Regular Article – Experimental Physics

First search for double-β decay of platinum by ultra-low background HP Ge γ spectrometry

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Abstract. A search for double-β processes in ^{190}Pt and ^{198}Pt was realized with the help of ultra-low background HP Ge 468 cm^3 γ spectrometer in the underground Gran Sasso National Laboratories of the INFN (Italy). After 1815 h of data taking with 42.5 g platinum sample, $T_{1/2}$ limits on 2β processes in ^{190}Pt ($\varepsilon\beta^+$ and 2ε) have been established on the level of 10^{14}–10^{16} y, 3 to 4 orders of magnitude higher than those known previously. In particular, a possible resonant double-electron capture in ^{190}Pt was restricted on the level of 2.9×10^{16} y at 90% C.L. In addition, $T_{1/2}$ limit on $2\beta^-$ decay of ^{198}Pt ($2\nu + 0\nu$) to the 2_1^+ excited level of ^{198}Hg has been set at the first time: $T_{1/2} > 3.5 \times 10^{18}$ y. The radiopurity level of the used platinum sample is reported.

1 Introduction

The investigation of the double-beta (2β) decay is considered now as one of the most sensitive probe of physics beyond the Standard Model of particles and interactions [1–8]. The observation of the neutrinoless (0ν) mode of 2β decay could establish an absolute scale of the neutrino mass and the neutrino mass hierarchy, clarify the nature of the neutrino (Dirac or Majorana particle) and check the conservation of the lepton number. The process also depends on the possible existence of light Nambu-Goldstone bosons (Majorons) and hypothetical admixture of right-handed currents in weak interaction.

At present, the two neutrino (2ν) $2\beta^-$ decay, process of nuclear transformation $(A, Z) \rightarrow (A, Z+2) + 2e^- + 2\widetilde{\nu}_e$, was observed in 10 isotopes with the half-lives in the range of 10^{18}–10^{24} y, while only the limits on $0\nu 2\beta^-$ decays up to 10^{23}–10^{25} y were set in numerous experiments [9–11].

The sensitivities of experiments to search for 2β processes, which transform (A, Z) nucleus to $(A, Z-2)$, double-electron capture (2ε), electron capture with emission of positron ($\varepsilon\beta^+$), and double-positron ($2\beta^+$) decay, are on the level of 10^{15}–10^{21} y [10–24]; even though allowed in the Standard Model, the two-neutrino mode is

still not observed[1]. At the same time, the investigation of neutrinoless 2ε and $\varepsilon\beta^+$ processes could give important information about the possible contribution of the right-handed currents to the neutrinoless double-β^- decay rate [26]. Therefore developments of experimental methods to search for "double beta-plus" processes are required.

The ^{190}Pt is one of the twenty-two potentially $\varepsilon\beta^+$ active nuclei [10,11]. The energy of the double-beta decay is $Q_{2\beta} = (1383 \pm 6)$ keV [27]. The decay scheme of the ^{190}Pt is presented in fig. 1.

It is also worth noting that in the capture of two electrons from external atomic shells, the energy release is close to the energy of the excited level of ^{190}Os with $E_{exc} = 1382.4$ keV [28]. Such a coincidence could give a resonant enhancement of the neutrinoless double-electron capture as a result of energy degeneracy. The possibility of the resonant neutrinoless double-electron capture was discussed in refs. [29–31], where an enhancement of the rate by some orders of magnitude was predicted for the case of coincidence between the released energy and the energy of an excited state. According to [32], high Z atoms are

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[1] An indication on the double-β decay of ^{130}Ba was obtained in [25] by the geochemical method. However, this result has to be confirmed by direct counting experiments.
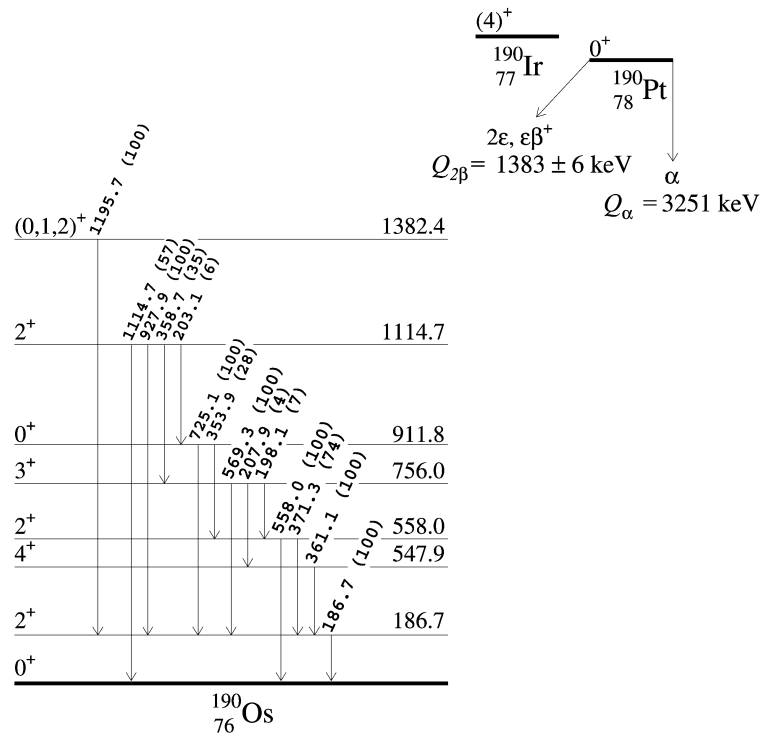
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Fig. 1. The decay scheme of the ^{190}Pt [28]. The energies of the excited levels and of the emitted γ quanta are in keV (the relative intensities of the γ quanta are given in parentheses). $Q_{2\beta}$ is the double-beta decay energy.

strongly favored to search for resonant 2ε decay. It should be noted that ^{190}Pt has the greatest Z value among the nuclei for which a resonant double-electron capture is possible. Isotope ^{190}Pt is noted as one of the most promising to search for resonant neutrinoless double-electron capture in the recent review [33].

Unfortunately, the natural abundance of ^{190}Pt is very low: $\delta = (0.014 \pm 0.001)\%$ [34]. Perhaps this fact, together with the rather high cost of platinum, explains the absence of experimental results (after the one and only old study [35]) to search for 2β decay of this nuclide. A modest limit on $\varepsilon\beta^+$ decay of ^{190}Pt on the level of 3.1×10^{11} y was calculated in [10,11] on the basis of the early experiment [35] with corrections on the decay energy, experimental efficiency and the natural isotopic abundance.

Another platinum isotope ^{198}Pt is potentially $2\beta^-$ active, with the energy of the decay $Q_{2\beta} = (1047 \pm 3)$ keV [27]. The isotopic abundance of ^{198}Pt is $(7.163 \pm 0.055)\%$ [34]. The decay scheme of ^{198}Pt is presented in fig. 2. As one can see, the $2\beta^-$ decay of ^{198}Pt to the first-excited 2^+ level of ^{198}Hg can be searched for by using an external γ detector.

The aim of this study was the search for double-β processes in ^{190}Pt and ^{198}Pt with the help of ultra-low background HP Ge γ spectrometry.

2 Experiment

Two platinum cups and one lid designed for chemistry purposes with the total mass of 42.53 g were used in the

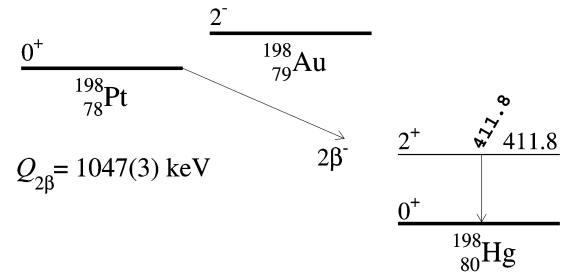**Fig. 2.** Decay scheme of ^{198}Pt [36]. The energy of the excited level and of the γ quantum are in keV. $Q_{2\beta}$ is the double-beta decay energy.

experiment. Taking into account the isotopic composition of the platinum, the sample contains 1.84×10^{19} nuclei of ^{190}Pt and 9.40×10^{21} nuclei of ^{198}Pt. The search for double-β decay of platinum was realized at the Laboratori Nazionali del Gran Sasso of the INFN (average overburden of ≈ 3600 meters of water equivalent) [37,38] with a p-type HP Ge detector (GeCris, 468 cm^3 of volume; we refer readers to ref. [39] where the detailed picture of the sample and detector set-up is given). The energy resolution of the spectrometer is FWHM = 2.0 keV for the 1333 keV γ line of ^{60}Co. The data with the sample were accumulated over 1815.4 h, while the background spectrum was taken over 1045.6 h. The spectra normalized on the time of measurements are presented in fig. 3.
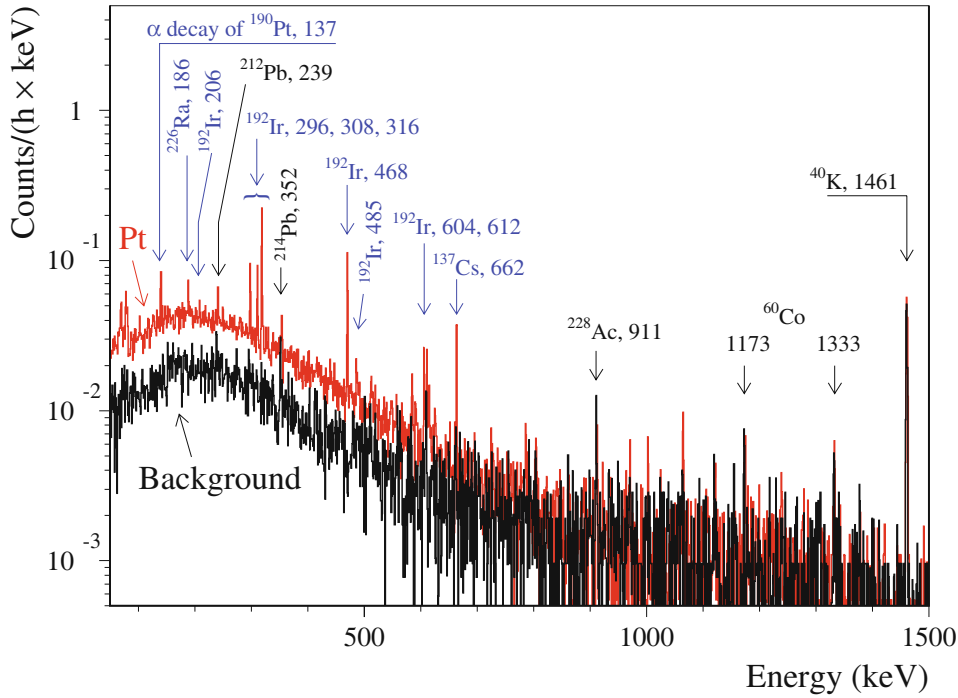
Fig. 3. (Color online) Energy spectra measured with the 42.5 g platinum sample over 1815 h (Pt) and without sample over 1046 h (Background) by ultra-low background HP Ge γ spectrometer. The energy of the γ lines is in keV.

3 Results and discussion

3.1 Radioactive contamination of platinum

There is a certain difference between the Pt and the background spectra, mainly due to the contamination of the platinum by ^{192}Ir ($T_{1/2} = 73.831$ d [40]). The activity of ^{192}Ir is equal to (40 ± 5) mBq/kg. The radioactive iridium could appear in platinum due to the cosmogenic activation of Pt by cosmic rays on the Earth surface. In addition, iridium usually accompanies platinum in ores. Therefore ^{192}Ir can be created as a result of neutron capture by ^{191}Ir which is one of two naturally occurring iridium isotopes ($\delta = 37.3\%$, the cross-section for thermal neutrons is $\sigma = 954$ b [40]). However, the half-life of the ground state of ^{192}Ir is $T_{1/2} = 73.8$ d [40], and the exponential decrease in time of the ^{192}Ir activity should be observed during our 75.6 d measurements. In fact, the behaviour of the counting rate in the f.e. 468.1 keV γ peak of ^{192}Ir cannot be explained by this assumption (see fig. 4). A fit of the data by the exponential function corresponding to the decay of ^{192}Ir gives a too large value of χ^2/n.d.f. = 22.6/9 = 2.5. At the same time the data is very well described (χ^2/n.d.f. = 7.8/9 = 0.87) by the exponential decay with the half-life 241 y (decay of ^{192m}Ir, see fig. 4). The same behaviour was observed for two other intensive γ lines of ^{192}Ir with energies 296.0 keV and 316.5 keV. Thus, the ^{192}Ir activity should be ascribed not to the ground state, but to the isomeric ^{192m}Ir level with $E_{exc} = 168.1$ keV and $T_{1/2} = 241$ y [41]. This isomeric state decays to the ground state of ^{192}Ir emitting 155.1 keV and 13.0 keV γ's which are, however, strongly
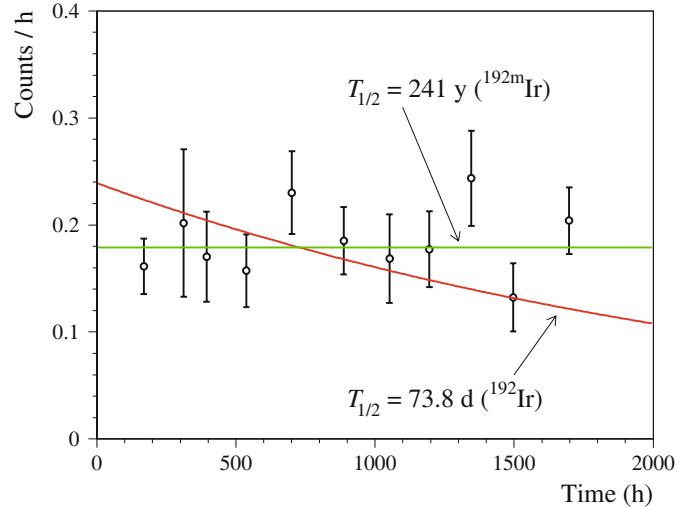

Fig. 4. (Color online) Behaviour in time of the counting rate in the 468.1 keV γ peak from the decays of ^{192}Ir in the platinum sample measured by ultra-low background HP Ge γ spectrometer. The dependence can be explained assuming the appearance of ^{192}Ir in the platinum sample after the decay of ^{192m}Ir. See text.

converted to electrons (coefficients of conversion are equal to $\alpha_{13} = 57000$ and $\alpha_{155} = 1026$ [41]). This explains the absence of the 155.1 keV peak in our data.

A peak at the energy 137 keV is caused by the α decay of ^{190}Pt to the lowest-excited 137.2 keV level of ^{186}Os (the first evidence is reported in [39]). We have also observed some excess of events in the 662 keV peak of ^{137}Cs. The

Table 1. Radioactive contamination of the platinum sample measured with HP Ge detector. Gamma emitters and energies of γ lines used to determine the activity of the isotopes are specified in the third column. The upper limits are given at 90% C.L., and the uncertainties of the measured activities are estimated at 68% C.L.

Chain	Nuclide (sub-chain)	γ emitters (E_γ, keV)	Activity (mBq/kg)
	^{40}K	(1460.8)	≤ 25
	^{60}Co	(1173.2)	≤ 1.5
	^{137}Cs	(661.6)	7 ± 1
	^{192}Ir	(296.0, 316.5, 468.1)	40 ± 5
^{232}Th	^{228}Ra	^{228}Ac (911.2)	≤ 7
	^{228}Th	^{212}Pb (238.6),	
		^{208}Tl (583.2, 2614.6)	≤ 7
^{235}U	^{235}U	^{235}U (185.7)	≤ 16
	^{231}Pa	^{231}Pa (283.7, 300.1)	≤ 66
	^{227}Ac	^{227}Th (236.0)	≤ 13
^{238}U	^{238}U	^{234m}Pa (766.4)	≤ 68
	^{226}Ra	^{214}Pb (352.0),	
		^{214}Bi (609.3, 1764.5)	≤ 3
	^{210}Pb	^{210}Pb (46.5)	≤ 34000

response functions of the detector to decays of ^{40}K, ^{60}Co, ^{137}Cs, ^{192}Ir, U/Th daughters in the platinum sample were simulated by EGS4 code [42] with initial kinematics given by the DECAY0 event generator [43]. The activity of ^{137}Cs and ^{192}Ir, as well as upper limits on contamination by ^{40}K, ^{60}Co and U/Th daughters are presented in table 1.

The investigation of radioactive contaminants in platinum samples is important also, *e.g.*, when selecting the materials to build platinum crucibles to be used for growing radiopure inorganic crystal scintillators.

3.2 Search for double-β processes in ^{190}Pt and ^{198}Pt

We do not observe any peaks in the spectrum accumulated with the platinum sample which could indicate double-β activity of ^{190}Pt or ^{198}Pt. Therefore only lower half-life limits ($\lim T_{1/2}$) can be set according to the formula: $\lim T_{1/2} = N \cdot \eta \cdot t \cdot \ln 2 / \lim S$, where N is the number of potentially 2β unstable nuclei, 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 given confidence level (C.L., all the limits obtained in the present study are given at 90% C.L.). The efficiencies
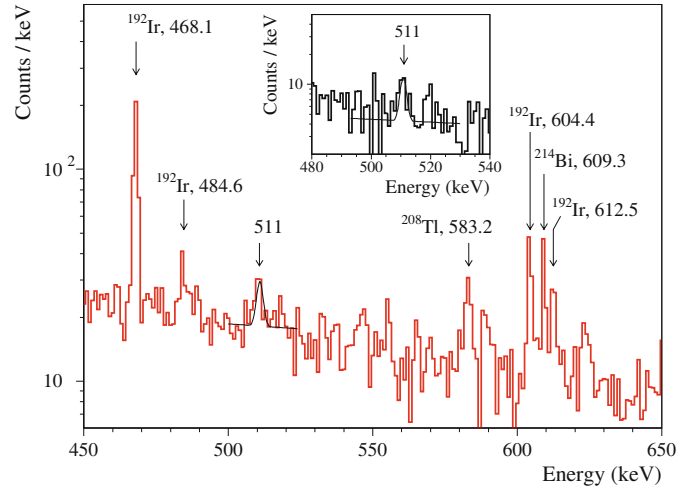

Fig. 5. (Color online) Fragment of the energy spectra accumulated with the platinum sample over 1815 h by ultra-low background HP Ge γ spectrometer. (Inset) The energy spectrum accumulated without sample over 1046 h. The fits of the 511 keV annihilation γ peaks are shown by solid lines. The energy of the γ lines is in keV.

of the detector to the double-β processes in the platinum isotopes were also calculated with the EGS4 code [42] and DECAY0 event generator [43].

3.2.1 Electron capture with positron emission in ^{190}Pt

One positron can be emitted in the $\varepsilon\beta^+$ decay of ^{190}Pt with energy up to (361 ± 6) keV. The annihilation of the positron will give two 511 keV γ's leading to an extra rate in the annihilation peak.

Part of the spectrum in the energy interval 450–650 keV is shown in fig. 5. There are peculiarities in both the spectra accumulated with the platinum sample (36 ± 13 counts at 510.9 ± 0.2 keV) and in the background spectrum (12 ± 5 counts at 510.8 ± 0.3 keV), which can be ascribed to annihilation peaks. The decays of ^{137}Cs and ^{192}Ir, present in the used platinum sample, provide no contribution to the peak. The difference in the areas of the annihilation peak (15 ± 16) counts, which can be attributed to electron capture with positron emission in ^{190}Pt, gives no indication on the effect. In accordance with the Feldman-Cousins procedure [44] (here and below we use this approach to estimate values of $\lim S$ for all the processes searched for) we should take $\lim S = 41$ counts which can be excluded at 90% C.L. Taking into account the detection efficiency ($\eta = 14.2\%$), we have estimated a limit on the half-life of ^{190}Pt relatively to two-neutrino $\varepsilon\beta^+$ decay as:

$$T_{1/2}^{2\nu\varepsilon\beta^+} (\text{g.s.} \rightarrow \text{g.s.}) \geq 9.2 \times 10^{15} \text{ y.}$$

The detection efficiency in a case of neutrinoless $\varepsilon\beta^+$ decay is slightly lower (13.9%), which leads to the limit

$$T_{1/2}^{0\nu\varepsilon\beta^+} (\text{g.s.} \rightarrow \text{g.s.}) \geq 9.0 \times 10^{15} \text{ y.}$$
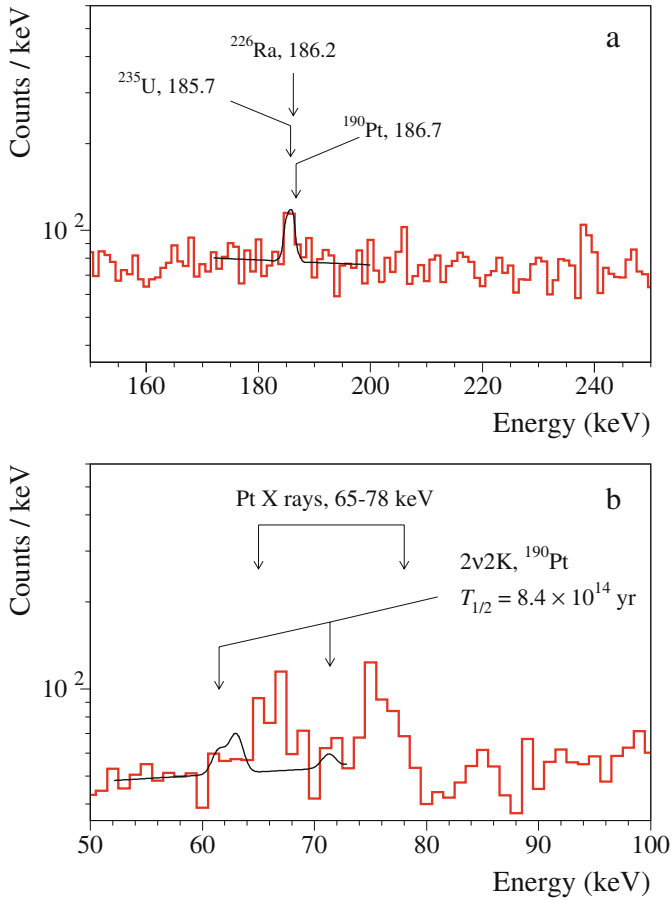
Fig. 6. (Color online) (a) Part of the energy spectrum accumulated with the platinum sample over 1815 h in the vicinity of γ peak expected for $\varepsilon\beta^+$ decay ^{190}Pt to the first-excited level of ^{190}Os with the energy 186.7 keV. Fit of the peak presented in the data is shown by solid line. (b) Low-energy part of the spectrum. The distribution expected for $2\nu2K$ decay of ^{190}Pt with the half-life 8.4×10^{14} y excluded at 90% C.L. is shown by solid line.

In the $2\nu\varepsilon\beta^+$ decay, the first-excited level of ^{190}Os could also be populated with emission of γ quanta with the energy 186.7 keV. There is a peak at the energy of (185.6 ± 0.3) keV with the area (86 ± 21) counts in the spectrum accumulated with the platinum sample (see fig. 6a). Most likely, this peak can be explained by the α decay of ^{235}U to the excited levels of ^{231}Th. Some parts of the peak can be due to the α decay of ^{226}Ra to the excited level 186.2 keV of ^{222}Rn. However, we cannot surely estimate a contribution to this peak either from the decay of ^{235}U or from the decay of ^{226}Ra. For ^{235}U we set only the limit on the activity of ^{235}U in the platinum (≤ 16 mBq/kg). Even a much stronger limit (≤ 3 mBq/kg) was obtained on the activity of ^{226}Ra by the analysis of ^{214}Pb and ^{214}Bi peaks. Because of certain difficulties to take into account the contribution of the natural activity to the area of the 186.7 keV peak, we ascribe, very conservatively, all the area of the peak to the $\varepsilon\beta^+$ decay of ^{190}Pt to the first-excited level of ^{190}Os. By using the Feldman-Cousins procedure we have obtained a

value $\lim S = 120$ events which can be excluded at 90% of C.L.[2]. This leads to the following limit on the process: $T_{1/2}^{2\nu\varepsilon\beta^+} (\text{g.s.} \rightarrow 186.7\,\text{keV}) \geq 9.0\times10^{14}$ y. However, a better limit can be set by using the estimation of the annihilation peak area (the detection efficiency for the process is $\eta = 13.0\%$)

$$T_{1/2}^{2\nu\varepsilon\beta^+} (\text{g.s.} \rightarrow 186.7\,\text{keV}) \geq 8.4 \times 10^{15}\,\text{y}.$$

The same limit was set for the neutrinoless $\varepsilon\beta^+$ decay of ^{190}Pt to the excited 186.7 keV level of ^{190}Os.

3.2.2 Double-electron capture in ^{190}Pt

The $2\nu2K$ capture in ^{190}Pt to the ground state of ^{190}Os will give a cascade of X-rays and Auger electrons with the individual energies up to 73.8 keV [40]. The most intensive X-ray lines should have the energies 61.5 (27.4%), 63.0 (46.7%), 71.1 (5.5%), 71.4 (10.6%) and 73.4 keV (3.7%). Contributions to the spectrum in this energy region (see fig. 6b) are expected from the decay of ^{192}Ir, the excitation of Pt by the decays of ^{137}Cs, and also from the decays of U/Th daughters contained in the materials of the detector. To set a limit on the process searched for, the energy spectrum was fitted by the model consisting of the sum of five Gaussians (to describe the expected 61.5, 63.0, 71.1, 71.4 and 73.4 keV peaks in ^{190}Os), two Gaussians (65.1 and 66.8 keV X-ray lines due to excitation of Pt), and a polynomial function of the first degree (background). A fit in the energy interval $(52–73)$ keV gives the area of the $2\nu2K$ effect as (26 ± 21) counts with the value $\chi^2 = 29.8/13 = 2.3$, the average background level ≈ 52 counts per keV, and the total area of the 65.1 and 66.8 keV X-ray peaks 137 ± 27 counts. According to the Feldman-Cousins procedure we should take $\lim S = 60$ counts at 90% C.L. Therefore, taking into account the efficiency to detect the expected effect (1.9%), we set the following limit on the $2\nu2K$ capture in ^{190}Pt:

$$T_{1/2}^{2\nu2K}(\text{g.s.} \rightarrow \text{g.s.}) \geq 8.4 \times 10^{14}\,\text{y}.$$

The distribution of the excluded effect of the $2\nu2K$ decay in ^{190}Pt is shown at fig. 6b.

The $Q_{2\beta}$ energy of ^{190}Pt allows also the population of several excited levels of ^{190}Os. In a subsequent de-excitation process, one or few cascade γ quanta (together with conversion electrons and e^+e^- pairs) will be emitted. To estimate the limits on the two-neutrino double-electron capture in ^{190}Pt, the energy spectrum accumulated with the platinum sample was analyzed in different energy intervals. The results of the estimations are presented in table 2, where the energies of the γ peaks, used to derive the limits, are also specified.

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[2] The Feldman-Cousins procedure gives, in this case, for $S = 86 \pm 21$ counts positive observation with $S = [49, 120]$ counts at 90% C.L., thus stating that $S < 120$ counts has in fact the confidence level even greater than 90% C.L. We adopt the same conservative politics also for other cases in this article.

Table 2. Half-life limits on 2β processes in ^{190}Pt and ^{198}Pt isotopes. The energies of the γ lines (E_γ), which were used to set the $T_{1/2}$ limits, are listed in column 4 with the corresponding efficiencies (η) in column 5. $T_{1/2}$ limits are derived in the present work at 90% C.L., while the limit from [10,11,35] is given at 68% C.L.

Process of decay	Decay mode	Level of daughter nucleus (keV)	E_γ (keV)	η	$T_{1/2}$ (y) Present work	$T_{1/2}$ (y) [10,11,35]
^{190}Pt $\rightarrow$ ^{190}Os						
$\varepsilon\beta^+$	2ν	g.s.	511	14.2%	$> 9.2 \times 10^{15}$	–
	0ν	g.s.	511	13.9%	$> 9.0 \times 10^{15}$	$> 3.1 \times 10^{11}$
	$2\nu + 0\nu$	2^+ 186.7	511	13.0%	$> 8.4 \times 10^{15}$	–
2K	2ν	g.s.	61.5–73.4	1.9%	$> 8.4 \times 10^{14}$	–
	2ν	2^+ 186.7	61.5–73.4	2.0%	$> 8.8 \times 10^{14}$	–
	2ν	2^+ 558.0	558.0	4.0%	$> 5.6 \times 10^{15}$	–
	2ν	0^+ 911.8	725.1	4.4%	$> 4.5 \times 10^{15}$	–
	2ν	2^+ 1114.7	1114.7	1.6%	$> 1.0 \times 10^{16}$	–
2K	0ν	g.s.	1229–1241	4.7%	$> 5.7 \times 10^{15}$	–
KL	0ν	g.s.	1290–1304	4.6%	$> 1.7 \times 10^{16}$	–
2L	0ν	g.s.	1351–1367	4.6%	$> 3.1 \times 10^{16}$	–
2ε	0ν	2^+ 186.7	186.7	3.1%	$> 6.9 \times 10^{14}$	–
	0ν	2^+ 558.0	558.0	3.1%	$> 4.5 \times 10^{15}$	–
	0ν	0^+ 911.8	725.1	3.6%	$> 3.6 \times 10^{15}$	–
	0ν	2^+ 1114.7	1114.7	1.5%	$> 9.8 \times 10^{15}$	–
Resonant MM, MN, NN	$2\nu + 0\nu$	$(0, 1, 2^+)$ 1382.4	1195.7	4.5%	$> 2.9 \times 10^{16}$	–
^{198}Pt $\rightarrow$ ^{198}Hg						
$2\beta^-$	$2\nu + 0\nu$	2^+ 411.8	411.8	7.5%	$> 3.5 \times 10^{18}$	–

In the neutrinoless double-electron capture to the ground state of the daughter nuclei, in addition to the X-rays, some other particle(s) should be emitted to take away the rest of the energy. Usually one bremsstrahlung γ quantum is assumed. The energy of the γ quantum is expected to be equal to $E_\gamma = Q_{2\beta} - E_{b1} - E_{b2}$, where E_{b1} and E_{b2} are the binding energies of the first and of the second captured electrons on the atomic shell. The binding energies on the K, L_1, L_2 and L_3 shells in Os are equal to $E_K = 73.9$ keV, $E_{L_1} = 13.0$ keV, $E_{L_2} = 12.4$ keV and $E_{L_3} = 10.9$ keV [40], respectively. Therefore, the expected energies of the γ quanta for the $0\nu2\varepsilon$ capture in ^{190}Pt to the ground state of ^{190}Os are in the intervals: i) $E_\gamma = (1229–1241)$ keV for the $0\nu2K$; ii) $E_\gamma = (1290–1304)$ keV for the $0\nu KL$; iii) $E_\gamma = (1351–1367)$ keV for the $0\nu2L$ process.

There is only one peak at the energy of 1238 keV with the area (15 ± 4) counts in the spectrum accumulated with the platinum sample in the energy interval of interest (see fig. 7a). More likely this is the γ peak 1238.1 keV from the decays of ^{214}Bi (daughter of ^{226}Ra from ^{238}U family). It is rather difficult to estimate a contribution of the ^{214}Bi γ quanta to the peak area because we do not know exactly

the location of the ^{226}Ra contamination in the materials of the set-up. Conservatively we assume that all the peak area is due to the neutrinoless $2K$ decay of ^{190}Pt. The procedure [44] gives $\lim S = 21.6$ events. Taking into account the calculated efficiency to detect γ quanta with the energy (1229–1241) keV ($\approx 4.7\%$), it gives the following limit on the process searched for:

$$T_{1/2}^{0\nu2K}(\text{g.s.} \rightarrow \text{g.s.}) \geq 5.7 \times 10^{15} \text{ y.}$$

There are no clear peaks in the energy intervals (1290–1304) keV and (1351–1367) keV expected for the $0\nu KL$ and the $0\nu2L$ processes in ^{190}Pt. The experimental spectrum was fitted in the energy interval 1280–1310 keV to find a peak which could be due to the $0\nu KL$ decay and in the energy interval 1340–1365 keV for the $0\nu2L$ decay. The fits given the maximal peak areas 3.3 ± 2.2 events at the energy ≈ 1294 keV, and 1.3 ± 1.6 events at ≈ 1350 keV. By using the Feldman-Cousins approach, we get $\lim S = 6.9$ and $\lim S = 3.9$ counts for the $0\nu KL$ and $0\nu2L$ decay, respectively. Taking into account the calculated detection efficiencies for γ quanta with the energies in these intervals (4.6%) we have obtained the following limits on the
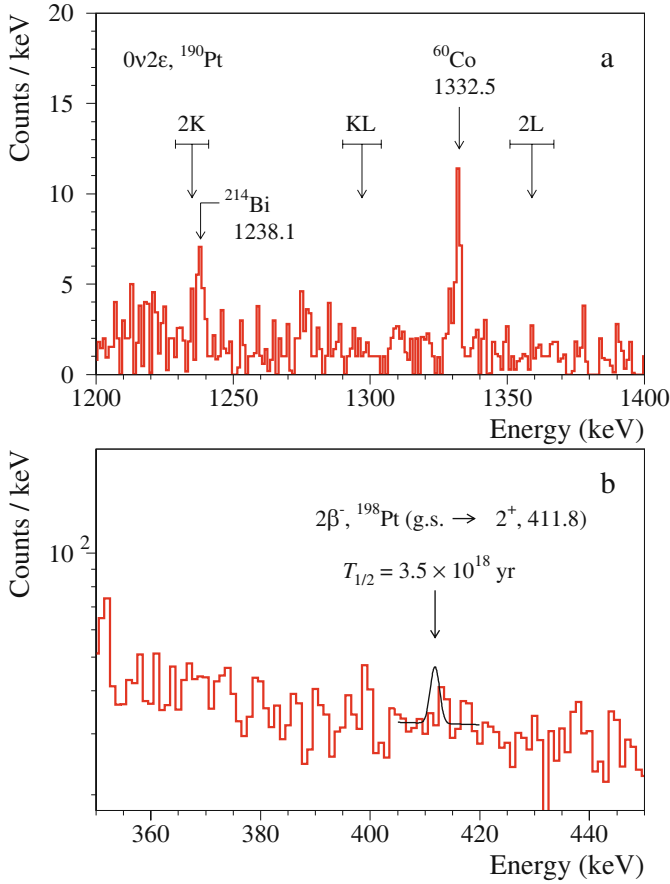
Fig. 7. (Color online) (a) Part of the Pt energy spectrum where peaks from the $0\nu2\varepsilon$ processes in ^{190}Pt ($2K$, KL and $2L$) to the ground state of ^{190}Os are expected. (b) Part of the spectrum in the energy interval (350–450) keV where a γ peak with the energy of 411.8 keV is expected for $2\beta^-$ decay of ^{198}Pt to the excited level 2^+_1 of ^{198}Hg. Area of the peak shown by solid line corresponds to the half-life 3.5×10^{18} y excluded at 90% C.L.

0ν double-electron captures in ^{190}Pt to the ground state of ^{190}Os:

$$T_{1/2}^{0\nu KL}(\text{g.s.} \to \text{g.s.}) \geq 1.7 \times 10^{16} \text{ y},$$
$$T_{1/2}^{0\nu 2L}(\text{g.s.} \to \text{g.s.}) \geq 3.1 \times 10^{16} \text{ y}.$$

Limits on the $0\nu2\varepsilon$ decay of ^{190}Pt to the excited levels of ^{190}Os were obtained in the similar way as for the 2ν mode by analysis of the experimental data in the energy intervals where intensive γ peaks from the processes are expected.

The limits on double-electron capture in ^{190}Pt to the ground and excited levels of ^{190}Os are presented in table 2.

3.2.3 Resonant 2ε capture in ^{190}Pt

The atomic-mass difference between ^{190}Pt and ^{190}Os atoms is very close to the energy of the excited level 1382.4 keV in ^{190}Os. A resonance transition could be fulfilled if two external electrons were captured. There is no

peak in the energy spectrum accumulated with the platinum sample with the energy 1195.7 keV expected from de-excitation of the 1382.4 keV level of ^{190}Os (see fig. 1). Taking into account the detection efficiency for 1195.7 keV γ quanta (4.5%) we set the following limit on the decay:

$$T_{1/2}^{2\varepsilon(0\nu+2\nu)}(\text{g.s.} \to 1382.4 \text{ keV}) \geq 2.9 \times 10^{16} \text{ y}.$$

3.2.4 Double-β^- decay of ^{198}Pt

To set a limit on the double-β^- transition of ^{198}Pt to the 2^+_1 excited level of ^{198}Hg with the energy of 411.8 keV, the energy spectrum (see fig. 7b) was fitted in the energy interval (405–420) keV by a straight line (which represents the background model) and the expected peak at 411.8 keV [3]. The fit gives the area of the peak (13 ± 10) counts, which allows to exclude 29 counts at 90% C.L. Taking into account the detection efficiency ($\eta = 7.5\%$) we have obtained the following limit on the process searched for:

$$T_{1/2}^{2\beta^-(2\nu+0\nu)}(\text{g.s.} \to 411.8 \text{ keV}) \geq 3.5 \times 10^{18} \text{ y}.$$

4 Conclusions

The measurements performed over 1815 h with a 42.5 g sample of platinum with the help of an ultra-low background HP Ge γ spectrometer (468 cm^3) were used to set limits on double-β processes in ^{190}Pt in the range of $T_{1/2} \sim 10^{14-16}$ y, which is 3–4 orders of magnitude higher than the limit on $0\nu\varepsilon\beta^+$ (g.s. $\to$ g.s.) decay obtained in [10,11] by a re-analysis of the early experiment [35].

The search for the possible resonant $0\nu2\varepsilon$ capture to the 1382.4 keV level was realized for the first time. For future applications, taking into account the strong dependence of the resonant process on the difference in atomic masses of ^{190}Pt and ^{190}Os, precise measurements of the atomic masses are required. Moreover, it would also be useful to precisely study the characteristics of the 1382.4 keV level of ^{190}Os (spin, parity, decay scheme).

Search for 2β transition of ^{198}Pt to the 411.8 keV excited level of ^{198}Hg was carried out at the first time giving the limit $T_{1/2} > 3.5 \times 10^{18}$ y.

The measurements allowed us to estimate the radioactive contamination of the used platinum sample. In particular, we have detected 7 mBq/kg of ^{137}Cs and 40 mBq/kg of ^{192}Ir in this platinum sample. The contamination of ^{60}Co, ^{226}Ra, ^{228}Ra and ^{228}Th does not exceed the level of a few mBq/kg, while the activities of ^{40}K, ^{235}U, ^{238}U

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[3] There is a peculiarity in the data at the energy 413.4 ± 0.3 keV with the area of 15 ± 10 counts. However it cannot be ascribed to the double-beta decay of ^{198}Pt to the first-excited level of ^{198}Hg. Moreover no intensive gamma quanta of such an energy are expected to be emitted in decays of U/Th daughters, ^{192}Ir or ^{192m}Ir. Therefore we assume the peculiarity is due to statistical fluctuation, which allow us to use a line as a background model.

are less than a few tens mBq/kg (we assume a broken equilibrium in U/Th chains).

To improve the sensitivity of the experiment we are going to increase the mass of the platinum by one order of magnitude. Further improvements can be achieved by increasing the detection efficiency and the exposition, and obviously by using enriched ^{190}Pt isotope. Specially developed multi-crystal HP Ge detectors could also be applied to reach a sensitivity to double-β processes in ^{190}Pt on the level of 10^{22-24} y. This would be particularly interesting also because of the possibility of a resonant double-electron capture in ^{190}Pt having such isotope the largest Z value (the nuclear parameter which greatly favors the process) among the nuclei where a resonant double-electron capture could occur.

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