Beta-Spectra and Internal Conversion Coefficients for Co^{60}, Nb^{95}, Au^{198}, and Hf^{181}

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Using a double-lens beta-ray spectrometer, the beta-spectra and the internal conversion coefficients for Co^{60}, Nb^{95}, Au^{198}, and Hf^{181} were investigated. All the beta-spectra were found to be of allowed shape with end-point energies 0.306±0.005 Mev, 0.160±0.003 Mev, 0.97±0.01 Mev, and 0.408±0.008 Mev, respectively. The extra activity presented in Au^{198} was identified to be Au^{199}, and the slow neutron absorption cross section of Au^{198} was estimated to be about 10^5 barns.

The internal conversion coefficients indicate that both of the gamma-rays from Co^{60} are E-2^2 radiations; the 0.77-Mev gamma-ray from Nb^{95} is either an M-2^1 or an E-2^2 radiation or a mixture of both; the 0.411-Mev gamma-ray from Au^{198} is an E-2^2 radiation; the 0.474-Mev gamma-ray from Hf^{181} is an E-2^2 radiation, the 0.134-Mev and 0.340-Mev gamma-rays are M-2^1 radiations, and the 0.130-Mev gamma-ray is doubtful but seems to be an E-2^2 radiation.

The results are discussed from the point of view of the beta-decay theory and the shell structure model (Mayer, Haxel, Jensen, and Suess).

I. INTRODUCTION

WITHIN the past few years the theory of internal conversion has been worked out in some detail by several investigators, and a complete table of the K-shell internal conversion coefficients was published recently by Rose and his collaborators.[1] The general purpose of such calculations is to evaluate the probability of the transition of K, L, etc., atomic electrons to states of the continuum under the influence of the electromagnetic radiation emitted by the nucleus when an isomeric transition occurs. An essential feature is that the probability of the transition depends upon the intensity of the electromagnetic field in the vicinity of the nucleus where the field decreases with a power of r determined by the multipole order of the radiation. Hence the measurement of the probability of the transition of atomic electrons from different shells affords a method of exploring the multipole order of the transition.

Two selection rules govern the nature of the electromagnetic radiation, from which it becomes possible to draw some conclusions about the nuclear states involved in the transition. The first is the rule that the photon which is emitted from a 2^l electric or magnetic multipole has angular momentum $l\hbar$ with respect to the position of the multipole.[2,3] Thus, if I and I' are the angular momenta in units of $\hbar$ of the initial and final states of the radiating system, then l must have the value

$$|I+I'| \geqslant l \geqslant |I-I'|, \qquad (1)$$

with the lowest value of l most probable.

The second selection rule governs the parity change between initial and final states. Electric 2^l-pole and/or magnetic 2^{l-1}-pole radiation occur only between states of the same parity if l is even, and only between states of opposite parity if l is odd.[2,3]

This paper describes the study of a number of cases in which a nucleus A is transformed by beta-decay into an excited state of a stable nucleus B. The transition from the excited state of B occurs by the emission of an internally converted gamma-ray.

In so far as Fermi's theory of beta-decay applies, it is usually possible to draw some conclusion about the spin and parity changes involved in the beta-ray process from the shape of the beta-spectrum and the ft-value.[4,5] Such studies of the beta-spectrum together with the coefficients of internal conversion provide a check of the theoretical predictions of the spins and parities of nuclei A and B, such as those given by the shell structure model.[6] The present work was undertaken to provide sufficient accuracy in the experimental determination of beta-spectra and internal conversion coefficients so that a significant test of the shell theory could be made.

II. APPARATUS AND CALIBRATION

1. Beta-Ray Spectrometer

All the measurements were carried out by using the double-lens beta-ray spectrometer[7] of this laboratory (Fig. 1). The spectrometer consists of two current carrying coils 95.8 cm apart. The source was placed at the center of one coil and the detector was placed at the center of the other. Electrons from the source were detected only when their trajectories were of the first order,[7] those having trajectories of higher orders being eliminated by a helical baffle at the center of the instrument. The transmission and the resolving power of the instrument were controlled by two iris diaphragms situated in close contact with the ends of the helical baffle. Because of the large dimensions of the instrument, scattering from the side walls was practically absent. Scattering from the helical baffle was eliminated

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[1] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).
[2] W. Heitler, Proc. Cambridge Phil. Soc. **32**, 112 (1936).
[3] E. Segrè and A. C. Helmholz, Revs. Modern Phys. **21**, 271 (1949).
[4] E. Fermi, Z. Physik **88**, 161 (1934).
[5] E. J. Konopinski, Revs. Modern Phys. **15**, 209 (1943).
[6] M. G. Mayer, Phys. Rev. **78**, 16 (1950).
[7] H. Agnew and H. L. Anderson, Rev. Sci. Instr. **20**, 869 (1949).

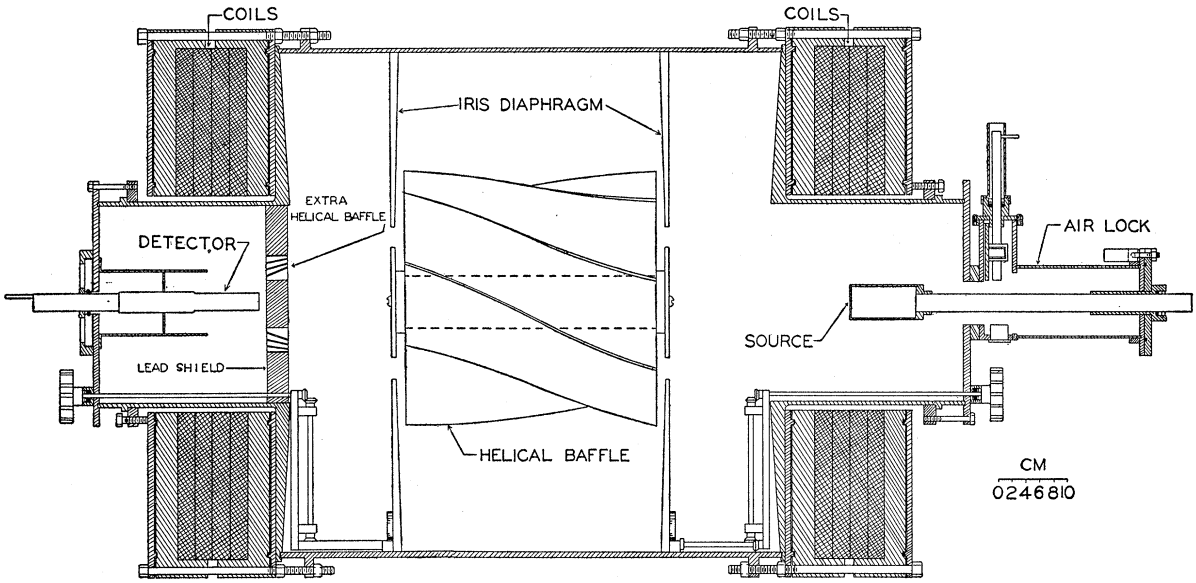

FIG. 1. The beta-ray spectrometer.

by introducing an extra helical baffle in front of the detector. The Compton electrons produced by gamma-rays from the source were reduced to a negligible amount when this device was installed.

2. Calibration

Although the spectrometer makes use of an iron shell to shield the orbits from the earth's magnetic field, its linearity is not noticeably impaired thereby and a single constant $k=I/p$ relates the momentum p with the current in the coil I. This constant was checked by observing the current at which the maxima of the conversion lines of Cs^{137} (624 kev)[8] and of Co^{60} (1163.1 kev and 1323.2 kev)[9] appeared. Values of the constant k from these calibration points were 16.92, 16.91, and 16.90, where p is in units of mc and I is in amperes. The value $k=16.91$ was adopted in the calculations.

The transmission of the spectrometer was determined by a beta-gamma coincidence measurement using a Co^{60} source. With the source and the beta-ray counter in the standard position in the spectrometer, a gamma-ray counter having a 2-mm brass window was placed 2 cm behind the source. The gamma-ray counting rate N_γ, the beta-ray counting rate N_β, and the beta-gamma coincidence rate $N_{\beta\gamma}$ were measured for various values of current in the spectrometer. Co^{60} decays by emitting a beta-ray followed by two gamma-rays in cascade; thus, at any given value of the current,

$$N_{\beta\gamma}/N_\beta=\epsilon_1(1-\alpha_1)+\epsilon_2(1-\alpha_2) \quad (2)$$

and

$$N_\gamma=\epsilon_1(1-\alpha_1)N+\epsilon_2(1-\alpha_2)N, \quad (3)$$

where ϵ_1 and ϵ_2 are the efficiencies of the counter for counting gamma-rays 1 and 2, respectively, α_1, α_2 are the total internal conversion coefficients corresponding to the two gamma-rays, respectively, and N is the total number of beta-disintegrations per second. Here, the possible beta-gamma angular correlation is taken to be zero.[10] The absolute strength of the source is given by

$$N=N_\beta N_\gamma/N_{\beta\gamma}. \quad (4)$$

Using this value and the area A under the momentum spectrum of Co^{60}, which is equal to the product of N, the resolution w, and the transmission t of the instrument [Eq. (6)], the transmission can be determined.

It was found that when the counter position was fixed, the transmission depended very sensitively on the source position. Thus, the position of the source and the control diaphragms were carefully adjusted to get (1) sufficient resolution and (2) the highest possible transmission. For a source 4 mm in diameter, the transmission was 1 percent with a resolution of 2 percent.

3. The Counter

A spherical G-M counter was originally used for the detector. This type of counter was designed particularly for a system in which low energy electrons receive an accelerating voltage at the detector to aid them in the penetration of the thin window.[7] Later, a cylindrical counter was used which had an efficiency which was more nearly independent of the energy of the electrons. Figure 2 shows a sketch of the cylindrical counter. It was constructed of a 4 in. long brass tubing with twenty-two $\frac{1}{16}$ in.$\times\frac{1}{2}$ in. slots distributed around the circumference, and a center wire along the axis of the tubing. Nylon foils, made conducting by evaporating

[8] L. M. Langer and R. D. Moffat, Phys. Rev. **78**, 75 (1950).
[9] Lind, Brown, and DuMond, Phys. Rev. **76**, 591 (1949).
[10] R. L. Garwin, Phys. Rev. **76**, 1876 (1949).

5 $\mu g/cm^2$ of aluminum on them, were used for the window.[11] The thickness of the window ranged between 80 $\mu g/cm^2$ to 100 $\mu g/cm^2$; it stopped electrons of energy below 5 kev.

A mixture of neon and amyl-acetate with a total pressure of 8 cm was generally used for the counter filling. On several occasions, ethyl-acetate at pressure 2.4 cm was used. With the latter, the vapor pressure could be kept constant by dipping liquid ethyl-acetate into ice, and the thickness of the counter window could be reduced to 50 $\mu g/cm^2$ which corresponds to a cut-off energy about 3 kev.[12]

The efficiency of counters was also studied at various pressures. Let m be the number of electrons per molecule and p be the pressure (in cm Hg) of the gas in the counter. It was found that the efficiency of the particular counter used was almost proportional to mp for beta-particles of energy ≈ 0.6 Mev when $mp<70$. For $mp \gtrsim 70$, the efficiency of the counter reached its maximum and remained constant. The value of mp for 8 cm of neon is 80, and it is 115 for 2.4 cm of ethyl-acetate.

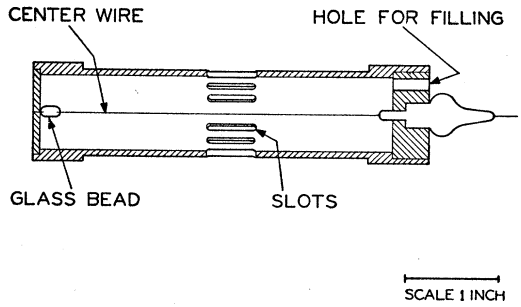

FIG. 2. A sketch of the cylindrical counter.

100 percent efficiency for electrons for the whole range in beta-ray spectrometer work was therefore assumed.

4. Sources

Sources were deposited on 15 $\mu g/cm^2$ Nylon foils having a thin backing of evaporated aluminum. The weight of the sources was determined by controlling the total amount of liquid deposited, and the uniformity was checked by radio-autograph. All the samples reported here were obtained from Oak Ridge National Laboratory except those mentioned specially.

[11] The method used to wrap thin foils on a cylinder was as follows: A wet foil was laid on a rectangular frame of a width slightly smaller than the length of the counter and a length slightly longer than that of the circumference of the counter. Then the counter, whose surface was painted with clear glyptal, was placed at one end of the frame. The foil could then be picked up and glued to the surface when the counter was rolled carefully over the frame. It was necessary to leave enough space on the counter shell without slots for the overlapped ends of the foil.

[12] The ethyl-acetate filling permitted the use of thinner window and more stable gas pressure. However, it was found that the counters with the ethyl-acetate filling had several minor drawbacks. They needed higher applied voltage (1700 v); they produced pulses less uniform in height and having a longer dead time (≈ 100 μsec). The plateau was also shorter, *viz.*, having about 30 volts with a 2 percent slope.

III. EXPERIMENTAL METHOD

1. Momentum Spectrum

By a momentum spectrum is meant the plot of the number of beta-disintegrations $n(p)$ per unit momentum interval per second *vs* the corresponding momentum p of the beta-particles. The quantity $n(p)$ cannot be measured directly because of the finiteness of the resolving power of a beta-ray spectrometer. However, it can be shown that $n(p)$ is related to the counting rate N_c of the detector, the transmission t, and the resolution w of the beta-ray spectrometer by the following approximate equation:

$$n(p)=N_c/pwt=kN_c/Iwt, \tag{5}$$

where k is the constant previously defined. Equation (5) is a good approximation for $0<p<p_{\max}(1-w)$ provided (1) an originally monoenergetic line of momentum p' appears as an isosceles triangle, and (2) the resolution w which is defined as the ratio of the width of the triangle at the half-maximum, $\Delta p'$, to the momentum p' is a constant. A detailed discussion is given in the appendix.

In the present measurement, the assumption (1) was justified by the shapes of single conversion lines measured, and the assumption (2) was justified by the linear relationship found between the coil current I and the magnetic rigidity p as shown in Agnew and Anderson's paper.[7] Since p was proportional to the coil current I, a momentum spectrum was thus obtained by plotting N_c/I *vs* p or $H\rho$. Consequently, the area under the momentum spectrum is

$$A=Nwt, \tag{6}$$

where N is the total number of beta-disintegrations per second.

2. Maximum Energy of a Beta-Spectrum

The maximum energy of a beta-spectrum was determined by means of a Fermi plot. Since Eq. (5) is no longer correct and the counting rate is low for p near $p_{\max}$, experimental points near the end of the beta-spectrum were ignored in curve-fitting according to the method of least squares. The percentage errors in the final results were obtained by the standard statistical method.[13]

3. Internal Conversion Coefficient

The ratio of the number of K-shell electrons ejected per second, N_K, to the number of gamma-quanta emitted per second, N_q, is defined to be the K-shell internal conversion coefficient, α_K, and similar definitions hold for α_L, etc. The ratio of the total number of internal conversion electrons ejected per second, N_e,

[13] H. Margenau and G. Murphy, *The Mathematics of Physics and Chemistry* (D. Van Nostrand Company, Inc., New York, 1943), pp. 487–502.

to N_q is called the total internal conversion coefficient α. However, in older literature, the internal conversion coefficient was defined as the ratio of the internal conversion electrons to the number of radiative transitions N_ν, i.e., $\alpha'=N_e/N_\nu=N_e/(N_q+N_e)$, where the "primed" quantity denotes the older definition. In this paper, the former definition is followed in agreement with the more recent custom.[14]

The measurement of the internal conversion coefficients for a given isotope does not require a knowledge of the true activity of the source if the decay scheme is known. Let N be the total number of beta-disintegrations per second. The area under the curve of the plot of the momentum spectrum is $A=Nwt$ according to Eq. (6) provided that the window absorption of the counter is negligible. The area under the K-shell internal conversion peak in the momentum spectrum is equal to $A_K=\alpha_K'N_\nu wt$. Therefore,

$$\alpha_K'=A_KN/AN_\nu, \tag{7}$$

where the quantity N_ν/N can be determined from the known decay scheme. The value of α_K is thus given by the following expression:

$$\alpha_K=\alpha_K'/(1-\alpha'), \tag{7'}$$

which is used to compare with the theoretical values of Rose *et al.*

A Fermi plot of the momentum spectrum is essential to decide what part of the curve is due to the beta-rays and what part is due to the conversion electrons, and to make the correction at low energies for the cutoff of the counter window. Accordingly, such plots were made in every instance. Where some degree of forbiddenness was revealed in a lack of straightness using the simple Fermi rules, an appropriate correction factor was applied.[5] However, because of the distortions due to the source thickness and the uncertainty of Fermi's theory over the low energy range, a correction curve for the window absorption thus obtained might not be correct. Hence, one-half of the difference of the area under the measured spectrum and that under the corrected spectrum was taken as the possible error of the measurement.

IV. RESULTS

1. Co^{60}

5.3-year Co^{60} decays into an excited state of Ni^{60} by negatron emission, with a maximum energy of 0.31 Mev. Subsequently two gamma-rays of energy 1.1715 Mev and 1.3316 Mev, respectively, are emitted in cascade. The beta-spectrum was found to be of allowed shape down to 0.17 Mev.[15]

The coefficients of internal conversion for Co^{60} were measured first by Deutsch and Siegbahn[16] and then by Waggoner, Moon, and Roberts.[17] Their measurements indicate that the two gamma-rays are both due to electric quadrupole radiations. However, in all these measurements, thick window counters were used. It was considered worth-while to study the same problem with thin window counters.

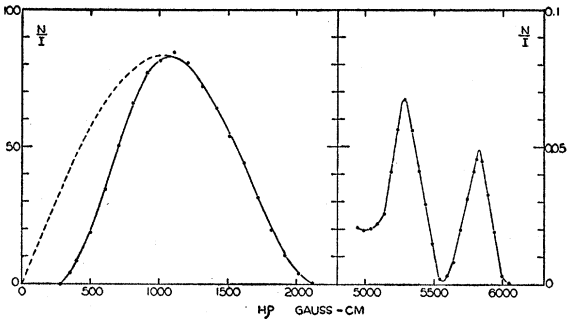

FIG. 3. The beta-spectrum of Co^{60}.

The measured beta-spectrum of Co^{60} is shown in Fig. 3. A 10-microcurie source of 0.2-mg/cm² thickness and 0.5-cm diameter was used. The internal conversion peaks are shown in the same figure with the scale enlarged 1000 times. The rising background on the left side of the first peak is known to be caused by Compton electrons produced in the source. It was found that the beta-spectrum is allowed down to 70 kev with an end-point energy 0.306±0.005 Mev as shown in Fig. 4. The dotted curve in Fig. 3 is the extension of the beta-spectrum corresponding to the extrapolation to zero energy of the straight Fermi plot. The total internal conversion coefficient is found to be $(1.72\pm0.17)\times10^{-4}$ for the 1.17-Mev gamma-ray and $(1.24\pm0.12)\times10^{-4}$ for the 1.33-Mev gamma-ray, where the experimental errors are taken according to the assumption stated in Sec. III. A comparison of the present measurement with those of the other authors is given in Table I, where the experimental errors assumed by the latter are also indicated.

The values of α_K are unknown because the K lines were not resolved from L, etc., lines. However, the

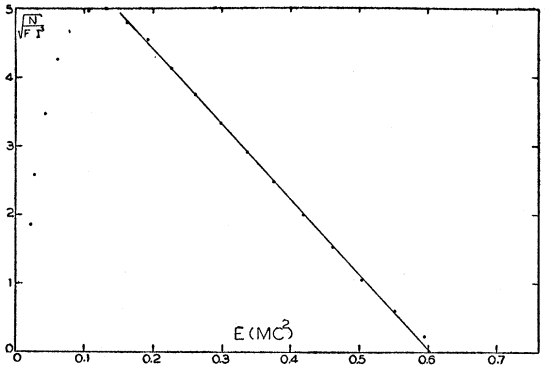

FIG. 4. The Fermi plot for the beta-spectrum of Co^{60}.

[14] See, for example, J. R. Reitz, Phys. Rev. **77**, 10 (1950).
[15] Deutsch, Elliot, and Roberts, Phys. Rev. **68**, 193 (1945).
[16] M. Deutsch and K. Siegbahn, Phys. Rev. **77**, 680 (1950).
[17] Waggoner, Moon, and Roberts, Phys. Rev. **80**, 420 (1950).

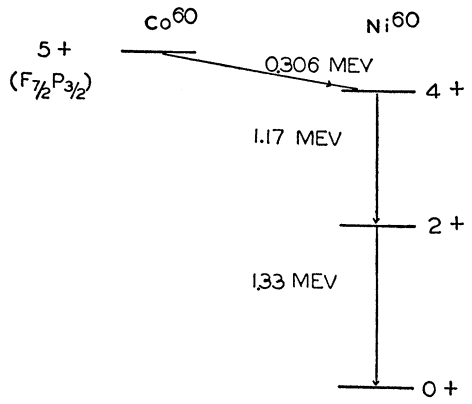

FIG. 5. The decay scheme of Co^{60} and the suggested level values.

values of α_K/α_L can be roughly estimated from Goldhaber and Sunyar's empirical curves;[18] they are high for all multipole radiations. If $\alpha_K/\alpha_L \approx 10$ is assumed for both gamma-rays, $\alpha_K=(1.55\pm0.15)\times10^{-4}$ for the 1.17-Mev gamma-ray and $\alpha_K=(1.12\pm0.11)\times10^{-4}$ for the 1.33-Mev gamma-ray. A comparison of these values with the theoretical ones in Table II[1] permits one to assign both gamma-rays to be more likely E-2^2 radiations. This is in agreement with the angular correlation measurement[19] and the polarization-direction correlation measurement[20] which assigned the states of Ni^{60} involved in the decay of Co^{60} as 0+, 2+, and 4+, respectively. However, the internal conversion measurements alone would not rule out the possibility of M-2^1 or mixed M-2^1 and E-2^2 transitions, if one interprets the experimental uncertainties rather conservatively.

The Fermi plot of the beta-spectrum is straight down to 70 kev. This, together with the absence of beta-gamma angular correlation,[10] allows one to conclude that the transition is allowed, though the ft-value is rather high ($\log ft=7.5$). This means, accepting the assignment 4+ for the state of Ni^{60} for Co^{60} decay, that

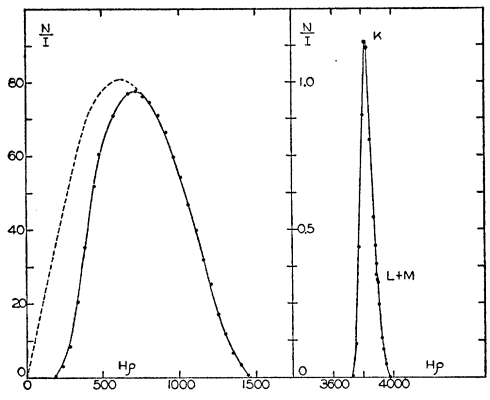

FIG. 6. The beta-spectrum of Nb^{95}.

[18] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[19] E. L. Brady and M. Deutsch, Phys. Rev. **78**, 558 (1950).
[20] F. Metzger and M. Deutsch, Phys. Rev. **78**, 551 (1950).

Co^{60} has a 5+, 4+, or 3+ ground state according to the G-T selection rules. The possibility of a 3+ state can be ruled out by the fact that there is no evidence for transitions from Co^{60} to the lower excited state of Ni^{60}. The experimental evidence at hand does not permit one to decide between the assignments 4+ and 5+. Shell structure model predicts 5+ for Co^{60} [21] and is thus not inconsistent with our results. A tentative conclusion about the level values involved in the decay of Co^{60} is shown in Fig. 5.

TABLE I. Total internal conversion coefficient$\times10^4$.

	1.17 Mev	1.33 Mev
Deutsch and Siegbahn	2.32±0.6	1.83±0.5
Waggoner *et al.*	1.73±0.061	1.286±0.035
The present measurement	1.72±0.17	1.24±0.12

2. Nb^{95}

The decay scheme of 37-day Nb^{95} is simple. The work of Mandeville, Scherb, and Keighton[22] indicates indeed that beta-decay of Nb^{95} into Mo^{95} is followed by a single gamma-ray. The beta-spectrum and the internal conversion coefficient have been determined by Hudgens and Lyon.[23] Figure 6 shows the beta-spectrum of Nb^{95} obtained from the present measurement using a two-microcurie source of 0.05-mg/cm^2 thickness and 0.5-cm diameter. The purity of the source was checked by observing the decay rate over 9 half-lives and was found to be sufficient for the purpose of the experiment. The internal conversion peak is plotted in the same figure on a scale enlarged eighty times. The Fermi plot is seen in Fig. 7 to be straight down to 40 kev. The dotted curve in Fig. 6 is the extrapolation of the beta-spectrum corresponding to a straight Fermi plot down to zero energy. The present results differ in some respects from those of Hudgens and Lyon. A comparison is given in Table III.

TABLE II. K-internal conversion coefficients$\times10^4$.

	E-2^1	E-2^2	E-2^3	M-2^1	M-2^2
1.17 Mev	0.72	1.55	3.01	1.38	2.87
1.33 Mev	0.58	1.17	2.07	1.03	2.07

The L and M conversion line can only barely be resolved from the K conversion line. By comparing the heights of the two tips as indicated in Fig. 6, or by decomposing the conversion line into two isosceles triangles and comparing their area, one obtains as a rough estimate for $\alpha_K/\alpha_L \approx 4$, i.e., $\alpha_K \approx 1.3\times10^{-3}$. The error introduced in measuring the area under the beta-spectrum is about 10 percent.

[21] Mayer, Moszkowski, and Nordheim, Argonne National Laboratory Report No. 4626 (1951).
[22] Mandeville, Scherb, and Keighton, Phys. Rev. **74**, 888 (1948).
[23] J. E. Hudgens, Jr., and W. S. Lyon, Phys. Rev. **75**, 206 (1949).

A comparison of the measured internal conversion coefficient with the theoretical values corresponding to different multipole transitions[1] is given in Table IV. Therefore, the 0.77-Mev gamma-ray is classified as magnetic dipole or electric quadrupole radiation, or a mixture of both.

Nb^{93} has a spin 9/2, and should, according to the shell structure model, be in a $g_{9/2}$ state. This is perhaps also the state of Nb^{95}, because adding two neutrons usually does not change the nuclear configurations.[21]

TABLE III. Results of Nb^{95}.

	Present measurement	Hudgens and Lyon
Beta-ray end-point energy	0.160±0.003 Mev	0.146 Mev
Gamma-ray energy	0.77±0.01 Mev	0.75 Mev
Total internal conversion coeff.	$(1.6\pm0.16)\times10^{-3}$	2.4×10^{-3}

The spin of Mo^{95} is known to be 5/2. Shell structure model predicts $d_{5/2}$.[21] The multipole order of the 0.77-Mev gamma-ray (M-2^1 and/or E-2^2) together with the shape of the beta-spectrum (allowed) and the ft value ($\log ft=5$) would then be consistent with 9/2+ or 7/2+ for the excited state of Mo^{95}. The shell structure favors a $g_{7/2}$ state, which is adopted in the tentative level values shown in Fig. 8.

3. Au^{198}

The beta-spectrum and the internal conversion coefficients of 2.69-day Au^{198} have been the subject of extensive investigations. In some earlier work by Levy and Grueling,[24] several gamma-rays with energies of 0.157, 0.208, and 0.411 Mev and two beta-ray components of maximum energies 0.601 and 0.97 Mev were observed. However, the spectrometric work of Peacock and Wilkinson,[25] Siegbahn and Hedgran,[26] and the latest work of Saxon and Heller[27] show that the beta-spectrum is simple with an end-point energy of 0.97

TABLE IV. K conversion coefficient for Nb^{95}.

Experimental value$\times10^3$	Theoretical values$\times10^3$			
	M-2^1	M-2^2	E-2^1	E-2^2
1.3±0.2	1.32	3.50	0.53	1.31

±0.01 Mev followed by an internally converted gamma-ray of 0.411 Mev. These conflicting results were explained by Hill and Mihelich,[28] who showed that the additional activities observed by Levy and Grueling were due to Au^{199} formed through successive neutron capture during the pile irradiation of gold as follows:

$$Au^{197}+n\rightarrow Au^{198},\quad Au^{198}+n\rightarrow Au^{199}.$$

[24] P. W. Levy and E. Grueling, Phys. Rev. **75**, 819 (1949).
[25] C. L. Peacock and G. Wilkinson, Phys. Rev. **74**, 297 (1948).
[26] K. Siegbahn and A. Hedgran, Phys. Rev. **75**, 523 (1949).
[27] D. Saxon and R. Heller, Phys. Rev. **75**, 909 (1949).
[28] R. D. Hill and J. W. Mihelich, Phys. Rev. **79**, 275 (1950).

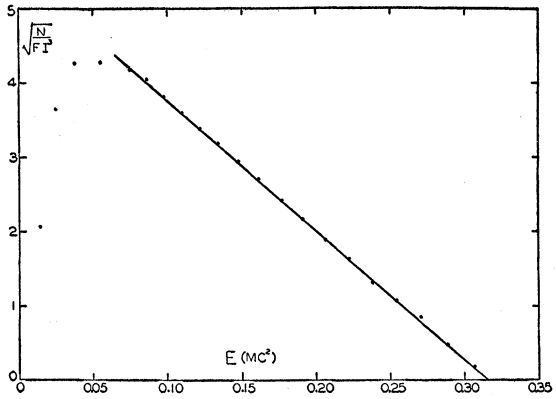

FIG. 7. The Fermi plot for the beta-spectrum of Nb^{95}.

The method used by Hill and Mihelich was to identify the photographic lines of conversion electrons taken with a spectrograph using secondary electrostatic focusing. The ratio of Au^{199} to Au^{198} was estimated to be 1:95 by comparing the intensities of the 0.157-Mev and 0.411-Mev gamma-rays; it was reduced to 0.6:100 later[29] by drastically changing the percentage of 0.157-Mev gamma-rays per beta-particle emission of Au^{199}. Here the same problem was studied by measuring simultaneously the total beta-disintegrations of both Au^{199} and Au^{198}. The result differs considerably from that of Hill and Mihelich.

Figure 9 shows the beta-spectrum of Au^{198}. The sample was obtained from Oak Ridge, where it had been irradiated for seven days with an average flux of approximately 6.5×10^{11} cm^{-2} sec^{-1}; the measurement was performed about three days after the irradiation. The spectrographic analysis of the particular piece of gold from which the sample was taken shows the presence of a small amount of Ag and very small amount of Ca, Cu, and Mg,[30] none of which should interfere with the present study.

The Fermi plot for the spectrum is shown in Fig. 10 and indicates clearly the superposition of two components. The end point of the high energy component is 0.97±0.01 Mev, while that of the low energy component is estimated from the Fermi plot as about 0.4 Mev. Besides the well-known 0.411-Mev gamma-ray, two

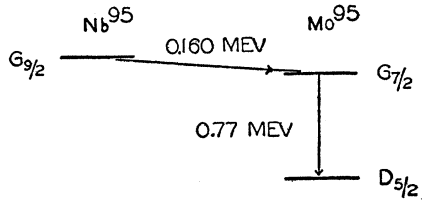

FIG. 8. The decay scheme of Nb^{95} and the suggested level values.

[29] R. D. Hill, Phys. Rev. **79**, 413 (1950).
[30] The author is indebted to Mr. J. A. Cox, Superintendent of the Radioisotope Control Department, Oak Ridge National Laboratory, for supplying this information.

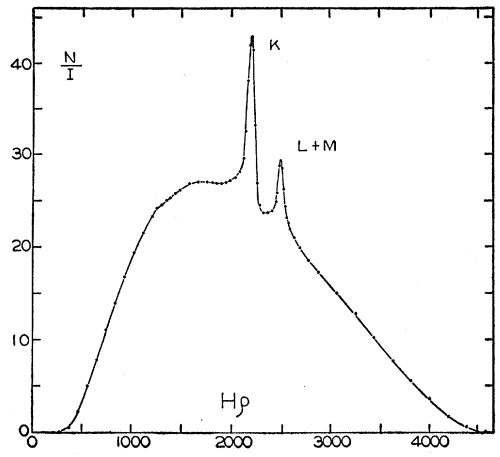

FIG. 9. The beta-spectrum of Au^{198}. Sample was obtained from Oak Ridge National Laboratory.

gamma-rays of energy 0.159 and 0.207 Mev, respectively, were also found. None of the impurities leads to the observed activities. Chemical purification was performed[31] and the activities were found to be unaffected. Therefore the activities must be due to some isotope of gold.

Figure 11 shows the beta-spectra of Au^{198} irradiated in the heavy water pile of Argonne Laboratory. In these measurements some older and therefore weaker sources were used. Hence the spectrometer was operated with lower resolution but higher transmission. Furthermore, an end-window mica counter of window thickness 1 mg/cm^2 was used instead of a Nylon window counter, because the supporting grid of the Nylon window counter cuts down the transmission by a factor of about one-half. Curve 1 in Fig. 11 is the spectrum of a sample irradiated for 45 hours in the pile. The measurement was done about one week after irradiation. Curve 2 corresponds to the measurement on the same sample 34 days after irradiation, with the high energy part normalized to coincide with that of the first measurement. It can be seen that the two activities have different half-lives. Assuming that the beta-spectrum of Au^{198} is simple as indicated by the dotted curve (No. 3) in the figure, the half-life of the low energy component can be estimated by comparing curves 1 and 2. It is found to be 3.2 days, which differs by about 4 percent from the accepted value of the half-life of Au^{199}, 3.3 days.

The end-point energy of the low energy component can be estimated from the Fermi plot which is shown in Fig. 12. It is 0.35±0.04 Mev. This agrees with the values for Au^{199} of 0.32 Mev by Beach, Peacock, and Wilkinson,[32] and of 0.38 Mev by Mandeville, Scherb, and Keighton.[33]

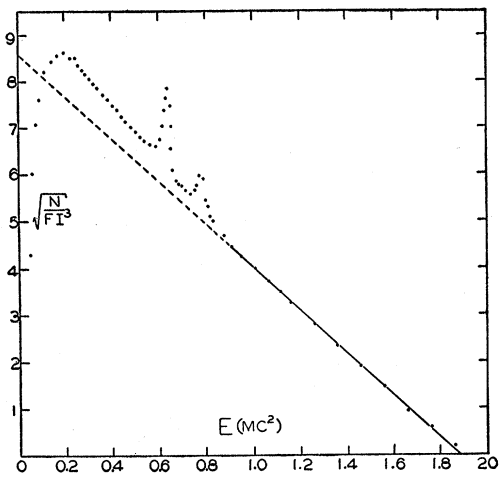

FIG. 10. The Fermi plot for the beta-spectrum of Au^{198} shown in Fig. 9.

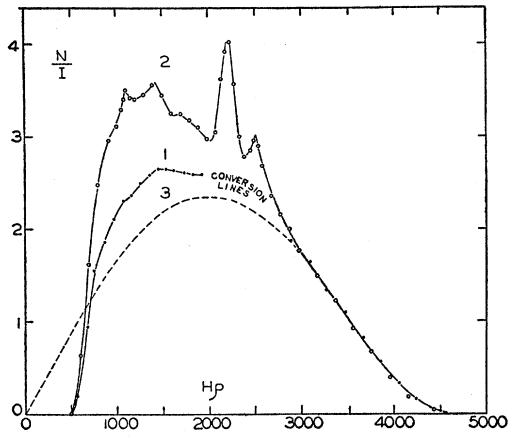

FIG. 11. The beta-spectra of Au^{198}, sample was irradiated in the heavy water pile of Argonne National Laboratory. Curves 1 and 2 are the spectra measured 7 and 34 days after irradiation, respectively. Curve 3 is the spectrum corresponding to a straight Fermi plot.

The internal conversion peaks in curve 2 are readily identified with the energy spectrum of Au^{199} found by Beach *et al.*[32] with a thinner counter and a better resolution. The first peak can be identified with the K conversion line of the 0.157-Mev gamma-ray. The second peak is the L line corresponding to the 0.157-Mev gamma-ray, the K line of the 0.207-Mev gamma-ray, and the K line of the 0.230 gamma-ray, which are not resolved. The third peak is the L line of the 0.207-Mev gamma-ray. All these findings show that the extra activity is due to Au^{199}. The neutron absorption cross section of Au^{198} can be estimated from the comparison of the beta-activities of Au^{198} and Au^{199} and the neutron flux.

After making the correction for window absorption, the total amount of Au^{199} compared to Au^{198} under the curve in Fig. 9 was about 16 percent, while that under

[31] R. S. Krishnan, Proc. Cambridge Phil. Soc. **37**, 186 (1941). Thanks are due to Mr. J. B. Niday and Mrs. A. Tompkins for their help in performing the chemical process.

[32] Beach, Peacock, and Wilkinson, Phys. Rev. **76**, 1624 (1949).

[33] Mandeville, Scherb, and Keighton, Phys. Rev. **74**, 601 (1948).

curve 1 in Fig. 11 was about 11 percent and that under curve 2 was about 30 percent. The first value gives a neutron absorption cross section of Au^{198} of $\approx 8.8\times10^5$ barns, and the last two give an average result of 9.1×10^5 barns if a neutron flux is 10^{12} cm^{-2} sec^{-1} is assumed. This is much higher than the value of Hill and Mihelich, i.e., 1.6×10^4 barns. This large discrepancy might partly be due to the uncertainty in the pile neutron flux on which the calculations are based.

The K internal conversion coefficient for 0.411-Mev gamma-ray is 3.0 percent. The L and M conversion coefficients are 1.3 percent. All these values agree exactly with the results of Siegbahn and Hedgran.[22] The radiation is classified as electric quadrupole radiation. This classification is consistent with the lifetime of the isomeric state (10^{-11} sec).[34]

The multipole order of the 0.411-Mev gamma-ray, together with the fact that Hg^{198} has zero spin and presumably even parity, enable one to assign 2+ to the excited state of Hg^{198}. The Fermi plot of Au^{198} is

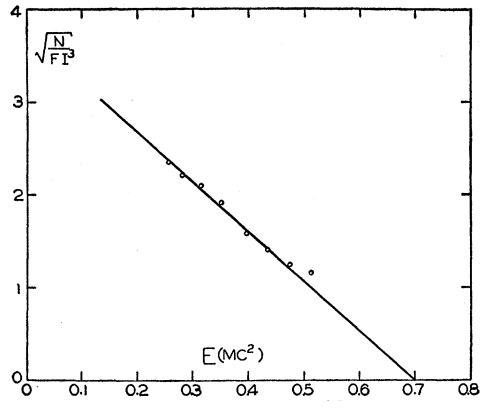

FIG. 12. The Fermi plot for the beta-spectrum of Au^{199}.

straight, which could mean that this is an allowed spectrum or a first-forbidden spectrum ($\Delta J=0$ or 1, yes) with allowed shape which happens more often for high Z isotopes. Indeed, the condition that the correction factor for forbiddenness reduces to a constant is given by Konopinski[5] as

$$Z \gg 1.6W_0^{1.5},$$

where W_0 is the maximum energy (including rest mass) in unit of mc^2 of the beta-particles. For Au^{198}, $1.6W_0^{1.5} \approx 8$, and this condition is thus fulfilled. Therefore the ground state of Au^{198} could be 1±, 2±, or 3±. The ft value ($\log ft=7.3$) tends to indicate a first-forbidden spectrum and hence a negative parity of the ground state.

The shell structure model favors a 2− state[21] for the ground state of Au^{198}. Adopting this assignment, one obtains the tentative level scheme shown in Fig. 13.

[34] R. L. Graham and R. E. Bell, Phys. Rev. **84**, 380 (1951).

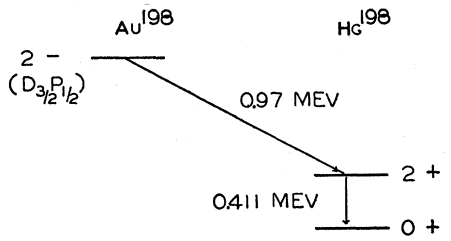

FIG. 13. The decay scheme of Au^{198} and the suggested level values.

4. Hf^{181}

The decay of Hf^{181} and the internal conversion coefficients have been investigated by Chu and Wiedenbeck[35] and lately by Hedgran and Thulin.[36] The decay scheme proposed by Chu and Wiedenbeck is shown in Fig. 14. It fits with the delayed nature of the gamma-rays[37] following the beta-decay and the cascade nature of the 0.130-Mev and the 0.471-Mev gamma-rays.[38,39]

Later, Jensen found another gamma-ray of energy 0.087 Mev.[40] This gamma-ray was identified by Cork *et al.*[41] as due to the activity of Hf^{175}. The total amount of Hf^{175} relative to the activity of Hf^{181} one month after four weeks of irradiation was determined by Hedgran and Thulin[36] as 2.1 percent. However, Cork *et al.*[41] found one other gamma-ray of energy 0.601 Mev by using a photographic registration. This was also confirmed by Burson *et al.*[42] They assigned the gamma-ray to the direct transition from the highest excited state of Ta^{181} to the ground state. Since this gamma-ray has not been reported by Chu and Wiedenbeck and Hedgran and Thulin, it is presumably of very low intensity.[43] The

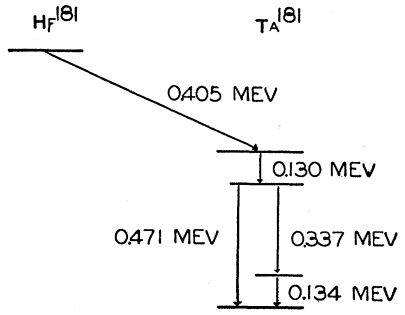

FIG. 14. The proposed decay scheme of Hf^{181} of Chu and Wiedenbeck.

[35] K. Y. Chu and M. L. Wiedenbeck, Phys. Rev. **75**, 226 (1949).
[36] A. Hedgran and S Thulin, Phys. Rev. **81**, 1072 (1951).
[37] S. De Benedetti and F. K. McGowan, Phys. Rev. **75**, 728 (1948).
[38] Mandeville, Scherb, and Keighton, Phys. Rev. **75**, 221 (1949).
[39] Bunyan, Lundby, and Walker, Proc. Phys. Soc. (London) **A62**, 253 (1949).
[40] E. N. Jensen, Phys. Rev. **76**, 958 (1949).
[41] Cork, Stoddard, Rutledge, Branyan, and Le Blanc, Phys. Rev. **78**, 299 (1951).
[42] Burson, Blair, Keller, and Wexler, Phys. Rev. **83**, 62 (1951).
[43] The branching ratio is less than 2 percent according to Mr. S. B. Burson's private communication.

TABLE V. Number of conversion electrons for Hf^{181}.

	Chu and Wiedenbeck	Hedgran and Thulin
$N_{K_1}+N_{K_2}$	0.78	0.32
$N_{K_1}:N_{K_2}$	2.5:1	2:1
$N_{L_1}+N_{L_2}+N_{M_1}+N_{M_2}$	0.514	0.36
N_{K_3}	0.019	
N_{L_3}	0.0053	
N_{K_4}	0.021	
N_{L_4}	0.0071	

proposed decay scheme seems to be well established, except that there are some small differences in the energies of gamma-rays as measured by different authors. But the problem of the internal conversion coefficients is still an open one as is shown in Table V, where N_{Ki} $(i=1, 2, 3, 4)$ is the number of K conversion electrons of the ith gamma-ray emitted per second per unit beta-disintegration, and N_{Li} and N_{Mi} denote the number of L and M conversion electrons, respectively. The gamma-rays are numbered according to increasing energies. The purpose of the present measurement, which was done before the publication of Hedgran and Thulin's paper, was to measure the conversion coefficients of the gamma-rays to compare with Chu and Wiedenbeck's results. It was found that the results were in agreement with Hedgran and Thulin's and, in spite of the complications of the problem and the uncertainties of the measurements, could be taken in conjunction with the determinations of the branching ratio, the ratio of $N_{K1}:N_{K2}$ and the ratio of $N_{K2}:N_{L2}$ of the latter, to give a complete comparison with the theoretical predictions of the shell structure model and Goldhaber and Sunyar's empirical classification of nuclear isomers.[18]

The beta-spectrum of the present measurement is shown in Fig. 15, where the dotted curve corresponds to a straight Fermi plot. The sample was in the form of Hf_2O_3 powder irradiated in Oak Ridge pile for four weeks. The source was prepared by depositing very fine Hf_2O_3 powder of 2 microcurie strength on a conducting Nylon foil and then covering the source with a piece of zapon foil of thickness ≈ 5 $\mu g/cm^2$ to hold the

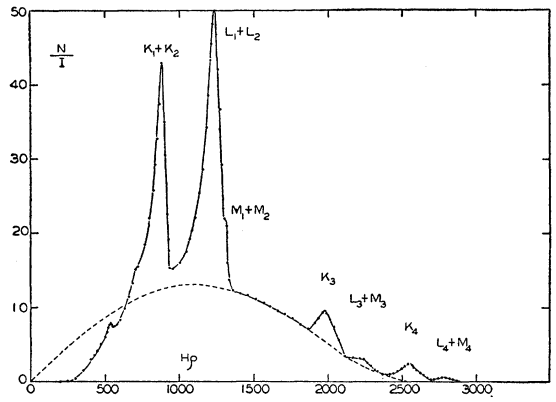

FIG. 15. The beta-spectrum of Hf^{181}.

powder in place. The size of the source was ≈ 0.5 cm in diameter and the thickness was ≈ 0.3 mg/cm^2. The energies of the gamma-rays of Hf^{181} and their internal conversion coefficients are listed in Table VI, where $N_{K1}:N_{K2}\approx 2:1$, $N_{K2}:N_{L2}\approx 8$, the branching ratio for the 4th gamma-ray ≈ 87 percent, and the activity of Hf^{175} to that of $Hf^{181}\approx 2.1$ percent just after four week irradiation, $\alpha_K\approx 40$ percent, and $\alpha_K/\alpha_L\approx 4.5$ for the 0.342-Mev gamma-ray of Hf^{175} [36,44] were used for the calculation. The errors of the internal conversion coefficients were calculated from the experimental errors of α_K', etc., which are about 10 percent. The errors of the internal conversion coefficients for the third gamma-ray could not be given because of the uncertainty of Wilkinson and Hicks' measurements.

The theoretical K-shell internal conversion coefficients for different gamma-rays for various multipole radiations from the table of Rose *et al.*[1] and the values of α_K/α_L from Goldhaber and Sunyar's empirical curves[18] are listed in Table VII.

In connection with the comparison of the experimental results with the theoretical values two remarks must be

TABLE VI. γ-ray energies and internal conversion coefficients for Hf^{181}.

Energy of gamma-rays	Internal conversion coefficients	
0.130 Mev	K_1	0.34±0.06
	L_1+M_1	0.56±0.10
0.134	K_2	$3.1^{+2.2}_{-1.1}$
	L_2+M_2	$0.38^{+0.28}_{-0.16}$
0.340	K_3	0.13 (very poor estimation)
	L_3+M_3	0.02 (very poor estimation)
0.474	K_4	0.024±0.003
	L_4+M_4	0.006±0.0006

made: (1) The conversion peaks of the first two gamma-rays occur in the low energy range; considerable uncertainties could thus be involved in the corrections for the counter window absorption. (2) The theoretical values of the K-shell internal conversion coefficients for the first two gamma-rays were obtained by means of extrapolation of the curves of Rose *et al.*, and errors could thus be introduced. However, all those uncertainties do no completely obscure the nature of the gamma-rays. In fact, one can readily assign the fourth gamma-ray as an electric quadrupole radiation, both the second and the third gamma-ray as magnetic dipole radiations, and the first gamma-ray as most likely an electric quadrupole radiation. The classification of the first gamma-ray is consistent with the lifetime of the isomeric transition (22 μsec).

The Fermi plot of the beta spectrum is shown in Fig. 16. It is straight with an end-point energy of 0.408±0.008 Mev. This straight Fermi plot could be an indication of either an allowed spectrum or a first-forbidden spectrum with a constant correction factor

[44] G. Wilkinson and H. G. Hicks, Phys. Rev. **75**, 696 (1949).

for the forbiddenness as in the case of Au^{198}. However, a first-forbidden spectrum is compatible with the nature of the first gamma-ray from the following theoretical consideration.

The spin of Hf^{177} and Hf^{179} are both 1/2. Shell structure model predicts the value $p_{1/2}$ as the state of Hf^{177}, Hf^{179}, and for Hf^{181}.[21] Because of the beta-spectrum is of an allowed shape, the highest excited is likely to be of state $3/2\pm$ or $1/2\pm$. The spin of Ta^{181} is 7/2. Shell structure model favors a $g_{7/2}$ for the ground state.[21] The multipole orders of the second, third, and fourth gamma-ray indicate the first excited state and the second excited state as 5/2+ and 3/2+, respectively. Therefore, the nature of the first gamma-ray requires 3/2+ or 1/2+ as the highest excited state. This fits with a first-forbidden spectrum and high ft value ($\log ft=7.2$).

This case is quite interesting purely from the point of view of the shell structure model, because the states of Ta^{181} seem to cover all the term values in the sixth shell, ($g_{7/2}$, $d_{5/2}$, $d_{3/2}$, and $s_{1/2}$) except the last one ($h_{11/2}$). This is different from the scheme of Goldhaber and Sunyar.[18] They assigned the first excited state as 7/2+ and the first, third, and the fourth gamma-ray all

TABLE VII. K-shell internal conversion coefficients and K–L ratios for the γ-rays from Hf^{181}.

Energy of gamma-rays in Mev	K-shell internal conversion coefficients				α_K/α_L			
	E–2^1	E–2^2	E–2^3	M–2^1	E–2^1	E–2^2	E–2^3	M–2^1
0.130, 0.134	0.15	0.52	1.5	1.9		0.5	0.35	7.6
0.340	0.014	0.041	0.11	0.14		2.7	1.7	7.9
0.474	0.0066	0.0185	0.0475	0.060		4.3	3.0	8.0

as electric quadrupole radiations and did not mention the nature of the second one.

The author wishes to express his sincere appreciation to Professor H. L. Anderson for suggesting this problem and for his kind guidance during its progress. Thanks are also due to Mr. S. D. Bloom, Mr. S. A. Moszkowski, and Mr. D. Kahn for their friendly help.

APPENDIX

In most of the beta-ray spectrometer measurements, a single conversion line can be represented by an isosceles triangle having a width at half-maximum proportional to the momentum of the conversion electrons and a height for a source of unit strength (one particle per second) equal to the transmission of the machine t. Let p' be the momentum of the conversion electrons and w the resolution; the width of the triangle at half-maximum will be wp' if the magnetic field of the instrument is linearly proportional to the current through the coil and there is no distortion of any kind.

The shape of a single conversion line of momentum p', denoted by $K(p, p')$, is essentially the probability that an electron of momentum p' from the source appears at the slit of the detector of a beta-ray spectrometer when

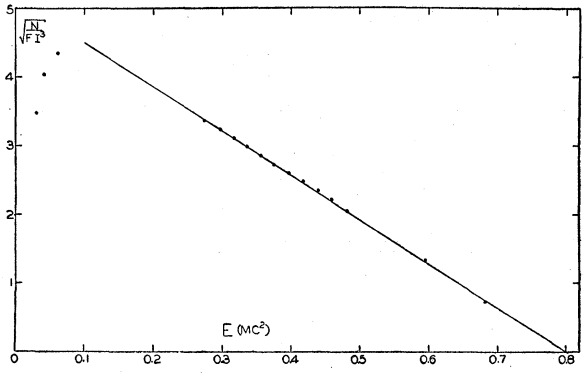

FIG. 16. The Fermi plot for the beta-spectrum of Hf^{181}.

the current through the coil is I, appropriate to the magnetic rigidity p. If $n(p')dp'$ is the number of beta-particles of momenta between p' and $p'+dp'$ emitted per second and $N_c(p)$ is the counting rate of the detector when the coil current is I, then

$$N_c(p)=\int_0^\infty K(p, p')dp'n(p').$$

In the case of $K(p, p')$ being an isosceles triangle,

$$N_c=\int_a^b tn(p')dp'\frac{wp'-(p-p')}{wp'}+\int_b^c tn(p')dp'\frac{wp'-(p'-p)}{wp'}, \quad \text{(i)}$$

where $a=p/(1+w)$, $b=p$, and $c=p/(1-w)$.

Equation (1) can be easily transformed into a differential equation. After terms containing the third or higher power of w being neglected, it reads

$$(1/wt)d^2N_c/dp^2+2dn/dp+pd^2n/dp^2, \quad \text{(ii)}$$

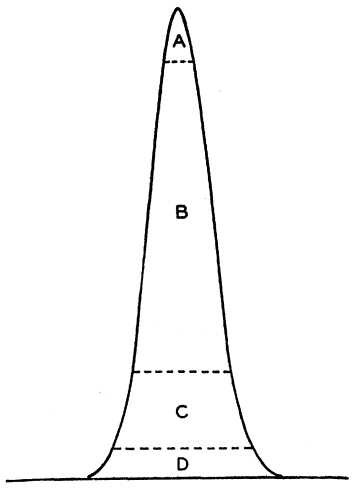

FIG. 17. A Gaussian curve, considered as a sum of an isosceles triangle and three isosceles trapezoids.

with the following solution

$$n=(1/wt)(N_c/p). \quad \text{(iii)}$$

This is Eq. (5). The correction term to N_c/p is of order w^2, which is small for $w=2$ percent.

DISCUSSION

Equation (3) can be generalized to the case that the shape of a single conversion line is symmetrical with respect to p' but other than an isosceles triangle, because it is always possible to divide a symmetrical shape into isosceles triangles so that they can be treated separately as the special case given above. For instance, a Gaussian curve can be divided into an isosceles triangle A and three isosceles trapezoids B, C, and D are illustrated in Fig. 17. An isosceles trapezoid may be regarded as the difference of two isosceles triangles whose apexes are at the position p'. Equation (3) is thus applicable to the general case, except that the width w will be an average of widths of different triangles with proper weights attached.

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Energy Levels in C^{12}, Al^{27}, and Na^{23} Using a Scintillation Spectrometer for Heavy Particles*

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(Received March 24, 1952)

Energy levels in carbon, aluminum, and sodium have been measured using a "proportional" sodium iodide scintillation counter to determine energies of outgoing protons from inelastic proton scattering at 7.26 Mev with the following results: C^{12}: 4.45 Mev; Al^{27}: 0.84, 1.01, 2.23, 2.77, 3.03, 3.71, 4.00, 4.47, 4.60, 4.87, 5.43 Mev; Na^{23}: 2.10, 2.37, 2.69, 3.01, 3.70, 3.92, 4.45 Mev.

Factors limiting the resolution are presented, and it is found that the standard deviations of the distributions are consistent with 2.4 photoelectrons produced at the photocathode of the RCA 5819 per kev of particle energy lost.

1. INTRODUCTION

CONTINUED interest in the level structure of the nuclides has led to many recent experimental determinations of excited states. Usually the energy assignments are based on range measurements of the outgoing particles in nuclear scattering experiments or by deflection of these particles in known magnetic fields. This paper reports energies for excited states of several nuclides found by a different method which takes advantage of the "proportional" characteristics of the scintillations produced in a NaI(Tl) crystal when it is excited by protons.[1] By using a crystal of sufficient thickness all of the particle range is absorbed and, in principle, all groups can be examined simultaneously.

2. EXPERIMENTAL

The object in designing such a counter is to get as much of the light as possible from the crystal into the phototube since this is one of the factors which determines the resolution. For this purpose a simple aluminum reflector was designed as shown in Fig. 1. Because of total reflection at the crystal surface, a large part of the light comes out of the edge of the crystal, and the aluminum reflector is designed to utilize as much of this as possible. The volume of the reflector is filled with Dow-Corning DC-703 silicone oil to provide optical matching between the glass crystal holder and the glass tube face. Mineral oil, which is usually used in this application, is unsatisfactory in vacuum systems because of its high vapor pressure. The transmission of both silicone oil and mineral oil was checked and in the region of interest the silicone oil was actually found to be better.

The sodium iodide crystal was cleaved on all six sides from a larger single crystal obtained from Harshaw Chemical Company. Cleaving gives a thin flat plate with the smoothest, most uniform surface possible. The cleaving operations were carried out in a dry box. Difficulty with gamma-ray background is minimized by making the crystal thin and hence increasing the ratio of efficiency for detecting charged particles to efficiency for detecting gamma-rays.

The counter is mounted on one of the movable arms in the MIT scattering chamber.[2] No window is used between the crystal and the target, and this necessitates filling the whole chamber volume with dry nitrogen

* This work has been supported in part by the joint program of the ONR and the AEC.

[1] Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).

[2] Boyer, Gove, Harvey, Deutsch, and Livingston, Rev. Sci. Instr. **22**, 310 (1951).