Determination of the Low-Energy Region of the Tritium Beta Spectrum*

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The tritium beta spectrum has been determined in a proportional counter spectrometer fitted with field-adjusting electrodes down to an energy of 200 ev. Below 1.2 kev the experimental spectra exhibit positive deviation from the theoretical spectrum which was corrected for the screening effect and for counter resolution. Various possible explanations for the deviation, which amounts to +6.5% at 0.3 kev, are examined. A possible explanation is that w, the energy to produce an ion pair in the counting gas, increases by several percent in the energy interval 1.2 to 0.25 kev.

I. INTRODUCTION

FOLLOWING the publication by Fermi[1] of his theory of beta decay, many spectra were examined to test the theory in allowed transitions. With the exception of the few cases noted in the Discussion, Sec. IV, it was found that the experimental results verified the theoretical (corrected for screening and the finite nuclear size) except for distortion at low energies, which in the case of solid samples was caused by finite sample thickness, back scattering, window absorption, etc.[2] These causes of distortion can be avoided by use of a proportional counter spectrometer with an internal gaseous source. However, at the ends of a simple counter the electric field strength and, therefore, the gas gain is too low. This effect decreases the pulse heights of the betas absorbed in this region so the experimental activity is too large (positive deviation) at the low-energy end of the spectrum. This distortion can be eliminated by the use of field-adjusting electrodes to adjust the field at the ends to the same intensity as it is at the center of the counter.[3,4]

An interesting spectrum examined by this technique is the allowed (favored) transition of tritium which provides a check of Fermi's theory at the lowest possible energy. The spectrum obtained exhibited negative deviation at low energies, e.g., the experimental activity was less than one-half of the theoretical activity at 200 ev.[5] However, on the basis of additional experiments Curran now believes that the deviation may have been due to deficiencies in the analyzer.[3] Because the results of these latter experiments have never been published, to the best of the authors' knowledge, and it seemed desirable to have an independent determination of this spectrum, the spectrum has been examined again.

II. EXPERIMENTAL PROCEDURE

The brass counter (6.2-in. i.d. by 37.8-in. active length) was fitted with an Al liner as the cathode and with field adjusting electrodes similar to those described by Cockcroft and Curran[4] except for the thinner (0.01-in.) electrode walls. The counter was fitted with two Mylar windows for x-ray calibration, one in the center of the counter and one 0.2 in. from the end of one field-adjusting electrode. Since the gas gain is very dependent on the anode radius, a close tolerance ($\pm\frac{1}{2}\%$) 0.002-in. wolfram wire was used as the anode.

Tritium was counted as hydrogen gas. A trace of HOT was reduced by $LiAlH_4$ and passed through a liquid N_2 trap packed with broken Pyrex tubing into the counter. Then a few mm of carrier H_2 and one atmos of P-10 (90% Ar, 10% CH_4) gas were added.

In counting, the center wire was grounded, and negative potential was applied to the counter wall by a very stable commercial high-voltage supply to avoid high-voltage breakdown noise. The pulses from the counter were fed into a low-noise 40 gain pre-amplifier with an input stage similar to that used by Engelkemeir and Magnusson.[6] At first the pre-amplifier was connected to a commercial proportional pulse amplifier, but the recovery time of this particular amplifier is 500 μsec at 10 times overload. If another pulse appeared during this recovery time, its pulse height would be decreased, which would cause positive deviation in the experimental spectrum. With this amplifier an upper limit would therefore be set on the counting rate which would set a limit on the accuracy of the determination in a reasonable counting time. This long recovery time is inherent in the capacitive coupling used in this type amplifier. Therefore, a completely dc-coupled proportional pulse amplifier was constructed with a gain of 1000 and a linearity better than 1.2%. With a 5-μsec differentiating RC at the input to this amplifier, the recovery time for a 10 times overload pulse is 84 μsec.

The amplifier was dc-coupled to the RIDL Model 3300 100-channel analyzer[7] to avoid differentiation of the pulses which would cause overshoot pulses. The

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[1] E. Fermi, Z. Physik **88**, 161 (1934).
[2] M. E. Rose, *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (Interscience Publishers, Inc., New York, 1955), pp. 279–284.
[3] S. C. Curran, *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (Interscience Publishers, Inc., New York, 1955), pp. 165–182.
[4] A. L. Cockcroft and S. C. Curran, Rev. Sci. Instr. **22**, 37 (1951).
[5] G. M. Insch and S. C. Curran, Phil. Mag. **42**, 892 (1951).
[6] D. W. Engelkemeir and L. B. Magnusson, Rev. Sci. Instr. **26**, 295 (1955).
[7] Radiation Instrument Development Laboratory, 5737 South Halsted Street, Chicago, Illinois.

analyzer was modified so that the store pulse was 104 μsec wide. This had the effect of blocking out the time following every pulse so the dc amplifier was fully recovered before the next pulse could be analyzed. Therefore, there can be no distortion of the type described above.

The potential of the field adjusting electrodes was adjusted until the Mn K x-ray peak (from electron capture in Fe^{55}) was at the same channel number at the edge of the electrode as at the center of the counter. At this potential the ratio of the field electrode voltage V_p to the cathode voltage V_c was 0.535 compared to the theoretical ratio calculated from the known counter dimensions of 0.550. This difference is outside the 1.5% uncertainty in measuring the ratio and may indicate a slight error in centering the center wire. However, even when V_p/V_c was changed from 0.535 to 0.550 in one experiment (counting error $\pm 0.5\%$), the positive deviation at 250 ev was only decreased by 1.0%. (See Results, Sec. III.)

The energy scale was calibrated by determining the position of the 5.93-kev (energy calculated from the energies and abundances of the various K x-rays[8,9]) Mn K x-ray peak as a function of amplifier gain. The channel widths varied from 30 to 170 ev.

Three samples with activities of 86 000, 73 000, and 18 700 disintegrations/min were measured at 2385 v, and two samples with activities of 42 000 and 85 000 dis/min were measured at 2490 v.

III. RESULTS

The equation relating the number of counts per unit energy interval, $N_{(W)}$, to the total energy of the beta particle, W, is[2]

$$N_{(W)}=F_{(Z,W)}pWK^2(W-W_0)^2,$$

where p is the momentum, W_0 is the total disintegration energy of[10] 1.0352 in mc^2 units, and K is a constant independent of W. The nonrelativistic form of the Fermi function evaluated for tritium decay,

$$F_{(Z,W)}=\frac{(2.15/E)^{\frac{1}{2}}}{1-\exp(-2.15/E)^{\frac{1}{2}}},$$

was used below 2.5 kev where E is the kinetic energy in kev.[11] Above this energy F was calculated from the tabulated values of p^2F by interpolation.[11]

A preliminary spectrum was taken for each sample and a Fermi plot constructed from the data. (See the Appendix for a discussion of the end-point energies obtained.) Above 1 kev all the points in each plot fell on a straight line (maximum deviation from 1–10 kev $\pm 2\%$ in $N_{(W)}$) except that the $T70$ and $T71$ plots showed slight negative deviation above 12 kev. This was caused by a decrease in the gas gain for the higher energy betas at the large gas gain,[12] but this does not effect the shape of the spectrum in the region of interest. Because the Fermi plots indicated that the experimental spectra agreed with theory at 2 kev and Curran had reported deviation at lower energies, the region below 2.2 kev was examined carefully.

The theoretical spectrum in this region was corrected for the screening effect of the atomic electrons[2,13,14] and for the counter resolution. Dzhelepov and Zyryanova discuss the various equations which have been used for the screening correction. However, in the nonrelativistic region examined in the case of tritium, all are equivalent. With a change in potential due to screening, V_0, of 30 ev, the correction is a maximum of 0.45% at 300 ev when the corrected curve is normalized to the uncorrected curve at 2.00 kev.

The two causes for the counter resolution are statistical variations (1) in the number of original ions produced and (2) in the gas gain.[15] It was assumed that only the first effect need be considered under the present conditions (gas gain$\gg$number of original ions) in calculating the resolution (full width at half height) of the Poisson or Gaussian curves representing a monoenergetic source. (The experimental resolution of the Mn K x-ray peak of 17% is in agreement with this assumption.) In correcting the theoretical spectrum for counter resolution, the procedure was first to divide the spectrum into small energy intervals. Then a Poisson (below 270 ev) or a Gaussian curve was calculated at each energy with the area of each curve normalized to the $N_{(W)}$ for that energy. Then if $N_{i(W)}'$ is the theoretical counts per unit energy interval at the ith energy corrected for counter resolution and $N_{ij(W)}$ is the counts per unit energy interval at the ith energy for the Gaussian (Poisson) calculated for the jth energy,

$$N_{i(W)}'=\sum_j N_{ij(W)}.$$

This resolution correction amounted to 0.8% at 200 ev when the corrected curve was again normalized at 2.00 kev. Since the correction for finite nuclear size is insignificant at low atomic numbers,[16] it is felt that this final theoretical spectrum which has been corrected for the screening effect and the counter resolution, is accurate to 0.5%.

The experimental spectra are compared to the corrected theoretical spectrum in Figs. 1 and 2 with the curves normalized in the region 1.8 to 2.2 kev. The lengths of the arrows indicate the average deviation in

[8] S. Fine and C. F. Hendee, Nucleonics **13**, No. 3, 36 (1955).
[9] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1935), second edition, p. 640.
[10] L. M. Langer and R. J. D. Moffat, Phys. Rev. **88**, 689 (1952).
[11] I. A. Stegun, *Tables for the Analysis of Beta Spectra*, U. S. Department of Commerce, Applied Mathematics Series 13, (U. S. Government Printing Office, Washington, D. C., 1952).
[12] D. West, *Progress in Nuclear Physics*, edited by O. R. Frisch (Academic Press, Inc., New York, 1953), Vol. III, p. 24.
[13] R. H. Good, Phys. Rev. **94**, 931 (1954).
[14] B. S. Dzhelepov and L. H. Zyryanova, *Influence of the Atomic Electric Field on Beta Decay* (Academy of Sciences, U.S.S.R. Khlopin Radium Institute, Leningrad, 1956), pp. 37–39.
[15] Curran, Cockcroft, and Angus, Phil. Mag. **40**, 929 (1949).
[16] M. E. Rose and D. K. Holmes, Phys. Rev. **83**, 190 (1951).

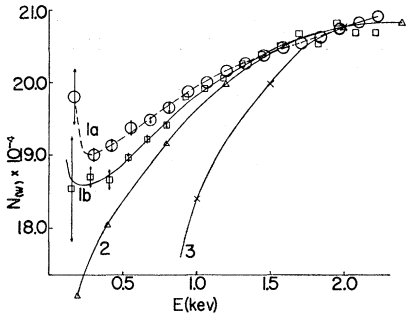

FIG. 1. Tritium spectra: (1) $T61$, $T67$, and $T68$ at 2385 v (a) at gain 1000 and (b) at gain 500; (2) theoretical spectrum corrected for screening and counter resolution; (3) Insch and Curran data.[5,3]

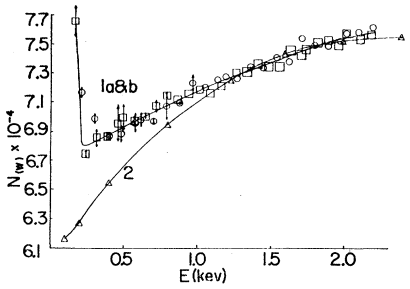

FIG. 2. Tritium spectra: (1) $T70$ and $T71$ at 2490 v (a), ○, at gain 500 and (b), □, at gain 200 and (2) theoretical spectrum corrected for screening and counter resolution.

$N_{(W)}$ for the various spectra. Most of the deviation is due to the statistical counting error (0.5%).

At each gain the gross counts were corrected to the same actual (live) counting time using the analyzer dead time meter which was later calibrated against an RIDL Model A-268 live timer. The dead time correction was good to 1% so spectra taken at two different gains should agree to within 1%. However, it was found that at low energies the spectra of samples $T61$, $T67$, and $T68$ taken at gain 500 were lower than those at gain 1000 (3.3% at 285 ev). At least part of the difficulty was traced to analyzer nonlinearity. The linearity of the analyzer is dependent on the slope of the linear sweep circuit being constant over the entire range from 0 to 90 v.[17] When the "saw tooth" pulse from this circuit was displayed on a Model 535 Tetronix oscilloscope and both the x (time) and y scales changed by the same factor, it was found that the slope increases over the first 5 μsec (10 channels) and is constant thereafter. This nonlinearity causes the lower channel widths to be too small and, therefore, $N_{(W)}$ to be too low. Of course, at the same beta energy the distortion is greater, the lower the gain.

The spectra shown in Fig. 1 have been corrected for this distortion effect by the following method: Long counts of the same spectrum were taken with channel 2 and with channel 10 equal to zero energy. Since the two spectra should have the same shape and the second spectrum is above the region of distortion, correction factors at each channel could then be determined to correct the first spectrum to the second. These correction factors, which were accurate to ±0.5%, were used to correct the $T61$, $T67$, and $T68$ data. Samples $T70$ and $T71$ were examined with zero energy at channel 10.

The counter became slightly contaminated during each determination. The contamination could be removed by exchange with inactive water at 2.5-cm pressure, but was much worse on the following determination if the counter was not first well degassed to remove adsorbed water. Therefore, it was assumed that the contamination resulted from a counter discharge induced exchange during the very long counting times between HT and traces of water or hydroxyl groups on the counter wall. A background was used in each run which was weighted assuming the contamination was picked up linearly with time. This correction for contamination affected the shape of the spectra by less than 1% except for $T70$ where it changed it by 1.8%.

IV. DISCUSSION

Within 1% the counting rate has no effect on the shape of the spectra. Although the comparison spectra are not shown, the small average deviations in Figs. 1 and 2 indicate this fact.

It is seen in Fig. 1 that there is a disagreement in the shapes of the spectra taken at two gains at 2385 v. The discrepancy could have been caused by two or three of the following effects: (1) an error in the correction for analyzer nonlinearity, (2) an error in the dead time correction which would shift one curve relative to the other, and (3) amplifier nonlinearity at the highest gain. At 2490 v the agreement between the spectra taken at the two gains is excellent, which is an indication of amplifier linearity.

The spectra are also independent of the counter voltage within 1%. The degree to which the shapes of the spectra are independent of the various experimental parameters is indicated by the average deviation of the four experimental spectra shown in Figs. 1 and 2 from the corrected theoretical spectrum. These deviations are 6.5±0.6% at 300 ev and 3.8±0.5% at 500 ev.

It is interesting to note that recently several beta spectra have been examined carefully and found to exhibit positive deviation at low energies.[18,19] The Kurie plots were made linear by dividing $N_{(p)}$, the counts per unit momentum interval, by $1+b/W$, where $0.4>b>0.2$. Since W varies by less than 1% over the energy interval covered in the present investigation, this type of correction factor would have no effect on the tritium spectrum.

Various possible explanations for the deviation below 1.2 kev will now be examined. The total activity

[17] R. W. Schumann and J. P. McMahon, Rev. Sci. Instr. **27**, 675 (1956).

[18] Johnson, Johnson, and Langer, Phys. Rev. **112**, 2004 (1958).
[19] Hamilton, Langer, and Smith, Phys. Rev. **112**, 2010 (1958).

in excess of the theoretical spectrum above 200 ev is only 0.2% of the total tritium activity. Thus if only a small fraction of the overload pulses had small positive overshoots which were counted, the deviation would be accounted for, but the use of the blocking pulse following each pulse negates this type of explanation. In addition, the deviation is not caused by noise as even at the lowest energy the correction for noise plus background was less than 6% of $N_{(W)}$ except for $T68$ in which it was 26%.

Two additional sources of distortion are the wall and end effects which give degraded pulses in the low-energy region when only a portion of the beta particle's ionization path lies in the active counter volume. If it is assumed that one-half of the betas approach the wall normally, the weighted fraction of the higher energy betas which give pulses in the region 0–1 kev because of the wall effect is

$$2\pi r \sum_{0}^{18\text{ kev}} N_{i(W)}\Delta r_i / 2\pi r^2 \sum_{0}^{18} N_{i(W)} = \sum_{0}^{18} N_{i(W)}\Delta r_i / r \sum_{0}^{18} N_{i(W)} = f,$$

in which r is the counter radius and Δr_i is the path length for 1 kev of energy absorption in the counting volume for the ith energy beta. The value of Δr_i was calculated from the range-energy relation of Lane and Zaffarano.[20] The average percent deviation over the interval 0–1 kev is then $100f\sum_0^{18} N_{i(W)}/\sum_0^{1} N_{i(W)}$. The same calculation was made for the interval 1–2 kev. The result of these calculations show that the net distortion from 0 to 1 kev from the wall effect should be less than 0.4%. A similar calculation for the betas entering the active volume from the ends shows that distortion from this effect should also be less than 0.4%. If a correction is made for the fact that the betas actually approach the wall (end) at an angle, these distortions are approximately halved. Thus the positive deviation cannot be explained by wall or end effect.

The pulse heights of the proportional pulses are proportional to the number of ions released by each beta particle on being absorbed by the counter gas. Thus the ordinate of the experimental spectra is actually the number of counts per unit electron interval, $N_{(e)}$. The relation between the experimental $N_{(e)}$ and the theoretical $N_{(W)}$ is

$$N_{(e)}/w = kN_{(W)},$$

in which w is the energy to produce an ion pair in the counter gas and k is a constant. If w were to increase as the beta energy decreases from 1.2 to 0.2 kev, the experimental spectra would exhibit positive deviation at low energies. Much experimental evidence indicates that w is constant over the range 2.8 to 0.24 kev.[21–23] However, the limits of error are large (5%), and indeed the lowest limit of error was obtained for N_2 by assuming that w is constant in Ar and comparing the Ar^{37} K/L peak height ratio in N_2 to the ratio in Ar.[22] In addition, all the evidence does not support the constancy of w.[24,21] Thus it is barely possible that the deviation between the experimental spectra and the theoretical spectrum is caused by a 4–6% increase in w for beta energies between 1.2 and 0.25 kev. If additional experimental evidence should establish that w is constant to 4% to 0.25 kev, the present experimental results can be considered to be at variance with the theoretical prediction. The authors have no explanation for the large deviation below 0.2 kev, as it seems unlikely that the variation in w can be of such a magnitude.[21]

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APPENDIX

The average of the end-point kinetic energies (E_0) obtained by the Fermi plots is 18.78±0.05 kev. This value would be decreased by about 0.3 kev if the spectra were corrected for the finite counter resolution.[25] However, when the anode in another counter was divided by a glass bead, the difference between the positions of the Mn K x-ray peaks in the two counting volumes for five fillings was (2±1)% greater than the amount due to the capacitance difference. If this effect be ascribed to nonuniformity in anode diameter, the energy scale is uncertain to about ±2%. Therefore, the more accurate E_0 of Langer and Moffat was used in calculating the theoretical spectrum.‡

[20] R. V. Lane and D. J. Zaffarano, Phys. Rev. **94**, 960 (1954).

[21] J. M. Valentine and S. C. Curran, *Reports on Progress in Physics* (The Physical Society, London, 1958), Vol. 21, p. 1.

[22] J. M. Valentine, Proc. Roy. Soc. (London) **A211**, 75 (1952).

[23] J. F. Lehman, Proc. Roy. Soc. (London) **A115**, 624 (1927).

[24] M. Langevin and P. Radvanyi, Compt. rend. **241**, 33 (1955).

[25] G. C. Hanna and B. Pontecorvo, Phys. Rev. **75**, 984 (1949).

‡ *Note added in proof.*—An E_0 of 18.6±0.1 kev was recently obtained by a careful examination of the spectrum in a magnetic lens spectrometer using Al backings to provide certain electrical grounding [F. T. Porter, Phys. Rev. **115**, 450 (1959)]. With this value of E_0 the deviations at 300 ev and 500 ev are 7.3±0.6% and 4.4±0.5%, respectively.

Porter calculated an E_0 of 18.21±0.03 kev from the average energy of 5.52±0.01 kev obtained by Popov, *et al.* [Popov, Gagarinskii, Senin, Mikhalenko, and Morozov, Atomnaya Energiya **4**, 296 (1958)]. As noted by Porter, the discrepancy in E_0 values could be explained if the tritium spectrum deviated from the assumed theoretical shape at low energies. However, when the experimental spectra were extrapolated to zero from 300 ev, it was shown by graphical integration that the observed deviation amounts to less than a 0.01-kev decrease in the E_0 calculated from the average energy. Indeed, even if $N_{(W)}$ be zero below 300 ev, the average energy obtained by Popov, *et al.* is incompatible with an E_0 of 18.6 kev.