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The Beta- and Gamma-Radiations from Copper^{64} and Europium^{152}

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The beta- and gamma-radiations from Cu^{64} and Eu^{152} have been studied with the aid of a magnetic spectrometer of high resolution. Copper emits both positrons and electrons with a maximum energy of 0.659±0.003 and 0.578±0.003 Mev, respectively. The effect on the shape of the spectra of scattering within the source was investigated. The spectra obtained with an extremely thin source were found to contain fewer low energy particles than those obtained with a thicker source. The thin source results in much better agreement with the original Fermi theory of beta-decay than with the later modification introduced by Konopinski and Uhlenbeck. As the source is made thicker there is a gradual change in the shape of the spectra which eventually brings about better agreement with the K-U theory than with the Fermi theory. Eu emits electrons with a maximum energy of 1.885±0.012 Mev and several gamma-rays. The energies of three of the gamma-rays have been determined as 0.123±0.001, 0.163±0.001 and 0.725±0.003 Mev.

INTRODUCTION

NO THEORETICAL treatment of the continuous energy distribution of the beta-particles emitted by radioactive materials has been entirely satisfactory. The original Fermi theory[1] apparently predicts fewer low energy beta-particles than are found experimentally. Konopinski and Uhlenbeck[2] pointed out that several modifications of the theory are possible. One modification in particular yields a distribution which agreed quite well over the major portion of the spectrum with the data available at that time. However a serious discrepancy between theory and experiment appears near the upper limit of the spectrum. It is rather improbable that this can be due to experimental error. On the other hand, it is quite possible that the Fermi theory can be reconciled to experiment because the discrepancy is in the low energy region of the spectrum where experimental errors are likely to be large.

A magnetic spectrometer of high resolution has been designed and constructed with especial attention to the reduction of the experimental error in the measurement of a spectrum. Scattering from the walls of the vacuum chamber has been reduced to a negligible value and corrections for the efficiency of the Geiger-Müller counter and for the absorption in the counter window have been determined. A detailed description of the spectrometer and a discussion of the corrections is being published.

COPPER

When copper is bombarded with deuterons an intense radioactivity which decays with a 12.8

[1] E. Fermi, Zeits. f. Physik **88**, 161 (1934).
[2] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **48**, 7 (1935).

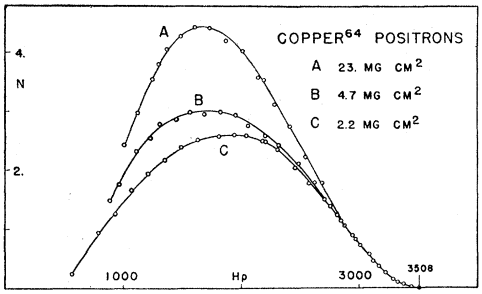

FIG. 1. Positron spectrum of copper from sources of different thickness.

hour half-life is formed. Van Voorhis[3] studied this reaction and found that the activity is due to Cu^{64}, which decays with the emission of either an electron to $zinc^{64}$ or a positron to $nickel^{64}$. He also measured the energy distribution of the particles with a cloud chamber and reported upper limits of 0.79 and 0.83 Mev for the positron and electron spectra, respectively. Copper was chosen for further study with the magnetic spectrometer because it can be activated quite strongly, it has a conveniently long half-life and it is one of the few elements which emits both positrons and electrons.

A thin copper foil was bombarded in the cyclotron for several hours. A small portion of the activated foil was dissolved in nitric acid and electroplated onto a thin copper strip 3 mm wide. This strip was then mounted on an aluminum frame which holds it in place in the spectrometer. The thickness of the source was about 7 mg/cm^2 and that of the copper support about 16 mg/cm^2. Intensity measurements were made at intervals of approximately 100 $H\rho$. Each reading was extended over a sufficient number of counts to obtain a statistical accuracy of about 1–2 percent for the major portion of the spectrum and 10–20 percent near the upper limit.

In order to determine whether the method of preparing the source has any effect on the shape of the spectrum a second source was prepared as follows: Scrapings from an activated copper target were dissolved in nitric acid and precipitated as $Cu(OH)_2$. This was converted to CuO by boiling and then filtered. The filtering device was arranged to deposit the precipitate over a 3×20 mm area. The filter paper was cut along the edges of the source and the strip mounted as before. This source weighed 12.2 mg/cm^2 of which 7.5 mg/cm^2 was due to the filter paper backing. The spectra obtained with this source were quite different in shape from the first. Neither the electron nor the positron curves could be made to coincide by any multiplicative constant. Consequently a third and thinner source was prepared in the same manner as the second except that the filter paper backing was replaced by thin tissue paper. The CuO source weighed only 2.2 mg/cm^2 and the paper support 1.8 mg/cm^2. The shapes of the spectra from this source were again unlike those obtained with the other sources. Fig. 1 shows the positron spectra obtained with the three sources. The curves have been normalized at about 2900 $H\rho$.

The half-life of each source was checked at several points and was found to be 12.8 hours in all the energy regions of all spectra. Thus there is no appreciable amount of impurity in the copper which might account for the difference in shape. The absorption in the source will cause a general shift of the spectrum towards lower energy but this could not introduce as much distortion in the spectra as is observed. The only other reasonable assumption is that the effect is due to scattering within the source and from its support. If the energy of the particle is not changed appreciably by the scattering process, as one would expect in the case of electrons scattered in a thin layer of material, the shape of the spectrum should be altered only by the back scattering from the support. Scattering within the source itself should not affect the shape since as many particles would be scattered out of the selected solid angle as are scattered into it. Back scattering from the support will increase

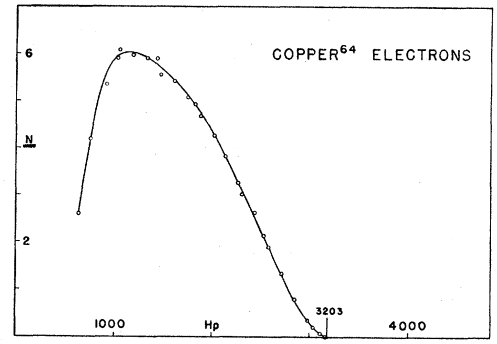

FIG. 2. Electron spectrum from thinnest copper source.

[3] S. N. Van Voorhis, Phys. Rev. 50, 895 (1936).

the intensity in the forward direction by a greater percentage for low than for high energy regions. This could account for the distortion of curve A, Fig. 1, but not for the difference between curves B and C. The latter must be due mostly to the difference in thickness of the two sources, since these supports were made from a material of low atomic number and should produce negligible back scattering. The complete scattering phenomenon is apparently more complicated. Its effect on the shape of the spectrum is greater than has generally been supposed. Curve C represents the best approximation to the true shape of the positron spectrum of Cu^{64}. The observed upper limit corrected for the resolution of the spectrometer is 0.659 ± 0.003 Mev.

The distortion of the electron spectrum is almost identical to that of the positron spectrum. The same normalizing factors are necessary to make the curves coincide near the upper limit. Fig. 2 shows the electron spectrum obtained with the thinnest source. The observed upper limit is 0.578 ± 0.003 Mev. There is no positive indication of a gamma-ray associated with Cu^{64} other than the annihilation radiation. It is possible that a photoelectric conversion group of electrons may fall between two measured points of the electron spectrum and hence escape detection. However this is not very probable in view of the large number of measurements. The difference in the maximum energy of the positron and electron spectra is 81 kev. Thus the isobars Zn^{64} and Ni^{64} should differ in mass by 8.7×10^{-5} mass unit.

The distribution in momentum of beta-particles is given by the Fermi theory as:

$$N(\eta)d\eta = F(Z, \eta)(W_0 - W)^2\eta^2 d\eta \qquad (1)$$

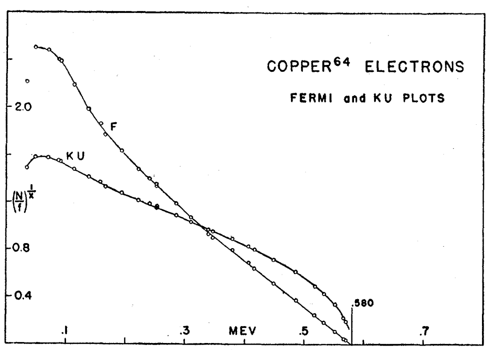

FIG. 3. Fermi and K-U plots of electrons from thinnest copper source.

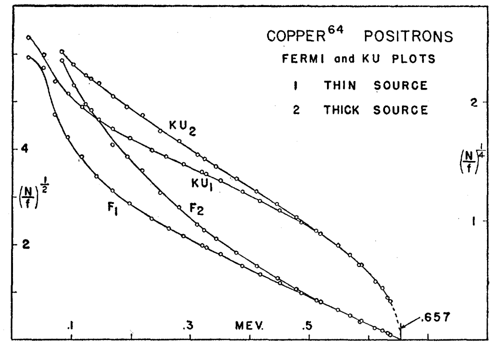

FIG. 4. Fermi and K-U plots of positrons from thick and thin copper source.

where W is the energy and η the momentum of the beta-particles and W_0 is the total energy available in a disintegration. $F(Z, \eta)$ is a function of η and the nuclear charge only. The modification of Konopinski and Uhlenbeck changes the factor $(W_0 - W)^2$ to $(W_0 - W)^4$. Kurie, Richardson and Paxton[4] have pointed out that, if $(N/\eta^2 F)^{1/k}$ is plotted against energy, a straight line should result if the Fermi theory ($k=2$) or the K-U theory ($k=4$) is valid. The intercept of the line on the energy axis is W_0. Furthermore they have shown that $F(Z, \eta)$ can be approximated by

$$F(Z, \eta) \backsimeq \lambda/(1 - e^{-\lambda}) \qquad (2)$$

where $\lambda = 2\pi Z W / 137\eta$ for values of Z up to about 29. Fig. 3 shows these plots of the electron spectrum obtained from the thinnest copper source. The abscissa is the kinetic energy of the electrons in Mev $[= 0.51(W-1)]$. The high energy portion of the Fermi curve is exceedingly straight and intercepts the axis at 0.580 Mev, which is very nearly the observed upper limit. The K-U curve is quite straight at low energies but is definitely bad near the upper limit. If the straight portion of the K-U curve is extrapolated to the energy axis, as has often been done to obtain the upper limit, the intercept is 1.38 times the observed end point.

Figure 4 shows the Fermi and K-U plots of the positrons from copper. Curves 1 and 2 are constructed from the data obtained with the thinnest source and the thickest source, respectively. The intercept of the Fermi plot is 0.657 Mev. The Fermi plot for the thick source diverges from a

[4] Kurie, Richardson and Paxton, Phys. Rev. **49**, 368 (1936).

straight line at a much higher energy than that of the thin source. If one disregards the discrepancy near the upper limit the K-U plot for the thick source is almost a straight line. This probably explains in part why many observers have obtained substantial agreement with the K-U theory. However, the spectrum obtained with the thin source, which is undoubtedly more nearly the true spectrum, definitely disproves the validity of the K-U theory. A thick source tends to favor the K-U theory hence any remaining distortion due to the thickness of the source adds more weight to the disproof of the theory.

The departure from linearity of curve F_1, Fig. 4, and curve F, Fig. 3, in the region 0.2 to 0.35 Mev may still be due to the thickness of the source. It is difficult to make a source of sufficient intensity much thinner than 1 or 2 mg/cm². It might be possible to obtain the spectrum of a source of infinitesimal thickness by extrapolating a series of spectra obtained from sources of different thicknesses. In order to do this one would need to know the relative source strengths. This information is not available for the present data. A thorough theoretical treatment of this scattering phenomenon would certainly be of great advantage.

The discrepancy below 0.2 Mev, however, is rather large to be explained by the thickness of the source. There are several possible explanations which may account for the large number of low energy electrons. The experimental accuracy becomes progressively worse at lower energies (1 percent at 0.2 Mev and 10 percent at 0.07 Mev). Bethe *et al.*[5] have recently suggested that each observed spectrum may consist of several superimposed spectra of the same half-life but of different maximum energy. This would occur if the radioactive nucleus could decay to one of several energy states of the product nucleus. Thus, although the Fermi plot of the composite spectrum is not a straight line, that of each component might be. The composite Fermi plot may be resolved into its components by extrapolating the straight portion of the curve to zero energy and subtracting the number of counts it predicts at each point from the corresponding total number of counts. If the Fermi plot of this residue is not a straight line the process may be repeated. One would expect to find gamma-rays emitted during the transitions of the nuclei from the excited states to the ground state. Bethe has found several cases in which a spectrum could be resolved reasonably well into two or more components and in which there are associated gamma-rays of approximately the correct energies. It was found impossible to resolve the present Fermi curves into two or more linear components. This does not rule out the possibility of a composite spectrum since the distortion which probably still remains below 0.3 Mev would make the low energy components nonlinear.

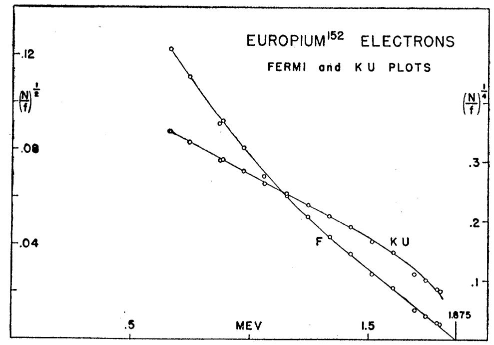

FIG. 5. Fermi and K-U plots of electrons from europium source.

It can readily be seen by a comparison of curve F, Fig. 3, and curve F_1, Fig. 4, that the departure from the Fermi theory at low energies is greater for the positron spectrum than for the electron spectrum. Since the scattering of positrons has been found to be essentially the same as that of electrons it would be impossible to reconcile both spectra to the Fermi theory by a scattering phenomenon. Furthermore it might be well to point out that, if either spectrum is composite, the difference in energy of the two end points would be very nearly 0.5 Mev and the expected gamma-radiation would be concealed by the annihilation radiation. This difference between the positron and electron spectra leads one to suspect that the Fermi theory is not adequate at low energies and in particular does not take into account correctly the effect of the nuclear charge.

EUROPIUM

When europium is exposed to slow neutrons a strong electron radioactivity of 9.2 hours half-

[5] Bethe, Hoyle and Peierls, Nature **143**, 200 (1939).

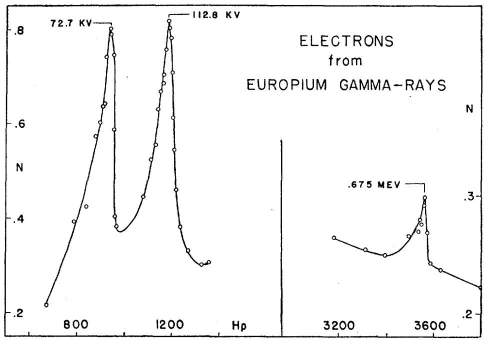

FIG. 6. Secondary electron groups from europium gamma-rays.

life is formed. Pool[6] has excited this activity by fast neutron bombardment of Eu and has established that it is in the Eu isotope of mass 152. The upper limit of the spectrum was first reported as 2.0 Mev by Sugden,[7] who also detected the presence of gamma-radiation. Hevesy and Levi[8] also studied this reaction and noticed that the slow neutron absorption cross section was larger than could be accounted for by the activity that was produced. They concluded that the resonance absorption of slow neurtrons formed a stable as well as a radioactive isotope. D. W. Stewart, formerly of the Chemistry Department of the University of Michigan, detected in Eu a slow neutron induced activity of long half-life (>two years). He assigned this activity to the isotope of mass 154 and suggested that it may account for the observations of Hevesy and Levy. His work has not yet been published. He also measured the spectrum of the 9.2-hour period. Absorption measurements indicated a beta-spectrum with an upper limit of 1.9 Mev and strong gamma-radiation of approximately 80 kev. The beta-spectrum measured with a cloud chamber had an upper limit of 1.83 Mev. The shape of the spectrum was determined roughly with a magnetic spectrometer by Alichanow *et al.*[9] They observed an upper limit of 2.00 Mev. The spectrum which they obtained from a thick source contained relatively fewer low energy electrons than that from a thin source. This is in definite disagreement with the present results obtained with copper.

Pure Eu_2O_3 was spread in a thin layer on a 3-mm strip of tissue paper stretched on the aluminum mounting frame and held in place by a small amount of collodion. The source was surrounded with paraffin blocks and exposed to the neutrons produced by deuterons striking a beryllium target. The Eu_2O_3 source weighed 11.7 mg/cm^2 and the paper support 1.8 mg/cm^2. Since this source is quite thick it is not suitable for a determination of the shape of the spectrum except near the upper limit. The low energy portion of the spectrum is also distorted by the secondary electrons from the gamma-rays. The K-U and Fermi plots of the spectrum are shown in Fig. 5. The Fermi curve is linear near the upper limit and intercepts the axis at 1.875 Mev. The observed upper limit is 1.885 ± 0.012 Mev. The departure from linearity below 1.4 Mev could easily be due to the thickness of the source. Whereas scattering is much less in this higher energy region, Eu has a higher atomic number than Cu (63 *vs.* 29) and hence is much more effective as a scatterer. The K-U curve is quite linear below 1.4 Mev. This again is probably a consequence of the thick source, similar to the case of copper.

There are three processes by which a gamma-ray can give up its energy to an electron: the Compton effect, the photoelectric effect and internal conversion. Compton electrons are spread over a large range of energy while photoelectrons appear in definite energy groups. Because of this the spectrometer can detect a much lower intensity of photoelectrons than of Compton electrons. This is particularly true if the secondary electrons are superimposed on a continuous spectrum. Moreover, because of the relatively low cross section for the photoelectric process at all except quite low energies, it is probable that any electron groups which can be detected with the present arrangement and strength of the source are produced by the internal conversion of gamma-rays.

The region of low energy was investigated in small intervals in order to detect secondary electron groups. Three such groups were resolved above the continuous spectrum. They are shown in Fig. 6. The measured energy values are

[6] M. L. Pool, Phys. Rev. **53**, 437 (1938).
[7] S. Sugden, Nature **135**, 469 (1935).
[8] Hevesy and Levi, Nature **137**, 185 (1939).
[9] Alichanow, Alichanian and Dzelepow, Physik. Zeits. Sowjetunion **11**, 204 (1937).

0.0727 ± 0.0005, 0.1128 ± 0.0007 and 0.675 ± 0.003 Mev. These peaks decayed with the same half-life as the continuous spectrum. This indicates that the gamma-rays originate from the decay product of Eu. The product nucleus is formed in an excited state and immediately falls to the ground state with the emission of one or more gamma-rays. The lifetime of an excited state is so short that the decay of the observed gamma-ray intensity is determined entirely by the half-life of the Eu. If the gamma-ray were emitted first this would not be the case.

The two low energy peaks are separated by 40.1 ± 1.0 kev. The K minus the L binding energy of Eu is $48.5 - 8.0 = 40.5$ kev. These peaks have the correct separation to be due to the K and L photoelectrons ejected from Eu by a gamma-ray with an energy of 0.1210 ± 0.001 Mev. If the gamma-ray were internally converted, the peaks should be separated by the K minus the L binding energy of Gd, the nucleus from which the gamma-ray originated, which is $50.3 - 8.4 = 41.9$ kev. The difference is greater than the probable error in the present measurements but not sufficiently greater to rule out this process. One serious objection to both of these possibilites is that the two groups are of equal intensity, whereas usually the K group is much the stronger in accordance with the theoretical predictions. This is perhaps reconcilable but it leads one to suspect that the peaks might be due to the K electron groups of two gamma-rays, which happen to differ in energy by 40 kev. The electron group formed by conversion of the lower energy gamma-ray in the L shell would be masked by the K group of the other gamma-ray. The L group of the higher energy gamma-ray should appear at about 0.155 Mev. There was an indication of a small peak in this region but its existence cannot be established definitely until a more intense source is obtained. If there are two gamma-rays present their energies are 0.123 ± 0.001 and 0.163 ± 0.001 Mev.

These two low energy peaks account for the exceedingly large number of low energy electrons in the Eu spectrum reported by Alichanow. His spectrometer had a very poor resolving power and could not resolve these lines from the continuous spectrum.

The electron group at 0.675 Mev is probably the K conversion electrons ejected from Gd by a 0.725 ± 0.003-Mev gamma-ray. The L group would be too weak to be detected. There are several indications of other gamma-rays but definite peaks could not be resolved with the intensity available. The relative intensities of the three gamma-rays can be estimated from the intensities of the electron groups with the help of the theoretical internal conversion coefficients. If all the gamma-rays are dipole radiation the relative intensities are 1 : 2 : 6 for the 0.123, 0.163 and 0.725-Mev lines, respectively. If the 0.725-Mev gamma-ray is quadripole radiation its relative intensity is 2 instead of 6.

J. R. Richardson[10] has investigated the gamma-radiation from Eu with a cloud chamber and has reported three lines at 0.045, 0.31 and 0.90 Mev of roughly equal intensity. In his measurements the Compton electrons from the 0.9-Mev gamma-ray would obscure the photoelectron group from the 0.725-Mev line. The Compton electron distribution from the latter gamma-ray would blend into that of the 0.31-Mev line and be difficult to resolve without a strong source. The absence of photoelectron groups in the vicinity of 1000 $H\rho$ indicates that the two such groups found with the spectrometer result from two highly internally converted gamma-rays. It was pointed out before that the separation of the groups indicates that, if they are formed by the internal conversion of gamma-radiation in Gd, they are probably due to two different gamma-rays. He suggested that the 45 kev line is the K radiation from Sm^{152}, which would be formed if Eu decayed also by the capture of a K electron and the emission of a neutrino. If a gamma-ray is internally converted in Gd approximately 45 kev radiation would also be present, so it is not necessary to postulate K-electron capture in order to explain the presence of K radiation.

I wish to express my gratitude to Professor J. M. Cork, Professor S. A. Goudsmit, Dr. H. R. Crane and other members of the Physics Department for many helpful discussions of the problems met with in this research. This work has been made possible by the Horace H. Rackham endowment fund.

[10] J. R. Richardson, Phys. Rev. **55**, 609 (1939).