Radiations from Zr^{89}*†

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A betatron-induced γ, n reaction on Zr^{90} has been found to produce a third branch in the decay of the 4.4-minute state of Zr^{89}. The new branch includes a 1.53-Mev gamma ray (8 percent as intense as the 588-kev gamma ray) coincident with positrons of approximately 850-kev maximum energy. These particles (log ft$=4.2\pm0.1$) are in competition with the 2.4-Mev positron group (log ft$=6.9\pm0.3$) emitted in the decay, to the ground state of Y^{89}, of the 4.4-minute Zr^{89m}. Anomalous behavior of the comparative half-lives is indicated, since both the 2.4-Mev and the 901-kev, 79-hour positron transitions are thought to be allowed. $K/(L+M)$ conversion ratios for the 4.4-minute, 588-kev and the 79 hour, 913-kev gamma radiations are, respectively, 5.4 ± 0.7 and 7 ± 2. A ten-channel scintillation spectrometer, based on a cathode-ray tube, light pipes, and photomultiplier tubes, to effect pulse-height selection, is also described.

I. INTRODUCTION

RECENT experiments by Shure and Deutsch,[1] and Goldhaber *et al.*[2] have shown that the 79-hour activity in Zr^{89} decays mainly with the emission of a positron of about 900-kev maximum energy, followed by a 14-second gamma ray of similar energy. As a result of considerations of lifetime, energy, and total conversion coefficient for the gamma ray, the multipolarity of the radiation was set by them as magnetic 2^4-pole ($M4$). Preliminary findings concerning the associated 4.4-minute decay process in Zr^{89} have been reported[3] by this laboratory. In this work it was found that the 4.4-minute state of zirconium (interpreted as an isomer of Zr^{89}, and designated as Zr^{89m}) decayed, principally by the emission of partially-converted, 588-kev gamma radiation to the ground state of Zr^{89}, and somewhat by the emission of a high-energy positron of about 2.4-Mev maximum energy. In the course of that work some of the salient features of the 79-hour activity were confirmed.

Subsequent measurements in this laboratory, of the 4.4-minute activity, have revealed the presence of an additional gamma ray of 1530-kev energy, apparently following a positron group of about 850-kev maximum energy. The intensity of the 850-kev transition is deduced to be over ten times that of the 2.4-Mev transition to the ground state of Y^{89}. Consequently an abnormally low log ft value results if the low-energy group is indeed concerned with Zr^{89}, and not with another isotope or an impurity. Since the association of the low-energy group with Zr^{89m} has such disturbing consequences, it was necessary to investigate in detail whether or not the transition is concerned with Zr^{89m}. A number of experiments, performed in order to accomplish this end, are described below.

A ten-channel scintillation spectrometer, employing a cathode-ray tube, together with associated light pipes and phototubes as a discriminator, found extensive use in the present experiments. A discussion of the design and properties of this instrument comprises most of Sec. II.

II. APPARATUS

In the course of this work use was made of two magnetic beta-ray spectrometers, a 180° type previously described,[4] and a 255° double-focusing type with a 15-cm mean-radius of curvature. Coincidence measurements were made with scintillation detectors employed in conjunction with either of two coincidence circuits. The faster of these had a resolving time of 2×10^{-8} second. Coincident pulses were selected in a Rossi type mixer using crystal diodes. The second circuit, with a resolving time of approximately a microsecond, employed blocking oscillators for pulse shaping, and a $6AS6$ tube to select coincident pulses.

Extensive use was made of a 10-channel scintillation spectrometer in which pulse-height selection was effected by means of a gated-beam cathode-ray tube (C.R.T.) on the screen of which were disposed 10 light pipes, and associated photomultiplier tubes and circuits. In Fig. 1(a) there is presented a block diagram which indicates the arrangement of the parts of the pulse-height discriminator. In Fig. 1(b) is shown the time sequence and flat-topped shape[5] of pulses which occur on the vertical deflecting plates and intensifier grid of the C.R.T. for two input pulses of different amplitude. Corresponding to two input pulses of Fig. 1(b), there are observed on the screen of the C.R.T., two stationary light spots. These differ in vertical

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[1] K. Shure and M. Deutsch, Phys. Rev. **82**, 122 (1951).

[2] Goldhaber, der Mateosian, Scharff-Goldhaber, Sunyar, Deutsch, and Wall, Phys. Rev. **83**, 661 (1951).

[3] Shore, Bendel, and Becker, Phys. Rev. **83**, 688 (1951).

[4] Brown, Bendel, Shore, and Becker, Phys. Rev. **84**, 292 (1951).

[5] The pulse shaping circuit is that of D. A. Watkins, Rev. Sci. Instr. **20**, 195 (1949).

displacement and temporal occurrence, but are the same in duration and brightness. These light pulses are conducted through different light pipes to separate photomultiplier tubes (Type 931-A). The light pulses are than transformed into electrical signals which actuate scales-of-10 and mechanical registers.

A channel acceptance window is determined mainly by the relative sizes of the light spot and of the defining aperatures for the light pipe. When the light spot is only partially within the acceptance window of the light pipe, an uncertainty arises in the triggering of the scalers. This uncertainty may be large if the spot size is comparable to that of the light pipe. It was found that with a channel acceptance window having approximately 6 times the diameter of the spot, and with channel-center separations of 8 spot diameters, a pulse-height discriminator resulted for which effective widths of single channels varied by as much as 1 percent per hour. This permitted energy and intensity measurements to be made over periods of a few hours, after which it was advisable to check the channel widths.

A type 5CP5A C.R.T. was employed with the cathode at -1800 v and the post-accelerating electrode at $+1800$ v. The positive intensification pulses which were applied to the C.R.T. control grid were flat-topped with an amplitude of 35 v and a duration of 1.5 microseconds.

In order to test the performance of the discriminator, a pulse generator was employed. This gave flat-topped pulses of variable amplitude and 1-microsecond duration at a repetition rate of 60 per second. Shown in Fig. 2 is a typical channel profile. The shape is essentially that of a rectangle with narrow triangular wings, the latter comprising about 3.5 percent of the total area under the profile curve.

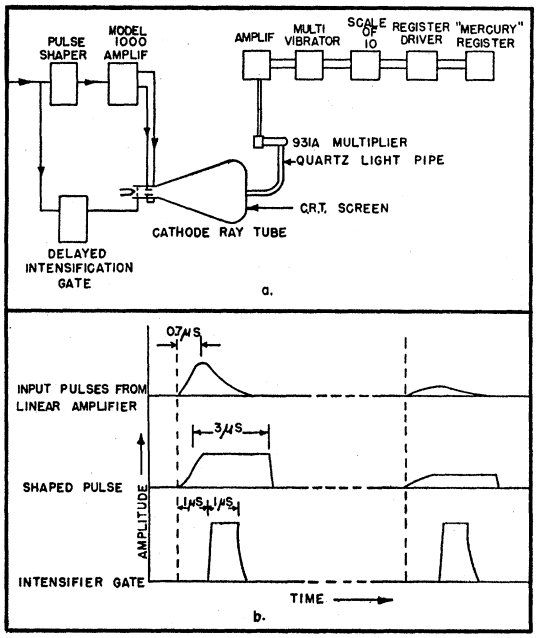

FIG. 1(a). Block diagram of the differential pulse-height discriminator. One channel is shown in the diagram; (b) Sequence of input, shaped, and intensifier pulses. Two different pulse heights are shown. [Note that the final shaped pulse arriving at the intensifier gate (bottom curves) is the same in both cases.]

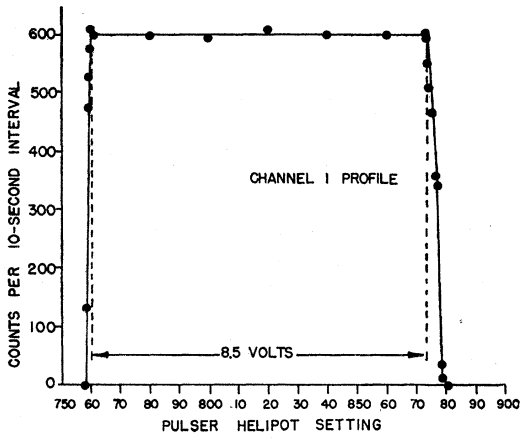

FIG. 2. Typical channel profile. For a given point, counts are recorded for 10 seconds with input pulses of fixed amplitude. The amplitude then is varied to obtain the remaining points.

The linearity of the positions of the channel centers was investigated by employing a constant amplitude input pulse and varying the position of the spot for zero pulse amplitude. It was found that the deviation of the separation of adjacent channels from the average separation did not exceed 2 percent. Keeping the sensitivity constant and varying the position of the spot for zero pulse amplitude, the instrument could be made to examine spectra comprising 28 channel-center separations with a linearity of better than 2 percent.

The scintillation detectors usually were NaI(Tl) crystals. These were mounted inside of air-tight, diffusely-reflecting units placed on top of 5819 photomultiplier tubes. The reflectors were thin Al hemispheres smoked on the inside with magnesium oxide. Both the crystal (cleaved in a dry box into parallelepipeds) and reflector were cemented to 1-mm thick glass disks by means of Gelva.[6] The power supply for the 5819 photomultiplier tube was stabilized to such an extent that over a period of 5 days the drift in output voltage was less than 0.05 percent. An Atomic Instrument Company, 204-C linear amplifier was employed to amplify the signals. It was modified by including crystal-diode limiters between certain stages in order to allow detection of small pulses in the presence of large ones.

III. DATA

Both the long and the short-period activities were produced in the 22-Mev betatron by means of the probe

[6] Made by Shawinigan Products Corporation, 350 Fifth Avenue, New York. The technique of J. S. Moenich and R. Swank of Argonne Laboratory, described by W. L. Buck in a private communication, was employed.

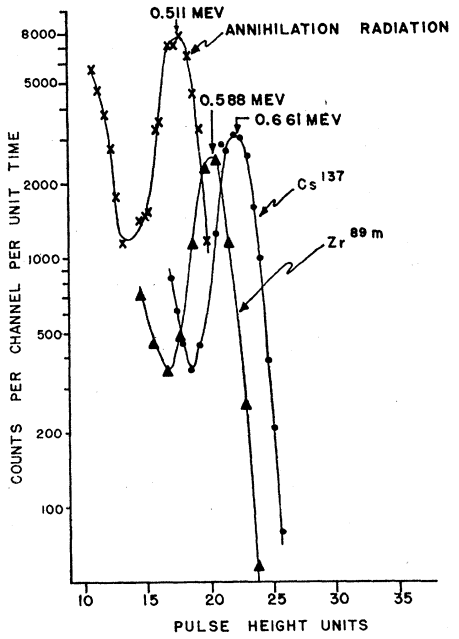

FIG. 3. Scintillation-spectrometer determination of the 588-kev gamma ray in Zr^{89m}. Comparison is made with annihilation radiation from Ag^{106} and with the 661-kev gamma ray following the decay of Cs^{137}.

technique[7] previously described. The sources usually were metallic foils (0.5 mil and 1.0 mil in thickness) of zirconium. For some of the irradiations, in order to check for the effect of impurities, chemically pure ZrO_2 was activated, due account being taken of the 126-second oxygen activity which was also produced. Finally, separated Zr^{90} was irradiated in order to eliminate the possibility of some of the radiations being associated with isotopes of zirconium other than Zr^{89}.

4. 4-Minute Radiations

1. Energy and Lifetime Measurements of the Gamma Radiation

The energies of the gamma radiation associated with the 4.4-minute activity were measured with the scintillation spectrometer mentioned above. The detector was a 1.2-cm cube of NaI(Tl). The instrument was calibrated by means of the 661-kev gamma ray of Ba^{137}, annihilation radiation, and the 1277-kev radiation from Ne^{22}. The results for the lower-energy range are displayed in Fig. 3. The peaks shown are produced by photoelectric absorption in the iodine of the crystal. Energywise they represent the total energy of the primary gamma ray, since most of the low-energy secondaries would be reabsorbed in the crystal. The center peak is due to 4.4-minute Zr^{89m}. Here a 1-mil zirconium foil was irradiated in the 22-Mev betatron. A mean of five determinations yielded a value of 588±3 kev. A measurement, described next, of the associated internal conversion line, with the 255° spectrometer yielded a value of 586±3 kev. Both figures agree with the previously reported value.[3]

Figure 3 furnishes a good picture of the resolution attainable with the scintillation spectrometer. The full width at half-maximum for the Ba^{137m} line is 12 percent, with a peak-to-valley ratio of 8.5. The upward trend of the points below the peaks is attributed to Compton absorption of the primary radiation. The peak counting rate of the 588-kev line in Fig. 3 was found to decay with a half-life of 4.5±0.5 minutes. This is in good agreement with the value 4.40±0.04 minutes found as a mean of four determinations of the half-life (primarily of the internal conversion electrons) employing a Geiger counter having a mica window of thickness 3.5 mg/cm².

The scintillation detector also revealed a photo peak corresponding to a high-energy gamma-ray line (not shown in Fig. 3). A mean of six determinations of the energy (including the use of separated zirconium isotopes, to be described below) yielded a value of 1.53±0.03 Mev. A mean of four determinations of the half-life of the high-energy radiation gave a value of 4.4±0.3-minutes, in good agreement with the half-life of the particles and of the 588-kev line. This suggests that the 1530-kev gamma ray is also associated with Zr^{89}.

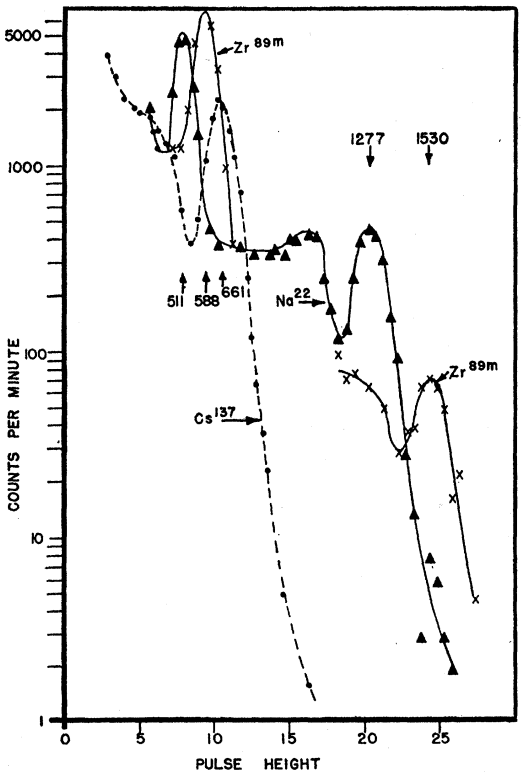

FIG. 4. Scintillation-spectrometer comparison of the gamma rays following the decay of Na^{22}, Cs^{137} with those of Zr^{89} produced from enriched $Zr^{90}O_2$.

[7] R. A. Becker, Rev. Sci. Instr. **22**, 773 (1951).

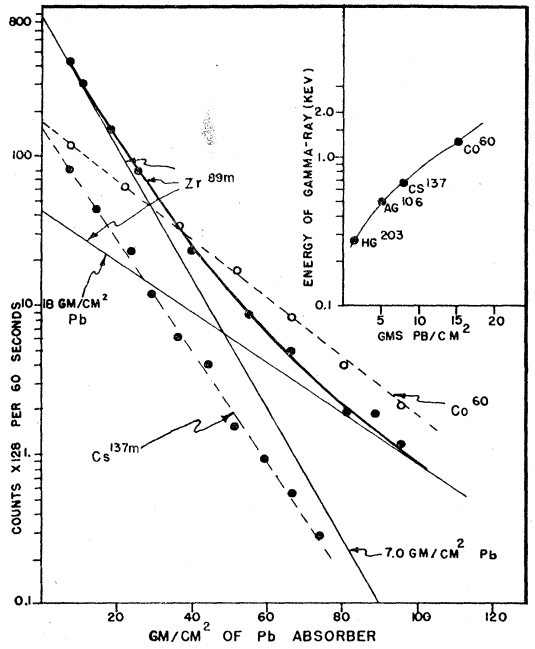

FIG. 5. Absorption in lead of the gamma radiation from Zr^{89m}. Shown for comparison are curves for Cs^{137} and Co^{60}. The inset shows the energy as a function of absorber half-thickness. The Zr^{89m} curve is decomposed into two straight lines corresponding to energies of 588 and 1530 kev.

2. *Confirmation of the 1530-kev Radiation*

It will be pointed out in Sec. IV, that the presence of the 1530-kev gamma ray, with an intensity as high as is here observed, introduces difficulty into the disintegration scheme. Consequently it was necessary to check carefully whether the 1530-kev radiation was real and associated with the decay of Zr^{89m}. That the radiation in question concerns the Zr^{90} isotope is supported by the fact that a source produced from enriched Zr^{90} ($Zr^{90}O_2$) yielded the same relative intensity for the 588 and 1530-kev radiation as did a source produced from commercial zirconium foil.

Data obtained from such an enriched source are presented in Fig. 4. The bump on the sodium calibration curve near 17 on the horizontal scale is interpreted as arising from Compton absorption of the 1277-kev radiation. Such "Compton peaks" accompany the photo lines in this type of instrument.[8] The effect of source decay was taken into account, and the relative peak heights at 588 and 1530 kev represent the apparent relative intensity of the two radiations, uncorrected for the relative efficiency of the detector.

Attention was also paid to such considerations as the adding together, or "pileup," of several pulses which occur close together in time, in such a way that the amplitude recorded for any one of them is increased by the presence of the others. Simultaneous detection of two quanta by the crystal may occur either if both are randomly distributed in time, or if they are "truly coincident" (such as being in cascade). The pulse amplitude which results in this case should be the sum of those for the component radiations. Evidence that this new gamma ray is not caused by such "pileup" in the crystal is furnished by the fact that the high-energy edge of the Ba^{137m} gamma ray is roughly a straight line over nearly three cycles of intensity in the semilog plot. In contrast it was found that the upper edge of the 588-kev curve from zirconium became nonlinear at 3 percent of the peak counting rate.

Additional verification that the 1530-kev gamma ray is real is obtained by noting that the decay rate and relative intensity of the two zirconium radiations are unchanged when the solid angle subtended by the detector at the source is varied.

The presence of radiation of energy greater than 588 kev is further confirmed by the Pb absorption curves, employing a scintillation detector, shown in Fig. 5. The source was 5 inches above the detector, which in turn was 15 inches above the top of a table. A Bakelite-lined Pb shield $\frac{5}{8}$ in. to $1\frac{1}{4}$ in. thick surrounded the detector in order to minimize scattering. The system was calibrated by means of data obtained with Hg^{203}, Cs^{137}, Ag^{106}, and Co^{60} sources in the decay of which are involved gamma rays, respectively, of energy 0.28, 0.66, 0.51,[9] and approximately 1.25 Mev. It is seen that the absorption curves for the Ni^{60} and Ba^{137m} radiations are linear over the ranges of absorber used, whereas that of the zirconium radiation is not, supporting the existence of radiation of energy higher than 588 kev. Making the assumption that the higher-energy radiation is indeed 1530 kev (18 g/cm²), results are obtained which are consistent with an energy of about 600 kev for the soft radiation.

3. *Relative Intensity of the Gamma Rays*

From the ratio of the extrapolated intercepts at zero absorber thickness, the relative intensity of the gamma rays was derived. A correction was made for the total absorption in the NaI of the two different energies. The result was $I_{588}/I_{1530}=12\pm3$.

The relative intensity was also determined with the scintillation spectrometer by means of a comparison of the zirconium radiation with the 1277-kev and annihilation quanta from Na^{22}. The ratio of the peak counting rates of the two 4.4-minute gamma rays, corrected for background and absorption in NaI, was compared to the two photopeaks obtained from Na^{22}. The ratio I_{511}/I_{1277} for Na^{22} was taken as $1.80_{-0.05}{}^{+0.25}$, since the gamma-ray transition in Ne^{22} follows positron emission from Na^{22} (10 percent K-capture assumed, although there is some experimental evidence[10]

[8] R. Hofstadter and J. A. McIntyre, Phys. Rev. **79**, 390 (1950); **80**, 631 (1950).

[9] Way, Fano, Scott, and Thew, *Nuclear Data*, Natl. Bur. Standards Circ. 499 (Government Prinitng Office, Washington, D. C., 1950).

[10] Good, Peaslee, and Deutsch, Phys. Rev. **69**, 313 (1946).

that there is no K-capture). By comparing the two ratios of corrected photopeak counting rates, the effect of counts appearing under the photopeaks via absorption of the secondary Compton photons in the NaI crystal can be largely canceled out. A small correction was applied, in comparing the two ratios, to account for the variation with energy of the photoelectric absorption of Compton secondary photons. Employing a $(1.2\text{-cm})^3$ crystal, the peak counting rates for the zirconium radiation were estimated to correspond to an intensity ratio of $I_{588}/I_{1530}=14_{-4}{}^{+5}$. An average of this value with that of the Pb absorption result gave a figure of 13 ± 3 for the relative intensity.

The effect of betatron energy on the relative intensity of the gamma rays was also considered. Irradiation with x-rays of maximum energy 23.5, 22, 21, 17.5, and 14 Mev gave no appreciable change in the relative intensity of the gamma rays. Thus the ratio remains essentially constant even close to the experimental threshold of about 12.5 Mev for the Zr^{89} activity. Accordingly, detailed considerations involving experimental thresholds, calculated thresholds using the tables of Metropolis and Reitwiesner,[11] and estimated Coulomb barrier heights show that the 1530-kev gamma radiation cannot arise from a $(\gamma, 2n)$, (γ, d), (γ, t), (γ, p), or (γ, α) photoreaction. (The γ, p threshold also can be deduced empirically from the present work. See Fig. 9 below.)

Evidence tending to disprove that the 1530-kev radiation arises from an n-capture reaction is afforded by varying the probe material and/or probe geometry in the betatron irradiations. Since the bremsstrahlung are confined to a narrow cone in the forward direction, and since the neutron flux is approximately spherically symmetric, it is to be expected that variations in the position[7] of the material being activated behind the probe will produce large variations in the relative intensity of photo to n-capture processes. Such variations were not found for the 588- and 1530-kev radiations. Actually when the target was moved $\frac{1}{2}$ in. back from the probe tip no appreciable activity was observed at all. Further evidence that an n-capture process is not involved is given by changing the converter material at the probe tip from lead to copper. Again no variation in the relative intensity of the 1530- and 588-kev radiation was observed. Evidence disproving the existence of an n-capture-produced activity of half-life less than 5 minutes in separated zirconium isotopes is also given elsewhere.[9]

4. *High-Energy Positrons*

Indications have been cited previously[3] for the existence of a weak positron group at about 2.5-Mev maximum energy. The existence of 4.4-minute positrons has been confirmed in the present work by the detection of 180° coincidences which were ascribed to pairs of annihilation quanta. Two anthracene crystals were situated $4\frac{1}{2}$ inches apart and a Zr source was situated midway between the crystals in a small hole in a lead sheet. The sheet was normal to the line joining the crystals and was covered with a thick enough sheet of aluminum to stop 2.5-Mev positrons. The lead sheet was of sufficient thickness to attenuate backscattered Compton radiation by a factor of 10^3. The coincidence and the single-counting rates both decayed with a half-life of 4.4 minutes. (A small isotropic coincidence rate was also observed when the source was slightly displaced laterally from the position in line with the crystals. This is confirmed below by the measurement of particle-gamma coincidences.)

In order to fix the energy of the 4.4-minute particles, two anthracene crystals (1 in.$\times$1 in.$\times\frac{1}{2}$ in.), separated by about $\frac{1}{16}$ in. were attached to the end of a 5819. With sources placed between the crystals, particles which were backscattered out of one crystal were detected in the other. In this way the pulse heights for the positrons from Ag^{106}, Cu^{62}, and Zn^{63} were compared with those from Zr^{89m}. Figure 6 presents Kurie plots, uncorrected for resolution, of the spectra. The three runs, shown for Zr^{89m}, were taken with the same source at different times. The inset shows ϵ_0 (maximum total energy) for the three known end points. From this the end point for Zr^{89m} is deduced to be 2.43 ± 0.06 Mev. The apparent nonlinearity of the derived curve for ϵ_0 may be a property of the two-crystal geometry.

In addition, the maximum energy and the sign of

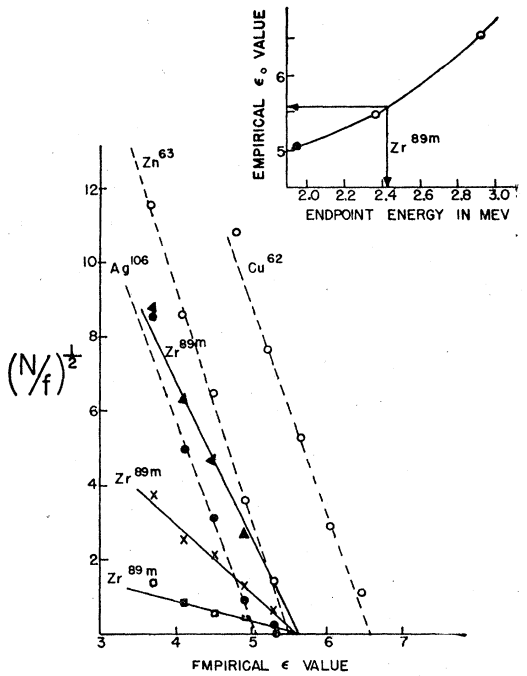

FIG. 6. Kurie plots comparing positrons from Cu^{62}, Zn^{63}, and Ag^{106} with Zr^{89m} in the split-crystal spectrometer.

[11] N. Metropolis and G. Reitwiesner, U. S. Atomic Energy Commission Report NP-1980 (unpublished).

the charge of the Zr^{89m} particles was confirmed by means of a Feather-absorption comparison with Cu^{62}, Zn^{63}, and Ag^{106} in the presence of a magnetic field.

5. *Particle-Gamma Coincidences*

For these measurements a resolving time of one microsecond was employed. Both the gamma and particle detectors were NaI(Tl) crystals, the former being 1.75 cm, and the latter 0.2-cm thick. A 6-g/cm² Pb absorber, interposed between the source and the gamma detector, attenuated the backscattered Compton photons. The source was a small piece of 1-mil zirconium foil. Aluminum absorbers were used to estimate the energy of the coincident particles. A study of the system was made with Co^{60}, Cs^{137}, Ag^{106}, and Hg^{203} to determine the counting rate *versus* amplifier gain characteristic of the gamma detector. The gain was so adjusted that the counting rate for 511-kev radiation was down by a factor of 2000 when the corresponding rate for the Co^{60} radiation was down by a factor of 5. In this way particle-annihilation-quanta coincidences were discriminated against.

Figure 7 shows the decay of the coincidences, for Al absorber thicknesses, on the particle side, of 0, 66, and 138 mg/cm². A half-life between 4 and 5 minutes is evident for each case. The data obtained with the singles rates (not shown), for both particles and gamma rays, confirmed the 4.4-minute half-life. The points plotted in the inset correspond to coincidence rates at zero time. A value of 59±6 mg/cm² of aluminum absorber was estimated as the half-thickness for the particles.

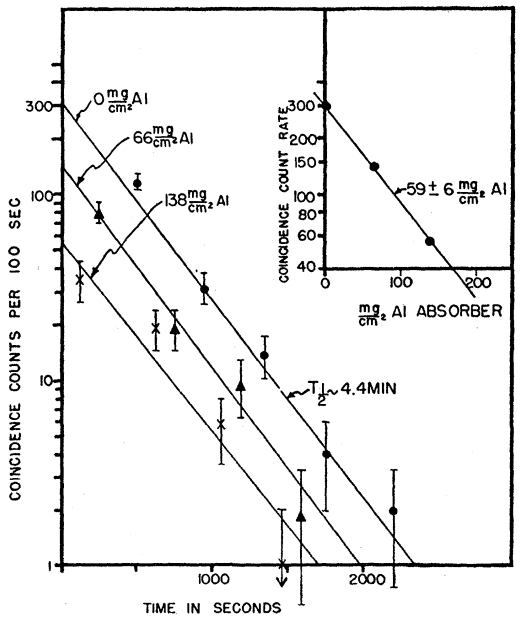

FIG. 7. Decay with time of coincidence counts for Zr^{89m} as a function of Al absorber placed in front of the particle counter. The gamma-ray counts are strongly biased against annihilation radiation. The inset shows the absorption curve of the particles.

Employing the same geometry, half-thicknesses were obtained (data not shown) for the positrons associated with Hg^{203}, and with 79-hour Zr^{89}. The measured half-thicknesses for these furnished the basis for an estimate of 850 kev as the end-point energy of the 4.4-minute particles. Similar results were obtained with the gamma counter biased still more strongly (annihilation radiation down by about 10^5). However, with the smaller coincidence rates the statistical uncertainties were larger.

Since the gamma spectrum in the scintillation spectrometer shows the presence of 588- and 1530-kev gamma rays only, the 1530-kev gamma ray must be involved in the coincidences with the 850-kev particle group since, in the present experiment, the 588-kev gamma rays would not be detected. As will be seen below, when the decay scheme is considered, this end-point energy is consistent with interpreting this group of particles as positrons. Additional support for this is obtained from the consideration of the relative intensities of the 4.4-minute x-rays from zirconium and yttrium.

6. *Relative Intensities of the Particles as Determined by Absorption*

The intensity of the 2.4-Mev positrons relative to the other 4.4-minute particles was derived from Al absorption measurements made with an end-window Geiger counter. In addition to penetrating radiation, there were two soft radiations which had half-thicknesses corresponding to beta energies of 2.9±0.6 Mev and 0.9±0.2 Mev. The latter group is thought to be comprised of both the 850-kev positrons and the conversion electrons from the 588-kev radiation, whereas the higher energy group is identified with the 2.4-Mev positrons. From the extrapolated counting rates at zero thickness, the relative numbers of 2.4-Mev to 850- and 570-kev particles is $0.06_{-0.02}^{+0.04}$.

7. *Critical Absorption of X-Rays and x/γ Intensity Ratio*

Since the decay of Zr^{89m} involves positron emission, K-capture, gamma radiation, and internal conversion, a knowledge of the relative intensity of x-rays to other radiations is desirable. The scintillation spectrometer revealed 4.4-minute x-rays of approximately 15 kev but, owing to the poor resolution, it was not possible, from the pulse-height distribution, to determine whether the radiation was characteristic of zirconium or yttrium. Critical absorption measurements, employing NaBr and RbCl, showed the presence of both Y and Zr x-radiation. The detector was a 0.1-cm thick NaI crystal on a 5819. The absorbers were placed close to the source to reduce detection of fluorescent x-rays characteristic of the absorber. An analysis of the results showed that the ratio of the intensities of the Y to the Zr x-rays is 1.4±0.5. The large uncertainty resulted mainly from

the difficulty in estimating how much background to subtract because of Compton events in the crystal.

An estimate was made of the intensity of the 4.4-minute x-rays relative to the 588-kev gamma rays in terms of the x-rays and 79-hour, 913-kev gamma rays. The method was to use two NaI scintillation detectors, one detector for x-rays (0.2 cm thick) and one for gamma rays (1.75 cm thick). The photomultipliers were so arranged that x and gamma radiation from the source alternately could be studied with the pulse-height discriminator. The data obtained were corrected for the instrumental variation, with energy, of the photopeak heights. This arose because of different primary absorption, in the crystal, of the two gamma rays via the photoelectric effect, and for the counts which appear under the photopeak via photoelectric absorption of the Compton secondary photons. The ratio of the two gamma-ray to x-ray ratios, $(N_g/N_x)_1$ and $(N_g/N_x)_2$, which were observed, respectively, for the 4.4-minute and 79-hour radiations, after the corrections were applied, was found to be 6.3 ± 2.

8. Relative Intensity of Positrons and Gamma-Rays

This was determined from 180° coincidence data following the method of Griffiths.[12] The anthracene crystals (1 in.$\times$1 in.$\times\frac{1}{2}$ in.) were 13 inches apart.

The relative efficiency of the detectors was determined from a knowledge of the singles and 180°-coincidence-counting rates when 24-minute Ag^{106} was studied with the same geometry. For this source, there is a 0.51-Mev gamma ray about 13 percent as abundant as the annihilation radiation which is present.[13] The relative efficiencies for the annihilation radiation, 588-kev, and 1530-kev gamma rays was estimated from the absorption data of Gillette[14] for anthracene.

The ratio of the positrons to 588-kev quanta was determined to be 0.019 ± 0.002. This included a correction for the presence of 1530-kev quanta to the extent of 8 percent of the 588-kev line.

9. $K/(L+M)$ Ratio for the 588-kev Radiation

Conversion electron spectra [see Fig. 8(a)] were studied with the 255° spectrometer using $\frac{1}{2}$-mil metal foils (8m/cm²). The solid curve represents the contributions from all shells. The dashed curves represent the derived K and $L+M+\cdots$ electron lines. The ratio was found to be $K/(L+M)=5.4\pm0.7$. This is less than the previously reported value of 7 ± 2 obtained with the 180° spectrometer.[3]

10. Total Conversion Coefficient for the 588-kev Gamma Ray

The total conversion coefficient for the 588-kev gamma ray was estimated by using two detectors.

[12] G. M. Griffiths, Phys. Rev. **83**, 852 (1951).
[13] W. L. Bendel (private communication).
[14] R. Gillette, Rev. Sci. Instr. **21**, 294 (1950).

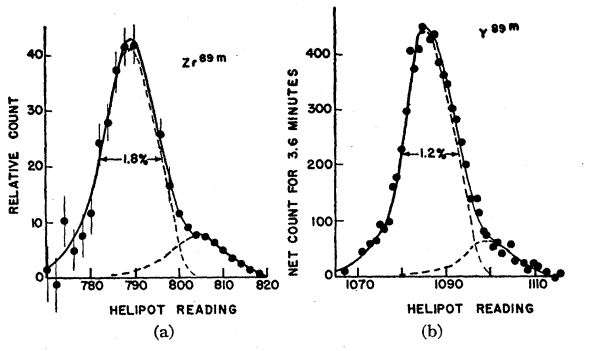

FIG. 8(a). Internal conversion electrons associated with the 4.4-minute, 588-kev transition; (b) Internal conversion electrons associated with the 913-kev transition following the 79-hour decay of Zr^{89}.

The intensity of the Zr^{89m} conversion electrons measured with one detector (less than 1-mm thick NaI) was compared to the intensity of the gamma rays measured in the other detector (1.2 cm-thick NaI). Employing the same geometry, a similar comparison was made for the Ba^{137m} 661-kev gamma ray. The particle line-spectra were superimposed on weak continua, and, for the Cs^{137} case, 5 percent of the area under the peak was subtracted to account for the 1.18-Mev particle continuum. For the Zr^{89m} case, a 10-percent contribution to the area under the electron peak, ascribed to the 850-kev positron group, was subtracted. The total conversion coefficient for Zr, computed with the aid of the results of Waggoner[15] and Bendel,[13] was $\alpha=0.076\pm0.014$.

79-Hour Radiations

1. Positrons and Conversion Electrons

The particle spectrum of the 79-hour activity was measured with the 180° magnetic spectrometer. The source was a 2-mm-wide strip of 1-mil ($\sim$16 mg/cm²) zirconium. The positron group reported previously[1,3] was confirmed, and found to have an end-point energy of 901 ± 10 kev, the difference between this value and the previous one[3] arising as a result of a better energy calibration of the spectrometer. The half-life for the particles was in agreement with the value of Shure and Deutsch.[1]

The 79-hour conversion line at 896 kev (913 ± 5 kev, upon adding the K binding energy of yttrium) has already been reported.

2. Gamma Radiation

The gamma radiation associated with the 79-hour activity was also investigated with the scintillation spectrometer, employing the 661-kev radiation of Ba^{137m} as a calibration. The measured value is 0.92 ±0.01 Mev, in good agreement with the more accurate measurement of the magnetic spectrometer.

[15] M. A. Waggoner, Phys. Rev. **82**, 906 (1951).

A careful search was made for gamma radiation of energy in the range 0.95 to 2.0 Mev, employing a separated Zr^{90} sample. None was found to be present. In particular, the gamma ray reported by Hyde and O'Kelley,[16] at 1256 kev, was not detected. The present measurements indicate that its intensity, if present, cannot be greater than one percent of the 913-kev gamma ray.

The relative intensity of the 913-kev and annihilation radiation was estimated in the same manner as that employed above for the 4.4-minute gamma radiation. The value for the ratio of 913-kev gamma rays to positrons was found to be 5 ± 1. This compares favorably with the value of Goldhaber *et al.*[2]

The decay with time of the 913-kev radiation was followed for several days with the scintillation spectrometer. By comparing the peak counting rates obtained with the zirconium to that with a Na^{22} source, a value of 79 ± 2 hours was obtained for the half-life. This is in good agreement with the more accurate value given by Shure and Deutsch.[1]

3. $K/(L+M)$ Ratio for the 913-kev Radiation

Figure 8(b) shows the conversion electron spectrum as obtained with the 255° spectrometer, employing an 8-mg/cm² source. The presence of the $(L+M)$ peak is derived by comparing, for comparable conditions, with the conversion lines associated with Cs^{137}. On the basis of relative peak heights the ratio is found to be 7 ± 2.

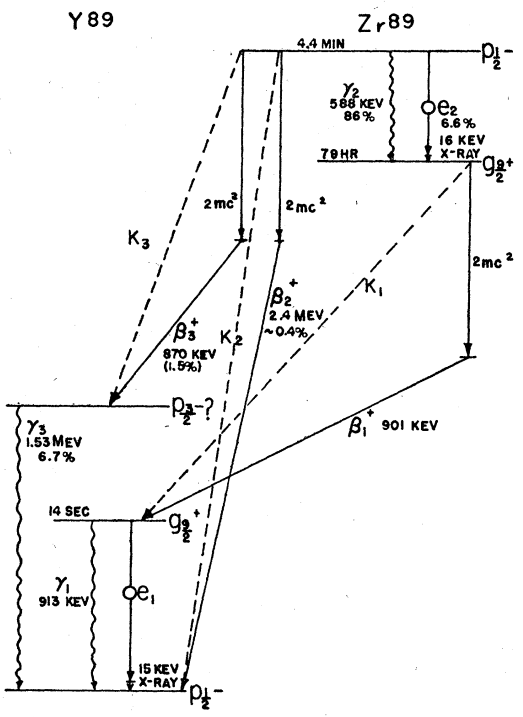

FIG. 9. Suggested decay scheme for the two zirconium activities. The intensities indicated are percentages of the total modes of decay of the 4.4-minute state of Zr^{89}.

IV. DISCUSSION

Figure 9 shows a tentative level scheme embodying the results of the present work and of the work previously reported.[1–3] The spin assignments and parities of the ground states and of the first excited states are as given before.[3] For the 588 and 913-kev gamma rays the conversion ratios determined in the present experiment provide additional confirmation that the two transitions are of the $M4$ type. The 1530-kev gamma ray and the 850-kev particles are indicated in the figure as a new branch of decay for 4.4-minute Zr^{89m}. The maximum energy of the particles is given in the diagram as 870 kev in order to yield consistent results for the energy difference between Zr^{89m} and Y^{89}, as determined from the remaining two branches of decay. It is to be recalled that the measured value, 850 kev, was subject to an uncertainty of 50 kev.

A compelling reason for placing the 4.4-minute state of Zr^{89} above the 79-hour state is the existence of several 4.4-minute branches, all of which have energetics consist with such a level scheme. Energy considerations also confirm that the 870-kev particles are positrons since the Nb^{89} ground state lies about 2.9 Mev above the Zr^{89} ground state (calculated from the tables of Metropolis and Reitwiesner[11]). Intensities are given for the radiation proceeding from the 4.4-minute state, each number representing a percentage of the total modes of decay of Zr^{89m}.

Much of the data employed in determining the relative intensities suffers because of limited counting rates and the short half-life involved. The gamma-ray coincidences at 180° gave, for the 4.4 minute radiations, $(\beta_2+\beta_3)/\gamma_2=0.019\pm0.002$ positron per 588-kev quantum. The absorption experiment yielded $\beta_2/(e_2+\beta_3)=0.06_{-0.02}{}^{+0.04}$. This, combined with the total conversion coefficient, $e_2/\gamma_2=0.076\pm0.014$, gave $\beta_2/\gamma_2=0.0054_{-0.0025}{}^{+0.0046}$. The experimental ratio, $\gamma_2/\gamma_3=13\pm3$, when combined with the theoretical[17] ratio of K-capture to positron emission, yields $\beta_3/\gamma_2=0.017\pm0.005$. Combining these, the relative intensities of the various 4.4-minute radiations are estimated to be $\gamma_2=0.86\pm0.03$, $\beta_2=0.0035_{-0.0015}{}^{+0.0024}$, $e_2=0.066\pm0.012$, $\gamma_3=0.067\pm0.012$, and $\beta_3=0.015\pm0.005$. These determine the comparative half-lives to be log $ft=6.9\pm0.3$ for β_2, and log $ft=4.2\pm0.1$ for (β_3+K_3).

The general features of the decay scheme are consistent with the measured x-ray intensities. The critical absorption experiment indicated that 4.4-minute x-rays of yttrium are 1.4 ± 0.5 times as numerous as are those of zirconium. An examination of the decay scheme reveals that only a negligible amount of the x-radiation

[16] E. K. Hyde and G. O'Kelley, Phys. Rev. 82, 944 (1951); University of California Radiation Laboratory Report UCRL-1064, 1950 (unpublished).

[17] E. Feenberg and G. L. Trigg, Revs. Modern Phys. 22, 399 (1950).

arises in transitions other than K_3 and e_2. Hence, as the fluorescent yields[18] for zirconium and yttrium are nearly equal, this gives $K_3/e_2=1.4\pm0.5$. Employing the theoretical β_3/K_3 ratio,[17] we have $\gamma_3/e_2=1.8\pm0.6$. Both L-capture and L-conversion have been neglected here. The branching ratios previously assigned indicate $\gamma_3/e_2=1.0\pm0.4$, which agrees roughly with the x-ray estimate.

When one examines the ft values for the three positron groups, anomalous behavior is apparent. Both the 901-kev group, $\log ft=6.1$, and the 2.4-Mev group, $\log ft=6.9$, are involved in allowed transitions, with no angular momentum or parity changes. It is evident that the comparative half-life in the second case is substantially larger than that for the first.

Even more unusual is the case of the comparative half-life for the transition to the 1.53-Mev Y^{89} state. In this instance $\log ft=4.2$, and evidently we are concerned with a highly allowed transition. Hence $\Delta I=0, 1$ and, from Gamow-Teller rules, no parity change is expected. Thus the 1.53-Mev state must be either $p_{\frac{1}{2}}$ or $p_{\frac{3}{2}}$. The choice of $p_{\frac{3}{2}}$ would be consistent with Mayer's shell model, since the ground state has already been designated as a $p_{\frac{1}{2}}$ state. However, the comparative half-life is 600 times smaller than that for the $p_{\frac{3}{2}}$ to $p_{\frac{1}{2}}$ transition to the ground state, for which $\Delta I=0$.

If, following Mayer's shell model, we examine the nuclear configurations corresponding to the various states of zirconium and yttrium involved in these transitions, it becomes difficult to explain all three of the comparative lifetimes solely on the basis of single-nucleon transformations.

The anomalous behavior noted for the present Zr^{89} transitions is similar to that in two other apparent exceptions, recently found, to the usual rules of beta decay. The other cases are Y^{87} to Sr^{87}, studied by Mann and Axel,[19] and Kr^{85} (and Sr^{85}) to Rb^{85}, studied by Sunyar, Mihelich, Scharff-Goldhaber, and Goldhaber.[20] The latter authors have suggested that the large ft values, found in these three cases for transitions which presumably are allowed, and in which all of the nuclei are in the region of near-closed shells, each can be interpreted as being due to a rearrangement of nucleons in the even-even core of an odd-A nucleus.

[18] Steffen, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949).

[19] L. G. Mann and P. Axel, Phys. Rev. **84**, 221 (1951).

[20] Sunyar, Mihelich, Scharff-Goldhaber, and Goldhaber, Phys. Rev. **86**, 1023 (1952).