APPENDIX II

In this appendix we show how I_μ can be evaluated in detail to order $|n|^2$ even if $\mu>0$. Consider, for example, I_1.

$$I_1=(2\pi)^{-3}N_-^*N_+\int d\tau \exp\{i(\mathbf{k}_+-\mathbf{k}_-+\boldsymbol{\sigma})\cdot\mathbf{r}+iqr\}$$
$$\times F^*(-n_-,1;\rho_-)F(-n_+,1;\rho_+)r^{-2}.$$

We now use the fact that

$$F(-n,1;\rho)=1-n\,\mathrm{Ei}(\rho)+O(|n|^2),$$

where $\mathrm{Ei}(\rho)$ is the exponential integral function. It is easily shown that correct to order $|n|^2$,

$$I_1=\int d\tau \exp\{i(\mathbf{k}_+-\mathbf{k}_-+\boldsymbol{\sigma})\cdot\mathbf{r}+iqr\}$$
$$\times[F^*(-n_-,1;\rho_-)+F(-n_+,1;\rho_+)$$
$$+|n_+n_-|\,\mathrm{Ei}(\rho_+)\,\mathrm{Ei}^*(\rho_-)-1]r^{-2}.$$

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Decay of UX_1, UX_2, and UZ†

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The decay of Th^{234}(UX_1) and Pa^{234}(UX_2 and UZ) has been investigated by means of a coincidence scintillation spectrometer. Three gamma rays at 29, 63, and 92 kev were found in UX_1. Four gamma rays at 250, 750, 1000, and 1810 kev were found in UX_2, and four gamma rays at 250, 760, 910, and 1680 kev were observed in UZ. The coincidence relations between the gamma rays, and also between the gamma rays and the beta radiation, have been investigated. A decay scheme is proposed and spin and parity assignments have been made for some of the levels of Pa^{234} and U^{234}.

THE beta-emitting substance in natural uranium was isolated by Crookes[1] as early as 1900 and called UX. Later on Fajans and Göhring[2] showed that this substance is a radioactive equilibrium of two isotopes, UX_1 and UX_2, in modern nomenclature Th^{234} and Pa^{234}, respectively. Hahn[3] showed that Pa^{234} decays with two different half-lives and that both activities emit beta and gamma rays. The long-lived activity was given the symbol UZ. Actually, UX_2 and UZ were the first example of an isomeric pair.

The radiation from UX_1, UX_2, and UZ has been studied extensively for a long time. At first absorption technique was used, but more recently the beta radiation has been investigated by means of magnetic spectrometers, and is therefore fairly well known. Almost nothing is known about the gamma radiation, however, and it has been difficult to set up a consistent decay scheme. It seemed therefore worthwhile to make a complete reinvestigation of the radiation from UX_1, UX_2 and UZ, using a coincidence scintillation spectrometer.

The apparatus used in this work has been described elsewhere.[4] It consists of two scintillation spectrometers. Each one can be used as a beta- or gamma-ray spectrometer, depending on the crystal material. The pulses from the first spectrometer are led to a single channel analyzer, which selects pulses corresponding to a certain energy-range. The pulses from the second spectrometer are displayed on an oscilloscope screen and are recorded by photographic methods. The oscilloscope sweep is triggered by coincidences between the selected pulses from the first spectrometer and pulses from the second spectrometer. Hence, a recorded pulse distribution shows the spectrum of the radiation which is in coincidence with the radiation component selected by the analyzer. One can, for example, select beta rays within a certain energy range and study the corresponding coincidence gamma-ray spectrum. It is also possible to select a certain gamma ray and record the corresponding beta component. One can finally study gamma-gamma coincidences by selecting a certain gamma ray and recording the corresponding coincidence gamma spectrum. Hence, this apparatus makes possible a complete investigation of the coincidence relations in a radioactive decay.

The radioactive material used for making sources was isolated from natural uranium by an ether-water extraction method. It was precipitated as the fluoride with lanthanum as the carrier. After a short while it contains UX_1, UX_2, and UZ in equilibrium. The work on UX_1 and UX_2 has been done with sources made up of this material. The activity of UZ is so low that it does not interfere with the measurements on UX_1 and UX_2. The radiation of UX_1 is of low energy, that of

† Contribution from the Institute for Atomic Research and Department of Physics, Iowa State College, Ames, Iowa. Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.
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[1] W. Crookes, Proc. Roy. Soc. (London) **A66**, 409 (1900).
[2] K. Fajans and O. Göhring, Naturwiss. **1**, 399 (1913).
[3] O. Hahn, Naturwiss. **9**, 84 (1921).
[4] S. A. E. Johansson, Ames Laboratory Report ISC-431 (unpublished).

UX_2 of high energy. It is therefore possible to investigate their radiation without being bothered by overlapping spectra.

UZ was isolated from the equilibrium material by a separation of protactinium from thorium. The protactinium fraction contains both UX_2 and UZ, but UX_2 decays very fast (half-life 1.18 min) giving a pure UZ source. The separation was made in the following way using tantalum as the carrier: The radioactive material, containing the equilibrium amount of UZ, was dissolved in dilute sulfuric acid. A solution of potassium tantalate was added and the solution was heated. The tantalum oxide precipitate was filtered, washed, and dried.

UX_1

UX_1 (Th^{234}) has a half-life of 24.1 days. It emits low-energy beta rays and a strong low-energy gamma radiation. The continuous beta rays have been investigated several times by absorption technique and cloud chambers. More recently Bradt and Sherrer,[5] using a 180° magnetic spectrometer, determined the end point as 205 kev. Jnanananda,[6] with a lens spectrometer, found the end point to be at 190 kev. Quite recently Stoker, Heerschap, and Hok[7] reported a careful investigation of the UX_1 beta spectrum, using a double focusing spectrometer. They were able to resolve the continuous beta spectrum into two components with end points at 193 and 103 kev.

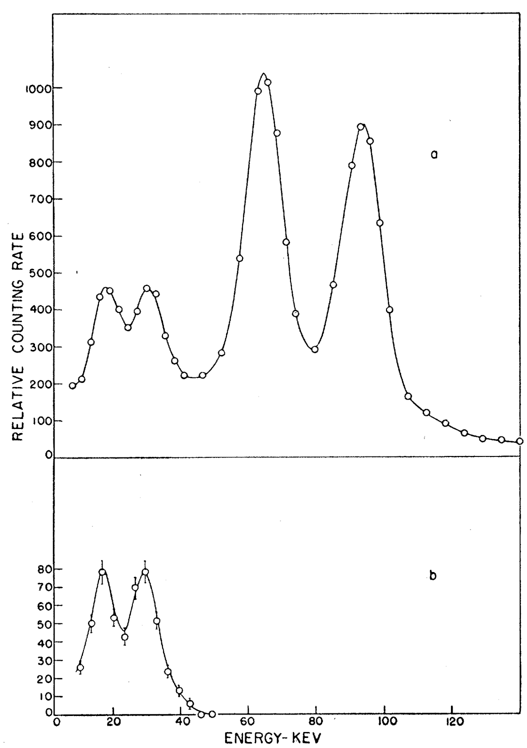

FIG. 1. (a) Gamma-ray spectrum of UX_1. (b) Spectrum of the gamma radiation in coincidence with the 63-kev gamma ray.

Superimposed upon the continuous beta spectrum are several internal conversion lines. Very early, Meitner[8] reported some lines due to the conversion of a 92 kev gamma-ray. Bradt and Sherrer[5] arrived at the same results. Stoker, Heerschap, and Hok[7] in more recent work have found very many internal conversion and Auger lines. Besides strong lines due to a 91.5-kev gamma ray, there are also weaker lines assigned to gamma rays at 63, 47, 43, and 29 kev.

A sodium iodide scintillation spectrometer was used to find the gamma spectrum of UX_1, shown in Fig. 1(a). It consists of two main peaks at 94 and 64 kev, and two weaker ones at 30 and 17 kev. The 94-kev peak is presumably made up of two components. Part of it is due to K x-rays emitted in internal conversion of the UX_2 gamma rays. The mean energy of the uranium K x-rays is 96 kev. The 94-kev peak is so strong, however, that it cannot be solely due to the x-rays. The greater part of it must be due to a gamma ray. In order to determine its energy one must know how much of the peak is due to x-rays and then subtract this part of the peak. The x-ray intensity is not known accurately, however, and therefore it is only possible to say that the energy is a little less than 94 kev, probably 92–93 kev. This value is close to the value 91.5 kev found from the energy of the conversion lines in the beta spectrum.[7]

The 64-kev peak must be due to a gamma ray. The energy is close to the value 63 kev found from the internal conversion lines. The 30-kev peak is probably made up of two components. Part of it is an "escape peak" from the 63-kev gamma ray, having an energy of about 34 kev. The intensity of this escape peak should be about 10 percent of the 64-kev peak. The greater part of the 30-kev peak must therefore be due to a gamma ray with an energy of about 29 kev. This value coincides with one of the gamma-ray energies found in the internal conversion spectrum. The 17-kev peak finally is attributed to L x-rays of protactinium, emitted in the internal conversion of the UX_1 gamma rays. The L_α and L_β x-rays have the energy values 13 and 17 kev, respectively.

In order to establish a decay scheme some coincidence measurements have been performed. The relation between the beta and gamma radiation was investigated in the following way: One of the gamma rays was picked out from the spectrum by means of a sodium iodide scintillation spectrometer and the single-channel analyzer. Those pulses from an anthracene spectrometer, which were in coincidence with the selected gamma ray, were recorded. The results are rather qualitative because of the low counting rate and the poor resolution of the anthracene scintillation

[5] H. Bradt and P. Sherrer, Helv. Phys. Acta **19**, 307 (1946).
[6] S. Jnanananda, Phys. Rev. **69**, 570 (1946).
[7] Stoker, Heerschap, and Hok, Physica **19**, 433 (1953).
[8] L. Meitner, Z. Physik **17**, 54 (1923).

spectrometer at low energies. They definitely show, however, that the gamma rays at 92, 63, and 29 kev are all in coincidence with a beta group having an end point at about 100 kev.

To get a further check on the decay scheme some gamma-gamma coincidence measurements were performed. With the channel set to pick out the 92-kev gamma ray, no coincidences were obtained except for a few accidental ones. With the channel selecting the 63-kev gamma ray, the coincidence spectrum shown in Fig. 1(b) was obtained. It consists of a peak from the 29-kev gamma ray and a peak from the L x-rays emitted in the internal conversion of the gamma ray.

These coincidence measurements show that all three gamma rays are in coincidence with the same beta-ray group. The 63-kev and 29-kev gamma rays form a cascade with the 92-kev ray as a cross-over transition.

UX

The isotope $UX_2(Pa^{234})$ has a half-life of 1.18 min. It emits high-energy beta rays and a very weak gamma radiation. Except for absorption and cloud-chamber measurements the first investigation of the continuous beta spectrum was made by Marshall[9] using a 180° magnetic spectrometer. He found the end point to be at 2320 kev and some evidence for a low-energy beta group with an end point at 1500 kev. Recently the continuous beta spectrum was measured by Stoker *et al.*[7] They report three beta groups with end points at 2305, 1500, and 580 kev and relative intensities of 90 percent, 9 percent, and 1 percent, respectively.

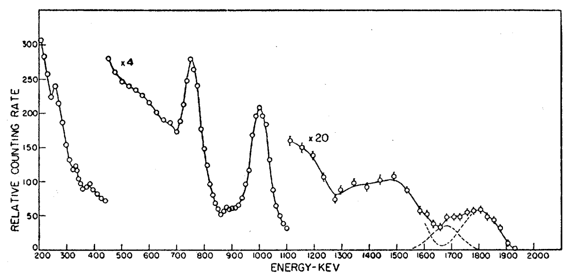

FIG. 2. Gamma-ray spectrum of UX_2 and UZ.

The most striking feature of the UX_2 beta spectrum is the presence of some strong internal conversion lines at about 800 kev. They were first found in 1933 by Meitner[10] and have been remeasured several times since then. The agreement between the various measurements is very poor, however. Most probably there is just one transition with an energy of 810 kev. If there are some more lines, they are very weak. The conversion lines of the 810-kev transition are surprisingly strong.

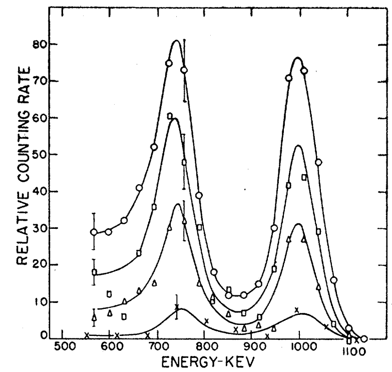

FIG. 3. Spectra of the gamma radiation in coincidence with beta radiation of different energies. The energies are (from the top) 690, 930, 1170, and 1410 kev, respectively.

The intensity of the K line is 0.52 percent of the UX_2 beta rays.[7]

The gamma-ray spectrum of UX_2 from a sodium iodide scintillation spectrometer is shown in Fig. 2. There are well-defined photopeaks at 250, 750, and 1000 kev, indicating gamma rays of these energies. There are also two small bumps at 330 and 380 kev. The corresponding gamma rays must be very weak and it is uncertain whether they belong to UX_2 or to some impurity. They will not be dealt with any further in this work. At 900 kev there is another small peak. It belongs to UZ and will be discussed later in connection with the decay of UZ. There is also some high-energy gamma radiation. The pulse distribution is most easily interpreted as due to two gamma rays of 1680 and 1810 kev. The two components are shown with dotted lines in Fig. 2.

A series of coincidence measurements was performed in order to find out the coincidence relations in the decay of UX_2. The first type is beta-gamma coincidences. A thin source was mounted between two scintillation spectrometers with anthracene and sodium iodide crystals, respectively. Figure 3 shows a series of gamma spectra obtained with the channel set to select beta particles at 1410, 1170, 930, and 690 kev. The channel width was 240 kev. The spectra are all of the same shape. This fact shows that the two gamma rays at 750 and 1000 kev are in coincidence with the same beta-ray group.

Next, some gamma-beta coincidence measurements were performed. With the channel set on the 1000-kev peak, a spectrum was obtained which is shown in Fig. 4 as a Fermi plot. The end point is around 1350 kev. Adding the gamma-ray energy one gets a disintegration energy of 2350 kev, which is approximately the same as the end point of the high-energy beta group.

An attempt was also made to select the gamma rays at 1800 kev and record the corresponding coincidence beta spectrum. The result was rather qualitative due to the low counting rate. It shows, however, that these

[9] J. S. Marshall, Proc. Roy. Soc. (London) **173**, 391 (1939).
[10] L. Meitner, *Handbuch der Physik* (Springer, Berlin, 1933), Vol. 22.

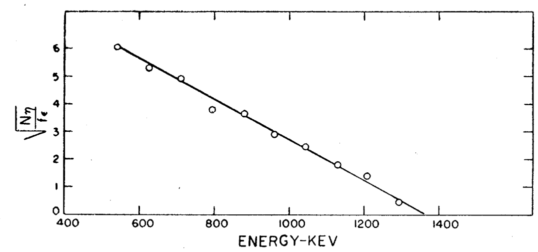

FIG. 4. Fermi plot of the beta radiation in coincidence with the 1000-kev gamma ray.

high-energy gamma rays are in coincidence with beta rays of about 600-kev maximum energy.

Finally, the coincidence spectrometer was used to make some gamma-gamma coincidence measurements. The source was mounted between two sodium iodide spectrometers. A lead shield was placed between the two crystals in order to prevent back-scattered radiation from one crystal from reaching the other crystal. Figure 5 shows the coincidence spectrum obtained with the channel of the analyzer set on the 250-kev peak. Apparently the 750-kev gamma ray is in coincidence with the 250-kev ray. There is also a weak peak at 900 kev, indicating that the gamma ray of this energy, which belongs to UZ, also is in coincidence with the 250-kev gamma ray.

From these coincidence measurements one can conclude that the 1000-kev gamma ray is a cross-over transition to a cascade of the 750- and 250-kev gamma rays. The gamma rays are in coincidence with a beta-ray group having an end point at about 1350 kev.

UZ

An isomeric state of Pa^{234} with a half-life of 6.7 hours is UZ. The decay has been investigated by Feather, Bretscher, and Dunworth[11,12] using absorption and coincidence technique. They found a complex beta spectrum with the end point at 1200 kev. The gamma radiation, with a mean energy of about 700 kev, showed

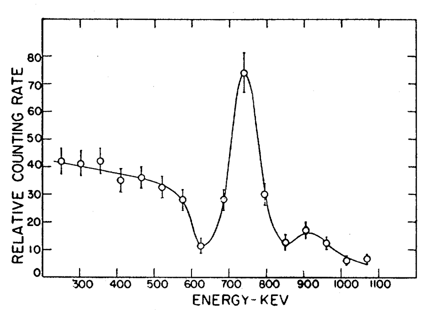

FIG. 5. Spectrum of the gamma radiation in coincidence with the 250-kev gamma ray.

[11] N. Feather and E. Bretscher, Proc. Roy. Soc. (London) **A165**, 530 (1938).

[12] N. Feather and J. V. Dunworth, Proc. Roy. Soc. (London) **A168**, 566 (1939).

a high coincidence rate. Bradt and Sherrer[13] made further experiments using the same technique. In addition they investigated the beta radiation using a 180° magnetic spectrometer. They were able to resolve the spectrum into two groups. For the low-energy group they report an end point at 450±30 kev. The analysis of the spectrum is rather unreliable, however, because of the low counting rate.

Figure 6(a) shows the gamma-ray spectrum of UZ. It has been resolved into two components belonging to gamma rays at 760 and 910 kev. They are shown with dotted lines in the figure. The intensity ratio of the two rays is 0.8. The UZ source was very weak. Therefore, it was difficult to get a reliable pulse distribution at lower energies because of the background of the crystal.

The coincidence relations between the gamma rays were investigated using the coincidence spectrometer. The channel was set on the 910-kev peak and the

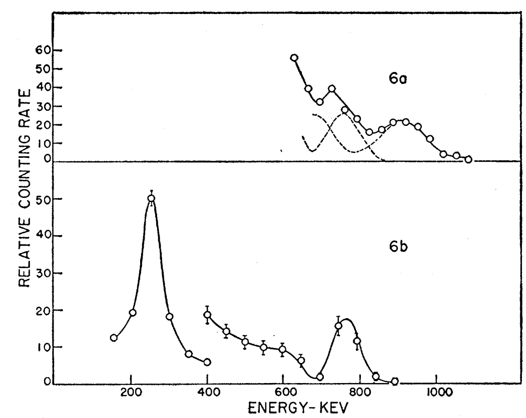

FIG. 6. (a) Gamma-ray spectrum of UZ. (b) Spectrum of the gamma radiation in coincidence with the 910-kev gamma ray.

corresponding coincidence gamma spectrum was recorded. It is shown in Fig. 6(b). The 760-kev gamma ray shows up much better in this coincidence spectrum because there is no interference from the 910-kev ray. The spectrum also contains a 250-kev gamma ray, which was completely hidden in the background of the spectrum shown in Fig. 6(a). The intensity is roughly the same as that of the 760-kev gamma ray.

Hence UZ decays by emission of three gamma rays in cascade. It is interesting to compare the present results with the absorption and coincidence measurements of Feather *et al.*[11,12] They found the mean energy of the gamma rays to be 700 kev. This value is definitely lower than the mean energy of the two gamma rays at 760 and 910 kev. It shows that there must be some low-energy gamma ray present. The coincidence measurements gave a coincidence rate which was higher than could be expected from a decay with only

[13] H. Bradt and P. Sherrer, Helv. Phys. Acta **18**, 405 (1945).

two gamma rays in coincidence. This, too, can be explained by the presence of a third gamma ray in coincidence with the other two.

DECAY SCHEME

The results obtained in the present work make it possible to set up a consistent decay scheme, which is shown in Fig. 7.

The gamma ray at 1810 kev is assumed to belong to UX_2 because the spectrum of UZ does not contain a high-energy gamma ray of this intensity. It is in coincidence with a 600-kev beta group and must therefore go directly to the ground state. The 1680-kev gamma ray is assumed to be a cross-over transition in UZ. Its energy is close to the sum of 760 and 910 kev. Furthermore, the spectrum of UZ contains some high-energy gamma radiation although it is too weak to permit accurate energy measurements. If there is any high-energy gamma radiation present in UZ one would actually expect it to go to the 250-kev level, from spin considerations.

The uncertain point in this decay scheme is the position of the UZ level with respect to the UX_2 level. It depends on the accuracy of the energy values of the UZ beta radiation. Adopting the value[11] 1200 kev for the end point of the high-energy group, one obtains a disintegration energy of 2360 kev for UZ. The low-energy beta group will then have an energy of 440 kev, which is in good agreement with the value reported by Bradt and Sherrer.[13] This places the UZ level above the UX_2 ground state with an energy difference of 40 kev. The uncertainty in this value is considerable, and the UZ level might quite well be the ground state of Pa^{234}. The two levels must be fairly close together, however.

It is known from the decay of Pu^{238} [14] that there is a level in U^{234} at 45 kev. It is therefore possible that the gamma rays at 250, 1000, and 1810 kev do not go to the ground state but to this excited state. In this case there would be a gamma ray at 45 kev almost completely converted. It should show up as a conversion line in the beta spectrum of Stoker *et al.*[7] There is, however, no line corresponding to a gamma ray at about 45 kev with the intensity required in this case. It therefore seems probable that the gamma rays at 250, 1000, and 1810 kev go directly to the ground state.

As mentioned above, the UX_2 beta spectrum is characterized by the presence of some strong internal conversion lines arising from a gamma transition at 810 kev. No trace of a gamma ray of this energy can be found in the gamma spectrum of Fig. 2. Hence the gamma ray must be completely converted. The only way to account for this is to assume a 0–0 transition going from an excited state at 810 kev to the ground state of U^{234}.

[14] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

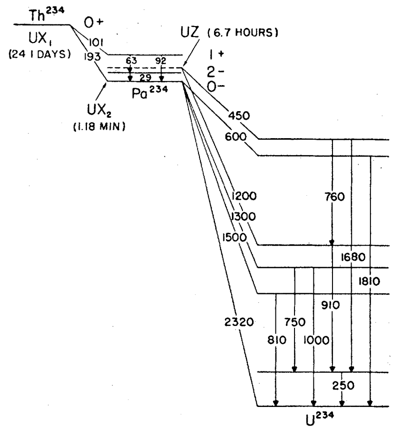

FIG. 7. Proposed decay scheme for Th^{234} and Pa^{234}.

The intensities of the various gamma rays were measured in the following way. By using an anthracene spectrometer and a thin source in a known geometry, the number of beta particles from UX_2 was measured. The number of gamma-quanta from the same source was measured by a sodium iodide spectrometer. The intensities were calculated from these measured values, the shape of the spectrum, the crystal size and shape, and the absorption coefficients of sodium iodide. Secondary absorption processes in the crystal were taken into account. The intensities of the internal conversion lines[7] were added in order to get the total intensities. The intensity values of the gamma-ray peaks and the conversion lines have been used to calculate approximate values on the L-conversion coefficients of the UX_1 gamma rays. Table I gives the energy values, the intensities in percent of the total number of disintegrations, and the L-conversion coefficients for the UX_1 gamma rays.

The branching ratios of the various beta groups have been obtained from the gamma intensities. The corresponding log ft values have been calculated using the graphs and nomograms of Moszkowski.[15] The branching ratios and log ft values are tabulated in Table II.

TABLE I. Gamma-ray intensities and L-conversion coefficients.

Decay	Energy kev	Intensity in percent of total disintegrations	L-conversion coefficient
UX_1	29	6.5	10
UX_1	63	6.5	0.25
UX_1	92	14.8	2.5
UX_2, UZ	250	0.19	
UX_2	750	0.12	
UZ	760	0.06	
UZ	910	0.07	
UX_2	1000	0.37	
UZ	1680	0.02	
UX_2	1810	0.04	

[15] S. A. Moszkowski, Phys. Rev. **82**, 35 (1950).

In assigning spin and parity to the levels of Pa^{234} and U^{234} one encounters several difficulties. The main problem is to make an assignment for the ground state of Pa^{234}. As already discussed by Feather and Richardson[16] in the terms of a Sargent diagram, the difficulty is that the ground state beta transitions of UX_1 and UX_2 have different log ft values, 6.4 and 5.5, respectively. The first one starts from a 0+ state and the second one goes to a 0+ state. Hence they must have the same degree of forbiddeness. However, from the log ft values one would conclude that the first one is first forbidden and the second one allowed. If we assume that both of them are allowed the spin of the Pa^{234} ground state is 1+ according to the Nordheim rule.[17] This assumption creates several difficulties, however. The 1500-kev transition to the 0+ state of U^{234} must then also be allowed but its log ft value is 7.1. The first excited state of an even-even nucleus is known to be a 2+ state almost without exception. A transition to a 2+ state must be allowed if the spin of the Pa^{234} ground state is 1+. There is however no transition with an allowed log ft value except the ground state transition.

Therefore it seems more natural to assume that the ground state of Pa^{234} has the spin 0− and to regard the 2320-kev beta group as an unusual fast first forbidden transition. The 1300- and 600-kev beta transitions have log ft values which characterize them as first forbidden. We therefore assume even parity for these levels.

TABLE II. Branching ratios and log ft values for the beta groups.

Decay	Energy	Branching ratio %	Log ft
UX_1	101	28	5.9
UX_1	193	72	6.4
UX_2	600	0.04	6.8
UX_2	1300	0.49	7.0
UX_2	1500	0.63	7.1
UX_2	2320	98.8	5.5
UZ	450	90	5.6
UZ	1200	10	8.1

The 193-kev beta transition is according to its log ft value first forbidden, $\Delta I=0$, in agreement with the spin assignment 0− for the Pa^{234} ground state. The 103-kev beta transition is probably allowed and the spin of the 92 kev level 1+. The 29-kev level is assumed to have spin 2−. The corresponding beta transition is then first forbidden, $\Delta I=2$, and must be very weak compared to the other two transitions. This assignment makes the 63-kev gamma ray an electric dipole and the 29-kev gamma ray an electric quadrupole transition, giving good agreement between the theoretical[18] and experimental values of the internal conversion coefficients. However, the 92-kev gamma ray is an electric dipole transition according to the spin assignments but an electric quadrupole transition according to its conversion coefficient. There seems to be no way of removing this discrepancy without creating new ones.

Very little can be said about the spin of the UZ level of Pa^{234} without knowing its exact position. A value of 3 or 4 seems rather likely.

The spin values proposed here are only tentative. The main difficulty is, as pointed out above, to make an assignment for the ground state of Pa^{234}. No spin value seems to give complete agreement with the experimental results. This state might be an example of the mixed configurations recently discussed by de-Shalit and Goldhaber.[19]

ACKNOWLEDGMENTS

The author is greatly indebted to Dr. D. J. Zaffarano for his interest and assistance in this work. The technical assistance of Mr. G. Eakins and Mr. J. Powers is gratefully acknowledged.

[16] N. Feather and H. O. W. Richardson, Proc. Phys. Soc. (London) **61**, 452 (1948).
[17] L. W. Nordheim, Revs. Modern Phys. **23**, 3321 (1951).
[18] Gellman, Griffith, and Stanley, Phys. Rev. **50**, 866 (1950).
[19] A. de-Shalit and M. Goldhaber, Phys. Rev. **92**, 1211 (1953).