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Energies of the Soft Beta-Radiations of Rubidium and Other Bodies. Method for Their Determination

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A method for measuring the upper limits of soft beta-spectra by use of screen wall tube counters filled with hydrogen gas is described. Illustrative results are given for Rb^{87}, Na^{22}, S^{35}, and Au^{198}.

INTRODUCTION

THE necessity of checking any relation between the decay constants of beta-radioactive nuclei and the energies of the emitted radiations of the type Sargent[1] has suggested makes data on the energies of soft beta-spectra valuable. The difficulties of such a measurement, arising principally from the high probability of scattering for low energy electrons, have previously prevented our obtaining of data of desirable reliability for the rather numerous long-lived soft emitters.

The following discussion is intended to be a brief presentation of a method which apparently will serve for beta-radiation harder than about ten or twenty kilovolts. Illustrative data are given.

APPARATUS AND METHOD

The method[2] employs a screen-wall Geiger-Müller counter placed along the axis of a surrounding cylinder which has the radioactive body mounted on its inner surface. Fig. 1 shows the counter. The whole chamber is evacuated and filled with hydrogen gas at a few centimeters of Hg pressure and is placed in a solenoidal magnet with its axis parallel to the field. The effect of the sample on the counter (obtained by movement of the sample cylinder inside the counter chamber) is studied as a function of the field and, in general, is found to disappear at a certain value of the field. If gamma-radiation is present there is a constant residual effect. The maximum energy of the beta-spectrum is determined by the field at which the effect of the sample on the counter is just eliminated. The $H\rho$ (product of field and radius of curvature of electron track) value for the upper limit of the spectrum is this limiting field multiplied by half of the shortest distance from the sample surface to the counter, because any smaller field allows some of the curved tracks to penetrate the counter. The geometrical arrangement used in this method was suggested and used by Bocciarelli and Occhialini[3] on the potassium radiation with a solid-wall counter as detector.

The amplifying and recording units used with the counters are of conventional design for use with solid-wall counters. In fact, screen-wall

[1] B. W. Sargent, Proc. Roy. Soc. **A139**, 659 (1933).
[2] W. F. Libby, Phys. Rev. **46**, 196 (1934).
[3] D. Bocciarelli, Nature **128**, 375 (1931).

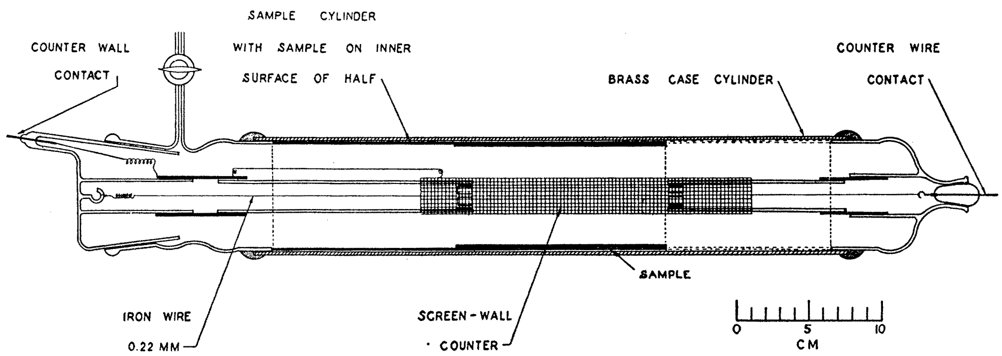


FIG. 1. Screen wall counter assembly.

counters differ in only one essential respect from solid-wall counters insofar as operation is concerned. It is necessary to provide an electric field between the counter and the surrounding chamber wall, presumably to prevent the accumulation of ions in the intervening space some of which would be free to move back into the counting volume and trip the counter. Under normal operating conditions the chamber wall (and sample cylinder) is kept about fifty volts positive relative to the counter wall by means of an insulated battery. The size of this potential drop seems to be rather unimportant providing it is sufficiently small to prevent sparking across the gap. It is convenient occasionally to ground the chamber for purposes of handling and to allow the full counter voltage to serve as a clearing voltage. Fig. 2 shows the effects produced by changing the sign and magnitude of this clearing field for a standard counter with diameter of 2.5 cm and sample cylinder diameter of 6 cm. The gas was helium at a pressure of 6 cm of mercury, with a trace of air impurity. The three curves are for the background, potassium radiation plus background, and aluminum photoelectrons plus background. The aluminum photoelectrons were obtained by illuminating an aluminum sample cylinder with visible light. The rise observed with the chamber wall more negative than the counter was due to an effective increase in size of the counter. In a certain sense, the screen is replaced as the counter wall by the chamber wall and sample cylinder. Various uses of this arrangement for construction of large sensitive counters suggested themselves. In particular, a soft radiation incident at the wall of a counter of large diameter might not produce enough ion pairs to initiate the counter discharge in the case of the ordinary counter. However, in the case of the arrangement described above with a sufficiently large negative potential on the sample cylinder with respect to the screen, the initial ionization will be increased more than it would have been in the standard counter because of the larger fields near the outer wall. The chances of counting are necessarily increased. Evidence for this appears in the figure in the relative effects of changing the sign of the clearing field on the potassium beta-radiation and the aluminum photoelectrons. The effect on the latter is an increase of considerable size while that on the potassium-plus-background radiation is little more than is observed with the background radiation alone. When the clearing voltage is removed entirely the counters have a tendency to develop high backgrounds which finally grow until they prevent the counter's operating at all. This may require only a few seconds with small assemblies.

The magnetic fields used (6000 gauss at most) have two effects on the operation of the counter which, though small, must be considered. The operating voltage is reduced by placing the

TABLE I. *Counter dimensions.*

Counter Name	Counter Diameter (cm)	Counter Length (cm)	Counter Wire Diameter (mm)	Sample Cylinder Diameter (cm)	Value of ρ (cm)
Large	2.5	14	0.22	13.5	2.75
Medium	2.5	14	0.22	10.1	1.90
Small	2.5	14	0.22	6.1	0.90

screen-wall counter in a field; the effect is about two percent at 6000 gauss. The explanation of this fact is not clear, though it is possible that it is due to an increase in probability of recombination of ion pairs formed in the space between the counter wall and the chamber walls before the positive ions so formed have time to move to the negatively charged counter wall. In the absence of the field these ions would tend to collect on the screen and reduce the negative

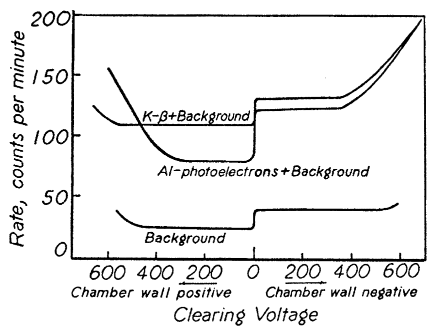


FIG. 2. Effects of clearing fields in small counter (6 cm He as gas).

potential. The effect apparently is associated with the curving of the paths traversed by the ions and the consequent decrease in their chance of reaching the walls before recombination occurs. The result is that the field allows the screen to maintain a higher average negative potential and requires a reduction of the applied voltage relative to the field-free value. This effect is not observed with solid-wall counters. The adjustment is made in practice by setting the voltage so the shape of the kicks, as shown on an oscillograph, remains constant over the whole range of variation of the field.

The other effect of the field is a linear reduction of the background as the field is applied which has an obvious explanation in the elimination of electron secondaries ejected from the chamber wall and sample cylinder by background gamma-radiation, etc. This effect amounts to a reduction of about thirty percent in most cases. The data presented later show this effect and are quite typical.

THE MAGNET

The magnet used was a solenoid with central hole six inches in diameter and one foot long.

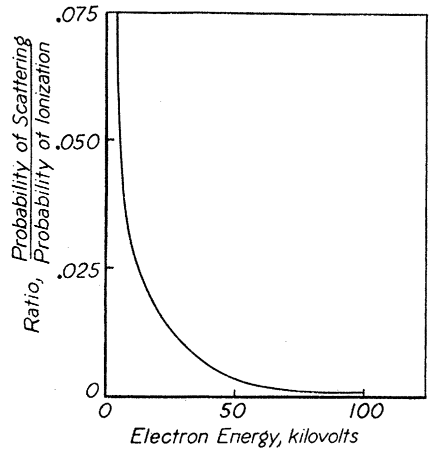


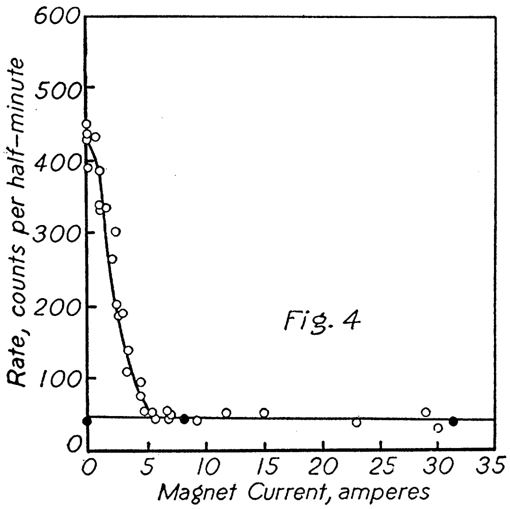
FIG. 3. Relative probabilities of scattering (≥5°) and production of one ion pair in H_2.

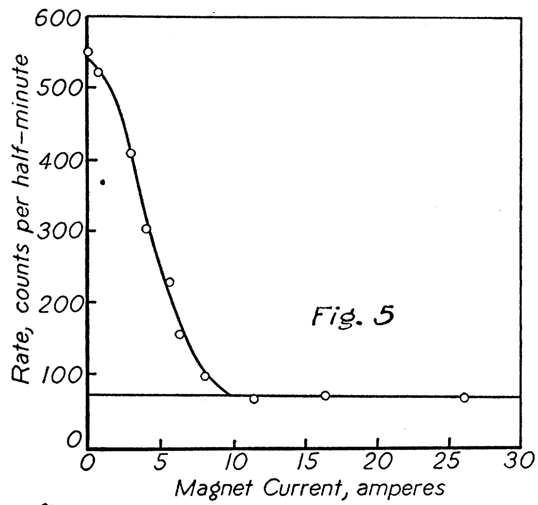
The coil is cooled by a kerosene circulation system. The assembly is of standard type for obtaining the fields used in Zeeman effect work. The pole pieces were removed and only one of the two coils used in any one determination in this work. Both coils were calibrated, however (by bismuth spiral and flip coil measurements), and both used in the course of the work. Maps were made of the fields in the cylindrical holes which showed them to be homogeneous within five percent over a distance of about 20 cm along the axis of the solenoid. The variations along the perpendicular direction in this region were smaller.

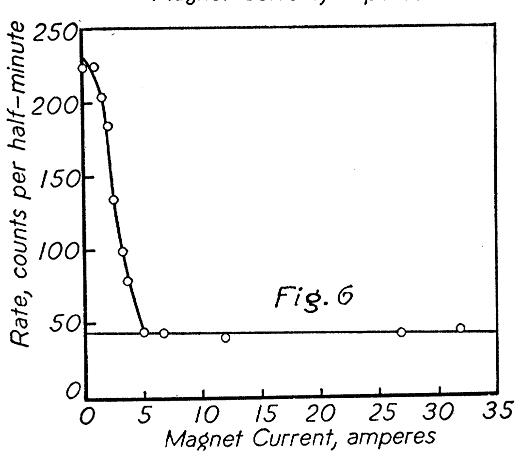
THE COUNTERS

Counters of the type shown in Fig. 1 with the characteristics given in Table I were used. All counters were constructed of copper screen with fourteen wires (0.010″ diameter) to the inch. The counter gas was hydrogen at a pressure of about 4 cm of mercury with a little air, water, or ethyl alcohol vapor introduced to improve the counting characteristics.[4] The sixth column of Table I gives the value of the radius of curvature, ρ, of the electrons which leave the sample surface tangentially and hit the counter surface tangentially, traveling in a plane perpendicular to the direction of the magnetic field. This is half of the shortest distance between the sample and counter surfaces. As given in the table it is computed for thin samples. In the cases where

[4] A. Trost, Zeits f. Physik **105**, 399 (1937).







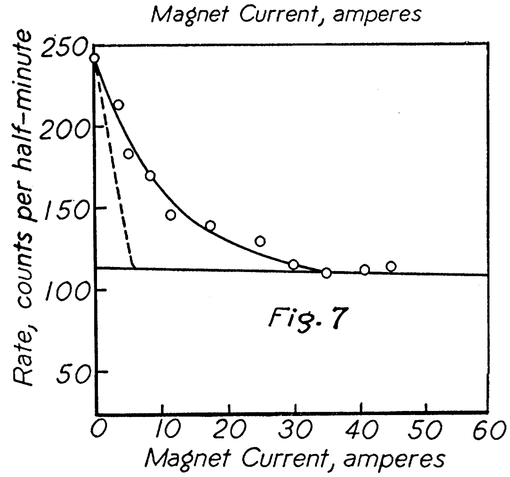


FIG. 4. Beryllium sample in medium counter. (1.85 cm H_2). Open circle, exposed. Solid circle, unexposed. $i_{max}=5.5\pm1$ amperes. $H_{max}=220\pm40$ gauss. $(H\rho)_{max}=410\pm75$. $E_{max}=15{,}000\pm7{,}000$ ev.

FIG. 6. Beryllium sample in medium counter. (5.1 cm H_2 as gas). $\rho=1.85$ cm. $i_{max}=5.0\pm1$ amperes. $H_{max}=200\pm40$ gauss. $(H\rho)_{max}=370\pm75$. $E_{max}=12{,}000\pm6{,}000$ ev.

FIG. 5. Beryllium sample in small counter. (2.8 cm H_2 as gas). $\rho=0.85$ cm. $i_{max}=10\pm2$ amperes. $H_{max}=400\pm$gauss. $(H\rho)_{max}=340\pm70$. $E_{max}=11{,}000\pm5{,}000$ ev.

FIG. 7. Straggling effect. Beryllium sample in medium counter. (4.0 cm A as gas).

the samples were of appreciable thickness, the proper ρ value was computed and used. The uncertainty of this quantity seems to be about 0.5 mm. This information was obtained by running a given spectrum in counters with different ρ values. The uncertainty of measurement of the distance from sample to counter is of that order.

The Straggling Effect and Theoretical Limitations of the Method

The problem of fixing the curvature in a magnetic field and recording a low energy electron is difficult because the probability of scattering is high. The method used in this research requires that the number of collisions along the path from the sample to the counter be as small as is compatible with the production of one or more ion pairs in the counter volume proper. Obvious steps in this direction are the reduction of the distance from sample to counter and the use of hydrogen as counter gas.

Since the essential quantity involved is the ratio of the probability of serious scattering ($\geq 5°$, for example) to the probability of production of an ion pair, this quantity has been

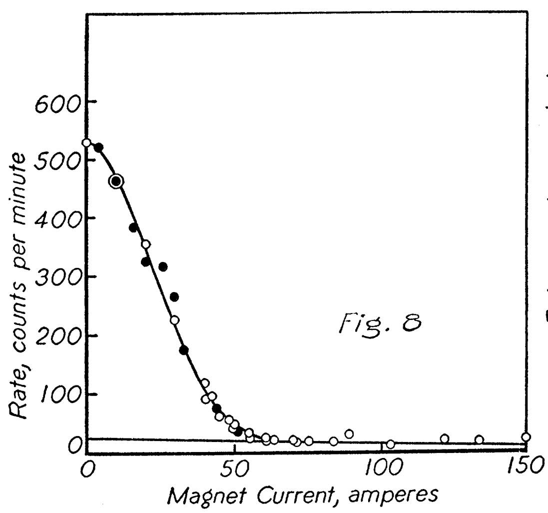


FIG. 8. Radioactive phosphorus in large counter. (3.8 cm H_2+0.2 cm C_2H_5OH). $i_{max}=65\pm5$ amperes. $H_{max}=2600\pm200$. $\rho=2.75$ cm. $(H\rho)_{max}=7150\pm550$. $E_{max}=1.69\pm0.08$ Mev.

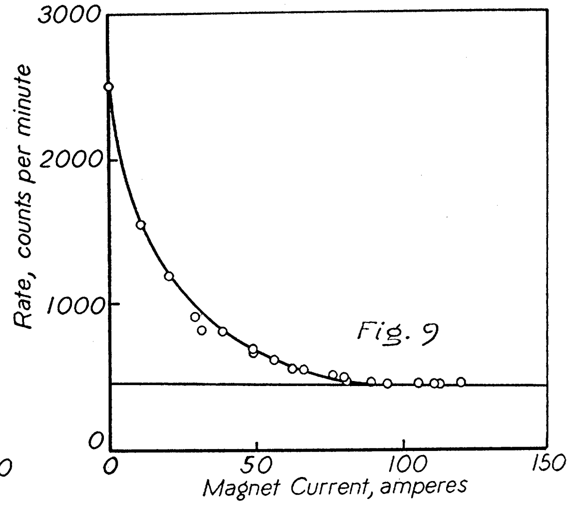


FIG. 9. Mesothorium 2 in medium counter. (Approximately 4 cm H_2+trace of C_2H_5OH). $\rho=1.90$ cm. $i_{max}=88\pm5$ amperes. $H_{max}=3520\pm200$. $(H\rho)_{max}=6680\pm380$. $E_{max}=1.55\pm0.07$ Mev.

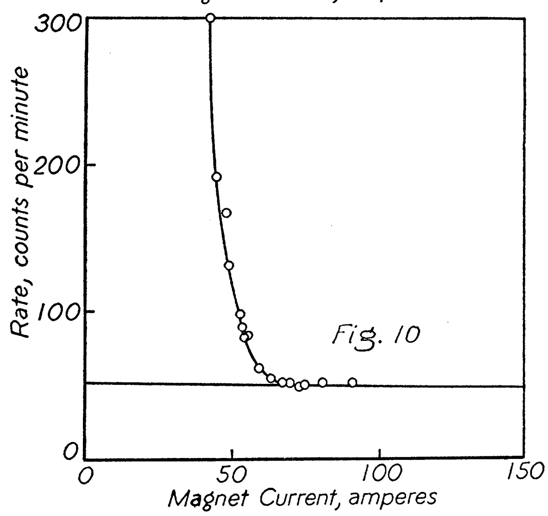


FIG. 10. RaE in medium counter. (Approximately 4 cm H_2+trace of C_2H_5OH). $\rho=1.90$ cm. $i_{max}=68\pm3$ amperes. $H_{max}=2720\pm120$. $(H\rho)_{max}=5150\pm250$. $E_{max}=1.16\pm0.06$ Mev.

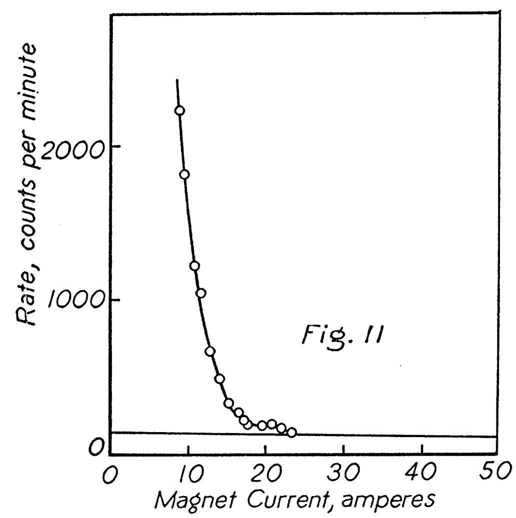


FIG. 11. RaD in small counter. $(H\rho)_{max}=750\pm50$. $E_{max}=48{,}000\pm6000$ ev.

calculated approximately for hydrogen gas from the formula for energy loss given by Bethe,[5] as modified by Bloch's summation method, and the formulae for electronic and nuclear scattering given by Mott.[6] Fig. 3 shows the results as a function of the energy of the electrons. This curve allows an estimate to be made of the energy of the softest beta-radiation for which the method will be valid. The ratio of path length through the counter to the path length before reaching the counter must be at least as large as the ratio of the probability of serious scattering to the probability of ionization. For the small counter used this ratio is about one to twenty; the chief uncertainty arises from the estimation of the path length through the counter. This corresponds to an electron energy of about six

[5] H. Bethe, *Handbuch der Physik* Vol. 24, No. 1 (Berlin, 1933), p. 519.
[6] N. F. Mott, Proc. Roy. Soc. **125**, 222; **126**, 259 (1929).

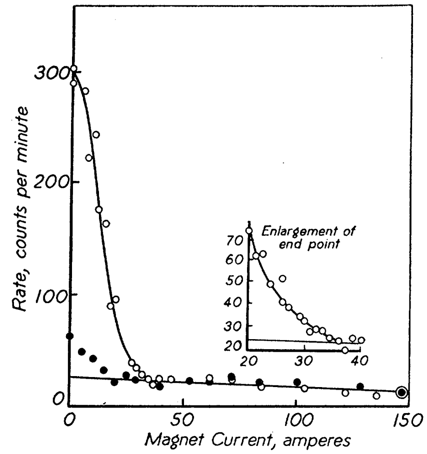


FIG. 12. Rubidium nitrate in small counter. (3.6 cm H_2+0.4 cm A). $\rho=0.9$ cm. Open circles, exposed. Solid circles, unexposed. $i_{max}=36.0\pm3$ amperes. $H_{max}=40.0i_{max}=1440\pm120$. $(H\rho)_{max}=1296\pm110$. $E_{max}=132{,}000\pm20{,}000$ ev.

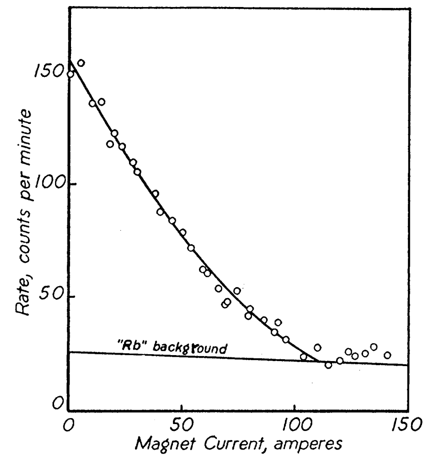


FIG. 13. Potassium nitrate in small counter. (3.6 cm H_2+0.4 cm A). $i_{max}=110\pm10$ amperes. $H_{max}=4400\pm400$. $\rho=0.85$ cm. $(H\rho)_{max}=3740\pm340$. $E_{max}=725{,}000\pm100{,}000$ ev.

kilovolts. However, because an error of a factor of two may have been made in the estimation of the critical ratio and the calculations themselves are somewhat approximate, the limit cannot safely be set lower than about ten kilovolts.

To summarize, with the best conditions of distance (small) from sample cylinder to counter wall, H_2 gas (with a small amount of heavy impurity to improve the counting characteristics) and the lowest pressure of gas insuring formation of a sufficient number of ion pairs in the counter, it should be possible to measure the upper limits of beta-spectra if they are no lower than about ten kilovolts.

These conclusions were tested experimentally by means of a source of soft electrons (kindly loaned by Professor E. M. McMillan of the Physics Department). It was a piece of metallic beryllium which had been bombarded for several months with deuterons in the cyclotron. The radiation consists of negative electrons (determined by placing sloping vanes between the sample cylinder and the counter and reversing the field direction to find the direction of bending of the radiation around the vanes.[1] Runs were made in both the small and medium counters. Of course, the test for absence of the straggling and scattering effect was to find conditions under which the product $(H\rho)$ for the end point would be the same for both counters. Figs. 4, 5 and 6 show this to be true approximately for H_2 gas at pressure of 2 to 5 cm of Hg. Fig. 7 shows that the effect of replacing the hydrogen with argon is the expected smearing and shifting of the end point to higher values. Similar results have been obtained with oxygen and air. The $(H\rho)_{max}$ values show the upper limit to be $13{,}000\pm5000$ electron volts.

The mechanism of counting for the electrons of maximum energy when the field is set for them to hit the edge of the counter may not involve the production of ions in the gas by single collisions so seriously as reflection by the screen into the counter volume or possibly the ejection of electrons from the screen itself. The data afford some evidence for this. It seems difficult to account for the observed counting near the end point at some of the gas pressures used without invoking some mechanism of this sort. Of course such an effect makes the effect of scattering less important.

TABLE II. *Energy of β-rays.*

Element	Energy (this research) (Mev)	Energy (other methods) (Mev)
P^{32}	1.69 ± 0.08	1.69
MsTh 2	1.55 ± 0.07	1.60
RaE	1.16 ± 0.06	1.16
RaD	0.048 ± 0.006	0.046

Checks Against Known Spectra

In order to prove that the upper limits determined by this method are not seriously in error a number of determinations have been made for spectra whose upper limits are obtainable by other methods. Figs. 8, 9 and 10 present typical determinations for P^{32}, MsTh 2, and RaE. Fig. 11 for RaD gives the upper limit of 46 kilovolts found by the standard semi-circular focusing method[7, 8, 9] which is applicable in this low energy range in this case because the radiation is intense. The 46 kilovolt radiation is emitted by conversion of a 47.1-kilovolt gamma-ray. The semi-circular focusing method which uses a photographic plate is inherently a very good apparatus for studying low energy spectra providing there is sufficient intensity available to give a trace on the plate. The whole space between source and plate can be evacuated and the scattering by the gas eliminated. There certainly is little reason to doubt the correctness of its result on the RaD conversion electrons.

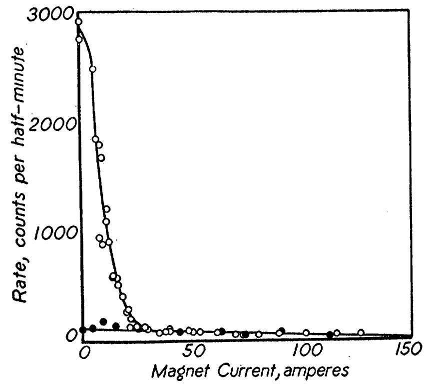


FIG. 15. Radioactive gold in large counter. (4 cm H_2+trace of air). Open circles, exposed. Solid circles, unexposed. $i_{max}=33\pm4$ amperes. $\rho=2.70$ cm. $H_{max}=41.1i+80=1440\pm165$. $(H\rho)_{max}=3890\pm450$. $E_{max}=770{,}000\pm160{,}000$ ev.

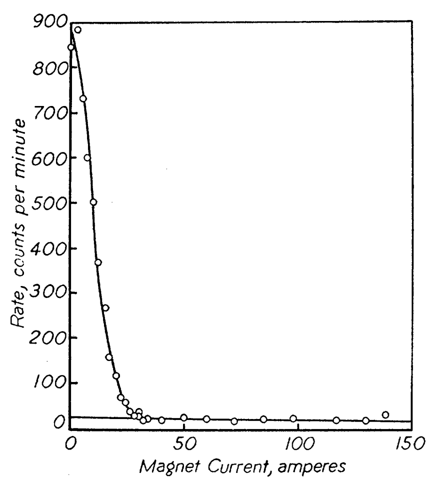


FIG. 14. Radioactive sulfur ($BaSO_4$) in small counter. (3.6 cm H_2+0.4 cm C_2H_5OH). $\rho=0.9$ cm. $i_{max}=32\pm3$ amperes. $H_{max}=40.0i_{max}=1280\pm120$. $(H\rho)_{max}=1152\pm108$. $E_{max}=107{,}000\pm20{,}000$ ev.

Table II presents the results obtained by the present method together with the accepted values from other methods.

RESULTS

Figure 12 gives data obtained for the natural radioactivity of rubidium, due to Rb^{87}. The result is $132{,}000\pm20{,}000$ ev and is considerably lower than the value of 250,000 ev previously used.[10]

Figure 13 shows the data for the natural activity of potassium, due to K^{40}, which give a value of $725{,}000\pm100{,}000$ ev in agreement with the older determinations. No serious attempt was made to make this accurate because of the agreement with the older work and the low intensity of the radiation.

Figure 14 presents data for radioactive sulfur (S^{35}) and Fig. 15 for radioactive gold. The result on radioactive sulfur is new; that for radioactive gold checks the value given by McMillan, Kamen, and Ruben[11] from absorption.

Table III is a summary of results obtained by this method.

TABLE III. *Summary of results.*

ELEMENT	HALF-LIFE	ENERGY OF UPPER LIMIT (Mev)	REFERENCE
Ac	13.4 yr.	0.220 ± 0.05	D. E. Hull, W. F. Libby and W. M. Latimer, J. Am. Chem. Soc. **57**, 1649 (1935)
MsThI	6.7 yr.	0.053 ± 0.005	D. D. Lee and W. F. Libby, following article
S^{35}	88 days	0.107 ± 0.020	This paper*
Au^{198}	2.7 days	0.77 ± 0.16	This paper
Rb^{87}	10^{11} yr.	0.132 ± 0.02	This paper

* The authors are indebted to Professor D. M. Yost of the California Institute of Technology for communicating this value of the half-life of radioactive sulfur checking that given first by E. B. Andersen, Zeits. f. physik. Chemie, **32**, 237 (1936).

7 L. Danysz, Le Radium **9**, 1 (1911).
8 L. Meitner, Zeits. f. Physik **11**, 35 (1922).
9 D. H. Black, Proc. Roy. Soc. **A109**, 166 (1925).
10 O. Klemperer, Proc. Roy. Soc. **148**, 638 (1935).
11 E. McMillan, M. Kamen and S. Ruben, Phys. Rev. **52**, 375 (1937).