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Quest for double beta decay of ^{160}Gd and Ce isotopes

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Abstract

The 2β decay study of ^{160}Gd has been performed in the Solotvina Underground Laboratory with the help of $\text{Gd}_2\text{SiO}_5:\text{Ce}$ crystal scintillator (volume 95 cm^3). The background of the detector in the vicinity of the $Q_{\beta\beta}$ energy of ^{160}Gd was reduced to $1.0\text{ cpd}/(\text{keV kg})$. The new improved half-life limits have been established for $0\nu 2\beta$ decay of ^{160}Gd to the ground (0^+) and first excited (2^+) levels of ^{160}Dy : $T_{1/2}^{0\nu} \geq 2.3(1.3) \times 10^{21}\text{ yr}$ at 68%(90%) C.L. The $T_{1/2}$ bounds have been also set for 2ν , $0\nu\chi$ and $0\nu\chi\chi$ modes of ^{160}Gd decay, as well as for different 2β decay processes in ^{136}Ce , ^{138}Ce and ^{142}Ce . © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The exceptional interest in the neutrinoless double-beta ($0\nu 2\beta$) decay is explained by the great potential of this process — which violates the lepton number conservation — in the search for the neutrino mass (m_ν) and by its nature as a sign of a possible new physics beyond the standard model (SM) [1–6]. The absence of the $0\nu 2\beta$ decay, established at the required level of sensitivity, yields strong restrictions on m_ν , lepton violation constants and other parameters of the manifold SM extensions, which allow to narrow a wide choice of theoretical models and to touch the multi-TeV energy range competitive to the accelerator experiments [3–6].

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In fact, the most sensitive $0\nu 2\beta$ results were obtained by using the so called “active source” technique, in which a detector (containing 2β candidate nuclei) serves as source and detector simultaneously [1,2]. As examples, we recall the impressive half-life limits $T_{1/2}^{0\nu}$ in the range of $(1-4) \times 10^{23}$ yr ($m_\nu \leq 1.3-4$ eV) established for ^{136}Xe (high-pressure Xe TPC) [7], ^{130}Te (low-temperature bolometers TeO_2) [8], ^{116}Cd (enriched $^{116}\text{CdWO}_4$ scintillators) [9], and the highest limit reached for ^{76}Ge (enriched HP ^{76}Ge detectors): $T_{1/2}^{0\nu} \geq 1.8 \times 10^{25}$ yr [10,11]. Therefore, it is apparent that application of the “active source” technique with a new nucleus extends the number of 2β candidates studied with a high sensitivity.¹

During last years cerium-doped gadolinium silicate $\text{Gd}_2\text{SiO}_5:\text{Ce}$ (GSO) crystal scintillators have been developed [13,14]. These scintillators are nonhygroscopic and have a high density (6.71 g/cm^3), fast response (primary decay time about 30–60 ns), quite high light output (20% of NaI(Tl)), and a wavelength of emission with a maximum at 440 nm). Moreover, it was already demonstrated [15–17] that GSO crystals can be applied in the 2β decay search of ^{160}Gd , which is one of the interesting candidate nuclei for the following reasons. First, despite a rather low 2β decay energy release ($Q_{\beta\beta} = 1729.7(13)$ keV [18]), its theoretical value of $T_{1/2}^{0\nu} \cdot \langle m_\nu \rangle^2 = 8.6 \times 10^{23}$ yr is nearly three times lower than that for ^{76}Ge and ^{136}Xe [19], thus for the same lower bound on $T_{1/2}^{0\nu}$, an experiment with ^{160}Gd will yield a more stringent constraints on the neutrino mass and other parameters of the theory. Secondly, a recent calculation [20] shows that two-neutrino 2β decay of ^{160}Gd is forbidden due to the large deformation of this nucleus. Meanwhile, the suppression of the $0\nu 2\beta$ decay mode would be not so strong due to different sets of intermediate states involved in both transitions. Therefore the energy region of $0\nu 2\beta$ signal of ^{160}Gd could be free from the background from $2\nu 2\beta$ decays, which is a very serious problem for 2β detectors with the poor energy resolution [2]. Thirdly, the natural abundance of ^{160}Gd is rather large (21.86% [21]) allowing the construction of a sensitive apparatus with natural GSO crystals.

The present paper describes new and improved half-life limits on 2β decay of ^{160}Gd obtained with a 95 cm^3 GSO crystal scintillator and with about 3 times longer running time than that of Ref. [22], where preliminary results of this experiment have been already published.

2. Experimental set up, background measurements and data analysis

A cerium-doped gadolinium silicate crystal (5.4 cm long, 4.7 cm in diameter) grown by Czochralski method was used in the measurements. The mass of the crystal is 635 g, and the number of ^{160}Gd nuclei is 3.951×10^{23} . The first 630 h of measurements had been carried out with a crystal of mass 698 g, and then its side surface was ground by 1–1.5 mm.

The experiment was performed in the Solotvina Underground Laboratory (SUL) of the INR in a salt mine 430 m underground ($\simeq 1000$ mwe, with a cosmic muon flux of

¹ For example, the first interesting results were obtained recently for 2β decay processes in ^{40}Ca and ^{46}Ca with the help of newly developed low radioactive $\text{CaF}_2(\text{Eu})$ crystal scintillators [12].

$1.7 \times 10^{-6} \text{ cm}^{-2} \text{ s}^{-1}$; a neutron flux $\leq 2.7 \times 10^{-6} \text{ cm}^{-2} \text{ s}^{-1}$, and a radon concentration in the air $< 30 \text{ Bq m}^{-3}$ [23]. In the low background installation, the GSO crystal is viewed by a FEU-110 photomultiplier through a plastic light-guide 8.6 cm in diameter and 18.2 cm long. The energy resolution of the detector was measured in the energy region 60–2615 keV by using γ lines of ^{22}Na , ^{137}Cs , ^{207}Bi , ^{226}Ra , ^{232}Th and ^{241}Am sources. For example, the resolutions (FWHM) are: 16.8%, 13.5%, 11.2% and 10.7% at energies of 662, 1064, 1770 and 2615 keV, respectively. In the course of the measurement, the energy calibration was carried out weekly with a ^{207}Bi source. The passive shield made of high-purity copper (5 cm thickness), mercury (7 cm), and lead (15 cm) surrounds the GSO scintillator to reduce the external background. An event-by-event data acquisition system consists of an IBM PC compatible personal computer and CAMAC crate with electronic units, to record the amplitude (energy) and arrival time of each event [24].

The total running time of the experiment was 13 949 h (1.015 yr/kg of exposure). The measured background spectrum of the GSO crystal is depicted in Fig. 1, where the following peculiarities exist: a clear peak at the energy 420 keV, a comparatively wide peak at the energy around 1050 keV, and two broad distributions at the energies 2.4 and 5.5 MeV. Taking into account the relative light yield for α particles as compared with that

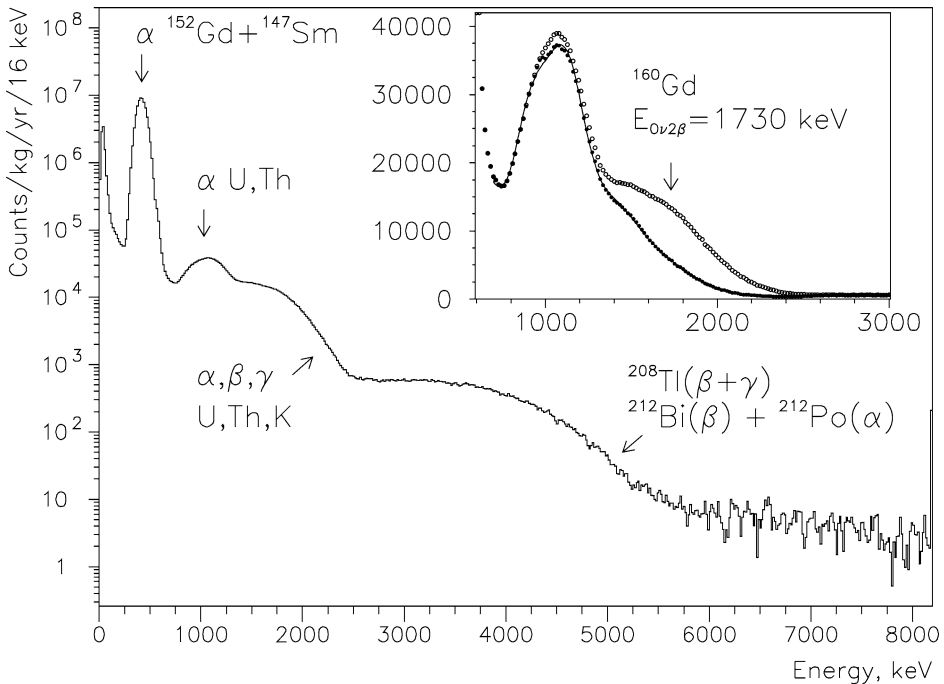


Fig. 1. Background spectrum of the GSO crystal (95 cm^3) collected during 13 949 h. In the insert: part of the measured distribution in the energy interval 600–3000 keV (open circles) together with the residual after subtraction (see text) of the decays from the ^{226}Ra , ^{227}Ac and ^{228}Th intrinsic contamination of the crystal (filled circles) and fitting curve (solid line).

for electrons (α/β ratio) for the GSO scintillator,² the first peak is attributed to α particles from ^{152}Gd ($T_{1/2} = 1.08 \times 10^{14}$ yr; $E_{\alpha} = 2140$ keV; abundance $\delta = 0.20\%$) and ^{147}Sm ($T_{1/2} = 1.06 \times 10^{11}$ yr; $E_{\alpha} = 2233$ keV; $\delta = 15\%$; samarium can be present as impurity of the GSO crystal at a level of ≈ 8 ppm [15]). The peak near 1050 keV as well as the broad distribution up to 2.4 MeV is mainly due to the radioactive contamination of the crystal by the nuclides in the chains of ^{232}Th , ^{235}U and ^{238}U . The second distribution up to 5.5 MeV is caused by decays of ^{232}Th daughter isotopes: (a) the β decay of ^{208}Tl ($Q_{\beta} = 5.00$ MeV); (b) the β decay of ^{212}Bi ($Q_{\beta} = 2.25$ MeV), followed by the α decay of its daughter ^{212}Po ($T_{1/2} = 0.3$ μs ; $E_{\alpha} = 8.78$ MeV corresponding to ≈ 2.7 MeV in β energy).

To recognize and reduce the background from the intrinsic radioactive impurities of the crystal, the off-line analysis of the arrival times of measured events was made as described in detail elsewhere [22,24]. Using this method, fast sequences of decays, belonging to the natural radioactive chains, were searched for as, for example, the sequence of two α decays from the ^{232}Th family: ^{220}Rn ($Q_{\alpha} = 6.41$ MeV, $T_{1/2} = 55.6$ s) \rightarrow ^{216}Po ($Q_{\alpha} = 6.91$ MeV, $T_{1/2} = 0.145$ s) \rightarrow ^{212}Pb . Because the energy of ^{220}Rn α particles corresponds to 1.7 MeV in β energy in the GSO detector, the events within the energy region 1.2–2.2 MeV were used as triggers. Then all events following the triggers in the time interval 10–1000 ms (it contains the part $\eta_t = 0.945$ of the total number of the ^{216}Po decays) were selected. As an example, the spectra of the ^{220}Rn and ^{216}Po α decays obtained in this way — as well as the distribution of the time intervals between the first and second events — are presented in Fig. 2. It is evident from this figure that the selected spectra and time distribution are in the excellent agreement with those expected from α particles from ^{220}Rn and ^{216}Po . Taking into account the efficiency of the time-amplitude analysis, the number of accidental coincidences, and the interfering chain $^{219}\text{Rn} \rightarrow ^{215}\text{Po}$ (it is selected by the applied procedure with 2.04% efficiency), the ^{228}Th activity in the GSO crystal was determined to be 2.287(13) mBq/kg. Then in the next step of the analysis, the found fast couples (^{220}Rn and ^{216}Po) were used as triggers to search for preceding α decays of ^{224}Ra ($Q_{\alpha} = 5.79$ MeV, $T_{1/2} = 3.66$ d). The time window was set as 1–30 s (it contains $\eta_t = 0.30$ of ^{220}Rn decays). The resulting distribution, which includes the accidental coincidences (calculated ratio of the effect to the accidental background is equal to 0.825), is also in a good agreement with the expected α peak of ^{224}Ra .

The same technique was applied to the sequences of α decays from the ^{235}U family: ^{223}Ra ($Q_{\alpha} = 5.98$ MeV, $T_{1/2} = 11.44$ d) \rightarrow ^{219}Rn ($Q_{\alpha} = 6.95$ MeV, $T_{1/2} = 3.96$ s) \rightarrow ^{215}Po ($Q_{\alpha} = 7.53$ MeV, $T_{1/2} = 1.78$ ms) \rightarrow ^{211}Pb . For the fast couple (^{219}Rn and ^{215}Po), events within 1.3–2.4 MeV were used as triggers and the time interval of 0.5–10 ms (containing $\eta_t = 0.803$ of ^{215}Po decays with energy between 1.5 and 2.6 MeV) was chosen.³ The obtained α peaks correspond to activity of 0.948(9) mBq/kg for the ^{227}Ac

² The energy dependence of the α/β ratio was determined by using the α peaks of ^{214}Po , ^{215}Po , ^{216}Po and ^{220}Rn from the internal contamination of the crystal as following: $\alpha/\beta = 0.152 + 0.01765 \cdot E_{\alpha}$, where E_{α} is in MeV. The mentioned peaks were selected from the background with the help of the time-amplitude analysis described below.

³ The used procedure selects also pairs $^{220}\text{Rn} \rightarrow ^{216}\text{Po}$ and $^{214}\text{Bi} \rightarrow ^{214}\text{Po}$ with efficiencies 4.64% and 12.08%, respectively. These contributions were taken into account in the calculation of activities.

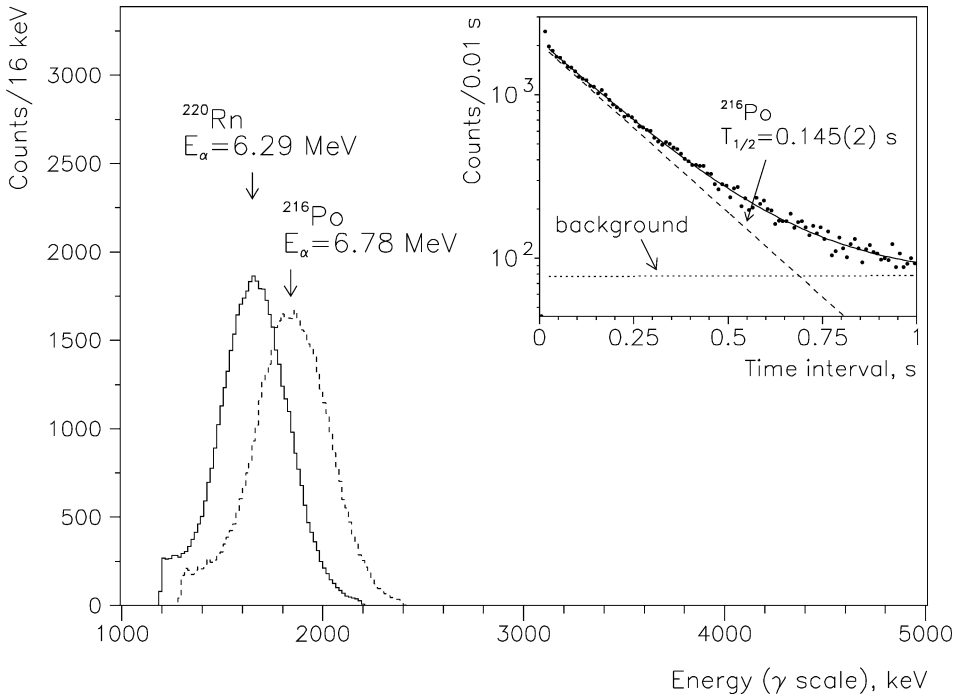


Fig. 2. The energy spectra of the first and second α particles in the decay chain $^{220}\text{Rn} \rightarrow ^{216}\text{Po} \rightarrow ^{212}\text{Pb}$ which were found by means of the time-amplitude analysis of the data recorded over 8609 h. In the insert: the distribution of the time interval between the first and second events together with its fit (solid line) by the sum of exponent (dashed line) with $T_{1/2} = 0.145$ s (table value $T_{1/2} = 0.145(2)$ s [25]) and background (dotted line).

impurity in the crystal. Then the procedure analogous to that described for the ^{224}Ra was applied for to find the preceding ^{223}Ra α decays.

For the analysis of the ^{226}Ra chain (^{238}U family), the following sequence of β and α decays was used: ^{214}Bi ($Q_\beta = 3.27$ MeV, $T_{1/2} = 19.9$ m) \rightarrow ^{214}Po ($Q_\alpha = 7.83$ MeV, $T_{1/2} = 164.3$ μs) \rightarrow ^{210}Pb . For the first event, the lower energy threshold was set at 0.5 MeV, while for the second decay, the energy window 1.3–3.0 MeV was chosen. Time interval of 2–500 μs ($\eta_t = 0.872$ of ^{214}Po decays) was used. The selection efficiency is also decreased a little by the energy threshold applied to the first event (in the procedures described above the selection efficiency of the energy windows η_E is equal to 1). By the Monte Carlo simulation the part η_E of the ^{214}Bi spectrum above 500 keV (as compared with the total spectrum) has been determined as $\eta_E = 0.794$. The obtained spectra for ^{214}Bi ⁴ and ^{214}Po are shown in Fig. 3 and lead to the ^{226}Ra activity in the GSO crystal equal to 0.271(4) mBq/kg.

⁴ Peak observed in the β spectrum of ^{214}Bi (see Fig. 3) is the part (17.7%) of ^{219}Rn α decays (from ^{235}U family), which corresponds to the chosen time interval 2–500 μs . Due to known activity of ^{227}Ac the contribution of $^{219}\text{Rn} \rightarrow ^{215}\text{Po}$ (as well as 0.24% of the chain $^{220}\text{Rn} \rightarrow ^{216}\text{Po}$) was calculated accurately and subtracted from the activities of ^{214}Bi and ^{214}Po .

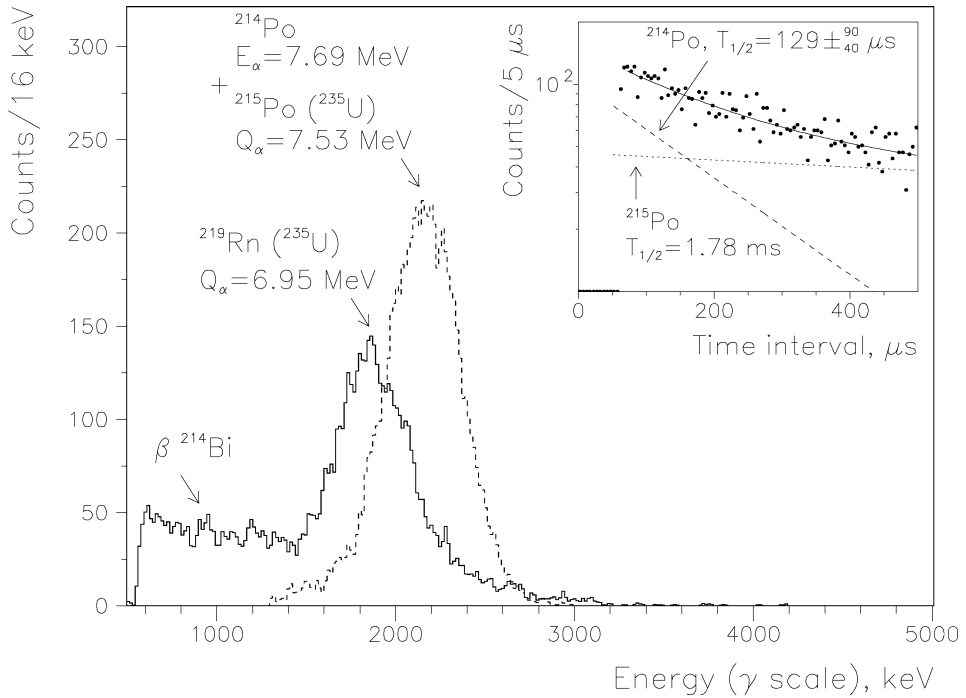


Fig. 3. The energy spectra of the sequence of β and α decays in the decay chain $^{214}\text{Bi} \rightarrow ^{214}\text{Po} \rightarrow ^{210}\text{Pb}$ which were found by means of the time-amplitude analysis of 8609 h data. In the insert: the distribution of the time interval between the first and second events together with its fit (solid line) by the sum of exponent (dashed line) with $T_{1/2} = 129 \mu\text{s}$ (table value $T_{1/2} = 164.3(20) \mu\text{s}$ [25]) and exponent with $T_{1/2} = 1.78 \text{ ms}$ related with the chain $^{219}\text{Rn} \rightarrow ^{215}\text{Po} \rightarrow ^{211}\text{Pb}$ (dotted line).

Besides determination of the background components, the time-amplitude analysis is used for reduction of the background. In this case, the attention is paid, first, to selection of time-correlated decays with minimal loss of the exposure, and, secondly, to prevention of the double selection of the events. With these aims four “removing” procedures were applied (the subscripts indicate the events in the chain):

- (i) $E_1 \geq 500 \text{ keV}$, $\Delta t = 2\text{--}500 \mu\text{s}$, $E_2 = 1000\text{--}3000 \text{ keV}$.

This cut removes mainly decays of ^{214}Bi and ^{214}Po ($\eta_t = 0.872$, $\eta_{E_1} = 0.794$, $\eta_{E_2} = 1.00$, thus the total efficiency of selection is $\eta = \eta_t \eta_{E_1} \eta_{E_2} = 0.692$), while efficiencies for the pairs $^{219}\text{Rn}\text{--}^{215}\text{Po}$ and $^{220}\text{Rn}\text{--}^{216}\text{Po}$ are rather low: 0.177 and 0.024, respectively.

- (ii) $E_1, E_2 = 1200\text{--}3000 \text{ keV}$, $\Delta t = 500 \mu\text{s}\text{--}1 \text{ s}$.

The corresponding efficiencies are: $\eta(^{219}\text{Rn}\text{--}^{215}\text{Po}) = 0.823$, $\eta(^{220}\text{Rn}\text{--}^{216}\text{Po}) = 0.989$, and $\eta(^{214}\text{Bi}\text{--}^{214}\text{Po}) = 0.067$. It is also necessary to take into account the pairs $^{223}\text{Ra}\text{--}^{219}\text{Rn}$ ($\eta = 0.161$) and $^{224}\text{Ra}\text{--}^{220}\text{Rn}$ ($\eta = 0.0124$).

- (iii) In the third cut, a fast chain of decays ($^{220}\text{Rn}\text{--}^{216}\text{Po}$) is used as a trigger to select the previous event of ^{224}Ra . The parameters taken for ^{224}Ra selection are the following:

$E_1 > 1000$ keV, $\Delta t_{12} = 1\text{--}30$ s, $E_2 = 1200\text{--}2200$ keV, $\Delta t_{23} = 0.01\text{--}0.5$ s, $E_3 = 1300\text{--}2400$ keV. The final efficiency for selection of ^{224}Ra is $\eta = 0.258$.

- (iv) In the fourth cut procedure, a fast chain of decays ($^{219}\text{Rn}\text{--}^{215}\text{Po}$) is used as a trigger to select the previous event from ^{223}Ra . The parameters are: $E_1 > 1000$ keV, $\Delta t_{12} = 1\text{--}10$ s, $E_2 = 1300\text{--}2400$ keV, $\Delta t_{23} = 500$ $\mu\text{s}\text{--}10$ ms, $E_3 = 1500\text{--}2600$ keV. It removes 53.4% ($\eta = 0.534$) of ^{223}Ra decays.

Since the time windows of the cuts do not overlap, the probability that one event could be selected more than once is rather low. Indeed, from the total number of 345 169 events being attributed to the time-correlated background, only 318 (i.e. 0.092%) are double-selected.

The part of the measured distribution in the energy interval 600–3000 keV is depicted in the insert of Fig. 1 (open circles) together with the resulting spectrum after subtraction of the selected decays (filled circles). It corresponds to 0.969 kg yr of exposure ($\approx 13\,400$ h), or 95.7% of the initial value that was decreased by the “removing” procedures. At the same time, due to these procedures, the background rate in the vicinity of the energy release of the $0\nu 2\beta$ decay of ^{160}Gd (1648–1856 keV) has been reduced by 2.3 times to the value of 1.01(1) cpd/(keV kg).

3. Background simulation

With the aim to evaluate the 2β decay processes in ^{160}Gd , the measured background spectrum (after subtraction of the time-correlated decays) was simulated with the help of GEANT3.21 package [26]. The event generator DECAY4 [27] was used to describe initial kinematics of decays (number and types of emitted particles, their energies, directions of movement and times of emission). It takes into account decays to ground state as well as to excited levels of daughter nuclei with the subsequent complex deexcitation process [25]. The possibilities of emission of conversion electrons and e^+e^- pairs instead of γ quanta in nuclear transitions and the angular correlation between emitted particles are also taken into consideration.

The background model was built up as a result of the procedure, in which the experimental spectrum was fit by the sum of simulated response functions. The coefficients of the latest were determined on the basis of the following data:

- (i) Activity values for ^{228}Th , ^{227}Ac and ^{226}Ra (and their short-lived daughters) that are present as intrinsic contamination in the GSO crystal. These activities were determined firmly and accurately (with uncertainty less than 1%) with the help of the time-amplitude analysis, as described above. The part of decays of ^{224}Ra , ^{220}Rn , ^{216}Po ; ^{223}Ra , ^{219}Rn , ^{215}Po ; ^{214}Bi and ^{214}Po was removed from the background spectrum, and the remaining part is calculated with high precision for all these nuclides. Thus, one can describe the spectrum of, for example, ^{228}Th + daughters as a sum of the simulated spectra with exactly known areas. If we suppose the secular equilibrium within the natural radioactive chains for these contaminations, the activities of the remaining long-lived members of the

^{232}Th , ^{235}U and ^{238}U families would be known, too. However, it is known that chemical procedures and crystal growth usually break the equilibrium in the natural radioactive series. To account this possibility, the activities of the mentioned remaining members of the radioactive chains (namely, ^{232}Th , ^{228}Ra , $^{235}\text{U} + ^{238}\text{U} + ^{234}\text{U}$, ^{231}Pa , ^{230}Th , and ^{210}Pb) were taken as free parameters for the fitting procedure.⁵

- (ii) The radioactive impurities of the photomultiplier (PMT) are the main source of the external background. The values for PMT FEU-110 were measured previously [24] as 3.0(3) Bq (^{40}K), 0.8(2) Bq (^{226}Ra) and 0.17(7) Bq (^{228}Th). In the fit, these activities were taken as free parameters and varied within their errors.

In addition, simulated spectra of ^{40}K and ^{138}La — natural radionuclides which could be present in the GSO crystal — were also included in the fitting procedure. The last component of the background model is the exponential function (with two free parameters) which describes the residual external background (multiple scattering of γ quanta, influence of weak neutron flux, etc.). The exponential behavior of this component was confirmed by the measurements with the high-radiopurity CdWO_4 crystal scintillator (454 g) performed in the same set up [28].

The fit of the experimental spectrum in the energy region 0.1–3.0 MeV by the sum of the described components gives the following activities of the additional intrinsic contamination of the GSO crystal: $^{40}\text{K} \leq 14$ mBq/kg, $^{138}\text{La} \leq 55$ mBq/kg,⁶ $^{232}\text{Th} \leq 6.5$ mBq/kg, $^{228}\text{Ra} \leq 9$ mBq/kg, $^{238}\text{U} \leq 2$ mBq/kg, $^{231}\text{Pa} \leq 0.08$ mBq/kg, $^{230}\text{Th} \leq 9$ mBq/kg, $^{210}\text{Pb} \leq 0.8$ mBq/kg. The parameters of the exponent were also found in the fit procedure and its contribution to the experimental distribution was revealed to be small ($\approx 2\%$ for the energy interval 1–2 MeV). For illustration, the fitting curve in the energy region 0.7–2.4 MeV is presented in the insert of Fig. 1 together with the experimental data.

4. Half-life limits on the 2β decay of ^{160}Gd

Since in the measured spectrum the $0\nu 2\beta$ decay peak of ^{160}Gd is evidently absent, only the limit for the probability of this process can be set with the present experimental data. To estimate the half-life limit, $T_{1/2}$, we use the formula $\lim T_{1/2} = \ln 2 \cdot \eta \cdot N \cdot t / \lim S$, where η is the detection efficiency, N is the number of ^{160}Gd nuclei, t is the measuring time and $\lim S$ is the maximum number of $0\nu 2\beta$ events which can be excluded with a given confidence level on the basis of the data. To calculate the values of η and $\lim S$, the response function of the GSO detector for the effect being sought has been simulated with the help of GEANT3.21 and DECAY4 programs. It was found that for the $0\nu 2\beta$ decay, the response function is the Gaussian centered at 1730 keV and its width (FWHM)

⁵ Three isotopes of uranium ^{235}U , ^{238}U and ^{234}U are not chemically separable, and their relative activities (0.046 : 1 : 1) do not change, so we can take only one parameter for these nuclides.

⁶ This limit of ^{138}La activity corresponds to the possible La impurity in the GSO crystal at the level of 67 ppm, which does not contradict with the results of the chemical analysis.

equals 176 keV. The edge effects (escape of one or both electrons and bremsstrahlung quanta from the crystal) remove $\approx 5\%$ of events from the peak, thus $\eta = 0.95$. The values of $\lim S$ were determined in two ways. First, by using the so called “one σ approach”, in which the excluded number of real events that could be invisible in the spectrum is estimated simply as square root of the number of background counts in a suitably chosen energy window ΔE . Notwithstanding its simplicity, this method gives the right scale of the sensitivity of the experiment. For instance, in the measured spectrum within the energy interval 1648–1856 keV (it contains 82% of the expected peak area) there are 74 500 counts; thus, the square root estimate gives $\lim S = 273$ events. Using this value of $\lim S$, the total exposure related to ^{160}Gd nuclei ($N \cdot t = 6.04 \times 10^{23}$ nuclei yr), and the calculated efficiency ($\eta = 0.78$), we obtain the half-life limit: $T_{1/2} \geq 1.2 \times 10^{21}$ yr (68% C.L.). Further, the value of $\lim S$ was determined by using the standard least squares procedure, where the experimental energy distribution in the vicinity of the peak searched for was fitted by the sum of the background model (as described above) and $0\nu 2\beta$ decay peak being sought. It should be stressed that for the energy interval of interest the most important contributions ($\approx 73\%$ of the experimental spectrum within 1600–1900 keV) are from the activities of ^{226}Ra , ^{227}Ac and ^{228}Th (and their short-lived daughters) in the intrinsic contamination of the GSO crystal, whose values were determined accurately (less than 1% uncertainty).

As a result of the fitting procedure in the energy region 1.3–2.1 MeV, the obtained area for the $0\nu 2\beta$ decay peak is -160 ± 233 counts (χ^2 value equals to 0.85), thus giving no evidence for the effect. The difference between measured and simulated spectra in the energy region of the hypothetical $0\nu 2\beta$ decay peak of ^{160}Gd is shown in Fig. 4, where the excluded effect is also depicted (solid line). The maximum number of real events, which can be excluded with 90% (68%) C.L. was calculated [29] as 298 (169). It yields the following half-life limit:

$$T_{1/2}(0\nu 2\beta) \geq 1.3(2.3) \times 10^{21} \text{ yr at 90\% (68\%) C.L.}$$

Comparing our limit with the theoretical calculations [19], one can compute the constraint on the neutrino mass $\langle m_\nu \rangle \leq 26(19)$ eV at 90% (68%) C.L.

The same $T_{1/2}$ limit has been set for the $0\nu 2\beta$ transition to the first excited level of ^{160}Dy (2^+ , 87 keV) because 87 keV γ quanta following this process are almost fully absorbed inside the scintillator. The obtained limits are several times larger than those already published [15–17,22].

The two-neutrino 2β -decay rate was evaluated in two ways. For a very conservative estimate, the model spectra of the exactly measured contamination in the crystal and PMT were subtracted from the experimental data. The residual in the chosen energy region was equated to the simulated $2\nu 2\beta$ decay distribution and the latest was taken as an excluded effect with $\lim S = 7.6 \times 10^5$ events corresponding to the $T_{1/2}^{2\nu}(\text{g.s.}) \geq 5.5 \times 10^{17}$ yr (99% C.L.) for $2\nu 2\beta$ decay of ^{160}Gd . However, with this simple approach, it was impossible to reproduce adequately the measured spectrum, thus the background model and the fitting procedure described above were also applied for the 2ν decay mode. The set of fits were performed by changing the fitting energy region from (100–760) keV

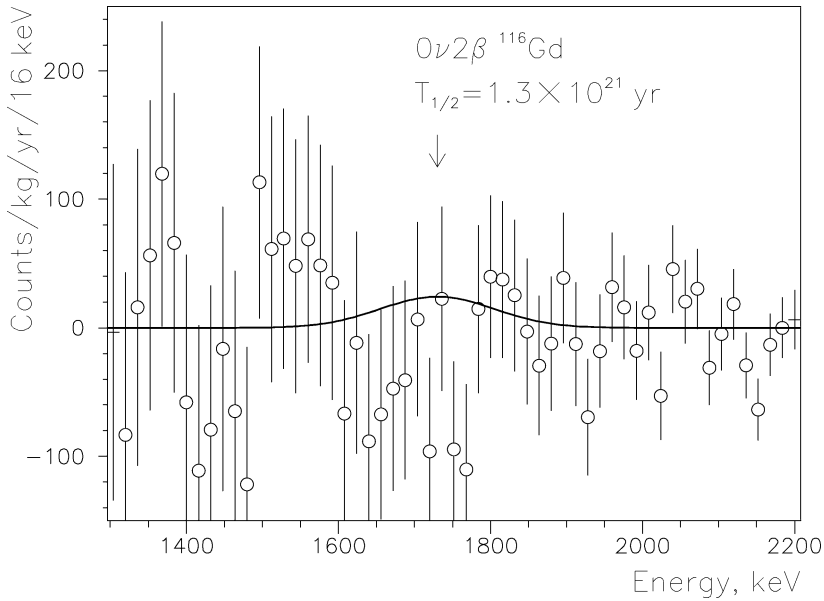


Fig. 4. The residual between the experimental and simulated spectra in the energy region of the $0\nu 2\beta$ decay of ^{160}Gd (open circles with error bars). The solid line represents the excluded $0\nu 2\beta$ peak with $T_{1/2}^{0\nu} = 1.3 \times 10^{21}$ yr (90% C.L.).

to (2400–3000) keV. The maximum value of $\lim S = 2.2 \times 10^4$ events (at 90% C.L.) was found for the energy interval 760–2600 keV. The corresponding Monte Carlo simulated spectrum of $2\nu 2\beta$ decay of ^{160}Gd (g.s. \rightarrow g.s.) is shown in Fig. 5 together with the fitting curve and the most important background components. It is visible from this figure that our background model reproduces the experimental data quite well even outside the region of the fit. The final lower limit for the process searched for is equal to

$$T_{1/2}^{2\nu}(\text{g.s.}) \geq 1.9(3.1) \times 10^{19} \text{ yr} \quad \text{at 90\% (68\%) C.L.}$$

The same method gives the limit for $2\nu 2\beta$ transition to the first (2^+) excited level⁷ of ^{160}Dy :

$$T_{1/2}^{2\nu}(2^+) \geq 2.1(3.4) \times 10^{19} \text{ yr} \quad \text{at 90\% (68\%) C.L.}^8$$

The similar fitting procedure was used to set limits on double beta decays with one or two Majoron emission. The estimated lower bounds on half-life are:

⁷ The shape of distribution for $2\nu 2\beta$ decay to 2^+ level differs from $2\nu 2\beta$ transition to ground (0^+) state [2].

⁸ There are only three theoretical works where $2\nu 2\beta$ decay of ^{160}Gd was considered [19,20,30]. The nuclear matrix elements of ^{160}Gd were calculated within the quasiparticle random phase approximation (QRPA) in Ref. [19] and by using the operator expansion method (OEM) in Ref. [30], which both yield the $T_{1/2}^{2\nu}$ values from 5×10^{18} yr to 3×10^{21} yr. In [20] it was found that due to the large deformation of ^{160}Gd two neutrino 2β decay is simply forbidden ($T_{1/2}^{2\nu} = \infty$). Therefore, part of the interval for the $2\nu 2\beta$ half-life values predicted by the QRPA for ^{160}Gd is excluded by our experimental bound.

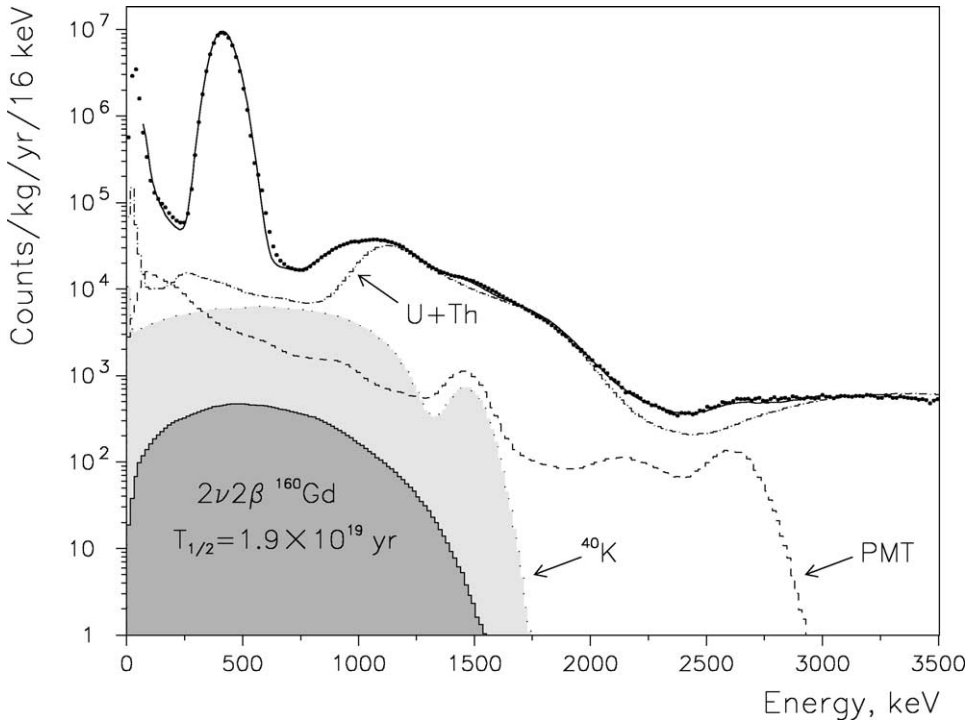


Fig. 5. The background spectrum of the GSO detector for 0.969 yrkg of exposure (points) and the model of background (solid line) obtained by the fitting procedure in 760–2600 keV energy interval (see text). The most important internal (^{40}K and sum of ^{238}U , ^{235}U , ^{232}Th) and external (γ radiation from PMT) components of background are shown. The excluded with 90% C.L. distribution of $2\nu 2\beta$ decay of ^{160}Gd to the ground level of ^{160}Dy corresponds to the half-life limit $T_{1/2}^{2\nu}(\text{g.s.}) = 1.9 \times 10^{19}$ yr.

$$T_{1/2}^{0\nu X} \geq 3.5(5.3) \times 10^{18} \text{ yr} \quad \text{at 90\% (68\%) C.L.}$$

$$T_{1/2}^{0\nu XXX} \geq 1.3(2.0) \times 10^{19} \text{ yr} \quad \text{at 90\% (68\%) C.L.}$$

5. Limits on 2β decay processes of Ce isotopes

The concentration of cerium in the GSO(Ce) crystal (0.8%) is known from the crystal growth conditions and from the results of the chemical analysis. It allows a search for the 2β processes in three cerium isotopes: double positron decay ($2\beta^+$), or electron capture and positron decay ($\epsilon\beta^+$), or double electron capture (2ϵ) in ^{136}Ce (the mass difference between parent and daughter atoms $\Delta M_A = 2397(48)$ keV; the abundance of the parent nuclide $\delta = 0.185\%$); for double electron capture in ^{138}Ce ($\Delta M_A = 693(11)$ keV, $\delta = 0.251\%$); and $2\beta^-$ decay in ^{142}Ce ($\Delta M_A = 1417(2)$ keV, $\delta = 11.114\%$). The total numbers of ^{136}Ce , ^{138}Ce and ^{142}Ce nuclei in the crystal are 4.1×10^{19} , 5.4×10^{19} and 2.4×10^{21} , respectively. The response functions of the detector for the different possible 2β

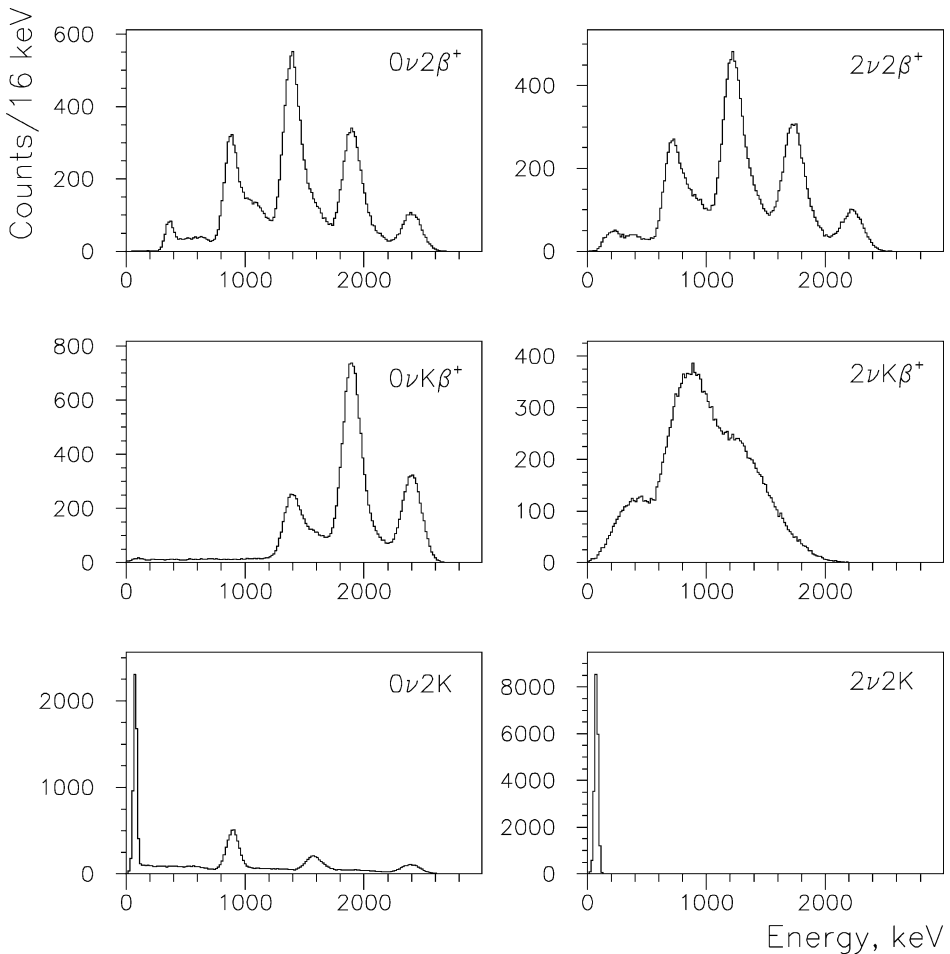


Fig. 6. The response functions of the GSO(Ce) detector for neutrinoless and two-neutrino $2\beta^+$ ($K\beta^+$ and $2K$) decays of ^{136}Ce simulated with the help of GEANT3.21 code and event generator DECAY4.

processes in these cerium isotopes were simulated with the help of GEANT3.21 code and event generator DECAY4. As an example, the simulated energy distributions for neutrinoless and two-neutrino $2\beta^+$ ($K\beta^+$ and $2K$) decays of ^{136}Ce are presented in Fig. 6. Limits on half-lives with respect to different modes of 2β processes of cerium isotopes were calculated in the way analogous to that for ^{160}Gd — they are summarized in Table 1. Except two limits for $0\nu 2\beta^+$ decay of ^{136}Ce and $0\nu 2\beta^-$ decay of ^{142}Ce (already obtained by using CeF_3 scintillators [31]), all presented results for cerium isotopes are set for the first time.

6. Discussion and conclusions

Despite the numerous efforts to detect $0\nu 2\beta$ decay, this process still remains unobserved [1,2]. However, the highest half-life limits set in direct experiments for ^{100}Mo ,

Table 1

Half-life limits on the 2β decay processes in ^{160}Gd , ^{136}Ce , ^{138}Ce and ^{142}Ce

Nuclide	Decay mode		Limit on $T_{1/2}$ (yr)				
			present work 90 (68)% C.L.	other works (C.L.)			
^{160}Gd	$2\beta^-$	0ν	g.s.–g.s., 2^+	$1.3(2.3) \times 10^{21}$	1.4×10^{19} (90%) [15]		
					3.0×10^{20} (68%) [16]		
							8.2×10^{20} (90%) [22]
		2ν	g.s.–g.s.	$1.9(3.1) \times 10^{19}$	1.3×10^{17} (99%) [15]		
		2ν	g.s.– 2^+	$2.1(3.4) \times 10^{19}$			
^{136}Ce	$2\beta^+$	$0\nu\chi$	g.s.–g.s.	$3.5(5.3) \times 10^{18}$	2.7×10^{17} (99%) [15]		
		$0\nu\chi\chi$	g.s.–g.s.	$1.3(2.0) \times 10^{19}$			
		0ν	g.s.–g.s.	$1.9(3.2) \times 10^{16}$	6.9×10^{17} (68%) [31]		
	$K\beta^+$	2ν	g.s.–g.s.	$1.8(3.8) \times 10^{16}$			
		0ν	g.s.–g.s.	$3.8(6.0) \times 10^{16}$			
	^{138}Ce	$2K$	2ν	g.s.–g.s.	$1.8(3.0) \times 10^{15}$		
0ν			g.s.–g.s.	$6.0(8.0) \times 10^{15}$			
2ν			g.s.–g.s.	$0.7(1.1) \times 10^{14}$			
^{142}Ce	$2\beta^-$	0ν	g.s.–g.s.	$1.8(1.9) \times 10^{15}$			
		2ν	g.s.–g.s.	$0.9(1.5) \times 10^{14}$			
^{142}Ce	$2\beta^-$	0ν	g.s.–g.s.	$2.0(3.3) \times 10^{18}$	1.5×10^{19} (68%) [31]		
		2ν	g.s.–g.s.	$1.6(2.6) \times 10^{17}$			

^{116}Cd , ^{130}Te , ^{136}Xe , and ^{76}Ge have already brought the most stringent restrictions on the values of the Majorana neutrino mass $m_\nu \leq (0.5\text{--}5.0)$ eV, right-handed admixtures in the weak interaction $\eta \approx 10^{-7}$, $\lambda \approx 10^{-5}$, the neutrino-Majoron coupling constant $g_M \approx 10^{-4}$, and the R -parity⁹ violating parameter of minimal SUSY standard model $\varepsilon \approx 10^{-4}$. However, on the basis of current status of astroparticle physics, it is very desirable to improve the present level of sensitivity by one-two orders of magnitude [5,6,32].

Many projects were proposed during a past few years with regard to these goals. First of all, there are two projects NEMO-3 [33] and CUORICINO [34] under construction now. The sensitivity of the NEMO-3 tracking detector with a passive 10 kg of ^{100}Mo source (plastic scintillators are used to measure β energies) would be on the level of $\approx 4 \times 10^{24}$ yr ($m_\nu \leq 0.3\text{--}0.5$ eV) [35]. The CUORICINO set up consists of 60 low-temperature bolometers made of TeO_2 crystals (750 g mass each) and is designed as a pilot step for a future CUORE project with one thousand of TeO_2 bolometers (total mass of 750 kg), which could touch ≈ 0.05 eV neutrino mass domain in the 2β decay quest of ^{130}Te [34,36].

An interesting approach to study 2β decay of ^{136}Xe ($Q_{\beta\beta} = 2468$ keV) makes use of the coincident detection of $^{136}\text{Ba}^{2+}$ ions (the final state of the ^{136}Xe decay on the

⁹ R -parity is defined as $R_p = (-1)^{3B+L+2S}$, where B , L and S are the baryon and lepton numbers, and the spin, respectively.

atomic level) and the $0\nu 2\beta$ signal with the energy of 2.5 MeV in a time projection chamber (TPC) filled with liquid or gaseous Xe [37–39]. Recently, the EXO project has been considered [40], where the resonance ionization spectroscopy for the $^{136}\text{Ba}^{2+}$ ions identification would be applied in a 40 m^3 TPC operated at 5–10 atm pressure of enriched xenon ($\approx 1\text{--}2$ tons of ^{136}Xe). Estimated sensitivity to neutrino mass is $\approx 0.01\text{ eV}$ [40]. Another proposal (originated from [41]) is to dissolve $\approx 80\text{ kg}$ (≈ 1.5 tons) of enriched (natural) Xe in the liquid scintillator of the BOREXINO [42] Counting Test Facility (CTF) in order to reach the $T_{1/2}^{0\nu}$ limit in the range of $10^{24}\text{--}10^{25}\text{ yr}$ [43].

The project MOON aims to make both the study of $0\nu 2\beta$ decay of ^{100}Mo ($Q_{\beta\beta} = 3034\text{ keV}$) and the real time studies of low-energy solar ν by inverse β decay [44]. The detector module will be composed of $\approx 60\,000$ plastic scintillators ($6\text{ m} \times 0.2\text{ m} \times 0.25\text{ cm}$), the light outputs from which are collected by 866 000 wave length shifter fibers ($\varnothing 1.2\text{ mm} \times 6\text{ m}$), viewed through clear fibers by 6800 16-anode photomultiplier tubes. The proposal calls for the use of 34 tons of natural Mo (i.e. 3.3 tons of ^{100}Mo) per module in the form of foil ($\approx 50\text{ mg/cm}^2$). The sensitivity of such a module to the neutrino mass could be of the order of $\approx 0.05\text{ eV}$ [44].

Using future large-scale Yb-loaded liquid scintillation detectors for solar neutrino spectroscopy [45] it is supposed to search for $2\beta^-$ decay of ^{176}Yb ($Q_{\beta\beta} = 1087\text{ keV}$) and $\varepsilon\beta^+$ decay of ^{168}Yb ($Q_{\beta\beta} = 1422\text{ keV}$). With about 20 tons of natural Yb (≈ 2.5 tons of ^{176}Yb) the limit $T_{1/2}^{0\nu} \geq 10^{26}\text{ yr}$ could be set on $0\nu 2\beta$ decay of ^{176}Yb ($m_\nu \leq 0.1\text{ eV}$) [46].

Recently a project CAMEO has been suggested [47], where the unique features (superlow background and large sensitive volume) of the CTF (already existing device) and the BOREXINO set up (under construction) are used to study ^{116}Cd . It is supposed that $\approx 100\text{ kg}$ of enriched $^{116}\text{CdWO}_4$ crystal scintillators will be placed in the liquid scintillator of the CTF. The calculated sensitivity of the CAMEO experiment (in terms of the $T_{1/2}^{0\nu}$ limit) is $\approx 10^{26}\text{ yr}$, which translates to the neutrino mass bound $m_\nu \leq 0.06\text{ eV}$. Similarly with one ton of $^{116}\text{CdWO}_4$ crystals located in the BOREXINO apparatus the constraint on the neutrino mass can be pushed down to $m_\nu \leq 0.02\text{ eV}$ [47].

There are also two proposals (MAJORANA [48] and GENIUS [49]) for the 2β decay quest of ^{76}Ge ($Q_{\beta\beta} = 2039\text{ keV}$). In the MAJORANA proposal it is scheduled to use around 500 kg of HP Ge semiconductor detectors (enriched in ^{76}Ge to $\approx 86\%$) in more or less standard cryostats and set up [48]. The GENIUS project intends to operate one ton of “naked” HP Ge (enriched in ^{76}Ge to $\approx 86\%$) detectors placed in an extremely high-purity liquid nitrogen, simultaneously serving as cooling medium and shielding for the detectors. Due to the latest, the very low background could be reached, which together with a large amount of ^{76}Ge will result in a sensitivity enhancement up to the level of $m_\nu \leq 0.01\text{ eV}$ [49].

Therefore, one can conclude that 2β decay research is really entering a new era of the ultimate sensitivity experiments. However, it is obvious that all mentioned projects (except CAMEO and MAJORANA, which both have no technical risk) require a significant amount of R&D to demonstrate their feasibility, thus the strong efforts and perhaps long time will be needed before their approval. To this effect, let us consider “pro et contra” of the GSO crystals scintillator for the advanced double β decay study of ^{160}Gd .

It is evident on the basis of present experiment, that current level of sensitivity is limited mainly by the intrinsic contamination of the GSO crystal (^{232}Th , ^{235}U and ^{238}U families), therefore further development (which is in progress now) to remove these impurities from the crystals is of most importance. Careful purification of raw materials from actinides and their daughters (technically available now) could decrease radioactive contaminations of the GSO crystals by two to three orders of magnitude down to the level of several $\mu\text{Bq/kg}$.¹⁰ Another possibility to reduce the background is pulse-shape discrimination. Our preliminary measurement shows the difference of decay times for pulses induced in GSO by α particles and γ quanta: 50.8(4) and 48.3(4) ns, respectively.¹¹ Although this difference is not so large, the proper pulse shape discrimination technique, which is under development now, would allow us to eliminate α background from the decays of ^{147}Sm , ^{152}Gd and members of ^{232}Th , ^{235}U and ^{238}U chains.

On the other hand, due to the high abundance of ^{160}Gd ($\approx 22\%$) the GSO crystals could be grown up from natural Gd, thus such detectors should be well less expensive than those made of enriched 2β decay candidate isotopes. Therefore, the realization of the high-sensitivity experiment with ^{160}Gd would be possible by using the GSO multicrystal array with a total mass of about 2000 kg (≈ 400 kg of ^{160}Gd).

The background of such a detector could be reduced further by placing these crystals into a high-purity liquid (water or scintillator)¹² serving as shield and light guide simultaneously, as it has been recently proposed in CAMEO project [47]. Moreover, the strong dependence of the light collected by each PMT versus coordinate of the emitting source in the crystal has been found. Such a dependence is explained by difference of the refraction indexes of crystal ($n = 2.3$ for CdWO_4) and liquid scintillator ($n = 1.58$), which leads to the redistribution between reflected and refracted light due to change of the source position. By means of the GEANT Monte Carlo simulation it was shown that spatial resolution of 1–5 mm (depending on the event location and the energy deposition) can be reached with CdWO_4 crystals ($\varnothing 7 \times 9$ cm) placed in the liquid scintillator of the CTF and viewed by 200 distant PMTs [47]. With the GSO detectors (refractive index $n = 1.85$) placed in a liquid ($n \approx 1.5$), the simulation of light propagation gives the spatial resolution in the range of 4–10 mm. In any case, it would certainly allow the reduction in background in the energy region of interest (roughly by factor of 10–50).

We estimate that sensitivity of the experiment with about two tons of the GSO crystals (placed, for example, in the SNO, KamLand or BOREXINO set ups) and for 5–10 years of exposition would be of the order of $T_{1/2}^{0\nu} \approx 2 \times 10^{26}$ yr, hence the restriction on the Majorana neutrino mass can be reduced down to $m_\nu \leq 0.07$ eV. It is comparable with the sensitivities of the most mentioned above projects for 2β decay study, whose results could

¹⁰ Such a radiopurity has been already reached in the CdWO_4 crystal scintillators used for the 2β decay study of ^{116}Cd [24,50,51].

¹¹ These results are in a good agreement with Ref. [52].

¹² The existing and future large underground neutrino detectors (SNO [53], BOREXINO [42], KamLand [54]) could be appropriate for our proposal with one-two tons of the GSO crystals. The latest located in the water or liquid scintillator would be homogeneously spread out on a sphere with diameter 3–4 m and viewed by the distant PMTs.

provide crucial tests of the certain key problems and theoretical models of the modern astroparticle physics.

Moreover, the experiment with superlow background GSO crystal scintillators could be of great interest for the solar neutrino spectroscopy with ^{160}Gd . It is because that solar neutrino capture by ^{160}Gd (with the low energy threshold) can be easily distinguished from background due to highly specific time signature of this reaction [45]. Hence, if all mentioned developments concerning intrinsic contamination of the GSO crystals and scheduled background improvement would be successfully fulfilled, even larger experiment with ≈ 50 t of the GSO crystals (needed for the solar neutrino spectroscopy) is become available [45]. The latest would further enhance the sensitivity of the by-product $0\nu 2\beta$ decay study with ^{160}Gd .

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