

Investigation of $\beta\beta$ decay in ^{150}Nd and ^{148}Nd to the excited states of daughter nuclei

A. S. Barabash,^{1,*} Ph. Hubert,² A. Nachab,² and V. I. Umatov¹¹*Institute of Theoretical and Experimental Physics, B. Cheremushkinskaya 25, RU-117259 Moscow, Russian Federation*²*Centre d'Etudes Nucléaires, IN2P3-CNRS et Université de Bordeaux, F-33170 Gradignan, France*

(Received 5 March 2009; published 10 April 2009)

Double beta decay of ^{150}Nd and ^{148}Nd to the excited states of daughter nuclei have been studied using a 400 cm³ low-background HPGe detector and an external source consisting of 3046 g of natural Nd₂O₃ powder. The half-life for the two-neutrino double beta decay of ^{150}Nd to the excited 0_1^+ state in ^{150}Sm is measured to be $T_{1/2} = [1.33_{-0.23}^{+0.36}(\text{stat})_{-0.13}^{+0.27}(\text{syst})] \times 10^{20}$ yr. For other $(0\nu + 2\nu)$ transitions to the 2_1^+ , 2_2^+ , 2_3^+ , and 0_2^+ levels in ^{150}Sm , limits are obtained at the level of $\sim(2-8) \times 10^{20}$ yr. In the case of ^{148}Nd only limits for the $(0\nu + 2\nu)$ transitions to the 2_1^+ , 0_1^+ , and 2_2^+ excited states in ^{148}Sm were obtained and are at the level of $\sim(4-8) \times 10^{20}$ yr.

DOI: 10.1103/PhysRevC.79.045501

PACS number(s): 23.40.-s, 14.60.Pq

I. INTRODUCTION

The experiments with solar, atmospheric, reactor, and accelerator neutrinos have provided compelling evidence for the existence of neutrino oscillations driven by nonzero neutrino masses and neutrino mixing (see recent reviews [1–3] and references therein). These results are impressive proof that neutrinos have a nonzero mass. However, the experiments studying neutrino oscillations are not sensitive to the nature of the neutrino mass (Dirac or Majorana) and provide no information on the absolute scale of the neutrino masses, since such experiments are sensitive only to the difference of the masses, Δm^2 . The detection and study of neutrinoless double beta ($0\nu\beta\beta$) decay may clarify the following problems of neutrino physics (see discussions in [4–6]): (i) neutrino nature: whether the neutrino is a Dirac or a Majorana particle; (ii) absolute neutrino mass scale (a measurement or a limit on m_1); (iii) the type of neutrino mass hierarchy (normal, inverted, or quasidegenerate); (iv) CP violation in the lepton sector (measurement of the Majorana CP-violating phases).

Double beta decay with the emission of two neutrinos ($2\nu\beta\beta$) is an allowed process of second order in the standard model. The $2\nu\beta\beta$ decays provide the possibility of an experimental determination of the nuclear matrix elements (NME) involved in the double beta decay processes. This leads to the development of theoretical schemes for NME calculations both in connection with the $2\nu\beta\beta$ decays as well as the $0\nu\beta\beta$ decays (see, for example, [7–9]). At present, $2\nu\beta\beta$ decay to the ground state of the final daughter nucleus has been measured for ten nuclei: ^{48}Ca , ^{76}Ge , ^{82}Se , ^{96}Zr , ^{100}Mo , ^{116}Cd , ^{128}Te , ^{130}Te , ^{150}Nd , and ^{238}U (a review of the results is given in Refs. [10–12]).

The $\beta\beta$ decay can proceed through transitions to the ground state as well as to various excited states of the daughter nucleus. Studies of the latter transitions allow one to obtain supplementary information about $\beta\beta$ decay.

Because of smaller transition energies, the probabilities for $\beta\beta$ -decay transitions to excited states are substantially suppressed in comparison with transitions to the ground state. But as was shown in Ref. [13], by using low-background facilities utilizing high purity germanium (HPGe) detectors, the $2\nu\beta\beta$ decay to the 0_1^+ level in the daughter nucleus may be detected for such nuclei as ^{100}Mo , ^{96}Zr , and ^{150}Nd . For these isotopes the energies involved in the $\beta\beta$ transitions are large (1903, 2202, and 2627 keV, respectively), and the expected half-lives are of the order of 10^{20} – 10^{21} yr. The required sensitivity was reached for ^{100}Mo in four independent experiments [14–18]. It was also obtained in ^{150}Nd [19]. Recently additional isotopes (^{82}Se , ^{130}Te , ^{116}Cd , and ^{76}Ge) have also become of interest to studies of the $2\nu\beta\beta$ decay to the 0_1^+ level (see reviews in Refs. [20–22]).

The $0\nu\beta\beta$ transition to excited states of daughter nuclei provide a clear-cut signature of such decays, and is worthy of a special note here. In addition to two electrons with fixed total energy, one ($0^+ \rightarrow 2^+$ transition) or two ($0^+ \rightarrow 0^+$ transition) photons appear, with their energies being strictly fixed. In a hypothetical experiment detecting all decay products with a high efficiency and a high energy resolution, the background can be reduced to nearly zero. This zero background idea will be the goal of future experiments featuring a large mass of the $\beta\beta$ sample (Refs. [20,21,23]). In Ref. [24] it was mentioned that detection of this transition will give us the additional possibility of distinguishing the $0\nu\beta\beta$ mechanisms (the light and heavy Majorana neutrino exchange mechanisms, the trilinear R -parity breaking mechanisms, etc.). So the search for $\beta\beta$ transitions to the excited states has its own special interest.

In this article, results of an experimental investigation of the $\beta\beta$ decay of ^{150}Nd and ^{148}Nd to the excited states in ^{150}Sm and ^{148}Sm are presented. The decay schemes for the triplets ^{150}Nd – ^{150}Pm – ^{150}Sm [25] and ^{148}Nd – ^{148}Pm – ^{148}Sm [26] are shown in Figs. 1 and 2, respectively. The search for $\beta\beta$ transitions of ^{150}Nd and ^{148}Nd to excited states has been carried out using a HPGe detector to look for γ -ray lines corresponding to their decay schemes. A preliminary result for $\beta\beta$ decay of ^{150}Nd to the 0_1^+ excited state of ^{150}Sm was published in Ref. [19].

*barabash@itep.ru

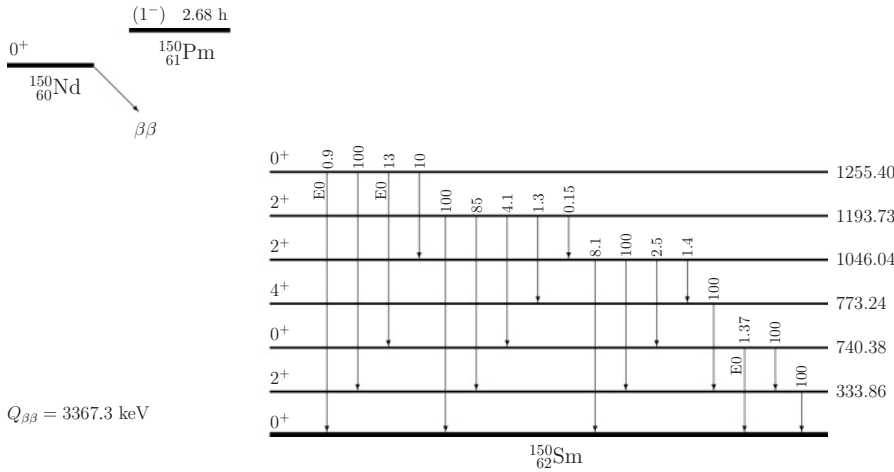


FIG. 1. Decay scheme of ^{150}Nd are taken from [25]. Only the investigated levels of 0^+ and 2^+ and levels associated with transitions to the first ones are shown. Relative branching ratios from each level are presented.

II. EXPERIMENTAL STUDY

The experimental work was performed in the Modane Underground Laboratory (depth of 4800 m w.e.). A 400 cm³ low-background HPGe detector was used to measure a 3046 g sample of Nd₂O₃ powder in a special Marinelli delrin box which was placed on the detector endcap. Taking into account the natural abundance there are 153 g of ^{150}Nd (5.64%) and 154 g of ^{148}Nd (5.76%) in the sample. Data were collected for 11320.5 h.

The Ge spectrometer is composed of a *p*-type crystal. The cryostat, endcap, and the other mechanical parts have been made of a very pure Al-Si alloy. The cryostat has a *J*-type geometry to shield the crystal from radioactive impurities in the dewar. The passive shielding consisted of 2 cm of archeological lead, 10 cm of OFHC copper, and 15 cm of ordinary lead. To remove ^{222}Rn gas, one of the main sources of the background, a special effort was made to minimize the free space near the detector. In addition, the passive shielding was enclosed in an aluminum box flushed with high-purity nitrogen.

The electronics consisted of currently available spectrometric amplifiers and a 8192 channel ADC. The energy calibration was adjusted to cover the energy range from 50 keV to 3.5 MeV, and the energy resolution was 2.0 keV for the

1332-keV line of ^{60}Co . The electronics were stable during the experiment due to the constant conditions in the laboratory (temperature of $\approx 23^\circ\text{C}$, hygrometric degree of $\approx 50\%$). A daily check on the apparatus assured that the counting rate was statistically constant.

The current data of accepted values for different isotopes published in Nuclear Data Sheets were used for analysis of the energy spectrum. The photon detection efficiency for each investigated process has been calculated with the CERN Monte Carlo code GEANT 3.21. Special calibration measurements with radioactive sources and powders containing well-known ^{226}Ra activities confirmed that the accuracy of these efficiencies is about 10%.

The dominate detector backgrounds come from natural ^{40}K , radioactive chains of ^{232}Th and $^{235,238}\text{U}$, manmade and/or cosmogenic activities of ^{137}Cs and ^{60}Co . The sample was found to have a large activity of ^{40}K (46.3 mBq/kg). Additionally long-lived radioactive impurities were observed in the sample, but with much weaker activities, i.e., ^{137}Cs (0.089 mBq/kg), ^{176}Lu (0.450 mBq/kg), ^{138}La (0.068 mBq/kg), ^{133}Ba (0.155 mBq/kg), etc. In our case the most important isotopes contributing to energy ranges of the investigated transitions are ^{214}Bi (1.15 mBq/kg), ^{228}Ac (0.93 mBq/kg), ^{227}Ac (0.62 mBq/kg), and their daughters.

Figures 3, 4, 5, and 6 show the energy spectra in the ranges of interest.

III. ANALYSIS AND RESULTS

A. Search for $\beta\beta$ processes in ^{150}Nd

Double beta decays of ^{150}Nd to 2_1^+ (333.86 keV), 0_1^+ (740.38 keV), 2_2^+ (1046.04 keV), 2_3^+ (1193.73 keV), and 0_2^+ (1255.40 keV) levels in ^{150}Sm have been investigated.

1. Decay to the 0_1^+ excited state

The transition is accompanied by two γ -rays with energies of 333.9 keV and 406.5 keV. The detection photopeak efficiencies are equal to 2.30% at 333.9 keV and 2.29% at 406.5 keV. Figures 3 and 4 show the energy spectrum in the ranges of interest. As one can see there is an excess of

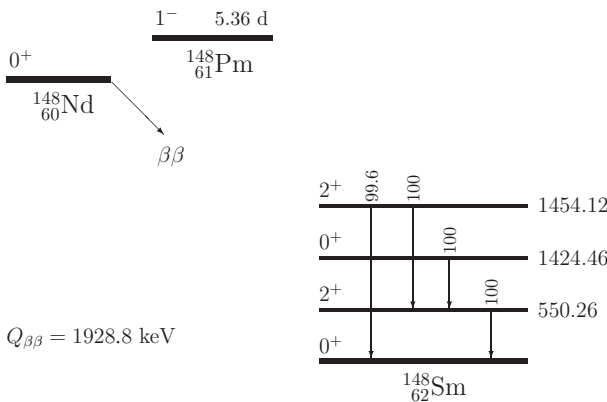


FIG. 2. Decay scheme of ^{148}Nd [26]. Only the investigated levels of 0^+ and 2^+ are shown. The relative branching from each level are also given.

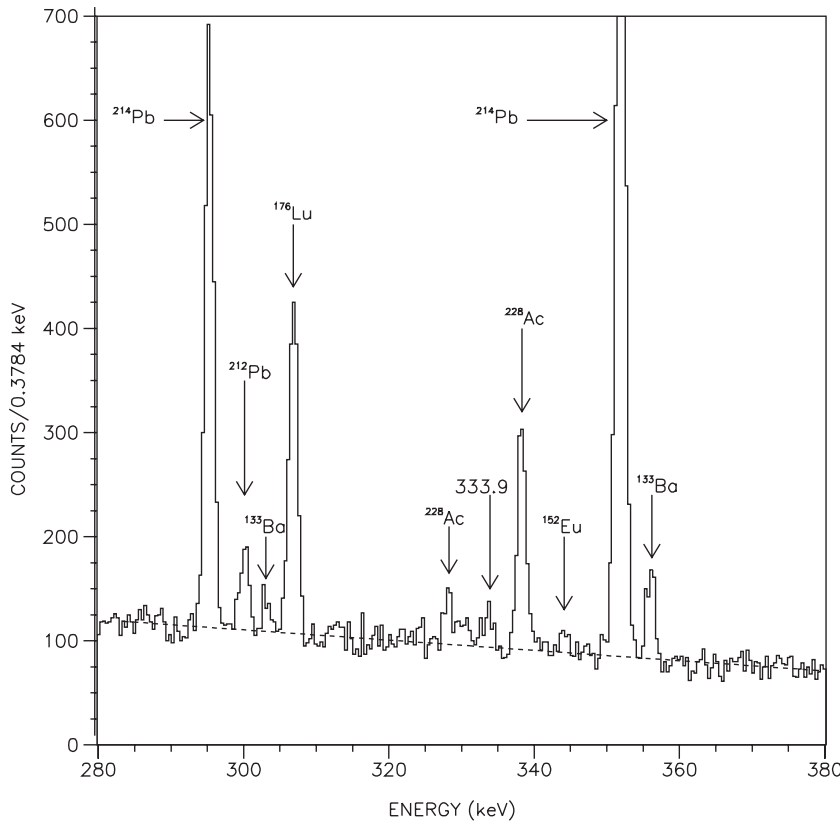


FIG. 3. Energy spectrum with natural Nd_2O_3 powder of observed γ -rays in the range [280–380] keV.

events above the averaged continuous background at the investigated energies. Isotopes of natural radioactivity (^{211}Pb , ^{214}Bi , ^{227}Th , and ^{228}Ac), found in the spectrum, have γ -lines near these energies. ^{214}Bi contributes to both investigated ranges through γ -rays with energies of 333.37 keV (0.065%) and 334.78 keV (0.036%) for the 333.9-keV peak, and 405.72 keV (0.169%) for the 406.5-keV peak. ^{228}Ac touches the 333.9-keV peak range with its γ (332.37 keV, 0.40%). Appropriate γ -rays from ^{227}Th and ^{211}Pb are 334.37 keV (1.54%) and 404.853 keV (3.78%), respectively. Both of the mentioned isotopes are daughters of ^{227}Ac ($T_{1/2} = 21.772$ yr; ^{235}U chain). Activity of ^{227}Ac was estimated using the most intensive γ -lines of its daughters, ^{227}Th (235.96 keV, 12.9%; 256.23 keV, 7.0%), ^{223}Ra (269.46 keV, 13.9%), and ^{219}Rn (271.23 keV, 10.8%; 401.81 keV, 6.6%).

There is also the cosmogenic isotope, ^{150}Eu ($T_{1/2} = 36.9$ yr), which decays to ^{150}Sm , with γ -rays of 333.9 keV

(96%), 406.5 keV (0.14%), 439.4 keV (80%), 584.3 keV (52.6%). The line at 439.4 keV is within one standard deviation of the continuous background, therefore it can be taken into account as a systematic error.

Table I presents the results of the analysis for the two peak energy ranges being studied. A peak's shape is described by a gaussian with a standard deviation of 0.58 keV at 333.9 keV and 0.61 keV at 406.5 keV. For the analysis a peak's range is taken to within approximately four standard deviations ($E \pm 2\sigma$), i.e., 94.82% of full peak at 333.9 keV and 92.75% of full peak at 406.5 keV. As one can see there is an excess of events for each peak. Summing the two peaks we obtain a signal of (177.5 ± 37.6) events, corresponding to a half-life of ^{150}Nd to the first 0^+ excited state of ^{150}Sm given by $T_{1/2} = [1.33^{+0.36}_{-0.23}(\text{stat})^{+0.23}_{-0.13}(\text{syst})] \times 10^{20}$ yr.

The primary systematics come from the GEANT calculations (10%), continuous background estimation (2.6%), and

TABLE I. Analysis of events in the energy ranges of the peaks under study.

| Peak (keV) | 333.9 | 406.5 |
|----------------------------------|-----------------|--|
| Number of events | 779 | 603 |
| Continuous background | 656.4 ± 1.6 | 484.6 ± 1.2 |
| Isotope contributions | | |
| $^{214}\text{Bi}(333.37;334.78)$ | 7.9 ± 2.2 | $^{214}\text{Bi}(405.72)$ 9.3 ± 2.1 |
| $^{227}\text{Th}(334.37)$ | 30.7 ± 4.7 | $^{211}\text{Pb}(404.85)$ 10.2 ± 1.1 |
| $^{228}\text{Ac}(332.37)$ | 5.6 ± 0.7 | |
| Excess of events | 78.5 ± 28.4 | 99.0 ± 24.7 |

TABLE II. Experimental results for $(0\nu + 2\nu)\beta\beta$ decay of ^{150}Nd to the excited states of ^{150}Sm . All limits are given at the 90% C.L.

| Excited state | Energy of γ -rays (efficiency) | $(T_{1/2}^{0\nu+2\nu})_{\text{exp}} (10^{20} \text{ yr})$ | |
|------------------|---------------------------------------|--|---|
| | | This work | Other works |
| $2_1^+(333.86)$ | 333.9(2.60%) | >2.2 | >0.91 [27] $>24^a$ [31] |
| $0_1^+(740.38)$ | 333.9(2.30%) 406.5(2.29%) | $= [1.33^{+0.36}_{-0.23}(\text{stat})^{+0.27}_{-0.13}(\text{syst})]^b$ | >1.5 [28] >1 [27] $>2.4^a$ [31] |
| $2_2^+(1046.04)$ | 712.2(1.78%) | >8.0 | >1.4 [32] |
| $2_3^+(1193.73)$ | 1193.7(0.95%) | >5.4 | >0.027 [33] |
| $0_2^+(1255.40)$ | 921.5(1.45%) | >4.7 | >2 [27] |

^aOnly 0ν decay mode.

^bHalf-life value for 2ν decay (see text for the details).

the possible contribution of ^{150}Eu in the 333.9-keV peak (8.5%).

Previous experiments gave only limits on this transition, $>1 \times 10^{20} \text{ yr}$ [27] and $>1.5 \times 10^{20} \text{ yr}$ [28]. Taking into account all errors, our “positive” result is not in contradiction with the previous limits.

2. Decay to the 2_1^+ excited state

To search for this transition one has to look for a γ -ray with an energy of 333.9 keV. The detection efficiency is 2.60%. The analysis given above shows that the excess of events at 333.9 keV is mainly due to the double beta decay of ^{150}Nd to the 0_1^+ excited state of ^{150}Sm . So one can only give the lower half-life limit on the transition to the 2_1^+ excited state of ^{150}Sm . The limit has been calculated using the likelihood function described in Refs. [29,30] which takes into account all the peaks identified above as background. This result together with available data on $\beta\beta$ decay of ^{150}Nd from other experimental works are presented in Table II.

3. Decays to the 2_2^+ , 2_3^+ , and 0_2^+ excited states

To search for these transitions one has to look for γ -rays with energies of 712.2, 921.5, and 1193.7 keV (Fig. 1). As one can see from Figs. 5 and 6, there are no statistically significant peaks at these energies. Using the same technique as above [29,30] the lower half-life limits are found within $(4.7\text{--}8.0) \times 10^{20} \text{ yr}$ for the transitions (Table II). Table II also presents other valuable data on these transitions.

B. Search for $\beta\beta$ processes in ^{148}Nd

A search for the double beta decays of ^{148}Nd to the 2_1^+ , 0_1^+ , and 2_2^+ excited states of ^{148}Sm was carried out by looking for γ -rays with energies of 550.3, 874.2, and 903.9 keV accompanying these transitions (Fig. 2). Figures 5 and 6 show no statistically significant peaks at these energies. The lower half-life limits reported in Table III have been calculated using the same procedure as in Sec. III A2. Available data

on $\beta\beta$ decay of ^{148}Nd from other experimental works are also presented in Table III.

IV. DISCUSSION

Because the technique used in the present work does not allow for a distinction between $0\nu\beta\beta$ and $2\nu\beta\beta$ decay, our result for double beta decay of ^{150}Nd to the excited 0_1^+ state in ^{150}Sm is the sum of the $0\nu\beta\beta$ and $2\nu\beta\beta$ processes. However we believe that we detected only the $2\nu\beta\beta$ decay. This conclusion is supported by two arguments. First, in the recent NEMO paper [31] the limit on $0\nu\beta\beta$ decay of ^{150}Nd to the excited 0_1^+ state was established as $2.4 \times 10^{20} \text{ yr}$, which is stronger than the half-life value obtained here. Second, the experimental limit for the $0\nu\beta\beta$ decay of ^{150}Nd to the ground 0^+ state of ^{150}Sm is about two orders of magnitude larger [31] than the value reported here. Therefore, considering the reduced phase space factors available for the transition to the excited 0_1^+ state, it is safe to assume that our result for $T_{1/2}$ refers solely to the $2\nu\beta\beta$ decay. As to the possible contribution of $(0\nu + 2\nu)\beta\beta$ decay to the 2_1^+ excited state to the peak at 333.9 keV one can conclude that this contribution is small. First, the experimental limit on neutrinoless mode ($T_{1/2} > 2.4 \times 10^{21} \text{ yr}$ [31]) is much stronger than the obtained “positive” result for the decay of ^{150}Nd to the 0_1^+ excited state of ^{150}Sm . And second, the theoretical prediction for two neutrino

TABLE III. Experimental results for $(0\nu + 2\nu)\beta\beta$ decay of ^{148}Nd to the excited states of ^{148}Sm . All limits are given at the 90% C.L. (See text for details.)

| Excited state | Energy of γ -rays (efficiency) | $(T_{1/2}^{0\nu+2\nu})_{\text{exp}} (10^{20} \text{ yr})$ | |
|------------------|---------------------------------------|---|---------------|
| | | This work | Other works |
| $2_1^+(550.26)$ | 550.3 (2.36%) | >6.6 | >0.03 [33] |
| $0_1^+(1424.46)$ | 550.3 (2.16%) 874.2 (1.83%) | >7.9 | - |
| $2_2^+(1454.12)$ | 550.3 (1.11%) 903.9 (0.87%) | >3.8 | >0.027 [33] |

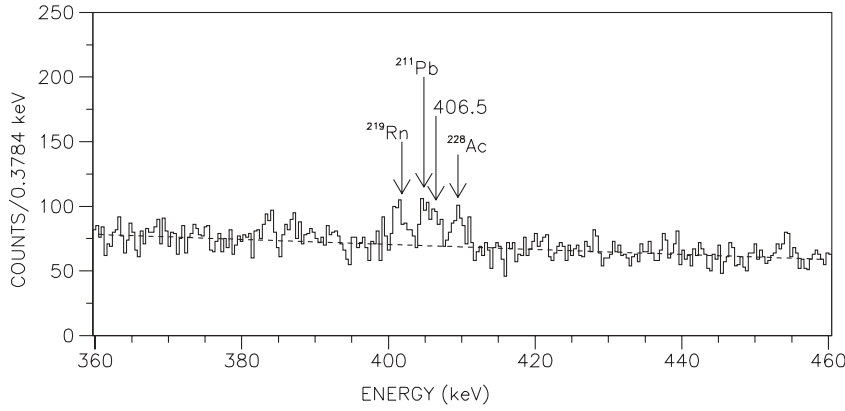


FIG. 4. Energy spectrum with natural Nd_2O_3 powder of observed γ -rays in the range [360–460] keV.

modes ($T_{1/2} \approx 7.2 \times 10^{24} - 1.2 \times 10^{25}$ yr [34]) is very far from the observed value.

Using the phase space factor value $G = 1.2 \times 10^{-17} \text{yr}^{-1}$ (for $g_A = 1.254$) [35] and the measured half-life $T_{1/2} = [1.33^{+0.36}_{-0.23}(\text{stat})^{+0.23}_{-0.13}(\text{syst})] \times 10^{20}$ yr, one obtains a NME value for the $2\nu\beta\beta$ transition to the 0_1^+ excited state of $M_{2\nu}(0_1^+) = 0.025 \pm 0.003$ (scaled by the electron rest mass). One can compare this value with the NME value for the $2\nu\beta\beta$ transition to the ground state of ^{150}Sm , $M_{2\nu}(0_{\text{g.s.}}^+) = 0.033^{+0.001}_{-0.002}$ (here we used the average half-life value $T_{1/2} = (7.8 \pm 0.7) \times 10^{18}$ yr from [11] and $G = 1.2 \times 10^{-16} \text{yr}^{-1}$ (for $g_A = 1.254$) from [35]). One can see that $M_{2\nu}(0_{\text{g.s.}}^+)$ is $\sim 25\%$ greater than $M_{2\nu}(0_1^+)$. Nevertheless, these values are very close and taking

into account errors and possible uncertainties their equality can not be excluded. From the viewpoint of the theory it is important to confirm this difference or to rule it out. Future more precise measurements for both transitions and especially for the transition to the excited state are needed.

For double beta decay of ^{150}Nd to the 2_1^+ , 2_2^+ , and 0_2^+ excited states of ^{150}Sm the limits obtained are ~ 2 – 5 times better than the best previous results [27,32,33]. The limit obtained for the transition to the 2_3^+ excited state is 200 times better than the previous limit [33].

The ^{148}Nd limit for the transition to the 0_1^+ excited state was obtained for the first time and for transitions to 2_1^+ and 2_2^+ states a sensitivity was achieved that is ~ 200 times better than in Ref. [33].

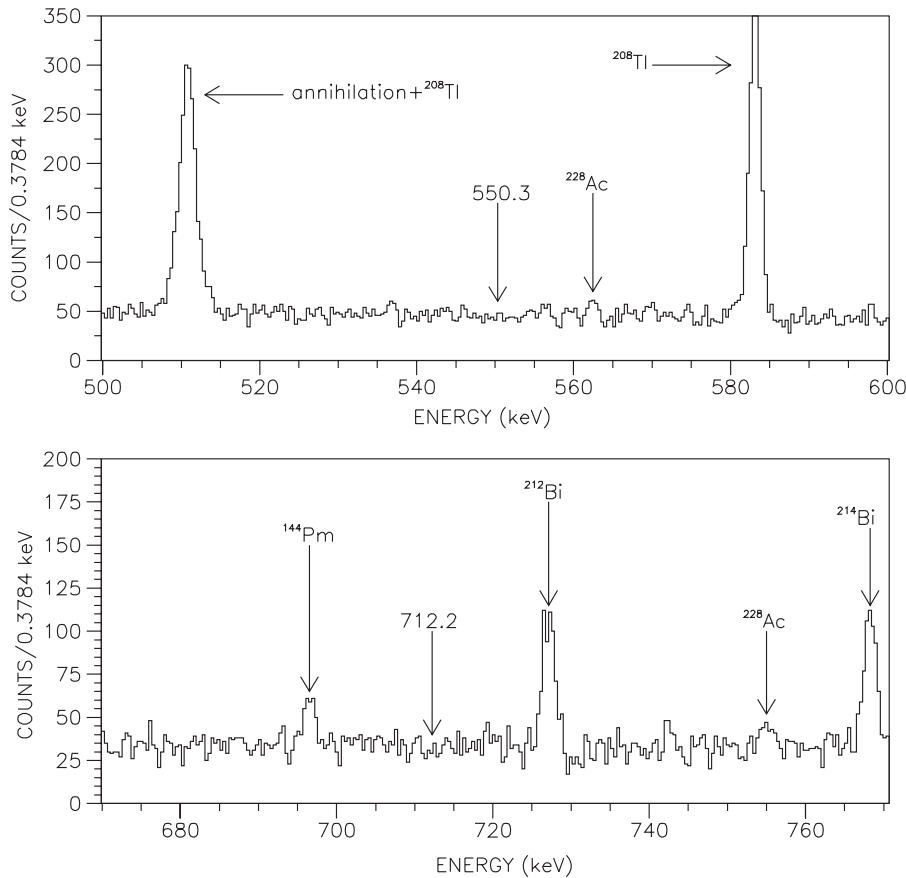


FIG. 5. Energy spectrum with natural Nd_2O_3 powder in the ranges of investigated γ -rays ([500–600] and [670–770] keV).

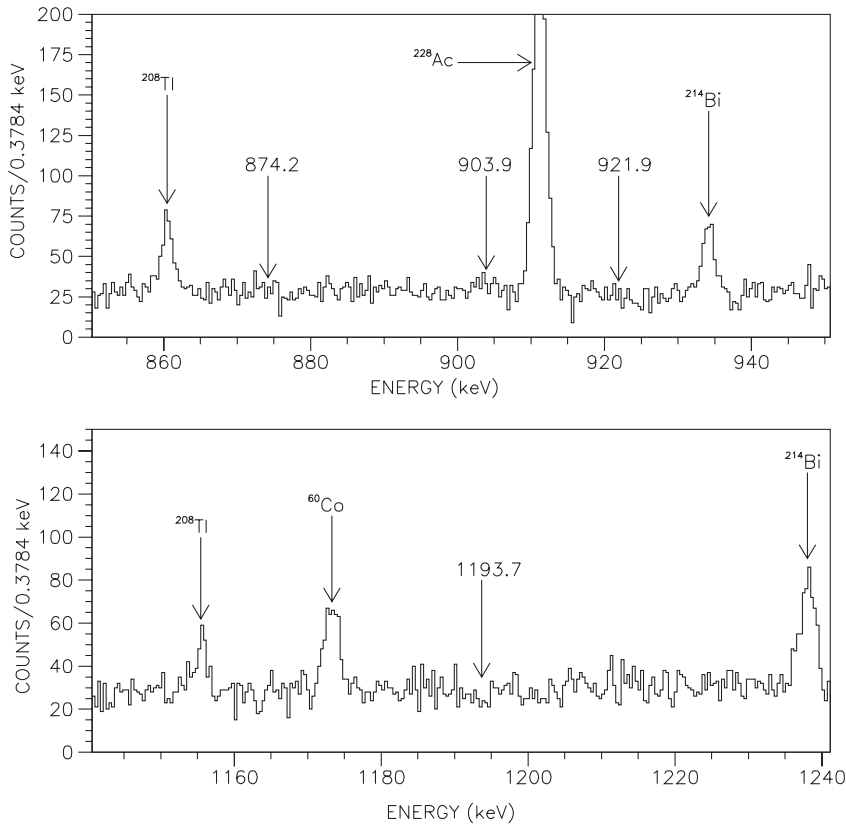


FIG. 6. Energy spectrum with natural Nd_2O_3 powder for γ -rays in the ranges [850–950] and [1140–1240] keV.

V. CONCLUSION

Double beta decay of ^{150}Nd and ^{148}Nd to the excited states of daughter nuclei was investigated with a high level of sensitivity. The half-life for the $2\nu\beta\beta$ decay of ^{150}Nd to the excited 0_1^+ state in ^{150}Sm is measured to be $T_{1/2} = [1.33^{+0.36}_{-0.23}(\text{stat})^{+0.27}_{-0.13}(\text{syst})] \times 10^{20}$ yr. The strongest limits for other transitions were established. The sensitivity of this experiment could still be increased by a few times using a pure Nd_2O_3 (or Nd) sample. Also further increases in the sensitivity could be reached using an enriched Nd sample and

a multicrystal HPGe installation to study larger masses of Nd samples.

ACKNOWLEDGMENTS

The authors would like to thank the Modane Underground Laboratory staff for their technical assistance in running the experiment. We are very thankful to Prof. S. Sutton for his useful remarks. Portions of this work were supported by grants from RFBR (no. 06-02-72553) and CRDF (RUP1-2892-MO-07). This work was also supported by the Russian Federal Agency for Atomic Energy.

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