Search for double beta decays of palladium isotopes into excited states

INTERNATIONAL SCHOOL OF NUCLEAR PHYSICS
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Outline

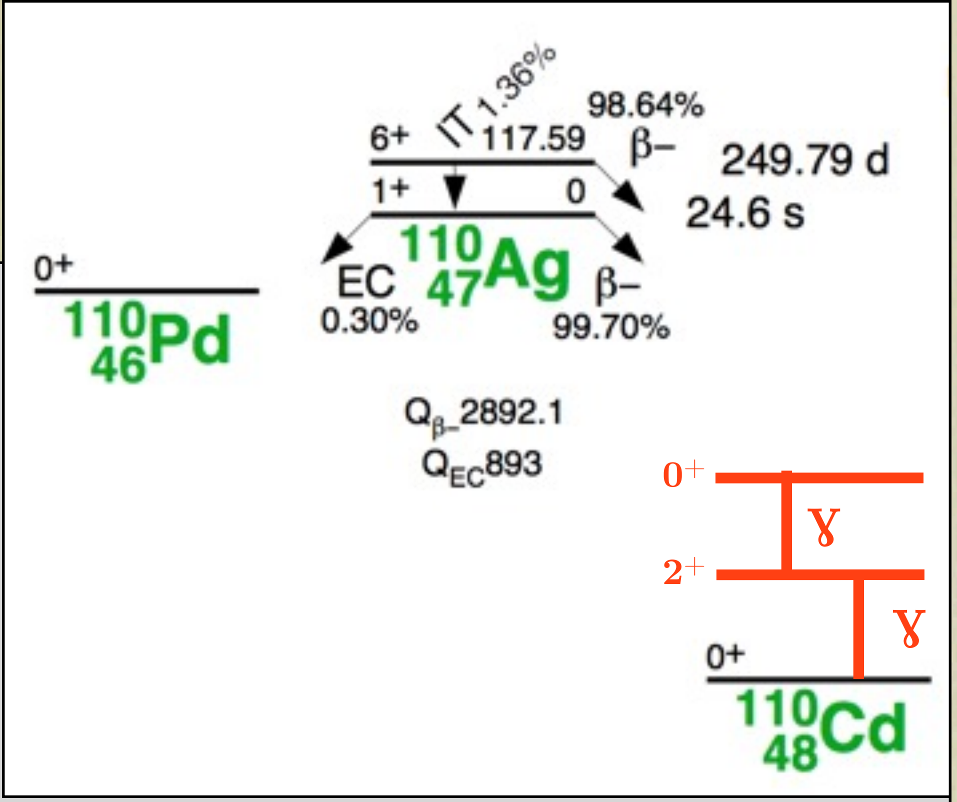
- Why double beta decays into excited states
- Palladium 101
- Three experiments...
- Outlook

Experiments:

1. Felsenkeller **Physics Letters B 705 47–51 (2011)**
2. HADES **Physical Review C 87, 034312 (2013)**
3. LNGS

Why DBD into excited states?

- Adds nuclear structure information
- Eventually helps constraining NME calculations for $0\nu\beta\beta$
- Potential resonance enhancement for $0\nu ECEC$
- Convenient experimental signature (ɣ-lines)
- So far only discovered in ^{100}Mo (1995) and ^{150}Nd (2004)



Compilation of 2nubb 0_1^+ transitions

Nuclei	$E_{2\beta}$, keV	Experiment $T_{1/2}$, y	Theory [18, 19] (Suhonen et al.)	Theory [23] (Stoica et al.)
^{150}Nd	2627.1	$= 1.4^{+0.5}_{-0.4} \times 10^{20}$ [8]	-	-
^{96}Zr	2202.5	$> 6.8 \times 10^{19}$ [22]	$(2.4 - 2.7) \times 10^{21}$	3.8×10^{21}
^{100}Mo	1903.7	$= 6.2^{+0.9}_{-0.7} \times 10^{20}$	1.6×10^{21} [29]	2.1×10^{21}
^{82}Se	1507.5	$> 3.0 \times 10^{21}$ [24]	$(1.5 - 3.3) \times 10^{21}$	-
^{48}Ca	1274.8	$> 1.5 \times 10^{20}$ [20]	-	-
^{116}Cd	1048.2	$> 2.0 \times 10^{21}$ [26]	1.1×10^{22}	1.1×10^{21}
^{76}Ge	916.7	$> 6.2 \times 10^{21}$ [30]	$(7.5 - 310) \times 10^{21}$	4.5×10^{21}
^{130}Te	735.3	$> 2.3 \times 10^{21}$ [31]	$(5.1 - 14) \times 10^{22*)}$	-

A.S. Barabash
arXiv:0710.2194 (2007)

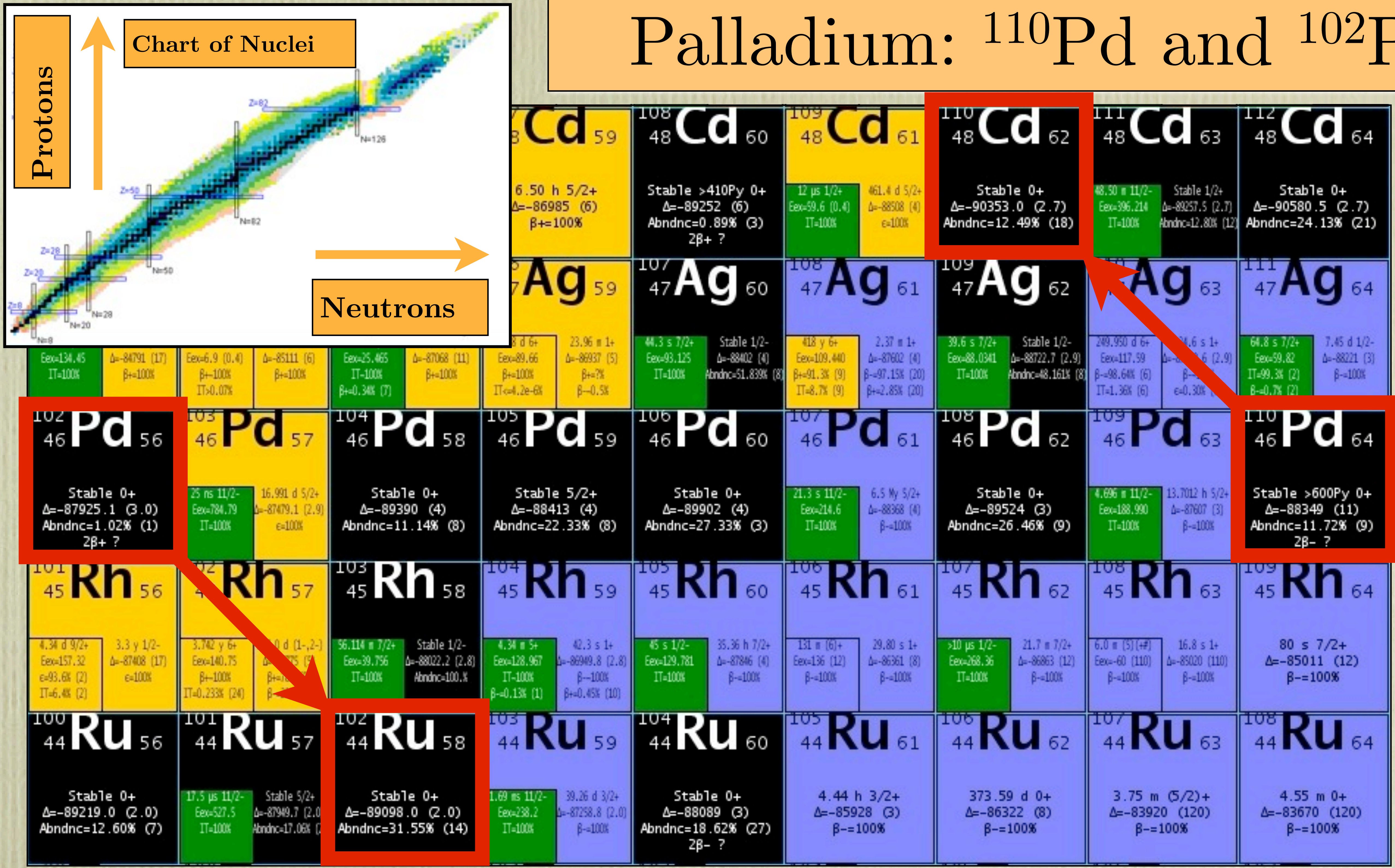
Why and why not Palladium?

Isotope	Q (MeV)	Percent natural abund.	Element cost [5] ($/kg)	$G^{0\nu}$ (10^{-14}/yr) [6]	$M^{0\nu}$ (avg) [7]	Annual world production [5] (tons)	$0\nu/2\nu$ rate [2,8] (10^{-8})
^{48}Ca	4.27	0.19	0.16	6.06	1.6	2.4×10^8	0.016
^{76}Ge	2.04	7.8	1650	0.57	4.8	118	0.55
^{82}Se	3.00	9.2	174	2.48	4.0	2000	0.092
^{96}Zr	3.35	2.8	36	5.02	3.0	1.4×10^6	0.025
^{100}Mo	3.04	9.6	35	3.89	4.6	2.5×10^5	0.014
^{110}Pd	2.00	11.8	23000	1.18	6.0	207	0.16
^{116}Cd	2.81	7.6	2.8	4.08	3.6	2.2×10^4	0.035
^{124}Sn	2.29	5.6	30	2.21	3.7	2.5×10^5	0.072
^{130}Te	2.53	34.5	360	3.47	4.0	~150	0.92
^{136}Xe	2.46	8.9	1000	3.56	2.9	50	1.51
^{150}Nd	3.37	5.6	42	15.4	2.7	$\sim 10^4$	0.024

PHYSICAL REVIEW D
87, 071301(R) (2013)

- ^{110}Pd is one out 11 DBD candidates with a Q-value above 2 MeV
- ^{110}Pd has the 2nd highest natural abundance
- ^{102}Pd is a double EC candidate
- But: Extremely expensive and no Pd detector technology

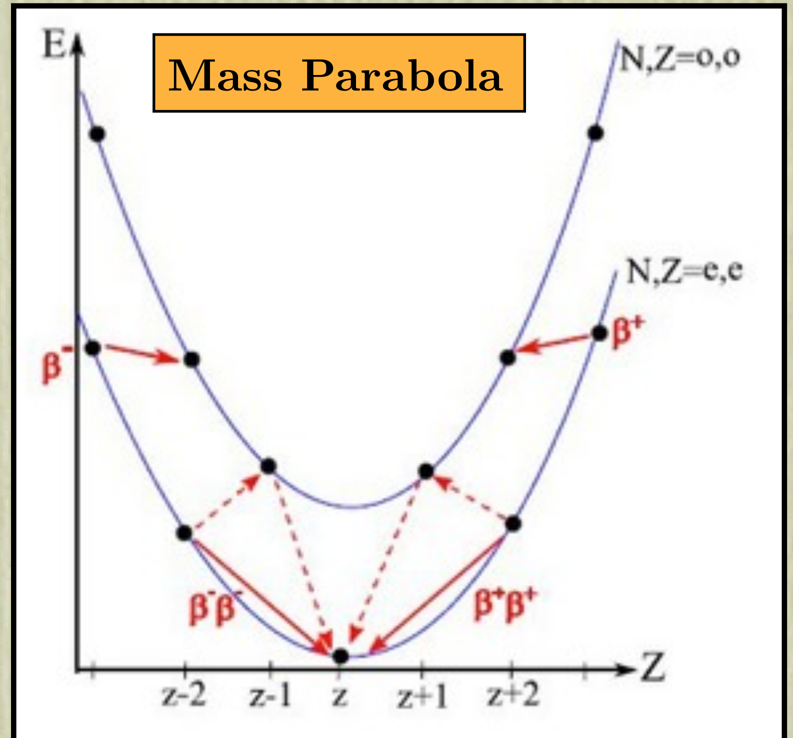
Palladium: ^{110}Pd and ^{102}Pd



- ^{102}Pd: 2νECEC, 2νECβ^{+}
- nat abundance: 1.02 %
- Q-value: 1172 keV

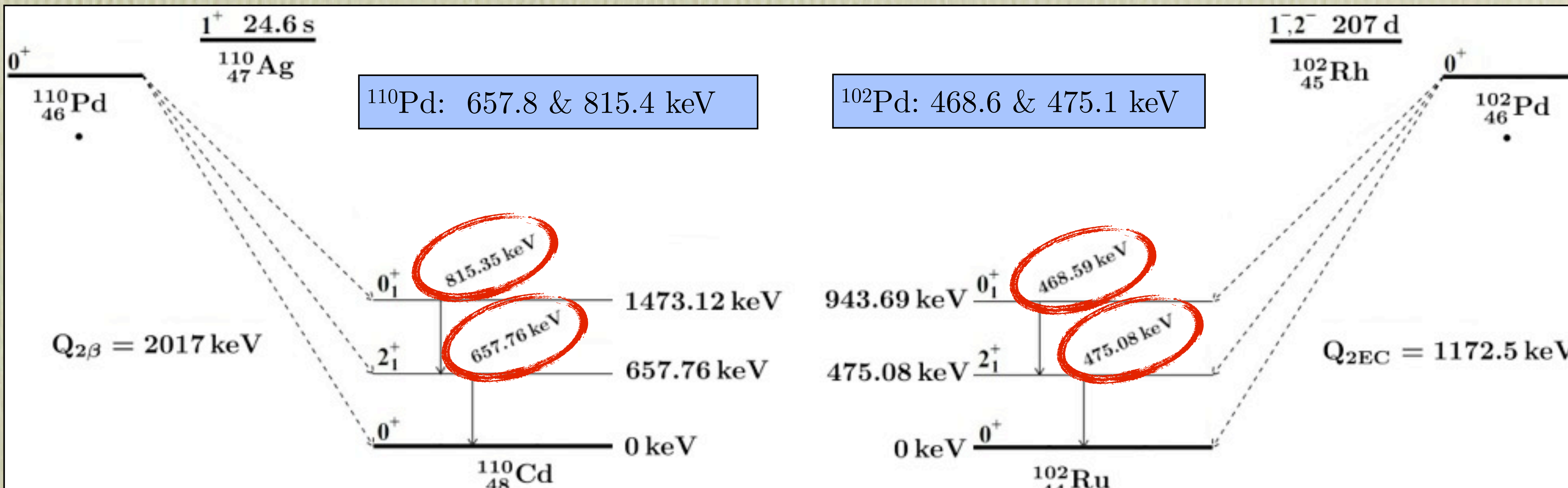
- ^{110}Pd: 2νβ^{-}β^{-}
- nat abundance: 11.72 %
- Q-value 2017.9 keV

PRL 108, 062502 (2012)



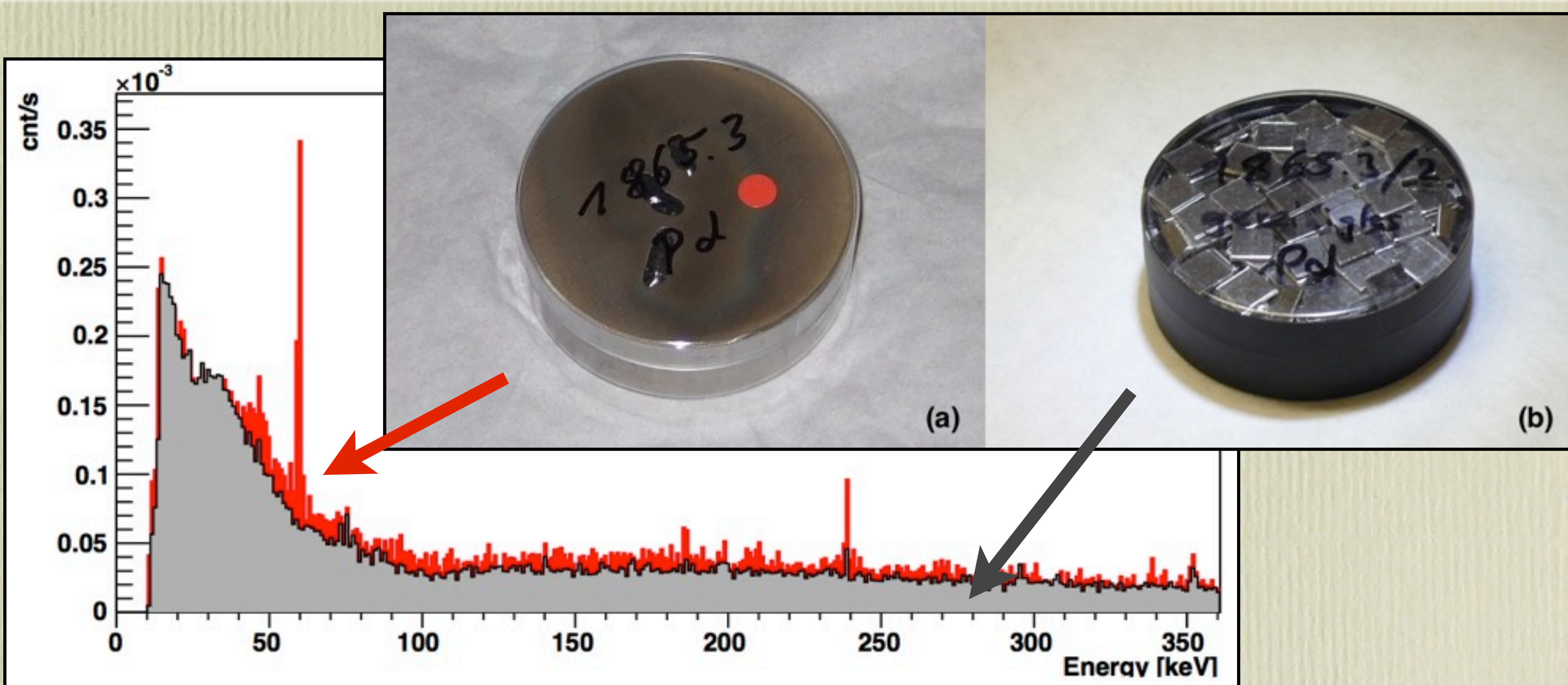
Decay Schemes: ^{110}Pd and ^{102}Pd

- ^{110}Cd and ^{102}Ru have excited 2^+ and 0^+ states
- Probability depends on phase space and spin (highest rate expected from 0_1^+)
- Expected γ-cascade



- Idea: Measure sample with HPGe γ-spectroscopy in low background env.
- Use γ-lines from excited state transitions as experimental signatures
- Not sensitive to g.s. transitions; No separation between 2νββ and 0νββ

Palladium Sample



- (a) Pd block from unknown origin
- Contaminated with ^{241}Am
- Cleaned by *C. HAFNER GmbH+Co. KG* >99.95% purity
- (b) 802.35 g Pd in 1cm x 1cm x 1mm plates (ρ_{eff} =10.2 g/cm^3)

Pd DBD Knowledge

^{110}Pd

- Best & only experimental half-life limit for g.s. transition from 1952
- No previous experimental limit for excited state transitions
- Theoretical expectations starting with half-lives $O(10^{23})$ for 0_1^+

^{102}Pd

- No previous experimental limit
- No theoretical calculation

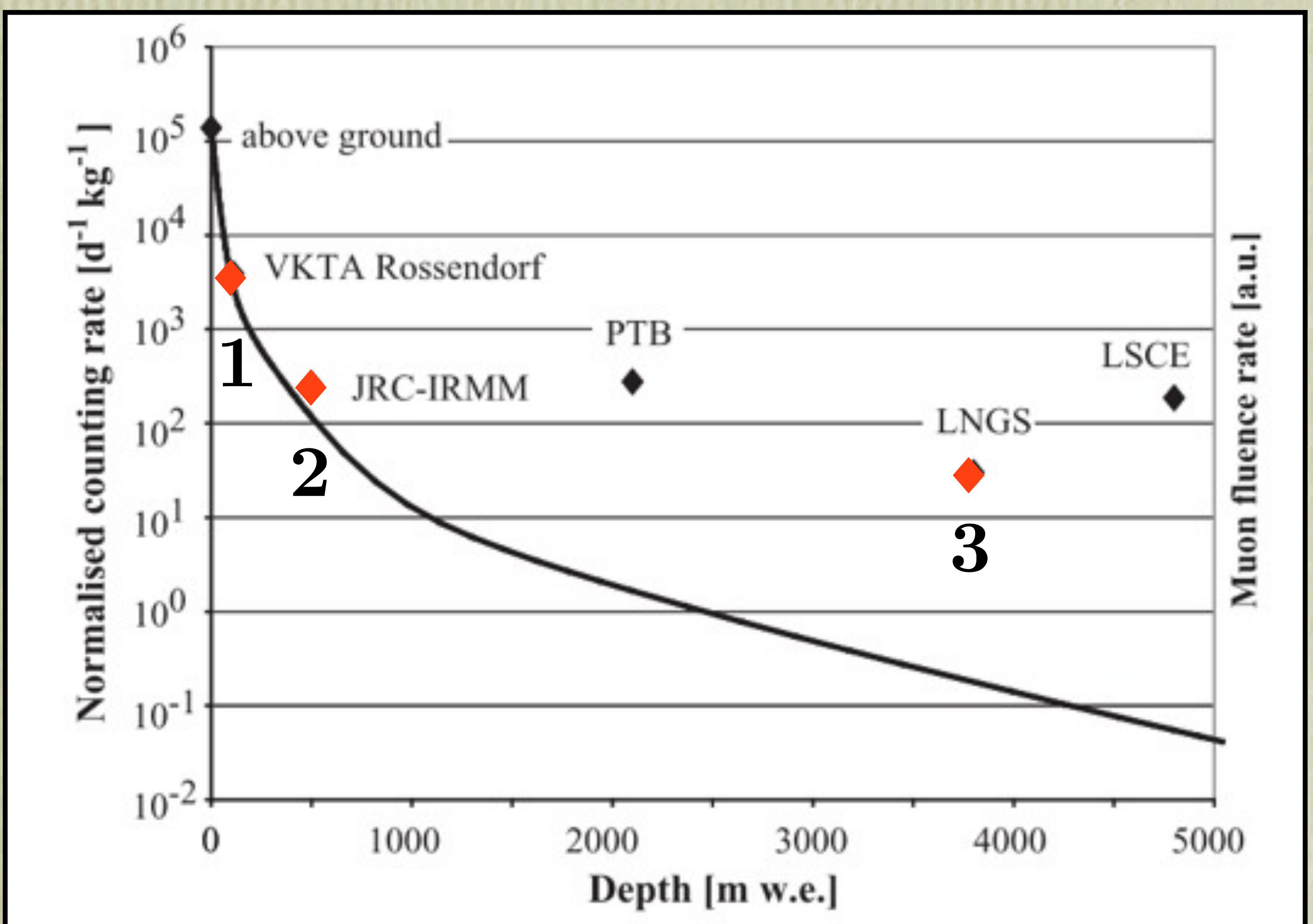
Physical Review C 87, 034312 (2013)

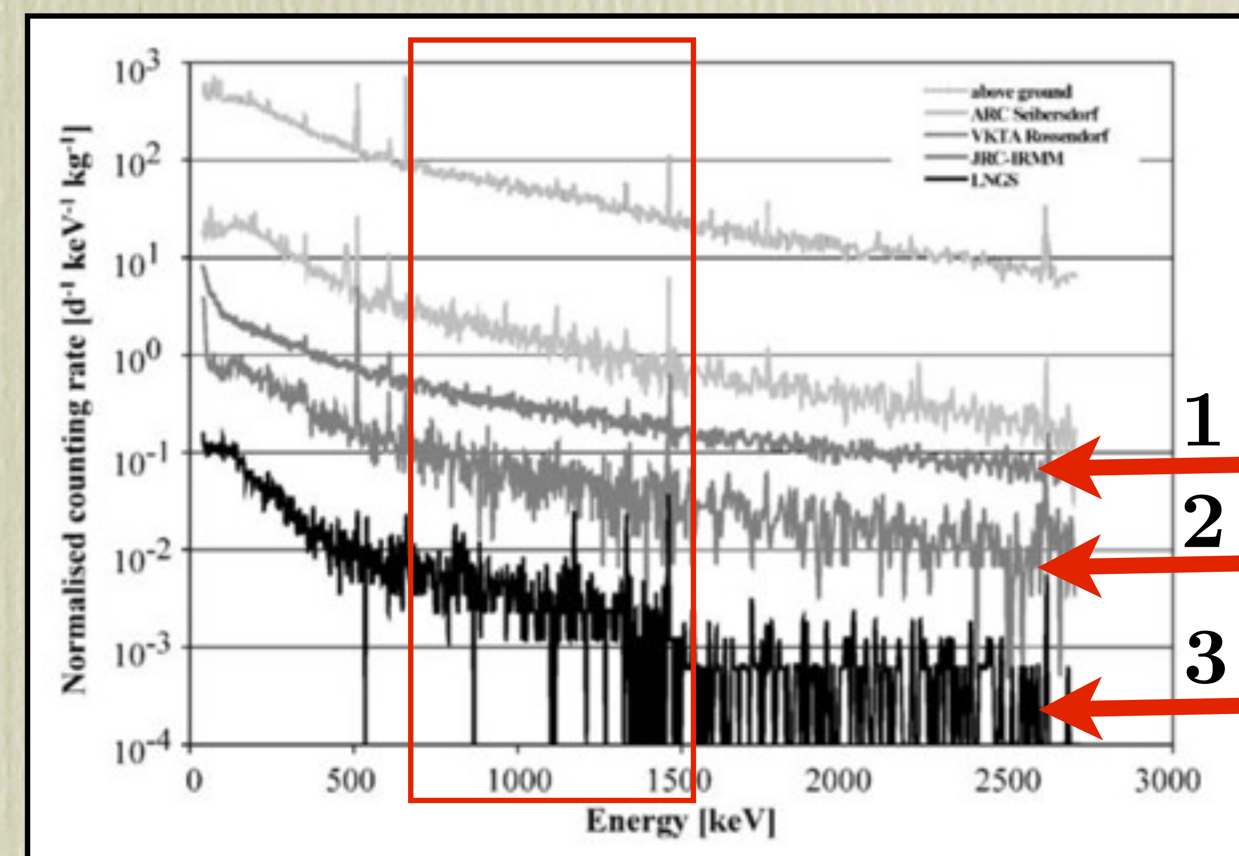
Expt./Th. model	Lower limit $T_{1/2}$ (yr)	Reference	Year of publication
^{110}Pd ground state			
Expt.	1×10^{17} (68% CL)	[24]	1952
PHFM	1.41×10^{20} and 3.44×10^{20} [a]	[26]	2005
SSDH	1.75×10^{20}	[27]	2000
SSDH	$1.2–1.8 \times 10^{20}$ [b]	[28]	1998
SRPA	1.6×10^{20}	[29]	1994
OEM	1.24×10^{21}	[30]	1994
QRPA	1.16×10^{19}	[31]	1990
SSD	1.2×10^{20}	[32]	2005
*pn*QRPA	1.1×10^{20} and 0.91×10^{20} [c]	[33]	2011
^{110}Pd 2_1^+ @ 657.76 keV			
Expt.	4.40×10^{19} (95% CL)	[25]	2011
SSD	4.4×10^{25}	[32]	2005
SRPA	8.37×10^{25}	[29]	1994
*pn*QRPA	1.48×10^{25}	[34]	2007
*pn*QRPA	0.62×10^{25} and 1.3×10^{25} [c]	[33]	2011
^{110}Pd 0_1^+ @ 1473.12 keV			
Expt.	5.89×10^{19} (95% CL)	[25]	2011
SSD	2.4×10^{26}	[32]	2005
*pn*QRPA	4.2×10^{23} and 9.1×10^{23} [c]	[33]	2011
^{110}Pd 2_2^+ @ 1475.80 keV			
SSD	3.8×10^{31}	[32]	2005
*pn*QRPA	11×10^{30} and 7.4×10^{30} [c]	[33]	2011
^{110}Pd 0_2^+ @ 1731.33 keV			
SSD	5.3×10^{29}	[32]	2005
^{110}Pd 2_3^+ @ 1783.48 keV			
SSD	1.3×10^{35}	[32]	2005

Three Experiments

The Palladium world tour:

1. Felsenkeller (Dresden, Germany)
2. HADES (Mol, Belgium)
3. LNGS (L'Aquila, Italy)

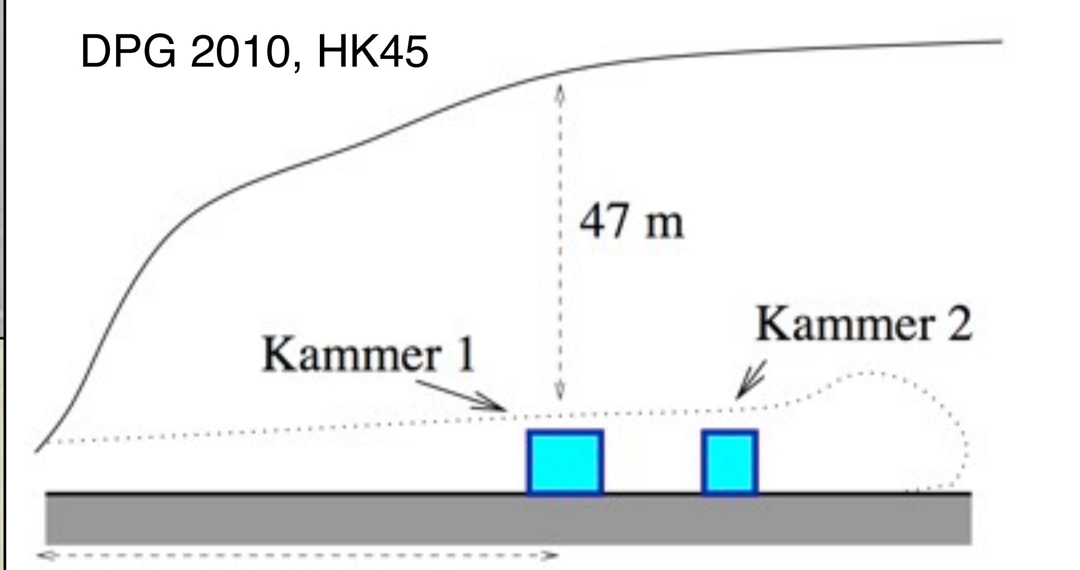




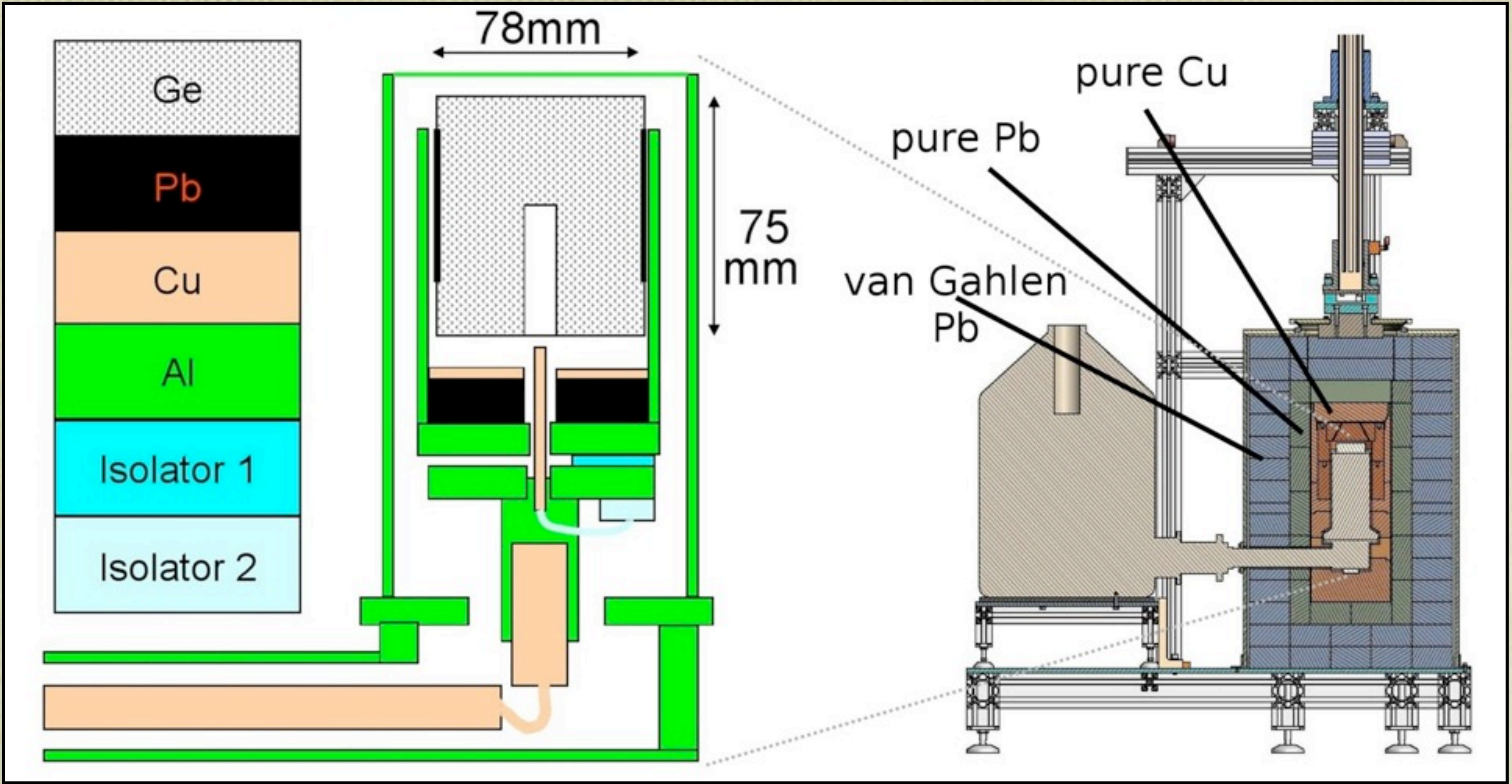
Applied Radiation and Isotopes 61 (2004) 167–172

VKTA Rossendorf - The Felsenkeller

- Built in 1982 in cavity of old brewery in Dresden, Germany
- Used to help decommissioning nuclear facilities
- 47 m Monzonite (120 m w.e.)
- μ-flux reduced by factor 20
- Up to 10 experimental HPGe setups in 2 chambers



The Detector



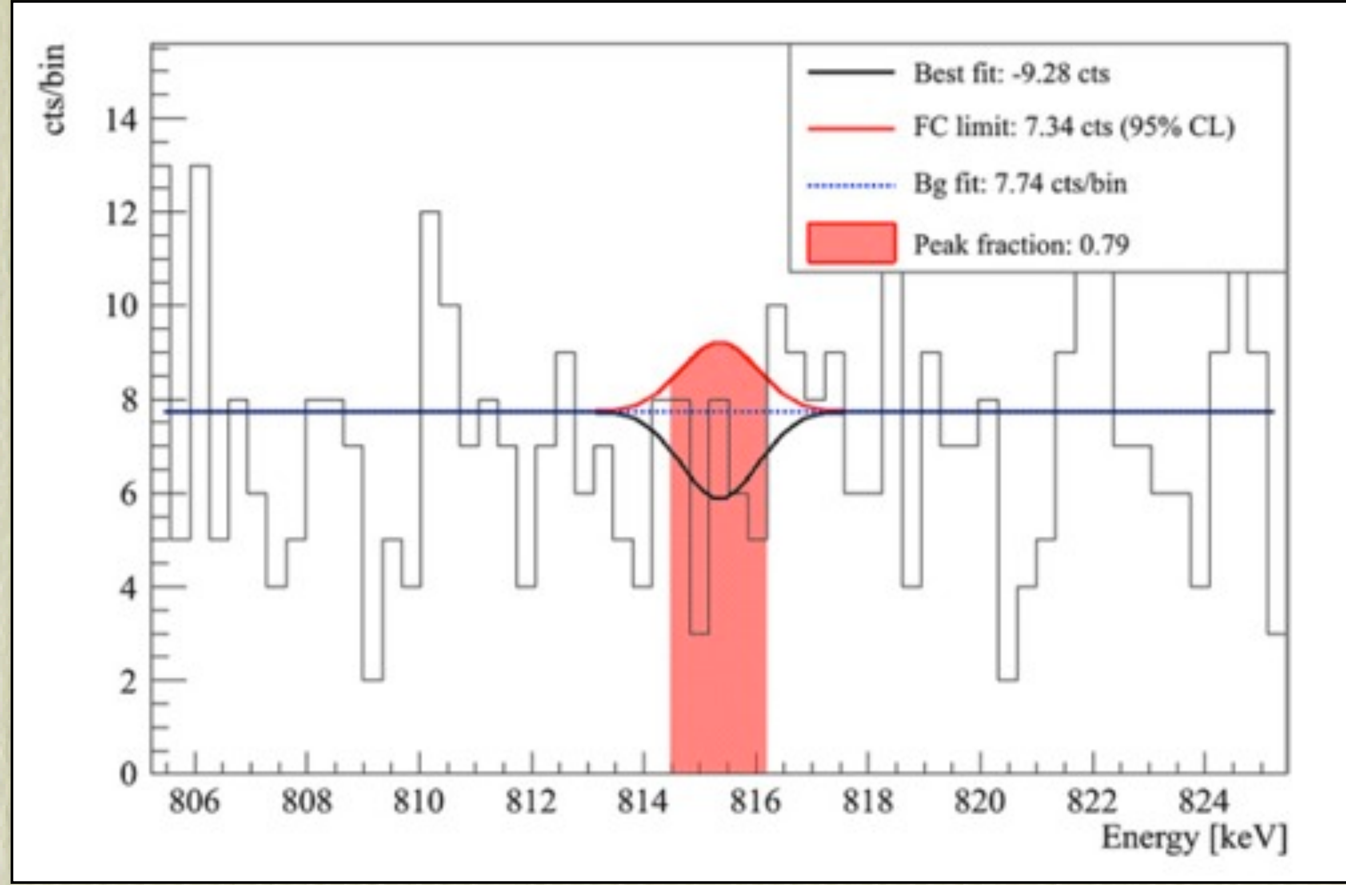
- HPGe Canberra 90 % efficiency
- Detector simulation with AMOS (Radiat. Protect. Dosim. 119 (2006) 479)

Analysis: Felsenkeller

Physics Letters B 705 47–51 (2011)
B. Lehnert, K. Zuber

- 16.2 d measurement
- 13.0 kg · y exposure

- Energy and efficiency calibration with 8 nuclides in SiO_2 sample geometry
- Correction for self-absorption with MC simulations with AMOS code
  - 1.6 keV FWHM @ 815 keV
  - 3.9 % FEP efficiency @ 815 keV



- Background model: Constant likelihood fit in side bands w/o γ-line
- Fit of gaussian signal can result in under-fluctuation
- Limit calculation using Feldman-Cousins in single analysis bin
- Quoting half-life limit for γ-line with best limit
- No systematic uncertainties considered for limit

Results:

Lower $T_{1/2}$ limits (95 % CL)

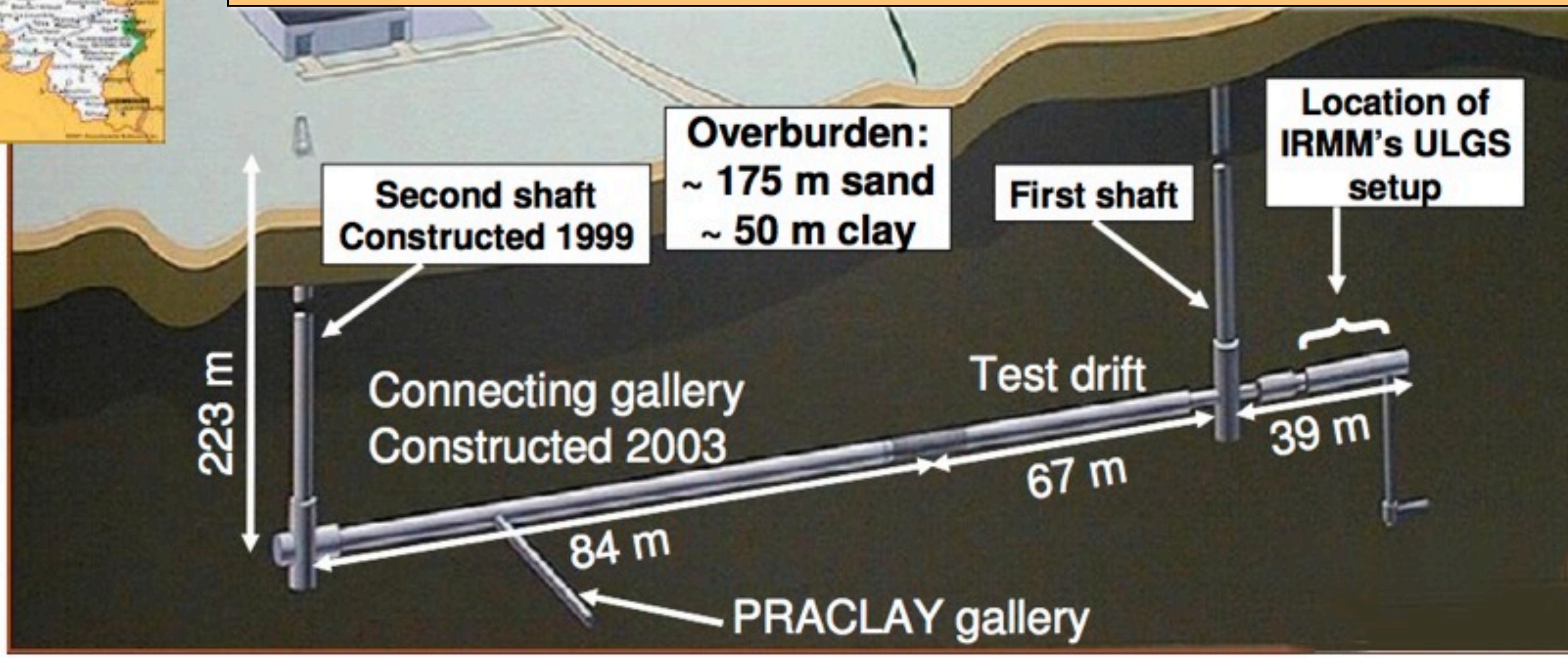
^{110}Pd 0_1^+: $> 5.89 \cdot 10^{19}$ yr

^{110}Pd 2_1^+: $> 4.40 \cdot 10^{19}$ yr

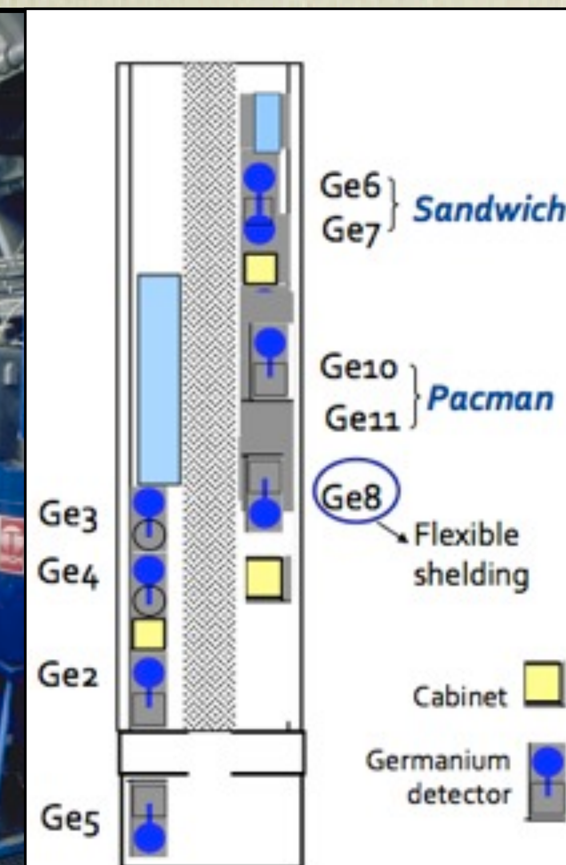
^{102}Pd 0_1^+: $> 7.64 \cdot 10^{18}$ yr

^{102}Pd 2_1^+: $> 2.68 \cdot 10^{18}$ yr

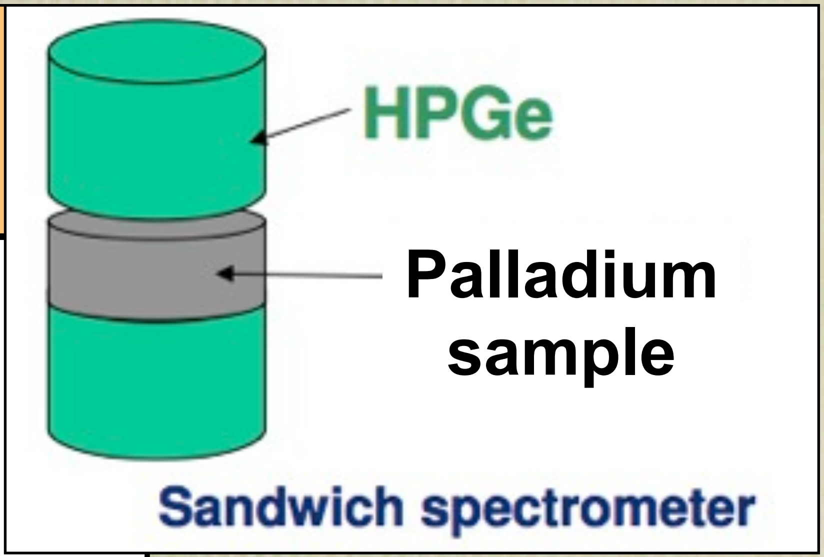
HADES - High Activity Disposal Experimental Site

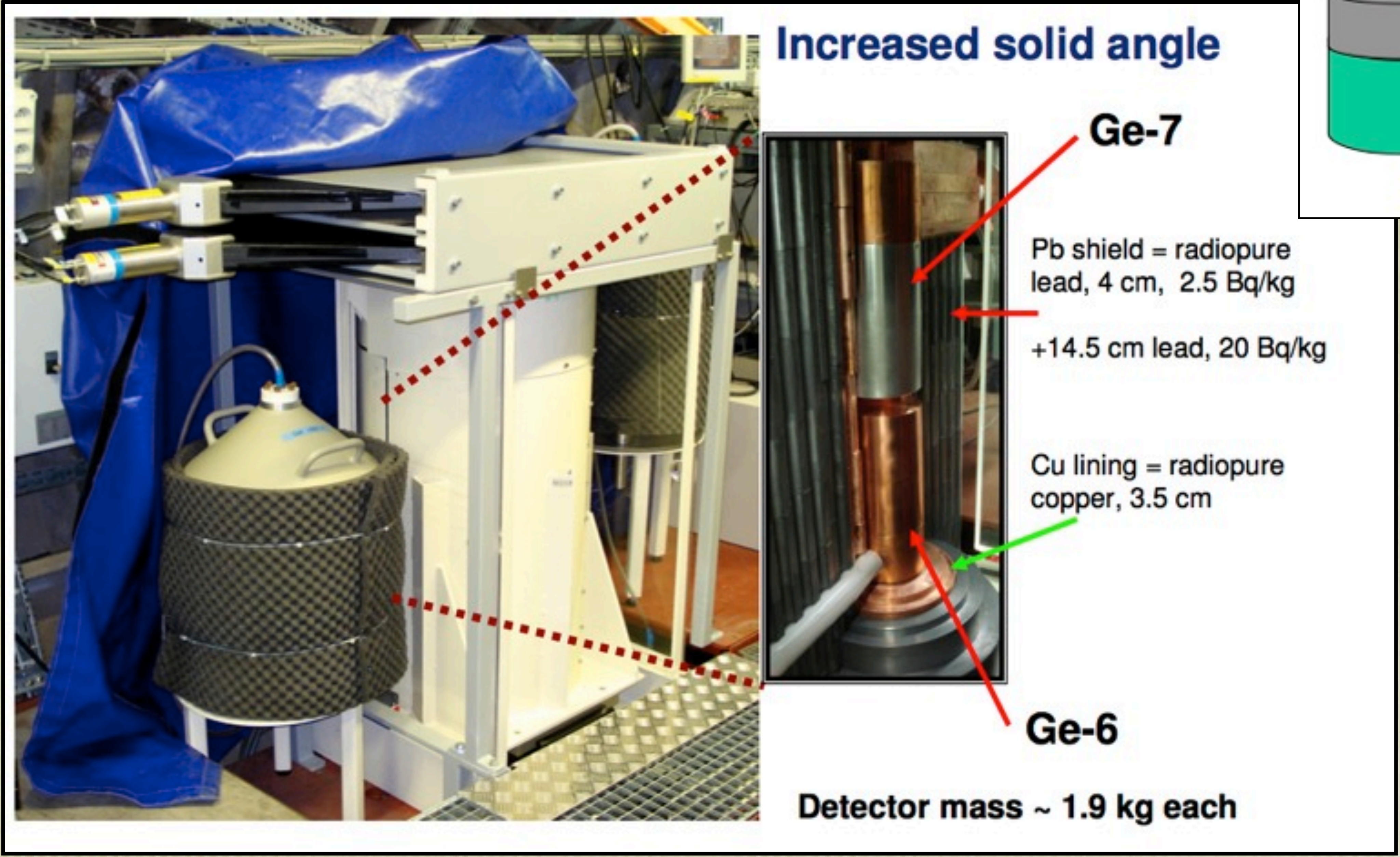


- Operated by SCK · CEN in Mol, Belgium
- Used to study disposal of nuclear waste in clay
- IRMM low-background laboratory
- 175 m overburden (500 m w.e.)
- μ-flux reduced by factor 1000



IRMM Sandwich Spectrometer





- 2 HPGe Canberra p-type with 80% and 90% efficiency; Muon-veto above detector
- Two DAQ:
  - Multi parameter system in list mode (offline)
  - MCA in histogram mode
- Detector simulation with EGS4

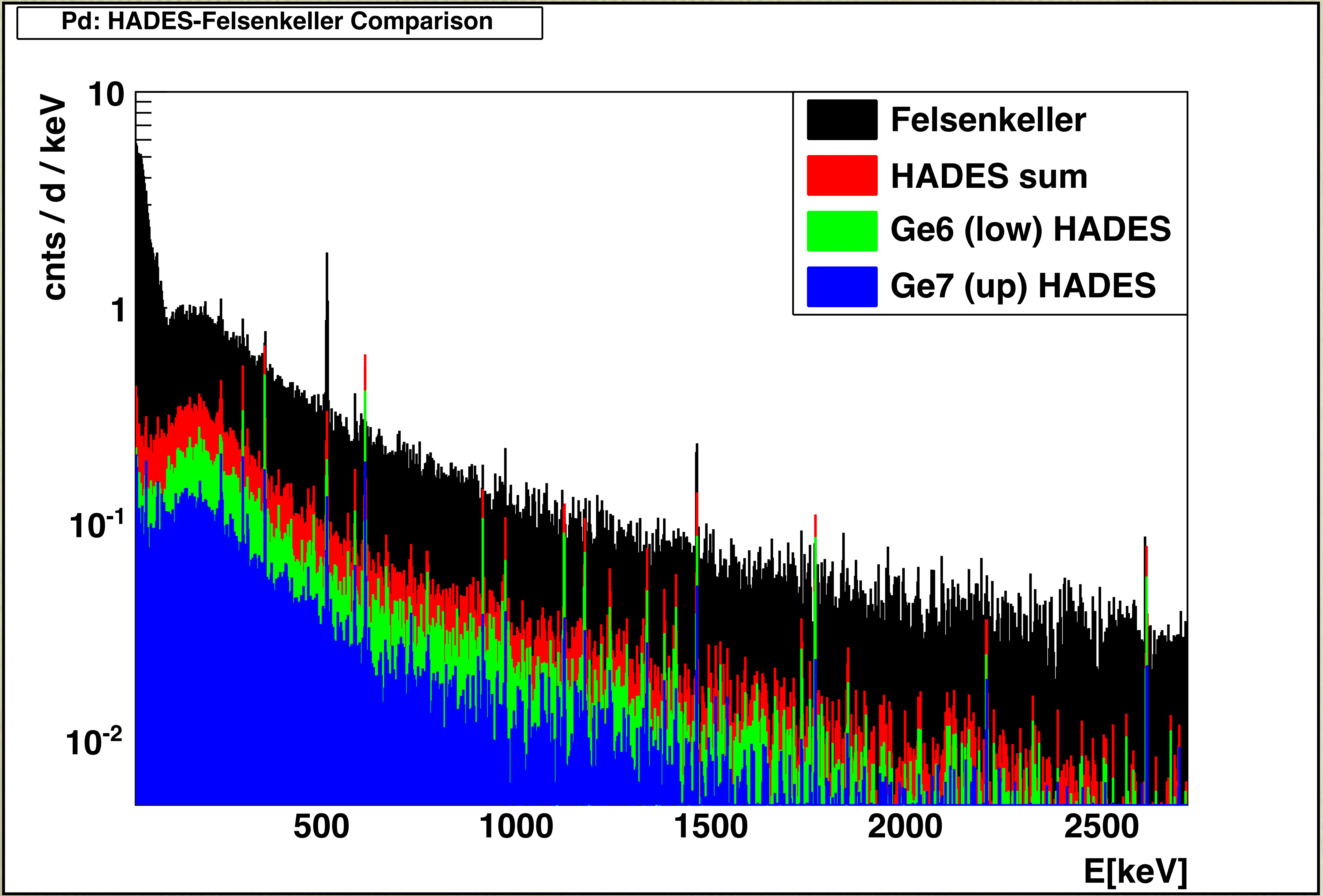
Analysis: HADES

Physical Review C 87, 034312 (2013)
B. Lehnert, K. Zuber, E. Andreotti, M. Hult

- Analysis only with single detector spectra (no coincidence possible)
- 46.5 d measurement; 35.9 kg y exposure
- Analysis analog to Felsenkeller measurement
- Limits for all possible decay branches were calculated
- Limit of best decay branch was quoted as limit of decay mode

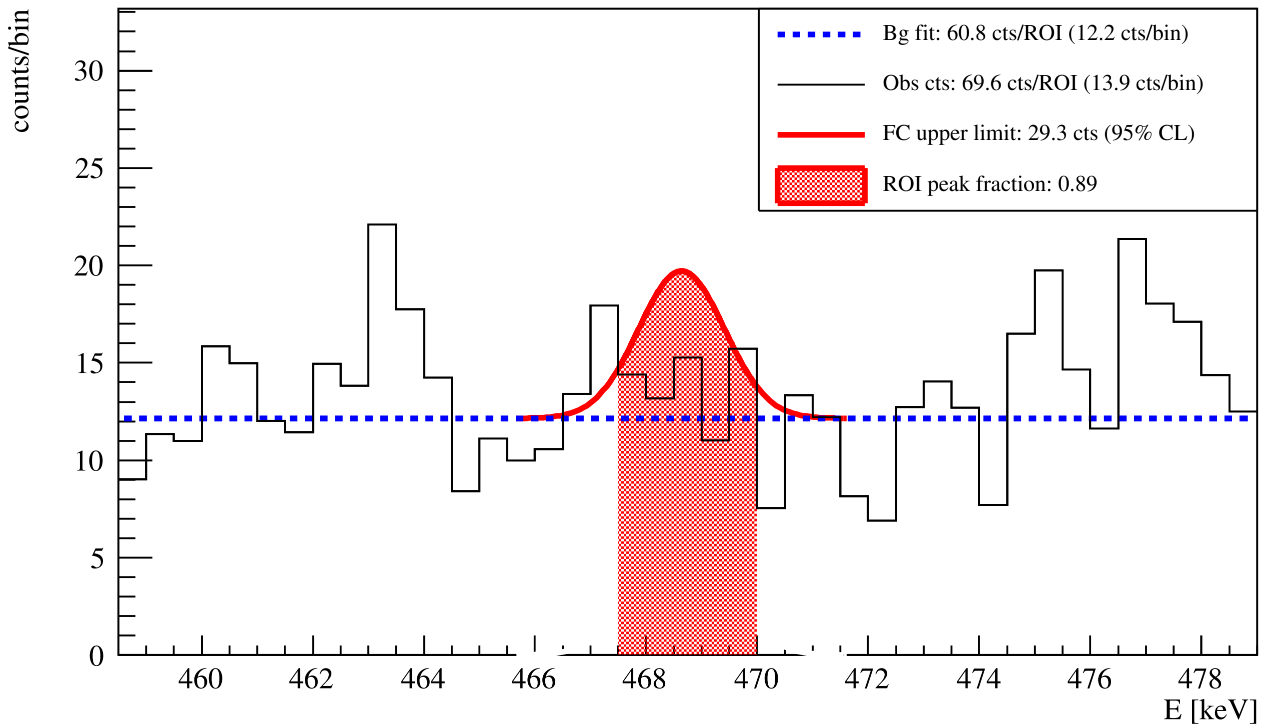
Decay mode	γ line energy (keV)	Emission probability	Detection efficiency	Signal count limit	$T_{1/2}$ limit (yr)
^{110}Pd 2_1^+ 657.76 keV	657.76 keV	100%	4.70%	12.4	1.72×10^{20}
^{110}Pd 0_1^+ 1473.12 keV	815.33 keV	100%	3.84%	8.4	1.98×10^{20}
	657.76 keV	100%	3.94%	12.4	1.44×10^{20}
^{110}Pd 2_2^+ 1475.80 keV	1475.80 keV	35.25%	1.32%	11.5	5.17×10^{19}
	818.02 keV	64.75%	2.40%	16.3	6.67×10^{19}
	657.76 keV	64.75%	2.53%	12.4	9.26×10^{19}
^{110}Pd 0_2^+ 1731.33 keV	1073.7 keV	86.73%	1.89%	10.1	8.50×10^{19}
	657.76 keV[a]	95.32%	3.78%	12.4	1.38×10^{20}
	255.49 keV	13.27%	0.36%	25.3	6.46×10^{18}
	1475.80 keV	4.68%	0.12%	11.5	4.87×10^{18}
	818.02 keV	8.59%	0.24%	16.3	6.63×10^{18}
^{110}Pd 2_3^+ 1783.48 keV	1783.48 keV	21.57%	0.88%	6.2	6.45×10^{19}
	1125.71 keV	78.43%	2.48%	12.0	9.41×10^{19}
	657.76 keV	78.43%	2.99%	12.4	1.09×10^{20}

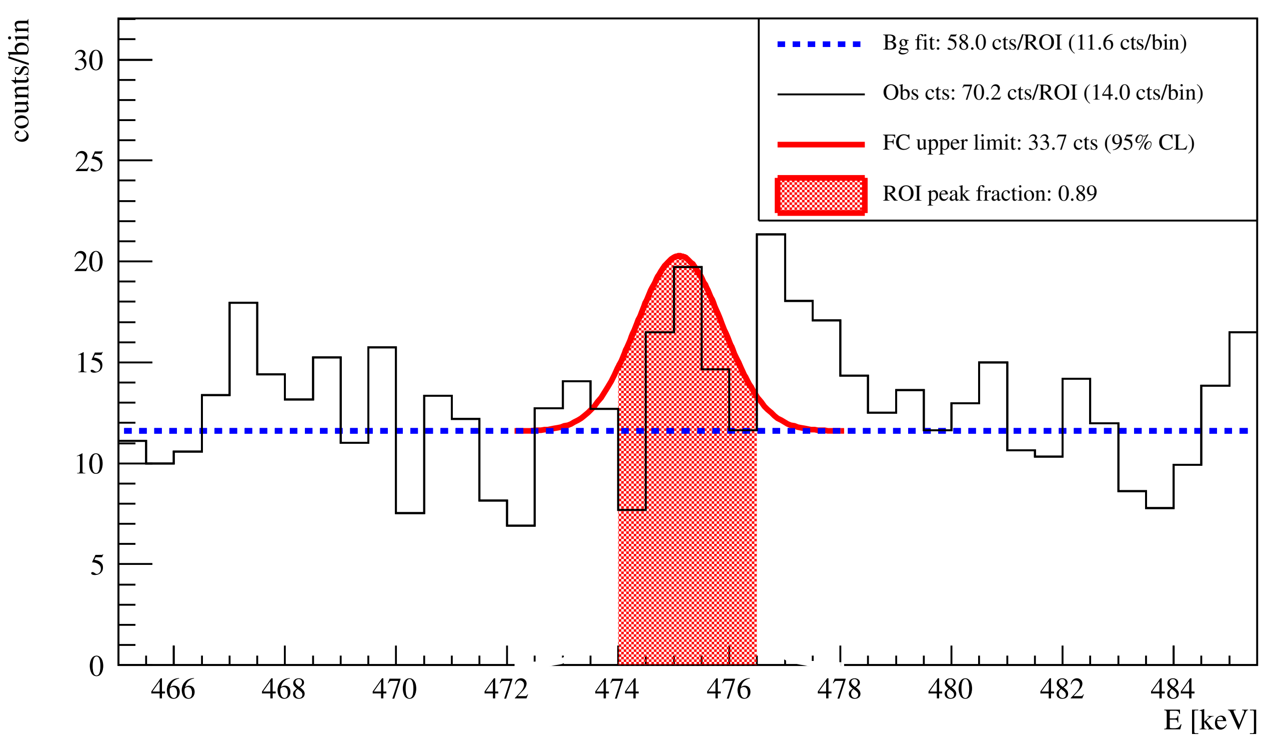
Full Spectra Comparison



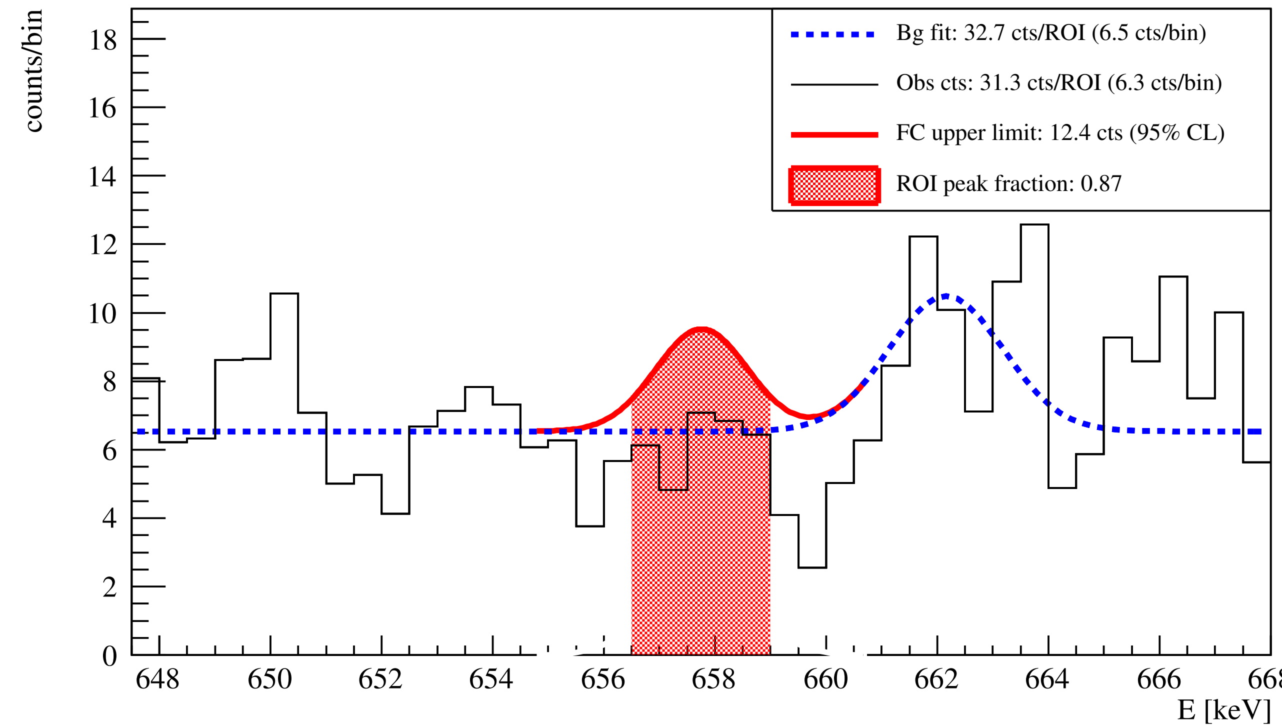
Region of Interests

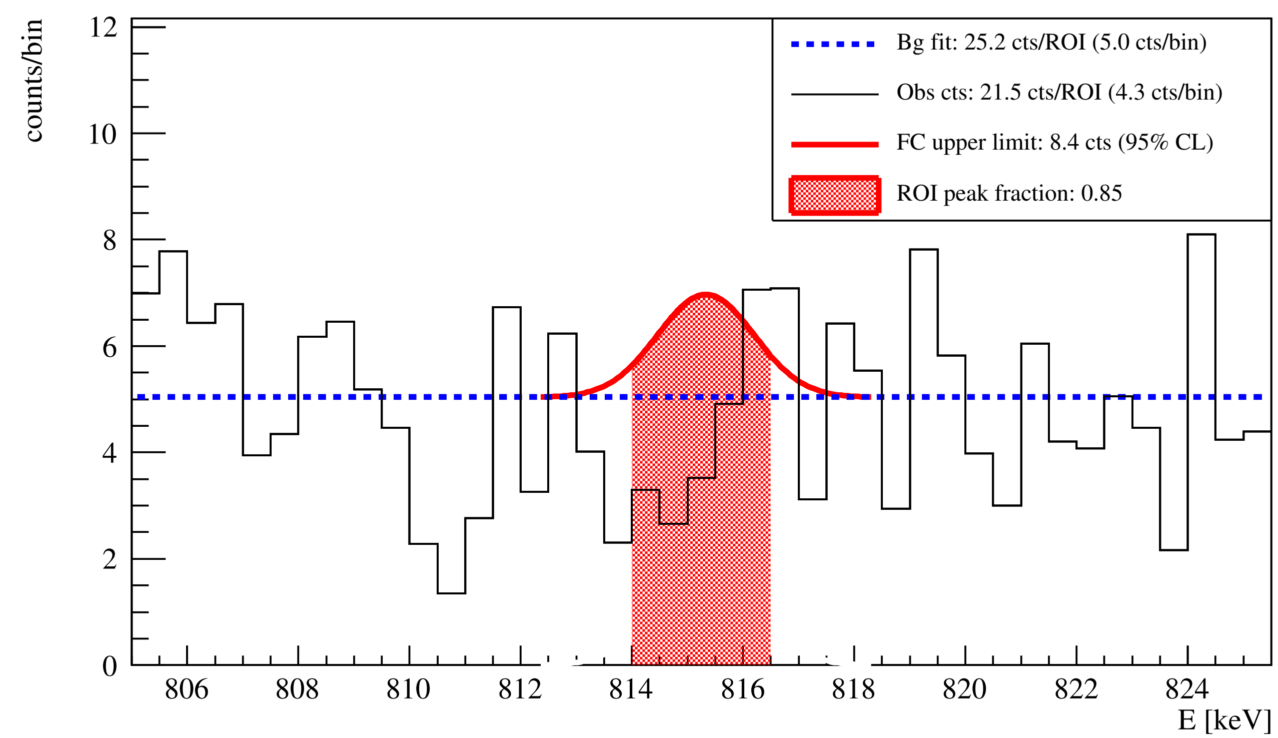
^{102}Pd





^{110}Pd

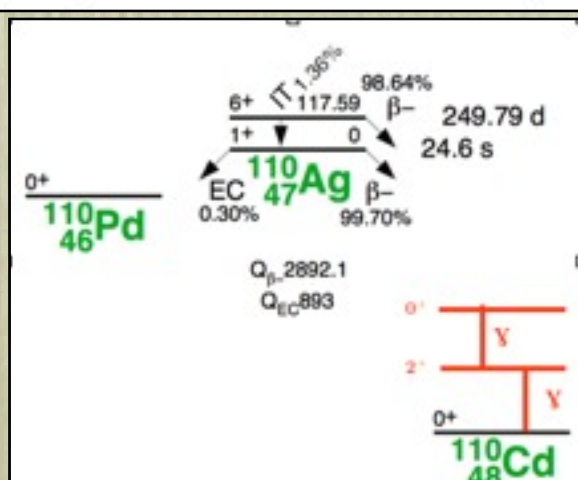


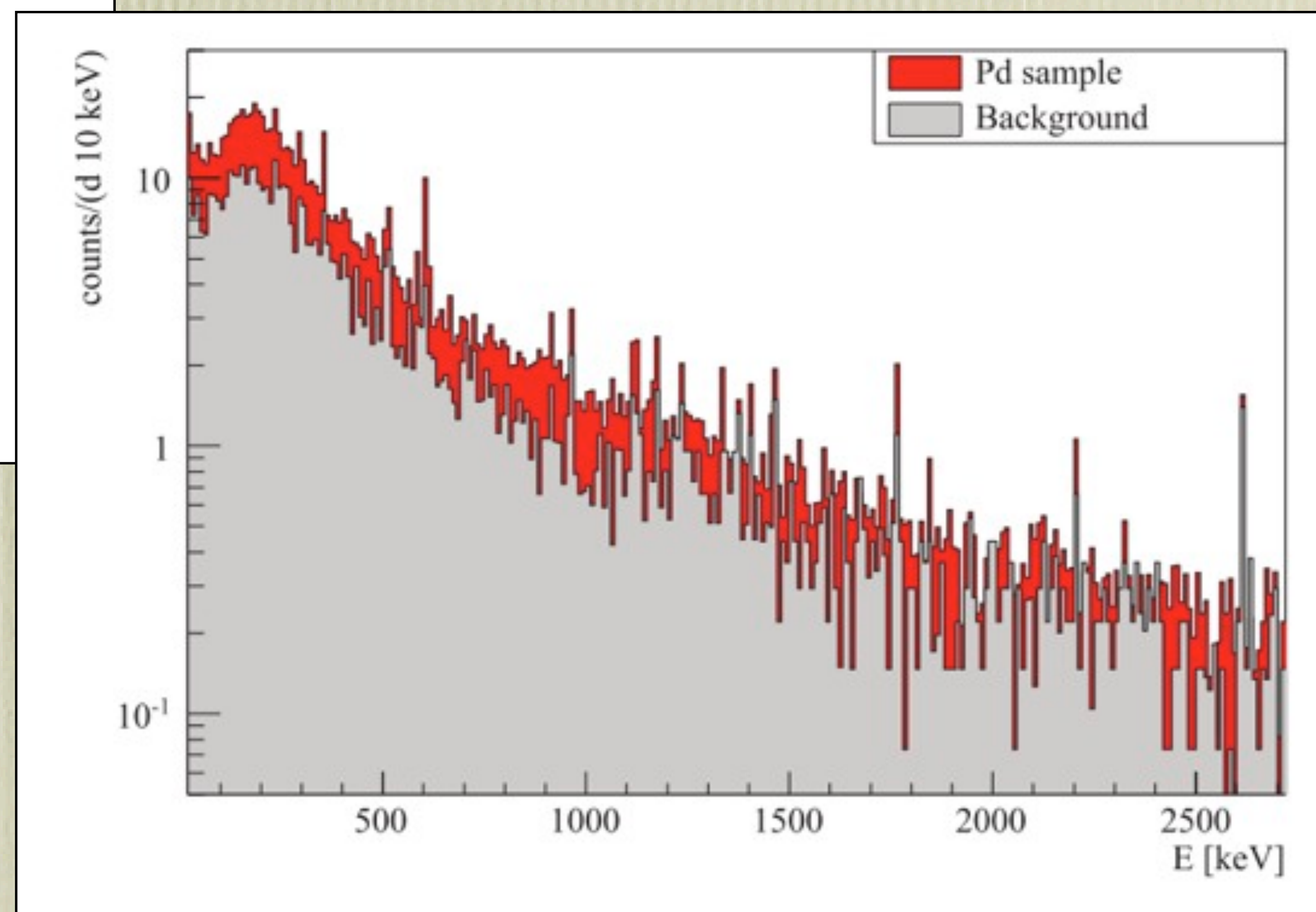


Background of Pd Sample

Nuclide	E (keV)	Massic activity (mBq/kg)	Decision threshold ($\alpha = 95\%$) (mBq/kg)	Weighted mean massic activity (mBq/kg)
^{214}Pb	295.22	1.9 ± 1.0	1.4	1.4 ± 0.4
	351.93	1.3 ± 0.5	0.6	
^{214}Bi	609.32	1.9 ± 0.4	0.4	1.9 ± 0.4
	1120.29	2.0 ± 0.8	0.9	
	1238.11	—	2.2	
	1377.67	—	2.7	
	1764.54	—	3.2	
^{210}Pb	46.54	—	414.3	
^{228}Ac	911.20	—	0.5	
	968.97	—	0.9	
^{212}Pb	238.63	—	0.7	
^{208}Tl	583.19	—	0.6	
	2614.51	—	0.3	
^{40}K	1460.82	—	1.0	
^{137}Cs	661.66	—	0.2	
^{60}Co	1173.23	—	0.2	
	1332.49	—	0.1	

- Only ^{214}Pb and ^{214}Bi with measured activity
- Background dominated by environment and not by sample
- No indication for irreducible background of ^{110m}Ag, ^{102}Rh, ^{102m}Rh
- Improvement possible external bg reduction





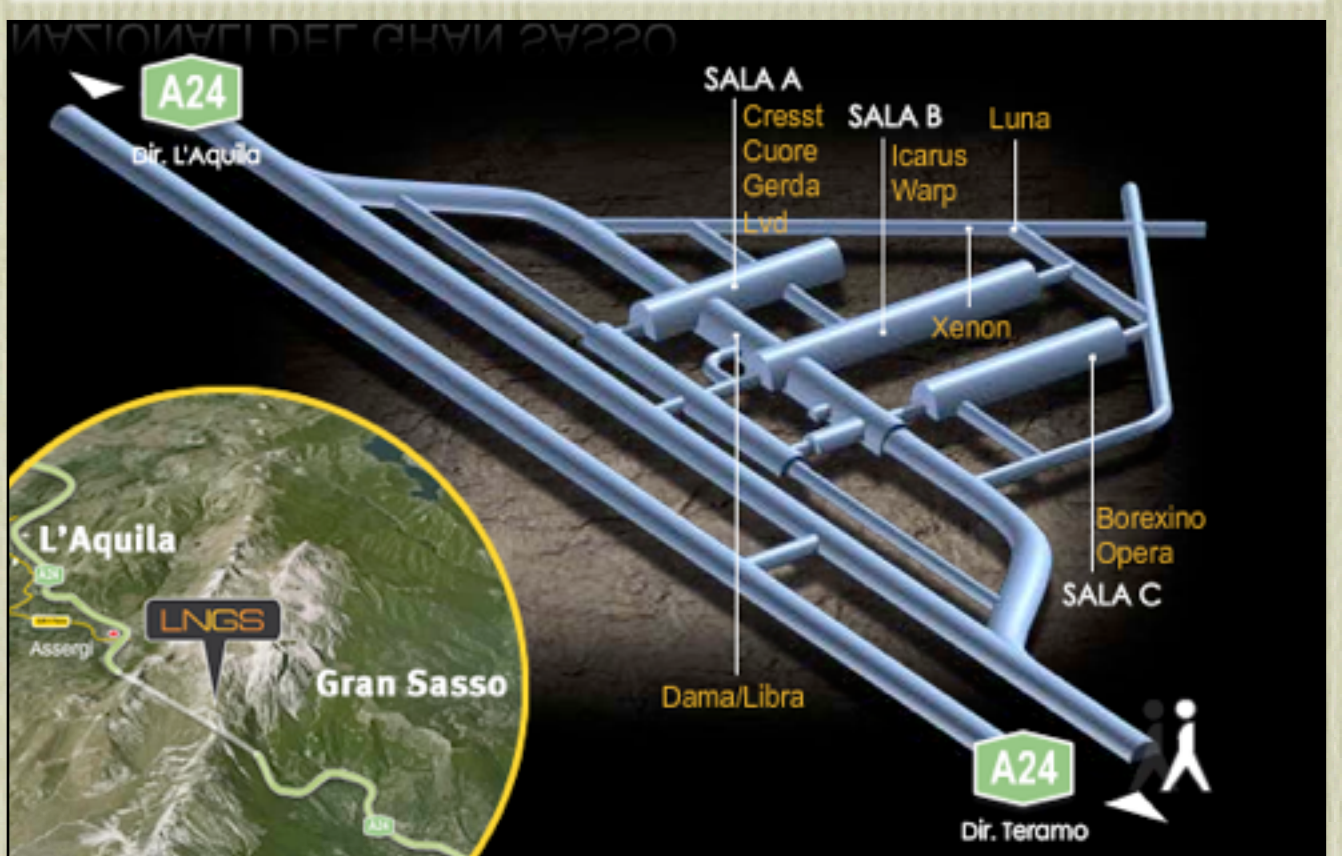
New Half-Life Limits for ^{110}Pd and ^{102}Pd

TABLE VI. Summary of measured half-life limits for all ^{110}Pd and ^{102}Pd double-β-decay excited-state transitions.

Decay mode	$T_{1/2}$ limit (yr) (95%)
^{110}Pd 2_1^+ 657.76 keV	1.72×10^{20}
^{110}Pd 0_1^+ 1473.12 keV	1.98×10^{20}
^{110}Pd 2_2^+ 1475.80 keV	9.26×10^{19}
^{110}Pd 0_2^+ 1731.33 keV	1.38×10^{20}
^{110}Pd 2_3^+ 1783.48 keV	1.09×10^{20}
^{102}Pd 2_1^+ 475.10 keV	5.95×10^{18}
^{102}Pd 0_1^+ 943.69 keV	5.81×10^{18}
^{102}Pd 2_2^+ 1103.05 keV	8.55×10^{18}

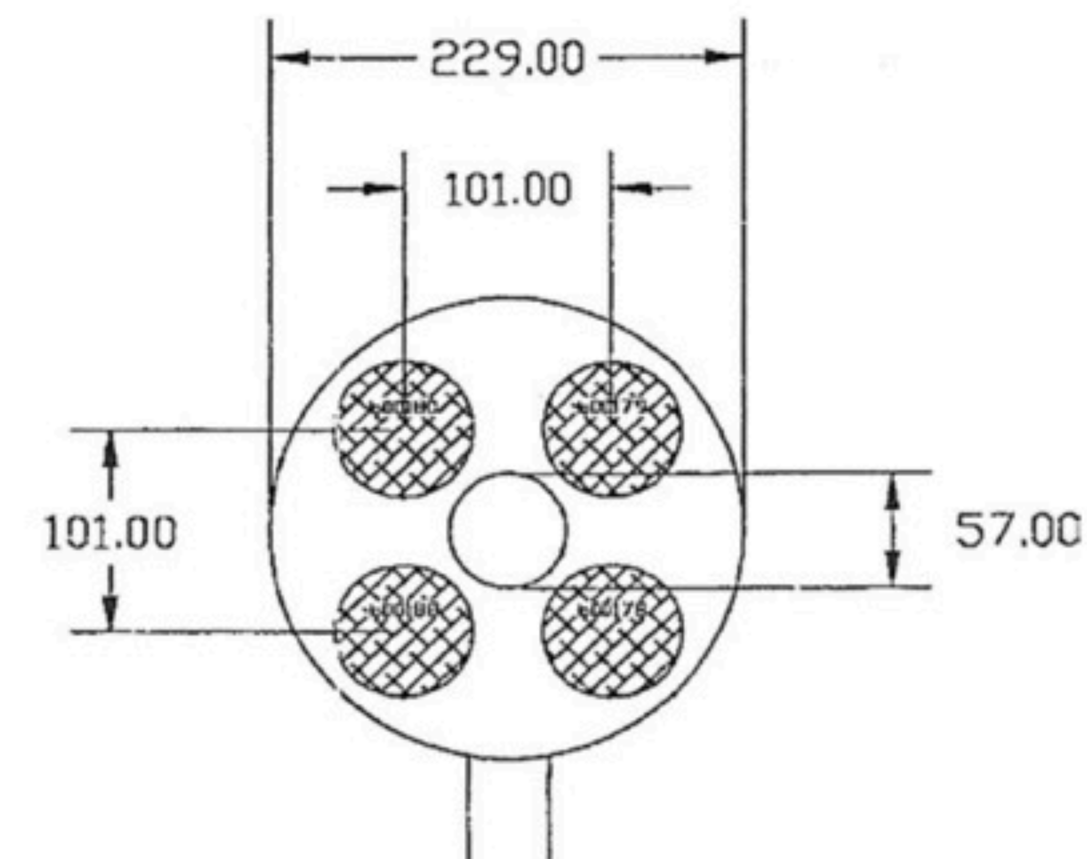
- Limit improved by factor 3 for 0_1^+ and 2_1^+ state in ^{110}Pd
- Experimental limits for all possible excited state transitions in ^{110}Pd and ^{102}Pd

Outlook @ LNGS



- 85 d with 4-HPGe setup @ LNGS
- μ-flux reduced by factor 10^6
- Higher efficiency
- True coincidence analysis will reduce the background significantly
- Further measurement planed with Pd plates in thin layer around n-type HPGe: Detection of EC X-rays

	Detectors			
	ge178	ge179	ge180	ge188
Volume (cm^3)	225.2	225.0	225.0	220.7
Endcap and holder material	Electrolytical copper			
Energy resolution (FWHM) at 1332 keV	2.1	2.0	2.0	2.0



Conclusions

- World first experimental limits for DBD into excited states in ^{110}Pd and ^{102}Pd
- Measurements in three state of the art underground gamma spectrometers
- Two measurements finished and published
- Third analysis is ongoing
- Challenge: 3 orders of magnitude improvement if theory is right

0_1^+ state

Nuclei	$E_{2\beta}$, keV	Experiment $T_{1/2}$, y	Theory [18, 19]	Theory [23]
^{150}Nd	2627.1	$= 1.4^{+0.5}_{-0.4} \times 10^{20}$ [8]	-	-
^{96}Zr	2202.5	$> 6.8 \times 10^{19}$ [22]	$(2.4 - 2.7) \times 10^{21}$	3.8×10^{21}
^{100}Mo	1903.7	$= 6.2^{+0.9}_{-0.7} \times 10^{20}$	1.6×10^{21} [29]	2.1×10^{21}
^{82}Se	1507.5	$> 3.0 \times 10^{21}$ [24]	$(1.5 - 3.3) \times 10^{21}$	-
^{48}Ca	1274.8	$> 1.5 \times 10^{20}$ [20]	-	-
^{116}Cd	1048.2	$> 2.0 \times 10^{21}$ [26]	1.1×10^{22}	1.1×10^{21}
^{76}Ge	916.7	$> 6.2 \times 10^{21}$ [30]	$(7.5 - 310) \times 10^{21}$	4.5×10^{21}
^{130}Te	735.3	$> 2.3 \times 10^{21}$ [31]	$(5.1 - 14) \times 10^{22*)}$	-
^{110}Pd	**544.8**	**$>2.0 \times 10^{20}$**	**$(4.2 - 9.1) \times 10^{23}$**	**-**

arXiv:0710.2194

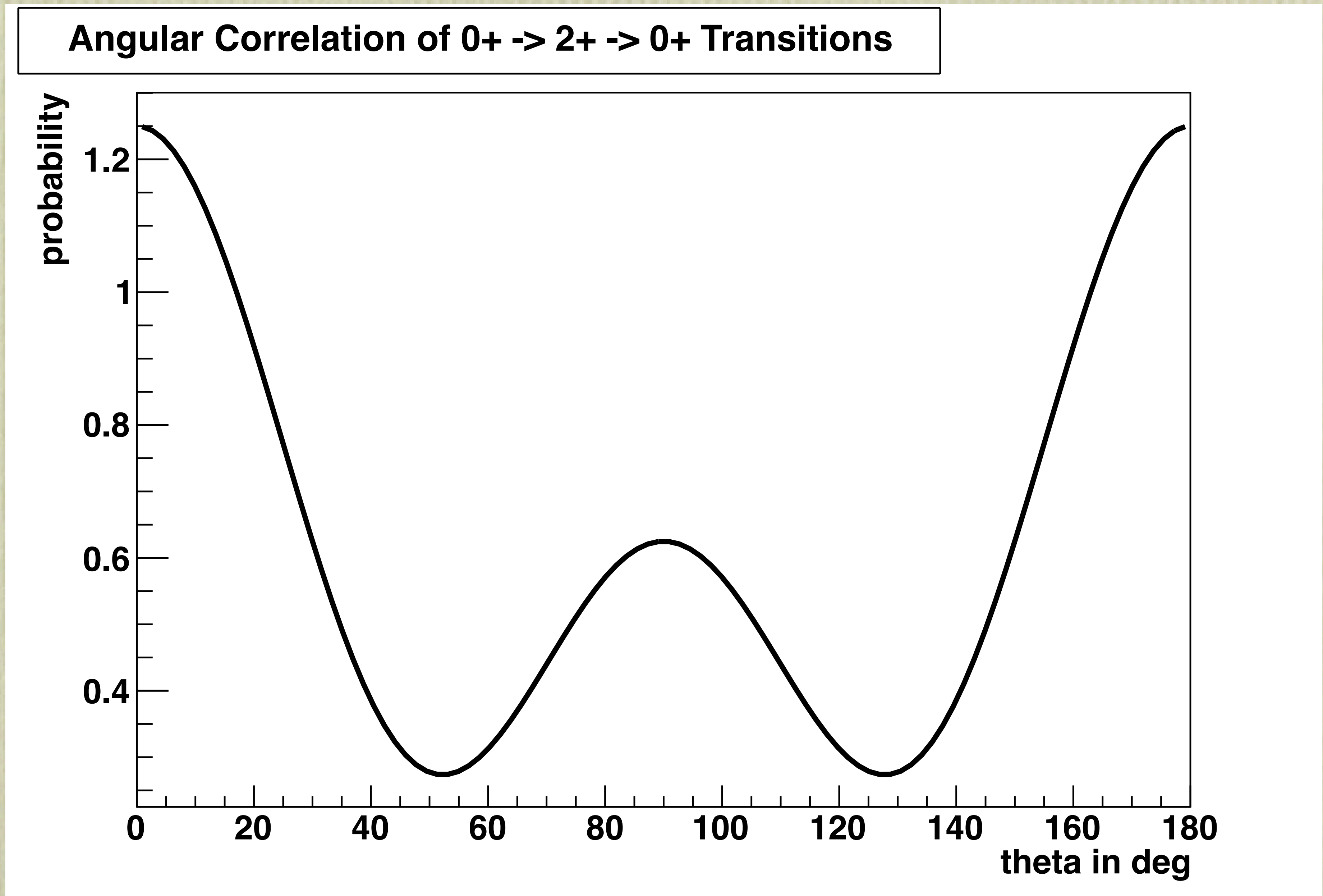
2_1^+ state

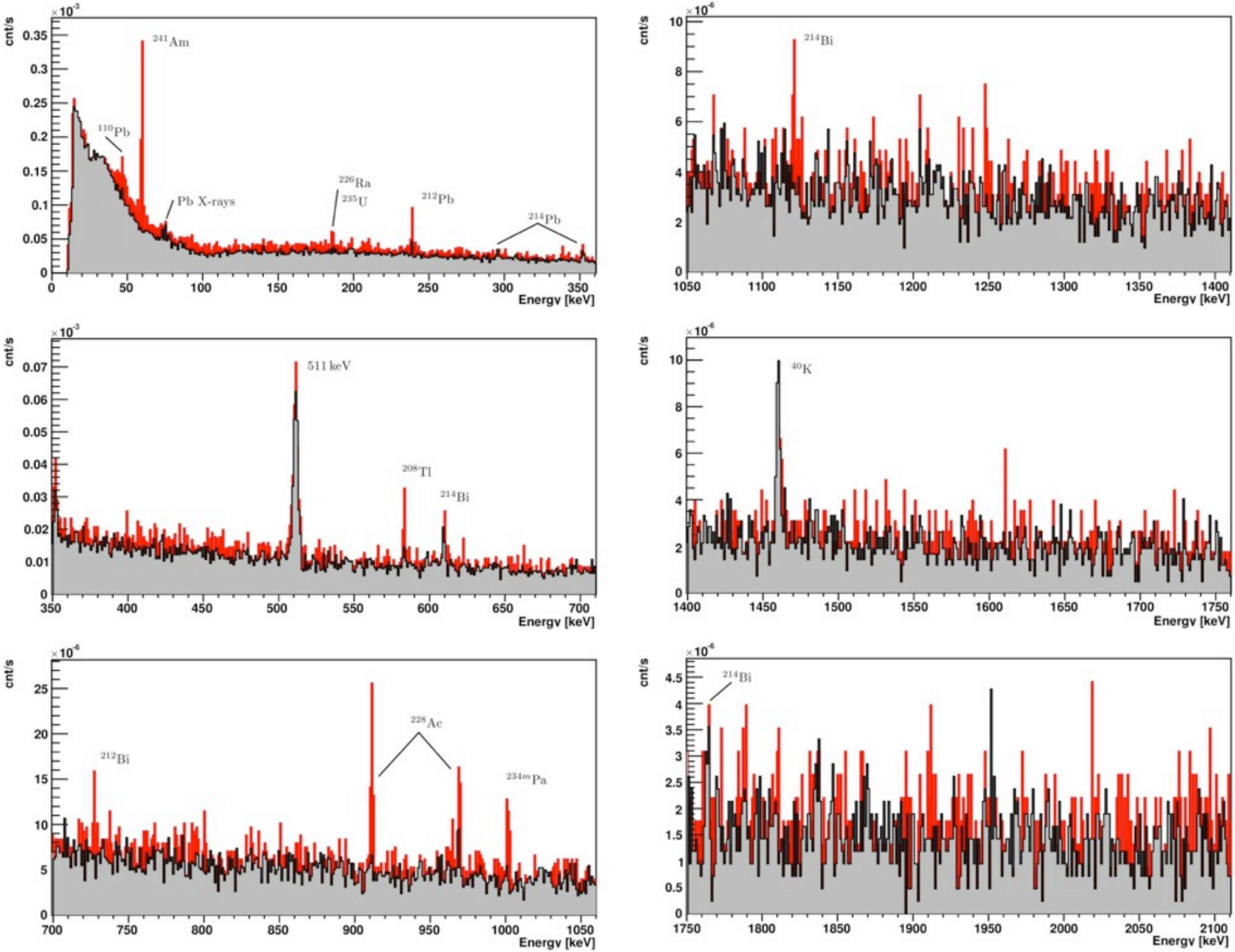
Nuclei	$E_{2\beta}$, keV	Experiment $T_{1/2}$, y	Theory [17]	Theory [18, 19]
^{48}Ca	3288.5	$> 1.8 \times 10^{20}$ [20]	1.7×10^{24}	-
^{150}Nd	3033.6	$> 9.1 \times 10^{19}$ [21]	-	-
^{96}Zr	2572.2	$> 7.9 \times 10^{19}$ [22]	2.3×10^{25}	$(3.8 - 4.8) \times 10^{21}$
^{100}Mo	2494.5	$> 1.6 \times 10^{21}$ [4]	1.2×10^{25}	3.4×10^{22} [23]
^{82}Se	2218.5	$> 1.4 \times 10^{21}$ [24]	-	2.8×10^{23}-3.3×10^{26}
^{130}Te	1992.7	$> 2.8 \times 10^{21}$ [25]	6.9×10^{26}	$(3.0 - 27) \times 10^{22}$
^{116}Cd	1511.5	$> 2.3 \times 10^{21}$ [26]	3.4×10^{26}	1.1×10^{24}
^{76}Ge	1480	$> 1.1 \times 10^{21}$ [27]	5.8×10^{28}	$(7.8 - 10) \times 10^{25}$
^{110}Pd	**1360.1**	**$>1.7 \times 10^{20}$**		**0.6-1.3×10^{25}**

BACKUP

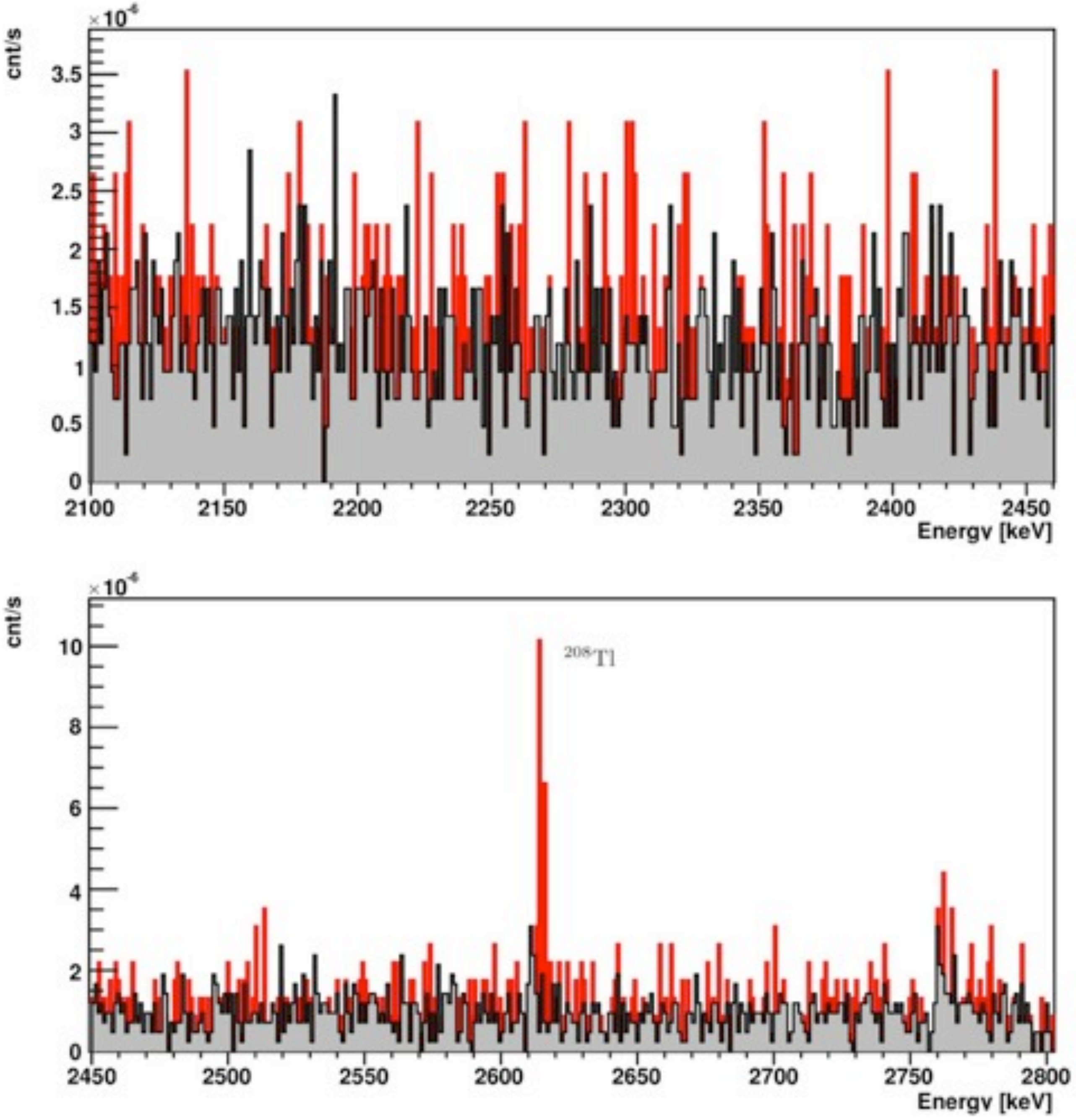
Angular Correlation

$$W(\theta) = \frac{5}{4}\left(1 - 3\cos^2\theta + 4\cos^4\theta\right).$$





cnt/s
Energy [keV]
²⁴¹Am
¹¹⁰Pb
Pb X-rays
²²⁶Ra
²³⁵U
²¹²Pb
²¹⁴Pb
²¹⁴Bi
511 keV
²⁰⁸Tl
⁴⁰K
²¹²Bi
²²⁸Ac
²³⁴ᵐPa


cnt/s
×10⁻⁶
Energy [keV]
cnt/s
×10⁻⁶
208Tl
Energy [keV]