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SPONTANEOUS DOUBLE ALPHA DECAY: FIRST EXPERIMENTAL LIMIT AND PROSPECTS OF INVESTIGATION

Nuclear decays with simultaneous emission of two alpha particles are energetically possible for a number of nuclides. Prospects of searching for such kind of decay for nuclides present in the natural isotopic composition of elements are discussed here. The first experimental limit on half-life for 2α decay is set for ^{209}Bi as $T_{1/2} > 2.9 \cdot 10^{20}$ y at 90 % C.L., using the data of work [P. de Marcillac et al. Nature 422 (2003) 876]. Theoretical $T_{1/2}$ estimations for the process are also given. Using these values, which are on the level of 10^{33} y or more, one can conclude that the prospects of experimental observation of 2α decay are very pessimistic.

Keywords: double alpha decay, low background experiments, theoretical and experimental half-lives.

1. Introduction

We know today several kinds of decay of atomic nuclei in which two particles of the same nature are emitted simultaneously.

Double beta (2β) decay, in which two electrons are emitted together with two antineutrinos, was predicted in 1935 by Goeppert-Mayer [1] and is registered to-date for 14 nuclides [2, 3]. Neutrinoless mode of this decay, in which only two electrons are emitted without antineutrinos, was first discussed by Furry in 1939 [4]. It is forbidden in the Standard Model (SM) of particles because it violates the lepton number by two units, but is predicted by many theories which consider the SM as a low-energy approximation; it is not observed yet but is actively searched for in many worldwide experiments [3, 5].

Deexcitation of an excited nuclear state with emission of two gammas, 2γ decay, was considered by Goeppert-Mayer in 1929 [6] (see also [7]). It was observed in 1963 for nuclei with $0^+ \rightarrow 0^+$ transitions, like ^{16}O, ^{40}Ca and ^{90}Zr, where emission of single γ is forbidden, and allowed are emissions of a conversion electron, or electron-positron pair, or two gammas [8]. In 2015, it was also observed at the first time for ^{137}Ba, where emission of a single γ is allowed, as a competitive mode on the level of 10^{-6} [9].

Also, nuclear decays with simultaneous emission of two protons (2p) and two neutrons (2n) are known; see [10] and Refs. therein.

However, possibility of nuclear decay with emission of two alpha particles at the same time (2α decay) was not analyzed on the same level of attention as the 2β, 2γ, 2p and 2n radioactivities. To our knowledge, it was discussed at the first time by Novikov in [11], with estimation of half-lives for few nuclides in [12]. It was considered also in theoretical article by Poenaru & Ivascu [13] (see also [14]). There were no experimental searches for such processes yet.

Here we analyze possible experimental investigations of nuclear 2α decay (Section 2), give theoretical estimations of half-lives for a number of nuclides present in the natural isotopic composition of elements (Section 3) and set the first experimental limit for such a process (Section 4). It should be noted that in interactions of nuclei with high-energy particles the defragmentation of nuclei with emission of few α particles is possible [15] but we are interested here in the spontaneous 2α decay.

2. Possible experimental approaches to search for 2α decay

Analyzing atomic masses from the last atomic mass evaluation [16], one can see that 2α decay is energetically possible for many nuclei (for 1459 isotopes from 3436 listed in [16]). Energy releases $Q_{2\alpha}$ can reach quite big values (maximal value is 22.7 MeV for ^{276}Cn) that, supposing an exponential dependence of half-life $T_{1/2}$ on $Q_{2\alpha}$ similar to single alpha decay, increases probability of this process. However, many of such nuclides are unstable with short half-lives respective to their main decay branch; they should be created at accelerators or extracted from fission products at nuclear reactors. It would be quite difficult to see a rare 2α process on the big radioactive α and β backgrounds. Instead, we will concentrate here on techniques which are typical in searches for rare nuclear decays, like 2β [2, 3, 5], single α and β [17], dark matter [18, 19], or in neutrino measurements [20].

In such an approach, massive (from ~0.1 kg to ~1 t) samples consisting of nuclides present in the natural isotopic composition of elements [21] are measured, usually in deep underground laboratories to decrease the background from cosmic muons.

Detectors, massive shieldings and all surrounding details are built from super-pure materials to suppress natural radioactivity that is necessary to see a small useful signal. Nuclides present in the natural composition of elements which are potentially unstable in relation to 2α decay are listed in Table.

Naturally abundant 80 nuclides candidates for 2α decay

Mother isotope	δ, % [21]	Q$_{2\alpha}$, keV [16]	Daughter isotope (T$_{1/2}$) [22]	Additional possible α and β decay modes of mother isotope	Theor. T$_{1/2}$, y [23]
^{144}Nd	23.798	289.1 ± 1.4	^{136}Ba (stable)	α (^{144}Nd $\rightarrow$ ^{140}Ce $\nrightarrow$ ^{136}Ba)	
^{145}Nd	8.293	1439.5 ± 1.4	^{137}Ba (stable)	α (^{145}Nd $\rightarrow$ ^{141}Ce $\nrightarrow$ ^{137}Ba)	
^{146}Nd	17.189	2485.9 ± 1.4	^{138}Ba (stable)	$2\beta^-$, α (^{146}Nd $\rightarrow$ ^{142}Ce $\rightarrow$ ^{138}Ba)	
^{148}Nd	5.756	1011.5 ± 8.2	^{140}Ba (12.7 d)	$2\beta^-$, α (^{148}Nd $\rightarrow$ ^{144}Ce $\rightarrow$ ^{140}Ba)	
^{147}Sm	15.00	2831.7 ± 7.5	^{139}Ce (137.6 d)	α (^{147}Sm $\rightarrow$ ^{143}Nd $\rightarrow$ ^{139}Ce)	
^{148}Sm	11.25	**3890.0 ± 2.1**	^{140}Ce (stable)	α (^{148}Sm $\rightarrow$ ^{144}Nd $\rightarrow$ ^{140}Ce)	$1.3\cdot10^{58}$
^{149}Sm	**13.82**	**3447.3 ± 2.1**	**^{141}Ce (32.5 d)**	α (^{149}Sm $\rightarrow$ ^{145}Nd $\rightarrow$ ^{141}Ce)	$4.7\cdot10^{66}$
^{150}Sm	7.37	2632.2 ± 2.8	^{142}Ce (stable)	α (^{150}Sm $\rightarrow$ ^{146}Nd $\rightarrow$ ^{142}Ce)	
^{152}Sm	26.74	819.4 ± 3.1	^{144}Ce (284.9 d)	α (^{152}Sm $\rightarrow$ ^{148}Nd $\rightarrow$ ^{144}Ce)	
^{151}Eu	**47.81**	**3565.6 ± 2.3**	**^{143}Pr (13.6 d)**	α (^{151}Eu $\rightarrow$ ^{147}Pm $\rightarrow$ ^{143}Pr)	$1.5\cdot10^{66}$
^{153}Eu	52.191	1408.9 ± 7.3	^{145}Pr (6.0 h)	α (^{153}Eu $\rightarrow$ ^{149}Pm $\rightarrow$ ^{145}Pr)	
^{152}Gd	0.20	**4191.2 ± 1.8**	^{144}Nd (2.3e15 y)	2ε, α (^{152}Gd $\rightarrow$ ^{148}Sm $\rightarrow$ ^{144}Nd)	$4.1\cdot10^{56}$
^{154}Gd	2.18	2370.1 ± 1.8	^{146}Nd (stable)	α (^{154}Gd $\rightarrow$ ^{150}Sm $\rightarrow$ ^{146}Nd)	
^{155}Gd	14.80	1227.0 ± 1.8	^{147}Nd (11.0 d)	α (^{155}Gd $\rightarrow$ ^{151}Sm $\rightarrow$ ^{147}Nd)	
^{156}Gd	20.47	23.3 ± 2.4	^{148}Nd (stable)	α (^{156}Gd $\rightarrow$ ^{152}Sm $\rightarrow$ ^{148}Nd)	
^{156}Dy	0.056	**3957.5 ± 1.8**	^{148}Sm (7.0e15 y)	$\varepsilon\beta^+$, α (^{156}Dy $\rightarrow$ ^{152}Gd $\rightarrow$ ^{148}Sm)	$1.9\cdot10^{64}$
^{158}Dy	0.095	1794.0 ± 2.7	^{150}Sm (stable)	2ε, α (^{158}Dy $\rightarrow$ ^{154}Gd $\rightarrow$ ^{150}Sm)	
^{160}Dy	2.329	240.0 ± 1.4	^{152}Sm (stable)	α (^{160}Dy $\rightarrow$ ^{156}Gd $\nrightarrow$ ^{152}Sm)	
^{162}Er	0.139	2521.6 ± 1.5	^{154}Gd (stable)	$\varepsilon\beta^+$, α (^{162}Er $\rightarrow$ ^{158}Dy $\rightarrow$ ^{154}Gd)	
^{164}Er	1.601	1742.2 ± 1.4	^{156}Gd (stable)	2ε, α (^{164}Er $\rightarrow$ ^{160}Dy $\rightarrow$ ^{156}Gd)	
^{166}Er	33.503	913.7 ± 1.7	^{158}Gd (stable)	α (^{166}Er $\rightarrow$ ^{162}Dy $\rightarrow$ ^{158}Gd)	
^{167}Er	22.869	420.4 ± 1.7	^{159}Gd (18.5 h)	α (^{167}Er $\rightarrow$ ^{163}Dy $\nrightarrow$ ^{159}Gd)	
^{168}Er	26.978	100.7 ± 1.7	^{160}Gd (stable)	α (^{168}Er $\rightarrow$ ^{164}Dy $\nrightarrow$ ^{160}Gd)	
^{169}Tm	100	1336.5 ± 1.6	^{161}Tb (6.9 d)	α (^{169}Tm $\rightarrow$ ^{165}Ho $\rightarrow$ ^{161}Tb)	
^{168}Yb	0.123	**3241.0 ± 1.4**	^{160}Dy (stable)	$\varepsilon\beta^+$, α (^{168}Yb $\rightarrow$ ^{164}Er $\rightarrow$ ^{160}Dy)	
^{170}Yb	2.982	2567.7 ± 0.8	^{162}Dy (stable)	α (^{170}Yb $\rightarrow$ ^{166}Er $\rightarrow$ ^{162}Dy)	
^{171}Yb	14.086	2224.5 ± 0.8	^{163}Dy (stable)	α (^{171}Yb $\rightarrow$ ^{167}Er $\rightarrow$ ^{163}Dy)	
^{172}Yb	21.686	1862.7 ± 0.8	^{164}Dy (stable)	α (^{172}Yb $\rightarrow$ ^{168}Er $\rightarrow$ ^{164}Dy)	
^{173}Yb	16.103	1211.6 ± 0.8	^{165}Dy (2.3 h)	α (^{173}Yb $\rightarrow$ ^{169}Er $\rightarrow$ ^{165}Dy)	
^{174}Yb	32.025	790.4 ± 0.9	^{166}Dy (81.6 h)	α (^{174}Yb $\rightarrow$ ^{170}Er $\rightarrow$ ^{166}Dy)	
^{176}Yb	12.995	218 ± 140	^{168}Dy (8.7 m)	$2\beta^-$, α (^{176}Yb $\rightarrow$ ^{172}Er $\nrightarrow$ ^{168}Dy)	
^{175}Lu	97.401	2265.2 ± 5.4	^{167}Ho (3.1 h)	α (^{175}Lu $\rightarrow$ ^{171}Tm $\rightarrow$ ^{167}Ho)	
^{176}Lu	2.599	1829 ± 30	^{168}Ho (3.0 m)	α (^{176}Lu $\rightarrow$ ^{172}Tm $\rightarrow$ ^{168}Ho)	
^{174}Hf	0.16	**4231.7 ± 2.5**	^{166}Er (stable)	$\varepsilon\beta^+$, α (^{174}Hf $\rightarrow$ ^{170}Yb $\rightarrow$ ^{166}Er)	$8.2\cdot10^{69}$
^{176}Hf	5.26	**3565.0 ± 1.9**	^{168}Er (stable)	α (^{176}Hf $\rightarrow$ ^{172}Yb $\rightarrow$ ^{168}Er)	
^{177}Hf	**18.60**	**3192.7 ± 1.8**	**^{169}Er (9.4 d)**	α (^{177}Hf $\rightarrow$ ^{173}Yb $\rightarrow$ ^{169}Er)	$2.4\cdot10^{93}$
^{178}Hf	27.28	2823.6 ± 2.1	^{170}Er (stable)	α (^{178}Hf $\rightarrow$ ^{174}Yb $\rightarrow$ ^{170}Er)	
^{179}Hf	13.62	2406.2 ± 2.1	^{171}Er (7.5 h)	α (^{179}Hf $\rightarrow$ ^{175}Yb $\rightarrow$ ^{171}Er)	
^{180}Hf	35.08	1854.4 ± 4.3	^{172}Er (49.3 h)	α (^{180}Hf $\rightarrow$ ^{176}Yb $\rightarrow$ ^{172}Er)	

Continuation of the Table

Mother isotope	δ, % [21]	$Q_{2\alpha}$, keV [16]	Daughter isotope ($T_{1/2}$) [22]	Additional possible α and β decay modes of mother isotope	Theor. $T_{1/2}$, y [23]
^{180}Ta	0.01201	**3591.7 ± 5.8**	**^{172}Tm (63.6 h)**	α (^{180}Ta → ^{176}Lu → ^{172}Tm)	$1.2\cdot10^{85}$
^{181}Ta	99.98799	2967.9 ± 4.6	^{173}Tm (8.2 h)	α (^{181}Ta → ^{177}Lu → ^{173}Tm)	
^{180}W	0.12	**4769.5 ± 1.4**	^{172}Yb (stable)	2ε, α (^{180}W → ^{176}Hf → ^{172}Yb)	$1.4\cdot10^{64}$
^{182}W	26.50	**3848.6 ± 0.7**	^{174}Yb (stable)	α (^{182}W → ^{178}Hf → ^{174}Yb)	
^{183}W	**14.31**	**3480.1 ± 0.7**	**^{175}Yb (4.2 d)**	α (^{183}W → ^{179}Hf → ^{175}Yb)	$6.4\cdot10^{89}$
^{184}W	30.64	2936.1 ± 0.7	^{176}Yb (stable)	α (^{184}W → ^{180}Hf → ^{176}Yb)	
^{186}W	28.43	2337 ± 10	^{178}Yb (74 m)	$2\beta^-$, α (^{186}W → ^{182}Hf → ^{178}Yb)	
^{185}Re	**37.40**	**3714.9 ± 1.5**	**^{177}Lu (6.7 d)**	α (^{185}Re → ^{181}Ta → ^{177}Lu)	$1.2\cdot10^{86}$
^{187}Re	**62.60**	**2992.6 ± 5.2**	**^{179}Lu (4.6 h)**	α (^{187}Re → ^{183}Ta → ^{179}Lu)	$7.9\cdot10^{105}$
^{184}Os	0.02	**5474.0 ± 1.7**	^{176}Hf (stable)	$\varepsilon\beta^+$, α (^{184}Os → ^{180}W → ^{176}Hf)	$1.4\cdot10^{57}$
^{186}Os	1.59	**4285.5 ± 1.6**	^{178}Hf (stable)	α (^{186}Os → ^{182}W → ^{178}Hf)	$8.0\cdot10^{75}$
^{187}Os	1.96	**4394.1 ± 1.6**	^{179}Hf (stable)	α (^{187}Os → ^{183}W → ^{179}Hf)	$6.9\cdot10^{73}$
^{188}Os	13.24	**3792.3 ± 1.6**	^{180}Hf (stable)	α (^{188}Os → ^{184}W → ^{180}Hf)	
^{189}Os	**16.15**	**3566.3 ± 1.6**	**^{181}Hf (42.4 d)**	α (^{189}Os → ^{185}W → ^{181}Hf)	$4.0\cdot10^{91}$
^{190}Os	26.26	2491.9 ± 6.2	^{182}Hf (9.0e5 y)	α (^{190}Os → ^{186}W → ^{182}Hf)	
^{192}Os	40.78	767 ± 40	^{184}Hf (4.1 h)	$2\beta^-$, α (^{192}Os → ^{188}W → ^{184}Hf)	
^{191}Ir	**37.3**	**3734.2 ± 1.9**	**^{183}Ta (5.1 d)**	α (^{191}Ir → ^{187}Re → ^{183}Ta)	$2.8\cdot10^{89}$
^{193}Ir	62.7	2008 ± 14	^{185}Ta (49.4 m)	α (^{193}Ir → ^{189}Re → ^{185}Ta)	
^{190}Pt	0.012	**6089.8 ± 1.0**	^{182}W (stable)	$\varepsilon\beta^+$, α (^{190}Pt → ^{186}Os → ^{182}W)	$2.7\cdot10^{52}$
^{192}Pt	0.782	**4567.1 ± 2.7**	^{184}W (stable)	α (^{192}Pt → ^{188}Os → ^{184}W)	$1.4\cdot10^{74}$
^{194}Pt	32.864	2898.6 ± 1.3	^{186}W (stable)	α (^{194}Pt → ^{190}Os → ^{186}W)	
^{195}Pt	33.775	2263.3 ± 1.3	^{187}W (23.7 h)	α (^{195}Pt → ^{191}Os → ^{187}W)	
^{196}Pt	25.211	1173.5 ± 3.1	^{188}W (69.4 d)	α (^{196}Pt → ^{192}Os → ^{188}W)	
^{197}Au	100	1989.5 ± 8.2	^{189}Re (24.3 h)	α (^{197}Au → ^{193}Ir → ^{189}Re)	
^{196}Hg	0.15	**4461.5 ± 3.0**	^{188}Os (stable)	2ε, α (^{196}Hg → ^{192}Pt → ^{188}Os)	$3.4\cdot10^{79}$
^{198}Hg	10.04	2903.6 ± 0.8	^{190}Os (stable)	α (^{198}Hg → ^{194}Pt → ^{190}Os)	
^{199}Hg	16.94	1999.3 ± 0.8	^{191}Os (15.4 d)	α (^{199}Hg → ^{195}Pt → ^{191}Os)	
^{200}Hg	23.14	1529.1 ± 2.4	^{192}Os (stable)	α (^{200}Hg → ^{196}Pt → ^{192}Os)	
^{201}Hg	13.17	881.9 ± 2.4	^{193}Os (30.5 h)	α (^{201}Hg → ^{197}Pt → ^{193}Os)	
^{202}Hg	29.74	240.0 ± 2.5	^{194}Os (6.0 y)	α (^{202}Hg → ^{198}Pt → ^{194}Os)	
^{203}Tl	29.44	1081.0 ± 1.8	^{195}Ir (2.5 h)	α (^{203}Tl → ^{199}Au → ^{195}Ir)	
^{204}Pb	1.4	2684.8 ± 1.3	^{196}Pt (stable)	α (^{204}Pb → ^{200}Hg → ^{196}Pt)	
^{206}Pb	24.1	1268.6 ± 2.4	^{198}Pt (stable)	α (^{206}Pb → ^{202}Hg → ^{196}Pt)	
^{207}Pb	22.1	86.8 ± 2.4	^{199}Pt (30.8 m)	α (^{207}Pb → ^{203}Hg ↛ ^{199}Pt)	
^{208}Pb	52.4	0.7 ± 20	^{200}Pt (12.5 h)	α (^{208}Pb → ^{204}Hg ↛ ^{200}Pt)	
^{209}Bi	**100**	**3292.2 ± 3.5**	**^{201}Au (26 m)**	α (^{209}Bi → ^{205}Tl → ^{201}Au)	$4.3\cdot10^{113}$
^{232}Th	**99.98**	**8151.9 ± 9.9**	**^{224}Rn (107 m)**	$2\beta^-$, α (^{232}Th → ^{228}Ra → ^{224}Rn)	$6.3\cdot10^{46}$
^{231}Pa*	**100**	**10192.2 ± 2.6**	**^{223}Fr (21.8 m)**	α (^{231}Pa → ^{227}Ac → ^{223}Fr)	$1.5\cdot10^{33}$
^{234}U	0.0054	**9627.4 ± 2.2**	^{226}Ra (1600 y)	α (^{234}U → ^{230}Th → ^{226}Ra)	$6.3\cdot10^{37}$
^{235}U	**0.7204**	**8891.3 ± 2.2**	**^{227}Ra (42.2 m)**	α (^{235}U → ^{231}Th → ^{227}Ra)	$1.2\cdot10^{43}$
^{238}U	**99.2742**	**7941.6 ± 10.4**	**^{230}Ra (93 m)**	α (^{238}U → ^{234}Th → ^{230}Ra)	$9.3\cdot10^{50}$

* While ^{231}Pa is listed in [21] as present in the natural isotopic composition with abundance δ = 100 %, in fact, it is unstable nuclide with quite short half-life $T_{1/2}$ = 32760 y [22].

In Table δ is the natural abundance of the mother isotope [21]. Energy release $Q_{2\alpha}$ is calculated with atomic masses [16]. Half-life $T_{1/2}$ is given for the daughter isotope [22]. If value of $Q_{2\alpha}$ is greater than 3 MeV, it is marked in bold (red); in this case big abundance (if so) and unstable daughter isotope (if so) are also marked in bold (red). In the last column, the theoretical estimation of $T_{1/2}$ is given, calculated with receipt from [23] for ^{8}Be emission. 2ε is for double electron capture, $\varepsilon\beta^{+}$ is for electron capture with positron emission.

One can see from the Table that all the 2α candidates also are potentially unstable in relation to single α decay. Intermediate (A–4,Z–2) nuclides always live long enough [22] not to imitate 2α decay in fast chain of two single α decays; and in few cases (^{144,145}Nd, ^{160}Dy, ^{167,168}Er, ^{176}Yb, ^{207,208}Pb) α decay of the intermediate nuclide is energetically forbidden.

Different methods can be used to look for the 2α decay, e.g.:

1) Detection of nuclear recoils which in case of 2α decay can have energies higher than those in single α decay (depending on the angle between the emitted α's) [11, 12];

2) Si detectors (or some others, e.g. nuclear emulsions) can register two α particles emitted from an external source and measure their energies; however, samples in this case should be very thin that restricts the mass that can be measured;

3) An external source can be placed on e.g. HPGe detector; if the daughter nuclide is unstable, one can register characteristic γ quanta emitted in its decay. In this case, mass of a sample could be a few kilograms; however, efficiency of HPGe detectors is on the level of only few percents. Other possible origins of the daughter nuclides should be also taken into account (resulting from e.g. cosmogenic production, or due to fission of U/Th present in some amounts in the investigated source);

4) Very promising is a "source = detector" approach, when a mother nuclide is embedded in a detector itself as its main component (like W isotopes in $CdWO_4$ or ^{209}Bi in $Bi_4Ge_3O_{12}$) or as a dopant (like ^{203}Tl in NaI(Tl)). This approach gives possibility to use detectors with big masses (up to ~1 t) and ensures high efficiency of registration of the 2α process (practically 100 %). Scintillators can be used; however, in this case one cannot expect high-energy resolution (it will be on the level of few tens or hundreds keV). In addition, scintillators have different light yields for β and γ particles comparing to those for α particles of the same energy (quenching effect, see e.g. [24]). Thus, 2α light signal will be quenched and shifted to lower energies, where α and β/γ backgrounds are higher. Instead, scintillating bolometers, devices able to measure simultaneously light and heat signals for the same event (see e.g. [25]), are very promising techniques. A signal in the heat channel is not quenched (thus, one will see 2α signal at $Q_{2\alpha}$ value but not at lower energies). In addition, energy resolution in the heat channel is on the level of few keV.

3. Theoretical estimation of $T_{1/2}$ for 2α decay

Half-lives for ^{148}Sm, ^{152}Gd, ^{156}Dy, ^{190}Pt and ^{234}U were estimated in [12]. There is a good agreement between the values of [12] and results obtained in this work (within 1 – 2 orders of magnitude, if to take also into account difference in $Q_{2\alpha}$ values known in 1986 and the current $Q_{2\alpha}$'s [16]).

Calculations of 2α decay half-lives for some nuclei in framework of the superasymmetric fission model were performed in [13]. The results are presented in table [13, 14], and also in graphical form as a logarithm of ratio of probability to emit two α's to that for single α emission. For nuclides presented in Table, results are absent.

To estimate the half-life values for 2α emission, we use here semi-empirical formulae for cluster decays developed in [23] and applied for emission of ^{8}Be nucleus. As it is known, ^{8}Be is highly unstable decaying to two α's with $T_{1/2}$ ~ 10^{-16} s and Q_α = = 91.8 keV [22]. Thus, the energy releases in 2α decay should be higher on 91.8 keV comparing to decay with ^{8}Be emission, and corresponding $T_{1/2}$ values should be slightly smaller. However, the difference is not big [13] and is acceptable for our aims of $T_{1/2}$ estimation. The results for some prospective nuclei are given in Table. One can see that even for the most promising cases, half-life values are too big for 2α decay's experimental observation.

4. First experimental $T_{1/2}$ limit for 2α decay of ^{209}Bi

To obtain the first limit on 2α decay of ^{209}Bi, we can use data from the experiment [26] in which single α decay of ^{209}Bi was observed at the first time. In particular, a BGO ($Bi_4Ge_3O_{12}$) scintillating bolometer with mass of 45.7 g was measured during 100 h in this work. Because of different light yields for β and γ particles comparing to those for α particles of the same energy, energy spectrum of α particles and nuclear recoils is effectively discriminated from much more intensive β and γ background. Because of small range of α particles in the BGO crystal, they are absorbed in the crystal with practically 100 % efficiency. In the heat channel, we should observe a peak for the single α decay of ^{209}Bi at Q_α = 3137.3 ±

± 0.8 keV [16], and for the double α decay at $Q_{2\alpha}$ = = 3292.2 ± 3.5 keV (see Table). In the data presented in Fig. 2b of Ref. [26], one really sees the peak for ^{209}Bi single α decay, while the peak at $Q_{2\alpha}$ is absent. In fact, no events are registered in the energy range 3.2 - 3.4 MeV which fully contains the expected 2α peak (full width at half maximum, FWHM is ~15 keV [26]). Thus, we can estimate only $T_{1/2}$ limit for the ^{209}Bi 2α decay with the formula:

$$\lim T_{1/2} = \ln 2 \cdot \varepsilon \cdot N_{209} \cdot t / \lim S,$$

where ε is the efficiency to detect the expected 2α decay (ε = 1), N_{209} is the number of ^{209}Bi nuclei in the 45.7 g $Bi_4Ge_3O_{12}$ crystal ($N_{209} = 8.84 \cdot 10^{22}$), t is the time of measurements (t = 100 h), and lim S is the upper limit on the number of events of the effect searched for which can be excluded at a given confidence level (C.L.). In accordance with the Feldman-Cousins recommendations [27], for 0 registered events (and with 0 supposed background), lim S = = 2.44 at 90 % C.L. Substituting all the values in the formula above, we obtain the following constraint on the ^{209}Bi 2α decay:

$$T_{1/2} > 2.9 \cdot 10^{20} \text{ y at 90 \% C.L.}$$

The obtained experimental limit is very far from the theoretical expectations presented in Section 3 (for ^{209}Bi, $T_{1/2} = 4.3 \cdot 10^{113}$ y).

Half-life limit for 2α decay of ^{232}Th, nuclide with quite big energy release $Q_{2\alpha}$ = 8152 keV and high natural abundance near 100 %, could be derived from e.g. measurements [28] of 2 kg ThO_2 sample with HPGe detector at the Yangyang underground laboratory (Korea). The daughter nuclide ^{224}Rn is unstable and β decays further to ^{224}Fr with Q_β = 800 keV and $T_{1/2}$ = 107 m [22]. One can look for characteristic γ quanta emitted in ^{224}Rn (and ^{224}Fr) decay.

5. Conclusions

Nuclear decay (A,Z) → (A–8,Z–4) + 2α with simultaneous emission of two alpha particles is energetically allowed for near 40 % of known isotopes (1459 from 3436 listed in [16]). Among them, 80 nuclides are present in the natural isotopic composition of elements [21]. First experimental limit for this kind of radioactivity is given for ^{209}Bi as $T_{1/2} > 2.9 \cdot 10^{20}$ y at 90 % C.L. Theoretical $T_{1/2}$ estimations are calculated for the most prospective candidates. However, the calculated $T_{1/2}$ values are very big, 10^{33} y or more, making prospects for future observation of such processes very pessimistic.

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Note added in proofs. After appearance of eprint version of this work [29], very recently 2α decay of ^{212}Po and ^{224}Ra was described in a microscopic framework based on energy density functional [30]. The calculated half-lives for two alpha particles emitted in opposite directions were found to be 20 (13) orders of magnitude lower for ^{212}Po (^{224}Ra) in comparison with those obtained with semi-empirical formula for ^{8}Be emission [23]. This gives more hopes for experimental investigation of the process.

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СПОНТАННИЙ ПОДВІЙНИЙ АЛЬФА РОЗПАД: ПЕРШЕ ЕКСПЕРИМЕНТАЛЬНЕ ОБМЕЖЕННЯ ТА ПЕРСПЕКТИВИ ДОСЛІДЖЕНЬ

Ядерні розпади з одночасним випромінюванням двох альфа частинок енергетично можливі для ряду нуклідів. Обговорюються перспективи пошуку такого розпаду для нуклідів, присутніх у природному ізотопному складі елементів. Отримано перше експериментальне обмеження на період напіврозпаду для 2α розпаду ^{209}Bi $T_{1/2} > 2{,}9 \cdot 10^{20}$ р. при 90 % довірчій імовірності, з використанням даних роботи [P. de Marcillac et al. Nature 422 (2003) 876]. Також наводяться теоретичні оцінки $T_{1/2}$ для такого процесу. Ці значення знаходяться на рівні 10^{33} і більше років, з чого можна зробити висновок, що перспективи експериментального спостереження 2α розпаду дуже песимістичні.

Ключові слова: подвійний альфа розпад, експерименти з низьким фоном, теоретичні та експериментальні періоди напіврозпаду.

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