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ANL/NDM-69

Fast-Neutron Scattering Cross Sections of Elemental Zirconium

by

A.B. Smith and P.T. Guenther

December 1982

ARGONNE NATIONAL LABORATORY,
ARGONNE, ILLINOIS 60439, U.S.A.

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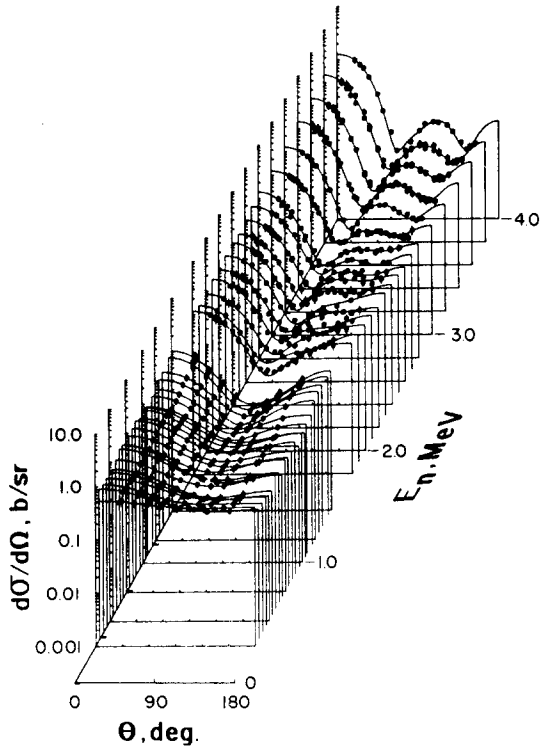

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*This work supported by the U.S. Department of Energy

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FAST-NEUTRON SCATTERING CROSS SECTIONS of ELEMENTAL ZIRCONIUM*

by

A. B. Smith and P. T. Guenther

ABSTRACT

Differential neutron-elastic-scattering cross sections of elemental zirconium are measured from 1.5 to 4.0 MeV at intervals of $\leq$ 200 keV. Inelastic-neutron-scattering cross sections corresponding to the excitation of levels at observed energies of; 914 ± 25, 1476 ± 37, 1787 ± 23, 2101 ± 26, 2221 ± 17, 2363 ± 14, 2791 ± 15 and 3101 ± 25 keV are determined. The experimental results are interpreted in terms of the optical-statistical model and are compared with corresponding quantities given in ENDF/B-V.

*This work supported by the U.S. Department of Energy.

I. INTRODUCTION

The present work was undertaken as a part of a comprehensive study of the interaction of few-MeV neutrons with light-mass fission products. The objectives were the explicit determination of zirconium neutron-scattering cross sections and the derivation of a spherical optical-statistical model (OM) generally applicable to this mass-energy region and suitable for extrapolation to other and experimentally inaccessable processes in radioactive fission products. Previous portions of this work dealing with palladium, rhodium and silver are described in Refs. 1, 2 and 3.

Elemental zirconium consists of the five isotopes ^{92}Zr(51.5%), ^{91}Zr(11.2%), ^{92}Zr(17.1%), ^{94}Zr(17.4%) and ^{96}Zr(2.8%). These isotopes are significant fission-products. For example, the yields from ^{239}Pu fission induced by fission-spectrum neutrons are: $\approx$ 2.2%(^{90}Zr), $\approx$ 2.6%(^{91}Zr), $\approx$ 3.1%(^{92}Zr), $\approx$ 4.4%(^{94}Zr) and $\approx$ 5.1%(^{96}Zr). For all of the isotopes compound-elastic (CE) scattering is relatively large up to several MeV, and particularly so for ^{90}Zr which is magic in neutron number. Therefore, compound-nucleus (CN) processes are a consideration throughout the energy range of the present measurements.

Subsequent portions of this report deal with: (Sec. II) a brief outline of the experimental method, (Sec. III) the experimental results, (Sec. IV) the model derivation, and (Sec. V) comparisons with respective values given in ENDF/B-V.[4]

II. EXPERIMENTAL METHODS

The neutron-scattering samples were 2 cm diameter, 2 cm long cylinders fabricated of high-purity elemental metal. All of the measurements were made using the time-of-flight-technique and the Argonne ten-angle detection apparatus. The neutron source was the ^{7}Li(p;n)^{7}Be reaction, pulsed on for durations of $\approx$ 1 nsec at a repetition rate of 2 MHz. The scattered neutrons were detected with proton-recoil scintillators placed at the ends of $\approx$ 5.5 m flight paths. The measurements were made relative to the neutron total cross sections of carbon[5] using the method described in Ref. 6. All experimental results were corrected for multiple events, beam attenuation and angular resolution as described in Ref. 7. Generally, the experimental methods were analogous to those employed at the Argonne National Laboratory for a number of years and described in detail in Refs. 7, 8 and 9.

III. EXPERIMENTAL RESULTS*

A. Neutron Elastic Scattering

The experimental measurements were made at incident energies from 1.5 to 4.0 MeV. At the low-energy extreme of the measurements fluctuations are a

*The experimental results have been transmitted to the National Nuclear Data Center, Brookhaven National Laboratory.

concern, therefore relatively broad incident-neutron energy spreads of 50 to 70 keV were employed and the measurements were made at intervals of $\approx$ 50 keV below 3.0 MeV. Distributions measured at adjacent energies were averaged to further smooth any residual fluctuations. The statistical uncertainties of the individual differential values were $\lesssim$ 3%. The detector-calibration uncertainties were similarly $\lesssim$ 3%. Relative angular uncertainties were $\lesssim \pm$ 0.1 degrees and absolute angular uncertainties were $\lesssim \pm$ 0.7. The correctional procedures introduced $\lesssim$ 1% uncertainties. Thus the overall differential-elastic-scattering-cross-section uncertainties were $\lesssim$ 5%. The experimental results are summarized in Fig. 1. They generally behave in an energy smooth manner with no indication of fluctuations. The measured differential-elastic-scattering results were least-square fitted with 6th-order Legendre-polynomial series in order to obtain the angle-integrated elastic-scattering cross sections. The quality of the Legendre-polynomial-series fits to the data is illustrated in Fig. 1. The resulting angle-integrated elastic-scattering cross sections are shown in Fig. 2 Their uncertainties are $\lesssim$ 5%.

The present differential-elastic-scattering results are consistent with the isotopic ^{90}Zr and ^{92}Zr values of Ref. 10, considering the isotopic content of the element. The angle-integrated elastic-scattering results reasonably extrapolate to the lower-energy elemental values of Ref. 11 and are consistent with the recent neutron total cross sections of Refs. 12 and 13, as illustrated in Fig. 2.

B. Neutron Inelastic Scattering

The broad resolutions necessary for reliable energy-averaged elastic-scattering cross sections precluded optimum resolution of inelastically-scattered neutrons. Moreover, the element contains four isotopes, one of which is odd, resulting in a high elemental level density and, consequently, complex, scattered-neutron spectra. Despite these obstacles, nine inelastic-scattered neutron groups were observed corresponding to the apparent elemental level structure given in Table 1. The cited excitation energies are simple averages of a number of measurements with the uncertainties defined as the RMS derivations from the averages. The first few observed levels closely correspond to reported levels[14] in ^{90}Zr, ^{92}Zr and ^{94}Zr as outlined in Table 1. Above $\approx$ 2.2 MeV this correspondence becomes increasingly complex and uncertain.

The neutron inelastic-excitation cross sections were deduced from the observed differential values as described above for elastic scattering. The angular distributions were nearly isotropic thus the Legendre-polynomial series fitted to the data were of low order (e.g., $\lesssim$ 2). The corresponding uncertainties were as outlined above for elastic scattering with a much larger statistical component in the 10 to 30% range. Some of the inelastic-excitation cross sections were distorted by the second neutron group from the source reaction. In those cases appropriate corrections were made. The resulting angle-integrated inelastic-scattering cross sections are summarized in Fig. 3. These results are reasonably consistent with the ^{90}Zr and ^{92}Zr values of Ref. 10 at low energies, particularly as ^{92}Zr and ^{94}Zr level properties are

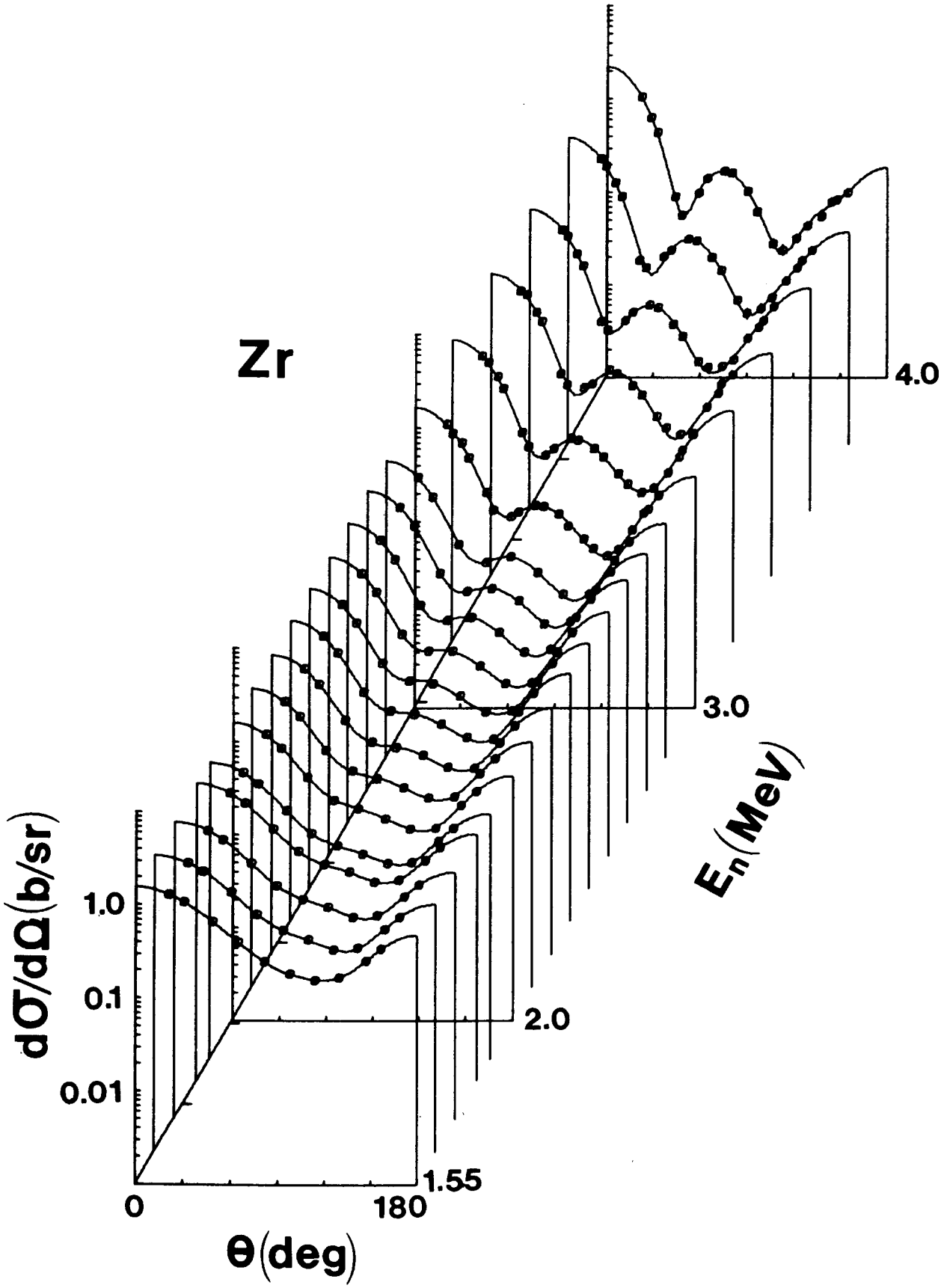

Fig. 1. Differential-Neutron-Elastic-Scattering Cross Sections of Elemental Zirconium. The present measured values are indicated by data symbols. Curves indicate the results of Legendre-polynomial-series fits to the measured values.

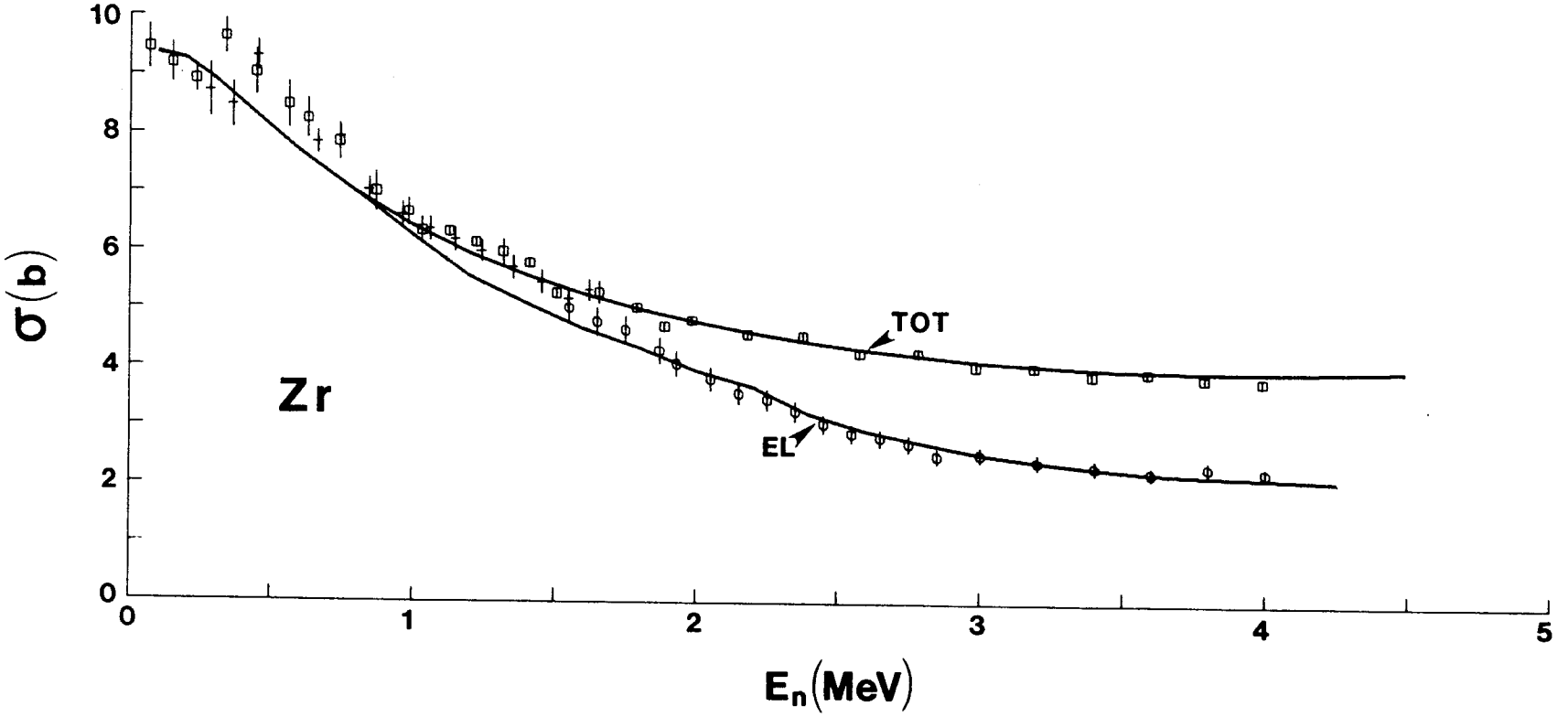

Fig. 2. Neutron Total and Elastic-Scattering Cross Sections of Elemental Zirconium. The present measured elastic-scattering values are indicated by 0. 100-keV averages of the neutron total cross sections of Refs. 12 and 13 are indicated by □ and +, respectively. Curves indicate the results of model calculations as discussed in the text.

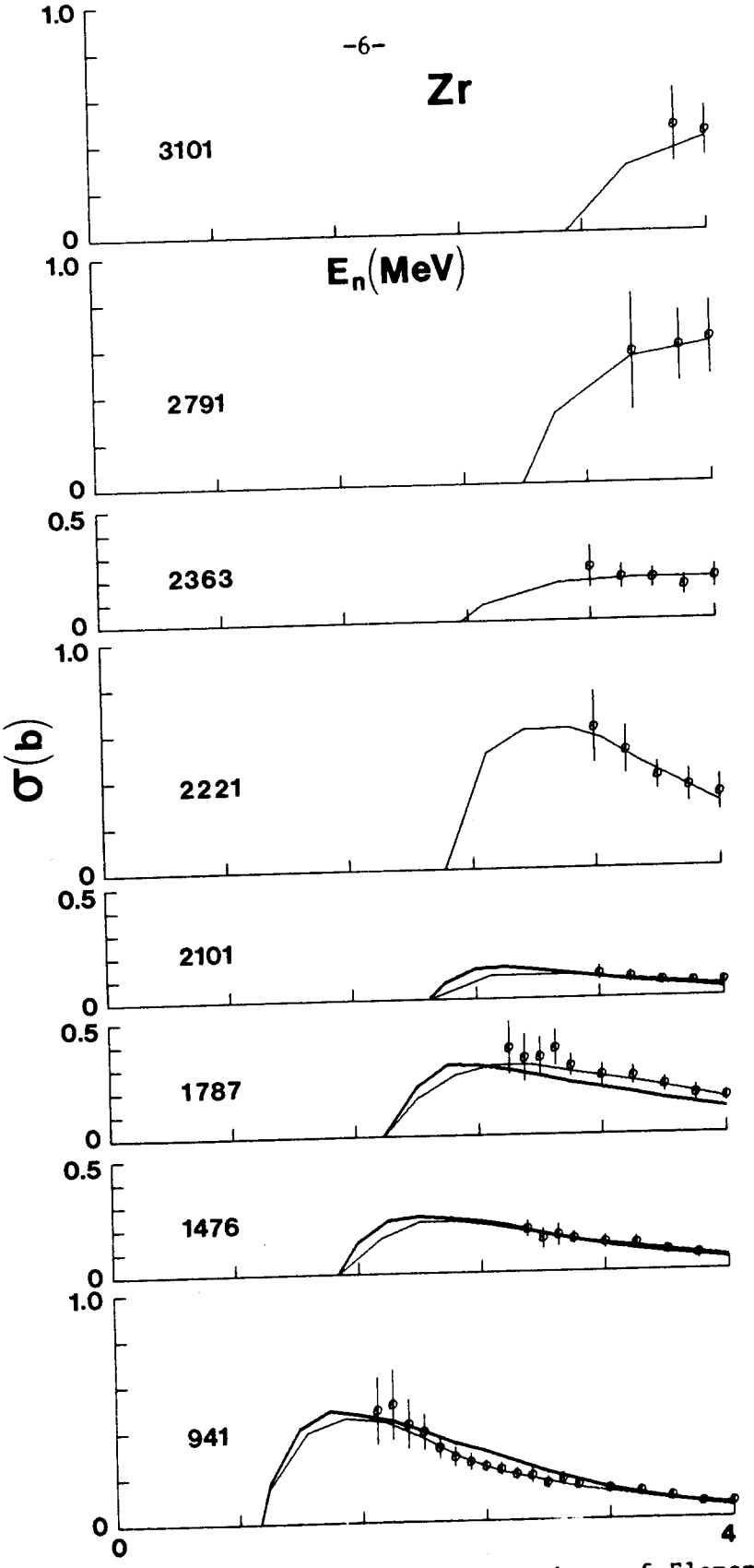

Fig. 3. Neutron Inelastic-Scattering Cross Sections of Elemental Zirconium. The present experimental values are indicated by O. Light curves are "eyeguides" and heavy curves the results of model calculations. Excitation energies in keV are numerically cited in the respective sections of the figure.

similar in this region. The sum of the "eyeguides" of Fig. 3 is slightly smaller than the nonelastic cross section implied by the above elastic-scattering results and the neutron total cross sections of Ref. 12. This is not surprising since the present experiments probably were not sensitive to very small contributions from the complex level-structure of ^{91}Zr.

IV. INTERPRETATION AND DISCUSSION

It was assumed that the experimental results could be described in terms of a spherical OM[15]. The derivation of the OM parameters was complicated by the isotopic composition of the element as the level structure differs from isotope to isotope. A consequence is different CN, particularly CE, contributions from the different isotopes and these can be large at the energies of the present experiments. These obstacles were managed by assuming a reasonable set of OM parameters, calculating the CE cross sections for each isotope and combining them to obtain the elemental CE cross sections. The latter were subtracted from the measured differential-elastic-scattering cross sections to obtain the "experimental" differential shape-elastic cross sections (SE). The differential SE cross sections were then concurrently chi-square fitted with the OM, simultaneously adjusting the six parameters; real and imaginary strengths, radii and diffusenesses. The resulting parameters were then used to re-determine the CE and SE cross sections and the fitting procedure repeated. The process was followed through several cycles until the resulting OM parameters appeared to be stable. The CN calculations used the Hauser-Feshbach formula[16], as modified by Moldauer[17]. All the calculations employed the computer code ABAREX[18]. Discrete-level properties were taken from Ref. 14. The level structure is well defined for ^{90}Zr throughout the energy range of interest. Only the lower-energy levels are reasonably known for the other isotopes. Where the level structure became uncertain the statistical formalism and parameters of Gilbert and Cameron[19] were used. A correlation of the experimental observations with reported levels is indicated in Table 1.

The OM parameters resulting from the above fitting procedures are defined in Table 2. The real strength (measured as the integral per nucleon (J_V/A) or $V \times r^2$) is similar to that found generally applicable in this mass-energy region[20]. The imaginary strength is relatively small as frequently encountered near shell closures (N=50)[21]. The imaginary radius is larger than the real radius as has been reported from considerations of low-energy strength functions[22]. The parameters of Table 2 provide a good description of the measured elastic-scattering cross sections as illustrated in Fig. 4. More than 90% of the measured values are consistent with the calculated quantities to within the experimental uncertainties alone. The calculations also reasonably describe the angle-integrated elastic scattering cross sections and the neutron total cross sections of Refs. 11 and 12 as illustrated Fig. 2. The calculated angle-integrated elastic-scattering cross sections are consistent with the measured values to within the 5% experimental uncertainties throughout the measured energy range. There may be some discrepancy between measured and calculated neutron total cross sections at low energies but the measured values are subject to large fluctuations, even in the illustrated 100-keV averages, and thus the differences may be due to residual fluctuation effects. It is noted that the low-energy strength functions of the various zirconium isotopes are quite different[23]; possibly reflecting large fluctuations.

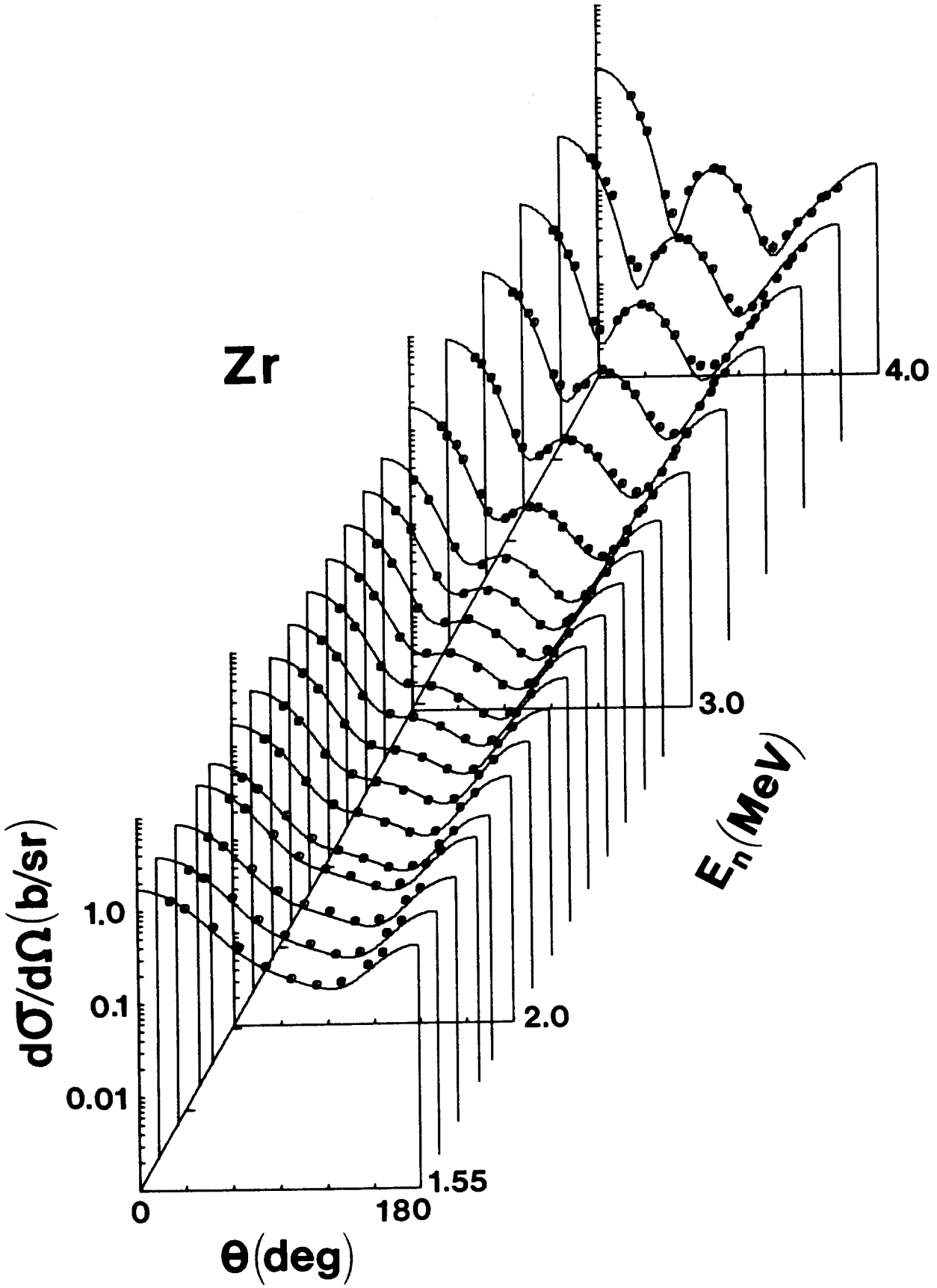

Fig. 4. Comparison of Measured (0) and Calculated (Curves) Elastic-Scattering Cross Sections of Elemental Zirconium.

The measured inelastic-scattering cross sections were not particularly well resolved, as noted above. However, assuming the correlations between observed and reported ^{90}Zr, ^{92}Zr and ^{94}Zr levels indicated in Table 1, the first few inelastic-excitation cross sections were calculated with the results shown in Fig. 3. Generally, the agreement between measured and calculated values is qualitatively acceptable. The calculated results appear to reasonably account for channel competition suggesting that the statistical parameters of Ref. 19 are suitable. Calculations of higher-energy excitations were not attempted due to the uncertain nature of the contributing levels.

Nearly ten years ago measurements of ^{90}Zr and ^{92}Zr scattering cross sections were made at this laboratory[10]. These isotopic results are not as comprehensive nor of as good quality as the present work but their interpretation is free of the elemental complexities cited above. Therefore, the previously reported ^{90}Zr and ^{92}Zr data were re-analyzed using fitting procedures identical to those employed above. The resulting potential parameters were similar to the elemental values given in Table 2. The respective real and imaginary strengths are noted in footnote "d" of Table 2. Again, the imaginary strengths are somewhat smaller than commonly encountered in global potentials.

V. COMPARISONS WITH ENDF/B-V

The ENDF/B-V file system[4] presents zirconium neutron cross sections on an isotopic basis. Those isotopic components were combined, weighted by isotopic abundance, to obtain the elemental neutron total, elastic- and inelastic-scattering cross sections for comparison with the present experimental results. These evaluated elemental cross sections should be representative of the previously reported data base. The measured and evaluated neutron total and elastic-scattering cross sections are compared in Fig. 5. The agreement is good. This implies that the nonelastic cross sections deduced from the present measurements are consistent with the total inelastic-scattering cross sections given in the file. This agreement may be somewhat fortuitous as the file appears to contain identical neutron total cross sections for ^{90}Zr, ^{91}Zr, ^{92}Zr and ^{94}Zr, even to the fluctuating structure, while in the few-MeV region the evaluated elastic-scattering cross sections of the isotopes differ by as much as one barn ($\approx$ 30%) in some instances.

VI. SUMMARY COMMENT

The present measurements well define the energy-averaged differential-elastic-scattering cross sections of elemental zirconium from 1.5 to 4.0 MeV. They also improve the understanding of elemental inelastic-scattering cross sections over the same energy range. The present elastic scattering results and the neutron total cross sections of Refs. 12 and 13 imply nonelastic cross sections from 1.5 to 4.0 MeV which are consistent with the present measured inelastic-scattering cross sections. An optical-statistical model deduced from the measurements provides a good description of the observables. The model parameters are similar to those found generally applicable in this mass-energy region and are an input to the formulation of a "regional" potential suitable for the description of fast-neutron interactions with light-mass fission products[20]. The experimental results are in good agreement with the elemental

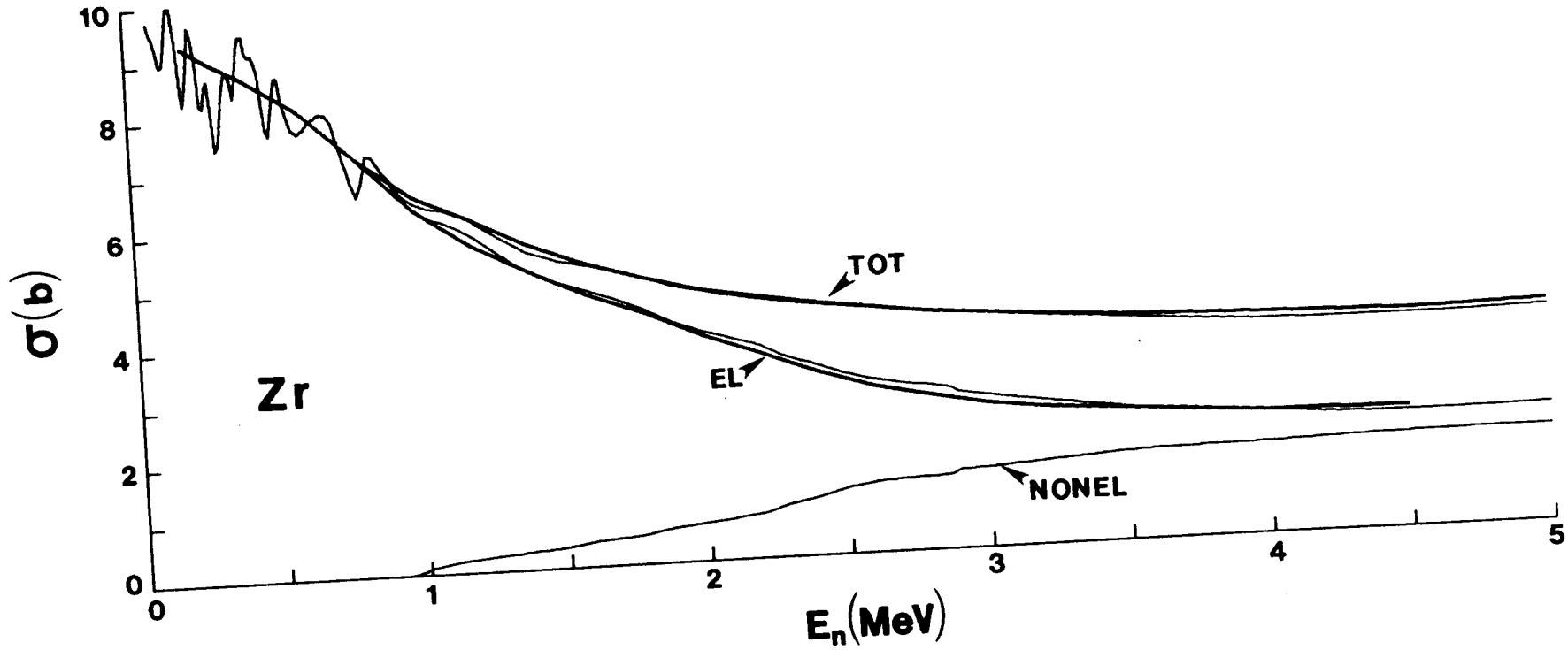

Fig. 5. Comparisons of Measured and Evaluated Neutron Total and Elastic Scattering Cross Sections of Elemental Zirconium. The heavy curves are "eyeguides" constructed through the present experimental values and the neutron total cross sections of Ref. 12 and 13. The light curves are taken from the evaluated quantities of ENDF/B-V[4]. The nonelastic cross section implied by the evaluation is also indicated.

elastic-scattering cross sections constructed from the contributing isotopic evaluations of ENDF/B-V[4].

ACKNOWLEDGEMENTS

The authors are indebted to Drs. P. Moldauer, E. Pennington and W. Poenitz for helpful comments and for making available experimental information prior to publication.

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TABLE 1. Observed Excitation Energies of Elemental Zirconium (in keV)

	Excitation Energies			
No.	Exp.	^{90}Zr[a]	^{92}Zr[a]	^{94}Zr[a]
1	941 ± 25	-	935(2+)	918(2+)
2	1476 ± 37	-	1382(0+)	1300(0+)
			1496(4+)	1469(4+)
3	1787 ± 23	1751(0+)	1847(2+)	1670(2+)
4	2101 ± 26	-	2056(2+)	2057(3-)
5	2221 ± 17	2186(2+)	2150(?)	2150(2+)
6	2363 ± 14	2319(5-)	-	many -
7	2791 ± 15	2736(4-)	-	many -
		2748(3-)		
8	3101 ± 25	3077(4+)	-	many -
9	3331 ± ?	3309(2+)	-	many -

[a]See Ref. 14.

TABLE 2. Optical-Model Parameters for Elemental Zirconium

Real Potential[a]

V_o = 49.41 MeV

r_v = 1.221 F[b]

a_v = 0.748 F

Vr^2 = 73.66 MeV-F^2

J_v/A^d = 445.42 MeV-F^3

Imaginary Potential[c]

W = 7.68 MeV

r_w = 1.416 F[b]

a_w = 0.398 F

Wa = 3.05 MeV-F

J_w/A^d = 69.21 MeV-F^3

[a]Saxon form, assume energy dependence of the form $V=V_o-0.3 \times E(MeV)$, assume real spin orbit potential of the Thomas form with 6 MeV strength and $r = r_v$.

[b]All radii expressed as $R = r \times A^{1/3}$.

[c]Saxon-derivative form.

[d]Comparable values for ^{90}Zr are J_v/A=474.04 and J_w/A=62.69 MeV-F^3 and for ^{92}Zr J_v/A=438.70 and J_w/A=57.62 MeV-F^3 derived from fits to the data of Ref. 10 as described in the text.