

FINE STRUCTURE OF Pu<sup>239</sup> ALPHA RADIATION

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The radioactive decay of Pu<sup>239</sup> was investigated with a magnetic  $\alpha$  spectrograph. Analysis of the Pu<sup>239</sup> spectrum revealed the existence of more than 20 fine-structure  $\alpha$  groups, many of which were detected for the first time. The U<sup>235</sup> energy-level scheme is presented, with discussion of a possible interpretation indicating the existence of a new band with  $\frac{5}{2}^+[633]$  base state.

## INTRODUCTION

NILSSON'S well-known theoretical schemes of single-particle states in odd-mass heavy elements ( $N > 126$ ) can be used to predict many U<sup>235</sup> levels qualitatively. Earlier investigations of Pu<sup>239</sup>  $\alpha$  decay<sup>[1-6]</sup> revealed fine-structure  $\alpha$  lines representing transitions to U<sup>235</sup> levels. The experimental data indicated the existence of two rotational bands, which will be discussed below. The other theoretically predicted levels have hitherto not been observed. We therefore undertook a thorough investigation of Pu<sup>239</sup>  $\alpha$  decay in order to detect new  $\alpha$  groups and to construct a more complete level scheme for U<sup>235</sup>.

We used a double-focusing ( $\pi\sqrt{2}$ )  $\alpha$  spectrograph<sup>[7,8]</sup> possessing good resolution and transmission.

## 1. PREPARATION OF RADIOACTIVE SOURCES

After careful chemical purification of plutonium samples having different isotopic concentrations of Pu<sup>239</sup> and Pu<sup>240</sup>, sources for  $\alpha$ -spectrograph study were prepared by plutonium chloride evaporation in vacuo on glass plates and very thin Al<sub>2</sub>O<sub>3</sub> films. Five sources having different effective areas were prepared ( $S_{\max} = 3 \times 100$  mm,  $S_{\min} = 0.5 \times 50$  mm). The active surface densities on the two extreme areas were 5 and 0.6  $\mu\text{g}/\text{cm}^2$ , respectively; the film thickness did not exceed 0.05  $\mu$ . The use of these films as a backing for the sources made it possible to exclude spurious  $\alpha$  groups resulting from the diffusion of plutonium atoms into the backing.

## 2. EXPERIMENTAL RESULTS

We confined our investigation of the Pu<sup>239</sup> spectrum to fine-structure groups in the energy range 4600-5200 keV with relative intensities  $\eta \geq 2$

$\times 10^{-6}$ . The 5495.0-keV  $\alpha_0$  group of Pu<sup>238</sup> was used as a standard.<sup>[9]</sup>

Figure 1 shows  $\alpha$ -ray spectrograms of Pu<sup>239</sup> obtained in three different runs using samples having different concentrations of Pu<sup>239</sup> and Pu<sup>240</sup>. The lower right-hand portion of the figure shows the three most intense Pu<sup>239</sup>  $\alpha$  groups and the two main groups ( $\alpha_0^{40}$  and  $\alpha_1^{40}$ )<sup>1)</sup> of Pu<sup>240</sup>. Here the minimum  $\alpha$ -line half-width is 2.3 keV (exposure time 25 hr), permitting a very precise determination of group intensities as well as the calculation of the energy difference  $E(\alpha_0^{40}) - E(\alpha_0^{39}) = 12.10 \pm 0.15$  keV. The lower left-hand portion of the figure shows the  $\alpha$  spectrum in the range 4600-5080 keV obtained from a stronger source backed with an Al<sub>2</sub>O<sub>3</sub> film.

More than 17 weak groups were observed in the last-mentioned energy range; some of these evidently have a complex structure. Control runs were performed to check the reproducibility of individual lines and to refine the structure of some of them. A portion of the spectrum obtained in one of these runs is shown in the upper part of Fig. 1. A comparison of the last two spectra confirms the complex structure of some  $\alpha$  lines. The source used in the last case differed in its plutonium-isotope composition.

The data obtained in five runs are shown in the accompanying table, which gives the energies and intensities of individual Pu<sup>239</sup>  $\alpha$  groups, the calculated hindrance factors  $m$  and the U<sup>235</sup> energy levels.

The  $\alpha$ -group energies in the table differ considerably from those given by other authors<sup>[1-6]</sup> ( $\Delta E = 7-10$  keV). The discrepancies are associated with the selection of a "standard energy." For example, Rytz<sup>[10]</sup> recently determined the

<sup>1)</sup>The intensities of these groups were 75.3 and 24.7%, respectively.

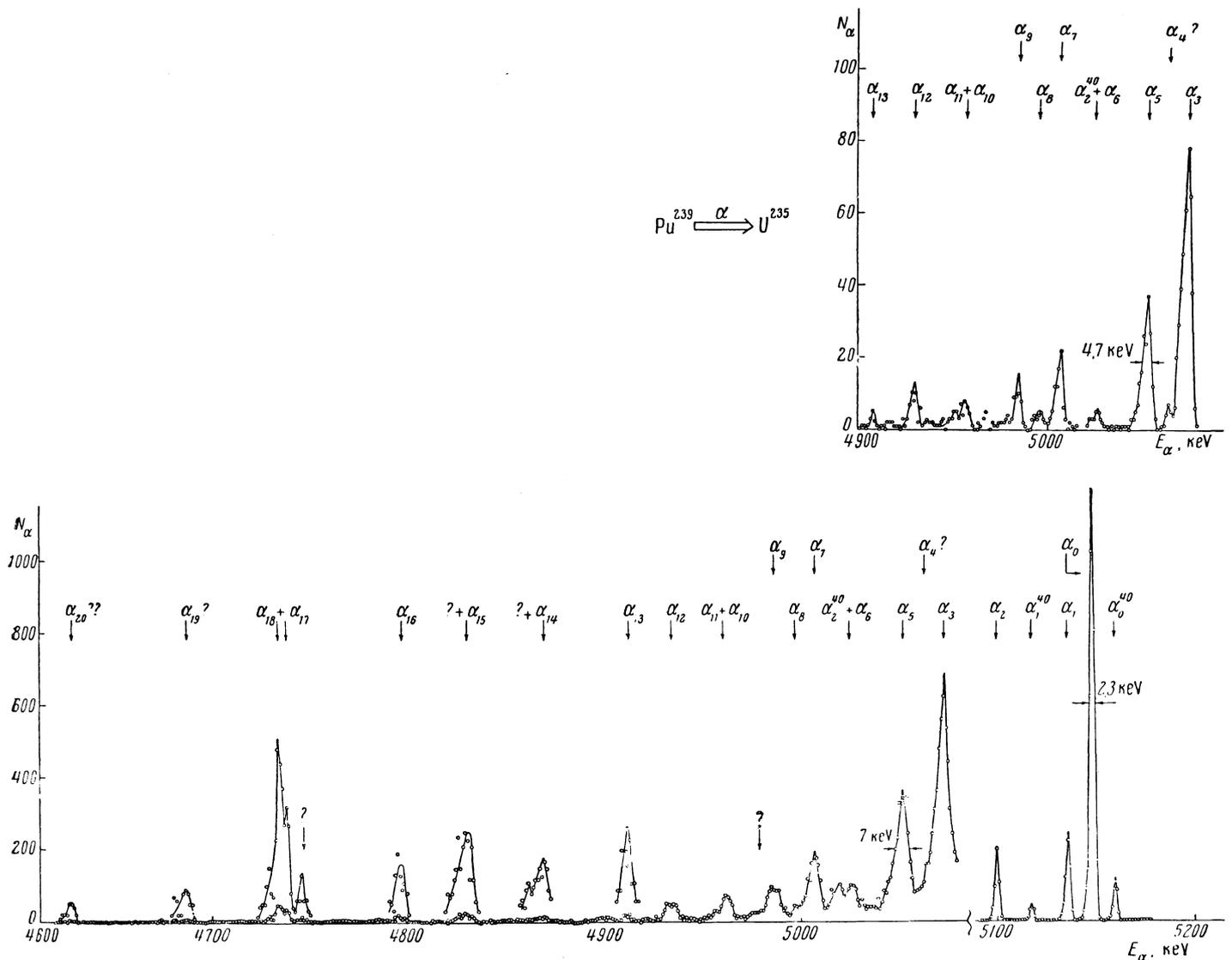


FIG. 1.  $\alpha$ -ray spectrograms of  $\text{Pu}^{239}$ . Superscripts denote  $\text{Pu}^{240}$   $\alpha$  groups.

absolute energy of  $\text{Po}^{210}$   $\alpha$  particles as  $E_{\alpha_0} = 5304.81 \pm 0.62$  keV, whereas several other investigators<sup>[2,3]</sup> used as the standard value  $E_{\alpha_0}(\text{Po}^{210}) = 5298.0$ .<sup>[11]</sup> We avoided the use of a  $\text{Po}^{210}$  source in our work for several reasons.

### 3. ENERGY LEVELS OF $\text{U}^{235}$

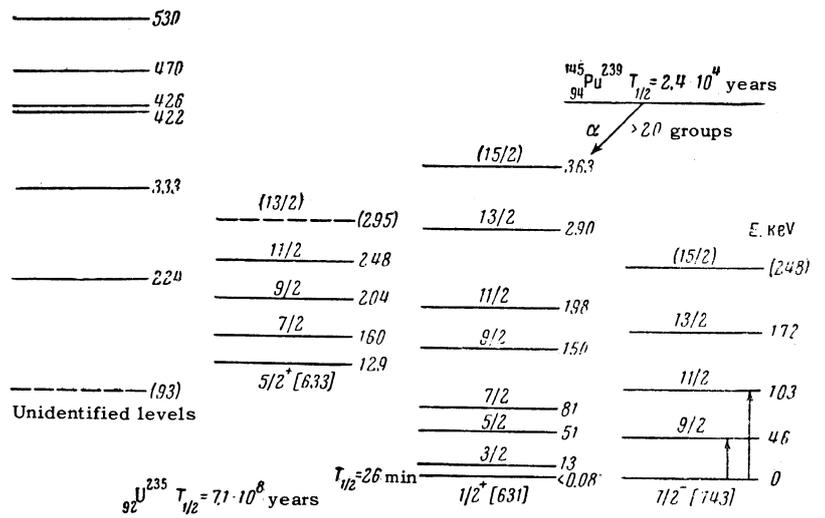
The most complete level schemes of  $\text{U}^{235}$  have been given by Perlman and Rasmussen,<sup>[5]</sup> Dzhele-pov et al,<sup>[6]</sup> Tret'yakov et al,<sup>[12]</sup> and Mottelson and Nilsson.<sup>[13]</sup> Our data enable the construction of a considerably more complete level scheme.

The level scheme of  $\text{U}^{235}$  based on our data is shown in Fig. 2. The vertical arrows indicate levels obtained by Newton<sup>[14]</sup> from Coulomb excitation of  $\text{U}^{235}$ . Let us examine the proposed

scheme in detail. The  $\text{U}^{235}$  ground state has spin  $7/2$  and negative parity,<sup>[15,16,13]</sup> and according to Nilsson's scheme for odd N can be assigned to a  $7/2^- 7/2 [743]$  orbital. The  $\sim 46$ - and  $\sim 103$ -keV levels observed by Newton<sup>[14]</sup> have spins and parities  $9/2^-$  and  $11/2^-$ , respectively, and belong to the indicated ground-state rotational band of  $\text{U}^{235}$ . This band also possibly includes the levels 172 keV ( $13/2^-$ ) and 248 keV ( $15/2^-$ ).

Some comments regarding this band are in order. First, it must be noted that the  $\alpha$  transition to the  $\text{U}^{235}$  ground state in our work could not be discriminated from the  $\alpha_0$  transition to an isomeric state of  $\text{U}^{235}$  ( $E < 80$  keV,  $T_{1/2} = 26$  min)<sup>[17-19]</sup> having the characteristics  $1/2^+ 1/2 [631]$  similar to those of the  $\text{Pu}^{239}$  ground state. Secondly, we were unable to detect an  $\alpha$  group cor-

FIG. 2. Energy levels of U<sup>235</sup>. The energies and intensities of α transitions to these levels are given in the table.



responding to a transition to the ~ 46-keV level (I = 9/2<sup>-</sup>) which had been observed by Newton,<sup>[14]</sup> despite the favorable conditions of our control run. We estimate the intensity of the α transition to the 46 keV level as less than 3 × 10<sup>-2</sup>%, whereas the transition to 103 keV (I = 11/2<sup>-</sup>, η ≈ 2 × 10<sup>-2</sup>%) is seen clearly in Fig. 1 (α<sub>5</sub>). Finally, although a calculation based on the Bohr-Mottelson formula for the ground-state rotational band with K = 7/2 predicts levels at about 172 keV (I = 13/2<sup>-</sup>) and 248 keV (I = 15/2<sup>-</sup>), the excessive quantum differences between the Pu<sup>239</sup> ground state (I = 1/2<sup>+</sup>) and the U<sup>235</sup> states I<sub>i</sub> = 13/2<sup>-</sup>, 15/2<sup>-</sup> hardly permits us to associate these two groups with transitions to levels of the ground-state rotational band 7/2<sup>-</sup> 1/2<sup>-</sup> [743].

We shall show subsequently that the α group associated with a transition to 248 keV can be assigned more properly to transitions going to a level of our newly discovered rotational band 5/2<sup>-</sup> [633].

As indicated above, the α transition to a level < 80 eV (U<sup>235m</sup>) is favored; we have the assignment 1/2<sup>+</sup> 1/2<sup>-</sup> [631] for this level. The rotational band based on this state is highly developed and extends to a 15/2<sup>+</sup> level (Fig. 2). The energies, spins, and parities of levels in this band with K = 1/2 are < 80 eV (I = 1/2<sup>+</sup>), and 13 (3/2<sup>+</sup>), 51 (5/2<sup>+</sup>), 81 (7/2<sup>+</sup>), 150 (9/2<sup>+</sup>), 198 (11/2<sup>+</sup>), 290 (13/2<sup>+</sup>), and possibly 363 keV (15/2<sup>+</sup>). Our results relative to this band do not disagree with those given by other authors.<sup>[6,12,19]</sup> A certain easily noted discrepancy in the energy levels evidently lies within experimental error.

Figure 2 shows that the energy range 120–300 keV also includes levels at 129, 160, 204, 248, and 295 keV. The first of these levels can be assigned by Newton's scheme to either 5/2<sup>+</sup> 5/2<sup>-</sup> [622] or 5/2<sup>+</sup> 5/2<sup>-</sup> [633]. If our hypothesis regarding this level is correct, the last members of this new rotational band can be the additional levels given above with

Fine structure of Pu<sup>239</sup> α spectrum

α-group	α-particle energy, keV*	Intensity, %	Hindrance factor, m	Level energy, keV	α-group	α-particle energy, keV	Intensity, %	Hindrance factor, m	Level energy, keV
α <sub>0</sub>	5157	73.3	3	0	α <sub>11</sub>	4957	5 · 10 <sup>-4</sup>	25000	204
α <sub>0</sub>				< 80 eV	α <sub>12</sub>	4937	3 · 10 <sup>-3</sup>	3200	224
α <sub>1</sub>	5145	15.1	12.8	12.7	α <sub>13</sub>	4914	8 · 10 <sup>-4</sup>	7800	248
α <sub>2</sub>	—	< 3 · 10 <sup>-2</sup>	—	~ 46	α <sub>14</sub>	4873	7 · 10 <sup>-4</sup>	4900	290
α <sub>3</sub>	5107	11.5	9.4	51.0	α <sub>15</sub>	4868	—	—	~ 295
α <sub>4</sub>	5078	3.2 · 10 <sup>-2</sup>	2100	81	α <sub>16</sub>	4830	1.5 · 10 <sup>-3</sup>	1200	333
α <sub>5</sub>	5066	9 · 10 <sup>-4</sup>	65000	93	α <sub>17</sub>	4801	6 · 10 <sup>-4</sup>	1800	363
α <sub>6</sub>	5056	2.1 · 10 <sup>-2</sup>	2500	103	α <sub>18</sub>	4743	2.6 · 10 <sup>-3</sup>	—	422
α <sub>7</sub>	5031	5 · 10 <sup>-3</sup>	6900	129	α <sub>19</sub>	4739	—	—	426
α <sub>8</sub>	5010	8 · 10 <sup>-3</sup>	3100	150	α <sub>20</sub>	4695	4 · 10 <sup>-4</sup>	500	470
α <sub>9</sub>	5001	6 · 10 <sup>-4</sup>	40000	160		4636	2 · 10 <sup>-4</sup>	300	530
α <sub>10</sub>	4988	5 · 10 <sup>-3</sup>	4100	172					
	4963	3 · 10 <sup>-3</sup>	4100	198					

\*The 5495.0-keV α group of Pu<sup>238</sup> was used as a standard.<sup>[9]</sup> The table does not give the energies of the Pu<sup>239</sup> α<sub>0</sub> and α<sub>1</sub> groups, which are 51695 and 51259 keV, respectively.

spins and parities  $7/2^+$ ,  $9/2^+$ ,  $11/2^+$ , and  $13/2^+$ (?). The value of  $\hbar^2/2J$  for this band is  $\sim 4.4$  keV.

The number of excited  $U^{235}$  states is not confined to the foregoing. Analysis of the  $Pu^{239}$   $\alpha$  spectrum indicates the presence of  $\alpha$  groups representing transitions to 93 (?), 224, 333, 422, 426, and 530 keV (Fig. 2). These levels cannot be identified at the present time. The experimental data also suggest the existence of several  $\alpha$  lines having intensities  $< 2 \times 10^{-6}$ , which are not included in the table.

Some  $\alpha$  groups ( $\alpha_9$ ,  $\alpha_{14}$ , and  $\alpha_{15}$  in Fig. 1) are evidently sums of at least two components. It is therefore possible that the  $U^{235}$  scheme should include a few levels not shown in Fig. 2. Confirmation of the last two statements will depend on further experimental work.

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