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Reaction $Zr^{90}(n,n')Zr^{90m}$†

ROBERT T. WAGNER, E. R. SHUNK, AND ROBERT B. DAY
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico
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The cross section for the excitation of the 2.3-MeV metastable state in the $Zr^{90}(n,n')Zr^{90m}$ reaction has been measured from threshold to 5 MeV. An irradiate-count cycle was used in which a Zr sample was first bombarded by monoenergetic neutrons and then transferred by a pneumatic transport system to a shielded counting station where the 2.3-MeV gamma rays from the decay of the metastable state were measured. The half-life of this state was found to be $T_{1/2}=0.801\pm0.005$ sec. The excitation curve has a threshold at 2.3 MeV and increases monotonically to 300 mb at 5 MeV. In addition to the threshold rise there are also marked increases in cross section that begin at 2.75 and 4.4 MeV. These increases are due to states at these energies that cascade through the 2.3-MeV state. The state at 2.75 MeV is probably the hitherto missing 4− (four-minus) state of the $(p_{1/2}g_{9/2})$ configuration and it is essentially equal in energy to the 3− (three-minus) state at 2.75 MeV that has been observed in other reactions.

I. INTRODUCTION

BECAUSE of its relatively simple structure, the study of the nucleus Zr^{90} is instructive. Ford[1] was the first to point out that it is very similar to a doubly magic nucleus since the subshell at 40 protons behaves much like a closed shell. On this basis he predicted that the low-energy levels of Zr^{90} should consist of almost pure configurations of the form $(p_{1/2})^2$, $(p_{1/2}g_{9/2})$, and $(g_{9/2})^2$. Subsequent experimental work bore out his predictions, and now all of the possible levels represented by these configurations have been identified with the exception of the 4− level from the $(p_{1/2}g_{9/2})$ configuration. Furthermore, Bayman[2] has done more detailed calculations and has been able to obtain fairly good agreement betweeen the calculated and experimentally observed positions of these levels.

In view of the success of the theory in predicting the positions of the other low-lying levels in Zr^{90}, it is interesting that the 4− level has not been experimentally verified. The most extensive experiments on Zr^{90} have been on the radiations following the β decay of Nb^{90}, but because of the spins of the states involved it is not surprising that the 4− state was not found. However, experiments on neutron inelastic scattering by Zr^{90} have been carried out at neutron energies up to 3.7 MeV. Since neutron scattering at these energies appears to excite all possible levels without regard to their specific character, the 4− state should have been found unless it is nearly equal in energy to one of the other states.

One of the characteristics of the 4− level that is expected from elementary theoretical considerations is that it should decay to the 5− state of the $(p_{1/2}g_{9/2})$ configuration by means of an $M1$ transition. Thus, if one measured the cross section for excitation of the 5− level by neutron inelastic scattering as a function of energy, the position of the 4− level should be revealed by the usual threshold rise that is observed when another level emits a gamma ray that cascades to the level under observation. Since the 5− level is metastable, with a half-life of 0.8 sec[3], the technique described below permits a greatly improved signal-background ratio over the techniques used in the earlier neutron inelastic scattering measurements. With this improvement one can measure the shape of the excitation curve of the 5− state well enough so that this indirect method is probably more sensitive than a direct measurement of the 4− level.

Campbell and Stelson[3–5] have previously studied the excitation curve for the 5− level. Their results show a rise at 2.75 MeV in addition to the threshold of the 5− state itself at 2.3 MeV. The level at 2.75 MeV is believed to be a 3− level because (1) its energy is in agreement with the systematics of such levels, and (2) this level is found in the same type of reaction in which the 3− states are found in other even-even nuclei.[6,7] Furthermore, it shows a strong transition to the 2+ state at 2.18 MeV in addition to a transition to the 5− state. This indicates that if there is a single state here, it cannot be the missing 4− state.

A rough excitation curve for the 5− state has also been measured previously by Lind and Day.[8] The shape of their curve is probably wrong because of the difficulties they experienced in analyzing their data due to the presence of the strong 2.18-MeV gamma ray. However, there was also a fairly large discrepancy in cross section between these two sets of measurements in the region where the results of Lind and Day were not so uncertain. Since Zr^{90} provides a useful example for testing the statistical theory of nuclear reactions, it was felt that

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

[1] K. W. Ford, Phys. Rev. **98**, 1516 (1955); see also I. Talmi and I. Unna, Nucl. Phys. **19**, 225 (1960).

[2] B. F. Bayman, A. S. Reiner, and R. K. Sheline, Phys. Rev. **115**, 1627 (1959).

[3] E. C. Campbell, R. W. Peelle, F. C. Maienschein, and P. H. Stelson, Phys. Rev. **98**, 1172 (1955).

[4] *Neutron Cross Sections*, complied by D. J. Hughes and R. B. Schwartz (U. S. Government Printing Office, Washington, D. C., 1958), BNL-325, 2nd ed.

[5] P. H. Stelson (private communication).

[6] B. L. Cohen and A. G. Rubin, Phys. Rev. **111**, 1568 (1958); B. L. Cohen, *ibid.* **125**, 1358 (1962).

[7] M. Crut, D. R. Sweetman, and N. S. Wall, Nucl. Phys. **17**, 655 (1960).

[8] D. A. Lind and R. B. Day, Ann. Phys. (N. Y.) **12**, 485 (1961).

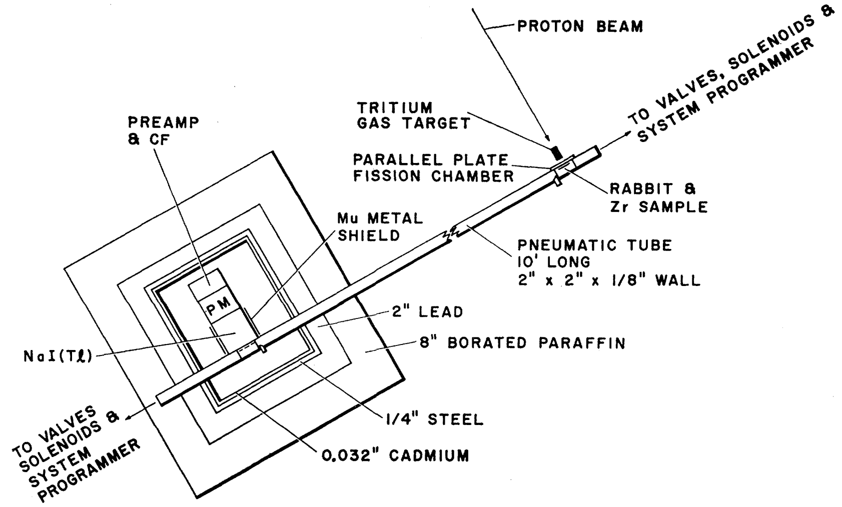

FIG. 1. Schematic diagram of experimental arrangement.

this discrepancy should be cleared up in order that meaningful comparisons of experiment with the theory might be made.

II. EXPERIMENTAL PROCEDURE

The 0.8-sec half-life of the 2.3-MeV state of Zr^{90} made possible a technique that gave a much better signal-to-background ratio than would have been possible for states emitting prompt gamma rays. In this method the sample was alternately bombarded by neutrons and the activity then counted, with the beam being turned off during the counting period. In this way the background produced by the beam, neutrons, and prompt gamma rays was eliminated. In addition, after bombardment the Zr sample was transported by a pneumatically operated system to a shielded area where the counting could take place with considerably lower background than would otherwise have been possible. This irradiate-count cycle was repeated with the help of an electronic programmer until sufficiently good satistical accuracy had been obtained.

Monoenergetic neutrons from 2 to 5 MeV were obtained from an electrostatic accelerator using the $T(p,n)He^3$ reaction. The neutron flux through the Zr sample was monitored by using a specially calibrated U^{238} fission counter positioned immediately in front of the sample. Some of the various parts of the experiment are described in the following sections.

(a) Electromechanical System

Figure 1 shows a sketch of the experimental arrangement. The pneumatic transport system was fairly standard, and it was equipped with several microswitches to announce the arrival of the rabbit at the irradiation position and the counting position. In this manner it was possible to turn off the beam during the counting interval as well as to turn off the photomultiplier voltage when the beam was on.

The irradiation-counting cycle is shown in Fig. 2. The cycle was automatically repeated until at least 10 000 counts had been accumulated in both the neutron detector and the gamma-ray detector. The data from these detectors were fed into an RIDL Model 34–12 400-channel analyzer that was operated in the time analyzer mode. Each channel width was 0.1 sec. In channels 160–200 were stored neutron monitor counts as a function of time so that one could correct for possible fluctuations in the neutron flux during sample irradiation. Figure 3 shows a block diagram which illustrates the electronic apparatus used to obtain the data.

(b) Scintillation Detector

A Harshaw 3-in.×3-in. NaI(Tl) crystal mounted on a DuMont K1197 photomultiplier was used to detect the gamma rays from the decay of the 2.3-MeV metastable state. The efficiency of the detector was measured using a calibrated Na^{24} source whose activity was determined by the method of beta-gamma coincidence counting. The source was spread over an area as large as the sample (1-in. diam). It was placed in the rabbit in the position normally occupied by the Zr sample; the rabbit was then transferred inside the pneumatic tube to the counting position. In this manner the scattering and attenuation environment seen by the Zr sample

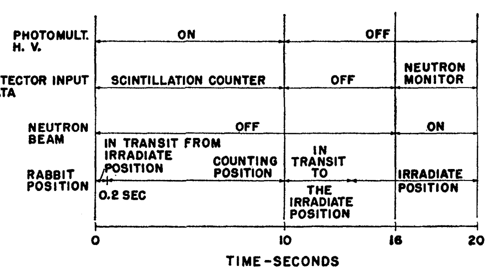

FIG. 2. Time sequence for single cycle.

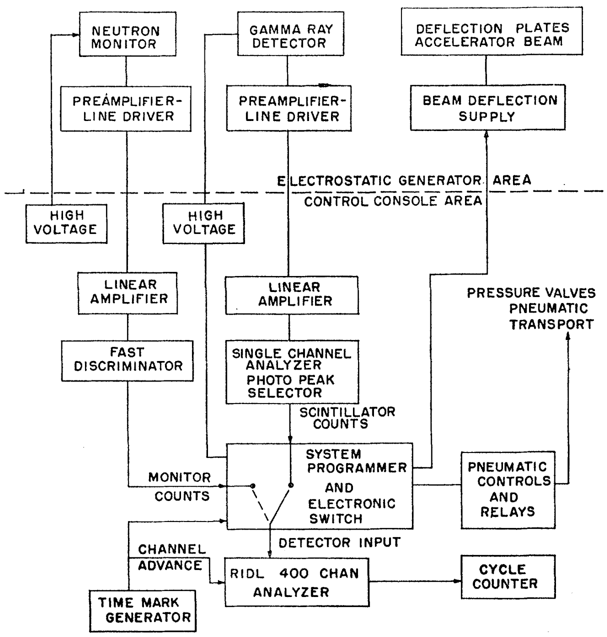

FIG. 3. Block diagram of electronic apparatus.

during the experiment was duplicated during the course of the detector calibration. Simultaneous detection of the 1.37- and 2.75-MeV gamma rays from the source had the effect of reducing the apparent photopeak efficiency as well as introducing a background. The background was estimated and subtracted. A correction for photopeak efficiency reduction was obtained by calculating the probability that a second gamma ray would interact with the crystal in any way. This correction amounted to 15% for the 2.75-MeV gamma ray and 12% for the 1.37-MeV gamma ray. The efficiency for the 2.3-MeV gamma ray was interpolated from the plot of photopeak efficiency vs gamma-ray energy. Actually a gamma ray of somewhat less than 2.3 MeV was used since 13%[9,10] of the time the 2.3-MeV state decays to a state of 2.218 MeV, which then emits a gamma ray of that energy. Finally, small corrections were made for the fact that the single-channel analyzer included a pulse-height region somewhat wider than the photopeak, and for the self-absorption in the Zr sample.

(c) Neutron Source and Monitor

The neutrons were produced by bombarding a target 3 cm long filled with tritium at a pressure of 3 atm. A molybdenum foil 10 mg/cm^2 thick served as the entrance window to the target. During the irradiation period the Zr sample was located at 0° to the beam 1 in. from the end of the target with its axis collinear with the beam. Effects due to straggling in the molybdenum foil, energy loss in the tritium, and variation of neutron energy with angle cause a spread in the effective neutron spectrum of approximately ± 90 keV at all energies. Deviations from this spread were only of the order of 5 keV or less. In the results that follow, the neutron energy quoted is the average neutron energy.

The neutron flux was measured with a parallel plate fission ionization chamber that was specially designed for this purpose. It contained a fission foil consisting of 2.955 mg of U^{238} deposited on a 0.005-in.-thick platinum foil 1 in. in diameter. The chamber was constructed with thin walls in order to minimize scattering and absorption of neutrons. It was placed on the beam axis immediately in front of the pneumatic tube containing the Zr sample. Since the diameters of the fission foil and the Zr sample were both 1 in., the fission chamber subtended essentially the same number of neutrons as the Zr sample. A small correction was applied to the data to correct for the difference in solid angle subtended by the fission foil and the sample. The fact that the neutron source was distributed along the length of the gas target was taken into account in calculating this correction.

From a knowledge of the neutron-induced fission cross section of U^{238} as a function of energy the neutron flux through the fission foil per monitor count could be calculated. Several corrections had to be made, however, for the following effects:

(1) Stopping of fission fragments in the foil (3%).
(2) Deviation from isotropy of fission fragments due to momentum imparted by bombarding neutron ($\leqslant 1\%$).
(3) Low-energy part of pulse-height distribution that cannot be counted because it is in the amplifier noise (4%).
(4) Absorption of neutrons by material between the fission foil and the Zr sample (1.5–2.0%).

The first two corrections were calculated for the foil used here. The third was obtained by measuring the pulse-height distribution for the chamber and extrapolating this to zero pulse height from the region above the amplifier noise. An auxiliary experiment was done to determine the actual absorption introduced by the

TABLE I. Cross section for excitation of 2.3-MeV state.[a]

E_n (MeV)	σ (mb)	E_n (MeV)	σ (mb)	E_n (MeV)	σ (mb)
2.3	1.9	3.2	115.9	4.1	184.9
2.4	12.4	3.3	124.6	4.2	188.4
2.5	23.3	3.4	125.3	4.3	197.1
2.6	30.3	3.5	137.9	4.4	196.3
2.7	41.9	3.6	137.1	4.5	219.1
2.8	53.4	3.7	149.8	4.6	238.5
2.9	77.3	3.8	153.5	4.7	261.0
3.0	101.6	3.9	167.1	4.8	279.0
3.1	115.9	4.0	170.1	4.9	299.2
				5.0	308.3

[a] The cross sections are estimated accurate in absolute value to within 10%.

[9] S. Bjørnholm, O. B. Nelson, and R. K. Sheline, Phys. Rev. **115**, 1613 (1959).
[10] N. H. Lazar, G. D. O'Kelley, J. H. Hamilton, L. M. Langer, and W. G. Smith, Phys. Rev. **110**, 513 (1958).

walls of the fission chamber and pneumatic tube plus the polyethylene rabbit. The correction for this varied between 1.5 and 2% depending on the energy used. The over-all accuracy in the neutron flux measurements was 5%.

III. DATA ANALYSIS

At the end of an experimental run at a given neutron energy, the data stored in the RIDL analyzer were punched onto paper tape and later transferred to punched cards for analysis by an IBM 7090 computer. The computer first corrected the data for the 4.4-μsec dead time in the electronics system. It next fitted the data from the gamma-ray detector to a function of the form

$$f(t)=Be^{-\lambda t}+C.$$

Here B is the counting rate from the Zr sample extrapolated to time zero, and C is the background counting rate. The computer then calculated the cross section for excitation of the 2.3-MeV level from the formula

$$\sigma=\frac{AB}{N_0M\epsilon K[1-\exp(-\lambda\Delta t)]}\times\{\textstyle\sum_i N_i\exp[-\lambda\Delta t(n-i)]\}^{-1},$$

where σ = cross section (cm^2), A = atomic mass number of sample, N_0 = Avogadro's number, M = mass of Zr^{90} in sample (grams), ϵ = efficiency of gamma-ray detector, K = calibration factor of neutron monitor (neutrons/count), λ = decay constant (sec^{-1}), Δt = time interval in which data are stored (0.1 sec), N_i = number of monitor counts in ith interval, and n = total number of intervals (40). In this formula the quantities ϵ and K contain all the corrections discussed in the previous section that are necessary to convert B and N_i to the total disintegration rate and neutron flux at the sample, respectively.

IV. RESULTS AND DISCUSSION

From the least-squares fit to the gamma-ray decay of the 2.3-MeV state a half-life of 0.801 ± 0.005 sec was obtained. This is in agreement with the value of 0.83 sec previously obtained by Campbell *et al.*[3] The error quoted for our result is the weighted standard deviation obtained from a consideration of 35 separate half-life determinations.

The cross section for excitation of the 2.3-MeV state is shown as a function of neutron energy in Fig. 4 and in Table I. The shape of the curve is very similar to that measured by Campbell and co-workers; however, our cross sections are almost exactly a factor of two smaller than theirs. The reason for this large discrepancy between our results and theirs is not understood; however, an analysis of our method indicates that our cross sections have an accuracy of $\pm10\%$. The usual threshold rise is observed here at 2.3 MeV. In addition, there are

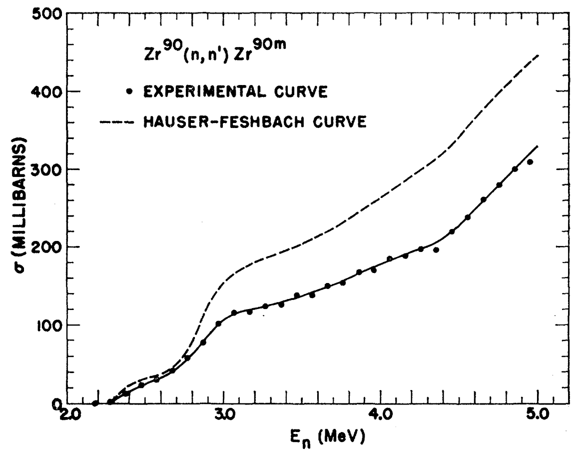

FIG. 4. $Zr^{90}(n,n')Zr^{90m}$ cross section as a function of energy.

marked rises in the cross section beginning at 2.75 MeV and at 4.4 MeV that are superimposed on the gradual increase in cross section. The first of these is due to a state at 2.75 MeV that had previously been observed by Day and Lind.[11] The second is from a state at 4.4 MeV that decays by cascading either to the 2.3-MeV state or to a higher state that decays in turn to the latter. The 4.4-MeV state had previously been observed in experiments[9,10] on the decay of Nb^{90} although evidence for it here was rather weak. From its mode of decay one would infer that it had a spin of 5 or greater. However, the shell model suggests that certain low states will be found in Zr^{90} whose transition to the excited states of the $(g_{9/2})^2$ configuration will be forbidden. On this basis it is conceivable that the 4.4-MeV state could have a spin as low as 3.

Figure 4 also shows a theoretical curve for the excitation of the 2.3-MeV state that is obtained from Hauser-Feshbach[12] theory. Moldauer[13] has recently shown that this theory is only an approximation to the proper statistical model theory of reactions, since it omits several important effects. However, it is instructive to use it to compare with experiment, and experience has shown it often works surprisingly well. In the calculation of this curve we have used transmission coefficients obtained from a Woods-Saxon[14] optical-model potential using parameters that were obtained by Ford and Gursky[15] from a detailed analysis of the available information on neutron elastic scattering by zirconium. These parameters are

$$V=45\text{ MeV},\qquad r_0=1.33\text{ F},$$
$$W=2.25+0.32E\text{ MeV},\qquad a=0.50\text{ F},$$

where E is the neutron channel energy. The calculation

[11] R. B. Day and D. A. Lind (private communication).
[12] W. Hauser and H. Feshbach, Phys. Rev. **87**, 366 (1952).
[13] P. A. Moldauer, Phys. Rev. **123**, 968 (1961).
[14] R. D. Woods and D. S. Saxon, Phys. Rev. **95**, 577 (1954).
[15] K. W. Ford and M. L. Gursky (private communication).

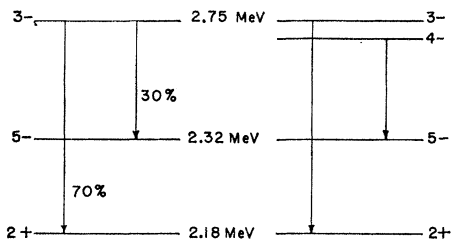

FIG. 5. Alternative level schemes for Zr^{90} near 2.7 MeV.

here takes into account all of the levels of Zr^{90} below 5 MeV that are given in *Nuclear Data Sheets*.[16] In addition, we have assumed that there are two degenerate levels at 2.75 MeV having odd parity and spins of 3 and 4. Only the latter is assumed to decay to the 5− state. If we assume that there is only a single state at 2.75 MeV with spin and parity 3−, and that this state decays to the 5− state with the branching ratio (see Fig. 5) observed by Day and Lind,[11] the results of the calculation remain essentially unchanged. The state at 4.4 MeV is assumed to be 3−.

Except near threshold, the experimental and theoretical curves have similar shapes, but the data for the experimental curve are 30% smaller than the latter. One can reduce the theoretical cross sections by reducing the magnitude of the imaginary part of the potential; however, when one does this, the agreement in shape of the curves is destroyed. In view of the approximations in the theory, it was not felt that forcing a better agreement would be significant.

From the increase in slope of the cross-section curve above 4.4 MeV, one can learn something about the spin of the 4.4-MeV level. Calculations with various spins assumed for this level showed that the higher the spin (for spins $\geqslant 3$) the smaller the increase in cross section. The magnitude of the experimentally determined increase in cross section appears to require a spin less than 4 when compared with the theoretical calculations, while considerations based on gamma-ray decay rates require a spin $\geqslant 3$. Thus, the most likely spin is 3. For Zr^{90} the calculations also show a dependence of the cross section on the parity of the level. This dependence arises because of the fact that a resonance in the optical model exists near here for odd-parity angular momentum waves at the same time that there is an antiresonance for even-parity waves. Thus, there is a preference for both the ingoing and outgoing neutrons to have odd parity, and as a result levels for which no parity change is required are excited with a greater probability than would exist if a parity change were necessary. However, this dependence on parity is not large enough in comparison with the uncertainties introduced in the approximations in the theory to enable one to come to a conclusion about the parity of the levels.

From a qualitative examination of the cross section results in Fig. 4, one can obtain some information on the 4− ($p_{1/2}g_{9/2}$) level that was discussed in the introduction. The only possible locations for this level appear to be close to 2.3 MeV (essentially equal in energy to the 5− level), at 2.75 MeV (equal in energy to the 3− level observed in proton and alpha-particle inelastic scattering),[6,7] or at 4.4 MeV. The threshold rise at 2.3 MeV does not appear to be large enough in comparison with the Hauser-Feshbach calculations to permit both a 5− and a 4− level to be present there. The level at 4.4 MeV appears to have too large a cross section to be accounted for by a 4− level. Furthermore, the spacing of 2.1 MeV between this and the 5− level is larger than the calcula- of Bayman[2] would indicate to be possible. Thus, the evidence is against the possibility that this is the 4− ($p_{1/2}g_{9/2}$) level, although this evidence is not conclusive.

It is most likely that the 4− level is at 2.75 MeV. As mentioned above, the Hauser-Feshbach calculations show that essentially no difference would be expected in this experiment from having either two levels at 2.75 MeV (3− and 4−) or a single 3− level that decays both to the 2+ level at 2.18 MeV and the 5− level with the branching ratio observed by Day and Lind.[11] Since it is hard to see how a collective 3− level could decay to the 5− state in competition with the $E1$ decay to the 2+ state, we conclude that the 4− level is at 2.75 MeV, essentially equal in energy to the collective 3− level observed in other experiments.[6,7]

V. ACKNOWLEDGMENTS

Most of the experimental work was done on the large Los Alamos electrostatic generator which was operated by R. E. Luders and Marie Murphy. We were greatly aided by R. K. Smith who built and calibrated the parallel plate fission chamber. We wish to thank Blair K. Swartz for preparing the code for handling the experimental data and programming it for the IBM 7090 computer. The rabbit transport system and its associated electronics were designed and built by R. C. Peck.

[16] *Nuclear Data Sheets*, complied by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C., 1961), NRC 60-4-27.