A LOW ENERGY BETA-RAY SPECTROMETER AND THE BETA-RAY SPECTRUM OF $EU^{152-154}$

by

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A THESIS SUBMITTED IN PARTIAL FULFILMENT OF
THE REQUIREMENTS FOR THE DEGREE OF
MASTER OF APPLIED SCIENCE
in the Department
of
PHYSICS

We accept this thesis as conforming to the standard required from candidates for the degree of MASTER OF APPLIED SCIENCE

Members of the Department of PHYSICS

THE UNIVERSITY OF BRITISH COLUMBIA

January, 1953

ABSTRACT

Modifications have been made to a semi-circular focussing spectrometer to facilitate its operation and to improve its performance. Geiger counters filled with the saturated vapour of heptane kept in an ice bath are used to detect the beta-particles. The windows of the counters are made of zapon films about 10 micrograms/cm^2 in thickness. The sources are mounted on similar films and have a total thickness less than 100 micrograms/cm^2. The combination of thin source and thin windows permits the measurement of beta-particle energies down to 2 Kev.

An examination of the beta-spectrum of Eu^{152-4} has been carried out. It consists of 7 peaks corresponding to the energies 8.0, 15.0, 26.4, 33.2, 38.3, 73.1 and 74.8 Kev. The two upper peaks are assigned as K conversion lines for gamma-rays of 121.5 and 123.2 Kev. The 33.2 and 38.3 Kev lines are assigned as L and M Auger electrons in Sm. The 8.0 Kev line is assigned as an M Auger electron in Sm. The 15.0 Kev peak is assigned tentatively as an M conversion line corresponding to a gamma-ray of 16 Kev. The 26.4 Kev line is tentatively assigned as either a K conversion line corresponding to a gamma-ray of 73 Kev or an L conversion line corresponding to a gamma-ray of 34 Kev.

ACKNOWLEDGEMENT

The research described in this thesis was made possible by the award of a National Research Council Bursary and a National Research Council Summer Scholarship.

I am deeply indebted to Dr. K. C. Mann for his invaluable advice and assistance and also to Dr. H. Brown for familiarising me with the apparatus and the techniques involved in its operation.

Acknowledgement is made also to Dr. K. Starke for his advice on the preparation of radio-active sources and to Messrs. J. Lees and A. Fraser for their technical assistance.

TABLE OF CONTENTS

TABLE OF ILLUSTRATIONS

I

INTRODUCTION

Particle spectrometers were first introduced in order to resolve the energy distribution of charged particles emitted by naturally radio-active substances. A collimated beam of alpha-particles deflected by an electric or magnetic field was made to fall on a photographic plate or scintillation screen. The energy of these particles could be calculated from a measurement of the field and the deflection of the particle. Baeyer and Hahn[1] built the first spectrograph of this type for the analysis of the energy spectrum of beta-particles from radio-active nuclei. No use was made of any focussing properties, the result being that most of the particles emitted by the source were not permitted to enter the spectrograph, i.e., only particles with the same velocity and the same initial direction would strike the same point on the photographic plate.

To circumvent this defect, focussing spectrometers of many types have been built. A focussing spectrometer is an instrument in which the resolution is not critically dependent on the transmission (percent solid angle subtended by the entrance slit at the source). Danysz[2] pointed out that two equal circles drawn about points separated by a

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1. G. Baeyer and O. Hahn, Phys. Zeit., 11,488(1910).
2. J. Danysz, Comptes Rendus, 153,339,1066(1911).

distance small with respect to the radius would intersect at two points approximately diametrically opposite. Because the path of an electron in a uniform magnetic field is a circle, focussing can be obtained by allowing the electrons to travel through a semi-circle before striking a photographic plate. Fig. 1 shows such an arrangement, in which monoenergetic electrons leaving the source at point S will travel through the baffle and arrive at point P on the photographic plate. These electrons are said to be focussed since they will arrive near the point P so long as they are emitted within the angle A defined by the baffle width. Electrons with higher energies will be focussed to the right of P, those with lower energies to the left. This arrangement, although an improvement, is not perfect since the central ray in Fig. 1 does not strike the photographic plate at exactly the same point as the two outer rays.

The radius of curvature of an electron in a uniform magnetic field is related to the momentum of the electron by the formula

$$H\rho = \frac{mvc}{e} \qquad (1)$$

where H is the magnetic field in gauss, ρ is the radius of curvature in cm., mv is the momentum in c.g.s. units, c is the velocity of light and e is the charge on the electron in e.s.u. The momentum is related to the energy of the particle by the relativistic formula

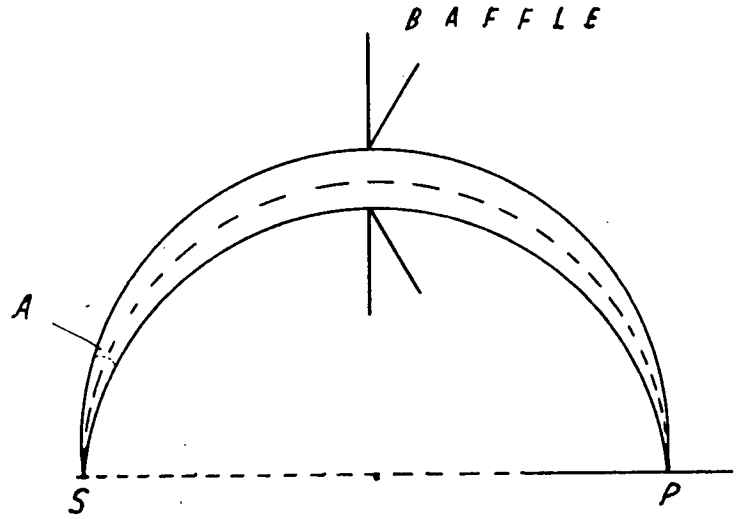

FIGURE 1

SEMICIRCULAR FOCUSSING

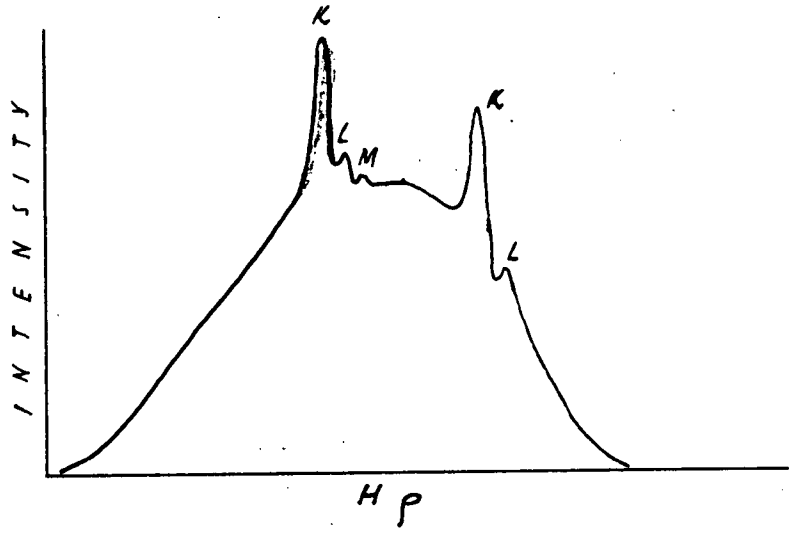

FIGURE 2

ANORMAL BETA SPECTRUM

$$H\rho = 10^4/3 \sqrt{T(T + 1.02)} \qquad (2)$$

where T is the kinetic energy of the electron in Mev.

A spectrograph of the type illustrated in Fig. 1 has one objectionable feature. The photographic plate is not equally sensitive to all energies of electrons. This difficulty may be overcome by placing a Faraday cage or a Geiger counter behind an exit slit as a detector, and varying the magnetic field to select the energy region under investigation.

Using a spectrometer of this type Chadwick[3] showed that in addition to groups of electrons of discrete energies there was also a continuous distribution of electrons, as illustrated in Fig. 2.

As a result of many experiments conducted during the past forty years a comprehensive theory of beta decay has been developed. In addition to nuclear beta emission, atomic beta particles are known to be emitted from atoms with excited nuclei, the latter phenomenom being known as internal conversion. A third process of interest in beta-ray spectroscopy is called orbital electron capture and occurs when a nucleus absorbs one of the orbital electrons of the atom.

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3. J. Chadwick, Vorh.d.D. Phys. Ges., 16,383(1914).

In nuclear beta emission a radio-active nucleus emits an electron thereby decaying to a daughter nucleus with atomic number one greater (negatron emission), or one less than (positron emission or orbital electron capture) the parent. The total energy involved in such a transition is discrete although the emitted electrons have a continuous distribution of energies up to this maximum total energy E_o. Furthermore experiment has shown that the electron does not necessarily leave in the opposite direction to the recoil nucleus. It therefore seems that neither energy nor momentum is conserved in beta decay processes, nor is there conservation of angular momentum. The electron, being a particle of intrinsic spin $\frac{1}{2}\hbar$, must carry away a half-integral multiple of $\hbar$ of total angular momentum since both parent and daughter are isobaric, i.e., either both have integral spins or both have half-integral spins.

A possible solution to the dilemma is provided by the neutrino hypothesis of Pauli. The neutrino hypothesis proposes the existence of a neutral particle (so far unobserved) of small, possibly zero mass and half-integral spin. This particle, called the neutrino, is assumed to be emitted simultaneously with the observed beta-particle. The electron, neutrino and product nucleus share among them the energy, momentum and angular momentum available from the nuclear transition. The beta-particle has its maximum momentum when the neutrino is emitted with zero momentum.

Fermi[4] has developed a mathematical analysis of beta decay based on the neutrino hypothesis which results in a theoretical energy distribution for the emitted beta-particles which is in substantial agreement with experiment. The momentum spectrum of the beta-particles for the so-called "allowed" or most probable transitions is given by

$$N(p)dp = CF(Z,E)p^2(E_o - E)^2\, dp \qquad (3)$$

where $N(p)dp$ is the number of beta-particles emitted with momentum in the range dp at p, C is a constant which in general depends on the specific nucleus involved in the decay, $F(Z,E)$ is a relativistic Coulomb correction factor, E is the energy of the beta-particle, and E_o is the total energy available from the nuclear transition. It follows from (3) that a plot of $(N(p)/Fp^2)^{\frac{1}{2}}$ against energy E is a straight line intersecting the abscissa at $E = E_o$. Such a plot is called a Kurie plot and has the advantage that it may be used to find E_o by a straight line extrapolation.

After emission of a nuclear beta-particle the daughter nucleus is often left in an excited state. The excitation energy may be dissipated either by emission of a gamma-ray or by liberation of an orbital electron. The latter process is termed internal conversion. In order to determine the energy involved in de-excitation by internal conversion it is necessary to add the atomic shell binding energy of the converted electron to its measured kinetic energy. The most probable internal conversion process

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4. E. Fermi, Zeits. fur Phys., 88,166(1934).

involves a K shell electron. The K conversion coefficient is defined as the ratio of the probability of K conversion to gamma emission. In the internal conversion process energy is transmitted from the nucleus to the electron by the electromagnetic interaction of the nucleus and the electron. The main contribution comes from the electrostatic Coulomb interaction. It is possible to make exact calculations of internal conversion coefficients using the relativistic wave functions of the electrons. In general internal conversion coefficients increase as the atomic number and the multipole order of the transition increase and decrease as the energy of the transition increases. The multipole order of a transition is a measure of the vector angular momentum change, i.e., transitions involving a vector angular momentum change of $\hbar$ are termed dipole, those involving a change of $2\hbar$, quadrupole, etc.

Exact calculations of K conversion coefficients have been made by Hulme et al[5], Taylor[6], Fisk[7], Rose et al[8], Griffith[9], and Reitz[10]. Experimental support of these

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5. H. R. Hulme, N. F. Mott, F. Oppenheimer, and H. M. Taylor, Proc. Roy. Soc. (London), A155,315(1936).

6. H. M. Taylor, N. F. Mott, Proc. Roy. Soc. (London), A142,215(1933).

7. J. B. Fisk, H. M. Taylor, Proc. Roy. Soc. (London), A146,178(1934).

8. M. E. Rose et al, G. Goertzel, B. I. Spinrad, J. Harr, and P. Strong, Phys. Rev., 76,1883(1949).

9. B. A. Griffith, J. P. Stanley, Phys. Rev., 75,534(1949).

10. J. R. Reitz, Phys. Rev., 77,10(1950).

calculations has been obtained by Waggoner[11], Petch[12], and others. By measuring experimentally the K conversion coefficient and referring to the conversion coefficient tables mentioned above, it is often possible to infer the multipole order and thus the vector nuclear spin change involved in the transition.

The theory absolutely forbids quantum transitions of multipole order zero. In this case the nucleus can de-excite only by internal conversion, i.e., the internal conversion coefficients are all infinite.

It is interesting to speculate on the possible effect that orbital electron capture might have on the mean life of subsequent internal conversion transitions. The de-excitation of a nucleus by internal conversion is independent of the competing gamma-decay process. We may write

$$\frac{1}{\tau} = \frac{1}{\tau_g} + \frac{1}{\tau_K} + \frac{1}{\tau_L} + \frac{1}{\tau_M} + \cdots$$

where τ is the mean life of the excited state of the nucleus, τ_g is the mean life for decay by photon emission, τ_K is the mean life for decay by internal conversion in the K shell, and τ_L, τ_M, etc. are respectively the mean lives for decay by internal conversion in the L, M, etc. shell. It is assumed that the probability for emission of an internal

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11. M. A. Waggoner, M. L. Moon, and A. Roberts, Phys. Rev., 80,420(1950).

12. H. E. Petch, M. W. Johns, Phys. Rev.,80,478(1950).

conversion electron by a particular excited nucleus is directly proportional to the number of electrons in the atomic shell. This may be written

$$\lambda_{2K} = 2\lambda_{1K}$$

where λ_{2K} is the probability per unit time of emission of a K conversion electron from an atom with a full K shell, and λ_{1K} is the probability per unit time of emission of a K conversion electron from an atom with only one K electron. Consider N nuclei at time t = 0 which have just undergone a K capture transition to an excited state. If λ_x is the probability per unit time of filling a vacancy in the K shell by an x-ray transition, we may write the probability of there being one electron in the K shell at time t,

$$P_{1K} = e^{-\lambda_x t}$$

and the probability for two K electrons,

$$P_{2K} = 1 - e^{-\lambda_x t}$$

The probability per unit time of emission of a K conversion electron at time t is then

$$\lambda_K(t) = \lambda_{2K} P_{2K} + \lambda_{1K} P_{1K}$$

$$= \lambda_{2K}\left(1 - \tfrac{1}{2} e^{-\lambda_x t}\right)$$

The rate at which nuclei of this species decay by K

conversion is then given by

$$\alpha N = \lambda_K (t) N$$

which on integration yields

$$\ln \frac{N}{N_0} = - \lambda_{2K} \left[t + \frac{1}{2\lambda_x} \left(e^{-\lambda_x t} - 1 \right) \right]$$

When ln $N/N_0 = -1$, only N_0/e of the original excited nuclei remain. We define the corresponding time τ_K as the mean life for K conversion. Solving the previous equation yields

$$1 = \lambda_{2K} \left[\tau_K + \frac{1}{2\lambda_x} \left(e^{-\lambda_x \tau_K} - 1 \right) \right]$$

By substituting $\tau_x = \frac{1}{\lambda_x}$ and rearranging we get

$$\tau_K = \tau_{2K} + \frac{\tau_x}{2} \left(1 - e^{-\frac{\tau_x}{\tau_K}} \right)$$

which may be solved approximately in three cases:

(1) $\frac{\tau_K}{\tau_x} \ll 1$

$$\therefore \tau_K = \tau_{2K} + \frac{\tau_K}{2} = 2\tau_{2K}$$

(2) $\frac{\tau_K}{\tau_x} \approx 1$

$$\therefore \tau_K = \tau_{2K} + \frac{\tau_K}{2} \left(1 - \frac{1}{e} \right) \approx \frac{3}{2} \tau_{2K}$$

(3) $\frac{\tau_K}{\tau_x} = A > 2$

$$\therefore \tau_K = \tau_{2K} + \frac{\tau_K}{2A} = \left(\frac{2A}{2A-1} \right) \tau_{2K}$$

Since the effect of a single missing electron in the L shell is relatively small, the effect of K capture on the mean life of the subsequent nuclear state, if appreciable, should be most easily detected by measurement of K/L ratios. We may thus say with assurance that, so long as the mean life for x-ray transitions to the K shell is greater than the mean life for normal K conversion the K/L ratio for a nuclear process following K capture will be smaller than that for the same process following beta emission. If the mean life for the x-ray processes which fill the K shell is shorter than the normal K conversion lifetime, and if this in turn is shorter than the mean life for x-ray processes that fill the L shell, then it is possible that the K/L ratio may become larger than that which would normally be expected. Similar effects to those noted above might be found in cascade decay schemes where previous transitions involving internal conversion have robbed the atom of some of its orbital electrons.

Beta-ray spectroscopy has provided a method for determining the energies of particles and photons emitted by radio-active nuclei and has thus increased the knowledge of nuclear energy levels. Most spectrometers have been designed to cover the energy range from 0.1 to 3 Mev, the lower limit being an instrumental one, caused by excessive scattering of low energy electrons from baffles and residual gas molecules, by the increasingly serious defocussing effect due to the uncompensated earth's magnetic field, and by

absorption of the low energy particles both in the source and in the counter window. The upper limit is set by the power demands of the focussing magnet. A few spectrometers have been built to measure energies greater than 10 Mev and some to extend the lower limit to 10 Kev or less.

There are important reasons for the investigation of the low energy region. Cook and Langer[13] and other experimenters[14,15,16] report that in a number of cases the Kurie plot deviates from a straight line at low energies. More recent work[17,18,19] indicates that these results were entirely due to self-absorption in the source. The presence of a low energy beta group would cause an apparent curvature in the Kurie plot at low energies. The presence of Auger and internal conversion electrons can so complicate the low energy beta spectrum as to make it difficult to determine the shape of a low energy beta group. Low energy internal conversion lines are very common. Lines at less than 50 Kev have been found in the beta spectrum of almost all naturally

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13. C. S. Cook and L. M. Langer, Phys. Rev., 73,601(1948).
14. A. W. Tyler, Phys. Rev., 56,125(1939).
15. J. L. Lawson, Phys. Rev., 56,131(1939).
16. A. A. Townsend, Proc. Roy. Soc., A1777,357(1941).
17. R. D. Albert and C. S. Wu, Phys. Rev., 74,847(1948). 75,1107(1949).
18. L. Feldman and C. S. Wu, Phys. Rev., 76,697(1949).
19. L. M. Langer, J. W. Motz and H. C. Price Jr., Phys. Rev., 77,798(1950).

radio-active substances[20] and in some artificially prepared isotopes[21]. In substances of low Z the flourescence yield is found to be small[22], that is to say, an atom with a missing K electron is more likely to de-excite by emission of an Auger electron than by emission of an x-ray. If a nucleus is in the ground state on capturing a K electron the soft x-rays and the Auger electrons offer the only means of detecting the transition. Many postulated decay schemes based on the measurement of high-energy gamma-rays and conversion electrons have energy levels within 50 Kev of each other. Most spectrometers now in use cannot detect transitions between such levels. An instrument which could do so would be a valuable tool in checking the proposed spin and parity values assigned to these states on the evidence of the high energy transitions alone.

This research describes the redesign and use of a spectrometer for the measurement of beta particles in the energy range 1 to 100 Kev. For the reasons previously stated it is essential to investigate this energy region for a complete understanding of the mechanism of nuclear processes.

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20. Radiations from Radio-active Substances. Rutherford, Chadwick and Ellis, pp.360-380.

21. R. D. Hill, Phys. Rev., 74,78(1948).

22. X-rays in Theory and Experiment. Compton and Allison, pp.477-492.

II

THE SPECTROMETER

A. Design considerations.

(1) Detection of beta-particles.

Beta-particles may be detected with photographic plates, and in spite of the objection to a spectrograph, plates would be used if they were sensitive to electrons of low energy. Unfortunately the sensitivity of even the best plates falls off very rapidly as the energy of the impinging electrons is decreased[23]. The sensitivity of the scintillation crystal and photomultiplier combination falls off greatly at low energies. By cooling the photomultiplier in liquid air it is possible to detect electrons with energies as low as 2 Kev, but the operation of a photomultiplier is seriously effected by the focussing magnetic field. Electron multipliers can be made with high efficiency for beta energies between 0.1 and 5 Kev. One of the better detection methods involves post-acceleration of the electrons after they have passed through the exit slit of the spectrometer. This method shows every sign of more frequent use in low-energy beta-ray spectrometry. In a spectrometer of the type described in this thesis post acceleration would be

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23. L. Cranberg and J. Halpern, R.S.I., 20,641(1949).

impracticable. For our purpose a thin window Geiger counter seems best fitted to the task. Geiger counters are known to be sensitive to electrons of almost negligible energy and therefore the problem resolves itself into one of finding some method of introducing the electrons into the sensitive volume of the counter. If a window is to be used it must be sufficiently thin to transmit electrons of low energy and yet strong enough to withstand the pressure of the filling gas.

Backus[24] has described a method for making films as thin as a few micrograms/cm^2. Windowless counters have been used with success by Langer, Motz and Price[19] with an equivalent window thickness due to diffusing gas of about one microgram/cm^2.

The counters used in the present equipment were built by Brown[25] and utilize zapon film of 3 to 5 micrograms/cm^2 covering an entrance slit 0.025 cm. wide. The combination of the narrow window and the low gas pressure used in the counter (1.1 cm. Hg.) makes the use of such films entirely satisfactory.

(2) The Source.

The preparation of low energy beta-sources is an extremely delicate operation since the optimum thickness of

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24. J. Backus, Phys. Rev., 68,59(1945).

25. H. Brown, Ph.D. Thesis, University of British Columbia, (1951).

source plus backing is vanishingly small. The thickness of the source can have a great influence on the shape of the low energy beta spectrum[19]. Because of the fragility of the source its area should preferably be small. A small source poses an additional problem: since the resolution of the spectrometer is in general inversely proportional to the luminosity (transmission times source area), it can be seen that once the resolution has been specified, the overall counting rate of the spectrometer is then dependent entirely on the total source strength. It is important that the overall counting rate be reasonably high since only in this way can good statistics be established quickly. A practical lower limit in counting rate is set by the cosmic-ray background. Having specified the resolution and thus the luminosity and having set a lower limit on the useful counting rate, we may estimate the activity per unit area of the source necessary to produce this counting rate. If we now demand a source thickness which will not too badly distort the low energy end of the spectrum, we may then compute the minimum acceptable specific activity of the source material.

(3) Limitations due to available material and techniques.

The thinnest sources so far produced seriously broaden spectral lines below 10 Kev. Because of this fact there is little point in designing a low energy spectrometer with a resolving power better than one percent. The spectrometer should be designed to utilize source material of that specific activity which may readily be produced. The specific activity and the size of the source are the factors

which determine the minimum overall size of the spectrometer.

The magnetic field of the spectrometer should be large compared to the earth's field and the fluctuating magnetic fields of nearby electrical equipment. Large fields produce short electron trajectories at low energies and since other considerations have placed a lower limit on the size of the spectrometer, it may therefore be necessary to effect a compromise.

B. The Spectrometer Chamber.

A semi-circular focussing type of instrument which best fulfils the above conditions was designed and built by Brown[25]. The internal construction of this spectrometer is illustrated in Fig. 3.

The radius of the curvature of this instrument was kept small in order to reduce the path length of the detected electrons and thus keep to a minimum scattering both from residual gas molecules and from the walls and baffles. In order to have a large transmission this spectrometer was designed to accept electrons from the source in four different directions. Geiger counters with zapon windows are used for detection, the pressure being kept constant by a dynamic filling system. (See Appendix III)

The source material is deposited in a line about 0.1 cm. wide and 1.5 cm. long on a thin zapon film which is supported on a lucite holder. The holder is so mounted that the source is coincident with the axis of the spectrometer. In Brown's arrangement, four sets of baffles were

placed so that electrons leaving the source at angles of $45^{\circ} \pm 5.7^{\circ}$ to the plane of the backing would travel a circular path of $3.05 \pm .03$ cm. radius to strike the window of one of four symmetrically placed Geiger counters. The baffles were cut on a lathe to the required radius and grooves cut in the surfaces to reduce the reflection of electrons into the counters.

Each counter was made by drilling a 0.625 inch diameter hole through a block of brass 1.5 inches long. The brass was then trimmed down with a shaper, particular care being taken with the face containing the window. This face was cut as smooth as possible to leave a thickness of 0.030 inches in the center. Through the narrowest part a longitudinal slot about 5/8 inches long and 0.010 inches wide was cut. The inside of the counter was then polished thoroughly with emery paper and crocus cloth. Another hole of about the same size was drilled through the brass, parallel to the first, for the purpose of filling the counter, so that about 0.050 inches of metal separated the two. Small holes were drilled through this separating wall to allow the filling gas to enter the counter proper. The auxiliary hole was then plugged at one end with a brass plate and at the other with a copper tube to allow connection to the filling system.

The entire counter was then immersed in boiling nitric acid (0.1 Normal) for a few minutes until the surfaces appeared clean. It was then washed, first in distilled water, and then in absolute alcohol and dried. One end was

closed with a Kovar seal and the other with a pyrex cap sealed with deKhotinsky wax. A 0.005 inch tungsten wire was used for the anode.

The spectrometer chamber is a brass cylinder of 6.5 inches inside diameter. The end plates are of 5/16 inch brass. Soft rubber rings are used as vacuum seals. In the bottom plate holes are drilled for the admission of the counter filling gas for the admission of the high voltage lead for connection to the anode of the counters, and for connection to the vacuum system.

The internal assembly, with the exception of the counters, is mounted on a 1/8 inch brass base plate one inch above the lower end plate of the spectrometer chamber. The base plate was highly polished and the position of the source, the positions of the baffles and the location of the entrance slits of the counters were marked on it. The counters were aligned with the baffle system by observing that the counter window, its reflected image, and the mark on the base plate, were in one straight line. The counters are connected to a four arm glass tee by lengths of Tygon tubing to allow movement during alignment.

All exposed metal surfaces are coated with a material of low atomic number in order to reduce scattering as much as possible. This was done by dissolving vacuum wax in carbon tetrachloride and applying several coats of the paint.

In the course of the present work three modifications have been made to the inner spectrometer, two of which had been suggested by Brown[25].

The baffles have been redesigned to permit attachment of new windows without removal of the baffles and source from the instrument. The brass baffle system was completely removed and replaced by four simple lucite baffles made as illustrated in Fig. 3. These baffles were mounted on the base plate midway between the source and the counters, leaving the face of the counters freely accessible for attachment of windows.

In order to reduce the background counting rate due to gamma-rays emitted by the source lead shields were installed on the inner side of each counter. The thickness of these shields was dictated by the available space between the counters and the electron trajectories. (See Fig. 3)

Because the counter anodes had been connected in parallel inside the spectrometer chamber the counters could not easily be tested individually, nor could faulty ones be disconnected without terminating an experimental run. This fault was rectified by introducing four separate leads into the spectrometer chamber through a rubber vacuum seal.

C. The Magnetic Field.

Production of a magnetic field without the use of iron eliminates the need for measuring small fields since the field must vary linearly with the current in the magnet

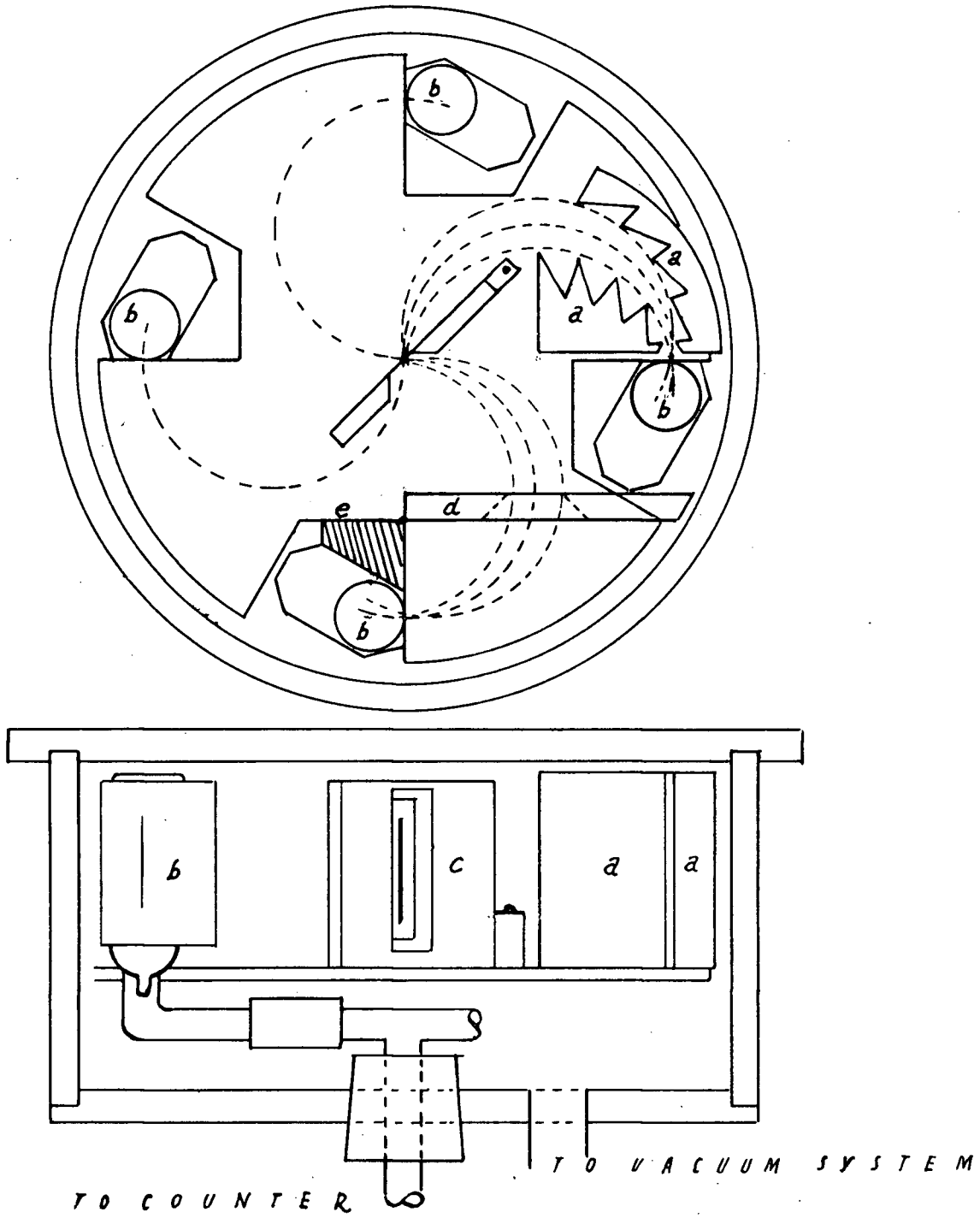

FIGURE 3

INTERNAL CONSTRUCTION OF SPECTROMETER

a BAFFLES (ORIGINAL)

b COUNTERS

c SOURCE HOLDER

d BAFFLE (RE-DESIGNED)

e LEAD SHIELD

coil. The magnetic field required by this spectrometer should be uniform over a cylindrical region, 12 cm. in diameter and about 2 cm. deep, with the magnetic vector parallel to the axis of the cylinder. A set of coils has been designed and built by Brown[25] as illustrated in Fig. 4 in order to produce such a field. The coil system consists of three coplanar coils, one of 1400 turns of radius 17.5 cm., a second of 270 turns of radius 11 cm., and a third of 12 turns of radius 8.5 cm. Passing 10 amperes of current through these coils, with the current in the second coil flowing in the opposite sense to the current in the other two coils, was found to produce a magnet field of approximately 360 gauss with a maximum inhomogeneity of less than 1% over the desired region.

The uniformity of the magnetic field was tested by means of two inverse connected matched search coils and a ballistic galvanometer. The results of this test are shown in Fig. 5. The inner coil has been eliminated because this test showed the field to be more uniform without it.

D. Mathematical Treatment of a Spectrometer.

Geoffrion[26] has deduced mathematically the characteristics of a semi-circular focussing spectrometer. In the spectrometer shown in Fig. 6 electrons of a given momentum will form an image of the source at the exit slit.

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26. C. Geoffrion, R.S.I., 20,638(1949).

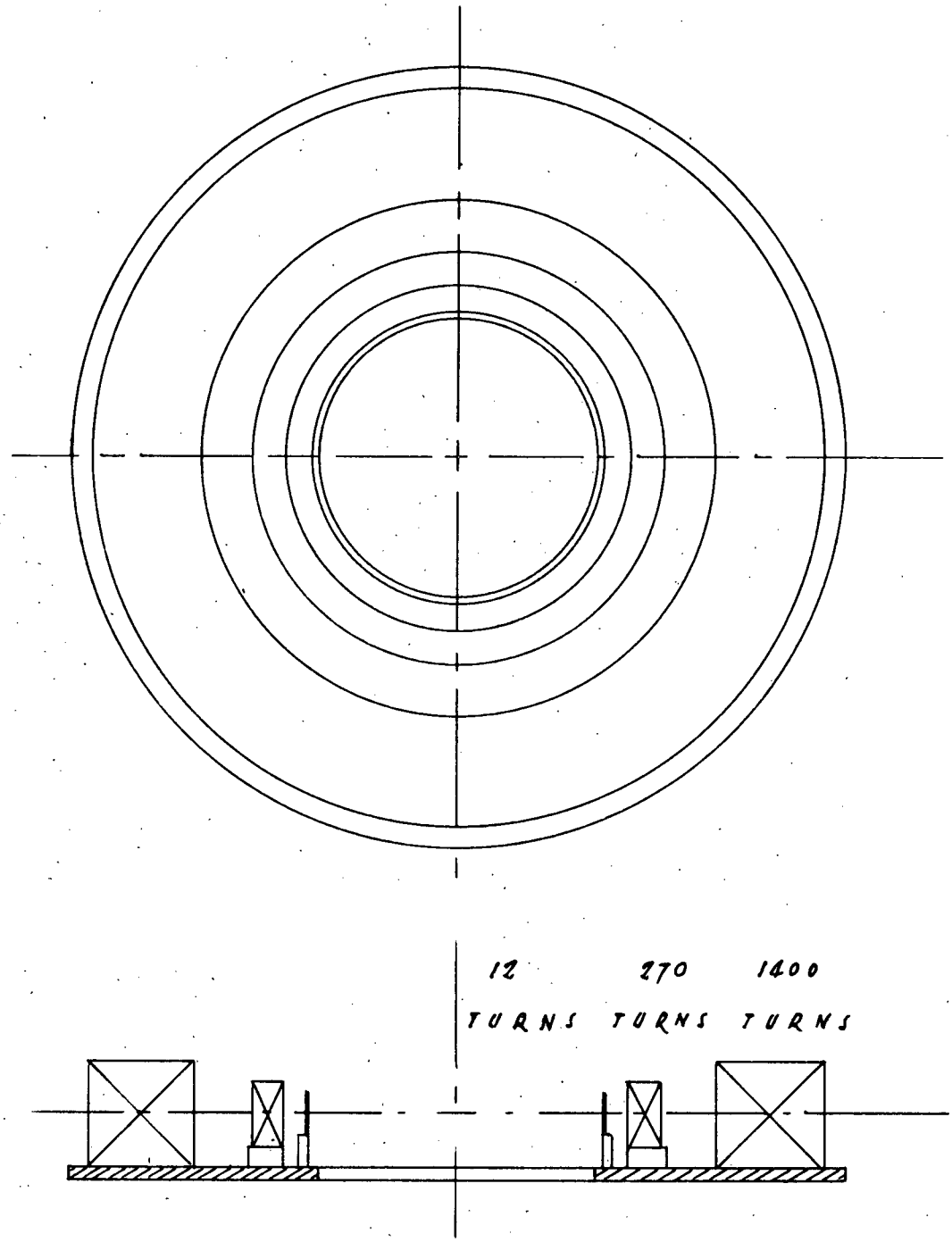

FIGURE 4

CONSTRUCTION OF THE MAGNET

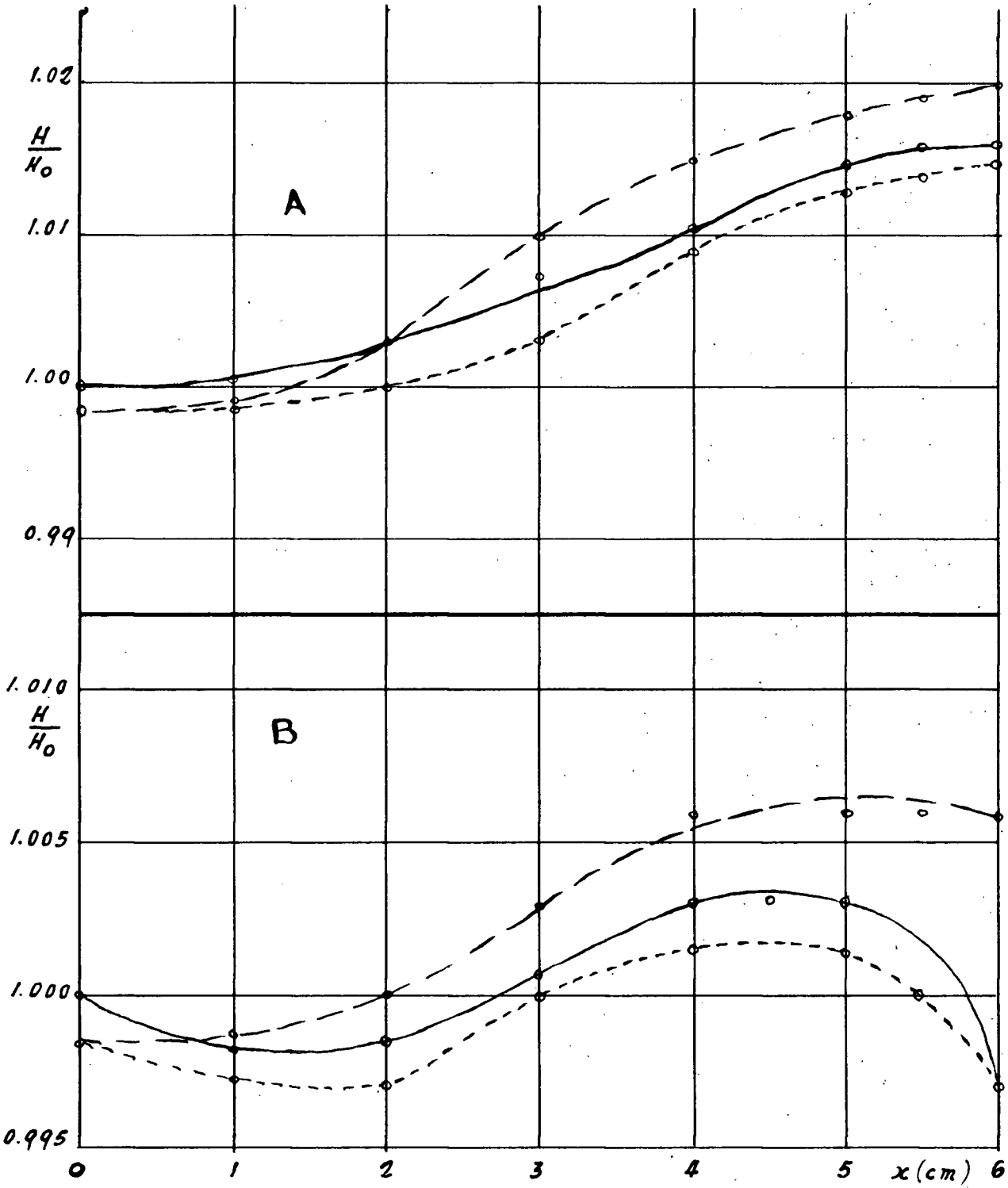

FIGURE 5

OBSERVED VARIATION OF MAGNETIC FIELD IN SPECTROMETER REGION

A INNER COIL CONNECTED

B INNER COIL DISCONNECTED

The width of this image is given by

$$Q^1 = Q + 2r(1 - \cos A_o \cos B_o) \qquad (1)$$

where Q^1 is the width of the image, Q is the width of the source, r is the radius of the electron trojectory, A_o is one-half the angle subtended by the baffles at the source, and B_o is the maximum angle with the median plane of the spectrometer at which an electron may enter the exit slit.

The right hand edge of this image is located a distance 2r from the right hand edge of the source. Since r depends on the momentum of the elctrons in a constant field H, a number of images will be formed with different values of r for different values of the momenta.

If a detector is placed behind the exit slit it will receive electrons with a certain range of values of momenta. The width of the exit slit adds to the effective width of the image of the source so that it now becomes equal to

$$F + Q + 2r(1 - \text{Cos } A_o \text{ Cos } B_o) \qquad (2)$$

where F is the width of the exit slit and (2) is the uncertainty in the measurement of 2r for a constant field H.

The limit of resolution of a spectrometer is defined as the ratio $\Delta p/p$ where Δp is the range of the momenta of those electrons passing through the exit slit.

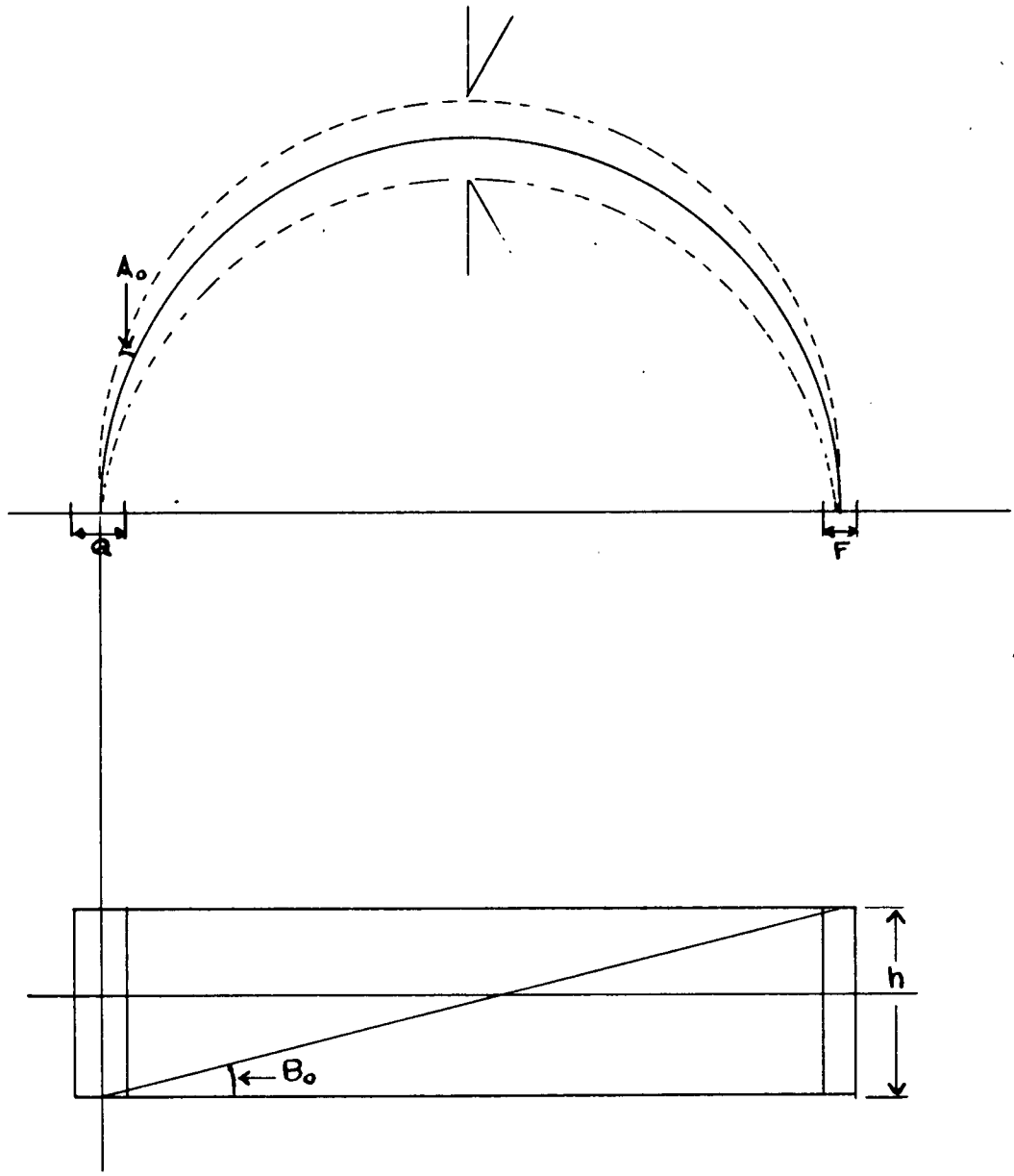

FIGURE 6

DIAGRAM OF ELECTRON PATHS

IN THE SPECTROMETER

Since

$$p = cHr/e \tag{3}$$

$$\Delta p = (H\Delta r + r\Delta H)c/e \tag{4}$$

and since H is constant

$$\Delta p = cH\Delta r/e$$

$$= cH/2e(F + Q + 2r(1 - \text{Cos } A_o \text{ Cos } B_o)) \tag{5}$$

and therefore

$$\frac{\Delta p}{p} = \frac{F + Q + 2r(1 - \text{Cos } A_o \text{ Cos } B_o)}{2r} \tag{6}$$

and since A_o and B_o are small this can be simplified to

$$\frac{\Delta p}{p} = \frac{F + Q}{2r} + \frac{A_o^2 + B_o^2}{2} \tag{7}$$

The spectrometer described in this report has a trajectory radius of curvature of 3.05 cm. and A_o was chosen as 0.1. The minimum value of Q obtainable is about 0.07 cm. since this is the projection of a 0.10 cm. wide line at 45° to the plane of the source backing. The width of the exit slit (width of the counter window) is about 0.025 cm. Substituting the actual values of Q, F, A_o and B_o, the above formula yields a resolution of about 1.5%.

The luminosity of a spectrometer is defined as the product of the transmission times the source area. For this instrument the luminosity is 0.58×10^{-3} cm^2, which is larger than most comparable spectrometers.

Geoffrion[26] shows a curve of the intensity to be expected under optimum conditions using an exit slit of infinitesimal width. The theoretical line profile for an exit slit of a given width F can be obtained by partial integration of Geoffrion's curve for F = 0.025 cm. and F = 0.06 cm. These profiles are shown in Fig. 7. It is evident from these curves that the resolution for the wider exit slit is appreciably the same as for the narrower one (the one we are using). Attempts to use the wider exit slit with its resultant increase in transmission were unsuccessful because the wider zapon windows were unable to withstand the counter filling pressure.

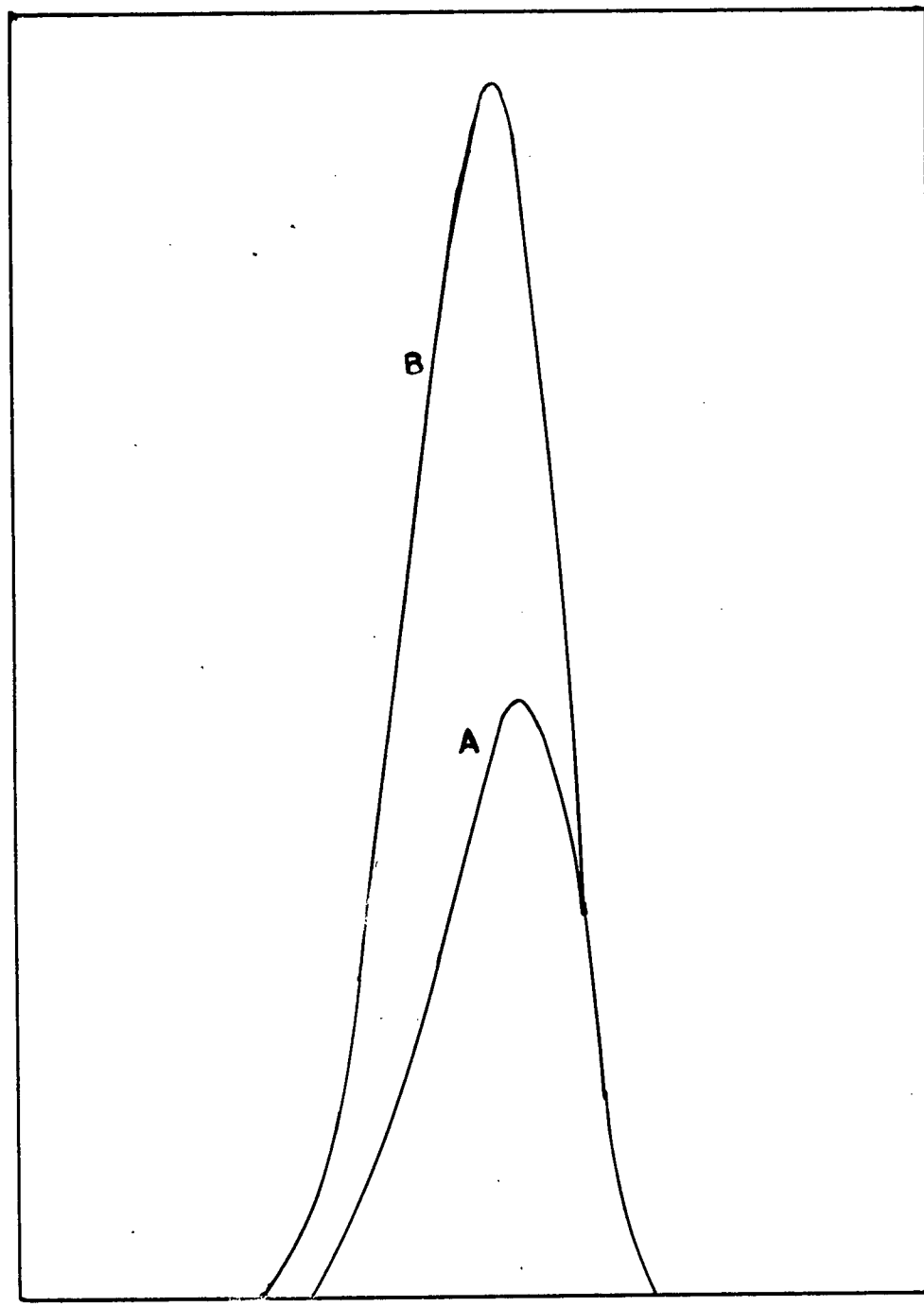

FIGURE 7

THEORETICAL LINE PROFILES

A EXIT SLIT 0.025 CM. WIDE

B EXIT SLIT 0.06 CM. WIDE

III

RESULTS

A. Preliminary Considerations.

Before attempting the analysis of the low energy beta spectrum of Eu^{152-4} it is necessary to clarify two important points. Firstly, some attempt should be made to estimate the effect of source absorption on the broadening and shifting of spectral peaks, and secondly, some proof should be shown for the validity of the spectrometer calibration. Although it is difficult, if not impossible, to determine the effect of source absorption from a purely theoretical argument, nevertheless it is possible to estimate these effects in an empirical way. White and Millington[27] have derived from experimental results the momentum distribution of an initially monochromatic beam of electrons after passing through thin foils. After taking into account the finite resolving power of their equipment they plot the natural line profiles for electrons after being straggled by various thicknesses of foil. From these results they are able to plot a fundamental line profile curve from which all other straggling curves may be obtained from a simple relationship. These calculations show that the displacement in momentum of the maximum of any straggled curve from the true value of the momentum of the incident

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27. F. White and G. Millington, Proc. Roy. Soc., A120,701(1928).

monochromatic electrons is a constant multiple (1.3) of the half-width of the straggled curve, where the half-width is defined as the percentage width of the peak at half-intensity on a momentum scale. Although electrons emitted by a thin radio-active source do not all travel through the same thickness of straggling material, the relationship regarding peak displacement may be used since it is independent of the thickness of the straggling material. By taking into account the natural line width due to the finite resolving power of the spectrometer employed in this research we obtain the corrected value of the momentum corresponding to a spectral peak:

$$H\rho = (H\rho)_0(1 + 0.013(P - 1.5))$$

where H is the corrected value of the momentum, $(H\rho)_0$ is the momentum value corresponding to the maximum of the experimental peak, and P is the percentage half-width of the experimental curve, as defined above.

The original calibration of the spectrometer made by Brown is considered by this writer to be somewhat in error. Brown's determination of the energy of the dominant internal conversion line in Ra D (600 gauss-cm), although in substantial agreement with the work of Rutherford, Chadwick and Ellis[28], Richardson

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28. Sir E. Rutherford, J. Chadwick and C. D. Ellis, Radiations from Radio-active Substances, 1930.

and Leigh-Smith[29], Tsien San-Tsiang[30] and others, does not take into account the peak shift phenomenum discussed above. The calibration used in the present work has been established by accepting the experimental work of Brown reinterpreted in the light of the previous paragraph, together with the well substantiated value for the dominant L conversion line in Ra D. This calibration in terms of the control potentiometer setting is 980 ± 15 gauss-cm. per volt. Using this calibration, together with the method of analysis described above, none of the peaks in Ra D reported by Brown is appreciably shifted from the values he has assigned to them.

B. Europium$^{152-154}$

When the stable isotopes of Eu (mass numbers 151 and 153) are irradiated by slow neutrons, active isotopes of atomic weights 152 and 154 are formed by (n,γ) processes. A mass spectrographic analysis by Ingham and Hayden[31] showed that activities could be assigned to these isotopes as follows:

TABLE I

Atomic wt.	Half-life
152	9.2 h
152	5-8 y
154	5-8 y

29. H. O. W. Richardson and A. Leigh-Smith, Proc. Roy. Soc., 160,454(1937)

30. Tsien San-Tsiang, Phys. Rev., 69,38(1946).

Cork, Shreffler and Fowler[32] studied the long-lived activities in a photographic 180° spectrometer, and reported internal conversion lines corresponding to transitions of 122, 247, 286, 343 and 400 Kev, as well as a continuous beta spectrum of energy 0.93 Mev.

Wiedenbeck and Chu[33], using a coincidence counting technique, reported the continuous beta spectrum of long-lived Eu to be complex with upper limits 0.62 and 1.0 Mev.

Shull[34], using a magnetic double-focussing beta-ray spectrometer, reported internal conversion lines corresponding to transitions of 123, 124, 247, 286, 344 and 412 Kev, as well as additional external conversion lines corresponding to transitions of 442, 772, 959, 1082 and 1402 Kev.

Cork et al[35] summarize their investigation of Eu by proposing the decay schemes illustrated in Fig. 8. The same workers report Auger lines at 32.0 and 37.9 Kev corresponding to an atomic transition in Sm of 39.1 Kev.

For the present investigation the source was made from europium oxide (see Appendix II), the active material having been left standing several months to

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32. J. M. Cork, R. G. Shreffler and G. M. Fowler, Phys. Rev., 72, 1209(1947); 73,78L(1948).

33. M. L. Wiedenbeck and K. Y. Chu, Phys. Rev., 72, 1164(1947).

34. F. R. Shull, Phys. Rev., 74,917(1948).

35. J. M. Cork et al, Phys. Rev., 77,848(1950).

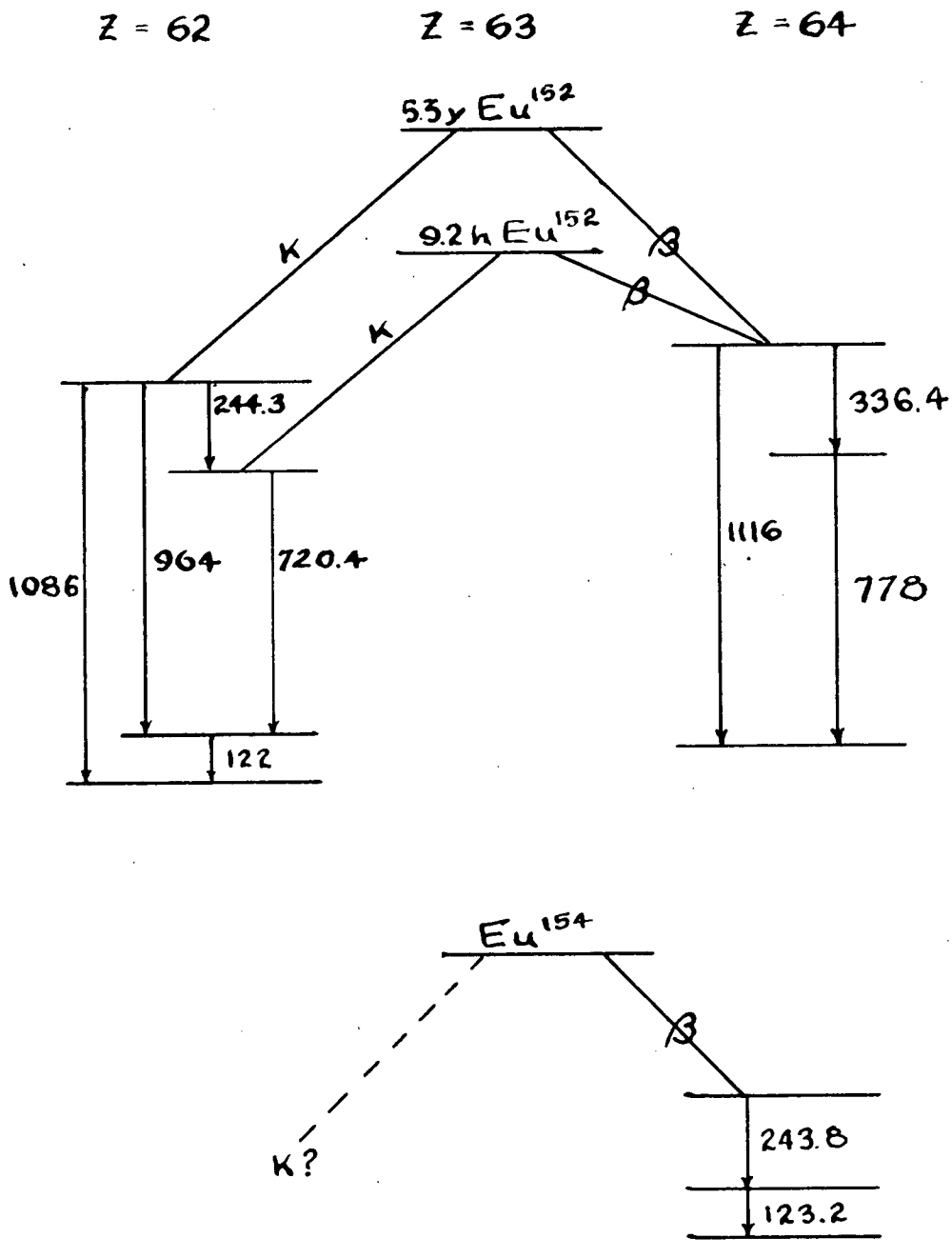

FIGURE 8

PROPOSED DECAY SCHEMES FOR Eu^{152-4}

eliminate the short-lived isomer of Eu^{152}.

Table II shows the binding energies of the orbital electrons in Sm and Gd.

TABLE II

Shell	62 Sm Kev	64 Gd Kev
K	46.7	50.1
L	7.3	7.9
M	1.4	1.6

The four highest energy peaks of the spectrum (Fig. 9) correspond to the four lowest energy peaks reported by Cork et al[35] (Table IV). The seven peaks found in the present investigation have tentatively been assigned as follows:

TABLE III

Peak No.	Electron energy Kev	Interpretation	Energy Sum Z = 62 Kev	Energy Sum Z = 64 Kev
1	8.0 ± 1.0	M(Auger(62)	9.4 ± 1.0	
2	15.0 ± 1.0	M(62)(64)	16.4 ± 1.0	16.6 ± 1.0
3	26.4 ± 1.2	K(62)(64) L(62)(64)	73.1 ± 1.2 33.7 ± 1.2	76.5 ± 1.2 34.3 ± 1.2
4	33.2 ± 1.4	L(Auger)(62)	40.5 ± 1.4	
5	38.3 ± 1.5	M(Auger)(62)	39.7 ± 1.5	
6	73.1 ± 2.2	K(64)		123.2 ± 2.2
7	74.8 ± 2.2	K(62)	121.5 ± 2.2	

(Third column numbers in brackets refer to mass numbers of assigned daughter nucleus).

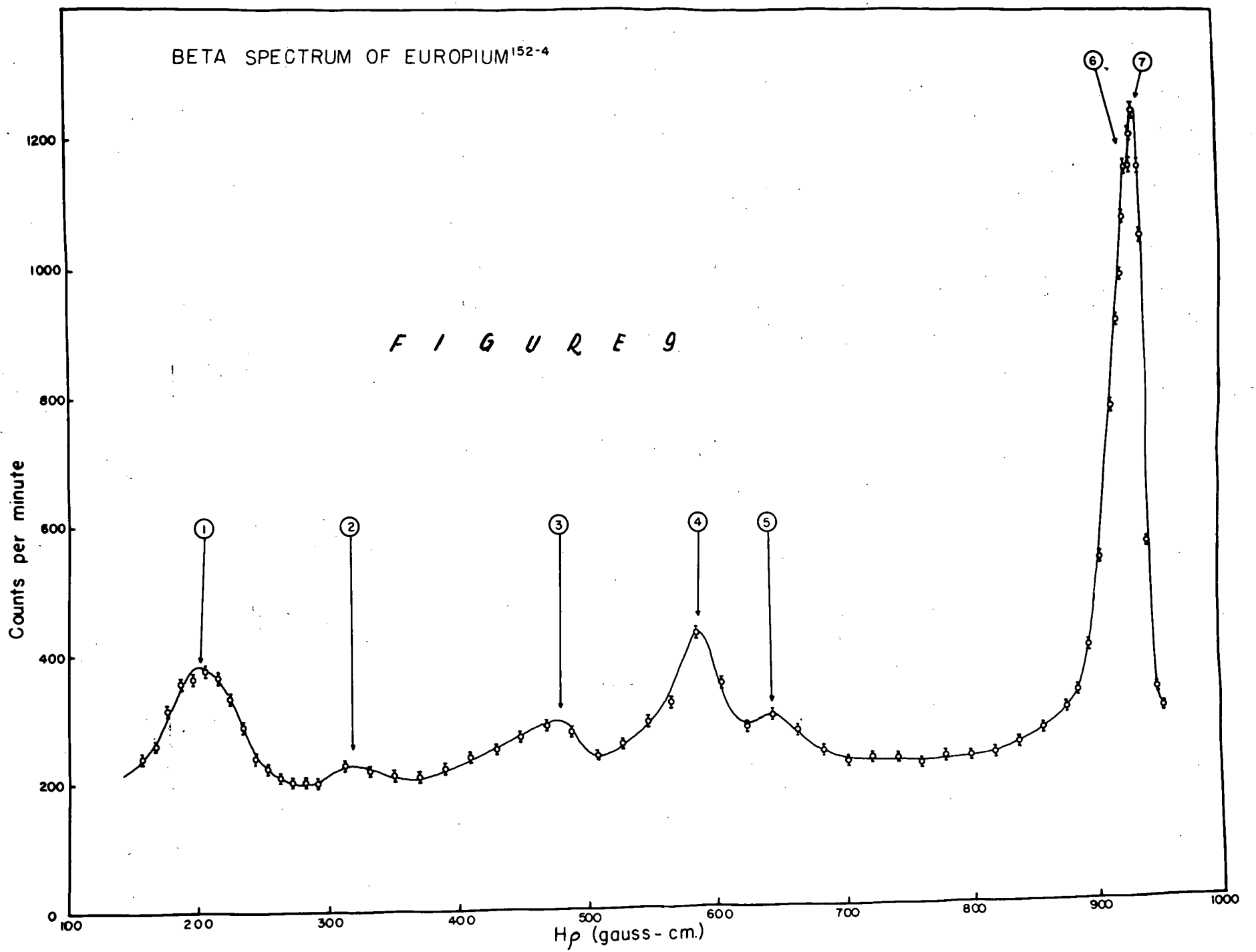
BETA SPECTRUM OF EUROPIUM 152-4
FIGURE 9
Counts per minute
Hρ (gauss - cm.)
0
200
400
600
800
1000
1200
100
200
300
400
500
600
700
800
900
1000
1
2
3
4
5
6
7

TABLE IV

Electron energy Kev	Interpretation	Energy Sum	
		$Z = 62$ Kev	$Z = 64$ Kev
32.0	L(Auger)(62)	39.1	
37.9	M(Auger)(62)	39.1	
72.9	K(64)		123.2
75.0	K(62)	121.8	

Peaks (1) and (2) lie at approximately the correct energies for L and M conversion of a 16 Kev gamma-ray transition in Gd or Sm. Some support is given for this assignment by the work of Mr. R. Azuma (privately communicated) using a sodium iodide proportional scintillation spectrometer. Azuma finds distinct evidence of a weak gamma-ray at this energy. Peak (1) is very broad. This may be partly caused by source-absorption but it should be noted that peak (1) corresponds roughly to the energy at which one would expect to find the Auger electrons due to atomic transitions which fill the L shell. Peak (2) cannot be ascribed to any Auger electron group in Gd or Sm because of energy considerations.

Peak (3) may be either a K conversion line corresponding to a gamma transition of about 73 Kev or an L conversion line corresponding to a gamma transition of about 34 Kev. If it is the former, an L conversion line would be expected at about 66 Kev. This was not detected

If peak (3) is an L conversion line an M conversion line would be expected at about 32 Kev. This line, if it exists, is possibly hidden by Auger peak (4). There is insufficient information available to determine in which daughter the transition occurs or which isotope of Eu it is due to. Because of energy considerations peak (3) cannot be ascribed to Auger groups in Gd or Sm.

Peaks (4) and (5) have been reported by Cork et al[32], and peaks (6) and (7) have been reported by these same workers as well as by Shull[34]. The present work is in good agreement with these experiments. Peak (6), although not fully resolved, is justified since the best smooth curve through the experimental points misses two consecutive experimental points by more than four times the probable error of these points.

The present work has extended the low-energy beta spectrum of Eu^{152-4} to energies lower than any previously reported and although insufficient information exists to fit the newly found transitions into an energy level scheme it is clear from this work that the decay schemes proposed by Cork et al[32] are incomplete, if not in error.

It might prove fruitful to repeat this experiment using, if possible, a thinner source of higher specific activity together with a more accurate calibration. Such an experiment could possibly resolve the two components suggested in peak (1) and thus provide conclusive evidence for the 16 Kev transition.

The fact that the source contains two activities, Eu^{152} and Eu^{154} of almost identical half-lives, plus the added difficulty of ascribing the gamma-ray transitions to the proper daughter product, i.e., Gd^{152}, Gd^{154}, Sm^{152}, or Sm^{154} makes the task of assigning a reasonable decay scheme to any of these elements impossible. It would appear that many gamma-rays are still undetected and the possibility still exists that the beta-spectra of both parent nuclei may contain more groups than the two already reported. Until more information is available over the entire energy region, the level sequences must remain unknown.

IV

RECOMMENDATIONS

The modifications incorporated into the spectrometer have satisfactorily facilitated its operation, nevertheless, more work must be done if this equipment is to produce accurate and reliable results quickly and efficiently.

The magnet coil, because it seriously overheats, cannot be operated continuously with currents in excess of 3 amperes. The necessity for intermittent operation reduces the efficiency of the spectrometer so that it is necessary to spend several weeks obtaining a single spectrum. Counter windows were found to last two or three days on the average. If continuous operation of the spectrometer were possible it is likely that a complete spectrum could be run without rupture of a counter window and its consequent laborious and time-consuming repair. To this end the magnet coil should be redesigned to permit continuous operation carrying the current necessary to produce a magnetic field of about 400 gauss.

Several improvements can be suggested for the spectrometer chamber. Firstly, a reduction in reflection of electrons from the walls could be effected by lining the chamber with lucite. For this reason the base plate as well should be made of lucite. Secondly, the leads from the

anodes of the counters should be brought out of the chamber through kovar seals to a shielded preamplifier mounted directly beneath the chamber. This would minimize pick-up as well as guard against sparkover inside the chamber and at the same time provide an adequate vacuum seal. Provision against source charging could be made by mounting a small filament above the source holder. Such a filament could likely provide sufficient thermal electrons near the source to discharge it without resorting to the use of an accelerating electrode. The chamber lid could be more quickly and reliably sealed to the chamber by using an O-ring rather than a flat rubber washer.

The counting rate of the Geiger counters was noted to deteriorate slightly with time. This effect is probably due to impurities in the heptane, e.g., dissolved oxygen could effect the counter characteristics. In any case an experiment should be performed to determine the cause of this defect so that it may be rectified. A jig could be constructed to facilitate the application of the counter windows. This would consist of a hinged support for the film holder and would permit the wet film to be swung uniformly into contact with the counter face where it could be left until dry. In this way reliable windows could be applied to the counters without taxing the dexterity of the operator.

Attempts should be made to devise new techniques for making thinner and more uniform sources, for without

question this is the crux of the problem of producing accurate results at energies below 20 Kev. An accurate calibration of the instrument should be made either by a direct measurement of the magnetic field or by use of an accurately measured internal conversion line.

APPENDIX I

COUNTER WINDOWS

The windows are produced by the method first published by Backus[24]. About 20 microlitres of a solution of one part zapon lacquer in two parts amyl acetate is dropped on the clean surface of distilled water. The solution spreads out over the surface of the water and the solvent evaporates leaving a thin film of zapon on the surface. This film is picked up on a wire frame so that the film falls on both sides of it making a double layer. The film was found to become brittle when left on the water surface for more than 20 seconds.

In the process employed by Brown these windows were permitted to dry before applying them to the counters. The counter face was prepared by cleaning it with amyl acetate and applying to it a thin film of vinylite resin prepared in the same manner as the zapon films. This film was applied immediately after it was stripped from the water. The entrance slit was then cleaned out with the sharp corner of a piece of paper and a dry zapon film was placed on the counter face, the vinylite acting as an adhesive.

Fortunately it has been found that the wet zapon films will adhere to the counter face quite adequately without the use of any bonding material. This is an advantage not only because it is a simplification in the window application technique, but also because it had been found difficult to clean out the counter window slot without disrupting the whole

vinylite film, thus rendering it useless.

Brown has shown that zapon films prepared in this manner are less than 10 micrograms/cm^2 thick and that their threshold for beta-particle transmission is below 2 Kev. For operation in the energy range above 10 Kev laminated windows of two or three zapon films have been utilized to good effect.

The windows, once applied, are tested by reducing the pressure inside the counters by about 1 cm of mercury. A small mercury manometer is provided in the system for measuring this pressure. After 5 or 10 minutes any leakage through the windows is readily detected by a change in reading of this manometer.

APPENDIX II

PREPARATION OF SOURCES

The source holder is made from a piece of 3/16 inch lucite 2 inches square. A rectangular slot 2 cm. long by 0.5 cm. wide is milled through the lucite with the back cut away at an angle of 45°, leaving a narrow edge around the slot. A hole is drilled in the lucite for mounting the source holder on the base plate of the spectrometer and a line is scribed on the lucite to indicate the position of the source.

The source backing is made of zapon in the same manner as the counter windows. The film is stripped from the surface of the water on a frame made of 1/16 inch lucite and laid on the source holder while still wet. The film is then allowed to dry before further work is done in preparing the source. Attempts to use LC600 films failed because this lacquer did not form a homogeneous solution in amyl acetate. The thinnest films that could be made with nylon were found to be about ten times as thick as those made with zapon.

A solution of one part zinc insulin[36] to eight parts distilled water is made up and about 0.25 microlitres of it is deposited with a pipette at each end of the film in line with the mark on the holder. The pipette is made

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36. L. M. Langer, R.S.I., 20,216(1949).

from tubing pulled down to 0.5 mm. outside diameter and 0.2 mm. inside diameter. The solution is brought up into the pipette and the end wiped clean with a paper towel. By carefully blowing into the pipette, the liquid can be made to bulge from the end without forming a drop. The end of the pipette when touched to the film leaves a droplet about 0.5 mm. in diameter.

The source is now placed in a jig constructed so as to permit the droplets to be run along the film in a thin line. The jig is made of brass and comprises a milled slideway beneath an adjustable ball-point pen which may be raised or lowered by turning a screw. The ball-point is carefully lowered to contact the film at the insulin droplet and the source holder is then moved in the slideway of the jig so that the ball-point is drawn along the source film. With care the droplet may be drawn back and forth along the film to form a line of insulin about 1 mm. wide.

A pipette similar to that used for depositing the insulin solution but made in the form of an eye-dropper is then used to deposit the active material. The active solution was made by dissolving about 0.5 mg. of europium oxide in a few drops of concentrated HCl. The europium oxide had been activated in the Chalk River pile and was subsequently left standing for about six months, by which time the short-lived isomer of Eu^{152} had been reduced to a negligible amount. The solution was concentrated by evaporation under a heat lamp until about 10 microlitres remained. The total activity

of the europium oxide was about 500 microcuries, so that pipetting off 0.25 microlitres of solution onto the source film would produce a source of total activity between 10 and 15 microcuries. By carefully manipulating the pipette one can make the active material run along the insulin line without flowing to the remaining film. After drying for a short time the source is ready for use.

APPENDIX III

THE COUNTER FILLING SYSTEM

The counters are filled with heptane (C_7H_{16}) maintained at a pressure of 1.1 cm. of mercury by keeping it in contact with liquid heptane in an ice bath. Commercial heptane was found to contain a small amount of water which required removal in order to permit proper operation of the counters. This was done by connecting a flask containing a few grams of phosphorous pentoxide to the neck of the flask which contains the liquid heptane. Because heptane is a good solvent for stopcock grease it was necessary to connect the heptane reservoir to the rest of the counter filling system by means of a graphite lubricated mercury seal stopcock.

The liquid heptane is introduced into the system by filling a small flask with heptane, freezing it in liquid air, and then sealing it to the spectrometer glasswork by means of a torch. The heptane flask is then carefully pumped while still frozen to remove all the air from it, the pumping being continued until evaporating heptane has completely flushed the system.

The characteristics of the heptane filled Geiger counters have been determined by Brown[25]. Although they do not exhibit a long plateau, they may be operated satisfactorily at about 1250 volts giving uniform pulses of about 4 volts peak amplitude with a rise time of less than one microsecond and a length of about 50 microseconds.

APPENDIX IV

AUXILIARY ELECTRONIC APPARATUS

A. Current Regulator.

In order to set and maintain a constant magnetic field to focus the beta-particles in the spectrometer it is necessary to be able to regulate and measure the current in the field coil. Since there is no iron in the system the magnetic field is linear with current. It is therefore possible to make a secondary calibration of the spectrometer at a single check point using an internal conversion line of accurately known energy or alternatively a primary calibration may be made by measuring the magnetic field produced by a measured current. Since the resolution of the spectrometer is of the order of 1%, the magnetic field should be regulated to at least 0.1%.

The coil current is regulated by passing it through a standard resistor and comparing the voltage developed across this resistor with the control voltage which is generated in a Student Potentiometer. The difference voltage is then amplified and fed back to control and grids of the current regulating tubes.

The standard resistor has a value of about 0.1 ohm and is made of 10 feet of 1 inch manganin strip. This resistor can carry the largest currents used with a temperature increase of less than 1°C, consequently its resistance

will stay constant to about 1 part in 10^5.

The amplifier comprises two channels. One channel amplifies the frequency range 0-10 cps. This channel is called the D.C. amplifier. The A. C. channel amplifies the frequency range 5-1000 cps. Amplification at higher frequencies is unimportant since the load is highly inductive. The signals from the two channels are mixed in a 6SL7 double triode which is used to drive the grids of a bank of eight 6AS7's in series with the magnet and the standard resistor. The input to the D.C. amplifier is 'chopped' in a Brown Converter, fed into an A.C. amplifier and subsequently detected in a phase sensitive detector. The overall voltage gain from the input of the D.C. amplifier to the output of the mixer is 5×10^5.

The eight 6AS7's will conduct currents up to 2 amperes. For current greater than this it is necessary to shunt the 6AS7's with an external shunt resistance. An additional variable resistance in series with the 6AS7's permits operation from 0 to 10 amperes from the 250 volt D.C. mains.

A block diagram of the regulating system is shown in Fig. 10 and a circuit diagram in Fig. 11.

The potentiometer output voltage is accurage to 5 parts in 10^4. The loop gain for the servo-mechanism is about 1.5×10^4. The voltage generated across the standard resistor is

$$V = V_o (1 + 1/G)$$

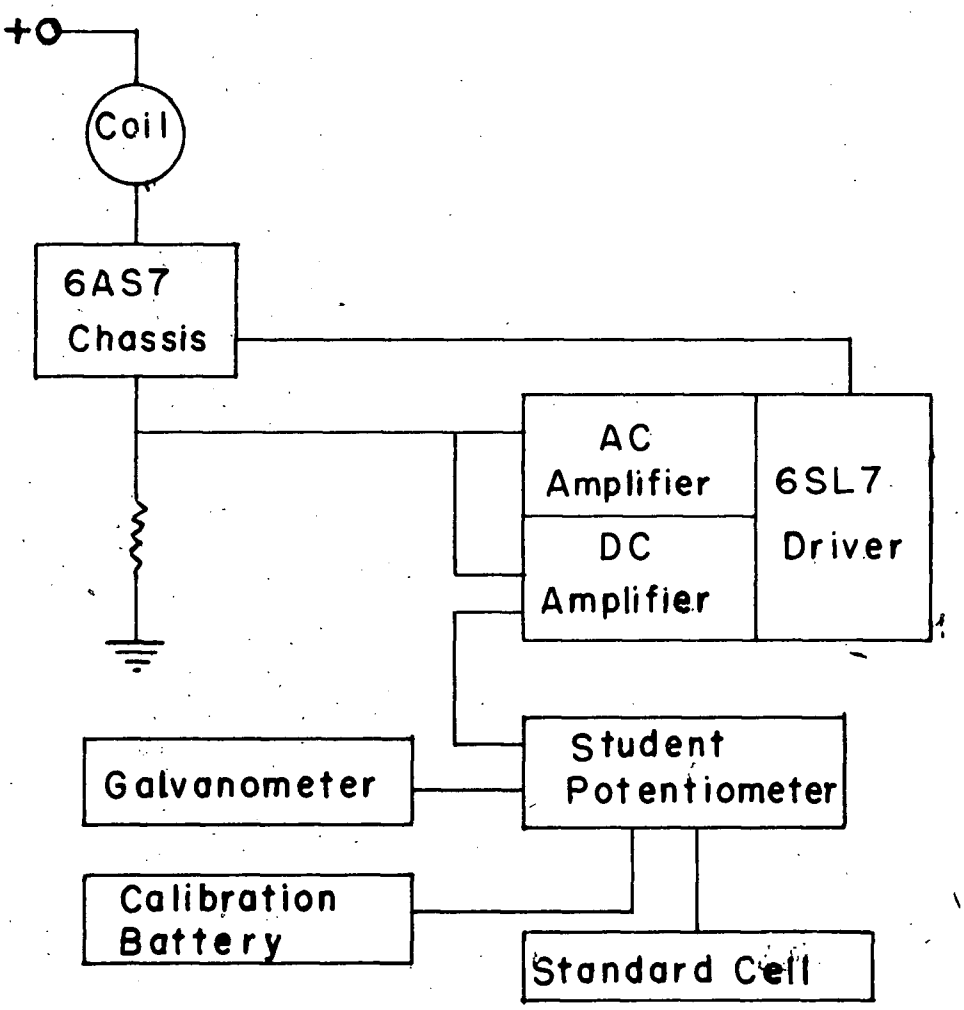

FIGURE 10

BLOCK DIAGRAM OF CURRENT

REGULATING SYSTEM

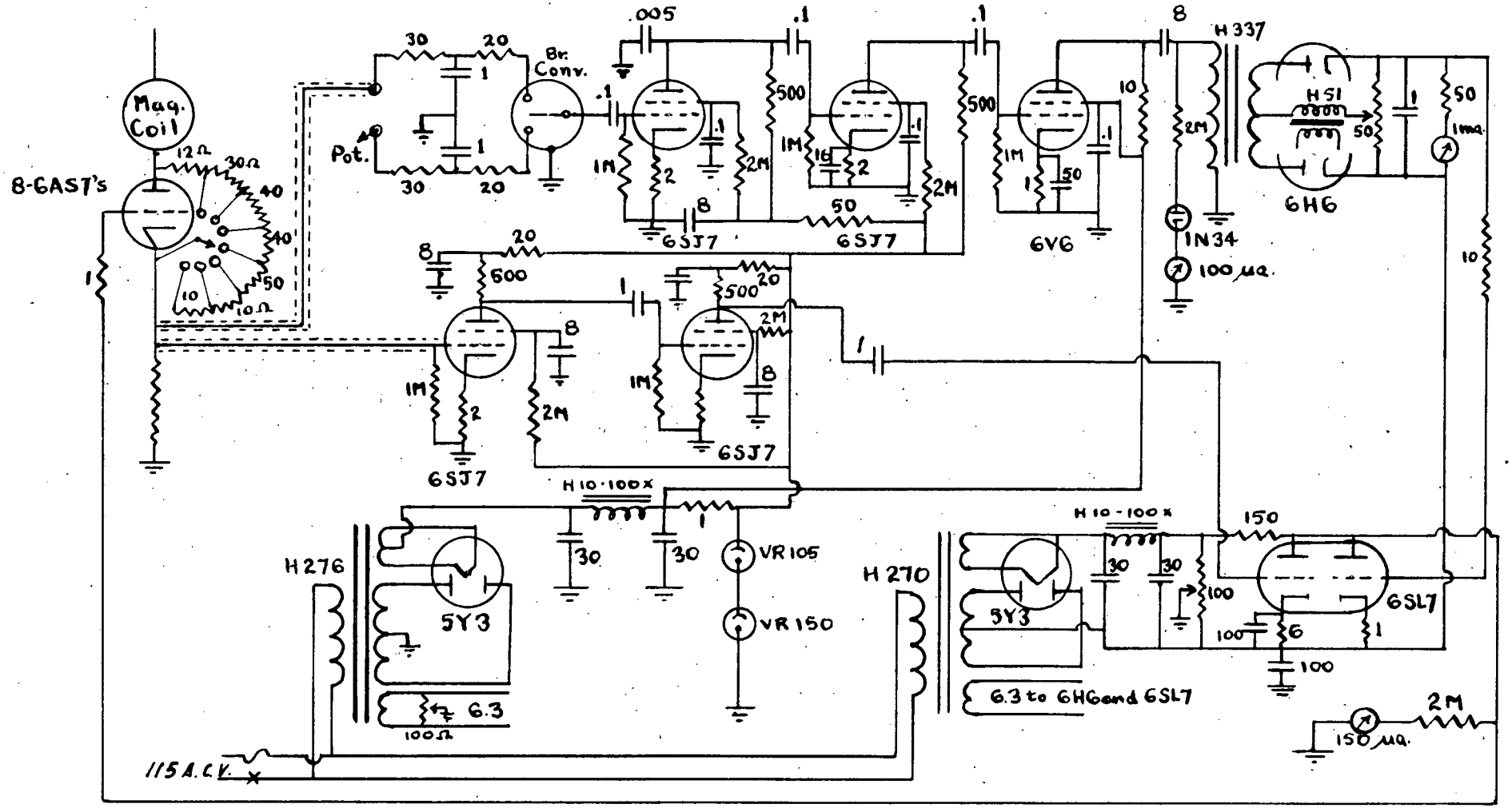

FIGURE 11

CIRCUIT DIAGRAM OF CURRENT REGULATING SYSTEM

where V_o is the potentiometer voltage, and G is the loop gain. Therefore the current is regulated to better than 1 part in 10^4 of the value corresponding to the setting on the control potentiometer. The A. C. ripple current in the magnet coil was measured to be less than 1 part in 10^3.

B. H. T. Regulation.

The voltage on the counter anodes must be well stabilized, since these counters do not exhibit a well defined plateau. A voltage regulator has been built by Brown[25] for this purpose. Tests made on this regulator show a change in output voltage of less than 5 volts for a change of 20% in A.C. input. The A.C. ripple in the output is less than 0.1%.

C. Pulse Amplifier and Scaler.

A simple 2 tube pulse amplifier with high input impedance and low output impedance was designed utilizing a double triode to serve half as an input stage and half as an output stage. It was necessary to use a shielded lead from the counters to the amplifier to reduce pickup. The output of the amplifier was fed into a scale of 1000, composed of 3 Berkeley scales of 10 followed by a mechanical register.