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Search for 2β decays of ^{96}Ru and ^{104}Ru by ultralow-background HPGe γ spectrometry at LNGS: Final results

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An experiment to search for double-β decay processes in ^{96}Ru and ^{104}Ru, which are accompanied by γ rays, has been realized in the underground Gran Sasso National Laboratories of the I.N.F.N. (Italy). Ruthenium samples with masses of ≈0.5–0.7 kg were measured with the help of ultralow-background high-purity Ge γ-ray spectrometry. After 2162 h of data taking the samples were deeply purified to reduce the internal contamination of ^{40}K. The last part of the data has been accumulated over 5479 h. New improved half-life limits on $2\beta^+/\varepsilon\beta^+/2\varepsilon$ processes in ^{96}Ru have been established on the level of 10^{20} yr, in particular for decays to the ground state of ^{96}Mo: $T_{1/2}^{2\nu2\beta^+} \geqslant 1.4 \times 10^{20}$ yr, $T_{1/2}^{2\nu\varepsilon\beta^+} \geqslant 8.0 \times 10^{19}$ yr, and $T_{1/2}^{0\nu2K} \geqslant 1.0 \times 10^{21}$ yr (all limits are at 90% C.L.). The resonant neutrinoless double-electron captures to the 2700.2 and 2712.7 keV excited states of ^{96}Mo are restricted as $T_{1/2}^{0\nu KL} \geqslant 2.0 \times 10^{20}$ yr and $T_{1/2}^{0\nu2L} \geqslant 3.6 \times 10^{20}$ yr, respectively. Various two-neutrino and neutrinoless 2β half-lives of ^{96}Ru have been estimated in the framework of the quasiparticle random-phase approximation approach. In addition, the $T_{1/2}$ limit for $0\nu2\beta^-$ transitions of ^{104}Ru to the first excited state of ^{104}Pd has been set as $\geqslant 6.5 \times 10^{20}$ yr.

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I. INTRODUCTION

Double-beta (2β) decay is a process of transformation of a nucleus (A, Z) either to $(A, Z+2)$ with simultaneous emission of two electrons ($2\beta^-$ decay) or to $(A, Z-2)$ through one of the following ways: emission of two positrons ($2\beta^+$), capture of electron and emission of positron ($\varepsilon\beta^+$), or double-electron capture (2ε). The two-neutrino (2ν) double-β decay, in which two (anti)neutrinos are also emitted, is allowed in the standard model (SM); however, being a second-order process in the weak interactions, it is characterized by very long half-lives in the range of 10^{18}–10^{24} yr [1]. There are 35 known nuclei candidates for $2\beta^-$ and 34 candidates for $2\beta^+/\varepsilon\beta^+/2\varepsilon$ decays [2]. To date, two-neutrino 2β decays are observed for several $2\beta^-$ decaying nuclei (see reviews [2,3] and recent original works [4,5]), while indications of double-electron capture have been obtained for ^{130}Ba in geochemical experiments [6,7].

The neutrinoless (0ν) mode of the 2β decay is forbidden in the SM because it violates the lepton number by two units. It is, however, naturally expected in many SM extensions which describe the neutrino as a Majorana particle with nonzero mass. The neutrino oscillation experiments indicate that neutrinos are massive. Nevertheless, since they are sensitive to the difference in ν masses, the absolute ν mass scale is unknown [8]. The $0\nu2\beta$ decay is considered a powerful tool for checking lepton number conservation, determining the absolute ν masses and their hierarchy, establishing the nature of the neutrino (Majorana or Dirac particle), and finding a possible contribution of right-handed admixtures to weak interaction and the existence of Nambu-Goldstone bosons (Majorons). A particular analysis of data on ^{76}Ge provided evidence for $0\nu2\beta$ decay [9]; several experiments with the aim to test it and to explore the inverted hierarchy of the Majorana neutrino mass region ($m_\nu \sim 0.1$–0.05 eV) are now in progress or under development [1]. Studies of neutrinoless $2\beta^-$ and $2\beta^+/\varepsilon\beta^+/2\varepsilon$ decays are mutually complementary, helping to distinguish contributions from the neutrino mass and right-handed admixture mechanisms [10].

^{96}Ru is one of the only six isotopes where decay with emission of two positrons is allowed [2] thanks to the high-energy release: $Q_{2\beta} = (2714.51 \pm 0.13)$ keV [11]. It also has a quite large natural abundance: $\delta = 5.54\%$ [12]. Moreover, in the case of the capture of two electrons from the K and L shells (the binding energies are $E_K = 20.0$ and $E_{L1} = 2.9$ keV [13]) or both from the L shell, the decay energies (2691.61 ± 0.13) and (2708.71 ± 0.13) keV are close to the energy of the excited levels of ^{96}Mo ($E_{\rm exc} = 2700.21$ and 2712.68 keV [14]). Such a situation could give rise to a resonant enhancement of

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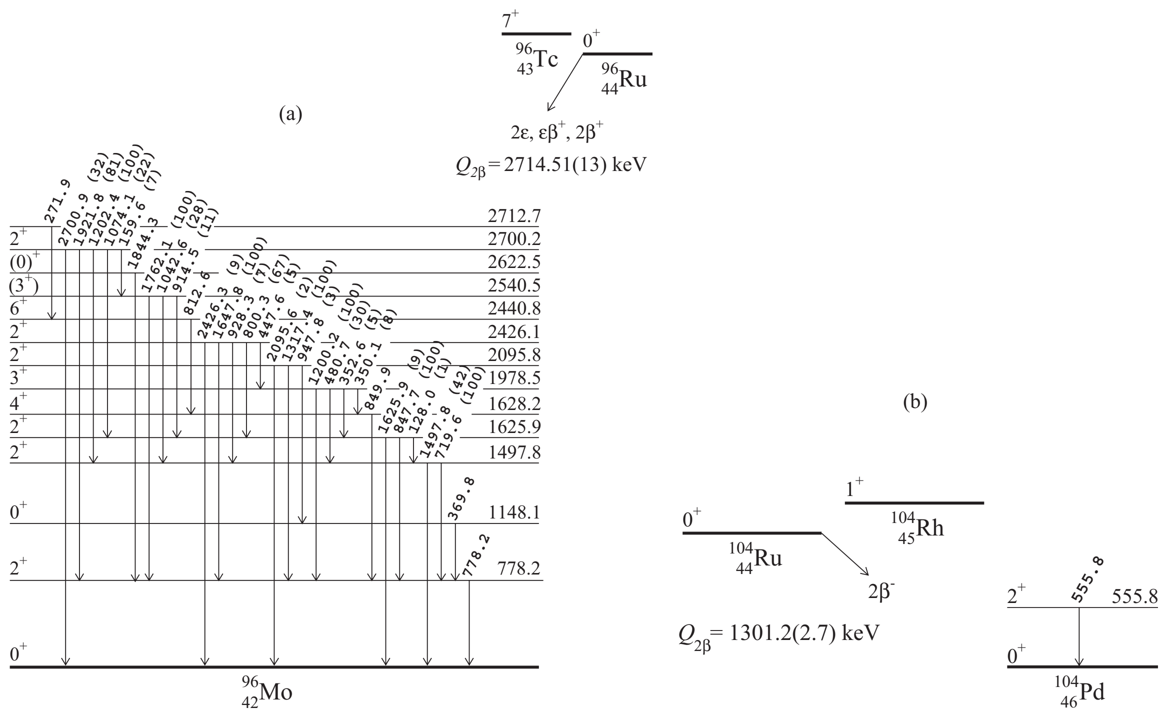

FIG. 1. Decay schemes of ^{96}Ru (a) and ^{104}Ru (b). Energies of the excited levels and emitted γ quanta are in keV. The relative intensities of γ quanta are given in parentheses [13,14,18].

the neutrinoless KL and $2L$ capture to the corresponding level of the daughter nucleus as a result of the energy degeneracy [15].[1]

In addition, another isotope of ruthenium, ^{104}Ru, is potentially unstable with respect to the $2\beta^-$ decay [$Q_{2\beta} = (1301.2 \pm 2.7)$ keV [17], $\delta = 18.62\%$]. The decay schemes of ^{96}Ru and ^{104}Ru are shown in Fig. 1.

Despite the high energy release and the high abundance, only one search for $2\beta^+/\varepsilon\beta^+$ processes in ^{96}Ru was performed in 1985, giving $T_{1/2}$ limits on the level of 10^{16} yr [19]. The efforts were renewed only in 2009, when a Ru sample with a mass of 473 g was measured for 158 h with an HPGe detector (468 cm^3) in the underground conditions of the Gran Sasso National Laboratories (Laboratori Nazionali del Gran Sasso, LNGS) of the I.N.F.N. [3600 meters (water equivalent, w.e.)] [20] (an updated statistics of 2162 h was then reported in [21]). The achieved sensitivity for the $2\beta^+/\varepsilon\beta^+/2\varepsilon$ decays was 10^{18}–10^{19} yr; for several modes of 2β decay of ^{96}Ru (and ^{104}Ru) $T_{1/2}$ limits were established for the first time. A search for 2β decays of Ru was also performed in the HADES underground laboratory [500 m (w.e.)] where a sample of Ru with mass of 149 g was measured during 2592 h; $T_{1/2}$ limits were obtained on the level of 10^{19} yr [22].

Our previous measurements [20,21] showed that the used Ru sample was contaminated by ^{40}K at $\simeq 3$ Bq/kg, and better results are possible only with purified Ru. Here we report the final results of the search for $2\beta^+/\varepsilon\beta^+/2\varepsilon$ processes in ^{96}Ru and for $2\beta^-$ decay in ^{104}Ru obtained with a purified sample of Ru (720 g) in measurements during 5479 h.

II. PURIFICATION OF RUTHENIUM AND LOW-BACKGROUND MEASUREMENTS

The ruthenium (with natural isotopic composition) of 99.99% grade produced by powder metallurgy was provided by Heraeus [23]. At the first stage [20,21], the Ru sample with total mass of 473 g was in the form of pellets (50 tablets $\oslash 16 \times 5$ mm, density ≈ 8.7 g/cm^3). The analysis of the data showed a high level of ^{40}K contamination in the ruthenium (3.4 Bq/kg), and for further measurements the Ru sample with an increased total mass of 946 g was purified by an electron beam melting method.

The ruthenium was divided into five parts, each with a mass of almost 0.2 kg, which were slowly melted (to avoid intensive sprinkling) using an electron beam and kept in a liquid state under vacuum (0.01–0.05 Pa) for $\simeq$1.5–2 h. The purification occurs through the evaporation of the impurities from the melted ruthenium. More details about the purification process can be found in [24]. As a result, five Ru samples in the form of oval disks (total 719.5 g, density ≈ 10.0 g/cm^3) were obtained.

[1]In accordance with older atomic masses [16], the energy release $Q_{2\beta} = (2718 \pm 8)$ keV gave the decay energies for KL and $2L$ captures as (2695 ± 8) and (2712 ± 8) keV, respectively, compatible within uncertainties to the energies of the ^{96}Mo excited levels (2700.2 and 2712.7 keV), and ^{96}Ru was considered as a very promising candidate in looking for resonant $0\nu2\varepsilon$ captures. After the recent high-precision measurement of Ref. [11], ^{96}Ru is no longer considered a promising candidate in the search for this process.

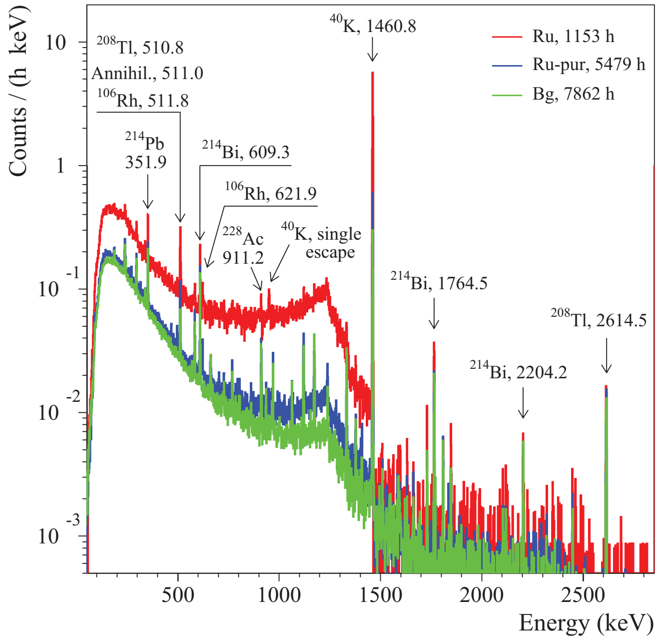

FIG. 2. (Color online) The energy spectra above 20 keV accumulated with the initial Ru sample over 1153 h (Ru) and with the purified Ru over 5479 h (Ru-pur) in comparison with the background (Bg) of the GeMulti ultralow-background HPGe γ spectrometer measured over 7862 h. The energies of γ lines are in keV.

The purified ruthenium samples were measured over 5479 h in the GeMulti setup (made of four HPGe detectors; $\simeq$225 cm^3 each one) installed deep underground at the LNGS. The detectors are surrounded by a passive shield made of low-radioactivity copper ($\simeq$5 cm thick) and low-radioactivity lead ($\simeq$25 cm). The setup was continuously flushed with high-purity nitrogen to remove radon. The typical energy resolution of the detectors is 2.0 keV at the 1332.5 keV line of ^{60}Co. The energy spectra without samples were accumulated with the GeMulti spectrometer over 7862 h. The results of the measurements are presented in Fig. 2, where the effect of the purification is clearly visible (here and in the following, the spectra of the GeMulti setup are the sum of the spectra of the four individual HPGe detectors). Table I gives a summary of the measured radioactive contaminations in the used Ru before and after the purification process. The purification allowed us to decrease the ^{40}K contamination by $\simeq$20 times; the contamination by ^{226}Ra was also suppressed by $\simeq$6 times, while the activity of ^{106}Ru was decreased by $\simeq$5 times due to decay with the half-life $T_{1/2} = 371.8$ d [25].

III. NEW EXPERIMENTAL $T_{1/2}$ LIMITS ON 2β DECAY OF RUTHENIUM

We did not observe any peak in the spectra accumulated with the ruthenium sample which could be unambiguously attributed to the 2β processes in ^{96}Ru and ^{104}Ru. Therefore only lower half-life limits are given using the formula

$$\lim T_{1/2} = N\eta t \ln 2/\lim S, \qquad (1)$$

TABLE I. Radioactive contamination of the Ru sample used in [20] (473 g, 158 h) and of the purified Ru sample (720 g, 5479 h, measured here). For comparison, the results of the sample used in [22] (149 g, 2592 h) are also presented. The limits are given at 90% C.L. (95% C.L. for [22]). Activity of ^{103}Ru ($T_{1/2} = 39.26$ d [13]) is quoted for the beginning of the present measurements.

Chain	Nuclide	Activity (mBq/kg)		
		Ru [20]	Purified Ru	Ru [22]
^{228}Th	^{228}Ra	$\leqslant$7.1	$\leqslant$1.0	8.7 ± 0.7
	^{228}Th	$\leqslant$3.4	1.4 ± 0.4	8.8 ± 0.6
^{235}U	^{235}U	$\leqslant$6.9	$\leqslant$4.0	–
^{238}U	^{234}Th	$\leqslant$390	–	$\leqslant$36
	^{234}Pa^m	$\leqslant$260	$\leqslant$23	–
	^{226}Ra	6.4 ± 1.7	1.0 ± 0.3	14.6 ± 0.7
	^{210}Pb	–	–	$\leqslant$100
	^{40}K	3400 ± 600	153 ± 4	169 ± 7
	^{60}Co	$\leqslant$1.7	$\leqslant$0.1	$\leqslant$0.2
	^{137}Cs	$\leqslant$2.6	$\leqslant$0.1	$\leqslant$0.2
	^{103}Ru	–	3.3 ± 0.7	–
	^{106}Ru	24 ± 7	5.0 ± 0.6	$\leqslant$1.7

where N is the number of potentially 2β unstable nuclei in the Ru sample, η is the detection efficiency, t is the measuring time, and lim S is the number of events of the effect searched for which can be excluded at a given confidence level (C.L.; all the limits in the present study are given at 90% C.L.). The efficiency of the detectors for the double-β processes in ^{96}Ru and ^{104}Ru has been calculated by using the EGS4 code [26] with initial kinematics given by the DECAY0 event generator [27]. The procedure of the analysis, in particular in determining the lim S values, is well described in [20].

A. Search for $2\beta^+$ decay of ^{96}Ru

Only the ground state of ^{96}Mo can be populated in the $2\beta^+$ decay of ^{96}Ru, and thus only annihilation γ quanta with energy 511.0 keV could be registered by our detectors. A possible extra rate in the annihilation peak in the spectrum accumulated with the purified Ru sample (see Fig. 3) could be related to the $2\beta^+$ (and $\varepsilon\beta^+$) decay of ^{96}Ru.

The area of the annihilation peak in the measurements with the purified Ru during 5479 h is equal to (1461 ± 39) counts, while in the background spectrum it is (535 ± 27) counts during 3362 h;[2] this gives (589 ± 58) counts of the extra events. The excess is explained by the following contributions:

(i) The 511.8 keV γ line from ^{106}Rh which is the daughter radionuclide of the cosmogenic ^{106}Ru; this contribution is estimated to be (433 ± 52) counts using the supplementary ^{106}Rh peak at 621.9 keV with an area (197 ± 24) counts, taking into account the different yields of these γ quanta

[2]We use here the last series of the background measurements with the GeMulti setup close to the data accumulated with the purified Ru sample.

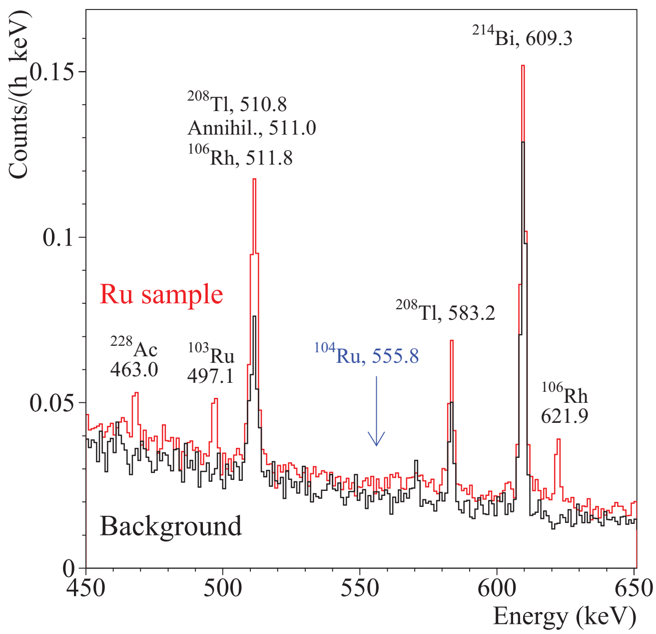

FIG. 3. (Color online) Fragment of the energy spectra accumulated with the ruthenium sample over 5479 h (Ru sample) and without the sample over 3362 h (Background; see footnote[2]) in the vicinity of the annihilation peak.

per decay ($\gamma_{512} = 20.4\%$ and $\gamma_{622} = 9.93\%$ [13]) and their detection efficiencies ($\eta_{512} = 3.0\%$ and $\eta_{622} = 2.8\%$).

(ii) The 510.8 keV γ line from the ^{208}Tl contamination in the Ru sample; this contribution is estimated in a similar way as before to be (49 ± 11) counts using the area of the 583.2 keV peak of ^{208}Tl (with subtraction of the contribution from the corresponding background peak).

(iii) The e^+e^- pairs created by the 1460.8 keV γ quanta emitted in the ^{40}K decay; this contribution is estimated to be (165 ± 4) counts using the area of the 1460.8 keV peak, (5266 ± 132) counts (after subtraction of the corresponding background peak), and results of simulation with the EGS4, which give a ratio of 1:31.9 between the 511.0 and 1460.8 keV peaks from ^{40}K in our measurements.

The difference between the measured number of events in the 511.0 keV peak and the estimated contributions from the known sources, (-58 ± 79) counts, could eventually be ascribed to the effect searched for. Obviously, there is no evidence of $2\beta^+$ (and $\varepsilon\beta^+$) decay of ^{96}Ru to the ground state of ^{96}Mo. In accordance with the Feldman-Cousins procedure [28], it results in a limit $\lim S = 79$ counts for the effect, which can be excluded at 90% C.L. Taking into account the calculated efficiency of $2\beta^+$ processes (10.36% for 2ν mode and 10.31% for 0ν) and the number of ^{96}Ru nuclei ($N = 2.38 \times 10^{23}$), this gives

$$
\begin{aligned}
T_{1/2}^{2\nu2\beta^+}(\text{g.s.} \to \text{g.s.}) &\geqslant 1.4 \times 10^{20}\ \text{yr},\\
T_{1/2}^{0\nu2\beta^+}(\text{g.s.} \to \text{g.s.}) &\geqslant 1.3 \times 10^{20}\ \text{yr}.
\end{aligned} \tag{2}
$$

If the observed number of events is less than the expected background and thus the estimated effect is negative, Feldman and Cousins [28] recommended giving, in addition to the upper limit, the so-called sensitivity of the experiment defined as "the average upper limit that would be obtained by an ensemble of experiments with the expected background and no true signal." Using the total number of events in the range of 509–514 keV as the background ($B = 2522$ counts) and extrapolating Table XII of [28] (in terms of $\sqrt{B}$), we obtain $\lim S_s = 93$ counts at 90% C.L. This gives the "sensitivity" $T_{1/2s}$ value, e.g., for the $0\nu2\beta^+$ decay as $T_{1/2s}^{0\nu2\beta^+}$(g.s. → g.s.) $\geqslant 1.1 \times 10^{20}$ yr, which is very close to the obtained above value of 1.3×10^{20} yr. We accept the values (2) as the final ones (also to compare with results from other experiments where only the upper limits are given).

In addition to the analysis of the usual one-dimensional spectrum, the GeMulti setup with its four HPGe detectors offers the possibility of using coincidences between different detectors for γ quanta emitted simultaneously (annihilation γ quanta in $2\beta^+$ and $\varepsilon\beta^+$ decays, and γ's from cascades in the deexcitation of the excited ^{96}Mo levels). The setup, with and without a Ru sample, has been operated in coincidence mode over 5479 and 2490 h, respectively. The procedure of the analysis is the same as described recently in [29], where a two-dimensional spectrum was used to detect the $2\nu2\beta^-$ transition of ^{100}Mo to the first excited 0_1^+ state of ^{100}Ru. So, fixing the energy of one of the detectors to the expected 511 ± 3 keV (in accordance with the energy resolution for the annihilation peak), we observe the coincidence peak at the corresponding energy 511 ± 3 keV. There are 18(4) counts in both the spectra. The Monte Carlo simulations give the efficiencies to get coincidences of two γ quanta with energy 511.0 keV: 0.30% for $0\nu2\beta^+$ decay of ^{96}Ru and $8 \times 10^{-5}\%$ for ^{40}K (which gives the biggest contribution of 2 counts during 5479 h). The difference between the observed and the expected number of counts (-24 ± 10) corresponds to $\lim S = 3.3$ counts at 90% C.L., which results in $T_{1/2}^{0\nu2\beta^+}$(g.s. → g.s.) $\geqslant 9.3 \times 10^{19}$ yr. This value is comparable to but slightly lower than that obtained above from the analysis of the one-dimensional spectrum. This also concerns other limits obtained from the analysis of coincidences: while they are comparable to those derived from the one-dimensional spectrum, in general they are lower due to a lower detection efficiency.

B. $\varepsilon\beta^+$ processes in ^{96}Ru

The limit obtained above for the $2\beta^+$ decay considering the 511.0 keV peak ($\lim S = 79$ counts at 90% C.L.) can be used to estimate a half-life limit on the $\varepsilon\beta^+$ decay to the ground state of ^{96}Mo. Taking into account the efficiencies for the $\varepsilon\beta^+$ decay of ^{96}Ru (6.12% for 2ν mode and 5.89% for 0ν), we obtain

$$
\begin{aligned}
T_{1/2}^{2\nu\varepsilon\beta^+}(\text{g.s.} \to \text{g.s.}) &\geqslant 8.0 \times 10^{19}\ \text{yr},\\
T_{1/2}^{0\nu\varepsilon\beta^+}(\text{g.s.} \to \text{g.s.}) &\geqslant 7.7 \times 10^{19}\ \text{yr}.
\end{aligned}
$$

In addition to the transition to the ground state, a few excited levels of ^{96}Mo can be populated in $\varepsilon\beta^+$ decay of ^{96}Ru (up to the level 2^+, 1625.9 keV). To estimate the number of events ($\lim S$), the experimental energy spectrum was fitted in different energy intervals with the sum of components representing the background (internal ^{40}K, U/Th, external γ

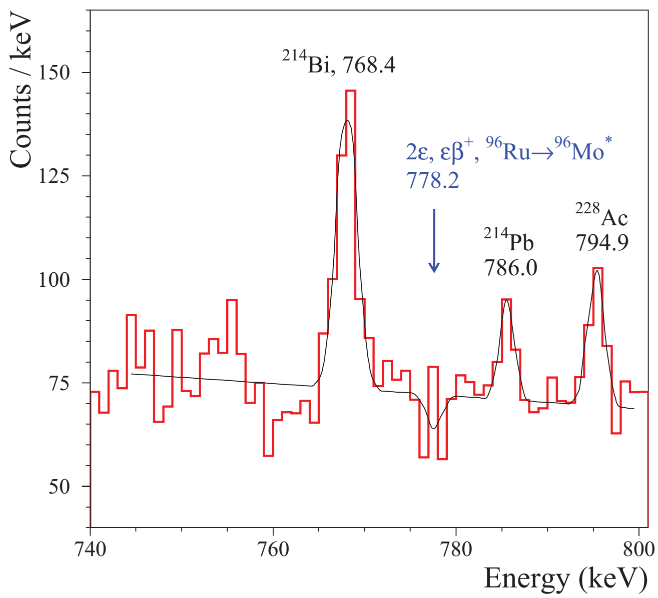

FIG. 4. (Color online) Fragment of the energy spectrum accumulated with the purified Ru sample over 5479 h with the ultralow-background HPGe γ spectrometer. The fit is shown by a solid line. The arrow shows the energy of the peak expected in the decay of ^{96}Ru through the 2^+, 778.2 keV level of ^{96}Mo. The energies of the γ lines are in keV.

from the details of the setup) and the EGS4-simulated models for 2β processes in ^{96}Ru. The used fitting approach is described in detail in Ref. [20].

For example, in the case of the transition to the 778.2 keV level of ^{96}Mo, a peak at 778.2 keV should be present in the energy spectrum accumulated with the Ru sample. To estimate an upper limit the spectrum was fitted in the energy interval 744–799 keV using a model made of four Gaussian functions at the energies of 768.4 keV (γ peak from ^{214}Bi), 786.0 keV (^{214}Pb), 794.9 keV (^{228}Ac) and 778.2 keV (the expected effect) with the energy resolution FWHM = 2.0 keV, and a linear function representing the background (see Fig. 4). The fit using the chi-square method ($\chi^2/\mathrm{ndf} = 38.8/42 = 0.92$, where ndf is the number of degrees of freedom) results in a peak area of $S = (-16.9 \pm 14.6)$ counts, which gives no evidence for the effect. In accordance with the procedure in Ref. [28], one should take 10.5 counts which can be excluded at 90% C.L. (fits in other energy intervals give close results). Taking into account the detection efficiency (2.38%), we have obtained the following limit:

$$T_{1/2}^{(2\nu+0\nu)\varepsilon\beta^+}(\text{g.s.} \to 2^+, 778.2\ \text{keV}) \geqslant 2.3 \times 10^{20}\ \text{yr},$$

Similar fits allow us to set limits on possible transitions to other excited levels in the $\varepsilon\beta^+$ decay of ^{96}Ru; obtained results are listed in Table II.

C. Double-electron captures in ^{96}Ru

Double-electron captures in ^{96}Ru lead to the creation of holes in the atomic shells of ^{96}Mo. In the $2\nu2\varepsilon$ process, all the energy release (except the part spent on atomic shell excitation) is taken away by two neutrinos. The energy threshold in the current measurements (around 50 keV) does not allow us to search for the deexcitation processes in the atomic shell (which have energies less than 20 keV), thus we cannot derive limits for the g.s. to g.s. $2\nu2\varepsilon$ capture.

In neutrinoless 2ε capture, we suppose (as do other articles on the subject) that the energy excess is taken away by (bremsstrahlung) γ quanta with energy $E_\gamma = Q_{2\beta} - E_{b1} - E_{b2} - E_{\mathrm{exc}}$, where E_{bi} is the binding energy of the ith captured electron on the atomic shell, and E_{exc} is the energy of the populated (g.s. or excited) level of ^{96}Mo. In the case of a transition to an excited level, in addition to the initial γ quantum, other γ's will be emitted in the nuclear deexcitation process.

We did not observe peaks with energies expected in the 2ε decays of ^{96}Ru in the experimental data. Limits on the areas of the peaks were obtained using the fitting procedure as explained in the previous section. Figure 5 shows an interval of the spectrum around 1921.8 keV and its fit by the sum of a straight line (representing the background) and the peak with an energy of 1921.8 keV expected in the ^{96}Ru resonant decay to the 2700.2 keV level of ^{96}Mo.

All the obtained $T_{1/2}$ limits, together with the energies of the γ lines which were used to set the $T_{1/2}$ limits and corresponding detection efficiencies, are summarized in Table II.

D. $2\beta^-$ decay ^{104}Ru → ^{104}Pd*

For the $2\beta^-$ decay of ^{104}Ru, only one excited level (2^+, 555.8 keV) can be populated [see Fig. 1(b)]. The peak at the energy 555.8 keV is absent in the experimental data (see Fig. 3). The fit of the spectrum accumulated over 5479 h was bounded within the 530–600 keV interval. The best fit ($\chi^2/\mathrm{ndf} = 38.2/43 = 0.89$) was achieved in the energy interval 540–590 keV. The derived area of the effect, (-17.4 ± 18.8) counts, corresponds to $\lim S = 16.2$ counts at 90% C.L. [28]. Using the number of ^{104}Ru nuclei in the purified ruthenium sample ($N = 7.98 \times 10^{23}$) and the very close detection efficiencies for the 555.8 keV γ quanta in the case of the 2ν and 0ν modes (3.09% and 3.06%, respectively), the following half-life limits were reached:

$$T_{1/2}^{2\nu2\beta^-}(\text{g.s.} \to 2^+, 555.8\ \text{keV}) \geqslant 6.6 \times 10^{20}\ \text{yr},$$
$$T_{1/2}^{0\nu2\beta^-}(\text{g.s.} \to 2^+, 555.8\ \text{keV}) \geqslant 6.5 \times 10^{20}\ \text{yr}.$$

E. Theoretical estimates

The theoretical estimations of Table II were obtained by using a higher quasiparticle random-phase approximation (QRPA) framework [30,31] with detailed expressions given in [32,33]. For the neutrinoless modes of decay the unitary correlation operator method (UCOM) short-range correlations [34] were used. All computational details are given in a recent article [35]. Estimates coming from other sources [36–38] are indicated in the table. In addition, a summary of other theoretical results can be found in our previous work [20].

IV. DISCUSSION

Experimental searches for $2\beta^+/\varepsilon\beta^+/2\varepsilon$ processes are not as popular as those for $2\beta^-$ decays. There are three reasons for such a situation: (i) These nuclei mostly have low natural

TABLE II. The half-life limits on 2β processes in ^{96}Ru and ^{104}Ru isotopes together with theoretical predictions. The energies of the γ lines (E_γ), which were used to set the $T_{1/2}$ limits, are listed with the corresponding detection efficiencies (η). The theoretical $T_{1/2}$ values for 0ν mode are given for $m_\nu = 1$ eV.

Process of decay		Level of daughter nucleus (keV)	E_γ (keV)	η (%)	Expt. $T_{1/2}$ (yr) at 90% C.L.		Theor. $T_{1/2}$ (yr)
					Present work	Ref. [22]	
^{96}Ru $\to$ ^{96}Mo							
$2\beta^+$	2ν	g.s.	511.0	10.36	$\geqslant 1.4 \times 10^{20}$	$\geqslant 5.0 \times 10^{19}$	$1.2 \times 10^{26} - 1.0 \times 10^{27}$
	0ν	g.s.	511.0	10.31	$\geqslant 1.3 \times 10^{20}$	$\geqslant 5.0 \times 10^{19}$	$5.9 \times 10^{27} - 1.0 \times 10^{28}$
$\varepsilon\beta^+$	2ν	g.s.	511.0	6.12	$\geqslant 8.0 \times 10^{19}$	$\geqslant 5.5 \times 10^{19}$	$2.0 \times 10^{21} - 2.3 \times 10^{22}$
		2^+ 778.2	778.2	2.38	$\geqslant 2.3 \times 10^{20}$	$\geqslant 2.7 \times 10^{19}$	$1.3 \times 10^{27} - 1.2 \times 10^{31}$
		0^+ 1148.1	778.2	2.26	$\geqslant 2.1 \times 10^{20}$	$\geqslant 1.8 \times 10^{19}$	$6.1 \times 10^{24} - 1.9 \times 10^{26}$
		2^+ 1497.8	778.2	1.56	$\geqslant 1.5 \times 10^{20}$	$\geqslant 1.3 \times 10^{19}$	$2.1 \times 10^{33} - 1.6 \times 10^{37}$
		2^+ 1625.9	847.7	1.96	$\geqslant 3.1 \times 10^{20}$	$\geqslant 1.6 \times 10^{19}$	$>3.4 \times 10^{38}$
	0ν	g.s.	511.0	5.89	$\geqslant 7.7 \times 10^{19}$	$\geqslant 5.5 \times 10^{19}$	$5.0 \times 10^{26} - 1.0 \times 10^{27}$
		2^+ 778.2	778.2	2.39	$\geqslant 2.3 \times 10^{20}$	$\geqslant 2.6 \times 10^{19}$	–
		0^+ 1148.1	778.2	2.26	$\geqslant 2.1 \times 10^{20}$	$\geqslant 1.8 \times 10^{19}$	$(1.0 - 8.2) \times 10^{28}$
		2^+ 1497.8	778.2	1.56	$\geqslant 1.5 \times 10^{20}$	$\geqslant 1.3 \times 10^{19}$	–
		2^+ 1625.9	847.7	1.96	$\geqslant 3.1 \times 10^{20}$	$\geqslant 1.6 \times 10^{19}$	–
2ε	2ν	g.s.	–	–	–	–	$4.7 \times 10^{20} - 3.9 \times 10^{21}$
		2^+ 778.2	778.2	2.83	$\geqslant 2.6 \times 10^{20}$	$\geqslant 6.5 \times 10^{19}$	$4.2 \times 10^{28} - 2.2 \times 10^{32}$
		0^+ 1148.1	778.2	2.64	$\geqslant 2.5 \times 10^{20}$	$\geqslant 4.2 \times 10^{19}$	$4.2 \times 10^{21} - 9.2 \times 10^{22}$
		2^+ 1497.8	778.2	1.82	$\geqslant 1.7 \times 10^{20}$	$\geqslant 3.0 \times 10^{19}$	$1.8 \times 10^{29} - 6.5 \times 10^{32}$
		2^+ 1625.9	848.2	2.29	$\geqslant 3.6 \times 10^{20}$	$\geqslant 3.9 \times 10^{19}$	$>1.6 \times 10^{29}$
		2^+ 2095.8	778.2	2.56	$\geqslant 2.4 \times 10^{20}$	$\geqslant 4.3 \times 10^{19}$	–
		2^+ 2426.1	778.2	2.28	$\geqslant 2.1 \times 10^{20}$	$\geqslant 3.5 \times 10^{19}$	–
		$(0)^+$ 2622.5	778.2	2.61	$\geqslant 2.4 \times 10^{20}$	$\geqslant 4.6 \times 10^{19}$	–
2ε	0ν	2^+ 778.2	778.2	2.61	$\geqslant 2.4 \times 10^{20}$	$\geqslant 6.4 \times 10^{19}$	–
		0^+ 1148.1	778.2	2.46	$\geqslant 2.3 \times 10^{20}$	$\geqslant 4.1 \times 10^{19}$	–
		2^+ 1497.8	778.2	1.67	$\geqslant 1.6 \times 10^{20}$	$\geqslant 2.9 \times 10^{19}$	–
		2^+ 1625.9	847.7	2.12	$\geqslant 3.3 \times 10^{20}$	$\geqslant 3.8 \times 10^{19}$	–
		2^+ 2095.8	778.2	2.39	$\geqslant 2.2 \times 10^{20}$	$\geqslant 4.3 \times 10^{19}$	–
		2^+ 2426.1	778.2	2.20	$\geqslant 2.1 \times 10^{20}$	$\geqslant 3.4 \times 10^{19}$	–
		$(0)^+$ 2622.5	778.2	2.60	$\geqslant 2.4 \times 10^{20}$	$\geqslant 4.5 \times 10^{19}$	–
$2K$	0ν	g.s.	2674.5	1.56	$\geqslant 1.0 \times 10^{21}$	$\geqslant 5.4 \times 10^{19}$	2.8×10^{34} [37]
KL	0ν	g.s.	2691.6	1.58	$\geqslant 2.3 \times 10^{20}$	$\geqslant 6.9 \times 10^{19}$	–
$2L$	0ν	g.s.	2708.7	1.55	$\geqslant 2.3 \times 10^{20}$	$\geqslant 6.9 \times 10^{19}$	–
Resonant KL	0ν	2^+ 2700.2	1921.8	0.60	$\geqslant 2.0 \times 10^{20}$	$\geqslant 2.7 \times 10^{19}$	$3.0 \times 10^{26} - 6.0 \times 10^{34}$ [38]
Resonant $2L$	0ν	2712.7	812.6	2.28	$\geqslant 3.6 \times 10^{20}$	$\geqslant 2.0 \times 10^{19}$	$4.4 \times 10^{31} - 2.3 \times 10^{32}$
^{104}Ru $\to$ ^{104}Pd							
$2\beta^-$	2ν	2^+ 555.8	555.8	3.09	$\geqslant 6.6 \times 10^{20}$	$\geqslant 1.9 \times 10^{20}$	$>1.8 \times 10^{28}$ [36]
	0ν	2^+ 555.8	555.8	3.06	$\geqslant 6.5 \times 10^{20}$	$\geqslant 1.9 \times 10^{20}$	–

abundance, usually less than 1%, with few exceptions [2] (and ^{96}Ru with $\delta = 5.54\%$ is among them); (ii) energy available for positrons is related to the energy release $Q_{2\beta}$ as $Q_{2\beta} - 4m_ec^2$ (for $2\beta^+$ decay) or $Q_{2\beta} - 2m_ec^2 - E_b$ (for $\varepsilon\beta^+$), where m_ec^2 is the electron rest mass, and E_b is the binding energy of the captured electron on the atomic shell. This leads to smaller phase space factors in comparison with $2\beta^-$ decay, and thus in lower probabilities for $2\beta^+/\varepsilon\beta^+$ processes; and (iii) in searches for x rays emitted in the deexcitation of atomic shells in $\varepsilon\beta^+/2\varepsilon$ decays, detectors with low energy thresholds (and good energy resolution) are needed; in addition, it is difficult to ensure high efficiency for detection of low-energy x rays when external 2β sources are investigated.

As a result, while in searches for neutrinoless $2\beta^-$ decay the sensitivity of $T_{1/2} > 10^{25}$ yr was achieved (for ^{76}Ge [39] and ^{136}Xe [40]), the best $T_{1/2}$ limits achieved in $2\beta^+/\varepsilon\beta^+/2\varepsilon$ experiments are much more modest. Sensitivity $T_{1/2} > 10^{20}$ yr was reached in direct experiments for ^{54}Fe [41], ^{58}Ni [42], ^{64}Zn [43], and ^{92}Mo [44]; and limits $T_{1/2} > 10^{21}$ yr were obtained for ^{40}Ca [45], ^{78}Kr [46], ^{106}Cd [47], ^{112}Sn [48], ^{120}Te [49], and ^{132}Ba [6]. Geochemical experiments currently give an indication on $2\nu2\varepsilon$ capture in ^{130}Ba with $T_{1/2} = (2.2 \pm 0.5) \times 10^{21}$ yr [6] and $T_{1/2} = (6.0 \pm 1.1) \times 10^{20}$ yr [7] (also limit $> 4.0 \times 10^{21}$ yr is known [50]). In addition, an observation of $2\nu2K$ capture in ^{78}Kr was recently claimed; the obtained half-life is $T_{1/2} = 1.4^{+2.2}_{-0.7} \times 10^{22}$ yr (however, a

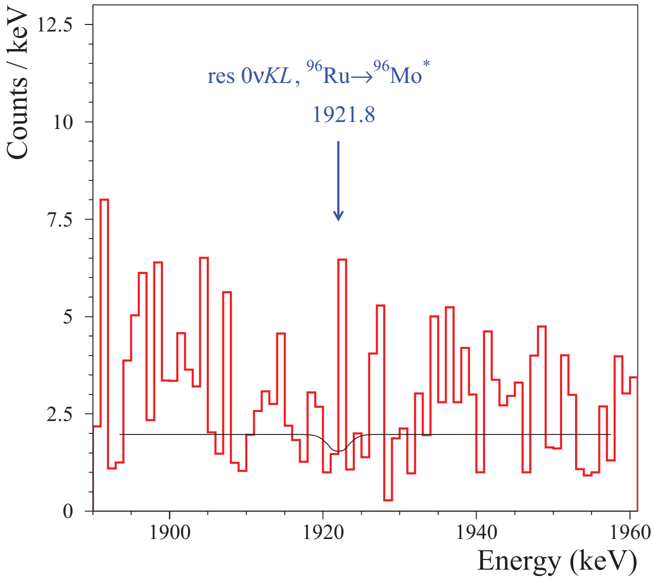

FIG. 5. (Color online) Fragment of the energy spectrum accumulated with the purified Ru sample over 5479 h by the ultralow-background HPGe γ spectrometer. The fit is shown by a solid line. The arrow shows the energy of a peak at 1921.8 keV due to possible resonant $0\nu KL$ capture in ^{96}Ru and further deexcitation of the 2^+, 2700.2 keV level of ^{96}Mo.

cautious limit is also given as $T_{1/2} > 7.0 \times 10^{21}$ yr at 90% C.L.) [51].

As for resonance $0\nu 2\varepsilon$ capture, intensive high-precision measurements of $Q_{2\beta}$ values during last few years (see reviews [52] and references therein) excluded many nuclei from the list of perspective candidates in searches for this exotic process, leaving in the list only ^{152}Gd and ^{156}Dy. While for ^{152}Gd experimental investigations have not been performed to date, for ^{156}Dy the first experimental limits [53] have been set on the level of only $T_{1/2} > 10^{16}$ yr (this is related to the ^{156}Dy low natural abundance $\delta = 0.056\%$ [12]).

Comparing the above described $T_{1/2}$ limits for different isotopes with the values obtained in the present measurements, one could conclude that the latter are on the level of the best results achieved to date in other experiments.

V. CONCLUSIONS

A low-background experiment to search for 2β processes in ^{96}Ru and ^{104}Ru isotopes was carried out over more than 7,600 h in the underground Gran Sasso National Laboratories of the I.N.F.N. measuring ruthenium samples with ultralow-background HPGe detectors. The total exposure of the experiment is 0.56 kg $\times$ yr. Purification of the ruthenium using the electron beam melting method allowed us to reduce the potassium contamination by more than 20 times; activities of ^{226}Ra and ^{106}Ru were decreased as well.

The new improved half-life limits on double-β processes in ^{96}Ru have been set at the level of 10^{20}–10^{21} yr. Moreover, the $2\beta^-$ transition of ^{104}Ru to the excited 2^+ level of ^{104}Pd has been investigated with the same sensitivity. All results give higher values than those recently published [20–22]. However, the limits are still far from the theoretical predictions, with the exception of the $2\nu\varepsilon\beta^+$ channel for ^{96}Ru (g.s. to g.s. transition) and $2\nu 2\varepsilon$ decays with population of the 0^+ levels (g.s. and the first 0^+ level with $E_{\rm exc} = 1148.1$ keV), for which half-lives $T_{1/2} \simeq 10^{21}$–10^{22} yr have been estimated.

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