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

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Abstract

The $2\beta$ decay study of $^{160}$Gd has been performed in the Solotvina Underground Laboratory with the help of Gd$_2$SiO$_5$:Ce crystal scintillator (volume 95 cm$^3$). The background of the detector in the vicinity of the $Q_{2\beta}$ energy of $^{160}$Gd was reduced to 1.0 cpd/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: $T_{0^{+}/2}^{0\nu} \geq 2.3(1.3) \times 10^{21}$ yr at 68%(90%) C.L. The $T_{1/2}$ bounds have been also set for $2\nu$, $0\nu \chi$ and $0\nu \chi \chi$ modes of $^{160}$Gd decay, as well as for different $2\beta$ decay processes in $^{136}$Ce, $^{138}$Ce and $^{142}$Ce.

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1 Introduction

An exceptional interest to the neutrinoless double beta ($0\nu 2\beta$) decay is explained by the great potential of this process – which violates the lepton number conservation – to search for the neutrino mass ($m_\nu$) and its nature as a sign of a possible new physics beyond the standard model (SM) [1, 2, 3, 4, 5, 6]. The absence of the $0\nu 2\beta$ decay, established at the required level of sensitivity, yields strong restrictions on $m_\nu$, 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 [1, 2, 3, 4].

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 $2\beta$ 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) [4, 5], $^{130}$Te (low temperature bolometers TeO$_2$) [5], $^{116}$Cd (enriched $^{116}$CdWO$_4$ scintillators) [5], 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 allows to extend the number of $2\beta$ candidates studied with a high sensitivity.

During last years cerium-doped gadolinium silicate Gd$_2$SiO$_5$:Ce (GSO) crystal scintillators have been developed [13, 14]. These scintillators are non-hygrosopic and have a large density (6.71 g/cm$^3$), fast response (primary decay time about 30–60 ns), quite high light output (20% of NaI(Tl), wavelength of emission maximum 440 nm). Moreover, it was already demonstrated [15, 16, 17] that GSO crystals can be applied for the $2\beta$ decay search of $^{160}$Gd, which is one of the interesting candidate nucleus due to the following reasons. First, despite rather low $Q_{\beta\beta} = 1729.7(13)$ 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 equal measured $T_{1/2}^{0\nu}$ limits the experiment with $^{160}$Gd will yield the more stringent restrictions on the neutrino mass and other parameters of the theory. Secondly, recent calculation [20] shows that two-neutrino $2\beta$ decay of $^{160}$Gd is strongly forbidden due to heavy deformation of this nucleus. Meanwhile, the suppression of the $0\nu2\beta$ decay mode would be not so strong due to different sets of intermediate states involved in both transitions. Therefore the energy region of $0\nu2\beta$ signal of $^{160}$Gd could be free of the background from $2\nu2\beta$ decays, which is very serious problem for $2\beta$ detectors with the poor energy resolution [2]. Thirdly, the natural abundance of $^{160}$Gd is rather large (21.86% [21]) allowing to build up the high sensitive apparatus with natural GSO crystals.

The present paper describes the new and further improved half-life limits on $2\beta$ decay of $^{160}$Gd obtained with the help of 95 cm$^3$ GSO crystal scintillator and with about 3 times larger running time than in ref. [22], where preliminary results of this experiment have been already published.

## 2 Set up, background measurements and data analysis

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 $3.951 \times 10^{23}$. The first 630 h of measurements had been carried out with the mass of the crystal equal to 698 g, then its side surface had been ground on 1–1.5 mm.

The experiment was performed in the Solotvina Underground Laboratory (SUL) of the INR in a salt mine 430 m underground ($\approx$1000 m w. e., cosmic muon flux $1.7 \times 10^{-6}$ cm$^{-2}$ s$^{-1}$, neutron flux $\leq 2.7 \times 10^{-6}$ cm$^{-2}$ s$^{-1}$, radon concentration in air $< 30$ Bq m$^{-3}$) [23]. In the low background installation the GSO crystal is viewed by the photomultiplier FEU-110 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 $\gamma$ lines of $^{22}$Na, $^{137}$Cs, $^{207}$Bi, $^{226}$Ra, $^{232}$Th and $^{241}$Am sources. As an example, the resolution equals 16.8%, 13.5%, 11.2% and 10.7% at the energy 662, 1064, 1770 and 2615 keV, respectively. In course of measurement the energy calibration was carried out with $^{207}$Bi source weekly. 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. Event-by-event data acquisition system consists of the IBM PC compatible personal computer and CAMAC crate with electronic units, which allow to record the amplitude (energy) and arrival time of each event [24].

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2For example, the first interesting results were obtained recently for $2\beta$ decay processes in $^{40}$Ca and $^{46}$Ca with the help of newly developed low radioactive CaF$_2$(Eu) crystal scintillators [12].
Total statistics collected in the experiment is 13949 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: the clear peak at the energy 420 keV, the comparatively wide peak at the energy around 1050 keV and two broad distributions dropped down at the energies 2.4 and 5.5 MeV. Taking into account the relative light yield for α particles as compared with that for electrons (α/β ratio) for the GSO scintillator, the first peak is attributed to α particles of $^{152}\text{Gd}$ ($T_{1/2} = 1.08 \times 10^{14}$ yr; $E_{\alpha} = 2140$ keV; abundance $\delta = 0.20\%$) and $^{147}\text{Sm}$ ($T_{1/2} = 1.06 \times 10^{11}$ yr; $E_{\alpha} = 2233$ keV; $\delta = 15\%$; samarium can be present as impurity of the GSO crystal at the level of $\approx 8$ ppm [15]). The peak near 1050 keV as well as the broad distribution up to the energy 2.4 MeV is mainly due to the radioactive contamination of the crystal by the nuclides from the $^{232}\text{Th}$, $^{235}\text{U}$ and $^{238}\text{U}$ families. The second distribution up to the energy 5.5 MeV is caused by decays of $^{232}\text{Th}$ daughter isotopes: a) β decay of $^{208}\text{Tl}$ ($Q_\beta = 5.00$ MeV); b) β decay of $^{212}\text{Bi}$ ($Q_\beta = 2.25$ MeV) followed by α decay of its daughter $^{212}\text{Po}$ ($T_{1/2} = 0.3$ µs; $E_{\alpha} = 8.78$ MeV or $\approx 2.7$ MeV in β scale).

![Figure 1: Background spectrum of the GSO crystal (95 cm$^3$) collected during 13949 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 $^{226}\text{Ra}$, $^{227}\text{Ac}$ and $^{228}\text{Th}$ intrinsic contamination of the crystal (filled circles) and fitting curve (solid line).](image)

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 fulfilled (as described in details 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 $^{214}\text{Po}$, $^{215}\text{Po}$, $^{216}\text{Po}$ and $^{220}\text{Rn}$ from the internal contamination of the crystal as following: $\alpha/\beta = 0.152 + 0.01765 \cdot E_{\alpha}$, where $E_{\alpha}$ is in MeV. The mentioned peaks were selected from the background with the help of the time-amplitude analysis described below.

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3The energy dependence of the α/β ratio was determined by using the α peaks of $^{214}\text{Po}$, $^{215}\text{Po}$, $^{216}\text{Po}$ and $^{220}\text{Rn}$ from the internal contamination of the crystal as following: $\alpha/\beta = 0.152 + 0.01765 \cdot E_{\alpha}$, where $E_{\alpha}$ is in MeV. The mentioned peaks were selected from the background with the help of the time-amplitude analysis described below.
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 $\alpha$ particles corresponds to 1.7 MeV in $\beta$ scale of 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 $\alpha$ 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 selected spectra and time distribution are in the excellent agreement with those expected from $\alpha$ particles of $^{220}$Rn and $^{216}$Po. Taking into account the efficiency of the time-amplitude analysis, the number of accidental coincidences, and the interfered chain of $^{219}$Rn $\rightarrow$ $^{215}$Po (it is selected by the applied procedure with 2.04% efficiency), the $^{228}$Th activity in the GSO crystal was determined as 2.287(13) mBq/kg. Then on the next step of analysis the fast couples found ($^{220}$Rn and $^{216}$Po) were used as triggers to search for preceding $\alpha$ 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). 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 $\alpha$ peak of $^{224}$Ra.

Figure 2: The energy spectra of the first and second $\alpha$ particles in the decay chain $^{220}$Rn $\rightarrow$ $^{216}$Po $\rightarrow$ $^{212}$Pb which were found with 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 [23]) and background (dotted line).

The same technique was applied to the sequences of $\alpha$ 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) was used as a trigger.
decays with energy between 1.5 and 2.6 MeV) was chosen\(^4\). The obtained \(\alpha\) peaks correspond to activity of 0.948(9) mBq/kg for the \(^{227}\)Ac impurity in the crystal. Then the procedure analogous to that described for the \(^{224}\)Ra was applied to find the preceding \(^{223}\)Ra \(\alpha\) decays.

For the analysis of the \(^{226}\)Ra chain (\(^{238}\)U family) the following sequence of \(\beta\) and \(\alpha\) decays was used: \(^{214}\)Bi \((Q_\beta = 3.27\) MeV, \(T_{1/2} = 19.9\) m) \(\rightarrow\) \(^{214}\)Po \((Q_\alpha = 7.83\) MeV, \(T_{1/2} = 164.3\) \(\mu\)s) \(\rightarrow\) \(^{210}\)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 \(2 - 500\) \(\mu\)s \((\eta_t = 0.872\) 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 \(\eta_E\) is equal to 1). By the Monte Carlo simulation the part \(\eta_E\) 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\(^5\) 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.

Figure 3: The energy spectra of the sequence of \(\beta\) and \(\alpha\) decays in the decay chain \(^{214}\)Bi \(\rightarrow\) \(^{214}\)Po \(\rightarrow\) \(^{210}\)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 \(T_{1/2} = 129\) \(\mu\)s (table value \(T_{1/2} = 164.3(20)\) \(\mu\)s [25]) and exponent with \(T_{1/2} = 1.78\) ms related with the chain \(^{219}\)Rn \(\rightarrow\) \(^{215}\)Po \(\rightarrow\) \(^{211}\)Pb (dotted line).

Besides determination of the background components, the time-amplitude analysis is used

\(^4\)The used procedure selects also pairs \(^{220}\)Rn \(\rightarrow\) \(^{216}\)Po and \(^{214}\)Bi \(\rightarrow\) \(^{214}\)Po with efficiencies 4.64\% and 12.08\%, respectively. These contributions are taken into account in the calculation of activities.

\(^5\)Peak observed in the \(\beta\) spectrum of \(^{214}\)Bi (see fig. 3) is the part (17.7\%) of \(^{219}\)Rn \(\alpha\) decays (from \(^{235}\)U family), which corresponds to the chosen time interval 2–500 \(\mu\)s. Due to known activity of \(^{227}\)Ac the contribution of \(^{219}\)Rn \(\rightarrow\) \(^{215}\)Po (as well as 0.24\% of the chain \(^{220}\)Rn \(\rightarrow\) \(^{216}\)Po) was calculated accurately and subtracted from the activities of \(^{214}\)Bi and \(^{214}\)Po.
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 numerate events in the chain):

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

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

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

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

(iii) In the third cut a fast chain of decays \(^{220}\text{Rn} - ^{216}\text{Po}\) is used as a trigger to select the previous event of \(^{224}\text{Ra}\). The parameters taken for \(^{224}\text{Ra}\) selection are the following: \( E_1 > 1000 \text{ keV}; \Delta t_{12} = 1 - 30 \text{ s}; E_2 = 1200 - 2200 \text{ keV}; \Delta t_{23} = 0.01 - 0.5 \text{ s}; E_3 = 1300 - 2400 \text{ keV}. \) The final efficiency for selection of \(^{224}\text{Ra}\) is \( \eta = 0.258 \).

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

Since the time windows of the cuts are not overlapping, the probability that one event could be selected more than once is rather low. Indeed, from the total number of 345169 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 yr·kg of exposure (\( \approx 13400 \text{ h} \)), 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 \( 0\nu2\beta \) decay of \(^{160}\text{Gd} (1648 – 1856 \text{ keV})\) has been reduced by 2.3 times up to the value of 1.01(1) cpd/keV·kg.

### 3 Background simulation

With the aim to evaluate the \( 2\beta \) decay processes in \(^{160}\text{Gd} \) 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 de-excitation process \(^{25}\). The possibilities of emission of conversion electrons and \( e^+ e^- \) pairs instead of \( \gamma \) 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 fitted 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 \(^{228}\text{Th}, ^{227}\text{Ac}\) and \(^{226}\text{Ra}\) (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 $^{224}\text{Ra}$, $^{220}\text{Rn}$, $^{216}\text{Po}$, $^{223}\text{Ra}$, $^{219}\text{Rn}$, $^{215}\text{Po}$; $^{214}\text{Bi}$ and $^{214}\text{Po}$ 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, $^{228}\text{Th}+\text{daughters}$ 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 $^{232}\text{Th}$, $^{235}\text{U}$ and $^{238}\text{U}$ 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, $^{232}\text{Th}$, $^{228}\text{Ra}$, $^{235}\text{U}+^{238}\text{U}+^{234}\text{U}$, $^{231}\text{Pa}$, $^{230}\text{Th}$, and $^{210}\text{Pb}$) were taken as free parameters for the fitting procedure.

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

Besides, simulated spectra of $^{40}\text{K}$ and $^{138}\text{La}$ – natural radionuclides which could be present in the GSO crystal – were included into the fitting procedure too. The last component of the background model is the exponential function (with two free parameters) which describes the residual external background (multiple scattering of $\gamma$ quanta, influence of weak neutron flux and so on). The exponential behavior of this component was confirmed by the measurements with the high radiopurity $\text{CdWO}_4$ 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; $^{232}\text{Th} \leq 6.5$ 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 $2\beta$ decay of $^{160}\text{Gd}$

Since in the measured spectrum the $0\nu2\beta$ decay peak of $^{160}\text{Gd}$ is evidently absent, only the limit for the probability of this process can be set on the base of the experimental data. To estimate the half-life limit $T_{1/2}$, we use the formula $T_{1/2} = \ln 2 \cdot \eta \cdot N \cdot t / \lim S$, where $\eta$ is the detection efficiency, $N$ is the number of $^{160}\text{Gd}$ nuclei, $t$ is the measuring time and $\lim S$ is the number of effect’s events which can be excluded with a given confidence level. To calculate the values of $\eta$ 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 $0\nu2\beta$ decay the response function is the Gaussian centered at 1730 keV and its width (FWHM) equals 176 keV. The edge effects (escape of one or both electrons and bremsstrahlung quanta...
from the crystal) remove from the peak $\approx 5\%$ of events, thus $\eta = 0.95$. The $\lim S$ values were determined in two ways. First, by using the so called ”one $\sigma$ approach”, in which the excluded number of effect’s events is estimated simply as square root of the number of background counts in a suitably chosen energy window $\Delta E$. 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 74500 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 $\lim S$ value 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 effect’s peak being sought. It should be stressed that for the energy interval of interest the main important contributions ($\approx 73\%$ of the experimental spectrum within $1600 - 1900$ keV) are the activities of $^{226}$Ra, $^{227}$Ac and $^{228}$Th (and their short-lived daughters) from the intrinsic contamination of the GSO crystal, whose values were determined accurately (better than 1% uncertainty).

As a result of the fit procedure in the energy region 1.3–2.1 MeV, the obtained area for the $0\nu 2\beta$ decay peak is $-160 \pm 233$ counts ($\chi^2$ 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 number of effect’s events, which can be excluded with 90%(68%) C.L. was calculated \cite{29} as 298(169). It gives the half-life limit:

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

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

The same $T_{1/2}$ limit has been set for the $0\nu 2\beta$ transition to the first excited level of $^{160}\text{Dy} (2^+, 87 \text{ keV})$ because $87 \text{ keV } \gamma$ quanta following this process will be almost fully absorbed inside the scintillator. The obtained limits are several times larger than those already published [15, 16, 17, 22].

The two neutrino $2\beta$ decay rate was evaluated in two ways. For the 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 $\text{lim } S = 7.6 \times 10^5 \text{ events corresponding to the } T_{1/2}^{2\nu} \text{(g.s.) } \geq 5.5 \times 10^{17} \text{ yr (99\% C.L.) for } 2\nu 2\beta \text{ decay of } ^{160}\text{Gd.}$ However, within this simple approach it was impossible to reproduce adequately the measured spectrum, thus the background model and the fitting procedure described above were applied for the $2\nu$ decay mode too. The set of fits were performed by changing the fitting energy region from $(100 – 760) \text{ keV to } (2400 – 3000) \text{ keV.}$ The maximum value of an excluded effect ($\text{lim } S = 2.2 \times 10^8 \text{ events at 90\% C.L.}$) was found for the energy interval $760 – 2600 \text{ keV.}$ The corresponding Monte Carlo simulated spectrum of $2\nu 2\beta$ decay of $^{160}\text{Gd (g.s. } \rightarrow \text{g.s.)}$ 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:

![Figure 5: The background spectrum of the GSO detector for 0.969 yr-kg 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 ($^{40}\text{K}$ and sum of $^{238}\text{U}, ^{235}\text{U}, ^{232}\text{Th}$) and external ($\gamma$ radiation from PMT) components of background are shown. The excluded with 90\% C.L. distribution of $2\nu 2\beta$ decay of $^{160}\text{Gd to the ground level of } ^{160}\text{Dy corresponds to the half-life limit } T_{1/2}^{2\nu} \text{(g.s.) } = 1.9 \times 10^{19} \text{ yr.}
\[ T_{1/2}^{2\nu}(\text{g.s.}) \geq 1.9(3.1) \times 10^{19} \text{ yr} \quad \text{at 90\% (68\%) C.L.} \]

The same method gives the limit for \( 2\nu2\beta \) transition to the first \( (2^+) \) excited level of \( ^{160}\text{Dy} \):

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

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

\[
\begin{align*}
T_{1/2}^{0\nu} &\geq 3.5(5.3) \times 10^{18} \text{ yr} \quad \text{at 90\% (68\%) C.L.} \\
T_{1/2}^{0\nu\chi} &\geq 1.3(2.0) \times 10^{19} \text{ yr} \quad \text{at 90\% (68\%) C.L.}
\end{align*}
\]

## 5 Limits on \( 2\beta \) 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 to search for the \( 2\beta \) processes in three cerium isotopes: double positron decay (\( 2\beta^+ \)), or electron capture and positron decay (\( \epsilon\beta^+ \)), or double electron capture (\( 2e \)) in \( ^{136}\text{Ce} \) (mass difference between parent and daughter atoms \( \Delta M_A = 2397(48) \text{ keV} \); abundance of parent nuclide \( \delta = 0.185\% \)); double electron capture in \( ^{138}\text{Ce} \) (\( \Delta M_A = 693(11) \text{ keV} \); \( \delta = 0.251\% \)); and \( 2\beta^- \) decay in \( ^{142}\text{Ce} \) (\( \Delta M_A = 1417(2) \text{ keV} \); \( \delta = 11.114\% \)).

The total numbers of \( ^{136}\text{Ce} \), \( ^{138}\text{Ce} \) and \( ^{142}\text{Ce} \) nuclei in the crystal are \( 4.1 \times 10^{19}, 5.4 \times 10^{19} \) and \( 2.4 \times 10^{21} \), respectively. The response functions of the detector for the different possible \( 2\beta \) 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 \( ^{136}\text{Ce} \) are presented in fig. 6. Limits on half-lives with respect to different modes of \( 2\beta \) processes of cerium isotopes were calculated in the way analogous to that for \( ^{160}\text{Gd} \) – they are summarized in the Table. Except two limits for \( 0\nu2\beta^+ \) decay of \( ^{136}\text{Ce} \) and \( 0\nu2\beta^- \) decay of \( ^{142}\text{Ce} \) (already obtained by using CeF\(_3\) scintillators \[3\]), all presented results for cerium isotopes are set for the first time.

## 6 Conclusions

The present experiment performed in the Solotvina Underground Laboratory with the help of GSO scintillator has shown “pro et contra” of these crystals for the advanced double \( \beta \) decay study of \( ^{160}\text{Gd} \).

The current level of sensitivity is limited mainly by the intrinsic contamination of the GSO crystal (\( ^{232}\text{Th}, ^{235}\text{U} \) and \( ^{238}\text{U} \) families), therefore further development (which is in progress now) to remove these impurities from the crystals is at 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-three orders of magnitude down to the level of several \( \mu\text{Bq/kg} \).[8] Another possibility to reduce background is the pulse-shape

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8It should be noted that shape of distribution for \( 2\nu2\beta \) decay to \( 2^+ \) level differs from \( 2\nu2\beta \) transition to ground \( (0^+) \) state.[8]

9Such a radiopurity has been already reached in the CdWO\(_4\) crystal scintillators used for the \( 2\beta \) decay study of \( ^{116}\text{Cd} \).[24, 21, 82].

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Figure 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.

discrimination. Our preliminary measurement shows the difference of decay times for pulses induced in GSO by $\alpha$ particles and $\gamma$ quanta: 50.8(4) and 48.3(4) ns, respectively\textsuperscript{10}. Although this difference is not so large, the proper pulse shape discrimination technique, which is under development now, would allow to eliminate $\alpha$ background from decays of $^{147}$Sm, $^{152}$Gd and members of $^{232}$Th, $^{235}$U and $^{238}$U families.

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 $2\beta$ decay candidate isotopes. Therefore, the enforcement of the large scale and high sensitivity experiment with $^{160}$Gd would be really possible by using the GSO multi-crystal array with total mass of about 2000 kg ($\approx$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)\textsuperscript{11} serving as shield and light guide simultaneously. The similar idea has been recently proposed in CAMEO project\textsuperscript{37} with aim to study $2\beta$ decay of $^{116}$Cd by using $^{116}$CdWO\textsubscript{4} crystals placed in the liquid scintillator of the BOREXINO Counting Test Facility (CTF). It was evidently demonstrated by pilot measurements with $^{116}$Cd\textsuperscript{38} and by Monte Carlo simulation that sensitivity of the CAMEO experiment (in term of the $T_{1/2}$ limit for $0\nu2\beta$ decay) with $\approx$ 100 kg of $^{116}$CdWO\textsubscript{4} crystals is $\approx$10\textsuperscript{26} yr which translates to the constraint on the neutrino mass $m_\nu \leq$ 0.06 eV\textsuperscript{37}. Moreover, the strong dependence of the

\textsuperscript{10}These results are in a good agreement with ref. \textsuperscript{33}.

\textsuperscript{11}The existing and future large underground neutrino detectors (SNO\textsuperscript{34}, BOREXINO\textsuperscript{32}, KamLand\textsuperscript{36}) 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.
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 \((n = 2.3 \text{ for CdWO}_4)\) 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 deposit) can be reached with CdWO_4 crystals \((\varnothing 7 \times 9 \text{ cm})\) placed in the liquid scintillator of the CTF and viewed by 200 distant PMTs \[37\]. 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. Anyhow, it would certainly allow to reduce background in the energy region of interest additionally (roughly by factor of 10–50).

We estimate that sensitivity of the experiment with about two tons of the GSO crystals (placed in the SNO or BOREXINO set ups) and for 5–10 years of exposition would be of the order of \(T_{1/2}^{\nu} \approx 2 \times 10^{26} \text{ yr}\), hence the restriction on the Majorana neutrino mass can be reduced down to \(m_\nu \leq 0.07 \text{ eV}\). It is comparable with the sensitivities of the recently proposed large scale projects for \(2\beta\) decay study, like MOON \[11\], EXO \[11\], CAMEO \[37\], CUORE \[38\], MAJORANA \[12\], GENIUS \[39\], whose results could provide crucial tests of the certain key problems and theoretical models of the modern astroparticle physics. Note, however, that cost of the GSO experiment would be well lower than those of mentioned projects.

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Table 1: Half-life limits on the $2\beta$ decay processes in $^{160}\text{Gd}$, $^{136}\text{Ce}$, $^{138}\text{Ce}$ and $^{142}\text{Ce}$

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