and we get for the energy loss due to bremsstrahlung

$$\frac{4Z^2e^4}{137\mu}\left[\frac{1}{3}\log^2 2\gamma+\frac{29}{12}\log^2 2\gamma-\left(\frac{217}{36}+\frac{\pi^2}{8}\right)\log 2\gamma-\left(\frac{\pi^2}{3}-\frac{7}{4}\right)\log\frac{Q}{\mu}+\frac{899}{108}+\frac{\pi^2}{12}-\frac{39}{16}\sum_{n=1}^{\infty}\frac{1}{n^3}-\frac{1}{4}\sum_{n=1}^{\infty}\frac{1}{2^n n^3}\right]. \quad (13)$$

Since energy lost in elastic collisions is given by the Bethe-Bloch formula,[7]

$$-\frac{dw}{dx}=n\frac{4\pi Z^2e^4}{\mu\beta^2}\log\frac{2\mu\beta^2\gamma^2}{I}, \quad (14)$$

we get for the fractional radiative correction for an infinitely heavy particle

$$\Delta=\frac{1}{137\pi\log(2\mu\beta^2\gamma^2/I)}[0.333\log^2 2\gamma+2.42\log^2 2\gamma-7.26\log 2\gamma-1.54\log(Q/\mu)+6.18]. \quad (15)$$

[7] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944), second edition, p. 218.

Unfortunately, the division Q between small and large momenta does not quite cancel out of the final formula, but Δ depends only logarithmically on Q so that it seems to be reasonable to expect that a reliable result would be obtained by choosing Q somewhere between the extremes $\mu<Q<2E_0$. However, since the Bethe-Heitler formula does not take into account the recoil of the scattering center, formula (15) may be in error when $m\gamma$ is larger than or of the order of M.

Taking collisions of pi-mesons ($M=270m$) with argon atoms as an example, we obtain the values in Table I for the percent radiative correction. The correction is positive, as one would expect, since mesons are losing some additional energy to radiation. Also, the correction is small for mesons of available energy.

Radiation observed in the laboratory should not show a marked anisotropy and the maximum energy of the emitted photons should be of the order of $\frac{1}{2}$ Mev, except for a few hard photons close to the direction of the incident heavy particle.

The author is indebted to Professor L. I. Schiff for suggesting this problem and for his continued interest and advice throughout the solution.

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Radiations of Pu^{243}

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The radiations of Pu^{243} were studied with beta- and gamma-scintillation spectrometers alone and in coincidence. An incomplete disintegration scheme is deduced which leads to a total beta-disintegration energy of 560 kev. A half-life of 4.98±0.02 hours was observed.

INTRODUCTION

THE radiations of Pu^{243} were first studied[1] by absorption measurements which indicated a maximum beta-energy of 0.5 Mev. O'Kelley and Orth[2] reported a preliminary value of 0.39 Mev for the maximum beta-energy and gammas of 0.095 and 0.12 Mev. The purpose of this research was to determine the total decay energy of Pu^{243}. Therefore, in addition to examining the beta- and gamma-spectra, beta-gamma and gamma-gamma coincidence measurements were undertaken.

SAMPLE PREPARATION

Samples of a nitrate solution of plutonium enriched in Pu^{242} were evaporated to dryness in a quartz tube and irradiated in the thimble of the Argonne Heavy Water Reactor for approximately 15 hours.

Immediately after irradiation the plutonium was purified from all extraneous activity with a resin column, a series of precipitations, and solvent extractions. Several irradiations were made to complete the experiments reported.

APPARATUS

Scintillation spectrometers were employed for the measurement of the beta- and gamma-ray spectra. Thallium-activated sodium iodide crystals $1\frac{1}{4}$ inches in diameter and $\frac{1}{2}$ inch thick were used for gamma-detection, and an anthracene crystal of the same diameter and $\frac{1}{4}$ inch thick was used for the beta-counter. The sodium iodide crystals were sealed in cylindrical 17 ST aluminum cups turned to a thickness of 0.013 inch on the end facing the sample and closed on the other end with a Pyrex window. A similar assembly was used for

[1] Sullivan, Pyle, Studier, Fields, and Manning, Phys. Rev. 83, 1267 (1951).
[2] G. D. O'Kelley and D. A. Orth, quoted by Thompson, Street, Ghiorso, and Reynolds, Phys. Rev. 84, 165 (1951).

the anthracene crystal except that the aluminum window was 0.001 inch thick. Short Lucite light pipes coupled the crystals to RCA type 5819 photomultipliers, which were attached to cathode follower preamplifiers. After further amplification the pulses from each detector were fed into a single channel differential pulse-height analyzer (designed by Robert K. Swank of this laboratory) and also into a coincidence circuit having a resolving time, $2T$, of 0.14 microsecond. The outputs of the two analyzers and the coincidence circuit were mixed in a threefold coincidence circuit with a resolving time of about 4 microseconds.

Photopeak widths of 11 percent (full width at half-height) for the 661-kev gamma of Ba^{137} and conversion electron peak widths of 15 percent for the 622-kev Ba^{137} conversion electrons were observed.

GAMMA-SPECTRUM

In order to obtain the gamma-spectrum of Pu^{243} free from interference from the radiations emitted by the alpha-emitting plutonium isotopes in the sample, the spectrum of gammas in coincidence with all betas with energies greater than 100 kev was measured. A beryllium absorber weighing 235 mg/cm^2 was placed over the gamma-counter. This, added to the 0.013-inch aluminum window covering the crystal, was sufficient to absorb betas with energies less than 0.8 Mev. The 0.001-inch window on the beta-counter was sufficient to absorb the alpha-particles. Samples weighing less than 0.1 mg, spread over an area of 2 cm^2 on a thin mica sheet, were placed $\frac{1}{8}$ inch from the face of the sodium iodide crystal and $\frac{5}{16}$ inch from the anthracene crystal. Gamma-energy calibration was made with the Ag^{109} gamma, for which the value of 89 kev given by Bradt *et al.*[3] was used rather than 87 kev given by Siegbahn *et al.*[4] since critical absorption measurements by one of us (D.W.E.) have shown that its energy lies between the K absorption edges of lead and bismuth or between 88.2 and 90.6 kev. The gamma-spectrum obtained is shown in Fig. 1, curve A. Prominent peaks at 19 kev and 85 kev are observed, in addition to a low peak at 55 kev which is attributed to the escape of iodine K x-rays following photoelectric absorption of the 85-kev gamma. The peak at 19 kev is probably due to L x-rays of americium. When the coincidence circuit was not used, the spectrum shown in Fig. 1, curve B, was obtained. The only observable difference is the large excess of L x-rays arising from the alpha-emitting plutonium isotopes.

If the entire 19-kev peak in the coincidence spectrum is assumed to be due to L x-rays from the conversion of the 85-kev gamma, the L conversion coefficient may be calculated from the ratio of the areas under the two peaks of curve A, Fig. 1. The observed intensity of the x-rays must be corrected for absorption and for the fluorescence yield. Correction for absorption is complicated by the fact that the L x-rays are distributed over such a wide range of energies.

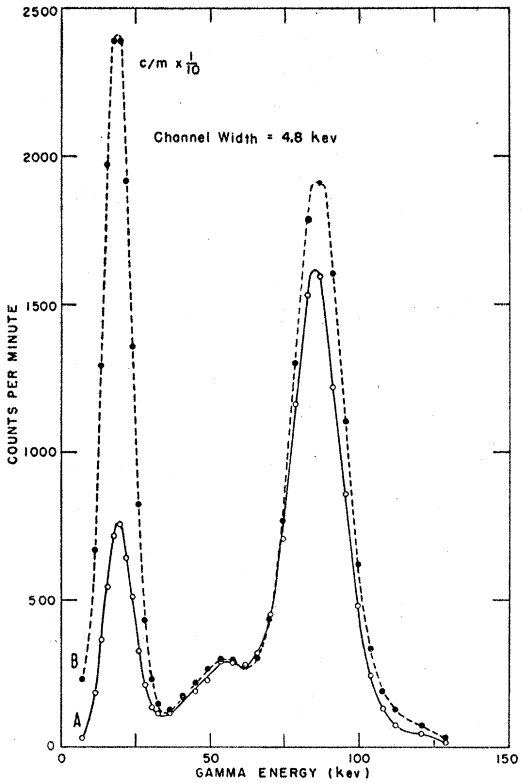


FIG. 1. Gamma-spectrum of Pu^{243}. A. Beta (>100-kev)—gamma coincidences; B. single gamma counts.

In order to correct for absorption the relative intensities of the different L x-ray lines were calculated by assuming that equal amounts of conversion occurred in the three L subshells. The fluorescence yields for the three L subshells were obtained by extrapolating the fluorescence yield and Coster Kronig coefficient data given by Kinsey[5] to $Z=95$. The relative intensities of the x-rays arising from vacancies in each of the L subshells were assumed to be the same as were observed in uranium by Compton and Allison.[6] In calculating the transmission of the x-rays through the absorber it was necessary to calculate the transmission as a function of angle through the absorber and then to integrate over the entire solid angle subtended by the detector (0.40 of 4π). An over-all efficiency of 0.062 was obtained for the number of L x-rays detected per L vacancy.

The efficiency is not very sensitive to assumptions as to the relative amounts of conversion in the three L subshells. The efficiency for the 85-kev gamma was calculated to be 0.38.

After application of these corrections, the ratio of L conversions to unconverted gammas was found to be

[3] Bradt, Gugelot, Huber, Medicus, Preiswerk, Scherrer, and Steffen, Helv. Phys. Acta **20**, 153 (1947).

[4] Siegbahn, Kondaiah, and Johansson, Nature **164**, 405 (1949).

[5] B. B. Kinsey, Can. J. Research **A26**, 404 (1948).

[6] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), pp. 643–645.

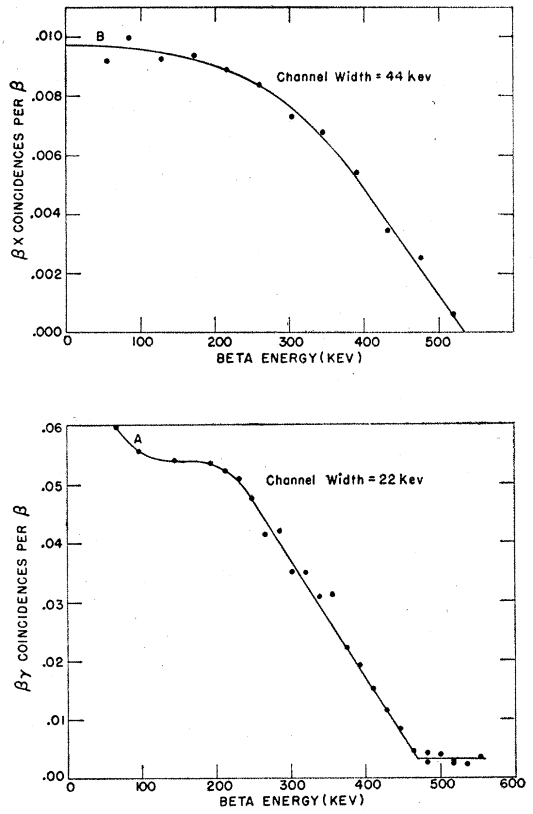


FIG. 2. Coincidences per beta as a function of beta-energy. A. Beta—85-kev gamma coincidences; B. beta—L x-ray coincidences.

1.3. This value is probably reliable to within 30 percent and represents an upper limit to the L conversion coefficient of the 85-kev gamma since some of the x-rays may arise from other transitions.

BETA- AND BETA-GAMMA COINCIDENCE SPECTRA

The total beta-spectrum and the beta-spectrum in coincidence with the 85-kev gamma were measured with the anthracene scintillation spectrometer. The spectrometer was calibrated with the 622-kev conversion electron line of Ba^{137} and was used with a channel width of 22 kev. Alpha-particles from the other plutonium isotopes were absorbed in the 0.001-inch aluminum window to prevent interference with the beta-measurements. The pulse-height selector on the gamma pulse-height analyzer was set to accept only those pulses between 65 kev and 130 kev in height in order to count nearly all of the 85-kev gamma-pulses and to eliminate any x-ray pulses. In Fig. 2, curve A, the number of beta-gamma coincidences per beta is plotted *vs* the beta-energy. The value of $\beta\gamma/\beta$ remains relatively constant up to 200 kev and then drops to a much lower, constant value at 470 kev. This behavior indicates that the 85-kev gamma is coincident with a group of beta-particles having a maximum energy of about 470 kev. The small number of coincidences observed above 470 kev may be due to a slight impurity.

Figure 3 shows the Kurie plots of the total beta-spectrum and of the spectrum in coincidence with the 85-kev gamma. The total beta-spectrum exhibits a continuous curvature which does not permit an accurate estimation of the upper energy limit. The coincidence spectrum does show a fairly straight portion which intercepts the energy axis at 455 kev. To this energy must be added the energy loss in the aluminum absorber,[7] 13 kev, to get the maximum beta-energy of 468 kev. In order to obtain the maximum beta-energy of the total beta-spectrum, the lower energy component was subtracted from the total spectrum in arbitrary amounts until the residual spectrum gave a straight line Kurie plot. In Fig. 3, the observed coincidence spectrum was multiplied by 8 and then subtracted from the total spectrum to give a spectrum having an observed end-point energy of 553 kev which, when corrected for energy loss in the aluminum absorber, gives a maximum beta-energy of 566 kev.[8] If the spectra are assumed to have allowed shapes, this breakdown of the spectrum assigns 60 percent of the betas to the 566-kev branch and 40 percent to the 468-kev branch.

In order to establish whether or not the 566-kev beta-branch is followed by a highly converted transition, beta—L x-ray coincidences were studied as a function of beta-energy. The pulse-height selector associated with the gamma-detector was set to accept only pulses corresponding to gamma-energies between 11 and 27 kev and, thus, counted almost all of the L x-rays and prac-

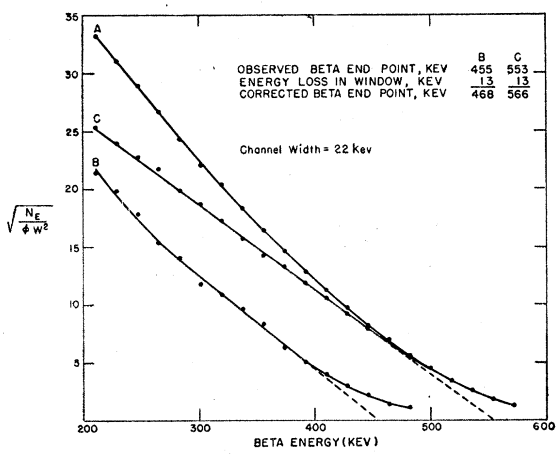


FIG. 3. Kurie plots of Pu^{243} beta-spectra. A. Total beta-spectrum; B. betas in coincidence with 85-kev gamma; C. subtraction of coincidence spectrum from total beta-spectrum.

[7] H. Bethe, *Handbuch der Physik* (Springer-Verlag, Berlin, 1933), Vol. XXIV–1, pp. 519–523.

[8] Multiplication of the coincidence spectrum by eight before subtraction from the total spectrum is equivalent to assuming a detection efficiency of 0.125 for the 87-kev gamma transition, whereas the calculated efficiency, taking into account the conversion coefficient and the fraction of the total 87-kev gamma-spectrum accepted by the analyzer, equals 0.16. The end point of the high energy component is not significantly altered if the subtraction is based upon the calculated detection efficiency.

tically none of the 85-kev gammas. The plot of beta—L x-ray coincidences per beta as a function of beta-energy is shown in Fig. 2, curve B. The steady decrease as the beta-energy is increased indicates clearly that the 566-kev beta is not followed by a gamma-transition highly converted in the L shell. If the 566-kev beta were followed by a gamma-transition completely converted in the L shell, the ratio of $\beta X/\beta$ would be expected to approach a limiting value of 0.06 at high beta-energies. The value of 0.01 observed at low beta-energies is consistent with the assumption that most of the L x-rays are due to conversion of the 85-kev gamma.

Since the 566-kev beta is not in coincidence with the 85-kev gamma and since no other gamma of appreciable intensity is present, the 566-kev beta must represent the total decay energy unless it is followed by a gamma of lower energy than the L binding energies of americium. It is plausible to assume that the 85-kev gamma-transition also proceeds to the ground state of Am^{243} since the sum of the energies of the gamma-transition and the 468-kev beta in coincidence with it equals 553 kev, which is in fair agreement with 566 kev.

GAMMA—85-KEV GAMMA COINCIDENCE SPECTRUM

The spectrum of gammas in coincidence with the 85-kev gamma was measured with two sodium iodide scintillation spectrometers in coincidence. Counter B was $\frac{9}{32}$ inch from the sample and was set to accept pulses from 70 to 89 kev in height. This setting was held fixed and was chosen in order to discriminate against higher energy gammas and at the same time to maintain a high counting efficiency for the 85-kev gamma. An aluminum absorber weighing 713 mg/cm^2 was placed between the sample and counter B. Counter A was $\frac{5}{32}$ inch from the sample and was covered with 71.9 mg/cm^2 of beryllium in addition to the 0.013-inch 17 ST aluminum window in order to absorb the betas. The analyzer for counter A was used with a 5.4-kev channel whose position was varied to obtain the gamma-spectrum coincident with the gammas detected in counter B.

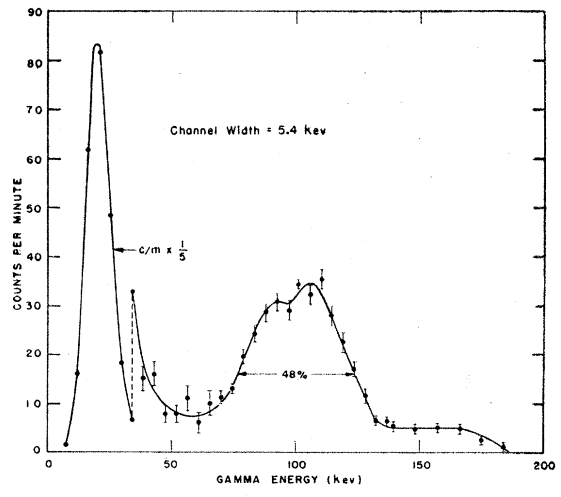


FIG. 4. Pu^{243} 85-kev gamma-gamma coincidence spectrum.

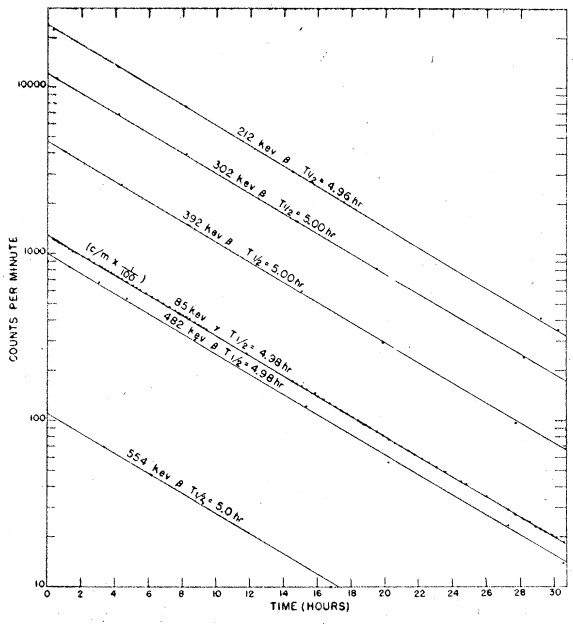


FIG. 5. Decay of the 85-kev gamma and of selected points on the beta-spectrum of Pu^{243}.

Figure 4 shows the spectrum obtained. In addition to a prominent line at the position expected for L x-rays, a low intensity broad unresolved peak is obtained at about 100 kev and a low broad continuum extending to 185 kev. The peak at 100 kev must be due to two or more gammas since its width is 48 percent as compared with 25 percent obtained for the 85-kev peak in Fig. 1. The peak shape is consistent with that expected for two gammas of nearly equal intensities with energies of about 92 kev and 107 kev. The high energy shoulder could be caused by a single gamma with an energy of about 160 kev.

If every 85-kev gamma were in coincidence with a completely L converted gamma, the integral number of 85-kev gamma—L x-ray coincidences per 85-kev gamma detected in counter B should equal the detection efficiency of counter A for L vacancies of 0.06 (see section on gamma-spectrum); the observed ratio is 950/48 000 $=0.020$, thus indicating that the 85-kev gamma is in coincidence with L vacancies one-third of the time. Similarly, if every 85-kev gamma were in coincidence with an unconverted 100-kev gamma, the integral number of gamma-gamma coincidences per gamma detected in counter B should be approximately equal to the geometrical efficiency of counter A or 0.38; the observed ratio of 280/48 000$=0.0058$ indicates that very few unconverted gammas with energies of about 100 kev are in coincidence with the 85-kev gamma. It follows from this that the gamma-transitions responsible for the 85-kev gamma—L x-ray coincidences must have high conversion coefficients. Since 566-kev

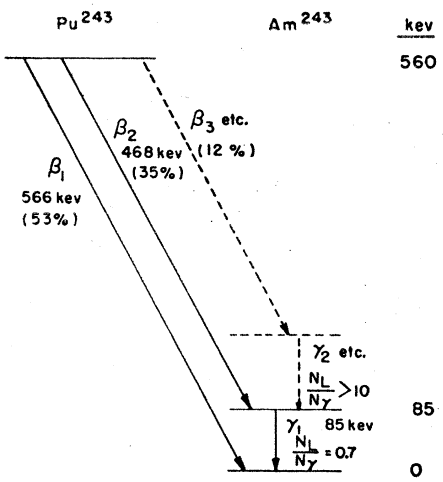


FIG. 6. Proposed decay scheme of Pu^{243}.

beta—L x-ray or gamma-coincidences were not observed, and since an L vacancy accompanies the 85-kev gamma only one-third of the time, these highly converted gamma-transitions must precede the 85-kev gamma. This implies that lower energy beta-groups are present with a total abundance at least one-third as great as that of the 468-kev beta or about 12 percent.

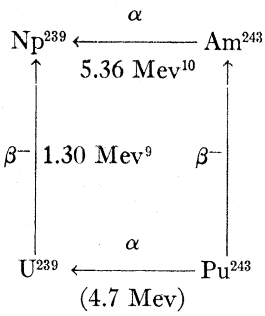
The L conversion coefficient of 1.3 obtained by assuming that all of the L x-rays arise from the conversion of the 85-kev gamma must be corrected for the L x-rays observed to be in coincidence with the 85-kev gamma. Upon making this correction, the conversion coefficient is lowered to 0.72. This value is still an upper limit since L x-rays may be present which are not in coincidence with the 85-kev transition.

HALF-LIFE OF Pu^{243}

The decay curves of the 85-kev gamma and of selected points on both branches of the beta-spectrum of Pu^{243} are shown in Fig. 5. The 85-kev gamma-decay curve was obtained by selecting a channel width great enough to accept almost the entire photopeak so that slight fluctuations in pulse height would not affect the counting rate appreciably. Decay curves of the beta-gamma coincidences and of gamma-gamma coincidences also showed a 5-hour half-life. The best value for the half-life is considered to be 4.98 ± 0.02 hours.

DISCUSSION OF RESULTS

A disintegration scheme which agrees with the experimental observations is shown in Fig. 6. The dotted lines represent transitions which are inferred from the observation of 85-kev—L x-ray coincidences. The transitions marked γ_2, etc., may be any combination of the gammas observed in the gamma-gamma coincidence measurements or some other highly converted gamma which was not observed. It is believed that the total disintegration energy of 560 kev cannot be in error by more than 30 kev. A total disintegration energy of approximately 0.6 Mev is expected on the basis of the following energy cycle, where the alpha-energy of Pu^{243} is estimated as *ca* 4.7 Mev from alpha-decay systematics.



The conversion coefficient of the 85-kev gamma may be compared with the theoretical values calculated by Gellman *et al.*[11] An interpolation of their data for uranium gives values of the total L conversion coefficient of 0.17 for electric dipole, 10 for magnetic dipole, and 30 for electric quadrupole. The observed upper limit of 0.7 indicates that the transition is probably electric dipole. Measurement of the relative intensities of the L conversion electron lines should permit an unambiguous assignment since the relative values of the L_1, L_2, and L_3 conversion coefficients calculated by Gellman *et al.*[11] vary markedly with the kind of transition.

A calculation of the logft values for the two main beta-transitions gives 6.0_8 for the ground-state transition and 5.9_4 for the transitions to the excited state, thus indicating that both transitions are first forbidden with change of parity and $\Delta J=0$ or ±1. This is in agreement with the strong spin-orbit coupling single particle model[12] which predicts for the ground-state transition, $g_{7/2}\rightarrow f_{7/2}$. Possible spin assignments for the first excited state are $f_{5/2}$, $f_{7/2}$, $h_{9/2}$, which would lead to magnetic dipole radiation to the ground state. The most probable assignment to the first excited state on the basis of the single particle model would be $f_{5/2}$. No spin and parity assignments are possible which will allow electric dipole radiations and at the same time assign equal forbiddenness to the two beta-transitions. Further experiments, particularly with magnetic focusing beta-ray spectrometers, will be needed to elucidate the details of the decay scheme.

The authors wish to thank M. S. Freedman for helpful discussions and Gray L. Pyle for assistance in some of the counting.

[9] Huizenga, Magnusson, Freedman, and Wagner, Phys. Rev. **84**, 1264 (1951).
[10] G. T. Seaborg (private communication).
[11] Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).
[12] Maria Goeppert Mayer, Phys. Rev. **75**, 1969 (1948); **78**, 16 (1950).