ELSEVIER Nuclear Physics B (Proc. Suppl.) 48 (1996) 235–237

NUCLEAR PHYSICS B
PROCEEDINGS
SUPPLEMENTS

Quest for neutrinoless double beta decay of ^{160}Gd

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The experiment with a large (≈95 cm^3) Gd_2SiO_5:Ce crystal scintillator has been performed in the Solotvina Underground Laboratory to search for neutrinoless double beta decay of ^{160}Gd. The background rate was reduced to a level of 0.045 counts/h/keV/kg in the vicinity of $\beta\beta$ decay energy of ^{160}Gd (1730 keV). The half-life limits have been set for $0\nu\beta\beta$ decay of ^{160}Gd: $T^{0\nu}_{1/2}$ (g.s.→ g.s.) $\geq 8.2\cdot10^{20}$ y (90% CL); $T^{0\nu}_{1/2}$ $(0^+\rightarrow2^+) \geq 8.2\cdot10^{20}$ y (90% CL).

1. DETECTOR, MEASUREMENTS AND ANALYSIS OF BACKGROUND

The $\beta\beta$ decay energy of ^{160}Gd equals 1729.7(13) keV [1], natural abundance of ^{160}Gd is large as 21.86(4)% [2]. Calculated $T_{1/2}$ for $2\nu\beta\beta$ decay of ^{160}Gd is in the range of $5\cdot10^{18}$ - 10^{21} y [3]. It is known that background from $2\nu\beta\beta$ decay in the region of $0\nu\beta\beta$ signal is very serious problem for detectors with poor energy resolution [4]. In recent work [5] it was shown that $2\nu\beta\beta$ decay of ^{160}Gd is strongly forbidden due to heavy deformation of ^{160}Gd. If so the background from $2\nu\beta\beta$ mode will be absent. The calculated value of $T^{0\nu}_{1/2}\cdot\langle m_\nu\rangle^2 = 8.6\cdot10^{23}$ y·eV^2 is nearly three times lower than for ^{76}Ge and ^{136}Xe [3].

Cerium-doped gadolinium silicate Gd_2SiO_5:Ce (GSO) crystal grown by Czochralski method with a mass of 698 g (volume of 104 cm^3) was used in the measurements. The GSO scintillator has a large density (6.71 g/cm^3), fast response (decay constant is equal to 40-60 ns) and quite large light output (20% of NaI(Tl); peak emission - 440 nm). The number of ^{160}Gd nuclei in this crystal is $4.346\cdot10^{23}$. The energy resolution of 12% at energy of 662 keV was obtained with photomultiplier XP2412 (Philips). Experiment has been carried out in the Solotvina Underground Laboratory of INR built in the salt mine on the depth more than 1000 m w.e. where cosmic muon flux is suppressed by a factor of 10^4 [6]. In the low background installation the GSO crystal is viewed by PMT (FEU-110) through a plastic light-guide 18 cm long. The energy resolution in these conditions is little worse: 15.5%, 12.3%, 9.3% and 7.9% at energy 662, 1064, 1770 and 2615 keV, respectively. The passive shielding of OFHC copper (5 cm), mercury (7 cm) and lead (15 cm) surrounds the GSO scintillator to reduce the external background. The data acquisition system consists of a microcomputer, a magnetic tape recorder and a CAMAC crate with electronic units which allow to record the amplitude and arrival time of each event. The energy calibration has been performed with ^{207}Bi weekly.

The background spectrum measured during 5421 h is shown in Fig. 1 (upper line) where the following peculiarities exist: the clear peak at the energy 425(15) keV, the weak and comparatively wide peak at the energy around 1050 keV and the broad distribution up to the energy 2400 keV. The first peak can be attributed to α particles of ^{152}Gd ($T_{1/2} = 1.08(8)\cdot10^{14}$ y, E_α=2140(30) keV, natural abundance of ^{152}Gd is 0.20%) if the α/β ratio for GSO crystal (equal to 0.20 (2) at this energy) was taken into account. However, the area of this peak is about two times greater than that expected from ^{152}Gd. The difference can be explained if assume the presence of ^{147}Sm ($T_{1/2} = 1.06(2)\cdot10^{11}$ y, E_α=2233(5) keV) in the crystal at the level of 7.6(9) ppm. The peak near 1050 keV as well as the broad distributions up to the energy 2.4 MeV could be due to the radioactive contamination of the crystal by the nuclides from ^{232}Th, ^{235}U and ^{238}U families. The second continuum up to the energy 5.2 MeV could be assigned by the ^{232}Th family: a) $\beta\gamma$ decay of ^{208}Tl; b) β decay of ^{212}Bi (Q_β=2.25 MeV) plus α decay of its daughter ^{212}Po ($T_{1/2}$ = 0.3 µs, E_α=8.95 MeV or ≈ 1.8 MeV in β scale). Due to the short half-life of ^{212}Po its α-line and the ^{212}Bi β-continuum can not be time resolved and result in the observed broad distributions till the energy of ≈4 MeV.

The spectrum in Fig. 1 was accumulated during two runs. The first run (630 h) was measured with crystal of initial volume of 104 cm^3. Then it was assumed that U

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PII: S0920-5632(96)00248-4

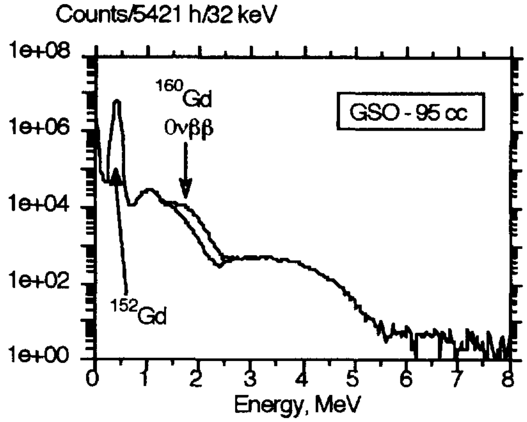

Figure 1. Background spectrum of GSO crystal.

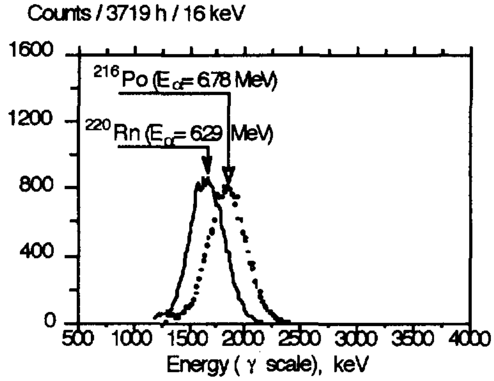

Figure 2. Distributions of α decays of ^{220}Rn and ^{216}Po.

impurities were located in a thin surface layer as it was found in the CdWO$_4$ crystals during the study of ββ decay of ^{116}Cd [7]. The GSO crystal was ground on 1-1.5 mm and its volume was decreased down to 95 cm^3. Nevertheless the reduction of the background had not been achieved and the rest part of statistics was accumulated with crystal volume of 95 cm^3.

To recognize and reduce the background from contamination of the crystal the of-line analysis of time distribution of measured events was developed and fulfilled. For example, the sequence of two α decays belonging to ^{232}Th family was searched for:
^{220}Rn (E_α=6.29 MeV, $T_{1/2}$=55.6 s) →
^{216}Po (E_α=6.78 MeV, $T_{1/2}$=146 ms) → ^{212}Pb.

Because α particle energy of ^{220}Rn corresponds to 1.65 MeV in β scale the background events with the energy in the interval 1.2 - 2.2 MeV were used as triggers. Then all events following the triggers in the time interval 10 ms - 500 ms (it contains ≈86% of all decays from ^{216}Po) were selected. The obtained spectra (Fig. 2) are in excellent agreement with what expected from α particles of ^{220}Rn and ^{216}Po decay (their energy has to be equal to 1.65 and 1.83 MeV in β scale). The distribution of the time interval between the first and second α particles is also in clear accordance with expected half-life value of 146 ms. Using these results and taking into account the efficiency and number of random coincidences, the ^{228}Th content was found to be equal to 2.79(5) mBq/kg. The same technique was applied for the sequences of α decays belonging to the ^{235}U family: (E_α≈5.6 MeV, $T_{1/2}$=11.4 d) → ^{219}Rn (E_α=6.82 MeV, $T_{1/2}$=3.96 s) → ^{215}Po (E_α=7.39 MeV, $T_{1/2}$=1.78 ms) → ^{211}Pb. The ^{227}Ac content was found to be equal to 0.995(23) mBq/kg.

For ^{226}Ra family the sequence of β and α decays was used: ^{214}Bi (Q_β=3.27 MeV, $T_{1/2}$=19.9 m) → ^{214}Po (E_α=7.69 MeV, $T_{1/2}$=164 μs) → ^{210}Pb, which leads to the ^{226}Ra activity in the crystal equal to 0.53(3) mBq/kg.

All selected events were excluded from the measured spectrum of the GSO crystal. The resulting spectrum is shown in Fig. 1 (lower line) and corresponds to 3469 h×kg of exposure, if the total efficiencies of analysis and random coincidences were taken into account. The background rate in the region of 0νββ decay of ^{160}Gd (1648 - 1813 keV) was reduced to 0.045 counts/h·kg·keV.

2. THE HALF LIFE LIMITS ON 0νββ DECAY OF ^{160}Gd

The data (Fig. 1) were used for evaluations of 0νββ half-life. The product of the number of ^{160}Gd nuclei by measuring time is equal to 2.469 · 10^{23} nuclei·y. The half-life limits were calculated by known formula:

$$\lim T_{1/2} = \ln 2 \cdot \varepsilon \cdot t \cdot N_n / \lim S_e,$$

where N_n is the number of ^{160}Gd nuclei, ε is the detection efficiency, t is the measuring time and lim S_e is the number of effect's events which can be excluded with a given confidence level. To calculate the detector efficiency its response function for 0νββ events was simulated by Monte Carlo method with help of programs

[8] and CERN code GEANT 3.14. It was found that the response function is the Gaussian peak with maximum at 1730 keV and FWHM is equal to 165 keV. The edge effects remove from peak 5% of events. The value of lim S_e was evaluated by using two kind of procedure. The simplest "one sigma" estimate based on the background rate (0.045 counts/h·kg·keV), number of nuclei and measuring time (5421 h) leads to the following half-life limit of ^{160}Gd: $T^{0\nu}_{1/2}\geq1.0\cdot10^{21}$ y (68% CL). Also the standard least-squares method [9] was applied in assumption that the experimental spectrum can be described in the region of 760 - 2400 keV by a sum of two functions: $0\nu\beta\beta$ peak of ^{160}Gd and background model. Last one was simulated by code GEANT 3.14 using the firm established contents of the ^{226}Ra, ^{228}Th and ^{227}Ac in the crystal. To describe the other possible contamination a generator for the decays of radioactive nuclides, which may be sources of background, was used [10]. Quantities of possible contamination were used as free parameters in fit. The exponential component was introduced as well to describe the residual background and can be explained by external background. The exponential behavior of the external background was confirmed with high purity $CdWO_4$ crystal (452 g) measured in the same conditions. The parameters of the exponent were also found in the fit procedure and the contribution of this component to the experimental spectrum was revealed to be small (5.3% in the energy interval of 1-2 MeV). The best fit (χ^2=0.88) was achieved in the region of 760 - 2400 keV with following model of the main additional intrinsic contamination of the GSO crystal: ^{40}K- 0.006(1) mBq/kg; ^{228}Ac - 0.8(2) mBq/kg; ^{232}Th - 5.9(3) mBq/kg; ^{235}U - 0.9(1) mBq/kg; ^{230}Th - 11.2(6) mBq/kg; ^{238}U - 2.9(4) mBq/kg. It should be stressed that for the energy interval of the $0\nu\beta\beta$ decay of ^{160}Gd the main important contribution is ^{238}U due to β decay of ^{234m}Pa. The area of this contribution does not exceed 4% of the experimental spectrum in the vicinity of $0\nu\beta\beta$ decay of ^{160}Gd.

The least-squared fit of experimental spectrum by the set of above mentioned functions in the region 1000 - 2400 keV (χ^2=0.86) gives for the area of $0\nu\beta\beta$ peak the values of -11±120 counts. In accordance with [9] it leads to the value of lim S_e = 198(125) counts excluded with the confidence level of 90% (68%). This estimate yields the following limit: $T^{0\nu}_{1/2}\geq8.2(13)\cdot10^{20}$ y - 90(68)% CL. The same half-life limit has been set for $0\nu\beta\beta$ decay of ^{160}Gd to the first excited level of ^{160}Dy because 87 keV γ-rays following this process will be almost fully absorbed inside GSO crystal.

Comparing this limit with theory [3] the restrictions on neutrino mass can be computed: $\langle m_\nu\rangle \leq 32$ eV.

Recently another experiment with large GSO crystal (353 cm^3) was made [11]. The background rate in the energy interval 1650 - 1820 keV is 1.98 counts/h·kg·keV (44 times higher than in our measurements) and obtained result is $T^{0\nu}_{1/2}\geq 3\cdot10^{20}$ y (68% CL).

The main conclusion of present work is the possibility to perform the more sensitive experiment on $\beta\beta$ decay of ^{160}Gd using the GSO crystals. The INR (Kiev) and MPI (Heidelberg) collaboration has recently proposed such a project with approximately one thousand of pure GSO crystals and with full mass of ^{160}Gd about 200 kg. Due to high abundance of ^{160}Gd the GSO crystals could be grown up from natural Gd and as a result the cost of such a multi-detector array should be well less expensive than for experiments based on the enriched isotopes and detectors. The reduction of the internal impurities of the crystals by more than order of magnitude is under progress now. With further improved background of such a multi-crystal array the limit of $T^{0\nu}_{1/2}\approx10^{25}$ y could be reached that corresponds to the restriction of the Majorana neutrino mass less than 0.3 eV which is comparable with the sensitivity of the most advanced experiments with ^{76}Ge [12, 13].

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