The resultant curve is shown in Fig. 4 (curve C) and is the proton yield attributed to $K^{41}(d,p)K^{42}$. The Q-values calculated from this curve are less accurate because of the graphical subtraction. An additional curve of the $K^{41}(d,p)K^{42}$ spectrum of protons was obtained by comparing the yield from a target of natural KCl with the yield from a KCl target enriched in K^{41}. This curve verified the results shown in Fig. 4, curve C.

The Q-values from $K^{41}(d,p)K^{42}$ are listed in Table IV. The mass difference between K^{42} and K^{41} computed from Q_0 is $K^{42}-K^{41}=1.00109\pm0.00010$ mass units.

The spacings between the levels in K^{42} are similar to those in K^{40} and the relative yields of the proton groups show similar variations. Many more levels in K^{42} exist above Q_3 but the proton spectrum was so complicated that they could not be accurately located with the available resolution.

TABLE IV. Q-values from $K^{41}(d,p)K^{42}$ and energy levels in K^{42}.

	Q	Excitation of K^{42}	Relative proton yield
Q_0	5.12±0.10 Mev	0 Mev	1
Q_1	4.50±0.12	0.62±0.07	1.2
Q_2	3.94±0.12	1.18±0.07	0.9
Q_3	3.15±0.18	1.97±0.15	0.9
Q_4	2.83±0.12	2.29±0.07	7.5

ACKNOWLEDGMENTS

The author wishes to express his appreciation to Professor E. C. Pollard who suggested this problem and who constantly assisted with valuable discussion and advice. Professor R. F. Humphreys and H. T. Motz also contributed much valuable information the treatment of data.

Low Energy Beta-Ray Spectra: Pm^{147} S^{35} *

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(Received November 7, 1949)

The beta-spectra of S^{35} and $_{61}Pm^{147}$ have been measured in order to study further the nature of any low energy deviation from the Fermi theory of beta-decay. Measurements were made with thin, relatively uniform sources in both the 40-cm radius of curvature spectrometer and also in a small 180 degree focusing Helmholtz coil spectrometer designed specifically for low energy spectra. The thinnest sources were less than 10 micrograms/cm². Using Zapon counter windows ranging from 15 to 3 micrograms/cm² and also a windowless counter technique, Fermi plots were obtained which showed how the measured distribution of particles at low energy depends on both source and counter window thickness. Favorable experimental conditions yielded a straight line Fermi plot for Pm^{147} above 8 kev. Less favorable conditions resulted in a straight line plot for S^{35} down to at least 50 kev. Thus, S^{35}, which is allowed, and Pm^{147}, which is probably once forbidden, are found to have spectra of the allowed shape. It is concluded that under very favorable experimental conditions there is probably no real disagreement between the observed momentum distribution and that predicted for an allowed transition by the Fermi theory. On the basis of an improved calibration, the following end points are obtained: Pm^{147}, 223.2±0.5 kev and S^{35}, 167.0±0.5 kev.

I. INTRODUCTION

RECENT experimental investigations,[1-4] particularly of the beta-ray spectra of Cu^{64}, Cu^{61}, N^{13}, and S^{35}, have suggested possible deviations from the Fermi theory in the low energy region. Some experiments[3, 5-7] indicated that this excess of low energy particles might be a function of source thickness. Other experiments showed no observable increase in the number of low energy particles as the thickness of the source was varied.[2] Still other investigations[8-10] resulted in straight line Fermi plots extending to quite low energies even when extremely thick sources were employed.

Further studies in the low energy region were undertaken in order to clarify this question and in order to try to determine the extent to which instrumentation may distort the shape of a spectrum. Some instrumental effects which may influence the shape of a spectrum at low energies are: source thickness and uniformity, source backing, scattering from the residual gas and walls of the spectrometer chamber, distortion of the analyzing field by saturation or remanence, counter window thickness, and counter response as a function of particle energy. In the present investigation, thin and relatively uniform sources deposited on extremely thin backings were employed, and the experiments were designed so as to make the other possible distortion factors negligible. Detailed measurements were made on the spectra of the allowed S^{35} transition (87 day) and the empirically once forbidden Pm^{147} transition (3.7 year) under favorable experimental conditions in both the 40-cm radius of curvature spectrometer[11] and in a small Helmholtz coil spectrometer designed specifically for low energy measurements.

* Assisted by the Joint Program of ONR and AEC.
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10 D. E. Alburger, Phys. Rev. **75**, 1442 (1949).
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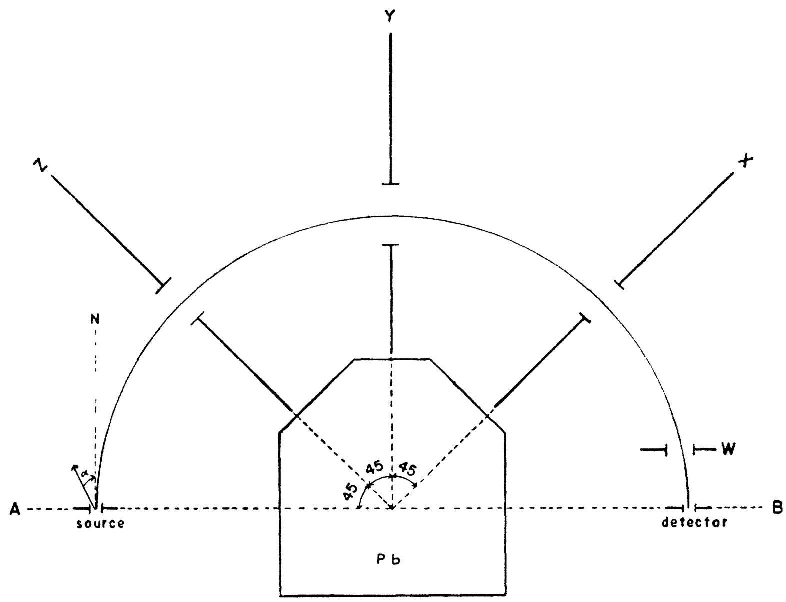

FIG. 1. Schematic arrangement of baffles and slits in small Helmholtz coil spectrometer.

II. EXPERIMENTAL METHOD

A. The Helmholtz Coil Spectrometer

The feasibility of using a windowless Geiger counter in a spectrometer to measure low energy spectra was first investigated. This necessitates a constant supply of gas to the counter and rapid pumping to remove the diffusing gas from the spectrometer proper. To this end, ethyl alcohol at 20-mm Hg pressure was used as the counting medium. This was supplied by evaporation from a constant temperature bath controlled at 23°C at which the vapor pressure of alcohol is 45 mm. The adjustment to a dynamic pressure of 20 mm was made with a Hoke valve and monitered with a manometer.

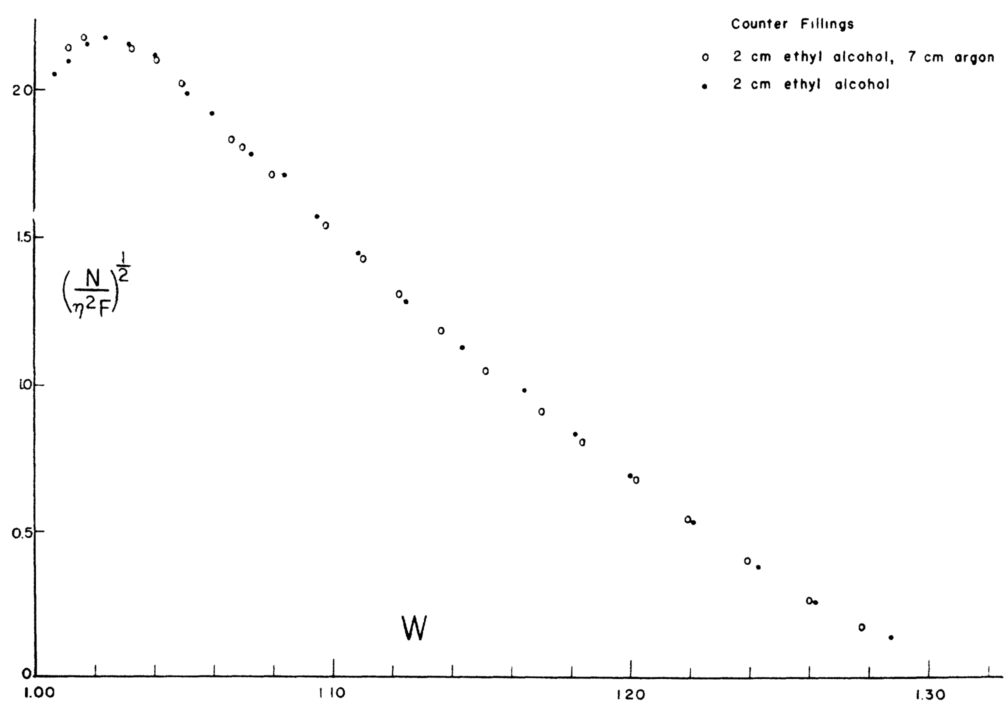

FIG. 2. Fermi plot of momentum distribution of C^{14} electrons. The open circles are for data recorded with a counter filled with a normal mixture of 2-cm alcohol plus 7-cm argon. The closed circles are for data obtained under identical conditions except that the counter filling was 2-cm alcohol only.

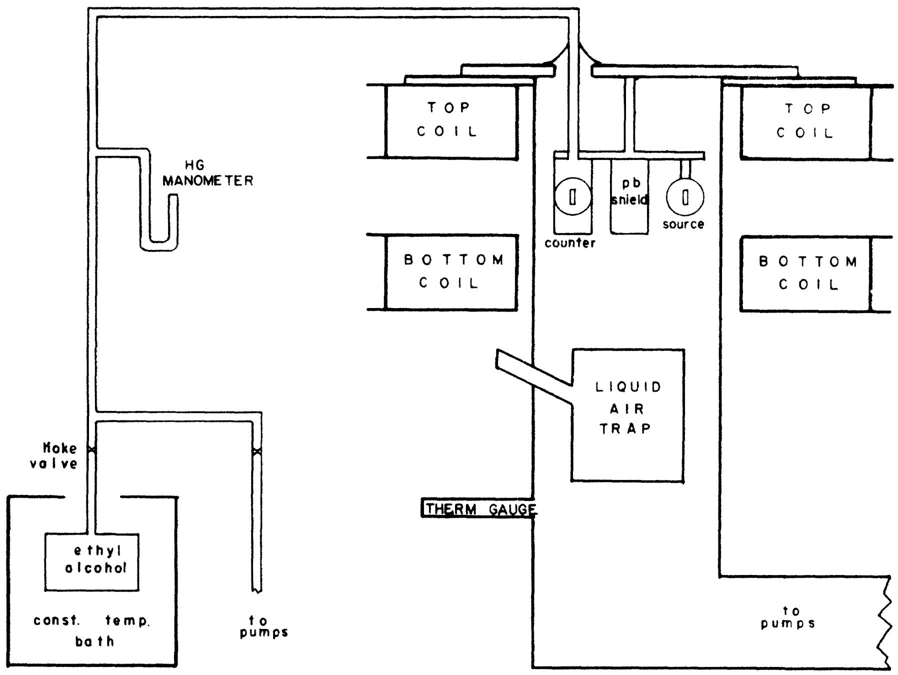

FIG. 3. Schematic arrangement of components in Helmholtz coil spectrometer.

Instead of a window, a detector slit 0.5 mm×10 mm located in the side of the counter separated it from the main spectrometer chamber. Pumping continuously with a liquid air trap and an oil diffusion pump, the pressure gradient was measured to be such that the constant pressure of 20-mm Hg in the counter dropped to 0.1-mm Hg at a distance 0.8 mm away and to a fairly constant value somewhat less than 0.01-mm Hg for distances greater than 20 mm from the slit. An approximate integration indicated that this pressure gradient was equivalent to a window thickness of about 1.5 micrograms/cm^2. A mean radius of 5.0 cm was chosen for the electron path so as to avoid excessive scattering of the beam by gas molecules. At this radius, electrons with an energy of 230 kev can be focused by the maximum available field of 360 gauss. As shown in Fig. 1, aquadag coated polystyrene baffles Z, Y, and X were placed at 45°, 90°, and 135° respectively to line AB joining the source and detector slits. The defining slit is contained in baffle Y, while the baffles Z and X, with slit widths about $\frac{1}{32}$ inch larger than the width of the beam, help reduce wall scattering. An additional baffle W was located near the counter slit to reduce wall scattering further. The fact that the top and bottom of the spectrometer are far removed from the beam is also helpful in minimizing scattering. Other possible sources of scattering in a conventional spectrometer are the edges of the slits and baffles. In this case, since the instrument is designed for use only at low energies, these edges could be restricted to a thickness of $\frac{1}{8}$ inch. With a 2-mm wide source, the resolution was calculated to be 2.5 percent. The experimental resolution of the thorium B line, which was used for calibration, was 2.6 percent for the full width at half-maximum. The transmission of the spectrometer was calculated at 0.1 percent.

Helmholtz coils were used to produce the field since they avoid the effect of residual ferromagnetism at the weak fields employed. Each coil was layer wound with 15,600 turns of round No. 22 Formex wire on brass spools with a rectangular cross section 3.25 inches×3.84 inches. Optimum coil separation was found to be 125 mm between coil centers. Under these conditions the field in the median plane showed a variation of less than 0.1 percent in the 20-mm range about the 50-mm mean radius, and no measurable variation within 10 mm above and below the median plane. The current for the Helmholtz coils was provided by an electronic constant current supply such that the coil current could be held constant over a period of at least an hour to better than one part in ten thousand.

It was calculated that counters filled with ethyl alcohol at 20-mm Hg should count with constant efficiency all electrons with energies up to at least 200 kev. In a preliminary experiment, an end window counter with its window removed was placed directly above a C^{14} source in a small vacuum chamber. The counter was tested with alcohol fillings ranging from 10-mm to 40-mm Hg, all of which gave satisfactory results. The counter plateau ranged from 50 to 100 volts depending on the pressure and purity of the alcohol vapor. For a high enough operating voltage and pure alcohol vapor the pulses were clean and of uniform height. The operating voltage for an alcohol filling of 20 mm was 1100 volts. Total efficiency was first checked in the above arrangement by establishing that the same total number of counts was recorded in a given time by the counter filled with alcohol vapor (at 20 mm) and by the counter filled with the normal mixture of 20-mm alcohol and 70-mm argon. A check on the counter efficiency as a function of energy over the range up to 150 kev was accomplished by comparing the beta-spectra of C^{14} obtained in the Helmholtz coil spectrometer with its side window counter filled to the same pressures as used above in the end window counter.

A Fermi plot of the C^{14} spectrum for each case is shown in Fig. 2. It is apparent from the close agreement of the two curves (no adjustments were made) that the alcohol counter may be considered 100 percent efficient up to at least 150 kev. The shape of the Fermi plot in this case has no absolute significance since the source used was thick.

The counters used in the spectrometer were cylindrical side window counters with an inside diameter of 1 inch and a length of 3.625 inches. The central wire was 0.005-inch diameter tungsten. Side window counters offer a more positive detection of low energy particles than end window counters because of the finite distance particles must travel past an end window to feel the full electric field. For the windowless counter the slit was 0.5 mm by 10 mm covered by copper Lektromesh[12] grid, 0.004 inch thick having 0.006 inch openings which yields a transmission of 23 percent. Similar counters were used with Zapon windows down to 3 micrograms/cm^2; however, in this case the slit was 10 mm by 2.0 mm.

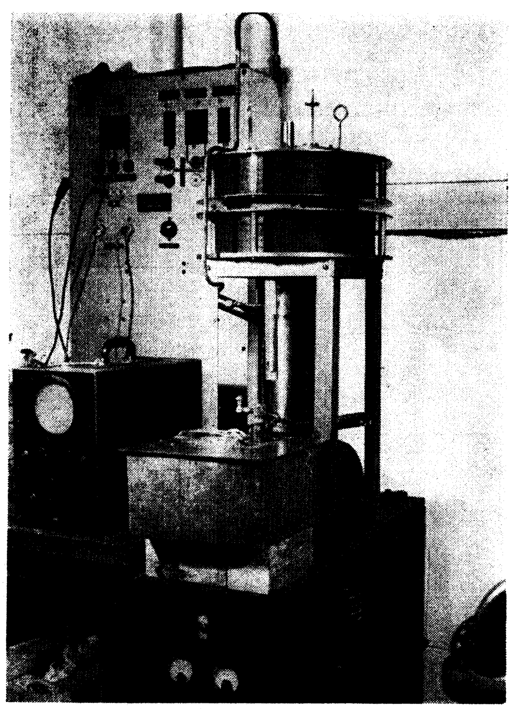

FIG. 4. Helmholtz coil spectrometer.

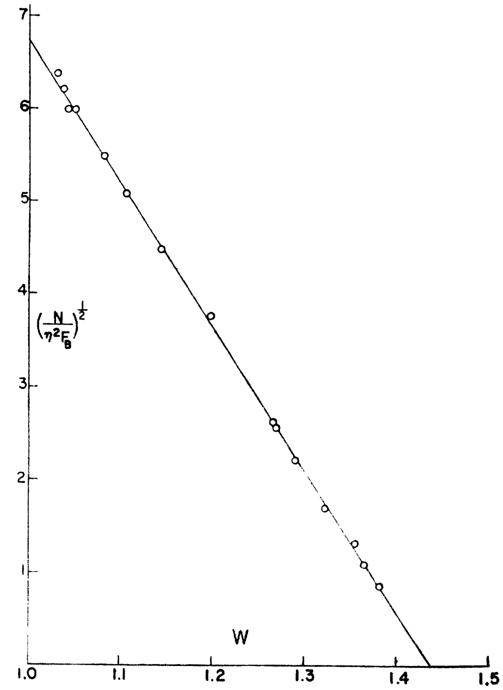

FIG. 5. Fermi plot of data for Pm^{147} spectrum obtained with the large shaped field spectrometer using a 6-microgram/cm^2 window supported on a Lektromesh grid. The source was about 10 micrograms/cm^2 thick.

It was possible to make counters with Zapon windows as thin as 1.5 micrograms/cm^2. However, the excessive rate of diffusion through such windows makes the counters inconvenient to use unless special means are taken to keep the pressure constant.[13] Figure 3 shows a schematic drawing of the experimental arrangement. Figure 4 shows a photograph of this spectrometer.

B. The 40-Cm Radius of Curvature Spectrometer

This spectrometer was adjusted for a resolution of 0.5 percent.[11] Spectra were recorded with counters having Zapon windows of 6 micrograms/cm^2 and 3 micrograms/cm^2 supported by a Lektromesh[12] grid or a Lucite-tantalum combination of a type described elsewhere.[11] The counters with the thinner windows were filled with a mixture of 20-mm ethylene and 10-mm argon, and the others at higher pressures.

In addition to its high resolution, this spectrometer is particularly suited for low energy measurements because of negligible wall scattering and a large source area (4 mm×25 mm at the above resolution) which makes it possible for a source of given intensity to be spread relatively thin.

[12] Obtainable from C. O. Jelliff Manufacturing Corporation, Southport, Connecticut.

[13] Ter-Pogossian, Robinson, and Townsend, Rev. Sci. Inst. 20, 289 (1949).

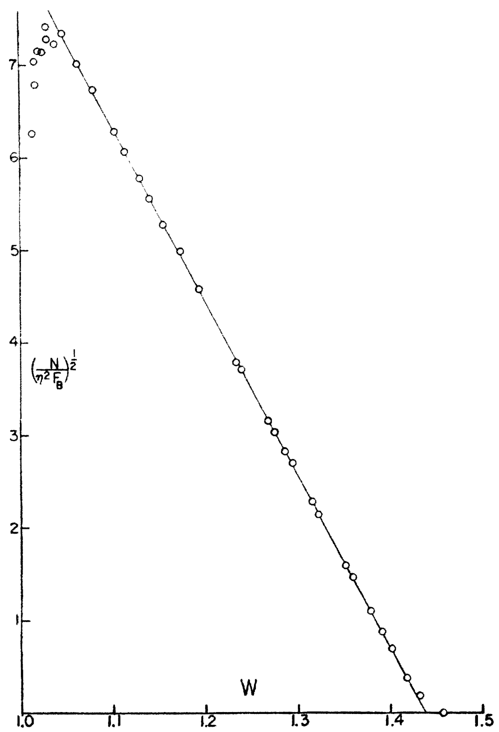

FIG. 6. Fermi plot of the Pm^{147} spectrum obtained with the large shaped field spectrometer using a tantalum grid and a 6-microgram/cm² Zapon window. The source was about 10 micrograms/cm² thick.

C. Preparation of Sources

In view of the conflicting reports in the literature on the effect of source thickness, we made a study of autoradiographs of sources prepared from chemical solutions. By this technique it was found that in general such sources, though appearing uniform, may in many cases have variations of intensity of as much as one hundred-fold. Under these circumstances the *average* source thickness as reported by various investigators does not have much meaning. We are not as yet able to deposit completely uniform sources from chemical solution. Our best technique consists of wetting the portion of the backing foil on which the source is to be placed with one drop of 5 percent solution of Lilly Insulin (40 units per cc) in water. A drop of radioactive solution applied any place in this area will spread over the entire region defined by the insulin. In the case of water solutions, it seems to be of no consequence whether or not the insulin layer is first dried. However, after the application of the radioactive material we find it best to quickly dry the source under an infra-red lamp, preferably while gently agitating (rocking) the liquid sample in order to prevent the formation of large crystals. Calibration of the medium Eastman Kodak lantern slide plates used for obtaining autoradiographs showed that sources could be made in this manner which were uniform to within a factor of four. In every case a narrow strip of 0.18 milligram/cm² grounded aluminum leaf was placed near the end of the deposit to minimize electrostatic effects of the source's charging on the dielectric film.

In the case of the narrow 1-mm and 2-mm sources for the Helmholtz coil spectrometer the width was controlled by applying full strength insulin from a stainless steel stylus. With the aid of a ruling jig, the stylus was placed directly above the Zapon film so that a suspended drop of insulin just touched the film, and finally, by moving the stylus across the film a narrow insulin line was obtained. A small drop of the liquid source was then applied and dried as before. Fairly uniform sources are easily made on larger areas by covering the region with narrow insulin lines and putting a small drop of liquid source on each. Extremely uniform sources can, if the activity permits, be successfully prepared by thermal evaporation in vacuum.

III. RESULTS

A. The 40-Cm Radius of Curvature Shaped Field Spectrometer

Figure 5 shows the Fermi plot for the spectrum of Pm^{147}. N is the number of particle per minute per unit momentum interval normalized to constant detector slit acceptance by dividing the actual number detected at any magnetic field strength by the value of the field. η is the electron momentum in units of m_0c and W is the electron energy in units of m_0c^2. F_B is the Coulomb factor which has been evaluated by means of the very good approximation given by Bethe and Bacher.[14] This data is for a typical run obtained with a source of about 10 micrograms/cm² and a Lektromesh grid counter with a 6-microgram/cm² window and filled with 20-mm ethylene and 10-mm argon.

Figure 6 shows data from the same source recorded in a tantalum grid counter with the same window

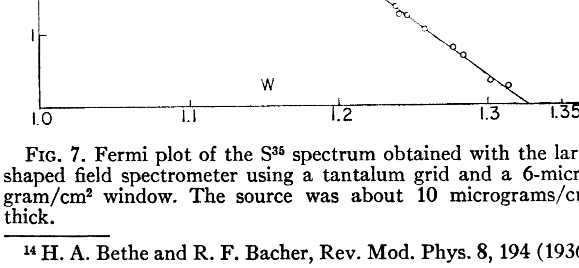

FIG. 7. Fermi plot of the S^{35} spectrum obtained with the large shaped field spectrometer using a tantalum grid and a 6-microgram/cm² window. The source was about 10 micrograms/cm² thick.

[14] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. 8, 194 (1936).

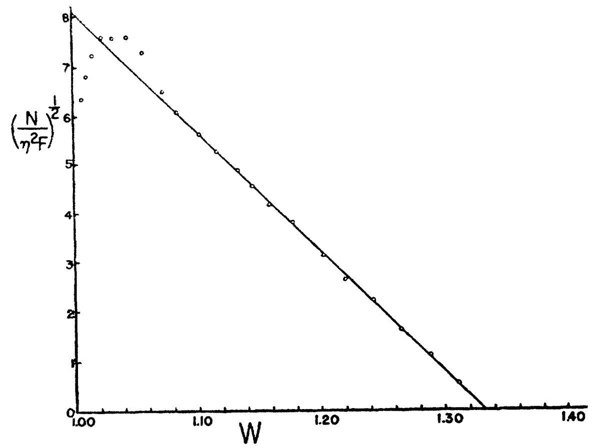

FIG. 8. Fermi plot of the S^{35} spectrum obtained with the small Helmholtz coil spectrometer.

thickness but filled to a higher pressure. The Fermi plot is certainly straight back to at least 20 kev and the end point is well determined at 223.2±0.5 kev (both verified by several other runs not shown). This may be compared with the 223 kev reported by Levy.[15]

Figure 7 shows the Fermi plot of the data for S^{35} obtained with a source of 10 micrograms/cm^2. One sees this plot is straight in agreement with the Fermi theory from the end point to energies at least as low as 50 kev, a lower value than was obtained with thicker sources.[3] Wu and Albert[5] report a rise at 100 kev for a source of 5 micrograms/cm^2 which may occur because of the difficulty of preparing uniform sources from chemical solutions as previously discussed in this paper. The new value of the end point of 167.0±0.5 kev is based on an improved calibration of the instrument. This may be compared with the 166 kev found by Wu and Albert.[5]

B. The Helmholtz Coil Spectrometer

Figure 8 shows the Fermi plot for the S^{35} spectrum obtained with a source of about 50 micrograms/cm^2 and a Zapon counter window of 3 micrograms/cm^2. The end point obtained with the large instrument is confirmed and the plot appears straight back to 40 kev.

Figure 9 shows Fermi plots obtained with the above counter for various thicknesses of Pm^{147} sources. The

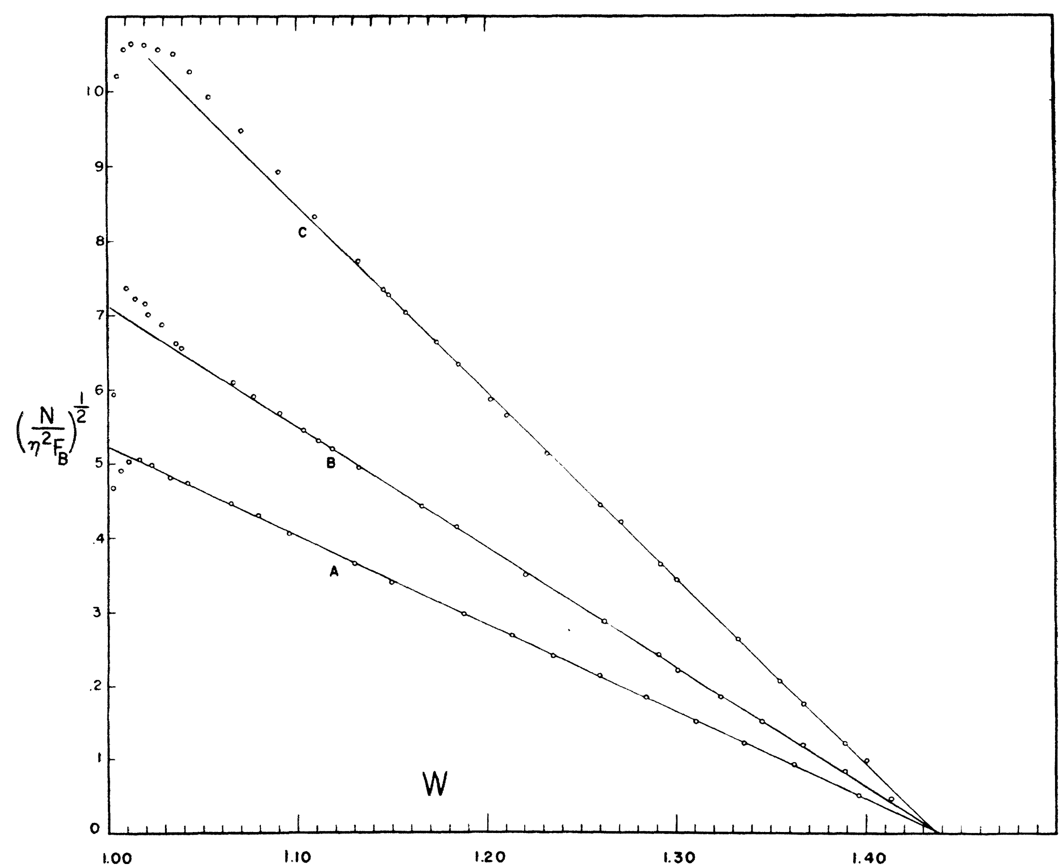

FIG. 9. Fermi plots of the Pm^{147} spectra obtained in the small Helmholtz coil spectrometer from sources of different thickness. Curve A is for a source somewhat less than 11 micrograms/cm^2. Curve B is for a source of about 43 micrograms/cm^2. Curve C is for a source somewhat thicker than 50 micrograms/cm^2.

[15] P. W. Levy, Plutonium Project Report, Mon. P-228, p. 29 (December, 1946).

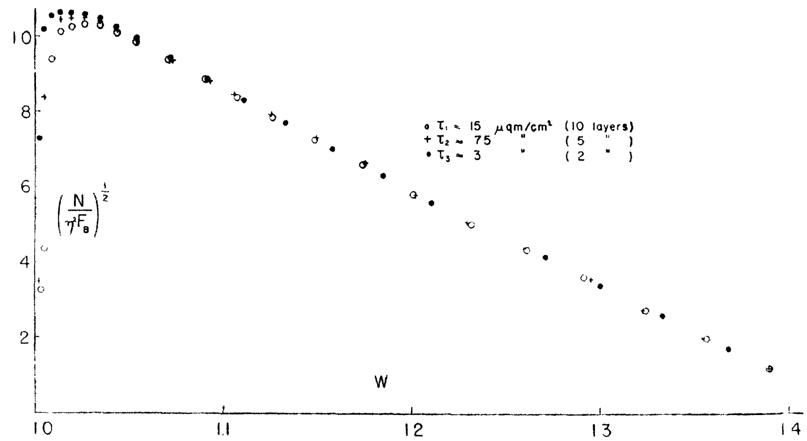

FIG. 10. Fermi plot of Pm^{147} spectra obtained in the small Helmholtz coil spectrometer showing effect of counter window thickness.

backings were all 1.5 micrograms/cm² Zapon. For the thinnest source, less than 11 micrograms/cm², there is no deviation from the straight line allowed shape above 8 kev. As the source thickness is increased the energy at which a deviation occurs also rises. The value of the end point is well confirmed.

Figure 10 shows the effect of window thickness on the measured spectral distribution of Pm^{147}. These data were taken with the thickest source (somewhat thicker than 50 micrograms/cm²) shown in Fig. 9. One sees clearly the expected effect of increased particle absorption at low energies for increasing counter window thickness.

For the same source, Fig. 11 compares the observed distribution with and without counter window. The Fermi plot of the data taken with a 3-microgram/cm²

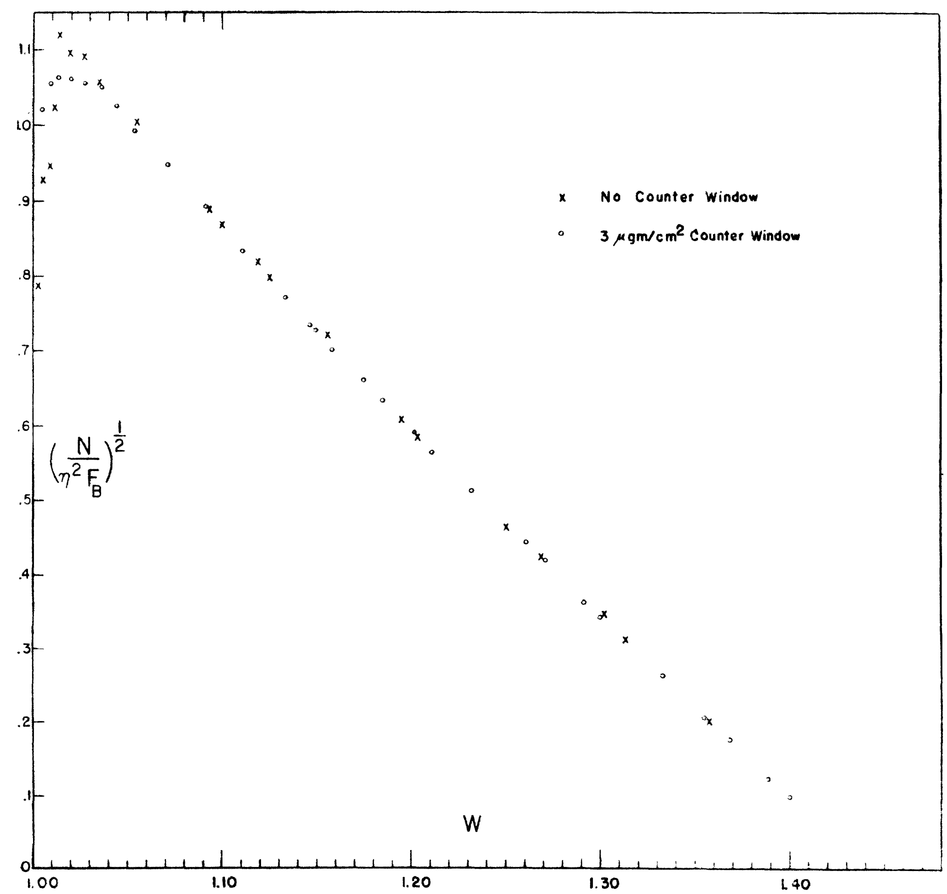

FIG. 11. Fermi plot of Pm^{147} spectra obtained in small Helmholtz coil spectrometer with a 3-microgram/cm² window and with the windowless counter.

window is in good agreement with that obtained with no window down to about 8 kev, where noticeable window absorption apparently sets in. The windowless counter continues to accept most electrons down to about 6 kev where the diffusing gas seems to become effective as a "window." The crossing of the curves at extremely low energy is probably not real but arises from poor statistics at the lowest energies.

IV. CONCLUSIONS

Fermi plots of the beta-spectra of S^{35} and Pm^{147} yield extrapolated end points of 167.0±0.5 kev, and 223.2 ±0.5 kev, respectively.

The S^{35} spectrum is in agreement with the Fermi theory for an allowed transition at least for energies above 50 kev. Wu and Albert[5] found the excess of particles at low energies to be a function of source thickness. For an average source thickness reported at 1 microgram/cm^2 their curve appears straight back to at least 20 kev. Coupled with the present findings on the effect of source thickness, S^{35} appears to offer no contradiction to the Fermi theory.[16]

The Pm^{147} Fermi plot is indistinguishable from a straight line for all energies above 8 kev. Empirically forbidden with a $ft=4.3\times10^7$, it is probably at most once forbidden since an allowed shape is highly improbable for higher forbidden transitions.[17] An allowed shape for a once forbidden transition may be taken to infer that the spin change is not two units which would be expected to lead to the unique first forbidden shape found recently.[18]

According to the nuclear shell scheme of M. G. Mayer,[19] the disintegration of $_{61}Pm^{147}$ into $_{62}Sm^{147}$ may be described as a transition from a $g_{7/2}$ to a $h_{9/2}$ state involving a change of parity. This is consistent with a once forbidden transition involving a spin change of one unit.

Thin uniform sources are difficult to prepare, and their attainment can be most reliably confirmed by some measurement of the relative activities of the various parts of the source, such as the autoradiographs described, coupled with a mass determination. Reasonably uniform sources can be made by the method described and extremely uniform sources result from thermal evaporation in vacuum.[16] Inasmuch as sources prepared by chemical deposit may in general be extremely non-uniform, the *average* thickness as reported in different laboratories does not have much absolute meaning. This undoubtedly accounts for the different results found at low energy by different observers working with sources which were reported to be of comparable *average* thickness.

Counters filled with ethyl alcohol at a pressure of 20-mm Hg can detect electrons up to at least 220 kev with 100 percent efficiency. This filling may be used with Zapon counter windows of 3 micrograms/cm^2 without excessive diffusion. Windowless counters of 100 percent efficiency may be used under the conditions of our experiment with an "effective window" of as little as 1.5 micrograms/cm^2. Some electrons with energies less than 1 kev are detected by both methods.

Thickness of counter window has the qualitatively expected effect at low energies of increasingly absorbing more particles as the window is made thicker. A 1.5-microgram/cm^2 "effective window" shows no indication of spectral disturbance above 6 kev; a 3-microgram/cm^2 Zapon window, none above 10 kev; a 6-microgram/cm^2, none above 20 kev; a 7.5-microgram/cm^2, none above 30 kev; a 15-microgram/cm^2, none above 80 kev. It is interesting to note that under certain conditions a thick counter window can just cancel the excess of low energy particles which arises because of source thickness. Partial transmission through the corners of a counter window supporting grid may have the same effect.

This work was partially supported by a grant from the Frederick Gardner Cottrell fund of the Research Corporation and by the Joint Program of ONR and AEC. The radioactive S^{35} and Pm^{147} were obtained from Oak Ridge.

[16] Since the completion of the work reported in this paper, measurements made on Cu^{64} with extremely thin and uniform sources prepared by evaporation of metallic copper in vacuum show no disagreement with the Fermi theory. Langer, Moffat, and Price, Phys. Rev. **76**, 1725 (1949); G. E. Owen and C. S. Cook, Phys. Rev. **76**, 1726 (1949).

[17] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **60**, 308 (1941).

[18] L. M. Langer and H. C. Price, Jr., Phys. Rev. **76**, 641 (1949).

[19] M. G. Mayer, Phys. Rev. **75**, 1969 (1949).

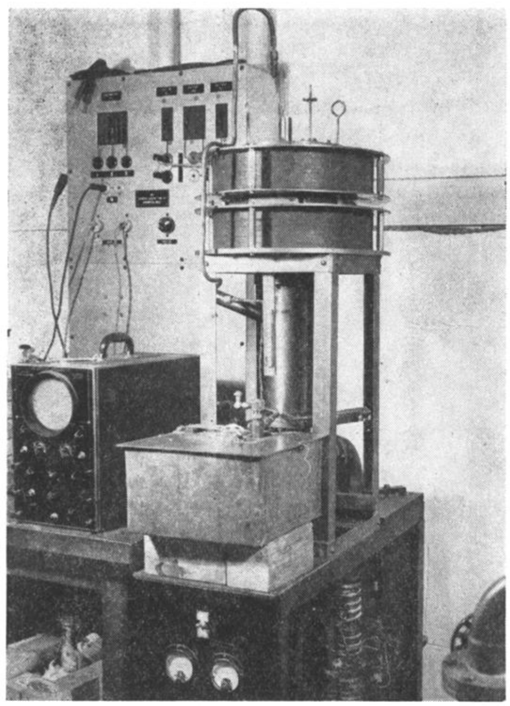

FIG. 4. Helmholtz coil spectrometer.