Nuclear data measurements with slow neutrons at Institut Laue Langevin

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Institut Laue Langevin

- founded 1967
- today 13 member states: F, D, UK, E, CH, A, I, CZ, S, HU, B, SK, DK
- operates 58 MW high flux reactor with most intense extracted neutron beams
- over 40 instruments, mainly for neutron scattering
- user facility: 2000 scientific visitors from 45 countries per year
- Director General: Richard Wagner
- “Nuclear data instruments”: LOHENGRIN, GAMS, PF1, S18, (V4),…

The LOHENGRIN fission fragment separator

mass-separated fission fragments,

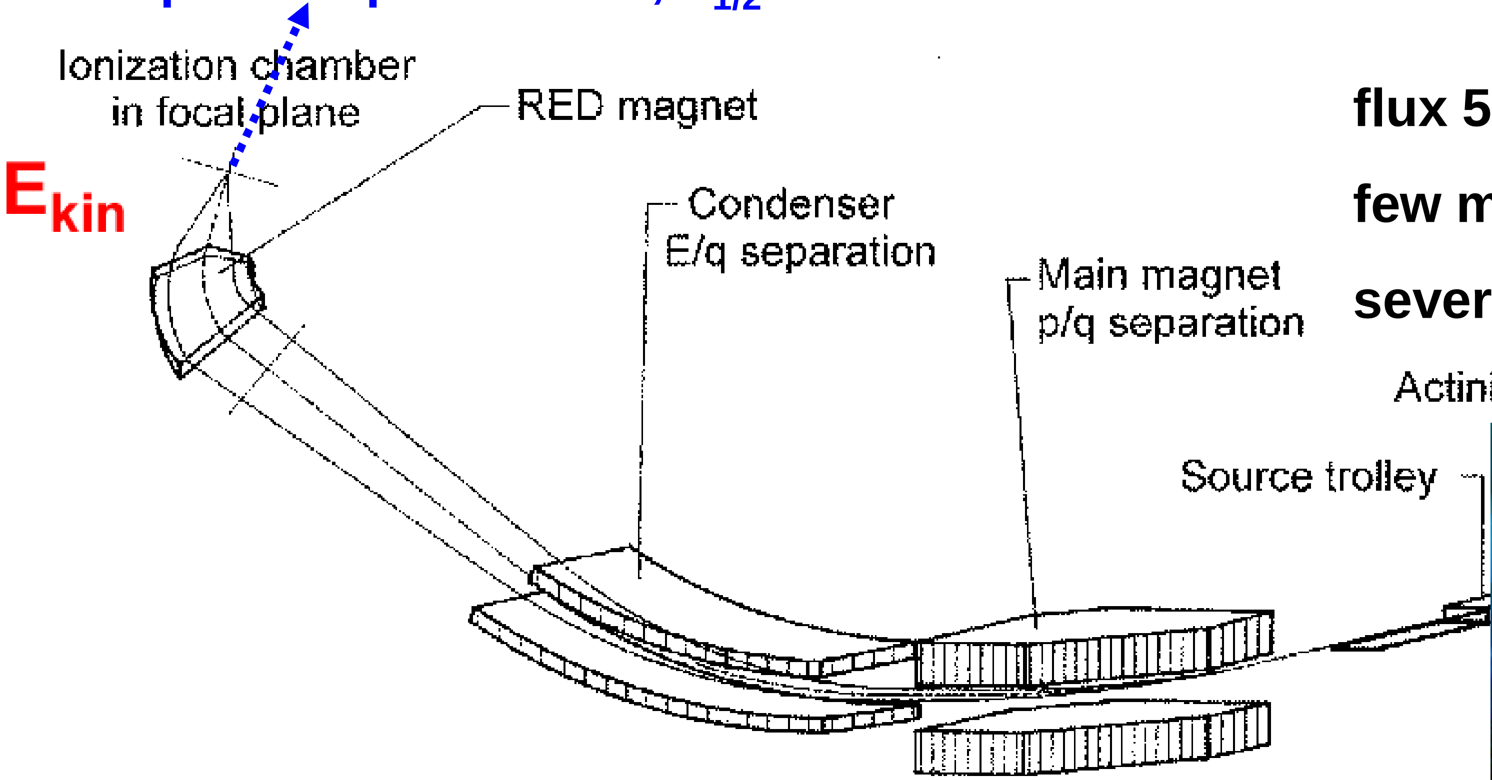
up to 10^5 per second, $T_{1/2} \geq$ microsec.

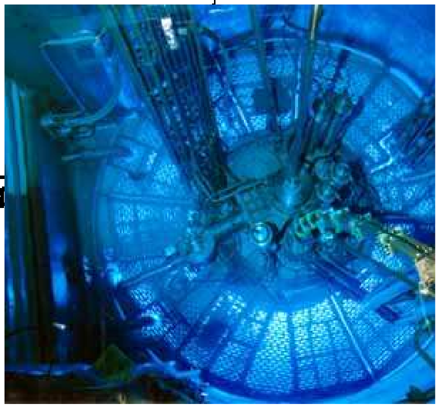
$\Delta A/A$ = 3E-4 – 3E-3
$\Delta E/E$ = 1E-3 – 1E-2

flux $5.5 \cdot 10^{14}$ n./cm^2/s

few mg fission target

several 10^{12} fissions/s





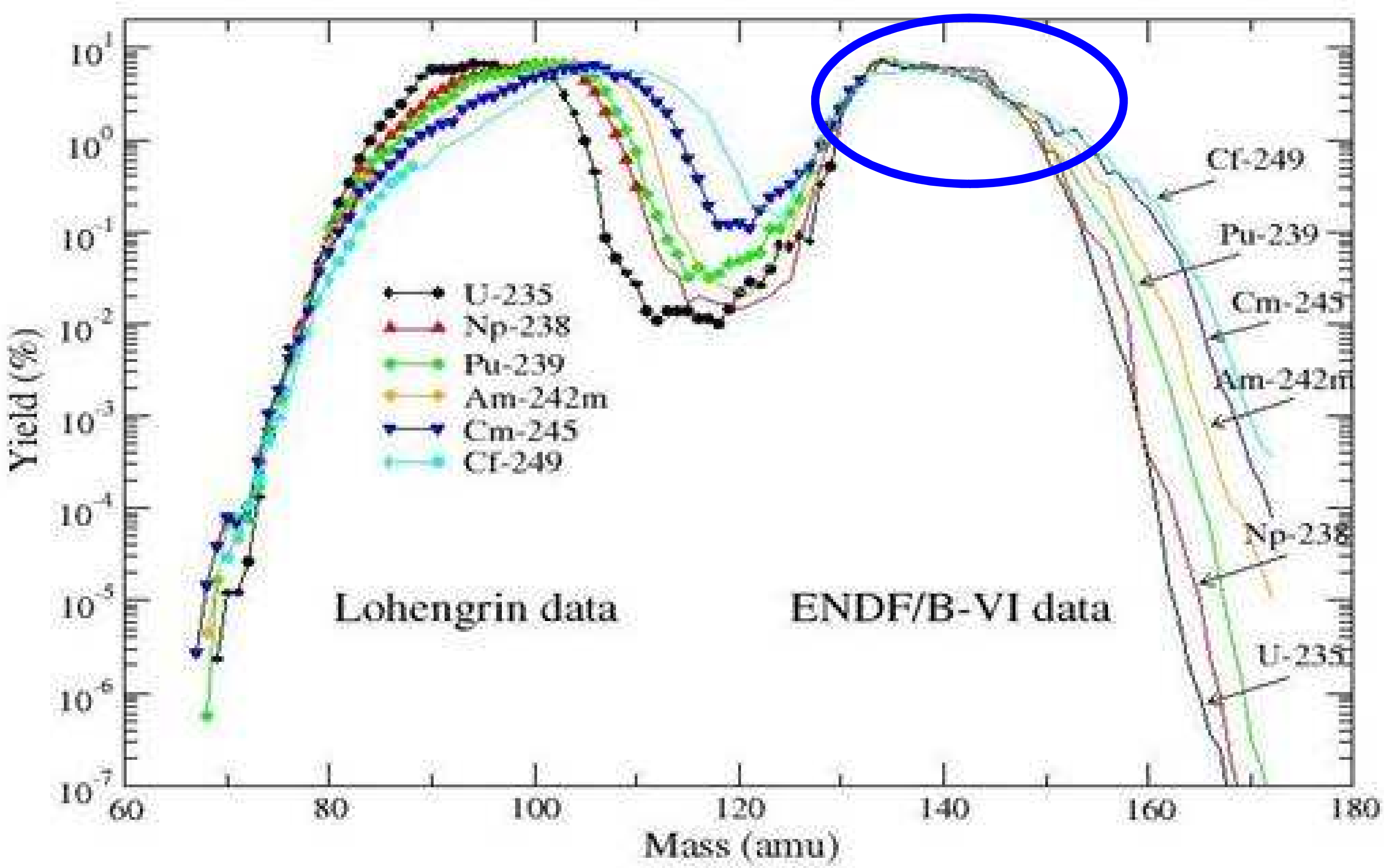
$$m v^2 / r_{el} = q E \qquad m v^2 / r_{magn} = q v B$$

$$E_{kin} / q = E / 2 r_{el} \qquad m v / q = B r_{magn}$$

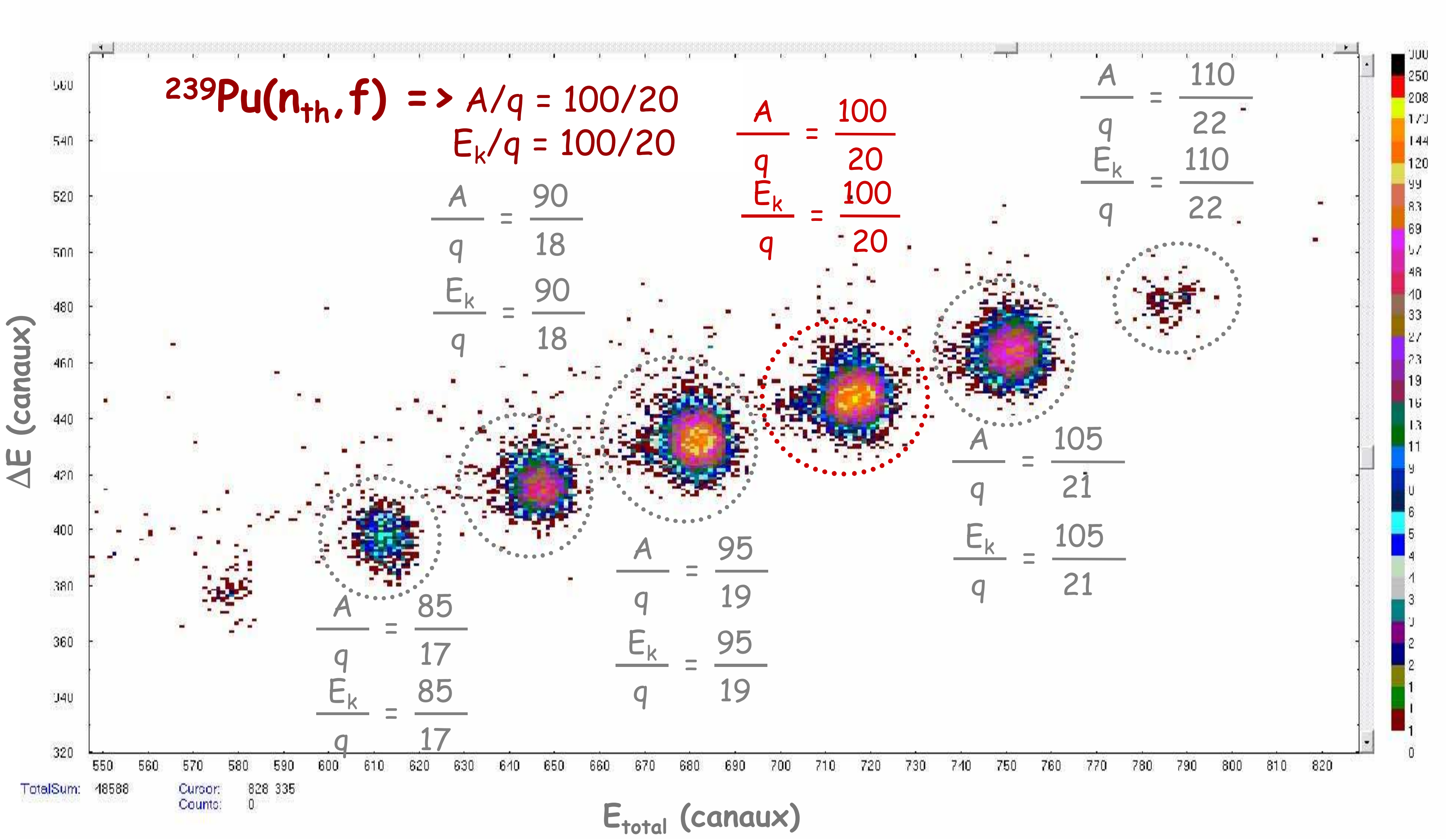
P. Armbruster et al., Nucl. Instr. Meth. 139 (1976) 213.

Is a 36 year old nuclear physics instrument still competitive?

Fission yield measurements

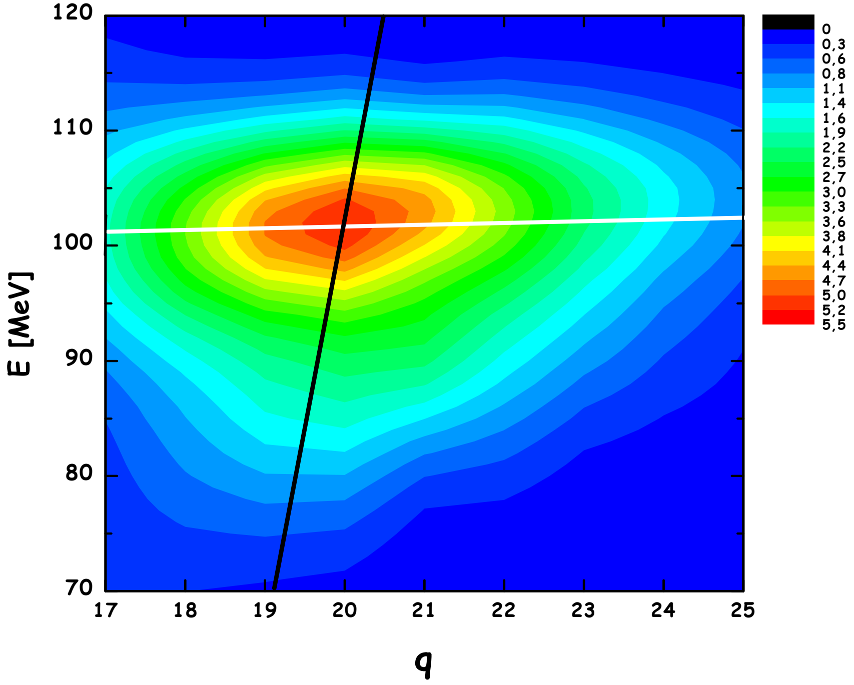


Mass identification with ionization chamber

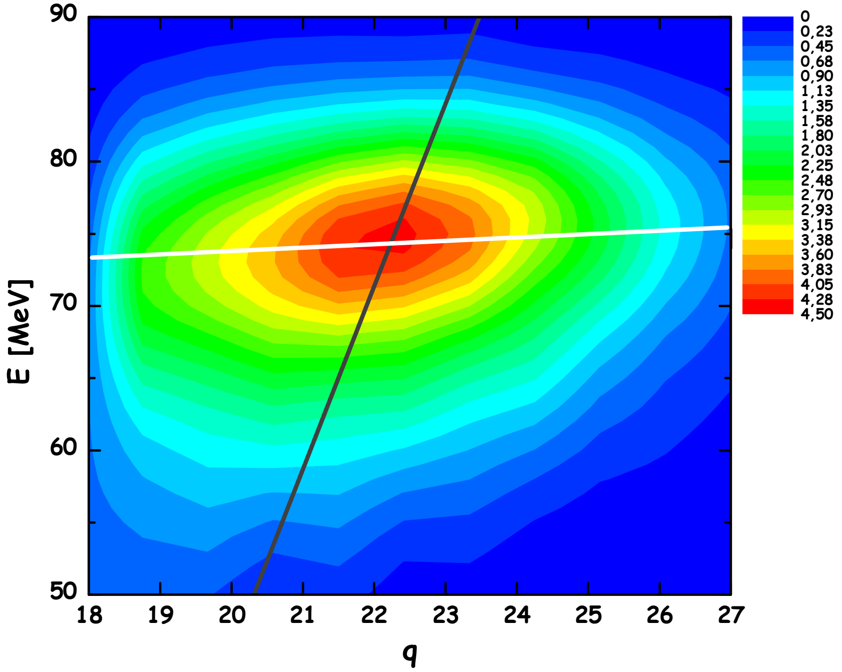


Distributions in E and q

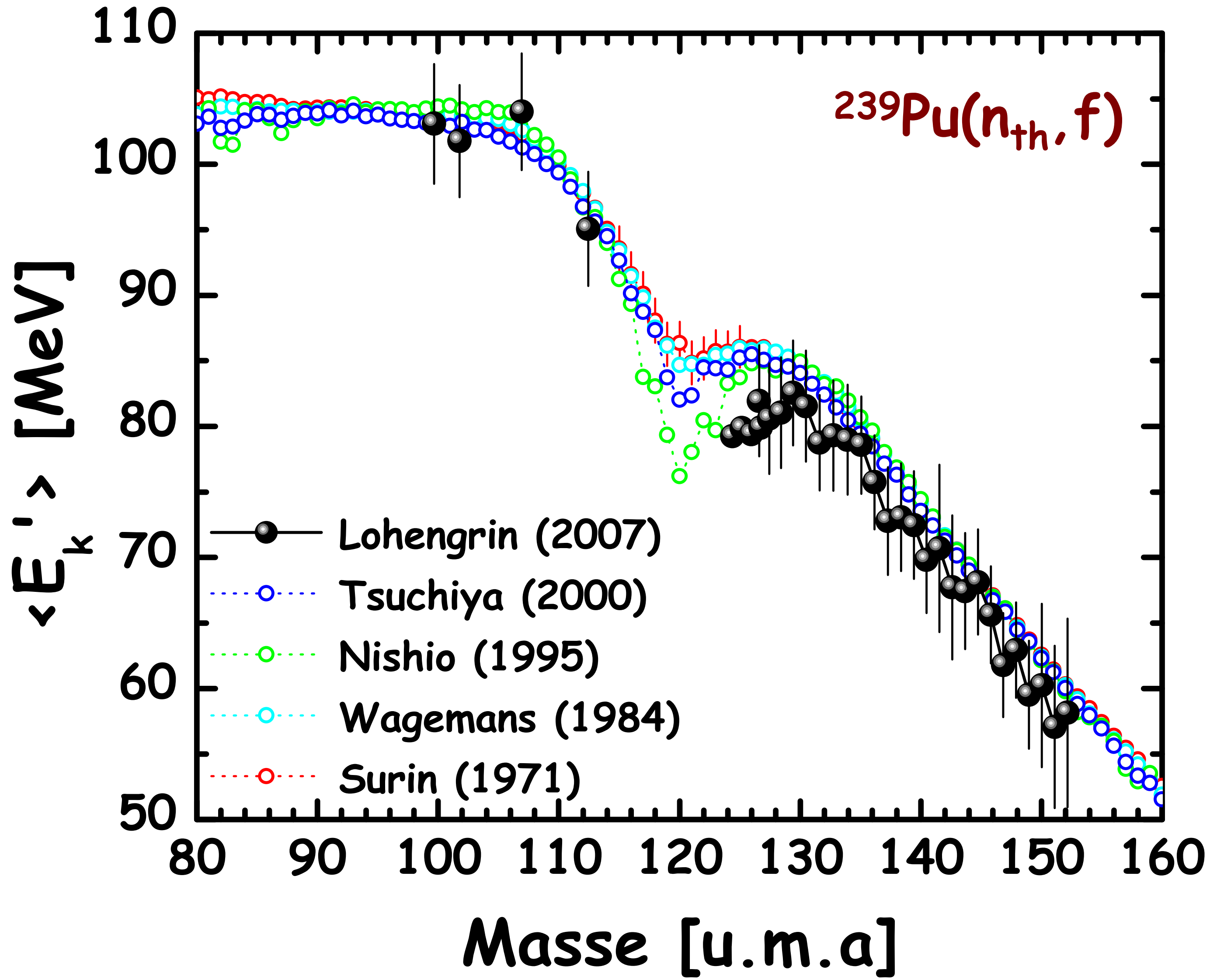
A = 98



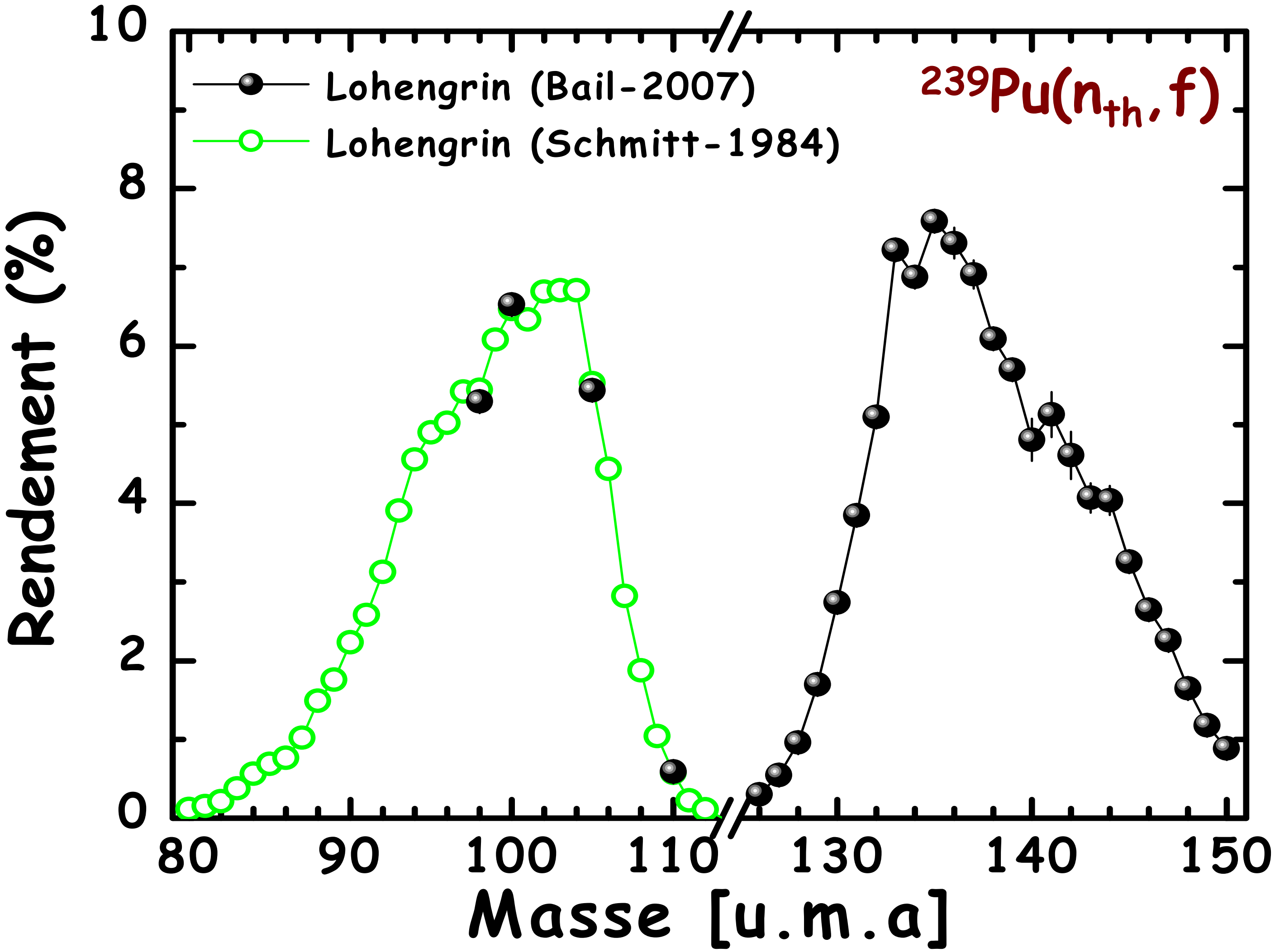
A = 136



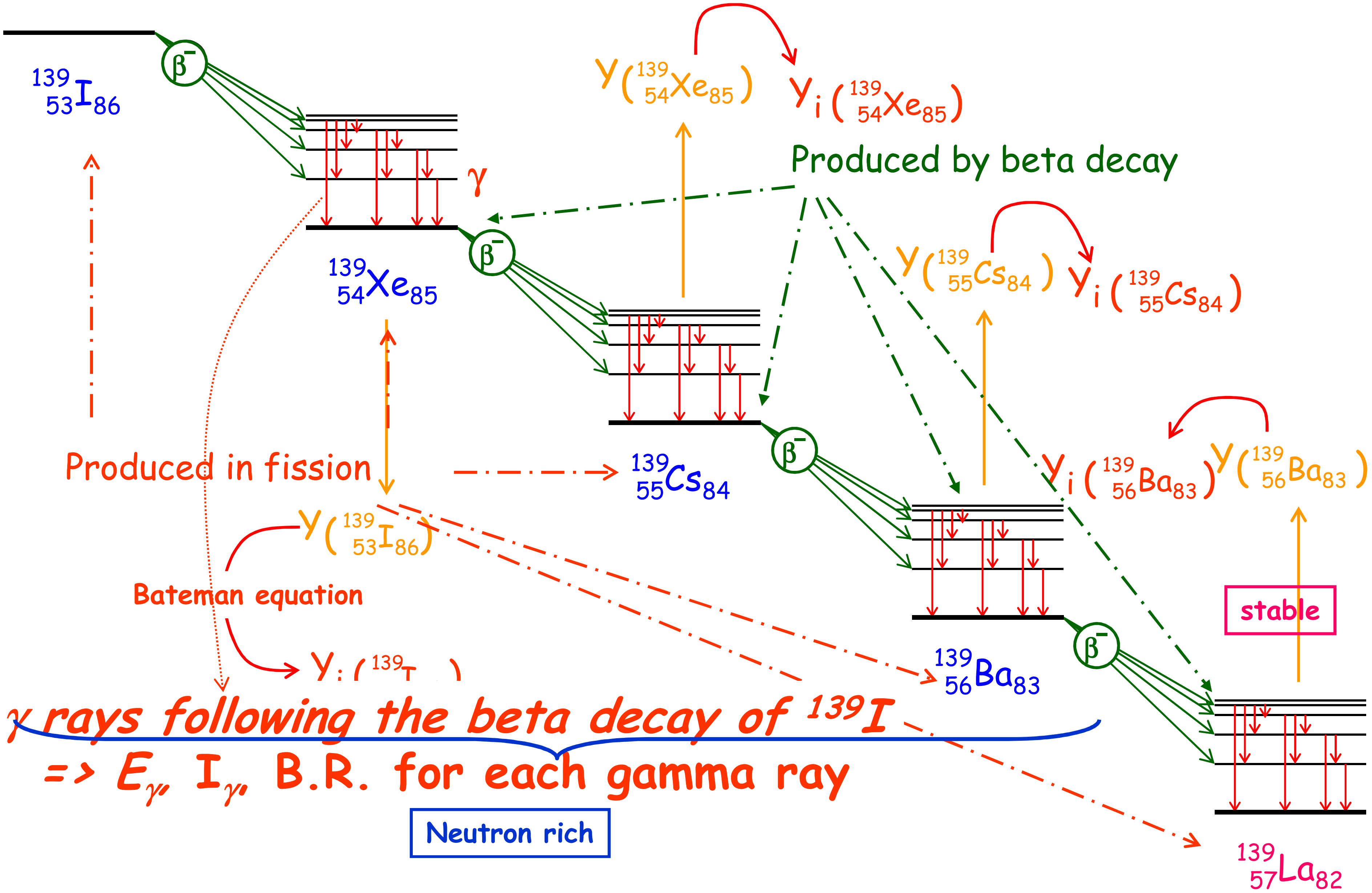
Kinetic energy distributions



Mass yields


Lohengrin (Bail-2007)
Lohengrin (Schmitt-1984)
^{239}Pu(n$_{th}$,f)
Rendement (%)
Masse [u.m.a]
0
2
4
6
8
10
80
90
100
110
130
140
150

Measurement of isotopic yields

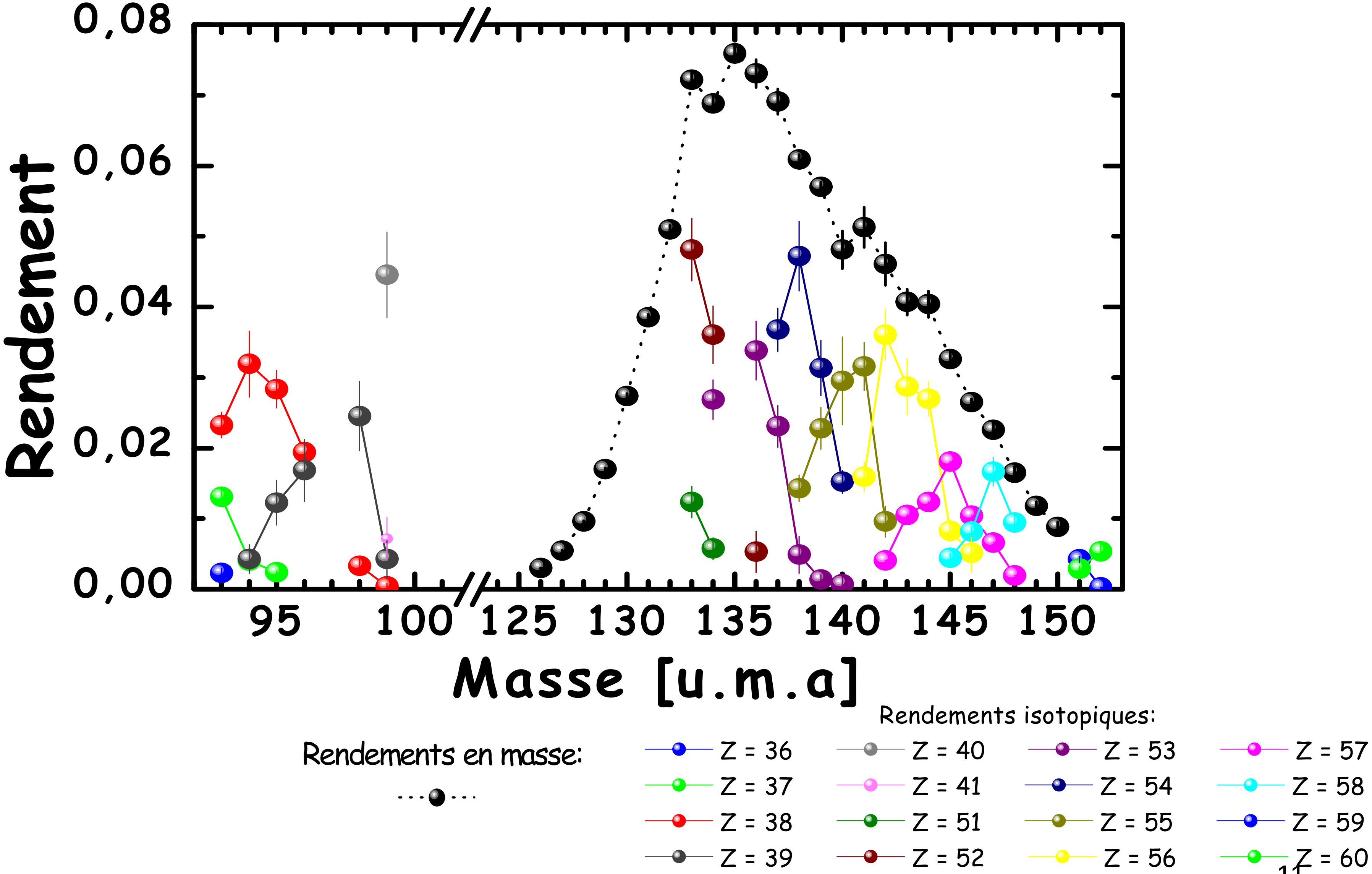


γ rays following the beta decay of ^{139}I

=> E_γ, I_γ, B.R. for each gamma ray

Neutron rich

Isotopic yields



Fission yield measurements

Measurement of mass and isotopic yields of heavy fission fragments:

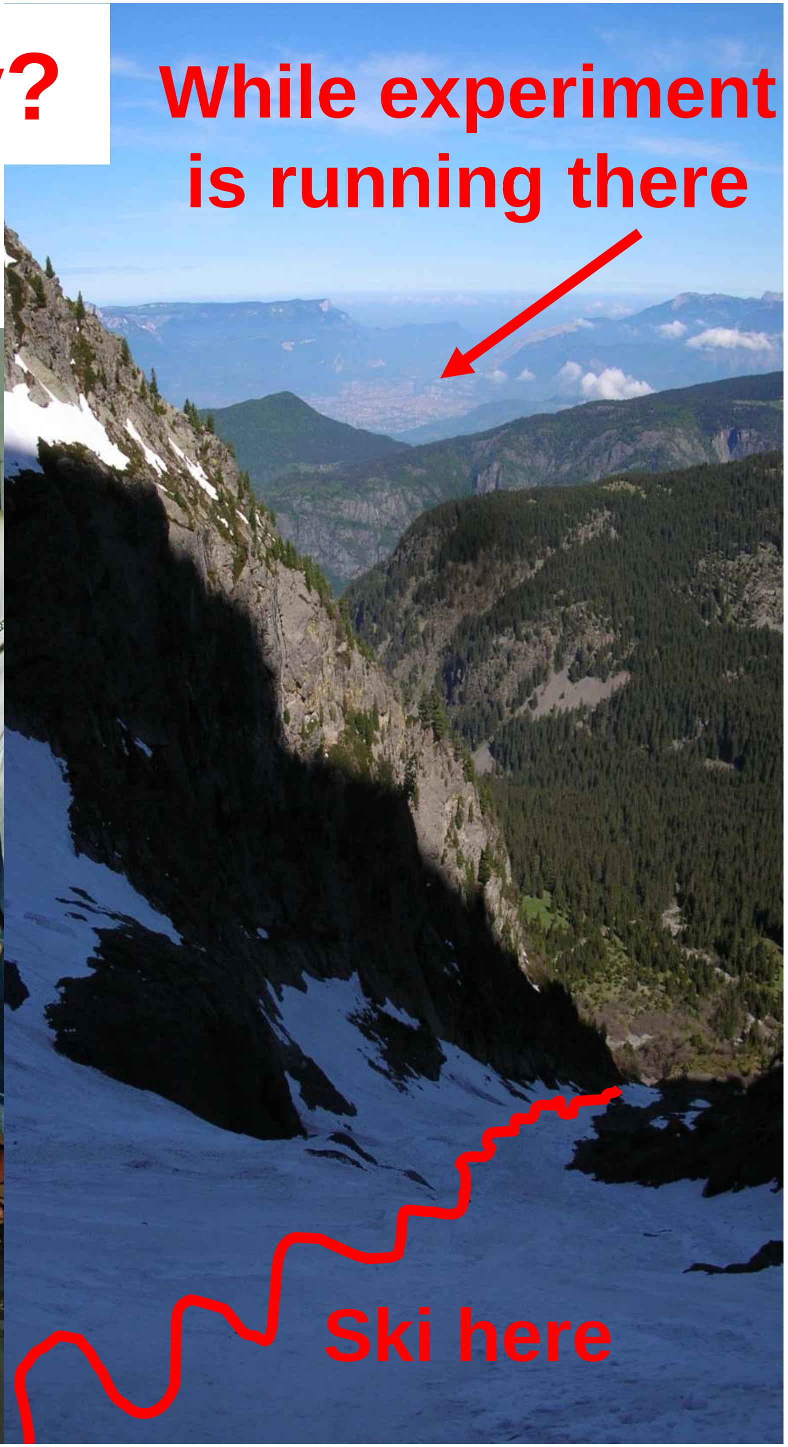
^{239}Pu(n,f) Adeline Bail, PhD thesis, Univ. Bordeaux, 2009.

Recent improvements:

- powerful Ge detectors
- new high voltage system
- independent monitoring of high voltage stability
- automated scans
- semi-automatic analysis

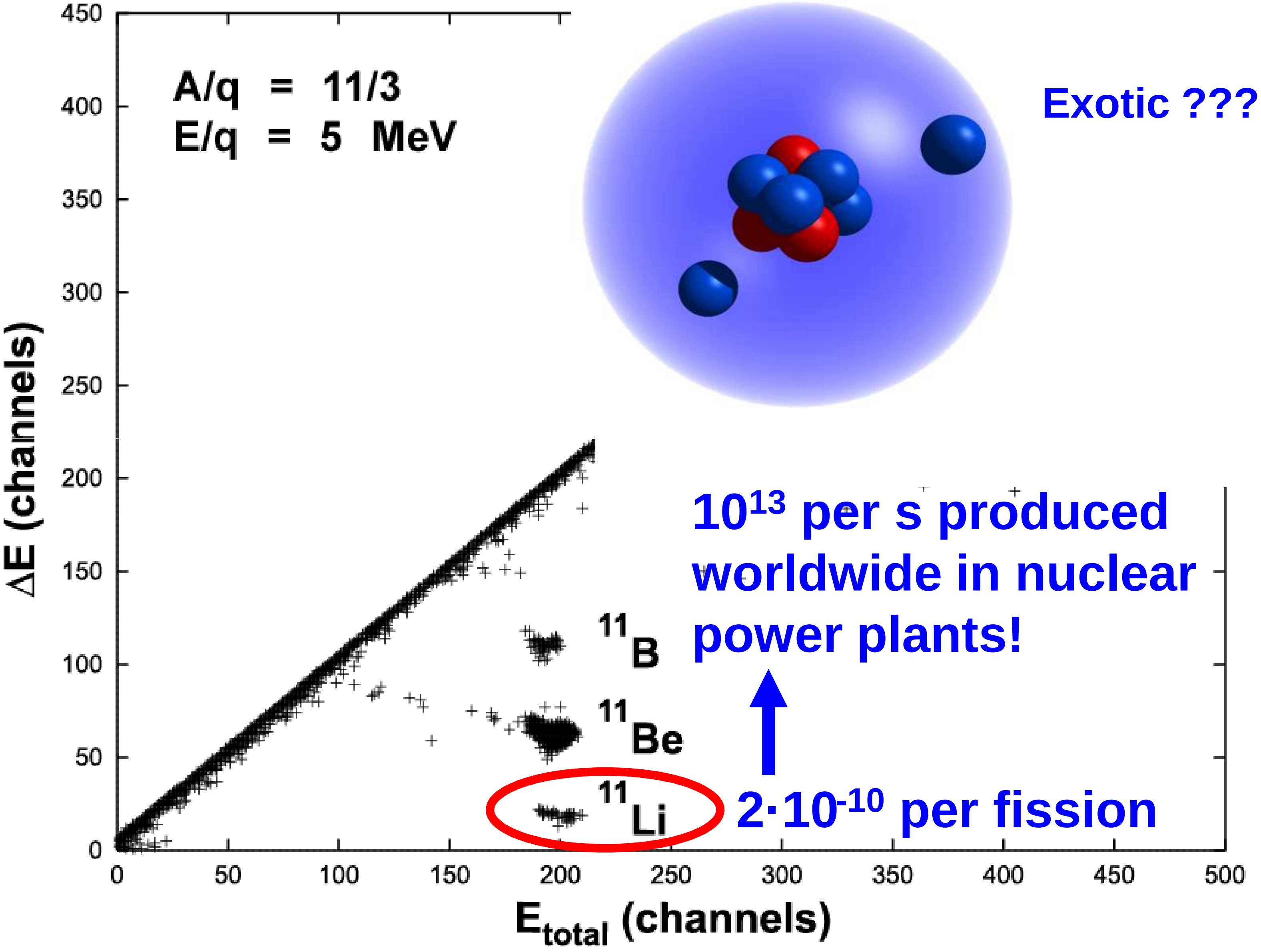
Future plans: ^{233}U(n,f), ^{241}Pu(n,f),...

Reliability?


While experiment is running there

Ski here

Detection of rare ternary particles

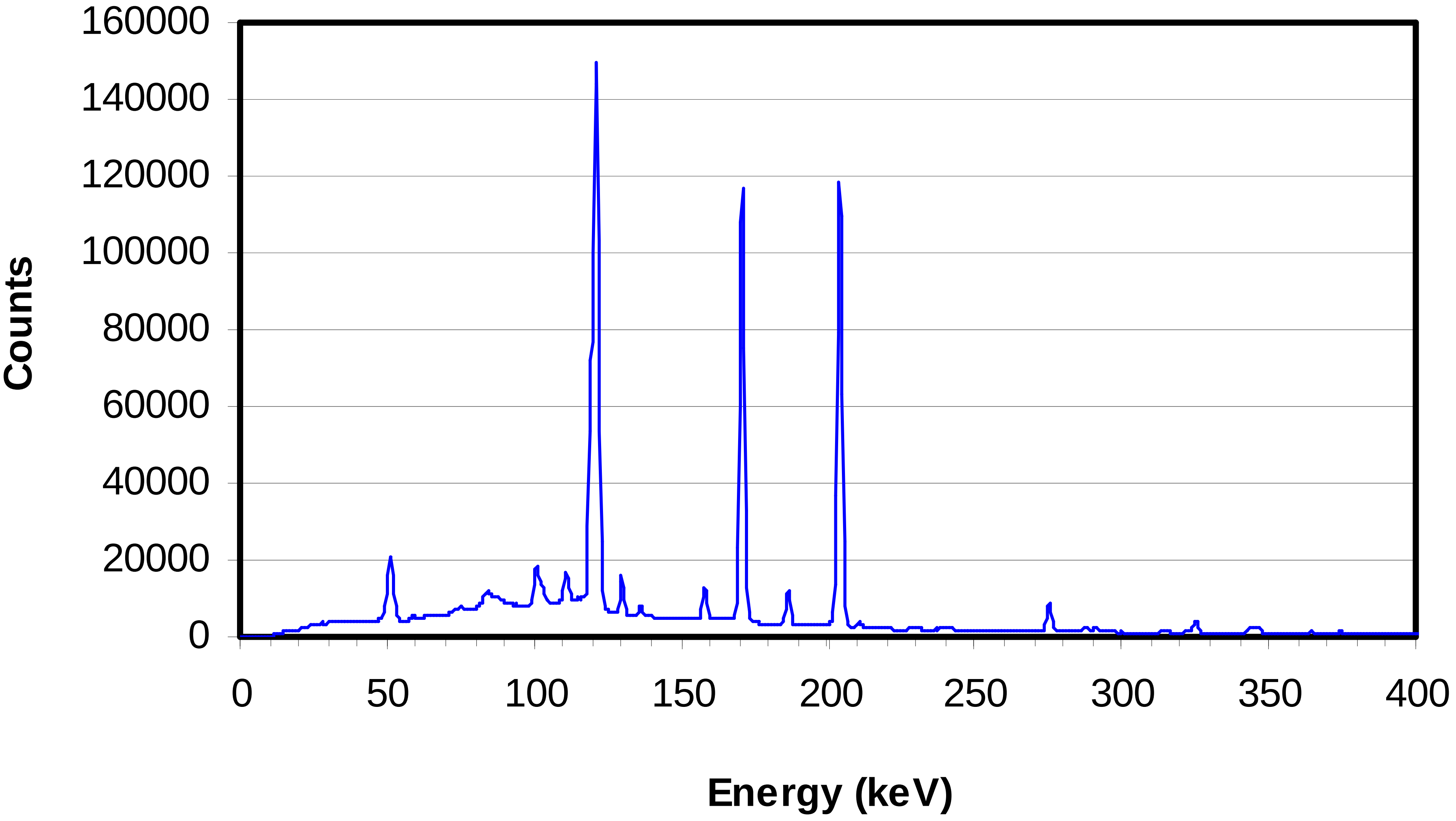


Exotic ???

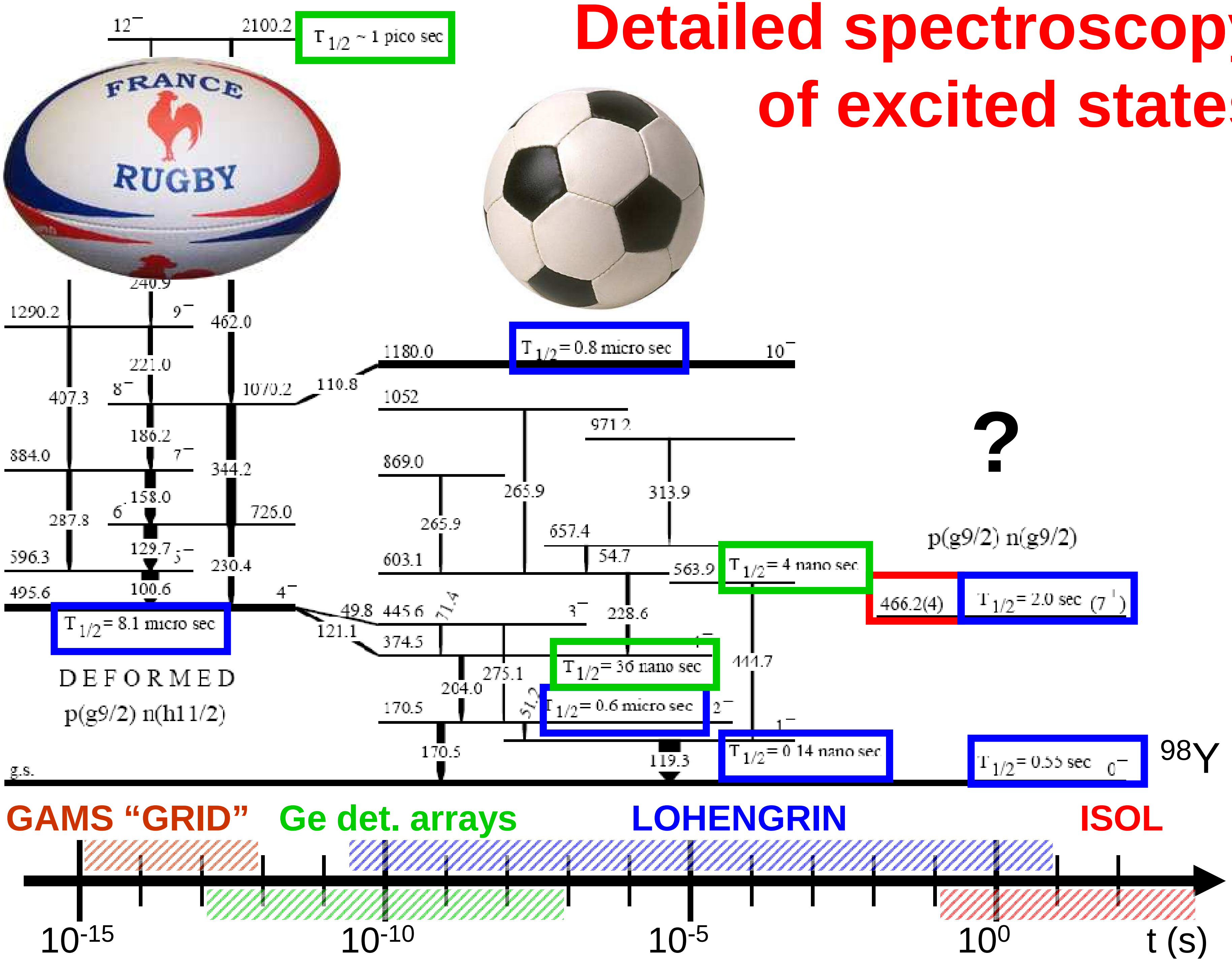
10^{13} per s produced worldwide in nuclear power plants!

$2 \cdot 10^{-10}$ per fission

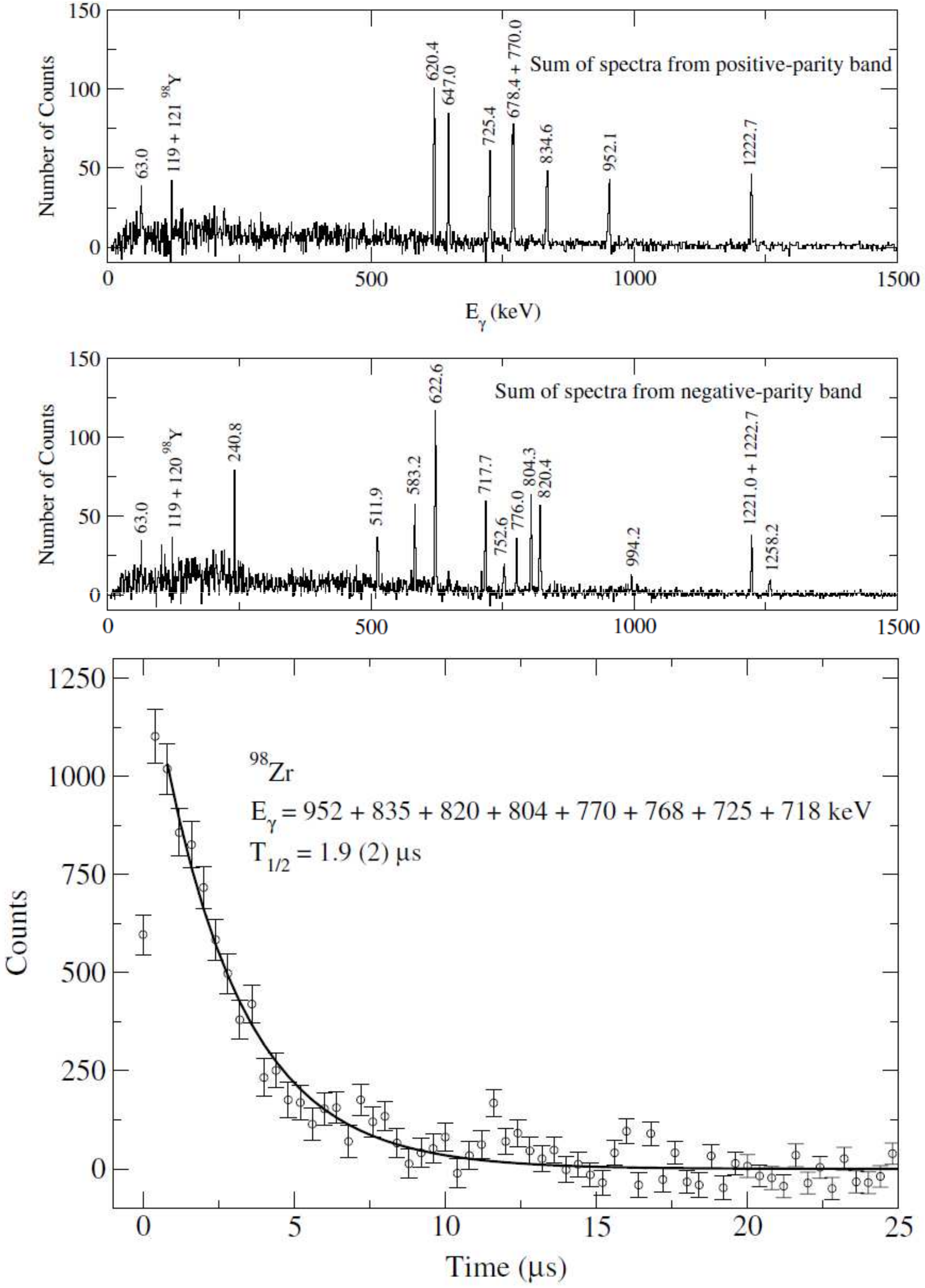
Gamma decay of 7.6 μs ^{98}Y isomer

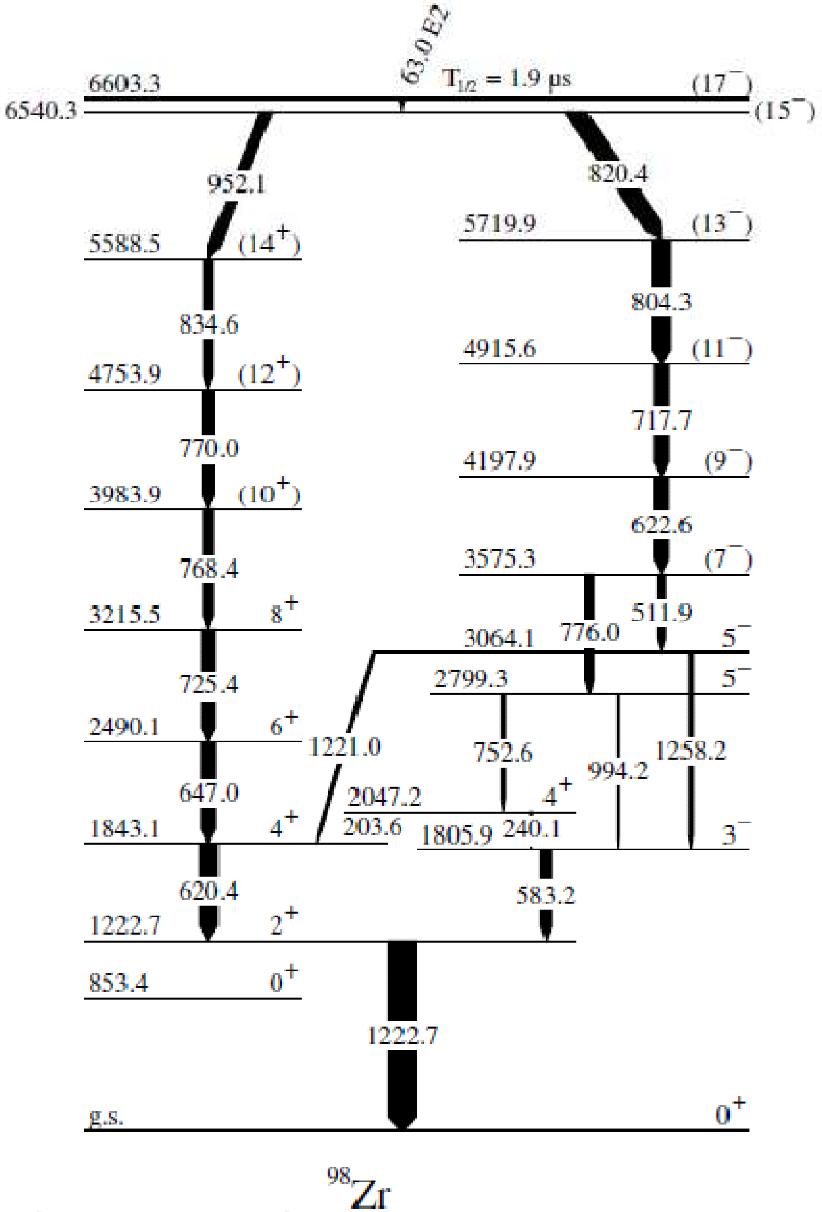


Detailed spectroscopy of excited states



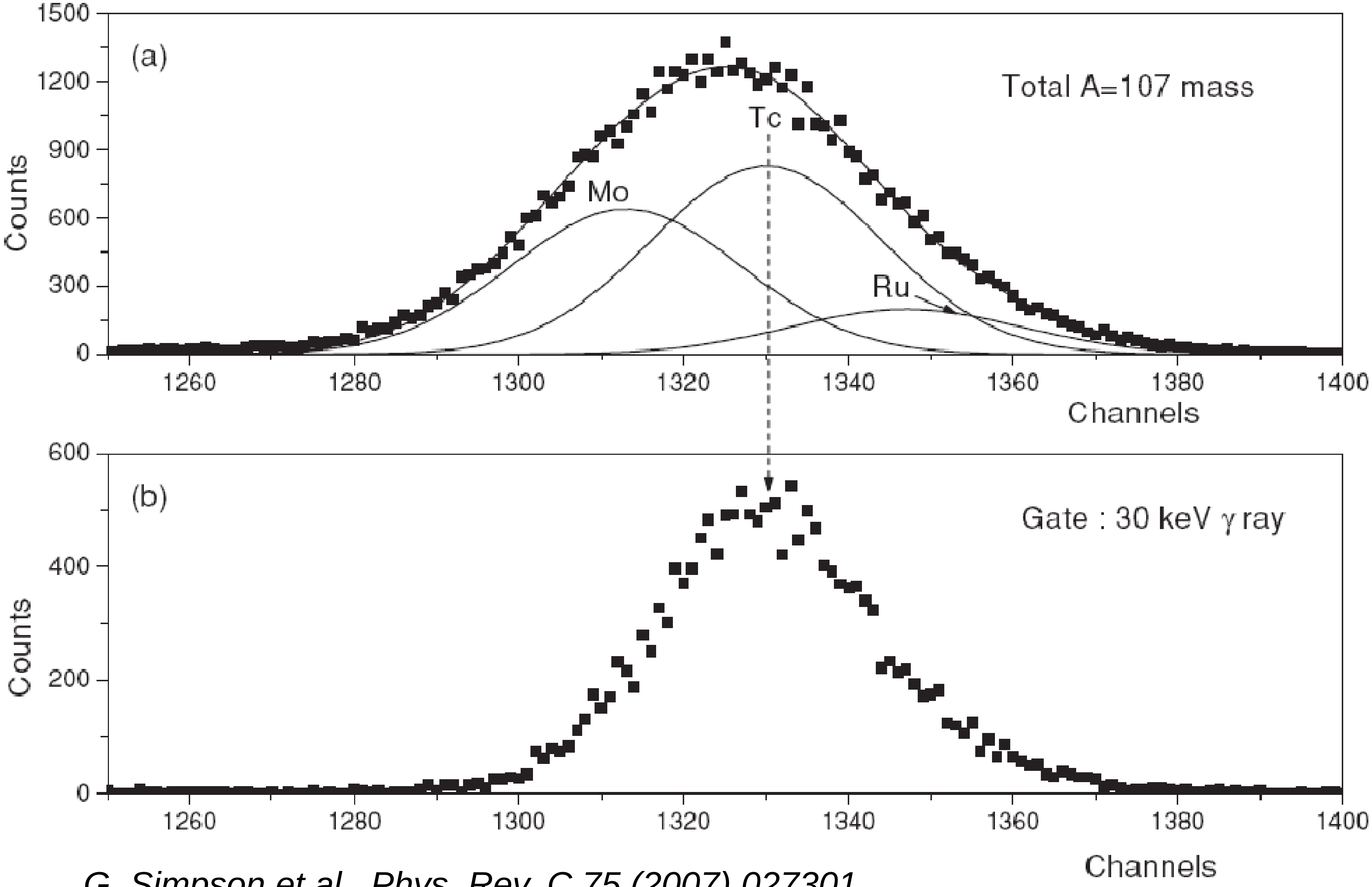
17^- isomer at 6.6 MeV in ^{98}Zr





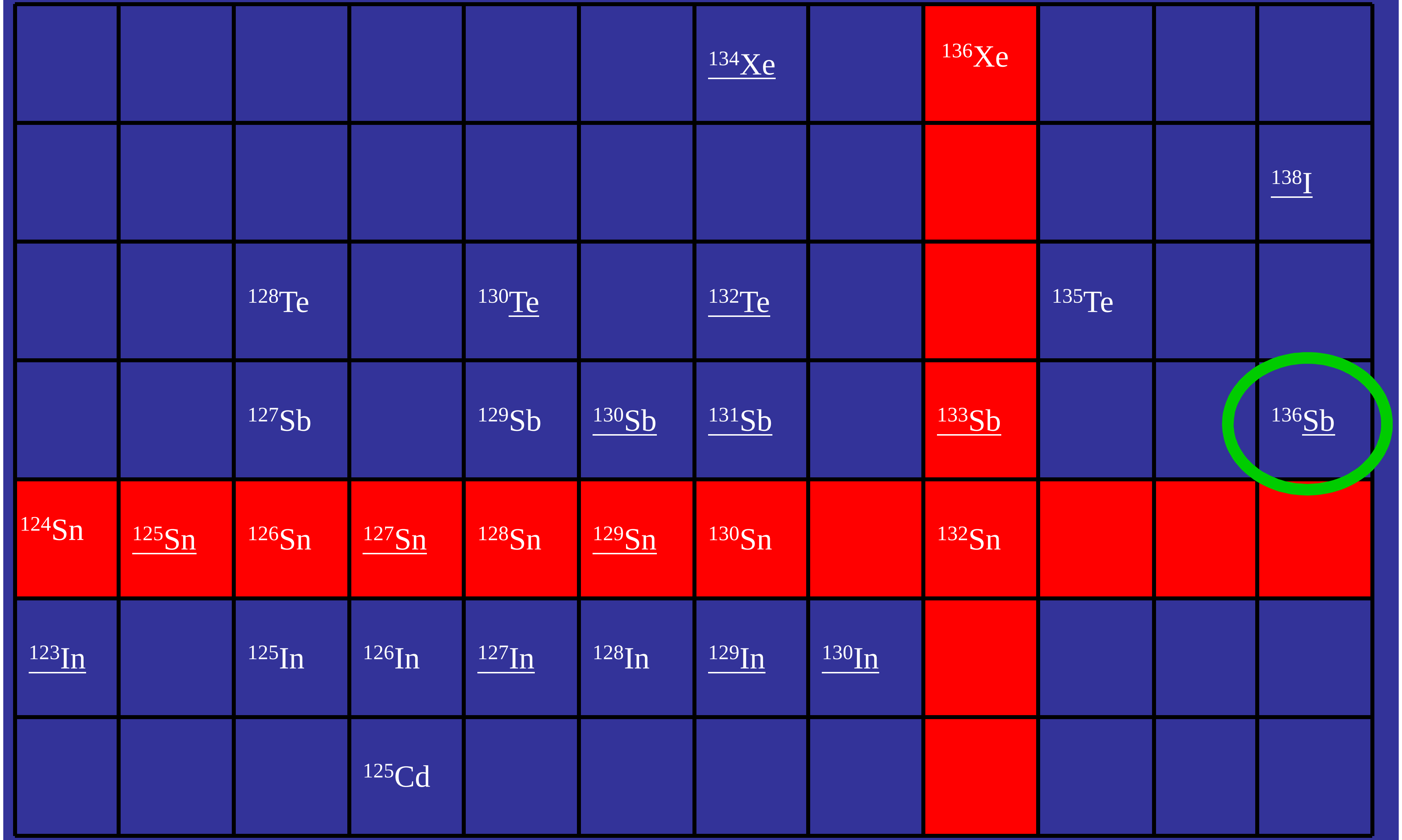
G. Simpson et al.,
Phys. Rev. C 74 (2006) 064308.

Z identification with specific energy loss



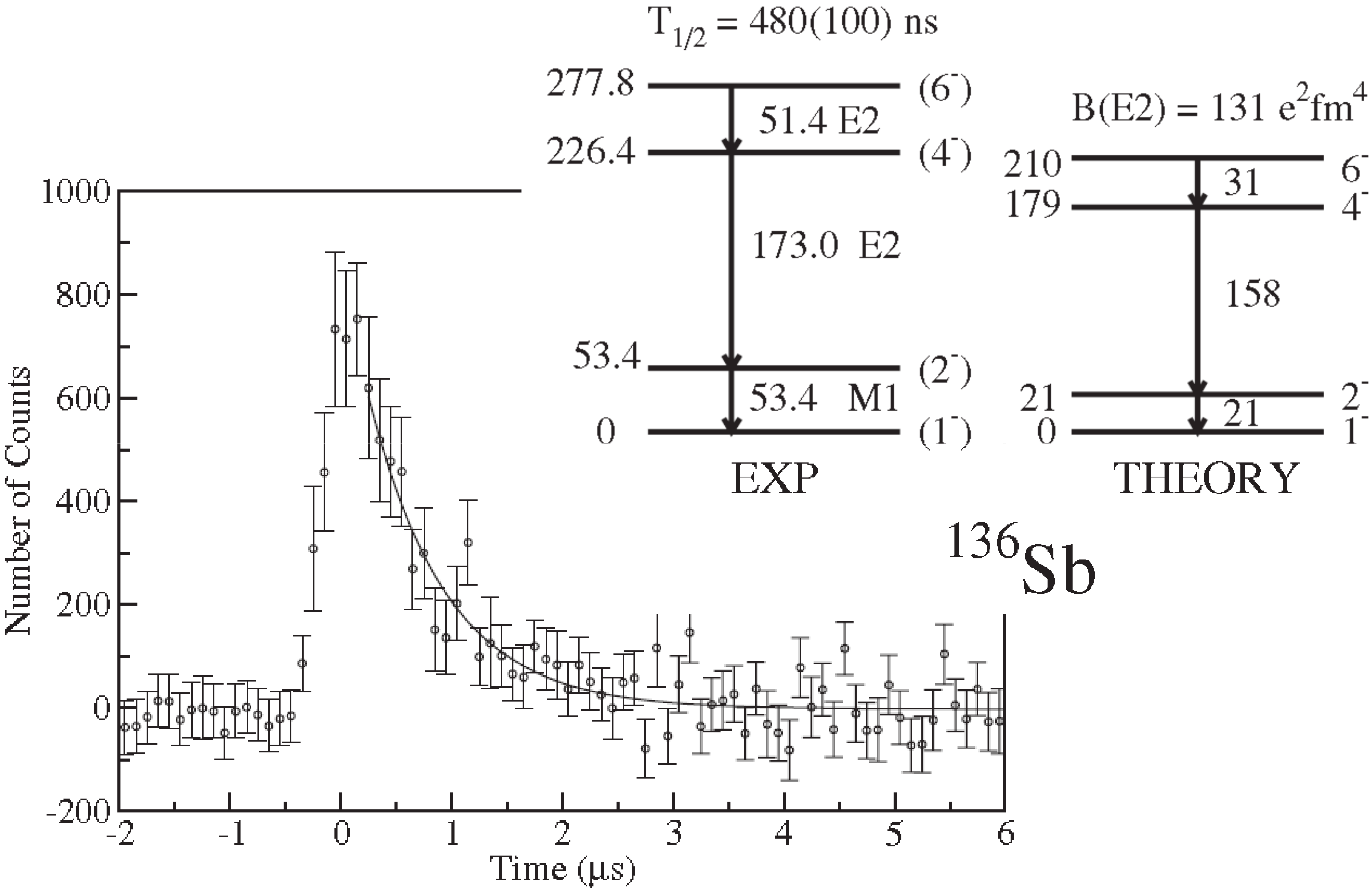
G. Simpson et al., Phys. Rev. C 75 (2007) 027301.

Microsecond isomers in the ^{132}Sn region



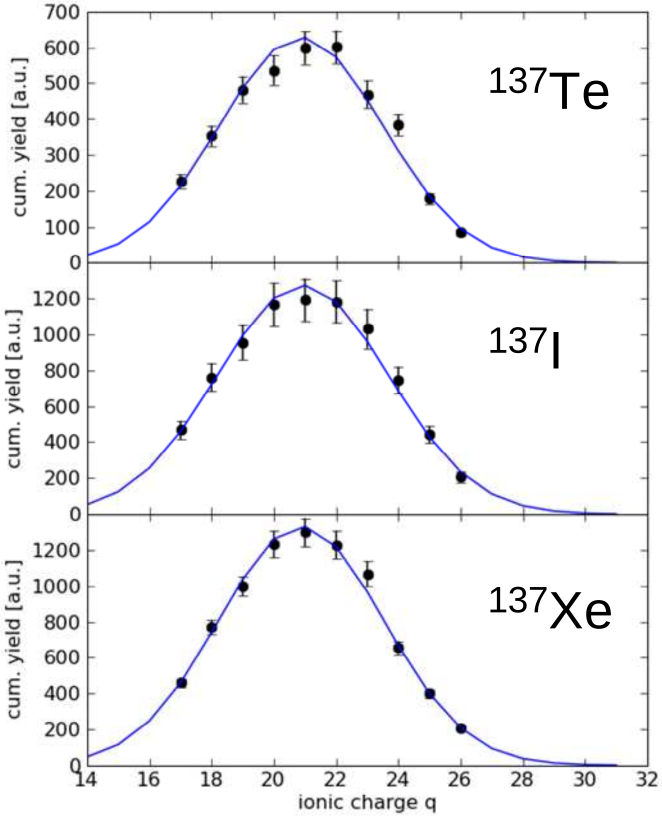
						^{134}Xe		^{136}Xe			
											^{138}I
		^{128}Te		^{130}Te		^{132}Te			^{135}Te		
		^{127}Sb		^{129}Sb	^{130}Sb	^{131}Sb		^{133}Sb			^{136}Sb
^{124}Sn	^{125}Sn	^{126}Sn	^{127}Sn	^{128}Sn	^{129}Sn	^{130}Sn		^{132}Sn			
^{123}In		^{125}In	^{126}In	^{127}In	^{128}In	^{129}In	^{130}In				
			^{125}Cd								

^{136}Sb isomer at LOHENGRIN

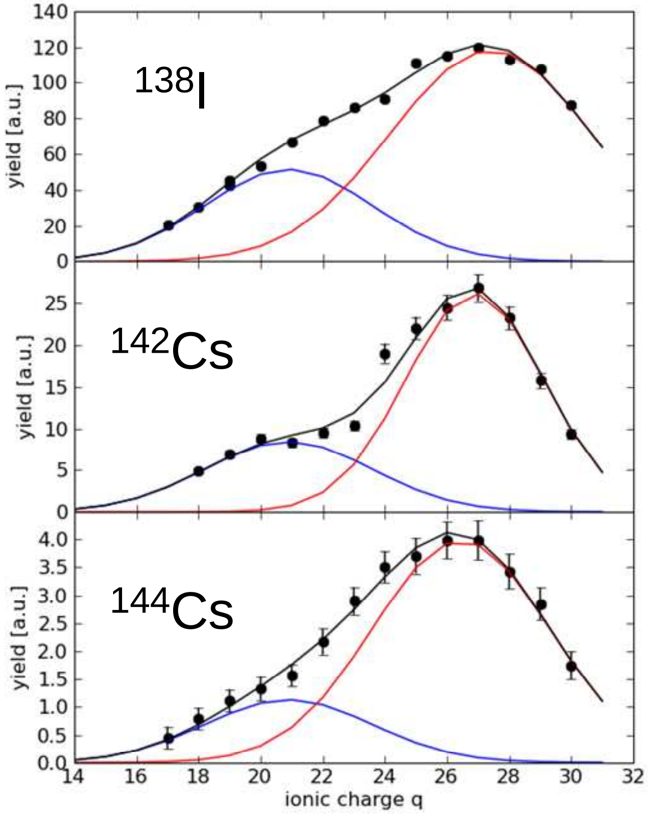


G. Simpson et al., Phys. Rev. C 76 (2007) 041303(R).

Identification of nanosecond isomers using their ionic charge distribution in the mass spectrometer



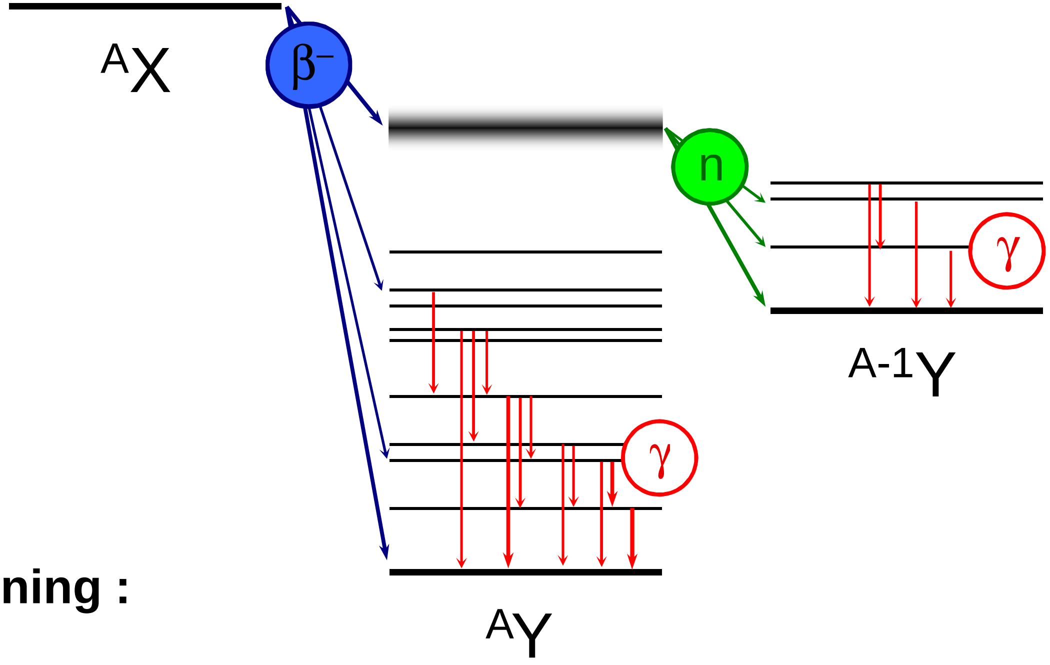
No evidence for ns-isomers in ^{137}Te, ^{137}I and ^{137}Xe



New nanosecond isomers for the exotic nuclei ^{138}I, ^{142}Cs and ^{144}Cs

Thomas Materna et al, Fission 2009, Cadarache.

Measurement of P_n values



P_n = ratio of **(β^-,n)** decay over total β^- decay

$$P_n = \frac{n}{\beta}\,\frac{\varepsilon_\beta}{\varepsilon_n}$$

Not known : determined with well known P_n values

Warning :

$\frac{\varepsilon_\beta}{\varepsilon_n}$ must be **constant** → ε_β and ε_n must be constant

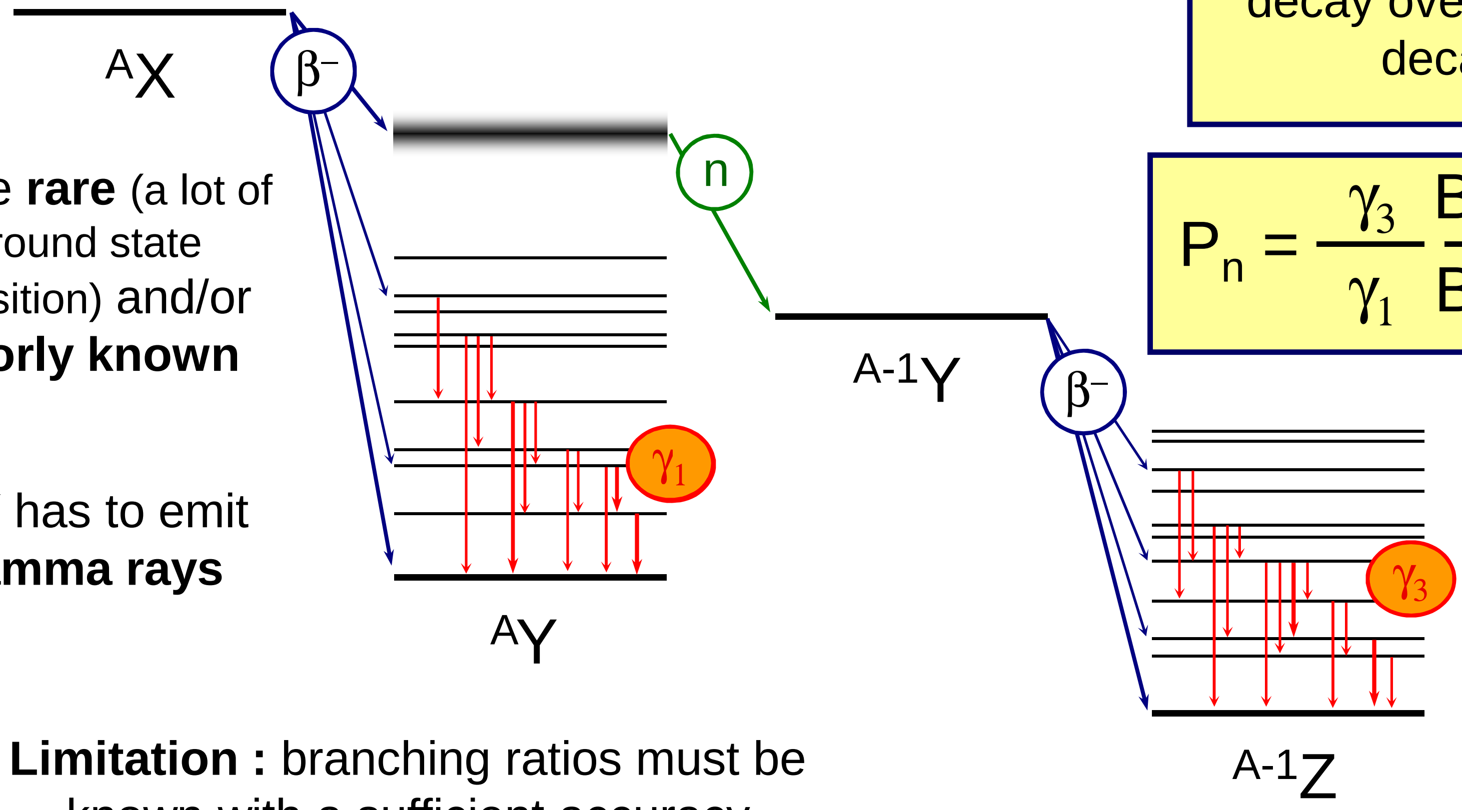
→ specific design of detectors

Analysis of the **β and n decay** required to **discriminate** other emitters

Measurement by gamma ray detection

P_n = ratio of **(β^-,n)** decay over total β^- decay

$$P_n = \frac{\gamma_3}{\gamma_1} \; \frac{BR_1 \,.\, \varepsilon_{\gamma(E1)}}{BR_3 \,.\, \varepsilon_{\gamma(E3)}}$$



γ_2 are **rare** (a lot of ground state transition) and/or **poorly known**

$^{A-1}$Y has to emit **gamma rays**

Limitation : branching ratios must be known with a sufficient accuracy

Main problems with the **less stable** nuclei (^{A}X)

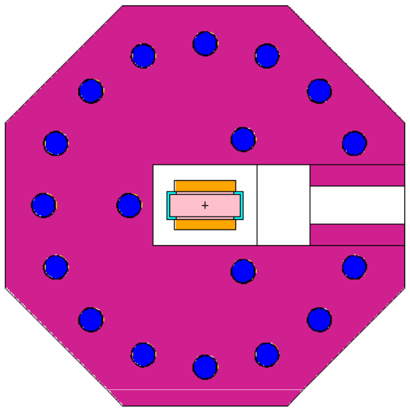
→ **independant** method

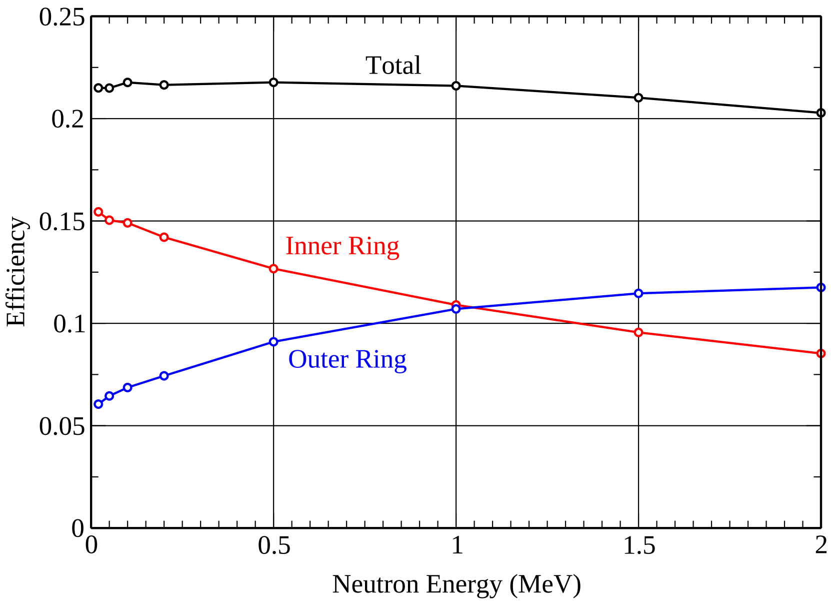
only usable for a **limited number** of P_n values

New neutron detector design for LOHENGRIN

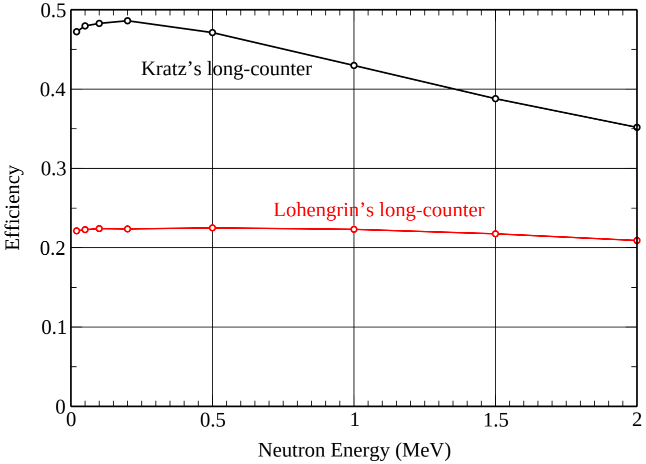
Simulations with MCNP code

Lohengrin's long-counter





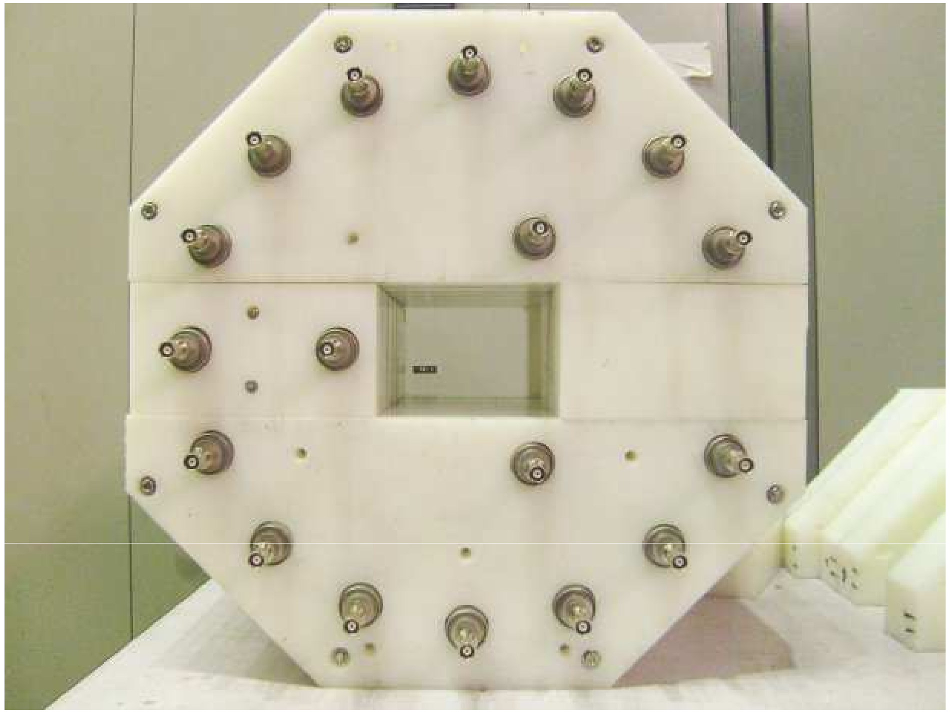
Kratz's VS Lohengrin's



➡ Negligible differences in ε_n

Lohengrin's long-counter: lower efficiency but better characteristics for P_n measurements

New neutron detector



June 2009:

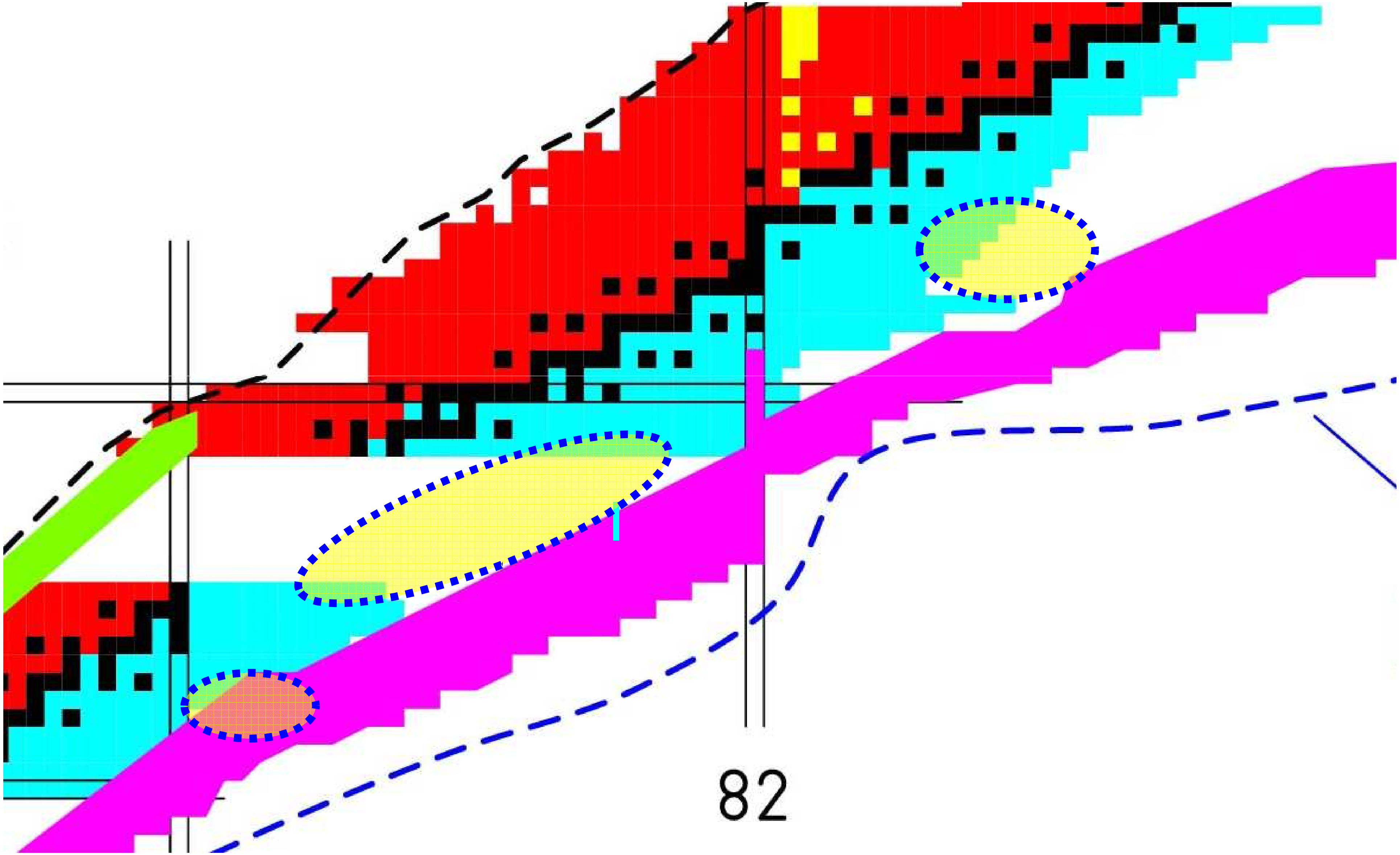
- **first use of the long-counter**
- **P_n measurement at Lohengrin**

Future:

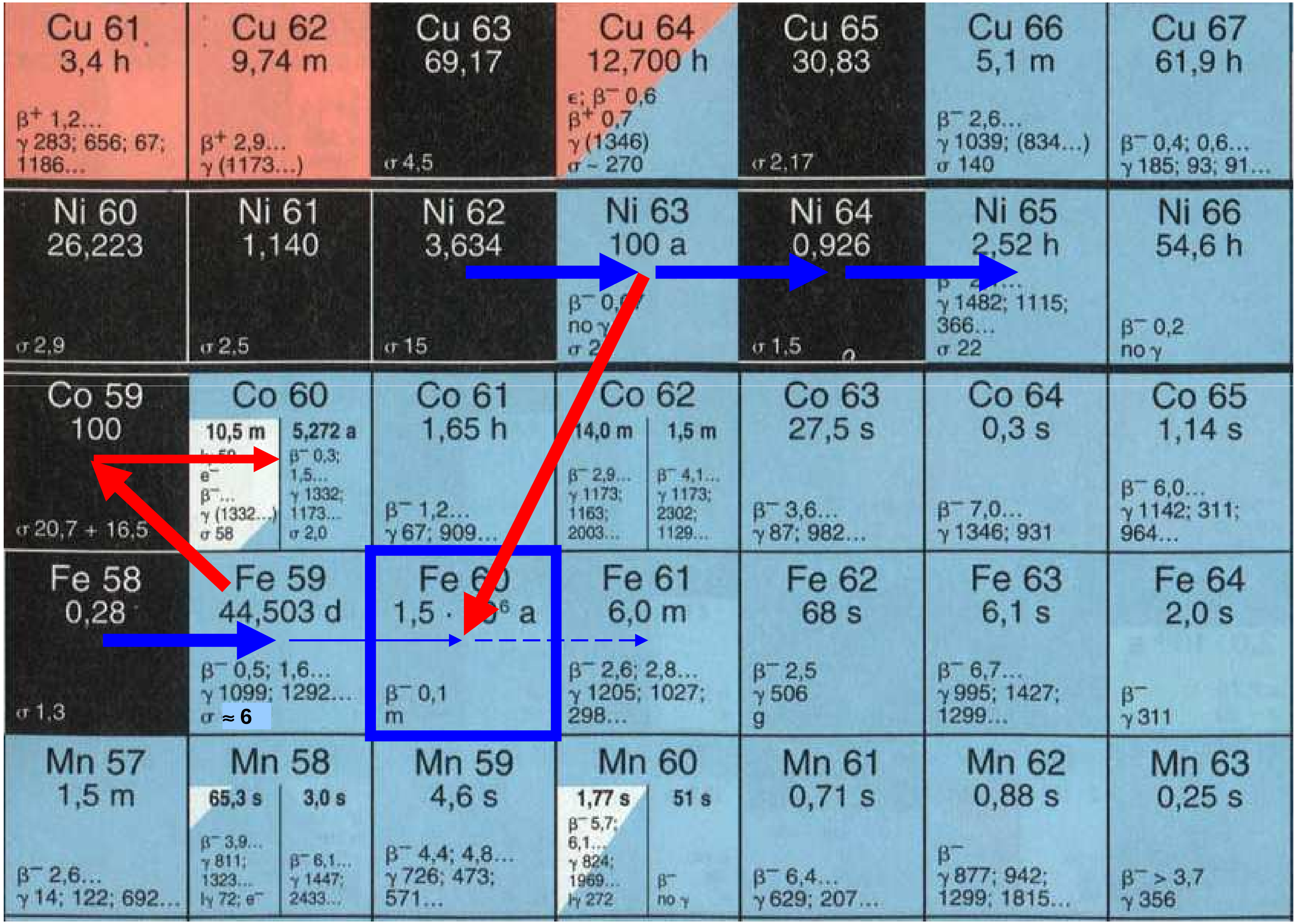
- **improved shielding against background**
- **improved beta detectors**
- **measurement of $Y*P_n$ for very neutron-rich isotopes**

Ludovic Mathieu et al., CEA Cadarache > CENBG

Nuclear chart at ISOLDE

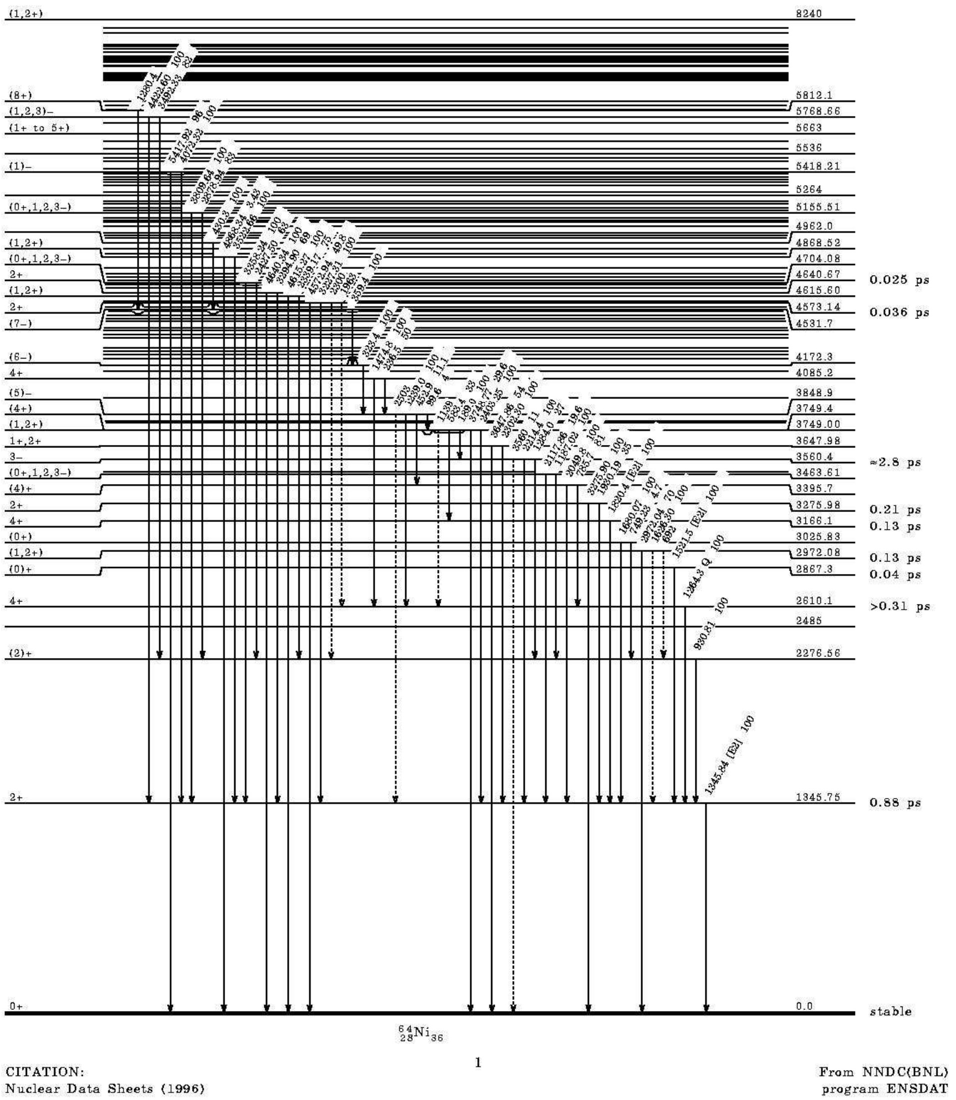


^{58}Fe(n,γ)^{59}Fe(n,γ)^{60}Fe and ^{62}Ni(n,γ)^{63}Ni(n,α)^{60}Fe



^{64}Ni level scheme

S_n = 9658 keV



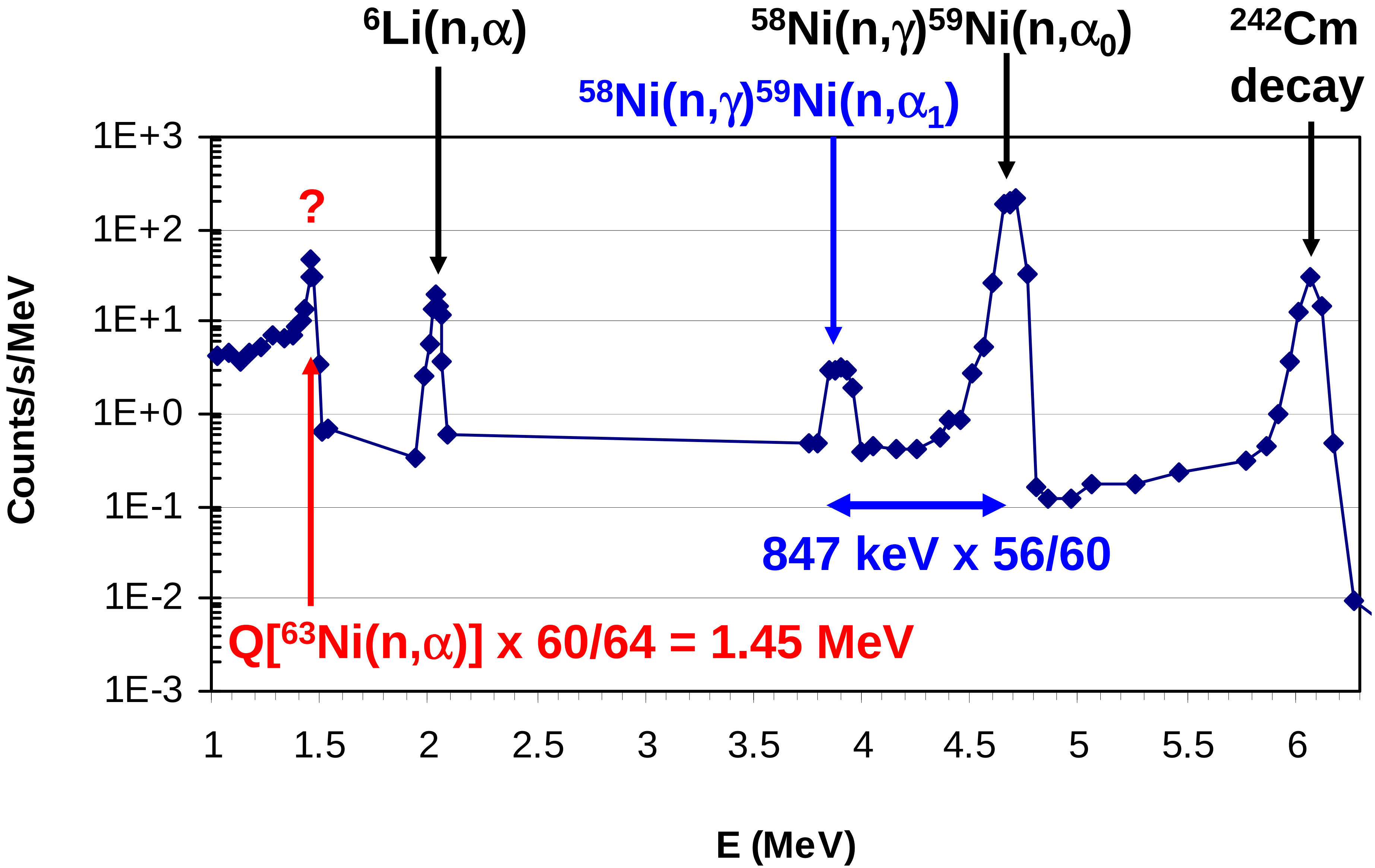
CITATION:
Nuclear Data Sheets (1996)

From NNDC(BNL)
program ENSDAT

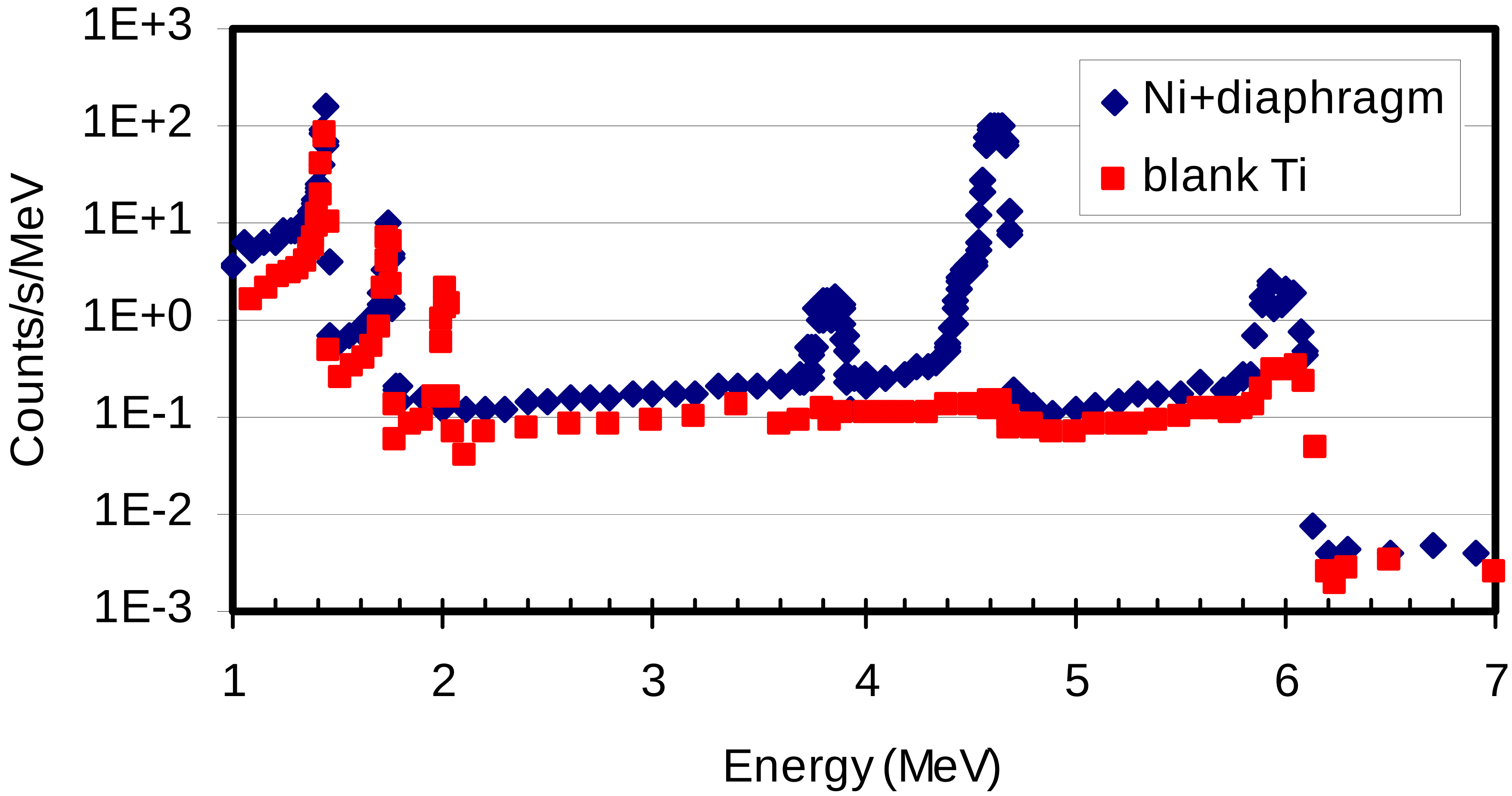
8240 keV

0 keV

$^{nat}Ni(n,\gamma)(n,\alpha)$

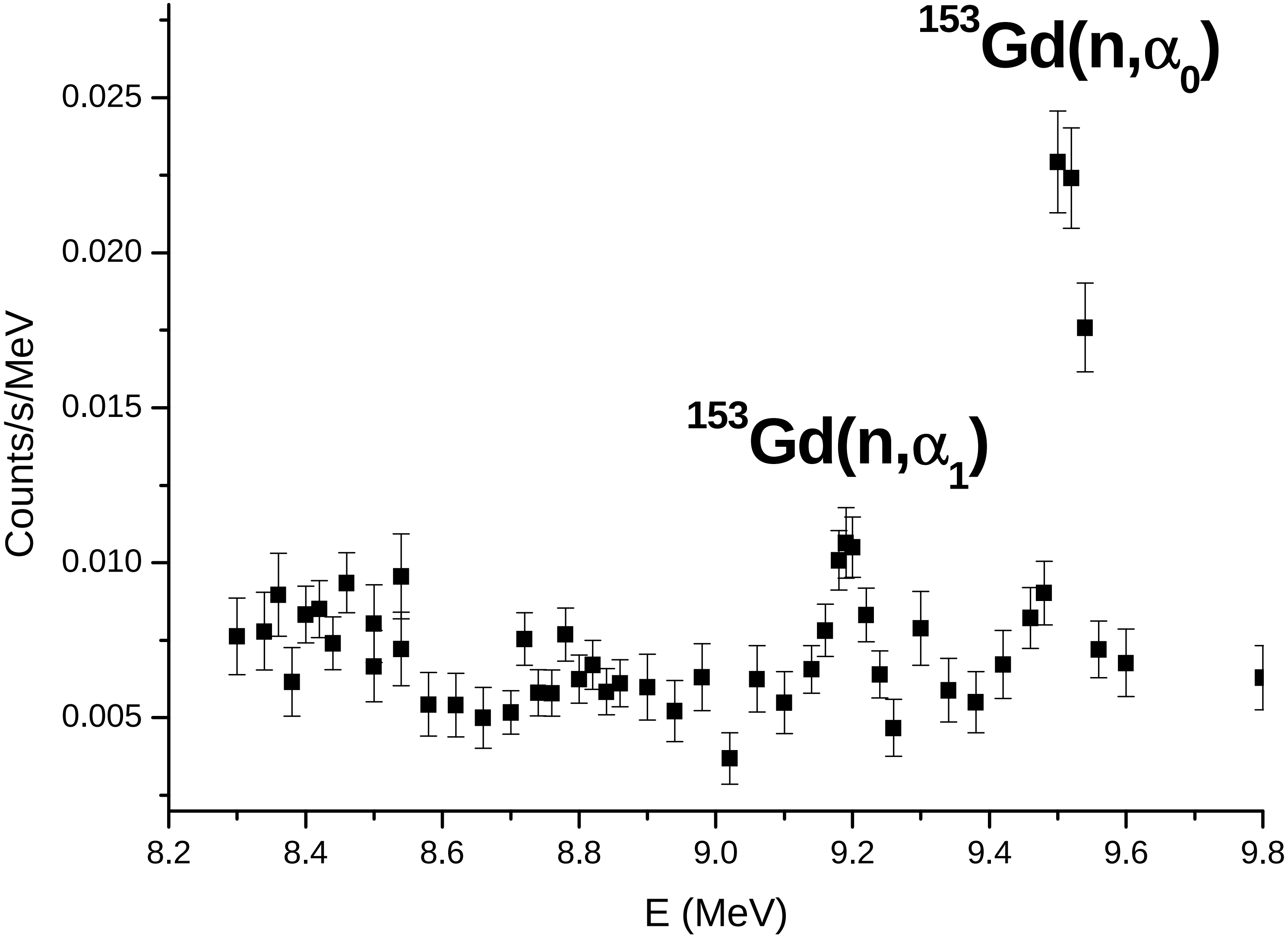


$^{nat}Ni(n,\gamma)(n,\alpha)$

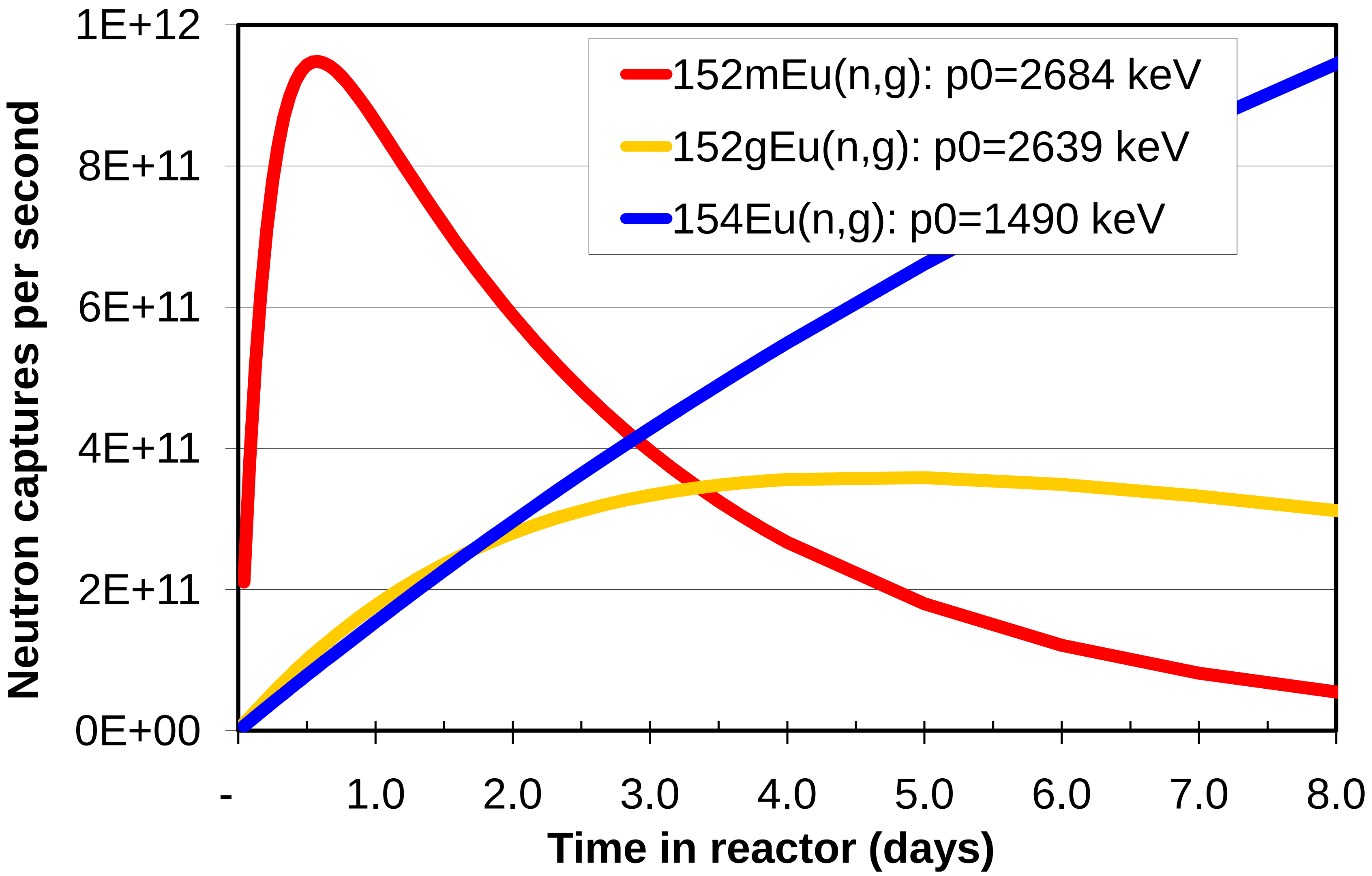


After irradiation to 10^{22} n/cm^2 off-line analysis of ^{60}Fe content by accelerator mass spectrometry ⇒ μbarn sensitivity

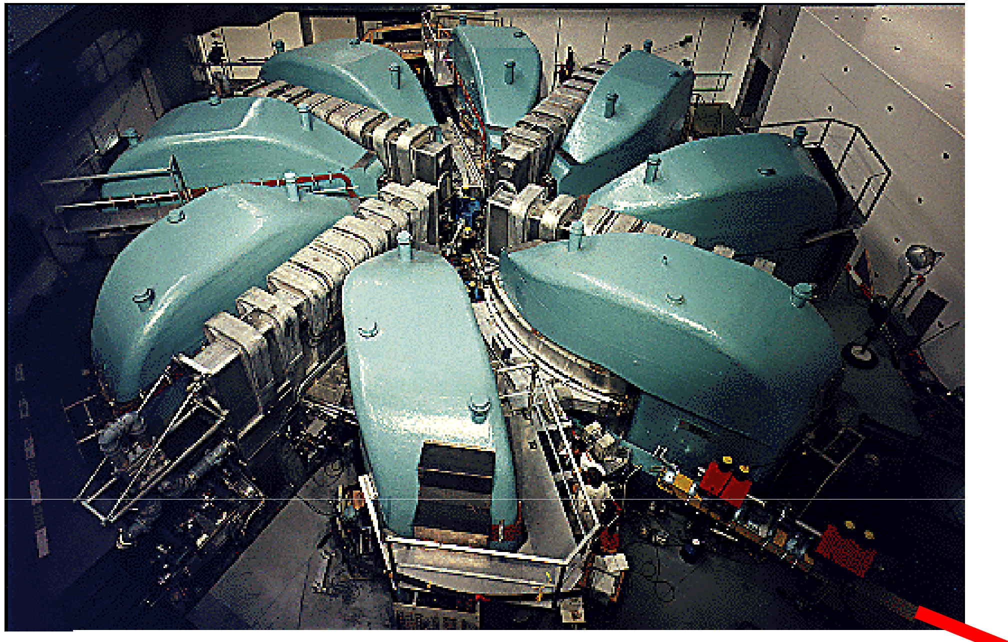
(n,α) spectroscopy with LOHENGRIN

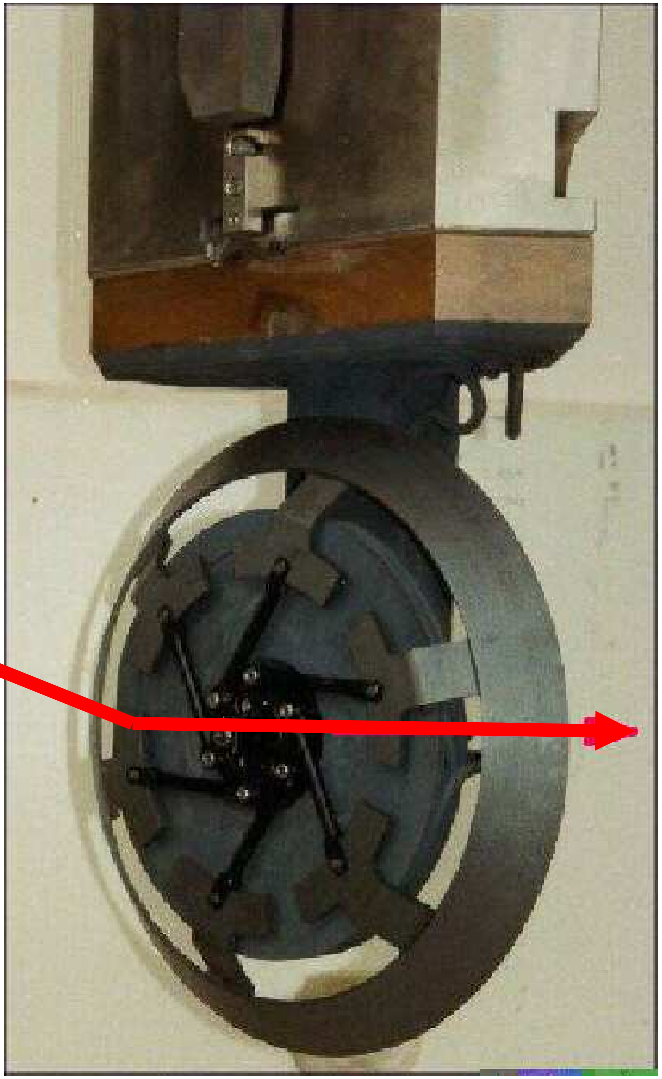


Identification of ^{152g}Eu(n,p_1) and ^{152m}Eu(n,p_0)



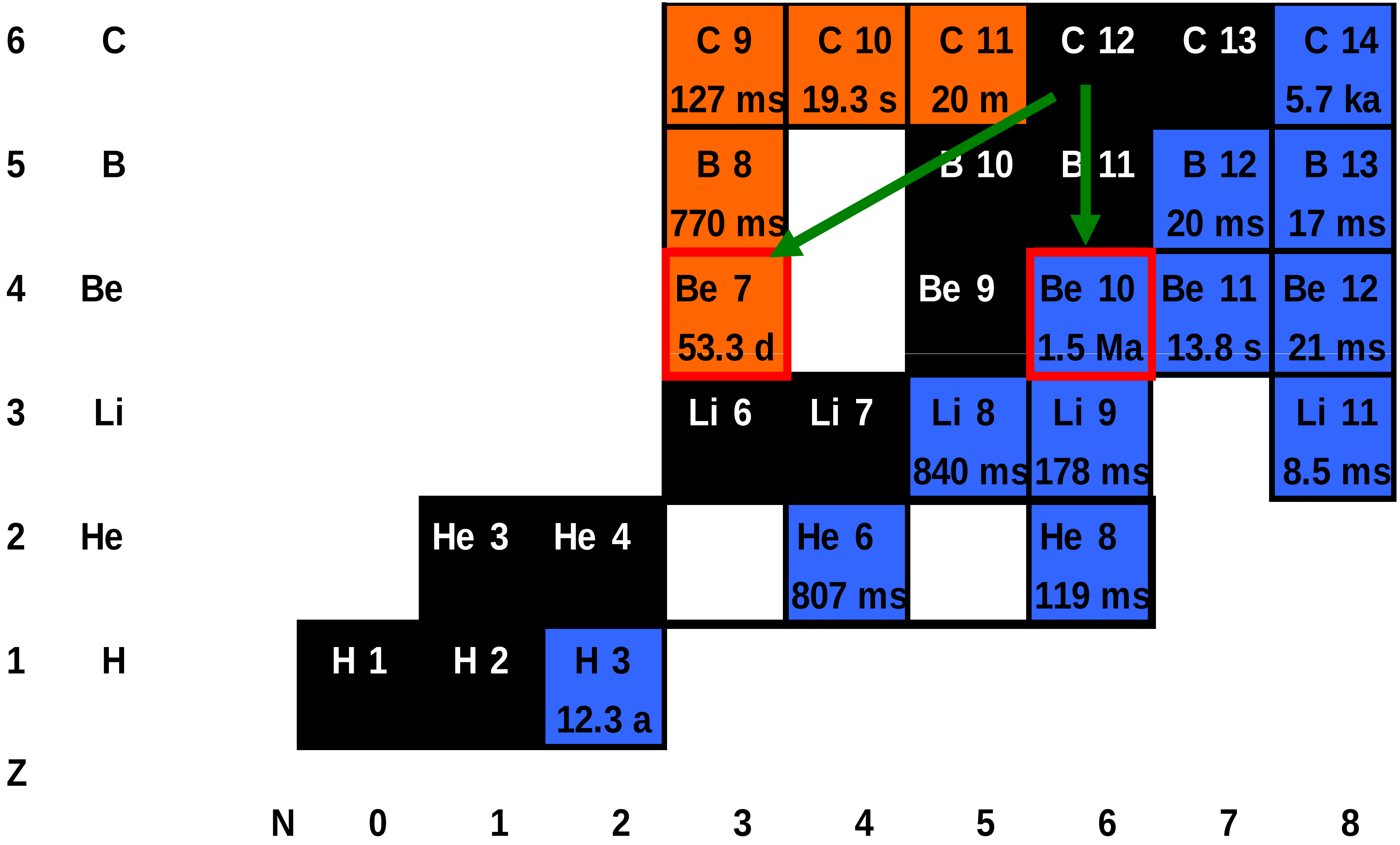
Intense ^{7}Be beam at ISOLDE





PSI: 2 mA 590 MeV protons onto graphite target for pion production

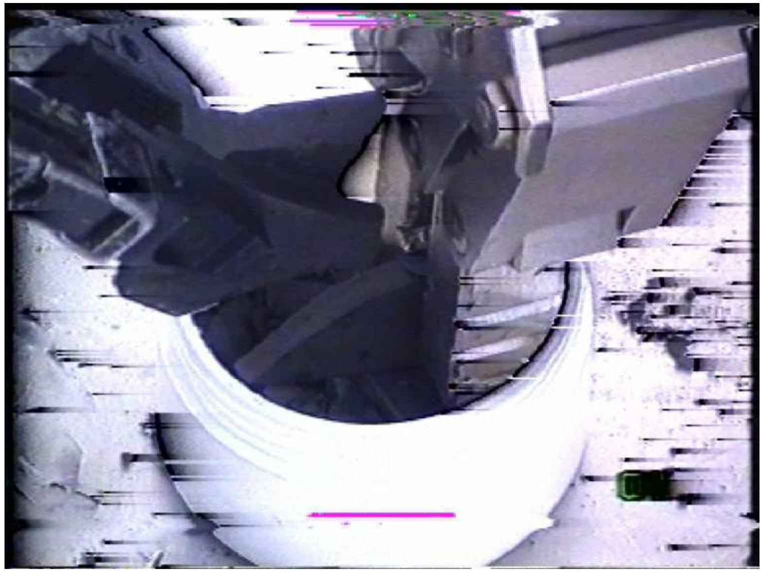
Spallation products



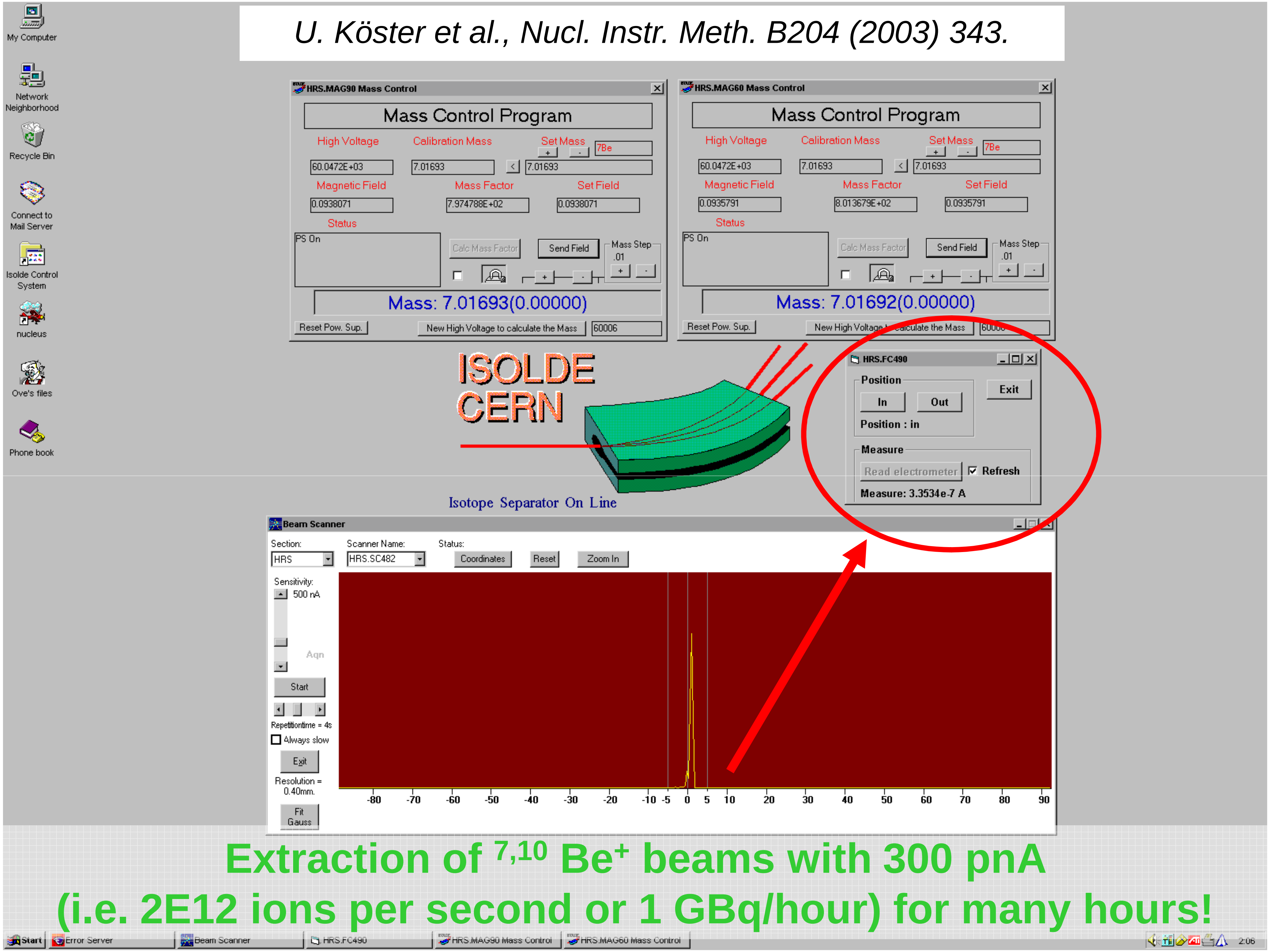
Z		0	1	2	3	4	5	6	7	8
6	C				C 9 127 ms	C 10 19.3 s	C 11 20 m	C 12	C 13	C 14 5.7 ka
5	B				B 8 770 ms		B 10	B 11	B 12 20 ms	B 13 17 ms
4	Be				Be 7 53.3 d		Be 9	Be 10 1.5 Ma	Be 11 13.8 s	Be 12 21 ms
3	Li				Li 6	Li 7	Li 8 840 ms	Li 9 178 ms		Li 11 8.5 ms
2	He			He 3	He 4		He 6 807 ms		He 8 119 ms	
1	H		H 1	H 2	H 3 12.3 a					

Procedure

1. **Break graphite into pieces**
2. **Put into Pb-shielded container**
3. **Transport to ISOLDE**
4. **Fill ISOLDE target container**
5. **Heat container to 1700 ℃**
6. **Ionize Be with RILIS**



U. Köster et al., Nucl. Instr. Meth. B204 (2003) 343.



HRS.MAG90 Mass Control

Mass Control Program

High Voltage: 60.0472E+03
Calibration Mass: 7.01693
Set Mass: 7Be
7.01693
Magnetic Field: 0.0938071
Mass Factor: 7.974788E+02
Set Field: 0.0938071
Status: PS On
Mass Step: .01
Mass: 7.01693(0.00000)
60006

HRS.MAG60 Mass Control

Mass Control Program

High Voltage: 60.0472E+03
Calibration Mass: 7.01693
Set Mass: 7Be
7.01692
Magnetic Field: 0.0935791
Mass Factor: 8.013679E+02
Set Field: 0.0935791
Status: PS On
Mass Step: .01
Mass: 7.01692(0.00000)

HRS.FC490

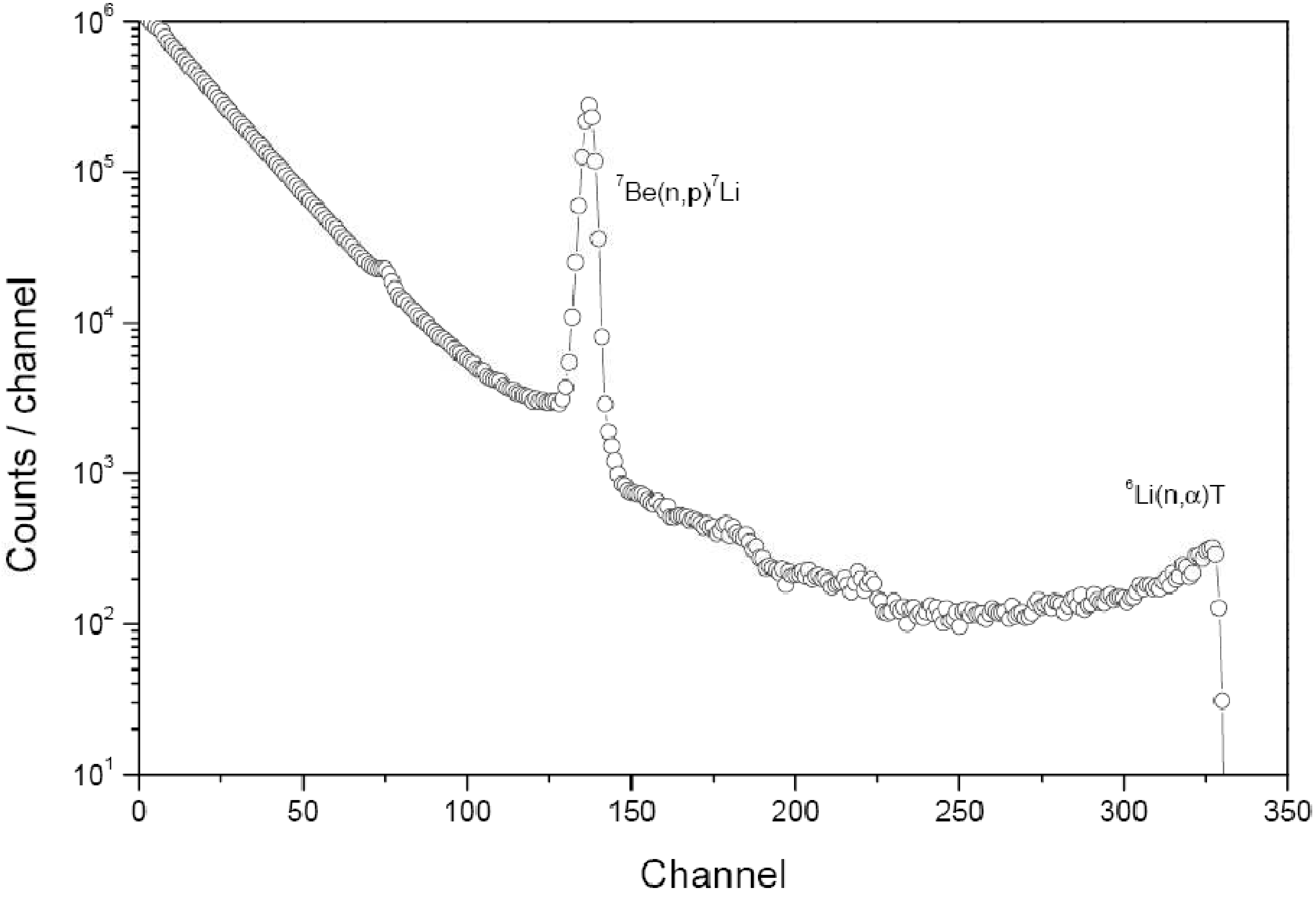
Position : in
☑ Refresh
Measure: 3.3534e-7 A

Beam Scanner

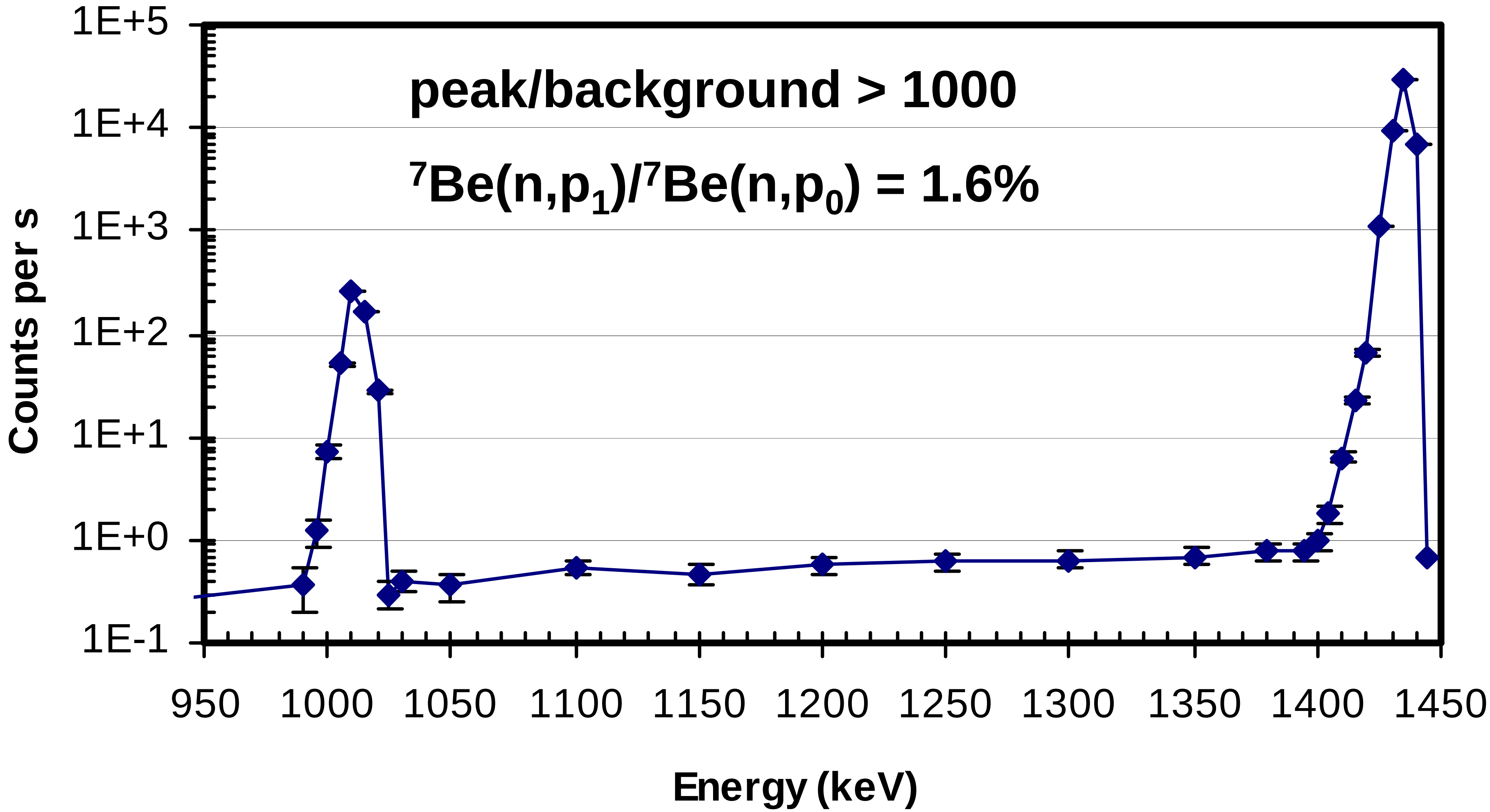
Section: HRS
Scanner Name: HRS.SC482
Status:
Sensitivity: 500 nA
Repetitiontime = 4s
☐ Always slow
Resolution = 0.40mm.

Extraction of ^{7,10} Be^{+} beams with 300 pnA
(i.e. 2E12 ions per second or 1 GBq/hour) for many hours!

^{7}Be(n,p) spectrum measured at neutron beam



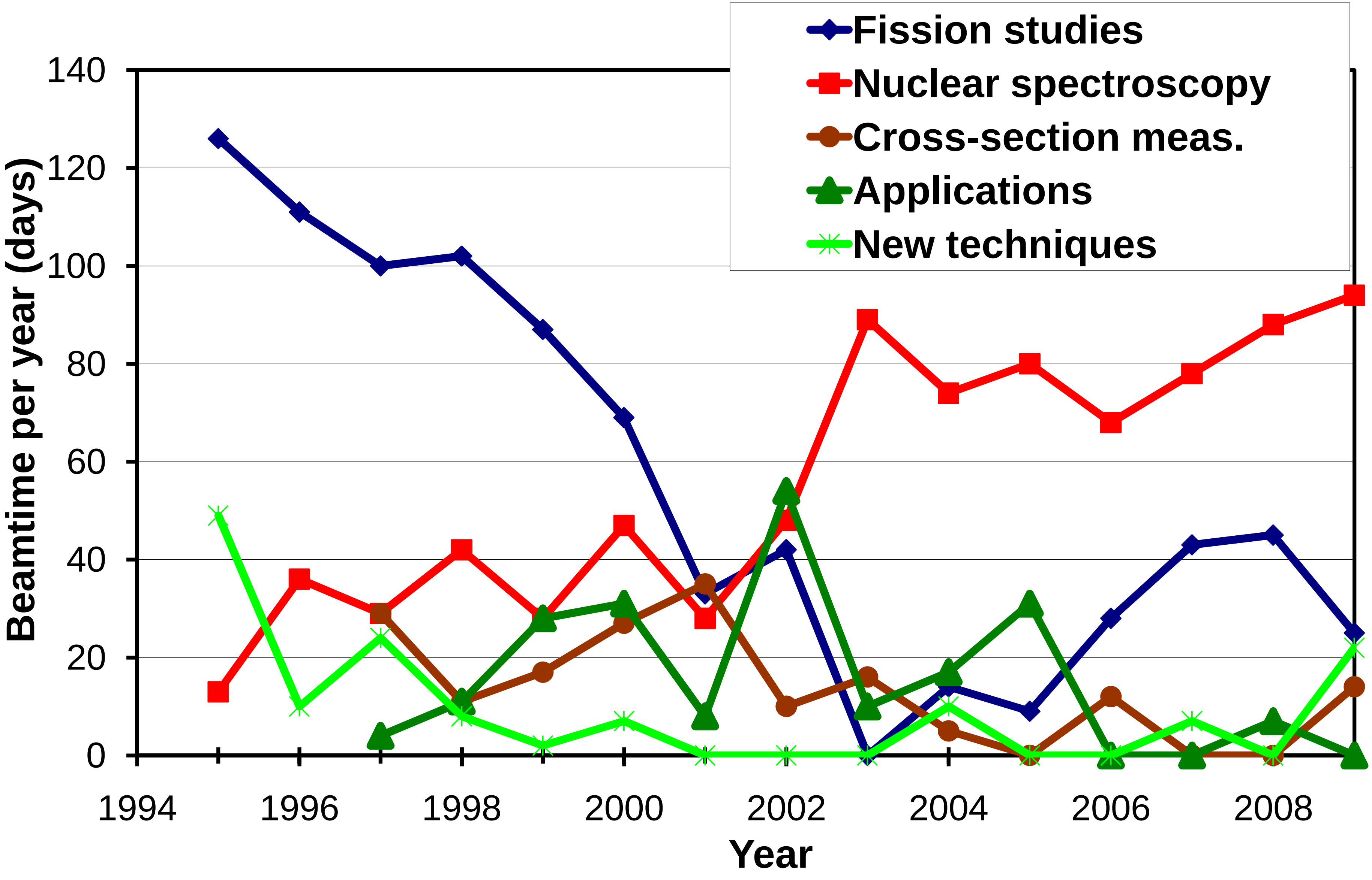
^{7}Be(n,p) measured at LOHENGRIN



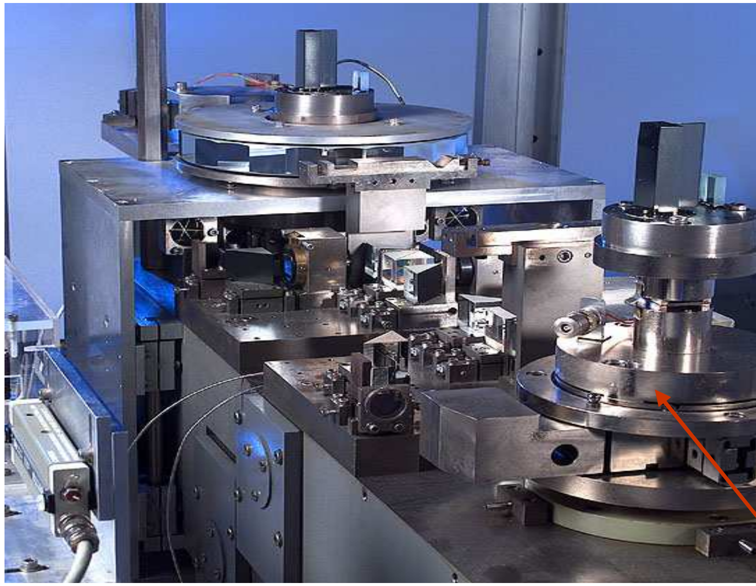
peak/background > 1000

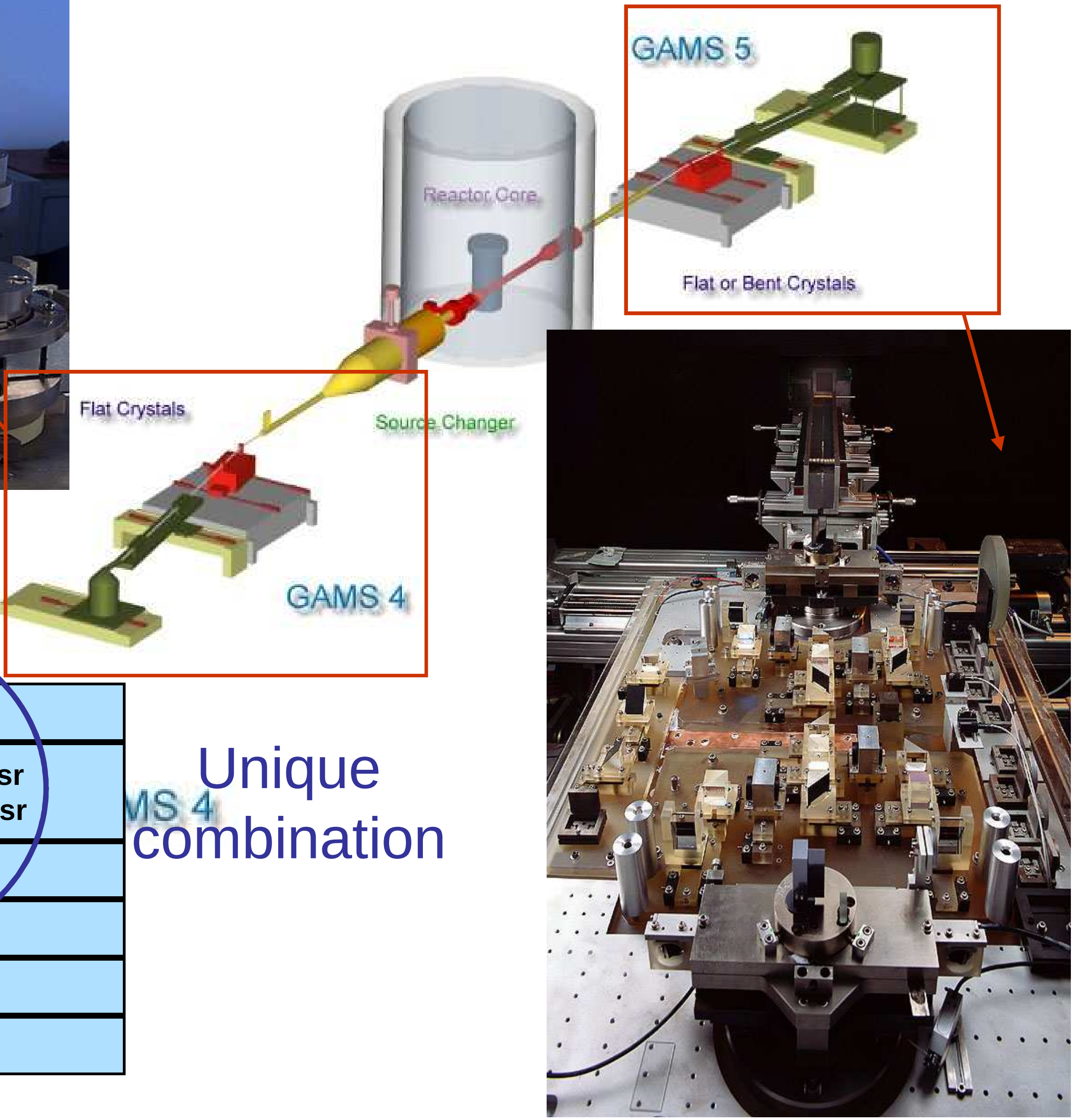
^{7}Be(n,p_1)/^{7}Be(n,p_0) = 1.6%

LOHENGRIN use



The GAMS spectrometers

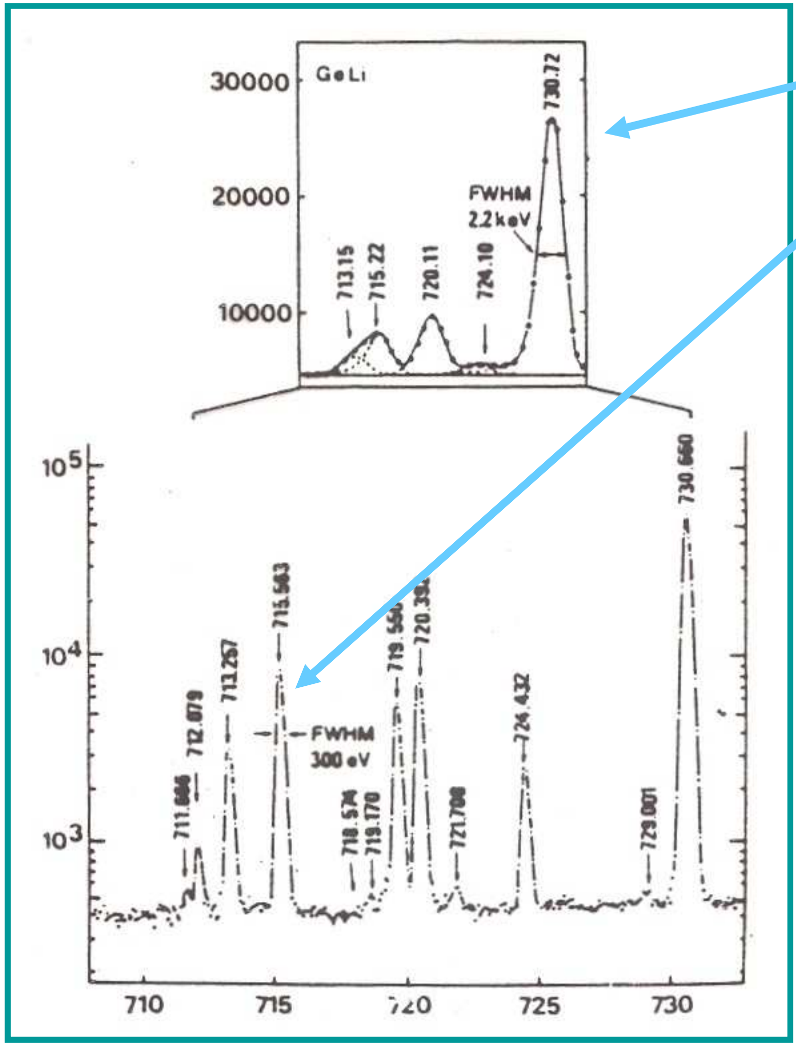




Neutron Flux:	**5 x 10^{14} n cm^{-2} s^{-1}**
Solid Angle:	**Flat crystal: 10^{-11} sr** **Bent crystal: 10^{-7} sr**
Resolution:	**0.008 arc sec**
Angle Precision	**<0.001 arc sec**
Energy Range:	**50 keV – 8 MeV**
Target Material:	**Stable isotopes**

Unique combination

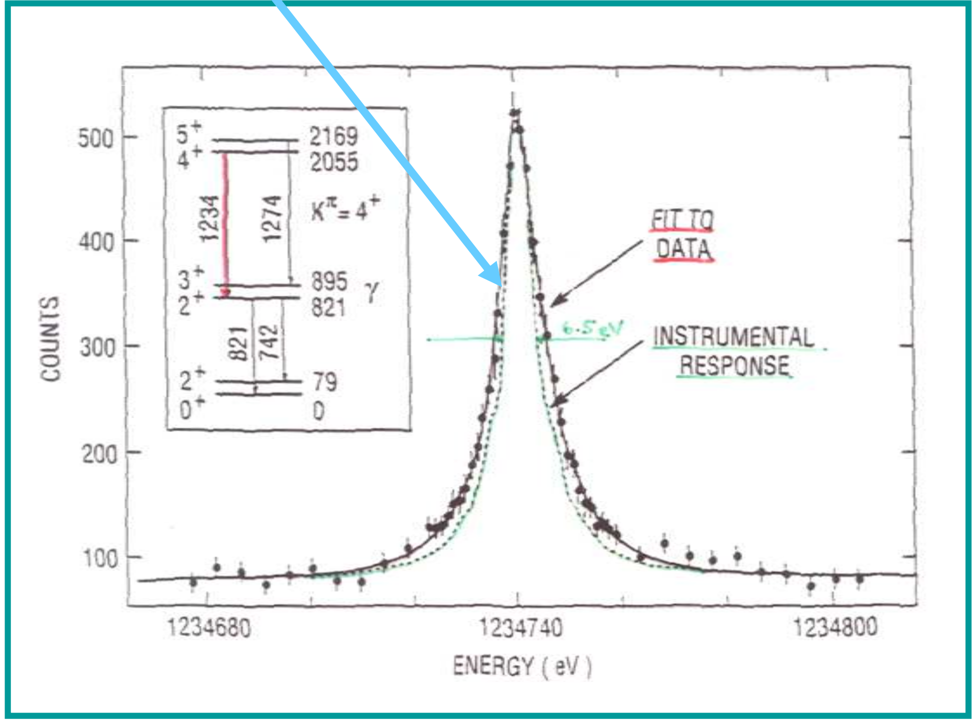
Ultra High Resolution Gamma Spectroscopy



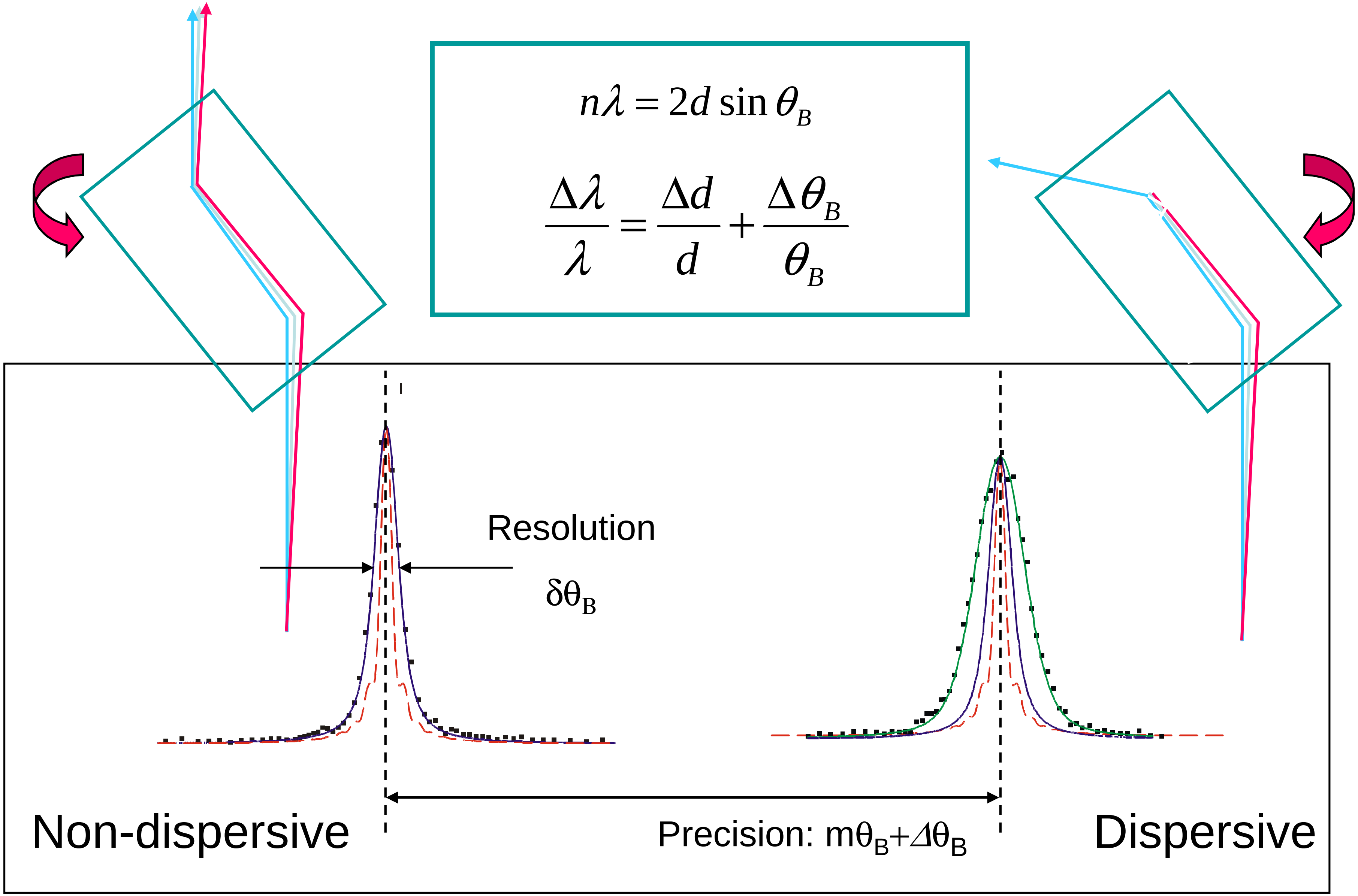
Ge-Detector

DuMond Crystal Spectrometer

Double Flat Crystal Spectrometer



Double Flat Crystal Spectrometers



$$n\lambda = 2d\sin\theta_B$$

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta d}{d} + \frac{\Delta\theta_B}{\theta_B}$$

Resolution

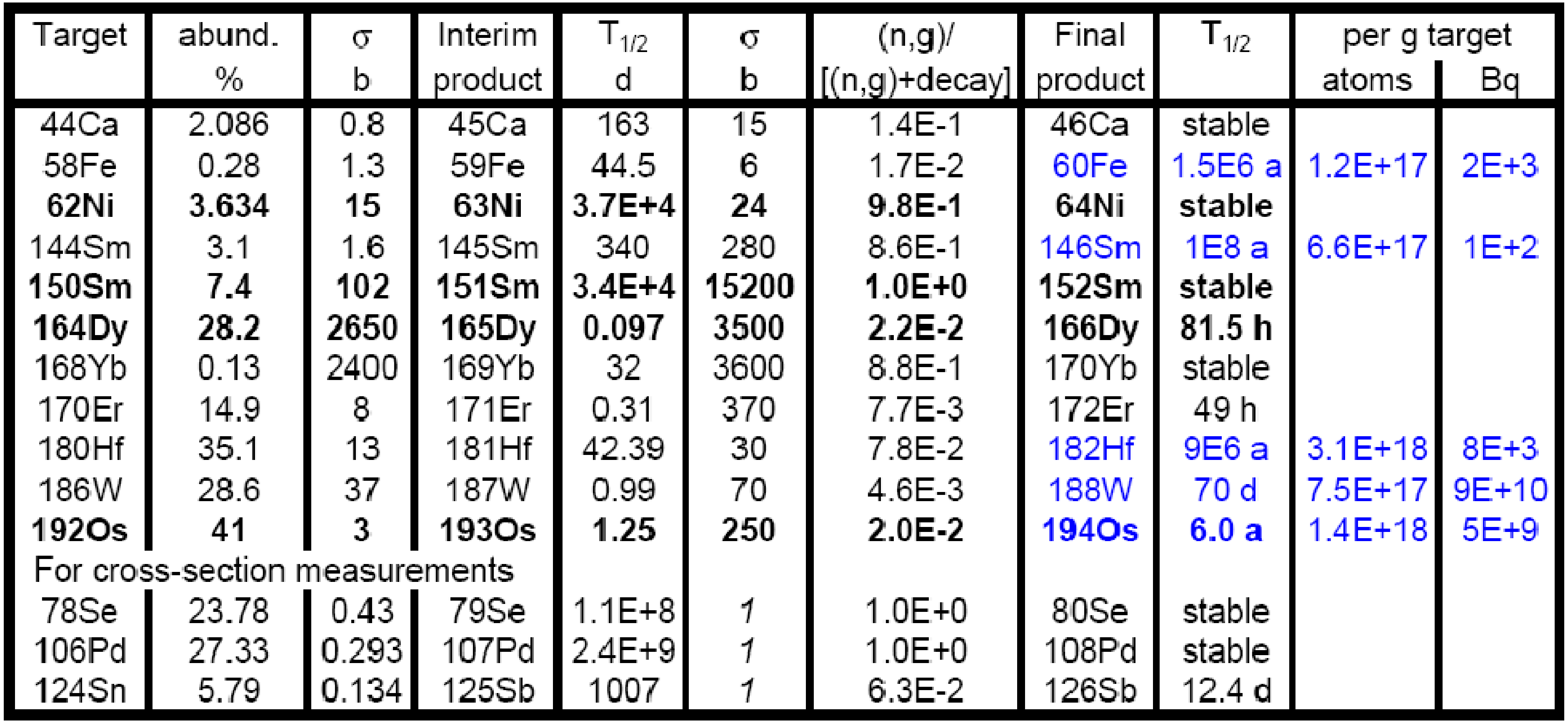
$\delta\theta_B$

Non-dispersive

Precision: $m\theta_B + \Delta\theta_B$

Dispersive

Double neutron capture at GAMS



Target	abund. %	σ b	Interim product	$T_{1/2}$ d	σ b	(n,g)/ [(n,g)+decay]	Final product	$T_{1/2}$	per g target	
									atoms	Bq
44Ca	2.086	0.8	45Ca	163	15	1.4E-1	46Ca	stable		
58Fe	0.28	1.3	59Fe	44.5	6	1.7E-2	60Fe	1.5E6 a	1.2E+17	2E+3
62Ni	**3.634**	**15**	**63Ni**	**3.7E+4**	**24**	**9.8E-1**	**64Ni**	**stable**		
144Sm	3.1	1.6	145Sm	340	280	8.6E-1	146Sm	1E8 a	6.6E+17	1E+2
150Sm	**7.4**	**102**	**151Sm**	**3.4E+4**	**15200**	**1.0E+0**	**152Sm**	**stable**		
164Dy	**28.2**	**2650**	**165Dy**	**0.097**	**3500**	**2.2E-2**	**166Dy**	**81.5 h**		
168Yb	0.13	2400	169Yb	32	3600	8.8E-1	170Yb	stable		
170Er	14.9	8	171Er	0.31	370	7.7E-3	172Er	49 h		
180Hf	35.1	13	181Hf	42.39	30	7.8E-2	182Hf	9E6 a	3.1E+18	8E+3
186W	28.6	37	187W	0.99	70	4.6E-3	188W	70 d	7.5E+17	9E+10
192Os	**41**	**3**	**193Os**	**1.25**	**250**	**2.0E-2**	**194Os**	**6.0 a**	1.4E+18	5E+9
For cross-section measurements										
78Se	23.78	0.43	79Se	1.1E+8	*1*	1.0E+0	80Se	stable		
106Pd	27.33	0.293	107Pd	2.4E+9	*1*	1.0E+0	108Pd	stable		
124Sn	5.79	0.134	125Sb	1007	*1*	6.3E-2	126Sb	12.4 d		

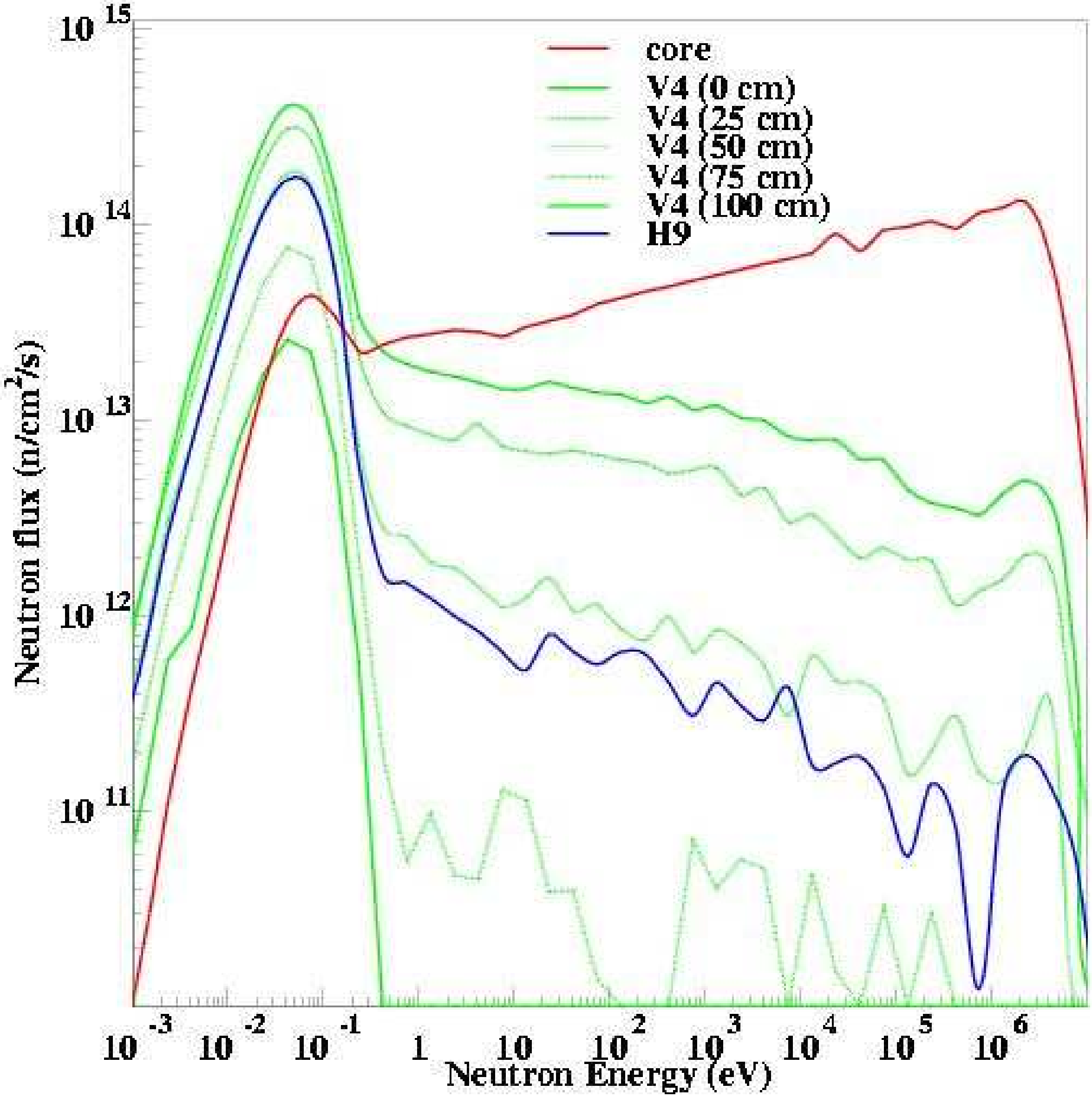
Lines in bold mark experiments that have already been performed

(also (n,g) cross-section of ^{75}Se, ^{147}Nd, ^{152}Eu, ^{170}Tm, ^{171}Tm, ^{194}Ir

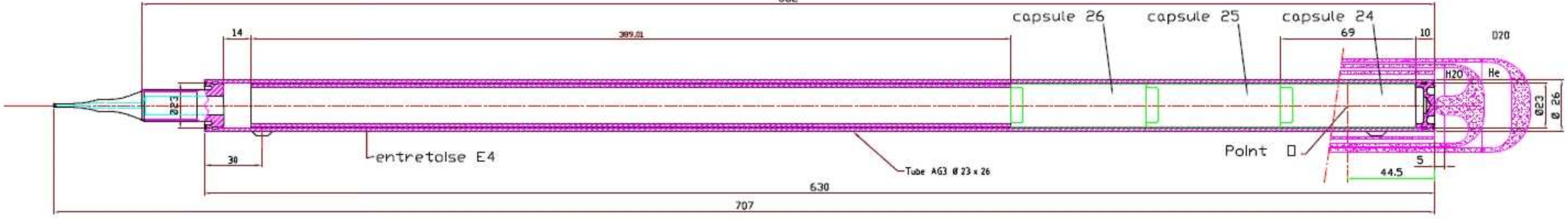
Lines in blue lead to final products that are long-lived.

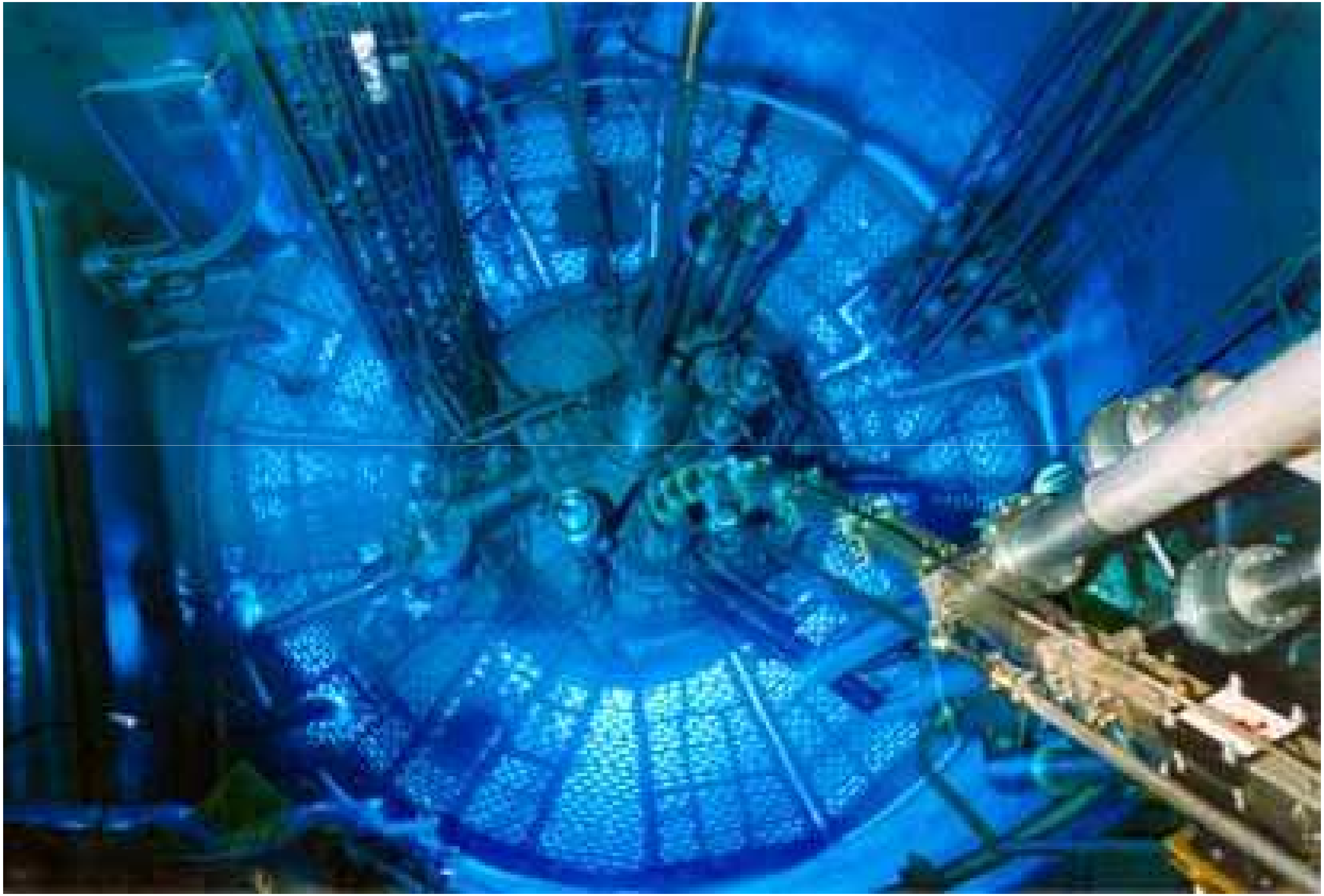
Flux distribution

(Differential flux per energy bin)

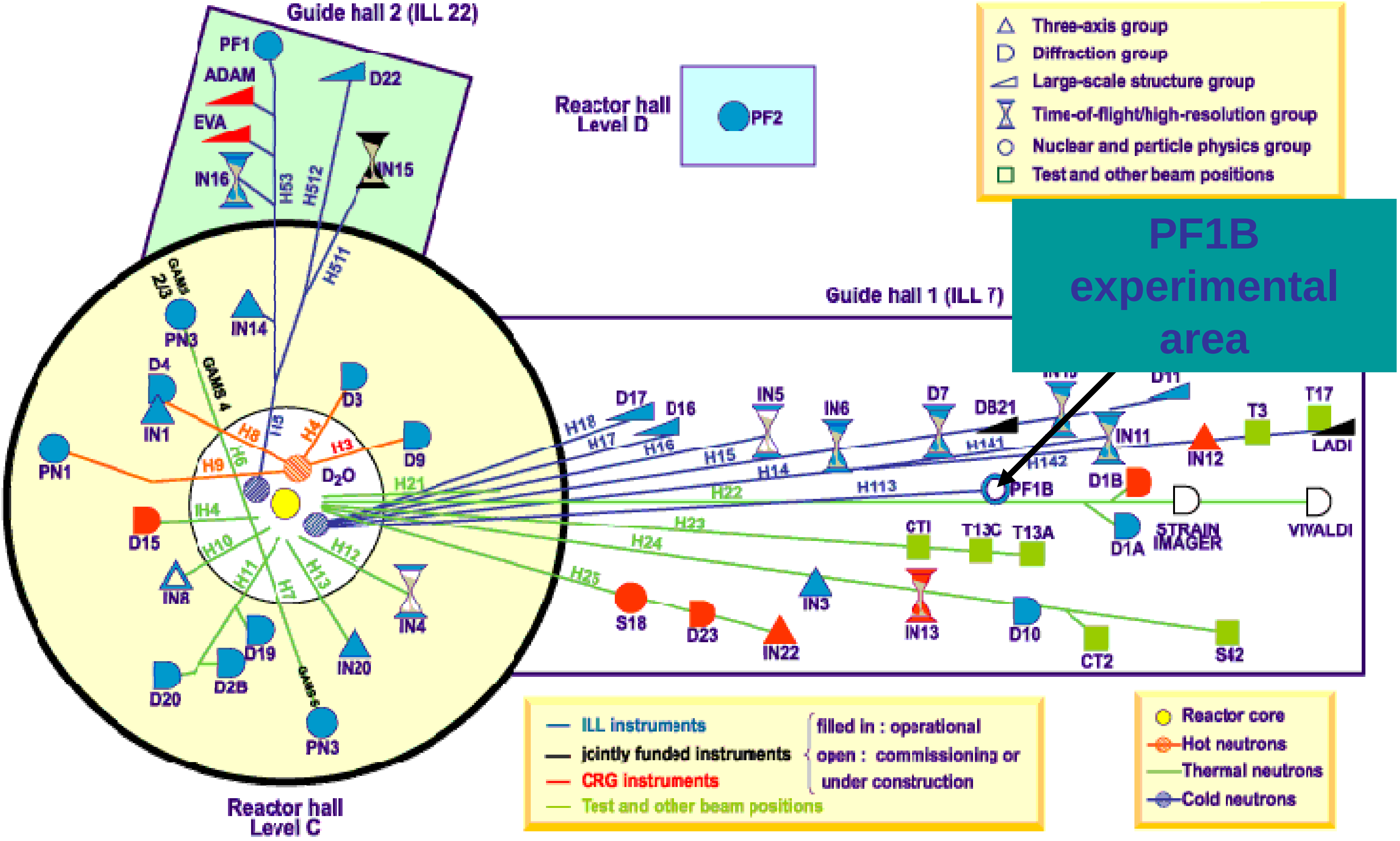


V4 irradiations



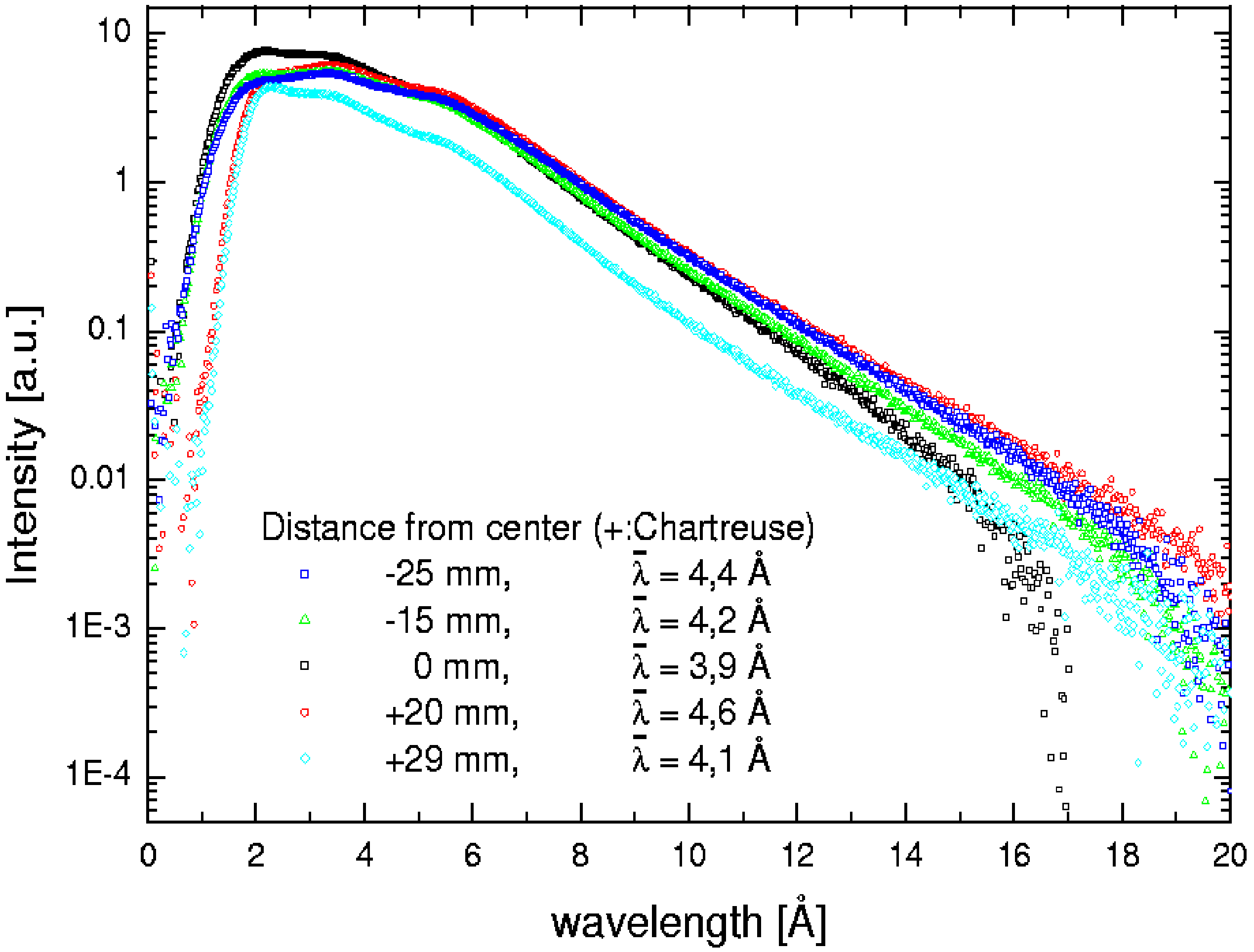


ILL instruments



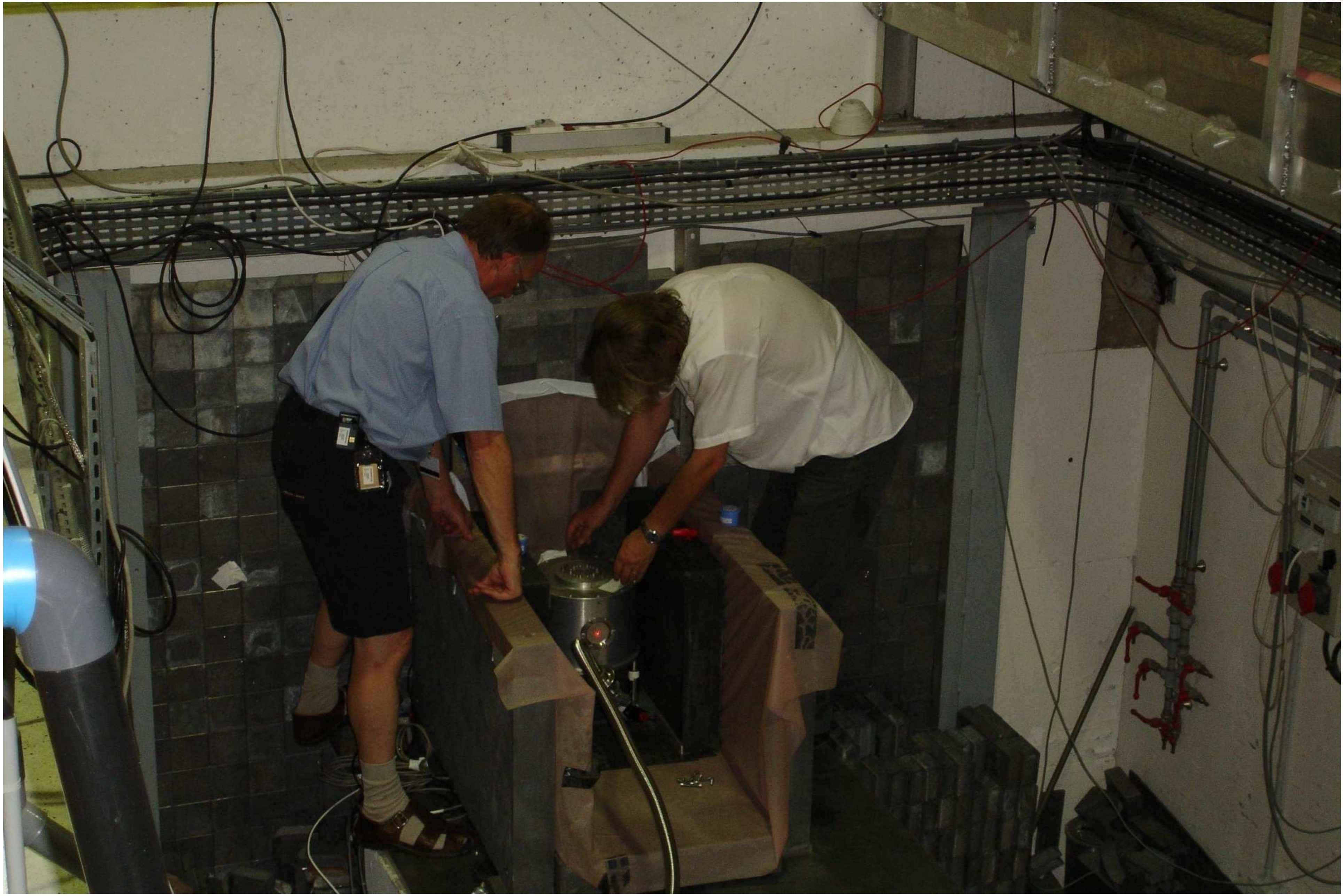
PF1B experimental area

Cold (polarized) neutron beam PF1B

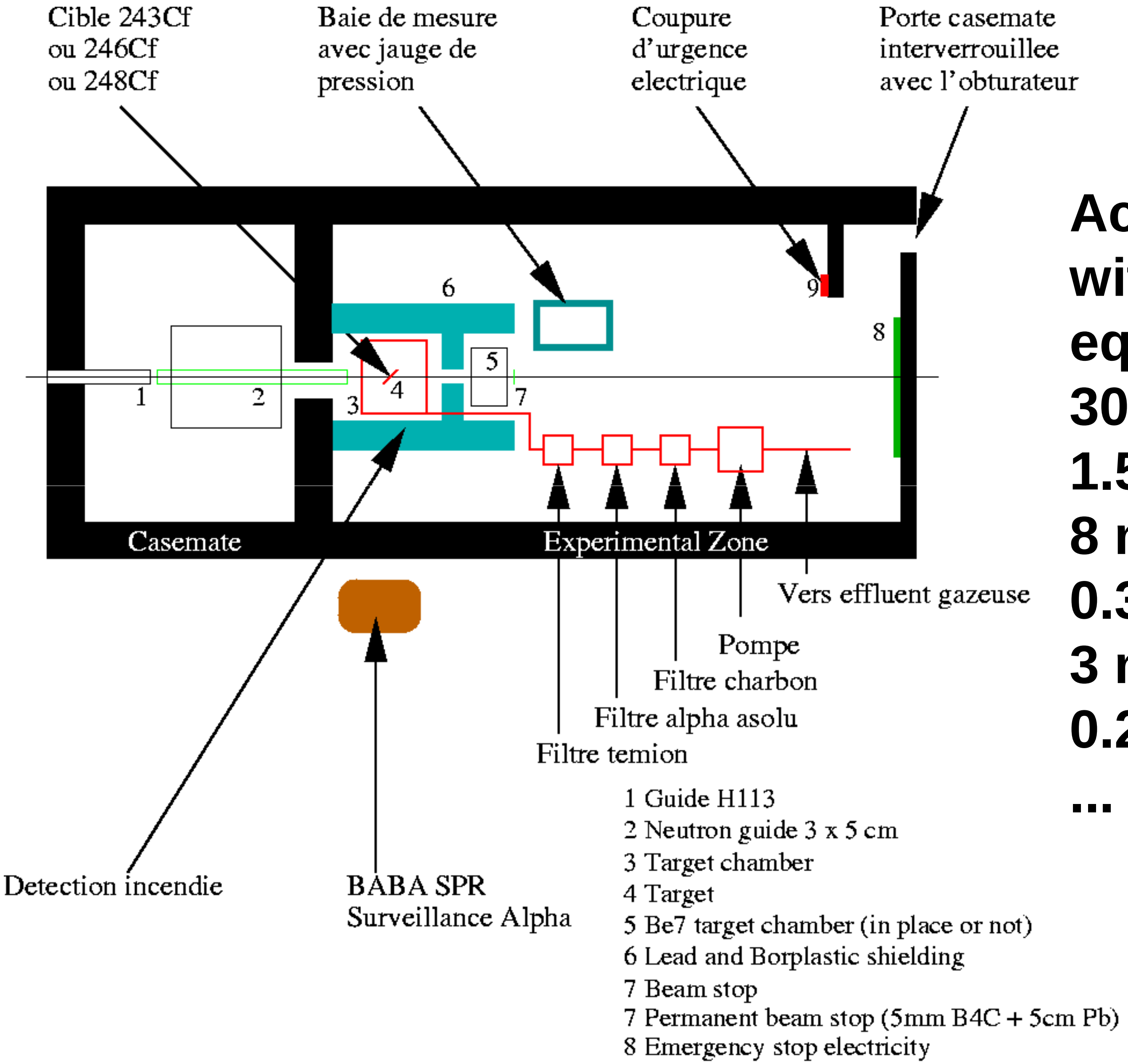


99.7% polarized neutron flux: 3E9 cm^{-2}s^{-1}

- Ballistic supermirror neutron guide H113: 76 m length
- **2E10 n/cm^2/s on 20x6 cm^2**
- **Gamma ray flux from the reactor: negligible**
- Ratio slow neutrons to fast neutrons is ~10^6
- **Average neutron energy:**
  <E>=5.38 meV
  <λ>=3.9 Å
  <T>=62.42 K



Use of actinide targets at PF1B

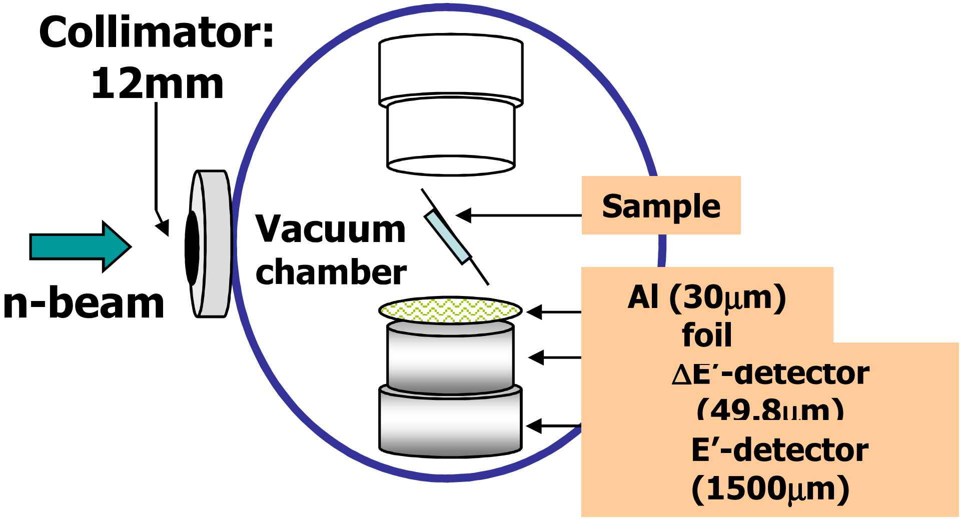


1 Guide H113
2 Neutron guide 3 x 5 cm
3 Target chamber
4 Target
5 Be7 target chamber (in place or not)
6 Lead and Borplastic shielding
7 Beam stop
7 Permanent beam stop (5mm B4C + 5cm Pb)
8 Emergency stop electricity

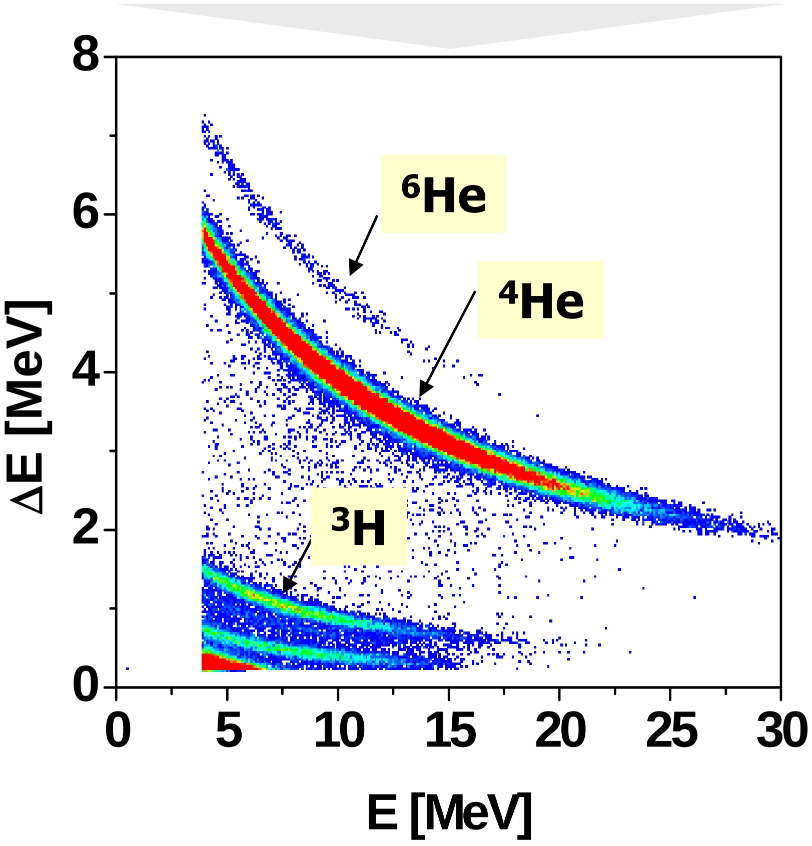
Actinide samples with 20 MBq ^{239}Pu equivalent:

300 mg ^{233}U
1.5 g ^{235}U
8 mg ^{239}Pu
0.3 mg ^{241}Pu
3 mg ^{245}Cm
0.2 mg ^{251}Cf
...

^{249}Cf(n,f) measurement



3rd Step: Measurement of the triton counting Rate



- Sample was turned over an angle of 90° in order to place it in front the other telescope

- Telescope: 49.8 μm ΔE′ and 1500 μm E′ used to measure the ternary triton and alpha yields simultaneously
- Better separation between the ternary particles, but energy threshold higher than the previous telescope

- N_{LRA} = (0.876 ± 0.027) LRA/s
- N_t = (0.069 ± 0.009) t/s
- t/LRA = (7.9 ± 1.2) %
- t/B = (t/B)/(LRA/B)=$(2.20 \pm 0.35) \times 10^{-4}$

O. Serot, S. Vermote, C. Wagemans, et al.

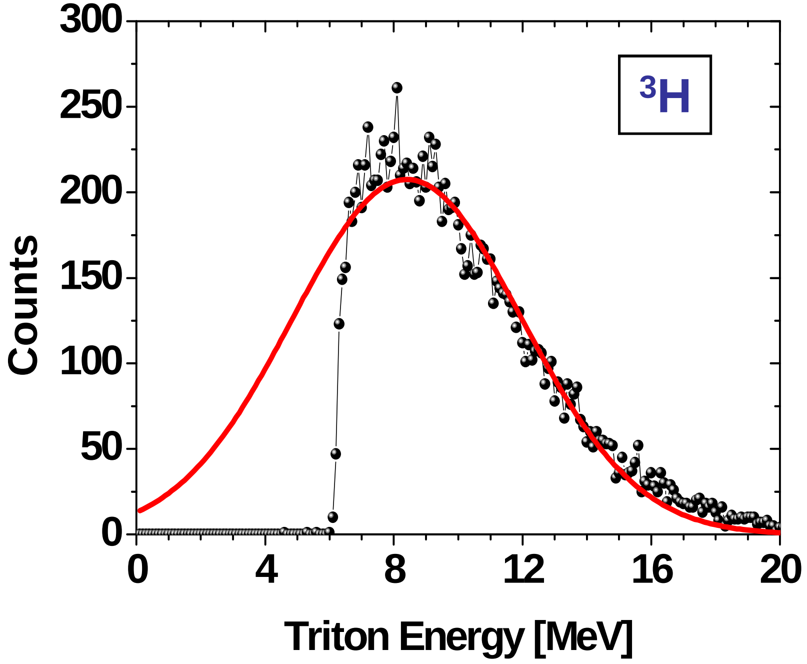
^{249}Cf(n,f) measurement

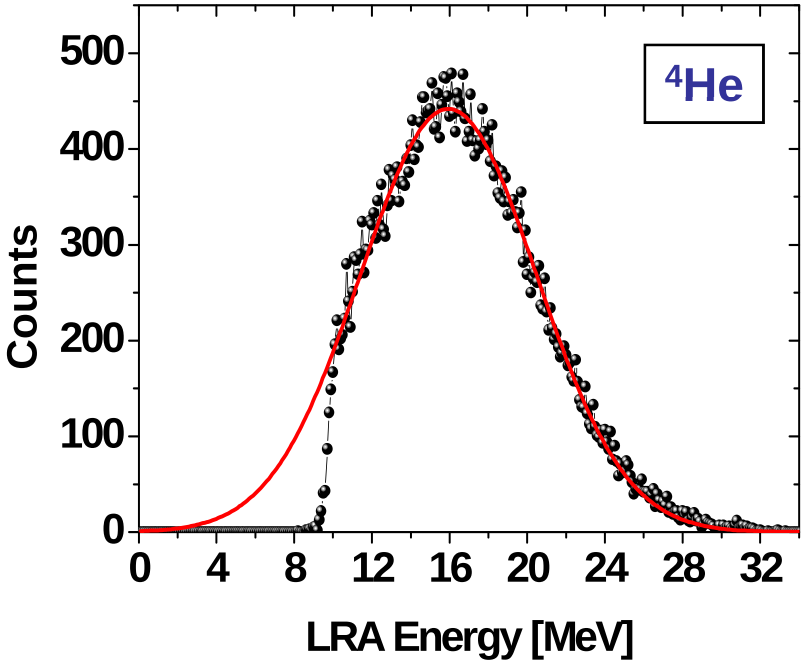
Ternary Triton

- <E>=(8.41 ± 0.17) MeV
- fwhm=(8.41 ± 0.99) MeV
- N_{3H}=(0.069 ± 0.009) 3H/s
- t/B=(2.20 ± 0.35) × 10^{-4}

Ternary Alpha

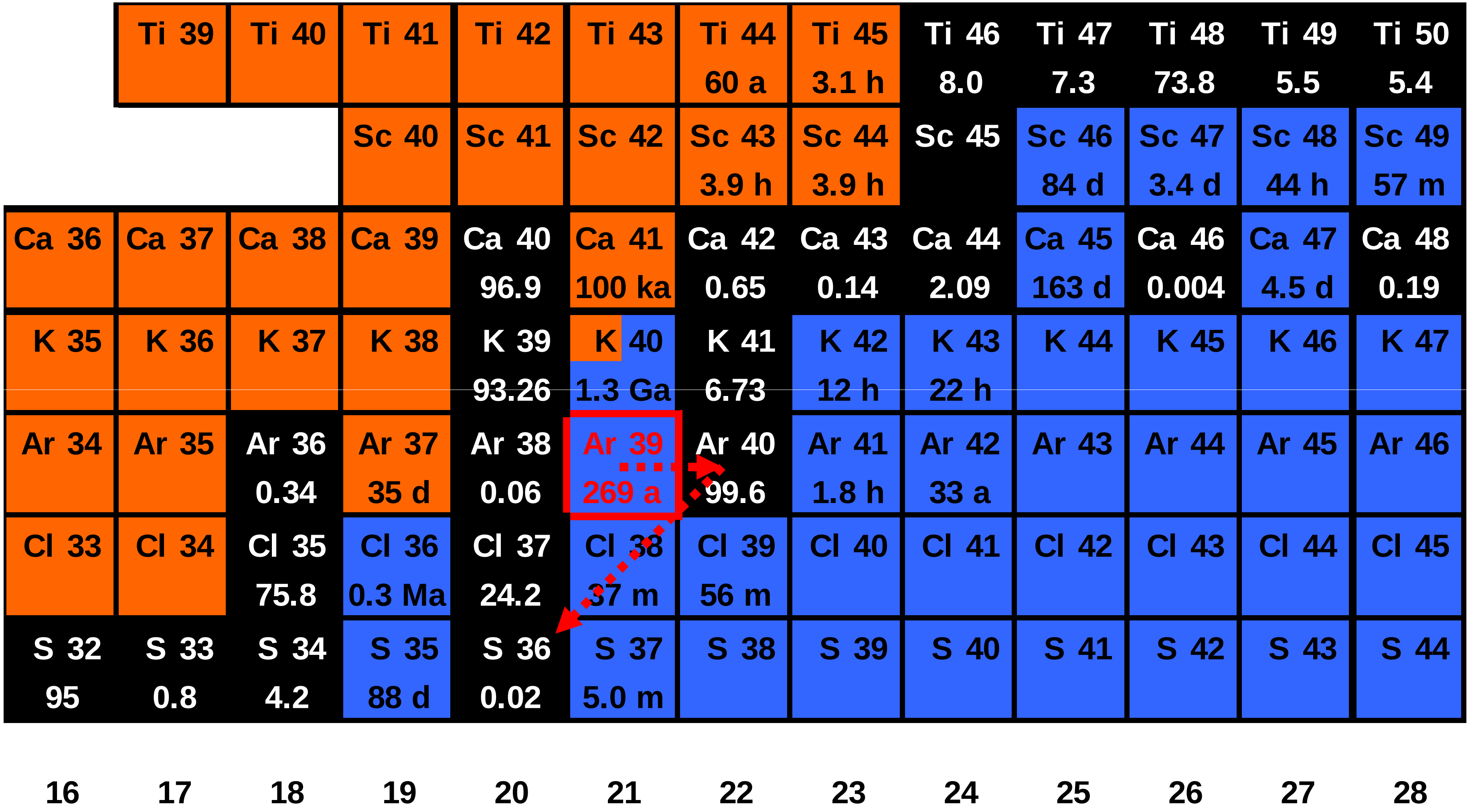
- <E>=(15.94 ± 0.27) MeV
- fwhm=(10.70 ± 0.31) MeV
- N_{lra}=(0.912 ±0.040) LRA/s
- LRA/B=(2.79 ± 0.12) × 10^{-3}





High energy part of ternary particle energy distributions : complementary to LOHENGRIN measurements

$^{39}\text{Ar}(n,\alpha)^{36}\text{S}$



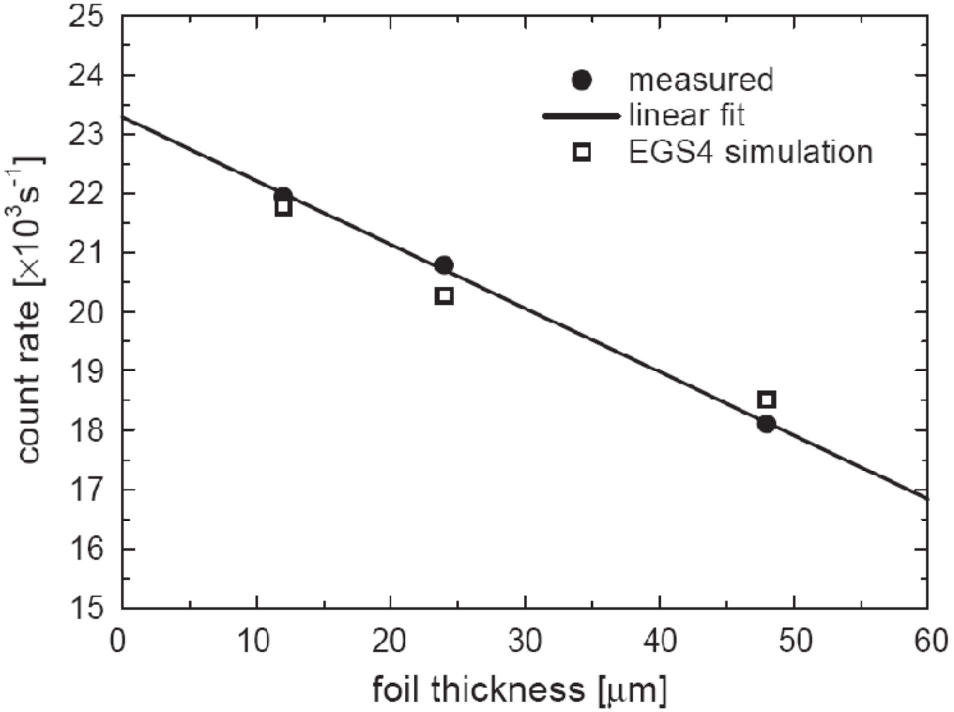
	Ti 39	Ti 40	Ti 41	Ti 42	Ti 43	Ti 44 60 a	Ti 45 3.1 h	Ti 46 8.0	Ti 47 7.3	Ti 48 73.8	Ti 49 5.5	Ti 50 5.4
			Sc 40	Sc 41	Sc 42	Sc 43 3.9 h	Sc 44 3.9 h	Sc 45	Sc 46 84 d	Sc 47 3.4 d	Sc 48 44 h	Sc 49 57 m
Ca 36	Ca 37	Ca 38	Ca 39	Ca 40 96.9	Ca 41 100 ka	Ca 42 0.65	Ca 43 0.14	Ca 44 2.09	Ca 45 163 d	Ca 46 0.004	Ca 47 4.5 d	Ca 48 0.19
K 35	K 36	K 37	K 38	K 39 93.26	K 40 1.3 Ga	K 41 6.73	K 42 12 h	K 43 22 h	K 44	K 45	K 46	K 47
Ar 34	Ar 35	Ar 36 0.34	Ar 37 35 d	Ar 38 0.06	Ar 39 269 a	Ar 40 99.6	Ar 41 1.8 h	Ar 42 33 a	Ar 43	Ar 44	Ar 45	Ar 46
Cl 33	Cl 34	Cl 35 75.8	Cl 36 0.3 Ma	Cl 37 24.2	Cl 38 37 m	Cl 39 56 m	Cl 40	Cl 41	Cl 42	Cl 43	Cl 44	Cl 45
S 32 95	S 33 0.8	S 34 4.2	S 35 88 d	S 36 0.02	S 37 5.0 m	S 38	S 39	S 40	S 41	S 42	S 43	S 44
16	17	18	19	20	21	22	23	24	25	26	27	28

IS382 experiment: $^{39}Ar(n,\alpha)^{36}S$

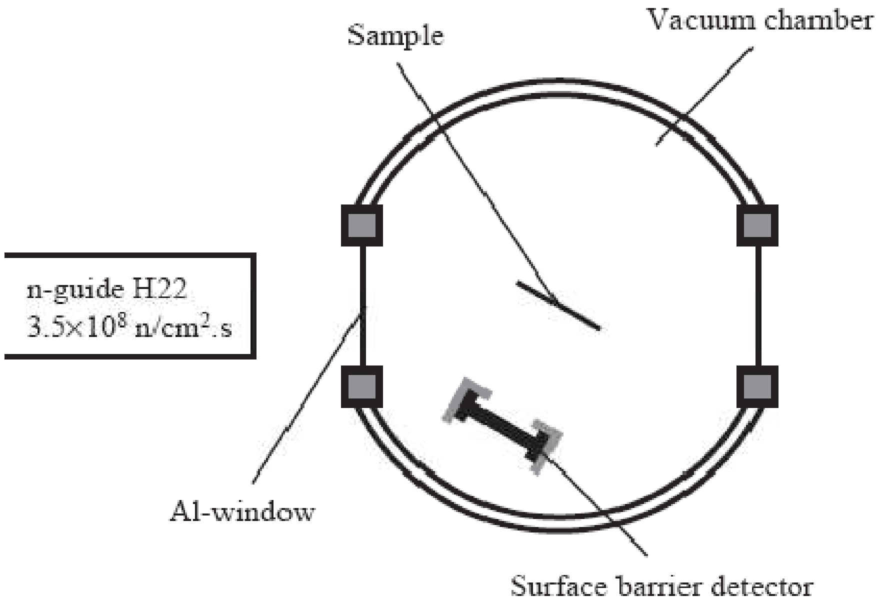
1. **first sample: 24 hours collection with CaO target at ISOLDE for a sample with 8E12 atoms of ^{39}Ar**

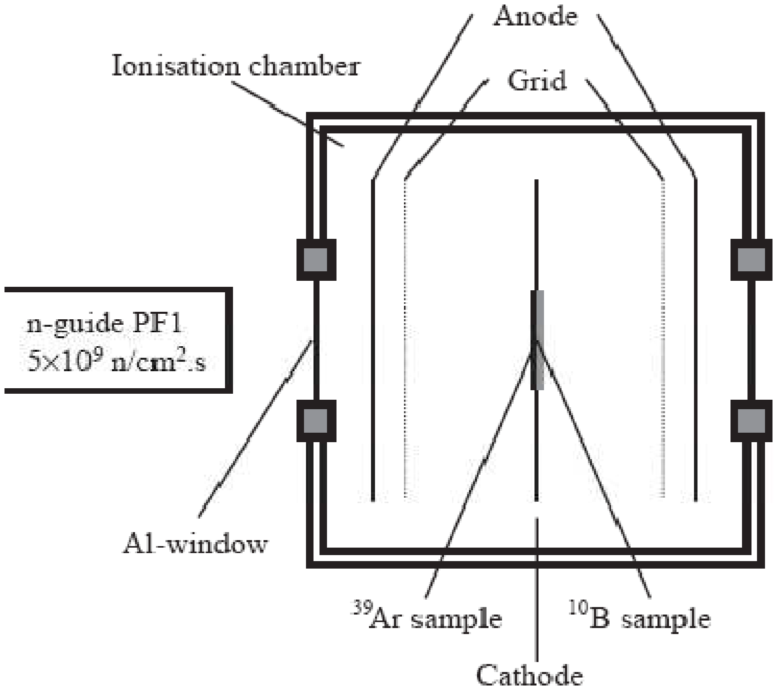
   ⇒ too weak to see (n,α)

2. **second sample: 3 days collection with TiO_2 target at ISOLDE (up to 4.1 μA of 1 GeV protons) for a sample with 2.85E14 atoms of ^{39}Ar**



Search for ^{39}Ar(n,α)^{36}S at ILL





$\sigma[^{39}\mathrm{Ar}(n_{th},\alpha)^{36}\mathrm{S}] < 0.29$ b

G. Goeminne et al., Nucl. Phys. A688 (2001) 233c.

G. Goeminne et al., Nucl. Instr. Meth. A489 (2002) 577.

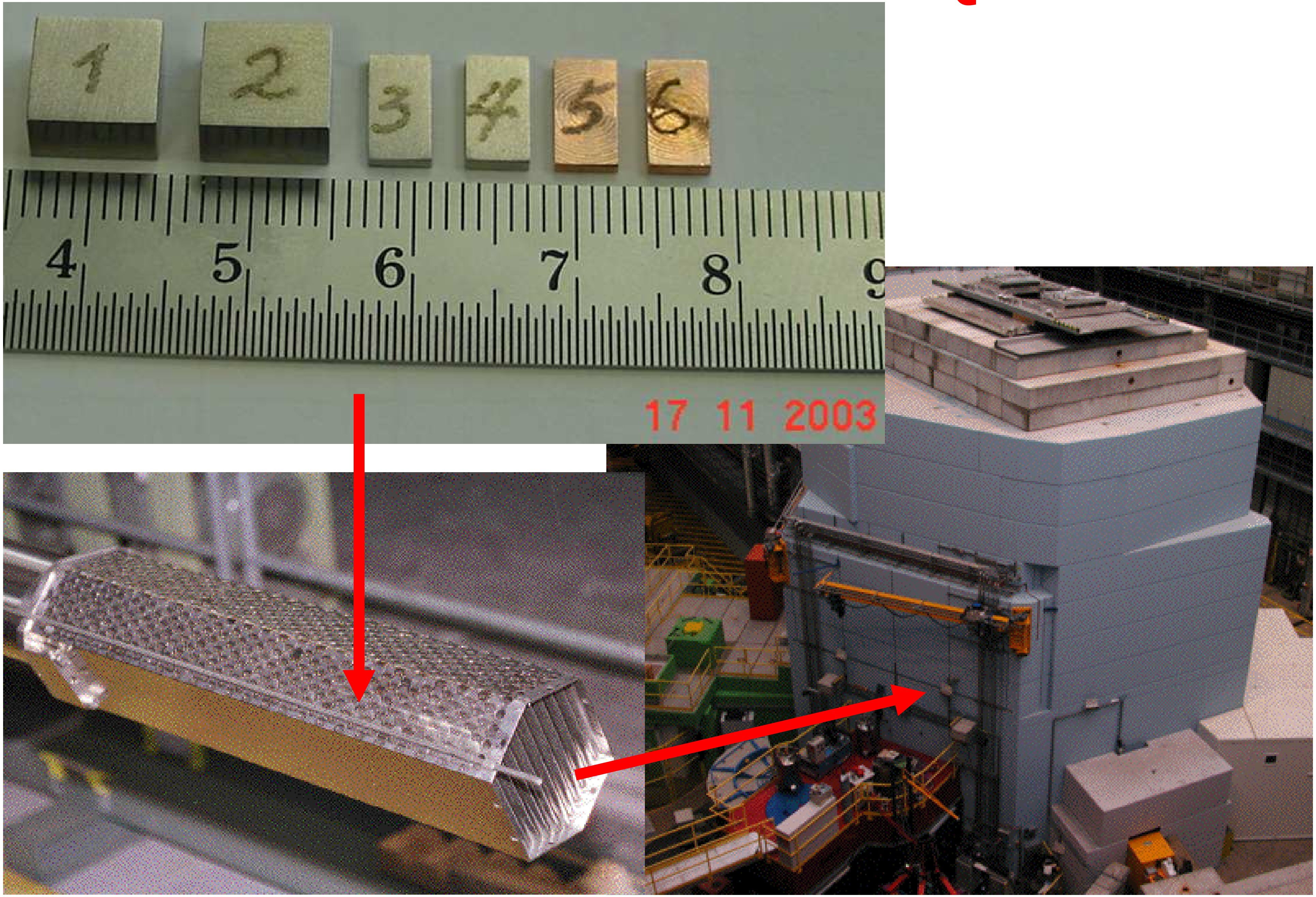
⇒ **more intense sample needed to determine real value and to measure at astrophysical energies with neutron time of flight facility**

Possible improvements

Modification	Gain
• replace MK7 FEBIAD (4% ionization eff.) with Mono-ECRIS (40% ionization eff.)	10
• Ti disk target (4 g/cm^3) instead of TiO_2 fiber target (0.4 g/cm^3)	10
• 0.7 cm long target instead of 20 cm long	0.035
• Proton current 40 μA instead of 2.2 μA	18
• Beam time 420 days	140
Total	8000

⇒ Sample with 2E18 atoms

Ti irradiation at SINQ

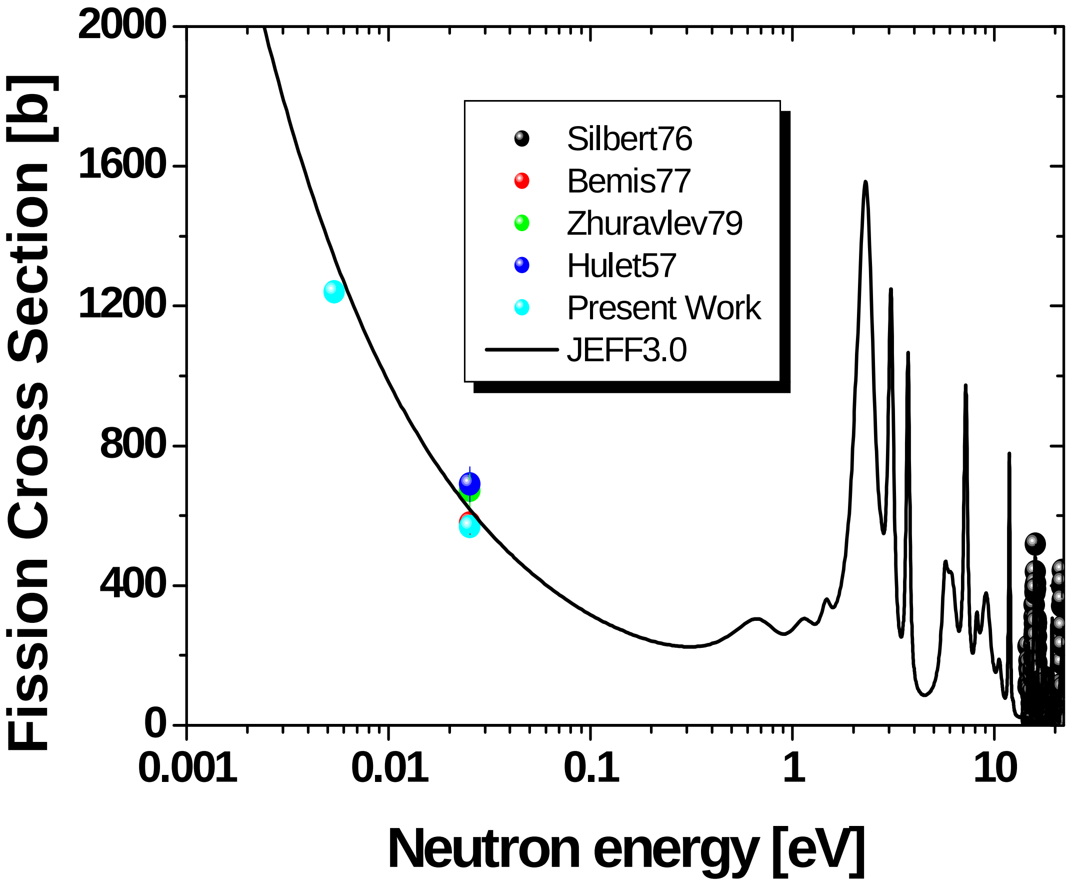


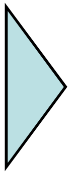
Ti irradiation at SINQ, ion implantation at ISOLDE

- irradiation of 2.56 g Ti in STIP-IV, target 6:
  April 2004 to December 2005
- integrated dose about 0.4 Ah/cm^2 (>8E21 p/cm^2)
- decay of short-lived activities for 4 years
- dose rate initially dominated by ^{46}Sc (84 days)
- implantation during ISOLDE shutdown once the MonoECRIS is working reliably
- 3-4E18 atoms of ^{39}Ar (0.3 GBq)
- simultaneous extraction of ≈1E17 atoms of ^{42}Ar (70 MBq)

^{243}Cm(n,f) measurement

Hulet (1957)	690 ± 50
Bemis (1977)	609.6 ± 26.9 (gf=1) 579 ± 31 (gf=0.95)
Zhuravlev (1979)	672 ± 60
Mughabghab	617 ± 20
Present work	569 ± 25

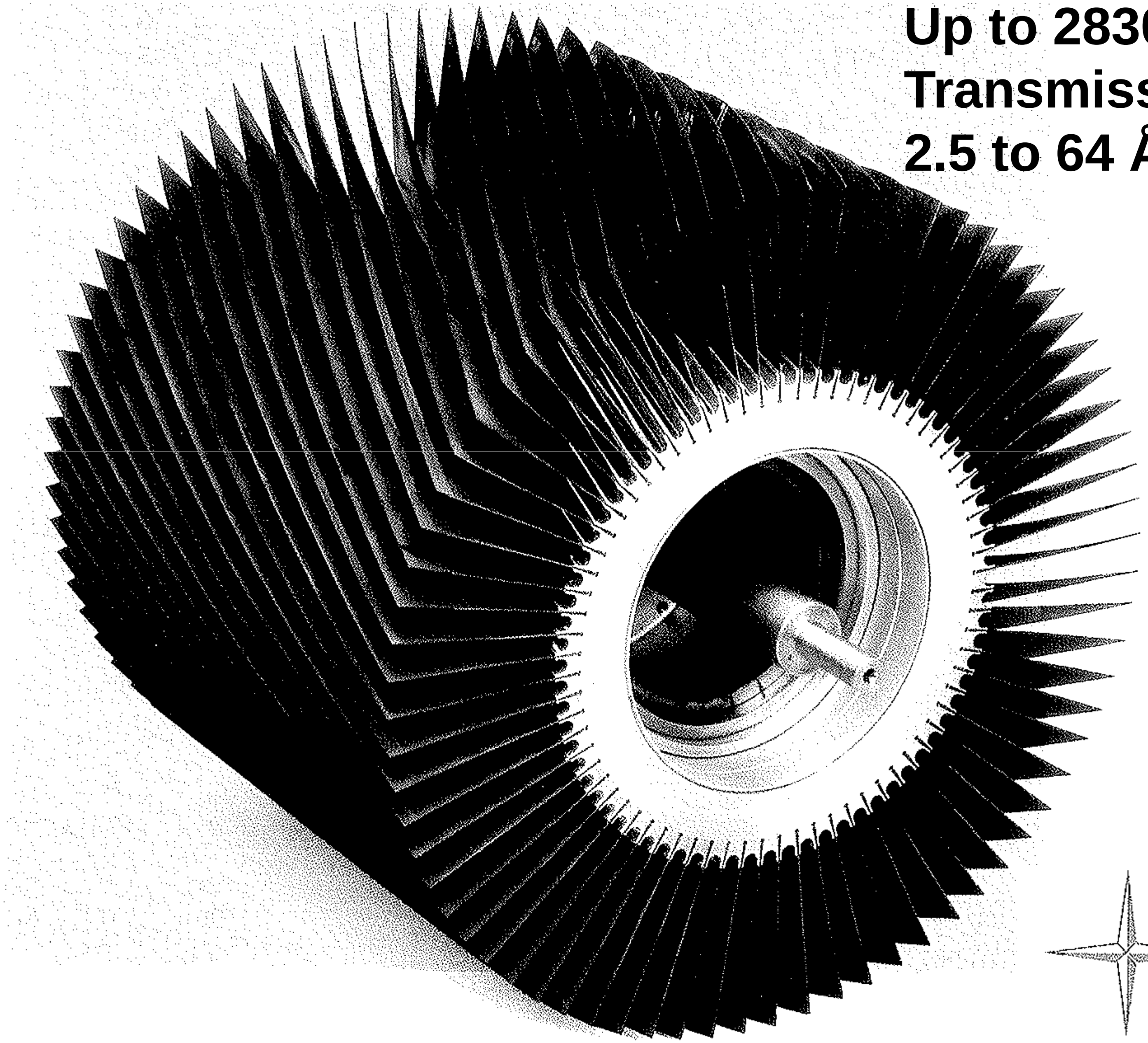




In agreement with the measurement performed by Bemis in 1977

O. Serot, S. Vermote, C. Wagemans, et al.

Velocity selector



Up to 28300 rpm

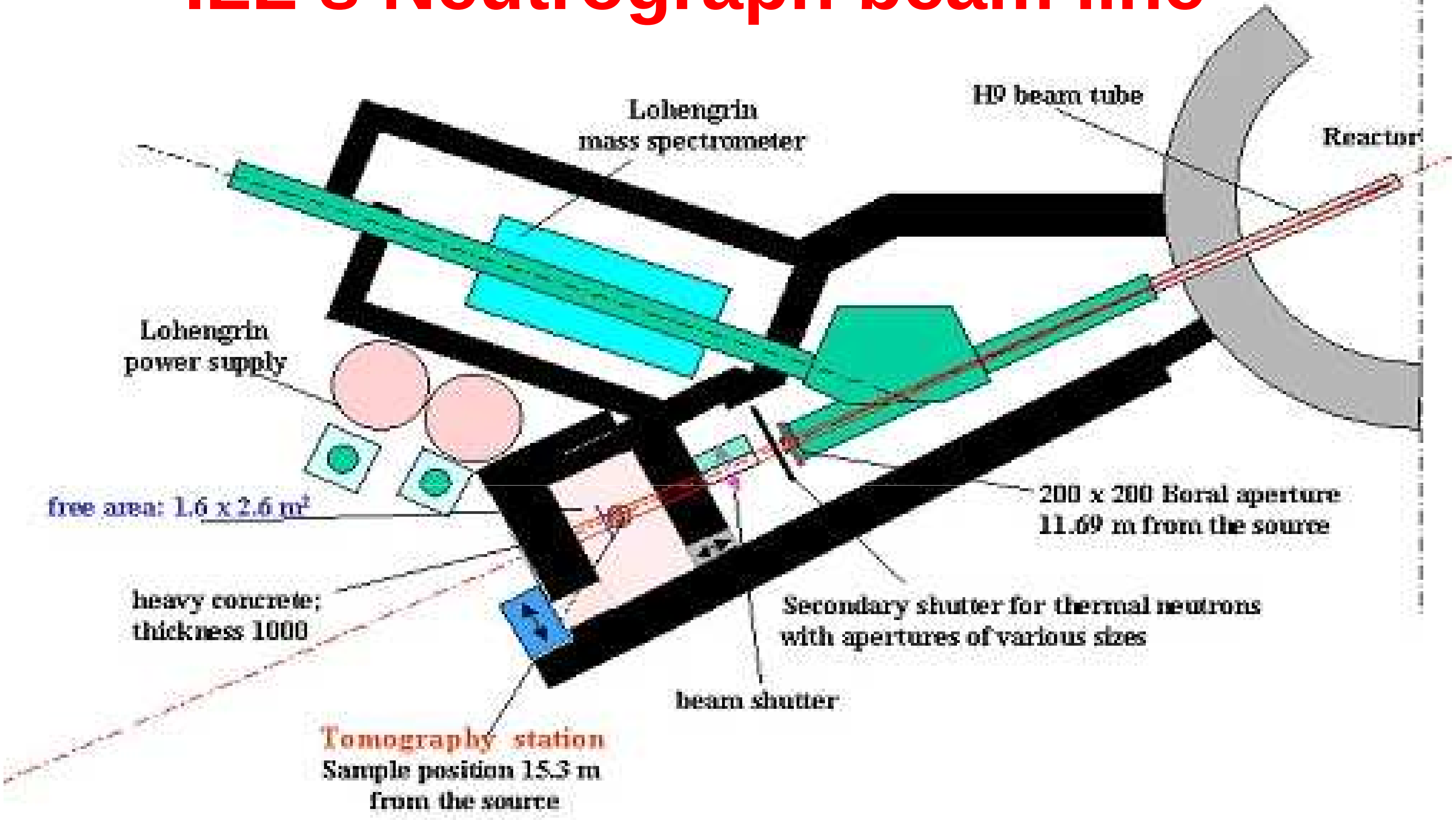
Transmission 77 to 94%

2.5 to 64 Å

Dornier

Deutsche Aerospace

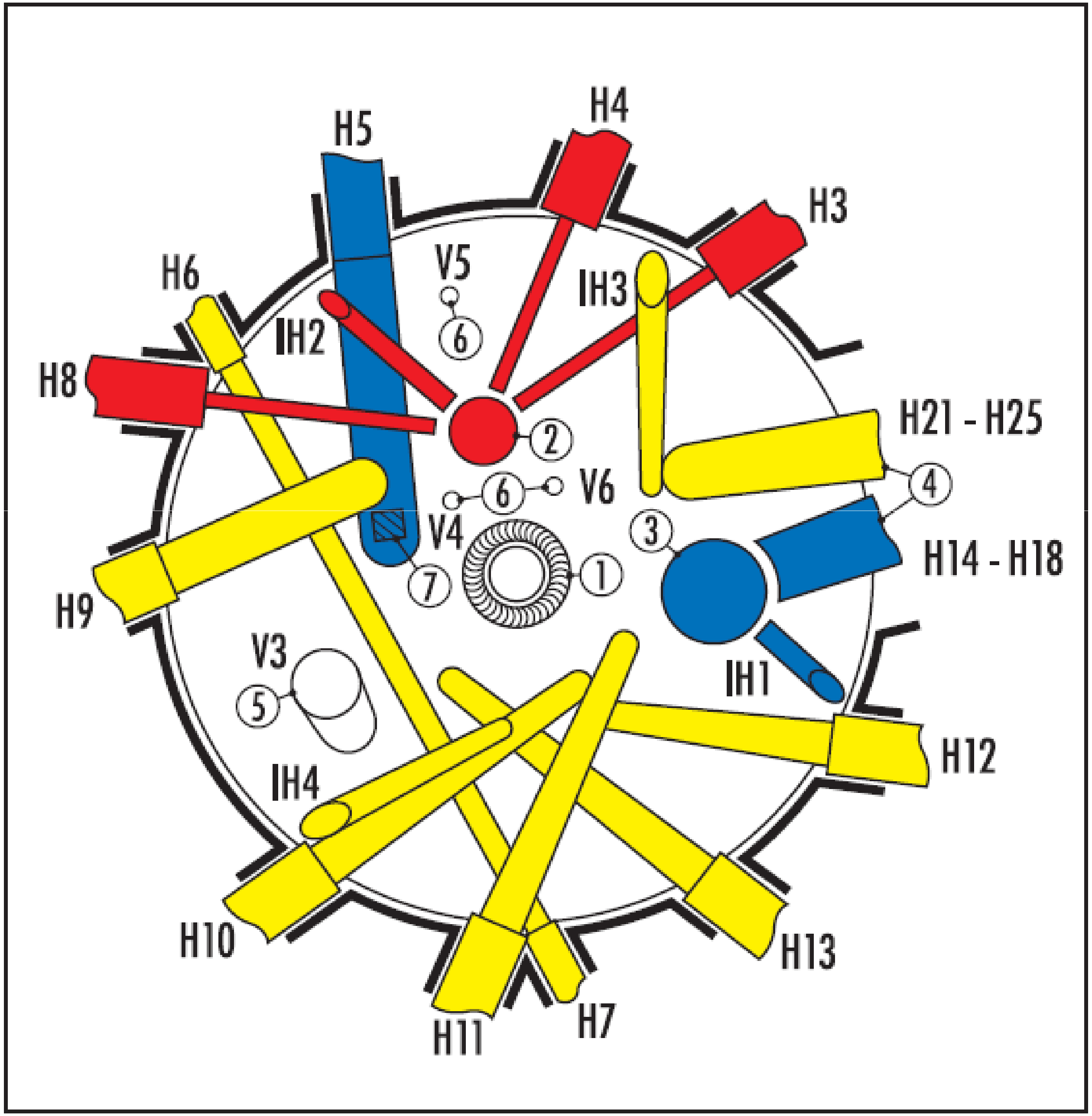
ILL's Neutrograph beam line



Thermal neutron flux: 3E9 cm^{-2}s^{-1}

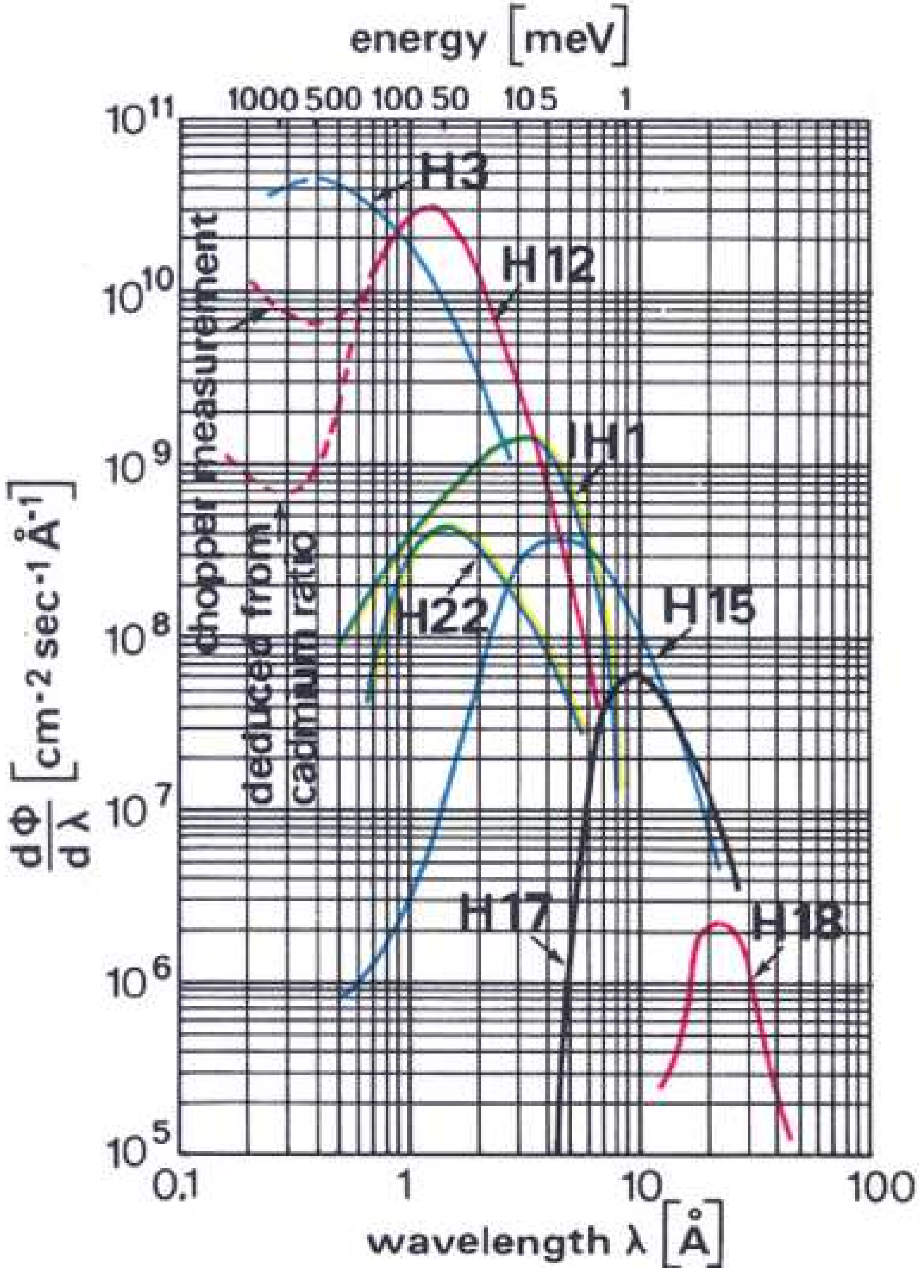
Direct view to core: fast neutrons, high gamma background

Other beamlines



Hot neutrons

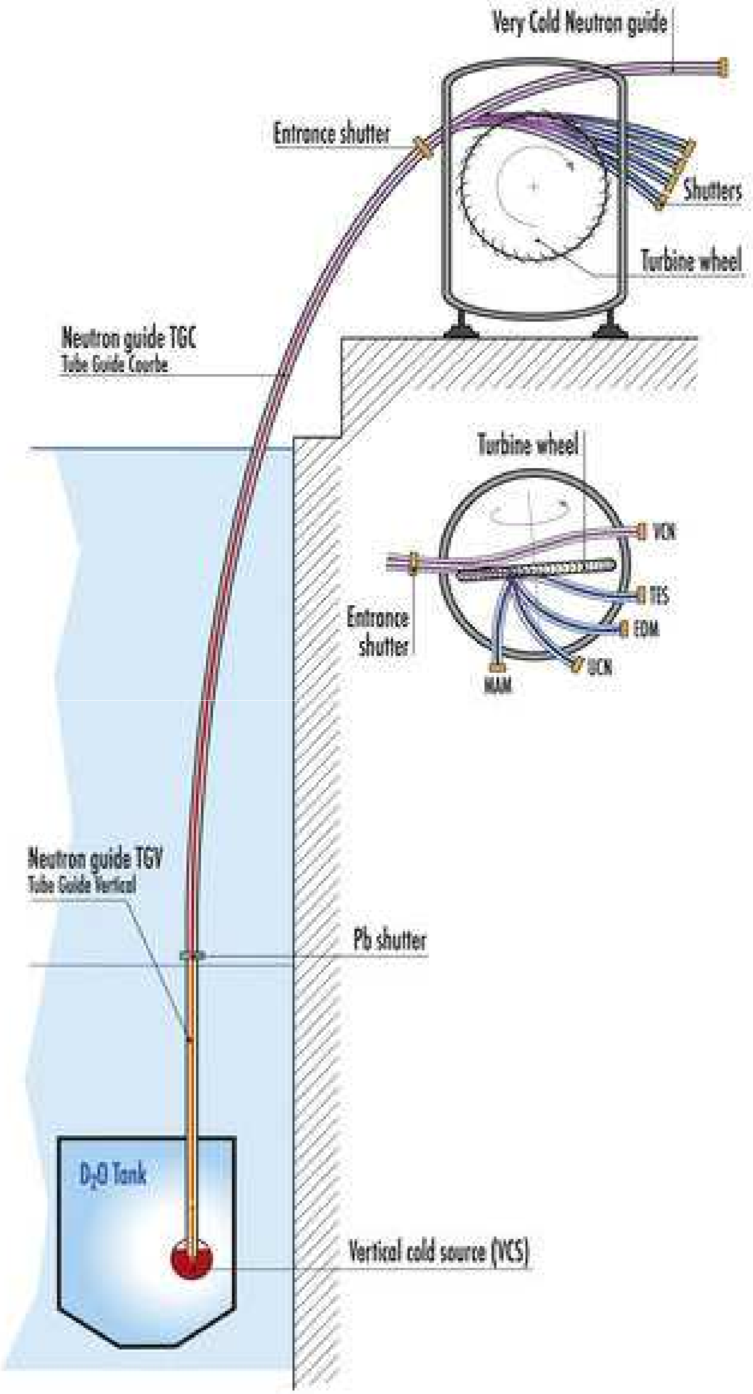
- **hot neutrons:**
  4E7 n/cm^2/s at 0.1 eV
  1E6 n/cm^2/s at 1 eV



PF2

- **ultracold neutrons:**
  3E4 n/cm^2/s for E=0 to 250 neV,
  beam size up to 14x10 cm^2

- **very cold neutrons:**
  4E6 n/cm^2/s at 8 μeV
  beam size up to 7x3.4 cm^2

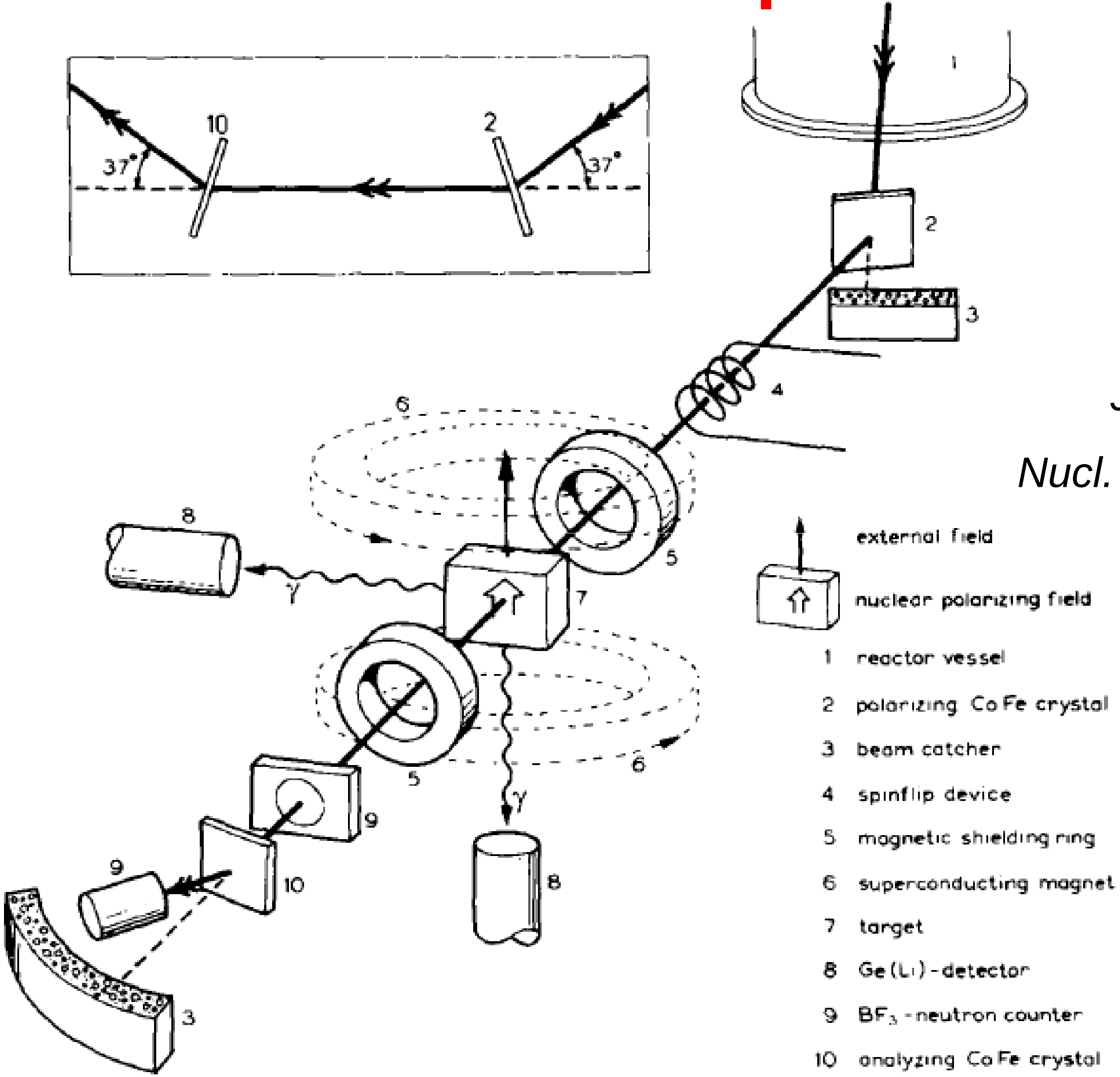


Sample environment

Available for experiments at ILL:

- **dilution refrigerators down to 15 mK**
- **superconducting split coil magnets up to 15 T**

Polarized neutron capture on oriented nuclei

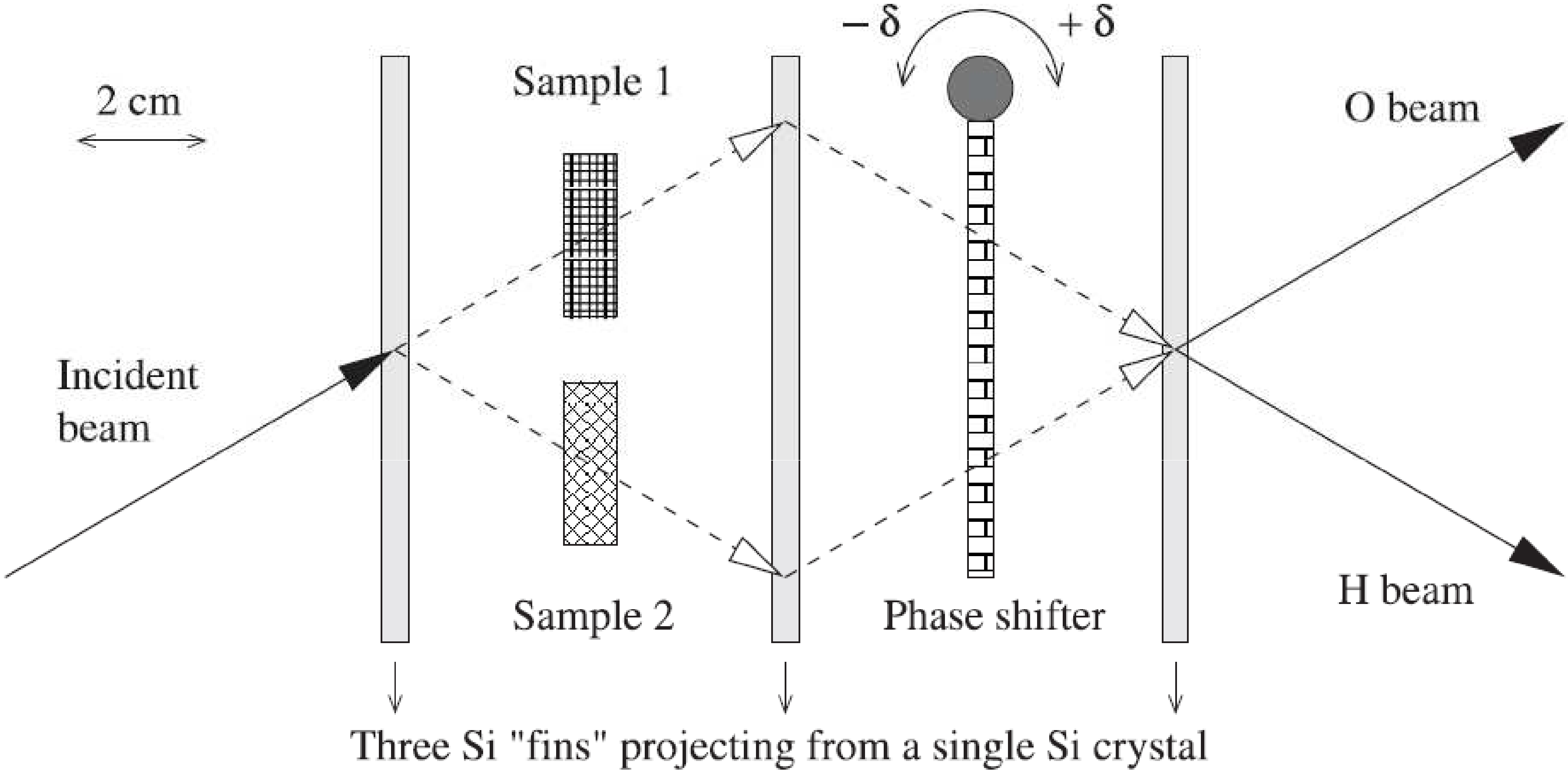


J.J. Bosman and H. Postma,

Nucl. Instr. Meth. 148 (1978) 331.

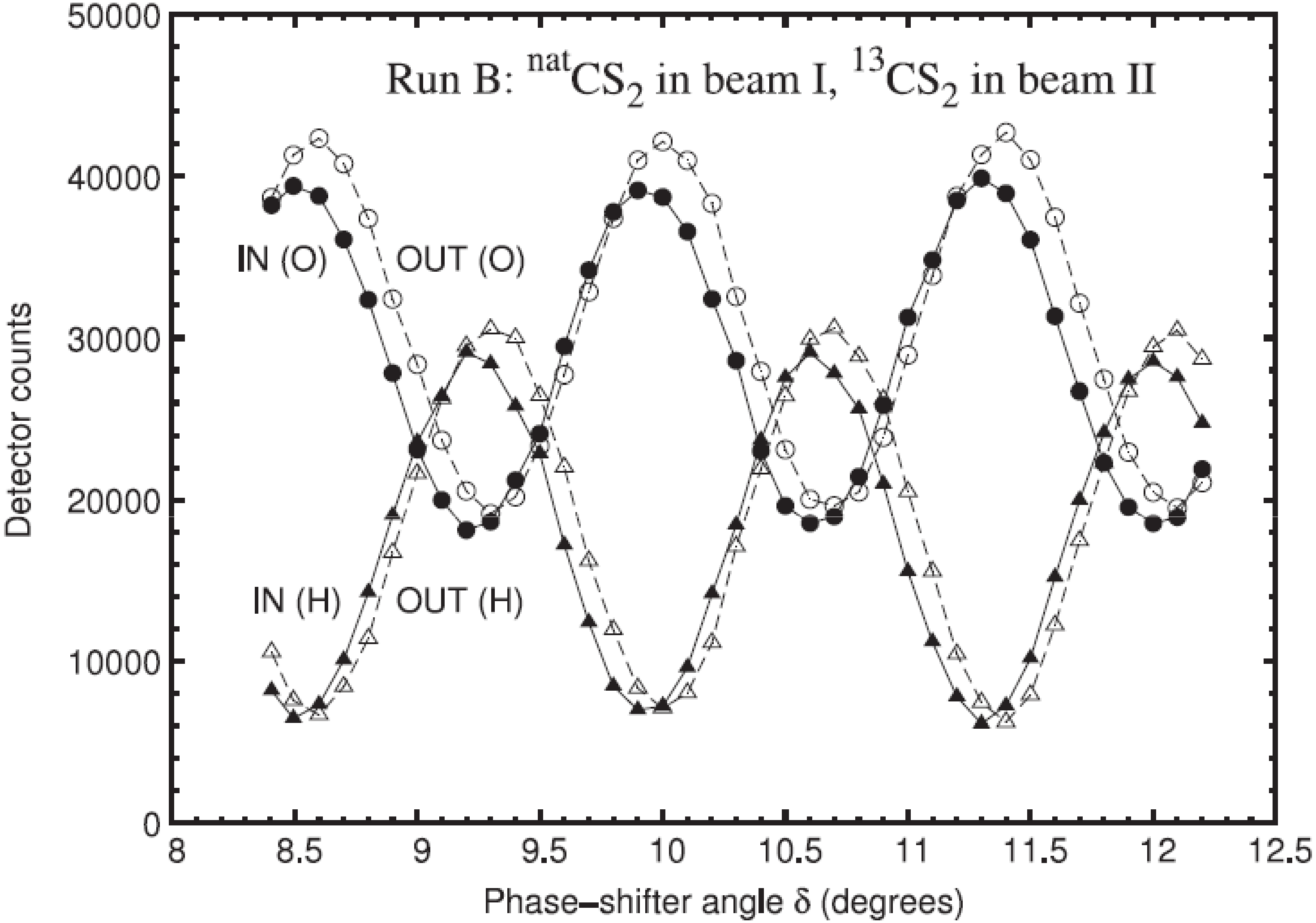
Fig. 3. Schematic view of the polarized beam system, target with polarizing split-coil magnet, neutron spin-flip device, neutron polarization analyzer and Ge(Li) detectors (not to scale).

Neutron interferometer S18



$$\Delta\phi = n\lambda D\Delta\overline{b},$$

Coherent scattering length measurement



$$b_{\mathrm{coh},13\mathrm{C}} = 6.542(3)$$

H.E. Fischer et al., J. Phys. Cond. Matter 20 (2008) 045221.

How to get beam time at ILL?

- experiments at LOHENGRIN, GAMS, PF1B or S18 via proposals to ILL, discuss with instrument responsibles
- proposal deadlines 15 February and 15 September
- (co-)proposers affiliated to member state lab

- study of short-lived products at MINI-INCA in collaboration with CEA Saclay and proposal to ILL

- experiments at Neutrograph (thermal neutron beam of 3E9 n/cm^2/s) or irradiations in V4 (up to 1.5E15 n/cm^2/s) to be discussed

- Possible “abuse” of diffraction instrument to access monochromatic “hot” neutron beams up to 1.3 eV (few 10^6 to 10^7 n/cm^2/s)