Investigation of double-beta decay of ^{150}Nd to excited states of ^{150}Sm by the low-counting γ spectrometry

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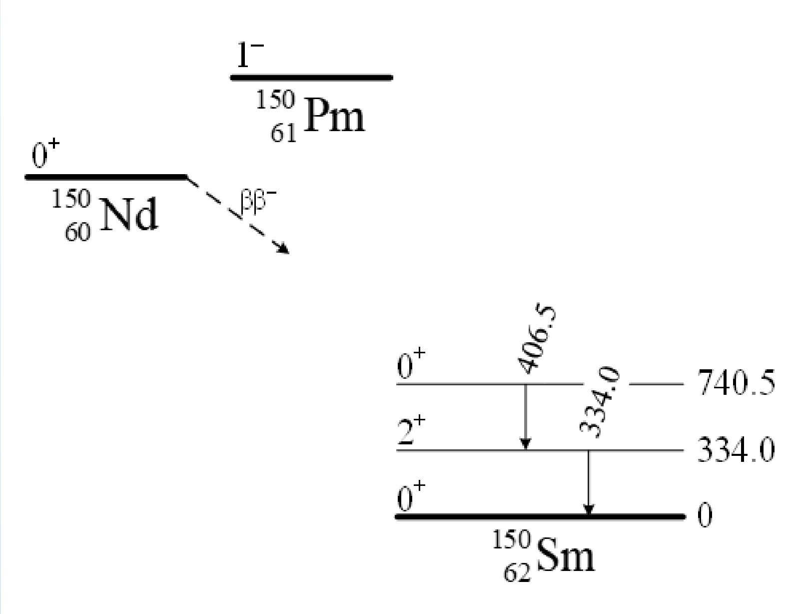
2β decay of ^{150}Nd



^{150}Nd 2β decay scheme

^{150}Nd is one of the most prospective isotopes for investigation of 2β decay:

- Energy release

  $Q_{\beta\beta}$ = 3371.38(20) keV [1];

- Natural isotopic abundance

  δ = 5.638(28)% [2];

- Possibility to investigate the decay to excited levels of ^{150}Sm with high energy resolution (HP Ge spectrometry).

[1] V.S. Kolhinen et al., Phys. Rev. C 82 (2010) 022501.

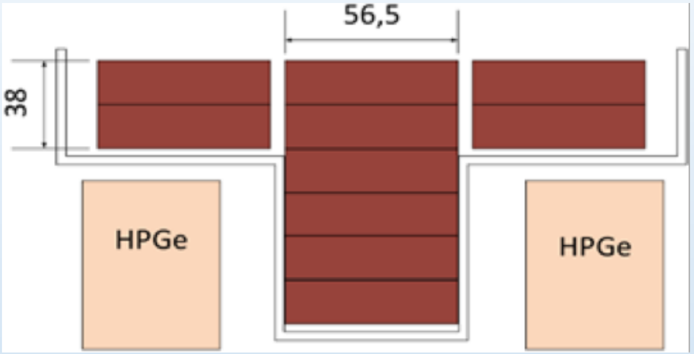
[2] J. Meija et al., Pure Appl. Chem. 88 (2016) 293.

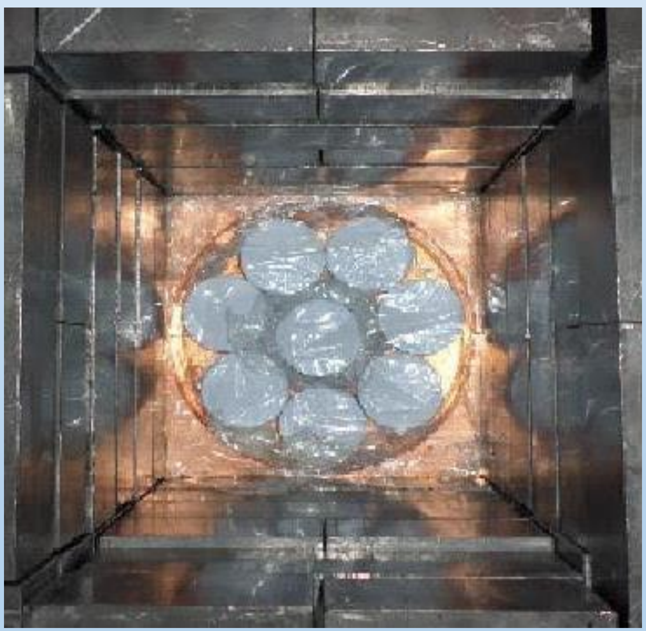
Experimental results for $^{150}Nd \rightarrow ^{150}Sm$ (0^+, 740.5 keV)

Short description	$T_{1/2}$, 10^{19} y (68% C.L.)	Year [Ref.]
Modane underground laboratory (4800 m w.e.), HP Ge 400 cm^3, 3046 g of Nd_2O_3 (δ = 5.638%), 11321 h, 1-d spectrum (Re-estimation)	14^{+5}_{-4} ($13.3^{+4.5}_{-2.6}$)	2004 [1] (2009 [2])
Kimballton Underground Research Facility, 2 HP Ge (~304 cm^3 each one), 50 g $^{150}Nd_2O_3$ (δ = 93.6%), 15427 h, coincidence spectrum	$10.7^{+4.6}_{-2.6}$	2014 [3]
Modane underground laboratory (4800 m w.e.), NEMO-3 detector, foil with 57.2 g of $^{150}Nd_2O_3$ (δ = 91.0%), 40774 h, e^- and γ energy, e^- tracks (*preliminary result*)	$11.1^{+2.6}_{-2.1}$	2022 [4]

[1] A.S. Barabash et al., Phys. Atom. Nucl. 67 (2004) 1216;
[2] A.S. Barabash et al., Phys. Rev. C 79 (2009) 045501;
[3] M.F. Kidd et al., Phys. Rev. C 90 (2014) 055501;
[4] V. Tretyak et al., Abstracts of "Nucleus-2021", p. 257.

Experimental setup

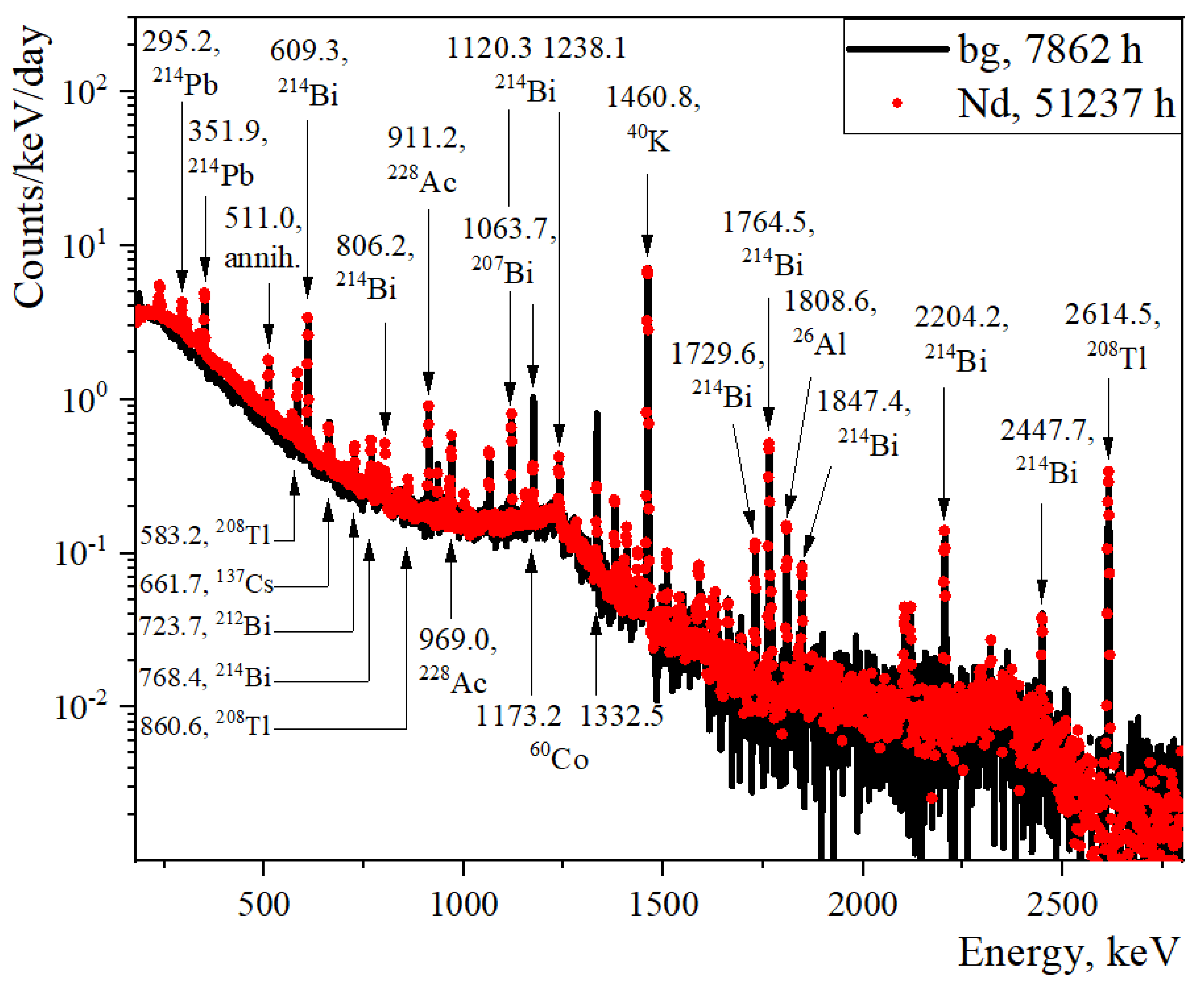




- 2381-g Nd_2O_3 sample (average density ~2.84 g/cm^3), used in previous experiment [1] and additionally purified before the measurements [2].
- Gran Sasso underground lab (~3.6 km of w.e.)
- 4 HP Ge detectors (≃225 cm^3 each) in a cryostat with cylindrical well in the center;
- Shield: copper (10 cm), lead (20 cm);
- Removing radon: Plexiglas container flushed with high-purity nitrogen gas;
- Measurement time: 51237 hours (~5.85 y.).
- Detection of energy and time of events (count coincidence (CC) analysis is possible)

[1] A.S. Barabash et al., Phys. Atom. Nucl. 67 (2004) 1216.
[2] R.S. Boiko, Int. J. Mod. Phys. A 32 (2017) 1743005.

Nd_2O_3 vs. background spectra



Radioactive contamination of Nd_2O_3 sample

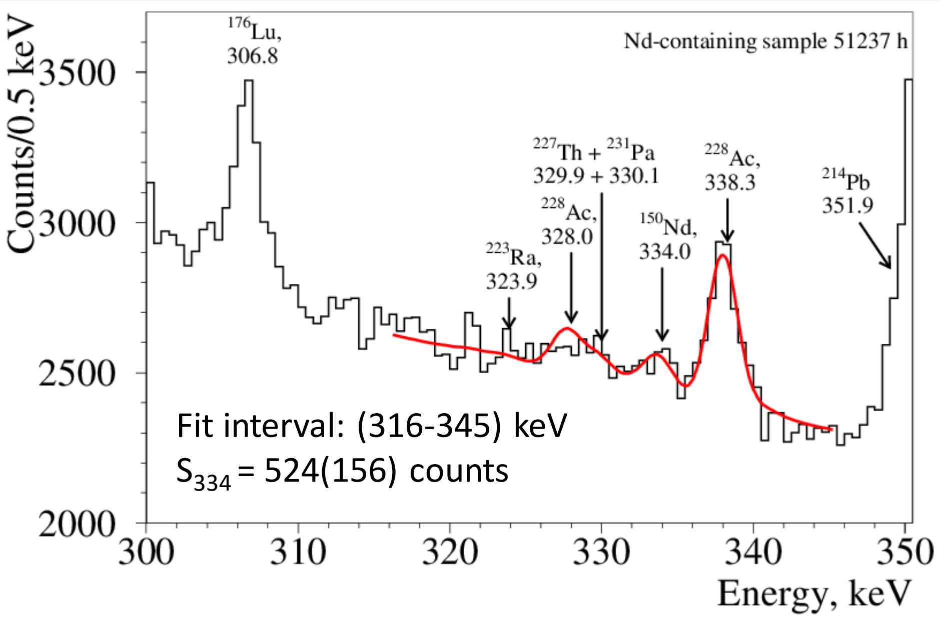
Sub-chain	A_m, mBq/kg *	
	This work	Before purification [1]
^{238}U chain		
^{238}U	≤ 3.7	≤ 28
^{226}Ra	**≤ 0.16**	1.5(8)
^{232}Th chain		
^{228}Ra	≤ 0.3	≤ 2.1
^{228}Th	0.33(5)	≤ 1.3
^{235}U chain		
^{235}U	≤ 0.7	≤ 1.7
^{231}Pa	≤ 1.0	—**
^{227}Ac	≤ 0.17	—**

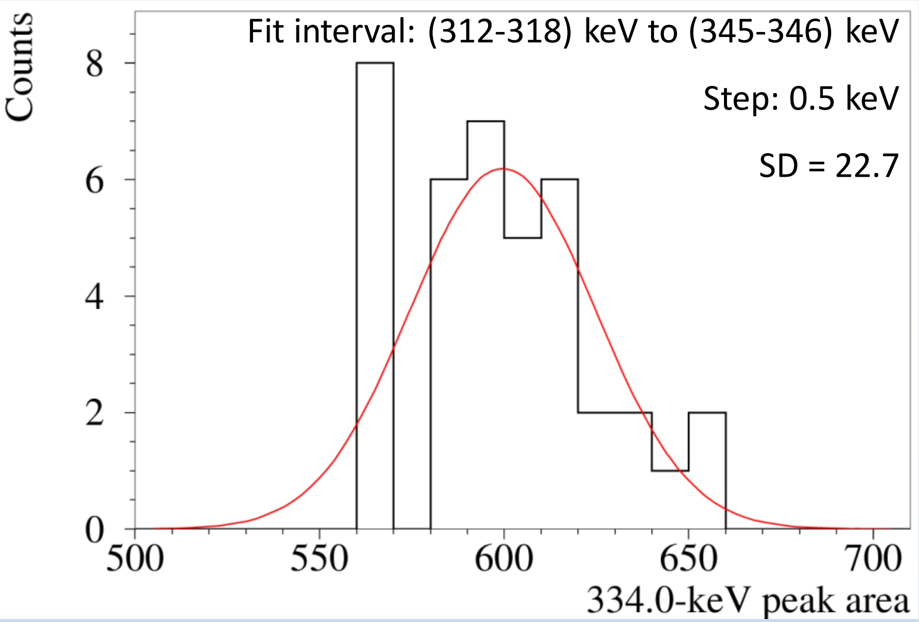
Sub-chain	A_m, mBq/kg *	
	This work	Before purification [1]
^{40}K	**3.1(7)**	16(8)
^{60}Co	≤ 0.03	—**
^{108m}Ag	≤ 0.018	—**
^{133}Ba	≤ 0.08	—**
^{137}Cs	≤ 0.018	≤ 0.8
^{138}La	0.085(7)	—**
^{150}Eu	≤ 0.06	—**
^{152}Eu	≤ 0.13	—**
^{154}Eu	≤ 0.031	—**
^{176}Lu	**0.33(3)**	1.1(4)
^{207}Bi	≤ 0.07	—**

- * Here and below: 68% C.L. for uncertainties; 90% C.L. for limits
- ** The measurement time was too low to observe corresponding peaks

[1] R.S. Boiko, Int. J. Mod. Phys. A 32 (2017) 1743005.

334.0-keV peak





$T_{1/2} = 7.1^{+3.0}_{-1.7} \times 10^{19}$ y (statistical uncertainties only);

Interval of fit: $\sigma_{\mathrm{fit}} = \pm 0.3 \times 10^{19}$ y;

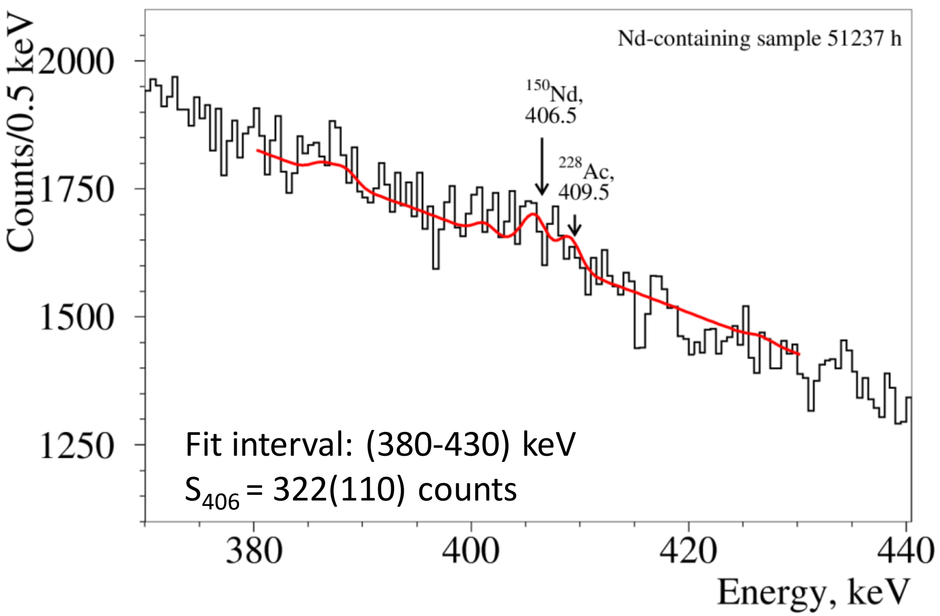
Monte Carlo simulation: $\sigma_{\mathrm{MC}} = \pm 0.7 \times 10^{19}$ y;

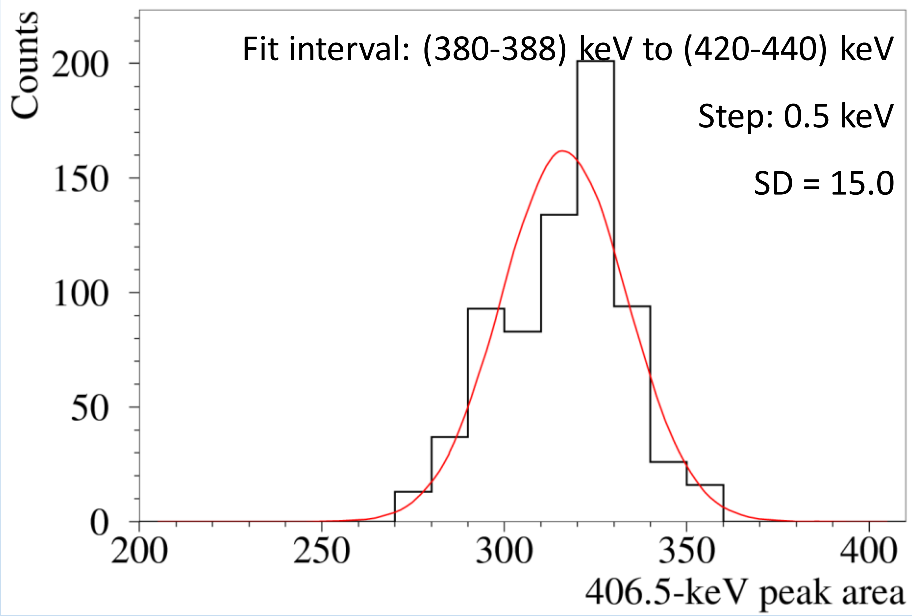
Number of nuclei: $\sigma_{\mathrm{nucl}} = \pm 0.05 \times 10^{19}$ y;

$A_{\mathrm{m}}(^{150}\mathrm{Eu}) \leq 0.031$ mBq/kg (68% C.L.) → $\Delta S \leq 283$ counts → $\sigma_{^{150}\mathrm{Eu}} = +8.3 \times 10^{19}$ y

$$T_{1/2} = 7.1^{+3.0}_{-1.7}(\mathrm{stat.}) \pm {}^{+8.3}_{-0.8}(\mathrm{syst.}) \times 10^{19}\ \mathrm{y}$$

406.5-keV peak





$T_{1/2} = 16.5^{+19.0}_{-5.8} \times 10^{19}$ y (statistical uncertainties only);

Interval of fit: $\sigma_{\rm fit} = ^{+1.1}_{-1.0} \times 10^{19}$ y;

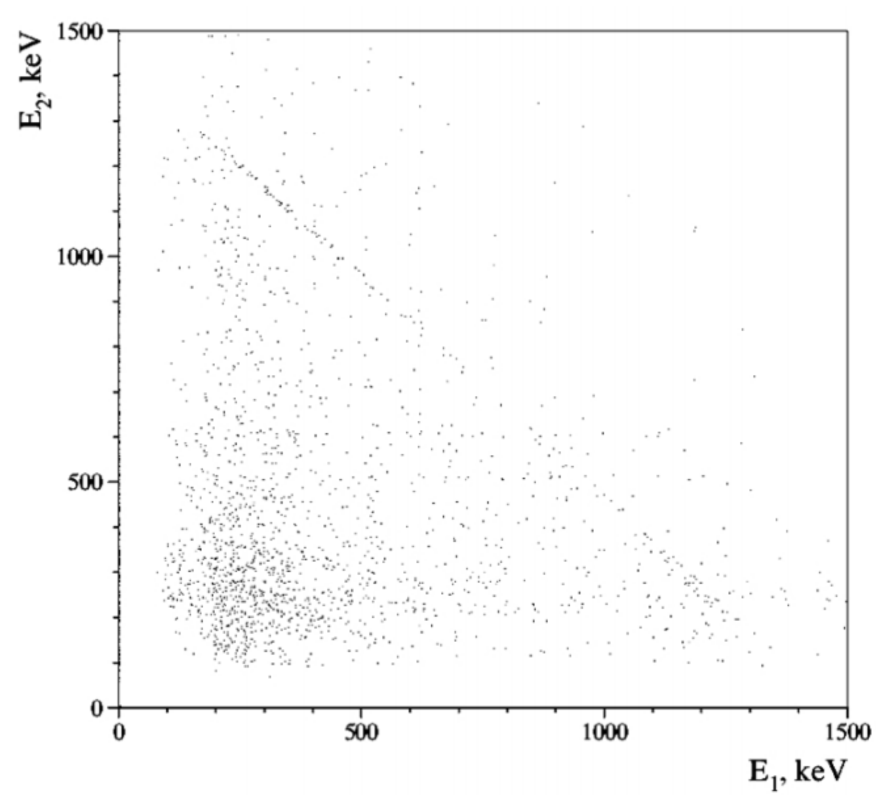
Monte Carlo simulation: $\sigma_{\rm MC} = \pm 1.7 \times 10^{19}$ y;

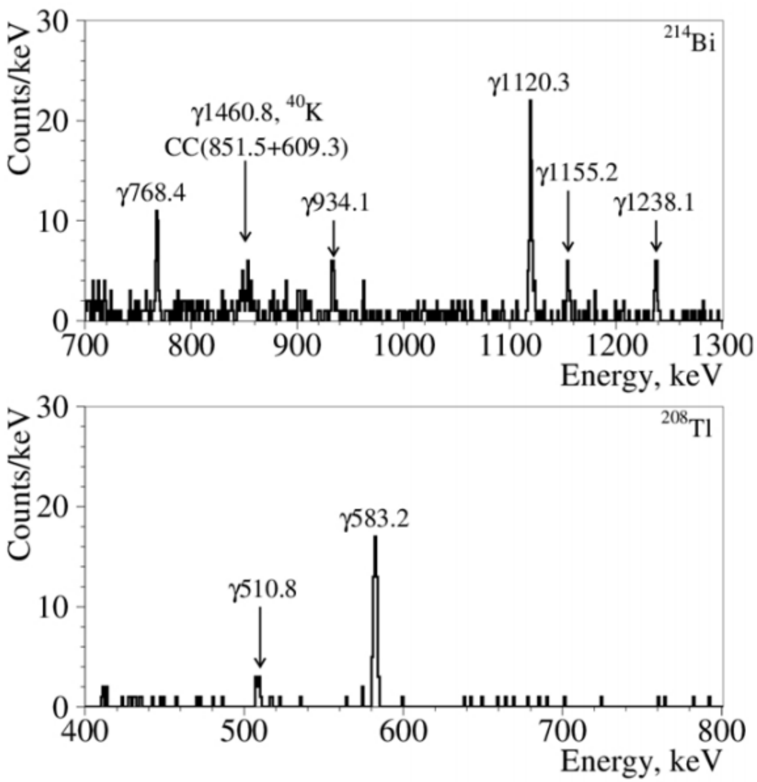
Number of nuclei: $\sigma_{\rm nucl} = \pm 0.12 \times 10^{19}$ y;

$A_{\rm m}(^{150}{\rm Eu}) \leq 0.031$ mBq/kg (68% C.L.) → $\Delta S \leq 0.4$ counts → $\sigma_{^{150}{\rm Eu}} = +0.03 \times 10^{19}$ y

$$T_{1/2} = 16.5^{+19.0}_{-5.8}({\rm stat.}) \pm 2.0({\rm syst.}) \times 10^{19}\ {\rm y}$$

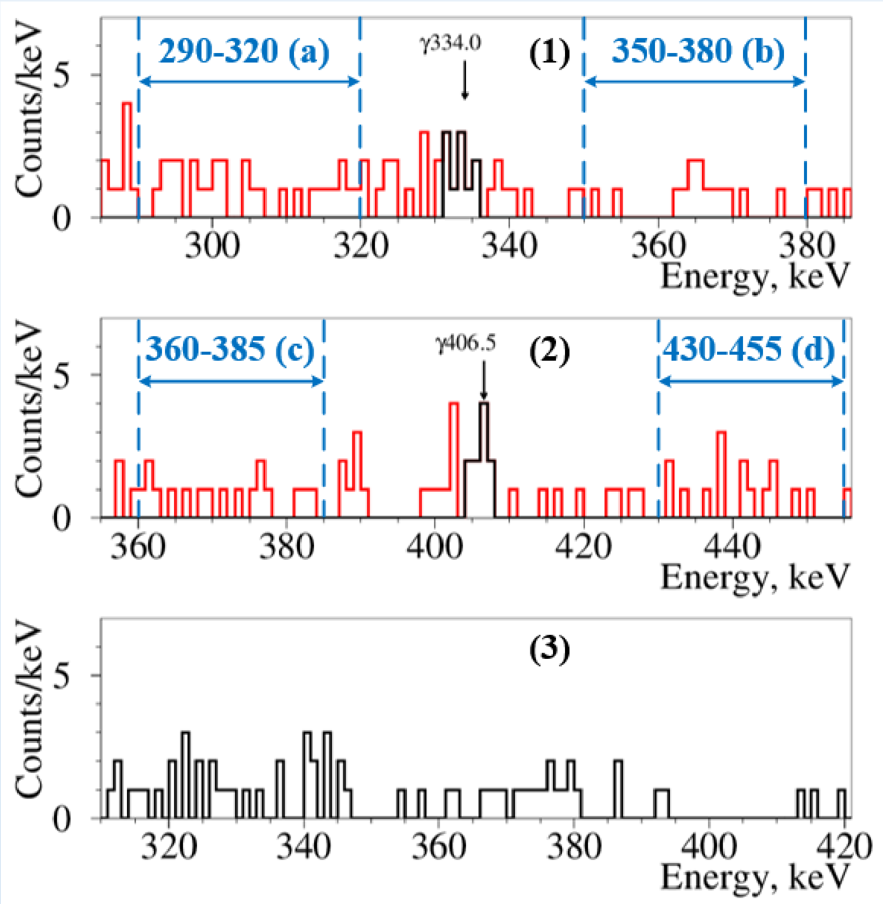
Coincidence analysis





- 2-dimensional energy spectrum of coincidences (*left*);
- Energy in one detector is fixed as (609 ± 5) keV (^{214}Bi, *top right*)
- Energy of one detector is fixed as (2615 ± 5) keV (^{208}Tl, *bottom right*).

334.0 + 406.5 CC (±1.8σ selection interval)

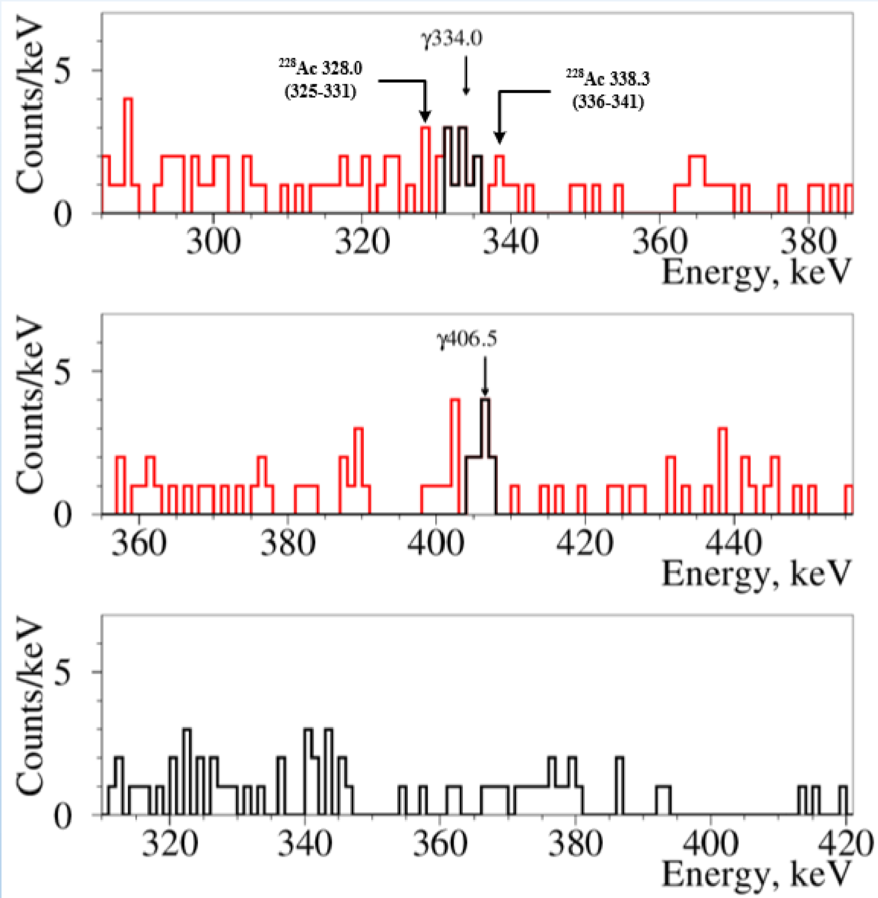


- N = 10
- $Int_{334} = 1.8 \cdot 2 \cdot \frac{\sum_{i=1}^{4} \sigma_{\text{det}\_i}}{4} = 4.22$ keV
- $N_{bg}(1) = N_{bg}(\text{a})+N_{bg}(\text{b})=45$ →

→ b = 0.742 counts/keV

- $B(1) = Int_{334} \bullet b$ = 3.13 counts
- $B(2)$ = 2.56 counts
- B = (B(1) + B(2))/2 = 2.85 counts
- $S_{CC}^{0} = [5.1 \ldots 12.1] = 7.2^{+3.8}_{-3.3}$ counts
- (F.-C., 68% C.L.)
- CC selection efficiency:
- $\eta = \left(\int_{-1.8\sigma}^{1.8\sigma} g\,(0,\sigma)\,\text{dx}\right)^2 = 0.8614$
- $S_{CC\_norm} = {S_{CC}^{0}}/{\eta} = 8.3^{+4.4}_{-3.8}$ counts

334.0 + 406.5 CC (estimation of background from ^{228}Ac)



- Interval: (325…331) keV
- $N_{328} = 6$
- $B_{328} = 4.13$
- $S^0_{328} = [0.37 \ldots 5.30] = 2.0^{+3.3}_{-1.7}$ counts
- $\eta_{328} = \dfrac{\sum_{i=1}^{4} \int_{325}^{331} g\,(328.0, \sigma_i)\mathrm{dx}}{4} = 0.9727$
- $S_{328} = {S^0_{328}}/{\eta_{328}} = 2.1^{+3.4}_{-1.7}$ counts
- Part of S_{328} inside CC selection interval:
- $\delta = \dfrac{\sum_{i=1}^{4} \int_{334-1.8\sigma_i}^{334+1.8\sigma_i} g\,(328.0, \sigma_i)\mathrm{dx}}{4} = 1.021\%$
- $\Delta S_{328} = S_{328} \cdot \dfrac{\delta_{328}}{\eta} = 0.025^{+0.040}_{-0.021}$ counts

The same procedure performed for 338.3-keV peak (selection interval (336-341) keV):

$$\Delta S_{338} = S_{338} \cdot \frac{\delta_{338}}{\eta} = 0.18^{+0.30}_{-0.15} \text{ counts}$$

CC systematics

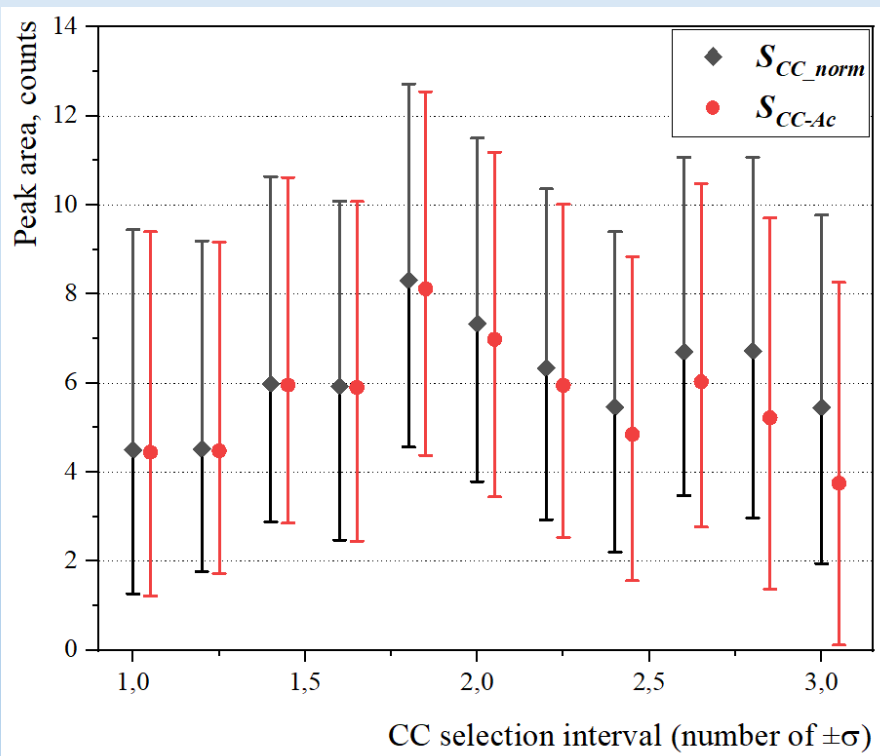
$S_{CC-Ac} = S_{CC\_norm} - \Delta S_{328} - \Delta S_{338} = 8.1^{+4.4}_{-3.7} \;\; \rightarrow \;\; T_{1/2} = 8.8^{+7.6}_{-3.1}(\text{stat.}) \times 10^{19}$ y.

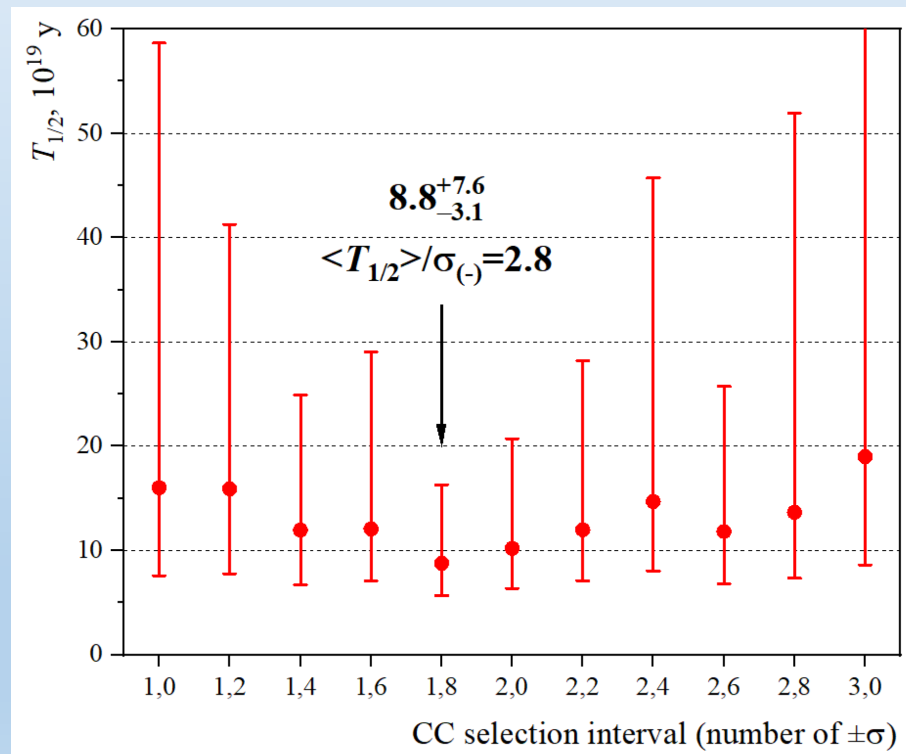
The same procedure for intervals ± (1.0σ, 1.2σ, 1.4σ, ..., 3.0σ).

CC selection interval: SD ($T_{1/2}$) = 3.0 × 10^{19} y

Number of nuclei: $\sigma_{nucl} = \pm 0.07 \times 10^{19}$ y;

MC simulation: $\sigma_{MC} = \pm 1.3 \times 10^{19}$ y



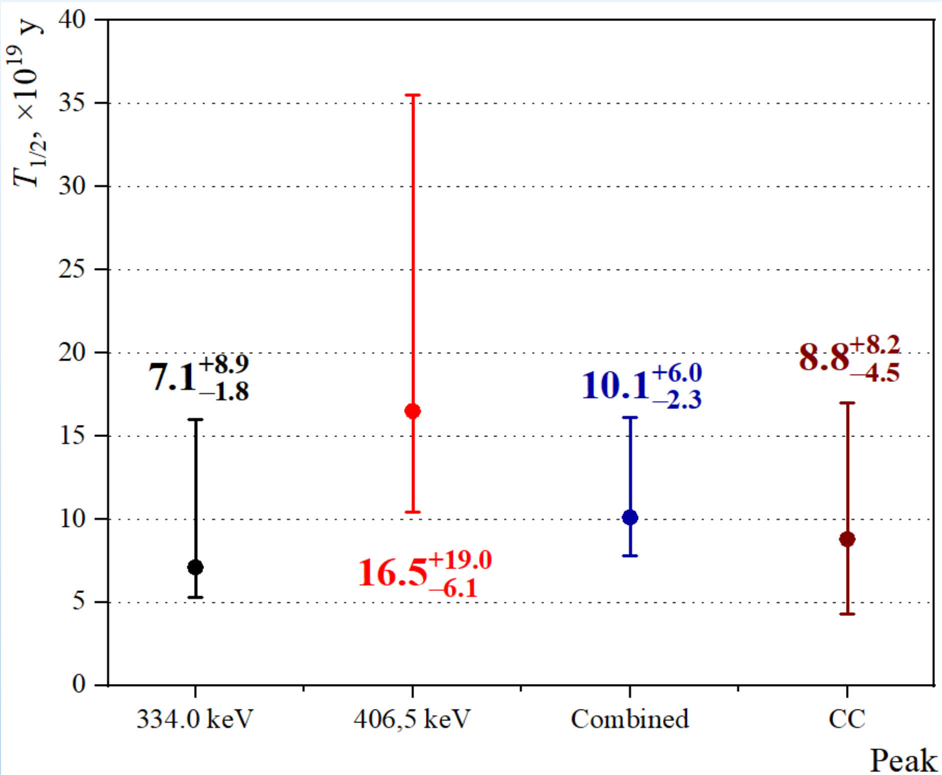


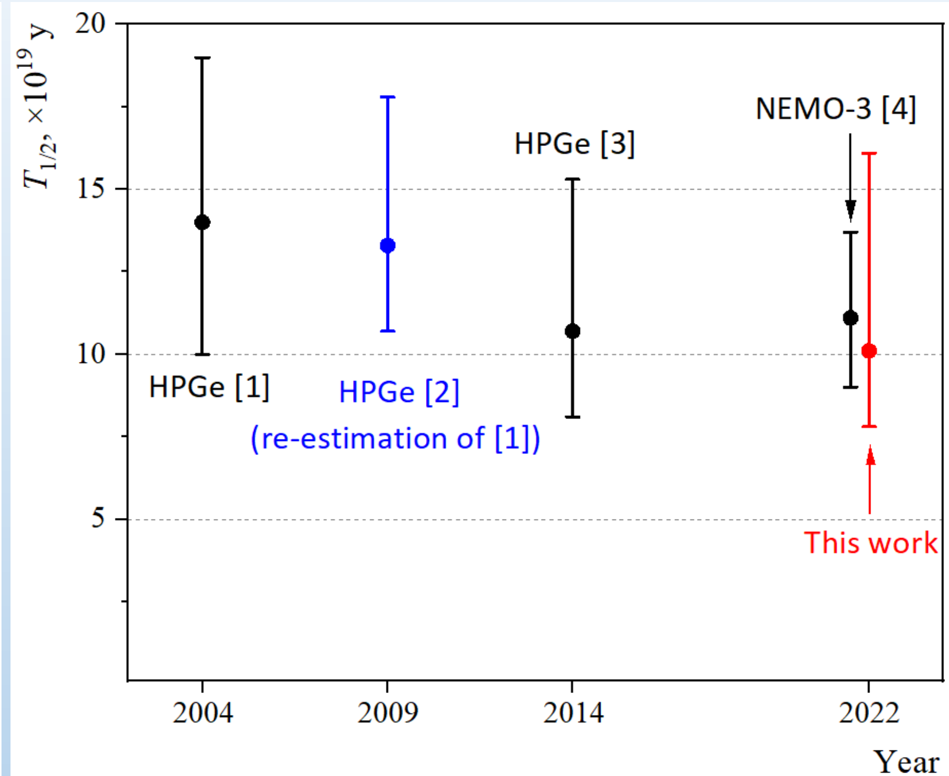
$T_{1/2}$ combined value & systematics summary

- $T_{1/2}^{\text{comb}} = \frac{\ln 2 \cdot N \cdot t \cdot \sum \varepsilon_i}{\sum S_i} = 10.1^{+3.6}_{-2.1}(\text{stat}) \times 10^{19}$ y

Value, × 10^{19} y	334.0	406.5	combined	CC
$\langle T_{1/2} \rangle$	7.1	16.5	10.1	8.8
Statistical uncertainty	$^{+3.0}_{-1.7}$	$^{+19.0}_{-5.8}$	$^{+3.6}_{-2.1}$	$^{+7.6}_{-3.1}$
Systematics				
Number of nuclei	±0.05	±0.12	±0.08	±0.07
Interval of fit (CC selection)	±0.3	$^{+1.1}_{-1.0}$	±0.4	±3.0
MC simulation	±0.7	±1.7	±0.7	±1.3
^{150}Eu possible presence	+8.3	+0.03	+4.8	—
Sum systematics	$^{+8.3}_{-0.8}$	±2.0	$^{+4.9}_{-0.8}$	±3.2
Sum uncertainty	$^{+8.9}_{-1.8}$	$^{+19.0}_{-6.1}$	$^{+6.0}_{-2.3}$	$^{+8.2}_{-4.5}$

Our result vs. previous ones





[1] A.S. Barabash et al., Phys. Atom. Nucl. 67 (2004) 1216;
[2] A.S. Barabash et al., Phys. Rev. C 79 (2009) 045501;
[3] M.F. Kidd et al., Phys. Rev. C 90 (2014) 055501;
[4] V. Tretyak et al., Abstracts of "Nucleus-2021", p. 257.

Origin of 334.0-keV peak area excess?

- S_{334} = 524(156) counts; should be 298(102) counts (calculated from S_{406}).
- Peak area excess: ΔS_{334} = 226(186) counts.
- Three possible sources:

1. Statistical 'artifact' → the probability of such statistical effect should be tested.
2. ^{150}Eu in the sample (I_{334}=95.16%; I_{406}=0.139%). Then, $A_{\rm m}$ ≤ 0.05 mBq/kg (90% C.L.). From analysis of the other peaks: $A_{\rm m}$ ≤ 0.06 mBq/kg → ^{150}Eu possible content in the sample should be estimated by using data of the mass-spectromtry measuremnts (in progress), and calculations of possible generation of ^{150}Eu in the sample by cosmic rays and neutrons..
3. Indication of ^{150}Nd→^{150}Sm(2^+, 334.0 keV). Then, $T_{1/2}$≥0.8×10^{20} y (90% C.L.) or $T_{1/2} = 1.8^{+8.5}_{-0.9}(\text{stat.}) \times 10^{20}$ y. In [1] $T_{1/2}$≥2.2×10^{20} y is reported; the decay could be much more suppressed → theoretical calculations are in progress.

[1] A.S. Barabash et al., Phys. Rev. C 79 (2009) 045501

Conclusions

- Experimental investigation of 2β decay of ^{150}Nd to the first 0^+ excited state of ^{150}Sm has been performed by using low-background 4-crystal HPGe γ-spectrometer at the Gran Sasso UL.
- Gamma quanta expected in the decay have been observed in the spectrum of single events, as well as in the coincidence spectra. The half-life of ^{150}Nd relatively to the 2β decay to the 0_1^+ excited state of ^{150}Sm is estimated (preliminary) as

$$T_{1/2} = [10.1^{+3.6}_{-2.1}(\text{stat.})^{+4.9}_{-0.8}(\text{syst.})] \times 10^{19}\ \text{y}$$

- Possible indication of ^{150}Nd decay to the 2^+(334.0 keV) has been observed, but the effect could have different origin; additional analysis and theoretical calculations are in progress.