

DISINTEGRATION OF CERIUM OF MASS 144

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by

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SUMMARY

A study of the beta decay of 275 day Ce^{144} and its daughter 17.5 min. Pr^{144} , has been made using a thin lens magnetic spectrometer.

Analysis of the beta-spectrum indicates the presence of five spectra with end-points at 3.06 ± 0.09 , 1.05 ± 0.05 , 0.517 ± 0.036 , 0.303 ± 0.020 , and 0.179 ± 0.015 Mev. The first three have been assigned to Pr^{144} , the remaining two to Ce^{144} .

Conversion lines at 90.1 ± 0.9 and 125 ± 1.3 Kev. are attributed to internal conversion of a 132 Kev. gamma-ray following the 0.179 Mev. beta-disintegration of Ce^{144} . Measurement of gamma-ray energies by spectrometric methods proved impractical with the activity available, but absorption methods reveal three other gamma-rays of energies about 2, 0.5 and 0.22 Mev.

The results of coincidence experiments are, on the whole, inconclusive, but indicate in general the complexity of the spectra.

Tentative disintegration schemes are presented.

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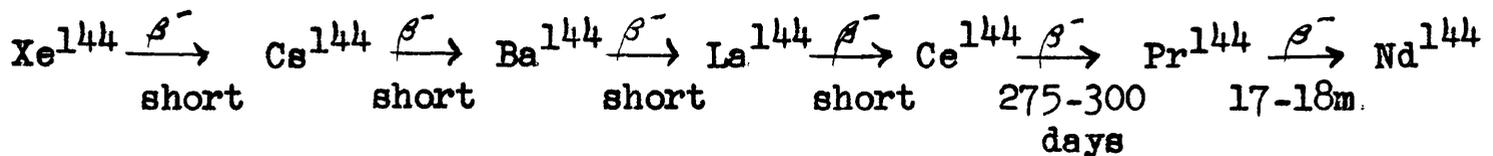
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I. INTRODUCTION

(a) Previous Studies.

The most complete listing of the production and characteristics of Ce^{144} and Pr^{144} is contained in a recent publication^(A1) based upon information issued by the Plutonium Project (U.S.A.). This report lists Ce^{144} as a fission product descended from Xe^{144} through a long chain of short lived beta-disintegrations. Pr^{144} is descended from Ce^{144} by beta-decay, and in turn decays by beta-emission to Nd^{144} which is stable. The entire process may be represented as follows:



The half lives of the respective isotopes are indicated under the arrows.

Most of the information regarding the characteristics of Ce^{144} and Pr^{144} which is listed in the paper referred to is unfortunately not available in published form, derived as it is, from secret reports which to the author's knowledge have not as yet been declassified. Non-secret publications are confined to three papers^(A2-A4) which reveal that Ce^{144} has a half-life of 300 days, decaying to Pr^{144} the half life of which is given as 17 minutes. The end point of the beta-spectrum of Pr^{144} is given as 3.1 Mev, as determined by absorption in aluminum.

The data collected by the Plutonium Project is a more copious source of information. Ce^{144} is listed as having a 275 day half-life. The end point of its beta-ray spectrum is found to be 0.348 Mev. (spectrometer determination) or ~ 0.3 Mev. (absorption in aluminum) and it is reported as emitting no gamma radiation. Pr^{144} is reported as having a half life 17.5 minutes or 18 minutes and yielding beta-rays of maximum energy 3.07 Mev by spectrometric determination, or 3.0 Mev, 3.1 Mev, 3.2 Mev, or 2.8 Mev, all measured by absorption in aluminum. Gamma radiation of

energies 0.135 Mev, 0.22 Mev, and 1.25 Mev (low intensity) are attributed to it, the first determined by spectrometric study of conversion electrons, the other two by absorption in lead. A doubtful gamma-ray of 0.145 Mev and conversion electrons of energies 0.091 and 0.128 Mev with a doubtful group at 0.103 Mev are also reported.

The Segre chart (Sept. 1946)^(A-5) yields essentially the same information with the exception that the 1.25 Mev. gamma ray is attributed to Ce^{144} .

The present research was undertaken with the aim of securing further information regarding the disintegration of Ce^{144} and Pr^{144} leading if possible to the disintegration schemes of both radioactive isotopes. The results confirm those of the Plutonium Project only in part. It is found that the end-point of the Pr^{144} spectrum is 3.06 Mev, in agreement with that reported by the Plutonium Project, but the end point of the Ce^{144} spectrum is found to be 0.303 Mev as compared with the value 0.348 Mev given by the Plutonium Project. Evidence is presented to show that the conversion lines at 0.091 Mev and 0.128 Mev are probably due to Ce^{144} and not Pr^{144} . In addition it is demonstrated that the Pr^{144} beta spectrum is complex, with the high energy spectrum (end point 3.06 Mev) leading to the ground state of Nd^{144} . The Pr^{144} spectrum appears to consist of three components, the Ce^{144} spectrum of two. Tentative decay schemes are presented.

A beta-ray spectrometer of the thin lens type has been used in the investigation. The remainder of this section is given over to a brief history of the development of beta-ray spectrometers and spectrographs, and to an outline of the theory of beta-decay.

(b) Historical

Early work in beta-ray spectroscopy commenced with the researches of Baeyer and Hahn⁽¹⁾ in 1910 who investigated the velocity of individual beta-rays using the small deviations produced by a magnetic field. The first use of the magnetic spectrograph, in which beams of particles were actually focussed, occurs in the work of Danysz⁽²⁾ in 1911, who studied the velocity distributions of beta-rays with a spectrograph of the semi-circular or 180° focussing type. Rutherford and Robinson⁽³⁾ also employed this type of spectrograph to investigate velocities of beta-rays, and for the next decade, all work in beta-ray spectroscopy was carried out with instruments employing the same principle. Electrons leaving a point or line source traverse circular paths owing to the action of a uniform magnetic field. Electrons of equal velocity travelling perpendicular to the direction of the field will describe circular paths of equal radii, and to a first order approximation will meet at a common point or line after having traversed a semi-circle. A photographic plate placed at this point acts as a recording instrument.

The difficulties met with in reproducing accurately the magnetic fields of the required area and intensity, and the problem of securing sufficient intensity for sources of low activity led in 1923 to a suggestion by Kapitza to employ a helical method of focussing, a suggestion carried out by Tricker⁽⁴⁾. A uniform field is generated by means of a solenoid, the source being placed on the axis at one end, and a photographic plate at the other with its long axis coinciding with the axis of the solenoid. Helical paths are traversed and electrons come to a focus at some point along the long axis of the plate. A consideration of the distances along the plate at which various groups of electrons are focussed leads to a

determination of the velocities of these groups.

The focussing action of a "thin" magnetic lens, as distinct from that of a solenoid, was first employed by Klemperer⁽⁵⁾ in 1935. In a spectrometer of this type, the lens can be placed anywhere between the source and the recording instrument, and as the laws of geometrical optics apply, magnification or reduction in size of the image as compared with the source is possible. Davis and O'Ceallaigh⁽⁶⁾ in 1937 applied such a lens to cloud chamber photography and Cosslett⁽⁷⁾ in 1940 utilized the action of a short magnetic lens in showing that beta-rays were not emitted in the proton bombardment of lithium.

In 1941 a very flexible instrument utilizing a magnetic lens was built by Deutsch, Elliott and Evans who have given a detailed account of its theory, design, and applications⁽⁸⁾. The spectrometer has proven a very popular tool since it is so well suited for use in coincidence studies, and possesses in addition such advantages as the use of circular sources and easy accessibility of both counter and source. In general, relative to the semi-circular focussing instruments, the magnetic lens spectrometer is characterized by a higher transmission but by a somewhat poorer resolution. Although the 180° focussing spectrometer can be built to give a high transmission by using properly designed non-uniform fields⁽⁹⁾, two cannot be employed simultaneously working from the same source because of the mutual interactions of their fields. For a single accurate spectrogram, however, there seems no doubt that the semi-circular type spectrometer is preferable.

(c) Theoretical

The salient feature of beta-decay is the emission, not of mono-kinetic electrons, but of electrons the energy distribution of which is a continuum reaching from zero or near-zero to an upper limit. This has long constituted a major problem, since apparently it constitutes a

violation of the law of conservation of energy. All nuclei of the beta-emitter must be assumed to possess the same initial energy, and in some cases the emission of the electron results in the nucleus being left in its ground state. In others, beta-emission is followed by the radiation of monochromatic gamma quanta prior to arrival at the ground state. Since the final states of the nuclei are identical, it must be assumed either that all the electrons possess initially the same energy, or that another process occurs at the time of beta-emission, the effect of which in some manner accounts for the discrepancy.

The possibility that all the electrons emitted are initially monoenergetic has been investigated by Ellis and Wooster⁽¹⁰⁾ and by Meitner and Orthmann⁽¹¹⁾ with negative results. By means of a calorimeter the energy of the electrons emitted by a known number of atoms of RaE was measured. The energy per electron found in this manner agreed very well with the average energy as deduced from the spectrum of RaE and not at all with the upper energy limit of the spectrum.

The question as to whether beta-rays are identical with ordinary electrons has also been raised, but this possibility has, in the main, *not* been ~~discarded~~^{seriously considered}⁽¹²⁾. The ratio e/m for beta-rays agrees very well with that for ordinary electrons⁽¹³⁾ but the proof that both beta-rays and electrons possess identical spins has not as yet been demonstrated⁽¹²⁾. In addition to the problem of conservation of energy (and momentum) during beta-decay it should also be mentioned that difficulties exist with regard to conservation of spin and statistics. There are very good reasons for believing that electrons do not exist as such in the nucleus prior to disintegration⁽¹⁴⁾ so that negative beta-decay consists in the conversion of a neutron into a proton and negatron⁽¹⁵⁾. Since all three have spin $1/2$ and obey Fermi statistics, the emission of a negatron alone will not conserve

spin and statistics. If the beta-ray is not identical with an ordinary electron it might conceivably possess a spin such that the process of beta-decay as outlined above does not result in non-conservation of spin. The possibility that such a beta-particle could possess sufficient mass to account for the discrepancy in energy is however untenable⁽¹²⁾.

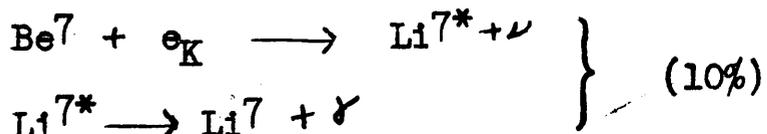
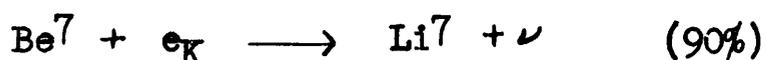
A suggestion by Pauli in 1933⁽¹²⁾ that the process of beta-decay results in the creation of an undetected particle in addition to the electron led to a theory in which the laws of conservation of energy, momentum, spin, statistics, and charge could all be retained. The particle has been given the name neutrino for positron decay and anti-neutrino for negatron decay, although they are otherwise indistinguishable⁽¹⁴⁾. The neutrino in this hypothesis possesses zero charge, spin $1/2$, very small mass, and obeys Fermi statistics. Its function is to share energy with the emitted electron, such that their sum corresponds to the upper limit of the beta-spectrum, since very strong evidence is at hand to support the belief that the energy lost by the nucleus does in fact correspond to the upper limit of the beta-spectrum⁽¹⁶⁾,⁽¹⁷⁾. The accuracy of the correspondence in these experiments sets an upper limit to the mass of the neutrino of about $1/7$ the electron rest mass. Theoretical estimates assign a mass of $1/30 - 1/45$ of the electron mass to the neutrino⁽¹⁸⁾.

The properties attributed to the neutrino make its observation very difficult. Since it undoubtedly possesses no charge - due to the fact that charge is conserved by the other particles - there is no possibility of strong ionization by neutrinos. Ionization due to a magnetic moment associated with the spin of the neutrino is conceivable, but an experiment⁽¹⁹⁾ designed to detect such ionization has yielded negative results. In this experiment strong radioactive sources were shielded with 1 metre of lead and

a search made for ionization produced by neutrinos. An evaluation of the results⁽¹⁴⁾ shows that neutrinos cannot form more than one ion per 500,000 Km. path in air. Their magnetic moment on this basis is estimated at less than 1/7000 Bohr magnetrons and probably zero.

The most successful experiment supporting the neutrino hypothesis has been carried out by Allen⁽²⁰⁾, the recoil of a nucleus undergoing K capture being utilized. K-capture is an extension of positron emission, the nucleon however absorbing not one of the infinite number of electrons in negative energy states, but rather one of the bound extra-nuclear electrons from the K-shell. As for positron emission, a neutrino is created in the process. Since the momentum of the K-electron is negligible, a measurement of the momentum of the recoiling nucleus will also be a measure of the momentum of the neutrino. It is, of course, assumed that momentum is conserved, and experiments of this type have been criticized on such grounds⁽¹²⁾.

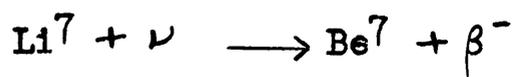
The reaction chosen employed Be^7 which decays in two ways:



in which e_K and ν represent the K-electron and the neutrino respectively. The mass difference between Be^7 and Li^7 is known to be 1 Mev and utilizing this fact, 90% of the recoils should have 58 ev. energy, and the other 10% energies less than this maximum. The recoil energies were very conveniently measured by means of a retarding electric field, a method applicable since the process of K-capture leaves the atom in an ionized state. The results indicated a maximum recoil energy about 10 to 15 volts lower than the neutrino hypothesis would require, but in view of experimental errors, the value is quite

good. The experiment does not answer the question as to whether the decay process involves the generation of single or multiple neutrinos.

Other recoil experiments have been performed^{(21), (22), (23), (24)} the conclusions resulting from such experiments showing, in general, that momentum is not conserved in a system consisting of the electron and nucleus alone. The critical experiment, yet to be performed, will probably utilize the large neutrino flux from a chain reacting pile in an endeavour to observe the inverse beta process, e.g.



The cross-section for such a reaction as estimated by Bethe⁽¹⁵⁾ is of the order of 10^{-44} cm², hence the necessity for a large neutrino flux. Successful completion of such an experiment would represent positive proof of the existence of the neutrino.

Despite the lack of experimental proof of the existence of the neutrino, the success attending the theory of beta-decay, as formulated by Fermi⁽²⁵⁾, has thus far justified the neutrino hypothesis. The Fermi theory regards the process of beta-decay as the transformation of a neutron into a proton, a negatron and a neutrino, the available energy being shared by the negatron and neutrino. In order to calculate the probability of these processes, Fermi introduced a new force or interaction between the nucleon, the electron, and the neutrino in analogy with the interaction between charges and the electromagnetic field in quantum electrodynamics.

Following Konopinski⁽²⁶⁾ the theory is developed as follows:

If the Hamiltonian of the interaction between the nucleon and electron-neutrino field is H , then the probability per unit time PdW of the emission of a beta-particle with energy between W and $W + dW$ is given in rationalized relativistic units⁽²⁷⁾ by

$$PdW = (G^2/2 \pi^3) \langle |H|^2 \rangle_{av} pW(W_0 - W)^2 dW \dots\dots\dots(1)$$

G is a constant which measures the strength of the coupling between the

nucleon and the electron-neutrino field and has a value $\sim 10^{-13}(27)$; p represents the electron momentum:

$$\langle |H|^2 \rangle_{av.} = (2p^2)^{-1} \int d\omega \sum_{lmj} \left| \int dv \sum_K V^* E_K U \right|^2 \dots\dots\dots(2)$$

where $G H_K$ is defined as

$$H = G \left\{ (O^L \varphi \psi^*) O^H Q + (O^L \varphi \psi^*)^* O^{H*} Q^* \right\} \dots\dots\dots(3)$$

as applied to the K th nucleon.

In (3) above Q is defined as an operator which, when applied to a wave function describing the initial nucleus, substitutes for it one in which a proton replaces a neutron. Q^* causes a nucleon to make the opposite transition. ψ and φ are wave functions of the electron and neutrino respectively, each being evaluated at the position of the transforming nucleon. O^L and O^H are operators, O^L affecting φ and ψ and O^H the wave function of the nucleon.

The integration $\int d\omega \dots$ of (2) is over the directions of the neutrino momentum, $\sum_{lmj} \dots$ is a summation over the angular momentum quantum numbers of the electron, while $\int dv$ represents an integration over all internal nuclear coordinates including those of the electron and neutrino. U and V are the wave functions of the initial and final nuclei respectively. The operators O^L and O^H were determined by imposing two conditions: the requirement of relativistic invariance and the requirement that the energy H remain a scalar. These requirements do not however determine O^L and O^H uniquely and a further choice must be made. Fermi chose $(O^L \varphi \psi^*)$ to consist of relativistically invariant bilinear combinations of the Dirac components $\psi_1 \dots \psi_4$ with $\varphi_1 \dots \varphi_4$. The ψ and φ components can be arranged into 5 combinations having transformation properties of (a) a scalar (b) a polar vector (c) a tensor (d) an axial vector (e) a pseudo-scalar. The requirement that the energy H remain a scalar results in five possible forms

for (V^*H_KU) in (2) depending on the choice made for the ψ , φ combination given above. Once a choice is made $\langle |H|^2 \rangle_{av.}$ can be calculated.

Fermi chose the polar vector form in analogy with the electromagnetic field. One of the other forms, or a linear combination of two or more of them might be used, the result manifesting itself in a different set of selection rules governing the transitions possible in beta-decay. Yukawa's meson field theory of beta-decay, for instance, requires interaction forms which are linear combinations of (a) and (b), (b) and (c), (c) and (d), or (d) and (e).

The terms:

$$pW(W_0 - W)^2 dW$$

in (1) are a result of the sharing of energy between the electron and the neutrino, and occur regardless of which one of the 5 forms for V^*H_KU is used. W denotes the electron energy including its rest mass and W_0 the energy released by the decaying nucleus.

In order to obtain PdW in explicit form further approximations are made and categories of spectra, viz. allowed, first forbidden, second forbidden, etc., are determined by the degree of approximation required. The first approximation made is to neglect the second terms, if any, in (V^*H_KU) . Justification for this rests upon the belief that nucleons have velocities of the order of $1/10 c$ and as a consequence the second terms, called "velocity terms" are $\sim 1/10$ the first term. The second approximation is the neglect of all terms but the first in the expansion of the light particle wave functions ψ and φ in powers of r the position vector of the transition. With these approximations, taking plane waves for the neutrino and Coulomb field solutions for the electron wave function, the expression secured for $\langle |H|^2 \rangle_{av.}$ in the Fermi formulation (Polar Vector) is

$$\langle |H|^2 \rangle_{av.} = F(ZW) |f|^2 \dots\dots\dots(4)$$

where $|\int 1|^2 \equiv \left| \int dV \sum_K V^* \dots Q_K U \right|^2$

and $F(ZW)$, the Fermi function, is a term indicating the effect of the Coulomb field on the emission of the electrons. Hence

$$PdW = ((G^2) \left| \int \dots \right|^2 / 2\pi^3) \times F(ZW) pW(W_0 - W)^2 dW \dots\dots\dots(5)$$

and the nuclear matrix elements $|\int \dots \dots \dots|$ are expected to be independent of W and Z and of order unity for the allowed spectrum. The expression for $F(ZW)$ as given by Konopinski⁽²⁶⁾ is:

$$F(ZW) = \frac{4(2pR)^{2s-2} e^{\pi \alpha ZW/p} |\Gamma(s + i \alpha ZW/p)|^2}{[\Gamma(2s + 1)]^2} \cdot \frac{(1+s)}{2} \dots(6)$$

R is the nuclear radius i.e. $\frac{1.5 \times 10^{-13} A^{1/3}}{\hbar/mc}$, α the fine structure constant (1/137), Z the nuclear charge (to be taken negative for positron emitters), $s = (1 - \alpha^2 Z^2)^{1/2}$.

It should be mentioned that (6) differs from the expression given by Konopinski and Uhlenbeck in an earlier paper⁽²⁷⁾. In the earlier work the factor $\frac{1+s}{2}$ has been omitted. This term is also absent in Fermi's original work (equation 42, reference 25).

It was noted on developing (5), that two simplifying approximations were made in obtaining an explicit expression for $\langle |H|^2 \rangle_{av}$. These approximations involved the neglect of terms of higher order than the first in \underline{r} , as well as the second terms in $(V^* H_K U)$. If it occurs that the first terms vanish, some selection rule is violated, and the second terms must be considered, leading to the appearance of \underline{r} in the nuclear matrix element. Transitions for which this occurs are known as first forbidden. Second forbidden, third forbidden, etc., transitions require the introduction of \underline{r} into the nuclear matrix element in successively higher powers.

Each of the categories of spectra is characterized by certain selection rules. The two quantum numbers employed in this respect are angular momentum and parity, both of which are conserved in beta-decay. The

angular momentum of nuclei, of nucleons, electrons and neutrinos are all integral or half-integral quantities, in units of \hbar . The concept of parity requires perhaps a word of explanation. If a system be represented by the wave function $\psi(xyz)$, then $|\psi(xyz)|^2$ represents a real quantity and is independent of the coordinate system.. Consider the transformation in which the coordinates become $x'y'z'$ where $x' = -x$, $y' = -y$, $z' = -z$.

$$\text{If } |\psi(xyz)|^2 = |\psi(x'y'z')|^2 = |\psi(-x, -y, -z)|^2$$

then two possibilities exist. Either

$$\psi(xyz) = \psi(-x, -y, -z) \quad \dots(a)$$

$$\text{or } \psi(xyz) = -\psi(-x, -y, -z) \quad \dots(b)$$

If (a) applies, the system is said to have even parity, whereas if (b) applies the system is said to have odd parity.

In order to determine whether or not a transition is allowed, selection rules with respect to angular momentum and parity may be invoked. Such selection rules constitute therefore a means of determining whether the nuclear matrix elements discussed above vanish unless higher terms than the first are taken into account. Conversely, a knowledge of the order of a spectrum can be expected to yield information as to the selection rules obtaining. Each of the five possible forms of $(V^*H_K U)$ will yield different sets of selection rules, although within each set there may be some rules identical with some in other sets. The polar vector forms yields what is known as Fermi selection rules while the tensor form leads to Gamow-Teller or "G-T" rules.

The derivation of the rules can be effected from a knowledge of the wave functions U and V. The initial and final nuclei will each have a specific total angular momentum, J_i and J_f , and either even or odd parity. The eigenfunctions U and V are orthogonal unless $J_i = J_f$ and the parity of the two states the same. It can be seen that the

nuclear matrix elements will vanish unless $\Delta J = 0$, and there is no parity change. It is possible however, for the operation represented in the matrix elements $|\int \dots| = |\int dv \sum_K v^* \dots \dots \dots G_K U|_{by} \dots$ to alter the symmetry of U to one corresponding to the J and parity of V. This type of operation corresponds to the light particles carrying off the difference $(J_f - J_i)$ in angular momentum and the change in parity.

The polar vector form of interaction adopted by Fermi leads to the nuclear matrix element 1 which cannot possibly change the symmetry properties of U, and hence we have the Fermi selection rule that for allowed transitions $\Delta J = 0$ (no) --- the "(no)" signifying no parity change. The scalar interaction form leads to a scalar operator, so that Fermi rules are also obeyed for this form of interaction. G-T selection rules on the other hand require $\Delta J = 0, \neq 1$ (no) with $0 \rightarrow 0$ forbidden for allowed transitions.

From an empirical standpoint, allowed and forbidden transitions are determined through a comparison of the half-lives of beta-emitters in relation to their nuclear charge Z and energy release W_0 . Since the mean life τ of beta-decay is given by

$$\frac{1}{\tau} = \int_1^{W_0} P dW \dots \dots \dots (7)$$

we have from (5)

$$\frac{1}{\tau} = (G^2 |\int \dots|^2 / 2 \pi^3) f(Z, W_0) \dots \dots \dots (8)$$

where

$$f(Z, W_0) = \int_1^{W_0} dW \cdot W \cdot (W^2 - 1)^{1/2} (W_0 - W)^2 F(Z, W) \dots \dots \dots (9)$$

Various approximations⁽²⁶⁾ are available for $f(Z, W_0)$. For $W_0 \gg 1 + (\alpha Z)^2/2$:

$$f(Z, W_0) \approx u(Z) (\bar{p})^{2s-2} x \left[v(W_0) - w(Z) (W_0 - 1)^3 \right] \dots \dots \dots (10)$$

with
$$u(Z) = \frac{4}{(2s!)^2} (2R)^{2s-2} \frac{\pi |\alpha Z| (1+s)}{|1 - e^{-2\pi\alpha Z}|}$$

$$v(W_0) = (W_0^5 - 10W_0^2 + 15W_0 - 6)/30$$

$$w(Z) = (1/3) \pi |\alpha Z| |e^{2\pi\alpha Z} - 1|^{-1}$$

\bar{p} is some average of the electron momentum. Curves for u , v , and w have been plotted(26).

For $Z \neq 0$, $(W_0-1) \ll 1$, $(W_0-1) \ll 2\pi^2\alpha^2 Z^2$ and $Z > 0$

$$f(ZW_0) \approx u(Z) (\bar{p})^{2s-2} v(W_0) (1 - e^{-2\pi\alpha Z}) \dots\dots\dots(11)$$

for negatron emitters only. While approximations for other cases are given, the above two cover most needs and are quoted here because of their use in a later section of this thesis.

The magnitude of ft constitutes the criterion as to which order of forbiddenness a spectrum belongs. For these purposes three sub-divisions are made; A, constituting the lighter elements, B, consisting of medium and heavy weight elements, and C, which is comprised almost entirely of the naturally occurring radioactive elements. Within each sub-division, distinctions are made as to the order of spectra. Thus group OA constitutes those light beta-active elements, the spectra of which are allowed; group 1A, the spectra of which are first forbidden, etc. A rather extensive table of this type has been compiled by Konopinski(26).

It is interesting to note that the well-known Sargent diagram(28) viz. a plot of $\log \tau$ vs $\log E_{\max}$ for the naturally occurring beta-emitters yields three straight lines. These lines, in the light of present knowledge, may be identified with the 0th, 1st and 2nd order spectra of sub-division C. The relationship is completely obscured if the lighter artificially radioactive beta-emitters are included in the diagram.

II. METHODS OF BETA-RAY SPECTROSCOPY

(a) Counting Procedures

The process of counting is essentially a simple one, involving only corrections for background. Background is due to a number of causes, and is manifested in a definite counting rate which may be a function of the magnetic field of the spectrometer. The materials out of which the counter are made have some slight radioactivity due to contamination, the counter may be photo-sensitive, and in addition cosmic rays will contribute to the background of the counter. Primary or secondary electrons scattered into the counter by the walls of the spectrometer tube also account for some of the background. In order to correct for the background it is usual to determine the counting rate with zero current flowing through the magnet coil, or alternatively, with the spectrometer focussed for electronic energies greater than that given by the upper limit of the spectrum. The difference in the background counting rate affords some measure of the amount of scattering within the spectrometer. The author has assumed a linear relationship between magnet current and background in order to effect a first order correction for the variation of background with the magnetic field. This procedure appears to be a fairly logical one under the circumstances.

Each counting rate determination is made subject to an error due to statistical fluctuations. If it is assumed that the emission of an electron by a radioactive nucleus is a random event, then the laws of probability⁽²⁹⁾ afford a simple means of determining the probable error in the counting rate. If the total number of counts recorded is n , the standard deviation is given by \sqrt{n} , so that the probable error is $0.67\sqrt{n}$. The percentage probable error is therefore $\frac{0.67}{\sqrt{n}} \times 100$. When the background

is taken into account the percent probable error becomes $\frac{\sqrt{n + n_1}}{n - n_1} \times 0.67 \times 100$ where n is the total number of counts and n_1 the number of counts due to the background. The desirability of obtaining a high counting rate relative to the background rate is obvious.

Other considerations apply when coincidence techniques are employed. Dunworth⁽³⁰⁾ has shown that optimum conditions are not obtained by an indefinite increase in source strength. If the efficiency of the beta-counter is ϵ_β (this is determined by the geometry and focussing action of the spectrometer) and that of a gamma counter connected in coincidence with it is ϵ_γ , then the number of beta counts per minute is $N \epsilon_\beta$ and the number of coincidence counts per minute is $N \epsilon_\beta \epsilon_\gamma$, where N is the number of beta disintegrations per minute. It is assumed that every beta-disintegration is followed by one gamma-ray. There may be a background coincidence counting rate due to cosmic rays, but since the gamma counter is placed behind the source at one end of the spectrometer and the beta-counter at the other end at the same level, this coincidence counting rate is negligible. The apparatus employed to distinguish which beta and gamma counts occur simultaneously is termed a coincidence mixer, and due to the fact that the mixer has a finite resolving time, the actual number of coincidence counts recorded will be larger than the number of true coincidences.

$$\text{Let } C_\beta = N \epsilon_\beta$$

$$C_\gamma = N \epsilon_\gamma$$

τ = resolving time of the coincidence mixer in minutes.

Then the recorded counting rate D is given by

$$D = N \epsilon_\beta \epsilon_\gamma + 2C_\beta C_\gamma \tau$$

It is desirable to keep the second term, due to chance counts, small, but for a given ϵ_β , ϵ_γ and τ , this can only be done by reducing N

which will at the same time reduce the first term, representing the coincidence counting rate. Analysis reveals that the maximum useful source strength, or optimum value, is given by

$$N = 1/2 \tau$$

in which case the number of true coincidences is equal to the number of chance counts. The importance of reducing τ is clear.

Calculations of probable errors due to statistical fluctuations are made in a manner similar to that outlined above for counting rate determinations.

(b) Continuous and Line Spectra.

One of the more critical factors involved in the determination of the shapes of continuous spectra is concerned with the preparation and mounting of the source. There are available a number of methods of mounting sources among which the most common are (a) evaporation to dryness of a solution of a salt of the radioactive element, (b) precipitation of the salt of the element out of solution, (c) electrolytic plating of the source material, (d) evaporation of the radioactive material in vacuo, and, (e) in special cases, such as ThB, the collection of the active deposit of a radioactive gas on a metallic foil. Methods (c), (d), and (e) are not always available or desirable and a choice is usually made between methods (a) and (b). As pointed out by Deutsch, Elliott and Evans⁽⁸⁾ the second of these two methods is preferable, when applicable, since it results in a more uniform source. Attempts are made to obtain "thin" sources, since scattering of electrons within the source can change the shape of the spectrum considerably. A source may be regarded as thin when further reduction in thickness does not alter the shape of the spectrum. Once the source has been deposited on a suitable backing it must be covered to prevent loss of material and consequent contamination of the spectrometer.

A convenient process is to dry the source thoroughly and apply a thin coating of a solution of collodion in amyl acetate. The thickness of the backing is also of importance since undue emphasis may be placed on the number of low energy electrons by scattering of electrons through large angles by the backing. Backing thicknesses of the order of 1 mg/cm^2 of mica or aluminum are customary, although very much thinner material has also been used(31),(32). The mounting of the backing is also of some importance, and the general rules to be applied are those which will lead to a minimum number of scattered primary and secondary electrons.

Baffles are employed to reduce the number of scattered electrons entering the counter window. When the shapes of the low energy portion of spectra are being determined, a thin window for the counter is essential. Mica is a very convenient material for windows since it is readily split to obtain laminae of the order of 0.5 mg/cm^2 but laminae thinner than this are obtained only with difficulty and good fortune.

The adjustment of the spectrometer when continuous spectra are studied is discussed in detail by Deutsch, Elliott and Evans⁽⁸⁾. Good resolution is not essential while high intensity is desirable. As a consequence a high degree of magnification may be employed. Nevertheless if end-points are to be determined accurately it is a matter of some importance to use as good a resolution as possible.

For line spectra, good resolution is required if accurate measurements are to be made. Line spectra arise when extranuclear electrons are ejected from their shells by gamma quanta which are radiated from a nucleus in an excited state. The gamma quanta are monochromatic, and each ejected electron receives the entire energy of a gamma quantum, less the binding energy of the electron in its shell. As a consequence, when this process (internal conversion) occurs groups of monokinetic electrons corresponding to K electrons (electrons ejected from the K shell),

M electrons, N electrons, etc., are focussed by the spectrometer and appear as lines superposed on the continuum. The accurate measurement of the energies corresponding to these lines affords an accurate knowledge of the energy of the converted gamma ray, since binding energies are known very accurately from X-ray absorption measurements(33). The extent to which internal conversion occurs is expressed by means of the total conversion coefficient, α , which is defined as the ratio of the number of electrons ejected to the number of quanta emitted. If only those electrons ejected from the K shell are considered, the above definition yields the K conversion coefficient α_K and similarly for the other shells. Therefore

$$\alpha = \alpha_K + \alpha_{L_I} + \alpha_{L_{II}} + \alpha_{L_{III}} + \dots$$

(c) The Kurie Plot.

Beta-ray spectroscopy has as one of its ultimate aims the determination of the disintegration schemes of radioactive nuclei, from which energy levels of nuclei may be determined. Analysis of a beta-ray continuum may be expected to yield some information of value. The Fermi theory of beta-decay outlined previously is concerned with a single spectrum. In essence the theory purports to describe a case of beta-decay in which a nucleus decays to a single lower level by the emission of an electron and neutrino. The resultant spectrum is referred to as simple. If it is possible for the nucleus to decay by beta-emission to more than one lower level, the resultant spectrum is termed complex and consists of the sum of a number of simple spectra, one corresponding to each of the possible modes of decay.

By the application of the Fermi theory it is possible in principle to resolve a complex spectrum into its simple components. The fundamental assumption involved is that the Fermi theory does in fact describe the process. The most convenient way of applying the Fermi

theory in any particular case is by means of the Kurie Plot⁽³⁴⁾.

Referring to equation 42 of Fermi's original paper⁽²⁵⁾ we have, converting to rationalized relativistic units,

$$N' \propto F(\eta, Z) \eta^2 (W_0 - W)^2 \dots\dots\dots(12)$$

where N' is the number of electrons per unit momentum interval, and

$$F(\eta, Z) = \frac{4(2R\eta)^{2s-2} e^{\pi\alpha ZW/\eta} |\Gamma(s + i\alpha ZW/\eta)|^2}{[\Gamma(2s + 1)]^2} \dots(13)$$

(The other symbols have the same significance as those of equation (5) with η replacing p as the symbol for electron momentum).

It should be noted as mentioned previously, that the expression for $F(\eta, Z)$ differs from that given by Konopinski⁽²⁶⁾, which contains an added factor $\frac{1+s}{2} = \frac{1 + (1 - \alpha^2 Z^2)^{1/2}}{2}$. However, for purposes of a single spectrum, in which Z is constant, the factor $\frac{1+s}{2}$ merely adds another multiplicative constant and is not of great importance.

From equation (12)

$$\frac{N'}{\eta^2 F(\eta, Z)} \propto (W_0 - W)^2$$

$$\therefore \sqrt{\frac{N'}{\eta^2 F(\eta, Z)}} \propto (W_0 - W) \dots\dots\dots(14)$$

Therefore a plot of $\sqrt{\frac{N'}{\eta^2 F(\eta, Z)}}$ vs. W should yield a straight line. By extrapolation W_0 may be found. For use with spectrometric data, equation (14) can be put into a more convenient form. Since the momentum interval of the lens spectrometer is a constant fraction of the momentum

$$N' \propto \frac{N}{\eta}$$

where N is the net counting rate. But η is proportional to the magnet current which is in turn proportional to the voltage V developed across standard resistance in series with the magnet coil.

Hence $V \propto \eta$

and $\sqrt{\frac{N}{V^3 F(\eta, Z)}} \propto (W_0 - W)$

or $\sqrt{\frac{N}{V^3 F(\eta, Z)}} = K(E_0 - E) \dots\dots\dots(15)$

where E is the electron energy, and E_0 the maximum electron energy, both in Mev. K is a constant.

For a simple spectrum, the application of equation (15) constitutes a test of the Fermi theory without prior knowledge of E_0 and in addition possesses the virtue that the test requires only the judgment as to how well experimental data are represented by a straight line. It is very similar to the plotting of an exponential function on logarithmic graph paper. Not all spectra can be expected to yield straight lines however. The assumption made (equation (5)) that the nuclear matrix elements are independent of W is doubtless true for allowed spectra but may not be true for all forbidden spectra. In any case, straight lines are yielded for a large part of the distribution of all allowed and many forbidden spectra(32). It is therefore of some significance if a straight line is obtained.

If a spectrum is complex, resolution can be effected by extrapolation of the straight line followed by "geometrical" subtraction of the straight line from the original curve, i.e., by squaring the ordinates at selected abscissae, subtraction of these ordinates, taking the square roots of the differences, and replotting. In this way a number of straight lines, corresponding to the number of component spectra, will be secured. From the straight lines one can obtain the end-points of the respective distributions and in addition one can use the data therefrom to plot the individual spectra.

Because of the compounding of errors the accuracy with which the

end points may be determined becomes poorer at each successive subtraction. In addition caution must be exercised in attributing spectra to every straight line obtained. Quite often, for instance, the Fermi theory predicts fewer lower energy electrons than are obtained in practice. This may be due to scattering effects at the source, or as a recent paper indicates⁽³²⁾, it may be due to inadequacy of the Fermi theory. The plot may therefore curve upward in the low energy region, and by following out the procedure outlined above, it is possible to construct straight lines ad infinitum. This can be avoided by making certain that the straight line drawn does take in a considerable region of the spectrum.

The Fermi function (equation (13)) must of course be determined for the particular Z required. This involves a good deal of labour because of the factor containing the Γ function with complex argument. For very low Z , the approximation $Z = 0$ may be made, for which $F(\eta, 0) = 1$. Kurie, Paxton and Richardson⁽³⁴⁾ have given an approximation valid for $Z < 29$. Doubtless other approximations could be obtained but the total labour involved would probably exceed that required to calculate the exact function. Calculations have been made for the exact function for $Z = 58$ and $Z = 59$ (Figure 3) utilizing an asymptotic expansion for $\Gamma(z)$ given by Copson⁽³⁵⁾. Some saving in time is effected by converting to polar coordinates and employing the identity

$$|\Gamma(z)|^2 = \Gamma(z) \Gamma(\bar{z}).$$

(d) Gamma-Ray Measurements.

The existence of possible gamma rays may be deduced from the Kurie Plot but in any case a systematic search should be conducted for gamma rays. Spectrometric determinations of gamma ray energies are made

by a study of the secondary electrons which they produce. Use is made of three effects produced by the interaction of gamma rays with matter: (1) photoelectric effect, (2) Compton effect, and (3) pair production. The latter effect is possible only with gamma rays of energy greater than 1.02 Mev, and involves the creation of a positron and negatron, the surplus energy being shared between the two. The probability of pair formation increases rapidly with nuclear charge and gamma ray energy and is of major importance for hard gamma rays. Photoelectric effect is similar to internal conversion, the gamma quantum ejecting an electron from one of the inner shells of the atom, and being completely absorbed in the process. The ejected electron has therefore the entire energy of the gamma ray less the binding energy of the electron in the shell whence it came. The photoelectric effect can occur in any shell of the atom but is preponderant for the K shell, the probability of the ejection of an L electron being about 1/5 that of a K electron. The Compton effect involves the transmission of a portion of the energy of a gamma quantum to a free electron in an elastic collision, the gamma quantum being scattered with a reduced frequency. The spectrum of Compton electrons is therefore a continuum with a fairly sharp maximum corresponding to electrons emerging at an angle of 0° to the initial direction of the gamma quanta. This energy is given by

$$E = \frac{2\alpha h\nu}{1 + 2\alpha}$$

where ν is the frequency of the gamma quantum, h is Planck's constant, and $\alpha = h\nu/mc^2$.

The photoelectric effect allows the most accurate determinations of gamma ray energies. The general procedure is to enclose the source in a capsule of low Z (such as copper) sufficiently thick to stop all primary

beta-rays, and then to place in front of the capsule a thin radiator of high Z such as lead. The spectrum of the secondary electrons is taken, and will consist of photoelectrons and Compton electrons. Replacing the lead radiator by a thick radiator of low Z (aluminum), the secondary spectrum consisting only of Compton electrons, is found. Comparing the two spectra it becomes possible to distinguish the photoelectric lines with certainty. The photoelectric cross-section is proportional (approximately) to Z^4 (36), whereas the Compton cross-section is largely independent of Z. This explains the reason for using a heavy element for the radiator when photoelectrons are sought. The thickness of the radiator must be chosen to suit the gamma rays being studied, and must be thin enough so that no photoelectrons are slowed down into the Compton region. The relative intensities of Compton and photoelectrons are determined by the energy of the gamma rays. The cross section for the production of Compton electrons is roughly proportional to $1/\nu$, whereas that for photoelectrons is approximately proportional to $(Z/\nu)^3$. Where numbers of gamma-rays of differing energies are emitted a corresponding number of K and L lines will be observed. In such cases it is possible that a K line of one gamma ray may coincide with the L line of another, in which case two radiators of differing Z will have to be employed to allow unambiguous interpretation. Relative intensities may be found if allowance is made for the energy variation of the photoelectric cross-section.

Using the three effects discussed above it is possible to find complete solutions to all the problems of gamma ray spectroscopy (37). It must be borne in mind that the intensity of secondary electrons is very much smaller than that of primary electrons and the use of the spectrometer in such studies requires, therefore, sources of high activity (38).

give a coincidence rate per beta particle which is independent of beta energy. A deviation from constancy is a definite proof of complexity. In the latter case, the end-points of the component spectra may be determined if the entire spectrum is investigated.

III. APPARATUS

(a) The Spectrometer

The spectrometer used in these experiments is of the thin lens type described by Deutsch, Elliott and Evans⁽⁸⁾ and was constructed by the National Research Council of Canada. A view of the spectrometer is presented in Plate I and a schematic diagram is given in Figure I.

The magnet consists of a coil wound in four sections. Each section, of 900 turns of #10 gauge double cotton covered copper wire, is wound over the previous section with a winding of 1/4" O.D. copper tubing separating successive sections. The copper tubing is provided for purposes of cooling. The drum on which the coil is wound is formed from two brass annuli 5/8" thick. The inner diameters of the annuli are 8" and outer diameters 24". A distance of approximately 7" separates the annuli. The spectrometer tube, about 40" in length, outer diameter 7 7/8" and wall thickness 1/8", fits through the centre of the coil drum and is held in position by means of two sets of three point suspension screws mounted on the brass annuli, one set on each annulus. The tube can therefore be moved along the axis of the coil to secure a wide range of magnification.

The tube is terminated at each end by brass discs 5/8" thick, which have circular grooves cut in them to receive rubber gaskets. Another set of brass discs with flanges corresponding to the grooves are provided one of which is used as a source holder, the other as a Geiger counter holder. Vacuum connections are made with the aid of a stopcock grease. The baffles are made of aluminum 1/8" in thickness and held in position by an aluminum tube which fits into the interior of the spectrometer tube. A cylinder of lead 6" in length is mounted at the centre of the defining baffle to prevent gamma radiation from reaching the counter.

The theory and operation of the instrument have been described very fully by Deutsch, Elliott and Evans⁽⁸⁾, and need not be given here. One point that should be noted is that since no iron is employed in the construction of the spectrometer, the magnetic field is proportional to the magnet current, and therefore the instrument may be calibrated in terms of the current.

(b) Power Supply and Magnet Current Regulator.

The resistance of the entire magnet coil is about 15 ohms, and can carry a maximum of about 20 amperes, provided the proper degree of cooling is maintained. An adequate supply for all purposes must then deliver 6 Kilowatts at 300 volts. A motor-generator set capable of satisfying these requirements was assembled by utilizing a 20 H.P., 220 volt, three phase, 1200 r.p.m. induction motor which was available. This motor is coupled to a 12 H.P., 440 volt, 1600 r.p.m. shunt wound D.C. generator. By supplying the field with separate excitation at 440 volts, the generator was found to deliver somewhat over 300 volts. The excess power supplied by the motor is used to drive a similar generator coupled to the first by an extension of the drive-shaft. The excitation for the generator field is obtained by connecting in series the armatures of two 1/3 H.P., compound wound, 220 volt, D.C. motors operated as generators, with the series portions of the field windings removed. As a consequence of the change from compound wound to shunt wound field windings, these generators exhibit poor regulation. Whereas their nominal speed is 1800 r.p.m. it was found necessary to raise the speed to 2500 r.p.m. to secure the rated output. In addition it was found desirable to supply them with separate excitation in order to reduce the current drain. This is done from a rectifier unit the output of which is controlled by the magnet current regulator. As a matter of convenience the field windings

are connected in parallel for this purpose. The small generators are driven from the main shaft by belt and pulley coupling.

Measurement and stabilization of the magnet current is effected by means of a standard resistance - potentiometer arrangement. The standard resistance is wound from #12 gauge manganin wire in 2 sections of equal resistance (0.14Ω) and is water cooled. By means of a switching arrangement, the two sections of the standard resistor may be used in series, parallel, or singly. In this way the entire current range may be covered. A Rubicon Type S potentiometer, Eppley standard cell, and Rubicon Spotlight model galvanometer (resistance 25Ω , sensitivity 0.25μ amps/mm) are used in conjunction with the standard resistance. The Brown convertor of the regulator is inserted in series with the galvanometer as shown at the points A and B of the circuit diagram (Figure 2).

The mode of operation of the regulator is as follows. Provided that the voltage developed across the standard resistor is exactly balanced by the potentiometer, no current will flow through the galvanometer or converter. If, however, the magnet current falls or rises, so that the balance is destroyed, an error voltage is developed across the input to the Brown converter, and a small 60 cycle (square wave) current will flow through the primary of the transformer. The signal is amplified through two stages and applied to the 6SC7 phase inverter. The resulting two signals, 180° out of phase, are applied to the grids of a 6N7 operated as a grid controlled rectifier. The amplified signal, thus converted to D.C., is applied to the grids of two 6L6 tubes connected in parallel, with the exciter field coil as load resistance. The plates of the 6L6 tubes are supplied by a 400 volt, 150 ma. power pack.

Provided the phasing is correct, the error voltage causes a change in exciter field current, the effect of which is to change the output voltage of the main generator in an attempt to reduce the error voltage to zero.

Under these circumstances, the error voltage is never actually reduced to zero but can be kept within a limit which depends upon the overall amplification of the circuit. It will approach zero only for infinite amplification.

It will be noted that a form of voltage regulator is also employed, the input being derived from the output of the exciter, and after amplification being applied to the grids of the 6L6 tubes. This was found necessary in order to reduce "hunting". The cause of hunting is attributed to the inherent time lags possessed by all components of the circuit, including magnet and generator, and hence the action of the regulator is always such as to correct for a past error. Over-correction results with consequent sustained oscillations. There are a number of methods available for damping out sustained oscillations⁽⁴⁹⁾, the one employed being the most successful in the present case.

A constancy of magnet current to within one part in one thousand has been aimed at and secured with the regulator. One difficulty encountered has been concerned with the reduction of a 20 cycle component present in the output of the generator. This component is constant in magnitude, and is probably due to permanent magnetization of the generator casing, since the frequency of the component is identical with the speed of rotation of the generator. It may also be due to non-uniform rotation of the driving motor. Attempts to reduce it by filtering have proven useless, owing to the low impedance of the generator armature. Instead, use is made of the fact that the magnitude of the component is constant,

and the generator is run at a sufficiently high voltage output to cause the 20-cycle variation to be less than 0.1% of the D.C. component. This involves the dissipation of a good deal of power (by means of resistors in series with the magnet) but appears to be the best that can be done under the circumstances.

The final criterion as to performance of the regulator was made in three ways. For slow variations, the galvanometer was employed, while for the higher frequencies, an oscilloscope connected across a resistor in series with the magnet was utilized. Finally a coil of 1500 turns of copper wire, 6" in diameter, was placed at the centre of the magnet coil, and the induced emf measured on an oscilloscope. In this way it was found that the r.m.s. magnetic field due to components of frequencies \geq 20 cycles could be reduced to less than 0.05% of the steady magnetic field. This is not surprising in view of the presence of the heavy brass plates forming the magnet coil. The effect of eddy currents within these plates is considerable.

In general, the regulator has proven quite adequate, but not ideal, in its performance. There remains a slight instability in its action which can probably be removed, but a more serious difficulty is due to the poor characteristics of the exciter - a large increase in exciter input resulting in a disappointingly small increase in generator output. This condition can be rectified and the writer takes this opportunity of suggesting the following method for doing so. The exciter may be dispensed with entirely, and the generator operated in shunt, with a rectifier delivering 0.5 amps at about 250 volts in series with the field winding. Regulation could be effected by inserting two 6AS7 twin triodes (connected in parallel) in series with the field winding and power pack. The output of the phase sensitive rectifier could then be applied to the grids of the 6AS7 tubes. Such a procedure would also

allow reduction in the gain of the amplifier since at present the amplifier is operated at high gain in order to compensate for the poor performance of the exciter.

(c) Neutralization of stray fields.

At low and moderate electronic energies, the focussing of the spectrometer is adversely affected by even small magnetic fields which are not parallel to the axis of the spectrometer tube. For this reason the spectrometer is aligned so that its axis lies in the earth's magnetic meridian. The vertical component of the earth's field is neutralized by the use of a pair of coils wound on a frame about the spectrometer, one above and one below the spectrometer.

The spectrometer is, however, very poorly situated with respect to the McGill cyclotron, and when the cyclotron is in operation the stray field near the spectrometer bears no relation either in magnitude or direction to the earth's field. The vertical field is increased about 350% and the component of the horizontal field perpendicular to the axis of the spectrometer assumes a value about 200% greater than the normal vertical component of the earth's field. For this reason a second pair of coils has been wound around the spectrometer, one on each side of the spectrometer.

The problems met in this regard are best summed up in the following table, in which the ampere-turns required to neutralize all stray fields are given for various positions along the axis of the spectrometer. The positions referred to are given in inches from the median plane of the spectrometer coil. S refers to south and N to north. The two sets of measurements were taken 6 weeks apart.

<u>Position</u>	<u>Ampere-Turns</u>			
	<u>Cyclotron Off</u>		<u>Cyclotron On (620 Amps)</u>	
	<u>Vertical Fd.</u>	<u>Horiz. Fd.</u>	<u>Vertical Fd.</u>	<u>Horiz. Fd.</u>
20S	40	4.0	138	-
10S	37	2.4	135	-
10N	36	1.8	133	-
20N	32	1.6	128	-
				<u>(580 Amps)</u>
20S	39	5.0	117	84
10S	36	1.8	112	87
0	32	2.5	111	85
10N	32	3.0	111	76
20N	28	4.0	103	77

It will be noted that at the time of taking the measurements the horizontal field within the room had assumed a direction slightly different from the magnetic meridian. Several effects are noted: (1) a slight dependence of the field within the room (cyclotron off) with the previous history of operation of the cyclotron (2) a definite dependence upon cyclotron current (3) a rather large variation of the magnitude of the field with distance along the axis of the spectrometer.

The problem was partially solved by using the spectrometer only when the cyclotron was not in operation. A careful check was kept on the proper value of current employed to secure neutralization of the stray fields. Insofar as the variation of field with distance along the axis is concerned, the best that could be done was to use an average value.

Measurements were made using a galvanometer in conjunction with a 'flip' coil 6" in diameter, and wound with 1500 turns of #29 gauge copper wire.

Current for the compensating field coils is supplied by a 2 KW, 30 volt D.C. generator driven from the shaft of the main generator. Controls and meters are mounted on the main panel, a view of which is presented in Plate II.

(d) The Vacuum System.

For a good part of the work, a pressure less than 1 micron was maintained within the spectrometer tube using only Megavac mechanical pump. When the low energy region of the beta spectrum was studied, a glass, three-stage, fractionating diffusion pump was added, with which pressures were reduced to 5×10^{-5} mm or lower. The speed of the diffusion pump (Distillation Products Type GF-25A) is rated at 25 litres/sec. at 10^{-4} mm.

Two vacuum gauges were used: a thermocouple type gauge with a lower limit of ~ 1 micron (calibrated against a McLeod gauge) and an ionization gauge (VG-1A Distillation Products) with control unit (EG200 Distillation Products).

(e) Beta and Gamma Counters.

Schematic diagrams of the types of beta and gamma counters used are given in Figure 4 (A and C). Two gamma counters were employed differing only in their cathodes, one of which was copper, wall thickness 0.050", the other of lead, wall thickness ~ 0.007 ". Lack of source material prevented complete calibration of these counters, but the efficiencies were measured at ~ 1.2 Mev. using the gamma rays from a small sample of Co^{60} (0.127 ± 0.001 rd). For the copper counter the calibration curve was deduced using the results of von Droste's⁽⁵⁰⁾ theoretical and experimental investigations for a brass-walled counter. The lead counter

is thought to be relatively more efficient than the copper counter for the lower energy gamma rays, but the shape of the curve is unknown. The results are shown in Figure 4(B). For the copper and lead counters the data have been corrected to give the efficiency for a total solid angle of 4π steradians.

The beta counters are of the end-type, copper walled, with brass bases shaped to fit into the end plate of the spectrometer. A window diameter of 0.5 cm was used throughout, with mica windows 0.5 mg/cm^2 and 0.75 mg/cm^2 thick. The counters were normally filled to a pressure of 10 cm with a 90% argon, 10% ethyl alcohol mixture. Lower pressures were tried with the beta-counters, particularly for the low energy end of the beta spectrum, but with unsatisfactory results. At the lower pressures spontaneous discharge was very prone to occur. Plateaus of the order of 200 volts in extent with a 3% rise per 100 volts were secured for the gamma counters. For the beta counters a rise of 7 or 8% per 100 volts over the plateau was considered satisfactory. Usually the plateaus were very narrow - of the order of 50 volts.

(f) Counting Circuits.

Components of the counting circuit consisted of high tension supplies for the Geiger counters, grounded grid preamplifiers, scaling units and an oscilloscope. For coincidence work three scaling units were used, separate supplies for the counters, and a coincidence mixer.

The National Research Council MK III voltage doubler rectifying units were at times employed as H.T. supplies. While the output of this unit is well filtered, the voltage is unregulated and shows a tendency to drift. A supply using a cathode ray transformer and simple regulating circuit was built and proved much more satisfactory. It shows, however, a slight drift of voltage with time. The output is constant to within

$\pm 0.3\%$ for line voltage variations of $\pm 10\%$. The performance could undoubtedly be improved by regulation of the filament current, if this were desired. Ripple is of the order of 0.001% . The Atomic Instrument Company scale of 64 units were employed with preamplifiers using cathode follower output, placed near the counters. For high counting rates a National Research Council scale of 128 unit was used.

The resolving time of the coincidence mixer (NRC) was measured using the relation $C_{\text{random}} = 2C_1 C_2 \tau$ with two independent sources. It was found to be $(0.62 \pm 0.03) \times 10^{-6}$ sec.

IV. EXPERIMENTAL

(a) The Source.

The cerium source used was obtained from the Atomic Energy Laboratory of the National Research Council. The method of separation of the cerium from other fission products is unknown to the author. It was ascertained, however, that the source had been prepared by evaporation of a salt of the cerium from solution. The material was deposited on a small piece of mica, $\frac{5}{8}$ " square and about 10 or 11 mg/cm² thick. It covered a circular area about 1 mm in diameter, and was covered with a thin layer of collodion. During the process of covering, some of the cerium had migrated away from the main body to a maximum distance of 3 or 4 mm, but the majority still remained concentrated at the original point of deposition. It is assumed that the method of preparation resulted in a "thin" source. The activity was reported to the writer as approximately 5 microcuries. The source mounting consisted of a brass cup 1" in diameter filled with apiezon Q sealing compound to which the mica backing was affixed.

The writer was given the opportunity of measuring the K conversion line (section V) on one of the NRC lens spectrometers at the time the source was obtained. This measurement served as a fiducial point for purposes of temporary calibration of the McGill spectrometer.

(b) Alignment and Calibration of the Spectrometer.

It is essential that the axis of the spectrometer tube and the magnetic axis of the coil coincide accurately and the satisfaction of this condition constitutes proper alignment of the spectrometer. The tube is not of perfectly circular cross-section, but this is immaterial. The axis of the spectrometer tube may be defined as the straight line joining the centres of the source and counter window. It is desirable

of course that the circular baffles have centres which lie on this axis.

The experimental procedure for aligning the tube involves the measurement of a monoenergetic group of electrons, the group in this case constituting the conversion electrons of the K line mentioned above. The tube is moved radially until maximum intensity and theoretically predicted line width and shape are obtained.⁽⁸⁾ Inasmuch as the author had the good fortune to perform such a measurement on a properly aligned spectrometer of almost identical construction, it was merely necessary in the present case to achieve a similar line shape, width, along with maximum intensity. This procedure allowed alignment and calibration to be achieved simultaneously.

At a much later date a source of 5 mc of mesothorium₂ became available and a more accurate calibration of the spectrometer was performed. The MsTh₂ was enclosed in an "activator" and the active deposit collected on a small aluminum foil 0.0005" thick. After about 10 hours exposure, a square of the foil 1/8" on an edge was removed, and used as a source. Both sides of the foil were activated. The standard used was the F line of conversion electrons from ThB which has been measured by Siegbahn⁽⁵¹⁾ as 1383.8 gauss cm. with a probable error of 0.2 - 0.3%. All data have been corrected to this calibration.

Ideally, a thorough calibration should entail the measurement of two lines differing appreciably in energy. This is desirable since improper alignment can be compensated for by improper neutralization of stray fields at one particular electron energy. Calibrations at two points which agree within 1% is considered satisfactory⁽⁸⁾. Often, in addition to the Thorium F line, secondary electrons produced by annihilation radiation (0.510 Mev) of Cu⁶⁴ positrons is used for this purpose. Since

activity of this sort was lacking the procedure followed was simply to neutralize stray fields as accurately as possible, and then to proceed with alignment. The alignment achieved was maintained throughout all experiments. When the arrival of the MsTh_2 made possible the measurement of two accurately known lines differing in momentum by 367 gauss-cm, an assessment of the value of the procedure could be made.

The dimensions of the defining baffle annulus were 15.0 cm O.D. and 14.0 cm I.D. This baffle was placed midway between source and counter at a distance of 52.8 cm from the source. A solid angle of 0.065% of 4π steradians was thus utilized. The counter window was 0.5 cm in diameter, and 0.5 mg/cm^2 thick. (Figure 4C).

(c) The Beta-Ray Spectrum.

For the portion of the spectrum $> 90 \text{ Kev.}$ a counter with a mica window 0.75 mg/cm^2 thick was used. A pressure < 1 micron was maintained in the spectrometer tube using the mechanical pump alone. Conditions were otherwise identical with those employed for the calibration.

Particular care was taken in measuring the K and L conversion lines, and a good deal of time spent in determining the end point of the spectrum. Measurements were made at the peak of the K line at the beginning and end of each day's work in order to make certain that the stray fields were being neutralized. Every day's measurements were corrected for decay of the source and the rather rare changes in counting rate by relating the measurement of the K line to that taken on the first day. It was noticed that counters had a tendency to give a higher counting rate on initial filling, but that constancy in this regard was obtained after about one day.

The value of the half-life was found incidentally to be about 285 days, and although this value is based upon intensity measurements

of the K line (time interval 80 days), it must necessarily be the half life of Ce^{144} even though the line may be due to Pr^{144} . This follows from the fact that Pr^{144} is in secular equilibrium with Ce^{144} .

Since the momentum interval of the spectrometer is a constant fraction of the momentum, economy was effected in taking measurements by employing a logarithmic momentum scale. Counts were recorded for successive momentum values each 2% greater than the preceding value. At each point two or three runs of from 2 to 5 minutes each were made. The spectrum was examined twice and for the second determination, the momentum values used differed from the first set by 1%. Accordingly the spectrum has been plotted for momentum values each of which is 1% greater than the value immediately preceding it. The two sets of curves agreed for the most part within 3% or better. Average values at each point are employed to plot out the final spectrum. The counter background was about 16 counts per minute at zero current, and about 8 counts per minute beyond the upper end of the spectrum. The line width was 0.034.

For the lower end of the spectrum which was measured at a later period the diffusion pump was added. Attempts to use lower pressures in the counter failed completely however. Behaviour of the counter at low pressures (< 6-7 cm Hg) was characterized by a slight plateau, if any, and a tendency to break down spontaneously. Little difference in counting rate was noted when the 0.75 mg/cm² window was replaced by the 0.5 mg/cm² window. Counts were recorded to energies as low as 530 gauss-cm (24 Kev).

(d) Measurement of Gamma Ray Energies.

Initial rough measurements indicated that the gamma radiation emitted by the source was mainly soft with a weak hard component. At the time of taking the measurements the source had decayed to about 85% of the

initial 5 microcuries. Since the lowest practicable activity for use in the measurement of gamma rays with the spectrometer is 10^7 quanta per minute of each gamma ray (5 microcuries)(8) there did not seem much likelihood of success in the measurement of gamma ray energies using the spectrometer.

Nevertheless an attempt was made. The mica on which the source was mounted was cut down to a circle 1 cm. in diameter with the source at the centre of the circle. A copper capsule was made in the form of a shallow cup, with inside diameter 1 cm, and the source placed in the cavity. A copper cover with a short stem was placed over the back and the stem pressed into the apiezon Q which filled the source holder. The wall thickness of the capsule and backing was 1.6 grams/cm^2 , calculated(36) to stop all primary electrons. A lead radiator 29 mg/cm^2 was placed over the face of the capsule.

The counter was replaced by one with a 1.5 cm diameter window (effective source size ~ 1.3 cm diameter) and thickness 1.5 mg/cm^2 . The baffles were changed so that the solid angle subtended at the source was increased to 0.31% of 4π steradians. This represented an increase in solid angle of about 470%. With the baffle system in use the solid angle could not have been increased very much more without increasing the magnification of the spectrometer. A magnification of 2 would have required a very much thicker window about $2\frac{1}{2}$ to 3 cm. in diameter, so further changes were not made.

As will be seen in the next section, these experiments were not fruitful, and an attempt to obtain information by means of absorption in lead was made. A copper cathode gamma counter was placed in a lead castle and the source inside its copper capsule was placed about 4 inches distant from it. This entailed very poor geometry but was necessary in

order to secure a counting rate significantly different from the background count, which was 28 counts per minute. The net rate without absorber was 72 c.p.m. Thickness of lead up to 5/8" were used, placed rather closer to the source than to the counter.

(e) Coincidence Experiments.

Two types of coincidence experiments were performed. A disintegration electron-conversion electron experiment was performed to determine whether or not the gamma rays giving rise to the conversion electrons were associated with the high energy beta ray spectrum. Since the source backing was $\sim 11 \text{ mg/cm}^2$ thick no coincidence experiments involving low energy beta rays were attempted. In this experiment the solid angle subtended at the source by the baffle was maintained at 0.31% of 4π steradians, and a counter with 0.5 cm diameter window, 0.5 mg/cm^2 thick was used. Magnification of the spectrometer was unity. A beta counter with window 1.5 cm in diameter, 10 mg/cm^2 thick, was placed behind the source with 0.5 g/cm^2 of aluminum between the counter and source. In this way only those beta rays with energy greater than about 1.1 Mev. were measured by this counter. The spectrometer was set on the K conversion line and coincidences recorded for 92 minutes.

A second set of coincidence experiments was attempted in an endeavour to verify the complexity of the beta spectrum as indicated by the Kurie Plot. The spectrometer was set up as for the previous experiment, but the beta counter behind the source was replaced by a lead cathode gamma counter. A sheet of brass 1/16" thick placed between the gamma counter and source prevented the high energy electrons from penetrating to the sensitive volume.

Beta-gamma coincidences were recorded at 5 points of the beta-spectrum. The points chosen corresponded to electron energies of 1.53,

0.697, 0.312, 0.233 and 0.070 Mev. At the last named point, a sheet of lead 1/8" thick was placed between the gamma counter and source and coincidences again recorded.

V. RESULTS

(a) Calibration.

The results of the calibration of the spectrometer are presented in graphical form in Figure 5. Taking the thorium F line at 1383.8 gauss-cm as standard, the I line is measured at 1764 gauss-cm, i.e., 3/4% higher than the accepted value of 1751 gauss-cm. The relation $H\rho = 4885 V$ is obtained, where $H\rho$ represents momentum in gauss-cm, and V the voltage across the standard resistance in series with the magnet coil, as measured by the potentiometer.

(b) The Beta-Ray Spectrum.

The spectrum is shown in Figure 6. The method of plotting the spectrum distorts the shape, since the momentum is plotted on a logarithmic scale.

In order to maintain clarity, the complete curve has not been drawn through the points. For the same reason the standard practice of drawing vertical lines through the points to indicate probable errors has not been followed for every point. Instead this has been done for every tenth point only, but this procedure will give an indication of the magnitude of the errors existing throughout the spectrum.

The end point of the spectrum as obtained by inspection is 3.19 ± 0.14 Mev. The error has been estimated from the known resolution of the spectrometer (0.034) and allowance made for an experimental uncertainty of 1% in the calibration.

The K conversion line is measured at 1056 gauss-cm. (90.1 Kev) and the L conversion line at 1264 gauss-cm (125 Kev). It is believed that these measurements are accurate to within 1%, the experimental uncertainty in calibration.

The Kurie Plot for the combined spectra is given in Figure 7.

It is immediately evident from Figure 6 that two major distributions are involved in the complete distribution, one of which must be due to Ce^{144} and the other to Pr^{144} . There are clear indications that one of these distributions has an end point at about 0.3 Mev. and the other at about 3.19 Mev. Since the half-life of Pr^{144} is very short compared with that of Ce^{144} , it was assumed that the upper end of the distribution was due to Pr^{144} negations. Consequently the Kurie plot has been drawn up using the Fermi function calculated for $Z = 59$ for $H\rho > 2000$ gauss-cm (0.275 Mev), and for $Z = 58$ for $H\rho < 2000$ gauss-cm. At $H\rho = 2000$ gauss-cm an average value for $F(\eta, Z)$ has been used. This procedure, it is believed, introduces only slight errors in the region about $H\rho = 2000$ gauss-cm.

The existence of five separate spectra is indicated with end-points at 3.06 ± 0.09 Mev., 1.05 ± 0.05 Mev., 0.517 ± 0.036 Mev., 0.303 ± 0.020 Mev., and 0.179 ± 0.015 Mev. The errors have been estimated on a basis of a 1% error in calibration of the spectrometer, and allowance made for the uncertainty involved in representing the data by straight lines. This is particularly true for the two spectra with the lower energy end-points.

(c) Gamma-Ray Energies.

The presence of conversion lines makes possible the energy determination of one of the gamma-rays with an accuracy corresponding to that with which the line itself is measured. Reasons are advanced in the next section for belief that the K and L lines are due to conversion of a gamma-ray emitted from the Pr nucleus. This leads to an energy of $0.132 \pm .001$ Mev. for the gamma-ray in question. The error in the K binding energy of 41.8 Kev⁽³³⁾ is assumed to be negligible compared with the 1% error in the determination of the energy of the K line (90.1 Kev).

Little information of significance was obtained from the attempt to measure the energies of photoelectrons with the spectrometer. For the most part counting rates about 25% of the counter background were recorded. In a region of poorly defined extent about 130 Kev., the net counting rate rose to about 75-100% of the background. There seems a slight indication, therefore, that a gamma-ray of about 0.22 Mev. is emitted.

The results of the absorption experiment are presented in Figure 9 in which $\ln 10N$, where N is the net counting rate, is plotted against thickness of lead absorber in cm. By the usual form of analysis, the existence of three gamma-rays is indicated with absorption coefficients of 0.70 cm^{-1} , 3.92 cm^{-1} and 12.5 cm^{-1} . The corresponding gamma-ray energies⁽⁵²⁾ are 1.20 Mev., 0.30 Mev., and 0.17 Mev. respectively. These values are believed to be highly underestimated because of the poor geometry, and are discussed further in the next section.

(d) Coincidence Experiments.

Results of the coincidence experiments are presented in the following table in which Min. represents duration of the experiment in minutes; $H \rho$ the electron momenta in gauss-cm; E the electron energies in Mev; C_e , C_β , C_γ , C_t , C_r and C_c are the counting rates in counts per minute for conversion electrons, disintegration electrons, gamma-rays, total coincidences, random coincidences and true coincidences, respectively. The coincidence mixer resolving time is $(0.62 \pm 0.03) \times 10^{-6}$ sec.

Probable errors are shown for all coincidence counting rates. The errors in C_e , C_β and C_γ are negligible compared with those for the coincidence counting rates.

VI. DISCUSSION

One of the points of interest is the identification of the element giving rise to the K and L conversion lines. An obvious method for settling this question is to compare the energy difference between the K and L lines as measured on the spectrometer with that calculated from X-ray data.

The energy of the K group of electrons has been measured as 90.1 Kev. This is based upon the calibration of the spectrometer as given in Figure 5 in which the thorium F line is taken as the calibration point. It is to be noted that the measured value of the I line on this basis is 3/4% too high, a comparatively large error considering the small energy difference. An error of this magnitude can also be expected in the measurement of the L conversion line relative to the K conversion line, or perhaps a somewhat lesser error since the K-L energy difference is smaller than the F-I energy difference. Accepting the energy of the K group as 90.1 Kev, the energy of the L group is 125.2 Kev with an estimated error of less than 3/4% (i.e. less than 1.7 Kev). Conversion can take place in either the Pr or Nd atom, and referring to⁽³³⁾ the following table can be constructed:

<u>Element</u>	<u>Energy Difference (Kev)</u>			
	<u>K-L_I</u>	<u>K-L_{II}</u>	<u>K-L_{III}</u>	<u>K-L_{mean}</u>
Pr	35.00	35.40	35.87	35.45
Nd	36.25	36.67	37.18	36.72

It would appear from the measured value of (35.1 ± 1.7) Kev that the conversion takes place in the Pr atom, but the limits of error make it impossible to be completely certain. However, the 3/4% error

assigned to the measurement of the L line relative to the K line is felt to be somewhat overestimated. On the whole it seems most probable that the lines are due to gamma rays emitted by the Pr nucleus. Another reason for believing this to be so is revealed upon analysis of the Kurie Plot (Figure 7).

If the "straightness" of lines in a Kurie Plot is to be accepted as a criterion of the existence of spectra in a complex distribution, then there remains little doubt that there are present three spectra with end points 3.06, 1.05 and 0.517 Mev. in the high energy region of the distribution. These data are indeed well represented by straight lines. There is however a little uncertainty as to how the remainder of the data is to be treated. In the region about 0.3 Mev. the line as drawn falls somewhat below the data. This may be attributed to a number of reasons. Firstly there is a sharp break in the distribution in this region (See Figure 8) and owing to the finite resolution of the spectrometer the data in all probability do not indicate faithfully the sharpness of the break. Secondly, it is in the region where the change from $F(\eta, 59)$ to $F(\eta, 58)$ is made. Thirdly, the process of subtraction in this neighborhood involves the difference of two quantities which are of almost equal magnitude, and finally, as a recent paper by Cook and Langer⁽³²⁾ shows, the Fermi theory as applied to Cu^{64} predicts too few negatrons in the low energy portion of the spectrum. If this is true in general one might expect that in the region in question, (due to the inadequacy of the Fermi theory), there would be an excess of negatrons belonging to the three higher energy spectra. For energies less than 0.3 Mev however, the number of negatrons due to the lower energy distribution becomes very much greater than those due to the higher energy dis-

assuming it to be so for Ce^{144} and Pr^{144} , certain rather reasonable results are obtained.

It is found that the sum of the areas bounded by the two lowest energy spectra is 5% less than the sum of the areas bounded by the three highest energy spectra. Any other grouping involves a much larger difference. If, for instance, the spectrum with the 0.517 end point is grouped with the two lowest energy spectra, the difference becomes 12%. The relative half-lives of Ce^{144} and Pr^{144} make it certain that the spectrum with the 3.06 Mev. end point is due to the disintegration of Pr^{144} . On this basis then, the 0.303 Mev and 0.179 Mev beta-disintegrations are attributed to Ce^{144} , and the others to Pr^{144} . It should be noted also that a beta-disintegration energy greater than 0.5 Mev is very high for a beta-emitter with a half life of 275 days.

The difference between the maximum beta-ray energies of the two lowest spectra is 124 Kev. Subtracting from this the K binding energy for Pr^{144} a figure of 82 Kev. is obtained. In view of the errors inherent in determining the end-points, this value is in remarkably good agreement with the measured value of the 90.1 Kev K conversion line exhibited by the main spectrum (Figure 6). In addition, agreement is secured with previous indications that conversion takes place in the Pr^{144} nucleus, since the two spectra in question have been assigned to Ce^{144} .

Further results of the integrations are shown in the tentative decay scheme (Figure 10) in which the fractional occurrences of the various groups of electrons are given. Estimation of the fractional occurrence of the 3.06 Mev. disintegration can be made fairly accurately since such a large portion of the spectrum lies in the high energy range where measurements are more easily made. For the lower energy spectra the errors are necessarily larger.

Because of the poor geometry, the gamma-ray energies as determined by the absorption experiment are believed to be substantially below the actual energies. Determination of accurate values by applying corrections is a very difficult task. However one obvious source of error can be taken into account. Since the absorber in the experiment was very close to the source, the effective path length in lead is a good deal greater than the nominal path difference. Errors are likely to be larger for the higher energy gamma-rays since at the higher energies slight changes in the determined absorption coefficient result in relatively large changes in the deduced energy. If the effective path length is 20% greater than the nominal path length, (a reasonable figure) then the calculated energy of the hard component is 1.6 - 1.7 Mev. The actual energy of this component is believed to be between 1.5 and 2.5 Mev. On a similar basis, the two softer components are 0.35 - 0.55 Mev., and 0.20 - 0.25 Mev., where the limits assigned are much narrower because of the relative insensitivity of the deduced energy with errors in the measurement of the absorption coefficient. The existence of the very soft 132 Kev component is not evident from Figure 9, due probably to the low resolution of the method and inefficiency of the gamma counter.

Using the calibration curve (Figure 4 B) for the copper cathode counter, the relative intensities are found to be 1:1:8, assuming energies of 2, 0.5 and 0.22 Mev. respectively. The tentative decay scheme would require that the 2 Mev. radiation be twice as intense as the 0.5 Mev. radiation. Bearing in mind the uncertainties connected with the absorption experiment and the variation of counter efficiency with energy, it is concluded that no real inconsistency is implied. The 0.22 Mev. radiation, from the decay scheme, should be four times as intense as the sum of the

intensities of the other two components. The excellent agreement is to be regarded as fortuitous.

The large errors associated with the coincidence counting rates make interpretation of the results of the coincidence experiments rather difficult. It is certain however, that the conversion electrons do not arise from internal conversion of a gamma-ray following the 3.06 Mev. beta-disintegration. Further, comparison of the total and random rates (0.0400 vs. 0.0393) for the first beta-gamma coincidence experiment makes it clear that the 3.06 Mev. beta-disintegration leads directly to the ground level of the Nd^{144} nucleus. There are definite indications that the Pr^{144} and Ce^{144} spectra are both complex, but the errors are such that this cannot be stated with certainty. The existence of separate distributions for Ce^{144} and Pr^{144} is clearly shown. The drop in the ratio C_c/C_β when 1/8" of lead is interposed between the source and gamma-counter at 0.070 Mev. indicates that the gamma radiation associated with the Ce^{144} negatrons is soft. For this reason, along with the fact that the 0.22 Mev gamma-radiation is the most abundant, it is believed that the 0.22 Mev. quanta are emitted following the 0.303 Mev. beta-disintegration, as shown in Figure 10. Assuming that the decay scheme is correct, estimates of the conversion coefficients α_K and α_L may be made. The number of quanta emitted is equal to the number of 0.179 Mev. disintegration negatrons. This number can be obtained by integration of the area under the corresponding spectrum in Figure 8. If N is the net counting rate, and $\Delta(Hf)$ the line width, then the required number of negatrons

is

$$\int_{Hf=0}^{Hf(\max)} \frac{N}{\Delta(Hf)} d(Hf)$$

where

$$\frac{\Delta(Hf)}{Hf} = 0.034.$$

The area has been measured with a planimeter, and the relation $H \rho = 4885 V$ applied.

This leads to a value of 2100 negatrons/min for the integral. A 10% error is estimated.

Neglecting spherical aberration, the number of K electrons emitted into the same solid angle is given by the height of the K line above the continuum, i.e., 136 counts/min. A rough correction for spherical aberration can be made by increasing this value by 25-30% (Figures 11 B and 11 C reference (8)):

α_L is found by determining the ratio of the height of the L line to the height of the K line. The values obtained are

$$\alpha_K = 0.082 \pm 0.025 \qquad \frac{\alpha_L}{\alpha_K} = 0.216 \pm 0.054$$

The estimates of errors are based upon uncertainties in measuring the area, probable errors associated with the determination of the heights of the lines, and uncertainties as to the height of the continuum below the peaks.

Incidentally, the value of 2100 negatrons/min. emitted into the solid angle of 0.067×10^{-2} of 4π steradians yields a value of 4 microcuries for the Ce^{144} beta-activity, taking the fractional occurrence of the 0.179 beta-disintegration into account.

It may be of value at this point to determine the order of the various spectra by computing the ft values, and thence determining the grouping to which each belongs (Table II, reference (26)). The partial half-lives (t) have been determined from the relative intensities of the spectra based upon measurements of the relative areas bounded by the spectra (Figure 8), using the relation

VII. CONCLUSIONS

The conclusions of this investigation are given in most succinct form in the disintegration schemes of Figure 10. Lack of precise knowledge of the gamma-ray energies and insufficient coincidence data make it desirable however, to regard the schemes as tentative. The schemes are nevertheless consistent with the information obtained from analysis of the complete beta-spectrum, from the gamma-ray absorption experiments and from the coincidence experiments. They are also in accord with the known half-lives of Ce^{144} and Pr^{144} .

The results are not in complete agreement with those of the Plutonium Project^(A-1). The end-point of the high energy Pr^{144} spectrum, 3.06 ± 0.09 Mev., agrees very well with that reported by the Plutonium Project viz. 3.07 Mev. The conversion lines are found to occur at energies of 90.1 Kev and 125 Kev, compared with the Plutonium Project measurements of 91 Kev and 128 Kev. respectively. In addition the Plutonium Project attributes these lines to Pr^{144} whereas this thesis attributes them to Ce^{144} , i.e., to internal conversion of gamma-radiation in the Pr^{144} nucleus. The 0.22 Mev gamma-radiation assigned to Ce^{144} is in disagreement with the report of the Plutonium Project which assigns this radiation to Pr^{144} . The end-point of the Ce^{144} beta-spectrum is found to be 0.303 ± 0.020 Mev., about 13% lower than the Plutonium Project value of 0.348 Mev. Finally, the Plutonium Project reports no gamma radiation from Ce^{144} in conflict with the findings of this thesis.

VIII. REFERENCES

- (A1). Plutonium Project, Jour. Am. Chem. Soc. 68, 2411 (1946).
- (A2). H.J. Born and W. Seelman - Eggebert, Naturwissenschaften 31, 201 (1944).
- (A3). O. Hahn and F. Strassman, Naturwissenschaften, 31, 499 (1943).
- (A4). O. Hahn and F. Strassman, Naturwissenschaften, 27, 11 (1939).
- (A5). E.H. Segre, "Segre Chart", Addison-Wesley Press Inc. Cambridge, Mass
- (1). O. von Baeyer and O. Hahn, Phys. Zeit, 11, 488 (1910).
- (2). J. Danysz, Le Radium 9, 1 (1912).
- (3). E. Rutherford and H. Robinson, Phil. Mag. 26, 717 (1913).
- (4). R.A.R. Tricker, Proc. Camb. Phil. Soc. 22, 454 (1924).
- (5). O. Klemperer, Phil. Mag. 20, 545 (1935).
- (6). W.T. Davis and C. O'Ceallaigh, Proc. Camb. Phil. Soc. 33, 540 (1937).
- (7). V.E. Cosslett, Jour. Sci. Insts. 17, 299 (1940).
- (8). M. Deutsch et al., Rev. Sci. Inst. 15, 178 (1944).
- (9). M. Korsunskii et al., Jour. Exptl. Theoret. Phys. (U.S.S.R.) 14, 394 (1947).
- (10). C.P. Ellis and W.A. Wooster, Proc. Roy. Soc. A117, 109 (1927).
- (11). L. Meitner and W. Orthmann, Zeits. f. Phys. 60, 143 (1930).
- (12). H.R. Crane, Rev. Mod. Phys. 20, 278 (1948).
- (13). C.T. Zahn and A.W. Spees, Phys. Rev. 53, 65 (1938).
- (14). H.A. Bethe and R.F. Bacher, Rev. Mod. Phys. 8, 184 (1936).
- (15). H.A. Bethe, "Elementary Nuclear Theory" Wiley and Sons, New York (1947).
- (16). C.D. Ellis, International Conference on Physics, London (1934).
- (17). R.O. Haxby et al., Phys. Rev. 58, 1035 (1940).
- (18). E.J. Konopinski, Phys. Rev. 72, 518 (1947).
- (19). M.E. Nahmias, Proc. Camb. Phil. Soc. 31, 99 (1935).
- (20). J.S. Allen. Phys. Rev. 61, 692 (1942).
- (21). B.T. Wright, Phys. Rev. 71, 839 (1947).
- (22). A.I. Leipunski, Proc. Camb. Phil. Soc. 32, 301, (1936).

- (23). J.C. Jacobsen and O. Kofoed - Hansen, Det. Kgl. Danske Vidensk. Selskab. Mat.-Fys. Med. 23, No. 20 (1945).
- (24). H.R. Crane and J. Halpern, Phys. Rev. 53, 789 (1938); 56, 232 (1939)
- (25). E. Fermi, Zeits. f. Physik 88, 161 (1934).
- (26). E.J. Konopinski, Rev. Mod. Phys. 15, 209, (1943).
- (27). E.J. Konopinski, and G.E. Uhlenbeck, Phys. Rev. 48, 7 (1935).
- (28). B.W. Sargent, Proc. Roy. Soc. 139, 659 (1933).
- (29). S.A. Korff, "Electron and Nuclear Counters" D. Van Nostrand, New York (1946).
- (30). J. Dunworth, Rev. Sci. Inst. 11, 167 (1940).
- (31). W.C. Peacock, Phys. Rev. 72, 1049 (1947).
- (32). C.S. Cook and L.M. Langer^{Phys. Rev.}, 73, 601 (1948).
- (33). A.H. Compton and S.K. Allison "X-Rays in Theory and Experiment", D. Van Nostrand, New York, (1935).
- (34). F.N.D. Kurie et al., Phys. Rev. 49, 368 (1936).
- (35). E.T. Copson "Introduction to the Theory of Functions of a Complex Variable" p. 224, Oxford Univ. Press (1935).
- (36). R.D. Evans, "The Science and Engineering of Nuclear Power" Addison-Wesley Press Inc., Cambridge, Mass. (1947).
- (37). G.D. Latyshev, Rev. Mod. Phys. 19, 132 (1947).
- (38). A.G. Mitchell, Rev. Mod. Phys. 20, 296 (1948).
- (39). J.R. Downing et al., Phys. Rev. 60, 470 (1941).
- (40). A. Roberts et al., Phys. Rev. 60, 544 (1941).
- (41). J.R. Downing, et al., Phys. Rev. 61, 686 (1941).
- (42). M. Deutsch et al., Phys. Rev. 62, 3 (1942).
- (43). M. Deutsch and L.G. Elliott, Phys. Rev. 62, 558 (1942).
- (44). M. Deutsch and L.G. Elliott, Phys. Rev. 62, 558A (1942).
- (45). L.G. Elliott et al., Phys. Rev. 63, 386 (1943).

- (46). A. Roberts et al., Phys. Rev. 64, 268 (1943).
- (47). L.G. Elliott and M. Deutsch, Phys. Rev. 64, 321 (1943).
- (48). L.G. Elliott and R.E. Bell, Phys. Rev. 72, 979 (1947).
- (49). M.I.T. Radar School "Principles of Radar" McGraw-Hill Book Co.,
New York (1946).
- (50). G.F. von Droste, Zeits. f. Phys. 100, 529 (1936).
- (51). K. Siegbahn, Ark. Mat. Astr. Fys. 30A, (Paper 20) (1944).
- (52). R.D. Evans and R.O. Evans, Rev. Mod. Phys. 20, 305 (1948).

CROSS SECTION OF SPECTROMETER

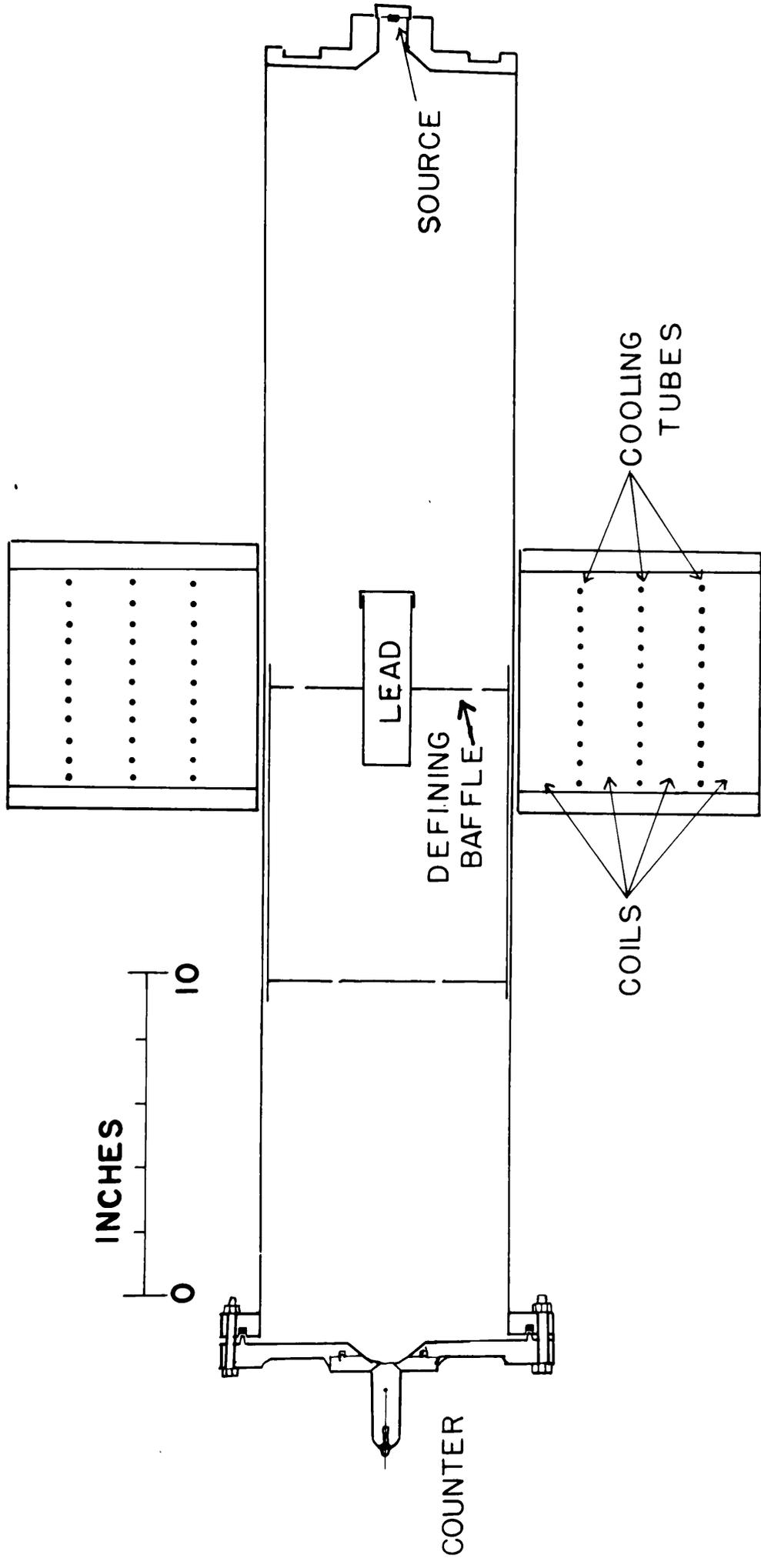
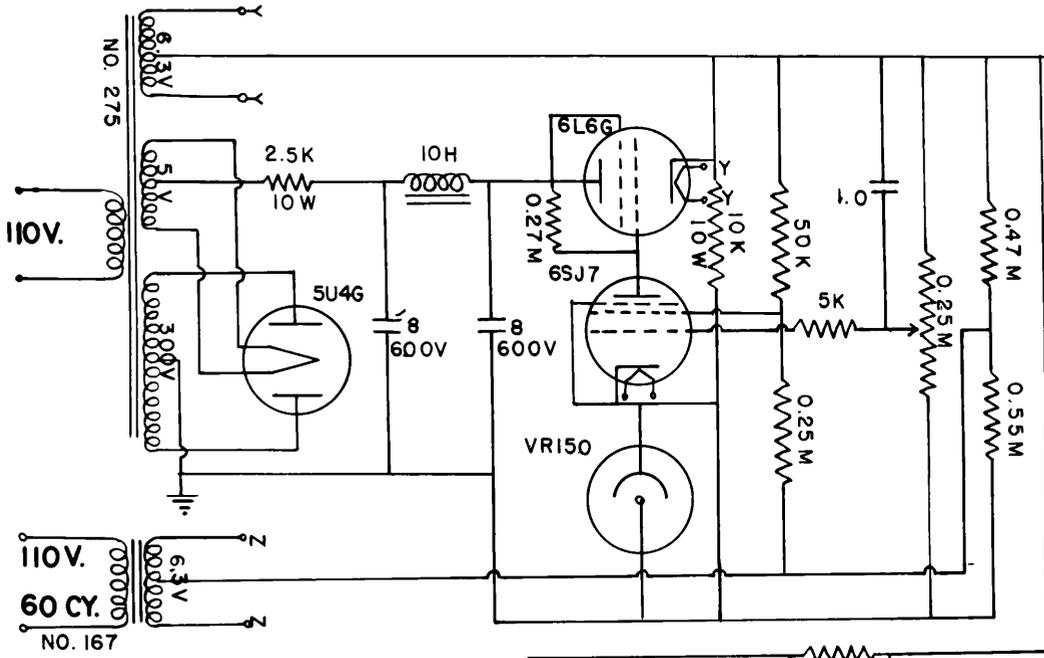
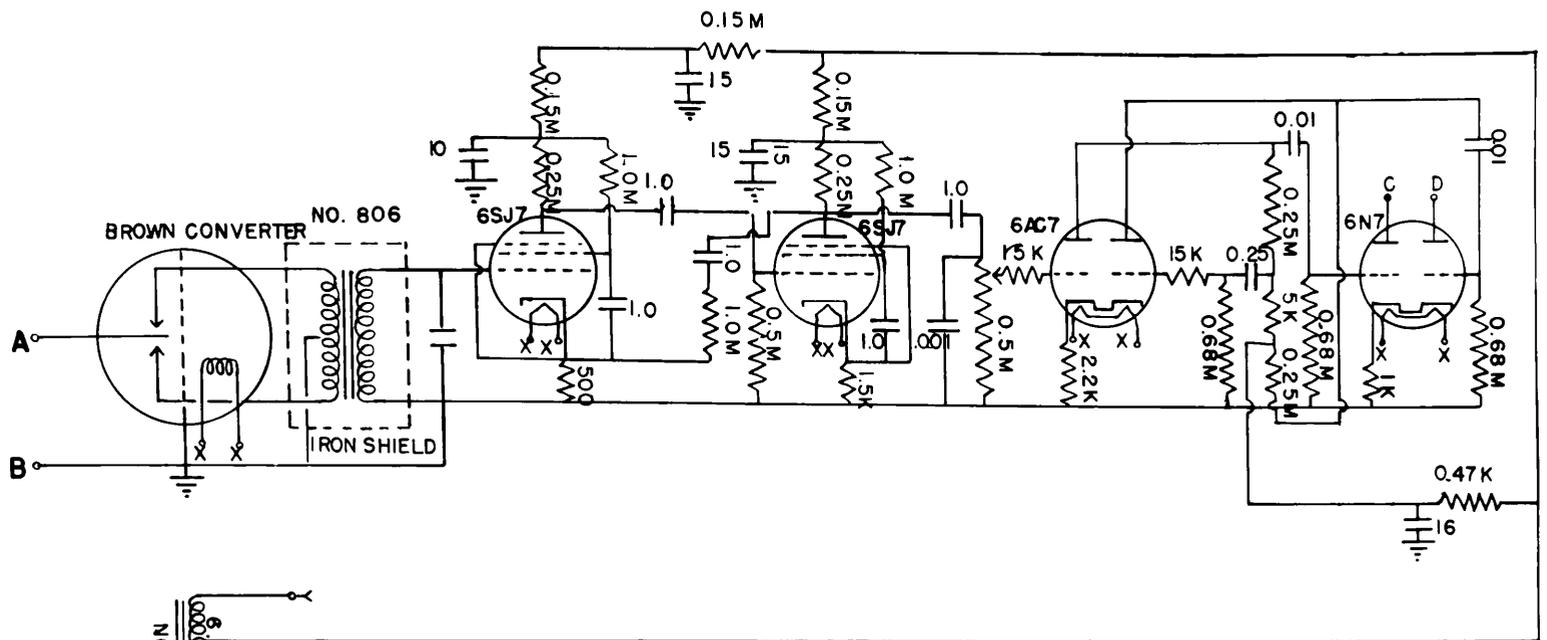


FIGURE I.



POINTS A AND B RECEIVE
ERROR VOLTAGE FROM
STANDARD RESISTANCE -
POTENTIOMETER CIRCUIT.

M = MEGOHMS
K = KILOHMS
CAPACITANCE IN MICRO-
FARADS

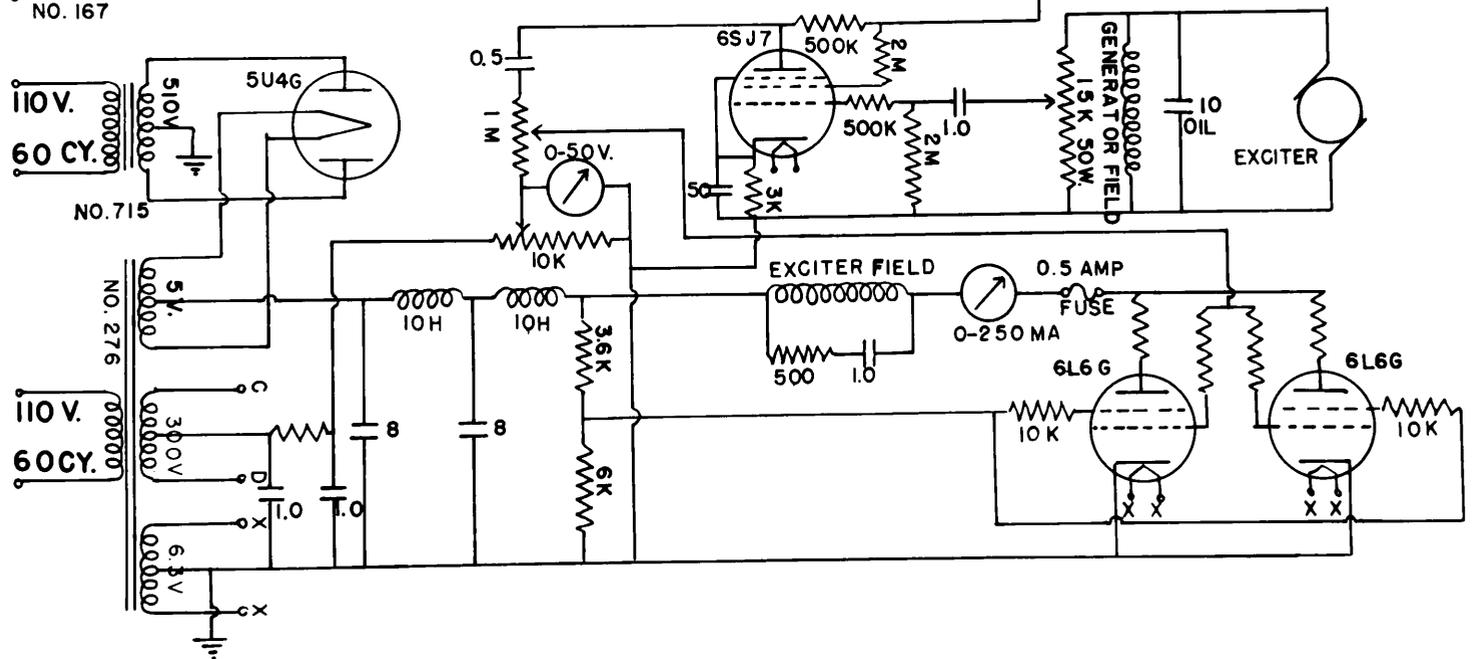


FIGURE 2 MAGNET CURRENT
REGULATOR

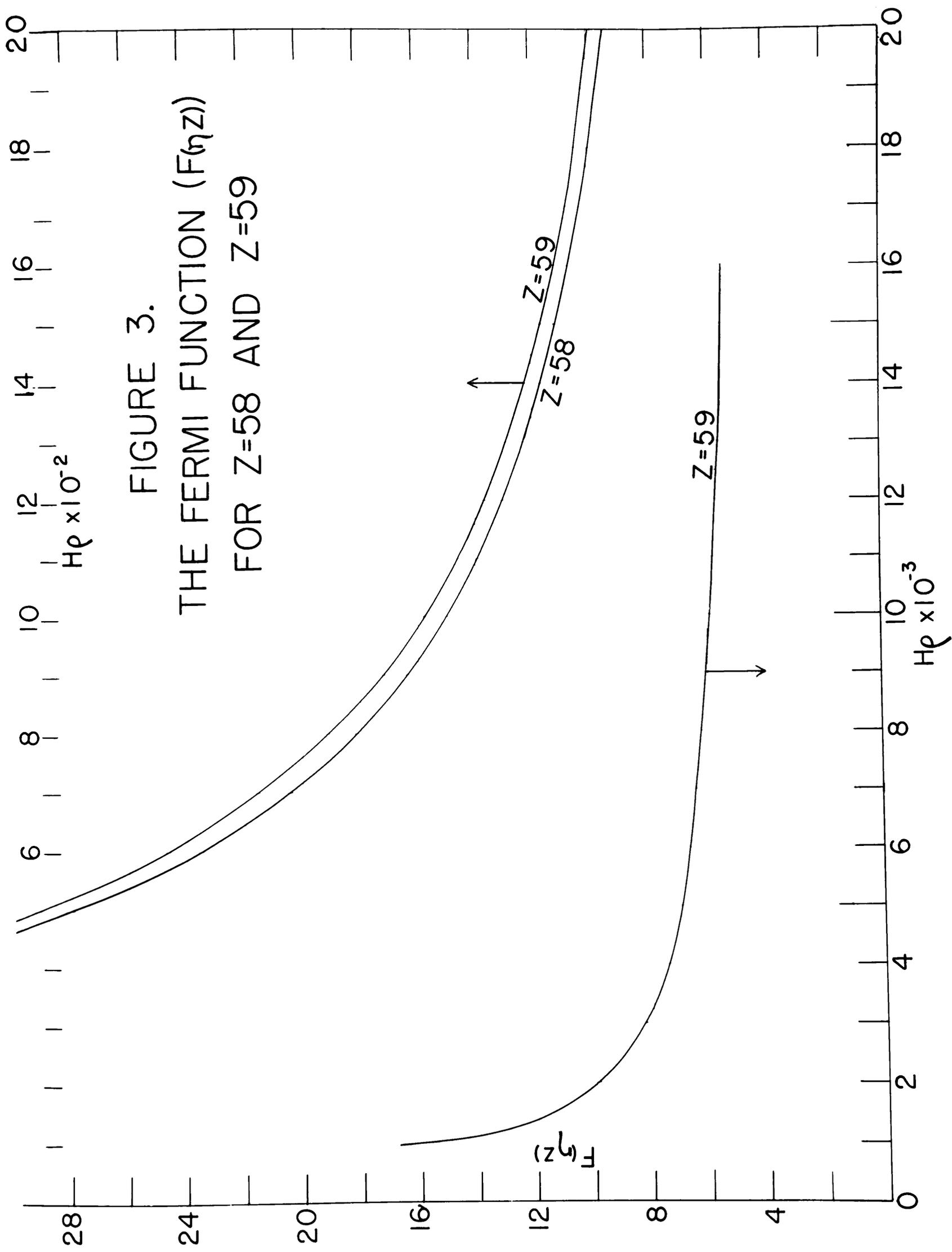
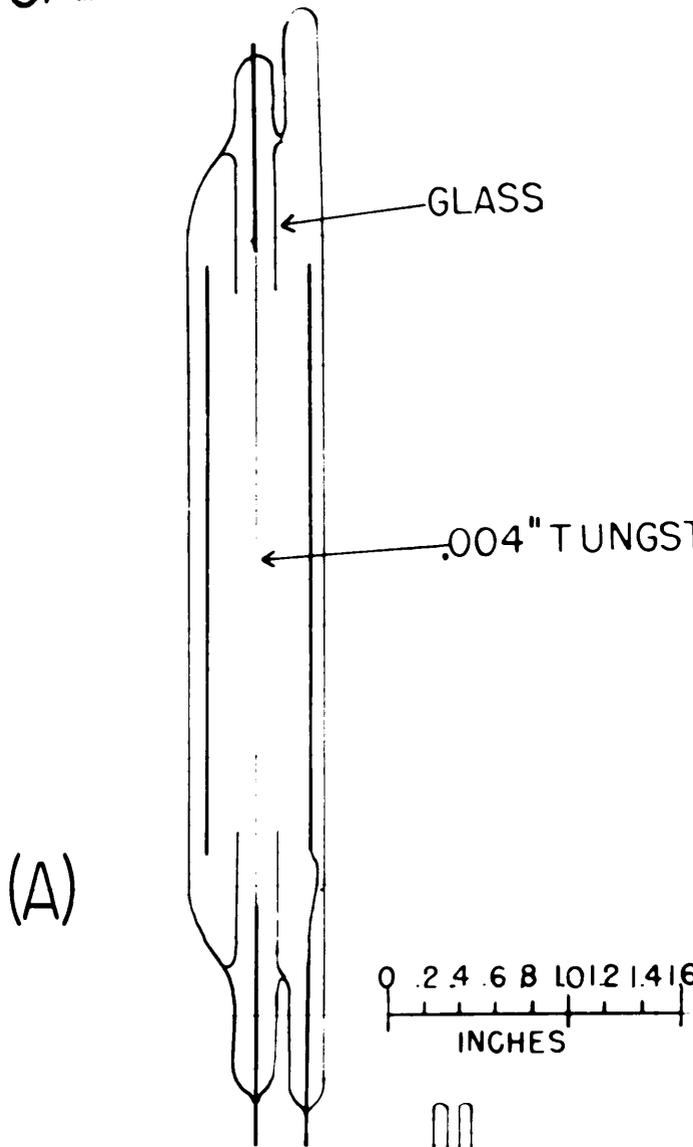


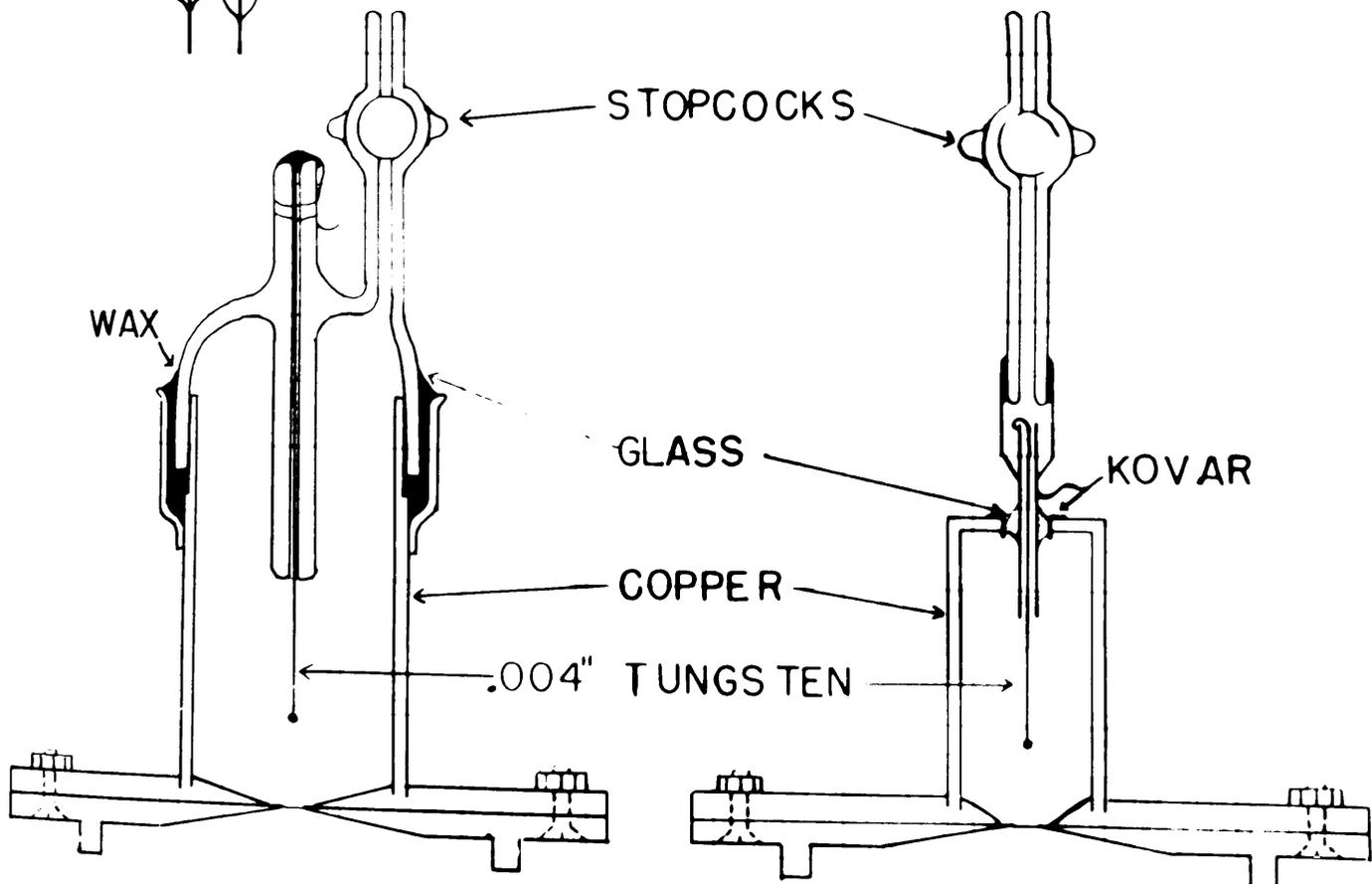
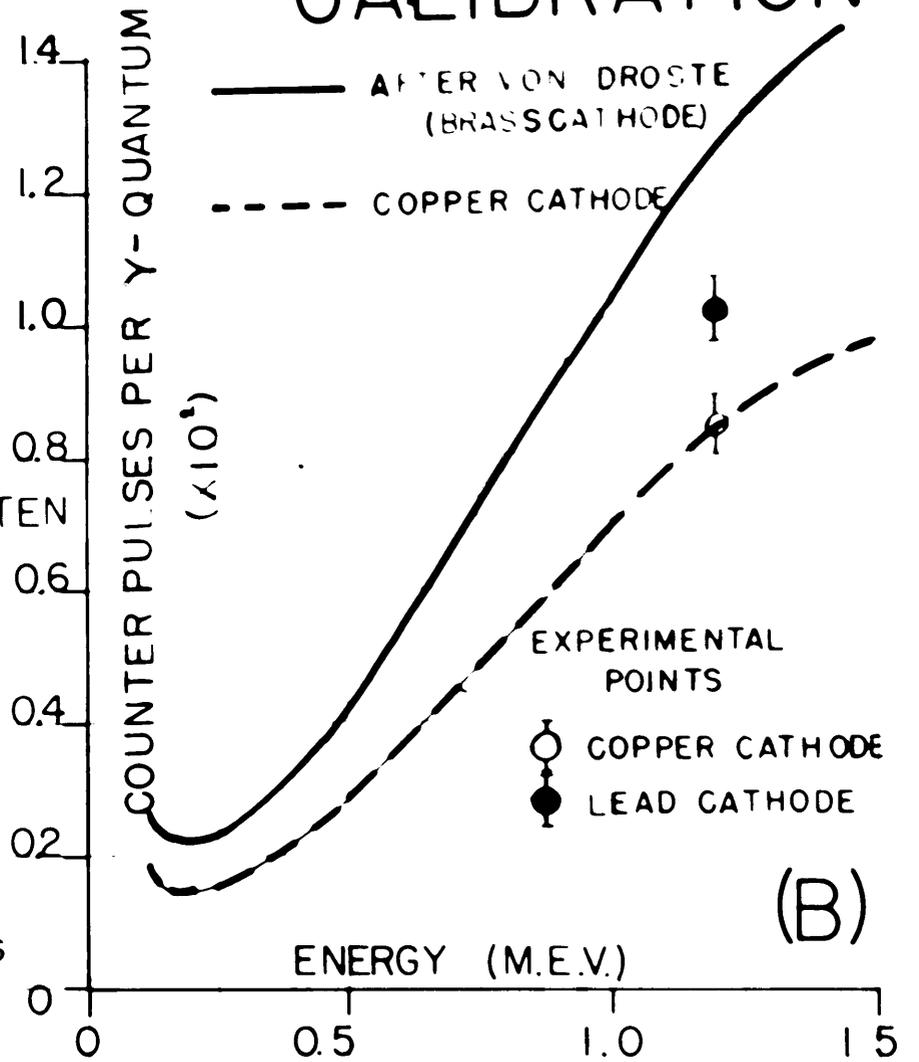
FIGURE 3.
 THE FERMI FUNCTION ($F(\eta z)$)
 FOR $Z=58$ AND $Z=59$

FIGURE 4

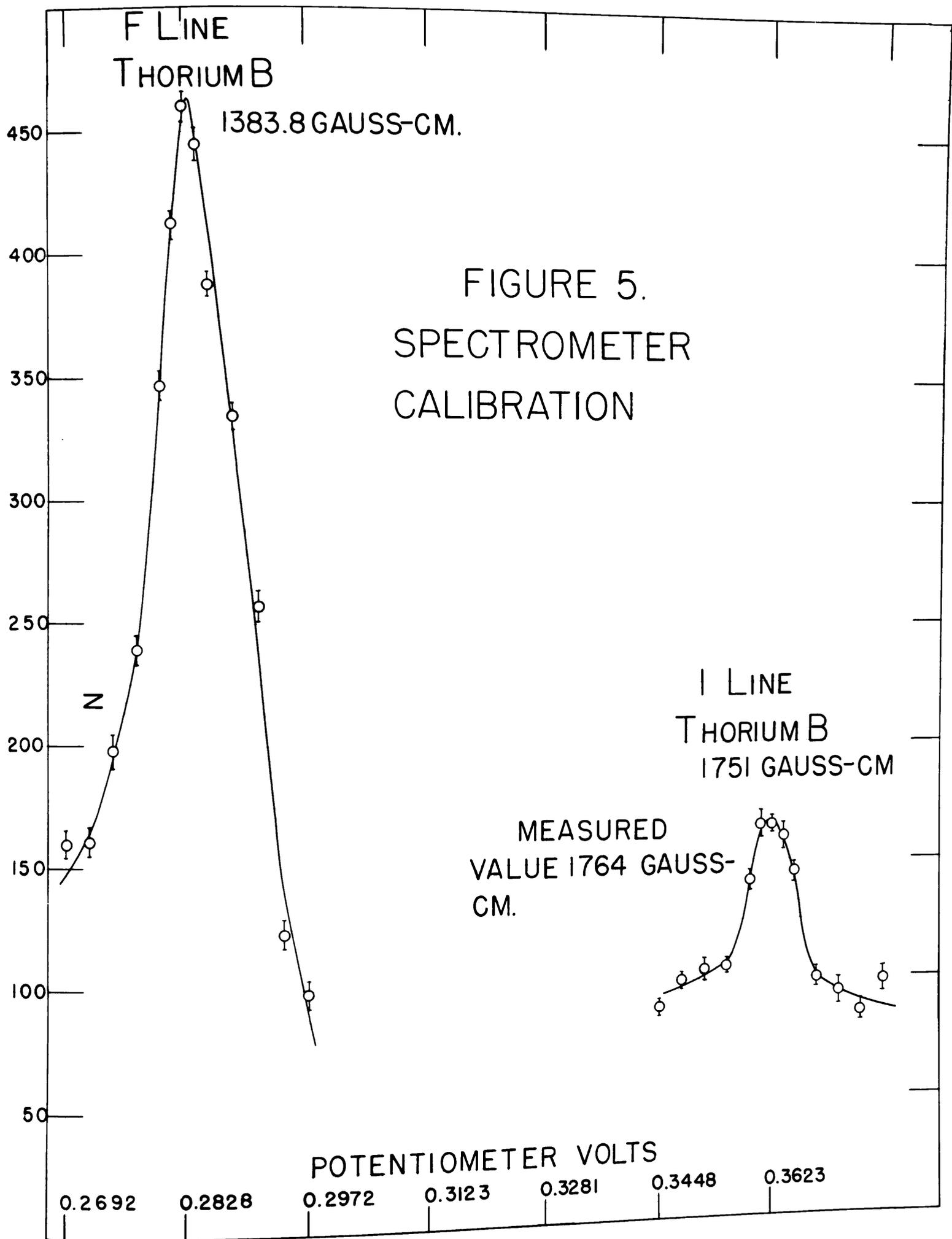
GAMMA COUNTERS



GAMMA COUNTER CALIBRATION



TYPES OF BETA COUNTERS



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