

Small Deviations Observed in Beta Spectra:  $\text{Na}^{22}\dagger$ 

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The intense positron spectrum of  $\text{Na}^{22}$  has been carefully measured in a magnetic spectrometer. This transition proceeds from a  $3+$  to a  $2+$  level. The observed  $\text{Na}^{22}$  spectrum has a nonstatistical shape corresponding to an excess of low-energy electrons. The well-measured spectrum of  $\text{Pm}^{147}$  was reinvestigated and found to have a statistical shape. The many tests of the experimental procedures all indicate that the observed nonstatistical spectrum of  $\text{Na}^{22}$  is not the result of instrumental distortions. Theoretical refinements for finite de Broglie wavelength, screening, and possible contributions from the twice forbidden matrix elements were considered and found to be much too small to explain the deviation from the statistical shape observed in  $\text{Na}^{22}$ . The Fermi-Kurie plot of  $\text{Na}^{22}$  can be linearized by a  $(1+b/W)$  correction factor. This same factor has been used to linearize the Fermi-Kurie plots of  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$  (in addition to the once forbidden, unique shape factor in the case of  $\text{Y}^{90}$ ). In all four cases, the value of the parameter  $b$  to yield a linear F-K plot is in the range  $0.2 < b < 0.4$ . At present, no theoretical explanation is offered for the correction  $(1+b/W)$ . It may be regarded as an empirical correction capable of explaining the observed shapes in the case of these four Gamow-Teller transitions. A search was also made for negative electrons accompanying the normal positron decay of  $\text{Na}^{22}$ . A weak, low-energy electron distribution was observed. This distribution may be explained by the theory of "shake-off" electrons.

## INTRODUCTION

WITH the improvement of experimental techniques, confidence has grown in the possibility of detecting in the shapes of beta spectra small deviations from the predictions of the present theory. Such deviations may correspond to higher order effects previously neglected in the theoretical calculations or to refinements which should be made in the theory. Deviations from the present theory have been reported<sup>1-4</sup> in the spectra of  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$ . All three of these isotopes are electron emitters which decay by pure Gamow-Teller radiations.

The Fermi-Kurie (F-K) plots of the above three spectra are reported to exhibit deviations from linearity corresponding to an excess of low-energy electrons (for  $\text{Y}^{90}$  after correction with the once-forbidden, unique shape factor). Before the advent of parity nonconservation, deviations could be interpreted in terms of Fierz interference between the possible beta decay interactions. Other evidence indicated that Fierz interference is much less<sup>5</sup> or even nonexistent. If the deviations observed in the three electron spectra were the result of Fierz interference, the shape factor  $(1+b/W)$  observed for these electron emitters would become  $(1-b/W)$  for a positron emitter (the Fierz parameter has a different sign for positrons and electrons). A positron spectrum

would be expected to exhibit deviations corresponding to a deficiency of low-energy electrons. Thus, more definitive evidence for the possible existence of Fierz interference in Gamow-Teller radiations can be obtained from the study of both positron and electron spectra. To make a more meaningful interpretation of the deviations observed in  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$ , it is of value to know whether deviations are present in positron spectra and what is the direction of the deviations.

A careful investigation has been made of the intense, allowed positron spectrum of  $\text{Na}^{22}$ . This decay also proceeds by pure Gamow-Teller radiations. The allowed  $\text{Na}^{22}$  spectrum has been reported to have a linear F-K plot from its endpoint down to about 100 keV.<sup>6-8</sup> Because of its abnormally high  $\log ft$  value of 7.4, the allowed positron spectrum of  $\text{Na}^{22}$  is perhaps not an ideal case for comparison with the electron spectra (although the  $ft$  value of  $\text{P}^{32}$  is also relatively high). From an experimental point of view, however, it is very suitable. Because of the low  $Z$ , it has the added virtue of small Coulomb corrections. The spectrum was measured under many different experimental conditions. With the improved techniques employed, a nonstatistical spectrum has been observed for the allowed  $\text{Na}^{22}$  positron spectrum.<sup>9</sup>

Many new tests of the experimental techniques were made during the course of these studies. The well-measured beta spectrum of  $\text{Pm}^{147}$  was carefully reinvestigated under various experimental conditions. The beta spectrum of  $\text{Pr}^{143}$  was also carefully measured

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<sup>1</sup> Johnson, Johnson, and Langer, preceding paper [Phys. Rev. **112**, 2004 (1958)]; O. E. Johnson, Ph.D. thesis, Indiana University, January, 1956 (unpublished).<sup>2</sup> Porter, Wagner, and Freedman, Phys. Rev. **107**, 135 (1957).<sup>3</sup> Yuasa, Laberriquer-Frolow, and Feuvrais, J. phys. radium **18**, 498 (1957).<sup>4</sup> Graham, Geiger, and Eastwood, Can. J. Phys. (to be published).<sup>5</sup> R. Sherr and R. H. Miller, Phys. Rev. **93**, 1076 (1954).<sup>6</sup> Macklin, Lidofsky, and Wu, Phys. Rev. **78**, 318(A) (1950).<sup>7</sup> B. T. Wright, Phys. Rev. **90**, 159 (1953).<sup>8</sup> Alburger, Hughes, and Egger, Phys. Rev. **78**, 318(A) (1950).<sup>9</sup> J. H. Hamilton, Ph.D. thesis, Indiana University, June, 1958 (unpublished). See also, L. M. Langer, *Proceedings of Rehovoth Conference on Nuclear Structure*, (North-Holland Publishing Company, Amsterdam, 1958), p. 437.

with the magnetic spectrometer and with a  $4\pi$  scintillation spectrometer.<sup>10</sup>

A thorough search was made for possible negative electrons associated with the normal positron decay of Na<sup>22</sup>. These studies yielded important information regarding the possibility of the deviations observed in the above three electron spectra arising from some secondary process associated with the normal decay or from some scattering process. Evidence was found for a weak, low-energy electron distribution accompanying the positron decay.<sup>9,11</sup>

## EXPERIMENTAL PROCEDURES

### Magnetic Spectrometer

The spectrometer used in these investigations was described in the preceding paper.<sup>1</sup> All of the precautions described in the preceding paper concerning the operation of the equipment were followed. Certain improvements were made and these have been described elsewhere.<sup>10</sup> The principal modification was the use of a specially designed end-window, loop-anode, stainless steel cathode, proportional counter as the detector.<sup>10</sup> This counter was extremely stable and trouble-free in operation. Although very high counting rates may be detected with essentially no loss, the counting rates employed in these studies were all less than 5000 per minute. For energies greater than 30 keV, the counter window was an unsupported, aluminum-coated Zapon film with a measured thickness of 100–150  $\mu\text{g}/\text{cm}^2$ . The cutoff energy of such windows was about 5–6 keV.<sup>10</sup> For the low-energy measurements, the counter window was a Zapon film ( $<10 \mu\text{g}/\text{cm}^2$ ) supported on a 56% transmission Lektromesh grid. The measured cutoff energy of some of these windows was less than 1 keV. In some cases, a gas pressure of 7–8 cm of Hg was used in the counter for the low-energy measurements because of the thin windows.

At the beginning of the studies of the Na<sup>22</sup> positron spectrum, several different types of counters were used in measuring the spectrum. The spectrum was measured with each of the following types of counter: end-window GM-counter with a loop anode, end-window GM-counter with a straight wire, glass beaded anode, side-window proportional counter, and an end-window proportional counter with a loop anode (this is the counter described in detail).<sup>10</sup> The measurements made with each of these counters yielded essentially the same spectral shape. These results indicate that the spectrum was not being distorted by the counters. The loop-anode proportional counter, however, had the best operating characteristics and was used in all of the later studies.

The spectrometer was calibrated in terms of the

$K$  conversion line of the 661.6-keV gamma ray<sup>12</sup> of Cs<sup>137</sup> and the  $K$  conversion line of the 1064-keV gamma ray<sup>13</sup> of Bi<sup>207</sup>. The low-energy calibration was in terms of the internal conversion lines of the 238.6-keV gamma ray<sup>14</sup> of ThB (the  $F$  and  $I$  lines).

Measurements of the Na<sup>22</sup> positron spectrum were made with the normal baffle system and with a modified beam-defining baffle which decreased the area of the accepted beam by more than 50%. This checked on scattering of beta particles and gamma rays in the vacuum chamber and on the transmission properties of different regions of the magnetic field. Measurements were made with a counter slit opening of 8 mm as well as the usual 5 mm. The thickness of the material used to define the slit was decreased by a factor of two to check on scattering in the slit edges. The pressure in the chamber was varied over a factor of ten in some experiments to test for possible scattering in the residual gas in the chamber. The normal vacuum was between  $5 \times 10^{-6}$  and  $10^{-5}$  mm of Hg. In all of these experiments, no change was observed in the shape of the Na<sup>22</sup> positron spectrum.

### Source Preparation

In these experiments, liquid-deposited and thermally evaporated sources were used. The process of preparing these sources is described in the preceding paper.<sup>1</sup> All of the sources used were 5 mm wide.

Seven Pm<sup>147</sup> sources were prepared; two were liquid deposited (thickness  $<40 \mu\text{g}/\text{cm}^2$ ) and five were thermally evaporated in vacuum (thickness  $\ll 10 \mu\text{g}/\text{cm}^2$ ). Selective evaporation was used to further increase the specific activity which made possible the very thin sources. The liquid deposited sources were on  $\approx 5 \mu\text{g}/\text{cm}^2$  of Zapon and 180  $\mu\text{g}/\text{cm}^2$  of Al foil. The five evaporated sources were prepared on backings of 180  $\mu\text{g}/\text{cm}^2$  of Al, 100–150  $\mu\text{g}/\text{cm}^2$  of Zapon-LC600,  $\approx 20 \mu\text{g}/\text{cm}^2$  of Zapon,  $\approx 10 \mu\text{g}/\text{cm}^2$  of Zapon, and  $\approx 6 \mu\text{g}/\text{cm}^2$  of Zapon. All of the sources were covered with  $\approx 2\text{--}3 \mu\text{g}/\text{cm}^2$  Zapon films with two exceptions for which no covers were used. Later a cover was added to one of these. These sources were prepared from two different shipments of Pm<sup>147</sup> from Oak Ridge National Laboratory. Careful searches for gamma rays confirmed that there were no impurities, at least with gamma rays present.

Carrier-free Na<sup>22</sup> was obtained from Nuclear Science and Engineering Corporation, Pittsburgh. The first shipment contained more than the specified solids so a second sample was ordered. Two sources were prepared from the first shipment. Source 1 was liquid-deposited and source 2 vacuum-evaporated. The backings for both sources were 180- $\mu\text{g}/\text{cm}^2$  Al foil covered with LC600  $<25 \mu\text{g}/\text{cm}^2$ . The source thickness was  $<100 \mu\text{g}/\text{cm}^2$  and each source was covered with a

<sup>10</sup> Hamilton, Langer, Robinson, and Smith, Phys. Rev. **112**, 945 (1958).

<sup>11</sup> Hamilton, Langer, and Smith, Bull. Am. Phys. Soc. Ser. II, **3**, 61 (1958).

<sup>12</sup> Lindström, Siegbahn, and Wapstra, Proc. Phys. Soc. (London) **B66**, 54 (1953).

<sup>13</sup> D. E. Alburger, Phys. Rev. **92**, 1257 (1953).

<sup>14</sup> K. Siegbahn and K. Edvarson, Nuclear Phys. **1**, 137 (1956).

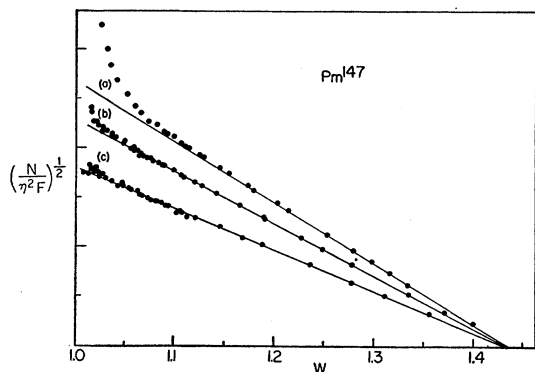


FIG. 1. Fermi-Kurie (F-K) plots for  $\text{Pm}^{147}$ . The three evaporated sources were identical except for backings which were (a) 180- $\mu\text{g}/\text{cm}^2$  Al foil, (b)  $\approx 10\text{-}\mu\text{g}/\text{cm}^2$  Zapon, and (c)  $\approx 6\text{-}\mu\text{g}/\text{cm}^2$  Zapon.

Zapon film ( $< 10\ \mu\text{g}/\text{cm}^2$ ). Two sources were also prepared from the second shipment; both were thermally evaporated in vacuum. Source 3 was on a backing of 180- $\mu\text{g}/\text{cm}^2$  Al foil covered with LC600 and source 4 on an aluminum-coated Zapon-LC600 laminate. The thickness of the laminate was measured with an alpha absorption technique to be  $20 \pm 10\ \mu\text{g}/\text{cm}^2$ . Both sources were covered with Zapon films ( $< 10\ \mu\text{g}/\text{cm}^2$ ). A radioautograph of source 3 indicated that the activity was uniformly distributed. After many measurements had been made of the positron spectrum and electron distribution, source 3 was covered with a 10- $\mu\text{g}/\text{cm}^2$  Zapon film. Then, after more measurements, it was covered with an additional  $(60 \pm 10)\text{-}\mu\text{g}/\text{cm}^2$  film for further studies. These measurements yielded information regarding possible distortions produced by some forward scattering process.

#### PROMETHEUM-147

The spectrum of  $\text{Pm}^{147}$  was measured under various experimental conditions to test for possible instrumental distortions.  $\text{Pm}^{147}$  was selected because it has been well studied<sup>15-17</sup> and has been found to have a linear F-K plot down to 8 keV.<sup>15</sup> Essentially 100% of the transitions go by electron emission to the ground state of  $\text{Sm}^{147}$  with a half-life of 2.64 years.<sup>18</sup> Transitions to a possible excited state at 121 keV ( $3 \times 10^{-5}$  photon per beta decay<sup>19,20</sup>) would produce no observable distortion of the ground state spectrum. The ground state decay is a once-forbidden, nonunique transition ( $\frac{5}{2}^+ \rightarrow \frac{7}{2}^-$ ) with a  $\log ft$  value of 7.6. The statistical shape may be expected for the F-K plot from the theoretical point of view.<sup>21</sup> This is because the energy-independent Coulomb

terms which are proportional to  $\alpha Z/R$  would be expected to dominate in the shape factor, especially since  $W_0$  is so small.

Measurements were made from 8 keV to beyond the end point of the  $\text{Pm}^{147}$  spectrum. This complete energy range was measured with the aid of two counters; one with an unsupported window and the other with a Lektromesh supported window. The data taken with the supported window were normalized to the data taken with the unsupported window. The energy region of overlapping data for normalization was 30 to 65 keV. This energy interval is long enough to make a reliable overlap possible. The normalization constants at 1.06, 1.08, 1.10, and 1.12  $m_0c^2$  were constant to within 0.2%. The measurements were made over a period of 8 months. The decay of the sources agreed with the reported half-life of 2.64 years. No decay correction was necessary for any single spectral measurement. Essentially every point had a mean statistical error in the counting rates of 1%. Below 20 keV, this value increased towards 2%.

Fermi-Kurie plots were constructed with the aid of tables<sup>22</sup> and graphs.<sup>23</sup> Figure 1 gives F-K plots of the data obtained from three different sources. The three sources (each vacuum-evaporated,  $< 10\ \mu\text{g}/\text{cm}^2$  thick) are identical except for backing. Their backings were Al foil, (b)  $\approx 10\ \mu\text{g}/\text{cm}^2$  of Zapon, and (c)  $\approx 6\ \mu\text{g}/\text{cm}^2$  of Zapon. A comparison of the three plots makes it quite obvious that there are large distortions possible from backing thickness effects. As the backing thickness decreased, the linearity of the F-K plot extended to a lower energy. F-K plot (c) in Fig. 1 is linear down to about 15 keV where a slight rise in the plot begins. The linear F-K plot corresponds to a linear shape correction factor,  $C = N/\eta^2 F(W_0 - W)^2$  (Fig. 2). The F-K plot reported linear to 8 keV<sup>15</sup> was obtained with a source on a 1.5- $\mu\text{g}/\text{cm}^2$  backing. Thus the present data are not inconsistent with past measurements. The deviations

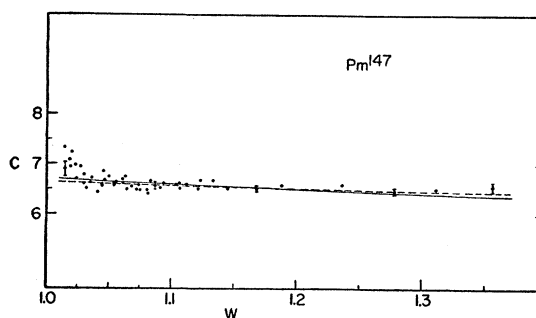


FIG. 2. Shape factor,  $C = N/\eta^2 F(W_0 - W)^2$ , plot for  $\text{Pm}^{147}$ . The dashed curve is the factor  $(1 + 0.1/W)$  and the solid curve  $(1 + 0.3/W)$ .

<sup>15</sup> Langer, Motz, and Price, *Phys. Rev.* **77**, 798 (1950).

<sup>16</sup> Lidofsky, Macklin, and Wu, *Phys. Rev.* **76**, 1888 (1949).

<sup>17</sup> H. M. Agnew, *Phys. Rev.* **77**, 655 (1950).

<sup>18</sup> Merritt, Campion, and Hawkings, *Can. J. Phys.* **35**, 16 (1957).

<sup>19</sup> H. Langevin-Joliot and M. Lederer, *J. phys. radium* **17**, 497 (1956).

<sup>20</sup> N. Starfelt and J. Cederlund, *Phys. Rev.* **105**, 241 (1957).

<sup>21</sup> H. Mahmoud and E. Konspinski, *Phys. Rev.* **88**, 1266 (1952).

<sup>22</sup> *Tables for the Analysis of Beta Spectra*, National Bureau of Standards, Applied Mathematics Series No. 13 (U. S. Government Printing Office, Washington, D. C., 1952).

<sup>23</sup> S. A. Moszkowski, University of Chicago (privately circulated graphs).

begin in plot (b) at about 25 keV and in (a) between 75 and 100 keV. From this work and the Na<sup>22</sup> studies, it is concluded that backings <180  $\mu\text{g}/\text{cm}^2$  are required to make undistorted measurements below 100 keV and preferably backings of 10–20  $\mu\text{g}/\text{cm}^2$  should be used for studies down to 30–50 keV.

A least-squares fit of the data for  $1.14 m_0c^2 < W < 1.34 m_0c^2$  yielded an end-point energy of  $(1.439 \pm 0.002) m_0c^2$ . Considering uncertainties in the absolute calibration, this corresponds to an end point of  $224.3 \pm 1.3$  keV.

Various experimental parameters were varied to determine their effect on the spectrum. No change was observed in the spectrum for pressures in the vacuum chamber as high as  $3 \times 10^{-4}$  mm of Hg. The effects of source charging were studied. It was confirmed that either method of source grounding described in the previous paper is sufficient. Without proper grounding, the spectrum was greatly distorted with a deficiency of low-energy electrons and a shifted end point (215 keV).

Corrections for outer screening<sup>22,24</sup> were found to be negligible over the momentum range measured.

An analysis was made to set limits on Fierz-type interference in the Pm<sup>147</sup> spectrum. This was previously done by Mahmud and Konopinski.<sup>21</sup> It should be noted that Pm<sup>147</sup> is not to be considered in the same manner as In<sup>114</sup>, Y<sup>90</sup>, and P<sup>32</sup> since the Pm<sup>147</sup> decay is once-forbidden, nonunique. It has been suggested<sup>25</sup> that a shape factor of the form  $(1+aW+b/W)$ , where  $a$  and  $b$  are adjustable parameters, should be used to fit the experimental spectra of once-forbidden, nonunique transitions. This equation was fitted to the Pr<sup>143</sup> spectrum<sup>10</sup> which is a  $\frac{5}{2}^+$  to  $\frac{7}{2}^-$  transition like Pm<sup>147</sup>. Thus a shape factor of the form  $(1+b/W)$  with  $a \approx 0$  might easily be expected from a theoretical standpoint.

Figure 2 gives a shape factor plot of the data [from plot (c) of Fig. 1] where  $C = N/\eta^2 F(W_0 - W)^2$ . The curves represent the factor  $(1+b/W)$  for  $b$  values of 0.1 and 0.3 and the flags represent one mean statistical deviation in the counting rate. A straight line is the best fit to the data down to about 15 keV, however, the correction for  $b=0.1$  would be difficult to exclude.

The data are plotted for  $W_0 = 1.439 m_0c^2$ . To compare factor  $(1+b/W)$  for  $b=0.3$  to the data,  $C$  should be calculated for  $W_0 = 1.442 m_0c^2$  (as obtained below). For this higher value of  $W_0$ , the high-energy points are brought down relative to the low-energy ones for a better comparison. This  $b$  value, however, is the approximate upper limit of permissible values to fit the data.

In Fig. 3 are F-K plots of the same data as in Fig. 2 corrected with  $(1+b/W)$  for  $b=0$  and 0.3. The straight lines are least squares fits of the data in the energy range  $(1.14-1.34) m_0c^2$ . Note that a slightly higher endpoint of  $1.442 m_0c^2$  is obtained with the data corrected for

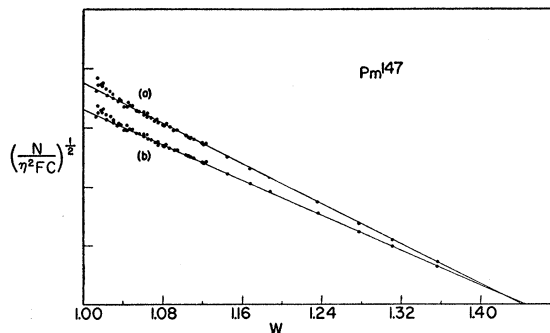


FIG. 3. F-K plots for Pm<sup>147</sup> corrected with  $C = 1 + b/W$ : (a)  $b=0$  and (b)  $b=0.3$ .

$b=0.3$ . This  $b$  value of 0.3 is approximately the upper limit of the parameter  $b$  which will yield a linear F-K plot. The short energy range makes a more definitive limit difficult. It should be restated that even if a shape factor  $(1+b/W)$  is needed to fit the spectrum it could be explained by the once-forbiddenness of the transition and would not necessarily be of the same nature as the effect found in In<sup>114</sup>, Y<sup>90</sup>, and P<sup>32</sup>.

## SODIUM-22

### Positron Spectrum

Na<sup>22</sup> decays by positron emission and K-capture (half-life 2.58 years<sup>18</sup>) to an excited state at 1.28 MeV which in turn decays to the ground state of Ne<sup>22</sup> through emission of a 1.28-MeV gamma ray.<sup>5,7</sup> This mode of decay represents essentially 100% of the transitions. The transition to the ground state is approximately 0.06%<sup>7</sup> and is completely negligible in these studies. The measured spin and magnetic moment of Na<sup>22</sup> lead to a  $3^+$  assignment for its ground state.<sup>26</sup> The ground state of the even-even nucleus Ne<sup>22</sup> is  $0^+$ . With this  $0^+$  assignment, the 1.28-MeV level in Ne<sup>22</sup> is  $2^+$  as determined from the measured internal conversion coefficient of the gamma transition.<sup>27</sup> With these level assignments for Na<sup>22</sup> and Ne<sup>22</sup>, the Na<sup>22</sup> decay to the 1.28-MeV level should be allowed.

The reported statistical spectrum of Na<sup>22</sup> is consistent with that of an allowed transition but the  $\log ft$  of 7.4 is high for such transition. “ $l$ -forbiddenness” or the accidentally poor overlap of the initial and final nucleon wave functions has been assumed to account for the high  $\log ft$  value. Although this transition is slower than most allowed transitions, it is still fast in comparison to twice forbidden ones. There should be negligible contribution to the Na<sup>22</sup> decay from the twice forbidden matrix elements. This will be discussed in greater detail below.

The initial measurements of the positron spectrum were made with sources 1 and 2 described above. A similar nonstatistical spectral distribution was observed

<sup>24</sup> J. R. Reitz, Phys. Rev. **77**, 10 (1950).

<sup>25</sup> T. Kotani and M. Ross [submitted to Progr. Theoret. Phys. (Kyoto)].

<sup>26</sup> L. Davis, Phys. Rev. **74**, 1193 (1948).

<sup>27</sup> R. D. Leamer and G. W. Hinman, Phys. Rev. **96**, 1607 (1954).

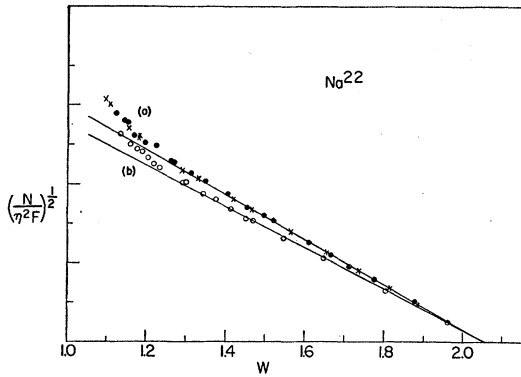


FIG. 4. F-K plots for  $\text{Na}^{22}$ : (a) The crosses represent data taken four months after that plotted as solid circles for source 3. (b) The open circles represent data from source 2.

with each source. (Recall that source 1 was liquid-deposited and source 2 vacuum-evaporated.) Measurements made with source 3, which was a factor of 10 or more thinner than sources 1 and 2 but otherwise identical, agreed with the data from sources 1 and 2 (see Fig. 4). The straight lines in Fig. 4 are fitted to the points with  $W > 1.3 m_0c^2$ . Both plots exhibit a very similar deviation from linearity beginning between  $1.25 m_0c^2$  and  $1.35 m_0c^2$ . This agreement indicates there is little if any distortion from source thickness in the measured energy range ( $E > 70$  kev). The normalized repeat spectrum obtained four months later with source 3 indicates that the deviation is decaying with the  $\text{Na}^{22}$  half-life as is the remainder of the spectrum.

Measurements were made from about 50 kev beyond the end point of the allowed  $\text{Na}^{22}$  spectrum with source 4. Seven measurements were made over a period of 4 months. Figure 5 gives a F-K plot of data taken with source 4 which had the thin backing ( $20 \mu\text{g}/\text{cm}^2$ ). The straight line is least-squares fitted to the data with  $W > 1.5 m_0c^2$  and yields an end point energy of  $(2.0626 \pm 0.006) m_0c^2$  ( $543 \pm 3$  kev). The deviation from linearity for source 4 begins in the same energy region as for sources 2 and 3 but is considerably less below

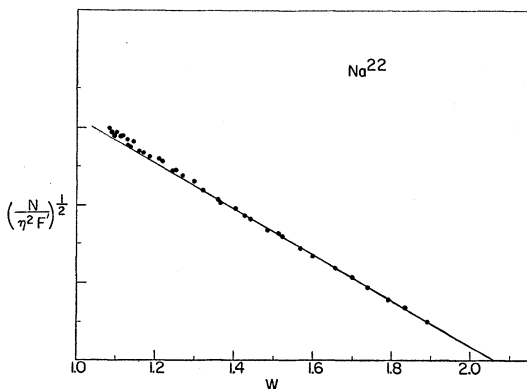


FIG. 5. F-K plot for  $\text{Na}^{22}$  source 4.  $F'$  includes the screening correction.

100 kev for the other sources. The only difference between sources 3 and 4 is the backing thickness which must be responsible for the added deviation in the data for source 3. Studies were made of the effects of source backing with  $\text{Pm}^{147}$  sources. All of the evidence indicates that there should be no distortion in the spectrum measured with source 4 for  $W \gtrsim 1.1 m_0c^2$ . These studies also confirmed that backings  $\geq 180 \mu\text{g}/\text{cm}^2$  thick would produce spectral distortions which begin around 100 kev. An added  $15 \mu\text{g}/\text{cm}^2$  of Al was vaporized on the back of source 4 to insure that source charging was not distorting the spectrum. The measured distribution before and after the extra Al was added agreed down to about 64 kev. Below this energy, the distribution after adding the Al exhibited a slight excess of particles as would be expected for the thicker backing.

Two tests were made for possible effects arising from scattering or annihilation of the positrons in the material used to define the detector acceptance slit which is located directly in front of the counter. The thickness of the material was decreased a factor of two and measurements were made for slit openings of 5 and 8 mm. There was no observable change in the shape of the spectrum. A further test for possible scattering in the spectrometer chamber was made by decreasing by more than 50% the area of the accepted beam. The extra baffle decreased the opening both radially and axially. The spectrum measured with the extra baffle agreed well with that taken without the baffle, confirming the absence of scattering in the spectrometer.

To determine whether the observed deviation from linearity was in any way the result of some forward scattering process in the thin cover film or even thinner source, the  $\text{Na}^{22}$  spectrum was measured with two additional Zapon cover films (total thickness  $70 \pm 15 \mu\text{g}/\text{cm}^2$ ) over the source. The F-K plots (Fig. 6) of the data taken with and without the two cover films agree. The good agreement of the data indicates that there is negligible distortion produced by the cover films. The possibility of compensating effects yielding

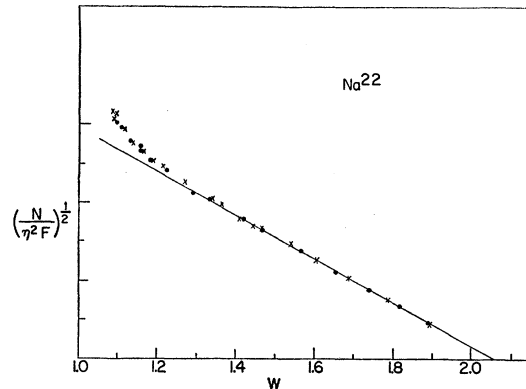


FIG. 6. F-K plots for  $\text{Na}^{22}$  source 3. The data plotted as crosses were taken after a  $70\text{-}\mu\text{g}/\text{cm}^2$  Zapon cover film was added over the source.

the same spectrum would be most unlikely for a positron spectrum measured in a 180-degree focusing spectrometer.

Two theoretical refinements of the data were considered. The correction for the finite de Broglie wavelength,<sup>28,29</sup> although in the right direction, is much too small to account for the observed deviation. The data were corrected for outer screening<sup>22,24</sup> with the aid of tables<sup>22</sup> for  $Z=10$  and  $V_0=0.7$ . This correction was somewhat larger but still resulted in less than 1% relative change in the spectrum over the measured energy range. This correction is also in the right direction to linearize the F-K plot but is again much too small.

The high  $ft$  value for the allowed  $\text{Na}^{22}$  decay may imply that there is some destructive interference among the radiations contributing to the decay. If this were so, it might be possible for small energy-dependent terms from the twice forbidden matrix elements to exert their presence in the spectrum. From the known  $ft$  values of twice-forbidden transitions, the possibility that there would be any contribution of the required magnitude to explain the  $\text{Na}^{22}$  spectrum appears extremely unlikely. There would be a cross term between the allowed and second-forbidden terms which would introduce a shape factor correction. Although the contributions of this cross term is approximately 100 times larger than that of the second-forbidden matrix elements, approximate calculations for  $\text{Na}^{22}$  indicate that even this is completely negligible.

The  $\text{Na}^{22}$  spectrum can be linearized by a correction of the form  $(1+b/W)$ . In Fig. 7 are plots of the data from source 4 corrected with  $(1+b/W)$  for various values of  $b$ . The lines are least-squares fits to the points with  $W > 1.5 m_0c^2$ . The best value of  $b$  to yield a linear F-K plot is in the range  $0.25 \leq b \leq 0.35$ . The data from source 3, corrected with  $(1+0.3/W)$ , are also plotted in Fig. 7. Note that this correction does not linearize the F-K plot below about 100 kev. This again emphasizes the difference in the data from sources 3 and 4. This distortion in the source 3 data is attributed to effects arising from the thicker backing. Figure 8 gives a plot of the shape correction factor  $C = N/\eta^2 F'(W_0 - W)^2$ . The curves are the factor  $(1+b/W)$  for  $b=0.25$  and  $0.30$ . The data are plotted as closed circles for  $W_0 = 2.0626 m_0c^2$  as obtained from a least-squares straight line of the data without the factor  $(1+b/W)$  and as crosses (normalized to fit the curves) for  $W_0 = 2.0680 m_0c^2$  as obtained from a least-squares fit of the data corrected with  $(1+0.3/W)$ . A correction factor of this form is a reasonable fit to the data. The flags represent the standard deviation in the counting rates which was  $\approx 1.7\%$  for all points.

A search was made for possible negative electrons associated with the normal decay of  $\text{Na}^{22}$ . It is possible

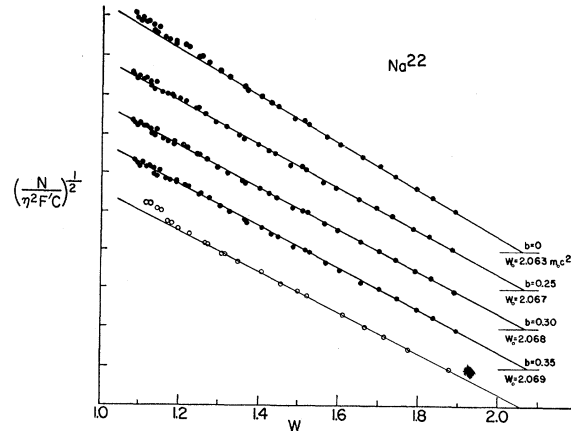


Fig. 7. F-K plots for  $\text{Na}^{22}$  source 4 corrected for screening and for  $(1+b/W)$  for various values of  $b$ . The open circles represent data from source 3 corrected with  $(1+0.3/W)$ .

that some secondary process of electron emission might be responsible for the deviations observed in the spectra of  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$ . Positron decay offers a good opportunity to search for secondary processes of electron emission. This search provided useful information relating to the principal problem of this paper. These studies indicated that the films normally used to cover the sources should not be distorting any of the spectral measurements. A further important conclusion is that there do not appear to be any secondary modes of decay or scattering processes with magnitudes large enough to explain the observed deviations in the electron spectra of  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$  or in the positron spectrum of  $\text{Na}^{22}$ . An electron distribution was observed accompanying the positron decay. The results and conclusions of this investigation are found in the Appendix.

## CONCLUSIONS

The allowed positron spectrum of  $\text{Na}^{22}$ , which decays by pure Gamow-Teller radiations, has been measured and found to exhibit a nonstatistical shape. The spectrum of  $\text{Pm}^{147}$  was measured as a further test of the

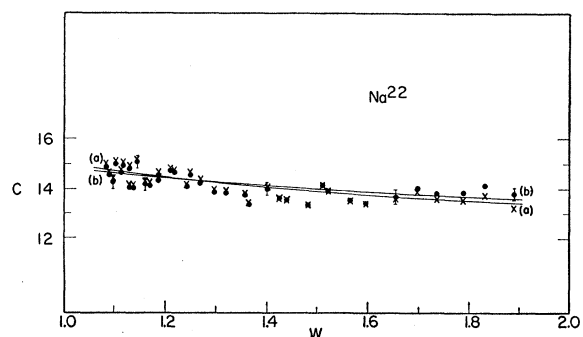


Fig. 8. Shape factor,  $C = N/\eta^2 F'(W_0 - W)^2$ , plot for  $\text{Na}^{22}$  source 4. The closed circles are for  $W_0 = 2.0626 m_0c^2$  and the crosses for  $W_0 = 2.0680 m_0c^2$ . The curves represent the factor  $(1+b/W)$  for (a)  $b=0.30$  and (b)  $b=0.25$ .

<sup>28</sup> M. E. Rose and C. L. Perry, Phys. Rev. **90**, 479 (1953).

<sup>29</sup> Rose, Perry, and Dismuke, Oak Ridge National Laboratory Report ORNL-1459, Special, 1953 (unpublished).

experimental techniques. The spectrum of  $\text{Pr}^{143}$  was measured with magnetic and scintillation spectrometers<sup>10</sup> during the course of this work. The good agreement of the  $\text{Pr}^{143}$  data taken with these two different spectrometers is strong evidence that the measurements are not being distorted by instrumental effects. All of the evidence indicates that the observed nonstatistical spectra of  $\text{Na}^{22}$ ,  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$  represent the undistorted beta distributions for each isotope.

All four of these isotopes decay by pure Gamow-Teller radiations. It was found that a shape factor of the form  $(1+b/W)$  would linearize the F-K plots of each of the four isotopes. The value of the parameter  $b$  to yield a linear F-K plot is in the range  $0.2 < b < 0.4$  in each case.

It was mentioned earlier that the shape factor for Fierz interference  $(1+b/W)$  has a different sign for  $b$  for positrons and electrons. The deviations observed in these four spectra are not consistent with an interpretation of Fierz interference since the experimental shape factor does not change sign for positrons and electrons.<sup>30</sup>

At the present time, the theory provides no explanation for a shape factor of the form  $(1+b/W)$ . The finite de Broglie wavelength correction included in the analysis is in the right direction to account for the observed deviations but the magnitude of the correction is much too small to explain the observations. It is possible that further considerations of refinements of this type may offer an explanation of the observations.

The outer-screening correction is in the right direction to explain the deviation observed in the  $\text{Na}^{22}$  spectrum but in the wrong direction to explain the spectra of  $\text{In}^{114}$ ,  $\text{Y}^{90}$ , and  $\text{P}^{32}$ . Because of the low  $Z$  of  $\text{Na}^{22}$ , it is unlikely that even more exact calculations of the screening correction would account for the observed deviation in this spectrum.

Other experiments have confirmed the deviations in  $\text{Y}^{90}$  and  $\text{P}^{32}$  but there appears to be some disagreement about the magnitudes of the deviations observed in these spectra. Since the observed effect is small, the differences possibly arise in part from differences in the treatment of the data.

At present, the term  $(1+b/W)$  may be regarded as an empirical correction which is capable of explaining the observed shapes in the case of these four pure Gamow-Teller transitions.

#### ACKNOWLEDGMENTS

The authors wish to thank Professor E. Konopinski, Professor T. Kotani, and Professor M. Ross for helpful discussions concerning the theoretical aspects of this problem. Professor J. S. Levinger is also thanked for his helpful suggestions and comments. D. Camp and D. Smith helped in the preparation of the figures.

<sup>30</sup> Of course, in the light of recent experiments which indicate that the interaction strengths are such that  $C_V = C_V'$  and  $C_A = C_A'$ , Fierz interference is expected to be zero.

#### APPENDIX. $\text{Na}^{22}$ ELECTRON DISTRIBUTION

It was pointed out earlier in the paper how these studies were correlated to the principal problem of this paper. This work was also motivated by the work of Bruner who reported<sup>31</sup> an electron distribution accompanying the positron decay of  $\text{Sc}^{44}$ . He observed an electron distribution in the energy range 30–150 keV with an intensity of  $\approx 4\%$  relative to the intensity of the positron spectrum. There was no satisfactory explanation given for the observed electrons. Blue and Bleuler repeated the work on  $\text{Sc}^{44}$  and reported<sup>32</sup> fewer electrons by a factor of 5–10 than Bruner reported. They felt that their results were consistent with no electrons observed in this energy range 30–150 keV. They also reported<sup>33</sup> electrons observed in the decay of  $\text{Na}^{22}$ . Their  $\text{Na}^{22}$  measurements were made, however, only as tests of the procedures and little was reported concerning this work.

$\text{Na}^{22}$  sources 3 and 4 were used in this investigation. Six measurements were made from 20 to about 200 keV with a proportional counter which had a thin unsupported window (cutoff  $\approx 5$  keV) and six measurements from 2–60 keV with Lektromesh-supported Zapon windows with cutoffs either  $\lesssim 1$  keV or  $\lesssim 2$  keV. The two sets of data were normalized.

An electron distribution was observed which decreased rapidly with energy, approaching the background counting rate in the region between 40 and 150 keV.<sup>9,11</sup> In Fig. 9, the observed electron distribution as a function of momentum is plotted for the two sources. The curves are the best fit to the data. Only a few points are shown in order to indicate the size of the errors. The limits of error, which include the statistical deviation in the counting rates and background, are large because of the low counting rates (maximum counting rate was 10 counts/min above a background of about 25 counts/min).

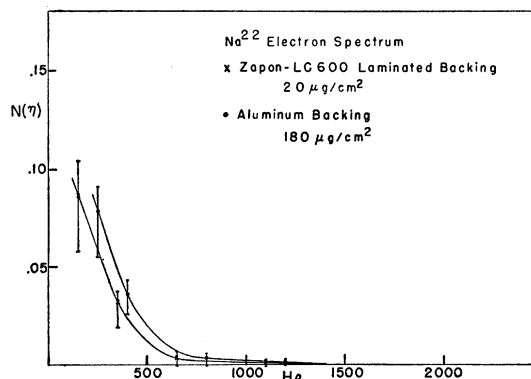


FIG. 9.  $\text{Na}^{22}$  electron distribution as a function of momentum for sources 3 and 4.

<sup>31</sup> J. A. Bruner, *Phys. Rev.* **84**, 282 (1951).

<sup>32</sup> J. W. Blue and E. Bleuler, *Phys. Rev.* **100**, 1324 (1955).

<sup>33</sup> W. Paul and H. Steinwedel, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 1, Eq. (2).

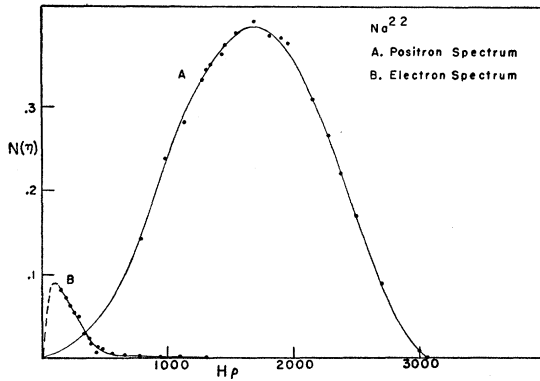


FIG. 10.  $\text{Na}^{22}$  positron spectrum and electron distribution as a function of momentum for source 4.

Figure 10 gives a comparison plot of the positron spectrum and the electron distribution obtained with source 4. The extrapolation of the electron distribution will be discussed later. The measured ratio  $N_{\beta^-}/N_{\beta^+}$  is  $(2.2_{-1.1}^{+0.9}) \times 10^{-2}$  for source 4. The electron intensity,  $N_{\beta^-}$ , was measured only for energies  $> 2$  kev (the lowest energy at which reasonably reliable electron data were measured) and  $N_{\beta^+}$  is the total positron intensity. The errors quoted arise from the different areas under the electron distribution curves drawn through the upper and lower limits of error placed on the experimental points. The distribution measured with the more thinly backed source fell lower (Fig. 9). Other studies of the effects of source backing indicate that the value 2.2 should be decreased by possibly 10–20%. Several tests were made to determine the origin of the observed electrons. In Fig. 11, one notes that the counting rates do go to zero as the energy decreases. This is because of counter window cutoff. Although not plotted, the data taken with the unsupported window decreased to zero at  $\approx 5$  kev as expected. This indicates that the observed electrons are not particles produced in the walls of the chamber by positron scattering, Compton scattering, or pair production of the 1.28-Mev gamma ray. Such particles scattered into the detector would not result in a window cutoff. Careful measurements were made of the background in the spectrometer both with and without the  $\text{Na}^{22}$  sources in the spectrometer.

A test was made to see whether the electrons originate through some forward scattering process of the positrons or gamma rays in the source material or cover film by placing additional cover films over the source. An additional Zapon cover film of about  $10 \pm 5 \mu\text{g}/\text{cm}^2$  was placed over source 3 and then a second Zapon film of  $60 \pm 10 \mu\text{g}/\text{cm}^2$  was added. A plot of the data taken with and without the two cover films ( $70 \pm 15 \mu\text{g}/\text{cm}^2$ ) is seen in Fig. 11. Measurements were made only one day apart with the 10- and then with the additional 60- $\mu\text{g}/\text{cm}^2$  cover with the same counter and window. The two sets of data agreed above 10 kev with each other and with the previous data (Fig. 11). The thicker

counter window used in these measurements is probably responsible for the drop in intensity beginning at 10 kev in comparison to the data without the extra cover films. From the experimental values of counter window cutoff, the range-energy relation of Flammersfeld<sup>33</sup> (which appears to give slightly higher values for the cutoff energy as a function of the thickness below  $100 \mu\text{g}/\text{cm}^2$ ) and the energy loss equation,<sup>33</sup> it seems safe to conclude that the 10- $\mu\text{g}/\text{cm}^2$  film and most probably even the 70- $\mu\text{g}/\text{cm}^2$  film should not distort the observed distribution above 10 kev. It seems safe to conclude that the electrons do not originate through some forward scattering process of the positrons or gamma rays in the cover films. If the electrons did originate in this manner, then the electron distribution would be expected to increase rather sharply with the added cover films over the source and it does not.

The decay of the electron distribution was checked over a period of five months. The half-life of the decay agreed with that of  $\text{Na}^{22}$ . The shape of the distribution excludes the interpretation of the electrons resulting from a beta disintegration process in either  $\text{Na}^{22}$  or an impurity. All of the evidence indicates that the observed electrons are directly associated with the decay of  $\text{Na}^{22}$ .

The intensity and distribution of the observed electrons from  $\text{Na}^{22}$  are in sharp disagreement with the observations of Bruner<sup>31</sup> on  $\text{Sc}^{44}$ . The large number of electrons observed by Bruner in the range 30–150 kev is difficult to explain and appears from the work of Blue and Bleuler<sup>32</sup> to be at least partially of instrumental origin. Their results are essentially in agreement with the present  $\text{Na}^{22}$  studies although they did not go to very low energies in their measurements.

A possible theoretical explanation of the electrons observed in the  $\text{Na}^{22}$  decay is found in the independent

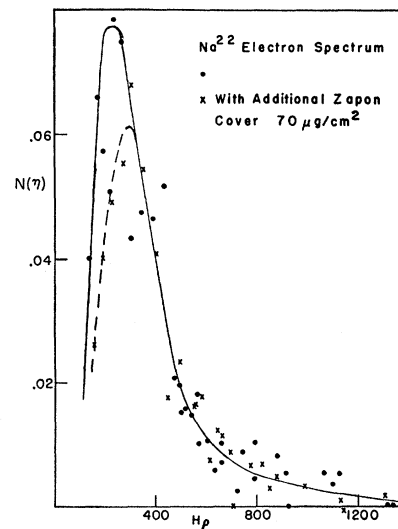


FIG. 11.  $\text{Na}^{22}$  electron distribution as a function of momentum for source 3.



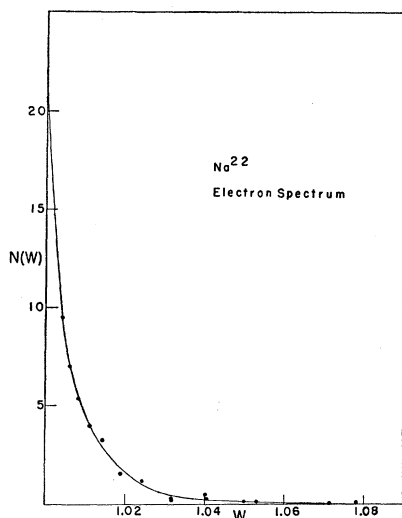


FIG. 12.  $\text{Na}^{22}$  electron distribution plotted as a function of energy. The extrapolation is made in a manner consistent with the theory of "shake-off" electrons.

work of Feinberg,<sup>34</sup> Migdal,<sup>35</sup> and Levinger.<sup>36</sup> They have shown that, because of the sudden change in the nuclear charge in the process of beta decay, excitation and ionization of the atomic electrons are expected. In this process, the energy release of the decay is shared between the beta particle, neutrino, and atomic electron. This small effect does not measurably change the distribution of the particles in beta decay.

The probabilities of atomic electrons being excited or ejected in the process of beta decay were calculated (Feinberg for the  $K$  shell, Levinger for the  $K$  and  $L$  shells, and Migdal for the  $K$ ,  $L$ , and  $M$  shells). The  $K$ -shell calculations agree with each other, but the  $L$ -shell calculations do not. Primakoff and Porter have treated the case of  $K$  capture.<sup>37</sup> The ejected electrons are sometimes referred to as "shake-off" electrons.

To compare the  $\text{Na}^{22}$  electron data with the probabilities predicted for "shake off" electrons, a plot of  $N(W)$  vs  $W$  was made (Fig. 12). The extrapolation of the data from 2 keV to zero energy in this plot was made in a manner consistent with the distribution predicted for "shake-off" electrons. The extrapolation is still somewhat arbitrary. The extrapolation in Fig. 10 was constructed from that in Fig. 12. If this extrapolation of the experimental curve to zero energy is included, then the ratio of the total number of electrons to the total number of positrons,  $N_{\beta^-}/N_{\beta^+}$ , becomes  $(4.3_{-0.4}^{+1.4}) \times 10^{-2}$  for the data from source 4. The limits of error do not include possible changes in the extrapolation but are statistical deviations about the average curves. Changes in the extrapolation of not too unreasonable a nature could increase the 4.3 by a factor

of 2 or possibly more but could not decrease the value much below 3.

If screening effects and the change of one in  $Z$  are neglected, the ionization probabilities for the  $K+L$  shells per positron decay ( $N_{\beta^-}/N_{\beta^+}$ ) are  $7.4 \times 10^{-2}$  from Migdal's work and  $2.75 \times 10^{-2}$  from Levinger's work. The agreement between theory and experiment is most likely partly fortuitous considering the large experimental and theoretical uncertainties. The neglected effects mentioned above are quite large for  $\text{Na}^{22}$ . The correction to the theoretical numbers for the  $\approx 10\%$   $K$ -capture in  $\text{Na}^{22}$  is quite small.

The observed  $\text{Na}^{22}$  electron distribution does not appear to peak as strongly at low energy as predicted by the theory of "shake-off" electrons. The theory predicts that the mean energy of the electrons from a given shell is approximately the binding energy of the shell and that the distribution should fall off as  $1/W^4$  for energies much greater than the binding energy of the shell. The best fit to the  $\text{Na}^{22}$  data for  $W/E_K > 5$  is  $W^x$ , where  $x = -2.3_{-0.8}^{+1.8}$  and  $E_K$  is the  $K$ -shell binding energy in  $\text{Ne}^{22}$ . The errors represent the curves drawn through the limits of statistical error. The theoretical value is just within the experimental limits of error. The intensity of electrons observed for energies  $> 2$  keV appears to be larger than predicted by Levinger or Migdal. Two of the approximations made in the general calculations of Levinger and Migdal are poor for  $\text{Na}^{22}$ . From Feinberg's paper, it is possible to estimate the error introduced in the  $K$ -shell probability arising from neglecting the change of one in  $Z$ . For  $\text{Na}^{22}$ , taking into account the change in  $Z$ , the probability of  $K$ -shell ionization becomes  $0.92/Z^2$  in comparison to the value of  $0.64/Z^2$  given by Levinger and Migdal. This is about a 45% increase. Further corrections of this nature would be expected for the  $L$ -shell probabilities.

The other poor approximation is the neglect of screening of the electrons. Because of screening, the probabilities are proportional to  $1/(Z-\sigma)^2$ , where  $\sigma$  is the screening constant, instead of  $1/Z^2$ . This correction is small for the  $K$  shell but becomes large for the  $L$  shell. If the theoretical probabilities are calculated from Migdal's equations with screening constants given by Hartree,<sup>38</sup> the total probability of ionization for the  $K+L$  shells becomes 15.8% for  $Z=11$  and 20.8% for  $Z=10$ . These values do not include the change in  $Z$  correction mentioned in the last paragraph.

In addition to the work of Bruner, and of Blue and Bleuler, there are two other measurements of electron distributions accompanying positron decay or  $K$  capture. Soltysik observed electrons accompanying the  $K$ -capture decay of  $\text{Be}^7$  with a  $180^\circ$  magnetic spectrometer.<sup>39</sup> Miskel and Perlman observed electrons accompanying the  $K$ -capture decay of  $\text{A}^{37}$  with a proportional

<sup>34</sup> E. L. Feinberg, J. Phys. U.S.S.R. 4, 423 (1941).

<sup>35</sup> A. Migdal, J. Phys. U.S.S.R. 4, 449 (1941).

<sup>36</sup> J. S. Levinger, Phys. Rev. 90, 11 (1953).

<sup>37</sup> H. Primakoff and F. T. Porter, Phys. Rev. 89, 930 (1953).

<sup>38</sup> D. R. Hartree, *The Calculation of Atomic Structures* (John Wiley and Sons, Inc., New York, 1957).

<sup>39</sup> E. A. Soltysik, Ph.D. thesis, Indiana University (unpublished).

counter.<sup>40</sup> The total number of electrons observed in both experiments agreed reasonably well with the "shake-off" theory but, in both cases, the observed distributions were in disagreement with the theory. In Soltysik's work, the total experimental and theoretical probabilities agreed only if the experimental curve was extrapolated in a manner consistent with the experimental data but contrary to the theoretical curve. In the measured electron momentum range  $(0.20-1.50)m_0c$ , the experimental value of the electron intensity in  $\text{Be}^7$  is 10-40 times greater than predicted by theory. The proportional counter experiment would not be expected to give a good measure of the electron distribution but the magnetic spectrometer measurements should have. In both experiments, the experimental distributions do not peak as sharply at low energies as the theory predicts.

Other investigators have looked at the  $K$  and  $L$  x-rays accompanying beta decay or  $K$  capture in other isotopes. In such studies,<sup>41</sup> reasonable agreement has been found between the total theoretical ionization probabilities for the  $K$  and  $L$  shells and the experimental values.

Two other recent papers concerning the ionization of the atomic electrons following the beta decay of  $\text{Kr}^{85}$  are of interest in this problem. Snell and Pleasonton studied the ionization of the atomic electrons following the beta decay of  $\text{Kr}^{85}$  by observing the charge spectrum of the recoiling ions.<sup>42</sup> Experimentally, they found the

ionization of the  $K+L+M$  shells to be 9.9% and that of the  $N$  shell 10.9%. Migdal's equations predict the probability of ionization of the  $K+L+M$  shells to be 1.65% (for the probability  $\sim 1/Z^2$ ) or 3.17% [probability  $\sim 1/(Z-\sigma)^2$ ]. The predictions of Levinger are even lower than Migdal's for the  $K+L$  shells.

To explain the experimental results, Green calculated for  $\text{Kr}^{85}$  the probability of ionization of the  $K$ ,  $L$ ,  $M$ , and  $N$  shells.<sup>43</sup> For his more exact calculations, Green used Hartree wave functions for the initial and final states of the specific nuclei in question and took into account the electron screening. He obtained a value of 9% for the  $N$ -shell ionization probability and 4.4% for the  $K+L+M$  shells. He also suggested how this value of 4.4 might be increased to 6.5. His work suggests that the results of the general calculations of Levinger and Migdal are too low and that for a comparison of theory with experiment the ionization probabilities should be calculated for the specific isotope.

In conclusion, an electron distribution from  $\text{Na}^{22}$  has been measured for energies  $>2$  kev. The intensity of the electron distribution in this range relative to the positron spectrum is  $(2.2_{-1.1}^{+0.9}) \times 10^{-2}$  (excluding a few percent decrease for backing effects). On the basis of the  $\text{Na}^{22}$  and  $\text{Kr}^{85}$  results, it seems likely that the ionization probabilities predicted by the general calculations of Levinger and Migdal are too low. The distribution predicted for "shake-off" electrons may also be questioned on the basis of the present results. For a more meaningful comparison of theory and experiment, ionization probabilities should be calculated for the isotope being studied as was done for  $\text{Kr}^{85}$ .

<sup>40</sup> J. A. Miskel and M. L. Perlman, Phys. Rev. **94**, 1683 (1954).

<sup>41</sup> For references to these works see reference 37, F. Boehm and C. S. Wu, Phys. Rev. **93**, 518 (1954), and W. Rubinson and J. J. Howland, Phys. Rev. **96**, 1610 (1954).

<sup>42</sup> A. H. Snell and F. Pleasonton, Phys. Rev. **107**, 740 (1957).

<sup>43</sup> A. F. S. Green, Phys. Rev. **107**, 1646 (1957).