α-nitroso-β-naphthol and was converted into Co_3O_4 for measurement. All active impurities separated from cobalt were measured with a G-M immersion tube. When the activities were found to be considerable the chemical separations were repeated.

The lead absorption coefficient of the gamma-ray activity was 0.54 cm^{-1}, corresponding to an energy of 1.7 ± 0.2 Mev. Lower energy gamma-rays were looked for but not observed. Beta-ray ranges in aluminum varied from 44 mg/cm^2 to 55 mg/cm^2, corresponding to an energy of 220 ± 20 kev. As previously reported,[3] the shape of the absorption curve indicated that the beta-rays were probably continuous. The presence of beta-rays with an absorption limit at 300 mg/cm^2 to 400 mg/cm^2 was observed only in samples in which no chemical separations had been made. Hence, it is probable that this group of beta-rays does not belong to Co^{60}.

In order to estimate the relative activities due to the beta- and gamma-rays an ionization chamber filled with Freon at atmospheric pressure and having a 3 mg/cm^2 paper window was used. From the absorption curve taken with this chamber it was estimated that approximately one gamma-ray was emitted for each beta-ray.

The ratio of the activities (5.3-year : 10.7-minute) which would have been produced by an infinitely long deuteron bombardment was greater than 200 : 1. The ratio of the slow neutron cross sections for the two periods[6] (5.3-year : 10.7-minute), when corrected to account for their production from the same stable isotope, is about 40 : 1.

Isomeric decay of the 5.3-year period into the 10.7-minute period is excluded by the fact that no 1.35-Mev electrons are observed in the 5.3-year activity. Isomeric decay of the 10.7-minute period into the 5.3-year period is improbable since this level decays by continuous beta-emission. An interpretation consistent with the data is that Co^{60} consists of isomeric nuclei in which the two activities decay independently.

ACKNOWLEDGMENTS

We wish to express our appreciation to Dr. John W. DeWire for making the spectrometer measurements on the 10.7-minute period. It is a pleasure to acknowledge the support received from Mr. Julius F. Stone, the University Development Fund, and the American Association for the Advancement of Science. Valuable assistance through the W.P.A. cyclotron project No. 65-1-42-89 is also acknowledged.

[6] K. Sinma and F. Yamasaki, Phys. Rev. **59**, 402 (1941)

JULY 1 AND 15, 1942 PHYSICAL REVIEW VOLUME 62

Disintegration Schemes of Radioactive Substances. IV. Fe^{59}

M. Deutsch, J. R. Downing, L. G. Elliott, J. W. Irvine, Jr., and A. Roberts
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received May 8, 1942)

The mode of disintegration of the 47-day isotope of iron Fe^{59} has been investigated by spectrometer and coincidence techniques. The beta-ray spectrum is complex, consisting of two components of approximately equal intensity with end points 0.257 ± 0.008 Mev and 0.460 ± 0.007 Mev. The low energy group is accompanied by gamma-rays of energy 1.30 ± 0.02 Mev, and the high energy group by gamma-rays of energy 1.10 ± 0.02 Mev. No beta-rays of energy greater than 0.460 Mev are present to as much as 0.25 percent of the main group. The yield of Fe^{59} from an iron target bombarded by 12-Mev deuterons is 0.05 microcurie per microampere hour. Methods of purification and preparation are described.

INTRODUCTION

THE 47-day activity induced in iron by deuteron bombardment is uniquely assigned to Fe^{59}.[1] The use of this isotope in biological work has been widespread, although it has been considered a difficult substance to use because of the low energy of the beta-radiation. The widely used dipping counter[2] has recently

[1] J. J. Livingood and G. T. Seaborg, Phys. Rev. **54**, 51 (1938).

[2] W. F. Bale, F. L. Haven, and M. L. Le Fevre, Rev. Sci. Inst. **10**, 193 (1939).

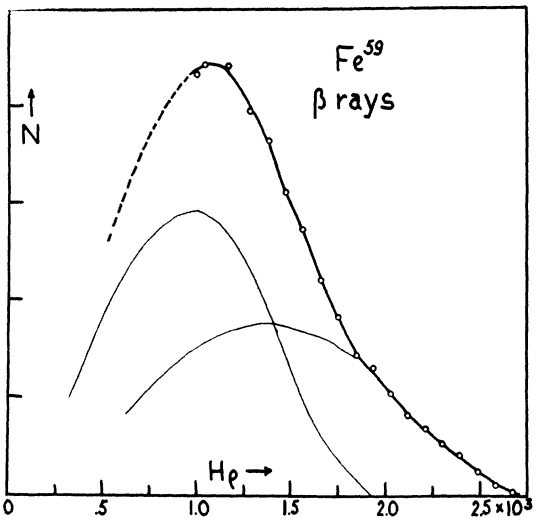

FIG. 1. The beta-ray spectrum of Fe^{59}, showing the two-component spectra. The high energy tail due to secondary electrons from the gamma-rays has been subtracted. The source was mounted on 0.001″ Al and had a surface density of 1.5 mg/cm².

been shown to detect less than 5 percent as many disintegrations from a given sample as a properly constructed thin window counter.[3] In view of the ray spectroscopy were prepared by electroplating the iron upon a suitable backing material.

After activation, 100 mg/cm² were removed from the iron target face with an end mill. The coarse powder thus obtained was dissolved in 6 *M* nitric acid, 10 mg of phosphorus added as phosphoric acid and the phosphate precipitated as ammonium phosphomolybdate. The precipitate was filtered off, 10 mg more phosphorus added, and the precipitation repeated. After removal of excess molybdenum with hydrogen sulphide the iron was precipitated three times with pyridine.[4] Each time 1 mg of manganese and 1 mg of cobalt were added as chlorides for carriers and the ferric hydroxide precipitate was dissolved in hydrochloric acid. The final precipitate was ignited and weighed as ferric oxide.

Upper limits of radioactive impurities of iron thus obtained were: radiophosphorus <0.01 percent of the total activity; radiocobalt plus radiomanganese <0.04 percent. A decay curve of this iron corresponded to $_{26}Fe^{59}$ with no long lived $_{26}Fe^{55}$ detectable in $4\frac{1}{2}$ half-periods of $_{26}Fe^{59}$. The observed half-period was 46 days.

An electrolyte consisting of 10 percent saturated oxalic acid solution and 90 percent saturated ammonium oxalate solution was added to one to ten milligrams of radio-iron in the form of ferric chloride. A rotating platinum anode stirred the solution in a cylindrical cell. Cathodes of thin aluminum or brass foil formed the bottom of the cell making it possible to deposit the iron in a circular area of any desired size. A current density of 0.3 amp./cm^2 over 3 cm^2 or less for 75 minutes leaves approximately 0.005 mg of iron in solution. One to ten milligrams of iron are usually plated out.

Gamma-ray sources were prepared by sealing ferric oxide into small thin wall glass capsules of approximately 0.2-ml volume. Secondary electron radiators were fastened to a flattened end of the capsule with wax.

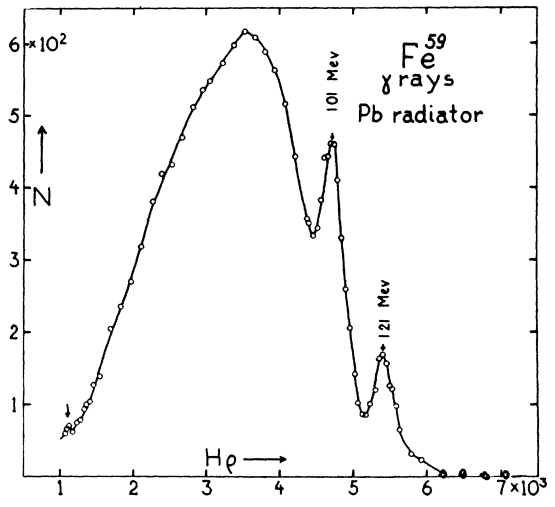

FIG. 4. Spectrum of secondary electrons from a lead radiator due to the gamma-rays of Fe^{59}. The energies of the gamma-rays corresponding to the photoelectron lines are 1.10±0.02 and 1.30±0.02 Mev. The arrow shows a line due to a 2-percent radiocobalt contamination, at $H\rho = 1010$.

RESULTS

1. The Beta-Ray Spectrum

The beta-ray spectrum obtained with the magnetic lens beta-ray spectrometer by the use of a counter with a mica window of 7 microns thickness is shown in Fig. 1. It is seen that the main group of beta-rays has a maximum energy of 0.460±0.007 Mev. There is also a very much less abundant group (about 1 percent with maximum energy about 1.1 Mev, not shown in Fig. 1). Experiments with sources of different thicknesses and different backing materials showed varying relative intensity of the high energy group. It was further noted that the end point of the high energy electrons corresponded to the maximum energy of the Compton recoils from the highest energy gamma-ray present. Therefore, on the supposition that the high energy group might be gamma-ray secondaries and not a primary beta-ray spectrum at all, the source was covered with 500 mg/cm^2 of Cu, sufficient to stop all these electrons if they were primary beta-rays, and again placed in the spectrometer. The high energy group remained unaltered with the copper absorber in place. We conclude that the high energy group is not due to primary beta-rays but to secondaries from the gamma-rays.

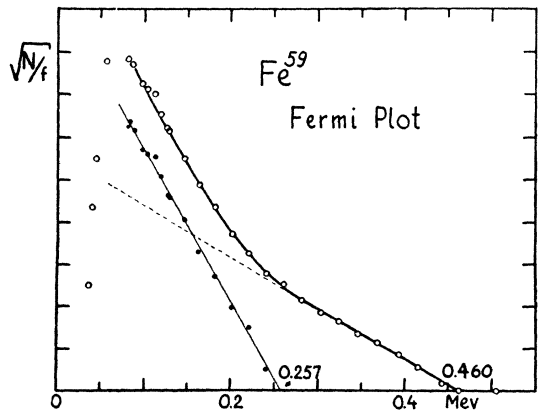

FIG. 3. Fermi plot, (for $Z = 26$) of the beta-spectrum of Fe^{59}. The secondary electron tail has been subtracted.

In addition, absorption measurements were taken with two arrangements shown in Fig. 2. Arrangement *A*, in which the source was suspended by strings, was considered satisfactory as to collimation and absence of scattering material. With this arrangement the high energy group is completely absent, while other arrangements, such as *B*, show high energy tails similar to that observed by Livingood and Seaborg.[1] We conclude that if any high energy spectrum exists at all, its intensity must be less than 0.25 percent of the main spectrum.

The Fermi plot of the beta-spectrum after correction for the gamma-ray secondaries is shown in Fig. 3. It is seen to break into two

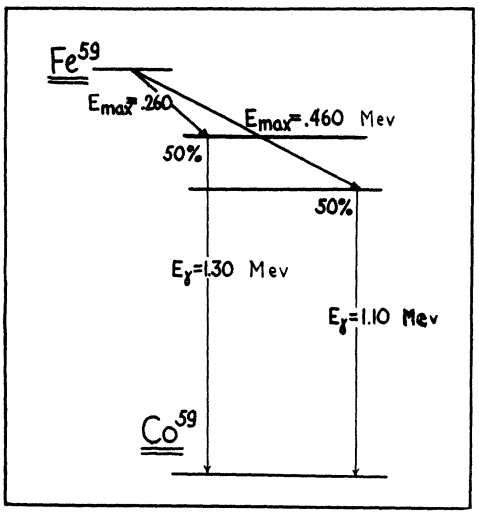

FIG. 5. Probable mode of disintegration of Fe^{59}.

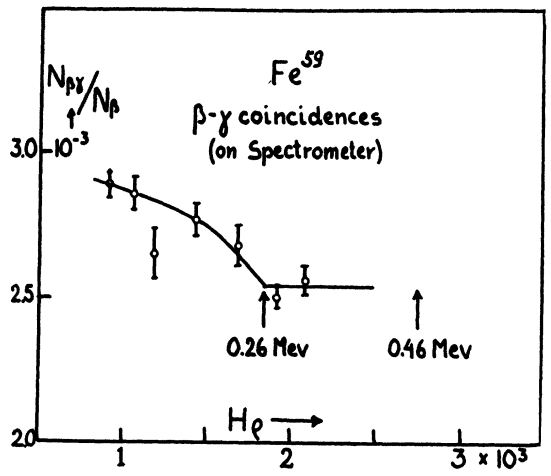

FIG. 6. Coincidence spectrum of Fe^{59}. The full line shows the shape of the curve to be expected from the energy level scheme of Fig. 5, and the known shape of the efficiency curve of the gamma-ray counter. The curve is normalized to the experimental points for the higher energy spectrum (above 0.26 Mev).

straight lines corresponding to partial spectra whose end points are 0.257 and 0.460 Mev.

The beta-decay of Fe^{59} is no doubt forbidden, and thus the significance of the breaking of the Fermi plot of the beta-ray spectrum into two linear components is not to be given too much significance, especially in view of the tentative character of current theory. In the usual interpretation, each component represents a branch to a different level in the product Co^{59} nucleus. The verification of this interpretation in the present case is thus of considerable interest.

The spectrum of secondary electrons excited in a 35 mg/cm² lead radiator by the gamma-rays is shown in Fig. 4. As previously reported[5] there are 2 gamma-rays of energies 1.10 and 1.30.

2. Coincidence Measurements

The number of beta-gamma coincidences per beta-ray showed immediately that the two gamma-rays are not in cascade, since the coincidence rate observed with a gamma-ray counter of known efficiency was characteristic of only one gamma-ray of about 1.2 Mev per beta-ray, and half what it would have been had the two gamma-rays been in cascade. An attempt to disclose the complexity of the beta-spectrum by finding a variation in the beta-gamma coincidence rate* as absorber was interposed before the beta-counter was unsuccessful, as a rise at low energies could not be unequivocally established. It was also found, with the "poor" geometry necessary in coincidence experiments, that as absorber was interposed the coincidence rate fell off, gradually approaching zero at the 0.460-Mev end point, since the secondary electrons constituting the high energy tail do not coincide with gamma-radiation. This observation is of no use in determining the origin of the tail, however, since these electrons would not coincide with gamma-rays if they were either a high energy branch to the ground state of Co^{59} or secondaries from a gamma-ray not coinciding with other gamma-rays.

Since the gamma-rays apparently do not coincide with each other, one might suppose that they represent transitions directly to the ground state from excited levels, respectively, 1.10 and 1.30 Mev above the ground state. The beta-ray transitions would end upon these levels, as in Fig. 5, the difference between their end points corresponding to the difference between the gamma-ray energies within the experimental error. This scheme does predict, however, a somewhat higher beta-gamma coincidence rate for the low energy beta-ray spectrum since the efficiency of the gamma-ray counter is higher for the higher

[5] M. Deutsch, A. Roberts, and L. G. Elliott, Phys. Rev. **61**, 389A (1942).

* Throughout this section beta-gamma coincidence rates are taken per recorded beta-ray and gamma-gamma rates are taken per recorded gamma-ray.

energy gamma-ray. The failure to observe this rise in the absorption experiment is not conclusive, since the number of electrons in the low energy spectrum counted is less than half of the total even with zero absorber, because of source thickness and counter window absorption. A much better chance of observing the rise at low energies will be obtained by measuring the beta-gamma coincidence rate on the spectrometer, where the relative number of electrons from each spectrum at low energies is much more favorable. The beta-gamma coincidence spectrum so taken is shown in Fig. 6, and the predicted rise at low energies verified.

A considerable number of attempts to detect gamma-gamma coincidences were made. According to the disintegration scheme of Fig. 5 none is to be expected. Careful repeated measurements gave a weighted mean value of $(0.050\pm0.033)\times10^{-3}$ gamma-gamma coincidence in the standard gamma-gamma set-up. Careful search of the gamma-ray spectrum for a 200-kev gamma-ray due to an occasional transition between the excited levels shows that such a gamma-ray cannot be present to more than five percent of the intensity of the competing 1.30-Mev gamma-ray. In fact Fig. 4 shows a peak in this neighborhood due to the L conversion in lead of the 131-kev gamma-ray of a 2-percent radiocobalt contamination which was subsequently removed. If one attempts to attribute the observed gamma-gamma coincidences to such a transition, the known efficiencies of the gamma-ray counters for gamma-rays of the energies involved predict a gamma-gamma coincidence rate of not more than 0.015×10^{-3}. We do not think our apparatus capable of greater accuracy than that already obtained in measuring so small a number of true coincidences (less than 1 percent of the coincidence background). We are therefore inclined to question the significance of the observed gamma-gamma coincidence rate.

DISCUSSION

The reasonably certain establishment of the disintegration scheme of Fig. 5 shows that the splitting of the Fermi plot may be of significance in establishing complexity of the beta-ray emission, even in cases of such low energy and highly forbidden disintegrations as this.

A word or two as to the angular momentum of the levels involved is in order. The ground state of Co^{59} is known to have a total angular momentum of 7/2.[6] The two beta-ray spectra have approximately equal probabilities, and since they do not differ greatly in total disintegration energy (0.77 and 0.97 Mev) it seems reasonable to suppose that they are about equally forbidden. On the other hand, beta-decay directly to the ground state is highly forbidden, and from the considerably greater energy available for such a transition it would seem that it must be forbidden at least twice more than the observed transitions. Unique angular momentum or parity assignments from these data are not possible. The indicated value for the angular momentum of the ground state of Fe^{59} is different from that of Co^{59} by at least two units, and thus may perhaps be satisfied by the values $\frac{1}{2}$ or $\frac{3}{2}$.

Yield of Fe^{59}

Since knowledge of the gamma-ray counter efficiency and the disintegration scheme permit the determination of the number of disintegrations per minute without comparison with curie standards an estimate of the total yield of Fe^{59} with 12-Mev deuterons was made. The principal errors are those of deuteron beam current measurement during the bombardment. At 12 Mev the yield is at least 0.05 microcurie per microampere hour. This is greater than the reported Berkeley yield of 0.03 microcurie per microampere hour[7] at 16 Mev.

ACKNOWLEDGMENTS

We are glad to acknowledge the cooperation of the M.I.T. cyclotron crew under Professor M. S. Livingston, and the continued interest and support of Professor R. D. Evans.

[6] K. R. More, Phys. Rev. **46**, 470 (1934); Kopfermann and Rasmussen, Zeits. f. Physik **49**, 58 (1935).
[7] J. G. Hamilton, J. App. Phys. **12**, 440 (1941).