The Beta-Spectra of Cs^{137}, Y^{91}, Pm^{147}, Ru^{106}, Sm^{151}, P^{32}, and Tm^{170}

HAROLD M. AGNEW*
Institute of Nuclear Studies, University of Chicago, Chicago, Illinois
(Received October 11, 1949)

The beta-spectra of Cs^{137}, Y^{91}, Pm^{147}, P^{32}, Ru^{106}, Tm^{170}, and Sm^{151} have been investigated using a large double-magnetic lens spectrometer. The investigation of these materials was carried out with sources whose thicknesses were less than 0.1 mg/cm². The sources were mounted on thin foils in such a manner that they did not accumulate charge. Cs^{137} and Y^{91} were found to have distinctly non-linear Fermi plots. Ru^{106}, Pm^{147}, and Sm^{151} have linear Fermi plots, and the Fermi plot of Tm^{170} is slightly curved. P^{32} had an unusual Fermi plot at the low energy end. Auger electrons have been found associated with the Ba^{137} decay product of Cs^{137}, and the internal conversion electrons have been found accompanying the continuous beta-radiation from Tm^{170}. The energy of the gamma-ray giving rise to the Yb^{170} conversion electrons is 85.4±0.6 kev. The end points of the continuous beta-spectra of these materials have been determined as follows: Low energy group of Cs^{137}, 0.523±0.004 Mev; Y^{91}, 1.56±0.010 Mev; Tm^{170}, 0.990±0.007 Mev; Sm^{151}, 0.0755±0.0010 Mev; P^{32}, 1.718±0.01 Mev; Pm^{147}, 0.229±0.001 Mev; Ru^{106}, 0.0392±0.0003 Mev.

INTRODUCTION

THE work reported here was begun in the hope of finding forbidden spectra which could be analyzed by using the existing theory of forbidden spectra.[1,2] At the time that this work was begun, there had been no reported instances of a beta-emitter exhibiting a Fermi plot which is concave toward the energy axis at the high energy portion of its spectrum. Actually, most of the experimental data indicated a straight line Fermi plot for the high energy portion of the spectrum, and then some deviation at the low energy end. Generally, the deviation at the low energy end was attributed to thickness of source and backing, lack of detection efficiency, and scattering in the spectrometer. In view of the advantages of the spectrometer[3] used in the work reported here—small scattering and high transmission—it was decided to investigate spectra using thin sources and a known detection efficiency.

Experimentally, allowed and forbidden transitions of nuclei undergoing beta-decay are distinguishable through the comparison of their half-lives after taking into account differences brought about by their nuclear charge and the maximum energy of the beta-particles emitted. This classification is given in terms of the so-called ft value. (The notation used here is that adopted by Konopinski.[2]) It should be understood that the ft value is only a rough guide to be used in distinguishing between various types of transitions. However, it is generally agreed that if the ft value lies between 10^3 and 10^4, the transition involved is an allowed one. Another indication of an allowed transition is obtained from the so-called Fermi or Kurie plot. This is a plot of $\{N/[p^2f(Z, W)]\}^{\frac{1}{2}}$ plotted against the total energy of the particles emitted. N is the number of beta-rays per unit momentum interval Δp. The energy-dependent function $f(Z, W)$ shows the effect of the Coulomb field on the emission of the beta-ray. A straight-line Fermi plot is an indication of an allowed transition. The Fermi plot of a forbidden transition should deviate from a straight line if one uses the $f(Z, W)$ corresponding to an allowed spectrum. Konopinski[2] gives in his review article a list of correction factors C_{ij}. According to the present theory, these should be used to correct $f(Z, W)$ in order to get a straight-line Fermi plot in the case of a forbidden transition.

During the progress of this work, several papers[4-6] have appeared reporting the observation of forbidden-type spectra. The work reported here is in agreement with the results reported on Cs^{137} and Y^{91}. In the case of

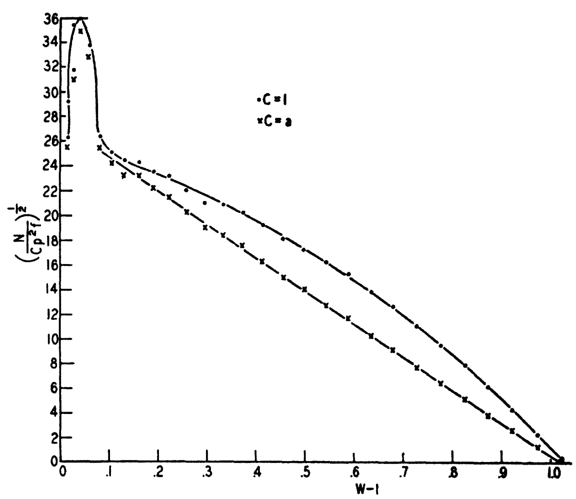

FIG. 1. Fermi plots of the low energy group of beta-rays of Cs^{137} with and without the correction factor a. The hump at the low energy end is due to Auger electrons.

* Now at Los Alamos Scientific Laboratory, Los Alamos, New Mexico.
[1] E. Fermi, Zeits. f. Physik **88**, 161 (1934).
[2] E. Konopinski, Rev. Mod. Phys. **15**, 209 (1943).
[3] H. M. Agnew and H. L. Anderson, Rev. Sci. Inst. **20**, 869 (1949).
[4] C. L. Peacock and A. C. G. Mitchell, Phys. Rev. **75**, 1272 (1949).
[5] L. M. Langer and H. Clay Price, Jr., Phys. Rev. **75**, 1109 (1949).
[6] E. N. Jensen and L. Jackson Laslett, Phys. Rev. **75**, 1949 (1949).

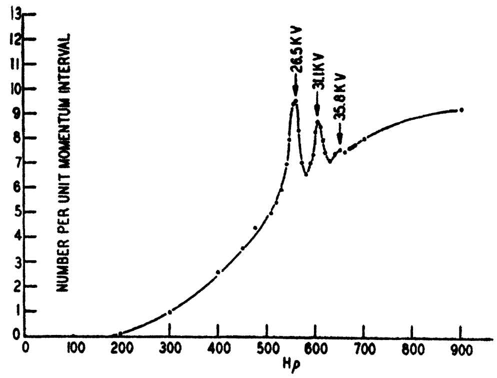

FIG. 2. Auger electrons identified as resulting from Ba^{137}.

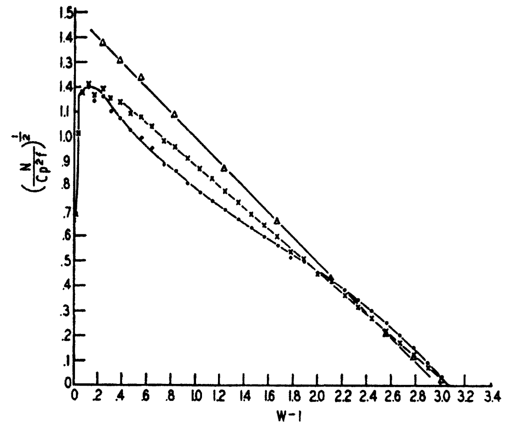

FIG. 3. Fermi plots of the beta-rays of Y^{91} corrected as follows: •$C=1$; ×$C=a$; Δ$C=c$.

Cs^{137}, very distinct Auger lines were detected. From their spacing, it can be shown that they result from the K internal conversion line of the Ba^{137} decay product. Ru^{106}, Pm^{147}, and Sm^{151} were found to have straight-line Fermi plots. Internal conversion electrons were detected accompanying the continuous beta-radiation of Tm^{170}. They result from an excited state of Yb^{170}, the emitted gamma-ray having an energy of 85.4±0.6 kev. Seaborg's[7] table reports, instead, a gamma-ray of energy 0.84 Mev associated with the Tm^{170} activity. If such a gamma-ray exists, its intensity is weaker by a factor of one thousand than the one with energy 85.4 kev. It is suggested that there is an error in the literature and in reality reference is made to a gamma-ray having an energy of 0.084 Mev. The Fermi plot of Tm^{170} was slightly concave toward the axis and an attempt has been made to apply one of the forbidden-type correction factors to it. P^{32} gave a Fermi plot of unexpected shape. The end point of P^{32} has long been used as an energy standard by many investigators to calibrate their spectrometers. It was believed that P^{32} had been thoroughly investigated.[8, 9] It was found, however, that the data at energies below 150 kev were not complete. The work here shows a sharp increase in slope of the Fermi plot at about 150 kev and a slight curvature from there to the end point. The source was followed over five half-lives with no change in the position of the break.

It is believed that all materials used were essentially free from radioactive impurities. However, it should be realized that the presence of an impurity in one of the materials could cause a change in the shape of its spectrum. Because of the long lives of the materials reported here, it was not feasible to take separate data from the same materials over a period of several half-lives.

[7] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).
[8] E. M. Lyman, Phys. Rev. **51**, 1 (1937).
[9] K. Siegbahn, Phys. Rev. **70**, 127 (1946).

APPARATUS

Spectrometer

The spectrometer[3] used in this work consisted of two large identical coils spaced 95.5 cm apart. The intervening space was evacuated and contained the baffle system. The source was placed in the center of one coil and the detector was placed in the center of the other. In addition to the ordinary type of annular baffles, a helical baffle was used to select the desired trajectories. In the work reported here, the spectrometer was used with a resolution corresponding to a half-width at half-maximum of 1.1 percent and a transmission of 1.4 percent using sources 0.12 cm² in area.

All measurements are based on a value of 629 kev for the K internal conversion line of Ba^{137}. This line has been measured by several workers.[4, 10, 11] The value of 629 kev agrees with their reported results to within one-half percent and, considering the resolution employed in these measurements, it was decided to use it as a standard because of its convenience. On the basis of this value, the energies of the Auger electrons from Ba^{137} agreed very well with their calculated values. The end point of P^{32} agreed with the accepted value of 1.71 Mev.

Detector

A Geiger counter of special design was used to detect the beta-rays. The counter had a thin Nylon window which would transmit electrons having an energy greater than 8.8 kev. The thin Nylon window was supported on a grid and the counter was one electrode of an accelerating system which made it possible to detect a known fraction of the low energy electrons. Electrons of the proper momentum passed through a defining annulus and by means of a potential difference maintained between the counter and the annulus could

[10] Townsend, Owen, Cleland, and Hughes, Phys. Rev. **74**, 99 (1948).
[11] M. Friedman (private communication).

be accelerated before entering the counter. By taking spectra with and without the applied accelerating voltage, it was found that the detecting system was 100 percent efficient for 30-kev electrons with no accelerating voltage.

Using the accelerating section, it was 100 percent efficient for electrons with an energy equal to 30 kev minus the applied accelerating voltage. An $8\frac{1}{2}$ percent correction had to be applied for electrons having an energy of 16 kev when an accelerating voltage of 7 kev was used. The extent of these corrections is illustrated in Figs. 4 and 5. Because of the finite thickness of the sources, it is probable that the spectrum is distorted below 20 kev due to scattering in the source itself. The counting rate, when the counter was operated on its plateau, was constant for counter filling pressures ranging from 3 to 9 cm. This was checked using cobalt gamma-rays and beta-particles from uranium.

Sources

The sources were mounted on either zapon or LC600 films* weighing less than 0.01 mg/cm². The actual weight of the source was estimated by using a measured fraction of a solution in which the total non-volatile mass was known. The sources were made to spread out uniformly over an area of about 0.12 cm² by first placing a dilute drop of insulin on the foils. Experience proved that a very satisfactory method of preventing a source of high specific activity from charging up on the type of mounting employed was to cover it with a thin conducting foil after the source had been vacuum dried. Since scattering for low energy electrons is proportional to Z^2, it is important not only to use a thin foil but to use a thin foil of low atomic number. For this reason, an organic material was used for the foil to give it strength, and the foil was made conducting by coating it with Be metal using standard evaporating techniques. Care was taken not to inhale any of the Be fumes or dust. Foils whose total weight including Be was less than 0.006 mg/cm² were prepared in this manner. Such a foil 2 in. in diameter has a resistance from edge to edge of between 50 and 800 megohms as measured with an ohmmeter. These foils were placed Be side down on top of the dried radioactive source and were grounded to the spectrometer. No effect on the spectrum other than the removal of accumulated charge could be detected when such a foil was placed over the source. At least two different sources were run on each material and one percent statistics were obtained at all points of the spectrum except where the counting rate was less than four times the counter's natural background. In these instances, the data checked to within two percent.

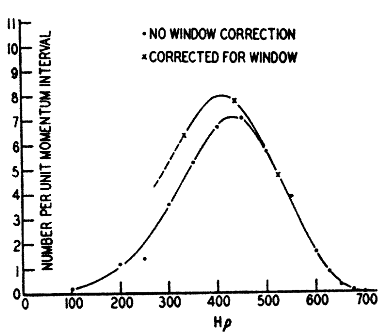

FIG. 4. Momentum distribution of the beta-rays of Ru^{106}.

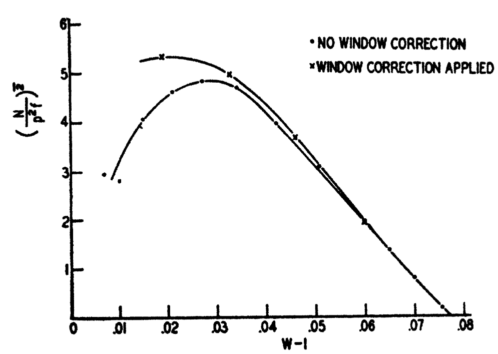

FIG. 5. Fermi plot of the beta-rays of Ru^{106}.

RESULTS

Cs^{137} (Half-Life† 37 Years: $ft = 4.41 \times 10^9$)

Three separate sources of Cs^{137} were prepared from material obtained from Oak Ridge. In each case, the source thickness was less than 0.05 mg/cm². The results are in agreement with those reported by Peacock and Mitchell.[4] The high energy group was not investigated. Figure 1 is a Fermi plot showing how the uncor-

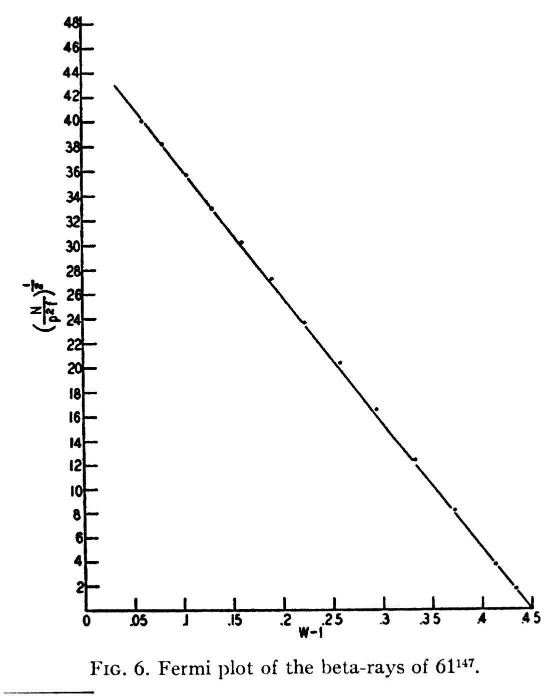

FIG. 6. Fermi plot of the beta-rays of 61^{147}.

* A plastic resin manufactured by the Lithgow Corporation, Chicago, Illinois.

† Half-lives are those given by Seaborg (see reference 7).

rected plot curves toward the energy axis and how using the correction factor $[(W_0-W)^2+W^2-1]$ transforms the curve into a straight line. The hump at the low energy end is due to the presence of Auger electrons. From the end point of 0.523 ± 0.004 Mev, one calculates ft to be equal to 4.41×10^9. Consequently, one would ordinarily classify this material as being second forbidden. Konopinski gives in his reference article various correction factors C_{ij} which one must apply to the function $f(Z, W)$, corresponding to an allowed spectrum, in order to get a straight line when $[N/C_{ij}p^2f(Z, W)]^{\frac{1}{2}}$ is plotted against the energy for an ith-forbidden spectrum. In the case of Cs^{137}, the correction factor a corrected the data extremely well. The only correction factors whose energy dependence is upon the factor a alone are C_{1A} and C_{1T} which are first-forbidden correction factors occurring with G-T selection rules corresponding to $\Delta J=\pm2$(yes). As pointed out by Konopinski, except for the case $\Delta J=\pm2$(yes), if $Z\gg Z_c$ with $Z_c\sim1.6\ W_0^{\frac{1}{3}}$ (W_0 is the total maximum energy of the beta-particle emitted) then all the C_{1j}'s are essentially independent of energy. For Cs^{137}, $Z=55$, whereas $Z_c=4.57$, hence, $Z\gg Z_c$. The correction factors C_{2j} all decrease as $(W_0-W)^2$ for $Z\gg Z_c$ except for $\Delta J=\pm3$(no). For this exception, the correction is determined by the factor

$$c=[3(W^2-1)+3(W_0-W)^4+10(W^2-1)(W_0-W)^2]/90.$$

When the factor c was applied as a correction factor, the Fermi plot was convex with respect to the energy axis. The use of $(W_0-W)^2$ as a correction factor did not give a straight-line Fermi plot. In view of this, one can exclude the possibility $\Delta J=\pm3$(no) and any of the other C_{2j}'s. Consequently, it may be assumed that the low energy group of Cs^{137} is first-forbidden and the large ft value is due to other reasons not included in the theory.

Figure 2 illustrates the Auger electrons occurring at 26.5 ± 0.2 kev, 31.1 ± 0.2 kev, and 35.8 ± 0.2 kev. Using an average of the values of the critical absorption edges of the L and M shells of Ba given by Compton and Allison,[12] one may calculate that Auger electrons associated with Ba should have energies corresponding to 26.1 kev, 31 kev, and 35.8 kev. The experimental results agree very well with these values. With the resolution used, it is very difficult to determine from the separation of the K and L internal conversion lines whether they occur in the Ba or Cs, since it is a problem of measuring 3 kev in 629 kev. However, in the case of the Auger electrons, it is easy to identify them as belonging to the Ba since one needs only to measure 2 kev in 25 kev.

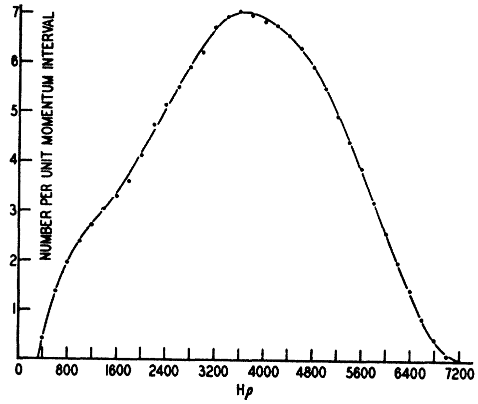

FIG. 8. Momentum distribution of the beta-rays of P^{32}.

It should be pointed out that this material, because of its long half-life, high specific activity, and the presence of the Auger and internal conversion electrons accompanying the continuous beta-spectrum, should be an extremely useful substance for workers in this field. A complete spectrum is shown in Fig. 5 in the report on the spectrometer.

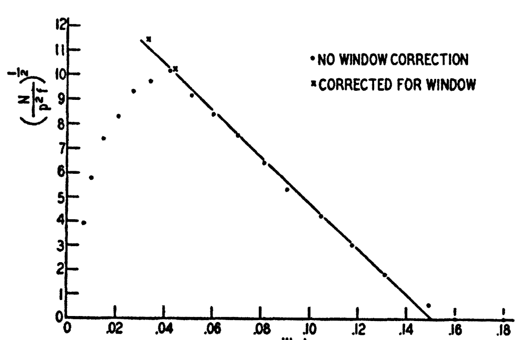

FIG. 7. Fermi plot of the beta-rays of Sm^{151}.

[12] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), p. 792.

Y^{91} (Half-Life 61 Days: $ft=5.7\times10^8$)

Sources less than 0.05 mg/cm² thick of Y^{91} were prepared from material obtained from Oak Ridge. Figure 3 is a Fermi plot of the spectra of Y^{91} showing the effect of using the correction factors a and c on the allowed $f(Z, W)$. From the end point of 1.56 ± 0.01 Mev, one may calculate Y^{91} to have an ft value equal to 5.7×10^8 and consequently it may be classified as being second forbidden. As in the case of Cs^{137}, the correction factor a, when applied to $f(Z, W)$, gives a straight-line Fermi plot.

The nuclear shell model examined by Feenberg and Hammack[13] predicts that Y^{91} be first forbidden with maximum spin change of ±2. This is consistent with the interpretation of the use of the correction factor a. However, by using the correction factor c, one obtains a Fermi plot which deviates only slightly from a straight line at the high energy end. This would indicate that Y^{91} belongs to the second-forbidden group with $\Delta J=\pm3$(no). This is in contradiction with the conclusions one would arrive at from the use of the factor a. In view of this contradiction, it seems unlikely that one

[13] E. Feenberg and K. C. Hammack, Phys. Rev. **75**, 1887 (1949).

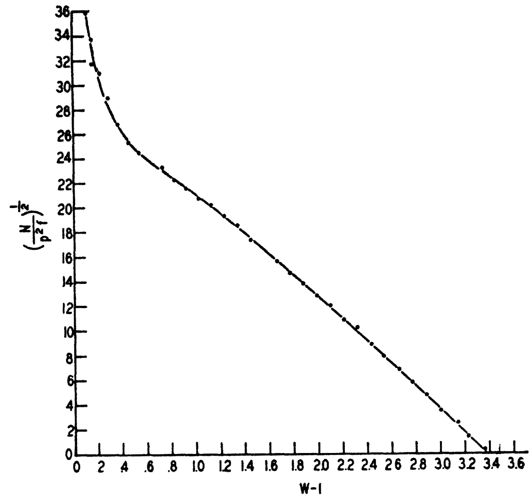

FIG. 9. Fermi plot of the beta-rays of P^{32}.

can determine the type of transition Y^{91} undergoes from an analysis of this type. Langer and Price[5] report using the correction factor a, but do not state whether they have tried any other factors on their data.

Ru^{106} (Half-Life 1 Year: $ft=2.14\times10^4$)

Ru^{106} was prepared by distillation of fission product solution. The author wishes to thank Dr. Ellis Steinberg of the Argonne National Laboratory for his assistance in preparing the material. It was hoped that the source would be free from solid matter except for the Ru^{106}. However, on evaporation, some deposit was visible. The sources used were less than 0.1 mg/cm^2, but their weights could only be estimated.

Figure 4 gives the momentum distribution of the electrons and shows the effect of the accelerator. Figure 5 is a Fermi plot of the data. From this, one obtains an end point of 0.0392 ± 0.0003 Mev, corresponding to an ft value of 2.14×10^4 which places it in the allowed group. This is an unusually low ft value for a substance with such a high atomic number. Because of the low upper energy limit of this spectrum, only a short stretch of the plot is experimentally significant. With this reservation, the data are compatible with a straight-line Fermi plot. No gamma-ray was observed.

Pm^{147} (Half-Life 3.7 Years: $ft=4.01\times10^7$)

Sources of Pm weighing less than 0.05 mg/cm^2 were prepared from material obtained from Oak Ridge. Figure 6 is a Fermi plot of the data. From the end point of 0.229 ± 0.001 Mev, an ft value of 4.01×10^7 was computed. This would place Pm^{147} in the first-forbidden group. However, Fig. 6 indicates that it has an allowed shape. As pointed out by Konopinski, if $Z\gg Z_c$, with $Z_c\sim1.6W_0^{\frac{1}{2}}$, then the first-forbidden spectra are nearly identical with allowed spectra for $\Delta J=0\pm1$. In this case, $Z_c=2.18$, whereas $Z=61$; therefore, $Z\gg Z_c$. Consequently, the data are not incompatible with the assumption that the β^--emission of Pm^{147} is first forbidden with $\Delta J=0\pm1$.

Sm^{151} (Half-Life 20 Years: $ft=7.5\times10^6$)

Two sources of Sm^{151} were prepared from material obtained from Dr. M. G. Inghram of the Argonne National Laboratory. Because of the extremely high activity of the material, it was possible to prepare sources weighing less than 0.03 mg/cm^2. Figure 7 is a Fermi plot of the data. An end point of 0.0755 ± 0.001 Mev was obtained. Using a half-life of 20 years, the corresponding ft value is 7.5×10^6 which classifies it as

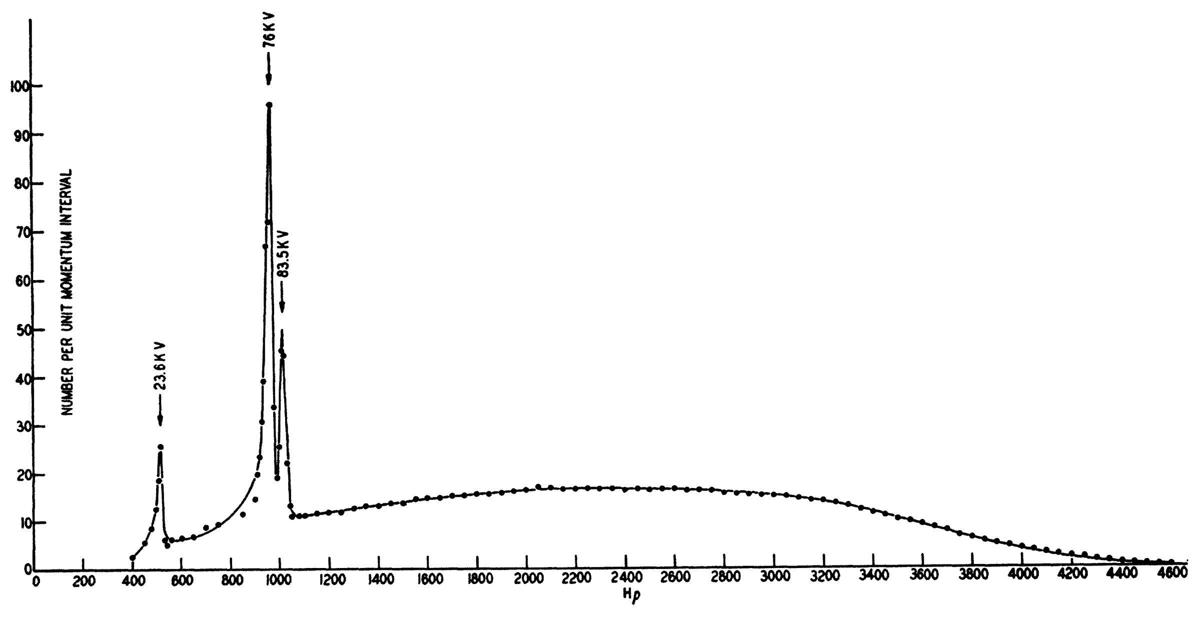

FIG. 10. Momentum distribution of the beta-rays of Tm^{170} showing the internal conversion lines of Yb^{170}.

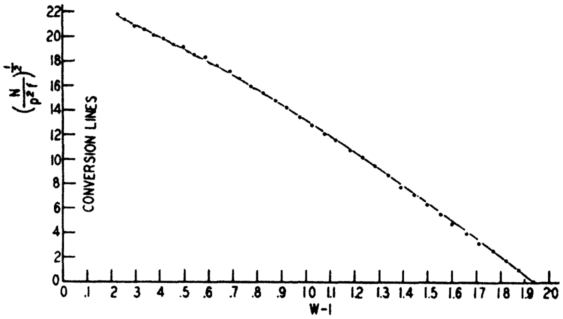

FIG. 11. Fermi plot of the beta-rays of Tm^{170}.

being first forbidden. However, Fig. 7 indicates an allowed-type Fermi plot. Z_c for Sm is 1.79. Hence, $Z \gg Z_c$ so it may be expected that the first-forbidden shape resembles the allowed shape. This would indicate that Sm^{151} is first forbidden with $\Delta J = 0, \pm 1$. Again it should be remarked that, because of the low upper energy limit of this spectrum, only a short stretch of the plot is experimentally significant. The situation is much better than in the case of Ru^{106}, but the errors involved should be realized.

P^{32} (Half-Life 14.3 Days: $ft = 8.60 \times 10^7$)

P^{32} was obtained in carrier-free form from Oak Ridge, and sources weighing less than 0.04 mg/cm^2 were prepared from it. Figure 8 shows the momentum distribution obtained. The large number of electrons at low energies was not expected, and its effect is shown clearly in Fig. 9 which is a Fermi plot of the data. From this plot, an end point of 1.718 ± 0.01 Mev was obtained. The work of Siegbahn[9] presents a Fermi plot which is only given down to 150 kev; however, there seems to be an indication of the rise which is so noticeable in Fig. 9. This rise is also evident in the momentum plot given by Lawson.[14] From the end point of 1.718 ± 0.01 Mev, P^{32} has an ft value of 8.60×10^7, which places it in the second-forbidden group. The P^{32} was followed for five half-lives with no change in the Fermi plot. Consequently, Fig. 9 must represent the spectrum of P^{32}. If it is complex with a low energy group, then S^{32} must have an isomeric state in which a gamma-transition is strictly forbidden since none is observed. On the basis of the assumption that it is not complex, an attempt has been made to apply a correction factor of the type C_{2j} to the data. The use of the factor c alone did not give a straight line; hence, this rules out the possibility of $\Delta J = \pm 3$(no). Since for these selection rules C_{2A} and C_{2T} depend upon the factor c alone. The use of the factor D_- was of no value, ruling out C_{2S} and C_{2P} corresponding to pure scalar and pseudoscalar interactions. This also rules out C_{2A} for $2 \leftrightarrow 0$ transitions. The use of the factor a alone is also unsatisfactory, ruling out the possibility of P^{32} being first forbidden with $\Delta J = \pm 2$(yes). The factor A_- alone was unsatisfactory, ruling out C_{1S} and C_{1P}. An attempt was made to use C_{2A} assuming various values for $(S_{ijk}/T_{ij})^2$, but no satisfactory fit was obtained.

Tm^{170} (Half-Life 127 Days: $ft = 7.05 \times 10^8$)

Tm^{170} was also obtained from Dr. M. G. Inghram of the Argonne National Laboratory, and sources were prepared which weighed less than 0.03 mg/cm^2. Figure 10 shows the momentum distribution obtained. Figure 11 is a Fermi plot of the data. The internal conversion lines correspond to a gamma-ray having an energy of 85.4 ± 0.6 kev being emitted by the Yb^{170} decay product. From the Fermi plot, an end point of 0.990 ± 0.007 Mev has been obtained which corresponds to an ft value of 7.05×10^8. This places Tm^{170} in the second-forbidden group. The Fermi plot is slightly concave toward the energy axis and an attempt has been made to apply one of the C_{ij} correction factors. The factors a and c distort the curve considerably thus ruling out the possibility of Tm^{170} being first forbidden with $\Delta J = \pm 2$(yes) or second forbidden with $\Delta J = \pm 3$(no). The factor D_- does not give satisfactory results nor does the factor A_-, thus ruling out C_{2S}, C_{1S}, and C_{1P}. For Tm^{170}, $Z_c = 4.48$, hence $Z \gg Z_c$. Consequently, there are really only two choices of C_{2j}; either $C \sim c$ or $C \sim (W_0 - W)^2$, neither of which is satisfactory in this case. It may be that Tm^{170} is actually first forbidden, and with $Z \gg Z_c$, the Fermi plot could resemble an allowed one. These results are in agreement with those recently reported by Saxon.[15]

ACKNOWLEDGMENTS

It is a pleasure for me to express my appreciation to Professor Enrico Fermi for his continued advice and encouragement. I am indebted to Professor Herbert L. Anderson for his interest in this work, and for initiating the construction of the spectrometer. I also wish to thank Mr. C. Y. Fan for his assistance in the latter stages of the work.

I wish to acknowledge the use of the tables of screening factors and the tables of $f(Z, W)$ made available to me by Dr. John Reitz and Mr. Steven Moszkowski.

A major portion of this work was made possible through the use of materials made available by the Isotope Division of the Oak Ridge Laboratory.

[14] J. L. Lawson, Phys. Rev. **56**, 131 (1939).

[15] D. Saxon and J. Richards, Phys. Rev. **76**, 186 (1949).