Absolute Energy Measurement of Alpha Particles from Po^{210}

F. A. WHITE, F. M. ROURKE, J. C. SHEFFIELD, AND R. P. SCHUMAN,* *Knolls Atomic Power Laboratory,† Schenectady, New York*

AND

J. R. HUIZENGA, *Argonne National Laboratory, Lemont, Illinois*

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The absolute alpha energy of Po^{210} alpha particles was measured by a 180° magnetic spectrometer. The value obtained was 5.3054±0.0010 Mev (absolute volts). Measurement of the energy (α_0) of the alpha particles from Cm^{244} leading to the ground state of Pu^{240} was made relative to the energy of the Po^{210} alpha particles. A value of 5.8025±0.002 Mev was obtained for this Cm^{244} α_0 energy. The energy difference between the ground state transition α_0 and the transition α_1 to the first excited level of Pu^{240} from Cm^{244} was found to be 43.5±1 kev.

INTRODUCTION

INTEREST in the determination of nuclear binding energies by mass spectrographic measurements and nuclear reaction Q values has been shared by many workers in recent years. The instruments used in measuring nuclear reaction energies have been calibrated in a number of ways including precise $B\rho$ measurements,[1,2] utilization of nuclear resonance,[3] and the reaction threshold of $Li^7(p,n)Be^7$ for electrostatic analyzers.[4] Comparison of nuclear binding energies computed from nuclear reaction energies with those derived from mass spectrographic measurements gives information on the original energy calibrations used for the nuclear reaction energy measurements. Additional recent measurements of the alpha energies of the transuranic elements, either by magnetic spectrographs or ion chambers with multi-channel analyzers has made desirable the absolute alpha energy determination of one or more radioisotopes which can be easily obtained as standards.

The measurement of alpha-particle energies with magnetic analyzers has previously been done by careful measurement of $B\rho$ values. The difficulties associated with these measurements, and the various sources of errors, have been discussed by previous investigators.[1–3] It is not practical to construct a magnet of large radius of curvature which has the desired uniformity of field strength over the complete pole-face area. Hence, very careful mapping is required in order to obtain the mean field strength over the precise trajectory of the alpha particle. Even if the variations in magnetic field are completely determined, it is impossible to maintain the relative field values along the trajectory if the mean field strength is varied over a large range, because of differential hysteresis.

The method presently described was conceived as one which would eliminate the necessity of knowing either the absolute or relative value of the field along the alpha-particle trajectory. For the large electromagnet described below, it seemed desirable to relate the alpha-particle energy to the kinetic energy of an ion of high atomic mass traversing an identical trajectory, utilizing a common magnetic field for both the high-mass ion and the alpha particle. The problem is thus resolved to that of making an accurate voltage measurement related to a standard cell in order to deterimine the kinetic energy of the accelerated positively charged ion of known mass.

By using nuclear magnetic resonance control to eliminate magnetic field drifts, the alpha-particle energy is determined in terms of the mass of the heavy ion and the standard cell reference voltage. This approach seemed to present the fewest possible sources of error. It is also more amenable to repeated experiments.

Ideally one would like to perform an experiment in which the alpha particle and heavy positive ions are detected simultaneously. Such an experiment is not practical. It is possible, however, to keep the radius ρ invariant with fixed source and detector slits, and to maintain the magnetic field B at an approximately constant value throughout an experiment. In addition, by using nuclear magnetic resonance very small drifts in field can be visually monitored with a frequency meter to an accuracy of one part in 10^6. In the actual comparison of the alpha particle and the heavy positive ion the small changes in the field were recorded and appropriate corrections for B made when necessary.

The magnet was designed and constructed primarily for relative measurements requiring only a high degree of uniformity along lines normal to the mean radius. Uniformity along these orthogonal lines was excellent. For large changes in B, the circumferential variations in magnetic field strengths from source to detector could not be reproduced. There was no way of circumventing this difficulty as it was impractical to map the

* Now at Phillips Petroleum Company, Idaho Falls, Idaho.

† Operated by the General Electric Company for the U. S. Atomic Energy Commission.

[1] S. Rosenblum and G. J. Dupouy, Compt. rend. **194**, 1919 (1932); J. phys. radium **4**, 262 (1933).

[2] Rutherford, Williams, Bowden, and Lewis, Proc. Roy. Soc. (London) **A142**, 347 (1933); W. B. Lewis and B. V. Bowden, Proc. Roy. Soc. (London) **A145**, 235 (1934).

[3] Collins, McKenzie, and Ramm, Proc. Roy. Soc. (London) **A216**, 219 (1953).

[4] W. J. Sturm and V. Johnson, Phys. Rev. **83**, 542 (1951).

entire pole-face area without lifting the top pole piece to remove the vacuum ring. Thus the magnet was cycled through large values of B with unacceptable reproducibility. There is, of course, no objection to this limitation for the usual relative measurements, but it leaves a wide margin of error for absolute energy determination.

Po^{210} was selected as a source for absolute energy calibration because of its general availability, high specific activity, and because of the fact that a number of previous absolute energy measurements had been carried out using this isotope.

In order to furnish a secondary standard of higher energy, the energy of the alpha particles from Cm^{244} was measured relative to the energy of the Po^{210} alpha particles. The difference in kinetic energy between the alpha particles leading to transitions to the ground state and to the first excited level of Pu^{240} was also measured. Results are discussed below.

THEORY

The determination of the energy of the alpha particle in this experiment is dependent upon a precise measurement of the kinetic energy of the accelerated heavy positive ion. The measurement of this latter is best obtained by means of a special potentiometer, a precision voltage divider, and a standard cell. A relativistic correction is required only for the alpha particle; the correction for the accelerated heavy ion is negligible.

If B is the magnetic induction, e the electronic charge, and m the mass of the ion moving at the velocity v, then

$$\mathrm{Bev}=mv^2/\rho. \tag{1}$$

For a constant value of $B\rho$, and using subscripts 1 and 2 for the heavy singly charged positive ion and alpha particle, respectively, one obtains the relation

$$B\rho=m_1v_1/e_1=m_2v_2/e_2. \tag{2}$$

Equating the velocity of the alpha particle, v_2 from Eq. (2), with the velocity derived from the relativistic equation, $E=m_0c^2(\gamma-1)$, where E is the kinetic energy of the alpha particle, m_0 is the rest mass of the alpha particle, c is the velocity of light, and $\gamma=(1-v_2^2/c^2)^{-\frac{1}{2}}$, gives

$$\frac{m_1v_1e_2}{m_2e_1}=c\left[1-\frac{m_0^2c^4}{(E+m_0c^2)^2}\right]^{\frac{1}{2}}. \tag{3}$$

Now $e_2=2e_1$, $m_2=m_0+E/c^2$, and $v_1=(2e_1V/m_1)^{\frac{1}{2}}$, where V is the accelerating voltage of the singly charged positive ion. Substitution of these quantities into Eq. (3) gives

$$E=\frac{4m_1e_1V}{m_0}-\frac{E^2}{2m_0c^2}, \tag{4}$$

or, by transposing terms,

$$E=\frac{4m_1e_1V}{m_0}\left(1-\frac{E}{2m_0c^2}+\frac{E^2}{4m_0^2c^4}-\cdots\right). \tag{5}$$

In order to determine V precisely, a precision voltage divider was constructed by utilizing a method previously described by Wenner.[5] In this method the percentage deviations of the precision resistors cancel out to the second order and an accuracy of one part in 10^6 in the divider ratio is easily obtainable. The voltage, V_1, across the low-resistance section of the divider may then be measured by a potentiometer referred to a standard cell. If R is the divider ratio, then the source voltage V is given by

$$V=RV_1+V_1.$$

DESCRIPTION OF APPARATUS

The analyzer is shown schematically in Fig. 1. The design was primarily for high-resolution work. An important feature is that alpha particles differing in energy by greater than 1 Mev could be simultaneously focused on a nuclear emulsion plate.

The top and bottom of the magnetic circuit consists of iron slabs of rectangular cross section, 30 cm×100 cm ×350 cm, in length. The top slab, to which one of the 180° sector pole pieces is affixed, rests on two iron cylinders, machined from special forgings, 62 cm in diameter and 53.3 cm in height. The exciting coils surround these cylinders. Top and bottom pole-piece sectors are 25.4 cm thick which gives a 2.54-cm gap in the magnetic analyzer region.

The pole pieces, which were plated with 0.0005-cm nickel, serve as an integral part of the vacuum chamber, i.e., there is no separate chamber or analyzer tube. A solid Duralumin ring, cut to the contour of the pole-piece sector, contains a source adapter, defining slits and baffles, pumping ports, and detector adapter. Sealing of this vacuum chamber is accomplished by a lead gasket and subsequent waxing. Pressures in the 10^{-7} mm Hg range were obtained with this arrangement. The 180° pole sectors have a cross-sectional area of approximately 6500 cm^2, and the mean radius of curvature is 76.2 cm.

In order to achieve a uniform gap the pole-piece sectors were ground flat within 50 parts in 10^6, and twelve Duralumin spacers, ground to the gap dimension, served as supports for the top pole face.

For maximum transmission, the variable baffle could be set to a half-angle of divergence of approximately $7\frac{1}{2}°$ which corresponds to a geometry of ~1 part in 5000. In order to utilize the highest possible geometry and reduce the spherical aberration of the magnet lens, a parabolic shim was constructed of pure sheet iron and inserted midway between the source and detector.

[5] F. Wenner, J. Research Natl. Bur. Standards **25**, 229 (1940).

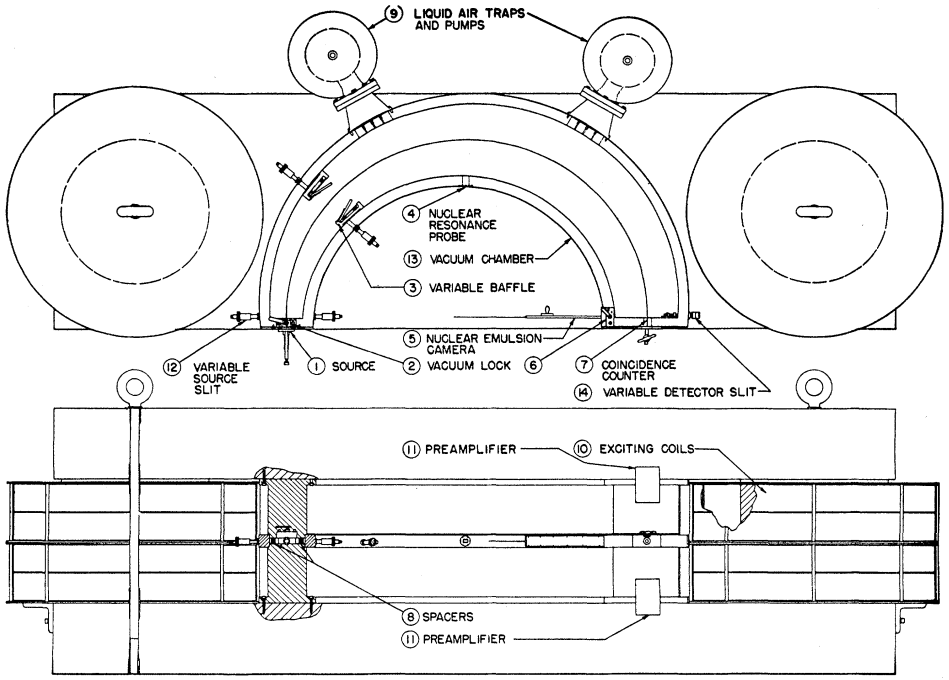

FIG. 1. Diagrammatic sketch of 180° magnetic spectrometer.

In the absolute energy measurement the shim was not used since the baffle was adjusted to a small acceptance angle.

For detecting alpha particles electronically, a coincidence-type ionization chamber was mounted directly in back of the adjustable detector slit. This was a cylindrical chamber of 3 cm diam$\times$3.8 cm length, having a 0.0005-cm rubber hydrochloride end-window and a parallel-plate-type geometry for the collecting electrodes. Rather than collect the ionization electrons on a single plate, dual collecting plates were used such that an alpha particle entering normally caused an equal number of ionization electrons to be collected on each of the two collecting electrodes. The pulses from each of the electrodes were fed through separate preamplifiers and discriminators and finally through a coincidence circuit. This technique reduced the background to 2 alpha counts/hour.

For measurement of the positive-ion beam current, a Faraday cage was used with a cylindrical housing similar to that of the alpha counter. The Faraday cage was connected to a vibrating reed electrometer and recorder.

The precision voltage divider was constructed of forty 2-megohm resistors in series coupled to four 12 500-ohm resistors in parallel, giving a nominal divider ratio R of 25 600. All the resistors were Shallcross precision resistors accurate to one part in 10 000 having a temperature coefficient less than 20 parts per million. In order to determine the divider ratio R precisely, the 2-megohm resistors were connected in parallel and the 12 500-ohm resistors in series, giving two groups of resistors each with a nominal value of 50 000 ohms. The two resistor groups were then compared by an interchange bridge method. In this method the degree of imbalance of the two resistor groups was measured within 0.1 ohm in 50 000 ohms, by using a galvanometer having a sensitivity of 6×10^{-9} amp/mm. The precision divider was inserted into a column of insulating oil to reduce any possible corona when connected to the high voltage.

The high-voltage supply was designed for regulation and stability within 0.01% per day up to 50 kilovolts output. This was achieved by means of temperature compensated precision resistors and five standard cells for potential reference in a temperature controlled oil bath. This unit also contained the transformer to supply the power for heating the filament of the surface ionization source.

The field was established by two sets of exciting coils in parallel having a total of 68 000 turns operating at currents slightly less than 0.5 ampere. An electronic regulated power supply furnished a maximum of 1 ampere at 600 volts. For more precise field regulation, an error voltage was fed to the power supply from a nuclear magnetic resonance sensing device. The magnetic field was changed by varying the nuclear magnetic oscillator frequency with a variable speed drive on the tuning capacitor. A frequency meter with a temperature-controlled crystal oscillator for reference, monitored the nuclear magnetic oscillator and measured the frequency to better than 1 part in 10^6. Except for oscillator frequency changes, the field was controlled by nuclear magnetic resonance to better than 1 part in 40 000.

PREPARATION OF SAMPLES

Po^{210}

The polonium sample was prepared from chemically purified polonium purchased from the Mound Laboratories. No further purification of the polonium was made. The sample used for the energy determination was plated on polished platinum. First a very thin film of copper was electroplated on the platinum; then the polonium was deposited on the platinum from a chloride solution with the copper simultaneously dissolving in the solution. The final sample showed no discoloration due to either impurities or residual copper.

Cm^{244}

The curium sample was produced by the very-long-term irradiation of plutonium in the Materials Testing Reactor. The curium was separated from other actinides and purified from fission products by a series of cation and anion ion exchange column processes. The final curium contained about 98% Cm^{244} alpha activity and 2% Cm^{242} alpha activity. Just before the source was prepared, plutonium daughter activity was separated from the curium by an anion exchange column process.

The curium sources were made by vacuum-subliming the curium from a tantalum filament through a defining slit onto the polished platinum source holder. The sources showed a slight discoloration of the platinum due either to sublimed tantalum or to solid impurities deposited on the filament with the curium.

EXPERIMENTAL PROCEDURE

In order to carry out the experiment as outlined above, it was necessary to check on the validity of certain assumptions and to determine the relative errors of a number of parameters. These included measurements in the variations of B and ρ, freedom from gas scattering for both the alpha particles and the singly charged positive ions, temperature variations of the precision voltage divider and standard cell, and frequency drifts of the nuclear magnetic resonance field control circuit.

Variations in the magnetic field strength along lines normal to the trajectory were measured and found to be within one part in 10^5 per centimeter. Thus the magnet was shown to be good for general alpha spectrometry, requiring only relative measurements for nuclear energy level determinations. Variations in magnetic field along the median radius and changes in these variations were not, however, insignificant and for very large excursions in magnetic field, ($\sim$4000 gauss), the reproducibility of B values was only 1 part in 1400. This, of course, is to be expected with a magnet of such a large pole-face area when the entire magnet is regulated from a single point and single probe as shown in the figure. Thus, during the course of the entire experiment, approximately one month, the magnet was continually energized at a field strength whose variations were restricted to a few gauss. The dimensional changes in the source-to-detector distance, 2ρ, were minimized by temperature control of the room within 2° centigrade. Mean variations of temperature in the iron yoke itself were probably closer to $\frac{1}{2}$° centigrade. The mean source-to-detector distance was measured and set to 152.40 cm by micrometer adjustments of source and detector slits. The actual value of the radius is, of course, cancelled out of the equations in the ion comparison method of determining absolute energy.

A frequency meter, which monitored the nuclear magnetic resonance oscillator was calibrated against radio station WWV and found to be accurate within 3 parts in 10^7. Any drifting of the oscillator was measured with the frequency meter. Thus the magnetic field strength was monitored for each specific point of the Po^{210} alpha line shown in Fig. 2.

The general procedure of the work can be summarized as follows. The Po^{210} source was mounted as shown in Fig. 1 in front of a 0.15-mm collimating slit; the detector slit was also set at 0.15 mm. The baffle restricted the half-angle of divergence to 0.013 radian, giving a transmission of 1 part in 50 000. The magnetic field was then varied by changing the nuclear magnetic resonance oscillator frequency at a linear rate in order to scan the alpha-particle peak and to determine the approximate position of the "peak" maximum. The magnetic field was then set corresponding to a number of selected frequencies on both sides of the peak maximum and the curve shown in Fig. 2 was obtained.

It was desirable to keep the radioactive source thin in order to obtain a narrow line shape. This resulted in low counting rates at the detector and made it necessary

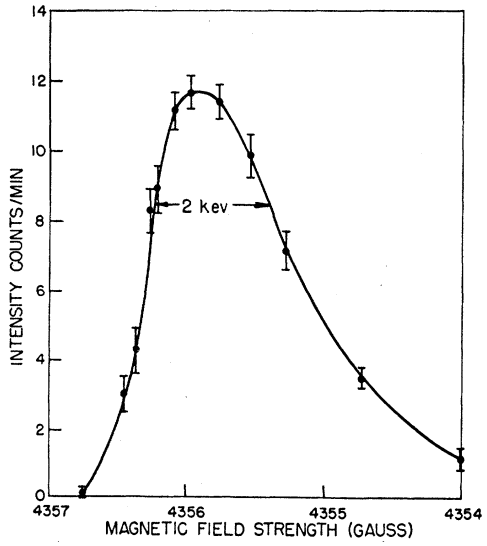

FIG. 2. Intensity of alpha particles from Po^{210} as a function of magnetic field strength.

to reduce noise pulses to a minimum. The use of the coincidence counter eliminated background alpha pulses but did not completely eliminate all noise despite excellent shielding. Thus most of the low-level counting was done at night when radiated signals were at a minimum.

The magnetic field was then precisely maintained at the value corresponding to the maximum intensity of alpha particles as illustrated in Fig. 2 and a mass spectrometer surface ionization source was inserted.

Lu^{175} was selected as the singly charged positive ion for the energy comparison. The selection was made for two primary reasons. First, for a magnetic field in the neighborhood of 4000 gauss Eq. (1) requires an ion of high mass because of limited accelerating voltages which can be attained with our available source voltage supply. Secondly, a copious emission of ions at low temperature and low filament current was desired in order that the IR drop of the source filament could be minimized.

Currents of 10^{-12} ampere were needed as an electron multiplier could not be shielded magnetically at the detector focal point. Previous mass spectrometric experience at this Laboratory indicated that large beam currents of the metallic ion of lutetium could be obtained by surface ionization.

An additional mercury diffusion pump was added to the spectrometer source to prevent voltage fluctuations due to arcing. The accelerating voltage was then varied to locate the position of the Lu^{175} peak maximum. At this position a large number of readings of the voltage, V_1, across the low-resistance section of the precision voltage divider were made on a Leeds and Northrup potentiometer which related the voltage divider to the standard cell.

The Po^{210} sample was placed in the source position after the completion of the Lu^{175} experiment and the above measurements repeated.

Temperature changes of the voltage divider under current load were measured and shown to yield a negligible change in resistance. The total resistance imbalance of the two resistor groups was measured both in air and in the column of insulating transformer oil. This check was made to determine any current by-passing the resistor string due to conductivity of the oil. The value of the precision divider ratio, R, was measured daily over a period of one month to determine the amount of secular change in the precision resistors.

RESULTS

The energy of the Po^{210} alpha particle is calculated from the equation

$$E=4\frac{m_1}{m_0}(RV_1+V_1)\left(1-\frac{E}{2m_0c^2}\right),$$

where m_1, the mass of the Lu^{175} ion, is taken to be 174.995 and m_0, the rest mass of the alpha particle, is taken to be 4.0028, the mass of the helium atom (4.00387) minus the mass of two electrons. Based on present mass systematics, there is little likelihood that the above assigned mass of Lu^{175} could be in error by more than 0.003 mass unit. The value of R is determined from

$$R=25\,600\ (1+d),$$

where

$$d=(R_p/R_s)-1,$$

R_p being the parallel resistance of the forty 2-megohm resistors and R_s the series resistance of the four 12 500-ohm resistors. The value of d was determined by the interchange bridge method over a period of one month, and was found to be -0.000052 ± 0.00002, yielding a value for R of $25\,598.7\pm0.5$. This error is the total range of the observed measurements of d.

The value of V_1 was found from the potentiometer measurement which was related to the standard cell. The standard cell was calibrated before and after the experiment and the measured value of 1.01906 absolute volts at 25°C was reproduced within 10 microvolts. The mean value of V_1 obtained from the two precision potentiometers was found to be 1.18598 ± 0.00007 absolute volts. The error quoted in V_1 is composed of the average deviation of the measurements plus the uncertainty in the potential of the standard cell used as the reference and the error associated with the precision potentiometer. The average deviation of the measurements taken on the Lu^{175} ion peak is caused principally by the uncertainty in locating the peak maximum. The half-width of the Lu^{175} ion beam was about one part in 10 000 of the ion energy.

When one uses the above values, the calculated energy of the Po^{210} alpha particle is 5.3054 Mev. The major source of error in this experiment is attributed to the uncertainty in locating the top of the Po^{210} alpha peak. The curve shown in Fig. 2 shows the relative counting rate of the Po^{210} alpha particles plotted against magnetic field strength. The half-width value as shown by the curve corresponds to about 2 kev in alpha-particle energy. After the measurements on the Lu^{175} ions were completed, this curve was again obtained and in both cases the position of maximum intensity was located within 1 part in 40 000 of the magnet field strength. The magnetic field strength producing the maximum intensity of alpha particles remained constant to within one part in 40 000 before and after the Lu^{175} experiment. Nevertheless, in selecting a best value for the magnetic field strength with which to compare the Lu^{175} ion energy, an error of one part in 10 000 or approximately one quarter of the half-width of the alpha-particle peak is a more reliable estimate of the uncertainty involved. Since the magnetic field was constantly monitored and controlled during the entire experiment, the selected mean value of B, the magnetic field strength, for both the alpha particles and the Lu^{175} ions may be considered invariant

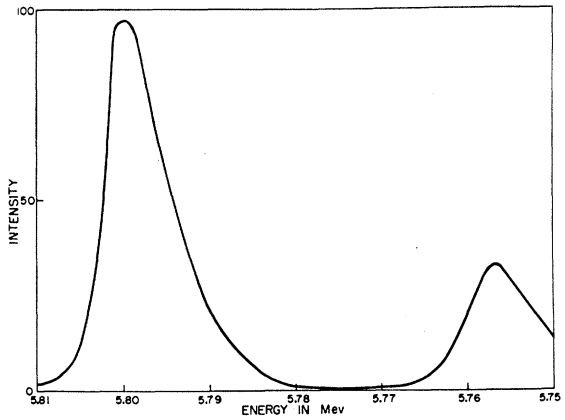

FIG. 3. Alpha spectrum of Cm^{244}.

within one part in 40 000, corresponding to an energy uncertainty of 0.25 kev for the Po^{210} alpha particles. With all of the above errors taken into consideration, the value obtained for the Po^{210} alpha particle energy is given as 5.3054±0.0010 Mev (absolute volts).

This value is in agreement with the absolute energy determination of Po^{210} alpha particles by Collins, McKenzie, and Ramm.[3] From the $B\rho$ measurements of the above authors, one calculates a Po^{210} alpha-particle energy of 5.3043±0.0029 Mev. Briggs,[6] in his review of the energies of natural alpha particles in 1954 in which he calculated a mean $B\rho$ value for Po^{210} alpha particles, assigned the greatest weighting factor to the $B\rho$ value of Collins *et al.*, the one most recently measured. The classical $B\rho$ measurements of the Po^{210} alpha particles made at the Cavendish Laboratory[2] and in Paris[1] give a lower value, 5.299 Mev, for the Po^{210} alpha particles, although the error in these measurements is somewhat uncertain and may be large enough to give agreement with the present results. Sturm and Johnson[4] obtained a Po^{210} alpha particle energy of 5.298±0.005 Mev by using 90° electrostatic deflection and a $Li^7(p,n)Be^7$ threshold energy of 1.882 Mev. Wapstra,[7] in evaluating the $B\rho$ data of several authors, preferred the older measurements of the energy of the Po^{210} alpha particles, based on the consistency of several $B\rho$ ratios, and as a result he chose a Po^{210} alpha-particle energy of 5.3009 ±0.0020 Mev. With additional mass spectrometric measurements, it may be possible by comparing these data with nuclear reaction energy data to give a better evaluation of the different absolute energy determinations of the Po^{210} alpha particles.

In order to provide a secondary standard, a comparison was made between the energy value (α_0) of the alpha particles from Cm^{244} leading to the ground state of Pu^{240}, and the energy of the Po^{210} alpha particles. For this measurement, a nuclear emulsion was used to record the dispersion of the two alpha groups. The Cm^{244} to Po^{210} ratio of alpha particle energies was found to be 1.0937. The principal source of error in this relative measurement was the uncertainty in locating the distance between the centers of the Po^{210} alpha peak and the Cm^{244} alpha peak. Examination of the photographic plate showed the separation of the two alpha-particle peaks to be 68.4±0.1 mm. Since the dispersion of the magnetic analyzer is 0.13 mm/kev in this region of alpha energies, the error quoted is equivalent to less than one kev in the energy difference between the Po^{210} and the Cm^{244}. Repeated exposures of the Po^{210} and the Cm^{244} alpha particles all gave results consistent with the above. Thus the Cm^{244} α_0 energy is calculated to be 5.8025±0.002 Mev. This may be compared with the value 5.798 Mev previously obtained by Asaro, Thompson, and Perlman.[8]

In order to obtain the energy difference between the Cm^{244} to Pu^{240} ground-state transition α_0, and the transition α_1 to the first excited level of Pu^{240}, the magnetic shim was inserted in the pole-face gap and a larger acceptance angle was utilized. The curve shown in Fig. 3 was obtained by alpha-particle track counting and shows the resolution obtainable when a half-angle of divergence of approximately 5 degrees is utilized. The energy difference of the two Cm alpha particles, α_0 and α_1, was found to be 43.5±1 kev. This value (when adjusted for Pu^{240} recoil) is in reasonably good agreement with the more precise determination of 42.88 ±0.05 kev obtained by Smith and Hollander[9] for the energy of the first excited level of Pu^{240}.

[6] G. H. Briggs, Revs. Modern Phys. **26**, 1 (1954).
[7] A. H. Wapstra, Physica **21**, 367 (1955).
[8] Asaro, Thompson, and Perlman, Phys. Rev. **92**, 694 (1953).
[9] W. G. Smith and J. M. Hollander, Phys. Rev. **101**, 746 (1956).