First observation of α decay of ^{190}Pt to the first excited level ($E_{\rm exc} = 137.2$ keV) of ^{186}Os

P. Belli,[1] R. Bernabei,[1,2,*] F. Cappella,[3,4] R. Cerulli,[5] F. A. Danevich,[6] A. Incicchitti,[3] M. Laubenstein,[5] S. S. Nagorny,[6] S. Nisi,[5] O. G. Polischuk,[6] and V. I. Tretyak[6]

[1]*INFN, Sezione di Roma Tor Vergata, I-00133 Rome, Italy*
[2]*Dipartimento di Fisica, Università di Roma Tor Vergata, I-00133 Rome, Italy*
[3]*INFN, Sezione di Roma La Sapienza, I-00185 Rome, Italy*
[4]*Dipartimento di Fisica, Università di Roma La Sapienza, I-00185 Rome, Italy*
[5]*INFN, Laboratori Nazionali del Gran Sasso, 67010 Assergi (AQ), Italy*
[6]*Institute for Nuclear Research, MSP 03680 Kyiv, Ukraine*

(Received 24 December 2010; published 9 March 2011)

The α decays of naturally occurring platinum isotopes, which are accompanied by the emission of γ quanta, have been searched for deep underground (3600 m water equivalent) at the Gran Sasso National Laboratories of the INFN (Italy). A sample of Pt with a mass of 42.5 g and a natural isotopic composition has been measured with a low background HP Ge detector (468 cm^3) during 1815 h. The α decay of ^{190}Pt to the first excited level of ^{186}Os ($J^\pi = 2^+$, $E_{\rm exc} = 137.2$ keV) has been observed for the first time, with the half-life determined as $T_{1/2} = 2.6^{+0.4}_{-0.3}({\rm stat.}) \pm 0.6({\rm syst.}) \times 10^{14}$ yr. The $T_{1/2}$ limits for the α decays of other Pt isotopes have been determined at the level of $T_{1/2} \simeq 10^{16}$–10^{20} yr. These limits have been set for the first time or they are better than those known from earlier experiments.

DOI: 10.1103/PhysRevC.83.034603 PACS number(s): 23.60.+e, 27.80.+w, 29.30.Kv

I. INTRODUCTION

The phenomenon of α decay has been known for more than 100 years [1] but the interest in this process is still great; in fact, it has even increased during the last decade, from both the theoretical and the experimental sides. Almost 30 theoretical papers can be found in the literature published from 2009 through 2010. Many of them were devoted to new semiempirical formulas for half-lives (see, for example, Ref. [2]) that successfully describe the accumulated to date experimental $T^\alpha_{1/2}$ values and allow one to understand in a more clear way the perspectives of new investigations, in particular, in studies of exotic very short-living isotopes close to the proton drip line [3] or decays of superheavy elements [4]. In the investigation of long-living rare nuclear decays, the field of interest of the authors of this article, the improvements in the experimental techniques have led to the enhancement of sensitivity and to the discovery of new α decays that were never observed previously because of the extra-long half-lives of the decaying nuclides: $T^\alpha_{1/2} = 1.9 \times 10^{19}$ yr was found for ^{209}Bi, which was considered before as the heaviest stable nuclide [5]; half-lives in the range of $T^\alpha_{1/2} = (1.0$–$1.8) \times 10^{18}$ yr were measured for ^{180}W in Refs. [6–9]; and the α decay of ^{151}Eu was also recently observed with $T^\alpha_{1/2} = 5.0 \times 10^{18}$ yr [10]. ^{209}Bi and ^{180}W are the keepers of two current world records: ^{209}Bi for the longest α half-life measured so far, and ^{180}W for the lowest observed specific α activity of only 2.3 disintegrations per year per gram of W of natural composition, which is much lower than that for ^{209}Bi (105 disintegrations per year per gram of Bi), due to the low natural abundance of ^{180}W ($\delta = 0.12\%$ compared to $\delta = 100\%$ for ^{209}Bi [11]).

All the six naturally occurring isotopes of platinum are potentially unstable in relation to α decay (see Table I). However, only for one of them, ^{190}Pt (with the biggest energy release of $Q_\alpha = 3251(6)$ keV), has this process been experimentally observed to date, with the first successful measurement in 1921 [12]. In that and in subsequent works the half-life of the ^{190}Pt was determined in the range of $(3.2$–$10) \times 10^{11}$ yr (see review, Ref. [13], and references therein); the currently recommended half-life value is: $T^\alpha_{1/2} = (6.5 \pm 0.3) \times 10^{11}$ yr [14]. In all the previous works, the ^{190}Pt α decay was observed only to the ground state (g.s.) of ^{186}Os. However, the first excited level of the daughter nuclide ^{186}Os ($J^\pi = 2^+$) has a quite low energy: $E_{\rm exc} = 137.2$ keV [15], and the energy available to the α particle in the decay to this level, $Q^*_\alpha = 3114(6)$ keV, is not much lower than that in the g.s. to g.s. transition. Our theoretical estimates of the corresponding half-life (see details in Sec. IV) gave values in the range of $T^\alpha_{1/2} = 10^{13}$–10^{14} yr. This allowed us to hope to discover the $^{190}{\rm Pt} \to {}^{186}{\rm Os}(2^+_1)$ decay through the observation of the 137.2 keV γ quantum emitted in the deexcitation of the ^{186}Os* nucleus with a well-shielded low-background HP Ge detector even using a Pt sample with natural isotopic composition with very low percentage of ^{190}Pt ($\delta = 0.014\%$).

The present article describes the first observation of the α decay $^{190}{\rm Pt} \to {}^{186}{\rm Os}(2^+_1)$ based on the measurements with a low-background HP Ge detector operated in the underground conditions of the Gran Sasso National Laboratories (LNGS) of the INFN (Italy). In addition to the ^{190}Pt decay, the γ quanta can be emitted in the α decays of other Pt isotopes: (1) when the excited levels of the daughter Os nuclei are populated, and (2) when the daughter Os nuclei are unstable and further decay with emission of some γ rays, as in the case of ^{195}Pt and ^{198}Pt. We also determine $T_{1/2}$ limits for such decays (to our knowledge, for the first time).

*rita.bernabei@roma2.infn.it

 ©2011 American Physical Society

TABLE I. Natural abundances (δ) of Pt isotopes and energy releases (Q_α) expected in the α decays of Pt to Os. N_i is the number of nuclei of the specific Pt isotope per 1 g of natural Pt.

Parent isotope	δ (%) [11]	Q_α (keV) [16]	N_i/1 g
^{190}Pt	0.014(1)	3251(6)	4.32×10^{17}
^{192}Pt	0.782(7)	2418.6(2.2)	2.41×10^{19}
^{194}Pt	32.767(99)	1518.3(1.6)	1.01×10^{21}
^{195}Pt	33.832(10)	1172.0(1.6)	1.04×10^{21}
^{196}Pt	25.242(41)	808.1(2.6)	7.79×10^{20}
^{198}Pt	7.163(55)	100(4)	2.21×10^{20}

II. MEASUREMENTS

The measurements were performed deep underground at LNGS; the average overburden is about 3600 m of water equivalent. Two platinum crucibles, exploited in the LNGS chemical laboratory, were used as the Pt sample with a total mass of 42.53 g. The bigger Pt cup [see Fig. 1(a)] has a maximal diameter of near 6.4 cm, a height of 2.8 cm, and a thickness of around 0.21 mm. Approximate sizes of the smaller Pt crucible are 3.0 cm (maximal diameter), 3.2 cm (height), and 0.25 mm (thickness).

The data with the Pt sample were collected with the HP Ge detector (GeCris, 468 cm^3; see Fig. 1(b) for simplified scheme of the measurements) over 1815.4 h. The Pt sample in a thin polyethylene bag was placed directly on the lid of the detector's cryostat. The background spectrum of the detector was measured during 1045.6 h. The energy resolution of the detector is full width at half maximum (FWHM) = 2.0 keV for the 1332-keV γ line of ^{60}Co. To reduce the external background, the detector was shielded by layers of low-radioactive copper ($\simeq$10 cm) and lead ($\simeq$20 cm). The setup has been continuously flushed by high-purity boil-off nitrogen to avoid the presence of residual environmental radon. Part of the spectrum accumulated with the Pt sample in comparison with the background in the energy range of 100–700 keV is shown in Fig. 2.

The comparison of the Pt and of the background spectra shows that the platinum sample practically is not contaminated by "usual" radioactive contaminants: U/Th series, ^{40}K, ^{60}Co,

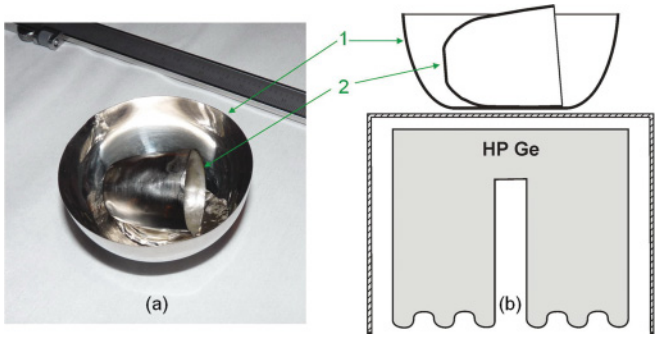


FIG. 1. (Color online) (a) Photo of the Pt sample and (b) simplified scheme of the measurements with the HP Ge detector. The bigger Pt cup and the smaller Pt crucible are labeled as 1 and 2, respectively.

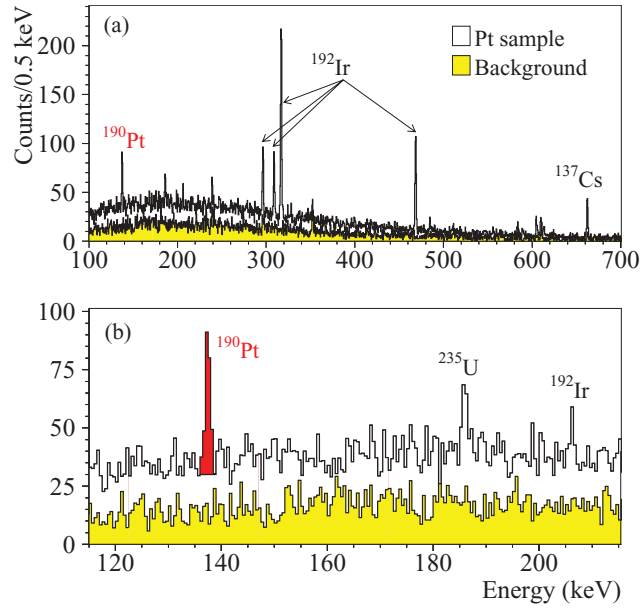


FIG. 2. (Color online) Energy spectrum of the Pt sample with a mass of 42.5 g measured during 1815 h in the 100–700 keV energy interval (a) and in more detail around the 137-keV region (b). The background spectrum (measured during 1046 h but normalized here to 1815 h) is also shown (filled histogram). A peak at 137 keV after α decay of ^{190}Pt $\rightarrow$ ^{186}Os(2_1^+) is clearly visible in the Pt spectrum but is absent in the background.

(for which mostly only limits are determined at the level of $\simeq$10 mBq/kg; see details in a companion paper on the search for double β processes in Pt [17]). However, the presence of the radioactive ^{192}Ir is evident; the corresponding activity is equal to 49 ± 3 mBq/kg. ^{192}Ir could appear in platinum as cosmogenic activation of Pt by cosmic rays at the Earth's surface. In fact, because iridium usually accompanies platinum in nature, ^{192}Ir can be created as a result of neutron capture by ^{191}Ir, which is one of two naturally occurring iridium isotopes ($\delta = 37.3\%$, cross section for thermal neutrons is $\sigma =$ 954 b [15]). However, while the half-life of the ground state of ^{192}Ir is $T_{1/2} = 73.8$ d [15], and the exponential decrease in time of the ^{192}Ir activity should be observed during our 75.6 d measurements, in fact, the rate of the corresponding peaks was consistent with constant. Thus, the ^{192}Ir activity should be ascribed not to the ground state but to the isomeric ^{192m}Ir level with $E_{\rm exc} = 168.1$ keV and $T_{1/2} = 241$ yr [18]. This isomeric state decays to the ground state of ^{192}Ir emitting 155.1- and 13.0-keV γ rays. The transitions, however, are strongly converted to electrons (coefficients of conversion are equal $\alpha_{13} = 57\,000$ and $\alpha_{155} = 1026$ [18]). This explains the absence of the 155.1-keV peak in our data (Fig. 2).

III. RESULTS

A. α decay of ^{190}Pt

In the spectrum collected with the Pt sample, the peak at energy 137.1 ± 0.1 keV is observed (see Fig. 2) while it is absent in the background spectrum. The fit of the Pt spectrum in the

energy region 120–170 keV with a Gaussian peak and linear background assumption gives a net area of 132 ± 17 counts, inconsistent with zero at about 8σ. Variations of the energy interval for the fit result in changes of the area inside the quoted uncertainty.

The 137.1-keV peak can be explained with the α decay of ^{190}Pt to the first excited level of ^{186}Os whose excitation energy is 137.159 ± 0.008 keV [14]. If populated, this level deexcites with the emission of a γ quantum with energy $E_\gamma = 137.157 \pm 0.008$ keV, which is in nice agreement with that of the observed peak, 137.1 ± 0.1 keV. The process of α decay of $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}^*$ was never observed previously.

Using the area of the 137-keV peak, the corresponding partial half-life for the transition to the first excited level of ^{186}Os can be calculated as

$$T_{1/2}[^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+, 137.2\,\text{keV})] = \frac{\ln 2 N_{190} \varepsilon t}{S(1+\alpha)}, \quad (1)$$

where $N_{190} = 1.84 \times 10^{19}$ is the number of the ^{190}Pt nuclei in the 42.53-g Pt sample, ε is the efficiency to detect the full energy γ with the HP Ge detector, $t = 1815.4$ h is the measurement time, $S = 132 \pm 17$ counts is the area of the peak, and α is the coefficient of conversion to electrons for the given nuclear transition.

The full peak efficiency at 137 keV was calculated with the EGS4 simulation package [19] and, for crosscheck, also with the GEANT4 [20] simulation package. The Pt crucibles have nontrivial shapes, and models used in each code (programmed independently) reproduced the real shape with some approximations. The calculations gave for the 137-keV peak values of $\varepsilon = 3.4\%$ with EGS4 and $\varepsilon = 2.6\%$ with GEANT4. For the final result, we use the average between these two values $\varepsilon = 3.0\%$, including the difference in systematic uncertainties.

Taking into account the electron conversion coefficient for the transition $\alpha = 1.29$ [14], the $T_{1/2}$ value is equal to $T_{1/2}[^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+, 137.2\ \text{keV})] = 2.6^{+0.4}_{-0.3}(\text{stat.}) \times 10^{14}$ yr.

Only the statistical uncertainty in the peak area was taken into account here. The systematic uncertainties are related with the uncertainty of the mass of the Pt sample (0.02%), of the calculation of the measurements' live time (0.01%), and of the knowledge on natural isotopic abundance of ^{190}Pt (7.1%), with the biggest contribution from the calculation of the efficiency. To estimate the latter uncertainty, we performed a comparison of the calculated and measured efficiencies for a voluminous ($\oslash 7.0 \times 1.1$ cm) water source in which, among several radioactive isotopes, also ^{57}Co was dissolved. This source was calibrated and distributed by the International Atomic Energy Agency (IAEA) within an open world-wide proficiency test. The radioactive ^{57}Co, in particular, emits γ quanta with an energy of 136.5 keV, very close to the peak under investigation. Disagreement between the experimental and the calculated efficiencies was 6% (0%) with EGS4 (GEANT4). Taking into account the previous experience of measurements with the used GeCris HP Ge detector and the more complicated geometry of the Pt sample in comparison with the simple cylindrical shape of the IAEA water source, the systematic uncertainty in the efficiency can be conservatively estimated as 20%.

Summing all the uncertainties in squares, we obtain the following value for the half-life of α decay of $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+)$ as the final value:

$$T_{1/2}[^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+, 137.2\,\text{keV})] = 2.6^{+0.4}_{-0.3}(\text{stat.}) \pm 0.6(\text{syst.}) \times 10^{14}\ \text{yr}. \quad (2)$$

It should be noted that ^{192}Ir, present in the Pt sample, also emits γ rays with an energy of 136.3 keV, however, with a very low yield of $I = 0.183\%$ [15]. Taking into account that the area of the most intensive ($I = 82.80\%$) peak of ^{192}Ir at 316.5 keV is (619 ± 32) counts (and considering also different efficiencies of 2.3% at 137 keV[1] and 5.5% at 316 keV), the contribution of ^{192}Ir to the 137-keV peak is 0.6 counts; this does not change the $T_{1/2}$ value presented in Eq. (2).

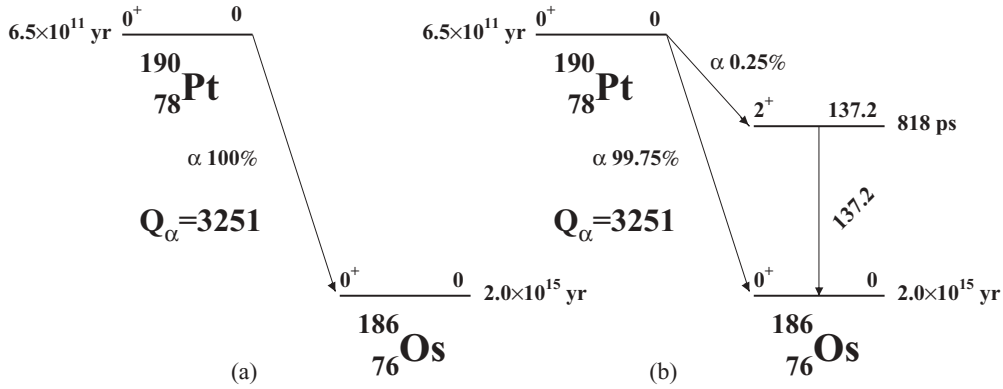
Gamma rays with energies close to 137 keV are emitted in some other nuclear processes (see, for example, Ref. [21]), and this could give an alternative explanation of the peak observed in the Pt experimental spectrum; however, usually additional γ rays are also emitted in such decays, and their absence allows us to exclude these effects. For example, ^{181}Hf ($T_{1/2} = 42.4$ d [15]), which could be created as a result of cosmogenic activation of Pt, emits 136.3-keV ($I = 5.85\%$) and 136.9-keV ($I = 0.86\%$) γ quanta but also 133.0-keV γ rays should be emitted with much higher yields of $I = 43.3\%$, which, however, are absent in the experimental data (see Fig. 2). Another example could be the 136.6-keV ($I = 0.012\%$) γ rays from ^{235}U but the peak at 143.8 keV ($I = 10.96\%$) is absent. The contributions from the nuclides of the U/Th natural radioactive chains: ^{214}Pb ($E_\gamma = 137.5$ keV, $I < 0.006\%$) and ^{228}Ac ($E_\gamma = 137.9$ keV, $I = 0.024\%$) are calculated as negligible using the data on the U/Th pollution of the Pt sample.

^{181}W ($T_{1/2} = 121.2$ d), another possible cosmogenic contaminant of Pt, cannot be estimated in the above-described way because its 136.3-keV γ rays are the most intensive with a yield of $I = 0.0311\%$. We calculated the ^{181}W-induced activity in Pt with the COSMO code [22]; the result was 1.5 decays per day per kg of $^{\text{nat}}$Pt. Taking into account the time of measurements (75.6 d), the small mass of our Pt sample (42.5 g), and the low yield of these γ rays, the contribution from ^{181}W to the 137-keV peak is estimated also as negligible (5×10^{-5} counts).

In conclusion, analyzing other possible sources of the 137-keV line, we did not find any real alternative that could mimic α decay of $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+)$. The old scheme of the ^{190}Pt α decay [14,15] and the updated scheme that follows from our observation of the $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+)$ transition is presented in Fig. 3.

In the process of ^{190}Pt α decay other excited levels of ^{186}Os could be populated too. Because the probability of the α decay exponentially decreases with the decrease of the energy, we consider here possible transition only to the next excited level of ^{186}Os ($J^\pi = 4^+$) with $E_{\text{exc}} = 434.1$ keV. If this

[1]The efficiency for the 137-keV peak of ^{192}Ir is lower than that for the single 137-keV γ quantum because of summing effects for ^{192}Ir γ rays emitted in cascade.



.

FIG. 3. Old (a) and new (b) schemes of α decay of ^{190}Pt. The energies of the levels and the deexcitation γ quantum are given in keV [14,15].

level is populated, two γ quanta are emitted in its deexcitation with energies of $E_{\gamma 1} = 296.9$ keV and $E_{\gamma 2} = 137.2$ keV. Both peaks are present in our data, and while we explain the 137-keV peak by the $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+)$ α decay, the 296-keV peak is related with ^{192}Ir. We can estimate the contribution to the 296-keV peak from ^{192}Ir using its nearby peak at 316.5 keV. Taking into account that the area of the 316-keV peak is 619 ± 32 counts, and the yields $I_{316} = 82.80\%$ and $I_{296} = 28.67\%$ [15], we could expect 193 ± 10 counts from ^{192}Ir. At the same time, in the real Pt spectrum the area of the 296-keV peak is $S = 200 \pm 23$ counts. The difference between these two values, 7 ± 25 counts, is consistent with 0, and, in accordance with the Feldman-Cousins procedure [23], results in the limit $S < 48$ counts at 90% C.L. Deriving (here and in the following) the $T_{1/2}$ limits for the Pt α decays, to be more conservative, we will use for the efficiencies the values obtained with the code giving systematically lower efficiency. Substituting in Eq. (1) the value of the electron conversion coefficient $\alpha = 0.095$ [14] and the efficiency $\varepsilon = 7.1\%$ for 296-keV γ quanta, we obtain the following half-life limit:

$$T_{1/2}[^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(4_1^+, 434.1\,\text{keV})] > 3.6 \times 10^{15}\ \text{yr at 90\% C.L.}$$

B. $T_{1/2}$ limits on α decays of other Pt isotopes

The data collected with the Pt sample also allow us to search for α decays of other Pt isotopes related with the emission of γ quanta. We consider here only transitions to the lowest excited levels of the daughter Os isotopes as the most probable. In the case of ^{195}Pt and ^{198}Pt, the daughter Os nuclei are unstable, and the search for γ quanta related with their decays gives the possibility to look for the g.s. to g.s. transitions of Pt to Os. In general, we do not see any of such decays, and only limits on the corresponding half-lives are determined.

^{192}Pt. The best half-life limit on α decay of ^{192}Pt was obtained in Ref. [24] searching for its specific activity: $T_{1/2} > 6.0 \times 10^{16}$ yr. This value could be considered as valid not only for the g.s. to g.s. transition but also for the decay to the lowest excited levels of ^{188}Os.

The population of the first excited level of ^{188}Os ($J^\pi = 2^+$, $E_{\text{exc}} = 155.0$ keV) will lead to the emission of γ quantum and to a peak at energy of 155.0 keV. The latter, however, is absent in the Pt spectrum (see Fig. 2), and we can give only a limit on its area. The value of $\lim S$ was calculated fitting the experimental Pt spectrum by the sum of a linear function (representing the near linear background) and a gaussian (representing the expected effect) with the center at 155.0 keV and a proper FWHM close to that of the nearby 137-keV peak. This procedure gave the value $S = -8 \pm 17$ counts for the area of the 155-keV peak that results in the limit [23] $\lim S = 21$ counts at 90% C.L. Using the number of ^{192}Pt nuclei, $N_{192} = 1.02 \times 10^{21}$, the electron conversion coefficient $\alpha = 0.82$ [25], and the calculated efficiency $\varepsilon = 3.5\%$, one gets with Eq. (1) the following:

$$T_{1/2}[^{192}\text{Pt} \rightarrow {}^{188}\text{Os}(2_1^+, 155.0\,\text{keV})] > 1.3 \times 10^{17}\ \text{yr at 90\% C.L.}$$

The α decay of ^{192}Pt to the next excited level of ^{188}Os ($J^\pi = 4^+$, $E_{\text{exc}} = 477.9$ keV) will lead to the emission of two deexcitation γ quanta with energies of 322.9 and 155.0 keV. The peak at the energy of 322.9 keV is absent in the data: its area found in a procedure similar to the one described above gave $S = 12 \pm 16$ counts, which results in $\lim S = 38$ counts at 90% C.L. The electron conversion coefficient for the 322.9-keV transition is equal $\alpha = 0.074$ [25], and efficiency is $\varepsilon = 7.2\%$. With these values we obtain

$$T_{1/2}[^{192}\text{Pt} \rightarrow {}^{188}\text{Os}(4_1^+, 477.9\,\text{keV})] > 2.6 \times 10^{17}\ \text{yr at 90\% C.L.}$$

^{194}Pt. We give below limits for possible ^{194}Pt α decays to the two lowest excited levels of ^{190}Os: $J^\pi = 2^+$, $E_{\text{exc}} = 186.7$ keV and $J^\pi = 4^+$, $E_{\text{exc}} = 547.9$ keV. Their population results in the emission of one γ quantum with $E_\gamma = 186.7$ keV or two γ rays with $E_{\gamma 1} = 186.7$ keV and $E_{\gamma 2} = 361.1$ keV, respectively.

There is a peak at energy 185.6 ± 0.1 keV in the experimental Pt spectrum with area $S = 79 \pm 16$ counts; most probably it belongs to ^{235}U which emits γ rays of 185.7 keV with a yield of 57.20%. It is difficult to estimate the contribution of

^{235}U to the 186-keV peak using other γ lines in the ^{235}U chain because they have lower yields. Thus, to derive a limit for α decay of $^{194}\text{Pt} \rightarrow {}^{190}\text{Os}(2_1^+)$, we very conservatively ascribe all the peak area to the process of α decay; taking also into account the uncertainty in the S value, we obtain $\lim S = 105$ counts at 90% C.L. Substituting in Eq. (1) the number of ^{194}Pt nuclei, $N_{194} = 4.30 \times 10^{22}$, the efficiency $\varepsilon = 4.9\%$, and the conversion coefficient $\alpha = 0.425$ [26], one finally obtains

$$T_{1/2}[^{194}\text{Pt} \rightarrow {}^{190}\text{Os}(2_1^+, 186.7\,\text{keV})] > 2.0 \times 10^{18}\ \text{yr at 90\% C.L.}$$

The peak at the energy of 361.1 keV, expected for $^{194}\text{Pt} \rightarrow {}^{190}\text{Os}(4_1^+)$ decay, is absent, and using the values $S = -9.2 \pm 8.3$ counts (thus $\lim S = 6.2$ counts), $\varepsilon = 7.2\%$, and $\alpha = 0.054$ [26], one gets

$$T_{1/2}[^{194}\text{Pt} \rightarrow {}^{190}\text{Os}(4_1^+, 547.9\,\text{keV})] > 6.8 \times 10^{19}\ \text{yr at 90\% C.L.}$$

^{195}Pt. The daughter nucleus ^{191}Os is unstable ($T_{1/2} = 15.4$ d [15]) and β^- decays to ^{191}Ir. This allows us to set the $T_{1/2}$ limit for ^{195}Pt α decay not only to some specific excited level of ^{191}Os but to all the ^{191}Os states (including g.s.). In the ^{191}Ir decay, a γ quantum with an energy of 129.4 keV and a yield of $I = 29.0\%$ is emitted [27]. A peak at this energy is absent in the Pt spectrum (see Fig. 2), its area $S = -15.7 \pm 10.9$ counts gives $\lim S = 6.4$ counts. Taking into account the number of the ^{195}Pt nuclei, $N_{195} = 4.42 \times 10^{22}$, and the efficiency $\varepsilon = 2.2\%$, we obtain

$$T_{1/2}[^{195}\text{Pt} \rightarrow {}^{191}\text{Os(all states)}] > 6.3 \times 10^{18}\ \text{yr at 90\% C.L.}$$

^{196}Pt. We give here the $T_{1/2}$ limit only for the decay to the lowest excited level of ^{192}Os ($J^\pi = 2^+$, $E_{\text{exc}} = 205.8$ keV). It should be noted that in the experimental Pt spectrum we have a peak at an energy of 205.9 ± 0.1 keV and an area of $S = 36 \pm 12$ counts. However, it is related with the contamination of the Pt sample by ^{192}Ir which emits γ rays of 205.8 keV with yield $I_{206} = 3.30\%$ [15]. We can estimate the contribution of ^{192}Ir to the 206-keV peak on the basis of another peak of ^{192}Ir at 316.5 keV ($I_{316} = 82.80\%$). Taking into account that $S_{316} = 619 \pm 32$ counts and the efficiencies $\varepsilon_{316} = 5.5\%$ and $\varepsilon_{206} = 4.3\%$, we can expect 19 ± 1 counts in the 206-keV peak from ^{192}Ir. The contribution from ^{235}U, which emits γ rays of 205.3 keV with $I = 5.01\%$, can be estimated from its 185.7-keV peak ($I = 57.20\%$) as 8 ± 2 counts. The difference in areas between the line observed in the experimental spectrum and the contributions from ^{192}Ir and ^{235}U is equal to 9 ± 12 counts, giving no evidence for the effect. With the number of ^{196}Pt nuclei, $N_{196} = 3.31 \times 10^{22}$, $\lim S = 29$ counts, the efficiency $\varepsilon = 5.5\%$, and the electron conversion coefficient $\alpha = 0.305$ [18], we obtain

$$T_{1/2}[^{196}\text{Pt} \rightarrow {}^{192}\text{Os}(2_1^+, 205.8\,\text{keV})] > 6.9 \times 10^{18}\ \text{yr at 90\% C.L.}$$

^{198}Pt. The daughter ^{194}Os nucleus is unstable ($T_{1/2} = 6.0$ yr [28]) and decays to ^{194}Ir, which is unstable too. In the decay chain ^{194}Os (6.0 yr, $Q_\beta = 96.9$ keV) $\rightarrow$ ^{194}Ir (19.28 h, $Q_\beta = 2233.8$ keV) $\rightarrow$ ^{194}Pt (stable) the most prominent γ line is at 328.5 keV with a yield of $I = 13.1\%$. A peak at this energy is absent in the Pt spectrum: $S = (12 \pm 9)$ counts and $\lim S = 27$ counts. Using the number of ^{198}Pt nuclei,

TABLE II. Summary of $T_{1/2}$ results. Limits correspond to 90% C.L. Theoretical half-lives are calculated in accordance with the findings of Refs. [29] and [30] taking into account additional hindrance factors. For ^{195}Pt and ^{198}Pt, the experimental $T_{1/2}$ limits are valid for the α decays to all the states of the daughter Os nuclei while the theoretical estimations are given for the g.s. to g.s. transitions.

α transition	Level of daughter nucleus	Experimental $T_{1/2}$ (yr) Previous	This work	Theoretical estimation
$^{190}\text{Pt} \rightarrow {}^{186}\text{Os}$	2^+, 137.2 keV	–	$=2.6 \pm 0.7 \times 10^{14}$	3.2×10^{13} [29] 7.0×10^{13} [30]
	4^+, 434.1 keV	–	$>3.6 \times 10^{15}$	7.4×10^{17} [29] 2.0×10^{18} [30]
$^{192}\text{Pt} \rightarrow {}^{188}\text{Os}$	2^+, 155.0 keV	$>6.0 \times 10^{16}$ [24]	$>1.3 \times 10^{17}$	9.1×10^{25} [29] 4.6×10^{26} [30]
	4^+, 477.9 keV	$>6.0 \times 10^{16}$ [24]	$>2.6 \times 10^{17}$	2.7×10^{33} [29] 1.9×10^{34} [30]
$^{194}\text{Pt} \rightarrow {}^{190}\text{Os}$	2^+, 186.7 keV	–	$>2.0 \times 10^{18}$	7.0×10^{51} [29] 1.9×10^{53} [30]
	4^+, 547.9 keV	–	$>6.8 \times 10^{19}$	4.3×10^{71} [29] 3.5×10^{73} [30]
$^{195}\text{Pt} \rightarrow {}^{191}\text{Os}$	All states	–	$>6.3 \times 10^{18}$	7.5×10^{59} [29] 3.2×10^{69} [30]
$^{196}\text{Pt} \rightarrow {}^{192}\text{Os}$	2^+, 205.8 keV	–	$>6.9 \times 10^{18}$	1.7×10^{106} [29] 2.1×10^{109} [30]
$^{198}\text{Pt} \rightarrow {}^{194}\text{Os}$	All states	–	$>4.7 \times 10^{17}$	1.6×10^{348} [29] 7.2×10^{358} [30]

$N_{198} = 9.40 \times 10^{21}$, and the efficiency $\varepsilon = 7.2\%$, one gets

$T_{1/2}[^{198}\text{Pt} \rightarrow {}^{194}\text{Os}, \text{all states})] > 4.7 \times 10^{17}$ yr at 90% C.L.

As in the case of ^{195}Pt, this limit is valid for the α decay of ^{198}Pt to all the states of ^{194}Os including the g.s. to g.s. transition. A summary of all the obtained results is given in Table II.

IV. COMPARISON WITH THEORETICAL ESTIMATIONS

We calculated half-life values for the α decays of the different Pt isotopes with the cluster model of Ref. [29] and with semiempirical formulas [30] based on a liquid drop model and a description of α decay as a very asymmetric fission process. The approaches of Refs. [29,30] were tested with a set of experimental half-lives of almost 400 α emitters and demonstrated good agreement between calculated and experimental $T_{1/2}$ values, mainly inside the factor of 2–3. We also successfully used these works to predict $T_{1/2}$ values in our searches for α decays of ^{180}W [6] and ^{151}Eu [10], obtaining adequate agreement between the first experimentally measured and calculated results. As another example, the calculated $T_{1/2}$ values for the g.s. to g.s. α decay of ^{190}Pt are equal: 5.0×10^{11} yr in accordance with Ref. [29] and 10.3×10^{11} yr in accordance with Ref. [30], while the experimental values were measured in the range of $(3.2\text{–}10) \times 10^{11}$ yr [13] (the recommended value is $(6.5 \pm 0.3) \times 10^{11}$ yr [14]).

The ground state of all the Pt isotopes has spin and parity $J^\pi = 0^+$, except for ^{195}Pt with $J^\pi = 1/2^-$. We study here the transitions to the daughter levels with $J^\pi = 2^+$ and 4^+ ($9/2^-$ for $^{195}\text{Pt} \rightarrow {}^{191}$Os g.s.). Because of the difference between the spins of the initial and of the final nuclear states, the emitted α particle will have nonzero angular momentum: values of $L = 2$ or $L = 4$ are the most preferable among the allowed ones by the selection rules for the transitions with $\Delta J^{\Delta\pi} = 2^+$ or 4^+, respectively. Thus, in the $T_{1/2}$ estimation, we take into account the hindrance factors of HF = 1.9 for $L = 2$ and HF = 9.0 for $L = 4$, calculated in accordance with Ref. [31]. The results of the calculations are given in Table II. For the α decay of $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2^+, 137.2$ keV) observed in this work, the semiempirical approach of Ref. [30] gives a better agreement with the measured $T_{1/2}$ value.

From the theoretical results, it is clear that the observation of other Pt α decays presented in Table II is out of reach of current experimental possibilities, except probably for the decay of $^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(4^+, 434.1$ keV). The latter will need 3 orders of magnitude higher sensitivity which, however, could be possible by using enriched ^{190}Pt instead of natural Pt with $\delta(^{190}\text{Pt}) = 0.014\%$.

V. CONCLUSIONS

The α decays of naturally occurring platinum isotopes, which are accompanied by the emission of γ quanta, were searched for with the help of a low-background HP Ge detector at the Gran Sasso National Laboratories of the INFN. The α decay of ^{190}Pt to the first excited level of ^{186}Os ($J^\pi = 2^+$, $E_{\text{exc}} = 137.2$ keV) was detected for the first time by the observation of the γ line at energy (137.1 ± 0.1) keV at the 8σ significance level while it is absent in the background spectrum of the detector. We did not find alternative processes that could mimic this effect. The half-life is determined as $T_{1/2}[^{190}\text{Pt} \rightarrow {}^{186}\text{Os}(2_1^+, 137.2\,\text{keV})] = [2.6^{+0.4}_{-0.3}(\text{stat.}) \pm 0.6(\text{syst.})] \times 10^{14}$ yr. This value is in relevant agreement with theoretical calculations based on the liquid drop model and the description of the α decay as a very asymmetric fission process [30].

In addition, $T_{1/2}$ limits for α decays with population of the lowest excited levels of Os nuclei (for ^{192}Pt, ^{194}Pt, ^{196}Pt) and for transition to the ground states of Os nuclei (for ^{195}Pt, ^{198}Pt) were determined at the level of $T_{1/2} \simeq 10^{16}$–10^{20} yr. These limits were set for the first time or they are better than those known from earlier experiments.

The α decay of ^{190}Pt to the second excited level of ^{186}Os ($J^\pi = 4^+$, $E_{\text{exc}} = 434.1$ keV) could be detected in a higher sensitivity experiment with platinum enriched in ^{190}Pt to $\simeq$10%. However, because its natural abundance is extremely low ($\delta = 0.014\%$), this is a very expensive task with the current technologies.

ACKNOWLEDGMENTS

The authors thank V. Yu. Denisov for interesting discussions and an anonymous referee for useful suggestions. The group from the Institute for Nuclear Research (Kyiv, Ukraine) was supported in part through the Project "Kosmomikrofizyka-2" (Astroparticle Physics) of the National Academy of Sciences of Ukraine.

[1] E. Rutherford, Philos. Mag. (Ser. 5) **47**, 109 (1899).
[2] V. Yu. Denisov and A. A. Khudenko, Phys. Rev. C **79**, 054614 (2009); **82**, 059901(E) (2010).
[3] C. J. Gross, W. Nazarewicz, and K. P. Rykaczewski (Eds.), Proceedings of the 4th International Conference on Exotic Nuclei and Atomic Masses, Eur. Phys. J. A 25, Suppl. 1 (2005).
[4] S. Hofmann, Lect. Notes Phys. **764**, 203 (2009).
[5] P. de Marcillac *et al.*, Nature (London) **422**, 876 (2003).
[6] F. A. Danevich *et al.*, Phys. Rev. C **67**, 014310 (2003).
[7] C. Cozzini *et al.*, Phys. Rev. C **70**, 064606 (2004).
[8] Yu. G. Zdesenko *et al.*, Nucl. Instrum. Methods A **538**, 657 (2005).
[9] P. Belli *et al.*, Nucl. Instrum. Methods A **626&627**, 31 (2011).
[10] P. Belli *et al.*, Nucl. Phys. A **789**, 15 (2007).
[11] J. K. Bohlke *et al.*, J. Phys. Chem. Ref. Data **34**, 57 (2005).
[12] G. Hoffman, Z. Phys. **7**, 254 (1921).

[13] O. P. Tavares, M. L. Terranova, and E. L. Medeiros, Nucl. Instrum. Methods B **243**, 256 (2006).
[14] C. M. Baglin, Nucl. Data Sheets **99**, 1 (2003).
[15] R. B. Firestone *et al.*, *Table of Isotopes*, 8th ed. (Wiley, New York, 1996), and CD update (1998).
[16] G. Audi, A. H. Wapstra, and C. Thibault, Nucl. Phys. A **729**, 337 (2003).
[17] P. Belli *et al.* (submitted to Eur. Phys. J A).
[18] C. M. Baglin, Nucl. Data Sheets **84**, 717 (1998).
[19] W. R. Nelson *et al.*, SLAC-Report-265, Stanford, 1985.
[20] S. Agostinelli *et al.*, Nucl. Instrum. Methods A **506**, 250 (2003); J. Allison *et al.*, IEEE Trans. Nucl. Sci. **53**, 270 (2006).
[21] WWW Table of Radioactive Isotopes [http://nucleardata.nuclear.lu.se/nucleardata/toi/radSearch.asp].
[22] C. J. Martoff and P. D. Lewin, Comput. Phys. Commun. **72**, 96 (1992).
[23] G. J. Feldman and R. D. Cousins, Phys. Rev. D **57**, 3873 (1998).
[24] G. Kauw, Forschungsber. Landes Nordrhein-Westfalen (1966) (No. 1640).
[25] B. Singh, Nucl. Data Sheets **95**, 387 (2002).
[26] B. Singh, Nucl. Data Sheets **99**, 275 (2003).
[27] V. R. Vanin *et al.*, Nucl. Data Sheets **108**, 2393 (2007).
[28] B. Singh, Nucl. Data Sheets **107**, 1531 (2006).
[29] B. Buck, A. C. Merchant, and S. M. Perez, J. Phys. G **17**, 1223 (1991); **18**, 143 (1992).
[30] D. N. Poenaru and M. Ivascu, J. Physique **44**, 791 (1983).
[31] K. Heyde, *Basic Ideas and Concepts in Nuclear Physics*, 2nd ed. (IOP Publishing, Bristol, 1999).