A Comparison of Several Nuclear Absolute Voltage Determinations*

WILLIAM J. STURM[†] AND VIRGIL JOHNSON[‡] University of Wisconsin, Madison, Wisconsin (Received April 23, 1951)

An evaluation of the consistency between the absolute voltage determination of the threshold energy of the $Li^{7}(p,n)Be^{7}$ reaction as measured by Herb, Snowdon, and Sala and earlier independent absolute alphaparticle energy measurements has been made. The experimental comparison was affected by employing the natural alpha-particles from polonium and radium C', the energy of which has been measured absolutely by several authors using magnetic deflection methods. In this experiment the alpha-particles from these substances have been electrostatically deflected in the 90° cylindrical electrostatic analyzer at the University of Wisconsin. Alpha-particle line measurements were analyzed by numerical integration of resolution and source functions and in themselves contributed negligibly to the uncertainties of the comparison. The resulting alpha-particle energies, evaluated by comparison with the energy of the $Li^{7}(p,n)Be^{7}$ threshold, agree with earlier measurements, and indicate that within the limits of accuracy of the experiments, all determinations give identical results.

INTRODUCTION

SEVERAL accurate measurements of the absolute energies of the alpha-particles emitted by radium C' and polonium have been made by electrostatic and magnetic deflection techniques.¹ Briggs' most recent precision measurement was made using improved apparatus as well as better values for the e/m for the alphaparticle and claims an accuracy of 7 parts in 100,000. More recently, further nuclear energy measurements have been made to establish a series of reference voltages using, among others, the energy of the $Li^{7}(p,n)Be^{7}$ threshold.² Herb, Snowdon, and Sala³ as a result of their recent absolute measurements, placed the energy of the threshold for this reaction at 1.882 Mev absolute ± 0.1 percent.

This paper reports a comparison of absolute measurements of Herb with earlier absolute measurements of alpha-particle energies. This was accomplished by comparing the proton energy of the Li(p,n) threshold with the energies of the radium C' and polonium alpha-particles. All were measured on a modification of the electrostatic analyzer⁴ used by Herb, Snowdon, and Sala in their absolute voltage determinations. The analyzer as used for this experiment employed a narrower gap, and was supplied with high voltage from an electronic source in place of the battery supply of the earlier work.

APPARATUS

The cylindrical electrostatic analyzer used for these measurements deflects the proton beam from the University of Wisconsin electrostatic generator, and the alpha-particles used in this experiment, through a 90° arc (Fig. 1). Curved metal plates were used to provide the radial electric field for the deflection of the particles and were separated by $\frac{3}{16}$ -in. gap. These plates were made by the same manufacturer and produced with the same tolerance requirements as were Herb's original $\frac{5}{16}$ -in. set. An electronic high voltage supply,⁵ controlled and measured by a standard cell checked for constancy at the University of Wisconsin Standards Laboratory, provided up to 25 kev ± 0.01 percent to each of the analyzer plates. Provision was made for measuring the lithium (p,n) threshold and alpha-spectra alternately while retaining identical analyzer geometry, slit settings and consequently resolving power for the comparative results desired here. A multiple heated target housing⁶ provided a simple method for bringing any one of a group of several lithium foils into the proton beam. Neutrons from the lithium (p,n) reaction were detected by means of a BF₃ long counter placed immediately behind the target.

A scintillation counter (Fig. 1) was employed to detect the polonium and radium C' alpha-particles as they emerged from the exit slit of the analyzer. This counter was an RCA 931-A photomultiplier tube. The fluorescent screen, a ZnS compound (RCA 33-A-20A lot No. 484), was simply dusted on the glass surface of the tube to form a sensitive area for particle detection. The tube was mounted in brass housing which, involving an O-ring to seal the junction at the tube base, permitted the glass portion of the phototube to be introduced into the vacuum system of the analyzer.

EXPERIMENTAL PROCEDURE

Lithium oxide targets were prepared by evaporation of metallic lithium on tantalum foils. Oxidation occurred

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[†] AEC Predoctoral fellow; now at Oak Ridge National Labora-

[†] AEC Predoctoral fellow; now at Oak Ridge National Laboratory, Oak Ridge, Tennessee.
‡ Now at Midwest Research Institute, Kansas City, Missouri.
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when the targets were exposed to air. Six foils were prepared, each used for one threshold measurement, the thinnest being a barely visible deposit.

Three of the polonium sources were prepared by electrodeposition by the Canadian Radium and Uranium Corporation. A fourth foil was prepared at this laboratory by evaporating part of the commercially prepared deposit on to a blank nickel foil in an effort to determine the effect of the age of the source upon the nature of the spectrum at the position of the alphaparticle line. These sources of strengths, varying from 0.25 to 3.0 millicurie, were deposited on nickel metal strips and covered one 0.050×0.250 -in. face of a $0.050 \times 0.250 \times 0.750$ -in. strip of the metal.

The RaC' source was prepared at the Radium Laboratory of the State of Wisconsin General Hospital. The active deposit of RaC' was produced by exposing one centimeter at the end of a 0.020-in. diameter platinum wire to approximately 250 millicuries of radon over mercury for 40 minutes. In order to increase the strength of the deposit a cylinder of copper mesh 1 cm in diameter was fitted about the platinum wire and raised to a positive potential of 300 volts with respect to the wire. The strength of the RaC' source at the beginning of the first measurement was about 25 millicuries.

A sequence of 13 measurements, selected in order to minimize variations in time and change in the experiment arrangements, alternated alpha-line and lithium (p,n) threshold measurements. The resolution selected in each case was the optimum one chosen in view of the strengths of the alpha-particle sources. Because these measurements did not themselves represent an absolute measurement, it was not necessary to measure accurately the spacings of the analyzer plates, or to measure the absolute value of the voltages on the plates.



FIG. 1. Schematic diagram of the cylindrical electrostatic analyzer. Alpha-particle sources were placed at the entrance slit, and detection was accomplished by scintillation counting. Insert: The housing for the photomultiplier showing phosphor screen and method of producing vacuum seal.



FIG. 2. The $\operatorname{Li}^7(p,n)\operatorname{Be}^7$ threshold, showing neutron yield in arbitrary units plotted as a function of potentiometer voltage. Six determinations were used to establish the position of the threshold on the potentiometer axis. Triangles represent the width and approximate shape of the incident proton spectrum at the potentiometer setting corresponding to the position of the apex.

(a) $Li^7(p,n)Be^7$ Thresholds

Six measurements of the Li(p,n) threshold were made, each on a different lithium target. Targets were maintained at a temperature of 250° to 300° Centigrade at all times, and a liquid air trap was mounted in the vacuum system near the targets in order to avoid contamination of the lithium foils. The theoretical resolutions⁴ for all of the threshold measurements was the same, 1000. The proton beam strength measured at the target was essentially constant during each run, and varied from 0.1 to 0.25 microampere for the various threshold measurements. The beam intensity for each point on the curves was measured by means of a current integrator circuit, and counting rates were recorded in arbitrary units of neutrons per microampere of proton beam. Each measurement represented the average of two cycles of data, taken in all cases in opposite directions and in equal current intervals over the range of proton energy shown (Fig. 2). The proton energy is proportional to the voltage applied to the plates and is reported here in terms of the reading of the potentiometer which controlled the voltage to the plates. The consistency of the results is not quite as great as that of Herb, all six points here lying within 0.04 percent of the mean threshold value. However, since each target upon repeated measurements gave a more constant threshold than the group would indicate, and since no systematic variation could be found, it was felt desirable to take the mean of a number of measurements as the closest approach to duplicating the value obtained by Herb. Combining these results with Herb's work, the potentiometer reading 0.64330 was taken to correspond to the energy 1.882 Mev absolute.

(b) The Polonium Alpha-Line

Preliminary measurements of the shape of the polonium alpha-line showed that its width depended upon the cleanliness of the surface of the polonium layer. The breadth of the line at $\frac{1}{2}$ maximum was in one instance halved by cleaning the source in a sequence of organic solvents (carbon tetrachloride, benzene, acetone) before beginning measurement of the spectrum. This process, however, produced no measurable effect in the shape or position of the high energy side of the line. A survey of the energy spectrum of the polonium alpha-particles (Fig. 3) taken at two resolutions, 1000 and 667, showed that the apparent width of the line depended upon resolution employed. However, the projection of the linear portion of the high energy side of the line upon the energy axis was not affected by varying the resolution over the range of interest in this experiment. It was considered essential, therefore, to locate the position of this projection as the prime datum for the determination of the energy of the alpha-particle. Corrections which were applied to account for the



FIG. 3. The spectrum of polonium alpha-particles in the region of the line at 5.298 Mev. The resolution for this measurement was 1000.

width of the resolution function will be described subsequently. A significant rise of the background counting rate in time was observed, the rate of increase varying from one count per minute per 15 hours to one count per minute per hour. It was consequently necessary to decontaminate the alpha-detector frequently during the runs, and to make a frequent record of the background counting rate.

The results obtained showed a very constant value for the potentiometer setting corresponding to the energy of the projection of the high energy side of the alpha-line. All five determinations of this point lay within 0.004 percent of the mean value and the width of the toe of curve again corresponded roughly to the value of the resolution used. Sources of varying strengths and methods of production were used to determine whether diffusion into the nickel support influenced this value. One energy measurement, made immediately upon completion of the deposition in order to assure that any time dependent diffusion into the support was minimized, yielded a result indistinguishable from the others in this series.

(c) The Radium C' Line

Most of the considerations discussed in evaluating the high energy extrapolation for the polonium line occurred in the case of RaC'. The source was cleaned in organic solvents and introduced into the vacuum system of the analyzer within 30 minutes of its preparation. The analyzer resolution was 667 for the radium C' measurements. Since the rate of alpha-particle emission was not constant, but followed the characteristic curve of growth of RaC from the parent substances, and since the manner of deposition was not known with sufficient accuracy to make a theoretical calculation satisfactory for correcting the data, a monitor method was used to evaluate the source strength continuously. A second scintillation counter was set up and a second source of RaC' was prepared by removing some of the deposit from the original source. The monitor counter was a second RCA 931A photomultiplier and zinc sulfide screen; the screen was, in this case, covered with a 0.0005-in. aluminum foil, and the tube was painted with an opaque layer of black lacquer. The monitor source was mounted 2 cm from the aluminum foil and thus, by aluminum and air absorption, all radon and RaA alpha-particles were removed without detection. During the measurement of the RaC' line, both the monitor and the scintillation counter on the analyzer recorded data for identical intervals. It was consequently possible to correct analyzer data either point by point in proportion to the total number of counts observed by the monitor, or by plotting the monitor data as a function of time. The second alternative was chosen as the more accurate. The monitor curves measured during each run showed decay periods in good agreement with the value calculated from the periods of the parent substances and indicate that the period of growth was completed before the measurements were begun.

Each RaC' curve was the result of two cycles of data taken in opposite directions on the energy scale over the region shown, and corrected for decay as indicated above. As in the case of polonium, the extrapolation intercept is reproducibly located, both measurements lying within ± 0.003 percent of the mean value. The second measurement was made shortly after the completion of the first and was preceded by remeasurement of the analyzer slit settings and replacement of the RaC' source at the entrance slit of the analyzer.

ANALYSIS OF RESULTS

The measurements reported here were directed toward comparing three points on the energy scale, the experimental determinations being made on the same instrument and under comparable experimental conditions. The first, that of the $\text{Li}^7(p,n)\text{Be}^7$ threshold, was taken to be the extrapolation of the linear portion of the neutron yield curve upon the energy axis and was measured in the same manner as was done by Herb *et al.*³



FIG. 4. The polonium alpha-line measured with a resolution of 1000. Curve represents the result of fitting the observed data with a combination of exponential source function and triangular resolution function. The period of the exponential was 1.05 times the base width of the triangle.

The second and third were the positions of the natural alpha-particle lines from polonium and RaC'. Since these energy values in the nonrelativistic region depend linearly upon the voltage applied to the deflctor plates, the first approximation to the energy is directly proportional to the setting of the potentiometer which determines this plate voltage. Analysis, then, involve determination of the potentiometer settings corresponding to these points from the data of the last sections, and conversion of these values to equivalent energies.

Li(p,n) threshold determinations were made under conditions which, using much of the same apparatus, duplicated closely the experimental conditions of Herb's measurements. Because of the nature of the quantitative definition of this threshold, a simple evaluation of the six extrapolated potentiometer values obtained here was taken to correspond to the absolute energy value of the threshold assigned by Herb. This potentiometer setting, 0.64330 was thus taken to correspond to the energy 1.882 Mev on the absolute scale, and the energies of the alpha-particle lines were calculated from this fiducial point.

The determination of the energy of the polonium alpha-particle line was not as direct, however, and involved assumptions about the nature of the resolution and source functions. A curve of the alpha-particle spectrum in the region of the polonium line was measured with resolution 1000 and attempts were made to fit the portion in the vicinity of the line with various combinations of resolution and source functions — triangular, gaussian, and truncated triangular resolution functions were used in combination with exponential and finite "delta-functions" source functions. Exponential functions increased with energy and terminated abruptly at the energy of the alpha-particle line. The alpha-intensity measured in the potentiometer setting P will be given by

$$Y(P) = \int f(P)g(P)dP,$$

where f(P) is the source function, g(P) the resolution function, and Y(P) the intensity measured. That curve which fitted the observed prototype data best (Fig. 4) was a combination of the triangular resolution function and exponential source function and was found to yield a value for the base width of the resolution function indistinguishable from the theoretical resolutions. The period of the exponential for this curve was 1.05 times the triangular base width and the position of the alpha-line was $P_0=0.90583$ potentiometer. And since an extrapolation value of the high energy linear position of this curve was $P_e=0.90710$ potentiometer, we take P_0 for polonium alpha-particle measurements made at a resolution 1000 as given by

$$P_0 = P_e - 0.00127$$

in which P_0 is the extrapolated value of the alpha-line for the mean of the 5 measurements, 0.90708 potentiometer. Thus P_0 , the potentiometer reading corresponding to the energy of the line, is 0.90581.

From the electrostatic analyzer relations between the applied voltage and the particle energy, given in terms of the geometrical parameters of the analyzer⁴ we have

$$V'=2V_0(1-\gamma)d/b$$

in which $\frac{1}{2}V'$ is the positive voltage supplied to the positive analyzer plate (the negative plate is charged



FIG. 5. The high energy side of the RaC' alpha-particle line. The average extrapolated potentiometer reading, 1.3147, corresponds to an energy of 7.683 Mev absolute.

to the same value), V_0 is the particle energy in volts, d is the separation of the plates, and b is the mean radius of the plates. The value of γ is determined by

$\gamma = eV_0/2m_0c^2,$

where eV_0 is the particle energy, m_0 the rest mass, and c the velocity of light. Since V' is directly proportional to the potentiometer reading $P_{e'}$, we can replace V' by $kP_{e'}$ and determine k from the known values of $P_{e'}$ (0.64330) and V_0 (1.882 Mev absolute) for lithium. Thus $k=2.7141\times10^4$. Introducing this value for k, together with $P_0'=0.90581$ and the mass of the alphaparticle into the above expression we find the energy of the polonium alpha-line to be 5.298-Mev absolute.

The third point established on this energy scale was the position of the RaC' alpha-line. Two measurements of this line were made (Fig. 5) at the same resolution, 667. However, because of the short half-life of RaC', a prototype curve was made at this resolution using the polonium alpha-particle line. This prototype line was fitted as already discussed and was again fitted best by a curve of intensity resulting from a combination of an exponential source function and triangular resolution function. For this curve the value of P_0 was found to be 0.90600 potentiometer; and since the value of P_e from the curves was 0.90710, the difference $P_e - P_0$ was 0.00110 potentiometer units. This difference was increased in the ratio of the potentiometer readings corresponding to the extrapolated linear portions, P_e for RaC' and that for polonium, and was subtracted from P_e for RaC'. Thus for RaC'

$P_0 = 1.31467 - 0.00159 = 1.31308.$

And the energy of the RaC' alpha-particle calculated as in the last section is 7.683 Mev absolute.

ESTIMATION OF ERRORS

A consideration of the limitations upon the accuracy of the various factors which enter into the determination of these energies and which are used in evaluating the probable errors in the results follows:

(a) The absolute determination of the Li(p,n) threshold.—This value was established by Herb *et al.* to be 1.882 Mev absolute with an uncertainty evaluated by him to be ± 0.1 percent. The uncertainty was conservatively determined and for the purposes of this paper we accept these values.

(b) Leakage to ground on the analyzer plates.—For all data reported here no leakage current was detectable on the microammeters in series with the analyzer plates. Assuming that 0.5 microampere was detectable in this manner, the only significant voltage drop was that across the protective resistors. The error in the voltage due to this cause was less than 0.004 percent and is negligible for this work.

(c) Constancy of the standard cell.—Each of two determinations of the voltage showed three-day variations of less than 2 in 10^5 , and measurements made at the beginning and end of the series differed by less than

5 in 10⁵. Thus the cell probably remained constant within ± 0.005 percent during the experiment, and errors due to this effect were minimized by cycling neutron and alpha-particle data.

(d) Potentiometer settings.—The same potentiometer as used by Herb was used here and its contribution to the error, 1 part in 150,000, could be neglected.

(e) Constancy of analyzer power supply.—This problem has been considered by Henkel and Petree⁵ and the value given by them is ± 0.01 percent.

(f) Width of slit apertures.—Width of the entrance, diaphragm and exit slits were measured with an accuracy greater than ± 0.005 cm, and were probably reproduced to about this value. The determination of the resolution on this basis was good to ± 5 percent, and the effect upon final evaluation of particle energy influenced only the alpha-particle measurements and that only in a secondary sense, as will be discussed (j) subsequently.

(g) Magnetic field in analyzer.—The effect of the vertical component of the magnetic value has been treated by Herb *et al.* We have estimated the error from this source by taking the field within the analyzer as uniform and equal to the maximum found at any interior point during the earlier work (0.3 gauss) and as wholly in the vertical direction which is that of greatest effect. Only the difference in the correction for a 2-Mev proton and a 5.3-Mev alpha-particle is significant here and is less than 0.01 percent. Likewise, the difference in correction for a 2-Mev proton and for a 7.7-Mev alpha-particle is less than 0.02 percent, and was considered in evaluating the resultant probable error.

(h) Relativistic effects.—The complete expression for the energy of an alpha-particle in terms of the voltage at the analyzer plates is given by

$$V' = 2V_0(1 - \gamma + 2\gamma^2 \cdots) \left(\frac{d}{b} + \frac{1}{12} \frac{d^3}{b^3} \cdots \right)$$

Since the value of d/b is 4.69×10^{-3} , terms in d^3/b^3 and higher order will produce negligible correction. However, terms in γ produce a change as great as 0.04 percent in the alpha-line energies and were introduced into the calculation. Terms in γ^2 and higher produced negligible effects.

(i) Determination of slope of linear portions of curves. —The effect of statistical uncertainty upon the determination of the position of the linear portion of the curve has been determined by Bessel's method for several typical curves. The uncertainty in the extrapolations thus evaluated prove to be less than ± 0.004 percent. However, assigning to each point on the curve a statistical uncertainty of $\pm N^{\frac{1}{2}}$, where N is the total number of particles detected for the point considered, and extrapolating through the extreme values thus computed, indicated that the errors could be as great as ± 0.01 percent. The latter value was used in estimating the total error. (j) The width and shape of the resolution function. —The shape of the resolution function, as long as symmetry is preserved, does not alter the extrapolated potentiometer values significantly; however, the width of this function must be established in the case of alphaparticle data. From the curve analysis in the manner indicated earlier, we conclude that the uncertainty in the extrapolated values arising from the width of this function is less than ± 0.01 percent. An additional uncertainty due to setting the slits, as discussed in (f), produced an error of 5 percent in the width of the resolution function, and affected the energy determinations by ± 0.005 percent. A total error of ± 0.015 percent seemed to be the maximum expected from these causes.

(k) The assumption of an exponential source function.—To determine the error arising from this cause several other types of source functions were considered. As one extreme, a source "delta-function," having a width corresponding to the sum of the natural width of the alpha-particle line and the degradation in energy occurring in the source itself, was integrated numerically with the gaussian and triangular resolution functions used earlier. This was compared with the results of integrating similarly a rectangular block source function extending down to zero energy and showed that a range of ± 0.015 percent in energy would include both possibilities as well as the exponential case assumed. A rather conservative value of ± 0.02 percent was taken for this uncertainty.

(1) Degradation of alpha-particle energy at the source.—For the thickest source used, assuming a stopping power of 4.5 relative to air and a density of 9.5 for Po, the greatest loss of energy possible is of the order of 200 electron volts, and is a negligible error. Because of the method of evaluating the alpha-particle line position by extrapolation of the high energy side of the line, diffusion into the source, unless a large fraction of the alpha-emitter diffused thus, was not a significant factor. This was established experimentally by producing a fresh source on nickel and measuring it immediately. No error in these measurements was taken to arise from this cause.

The uncertainties b through l are those which arise un the determination of the potentiometer voltage corresponding to the extrapolated alpha-lines. If these errors are assumed to be random, the combined probable error in this determination is ± 0.03 percent for the Po and ± 0.036 percent for RaC'. In establishing the potentiometer reading corresponding to the Li(p,n)threshold, many of the errors discussed above (b through e) were again operative. If the distribution of threshold measurements is assumed random and if the uncertainty of 0.1 percent in Herb's measurement is included, the probable error in the evaluation of the particle energies is ± 0.103 percent. And our final results for the energy of the alpha-particles are

 $RaC' energy = 7.683 \pm 0.008$ -Mev absolute,

Po alpha-energy = 5.298 ± 0.005 -Mev absolute.

DISCUSSION OF RESULTS

In order to compare the results obtained here with the independent absolute alpha-measurements made of the Po and RaC' alpha-particle lines by magnetic deflection techniques, we have selected for the comparison the latest value of Briggs¹ for the RaC' alphaenergy and that of Rosenblum and Dupouy¹ for the Po alpha-energy. Introducing the current values⁷ for the electronic charge, alpha-particle mass and the Faraday, changes Briggs' value of e/m for the alpha-particle by 0.001 percent, and hence his value for the RaC' energy insignificantly. The value he reported for the energy of the RaC' line was 7.6802-Mev absolute ± 0.007 percent.

For the Po alpha-line, Rosenblum's values for H have been reconverted using more recent values of the constants and the energy was converted to the absolute scale. The energy thus computed, 5.297 Mev with an experimental error of less than 0.2 percent, agrees very well with the value of 5.298 Mev reported in Halloway and Livingston.⁸ This latter value was obtained by recalculation of the data of Lewis and Bowden⁹ in terms of Briggs' absolute energy for the RaC' line.

Thus within the accuracy of the experiments, present values for the alpha-particle energies agree with the earlier independent magnetic deflection measurements. Since these determinations were made electrostatically and energies assigned by comparison with the absolutely determined Li(p,n) threshold energy, it is clear that all methods yield consistent results for the energy of these alpha-particles. Consequently, it appears improbable that a systematic error as large as 0.1 percent has been made in either magnetic or electrostatic absolute determinations and that it is reasonable to accept the precision of both with increased confidence.¹⁰

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¹⁰ Note added in proof:—It may be of interest to make the inverse comparison as well, accepting the claimed accuracy (0.007 percent) of Briggs' measurement of the RaC' alpha-energy, and use the present measurements (accuracy 0.036 percent) to fix the Li(p,n) threshold. Although the small uncertainty in Briggs' absolute energy may be optimistic, the $\text{Li}^{?}(p,n)\text{Be}^{?}$ threshold thus calculated from data of the present comparison is 1.8813 ± 0.0007 Mev absolute. (The polonium alpha-energy becomes 5.296 ± 0.002 Mev.) Combining this threshold value with the other accurate absolute determinations, 1.8812 ± 0.0019 Mev absolute (reference 3), and 1.8816 ± 0.0019 Mev absolute (HSS, reference 3), yields an unweighted mean of 1.8816 Mev absolute. An uncertainty of 0.05 percent (0.0009 Mev) assigned to this mean value will generously overlap all three of the absolute and independent determinations of the threshold.