THE BETA RAYS OF RADIUM E AND ANTIMONY 124

by

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ABSTRACT

A thin-lens beta-ray spectrometer is described and a brief analysis of its operation is given. A coincidence Geiger-Mueller counter to be used with this instrument is also described.

The spectrometer has been calibrated with a line of the thorium B spectrum, and used to obtain beta-ray spectra of radium E and antimony 124. The experimental spectra have been found to agree well with those previously published. Several methods of plotting beta-ray spectra are described and applied to radium E and antimony 124.

From the Fermi plot the endpoint of the radium E spectrum appears to be at 1.18 Mev, from the van der Held plot at 1.16 Mev. For antimony 124 four endpoints have been determined from the Fermi plot at .50, .65, .90 and 2.43 Mev. It is shown that the van der Held plot reduces to the Fermi plot for this spectrum.

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BETA-RAY SPECTRA

One of the most important methods of investigating the structure of atomic nuclei is the study of nuclear radiation. Beta-ray spectroscopy is a particular branch of this study and is concerned with the beta and gamma rays emitted during the disintegration of radioactive isotopes.

When a negative beta particle is emitted from a nucleus of atomic number Z, the nucleus is changed to one of atomic number Z + 1. Similarly the emission of a positive beta particle lowers the atomic number by one unit. Very often a gamma ray accompanies such a transition.

The present explanation of this process assumes first of all, in accordance with quantum theory, that nuclei can exist only in discrete energy states or energy levels, and that in a transition between the levels of a nucleus of atomic number Z the energy difference is accounted for by the emission or absorption of a gamma ray. An energy level diagram for this case is shown in figure 1. Such a picture is fully in accord with the observed line structure of gamma rays. If this picture is now extended to beta transitions, serious difficulties arise. An energy level diagram here is of the type shown in figure 2, where a nucleus of atomic number Z changes to an exited nucleus of atomic number Z + 1 by the emission of a negative beta particle and then drops to the ground state by the emission of a gamma ray. We would expect then that all the beta particles emitted in such a transition have the same energy E, equal to the energy difference between the ground state of the parent nucleus and the exited state of the daughter nucleus.

This does not agree with experiment; the observed beta rays show a continuous energy distribution of the form shown in figure 3, where the maximum energy E_0 is equal to the energy of the disintegration. To overcome this difficulty without giving up the law of conservation of energy requires the postulation of another particle, the neutrino, which carries away the difference between the energy of the beta particle and the disintegration energy E_0. Such a particle would have to have an extremely small mass and no charge, making its detection most difficult. So far no experiment on the existence of the neutrino has been decisive.

At the present time the most comprehensive theory of beta disintegration is that proposed by Fermi(1) on the basis of a quantum mechanical calculation of the probability of emission of an electron and a neutrino which share the energy E_0. According to this theory the probability of emission of an electron whose energy lies between E and E + dE is

$$F(Z,E)dE = G^2 |M|^2 E\sqrt{E^2 - 1}\,(E_0 - E)^2 \frac{4e^{\pi y}}{\Gamma^2(3+2\gamma)} (2\rho\sqrt{E^2-1})^{2\gamma} |\Gamma(1+\gamma+iy)|^2 dE$$

where G is a constant

$|M|$ is a matrix element of the transition

$\gamma = \sqrt{1 - \alpha^2 Z^2} - 1$

Z is the atomic number

$\alpha = 1/137$

$y = \frac{\alpha E Z}{(E^2-1)^{1/2}}$

ρ is the radius of the nucleus

(The factors have been made dimensionless by choosing energy units in $m_o c^2$ and momentum units in $m_o c$.)

For any one beta-ray spectrum this can be reduced to

$$\left[\frac{N}{\eta^2 f}\right]^{\frac{1}{2}} = E_o - E$$

where N is proportional to the intensity of beta radiation of energy E

$f = \eta^{2\gamma} e^{\pi y} |\Gamma(1+\gamma+iy)|^2$

η = momentum = $\sqrt{E^2-1}$

This relation can be readily checked for any spectrum if N is taken as the observed count of beta particles and $\left[\frac{N}{\eta^2 f}\right]^{\frac{1}{2}}$ is plotted against E. This should result in a straight line for "allowed" transitions with the intercept on the E-axis being equal to E_o. Such a curve is called a Fermi plot and is of great importance of beta-ray spectroscopy: if it turns out to be a straight line it adds considerable weight to the Fermi theory; regardless of theoretical considerations, however, it will be an important method of finding E_o by extrapolation, if at least the portion of the spectrum near E_o can be plotted as a straight line.

It should be pointed out here that the determination of E_o by inspection from the spectrum as in figure 3 is very

PLATE I

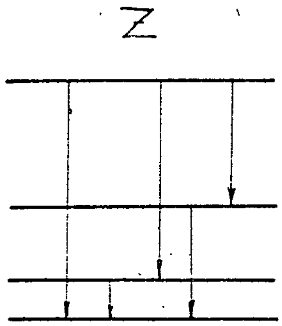

Fig. 1

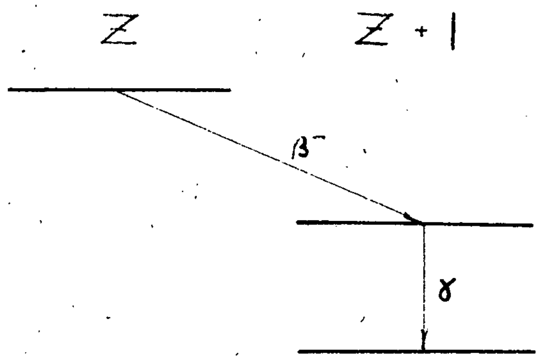

Fig. 2

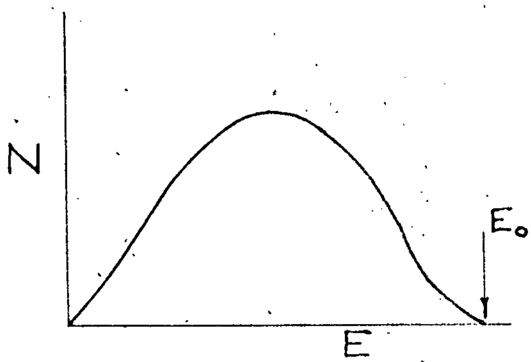

Fig. 3

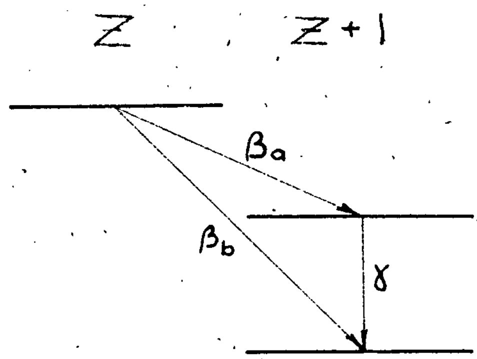

Fig. 4

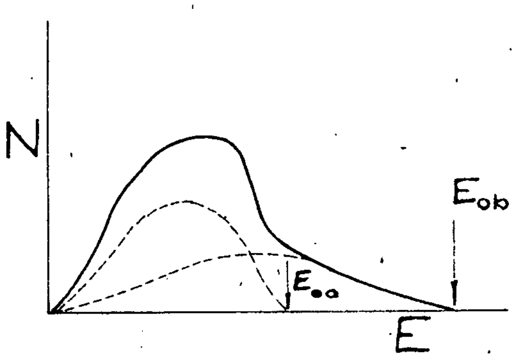

Fig. 5

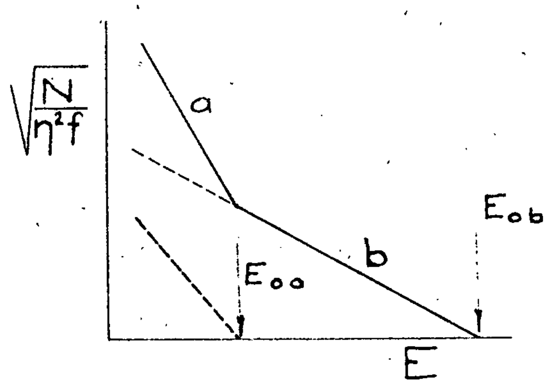

Fig. 6

unreliable, partly because of the small angle at which the curve approaches the E - axis, and partly because of the low counting rates in the neighbourhood of E_o.

The first Fermi plots showed considerable deviation from the straight line relation, and in 1935 Konopinski and Uhlenbeck[2] proposed a modification of the theory, which led to the new straight line relation

$$\left[\frac{N}{\eta^2 f}\right]^{\frac{1}{4}} = E_o - E$$

At first this seemed to fit the experimental curves better, but as more accurate determinations were made the original Fermi theory was found to be more generally successful.* The agreement is far from perfect, but in many cases the Fermi plot approximates a straight line, especially near the endpoint.

In an attempt to get better correlation with experimental spectra, van der Held[4] proposed using a linear combination of the terms arising out of the Fermi and the K - U theories. The Fermi plots are often straight near the endpoint, while the K - U plots are straight for lower

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*The ratio of K-capture to positron emission for Cadmium 107-1o9, as calculated from the Fermi theory, agrees well with experiment, while the K - U theory leads to a value sixty times too large.[3] On the other hand the half-life values calculated by integrating the Fermi formula do not, in general, agree with experimental values.

energies. A graph combining the two methods should therefore be straight over a greater range. In this case the function which should be plotted against E to give a straight line relationship is $\left[\frac{N}{\eta^2 f\left[1+c(E_o-E)^2\right]}\right]$. The constant c turns out to be approximately equal to 1.8 x 10^{-3} times the mass number for the sample curves made by van der Held. Whether this proportionality is of theoretical significance can not be decided at this time. For some elements it has been shown that the straight line character of the plot is retained over a larger region of energies than in the two other types of plot. This is of particular importance in the resolution of complex beta-ray spectra.

Consider an energy level diagram as in figure 4, where the emission of the beta particles may leave the daughter nucleus in one of two different states. To each of the two possible transitions will correspond a simple distribution as in figure 3, but it will be very difficult to separate two beta groups from a composite spectrum, which would have the form of figure 5. The resolution becomes very simple if there is a method of plotting separate beta groups as straight lines. If this can be done a straight line of a certain slope corresponds to each of the two beta groups, so that the plot of a double spectrum has the form shown in figure 6. Section b can be subtracted, leaving a straight line whose intercept on the E - axis gives the endpoint of the first beta group.

There is one other method of resolving complex spectra: that of coincidence measurements. This method is based on the fact that in a double spectrum (see figure 4) a beta ray of group a will in general be accompanied by a gamma ray, or followed by it after a time interval short compared to the resolving time of the coincidence circuit. The emission of a beta ray of group b leaves the daughter nucleus in the ground state without the emission of a gamma ray. If the recording apparatus is arranged in such a way that a beta particle registers only when it is accompanied by a gamma ray, no beta particles of energy greater than E_{0a} can be recorded. The chief difficulty of this method lies in the relatively low intensity available in coincidence studies.

It is obvious from this discussion that present theories of beta disintegration are not too satisfactory. It is the hope of beta-ray spectroscopists that as more investigations of energy level schemes are made, and as the methods of measurement become more precise, the shortcomings of the present theories will appear more definitely, thus leading to refinements in the theories, or possibly to a completely new theory to explain this very important fundamental process.

TECHNIQUES OF MEASUREMENT

The most direct method of measuring beta-ray energies is to observe the curvature of cloud-chamber tracks in a homogeneous magnetic field at right angles to the plane of the electron path. If the magnetic field is H gauss and

and the radius of curvature of the electron path is ρ, then the energy E in Mev can be found from the relation

$$H\rho = \frac{1}{3} \times 10^4 \sqrt{E(E+1.02)} \text{ gauss-cm}$$

Since the curvature of each track has to be measured separately a great many determinations have to be made in order to arrive at a reasonably accurate shape for the distribution curve.

Another method is to introduce various thicknesses of absorbers in the path of the rays and measuring the loss in intensity of the beta-ray beam. If R is the absorber thickness in grams per square centimeter which completely absorbs beta rays up to an energy E Mev then according to Feather[5]

$$R = .543\ E - .161$$

This is a purely empirical rule and is used normally for energies above .4 Mev.

For complex spectra the absorption and coincidence methods can be combined to correlate gamma rays with the proper beta-ray groups. This is a straightforward way of estimating the decay scheme of an isotope, but it is difficult to use it when the relative intensity of one of the beta groups is very small. For the most accurate determinations, however, beta-ray spectrometers are now universally used. These can be of various types, but all are based on the dispersion of charged particles in magnetic and electric fields.

The earliest and most common type of beta-ray spectrometer is the 180 degree or π-type. Here the rays

PLATE 2

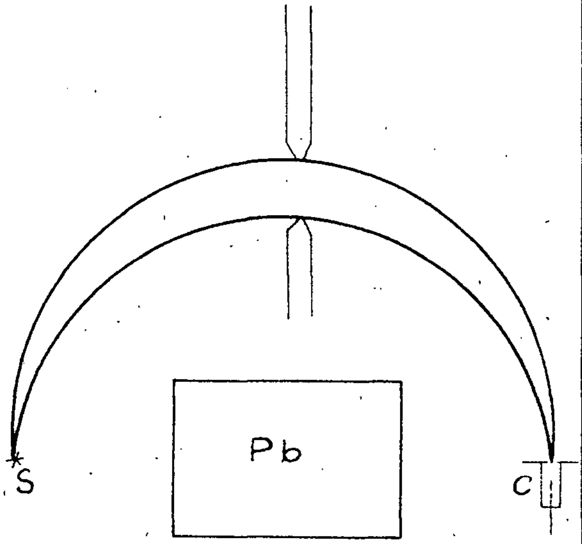


Fig. 7

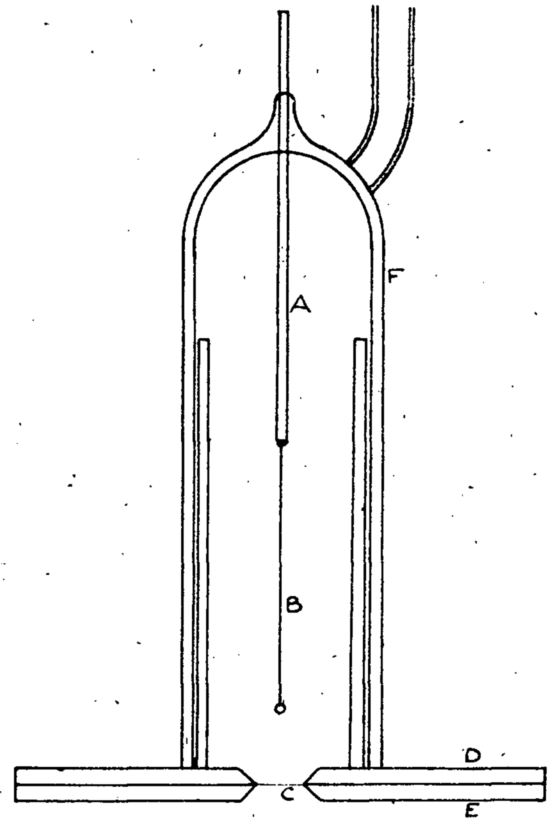


Fig. 9

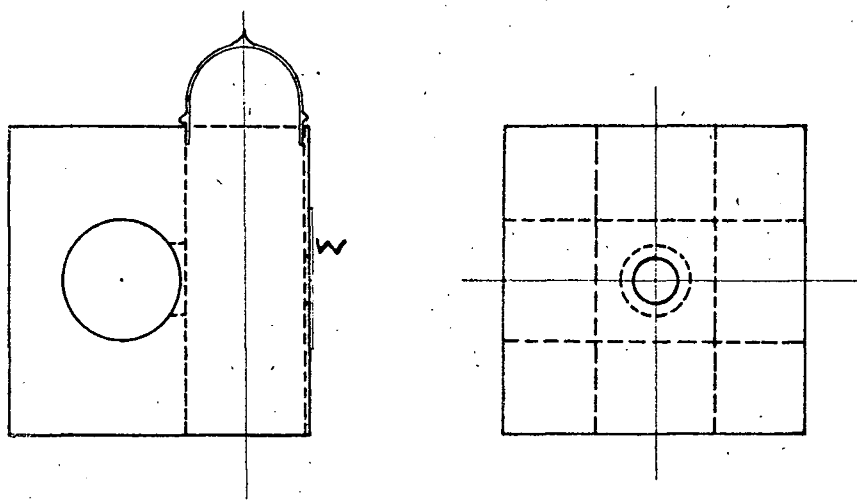


Fig. 10

describe circular paths in a magnetic field just as in the cloud-chamber determinations mentioned on page 6, so that if the radius of curvature is held fixed by suitable limitation of the beam (see figure 7) different energy bands can be made to arrive at the counter by varying the strength of the magnetic field.

If the magnetic field is in the same direction as the initial velocity of the electrons, as in a long solenoid, they will execute a spiral motion in that direction, returning to the axis after a certain distance. Here again different energies can be focussed at a counter, depending on the strength of the field.

A type of spectrometer using an inhomogeneous field is the thin lens type[6]. Since this is the type used in the present investigation, it will be desribed in detail under 'Description of Spectrometer'.

DESRIPTION OF THE SPECTROMETER

A photograph of the spectrometer is shown on plate three. Figure 8 on plate four is a schematic representation. A is a brass tube $7\frac{3}{4}$ inches in diameter and 40 inches long, which is evacuated to a pressure of about .001 mm Hg with a Cenco Megavac pump. An oil diffusion pump is also connected but is used only where lower pressures are desirable. C is a lead cylinder placed between F and J to absorb gamma rays from the source F which would otherwise register on the Geiger-Mueller counter J. The lead collimating baffle D

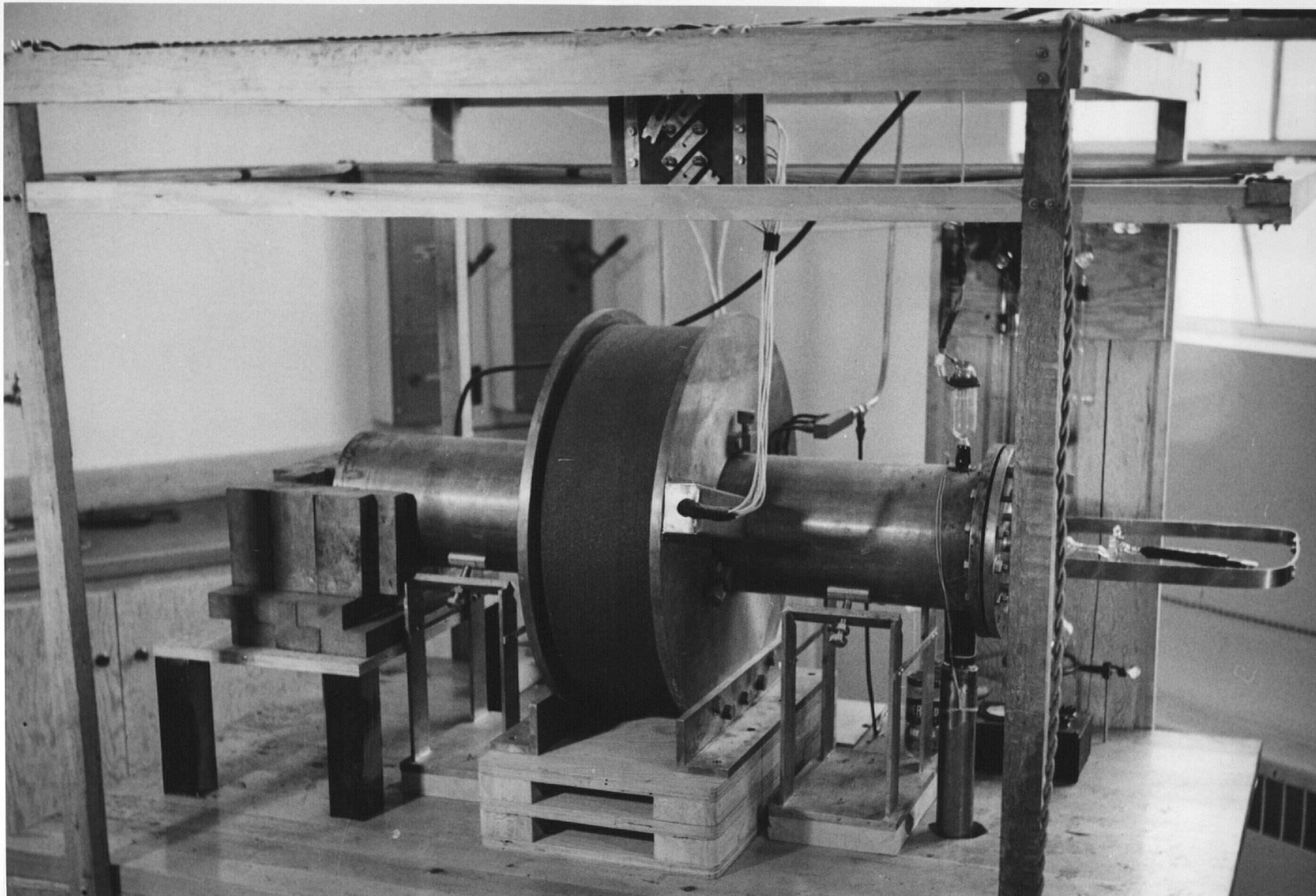


PLATE 4

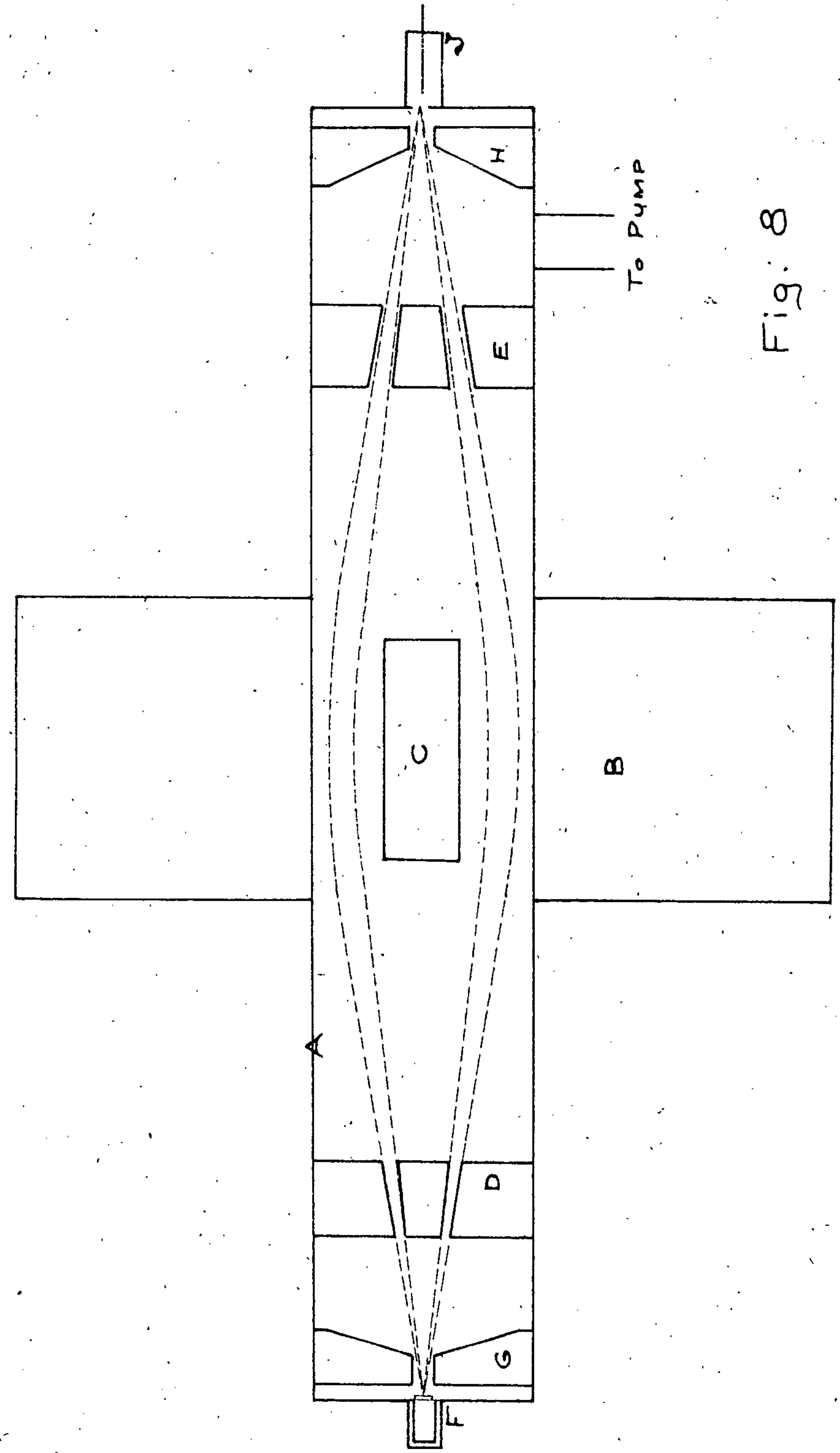


Fig. 8

allows only a conical shell of beta particles to pass through in the direction of the counter. In the region of the magnet B they are deflected in spiral paths, returning to the axis at the counter window. Such beta particles are said to be 'focussed'. The second lead baffle E is not necessary to limit the beam further but is introduced to reduce scattered beta and gamma radiation that without it would increase the background counting rate. For this purpose the baffles G and H have also been found to be quite effective.

The magnet is wound in four sections with number 10 wire with alternate layers of water cooling coils. The resistance of each section is about one ohm. The current is supplied by a 5 kva motor-generator and is stabilized by an electronic regulator, using 38 6AS7 triodes in parallel. The current is measured as the potential drop across a manganin resistance of about .8 ohms with a Rubicon potentiometer.

In order to eliminate the effect of the earth's magnetic field the spectrometer is aligned along the horizontal component of the earth's field, and two Helmholtz coils are used to compensate for the vertical component.

The Geiger-Mueller counters are of the bell type, shown in figure 9. A is a .020 inch tungsten wire to which the .005 inch tungsten wire B is attached with a drop of silver solder. At the end of wire B is a small glass bead. The window C is of thin mica which is sealed between the two brass plates D and E with a mixture of equal parts of beeswax and rosin. The mica used had a thickness of about two mg/cm^2, although it was found possible to split the mica to less than one mg/cm^2. The glass envelope F was waxed to the base D with Apiezon wax "W". The counter was filled with a mixture of 9.5 cm argon and .7 cm ethyl alcohol.

The pulse from the Geiger-Mueller counter goes to a cathode-coupled preamplifier stage using a 6J6 miniature twin triode, and then to a scale of 64 scaler built by the Atomic Instrument Company. The scaler is connected to a mechanical register made by the Cyclotron Specialties Company.

In analogy to optics the spectrometer may be said to consist of a thin lens, an object and an image, with the image distance equal to the object distance and equal to twice the focal length of the lens. The focal length of the lens is determined by the field current. Just as in the optical case there is a chromatic dispersion, i. e. rays of different energies will be focussed at different points on the axis. The original beam is thus separated into rays of different energies and only a small bundle of such rays reaches the counter for any one value of current through the coil. The momentum interval of the focussed rays as a percentage of their average momentum is called the resolving power, and in this instrument it is about three per cent.

The radius of curvature ρ of the focussed rays is held constant by the position of the baffles so that from the equation $H\rho = 1/3 \times 10^4 \sqrt{E(E + 1.02)}$ gauss-cm there is thus a direct correspondence between the strength H of the field and the energy E of the rays registered by the counter. No attempt is made to arrive at an absolute value of either H or ρ and the instrument is calibrated with a gamma ray conversion line of known $H\rho$. Since no iron is present the magnetic field strength is directly proportional to the current through the coil. Hence for each value of current, beta rays of a definite energy or momentum are focussed on the counter.

COINCIDENCE COUNTER

In all work with the spectrometer there is a steady background counting rate which is caused to some extent by cosmic rays, but mainly by scattered beta and gamma rays. Since this background is an important limit to the accuracy of the determination, it is important to keep it as low as possible. Up to the present this has been done by lead baffles as described earlier. It is proposed to reduce it further by using two connected beta-ray counters next to one another and operated in coincidence. See figure 10.

A beta ray coming at the proper angle will pass through the window W and register in both counters. A gamma ray, however, will liberate a secondary electron which will register in the first counter but has only a small probability of going in the right direction to pass into the second counter and registering as a coincidence.

Such a counter has been made and tested outside the spectrometer with a beta-ray source. The following sample readings will serve to indicate its performance.

Source $4\frac{1}{4}$ inches from window

	total count	background
counter A	9468	77
counter B	3829	108
coincidences	3439	16

It will be seen that the coincidence counting rate and the counting rate of counter B are not greatly different so that it is expected that the introduction of this double counter will not appreciably affect the normal counting rate. The background for coincidences is seen to be considerably reduced over that of a single counter.

Due to mechanical difficulties it has not been possible to test this counter in the spectrometer before this time.

CALIBRATION

The spectrometer was calibrated with the F-line of thorium B, for which $H\rho$ is 1385.6 gauss-cm. This line has been determined very accurately [7,8], and is now widely used as a secondary standard. The source was prepared by precipitation as a sulfide from a thorium nitrate solution, and put on a backing of mica. It was covered with a drop of collodion solution to hold it in place. This isotope has a half-life of 10.6 hours so that all readings had to be corrected for decay.

Two sources were used, but only the second calibration was successful. The first source had a considerable amount of inert material added to it as carrier, with the result that the source was so thick that scattering broadened the line to the extent of being almost indistinguishable from the background.

EXPERIMENTAL RESULTS

A. The Beta-Ray Spectrum of Radium E

Radium E is a good example of a beta-ray emitter with a simple spectrum. It has been studied by many investigators(9,10,11), yet there is considerable variation in the reported endpoints. Most of the difficulty seems to be due to the fact that the Fermi plot, instead of being a straight line, is concave upwards. When the Konopinski - Uhlenbeck modification of the Fermi theory was first announced it was thought that it was more successful for radium E, but as more accqurate measurements were made it was shown that the K - U plot drops sharply near the endpoint. Thus extrapolated K - U plots give endpoints which are much too high.

Van der Held was able to get plots which approximate straight lines very closely, but his method of plotting is somewhat more complicated than for the other two cases. (See page 5) It is necessary to get a preliminary value of the endpoint from the Fermi plot, and also to use a constant c, determined from the slope of the line obtained when $\left(\frac{F}{E_o-E}\right)^2$ is plotted against $(E_o - E)^2$. (F here is $\left[\frac{N}{\eta' f}\right]^{\frac{1}{2}}$ as calculated for the Fermi plot.) It should be noted that the Fermi theory is a special case of the van der Held theory, with c equal to zero. Van der Held showed that for the few elements which he studied c was proportional to the atomic mass number A, with c/A approximately equal to 1.8×10^{-3}.

The source used was one of metallic radium D in equilibrium with its daughter products. The beta and gamma rays from radium D have very low energies (less than .05 Mev) so that they did not interfere. Since the energies in the region to be studied were near 1 Mev no particular precautions to reduce scattering had to be taken.

The high energy end of the spectrum is shown on plate 5. The difficulty in determining the endpoint from this is apparent. The Fermi plot is shown on plate 6. Its curvature is quite pronounced, so that extrapolation becomes unreliable. The endpoint is at 1.18 Mev. The van der Held plot (plate 7) shows a straight line, disregarding the last three points. With a three per cent resolving power of the instrument these points will be shifted to the right so that the endpoint appears 1½ per cent too high. Since the statistical accuracy of these points is fairly low, it seems best to disregard them in drawing the straight line. The endpoint is then at 1.16 Mev. The constant c has been taken as $210 \times 1.8 \times 10^{-3}$ which is the average value obtained by van der Held for radium E.

B. The Beta-Ray Spectrum of Antimony 124

The beta-ray spectrum of antimony 124 has been reported on by about ten research groups(12-21). Until recently it seemed to consist of two beta-ray groups, the first with an endpoint between .50 and .74 Mev, and the second between 1.53 and 2.54 Mev. It seemed desirable to determine these endpoints more closely, and a sample of antimony 124 was therefore ordered for the present investigation. In January 1948, however, two reports were published[20,21], giving the endpoints of five beta-ray groups emitted by this isotope. With this complication the investigation of this spectrum became even more interesting.

The source was prepared by irradiating antimony trioxide with slow neutrons in the heavy water pile at Chalk River. The antimony trioxide was specially purified in the department of chemistry, and spectroscopically tested. The spectroscopic analysis showed that the only impurity present was copper (.002 %) with sodium, calcium, tin and iron probably absent and arsenic, lead, bismuth, lithium, potassium, silicon, manganese, strontium and barium definitely absent. The irradiated oxide had a specific activity of one millicurie per gram. The source strength used was a few microcuries, on a backing of mica of thickness less than 1 mg/cm^2. The source was covered by a thin film of collodion, prepared by the method of Backus[22].

All readings were corrected for decay, assuming a half-life of 60 days, as given by Livingood and Seaborg[23]. Some difficulty was experienced in reproducing parts of the spectrum, and it was necessary to normalize two sections of the spectrum, since the intensity had dropped by about 2 per cent in the one case and by about 15 per cent in the second. The possibility of some antimony 122 (half-life 63 hours) being present can not be ruled out, although at least three weeks had elapsed after the end of irradiation of the sample.

The spectrum is shown on plate 9. It shows a conversion line at $.57^8$ Mev, corresponding to a gamma ray energy of $.61^0$ Mev. This line has also been reported by Kern, Zaffarno and Mitchell[20] who obtained a gamma-ray energy of .603 Mev. A Fermi plot is shown on plate 8. It consists of four straight line sections, with endpoints agreeing well with those reported earlier this year. A tabulation of results is shown below: (energies in Mev)

Kern, Zaffarno, Mitchell[20]	Cook, Langer[21]	Mann, Lindenfeld
.47	.50	.50 ± .02
.63	.68	.65 ± .03
.98	.98	.90 ± .06
1.58	1.50	--
2.31	2.37	2.43 ± .03

There is an indication of a group with an endpoint near 1.5 Mev, but it seems to be very weak. It must be stressed that these endpoints are derived wholly from the Fermi plot, and therefore depend entirely on the accuracy of the Fermi theory. They can not be regarded as well established until they are confirmed by coincidence measurements.

A plot of $\left(\frac{F}{E_0-E}\right)^2$ vs. $(E_0 - E)^2$ gave approximately a straight line with a slope either zero or at least very small.

Thus the van der Held plot for this case reduces to the Fermi plot. The van der Held plot was also plotted using $c = 124 \times 1.8 \times 10^{-3}$, but it showed no indication of being composed of straight line sections. This shows the importance of determining c separately for each spectrum, since the proportionality to the mass number, as suggested by van der Held, does not seem to hold in all cases.

CONCLUSIONS

The work on radium E has confirmed the work of many other investigators who have found that the Fermi plot for this case is not straight. The van der Held plot is apparently better in this respect. The endpoint from the Fermi plot is 1.18 Mev, from the van der Held plot 1.16 Mev.

The Fermi plot for antimony 124 shows four endpoints at .50, .65, .90, and 2.43 Mev. A fifth group, reported by Kern, Zaffarno, Mitchell[20] and by Cook, Langer[21] is too weak for significant evaluation in this experiment. The van der Held constant c is near zero for antimony 124, so that van der Held's suggestion that c is proportional to the mass number appears to be incorrect.

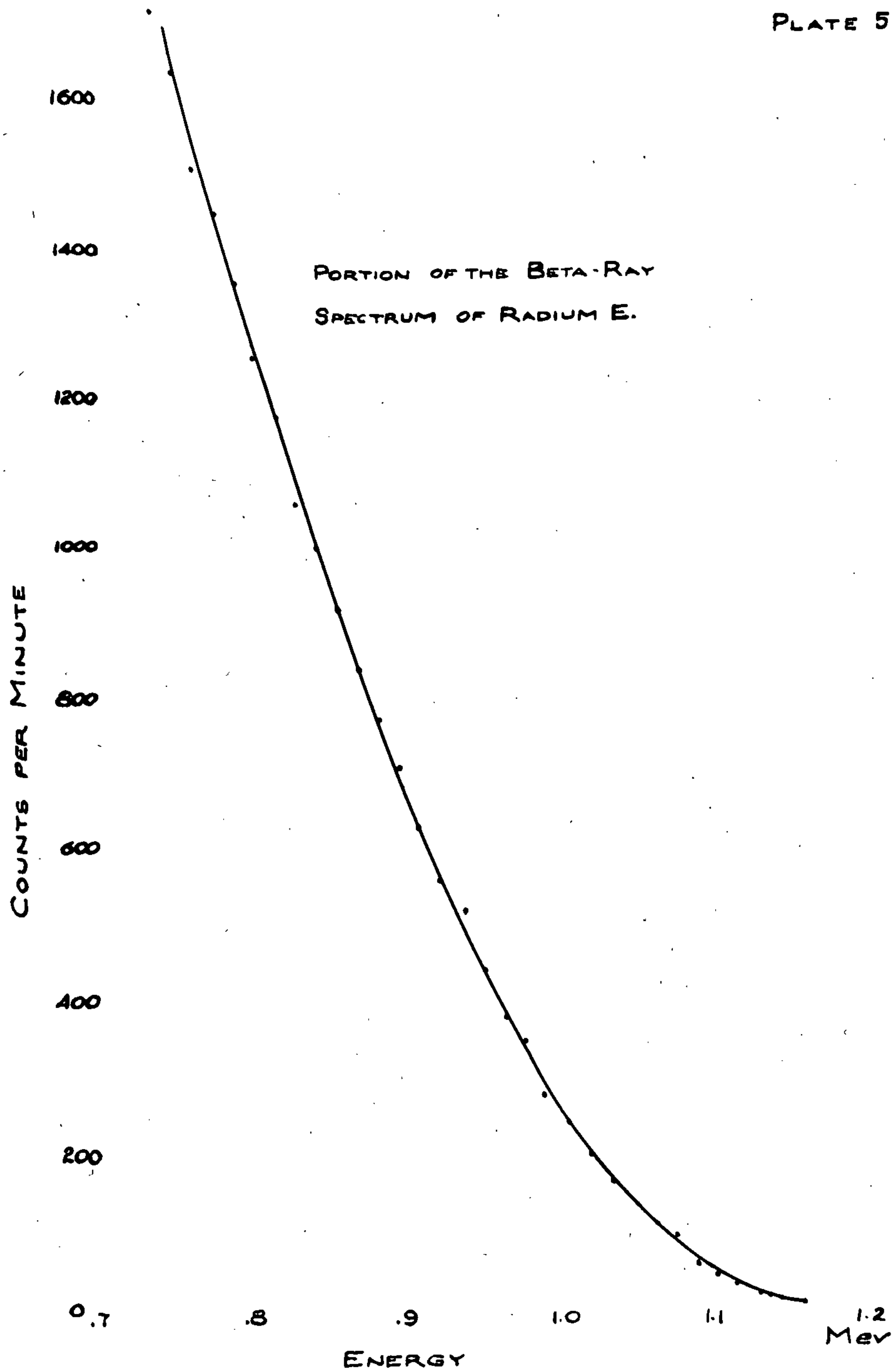
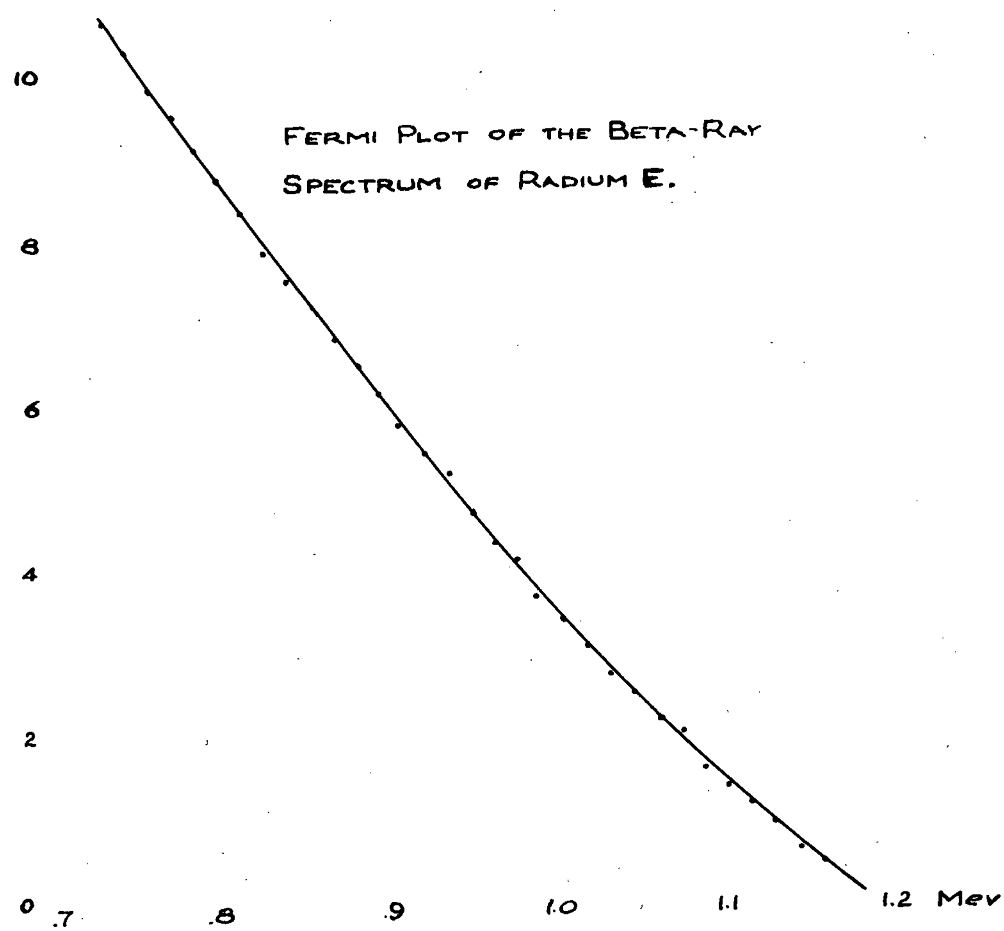
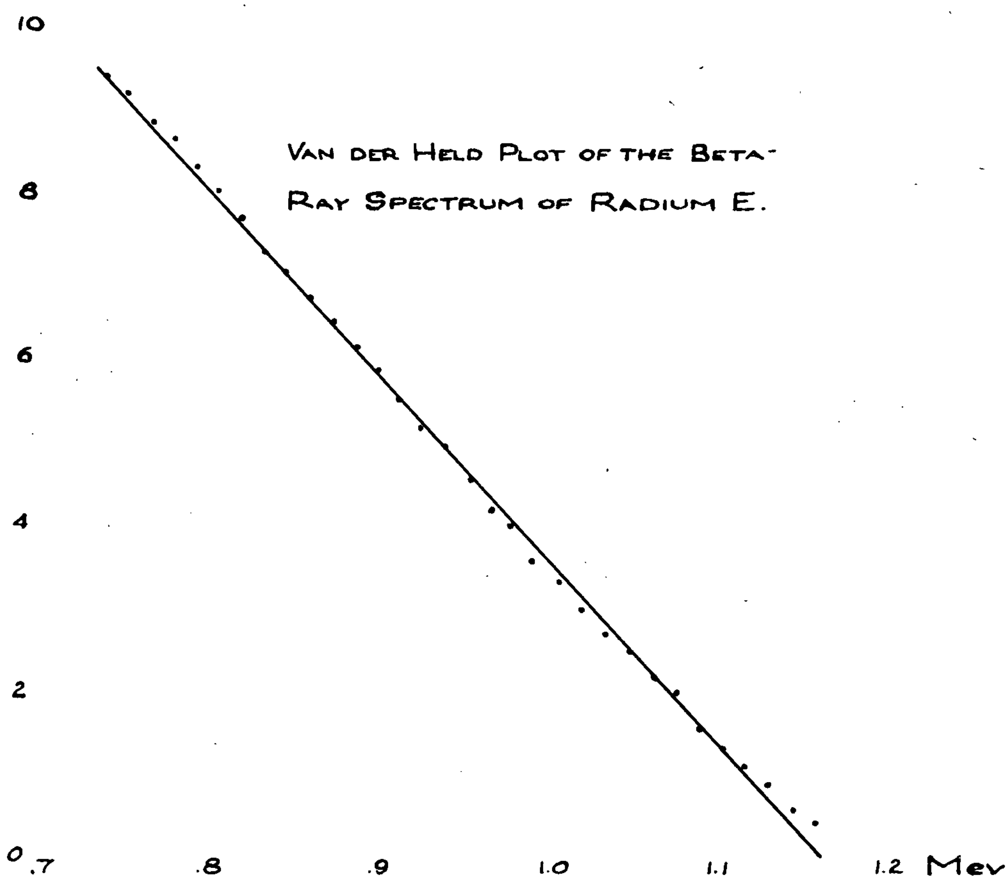

PORTION OF THE BETA-RAY
SPECTRUM OF RADIUM E.
COUNTS PER MINUTE
1600
1400
1200
1000
800
600
400
200
0
.7
.8
.9
1.0
1.1
1.2
Mev
ENERGY

PLATE 6



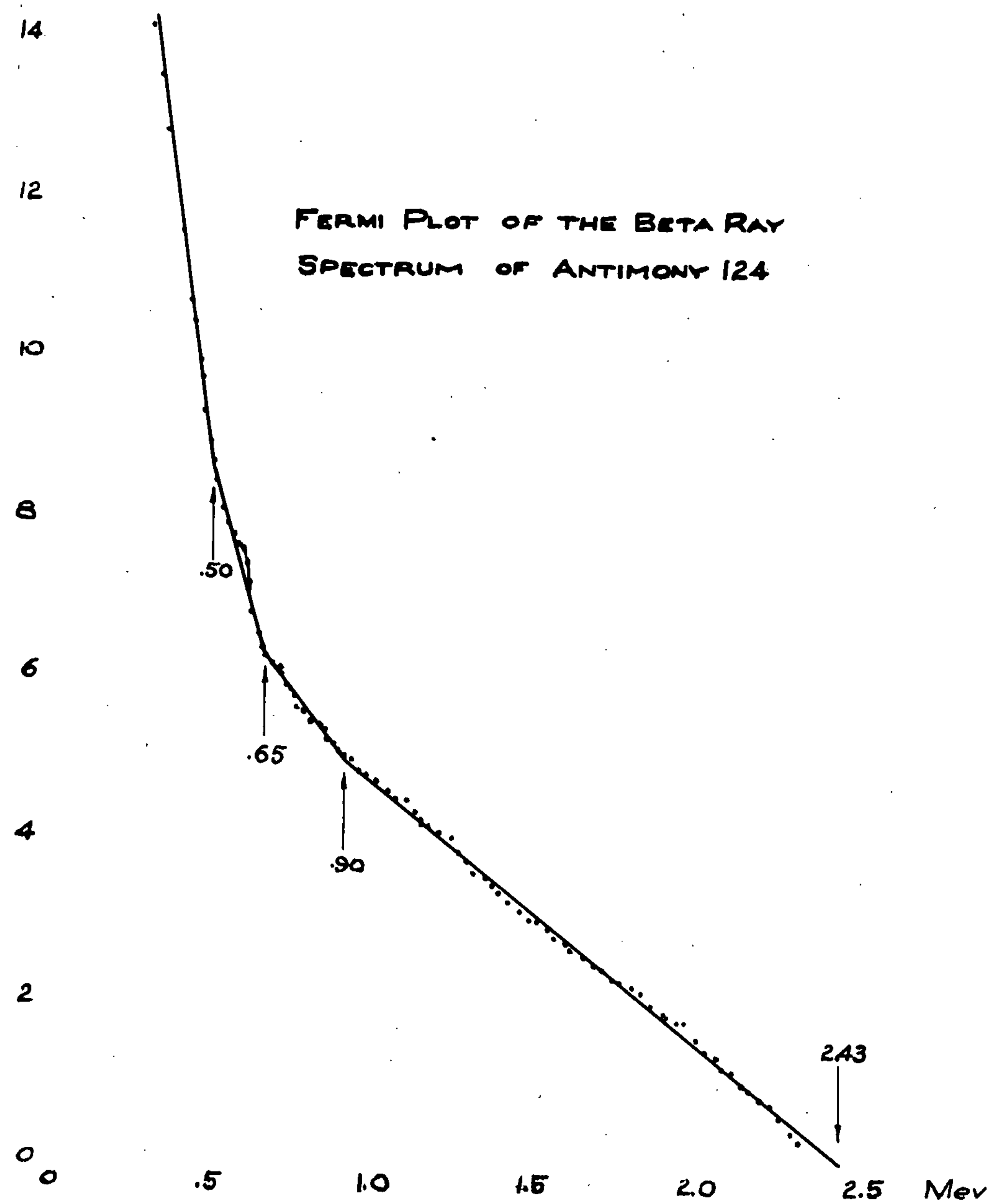
10
8
6
4
2
0
.7
.8
.9
1.0
1.1
1.2 Mev
FERMI PLOT OF THE BETA-RAY
SPECTRUM OF RADIUM E.

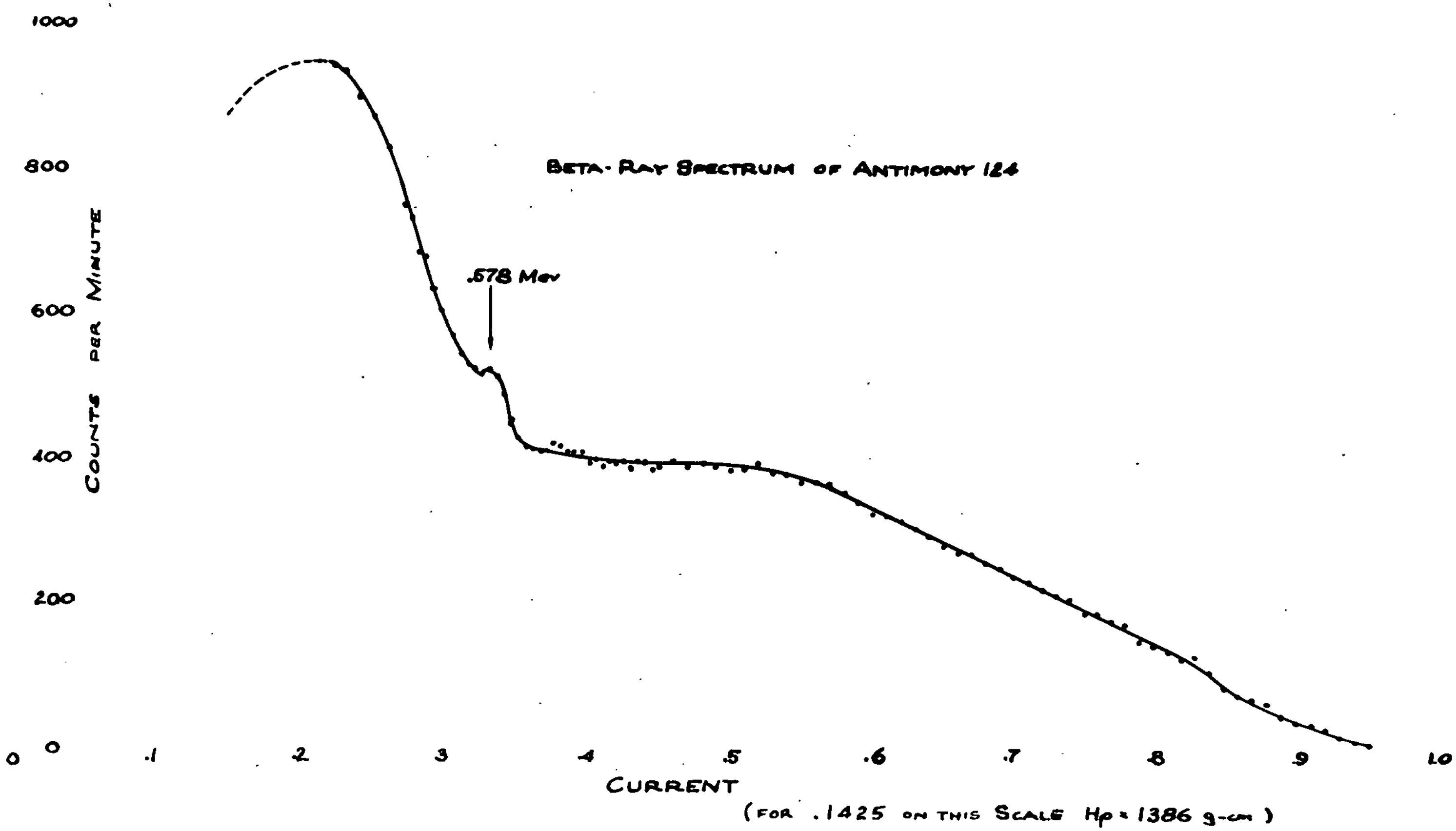
Plate 7



Van der Held Plot of the Beta-Ray Spectrum of Radium E.

PLATE 8


FERMI PLOT OF THE BETA RAY SPECTRUM OF ANTIMONY 124
14
12
10
8
6
4
2
0
0
.5
1.0
1.5
2.0
2.5
Mev
.50
.65
.90
2.43



Beta-Ray Spectrum of Antimony 124
.578 Mev
1000
800
600
400
200
0
Counts per Minute
0
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
Current
(for .1425 on this Scale Hρ = 1386 g-cm)

PLATE 8

ACKNOWLEDGEMENTS

The beta-ray spectrometer and auxiliary apparatus are part of a grant-in-aid to Dr. K. C. Mann from the National Research Council of Canada. The National Research Council has also awarded a studentship to me for the session 1947-48.

Two tons of lead have been donated by the Consolidated Mining and Smelting Company of Trail, B. C. for the protection of the experimenters.

Mrs. B. E. Speers of the Department of Chemistry has purified the antimony trioxide and prepared the two thorium B sources.

Dr. A. M. Crooker has made the spectroscopic analysis of the antimony trioxide.

Mr. A. W. Pye has made the glass parts of the Geiger-Mueller counters and the filling systems.

Their help is gratefully acknowledged.

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