Isomeric State of Y^{88} in the Decay of Zr^{88}†

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(Received November 29, 1954)

The 85-day activity Zr^{88} decays by electron capture to Y^{88m} which de-excites by emission of 395-kev gamma radiation with a half-life of 0.37 ± 0.03 millisecond. From the half-life and the conversion coefficient the transition is characterized as $E3$.

I. INTRODUCTION

A PRELIMINARY report on the nuclide Zr^{88} by Hyde and O'Kelley[1] showed that it decayed by electron capture with a half-life of many months into the well-known 105-day isotope Y^{88}. An accurate value of 85 days was given later.[2] Prominent in the decay was a single gamma ray of 406 kev as determined in a beta-ray spectrometer. No positrons were detected nor were any other gamma radiations observed.

Recently we have restudied the gamma radiation of this isotope in a scintillation spectrometer. The activity was prepared by bombarding 0.005-inch thick foils of pure niobium metal with 100-Mev protons in the 184-inch cyclotron, whereby Zr^{88} is produced by the $Nb^{93}(p,2p4n)Zr^{88}$ reaction. The target was set aside for two months to allow complete decay of all interfering zirconium activities, principally 80-hour Zr^{89}. At the end of this time a pure sample of Zr^{88} was isolated by the radiochemical procedures previously described.[1]

The gamma radiation was studied in a sodium iodide crystal-photomultiplier tube combination coupled, after suitable amplification, to a 50-channel electronic pulse-height analyzer. As shown in Fig. 1, a single gamma ray was detected. By repeated calibration against the gamma standards U^{235}(184 kev), Cs^{137}(662 kev), Am^{241}(59.8 kev), Cd^{109}(87 kev), Na^{22}(510 kev), Hg^{203}(279 kev), and Nb^{95}(745 kev), the energy was determined to be 395 kev. This is believed to be more accurate than the previous beta-ray spectrometer value of 406 kev. No other gamma radiation was detected nor was there any evidence of annihilation radiation from positron decay.

Isomerism in the Y^{88} daughter was established by an x-ray—gamma-ray coincidence experiment carried out with the assistance of Frank S. Stephens. A set-up was used in which the sample was placed between two sodium iodide-photomultiplier combinations facing each other. The gate crystal was encased in beryllium rather than aluminum to increase the transition of 15-kev x-rays. The output of this crystal-photomultiplier assembly was applied to a single-channel pulse-height analyzer to allow selection of pulses corresponding to the x-rays and these were used to gate the coincidence circuit. The output of the second crystal-photomultiplier combination was applied to the signal side of the coincidence circuit. Those pulses arriving in coincidence with the gate pulses within the rather slow resolving time of 5×10^{-6} second were displayed on the 50-channel pulse-height analyzer. Experiments performed in this apparatus with pure samples of Zr^{88} showed conclusively that the 395-kev gamma ray was not in coincidence with the x-rays, indicating a delayed state in Y^{88} with a half-life longer than 5×10^{-6} second.

Application of the Montalbetti[3] nomogram of the Weisskopf[4,5] formula for half-lives of isomers indicated the following predicted mean lives for various transition types: 2×10^{-9} second for $E2$, 2×10^{-7} second for $M2$, 8×10^{-4} second for $E3$, 0.8 second for $M3$, 13 minutes for $E4$, and 2 hours for $M4$. Conversion would affect these values but little.[6] Chemical experiments in which yttrium daughter activity was rapidly separated from the Zr^{88} parent activity and examined for the 395-kev gamma radiation were negative, indicating a half-life shorter than a few seconds. Hence it seemed apparent that the transition was $E3$ or $M3$ in nature and that the half-life might be expected to lie in the region of 10^{-2} to 10^{-4} second. A delayed-coincidence technique was applied to this problem with success.

II. MEASUREMENT OF THE HALF-LIFE

A weak sample of Zr^{88} with a disintegration rate of a few hundreds per minute and mounted on aluminum foil was placed between the two sodium iodide crystals mentioned above. The side of the aluminum foil on which the activity had been evaporated faced the beryllium covered crystal. As before, pulses corresponding to 15-kev x-radiation intercepted by the gate crystal were selected by a single-channel analyzer. The output of the analyzer was fed to the trigger input of a Tektronix-514D oscilloscope to start a trace across the oscilloscope face. The speed of this trace could be adjusted with the standard controls of the oscilloscope. The pulses from the signal crystal were fed to the vertical deflecting system of the oscilloscope. Signal pulses arriving in prompt coincidence with the 15-kev

† This research was carried out under the auspices of the U. S. Atomic Energy Commission.
[1] E. K. Hyde and G. D. O'Kelley, Phys. Rev. **82**, 944 (1951).
[2] E. K. Hyde, Phys. Rev. **92**, 927 (1953).
[3] R. Montalbetti, Can. J. Phys. **30**, 660 (1952).
[4] V. F. Weisskopf and J. M. Blatt, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).
[5] V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951).
[6] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

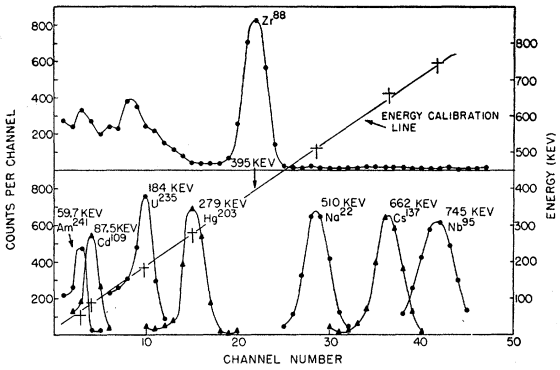

FIG. 1. Gamma spectrum of pure sample of Zr^{88}.

trigger pulses were displayed as dots at the start of the trace. The height of the dots above the base line was proportional to the energy of the detected gamma radiation. These signal pulses appeared as dots for the usual reason that the duration of the flat-topped pulse (7 microseconds) was greater than the rise and decay time. Any delayed gamma-ray signal pulses or randomly occurring chance pulses which arrived during the sweep appeared as dots at a corresponding distance from the start of the trace.

A Poloroid Land camera was used to take exposures of 4-minute duration of the scope face. When the sweep speed was set at 0.3 millisecond per centimeter a finished photograph showed a band of dots at a height corresponding to the 395-kev gamma ray decreasing in intensity across the horizontal time scale. Dots at lower energies corresponding to the Compton scattered radiation were also observed. An exact time scale was printed on the photographs by impressing a sine wave of 1200 cycles per second from a calibrated signal generator on the vertical deflection plates of the oscilloscope.

To extract the data recorded on the photographs the 395±20-kev energy band was marked off by penciled lines and the horizontal time scale was marked off in units of 333 microseconds ($\frac{1}{2}$ the period of the 1200-cycle sine wave). The dots in each square were then counted under a low-power microscope. The number of dots per square were plotted on semi-logarithmic graph paper as a function of the distance (time). Figure 2 is a composite plot of the data from 17 photographs. The half-life of 0.37 millisecond is believed to be good to ±0.03 millisecond.

A common difficulty in this type of experiment is that halation from the base line on the oscilloscope screen fogs the photograph during the necessarily long exposure times. This difficulty was avoided by not using the normal unblanking circuit of the Tektronix-514D oscilloscope. Instead, when the positive signal pulse from the crystal-photomultiplier amplifier combination was fed to the vertical deflection system as mentioned above, it was simultaneously fed to an inverter circuit which inverted and amplified the pulse. The resulting negative pulse was fed to the cathode input post on the oscilloscope to unblank the trace at the instant a signal appeared on the vertical deflector.

The Zr^{88} is an almost ideal case for the application of this delayed coincidence method inasmuch as no extraneous radiations are present, although we have applied the method successfully to the measurement of a 10-millisecond delay in a gamma ray associated with Mo^{90} where an appreciable chance background had to be considered.[7]

The 0.37-millisecond half-life corresponds most closely to the $E3$ half-life prediction, although $M3$ probably cannot be ruled out. The conversion electron spectrum of the 395-kev gamma transition is shown in Fig. 3. The observed K/L ratio is 8.4. With presently available empirical data on K/L ratios[8] or theoretical calculations of L-shell conversion coefficients no clear choice between $E3$ and $M3$ can be made on the basis of this K/L ratio although $M3$ appears to be favored.

III. MEASUREMENT OF THE CONVERSION COEFFICIENT

A sensitive test of transition type is the K-shell conversion coefficient. Exact theoretical value for the transition under discussion can be taken from the theoretical curves of Rose *et al.*[6] The expected K conversion coefficients are 0.03 for $E3$ and 0.065 for $M3$.

An experimental measurement of the total conversion coefficient (which, within the experimental error, can be equated to the K-shell conversion coefficient inasmuch as the K/L ratio is large) was made in the following way.

A sample of freshly purified Zr^{88} was mounted on aluminum and counted in a GM counter which had been calibrated with a Bureau of Standard's RaDEF beta standard. Beryllium and aluminum absorption curves were run to determine that 55 percent of the activity detected through no absorber was assignable

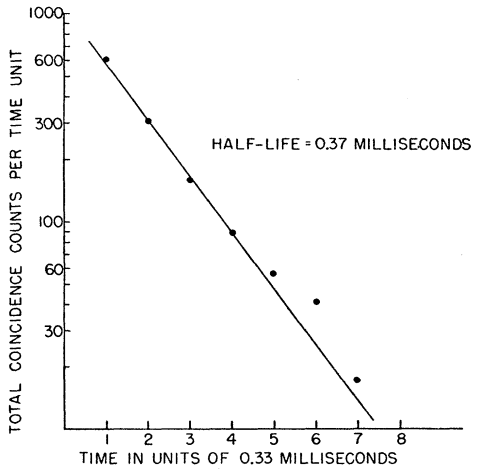

FIG. 2. Decay curve for Y^{88m}. Data compiled from 17 photographs as described in text.

[7] H. B. Mathur and E. K. Hyde, Phys. Rev. (to be published).
[8] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

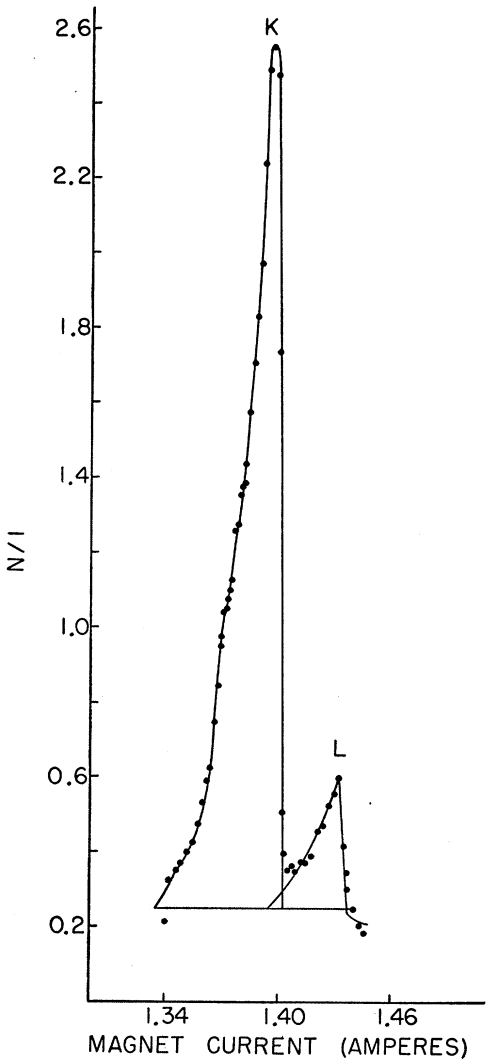

FIG. 3. Conversion electrons of 395-kev transition in Y^{88m}. Data taken by J. Schooley and J. Juliano. *Note added in proof.*—Recent recalibration of the beta-ray spectrometer shows that the best value of the γ-ray energy is 387 rather than 395 kev.

to conversion electrons, the remaining 45 percent being due to the much less efficiently counted x-ray and 395-kev gamma-ray photons. The electron counting rate was corrected for geometry and a backscattering to 10 percent, which yielded a value of 6350 electrons per minute for the sample.

The gamma-ray emission rate was measured by duplicating Fig. 1 with this particular sample, using a sample position about 4.5 cm away from the 1-inch $\times 1\frac{1}{2}$-inch sodium iodide crystal. The geometry factor of this position was known to be 3.8 percent from measurements of the photopeak of the 59.8-kev gamma ray emitted by an Am^{241} sample of known alpha disintegration rate. The area under the photopeak of the 395-kev gamma ray was corrected for this geometry factor and for a 27 percent photopeak efficiency to calculate an absolute gamma emission rate of 261,000 per minute.

The ratio of conversion electrons to gamma-ray photons was 0.024. The number of conversion electrons was checked by counting the same Zr^{88} sample in an anthracene crystal-photomultiplier combination coupled to the 50-channel pulse-height analyzer. Because of the low conversion coefficient the conversion electron peak was obscured somewhat by Compton scattered electrons, but the contribution to the observed electron spectrum from this source was easily found by rerunning the spectrum with a 120 mg/cm^2 aluminum absorber between the sample and the crystal to absorb the electrons.

The counting yield of the geometrical arrangement used in this measurement was calibrated by counting a sample of Cs^{137} under identical conditions. The conversion electron line at 625 kev was easily resolved from the β^- spectrum. The conversion coefficient of the 662-kev transition in the Cs^{137} decay is 0.1.[8] By counting the gamma-ray photons in the sodium iodide spectrometer, integrating under the photopeak, correcting for a crystal efficiency of 12 percent and correcting for geometry, the absolute number of conversion electrons expected from the sample was calculated and compared with the observed electron counting rate. The counting yield figure so obtained was 23 percent.

This number was used to convert the observed electron counting rate for the Zr^{88} sample to an absolute disintegration rate. This number was 17 percent higher than that found in the GM counter, yielding a conversion coefficient of 0.027.

Considering the errors in the measurements, the final value for the total conversion coefficient can be given within the limits $0.02_5 \pm 0.01$. Hence the conversion coefficient clearly favors $E3$ rather than $M3$ for the transition type.

IV. DISCUSSION

The 0.37-millisecond activity decays into and is isomeric with the 105-day Y^{88} whose decay scheme is well established.[9] The great majority of nuclear isomers can be related to the nuclear shell model as has been shown conclusively in the review of Goldhaber and Hill.[10] Even in the case of odd-odd nuclei the configurations of the individual odd particles correspond to the predictions of the single-particle model and isomers can occur when the odd neutron or proton lies just beneath a completed shell. In this new example the odd-proton and odd-neutron numbers are 39 and 49, respectively, and the isomerism can be attributed to the closed shell at 50 particles.

Empirical data from many nuclides show that $f_{5/2}$, $p_{3/2}$, $p_{1/2}$, and even $g_{9/2}$ levels are available to the 39th particle. The 49th particle may be either $g_{9/2}$ or $p_{1/2}$.

[9] For references see Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[10] M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179 (1952).

It cannot be predicted with certainty which of these possible orbitals will be occupied or how the odd neutron-odd proton orbitals will couple. Nordheim's rule[11] is useful for ground state predictions, but it has many exceptions.[12] In the case of the 105-day ground state of Y^{88} the spin is 4 with odd parity resulting from a $p_{1/2}$ proton and a $g_{9/2}$ neutron. Inasmuch as the 0.37-millisecond gamma transition is $E3(\Delta I=3$, yes), the isomeric level must be $+1$ or $+7$. Using the orbitals just mentioned, the only likely choice is $+1$ resulting from a $p_{1/2}$ proton and a $p_{1/2}$ neutron. An additional reason excluding $+7$ is that the even-even Zr^{88}, presumably with spin 0, would not be expected to show prominent electron capture decay to a $+7$ state.

There are several other examples of odd-odd isomerism in this mass region. The most closely related examples are $_{37}Rb_{49}^{86}$ (2 isomeric forms)[10,13,14] and $_{41}Nb_{49}^{90}$ (3 isomeric forms)[7] which differ from Y^{88} only in the subtraction or addition of 2 protons. In addition, Rb^{84} and Nb^{94} exist in isomeric forms.[10]

It perhaps should be mentioned that an isomeric form of Y^{88} with properties distinctly different from the one reported here (2-hour positron emitter) was published many years ago[9] but that later workers have shown this report to be in error.[15]

[11] L. W. Nordheim, Revs. Modern Phys. **23**, 322 (1951).
[12] M. H. L. Pryce, Repts. Progr. Phys. **17**, 26 (1954).
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V. ACKNOWLEDGMENTS

We wish to thank the crew of the 184-inch cyclotron for the proton bombardments of niobium foil required in this research. One of us (M.G.F.) acknowledges the support of the U. S. Air Force Institute of Technology, Wright-Patterson Air Force Base, Dayton, Ohio. We also wish to thank Frank S. Stephens for considerable help on the scintillation counter measurements. James Schooley and Jose Juliano determined the electron spectrum of Fig. 3.

[15] Robertson, Scott, and Pool, Phys. Rev. **76**, 1649 (1949).

PHYSICAL REVIEW VOLUME 97, NUMBER 5 MARCH 1, 1955

Angular Distribution of the Resonance Fluorescence Radiation from the 411-kev Excited State of Hg^{198}*

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(Received November 16, 1954)

By using the Doppler broadening due to thermal agitation at elevated temperatures as a means of restoring the resonance condition, the angular distribution of the resonance fluorescence radiation from the 411-kev excited state in Hg^{198} has been measured. The observed distribution has the form

$$1+(0.37\pm0.06)P_2(\cos\theta)+(1.07\pm0.08)P_4(\cos\theta),$$

which agrees very well with the theoretical expectation for the excitation of a state with spin 2 from a ground state with spin 0.

INTRODUCTION

IN his paper on the directional correlation of successive quanta, Hamilton[1] pointed out that the correlation functions $W(\theta)$ should also apply if one or both transitions are absorptions rather than emissions. Thus $W(\theta)$ should give the angular distribution of resonance radiation excited by a unidirectional, unpolarized beam of gamma rays.

Over the past few years the investigation of the angular correlation of successive gamma rays has provided very valuable information on the excited states of nuclei. The success has been especially good for even-even nuclei because the value 0 of the spin of the ground state reduces the number of possible complications due to mixtures of multipoles to a minimum. For odd-A nuclei the interpretation of angular correlation data is sometimes rather ambiguous due to the many parameters determining $W(\theta)$. In this respect the study of the angular distribution of resonance fluorescence radiation offers considerable advantages: If the spin of the ground state is known, the only parameters entering into the calculation of $W(\theta)$ for resonance radiation are the J-value of the excited state and the multipole character of the transition. In the angular correlation case the J-values of two different excited states and the multipole character of the two gamma rays define $W(\theta)$. The number of parameters involved in the angular correlation case is thus in general considerably greater than the number involved in the angular distribution of resonance radiation.

With an ingenious experiment on Hg^{198}, Moon[2] and co-workers have recently demonstrated that under suitable conditions resonance fluorescence becomes a very prominent phenomenon which can be studied in detail. Moon[2] used the large peripheral speed of a centrifuge to compensate via Doppler shift for the small energy

* Assisted by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.
[1] D. R. Hamilton, Phys. Rev. **58**, 122 (1940).

[2] P. B. Moon, Proc. Phys. Soc. (London) **A64**, 76 (1951); P. B. Moon and A. Storruste, Proc. Phys. Soc. (London) **A66**, 585 (1953); W. G. Davey and P. B. Moon, Proc. Phys. Soc. (London) **A66**, 956 (1953).