abundance or greater. The discrepancy between the abundance of the  $\sim$ 120-kev transition and the absence of a corresponding alpha-group is too close to the limits of error in measurement to be considered real at this stage. If it should prove to be real, this may be taken

as evidence that the ground-state transition for the alpha-decay of Pu<sup>239</sup> has not yet been seen.

We wish to acknowledge the assistance of Miss Beverly Turner and Mr. James Vanderveen in counting the alpha-tracks.

PHYSICAL REVIEW

VOLUME 88, NUMBER 4

NOVEMBER 15. 1952

# Radioisotopes of Osmium\*

J. B. SWAN<sup>†</sup> AND R. D. HILL Physics Department, University of Illinois, Urbana, Illinois (Received July 30, 1952)

<sup>1</sup> Osmium activities of 15-day and 32-hour half-lives have been identified with Os<sup>191</sup> and Os<sup>193</sup>, respectively. A 14-hour activity, decaying by a 74.2-kev gamma-transition into the 15-day activity, has been assigned to an isomeric state in  $Os^{191}$ . The decay scheme of  $Os^{191}$  has been investigated and a 7/2+ assignment has been made to the 15-day ground state and an  $i_{13/2}$  assignment to the 14-hour isomeric state. The 7/2+ state is attributed to a coupling of an odd number of  $i_{13/2}$  neutrons.

#### INTRODUCTION

SMIUM has seven stable isotopes, with mass numbers from 184 to 192, except for vacancies at 185 and 191. Three radioisotopes have so far been observed, and these have been allocated to mass numbers 185, 191 and 193.<sup>1-13</sup> We report in the present paper a new activity and a revised assignment of two of the already known activities.

#### Os191 AND Os191m: EXPERIMENTAL OBSERVATIONS

Ordinary osmium, in powdered metallic form, was irradiated by neutrons in the heavy water pile at Argonne. The internal conversion electron spectrum, measured with 180° magnetic spectrographs, showed three groups of conversion lines. Accurate energy determinations, which are given in Table I, indicate that the gamma-transitions of 41.7 and 129.1 key take place in iridium and that the 74.2-kev gamma-transition occurs in osmium.

- <sup>6</sup> Cork, Schreffler, and Fowler, Phys. Rev. 72, 1209 (1947).
  <sup>6</sup> L. I. Katzin and M. Pobereskin, Phys. Rev. 74, 264 (1948).
  <sup>7</sup> D. Saxon, Phys. Rev. 74, 1264 (1948); Atomic Energy Commission Report, AECD, 1860 (1948) (unpublished).
  <sup>8</sup> Mandeville, Sherb, and Keighton, Phys. Rev. 74, 888 (1948).
  <sup>9</sup> Bunker, Canada, and Mitchell, Phys. Rev. 80, 126 (1950);
  79, 610 (1950).
- <sup>10</sup> F. K. McGowan, Oak Ridge National Laboratory Report, ORNL-952 (1951) (unpublished); Phys. Rev. 79, 404 (1950).
   <sup>11</sup> T. C. Chu, Phys. Rev. 79, 582 (1950).
   <sup>12</sup> E. Kondaiah, Ark. Fys. 3, 47 (1951).
   <sup>13</sup> Wedger, Data Mathematical National Burgau of Standards, Circular National Strength (1974).

<sup>13</sup> Nuclear Data, National Bureau of Standards, Circular No. 499 (1950), p. 222.

Lifetime tests on the individual conversion lines showed that there was an initial rise in the intensity of the 41.7- and 129.1-kev lines, which changed over after a period of several days into a constant decay of 15-day half-life. On the other hand, the 74.2-kev transition lines decayed throughout with a half-life of  $14\pm 2$ hours. A set of decay curves for a particular source, which was bombarded one day in a pile, is shown in Fig. 1. It is clear that the growth of the 41.7- and 129.1kev transition intensities is consistent with a partial production of the 15-day activity from a parent of 14-hour half-life.

Osmium was also bombarded by gamma-rays from a 22-Mev betatron, using an internal target arrangement.<sup>14</sup> A conversion spectrum the same as, although weaker than, that from a pile irradiated source was obtained. The 15-day activity is therefore to be identified with mass number 191, instead of 193, and the 14-hour activity is thus an isomer of Os<sup>191</sup>. The present assignment of the 15-day activity is in accord with beta-disintegration energy systematics, as pointed out by Way.15, ‡

TABLE I. Energies in kev of electron conversion lines from Os<sup>191</sup> and Os<sup>191m</sup> activities.

Gamma-	Sub-shell origin of conversion electrons								
transition	Κ	$L_{\mathrm{I}}$	$L_{\mathrm{II}}$	$L_{\mathrm{III}}$	$M_{I}$	$M_{\rm II}$	$M_{\rm III}$	N	
41.70 kev 74.17 kev		 61.24	28.85 61.78	30.48 63.36	 71.09	38.80	39.19 71.72	41.15 73.45	
129.1 kev	53.15	115.49	116.14	117.97		126.29		128.76	

<sup>14</sup> R. A. Becker, Rev. Sci. Instr. 22, 773 (1951).

made on the basis of  $\gamma$ -irradiations.

<sup>15</sup> K. Way, private communication and reference 13. <sup>1</sup> Note added in proof:—A revision of the original Os<sup>191</sup>, Os<sup>193</sup> assignment by Seaborg and Friedlander (see reference 3) was later made by G. Friedlander (quoted in *Table of Isotopes*, Seaborg and Perlman, Revs. Modern Phys. **20**, 585 (1948)). According to J. M. Hollander (private communication) this revision was also

<sup>\*</sup> Assisted by the joint ONR and AEC program.

<sup>†</sup> Fulbright and University of Illinois Postdoctoral Fellow. On leave from University of Western Australia, Perth, Australia.

<sup>&</sup>lt;sup>1</sup>Kurtchatow, Latyschew, Nemenow, and Selinow, Physik Z. Sowjetunion 8, 589 (1935). <sup>2</sup> É. Zingg, Helv. Phys. Acta 13, 219 (1940)

<sup>&</sup>lt;sup>3</sup>G. T. Seaborg and G. Friedlander, Phys. Rev. 59, 400 (1941). <sup>4</sup> L. J. Goodman and M. L. Pool, Phys. Rev. **70**, 112 (1946); **71**, 288 (1946).

<sup>&</sup>lt;sup>5</sup> Cork, Schreffler, and Fowler, Phys. Rev. 72, 1209 (1947).



FIG. 1. Decay curves of conversion lines from the 74-kev gamma-transition of  $Os^{191m}$  and the 129- and 42-kev gammatransitions of Os<sup>191</sup>. Bombardment time of the source was one day. Zero time of the abscissa corresponds to the end of the bombardment.

In order to investigate whether there was any betadecay associated with the 14-hour activity, an osmium source prepared by gamma-irradiation was studied using a lens spectrometer. The spectrum observed several hours after an 8-hour irradiation in the 22-Mev betatron is shown in Fig. 2. We conclude that direct beta-decay from the 14-hour isomeric level in Os does not occur to the extent of 5 percent, if at all, of the isomeric transition intensity from this level.

Investigations of the beta-spectrum of the 15-day Os<sup>191</sup> activity were also made with lens and 180° spectrometers. However, our results, on account of the too-thick sources, did not lead to accurate values of the conversion coefficient of the 42- and 129-kev radiations. We are essentially in agreement with other workers<sup>7,9,12</sup> that there is a beta-spectrum with an upper end point in the neighborhood of 140 kev.

TABLE II. Relative intensities of conversion lines from Os<sup>191</sup> and Os<sup>191m</sup> activities.

transition	K	$L_{I}$	$L_{\mathrm{II}}$	$L_{III}$	$M_{I}$	$M_{\rm II}$	$M_{III}$	N
41.70 kev		•••	32	40		11	18.5	9.5
129.1 kev 74.17 kev	100	$\substack{29.5\\42}$	$10.5 \\ 23.5$	$\begin{array}{c} 6.0 \\ 100 \end{array}$	14	11.5	35	3.5 15

An important question to be resolved in connection with the decay scheme of Os<sup>191</sup> is whether the 42- and 129-kev transitions decay in cascade or parallel. Experiments to show whether gamma- or x-rays were or were not in coincidence were inconclusive. However, more elaborate experiments using a double slit 180° beta-ray spectrometer<sup>16</sup> showed that the 42-L and 129-L conversion electrons were definitely in coincidence.

The relative intensities of the 74.2- 41.7-, and 129.1-kev transition conversion lines were determined photographically and their values are given in Table II.

#### Os191: DISCUSSION

The multipolarities of the 129.1- and 41.7-kev transitions have been obtained from the following arguments.

Kondaiah's value<sup>12</sup> of the ratio of the L-conversion line intensity to the total Os<sup>191</sup> beta-intensity, in conjunction with the conversion line intensities of Table II, yields a value of 1.36 for the K-conversion coefficient of the 129-kev transitions. Using Rose's tables,<sup>17</sup> this value is in best agreement with an 0.42:0.58 mixture of M1:E2 multipole radiations. Our value of 2.2 for the K/L ratio of the 129-kev transition agrees excellently with Kondaiah's value of 2.1, and using the empirical K/L ratios of Goldhaber and Sunyar,<sup>18</sup> the ratio of the M1: E2 mixture is 0.64: 0.36. The ratios of the intensities of the  $L_{I}: L_{II}: L_{III}$  components of the 129-L conversion line, using Gellman's<sup>19</sup> unscreened theoretical L-shell conversion calculations, indicate an 0.8:0.2 mixture of M1:E2.

The multipolarity of the 42-kev transition is more difficult to obtain. Qualitatively, however, both our own scintillation counter experiments and photoelectron spectrometer studies of the 15-day Os<sup>191</sup> show that the 42-kev transition is highly converted. Beta-conversion electron coincidence experiments<sup>12</sup> also show that the 42-kev gamma transition is certainly of multipole order less than three. One possibility of determining the multipolarity of this transition is through a comparison of theoretical and experimental  $L_{I}: L_{II}: L_{III}$ ratios. According to Gellman's analysis<sup>19</sup> these ratios are as follows: 1.0:0.68:1.0 for E1, 0.03:0.90:1.0 for E2, and 1.0:0.065:0.011 for M1. The experimental ratios are 0.0:0.81:1.0, which agree well with pure E2. An assignment of M2, for which there are no theoretical data, can be ruled out on beta- and gamma-transition considerations. The theoretical<sup>19</sup> total L-shell conversion coefficient of a 42-kev E2 transition is  $\sim 200$ .

If we conclude that the 129-kev transition is a 3:1 mixture of M1:E2 and the 42-kev transition is pure E2, we find that the total intensity of the 129-kev

 <sup>&</sup>lt;sup>16</sup> Katz, Hill, and Goldhaber, Phys. Rev. **78**, 9 (1950).
 <sup>17</sup> Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951); Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023 (1951) (unpublished).
 <sup>18</sup> M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
 <sup>19</sup> Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).

transition is approximately twice the 42-kev transition intensity. Smaller mixtures of M1 in the 129-kev transition would necessarily increase the relative intensity of this transition.

Since it has been shown that the two transitions are in cascade it must be concluded that the 42-kev transition precedes the 129-kev transition. In this case, since the ground state  $Ir^{191}$  assignment<sup>20</sup> is  $d_{3/2}$ , the spins and parities of the 129.1- and 170.8-kev excited states are 5/2+ and 9/2+, respectively. The 5/2+ assignment is in accord with a  $d_{5/2}$  shell theory level, but there is no theoretical single-particle assignment for the 9/2 + level.

The Os<sup>191</sup> beta-transition of energy only 142 kev and half-life 15 days predicates an allowed transition. According to the cascade scheme proposed above, approximately equal intensity beta-branches would proceed to the 170.8-kev and 129.1-kev excited states. Both component spectra would necessarily be allowed, the lower energy component having  $\log ft = 5.2$  and the higher energy component having  $\log ft = 5.6$ . The level assignment of the Os<sup>191</sup> ground state would then be unequivocally 7/2+.

### Os191m: DISCUSSION

The multipolarity of the 74.2-key transition has been inferred from lifetime considerations. Probably the most uncertain factor in this analysis concerns the internal conversion coefficients,  $\alpha_{\rm T}$ , which influence the transition lifetimes,  $\tau_{\gamma}$ , through the relationship,  $\tau_{\gamma}$  $= \tau_{\text{expt}}(1+\alpha_{\text{T}})$ . Estimates of the internal conversion coefficients have been obtained in the following way.

As the gamma-transition energy is just in excess of the K-shell binding energy (73.87 kev), estimates of the threshold K-shell conversion coefficients for a number of multipole radiations were obtained from the tables of Spinrad and Keller.<sup>21</sup> Very approximate estimates of the intense L-shell conversions were then obtained using the empirical values of the K/L ratios of Goldhaber and Sunyar.18

For E3 and E4 transitions,  $\log_{10}(\tau_{\gamma}\rho^{2l})$  has the values of 9.4 and 11.6, respectively, and for M3 and M4 transitions,  $\log_{10}(\tau_{\gamma}\rho^{2l-2})$  has the values 8.5 and 10.9, respectively. These values for E3 and M3 are approximately a factor 10<sup>2</sup> higher than the "average" observed values for a 74-kev transition of either of these multipoles.<sup>18</sup> For E4 and M4, the values are a factor of approximately 10<sup>3</sup> lower than the average observed values. This fact alone probably precludes an M4assignment, since it is generally found that M4 transitions lie much closer than a factor of 10<sup>3</sup> to the average value.

As the Os<sup>191</sup> ground state has been given a 7/2+assignment, and as the isomeric transition is very probably an octopole transition, it follows that the spin

74L 20 OSMIUM SOURCE GAMMA IRRADIATION COUNTS PER SECOND × 10<sup>3</sup>/HP G 129k 42M 74M POSSIBLE **BETA- SPECTRUM** 129L 1.000 1,500 500 HP

FIG. 2. Beta- and conversion-electron spectra of Os<sup>191m</sup> and Os<sup>191</sup>.

of the isomeric state is either 1/2 or 13/2. Of the four states having these spins, the even parity, 1/2 spin, state can be ruled out on the grounds of the absence of a direct beta-transition from Os<sup>191m</sup> to the Ir<sup>191</sup> ground state. Both the odd parity, 1/2 spin, and even parity, 13/2 spin, states are in accord with  $p_{1/2}$  and  $i_{13/2}$  levels of nuclear shell theory. An isomeric transition from a  $p_{1/2}$  state would be E3, and from an  $i_{13/2}$  state would be M3. A decision on whether this transition is M3 or E3 can be reached using the empirical L sub-shell conversion ratios determined by Mihelich.<sup>22</sup> For M3,



FIG. 3. Proposed decay scheme of Os<sup>191m</sup> and Os<sup>191</sup>.

<sup>&</sup>lt;sup>20</sup> Brix, Kopfermann, and Siemens, Naturwiss. 37, 397 (1950); H. Kopfermann, Naturwiss. 38, 29 (1951).
 <sup>21</sup> B. I. Spinrad and L. B. Keller, Phys. Rev. 84, 1056 (1951).

<sup>&</sup>lt;sup>22</sup> J. W. Mihelich, Phys. Rev. 87, 646 (1952) and private communication.



FIG. 4. Aluminum absorption curves of the radiations from a neutron-irradiated source of Os. These curves show the strong 1-Mev beta-component of Os<sup>183</sup> which has an end point at approximately 350 mg/cm<sup>2</sup>.

the expected  $L_{III}$  intensity is about twice the  $L_{I}$  intensity, whereas for E3, the  $L_{II}$  intensity appears to exceed the  $L_{III}$  intensity, with the  $L_{I}$  intensity very weak. The experimental  $L_{I}:L_{II}:L_{III}$  ratios therefore decide strongly in favor of M3. This assignment is shown in Fig. 3.

The existence of a 7/2+ state in the region of Os<sup>191</sup>. which has even Z and 115 neutrons, is not predicted by the independent particle nuclear shell theory. Odd neutron levels in this shell are  $h_{9/2}$ ,  $f_{7/2}$ ,  $f_{5/2}$ ,  $p_{3/2}$ ,  $p_{1/2}$ , and  $i_{13/2}$ . As an odd number of neutrons can lead to an even parity state only if they are of even parity, the 7/2+ state is therefore attributed to a coupling of an odd number of  $i_{13/2}$  neutrons or possibly a coupling of an  $i_{13/2}$  neutron with the nuclear core.

There are two other examples<sup>23</sup> of 7/2 + levels in this region. W<sup>183</sup> and W<sup>185</sup>, with N = 109 and 111, respectively, exhibit 7/2+ isomeric states. It was pointed out that these states were anomalous, and it now seems reasonable that they may also arise from a coupling of an odd number of  $i_{13/2}$  neutrons.

Systematics of isomer levels<sup>23,24</sup> indicate that  $p_{1/2}$ levels are energetically close to  $i_{13/2}$  levels in the region of N=117. In fact, extrapolation of level trends indicates that the  $i_{13/2}$  levels might lie lowest in the region of Os<sup>191</sup>.

## **Os193: OBSERVATIONS AND DISCUSSION**

The identification of the 32-hour beta-activity as Os<sup>193</sup> was made on the basis of lifetime and absorption measurements. Osmium sources obtained from pile irradiations exhibited a strong 32-hour beta-activity, and aluminum absorption curves indicated a predominant 1-Mev beta-component which decayed with this halflife. Curves showing the decay of the 1-Mev component are given in Figs. 4 and 5. Osmium sources from betatron irradiations showed an initial decay of 14 hours half-life, with no 32-hour activity observable. Absorption curves also gave no indication of the 1-Mev betacomponent associated with the 32-hour activity. Curves showing the decay of the Os<sup>191</sup> radiations are given in Fig. 6.

In the pile irradiated sources several very weak conversion lines were observed to decay with half-lives intermediate between 14 hours and 15 days. The gamma-transitions identified from K and L conversion lines were of energies 215, 323, and 460 kev. There were also weak intensity lines of 55.2, 59.4, 60.0, and 67.2 kev. The 59.4- and 60.0-kev lines appear to be  $L_1$ and  $L_{11}$  conversion lines of a 72.4-kev gamma-transition. Some or all of these transitions could have half-lives consistent with 32 hours.

Gamma-transitions associated with the 32-hour activity have been reported previously<sup>4,8,10</sup> at energies



FIG. 5. Aluminum absorption curves of radiations from a neutron-irradiated source of Os. These curves were taken at later times and with the source closer to the counter than in the case of Fig. 4. The curves illustrate the disappearance of the Os<sup>193</sup> beta-component, as well as the increasing similarity with the curves of Fig. 6, as the Os source ages.

<sup>&</sup>lt;sup>23</sup> M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179

<sup>(1952).</sup> <sup>24</sup> R. D. Hill, Brookhaven Quarterly Report 11, 18 (1951)

of 1.58, 1.17, 0.22, and 0.065 Mev. If the 59.4- and 60.0kev conversion lines are to be identified with the 65-kev transition observed in delayed-coincidence by Mc-Gowan,<sup>10</sup> then it must be concluded that this transition is only very weak compared with the beta-intensity of Os<sup>193</sup>. McGowan has identified the 65-kev transition as E2. According to Gellman *et al.*<sup>19</sup> the total L conversion of such a transition is approximately nineteen. From the measured intensity of the 59.4- and 60.0-kev lines relative to the intensity of the 32-hour betaspectrum, we conclude that the "65-kev" transition



FIG. 6. Aluminum absorption curves of radiations from a gamma-irradiated source of Os. These curves show an electron component of end point approximately  $20 \text{ mg/cm}^2$  which is characteristic of Os<sup>191</sup>.

Energy (kev)	Interpretation	γ-energy (kev)	Activity
152	$\gamma_1 - L$ in Re	164	5
160	$\gamma_1 - M$ in Re	163	5
163	$\gamma_{0}-K$ in Re	235	2
223	T in Po	225	5
021	$\gamma_2 - L$ in Re	200	í.
231	$\gamma_2 - M$ in Ke	234	r
219	$\gamma_3 - K$ in Pt	297	Ir <sup>192</sup>
284	$\gamma_2 - I$ in Pt	207	Tr192
206	M = Dt	200	T. 192
290	$\gamma_3 - M$ III Ft	299	11
231	$\gamma_4 - K$ in Pt	309	Ir <sup>192</sup>
296	$\gamma_{i} - L$ in Pt	309	Tr <sup>192</sup>
304	$\gamma_{\rm H} = M$ in Pt	307	Tr192
504	74 m m 1	507	11
240	$\gamma_5 - K$ in Pt	318	Ir <sup>192</sup>
304	$\gamma_5 - L$ in Pt	317	Ir <sup>192</sup>
312	$\alpha = M$ in Pt	315	Tr192
512	ys minit	515	11
391	$\gamma_6 - K$ in Pt	469	Ir <sup>192</sup>
456	$\gamma_6 - L$ in Pt	469	Ir <sup>192</sup>
	/·	_07	
580	$\gamma_7 - K$ in Re	652	Os <sup>185</sup>
641	$\gamma - L$ in Re	653	Os185
652	M in Re	656	Oc185
055	y7-m m Ke	050	05.00

TABLE III. Internal conversion lines of long half-lives from neutron-irradiated osmium.

occurs to the extent of less than 2 percent of the betadecays from  $Os^{193}$ .

#### **Os185: OBSERVATIONS**

A two-month old neutron-irradiated Os source was examined in a lens spectrometer and conversion lines corresponding to previously reported<sup>9,25</sup> transitions of 0.65 and 0.88 Mev were observed. Prolonged exposures of the same source in 180° magnetic spectrographs brought out a number of conversion lines of weak intensities and also of long half-lives. An attempt has been made to analyze and identify these lines in Table III.

<sup>25</sup> M. M. Miller and R. G. Wilkinson, Phys. Rev. 82, 981 (1951).