INVESTIGATION OF RADIOACTIVE DECAY OF Np^{237}

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Results of an investigation of the radioactive decay of Np^{237} are presented. More than 20 fine-structure monoenergetic α groups have been established by analyzing the α spectrum of Np^{237}. Most of the groups have been detected for the first time. On the basis of the expermental data obtained with the help of a β and γ spectrometer it has been possible to detect twelve γ transitions in the Pa^{233} nucleus. An energy level scheme for the Pa^{233} nucleus is constructed on the basis of the data obtained.

1. INTRODUCTION

THE radioactive isotope Np^{237}, discovered by Wahl and Seaborg[1] in the early Forties, is transformed via α decay into the β-radioactive isotope Pa^{233}. The half-life of Np^{237} is 2.2×10^6 years. The low specific activity of this isotope has hitherto made difficult precise research of the fine structure of the α radiation and of the electron spectrum. Some information on the radiation produced in the decay of this nucleus and on the levels of the daughter nucleus Pa^{233} is contained in several papers.[2-10]

We undertook a more precise study of the radioactive decay of Np^{237}, using apparatus of considerable efficiency and good resolution.

2. APPARATUS AND PREPARATION OF RADIOACTIVE SOURCES

We used in our research magnetic α and β spectrometers with double focusing of the charged-particle beam in an angle $\pi\sqrt{2}$, and also spectrometric proportional counters, a scintillation spectrometer, and other devices. The α, β, and γ spectrometers were described in our earlier articles.[11-13] We shall therefore discuss briefly only the preparation of the radioactive sources.

The sources for the α, β, and γ spectrometric measurements were prepared on the same day when the neptunium was thoroughly rid of extraneous impurities by triple chemical purification. The source for the α spectrometer was obtained by evaporating the 4 $\mu g/cm^2$ of neptunium on a glass base in vacuum. The source area was 10×100 mm. For β-spectrometer measurements we used three sources of much greater surface density; the size of the strongest source was 1×4 cm. It must be noted that the solution used to prepare the sources was also monitored by an ionization chamber with a grid and by a scintillation γ spectrometer, to check on the possible presence of other α- and β-active isotopes. Within the accuracy of instruments of this type we could observe no other α-active impurities.

3. FINE STRUCTURE OF α RADIATION OF Np^{237}

To obtain detailed information on the α structure of Np^{237} with the aid of an α spectrograph it is necessary to use a source of relatively large effective area and apparatus of high resolution. In our experiments the effective source area was 1—3 cm^2 and the minimum apparatus line width was 4.4 kev. To investigate a relatively wider region of the spectrum (~500 kev) and to carry out control experiments, several exposures with total duration of 300 hours were necessary. The energy calibration of the instrument was described by us earlier.[14] The standard line employed was the known group of other particles from Np^{237} with energy 4787.0 kev.[7]

The results of the investigation of the fine structure of the α radiation from Np^{237} are shown in Fig. 1 and in Table I.

As can be seen from Fig. 1, the α spectrum of Np^{237} is very complicated and contains more than 20 monoenergetic α lines. Some groups of α particles are exceedingly close in energy, the spacing not exceeding 2—5 kev in many cases. The conclusion that several α groups exist is therefore based on analysis of the shapes and half-widths of the α lines. Thus, for example, a comparison of the α_5 line (see the spectrum in the upper part of Fig. 1) with the groups $\alpha_{0,1,2}$, $\alpha_{6,7}$, $\alpha_{13,14}$ etc clearly indicates that the latter have a complicated composition. To confirm the data obtained we made three control experiments, which established

Table I. Fine Structure of α spectrum of Np^{237}

α_i	E_α, kev	J, %	Hindrance coefficient	E_{lev}, kev	α_i	E_α, kev	J, %	Hindrance coefficient	E_{lev}, kev
α_0	4872.$_3$	0,441	2200	0	α_{11} ?	4698,$_2$	0,067	920	177,$_1$
α_1 ?	4869.$_8$	0,925	1000	~2—3	α_{12}	4693,$_4$	0,178	320	182,$_0$
α_2	4861,$_8$	0.242	3400	~10.6	α_{13}	4663,$_0$	1,605*	20	212.$_0$
α_3	4816.$_3$	1,487	270	57,0	α_{14}	4658,$_1$	0,573*	56	217,$_3$
α_4	4802.$_3$	1,565	210	71,2	α_{15}	4638,$_4$	4.617**	5	238.$_6$
α_5	4787,$_0$	51,42	5	86.3	α_{16}	4597,$_6$	0,063	180	279,$_5$
α_6	4769,$_8$	19,38	10	104.$_2$	α_{17}	4593,$_9$	0,085	130	283,$_2$
α_7	4764,$_7$	16.82	10	109.$_5$	α_{18}	4580,$_0$	0.024	360	297.$_3$
α_8	4740,$_3$	0.019	6400	~134.$_3$	α_{19}	4572,$_7$	0.054	140	304,$_8$
α_9 ?	4711,$_3$	0.126	610	163.$_8$	α_{20}	4513,$_5$	0.01	120	~365
α_{10}	4707.$_3$	0.293	250	167.$_9$	α?	~4385,$_0$	0,02	30	~496

$^*J_{13} + J_{14} + J_Z = 2.178$.
**Sum of three lines $\alpha_X + \alpha_y + \alpha_{15}$ (see Fig. 1).

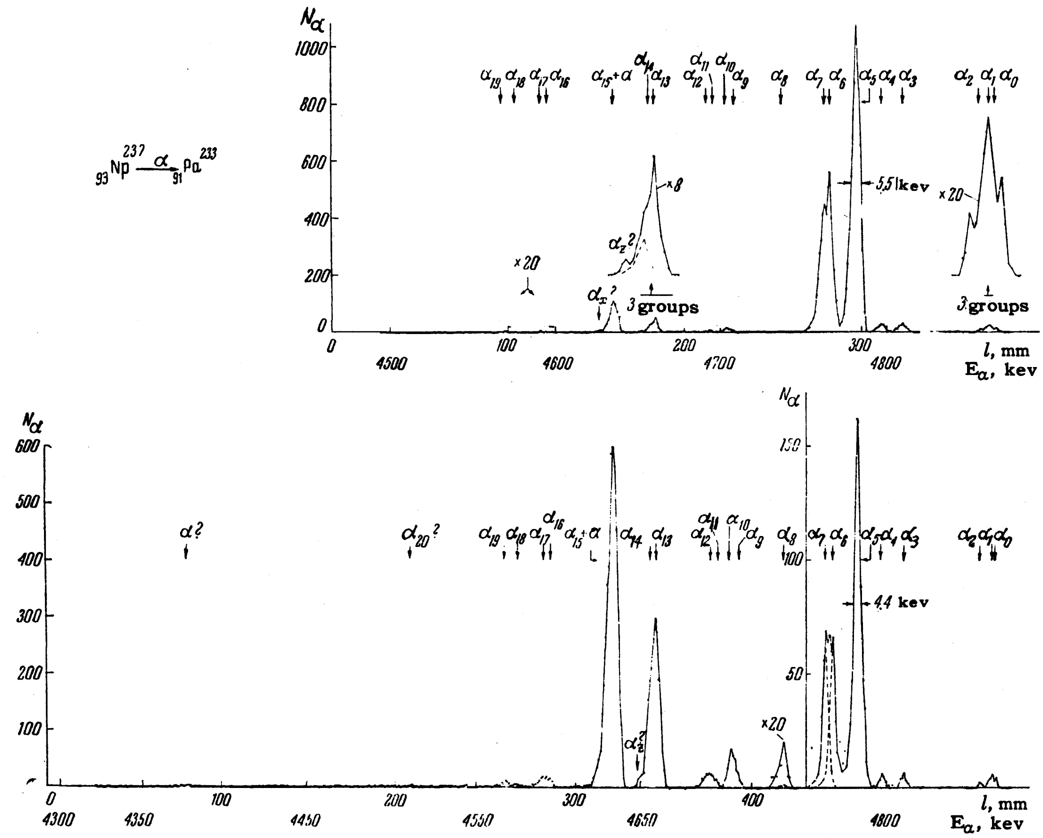

FIG. 1. Apparatus spectrum of the fine structure of α radiation of Np^{237}.

the reproducibility of the initial results. The lower half of Fig. 1 is part of the spectrum, obtained in one of the control experiments.

In the next experiment we investigated the portion of the α spectrum of Np^{237} in the energy range ~4350—4750 kev (see Fig. 1), using a wider and more intense source. The resolution of the α spectrograph was somewhat worse in this experiment than in the preceding ones. The purpose of the experiment was to confirm the series of α lines (α_9—α_{12}, α_{16}—α_{19}) with large statistical accuracy, and also to detect new ones with intensity ~0.01%.

An analysis of the plotted α spectrum has confirmed with great certainty the existence of the complex groups just mentioned, and also disclosed some other α lines (see Fig. 1).

It must be noted that the accuracy with which the energies of most individual components in the complicated groups were determined did not exceed 2 kev. It is also obvious that the intensities of these components (except α_6 and α_7) cannot be estimated accurately. Consequently the intensities listed in Table I for these lines are not exact. This inaccuracy is due primarily to the arbitrariness entailed in the graphic resolution of a complex group into individual components.

4. MEASUREMENT OF THE ELECTRON AND γ SPECTRA

The electron and γ spectra were investigated in order to detect new γ transitions in Pa^{233} and to determine the class of multipolarity of some of the more intense γ transitions. Measurements of the electron and γ spectra were made directly after radiochemical elimination from the Np^{237} of the daughter Pa^{233}, and therefore the activity of the Pa^{233} was small and did not affect the measurement results.

The β spectrometer used to investigate the electron spectrum permitted the use of sources with relatively large effective areas ($S_{max} = 4$ cm^2). However, the slight specific activity of Np^{237} ($T_{1/2} = 2.2 \times 10^6$ years) did not make it possible to obtain a source of required intensity with small surface density.

The low activity and the large thickness of the sources did not allow us to plot the electron spectrum with high degree of resolution. The spectra obtained in three different experiments, however, were identical and enabled us to separate reliably the conversion lines characterizing the γ transitions in Pa^{233}. In the analysis of the electron spectrum we took into account the measured γ spectrum, and also the data on the investigation of the α radiation of Np^{237}.

Table II summarizes some of the data on the γ transitions, obtained from the analysis of the electron and γ spectra for Pa^{233}.

The determination of the multipolarity of most γ transitions in Pa^{233} is impossible because of the low intensity and insufficient resolution of the conversion-electron lines. A comparison of the data obtained with a proportional counter, a β spectrometer, and a γ spectrometer enabled us to establish the multipolarity only for the 29 kev (E1), 57 kev (E2), and 86 kev (E1) γ transitions. The results obtained do not contradict the data of Magnusson et al.[7]

Table II. Energy (kev) of γ transitions in Pa^{233}

β spectrometer	Proportional counter	γ spectrometer	Multipolarity
29.6	29.6	30	$E1$
55.0?	—	—	$E2$?
56.8	57÷58	57÷60	$E2$
84.5? }	84—90	—	$E1$
85.9 }			
105.0	~104	} 106	—
108.4	~108		
133.5	—	} 130÷145	—
144.5	—		
?	—	~160?	—
207	—	—	—
240	—	~250	—

It is mentioned in the literature[15] that an anomalously high internal-conversion coefficient was obtained for 86-kev γ rays. The possible existence of an 84-kev γ transition can apparently change this anomaly.

5. ENERGY LEVELS OF Pa^{233}

The most complete among the energy level schemes listed in the literature[7,16-18] for Pa^{233} is found in the book by Mottelson and Nilsson[18]. This scheme is based on the data of an unpublished paper by Asaro, Perlman, and Stephens devoted to a study of the α decay of Np^{237} (see also [19]), and the known Nilsson diagrams. Our new data make it possible to supplement this scheme.

An analysis of the experimental data enables us to state that the Pa^{233} nucleus has more than 20 energy levels in a relatively narrow energy interval (0—400 kev). So large a number of levels indicates that this nucleus has several rotational bands with different values of K (spin projection on the symmetry axis of the nucleus). Of course, one cannot exclude the existence of levels belonging to bands with different K but having the same spins and parities. An "interaction of rotational levels"[20], corresponding to different states of the nucleus $K[N, n_z, \lambda]$, $K'[N', n'_z, \lambda']$ etc, should therefore take place in this nucleus. This may violate O. Bohr's interval rule for rotational bands and, in accordance with[21], probably causes a considerable increase (or decrease) in the intensities of some α transitions.

The data given above were used to construct the energy level scheme for Pa^{233} shown in Fig. 2. On the left side of this figure is the Nilsson diagram; the vertical dashed line on the diagram corresponds to $\delta = 0.24$, a value customarily used for Pa^{233}.[18] If we ascribe to the ground state of this nucleus an orbit $1/2^-$ [530] (see [18,22]), then, in accordance with this diagram, the final levels $5/2^+$ [642], $5/2^-$ [523], $3/2^-$ [521], and $7/2^+$ [633] should be observed. As indicated by Mottelson and Nilsson[18], the existence of a "hole" level, with characteristics $3/2^+$ [651] is likewise not excluded, from the point of view of the Nilsson diagram.

The ground state of Pa^{233} has a spin $3/2$[23] and

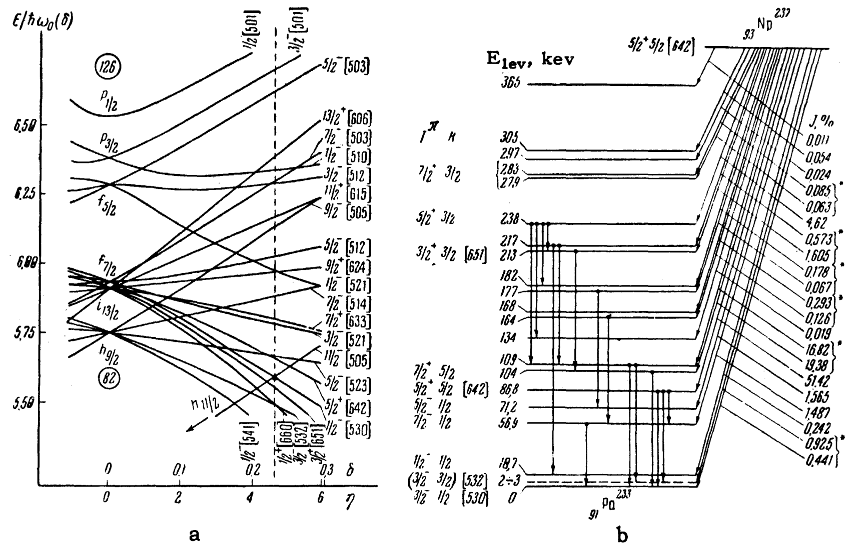

FIG. 2. Energy-level scheme of the Pa^{233} nucleus; a – Nilsson diagram for odd Z, b – experimentally obtained level scheme of Pa^{233}. Levels due to α_x, α_y, and α_z transitions are not indicated.

negative parity.[24] The levels with energies 51 and 71 kev[18,22] and the ~11-kev level observed in our experiments apparently belong to the main rotational band ($3/2^-$ $1/2$ [530]) with a spin sequence $3/2^-$, $1/2^-$, $7/2^-$, $5/2^-$, etc. It must be noted that although there is no doubt of the existence of the α_2 group, nonetheless the energy of the level (E_{lev} $\approx$ 11 kev) has not been determined with sufficient accuracy.

The next levels of this rotational band, with characteristics $11/2^-$ and $9/2^-$, can apparently be the 164- and 177-kev levels, respectively, but there is no rigorous proof of the correctness of this statement.

The favored transition α_5 actually gives grounds for assuming[18,22] that the single-particle 87-kev level can be assigned an orbit $5/2^+$ $5/2$ [642], so long as the ground state of Np^{237} is $5/2^+$ $5/2$ [642]. The members of this rotational bands are apparently the levels at 87 kev ($5/2^+$ $5/2$ [642]), 104 kev ($7/2^+$), 168 kev ($9/2^+$), and 249 kev ($11/2^+$). However, the assumption that the last two levels belong to the indicated rotational band is hardly correct, in view of the indicated interaction. It is not excluded that the third member of the rotational band referred to here is a level with energy 109 kev, since the intensity of the α_7 group, which goes to this level, is high.

As indicated above, the existence of a 'hole' level with characteristics $3/2^+$ [651] is expected from the point of view of the Nilsson diagram. The 213-kev level apparently confirms this assumption. It is possible that one of the lines of the complex groups ($\alpha_{15} + \alpha_y$ and $\alpha_{16} + \alpha_{17}$) determines accordingly the positions of the next levels ($5/2^+$ and $7/2^+$) of the new rotational band.

Analysis of the complex line group (α_0, α_1, α_2) (see Fig. 1) apparently indicates that there exists still another level with energy 2–3 kev. Naturally, the lines α_0 and α_1 are not sufficiently well separated and a certain arbitrariness in the interpretation of the experimental results can therefore not be excluded. We do not insist that this level exists. If it does, however, we must ascribe to it either an orbit $5/2^-$ $5/2$ [523] or $3/2^-$ $3/2$ [532].

The interpretation we presented for most Pa^{233} levels is quite arbitary. We cannot pretend that the proposed level scheme of this nucleus is complete or final.

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[1] A. C. Wahl and G. T. Seaborg, Phys. Rev. **73**, 940 (1948).

[2] J. Mack, Revs. Modern Phys. **22**, 64 (1950).

[3] D. Dunlavey, and G. Seaborg, Phys. Rev. **87**, 165 (1952).

[4] D. Engelkemeir, and L. Magnusson, Phys. Rev. **94**, 1395 (1954).

[5] Bleaney, Llewellyn, Pryce, and Hall, Phil. Mag. **45**, 992 (1954).

[6] D. Strominger, and J. Rasmussen, Phys. Rev. **100**, 844 (1955).

[7] Magnusson, Engelkemeir, Friedman, Porter, and Wagner, Phys. Rev. **100**, 1237 (1955).

[8] R. Thorne, Nature **178**, 484 (1956).

[9] Dobbs, Roberts, and Parker, Bull. Am. Phys. Soc. **1**, 207 (1956).

[10] Kondrat'ev, Novikova, Vorob'ev, and Gol'din, Izv. AN SSSR, ser. fiz. **20**, 875 (1956), Columbia Tech. Transl. p. 795.

[11] Baranov, Zelenkov, Shchepkin, Beruchko, and Malov, Atomnaya énergiya **7**, 262 (1959); Izv. AN SSSR ser. fiz. **23**, 1402 (1959), Columbia Tech. Transl. p. 1389.

[12] P. S. Samoilov, PTÉ (Instrum. and Exptl. Techniques) No. 6, 33 (1959).

[13] Baranov, Rodionov, Shishkin, and Chistyakov, JETP **34**, 1367 (1958), Soviet Phys. JETP **7**, 946 (1958).

[14] Baranov, Zelenkov, and Kulakov, Izv. AN SSSR ser. fiz. **24**, 1035 (1960), Columbia Tech. Transl. p. 1045.

[15] Asaro, Stephens, Hollander, and Perlman, Phys. Rev. Lett. **2**, 442 (1960).

[16] B. S. Dzhelepov and L. K. Peker, Skhemy raspada radioaktivnykh yader (Decay Schemes of Radioactive Nuclei), AN SSSR, 1958.

[17] D. Strominger and J. M. Hollander, UCRL-8289, Berkley, California (1958).

[18] B. Mottelson, and S. Nilsson, Mat-Fys. Scr. Dan. Vid. Selsk. **1**, 8 (1959).

[19] I. Perlman, and J. O. Rasmussen, Handbuch d. Physik **42**, 109 (1957).

[20] A. Kerman, Mat-Fys. Medd. Kyldaske Vid. Selsk. **30**, 15 (1955).

[21] O. Prior. Ark. Fysik. **16**, 15 (1959).

[22] Stephens, Asaro, and Perlman, Phys. Rev. **113**, 212 (1959).

[23] J. Hubbs and J. Winicur, Bull. Am. Phys. Soc. **11**, 319 (1958).

[24] Hamilton, Hollander, and Petterson, UCRL 9438, Berkley, California (1960).

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