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ALPHA DECAY OF Pu^{241}

B. S. DZHELEPOV, R. B. IVANOV, and V. G. NEDOVESOV

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The α spectrum of Pu^{241} was studied with a magnetic $\pi\sqrt{2}$ α spectrometer. The following α transitions were observed (in keV): 5042 (1.5%), 4973 (1.4%), 4899 (83%), 4862 (13%), and 4805 (1.1%). The decay scheme of Pu^{241} is discussed.

THE investigation of the α decay of Pu^{241} (T = 13 years)[1] is very complicated because:

1) Only 2.3×10^{-3} % of the decays are α decays,[2] corresponding to an α half-life of 5.7 $\times 10^5$ years.

2) The intensity of all Pu^{241} α transitions in the investigated samples was, as a rule, hundreds of times smaller than the α-transition intensities of Pu^{238} and Pu^{240} at higher energies E_α. This means that the Pu^{241} lines are located in the tails of the Pu^{238} and Pu^{240} lines.

3) The energies of the main Pu^{241} α transitions are very close to the energies of Pu^{242} α transitions (Table I). It follows that the possibility of studying the Pu^{241} α spectrum also depends on the α-activity ratio of Pu^{241} and Pu^{242} in a given sample.

The foregoing circumstances appear to account for the fact that very little information regarding Pu^{241} α decay is found in the literature. Strominger, Hollander, and Seaborg[3] give data for two α groups at 4893 and 4848 keV with 75% and 25% intensities, respectively. On the basis of data in [4] indicating the existence of a 145-keV γ line in the Pu^{241} γ spectrum it was suggested in [3] that this γ line represents a transition from the 145-keV level of U^{237}, reached by a 4893-keV α transition, to the ground state. The intensity of this transition is the fraction 2×10^{-6} of Pu^{241} decays; this corresponds to $2 \times 10^{-6}/2.3 \times 10^{-5}$ = 9% of the number of α decays. The γ spectrum also reveals a line at about 100 keV, which appears to be a K x-ray line of U^{237} with the intensity 1×10^{-5}, i.e., $1 \times 10^{-5}/2.3 \times 10^{-5}$ = 45% of the number of α decays.

Information regarding the U^{237} level scheme can also be obtained by studying the β decay of Pa^{237}. The first data regarding the β decay of this isotope, to which the half-life 11 min was assigned, appeared in [5]. Subsequently 39 ± 3 min was obtained for the half-life by Takahashi and Morinaga,[6] who observed three endpoints at 2.30, 1.35, and 0.8 MeV in the β spectrum of Pa^{237}, as well as 17 γ lines from 90 to 1420 keV. These included the aforementioned 145-keV transition. One possible Pa^{237} decay scheme, based on these data, was discussed.

EXPERIMENT

We studied the Pu^{241} α spectrum using the magnetic $\pi\sqrt{2}$ α spectrometer of the Radium Institute of the Academy of Sciences.[7] The measurements were obtained under the same conditions as in our

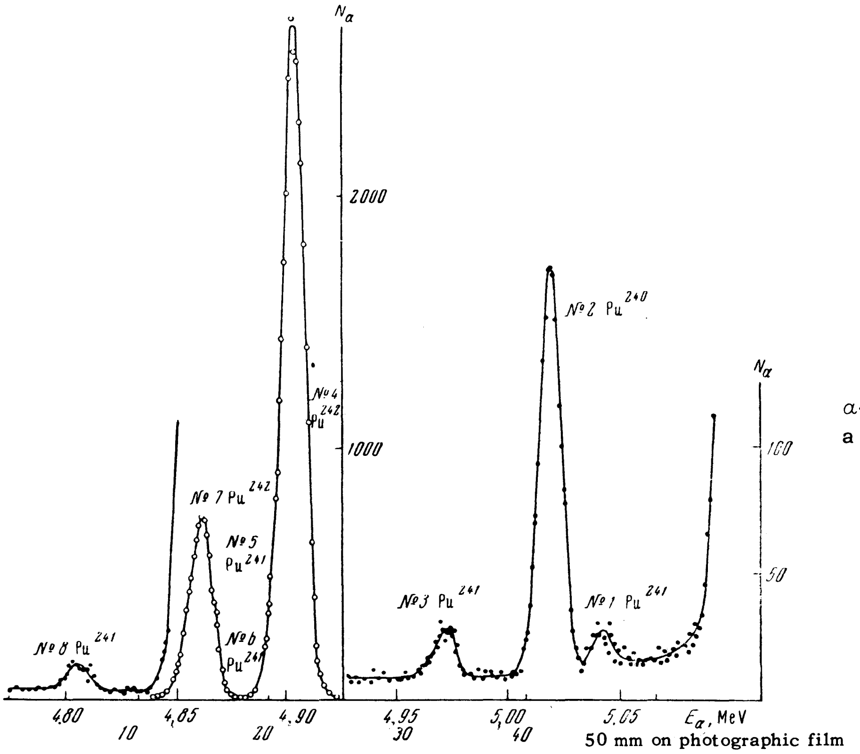


FIG. 1. Portion of Pu^{241} α spectrum. E_α – α-particle energy, N_α – number of α tracks in a band 300μ wide.

investigation of the α spectra of curium isotopes.[8] The 1 × 10-mm source was prepared by vacuum deposition on glass. The surface density of about 20 $\mu g/cm^2$ resulted in a broadening of the α lines (10–11-keV half-width) and some lengthening of their tails. This source thickness was required in order to obtain information about the relatively weak Pu^{241} lines.

Each of the four exposures lasted 90 hours. Figure 1 shows a portion of the α spectrum of plutonium isotopes in the energy range 4.75–5.10 MeV, obtained in one of the exposures. The spectrum was recorded when 5.05-MeV α particles moved in a circular orbit. During the other exposures the magnetic fields were somewhat differerent, and the α particles moved in circular orbits at the energies 5.15, 4.95, and 4.88 MeV.

Table I gives our results together with some previously tabulated data. The α spectra of the isotopes $Pu^{238,240,242}$ have been studied very thoroughly, so that any observed α line not belonging to any one of these isotopes can be assigned to Pu^{241}. In each instance the α energies were also compared with data for all possible α-active impurities. The table gives the α-transition energies. The reference line is Pu^{240} α_2 at 5020 keV (line No. 2), whose energy is known very accurately (± 2 keV).[3,9]

There is a notable disagreement with the values that we obtained in earlier work for the Pu^{242} α transitions and the most intense Pu^{241} transition. The discrepancy of about 6 keV can be attributed to a shift of the energy scale toward higher values. This shift has recently been observed in the entire

Table I

No. of line	Plutonium isotope to which the α transition is assigned	Tabulated data[3,9]		Our data	
		E_α, keV	Relative intensity in the isotope, %	E_α, keV	Relative intensity in the isotope, %
1	Pu^{241}			5042±4	1.5±0,5
2	Pu^{240}	5020	0,1	5020	0,1
3	Pu^{241}			4973±4	1,4±0.3
4	Pu^{242}	4898	76	4904±3	75±2
5	Pu^{241}	4893	75	4899±4	83±8
6	Pu^{241}	4848	25	4862±4	13±3
7	Pu^{242}	4853	24	4859±3	25±2
8	Pu^{241}			4805±4	1,1±0.3

range of α energies. Our reference line in these measurements was the 5020-keV Pu^{240} line; this value corresponds to the energy of the 5169-keV Pu^{240} main transition given in [9]. In earlier work the assigned energies of the Pu^{240} α_0 and α_2 groups were 5162 keV and 5014 keV, respectively.[3] This can possibly account for the too low energies of the main Pu^{242} and Pu^{241} transitions given in [3].

The experimental curve (Fig. 1) reveals, in addition to the single lines Nos. 1, 3, and 8, two composite lines 4—5 and 6—7, whose half-widths show that they are at least double lines. From earlier work it is known that these lines should include doublets of the main Pu^{241} and Pu^{242} lines. We were unable to resolve these lines experimentally because of low resolving power. The resolution shown in Fig. 2 is based on the following considerations.

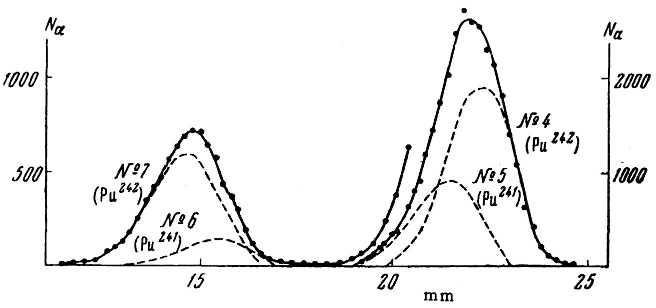


FIG. 2. Resolution of lines 4–5 and 6–7 (Pu^{241} and Pu^{242}) (on photographic plate).

First, the line shape in this region was determined from the line α_2 Pu^{240} (line No. 2). However, a resolution based only on knowledge of the line shape could result in a large error (greater than 20%) in both the intensities and energies of the transitions.

Secondly, lines 4 and 7 belong to the Pu^{242} α spectrum, representing transitions to the 0^+ and 2^+ levels of U^{238}. According to the literature these lines differ by 45 keV (from α spectra[3] and Coulomb excitation[10]) and their intensity ratio is 76/24.[3] Also, the intensity ratio of α transitions to the 0^+ and 2^+ levels of all neighboring even-even nuclei does not vary much; the average ratio is $(75 \pm 2)/(25 \pm 2)$. Lines 4—7 can therefore be resolved quite accurately ($\pm 10\%$), because, in addition to the line shape, we know the energy difference of lines 4 and 7, and their intensity ratio.

As a result of the foregoing line resolution it was found that

1) the energy difference of Pu^{242} line No. 4 and Pu^{241} line No. 5 agrees with the literature;

2) the energy of Pu^{241} α transition No. 6 is higher than the energy of Pu^{242} transition No. 7, thus conflicting with the results of Asaro et al.;[3]

3) the intensity ratio of Pu^{241} lines 5 and 6 is $(83 \pm 8)/(13 \pm 3)$, which also disagrees with the results of Asaro et al.,[3] who obtained 75/25.

At energies below 4.8 MeV we observed no Pu^{241} α-decay lines with intensities above 0.2%.

Table I shows our data for the relative intensities of α transitions in each plutonium isotope. Table II gives the excitation energies of U^{237} levels and the hindrance factors F for the α transitions, calculated from the data of Bohr et al.[11]

DISCUSSION OF Pu^{241} DECAY SCHEME

The Pu^{241} ground-state spin was found by means of magnetic resonance to be $5/2$.[12] This agrees well with the Nilsson scheme for single-particle states of deformed nuclei. The ground state of a nucleus containing 147 neutrons is a $5/2^+$ [622] level.

A. Transitions with small F

The hindrance factor for the 4899-keV α transition is 1.5, which indicates that the transition is favored and goes to a $5/2^+$ state. The 4862- and 4805-keV transitions are also characterized by small values of F (5.2 and 25.0); this is characteristic of transitions to levels of a single rotational band, which in the present case has the quantum number $K = 5/2$. The probability ratios of α transitions to rotational band levels as calculated from the formula in [11] for Pu^{241} are

$$J(5/2 \to 5/2) : J(5/2 \to 7/2) : J(5/2 \to 9/2) = 100 : 13 : 2.2.$$

This result agrees satisfactorily with our ratio 100:16:1.3, thus confirming the hypothesis that the three U^{237} levels reached by favored decays are members of the $K = 5/2$ rotational band which have spins and parities $5/2^+$, $7/2^+$, and $9/2^+$. The transition intensity ratio $J(5/2 \to 5/2) : J(5/2 \to 7/2) = 100:33$ given in [3] is evidently incorrect. However, the energy differences between these three levels are not in accord with the interval ratio derived from the formula for excited levels of the $K = 5/2$ rota-

Table II

α transition	E_α, keV	Energy level,* keV	Relative intensity, %	Hindrance factor, F
α_0	5042±4	0	1.5±0.5	700
α_1	4973±4	70±2	1.4±0.3	250
α_2	4899±4	145±2	83±8	1.5
α_3	4862±4	183±3	13±3	5.2
α_4	4805±4	241±3	1.1±0.3	25

*Assuming that α_0 is a ground-state transition of U^{237}.

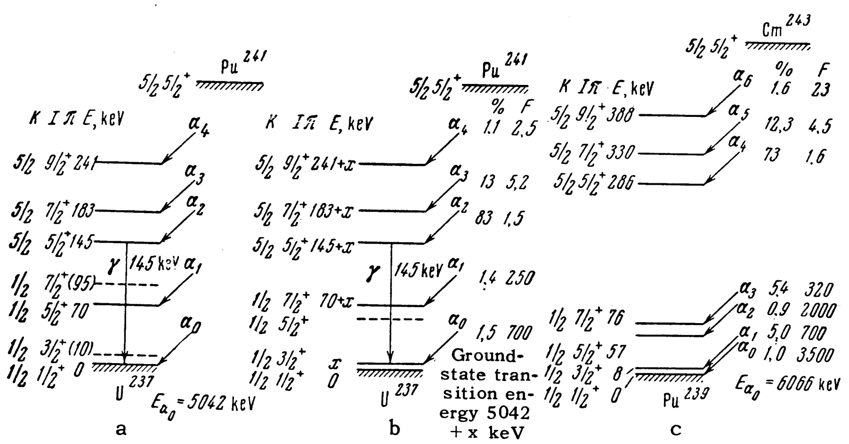


FIG. 3. a and b – two different Pu^{241} α-decay schemes; c – Cm^{243} α-decay scheme.

tional band. Indeed, $E_{rot}(9/2^+) = 96$ keV and $E_{rot}(7/2^+) = 38$ keV, giving the ratio 2.53 instead of the computed ratio 16/7 = 2.29. The moments of inertia based on these levels are $A = \hbar^2/2I$ = 5.42 keV from the $7/2^+$ level and 6.00 keV from the $9/2^+$ level.

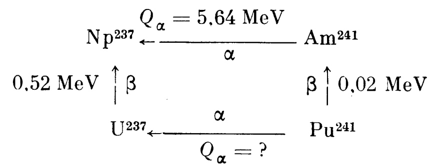
It is extremely likely that the foregoing energy levels can be accounted for by the interaction between the nuclear rotational motion and the motion of the odd nucleon. Kerman[13] has considered the interaction of levels belonging to different rotational bands of identical parity but with K differing by unity. He showed that this interaction can make the energy levels in rotational bands differ from the calculated values. In the case of U^{237} we consider the interaction of the levels $7/2^+$ (K = $5/2$ [622]) and $7/2^+$ [624], which causes a relative shift between them. The $7/2^+$[624] state is one of the closest to [622] in the Nilsson scheme. We did not observe this level experimentally, although it appears among the excited states of Pu^{239}, which resembles U^{237}, having an equal number of neutrons (N = 145).

B. Relation between the transitions α_2 and α_0

The rotational band mentioned in paragraph A is clearly not a ground state band, because the transitions α_0 and α_1 occur at higher energies than α_2. The α_2 transition is very intense (83% of the α decays) and goes to a level which should be deexcited through intense γ radiation. The only known γ line of Pu^{241} is $h\nu = 145$ keV. This γ line probably follows the α_2 group immediately; this would mean that there is a level lying 145 keV below the rotational band, and the decay energy of group α_0 actually is 145 keV above that of group α_2. Thus the groups α_2 and α_0 are consistent with the 145-keV γ rays with regard to energy.

C. Decay to the ground state of U^{237}

It is not clear from the foregoing whether the α_0 group represents decay to the ground state or to some low-lying state of U^{237}. In the latter case intense γ rays and conversion transitions should occur, but at a low transition energy they could remain unobserved in scintillation measurements. The fact that this level can have only a low excitation energy follows from consideration of a closed decay cycle. The pertinent cycle (of a 4n + 1 nucleus) is



which gives 5.14 MeV as the total decay energy of Pu^{241}, with $E_{\alpha_0} = 5.05$ MeV. This is very close to the α_0 transition energy (5042 keV), and it can therefore be assumed that this transition goes either to the ground state or to a close-lying state of U^{237}.

D. Ground state of U^{237}

From a rigorous point of view nothing is known about the ground state of U^{237}. The Nilsson scheme predicts the state $1/2^+$ [631] for the 145-th neutron. In the case of Pu^{239}, which also has 145 neutrons, this prediction is confirmed; the experimental ground-state spin is $1/2$ and the ground-state rotational band appears to have the quantum number K = $1/2$.[8] Figure 3c shows the lower portion of the $Cm^{243} \xrightarrow{\alpha} Pu^{239}$ decay scheme. The analogously constructed rotational band for U^{237} can appear either as in Fig. 3a (given by Takahashi and Morinaga[6]) or as in Fig. 3b. It should be noted that the hindrance factor for α_0 (F = 700) does not conflict with either of these two versions of the decay scheme.

The Pu^{241} α spectrum contains the still unconsidered 4973-keV transition α_1 (70 keV less decay energy than for α_0). It is reasonable to assume that, as in the decay $Cm^{243} \rightarrow Pu^{239}$, this transition goes to one of the ground-state rotational levels. The essential difference between the decay schemes of Fig. 3, a and b, lies in the fact that in the former α_1 goes to the $5/2^+$ level, while in the latter it goes to the $7/2^+$ level.

The following arguments can be advanced against Fig. 3a:

1. There is a great difference between the moment of inertia (A = 7.7 keV) of the ground-state rotational band with K = $1/2$ and that of the K = $5/2$ band (A = 6.0 keV). We recall that in the case of Pu^{239}, which resembles U^{237}, the moments of inertia for these two bands are practically equal (6.25 and 6.30 keV, respectively) although the $5/2^+$ energy level lies considerably higher (286 keV) in Pu^{239} than in U^{237} (145 keV).

2. The 145-keV γ transition occurs between $5/2^+$ and $1/2^+$ states and is thus of multipolarity E2. However, the total conversion coefficient for the 145-keV E2 transition is ~ 1.7, and the total intensity of the 145-keV transition is 24% of the number of α decays. This number is considerably smaller than the population of the 145-keV level.

3. Our measurements reveal no α transition to the 95-keV level. This means that if the transition does occur its intensity is at least one order smaller than that of the α transition to the 70-keV level. On the other hand, it follows from the data on $Cm^{243} \xrightarrow{\alpha} Pu^{239}$ α decay that decay to the $7/2^+$ level is more intense than to the $5/2^+$ level, and that decay to the $3/2^+$ level is more intense than to the $1/2^+$ level.

For the decay scheme of Fig. 3b the first objection can be removed by a suitable selection of the decoupling factor a; for a = −0.166 and $E(7/2^+) - E(3/2^+) = 70$ keV the moment of inertia is A = 6.0 keV as for the K = $5/2$ band. The second objection is removed because the 145-keV transition can be mainly M1. Then $\Sigma\alpha = 6.4$ and the total transition intensity can be as high as 67%. In $Cm^{243} \xrightarrow{\alpha} Pu^{239}$ this M1 transition is the main transition from the $5/2$ (K = $5/2$) level. The third objection disappears automatically.

It seems to us, however, that the aforementioned value a = −0.166 is not the most probable one. With this value of a, $x = E(3/2) - E(1/2)$ would have the value 15 keV, thus disagreeing with the energy balance in the foregoing cycle. By setting a = −0.620 we obtain A = 6.5 keV and $x = E(3/2^+) - E(1/2^+) = 7$ keV; for Pu^{239} we have A = 6.25 keV and x = 8 keV.

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