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TO A POSSIBILITY OF THE ^{178m2}Hf ISOMER ALPHA-DECAY

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An experiment was performed to search for α decay of the ^{178m2}Hf isomer. This high-spin (16^+) metastable state has a halflife of 31 years and its electromagnetic decay is well-known, principally occurring by a completely converted 12,7-keV transition. Its total *α*-decay energy $Q_\alpha = 4{,}53$ MeV should correspond to a halflife on the order of days. However, the angular momentum of the isomer should generate a drastic retardation toward *α* decay due to the influence of the centrifugal barrier. This is indeed the case, as established by the present work in which a partial α-decay halflife of $2{,}5 \cdot 10^{10}$ y was measured for ^{178m2}Hf. *K* hindrance in *α* decay is also discussed.

1. Introduction

As is known, the α-emission branch in the decay of the ^{178m2}Hf has not yet been observed, but in principle the successful detection is just a problem of a high sensitivity of measurements. Experiments with low activity require typically sufficient amount of the material studied, the deep suppression of the background radiation, good efficiency of the detector and long exposition time in order to reach statistically reliable estimation of the specific radiation yield, despite its very low level.

The Q_α value of 4,53 MeV is estimated according to nuclear mass tables, Ref. [1], and it corresponds to the transition from isomeric state to the ground state of the daughter ^{174}Yb nucleus, i.e. to the transition from 16^+ to 0^+ levels. Therefore, the emitted α-particle should release full angular momentum of 16 units, and a probability of that should be strongly suppressed by the centrifugal barrier. Another hindrance factor may also exist due to the change of the K-quantum number ΔK = 16. The K-hindrances in α-decay were not yet well understood until now, because of a deficit of the experimental and theoretical works on such a problem.

There is some challenge to get experimental information on K-hindrance manifestation in nuclear α-decay after the measurements of the partial α-decay halflife for ^{178m2}Hf. The ΔK hindrance can be the same high as the retardation due to the spin difference ΔI, or smaller but comparable, or even almost negligible. Remind that K quantum number arises only for the axially symmetric nuclear shapes. But in α-decay of a well-deformed axial nucleus, the emission of α-particle leads to the broken on the way axial symmetry. The triaxial shapes should be passed inevitably, when α takes off the nucleus. Means, α-decay may be a process out of the selection rules by K-quantum number. Emission along the axis does not break the symmetry, but these events have too low phase volume.

If one neglects the ΔI and ΔK hindrances, the estimated halflife of ^{178m2}Hf should be as short as days. However, the forbiddenness due to the angular momentum change ΔI should be taken in the account in any case, and it influences the halflife $T_{½}$ increasing it by many orders of magnitude. Below we reduce our estimations of expected ^{178m2}Hf α-decay $T_{½}$ with the account of all possible branches populating the rotational band levels with spins from 0 to 14 in the daughter ^{174}Yb nucleus. The forbiddenness due to the ΔI centrifugal barrier was determined using the semiempirical systematics. The deduced $T_{½}$ value does not contain the ΔK hindrance factor. If the experimentally measured $T_{½}$ for ^{178m2}Hf appears to be longer than the estimated value, this may allow a conclusion on the role of K-hindrance in nuclear α-decay.

2. Halflife estimates

For medium Z elements, the α-decay mode was experimentally observed typically for short-lived neutron deficient isotopes. Existing data on the α-decay energies E_α are reduced in Fig. 1 as a function of neutron number for elements from Nd to Pb. Only experimentally measured E_α values were used according to Ref. [2] to plot Fig. 1, the partial α-decay lifetimes were also measured for them. For many other isotopes, E_α can be calculated basing on known masses, but halflife values are unknown yet. In Fig.1, the range of nuclei corresponds to the area restricted by magic numbers of closed nuclear shells: N > 82 and Z ≤ 82. The Hf isotopes are in the center of this range, far from magic numbers in N and Z coordinates. As a result, they are characterized by well-deformed, axially symmetric, elongated shape and among them is also the nucleus of interest: ^{178}Hf. One can see in Fig. 1 very regular E_α dependence on N and Z. The record-long chain of α-decaying isotopes was established for Pt with Z = 78 and N varying from 90 to 112.

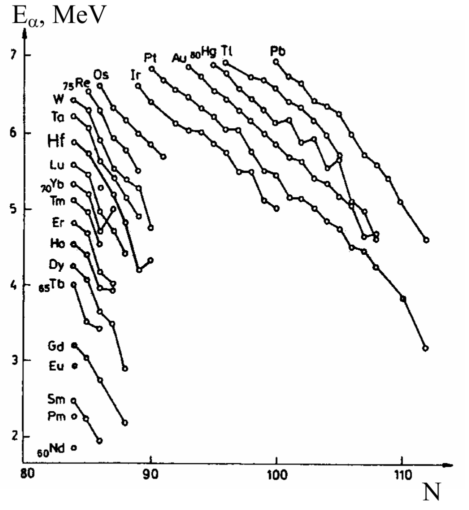

Fig. 1. Energies of α-particles emitted in the decay of Z = 60 to 82 nuclei. Experimental data are given according to Ref. [2].

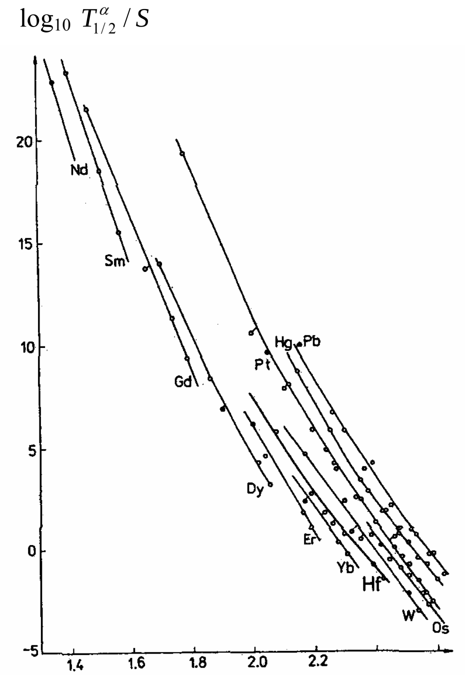

Fig. 2. The Geiger-Nuttall type systematics of the α-decay halflives in the range of Z = 60 to 82 nuclei.

Rich experimental data allows the empirical systematics, and the Geiger-Nuttall law was one of the first regularities found in nuclear physics by 90 years ago. A Geiger-Nuttall plot is shown in Fig. 2 for Z-even nuclei, one can see really almost linear dependence of log $T_{½}$ on the square root of Q_α. The slope of curves may be changing a little, but it is natural, when the exponential type function covers many order of magnitude, for instance ~ 10^{23} for Pt isotopes. The systematics of Fig. 2 can be very helpful for estimation of a halflife of some nuclide, if Q_α is known, but $T_{½}$ is not yet measured. We will use this for α-halflife estimation at the case of ^{178m2}Hf.

The Geiger-Nuttall plot is normally constructed for well allowed decay transitions; it does not contain significant structure or angular momentum hindrances. But in the decay of ^{178m2}Hf, the high-spin of initial state should produce very strong influence on the α-halflife, this is a major effect in the lifetime value of our interest. So that, in addition to Fig.2, we have to attract the data characterizing a centrifugal barrier contribution into α-decay halflife and to take into account the corresponding regular function. After that, one can be ready to produce some realistic estimations for the high-spin isomer decay.

Decay rate for α-transition from initial to final spin states I_i to I_f may be expressed as follows:

$$R = \frac{N_{at} \ln 2}{T^{\alpha}_{1/2}(Q_\alpha)} \sum_{m=-I_f}^{I_f} \frac{1}{F(|I_i - m|)}, \qquad (1)$$

where $T_{½}(Q_\alpha)$ corresponds to zero spin change and depends only on Q_α, like the functions shown in Fig. 2, while F(ΔI) is defined as spin forbiddenness factor dependent only on the spin difference. At the partial case of $I_i = 0$, the sum in Eq. (1) is naturally replaced by the spin volume factor $(2I_f + 1)$

$$\mathrm{R} = \frac{(2I_f + 1)\mathrm{N_{at}} \ln 2}{\mathrm{T}_{1/2}(Q_\alpha)\mathrm{F}(\mathrm{I}_f)}. \qquad (2)$$

In literature, one can find the quantitative data on relative intensities for the ground state rotational band levels population in α-decay of the even-even nuclei. So, one can compare quantitatively the decay rate correspondent to ΔI = 0 and to ΔI = I_f. This case is most convenient for the analysis because the initial and final states have zero K quantum number and the structure hindrances do not appear. The data on ^{230}Th and ^{238}Ph taken from the Ref. [2] were analyzed using Eq. (2) and the corresponding Geiger-Nuttall functions for Th and Pu. For mentioned isotopes the population of levels with I_f up to 8 was observed, unfortunately the similar detailed data are absent in the range of elements near Hf. However, Th and Pu are also well deformed nuclei, and we suppose that not much difference arises due to the mass-number change. Really, the F(ΔI) value should depend on the reduced mass of α-particle and on the nuclear radius, proportional to $A^{1/3}$, and both are varied weakly with A. As a result, the spin hindrance function F(ΔI) was numerically characterized up to spin difference ΔI = 8. The ΔI in α-decay is released in a form of the α-particle orbital momentum l, so one can introduce also the function F(l) equal numerically to F(ΔI).

For the decay of ^{178m2}Hf, the spin difference may be as high as ΔI = 16, means the obtained above results are not enough. To extend F(ΔI) to higher argument values we used known data for the decay of high-spin isomers of ^{211m}Po, ^{212m}Po and ^{214m2}Rn. In the analysis, the decay rate of isomers is compared to that of the ground state of the same nuclide. The relative change of the spin difference is thus reduced for odd ^{211}Po nucleus, because even ground state decay is characterized by $\Delta I = 4$. But for even-even ^{212}Po and ^{214}Rn the spin difference corresponds to the isomer spin. In this manner we could get the resulted F(ΔI) value for $\Delta I = 18$, and could confirm the data obtained earlier for $\Delta I \leq 8$. In such analysis, Eq. (1) was used.

The isomers of Po and Rn are almost pure spin isomers, because K quantum number does not exist for near magic nuclides and their quasiparticle structure corresponds to well-allowed α-decay. Relative calibration by the decay rate of the ground states is also useful to exclude some possible systematical errors. We suppose that after all we succeed to isolate the spin forbiddenness factor in α-decay up to high values of parameter ΔI.

In Fig. 3 the F(l) = F(ΔI) function is shown. One can see the forbiddenness by many orders of magnitude for l values near 15, that is of our interest when we are going to estimate the α-decay of the 16^+ ^{178m2}Hf isomer. By analogy with the electromagnetic decay hindrances, one can introduce also the reduced forbiddenness factor (f) with the equation

$$F(l) = (f)^l. \qquad (3)$$

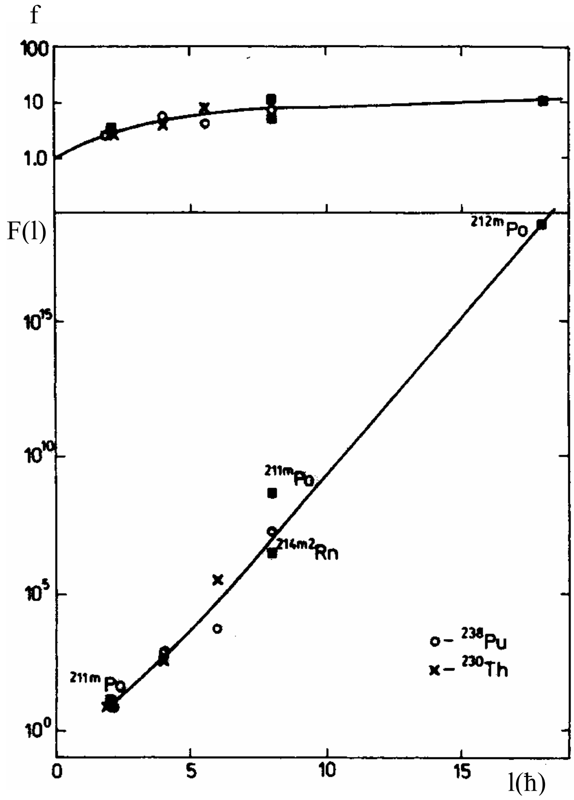

Fig. 3. Spin-forbiddenness factor F(l) versus angular momentum taken by α-particle and the reduced factor f(l) at the top panel of the Fig.

The f values are shown at the top panel in Fig. 3, they are not strongly varied at $l \geq 5$ saturating near f = 10. The reduced factors are used in the electromagnetic decay (and in our case) because they are more convenient being stable and easy applicable in calculations for the unstudied nuclei. Such procedure is used now in order to evaluate the properties of α-decay of the ^{178m2}Hf to the g.s. level of the daughter ^{174}Yb nucleus.

In the Table the estimated partial halflives are reduced for the transitions from $I_i = 16^+$ isomeric state to the levels with I_f^π varied from 14^+ to 0^+ in ^{174}Yb.

Calculated partial halflives* for α-decay of the ^{178m2}Hf isomer to the ^{174}Yb g.s. level

Transition	E_α, MeV	$T^i_{1/2}$, years
$16^+ \rightarrow 0^+$	4,43	$2{,}5 \cdot 10^{13}$
$16^+ \rightarrow 2^+$	4,35	$4{,}3 \cdot 10^{11}$
$16^+ \rightarrow 4^+$	4,18	$2{,}4 \cdot 10^{10}$
$16^+ \rightarrow 6^+$	3,91	$4{,}2 \cdot 10^{9}$
$16^+ \rightarrow 8^+$	3,56	$4{,}8 \cdot 10^{9}$
$16^+ \rightarrow 10^+$	3,12	$2{,}3 \cdot 10^{10}$
$16^+ \rightarrow 12^+$	2,61	$2{,}4 \cdot 10^{11}$
$16^+ \rightarrow 14^+$	2,03	$9{,}1 \cdot 10^{13}$

* Estimated total halflife $T_{1/2} = 1{,}8 \cdot 10^9$ y with account of all transitions.

The parity selection rule is satisfied because all levels have positive parity and I_i, I_f values are even. Eq. 1 was used and in the sum, main contribution gives the term correspondent to the minimum $(I_i - I_f)$ value because F(ΔI) is very steep function, as shown in Fig. 3.

One can see that shortest halflife is expected for the transitions to the 8^+ and 6^+ levels. In such cases, the optimum product of $T_{½}(Q_\alpha)$ and F(l) functions takes place. At higher I_f numbers, the F(l) is drastically reduced but $T_{½}(Q_\alpha)$ increases much. The opposite regulation appears for lower I_f numbers. The energy values of the g.s. level in ^{174}Yb are taken from Ref. [2]. Using the partial halflives $T^i_{1/2}$ one can calculate the full halflife for α-decay

$$T_{1/2}(\alpha) = \left(\sum_i \frac{1}{T^i_{1/2}} \right)^{-1}. \qquad (4)$$

The numerical value $T_{½}(\alpha) \approx 1{,}8\cdot 10^9$ y is predicted now. It would be important to compare this with the result of the experiment. The value would be in any assumptions much longer than the real halflife of ^{178m2}Hf: $T_{½} = 31$ y. It means that α-decay should be a branch of a very low probability in the isomer decay. But the comparison of experimental and estimated $T_{½}(\alpha)$ values can be conclusive for the K-hindrance manifestation in α-decay, thus may be of fundamental significance.

3. Experimental details

The source of ^{178m2}Hf isomer containing of about $3{,}5 \cdot 10^{13}$ isomeric atoms was prepared 10 years ago as a result of production series, Ref. [3], carried out at FLNR JINR using the ^{176}Yb (^{4}He, 2n) ^{178m2}Hf reaction. The Hf fraction was chemically isolated from the enriched ^{176}Yb material after exposure it to the ^{4}He-ion beam at an energy of 36 MeV produced by the U-200 cyclotron. The ^{178m2}Hf activity within the Hf fraction content was deposited onto the Be foil and formed a Hf oxide layer of small thickness allowing low energy losses at the transmission of α-particle. This was possible because the method of the Hf carrier was not applied to the chemical operations. Only Hf produced in nuclear reactions was contained in well purified and highly enriched ^{176}Yb target material.

The source includes definitely the equilibrium content of the 8^- ^{178m1}Hf isomeric nuclei in an amount of about $4 \cdot 10^{-9} \cdot N_{at}(^{178m2}\mathrm{Hf})$. Thus, the contribution of the m1 state into searched for α-activity of the source should be neglected. Though the spin forbiddenness F(ΔI) is reduced for m1 state, but the $T_{1/2}(Q_\alpha)$ factor produces an opposite influence.

It was important to control a presence of actinide α-active contaminations in the Hf source deposit and in the substrate foil. The α-spectroscopy using the Si surface barrier detector was applied for that. The ^{178m2}Hf source was placed in the vacuum chamber at a distance of 10 mm in front of Si detector with active area of 14 mm in diameter. The efficiency of the detector in such geometry was calibrated using ^{nat}U sample. Also it was found the energy resolution is better than 50 keV by the ^{234}U and ^{238}U α-particles. The ^{178m2}Hf source was kept during two weeks in the chamber pumped by the dry vacuum system repeatedly twice per week. The spectrum of α-particles was collected during this time and the background spectrum was also measured during two weeks in the identical condition, but without the source. Some radioactive contamination single events were observed in both spectra. Then, the alpha-energy range between 2,0 and 4,5 MeV was selected, it should cover all α-transition energies listed in the Table for assumed α-decay lines of ^{178m2}Hf to the ^{174}Yb rotational band levels, In the selected range all events were integrated and the “background” number of events (without source) was subtracted from the “effect” number of events (with source), The resulted value appeared to be negative, still within the statistical error

$$N_\alpha(^{178m2}\mathrm{Hf}) = (\,-17 \pm 25) \text{ events / 2 weeks.} \tag{5}$$

One may believe that such a measurement is not enough in sensitivity for the reliable estimation of the ^{178m2}Hf α-decay halflife. But it confirms the purity of the source from α-active contaminations because no statistically significant difference in the count rate is detected with and without source in front of the detector over whole range of α-energies from 1,5 to 9 MeV, as well.

Higher sensitivity method has been applied for detection of alphas emitted from ^{178m2}Hf. The source was kept during 7 months in close contact with the plastic track-detector foil of CR-39. The sandwich of the source in contact with the plastic detector was carefully packed with the plastic wrapping and placed then in the sealed plastic box. The penetration of the Rn gas should be stopped. Such kind of α-particle detection was well developed and calibrated by many groups using the detector foils of special production. This technique is used also in FLNR JINR. Tracks of α-particles should appear after exposure and after chemical etching of the detector foil. A number of tracks can be counted by optical-microscope visual registration. In principle, this method can be characterized as high efficiency and low background method.

However, in our case it was observed after the exposure and etching that the surface of the detector was moderately damaged. The roughness spot was seen directly by visual control without the microscope. This damage was created by high flux of low-energy electrons emitted from the source and it creates some continuous shade for the observation of α-particle tracks. Via the microscope the single tracks were detected and counted.

Series of more careful studies were carried out, as well. The CR-39 detecting foils were exposed during 1 and 3,5 months in contact with the source, and the etching time was slightly shortened. In such a condition, the detector surface damage due to electrons was significantly reduced and the tracks of α-particles were observed as clear images, in accordance with the standard α-tracks. Their number was in agreement with the ones previously detected in worse conditions. Total number of tracks was integrated over whole area contacted with the active spot of the source, and the out-exposure region was controlled for the background measurements at the same detectors.

Additional background measurements were taken with the similar CR-39 foils kept at the same environment, but in contact with the Be foil the same as a substrate of the source, with some other foils and without any inclusion. In all cases, the background track density was identical to the one observed at the detectors used for actual measurements with ^{178m2}Hf, but out of the active spot.

Finally, the background track density was measured with good statistical accuracy, and then was used for definition of the excess counts due to the presence of the ^{178m2}Hf activity. After integration of the results obtained in three exposure runs of about one year in total duration, it was possible to deduce the excess count rate due to α-activity of the source in a value of 2,1 alphas/day. This corresponds to the ^{178m2}Hf nuclei number of about $2{,}8 \cdot 10^{13}$ on the time of measurements. The resulted $T_{1/2}(\alpha)$ estimation follows:

$$T_{1/2}(\alpha) = (2{,}5 \pm 0{,}5)\ 10^{10}\ \text{y}. \qquad (6)$$

The statistical accuracy of about 10 % has been reached, but some systematical errors could not be excluded, for instance due to the estimated efficiency of the detectors.

The value is by a factor 15 longer than the calculated value, see in the previous section of the present report. One can understand such longer halflife as a manifestation of K-hindrance in α-decay, because it was not yet in account during the calculations. But a factor of 15 for the $\Delta K = 8$ transition to 8^+ level in ^{174}Yb seems relatively low as compared to the spin-hindrance $F(\Delta I = 8) \approx 10^6$ in Fig. 3. This may mean that really, K-hindrance is weakly manifested in α-decay, unlike to the γ-emission process.

Experimentally, the orders of magnitude improvement in sensitivity could normally be reached only by very strong efforts and high expenses. In principle, one can discuss a possibility to use as much as 10^{16} atoms of ^{178m2}Hf. It should be distributed over large area in order to prepare a thin source, and then inserted into a large area α-detector, probably an ionization chamber, or a proportional counter. After years of measurements, the sensitivity can be enhanced by a factor of 10^4. Thus, with further improved techniques more detail spectroscopic studies of ^{178m2}Hf α-decay will be possible.

Let's remind the calculated in Ref. [4] α-decay halflife for the ground state of ^{178}Hf: $T_{1/2}(\alpha) \approx 5 \cdot 10^{23}$ y. We have reached a level of $T_{½}(\alpha) \approx 2{,}5 \cdot 10^{10}$ y for the isomer, and this is a natural manifestation of the gain in Q_α value due to the isomeric level excitation energy of 2,446 MeV. This gain is partially suppressed by the spin hindrance factor, but the K-hindrance is weakly manifested. The $T_{1/2}$ value is quantitatively determined in the experiment with moderately good sensitivity because the halflife is found to be comparable with the age of the Earth.

4. Summary

The α-decay mode of the high-spin ^{178m2}Hf isomer was studied. Expected halflife was evaluated basing on known empirical regularities and with account of this isomer properties. After appropriate analysis, it was formulated that α-emission by ^{178m2}Hf being established experimentally will clarify a problem of K-hindrance manifestation in nuclear α-decay. The measurements were carried out using two independent methods for α-particle detection using the source containing $2{,}8 \cdot 10^{13}$ atoms of ^{178m2}Hf. The partial α-decay halflife was established experimentally to be $T_{½}(\alpha) \approx 2{,}5 \cdot 10^{10}$ y. This confirms strong retardation of α-decay due to the high changes of spin ΔI and remains also place to assume some forbiddenness due to the K quantum number changes ΔK. The estimated factor of 15 may confirm relatively weak manifestation of the K-hindrance in α-decay. The possibilities to increase a sensitivity of measurements are discussed.

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О ВОЗМОЖНОСТИ АЛЬФА-РАСПАДА ИЗОМЕРА ^{178m2}Hf

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Осуществлен поиск α-распада изомера ^{178m2}Hf. Это высокоспиновое (16^+) изомерное состояние, как известно, распадается путем конверсии электромагнитного перехода с энергией 12,7 кэВ и имеет период $T_{1/2}$ = 31 год. Полная энергия α-распада этого изомера Q_α = 4,53 МэВ должна соответствовать времени жизни порядка дней. Однако угловой момент изомерного состояния приводит к сильному запрету распада из-за центробежного барьера. Это и было установлено в эксперименте настоящей работы, поскольку измеренный парциальный полупериод α-распада оказался равным $2,5 \cdot 10^{10}$ лет. Обсуждается также возможный К-запрет в α-распаде.

ПРО МОЖЛИВІСТЬ АЛЬФА-РОЗПАДУ ІЗОМЕРУ ^{178M2}HF

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Здійснено пошук α-розпаду ізомеру ^{178m2}Hf. Цей високоспіновий (16+) ізомерний стан, як відомо, розпадається шляхом конверсії електромагнітного переходу з енергією 12,7 кеВ і має період $T_{1/2}$ = 31 рік. Повна енергія α-розпаду цього ізомеру Q_α = 4,53 МеВ повинна відповідати часу життя порядку днів. Однак кутовий момент ізомерного стану приводить до сильної заборони розпаду через відцентровий бар'єр. Це і було встановлено в експерименті даної роботи, оскільки виміряний парціальний напівперіод α-розпаду виявився рівним $2,5 \cdot 10^{10}$ років. Обговорюється також можлива К-заборона в α-розпаді.