The Decay Scheme of Cu^{66}

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The decay scheme of 5.2-minute Cu^{66} has been determined by beta-ray spectrometer, scintillation spectrometer, and coincidence measurements. It is found that 9.2±1 percent of the beta-ray transitions go to an excited state of Zn^{66} whose lifetime is less than 10^{-7} second and are followed by 1.044±0.010-Mev gamma-rays. The remaining 91 percent of the beta-decays go to the ground state of Zn^{66}, with an upper energy limit of 2.63 ±0.02 Mev. This high energy beta-group appears to have the allowed spectrum shape. Both beta-decays have *ft* values characteristic of allowed transitions. Spin and parity assignments for Cu^{66} and for the 1.044-Mev level of Zn^{66} are made, and some features of the decay of Ga^{66} to Zn^{66} are discussed.

I. INTRODUCTION

THE radiations from Cu^{66} have been investigated by means of lens spectrometer, scintillation spectrometer, and coincidence measurements. This study was originally undertaken because Langer and Moffatt,[1] in their decay scheme for Ga^{66}, had proposed a 1.03-Mev level in Zn^{66} with a lifetime greater than 10^{-6} second, and because it seemed of interest to establish if this level was involved in the decay of Cu^{66} also. The results of the present work indicate that about 9 percent of the beta-disintegrations of Cu^{66} do indeed go to a Zn^{66} level at 1.044±0.010 Mev, but that this level has a lifetime of less than 10^{-7} second. The remaining 91 percent of the Cu^{66} decays go to the ground state of Zn^{66}.

The half-life of Cu^{66} has been variously reported as 5 min,[2] 4.34±0.03 min,[3] and 5.18±0.10 min.[4] The maximum beta-particle energy was reported as 2.9 Mev from cloud-chamber measurements[5] and as 2.58 Mev from absorption data.[6] Meitner[7] observed gamma-rays and gave their energy as 1.32 Mev from lead absorption measurements.

II. EXPERIMENTAL WORK

Sample Preparation and Half-Life Determination

All samples of Cu^{66} were prepared by short (1 second to 6 minutes) neutron irradiations of copper in a pneumatic tube in the Brookhaven reactor. For the scintillation and lens spectrometer measurements of the gamma-rays, electrolytic copper was used in the form of 0.001-inch sheet. All other data were taken with copper films 0.2 to 2 g/cm² thick, evaporated in vacuum onto 0.001-inch Cellophane. To determine what corrections, if any, needed to be made for activities induced in the Cellophane backings, Cellophane blanks were irradiated along with the copper samples and their activities checked in the various experimental setups used in the copper measurements. The corrections required for the Cellophane activities were negligible in all cases.

The decay of the total beta-radiation from a copper source irradiated in the reactor for 2 minutes was followed for 90 minutes. After subtraction of the long-lived background (mostly 12.9-hour Cu^{64}), which accounted for about 1 percent of the initial counting rate, a simple exponential decay with a half-life of 5.2±0.1 min resulted, in good agreement with the value of Cameron and Katz.[4] The gamma-rays were found to decay with the same period.

To minimize the effects of Cu^{64} radiations, measurements were made after each irradiation during the first 10 to 15 minutes only.

Beta-Spectrum

The beta-spectrum of Cu^{66} was measured with a conventional lens type spectrometer with point focusing. Evaporated copper films 0.3 to 0.7 mg/cm² thick were used. Each film was irradiated for 2 minutes and then used for about 10 minutes of spectrometer measurements; this procedure made it possible to run, with fair statistics per point, over approximately half the spectrum with each source. A total of 10 sources was used. The effects of varying source strengths and of source decay were eliminated by an automatic monitoring technique which utilized the counting rate of a G-M counter in a fixed position to adjust each counting interval to the source strength. Since a vacuum gate was not available, the sources were located outside the spectrometer end plate immediately adjacent to a 10-mg/cm² aluminum window.

The Fermi-Kurie plot of the beta-spectrum, constructed with the use of the Fermi functions given by Feister,[8] is shown in Fig. 1. It is linear from about 1.5 Mev to the end point at 2.62 Mev. The maximum beta-energy, corrected for energy loss in the spectrometer window, is 2.63±0.02 Mev. The deviation from the

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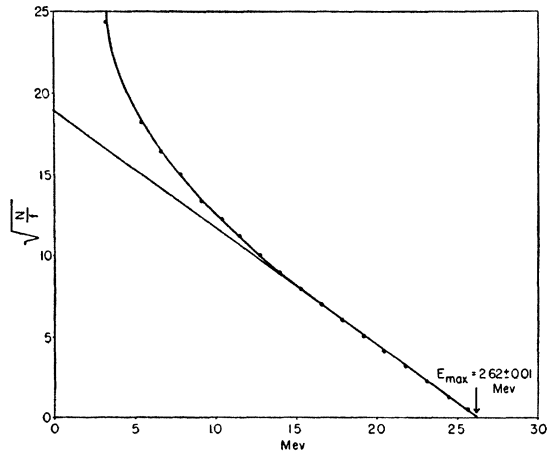

FIG. 1. Fermi-Kurie plot of the Cu^{66} beta-spectrum. With window absorption corrections, the upper energy limit becomes 2.63±0.02 Mev. The deviation from the straight line $F-K$ plot below 1.5 Mev is partly due to the 9 percent low energy beta-group (end point 1.59±0.03 Mev), partly due to the effect of the window between source and spectrometer.

straight line $F-K$ plot below 1.5 Mev is compatible with the presence of a beta-group of 1.5±0.1-Mev maximum energy with an intensity of less than 20 percent of that of the 2.63-Mev group. However, the observed deviation from linearity cannot be taken as proof of such a low energy group because at least part of the deviation is probably caused by the aluminum window between source and spectrometer as shown in experiments by H. Motz on the spectra of Al^{28} and P^{32} sources taken under the same conditions.

Gamma-Ray Measurements

In a preliminary search for gamma-radiation, a Cu^{66} source was placed near a NaI scintillation counter, and the pulse-height distribution as displayed on an oscilloscope screen was photographed. The pulse height distribution obtained about 5 minutes after a 2-minute irradiation is shown in Fig. 2, together with the spectrum of Cs^{137} which was used for energy comparison. A photoelectron line at 1.0±0.05 Mev and the associated Compton distribution were found to decay with a half-life of approximately 5 minutes. Another gamma-ray, of about 0.5-Mev energy, was initially weaker than the 1-Mev radiation, but decayed very little in a few hours; it is undoubtedly annihilation radiation associated with the positron decay of 12.8-hour Cu^{64}.

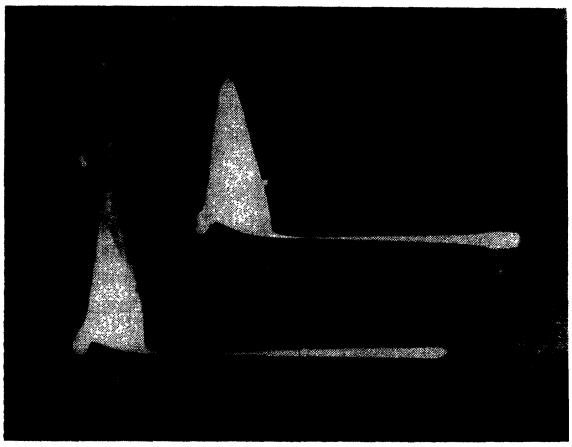

FIG. 2. Photograph of scintillation counter pulse distribution for Cu^{66} as displayed on an oscilloscope screen. Photopeaks and Compton distributions of the 1.04-Mev gamma-ray of Cu^{66} and of the Cu^{64} annihilation radiation can be seen. The Cs^{137} pulse distribution is shown at the right, for comparison.

To determine the gamma-ray spectrum of Cu^{66} more accurately, the photoelectrons ejected from a uranium radiator 27 mg/cm² thick were measured in the lens spectrometer with a resolution setting of 2.7 percent. Copper sources weighing about 50 mg each and about 2 mm in diameter were placed in an open-ended brass capsule whose walls were thick enough to absorb all the beta-rays. The closed end of the capsule was placed against the outside of the spectrometer window, and the uranium converter was attached to this window inside the spectrometer. The G-M counter used to monitor source strength was placed opposite the open end of the source capsule and was shielded with enough aluminum to absorb all of the Cu^{64} beta-radiation, so that the counting intervals for the spectrometer counts were automatically adjusted for the Cu^{66} decay.

The spectrum showed K and L photoelectron peaks and Compton electrons due to a gamma-ray of energy 1.044±0.010 Mev (Fig. 3). In addition, weak K and L photoelectron peaks from 0.51-Mev annihilation radiation were observed; but because of the monitoring technique the intensity of these peaks (which are associated with Cu^{64}) varied as a function of time relative to the gamma-ray spectrum due to Cu^{66}. A careful search above about 200 kev revealed no evidence for any other gamma-rays associated with the decay of Cu^{66}.

Search for Internal Conversion Electrons

After the energy of the gamma-rays had been established, a careful search for conversion electrons was made with the lens spectrometer. The experimental arrangement was the same as in the beta-spectrum measurements, except for the fact that the resolution was changed to 2.0 percent. Data were taken with each of 22 sources on 17 points in the energy range 900 to 1100 kev. Unfortunately, the internal conversion line of the 1.04-Mev gamma-ray would occur at the peak of the beta-distribution. The statistics were such that one conversion electron in 4000 beta-rays would have been detected. However, no internal conversion line was found; from this result and from the gamma-to-beta intensity ratio of 0.09 (see the following section) an upper limit of 3×10^{-3} can be set for the conversion coefficient of the 1.04-Mev gamma-ray. Langer and Moffatt[1] observed conversion electrons of the 1.03-Mev

gamma-ray of Ga^{66}, but gave no estimate of the conversion coefficient.

Relative Intensities of Beta- and Gamma-Rays

To determine the relative intensities of beta- and gamma-rays from Cu^{66}, a copper source mounted on 2.5-mg/cm^2 Cellophane was placed between a thin window G-M tube and a NaI scintillation counter shielded by about 1.2 g/cm^2 of aluminum. Counts were taken simultaneously with the two counters, and corrections were applied to the results for natural background, for the (very small) activity of the Cellophane backing, for the long-lived Cu^{64} activity, for the absorption of beta-rays in the air and counter window, and for the gamma-counting rate in the G-M counter. The resulting corrected ratio of gamma-to-beta counts in the particular experimental setup used was 0.0675. Measurements were taken under identical conditions on sources of Co^{60} and Cs^{137}, and with the appropriate background and absorption corrections the gamma-to-beta counting rate ratios turned out to be 1.54 for Co^{60} and 0.529 for Cs^{137}. In case of the Cs^{137} the "beta-rate" includes the contribution of the conversion electrons from the 0.66-Mev gamma-ray.

According to the well-known decay schemes[9] of Co^{60} and Cs^{137} the ratios of gamma-quanta to electrons emitted in the decays of these nuclides are 2.00 and 0.764, respectively. Therefore, the ratio of ratios is

$$(\gamma/\beta)_{Co}/(\gamma/\beta)_{Cs}]_{theor}=2.00/0.764=2.62.$$

The corresponding measured ratio of ratios is

$$[(\gamma/\beta)_{Co}/(\gamma/\beta)_{Cs}]_{meas}=1.54/0.529=2.91.$$

Assuming the counting efficiency of the G-M counter to be the same for all the beta-rays and conversion electrons concerned, this result indicates that the scintillation counter registered the Co^{60} gamma-rays (average energy 1.25 Mev) about 10 percent more efficiently than it did the 0.66-Mev Cs^{137} gamma-rays. By interpolation one may conclude that the 1.04-Mev Cu^{66} gamma-rays were counted about 6 percent more efficiently than the Cs^{137} gammas.

The measured ratio of γ/β-ratios for Cu^{66} and Cs^{137} was

$$[(\gamma/\beta)_{Cu}/(\gamma/\beta)_{Cs}]_{meas}=0.0675/0.529=0.128.$$

With the 6 percent difference in gamma-counting efficiency taken into account, the true ratio of ratios thus becomes

$$[(\gamma/\beta)_{Cu}/(\gamma/\beta)_{Cs}]_{theor}=0.94\times0.128=0.120.$$

Therefore, the ratio of the number of gamma-quanta to the number of beta-particles emitted by Cu^{66} is

$$0.120\times0.764=0.092.$$

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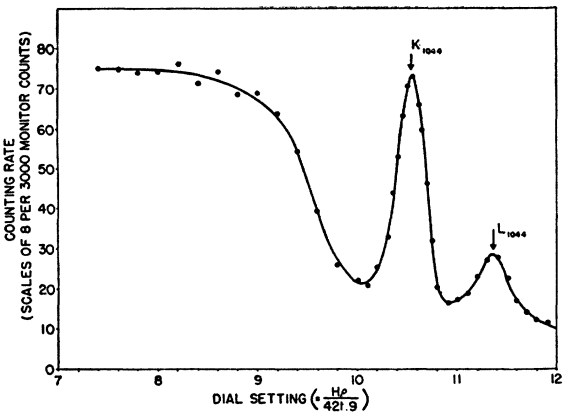

FIG. 3. External conversion spectrum of Cu^{66} gamma-radiation taken with a uranium converter in a lens spectrometer. The K and L photoelectron peaks and the Compton distribution due to the 1.044±0.010-Mev gamma-ray are shown.

A consideration of the experimental errors indicates that the uncertainty in this ratio is about 10 percent.

Coincidence Experiments

With an anthracene scintillation counter as a beta-particle detector and a NaI scintillation counter for gamma-rays, beta-gamma coincidences were observed from Cu^{66} sources. The coincidence circuit used had a resolving time of about 0.3 microsecond. The number of beta-gamma coincidences per beta-count was found to be 0.076 times as large in Cu^{66} as in Au^{198} which is known to emit one gamma-quantum per beta-particle. This is consistent with the gamma-to-beta ratio 0.092 for Cu^{66}, provided the variation with energy of the gamma-counter sensitivity is taken into account. This result alone would seem to exclude the possibility of a lifetime greater than 10^{-6} second for the 1.04-Mev state in Zn^{66} as postulated by Langer and Moffatt.[1] However, to confirm this conclusion, delayed coincidences were determined as a function of delay time by introducing variable time delays from 10^{-7} to 1.7×10^{-6} second in the beta-side of the coincidence

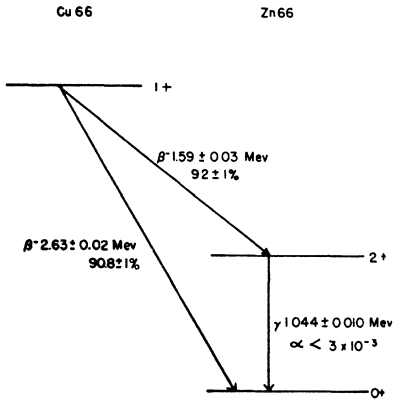

FIG. 4. Decay scheme of Cu^{66}.

arrangement. The curves obtained of coincidence counting rate *vs* delay time had identical shapes within statistics for Au^{198} and Cu^{66}, and from these data a conservative upper limit of 10^{-7} second can be set for the half-life of the 1.044-Mev state of Zn^{66}.

III. DISCUSSION

The experimental results reported lead to the decay scheme for Cu^{66} represented in Fig. 4. The abundance of 9.2 ± 1 percent for the low energy beta-group deduced from the γ/β intensity measurements is consistent with the $F-K$ plot (Fig. 1) of the beta-spectrum. Using the nomographs given by Moszkowski,[10] one finds log (ft) values of 5.35 and 5.45 for the high and low energy beta-groups, respectively. This clearly labels both transitions as allowed.[11] The ground state of the even-even nucleus Zn^{66} presumably has spin 0 and even parity; therefore, Cu^{66} may be assigned spin 0 or 1 and even parity, and the 1.044 Mev level in Zn^{66} must have even parity and spin 1 or 2, depending on whether the Cu^{66} spin is 0 or 1. This assignment is confirmed by the upper limit of 10^{-7} second for the 1.044-Mev excited state which would not be compatible with a spin change ≥ 3 for the 1.044-Mev transition.[12] The rather high upper limit of 3×10^{-3} for the conversion coefficient of the 1.044-Mev gamma-rays does not contribute to the spin assignments. It would be compatible even with a 2^4-pole transition.[13]

If the 1.044-Mev state of Zn^{66} observed in the Cu^{66} decay can be identified with the 1.03-Mev level reported by Langer and Moffatt,[1] the decay scheme of Ga^{66} given by these authors must be modified. The absence of coincidences between the most energetic positron group of Ga^{66} (4.144 Mev) and the 1.03-Mev gamma-ray was interpreted by Langer and Moffatt to indicate that these positrons lead to the 1.03-Mev level, but that this level has a lifetime in excess of 10^{-6} second. The present data contradict this assignment and lead to the conclusion that the 4.144-Mev positrons of Ga^{66} probably go to the ground state of Zn^{66}. Further confirmation for this interpretation is the recently published value[14] of 6.05 Mev for the $Zn^{66}(p, n)Ga^{66}$ threshold (or 5.96 Mev for the Q of the reaction); this corresponds to 5.18 Mev for the $Ga^{66}-Zn^{66}$ mass difference, or $5.18-1.02=4.16$ Mev for the maximum energy of the positron transition to the ground state of Zn^{66}. This is in excellent agreement with the measured value of 4.144 Mev.

This re-evaluation of Langer and Moffatt's Ga^{66} data allows a further narrowing of the Cu^{66} and Zn^{66} spin assignments. Since Langer and Moffatt did not find any evidence for a positron group with a maximum energy $4.144-1.044=3.10$ Mev, one must conclude that the positron transition from Ga^{66} to the 1.044-Mev state is more highly forbidden than the ground-state transition. This in turn would be very hard to explain if the spin difference between ground and first excited states in Zn^{66} were only one unit with even parities for both. Thus by far the most likely spin value for the 1.044 Mev level is 2; the Cu^{66} spin is then 1.†

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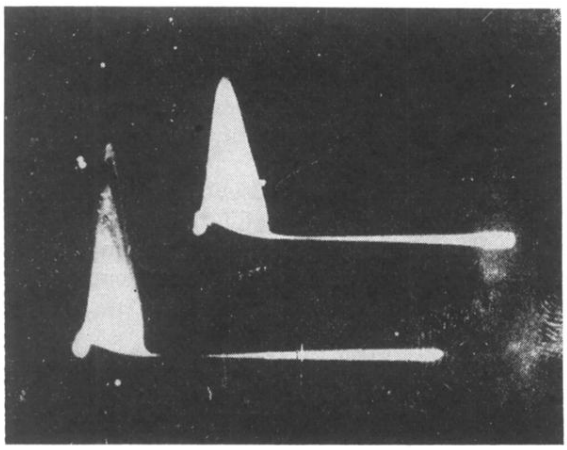
† *Note added in proof:*—Dr. W. E. Meyerhof has kindly called the authors' attention to the fact that the β^+ transitions from Ga^{66} to the 1.04-Mev and ground states of Zn^{66} may well have the same degree of forbiddenness. With a branch of the order of one percent to the 1.04-Mev level, the log (ft) values would be quite similar, and such a small branch could have escaped detection in Langer and Moffatt's work. The spin of 1 for the 1.04-Mev level can therefore not be conclusively ruled out, although spin 2 is still more probable.

The authors are very grateful to Dr. Meyerhof for sending them the results of Roderick, Meyerhof, and Mann on the Cu^{66} decay prior to publication. These results are in substantial agreement with the present work.



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