Some Measurements of Gamma-Ray Energies

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To examine the possibility that some radio-isotopes which have been considered to decay by K-capture do in part decay with the emission of very slow positrons careful measurements of the γ -rays emitted by two of them have been made. Preliminary measurements were made of the F and I lines of ThB as a check on the spectrometer used. Also the annihilation radiation line of Cu⁶⁴ was measured to establish its shape and position. In the course of this, good evidence was found for radiation resulting from annihilation while the positron is in flight. The weak high energy gamma-ray previously reported was found and its energy determined to be 1.34 ± 0.01 Mev. The γ -radiation of both Be⁷ and Cr⁵¹ consists of a single line at 485 ± 5 kev and 320 ± 5 kev, respectively. No evidence of annihilation radiation was found in either case.

1. INTRODUCTION

'HIS work began because of the necessity of examining experimentally the γ -radiaion of those radioisotopes which decay bv K-capture. If one plots the number of these isotopes as a function of the energy of γ -rays as recorded in the literature, one finds a clustering of energies in the neighborhood of the positron annihilation energies, 0.5 and 1 Mev. This suggests that in many cases, instead of actually disintegrating by K-capture, there might be the emission of slow positrons which could escape detection. These positrons would have a good chance, if they were slow enough, to be annihilated in the K-shell of the atom whose nucleus they had just left. This would leave a vacancy in the K-shell and, in the course of its being filled, characteristic K-radiation would be emitted. Thus the detection of K-radiation does not suffice to establish K-capture as the process whereby a particular radioisotope decays. Evidence for the emission of slow positrons might be obtained if annihilation radiation were observed in "K-capture" isotopes. Indeed, because of the proximity of a nucleus to the K-shell one should expect to find examples of single quantum annihilation perhaps somewhat more frequently than when fast positrons are annihilated. These considerations suggested that accurate determinations of γ -ray energies from isotopes which decay by K-capture might reveal such annihilation radiation.

The measurements described here include only two *K*-capture isotopes. Two other measurements were made for calibration purposes. The measurement of one of the internal conversion lines of Th B served to check the calibration of the magnetic field of the spectrometer, and those on Cu^{64} provided a reference line in the neighborhood of 0.5 Mev for comparison with other annihilation radiation lines, if such were found.

2. EXPERIMENTAL DETAILS

The magnet used for this work consists of two Armco iron forgings with a total weight of about two tons. The pole faces are circular, about 16" in diameter; the gap is 4". In this work the magnetizing current, which passed through coils of copper strip, was supplied by a bank of automobile storage batteries. No regulation other than manual was employed. Despite its somewhat massive construction and the comparatively low magnetic fields used, there was enough hysteresis in the magnet so that the same current did not give the same magnetic field. Consequently current measurements were used only to establish the increments between successive fields; the fields were measured with a 6000-turn flip coil and ballistic galvanometer. The calibration of the flip coil was made against two standard flip coils of large diameter (3'') which had been very carefully made. The area-turns of these two coils were compared in a fixed magnetic field (the remanent field of the above magnet was used) and found to be the same to one part in 1000. By mounting the flip-coil, which was used in these measurements, concentrically and consecutively in each of the standard coils it was possible to obtain an equivalent area-turn for it. The ballistic galvanometer was calibrated against a home-made mutual inductance which, in turn, was standardized against a Leeds and Northrup mutual inductance, using the value given by the Bureau of Standards who had recently calibrated it.

Two vacuum chambers were used in this work. The first of these was a modification of a chamber which had been used for some earlier work in this Department. The modification consisted in installing lead shielding between source and counter, arranging an easily accessible source mounting, and installing slits and a Geiger-Müller counter. A radius of curvature of 13 cm was chosen for the first chamber. This was checked in the actual instrument to about two parts in 1000. Because of the diameter of the pole faces the beam of electrons is always in a very nearly uniform field. Steel lids reduced the total gap to $2\frac{1}{4}''$ of which $1\frac{3}{4}''$ was the "vacuum" gap. There were only two slits: one at the half-way point 25.4 mm wide and the other immediately before the counter window 1 mm wide. The first defining slit was aluminum-clad lead. The counter was entirely in the main



FIG. 1. The F- and I-lines of ThB. The region between these two lines and on either side was not searched carefully. There are numerous small lines in these regions. The width of the F-line corresponds to a resolution of 0.6 percent. The energy values deduced from the extrapolation of the high side of the line to the background are in good agreement with previous determinations.

vacuum, but could be filled from outside. It consisted of a block of copper with a $\frac{1}{4}$ -in. hole drilled in it. To its ends were attached metal-toglass seals and between these was stretched the central wire of about 0.003-in. tungsten. In the side of the copper block and perpendicular to the axis a hole about 3×5 mm was cut and over this a thin window was clamped. In this work all windows were 0.001-in. aluminum made vacuumtight by being clamped against a 0.007-in. indium gasket. Such a thick window cannot pass electrons whose energy is much below 100 kev.

The second chamber differs from the first in being more carefully designed and constructed. The source is inserted through an air-lock and all of the slits can readily be removed so that their aperture can be changed, if desired. The counter in the second chamber is of the endwindow type and has a much larger window, $1\frac{1}{4}'' \times \frac{5}{16}''$. The arrangement of the slits, the length of the magnetic air gap, and the flip coil are all identical in the two chambers. The radius of curvature of the electrons was increased to 14 cm and this figure is known with the same accuracy as in the previous case. A minor feature of the second chamber is that everything including the cover lids is positioned by metal-tometal seats, through the use of radial O-ring seals. In general the vacuum used in this work was around 10⁻⁵ mm of mercury.

For the measurement of γ -ray energies the following procedures were used. With those substances which have particle radiation it is necessary that they be absorbed. The γ -rays are then measured by determining the energies of the secondary electrons which they eject from suitable radiators. In this paper we shall mostly be concerned with the photoelectrons ejected from a thin lamina of uranium metal. The radioactive material is placed in a small copper "boat" whose bottom is $\frac{1}{4}$ " $\times 1$ ". The long dimension is mounted parallel to the magnetic field. The interior is hollowed out to form a cavity $\frac{1}{8}'' \times \frac{7}{8}''$. The depth of this hollowing is such as to give a thickness of $\frac{1}{16}''$ to absorb the nuclear β -rays. This is quite adequate for most radioactive materials. To the bottom of this boat is glued a piece of uranium foil about 50-mg/sq. cm thick. When there is no particle radiation the radioactive substance is sealed in a thin glass tube

and mounted in a skeleton Lucite frame so that the glass tube is in contact with a similar piece of uranium foil.

Compton electrons are ejected from the material of the boat, from the source material and from the uranium foil. They are always present as a background. In addition photoelectrons from the uranium form sharp spires on this Compton distribution. From the known facts about the absorption and straggling of homogeneous electrons as they pass through matter, it is clear that the energy corresponding to one of the photoelectron lines is obtained by extrapolating the high energy side of such a line to the background. Usually the lines are so steep on their high energy side that this extrapolation presents no problem which might otherwise arise if there were uncertainty as to the location of this background. In all cases which have been examined both the K and the L photoelectrons are observed, and the γ -ray energies deduced from both have been found to be identical within experimental error. The γ -ray energies quoted will be those corresponding to the K-line. In all of the curves shown here irregularities on the low energy side of an intense peak are noticed. Because of their systematic regularity in all data taken with the first chamber (Cu⁶⁴, Be⁷) they are thought to be caused by scattering. For the binding energy of the K-shell of uranium the value 116.4 kev was used; for the L-shell, 22 kev.

No special care was taken in preparing the counters except to have them very clean. The central wire was either heated to incandescence under a good vacuum or else was cleaned electrolytically. Various conventional quenching vapors were used without there being much noticable difference among them. Lately this laboratory has been experiencing good results with ethylene as a quenching gas. Good evacuation with a diffusion pump appears to be desirable.

The counter was connected to a scaling circuit (Instrument Development Laboratories Model 161) with a scale of 256. This had been modified so that scales of 8, 16, 32, 64, 128 or 256 could be used. No quenching circuit was used. An electric timer and a message register completed the recording equipment. In general counts were taken for a fixed length of time, so that the



FIG. 2. The momentum distribution of photoelectrons (curve P) from a 50 mg/cm² uranium radiator due to the radiations from Cu⁴⁴. The lower curve, marked C, is the Compton distribution from the same γ -rays. The insert is a 100-fold magnification of the high energy end of both distributions. In the insert are seen the K- and L-photoelectron peaks from the nuclear γ -ray at 1.34 Mev. The arrow marks the position of the expected end of the Compton distribution if there were no annihilation radiation from moving positrons.

statistical accuracy is better at high counting rates than at low.

3. THORIUM-B

The original purpose of measurements on ThB was to check the accuracy of the magnetic field calibration, which was described earlier. The source was thorium active deposit on a 0.00025'' thick aluminum foil 1 mm wide. Since the gammarays of ThB are strongly internally converted, no radiator was needed to get the sharp peaks shown in Fig. 1. The taller of the two peaks is, in Ellis' notation, the *F*-line while the smaller is the *I*-line. These have been measured probably most accurately by Ellis¹ and Siegbahn² who give, respectively, 1385.8 and 1383.8 for the $H\rho$ of the *F*-line and 1749.6 and 1751.0 for that of the *I*-line.

The present work makes no claims to accuracy of the same order as that of the highly precise measurements to which reference has been made. Our values of 1395 $H\rho$ for the *F*-line and about 1770 $H\rho$ for the *I*-line merely indicates the differences between our flip-coil calibration and this more precise calibration based on these two lines. This difference of about 0.7 percent for

¹C. D. Ellis, Proc. Roy. Soc. A138, 318 (1932).

² K. Siegbahn, Arkiv f. Mst. Astr. och Fysik, **30A**, No. 20 (1944).

the K-line is corrected for in the data to be discussed later. The resolution of the spectrograph can be estimated by the half width of the F-line and is found to be about 0.6 percent. The calculated resolution is about 1 percent and this difference has been a matter of some concern to us. No use is to be made of the resolution in this paper, so it is only interesting in understanding the operation of a spectrometer. A reasonable explanation is that the extreme outside electron path is through a somewhat weaker magnetic field which, for monochromatic electrons, would tend to throw more of them toward the head of the line, thus making it appear narrower. This same effect has been noticed in another spectrometer in this laboratory.

4. COPPER-64

A source of Cu⁴⁴ was made by bombarding a copper plate with 11-Mev deuterons in the Washington University cyclotron. Some shavings were taken from this plate and, after they had aged for 4–5 hours to let the short-lived contaminating activities die off, they were placed in the copper boat with a uranium radiator on its bottom. The resulting distribution is shown in Fig. 2. Both the K- and L-peaks corresponding to the half-million electron volt annihilation radiation are observed on the curve marked P. The corrected H_{ρ} value as here observed is 2480 corresponding to an energy of 0.510 Mev. This establishes the position and appearance of an annihilation line in our particular spectrometer.

In the insert is shown a 100-fold magnification



FIG. 3. The momentum distribution of photoelectrons from the same uranium radiator due to the γ -radiation from Be⁷. There appears to be only one strong γ -ray from this element. The energy of it found here is 485 kev.

of the high energy end of the secondary electron spectrum, showing the K-line at 5550 $H\rho$ corresponding to 1.34 ± 0.01 Mev. This line has been observed by several authors.^{3,4} Our value is seen to be in good agreement with the figure 1.35 Mev obtained by Deutsch. No attempt has been made here to estimate the intensity of this line relative to that of the annihilation line. Also shown in Fig. 2 is the distribution curve of the Compton electrons knocked out of the copper boat. This is the lower curve marked C in both the main part of the figure and the insert. It is lower and very broad because about half of the Compton electrons are contributed by the uranium radiator and because the thickness of the copper spreads the Compton distribution by straggling. The arrow arising from the axis of abscissae indicates the position of the upper limit of the Compton electrons if the annihilation radiation had a sharp energy of 0.511 Mev. That the distribution extends beyond this point is indication, as Deutsch has also shown,3 of the existence of radiation caused by the positrons being annihilated in flight. As suggested by the curve on the insert to Fig. 2 this Compton distribution seems to extend at least up to $H\rho$ 5250. corresponding to an energy of 1.35 Mev, which is the maximum energy expected theoretically. The data are not sufficiently good statistically to argue that the extension beyond this point is real.

5. BERYLLIUM-7

Beryllium-7, which decays with a half-life of 53 days, can easily be made by bombarding lithium with deuterons according to the process: $\text{Li}^6(d,n)\text{Be}^7$. Such a source was prepared by bombarding pure lithium metal for 10,000 micro-ampere hours with 11 Mev deuterons in the cyclotron. The beryllium was chemically separated, following a procedure suggested by Dr. Gerhard Friedlander. The resulting activity was sealed in a thin-walled glass tube and placed in immediate contact with a thin uranium radiator of the same size as had been used in our other measurements. The energy distribution of the photoelectrons ejected from the uranium is shown in Fig. 3. The corrected $H\rho$ of the K-peak

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³ M. Deutsch, Phys. Rev. 72, 729 (1947).

⁴ Bradt, Gugelot, Huber, Medicus, Preiswerk, Scherrer, Steffen, Helv. Phys. Acta **19**, 219 (1946).

is 2380 corresponding to a gamma-ray energy of 0.485 ± 0.005 Mev.

Previous work on this γ -ray has followed two lines of attack, as summarized by Zlotowski and Williams⁵ and by Siegbahn.⁶ One of these has been, of course, the direct measurement of the γ -ray energy. The other has been to study the excited states of Li7 formed in the reactions: $Li^{6}(d,p)Li^{7}$, $Be^{9}(d,\alpha)Li^{7}$, $B^{10}(n,\alpha)Li^{7}$. Results obtained to date have not shown very good agreement with each other. In general the excitation energy of Li⁷ has been somewhat higher than the measured γ -ray energy. Thus the following values (quoted without limits of error) have been found for the energy of the excited state of Li⁷: (see reference 5 for bibliography) 455, 495, 494, 470, 480 kev. These are to be contrasted with the following γ -ray energies : 425, 459 kev. Zlotowski and Williams, using Bothe's coincidence method. found a value of 485 kev for this γ -ray, while Siegbahn, using a β -spectrometer, finds a value of 453 kev. Both of these authors had, as compared to ourselves, quite weak sources and, as suggested by Siegbahn, it would be possible to deduce appreciably different values from the data of Zlotowski and Williams. To get the value he gives, Siegbahn seems to have chosen the top of the K-line peak. Since there is some debate as to what is the proper point on a photoelectron line to use in deducing the γ -ray energy and since this point may differ for different types of spectrometers, it is possible that Siegbahn's data could be interpreted to give a higher energy. As previously stated, on the basis of measurements of the straggling of homogeneous electrons, we subscribe to the method of extrapolating the high energy side of a photoelectron peak to get its energy, at least in the case of a 180° spectrometer.

The value which has been found here seems to be in good agreement with the measurements on the excitation energy of Li⁷. The deviation of this value from that of the 0.511 Mev of annihilation radiation is well outside of experimental error so we must conclude that there is no evidence here for the process discussed at the beginning of this paper.

⁶ I. Zlotowski and J. H. Williams, Phys. Rev. **62**, 29 (1942). ⁶ K. Siegbahn, Arkiv f. Mat., Astr. o. Fysik **34B**, No. 6 (1946).



FIG. 4. The momentum distribution of photoelectrons from the same uranium radiator due to the γ -radiation from Cr⁵¹. There appears to be only one strong γ -ray from this element. The energy of it found here is 320 kev.

6. CHROMIUM-51

A radioactive substance whose half-life is about 26.5 days and which has been identified as Cr^{51} was discovered and studied by Walke, Thomson and Holt.⁷ These authors ascertained that it decays by *K*-capture and, by absorption, measured γ -ray energies of 0.5 and 1.0 Mev. They also report that the 1.0-Mev γ -ray is internally converted to the extent of 0.1 percent. Later Miller and Curtiss⁸ listed the $Cr^{51} \gamma$ -ray as 0.32 Mev. This appeared to be a likely substance to use to examine the notion of slow positron annihilation.

A sample of Cr⁵¹ was obtained from Oak Ridge and carefully purified chemically following a procedure suggested by W. H. Burgus. The purified sample was divided into several parts and the largest of these was mounted in a thinwalled lucite tube close to a uranium radiator as in the case of Be⁷. Figure 4 shows the momentum distribution of the photoelectrons from this radiator. It will be noticed that there appears to be only one γ -ray as is evidenced by the K- and L-peaks. The corrected H_{ρ} for the upper limit of the K-line is 1670, corresponding to a gamma-ray of 320 ± 5 kev. This is in very good agreement with the value found by Miller and Curtiss. The region where one would expect to find photoelectrons from a 1.0-Mev gamma-ray

⁷ H. Walke, F. C. Thomson and J. Holt, Phys. Rev. 57, 171 (1940). ⁸ L. C. Miller and L. F. Curtiss, Phys. Rev. 70, 983 (1946).

was very carefully searched but nothing above background was detected.

Another sample of the same material was mounted on a thin foil and used as a β -ray source to look for the conversion electrons reported by Walke, in an attempt to find some positron emission. In both cases no detectable particle radiation was found of sufficient energy to penetrate our counter window. These results indicate that Cr^{51} decays simply by K-capture with the emission of one soft γ -ray.

7. CONCLUSIONS

In the two K-capture elements tested there is no evidence of annihilation radiation resulting from the emission of very slow positrons. In our experiments no positron of energy lower than about 100 kev could enter our counter. It is believed that it would be profitable to pursue this problem further, however, since in many cases the evidence for \bar{K} -capture would not exclude the possibility of slow positron emission.

We take great pleasure in thanking Mr. J. E. Robinson for the very able assistance he rendered in the many long "runs" which were involved in this work and its preliminaries. Our grateful thanks are also due to Dr. Lin-Sheng Tsai, to Dr. Gerhard Friedlander, and to Mr. W. H. Burgus for their assistance and counsel on matters chemical.

This work has been supported in part by a grant from the Research Corporation to one of us (FNDK) and by the Office of Naval Research under Contract N6ori-117.

PHYSICAL REVIEW

VOLUME 74. NUMBER 6

SEPTEMBER 15, 1948

The Beta- and Gamma-Rays of Rb⁸⁶

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The beta- and gamma-rays of Rb⁸⁶ (19.5 d) have been measured in a magnetic lens spectrometer. Measurement of the energies of photoelectrons produced in a lead radiator shows one line corresponding to a gamma-ray of energy 1.081 Mev. The beta-ray spectrum is resolvable into two groups with end point energies of 1.822 and 0.716 Mev.

I. INTRODUCTION

WO rubidium isotopes of 18 minutes and 19.5-day half-lives were reported by Snell.¹ Later Helmholz, Pecher, and Stout² showed that the activity of 19.5 day half-life must be attributed to Rb⁸⁶. They measured the beta-ray end point by absorption in aluminum and obtained a value of 1.56 ± 0.05 Mev. Haggstrom³ measured the beta-ray spectrum of this isotope in a solenoid type magnetic analyzer. She made a Fermi plot of her data and found a beta-ray end point of 1.60 ± 0.03 Mev. The Fermi plot was not a straight line at the low energy end, but she attributed this to the fact that Rb⁸⁶ belongs to the second forbidden class.

Preliminary work in this laboratory showed that a low intensity gamma-ray accompanies this activity. A rough estimate of the energy of the gamma-ray was obtained by measuring the coincidence absorption of Compton electrons produced in an aluminum radiator. The value obtained for the energy of this gamma-ray was approximately 1 Mev. In order to determine the disintegration scheme, the authors decided to investigate the spectrum in a magnetic lens spectrometer. The spectrometer has been described elsewhere.

II. PURIFICATION OF SOURCE MATERIAL

Strong sources of Rb⁸⁶ were obtained from Oak Ridge. The main impurity in the source

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¹ A. H. Snell, Phys. Rev. **52**, 1007 (1937). ² A. C. Helmholz, C. Pecher, and P. R. Stout, Phys. Rev. **59**, 902 (1941).

³ E. Haggstrom, Phys. Rev. 62, 144 (1942).