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First limits on neutrinoless resonant 2ε captures in ^{136}Ce and new limits for other 2β processes in ^{136}Ce and ^{138}Ce isotopes

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Abstract

A small CeCl$_3$ crystal (6.9 g) was measured deep underground (3600 m w.e.) at the Gran Sasso National Laboratories of the INFN (Italy) with a low background HP Ge detector (244 cm^3) during 1280 h. The crystal is not polluted by ^{232}Th, ^{40}K, ^{60}Co, ^{137}Cs (limits were set on the activity at a level of 0.05–1 Bq/kg), but contains radioactive ^{138}La (0.68 Bq/kg) and, probably, is polluted by ^{235}U and ^{238}U (0.3–0.7 Bq/kg). The half life limits for different double β processes in ^{136}Ce and in ^{138}Ce were set in the range of $(1–6) \times 10^{15}$ yr. In particular, the 2ε captures in ^{136}Ce to various excited levels of ^{136}Ba were considered for the first time. The limits for the resonant neutrinoless transitions to the 2392.1 keV and 2399.9 keV levels of ^{136}Ba are: $T_{1/2} > 2.4 \times 10^{15}$ yr and $T_{1/2} > 4.1 \times 10^{15}$ yr at 90% C.L., respectively.
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1. Introduction

In recent years scintillating materials, which contain Ce, have attracted much attention in the searches for new fast scintillators with high density, large light yield, perfect linearity and good energy resolution for the use in fundamental and applied nuclear and particle physics, and in numerous applications in medicine and industry. This is related with the fact that the Ce^{3+} ions yield fast (10–100 ns) scintillation in the wavelength range of 300–500 nm due to electric dipole allowed 5d–4f transitions in the atomic shell [1,2]. Many Ce doped scintillating crystals (generally of modest size) were investigated; among them, we can mention:

– $LaCl_3$(Ce) with decay time of $\tau = 24$ ns for main decay component and light yield LY = 50 000 photons/MeV (to be compared with classical NaI(Tl) scintillator with $\tau = 230$ ns and LY = 48 000 photons/MeV, or CsI(Tl) with $\tau = 800$ ns and LY = 66 000 photons/MeV) [2];
– $LaBr_3$(Ce) with $\tau = 16$ ns and LY = 70 000 photons/MeV [2]; excellent energy resolution (FWHM) of 2.6% at 662 keV energy of ^{137}Cs was measured in comparison with typical 6.7% for NaI(Tl) [3];
– LuI_3(Ce) with $\tau = 24$ ns and LY = 95 000 photons/MeV [2];
– Lu_2SiO_5(Ce) with $\tau = 42$ ns and LY = 30 000 photons/MeV [2];
– GdI_3(Ce) with $\tau = 39$ ns and LY = 58 000 photons/MeV [4];
– $PrBr_3$(Ce) with $\tau < 10$ ns and LY = 21 000 photons/MeV [5].

Scintillating crystals where Ce is not a dopant but a main constituent were also studied. Measurements of $CeBr_3$ gave $\tau = 17$ ns and LY = 88 000 photons/MeV [6]. CeF_3 scintillator ($\tau = 5$ ns and $\tau = 31$ ns for two main components; LY = 4000 photons/MeV [1]) was intensively investigated in many works, in particular, by the Crystal Clear Collaboration (see also e.g. [7] and references therein).

$CeCl_3$ is another perspective scintillator which contains Ce as a main constituent. However, experimental information on its scintillation features is rather scarce. The density of $CeCl_3$ is 3.9 g/cm^3. The melting point is 817 °C. It is highly hygroscopic. The thermal and magnetic properties were studied in [8]. The electronic and optical properties were examined in [9,10]; in the latter work a refraction index near 2.2 was calculated at 360 nm (experimental value is unknown). To our knowledge, scintillation characteristics were measured only in one work [11] (see also [12]) where few components of scintillation signal were measured with decaying times 4.4 ns (6.6%), 23.2 ns (69.6%), 70 ns (7.5%), and > 10 µs (16.3%); total light output was 28 000 photons/MeV (i.e. much higher than that of much more intensively investigated CeF_3). Maximal emission wavelength is 360 nm.

Intrinsic radioactivity is one of the important characteristics of scintillators. While it is possible to decrease contributions from nuclides of the natural U/Th chains, of ^{40}K, of ^{60}Co, of ^{137}Cs and of other "usual" radioactive contaminants by the purification of all the materials and environmental conditions used for the crystal growth, some other contributions are unavoidable if they are related with natural radioactive isotopes of one of the elements which constitutes the crystal.

For example, La containing crystals include unstable ^{138}La (β^- decay with 33.6% and EC with 66.4%); its half life $T_{1/2} = 1.05 \times 10^{11}$ yr [13] and natural abundance $\delta = 0.09\%$ [14] result in activity of $A = 0.82$ Bq per 1 g of ^{nat}La. Internal radioactivity of $LaCl_3$ scintillators was investigated e.g. in [15,16].

Alpha emitting ^{144}Nd ($T_{1/2} = 2.29 \times 10^{15}$ yr) is present with natural abundance $\delta = 23.8\%$; the corresponding activity is $A = 9.5$ mBq per 1 g of ^{nat}Nd.

Natural Sm contains two α radioactive isotopes: ^{147}Sm ($T_{1/2} = 1.06 \times 10^{11}$ yr, $\delta = 14.99\%$, $A = 124.4$ Bq/g) and ^{148}Sm ($T_{1/2} = 7 \times 10^{15}$ yr, $\delta = 11.24\%$, $A = 1.4$ mBq/g).

Alpha unstable ^{152}Gd ($T_{1/2} = 1.08 \times 10^{14}$ yr) is present in natural Gd with $\delta = 0.2\%$; the corresponding activity is $A = 1.6$ mBq per 1 g of ^{nat}Gd. Such a value seems rather small; nevertheless it gave the most prominent peak observed in measurements with a Gd_2SiO_5 scintillator [17].

Presence of β^- radioactive ^{176}Lu ($T_{1/2} = 3.78 \times 10^{10}$ yr) in natural Lu ($\delta = 2.59\%$) gives an activity of $A = 51.8$ Bq per 1 g of ^{nat}Lu. Internal radioactivity of $Lu_3Al_5O_{12}$ scintillators was measured in [18].

Natural Eu consists of only 2 stable isotopes: ^{151}Eu and ^{153}Eu. However, both of them have quite big cross sections for thermal neutron capture (5900 b and 312 b, respectively). Capture of neutron will result in radioactive ^{152}Eu ($T_{1/2} = 13.537$ yr) and ^{154}Eu ($T_{1/2} = 8.593$ yr); their activities depend on the history of the materials used for the production of a crystal. This radioactivity is unavoidable for any scintillator which contains Eu as a main constituent or as a dopant; for $Li_6Eu(BO_3)_3$ crystals, contamination by ^{152}Eu and ^{154}Eu was studied in [19] and for CaF_2(Eu) in [20].

In addition it should be noted that, because of the similarity in the chemical properties, all the above mentioned radioactive isotopes could be present – at some level – in any lanthanide based scintillator.

The measurements of the Ce containing samples permit a search for double beta (2β) decays of Ce isotopes. The two neutrino (2ν) double beta decay is a process of transformation of atomic nucleus with simultaneous emission of two electrons and of two antineutrinos $(A, Z) \rightarrow (A, Z + 2) + 2\beta^- + 2\tilde{\nu}_e$ (or similar processes with emission of two positrons $2\beta^+$, or electron capture with simultaneous emission of positron $\varepsilon\beta^+$, or double electron capture 2ε) [21]. This process is allowed in the Standard Model (SM), but being of the second order in the perturbation theory, it is characterised by extremely low probability: to-date it is the rarest decay observed in direct laboratory experiments. It has been detected for only 10 nuclides; the corresponding half lives are in the range of 10^{18}–10^{21} yr [22,23]. In contrast, neutrinoless (0ν) double beta decay is forbidden in the SM because it violates lepton number by two units. However, it is predicted in many SM extensions. Its existence can lead to the extremely important conclusion that the neutrino is a Majorana particle ($\nu = \tilde{\nu}$) and permits the measurement of the absolute scale of the neutrino mass (while the neutrino oscillation experiments measure only the differences between neutrino masses).

Three Ce isotopes are candidates for 2β decay: ^{136}Ce with natural abundance $\delta = 0.185\%$ [14] and an energy release $Q = 2419(13)$ keV [24]; ^{138}Ce with $\delta = 0.251\%$ and $Q = 693(10)$ keV; and ^{142}Ce with $\delta = 11.114\%$ and $Q = 1416.7(2.1)$ keV. ^{136}Ce isotope is the most interesting one because the high energy release allows its $2\beta^+$ decay, which is energetically possible only for 6 nuclei-candidates [22]. As well, some resonant neutrinoless 2ε captures in ^{136}Ce to excited states of ^{136}Ba are energetically possible (see e.g. [25]). Up to now, 2β decays of Ce isotopes to the ground states of daughter nuclides were searched for only in 3 experiments [7,17,26]. The processes were not detected and only limits on half lives were set; for ^{136}Ce and ^{138}Ce, of interest in this work, they are in the range of 10^{15}–10^{17} yr.

In this paper we report results on the internal impurities of a $CeCl_3$ sample – to our knowledge, for the first time – based on the measurements of a $CeCl_3$ crystal with HP Ge detector. The measurements also allow the improvement of some limits on 2β processes in ^{136}Ce and ^{138}Ce

and, in particular, to set the first $T_{1/2}$ limits for the decays of ^{136}Ce to excited states of ^{136}Ba (including those with possible resonant enhancement).

2. Measurements and results on internal contamination of CeCl₃

The used CeCl₃ crystal has dimensions of $\oslash 13 \times 13$ mm and a mass of 6.9 g. Because of its hygroscopy, it was housed in combined plastic and copper container with external dimensions $\oslash 16 \times 20$ mm; total mass of the sample was 13.8 g. Radioactive contamination of the sample was measured with a HP Ge detector (244 cm³ volume) in the low-background set-up located deep underground (3600 m w.e.) in the Laboratori Nazionali del Gran Sasso (LNGS) of INFN (Italy). To reduce external background, the detector was shielded by layers of low-radioactive copper ($\simeq 10$ cm), lead ($\simeq 20$ cm) and polyethylene ($\simeq 10$ cm). The set-up has been continuously flushed by high purity nitrogen (stored deep underground for a long time) to avoid presence of residual environmental radon. Energy resolution (FWHM) of the HP Ge detector was 2 keV at 1332 keV γ line of ^{60}Co; it depends on energy, and for all cases considered in the following it was calculated using the FWHM values of nearby peaks. The CeCl₃ sample was measured during 1279.9 h; the background of the HP Ge detector was measured over 3047.7 h. Part of the experimental spectrum in the energy region 750–1500 keV together with the background for comparison is shown in Fig. 1.

The peaks found in the spectrum of the CeCl₃ crystal belong to: (1) standard contaminants: ^{40}K, ^{60}Co, ^{137}Cs and nuclides in chains of ^{232}Th, ^{235}U, ^{238}U; (2) non-standard but expected: ^{138}La.

The specific activities of the nuclides were calculated with the formula: $A = (S_s/t_s - S_b/t_b)/(y \cdot \eta \cdot m)$, where S_s (S_b) is the area of a peak in the sample (background) spectrum; t_s (t_b) is the time of the sample (background) measurement; y is the yield of the corresponding γ line [13]; η is the efficiency of the full peak detection; m is the mass of the sample. The registration efficiencies were calculated with the EGS4 package [27]. The values of the limits were obtained with the Feldman–Cousins procedure [28].

The comparison of the rates of the peaks of ^{40}K, ^{60}Co, ^{137}Cs and ^{232}Th in the CeCl₃ spectrum with those in the background gives no evidence for pollution of the CeCl₃ crystal by these nuclides: intensities of the peaks were equal to intensities of the peaks in the background spectrum within statistical uncertainties. For example, for line 1173.2 keV of ^{60}Co, (101 ± 19) counts were observed in the CeCl₃ spectrum, and (280 ± 33) counts in the background; the rates are (1.89 ± 0.36) counts/day and (2.21 ± 0.26) counts/day, respectively. Taking into account the γ yield $y = 99.97\%$ [13] and the efficiency $\eta = 1.9 \times 10^{-2}$, this gives activity of (-27 ± 38) mBq/kg, or limit < 38 mBq/kg at 90% C.L. with the Feldman–Cousins procedure [28]. Analysis of the second line of ^{60}Co at 1332.5 keV gives a very close result (< 39 mBq/kg at 90% C.L.).

For the ^{235}U and ^{238}U chains, additional pollution is observed. Taking into account the strong interference between the most intensive line in ^{235}U chain at 185.7 keV (^{235}U, $y = 57.20\%$) with the line at 186.2 keV in ^{238}U chain (^{226}Ra, $y = 3.59\%$), and the line of 351.1 keV (^{211}Bi in ^{235}U chain, $y = 12.91\%$) with the line 351.9 keV (^{214}Pb in ^{238}U chain, $y = 37.60\%$), we estimated the ^{238}U activity using strong peaks of ^{214}Pb (242.0, 295.2 keV) and ^{214}Bi (609.3, 1120.3, 1238.1, 1764.5, 2204.2 keV) as (700 ± 70) mBq/kg. The activity of ^{235}U was calculated – considering the peaks at 236.0 keV (^{227}Th), at 269.5 keV (^{223}Ra) and at 271.2 keV (^{219}Rn) to be: (360 ± 190) mBq/kg. The activities, derived in this way, give also correct results for the total area of the observed 186 and 352 keV peaks in the assumption of equilibrium in the ^{235}U
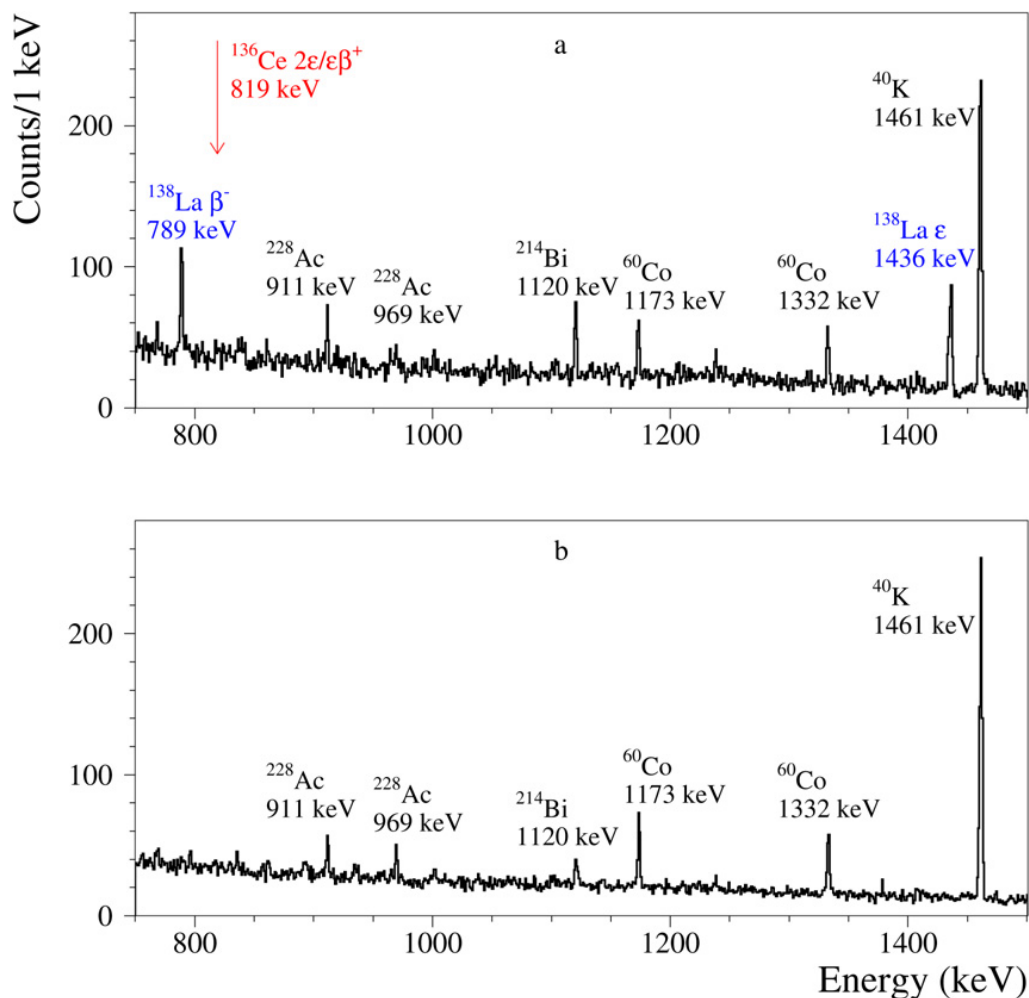
Fig. 1. (Color online.) Spectra measured underground in the LNGS with the low-background HP Ge detector (244 cm^3): (a) CeCl$_3$ sample during 1279.9 h; (b) background of the HP Ge detector during 3047.7 h (normalized here to 1279.9 h). Peaks of 789 keV (^{138}La β^- decay) and 1436 keV (^{138}La electron capture) are evident in the CeCl$_3$ spectrum. The arrow (red online) corresponds to energy of the first excited level of ^{136}Ba (819 keV) which could be populated in 2ε and $\varepsilon\beta^+$ decay of ^{136}Ce.

and ^{238}U chains. For example, the contribution to the peak at 186 keV is equal to 37 counts from ^{226}Ra and 305 counts from ^{235}U; the sum of 342 counts is in perfect agreement with the total observed 346 counts.

As regards the pollution of the CeCl$_3$ crystal by elements from the lanthanide series, the presence of the radioactive ^{138}La is evident: see Fig. 1 where two prominent peaks are present at 788.7 keV (β^- decay of ^{138}La, 33.6%) and at 1435.8 keV (EC of ^{138}La, 66.4%). The corresponding activity of ^{138}La is equal to (680 ± 50) mBq/kg. The presence of other radioactive lanthanides which decay with emission of γ quanta and, thus, could be registered in the measurements with HP Ge detector, was not seen, and only limits for their activities were calculated using the most intensive expected lines: < 50 mBq/kg for ^{176}Lu (lines at 201.8 and 306.8 keV); < 130 mBq/kg for ^{152}Eu (lines at 344.3 and 1408.0 keV); < 60 mBq/kg for ^{154}Eu (1274.4 keV line). The summary of the results is given in Table 1. It should be noted that, because of the chemical similarity of the elements in the lanthanide series, it is quite natural to suppose the presence of ^{138}La in the CeCl$_3$ crystal itself; however, some La contamination might have also been arisen during the CeCl$_3$ crystal manufacturing. In addition, some radioactive ^{138}La could also be created as result of cosmogenic activation of Ce on the Earth surface. Finally, because of

Table 1
Radioactive contamination of the $CeCl_3$ crystal measured with the HP Ge spectrometry. The limits are given at 90% C.L.

Chain	Nuclide	Activity (mBq/kg)
^{232}Th	^{228}Ac	$\leqslant 210$
	^{212}Pb	$\leqslant 230$
	^{208}Tl	$\leqslant 73^*$
^{235}U	^{227}Th	$= 361(340)$
	^{223}Ra	$= 365(229)$
	^{219}Rn	$= 365(229)$
^{238}U	^{214}Pb $+ ^{214}$Bi	$= 700(70)$
	^{40}K	$\leqslant 1700$
	^{60}Co	$\leqslant 39$
	^{137}Cs	$\leqslant 58$
	^{138}La	$= 680(50)$
	^{152}Eu	$\leqslant 130$
	^{154}Eu	$\leqslant 60$
	^{176}Lu	$\leqslant 50$

* ^{208}Tl branching in ^{232}Th chain is 35.94%. Thus ^{208}Tl activity of 73 mBq/kg corresponds to 203 mBq/kg in ^{232}Th.

its hygroscopy, the crystal was measured together with the plastic and copper container which hosted the $CeCl_3$ and which also could be contaminated; thus, the observed additional activities of ^{235}U and ^{238}U should be considered as an upper limits which could be present in the $CeCl_3$ crystal.

3. New limits on 2β decays of ^{136}Ce and ^{138}Ce

As was already stated in the Introduction, three Ce isotopes could decay through double beta processes: ^{136}Ce ($2\beta^+/\varepsilon\beta^+/2\varepsilon$ decay, $Q = 2419(13)$ keV), ^{138}Ce (2ε capture, $Q = 693(10)$ keV), and ^{142}Ce ($2\beta^-$ decay, $Q = 1417(2)$ keV). Because the first excited state of ^{142}Nd has an energy of 1576 keV and cannot be populated in $2\beta^-$ decay of ^{142}Ce, we cannot search for this process in the present measurements. With natural abundances of $\delta = 0.185\%$ for ^{136}Ce and $\delta = 0.251\%$ for ^{138}Ce, the investigated $CeCl_3$ crystal (mass of 6.9 g) contains the following number of ^{136}Ce and ^{138}Ce nuclei: $N_{136} = 3.12 \times 10^{19}$ and $N_{138} = 4.23 \times 10^{19}$, respectively. For ^{138}Ce, only 2ε capture, and only to the ground state of ^{138}Ba is possible. The physics for the ^{136}Ce is more rich. Two positrons could be emitted in the $2\beta^+$ decay of ^{136}Ce with the total energy of 375 keV (thus, only the ground state of ^{136}Ba can be populated, see Fig. 2). Annihilation of these positrons will give four γ's of 511 keV leading to extra rate of the 511 keV peak in the HP Ge detector. In the $\varepsilon\beta^+$ decay, not only the ground state but also the first excited level of ^{136}Ba at 818.5 keV could be populated. In the 2ε capture, many excited 0^+ and 2^+ levels of ^{136}Ba – up to the level of 2399.9 keV – could be populated with the subsequent emission of different γ quanta in the deexcitation process; see scheme on Fig. 2.

In general, we do not observe any additional activity in the spectrum of the $CeCl_3$ crystal which could indicate the presence of 2β processes in ^{136}Ce and ^{138}Ce, thus in the following only limits on the corresponding $T_{1/2}$ values are given.

The $2K2\nu$ capture of the ^{136}Ce (^{138}Ce) to the ground state of ^{136}Ba (^{138}Ba) is not considered here because as result of such a process only low energy X rays are emitted ($E \simeq 32$–37 keV) which are effectively absorbed in the $CeCl_3$ crystal, in its housing and in the holder of the HP
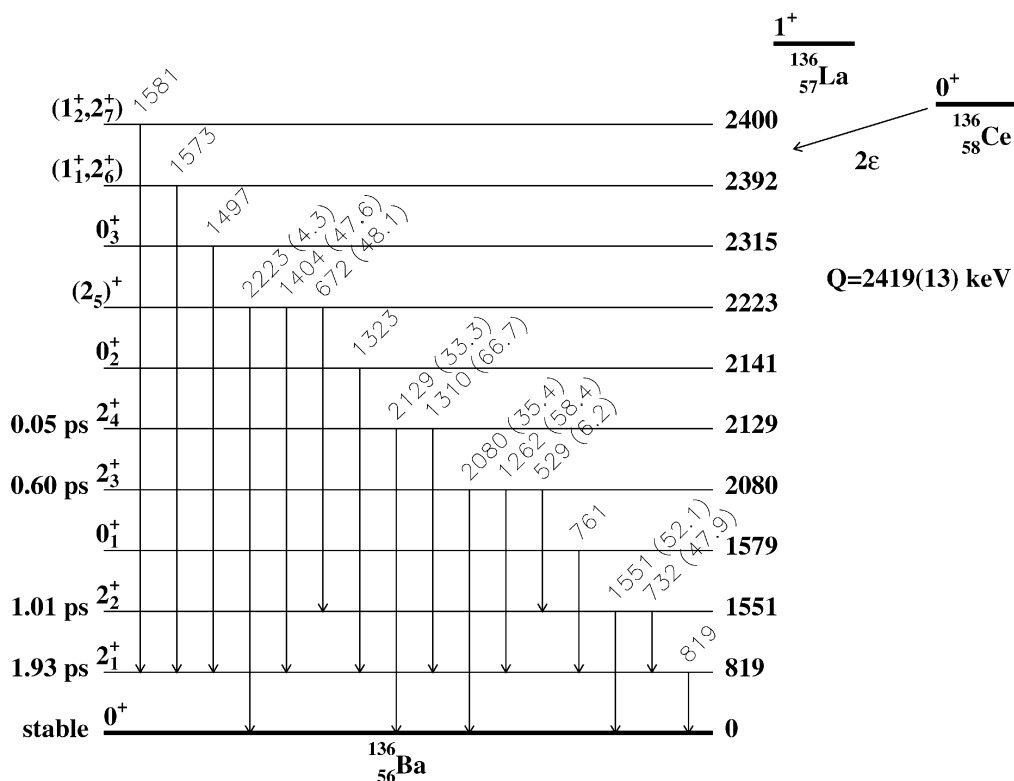
Fig. 2. Scheme of the nuclear triplet ^{136}Ce–^{136}La–^{136}Ba [13,29]. Energies of the excited levels of ^{136}Ba and emitted in deexcitation process γ quanta are rounded to 1 keV. Relative intensities of γ's are given in parentheses.

Ge detector. This gives a low detection efficiency and ensures low sensitivity in comparison with that obtained in experiment [7], where a CeF$_3$ crystal scintillator was used as the detector itself of such processes (the so-called "source = detector" approach); efficiency for X rays was close to 1 that resulted in the limits: $T_{1/2}(2K2\nu) > 2.7 \times 10^{16}$ yr for ^{136}Ce and $T_{1/2}(2K2\nu) > 3.7 \times 10^{16}$ yr for ^{138}Ce at 90% C.L. [7].

3.1. $2\varepsilon0\nu$ captures in ^{136}Ce (^{138}Ce) to the ground state of ^{136}Ba (^{138}Ba)

In the neutrinoless process, in addition to the X rays described above and visible in our measurements with very low efficiencies, some other particle(s) should be emitted to take away the energy released by the two neutrinos in the 2ν process. Usually a (bremsstrahlung) gamma quantum is assumed, and below we also suppose deexcitation by one γ quantum. Its energy will be equal to $E_\gamma = Q - E_{b1} - E_{b2}$, where E_{b1} and E_{b2} is the binding energy of the first and of the second captured electron on the corresponding atomic shells. For Ba atom, the binding energies on the K and L_1, L_2, L_3 shells are equal: $E_K = 37.4$, $E_{L_1} = 6.0$, $E_{L_2} = 5.6$ and $E_{L_3} = 5.2$ keV. Thus, the expected energy of the γ quantum for the $2\varepsilon0\nu$ capture: ^{136}Ce $\rightarrow$ ^{136}Ba(g.s.), is equal to: (i) $E_\gamma = 2344$ keV for $2K0\nu$; (ii) $E_\gamma = 2376$ keV for $KL0\nu$ (energy resolution of the HP Ge detector does not allow to distinguish between L_1, L_2 and L_3 transitions); (iii) $E_\gamma = 2407$ keV for $2L0\nu$ processes.

Fig. 3 shows the energy spectrum of the CeCl$_3$ sample in the region of $2K0\nu$, $KL0\nu$ and $2L0\nu$ decays of ^{136}Ce and ^{138}Ce. The expected peaks are not evident, and we can give only a limit on the half life:

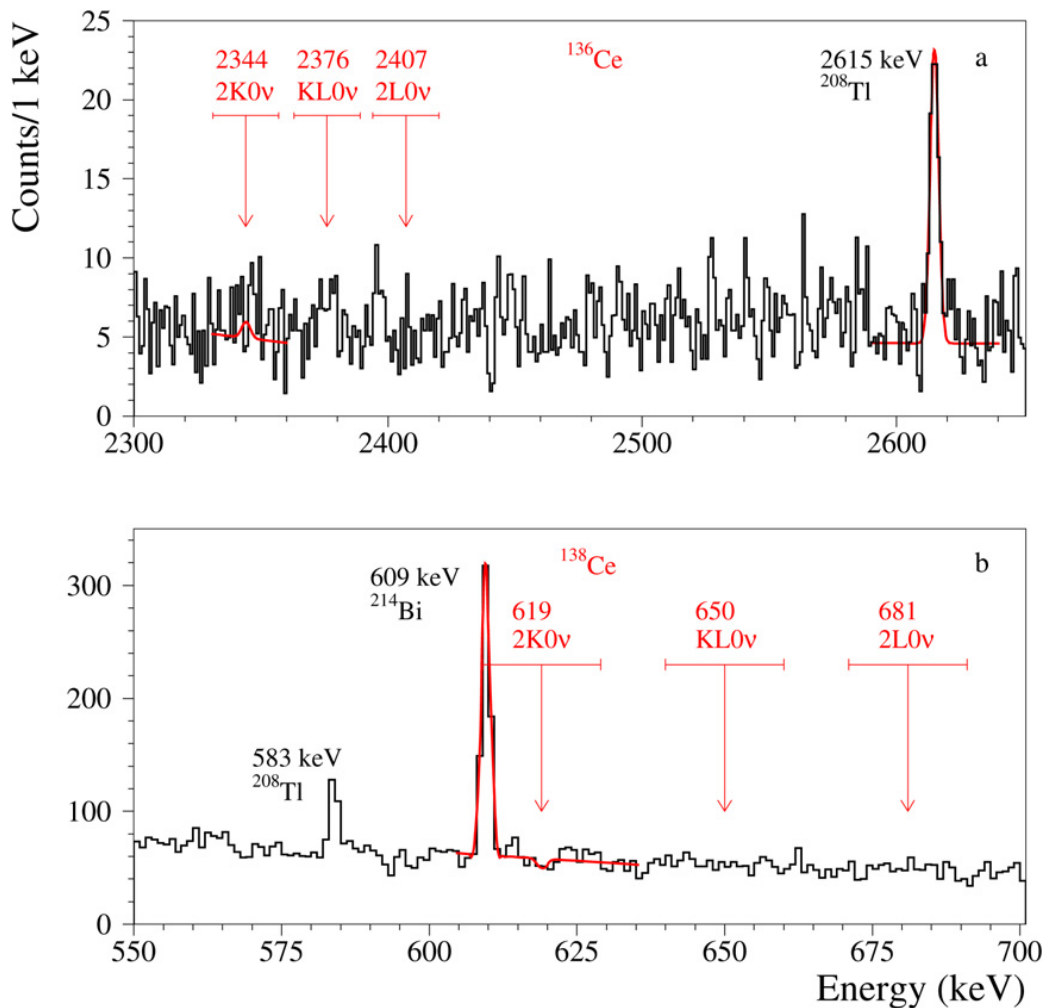$$\lim T_{1/2} = \ln 2 \cdot \eta \cdot N \cdot t / \lim S, \tag{1}$$

Fig. 3. (Color online.) Part of the CeCl$_3$ experimental spectrum in the energy region of the $2\varepsilon 0\nu$ captures in ^{136}Ce (a) and ^{138}Ce (b). The arrows show the expected energies for the $2K0\nu$, $KL0\nu$ and $2L0\nu$ decays; their error bars reflect the uncertainty in the knowledge of the atomic mass differences: 13 keV for ^{136}Ce–^{136}Ba and 10 keV for ^{138}Ce–^{138}Ba. The results of the fit of some peaks are also shown as examples: (a) the 2615 keV peak of ^{208}Tl and the expected peak at 2344 keV (^{136}Ce $2K0\nu$); (b) the 609 keV peak of ^{214}Bi and the expected peak at 619 keV (^{138}Ce $2K0\nu$).

where η is the efficiency to detect a corresponding γ quantum; N is the number of ^{136}Ce or ^{138}Ce nuclei; t is the time of measurement ($t = 1279.9$ h); and lim S is a limit on the peak area which could be excluded at some confidence level for given experimental data.

In the so-called "1σ approach", the excluded number of real events that could be invisible in the spectrum is estimated as a square root of the number of background counts in a suitably chosen energy region; such an evaluation is simple but gives the right scale of sensitivity of an experiment. Taking into account that the background rate at the energies of $2\varepsilon 0\nu$ decay of ^{136}Ce is near 5 counts/keV (see Fig. 3a) and the expected width of the peaks in this region is 5 keV, we obtain lim $S = 8$ at 90% C.L. Together with $N_{136} = 3.12 \times 10^{19}$ and the efficiency $\eta = 1.1\%$, it gives $T_{1/2} = 4.3 \times 10^{15}$ yr.

Further, to obtain the limit on the $2K0\nu$ capture of ^{136}Ce (the expected energy of the peak is 2344 keV), the experimental spectrum in the range of 2330–2360 keV was fitted, using the standard least square procedure, by the sum of the straight line (representing the background here) and a Gaussian with fixed centre and width. As a result of the fitting (see Fig. 3a, χ^2/n.d.f. $= 1.1$), the obtained area for the 2344 keV peak is equal (4.3 ± 4.4) counts, giving no evidence for the

effect. The maximum number of excluded events was calculated with the Feldman–Cousins procedure [28] as $\lim S = 11.5$ at 90% C.L. This gives the limit: $T_{1/2} = 3.0 \times 10^{15}$ yr. We also have to take into account that, because the atomic mass difference ^{136}Ce–^{136}Ba is known currently with an uncertainty of 13 keV, the same uncertainty has the energy of the expected $2K0\nu$ peak: (2344 ± 13) keV. Because of this, we repeat the fitting procedure with the centre of the Gaussian in the range of 2331–2357 keV with step of 1 keV recalculating the $T_{1/2}$ limit. The final conservative limit on $2K0\nu$ decay of ^{136}Ce is:

$$T_{1/2}\left(^{136}\text{Ce}, 2K0\nu, \text{g.s.}\right) > 1.6 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

The $T_{1/2}$ limits on $KL0\nu$ and on $2L0\nu$ decays of ^{136}Ce were obtained in the same way as:

$$T_{1/2}\left(^{136}\text{Ce}, KL0\nu, \text{g.s.}\right) > 1.4 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.,}$$

$$T_{1/2}\left(^{136}\text{Ce}, 2L0\nu, \text{g.s.}\right) > 1.1 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

For $2\varepsilon0\nu$ decays of ^{138}Ce, the expected energies of γ quanta have the following values: $E_\gamma = 619$ keV for $2K0\nu$, $E_\gamma = 650$ keV for $KL0\nu$ and $E_\gamma = 681$ keV for $2L0\nu$ processes (see Fig. 3b). With background of near 55 counts/keV in this region and width of the peak of near 3 keV, expected $\lim S$ is 21 events at 90% C.L. With efficiency $\eta = 2.7\%$ and number of nuclei $N_{138} = 4.23 \times 10^{19}$, we could expect limits at the level of $T_{1/2} > 5.5 \times 10^{15}$ yr.

For $2K0\nu$ decay of ^{138}Ce, the spectrum was fitted in energy region 605–635 keV by straight line (as background), Gaussian with centre at 609.3 keV (^{214}Bi) and Gaussian at 619 keV (expected $2K0\nu$ peak); this gives $S = (-17.1 \pm 12.4)$ counts (see Fig. 3b). With the Feldman–Cousins procedure one obtains $\lim S = 7.5$ at 90% C.L. which corresponds to limit $T_{1/2} = 1.5 \times 10^{16}$ yr. However, due to uncertainty of 10 keV in the atomic mass difference ^{138}Ce–^{138}Ba and to the existence of a peak-like structures at energies of 614, 623 and 626 keV, the final conservative result is more modest:

$$T_{1/2}\left(^{138}\text{Ce}, 2K0\nu, \text{g.s.}\right) > 1.9 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

Nevertheless, it is slightly better than the limit obtained in Ref. [17]: $T_{1/2} > 1.8 \times 10^{15}$ yr. Similar procedure applied in the case of the $KL0\nu$ and $2L0\nu$ decays of ^{138}Ce gives the limits:

$$T_{1/2}\left(^{138}\text{Ce}, KL0\nu, \text{g.s.}\right) > 4.4 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

$$T_{1/2}\left(^{138}\text{Ce}, 2L0\nu, \text{g.s.}\right) > 4.6 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

3.2. 2ε captures in ^{136}Ce to excited levels of ^{136}Ba

The high energy release in the ^{136}Ce 2ε capture allows the population of many excited 0^+ and 2^+ levels of ^{136}Ba (see Fig. 2). In a subsequent deexcitation process, a cascade of a few γ quanta (together with conversion electrons and e^+e^- pairs) will be emitted. To calculate the efficiencies of the HP Ge detector for these γ's, corresponding cascades were simulated with the EGS4 code [27] with initial kinematics given by the DECAY0 event generator [30] where the total scheme of the decay was described. In particular, it gave the possibility to take into account summing effects when few γ's were emitted (this decreases to some extent the efficiencies for specific γ's in comparison as they would be emitted as single particles).

The limits on $T_{1/2}$ values were obtained in a way similar to the one described in the previous section: fitting the proper energy region by the sum of the background (straight line) and of the Gaussian searched for. Widths of the Gaussians were known from those of neighbouring

peaks from natural radioactivity. However, in contrast to the quite big uncertainty in position of the expected peaks in the $2K0\nu$, $KL0\nu$, $2L0\nu$ decays of ^{136}Ce to the ground state of ^{136}Ba (13 keV), energies of deexcitation γ quanta are known with high precision (< 0.2 keV) [13]; this allows to fix the positions of the searched Gaussians.

In the $2\varepsilon2\nu$ capture in ^{136}Ce to the first excited level of ^{136}Ba (2_1^+) at 819 keV, in addition to X rays only one γ quantum will be emitted with $E_\gamma = 819$ keV. Peak at 819 keV is not observed (see Fig. 1). Excluded peak area is equal to $\lim S = 29$ at 90% C.L., and the related efficiency is $\eta = 2.3\%$. This gives the following limit:

$$T_{1/2}\big(^{136}\text{Ce}, 2\varepsilon2\nu, 819\text{ keV}\big) > 2.5 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

This limit is valid for any $2\varepsilon2\nu$ capture ($2K$, KL, $2L$ and others) because it was derived by using only the energy of the deexcitation γ.

In the neutrinoless ($2\varepsilon0\nu$) process ^{136}Ce $\rightarrow$ ^{136}Ba(819 keV), two γ's are emitted: one is the deexcitation γ with $E_\gamma = 819$ keV, and the second one takes off rest of the energy release; for $2K0\nu$ decay its energy is $E_\gamma = (1526 \pm 13)$ keV. Thus, it is more preferable to use the peak at 819 keV to derive the limit on $T_{1/2}$ for this process, since: (1) the energy of the γ is known exactly, and (2) in this case the limit will be valid for any $2\varepsilon0\nu$ capture. (For these reasons we will use only deexcitation γ's also in the following.) However, the emission of the second γ decreases the efficiency from $\eta = 2.3\%$ to $\eta = 2.0\%$ that results in the limit:

$$T_{1/2}\big(^{136}\text{Ce}, 2\varepsilon0\nu, 819\text{ keV}\big) > 2.2 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

In the $2\varepsilon2\nu$ decay of ^{136}Ce to the second excited level of ^{136}Ba (2_2^+, 1551 keV), three deexcitation γ's are emitted with corresponding probabilities [13,29] and energies $E_{\gamma_1} = 732$ keV, $E_{\gamma_2} = 819$ keV and $E_{\gamma_3} = 1551$ keV. The best sensitivity to the effect searched for gives absence of the 732 keV peak with $\lim S = 5.9$ and efficiency $\eta = 1.0\%$:

$$T_{1/2}\big(^{136}\text{Ce}, 2\varepsilon2\nu, 1551\text{ keV}\big) > 5.4 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

In the $2\varepsilon0\nu$ decay, emission of an additional γ quantum, which takes off the rest of the energy instead released by the 2 neutrinos in the 2ν process, leads to decrease in efficiency to $\eta = 0.89\%$ due to summing effects, and gives the $T_{1/2}$ limit:

$$T_{1/2}\big(^{136}\text{Ce}, 2\varepsilon0\nu, 1551\text{ keV}\big) > 4.8 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

Limits for transitions to other excited levels of ^{136}Ba were obtained in similar way. A summary of the results is given in Table 2.

3.3. Resonant 2ε captures in ^{136}Ce

The transitions to the excited ^{136}Ba levels with energies of 2392.1 keV and 2399.9 keV are the most interesting ones because of the possible resonant enhancement of the process in result of energy degeneracy. The energy release in the $2L$ capture of ^{136}Ce is equal to (2407 ± 13) keV which is very close to the energy of these excited levels. In addition, both of them have spin and parity of $(1^+, 2^+)$ [29], and the $2L$ decays are not suppressed by big change in spin and parity.

The possibility of resonant neutrinoless double electron capture was discussed long ago in Refs. [31–33], where an enhancement of the rate by few orders of magnitude was predicted for perfect energy coincidence between the released energy and the energy of an excited state ($\simeq 10$ eV). In this case the expected half lives could be as low as 10^{24} yr, and the corresponding

Table 2

Half life limits on 2β processes in ^{136}Ce and ^{138}Ce isotopes. The energies of the γ lines, which were used to set the $T_{1/2}$ limit, are listed in column 3.

Process of decay	Level of daughter nucleus	E_γ (keV)	$T_{1/2}$ limit (yr) at 90% C.L.	
			Present work	Best previous result
^{136}Ce $\rightarrow$ ^{136}Ba				
$2\beta^+0\nu$	g.s.	511	$> 4.2 \times 10^{15}$	$> 6.9 \times 10^{17}$ [26][a]
$2\beta^+2\nu$	g.s.	511	$> 4.2 \times 10^{15}$	$> 1.8 \times 10^{16}$ [17]
$\varepsilon\beta^+0\nu$	g.s.	511	$> 2.6 \times 10^{15}$	$> 3.8 \times 10^{16}$ [17]
$\varepsilon\beta^+2\nu$	g.s.	511	$> 2.6 \times 10^{15}$	$> 1.8 \times 10^{15}$ [17]
$\varepsilon\beta^+0\nu$	2_1^+ 819 keV	511	$> 2.4 \times 10^{15}$	–
$\varepsilon\beta^+2\nu$	2_1^+ 819 keV	511	$> 2.4 \times 10^{15}$	–
$2K0\nu$	g.s.	2331–2357	$> 1.6 \times 10^{15}$	$> 6.0 \times 10^{15}$ [17]
$KL0\nu$	g.s.	2363–2389	$> 1.4 \times 10^{15}$	–
$2L0\nu$	g.s.	2394–2420	$> 1.1 \times 10^{15}$	–
$2K2\nu$	g.s.	–	–	$> 2.7 \times 10^{16}$ [7]
$2\varepsilon0\nu$	2_1^+ 819 keV	819	$> 2.2 \times 10^{15}$	–
$2\varepsilon2\nu$	2_1^+ 819 keV	819	$> 2.5 \times 10^{15}$	–
$2\varepsilon0\nu$	2_2^+ 1551 keV	732	$> 4.8 \times 10^{15}$	–
$2\varepsilon2\nu$	2_2^+ 1551 keV	732	$> 5.4 \times 10^{15}$	–
$2\varepsilon0\nu$	0_1^+ 1579 keV	761	$> 5.4 \times 10^{15}$	–
$2\varepsilon2\nu$	0_1^+ 1579 keV	761	$> 6.3 \times 10^{15}$	–
$2\varepsilon0\nu$	2_3^+ 2080 keV	2080	$> 1.2 \times 10^{15}$	–
$2\varepsilon2\nu$	2_3^+ 2080 keV	2080	$> 1.3 \times 10^{15}$	–
$2\varepsilon0\nu$	2_4^+ 2129 keV	1310	$> 2.9 \times 10^{15}$	–
$2\varepsilon2\nu$	2_4^+ 2129 keV	1310	$> 3.2 \times 10^{15}$	–
$2\varepsilon0\nu$	0_2^+ 2141 keV	1323	$> 5.3 \times 10^{15}$	–
$2\varepsilon2\nu$	0_2^+ 2141 keV	1323	$> 6.1 \times 10^{15}$	–
$2\varepsilon0\nu$	$(2_5)^+$ 2223 keV	1404	$> 5.4 \times 10^{15}$	–
$2\varepsilon2\nu$	$(2_5)^+$ 2223 keV	1404	$> 5.6 \times 10^{15}$	–
$2\varepsilon0\nu$	0_3^+ 2315 keV	819	$> 5.4 \times 10^{15}$	–
$2\varepsilon2\nu$	0_3^+ 2315 keV	819	$> 5.6 \times 10^{15}$	–
$2\varepsilon0\nu$	$(1_1^+, 2_6^+)$ 2392 keV	1573	$> 2.4 \times 10^{15}$	–
$2\varepsilon2\nu$	$(1_1^+, 2_6^+)$ 2392 keV	1573	$> 2.4 \times 10^{15}$	–
$2\varepsilon0\nu$	$(1_2^+, 2_7^+)$ 2400 keV	1581	$> 4.1 \times 10^{15}$	–
$2\varepsilon2\nu$	$(1_2^+, 2_7^+)$ 2400 keV	1581	$> 4.1 \times 10^{15}$	–
^{138}Ce $\rightarrow$ ^{138}Ba				
$2K0\nu$	g.s.	609–629	$> 1.9 \times 10^{15}$	$> 1.8 \times 10^{15}$ [17]
$KL0\nu$	g.s.	640–660	$> 4.4 \times 10^{15}$	–
$2L0\nu$	g.s.	671–691	$> 4.6 \times 10^{15}$	–
$2K2\nu$	g.s.	–	–	$> 3.7 \times 10^{16}$ [7]

[a] At 68% C.L.

experiments even could compete with searches for neutrinoless $2\beta^-$ decay in sensitivity to the neutrino mass.

Theoretical expectations were given in Ref. [33] for the transition ^{112}Sn($2K0\nu$) $\rightarrow$ ^{112}Cd(1871 keV). In Ref. [34] the following resonant captures were calculated: ^{112}Sn($2K0\nu$) $\rightarrow$

^{112}Cd(1871 keV), ^{136}Ce($2K0\nu$) $\rightarrow$ ^{136}Ba(2315 keV), ^{152}Gd($KL0\nu$) $\rightarrow$ ^{152}Sm(g.s.), ^{162}Er($2K0\nu$) $\rightarrow$ ^{162}Dy(1745 keV), ^{164}Er($2L0\nu$) $\rightarrow$ ^{164}Dy(g.s.), ^{180}W($2K0\nu$) $\rightarrow$ ^{180}Hf(g.s.). The level at 2315 keV, considered in [34] for the decay: ^{136}Ce($2K0\nu$) $\rightarrow$ ^{136}Ba, is quite far from resonance conditions ($\Delta(Q - E_{\mathrm{exc}}) = (29 \pm 13)$ keV) and half life estimation was quite pessimistic (5.0×10^{29} yr for $m_\nu = 1$ eV). It is interesting to note perfect resonant conditions for the decay ^{152}Gd($KL0\nu$) $\rightarrow$ ^{152}Sm(g.s.) with $\Delta(Q - E_{\mathrm{exc}}) = (0.1 \pm 1.2)$ keV and estimated half life of 5.0×10^{24} yr [34].

There were only few searches in this field to-date. The following direct experiments should be listed:

- ^{74}Se($2L0\nu$) $\rightarrow$ ^{74}Ge(1204 keV) – $T_{1/2} > 5.5 \times 10^{18}$ yr [35];
- ^{106}Cd($KL0\nu$) $\rightarrow$ ^{106}Pd(2741 keV) – $T_{1/2} > 3.0 \times 10^{19}$ yr [36];
- ^{112}Sn($2K0\nu$) $\rightarrow$ ^{112}Cd(1871 keV) – $T_{1/2}$ limits from 1.4×10^{18} to 9.2×10^{19} yr [37–40];
- ^{180}W($2K0\nu$) $\rightarrow$ ^{180}Hf(g.s.) – $T_{1/2}$ limits from 5.0×10^{16} to 8.6×10^{17} yr [41–43].

Geochemical results for the transitions ^{130}Ba($2K0\nu$) $\rightarrow$ ^{130}Xe(2544 keV) and ^{130}Ba($2L0\nu$) $\rightarrow$ ^{130}Xe(2608 keV) also have to be mentioned with limit $T_{1/2} > 4.0 \times 10^{21}$ yr [44] and a positive observation declared in [45] with $T_{1/2} = (2.16 \pm 0.52) \times 10^{21}$ yr.

Continuing our consideration of ^{136}Ce 2ε decays, we give below the first experimental limits on resonant transitions to the excited 2392.1 keV and 2399.9 keV levels of ^{136}Ba.

Only two high energy γ's will be emitted after population of the 2392 keV level, $E_{\gamma_1} = 1573$ keV and $E_{\gamma_2} = 819$ keV (see Fig. 2). Both peaks are not observed in the experimental spectrum of the CeCl$_3$ sample, and only limits on their areas can be derived as explained in previous sections: lim $S = 29$ counts for 819 keV and lim $S = 17$ counts for 1573 keV at 90% C.L. Related efficiencies are $\eta = 2.0\%$ and $\eta = 1.3\%$, respectively. Using Eq. (1) with $t = 1279.9$ h, $N_{136} = 3.12 \times 10^{19}$ and lim $S = 17$ for the 1573 keV peak, we get:

$$T_{1/2}\left(^{136}\mathrm{Ce}, 2\varepsilon, 0\nu + 2\nu, 2392 \text{ keV}\right) > 2.4 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

Absence of the 819 keV peak gives a very close result ($> 2.2 \times 10^{15}$ yr).

In the transition to the 2400 keV level of ^{136}Ba, two γ's are emitted, $E_{\gamma_1} = 1581$ keV and $E_{\gamma_2} = 819$ keV. Peak at 1581 keV is also absent (lim $S = 10$ counts), and half life limit is:

$$T_{1/2}\left(^{136}\mathrm{Ce}, 2\varepsilon, 0\nu + 2\nu, 2400 \text{ keV}\right) > 4.1 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

In recent years resonant $2\varepsilon 0\nu$ captures draw the increasing attention; see [25] for discussion of perspectives to improve sensitivity of such experiments to the level of $T_{1/2} \sim 10^{26}$ yr.

3.4. $\varepsilon\beta^+$ decay of ^{136}Ce

The $\varepsilon\beta^+$ decay of ^{136}Ce is energetically possible to the ground state and to the first excited level of ^{136}Ba (819 keV).

The $T_{1/2}$ limit to the ground state can be obtained from the excess of events in the annihilation peak at 511.0 keV. In fact, this peak is a superposition of the annihilation peak with the peak of 510.8 keV from ^{208}Tl. However, the activity related with the ^{232}Th chain (based on other peaks in this chain) was not found earlier (Section 2). In the spectrum of the CeCl$_3$ sample, (72 ± 20) events were detected in the 511 keV peak during 1279.9 h; in the background spectrum we have (125 ± 53) events during 3046.7 h. Scaled to 1279.9 h and subtracted, it gives a difference

of (19 ± 30) events, consistent with 0. With $\lim S = 68$ at 90% C.L. in accordance with the Feldman–Cousins procedure and efficiency $\eta = 5.6\%$, we obtain the half life limit:

$$T_{1/2}\left(^{136}\text{Ce}, \varepsilon\beta^+, 0\nu + 2\nu, \text{g.s.}\right) > 2.6 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

For the $\varepsilon\beta^+$ decay to the 819 keV level of ^{136}Ba, the efficiency for the 511 keV peak is $\eta = 5.1\%$. It gives the limit:

$$T_{1/2}\left(^{136}\text{Ce}, \varepsilon\beta^+, 0\nu + 2\nu, 819 \text{ keV}\right) > 2.4 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

Estimation based on the absence of the 819 keV peak gives a slightly worse half life limit of 1.9×10^{15} yr.

3.5. $2\beta^+$ decay of ^{136}Ce

The $2\beta^+$ decay of ^{136}Ce is possible only to the ground state of ^{136}Ba. The efficiency for the 511 keV peak is equal in this case to $\eta = 9.0\%$. Together with $\lim S = 68$ counts derived above, it gives:

$$T_{1/2}\left(^{136}\text{Ce}, 2\beta^+, 0\nu + 2\nu, \text{g.s.}\right) > 4.2 \times 10^{15} \text{ yr} \quad \text{at 90\% C.L.}$$

4. Conclusions

The measurements performed with a small CeCl$_3$ crystal and a low-background HP Ge detector (244 cm^3) were used to determine the internal radioactive contamination of this crystal. Only limits (90% C.L.) were set for activity of radioactive nuclides in the ^{232}Th chain (< 0.2 Bq/kg), of ^{40}K (< 1.7 Bq/kg), of ^{60}Co (< 0.04 Bq/kg), of ^{137}Cs (< 0.06 Bq/kg), and for some radioactive isotopes in the lanthanide series: ^{152}Eu (< 0.13 Bq/kg), ^{154}Eu (< 0.06 Bq/kg), ^{176}Lu (< 0.05 Bq/kg). However, activity was observed for ^{235}U as 0.36(19) Bq/kg, ^{238}U as 0.70(7) Bq/kg, ^{138}La as 0.68(5) Bq/kg (U activities could be related with the plastic and copper container of the CeCl$_3$ crystal). These results are important for further applications of CeCl$_3$ as crystal scintillators.

The measurements have allowed to set, for the first time, half life limits for different double electron captures in ^{136}Ce to the excited levels of ^{136}Ba; the obtained results are in the range of $(1$–$6) \times 10^{15}$ yr. In particular, the resonant $2\varepsilon0\nu$ captures to the levels of 2392.1 keV and 2399.9 keV of ^{136}Ba are limited as $T_{1/2} > 2.4 \times 10^{15}$ yr and $T_{1/2} > 4.1 \times 10^{15}$ yr at 90% C.L., respectively. The sensitivity is restricted by the small mass of the CeCl$_3$ sample (6.9 g) and by the low natural abundance of ^{136}Ce isotope (0.185%). These could be improved in future experiments. In the simplest approach, a Ce sample with mass of $\simeq 1$ kg could be measured underground with a HP Ge detector. More sophisticated scheme with use of big CeCl$_3$ (or CeF$_3$, or CeBr$_3$) scintillator working in coincidence with a HP Ge detector is under consideration too (also in relation with possibility to investigate 2β processes in other scintillators). Expected sensitivity with use of natural Ce is at least around 10^{18} yr, and would be better with Ce enriched in ^{136}Ce isotope.

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