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Search for Double Electron Capture of ¹⁰⁶Cd*

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Abstract—A search for double electron capture of ¹⁰⁶Cd was performed at the Modane Underground Laboratory (4800 m w.e.) using a low-background and high-sensitivity multidetector spectrometer TGV-2 (Telescope Germanium Vertical). New limits on β^{+}/EC , EC/EC decays of ¹⁰⁶Cd were obtained from preliminary calculations of experimental data accumulated for 4800 h of measurement of 10 g of ¹⁰⁶Cd with enrichment of 75%. They are $T_{1/2}^{2\nu\beta^+EC}>9.1\times10^{18}$ yr, $T_{1/2}^{2\nu EC/EC}>1.9\times10^{19}$ yr for transitions to the first 2^+ , 511.9 keV excited state of $^{106}\rm{Pd}$, and $T_{1/2}^{2\nu\beta^+\rm{EC}}>1.3\times10^{19}$ yr, $T_{1/2}^{2\nu\rm{EC/EC}}>6.2\times10^{19}$ yr for transitions to the ground 0^+ state of ¹⁰⁶Pd. All limits are given at 90% C.L.

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1. INTRODUCTION

Investigation of ultrarare double-beta decay processes (e.g., $\beta^{-}\beta^{-}$, $\beta^{+}\beta^{+}$, β^{+}/EC , EC/EC) is of major importance for particle and nuclear physics as a powerful tool for the study of lepton conservation in general and of neutrino properties in particular [1, 2]. Up to now, more attention has been given to the β ⁻ β ⁻ channels (two-neutrino emitting and neutrinoless modes). There are also other channels of double-beta decay, in particular, the double capture of two bound atomic electrons (EC/EC), capture of the bound electron with emission of a positron (β^+/EC) and decay with emission of two positrons $(\beta^+\beta^+)$. In comparison with the $\beta^{-}\beta^{-}$ decay, these channels are disfavored by smaller available kinetic energy, by Coulomb repulsion on the positron, or due to a small overlap of the bound electron wave function with the nucleus. In principle, the neutrinoless $0\nu\beta^{+}\beta^{+}$, $0\nu\beta^{+}/EC$, and $0\nu EC/EC$ decays can yield the same information as the $0\nu\beta^{-}\beta^{-}$ process.

Prior to now, only the two-neutrino modes of the EC/EC, β^+ /EC, and β^+ β^+ decays were of experimental interest. Owing the low Q values of the $\beta^+\beta^+$ candidates and difficulties connected with experimental detection, the current level of experimental sensitivity to obtain half-life limits is in the range $10^{18}-10^{20}$ yr [3, 4]. On the other hand, the $2\nu EC/EC$ mode ($0^{+}_{\text{g.s}} \rightarrow 0^{+}_{\text{g.s}}$), which is characterized by emission of only two x rays and has a comparatively higher probability, was measured at the level of 10^{17} yr [5, 6].

The double-beta decay is a very rare process and therefore various techniques and methods for background suppression, including elaborate systems of shielding, and of extremely pure construction materials have to be used at the facilities designed to investigate such processes. The measurements are performed in underground laboratories.

On the basis of the experience gained from application of the Ge multidetector spectrometer TGV [7, 8] for studying double-beta decay $(2\nu\beta^{-}\beta^{-}, 0\nu\beta^{-}\beta^{-})$ of ⁴⁸Ca [9], a new low-background multidetector spectrometer TGV-2 has been developed [10]. The spectrometer TGV-2 (like TGV) is intended for the investigation of ultrarare nuclear processes (doublebeta decay, double electron capture, etc.). The main advantage of the TGV-2 is a significantly greater sensitive volume and efficiency of the detectors. Consequently, a higher mass of nuclide to be studied can be used. To prevent an increase in the background

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Fig. 1. Decay scheme of 106Cd (energy in keV).

in the TGV-2 due to a higher mass of construction materials, only those with a very low level of radioactive impurities were used for the detector part. The principal changes were made in the design of the cryostat and detector holders in comparison with the TGV [7, 8]. Two different configurations of the TGV-2 electronics were developed for studying double-beta decay and double electron capture processes.

The most important feature of the TGV-2 is its ability to study a small amount of materials with a high efficiency (especially samples made of isotopes available in small quantities, e.g., ^{48}Ca). The ^{106}Cd isotope is one of those most favorable candidates for studying the EC/EC-decay channel, due to the high decay energy ($Q_{\text{EC/EC}} = 2778 \text{ keV}$). From the point of view of the energy region considered (x rays at about 21 keV), the TGV-2 is the only setup allowing measurement of such an exotic-decay mode as the $2\nu \text{EC}/\text{EC}$ decay from the $0^{+}_{\mathrm{g.s}}$ level of ^{106}Cd to the $0^{+}_{\mathrm{g.s}}$ level of ^{106}Pd (Fig. 1).

The QRPA predictions for the 2ν EC/EC decay half-life of ¹⁰⁶Cd are in the range from 1.0×10^{20} [11] to 8.7 \times 10²⁰ yr [1]. Recently, by considering the SSD hypothesis and dependence of energy denominators on lepton energies, the half-life of the EC/EC decay of 106 Cd was found to be above 4.4×10^{21} yr [12].

2. TGV-2 EXPERIMENT

The TGV-2 experiment was performed at the Modane Underground Laboratory (depth 4800 m w.e.). The search for double electron capture of $106Cd$ was carried out using the low-background multidetector spectrometer TGV-2 (Telescope Germanium Vertical) $[10]$.

The detector part of the TGV-2 (Figs. 2 and 3) is composed of 32 HPGe planar-type detectors 60 mm in diameter and 6.5 mm thick. The detectors are mounted one over another together with double-beta emitters in a low-background cryostat. The sensitive area of each detector is about 2040 mm² (diameter

Fig. 2. Detector part of the TGV-2 cryostat. HPGe planar-type Ge detectors; Al—construction details made from Al–Si alloy; Cu—construction details made from copper; LN—liquid nitrogen; PA—preamplifiers.

51 mm) and the sensitive layer is about 6 mm thick. The total sensitive volume of the TGV-2 detectors is as large as 400 cm^3 . The total mass of the detectors is about 3 kg. The main construction details surrounding the detectors were made from materials with low radioactive contamination $(U + Th < 0.1$ ppb) Al Si 1% alloy, oxygen free copper, and Teflon. The detectors and the preamplifiers were connected by 32 pairs of twisted wires which were arranged to avoid cross talk between the neighboring detectors. Double beta emitters were the foils (thickness \sim 50 µm for Cd) with a diameter of 52 mm, inserted between the entrance windows of the neighboring detectors. The samples for investigations were made by rolling metallic Cd to a thickness of 50 μ m. The thickness of Cd foils was chosen on the basis of Monte Carlo simulations performed with the GEANT 3.21 CERNlib package.

The distance between the detectors and the emitters was fixed to be $<$ 1.5 mm. The energy resolution of the detectors ranged from 3.0 to 4.0 keV at 1332 keV (60 Co). The total efficiency of the TGV-2 spectrometer is 50–70% depending on the energy threshold. ²²⁸Th and ²⁴¹Am radioactive sources were used to calibrate the TGV-2. For this purpose, a Teflon tube allowing sources to move along the stack

Fig. 3. Section view of the stack of HPGe detectors: (*1*) cylindrical holders of detectors, (*2*) investigated samples (double-beta emitters), (*3*) electric contacts (bronze wires in Teflon insulators).

of the detectors was set to run through the passive shielding.

The detector part of the TGV-2 was surrounded by copper shielding $(>20 \text{ cm})$, a steel airtight box against radon, a lead shielding $(>10$ cm), and an antineutron shielding made of polyethylene filled with boron (16 cm). The basic circuit arrangement of the TGV-2 electronics was described in [10]. To suppress both the electronic and the microphone noise in the low-energy region, a method of comparison of two signals generated by processing with two different shaping times (2 and 8 μ s) is used at the TGV-2 [10].

The search for double electron capture of $106Cd$ decay has been carried out since the beginning of 2004. Several runs were performed within the framework of this investigation: (a) measurements of 16 samples $(14.5 g)$ of natural Cd for 903 h (Figs. 4) and 5); (b) three measurement runs with enriched ¹⁰⁶Cd of different origin and enrichment. In the first run, 13 samples of ¹⁰⁶Cd (enrichment ~60%) with a total mass of ∼11.3 g and 3 samples of natural Cd with a total mass of \sim 2.4 g were investigated for 1768 h. In the second run (Fig. 6), 12 samples of $106Cd$ (enrichment 67.9%) with a total mass of ∼10 g and 4 samples of natural Cd with a total mass of ∼3.2 g were studied for 3277 h. The third run was performed with 12 samples of $106Cd$ (enrichment 75.0%) with a total mass of \sim 10 g and 4 samples of natural Cd previously used in the second run.

The experimental run with 16 samples of natural Cd was performed mainly for the measurement of the TGV-2 background conditions. The parameters of the background in the regions of interest $(K_{\alpha}Pd, K_{\beta}Pd, K_{\alpha}Pd + K_{\beta}Pd, K_{\alpha}Cd, 511 \text{ keV}, \text{etc.})$

Fig. 4. Energy spectra of single events (*a*), double coincidence events (*b*), and double coincidences with selection of the events from the energy region corresponding to the $K_\alpha(Pd)$ rays in one of the neighboring detectors (*c*) obtained in the measurement of natural $Cd(14.5 g)$ for 903 h.

Fig. 5. Suppression of electronic noise in the run with natural Cd for 903 h. Upper spectrum is all events; lower spectrum is selected events.

were obtained from spectra of single events, double coincidence events from neighboring detectors, and double events with selection of the corresponding energy regions and were used in further calculations

of experimental runs with ¹⁰⁶Cd. The background was analyzed both for separate detectors (or pairs of detectors) and for the total energy spectra of all detectors (and pairs) (Fig. 4). The TGV-2 sensitivity was

Fig. 6. The spectra of single- and double coincidence events between the neighboring detectors obtained in the second run for samples of natural Cd (3.2 g) and ¹⁰⁶Cd (10.0 g) with enrichment of 67.9% for $T = 794$ h.

found to be $T_{1/2}^{\rm EC/EC}$ $> 3\times 10^{19}$ yr (90% C.L.) for the search for double electron capture of $^{106}\mathrm{Cd}$ ($0^{+}_{\mathrm{g.s}}$ \rightarrow $0^{+}_{\text{g.s}}$ transition) in the experiment with natural Cd.

The method of additional suppression of electronic noise in the low-energy region was also tested. The efficiency of this method may be demonstrated by a comparison of the low-energy part $(<50 \text{ keV})$ of two spectra (Fig. 5) obtained in the run with natural Cd. The upper spectrum contains all events of the run; the lower one contains a part of these events selected by correlations between "true" pulses with a different shaping time (2 and 8 μ s) in each detector (see [10] for details).

The first two attempts to search for double electron capture of 106 Cd in the direct measurements with enriched ¹⁰⁶Cd failed. In both cases, the foils of enriched $106Cd$ were found to be contaminated with $113mCd$. As a result, additional β and KX radiation (Fig. 6) in the experimental spectra significantly decreased the sensitivity of the spectrometer. The presence of $113mCd$ in the samples of $106Cd$ was not indicated in

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their certificates. Our attempts to clarify the origin of this radioactive contamination were not successful. It should only be mentioned that such type of contamination is very difficult to find in relatively thick samples because only β radiation with a maximal energy of ∼580 keV is available in the ¹¹³mCd decay. The thickness of enriched $106Cd$ was decreased by many times (down to 50 μ m) for the TGV-2 experiment. In this case, both β radiation and Kx rays of Cd arising due to the interaction of β radiation with Cd foils leave the thin samples and are detected by the TGV-2 with a high efficiency. Four foils of natural Cd were used to test background in the reported measurements with enriched ¹⁰⁶Cd. The spectra obtained from the detectors with natural Cd had a usual background without additional contaminations (Fig. 6).

Enriched ¹⁰⁶Cd used in the third run of the search for double electron capture of ¹⁰⁶Cd was without additional contaminations. The measurement of 12 samples (10 g) of enriched 10^{6} Cd and 4 samples (3.2 g) of natural Cd is in progress now. The energy spectra obtained in this experimental run

Fig. 7. Spectra of single events (top), double coincidence events (middle), and double coincidences with selection of the events from the energy region corresponding to the $K_{\alpha}(\text{Pd})$ series in one of the neighboring detectors (bottom) obtained in the third run with 10 g of 10^{6} Cd (enrichment 75%) for 4800 h.

Fig. 8. Low-energy region of the spectra of double coincidence events with selection of $K_{\alpha}(\text{Pd})$ rays (lower spectra in Fig. 7) obtained in the third run with 10 g of 10^{6} Cd (enrichment 75%) during the 4800-h exposure. At the top of the figure, the calculated shape of the possible Kx Pd and Kx Cd radiations.

after 4800 hours of exposure are presented in Fig. 7. The γ peaks in the spectra are due to isotopes of U and Th rows. This is a total effect coming from the external background, the intinsic background of the TGV-2 caused by contamination of the detectors and construction details, and contamination of the investigated foils. The internal radioactive contamination in the present measurement will cause Kx rays of Cd which increase background in the region of interest (K_{α} and K_{β} series of Pd). It is impossible to separate K_{α} Cd (22.98 and 23.17 keV) from K_{β} Pd (23.81 and 24.30 keV). In our preliminary off-line calculation of double coincidences the energy window was set at 19–22 keV to reduce background from the Kx rays of Cd (lower spectra in Fig. 7). This means that we took into account only the K_{α} series of Pd (21.02 and 21.18 keV), which was 68.4% of all Kx rays of Pd.

3. PRELIMINARY RESULTS

The analysis of the low-energy part of the double coincidence spectrum of ¹⁰⁶Cd with the "window" of $19-22$ keV (Fig. 8) shows a small increase in the number measured events in the regions of \sim 21 and ∼23 keV. The additional events with energy of \sim 23 keV may be explained by deexcitation of Kx rays of Cd induced by radiation of the contamination in the detectors, details of the cryostat, and investigated foils. The additional events above the background in the region of ∼21 keV (if they really occur) may point to the presence of the process of double electron capture of ¹⁰⁶Cd. Larger statistics and a highly accurate long-term background measurement are needed for the careful analysis of these events.

A preliminary calculation of the experimental data accumulated for 4800 h with 10 g of 10^{6} Cd (enrichment 75%) was performed. The calculation was based on the analysis of the $Kx-Kx$, $Kx-511$ keV, and 511 keV–511 keV coincidence spectra. The new limits on the half-lives different branches (β^+/EC , EC/EC) of ¹⁰⁶Cd decay were obtained. They are $T_{1/2}^{2\nu\beta^+EC} > 9.1 \times 10^{18}$ yr (90% C.L.), $T_{1/2}^{2\nu EC/EC} >$ 1.9×10^{19} yr (90% C.L.) for transitions to the first $2^+,511.9$ -keV excited state of $^{106}\text{Pd,}$ and $T_{1/2}^{2\nu\beta^+\text{EC}}$ $>$ 1.3×10^{19} yr (90% C.L.), $T_{1/2}^{2\nu EC/EC} > 6.2 \times 10^{19}$ yr (90% C.L.) for transitions to the ground 0^+ state of $106Pd$. Our value for the $2\nu EC/EC$ decay mode of ¹⁰⁶Cd ($0^{+}_{\text{g.s}} \rightarrow 0^{+}_{\text{g.s}}$) is more than one order of magnitude higher than those obtained in recent experiments [13, 14].

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REFERENCES

- 1. W. C. Haxton and G. J. Stephenson, Jr., Prog. Part. Nucl. Phys. **12**, 409 (1984).
- 2. V. I. Tretyak and Yu. G. Zdesenko, At. Data Nucl. Data Tables **80**, 83 (2002).
- 3. A. Barabash et al., Nucl. Phys. A **604**, 115 (1996).
- 4. P. Belli et al., Astropart. Phys. **10**, 115 (1999).
- 5. E. B. Norman and M. A. DeFaccio, Phys. Lett. B **148**, 31 (1984).
- 6. A. Sh. Georgadze et al., Phys. At. Nucl. **58**, 1093 (1995).
- 7. Ch. Briancon et al., Nucl. Instrum. Methods Phys. Res. A **372**, 222 (1996).
- 8. V. B. Brudanin et al., Izv. Ross. Akad. Nauk, Ser. Fiz. **60** (1), 137 (1996).
- 9. V. B. Brudanin et al., Phys. Lett. B **495**, 63 (2000).
- 10. V. B. Brudanin et al., Izv. Ross. Akad. Nauk, Ser. Fiz. **67**, 618 (2003) [Bull. Russ. Acad. Sci., Phys. **67**, 680 (2003)].
- 11. M. Aunola and J. Suhonen, Nucl. Phys. A **602**, 133 (1996).
- 12. P. Domin, S. Kovalenko, F. Šimkovic, and S. V. Semenov, Nucl. Phys. A **753**, 337 (2005).
- 13. F. A. Danevich et al., Phys. Rev. C **68**, 035501 (2003).
- 14. H. Kiel et al., Nucl. Phys. A **723**, 499 (2003).