

## Nuclear Recoil Following Neutrino Emission from Beryllium 7\*

RAYMOND DAVIS, JR.

*Chemistry Department, Brookhaven National Laboratory, Upton, New York*

(Received March 6, 1952)

The nuclear recoil spectrum of the electron capture isotope  $\text{Be}^7$  was measured from nearly monolayer deposits of the isotope on lithium fluoride and wolfram surfaces. The recoil spectrum was measured with an electrostatic analyzer having a two percent resolution. The spectra exhibited a peak at the high energy end of the spectrum consistent with the hypothesis of single neutrino emission in  $\text{Be}^7$ . The maximum recoil ion energy observed was  $55.9 \pm 1.0$  electron volts.

### I. INTRODUCTION

IN the theory of beta-radioactivity it has been assumed that part of the energy of the decaying nucleus is carried away by a single neutrino. Crane,<sup>1</sup> in a review of the subject of energy and momentum relations in beta-decay, pointed out that this assumption could be examined experimentally by studying the nuclear recoil spectrum of an electron capture isotope. If the energy is emitted as a single neutrino, the momentum of the recoiling nucleus must be equal to the momentum of the emitted neutrino, with the result that all nuclei should recoil with the same momentum. However, if the energy is emitted in the form of several neutrinos in random directions, the momentum of the recoiling nucleus will be equal and opposite to the resultant momenta of the neutrinos, and the recoil spectrum will be continuous extending from zero to a maximum corresponding to the emission of all neutrinos in the same direction. Another method of testing the single neutrino hypothesis is to examine the nuclear recoil spectrum from a beta-emitting nucleus by defining the direction and energy of the electron and the direction of the recoil. Single neutrino emission should then lead to monoenergetic recoils. This point has been tested by the experiments of Sherwin with  $\text{P}^{32}$ . He concludes that the missing energy is carried away by a single neutrino.<sup>2</sup>

Two experiments have been conducted with the electron capture isotope  $\text{Be}^7$ , by Allen<sup>3</sup> and by Smith and Allen.<sup>4</sup> This isotope has the largest recoil energy of the known pure electron capture isotopes and is therefore a good choice for the experiment. The recoil spectra observed by these experimenters, from a source of  $\text{Be}^7$  on the surface of a solid, showed a distribution in recoil ion energies which was attributable to the recoiling ion losing energy by collisions with neighboring atoms on the surface. Because of this difficulty, definite conclusions could not be reached concerning the question of single or multiple neutrino emission. In the later work, the end point of the recoil spectrum was measured

at  $56.6 \pm 1.0$  ev, a value that agrees with the expected result.

More recently, Rodeback and Allen<sup>5</sup> have reported an experiment with the electron capture isotope  $\text{A}^{37}$  in which the recoil spectrum was measured in the gas phase. A peak was observed at the correct energy, 9.7 electron volts, which had a width corresponding to the resolution of the apparatus and was therefore in agreement with the hypothesis of single neutrino emission. A further experiment with  $\text{A}^{37}$  is in progress,<sup>6</sup> but to date only the experimental method has been reported.

So far, no recoil experiments have been reported with positron-emitting isotopes, although many of them have high recoil energies and would be well suited for an experiment. These isotopes should exhibit a continuous recoil spectrum displaying a sharp peak beyond the continuous spectrum resulting from single neutrino emission in the  $K$ -capture branch. Isotopes with high transition energies will in general have a weak  $K$ -capture branch, and the peak will be small compared to the continuous spectrum. However, if future recoil experiments reveal this peak cleanly, it will serve as an added verification of the single neutrino hypothesis.

It might be pointed out that recoil experiments of the type discussed can only show that the missing energy disappears in one parcel in a defined direction, but they are not capable of conclusively demonstrating that the parcel is composed of one or more particles. The only means of examining this possibility is to measure the cross section for the absorption of neutrinos and compare the results with the theoretically expected cross section for single neutrino emission. But neutrino absorption experiments are barely within the limit of available experimental techniques.

The present study is a reinvestigation of the recoil spectrum of  $\text{Be}^7$ . In this work the spectrum was observed from a solid surface with an electrostatic analyzer of relatively high resolution,  $\Delta E/E = 0.018$ . A study was made of the methods of preparing the recoil sources in an effort to obtain a monolayer distribution of  $\text{Be}^7$  on a surface, and thereby obtain a recoil spectrum free from surface effects. This was accomplished to a

\* Research carried out under the auspices of the AEC.

<sup>1</sup> H. R. Crane, *Revs. Modern Phys.* **20**, 278 (1948).

<sup>2</sup> C. W. Sherwin, *Phys. Rev.* **75**, 1799 (1949).

<sup>3</sup> J. S. Allen, *Phys. Rev.* **61**, 692 (1942).

<sup>4</sup> P. B. Smith and J. S. Allen, *Phys. Rev.* **81**, 381 (1951).

<sup>5</sup> G. W. Rodeback and J. S. Allen, *Phys. Rev.* **83**, 215 (1951).

<sup>6</sup> T. Ohkawa, *J. Phys. Soc. Japan* **6**, 197 (1951).

degree that permitted conclusions to be reached in favor of single neutrino emission in Be<sup>7</sup>.

The nuclear recoil energy  $E_R$  resulting from the emission of a single neutrino of momentum  $E_\nu/c$  is given by

$$E_R = E_\nu^2 / 2Mc^2, \quad (1)$$

where  $M$  is the mass of the recoiling atom and  $c$  is the velocity of light. The isotope Be<sup>7</sup> decays according to the scheme given in Fig. 1.<sup>7-9</sup> In 89 percent of the decays 0.864±0.003 Mev of energy is emitted, and if this amount of energy is carried away by a single neutrino all nuclei should recoil with 57.3±0.5 electron volts energy according to (1).

The 11-percent branch which goes to the excited state of Li<sup>7</sup> would exhibit a continuous recoil spectrum extending from zero to 57.3±0.5 eV from the nearly simultaneous emission of a neutrino and a gamma-ray.<sup>10</sup>

To study the neutrino recoil spectrum of Be<sup>7</sup> one observes the recoils from a solid surface, since it is very difficult to prepare carrier-free gaseous compounds of beryllium that would be suitable for the experiment. In using a surface as a site of recoils one must prepare the radioactive isotope as an extremely thin layer on the surface, approaching a bare monolayer if the observed recoil spectrum from the surface is to be a true representation of the initial recoil spectrum. In the present case, the recoil energy is low, only about twenty times the magnitude of chemical bond energies, and the surface effects must be made small if one is to distinguish between single and multiple neutrino emission. Thin layers of radioactive atoms on surfaces can be prepared by selectively evaporating the contaminants from a thick deposit on a surface leaving the desired radioactive atoms on the surface, or by evaporating the radioactive atoms and condensing them on a fresh surface. The first method was used by Allen<sup>3</sup> in the first experiment with Be<sup>7</sup>. A combination of these two means has been employed successfully by Sherwin<sup>11</sup> in preparing monolayer sources of both P<sup>32</sup> and Y<sup>91</sup> for recoil studies of electron-neutrino angular correlations and by Smith and Allen<sup>4</sup> in their work on Be<sup>7</sup>. In the present work, both of these means of preparing surfaces were used.

Since the energy spectrum was measured by electrostatic means, a further requisite is that excited lithium atoms leaving the surface and traveling at  $4 \times 10^6$  cm/sec be ionized. If the time required for the atom to move away from the surface is long compared to the time of adjustment of the electrons on the surface, then the recoiling atom will be in equilibrium with the

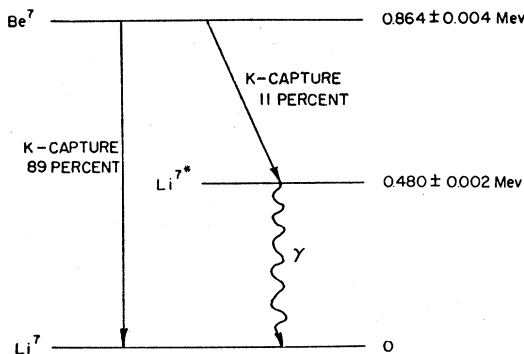


FIG. 1. The decay scheme of Be<sup>7</sup>. The Be<sup>7</sup>-Li<sup>7</sup> mass difference of 0.864±0.003 Mev was computed from the  $Q$  value (see references 7 and 8) of 1.646±0.002 Mev for the reaction  $\text{Li}^7(p,n)\text{Be}^7$  and the neutron-proton mass difference of 0.782±0.002 Mev (see reference 9).

surface, and the ratio of the number of ionized atoms to neutral atoms will be given by

$$n_+/n_a = (\omega_+/\omega_a) \exp[-(I-\phi)/kT], \quad (2)$$

where  $\omega_+$  and  $\omega_a$  are statistical weights,  $I$  is the ionization potential of the atom, and  $\phi$  the work function of the surface. This equation has been verified for potassium atoms in thermal equilibrium with tungsten surfaces.<sup>12</sup> In recoil experiments one insures that the conditions are favorable for ionization by employing surfaces with high work functions. For experiments with Be<sup>7</sup> the work function of the surface should be greater than 5.36 volts, the ionization potential of lithium. Surfaces of high work function, oxygenated wolfram and lithium fluoride, were used in the present experiments.

## II. EXPERIMENTAL

### Chemical Separation

The Be<sup>7</sup> was prepared by bombarding cp metallic lithium for 500 microampere hours with the 8.5-Mev proton beam from the University of Pittsburgh cyclotron. The portion of the target struck by the beam was removed and dissolved in water. The solution was made acid and 50 mg of CuCl<sub>2</sub> and FeCl<sub>3</sub> were added as carrier. After adjusting the solution to 0.3*N* in HCl, copper was precipitated and removed as sulfide. Following this the solution was boiled to remove hydrogen sulfide, and a few drops of hydrogen peroxide were added to oxidize the ferrous ion. The solution was made alkaline with ammonium hydroxide precipitating ferric hydroxide which serves to carry the Be<sup>7</sup>. The precipitate was dissolved in acid, and reprecipitated with ammonium hydroxide. This precipitation was repeated once more, and the final ferric hydroxide precipitate dissolved in about 5 milliliters of 7.8*N* HCl. The aqueous solution was extracted three times with 10 milliliter portions of isopropyl ether which serves to

<sup>7</sup> Herb, Snowden, and Sala, Phys. Rev. **75**, 246 (1949).

<sup>8</sup> Shoupp, Jennings, and Jones, Phys. Rev. **76**, 502 (1949).

<sup>9</sup> Taschek, Argo, Hemmendinger, and Jarvis, Phys. Rev. **76**, 325 (1949).

<sup>10</sup> R. E. Bell and L. G. Elliott, Phys. Rev. **76**, 168 (1949), have shown that the lifetime of the excited state of Li<sup>7</sup> is  $(0.75 \pm 0.25) \times 10^{-13}$  second.

<sup>11</sup> C. W. Sherwin, Phys. Rev. **82**, 52 (1951); Rev. Sci. Instr. **22**, 339 (1951).

<sup>12</sup> M. J. Copley and T. E. Phipps, Phys. Rev. **48**, 960 (1935).

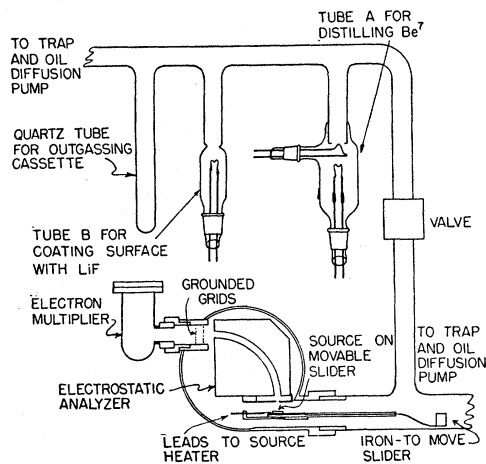


FIG. 2. Schematic diagram of the apparatus used to study the recoil spectrum of  $\text{Be}^7$ .

extract the iron leaving the  $\text{Be}^7$  in the solution. The solution was adjusted to pH 5.8 with ammonium hydroxide and extracted with a 5-milliliter portion of 10 percent solution of acetylacetone in benzene. The organic phase containing the activity was washed with 20-milliliter portions of distilled water until the wash water on evaporation showed no signs of a residue. The  $\text{Be}^7$  was back-extracted from the organic phase with a 1-milliliter portion of constant boiling hydrochloric acid or a 5 percent oxalic acid solution. The  $\text{Be}^7$  contained in 1 milliliter of solution was evaporated in a drying oven, a drop at a time, on a wolfram ribbon which could be installed in the vacuum system for the evaporations.

### Source Preparations

Sources were prepared by selective evaporation in vacuum from wolfram ribbons that were heated by passing a current through them. Wolfram was chosen because the evaporations were carried out at temperatures in the range  $1400^\circ$  to  $1600^\circ\text{C}$ , and it was necessary to use a metal with a very low vapor pressure at high temperatures. Either wolfram or tantalum would serve this purpose equally well, but these metals do have the disadvantage of reacting to some extent with acids. In some experiments, iridium was used and it was found that the metal evaporates slightly at temperatures above  $1650^\circ\text{C}$ , but it does have the advantage of resistance to acids. A wolfram ribbon  $0.005 \times 0.125$  inch was bent in the form of a flattened *M* as shown in Fig. 2 (lower ribbon of the tube *A*) and spot welded to two heavy wolfram leads held parallel by a glass bridge. This assembly was connected by means of steel couplings to wolfram leads sealed through a taper joint. The ribbon holder was arranged in this way so that it could be removed from the vacuum line and weighed. It was found that these ribbon assemblies could be weighed reproducibly with an Ainsworth microbalance to about  $2 \times 10^{-6}$  gram.

Preliminary studies were made of the distillation behavior of  $\text{Be}^7$ . Evaporations were made from the metal ribbons onto weighed platinum foils about two centimeters in diameter that could be removed from the vacuum line and weighed after the evaporation. By measuring the weight loss of the ribbon, the gain in weight of the platinum disks, and determining the amount of  $\text{Be}^7$  by counting, the vaporization properties of  $\text{Be}^7$  and its contaminants could be studied as a function of temperature. The temperature of the wolfram ribbon was measured with an optical pyrometer. In these preliminary experiments the optimum conditions for distilling a  $\text{Be}^7$  source were determined. It was found that upon evaporating a hydrochloric or oxalic acid solution containing the  $\text{Be}^7$ , the residues amounted to 100 to 200 micrograms. Upon heating the ribbon to around  $1400^\circ\text{C}$  in vacuum most of these residues distilled off leaving 70 to 90 percent of the  $\text{Be}^7$  on the ribbon. The weight of the wolfram ribbon upon heating from  $1400^\circ$  to  $1500^\circ$  loses about 5 to 10 micrograms, and the platinum disk receiving the distillate did not increase in weight within the limit of sensitivity of the weighing, less than 2 micrograms. By following the procedure of heating the ribbon to  $1400^\circ$  to remove contaminants, then distilling onto a surface by increasing the temperature to about  $1500^\circ$ , the activity could be transferred forming a deposit weighing less than  $2 \times 10^{-6}$  g/cm<sup>2</sup>.

The weighing technique used in these preliminary experiments was sufficiently accurate to establish the optimum conditions for separating the contaminants from the  $\text{Be}^7$  by evaporation. However, much more accurate weighings would be required to insure that the amount of material distilled onto a surface was less than an amount corresponding to a monolayer. A monolayer  $3 \times 10^{-8}$  cm thick with an average density of 3 g/cm<sup>3</sup> would weigh  $10^{-7}$  gram, and to detect weight changes of this order would require about a twenty-fold increase in sensitivity. It should be pointed out that the weight of  $\text{Be}^7$  itself distilled in the sources as only around  $4 \times 10^{-10}$  gram, an amount corresponding to 1/200th of a monolayer. In addition to the requirement of reducing the contaminants distilling along with the  $\text{Be}^7$  to less than  $10^{-7}$  g/cm<sup>2</sup>, it is essential that all beryllium atoms be on the surface, and not imbedded in crystalline aggregates. In the present experiments the only way of knowing whether the latter condition was attained was to examine the quality of the recoil spectrum.

The preparation of the recoil sources used in the measurements to be described were based upon the distillation behavior experienced in the preliminary trials. The recoil spectra will be reported for two sources prepared by quite different techniques. Source I was prepared by evaporation onto a freshly deposited lithium fluoride surface in which a double evaporation was employed to reduce the quantity of contaminants accompanying the  $\text{Be}^7$  evaporation. After preparation,

this source was transported through the vacuum system and placed under the electrostatic analyzer. Source II was prepared by evaporation of contaminants from a wolfram ribbon leaving some of the Be<sup>7</sup> remaining on the ribbon. The recoil spectrum of the Be<sup>7</sup> remaining on the wolfram ribbon was measured. The details of preparing these sources are as follows.

Source I, prepared by double vacuum evaporation. One milliliter of a 5 percent oxalic acid solution containing the separated activity was evaporated, a drop at a time, on the lower wolfram ribbon in tube *A* shown in Fig. 2. It was heated to 1380°C in vacuum to distill off the easily vaporizable impurities. The upper ribbon of tube *A* was heated to 2000°C to clean it thoroughly, and then rotated down over the lower ribbon. The lower ribbon was heated to 1830°C to distill the activity to the upper ribbon. The upper ribbon was then heated to 1300°C to remove contaminants vaporizing at this temperature that may have accompanied the activity on the first distillation. The upper ribbon was then rotated in position to distill a second time onto the lithium fluoride surface. The lithium fluoride surface was prepared by distilling the fused salt from a wolfram ribbon in tube *B* (Fig. 2). The lithium fluoride was distilled onto a one-inch disk of 0.005 inch platinum foil spot welded to a nickel ring so that it could be moved inside the vacuum line with an external bar magnet. Prior to coating with lithium fluoride this assembly was outgassed by heating it for 17 hours at 890°C. The lithium fluoride coated disk was then placed over the upper ribbon, and the second distillation of Be<sup>7</sup> was accomplished by heating the ribbon to 1525°C. The recoil source thus prepared was moved through the valve into the side of the vacuum system containing the electrostatic analyzer. Later the source was compared to a Co<sup>60</sup> standard and found to contain about  $5 \times 10^6$  disintegrations per second of Be<sup>7</sup>.

Source II, prepared by evaporation. A milliliter of hydrochloric acid solution containing Be<sup>7</sup> was evaporated on a wolfram ribbon. A ribbon was heated to 1560°C in a vacuum system to evaporate contaminants, and the cleaned-up ribbon used as a recoil source. The ribbon lost about 90 percent of the Be<sup>7</sup> activity from this treatment. The resulting source contained about  $5 \times 10^6$  disintegrations per second. The source was removed from the vacuum system and installed under the entrance slit of the electrostatic analyzer.

### The Vacuum System and the Electrostatic Analyzer

The apparatus, shown in Fig. 2, consists of two separately pumped vacuum systems interconnected by a valve. On one side of the valve was located apparatus for preparing the sources and on the opposite side the electrostatic analyzer and the electron multiplier for measuring the recoil ion spectra. The valve served to protect the measuring apparatus from becoming contaminated during the evaporation operations. The side of the vacuum system used to distill the sources was

pumped by a 2-stage oil diffusion pump through a liquid nitrogen cooled trap giving a vacuum of around  $5 \times 10^{-7}$  mm of Hg.

The recoil ion spectra were measured by means of a 90° cylindrical electrostatic analyzer of plate radii 3.750 and 4.125 inches. The plates of the analyzer were made of brass 1.63 inches thick, and were bolted to a one-half inch thick Pyrex glass plate. The analyzer was held in a cylindrical brass vacuum chamber 3 inches deep by 8 inches in diameter, having a 6-cm diameter pumping tube and a port into which the electron multiplier case is fitted. A vacuum of  $5 \times 10^{-7}$  mm of Hg was maintained in the chamber by means of a glass 3-stage oil diffusion pump and a large liquid nitrogen cooled trap. A track was located in the pumping tube on which rode a small platform slider that could be moved back and forth by means of an armature and an external bar magnet. The source fitted on a stainless steel hot plate attached to the sliding platform. A wolfram wire heating element attached to the brass chamber fitted under the hot plate when the source was located under the entrance of the analyzer. A calibration of the hot plate temperature at various settings of the heater current was made with a source disk identical to the one used in the experiment and having a fine wire thermocouple spot welded to its surface. By means of this hot plate the source could be heated to over 300°C.

An electron multiplier designed by Dr. Robert P. Stone<sup>13</sup> of the RCA laboratory was used to detect the recoiling ions. The multiplier was held in a case (Fig. 2) which was operated at a high negative potential and was insulated from the grounded vacuum case by a Lucite adapter. The multiplier was operated with its center dynode grounded, and a total applied potential across the tube of 4000 volts. The first dynode was made negative so that the low energy ions leaving the analyzer were accelerated by 2000 volts onto the first dynode. To screen the analyzer from the high potential two grids were placed between the multiplier and the exit of the analyzer. The analyzer was initially designed so that the source and the screen grid were located at the foci of the analyzer. With this arrangement the resolution of the analyzer,  $\Delta E/E$ , was 0.11. To improve the resolution a slit 0.199 inch wide was placed 0.375 inch from the entrance, and a second slit 0.063 inch wide by 0.75 inch long was placed 1.5 inches from the exit of the analyzer. With these slits in place the resolution  $\Delta E/E$  was increased to 0.018. A resistor, center tapped to ground, was connected between the plates of the analyzer so that the outer and inner plates ran at equal potentials above and below ground, respectively.

The energy  $E$  of an ion of charge  $z$  passing through

<sup>13</sup> The electron multiplier was loaned to me through the courtesy of Dr. Irving Wolff, and Dr. Stone of the RCA Laboratories Division, Radio Corporation of America, Princeton, New Jersey. The multiplier is described in Dr. Stone's article, *Rev. Sci. Instr.* **20**, 935 (1949).

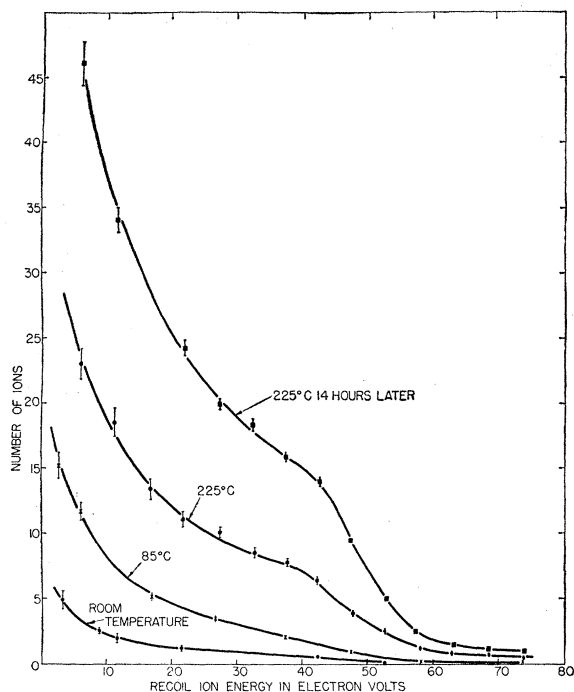


FIG. 3. Recoil spectra from a double distilled  $\text{Be}^7$  source on a lithium fluoride surface measured at a series of surface temperatures.

the center of the analyzer is related to the applied potential difference,  $V_1 - V_2$ , and the radii of the inner and outer plates  $r_1$  and  $r_2$  respectively by,

$$E = z(V_1 - V_2)/2 \ln(r_1/r_2). \quad (3)$$

For the analyzer used in these experiments the expression becomes for singly charged ions,

$$E = 5.246(V_1 - V_2). \quad (4)$$

The performance of the analyzer was checked with thermal ions from a heated wolfram ribbon placed under the entrance slit (Fig. 4). The ribbon was set at a measured negative potential with respect to the entrance slit to accelerate the ions into the analyzer. The peak position and shape were measured at several ion energies. The resolution expressed above was taken as the half-width of the peak divided by the ion energy. The position of the monoenergetic ion peaks for various ion energies agreed with Eq. (4) except that each peak was displaced an equal amount from the energy expressed by the equation as a result of the contact difference in potential between the thermal ion source and the brass analyzer.

Recoil spectra were measured by observing the counting rate on the electron multiplier at various voltage settings on the electrostatic analyzer. To obtain the energy spectrum as a plot of the number of ions in equal energy intervals along the energy axis, each observed counting rate was divided by the corresponding energy setting of the analyzer, and this quotient

plotted as the ordinate in the accompanying figures showing the recoil energy spectrum.

### III. RESULTS AND DISCUSSION

#### Results with a Double Evaporated Source on a Lithium Fluoride Surface

After preparing Source I it was transferred to the electrostatic analyzer and placed on the hot plate. Measurements were attempted with the source at room temperature with entrance and exit slits on the analyzer, but no measurable ion current was observed. The source was transferred back through the valve to the preparation side of the line where it was stored while the analyzer vacuum system was opened and both the entrance and exit slits removed. Upon obtaining a satisfactory vacuum the source was placed back in position under the analyzer. The set of measurements shown in the lower curve of Fig. 3 were obtained. Upon increasing the surface temperature of the source to  $85^\circ\text{C}$ , a much higher counting rate resulted. The measurements at this temperature shown in the figure were taken after the source had been at  $85^\circ\text{C}$  for about one hour, but it was noticed that the counting rate increased with time at a rate corresponding to an increase of 30 percent in an hour. The source was heated further until the surface temperature reached  $225^\circ\text{C}$  and again a large increase in counting rate was observed. The measurements shown in Fig. 3 were started at the time the source reached temperature, and at this time an increase of around 15 percent in counting rate per hour was observed. After this the source remained at  $225^\circ\text{C}$  for 14 hours and then the series of measurements shown in the upper curve of Fig. 3 were obtained. The counting rate exhibited an average increase of about 7 percent per hour during this heating period. The increase in counting rate in standing at  $225^\circ\text{C}$  for 14 hours was uniform over the whole spectrum, so that the treatment served to increase the yield of ions without effecting a change in the shape of the recoil spectrum. The end point of the recoil spectrum is about 51 eV as determined by the knee of the curve and subtracting 6 volts to allow for the resolution of the analyzer. The maximum recoil energy as observed must

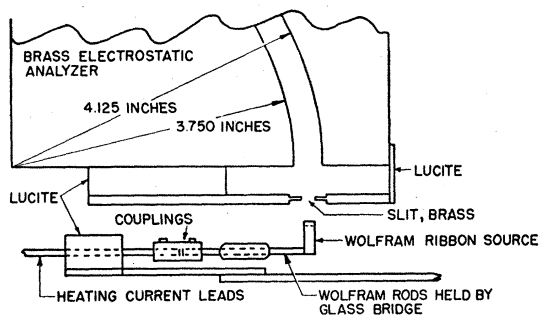


FIG. 4. Drawing showing the location of the wolfram ribbon recoil Source II under the entrance slits of the electrostatic analyzer.

be corrected for the difference in contact potential between the LiF surface and the brass analyzer and also for the binding energy of the recoiling lithium ion to the surface. Neither of these corrections could be made in the measurements on Source I. Also the shape of the recoil spectrum observed exhibited an increase in the number of ions with decreasing energy, a result that would be expected if a large fraction of the ions leaving to the surface were degraded in energy by collisions. The quality of the spectrum therefore does not allow conclusions to be drawn concerning single or multiple neutrino emission in Be<sup>7</sup>.

After making the measurements at 225°C just described, the analyzer was set for 35 electron volt ions and the heater current of the hot plate turned off. The counting rate was found to drop slowly and after forty minutes the counting rate had dropped to 87 percent of the value at 225°C. The surface temperature dropped to about 70°C in this period of time. The reduction in counting rate was probably due to the surface being covered with an adsorbed layer of gas, and this observation gives an approximate measure of the effect. This is reasonable when one notes that at a pressure of  $5 \times 10^{-7}$  mm of Hg the surface is bombarded by the residual gas molecules in the vacuum system at a rate sufficient to build up a monolayer in 15 seconds if none evaporated.

It was clear from the results that this source, although evaporated twice onto a fresh LiF surface, was of low quality, and that ions leaving the surface had to penetrate several layers of atoms. The increase in ion yield from the surface upon increasing the surface temperature indicated that the surface was covered with an adsorbed layer and to improve the results heating to even higher temperatures would be necessary. An attempt was made to heat the source to 320°C but at this temperature the LiF substrate vaporized partially, and measurements were discontinued.

It is well known that evaporated films on surfaces tend to agglomerate under certain conditions. For agglomeration of a condensed monolayer on a surface to take place, the condensed layer must be energetically more stable in the form of agglomerates, and the atoms or molecules composing the layer must have sufficient mobility to migrate over the surface and form crystalline agglomerates.<sup>14</sup> For the agglomerates to be more stable energetically the heat of evaporation of the substance from the agglomerates, which can be taken equal to the heat of evaporation of the bulk material, must be greater than the heat of evaporation of the adsorbed layer from the surface. The Be<sup>7</sup> activity evaporating from the wolfram ribbon exhibits a vapor pressure of beryllium oxide,<sup>15</sup> and lower than the vapor

<sup>14</sup> For a discussion see papers of G. I. Finch, E. T. S. Applegard, and J. E. Lennard-Jones from Report of a Conference on the Conduction of Electricity in Solids, Proc. Phys. Soc. (London) 49 (Extra Part) 113-154 (1937).

<sup>15</sup> N. D. Erway and R. L. Seifert, Manhattan District Declassified Contribution No. 1030 (1946).

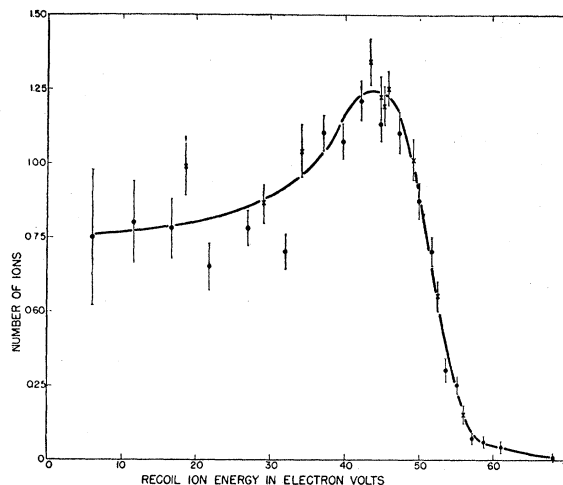


FIG. 5. Recoil spectrum from wolfram ribbon Source II at a surface temperature of 300°.

pressure of beryllium metal. Furthermore, upon considering the conditions under which the Be<sup>7</sup> was placed on the ribbon initially, and the subsequent heat treatment, one would expect that the beryllium would be in the form of the oxide. It is entirely possible that the poor recoil spectrum obtained from the LiF surface is a result of agglomeration of the Be<sup>7</sup> in the form of BeO crystals. If this conclusion is correct, a distilled source using BeO as a substrate may yield better results.

#### Results with an Evaporated Source on a Wolfram Surface

In view of the large increase in ion yield with temperature, an experiment was performed using the wolfram ribbon itself as a recoil source. The Be<sup>7</sup> activity remaining on the wolfram ribbon after vaporizing off the contaminants may be spread on the surface and be clean enough to serve as a recoil source. Further, a more vigorous thermal treatment could be given to clean the surface of absorbed gases by passing a current through the ribbon sufficient to heat it to 1000°C. With these possibilities in mind recoil Source II was prepared as described above.

Recoil Source II was installed under the entrance slits of the analyzer as shown in Fig. 4. After obtaining a satisfactory vacuum, a current was passed through the ribbon sufficient to heat it to 1000°C for 15 seconds to outgas its surface. Previously the ribbon had been heated to 1560°C, and by only heating to 1000°C there was no danger of distilling Be<sup>7</sup> from the surface and contaminating the analyzer and electron multiplier. The recoil spectrum was then measured while the ribbon was at room temperature with the result that only a very low counting rate was observed over the entire spectrum. The temperature of the ribbon was increased to about 300° and after a period of three and one-half hours the measurements shown in Fig. 5 were

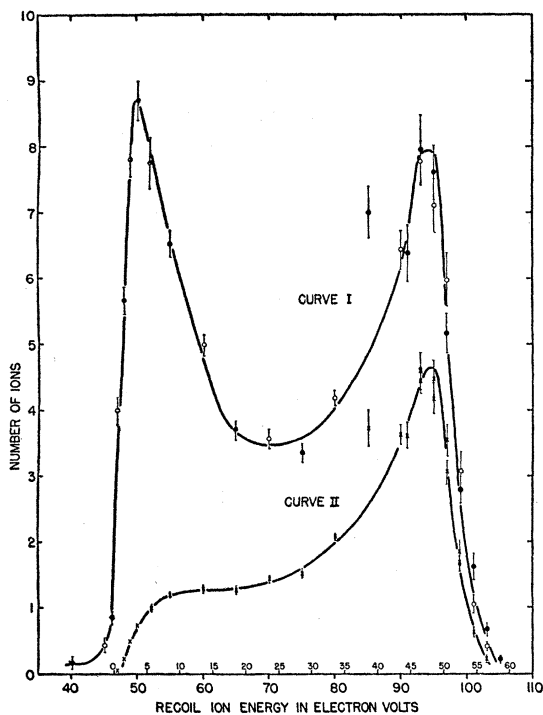


FIG. 6. Recoil spectrum observed from Source II with 46.4 volts between the source and the electrostatic analyzer. Curve I shows the experimentally observed spectrum, and Curve II represents the spectrum corrected for discrimination effects.

obtained. The spectrum shows a peak around 45 eV recoil energy. Comparison with the spectrum from the double evaporated source on lithium fluoride at 225°C surface temperature shows clearly that a relatively larger number of high energy ions was obtained from the wolfram surface. The counting errors are much greater at low energies and are indicated by the bars. The series of points indicated by the plane bars were taken in sequence from low to high energy, then following these the points marked with crosses were taken randomly over the spectra. A slight increase in counting rate with time was noticeable.

The temperature of the ribbon cannot be increased very much above 300° because thermal ions are given off in abundance at around 550°. However, a more vigorous thermal treatment of the surface could be achieved by flashing the ribbon for about 15 seconds at 1000°, then reducing the temperature of the ribbon to around 300° for counting. Experiments of this sort were conducted and it was found that the yield of ions increased by a factor of about three. The recoil spectra exhibited a sharper peak and the relative number of low energy ions was greatly reduced. The recoil spectrum shown in the upper curve of Fig. 7 illustrates these improvements. This spectrum was taken by flashing the ribbon at around 1000° for fifteen seconds and the recoil ions counted for a three minute period at 300° starting five seconds after the current was

reduced. The counting rate was observed as a function of the time, and it was found to remain constant during the three minute period within the counting statistics.

Measurements were made in which the recoil source was placed at a positive potential with respect to the grounded entrance slit of the analyzer. This accelerating potential gives each ion entering the analyzer an energy of  $V$  electron volts in addition to its initial recoil energy. In this way the entire recoil spectrum is shifted along the energy axis by  $V$  electron volts. The spectrum shown in the upper curve of Fig. 6 was obtained with 46.4 volts accelerating potential. This procedure had two advantages. First, by shifting the entire spectrum to higher energies the counting efficiencies for all ions entering the analyzer were made more nearly comparable. This follows from the fact that the aperture of the analyzer increases linearly with the energy. When potential between the source and the analyzer was zero, 50-volt ions were counted with fifty times greater efficiency than one-volt ions, and therefore, large errors were introduced in the measurements at the low energy side of the spectrum (Figs. 3, 5, and 7). But, by applying a potential comparable in magnitude to the maximum recoil energy, the aperture of the analyzer only changed by a factor of about 2 over the spectrum. There was, however, a discrimination effect in favor of low energy ions, which will be discussed later. The second advantage of the applied potential was that the maximum energy of the recoiling ions could be obtained free of a contact potential error.

The recoil spectrum shown in Fig. 6 was obtained following the above-mentioned flashing technique, and the counting rate observed as a function of the time for a three minute period. For each point a plot of counting rate *versus* time was made and the value taken by extrapolating to zero time. However, it was found that the counting rate did not change appreciably during the three minute period. The results presented represent two separate sweeps over the spectrum as indicated by the solid and open circles. The spectrum exhibits two distinct peaks, one at the low energy end that is very steep sided, and one at the high energy end which is similar in shape to the spectrum obtained without an accelerating potential. To obtain the maximum recoil energy of the ions from this spectrum, the following procedure was used. The low and high energy sides of the spectrum were extrapolated to the energy axis. The energies obtained were then corrected for the resolution of the analyzer by adding to the extrapolated energy at the low energy side the resolution half-width and subtracting the corresponding resolution half-width from the extrapolated value at the high energy side. The difference in these two corrected energies gave a maximum recoil energy of  $55.9 \pm 1.0$  eV. The largest error was in determining the end point at the high energy side. This value agrees with the results of Smith and Allen.<sup>4</sup> They observed a maximum recoil energy of

56.6±1.0 ev from an evaporated source of Be<sup>7</sup> on a tantalum substrate.

The strong peak at the low energy side of the spectrum in Fig. 6 results from a discrimination effect in favor of low energy ions. An ion leaving the surface with an energy  $E$  and direction  $\theta$  with respect to the normal was accelerated by the applied field in the direction normal to the surface. Upon passing through the entrance slit the ion had an energy equal to the sum of the initial energy and the applied potential,  $E+V$ , but also had its direction of motion changed to a

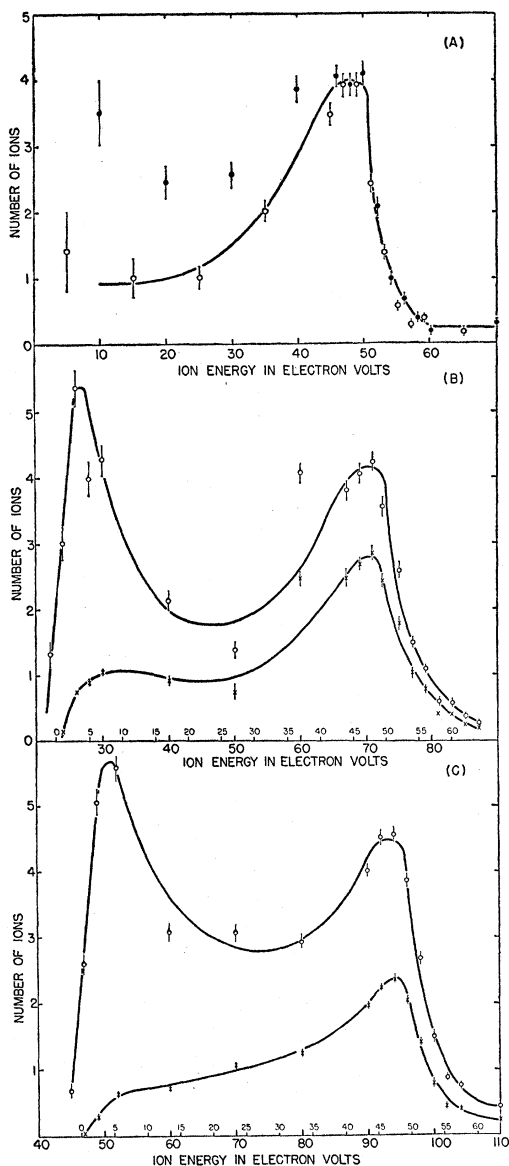


FIG. 7. A series of three recoil spectra observed successively at several accelerating potentials between the source and the entrance slit. These were at zero (Curve A), 23 (Curve B), and 46 (Curve C) volts. The recoil curves B and C were corrected for discrimination correction using the factor  $g$  plotted in Curves B and C of Fig. 9. The corrected spectra are shown below the observed spectra in each case.

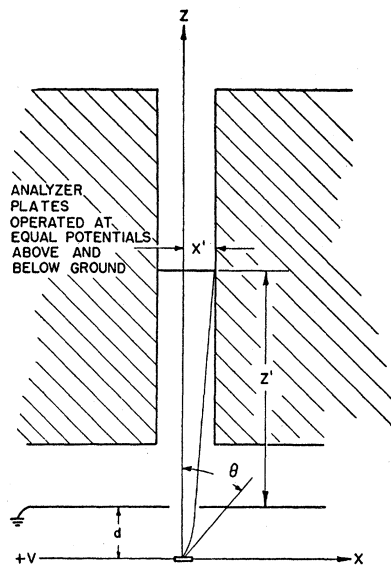


FIG. 8. The schematic representation of the relation between the source, slit, and analyzer used in calculating the discrimination effect.

smaller angle with respect to the normal. The change in direction of the ions was small for high energy ions and large for low energy ions, and thus gives rise to a greater relative collection of efficiency for low energy ions. An estimate of this effect was calculated in the following manner. The ion was considered to move in a uniform electric field between the wolfram ribbon and the entrance slit. After passing through the slit the ion travels a short distance and enters the radial field of the analyzer. For the purpose of making the geometrical efficiency correction the exact orbit of the ions through the analyzer was not used, but was regarded as a pure geometrical baffle having an acceptance angle as defined by half the distance between the plates at a distance halfway through the analyzer. The arrangement used for these calculations is shown in Fig. 8. The motion of the ion of initial energy  $E$  and initial direction  $\theta$  between the source and the slit is given by the expression

$$z = \frac{x}{\tan\theta} + \frac{Vx^2}{4dE \sin^2\theta} \tag{5}$$

After leaving the slit the ion was assumed to travel in a straight line, and after moving a distance  $z'$  from the slit, it will be displaced a distance  $x'$  in the X-direction.

$$x' = \frac{2dE}{V} \sin\theta \left[ -\cos\theta + \left( \cos^2\theta + \frac{V}{E} \right)^{\frac{1}{2}} \right] + \frac{z' \sin\theta}{\left( \cos^2\theta + \frac{V}{E} \right)^{\frac{1}{2}}} \tag{6}$$



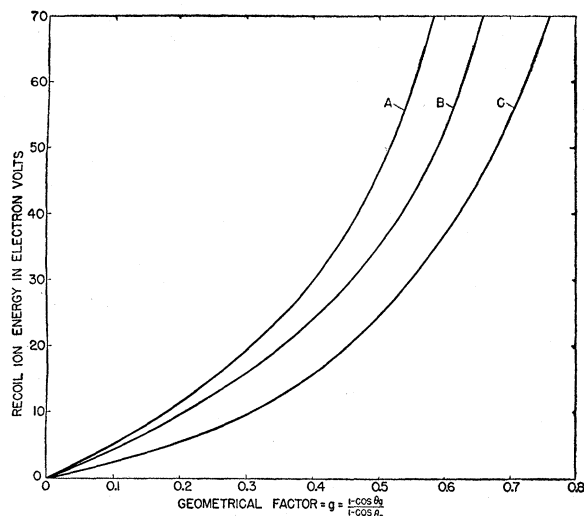


FIG. 9. Plot of the geometrical factor  $g$  for various ion energies calculated from Eqs. (6) and (7). Curve  $A$  is calculated for 46.4 volts between the source and the analyzer using the geometrical acceptance angle of the analyzer, three degrees. Curves  $B$  and  $C$  are for 46.4 and 23.3 volts, and using one-sixth the geometrical acceptance angle of the analyzer, around 30 minutes.

Setting the distance as one-half the spacing between the plates of the analyzer,  $z'$  the distance from the slit to the center of the analyzer, and the values for  $d$ , and

$$\text{Efficiency factor} = \frac{\text{volume of cone defined by critical angle}}{\text{volume of cone defined by geometrical acceptance angle}} = \frac{1 - \cos \theta_c}{g(1 - \cos \theta_a)} \quad (7)$$

Each measured value was multiplied by the factor  $g$  which corrected the spectrum for the variation of collection efficiency of the analyzer with initial ion energy. The results of applying the discrimination correction to the recoil spectrum are shown in the lower curve of Fig. 6. A plot of  $g$  versus  $E$  is shown in Fig. 9. Since the estimation of the defining geometry of the analyzer was only approximate, a calculation of the factor  $g$  was repeated using one-sixth the value of  $z'$  used in the first calculation and the results of this calculation are shown in Fig. 9.

To check the above described discrimination correction it was applied to recoil spectra measured successively at two different values of the accelerating potential, 23 and 46 volts, and also with the source grounded. The measurements were taken in sequence having the source at zero potential, then at 23 volts and finally at 46 volts. After this the spectrum at zero potential was repeated, the measurements being shown by the solid circles in Fig. 7(a). These points were higher at the low energy and this was attributed to a change in the source during the measurements which extended over a period of four and one-half hours. The observed spectra and the spectra corrected for discrimination are shown in Fig. 7. Comparison of the three

spectra shows that the discrimination correction reduces the observed spectra at 23 and 46 volts, respectively, to a spectrum that compares favorably with the spectra observed at zero accelerating potential. It can be noticed that the correction is perhaps too severe at the high energy end of the spectrum.

$V$ , a calculation was made of the angles  $\theta_c$  and the corresponding energies  $E$ . One has then as a result of the calculation the maximum angle from the normal that an ion of energy  $E$  leaving the surface, can have in order to enter the analyzer and be counted. The acceptance angle for the analyzer in the plane parallel to the plates is determined by the width of the exit slit and the total distance through the analyzer. For the analyzer used in these experiments the acceptance angle is almost identical in the radial and axial directions, and the critical angles  $\theta_c$  calculated by the above formula may be used as defining a cone at the surface within which ions of a specified energy are counted. It is clear that the cone will be much larger for low energy ions than for the high energy ions, and the calculation shows that cones for 2 and 42-volt ions have critical angles  $\theta_c$  of  $18^\circ$  and  $4^\circ$ , respectively, when a 46-volt potential is applied between the source and the slit. If it is assumed that the initial distribution of recoil ions from the surface is random in direction, then the collection efficiency of the analyzer for recoil ions of initial energy  $E$  is equal to the ratio of the surface area of the corresponding unit cone to the unit cone corresponding to the geometrical acceptance angle of the analyzer  $\theta_a$  ( $3^\circ$ ). The collection efficiency so defined is then calculated from the angles  $\theta_c$  and  $\theta_a$ .

#### IV. CONCLUSIONS

The recoil spectrum obtained from a wolfram surface under vigorous thermal treatment exhibits a peak at the high energy end of the recoil spectrum. The spectrum shown in Fig. 6 was taken under these conditions and represents the best series of measurements. The discrimination correction discussed above accounts for the presence of the sharp low energy peak, and the corrected recoil spectrum shown represents the true recoil spectrum from the surface. The high energy peak was located at 48 electron volts recoil energy, about 8 volts below the end point, and has a half-width in the neighborhood of 10 electron volts. The number of recoil ions leaving the surface with the full recoil energy was vanishingly small. The appearance of the peak at a lower energy indicates that the majority of the recoiling ions leaving the surface lose energy through

collisions with neighboring atoms. However, the quality of the recoil spectrum was good enough to allow one to draw conclusions concerning single or multiple neutrino emission in Be<sup>7</sup>. Multiple neutrino emission should lead to a bell-shaped recoil distribution spectrum, and if surface collision effects were present in the experimental curve it would be distorted toward larger numbers of low energy ions. Since the observed spectrum exhibits a peak near the high energy end of the spectrum one can conclude that Be<sup>7</sup> electron capture decays are accompanied by the emission of a single neutrino.

The observed end point of the spectrum  $55.9 \pm 1.0$  ev, should be compared to the initial recoil energy of the nucleus of  $57.3 \pm 0.5$  ev minus the binding energy of the lithium ion to the surface. The binding energy of lithium ions to the surface could not be measured experimentally, nor be evaluated theoretically because of the uncertainties in the knowledge of the nature of the surface from which the ion recoiled. The main reason for desiring an accurate value for the end point of the recoil spectrum and the surface binding energy is to obtain the mass of the neutrino directly from a measure of its recoil momentum. The recoil energy  $E_R$  for an ion of mass  $M$  resulting from the emission of a

single neutrino of rest mass  $m_\nu$  is given by

$$E_R = (E^2 - m_\nu^2 c^4) / 2Mc^2, \quad (8)$$

where  $E$  is the total energy available for the process ( $0.864 \pm 0.003$  Mev). Upon taking the observed end point of  $55.9 \pm 1.0$  ev as  $E_R$  one obtains from this experiment an upper limit for the mass of the neutrino of  $0.27 \pm 0.14$  electron masses. The limit obtained from this experiment is much higher than the upper limit of 0.001 electron masses set by observations of the shape of the end point of the He<sup>3</sup> beta-spectrum,<sup>16</sup> and the limit of  $(-0.04 \pm 0.05)$  set by nuclear reaction data.<sup>17</sup>

The author would like to express his appreciation to Dr. Irving Wolff and Dr. R. P. Stone of the RCA laboratories for the loan of the electron multiplier tube used in these experiments, and to Dr. Oliver A. Schaeffer and Dr. James A. Amick for their helpful discussions during the course of the experiments. The author would like further to thank Professor O. Kofoed-Hansen for reading the manuscript.

<sup>16</sup> Hamilton, Alford, and Gross, *Phys. Rev.* **83**, 215 (1951); H. A. Tolhoek, *Ned. T. Natuurkde* **15**, 96 (1949); Curran, Angus, and Cockcroft, *Phil. Mag.* **40**, 53 (1949); and O. Kofoed-Hansen, *Phil. Mag.* **42**, 1448 (1951).

<sup>17</sup> Shoupp, Jennings, and Sun, *Phys. Rev.* **75**, 1 (1949).

## On the Stress Tensor of the Electron\*

SIDNEY BOROWITZ† AND WALTER KOHN‡

*Department of Physics, Harvard University, Cambridge, Massachusetts*

(Received July 25, 1951)

A suggestion by Schwinger for dealing with problems of gauge invariance has been adapted for treating the radiative corrections to the stress tensor of an electron. The divergencelessness of the stress tensor implies that a certain divergent integral be invariant with respect to translation. Application of this rule gives a zero self-stress and shows that the matrix elements of the symmetric tensor between states of unequal momenta are finite. The canonical tensor however remains infinite. The connection with the results of Rohrlich, and Villars, and Pais and Epstein, is discussed.

### INTRODUCTION

RECENT developments in quantum electrodynamics<sup>1</sup> have shown that the difficulties connected with the infinite self-energy of the electron and the infinite charge induced in the electron vacuum by an external field can be circumvented by appropriate covariant renormalizations. It was of interest to investi-

gate whether these techniques were sufficient to give unambiguous and self-consistent answers to other questions in quantum electrodynamics.

The calculation of the self-stress of an electron by Pais and Epstein<sup>2</sup> seemed to show that this was not the case. These authors found that after carrying out the renormalization procedure, the self-stress emerged as a finite quantity, whereas Lorentz covariance demands that it must vanish. It was clear that this difficulty had its origin in the infinite integrals which occur in the expression for the stress-tensor and whose evaluation may lead to inconsistent results.

In the present paper the basic idea underlying the calculation was to investigate carefully the divergent integrals that occur and to use physical principles in

\* A preliminary report was given at the January, 1950 meeting of the Am. Phys. Soc., *Phys. Rev.* **78**, 345 (1950).

† Present address—Department of Physics, New York University, University Heights, New York 53, New York.

‡ Present address—Institute for Theoretical Physics, Copenhagen, Denmark; on leave of absence from Department of Physics, Carnegie Institute of Technology, Schenley Park, Pittsburgh, Pennsylvania.

<sup>1</sup> S. Tomonaga, *Prog. Theor. Phys.* **1**, 27 (1946); J. Schwinger, *Phys. Rev.* **74**, 1439 (1948); **75**, 651 (1949); **76**, 790 (1949). The last three papers will be referred to as SI, SII, and SIII, respectively.

<sup>2</sup> A. Pais and S. T. Epstein, *Revs. Modern Phys.* **21**, 445 (1949).