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Radiations from Radioactive Substances: Au^{198}, Eu^{152}, Ag^{106}, Cu^{64} and N^{13}

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By the use of a twelve-inch hydrogen-filled cloud chamber situated in a magnetic field, investigation has been made of the gamma-radiation from Au^{198} and Eu^{152}. Observations on the 2.7-day period of Au^{198} indicate three lines with energies 70 kv, 280 kv and 440 kv, and relative intensities 0.15, 1.0 and 1.2. The beta-spectrum has an inspection upper limit of 0.83 Mev and the distribution shows that the 440-kv gamma-radiation is internally converted with a coefficient of 0.1. Thus the 70-kv radiation is probably entirely K radiation emitted after internal conversion. The Eu^{152} gamma-distribution shows three main groups of 40 kv, 0.3 Mev and 0.9 Mev. The 40-kv radiation is ascribed to the K radiation of Sm emitted as a consequence of the orbital electron capture process in Eu^{152}. Evidence is also found for the K-electron capture process in the eight-day isomer of Ag^{106}. In fact the electron spectrum probably consists of secondary electrons from the gamma-radiation following the capture process. The distribution of electrons ejected from a lead radiator by a thin source of N^{13} is compared with that from a Cu^{64} source under the same geometrical conditions. The data seem to indicate the presence of a gamma-ray of 280 kv from N^{13} with the relative probability of roughly 0.4 quantum per positron. The necessity of employing radiators of different materials for the complete investigation of a gamma-ray spectrum is emphasized.

THE sources investigated in these experiments[1] were prepared by neutron or deuteron bombardment in the Michigan cyclotron. The radiations were examined principally by means of a cloud chamber. The chamber was twelve inches in diameter, and was situated in a magnetic field uniform to ± 0.5 percent throughout the volume of the chamber. The current through the coils producing the magnetic field was maintained constant and continuous in time. The chamber was filled with hydrogen at atmospheric pressure, and a mixture of ethyl alcohol and water was used as the condensible vapor. Illumination was obtained from three 110-volt, 500-watt lamps flashed on 200 volts dc. The control circuit was of the vacuum tube type described elsewhere.[2]

In the measurements on gamma-radiation, a beam of gamma-rays was formed by a lead collimator so that the source could not "see" the bottom or top of the chamber, but only the radiator. Different materials were used as radiators; the one called the "carbon" radiator was formed from paper dipped in paraffin.

Au^{198}

The source was formed by slow neutron bombardment of gold[3] and the 2.7-day activity was investigated. The beta-rays were first examined by hanging a piece of gold foil directly in the chamber. The momentum distribution of the

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[1] Some of these results have been reported previously, J. R. Richardson, Phys. Rev. **53**, 942 (1938).
[2] J. R. Richardson, Rev. Sci. Inst. **9**, 152 (1938).
[3] E. McMillan, M. Kamen and S. Ruben, Phys. Rev. **52**, 375 (1937).

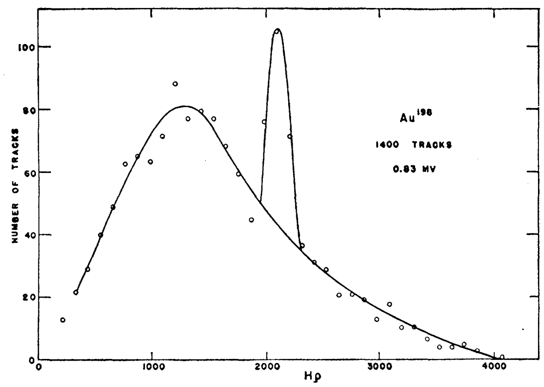

FIG. 1. The momentum distribution of the electrons from Au^{198}. The inspection upper limit is at 0.83 Mev. The internal conversion of the 440-kv gamma-ray is indicated with coefficient 0.1.

electrons obtained is shown in Fig. 1. The general shape of the distribution is similar to that usually obtained from an electron emitter of high atomic number with the exception, however, of the large number of electrons at 2100 $H\rho$. The obvious conclusion, of course, is that these are internal conversion electrons from a gamma-ray transition. This is born out when the gamma-radiation is examined. The internal conversion coefficient is then about 0.1 since there are 140 tracks in the line and 1400 in the whole distribution. The inspection upper limit of the beta-ray spectrum is 0.83 Mev.

The gamma-radiation from this source was examined with a lead radiator of surface density 20 mg/cm^2. The resulting distribution of the electrons ejected from this radiator is shown in Fig. 2. Two well-defined groups are indicated by the distribution. Considering the more energetic group we see that it must be produced by a gamma-ray of about 400 kv. Using a combination of theoretical and experimental results on the relative probabilities of the different processes we can calculate the shape and relative number of the distributions of electrons due to the Compton effect, the photoelectric effect in the K shell of lead, and the photoelectric effect in the L shell, named in the order of increasing energy. The curves in Fig. 2 have been drawn in accordance with these calculations. It must be emphasized that the data do not determine the relative size or shape of the curves. They are drawn for two reasons, first to determine if the data are consistent, second to separate out the distribution due to the K shell photoelectrons, from which the best determination of the energy can be made.

The energy is determined from the peak of the distribution of the K shell photoelectrons. This eliminates the broadening effect of the probable error of track measurement from the energy determination. Some account must be taken of the slowing down of the photoelectrons in the radiator. This can be estimated from the data of White and Millington[4] on the loss of momentum of electrons while traversing matter. For example, from their results, the loss of momentum $\Delta(H\rho)$ for the positron annihilation radiation photoelectrons (2610 $H\rho$) would be estimated as 70 $H\rho$ whereas the maximum of the K electrons experimentally occurs at 2520 $H\rho$, or a loss of 90 $H\rho$—a satisfactory check considering the errors involved.

Thus in the case of the more energetic line of gold, the maximum of the K electrons is at 2200 $H\rho$ so $\Delta(H\rho)$ is 100 $H\rho$, and the energy of the group from a very thin radiator would correspond to 2300 $H\rho$, or 348 kv. Taking account of the binding energy of the K shell of lead we see that the energy of the gamma-radiation must be 435 kv.

In the lower energy group one can ignore the contribution of the Compton effect in deter-

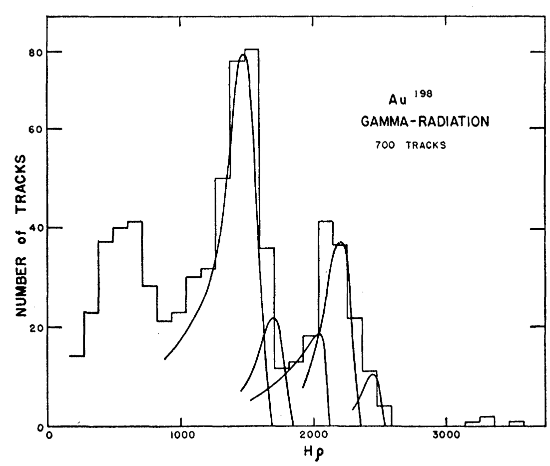

FIG. 2. The photoelectrons ejected from a lead radiator by the gamma-radiation of Au^{198}. Two gamma-ray lines are deduced at 280 kv and 440 kv and relative intensities 1.0 and 1.2. The low energy group of electrons is produced by 70-kv radiation with relative probability 0.15.

[4] P. White and G. Millington, Proc. Roy. Soc. **A120**, 701 (1928).

mining the K-electron distribution because of its much smaller probability relative to the photoelectric effect. The peak of the latter appears to be at 1470 $H\rho$. This corresponds to a gamma-ray energy of 280 kv.

The relative intensity of the two gamma-ray lines is also of interest. This can be obtained from a consideration of the relative probability of the photoelectric effect at the two energies compared with the experimental data. The result obtained is that there are 0.8 quantum of 280-kv radiation for every quantum of 440-kv radiation. This ratio is uncertain by about 30 percent.

Since internal conversion is observed only for the 440-kv line (as far as the sensitivity of the cloud chamber is concerned), and the coefficient is 0.1, it is logical to presume that the 440-kv radiation is quadrupole while the 280-kv radiation is dipole in character. There is some evidence from pictures taken with a carbon radiator for a weak line corresponding in energy to the sum of the energies of the two strong lines. The group of low energy electrons in Fig. 2 (relative probability 0.1) is undoubtedly produced by K radiation from the gold (more exactly Hg^{198}) emitted as a consequence of the internal conversion process. These x-rays would have an energy of about 70 kv and would eject photoelectrons of about 55 kv from the L shell of lead. This corresponds well with the electrons observed, both in energy and relative intensity.

The drawing of a level scheme for any β-radioactive nucleus is probably premature until more certain knowledge is obtained of the shape of a simple beta-spectrum for a given atomic number and spin change. If the beta-spectrum in the case of gold is simple, one must assume that the excited Hg^{198} nucleus subsides to the ground state in two steps—by emission of dipole radiation of 280 kv and by emission of 430-kv quadrupole radiation, or possibly in one step by the emission of a 0.7-Mev quantum.

Eu^{152}

The europium source was in the form of europium oxide which had been purified by Dr. D. W. Stewart. It was exposed to slow neutrons and the gamma-radiation associated with the 9.2-hour activity of Eu^{152} was investigated.

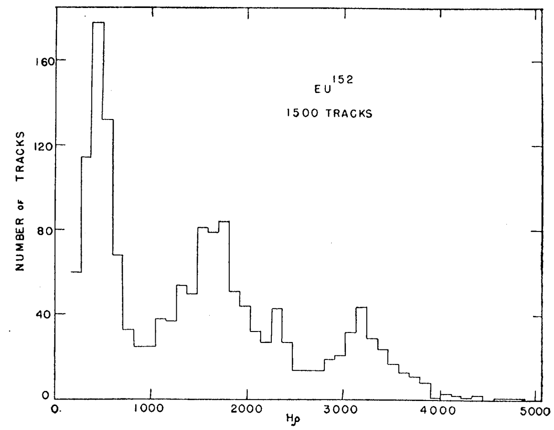

FIG. 3. Electrons ejected from a lead radiator by the gamma-radiation of Eu^{152}. The slow electrons correspond to an energy of 25 kv to be added to the binding energy of the electrons. Most of them originate in the L shell of lead, as is shown by comparison with a Cd radiator. The electrons above 2500 $H\rho$ are principally produced by the Compton process.

The momentum distribution of the electrons ejected from a lead radiator is shown in Fig. 3. The gamma-radiation is quite evidently complex, but the most interesting feature is the large number of low energy electrons. This group has an energy of about 25 kv and must obviously be composed of photoelectrons, so the question arises as to which shell in the lead atom is their origin. This can be ascertained by observing the electrons ejected from a cadmium radiator, since the binding energy of an electron in the K shell of cadmium is of the same order of magnitude as the binding for the L shell of lead. Observations on the cadmium radiator indicated the group as even more prominent and of a similar energy. Thus one may say that the energy of the radiation ejecting this group of electrons must be about 40 kv. This radiation is probably to be assigned as K radiation of Sm emitted after the capture of an orbital electron by Eu^{152}. Thus this isotope of europium has the possibility of either emitting an electron and decaying to Gd^{152} or capturing a K electron and forming Sm^{152}. The electron distribution has been investigated by Dr. Stewart.

The gamma-radiation itself is obviously complex, and it seems that one must wait for stronger sources and the use of thinner radiators before completely resolving the spectrum. It seems apparent, however, that there is one line at 0.8 Mev and a line or group of lines from 0.3 to 0.4 Mev. The number of electrons in these groups indicates that the relative number of quanta ejecting each group is roughly the same as the number of K quanta. Coincidence measurements could decide, of course, whether the gamma-radiation accompanied the K-electron capture process or the electron emission. The relative probability of the two processes seems to be approximately equal.

Ag^{106}

The eight-day activity induced in silver by fast neutron bombardment was assigned to Ag^{106} by Pool, Cork and Thornton[5] as isomeric with the 24.5-minute positron activity. Pool[6] later postulated the process of K-electron capture in the eight-day isomer to account for the abnormally large gamma to beta ratio that it exhibits. The results discussed here were presented previously before the American Physical Society.[7]

The electron spectrum of Ag^{106} was investigated by placing an activated silver foil of thickness 0.0025 cm in the cloud chamber. The momentum distribution appears as in Fig. 4. The striking feature of the distribution is the fact that forty-five percent of the tracks are in a small band of energies below 17 kv, while the remainder ranges up to 800 kv. Some of the low energy electrons were measured by means of their range in the gas of the chamber, after calibration of the method had been made. Pool did not observe these electrons because he had an aluminum window between the source and the chamber.

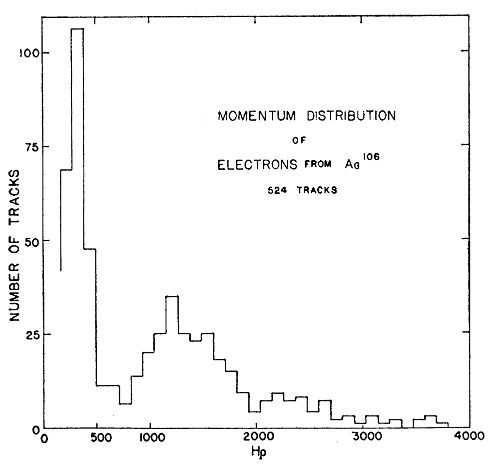

FIG. 4. The momentum distribution of the electrons from the eight-day isomer of Ag^{106}. The low energy group (17 kv) probably consists of Auger and photoelectrons from the K radiation of palladium. The other electrons are probably also secondary in nature.

It seems very unlikely that electrons of such low energy could arise from an ordinary beta-transition. The most probable explanation seems to be that the low energy group consists of Auger electrons and photoelectrons associated with the K-electron capture process in Ag^{106}. The process, of course, should be accompanied by emission of the K radiation of palladium. The writer was unable definitely to detect this radiation, however, while using a copper radiator in the cloud chamber. Other workers[8, 9] have also searched for this radiation with negative results. However, the gamma-ray activity is so intense that the method illustrated by Fig. 4 is undoubtedly much more sensitive.

From the known thickness of the foil (0.002 cm) the fluorescent yield (0.75), the absorption coefficient of Pd K radiation in silver (150 cm^{-1}), and the range of the electrons (0.6 mg/cm^2), one can determine the number of slow electrons observed per disintegration. Then taking account of the relative number of slow and fast electrons observed, one finds that there are roughly 50 transitions leading to the low energy group for every transition producing a higher energy electron.

Feather[9] estimates that there are four gamma-rays per disintegration, and in this connection it is interesting to ascertain whether or not it is necessary to assume that the higher energy electrons observed form an ordinary beta-spectrum.

If μ is the absorption coefficient of a gamma-ray, and t is the thickness of a foil throughout which the radioactive atoms are uniformly spread (e.g. by neutron activation), then the fraction of the gamma-quanta ejecting an electron from one side of the foil is given approxi-

[5] M. L. Pool, J. M. Cork and R. L. Thornton, Phys. Rev. **52**, 380 (1937).
[6] M. L. Pool, Phys. Rev. **53**, 116 (1938); M. L. Pool and E. C. Campbell, Phys. Rev. **53**, 272 (1938).
[7] J. R. Richardson, Phys. Rev. **55**, 236 (1939).
[8] L. W. Alvarez, Phys. Rev. **54**, 486 (1938).
[9] N. Feather and J. Dunworth, Proc. Roy. Soc. **A168**, 566 (1938).

mately by the expression

$$n=\{\mu t/2\}\{\ln (1/\mu t)+0.92\}.$$

This is strictly valid only for small values of μt and spherical symmetry of the gamma-ray electron interaction; but it is approximately applicable here.

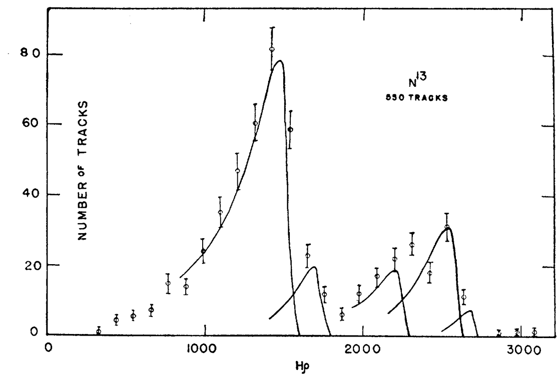

FIG. 5. The electrons ejected from a lead radiator by the radiation from a thin source of N^{13} arranged so that most of the positrons are annihilated behind the lead collimating shield. The energy of the gamma-ray is 280 ± 30 kv.

For 350-kv radiation in the foil used, one finds that $n=0.016$. This means that one would expect 1.6 percent of the gamma-quanta to eject an electron from one side of the foil. The corresponding number for a gamma-ray of 800 kv is 0.7 percent. Thus the number of electrons observed in the main part of the distribution can be easily accounted for by the ordinary processes of gamma-ray degradation. In addition, the calculations of Dancoff and Morrison[10] have made it clear that the internal conversion process would play an important role at the low energies, particularly for quadrupole and higher multipole transitions.

Sources of Ag^{106} have not yet been available which were sufficiently strong to make possible an accurate investigation of the gamma-ray spectrum. However, preliminary results with a lead radiator (200 tracks) indicate that the spectrum is certainly complex, with lines at least at 0.3 Mev, 0.5 Mev and 0.9 Mev. It is readily seen that this complex gamma-radiation could account for the "beta"-spectrum observed in Fig. 4.

[10] S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939).

N^{13} AND Cu^{64}

Evidence has previously been reported[11] which indicates the presence of gamma-radiation from N^{13} in addition to the positron annihilation radiation. An additional experiment has been performed by arranging a thin source of N^{13} so that most of the positrons were annihilated behind the lead collimating shield, but keeping the source itself in full "view" of the cloud chamber. The source was produced by bombarding charcoal with six-Mev deuterons.

The momentum distribution of the electrons ejected from a lead radiator is shown in Fig. 5. Again the components drawn in as smooth curves are determined from the theoretical ratios rather than the experimental points. Statistical probable errors are indicated by the vertical lines. The annihilation radiation, of course, is still present but the slower group of electrons is much more predominant than in Fig. 1 of reference 11, which shows the distribution obtained when all the positrons are stopped near the source. In order to examine the possibility that this group of electrons was due to some unrecognized scattering process, a source of Cu^{64} of equivalent strength was produced by deuteron bombardment of copper. The distribution obtained under the same geometrical conditions as in the case of N^{13} is shown in Fig. 6. No evidence of the low energy group is to be seen.

Assuming that this presents evidence for the existence of gamma-radiation from N^{13}, we may

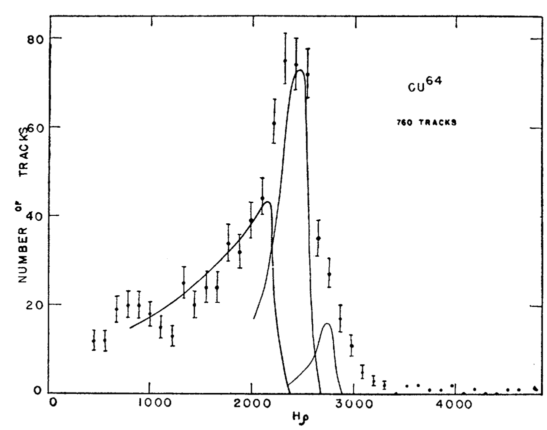

FIG. 6. The electron distribution obtained under the same geometrical conditions as in Fig. 5, but with an equivalent source of Cu^{64}.

[11] J. R. Richardson, Phys. Rev. **53**, 610 (1938).

inquire as to its energy. The radiation, of course, may consist of several lines, but if it is embodied in a single line we see from the data of Fig. 5 that the energy is about 280 kv. This is obtained from the maximum of the K-photoelectron distribution by using the method explained in the case of Au^{198}. The relative intensity of the gamma-ray must be determined from the data obtained with all the positrons of the source stopped in its immediate neighborhood. The result is found that there is 0.21 quantum of 0.3-Mev radiation for every quantum of annihilation radiation. In other words, there is 0.4 quantum of 0.3-Mev radiation for every positron emitted. This intensity estimate is uncertain by a factor of two. The result given previously was in error.

It is interesting to observe that in the case of Cu^{64} there is indication of a high energy tail to the annihilation radiation, as was found previously in the case of a carbon radiator.[12] This may mean the presence of some gamma-radiation from this source.

[12] J. R. Richardson, Phys. Rev. 53, 124 (1938).

Conclusion

It seems evident that for a complete investigation of a gamma-ray spectrum, it is necessary to employ radiators of various materials. A carbon radiator will give reliable information from 0.5 to 4 Mev, while an investigation of the photoelectrons ejected from a lead radiator will yield information about the region below 500 kv. If there are photoelectrons of energy 100 kv or less from the lead, then an experiment using some intermediate radiator such as cadmium is advisable. Only in this way can the complete spectrum be investigated properly.

The writer wishes to thank Professor J. M. Cork for his interest in this work. Thanks are due to Dr. R. L. Thornton and Dr. B. R. Curtis for their friendly cooperation. This research was supported by the Horace H. Rackham Trust Fund.

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The Penumbra at Geomagnetic Latitude 20° and the Energy Spectrum of Primary Cosmic Radiation

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The method of obtaining the penumbra presented in a previous paper is applied to the following energies: $r=0.385$, 0.400, 0.425, 0.450 and 0.500 Störmer at a geomagnetic latitude of 20°. Two graphs showing the variation of the penumbra with the energy are derived from the (γ_1, η) diagrams of these energies: one at a constant zenith angle of 60°, and the other along the east-west plane. If the number of primaries is assumed to vary inversely as the 2.8 power of their energy, the contribution of the penumbra to the directional intensity at a zenith angle of 60° is calculated, and is shown to be far from negligible. The calculated intensities are quite sensitive to the energy distribution used, and this suggests a possible method for determining the energy spectrum of primary cosmic rays.

The theory of the motion of charged primary cosmic particles developed by Lemaître and Vallarta leads to the conclusion that at every point of the earth there is a cone of many sheets within which all allowed directions are contained. In the terminology adopted by them,[1] the penumbra is the region of the allowed cone situated between the main cone and the Störmer cone, more precisely between the main cone and the simple shadow cone of Schremp.[2] In a previous paper[3] a description of methods for the determination of the penumbra was given, together with a brief summary of its structure. It was there pointed out that a complete analysis of this region, at least at one latitude, is imperative before definite theoretical conclusions, suit-

[1] G. Lemaître and M. S. Vallarta, Phys. Rev. 49, 719 (1936).

[2] E. J. Schremp, Phys. Rev. 54, 158 (1938).

[3] R. Albagli Hutner, Phys. Rev. 55, 15 (1939).