

determined by the magnetic spectrograph measurements are sufficiently conclusive to establish these genetic relationships.

#### V. Hf<sup>175</sup> DECAY SCHEME

The data given in Table III establish most of the features of the decay scheme shown in Fig. 5(b). From the relative transition intensity data given in Table III, it appears that little, if any, orbital electron capture takes place to the 113.6-keV level. To determine the  $K$ -capture ratios to the 342.9- and 432.2-keV levels a coincidence experiment was performed with the scintillation apparatus. With the "gate" channel set to cover the 342-keV photopeak, the coincidence spectrum of the 89-keV gamma ray and lutecium  $K$  x-rays was examined. The  $K$  x-rays are engendered partly by  $K$ -capture transitions to the 342.9-keV and 432.2-keV levels and partly by  $K$ -conversion of the 89-keV transition. From the measurement of the relative intensities of the 89-keV gamma and lutecium  $K$  x-ray and by utilizing the  $K$ -fluorescence yield,<sup>14</sup> the estimated  $L$ - to  $K$ -capture ratio of 17%,<sup>20</sup> and the estimated  $K$ -conversion coefficient and  $K/L$  ratio for the 89.3-keV transition,<sup>13,15</sup>  $K$ -capture branching ratios of ~80% to the 342.9-keV level and ~20% to the 432.2-keV level were obtained. Further studies of the "ungated" gamma-ray spectrum with the calibrated 2 inch×2 inch NaI(Tl) crystal indicated from the lutecium  $K$  x-ray intensity relative to that of the 342-keV gamma intensity that little, if any (<10%), orbital electron capture takes place to the ground state of Lu<sup>175</sup>.

By observing [Fig. 5(b)] that the 342.9-keV level decays by  $M1+E2$  radiation to the  $7/2+$  ground state, a spin and parity assignment of  $5/2+$ ,  $7/2+$ , or  $9/2+$

<sup>20</sup> M. E. Rose and J. L. Jackson, Phys. Rev. 76, 1540 (1949).

can be made for the 342.9-keV level. A  $5/2+$ -assignment for the 342.9-keV level would seem to be the most probable in view of the absence of observable  $K$ -capture to the  $7/2+$  ground state and to the  $9/2+$  excited state at 113.6 keV. From the  $M1+E2$  character of the 89.3-keV transition, a tentative assignment of  $5/2+$  is made for the 432.2-keV level. Perhaps one of the foregoing  $5/2+$  levels is to be identified with a  $d_{5/2}$  level available from shell theory.<sup>16</sup> It is to be noted that the remaining  $5/2+$  level excited by Hf<sup>175</sup> decay and the  $9/2-$  level excited by Yb<sup>175</sup> decay are not readily explainable on the basis of the shell model.

By utilizing the beta-energy systematics of Way and Wood<sup>21</sup> (which give a disintegration energy of ~900 keV) and the above  $K$ -capture branching ratios,  $\log ft$  values of 7.8 and 7.4 are determined for the 20% and 80%  $K$ -capture branches, respectively. This suggests that a spin change of one unit and a parity change are realized in both  $K$ -capture transitions. Since there is no observable  $K$ -capture to the  $7/2+$  Lu<sup>175</sup> ground state ( $\log ft > 8.8$ ), a spin and parity assignment of  $3/2-$  is therefore made for the ground state of Hf<sup>175</sup>. This ground-state assignment is probably to be identified with a  $p_{3/2}$  configuration which is available from shell theory.<sup>16</sup>

#### VI. ACKNOWLEDGMENTS

The authors wish to thank Mr. H. Maltrud for his assistance in the Yb<sup>175</sup> beta-decay measurements.

*Note added in proof.*—A detailed discussion of the nuclear levels and transitions in Lu<sup>175</sup>, according to the unified model, by L. Willets and D. M. Chase of the Los Alamos Scientific Laboratory, has been submitted for publication [Phys. Rev. (to be published)].

<sup>21</sup> K. Way and M. Wood, Phys. Rev. 94, 119 (1953).

## Spectrometry of the Neutrino Recoils of Argon-37

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The ions that recoil from neutrino emission in the electron capture decay of A<sup>37</sup> have been subjected to magnetic spectrometry with a resolution amounting to 2.8% in energy. They are found to have an energy of  $9.63 \pm 0.06$  eV, which is in agreement with the value  $9.65 \pm 0.05$  eV to be expected on the basis of a two-body breakup, the neutrino having an energy of  $815 \pm 2$  keV as determined by others from the threshold of the  $C^{13}(p,n)A^{37}$  reaction. The momentum balance sets an upper limit of about 5 keV for the rest mass of the neutrino. Auger electron emission following the orbital electron capture leaves the recoils mostly multiply charged, the percentage abundances in charge states 1 to 7 being  $6.2 \pm 0.1$ ,  $15.7 \pm 0.4$ ,  $39.2 \pm 0.5$ ,  $26.7 \pm 0.4$ ,  $10.0 \pm 0.2$ ,  $1.8 \pm 0.1$  and  $0.4 \pm 0.1$ , respectively. Neutrals were not measured. The natural width at half-intensity of the singly-charged recoil line is 1.7 eV, which is fully accounted for by the thermal motion of the argon atoms. The natural width of the triply-charged recoil line is 2.5 eV, which is mostly but not entirely accounted for by thermal motion plus recoil from the emission of one 2300-eV  $K$  Auger electron. The singly-charged recoils are thought to result from  $L$ -capture and from  $K$ -capture followed by  $K$  x-ray emission, neither of these processes involving recoil from the 2300-eV Auger electron.

ARGON-37 is an attractive substance for neutrino recoil experimentation because it possesses a number of simultaneously favorable properties. It is an

electron-capturing radionuclide with a half-life of 34 days, and its decay is unaccompanied by gamma emission, so the predominant mechanism giving recoil should

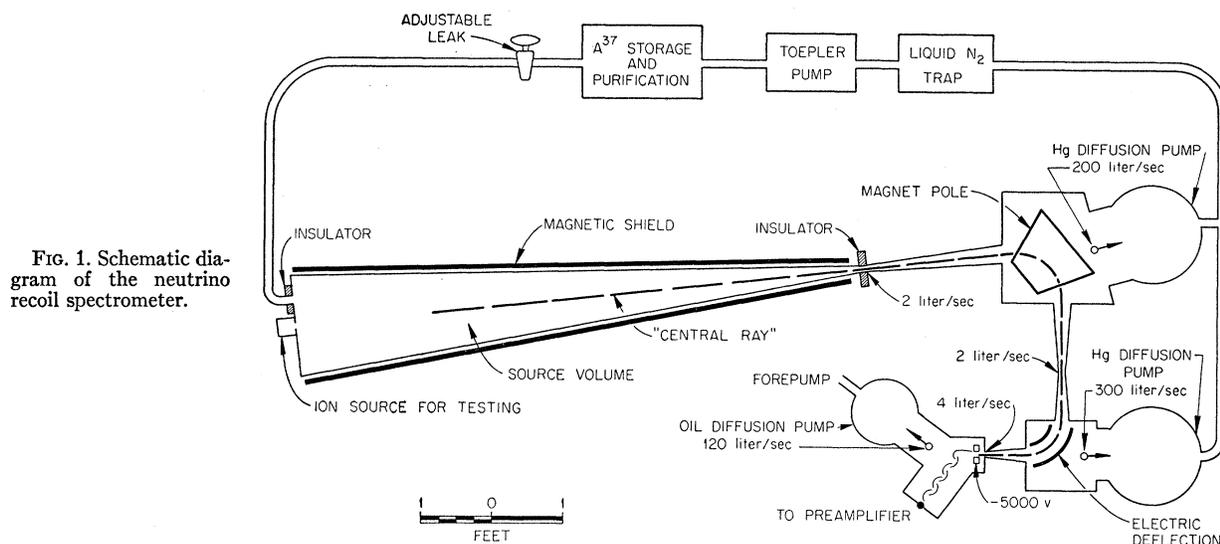


FIG. 1. Schematic diagram of the neutrino recoil spectrometer.

be the neutrino emission. It is a monatomic gas, so that surface effects and disturbing molecular effects are absent. It can in principle be prepared carrier free, it is chemically easy to extract and purify, and it does not contaminate the measuring system. Its main disadvantage lies in the low energy of the recoiling  $Cl^{37}$  atoms; this is predicted from the radioactive transition energy of  $815 \pm 2$  kev<sup>1</sup> to be  $9.65 \pm 0.05$  ev, assuming a two-body breakup with zero rest mass for the neutrino. This disadvantage has not prevented Rodeback and Allen<sup>2</sup> from observing the recoil velocity in a coincidence time-of-flight experiment, nor did it prevent Kofoed-Hansen<sup>3</sup> from studying the decay in greater detail in a recent integral experiment employing an ingenious crossed-field spectrometer. The work herein to be described paralleled Kofoed-Hansen's in objectives and to a large extent also in time; the experimental approach, however, has been considerably different.

At the outset one expects the monoenergetic recoils to have a distribution in charge arising from Auger processes accompanying the filling of the inner shell vacancy left by the orbital electron capture. One also expects a considerable natural width to the recoil lines, because the velocity of the argon atoms in their thermal motion amounts on the average to about 5% of the expected recoil velocity. That these expectations are realized has been shown rather qualitatively in an earlier communication of ours,<sup>4</sup> but since that time our technique has been greatly improved, and we are now able to present a much more detailed and precise experi-

mental study of the  $A^{37}$  recoils, using the methods of high resolution magnetic spectrometry.

#### EXPERIMENTAL

The recoil spectrometer is diagrammed in Fig. 1. A conical "source volume" contained the radioactive gas under a total pressure of  $2 \times 10^{-5}$  mm of mercury or less; this stainless steel container was 300 cm long and its interior was presumed to be free of electric and magnetic fields. A small fraction (about  $10^{-6}$ ) of the atoms recoiling from radioactive decay in this volume emerged from a hole  $\frac{1}{2}$  inch in diameter at the small end, and the resulting beam was subjected to analysis by a 96.5-degree deflection in a wedge-shaped, stigmatic-focusing magnetic field. The radius of curvature in the field was about 16.75 cm. The ions were then deflected and stigmatically focused a second time in an electrostatic deflector having spherical sector plates spaced  $\pm 3$  cm about a mean deflection radius of 20 cm. Finally, the ions were accelerated through 4600 volts for counting by means of a secondary electron multiplier of the design developed for particle counting by Allen,<sup>5</sup> but employing Ag-Mg plates.

No windows or foils could be used between source volume and detector, so the only means of forestalling an overwhelming background counting rate from gas flowing to the vicinity of the detector was to apply strong differential pumping, and to handle the gas on a continuous flow-through basis. The pumping requirements are stringent. The vacuum system must be narrowed at the nodes of the ion bundle, and the importance of two-directional focusing by the deflecting devices becomes compelling. The differential pumping can be traced out in Fig. 1, where measured pumping speeds are indicated. To conserve the gas for reuse, the mechanical forepumps were replaced by a Toepler pump which compressed the gas into a storage volume, where it could be purified by hot calcium or zirconium. The recovery system was use-

<sup>1</sup> Richards, Smith, Browne, Phys. Rev. **80**, 524 (1950) give  $816 \pm 4$  kev, and Schoenfeld, Duborg, Preston, and Goodman, Phys. Rev. **85**, 873 (1952) give  $816 \pm 2$  kev, both from  $Cl^{37}(p,n)A^{37}$  threshold measurements. Emmerich, Singer, and Kurbatov, Phys. Rev. **94**, 113 (1954) give  $815 \pm 20$  kev from the upper limit of the  $A^{37}$  internal bremsstrahlung spectrum. We here adopt the value of Schoenfeld *et al.* because it has the smaller quoted error, but we reduce their value by 1 kev because of a subsequent recalibration of the  $Li^{7}(p,n)Be^{7}$  threshold, which had been taken by them as a standard for their voltage scale. We are indebted to Dr. C. H. Johnson for informing us of the necessity for this step.

<sup>2</sup> G. W. Rodeback and J. S. Allen, Phys. Rev. **86**, 446 (1952).

<sup>3</sup> O. Kofoed-Hansen, Phys. Rev. **96**, 1045 (1954).

<sup>4</sup> A. H. Snell and F. Pleasonton, Phys. Rev. **97**, 246 (1955).

<sup>5</sup> J. S. Allen, Rev. Sci. Instr. **18**, 739 (1947).

ful, but best results were always obtained from a fresh batch of gas.

The argon-37 was prepared by the  $\text{Ca}^{40}(n,\alpha)\text{A}^{37}$  reaction, calcium oxide being irradiated in 150-gram batches in the Low Intensity Testing Reactor. This yielded samples several curies in strength,<sup>6</sup> and we estimate that 2 to 10 millicuries were typically present in the source volume when readings were being taken. Monitoring the source strength was a nuisance because there was no external signal; the best we could do was to follow its depletion by referring back to a standard position in the spectrum at intervals during the course of each run.

The source volume was insulated from ground, making it possible to apply predeflection acceleration to the ions before they entered the analyzer. We thus introduced a useful degree of flexibility in the effective resolution. When a positive bias of several hundred volts is applied to the source volume, the widths of the lines presented to the spectrometer become proportionately small, and are exceeded by the transmission width of the spectrometer, so the resolution is effectively low. At the other extreme, when only a few volts' bias is applied, the transmission width is smaller than the natural width, so the spectrometer acts with high resolution, and can examine the line shapes. The exploitation of this feature will become apparent as our discussion proceeds.

The use of both magnetic and electric deflection may seem puzzling because clearly both were not required for the energy analysis. The combination did however give complete  $e/m$  identification of the ions that were counted, and it will be of further utility for future experiments—particularly those involving continuous recoil spectra. Actually, the electric deflector served an indispensable function by merely supplying an enlargement of the system to which a stage of differential pumping could be applied. The magnet was necessary to resolve the charge spectrum.<sup>7</sup> We here put main emphasis upon the magnet as the analyzer, treating the electric deflector as a subsidiary that was always adjusted to transmit ions that were passed by the magnet. Actually, the method of taking readings was to match the electric deflector to the magnet for a given  $e/m$  value, and then to hold both deflecting fields fixed while sweeping over the line in question by slightly changing the voltage on the source volume. Thus our results will be presented as *energy* spectra obtained at fixed  $B\rho$  settings of the magnet, the abscissa scales being in terms of source volume voltage. Expressed in corresponding terms, the transmission width of the magnet was 2.8% *in energy* when half-inch object and image apertures were used. One reason for using this procedure was to save time in taking readings (the source is running out all the time!) for only one adjustment then had to be made between counts. As a consequence of the use of

fixed deflecting fields, the energy interval accepted by the spectrometer was constant, so the counting rates did not require division by the energy or momentum value to obtain the number of counts per unit energy or momentum interval.

The equation governing the magnetic deflection is

$$B\rho = \{2M(E_r + zeV_s)\}^{1/2}/ze, \quad (1)$$

where  $E_r$  is the recoil energy,  $V_s$  is the source voltage, and  $M$  and  $ze$  are respectively the ionic mass and charge. With  $B\rho$  in gauss-cm,  $E_r$  in ev, and  $V_s$  in volts, this becomes

$$B\rho = 143.94\{M(E_r + zV_s)\}^{1/2}/z, \quad (2)$$

where the atomic constants concerned have been taken from the values of Dumond and Cohen,<sup>8</sup> and the value  $M=36.977$  amu is pertinent to the present work. In the most accurate determination of the recoil energy, a proton nuclear resonance fluxmeter was used to determine  $B$ , and the calibration of the magnetic spectrometer consisted of the determination of the effective radius of curvature  $\rho$ , which would take into account deflection in the fringing fields and perhaps a small effect arising from the fact that the magnetometer probe was placed aside from the position of the ion paths. The form of Eq. (2) requires that the calibration be very carefully done, for with  $zV_s=75$  (a typical value),  $\rho$  must be accurate to 0.1% for 1% precision in  $E_r$ . Calibration was accomplished in two ways. At field strengths higher than those used for the recoil energy determination, positive ions produced in the ion source on the end of the source volume were deflected at accurately known voltages ( $\text{N}_2^+$  and  $\text{H}_2\text{O}^+$  at about 500 and  $\text{H}_1^+$ ,  $\text{H}_2^+$ , and  $\text{H}_3^+$  at about 1000 volts), care being taken to keep the voltage across the ion source itself low (10 volts) compared with the potential drop encountered on entering the magnet deflection chamber, so that uncertainties arising from space charge and point-of-origin effects in the ion source would be small compared with the total energy. In this calibration the ultimate standard was a standard cell with voltage measurement by means of a type  $K$  potentiometer and a string of intercalibrated wire-wound resistors. The value of  $\rho$  thus obtained was  $16.747 \pm 0.01$  cm. The second method of calibration was used at field strength-lower than those used in the recoil energy determination, and made use of the internal conversion lines of  $\text{Bi}^{207}$  and  $\text{Cs}^{137}$ . For this purpose the field had to be reversed, and detection was by means of a scintillator. For the  $B\rho$  value of the  $K$ -conversion line of  $\text{Bi}^{207}$  we took  $4656.8 \pm 1.1$  gauss-cm, this being derived from the mean of the gamma-ray measurements  $1063.9 \pm 0.3$  kev by Alburger<sup>9</sup> and  $1063.4 \pm 0.5$  kev by Yavin and Schmidt.<sup>10</sup> The  $\text{Bi}^{207}$  calibration yielded  $\rho=16.741 \pm 0.006$  cm. For the  $\text{Cs}^{137}$   $K$ -conversion line we adopted a  $B\rho$  value of  $3381.2 \pm 0.6$  gauss-cm, derived from the gamma-ray measurement  $661.60 \pm 0.14$  kev by Muller

<sup>6</sup> We are indebted to R. E. McHenry of Operations Division, Oak Ridge National Laboratory, for the high quality of the chemical separation and purification.

<sup>7</sup> The curves of reference 4 illustrate the incomplete charge resolution given by the electric deflector alone.

<sup>8</sup> J. W. M. Dumond and E. R. Cohen, *Revs. Modern Phys.* **25**, 691 (1953).

<sup>9</sup> D. Alburger, *Phys. Rev.* **92**, 1257 (1953).

<sup>10</sup> A. I. Yavin and F. H. Schmidt, *Phys. Rev.* **100**, 171 (1955).

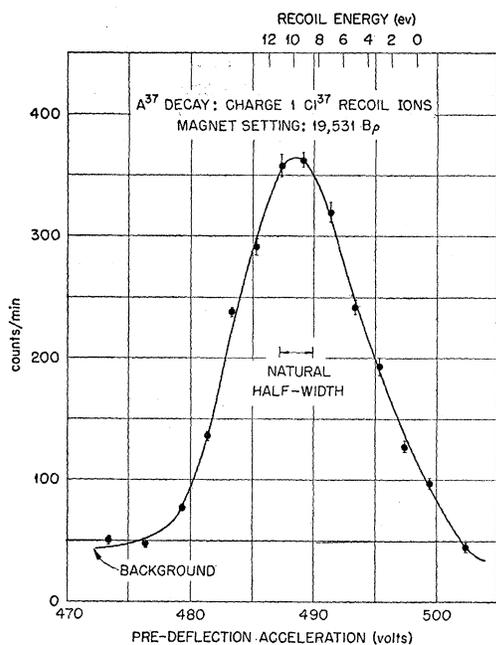


FIG. 2. The singly-charged recoil peak as observed with 490 volts positive bias applied to the source volume. The natural width is comparatively small, as indicated, and the curve traces the instrumental transmission function. The scale at the top indicates that the neutrino recoil energy is easily observable even at this high source voltage. Background has not been subtracted from this curve nor from Fig. 3, because it is instructive to show the signal-to-background ratio inasmuch as this is a measure of the adequacy of the differential pumping. The magnet was set at 19 531  $B\rho$ .

and others,<sup>11</sup> and obtained  $\rho = 16.746 \pm 0.007$  cm. As the mean of these three determinations we adopted finally  $\rho = 16.745 \pm 0.004$  cm, and used this value for the evaluation of the recoil energy.

## EXPERIMENTAL RESULTS AND DISCUSSION

### I. Charge Spectrum of the Recoils

With a potential bias of about +500 volts applied to the source volume, the transmission width of the analyzer encompasses the entire recoil line, and under these circumstances it is sufficient in measuring the relative intensities of the different charge states to take counting rate measurements at single settings upon the respective peaks. The lines could, however, be traced over at will, as is shown in Figs. 2 and 3 where the rather weak charge 1 and charge 5 peaks are shown. These plots of course trace out only the transmission curve of the analyzer, but they show how completely the lines are swallowed by the spectrometer. As will be observed from the scales at the top, the neutrino recoil energy was quite evident even when this rather high voltage was applied to the source volume.

It is unsafe to assume that the multiplier responds equally efficiently to the ions of varying charge, so it was necessary to evaluate corrections by the use of conventional integral bias curves. The curve for singly-

<sup>11</sup> Muller, Hoyt, and Klein, and DuMond, Phys. Rev. 88, 775 (1952).

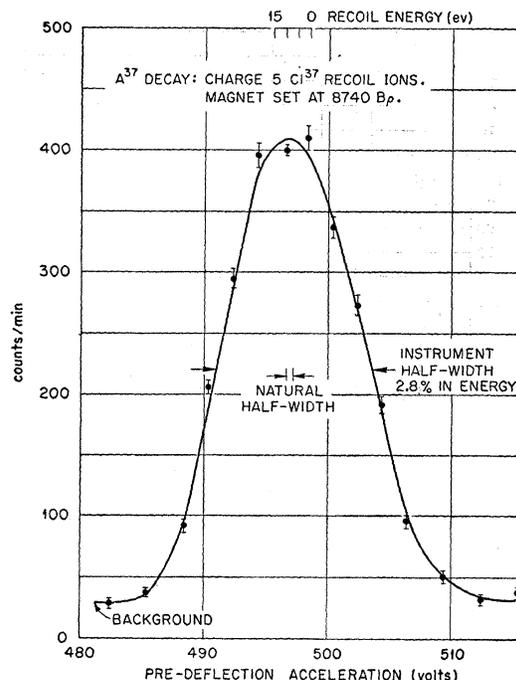


FIG. 3. The quintuply-charged recoil ions observed with 490 volts positive bias applied to the source volume. The curve is comparable with the singly charged ion peak shown in Fig. 2. Both figures show that at this source voltage, the lines are entirely accepted within the transmission width of the spectrometer. The magnet was set at 8740  $B\rho$ .

charged ions was accurately obtained by introducing carbon tetrachloride vapor into the ion source, but for the higher charges we used the  $A^{37}$  peaks themselves. Consequently the curves for the higher charges are rather rough (Fig. 4), but they do seem to indicate that ions of increasing charge produce pulses of slightly increasing average size. The upward corrections, read from these curves at the indicated operating point and applied to the observed counting rates, were 8.5% for charge 1 ions, 6.5% for charge 2, 3% for charge 3, 1% for charge 4, and no correction was assumed necessary for charges 5, 6, and 7.

The results of the relative intensity measurements are displayed in Fig. 5, and they are given in detail in the second column of Table I. The errors indicated are a combination of statistical uncertainty and uncertainty in the multiplier sensitivity correction. These relative intensity data were obtained by choosing the charge 3 line as a standard, and making several alternating settings on charge 3 and on the line under investigation, making the necessary allowance for source depletion and background. It is assumed that the ions of differing charge were equally efficiently transmitted through the apparatus.

Since the path lengths in the source volume were rather long, it was necessary to provide a magnetic shield (Fig. 1) to prevent the earth's field from preferentially affecting the paths of the recoils of high charge. It was also necessary to check the effect of pressure, lest collisions in the gas distort the charge spectrum.

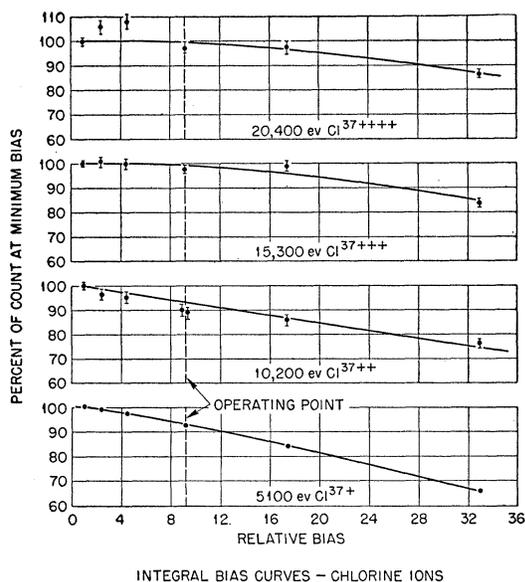


Fig. 4. Integral bias curves of the multiplier obtained with singly- and multiply-charged  $\text{Cl}^{37}$  ions.

This was done by running a second set of measurements at about double the source-volume pressure used for the first set of readings. The total source-volume pressures were in fact 6 to  $7 \times 10^{-6}$  mm Hg for the first run, and 12 to  $16 \times 10^{-6}$  mm Hg for the second; these are ionization gauge readings. The charge spectrum obtained at the higher pressure is given in the third column of Table I. Comparison of the results shows that at the higher pressure the charge 1 and 2 peaks are strengthened, while the charge 4, 5, 6, and 7 peaks are weakened; there is thus an indication that the effect of collisions is to "degrade" the charge spectrum. However, the observed differences are quite small, and it is likely that the intensities obtained at the lower pressure are not far from what they would be if there were no collisions.

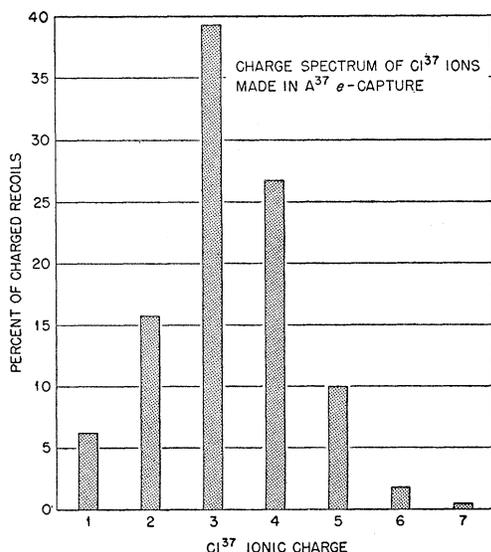


Fig. 5. The distribution in charge of the  $A^{37}$  neutrino recoils.

These observations differ from those of Kofoed-Hansen<sup>3</sup> mainly in that we find the charge 1 recoils to be considerably weaker than observed in his experiment. The mean charge as derived from our differential spectrum is  $3.26 \pm 0.03$ , which is in satisfactory agreement with the direct current measurements by Wexler<sup>12</sup> ( $3.0 \pm 0.2$ ) and by Perlman and Miskel<sup>13</sup> ( $3.41 \pm 0.14$ ).

The observation that  $0.4 \pm 0.1\%$  of the recoils have lost 7 of their 17 electrons presents a challenge in interpretation—the more so because this is a greater number than can be accounted for by the direct creation of double  $K$  (or simultaneous  $K$  and  $L$ ) vacancies in the capture event. Such a process is expected theoretically<sup>14,15</sup> and found experimentally<sup>16</sup> to occur in only 0.04% of the decays, and is thus far too weak to account for the present observation, especially inasmuch as there is no reason to expect those particular atoms to concentrate in the charge 7 state. We are accordingly faced with the problem of accounting for the emission of 7 electrons by conventional Auger processes, starting with a *single*  $K$ -shell vacancy. The transition is from the electronic configuration  $1s^2 2s^2 2p^6 3s^2 3p^6$  to the configuration  $1s^2 2s^2 2p^6$ ; that is, the atom loses all 8 of its  $M$  elec-

TABLE I. Relative intensities of the charge states of the  $A^{37}$  recoils.

Charge	Per cent of charged recoils	
	Pressure $7 \times 10^{-6}$ mm Hg	Pressure 12 to $16 \times 10^{-6}$ mm Hg
1	$6.2 \pm 0.1$	$6.9 \pm 0.1$
2	$15.7 \pm 0.4$	$16.8 \pm 0.4$
3	$39.2 \pm 0.5$	$39.0 \pm 0.5$
4	$26.7 \pm 0.4$	$25.9 \pm 0.4$
5	$10.0 \pm 0.2$	$9.6 \pm 0.2$
6	$1.8 \pm 0.1$	$1.5 \pm 0.1$
7	$0.4 \pm 0.1$	$0.2 \pm 0.1$
Mean charge of charged recoils	$3.26 \pm 0.03$	$3.20 \pm 0.03$

trons, one being used in the filling of the  $K$ -shell vacancy, and the other 7 leaving the atom. This can be accomplished only by the double use of the  $M_I$  positions; they are first emptied by Auger events, then filled from the  $M_{II}$  and  $M_{III}$  shells, and finally emptied again. The process can be plausibly traced in Fig. 6 as follows: The  $K$  vacancy at the left is filled by an  $L_I$  electron with the concomitant emission of the other  $L_I$  electron (arrow) as an Auger electron. The two  $L_I$  vacancies thus created are filled from the  $L_{II}$  or  $L_{III}$  shells; this transition does not provide enough energy to remove another  $L$  electron from the atom, so the best the atom can do is to throw off  $M$  electrons. We suppose these to be  $M_I$  electrons. At this stage we have vacancies in the four positions indicated in the third column. We now suppose that the two  $M_I$  vacancies are filled by ( $M_I - M_{II}$   $M_{III}$ ) transitions as shown in the fourth column. Simultaneously, one of the  $L_{II,III}$  vacancies can be filled by an Auger process from the  $M$  shell, as

<sup>12</sup> S. Wexler, Phys. Rev. **93**, 182 (1954).

<sup>13</sup> M. L. Perlman and J. A. Miskel, Phys. Rev. **91**, 899 (1953).

<sup>14</sup> H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).

<sup>15</sup> M. Wolfsberg, Phys. Rev. **96**, 1712 (1954).

<sup>16</sup> J. A. Miskel and M. L. Perlman, Phys. Rev. **94**, 1683 (1954).

shown at the top. We have now used all of the 8 originally available  $M$  electrons, but we have the two  $M_I$  positions filled, and we have not yet filled the other  $L_{II,III}$  vacancy in the third column. One of the two new  $M_I$  electrons can now fall into this  $L_{II,III}$  vacancy, causing the emission of the other (dotted lines). Thus the atom will have disposed of all its  $M$  electrons, and a count of the arrows will show that 7 electrons have left the atom.

The precise chain outlined above is not quite unique as to details, but the double use of the  $M_I$  positions is essential for the removal of seven electrons. The energetic possibility of the ( $M_I-M_{II} M_{III}$ ) transitions (twice used in the above scheme) is not clear, and we are compelled to conclude that the present experiment demonstrates this possibility, at least in these disturbed atoms.<sup>17</sup>

## II. Natural Widths of the Recoil Lines

While we were examining the charge 1 line, it became apparent that it was particularly narrow, and since this observation persisted under a variety of source voltage conditions, special attention was given in the experimental line shape analysis to its width compared to that of other lines, the easiest of which to study was the intense one of charge 3. The results of this comparison are given in Fig. 7. Here the points give experimental results obtained with very low applied source voltage (about 7 volts) so that the instrumental transmission width is much smaller than the natural widths of the lines. (The instrumental widths are separately indicated near the bottom of the figure.) The continuous line gives the theoretical peak shape that would be expected to result from the thermal motion of the A<sup>37</sup> atoms ( $kT=0.025$  ev) and the dotted line shows how the thermal peak is widened when one includes the recoil arising from the emission of randomly-directed, 2300-ev Auger electrons which, according to Kofoed-Hansen,<sup>3</sup> are the most energetic Auger electrons emitted. The theoretical line shapes have "folded into" them a small contribution (about 3%) from the appropriate finite instrumental resolution. We are indebted to Dr. J. M. Jauch for calculating the theoretical peak shapes, and to Miss Thelma Arnette who, under the guidance of Dr. T. A. Welton, was kind enough to compute the "folding in" effect.

Both theoretical peaks have been positioned in the figure on the assumption of a recoil energy of 9.66 ev. For clarity the experimental peaks have been shifted to an approximate centering on the theoretical line shapes. This was done because the data were obtained by the

<sup>17</sup> Note added in proof.—In a very recent article, Wolfsberg and Perlman [Phys. Rev. **99**, 1833 (1955)] suggest that there may be an electronic excitation arising from the changes of the electric fields within the atom that occur as Auger electrons are released. Such a process might excite the  $M$  electrons to the extent that the  $M_I-M_{II}M_{III}$  transition is definitely energetically possible, and so remove doubts about the cascade mechanism suggested above, or it might even cause ionization independently.

In estimating the energetic possibility of transitions we have used the compilation of x-ray absorption edges given by Hill, Church, and Mihelich [Rev. Sci. Instr. **23**, 523 (1952)].

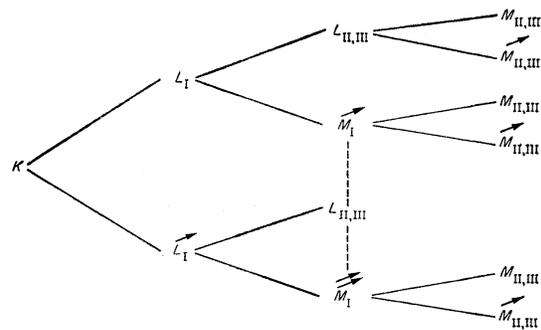


FIG. 6. Possible sequence of events leading to a Cl<sup>37</sup> charge 7 ion. The positions marked with arrows indicate the electrons emitted by Auger processes as described in the text.

use of an early fluxmeter (not a proton resonance device, but a null-reading, moving coil instrument) the calibration of which was very slightly nonreproducible. The calibration affects the relative positioning of the recoil energy abscissa scale and the source voltage scales, against which the counting-rate readings were

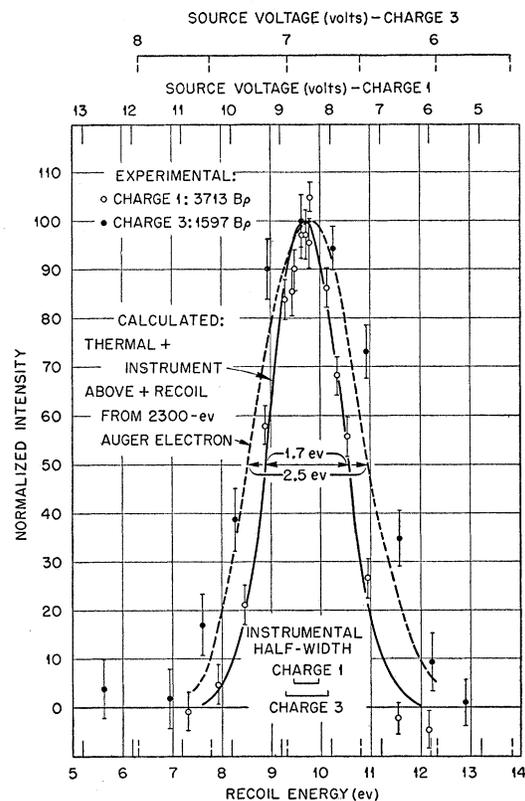


FIG. 7. Comparison of the natural shapes of the singly- and triply-charged recoil lines. The points are experimental and the curves are theoretical. The working scales for the experimental data are given at the top; it will be observed that the ion energies were very low, and the instrumental transmission width is much less than the natural widths of the lines. Background has been subtracted, the intensities have been normalized, and small shifts (within the fluxmeter errors) have been applied to the data to match them at 9.66-ev recoil energy so as to give a clear comparison of the line shapes. The singly-charged line is seen to be much narrower than the triply-charged line. A suggestion for the reason for this narrowness is given in the text.

actually obtained, but it does not affect the peak shape. Thus to obtain a clear picture for the comparison of line widths, the source voltage scale for the charge 1 readings was shifted 0.2 ev, and that for charge 3 0.35 ev, upward on the recoil energy scale. It should therefore be understood that Fig. 7 has been arranged purely for line shape comparisons, and it is not to be taken as presentation of accurate recoil energy measurement.

The result of the line shape study indicates that the charge 3 line is experimentally much wider than the charge 1 line, and indeed it is slightly wider than would be expected on the basis of thermal motion plus Auger recoil. The reason for the excess width is not clear, although some of it can be attributed to slight additional recoil from the emission of the second and third Auger electrons, which may have an energy of about 250 ev.<sup>18</sup> Thermal motion alone accounts for the width of the charge 1 line. This observation can hardly be instrumental, for it is difficult to think of an experimental

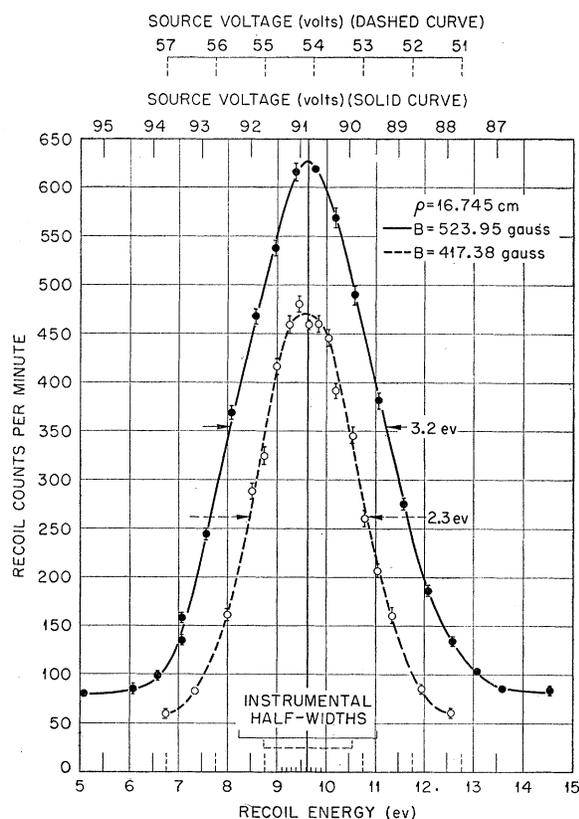


FIG. 8. Two separate determinations of the recoil energy. The working scales showing source volume voltage are given at the top, and these have been related to the recoil energy scale at the bottom, taking  $\rho = 16.745$  cm and  $B = 523.95$  gauss for the continuous curve and  $B = 417.38$  gauss for the dashed curve. The continuous curve, measured at the higher source voltage, is wider on the absolute scale because it has in it more of the instrumental width than does the dashed curve. Note that the zero of the recoil energy scale is far off the figure to the left.

<sup>18</sup> An extension of the line-width calculation has since shown that the ejection of a second Auger electron with an energy of 250 ev will broaden the line by less than 0.1 ev in the full width at half-intensity, so that some of the observed width remains unexplained.

effect that can make a line look *narrower* than it really is. Another line width comparison was made with about 90 volts on the source volume for the charge 1 ions, and 30 volts for the charge 3. The readings were statistically much more accurate than those of Fig. 7, but at these higher voltages the effect of folding in the (relatively) increased instrumental width becomes a major consideration, and the experimental comparison is slightly less direct. The result however was the same: Auger recoil is required to explain the breadth of the charge 3 peak, but it is not present in the charge 1 peak.

A plausible explanation for the narrowness of the charge 1 recoil line emerges when we consider the processes that can lead to singly charged recoils:

(1) *K*-capture, with the *K*-vacancy filled by an Auger process. This will lead immediately to two vacancies (probably in the *L*-shell), and no further Auger processes will be permitted.

(2) *K*-capture followed by "fluorescent" emission of the *K* x-ray. This is known to occur in about 9% of the decays,<sup>19</sup> and it will lead to a single vacancy in the *L*-shell. The atom is still neutral, so one subsequent Auger process is called for.

(3) *L*-capture, known to occur in 8% of the decays,<sup>20</sup> resulting in a single *L*-vacancy, and requiring one subsequent Auger process to give the ion its charge.

Now we have only to look at the preponderance of multiple charges in the spectrum to realize that the Auger processes in the *L*- and *M*-shells are very probable, and that the requirement of process (1) that *two* inner-shell vacancies be filled simultaneously with *no* Auger transitions might be very severe compared with the situations in (2) and (3) where there is only one vacancy and a subsequent Auger transition is actually called for. If the severity of this requirement is such as to outweigh the initial favorable chance of process (1), then the charge 1 peak will be fed mainly through processes (2) and (3), neither of which is accompanied by the emission of the 2300-ev Auger electron. Thus the charge 1 peak may have inappreciable natural spread other than that arising from thermal motion. There is sufficient intensity in processes (2) and (3) easily to account for all of the charge 1 recoils on the basis of our measurement of their strength in the charge spectrum.

### III. Energy of the Recoils

One chooses the charge 1 recoils as the most sensitive indicators of the recoil energy, and it is best to select a source voltage such that the proportional width of the line presented to the spectrometer approximately equals the instrumental transmission width. Under these conditions the line stands well above background, and little can be gained by going to lower voltages where the line starts to broaden out and fade into the background, nor to higher voltages where the recoil contribution becomes

<sup>19</sup> See the compilation of fluorescent yield data by Broyles, Thomas, and Haynes, *Phys. Rev.* **89**, 715 (1953), and E. H. S. Burhop, *The Auger Effect and Other Radiationless Transitions* (Cambridge University Press, Cambridge, 1952), p. 48.

<sup>20</sup> Pontecorvo, Kirkwood, and Hanna, *Phys. Rev.* **75**, 982 (1949).

a smaller fraction of the total energy. For the especially narrow charge 1 line this situation occurs when about 50 volts are applied to the source volume. Actually we made two careful examinations of the charge 1 peak, one at about 50 volts and one at about 90 volts. The results are plotted together in Fig. 8. Background has not been subtracted because it is instructive to leave it explicitly indicated. The working scales are indicated at the top, and through the calibration of the instrument these have been related to the recoil energy scale shown at the bottom. The two peaks are quite symmetric, and we have supposed it legitimate to use the center line as the indicator of the peak position; the same procedure was used in the calibration. The 90-volt peak centers at 9.64 ev with an uncertainty of  $\pm 0.02$  ev in the centering, and the 50-volt peak centers at 9.61 ev with an uncertainty of  $\pm 0.03$  ev. It seems acceptable to adopt  $9.63 \pm 0.02$  ev as the average, but to this we must attach an error of  $\pm 0.05$  ev as a possible calibration inaccuracy. Thus we arrive at a final value  $9.63 \pm 0.06$  ev for the energy of the A<sup>37</sup> recoils. It will be recalled that the value based upon the Cl<sup>37</sup>(*p,n*)A<sup>37</sup> threshold was expected to be  $9.65 \pm 0.05$  ev.

It is of interest to see what limit this determination places upon the rest mass of the neutrino. The momentum balance is expressed through the equation  $2ME_r = (T/c^2)(T+2\nu c^2)$ , whence it is apparent that if the neutrino rest energy  $\nu c^2$  were to amount to 1% of its kinetic energy *T*, that is, if it were to amount to 4 kev, then the expected recoil energy would be  $9.75 \pm 0.05$  instead of  $9.65 \pm 0.05$  ev, and this would be just outside

of the estimated errors so far as agreement with our determination is concerned. It is safer to state that from the evidence the neutrino rest energy is probably less than 5 to 6 kev. This limit is of course less restrictive than is the limit obtained from the shape of the high-energy end of the tritium beta spectrum, but it is based upon an independent experimental method and one that involves theoretical considerations no more complicated than the conservation of momentum. The conclusion also rests upon the assumption that the Cl<sup>37</sup>(*p,n*)A<sup>37</sup> threshold can legitimately be taken as determining the *Q*-value for that reaction.

In conclusion we may state that we have apparently encountered none of the expected difficulties that are supposed to beset the deflection of ions of low energy—that is, difficulties related to contact potentials and the charging-up of surfaces. Aside from reasonable (but not extreme) cleanliness, the only precautions that we took were to use mercury diffusion pumps to avoid internal surface films of insulating oil, and to have the ions see only stainless steel throughout their flight. Perhaps the saving features were that the apertures were large and the current densities extremely small. We expect to use the spectrometer in the measurement of continuous recoil spectra accompanying the beta decay of rare gases, but here an additional stage of differential pumping will have to be added because one then will forego the concentration of the recoil intensity into lines. Meanwhile, the method has proved itself under rigorous conditions in the detailed investigation of the very low-energy A<sup>37</sup> line spectrum.

### Radioactive Decay of the Isomers of Americium-242\*

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The decay characteristics of the two isomers of Am<sup>242</sup> have been investigated. The beta decay of the 16-hour Am<sup>242m</sup> has a branching ratio of  $51 \pm 5\%$  to the first-excited state (42.3 kev) of Cm<sup>242</sup> with the remainder of the decay going to the ground state. The electron capture decay of Am<sup>242m</sup> has a branching ratio of approximately 60% to first-excited state (44.8 kev) of Pu<sup>242</sup>. An upper limit of 6% has been set for the fraction of Am<sup>242m</sup> decay via isomeric transition. The beta decay of the 100-year Am<sup>242</sup> has a branching ratio of  $45 \pm 5\%$  to the first-excited state of Cm<sup>242</sup>, with the remainder of the decay going to the ground state. The beta-spectrum end points for Am<sup>242m</sup> and Am<sup>242</sup> have been measured to be  $620 \pm 10$  kev and  $585 \pm 10$  kev, respectively. A decay scheme for the two isomers has been proposed. *Logft* values have been calculated for beta and electron capture decay of the isomers and are discussed in conjunction with spin and parity assignments.

#### I. INTRODUCTION

THE two isomers, 16-hour Am<sup>242m</sup> and long-lived (approximately 100 years) Am<sup>242</sup>, were first observed as neutron-capture products of Am<sup>241</sup>.<sup>1</sup> The

decay characteristics of these isomers, particularly Am<sup>242m</sup>, have been studied by O'Kelley *et al.*<sup>2</sup> From the experimental data obtained by the authors, it is possible to formulate decay schemes for these isomers which differ in certain respects from those reported in

\* This work was performed under the auspices of the U. S. Atomic Energy Commission. The contents of this report have been presented in somewhat greater detail in University of California Radiation Laboratory Unclassified Reports UCRL-2325, UCRL-2528, and UCRL-2537 (unpublished).

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<sup>1</sup> Seaborg, James, and Morgan, *The Transuranium Elements*:

*Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, p. 1529; Seaborg, James, and Ghiorso, NNES, p. 1554; W. M. Manning and L. B. Asprey, NNES, p. 1595.

<sup>2</sup> O'Kelley, Barton, Crane, and Perlman, *Phys. Rev.* **80**, 293 (1950).