A Study of the Nuclear Radiations from Antimony and Arsenic

Allan C. G. Mitchell, Lawrence M. Langer and Paul W. McDaniel Department of Physics, Indiana University, Bloomington, Indiana (Received April 6, 1940)

Beta-gamma and gamma-gamma coincidences were investigated in As⁷⁶, Sb¹²² and Sb¹²⁴. Measurements were made on the gamma-ray energies by coincidence absorption of their Compton recoils. Beta-ray endpoints were determined by absorption in aluminum. In As, the measurements indicate three groups of beta-rays all going to excited states of Se⁷⁶. The endpoint of the highest energy group was found to be 3.24 ± 0.20 Mev. More than one gamma-ray was found to be present. The hardest gammaray has an energy of 2.05 ± 0.05 Mev. The effects due to $\mathrm{Sb^{122}}$ (2.5 days) and $\mathrm{Sb^{124}}$ (60 days) were separated. The beta-ray spectrum of Sb124 was found to be simple, consisting of one group going to an excited state of Te¹²⁴.

INTRODUCTION

'HE study of the radiations emitted from radioactive bodies has usually been carried out by examining the shape of the beta-ray spectrum and measuring the energies and, where possible, the intensities of the various gammaray lines emitted. From the results of beta-ray studies alone it is impossible to tell whether the product nucleus is left in the ground state or in some excited state from which it may proceed to the ground state by the emission of gammarays. In some cases the beta-ray spectrum appears to be complex and to consist of two or more groups. The maximum energy of each group can be determined with only a fair degree of accuracy. Furthermore, it is generally supposed that the difference in the endpoints of the several groups corresponds to the energy of gamma-rays which are emitted but in many cases correlations have been hard to find. On the other hand, the present technique in the measurement of gamma-rays does not afford very high resolving power so that two gamma-rays lying very close in energy might sometimes be taken for a single gamma-ray. In addition, when methods of gamma-ray measurement are used which are good in the high and medium energy range, low energy gamma-rays may be entirely overlooked. On account of the difficulties involved in the above methods, the number of gamma-rays emitted per disintegration has not,

The endpoint of this group is 1.53 ± 0.05 Mev. More than one gamma-ray was found. The highest energy gamma-ray has an energy of 1.82 ± 0.05 Mev. A soft gamma-ray was also found having an energy less than 69,000 volts. Evidence is offered which suggests the alternative process of K-electron capture with a transition to Sn^{124} . The beta-ray spectrum of the short period Sb¹²² was found to consist of two groups one of which goes to the ground state of Te¹²². The endpoint of the high energy group was found to be 1.76 ± 0.10 Mev. The endpoint of the lower energy group is estimated at 0.81 Mev. Only one gamma-ray was observed. Its energy is 0.96 Mev. A self-consistent energy level scheme for Sb122 is given.

in general, been uniquely determined and the energy level schemes for the various radioactive nuclei are therefore in doubt.

The application of coincidence counting methods to this field has contributed to the solution of many of the problems mentioned above. The principle of the method is to place two counters near the radioactive source and to measure coincidences between (1) the several gamma-rays which may be emitted, and (2) between the gamma-rays and beta-rays of varying energy. If the resolving time of the coincidence circuit is small and the strength of the source is reasonably high, considerable accuracy can be obtained by this method. To date the radiations from As⁷⁶ (26 hours),¹ Mn⁵⁶ (2.5 hours),^{2,3} In¹¹⁶ (54 minutes),⁴ and Na²⁴ (14.8 hours)⁵ have been investigated. In In and Na it was found that the number of beta-gamma coincidences, calculated per thousand beta-rays, did not change with the energy of the beta-rays, indicating that the beta-ray spectrum of these elements is simple. In Mn, on the other hand, there were found to be proportionately more beta-gamma coincidences associated with elec-

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

² F. Norling, Naturwiss. **27**, 422 (1939). ³ L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, ⁴L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, ⁴L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, Phys. Rev. **56**, 422 (1939).

 ⁶ L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, ⁶ L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, ⁷ Dhys. Rev. 56 (22)



FIG. 1. Schematic arrangement for counting beta-gamma and gamma-gamma coincidences.

trons of low energy, showing that there are two groups of beta-rays in Mn and that the lower energy group is accompanied by a gamma-ray which is not found with the high energy electrons. In all three cases, beta-gamma coincidences were found to persist all the way to the endpoint and in addition gamma-gamma coincidences were found. These facts show that, in these elements, there is more than one gamma-ray per disintegration and that the process of beta-ray emission leaves the product nucleus in an excited state. Norling's results on As were obtained with such a weak source that it is difficult to draw any definite conclusions from this work. The experiments are repeated in the present article.

In the present experiments, measurements of the coincidence type have been made on As, the two periods of Sb (2.5 days) and (60 days), Eu, and Dy. In addition the energies of certain gamma-rays in As and Sb have been determined as well as the beta-ray endpoints of the various substances.

1. Apparatus

The coincidence counting apparatus was used in two different modifications for the purposes of (1) measuring coincidences between beta- and gamma-rays and those between two or more gamma-rays, respectively; and (2) of determining the energy of gamma-rays by measuring the range of Compton electrons produced by them in aluminum.

The apparatus for measuring coincidences between beta- and gamma-rays is shown in Fig. 1. Two Geiger-Müller tube counters, 2 cm in diameter and 6 cm long, were placed with their centers 4.5 cm apart in a horizontal plane. The source of radioactive material was placed midway between the two counters. Between the source and one of the counters, usually designated as the gamma-ray counter, there was placed a block of aluminum of dimensions large enough to shield the counter from the radiations emitted by the source and of thickness great enough to stop all beta-rays coming from the source. In those instances in which coincidences between gamma-rays were to be measured, a similar block of aluminum was located between the source and the other counter. In those cases in which coincidences between beta-rays and gamma-rays were to be measured, there was placed between the source and the second counter, designated as the beta-ray counter, thin sheets of aluminum of thickness sufficient to stop beta-rays of energy less than a certain specified amount. The thickness of these sheets was varied from experiment to experiment so that the number of coincidences between betaand gamma-rays could be determined as a function of the energy of the beta-rays. The counter walls had a stopping power equivalent to 0.05 g/cm² of aluminum. The counters themselves were filled with a mixture of 8 cm of argon and 1 cm of ethyl alcohol. The pulses from the counter were fed into an improved resistancecapacity coupled coincidence amplifier similar to one previously described⁶ and thence to the scaling circuit and recorder. The apparatus was capable of counting with negligible losses at rates up to 30,000 per minute. The time constants of the apparatus were reduced in such a manner as to give a high resolving time for coincidences but at the same time to maintain the desirable feature of having all the pulses reaching the mixer tube of the same size. The resolving time for coincidences was determined by recording the chance coincidences when an independent source of beta-rays was placed over each counter.³



FIG. 2. Schematic arrangement for measuring gamma-ray energies by absorption in A of Compton recoils produced in R by a source S.

⁶L. M. Langer and M. D. Whitaker, Phys. Rev. 51, 713 (1937).

Throughout these experiments the resolving time, τ , was 0.54×10^{-7} min.

Before and after each coincidence run single counts were taken in each counter so that the chance rate could be determined. Before any coincidence measurements between beta- and gamma-rays were made, the gamma-gamma coincidence rate was determined so that this could be subtracted, together with the chance rate, in determining the true beta-gamma coincidence rate.

The arrangement for measuring the energy of the gamma-rays is shown in Fig. 2. Gamma-rays from the radioactive source, S, eject Compton electrons from the aluminum radiator, R. These electrons then go through both counters, being recorded as coincidences. Sheets of aluminum are then placed at the point A between the counters to absorb the electrons. The number of coincidences per minute is then measured as a function of the thickness of the absorber A.

The energy of the maximum energy gamma-ray may be determined by observing that thickness of aluminum which completely stops all coincidences above the background rate. The energy of these electrons E may then be calculated from their range in aluminum taking into account, of course, that they have had to traverse the thick-



FIG. 3. Absorption in Al of beta-rays from As⁷⁶.



FIG. 4. Coincidence absorption of Compton recoils produced by gamma-rays of As.

ness of three counter walls or 0.15 g/cm² of aluminum in addition to the absorber thickness. The energy of the gamma-ray $h\nu$ may best be calculated with the help of the curve published by Curran, Dee and Petrzilka⁷ which was obtained by measuring the range of Compton electrons ejected by gamma-rays of known energy.

The average energy of the gamma-rays, if there is more than one, can be obtained by measuring the thickness of aluminum necessary to diminish the coincidence rate to one-half its value with no absorber present. If D_i is the thickness in g/cm² necessary to cut the rate to one-half then^{8,9}

$$D_{j} = C_{j} \frac{(h\nu/mc^{2})^{2}}{(h\nu/mc^{2}) + 1}.$$
 (1)

⁷ S. C. Curran, P. I. Dee and V. Petrzilka, Proc. Roy. Soc. 169, 269 (1938). ⁸ F. Rasetti, Zeits. f. Physik 97, 64 (1935).

⁹ A. C. G. Mitchell and L. M. Langer, Phys. Rev. **52**, 137 (1937).



FIG. 5. Beta-gamma coincidences in As⁷⁶ as a function of beta-ray energy.

The value for $C_{\frac{1}{2}}$ is determined by calibration with a known gamma-ray and has been shown to be⁹ 0.0650 g/cm². A similar result holds for the quarter-value thickness, with $C_{1/4}$ taken as 0.110 g/cm^2 . In this work both methods were used and the results compared with each other in order to see how many gamma-rays might be present.

2. Measurements on As⁷⁶

A strongly activated sample of As⁷⁶, produced by bombarding arsenic with slow neutrons from the Berkeley cyclotron, was used throughout this work. During the course of the experiments, counting rates in the beta-ray counter and the gamma-ray counter were made separately at frequent intervals so that enough data were obtained to give an accurate value of the period. From these data, taken over a period of eight half-lives, a value of 26.3 ± 0.3 hours was obtained for the half-life, which is in excellent agreement with that obtained by other observers.

The endpoint of the beta-ray spectrum was determined by measuring the number of beta-ray counts as a function of the thickness of aluminum absorbers placed between the source and one of the counters. The results are shown in Fig. 3 from which the maximum range, including the thickness of the counter wall, was found to be 1.76 g/cm² of aluminum. From Feather's rule, this corresponds to a beta-ray endpoint of 3.24 ± 0.20 Mev. This is to be compared with an extrapolated K.U. endpoint obtained by Brown

and Mitchell¹⁰ of 3.4 Mev, a similarly determined value of 3.16±0.10 by Harteck, Knauer and Schaeffer,¹¹ and an observed endpoint of 2.71 ± 0.14 obtained by Weil and Barkas.¹²

The energy of the gamma-radiation was next examined by the method of absorbing the Compton electrons ejected from aluminum as described above. A curve showing the coincidence rate as a function of total absorber thickness is shown in Fig. 4. The maximum range of the Compton electrons is 0.95 ± 0.05 g/cm². From the data of Curran, Dee and Petrzilka,⁷ the value of the gamma-ray energy is found to be 2.05 ± 0.05 Mev. This, of course, represents the energy of the hardest gamma-ray. The value of the gamma-ray energy determined from the half-value thickness is 1.67 Mev, and from the quarter-value thickness is 1.62 Mev. Since the values obtained from the half- and quartervalue thicknesses give only the average energy of all the gamma-rays, it would appear that there are other gamma-rays of energy less than 2.05 Mev present. This supposition is borne out by the fact that we have found gamma-gamma coincidences in arsenic. The gamma-ray of energy 2.05 Mev found in this work agrees quite well with that of energy 2.16 Mev found by Harteck, Knauer and Schaeffer in their cloud-chamber experiments. The average energy of the gammarays obtained from the half-value thickness indicates that the gamma-ray of energy 1.5 Mev found by these authors is probably also present. The 3.15-Mev line mentioned by them was not found in the present work.

The sample was now placed in the coincidence counting arrangement and investigated for coincidences between gamma-rays. A thickness of aluminum (0.73 cm) sufficient to stop all betaparticles was placed on each side of the source. In a series of three experiments in which from 600 to 1400 coincidences were counted, the number of gamma-gamma coincidences per thousand gamma-ray counts was found to be 0.27 ± 0.02 .

Experiments were next performed in which the

¹⁰ M. V. Brown and A. C. G. Mitchell, Phys. Rev. 50, 593 (1936). ¹¹ P. Harteck, F. Knauer and W. Schaeffer, Zeits. f.

Physik 109, 153 (1938). ¹² G. L. Weil and W. H. Barkas, Phys. Rev. 56, 485

^{(1939).}

number of beta-gamma coincidences were measured as a function of the thickness of aluminum between the source and the beta-ray counter. Before and after each run the number of singles in the beta-counter and that in the gammacounter were measured. In addition the number of gamma-ray counts in the beta-counter was also determined. From these data the number of beta-ray counts in the beta-ray counter could be determined at the mean time of the run. The number of true beta-gamma coincidences per thousand beta-ray counts was then calculated. The results are shown in Fig. 5, in which the number of beta-gamma coincidences per thousand beta-rays recorded is plotted as a function of the thickness of absorbing material traversed.

It was difficult to obtain accurate results in the neighborhood of the endpoint since the number of beta-rays recorded was small and had to be determined in the presence of a large background of gamma-ray counts. The results indicate, however, that there are beta-gamma coincidences out to the endpoint of the beta-ray spectrum. As one proceeds from the lower energy end of the spectrum it is apparent that the number of beta-gamma coincidences decreases as electrons of low energy are cut out. This occurs in two well-defined groups, one of which is associated with electrons of energy less than 0.5 Mev (0.11 g/cm²), and the other with electrons of energy less than 1.5 Mev (0.66 g/cm^2). The beta-ray spectrum of As⁷⁶ should therefore consist of three beta-ray groups-one with an endpoint of about 0.5 Mev, another at 1.5 Mev and the third with an endpoint of 3.24 Mev. In addition the highest energy betaray does not lead to the ground state of Se^{76} since beta-gamma coincidences were found with this group as close to the endpoint as reliable results could be obtained.

The authors do not feel that a quantitative energy level scheme for Se^{76} can be drawn up from these data since the curve shown in Fig. 5 is an integral curve and it is impossible to determine the inner endpoints with any accuracy. Qualitatively, however, one can see that the nucleus of Se^{76} must consist of three levels above the ground level. The three upper levels should be approximately 0.5, 1.5, and 3.2 Mev below the ground level of As^{76} . The relation of the lowest of these levels and the ground level of Se^{76} is not at present known. This level scheme is in qualitative agreement with the one published by Schaeffer and Harteck¹³ except that they assumed that the 3.2-Mev beta-ray led directly to the ground state of Se^{76} . In addition, this scheme would provide for several gammarays, probably four. One of these gamma-rays, of energy 2.05 Mev, has been definitely established in this work, and evidence for others of lower energy has been obtained.

It should be noted that As^{76} has such a position in the periodic table that it may decay to Ge^{76} by positron emission or *K*-electron capture, or to Se^{76} by electron emission. Evidence for the emission of positrons was found by Harteck, Knauer and Schaeffer.^{11, 13} The number of positrons found, however, was only about one percent of the number of electron tracks. In the present experiments we have made no attempt to separate the positrons from the rest of the electrons, but we feel that this small number of positrons would not affect our results appreciably.

Finally, it should be pointed out that the ratio of the number of gamma-gamma coincidences per recorded gamma-ray to the number of betagamma coincidences per recorded beta-ray is much higher in these experiments than we have found in the elements that we have hitherto investigated. This indicates that there are a considerable number of gamma-rays per disintegration, some of which are presumably not accompanied by the emission of a beta-ray. The ratio is approximately unity when electrons of all energies are considered. The value of 0.27 ×10⁻³ gamma-gamma coincidences per gammaray is to be compared with the extrapolated value of 0.28×10^{-3} beta-gamma coincidences per beta-ray as shown in Fig. 5. It can be shown that (see Section 5 on theory), in the region in which the above-mentioned ratio is near unity, K (the number of gamma-rays per disintegration) cannot be determined with any accuracy, since the value of K is extremely sensitive to small variations in the value of this ratio.

¹³ W. Schaeffer and P. Harteck, Zeits. f. Physik **113**, 287 (1939).

3. MEASUREMENTS ON ANTIMONY

Antimony has two stable isotopes, Sb¹²¹ and Sb¹²³, from which two radioactive isotopes can be formed by the capture of slow neutrons. Both radioactive isotopes are electron emitters and their periods have been determined as 2.8 days for Sb¹²² and 60 days for Sb¹²⁴ by Livingood and Seaborg.¹⁴ The endpoint of the beta-ray spectrum of Sb¹²² has been determined with the help of a cloud chamber by Amaki and Sugimoto¹⁵ who obtained a value of 1.64 Mev. In addition gamma-rays have been reported from the two isotopes by Livingood and Seaborg.



FIG. 6. Coincidence absorption of Compton recoils produced by gamma-rays of Sb¹²² and Sb¹²⁴.

¹⁴ J. T. Livingood and G. T. Seaborg, Phys. Rev. 55, 414 (1939); Rev. Mod. Phys. 12, 30 (1940). ¹⁵ T. Amaki and A. Suzimata Latt. Phys. and Cham.



FIG. 7. Absorption in Al of beta-rays of Sb124.

Through the courtesy of the Radiation Laboratory in Berkeley, a sample of antimony was strongly irradiated with slow neutrons and shipped back to us for study. Upon investigation it turned out that both periods were strongly activated. We originally hoped that coincidence measurements taken at various times throughout the life of the sample would enable us to determine the effects due to both periods, but this turned out to be impossible since the points at low absorber thickness gave such a large count that they could not be determined until most of the activity due to the short period had died out. After measurements on the long period had been completed we obtained a sample from the University of Michigan, which had been irradiated for only two hours, in which the ratio of the betaactivity of the short period to that of the long period was about 30 to 1.

No attempt was made in this work to determine the half-life of the long period with any accuracy. Our points agree reasonably well with a half-life of 60 days as determined by others. Measurements on the short period, taken with greater care, give a value of 63 hours for its half-life.

Measurements of the energy of the gammarays from the two periods were made in the manner described above. Two series of measure-

¹⁵ T. Amaki and A. Sugimoto, Inst. Phys. and Chem. Research, Tokyo, Sci. Paper No. 853, 1650 (1938).



FIG. 8. Decay curve for the gamma-rays of Sb showing gamma-gamma coincidences with long period only.

ments were made with the strongly activated sample: one set taken at 22 hours after receiving the sample gave a composite curve for both periods, while the other, taken at 781 hours after the start of the work, gave results which are attributable to the long period only. These two curves, adjusted for decay of the source, are shown as curves I and II of Fig. 6. The difference curve (curve III), represents the measurements on the gamma-rays of the short period.

The maximum range for the Compton electrons from the long period is 0.852 g/cm^2 , corresponding to a gamma-ray energy⁷ of 1.82 ± 0.05 Mev. The energy of the gamma-ray determined from the half- and quarter-value thicknesses with the help of Eq. (1) is 1.85 and 1.89 Mev, respectively. One would therefore conclude that, if there are more than one gamma-ray of energy greater than 1 Mev, they are so closely spaced about 1.8 Mev as not to be resolved by this method. It will be shown later that there is another gamma-ray of much lower energy present.

The maximum range of the Compton electrons from the gamma-ray of the short period is 0.45 g/cm^2 , corresponding to a gamma-ray energy of 0.96 Mev. The half- and quartervalue thicknesses give 1.15 Mev and 1.10 Mev, respectively. Here again if there are more than one gamma-ray they must be closely spaced about 1.0 Mev.

a. Coincidence measurements on the long period

As a preliminary experiment the endpoint of the beta-ray spectrum of the long period was determined by measuring the range of the electrons in aluminum with a single counter. The results are shown in Fig. 7, in which the number of counts per minute is plotted as a function of the thickness of the aluminum absorber. The maximum range corresponds to a beta-ray energy of 1.53 ± 0.05 Mev. It should also be noted that the gamma-ray background decreases with increasing thickness of aluminum from 0.2 cm to 0.6 cm, indicating the presence of a low energy gamma-ray readily absorbed in aluminum.

With 0.6 cm of aluminum between the source and each counter, measurements of gammagamma coincidences were made. These observations were carried out on many occasions from the time that the source was received until it was 800 hours old. At the time of each measurement of the coincidence rate the gamma-ray counting rate in a single counter was also measured. The results of these experiments are shown in Fig. 8 in which both the gammagamma counting rate and the intensity of the gamma-rays, as recorded in a single counter, are shown as a function of the time. It will be seen at once that, whereas the intensity as measured with the single counter, decays in a manner which is a composite of both periods, the coincidence counting rate decays with the half-life of the long period. This is true even in the early part of the run when the intensity from the short period is approximately three times that from the long period. This indicates at once that there are no gamma-gamma coincidences connected with the short period.



FIG. 9. Beta-gamma coincidences in Sb¹²⁴ as a function of beta-ray energy.

After the short period activity had died out of the source beta-gamma coincidences were measured as a function of the thickness of aluminum between the source and the beta-ray counter, keeping 0.60 cm of aluminum between the source and the gamma-ray counter. The results are shown in Fig. 9, in which the number of beta-gamma coincidences per beta-ray recorded is plotted as a function of the thickness of aluminum absorber. On the same diagram is plotted the number of gamma-gamma coincidences per gamma-ray recorded when 0.60 cm of aluminum is placed between the source and each counter. All results are, of course, corrected for decay and chance counting rate in the usual manner.

It will be noticed that the number of betagamma coincidences per recorded beta-ray is independent of the energy of the beta-rays over the entire range which we were able to measure. The value of this ratio is 0.52×10^{-3} coincidence per recorded beta-ray. These results indicate that the shape of the beta-ray spectrum of Sb¹²⁴ is simple and that the resulting Te¹²⁴ nucleus is left in an excited state.

The number of gamma-gamma coincidences per recorded gamma-ray is 0.70×10^{-3} and is greater than the value 0.52×10^{-3} beta-gamma coincidence per recorded beta-ray by a factor of 1.35. As will be shown in Section 5, this result cannot be explained on the hypothesis that the parent nucleus emits a beta-ray which is followed by several gamma-rays. Since Sb124 may be transformed into Te¹²⁴ by electron emission or into Sn^{124} by K-electron capture, it appears likely that the second process is also taking place. If this were the case there would be gamma-rays emitted in the rearrangement of the Sn nucleus which are not associated with beta-emission, thus accounting for the high value of the gamma-gamma coincidence rate compared to that due to beta-gamma coincidences.

It appeared of interest to obtain more information on the gamma-ray of low energy which was observed at the tail of the beta-ray absorption curves. A new counter holder was therefore made in which the two counters were 6.5 cm apart between centers. With this apparatus it was possible to go to greater absorber thickness so



FIG. 10. Absorption in Al of gamma-rays of Sb¹²⁴. Curves I and II show absorption of singles. Curve III shows absorption of gamma-gamma coincidences.

that more points on the gamma-ray absorption curve could be obtained. In addition, with this symmetrical arrangement, the absorption of the gamma-gamma coincidences in various materials could be observed. The object of these experiments was to obtain a measure of the energy of the low energy gamma-ray and also to ascertain whether the low energy gamma-ray was involved in the gamma-gamma coincidences which were previously observed.

One can readily see that if there are two gamma-rays present the absorption curve of the singles will be given in the usual manner by

$$N_{\gamma} = N \left[S_{\gamma_1} e^{-\mu_1 x} + S_{\gamma_2} e^{-\mu_2 x} \right]$$

and the absorption coefficient of the low energy gamma-ray will have to be measured in the presence of a background due to the high energy gamma-ray. On the other hand, if the low energy gamma-ray is coincidental with another high energy gamma-ray, it is clear that if the soft gamma-ray is recorded in one counter the high energy one will be recorded in the other and vice versa, so that the intensity of the coincidences should be governed by the absorption coefficient of the softest gamma-ray. The absorption curve is then expressible as

$$N_{\gamma\gamma} = 2N S_{\gamma_1} S_{\gamma_2} e^{-(\mu_1 + \mu_2)x},$$

where $S_{\gamma_1}{}^0$ and $S_{\gamma_2}{}^0$ are the sensitivities of the counters for the gamma-rays 1 and 2, and where μ_1 and μ_2 are the absorption coefficients of the two gamma-rays.

The experiments were carried out with both lead and aluminum as absorbers, the coincidence curve being obtained by placing an equal amount of absorber on each side of the source. The results are shown in Fig. 10. Curves I and II show the intensity of the singles as a function of thickness of an aluminum absorber, while curve III is a similar curve for coincidences. It is apparent at once that the low energy gamma-ray is involved in the coincidences and that the absorption coefficient of the soft gamma-ray is dominating the coincidence curve. The mass absorption coefficient obtained for the aluminum curve is $0.232 \text{ cm}^2/\text{g}$ which corresponds to an energy of about 69,000 volts. Since the geometry of this arrangement was not particularly suitable for absorption coefficient work, the source subtending too large a solid angle at the counters, the value obtained for the mass absorption coefficient is a lower limit and the energy of the gamma-ray therefore an upper limit. Experiments in which lead was used as an absorber gave results in qualitative agreement with those mentioned above.

Beta-gamma coincidences were also measured in such a manner that there was no absorber between the source and the beta-ray counter, and varying thicknesses of aluminum were placed between the source and the gamma-ray counter. In this way the number of beta-gamma coincidences was obtained as a function of the absorption of the gamma-rays. In this case the absorption was slight and was definitely not so large as that for the gamma-gamma coincidences. It therefore appears that the beta-rays are not coincidental with a low energy gammaray alone. One can conclude from these experiments that the beta-rays are coincidental with a high energy gamma-ray alone or with a combination of a high energy and low energy gamma-ray.

b. Coincidence measurements on the short period, Sb¹²²

In order to make measurements on the 63-hour period, Sb^{122} , the source which had been activated for two hours, was used. Throughout the time necessary to make the measurements the ratio of the activity due to Sb^{122} to that of Sb^{124} was about 30 to 1.

The absorption of the beta-rays was measured

in a single counter and a range of 0.794 g/cm^2 of aluminum was obtained. This value corresponds to an endpoint energy of 1.76 ± 0.10 MeV, which agrees, within the limit of error, with that found by Amaki and Sugimoto¹⁵ and is somewhat greater than the value of 1.53 MeV found by us for Sb¹²⁴.

The number of gamma-gamma coincidences was found to be quite small and could be accounted for exactly from the small amount of the long period existing in the source. This result is in agreement with the fact that the gamma-gamma coincidences obtained from the other source were found to decay with a period of 60 days. Since there are no gamma-gamma coincidences in Sb¹²² and since we have shown that there is a gamma-ray present, it follows that there is only one gamma-ray per disintegration.

The number of beta-gamma coincidences per recorded beta-ray is plotted in Fig. 11 as a function of the thickness of aluminum absorber between source and beta-ray counter. It will be seen that there are no beta-gamma coincidences recorded when all electrons of range less than 0.28 g/cm^2 are cut out of the spectrum, but for the lower energy electrons there is a rise in the number of beta-gamma coincidences per recorded beta-ray. This indicates that the beta-ray spectrum of Sb¹²² consists of two groups, the most energetic of which leads to the ground state of Te¹²², and the other to an excited state from which a gamma-ray may be emitted. From Fig. 11 the endpoint of the lower energy group may be determined with fair accuracy as 0.81 Mev.

The data obtained so far enable one to construct a reasonable energy level scheme for the process Sb¹²²-Te¹²². This scheme is also shown in Fig. 11. The highest energy beta-ray group has an endpoint of 1.76 Mev which leads directly to the ground state of Te¹²² since no gammagamma coincidences are found. In addition there is a lower energy group of beta-rays having an endpoint of 0.81 Mev and one gamma-ray whose energy we found to be 0.96 Mev. The sum of the energies of the low energy beta-ray and the gamma-ray is equal to that of the high energy beta-ray within the limits of experimental error.



FIG. 11. Beta-gamma coincidences in Sb¹²² as a function of beta-ray energy.

4. Experiments on Eu and Dy

A sample of dysprosium oxide was irradiated with neutrons from a 200 mg radium-beryllium neutron source until the 2.5-hour period was strongly activated. Measurements showed that the gamma-ray activity, if any, was extremely weak, there being only 35 counts per minute recorded in the gamma-ray counter when there were 13,000 per minute in the beta-ray counter.

A sample of europium oxide was kindly sent us by Professor Cork. This sample was the one that had previously been used for the work on the very long period of Eu and still showed considerable activity. The sample showed some evidence of beta-gamma coincidences but its activity was not great enough to allow us to make precise measurements. The sample was then activated with the radium-beryllium neutron source with the hope of obtaining data on the 9.2-hour period. Here again the number of gamma-rays due to this period was small compared to the number of beta-rays so that coincidence work was impossible.

5. Discussion of the Theory

In order to find the number of gamma-rays per disintegration from coincidence experiments with two counters one may make the following considerations. Let there be N_{β} disintegrations per second and let K be the average number of gamma-rays per disintegration. Let S_{β} and S_{γ} be the sensitivity of the beta-ray counter and gamma-ray counter, respectively, the solid angle subtended by the counter being included. In addition suppose that S_{γ} is essentially constant over the region of gamma-ray energies investigated. The number of beta-gamma coincidences per second will be

$$N_{\beta\gamma} = N_{\beta}S_{\beta} \cdot KS_{\gamma}$$

and the number of beta-gamma coincidences per recorded beta-ray will be

$$N_{\beta\gamma}/N_{\beta}S_{\beta}=KS_{\gamma}.$$

If gamma-gamma coincidences are to be observed, then if one gamma-ray is recorded in one counter any one of K-1 will be recorded in the other. Since there are K gamma-rays per disintegration the number of gamma-gamma coincidences per second will be

$$N_{\gamma\gamma} = N_{\beta}KS_{\gamma} \cdot (K-1)S_{\gamma}.$$

The number of gamma-gamma coincidences per recorded gamma-ray will therefore be

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

In the case of a simple beta-ray spectrum, in which the number of beta-gamma coincidences per recorded beta-ray is independent of the energy of the beta-rays, the ratio of the number of gamma-gamma coincidences per recorded gamma-ray to the number of beta-gamma coincidences per recorded beta-ray is given by

$$R = \frac{N_{\gamma\gamma}}{N_{\gamma}S_{\gamma}} \Big/ \frac{N_{\beta\gamma}}{N_{\beta}S_{\beta}} = \frac{K-1}{K},$$

so that K may be calculated from this ratio.

The maximum value of the ratio R cannot be greater than unity and still comply with the suppositions on which the theory is based. For values of R greater than unity it is clear that other effects are coming in which were not taken into account in deriving the formula. This can only be the emission of gamma-rays not connected with recorded beta-rays. In the case of Sb¹²⁴ for which the value of R is 1.35, it would seem that the only reasonable way to account for this value would be to assume that in a certain fraction of the transitions Sb¹²⁴ is being transformed into Sn¹²⁴ by *K*-electron capture, and in the resulting rearrangement of the Sn nucleus gamma-rays are given out.

The authors wish to express their appreciation

to Professor E. O. Lawrence and Dr. E. Segrè of the Radiation Laboratory of the University of California and also to Professor J. M. Cork of the University of Michigan for furnishing them with the strong sources used in these experiments. They are also indebted to Professor E. J. Konopinski for many helpful discussions during the progress of the work. They also wish to express their thanks to the Penrose Fund of the American Philosophical Society for a grant.

JUNE 15, 1940

PHYSICAL REVIEW

VOLUME 57

Protons from the Transmutation of Boron by Deuterons

ERNEST POLLARD, W. L. DAVIDSON, JR. AND H. L. SCHULTZ Sloane Physics Laboratory, Yale University, New Haven, Connecticut (Received April 8, 1940)

The technique of studying proton groups produced by bombarding elements with deuterons from a cyclotron is described. Applied to the bombardment of boron by 3.1-Mev deuterons, the method shows the presence of four groups, three of which agree with those found by Cockcroft and Lewis while the fourth, a very prominent group of Q value +2.39 Mev is new and is ascribed to a third excited state of B¹¹.

INTRODUCTION

HE earliest experiments performed with the cyclotron were studies of charged particles emitted in nuclear reactions. However, since the discovery of artificial radioactivity for the production of which the cyclotron is so powerful an instrument, the emphasis in cyclotron work has shifted and little work with charged particles has been carried out. The cyclotron at the Sloane Physics Laboratory was designed to employ the beam mainly in direct studies of transmutations, rather than in production of therapeutic samples of radioactive materials. For this purpose a moderate sized cyclotron giving relatively small ion currents is quite satisfactory. Such a cyclotron has been developed at low cost of construction and put into operation. As a first experiment boron was bombarded, since the excellent work of Cockcroft and Lewis1 affords a means by which the interpretation of results can be checked. The agreement found is good and further additional information was added by the work.

EXPERIMENTAL EQUIPMENT

The cyclotron magnet has 28'' pole pieces, designed to fit the $27\frac{1}{2}''$ acceleration chamber $\overline{{}^{1}$ J. D. Cockcroft and W. B. Lewis, Proc. Roy. Soc. 154, 261 (1936).

formerly in use at Berkeley.2 The authors are highly appreciative of the opportunity to use this chamber which has proved quite satisfactory and, once the exigencies of large scale waxing were met, has given very little trouble. The source of R.F. power is a master oscillatoramplifier with an output stage consisting of four air-cooled RCA-833 tubes in parallel push-pull. We have been able to obtain one microampere of 3.1-Mev deuterons with a total input power of 3.5 kw. Of this a little over 1 kw appears in the cooling water flowing through the dees. For most of this work one microampere is more than enough, as it causes heating in volatile targets which are harmless with less intense bombardment.

The beam is deflected into an external bombardment chamber separated from the acceleration chamber proper by a brass wall and strikes a target of the element to be studied which is placed at 45° to the beam. A hole of 1 cm diameter, 8 cm from the target, covered with an aluminum foil, permits the protons ejected from the target at 90° to the incident beam to escape. The area of target struck by the beam varies somewhat with the conditions of operation of the cyclotron (for example, upon shims, and R.F.

 $^{^{2}}$ E. O. Lawrence and D. Cooksey, Phys. Rev. 50, 1131 (1936).