

## Study of the Beta-Spectra of $C^{14}$ and $S^{35}$

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Using sources of high specific activity, the beta-spectra of  $C^{14}$  and  $S^{35}$  have been investigated in a large magnetic spectrometer. Since the distributions are measured under essentially identical conditions, a direct comparison of the data is possible. The  $S^{35}$  spectrum is in agreement with that predicted by the Fermi theory for energies from  $W=1.15$  mc<sup>2</sup> to the end point at  $W_0=1.331$  mc<sup>2</sup>. Below this region, there is a deviation from the theory the exact nature of which is unfortunately somewhat masked by distortions due to source thickness and backing. The  $C^{14}$  spectrum has a shape which is slightly different from that which would be predicted for an allowed transition. The experimental shape is not consistent with that calculated for a second forbidden transition using Fermi selection rules. The extrapolated end point for  $C^{14}$  is at  $W_0=1.306$  mc<sup>2</sup>. From the shape of the  $S^{35}$  spectrum in the neighborhood of the end point, an upper limit is obtained for the mass of the neutrino as less than one percent the mass of the electron.

### 1. INTRODUCTION

THE momentum distribution of the neutrons emitted by  $C^{14}$  and  $S^{35}$  have been investigated in a magnetic spectrometer under rather favorable experimental conditions. Both of these isotopes emit beta-particles having approximately the same maximum energy and since the measurements of the spectra were accomplished under essentially identical conditions, a comparison of the results for the two elements may be made directly. The half lives are such that the 87 day  $S^{35}$  involves an allowed transition, whereas the 5100-year  $C^{14}$  is at least second forbidden.

Several experimenters<sup>1-7</sup> have made attempts to determine the end point energies of these transitions. The main purpose of the present investigation was to study the shape of the momentum distributions and to obtain some additional information on the influence of source thickness and backing on the experimental measurement of such low energy spectra.

Previous results<sup>8</sup> indicate that an allowed

transition does not necessarily yield a spectrum in agreement with the Fermi theory at low energies. Furthermore, many forbidden transitions which might be expected to yield spectra quite different from the allowed shape seem to give a straight line Fermi plot over a large part of the high end of the distribution.<sup>9</sup>

### 2. EXPERIMENTAL METHOD

The spectra were investigated in a high resolution, 40-cm radius of curvature, shaped field magnetic spectrometer.<sup>10</sup> The large size of this instrument and the use of strategic baffles make the effects of scattering completely negligible.

The same end window G-M counter which was used for the measurements on the shape of the  $Cu^{64}$  spectra,<sup>8</sup> was used for the measurements on  $S^{35}$  and  $C^{14}$ . It has a very thin Zapon window capable of transmitting electrons having an energy as low as 2.0 kev. The supporting grid for the Zapon foil is of such a thickness and design that the transmission is not dependent on energy.

In order to obtain adequate intensity from a thin source, a source and detecting slit were used which were somewhat wider than those used for the  $Cu^{64}$  work. In the present investigation sources 0.5 to 1.2 cm wide were used in conjunction with a detector slit width of 0.6 cm. Because of the large value of the radius of curva-

<sup>1</sup> A. K. Solomon, R. G. Gould, and C. B. Anfinsen, *Phys. Rev.* **72**, 1097 (1947).

<sup>2</sup> S. Ruben and M. D. Kamen, *Phys. Rev.* **59**, 349 (1941).

<sup>3</sup> P. W. Levy, *Phys. Rev.* **72**, 248 (1947).

<sup>4</sup> M. N. Lewis and M. Paul, *Phys. Rev.* **73**, 1269 (1948).  
 W. E. Stephens and M. N. Lewis, *Phys. Rev.* **72**, 526 (1947).

<sup>5</sup> W. F. Libby and D. D. Lee, *Phys. Rev.* **55**, 245 (1939).

<sup>6</sup> A. K. Saha, *Proc. Nat. Inst. Sci. India*, **12**, 159 (1946).

<sup>7</sup> M. D. Kamen, *Phys. Rev.* **60**, 537 (1941).

<sup>8</sup> C. S. Cook and L. M. Langer, *Phys. Rev.* **73**, 601 (1948).

<sup>9</sup> K. Siegbahn, *Phys. Rev.* **70**, 127 (1946).

<sup>10</sup> L. M. Langer and C. S. Cook, *Rev. Sci. Inst.* **19**, 257 (1948).

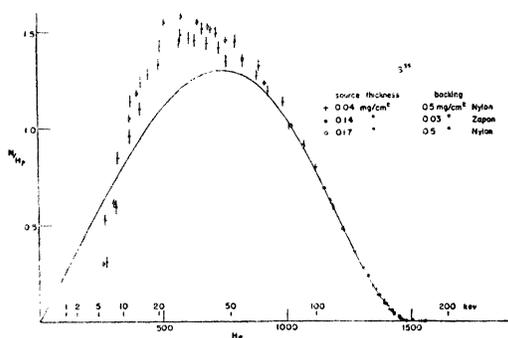


FIG. 1. Momentum distribution of the negatrons of  $S^{35}$ . The relative number per unit momentum interval is plotted as ordinate against the momentum in units of gauss-cm. The solid curve is the distribution for an allowed transition according to the Fermi theory. The experimental data have been adjusted to the same intensity at 100 kev.

ture, the resolution under these conditions is still quite adequate, e.g., of the order of one percent.

Relative measurements of the magnetic field were made with a flip coil and ballistic galvanometer. The absolute calibration is in terms of the photoelectrons ejected from a Pb radiator by the 0.5108 Mev annihilation radiation of  $Cu^{64}$ . The energizing current for the magnet is stabilized electronically, and the magnetic field remained constant to better than 0.1 percent during the one and two hour runs necessitated by the low counting rates. Several ballistic galvanometer readings were taken during each run, and the calibration of the ballistic galvanometer was checked after each such deflection against a standard mutual inductance. The magnetic field was further monitored continuously by a rotating pick-up coil whose generated voltage is inductively coupled to a crystal rectifier and type  $K$  potentiometer.

### 3. PREPARATION OF SOURCES

Both the  $C^{14}$  and  $S^{35}$  sources were obtained by  $(n,\gamma)$  reactions in the pile at Oak Ridge. The  $C^{14}$  was the very high specific activity material made available by Norris and Snell.<sup>11</sup> For each source which was prepared, a fraction of the finely divided barium carbonate sample was suspended in a few drops of alcohol. This suspension

<sup>11</sup> L. D. Norris and A. H. Snell, Phys. Rev. **73**, 254 (1948); L. D. Norris and A. H. Snell, Bull. Am. Phys. Soc., No. 3, p. 46, April 1948.

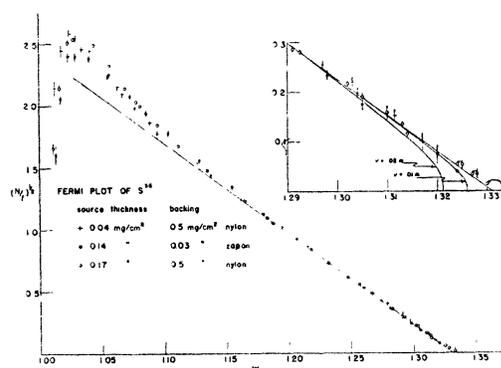


FIG. 2. Fermi plot of  $S^{35}$  negatron spectrum. The extrapolated end point is at  $W_0 = 1.331$  corresponding to a kinetic energy of 169.1 kev. The enlarged section in the vicinity of the end point shows the theoretical distribution calculated for a neutrino mass,  $\nu$ , equal to zero, one percent and two percent the mass of the electron. The arrows indicate the corrections applied for finite resolution.

was then diluted to seven times its original volume by the addition of distilled water. Several drops of the final suspension were then deposited on a  $0.02 \text{ mg/cm}^2$  Zapon backing. The source was dried under an infra-red lamp, and the barium carbonate seemed to settle out rather uniformly over the entire area. The several sources which were used were compared by measuring the counting rate in a fixed position under a thin window monitor counter. One source was weighed in order to determine its average thickness. The average thickness of the other sources was deduced from the relative activities.

The  $S^{35}$  was obtained as NaS in a 0.1 N solution of NaOH. Since it was found that a Zapon foil would not resist the NaOH, it became necessary either to use a different backing material or to neutralize the solution. Both methods were used. Some of the sources were prepared by spreading the solution of NaS in NaOH over a  $0.5 \text{ mg/cm}^2$  Nylon backing. Since this backing is considerably thicker than those usually used, it was thought desirable to investigate any effects caused by this change in the thickness of the backing. Other sources were therefore prepared by neutralizing the NaOH solution with  $HNO_3$  and spreading on  $0.03 \text{ mg/cm}^2$  Zapon. For the sulfur sources, the average thickness was calculated from the assay made at Oak Ridge and checked by weighing.

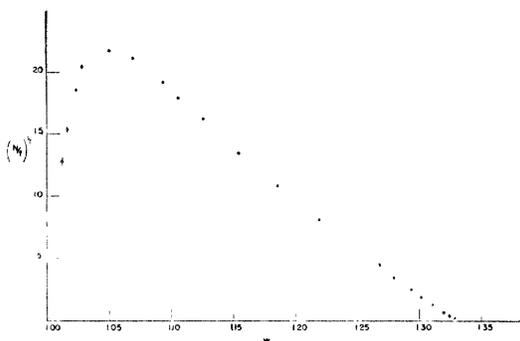


FIG. 3. Fermi plot of  $S^{35}$  spectrum for data obtained with a  $0.56 \text{ mg/cm}^2$  source on a  $0.5 \text{ mg/cm}^2$  Nylon backing.

#### 4. RESULTS

The great advantage of measuring  $C^{14}$  and  $S^{35}$  under essentially identical conditions is that in this way any difference between them may be compared, and a comparison with theory takes on added significance.

The momentum distribution of the  $S^{35}$  negatrons obtained for different source thicknesses and backings is shown in Fig. 1. The curve is the distribution predicted by the Fermi theory. Fig. 2 is a Kurie plot of the experimental data. It is significant that the experimental data for all the sources are in good agreement with the theory for all energies above  $W=1.15 \text{ mc}^2$ . Below this energy, the experimental points rise above the theoretical straight line and then fall rapidly for energies below about 15 kev. The deficiency at very low energies is probably due largely to the attenuation of the counter window.

It should be noted that it is possible under

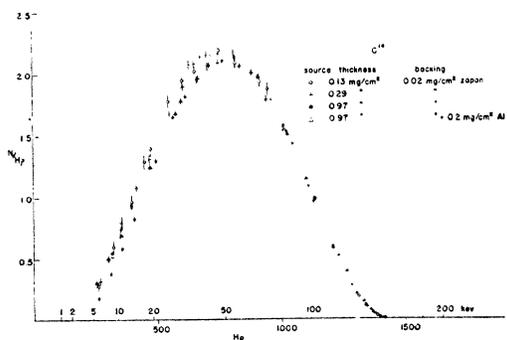


FIG. 4. Momentum distribution of the negatrons of  $C^{14}$ . The relative number per unit momentum interval is plotted as ordinate against the momentum in units of gauss-cm. The data have been adjusted to the same intensity at 90 kev.

certain conditions of thick source and backing to completely eliminate the rise observed below  $W=1.15 \text{ mc}^2$ . This effect is best illustrated in Fig. 3 which shows a Kurie plot of the data obtained with a  $0.56 \text{ mg/cm}^2$  source on a  $0.5 \text{ mg/cm}^2$  Nylon backing. The data, in this case, do not yield a straight line plot but instead there is a slight curvature away from the energy axis over the entire upper end of the distribution. If such a measurement were made with poor resolution and statistics, it might very well be misinterpreted as a straight line and the rise at lower energies would then not be recognized.

The momentum distributions of the negatrons of  $C^{14}$  are shown in Fig. 4. The corresponding Kurie plots are shown in Fig. 5. In this case it is significant that the data from all the sources give essentially identical results for all energies above  $W=1.12 \text{ mc}^2$ . In this region the Kurie plot is not a straight line but curves very slightly away from the energy axis. Since this curvature is the same for a wide variation of source thicknesses which have quite different influences at low energy, and since the sources of  $C^{14}$  were of comparable thinness and backing to those which yielded straight line plots over the same energy region for  $S^{35}$ , it is felt that this curvature in the case of  $C^{14}$  may indeed be real. One cannot, however, eliminate the possibility that this curvature may yet be caused by some unknown effect arising from the fact that the  $C^{14}$  source was prepared from a suspension whereas the  $S^{35}$  was deposited from a solution. It is quite possible that although the average thickness would indicate a "thin" source, the thickness of the individual grains may be such as to give a thick source effect.

The low energy region below  $W=1.12 \text{ mc}^2$  shows an effect caused by differences in source thickness and backing. It is seen that for the same backing, the drop in the low energy region of the Kurie plot occurs at somewhat higher energy for thicker sources. Further, for the same source thickness an increase in backing causes the fall off to occur at lower energy. This is illustrated by the solid circles and the triangles in Fig. 5. The run represented by the solid circles was taken with a source whose average thickness was  $0.97 \text{ mg/cm}^2$  on a Zapon backing of  $0.02 \text{ mg/cm}^2$ . The run represented by the triangles

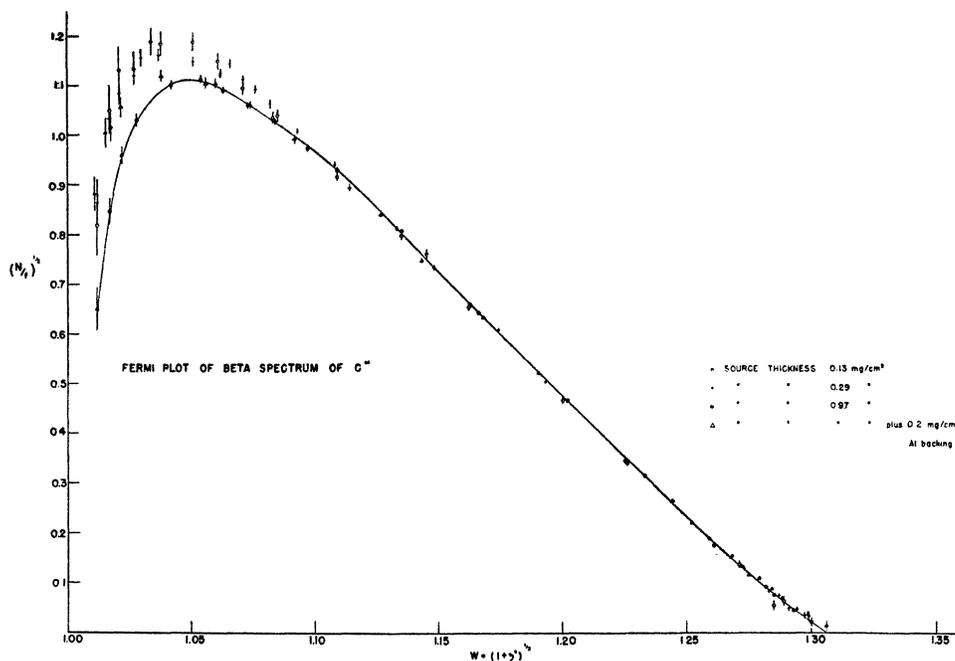


FIG. 5. Fermi plot of C<sup>14</sup> negatron spectrum. The extrapolated end point is at  $W_0 = 1.306 \text{ mc}^2$  corresponding to a kinetic energy of 156.3 kev.

was obtained with the same source after an aluminum foil  $0.18 \text{ mg/cm}^2$  had been placed in contact with the source.

The maximum energy of the negatron spectrum of S<sup>35</sup> obtained by extrapolating the straight line part of the Kurie plot is  $W_0 = 1.331 \text{ mc}^2$ . This corresponds to a kinetic energy of  $169.1 \pm 0.5 \text{ kev}$ . This value is in agreement with the values recently reported by Berggren and Osborne<sup>12</sup> and by Solomon, Gould, and Anfinsen.<sup>1</sup>

For C<sup>14</sup>, the end point obtained from the Kurie plot is  $W_0 = 1.306 \text{ mc}^2$  which corresponds to a kinetic energy of  $156.3 \pm 1.0 \text{ kev}$ . This is also in agreement with the value found by Berggren and Osborne.

Because of the finite width of the source and the detecting slit, those beta particles whose momenta are in the immediate vicinity of the end point enter the detector after traversing a trajectory having a slightly different *effective* radius of curvature. Because of the high resolution employed in the present instrument, this effect is very small. The magnitude of this correction has been calculated by integrating the theoretical

distribution over the width of the source and the detector slit. The theoretical allowed shape of the momentum distribution for zero neutrino mass was used for this calculation for S<sup>35</sup>. The shift in the position of the last three points is indicated by the arrows on the enlarged plot in Fig. 2.

## 5. CONCLUSIONS

### (a) Sulfur 35

As seen in Fig. 2, the S<sup>35</sup> spectrum shows agreement with the Fermi theory for an allowed transition for energies above  $W = 1.15 \text{ mc}^2$ . Below this energy, a rise begins which is very similar to the effect observed in the spectrum of the Cu<sup>64</sup> negatrons.<sup>8</sup> The effect involves much higher energy electrons in Cu<sup>64</sup> (up to  $1.37 \text{ mc}^2$ ) but leaves a major portion of the energy spectrum undisturbed, just as we find in S<sup>35</sup>. In the latter case, however, the real nature of the deviations from the Fermi theory is complicated by some distortion due to source thickness and backing.

The shape of the S<sup>35</sup> spectrum in the vicinity of the end point can be used to set an upper limit on the rest mass of the neutrino. In Fig. 2,

<sup>12</sup> J. L. Berggren and R. K. Osborne, Bull. Am. Phys. Soc. No. 3, p. 46, April 1948.

the region of the distribution in the neighborhood of the end point is shown on an enlarged scale. In addition to the experimental points, calculated curves are shown for the theoretical Fermi distribution for an allowed transition calculated for an assumed neutrino mass,  $\nu$ , of zero, one percent and two percent the mass of the electron. The distribution formula used here for  $N$ , the intensity in unit momentum interval, is given by:

$$Nd\eta = \text{const.} F(Z, \eta) \eta^2 K (K^2 - \nu^2)^{\frac{1}{2}} (1 - \nu/WK) d\eta$$

where  $F$  is the coulomb factor ( $F\eta^2 \equiv f$ , as used in the graphs),  $\eta$  is the electron momentum in units of  $mc$ ,  $W = (1 + \eta^2)^{\frac{1}{2}}$  and  $K$  is the neutrino energy in  $mc^2$  units. This formula includes the relativistic factor  $1 - \nu/WK$ . The sign in this factor conforms with the form of the theory as discussed by Pruett.<sup>13</sup> The inclusion of this factor makes the difference between the extrapolated end point and the theoretical true end point equal to  $\nu/2$ . If this relativistic factor is neglected, as was done by Kofoed-Hansen,<sup>14</sup> this difference would be equal to  $\nu$ .

The experimental points in Fig. 2 are consistent with a neutrino rest mass of zero. Within the limits of error one can certainly say that the mass of the neutrino is less than one percent the mass of the electron.

#### (b) Carbon 14

The  $C^{14}$  disintegration has an extremely long mean life for its energy release and is therefore classified as highly forbidden. On the other hand, recent measurements have determined the spin of  $C^{14}$  as zero.<sup>15</sup> This, together with the

known spin, 1, of  $N^{14}$ , shows that there is only a spin change of one unit in the transition. The so-called Gamow-Teller selection rules permit such a spin change to yield an allowed transition. On the basis of these rules, the  $C^{14}$  case must be regarded as an unsolved problem together with other members of the  $4n+2$  family. The Fermi selection rules permit no spin change in allowed transitions. Furthermore, if there is no parity change, they would make the  $C^{14}$  transition second forbidden, bringing into closer agreement the theoretical expectations with the observations.

Ordinarily, no unique shape can be predicted theoretically for a forbidden transition.<sup>16</sup> However, it turns out that in the special case of a second forbidden  $0 \rightarrow 1$  transition with the Fermi (polar-vector) form of interaction, the spectrum can be predicted without ambiguity. The calculation was carried out by Pruett,<sup>17</sup> who found a spectrum shape which, when plotted on a Kurie diagram, shows a concavity toward the energy axis, in disagreement with the present experimental observation of the spectrum. The latter, as shown above, shows a very slight convexity toward the energy axis. In comparison with the measurements on  $S^{35}$ , it would appear likely that this slight curvature in  $C^{14}$  is real.

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<sup>13</sup> J. R. Pruett, Phys. Rev. **73**, 1219 (1948).

<sup>14</sup> O. Kofoed-Hansen, Phys. Rev. **71**, 451 (1947).

<sup>15</sup> F. A. Jenkins, Phys. Rev. **73**, 639 (1948).

<sup>16</sup> E. J. Konopinski, Phys. Rev. **60**, 308 (1941).

<sup>17</sup> J. R. Pruett, private communications.