PHYSICAL REVIEW VOLUME 88, NUMBER 5 DECEMBER 1, 1952

The Beta-Spectra of Pu^{239}, Pu^{240}, and Pu^{241}

MELVIN S. FREEDMAN, F. WAGNER, JR., AND D. W. ENGELKEMEIR
Argonne National Laboratory, Chicago, Illinois

(Received April 24, 1952)

The beta-spectrum of Pu^{241} measured in a double lens spectrometer on samples containing Pu^{239}, Pu^{240}, and Pu^{241}, exhibits an allowed shape from $E_0=20.5$ kev to 14 kev, where instrumental effects distort it. Conversion lines found beyond the end point combined with gamma-scintillation counter measurements lead to the assignment of gammas of 39, 53, 100, 124, and 384 kev associated with the alpha-decay of Pu^{239}, and a gamma of 49.6 kev accompanying the alpha-decay of Pu^{240}; and the existence of 26.4- and 59-kev gammas in Am^{241} alpha-decay is confirmed. A conversion line corresponding to a highly converted 41-kev gamma in Am^{241} was found. Gammas of 100 and 145 kev probably associated with the alpha-branch in Pu^{241} were also found.

The influence of the orbital electronic rearrangement energy on the beta-decay is discussed.

EXPERIMENT

THE beta-spectrum of 10-yr Pu^{241} was examined on samples of radiochemically pure plutonium containing the isotopes Pu^{239}, Pu^{240}, and Pu^{241}, isotopically enriched in Pu^{241} by pile neutron irradiation. Isotopic compositions were determined mass spectrographically.

The plutonium was deposited on a *ca* 10-μg/cm^2 film of LC 600 by the evaporation of a solution of plutonium nitrate. Prepared samples had an average thickness of *ca* 10-μg/cm^2, but inspection with a microscope showed some local clumping.

The beta-spectrum was taken on a double lens spectrometer having a transmission of *ca* 2 percent and a resolution of *ca* 2 percent. An atmospheric pressure methane-flow proportional counter was used as detector. The counter window was built up of layers of thin Duco and Formvar films to a thickness of *ca* 60-μg/cm^2. A 50 percent transmission grid was used to support the window.

Window transmission characteristics of the counter were determined with the use of a sample of Pm^{147}. This sample was vacuum volatilized onto a 10-μg/cm^2 LC 600 film as a chelation compound of prometheum with trifluoroacetyl-acetone $[Pm(F_3C_5H_4O_2)_3]$, to produce a very uniform sample of an average thickness *ca* 10-μg/cm^2. Since it has been shown by Langer, Motz, and Price[1] that the Kurie plot of thin samples of Pm^{147} is straight to less than 8 kev, the Kurie plot was extrapolated for Pm^{147} back from 20 kev where it began to fall below the straight line, and the ratio of the observed to extrapolated data was used below 20 kev as a window transmission correction factor.

RESULTS

By using the correction factors so obtained, the Kurie plot (Fig. 1) (uncorrected for orbital electron screening) of the Pu^{241} sample exhibited an allowed shape ($\log ft=5.7$) from $E_0=20.5(+1.2, -0.3)$ kev to 14 kev, where source thickness effects and large window corrections tend to make the data unreliable. The *ft* value and shape are consistent with the spin-orbit coupling shell model predictions for the state assignments, $g_{9/2}$ and $f_{7/2}$ for Pu^{241} and Am^{241}, respectively, giving $\Delta J=-1$, yes, corresponding to a first forbidden beta-transition. The log*ft* value falls somewhat below the boundary value of 6.0[2] found for first forbidden beta-decays in lighter isotopes. Since $Z_c \approx 1.6 W_0^{\frac{3}{2}} \approx 1.6 \ll Z$,[3] the shape of a first forbidden transition with $\Delta J=1$ should almost exactly conform to the allowed shape.[3] Considering the experimental limitations on the quality of the data and the omission of shielding corrections to the Fermi function, any conclusions on the significance of the allowed shape must be drawn with reservations. This applies particularly in view of the possible effects of orbital rearrangement energy on the spectrum shape (see discussion below).

CONVERSION LINE AND GAMMA-SPECTRA

Seven very low intensity conversion lines were observed beyond the end point of the beta-spectrum. The maximum energy surveyed was 60 kev. In order to determine the assignment of these lines among the three plutonium isotopes or the Am^{241} [4] present, the

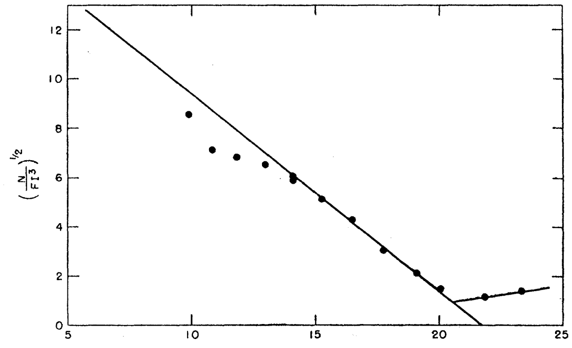

FIG. 1. Kurie plot for Pu^{241}.

---

[1] Langer, Motz, and Price, Phys. Rev. **77**, 798 (1950).

[2] Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. **23**, 315 (1951).

[3] E. J. Konopinski, Revs. Modern Phys. **15**, 209 (1943).

[4] We are indebted to G. T. Seaborg and Frank Asaro for pointing out this latter possibility to us.

TABLE I. Gamma-ray energies and relative intensities for Pu^{239} and Pu^{240} from conversion electrons, scintillation counter spectrometer, and proportional counter spectrometer.

Isotope to which gamma is assigned	Conversion electron energy (kev)	Conversion shell	Binding energy of (kev)	Gamma-energy conv. elec. (kev)	Gamma-energy NaI(Tl) (kev)	Gamma-energy prop. spect., (kev)	Gamma-energy best value (kev)	Relative intensity of gammas
Pu^{239}					17.5-$L_{\alpha, \beta, \gamma}$			100
					21.6-L_{γ}	20.9-L_{γ}		
						39	39	0.4
	31.4	*LI*	21.9	53.3	54.0	53.2	53.1	1.4
	35.3	*LIII*	17.2	52.5				
	47.1	*MI*	5.56	52.7				
	50.2	*MIV*	3.72	53.9				
					100		100	1.1
					124		124	0.5
					384		384	0.3
Pu^{240}						13.7-L_{α}		
					17.1-$L_{\alpha, \beta, \gamma}$	17.2-L_{β}		100
						21.2-L_{α}		
	32.9	*LIII*	17.2	50.1	47	48	49.6	3
	44.4	*MII*	5.20	49.6				
	27.6	*LI*	21.8	49.4				
Pu^{241}					100			100
					145			20
Am^{241}					18-$L_{\alpha, \beta, \gamma}$			25
					28	26.4	26.4	25
	23.8	*LIII*	17.6	41.4			41.4[a]	
	37.9	*LII*	21.7	59.6	61.1	60.2	59.0	100
	41.0	*LIII*	17.6	58.6				
	52.9	*MI*	5.75	58.7				

[a] C. A. Prohaska, University of California Radiation Laboratory Report 1395 (unpublished) (August, 1951) refers to observation of highly converted 41-kev gamma-ray in Am^{241} decay. Wagner, Freedman, Engelkemeir, and Huizenga (to be published) observed conversion lines of a 42-kev gamma in the beta-decay of U^{237}.

spectrum of a sample of almost isotopically pure Pu^{239} was examined, and four conversion lines were found, none identical with and all of still lower intensity than the first seven. A third sample containing a different relative isotopic abundance of Pu^{240}: Pu^{241} was run, and the intensities of the first seven conversion lines were observed to vary approximately with the amount of Pu^{240} present. However, it was noted that the Am^{241} which had grown in from the beta-decay of Pu^{241} was, in these samples, almost in constant ratio to the amount of Pu^{240}, and that some of the lines corresponded to previously observed lines[5] following alpha-decay of Am^{241}. From this and on the basis of the gamma-ray measurements (see below), three of the lines are assigned to Pu^{240} alpha-decay and four to Am^{241} alpha-decay. The data are tabulated in Table I, together with the gamma-ray assignments of these lines. Binding energies characteristic of uranium are used in calculating the gamma-energies for Pu^{239} and Pu^{240}, and of neptunium for Am^{241}, on the assumption that the gamma-emission follows alpha-decay.

Gamma-rays were also observed directly with both a sodium iodide scintillation spectrometer and an argon-CO_2 filled proportional counter spectrometer, using a pure Pu^{239} sample and three samples containing different ratios of Pu^{240} and Pu^{241}, from which the Am^{241} had been freshly separated. We are indebted to Professor G. T. Seaborg for pointing out to us the necessity for separating Am^{241} from samples containing Pu^{241} immediately before attempting to observe the plutonium gamma-spectra, since the intense 26.5- and 60-kev gammas of Am^{241} soon mask the weak gammas of the plutonium isotopes. In the first experiments in which americium was not separated, the most prominent gammas observed in a sample containing Pu^{240} and Pu^{241} were the 26.5- and 60-kev gammas of Am^{241}. In a later group of experiments, americium was separated and the measurements completed within 6 hours, during which time no perceptible growth of Am^{241} occurred.

L x-rays of uranium and gammas with energies of 39, 53, 100, 124, and 384 kev were observed in the pure Pu^{239} sample. In the three samples containing Pu^{240} and Pu^{241}, additional gammas with energies of 47, 100, and 145 kev were observed. The Pu^{238} and Pu^{239} contributions to the total gamma-spectrum were small in these samples and were subtracted off. Comparison of the intensity of the *L* x-ray and the 47-, 100-, and 145-kev gammas with the amount of each plutonium isotope present in the three samples indicated that the 47-kev gamma and most of the *L* x-rays are due to Pu^{240}. The 100- and 145-kev gammas are definitely not assignable to Pu^{240} but may be assigned fairly definitely to Pu^{241} on the basis of line intensities relative to isotopic abundances. The 100-kev line has a full width at half-height of 25 percent, whereas a width of 19 percent is expected (the 89-kev Ag^{109} gamma-line has a width of 20 percent). This line broadening is quite reproducible and must be due to the presence of two or more unre-

---

[5] G. D. O'Kelley and C. A. Prohaska (private communication from F. Asaro); Charles I. Browne, Jr., University of California Radiation Laboratory Report 1764 (unpublished) (June, 1952); Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952).

solved lines. The observed line width is very nearly equal to the width expected for uranium K x-rays which could arise from the internal conversion of the 145-kev gamma. Thus, the line may be entirely K x-rays.

Since approximately ten 100-kev gammas and two 145-kev gammas are emitted per 10^6 Pu^{241} betas, it is evident that the gammas cannot follow the Pu^{241} beta-disintegration. It is possible that these gammas are emitted in the alpha-branching of Pu^{241}; the gamma to total alpha-ratios are 0.35 for the 100-kev and 0.07 for the 145-kev gammas.

The gammas found and their assignments are given in Table I.

DISCUSSION

It has been pointed out by Schwartz[6] and by Edwards[7] that in beta-decay in a heavy element, where the beta-energy is low, the difference between the total orbital electron binding energies of parent and daughter atoms will be comparable to the maximum beta-energy. This energy difference, which is negative for a beta-minus-decay, must appear associated with the beta-transition, either partly or wholly as kinetic energy of the beta or neutrino or as quantum radiation (possibly converted).

It is predicted theoretically,[6,8] that this energy will, except for an experimentally indeterminable amount of the order of one hundred electron volts,[6] be emitted in the decay as kinetic energy of the beta-particle and neutrino. The manner by which this extra energy affects the energy distribution is taken into account by the use of the Fermi function $F(Z,W)$, *with shielding corrections*, using for the total energy of the transition the sum of the specifically nuclear contribution plus the electron binding energy difference, namely, the observed maximum energy.

A distribution of low energy excited states of the daughter atom will, in general, be formed, of average excitation energy about 100 volts for high Z. To each such state a beta-spectrum will be associated. Thus, the observed spectrum will be a composite of many distributions, differing only in their maximum energies by a small amount, and thus with present techniques, indistinguishable experimentally from a single normal spectrum. This is shown in curve *c*, Fig. 2, which exhibits, very closely, the characteristic allowed shape for an allowed beta-transition.

An alternative theoretical approach[9] assigns all the orbital binding energy difference to the beta-spectrum, on the basis that the coupling of the neutrino to the electrostatic field is negligible. In consequence, the beta-spectrum distribution (*a*, Fig. 2), characteristic of the decay of a bare nucleus, is shifted into curve *b* by this difference, resulting in a low energy cutoff. Further possibilities involve high energy excitation of the daughter atom, with subsequent radiation or extensive ionization.

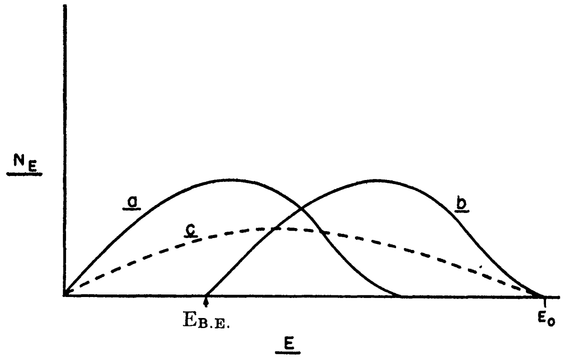

FIG. 2. Orbital rearrangement effect on beta-spectrum. (a) Beta-spectrum of bare nucleus. (b) Beta-spectrum of whole atom if only electron partakes of orbital electron binding energy ($E_{B.E.}$) difference. (c) Beta-spectrum of whole atom if orbital electron binding energy difference is shared between electron and neutrino.

The total orbital electron binding energy for high Z atoms is given by Foldy[10] as

$$-32.64Z^{7/5} \text{ ev}.$$

For the beta-transition $Pu^{241} \rightarrow Am^{241}$, the difference in binding energies is 19.0 kev. Other evaluations of this quantity give values between 14 and 22 kev. It can be seen from the Kurie plot that no low energy cutoff of the spectrum lies in this range, so the theory of Hebb[9] is not substantiated. Our past experience with other low energy spectra in lighter isotopes has shown that the cutoff observed below 10 kev is ascribable to sample and detector window absorption.

The approximate allowed shape of the upper half of the spectrum is in agreement with the predictions of references 6 and 8, and indicates that the neutrino does share in the orbital energy difference. However, detailed verification of the theoretical predictions as to the spectral shape cannot be made, as screening corrections were not included. Such corrections are not available to the desired precision for such low energies and high Z, except by uncertain extrapolation from the calculations of Reitz.[11] Although the screening correction accounts for the influence of the orbital electron binding energy difference on the shape of the spectrum,[8] since the *observed* maximum energy already includes the contribution of the orbital binding energy difference to the decay energy, application of the screening correction should not grossly affect the shape, i.e., it cannot alter the maximum energy or the low energy intercept. It would be desirable to have Reitz's calculations extended to cover this problem.

With Foldy's value, the specific nuclear contribution

---

[6] H. M. Schwartz, Phys. Rev. **85**, 733 (1952); Phys. Rev. **86**, 195 (1952).

[7] R. R. Edwards, Sixth Southwest Regional Meeting, American Chemical Society (1950).

[8] R. Serber and H. S. Snyder, Phys. Rev. **87**, 152 (1952); M. Hamermesh, private communication.

[9] M. H. Hebb, Physica **5**, 701 (1938).

[10] L. L. Foldy, Phys. Rev. **83**, 397 (1951).

[11] J. R. Reitz, Phys. Rev. **77**, 10 (1950).

to the decay energy is only *ca* 1.5 kev (20.5−19.0), but the spectrum shape and *ft* value are appropriate to a first forbidden, unfavored transition with $E_0=20.5$ kev. It is interesting to note that the experimental errors and the theoretical uncertainty in the value of the total orbital electronic binding energy may be such as to allow for a zero or even a negative value for the specifically nuclear contribution to the decay energy. Thus a stripped Pu^{241} nucleus is probably only slightly (1.5 kev) unstable with respect to beta-minus-decay, or a stripped Am^{241} nucleus may even be energetically unstable with respect to a stripped Pu^{241} nucleus, although neither positron emission nor electron capture processes could occur.

We wish to thank Mr. Maurice Rusnak for aid in the calculations.

PHYSICAL REVIEW VOLUME 88, NUMBER 5 DECEMBER 1, 1952

The Nuclear Moments of Ta^{181}

B. M. Brown* and D. H. Tomboulian
Department of Physics, Cornell University, Ithaca, New York

(Received August 18, 1952)

An analysis of the hfs pattern of several Ta II lines has made it possible to determine the separations of the three levels of the hfs multiplet associated with the $5d^36s\ {}^5F_1$ state of Ta II. For this multiplet, the constants A and B appearing in the energy expression $W=W_J+\frac{1}{2}AK+BK(K+1)$ have the numerical values $A=(-0.079\pm0.001)$ cm^{-1} and $B=(-0.77\pm0.4)\times10^{-3}$ cm^{-1}. Calculations are carried out in order to evaluate the magnetic and quadrupole moments of the Ta^{181} nucleus. On the basis of the above measurements the magnetic moment as calculated by the Goudsmit-Fermi-Segrè formula has the value of 1.9 nuclear magnetons. Taking into account effects due to the spatial extension of the nucleus, this result is raised to 2.1 nuclear magnetons when the correction factor of 12 percent as listed by Klinkenberg is applied. The result for the quadrupole moment as calculated by the Casimir formula is $+5.9\times10^{-24}$ cm^2. According to Sternhermer this moment should be increased by a factor of 10 percent in order to include the effect of an induced quadrupole moment in the closed shell electrons. With this correction the quadrupole moment has the value of $+6.5\times10^{-24}$ cm^2.

INTRODUCTION

It was deemed desirable to undertake an investigation of the hyperfine structure in the spectrum of Ta II taking advantage of recent identification of levels[1] arising from the $5d^36s$ configuration. These levels should exhibit a large hyperfine splitting due to the presence of a single *s* electron. For a term with a small *J*-value optimum conditions are provided for the measurement of deviations due to the presence of an electric nuclear quadrupole moment both from the point of view of pattern size and the number of hfs components. These considerations led to the study of the structure of the ${}^5F_1$ level—the lowest state arising from the $5d^36s$ configuration. Previous researches[2–4] indicate that the Ta nucleus has a spin of 7/2 and a magnetic moment of 2.1 nuclear magnetons. In addition, Schmidt[5] has assigned a value of $\sim6\times10^{-24}$ cm^2 to the nuclear quadrupole moment on the basis of hfs measurements in connection with the lowest states (${}^4F_{3/2}$ and ${}^4F_{5/2}$) of the Ta I spectrum. Values of the quadrupole moment obtained by him from the study of two other levels were found to disagree with the above value. The purpose of the present measurements was to attempt an independent determination of the nuclear magnetic and quadrupole moments.

Table I. Wavelengths and transitions of Ta II lines whose hyperfine patterns were investigated. In each case the final state arises from the $5d^36s$ configuration. The term values of the ${}^3P_0$ and ${}^5F_1$ states are equal to 4124.77 and 0.00, respectively.

Wavelength in air in angstroms	Transition
3379.49	$(33706.50)_1\rightarrow{}^3P_0$
3042.06	$(36987.73)_1\rightarrow{}^3P_0$
2965.92	$(33706.50)_1\rightarrow{}^5F_1$
2965.13	$(33715.15)_2\rightarrow{}^5F_1$
2763.37	$(36177.12)_2\rightarrow{}^5F_1$
2702.80	$(36987.73)_1\rightarrow{}^5F_1$
2595.59	$(38515.55)_2\rightarrow{}^5F_1$

EXPERIMENTAL

The Ta II spectrum was excited in a hollow cathode discharge tube containing argon. The hfs patterns were obtained by the use of a Fabry-Perot interferometer crossed with a 21-ft concave grating in a modified Wadsworth mounting. The interferometer plates were coated with aluminum and three separators having thicknesses of 3, 3.7, and 4 mm were utilized. Measurements were carried out on the seven lines whose wave-

---

* Now at the General Electric Research Laboratory, Schenectady, New York.

[1] Kiess, Harrison, and Hitchcock, J. Research Natl. Bur. Standards 44, 245 (1950).

[2] J. H. Gisolf and P. O. Zeeman, Nature 132, 566 (1933).

[3] N. S. Grace and E. McMillan, Phys. Rev. 44, 949 (1933).

[4] J. H. Gisolf, dissertation, Amsterdam (1935) (unpublished).

[5] T. Schmidt, Z. Physik 121, 63 (1943).