Measurement of the neutron differential elastic scattering from ⁹⁰Zr at 2.1 and 5.2 MeV

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> The differential elastic neutron cross section of 90 Zr was measured at 2.1 and 5.2 MeV. The Bettis pulsed Van de Graaff time-of-flight system was used in conjunction with a shielded neutron detector capable of γ ray rejection by pulse shape discrimination. Identical geometry measurements made on a polyethylene sample were used for normalization of the 90 Zr data to the H(n, n) cross section. Comparisons were made to the results of optical model calculations.

NUCLEAR REACTIONS 90 Zr(n, n), E = 2.1, 5.2 MeV; measured $\sigma(E, \theta)$; deduced optical-model parameters; resolution 100 keV; $\theta = 17-157^{\circ}$.

I. INTRODUCTION

The extensive use of zirconium as a structural material in nuclear reactors justifies a thorough study of its nuclear cross section description. Recent measurements^{1, 2} of inelastic scattering from the major zirconium isotopes together with recent total cross section measurements^{3,4} have provided new information. Prior elastic scattering measurements⁵⁻⁷ at energies near those used in this investigation have been made on natural zirconium. This paper is written to report elastic scattering measurements completed on the separated ⁹⁰Zr isotope at neutron energies of 2.11 and 5.17 MeV. It is hoped that the results of these measurements will supplement our knowledge of zirconium so that not only are more accurate cross sections provided at the energies measured, but an improved theoretical description will allow more accurate calculation of cross sections at energies where no measurements now exist. The Hauser and Feshbach statistical treatment corrected for level width fluctuations has been applied to provide a consistent framework for interpolation between the available data.

II. EXPERIMENTAL PROCEDURE

The Bettis Laboratory neutron time-of-flight facility was used to obtain elastic scattering data for a ⁹⁰Zr sample of the composition specified in Table I. The measurement system is shown in Fig. 1. The Bettis 3 MV Van de Graaff pulsed ion beam (~3 nsec duration at a 5 MHz repetition rate) was detected by a beam pickoff device prior to its incidence on the neutron producing target. Metal Zr³H and Ti²H targets were used to produce, through the ³H(p, n) and ²H(d, n) source reactions, respective neutron incident energies of 2.11 ± 0.11 and 5.17 ± 0.10 MeV. The respective neutron energy spreads indicated result from the ion beam degradation in the metal targets.

The right-circular cylinder scattering samples used in this investigation (i.e., 90 Zr and polyethylene) were positioned on the incident ion beam axis with their vertical axes 7.470 cm from target center. The 90 Zr sample was 1.94 cm high and 1.96 cm in diameter with a measured density of 6.43 g/cm³. The polyethylene sample, used for normalization of the 90 Zr scattering results to the H(*n*, *n*) scattering cross section, had effectively the same dimensions and a measured density of 0.914 g/cm³.

The detector used for observing the sample scattered neutrons, and shown in Fig. 1, consists of a 3.81 cm diam \times 3.81 cm thick shielded NE213 liquid scintillator. A 2.54 cm diam \times 2.54 cm thick stilbene detector was used for neutron flux normalization. A block diagram of the time-offlight electronics for these detectors is shown in Fig. 2. The pulse-shape discrimination properties⁸ of the NE213 scintillator are used to reject most of the pulses resulting from incident γ rays. This was found to be unnecessary in the case of the stilbene flux monitor aligned and shielded to view source neutrons only. A secondary neutron flux monitor consisted of a long counter which facilitated system calibration and stability checks performed routinely during the course of the measurements.

The 90 Zr scattering data were accumulated in alternate $\frac{1}{2}$ h periods, with and without the sample, at various scattering angles between 17.5° and 157.4° relative to the ion beam direction. Typical time-of-flight data accumulated at 90° are shown in Fig. 3. Data quality was continually

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Isotope ^a	Atomic composition (%)	
⁹⁰ Zr ⁹¹ Zr ⁹² Zr ⁹⁴ Zr ⁹⁶ Zr	97.72 1.07 0.51 0.56 0.15	

TABLE I. ⁹⁰Zr isotopic composition.

^a The ⁹⁰Zr sample was a solid metal casting provided by Oak Ridge National Laboratory. Dr. A. B. Smith at Argonne National Laboratory has not observed any light element contaminate scattering from this sample and estimates their presence at less than 2%.

monitored during these measurements with the aid of a PDP-9 computer utilizing a program which determined preliminary cross section results and associated statistics. Final data reduction includes elastic scattering peak integration, corrections for analyzer dead time, counter resolving time losses, and background subtraction. The technique of background subtraction used with the NE213 detector deserves special mention. Since the time-random background is a function of accelerator beam conditions [i.e., neutron production due to (d, n) and (p, n) reactions in the analyzing magnet area, etc.], it is determined separately for the sample-in and sample-out data obtained from the data region corresponding to flight times less than that of the maximum energy source neutron (i.e., time-random background range, Fig. 3). The average background/channel in each case is then subtracted from the respective flux normalized sample-in and sample-out data over the channel range used for the elastic scattering peak integration. The relative differential elastic cross section is then determined by subtracting the remaining sample-out background events from the remaining sample-in events. This two-stage technique of background subtraction was found to yield more reproducible results than those obtained by simply subtracting the flux normalized sample-out data from the sample-in data over the elastic scattering peak range.

Typical time-of-flight data accumulated at 50.2° for the polyethylene sample are shown in Fig. 4. As can be seen, the neutrons scattered from hydrogen are easily separated by the time-offlight system from those scattered from carbon. Before the observed relative differential scattering cross sections can be normalized to the H(n,n)reaction cross section, the relative efficiency of the NE213 detector is required. Separate measurements were conducted in which time-of-flight data were accumulated with this detector at 10° intervals between 0° and 150° for both the ${}^{2}H(d,n)$ and ${}^{3}H(p, n)$ source reactions with the target positioned at the pivot point. The relative efficiencies of the detector system at the two instrumental gains used were determined from the known kine-



FIG. 1. Measurement system.

matics⁹ of these reactions and the corresponding differential cross sections.^{10, 11} The respective relative efficiencies were then used to correct the H(n, n) scattered neutron counts to the efficiencies appropriate to the ⁹⁰Zr elastic scattered neutron counts in each case.

III. EXPERIMENTAL RESULTS

The relative [i.e., yet to be normalized to H(n, n) cross section] ⁹⁰Zr differential scattering data, corrected for background, were fitted by a Legendre polynomial program adapted from the work of Bevington.¹² The data were weighted in accordance with the counting statistics. This program provided the "measured" laboratory angular distribution at the 33 equally spaced values of the scattering angle, necessary for processing by the MAGGIE Monte Carlo program.¹³

The MAGGIE program, with this data as input, was then used to make the necessary corrections for flux attenuation and multiple scattering. Additional MAGGIE calculations were carried out for both the 90 Zr and polyethylene sample at 50.2 $^{\circ}$ to obtain accurate corrections for flux attenuation and multiple scattering needed for normalization to the H(n, n) cross section. The resulting total elastic cross sections obtained are 4.356 ± 0.35 and 2.21 ± 0.22 b at the respective energies of 2.11 and 5.17 MeV. Respective hydrogen cross sections used in the normalization were 2.814 and 1.585 b.¹⁴ The standard deviations of 10% placed on the measured cross sections are estimates based on the experimental statistics and the uncertainties in the measured detector efficiency, flux attenuation, and multiple scattering corrections.

The differential cross section results for 2.11



FIG. 2. Time-of-flight system electronics.



FIG. 3. Typical ⁹⁰Zr scattering data at 90°.





TABLE II. ⁹⁰Zr differential elastic scattering results for 2.11 and 5.17 MeV (corrected for multiple scattering effects).

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$\cos \theta_{ m lab}$	$ heta_{ ext{lab}}$ at 2.11 MeV (mb/sr)	$ heta_{lab}$ at 5.17 MeV (mb/sr)	
0.954	1456 ± 43	1604 ± 45	
0.870	966 ± 30	636 ± 20	
0.760	615 ± 21	108 ± 5	
0.640	424 ± 15	16.7 ± 1.4	
0.480		124 ± 6	
0.410		167 ± 4	
0.340	257 ± 12	191 ± 4	
0.199	232 ± 8	179 ± 8	
0.101	219 ± 9		
0.000	203 ± 7	90.4 ± 4.7	
-0.199	166 ± 8		
-0.261		15.9 ± 1.8	
-0.400	138 ± 5		
-0.520	135 ± 5	34.3 ± 3.3	
-0.640	147 ± 7	45.4 ± 3.8	
-0.801	206±8	49.1 ± 5.2	
-0.923	374 ± 12	66.6 ± 4.3	

and 5.17 MeV neutrons elastically scattered from 90 Zr are listed in Table II and shown in Figs. 5 and 6, respectively. The data and their associated Legendre polynomial fits, which represent the elastic angular distribution before and after multiple scattering corrections have been applied, are all normalized to the measured total elastic cross section. The standard deviations, appropriate for the relative angular distribution, were determined from the experimental counting statistics and do not include the H(n, n) normalization uncertainty discussed above. The curves included in Figs. 5 and 6 were obtained from Legendre polynomial fits weighted with the inverse squares of the standard deviations from Table II. The respective Legendre coefficients in the laboratory system are defined by the following equation:

$$\sigma(\theta_{1ab}) = \frac{\sigma_{s0}}{2\pi} \sum_{L=0}^{\infty} \frac{1}{2} (2L+1) f_L P_L(\cos\theta_{1ab}) , \quad (1)$$







0.4 0.2 0 -0.2 -0.4 COS (*θ*_{lab})

FIG. 6. ⁹⁰Zr differential elastic cross section at 5.17 MeV (laboratory system).

where $\sigma_{\rm S0}$ is the measured total elastic cross section.

10 -

0.8

0.6

σ(θ_{iab}) (mb/sr)

IV. OPTICAL-MODEL ANALYSIS

An optical potential, consistent not only with the measured angular distributions of elastically scattered neutrons, but also with measurements of total^{3, 4} and inelastic cross sections,^{1, 2, 15} was determined. For a spherical nucleus such as ⁹⁰Zr, this potential takes the form

$$U(r) = -Vf(r, a) + 4 ibW \frac{d}{dr} f(r, b)$$
$$-\frac{2}{r} V_s \frac{d}{dr} f(r, a)\vec{L} \cdot \vec{\sigma} , \qquad (2)$$

-0.8

-0.6

-1.0

where

$$f(r, x) = \left[1 + \exp\left(\frac{r - R_0}{x}\right)\right]^{-1},$$

and \vec{L} and $\vec{\sigma}$ refer to the orbital angular momen-

tum and spin of the bombarding neutron, respectively. In the present work, the values

$$a = 0.66$$
 fm, $b = 0.47$ fm, and $R_0 = 1.27 A^{1/3}$,
(3)

which were found by Elwyn *et al.*¹⁶ to give good agreement with their measured angular distributions of neutrons elastically scattered by Zr and Nb at energies below 0.9 MeV, have been adopted.

Preliminary values of the remaining parameters of Eq. (2) were fixed by fitting the differential elastic cross sections measured at 5.17 MeV. Shape elastic cross sections and transmission coefficients were calculated using the ABACUS-2 program¹⁷ while compound elastic cross sections, corrected for level width fluctuations, were obtained with the NEARREX code.¹⁸ In the latter calculations the Moldauer parameter Q^{19} was set to zero, the value found in Ref. 2 to give the best agreement with measured level excitation cross sections. The level scheme²⁰ employed in our optical-model calculations is given in Fig. 7. The best fit to the 5.17 MeV angular distribution was obtained using

$$V = 45.51$$
 MeV, $W = 7.61$ MeV,

and

 $V_{s} = 5.2$ MeV.

Previous work^{2, 3} indicated that over the energy range from 0.2 to 5 MeV the optical parameters, with the exception of V, could be taken as energy independent; V was found to be adequately represented by



FIG. 7. ⁹⁰Zr level structure and decay scheme.

(4)

E (MeV)	S0 (Expt.)	S0 (Calc.)	L	f_L (Expt.)	f _L (Calc.)
2.11 4.356±0.35 b	4.356±0.35 b	4.247 b	0	1	1
			1	0.3196 ± 0.0065	0.2929
			2	0.2728 ± 0.0056	0.2843
		3	0.0788 ± 0.0057	0.0928	
		4	0.0968 ± 0.0051	0.1006	
		5	0.0175 ± 0.0046	0.0153	
		6	0.0190 ± 0.0051	0.0148	
			7	0.0007 ± 0.0035	0.000232
5.17 2.21±0.21 b	2.21 ± 0.21 b	2. 173 b	0	1	1
			1	0.5831 ± 0.0074	0.5845
			2	0.4250 ± 0.0079	0.4448
			3	0.3068 ± 0.0068	0.3343
			4	0.3019 ± 0.0050	0.3093
			5	0.2237 ± 0.0039	0.2285
			6	0.1024 ± 0.0040	0.1128
			7	0.0134 ± 0.0033	0.0244
			8	0.0006 ± 0.0026	0.0106
			9		0.00172
			10		0.00040

 ${\tt TABLE \ III.} \ {\tt Experimental \ and \ calculated \ center-of-mass \ Legendre \ coefficients}.$

(5)

We, therefore, set $V_0 = 47.22$ MeV and tested the preliminary parameters by comparing computed and experimental angular distributions of scattered 2.11 MeV neutrons. Calculated and mea-

 $V = V_0 - 0.33E$ MeV.

sured integrated elastic scattering cross sections as well as the center-of-mass Legendre coefficients $f_L^{c.m.}$, defined by

$$\sigma(\theta_{c,m_*}) = \frac{\sigma_{S0}}{2\pi} \sum_{L=0}^{\infty} \frac{1}{2} (2L+1) f_L^{c,m_*} P_L(\cos\theta_{cm}) , \quad (6)$$



FIG. 8. 90Zr differential elastic cross section at 2.11 MeV (center-of-mass system).

are given in Table III. One notes that the calculated σ_{so} of 2.17 b is in good agreement with the measured value, 2.21 ± 0.21 b.

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In order to facilitate the comparisons of measured angular distributions with those obtained with the optical model, we have, in Figs. 8 and 9, normalized both sets of values by dividing out the total elastic cross section σ_{so} . Both of the figures emphasize the importance of compound elastic scattering which completely dominates the elastic scattering of 2.11 MeV neutrons in the vicinity of $\cos(\theta_{c.m.}) = -0.6$ and that of 5.17 MeV neutrons at the sharp minimum at $\cos(\theta_{c.m.}) = 0.68$.

As a further test of the applicability of the parameters of Eqs. (3) and (4), total cross sections were calculated for energies between 0.5 and 2.4 MeV. A comparison of these calculated values with the measurements of Stooksberry and Anderson³ and with preliminary results ob-

tained by Green, using the methods described in Ref. 4, is given in Fig. 10. The Green data, obtained with a ²⁵²Cf fission source, has been averaged over the energy blocks indicated. As is evident from this figure, the experimental cross sections display fluctuations, due to resonance scattering, which an optical-model calculation cannot reproduce. The model does, however, give a good estimate of the average total cross section for an energy interval, say 0.1 MeV, containing many resonances. Above 1.9 MeV, the calculated total cross sections are significantly greater than those measured by Stooksberry and Anderson, but agree well with Green's data when the latter are averaged over 50 keV intervals. This discrepancy is at present unresolved.

IV. CONCLUSIONS

As indicated in Table III, the parameters of Eqs. (3) and (4) have been used to calculate the



FIG. 9. ⁹⁰Zr differential elastic cross section at 5.17 MeV (center-of-mass system).





FIG. 10. ⁹⁰Zr total cross section vs neutron energy.

differential elastic cross section at 2.11 MeV. While the calculated total elastic cross section, 4.247 b, is in reasonable agreement with the measured 4.36 ± 0.36 b, the calculated Legendre coefficients are less satisfactory. In particular, the calculated f_L is 9% lower than the measured value. One possible explanation of this behavior is that, as suggested by the Stooksberry-Anderson data (Fig. 10), there is a resonance near 2.11 MeV. In the vicinity of such a resonance, the f_L are subject to dramatic fluctuations.²¹ An

alternative explanation is that energy dependence of the optical parameters is more serious than indicated in Eqs. (3) and (4). At the present time, we do not favor this interpretation since work on other isotopes in the vicinity of A = 90 indicates that such is not the case.

Finally, level excitation cross sections have been calculated and are in satisfactory agreement with experiment.^{2, 22} The calculated values are negligibly different from those previously reported in Ref. 2. The parameters of Eqs. (3) and

9.5

9.0

(4) are thus adequate to calculate neutron cross sections over the range 0.2 to 5.2 MeV. We, of course, make no claims of uniqueness for this particular set of parameters. Since, to the first order, scattering and reaction amplitudes are Fourier transforms of VR_0^2 and WR_0^2 , respectively, calculated cross sections are insensitive to variations of parameters which conserve these products. After the present investigations were completed, the authors became aware of recent measurements of 90 Zr total and elastic cross sections reported by P. Guenther, A. Smith, and J. Whalen of Argonne National Laboratory. Their 90 Zr total and elastic neutron cross sections are consistent with the results of the present work.

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