3.14-hr Isomeric Level in Y^{90}†

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A previously unreported 3.14-hr isomeric activity has been produced in Y^{90} by thermal neutron irradiation of Y^{89}. The thermal capture cross section for the production of the isomer was measured to be 1.0 ± 0.2 mb. The decay of the isomeric level is characterized by the emission of two cascade gamma rays of 0.482 and 0.203 Mev. Conversion coefficient measurements indicated that the isomeric level at 0.685-Mev decays by $M4$ or $E5$ radiation to an intermediate level at 0.203 Mev followed by a predominantly $M1$ transition to the ground state of Y^{90}. Gamma–gamma directional correlation measurements and internal conversion measurements indicated that the level at 0.203 Mev has spin 3 with odd parity and that the level at 0.685 Mev has spin 7 or 8 with even parity. A shell-model configuration of $(g_{9/2}d_{5/2})$ has been assigned to the isomeric state.

I. INTRODUCTION

IN a study of the decay of 3.6-hr Y^{92}, Cassatt and Meinke[1] observed two intense gamma rays of 0.21 and 0.475 Mev. Recent studies[2] at this laboratory of the decay of Y^{92} indicated that no gamma rays of these energies were associated with the decay of this nuclide. This discrepancy suggested the existence of another yttrium activity having a half-life of approximately 3 hr. In our work, the Y^{92} was chemically separated from gross fission products. Since Cassatt and Meinke used source material produced by chemically separating yttrium from deuteron-bombarded zirconium, the different methods of source preparation could account for the fact that this activity was not observed in our work.

As a result of this discrepancy, a search for a new yttrium activity was undertaken. It was found that neutron irradiation of Y^{89} produced a 3.14-hr activity characterized by the emission of gamma rays of 0.203 and 0.482 Mev. Lyon, Eldridge, and Bate[3] have also recently reported a 3-hr activity resulting from neutron irradiation of yttrium.

Extensive experimental investigations carried out at this laboratory have established that this activity arises from the decay of a previously unreported isomeric level at 0.685 Mev in Y^{90}.

II. EXPERIMENTAL METHOD AND RESULTS

A. Source Preparation

Sources of the short-lived activity were prepared by irradiating samples of Spectroscopic grade Y_2O_3 in a thermal-flux facility of the MTR and in the core of the EBR I.[4] The EBR (experimental breeder reactor) produces a fission spectrum with relatively few thermal neutrons. A chemical separation was performed on the irradiated material. The short-lived activity was observed to follow the yttrium fraction, indicating that the activity was associated with yttrium. Samples of this material irradiated with thermal neutrons also contained activities from rare-earth contaminants. It was possible to remove these contaminants by purification, using ion-exchange techniques. Sources free from contaminating activities were also produced by irradiation of mass-separated yttrium, obtained from the Oak Ridge National Laboratory. Analyses of the decay of sources produced both by thermal and fast neutrons indicated two major components with half-lives of approximately 3 hr and 64 hr (Y^{90}). Although the cross sections for the production of these activities were much reduced in the fast reactor, the ratio of the 3-hr activity to 64-hr Y^{90} was increased by a factor of about 10. This provided a cleaner source and permitted a more precise study of the conversion-electron spectrum associated with the decay of the 3-hr activity than would otherwise have been possible. For this reason, most of the sources used in these studies were prepared by irradiation with fast neutrons. Sources for both gamma-ray and beta-ray measurements were prepared by drying a Y_2O_3 slurry on VYNS films ($50\ \mu g/cm^2$).

B. Gamma-Ray Measurements

The gamma radiation emitted by irradiated yttrium samples was observed with 3-in.×3-in. cylindrical NaI(Tl) detectors. Gamma-ray sources were counted at 10 cm on the vertical axis of the detector, using a 1.18-g/cm^2 Be absorber to absorb the Y^{90} beta rays. The general procedures used at this laboratory for precision quantitative gamma-ray spectrometry have been previously described.[5]

A typical pulse-height spectrum obtained from the irradiated yttrium samples is shown in Fig. 1. Gamma rays were observed at 0.203 ± 0.003 and 0.482 ± 0.005 Mev. In addition, a peak was observed at 0.685 Mev

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] W. A. Cassatt and W. W. Meinke, Phys. Rev. **99**, 760 (1955).

[2] R. L. Heath, J. E. Cline, and S. D. Reeder, MTR-ETR Tech. Branches Quart. Progr. Rept.-1st Quarter, 1960, Atomic Energy Commission Report IDO-16633 (unpublished).

[3] W. S. Lyon, J. S. Eldridge, and L. C. Bate, Oak Ridge National Laboratory Rept. ORNL-2866 (unpublished).

[4] C. Eggler, C. M. Huddleston, V. E. Krohn, and G. R. Ringo, Nuclear Sci. and Engr. **1**, 391 (1956).

[5] R. L. Heath, C. W. Reich, and D. G. Proctor, Phys. Rev. **118**, 1082 (1960).

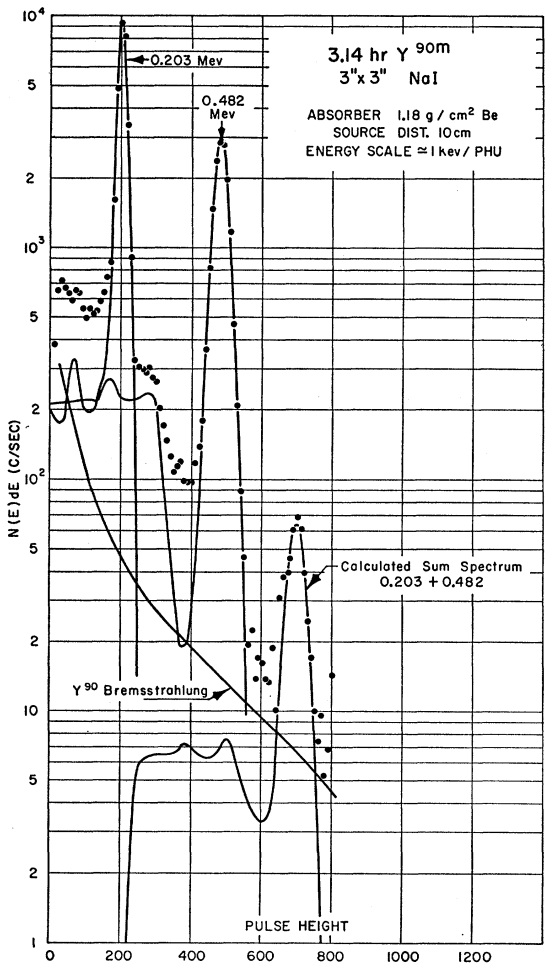

FIG. 1. Gamma-ray spectrum of 3.14-hr Y^{90} showing sum spectrum.

which was due to coincidence summing between the two gamma rays. Figure 2 shows a similar spectrum obtained with an increased amplifier gain to show the 0.015-Mev yttrium K x ray produced following internal conversion of the two gamma rays. To minimize the absorption of the x ray, this spectrum was obtained with only the 0.005-in. aluminum can surrounding the NaI detector. The figure shows the spectrum after removal of the contribution from Y^{90}.

Relative intensities of the gamma rays were obtained by successive subtraction of pulse-height distributions which represented the response of the detector to monoergic radiation for the particular geometrical arrangement used in making these measurements. This method is discussed in some detail in the *Gamma-Ray Spectrum Catalog*.[6] Relative emission rates were obtained using calculated detector efficiencies and experimentally determined photo peak efficiencies. Correction factors for absorption in the Be absorber and the aluminum detector can were experimentally determined using monoergic gamma-ray and x-ray sources. The results of the gamma-ray analyses are listed in Table I. In Fig. 1 the response of the detector to the individual gamma rays is represented by solid lines. The Y^{90} bremsstrahlung spectrum was obtained following the decay of the 3-hr isomer. The shape representing the coincidence sum spectrum produced by simultaneous detection of coincident events in the detector was calculated using a computer program to sum over the two gamma-ray distributions.[7] From a knowledge of the geometry, it was possible to calculate the shape and intensity of the coincidence sum spectrum produced by the two coincident gamma rays. Within the statistical fluctuations of the experimental measurement, the 0.685-Mev peak is entirely due to this coincidence summing effect. In view of the

TABLE I. Gamma rays and relative intensities.

E_γ (Mev)	Relative intensity
0.015 (x-ray)	0.109±0.001
0.203±0.003	1.00
0.482±0.005	0.957±0.004
0.685	<0.01

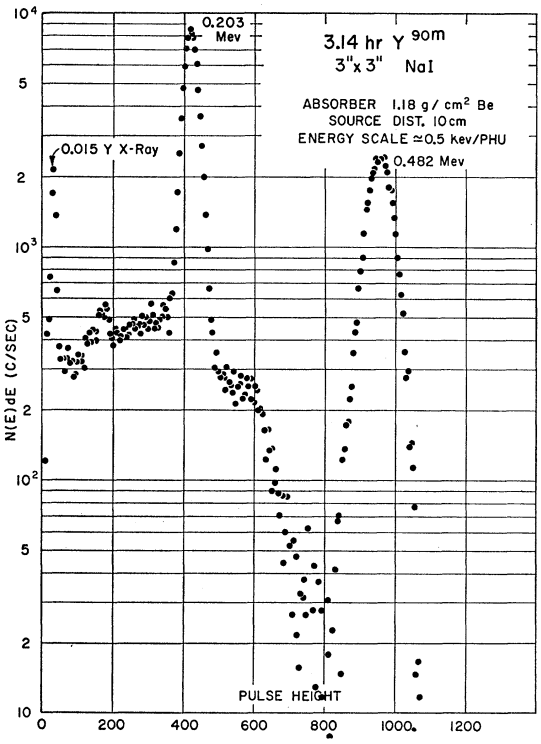

FIG. 2. Gamma-ray spectrum of 3.14-hr Y^{90} showing x-ray energy region.

[6] R. L. Heath, Atomic Energy Commission Rept. IDO-16408, (unpublished).

[7] A detailed description of this program is given in the following reference: E. C. Yates, R. L. Heath, and C. S. Pea, MTR-ETR Tech. Branches Quart. Progr. Rept.-3rd Quarter, 1960, Atomic Energy Commission Rept. IDO-16658 (unpublished).

theoretical significance of the intensity of the crossover gamma ray, additional measurements were made at reduced geometry to minimize summing. The upper limit for the intensity of this transition listed in Table I was obtained from these measurements.

C. Half-Life Determination

The half-life of the yttrium activity was obtained by observing the decay of the gamma-ray spectrum. Figure 3 shows a plot of the decay of the integrated pulse-height spectrum with background subtracted. Least-squares analysis of these data indicates the presence of two components with half-lives of 3.14±0.10 hr and 64.0 hr. The long-lived component is attributable to bremsstrahlung from the decay of Y^{90}.

D. Beta-Ray Measurements

Beta-ray and conversion electron measurements were made using a $1\frac{1}{2}$-in. diameter by $\frac{1}{4}$-in. anthracene detector covered with a 50 $\mu g/cm^2$ aluminum light reflector. Energy calibration of the scintillation detector was achieved by comparison with conversion lines of Ba^{137m} and Bi^{207m}.

A typical beta spectrum obtained from a sample of Y^{89} irradiated in the EBR I is shown in Fig. 4. The gamma-ray response of the detector has been subtracted. In addition to the 2.27-Mev ground-state beta ray associated with the decay of Y^{90}, conversion lines

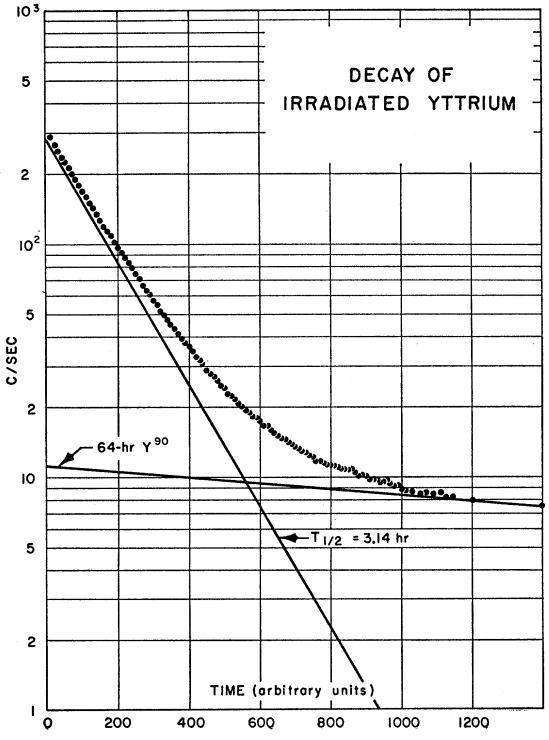

FIG. 3. Decay of 3.14-hr yttrium gamma rays for half-life determination.

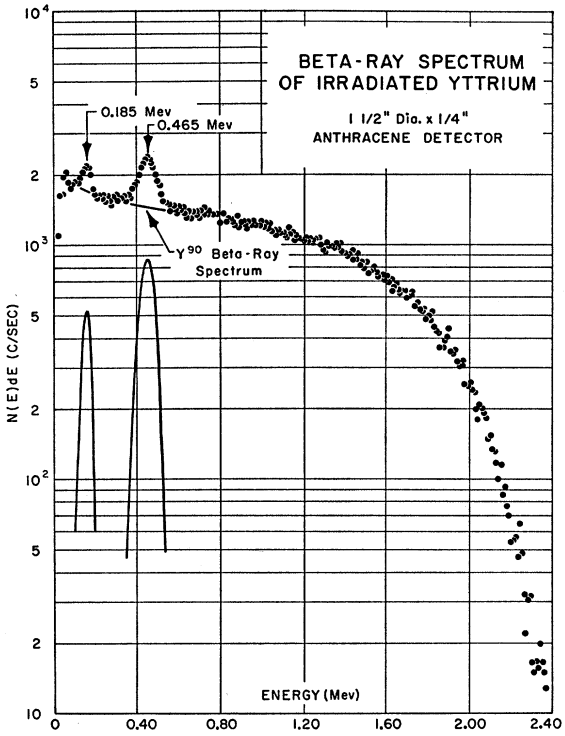

FIG. 4. Spectrum of beta radiation from yttrium sample.

are observed at 0.185 and 0.465 Mev corresponding to *K*-shell internal conversion of the 0.203- and 0.482-Mev gamma rays. No evidence of any beta group associated with the 3.14-hr activity was observed, and the conversion lines decayed with the 3.14-hr half-life.

E. Coincidence Measurements

Gamma-gamma coincidence measurements were made using a pair of 3-in.×3-in. NaI detectors. Beta–gamma coincidences were obtained using a $1\frac{1}{2}$-in.×$\frac{1}{4}$-in. anthracene detector in conjunction with one of the gamma-ray crystals. Graded back-scatter shields were used, and all measurements were made at 90° in the plane of the detectors. A "fast-slow" coincidence circuit with a fast resolution of 2×10^{-7} sec. was employed. The coincidence spectrometer consisted of a single-channel pulse-height analyzer operated in coincidence with a 256-channel transistorized analyzer of conventional design. Double-differentiated delay line linear amplifiers were used on both channels to allow the use of the "zero crossover point," characteristic of the pulse shape produced by this type of amplifier, to derive timing signals for the fast coincidence gates. This method, as developed by Fairstein,[8] eliminates the amplitude-dependent time jitter which is characteristic of conventional trigger circuits. Using this system, 100% co-

[8] E. Fairstein, Oak Ridge National Laboratory Instrumentation Division Annual Progress Report ORNL-2480, July 1, 1957 (unpublished).

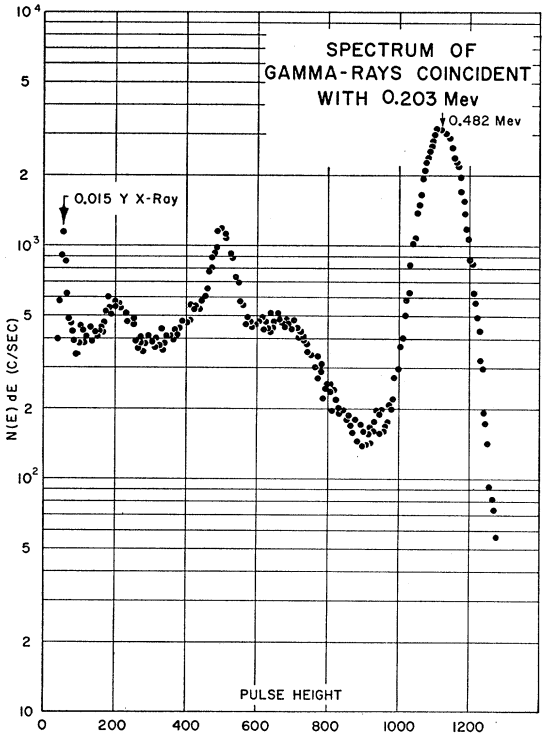

FIG. 5. Spectrum of gamma rays coincident with 0.203-Mev photopeak.

incidence efficiency for all pulses from 1 to 100 v in amplitude can be achieved with a fast resolving time of 2×10^{-7} sec. An example of the performance of this system is shown in Fig. 5, which is the spectrum of gamma rays in coincidence with the 0.203-Mev gamma ray. The 15-kev yttrium K x ray resulting from internal conversion of the 0.482-Mev gamma ray is clearly indicated.

Results of the gamma–gamma coincidence measurements indicated that the two gamma rays were 100% in coincidence. There was no evidence for any additional weak transitions. Beta–gamma coincidence measurements showed only the appropriate conversion line in coincidence with each gamma ray and showed no evidence for any beta ray associated with the decay of the 3.14-hr activity. The absence of any beta radiation associated with the observed activity indicates that it arises from the decay of an isomeric level.

F. Isotopic Identification and Measurement of Capture Cross Section

On the basis of arguments presented above, the 3-hr activity was attributed to the decay of an isomeric level in yttrium. To permit an isotopic assignment, irradiations of mass-separated Y^{89} were made in a thermal-flux facility of the MTR both with and without cadmium shields. The cadmium was used to attenuate neutrons with energies less than ~0.4 ev. The ratio of activity produced without the cadmium shield to the shielded irradiations was greater than 30, indicating that the activity can be produced with thermal neutrons. Furthermore, a decay curve for the Y^{90} ground-state activity was obtained using a $4\pi\beta$ flow proportional counter. These data are shown in Fig. 6. To eliminate the counter response to conversion electrons from the isomer, the source was placed between two 0.020-in.-thick absorbers. An analysis of the decay curve shows an initial buildup with a (3.05±0.11)-hr half-life followed by the decay of a single component with a (64.10±0.08)-hr half-life (Y^{90}). The total dead-time correction amounts to less than 5% of the observed effect. The observed buildup is attributed to the decay of the isomer to the ground state of Y^{90}. This, together with the fact that the activity can be produced by thermal neutrons, is felt to be sufficient proof that the isomer is in Y^{90}. The thermal cross section for the production of the isomer, measured by irradiating samples of yttrium together with thin gold foils to determine the thermal flux, was found to be 1.0±0.2 mb.

G. Internal Conversion Coefficient Measurements

Values of the total conversion coefficients of the 0.203- and 0.482-Mev gamma rays and the K-shell

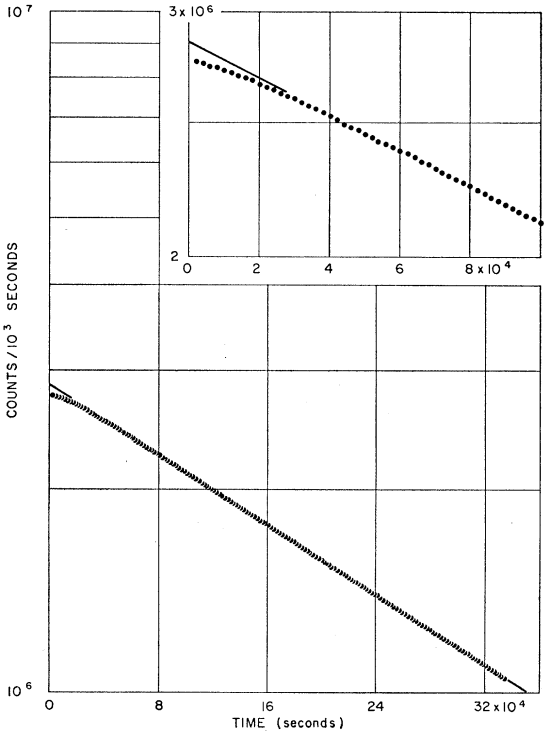

FIG. 6. Decay curve for Y^{90} ground-state beta activity.

coefficient of the 0.482-Mev gamma ray have been determined. The total conversion coefficients were obtained by measuring the absolute intensity of electrons and gamma rays with a $1\frac{1}{2}$-in. diam$\times\frac{1}{4}$-in. anthracene crystal and a 3-in.$\times$3-in. NaI gamma-ray detector. The efficiency of the electron detector was determined using Bi^{207m} and Ba^{137m} conversion electron sources, previously calibrated by absolute gamma-ray counting. A typical electron spectrum used in these measurements is shown in Fig. 4. Results obtained for α_T were: 0.11 ± 0.02 for the 0.482-Mev gamma ray and 0.03 ± 0.01 for the 0.203-Mev gamma ray. Comparison with the theoretical values of Rose[9] indicated that these values are consistent only with an $M4$ or $E5$ assignment for the 0.482-Mev transition and an $M1$ assignment with some $E2$ mixing for the 0.203-Mev transition.

Information on the K-shell coefficients for the two gamma rays was obtained from the relative intensities of the yttrium K x ray and the gamma rays in the singles spectra. The intensity of the yttrium K x ray, corrected for fluorescence yield,[10] represents internal conversion by both gamma rays. If one assumes an $M4$ assignment for the 0.482-Mev gamma ray and calculates the expected x-ray intensity from the theoretical value of α_k (0.084), a value of 0.030 is obtained for the K-shell conversion coefficient of the 0.203-Mev gamma ray. This compares with theoretical values of 0.026 and 0.068 for $M1$ and $E2$ radiation, respectively. An $E5$ assignment for the 0.482-Mev gamma ray would not materially alter this result, since the conversion coefficients for $M4$ or $E5$ radiation are nearly the same. The results of internal conversion coefficient measurements are summarized in Table II.

H. Directional Correlation Measurements

The directional correlation function of the 0.482–0.203-Mev gamma-ray cascade was measured with experimental techniques and equipment which have been described previously.[5] The "fast–slow" coincidence circuitry had a measured resolving time of $\sim8\times10^{-8}$ sec.

The random coincidence rate was measured several times during the course of the experiment and found to be less than 2% of the total coincidence rate. In the analysis of the data, the variation of this random coincidence rate with time (due to the decay of the source) was taken into account.

The measured directional correlation of the 0.482–0.203-Mev gamma-ray cascade is shown in Fig. 7. The solid curve is the least-squares fit of the data to a function of the form $W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$. The experimental values of the coefficients A_2 and A_4 are $-(0.153\pm0.006)$ and $+(0.0016\pm0.0063)$, respectively. After correction for the finite solid angle subtended by the detectors, these values are $-(0.178\pm0.007)$ and $+(0.003\pm0.011)$.

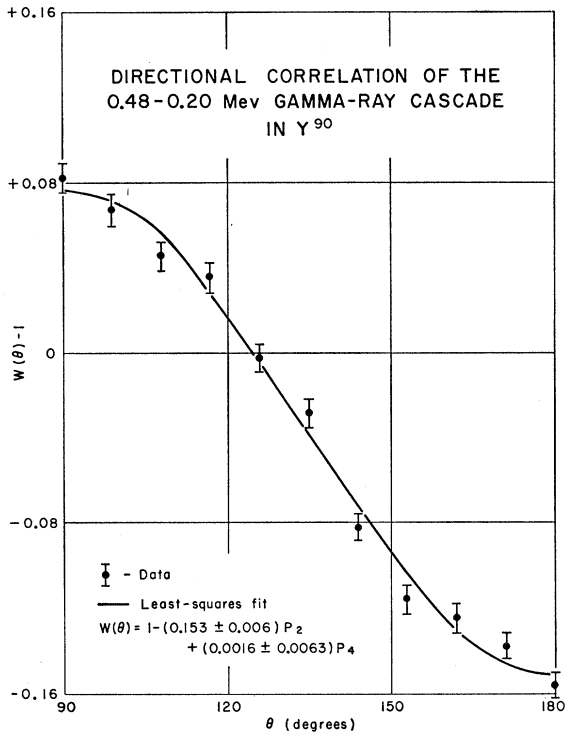

FIG. 7. Directional correlation of the 0.482–0.203 Mev gamma-ray cascade in Y^{90}.

TABLE II. Summary of conversion coefficient measurements.

E_γ (Mev)	α_T (experimental)	α_T (theoretical)			α_K (experimental) singles	α_K (theoretical)		
		$M1$		$E2$		$M1$		$E2$
0.203	0.03 ± 0.01	0.026		0.075	0.0298	0.023		0.068
		$E4$	$M4$	$E5$		$E4$	$M4$	$E5$
0.482	0.11 ± 0.02	0.038	0.100	0.112	0.084	0.032	0.084	0.090

Since the measured internal conversion coefficient of the 0.482-Mev gamma ray is consistent only with the assignment of either $M4$ or $E5$ to the transition, only spin sequences in which the multipolarity of this transition was 4 or 5 were considered. The negative value of A_2 definitely rules out an assignment of spin 4 to the 0.203-Mev state in Y^{90}. The only remaining assignment which is consistent with the half-life measurement and the absence of a crossover transition is 3. The data do not, however, provide unique assignments for the spin of the initial state or the mixing ratio of the second transition.

[9] M. E. Rose, *Internal Conversion Coefficients*, (North-Holland Publishing Company, Amsterdam, The Netherlands, 1958).

[10] C. D. Broyles, D. A. Thomas, and S. K. Haynes, Phys. Rev. **89**, 715 (1953).

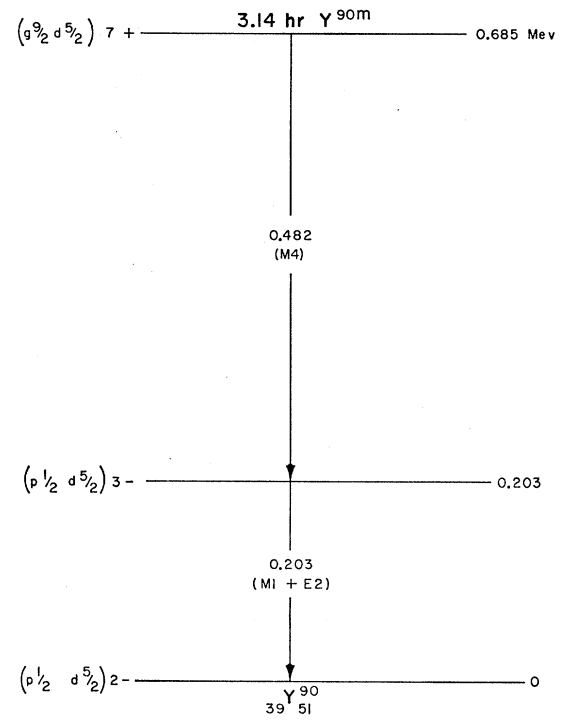

FIG. 8. Level scheme for Y^{90m}.

III. DISCUSSION

The decay scheme shown in Fig. 8 was constructed from the gamma-ray and beta-ray measurements previously described. The order of emission of the two gamma rays was established from the multipolarity assignments obtained from conversion coefficients and directional correlation measurements. Spin assignments for the 0.203- and 0.685-Mev levels are based on a knowledge of the ground-state spin[11] and the results of directional correlation measurements. Although neither the conversion coefficients nor directional correlation data unambiguously assigns a value for the spin of the initial state, the measured value for the half-life for gamma-ray decay of this level is more nearly in agreement with that expected for an $M4$ transition. This would require the assignment of spin 7 to this level.

It seems reasonable to expect that the Y^{90} nucleus can be described by the nuclear shell model.[11] The $g_{9/2}$ neutron shell is filled at 50 neutrons, leaving the odd neutron in Y^{90} in the $d_{5/2}$ level. Having 39 protons, the nucleus is left with one proton in the $p_{1/2}$ shell. On the basis of shape analysis of the beta spectrum of Y^{90}, the spin and parity of the ground state have been established as 2−. The next proton levels expected are $g_{9/2}$ and $g_{7/2}$. The order of neutron levels above the $d_{5/2}$ level would be $g_{7/2}$, $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$. Two possible configurations exist which could produce the 3− level at 0.203 Mev. The neutron could be elevated to the $g_{7/2}$ level to produce the configuration $(p_{1/2}g_{7/2})$, or the ground state configuration $(p_{1/2}d_{5/2})$ could recouple to give a spin of 3−. Similarly, three possible configurations exist for the isomeric level at 0.685 Mev. These configurations are $(g_{9/2}g_{7/2})$, $(g_{7/2}g_{7/2})$, and $(g_{9/2}d_{5/2})$.

Bartholomew *et al.*[12] have studied the level structure of Y^{90} by observing the capture gamma rays associated with the $Y^{89}(n,\gamma)Y^{90}$ reaction. The level scheme proposed in their work is shown in Fig. 9. Levels were reported at 0.2024, 0.247, and 0.7767 Mev. Shell-model configurations suggested for the 0.2024, 0.247, and 0.7767 Mev levels were $(p_{1/2}d_{5/2})$, $(p_{1/2}g_{7/2})$, and $(g_{9/2}d_{5/2})$, respectively, with assignments of 3− for the 0.2024 and 0.247-Mev levels and 2+ for the level at 0.7767 Mev. The assignment of 3− to both the 0.2024- and 0.247-Mev levels was based on the relative intensity arguments for gamma rays reportedly feeding these two levels from a level at 6.849 Mev and from the 0.7767-Mev level. These arguments depend upon the configuration assignment of the 0.7767-Mev level.

On the basis of the work of Bartholomew *et al.*,[12] the most reasonable configuration assignment for the level

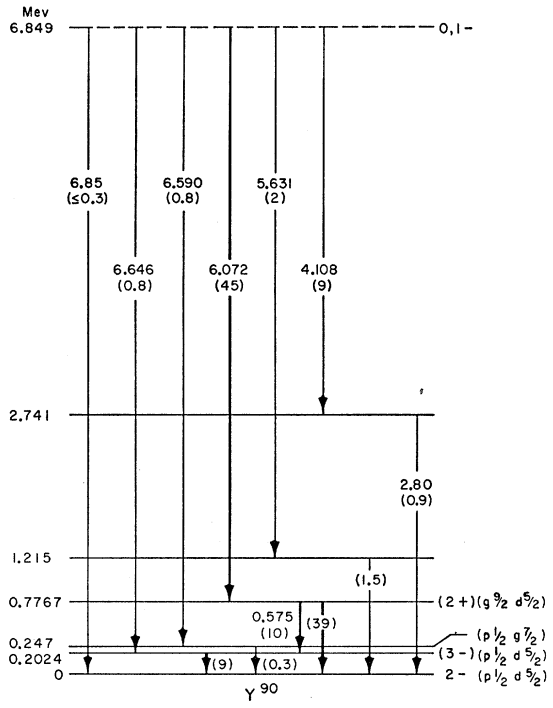

FIG. 9. Level scheme for Y^{90} proposed by Bartholomew *et al.*

[11] M. G. Mayer and J. H. D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley & Sons, Inc., New York, 1955).

[12] G. A. Bartholomew, P. J. Campion, J. W. Knowles, and G. Manning, Nuclear Phys. **10**, 590 (1959).

at 0.685 Mev would be $(g_{9/2}d_{5/2})$, since the isomeric transition would then involve a change in the state of only one nucleon. The transition to the 0.247-Mev state, involving a change in the state of both nucleons, should be considerably weaker than that to the state at 0.203 Mev. No 0.247-Mev gamma ray with an intensity greater than 1% of the 0.203 was observed in the present work.

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Nuclides Ar^{42} and Cl^{39}†

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The masses of Cl^{39} and Ar^{42} and the energies of their first excited states have been determined by an investigation of the reactions $Ar^{40}(t,\alpha)Cl^{39}$ and $Ar^{40}(t,p)Ar^{42}$. Charged reaction products were analyzed with a high-resolution magnetic spectrometer. The experimental results for Ar^{42} are: mass excess $(M-A)=-34.423\pm0.040$ Mev ($C^{12}=0$) or -21.990 ± 0.040 Mev ($O^{16}=0$); energy of first excited state $=1.138\pm0.030$ Mev. The experimental results for Cl^{39} are: mass excess $(M-A)=-29.772\pm0.040$ Mev ($C^{12}=0$) or -18.227 ± 0.040 Mev ($O^{16}=0$); energy of first excited state $=0.364\pm0.030$ Mev. The Q values of $Ar^{40}(t,p)Ar^{42}$ and $Ar^{40}(t,\alpha)Cl^{39}$ were found to be 7.046 ± 0.040 and 7.259 ± 0.040 Mev, respectively.

INTRODUCTION

THE masses and energy levels of a number of light nuclei have been measured at this laboratory in the past few years in experiments[1] using gas targets with tritons as the bombarding particles. The present experiment uses a triton beam to bombard an argon gas target in order to study the reactions $Ar^{40}(t,p)Ar^{42}$ and $Ar^{40}(t,\alpha)Cl^{39}$. A study of the proton and alpha energy spectra of these reactions should yield information on the masses of Ar^{42} (heretofore unknown) and Cl^{39} along with data on their energy levels. The information previously known about these nuclides mostly concerns the mass of Cl^{39} and the half-lives of the two nuclides.[2]

EXPERIMENTAL APPARATUS AND PROCEDURE

The experimental setup is identical with previous studies[1] and will not be described in detail. Briefly, a 2.6-Mev triton beam from an electrostatic accelerator bombarded a natural argon gas target. The reaction products emerging at a laboratory angle of 30° were analyzed with high resolution in a 16-in. double-focusing magnetic spectrometer and detected by a CsI scintillation counter. Mass-spectrometric analysis of the target gas showed that the argon concentration was always greater than 98 atom percent. Background runs were taken with nitrogen, oxygen, air, neon, and carbon dioxide. Energy determinations and other procedures are discussed in the previous papers.

RESULTS

Two alpha and two proton groups were observed that were assignable to the argon target. Energetic considerations eliminate all the argon isotopes except Ar^{40} as being responsible for these reactions. The peaks were about one-tenth the size of peaks seen[1] in reactions with targets of $Z=8$ or 10. This reduction in cross section is about what would be expected on the basis of Coulomb barrier penetration of the target nucleus by the incident triton. The peaks of interest were close to the ground-state alpha group from the $O^{16}(t,\alpha)N^{15}$ reaction and a small amount of oxygen was added to the target gas to provide an energy calibration. Analysis of the data has led to the results shown in Table I. No attempt was made to search for energy levels higher in excitation than the first excited states because of the increased complexity of background groups in the proton and alpha spectra. The estimates of errors are standard deviations. Values of known masses used in the calculations were taken from Everling *et al.*[3]

If one assumes that the even-even nuclide Ar^{42} has zero spin and even parity, then the beta decay to K^{42} (which has a 2^- ground state) is once forbidden. Only

† Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] N. Jarmie and M. G. Silbert, Phys. Rev. **120**, 914 (1960); M. G. Silbert and N. Jarmie, *ibid.* **123**, 221 (1961); M. G. Silbert, N. Jarmie, and D. B. Smith, Nuclear Phys. (to be published).
[2] P. M. Endt and C. M. Braams, Revs. Modern Phys. **29**, 683 (1957).
[3] F. Everling, L. A. König, J. H. E. Mattauch, and A. H. Wapstra, Nuclear Phys. **18**, 529 (1960).