

THE BETA AND GAMMA RADIATIONS
OF THULIUM 170

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by

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Summary

The beta and gamma radiations of 127 day Tm^{170} have been investigated using a thin lens spectrometer and coincidence techniques. The beta ray spectrum has been shown to consist of two groups with end points at (970 ± 5) kev. and (886 ± 5) kev. A single weak gamma ray of (83.9 ± 0.2) kev. has been found. This energy has been computed from the K, L and M conversion lines on the beta spectrum at 22.6, 73.5, and 81.2 kev. respectively as well as from L and M photoelectron lines using a lead radiator. A search for other gamma rays using lead and uranium radiators had a negative result. Beta-gamma coincidences were obtained for approximately 1% of the beta rays, confirming the complexity of the spectrum. The total conversion coefficient of the gamma ray is estimated at 0.8. A decay scheme consistent with these results is given. The decay of the activity has been followed for $3\frac{1}{2}$ months and the half-life found to be 126 ± 5 days.

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I. Introduction

(a) Previous Studies on Thulium.

Thulium is a rare earth element which forms a stable oxide, Tm_2O_3 . It has a nuclear charge number of 69 and has been shown by Aston⁽¹⁾ to have a single stable isotope of mass 169. Schuler and Schmidt⁽²⁾ state that the nuclear spin of $^{69}\text{Tm}^{169}$, as deduced from observations of the hyperfine spectral structure, is $\frac{1}{2}\hbar$.

In 1936, Neuninger and Rona⁽³⁾ reported that the slow neutron bombardment of Tm_2O_3 resulted in a negative beta-ray activity with a half-life of $4 \pm \frac{1}{2}$ months. A series of measurements by Bothe⁽⁴⁾ extending over about one year gave the more accurate value of 127 ± 5 days. Seren, Friedlander, and Turkel⁽⁵⁾ give a value of 105 days with an estimated error of 20%. Bothe's figure of 127 ± 5 days is undoubtedly the most reliable at the present time. Knox⁽⁶⁾ reports a half-life of 125 days.

The thermal neutron capture cross-section of Tm has been measured by Riezler⁽⁷⁾ using a D on D neutron source, the neutrons being slowed down by paraffin. His result was $69 \pm 25 \times 10^{-24} \text{ cm}^2$. A similar experiment by Bomke and Reddemann⁽⁸⁾ yielded a cross-section of $114 \times 10^{-24} \text{ cm}^2$. Bothe's⁽⁴⁾ result was $95 \times 10^{-24} \text{ cm}^2$. Using the thermal column of a chain reacting pile, as a neutron source, Seren et al⁽⁵⁾ have made rough measurements of the capture cross-section and half-life of a large number of isotopes. They obtained a value of $106 \times 10^{-24} \text{ cm}^2$ for the cross-section of Tm and estimated their error at 20%.

By absorption of the beta-rays in aluminium Bothe⁽⁴⁾ found the maximum energy to be about 1 Mev. and found no gamma radiation.

Dr. L.G. Stephens-Newsham⁽⁹⁾ while at this laboratory examined the beta spectrum of Tm with the thin lens magnetic spectrometer used in the present work. He found a conversion line at 68.2 kev. indicating the presence of a low energy gamma ray and presented evidence for the complexity of the beta spectrum. The end points of the three components were stated to be 953 ± 20 , 526 ± 20 , 304 kev. respectively. Absorption of the gamma rays in lead gave a curve which could be resolved into three components with energies of 100, 200, and 800 kev. respectively. These results could not be fitted into a disintegration scheme and further study using sources of higher specific activity was considered justified by the author. Since the start of this work other conflicting results on Tm^{170} have been published in the literature. Seaborg and Perlman in the recent Table of Isotopes⁽¹⁰⁾ list, for Tm^{170} , a single beta ray with a maximum energy of 980 kev. as reported by Ketelle and Boyd⁽¹¹⁾ and a single soft gamma ray reported by Ketelle⁽¹²⁾. Subsequently Ketelle⁽¹³⁾ stated the gamma ray energy to be 84 kev. as determined from the L conversion line and from photoelectrons ejected from various radiators. In addition, as a result of some beta-gamma coincidence measurements, he found that the gamma rays are in coincidence with only a small fraction of the beta particles, not greater than 10%. This suggests the existence of two beta groups whose end points differ by the energy of the gamma ray and that the lower energy group is of a lower intensity than the higher energy group. Consistent with the low beta-gamma coincidence rate is the inference of a large total conversion coefficient for the gamma ray. Saxon and Richards⁽¹⁴⁾, using a sample of Tm_2O_3 purified by the ion exchange technique at the Oak Ridge National Laboratory⁽¹³⁾, also found a single beta ray group of maximum energy 970 ± 10 kev. and a single gamma ray of 85.5 kev. The gamma energy was deduced from the K, L, and

M conversion lines observed at the low energy end of the beta spectrum. The K, L, and M conversion coefficients were given as 3.06, 4.28, 1.68% respectively. The most recent publication, by Grant and Richmond⁽¹⁵⁾, is only partially in agreement with the foregoing results. Using spectrometer and absorption methods they have found beta ray groups with maximum energies 1.00 ± 0.01 , 0.90 ± 0.015 , 0.79 ± 0.03 , and 0.45 ± 0.05 mev. and three gamma rays with energies of 82.7 ± 0.7 , 200 ± 10 , and 440 ± 20 kev. In these results there is again an indication of two high energy beta-ray groups whose end points differ by that of the internally converted gamma ray.

Before proceeding with a discussion of further work on the radiations from Tm^{170} a brief discussion of the salient features of beta ray theory and spectroscopy will be given.

(b) Theory of Beta-Decay.

The beta-ray spectra of radioactive nuclei are characterized by (i) a continuous distribution in energy of the particles from zero up to a well defined maximum and (ii) in many cases a line spectrum superposed on the continuum. When the line spectra are present gamma radiation is found to be emitted. The lines are readily interpreted as mono-kinetic electrons ejected from the electron shells surrounding the nucleus. The energy of these internal conversion electrons will be, according to this view, equal to the gamma-ray energy less the binding energy of the electron in the K, L, M or other shell of the disintegrating atom. In this way a series of lines may be generated by the conversion of gamma rays of a single energy.

The continuous distribution in energy of beta-rays is in striking contrast to the discrete energies observed in the spectra of gamma-rays and α -particles. This was shown by Ellis and Wooster⁽¹⁶⁾ not to be caused by any secondary effect of scattering or absorption of beta-particles all initially of the same energy. They measured calorimetrically the total energy released by a known amount of the beta-emitter RaE, which has no associated gamma-ray. The result was that the average energy per disintegration coincided with the average energy of the beta particles in the spectrum and not with the maximum energy. Thus the continuity of the beta-spectrum is a property of the disintegrating nucleus. The beta particle does not share its energy with a second beta ray since it can be shown by the location of the disintegrating nucleus and its product in the periodic table that only one beta particle is emitted per disintegration.

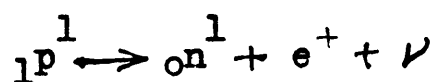
Another difficulty is that for a number of reasons (see reference 17) we must assume that the electrons observed in beta-decay

do not exist in the emitting nucleus.

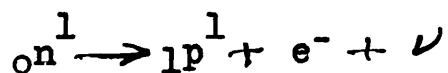
It is observed in beta-decay that the statistics of nuclei remain unchanged and the nuclear spin changes by integral multiples only of $\frac{1}{2}\hbar$. The beta-particle which is emitted has Fermi statistics and spin $\frac{1}{2}\hbar$.

These difficulties are resolved by the adoption of two hypotheses; (i) the emitted beta-particle is created at the moment of emission, and (ii) a new particle, the neutrino is also created and emitted at the same instant. The properties required of this particle are very small mass, spin $\frac{1}{2}\hbar$ and Fermi statistics. Apart from the successes of the Fermi theory of beta-decay the neutrino hypothesis has gained support through a series of measurements on the energies and angular correlation of recoil nuclei as reviewed by Sherwin⁽¹⁸⁾ and through recent measurements on the energy spectrum of meson decay particles by Leighton, Anderson and Seriff⁽¹⁹⁾.

In the Fermi theory (for a review see reference 20) the fundamental beta-process may be represented by a transition between neutrons and protons with the emission of electrons (positive or negative) and neutrinos. Thus



and



represent positron and negatron emission respectively. By constructing a Hamiltonian with operators representing these processes and by fitting it into a formalism analogous to the quantization of the electromagnetic field into photons, Fermi obtained for the probability per unit time PdW of the emission of a beta-particle into the energy range (W, W + dW),

$$\frac{G^2}{2^3} \left| \int \dots \right|^2 F(Z,W) pW (W_0 - W)^2 dW, \dots \quad (1)$$

where G is an empirical dimensionless coupling constant which measures the strength of the interaction between the nucleon and the electron-neutrino field and hence determines the scale on which beta-decay takes place. $\left| \int \dots \right|^2$ is a matrix element which measures the overlap of the initial and final nuclear wave functions and $F(Z,W)$ is a coulomb correction factor which allows for the effect on the energy distribution of the coulomb force between the beta-particle and the residual nucleus. W is the total energy in units of mc^2 of the electron including its rest energy, and p is its momentum in units of mc . The function $F(Z,W)$ is, exactly,

$$F(Z,W) = \frac{4(2pR)^{2S-2} e^{\pi \alpha ZW/p} |\Gamma(S + i \alpha ZW/p)|^2}{|\Gamma(2S + 1)|^2} \cdot \frac{1+S}{2} \dots \quad (2)$$

where R = nuclear radius

α = fine structure constant

$$S = (1 - \alpha^2 Z^2)^{\frac{1}{2}}$$

Γ = gamma function

An easily calculable relativistic approximation is given by Bethe and Bacher⁽¹⁷⁾ and discussed by Longmire and Brown⁽²¹⁾. It is

$$F(Z,W) = \frac{2\pi Z \alpha W/p}{1 - e^{-2\pi Z \alpha W/p}} \frac{(W^2(1 + 4Z^2 \alpha^2) - 1)^{(1-Z^2 \alpha^2)^{\frac{1}{2}} - 1}}{\dots} \quad (3)$$

$F(Z,W)$ for $Z = 70$ was calculated with the aid of this approximation for use in the present work. According to Longmire and Brown this approximation is accurate to about 1% for $Z = 84$ and to 0.25% for $Z = 29$ or less. Furthermore the error in this approximation varies only slowly with the energy W .

For the calculation of the matrix element it is possible to

set up five independent types of interaction potentials which behave, under Lorentz transformations, like a scalar, a polar vector, a tensor, a pseudovector, and a pseudoscalar respectively. Fermi originally adopted the polar vector form whose matrix element involved only an identity operator. Now the initial and final nuclear wave functions are orthogonal unless their total angular momentum and parity (symmetry) properties are the same. The identity operator will not alter these properties which means that with the polar vector form of interaction only those transitions will be allowed in which no angular momentum or parity changes take place. Thus in the usual notation the Fermi selection rules for allowed transitions are $\Delta I = 0$ (no) where the ΔI is the spin change and the (yes) or (no) indicates whether or not the parity changes. Gamow and Teller⁽²²⁾ using the pseudovector and tensor forms arrive at different rules for allowed transitions, viz., $\Delta I = 0, \pm 1$ (no) with $0 \rightarrow 0$ forbidden.

In the calculation of these selection rules (i.e., of the matrix elements) all terms except the first in the expansion of the light particle wave functions into powers of the position vector \vec{r} are neglected. If, now, in the consideration of those cases which are not allowed according to the selection rules given above, one uses the second terms in the particle wave functions, transition probabilities smaller than the allowed ones by a factor of approximately 100 are obtained. In this way the so-called first and second forbidden selection rules of the Fermi and G-T type are derived. These may be found tabulated in reference 20, on page 220 for the polar vector and tensor forms of interaction.

In addition to the selection rules, the calculation of the matrix elements yield certain correction factors. In the case of the

allowed spectra, that is to say those which are allowed according to either Fermi or G-T rules, these correction factors are independent of energy. There is then, no method of determining experimentally which of the various interaction potentials or linear combinations of them is the correct one for allowed spectra. Some of the correction factors for the forbidden spectra, however, show a sufficiently large energy dependence to make an identification possible. Pertinent to this subject is a recent group of measurements on Y^{91} by Langer and Price⁽²³⁾, C^{137} by Peacock and Mitchell⁽²⁴⁾, Y^{90} and Sr^{90} by Braden, Slack and Shull⁽²⁵⁾, Rb^{86} by Zaffrano, Kern and Mitchell⁽²⁶⁾ and K^{42} by Shull and Feenberg⁽²⁷⁾. In each of these cases the energy distribution for a single beta-ray group shows a marked deviation from that predicted by the Fermi theory for allowed transitions. At least in some of these cases, the shell structure of nuclei⁽²⁸⁾ predicts a spin change of ± 2 and a parity change. If the G-T selection rules govern the beta-process then the theory of forbidden spectra⁽²⁰⁾ predicts that the beta-spectrum differs from the allowed shape by a factor G which has the approximate form

$$G \cong (W_0 - W)^2 + W^2 - 1 \quad \dots\dots\dots(4)$$

It was found that if this correction factor were applied to these spectra, the shape could be made to coincide with the allowed shape. These results lend support to the G-T selection rules and to the shell structure theory of nuclei.

Allowed and forbidden transitions may be classified empirically through a comparison of half-lives after taking into account the nuclear charge Z and the energy release W_0 . The procedure for doing this is given by Konopinski⁽²⁰⁾. The mean life τ of the beta-decay is given by

$$\frac{1}{\tau} = \int_1^{W_0} P dW$$

$$= (G^2 |\int \dots|^2 / 2\pi^3) f(Z, W_0) \dots \dots \dots (5)$$

$$\text{where } f(Z, W_0) = \int_1^{W_0} dW W(W^2 - 1)^{\frac{1}{2}} (W_0 - W)^2 F(Z, W) \dots \dots \dots (6)$$

The quantity ft , where t is the half-life, should be independent of energy release for allowed transitions and distinctly higher for forbidden transitions. A classification of the then known beta-emitters was given by Konopinski⁽²⁰⁾ in 1943. For the purposes of the present work an approximation which is valid for $W_0 \gg 1 + (\alpha Z)^2 / 2$ was used:

$$f(Z, W_0) = u(Z)(\bar{p})^{2S-2} [v(W_0) - w(Z)(W_0 - 1)^3] \dots \dots \dots (7)$$

where \bar{p}_{mc} is some average of the electron momentum. The functions $u(Z)$, $v(W_0)$, and $w(Z)$ may be found from curves given by Konopinski⁽²⁰⁾.

It must be said that there are exceptions to this classification. In the case of Y^{91} (23), for instance, the "comparative lifetime" ft is given as 4.7×10^8 according to which it would be classified as twice-forbidden. The successful application of the G-T rules and the theory of the shell structure of nuclei indicate that this transition should be classified as once forbidden. The ft value should be regarded as only a rough guide to the forbidden nature of a transition.

II. Experimental Methods

(a) Counting Procedures.

A beta-spectrum is obtained in a spectrometer by measuring the counting rate as a function of the momentum of the electrons focussed on the counter. At each setting of the current the counting rate must be corrected for a background. The background may be due to cosmic rays, photo-sensitivity of the counter, contamination of the counter materials or of the laboratory and to scattering of primary and secondary electrons into the counter. The background may be a function of the focussing current. In the lens spectrometer used by the author the background counting rate with the spectrometer focussed for energies higher than the end point of the beta spectrum was less than that with zero current. To obtain a reasonable first order background correction in the region of the spectrum a linear decrease in background with increasing focussing current was assumed.

The measured counting rate is subject to an error due to statistical fluctuations. The probable error may be computed with the aid the laws of probability on the assumption that the emission of an electron by a radioactive nucleus is a random event. If the total number of counts taken is n , the standard deviation is \sqrt{n} , and the probable error is $0.67 \sqrt{n}$. The percentage probable error is thus $\frac{0.67}{\sqrt{n}} \times 100$. Since the statistical errors in the background determination are added to those of the true counts, the probable percentage error in the presence of a background is $67 \frac{\sqrt{n + n_1}}{n - n_1}$ where n is the total number of counts and n_1 is the number of background counts in the same interval of time. Clearly it is desirable to obtain a high counting rate relative to the background.

Errors due to drifting of the current regulator or slow changes in the counter characteristic may be decreased by sweeping over

a given region of the spectrum with shorter counting runs at each point until the total number of counts taken at a point is adequately large. This procedure was followed here when the counting rate was low.

(b) Sources.

In the thin lens spectrometer the sources have the shape of a disc. For the measurement of continuous and line spectra the radioactive material is commonly deposited on a very thin backing by evaporating to dryness a solution of a salt of the element or by precipitating the salt of the element out of solution. Other methods involve electro-plating of the source material, evaporation of the material in vacuo, or the collection of the active deposit of a radioactive gas on a metallic foil. According to Deutsch, Elliott and Evans⁽²⁹⁾ the precipitation method is to be preferred, when applicable, since a more uniform source results. The source should be as thin as is possible consistent with an adequate counting rate. A high specific activity therefore is desirable. Scattering of electrons within the source may distort seriously the shape of the spectrum. The source is considered thin when a further reduction in thickness does not change the shape of the spectrum. Albert and Wu⁽³⁰⁾ have found that with S^{35} sources on collodion films of surface density 3 micrograms/cm² the source had to be reduced below 5 micrograms/cm² before distortions were removed. This interpretation of their results, however, is not unambiguous. It is possible that with their source mounted on a very thin film of collodion, a non-conductor, that source charging⁽³¹⁾ distorted their beta-spectrum. Nevertheless one may say with assurance that the source for the beta-spectrum should be uniformly thin, electrically grounded to the spectrometer chamber, backed by material of low atomic number and low surface density, and mounted as far as possible from the walls of the spectrometer.

Gamma ray energies may be determined spectroscopically by measurements on the secondary electrons they produce. Use is made of three types of interaction of gamma rays with matter, viz., (i) the photoelectric effect, (ii) the Compton effect, and (iii) pair production. These three effects will be discussed later in connection with the analysis of the secondary electron spectra. A strong source of 10 to 50 millicuries (or more for less intense gamma rays) is mounted in a capsule of low Z (e.g., Cu) sufficiently thick to stop all of the primary beta-rays. In front of this is placed a thin radiator of high Z such as lead or uranium. The Compton electron spectrum is obtained by replacing the thin radiator of high Z by a thick radiator of low Z (e.g. Al). Since the photoelectric cross-section is approximately proportional to Z^5 (32) a heavy element is chosen to enhance the production of photo-electrons, especially for weak gamma rays. The radiator should not be so thick as to slow the electrons down into the Compton region.

(c) Analysis of the Spectra.

Beta-ray spectra are very often complex, and consist of two or more competing disintegrations with different end points. One of the aims of beta-ray spectroscopy is the resolution of these components and through this the determination of the disintegration schemes of radioactive nuclei. A method of ascertaining the complexity of a spectrum and of determining the end points of its components has been devised by Kurie, Richardson and Paxton⁽³³⁾. The assumption is made that expression⁽¹⁾ above for the Fermi distribution function is correct for a simple spectrum. In rationalized relativistic units

$$w^2 = p^2 + 1$$

$$\therefore wdw = pdp.$$

We may then write, where N' is the number of electrons per unit momentum interval,

$$N' \propto F(Z, W) p^2 (W_0 - W)^2.$$

This may be rearranged to read

$$\sqrt{\frac{N'}{p^2 F(Z, W)}} \propto (W_0 - W) \dots\dots\dots(8)$$

Therefore a plot of $\sqrt{\frac{N'}{p^2 F}}$ vs W should yield a straight line whose intercept with the energy axis will be the end point energy W_0 . Since the momentum interval of the spectrometer is a constant fraction of the momentum

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

where N is the net counting rate. The momentum of the focussed electrons is, in a thin lens spectrometer, proportional to the coil current, which is in turn proportional to the voltage developed across a standard resistance in series with the coil. Thus for the particular case of the lens spectrometer used here⁽⁸⁾ may be written

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

which is, in terms of kinetic energy E .

$$\sqrt{\frac{N}{V^3 F(Z, W)}} = K(E_0 - E) \dots\dots\dots(9)$$

where K is a constant.

The application of (9) to a simple spectrum constitutes a test of the Fermi theory as well as being a method for obtaining E_0 . A straight line is in fact obtained for allowed spectra and in many cases for forbidden spectra. In the several cases cited above in references (23) to (27) energy dependent nuclear matrix elements have been found

which fit the experimental data. The test is made by incorporating the forbidden correction factor G into (9) as follows:

$$\sqrt{\frac{N}{v^3 G(W) F(Z,W)}} = K(E_0 - E) \dots\dots\dots(10)$$

In these cases a plot of (9) yielded an S-shaped curve while (10) gave a straight line.

This method is known as the Kurie or Fermi plot. The distinction is sometimes made between those cases where a relativistic Coulomb correction factor $F(Z,W)$ is used (Fermi plot) and those where a non-relativistic approximation is used for $F(Z,W)$ (Kurie plot)⁽²¹⁾.

In some cases, complexity of a spectrum is indicated on a Kurie plot by a marked upward deviation from the straight line at energies below the end point. The ordinate is then $\sqrt{\frac{N_1 + N_2 + N_3 + \dots}{v^3 F(Z,W)}}$

and resolution into the components

$$\sqrt{\frac{N_1}{v^3 F}} , \sqrt{\frac{N_2}{v^3 F}} , \dots\dots$$

may be effected by a graphical method. The straight line portion at higher energies is extrapolated to the lower energy region and the squares of points on it are subtracted from the squares of the corresponding points on the total curve. The square roots of the differences are then plotted and this process is repeated until the last replotting yields a reasonably straight line. It must be borne in mind that the assumption is made that the distribution functions of the component spectra are identical with that of allowed transitions, that the errors are multiplied in successive replottings, and that deviations from a straight line plot may be instrumental, especially at low energies. The complexity of the spectrum should be corroborated by independent gamma-ray measurements and coincidence experiments where possible.

Internal conversion coefficients may be estimated from the beta-spectrum. By definition the conversion coefficient is the fraction of gamma rays which produce internal conversion electrons. The latter are mono-kinetic so their number will be proportional to the height of the conversion line above the continuum if one neglects those which lose sufficient energy in collisions to be thrown out of the pass band of the spectrometer. The total number of gamma rays may be obtained by estimating the number of beta particles emitted in cascade with the gamma rays. If $N(p)$ is the recorded net counting rate for this group of electrons at a momentum setting p then the counting rate per unit momentum interval will be $N/\Delta p$. For the lens spectrometer

$$\Delta p = Rp$$

where R is the fractional line width, so that the total number of electrons emitted into the solid angle of the spectrometer per second is

$$\frac{1}{R} \int_0^{p \text{ max}} \frac{N}{p} dp .$$

This may be put in more convenient form by utilizing the fact that the momentum p is proportional to the voltage v across the standard series resistor. Thus if N_c is the counting rate at the conversion line above the continuum then the conversion coefficient α is given by

$$\alpha = \frac{N_c}{\frac{1}{R} \int_0^{v \text{ max}} \frac{N}{v} dv} = \frac{N_c R}{\int_0^{v \text{ max}} \frac{N}{v} dv}$$

The ratio of the K conversion coefficient α_K to the L conversion coefficient α_L may often be measured more accurately than either α_K or α_L separately. This is done simply by taking the ratio of heights of the K and L conversion lines above the continuum; differential attenuation of the line heights by absorption in the counter window may have to be allowed for in the case of very low energy conversion

electrons. The ratio α_K/α_L is useful in determining the multipole character of the gamma radiation.

Gamma-ray energies are accurately measured by means of their photoelectron spectra. The photoelectron lines are superposed on a broad distribution of Compton electrons which extends from zero to a maximum energy given by

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

where $h\nu$ is the photon energy and α is $h\nu/mc^2$. A separate measurement of the Compton spectrum will serve to distinguish the photoelectron lines. Depending on the intensity of the gamma ray, K, L, and M photoelectron lines will appear with decreasing intensity. If more than one gamma ray is present the identification of the lines may be facilitated by taking a second spectrum using a radiator of different Z. The energy of the K photoelectrons will be shifted more than that of the L and M lines and this together with the knowledge of the K, L, and M differences for the two radiator elements is usually sufficient to make the identification complete.

(d) Coincidence Counting Techniques.

Tentative disintegration schemes may be deduced from the Kurie plot analysis together with the knowledge of the energies and relative intensities of the gamma rays. The use of coincidence counting techniques with or without the spectrometer affords a method of verifying the decay scheme. The method is based on the assumption that a gamma ray in cascade with a beta-ray is emitted following the beta ray (or vice-versa) in a time shorter than the resolving time of the coincidence measuring device. When two spectrometers are available one may be focussed on a conversion line while the other scans the whole spectrum. Alternatively a gamma counter may be placed behind the source and beta-gamma coincidences

recorded throughout the spectrum. The constancy of the coincidence rate per beta particle would be indicative of the simplicity of the spectrum.

Another simpler method, fundamentally similar but with decreased resolving power, is to replace the spectrometer with a beta-ray counter having absorbers placed between it and the source. A comprehensive analysis of coincidence techniques including a discussion of errors has been published by Dunworth⁽³⁴⁾. One interesting and important result is that the minimum error in the true coincidence counting rate is primarily determined not by the individual counting rates or the geometry of the experiment but by the relation of the source strength to the resolving time of the coincidence analyser. The optimum relation is

$$N = \frac{1}{2\tau}$$

where N is the source strength in disintegrations per second and τ is the resolving time of the coincidence circuit.

III. Apparatus

(a) The Spectrometer.

The beta-ray spectrometer used in these experiments is of the thin lens type described by Deutsch, Elliott and Evans⁽³⁵⁾. The coil, vacuum chamber and baffles were constructed by the National Research Council of Canada according to their own specifications. Plate I is a view of the spectrometer and a cross-sectional diagram is presented in Fig. I.

The focussing coil is wound in four sections each of 900 turns of #10 d.c.c. copper wire. Cooling coils of $\frac{1}{4}$ " O.D. copper tubing are placed between the sections of the coil and carry water supplied by the city mains. The spectrometer tube, about 40" long, 7 $\frac{7}{8}$ " O.D., and wall thickness $\frac{1}{8}$ ", is placed in the centre of the coil drum. On each of the two annular brass end plates of the coil drum are mounted three adjustable screws for suspending the spectrometer tube in its correct position relative to the field. The tube is terminated at each end by brass discs $\frac{5}{8}$ " thick, on one of which is mounted the source holder and on the other the end-window Geiger counter.

The baffles within the tube are made of $\frac{1}{8}$ " aluminum and are mounted on aluminum spiders in an aluminum tube which fits snugly in the chamber. A lead cylinder about 6" long is mounted on the axis of the tube to prevent gamma radiation from reaching the counter directly. A full description of the theory and operation of this type of spectrometer may be found in the paper by Deutsch et al⁽³⁵⁾.

A new source holder and vacuum lock for the spectrometer was designed by the author and constructed in the machine shop of the Radiation Laboratory. A vertical section of it is presented in Fig. 2. and in Plate II it may be seen mounted on the spectrometer. The vacuum

lock is made from a standard 2" Crane non-rising-stem gate valve as suggested by Kurie⁽³⁶⁾. The wedge-shaped gate is fitted with annular neoprene gaskets as shown in Fig. 2. The stem is sealed off with an "O" ring and the bonnet is made vacuum tight by an annular neoprene gasket. The gate valve is threaded onto a brass pipe hard-soldered to the end plate of the spectrometer and the threads are made vacuum tight with a rubber gasket compound which solidifies in about one day after application. At the other end of the valve another short length of brass pipe is screwed in and sealed with gasket compound. This pipe is fitted with a flange grooved to take a large "O" ring. The source holder is mounted on a sliding shaft which is, in turn, carried by a brass tube with a large flange at one end. A knurled brass retaining cap clamps this flange against the large "O" ring to serve as a vacuum seal. The flange has an annular shoulder which fits into the "O" ring groove to centre the source accurately each time it is replaced. The sliding shaft fits into the brass tube with a clearance of about 0.002" and is sealed off with two small "O" rings. The source itself is mounted on a mica disc 1 1/8" in diameter which is held by an aluminum cup. The cup is screwed to the sliding shaft and its walls were turned down to 0.010" to reduce the effects of back-scattering. A further improvement would be the use of about three aluminum rods to support the source mounting ring. The device has operated in a completely satisfactory manner since installation.

(b) The Vacuum System.

A Cenco Megavac rotary oil sealed pump backing a three stage fractionating oil diffusion pump are used to evacuate the spectrometer tube. The counter filling system is joined through a stopcock to the fore-vacuum line to enable the thin-window counter to be pumped down at

the same time as the spectrometer tube. Thermocouple gauges are mounted on the spectrometer and the filling system. An ionization gauge on the spectrometer serves to measure pressures below 1 micron when the diffusion pump is used. A four foot length of $\frac{1}{2}$ " O.D. copper sylphon tubing was used as a flexible vacuum connection between the filling system and the counter. At each end a flared copper tube was soft-soldered on and these were waxed onto the glass parts of the system.

(c) The Regulated Current Supply.

The lens current is supplied by a D.C. generator capable of delivering 20 amperes into the 15 ohm load of the lens coil. The current is controlled through the excitation of the generator as well as by a variable series element and stabilized by means of a high gain feedback circuit. The current supply was originally constructed by Dr. J.L. Wolfson⁽³⁷⁾ and subsequently the stabilizer circuit was modified and improved by Dr. D.G. Douglas⁽³⁸⁾.

A block diagram of the current supply is given in Fig. 3. A voltage proportional to the current is developed across a standard resistance in series with the lens coil. This voltage is compared with the voltage on the arm of a Rubicon Type S potentiometer which is calibrated by an Eppley standard cell, and the difference is indicated on a Rubicon spotlight galvanometer as well as being fed into a high gain D.C. amplifier. The output of the amplifier is applied to 6 type 6A3 triodes which control the current flowing into the generator field from a conventional full wave rectifier supply. The D.C. amplifier employs a 60 c.p.s. square wave signal to achieve the high gain. It is controlled in amplitude and phase by the magnitude and polarity respectively of the D.C. error signal voltage fed into a Brown Converter⁽³⁹⁾. The 60 c.p.s. signal is inverted

to a D.C. control voltage at a high level by a phase sensitive rectifier. In series with the lens coil is a bank of 8 type 6AS7 triodes connected in parallel and shunted by a variable power resistor. To the grids of these tubes is applied the same D.C. control voltage that appears on the 6A3 grids. The control is degenerative in both cases.

At a given potentiometer setting the galvanometer is brought to a null reading by manipulation of both a variac in the generator field current supply and the resistor shunting the 6AS7 tubes. The latter are made to carry as much current as possible to improve the control. By this method the coil current is made proportional to the potentiometer setting. A low pass RC filter in the generator output was found to reduce a small amplitude hunting which was otherwise present. The current was maintained constant to within 0.1% at any setting of the potentiometer.

(d) Stray Field Neutralization.

The earth's magnetic field must be neutralized so that the only field acting on the beta-rays is the field of the lens coil. This is accomplished approximately by putting the axis of the spectrometer parallel with the horizontal projection of the earth's field and by neutralizing separately the horizontal and vertical components of the earth's field by means of two sets of Helmholtz coils with mutually perpendicular axes. These coils are mounted on rectangular wooden frames mounted on the spectrometer table. They may be seen in Plate I. Current is supplied to them by storage batteries. The currents used were those determined by Stephens-Newsham⁽⁹⁾. The stray field from the cyclotron magnet upsets the neutralization and no attempt was made to use the spectrometer when the cyclotron was running.

(e) Counters and Scalers.

The beta-counter used on the spectrometer is of the end window

type. It has copper walls and is fitted with brass plates shaped to fit into the end plate of the spectrometer. A 0.004" tungsten wire with a glass bead on the unsupported end forms the anode. For most of the work nylon films were used to permit the counting of low energy electrons. These were prepared according to the method described by Brown, Felber, Richards and Saxon⁽⁴⁰⁾. The diameter of the window is 5 mm. and a small bar of brass with a central hole was placed across this to support the nylon film. Ethylene with argon in a 20/80 mixture at a total pressure of 6.5 cms was used. Lack of stability and poor plateaus was often found and was attributed to impurities in either the ethylene or the argon. Tank argon and ethylene were used throughout. Window thicknesses of 0.050 to 0.100 mgm/cm² were used. For the photoelectron spectra a mica window of 1.1 mgm/cm² with an alcohol-argon filling was used with more success.

For coincidence measurements a Victoreen mica-window beta-counter and brass-cathode gamma counters were used. The gamma counters have a wall thickness of 0.030" and have an effective length of approximately 4 cms.

A Berkeley Scientific Co. Model 1000-B decade scaler was used with the spectrometer. This includes a regulated R.F. power supply variable from 0-2500 volts. Even when used with a Sola voltage regulating transformer it showed drifts of about 1% for the first one or two hours of operation. For this reason it was left on as much as possible.

For the coincidence measurements apparatus manufactured by the Atomic Instrument Co. was available. It consisted of two Model 305 Regulated High Voltage Supplies, two Model 201 Pulse Amplifiers, two Model 101-A Scale of 64 units, one Model 502 Coincidence Analyser and one Model 307 Regulated Power Supply for the pulse amplifiers and

coincidence analyser. Nominal resolving times of $\frac{1}{4}$, $\frac{1}{2}$, 1 and 2 micro-seconds are available on this equipment.

IV. Experimental

(a) Adjustment and Calibration.

It is essential that the line joining the source and counter window accurately coincide with the magnetic axis of the lens. The fact that the tube may not be of perfect circular cross-section is immaterial. The procedure for aligning the tube involves the measurement of a conversion line, preferably a K-conversion line. L-conversion lines are measurably broader than K lines as a result of the separation in energy of the L_I , L_{II} , and L_{III} electron levels. Adjustment is made for maximum intensity and the realization of the predicted line shape.

These experiments began shortly after Dr. D.G. Douglas⁽³⁸⁾ completed his work on Lu^{177} . For the first beta-spectrum measured by the author the alignment was unchanged and found to be satisfactory as indicated by the shape of the F line of ThB. At a later date, however, an accident occurred which necessitated the realignment and recalibration of the spectrometer. On a very humid day, some water, which had condensed on the copper cooling tubes at the point where they emerge from the coil drum, ran down into the coil drum and shorted the second section of the coil to ground. An attempt was made to repair the damage by heating the coil drum with infra-red lamps and by blowing air through the space between the coil sections. By this method the insulation resistance was raised from 2 ohms to about 2000 ohms in one day. Continued treatment made no further improvement. About one month later the insulation resistance had risen to its normal value of about 2 megohms. It soon broke down again when a large current was put through all four sections of the coil. This was probably due to thermal expansion of the cooling coil.

For the remainder of the work only the two outer sections of

the coil were used. Realignment was performed using the L-conversion line of Tm^{170} . As stated above the shape of an L line is not ideal for this purpose but Tm^{170} had the advantage of a half-life long compared with that of ThB.

The calibration was achieved by measuring the well known F line of ThB which has been measured by Seigbahn⁽⁴¹⁾ as 1383.8 gauss-cm. with a probable error of 0.2 to 0.3%. The ThB source was prepared by placing MsTh_2 in an activator and collecting for 24 hours the active deposit on a small disc of aluminum foil 0.00025" thick and 4 mm in diameter. Only one side of the foil was activated so that it was ideally thin. This was then placed in the source holder at the centre of a mica annulus covered with one layer of aluminum 0.00025" thick.

In the first calibration, using the four coil sections, the peak of the F line of ThB was found with a potentiometer setting of 290.2 ± 0.2 millivolts. The spectrometer constant under these conditions is

$$H\rho = 1383.8 \frac{V}{290.2} = 4.768 V \text{ gauss-cm.}$$

where $H\rho$ is the momentum of the electrons focussed when the potentiometer setting is V millivolts.

With only the outer two coils in use, the second calibration located the same line at 603.5 ± 0.5 millivolts so that the momentum of the focussed electrons was given by

$$H\rho = 2.293 V \text{ gauss-cm.}$$

As a check on the linearity of the instrument the A line was located at a potentiometer setting of 233 ± 1 millivolts. Assuming linearity this corresponds to an $H\rho$ of 535 ± 2 gauss cm. C.D. Ellis⁽⁴²⁾ lists this line at 541.0 gauss-cm. but states that this value should be lowered by $\frac{1}{2}\%$. Furthermore his relative $H\rho$ measurements are based on the value

1385.8 gauss-cm. for the F line. These considerations should lead to value of about 538.3 gauss-cm. and the agreement is considered satisfactory. The contours of the A and F lines are shown in Fig. 5.

(b) Sources.

The thulium in the form of a fine white powder of Tm_2O_3 was prepared for investigation by neutron irradiation in the heavy water pile of the National Research Council at Chalk River, Ontario. Two samples were irradiated; one of 9 milligrams in an aluminum capsule suitable for the study of the gamma rays in the spectrometer, the other of 0.7 milligrams from which the beta sources were prepared was placed in an aluminum capsule with a screw top. Both of these capsules were placed in the standard aluminum irradiation capsule used by the National Research Council, and shipped to Chalk River. A total activity of 50mc or the maximum possible in a three month irradiation was requested. On the packing note returned with the sample after a three month irradiation the activity was stated to be 50.0 millicuries. Assuming this to be correct the specific activity should have been about 5.1 millicuries per milligram upon receipt of the sample.

The mounting for the first beta-ray source was a nylon film 0.030 mgm/cm^2 with a semi-opaque layer of aluminum evaporated on it in vacuo. The nylon film was prepared by dissolving Dupont Type FM-6501 nylon chips in isobutyl alcohol using a double boiler. A drop of the solution placed on distilled water spreads out and dries in a few minutes. An annulus of thin mica $1\frac{1}{4}$ " O.D., $3/8$ " I.D. was then placed gently on the film, the excess nylon around the edge torn away and the mica picked up. After drying, a second and third film are layed on until the desired thickness is obtained. The thickness was measured by the Michelson interferometer method developed by Prof. A.I. McPherson and Dr. D.G. Douglas⁽⁴³⁾

of this University. A very rough estimate of the thickness of the evaporated aluminum layer was made by comparison with an nearly opaque film which could be weighed on a chemical balance. The thickness was estimated at about 0.02 mgm/cm^2 so that the total backing thickness was no more than 0.05 mgm/cm^2 .

The source was prepared simply by putting one drop of water on the inside of the screw cover of the smaller capsule, stirring some of the Tm_2O_3 powder into suspension then dropping the suspension on the backing, taking care to centre it. When this was dried, without applying heat, it appeared to be sufficiently uniform in thickness to be usable. The diameter of the source was 5.5 mm., slightly larger than the counter window. Good electrical contact between the aluminum film and the grounded source mounting ring was assured by folding a narrow strip of aluminum foil around the periphery of the mica disc.

The second beta-ray source was mounted on an aluminum foil stretched over a mica annulus identical with the first. The aluminum foil, initially 0.00025 " thick was placed on the mica, folded over the edge about $1/8$ ", then etched in a dilute solution of sodium hydroxide until the thickness was approximately 0.5 mgm/cm^2 . The radioactive material was placed on the foil in the manner described above. A well grounded source was thus assured.

For the measurement of the gamma-rays the capsule containing 9 mgm. of Tm_2O_3 was used. A lead foil 15.2 mgm/cm^2 thick and 6.0 mm. in diameter was used as the first radiator. The activity was later transferred to a copper capsule. This had the shape of a cup with a flat bottom. The thickness of the bottom was 340 mgm/cm^2 and its diameter 6 mm. This is not thick enough to stop all of the primary beta-rays. However a uranium radiator in front of this whose thickness of 130 mgm/cm^2

added to that of the copper capsule was sufficient to stop the primary beta-rays whose maximum energy was found to be 970 kev. The diameter of the uranium metal foil was 8 mm. When this capsule was used with a lead foil of 15.2 mgm/cm^2 and a tin foil of 13.3 mgm/cm^2 a sheet of copper about 200 mgm/cm^2 was placed between the capsule and the foil.

For the half-life measurement a trace of the Tm_2O_3 was placed on a sheet of mica and put on the tray of a Lauritsen quartz fibre electroscope, Model I, made the Fred C. Henson Co., Pasadena, Calif. Measurements of the activity were taken periodically for $3\frac{1}{2}$ months.

(c) The Beta-ray Spectrum.

The counting rate measurements on the first beta-ray source were completed within 50 hours of starting. The corrections for decay of the source were therefore negligible for most of the counting rate measurements. The source was strong and gave, at the peak of the most intense conversion line, a counting rate of 240 counts per second and at the peak of the continuum a counting rate of 12.8 counts per second. Low statistical errors were readily obtained in a short time. At most of the settings between 8,000 and 12,000 total counts were taken and at no part of the spectrum were less than 4000 counts recorded. At the upper end of the spectrum where the counting rate was relatively low, the procedure was adopted of sweeping over the region several times until an adequate total count was registered. In this way the effects of poor current regulation in the coils and counter drifts were undoubtedly decreased.

Conversion lines were found at 22.51, 72.8 and 80.7 kev. The estimated uncertainty in these determinations is $\pm 1\%$.

The Kurie plot for this spectrum, using the Coulomb correction factor for $Z = 70$, is given in Fig. 7. It is evident that the Kurie plot

is, for the most part, essentially straight. Near the high energy end, however, a marked deviation from the straight line occurs exceeding the probable errors in the experimental points. For the sake of clarity, not all of the experimental points are shown. The lines have been transferred from a large scale drawing containing all of the points.

The end point of the spectrum is found at (970 ± 5) kev. If the straight line portion at the upper end is produced toward the low energy end and a subtraction is made according to the method discussed earlier, a second end point is found at (900 ± 20) kev. The uncertainty of this determination is large owing to the subtraction.

It will be recalled that the first beta-ray source was mounted on a nylon film covered with an evaporated film of aluminum. In order to check on the efficacy of the aluminum film as a ground, as well as to study the effects of self-absorption in the source, a second source of about 1/10 of the intensity of the first was mounted on an aluminum foil 0.5 mgm/cm^2 . The resulting spectrum is shown in Fig. 6. For these measurements the two outer coils were used, the spectrometer having been recalibrated as stated earlier. In this spectrum the second and third conversion lines are more completely resolved than in the first. In addition, the height of the conversion lines relative to the continuum has been increased. It is uncertain whether these effects were due to using a thinner source or to the slightly increased resolving power resulting from the use of only the two outer coils. Both may have been contributing factors. The thickness of the first source was approximately 0.25 mgm/cm^2 whereas the second (with lower specific activity) was about 0.10 mgm/cm^2 . To illustrate the improvement in resolution the half-width of the second conversion line in the first spectrum was 4.47% and in the second it was reduced to 3.32%.

In the second spectrum, the conversion lines were found at

22.75, 74.1, and 81.6 kev. respectively. These values, although consistently slightly higher than those from the first spectrum, do not reveal any significant source charging effects in the first spectrum. A Kurie plot of the second spectrum yielded an end point of (970 ± 10) kev. The larger error here is due to the large probable errors in the experimental points. The statistical errors were such that one could not say with certainty that a deviation from a straight line existed at the high energy end of this Kurie plot. At the low energy end, however, the Kurie plot was straight down to a lower energy than the first.

(d) The Photoelectron Spectra.

The photoelectron spectrum obtained with a lead radiator, 15.2 mgm/cm^2 revealed a group of lines attributed to L and M photoelectrons arising from X-rays and two lines at 68.2 and 79.8 kev. respectively. These lines may be seen in Fig. 4. In addition, a broad distribution of electrons was found extending up to approximately 900 kev. In a thorough search of the upper part of this distribution no lines were found. The active material was then transferred to the copper capsule described above and a uranium radiator of 130 mgm/cm^2 used. The broad distribution was now not present and only the lines in the low energy region were found. This effect was attributed to (i) possible contamination of the outer surface of the aluminum capsule with active Tm_2O_3 and (ii) possible insufficient stopping power of the aluminum capsule for the primary beta-rays. When the copper capsule with adequate stopping power was used consistent results were obtained with uranium, lead and tin radiators. With a tin radiator 13.3 mgm/cm^2 the K photoelectron yield was too low to be of any value for energy determination. The uranium radiator, 130 mgm/cm^2 gave L and M photoelectron lines which were less well defined

than those obtained with lead. It was used primarily to search for lines at higher energies.

The lead photoelectron lines at 68.2 and 79.8 kev. respectively may be attributed to a single gamma-ray. If the L and M binding energies of lead are added to these, gamma-ray energies of 83.7 and 82.5 kev. respectively are obtained.

(e) Coincidence Measurements.

Some coincidence experiments were performed in an effort to obtain evidence concerning the complexity of the decay scheme. A brass gamma-ray counter and a 2.1 mgm/cm² mica window beta-ray counter were placed on opposite sides of a Tm¹⁷⁰ source. A 16 mgm/cm² aluminum absorber was placed in front of the beta counter to avoid registering coincidences between conversion electrons and X-rays. Coincidences were counted for 48 hours and the separate beta and gamma counting rates were monitored from time to time. The total coincidence rate was found to be $(5.54 \pm 0.12) \times 10^{-3}$ per sec. The resolving time of the coincidence mixer, $(0.453 \pm 0.072) \times 10^{-6}$ secs., was measured by the method of two independent sources described by Dunworth⁽³⁴⁾. The random coincidence counting rate, given by the product of the beta and gamma counting rates and twice the resolving time was $(5.15 \pm 0.8) \times 10^{-3}$ per sec. Subtracting this from the total coincidence rate gives $(3.9 \pm 2) \times 10^{-4}$ per sec. as the true beta-gamma coincidence rate. This figure, divided by the beta counting rate, 125.1 per sec., gives $(3.1 \pm 1.6) \times 10^{-6}$ as the number of beta-gamma coincidences per beta ray. This experiment was repeated with the same gamma counter geometry using a thulium source of approximately one third the intensity. In a total counting time of 24 hours decreased statistical errors were obtained and the result was $(3.0 \pm .75) \times 10^{-6}$ beta-gamma

coincidences per beta ray.

For the case of a simple decay scheme consisting of one gamma-ray in cascade with a beta-ray, this ratio should be equal to the efficiency of the gamma counter for the geometry used in the experiment⁽³⁴⁾. In order to test whether this was true a rough calibration of the gamma counter was performed. The beta counter was replaced by a second brass-cathode gamma counter and the Tm^{170} source replaced by a small source of Co^{60} . It is known that Co^{60} emits two gamma-rays in cascade and the energies are given by Jensen, Laslett and Pratt⁽⁴⁴⁾ as 1.15 and 1.31 Mev. respectively. Gamma-gamma coincidence counts were registered for 9 hours. The significant result desired from this experiment was the efficiency of the first gamma counter for the average Co^{60} gamma ray energy of 1.23 Mev. and this was found to be $(1.45 \pm 0.05) \times 10^{-3}$ coincidences per gamma ray. From a semi-empirical calibration curve, for a brass-cathode gamma counter approximately the same dimensions used here, given by von Droste⁽⁴⁵⁾, it is estimated that the efficiency of the counter for 84 kev. radiation was approximately 20% of that for 1.23 Mev. gamma rays. Thus the efficiency of the gamma counter for an 84 kev. gamma ray in the experimental coincidence arrangement should be $(2.9 \pm 0.2) \times 10^{-4}$ coincidence per gamma ray.

(f) Half-life Measurement.

Time did not permit of an accurate measurement of the half-life of Tm^{170} . A decay curve was obtained by measurements of the activity of a sample of Tm^{170} extending over a period of $3\frac{1}{2}$ months in an effort to detect the presence of possible impurities. A plot of the logarithm of the activity versus time yields a value of the half-life of (126 ± 5) days in good agreement with Bothe's value of (127 ± 5) days. It is concluded that impurities did not appreciably contribute to the activity.

(g) Counter Window Correction.

An attempt was made to estimate the effects of absorption of low energy electrons by the counter window. The method used was that described by Witcher⁽⁴⁶⁾. A nylon counter window of 0.050 mgm/cm^2 was prepared and nylon absorbers of 0.050 , 0.100 , and 0.150 mgm/cm^2 respectively placed on the mica frame shown in Plate III. This mica frame may be rotated by a brass shaft mounted in the end plate of the spectrometer and sealed off by two "O" rings. In this way the absorbers could be placed conveniently before the counter window. A strong source was prepared and with the spectrometer focussed in the low energy region an absorption curve was obtained at several current settings. A fourth hole was cut in the mica frame to allow for scattering effects from the mica when no absorber was placed before the counter. The correction factors were obtained by extrapolating the absorption curves to zero thickness, and taking the ratio of the net counting rate corresponding to zero thickness to that corresponding to the window thickness employed. The correction factors for windows of 0.055 and 0.100 mgm/cm^2 respectively are shown in Fig. 9. The former window was used for the first beta-spectrum and the latter for the second.

There is one rather serious limitation in this method of obtaining the correction factors. It is probable that some electrons, although not stopped by the absorbers, may be deflected by a large angle and not be detected by the counter. For this reason the experimental correction factors must be regarded as only an upper limit to the true correction factors.

V. Discussion of Results.

The most reliable determination of the gamma-ray energy is to be found in the internal conversion lines visible in the beta-spectrum. Table I summarizes the energy values obtained from the beta-spectrum and the photoelectron spectrum taken with a lead radiator. The values stated for the conversion electron energies are the averages taken from the two measurements of the beta-ray spectrum.

Table I

Energy of the Gamma Ray

<u>Line</u>	<u>Location (kev)</u>	<u>Added Binding Energy (kev)</u>	<u>Energy (kev)</u>
Conversion	22.6	61.3 (K)	83.9
Conversion	73.5	10.5 (L)	84.0
Conversion	81.2	2.3 (M)	83.4
Photoelectron (Pb Radiator)	68.2	15.5 (L)	83.7
Photoelectron (Pb Radiator)	79.8	2.7 (M)	82.5

The binding energy values added to the corresponding conversion electron energies are those characteristic of ytterbium whose atomic number is one greater than that of thulium.

If the K and L binding energies of thulium, 59.3 and 10.1 kev. respectively are added to the K and L conversion electron energies, gamma ray energies of 81.9 and 83.6 kev. respectively are obtained. The results are, however, more self-consistent if the assumption is made that the gamma ray is converted in ytterbium, that is to say that the gamma ray follows the beta decay. Furthermore the possibility that Tm^{170} is a positron emitter is ruled out by the evidence for the gamma ray energy. If the gamma ray were internally converted in erbium, $Z = 68$, its K

binding energy of 57.5 kev. would lead to a gamma energy of 80.1 kev., in definite disagreement with the photoelectron data. The conclusion is that the single gamma ray has an energy of (83.9 ± 0.2) kev. and is partially internally converted in ${}_{70}\text{Yb}^{170}$. This result agrees well with Ketelle's measurement of 84 kev.⁽¹³⁾ whereas Saxon and Richards⁽¹⁴⁾ reported a value of (85.5 ± 0.5) kev.

The beta end point of (970 ± 5) kev. is to be compared with Saxon and Richards value of (970 ± 10) kev. and Ketelle's result of 980 kev. The error of ± 5 kev. is felt by the author to be justified in view of the close agreement obtained from the two spectra which were taken with independent calibration and different spectrometer adjustments. From the Kurie plot a lower energy beta group is found with an end point of (900 ± 20) kev. with an intensity estimated at 10% of the transitions.

The beta end points and the gamma-ray energy are consistent with the decay scheme illustrated in Fig. 8. The value 886 kev. is used for the end point of the lower intensity group, this being the difference in energy of the upper end point and the gamma-ray energy.

The complexity of the beta-spectrum is supported by the results of the coincidence experiments. Consider first the case of a simple decay scheme consisting of one beta-ray group followed by a gamma-ray. Then for a beta-gamma coincidence experiment the following relations should hold:

$$\text{Beta counting rate } C_{\beta} = N \epsilon_{\beta}$$

$$\text{Gamma counting rate } C_{\gamma} = N \epsilon_{\gamma}$$

$$\text{Coincidence counting rate } C_c = N \epsilon_{\beta} \epsilon_{\gamma}$$

where N = number of disintegrations per sec., and ϵ_{β} , ϵ_{γ} are the counter efficiencies. The ratio

$$\frac{C_c}{C_\beta} = \frac{N \epsilon_\beta \epsilon_\gamma}{N \epsilon_\beta} = \epsilon_\gamma$$

gives the efficiency of the gamma counter for the experimental geometry.

Consider now the case of a decay scheme such as shown in Fig.

8. Let N_1 , N_2 be the number of disintegrations per second in the upper and lower energy beta groups respectively per second. Let α be the total conversion coefficient for the gamma rays which we shall assume is not negligible with respect to one. Then

$$C_\beta = (N_1 + N_2) \epsilon_\beta$$

$$C_\gamma = N_2(1 - \alpha) \epsilon_\gamma$$

$$C_c = N_2(1 - \alpha) \epsilon_\beta \epsilon_\gamma$$

are the pertinent equations. In both cases it is assumed that the conversion electrons and a small fraction of the beta particles are not counted. An absorber, 16 mgm/cm² of aluminum, was used in the experiment. In this case the ratio

$$\frac{C_c}{C_\beta} = \frac{N_2(1 - \alpha)}{N_1 + N_2} \epsilon_\gamma$$

is smaller than ϵ_γ by the factor $\frac{N_2(1 - \alpha)}{N_1 + N_2}$.

The average of the two experimental values obtained for the ratio C_c/C_β is $(3.05 \pm 1.0) \times 10^{-6}$. The value to be expected in the case of a simple spectrum as obtained from the calibration of the coincidence apparatus is $(2.9 \pm 0.2) \times 10^{-4}$. The inference is that the factor $N_2(1 - \alpha)/N_1 + N_2$ is of the order of 0.01. The Kurie plot analysis of the beta-spectrum gives a value of approximately 0.1 for the ratio $N_2/N_1 + N_2$. Thus

$$1 - \alpha \cong 0.1$$

$$\text{and } \alpha \cong 0.9.$$

Table II gives approximate values for the conversion coefficients based on the assumption that the gamma rays follow the lower energy beta

ray group.

Table II

Conversion Coefficients

<u>Line</u>	<u>Conversion Coefficient</u>
K	0.08
L	0.55
M	<u>0.17</u>
Total	0.80

The K conversion coefficient has been corrected by 20% for window absorption. These values were taken from the second beta-spectrum for which a window of 0.100 mgm/cm^2 was used.

In view of the errors present in the beta-gamma coincidence rate and in the intensity estimate from the Kurie plot, the agreement of this value of the total conversion coefficient with that expected from the coincidence experiment is perhaps fortuitously good. The coincidence experiment does, nevertheless, lend strong support to the decay scheme shown in Fig. 8.

Evidence for the low intensity of the gamma-ray may be found also in the photoelectron spectrum, Fig. 4. The lower group of lines are attributed to x-ray photoelectrons. Table III lists the K_{α} and K_{β} x-rays characteristic of ytterbium as calculated from wavelengths given by Compton and Allison⁽⁴⁷⁾.

Table III

Energy of Characteristic K-lines (kev) of Ytterbium.

<u>Transition</u>	<u>Energy</u>
K-L _{II}	51.44
K-L _{III}	52.46
K-M _{II}	59.26
K-M _{III}	59.49
K-N _{II} . N _{III}	60.99

These two groups of x-rays should give rise to groups of L photoelectrons from the Pb radiator at about 35.5 and 44 kev. respectively. The peaks were located at 34.9 and 44.8 kev. respectively.

These x-rays arise, therefore, from transitions to the K level made vacant by the internal conversion of the gamma rays in the K level. The assumption of a simple decay scheme would lead to a value for the K conversion coefficient of less than 0.010 and a total conversion coefficient of less than 0.10, resulting in a low yield of K x-rays, relative to unconverted gamma radiation. Inspection of the photoelectron spectrum reveals that the K x-rays are certainly not less intense than the unconverted gamma-rays. The uncertain variation of the L photoelectron cross-section with energy makes a more accurate estimate of the relative intensities impossible but the variation of the momentum interval of the spectrometer will be a compensating factor.

The ft values⁽²⁰⁾ for the two component spectra indicated by the Kurie analysis have been calculated and are given in Table IV.

Table IV

ft Values

<u>Energy Point</u>	<u>Relative Intensity</u>	<u>Partial Half-Life t, (sec)</u>	<u>f</u>	<u>ft</u>	<u>Group</u>	<u>Order</u>
970	90	1.2×10^6	4.2×10^2	5.0×10^8	2B	2nd Forbidden
886	10	1.1×10^7	3.0×10^2	33.0×10^9	2B	2nd Forbidden

Not much information concerning the character of the gamma radiation can be deduced from the conversion coefficients. The ratios of the K, L and the L,M coefficients are

$$\frac{\alpha_K}{\alpha_L} = 0.15 ; \frac{\alpha_L}{\alpha_M} = 3.2.$$

From the curves given by Hebb and Nelson⁽⁴⁸⁾ one may say that the spin change in the gamma-transition is greater than one. Their calculation dealt only with electric multipole radiation; a recent paper by Drell⁽⁴⁹⁾ states that for elements of medium Z magnetic K-conversion is as important as electric K-conversion. Furthermore the large magnetic conversion will influence the experimentally observed K to L-conversion ratio. The above deduction concerning the spin, therefore, may have no significance.

VI. Conclusions.

1. The disintegration of $^{69}\text{Tm}^{170}$ is complex, and two beta-ray groups are found with end points at (970 ± 5) and (886 ± 5) kev. respectively.
2. A single weak gamma-ray has been observed and measured by the study of conversion electrons and the photoelectrons ejected from tin, lead, and uranium radiators. The energy has been found to be (83.9 ± 0.2) kev.
3. The complexity of the beta spectrum has been ascertained by a beta-gamma coincidence experiment. It was found that gamma rays are emitted in coincidence with about 1% of the beta rays. In the absence of an isomeric state⁽¹³⁾ this leads to the conclusion that the gamma rays are in coincidence with a beta-ray group of low intensity.
4. A decay scheme, consistent with these results, is proposed and is illustrated in Fig. 8.
5. The K, L, and M conversion coefficients for the gamma-ray have been found to be approximately 0.08, 0.55, 0.17 respectively.
6. In the absence of theoretical calculations applicable to this case no definite conclusion can be drawn concerning the spin change in the gamma transition.
7. The mounting of the source on a nylon film covered with an evaporated film of aluminum proved to be a satisfactory method of electrically grounding the source.
8. The absorption method described by Witcher⁽⁴⁶⁾ was of limited use in estimating the nylon window correction factors for the low energy region of the beta spectrum. As a result of possible scattering by the nylon absorbers, only an upper limit to the true correction factors could be deduced from the experimental data.

9. The decay of the activity was followed for $3\frac{1}{2}$ months and the half-life found to be 126 ± 5 days, consistent with Bothe's value of 127 days⁽⁴⁾.

VII. . Appendix

Tabulation of Coulomb Correction Factor for $Z = 70$

The values of $F(Z,W)$ are on an arbitrary scale.

<u>Kinetic Energy Kilovolts</u>	<u>$F(Z,W)$</u>
10	16.18
20	11.48
30	9.435
40	8.206
50	7.374
60	6.766
70	6.299
80	5.924
90	5.615
100	5.357
120	4.945
140	4.627
160	4.380
180	4.179
200	4.008
225	3.826
250	3.677
275	3.549
300	3.440
350	3.259
400	3.116
450	2.999

<u>Kinetic Energy Kilovolts</u>	<u>F(Z,W)</u>
500	2.900
550	2.817
600	2.743
650	2.679
700	2.622
750	2.570
800	2.523
850	2.480
900	2.444
950	2.405
1000	2.372
1050	2.341
1100	2.312

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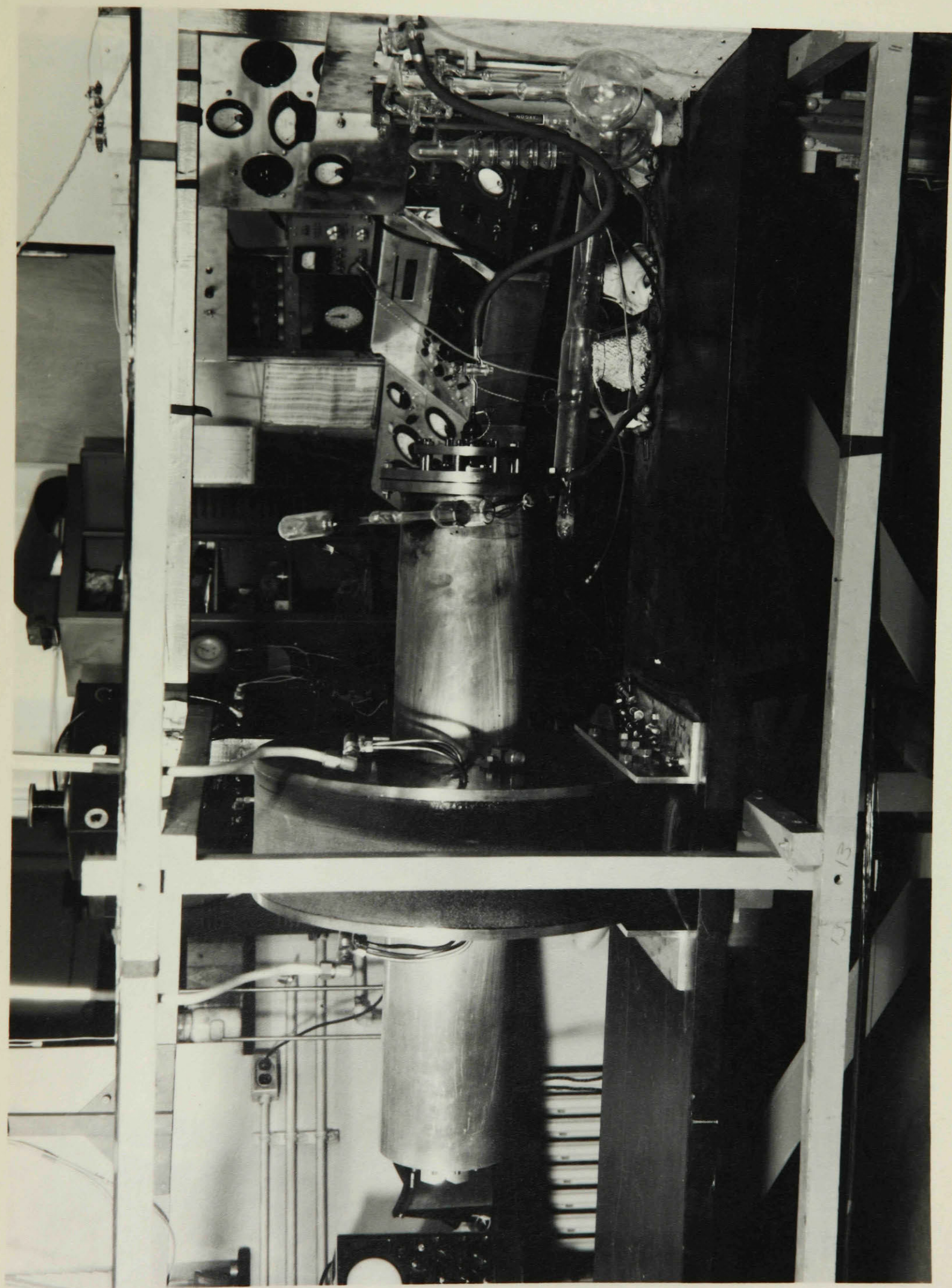


PLATE I SPECTROMETER

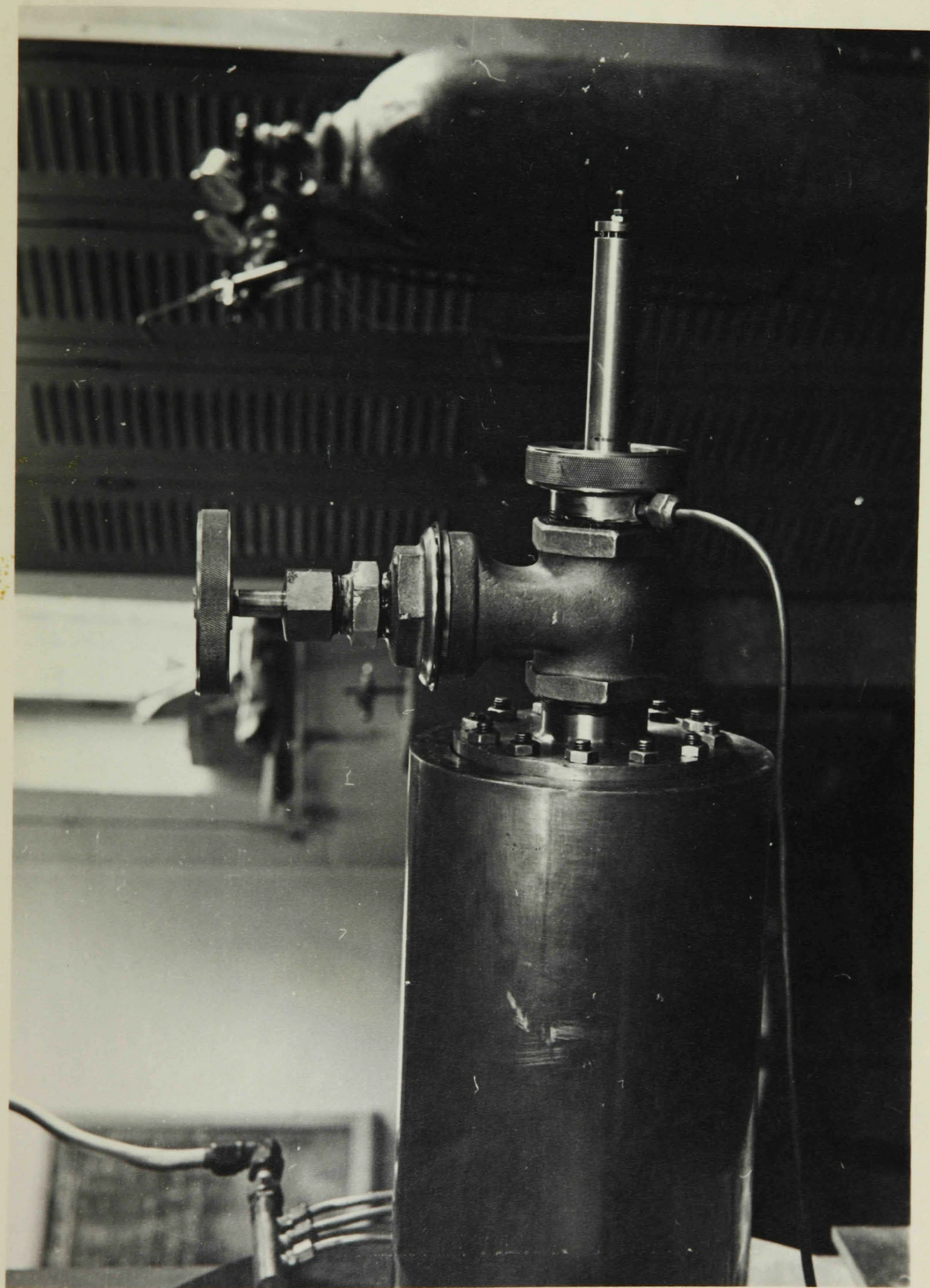


PLATE II SOURCE HOLDER

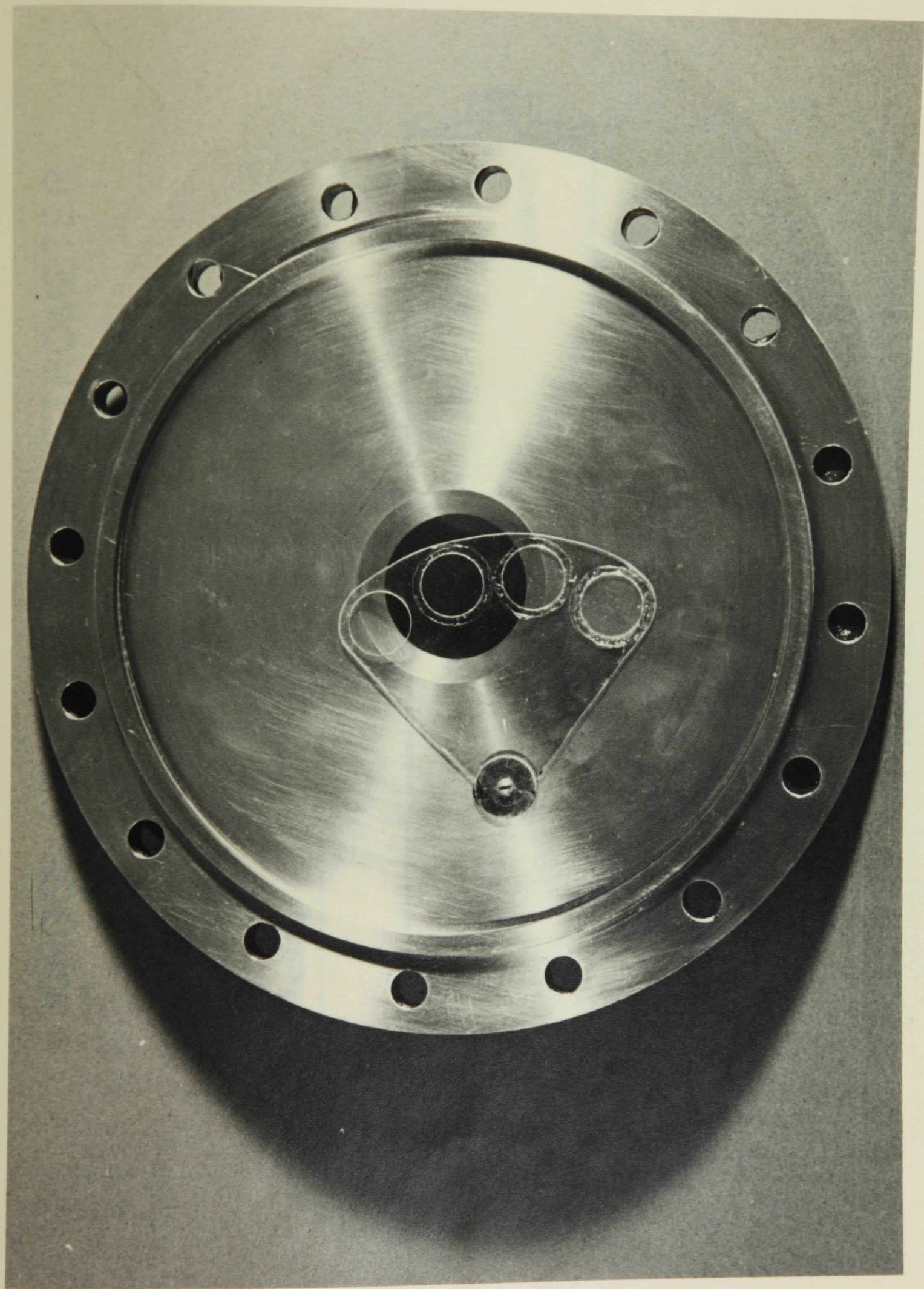


PLATE III NYLON ABSORBERS

CROSS SECTION OF SPECTROMETER

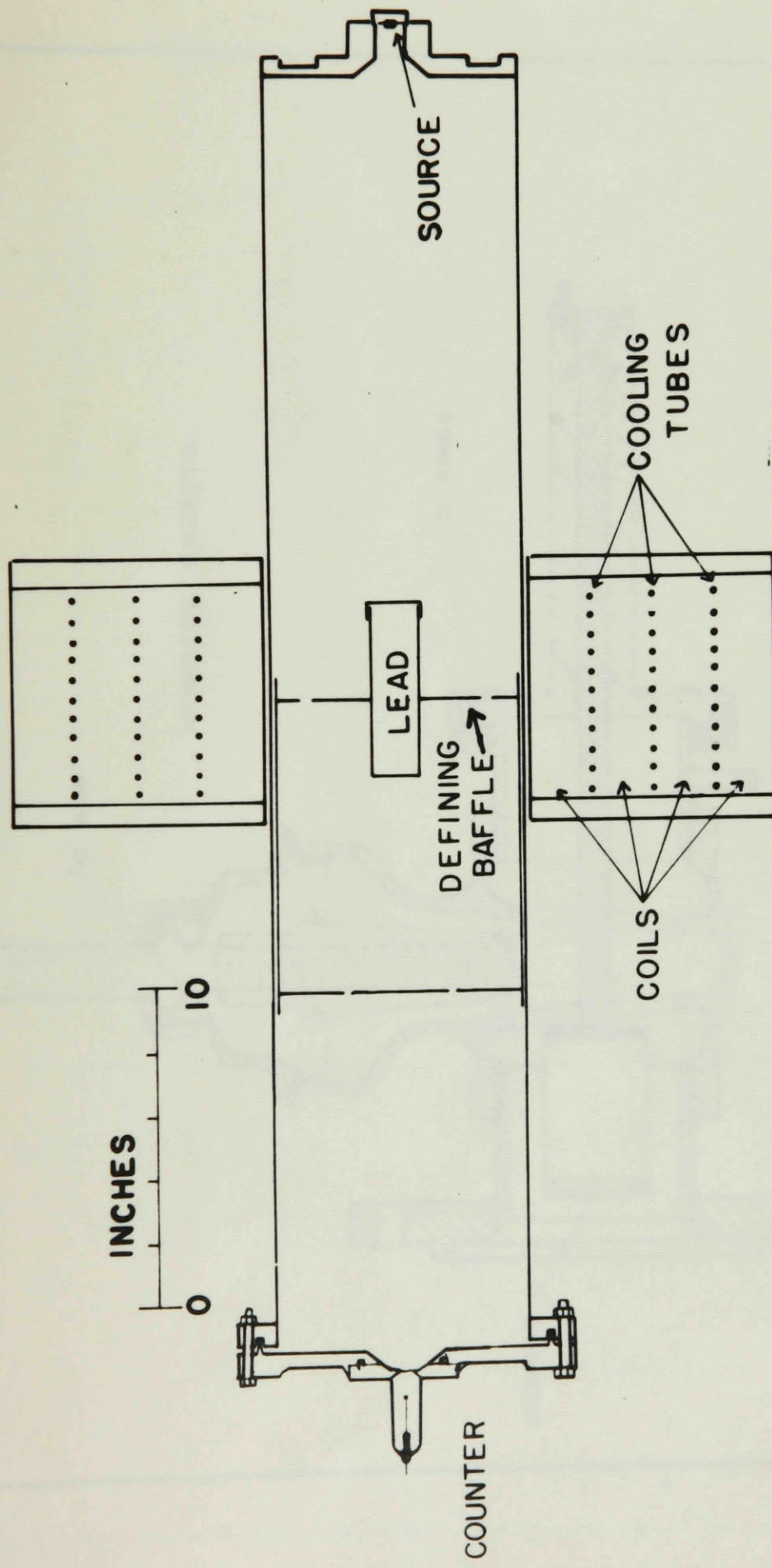


FIGURE 1.

SOURCE HOLDER
AND
VACUUM LOCK

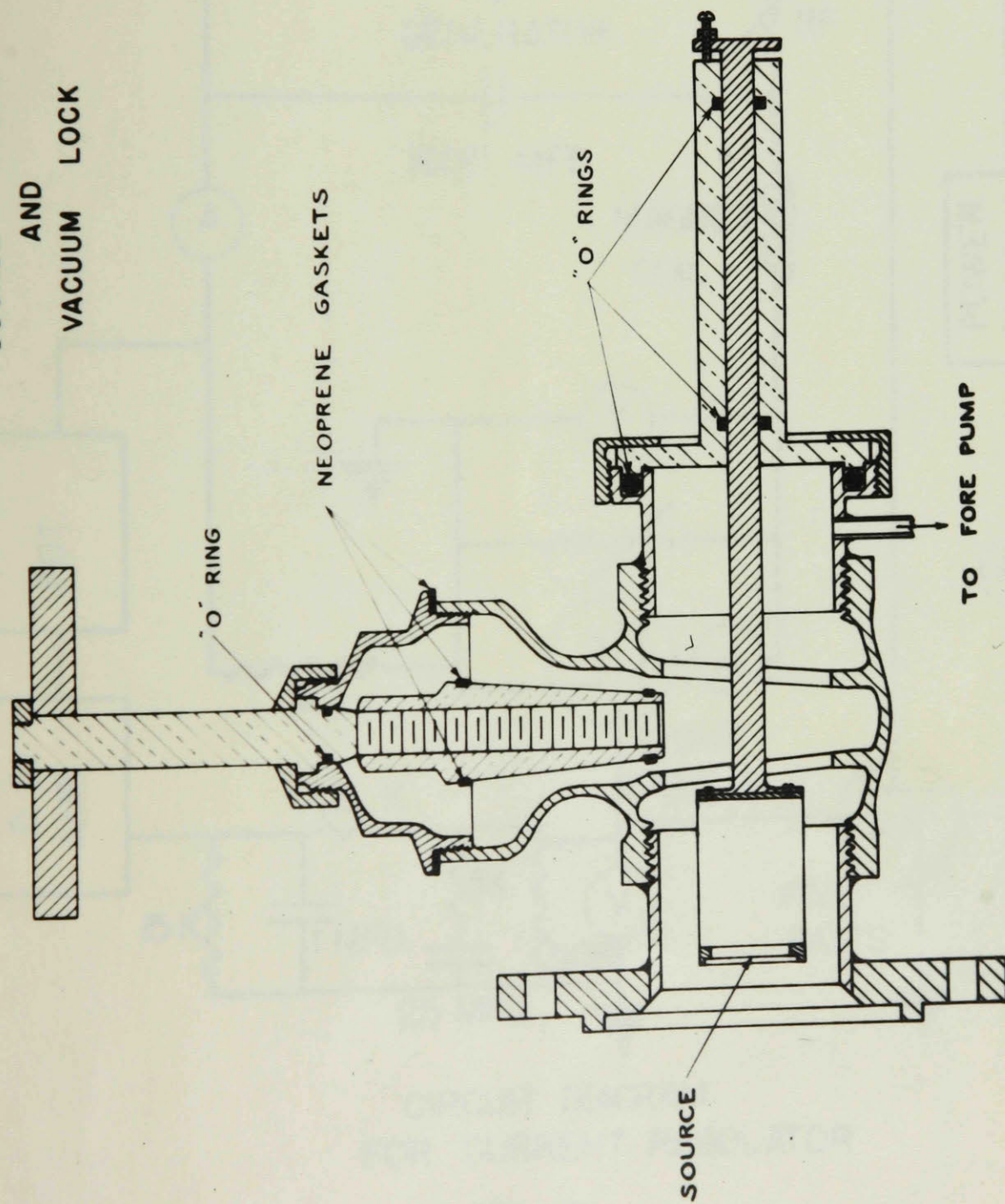
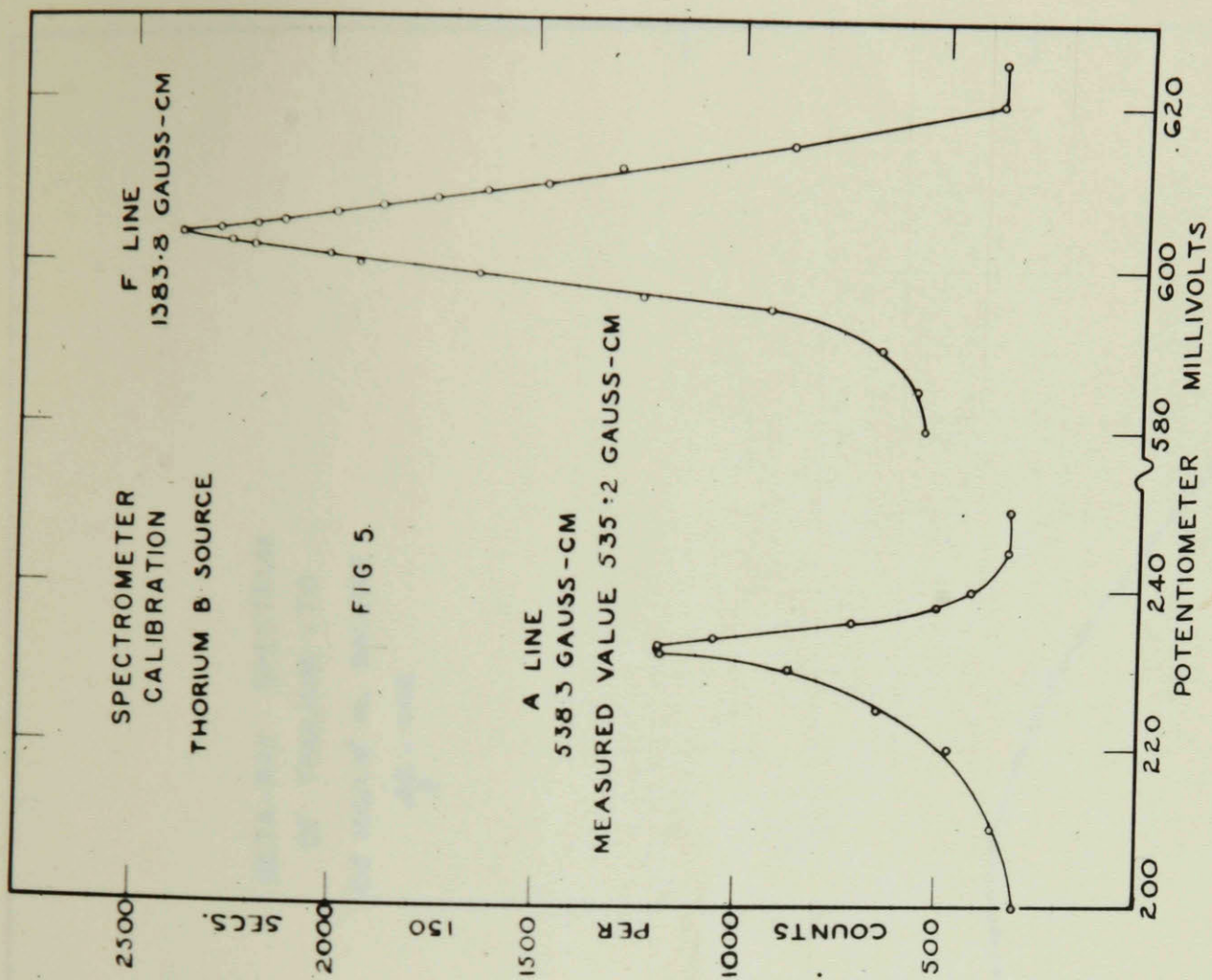
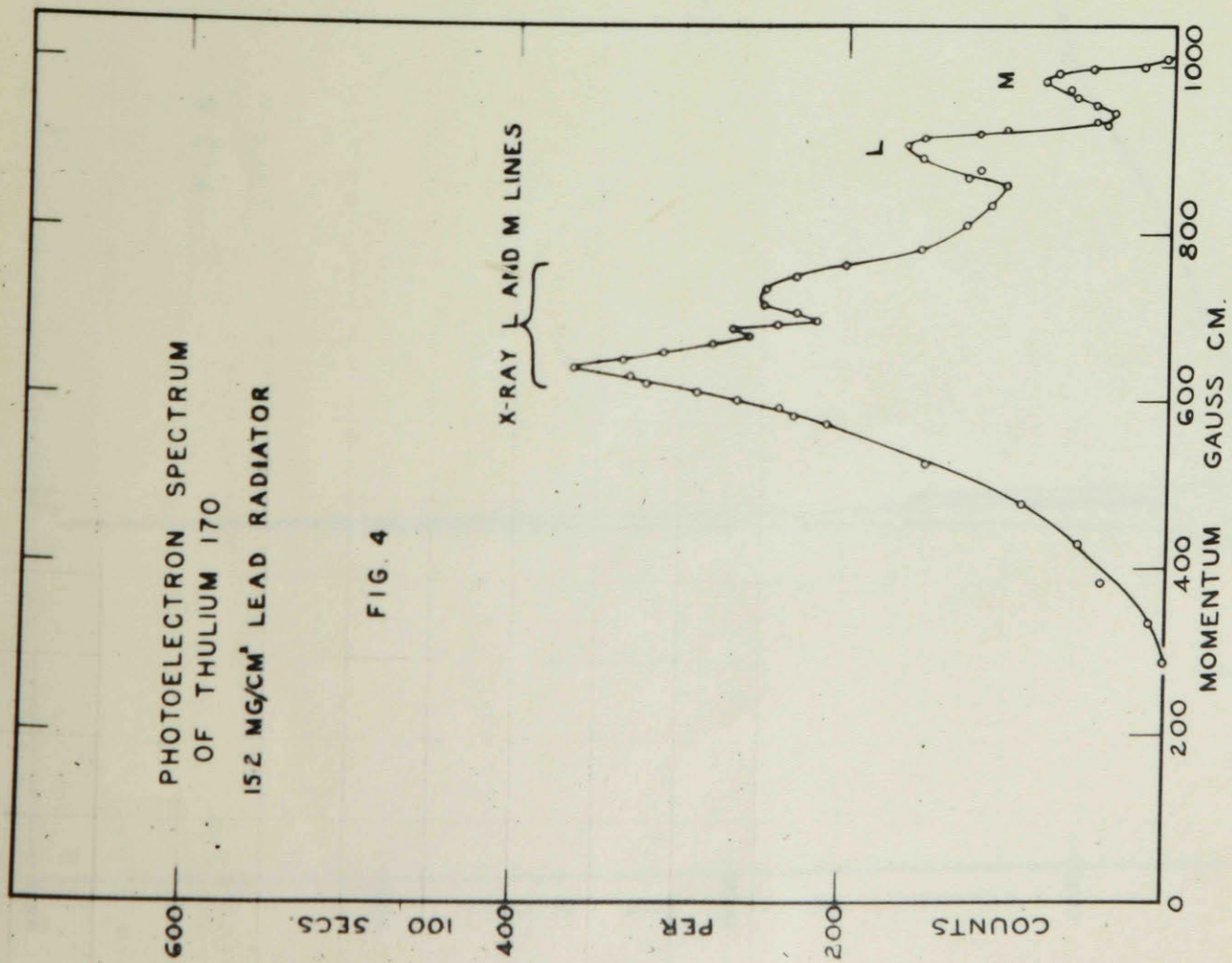
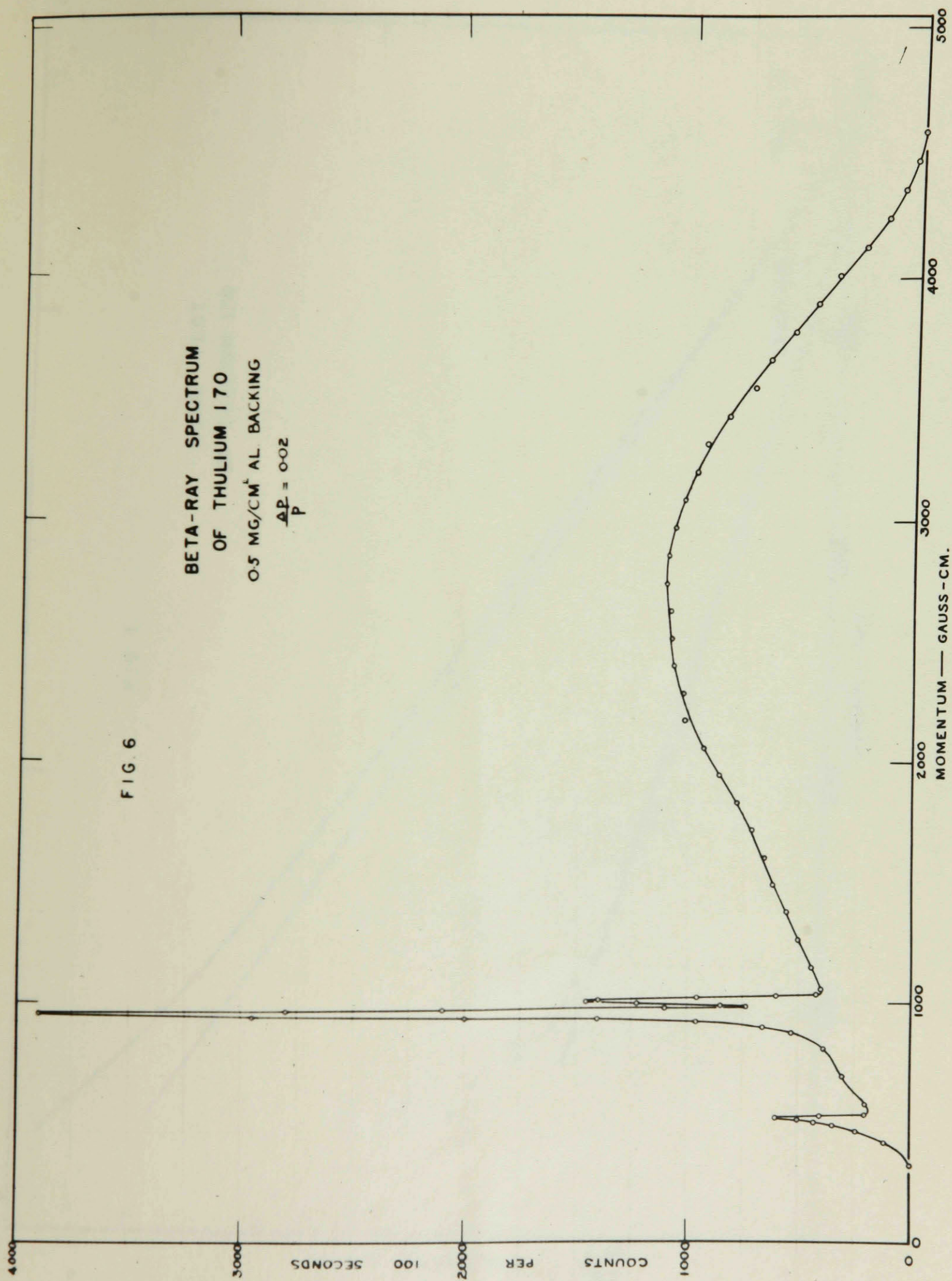


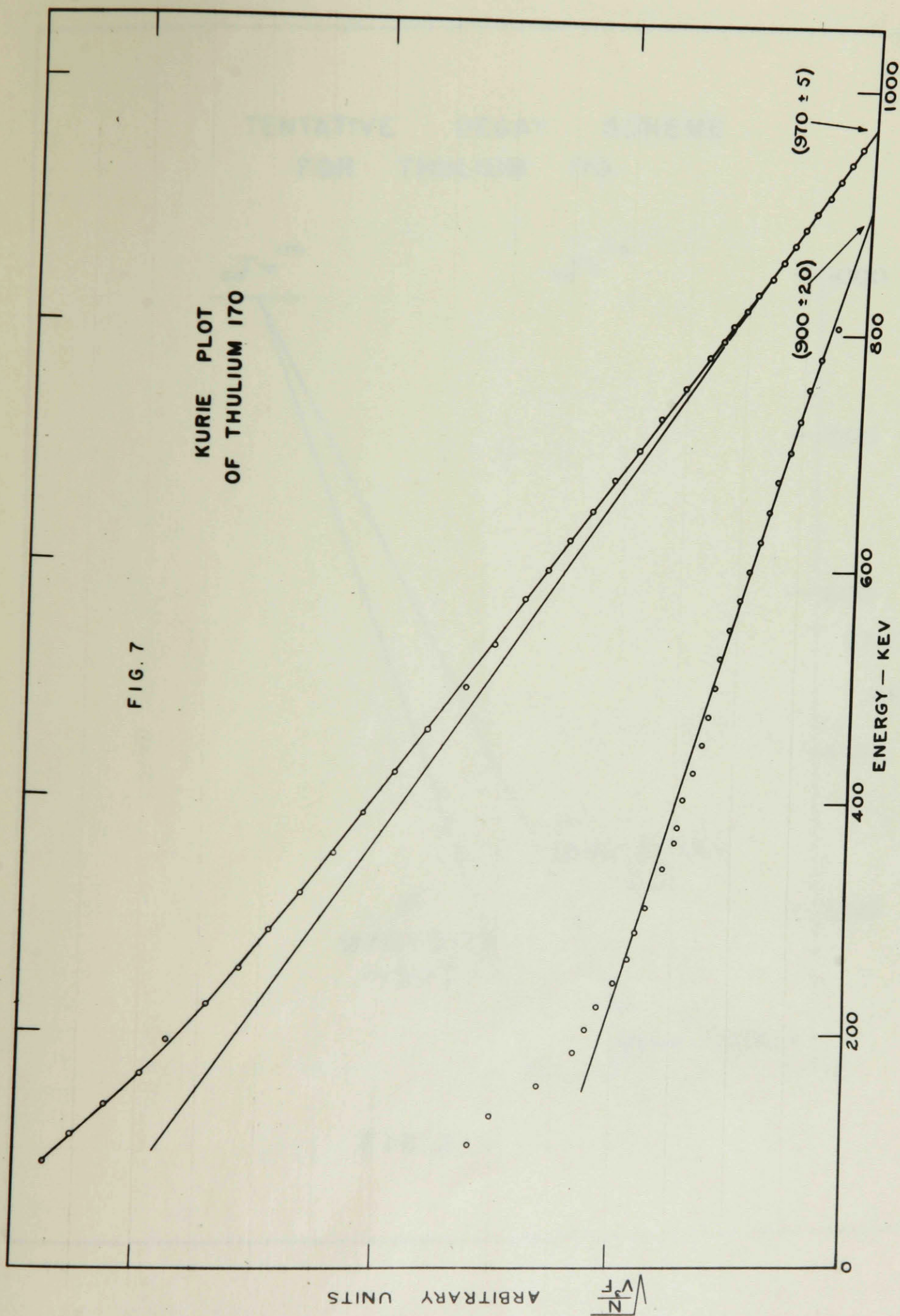
FIG. 2





KURIE PLOT
OF THULIUM 170

FIG. 7



TENTATIVE DECAY SCHEME FOR THULIUM 170

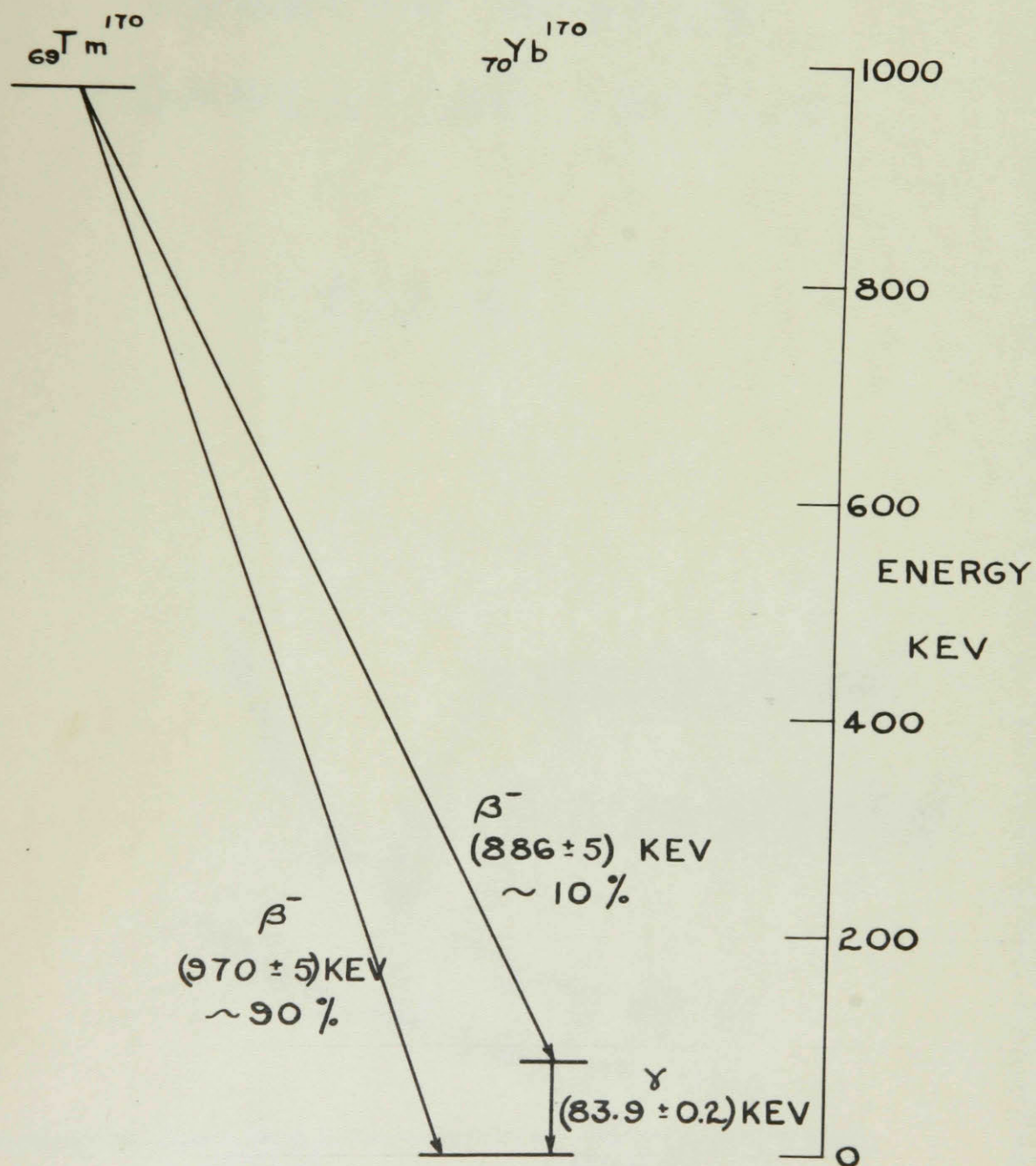
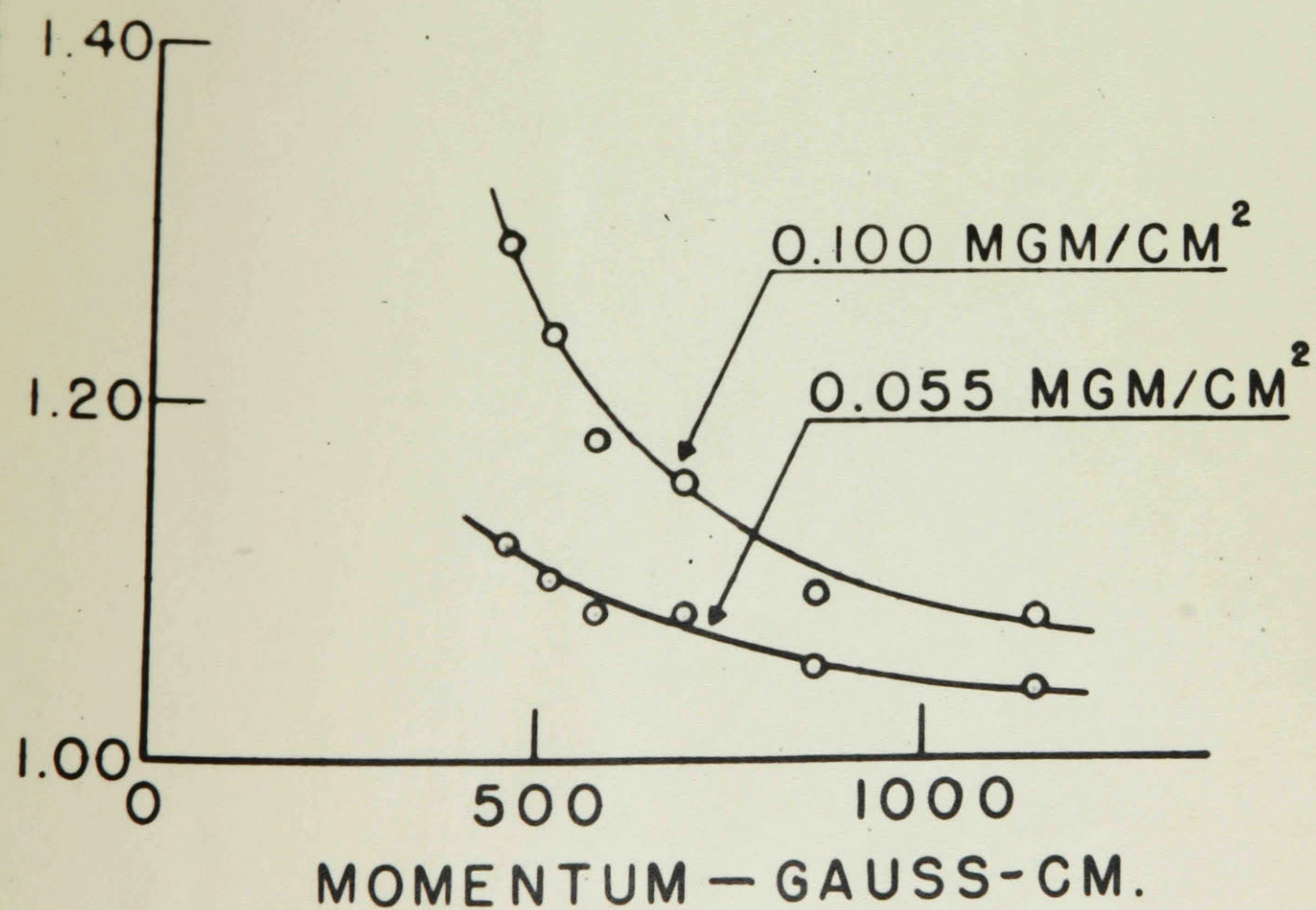


FIG. 8

COUNTER WINDOW CORRECTION FACTOR

FIG. 9



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