Available online at www.sciencedirect.com
ScienceDirect
Nuclear Physics A 859 (2011) 126–139
NUCLEAR PHYSICS A
www.elsevier.com/locate/nuclphysa

First search for double β decay of dysprosium

P. Belli [a], R. Bernabei [a,b,*], F. Cappella [c,d], R. Cerulli [e], F.A. Danevich [f], S. d'Angelo [a,b], M.L. Di Vacri [e], A. Incicchitti [c,d], M. Laubenstein [e], S.S. Nagorny [f], S. Nisi [e], A.V. Tolmachev [g], V.I. Tretyak [f], R.P. Yavetskiy [g]

[a] *INFN sezione Roma "Tor Vergata", I-00133 Rome, Italy*
[b] *Dipartimento di Fisica, Università di Roma "Tor Vergata", I-00133 Rome, Italy*
[c] *INFN sezione Roma "La Sapienza", I-00185 Rome, Italy*
[d] *Dipartimento di Fisica, Università di Roma "La Sapienza", I-00185 Rome, Italy*
[e] *INFN, Laboratori Nazionali del Gran Sasso, 67010 Assergi (AQ), Italy*
[f] *Institute for Nuclear Research, MSP 03680 Kyiv, Ukraine*
[g] *Institute for Single Crystals, 61001 Kharkiv, Ukraine*

Received 17 February 2011; received in revised form 10 March 2011; accepted 11 April 2011

Available online 22 April 2011

Abstract

A search for double β decay of dysprosium was realized for the first time with the help of an ultra-low background HP Ge γ detector. After 2512 h of data taking with a 322 g sample of dysprosium oxide limits on double beta processes in ^{156}Dy and ^{158}Dy have been established on the level of $T_{1/2} \geqslant 10^{14}$–$10^{16}$ yr. Possible resonant double electron captures in ^{156}Dy and ^{158}Dy were restricted on a similar level. As a by-product of the experiment we have measured the radioactive contamination of the Dy_2O_3 sample and set limits on the α decay of dysprosium isotopes to the excited levels of daughter nuclei as $T_{1/2} \geqslant 10^{15}$–$10^{17}$ yr.

Keywords: RADIOACTIVITY ^{156}Dy(2EC), $(\beta^+ EC)$, (α); ^{158}Dy(2EC), (α); ^{160,161,162}Dy(α); measured E_γ, I_γ; deduced $T_{1/2}$ lower limits for α and various 2β-decay modes. Natural Dy_2O_3 sample of 322 g. Ultra-low background HPGe detector at the Gran Sasso National Laboratory underground facility

* Corresponding author at: INFN sezione Roma "Tor Vergata", I-00133 Rome, Italy.
E-mail address: rita.bernabei@roma2.infn.it (R. Bernabei).

0375-9474/$ – see front matter
doi:10.1016/j.nuclphysa.2011.04.003

1. Introduction

The double beta (2β) decay experiments are considered to-date as the best way to determine an absolute scale of the neutrino mass and to establish the neutrino mass hierarchy, to clarify the nature of the neutrino (Majorana or Dirac particle), to look for existence of right-handed admixtures in the weak interaction and of hypothetical Nambu–Goldstone bosons (Majorons), and to test some other effects beyond the Standard Model [1]. The developments in the new experimental techniques during the last two decades lead to an impressive improvement of sensitivity to the neutrinoless (0ν) mode of $2\beta^-$ decay up to 10^{23}–10^{25} yr [2]. Allowed in the Standard Model the two neutrino (2ν) double beta decay was detected for 10 nuclides with the half-lives in the range of 10^{18}–10^{24} yr [2,3].

The sensitivity of the experiments to search for the double electron capture (2ε), the electron capture with emission of positron ($\varepsilon\beta^+$), and the double positron ($2\beta^+$) decay is substantially lower: the best counting experiments give only limits on the level of 10^{18}–10^{21} yr [2,4–6].[1] There is a strong motivation to develop experimental technique to search for these processes: study of neutrinoless 2ε and $\varepsilon\beta^+$ decays could clarify the contribution of the right-handed admixtures in weak interactions [8]. Dysprosium contains two potentially 2β active isotopes: ^{156}Dy with one of the largest releases $Q_{2\beta} = (2012 \pm 6)$ keV (therefore both 2ε and $\varepsilon\beta^+$ channels of decay are possible), and ^{158}Dy ($Q_{2\beta} = (284.6 \pm 2.5)$ keV, only double electron capture is energetically allowed) [9]. The decay schemes of the triplets ^{156}Dy–^{156}Tb–^{156}Gd and ^{158}Dy–^{158}Tb–^{158}Gd are presented in Figs. 1 and 2, respectively.

It should be mentioned the possibility of a resonant enhancement of the neutrinoless double electron capture in ^{156}Dy and ^{158}Dy due to energy degeneracy. The resonant double electron capture was discussed in Refs. [12–15], where an enhancement of the decay rate by some orders of magnitude was predicted for the case of coincidence between the released energy and the energy of an excited state. According to [15], high Z atoms are strongly favored to search for resonant 2ε decay. Dysprosium has one of the highest Z among nuclides for which resonant processes could occur.

Resonant captures are possible on a few excited levels of ^{156}Gd and one level of ^{158}Gd. The properties of the excited levels are listed in Table 1. Because transitions with difference in spin more than 2 are strongly suppressed, we consider in this study only the levels of ^{156}Gd with spin $\leqslant 2$. However, we left in the list the level of ^{158}Gd with the spin 4^+ to which the resonant capture is possible.

Unfortunately, the isotopic abundances of both potentially double beta active dysprosium isotopes are rather low: concentrations of ^{156}Dy and ^{158}Dy in the natural dysprosium are 0.056(3)% and 0.095(3)%, respectively [16].

To our knowledge there were no attempts yet to search for double β decays of ^{156}Dy and ^{158}Dy. The aim of the present work was the search for 2β processes in the dysprosium isotopes with the help of ultra-low background high purity (HP) Ge γ spectrometry. As a by-product of the experiment we have estimated the radioactive contamination of the dysprosium oxide sample and set limits on the α decay of the dysprosium isotopes to excited levels of the daughter nuclei.

[1] An indication for $2\beta^+$ decay processes in ^{130}Ba and ^{132}Ba was obtained in geochemical measurements [7]; however, this result has to be confirmed in a direct counting experiment.

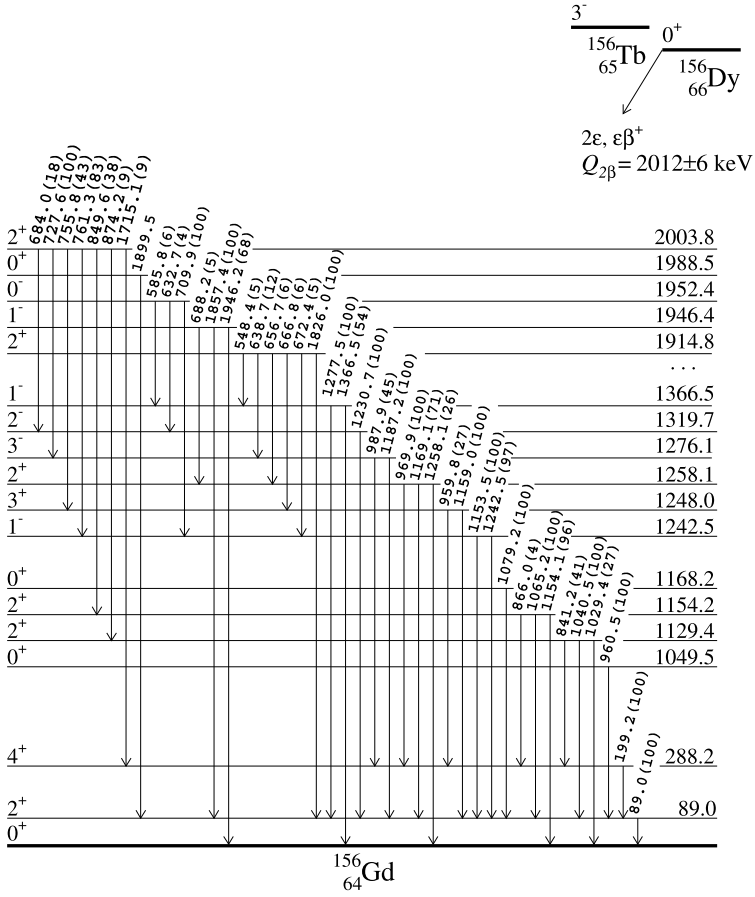

Fig. 1. Decay scheme of ^{156}Dy [10]. Energies of excited levels and emitted γ quanta are in keV (relative intensities of γ quanta, roughed to percent, are given in parentheses). Excited states related with possible population and subsequent de-excitation of 0^+ levels at $E_{exc} = 1851.2$ and 1715.2 keV, and 2^+ levels at $E_{exc} = 1827.8$ and 1771.1 keV are not shown. De-excitation branches with low probabilities ($\leqslant 1\%$) also are not shown.

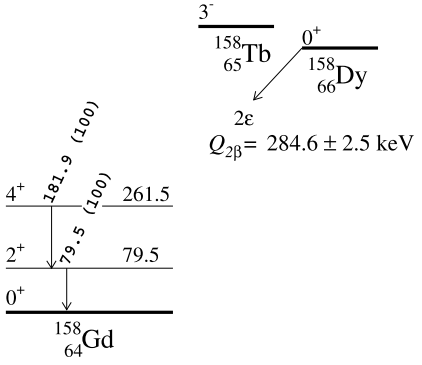

Fig. 2. Decay scheme of ^{158}Dy [11]. Energy of excited levels and γ quanta are in keV.

Table 1
Characteristics of possible resonant neutrinoless double electron capture in ^{156}Dy and ^{158}Dy. Difference $Q_{2\beta} - E_{exc}$ (where $Q_{2\beta}$ is the double beta decay energy, E_{exc} is the energy of an excited level) is denoted as δ.

Transition	Decay channel	Energy (keV) and parity of excited level	δ (keV)
^{156}Dy$\rightarrow^{156}$Gd	$2K$	1914.8, 2^+	-3 ± 6
	KL_1	1946.4, 1^-	7 ± 6
	KL_1	1952.4, 0^-	1 ± 6
	$2L_1$	1988.5, 0^+	7 ± 6
	$2L_3$	2003.8, 2^+	-6 ± 6
^{158}Dy$\rightarrow^{158}$Gd	$2L_1$	261.5, 4^+	6.3 ± 2.5

2. Measurements, results and discussion

2.1. Contamination of Dy_2O_3 measured by mass spectrometry

A sample of high pure dysprosium oxide (Dy_2O_3 of 99.98% purity grade) was provided by the Institute for Single Crystals (Kharkiv, Ukraine). The contamination of the material was measured with the help of High Resolution Inductively Coupled Plasma-Mass Spectrometric analysis (HR-ICP-MS, Thermo Fisher Scientific ELEMENT2). Potassium was measured in High Resolution mode. The calibration of the apparatus was performed by adding in one of the sample solutions of a known amount of a standard solution containing this element. All the other elements were determined in Low Resolution High Sensitivity mode. A semiquantitative analysis was performed, that is a single standard solution containing some elements at a known concentration level (10 ppb of Li, Ce, Tl) was used for calibration and quantification. In this case, the uncertainty can be estimated as about 30% of the given concentration value. The results of the HR-ICP-MS analysis are presented in Table 2. We have also calculated the activity of the radionuclides present in the measured elements.

2.2. Low background measurements

A Dy_2O_3 sample with the mass of 322 g in a polyethylene bag was placed directly on the end cap of the ultra-low background HP Ge detector (GeBer, 244 cm^3) installed deep underground ($\simeq$ 3600 m of water equivalent) at the Gran Sasso National Laboratories of the INFN (Italy). The detector was located inside a passive shield made of low radioactivity lead ($\approx$ 20 cm), copper ($\approx$ 10 cm) and borated polyethylene ($\approx$ 10 cm). To remove radon, the set-up was continuously flushed by highly pure nitrogen. The energy resolution of the spectrometer is 2 keV at 1332 keV γ line of ^{60}Co. The Dy_2O_3 sample was measured over 2512 h, and the background of the detector was collected during 6110 h.

The energy spectrum accumulated with the Dy_2O_3 sample by the HP Ge detector is shown in Fig. 3 together with the background data. The spectra are normalized on the times of measurements. The peaks in the spectra can be ascribed to γ quanta of the radionuclides ^{40}K, ^{60}Co, and daughters of U/Th. We have also observed γ lines of ^{176}Lu (201.8 and 306.8 keV) in the data accumulated with the dysprosium sample. To estimate radioactive contamination of the Dy_2O_3 sample, the detection efficiencies of the background components were simulated with the help of the EGS4 package [17]. The initial kinematics of the particles emitted in the decay of nuclei was given by an event generator DECAY0 [18]. Radioactive contamination of the dysprosium

Table 2
Contamination of Dy_2O_3 analyzed by ICP-MS analysis. The calculated activities of the most important radioactive isotopes presented in the radioactive elements are given.

Element	Concentrations of element (ppm)	Activity of radioactive isotopes (mBq/kg)
K	$\leqslant 0.5$	$\leqslant 15$ (^{40}K)
Ca	50	
Co	$\leqslant 0.05$	
Cu	$\leqslant 5$	
Sr	$\leqslant 0.02$	
Y	6	
La	0.25	0.21 (^{138}La)
Sm	17	2200 (^{147}Sm)
Gd	5	0.008 (^{152}Gd)
Tb	$\leqslant 5$	
Ho	30	
Er	60	
Yb	3	
Lu	0.5	26 (^{176}Lu)
Pb	0.16	
Th	0.036	147 (^{232}Th)
U	0.004	2.3 (^{235}U), 49 (^{238}U)

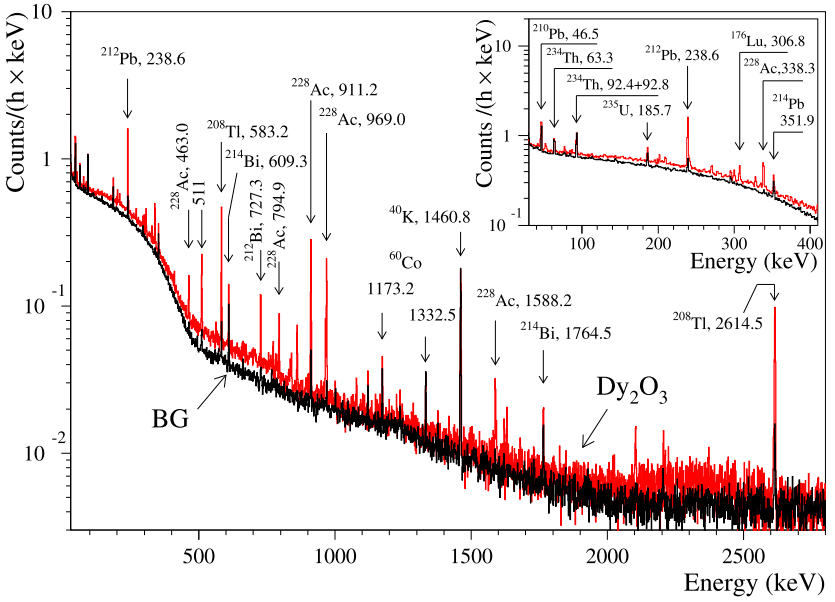

Fig. 3. (Color online.) Energy spectra accumulated with dysprosium sample over 2512 h (Dy_2O_3) and without sample over 6110 h (BG) by ultra-low background HP Ge γ spectrometer. (Inset) Low energy part of the spectra. Energy of γ lines are in keV.

oxide is presented in Table 3. The data agree with the results of the ICP-MS analysis taking into account accuracy of the measurements.

Table 3
Radioactive contamination of Dy_2O_3 sample measured by HP Ge γ spectrometry.

Chain	Nuclide	Activity (mBq/kg)
	^{40}K	$\leqslant 10$
	^{60}Co	$\leqslant 1$
	^{137}Cs	$\leqslant 2$
	^{138}La	$\leqslant 3$
	^{176}Lu	9 ± 2
^{232}Th	^{228}Ra	179 ± 40
	^{228}Th	157 ± 15
^{235}U	^{235}U	7 ± 3
	^{231}Pa	$\leqslant 80$
	^{227}Ac	$\leqslant 190$
^{238}U	^{234m}Pa	$\leqslant 115$
	^{226}Ra	5 ± 3
	^{210}Pb	$\leqslant 9400$

The observed contamination of the sample by U/Th is quite predictable taking into account that the principal ores for production of dysprosium are monazite and bastnaesite. Monazite contains considerable (typically a few %) amount of thorium and uranium. The presence of radioactive ^{176}Lu can also be expected due to the very similar chemical properties of lutetium and dysprosium, which lead to certain difficulties in chemical separation of the elements.

2.3. Search for 2ε and $\varepsilon\beta^+$ decay of ^{156}Dy

There are no clear peculiarities in the energy spectra accumulated with the dysprosium sample, which can be interpreted as double beta decay of ^{156}Dy or ^{158}Dy. Therefore only lower half-life limits can be set according to the formula:

$$\lim T_{1/2} = N \cdot \eta \cdot t \cdot \ln 2 / \lim S,$$

where N is the number of potentially 2β unstable nuclei, η is the detection efficiency, t is the measuring time, and $\lim S$ is the number of events of the effect searched for which can be excluded at a given confidence level (C.L.).

To estimate values of $\lim S$ for 2β processes in ^{156}Dy and ^{158}Dy, the energy spectrum accumulated with the Dy_2O_3 sample was fitted in different energy regions where γ peaks related with the decay processes are expected. The efficiencies of the HP Ge detector for the double β processes searched for were simulated with the help of the EGS4 code [17] and the DECAY0 event generator [18].

One positron can be emitted in the $\varepsilon\beta^+$ decay of ^{156}Dy with the maximal energy of (990 ± 6) keV. The annihilation of the positron will give two 511 keV γ's leading to extra counting rate in the annihilation peak. The energy spectra accumulated with and without the sample in the energy interval (400–700) keV are presented in Fig. 4. The measured area of the annihilation peak is (862 ± 44) counts. The area of the peak in the background spectrum is (149 ± 23) counts (normalized on the time of measurement with the Dy_2O_3 sample). The excess of events in the data accumulated with the dysprosium sample can be explained by radioactive contamination of the material. Indeed decays of ^{226}Ra and ^{228}Th daughters give in total (757 ± 72) counts in

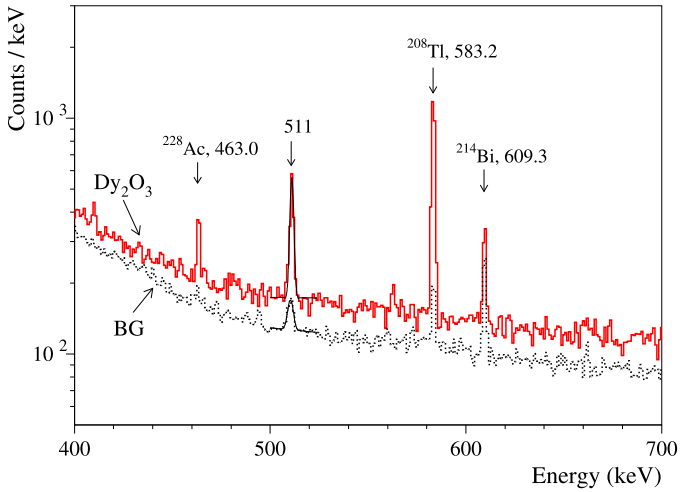

Fig. 4. (Color online.) Part of energy spectra accumulated with Dy_2O_3 sample over 2512 h by ultra-low background HP Ge γ spectrometer (Dy_2O_3). The energy spectrum accumulated without sample over 6110 h normalized on the time of measurements with the Dy_2O_3 sample (BG). Fits of 511 annihilation γ peaks are shown by solid lines.

the annihilation peak. The difference between the observed and the expected number of counts (-44 ± 87) gives no evidence for the effect of the $\varepsilon\beta^+$ decay of ^{156}Dy. In accordance with the Feldman–Cousins procedure [19], we obtain 102 counts which can be excluded at 90% C.L. (here and below all the half-life limits and values of $\lim S$ are given at 90% C.L.). Taking into account the number of ^{156}Dy nuclei in the sample ($N = 2.91 \times 10^{20}$) and the detection efficiency (3.4%), we have obtained the following limit on the half-life of ^{156}Dy relatively to the sum of 2ν and 0ν modes of $\varepsilon\beta^+$ decay:

$$T_{1/2}^{(2\nu+0\nu)\varepsilon\beta^+}\left(^{156}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant 1.9 \times 10^{16}\ \text{yr}.$$

The $\varepsilon\beta^+$ decay of ^{156}Dy is also allowed to the first excited level of ^{156}Gd with the energy of 89.0 keV. To estimate an upper limit on the transition, the energy spectrum was fitted in the energy interval (83–109) keV by the model constructed from a Gaussian function at the energy of 89 keV with the energy resolution FWHM = 1.6 keV (the γ peak searched for), a linear function describing the background, and a second Gaussian to take into account the γ peaks with the energies 92.4 and 92.8 keV (^{234}Th from ^{238}U chain). A fit by the chi-square method ($\chi^2/\text{n.d.f.} = 18.9/19 = 0.99$, where n.d.f. is number of degrees of freedom) gives the area of the peak searched for: $S = (86 \pm 88)$ counts, which gives no evidence for the effect. We took 230 counts which can be excluded at 90% C.L. Taking into account the simulated efficiency to 89 keV γ quanta (0.05%) we have obtained the limit on $2\nu\varepsilon\beta^+$ decay of ^{156}Dy to the first 2^+ 89.0 keV excited level of ^{156}Gd: $T_{1/2}^{2\nu\varepsilon\beta^+}(\text{g.s.} \to 89.0\ \text{keV}) \geqslant 1.3 \times 10^{14}$ yr. However, a much stronger restriction can be set by considering the annihilation peak. Indeed, the detection efficiency for the 511 keV γ for the $2\nu\varepsilon\beta^+$ (and $0\nu\varepsilon\beta^+$) decay to the 89.0 keV excited level is 3.4% which leads to the same limits as for the $\varepsilon\beta^+$ decay to the ground state:

$$T_{1/2}^{(2\nu+0\nu)\varepsilon\beta^+}\left(^{156}\text{Dy, g.s.} \to 89.0\ \text{keV}\right) \geqslant 1.9 \times 10^{16}\ \text{yr}.$$

In a case of the $2\nu 2K$ capture in ^{156}Dy a cascade of X-rays and Auger electrons with the individual energies up to 50 keV is expected. The most intensive X-ray lines are 42.3 keV (26.6%),

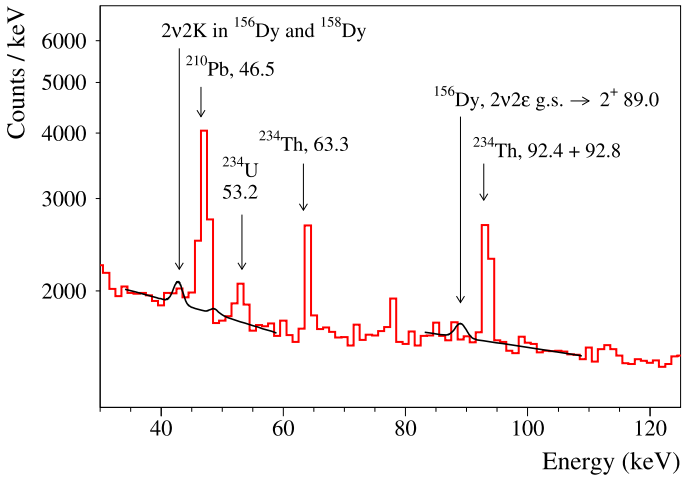

Fig. 5. (Color online.) Low energy part of energy spectrum accumulated with Dy_2O_3 sample over 2512 h. An excluded effect of $2\nu 2K$ process in ^{156}Dy (^{158}Dy) with the half-life $T_{1/2} = 6.1 \times 10^{14}$ yr ($T_{1/2} = 1.0 \times 10^{15}$ yr), and $2\nu 2\varepsilon$ transition of ^{156}Dy to the first excited level of ^{156}Gd (89.0 keV, $T_{1/2} = 1.8 \times 10^{14}$ yr) are shown by solid lines.

43.0 keV (47.5%), 48.6 keV (4.8%), 48.7 keV (9.3%) and 50.0 keV (3.1%) [20]. To derive a limit on the two neutrino double electron capture from K shells, the energy spectrum was fitted in the energy interval (35–60) keV by the model consisting of five Gaussians (to describe the expected X-ray peaks) plus two Gaussians (46.5 keV γ line of ^{210}Pb and 53.2 keV γ line of ^{234}U), and a polynomial function of the first degree (background; see Fig. 5). The fit gives the total area of the $2\nu 2K$ effect as (310 ± 102) counts. It is worth noting that the background in this energy region is quite complex to be safely estimated; therefore, conservatively we give here only a limit on the number of events: $\lim S = 477$ counts. Taking into account the detection efficiency of the effect (0.5%), one can calculate the following half-life limit:

$$T_{1/2}^{2\nu 2K}\left(^{156}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant 6.1 \times 10^{14} \text{ yr.}$$

To estimate a limit on the $2\nu 2\varepsilon$ decay of ^{156}Dy to the excited levels of ^{156}Gd, the energy spectrum accumulated with the dysprosium sample was fitted in energy intervals where peaks from the de-excitation γ-rays are expected. For instance, we have obtained the following limit on the $2\nu 2\varepsilon$ decay of ^{156}Dy to the first excited 89.0 keV level of ^{156}Gd:

$$T_{1/2}^{2\nu 2\varepsilon}\left(^{156}\text{Dy, g.s.} \to 89.0 \text{ keV}\right) \geqslant 1.8 \times 10^{14} \text{ yr.}$$

A peak expected for the $2\nu 2\varepsilon$ transition of ^{156}Dy to the excited 2^+ 89.0 keV level of ^{156}Gd is shown in Fig. 5.

The limits obtained for the $2\nu 2\varepsilon$ decay of ^{156}Dy to the excited levels of ^{156}Gd are presented in Table 4 where the energies of γ quanta used in the analysis, detection efficiencies and values of $\lim S$ are given too.

In case of $0\nu 2\varepsilon$ decay to the ground state of ^{156}Gd, we suppose here that only one bremsstrahlung γ quantum is emitted to carry out the transition energy (in addition to X-rays and Auger electrons from de-excitation of atomic shell). The energy of the γ quantum is expected to be equal to $E_\gamma = Q_{2\beta} - E_{b1} - E_{b2}$, where E_{b1} and E_{b2} are the binding energies of the first and of the second captured electron of the atomic shell. The binding energies on the K and L_1, L_2,

Table 4
Half-life limits on 2β processes in ^{156}Dy and ^{156}Dy.

Process of decay	Decay mode	Level of daughter nucleus (keV)	E_γ (keV)	Detection efficiency	lim S	Experimental limit (yr)
$^{156}\text{Dy} \to {}^{156}\text{Gd}$						
$\varepsilon\beta^+$	$2\nu + 0\nu$	g.s.	511	3.4%	102	$\geqslant 1.9 \times 10^{16}$
$\varepsilon\beta^+$	$2\nu + 0\nu$	2^+ 89.0	511	3.4%	102	$\geqslant 1.9 \times 10^{16}$
$2K$	2ν	g.s.	42.3–50.0	0.5%	477	$\geqslant 6.1 \times 10^{14}$
2ε	2ν	2^+ 89.0	89.0	0.07%	230	$\geqslant 1.8 \times 10^{14}$
2ε	2ν	0^+ 1049.5	960.5	1.1%	9	$\geqslant 7.1 \times 10^{16}$
2ε	2ν	2^+ 1129.4	1040.5	0.6%	24	$\geqslant 1.4 \times 10^{16}$
2ε	2ν	2^+ 1154.1	1154.1	0.5%	62	$\geqslant 4.7 \times 10^{15}$
2ε	2ν	0^+ 1168.2	1079.2	1.0%	65	$\geqslant 8.9 \times 10^{15}$
2ε	2ν	0^+ 1715.2	472.7	1.1%	21	$\geqslant 3.0 \times 10^{16}$
2ε	2ν	2^+ 1771.1	1682.2	0.7%	39	$\geqslant 1.0 \times 10^{16}$
2ε	2ν	2^+ 1827.8	1738.9	0.2%	6	$\geqslant 1.9 \times 10^{16}$
2ε	2ν	0^+ 1851.2	697.0	0.1%	39	$\geqslant 1.5 \times 10^{15}$
$2K$	0ν	g.s.	1906–1918	0.7%	24	$\geqslant 1.7 \times 10^{16}$
KL	0ν	g.s.	1947–1959	0.7%	24	$\geqslant 1.7 \times 10^{16}$
$2L$	0ν	g.s.	1989–2001	0.7%	18	$\geqslant 2.2 \times 10^{16}$
2ε	0ν	2^+ 89.0	89.0	0.06%	230	$\geqslant 1.5 \times 10^{14}$
2ε	0ν	0^+ 1049.5	960.5	1.0%	9	$\geqslant 6.4 \times 10^{16}$
2ε	0ν	2^+ 1129.4	1040.5	0.6%	24	$\geqslant 1.4 \times 10^{16}$
2ε	0ν	2^+ 1154.1	1065.2	0.5%	70	$\geqslant 4.1 \times 10^{15}$
2ε	0ν	0^+ 1168.2	1079.2	0.9%	65	$\geqslant 8.0 \times 10^{15}$
2ε	0ν	0^+ 1715.2	472.7	1.0%	21	$\geqslant 2.8 \times 10^{16}$
2ε	0ν	2^+ 1771.1	1682.2	0.6%	39	$\geqslant 8.9 \times 10^{15}$
2ε	0ν	2^+ 1827.8	1738.9	0.2%	6	$\geqslant 1.9 \times 10^{16}$
2ε	0ν	0^+ 1851.2	697.0	0.1%	39	$\geqslant 1.5 \times 10^{15}$
Resonant $2K$	$2\nu + 0\nu$	2^+ 1914.8	1826.0	0.5%	27	$\geqslant 1.1 \times 10^{16}$
Resonant KL_1	$2\nu + 0\nu$	1^- 1946.4	1857.4	0.4%	24	$\geqslant 9.6 \times 10^{15}$
Resonant KL_1	$2\nu + 0\nu$	0^- 1952.4	709.9	1.2%	27	$\geqslant 2.6 \times 10^{16}$
Resonant $2L_1$	$2\nu + 0\nu$	0^+ 1988.5	1899.5	0.7%	21	$\geqslant 1.9 \times 10^{16}$
Resonant $2L_3$	2ν	2^+ 2003.8	1715.1	0.02%	41	$\geqslant 2.8 \times 10^{14}$
Resonant $2L_3$	0ν	2^+ 2003.8	761.3	0.03%	58	$\geqslant 3.0 \times 10^{14}$
$^{158}\text{Dy} \to {}^{158}\text{Gd}$						
$2K$	2ν	g.s.	42.3–50.0	0.5%	477	$\geqslant 1.0 \times 10^{15}$
$2K$	0ν	g.s.	184.2	1.7%	40	$\geqslant 4.2 \times 10^{16}$
2ε	2ν	2^+ 79.5	79.5	0.04%	113	$\geqslant 3.5 \times 10^{14}$
2ε	0ν	2^+ 79.5	79.5	0.03%	113	$\geqslant 2.6 \times 10^{14}$
Resonant $2L_1$	$2\nu + 0\nu$	4^+ 261.5	181.9	1.3%	40	$\geqslant 3.2 \times 10^{16}$

L_3 shells in gadolinium atom are equal to $E_K = 50.2$, $E_{L_1} = 8.4$, $E_{L_2} = 7.9$ and $E_{L_3} = 7.2$ keV [20], respectively. Therefore, the expected energies of the γ quanta for the $0\nu 2\varepsilon$ capture in ^{156}Dy to the ground state of ^{156}Gd are: (i) $E_\gamma = (1912 \pm 6)$ keV for $0\nu 2K$; (ii) $E_\gamma = (1954 \pm 7)$ keV for $0\nu KL$; (iii) $E_\gamma = (1996 \pm 7)$ keV for $0\nu 2L$.

There are no clear peaks in the energy intervals (1906–1918), (1947–1959), and (1989–2004) keV expected in the g.s. $\to$ g.s. $0\nu 2\varepsilon$ decay of ^{156}Dy (see Fig. 6). We have estimated values of lim S by a fit of the data in the energy intervals by the simple model: a Gaussian function (to

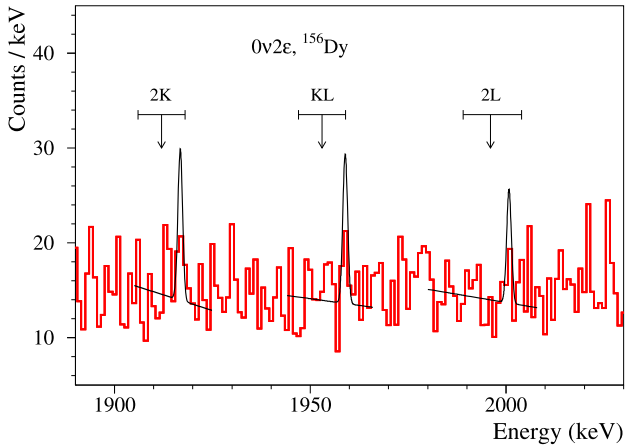

Fig. 6. (Color online.) Part of the energy spectra where peaks from $0\nu 2\varepsilon$ processes ($2K$, KL and $2L$) in ^{156}Dy to the ground state of ^{156}Gd are expected. The excluded peaks for the processes are shown.

describe the peaks searched for) plus polynomial function (continuous background). Taking into account the calculated efficiencies to detect γ quanta with energies $\approx$ 1.9–2.0 MeV ($\approx 0.7\%$), we set the following limits on the processes searched for:

$$T_{1/2}^{0\nu 2K}\left(^{156}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant 1.7 \times 10^{16}\ \text{yr},$$
$$T_{1/2}^{0\nu KL}\left(^{156}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant\ 1.7 \times 10^{16}\ \text{yr},$$
$$T_{1/2}^{0\nu 2L}\left(^{156}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant 2.2 \times 10^{16}\ \text{yr}.$$

The high $Q_{2\beta}$ energy of ^{156}Dy allows also population of several excited levels of ^{156}Gd with energies in the range of (89–2004) keV. To estimate limits on these processes, the energy spectrum accumulated with the dysprosium sample was analyzed in different energy intervals. A summary of the obtained results is given in Table 4.

2.4. 2ε decay of ^{158}Dy

An estimation of the half-life of ^{158}Dy relatively to the two neutrino double K electron capture was obtained by using the $\lim S$ and the detection efficiency for the $2\nu 2K$ decay of ^{156}Dy. Taking into account the number of ^{158}Dy nuclei in the sample ($N = 4.94 \times 10^{20}$) we have set the following limit on the $2\nu 2K$ decay of ^{158}Dy:

$$T_{1/2}^{2\nu 2K}\left(^{158}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant 1.0 \times 10^{15}\ \text{yr}.$$

The simulated response of the detector to the $0\nu 2\varepsilon$ decay of ^{158}Dy to the ground state of ^{158}Gd differs from the two neutrino process. We have set the following limit by analyzing the data in the vicinity of the 184.2 keV γ peak expected for the $0\nu 2K$ decay:

$$T_{1/2}^{0\nu 2K}\left(^{158}\text{Dy, g.s.} \to \text{g.s.}\right) \geqslant 4.2 \times 10^{16}\ \text{yr}.$$

Limits on the 2ν and 0ν modes of double electron capture to the first excited level 79.5 keV of ^{158}Gd were obtained by analysis of the data in the energy region around the energy 79.5 keV (see Table 4).

2.5. Search for resonant double electron capture in ^{156}Dy and ^{158}Dy

The double electron capture of ^{156}Dy to the excited levels of ^{156}Gd with energies of 1914.8, 1946.4, 1952.4, 1988.5 and 2003.8 keV may occur with higher probability due to a possible resonant enhancement. The limits on the resonant double electron capture in ^{156}Dy from K and L shells were obtained by analyzing the experimental spectrum in the energy intervals where the most intensive γ peaks from de-excitation of these levels are expected. For instance, we set the following limit on the resonant $2K$ capture in ^{156}Dy to the excited level 2^+ of ^{156}Gd with the energy 1914.8 keV (the dysprosium spectrum was fitted in the vicinity of the γ line 1826.0 keV):

$$T_{1/2}^{(2\nu+0\nu)2\varepsilon}\left(^{156}\text{Dy, g.s.} \to 1914.8\text{ keV}\right) \geqslant 1.1 \times 10^{16}\text{ yr.}$$

To search for a resonant double electron capture in ^{158}Dy from two L_1 shells to the excited level 261.5 keV of ^{158}Gd, the experimental spectrum was fitted in the energy interval 175–185 keV. The fit gives an area of a 181.9 keV peak (-48 ± 49) which corresponds to $\lim S = 40$ counts. The detection efficiency for γ quanta with the energy 181.9 keV irradiated in the 2ε decay is 1.3%, therefore we can set the following limit on the process:

$$T_{1/2}^{(2\nu+0\nu)2\varepsilon}\left(^{158}\text{Dy, g.s.} \to 261.5\text{ keV}\right) \geqslant 3.2 \times 10^{16}\text{ yr.}$$

All the half-life limits on 2β decay processes in dysprosium obtained in the present experiment are summarized in Table 4.

It should be stressed that the difference between the decay energy and the energy of an excited level is one of the most crucial characteristics for the resonant double electron capture. However, in most of the cases the accuracy of atomic mass data is still not enough to point out isotopes with minimal differences $Q_{2\beta} - E_{exc}$. Therefore searches for resonant transitions in different isotopes are strongly motivated.

2.6. Search for α decay of dysprosium isotopes

Five of the seven natural dysprosium isotopes are potentially unstable for α decay. Furthermore for all the isotopes transitions to excited levels of daughter nuclei are allowed. Characteristics of the possible alpha decays are presented in Table 5. The search for α decay of the dysprosium isotopes to the excited levels of the daughter gadolinium isotopes was realized by analyzing the low background energy spectrum measured with the Dy_2O_3 sample.

We do not observe any peculiarities in the experimental data which could be interpreted as γ peaks from α decays of the dysprosium isotopes to the lowest excited levels of the daughter nuclei, and set only limits on these decays. To estimate a limit on the α decay of ^{156}Dy to the excited 2^+ 344.3 keV level of ^{152}Gd, the energy spectrum of the Dy_2O_3 sample was fitted in the energy interval (341–350) keV by a simple model: Gaussian function (to describe γ peak with the energy of 344.3 keV) and polynomial function of the second degree (to approximate background, see Fig. 7). A fit by the chi-square method (χ^2/n.d.f. $= 5.3/5 = 1.1$) gives $S = (-48 \pm 42)$ counts for the area of the peak searched for, which provides no indication for the effect. According to the Feldman–Cousins procedure [19] we took 31 counts as $\lim S$. Taking into account the detection efficiency (2.1%) and the yield of γ quanta from the transitions from the excited level 2^+ 344.3 keV (0.962), we have obtained the following limit on the α decay of ^{156}Dy to the first excited level of ^{152}Gd:

$$T_{1/2}^{\alpha}\left(^{156}\text{Dy, g.s.} \to 344.3\text{ keV}\right) \geqslant 3.8 \times 10^{16}\text{ yr.}$$

Table 5
Characteristics of possible alpha decays of dysprosium isotopes to the lowest excited levels of the daughter nuclei.

Alpha decay Q_α (keV) [9] δ (%) [16]	Excited level of daughter nuclei (keV), yield of γ per α decay	Detection efficiency of the γ quanta	lim S	Experimental limit on $T_{1/2}$ (yr)	Theoretical estimation of $T_{1/2}$ (yr) [24]
^{156}Dy$\rightarrow^{152}$Gd 1759(6) 0.056(3)	2^+, 344.3 0.962 [21]	2.1%	31	$\geqslant 3.8 \times 10^{16}$	2.3×10^{34}
^{158}Dy$\rightarrow^{154}$Gd 879(3) 0.095(3)	2^+, 123.1 0.455 [22]	0.9%	31	$\geqslant 1.3 \times 10^{16}$	1.4×10^{68}
^{160}Dy$\rightarrow^{156}$Gd 439.2(13) 2.329(18)	2^+, 89.0 0.203 [10]	0.4%	230	$\geqslant 8.5 \times 10^{15}$	5.4×10^{126}
^{161}Dy$\rightarrow^{157}$Gd 344.6(12) 18.889(42)	$5/2^-$, 54.5 0.075 [23]	0.2%	83	$\geqslant 3.5 \times 10^{16}$	1.3×10^{147}
^{162}Dy$\rightarrow^{158}$Gd 85.0(12) 25.475(36)	2^+, 79.5 0.142 [11]	0.3%	113	$\geqslant 1.0 \times 10^{17}$	1.7×10^{1414}

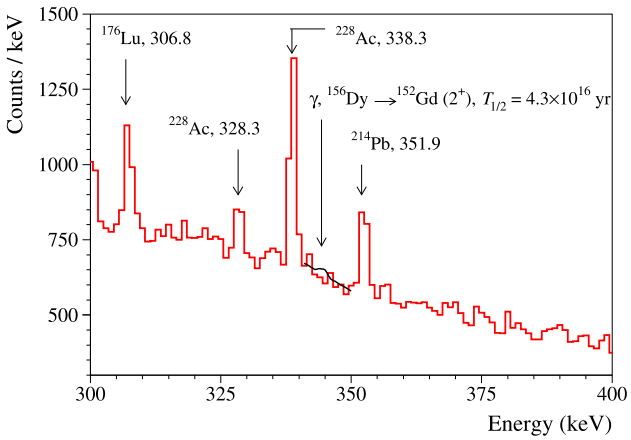

Fig. 7. (Color online.) A part of the energy spectrum of HP Ge spectrometer accumulated with Dy_2O_3 sample over 2512 h. An expected γ peak from α decay of ^{156}Dy to the first excited level of ^{152}Gd corresponding to the half-life $T_{1/2} = 4.3 \times 10^{16}$ yr (excluded at 90% C.L.) is shown by the solid line.

Limits on $T_{1/2}$ relatively to α decay to the lowest excited levels of the daughter nuclei for other dysprosium isotopes were obtained in the same way. The results are presented in Table 5 where theoretical estimations calculated by using the approach proposed in [24] are given too. The theoretical half-lives were found very big. Indeed, the predicted half-life is $T_{1/2} = 2.3 \times 10^{34}$ yr even for the decay with the biggest energy release (^{156}Dy$\rightarrow^{152}$Gd, 2^+, 344.3 keV).

Theoretical estimates give no hope for realistic observation of alpha decays of all naturally abundant dysprosium isotopes to the excited level of daughter nuclei. At the same time

the detection of the ^{156}Dy decay to the ground state of ^{152}Gd (supposing one will apply crystal scintillators from dysprosium enriched in ^{156}Dy) looks possible. Indeed we have estimated the half-life as 5×10^{24} yr by using the approach [24]. The important advantage of scintillation methods is an almost 100% detection efficiency to alpha particles and pulse-shape discrimination ability [25,26]. Development of enrichment methods for dysprosium isotopes and radiopure crystal scintillators containing Dy are requested to observe α activity of natural dysprosium.

3. Conclusions

The first experiment to search for 2β processes in ^{156}Dy and ^{158}Dy was carried out in the underground Gran Sasso National Laboratories of INFN by using ultra-low background HP Ge γ spectrometry. After 2512 h of data taking with a 322 g sample of highly pure dysprosium oxide (Dy_2O_3) limits on double beta processes in ^{156}Dy and ^{158}Dy have been established on the level of 10^{14}–10^{16} yr.

The search for the resonant $0\nu 2\varepsilon$ capture in ^{156}Dy and ^{158}Dy to excited levels of daughter nuclei was of particular interest. Taking into account the strong dependence of the process on the difference between the double beta decay energy and the energy of an excited level, a more accurate determination of the difference in atomic masses of ^{156}Dy–^{156}Gd and ^{158}Dy–^{158}Gd isotopes is strongly required. A precise study of the 1914.8, 1946.4, 1952.4, 1988.5 and 2003.8 keV levels of ^{156}Gd characteristics (spin, parity, decay scheme) is important too.

Theoretical calculations of double beta decay of dysprosium are scarce; in fact, the only known estimations for ^{156}Dy (g.s. to g.s. transitions) are $T_{1/2}^{0\nu\varepsilon\beta^+} = 7.0 \times 10^{27}$ yr (for the effective neutrino mass of 1 eV) [27], $T_{1/2}^{2\nu\varepsilon\beta^+} = (1.1\text{–}2.8) \times 10^{26}$ yr, $T_{1/2}^{2\nu 2\varepsilon} = (1.8\text{–}4.5) \times 10^{23}$ yr [28]. The present study is the first experimental attempt to search for double beta processes in ^{156}Dy and ^{158}Dy, which both are rather interesting isotopes for 2β experiments taking into account a possible resonant double electron capture on excited levels of daughter nuclei.

The sensitivity of the experiment can be advanced to the level of $T_{1/2} \sim 10^{23}$–10^{24} yr by using enriched ^{156}Dy and ^{158}Dy isotopes deeply purified from radioactive elements, and increasing the exposure and detection efficiency by application of multi-crystal HP Ge detectors.

The search for alpha decay of dysprosium isotopes to the excited levels of daughter nuclei was realized as a by-product of the experiment on the level of sensitivity $\lim T_{1/2} \sim 10^{15\text{–}17}$ yr. However, the obtained limits are very far from the theoretical predictions even for the α transition of ^{156}Dy with the highest energy of decay ($T_{1/2} \sim 10^{34}$ yr). At the same time the detection of α decay of ^{156}Dy to the ground state of ^{152}Gd (theoretical estimations of the half-life are on the level of $\sim 10^{24}$ yr) looks reasonably realistic by using crystal scintillators containing dysprosium enriched in ^{156}Dy. Such crystal scintillators could be also used to search for double beta processes in ^{156}Dy.

We have found that the Dy_2O_3 sample contains on the level of several mBq/kg ^{176}Lu, ^{235}U and ^{226}Ra; the activities of ^{228}Ra and ^{228}Th are ≈ 0.18 and ≈ 0.16 Bq/kg, respectively. The results of the γ spectrometry are consistent with the data of the ICP-MS analysis for radionuclides. The contamination of the dysprosium sample can be explained by the production of lanthanides from monazite, an ore with high concentration of uranium and thorium. Presence of radioactive ^{176}Lu can be due to the similar chemical properties of lutetium and dysprosium, which provides some difficulties in chemical separation of the elements.

Acknowledgements

The authors from the Institute for Nuclear Research (Kyiv, Ukraine) were supported in part by the Project "Kosmomikrofizyka-2" (Astroparticle Physics) of the National Academy of Sciences of Ukraine.

References

[1] Yu.G. Zdesenko, Rev. Mod. Phys. 74 (2002) 663;
S.R. Elliot, P. Vogel, Ann. Rev. Nucl. Part. Sci. 52 (2002) 115;
J.D. Vergados, Phys. Rep. 361 (2002) 1;
S.R. Elliot, J. Engel, J. Phys. G 30 (2004) R183;
F.T. Avignone III, G.S. King, Yu.G. Zdesenko, New J. Phys. 7 (2005) 6;
H. Ejiri, J. Phys. Soc. Jpn. 74 (2005) 2101;
H.V. Klapdor-Kleingrothaus, Int. J. Mod. Phys. E 17 (2008) 505;
F.T. Avignone III, S.R. Elliott, J. Engel, Rev. Mod. Phys. 80 (2008) 481.
[2] V.I. Tretyak, Yu.G. Zdesenko, At. Data Nucl. Data Tables 61 (1995) 43;
V.I. Tretyak, Yu.G. Zdesenko, At. Data Nucl. Data Tables 80 (2002) 83.
[3] A.S. Barabash, Phys. Rev. C 81 (2010) 035501.
[4] Ju.M. Gavriljuk, et al., Phys. At. Nucl. 69 (2006) 2124.
[5] P. Belli, et al., Nucl. Phys. A 826 (2009) 256.
[6] N.I. Rukhadze, et al., J. Phys. Conf. Ser. 203 (2010) 012072.
[7] A.P. Meshik, et al., Phys. Rev. C 64 (2001) 035205.
[8] M. Hirsch, et al., Z. Phys. A 151 (1994) 347.
[9] G. Audi, A.H. Wapstra, C. Thibault, Nucl. Phys. A 337 (2003) 729.
[10] C.W. Reich, Nucl. Data Sheets 99 (2003) 753.
[11] R.G. Helmer, Nucl. Data Sheets 101 (2004) 325.
[12] R.G. Winter, Phys. Rev. 100 (1955) 142.
[13] M.B. Voloshin, G.V. Mitselmakher, R.A. Eramzhyan, JETP Lett. 35 (1982) 656.
[14] J. Bernabeu, A. De Rujula, C. Jarlskog, Nucl. Phys. B 223 (1983) 15.
[15] Z. Sujkowski, S. Wycech, Phys. Rev. C 70 (2004) 052501.
[16] M. Berglund, M.E. Wieser, Pure Appl. Chem. 83 (2011) 397.
[17] W.R. Nelson, et al., SLAC-report-265, Stanford, 1985.
[18] O.A. Ponkratenko, et al., Phys. At. Nucl. 1282 (2000) 63;
V.I. Tretyak, in press.
[19] G.J. Feldman, R.D. Cousins, Phys. Rev. D 3873 (1998) 57.
[20] R.B. Firestone, et al., Table of Isotopes, 8th ed., John Wiley, New York, 1996; and CD update, 1998.
[21] A. Artna-Cohen, Nucl. Data Sheets 79 (1996) 1.
[22] C.W. Reich, Nucl. Data Sheets 110 (2009) 2257.
[23] R.G. Helmer, Nucl. Data Sheets 103 (2004) 565.
[24] D.N. Poenaru, et al., J. Phys. 44 (1983) 791.
[25] F.A. Danevich, et al., Phys. Rev. C 67 (2003) 014310.
[26] P. Belli, et al., Nucl. Phys. A 789 (2007) 15.
[27] P.K. Rath, et al., Phys. Rev. C 80 (2009) 044303.
[28] P.K. Rath, et al., J. Phys. G 37 (2010) 055108.