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First search for 2ε and $\varepsilon\beta^+$ decay of ^{162}Er and new limit on $2\beta^-$ decay of ^{170}Er to the first excited level of ^{170}Yb

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Abstract

The first search for double electron capture (2ε) and electron capture with positron emission ($\varepsilon\beta^+$) of ^{162}Er to the ground state and to several excited levels of ^{162}Dy was realized with 326 g of highly purified erbium oxide. The sample was measured over 1934 h by the ultra-low background HP Ge γ spectrometer GeCris (465 cm^3) at the Gran Sasso underground laboratory. No effect was observed, the half-life limits were estimated at the level of $\lim T_{1/2} \sim 10^{15}$–$10^{18}$ yr. A possible resonant $0\nu\, KL_1$ capture in ^{162}Er to the 2^+ 1 782.7 keV excited state of ^{162}Dy is restricted as $T_{1/2} \geqslant 5.0 \times 10^{17}$ yr at 90% C.L. A new improved half-life limit $T_{1/2} \geqslant 4.1 \times 10^{17}$ yr was set on the $2\beta^-$ decay of ^{170}Er to the 2^+ 84.3 keV first excited state of ^{170}Yb.

Keywords: 2β decay, double electron capture, low background experiment, rare events searches

(Some figures may appear in colour only in the online journal)

1. Introduction

The neutrinoless double beta ($0\nu2\beta$) decay is forbidden in the Standard Model of particle physics (SM) since the process violates the lepton number and allows one to investigate if the neutrino is a Majorana particle. Therefore, searches for this decay are considered as an unique way to study the properties of the neutrino and of the weak interaction, to test the lepton number violation, to search for effects beyond the SM [1–6]. While the two neutrino (2ν) mode of $2\beta^-$ decay has been already observed in several nuclei with the half-lives $T_{1/2}^{2\nu2\beta^-} \sim 10^{18}$–$10^{24}$ yr [7–9], the $0\nu2\beta^-$ decay is still under investigation. Even the most sensitive experiments give only half-life limits on the decay at the level of $\lim T_{1/2}^{0\nu2\beta^-} \sim 10^{24}$–$10^{26}$ yr (we refer reader to the reviews [5, 7, 10–13] and the recent original works [14–20]).

The achievements in investigations of the double beta plus processes, such as double electron capture (2ε), electron capture with positron emission ($\varepsilon\beta^+$) and double positron decay ($2\beta^+$) are much more modest [7, 21, 22]. The 'gap' can be explained by the typically very low isotopic abundance of the double beta plus isotopes, that does not exceed 1%, and the suppression of the decay probabilities by small phase space factors. This leads to a much lower sensitivity of the experiments to the effective Majorana neutrino mass. Even the allowed two neutrino double electron capture is not observed surely. There are only indications on the double electron capture in ^{130}Ba [23, 24] and ^{78}Kr [25, 26].

At the same time, the need to develop experimental methods to search for double beta plus processes is supported by the capability to distinguish between two possible mechanisms of the $0\nu2\beta^-$ decay if observed: whether it is due to the light Majorana neutrino mass or due to the right-handed currents' admixture in the weak interaction [27]. Another argument in favor of the neutrinoless double electron capture investigations is the possibility of resonant enhancement of the capture rate due to a mass degeneracy between the initial and final nucleus [28–30].

The isotope ^{164}Er was proposed as a candidate to search for the Majorana neutrino mass in the resonant $0\nu2\varepsilon$ process [31–33]. The $Q_{2\beta}$ value of the neutrinoless double electron capture transitions in ^{164}Er was precisely measured by Penning-trap mass-ratio spectrometry as 25.07(12) keV. The value results in a rather long theoretical prediction for the half-life of $\sim10^{30}$ yr for a 1 eV effective Majorana neutrino mass [34]. Taking into account that the sensitivity of the most recent $0\nu2\beta^-$ experiments is almost one order magnitude higher (e.g., the KamLAND-Zen experiment already reached an effective Majorana neutrino mass sensitivity $\lim\langle m_\nu\rangle \sim 0.1$ eV [15]) the corresponding theoretical half-life for ^{164}Er is on the level of $\sim10^{32}$ yr. Nevertheless, despite the precision mass measurements indicate that the actually most promising double electron capture candidates are ^{152}Gd, ^{156}Dy and ^{190}Pt [34–36], ^{164}Er remains an interesting nucleus in the list of resonant neutrinoless double electron capture candidates. So that was the reason to investigate the radiopurity level of erbium, and to estimate the possibilities of erbium purification from radioactive elements. In addition to ^{164}Er, erbium contains two other potentially double beta active isotopes: the double beta plus isotope, ^{162}Er, and the $2\beta^-$ ^{170}Er. Characteristics of these isotopes are given in table 1.

Unfortunately, the low energy release expected in the double electron capture of ^{164}Er does not allow the search for the decay by using the low background γ spectrometry applied in the present work. Nevertheless, in addition to the radiopurity investigations of the erbium sample, we have used the data of the low background measurements to derive new limits on double beta processes in ^{162}Er and ^{170}Er with emission of 511 keV γ quanta after β^+ annihilation, or γ quanta expected in the de-excitation of daughter nuclei.

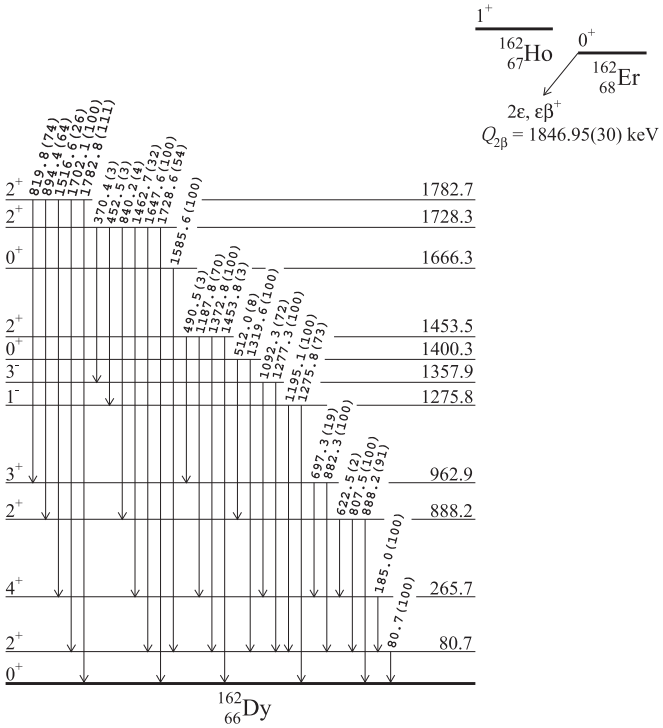

Figure 1. Simplified decay scheme of ^{162}Er [40]. The energies of the excited levels and of the emitted γ quanta are in keV (relative intensities of γ quanta, rounded to percent, are given in parentheses; only the γ transitions with the relative intensity of more than 2% are shown).

Table 1. Characteristics of 2β isotopes of erbium.

2β transition	$Q_{2\beta}$ (keV)	Isotopic abundance (%) [39]	Decay channel
^{162}Er $\rightarrow$ ^{162}Dy	1 846.95(30) [37]	0.139(5)	2ε, $\varepsilon\beta^+$
^{164}Er $\rightarrow$ ^{164}Dy	25.07(12) [34]	1.601(3)	2ε
^{170}Er $\rightarrow$ ^{170}Yb	655.2(15) [38]	14.910(36)	$2\beta^-$

A simplified scheme of the double beta decay of ^{162}Er is presented in figure 1 (in the daughter ^{162}Dy only the γ transitions with relative intensities of more than 2% are shown).

The double beta decay of ^{170}Er is possible to the ground and to the first 2^+ excited level of ^{170}Yb with energy 84.3 keV (see figure 2).

2. Experiment

2.1. Purification of erbium oxide

The erbium oxide (Er_2O_3) for the experiment was provided by the Stanford Materials Corporation. The purity level of the material was 99.5% (TREO, total rare earth oxides) and 99.999% (Er_2O_3/TREO). The compound was examined by Inductively Coupled Plasma

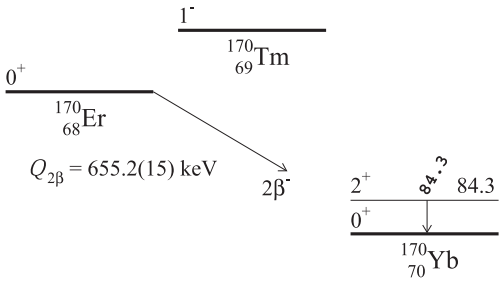

Figure 2. Simplified decay scheme of ^{170}Er [41].

Table 2. Contamination of the erbium oxide sample measured by ICP-MS before and after the purification, as well as contamination of the Er_2O_3 sediment after the fractional precipitation stage of the purification (see text). Errors on the measured values are at the level of 30% since the ICP-MS measurements have been carried out in semi-quantitative mode.

Element	Concentration (ppb)		
	Initial material before purification	Sediment after fractional precipitation	After purification by the liquid–liquid extraction method
K	<2000	424	283
Fe	—	1190	1797
Pb	10000	56	322
La	100	16	17
Lu	80	144	99
Th	1.6	13	<0.3
U	1.9	<0.2	<0.2

Mass Spectrometer (ICP-MS, model Element II from Thermo Fisher Scientific, Waltham, Massachusetts, USA). In order to overcome the drawback related to the well known isobaric interferences, K and Fe were measured, respectively, in high resolution (HR) and medium resolution mode. All the other elements have been analyzed in low resolution (LR) setting (see table 2). The initial contamination of the material by radioactive lanthanide elements is in agreement with the producer specification (concentrations of La and Lu less than 0.1 ppm). Thorium and uranium were also in the material as 1.6 ppb and 1.9 ppb, respectively.

The radioactive contamination of a 517 g sample of the erbium oxide was measured over 340 h with the p-type ultra-low background high purity germanium (HPGe) γ spectrometer GePV with an active volume of 363 cm^3 at the STELLA facility of the Gran Sasso underground laboratory of the INFN (Italy). The energy resolution of the detector is FWHM = 1.8 keV for 1333 keV γ quanta of ^{60}Co, the relative efficiency is 91% [44]. The detection efficiencies to γ quanta emitted in the decay of the radioactive contamination nuclides were calculated with the GEANT4 simulation package [42, 43] with initial kinematics given by the DECAY0 event generator [45, 46]. The results of the measurements (denoted as 'before purification') are presented in table 3. It should be noted that the low sensitivity of the measurements before purification (particularly to ^{234}Th) is due to a much lower detection efficiency to low energy γ quanta and higher background counting rate of the

Table 3. Radioactive contamination of the erbium oxide sample before and after the purification, measured with the help of an ultra-low background HP Ge γ spectrometer. The upper limits are presented at 90% C.L., the uncertainties are given at ≈68% C.L.

Chain	Nuclide	Activity (mBq kg^{-1})	
		Before purification	After purification
	^{40}K	⩽27	⩽1.7
	^{137}Cs	⩽2.1	1.4 ± 0.3
	^{176}Lu	6 ± 1	4.2 ± 0.4
^{232}Th	^{228}Ra	⩽7.2	⩽1.0
	^{228}Th	5 ± 2	⩽1.1
^{235}U	^{235}U	⩽12	⩽1.6
^{238}U	^{226}Ra	6 ± 2	1.1 ± 0.4
	^{234}Th	⩽1800	⩽91
	^{234m}Pa	⩽74	⩽16

GePV detector. E.g., presence of ^{234}Th in the sample was estimated by searching for γ quanta with energies 92.4 keV and 92.8 keV.

Traces of lutetium (^{176}Lu), radium (^{226}Ra) and thorium (^{228}Th) were detected in the sample with activities at the level[9] of 5–6 mBq kg^{-1}. Therefore, an additional purification of the material was decided.

The following scheme of purification procedure was applied to the Er_2O_3 purification: (1) dissolving of Er_2O_3; (2) fractional precipitation of $Er(OH)_3$ sediment; (3) liquid–liquid extraction; (4) precipitation of $Er(OH)_3$; and (5) final recovery of Er_2O_3.

As a first step, diluted nitric acid was added to a suspension of Er_2O_3 in deionized water to obtain a homogeneous aqueous solution of erbium. The initial amounts of water and nitric acid were calculated to obtain an acidic solution with a concentration of Er^{3+} at the level of 1.5 mol l^{-1}.

The fractional precipitation of erbium from the acidic solution was used to co-precipitate impurities like Th, taking into account that hydroxides of thorium are precipitated at a lower pH level than erbium. Ammonia gas has been injected into the solution till the pH reached 6.5 that led to the fractional precipitation of erbium hydroxide. Then, the amorphous $Er(OH)_3$ sediment was separated from the supernatant liquid using a centrifuge, and was annealed to Er_2O_3. The 43 g of erbium oxide were obtained, which is 8.3% of the initial mass. The oxide was analyzed by the ICP-MS to check the efficiency of the co-precipitation of the impurities (see table 2, 'sediment after fractional precipitation'). The contamination of the sediment testifies the efficiency of the purification process. For instance, the concentration of Th increased in the sediment by a factor of ≈8. However, it is a rather minor figure taking into account the requirements of the double beta experiments aiming at the achievement of an as low as possible level of background that is determined by the radioactive contamination of the sample. Therefore, the liquid–liquid extraction method was applied for further purification of the material. The sediment was excluded from the further purification process by the liquid–liquid extraction method since it accumulated impurities of the initial material.

[9] We would like to emphasize that the results of the ICP-MS and γ spectrometry on thorium and lutetium are in a good agreement (1.6 ppb of Th corresponds to an activity of ^{228}Th 6 mBq kg^{-1}, assuming the equilibrium of the ^{232}Th chain; 80 ppb of Lu corresponds to 4 mBq kg^{-1} of ^{176}Lu).

The liquid–liquid extraction method [47] proved to be the most effective one for the purification of lanthanides solutions from traces of uranium and thorium. To apply liquid–liquid extraction to the erbium solution, it was acidified with diluted nitric acid to pH = 1. Tri-n-octylphosphine oxide (TOPO) has been utilized as 'soft' organic complexing agent for binding U and Th, while toluene was used as liquor solvent. Considering the chemically very low concentration of the traces in the solution, the concentration of TOPO in toluene did not exceed $0.1\,\mathrm{mol\,l^{-1}}$. The two immiscible liquids (aqueous solution and organic solution) were placed in a separation funnel in the volumetric ratio of 1:1 and shaken for a few minutes. The uranium and thorium interact with TOPO forming organo-metallic complexes that have much higher solubility in organic phase than in water solution. This leads to the extraction of U and Th into the organic liquid. After the separation of the purified aqueous solution, the erbium was completely precipitated in form of hydroxide using ammonia. The impurities as alkali and alkali-earth cations were left in the supernatant liquid. Sediments were separated, dried and annealed at 900 °C for a few hours. Finally, 398 g of purified material were obtained, that is 77% of the initial material. The purified material was analyzed by the ICP-MS as reported in table 2.

2.2. Low counting experiment

The experiment was carried out at the STELLA facility by using the ultra-low background HPGe detector GeCris with a volume of 465 $\mathrm{cm^3}$. The detector is shielded by low radioactive lead (≈25 cm), copper (≈5 cm), and in the innermost part by archaeological Roman lead (≈2.5 cm). The set-up is enclosed in an air-tight poly(methyl methacrylate) box and flushed with high purity nitrogen gas to reduce the background from the environmental radon concentration. The energy resolution of the detector was estimated by using intensive background γ peaks with energies 238.6 keV (^{212}Pb), 338.7 keV (^{228}Ac), 463.0 keV (^{228}Ac), 583.2 keV (^{208}Tl), 661.7 (^{137}Cs), 727.3 keV (^{212}Bi), 911.2 keV (^{228}Ac), 1460.8 keV (^{40}K) and 2614.5 keV (^{208}Tl) in the data measured with the cerium oxide sample in the experiment [48]. It depends on the energy E_γ of the γ quanta as $\mathrm{FWHM(keV)} = \sqrt{1.41 + 0.001\,97 \times E_\gamma}$, where E_γ is in keV. A sample of the purified Er_2O_3 with mass 326 g, enclosed in a cylindric polystyrene box, was placed on the HP Ge detector end cap. The sample contained 1.43×10^{21} and 1.532×10^{23} nuclei of ^{162}Er and ^{170}Er, respectively. The data with the sample were accumulated over 1934 h, while the background spectrum was taken over 1046 h. The two spectra, normalized for their time of measurements, are presented in figure 3.

Some excess (in comparison to the background data) of ^{137}Cs, ^{176}Lu and ^{214}Bi (daughter of ^{226}Ra) was observed; this allowed the estimate of the residual contamination of the sample by these radionuclides. The activities of the nuclides in the Er_2O_3 sample after the purification are presented in table 3. The contamination by ^{176}Lu remained almost the same as before the purification due to the high chemical affinity between Er and Lu, while the activity of ^{226}Ra decreased by a factor 5.

2.3. Search for 2ε and εβ⁺ processes in ^{162}Er

There are no peculiarities in the energy spectrum accumulated with the Er_2O_3 sample that could be identified as double beta decay of the erbium isotopes. Therefore, the data were analyzed to estimate half-life limits for the 2ε and $\varepsilon\beta^+$ decay of ^{162}Er, and the 2β decay of ^{170}Er. Lower half-life limits were estimated with the help of the following equation:

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

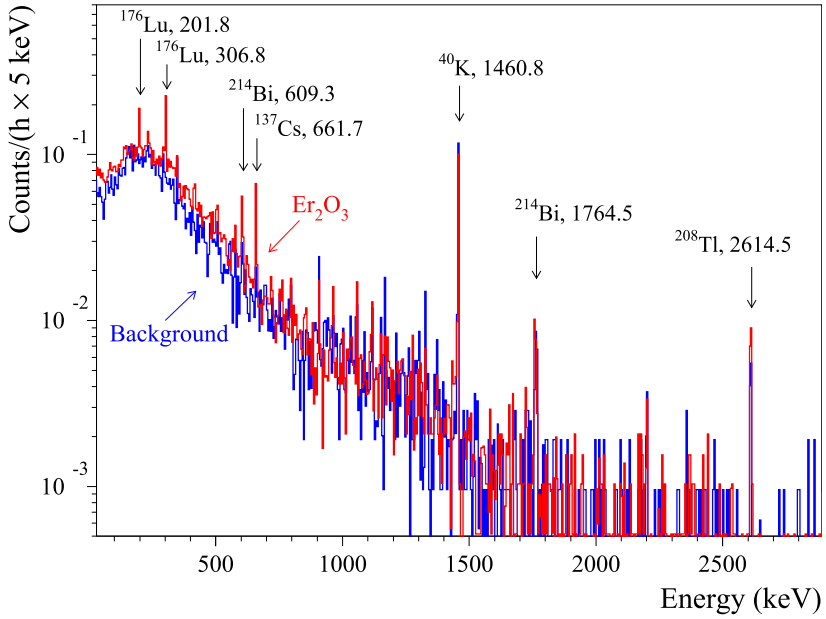

Figure 3. Energy spectra measured with the ultra-low background HPGe γ spectrometer with the purified Er_2O_3 sample over 1934 h (Er_2O_3) and without sample over 1046 h (Background). The energies of the γ peaks are in keV.

where N is the number of nuclei of interest in the sample, η is the detection efficiency (the yields of the γ quanta expected in the double beta processes are included), t is the time of measurement, and $\lim S$ is the upper limit on the number of events of the effect searched for that can be excluded at a given confidence level (C.L.). In the present work all the $\lim S$ and, therefore, the half-life limits are estimated at 90% C.L. The detection efficiencies to the effects searched for were simulated by Monte Carlo code using EGSnrc [49] package[10]. with initial kinematics given by the DECAY0 event generator [45, 46].

In the case of the $2\nu 2K$ capture in ^{162}Er, a cascade of x-rays and Auger electrons with energies up to 53.8 keV is expected. We took into account only the most intense x-rays of dysprosium [50]: 45.2 keV (the yield of the x-rays quanta is 26.8%), 46.0 keV (47.5%), 51.9 keV (4.9%), 52.1 keV (9.6%), and 53.5 keV (3.2%). The energy spectrum accumulated with the Er_2O_3 sample was fitted by the sum of five Gaussian functions ($2\nu 2K$ decay of ^{162}Er), a peak of ^{210}Pb with energy 46.5 keV, and a straight line to describe the continuous background. The best fit was achieved in the energy interval (35–63) keV with χ^2/n.d.f. $\simeq$0.59, where n.d.f. is number of degrees of freedom. The fit provides the area of the $2\nu 2K$ effect: (-6 ± 10) counts. Taking into account the recommendations given in [51], we took 11 counts as $\lim S$. The energy spectrum in the vicinity of the $2\nu 2K$ effect, the approximation by the model of background and the excluded effect are presented in figure 4. In this case the detection efficiency of the whole effect is calculated as: $\eta = \Sigma_i \eta_i$, where η_i are the efficiencies for the x-ray quanta. The detection efficiency was simulated by Monte Carlo code as $\eta = 0.016\%$. Taking into account the number of ^{162}Er nuclei in the sample, one can obtain

[10] It should be stressed that the calculations of the detection efficiencies with the help of the GEANT4 simulation package give similar results with a deviation of less than 16% in the worst case of the $2\nu 2K$ decay of ^{162}Er.

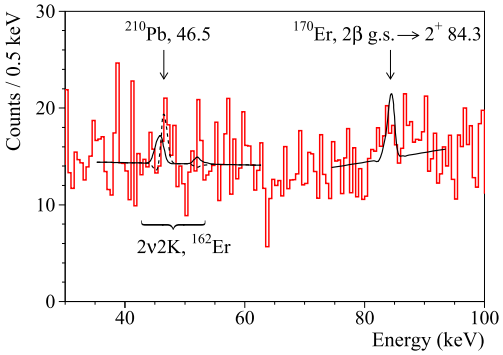

Figure 4. Low energy part of the spectrum accumulated with the Er_2O_3 sample over 1934 h. The approximation function (dashed line) and the excluded effect of $2\nu 2K$ decay of ^{162}Er (solid line) are shown. The excluded peak of 2β decay of ^{170}Er to the excited 2^+ level of ^{170}Yb with the energy 84.3 keV is also shown. The energies of the peaks are in keV.

Table 4. Half-life limits on 2β processes in ^{162}Er and ^{170}Er.

Process of decay	Decay mode	Level of daughter nucleus (keV)	E_γ (keV)	Detection efficiency (%)	lim S	Experimental limit (yr) at 90% C.L.
^{162}Er → ^{162}Dy						
$2K$	2ν	g.s.	45−53	0.016	11	$\geqslant 3.2 \times 10^{15}$
2ε	2ν	2^+ 80.7	80.7	0.014	2.6	$\geqslant 1.2 \times 10^{16}$
2ε	2ν	2^+ 888.2	888.2	1.25	6.5	$\geqslant 4.2 \times 10^{17}$
2ε	2ν	0^+ 1 400.3	1 319.6	2.03	3.3	$\geqslant 1.3 \times 10^{18}$
2ε	2ν	2^+ 1 453.5	1 187.8	0.86	6.0	$\geqslant 3.1 \times 10^{17}$
2ε	2ν	0^+ 1 666.3	1 585.6	1.96	5.6	$\geqslant 7.7 \times 10^{17}$
2ε	2ν	2^+ 1 728.3	1 647.6	0.99	2.3	$\geqslant 9.4 \times 10^{17}$
KL	2ν	2^+ 1 782.7	1 702.1	0.53	2.3	$\geqslant 5.0 \times 10^{17}$
$2K$	0ν	g.s.	1 739.1−1 739.7	1.87	3.9	$\geqslant 1.0 \times 10^{18}$
KL	0ν	g.s.	1 783.8−1 785.7	1.84	4.2	$\geqslant 9.6 \times 10^{17}$
$2L$	0ν	g.s.	1 828.6−1 831.7	1.82	3.1	$\geqslant 1.3 \times 10^{18}$
$2K$	0ν	2^+ 80.7	1 658.7	1.93	6.8	$\geqslant 6.2 \times 10^{17}$
$2K$	0ν	2^+ 888.2	851.2	2.38	8.8	$\geqslant 5.9 \times 10^{17}$
$2K$	0ν	0^+ 1 400.3	339.1	3.04	5.1	$\geqslant 1.3 \times 10^{18}$
$2K$	0ν	2^+ 1 453.5	285.9	2.86	6.9	$\geqslant 9.1 \times 10^{17}$
$2K$	0ν	0^+ 1 666.3	1 585.6	1.98	5.6	$\geqslant 7.7 \times 10^{17}$
$2K$	0ν	2^+ 1 728.3	1 647.6	0.98	2.3	$\geqslant 9.3 \times 10^{17}$
Resonant KL_1	0ν	2^+ 1 782.7	1 702.1	0.53	2.3	$\geqslant 5.0 \times 10^{17}$
$\varepsilon\beta^+$	2ν	g.s.	511	6.48	37	$\geqslant 3.8 \times 10^{17}$
$\varepsilon\beta^+$	2ν	2^+ 80.7	511	6.48	37	$\geqslant 3.8 \times 10^{17}$
$\varepsilon\beta^+$	0ν	g.s.	511	6.29	37	$\geqslant 3.7 \times 10^{17}$
$\varepsilon\beta^+$	0ν	2^+ 80.7	511	6.29	37	$\geqslant 3.7 \times 10^{17}$
^{170}Er → ^{170}Yb						
$2\beta^-$	$2\nu + 0\nu$	2^+ 84.3	84.3	0.017	9.6	$\geqslant 4.1 \times 10^{17}$

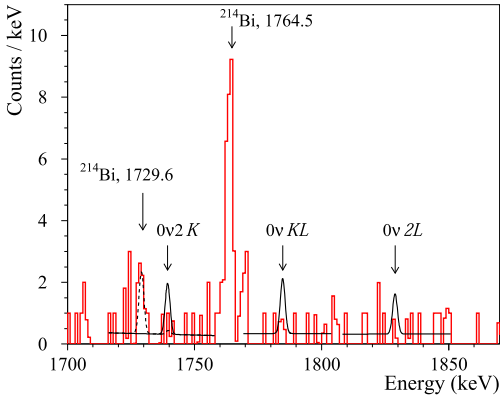

Figure 5. Part of the energy spectrum accumulated with the Er_2O_3 sample over 1934 h, where the γ peaks from the $0\nu 2K$, $0\nu KL$, and $0\nu 2L$ captures in ^{162}Er to the ground state of ^{162}Dy are expected. The excluded peaks at 90% C.L. are shown by solid lines. The fit in the energy interval (1716−1758) keV, that includes also the γ peak of ^{214}Bi with energy 1 729.6 keV, is shown by a dashed line. The energies of the peaks are in keV.

the half-life limit on the $2\nu 2K$ capture in ^{162}Er presented in table 4[11] . In the 0ν double electron capture in ^{162}Er (we consider here only capture from K and L shells) to the ground state of ^{162}Dy, we assume the energies of the γ quanta to be equal to $E_\gamma = Q_{2\beta} - E_{b1} - E_{b2}$, where E_{bi} are the binding energies of the captured electrons on the atomic shells of the daughter dysprosium atom. The energy spectrum accumulated with the Er_2O_3 sample was fitted by the sum of a Gaussian function (to describe the peak expected) and of a polynomial function of the first degree (to describe the background). The spectrum in the vicinity of the expected peaks is shown in figure 5. In the case of the $0\nu 2K$ decay of ^{162}Er, also the peak of ^{214}Bi with energy 1 729.6 keV was included in the fit to approximate the background in a wide enough energy interval around the peak searched for. The fit gives an area of expected $0\nu 2K$ peak with energy 1 739.4 keV $S = (0.3 \pm 0.9)$ counts, that corresponds to $\lim S = 1.8$ counts according to the recommendations [51]. However, we have used another, a more conservative approach (also recommended in [51] for experimental sensitivity estimated for expected background and no true signal). Taking into account that there are 2 counts in the energy interval of the expected peak with energy 1 739.4 keV, one should accept $\lim S = 3.9$ counts (see table XII in [51]). To estimate $\lim S$ for an expected $0\nu KL$ ($0\nu 2L$) peak with energy 1 784.7 keV (1 828.9 keV) we have utilized the recommendations [51] for measured mean of a Gaussian and its sigma. The fit gives an area of the peak $S = (1.7 \pm 1.5)$ (1.1 ± 1.2) counts that corresponds to $\lim S = 4.2$ ($\lim S = 3.1$) counts. The excluded peaks of the $0\nu 2K$, $0\nu KL$, and $0\nu 2L$ captures in ^{162}Er to the ground state of ^{162}Dy are shown in figure 5. The obtained half-life limits are given in table 4.

[11] It should be stressed that a possible effect of systematic errors on the obtained limit is rather weak. E.g., the error of the efficiency calculations (16%, the highest one in the case of the two neutrino double K capture, estimated from the difference between the simulations by using the EGSnrc and GEANT4 codes), is negligible in comparison to the statistical fluctuations of the excluded peak and its sigma. The above consideration is even more valid for all other limits reported below since the difference between the EGSnrc and GEANT4 detection efficiencies is smaller for all other double beta decay modes and channels analyzed in the present study. The contribution of other possible systematic errors, e.g., of the energy calibration and resolution uncertainties, are even smaller.

The double electron capture in ^{162}Er is also allowed to excited levels of ^{162}Dy with subsequent emission of gamma quanta that can be detected by the HP Ge spectrometer. In the 2ε process, the 2ν and 0ν modes cannot be distinguished[12]. However, the detection efficiencies for the decays are slightly different. The difference is due to emission of additional γ quanta in the 0ν process with energy $E_\gamma = Q_{2\beta} - 2E_K - E_{\rm exc}$, where $E_{\rm exc}$ is energy of the excited level of ^{162}Dy, and E_K is the binding energy of the captured electrons on the K atomic shell of the daughter dysprosium atom. The emission of the γ quanta will result in a small difference in the obtained half-life limits[13]. To estimate limits on the $2\nu2\varepsilon$ and $0\nu2K$ decays of ^{162}Er to the 0^+ and 2^+ excited levels of ^{162}Dy (see figure 1), the energy spectrum accumulated with the Er_2O_3 sample was fitted in the energy intervals where intense γ peaks from the de-excitation process are expected. The obtained limits for the double electron capture of ^{162}Er to the excited levels of ^{162}Dy are presented in table 4.

The $0\nu2\varepsilon$ capture in ^{162}Er to the 2^+ excited level of ^{162}Dy with the energy $E_\gamma = 1\,782.7$ keV could be much faster due to a resonant enhancement of the capture rate. However, the recent high precise measurements of the ^{162}Er $Q_{2\beta}$ value by the Penning-trap mass-ratio method have shown that the difference $Q_{2\beta} - E_K - E_L - E_{\rm exc} = 2.7$ keV is too big to result in a substantial resonant enhancement of the decay probability [37]. Nevertheless, we have estimated a limit on the $0\nu2\varepsilon$ decay of ^{162}Er to the 2^+ 1 782.7 keV excited level of ^{162}Dy as $T_{1/2} \geqslant 5.0 \times 10^{17}$ yr.

One positron can be emitted in the $2\nu\varepsilon\beta^+$ ($0\nu\varepsilon\beta^+$) decay of ^{162}Er with an energy up to $\approx$825 keV (depending on the binding energy of the atomic shell of the daughter atom). The annihilation of the positron should produce two 511 keV γ quanta resulting in an extra counting rate in the annihilation peak. A similar signature (annihilation γ quanta with energy 511 keV) is expected also in the case of $\varepsilon\beta^+$ decay of ^{162}Er to the first 2^+ 80.7 keV excited level of ^{162}Dy. To estimate $\lim S$ for the decay, the energy spectra accumulated with the Er_2O_3 sample and the background data were fitted in the energy interval (485–535) keV (see figure 6). There are (-6 ± 26) events in the 511 keV peak in the data accumulated with the erbium oxide sample (taking into account the area of the annihilation peak in the background). Since there is no evidence of the effect searched for, we took $\lim S = 37$ counts and set the limits $T_{1/2}^{\varepsilon\beta^+} \geqslant 3.8 \times 10^{17}$ yr (3.7×10^{17} yr) on two neutrino (neutrinoless) $\varepsilon\beta^+$ decay of ^{162}Er to the ground state and the first 2^+ 80.7 keV excited state of ^{162}Dy[14].

2.4. Search for $2\beta^-$ decay of ^{170}Er to the first 2^+ 84.3 keV excited level of ^{170}Yb

The double beta decay of ^{170}Er is possible to the ground state and to the first 2^+ excited level of ^{170}Yb with energy 84.3 keV. In our experiment only the transition to the excited level could be detected. The energy spectrum acquired with the erbium oxide sample was fitted in the energy interval (74–94) keV by a model consisting of a Gaussian function centered at 84.3 keV (to describe the effect searched for) and a straight line as a background model. The fit (see figure 4) gives an area of the 84.3 keV peak of (4.7 ± 3.0) counts; there is no evidence for the effect searched for. Therefore, according to [51] we took $\lim S = 9.6$ counts. Taking into account the number of ^{170}Er nuclei in the sample, and the detection efficiency $\eta = 0.017\%$, we have set the limit on the 2β decay of ^{170}Er to the first 2^+ excited level of

[12] In the present study we consider only $0\nu2K$ transitions to the excited levels expected to be the dominant channels of the decays.

[13] In some cases the $0\nu2K$ limits are substantially stronger due to expected intense γ quanta with energy $E_\gamma = Q_{2\beta} - 2E_K - E_{\rm exc}$.

[14] It should be noted that the half-life limits obtained by analysis of possible 80.7 keV peak are substantially weaker due to the much lower detection efficiencies on the level of $\sim$0.01%.

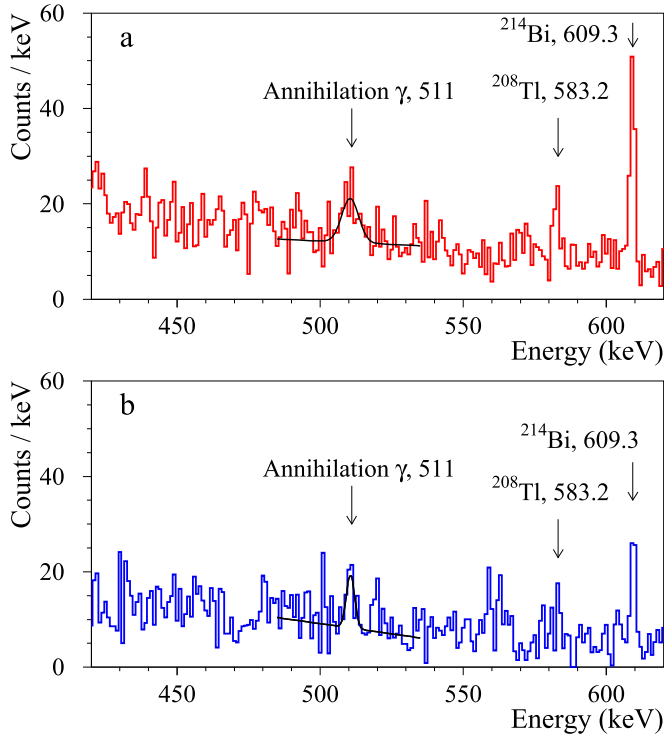

Figure 6. Energy spectra measured with the erbium oxide sample over 1934 h (a) and background over 1046 h (b) in the vicinity of the 511 keV annihilation peak.

^{170}Yb: $T^{2\beta^-}_{1/2} \geqslant 4.1 \times 10^{17}$ yr. The limit is for the sum of the 2ν and 0ν modes, since they cannot be distinguished with the γ-spectrometry method. The limit is slightly stronger than the one ($T^{2\beta^-}_{1/2} \geqslant 3.2 \times 10^{17}$ yr) obtained in the experiment [52] with a similar technique[15].

3. Conclusions

The double electron capture and the electron capture with positron emission in ^{162}Er, and the double beta decay of ^{170}Er to the first 2^+ excited level of ^{170}Yb were searched for in a highly purified 326 g Er_2O_3 sample using ultra-low background HP Ge γ spectrometer with volume of 465 cm^3 at the STELLA facility of the Gran Sasso underground laboratory. For the first time, limits on different modes and channels of double beta decay of ^{162}Er were set at the level of $T_{1/2} > 10^{15}$–10^{18} yr. A possible resonant neutrinoless double electron capture in ^{162}Er to the 2^+ 1 782.7 keV excited level of ^{162}Dy was restricted at the level of $T_{1/2} \geqslant 5.0 \times 10^{17}$ yr. The sensitivity is a few orders of magnitude weaker in comparison to the most sensitive 'double beta plus' experiments that already reached a level of $\lim T_{1/2} \sim 10^{21}$–$10^{22}$ yr with ^{36}Ar [53], ^{40}Ca [54], ^{58}Ni [55], ^{64}Zn [56], ^{78}Kr [25], ^{96}Ru [57], ^{106}Cd [58], ^{112}Sn [59], ^{120}Te [60], ^{124}Xe [62], ^{126}Xe [61, 62], ^{130}Ba [23, 24] and ^{132}Ba [23]. A new improved half-life limit $T_{1/2} \geqslant 4.1 \times 10^{17}$ yr was set on the $2\beta^-$ decay ($2\nu + 0\nu$ modes) of ^{170}Er to the first 2^+ 84.3 keV excited state of ^{170}Yb. A typical sensitivity to the 2β decays to 2^+ excited levels of

[15] It should be noted, that the result in [52] is given with 68% C.L. while the present limit is estimated at 90% C.L.

daughter nuclei is on the level of lim $T_{1/2} \sim 10^{21}$–10^{25} yr [63]. It should be stressed that the $2\nu2\beta$ decay to the first excited 0^+ levels of daughter nuclei is observed in ^{100}Mo and ^{150}Nd with $T_{1/2} \sim 10^{20}$–10^{21} yr [63].

A method of erbium purification from radioactive contamination based on the liquid–liquid extraction was developed. The obtained purified material is quite radiopure (as for other lanthanide elements that are typically contaminated by U and Th). Traces of ^{176}Lu, ^{137}Cs and ^{226}Ra were observed in the purified Er_2O_3 at the $\sim$(1–4) mBq kg^{-1} level, while other contaminations, in particular ^{40}K and ^{228}Th, are below the measurement's sensitivity of $\sim$1 mBq kg^{-1}.

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