

Yagoda, of the National Institute of Health, the author is indebted for the remainder of a powdered sample of pitchblende that he had analyzed for uranium. The National Museum, Washington, D. C., kindly supplied the sample of monazite through the good offices of Messrs. E. P. Henderson and Wm. F. Foshag, and thanks are due Dr. J. P. Marble for his advice in the choice of a suitable thorium mineral. Dr. E. G. Zies of the Geophysical Laboratory kindly contributed to the problem through his determination of the potash content of the Quincy granite.

Dr. Merle A. Tuve of the Department of Terrestrial Magnetism, Carnegie Institution of Washington, was responsible for arousing the author's interest in the problem during the course of an informal discussion concerning the possibility of obtaining for Dr. Hess a rock that would be essentially free of any radioactivity. The Harbison-Walker Refractories Company of Pittsburgh, Pennsylvania, kindly contributed several hundred pounds of such a rock in the form of dunite from their quarry at Addie, North Carolina.

PHYSICAL REVIEW

VOLUME 73, NUMBER 6

MARCH 15, 1948

## The Beta-Spectra of $\text{Cu}^{64}$ as a Test of the Fermi Theory

C. SHARP COOK AND LAWRENCE M. LANGER  
*Indiana University, Bloomington, Indiana*

(Received December 10, 1947)

The momentum spectra of the negatrons and positrons emitted by  $\text{Cu}^{64}$  have been measured with high resolution under such conditions that the usual distortions resulting from source thickness and backing, scattering, and finite counter window are completely negligible for almost the entire range of energies. Whereas the Fermi theory predicts the correct distribution for the high energy region, it is found that there are more negatrons and positrons than are predicted by the theory at low energies. The shapes of the spectra and the fact that no nuclear gamma-rays are observed preclude the possibility that they are complex spectra. Deviation from the Fermi theory occurs for energies below 0.270 Mev for the positrons and for energies below 0.190 Mev for the negatrons.

Incidental to these measurements, the end-point energies have been determined as 0.657  $\pm$  0.004 Mev for the positrons and 0.571  $\pm$  0.002 Mev for the negatrons. The ratio of the total number of electrons to positrons is 2.0.

### I. INTRODUCTION

A DETAILED investigation of the momentum distributions of the negatrons and positrons emitted by  $\text{Cu}^{64}$  has been made in order to test the validity of the Fermi theory of beta-decay.<sup>1</sup> When applied to reliable experimental data, the Fermi theory, using Fermi's original choice of interaction, has been remarkably successful in describing the high energy end of the beta-ray spectrum. The Kurie plot<sup>2</sup> of the experimental data yields a straight line for a large part of the distribution of all allowed and many forbidden transitions and is a valuable

tool for accurately determining the end-point energy.

Many experimenters,<sup>3</sup> however, have observed that there are apparently more particles in the low energy region of the distribution than are predicted by the Fermi theory. This deviation from the theory has usually been attributed to distortion resulting from instrumental difficulties such as finite source thickness and backing, absorption in the counter window, and scattering from the walls of the spectrometer. In some cases, the excess at low energies is caused by the fact that the spectrum is complex,

<sup>1</sup> E. Fermi, *Zeits. f. Physik* **88**, 161 (1934).

<sup>2</sup> Franz N. D. Kurie, J. R. Richardson, and H. C. Paxton, *Phys. Rev.* **49**, 368 (1936).

<sup>3</sup> A. W. Tyler, *Phys. Rev.* **56**, 125 (1939). J. L. Lawson, *Phys. Rev.* **56**, 131 (1939). K. Siegbahn and H. Slatis, *Ark. f. Mat., Ast., o Fysik* **32A**, No. 9 (1946). A. A. Townsend, *Proc. Roy. Soc.* **A177**, 357 (1941).

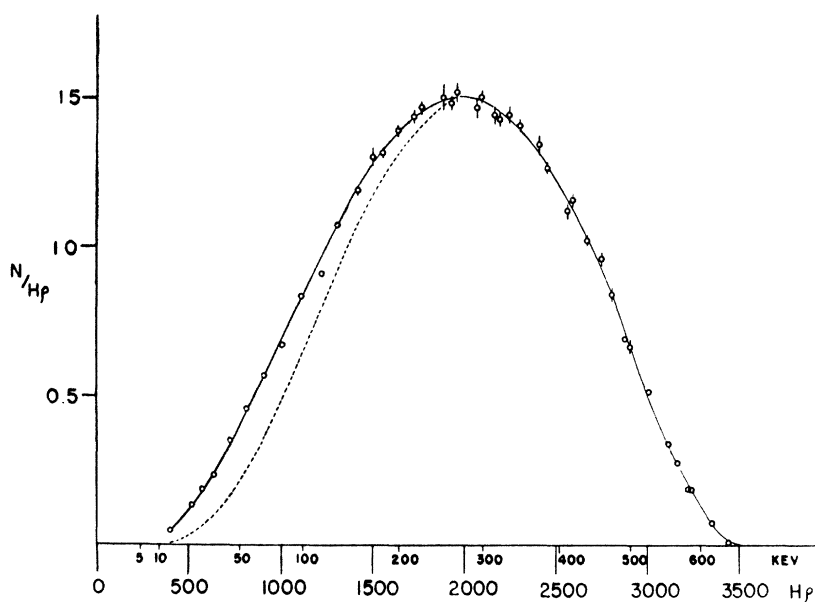


FIG. 1. Momentum spectrum of  $\text{Cu}^{64}$  positrons. Dashed curve is the distribution according to Fermi theory.

consisting of one or more transitions to excited levels of the product nucleus as well as to the ground state. Such complex spectra should, of course, be accompanied by gamma-radiation corresponding to the difference between the end-point energies.

Recently, Backus<sup>4</sup> measured the energy distribution of the positrons and negatrons from  $\text{Cu}^{64}$  at low energies, using an extremely thin source and backing and a counter window of negligible thickness. Because of the limitations of the electrostatic spectrometer which was used, he was not able to make measurements above 50 keV. Because of the proximity of the deflecting electrodes to the electron beam and because of the absence of baffles, one might expect a certain amount of scattering, which might or might not be the same for positrons and electrons.

Backus compared his results with the Fermi theory by plotting the log of the ratio of positrons to electrons as a function of the energy. He found a marked disagreement with theory. Lewis and Bohm<sup>5</sup> attempted to account for Backus' results by introducing a linear combination of the possible beta-interactions into the equation for the distribution. From an examina-

tion of Backus' data they concluded that the positron distribution was in agreement with the Fermi theory and that the negatron spectrum needed correcting. Since Backus' data cover such a small region of the total distribution, it seems dangerous to rely on deductions from them for the rest of the spectrum. Actually, as is shown below, neither the negatron nor the positron spectrum follows the Fermi theory. Whereas Backus' data for the ratio of positrons to electrons is apparently correct, the explanation offered by Lewis and Bohm is invalid.

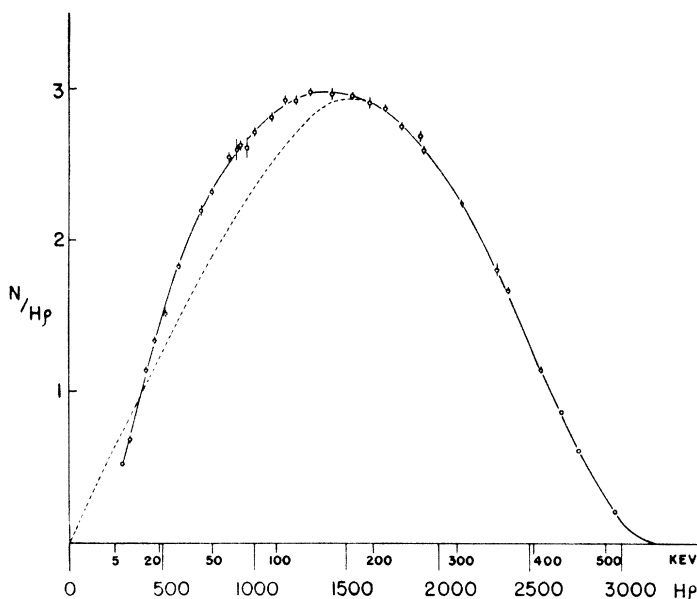
In the present experiment, an attempt was made to measure the beta-ray spectra of  $\text{Cu}^{64}$  over the entire energy range, under such conditions that any possible distortion of the true distribution would be negligible.

$\text{Cu}^{64}$  is a particularly suitable element for a test of the Fermi theory. Both beta transitions are allowed and should presumably give straight line Kurie plots. The end points and intensities of both the positrons and negatrons are comparable and so both may be measured under identical conditions.  $\text{Cu}^{64}$  activates strongly so that extremely thin sources of high specific activity are possible. The 12.8-hr. half-life is convenient so that complete positron and negatron spectra may be obtained from each source which is prepared. Except for an extremely weak radiation of about 1.3 Mev which follows

<sup>4</sup> J. Backus, Phys. Rev. 68, 59 (1945).

<sup>5</sup> H. Lewis and D. Bohm, Phys. Rev. 69, 129 (1946).

FIG. 2. Momentum spectrum of  $\text{Cu}^{64}$  negatrons. Dashed curve is the distribution according to the Fermi theory.



a very infrequent transition by  $K$  capture,<sup>6</sup> there seem to be no nuclear gamma-rays from  $\text{Cu}^{64}$  and the spectra are presumably not complex.

## II. EXPERIMENTAL METHOD

The measurements on the  $\text{Cu}^{64}$  spectra have been made with a large, high resolution magnetic spectrometer. This instrument is described in detail in another paper.<sup>7</sup> By using a magnetic field which varies radially in the proper manner, it is possible to obtain high resolution and good intensity in the large instrument. The nominal value of the radius of curvature is 40 cm. Using a source and a counter slit 0.4 cm wide and 2.54 cm high, the measured resolution for an internal conversion line is  $\Delta H\rho/H\rho = 0.5$  percent for the full width at half maximum. Under these conditions the transmission is about 0.1 percent of the total solid angle.

Because of the large size and the strategic baffling, background and scattering effects are completely negligible. The source is isolated at a considerable distance from any material which might introduce scattering. The main defining slit, located midway between the source and the counter slit, accepts a 2.54 cm-high beam with a

spread of 32 degrees in the plane perpendicular to the magnetic field. The edges of the defining slit are made of Polythene. The inside height of the aluminum vacuum chamber is 2.5 inches giving the beam a  $\frac{3}{4}$ -inch clearance above and below. Good evidence for the complete lack of scattering in the instrument is the extremely sharp high energy edge obtained for internal conversion and photo-lines and the fact that no counts are observed when the magnetic field is set for an energy lower than the calculated cut-off of the counter window or higher than the end point of a beta-spectrum.

Also because of the large size, it is possible to spread the active material over a larger area so as to minimize the self-absorption and scattering caused by the finite thickness of the source.

The particles are detected by means of an end window G-M counter. For a few preliminary runs, an unsupported mica window of 2.42 mg/cm<sup>2</sup> was used. Such a window has a range of about 35 keV and affects the shape of the electron distribution as high as 150 keV. For the final runs, a very thin Zapon window<sup>7</sup> capable of transmitting electrons having an energy as low as 2.0 keV was used. Such a window should not distort the spectrum above 10 keV (see also reference 4). The supporting grid for the Zapon foil is of sufficient thickness so that trans-

<sup>6</sup> H. Bradt, P. C. Gugelot, O. Huber, H. Medicus, P. Preiswerk, P. Sherrer, and R. Steffen, *Helv. Phys. Acta* **19**, 219 (1946); M. Deutsch, *Phys. Rev.* **72**, 729 (1947).

<sup>7</sup> L. M. Langer and C. S. Cook, *Rev. Sci. Inst.*, in press.

mission by the window is not dependent on energy. This grid is especially designed<sup>7</sup> to avoid partial transmission of rays coming through the window at an angle.

Relative magnetic field measurements were made with a flip coil and ballistic galvanometer. The absolute calibration of the magnetic field is in terms of the photoelectrons ejected by the 0.5108 Mev annihilation radiation. The energizing current for the magnet is stabilized electronically. If about one minute is allowed to elapse after each shift of the energizing current, then the magnetic field remains constant to better than 0.1 percent. At least two ballistic galvanometer readings were taken for each point counted. The calibration of the ballistic galvanometer was checked before and after each run in terms of a standard mutual inductance. In making measurements with a field of less than 20 gauss, care was taken to vary the current according to a prescribed cycle so as to avoid any change in the radial field distribution caused by remanence effects in the iron. This

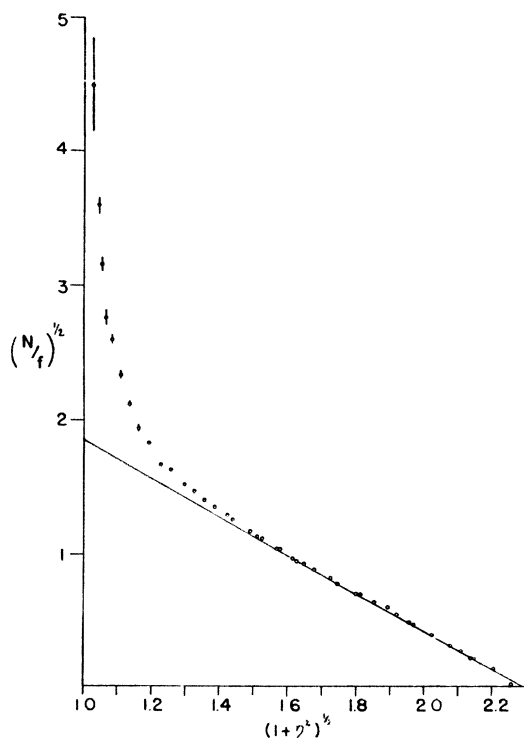


FIG. 3. Fermi plot of  $\text{Cu}^{64}$  positron spectrum. The extrapolated end point corresponds to  $0.657 \pm 0.004$  Mev. Deviation from the straight line begins at 0.270 Mev.

precaution is necessary in order to avoid distortion of the spectrum below about 30 kev.

Counts are recorded by a Higinbotham type scaler.<sup>8</sup> Counting rates are always such that losses are completely negligible.

The general procedure in making runs was to work through a complete spectrum and then put in arbitrary points at various parts of the distribution. It was found that with the preparation of the source described below, all parts of the  $\text{Cu}^{64}$  spectra decayed with the 12.8-hr half-life.

### III. PREPARATION OF SOURCES

$\text{Cu}^{64}$  can be prepared by deuteron bombardment of copper or zinc according to the reaction:  $\text{Cu}^{63}(d, p)$  and  $\text{Zn}^{66}(d, \alpha)$ . Although bombardment of zinc might seem preferable in order to obtain a separated source of high specific activity, it was found that the 11.5-Mev deuterons

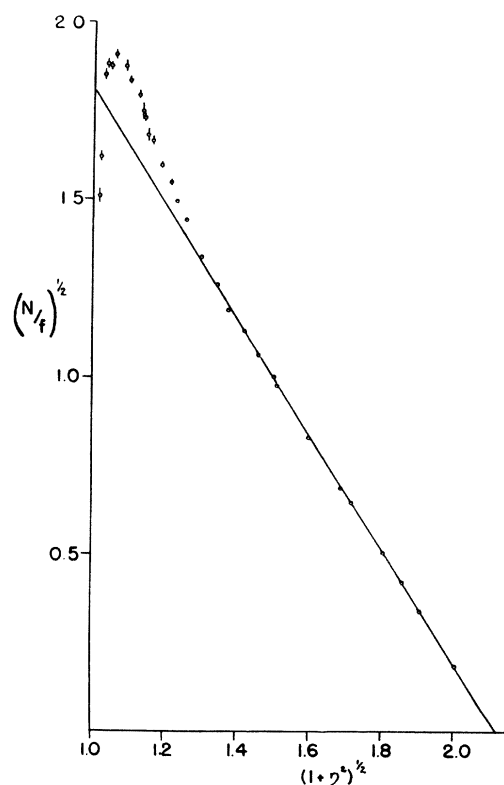


FIG. 4. Fermi plot of  $\text{Cu}^{64}$  negatron spectrum. The extrapolated end point corresponds to  $0.571 \pm 0.002$  Mev. Deviation from the straight line begins at 0.190 Mev.

<sup>8</sup> W. A. Higinbotham, James Gallagher, and Matthew Sands, *Rev. Sci. Inst.* **18**, 706 (1947).

available at the target chamber of the cyclotron produced a considerable quantity of  $\text{Cu}^{61}$  of 3.4-hr. half-life according to the reaction  $\text{Zn}^{64}$ . ( $d; \alpha, n$ ). The  $\text{Cu}^{61}$  cannot be chemically separated from the  $\text{Cu}^{64}$  and its period is sufficiently long so that it interferes with the measurements on  $\text{Cu}^{64}$ .

The method finally used, therefore, was to bombard copper on a probe. The probe was inserted to a radius which corresponds to 8-Mev deuterons so as to get maximum yield.<sup>9</sup> Bombardments were of from 4 to 8 hours duration with a circulating deuteron beam of several hundred microamperes.

A few flakes were scraped from the most active region of the bombarded target and dissolved in a small amount of nitric acid. To this was added 15 cc of distilled water and 25 cc of concentrated sulphuric acid. The solution was then heated to drive off the excess nitric acid. Copper was then separated from any zinc activities by electroplating onto a rotating tungsten cathode.<sup>10</sup> After plating for about two hours, the cathode was washed with distilled water and the copper deposit dissolved by one or two drops of concentrated nitric acid. The solution was picked up in a micropipette and spread over the surface of the source backing.

Source holders were prepared in the following way. A rectangular hole 2.5 cm by 0.4 cm was cut with a sharp needle in a sheet of mica 0.0005 inch thick. The mica was then cemented onto a Lucite ring 3.3 cm in diameter which mounts on the skeleton framework of the spectrometer source holder. The mica was then covered by two Zapon films each 0.01 mg/cm<sup>2</sup> thick. These films were made in the same manner as the films used for the counter window.<sup>7</sup> The Zapon sags very slightly into the rectangular opening of the mica. This serves well to confine the active solution to the desired region. The solution was then dried in a separate vacuum system after which it was covered by a single Zapon film 0.01 mg/cm<sup>2</sup> thick and placed in position in the spectrometer.

After each run, the sources were removed from the spectrometer and immediately weighed. The thinnest source used weighed somewhat less than 0.1 mg/cm<sup>2</sup>. The thickest was 0.35 mg/cm<sup>2</sup>.

<sup>9</sup> E. T. Clarke and J. W. Irvine, Phys. Rev. 69, 680 (1946).

<sup>10</sup> J. Steigman, Phys. Rev. 53, 771 (1938),

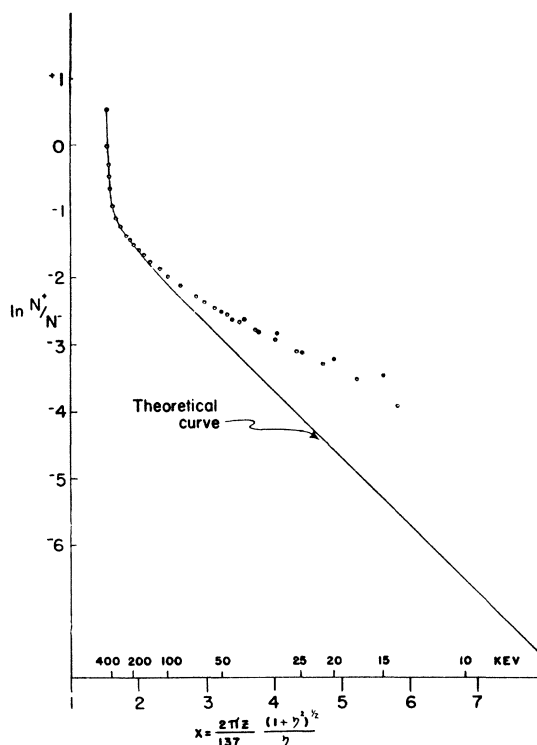


FIG. 5. The ratio of the number of positrons to the number of electrons as a function of the energy. The solid curve is the theoretical curve according to the Fermi theory. The open circles are our experimental data. The solid circles are the data of Backus adjusted to our data at 50 kev.

#### IV. RESULTS

In all, five sets of data were taken in which the intensity of the source was sufficient to yield good statistics. Three of these runs were made with the extremely thin Zapon counter window. The other two were made using the thicker mica window counter. In addition, several runs were made with much weaker sources yielding poorer statistics.

All the runs show that the momentum distribution is different from that predicted by the Fermi theory. In all cases the deviation from the theory occurs for positrons for energies below 0.270 Mev, while for negatrons the deviations begin for energies below 0.190 Mev.

The runs taken with the thin Zapon window counters have been adjusted to the same intensity and are plotted together.

Figures 1 and 2 show the momentum distributions for the positrons and negatrons. The relative counting rate, divided by the momentum

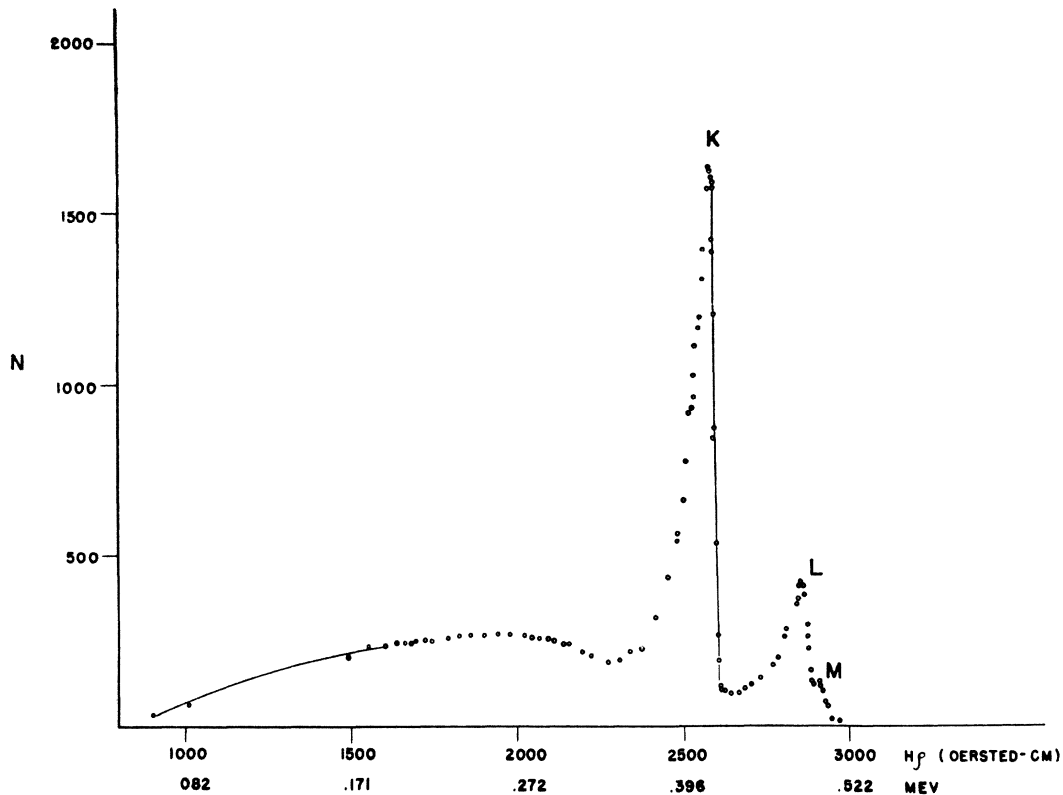


FIG. 6. Compton and photoelectrons ejected from a 0.0263 g/cm<sup>2</sup> Pb radiator by the 0.511-Mev annihilation radiation of Cu<sup>64</sup>. The ordinate is the relative number per minute.

is plotted against the momentum in terms of  $H\rho$ . An energy scale is indicated for convenience. The dashed curves are the distributions predicted by the Fermi theory. Figures 3 and 4 show the Kurie plots of the experimental data. The deviations from the theoretical straight lines occur at a total energy  $W = 1.53 \text{ mc}^2$  for the positrons and  $1.37 \text{ mc}^2$  for the negatrons.

The end-point energies determined from the straight line extrapolation of the Kurie plots are for the positrons  $0.657 \pm 0.004 \text{ Mev}$  and for the negatrons  $0.571 \pm 0.002 \text{ Mev}$ .

By integrating the areas under the experimental distributions, one gets for the ratio of the total number of negatrons to positrons a value of 2.0.

The difference between the experimental and theoretical distribution in Figs. 1 and 2 indicates that there are 9 percent more positrons and 6 percent more negatrons at low energy than is consistent with the theory.

If one uses the low  $Z$  approximation<sup>2</sup> for the Fermi coulomb factor,  $F$ , one may write for the ratio of the number of positrons to the number of electrons as a function of the energy,

$$N^+/N^- = Ke^{-x}[(W_+ - W)^2/(W_- - W)^2],$$

where  $K$  is a constant,  $W$  is the total energy in units of  $\text{mc}^2$ ,  $W_+$  and  $W_-$  are the end-point energies for the positrons and negatrons, respectively, and

$$x = 2\pi Z\alpha(W/\eta),$$

where  $Z$  is the atomic number,  $\alpha$  is the fine structure constant, and  $\eta = H\rho/1704$  is the electron momentum in units of  $\text{mc}$ .

In Fig. 5,  $\log N^+/N^-$  is plotted against  $x$ . The theoretical curve is shown as a solid line. The open circles are obtained from the experimental data. The solid circles show the data obtained by Backus adjusted to our data at 50 keV.

Our data is in good agreement with the results

of Backus over the region covered by him. Moreover, it is apparent from the curve that the deviation from the theory extends smoothly to the point where the positron spectrum first deviates from the Kurie straight lines, i.e., 270 kev.

If the excess of particles at low energies were due to the spectra being complex, one should expect to find gamma-radiation following the transitions to the excited states of Zn and Ni. The energy available for the gamma-ray turns out to be about the same for the positron and the negatron transitions. One should expect a gamma-ray of 0.387 Mev following 9 percent of the positron transitions and a gamma-ray of 0.381 Mev following 6 percent of the negatron transitions.<sup>11</sup>

A careful search for such radiations was made by examining the photoelectrons ejected from a thin Pb radiator 26.3 mg/cm<sup>2</sup> thick. Figure 6 shows the lines caused by the electrons ejected from the *K*, *L*, and *M* levels of lead by the 0.511 Mev annihilation radiation from the Cu<sup>64</sup> positrons. No other gamma-rays were observed of an energy between 0.190 and 0.51 Mev. By comparison with the intensity of the annihilation radiation of which there are two quanta for each positron emitted, one concludes that no nuclear gamma-ray of energy in the neighborhood of 0.38 Mev is emitted with an intensity of as much as 2 percent of the positron emission. If the transition to the ground levels were to proceed by two quanta of lower energy, then, because of the rapid increase in the photoelectric cross section, one should expect lines of even greater intensity. For example, one concludes from the data that no gamma-ray of 0.19 Mev is emitted with an intensity of as much as 0.2 percent of the positron emission.

It should also be noted that the general shapes of the Kurie plots do not readily allow resolution into complex spectra. The manner of deviation is quite different for positrons and negatrons.

<sup>11</sup> Low energy spectra having the abundance of the excess slow particles found here would have to be due to transitions which are allowed as are the high energy spectra. One should then expect only gamma-radiation corresponding to a transition direct to the ground level.

Whereas the positron distribution deviates more and more from the Fermi straight line at low energies, the negatron intensity first rises to a maximum and then decreases quite rapidly, actually being lower than that given by the Fermi theory for very low energies. Moreover, the decrease in the negatron curve is to be regarded as real since it begins at an energy of about 38 kev which is much higher than the region influenced by the counter window.

## V. CONCLUSIONS

The momentum distributions of the positrons and negatrons of Cu<sup>64</sup> have been measured with high resolution under such conditions that there is no distortion resulting from scattering, source thickness or backing, and counter window transmission. For both positrons and negatrons, more particles are observed at low energies than are predicted from the Fermi theory. The deviation from the theory begins at 0.270 Mev for positrons and 0.190 Mev for negatrons and is the same for several source thicknesses and two markedly different counter windows. The shapes of the spectra and the fact that no gamma-rays are observed eliminates the possibility that they are complex spectra. The fact that both positrons and electrons are measured under identical conditions and deviate in such different ways gives additional assurance that the effect is real and not instrumental.

One must conclude, therefore, that the Fermi theory does not predict the true distributions for either the negatrons or the positrons. Furthermore, a simple linear combination of the possible beta-interactions, as suggested by Lewis and Bohm, does not account for the experimental results.

It is planned to extend this work to other allowed transitions which can be measured under similar ideal conditions.

We are grateful to Professor E. J. Konopinski for many stimulating discussions.

This work is supported by a grant from the Frederick Gardner Cottrell Fund of the Research Corporation and by the U. S. Navy under Contract N6ori-48, T. O. I.