Isomeric State of W^{181}†

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An isomeric state of W^{181} is excited by a (γ,n) reaction on W^{182} using the x-ray beam of the Iowa State College 75-Mev synchrotron. The energy and half-life of the isomeric state are found to be 366 ± 4 kev and 14.4 ± 0.3 microseconds, respectively. A measurement of the K-shell conversion coefficient yields a value of 0.30 ± 0.03. These experimental data suggest that the transition is of mixed multipole order, 28% $E1+72\%$ $M2$. This assignment is in agreement with the transition probabilities computed from Nilsson's wave functions for the first excited state and the ground state of the W^{181} nucleus.

I. INTRODUCTION

In recent years the shell model for nuclear forces which was proposed by Mayer[1] and others has enjoyed considerable success in predicting the observed islands of isomerism and the spins and parities of the single particle levels of most nuclei. However, there are some discrepancies between theory and experiment, notably for those nuclei which have large intrinsic quadrupole moments. Recently, calculations of the energy levels of nuclei as a function of the deformation of the nucleus have been made by Nilsson.[2] A comparison[3] between these calculations and the known ground state spins of the deformed nuclei has demonstrated the success of Nilsson's model. However, there is at present little experimental verification of the ability of this model to predict the spins and parities of the low-lying excited states. Since the existence of isomeric states in nuclei depends critically upon the spins and parities of neighboring states, it is evident that a search for isomers in the region of the deformed nuclei will give rise to experimental evidence which can be used as an additional test of Nilsson's model. Accordingly, a search for isomeric states in nuclei from $Z=57$ to $Z=74$, with half-lives greater than 5 microseconds, has been instituted. The x-ray beam of the Iowa State College 75-Mev synchrotron is used to excite the isomeric states.

The purpose of this paper is to describe the experimental method adopted for the search and to report on the isomeric state found in W^{181}. Other isomeric states which have been found[4] will be the subject of another paper.

An observation made by Vegors and Axel of an isomeric state arising from the x-ray bombardment of sodium tungstate has been previously reported.[5] However, no effort was made to identify the element or the isotope, or to measure quantitatively the conversion coefficient associated with the isomer.

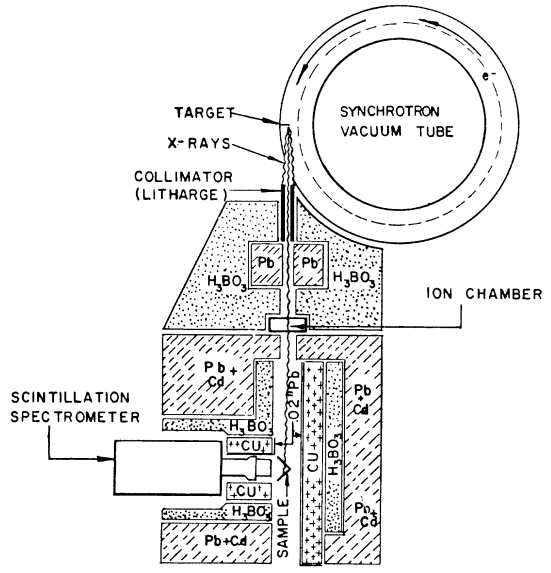

FIG. 1. Experimental arrangement. The Pb housing is interlaced with Cd sheeting.

II. EXPERIMENTAL METHOD

The Iowa State College synchrotron is a pulsed machine which can be made to deliver a 60-millimicrosecond burst[6] of x-ray radiation, fifty-two times per second. This accelerator is therefore ideal to use for the production of short-lived activities, because an accurate time base is achieved simultaneously with the limitation of the intense x-ray radiation from the synchrotron to a short interval of time. The geometry used for the bombardment of the various tungsten targets is shown in Fig. 1. The counter consists of a standard 1.5-inch diameter, 1-inch thick Harshaw NaI(Tl) crystal optically attached to the center of a three-inch DuMont 6363 photomultiplier tube. The photomultiplier tube is gated off during the entire

† Contribution No. 485. This work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

[1] Maria Mayer and D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley and Sons, Inc., New York, 1955).

[2] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 16 (1955).

[3] B. R. Mottelson and S. G. Nilsson, Phys. Rev. **99**, 1615 (1955).

[4] Stewart, Bureau, and Hammer, Bull. Am. Phys. Soc. Ser. II, **1**, 206 (1956).

[5] S. H. Vegors, Jr., and P. Axel, Phys. Rev. **101**, 1067 (1956).

[6] C. L. Hammer and A. J. Bureau, Rev. Sci. Instr. **26**, 594 and 598 (1955).

acceleration cycle of the synchrotron, including the x-ray burst, by pulsing the second dynode to the potential of the photosurface. This gating avoids any excessively large signals which result from scattered radiation during the acceleration cycle. However, it is found that the NaI crystal fluoresces for approximately 15 microseconds after the intense x-ray burst. It is therefore doubtful that this method could be used to measure isomeric transitions with half-lives of less than five microseconds.

The principal background encountered in this experiment is due to gamma rays which result from neutron capture by the NaI crystal and the surrounding materials. Since the neutrons have a definite moderation time, this background has an apparent half-life of approximately 400 microseconds. Moreover, the neutrons that become absorbed by the NaI crystal give rise to a spectral distribution centered about 135 kev. Figure 2 shows the pulse-height distribution of the background radiation which occurred within the first 200 microseconds after the x-ray beam burst, before adequate neutron shielding was used. Also in this figure

TABLE I. Half-life measurements.

Isotope	Half-life (microseconds) Our measurements	Previously reported values
Zn^{67}	9.4±0.3	8.5 9.5±1 8.8±1
Y^{88}	305±6	287±15

is shown the pulse-height distribution obtained by exposing the NaI crystal to thermal neutrons from a Po-Be source embedded in paraffin. This background is reduced to a reasonable level by the boric acid and cadmium shown in Fig. 1. (Also see Fig. 3.)

Three different pulse-height analyzers are used in this experiment. One, a 234-channel cathode ray oscilloscope pulse-height discriminator,[7] is used to determine the energy of the gamma rays. In addition, because this analyzer employs a cathode ray oscilloscope as its basic component, it is possible to convert the 234 pulse-height channels into 234 time channels by merely starting a calibrated sweep at the time the x-ray burst occurs. It is possible to measure half-lives in a range from one microsecond to ten milliseconds with this instrument. When this analyzer is used to measure half-lives, a single-channel pulse-height discriminator is used to preselect the energy of the gamma ray under investigation.

In order to test the validity of this method of measuring half-lives, the metastable states in Zn^{67} and Y^{88} were excited by the x-ray beam. The results of these

[7] Hunt, Rhinehart, Weber, and Zaffarano, Rev. Sci. Instr. **25**, 268 (1954).

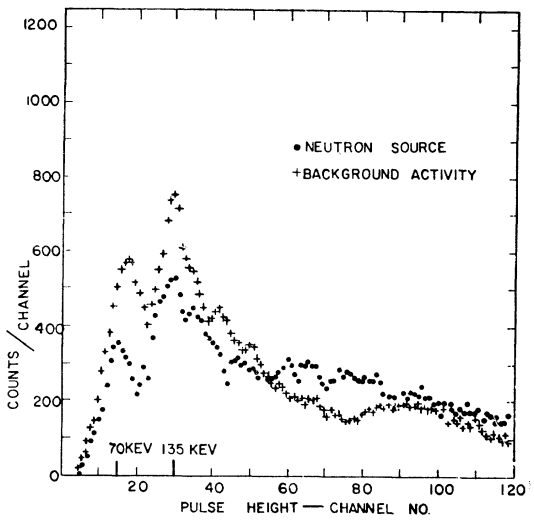

FIG. 2. Background radiation spectrum.

half-life measurements and previously reported values[5,8] are summarized in Table I.

The third pulse-height analyzer is a conventional ten-channel pulse-height discriminator which is used to follow the yield of the gamma rays as a function of the peak energy of the synchrotron. All three pulse-height selectors are electronically gated to accept pulses only during a time interval corresponding to several half-lives of the activity being measured. This second gating system reduces the effects of any long-lived activity induced in the target or surrounding materials by the x-ray beam.

III. RESULTS

A. Energy and Half-Life Measurement

The spectrum of the activity obtained by bombarding a five-mil tungsten target is shown in Fig. 3. The neutron-induced activity is kept essentially constant during a background run by using a 5-mil Au target in place of the W target. The broad peak shown at channel 60 is due to backscattering of the 366-kev gamma ray from the walls of the lead housing, while the small peak shown at channel 13 is the escape peak associated with the tungsten x-ray. The pulse-height analyzer is calibrated by using the known 425-kev and 174-kev γ rays and the 26.2-kev x-ray that arise from the decay of Sb^{125},[8] and the 357-kev, 300-kev and 82-kev γ rays that arise from the decay of Ba^{133}.[9] The calibration curve is shown in Fig. 4. The gamma-ray energy was thus determined to be 366±4 kev. An exhaustive search for additional gamma-ray activity from the tungsten target with half-lives between 5 microseconds and 1 hour, and energies from 20 kev to 2 Mev, yielded negative results. It is therefore necessary to conclude

[8] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[9] Hayward, Hoppers, and Ernst, Phys. Rev. **93**, 916(A) (1954).

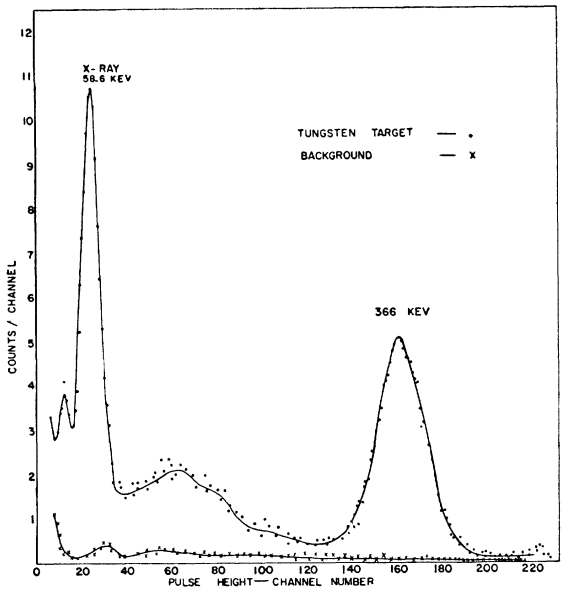

FIG. 3. W^{181} spectrum.

that either the 366-kev state is metastable or that there exists a metastable state which decays to the 366-kev state by internal conversion in the K shell. However, if this latter case were true, the ratio of the intensities of the tungsten x-ray to the 366-kev γ ray would have to be ≥ 1. A measurement of this ratio, which is described in part B of this section, gives a value of 0.3, thereby determining that the 366-kev gamma ray arises from an isomeric transition.

At least four half-life determinations (extended over five half-lives) were made using both the tungsten x-ray and the 366-kev γ ray. All of these measurements agree within five percent. The result was 14.4±0.3 microseconds.

The measurements of the energy of the gamma ray and the half-life of the transition are in good agreement with the recent values of 368±5 kev and 14.7±0.5 microseconds obtained by Vegors and Axel.[10]

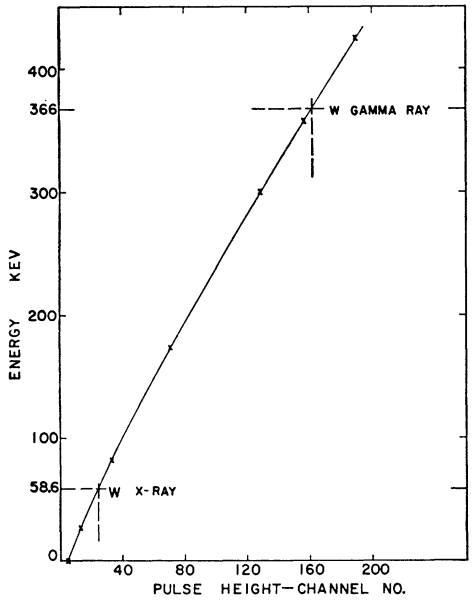

FIG. 4. Energy calibration curve.

B. Conversion Coefficient

Since the K x-rays coming from the target represent the decay of the isomeric state via internal conversion, it is possible to obtain a direct measure of the conversion coefficient by comparing the intensity of the x-rays to the intensity of the γ rays. However, this is a difficult measurement because of absorption by the target and Compton scattering in the target and surrounding materials. It is possible to measure the effects of the target on the observed intensity ratio by increasing the target thickness. If, for example, the target thickness is increased and no effect is observed on the ratio,[11] one would conclude that the effects of target thickness are negligible. Accordingly, both five- and ten-mil tungsten targets were bombarded and the ratio of the intensities was obtained. Three separate measurements were made and in no case did the ratio differ by more than five percent. Since the 5-mil target is used for all subsequent measurements, no correction is made for the target effects.

TABLE II. Intensity ratio measurements.

Intensity ratio for 5-mil tungsten	Intensity ratio for Ba	Conversion coefficient $(\alpha_K)_W$
1.08±0.02	0.96±0.01	0.31
1.03±0.02	0.94±0.02	0.30

The effects of the crystal efficiency and Compton scattering on the intensity ratio can be obtained in a single measurement if a source whose decay scheme is well known is substituted for the tungsten target and the intensities of the various gamma rays are measured. A Ba^{133} source is ideal for this purpose since it emits a 300-kev γ ray and a 357-kev γ ray, each in coincidence with an 82-kev γ ray.[9] The higher energy γ rays compare favorably with the energy of the 366-kev γ ray from tungsten while the 82-kev γ ray compares favorably with the energy of the tungsten x-ray. Unfortunately, the 82-kev γ ray is converted and the conversion coefficient is not well known. The reported value[9] of 3.5 seems too high to agree with any pure multipole order or any reasonable admixture. A measurement of the conversion coefficient, using the method of in-

[10] S. H. Vegors, Jr., and P. Axel (private communication).

[11] This would also be the case if the targets were opaque to both the x-ray and the γ ray. However, this does not apply here since the total absorption coefficient for 366-kev γ rays passing through W is only 3.5 cm^{-1}. Thus, only 10% of these γ rays are absorbed by 10 mils of W.

tensities employed by Hayward, Hoppers, and Ernst,[9] yields a value of 2.6. This value compares favorably with the values (including the effect of screening) quoted in Rose's tables[12] for an $M1+E2$ transition. A Ba^{133} point source was used for this measurement and care was taken to remove all materials from the vicinity of the source and counter in order to avoid back scattering of the γ rays. Bell's curves for crystal efficiency and peak-to-total ratio[13] were used to obtain an estimate of the crystal efficiency. In an attempt to obtain some experimental verification for these curves, the conversion coefficient was computed by using both the number of γ rays under the photoelectric peaks and the total number of γ rays. Both calculations agreed within the range of statistical error. The measured value of 2.6 is used in the measurement of the conversion coefficient for the 366-kev transition in tungsten.

To measure the effects of Compton scattering on the conversion coefficient for the 366-kev transition in tungsten, a Ba^{133} source was deposited in the shape of the x-ray beam at the target position on a thin aluminum backing. The source was then placed in the target holder and the ratio of the intensity of the 82-kev

TABLE III. Conversion coefficients.

$E1$	$M1$	$E2$	$M2$	$E3$
1.3×10^{-2}	0.14	3.5×10^{-2}	0.41	0.10

γ ray to the total intensity of the 300-kev and 357-kev γ rays was measured. The conversion coefficient $(\alpha_K)_W$ for the 366-kev transition in tungsten is then given by the relationship,

$$(\alpha_K)_W=\frac{R_W}{R_{Ba}[1+(\alpha)_{Ba}]}, \quad (1)$$

where R_W = intensity ratio for tungsten x-ray to γ ray, corrected for fluorescence yield, and R_{Ba} = intensity ratio for Ba^{133} γ rays as described above. Table II summarizes the results of these measurements.

It is estimated that a reasonable error for $(\alpha_K)_W$ is approximately 10%. Table III lists the values of the conversion coefficients taken from Rose's tables[14] for various multipole orders interpolated for $Z=74$ and $E=366$ kev.

The energy and half-life of the tungsten isomer are consistent with a transition which is predominantly $M2$.[15] The observed conversion coefficient (see Table III) can therefore be explained most reasonably by a 28% $E1$+72% $M2$ admixture, although an admixture consisting of 65% $M2$+35% $E3$ cannot be ruled out on the basis of this experiment. However, a "fast" $E3$ component has never been experimentally observed, whereas an inhibited $E1$ component is commonly observed for nuclei with large deformations.[16,17]

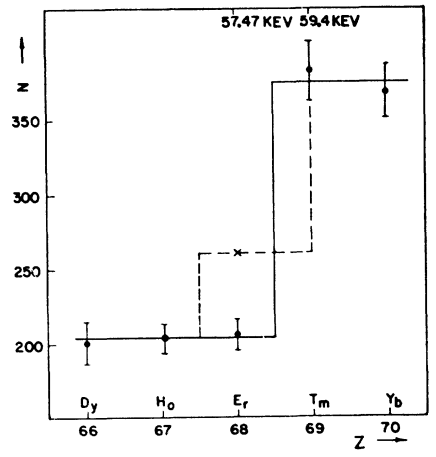

FIG. 5. Critical absorption of tungsten x-ray. The energy corresponding to the K absorption edge for Er and Tm is shown at the top of the graph.

C. Identification of the Isotope

Since the x-ray bombarding energy is above the $(\gamma,2n)$ and (γ,p) threshold for tungsten, it is necessary

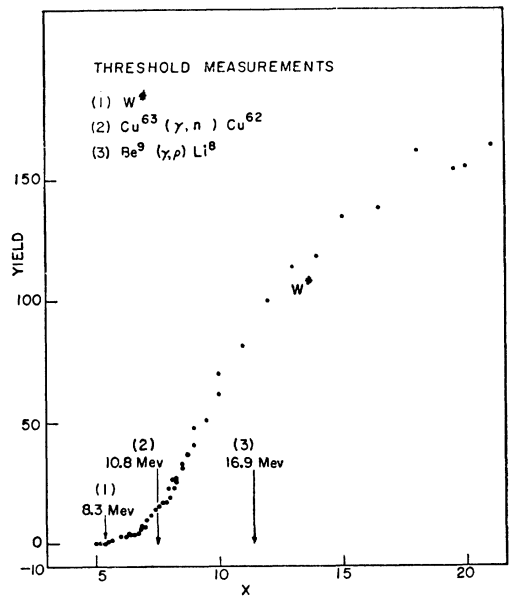

FIG. 6. Activation curve for the tungsten gamma ray. The peak energy of the x-ray beam in Mev is given by $E=1.53X-0.51$.

[12] M. E. Rose and G. H. Goertzel, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Appendix 4.

[13] P. R. Bell, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 5.

[14] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023 (unpublished).

[15] M. Goldhaber and A. W. Sunyar, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 16, Sec. II.

[16] J. O. Rasmussen and D. Strominger, Bull. Am. Phys. Soc. Ser. II, **1**, 206 (1956).

[17] D. M. Chase and L. Wilets, Phys. Rev. **101**, 1038 (1956).

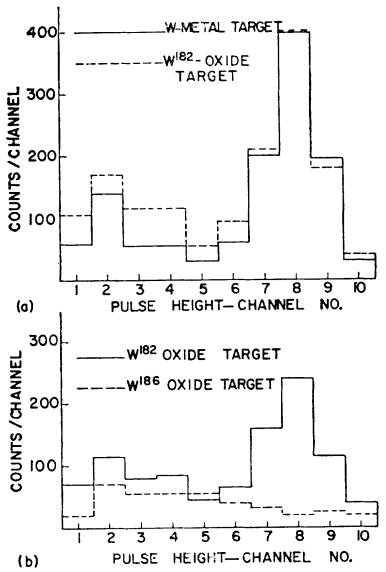

FIG. 7. W spectrum (a), A comparison of the energy spectrum obtained from a 5-mil metal W target and a W^{182} oxide target. (b), A comparison of the energy spectrum obtained from a W^{182} oxide target and a W^{186} oxide target.

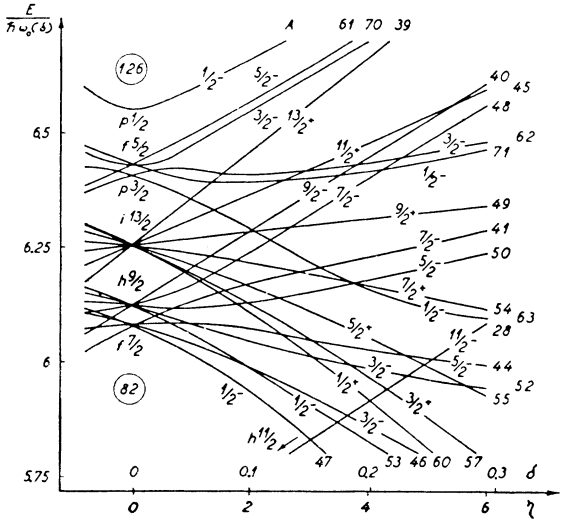

FIG. 8. Spectra for nuclei with an odd number of neutrons from $N=82$ to $N=126$ as a function of the nuclear deformation δ.

to perform separate measurements to establish which element and isotope contains the isomeric state. Figure 5 shows the results obtained from a critical absorption experiment on the x-ray activity observed in the tungsten target. The abscissa represents the various absorbers used while the ordinate is proportional to the number of transmitted x-rays. The dashed line is the expected absorption curve for the Ta x-ray while the solid line is the expected curve for the W x-ray.

Figure 6 shows the yield of the 366-kev gamma ray as a function of the peak energy of the x-ray beam. The standards used for this measurement are the known thresholds for the $Cu^{63}(\gamma,n)Cu^{62}$ and $Be^{9}(\gamma,p)Li^{8}$ reactions.[18]

The results of the measurements shown in Figs. 5 and 6 clearly indicate a (γ,n) reaction. The known energy level schemes for W^{182}, W^{183}, and W^{184},[19] coupled with the fact that the isomeric state arises from a (γ,n) reaction in a tungsten isotope, eliminates all isotopes but W^{181} and W^{185}. Figure 7 shows the spectrum obtained on the ten channel pulse height analyzer by bombarding separated isotopes of W^{182} and W^{186}. For comparison purposes, the spectrum obtained for the 5-mil tungsten target is also shown in Fig. 7. The results shown in Figs. 5, 6, and 7 clearly establish the fact that the isomeric state is in W^{181}.

IV. COMPARISON TO THEORY

The energy levels which have been calculated by Nilsson[2,3] for nuclei with an odd number of neutrons are shown in Fig. 8. The predicted ground state spin of W^{181}, assuming a deformation 0.2, is the $i_{13/2}$ $(\Omega=9/2)$ $(+)$ (No. 49) level shown in the figure. This spin assignment is also consistent with the decay scheme of W^{181}.[8] The only choice of a first excited single particle state which agrees both with the results of this experiment and the theoretical predictions is the $f_{7/2}$ $(\Omega=7/2)$ $(-)$ (No. 41) level shown in Fig. 8. This would mean that the excited nucleus prefers to have two neutrons in the $\Omega=9/2$ $(+)$ state, leaving behind a hole in the $\Omega=7/2$ $(-)$ state. With this interpretation, the inhibited $E1$ transition observed in this experiment can be explained as follows. At small deformations, the isomeric transition in question is between a pure $i_{13/2}$ state and a mixture of the $h_{9/2}$ and the $f_{7/2}$ nucleonic states. Hence, $|\Delta j|\geq 2$ and the $E1$ transition is forbidden. In the limit of large deformations, the components of the orbital angular momentum Λ, and spin Σ, along the axis of symmetry of the nucleus, are individually good quantum numbers. In this limit, the initial and final states for the isomeric transition have opposite spin components Σ, and the $E1$ transition is again forbidden. Even for intermediate deformations, a calculation shows that the theoretical transition rate remains quite small. In particular, for the deformation $\delta=0.2$, the $E1$ rate for the 7/2 $(-)$ to the 9/2 $(+)$ transition is inhibited relative to the corresponding spherical independent-particle model result computed by Moszkowski[20] by a factor of 10^{-4}. However, small changes in the coefficients of Nilsson's wave functions could easily change this result by a factor of $10^{\pm 2}$. For example, a 5% change of the coefficient of one of the wave functions causes the $E1$ transition probability to vanish.

[18] D. M. Van Patter and W. Whaling, Revs. Modern Phys. **26**, 408 (1954).

[19] Murray, Boehm, Marmier, and DuMond, Phys. Rev. **97**, 1007 (1955).

[20] S. A. Moszkowski, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 13.

An explanation must also be given for the fact that no rotational level is found in the observed spectrum. With the above interpretation, the rotational level would be expected to have a spin of 11/2 (+). The transition to this state from the 7/2 (−) state would, therefore, also be predominantly of multipole order $M2$. The ratio, R_T, of the transition probabilities for the emission of a given multipole radiation from a state, i, to two different members, f and f', of a rotational family is given by[21]

$$R_T=\left[\frac{E}{E'}\right]^{2L+1}\frac{\langle I_i, L, K_i, K_f-K_i|I_i, L, I_f, K_f\rangle^2}{\langle I_i, L, K_i, K_f-K_i|I_i, L, I_{f'}, K_f\rangle^2}, \quad (2)$$

where E=energy of the emitted gamma ray, I=total angular momentum, L=multipole order, K=the component of I along the symmetry axis of the nucleus, and the bracketed quantities are the Clebsch-Gordan coefficients for the addition of angular momenta. For $L=2$ and for a rotational state of energy greater than 120 kev, the ratio of the transition probabilities is less than eight percent. It is improbable that such a weak transition could be observed in the presence of the background produced by the Compton scattering of the 366-kev γ rays.

[21] Alaga, Alder, Bohr, and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 9 (1955).

V. ACKNOWLEDGMENTS

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Decay of 2.7-Hour Sr^{92}†

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The decay of 2.7-hr Sr^{92}, produced as a fission product activity, has been studied using the scintillation method. Gamma rays of energy 0.23±0.02, 0.44±0.04, and 1.37±0.05 Mev were found, with relative intensities of 0.039±0.004, 0.045±0.004 and 1.00, respectively. Beta-ray spectrometry using anthracene and beta-gamma coincidence studies indicate a major beta of 0.545±0.05 Mev in coincidence with the 1.37-Mev gamma ray. Comparison with 4π beta measurements indicate a branching ratio of 90±10% leading to the 1.37-Mev excited state ($\log ft=4.3$).

I. INTRODUCTION

THE 2.7-hr fission product Sr^{92} activity has been reported by Götte,[1] who determined its half-life by observing the production of the 3.5-hr Y^{92} daughter activity from uranium fission. Since the time of these early measurements, no further information on the decay characteristics of this nuclide has appeared in the literature. Because of this lack of detailed information a study was undertaken to determine the decay scheme, using the scintillation method.

II. EXPERIMENT AND RESULTS

Source Preparation

Samples of short-lived strontium fission-product activity were prepared by making short-time irradiations of uranyl nitrate in a high-flux facility of the MTR. The strontium chemical fraction was then obtained and samples mounted for observation. Since 9.7-hr Sr^{91} was also present in the samples and the decay of the two strontium activities gives rise to 55-min Y^{91} and 3.5-hr Y^{92}, activity was observed with a minimum time delay following the chemical separation.

Gamma-Ray Measurements

The scintillation gamma detectors used were cylinders of NaI(Tl), 3 inches in diameter and 3 inches long, mounted on DuMont type 6363 photomultipliers mounted in the manner described by Lazar and Klema[2] to produce the conditions for good gamma ray spectroscopy. Sources were mounted 3 cm from the surface of the detector on the central axis and a 1.6-g/cm² polystyrene absorber used to remove the beta rays. The detector assembly was housed in a "graded" lead shield with inside dimensions of 12×12×24 inches. Pulse analysis was accomplished by using a 20-channel pulse sorter of the type designed by Bell, Kelley, and Goss,[3] operated in conjunction with a 100-channel automatic readout system designed at this laboratory.

Figure 1 shows the pulse spectrum of the gamma

† Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] H. Götte, Naturwiss. **28**, 496 (1941).

[2] N. H. Lazar and E. D. Klema, Phys. Rev. **98**, 710 (1955).
[3] Bell, Kelley, and Goss, Oak Ridge National Laboratory Report ORNL-1278, 1951 (unpublished).