

# Rare $\alpha$ and $\beta$ decays

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## 1. Introduction

## 2. Recent searches and discoveries of rare $\alpha$ decays

( $^{151}\text{Eu}$ ,  $^{180}\text{W}$ ,  $^{178\text{m}2}\text{Hf}^*$ ,  $^{190}\text{Pt}^*$ ,  $^{204,206,207,208}\text{Pb}$ ,  $^{209}\text{Bi}$ ,  $^{209}\text{Bi}^*$ )

## 3. Investigations of rare $\beta$ decays

( $^{48}\text{Ca}$ ,  $^{50}\text{V}$ ,  $^{96}\text{Zr}$ ,  $^{113}\text{Cd}$ ,  $^{115}\text{In}^*$ ,  $^{123}\text{Te}$ ,  $^{180\text{m}}\text{Ta}$ ,  $^{222}\text{Rn}$ )

## 4. Observation of emission of $e^+e^-$ pairs in $\alpha$ decay of $^{241}\text{Am}$

## 5. Conclusions

# 1. Introduction

## Classification of radioactive decays:

### Old known $\alpha$ , $\beta$ , $\gamma$ decays

$\alpha$ :  $(A,Z) \rightarrow (A-4,Z-2)$ , starting from  $^{106}\text{Te}$  to superheavy;  
 $T_{1/2}$  from  $10^{-8}$  s ( $^{217}\text{Ac}$ ) to  $10^{19}$  y ( $^{209}\text{Bi}$ )

$\beta$ :  $(A,Z) \rightarrow (A,Z\pm 1)$ , from  $^3\text{H}$ ; from  $10^{-2}$  s ( $^{11}\text{Li}$ ) to  $10^{16}$  y ( $^{113}\text{Cd}$ )

$\gamma$ :  $(A,Z)^* \rightarrow (A,Z)$ , from  $10^{-12}$  s to  $10^5$  y ( $^{186\text{m}}\text{Re}$ )

**Cluster decays:** emission of nuclides heavier than  $\alpha$  particle, from  $^{14}\text{C}$  to  $^{34}\text{Si}$  (~40 mothers from  $^{221}\text{Fr}$  to  $^{242}\text{Cm}$ , residue close to double magic  $^{208}\text{Pb}$  – “lead radioactivity”),  $10^3 - 10^{20}$  y; predicted in 1980 (or earlier?), observed in 1984

**$2\beta$  decays:** **allowed** in SM  $2\beta 2\nu$  in 13 nuclei ( $^{48}\text{Ca}$ ,  $^{76}\text{Ge}$ ,  $^{82}\text{Se}$ ,  $^{96}\text{Zr}$ ,  $^{100}\text{Mo}$ ,  $^{116}\text{Cd}$ ,  $^{128}\text{Te}$ ,  $^{130}\text{Te}$ ,  $^{136}\text{Xe}$ ,  $^{150}\text{Nd}$ ,  $^{238}\text{U} + ^{78}\text{Kr}$ ,  $^{130}\text{Ba}$ ),  $10^{18} - 10^{24}$  y; **forbidden** in SM  $2\beta 0\nu$   $T_{1/2} > 10^{25}$  y (in best cases of  $^{76}\text{Ge}$ ,  $^{136}\text{Xe}$ ; claim for observation in  $^{76}\text{Ge}$ )

**Spontaneous fission:** heavy nuclei from  $^{232}\text{Th}$ ;  $T_{1/2}$  from  $10^{-3}$  s ( $^{264}\text{Hs}$ ) to  $10^{19}$  y ( $^{235}\text{U}$ )

**p, 2p, 3p, 2n, ...:** in short living isotopes (~40 mothers); from ps to s

## 2. Recent searches and discoveries of rare $\alpha$ decays

### Recently discovered $\alpha$ decays:

2003 –  $^{209}\text{Bi}$

2003 –  $^{180}\text{W}$

2007 –  $^{151}\text{Eu}$

2007 –  $^{178\text{m}2}\text{Hf}^*$

2011 –  $^{190}\text{Pt}^*$

2012 –  $^{209}\text{Bi}^*$

### Limits:

2012 –  $^{151}\text{Eu}^*$

2013 –  $^{204,206,207,208}\text{Pb}$

Until 2003,  $^{209}\text{Bi}$  was considered as the heaviest stable isotope. However, in 2003 its alpha decay was discovered by P. De Marcillac et al., Nature 422 (2003) 876.

To-date, it has the longest  $T_{1/2}^{\alpha} \approx 10^{19}$  y (for g.s. to g.s. transition).

Viewing: Atomic weight

1																	18	
1	H																	He
2	Li	Be											B	C	N	O	F	Ne
3	Na	Mg									Al	Si	P	S	Cl	Ar		
4	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
5	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
6	Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
7	Fr	Ra	Ac	Rf	Db	Sg	Bh	Hs	Mt	Uun	Uuu	Uub						

Lanthanide Series	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
	140.12	140.91	144.24	(144.9)	150.36	151.97	157.25	158.93	162.5	164.93	167.26	168.93	173.04	174.97
Actinide Series	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
	232.04	231.04	238.03	(237)	(244.1)	(243.1)	(247.1)	(251.1)	(252.1)	(257.1)	(258.1)	(259.1)	(262.1)	



**$^{209}\text{Bi}$**

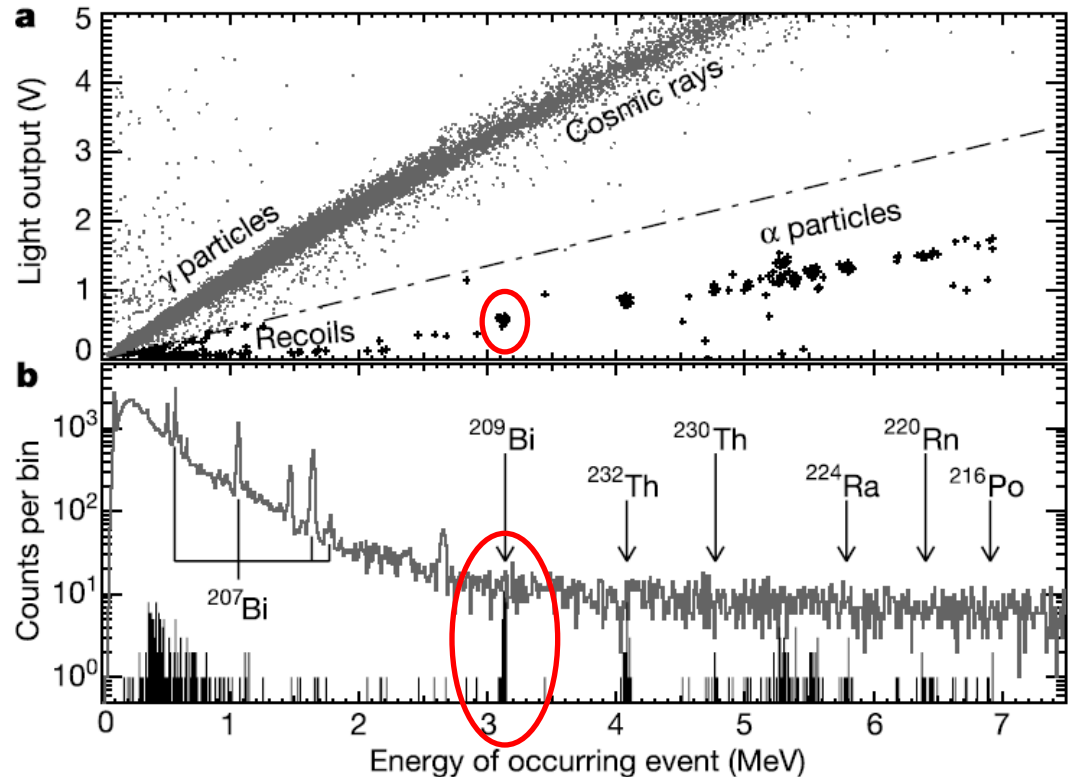
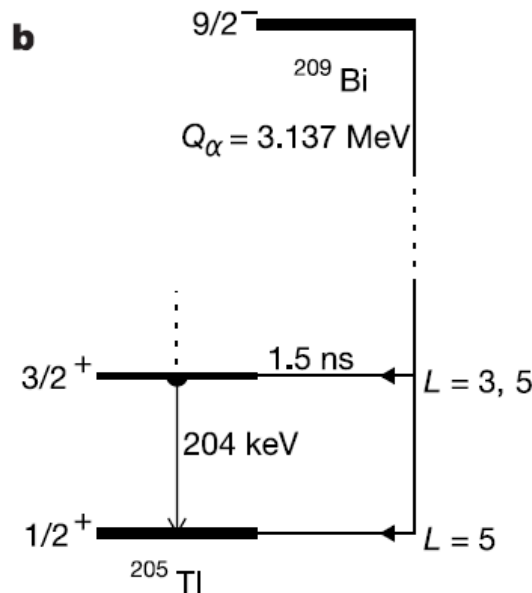
P. De Marcillac et al., Nature 422(2003)876

**$\text{Bi}_4\text{Ge}_3\text{O}_{12}$  scintillating bolometer 46 g, 20 mK (for  $^{209}\text{Bi}$  -  $\delta=100\%$ )**

**Heat and light signals – discrimination of  $\alpha$  and  $\beta/\gamma$  events by ratio of light/heat**

**Measurements (at Earth level) – 5 days, 128 observed events at  $Q_\alpha = 3.137$  MeV**

**$T_{1/2} = (1.9 \pm 0.2) \times 10^{19}$  y – the biggest half life ever measured for  $\alpha$  decays (g.s. to g.s.)**



**$^{180}\text{W}$**

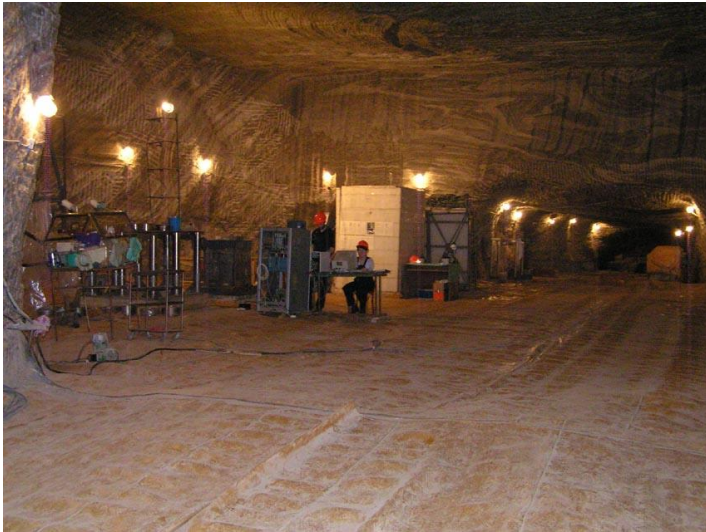
F.A. Danevich et al., Phys. Rev. C 67(2003)014310

**$\text{CdWO}_4$  scintillator, 330 g**

**Solotvina underground laboratory**

**(Ukraine, 1000 m w.e.)**

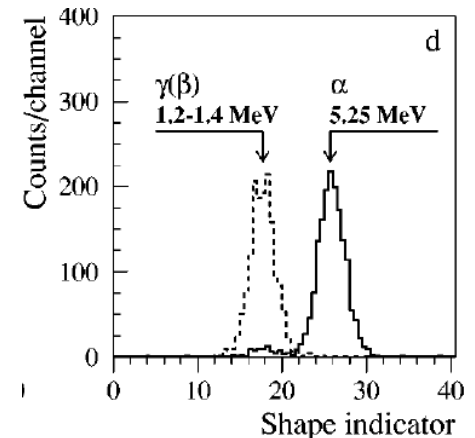
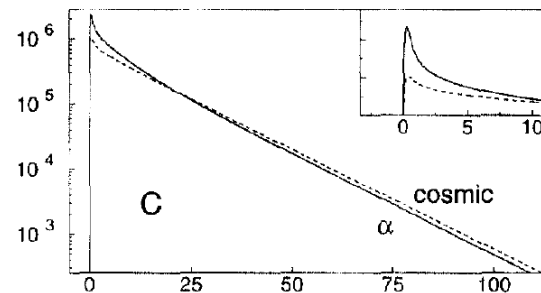
**2975 h of measurements in  
low background set-up**



**$\text{CdWO}_4$  crystals, Lviv, Ukraine, 2002**

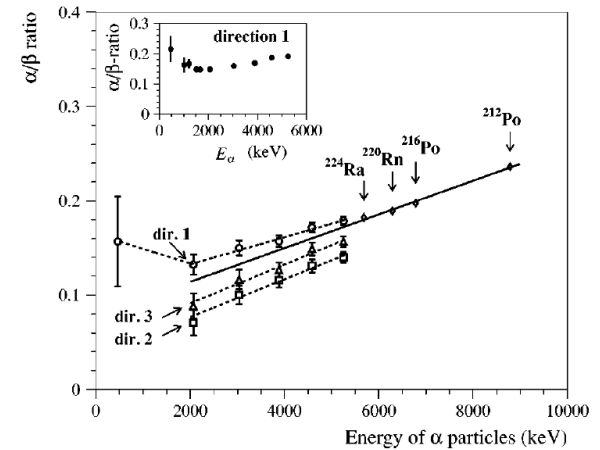
**Pulse shape discrimination  
between  $\alpha$  and  $\beta/\gamma$  events  
thanks to different evolution  
of scintillating signal in  
time:**

**$Q_\alpha = 2.516 \text{ MeV}$   
 $^{180}\text{W}$  ( $\delta = 0.12\%$ )**

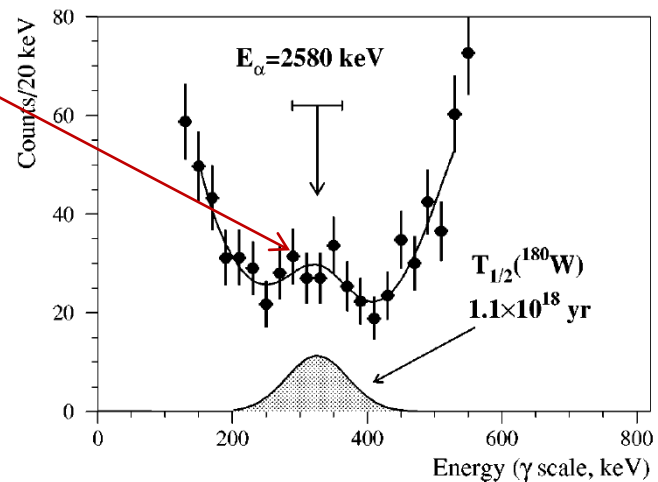
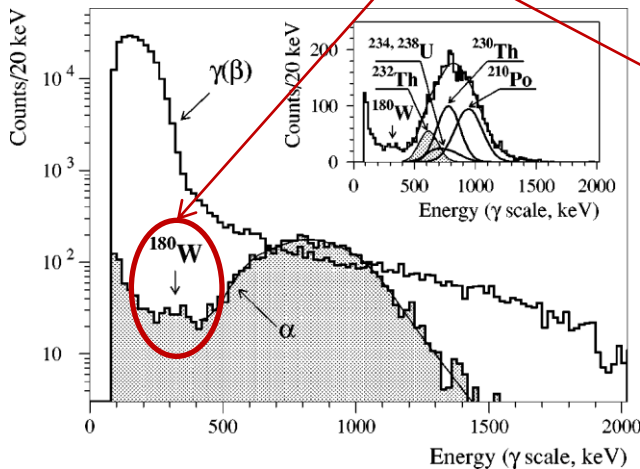




Quenching of scintillation signals from alpha particles (observed energy of  $\alpha$ 's is  $\sim 0.14$  of their real energy):



The effect is observed,  $T_{1/2} = (1.1^{+0.9}_{-0.5}) \times 10^{18} \text{ y}$



- (1) Peak belongs to  $\alpha$  particles (thanks to pulse-shape discrimination)
- (2) Correct energy
- (3)  $T_{1/2}$  in agreement with theoretical expectations

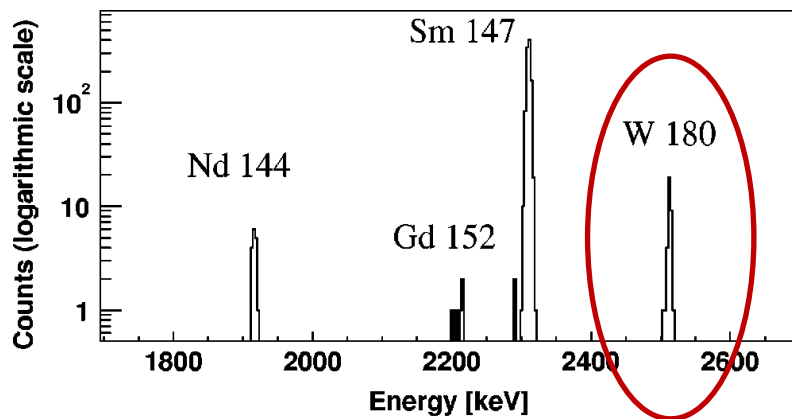
## Confirmation:

C. Cozzini et al., Phys. Rev. C 70(2004)064606

CRESST,  $\text{CaWO}_4$  scintillating bolometer 300 g,  $\sim 15$  mK, FWHM =  $\sim 18$  keV,  $\sim 2300$  h of measurements, low background set-up at LNGS (3600 m w.e. underground)

$$T_{1/2} = (1.8 \pm 0.2) \times 10^{18} \text{ y}$$

$$\text{Measured } Q_\alpha = 2516.4 \pm 1.1(\text{stat}) \pm 1.2(\text{syst}) \text{ keV}$$



## Further observations:

$T_{1/2} =$   
 $(1.0^{+0.7}_{-0.3}) \times 10^{18} \text{ y}$  –  $\text{CaWO}_4$ , Yu.G. Zdesenko et al., NIMA 538(2005)657  
 $(1.3^{+0.6}_{-0.5}) \times 10^{18} \text{ y}$  –  $\text{ZnWO}_4$ , P. Belli et al., NIMA 626-627(2010)31

Now it is routine observation in many rare events' experiments.

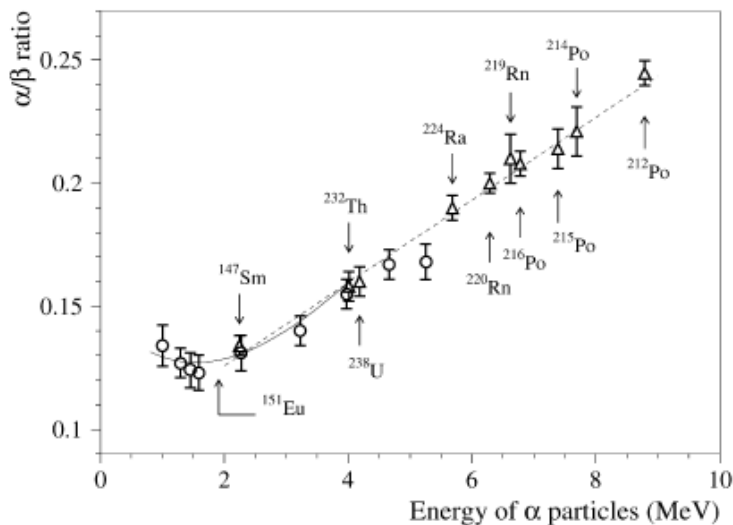
$\alpha$  decay <sup>151</sup>Eu (5/2<sup>+</sup>) → <sup>147</sup>Pm (7/2<sup>+</sup>),  $\delta = 47.81\%$ ,  $Q_\alpha = 1.964$  MeV

Our theoretical estimations with few models:  $T_{1/2} = 3.0 \times 10^{17} - 3.6 \times 10^{18}$  y

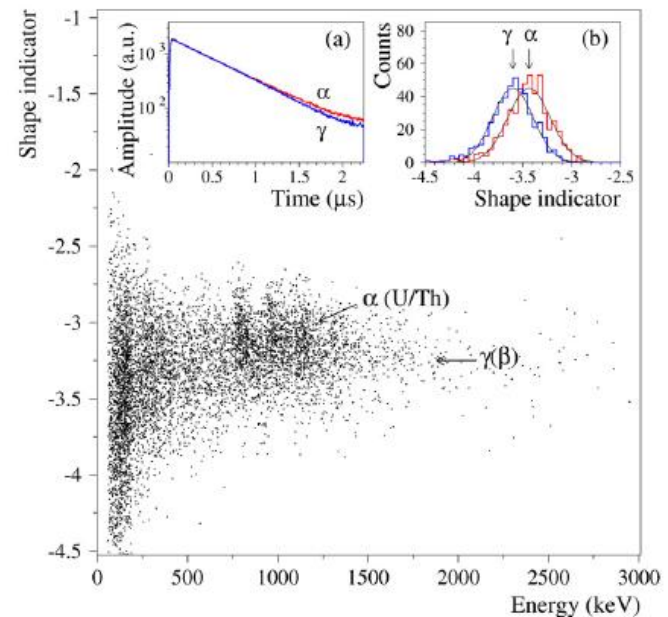
The effect could be observed with CaF<sub>2</sub>(Eu) scintillator with 0.4% Eu.

LNGS (3600 m w.e.), low background set-up, 7426 h, CaF<sub>2</sub>(Eu) 370 g

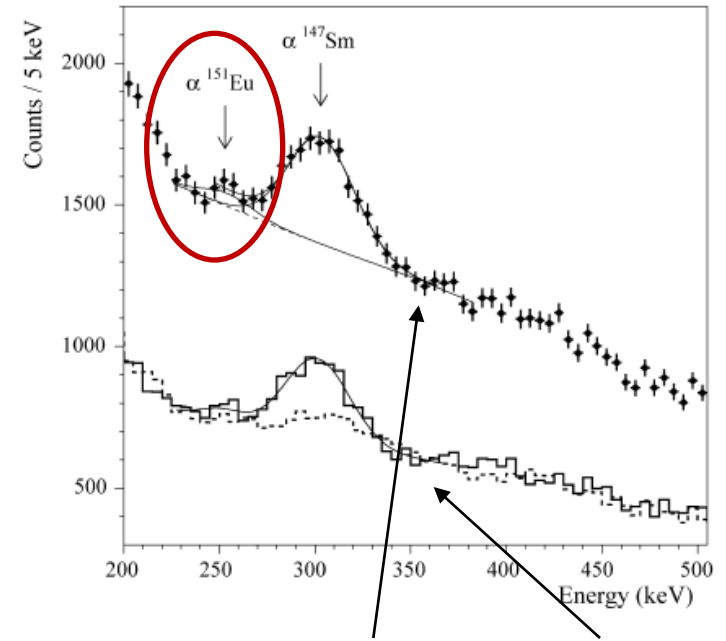
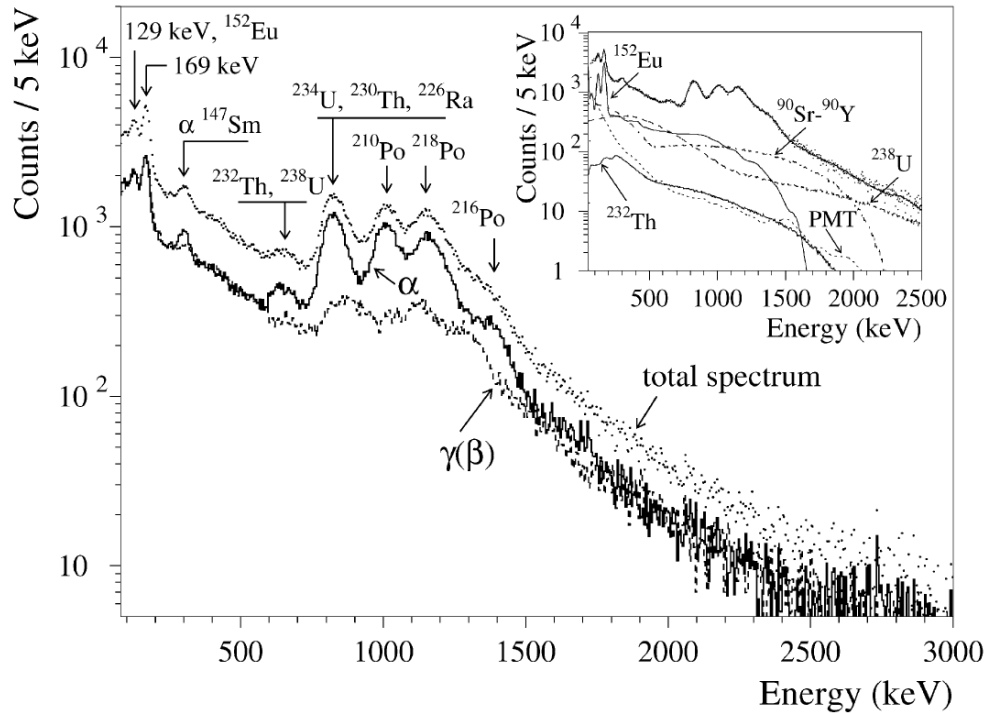
<sup>147</sup>Sm:  $\alpha/\beta$  ratio = 0.13 →  
expected energy for <sup>151</sup>Eu  
~250 keV



Pulse shape discrimination  
between  $\alpha$  and  $\beta/\gamma$  events:



# Experimental spectrum and its fit by simulated distributions:



**Spectra: total and divided on  $\alpha$  and  $\beta$  components**

**Peak's energy:  $255 \pm 7$  keV  $\rightarrow E_{\alpha} = 1.98 \pm 0.04$  MeV (expected  $E_{\alpha} = 1.912$ )**

**Number of  $^{151}\text{Eu}$  nuclei (ICP-MS):  $(2.8 \pm 0.7) \times 10^{21}$ ;  $S = 302 \pm 232$  counts**

$$T_{1/2} = 5^{+11}_{-3} \times 10^{18} \text{ y}$$

Later calculations:  $8.5 \times 10^{18}$  y – O.A.P. Tavares et al., Phys. Scr. 76(2007)C163

$1.3 \times 10^{18}$  y – Y.B. Qian et al., Phys. Rev. C 84(2011)064307

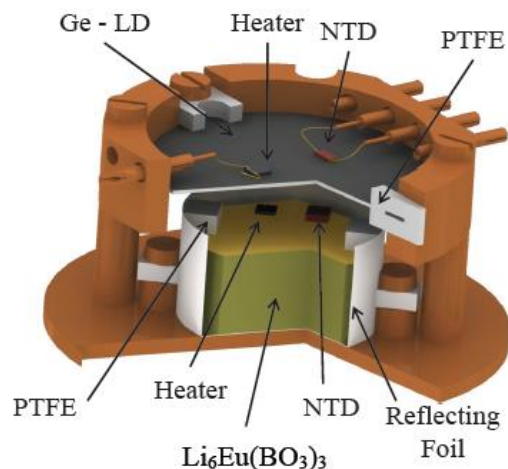
$1.0 \times 10^{19}$  y – Y.B. Qian et al., Phys. Rev. C 85(2012)027306

$8.0 \times 10^{17}$  y – K.P. Santhosh et al., Int. J. Mod. Phys. E 22(2013)1350081

## Confirmation:

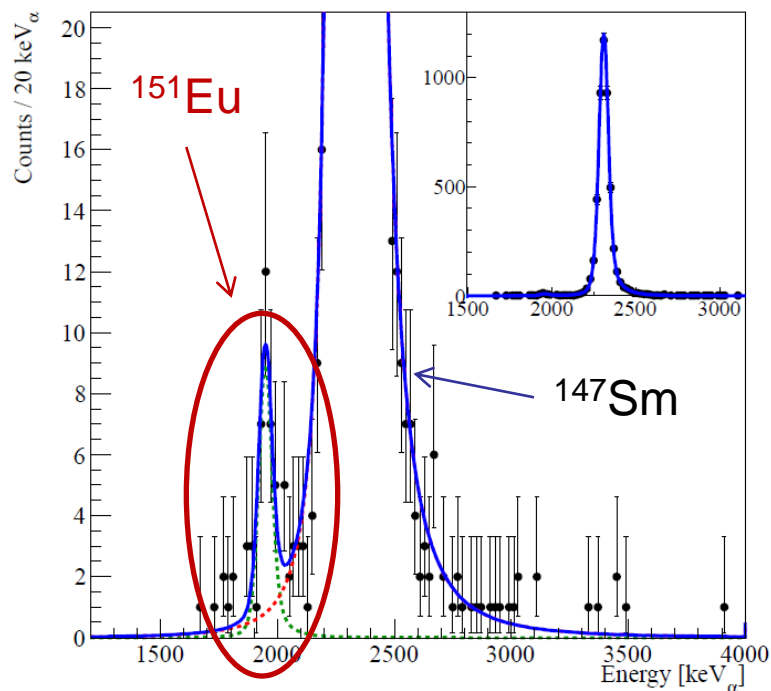
N. Casali et al., J. Phys. G 41(2014)075101

LUCIFER,  $\text{Li}_6\text{Eu}(\text{BO}_3)_3$  scintillating bolometer 6.15 g, FWHM = 65 keV, 462 h of measurements, low background set-up at LNGS (3600 m w.e. underground)

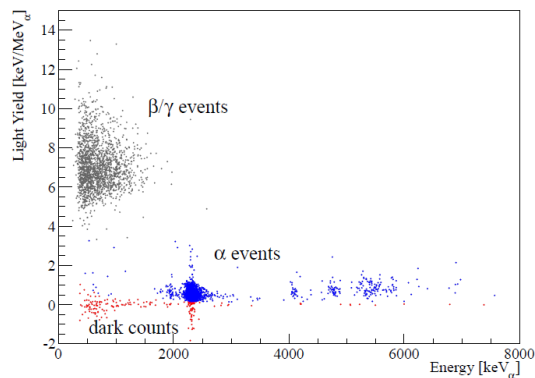


$S = 38 \pm 8$ ,  $T_{1/2} = (4.6 \pm 1.2) \times 10^{18} \text{ y}$   
Measured  $Q_\alpha = 1948.9 \pm 8.6 \text{ keV}$

L. Pattavina, talk at RPSaint'2013 workshop, Kyiv, 17-20.09.2013



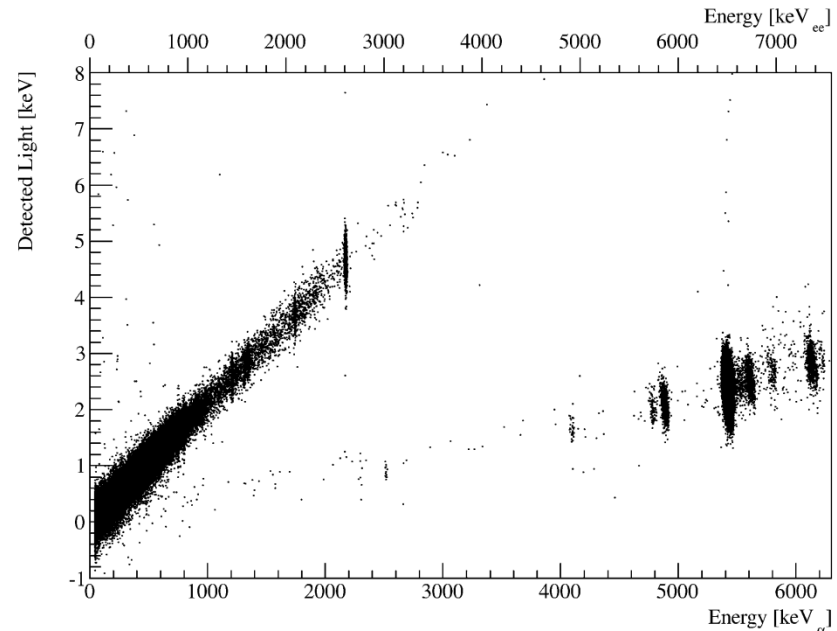
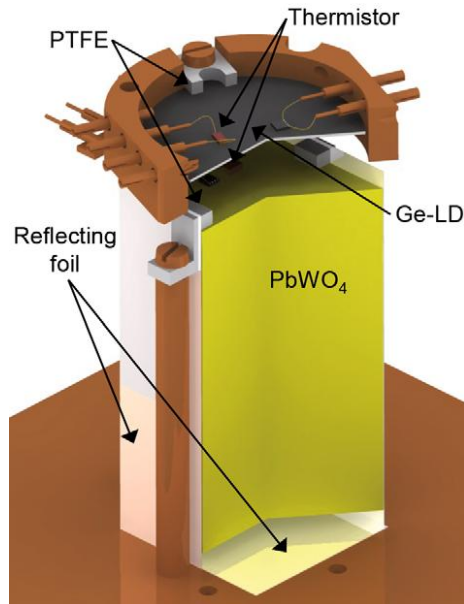
Excellent discrimination of  $\beta/\gamma$  events from  $\alpha$  events



All naturally occurring Pb isotopes are potentially  $\alpha$  decaying,  $Q_\alpha = 0.392 - 1.970$  MeV. Theoretical expectations:  $T_{1/2} = 10^{35} - 10^{189}$  y.

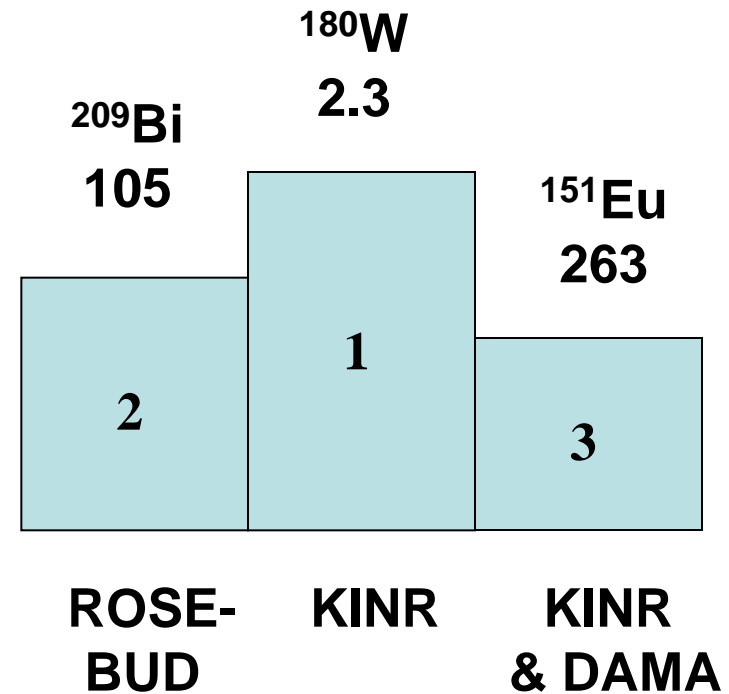
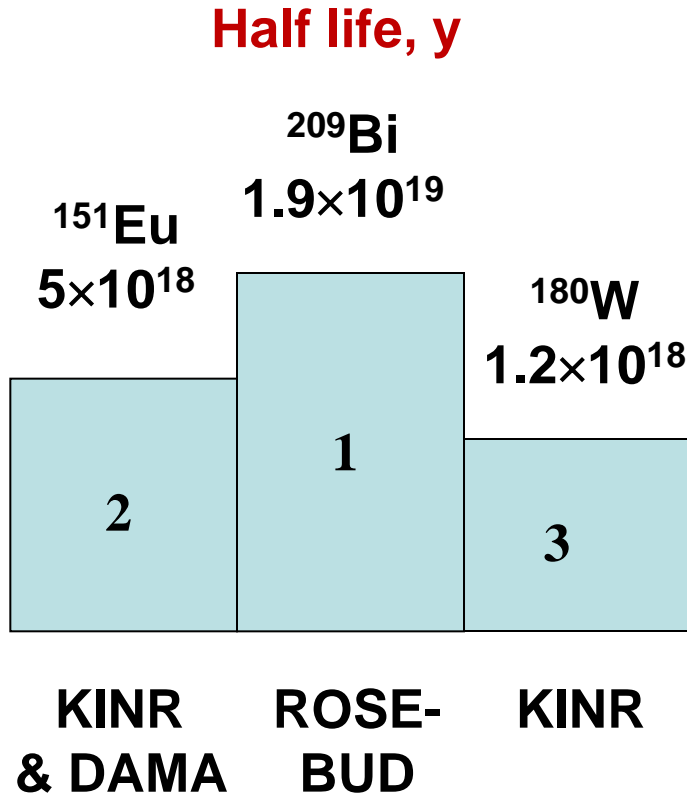
LUCIFER,  $\text{PbWO}_4$  scintillating bolometer (with ancient Roman lead: activity of  $^{210}\text{Pb} < 4$  mBq/kg, while for usual Pb it is  $\sim 10^2 - 10^3$  Bq/kg), 454 g, LNGS (3600 m w.e.), low background set-up, 586 h

Only limits are derived:  $T_{1/2} > 1.4 \times 10^{20} - 2.6 \times 10^{21}$  y  
(for  $^{204}\text{Pb}$  – 3 orders of magnitude better than previous exp. limit)



Observations of rare  
g.s. to g.s.  $\alpha$  decays  
(in sports terminology):

Activity, decays in 1 g  
of element (of natural  
isotopic composition)  
during 1 year



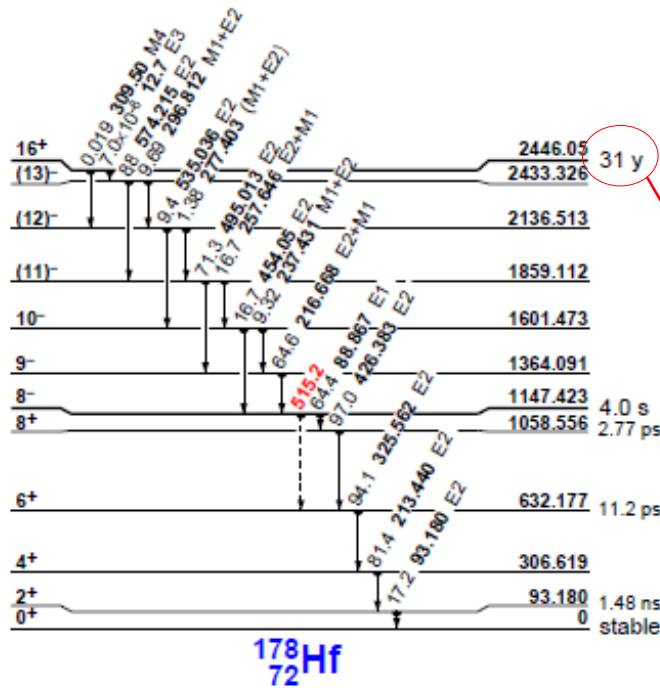
$$\delta(^{209}\text{Bi}) = 100\%$$

$$\delta(^{151}\text{Eu}) = 47.81\%$$

$$\delta(^{180}\text{W}) = 0.12\%$$

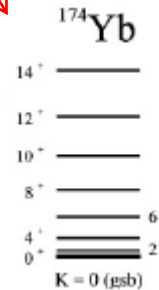


$^{178m2}\text{Hf}$  – extremely interesting nucleus:  $E_{\text{exc}} = 2446 \text{ keV}$  but  $T_{1/2} = 31 \text{ y}$



Usually it decays through IT, but potentially it is  $\alpha$  decaying,  $Q=4526 \text{ keV}$ ; several excited levels of  $^{174}\text{Yb}$  can be populated.

TABLE I. Estimated partial half-lives for  $\alpha$  decay of the  $^{178}\text{Hf}^{m2}$  isomer to levels in the ground-state band of  $^{174}\text{Yb}$ . The calculations are discussed in the text.



Transition $I_i \rightarrow I_f$	$E_\alpha$ [MeV]	$T_{1/2}^{\alpha, f}$ [yr]
$16^+ \rightarrow 0^+$	4.43	$8.6 \times 10^{10}$
$16^+ \rightarrow 2^+$	4.35	$3.0 \times 10^9$
$16^+ \rightarrow 4^+$	4.18	$3.4 \times 10^8$
$16^+ \rightarrow 6^+$	3.91	$1.2 \times 10^8$
$16^+ \rightarrow 8^+$	3.56	$2.8 \times 10^8$
$16^+ \rightarrow 10^+$	3.12	$2.7 \times 10^9$
$16^+ \rightarrow 12^+$	2.61	$7.2 \times 10^{10}$
$16^+ \rightarrow 14^+$	2.03	$5.5 \times 10^{13}$

most probable

First observed in 2007: source with  $3.5 \times 10^{13}$  nuclei of  $^{178m2}\text{Hf}$  ( $^{176}\text{Yb}$  target exposed to 36 MeV He ion beam) deposited on thin Be foil between 2 CR-39 foils,  $\sim 1 \text{ y}$  exposure, observation of  $\alpha$  tracks after CR-39 etching.

Result: 307 ( $\pm 25?$ )  $\alpha$  events in excess,  $T_{1/2}^* = (2.5 \pm 0.5) \times 10^{10} \text{ y}$

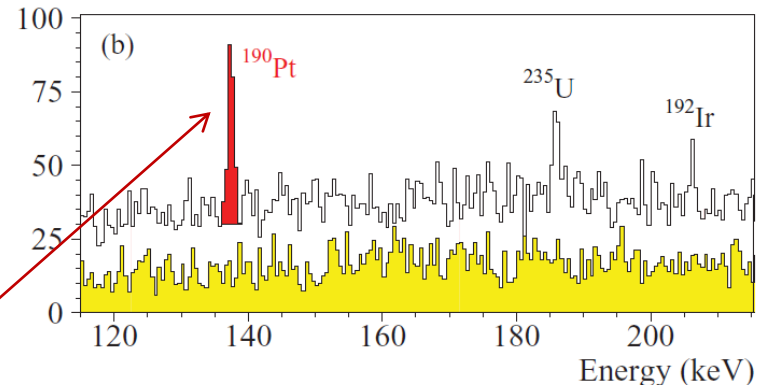
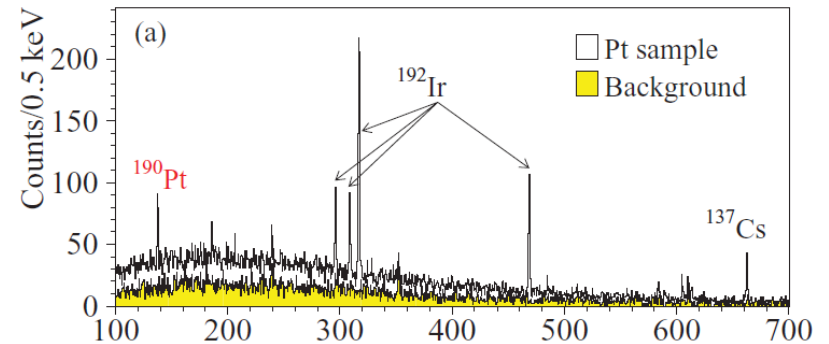
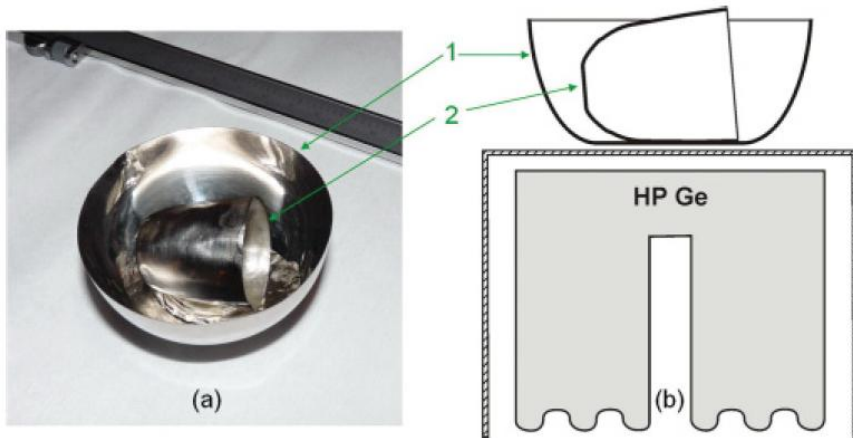


$^{190}\text{Pt} \rightarrow ^{186}\text{Os}^*$  ( $E_{\text{exc}}=137.2 \text{ keV}$ )

P. Belli et al., PRC 83(2011)034603

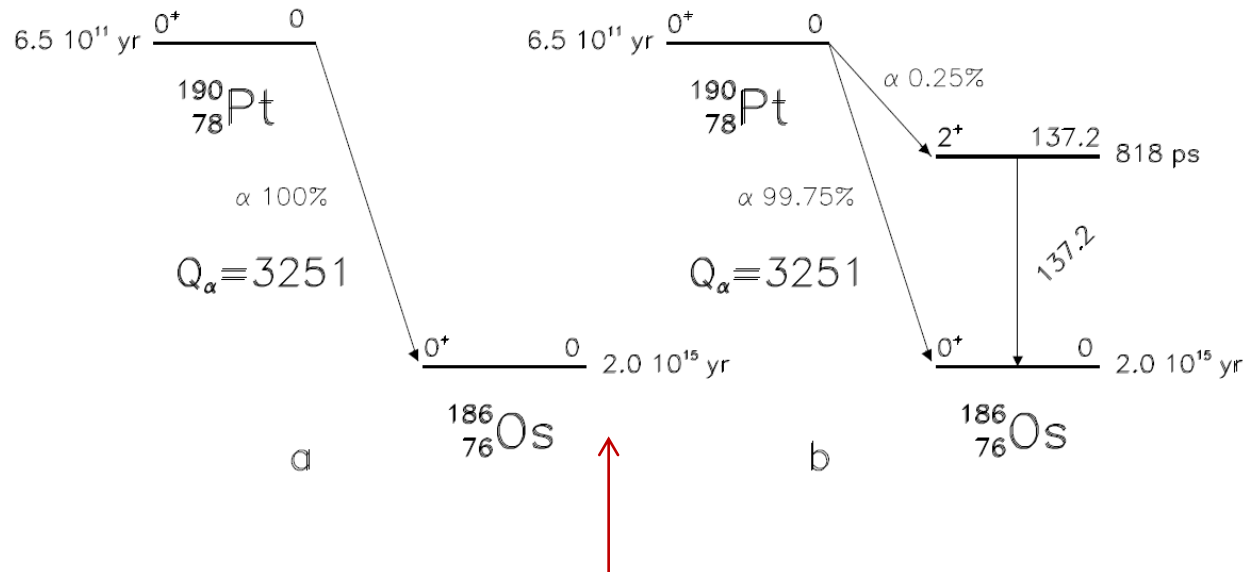
G.s. to g.s. decay  $^{190}\text{Pt} \rightarrow ^{186}\text{Os}$  is known since 1921:  $Q=3251 \text{ keV}$ ,  
 $T_{1/2}=(6.5\pm 0.3)\times 10^{11} \text{ y}$ ;  
Transition to the 1<sup>st</sup> excited level was observed only in 2011

LNGS (3600 m w.e.), HPGe 468 cm<sup>3</sup>, low background set-up, 1815 h,  
42.5 g of natural Pt ( $^{190}\text{Pt}$ :  $\delta=0.014\%$ ; new value (2011)  $\delta=0.012\%$ )



Measured energy =  $137.1\pm 0.1 \text{ keV}$   
Peak =  $132\pm 17$  counts ( $8\sigma$  effect)  
The peak is absent in background

$T_{1/2}^* = 2.6^{+0.4}_{-0.3}(\text{stat})\pm 0.6(\text{syst})\times 10^{14} \text{ y}$



## Old and new schemes of $^{190}\text{Pt}$ decay

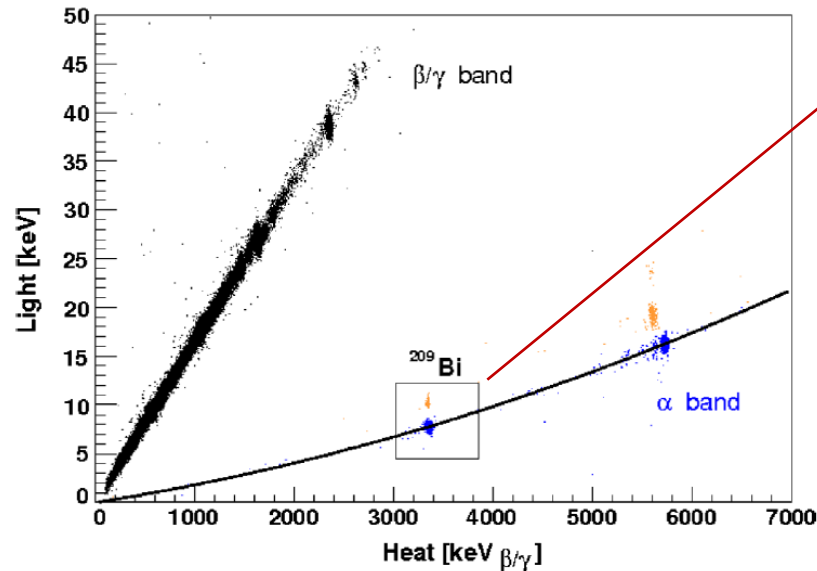
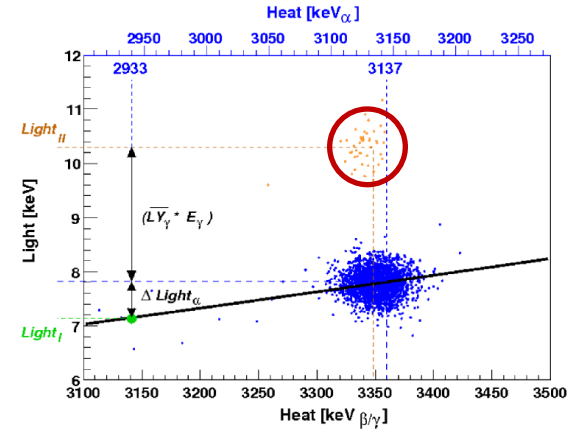
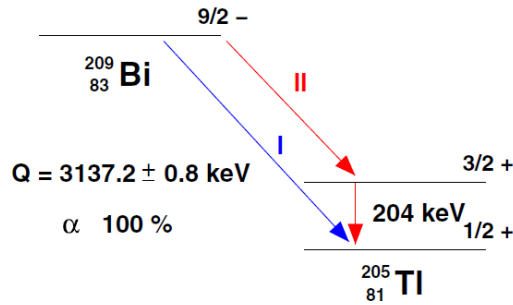
$T_{1/2}$  limits for other Pt isotopes were also set at the level of  $10^{16} - 10^{20}$  y

**It would be nice to remeasure with different detector and Pt sample**

$^{209}\text{Bi} \rightarrow ^{205}\text{Tl}^*$  ( $E_{\text{exc}}=204 \text{ keV}$ ) J.W. Beeman et al., PRL 108(2012)062501

G.s. to g.s. decay  $^{209}\text{Bi} \rightarrow ^{205}\text{Tl}$  – 2003:  $Q=3137 \text{ keV}$ ,  $T_{1/2}=(1.9\pm 0.2)\text{e}18 \text{ y}$   
 Transition to the 1<sup>st</sup> excited level was observed in 2012

LNGS (3600 m w.e.),  $\text{Bi}_4\text{Ge}_3\text{O}_{12}$  bolometer 889 g, few tens mK, 375 h  
**Heat and light signals – discrimination of  $\alpha$  and  $\beta/\gamma$  events by ratio of light/heat**



→ g.s.:  $\alpha + \text{recoil}$  (3137 keV)  
 → 204 keV:  $\alpha + \text{recoil}$  (2933 keV) +  $\gamma$  (204 keV)  
 The same heat signals, but different light signals because scintillation from  $\alpha$  is quenched, and from  $\gamma$  – not quenched  
 $T_{1/2} = (2.04\pm 0.08)\times 10^{19} \text{ y}$   
 $T_{1/2}^* = (1.4\pm 0.2)\times 10^{21} \text{ y}$

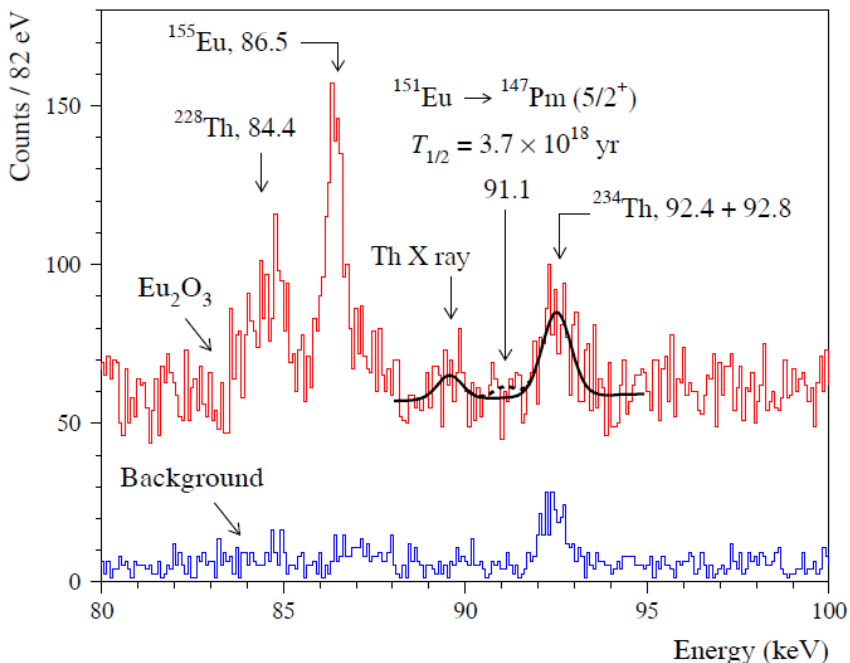
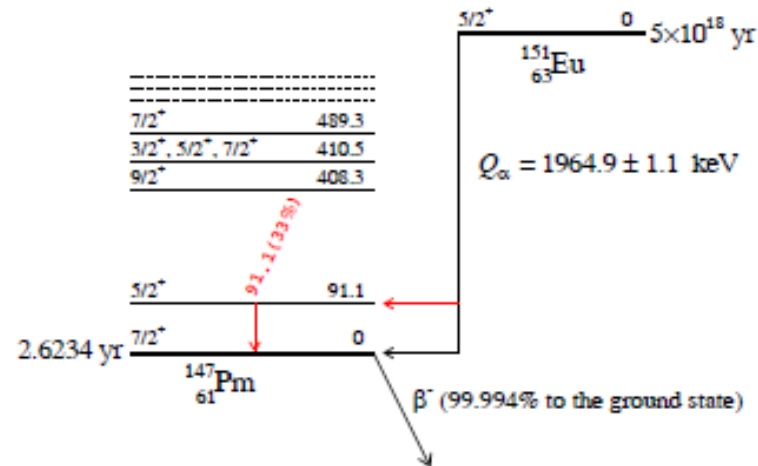
$^{151}\text{Eu} \rightarrow ^{147}\text{Pm}^*$  ( $E_{\text{exc}}=91 \text{ keV}$ )

F.A. Danevich et al., EPJA 48(2012)157

G.s. to g.s. decay:  $T_{1/2} \sim 5 \times 10^{18} \text{ y}$

Decays to excited levels are also possible, the most probable to 1<sup>st</sup> level,  $E_{\text{exc}}=91 \text{ keV}$

HADES (500 m w.e.), high purity  $\text{Eu}_2\text{O}_3$   
303 g, 2233 h in low-background set-up with HPGe 40 cm<sup>3</sup>



The effect is absent, only limit:

$T_{1/2} > 3.7 \times 10^{18} \text{ y}$

~1 order of magnitude better than previous exp. limits

Not far from theor. estimates  $10^{19} - 10^{20} \text{ y}$

### 3. Investigations of rare $\beta$ decays

$^{48}\text{Ca}$   $^{50}\text{V}$   $^{96}\text{Zr}$   $^{113}\text{Cd}$   $^{115}\text{In}^*$   $^{123}\text{Te}$   $^{180\text{m}}\text{Ta}$   $^{222}\text{Rn}$

**$^{48}\text{Ca}$**

$Q_{\beta}=282 \text{ keV}$

Could be populated:

ground state  $\Delta J^{\Delta\pi}=6^+$

level 131 keV  $\Delta J^{\Delta\pi}=5^+$

level 252 keV  $\Delta J^{\Delta\pi}=4^+$

$T_{1/2}$  - theoretical estimates and experimental limits (y)

( $T_{1/2}$  decreases for bigger Q as  $\sim 1/Q^5$ , but increases for bigger  $\Delta J^{\Delta\pi}$ ):

	Theory [1]	Theory [2]	Experiment [3]
$6^+(\text{g.s.})$	$=4.0\text{e}25$	$=1.5\text{e}29\text{-}1.3\text{e}31$	$>1.6\text{e}20$
$5^+(131)$	$=4.0\text{e}22$	$= (1.1^{+0.8}_{-0.6})\text{e}21$	$>2.5\text{e}20$
$4^+(252)$	$=3.0\text{e}23$	$=8.8\text{e}23\text{-}5.2\text{e}26$	$>1.9\text{e}20$

[1] R.K. Bardin et al., NPA 158 (1970) 337

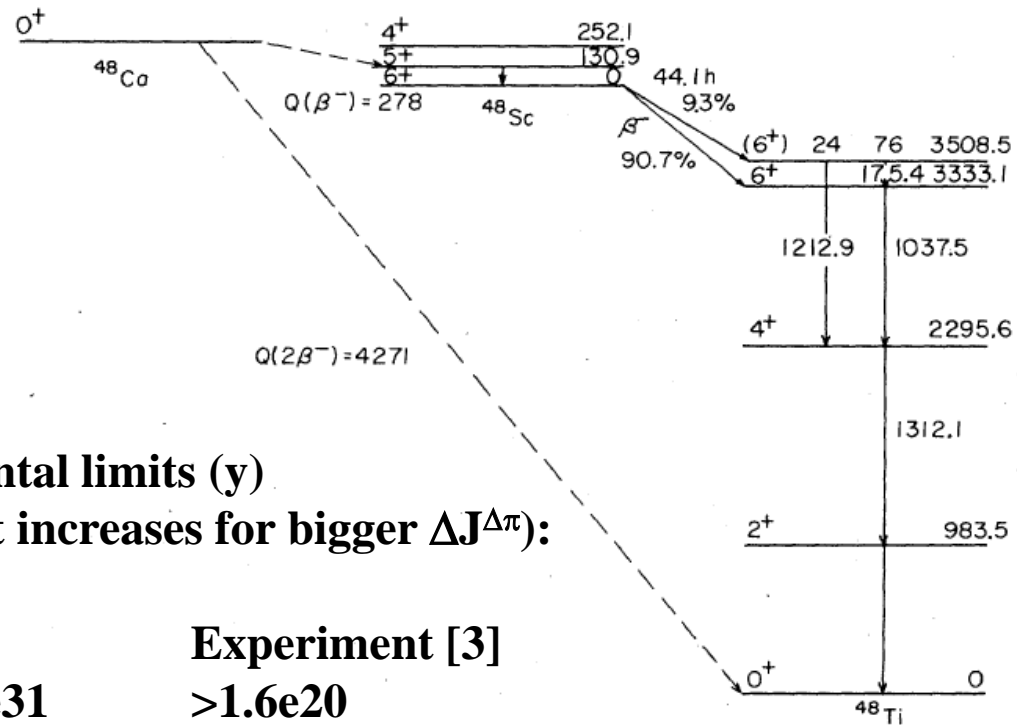
[2] M. Aunola et al., Europhys. Lett. 46 (1999) 577

[3] A. Bakalyarov et al., JETP Lett. 76 (2002) 545

(search for deexcitation  $\gamma$ 's of  $^{48}\text{Sc}$ ,  $^{48}\text{Ti}$  with Ge detector; however  $\delta(^{48}\text{Ca})=0.187\%$ )

At the same time,  $^{48}\text{Ca}$  can decay also through  $2\beta$  decay to  $^{48}\text{Ti}$  (2<sup>nd</sup> order process); already observed in few experiments:  $T_{1/2}(2\beta 2\nu, \text{g.s.}) = 4.3\text{e}19 \text{ y}$ .

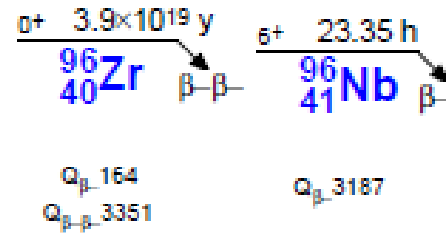
Thus **single  $\beta$  decay occurs even with lower probability than  $2\beta$  decay** - due to big  $\Delta J$



**the most probable**

**$^{96}\text{Zr}$**

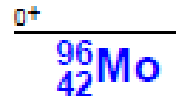
$Q_{\beta}=161 \text{ keV}$



**Could be populated:**

- ground state  $\Delta J^{\Delta\pi}=6^+$
- level 44 keV  $\Delta J^{\Delta\pi}=5^+$
- level 146 keV  $\Delta J^{\Delta\pi}=4^+$

**$T_{1/2}$  - theoretical estimates and experimental limits (y):**



	Theory [1]	Experiment [2]
$6^+(\text{g.s.})$	$=1.2\text{e}29$	$>3.8\text{e}19$
$5^+(44)$	$=2.4\text{e}20$	$>3.8\text{e}19$ <b>the most probable</b>
$4^+(146)$	$=4.9\text{e}22$	$>3.8\text{e}19$

[1] H. Heiskanen et al., J. Phys. G 34 (2007) 837

[2] M. Arpesella et al., Europhys. Lett. 27 (1994) 29

(search for deexcitation  $\gamma$ 's of  $^{96}\text{Mo}$  with Ge detector;  $\delta(^{96}\text{Zr})=2.80\%$  - **much higher than that for  $^{48}\text{Ca}$ ; worth to remeasure with higher sensitivity?**)

$2\beta$  decay of  $^{96}\text{Zr}$  to  $^{96}\text{Mo}$ :  $T_{1/2}(2\beta 2\nu, \text{g.s.})=(2.3\pm 0.4)\text{e}19 \text{ y}$  (NEMO-3'2008).

Geochemical  $2\beta T_{1/2}$ :  $=(3.9\pm 0.9)\text{e}19$  Kawashima'1993 and  $=(0.9\pm 0.3)\text{e}19$  Wieser'2001.

**Contribution of single  $\beta$  decay to geochemical  $T_{1/2}$ ?**

# $^{180m}\text{Ta}$

Extremely interesting case:

g.s. state quickly decays ( $T_{1/2} \sim 8 \text{ h}$ );

isomeric state ( $E_{\text{exc}} = 77 \text{ keV}$ ) has very big  $T_{1/2} > 2 \times 10^{16} \text{ y}$

$\delta(^{180m}\text{Ta}) = 0.012\%$

EC  $\Delta J^{\Delta\pi} = 3^-$

$\beta^-$   $\Delta J^{\Delta\pi} = 3^-$

Last experimental search:

M. Hult et al., Appl. Rad. Isot. 67 (2009) 918

1500 g of natural Ta, sandwich HP Ge,

underground HADES laboratory (500 m w.e.), 68 d

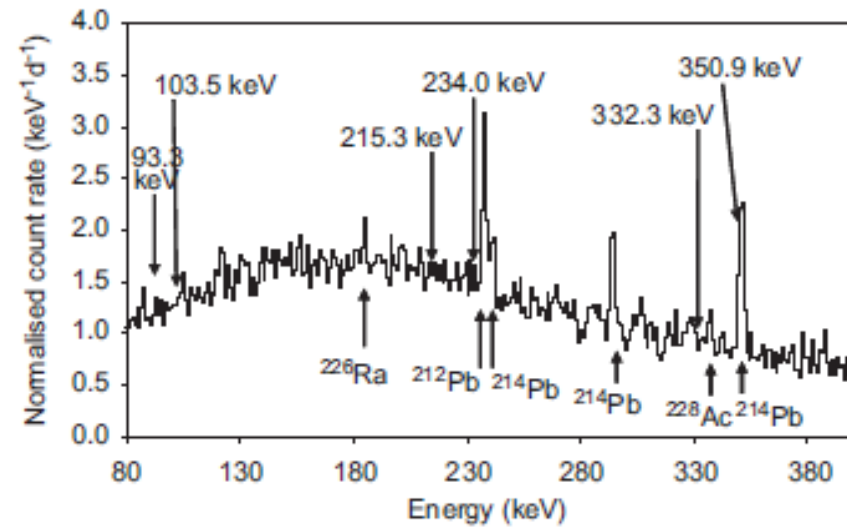
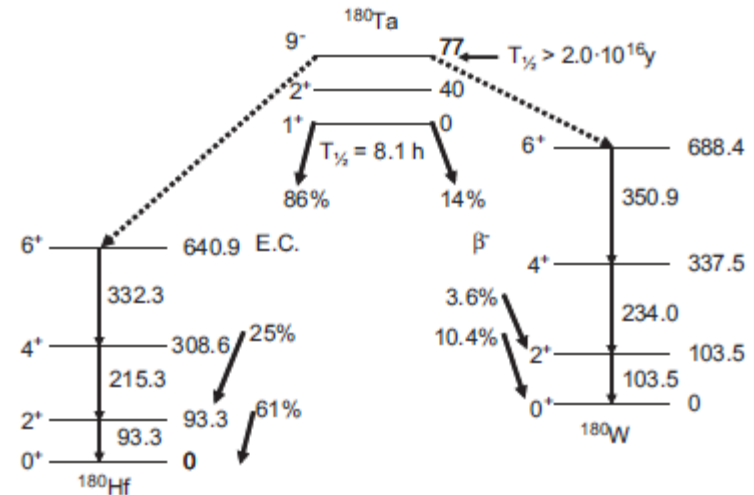
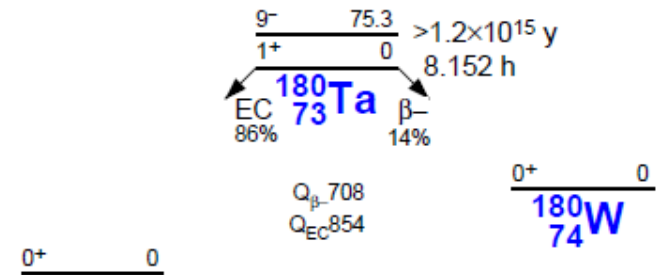
$T_{1/2}(\text{EC}) > 4.5 \times 10^{16} \text{ y}$

$T_{1/2}(\beta^-) > 3.7 \times 10^{16} \text{ y}$

Theoretical  $T_{1/2}$  estimations:

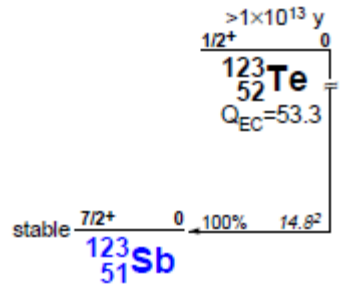
IT  $> 1 \times 10^{27} \text{ y}$  E.B. Norman, PRC 24(1981)2334

EC,  $\beta^-$  calculations are absent





$^{123}\text{Te}$   $\delta(^{123}\text{Te})=0.89\%$



**Many puzzling experimental situations (only K EC was searched for):**

1. D.N. Watt et al., Philos. Mag. 7 (1962) 105

Detection of Sb X rays  $E_X=26.1$  keV after EC with prop. counter,  $T_{1/2}=(1.24 \pm 0.10) \times 10^{13}$  y

This result was present in all nuclear tables many years

2. A. Alessandrello et al., PRL 77 (1996) 3319

Four 340 g  $\text{TeO}_2$  bolometers, underground measurements (LNGS, 3600 m w.e.), 1548 h

Peak at total energy release of 30.5 keV ( $E_K$  of Sb) is observed,

$T_{1/2}^{\text{K}}=(2.4 \pm 0.9) \times 10^{19}$  y - 6 orders of magnitude higher!

*Result of Watt'1962 was explained by excitation of Te atoms by cosmic rays and nat.*

*radioactivity that gives  $E_X=27.3$  keV, and by not enough good resolution of prop. counter*

3. A. Alessandrello et al., PRC 67 (2003) 014323

Twenty 340 g  $\text{TeO}_2$  bolometers, LNGS (3600 m w.e.), peak at 30.5 keV is not present,

$T_{1/2}^{\text{K}} > 5.0 \times 10^{19}$  y !

However, this peak appeared once more after all crystals were dismantled for surface cleaning at the sea level for ~2 months period and reinstalled underground.

*Explanation of Alessandrello'1996: peak at 30.5 keV is due to EC of  $^{121}\text{Te}$  ( $Q=1036$  keV,*

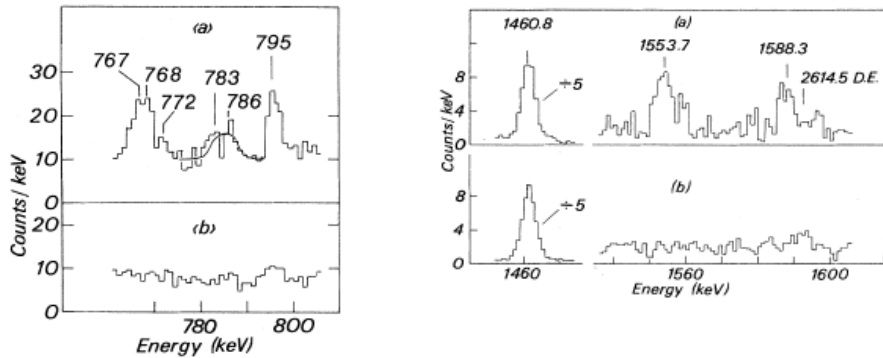
$T_{1/2}=16.78$  d);  $^{121}\text{Te}$  is produced by neutron capture on  $^{120}\text{Te}$  ( $\delta=0.09\%$ ) !

**$^{50}\text{V}$**   $\delta=0.250\%$

One of **only 3 nuclei** where  $\beta$  processes with  $\Delta J^{\Delta\pi}=4^+$  were observed (other two are  $^{113}\text{Cd}$  and  $^{115}\text{In}$ )

Low natural abundance ( $\delta=0.250\%$ ), big  $T_{1/2}$  (difficult to study)

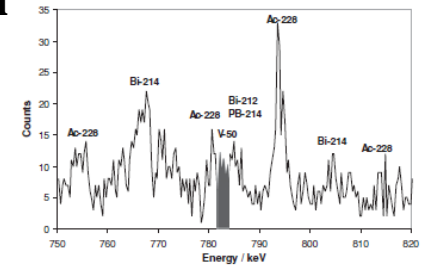
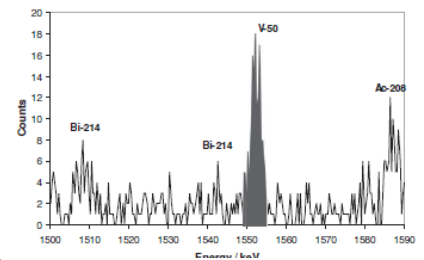
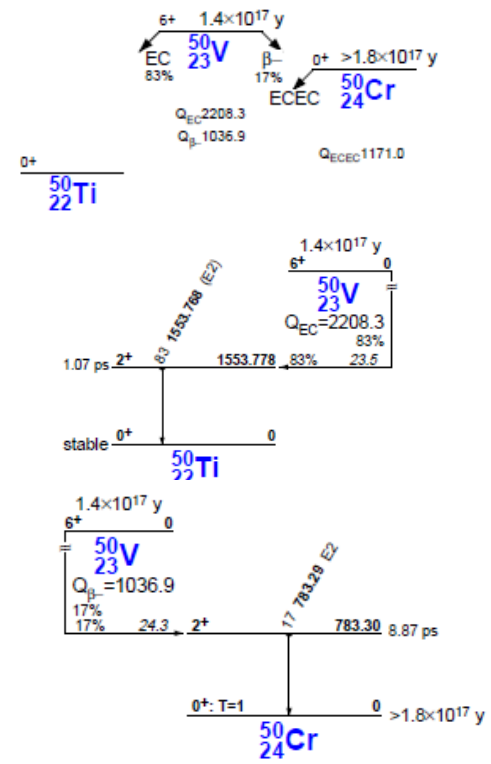
**Experiment 1989:** J.J. Simpson et al., PRC 39 (1989) 2367  
 3 Ge detectors, 337.5 g of natural V, salt mine, 1109 h  
 Search for  $\gamma$ 's of 1554 keV (EC) and 783 keV ( $\beta^-$  decay)



**Experiment 2011:** H. Dombrowski et al., PRC 83 (2011) 054322  
 Ge detector, 255.8 g of natural V, Asse salt mine (1200 m w.e.), 2347 h  
 Peak 783 keV is not observed:

$T_{1/2}(\text{EC})=(2.3\pm 0.3)e17$  y,  $T_{1/2}(\beta^-) > 1.7e18$  y

Only  $\gamma$ 's are detected;  $T_{1/2}$  is measured but not shape of  $\beta$  spectrum





Experimental spectrum is excellently described as 3 FU ( $\Delta J^{\Delta\pi} = 4^-$ ):

$$C(E) = P^6 + c_1 P^4 Q^2 + c_2 P^2 Q^4 + c_3 Q^6 \quad \text{with } c_1 = 7.112, c_2 = 10.493, c_3 = 3.034$$

(small puzzle ...)

Recent theoretical description as 4 FNU:

M.T. Mustonen et al., PRC 73 (2006) 054301 + PRC 76 (2007) 019901(E)

M.T. Mustonen et al., PLB 657 (2007) 38

(shape different from the experimental one)

Last experimental work:

J.V. Dawson et al., NPA 818 (2009) 264

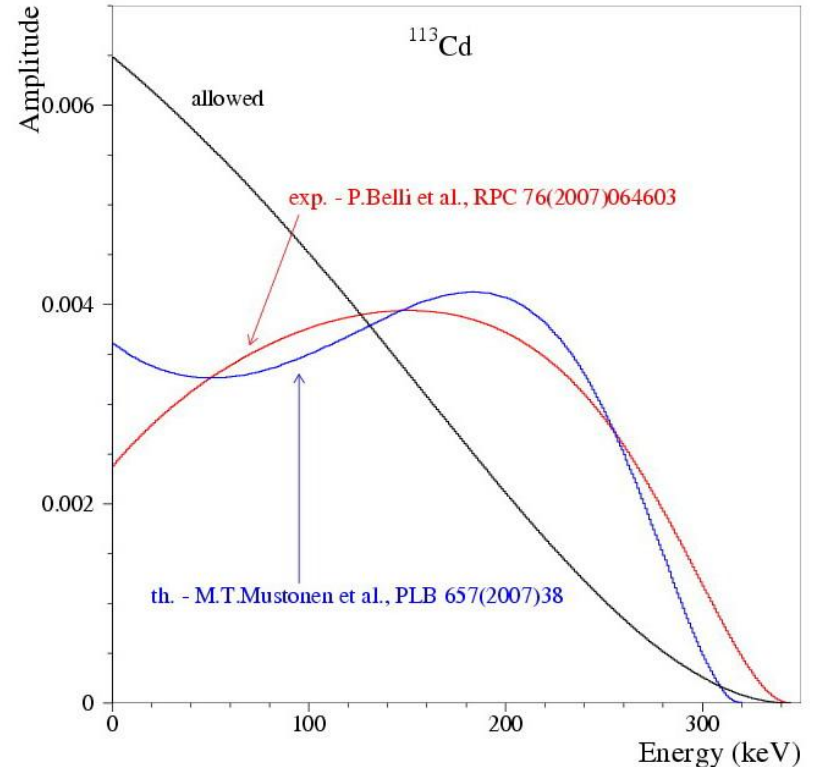
16 CdZnTe detectors, LNGS, 6.58 kg×d

Confirmed  $T_{1/2}$  and shape of spectrum,

but gave different  $Q_\beta$  value

(322 keV instead of 345 keV in Belli'2007)

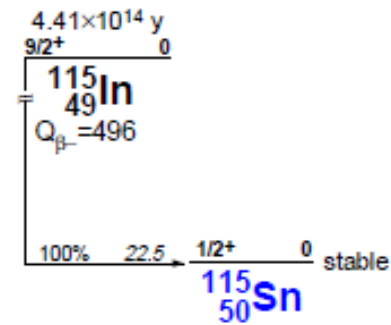
(another small puzzle ...)



**$^{115}\text{In}$**   $\delta=95.71\%$

$9/2^+ \rightarrow 1/2^+$   $\Delta J^{\Delta\pi} = 4^+$  classified as 4 FNU

On contrary to  $^{113}\text{Cd}$ , spectrum shape was measured only in one work:  
L. Pfeiffer et al., PRC 19 (1979) 1035



Liquid scintillator loaded by In at 51.2 g/l, measurements at the sea level

**What could be improved:**

- (1) Background, in particular n capture by  $^{115}\text{In}$  (and  $^{116}\text{In}$  is  $\beta^-$  unstable,  $Q=3275$  keV)
- (2) Strong quenching of low-energy electrons in liquid scintillator (was not discussed)
- (3) Response function (resolution) “is not known and is not readily measurable”
- (4) Q value was obtained as 492.7(13.6) keV and 470.6(5.2) keV; today value is 499(4) keV
- (5)  $T_{1/2}=(4.41\pm 0.24)e14$  y (since 1979 – in all tables), but in some disagreement with previous results (f.e. G.B. Beard et al., PR 122 (1961) 1576:  $T_{1/2}=(6.9\pm 1.5)e14$  y)
- (6) Energy threshold – around 50 keV
- (7) Shape is described as polynomial in E

**Remeasuring in low background conditions would be very interesting!**

Recent theoretical description as 4 FNU:

M.T. Mustonen et al., PRC 73 (2006) 054301 + PRC 76 (2007) 019901(E)

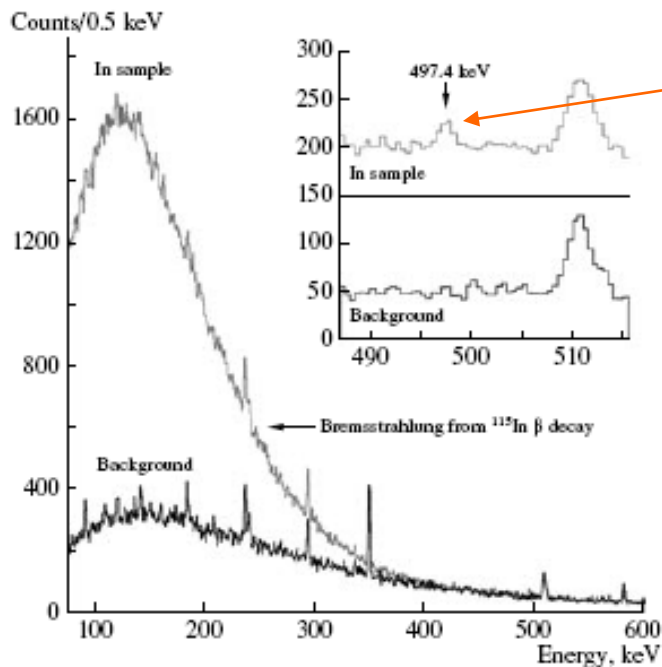
M.T. Mustonen et al., PLB 657 (2007) 38

$^{115}\text{In} \rightarrow ^{115}\text{Sn}^*$

First observation of  $\beta$  decay of  $^{115}\text{In}$  to the first excited level ( $E_{\text{exc}}=497.4$  keV) of  $^{115}\text{Sn}$ :

C.M. Cattadori et al., NPA 748 (2005) 333 + Phys. At. Nucl. 70 (2007) 127

LNGS, ~1 kg In, 4 HP Ge detectors 225 cm<sup>3</sup> each, 2762 h In + 1601 h background



De-excitation  $\gamma$ 's give peak at 497.4 keV (observation with  $4\sigma$ 's),  $(1.18 \pm 0.31) \times 10^{-6}$  yield,  $T_{1/2} = (3.7 \pm 1.0) \times 10^{20}$  y

**Situation in 2005:**

Implications for neutrino mass:

$\Delta M_a = 499 \pm 4$  keV (Audi et al., 2003)

$Q_\beta^* = \Delta M_a - E_{\text{exc}} = 1.6 \pm 4$  keV

our calculation:  $Q_\beta^* = 460$  eV

(possibly the lowest known measured  $Q_\beta$  value)

Evidently:  $m_\nu < Q_\beta$

Could be  $Q_\beta \sim 1$  eV?

Need to re-measure  $\Delta M_a$  ( $^{115}\text{In}$ - $^{115}\text{Sn}$ ,  $\delta=4$  keV) and  $E_{\text{exc}}$  ( $\delta=22$  eV) with greater accuracy

E.G. Myers, Florida St. Un.  
 $M_a$  with  $\delta \sim 10$  eV for  $A=100$ <sup>30</sup>

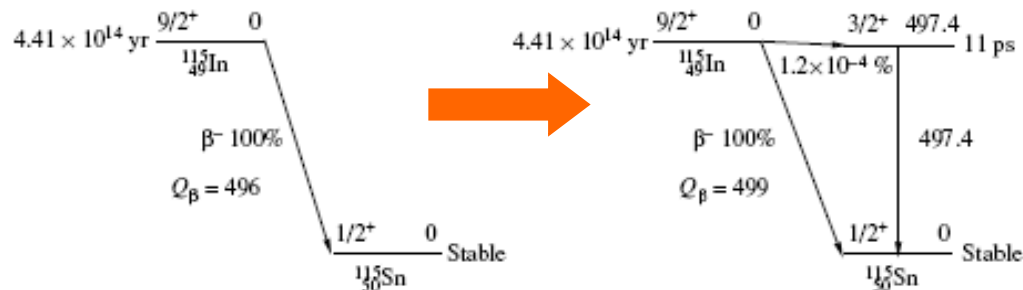


Fig. 2. Old (a) and new (b) schemes of  $^{115}\text{In} \rightarrow ^{115}\text{Sn}$   $\beta$  decay (energy in keV).

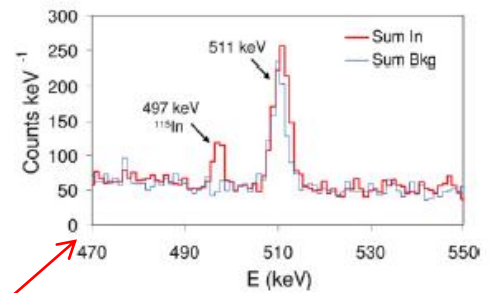
## Subsequent events:

### 1. Confirmation of observation of $^{115}\text{In} \rightarrow ^{115}\text{Sn}^*$ decay

HADES underground laboratory (500 m w.e.), 2566 g of In, 3 Ge:

$T_{1/2} = (4.1 \pm 0.6) \times 10^{20}$  y (E. Wieslander et al., PRL 103(2009)122501)

$T_{1/2} = (4.3 \pm 0.5) \times 10^{20}$  y (E. Andreotti et al., PRC 84(2011)044605)

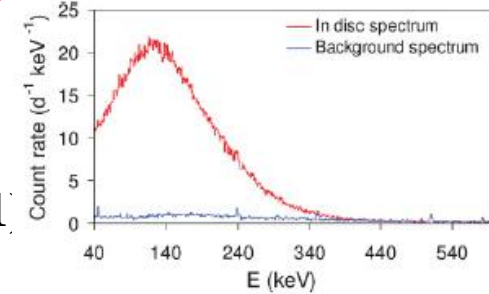


### 2. New measurements of difference $\Delta$ of $^{115}\text{In} - ^{115}\text{Sn}$ masses

$\Delta = 497.680 \pm 0.170$  keV (E. Wieslander et al., PRL 103(2009)122501)

$\Delta = 497.489 \pm 0.010$  keV (B.J. Mount et al., PRL 103(2009)122502)

Thus,  $Q^*$  value is:  $Q^* = \Delta - E_{\text{exc}} = (497.334 \pm 0.022) - (497.489 \pm 0.010) = 155 \pm 24$  eV



Really the lowest Q value of a known  $\beta$  decay ( $^{163}\text{Ho} - 2.555$  keV,  $^{187}\text{Re} - 2.469$  keV) (and highest (partial)  $T_{1/2}$ )

Paradoxical situation: masses of the nuclei ( $\sim 100$  GeV) are known with precision 10 eV while  $E_{\text{exc}}$  ( $\sim 500$  keV) – with precision 22 eV (needs to be remeasured)

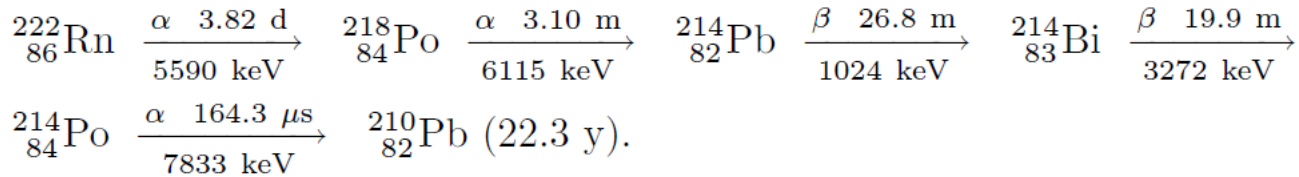
3. Influence of different chemical environment on  $T_{1/2}$  (In,  $\text{InCl}_3$ , etc.). If to use dependence  $T_{1/2} \sim 1/Q^5$  and change Q on 1 eV only, we will obtain  $(155/154)^5 = 1.03 - 3\%$  change in  $T_{1/2}$ . Difficult but maybe possible to see (current accuracy – 12%).

4. Deviations from theoretical spectrum due to non-zero  $\nu$  mass? Theoretical spectrum ( $\Delta J^{\Delta\pi} = 3^+ -$  classified as 2 FU) was calculated in R. Dvornicky, F. Simkovic, AIP Conf. Proc. 1417(2011)33. Very difficult experimentally.

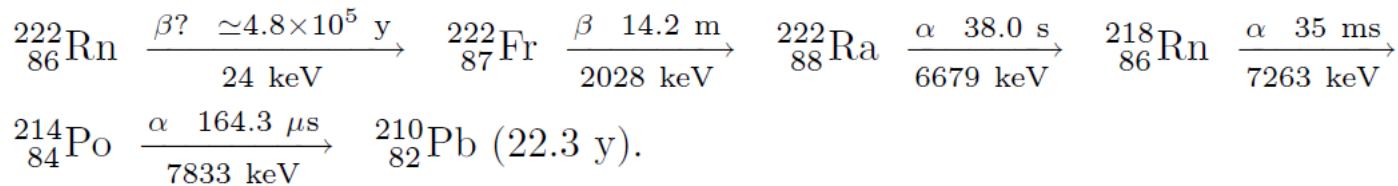
**BaF<sub>2</sub> scintillator, 1.714 kg, LNGS (3600 m w.e.), 101 h.**

**High contamination by  $^{226}\text{Ra}$  – 7.8 Bq/kg.**

**In all nuclear tables,  $^{222}\text{Rn}$  (in chain of  $^{238}\text{U}$ ) is 100%  $\alpha$  decaying. Usual chain:**



**However,  $\beta$  decay of  $^{222}\text{Rn}$  also is energetically allowed with  $Q=24\pm 21$  keV. In this case:**



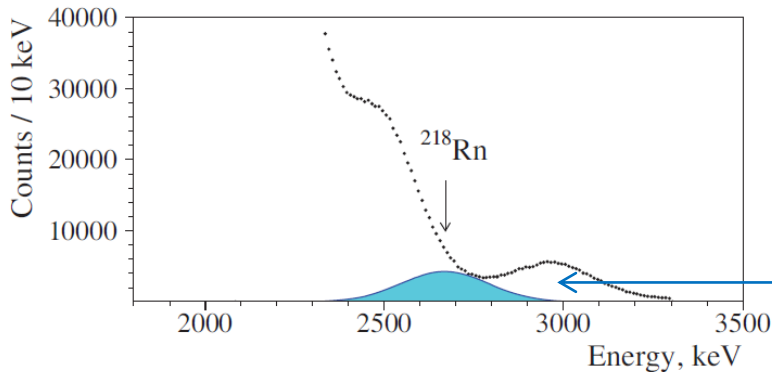
**$^{222}\text{Rn}(0^+) \rightarrow ^{222}\text{Fr}(2^-)$ ,  $\Delta J^{\Delta\pi}=2^-$ ;  $T_{1/2}$  can be estimated using average (for 216 known 1 FU  $\beta$  decays)  $\log ft = 9.5$  and LOGFT tool at NNDC as  $T_{1/2} = 4.8 \times 10^5 \text{ y}$  (for  $Q=24$  keV;  $6.7 \times 10^4 \text{ y}$  for  $Q=45$  keV and  $2.4 \times 10^8 \text{ y}$  for  $Q=3$  keV).**



Expected E and  $\Delta t$  are known, and it is possible to distinguish between  $\alpha$  and  $\beta$  events in  $\text{BaF}_2$  scintillator because of difference in their time shapes.

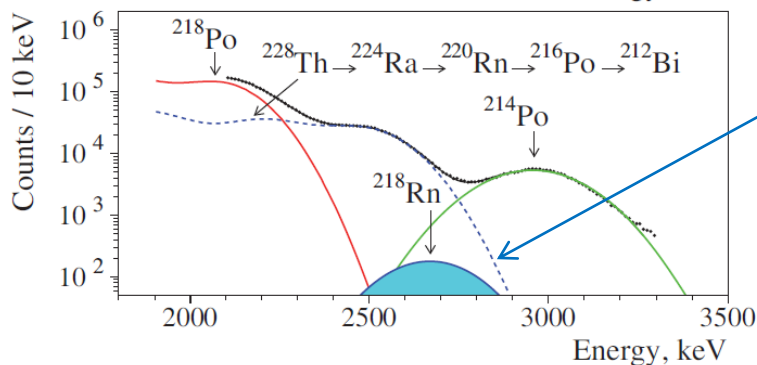
The following sequence of events was searched for ( $^{222}\text{Fr} \rightarrow ^{222}\text{Ra} \rightarrow ^{218}\text{Rn} \rightarrow ^{214}\text{Po}$ ):

- (1) event at 30 – 2207 keV ( $^{222}\text{Fr} Q_\beta + \text{FWHM}_\beta$ ) and with  $\beta$  time shape;
- (2) next event at 2109 – 2623 keV ( $^{222}\text{Ra} E_\alpha + \text{FWHM}_\alpha$  in  $\gamma$  scale), with  $\alpha$  time shape and in time interval [1.65 ms, 1.65 ms +  $5 \times 38.0$  s];
- (3) last event at 2398 – 2946 keV ( $^{218}\text{Rn} E_\alpha + \text{FWHM}_\alpha$  in  $\gamma$  scale), with  $\alpha$  time shape and in time interval [1.65 ms, 1.65 ms +  $5 \times 35$  ms].



$7.0 \times 10^5$  selected potential  $^{218}\text{Rn}$  events.

Maximal effect consistent with exp. data,  $T_{1/2}^\beta > 122$  d (too conservative limit)



Limit from fit by model (known  $\alpha$  peaks from contamination),  $T_{1/2}^\beta > 8.0$  y.

## 4. Observation of emission of $e^+e^-$ pairs in $\alpha$ decay of $^{241}\text{Am}$

$\beta$  decay - internal bremsstrahlung (IB) and internal pair production (IPP) are known effects  
 $\alpha$  decay - IB is known; **what about IPP?**

**In fact, it was observed previously in 3 experiments (1973, 1986, 1990):**

Source	Experiment				Theory		
	$\lambda (\times 10^{-9})$	Detectors	Year	Ref.	$\lambda (\times 10^{-9})$	Year	Ref.
$^{210}\text{Po}$	$5.3 \pm 1.7$	NaI(Tl)+Ge(Li)	1986	[10]	4.4	1978	[6]
$^{239}\text{Pu}$	$7 \pm 9$	NaI(Tl)+Ge(Li)	1986	[10]	2.2	1978	[6]
$^{241}\text{Am}$	$3.1 \pm 0.6$	NaI(Tl)+Ge(Li)	1973	[2]	1.2	1973	[2]
	$2.15 \pm 0.25$	NaI(Tl)+Ge(Li)	1986	[10]	2.3	1978	[6]
	$1.8 \pm 0.7^{(a)}$	Plastics+Ge	1990	[16]			
	$4.70 \pm 0.63$	NaI(Tl) pairs	2013	This work			

$$\lambda = \frac{A_{e^+e^-}}{A_\alpha}$$

[2] A. Ljubicic, B.A. Logan, Phys. Rev. C 7 (1973) 1541  
 [6] K. Pisk et al., Phys. Rev. C 17 (1978) 739  
 [10] J. Stanicek et al., Nucl. Instrum. Meth. B 17 (1986) 462  
 [16] T. Asanuma et al., Phys. Lett. B 237 (1990) 588

**Theory, which describes the effect as creation of bremsstrahlung  $\gamma$  during  $\alpha$  acceleration with  $E_\gamma > 1.022$  MeV which borns  $e^+e^-$ , gives  $\lambda$  value of a correct order of magnitude.**

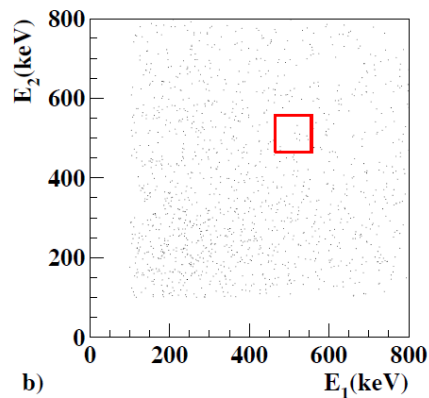
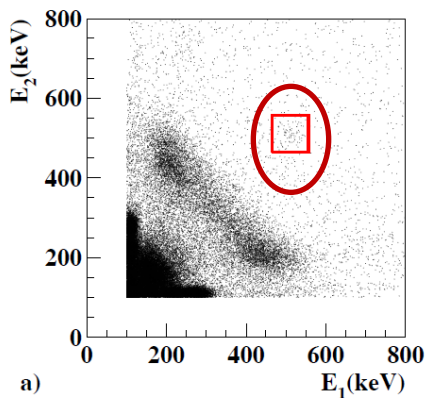
In the DAMA experiment [R. Bernabei et al., Int. J. Mod. Phys. A 28 (2013) 1330022],  $^{241}\text{Am}$  sources are used for weekly calibrations. So, an idea appeared to check the old  $\alpha$ -IPP results, **at the first time deep underground** (avoiding influence of cosmic rays) and **in low-background high-pure set-up** (suppressing presence of  $\beta^+$  contaminations).

**DAMA/LIBRA: 25 NaI(Tl) scintillators, 9.70 kg each, 10.2×10.2×25.4 cm.**

**1<sup>st</sup> run:** 6  $^{241}\text{Am}$  sources and 6 NaI(Tl) pairs (all other NaI(Tl)'s – as anticoincidence); 1.29 d with  $^{241}\text{Am}$  and 24.6 d background; total  $^{241}\text{Am}$  activity 200.8 kBq; result: excess rate of double coincidences in 465-557 keV ( $\pm 2\sigma$  region)  **$s = 4.87 \pm 0.87$  counts/d/NaI(Tl)pair**

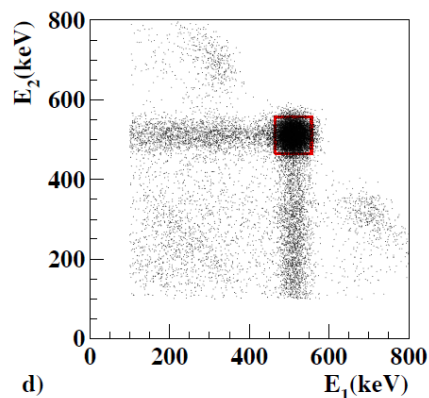
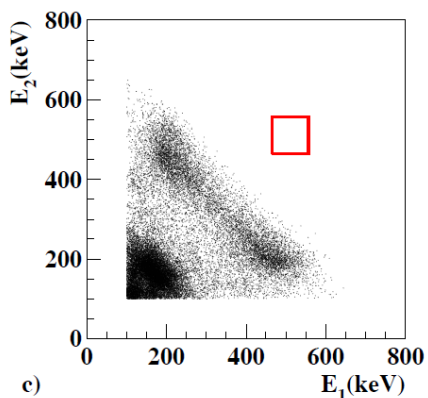
**2<sup>nd</sup> run:** 3  $^{241}\text{Am}$  sources and 3 NaI(Tl) pairs, 2.63 d ( $^{241}\text{Am}$ ) + 24.6 d (bkg), 98.9 kBq,  **$s = 5.23 \pm 0.90$  counts/d/NaI(Tl)pair**

Exp.  
spectrum  
with  $^{241}\text{Am}$



Exp.  
background

Simulated  
 $^{241}\text{Am}$   
decay

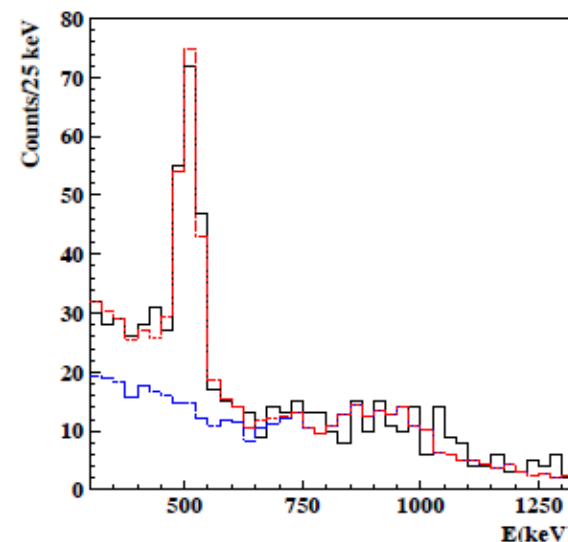


Simulated  
 $e^+$   
annihilation

Analysis of data:  
presence of  $^{243}\text{Am}$ ,  
 $^{233}\text{Pa}$ ,  $^{154}\text{Eu}$

Spectrum of one  
NaI(Tl) when  
energy of second  
is 465-557 keV

**$S=220\pm30$  counts**  
 **$\lambda=(4.70\pm0.63)\times 10^{-9}$**



**Many possible mimicking contributions were analyzed and excluded:**

- $\beta^+$  emitters in chain  $^{241}\text{Am} \rightarrow \dots \rightarrow ^{209}\text{Bi}$ ;
- high energy  $\gamma$  rays in chain  $^{241}\text{Am} \rightarrow \dots \rightarrow ^{209}\text{Bi}$ ;
- $\alpha$  decays of  $^{241}\text{Am}$  to high energy levels of  $^{237}\text{Np}$ ;
- $(\alpha, n)$  and  $(\alpha, p)$  reactions on isotopes of C, N, O, Cu in surrounding materials which lead to creation of  $\beta^+$  emitters;
- $(\alpha, n\gamma)$  and  $(\alpha, \gamma)$  reactions leading to high energy  $\gamma$ 's;
- $^{241}\text{Am}$  fission and cluster decays.

**The observed excess cannot be explained by any side process.**

**It would be interesting to repeat such studies with HPGe detectors with high energy resolution.**

# 5. Conclusions

**There was a little interest in investigations of rare  $\alpha$  and  $\beta$  decays since ~1970's – no  $T_{1/2}$  were measured with higher precision, no shapes of  $\beta$  spectra.**

**However, development of experimental technique lead to improvement in sensitivity, and new decays were observed with extreme characteristics ( $\alpha$  with longest  $T_{1/2}$  of  $10^{19}$  y for  $^{209}\text{Bi}$ ;  $\beta$  with lowest Q of 155 eV for  $^{115}\text{In}^*$ ; ...). Traditional (HPGe, NaI(Tl),...) but also new types of detectors ( $\text{Li}_6\text{Eu}(\text{BO}_3)_3$ ,...) are used in these investigations.**

**Interest to  $\beta$  shapes also is growing, in particular for nuclides which create background in rare events' searches.**

**Many theoretical works also appeared last time (especially in  $\alpha$  decay: tens of articles per year).**

**It could be concluded that investigations of rare  $\beta$  and  $\alpha$  decays experience revival now.**



**Thank you for attention!**