Comparison of Nuclear and Gamma-Ray Energy Scales*†

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(Received February 2, 1954)

Cylindrical and spherical electrostatic analyzers have been used to measure the ratio of the threshold proton energy for the $Li^7(p,n)Be^7$ reaction to the Q of the $Mg^{24}(p,p')Mg^{24*}$ reaction. This ratio was found to be 1.3734 ± 0.0007. The energy of the gamma ray from Mg^{24*} has been compared earlier by Hedgran and Lind to other gamma rays whose energies have been measured absolutely by either the curved crystal spectrometer method (Au^{198}-Caltech) or proton-moment calibrated magnetic field (Co^{60}-Lindstrom *et al.*). The present data fix the $Li(p,n)$ threshold as 1881.4 ± 1.1 kev in terms of the Caltech value for the Au^{198} gamma, or as 1879.7 ± 1.1 kev in terms of the Lindstrom *et al.* value for a Co^{60} gamma ray. The former value is in better agreement with currently used nuclear energy scales.

INTRODUCTION

NUCLEAR reaction energies are usually measured relative to certain easily reproduced calibration values such as the threshold proton energy of the $Li^7(p,n)Be^7$ reaction or one of the sharp $Al^{27}(p,\gamma)$ or $F^{19}(p,\alpha\gamma)$ resonances. Various of these calibration energies have been measured absolutely by electrostatic deflection of the charged incident particle[1] and by a radio-frequency speed gauge.[2] The estimated precision of these absolute measurements was in both cases approximately 0.1 percent. For some calibration purposes[3] alpha particles from naturally radioactive sources are preferred. These alpha energies have been measured absolutely by magnetic deflection and in some cases accuracies of 0.007 percent claimed.[4] Sturm and Johnson[5] have shown that Herb's scale and the old magnetic deflection measurements agree within 0.05 percent. Collins, McKenzie, and Ramm[6] have used a proton-moment calibrated magnetic field to remeasure some of the naturally radioactive alphas and also nuclear reaction Q's. Their energies are somewhat systematically higher than the old magnetic deflection measurements. Recently Famularo and Phillips[7] have measured nuclear reaction energies also in terms of the proton-moment calibrated field and find no systematic disagreement with earlier energy scales.

An attempt was made by Williamson *et al.*[8] to use the first excited state of Li^7 to intercompare the nuclear scale with the gamma-ray scale based upon DuMond's crystal spectrometer. Unfortunately the errors of comparison were at that time too large to display the 0.14 percent systematic error since discovered[9] in the original direct crystal measurement of the gamma ray from Au^{198}.

A more precise intercomparison of the nuclear and gamma scales might be of great help in suggesting the presence or absence of other systematic errors in one or both energy scales.

The recent precise comparison[10,11] of a Na^{24} gamma ray to other gammas whose energies are absolutely determined, makes practical another intercomparison of the nuclear reaction and gamma-ray scales. The gamma rays following the beta decay of Na^{24} correspond to transitions between states of a stable isotope. Therefore the location of these excited states can be readily examined by inelastic scattering of protons from Mg^{24}.

An earlier communication[12] from this laboratory reported measurement of the first excited state of Mg^{24} by inelastic scattering and also by the $Al^{27}(p,\alpha)Mg^{24}$ reaction. No inconsistency between the nuclear and gamma-ray scales was evidenced. The present measurements are an attempt to improve significantly the precision of the intercomparison.

Since the $Li^7(p,n)Be^7$ threshold is perhaps the most convenient and widely used nuclear calibration reaction, we attempt in the present work to determine accurately the ratio R of the proton energy for the $Li^7(p,n)Be^7$ threshold to the energy of the first excited state of Mg^{24}. The latter can then be easily related to the gamma-ray scale by the measurements of Hedgran and Lind.[10,11]

EXPERIMENTAL PROCEDURE

A cylindrical electrostatic analyzer[13] was used to select monoergic (±0.06 percent) incident protons. The energy of the incident protons T_1 is proportional

* Work supported in part by the Wisconsin Alumni Research Foundation and the U. S. Atomic Energy Commission.

† Preliminary report was given at the Washington Physical Society meeting [Phys. Rev. **91**, 482 (1953)].

[1] Herb, Snowdon, and Sala, Phys. Rev. **75**, 246 (1949).
[2] Shoupp, Jennings, and Jones, Phys. Rev. **76**, 502 (1949).
[3] E.g., Strait, Van Patter, Buechner, and Sperduto, Phys. Rev. **81**, 747 (1951).
[4] G. H. Briggs, Proc. Roy. Soc. (London) **157**, 183 (1936).
[5] W. J. Sturm and V. Johnson, Phys. Rev. **83**, 542 (1951).
[6] Collins, McKenzie, and Ramm, Proc. Roy. Soc. (London) **216**, 242 (1953).
[7] Famularo and Phillips, Phys. Rev. **91**, 1195 (1953).
[8] Williamson, Browne, Craig, and Donahue, Phys. Rev. **84**, 731 (1951).
[9] Muller, Hoyt, Klein, and DuMond, Phys. Rev. **88**, 775 (1952).
[10] D. Lind and A. Hedgran, Arkiv Fysik **5**, 29 (1952).
[11] A. Hedgran and D. Lind, Arkiv Fysik **5**, 177 (1952).
[12] Donahue, Jones, McEllistrem, and Richards, Phys. Rev. **89**, 824 (1953).
[13] Warren, Powell, and Herb, Rev. Sci. Instr. **18**, 559 (1947).

to $P_1/(1-\nu_1)$, where P_1 is the cylindrical analyzer potentiometer setting and $\nu_1=T_1/(2M_pc^2)$ is the first-order relativistic correction for the analyzer (see appendix of reference 1).

A spherical electrostatic analyzer[14] with angular opening of less than $\frac{1}{2}$ degree was used to observe inelastically scattered protons at an angle of 134°21′ with respect to the incident beam. The energy of the scattered particles, T_2, is proportional to $P_2/(1-\nu_2)$, where P_2 is the spherical analyzer potentiometer setting and $\nu_2=T_2/(2M_pc^2)$. (The relativistic correction for the spherical analyzer has the same form as the cylindrical, since in each case the mean orbit is a circle.) For the determination of the *ratio* R of the $Li^7(p,n)Be^7$ threshold proton energy to energy of the first excited state of Mg^{24}, it is not necessary to determine the proportionality constants for the two analyzers separately, since it is only the *ratio* of these two proportionality constants which is involved. This last ratio was found by using incident particles (selected by the cylindrical analyzer) to bombard platinum targets and observing with the spherical analyzer the high-energy cutoff of the elastic scattering.

Systematic errors arising from possible nonlinearity of the analyzers were minimized by the use of an incident *diatomic* beam for the intercalibration of the analyzers. If similar *cylindrical* analyzer potentiometer settings are used for the intercalibration (i.e., 0.65240) as for the $Li(p,n)$ threshold energy (~0.6529), then the calibrating protons after scattering from Pt require a *spherical* analyzer potentiometer setting (i.e., 0.4604) not much different (e.g., ~0.4108) from that for the final data on the protons scattered inelastically from Mg^{24}. Intercalibration platinum edges (again with incident diatomic beams) also were taken for the cylindrical analyzer potentiometer settings corresponding to the energy of the resonance in $Al^{25}(E_p=2.41$ Mev). No systematic change in calibration ratio for these different potentiometer settings could be detected.

These platinum intercalibration data were taken immediately before and after each set of data on Mg^{24} inelastic scattering and immediately before and after each $Li^7(p,n)Be^7$ threshold. During the total course of the measurements random changes of this calibration ratio were observed but the platinum data before and after each reaction measurement rarely differed by more than ~0.02 percent so that the correction for drift of analyzer constants is quite small.

Three different Mg targets were used. Two of enriched Mg^{24} were prepared by evaporation of $Mg^{24}O$ from a tantalum boat upon 1000A and 5000A nickel foils.

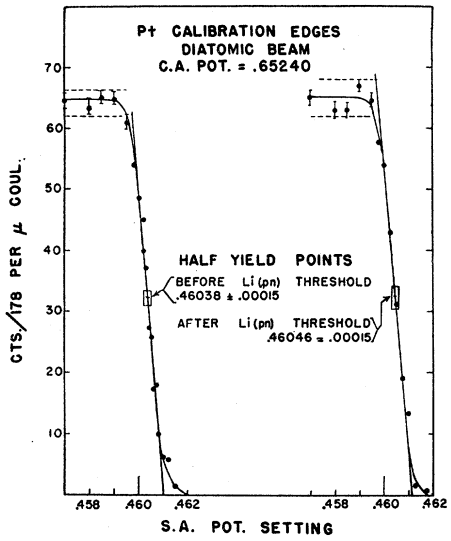

FIG. 2. Typical data for the intercalibration of the cylindrical analyzer (C.A.) and spherical analyzer (S.A.). A diatomic hydrogen ion beam, selected in energy by the C.A., bombarded a thick Pt target, and the energies of the elastically scattered protons at ~135° were measured by the S.A.

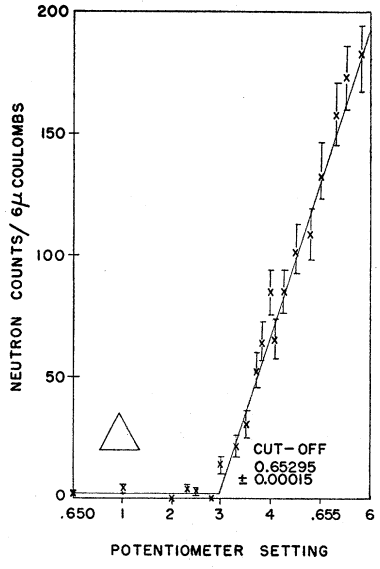

FIG. 1. Typical data on the $Li^7(p,n)Be^7$ threshold.

This method presumably gives metal rather than oxide targets,[15] but observation of the elastically scattered protons did show the presence of some oxygen (and also carbon) throughout the target. Although the concentration of these atoms increased somewhat toward the front surface of the target, no contamination buildup was detected during the course of the experiments. The other target was of ordinary evaporated Mg metal (77 percent Mg^{24}). Thin lithium targets were also evaporated on other 1000A nickel foils and were used for the $Li^7(p,n)Be^7$ threshold determinations. A BF_3 counter in paraffin was used as neutron detector. All targets were kept heated to 200°C to minimize contamination buildup. Two triple collision baffles,

[14] Browne, Craig, and Williamson, Rev. Sci. Instr. 22, 952 (1951).

[15] Russell, Taylor, and Cooper, Rev. Sci. Instr. 23, 764 (1952).

one water-cooled and the other liquid air-cooled, separated the diffusion pump from the system.

To get sufficient intensity of the protons scattered inelastically by Mg^{24}, it was advantageous to set the incident proton energy to correspond to the known resonance[16] at $E_p=2.41$ Mev. This resonance is only $\sim$300 ev wide and hence modifies appreciably the line shape as seen by the spherical analyzer. This modification is discussed below in the analysis of the results.

Typical data including a platinum intercalibration edge before and after the run are shown in Figs. 1, 2, and 3.

METHOD OF DATA ANALYSIS

The ratio R of the proton energy for the $Li^7(p,n)Be^7$ threshold to the energy of the first excited state of Mg^{24} can be expressed explicitly in terms of potentiometer settings for the two analyzers. The energy of the first excited state is just the $-Q$ for the $Mg^{24}(p,p')Mg^{24*}$ reaction and hence equals $T_1-T_2-T_3$, where the T's are the energies of incoming, outgoing, and residual nuclei, respectively. T_1 and T_2 are of course immediately expressible in terms of the cylindrical and spherical analyzer potentiometer settings, e.g.,

$$T_1=\frac{P_1}{1-\nu_1}k_c=P_1'k_c \quad \text{and} \quad T_2=\frac{P_2}{1-\nu_2}k_s=P_2'k_s,$$

where the unprimed P's refer to the actual potentiometer settings, and the primed P's are corrected for analyzer first-order relativistic effects; k_c or k_s is the energy proportionality constant for the cylindrical or spherical analyzer. Conservation of momentum permits T_3 to be expressed also in terms of cylindrical and spherical analyzer potentiometer settings since, for inelastic scattering,

$$T_3=\frac{M_1}{M_3}\left[T_1+T_2-2\left(T_1T_2\right)^{\frac{1}{2}}\cos\theta\right]+V,$$

where V is a small relativistic correction[17] which for the present case is only 0.00022 Mev; M_1 and M_3 are the masses of proton and Mg^{24}, respectively. Therefore, when values for the masses[18] and the $\cos\theta$ are substituted,

$$R=\frac{P_{\mathrm{Li}}'k_c}{1.04199P_2'k_s-0.958007P_1'k_c-0.058672(P_2'P_1'k_sk_c)^{\frac{1}{2}}+0.00022\ \mathrm{Mev}}.$$

Intercalibration of the two analyzers via the platinum scattered edges gives the ratio k_s/k_c, which is the only other constant necessary for the calculation of R from the data. (An approximate[19] value of k_c is of course necessary for the calculation of the small relativistic correction V.) If 0.98259 is taken as the relativistically correct ratio of scattered to incident energy when a proton is scattered elastically from platinum at $\theta=134°21'$, and if correction is made for the energy carried by the electron of the incident diatomic hydrogen ion, one can easily show that

$$k_s/k_c=0.491161P_{cd}'/P_{sp}'.$$

The P_{cd}' comes from the cylindrical analyzer potentiometer setting for the incident diatomic beam and P_{sp}' comes from the spherical analyzer potentiometer setting for the half-yield point on the high-energy cutoff for protons scattered from a thick platinum target (see Fig. 2).

P_{Li}, the cylindrical analyzer potentiometer setting corresponding to the $Li^7(p,n)Be^7$ threshold, is chosen as the linear extrapolated cutoff similar to the method of Herb, Snowdon, and Sala.[1]

The choice of the spherical analyzer potentiometer setting (P_2) appropriate to the energy of the protons scattered inelastically from Mg^{24} is more difficult because the resonant character of the scattering cross section invalidates earlier line-shape analysis[20] which showed that the half-yield point is the significant potentiometer setting when the reaction cross section is constant. To find P_2 one can, for this narrow resonance

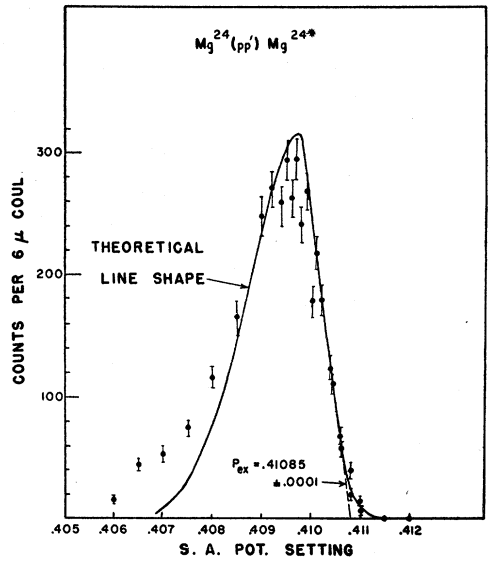

FIG. 3. Typical data on protons scattered inelastically by Mg^{24}. The theoretical line shape is discussed in the text and in Fig. 4.

[16] Mooring, Koester, Goldberg, Saxon, and Kaufmann, Phys. Rev. 84, 703 (1951).

[17] Craig, Donahue, and Jones, Phys. Rev. 88, 811 (1952).

[18] Li, Whaling, Fowler, and Lauritsen, Phys. Rev. 83, 517 (1951); C. W. Li, Phys. Rev. 88, 1038 (1952).

[19] k_c actually is known to better than 0.1 percent.

[20] R. M. Williamson, Ph.D. thesis, University of Wisconsin, 1951 (unpublished).

(~300 ev), treat the cross section for inelastically scattered protons as a δ function. Because of the finite spread in energy of the incoming beam, there will be some protons which after losing energy in the target arrive at the resonant energy. If one knows the energy distribution of the incoming beam, and the energy acceptance window of the spherical analyzer, then one can easily compute the expected line shape and the point on the curve which corresponds to the energy of the inelastically scattered protons. Graphical integration for the measured analyzer windows, etc., gives the line shape expected (see Fig. 4) and indicates that P_2 is $\sim 0.165 \pm 0.017$ percent less than the extrapolated cutoff. The error in this correction to the extrapolated cutoff was estimated by performing graphical integrations for reasonable deviations of analyzer windows from measured values and for slight deviations of the mean bombarding energy from the resonant energy.

P_1, which is proportional to the resonant energy for the inelastic scattering, is fixed approximately by varying the incident energy until the peak yield in the spherical analyzer is observed. A more sensitive test of the deviation of the mean energy from the resonant energy may be found by comparing the total area under the inelastic proton curve (e.g., Fig. 3) with the total area under a similar thick target curve when the mean incident energy is above the resonant energy by an amount large compared to the width of the resonance and the cylindrical analyzer resolution. If the incident energy is exactly at the resonant energy, the area should be precisely one-half that of the case where the mean incident energy is enough above resonance that all incident protons pass through the resonant energy while traversing the target.

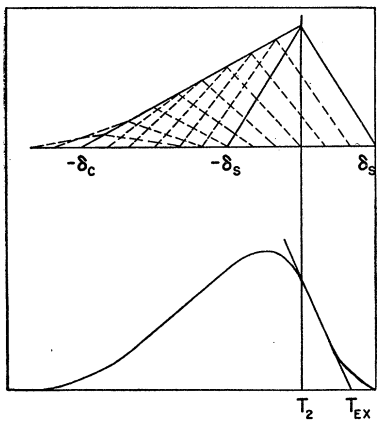

FIG. 4. Graphical integration for the theoretical line shape of the inelastically scattered protons. The incident proton energy T_1 is assumed equal to the energy of the very narrow resonance for inelastic scattering. The spherical analyzer "window" is $T_2 \times \delta_s$. The spread in T_2 arising from the finite energy spread of the incident beam is $\delta_c T_2$ and is equal to $(\Delta_c T_1) \times$(ratio of stopping cross sections of outgoing and incident protons). The cylindrical analyzer resolution is Δ_c.

Nonuniformities in target composition (particularly carbon and oxygen) give the chief systematic uncertainty to this method of finding P_1. The uncertainty in P_1 is estimated to be ~0.015 percent from this cause and from the fluctuations of the areas of different data curves such as shown in Fig. 3.

RESULTS

The mean value of the eight $Li^7(p,n)Be^7$ threshold determinations was $P_{Li}' = P_{Li}/(1-\nu_{Li}) = 0.65354 \pm 0.00012$, where the error is the standard deviation of the eight runs. The nine $Mg^{24}(p,p')Mg^{24}$ edges gave a mean $P_2' = P_2/(1-\nu_2) = 0.41033 \pm 0.00016$, the error including both the standard deviation and the 10 percent uncertainty in the correction to the extrapolated cutoff (see preceding). The calibration ratio k_s/k_c based upon 26 platinum edges was 0.69598 ± 0.00024. The potentiometer setting for excitation of the scattering resonance, $P_1' = P_1/(1-\nu_1)$, was taken as 0.83737 ± 0.00012. This last error estimate has already been discussed in the preceding paragraph.

A possible systematic error is that introduced by uncertainty in the angle of observation θ. This angle was measured to be $134°21 \pm 5'$. The angle measurement was achieved by comparing the energy of the alphas elastically scattered by a carbon target to those elastically scattered by a platinum target. The angle sensitivity of the former scattering process is such that this way of determining the mean angle is preferred to direct geometrical measurement. This method has, however, been earlier checked by direct measurements.[14] For the present experiment $\partial Q/\partial\theta = 0.08$ Mev/radian and hence the uncertainty from this source is only 0.009 percent.

The preceding input values give for the ratio R of the $Li^7(p,n)Be^7$ threshold to the energy of the first excited state of Mg^{24}:

$$R = \frac{E_{\text{thres}}[\text{Li}(p,n)\text{Be}^7]}{E_x(\text{Mg}^{24*})} = 1.3734 \pm 0.0007.$$

$E_x(Mg^{24*})$ from Gamma-Ray Data

The latest result of the Caltech group[9] for the absolute energy of the gamma ray following the beta decay of Au^{198} is

$$E_\gamma(\text{Au}^{198}) = 411.770 \pm 0.036 \text{ kev}.$$

Lind and Hedgran[10] and Hedgran and Lind[11] report the following ratios for momenta of the photoelectrons from various relevant gamma rays:

$$\frac{H\rho(\text{Au}^{198}(U_{LI}))}{H\rho(\text{Co}^{60}(U_K))} = 0.44924 \pm 0.00010,$$

$$\frac{H\rho(\text{Na}^{24},\ 1.37 \text{ Mev})}{H\rho(\text{Co}^{60},\ 1.33 \text{ Mev})} = 1.0229 \pm 0.0002.$$

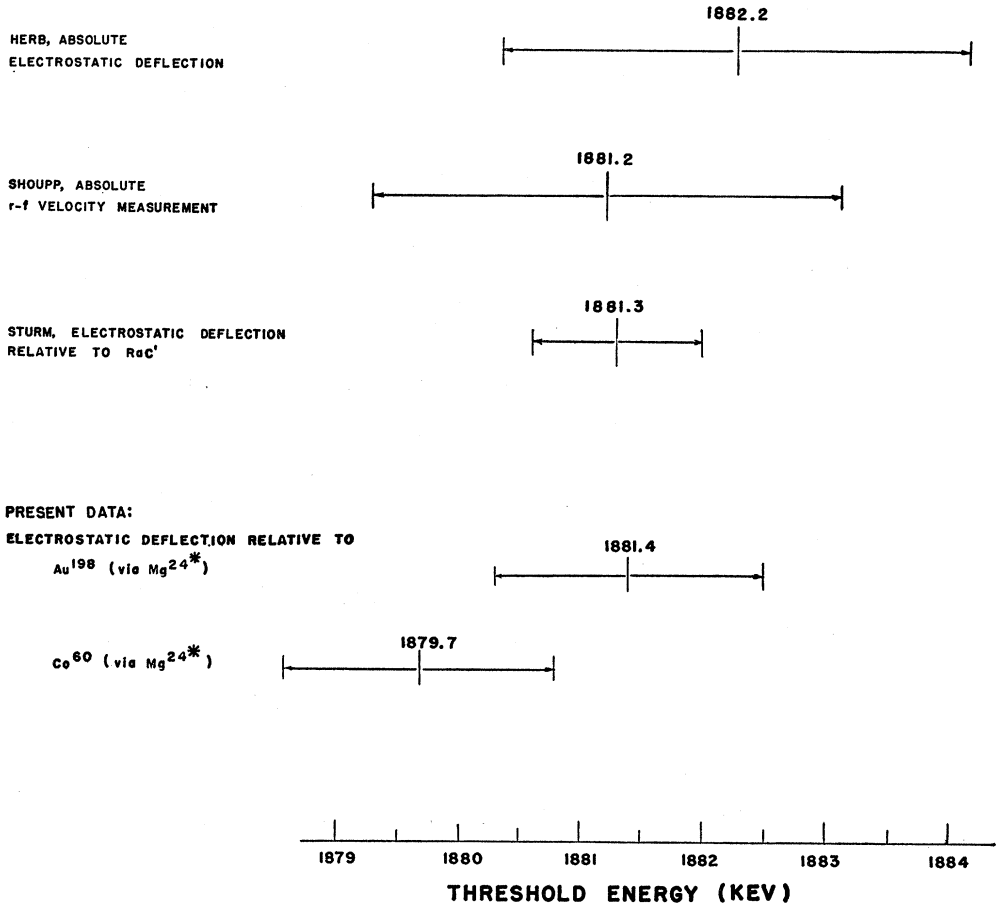

FIG. 5. Comparison of various independent determinations of the $Li^7(p,n)Be^7$ threshold.

When the binding energies of the U_K and U_{LI} electrons are taken as 115.62 kev and 21.76 kev,[21] respectively, the above data yield a value of 1369.91 $\pm$0.40 kev for the Na^{24} gamma-ray energy. When the energy of the recoiling Mg^{24} is included, $(h\nu)^2/(2M_{24}c^2)$, the energy of the first excited state of Mg^{24}, is

$$1369.95\pm0.40 \text{ kev},$$

based on the Caltech crystal spectrometer scale.

A value of comparable precision for the Mg^{24*} state can be found from other gamma-ray data. Recently Lindstrom, Hedgran, and Alburger[22] report 1332.5 $\pm$0.3 kev for the energy of a Co^{60} gamma ray. This value is obtained by comparison with the 1415.8$\pm$0.2 kev transition in RaC′ which was measured absolutely (in terms of the proton magnetic moment) by the same authors. If Hedgran and Lind's ratio (=1.0229 $\pm$0.0002) for momenta of the photoelectrons from Na^{24} and Co^{60} is used, then the energy of the Na^{24} gamma ray would be 1368.64$\pm$0.45 kev, and when the recoil energy of the Mg^{24} is included, the energy of the Mg^{24} state is 1368.68$\pm$0.45 kev. It will be noted that these two independent gamma-ray measurements differ by appreciably more than their assigned probable errors. Hedgran and Lind's ($H\rho$) ratio for Na^{24} and Co^{60} photoelectrons occurs in the determination of both values, but their $H\rho$ ratio for Au^{198} and Co^{60} photoelectrons occurs only in the former value. So the discrepancy between the two values for the Mg^{24*} could arise from Lind and Hedgran's $H\rho$ ratio[10] of Au^{198} to Co^{60} photoelectrons, or it could indicate a systematic error in one or both of the absolute gamma-ray measurements.

$Li^7(p,n)Be^7$ Threshold

If the abo ve values for the first excited state of Mg^2 are combined with our ratio R of the $Li^7(p,n)Be^7$ threshold to the $-Q[Mg^{24}(p,p')Mg^{24*}]$, then one finds

$$E_{\text{thres}}[Li^7(p,n)Be^7]=1881.4\pm1.1 \text{ kev}$$

[21] V. Cauchois, J. Phys. radium **13**, 113 (1952) but recomputed for latest atomic constants, Jessee W. M. Du Mond, Revs. Modern Phys. **25**, 691 (1953).

[22] Lindstrom, Hedgran, and Alburger, Phys. Rev. **89**, 1303 (1953).

in terms of the Caltech crystal spectrometer scale, but

$$E_{\text{thres}}[\text{Li}^7(p,n)\text{Be}^7] = 1879.7 \pm 1.1 \text{ kev}$$

in terms of Lindstrom's proton-moment calibrated scale.

Graphical comparison of various independent determinations of this threshold are shown in Fig. 5.

A weighted mean of these independent measurements of the $\text{Li}^7(p,n)\text{Be}^7$ threshold is 1881.1 ± 0.5 kev.

CONCLUSIONS

No appreciable systematic differences between the gamma-ray scales and current nuclear-reaction energy scales are indicated by the present work. There is, however, some suggestion of a systematic difference between two current gamma-ray scales. Somewhat better consistency with nuclear reaction data is found for the gamma-ray scale based upon the curved-crystal spectrometer measurements.

PHYSICAL REVIEW VOLUME 94, NUMBER 4 MAY 15, 1954

The Half-Life of U^{232}

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(Received February 5, 1954)

The half-life of U^{232} has been determined to be 73.6 ± 1.0 years by a method involving isotopic dilution, mass spectrometric analysis, and the determination of the specific activity of the diluted sample by conventional weighing and counting techniques.

THE half-life of U^{232} has been reported as 30 years[1] and 70 years.[2] The 30-year value was estimated from the growth of U^{232} alpha activity from its beta-emitting parent Pa^{232} and the 70-year value from the growth of U^{232} from its alpha-emitting parent Pu^{236}. No limits of error have been given for either value, but the 70-year value was based on a later measurement and considered to be more accurate than the 30-year value.

The half-life of U^{232} is sufficiently long that the direct measurement of its decay is difficult. On the other hand, the direct determination of the specific activity is difficult because of the small quantities which are available and is hazardous because of the high specific activity of U^{232}. However, isotopic dilution with a large excess of relatively inactive U^{238} enables one to determine the specific activity of the diluted sample by conventional weighing and counting techniques. A mass spectrometric determination of the ratio U^{238}/U^{232} then permits one to calculate the specific activity of U^{232}.

An aliquot (0.09905 g) of U_3O_8 from a Bureau of Standards sample of natural uranium was accurately weighed and transferred to a preweighed polyethylene bottle containing nitric acid and approximately 100 micrograms of U^{232}. After the oxide was dissolved, the solution was diluted with water to 102.61 g, from which was taken 1.0459 g which in turn was diluted to 101.54 g. From this final solution, about 100-milligram aliquots were weighed, evaporated on platinum plates, and counted in an argon-carbon dioxide counter with a geometry of 50.6 percent and τ of 4×10^{-8} minute. An average of three determinations gave a value of 56 530 disintegrations per minute per 100 mg of solution, with a probable error of 120 disintegrations per minute based on the deviations from the mean, which is a measure of the precision of weighing and counting the samples. This corresponds to a specific activity of the diluted sample of 6.61×10^{10} disintegrations per minute per gram of U^{238}. From the mass spectrometric analysis of the sample [$U^{232}/U^{238} = (1.436 \times 10^{-3}) \pm 1.0$ percent, and $U^{233}/U^{238} = (4.75 \times 10^{-2}) \pm 2.2$ percent, mole ratios] the specific activity of U^{232} was calculated to be 4.65×10^7 disintegrations per minute per microgram. (A mass spectrometric analysis of the original U^{232} sample showed that its U^{238} content was negligible.) This corresponds to a half-life of 73.6 years. The U^{233} was the only isotope other than U^{232} which contributed appreciably (1.5 percent of the total alpha activity) to the activity of the uranium sample, and a correction was applied. A correction was also made for the growth of the daughters of U^{232} by comparing the counting rates of the final samples with that of a sample taken immediately after purification of the U^{232} from its daughters. A consideration of the various errors leads to a probable error of about one year in the half-life.

The authors are grateful to Miss Lillie Mae Porter and Miss Marjorie M. Petheram for their help with the counting.

[1] J. W. Gofman and G. T. Seaborg, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 19.14, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, p. 1427.

[2] James, Florin, Hopkins, Jr., and Ghiorso, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.8, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, p. 1604.