The Radioactivity of Ga⁶⁶

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The disintegration of Ga⁶⁶ results in four positron groups with end-point energies and relative intensities of 4.144 Mev, 87 percent; 1.4 Mev, 4.3 percent; 0.878 Mev, 6.9 percent; and 0.403 Mev, 1.7 percent. Nuclear gamma-rays having energies of 1.03 Mev, 2.75 Mev and 4.8 Mev are observed. Measurement of the associated Auger electrons at 7.3 kev indicates that 34 percent of all disintegrations are by K-electron capture. The 4.14 Mev positron spectrum is found to have a shape characteristic of an allowed transition. In consideration of its high ft-value, it is therefore inferred that this transition is once forbidden with a change of parity and a spin change of 0 or 1 unit. Furthermore, this transition does not go to the ground state of Zn⁶⁶. but rather appears to be followed by the 1.03-Mev gamma-ray. The direct 5.17-Mev transition between ground states is found to proceed in less than 0.4 percent of the positron decays. This transition is apparently twice forbidden with a spin change of 3 units and is effectively outcompeted by the other less forbidden modes of decay.

I. INTRODUCTION

HE radioactivity of the 9.4-hr. transition from Ga⁶⁶ to Zn⁶⁶ has been investigated by means of a large, high resolution, magnetic spectrometer. The original interest in the problem arose from the fact that, on the basis of the previously reported¹ absorption end point of 3.1 MeV, the comparative half-life ($fl \sim 10^7$) suggested that the positron spectrum might exhibit a shape different from that generally associated with allowed transitions. Further support for this idea was based on the predictions of the nuclear shell models.^{2,3} which indicate that the transition from the ground state of Ga⁶⁶ to that of Zn⁶⁶ should involve no change of parity and should therefore fall into the twice forbidden class.

The results of the spectrometer measurements soon showed that the disintegration is quite complicated. The positron spectrum was found to be complex and was resolved into four groups with end-point energies of 4.144 Mev, 1.4 Mev, 0.878 Mev and 0.403 Mev. Furthermore, it appears that the intense 4.144-Mev group does not go to the ground state of Zn⁶⁶, but instead is followed by a 1.03-Mev gamma-ray. The direct 5.17-Mev positron transition between the ground states has a relative intensity of less than 0.4 percent. This transition was too weak to afford a reliable measurement of its spectrum shape. The momentum distribution of the 4.144-Mev positron group is such that the conventional Fermi plot is a straight line. This would be consistent with this transition's being once forbidden with a change of parity and a change of 0 or 1 unit of angular momentum.4

In addition to the 1.03-Mev transition, gamma-radiation was measured corresponding to energies of 2.75

Mev and 4.8 Mev. On the basis of the data, a reasonable energy level scheme is proposed for the Zn⁶⁶ nucleus.

From the high intensity of Auger electrons, it appears that the positron emission is in competition with considerable K-electron capture to one or more of the levels of Zn⁶⁶.

II. EXPERIMENTAL METHOD

The energy measurements were obtained with the aid of a high resolution, 180-degree focusing, 40-cm radius of curvature, shaped magnetic field spectrometer.⁵

For most measurements, a 2.3 mg/cm² mica end window counter was used as a detector. For studying the low energy Auger electrons, a second counter was employed whose window was one double layer of zapon, 150Å thick, supported by a grid of Lektromesh.⁶ Since the filling gas will slowly diffuse through such a thin window, the 2 cm total pressure of two parts ethylene to one part argon was automatically maintained constant by means of a Cartesian Manostat.⁷ This method proved to be completely satisfactory in keeping the counter operating under constant conditions. Since the apparatus is entirely mechanical, it does not suffer from the electrical contact difficulties encountered with other methods of pressure control.8

The Ga⁶⁶ was prepared by bombarding a copper probe with alpha-particles in the cyclotron. The probe tip was dissolved in acid and the Ga completely separated as chloride by a repeated ether extraction. In addition to the 9.4-hr. Ga⁶⁶ resulting from $Cu^{63}(\alpha, n)Ga^{66}$, there were also produced some 68-min. positron activity and 83-hr. gamma-activity from the reactions $Cu^{65}(\alpha, n)Ga^{68}$ and $Cu^{65}(\alpha, 2n)Ga^{67}$. The decay of the Ga⁶⁶ activities was monitored and was found not to be influenced by the other periods during the time of the spectrometer runs. The measured half-life was 9.45 hr.

A thin positron source was prepared on a 0.00025-in.

^{*} This work was assisted by a grant from the Frederick Gardner Cottrell Fund of the Research Corporation and by the joint program of the ONR and AEC.

¹W. B. Mann, Phys. Rev. 52, 405 (1937).

G. Mayer, Phys. Rev. 78, 16 (1950).
E. Feenberg and K. C. Hammack, Phys. Rev. 75, 1877 (1949).

⁴L. M. Langer and H. C. Price, Phys. Rev. 76, 641 (1946).

⁶ L. M. Langer and C. S. Cook, Rev. Sci. Inst. **19**, 257 (1948). ⁶ Langer, Motz, and Price, Phys. Rev. **77**, 798 (1950). ⁷ Obtainable from the E. Greiner Company.

⁸ Ter-Pogossian, Robinson, and Townsend, Rev. Sci. Inst. 20,

^{289 (1949).}



FIG. 1. Momentum distribution of the positrons of Ga⁶⁶.

Al backing by spreading, with the aid of insulin,⁹ a rectangle 0.3 cm by 2.5 cm. A second positron source, 0.5 cm wide and 25 times more intense was prepared in a similar manner for the investigation of the weak high energy group. This source was also used in a search for internal conversion electrons.

For the study of Auger electrons, two sources were used. One was 1.0 cm by 2.5 cm on a thin backing⁹ of LC 600. The upper and lower edges were grounded by tabs of Al leaf to prevent charging. The second Auger source was 0.4 cm wide and had a 0.00025-in. Al backing.

The photo- and Compton electrons from the gammaradiation were investigated by containing a strong Ga



FIG. 2. Fermi plot of the spectra of Ga⁶⁶. The solid circles represent data obtained with a source 25 times more intense.

31Ga66 Mev Intensity (%) 0.403 1.7 0.878 6.9 2 Beta 3 1.4 4.34.144 87.0 4 5 5.17(?)< 0.41.03 2.75 4.8 0.7 Gamma 2 . . .

TABLE I. Beta- and gamma-rays of Ga⁶⁵.

source in a rectangular copper box whose front face was covered by a 0.4 cm by 2.5 cm radiator of 28 mg/cm^2 uranium.

Coincidence measurements were performed with the source located between end window Geiger counters spaced 4 cm apart. The beta-gamma-coincidence measurements were corrected for the additional background arising from the paired quanta from positron annihilation. The resolving time of the coincidence circuit was $\tau = 0.44 \times 10^{-7}$ min. Weighed Al sheets were used for the absorbers.

III. RESULTS

The momentum distribution of the positrons of Ga⁶⁶ is shown in Fig. 1. The obviously complex spectrum has been resolved into four groups on the basis of the Fermi plot shown in Fig. 2. Here it is clear that the intense 4.144-Mev group results in a straight line. The solid circles in Fig. 2 are the unadjusted data obtained with the more intense source mentioned above. In the proposed decay scheme (see Fig. 7) there exists the possibility of a 5.17-Mev transition directly to the ground state of Zn⁶⁶. The data show that this transition occurs in less than 0.4 percent of the positron decays. The measured intensity of this group was too small to yield any information about the spectrum shape of this presumably twice forbidden transition. The maximum energies and relative positron intensities are shown in Table I.

The distribution of Compton and photo-electrons ejected from the uranium radiator is shown in Fig. 3. K and L photo-lines may be seen which correspond to the 0.511-Mev annihilation radiation and to a 1.03-Mev gamma-ray. In addition, there is a K photo-peak from a gamma-ray of 2.75 Mev and a Compton distribution whose maximum energy corresponds to a gamma-ray energy¹⁰ of 4.8 Mev. The areas under the 1.03 Mev and 2.75 photo-lines have been measured and compared, after correction by Gray's formula for the variation of photoelectric cross section with energy. The 2.75-Mev

⁹L. M. Langer, Rev. Sci. Inst. 20, 216 (1949).

 $^{^{10}}$ R. Hofstadter and J. A. McIntyre, in a private communication, confirm, by means of scintillation counting techniques, the existence of these gamma-rays. They also report a 4.25-Mev gamma which is not sufficiently resolved in the Compton distribution of Fig. 3. This gamma-ray could correspond to a transition to the ground state of Zn⁶⁶ following on the 0.878-Mev positron decay from Ga⁶⁶.



FIG. 3. Compton and photo-electrons ejected from a uranium radiator by the gamma-rays of Ga⁶⁶.

gamma-ray has 0.7 the intensity of that at 1.03 Mev. The intensity of the 0.511-Mev photo-line is here not indicative of the total positron activity because the copper box containing the source was not of sufficient thickness to cause annihilation of all positrons.

A careful search was made for internal conversion lines. The conversion electrons corresponding to the 1.03-Mev gamma-ray are shown in Fig. 4. Because of uncertainties in the distribution of K capture transitions and in the measurement of relative intensities, no reliable estimate could be made of the internal conversion coefficient.

Figure 4 also shows a continuous distribution of negatrons with most of the intensity concentrated in the region below 1500 Gauss-cm. The shape of this distribution shows no resemblance to that of a beta-ray spectrum and is attributed to bremsstrahlung and possibly to negatrons from pairs internally produced by the high energy gamma-rays.

Figure 5 shows a line of Auger electrons of 7.34 kev energy. Since the amount of internal conversion is relatively small, practically all of the Auger electrons are attributable to the rearrangement of the atomic electrons following K electron capture. The area under the line corresponds to 28 percent of the total positron intensity. Using a value of 0.46 for the fluorescence yield¹¹ in Zn, one concludes that 34 percent of all disintegrations are by K electron capture.

Beta-gamma-coincidences per recorded beta-ray are plotted in Fig. 6, as a function of Al absorber thickness placed before the beta-counter. The extremely low value of the ordinate for beta-energies above 1.4 Mev is interpreted to be indicative of a lifetime of the 1.03-Mev level in Zn⁶⁶ comparable to or longer than the resolving time of the apparatus.

The decay scheme of Fig. 7 is proposed after the following considerations. (1) The 2.75-Mev gamma-ray energy fits well the observed difference between the 4.144-Mev and 1.4-Mev positron end points. (2) Since the 1.03-Mev gamma-ray is the most intense, it must follow the 2.75-Mev gamma-ray and hence the 4.144-Mev beta-group. (3) This scheme provides a means of energy release following the lowest energy (0.403-Mev) beta-group and removes the necessity of associating the 4.8-Mev gamma-ray with a level fed only by K electron capture.



FIG. 4. Negatron distribution from a Ga⁶⁶ source showing the internal conversion of the 1.03-Mev gamma-ray.

¹¹ Compton and Allison, X-Rays in Theory and Experiment (D. Van Nostrand Company, Inc., New York, 1935), p. 489.





FIG. 7. Disintegration scheme for the Ga⁶⁶-Zn⁶⁶ decay.

FIG. 5. Auger electrons following K electron capture.

IV. CONCLUSIONS

The momentum distribution of the positrons from Ga⁶⁶ is complex. With the help of the Fermi theory, four groups are resolved with end points and relative intensities as shown in Table I. The most intense group, with an end point at 4.144 Mey, apparently does not represent a transition directly to the ground state of Zn⁶⁶, but rather to an excited state 1.03 Mev above the ground level. The intensity of the 5.17-Mev direct transition is less than 0.4 percent of the positron decays.

On the basis of the observed energies and intensities and taking into account the 34 percent of K electron capture, one finds, in order of decreasing end-point energy, the following values for $\log(fl)$, a measure of the comparative half-life of the individual positron groups: 7.84, 7.11, 6.29, 6.37.

The shape of the spectrum of the 4.144-Mev positron transition is of the allowed form. In consideration of the relatively high *ft*-value, one must infer therefore that



FIG. 6. Beta-gamma-coincidences as a function of positron energy.

this transition is of the once forbidden type, involving a change of parity and a spin change of 0 or 1; a spin change of two units should have resulted in a unique forbidden shape.4

According to the nuclear shell model,² the 31st proton and the 35th neutron in Ga⁶⁶ should each separately be in $P_{\frac{1}{2}}$ levels. According to Nordheim,¹² one might expect the resultant state of Ga⁶⁶ to have even parity and a spin of 3. Since the ground state of Zn⁶⁶ is that of an even-even nucleus, it is presumed to have even parity and zero spin. The transition between the ground states of Ga⁶⁶ and Zn⁶⁶ is then *twice* forbidden with a spin change of 3 units and is effectively outcompeted by the other less forbidden transitions. From the fact that the 4.144-Mev positron transition does not exhibit a forbidden shape, one may infer further that the 1.03-Mev excited state in Zn⁶⁶ has a spin of at least 2 and odd parity. The 1.03-Mev gamma-transition would then involve a change of parity and a change of at least 2 units of angular momentum. The character of the radiation would then be at least magnetic quadrupole or electric octopole. The experimental absence of coincidences between the 4.144-Mev positron group and the 1.03-Mev gamma-ray suggests a relatively long lifetime for the excited state of Zn⁶⁶. This, of course, is only reasonable if the 1.03-Mev gamma-radiation is of high polarity.13

In addition to the 1.03-Mev gamma-radiation, evidence was found for quanta of 2.75-Mev and 4.8-Mev energy.

Figure 7 shows an energy level scheme which appears to be reasonably consistent with the observed data.

The authors would like to take this opportunity to thank Professor M. B. Sampson and the cyclotron crew for their kind cooperation.

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