Once again, it is clear that the lines arising from L_I transitions ($L\gamma_2$ and $L\gamma_3$) are decreasing in intensity relative to a line ($L\gamma_1$) which has an L_{II} initial state.

VIII. CONCLUSIONS

The widths of lines arising from L_I transitions increase strikingly with atomic number in the range $73 \leq Z \leq 81$. At the same time the intensities of these lines relative to lines arising from L_{II} and L_{III} transitions show corresponding decreases. These observations are accounted for qualitatively by the role of the Auger effect. Only two Auger transitions are considered in the present discussion. Although beyond doubt these two are the principle ones involved in the topics of the present paper, there are certainly many more Auger transitions some of them with similar dramatic changes in probability from element to element. It is highly probable that many of the anomalies in x-ray line widths and relative intensities in other atomic number ranges will soon be shown to be due to Auger transitions.

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Radiations from Radioactive Gold, Tungsten, and Dysprosium*

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Level schemes are deduced for gold and tungsten from the measurement of beta-gamma and gamma-gamma coincidences, together with a knowledge of the energies of the various radiations emitted. The beta-ray spectrum of Au^{198} is simple with an end-point of 0.78 Mev. The residual nucleus emits two gamma-rays in cascade. W^{187} has a complex beta-ray spectrum. The high energy group has a maximum energy of 1.4 Mev while the one of lower energy has an end-point of 0.5 Mev and is followed by a 0.9 Mev gamma-ray and some other gamma-rays which give gamma-gamma coincidences. The dysprosium beta-ray end-point is 1.2 Mev and the maximum energy gamma-ray is 1.1 Mev.

IN order to obtain energy level schemes for radioactive nuclei, it is necesssry to know the energies and intensities of all gamma-rays emitted during the process, the energy distribution of all beta-rays emitted, and also any correlations between beta-rays emitted by the parent nucleus and the gamma-rays produced by the product. Up to the present our knowledge of all these factors, for any radioactive species, has been quite limited. In general one has available, or can determine with ease, data on the maximum energy of the electrons emitted during the process. In some cases the shape of the beta-ray spectrum is also known and information is available as to whether the spectrum is simple or complex. In those cases in which the spectrum is complex the value of the inner end point has not been determined with any great accuracy. Frequently gamma-rays have been found and the energies and intensities of these have been determined in a few instances.

Norling,[1] Mitchell, Langer and McDaniel,[2] and Dunworth[3] have applied coincidence counting methods to the latter problem and have been able to derive reasonable energy level schemes for several radioactive transformations. In particular, level schemes for the disintegration of Mn^{56} (2.5 hours), In^{116} (54 min.), Sb^{124} (60 days), and others have been determined. It is the pur-

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[1] F. Norling, Zeits. f. Physik **111**, 158 (1938).

[2] A. C. G. Mitchell, L. M. Langer, and P. W. McDaniel, Phys. Rev. **57**, 1107 (1940), also **56**, 380 (1939).

[3] J. V. Dunworth, Rev. Sci. Inst. **11**, 167 (1940).

pose of the present experiments to apply these methods to the investigation of radioactive gold (2.7 days), tungsten (24 hours), and dysprosium (2.5 hours).

APPARATUS

Counters were used singly to determine, by the method of absorption of beta-rays in aluminum, the beta-ray end-point. Two counters, in a coincidence circuit, were used to determine the energy of gamma-rays by measuring the range of Compton electrons ejected by them from aluminum. Coincidences between two or more gamma-rays and coincidences between beta- and gamma-rays were also measured.

Two types of counters were used in most of the experiments. Glass-walled counters of conventional design, having a brass cathode 0.0025 cm thick, were filled with argon to a pressure of 8.5 cm Hg, and 1 cm Hg of petrolic ether to aid in quenching the discharge. The sensitive region is about 7.5 cm long and 2.7 cm in diameter. The glass wall and brass cathode stop electrons with energies less than 300 kev.

In order to count electrons of energy less than this, especially designed counters having thin windows were made. These counters have a thick brass envelope and are fitted with thin aluminum windows. The windows are each 0.0025 cm thick which will absorb electrons of energy less than 70 kev and are supported on grids with transmitting areas of 1 cm^2. These counters were filled with argon at a pressure of 5 cm Hg and 1 cm of petrolic ether.

Because of the thin aluminum windows, the counters could not be baked out and were accordingly unstable. This defect was cured by a long period of evacuation and by passing a discharge through them to free the gas from the walls. After this treatment the counter thresholds increased only slightly in a period of four months.

The counters, source and absorbers were supported on two brass rods in a large copper-lined box. The apparatus could be so arranged that it could be used to measure beta-gamma and gamma-gamma coincidences on the one hand, or the energy of Compton recoil electrons from gamma-rays on the other. In either case the two counters were placed in a horizontal plane with their centers a known distance apart. If the energy of Compton recoils was to be measured, the radioactive source was placed on one side of the counters and an aluminum radiator, thick enough to stop all beta-rays, was interposed between the source and the counters. Sheets of aluminum of varying thickness were then placed between the two counters, and the number of coincidences per minute was measured as a function of the thickness of the aluminum. The energy of the gamma-ray was then obtained from the maximum range of the Compton electrons with the help of a curve published by Curran, Dee, and Petrzilka.[4]

In the cases in which beta-gamma or gamma-gamma coincidences were to be measured, the source was mounted on an aluminum block between the two counters. The block was of such a thickness that no beta-rays from the source could pass through it and at the same time it served as a radiator for Compton electrons from the gamma-rays of the source. In those cases in which coincidences between two or more gamma-rays were to be measured, a similar piece of aluminum was placed between the source and the other counter.

If coincidences between beta- and gamma-rays were to be measured the thick aluminum block between the source and one of the counters (hereafter called the beta-counter) was removed and arrangements made to place thin sheets of aluminum between this source and the counter so that only electrons with ranges greater than a certain specified amount would be detected. In this manner beta-gamma coincidences could be determined as a function of the energy of the beta-rays being recorded in the beta-counter.

In all cases the number of single counts in each counter was measured before and after each run. With the help of this information and the known resolving time of the circuit the chance coincidence rate could be calculated and subtracted from the total coincidence rate. The resolving time of the coincidence circuit was obtained in the usual manner by placing a separate source over each counter, measuring the counting rate of each counter singly and of the two in coincidence. If N_1, N_2, and N_c, are the single counting rate in each counter and the coincidence

[4] S. C. Curran, P. I. Dee, and D. Petrzilka, Proc. Roy. Soc. **169**, 269 (1938).

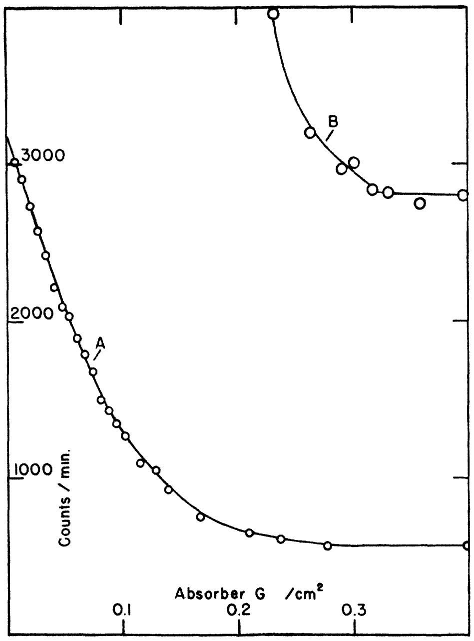

FIG. 1. Absorption of beta-rays from gold. Curve B, showing end-point, taken with stronger source than curve A.

counting rate, respectively, then

$$N_c = 2\tau N_1 \times N_2, \qquad (1)$$

where τ is the resolving time in question. In the experiments the value of 2τ varied between 9×10^{-8} min. and 7.5×10^{-8} min. depending on the counters used.

In addition, the coincidence rate due to cosmic rays was measured for every arrangement used and also subtracted from the total coincidence rate in any experiment. The cosmic-ray rate amounted to approximately 0.2 min.$^{-1}$.

The voltage source, amplifiers, and recording mechanism were those described by Mitchell, Langer, and McDaniel.[2] The apparatus could count at a rate of 30,000 per minute in each counter.

EXPERIMENTS AND RESULTS

Gold

Gold was bombarded with slow neutrons from the Indiana University cyclotron and experiments were performed on the radioactive species Au^{198} of half-life 2.7 days.

Using the method of absorption in aluminum, McMillan, Kamen, and Ruben[5] obtained a beta-ray end-point of 0.78 Mev. Richardson[6] examined the spectrum in a cloud chamber, and obtained 0.83 Mev as a maximum energy of the beta-rays. In addition, he found an internal conversion line at 0.44 Mev.

The gamma-rays have been investigated by Richardson,[6] who measured the energy of the Compton electrons and photoelectrons produced by suitable radiators in a cloud chamber. He found two lines with energies of 0.44 and 0.28 Mev of intensities 1.2 to 1, respectively. In addition, he suggested that there was a weak line at 0.7 Mev. Sizoo and Eijkman[7] obtained some evidence for a weak gamma-ray of 2.5 Mev by analyzing the absorption curve of the radiation in lead.

Absorption curves of the beta-rays were taken under various conditions. The two curves shown in Fig. 1 were taken with very different source strengths as is indicated by the disparity in intensity of the gamma-ray backgrounds. The background in Curve B was determined with the aid of other points at greater absorber thickness, not shown on the graph, and is therefore more accurate than it appears to be. The range of the beta-rays determined from this curve is 0.31 ± 0.01 g/cm² which is in good agreement with McMillan, Kamen, and Ruben. Using Feather's old rule,[8]

$$R = 0.511E - 0.091, \qquad (2)$$

which appears to be more accurate than the more

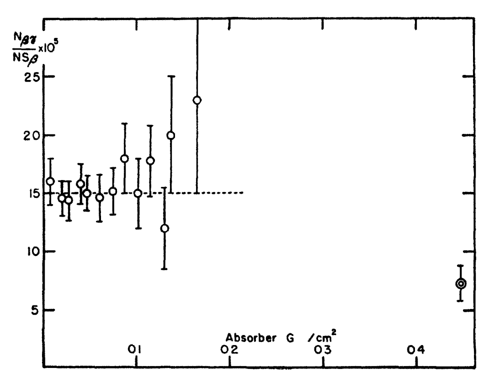

FIG. 2. Beta-gamma coincidences in gold. Double circle shows $N_{\gamma\gamma}/N_\gamma S_\gamma$.

[5] E. McMillan, M. Kamen, S. Ruben, Phys. Rev. **52**, 375 (1937).
[6] J. R. Richardson, Phys. Rev. **55**, 609 (1939).
[7] Sizoo and Eijkman, Physica **6**, 332 (1939).
[8] N. Feather, Phys. Rev. **35**, 1559 (1930).

recent one in the low energy region, one obtains an end-point energy of 0.78 Mev.

Experiments on beta-gamma and gamma-gamma coincidences were performed with both the thin aluminum-walled counters and the counters with glass envelopes. The number of gamma-gamma coincidences per recorded gamma-ray was found to be $(0.073\pm0.015)\times10^{-3}$.

The number of beta-gamma coincidences per recorded beta-ray taken with the thin-walled counters is shown as a function of the absorber thickness in Fig. 2. The vertical lines through the points indicated are calculated from the mean statistical errors, including the errors due to the background. It will be seen from the figure that the number of beta-gamma coincidences per recorded beta-ray is a constant independent of the energy of the beta-rays. Beta-gamma coincidences taken at larger absorber thickness than shown, but using the thick-walled counters, indicate that the coincidence rate per recorded beta-particle does not decrease at higher energies, but remains approximately constant all the way to the end point. From a curve of this type one would be led to suppose that the beta-ray spectrum of gold is simple, and that the disintegration of Au^{198} always leads to the same excited state of Hg^{198}, from which the gamma-rays follow. The average number of beta-gamma coincidences per recorded beta-particle can be taken from the curve and yields a value of $(0.15\pm0.01)\times10^{-3}$.

In order to calculate the number of gamma-ray quanta emitted per disintegration one employs the following considerations. Let there be N disintegrations per second and let K be the average number of gamma-quanta per disintegration. If S_β and S_γ are the sensitivities, including solid angle factor of the counters to beta- and gamma-rays, respectively, and if S_γ is supposed to be essentially constant in the region of gamma-ray energies involved, then the number of beta-gamma coincidences is given by

$$N_{\beta\gamma}=NS_\beta\cdot KS_\gamma \tag{3}$$

and the number per recorded beta-ray, by

$$N_{\beta\gamma}/NS_\beta=KS_\gamma. \tag{4}$$

Similarly the number of gamma-gamma coincidences is given by

$$N_{\gamma\gamma}=NS_\gamma^2K(K-1) \tag{5}$$

and the number per recorded gamma-ray by

$$N_{\gamma\gamma}/NKS_\gamma=S_\gamma(K-1). \tag{6}$$

The ratio of these quantities is

$$R=\frac{N_{\gamma\gamma}/NKS_\gamma}{N_{\beta\gamma}/NS_\beta}=\frac{K-1}{K}. \tag{7}$$

From these equations and the data given above

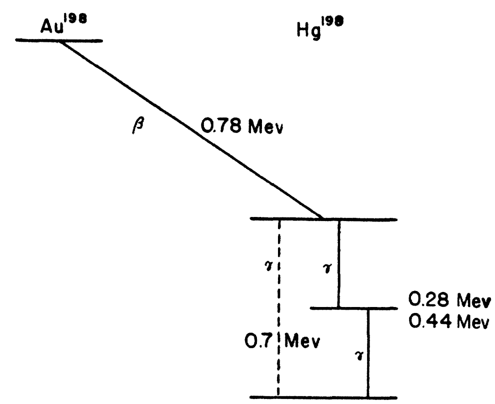

FIG. 3. Energy level scheme for gold.

one can determine a value of K, the number of gamma-quanta per disintegration. If, as a first approximation, one takes the efficiency of the counters for the various gamma-rays as equal, one obtains a value of $K=1.9$. This value of $K\approx2$ fits well with the level scheme proposed by Richardson, who assumed that the 0.28 Mev and 0.44 Mev gamma-rays are in cascade.

If, on the other hand, one assumes the level scheme of Richardson with $K=2$, one can calculate the gamma-ray efficiencies of the counters for the two gamma-rays in question.

Using formulas similar to Eqs. (3) and (5), one obtains for this apparatus

$$S_\gamma(0.44)=0.09\times10^{-3},\quad S_\gamma(0.28)=0.06\times10^{-3}.$$

An estimate of the solid angle of the gamma-counter allows one to solve for the absolute efficiencies of the gamma-rays. The results were in agreement with Norling's curve[9] to within the experimental errors.

[9] F. Norling, Phys. Rev. 58, 277 (1940).

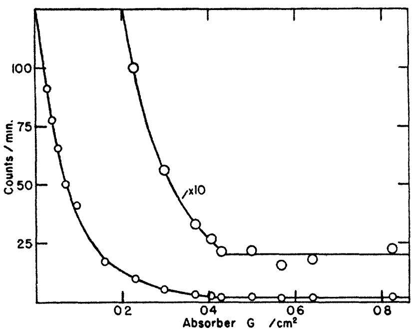

FIG. 4. Coincidence absorption of Compton electrons from tungsten gamma-rays.

The level scheme proposed by Richardson is shown diagrammatically in Fig. 3. Since no evidence has been found indicating the presence of a low energy group of electrons, it is supposed that Au^{198} goes to an excited state of Hg^{198} from which the two gamma-rays of energy 0.44 or 0.28 Mev are emitted in cascade. The weak 0.7 Mev gamma-ray may be placed hypothetically as a parallel mode in competition with the other gamma-rays in cascade. It will not affect the value of K within errors common to this type of experiment since it is relatively weak in intensity.

In view of the fact that, for ranges of electrons less than 0.1 g/cm², both internal conversion electrons and disintegration electrons are counted by the beta-ray counter, we tried to see if internal conversion electrons would have any effect on the number of beta-gamma coincidences per recorded beta-ray. One would expect a slight decrease in the number of beta-gamma coincidences per recorded gamma-ray for electrons of range less than 0.1 g/cm². Unfortunately the statistical errors could not be kept small enough to detect this effect, which should be about 5 percent. It is of interest, in this connection, to note that Norling,[10] found coincidences between internal conversion electrons and disintegration electrons in gold.

At the conclusion of these experiments, a recent paper of Norling[11] came to our attention in which he failed to find gamma-gamma coincidences in gold. The gamma-ray counters he used had walls of lead 1 mm thick which probably absorbed the 0.28 Mev gamma-ray to such an extent that no coincidences were observed. He did observe beta-gamma coincidences for two absorber thicknesses and the number of these per recorded beta-ray was independent of the energy.

Tungsten

The tungsten activity was induced with both slow and fast neutrons by Minakawa[12] who obtained 24.0 ± 0.1 hours for the half-life. He found 1.1 Mev for the maximum energy of the beta-ray spectrum by absorption in aluminum, Fajans and Sullivan[13] analyzed the spectrum in a cloud chamber and also by absorption in aluminum, obtaining 1.40 ± 0.05 and 1.40 to 1.44 Mev by the respective methods. In addition they measured a gamma-ray by the lead absorption method and gave 0.87 Mev for its energy. According to their experiments the activity is to be attributed to the isotope W^{187}. Since negative electrons are emitted, the resulting isotope is Re^{187}.

In the present experiments, a tungsten strip 0.025 cm thick was bombarded on a probe inserted into the deuteron beam in the cyclotron. Tungsten bombarded by deuterons give several radioactivities.[13] These possible contaminations include three rhenium periods with half-lives of 20 minutes, 90 hours, and 52 days, and a tungsten activity of 74.5 days. The short half-life had decayed before the observations started.

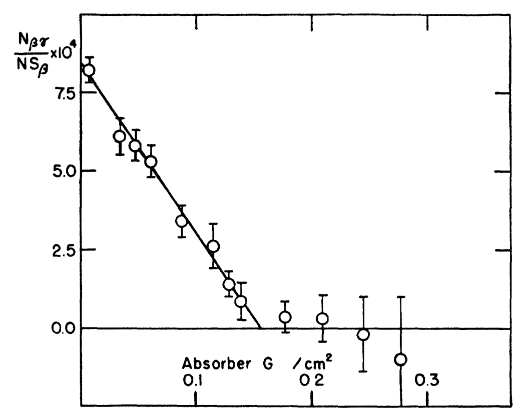

FIG. 5. Beta-gamma coincidences in tungsten.

[10] F. Norling, Naturwiss. **27**, 593 (1939).
[11] F. Norling, Arkiv f. Mat. Astr. och Fysik **B27**(3), 9 (1941).
[12] O. Minakawa, Phys. Rev. **57**, 1189 (1940).
[13] K. Fajans and W. H. Sullivan, Phys. Rev. **58**, 276 (1940).

The measurements were made without chemical separation. However, the activity followed the chemistry of tungsten and the residue had no gamma-activity. The final check on the contamination was an examination of the decay curves. The curves showed that there was a negligible amount of the 20-minute, 52-day, and 75-day activities present during the time coincidence runs were being taken. The rate of decay indicated that about six percent of the initial beta-singles was due to the 90-hour rhenium activity. Since this activity has no gamma-rays associated with it,[14] the only effect it would have is to increase the number of beta-counts and decrease the ratio $N_{\beta\gamma}/NS_\beta$. A correction was made for this contamination at various absorber thicknesses, by assuming that the shape of the beta-spectrum for rhenium is the same as for tungsten. The corrections were twelve percent or less, and have no effect on the inner end point.

No satisfactory beta-ray end-point was obtained because of the high gamma-ray background, but a rough determination giving 1.3 ± 0.1 Mev was sufficient to identify the spectrum.

The absorption of Compton electrons, determined with the thin-walled counters, is shown in Fig. 4. It will be seen that the range of the Compton electrons is 0.42 ± 0.03 g/cm^2. With the curve of Curran, Dee, and Petrzilka this corresponds to 0.90 Mev.

The results of experiments on beta-gamma coincidences with thin-walled counters are shown in Fig. 5. It is clear that the type of curve obtained here is entirely different from that shown in Fig. 2 for gold. The increase in beta-gamma coincidences in tungsten for low energy beta-particles shows that the beta-ray spectrum is complex. The absence of coincidences with thick absorbers indicates that there are no gamma-rays associated with high energy electrons. Points at still higher absorber thickness were reached with the help of thick-walled counters, and also indicated no beta-gamma coincidences. The straight line extrapolation of the beta-gamma coincidences per recorded beta-ray gives an end-point of 0.155 ± 0.015 g/cm^2. From Feather's old rule, the energy is 0.49 Mev of the lower energy group. Using values for the beta-ray end-point (1.4 Mev), the end-point of the lower energy group (0.5 Mev) and the energy of the gamma-ray (0.9 Mev), one can obtain a satisfactory level scheme for the disintegration of W^{187}. The level scheme is given in Fig. 6. The scheme is complicated, however, by the fact that we have observed gamma-gamma coincidences which suggest that there are other low energy gamma-rays in parallel with the 0.9 Mev gamma-transition.

To help in an attempt to obtain a value for K, the number of gamma-rays accompanying the low energy beta-group, as well as to get some idea of the relative intensity of the two groups, one may proceed as follows: the number of gamma-gamma coincidences per recorded gamma-ray was found to be

$$N_{\gamma\gamma}/(NKS_\gamma)_\beta=(K-1)S_\gamma=1.5\pm0.1\times10^{-3}. \quad (8)$$

The number of beta-gamma coincidences per recorded beta-particle at this absorber thickness

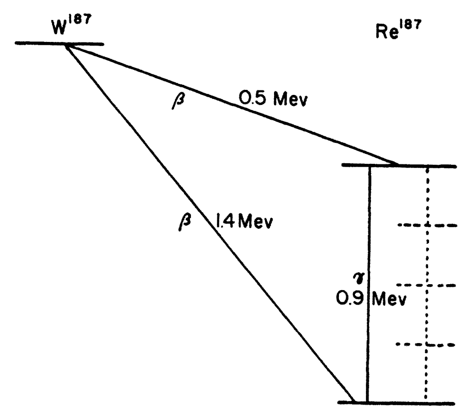

FIG. 6. Proposed energy level scheme for the disintegration of tungsten.

was

$$N_{\beta\gamma}/NS_\beta=\alpha KS_\gamma=0.84\pm0.05\times10^{-3}, \quad (9)$$

where α is the fraction of the total number of disintegrations belonging to the low energy group. If two groups are present, Eq. (7) is replaced by

$$R=(N_{\gamma\gamma}/NKS_\gamma)/(N_{\beta\gamma}/NS_\beta)_0$$
$$=(K-1)/\alpha K=1.79. \quad (10)$$

A rough value for the branching ratio α may be estimated from the slope of $N_{\beta\gamma}/NS_\beta$ as a

[14] G. T. Seaborg, Chem. Rev. **27**, 1 (1940).

function of absorber thickness to be expected if one assumes that the two beta-ray distributions have a shape to be expected from theory. Comparison of the calculated with the experimental slope led to the estimate $\alpha = 0.4 \pm 0.2$, which gives $K \approx 4$.

An independent estimate for K may be obtained by assuming a reasonable value for the average gamma-ray sensitivity. The dimensions of the experimental arrangement gave roughly 0.15 for the solid angle of the gamma-counter divided by 4π. Assuming an average absolute gamma-ray efficiency of 0.3×10^{-2}, one finds that $\bar{S}_\gamma$ becomes 0.5×10^{-3}. Then from Eq. (8), K is 4. These rough estimates seem to show that the data are consistent.

Dysprosium

The 2.5-hour activity induced by slow neutrons is probably due to Dy^{165}, since there is only one stable isotope of holmium (165) to which the dysprosium decays (emitting negative electrons), and since otherwise the isotope would be isomeric with a stable isotope of dysprosium.

Meitner[15] and Hevesy and H. Levy[16] agree on a beta-ray energy of 1.4 Mev. Mitchell, Langer, and McDaniel[2] suggested the existence of a gamma-ray. Meitner obtained the gamma-ray energy by rough absorption measurements in lead and found the energy to be of the order of magnitude of 0.6 Mev. This would be an estimate of the average gamma-ray energy.

A small amount of dysprosium oxide held on to an aluminum backing in a paraffin base was bombarded with slow neutrons from the cyclotron, to produce the source for this experiment.

The range of the beta-rays was measured with the thin-walled counters, and was found to be 0.49 ± 0.02 g/cm^2. This corresponds to an end-point of 1.20 Mev.

Compton recoils were measured to obtain the maximum energy gamma-ray. The glass-walled brass cathode counters gave a range of 0.5 ± 0.1 g/cm^2 which corresponds to a gamma-ray energy of 1.1 Mev.

Beta-gamma coincidences were found with the thin-walled counters with no absorber in front of the beta-counter. There were so many beta-rays per recorded gamma-ray that with a usable beta-rate there were too few gamma-rays to get an adequate number of coincidences for accurate work.

Gamma-gamma coincidences were also detected, indicating that the gamma-spectrum is probably complex.

The writer wishes to express his thanks to Professor Allan C. G. Mitchell, who suggested the problem, for his advice and interest in the work, and also to Professor E. J. Konopinski for many helpful suggestions concerning the correlation of the results. The author also is indebted to Professor J. M. Cork of the University of Michigan for irradiating the first samples used in the experiment, and to his colleagues at Indiana University for running the cyclotron and checking calculations.

APPENDIX

Errors

One would like to find criteria for a minimum relative error in beta-gamma coincidence experiments. On this basis Dunworth[3] has developed a formula for the maximum useful source strength for a given geometry:

$$N \approx 1/2\tau,$$

where N is the source strength and τ is the resolving time of the coincidence circuit.

Under certain conditions it may be better to use still weaker source strengths because the usable counting rate of a tube counter is limited at high rates by missing, and multiple pulses. If the source strength is too high one must sacrifice solid angle, to which the error is more sensitive than to the source strength. According to Dunworth's formula for f, the relative error due to statistical fluctuations in the total coincidence rate, this quantity depends on the source strength N approximately as $N^{-\frac{1}{2}}$. It depends on the solid angles subtended by the two counters, which are proportional to his ϵ_1 and ϵ_2, as $(\epsilon_1\epsilon_2)^{-\frac{1}{2}}$. It is evident that the best arrangement would be to have the counters as close as possible to increase ϵ_1 and ϵ_2 by increasing the solid angle and then to use such a source strength as to produce the maximum usable counting rate.

[15] L. Meitner, Arkiv. f. Mat. Astr. och Fysik **A27**(3), 17 (1941).

[16] G. Hevesy and H. Levy, Kgl. Dansk. Vid. Sels. **14**, 5 (1936).