of relative intensity two percent, logft is found to be 8.0. The same quantity for transitions to the excited levels has the value 6.5. These values of logft suggest that all the transitions to Tl^{203} are first forbidden[22] ($\Delta I=0\pm1$; ±2, yes!); this is additional confirmation that the ground state of Pb^{203} should be $5/2(-)$.

The complete level scheme of Tl^{203}, as derived from the present measurements, is shown in Fig. 8. There is a similarity between the level scheme of Tl^{203} and those of odd-A isotopes of Au and Ir.[23] Other recent measurements[24,25] as well as these seem to indicate that $M1+E2$ mixtures are a general feature of transitions in odd-Z—even-N nuclei.

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Disintegration of Y^{91}

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The radiations of the fission product Y^{91} have been investigated with beta- and gamma-scintillation spectrometers. In addition to the well-known 1.55-Mev ground-state beta transition, there is a weak (0.3 percent) beta transition of energy 0.330 Mev which is followed by a gamma ray of energy 1.22 ± 0.01 Mev. Definite assignment of these radiations to Y^{91} was established through exacting chemical procedures, measurement of the half-life of the gamma ray, and energy balance considerations. There is reason to believe that the 1.22-Mev level of $_{40}Zr^{91}$ is not a simple single-particle state.

INTRODUCTION

THERE has been considerable confusion in the literature as to whether or not there is weak γ radiation associated with the 60-day Y^{91} activity.[1–3] The recent measurements of Boley and Dunavan[3] suggest that the 1.2-Mev gamma ray ascribed to Y^{91} by Langer and Price is actually due to an impurity whose half-life is 160 ± 30 days. Boley and Dunavan base their impurity argument not only on the half-life discrepancy but also on the fact that they found a beta-to-gamma ratio of ~20, whereas Langer and Price had found this ratio to be >1000. However, since an examination of highly purified Y^{91} (prepared at this laboratory) indicated the presence of 1.2-Mev gamma rays, it seemed worthwhile to investigate in detail the source of this gamma radiation.

APPARATUS

Scintillation spectrometers were used for the measurement of the beta- and gamma-ray spectra. NaI(Tl) crystals 2 inches in diameter and 2 inches thick were used for gamma detection, and a Pilot Plastic Scintillator-B[4] phosphor $1\frac{3}{4}$ inches in diameter and $\frac{3}{4}$ inch thick was used for beta detection. The scintillators were coupled directly to Dumont type 6292 photomultiplier tubes. Each NaI(Tl) crystal was surrounded by a layer of MgO reflector. The MgO was held in place by a thin aluminum cup. The beta phosphor was covered with aluminum 0.0005 inch thick which served both as a light shield and a reflector.

A block diagram of the electronic circuitry used to obtain coincidence spectra is shown in Fig. 1. The twofold coincidence circuit has a resolving time of about 0.2 microsecond.

GAMMA-SPECTRUM

Preliminary examination of the gamma-ray spectrum of Y^{91} with the scintillation spectrometer revealed a weak photopeak at about 1.2 Mev and an intense, hyperbolic-shaped distribution at lower energy which had an apparent endpoint in the neighborhood of 1 Mev. The bulk of the latter distribution was attributed to internal and external bremsstrahlung resulting from

[1] N. E. Ballou, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1950), Paper No. 116, Natl. Nuclear Energy Ser., Plutonium Project Record **9**, Div. IV.
[2] L. M. Langer and H. C. Price, Phys. Rev. **76**, 641 (1949).
[3] F. I. Boley and D. S. Dunavan, Phys. Rev. **90**, 158 (1953).
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emission of the well-known 1.55-Mev beta-ray group of Y^{91}. The scintillation spectrum shown in Fig. 2 was obtained with a source-detector geometry intended to minimize the contribution of the external bremsstrahlung. The source, which had a surface density of <0.2 mg/cm^2, was mounted on a thin rubber hydrochloride film and was positioned about 6 inches in front of the NaI(Tl) crystal. The only material (other than air) within several feet of the source which intercepted direct beta particles was a 0.25 inch thick Lucite beta-ray shield attached to the front of the crystal.

The photopeak appearing in Fig. 2 at a pulse height of about 87 volts is from a gamma ray of energy 1.22 ± 0.01 Mev. This energy value is based on a calibration of the scintillation spectrometer with the 1.277-Mev[5] gamma ray of Na^{22}. No other photopeaks could be found. A very careful search was made in the vicinity of 0.2 Mev, since a gamma ray of this energy had previously been assigned to Y^{91}.[2] The pulse height at which a 0.2-Mev photopeak would occur is indicated in Fig. 2. If there were equal numbers of quanta of energy 1.22 and 0.2 Mev, the 0.2-Mev photopeak would have a height about four times the height of the observed continuum at 0.2 Mev. If a 0.2-Mev gamma does exist, which seems very doubtful on the basis of gamma-gamma coincidence measurements described below, its intensity is certainly less than 5 percent of that of the 1.22-Mev gamma.

The source used in the above experiment was prepared by evaporating several drops of a stock solution of carrier-free Y^{91} obtained from Oak Ridge National Laboratory. As a preliminary check on whether any part of the spectrum of Fig. 2 could be due to source contamination, a portion of the Oak Ridge source material was subjected to additional chemical purification cycles and then reexamined with the scintillation spectrometer. No change was observed in the gamma spectrum or in the beta-to-gamma ratio. In addition, a source of long-lived fission product yttrium was prepared independently at this laboratory by chemical extraction from the gross fission products resulting from thermal neutron bombardment of a sample of U^{235}. The gamma spectrum of this source was found to be identical to that of the Oak Ridge source.

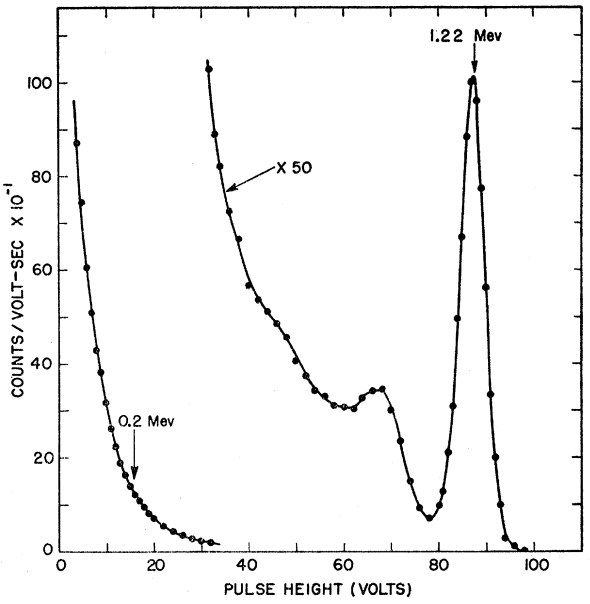

FIG. 2. γ-ray spectrum of Y^{91}, taken with a 2-inch NaI(Tl) crystal mounted on a Dumont type 6292 photomultiplier tube.

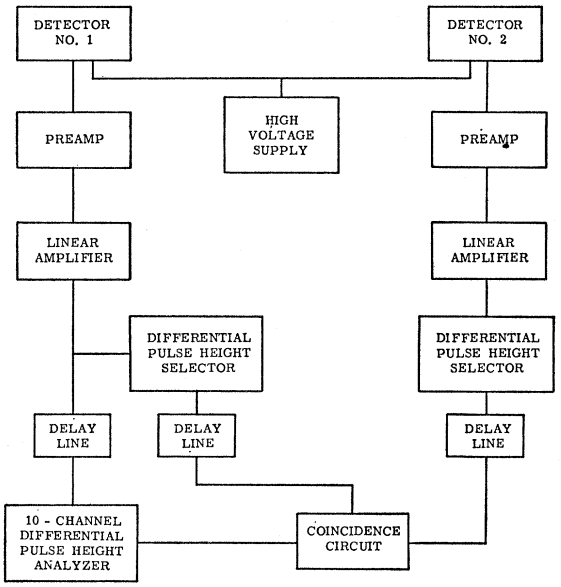

FIG. 1. Block diagram of the coincidence scintillation spectrometer.

Although the above experiments almost conclusively prove that the observed gamma spectrum is due entirely to the decay of a long-lived yttrium fission product (presumably Y^{91}), it was felt desirable to measure the half-life of the 1.22-Mev gamma ray before definitely assigning this gamma to Y^{91}. The decay data were taken with the scintillation spectrometer. The Y^{91} source used for this experiment was mounted rigidly inside a Lucite capsule which had wall thickness sufficient to stop the beta rays. An easily reproducible source-detector geometry was established which allowed alternate counting of the Y^{91} source and a Na^{22} monitor source. Each point on the decay curve (see Fig. 3) was obtained by first accurately calibrating the spectrometer and then measuring the total counting rate (minus background) above a discriminator setting corresponding in energy to 1.08 Mev which, in terms of pulse height, is at the minimum of the "valley" just to the left of the 1.22-Mev photopeak. Thus, the data of Fig. 3 represent essentially the decay of the 1.22-Mev photopeak. As a check

[5] D. E. Alburger, Phys. Rev. **76**, 435 (1949).

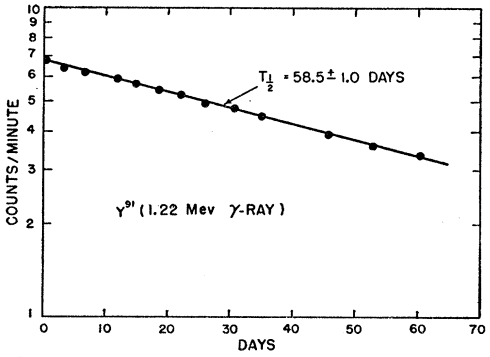

FIG. 3. Decay curve of the 1.22-Mev photopeak shown in Fig. 2.

on the long-term reliability of this counting arrangement, each time a Y^{91} measurement was made, the Na^{22} source was also counted using the same discriminator setting. All of the Na^{22} points lay within 2 percent of the calculated decay curve. Decay measurements were made over a period of two months, yielding a half-life for the 1.22-Mev gamma of 58.5±1.0 days which agrees quite well with the values reported for the half-life of Y^{91} (57 and 61 days).[6]

GAMMA-GAMMA COINCIDENCE SPECTRUM

Although there is no evidence from the simple pulse-height spectrum for any gamma rays except the one at 1.22 Mev, earlier measurements on Y^{91} had indicated that there were 1.2-Mev gamma—0.2-Mev gamma coincidences.[2] To investigate this possibility, a study was made of the gamma spectrum in coincidence with the 1.22-Mev gamma. The axes of the two scintillators were oriented at 90° to one another to eliminate coincidences resulting from 180° Compton scattering of the 1.22-Mev quanta. With the so-called "gate" spectrometer set to count all pulses representing 1.08 Mev or greater, all coincidences observed in the energy region 20 kev to 1 Mev were attributable to background plus chance coincidences. It is concluded that if gammas of energy other than 1.22 Mev are involved in the decay of Y^{91}, they are quite weak compared to the 1.22-Mev gamma and are probably not in coincidence with it.

BETA-GAMMA COINCIDENCE SPECTRUM

The above experiments indicate that there is a low intensity transition from Y^{91} to a 1.22-Mev excited state of Zr^{91}, implying that Y^{91} emits a weak beta group of end-point energy ~0.33 Mev. In order to verify this, the beta spectrum in coincidence with the 1.22-Mev photopeak was measured with the beta-scintillation spectrometer. The spectrometer was calibrated with the 624-kev K-conversion electron line of Ba^{137}. The coincidence counting rate was quite low, but the ratio of true to chance coincidences was >10 for points on the beta spectrum in the energy region below 200 kev. After correcting each point for energy loss[7] in the 0.0005-inch aluminum foil covering the beta scintillator, a Fermi plot was made of the coincidence spectrum. This is shown in Fig. 4. The end-point region has been corrected for the resolution of the spectrometer by the method of Palmer and Laslett.[8] The Fermi plot reveals only one beta-ray group with end-point energy 0.330±0.010 Mev, which confirms the hypothesis made at the beginning of this section.

DETERMINATION OF THE BRANCHING RATIO

The relative intensity of the 0.33-Mev beta group was determined by methods of absolute and comparison counting. A Na^{22} sample of known disintegration rate served as the comparison source. The source strength of 1.277-Mev gamma rays from this sample was determined by measuring the positron disintegration rate in a 4π proportional counter and then multiplying this result by 1.11 to correct for the K-capture branch to the 1.277-Mev level.[9] It was assumed, in agreement with the experiments of Sherr and Miller,[9] that Na^{22} K-capture events are not detected by the 4π counter. Two Y^{91} sources of very different strength were prepared from the same stock solution. The ratio of the activities of these two sources was known to be 5190 (±1 percent) by careful weighing of the amount of stock solution used for each source. The disintegration rate of the weaker source was measured in the 4π beta counter. Multiplication by 5190 then gave the disintegration rate of the stronger source. Next, the ga nma-ray spectra of the stronger Y^{91} source and the Na^{22} source were examined with the scintillation spectrometer. Identical source-detector geometries were used for each sample. The ratio of respective emission

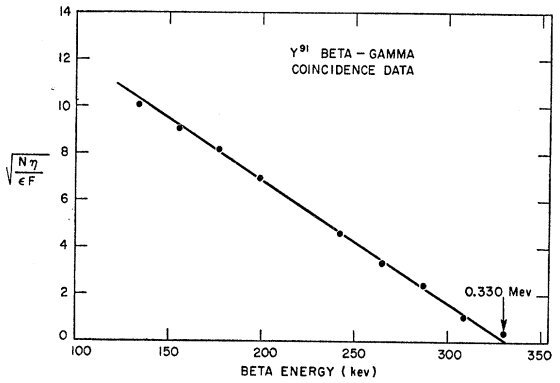

FIG. 4. Fermi plot of the Y^{91} beta spectrum in coincidence with the 1.22-Mev gamma ray.

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[7] H. Bethe, *Handbuch der Physik* (Verlag Julius Springer, Berlin, 1933), Vol. 24, Part 1, pp. 519–523.

[8] J. P. Palmer and L. J. Laslett, Atomic Energy Commission Report AECU-1220, March 14, 1951 (unpublished).

[9] R. Sherr and R. H. Miller, Phys. Rev. **93**, 1078 (1954). Private communication from Dr. R. Sherr to Dr. R. B. Day. We wish to thank Dr. R. Sherr and Mr. R. H. Miller for making their results available before publication.

rates of Y^{91} 1.22-Mev gamma rays and Na^{22} 1.277-Mev gamma rays was determined by comparing the areas of the two photopeaks and correcting for photopeak sensitivity of the crystal according to an empirical curve based on previous measurements of monoenergetic gamma-ray activities of known intensities. When the results of the experiments described above are combined, a value of 0.30 ± 0.05 percent is obtained for the strength of the Y^{91} decay branch involving the 0.33-Mev beta transition and the 1.22-Mev gamma transition.

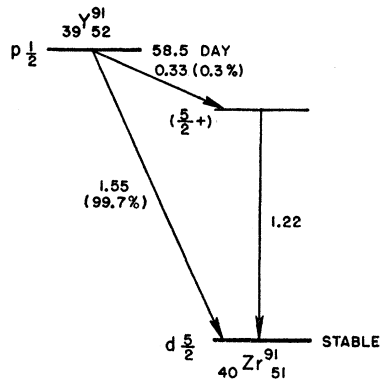

FIG. 5. proposed decay scheme of Y^{91}.

DISCUSSION

The decay scheme indicated by the measurements described above is shown in Fig. 5. Since the $_{40}Zr^{91}$ nucleus has only one particle outside a "magic" core, it seems reasonable to expect that the single-particle shell model would be particularly well suited for helping choose possible spin states for the low-lying levels of this nucleus. Odd-neutron levels in this shell are $d_{5/2}$, $g_{7/2}$, $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$. The measured ground-state spin of Zr^{91} is 5/2, in accord with the $d_{5/2}$ shell-theory level. One can arrive at possible spin assignments for the 1.22-Mev level through consideration of beta-ray selection rules. The 1.55-Mev beta transition from Y^{91} has been identified as first forbidden $\Delta I=2$(yes),[10] so that the ground state of Y^{91} must be $p_{1/2}$, which is again consistent with shell-model predictions. The 0.330-Mev beta transition of Y^{91} has an associated logft value of 8.80, which classifies the transition as first forbidden. If the spin change involved is $\Delta I=0$, 1(yes), a single-particle assignment of $d_{3/2}$ or $s_{1/2}$ could be made for the 1.22-Mev level. However, it is noted that a logft value of 8.80 is considerably higher than one would expect on an empirical basis[11,12] for a first forbidden transition involving a spin change of 0 or 1. Instead, since $\log[(W_0^2-1)ft]=9.03$, the value of the comparative half-life falls well within the range of values for transitions of the type $\Delta I=2$(yes). Thus, the 0.330-Mev beta spectrum may have a unique first forbidden shape. It is not possible to answer this question on the basis of the beta-ray data shown in Fig. 4 because of the relatively poor resolution of the spectrometer. If the low-energy beta transition is indeed of the $\Delta I=2$(yes) type, one would expect the spin and parity of the 1.22-Mev level of Zr^{91} to be 5/2+. Since the $d_{5/2}$ level has already been assigned to the ground state, a 5/2+ excited state is not interpretable as a simple odd-particle level. It is quite possible that the 1.22-Mev level is associated with an excitation of the even-even core. In this connection, it is interesting to note that the first three excited states of Mo^{93} (also a 51-neutron nucleus) seem to result from core excitation.[13,14] The fact that study of the proton groups associated with the reaction $Zr^{90}(d,p)Zr^{91}$ seems to give no evidence for a level in Zr^{91} at 1.22 Mev[15] may be taken as an indication that this state has an unusual character.

The authors are indebted to Dr. Charles W. Stanley and Dr. Rene J. Prestwood for making the necessary chemical separations.

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[11] Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. **23**, 315 (1951).
[12] L. W. Nordheim, Revs. Modern Phys. **23**, 322 (1951).
[13] M. Goldhaber, Phys. Rev. **89**, 1146 (1953).
[14] J. J. Kraushaar, Phys. Rev. **92**, 318 (1953).
[15] F. B. Shull and C. E. McFarland, Phys. Rev. **87**, 216 (1952).