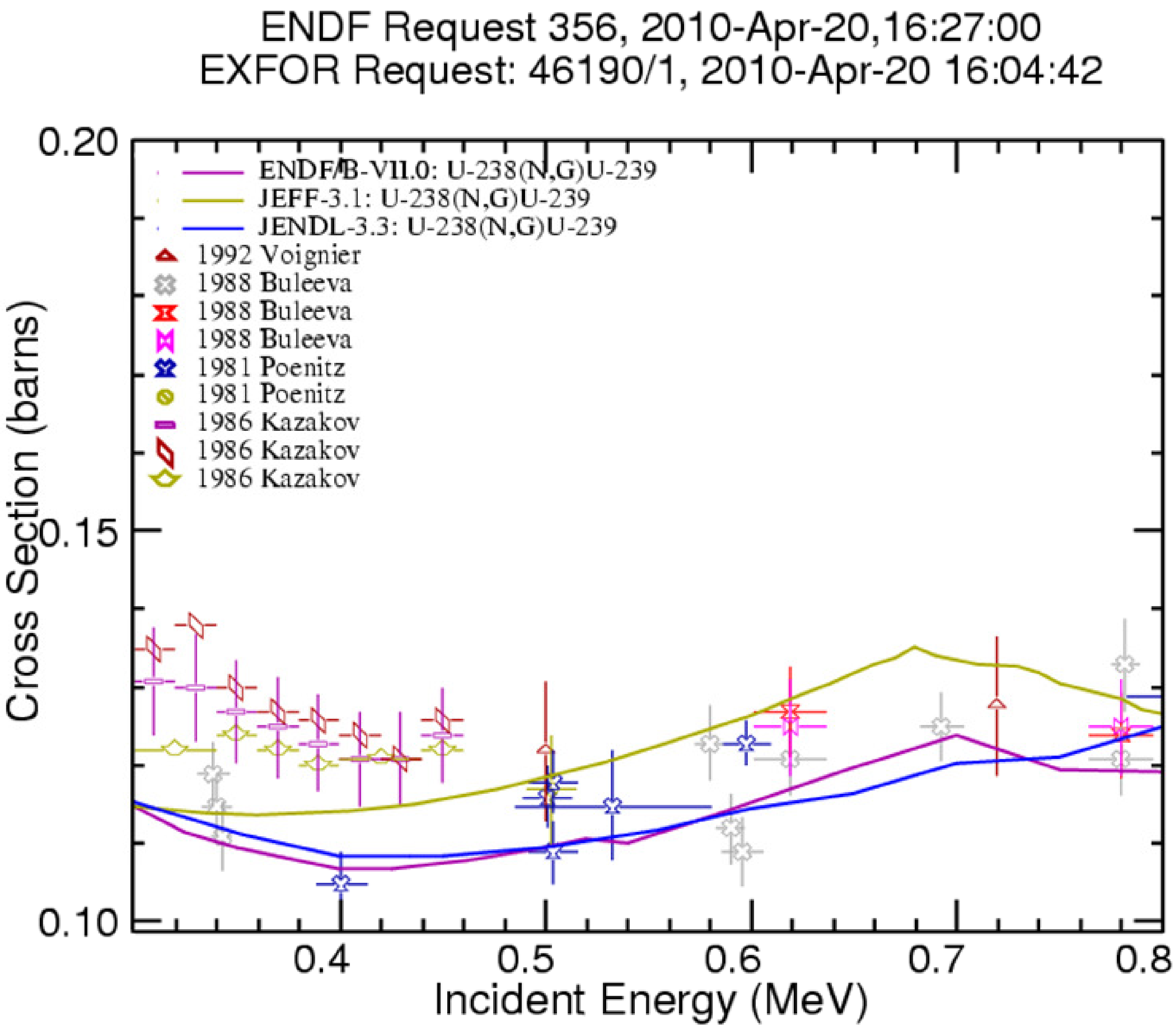
Results

$^{235}U(n,\gamma)^{236}U$

EXFOR database

ENDF = JEFF

300 – 800 keV

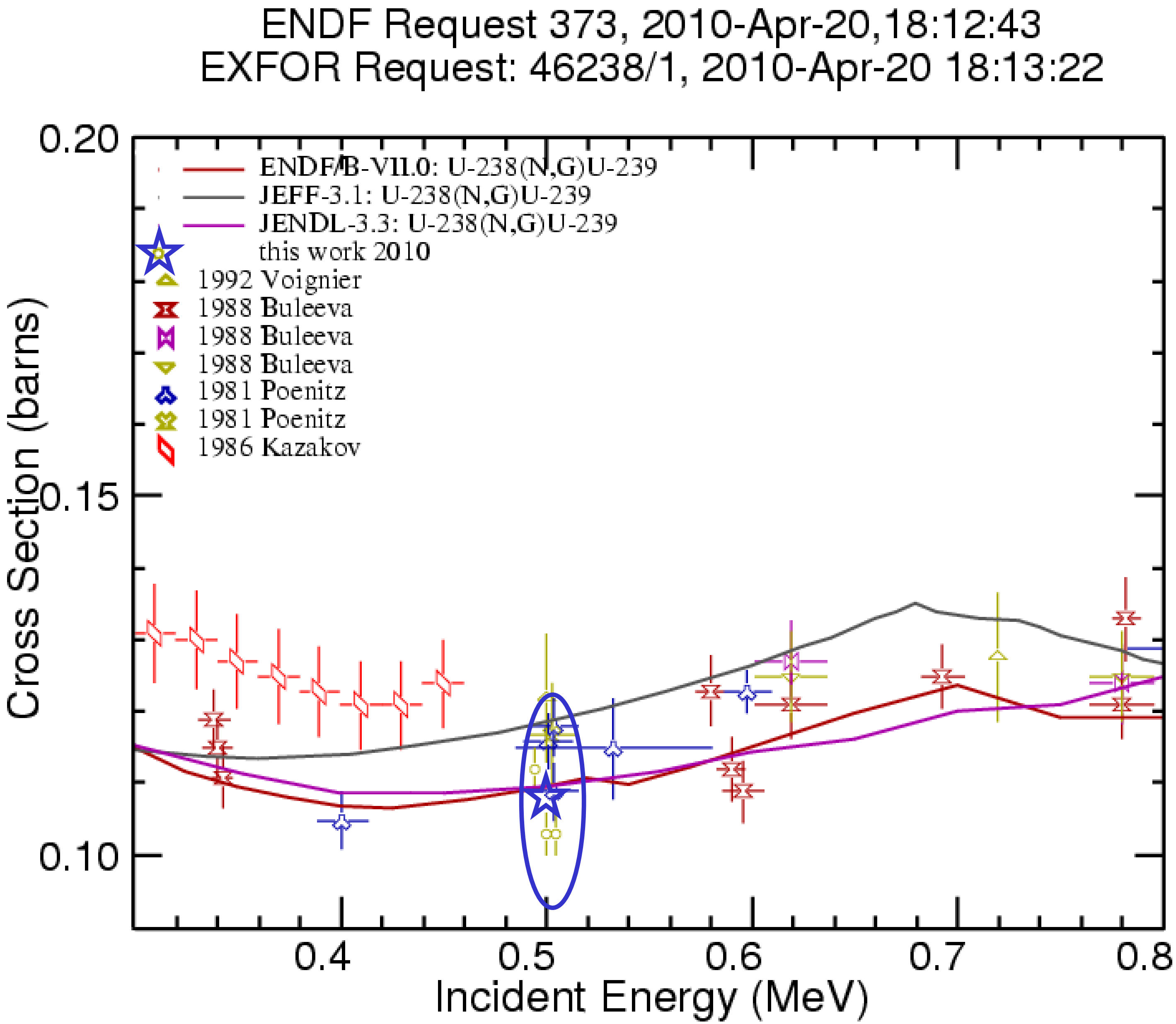


$^{238}U(n,\gamma)^{239}U$

EXFOR database

300 – 800 keV

exp. data > 1980



new data

$^{238}U(n,\gamma)^{239}U$

EXFOR database

300 – 800 keV

exp. data > 1980

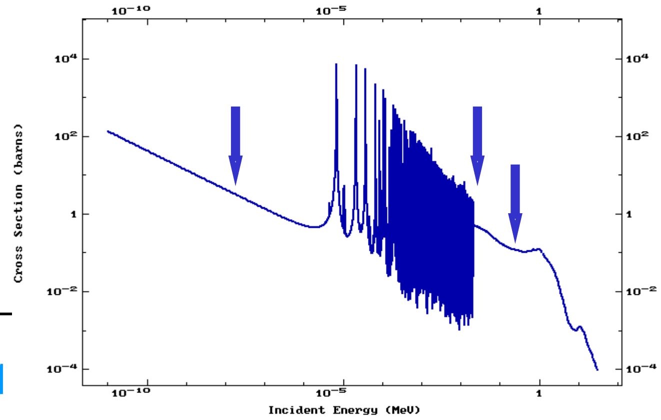
Status – preliminary data

	$^{235}U(n,\gamma)^{236}U$ this work	$^{235}U(n,\gamma)^{236}U$ „recommended“	$^{238}U(n,\gamma)^{239}U$ this work	$^{238}U(n,\gamma)^{239}U$ „recommended“
$\sigma_{thermal}$	IKI-1: 102 ± 1 barn* IKI-2: 103 ± 2 barn*	98.96 barn 98.96 barn	IKI-1: -- IKI-2: --	2.68 barn (2.72 barn JENDL)
$\sigma_{25\ keV}$	FZK-1: 591±25 mbarn	(ENDF = JEFF) ENDF: $\sigma_{U-8}/\sigma_{U-5} = 0.61$ this work (AMS ratio): $\sigma_{U-8}/\sigma_{U-5} = 0.63$	rn	360 mbarn calculated (ENDF = JEFF)
$\sigma_{500\ keV}$	FZK-2: 158 ± 7 mbarn	160-185 mbarn (ENDF, JEFF) ?? Spectrum??	FZK-2_a: 112 ± 2 mbarn** F… rn** mean: 109 mbarn FZK-2_c: 104 ± 2 mbarn**	109 mbarn (ENDF, JENDL) 119 mbarn (JEFF)

* relative to $^{236}U/^{238}U$ in-house standard

= data relative to ^{233}U-spike - 3.5% lower - ok

** slightly different values expected due to inhomogenous activation



AMS: limitations

- isotope ratios (e.g. $^{236}U/^{238}U$) measured:
  - interference with background: VERA ca. < 10^{-12} for ^{236}U
    - isobaric interference (e.g. ^{235}UH) – not an issue for AMS
    - isotopic interference (e.g. ^{235}U, ^{238}U): e.g. $^{236}U/^{238}U < 10^{-12}$
    - ev. m/q interference (count-rate interference - detector busy)

- number of nuclei in sample
  - determines statistical uncertainty
  - typical overall efficiency: 10^{-3} - 10^{-4}

Examples - limitations

U: isotope ratios

20 mg irradiated with 10^{15} n; 700 mb

$^{236}U/^{235}U$ = $7*10^{-10}$

$^{236}U/^{238}U$ = $5*10^{-12}$

N_{236} = $3*10^{8}$ -> AMS: > 10^{5} counts in detector

U: number of nuclei; example: $^{238}U(n,\gamma)^{239}U$ -> (0.5 h) -> ^{239}Np -> (2.4 d) -> ^{239}Pu ($t_{1/2}$ = 24 000 a)

N_{239} = 10^{7} ! (required for AMS counting -> ≈ 10^{3} detected)

$\sigma\phi$ = 1barn*10^{15} n cm^{-2}

= 10^{-9}

N_{238} = $N_{239} / \sigma\phi$ (required U-atoms)

= 10^{16} ats (4 ug material) + extraction of ^{239}Pu from U-bulk (with well-known spike)

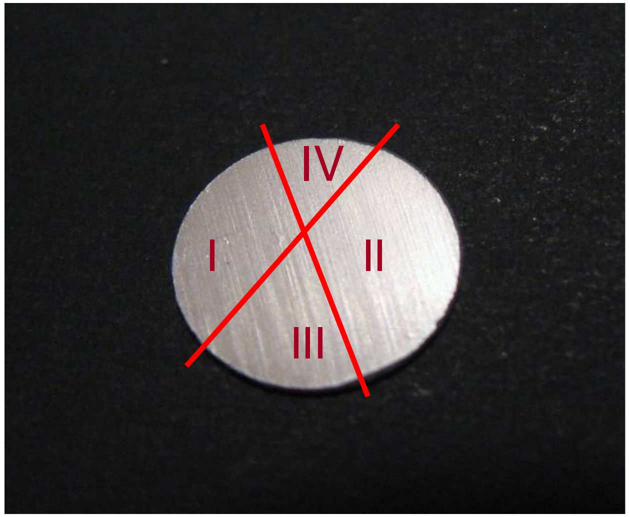
Collaboration:

- **VERA (Univ. Vienna):** **A. Wallner, O. Forstner, R. Golser, W. Kutschera, A. Priller, P. Steier, F. Quinto, K. Buczak, C. Lederer, G. Wallner**
- **FZK (Karlsruhe) / GSI:** **F. Käppeler, I. Dillmann**
- **IKI Budapest** **T. Belgya, L. Szentmiklosi**
- **IAEA (Vienna):** **A. Mengoni**
- **Atominstitut (Vienna):** **M. Bichler**
- **...**

$^{238}U(n,\gamma)^{239}U$ -> $^{239}Pu/^{242}Pu$ with AMS

- pellet was divided into 3 to 4 individual pieces
- dissolved
- well-known ^{233}U + ^{242}Pu spike added
- U / Pu column separation
- U- and Pu-Oxide powder
- pressed into AMS sample holder

- AMS: U: ^{236}U vs ^{233}U and vs $^{238(235)}U$
- AMS: Pu: ^{239}Pu vs ^{242}Pu



$$Z = N \cdot \sigma_{E_n} \cdot \Phi_{tot} \cdot f_b$$

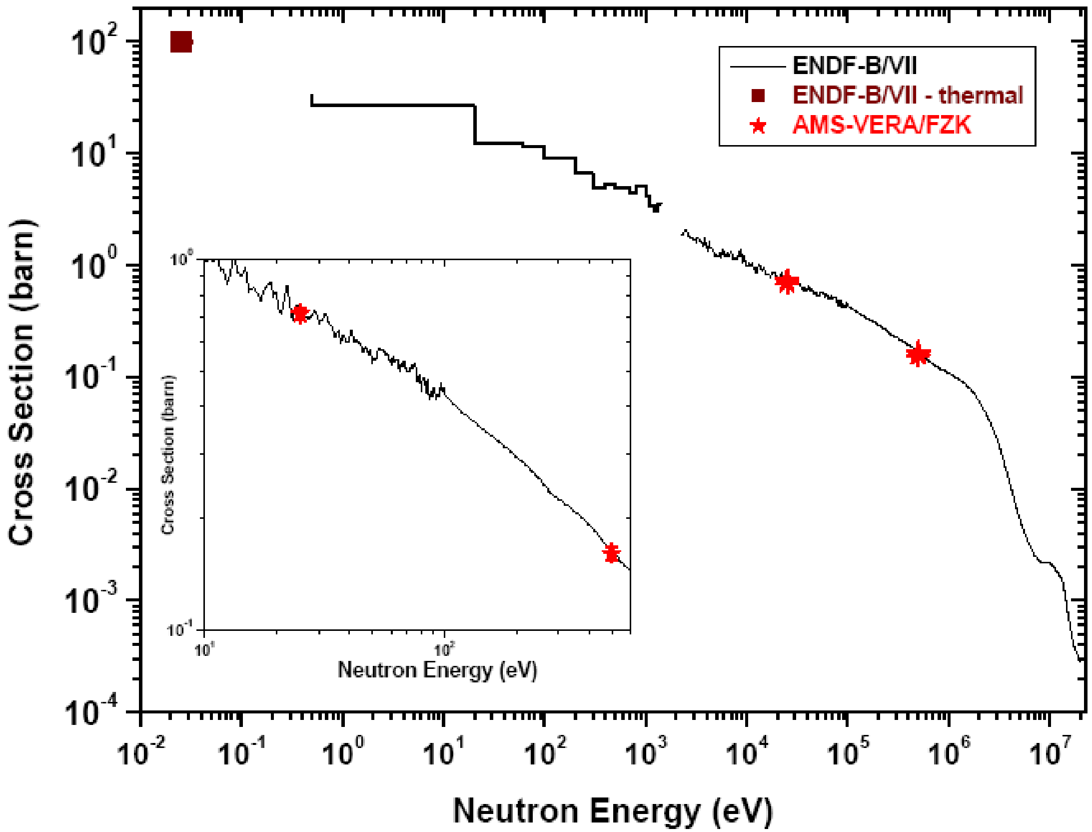
$$\sigma_{E_n} = \frac{Z}{N} \cdot \frac{1}{\Phi_{tot} \cdot f_b}$$

$$e.g.\ \sigma_{E_n} = \frac{^{239}U}{^{238}U} \cdot \frac{1}{\Phi_{tot}} = \frac{^{239}Pu}{^{238}U} \cdot \frac{1}{\Phi_{tot}} \quad \text{for } T_{wait} >> T_{1/2}(^{239}U)$$

$$= \frac{^{239}Pu}{^{242}Pu} \cdot \frac{^{242}Pu}{^{238}U} \cdot \frac{1}{\Phi_{tot}} \quad \text{for } ^{238}U(n,\gamma)^{239}U$$

AMS **mass** **Au**

$^{235}U(n,\gamma)^{236}U$



preliminary data

+ adjust for broad neutron energy distribution

Summary U-activations

- new AMS measurements for thermal, 25 keV (MB) and 500 keV neutron energy

- data evaluation in progress: 500 keV -> < 5% (3%) uncertainty seems reasonable

$^{235}U(n,\gamma)^{236}U$:

- thermal value as reference for 25/500 keV
- in-house AMS standard
- ^{233}U spike as additional standard

$^{238}U(n,\gamma)^{239}U$:

- thermal value as reference for 25/500 keV
- ^{242}Pu spike as additional standard

Extension to other minor actinides

U: isotope ratios

20 mg irradiated with 10^{15} n cm^{-2}; 700 mb

$^{236}U/^{235}U$ = $7*10^{-10}$

$^{236}U/^{238}U$ = $5*10^{-12}$ (natural sample)

N_{236} = $3*10^{8}$ atoms -> AMS: ~ 10^{6} counts in detector

Am: example: $^{241}Am(n,\gamma)^{242g}Am$ -> (16h) -> ^{242}Cm (163 d)

N_{242} = 10^{7} ! (required for AMS counting -> ≈ 10^{3} detected)

$\sigma\phi$ = 700 mb*10^{15} n cm^{-2}

= $7*10^{-10}$

N_{241} = $N_{242} / \sigma\phi$

= 10^{16} ats (< 0.01 mg material) + !! Extract Cm from Am-bulk!!

(400 cts ... 5 % statistics: 10^{15} ats (< µg!) ↔ Ion source contamination! (^{241}Pu)