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Neutron Total Cross Sections, 2.5–15 MeV. I. Experimental*

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We have completed measurements of the total cross section, for neutrons between 2.5 and 15 MeV, of 78 natural elements and 14 artificial or isotopically enriched samples spanning the range from hydrogen to plutonium. The measurements were made by transmission in good geometry (inscattering <0.3%), using time-of-flight analysis of the continuous spectrum of neutrons from a pulsed Li(d,n) source. Data were taken with moderate energy resolution (2 to 4%) and moderate precision (1 to 3% at each point) at a density of three or four points per resolution interval, in order to avoid leaving gaps. We believe systematic errors do not exceed 2%, except in particular cases. Our method leads naturally to small samples, so that we were able to include all of the stable elements except the noble gases. The resulting homogeneous set of data supplies a reference surface for the intercomparison of results from other laboratories. We have made detailed comparisons with virtually all previous measurements in this energy range, and we summarize the systematic properties of the major ones. The completeness of the set helps to reveal a number of systematic properties of the interaction of fast neutrons with nuclei over the entire range of masses.

I. INTRODUCTION

In a previous paper[1] (hereafter referred to as M) we described a method suitable for the systematic measurement of total cross sections between 2.5 and 15 MeV, under conditions of moderate resolution and precision appropriate to filling major gaps in the previous body of measurements. In the present paper (described as I) we shall bring the description of the experimental method up to date, and present the main experimental results. Two short papers derived from this series of measurements have already appeared. The first,[2] which dealt with a spectroscopic problem in B^{10}, grew out of our routine use of the $Be^9(d,n)B^{10}$ reaction for calibration purposes. The second[3] applied our measurement of the neutron total cross section of deuterium to the question of the nonexistence of the dineutron.

In the sections which follow, we first review briefly the status of fast-neutron total cross sections at the time the work described here was undertaken, and present the rationale for conducting a factory-scale measurement under the particular conditions which we chose. We next review our experimental technique and apparatus, including improvements incorporated since the publication of M. Then we present the results of the measurements as succinctly as possible, together with a discussion of the random and systematic errors of the measurement. Because of our relatively complete coverage of the mass scale within this energy range, we shall devote considerable attention to comparisons with the major groups of previous measurements. Our discussion of the interpretation of the results, on the other hand, will be primarily qualitative and relatively brief. A detailed comparison[4] between these measurements and nonlocal optical-model calculations is presented in a companion paper, hereafter referred to as II.

The energy range covered by our work is a transition region. For the lightest elements, the total cross section still displays resonances in this range, arising from individual levels in the compound nucleus. For somewhat higher mass numbers the heavily overlapped resonances fuse into one or another type of fluctuation at the low end of our energy range, and the fluctuations damp out altogether at the high end. The heaviest elements display only the broad diffraction structure. This structure is generated as the neutron wave that penetrates the nucleus interferes with the wave which has swept around the outside of the nucleus.

Correspondingly, our moderate-resolution measurements should furnish a limited amount of spectroscopic information for the lightest elements, and at least some qualitative information regarding the cross-section fluctuations in somewhat heavier elements. Their major application, however, should be in testing improved models of the many-nucleon nucleus, since a successful model must describe the diffractionlike giant resonances accurately and in detail with a minimum number of parameters.

Although total cross sections have been under investigation for more than 30 years, systematic measurements of good quality in the MeV region did not begin to appear until the early 1950's. In 1952, Barschall[5] first displayed portions of the smooth surface exhibited by a three-dimensional plot of total cross section vs neutron energy vs

nuclear mass (or radius). His observation was based primarily on the pioneering measurements on 23 elements by Miller *et al.*,[6] which concentrated on lower energies but included isolated[7] points from 1.4 to 3.4 MeV [the upper limit of the $H^3(p,n)$-He^3 reaction with 4-MeV protons]. In the same year Coon, Graves, and Barschall[8] presented a 58-element slice across the surface, parallel to the mass axis, using $H^3(d,n)He^4$ neutrons at 14.1 MeV. The former measurement showed diffractionlike maxima which moved towards higher energies with increasing nuclear radius. The latter demonstrated that the smooth variation with radius persisted at least as high as 14 MeV. The unexpected regularity of these results stimulated immediate application of early versions of the optical model[9] to the few-MeV region, with considerable success.

During the remainder of the 1950's, measurements continued on many elements, predominantly below 3 MeV and at 14 MeV. The region in between, which contains a diffraction maximum for half of the elements and a minimum for the rest, received comparatively little attention, however, because it was inaccessible to most of the electrostatic accelerators then available. Two major surveys attempted to bridge this gap. Nereson and Darden[10] employed neutrons from a reactor, together with a recoil-proton telescope, to complete a 40-element survey using adjacent points[7] at low resolution (~10%) and low precision (quoted as ~10%). Bratenahl, Peterson, and Stoering (BPS)[11] used $H^2(d,n)He^3$ neutrons from a cyclotron at a few isolated points between 7 and 14 MeV for 40 elements, with good resolution and high precision. Less systematic measurements, some isolated and some adjacent, were made up to 8 MeV by a number of other laboratories.[12-16] Isolated measurements[17,18] from 15 to more than 100 MeV were published also, using cyclotrons for neutron sources, so that by 1962 Peterson[19] was able to give a comprehensive theoretical discussion of as many as three diffraction maxima for a single element.

Nevertheless, after 1955 the primary focus of attention at the laboratories which were capable of reaching the energies between 4 and 14 MeV shifted from total cross sections to the more challenging task of measuring the partial cross sections. In 1958 Howerton, after completing a compilation[20] of available data up to 14.5 MeV, pointed out the existence[21] of numerous "areas of ignorance," even in the total cross sections. Howerton defined an area of ignorance as a gap of more than 0.5 MeV between measurements. In a survey which we completed in 1960, we added the restriction that acceptable measurements must have an accuracy better than 5%, and concluded that hydrogen was the only nucleus whose total cross section had no areas of ignorance below 15 MeV (assuming the validity of the effective-range theory). We concluded that a gap of more than 5 MeV existed for about half of the elements. An important factor in reaching this pessimistic conclusion was the systematic disagreement among the three sets of measurements which dominated the second edition of BNL-325[22]; namely, those of Nereson and Darden,[10] Weil and Jones,[16] and BPS.[11] The most dramatic gap in the mass scale occurred for the rare earths, which were largely untouched because samples were scarce and expensive.

By 1960, neutron total cross sections calculated from the optical model were reproducing the measured values with accuracies of the order of 3 to 5%, and thus appeared to be as good as the existing data.[23] Accordingly, we undertook a program designed to eliminate most of the existing "areas of ignorance" in the total cross sections within the 2.5–15-MeV region, largely because a method became feasible using the 2-MV accelerator at Hanford. An additional factor was the large number of separated isotopes which were then becoming available in sufficient quantities for reliable transmission measurements in this range.

II. EXPERIMENTAL METHOD

The use of monoenergetic neutrons to cover the desired energy range requires charged particles up to 9 MeV, and thus was not possible with our accelerator. In addition, a factory-scale measurement with overlapped[7] points becomes excessively tedious with monoenergetic neutrons (an objection which has recently been alleviated by relegating operation of the accelerator and apparatus to a computer). On the other hand, experience from the eV region to 130 MeV had demonstrated the effectiveness of a pulsed source yielding a continuous spectrum of neutron energies, in conjunction with a time-of-flight analyzer,[18,24] so that it was only necessary to adopt a suitable source reaction and pulsing technique.

We selected deuteron bombardment of a thick lithium target for the source of neutrons. Metallic lithium targets are easy to prepare and use. The high thick-target yield is an advantage with an accelerator limited to 2 MV and 20 μA, with no pulse-bunching system. Most important, however, the high Q and multiplicity of reactions, both two- and three-body, together with the thickness of the target insure that even for 1.5-MeV deuterons the neutron spectrum is continuous from less than 2 MeV to more than 15 MeV. A typical time-of-flight spectrum is shown in the lower portion of

Fig. 1, which clearly illustrates the major disadvantage of this source; namely, that although it is indeed continuous, the spectrum is very heavily structured. Indeed, the maximum slope in our case exceeded 20%/channel, and gave rise to our most persistent form of systematic error.

The apparatus has already been described fully in M, so we will limit ourselves to a few additional comments here. The general layout is diagramed in Fig. 2, and the details are listed in Table I. It constituted a closed-geometry narrow-aperture time-of-flight system for good-geometry transmission measurements, using a flight path of 6.14 m. The over-all time resolution averaged 2.2 nsec. The background at a recoil-proton threshold of 1.8 MeV averaged 2.5% of the open beam at 8 MeV and about 5% at 3 MeV, and consisted almost entirely of air-scattered neutrons synchronous with the neutron burst from the target. The only unusual feature of the equipment was a mechanical beam-alignment system which completely eliminated background from spilled beam, and facilitated maintaining the positioning tolerances listed in Table I. However, a major part of the background which triggered the time-of-flight system reached the detector in the form of photons, so that reliable operation of the pulse-shape discriminator was essential to a stable background level.

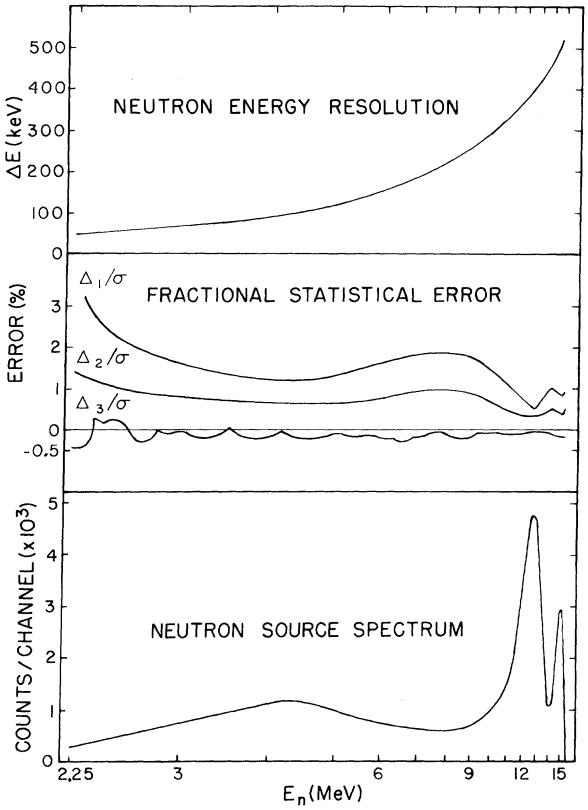

FIG. 1. Time-of-flight spectrum of the Li(d,n) neutron source used in this work, and typical values of the energy resolution and statistical errors of the resulting total cross sections. The spectrum is shown in the bottom third of the figure, with the abscissa marked in units of energy but linear in time of flight (which runs from right to left). The top third displays the composite energy resolution (FWHM). The middle third, which is explained more fully in the text, shows quantities related to the main-diagonal elements of the covariance matrix and the corresponding elements from the two adjacent diagonals. Δ_1 is simply the standard deviation.

Approximately three quarters of our data were taken with the circulating-line vernier chronotron referred to in M. The remainder were taken with a commercial time-to-pulse-height converter and analog-to-digital converter, using a small computer for data storage and control of the equipment. The vernier chronotron was eliminated primarily because its time zero varied with the gross counting rate, and hence it was excessively vulnerable to instabilities in the Van de Graaff accelerator. The computer was much more reliable than our previous storage systems, and facilitated making routine checks on the stability of the complete system by analyzing prominent features of the time-of-flight spectrum.

For both systems the average channel width of the time-of-flight system was calibrated frequently against the crystal-controlled period of the beam-chopping oscillator, using the sharp peaks in the thin-target $Be^9(d,n)B^{10}$ spectrum as markers. Since the range of the time-of-flight system was necessarily slightly less than one full rf period, this comparison had to be made using two bursts per rf cycle. Because of asymmetries in the beam-deflection system, the two half cycles

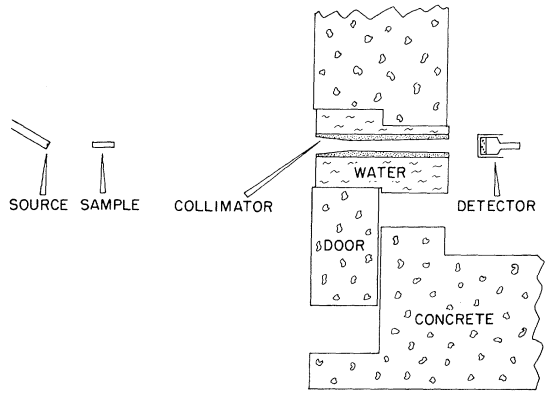

FIG. 2. Diagram of the experimental layout, drawn to scale except for exaggerated diameters of the beam piping and sample. The distance from source to detector is 6.14 m, the shield is 1.2 m thick, and the neutron flight path makes an angle of 25° with the deuteron beam.

were slightly different in length, so a two-step calibration against the full period was essential. The distribution of the differential channel width was measured using uncorrelated *stop* pulses from an independent oscillator, and the resulting nonlinear time scale was reconstructed numerically, using this distribution and the average channel width. The $Be^9(d,n)B^{10}$ spectrum also gave a direct measure of the over-all energy resolution.

The zero point of the time scale varied with counting rate in the vernier chronotron, and even after replacing the chronotron with the time-to-amplitude converter the phase-shifting network used in our time-mark generator was never exactly reproducible in resetting the time zero. Accordingly, the time scale was referred to the centroid of the 12.5-MeV peak in the $Li(d,n)$ spectrum as a secondary standard, and the flight time corre-

TABLE I. Summary of apparatus.

Pulsing system	
Location	Post acceleration
Frequency	3.330 MHz sinusoidal, crystal-controlled
Burst duration	1.5 nsec FWHM
Peak deuteron current	10–20 μA
Bursts per rf cycle	1 or 2
Neutron source	
Reactions	$Li(d,n)x$
Target	0.5-mm lithium metal
Beam spot	3 mm diam, position stable ±0.5 mm
Deuteron energy	1.5–2.0 MeV
Angle of emergence	25°
Detected counts/burst	$\lesssim 10^{-4}$
Samples	
Shape	Right circular (or square) cylinder
Diameter	0.95–2.54 cm
Distance from source	26–105 cm (center of sample)
Can wall thickness (when required)	Stainless steel (<10% of radius), or 0.05-mm brass foil
Shadow bar	30-cm steel, tapered to solid angle of front of sample
Sample-support frame	Perforated stainless steel, on stranded stainless-steel wires
Repositioning accuracy	±0.2 mm
Detector	
Distance from source	614 cm
Scintillator	NE-213 liquid, glass cell
Cell dimensions	13 cm diam × 5 cm deep
Collimator	Double cone (Langsdorf), paraffin-soaked boric acid
Working aperture	9 cm
Photomultiplier	58AVP
Instrumental time resolution	1.3 nsec FWHM
Flight-path resolution	1.6% of E_n
Threshold	1.8 MeV
Shielding	1.2-m concrete or water + 0.6-cm Pb
Pulse-shape discriminator	"Space-charge" type
Thermal insulation	5-cm foamed-polystyrene box
Typical counting rate	250/sec (sum for all energies)
Beam monitor	
Type	Integrated deuteron charge to target
Secondary-electron suppression	Baffled Faraday cup at ground potential
Integrator	Current-to-frequency converter + scaler
Dead-time correction	Integrator gated by analyzer live time
Stability of n/d ratio	Maintained by mechanical scraping of lithium *in situ*, as required

sponding to this centroid was determined in a final calibration measurement by switching off the pulse-shape discriminator and measuring the time interval between the centroid and the synchronous γ-ray flash from the target.[25]

The dimensions shown in Table I indicate that our samples were located much closer to the neutron source than the traditional position[26] halfway between the source and the detector. This position was dictated by inscattering-minimization calculations, which we have already discussed in an Appendix to M. The crucial point in these calculations is that the diameter of the sample must be varied along with its position in such a way that the sample just barely shadows the detector completely from the source. For our detector and source dimensions, the minimum inscattering error occurs about 95 cm from the source, where it is about a factor of 2 smaller than at the traditional 307-cm position. More importantly, even at 26 cm from the source the inscattering is 20% less than at the traditional position.

Following the treatment originated by McMillan and Sewell,[11,27] the relative inscattering error $\Delta\sigma_T/\sigma_T$ is proportional to a geometrical inscattering index G, defined by

$$G = \Omega_{12}\Omega_{23}/\Omega_{13}, \tag{1}$$

in which

Ω_{12} = solid angle subtended by sample at source,

Ω_{23} = solid angle subtended by detector at sample,

Ω_{13} = solid angle subtended by detector at source.

Table II lists values of G for some representative previous measurements, together with the corresponding values for two of our own sample positions. At the time we undertook this work, our value of G was approximately a factor of 2 smaller than the smallest previously reported[11] in this energy range.

The importance of minimizing inscattering arises, as is well known, from the very strong forward peak in the elastic scattering from heavy nuclei above 5 MeV. For our value of $G \approx 0.8\times10^{-3}$, the maximum error was approximately 0.3% for plutonium at 15 MeV, and hence we found it unnecessary to apply any inscattering corrections. We implied in M that this was the result of moving the sample closer to the source, but this is misleading, since only a factor of 2 can be attributed to this step. The basic reason for the small inscattering is the narrow-beam geometry forced on us by the small detector diameter and long flight path required to achieve adequate energy resolution. The penalty paid for these advantages is the very long running time required to achieve a reasonable precision; namely, about 8 h for a run on a single sample. The low counting rate has the compensating advantage, however, of completely eliminating dead-time distortion[28] of the time-of-flight spectrum.

A much more valuable consequence of moving the sample closer to the source lies in the reduction in the required area of the sample. In our case, the required area at 26 cm was a factor of 37 smaller than that needed at 307 cm. This made it feasible to perform measurements on samples which were too expensive or too scarce to do at the traditional position. For example, by using a diameter of 0.96 cm at 26 cm from the source, we were able to make a transmission measurement on a 14-g sample of $Pm^{147}{}_2O_3$, which at the time contained most of the world's supply of chemically separated promethium. On this scale, in 1965 there were sufficient quantities in the United States Atomic Energy Commission Cross-Section

TABLE II. Geometrical inscattering index G for representative measurements.

Authors	Reference [a]	Year	Detector	Sample area (cm^2)	Distance from source to sample (cm)	Distance from source to detector (cm)	Inscattering index (10^{-3})
Miller *et al.*	6	1952	H ionization chamber	15.1	18	36	187
Weil and Jones	16	1958	Thin plastic scintillator	5.1	15	30	90
Carlson and Barschall	53	1967	Stilbene	5.1	13	40	66
Fowler	83	1960	Propane prop. counter	6.4	38	76	18
Fasoli *et al.*	59	1966		7.1	30	300	9.7
Nereson and Darden	10	1954	Radiator + ion. chamber	2.8	107	130	9.6
Coon, Graves, and Barschall	8	1952	Stilbene	5.1	82	165	3.0
Bratenahl, Peterson, and Stoering	11	1958	Plastic scintillator	5.1	152	228	2.0
Galloway and Shrader	28	1966	Liquid scintillator	20.5	300	590	0.94
Present work	1	1970	Liquid scintillator	0.71	26	614	1.14
				5.1	96	614	0.78

[a] The references are given in this paper as footnotes to the text.

Isotope Pool to make such measurements on more than 60 separated isotopes.

Our data were taken in cycles of *blank*, *sample*, and *background*, using the monitor-count ("live" deuteron charge) ratios 1.0:4.0:0.4, respectively. The order of the *blank* and *sample* was reversed in successive cycles, in order to average out slow drifts, and a normal run comprised three cycles plus the standard set of calibrations discussed above. These particular counting-time ratios resulted from optimization calculations performed simultaneously on this distribution and the thickness of the sample, following the same general approach as Rose and Shapiro[29] or BPS.[11] The corresponding sample thickness for our background levels was approximately 1.8 mean free paths (mfp), which gives a transmission of approximately 17%. However, the same calculations show that the statistical precision attainable in a given counting time is only slightly degraded[30] for thicknesses as small as 1.0 mfp. Insofar as possible, we used 1.8 mfp as a maximum thickness rather than an average thickness, because at higher transmissions the measurement is less sensitive to drifts in background rate. However, we performed test measurements[31] on lead up to 2.6 mfp, at which thickness the transmitted counting rate of the foreground below 3 MeV was equal to the background, without detecting any change in the measured cross sections.

In addition to the latter test, we also made test measurements[31] to ensure that the measured background rate was insensitive to the diameter of the shadow bar, that absorption of background by a can surrounding a sample did not produce a measurable error, and that there was no detectable inscattering from the can. Since β decay of Li^8 following the $Li^7(d,p)Li^8$ reaction produces energetic bremsstrahlung which is asynchronous with the pulsed beam, we performed a differential-absorption test with steel and polyethylene shadow bars, but failed to detect any measurable asynchronous photon contamination in the beam. The synchronous γ flash, which was barely detectable through the pulse-shape discriminator, lay outside the working time-of-flight range, and so could not give rise to any error.

A straightforward calculation[32] shows that transmission measurements are not very sensitive to internal voids in the sample, provided that the average density of the sample is used in calculating the areal density (the average was from 0.5 to 2% lower than the "handbook" density in most of our cast samples), and provided that the aggregate length of a chain of voids which is parallel to the beam is not a large fraction of the thickness of the sample. We radiographed all of our cast samples and found numerous small voids, especially in the rare earths, but in no case did their calculated effect prove significant.

When the sample contains other nuclei than those of interest, the cross section of the contaminants can either be subtracted algebraically or else eliminated by using a *blank* which duplicates the areal density of the contaminants. The latter procedure is preferable, because it insures that the energy scale and resolution of the correction are exactly matched to those of the measurement on the composite sample. In addition to providing dummy cans for 35 samples, we have used both procedures in dealing with internal contaminants.

Table III lists all of the samples which were deliberately composite or involved more than minor contamination.[33] Since at the completion of our work we had measured the cross sections of all possible contaminants, it was straightforward to correct for the minor contaminants algebraically, using the chemical analyses provided by the manufacturers. Similarly, for well-defined compounds it was straightforward to prepare matched physical blanks for many of the composite samples (and to correct for any minor mismatch algebraically). Unfortunately, the latter category included only CF_4, CCl_4, the ingenious 5-aminotetrazole ($NH_2 \cdot N_4CH$) sample devised by Johnson, Petree, and Adair,[34] and the B_2O_3 glass which we prepared by roasting chemically pure boric acid.

Our results on both hydrogen and deuterium exhibit traces of residual structure from the cross section of carbon, although the average number of nuclei per cm^2 in the blanks was nominally matched to the samples[33] within a few parts per thousand. In addition, the cross sections for both elements fall several percent below most other measurements[3] for energies less than about 4 MeV. We shall discuss the hydrogen results further in Sec. VI. As noted in Table III, we did not have an analysis performed on the batch of polyethylene from which our hydrogen sample was machined. However, the absence of spurious structure near 8 MeV in our results for both isotopes argues against a mismatch of the blank, either because the sample was not exactly CH_2 or CD_2 or because of density gradients[33] in the blank. The thickness of both samples was about 2.5 mfp in the peaks of the carbon cross section near 3 MeV. Although the tests on lead described above gave the same results within about 1% up to 2.6 mfp, and our measurements on beryllium gave apparently correct results at 3.1 mfp in the 2.75-MeV peak, the internal evidence for both hydrogen and deuterium suggests an instability in background level which was equivalent, in the region below 3 MeV, to a systematic uncertainty of about 3% in the resulting

cross sections.

Our samples of Ca^{42}, Ca^{44}, and Pm^{147} were all in the form of oxides, for which we made the correction algebraically.[35] The latter two give no evidence of any resulting difficulty, but our result on Ca^{42} exhibits structure which is strongly suggestive of contamination by the cross sections of carbon and/or oxygen, and the average cross section is nearly 20% higher than would be inferred from the systematic behavior of neighboring elements. Since there is also some uncertainty regarding the completeness of conversion of this sample from the carbonate to the oxide, we have been forced to discard our measurements on this sample.

Table III lists six cases[36] of accidental contamination for which we made an empirical correction. We discovered all of these by the appearance of a flat-topped peak between 3.2 and 3.7 MeV, which is characteristic of oxygen. For each case we used the cross section of a neighboring element for a template (to supply the approximate shape of the expected cross section), and deduced the magnitude of the contamination from the amplitude of the characteristic structure. For strontium and tellurium the contamination proved to be a fixed quantity of oxygen incorporated at the outset, which amounted to 23.5 at% in the case of tellurium. In W^{182} and W^{186}, comparison of the cross sections at high and low energies with those of natural W showed that the isotopically enriched metal powders had adsorbed water vapor rather than oxygen alone. Similarly, the powdered samples of arsenic and rhenium adsorbed water vapor continuously in spite of the press-fitted caps of the cans, as evidenced both by the growing oxygen structure in the apparent cross sections and by the gain in weight of the samples. In all cases except arsenic, the correction deduced from the amplitude of the oxygen structure also brought the average cross section into close agreement with nuclear systematics, so that we could make the correction with considerable confidence. The situation in arsenic is ambiguous, and the correction which we have adopted yields a consistent average cross section but leaves a trace of possibly spurious structure.

III. REDUCTION OF DATA

Our procedure for the basic data reduction has remained substantially as described in M, except for the addition of a calculation of the covariance matrix throughout the reduction process. Since the unstable time zero discussed in Sec. II produced systematic errors in the neighborhood of the 12.5-MeV peak in the spectrum which were an order of magnitude larger than the counting statistics,[37] we were forced to refer each time spectrum to the centroid of that peak as a secondary zero point, as we have described above. Since the *blank* spectrum was displaced from the *sample* spectrum by a nonintegral number of channels, it was necessary to interpolate between channels in the *blank* spectrum, which we accomplished by fitting a second-degree polynomial to the pooled data from three adjacent channels in the *blank* spectra of all of the cycles devoted to a given sample in a given run. This least-squares fitting procedure furnished a χ^2 test for identity of the shapes of the individual spectra, which normally indicated a scatter of 1.0 to 1.3 times counting statistics. The transmission ratios deduced from each *sample* spectrum were required to show a comparable consistency. For larger discrepancies (indicating the presence of systematic drift) we processed the cycles separately and attempted to combine them later as if they were independent runs.

As we pointed out in M, the centroid of the highest peak in the *sample* spectrum is systematically displaced if the cross section has a nonzero slope at that energy. This leads to an error in the resulting cross section which is proportional to the relative slope $(1/S)(dS/dE)$, where S is the intensity of the source spectrum. In our original data-reduction scheme most of this error was removed by iteratively adjusting the shift in time zero so as to minimize fine structure above 11 MeV. After the publication of M we discover that some residual structure remained, with an amplitude approximately proportional to the cross-section slope. Accordingly, we applied an additional correction empirically,[38] using a more sophisticated covariant fit to the cross section and minimizing the resulting generalized χ^2. In almost all cases, this reduced the amplitude of the spurious structure below 1.5%. Coincidentally, the relative slope of the spectrum is nearly zero at 14.1 MeV, so that comparison to the many 14-MeV measurements in the literature is almost invariant under this empirical correction procedure.

The relative-slope correction is valid only if a pure shift in time zero is present. Under unstable operating conditions, however, the continual wandering of the time zero during the accumulation of a spectrum produced resolution broadening instead, for which we have found no satisfactory *a posteriori* correction. The worst examples of this in our final results, with amplitudes $\geq 2\%$, include hydrogen, deuterium, palladium, and about half of the rare earths.

The interpolation procedure which we have used makes it possible to recover usable data from the 11–15-MeV region, but only at the expense of in-

troducing covariance into the resulting cross sections. The correlation arises because the counts in a given channel of a *blank* spectrum contribute to the transmission ratio attributed to three successive channels. The result is that the symmetric covariance matrix, whose elements are defined by

$$V_{ij} = \langle d\sigma_i d\sigma_j \rangle, \quad (2)$$

has nonzero elements for $i-2 \leq j \leq i+2$. Clearly, V_{ii} is the variance according to the usual definition. Typical ratios V_{ij}/V_{ii} are of the order of 0.25 for $j = i \pm 1$ and -0.005 for $j = i \pm 2$. When separate runs with slightly different energy scales are averaged together, the correlation is extended weakly to $j = i \pm 3$.

The covariance has one important effect on the results of these measurements, aside from the nuisance of computing and storing the elements V_{ij}. Since the correlation between nearest-neighbor points is positive, a point which is accidentally high tends to be flanked by points which are also high. Because the resolution function is only three or four channels wide at half height, the effect is to simulate a weak peak where none exists. The effect is enhanced somewhat because the next-nearest neighbors tend to be lower than normal because of their negative correlation. This phenomenon can be disastrous to the intuition in examining our results for evidence of small fluctuations in the cross section, but it can be dealt with unambiguously by a least-structure calculation,[39] which substitutes rigorous calculation for the intuition. We have written[40] a least-structure computer program, generalized to accept covariant input data, but have not yet applied it to our measured cross sections. Accordingly, we will give only qualitative conclusions regarding fluctuations in our discussion in Sec. VII below.

IV. RESOLUTION AND ERRORS

The characteristics of a typical measurement are summarized in Fig. 1, which preserves the original time-of-flight scale of the abscissa, and hence has a nonlinear energy scale. Three components contributed directly to the time resolution of the system; namely, burst duration [~1.5 nsec full width at half maximum (FWHM)], instrumental resolution (~1.3 nsec), and channel width (~0.75 nsec), with a composite resolution of about 2.2 nsec. The 5-cm depth of the detector accounted for a constant 1.6% spread in energy, rather than a constant time resolution, and was the dominant component of the energy resolution at the low-energy end of the scale. The other three components

TABLE III. Composite and contaminated samples.

Element	Compound	Form	Blank	Principal contaminant Element	Principal contaminant at.%	Footnotes
H	CH_2	Cast	C			a
D	CD_2	Cast	$C + CH_2$	CH_2	3.5	
N	$NH_2 \cdot N_4CH + \frac{1}{2}C$	Powder	CH_2			
O	B_2O_3	Glass	B			
F	CF_4	Cast	C			a
Cl	CCl_4	Liquid	C			
Ca^{42}	CaO	Powder		C?	?	a
Ca^{42}	CaO	Powder				a
Mn		Powder		O	1.8	
As		Powder		H_2O	6.0	b, c
Sr		Cast		O	13.4	b
Te		Powder		O	23.5	b
Pm^{147}	Pm_2O_3	Powder		Sm	2.5	d
Eu		Cast		O	2.9	
Tb		Cast		O	2.5	
Yb		Cast	CaO	Ca, O	2.5, 1.6	
Lu		Cast		O	3.3	
Hf		Cast	Zr	Zr	6.2	
W^{182}		Powder		H_2O	12.2	b
W^{186}		Powder		H_2O	15.2	b
Re		Powder		H_2O	4.5	b, c

[a] Composition assumed without actual analysis.
[b] Concentration of contaminant estimated from oxygen structure.
[c] Increased with time, final value given.
[d] Radioactive daughter, concentration at middle of measurement.

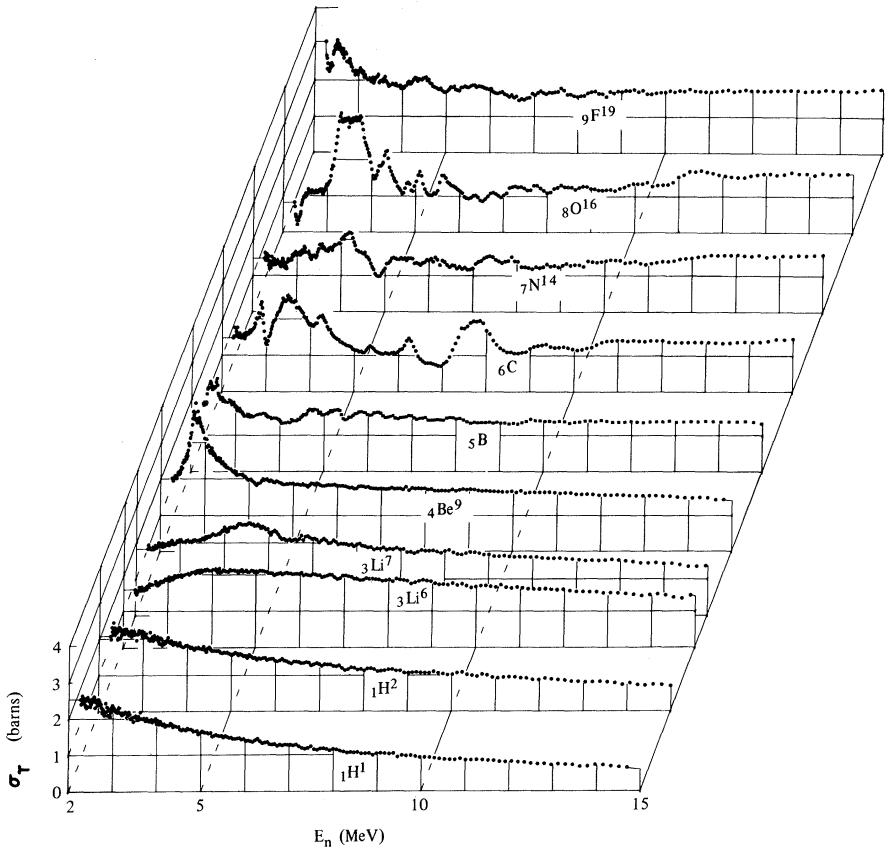

FIG. 3. Three-dimensional plot of the total cross sections measured in this work, for $Z=1$ to 9. Nominally, the third axis is linear in atomic number, but for greater clarity the lithium isotopes have been displaced equal distances on each side of $Z=3$ and deuterium has been moved into the vacant spot at $Z=2$, since we did not make measurements on helium. The statistical standard deviation is smaller than the plotted symbols.

dominated at the high-energy end. The upper section of Fig. 1 illustrates the over-all resolution. With the channel-width set near 0.75 nsec there were approximately 240 channels between 2.25 and 15 MeV, and the resolution width ranged between 3 and 4 channels, thus fulfilling our requirement for "overlapped" points.[7]

The counting statistics for a typical case are shown in the middle portion of Fig. 1, from which it is clear that they are dominated by the neutron spectrum. The quantities actually plotted are $\pm\Delta_{ij}/\sigma_i$, where

$$(\Delta_{ij})^2 = V_{i,i+j-1}\,, \tag{3}$$

and the sign is chosen to preserve the sign of $V_{i,i+j-1}$. The index i is suppressed in Fig. 1, and the Δ's are plotted as continuous functions $\Delta_j(E)$. Thus, Δ_1/σ is simply the relative standard deviation, and Δ_2/σ is an equivalent presentation of the correlation coefficient between points i and $i\pm1$. The second off-diagonal element was always negative after the initial data-reduction step, but the scalloped appearance of Δ_3/σ in Fig. 1 shows the effect of combining two independent runs which had slightly different channel widths. The essential points are that the standard deviations typically ranged between 1 and 3%, and that each point was fairly strongly positively correlated with its nearest neighbors and weakly negatively correlated with its next-nearest neighbors. Averaging groups of points together improves the precision more slowly than it would if the points were uncorrelated. This compensates for the improved precision achieved in the initial pooling of the *blank* spectra.

We have already noted that each step in the combination of transmission cycles into runs, and of independent runs into final results, gave evidence of discrepancies which typically ranged between 1.0 and 1.3 times statistics. The excess is, of course, caused by the minor systematic errors which provided the motivation for breaking the

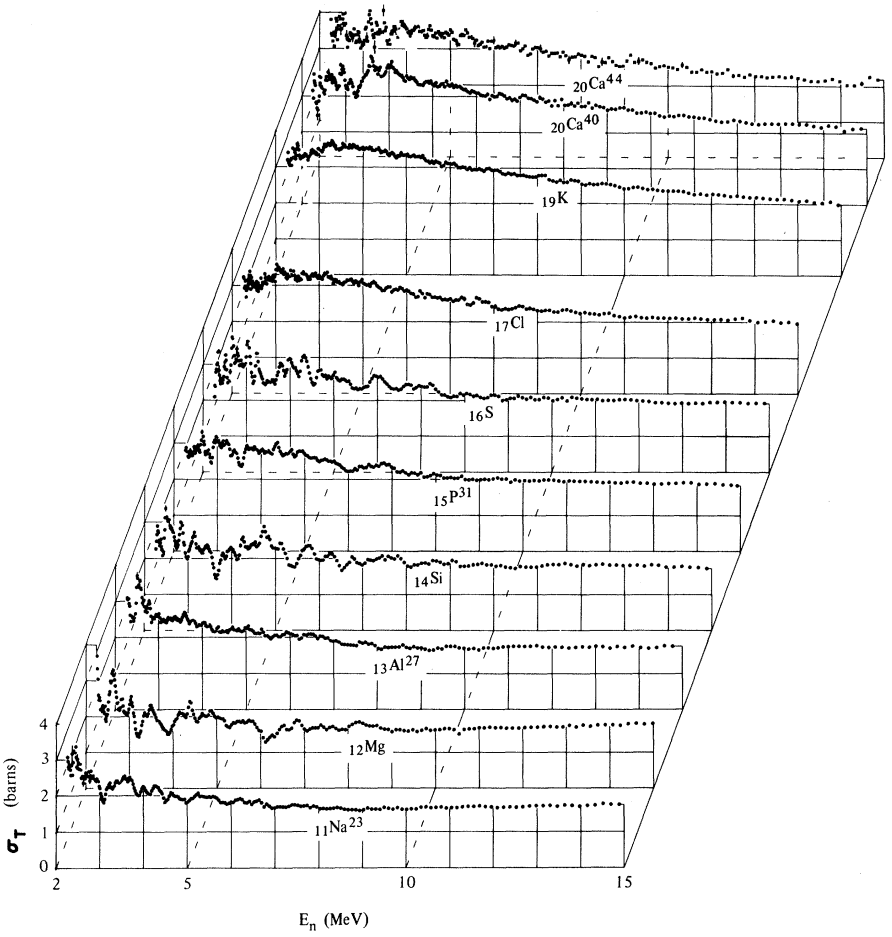

FIG. 4. Three-dimensional plot of the present results, for $Z = 11$ to 20, drawn to the same scale as Fig. 3. Natural calcium and Ca^{42} have been omitted, and the standard deviations (shown on every 10th point) are large enough to be visible for Ca^{44}.

measurement up into short cycles in the first place. The dominant causes are instabilities in zero time, background, detector threshold, and occasionally channel-width distribution. Since the typical final result is the average of only six data cycles and two calibration cycles, some of these uncertainties remain in the final result, with magnitudes which usually depend on the energy range involved. However, the entire range is affected equally by errors in sample thickness, for which we estimate uncertainties of 0.5% in most cases, and 1% for the alkali metals and other highly reactive metals.

We have already discussed at some length the spurious structure in the cross sections above 11 MeV. The amplitude of the residual structure is typically of the order of 1.5%, and is thus larger than the statistical errors. The effect of drifts in background and detector threshold, on the other hand, is largely concentrated below 3 MeV, in the long low-intensity tail of the neutron spectrum, where the errors due to counting statistics are also greatest. Thus, we can give a composite estimate of systematic errors below 11 MeV as one third of the quoted standard deviation of a single point. We emphasize that our quoted statistical errors contain only the random error with no allowance for this possible systematic error. Additional systematic errors apply to particular cases. The measurements on hydrogen and deuterium show traces of mismatch between the sample and its carbon blank (probably induced by instabilities in background rather than physical mismatch). Accordingly, we estimate the systematic errors for these samples (below 11 MeV) at one half of the random errors. In the case of samples contaminated by oxygen or water, for which we had to estimate the contamination from the amplitude of the characteristic structure in the cross section of oxygen, we would be even more pessimistic, but

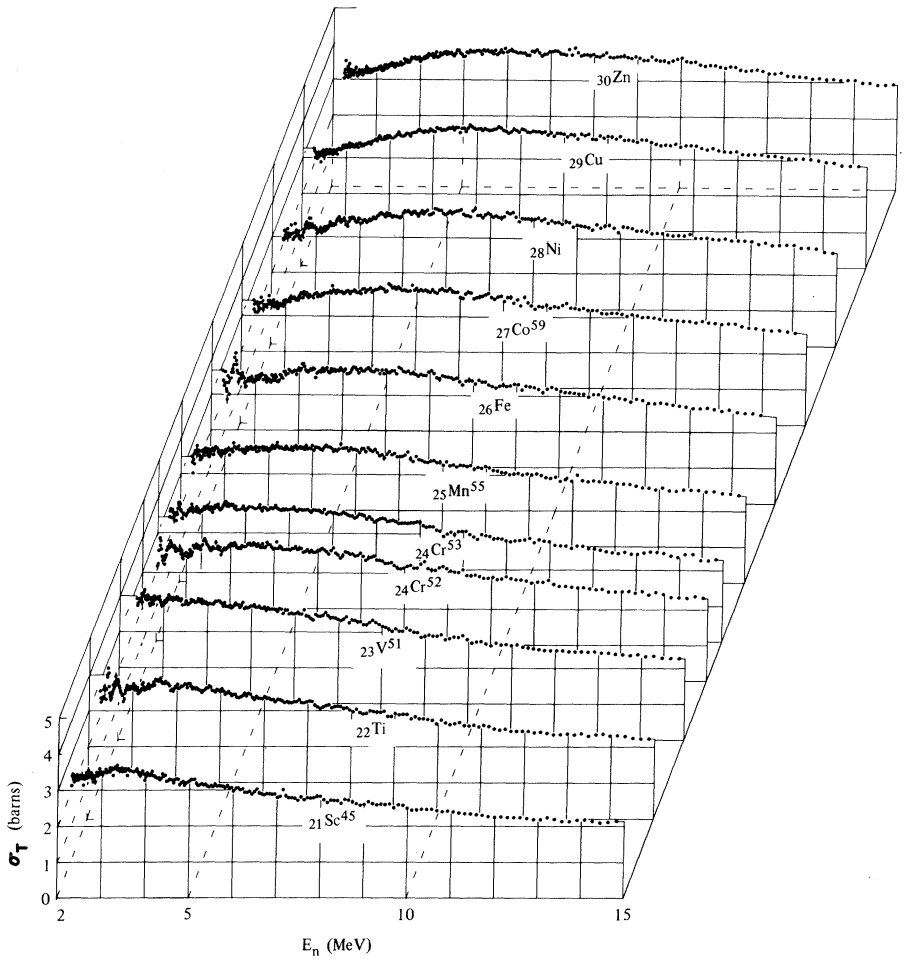

FIG. 5. Three-dimensional plot of the present results, for $Z=21$ to 30, drawn to the same scale as Fig. 3. The standard deviations are smaller than the plotted symbols.

the internal evidence from the systematic behavior of the cross sections of neighboring elements suggests that this is unnecessary. Accordingly, except for arsenic we do not believe the systematic uncertainty in the contaminated samples is worse than one half of the random error of an individual point. There are a few additional cases of apparently larger errors, which we will discuss below.

V. RESULTS

The complete set of results covers:
(1) all of the stable natural elements except the noble gases;
(2) the stable isotopes D^2, Li^6, Li^7, Ca^{44}, Cr^{53}, W^{182}, W^{186}, and radiogenic lead (mostly Pb^{206});
(3) by combining the second group with the measurements on the natural elements, the stable isotopes Ca^{40}, Cr^{52}, W^{184} (68%), and Pb^{208} (70%); and
(4) the radioactive isotopes Tc^{99}, Pm^{147}, Th^{232}, U^{233}, U^{235}, U^{238}, and Pu^{239} (90%).

The maximum energy range is $2.25 < E_n < 15$ MeV (the highest minimum energy is 2.5 MeV), and the complete set totals approximately 24 000 points, including the algebraically enriched isotopes.

Most of these results are plotted in Figs. 3–11, which form a single three-dimensional plot of the Barschall[5] type, broken into nine sections. The third axis is linear in atomic number (rather than mass number or nuclear radius), except for the separated isotopes and the heaviest elements, which have been displaced for easier plotting. The standard deviations are shown on every 10th point, unless they are smaller than the plotted symbol. We have omitted plots of Ca^{42} and of the natural elements for which we show results for several isotopes instead. Arbitrarily, we have labeled elements which are more than 99.0% monoisotopic with the mass number of the dominant isotope. Tabulations of the results are available

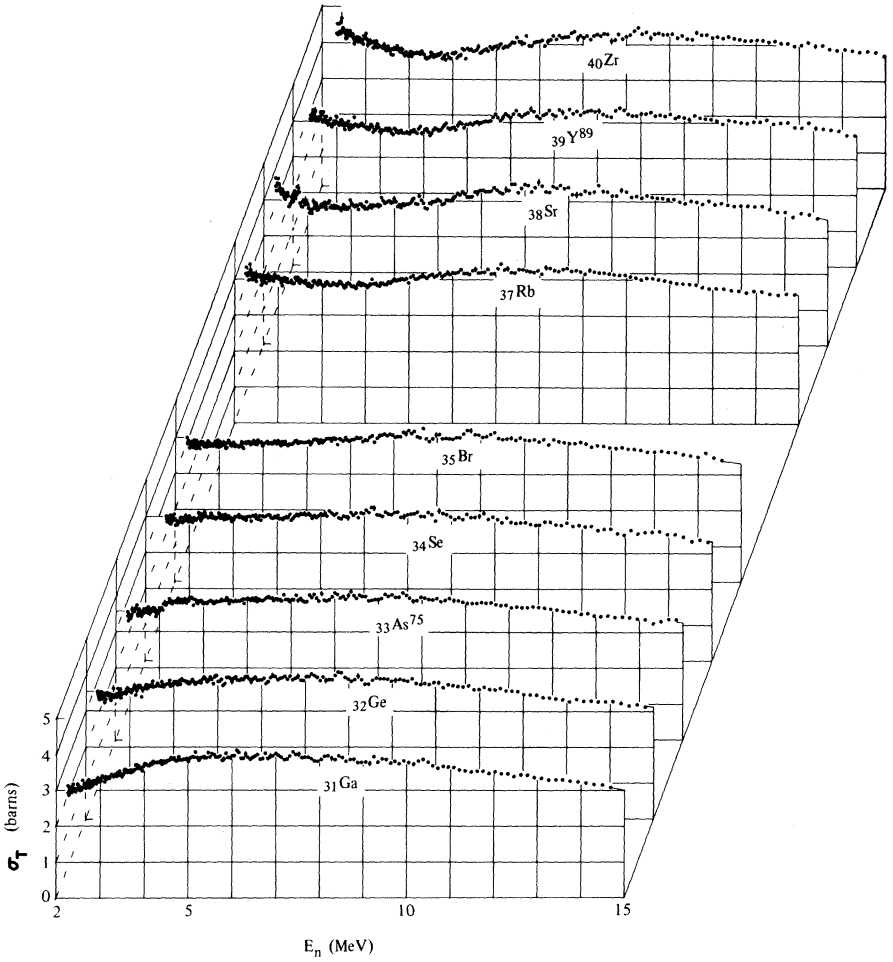

FIG. 6. Three-dimensional plot of the present results, for $Z = 31$ to 40, drawn to the same scale as Fig. 3. The standard deviations are shown on every 10th point.

through the National Neutron Cross Section Center at Brookhaven National Laboratory and the Centre ENEA de Compilation de Données Neutroniques at Saclay.[41]

Figures 3–11 are designed to emphasize the striking regularities in the neutron total cross sections, first pointed out by Barschall,[5] which have been the inspiration for so much nuclear model building. In our concluding section we will take advantage of the well-known regularities to pinpoint some irregularities. For the moment, however, let us exploit the regularities themselves.

It is convenient to define the geometric cross section,[9]

$$\sigma_g = \pi(r + \lambda\!\!\!^{-})^2 , \tag{4}$$

in which r is a suitably chosen effective radius of the nucleus and $\lambda\!\!\!^{-}$ is the wavelength of the neutron in the c.m. system divided by 2π. The geometric cross section approximates the gross behavior of the neutron-nucleus interaction at energies above the resonance region. Peterson[19] has pointed out that the nonelastic plus compound-elastic cross section is roughly equal to σ_g, while the shape-elastic cross section oscillates around σ_g as a result of interference between the neutron wave which passes through the nucleus and the wave which sweeps around the outside. Thus, the total cross section as a function of mass number should be approximately equal to $2\sigma_g$ when averaged over the diffraction oscillations. Our energy range is too small to cover a full cycle of the diffraction structure, but in most cases includes one maximum and one minimum. From these extrema, we find that an effective nuclear radius of

$$r = 1.28A^{1/3} + 0.53 \text{ fm} \tag{5}$$

best describes all of our data for nuclei heavier than chlorine, yielding a somewhat smaller con-

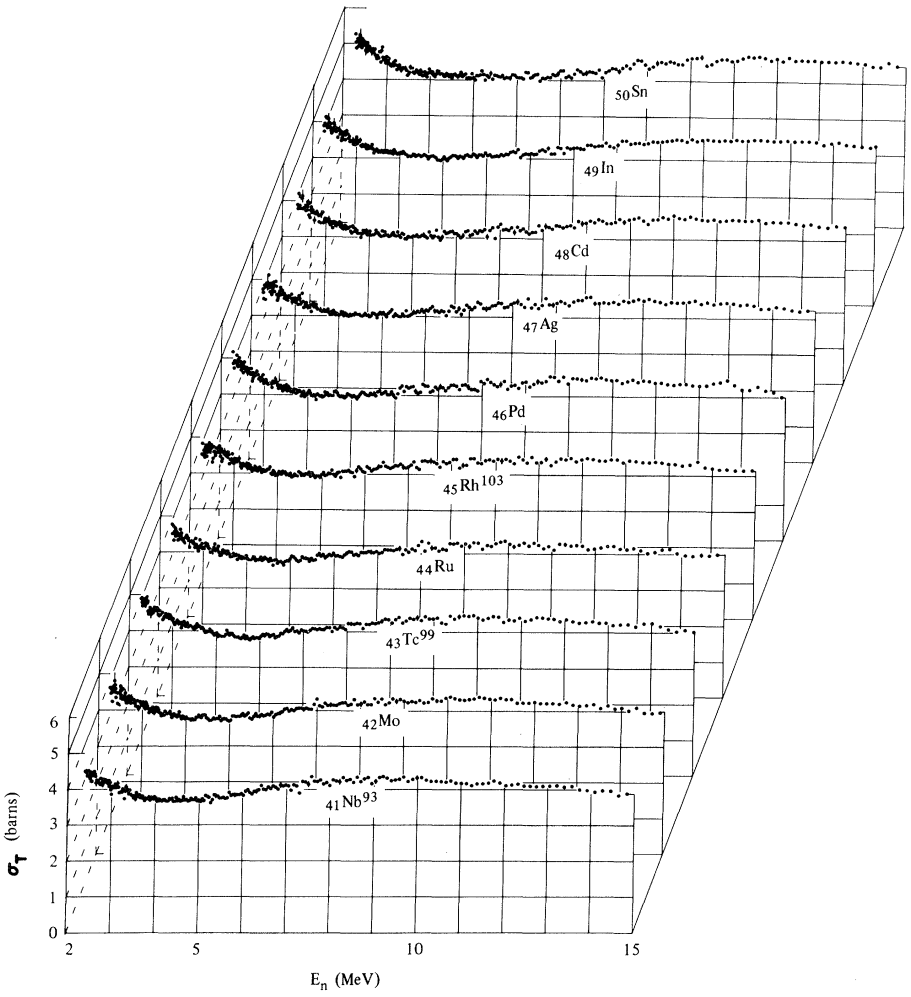

FIG. 7. Three-dimensional plot of the present results, for $Z=41$ to 50, drawn to the same scale as Fig. 3. The standard deviations are shown on every 10th point.

stant term than originally proposed by Drell.[42]

We have calculated values for the ratio $\Sigma=\sigma_T/\sigma_g$, averaged on a linear energy scale over our range of energies. Omitting Ca^{42}, we find that the average for all of our results with $Z>17$ is $\bar{\Sigma}=1.99$, and that all but two elements in this range lie within ±5% of $\bar{\Sigma}=2.0$. A plot of $\bar{\Sigma}$ vs $A^{1/3}$ is shown in Fig. 12. Most of the residual oscillations around 2.0 are due to the fact that our average does not span a complete cycle of the diffraction pattern and ignores the damping of the oscillations with increasing energy. Figure 12 is the basis for our earlier remarks regarding systematics, for it suggests a regularity for the elements heavier than calcium which is so sweeping that it can be used to diagnose systematic errors of the order of 2%. Thus, we suspect possible errors in sample, thickness[43] or purity for Ru, Yb, and Os (or alternatively, seek to construct models which explain their irregular behavior), but are reassured about the corrections for contamination applied to As, Sr, and Te. The value of $\bar{\Sigma}$ for scandium reinforces our confidence in the handling of the $Ca^{44}O$ sample, as well as pointing up a discontinuity upon crossing the closed proton shell at $Z=20$. The anomalous behavior of $\bar{\Sigma}$ for Ca^{42}, as illustrated in Fig. 12, was the principal reason for our discarding the measurements on this sample, as noted in Sec. II.

This rather general internal consistency from element to element, together with the fact that the data were taken under uniform conditions throughout the Periodic Table, suggests the use of our results as a reference surface for interrelating the measurements of others, even if the latter do not actually overlap each other. Our data form a homogeneous set, in which any unknown systematic errors that are intrinsic to the method and ap-

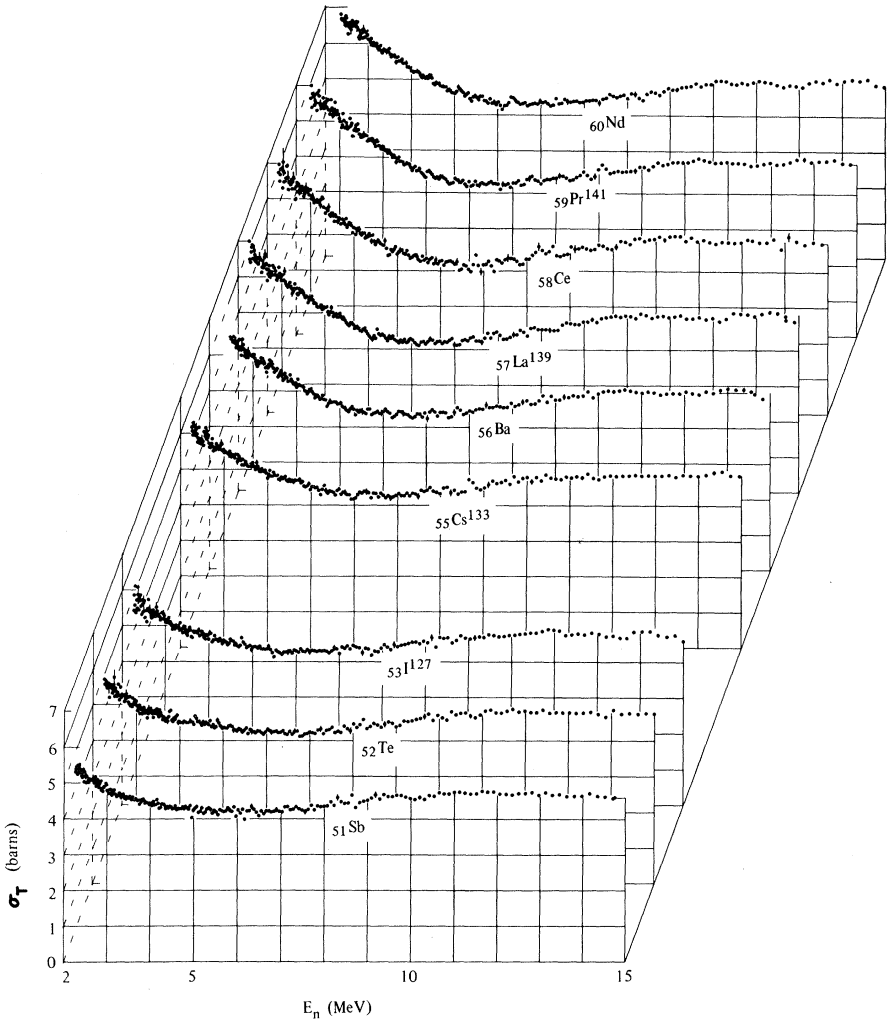

FIG. 8. Three-dimensional plot of the present results, for $Z=51$ to 60, drawn to the same scale as Fig. 3. The standard deviations are shown on every 10th point.

paratus should vary smoothly with mass and energy. In the next section, in the course of a detailed discussion of comparisons with other work, we shall attempt to show that a number of recent higher-precision better-resolution measurements lie very nearly on the same surface as ours, and thus tend to validate portions of that surface. We then argue, from the internal consistency of our work, that the entire surface is nearly free from unknown systematic errors.

VI. SYSTEMATIC COMPARISON WITH OTHER WORK

We have conducted detailed comparisons between our work and approximately 150 previously published sets of measurements, totaling more than 18 000 points. The exact number of references is somewhat ambiguous, because we have arbitrarily grouped some closely related papers, and there is further ambiguity because some measurements have never been published in an archival journal and others have appeared more than once.

Our basic source, both as a bibliography and for the data themselves, has been the compilation by Howerton[20] at the Livermore site of the Lawrence Radiation Laboratory. We chose this source originally because it was the first *tabulation* to become widely available, and subsequently because it was the first compilation to be made available to us on magnetic tape. In addition to the magnetic tapes, we located further references from the original edition of UCRL-5226,[20] from the various editions of BNL-325,[22] from CINDA,[44] and from our own literature search. Generally speaking, none of these sources completely duplicates the others,

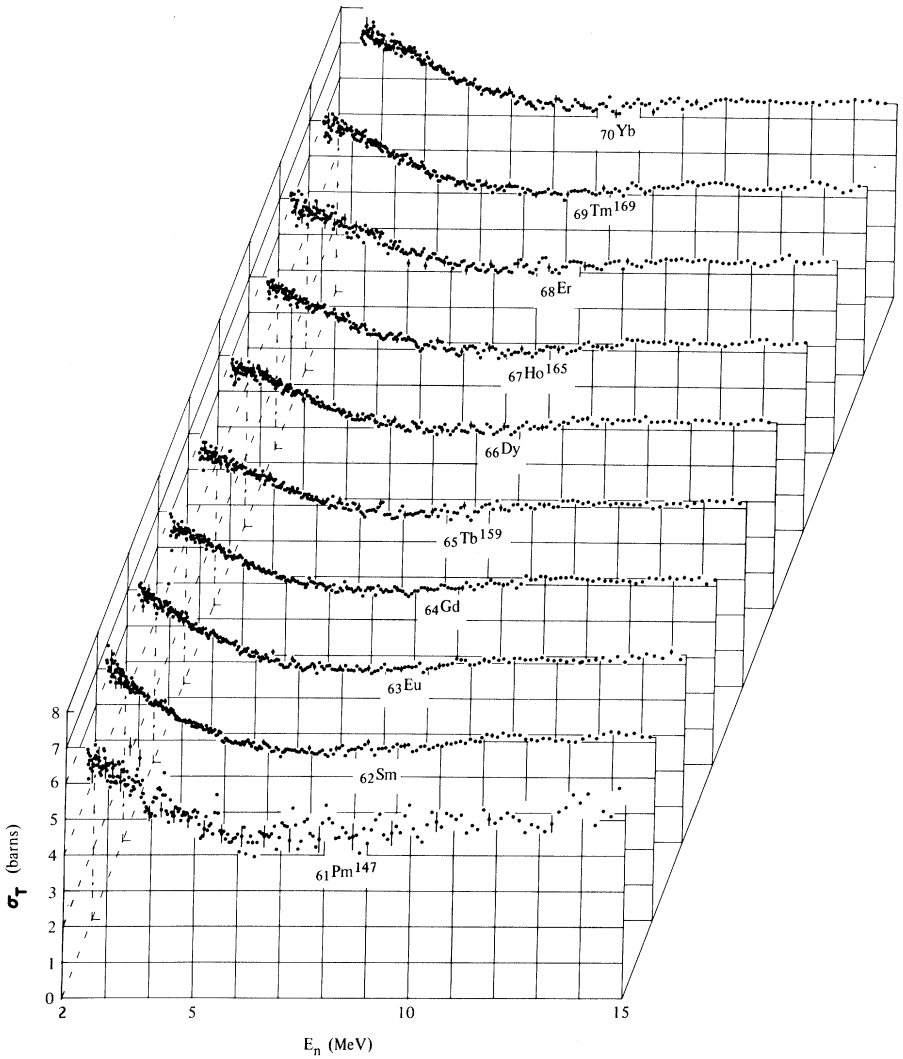

FIG. 9. Three-dimensional plot of the present results, for $Z = 61$ to 70, drawn to the same scale as Fig. 3. The standard deviations are shown on every 10th point. Note the large random errors for Pm^{147}, for which the sample was very thin, and the spurious fine structure above 11 MeV (discussed in the text) for several of the heavier elements in this plot.

and we found errors of inclusion, exclusion, and transcription in all of them.

For the data themselves we have supplemented the Livermore tapes with tabulations from the Brookhaven center, the original publications, or the authors. In a few instances we have resorted to enlargements of published graphs. For the most part, however, we have relied on secondary sources, both because of the convenience to ourselves and to the original authors, and because we hoped that direct communication *via* magnetic tape would eliminate further errors in transcribing the data. After the initial round of comparisons with our reference surface, many anomalous points became conspicuous. These were traced back to the original source, or cross-checked between compilations, and corrected wherever errors could be established. Many records lacked standard deviations, because they were either omitted by the authors or dropped during compilation. We have restored those which were dropped and in a few cases have approximated missing standard deviations from the apparent scatter of the points.

Although we have not used a fixed cutoff date, we have few entries that were published after June, 1968. There are a few sets of data which we have not acquired, of which the Karlsruhe work[45] is much the most conspicuous. We have checked only a small fraction of the entries for errors in transcribing the data.

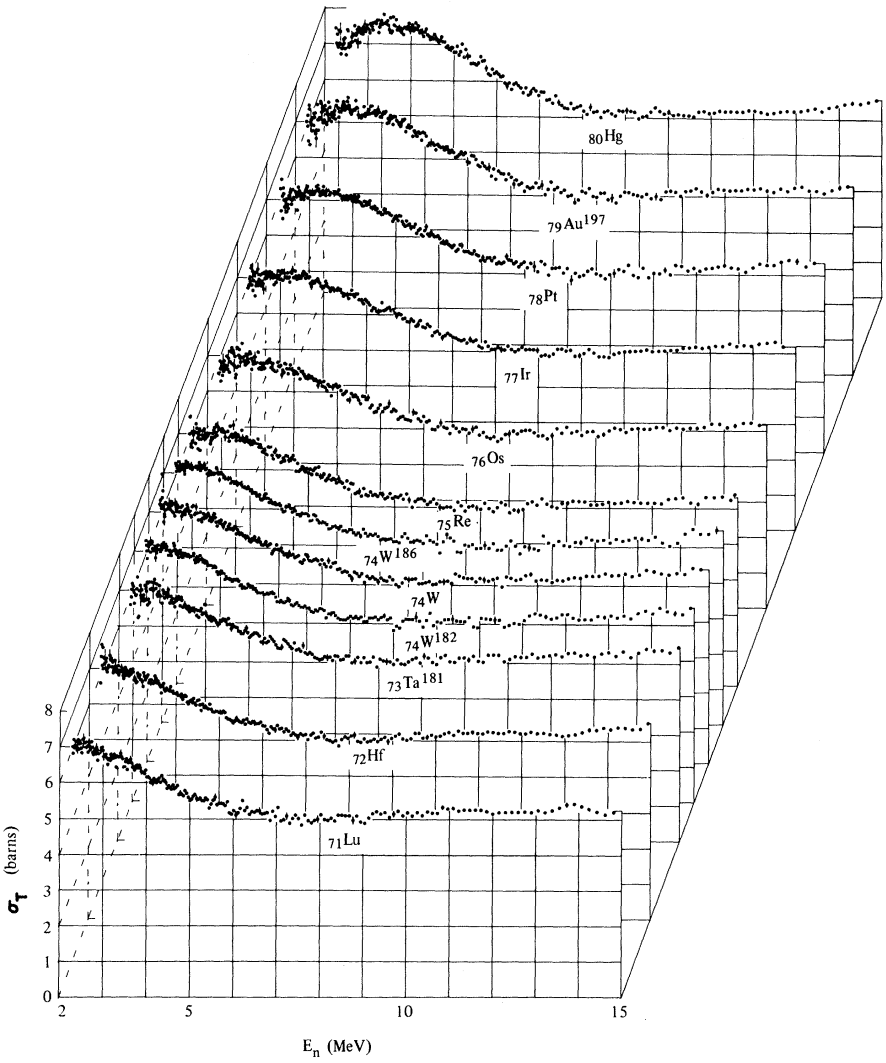

FIG. 10. Three-dimensional plot of the present results, for $Z=71$ to 80, drawn to the same scale as Fig. 3. The standard deviations are shown on every 10th point. W^{182} and W^{186} are shown clustered around natural tungsten (which is mostly ^{184}W) at $Z=74$. Note the spurious fine structure above 11 MeV (discussed in the text) for several of these elements.

Energy Scale

In an effort to verify our energy scale, we have compared the prominent features in the highly structured cross sections of the elements lighter than scandium with the corresponding features as measured by other laboratories. With suitable allowance for differences in energy resolution, we find that our energy scale agrees with most other authors within ±15 keV for all elements except oxygen. In the case of oxygen our energy scale appears to be about 25 keV low up to 6 MeV, although the data in the literature are not consistent among themselves. The 15-keV limit is approximately what is expected from the counting statistics of our differential-channel-width calibration. It amounts to one channel at 2.5 MeV and $\frac{1}{3}$ channel at 6 MeV.

In one important respect, not all of our data were taken under strictly equivalent conditions. Our earliest measurements used an average channel width based on the interval between two successive beam bursts, when operating with two bursts/cycle. This interval is usually not exactly one-half rf period. In the 29 elements for which one contributing run lacked the more exact full-period calibration, only three cases (carbon, sulfur, and calcium) required a correction in order to match the fine structure with that found in the later runs. Of these, carbon presented a "double

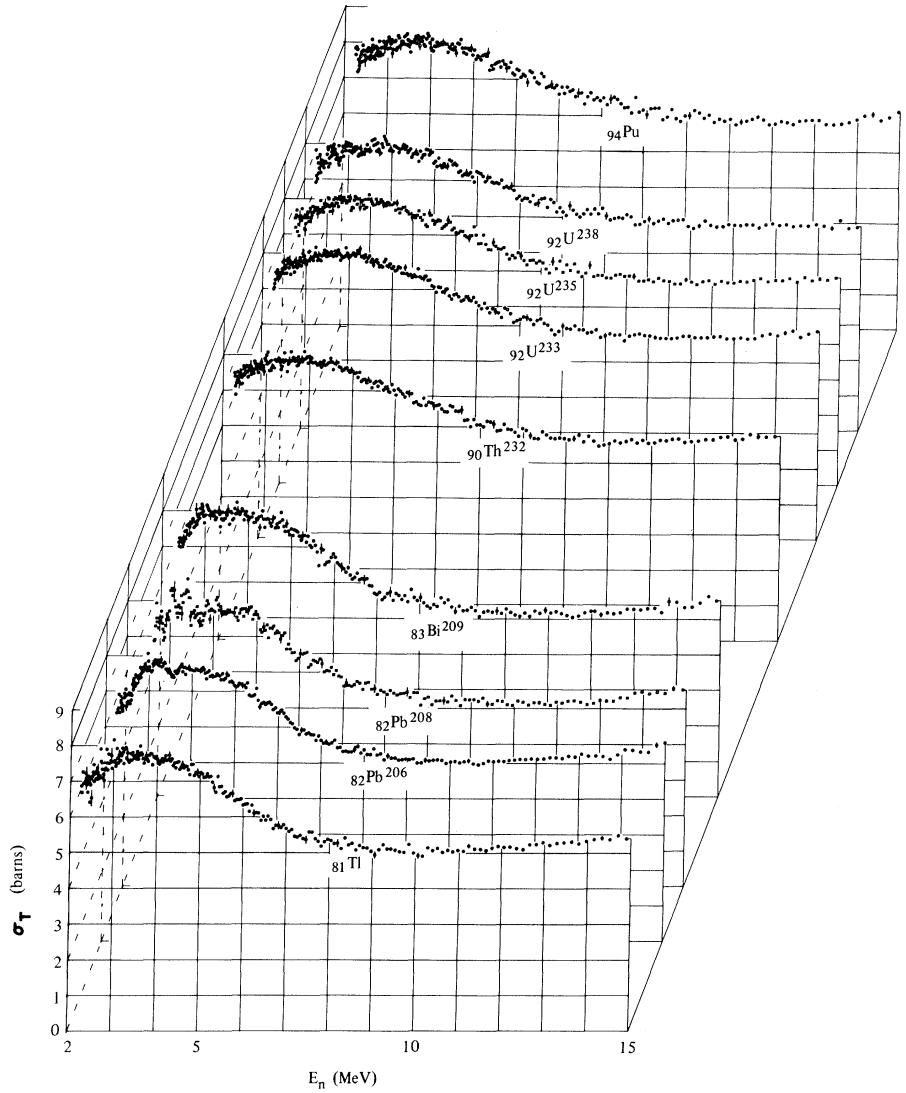

FIG. 11. Three-dimensional plot of the present results, for $Z=81$ to 94, drawn to the same scale as Fig. 3, except that eight units in Z have been omitted between bismuth and thorium, and the isotopes of lead and uranium have been spread rather widely around their nominal positions for greater clarity. The standard deviations are shown on every 10th point. The structure shown at low energies in the lead isotopes is real.

coincidence," in which internal evidence established that the *differential* channel-width distribution observed for the *later* run was incorrect also, thus jeopardizing the final energy scale by about 50 keV between 3 and 6 MeV. Using the differential channel width from either the preceding or the succeeding element in the later run yielded an energy scale consistent with other published measurements. We have made this substitution, but this empirical procedure necessarily increases the uncertainty in the energy scale. Indeed, Davis and Noda[46] report a 50-keV (0.7 channel) disagreement with our energy scale at 8 MeV.

The only elements heavier than scandium for which fine structure proved sufficiently pronounced to serve as a test of our energy scale were iron and lead. The comparison for iron is reasonably satisfactory. For lead, however, the agreement with the literature is relatively poor, but internal evidence for a systematic error in the earlier run is insufficient to justify the correction procedure which we used on carbon, sulfur, and calcium, so we have left it in its original form.

Technique of Comparison

The systematic quantitative comparisons with the previous literature were carried out on a digi-

tal computer, using only points which lay within our energy range. We made no effort to smooth either our data or the set being compared, to adjust them to the same resolution, or to apply corrections which the authors omitted that later were shown to be necessary. The effect of differences in resolution was estimated visually after completing the comparison. At each point measured by another author, we interpolated linearly between the adjacent points from our results, and computed the difference d and the standard deviation δd of the difference (including the effect of the covariance in our data). The averages of (d/σ_T) and $(d/\delta d)$ (with the sign of d retained in the averaging) and the rms value of $(d/\delta d)$, taken over $\frac{1}{2}$- or 1-MeV bins and over the entire range of energies, constituted our principal tools in characterizing each comparison.

The average difference $\langle d/\sigma_T \rangle_{\rm av}$ is, in effect, a measure of "normalization error." A systematic disagreement of this type arises primarily from errors in sample thickness or composition, inscattering errors, rate-dependent counting losses in the apparatus, or an incorrect determination of background, either in our own work or in the result to which it is being compared. It may also occur because of differences in resolution, particularly if a higher density of points is used in measuring narrow peaks than in the smooth regions between peaks (as is often the case). For data having the same resolution as our own, the rms disagreement $\langle d/\delta d \rangle_{\rm rms}$ (for which the expectation value is unity) is the primary index of compatibility, but the averages are necessary in assessing the effect of differences in resolution. Unfortunately, many authors do not separate random errors from estimates of systematic errors in reporting their results, so that the meaning of the differences divided by their standard deviations is somewhat ambiguous.

Some of the more important results of this systematic process of comparison are summarized in Table IV. We have grouped together results obtained at a given laboratory under substantially the same conditions over a reasonably short interval. The first five numbered entries refer to measurements which agree closely with our own, and which we regard as validating our own results. The remaining entries are ranked roughly in order of the "size" of the overlap with our measurements, giving relatively more weight to the number of different elements in common than to the total number of points lying within our energy range. With a few exceptions, we include work covering fewer than 10 elements (or isotopes) only if it has a large total number of points, and similarly include work which overlaps ours with fewer than 100 points only if it includes an unusually large number of elements.

For each laboratory and series of measurements, the table gives the energy range covered (with the understanding that the entire range may not have been covered for each element), the number of elements covered, the range of elements covered, and the total number of points used for comparison. The rms disagreement, expressed in units of the standard deviation per individual compari-

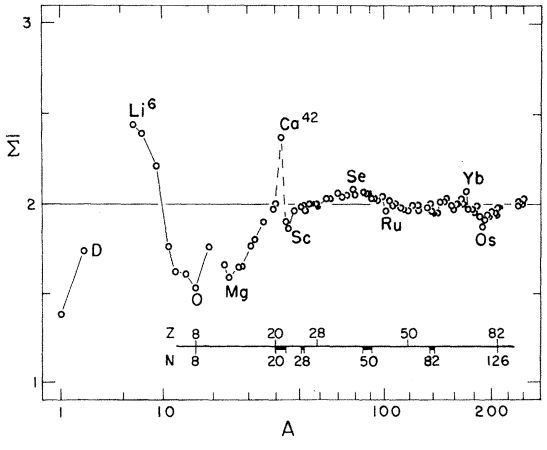

FIG. 12. The average ratio $\bar{\Sigma}$ of the measured total cross section to the geometric cross section, as a function of atomic weight. The abscissa is linear in $A^{1/3}$, and the inset scales indicate the location of closed shells.

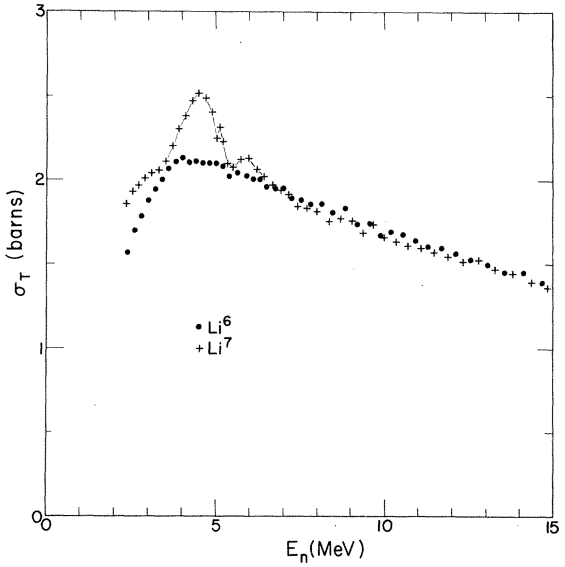

FIG. 13. Two-dimensional plot of the present results for the isotopes of lithium. The points are averages of 3 to 15 channels. The barely resolved peak in Li^7 at 5.1 MeV appeared in both natural lithium and enriched Li^7.

TABLE IV. Systematic comparisons with results of other laboratories.

Entry No.	Laboratory	References[a]	Energy range (MeV)	Elements covered No.	Range	Total points compared	rms Difference[b]	Average percent difference Low[c]	Av	High[c]	Agreement[d]
1	Theoretical fit	46–51	2.2–15	1	H	124	1.15		0.1		G
2	Wisconsin (3)	53,54	2.5–15	23	Be–Bi	4141	2.64	−1.4 Be	1.2	3.2 Pr	G
				17	(omit C–S)	1672	1.39	−1.4 Be	0.9	3.2 Pr	G
3	Los Alamos (1a)	8,49	14.1	58	H–Pu	58	0.94	−4.6 H	0.3	7.5 K	G
	(1b)	55	13.1–15	13	C–Pb	40	1.60	−3.4 La	0.6	3.2 Au	F
				11	(omit La, Gd)	34	1.18	−1.7 C	1.0	3.2 Au	G
4	Harwell	56,57	2.2–15	3	Na, Ho, U^{235}	752	1.16	−2.0 Na	0.5	3.2 U^{235}	G
5	Padova	59	<8.6	6	Na–S, Th	659	2.22	−6.6 Na	−0.8	2.0 Si	F
				5	(omit Na)	544	2.05	−2.0 P	0.4	2.0 Si	G
6	Los Alamos (2)	10	2.7–14	40	H–U	1144	0.78	−6.6 Cl	−1.6	3.4 Zr	F
7	Wisconsin (1)	6,68–70	<3.4	45	D–Th	508	2.60	−16.6 Ba	−2.2	15.9 As	P
				27	D–I	422	2.51	−9.3 B	−1.1	15.9 As	P
				18	Ba–Th	86	3.04	−16.6 Ba	−7.6	−1.0 Nd	P
8	Oak Ridge	83,75	<5	13	D–Cd, Hg–Pb^{208}	798	1.66	−8.7 Fe	0.4	6.1 N	F
				10	(omit N, F, Fe)	481	1.18	−3.6 Pb^{208}	−0.5	3.1 Se	G
9	Tokai-Mura (1)	73	3.0–5.1	9	B–Cl, Th	629	2.69	−3.4 C	4.4	13.7 O	P
	(2)	74	12.5–14.0	3	C, Al, Si	72	2.37	−3.5 C	2.5	12.5 Si	P
10	Livermore (LRL)	11	7.0–14.5	40	H–Pu	193	1.53	−3.6 Ta	0.9	6.3 D	F
				38	(omit D, Ta)	183	1.41	−1.8 C	1.0	3.4 Zr	F
11	Case Western Reserve	28	2.2–15	6	Mg–Fe, Pb	1849	2.14	−5.2 Mg	−2.2	−0.5 V	P
12	Rensselaer	76	2.2–15	6	C, Ta–Bi	1627	1.44	−2.9 W^{182}	0.2	3.4 Ta	F
				5	(omit C)	1256	1.27	−2.9 W^{182}	−0.5	3.4 Ta	F
13	Zürich	77	<3.8	9	Be–Si	506	2.55	−6.0 Be	4.2	9.6 Si	P
14	Wisconsin (2)	71,72	3.4–8.2	7	Li–F	618	2.10	0.2 Li	5.9	17.5 N	P
15	Osaka	62	2.3–2.8	35	H–Bi	87	4.34	−44.8 Ti	−7.2	50.4 N	P
16	Los Alamos (3a)	12	<8, (14.8)	3	D, C, O	252	e	5.6 D	13.2	19.0 O	P
	(3b)	13	<8	4	Al, U^{235}–Pu	226	(1.10)[e]	−2.0 U^{235}	1.2	2.2 Al	G
17	Madrid	78,79	3.2–5.2	8	V–I, Bi	363	1.60	−4.4 Mn	−0.1	2.6 In	F
				7	(omit Mn)	301	1.38	−0.9 Co	0.8	2.6 In	F
				3	V, I, Bi	112	1.10	0.3 V	1.2	1.8 I	G
18	Catania	84	2.4–5.2	6	Na–S	564	2.84	−2.0 Mg	2.7	7.6 Si	P
				5	(omit Si)	480	2.44	−2.0 Mg	1.9	6.6P	F
19	Columbia U. (1)	f	2.9	22	H–Sn, Hg, Pb	28	2.38	−26.8 Hg	−0.3	11.6 O	P
				20	(omit Cu, Hg)	26	1.71	−9.5 Sn	1.2	11.6 O	F
20	Columbia U. (2)	16	4.0–8.2	7	Mg–Mo, Pb	268	1.49	3.6 Mg	6.9	8.5 Pb	P
21	Louvain (1)	67,80	13.8–14.8	9	C–Ag, Pb, U	171	1.34	−2.7 U	−0.9	1.0 Ag	F
	(2)	g	<3.2	5	Mg, Ca, Cu, Pb, Bi	39	1.65	−7.7 Pb	1.3	8.4 Mg	F
22	Texas	81	<2.8	11	C–Br, Ba, Pb	140	2.86	−4.7 Mg	2.5	16.3 Ti	P
				8	(omit C, K, Ti)	91	2.05	−4.7 Mg	0.3	8.5 Pb	P

TABLE IV *(Continued)*

Entry No.	Laboratory	References[a]	Energy range (MeV)	Elements covered No.	Elements covered Range	Total points compared	rms Difference[b]	Average percent difference Low[c]	Av	High[c]	Agreement[d]
23	Hamburg	82	4.1–6.3	5	Li–Cu	207	2.35	−8.4 K	−3.0	0.8 Li	P
24	U.S. Nav. Rad. Lab.	15	3.8–8.0	5	Be–Ge	204	3.32	5.7 Co	7.1	10.4 F	P

[a] The references are given in the footnotes to the text (with the exception of e and f, which are to follow).

[b] In units of the standard deviation of an individual-point comparison.

[c] The most negative and the most positive difference are given together with the element in which each occurs.

[d] G = good, F = fair, P = poor.

[e] Most standard deviations not given.

[f] W. H. Zinn, S. Seely, and V. W. Cohen, Phys. Rev. 56, 260 (1939).

[g] G. DeConninck and A. Martegani, Bull. Classe Sci. Acad. Roy. Belg. 44, 851 (1958); Ann. Soc. Bruxelles 74, 64 (1960).

son, is given as an over-all index of the difference between each set of measurements and our reference surface. The columns which follow give the average percentage difference for the set, as well as the most negative and most positive average differences from our surface and the elements for which they occur. In many cases a few elements stand out as particularly divergent from our results. For these cases we have added a second line, which summarizes the comparison when these are omitted from the set.

Table IV is intended primarily to emphasize *systematic* disagreements, which we have found to be far more prevalent than would be expected from the simplicity of a transmission measurement. Unfortunately, such a condensed summary can be misleading, especially when the resolution differs very much from our own or when the density of points is nonuniform. Accordingly, in the final column we give a qualitative assessment of the agreement, which takes such complications into account. In the absence of complications, we consider the agreement *good* if the difference averages less than 2%, and the rms difference is less than 1.5 standard deviations. If the differences lie outside of both of these limits, we consider the agreement *poor*; otherwise it is labeled *fair*. For brevity, we will cite the entries in Table IV by the entry numbers given in the first column.

Validation of the Reference Surface

The most accurate set of cross sections for comparison in the MeV range is undoubtedly that provided by calculating the cross section of hydrogen from effective-range theory fitted to the existing precise measurements[46–50] at selected energies (entry 1). Using a revised fit by Gammel,[51] we find that our average disagreement with the fit is 0.1%, and the rms disagreement is 1.15 standard deviations.[52] This gives us some confidence in our method, but these summary figures overstate the agreement. Our data have residual spurious structure above 11 MeV with an amplitude of 4.6%, which gives rise to correspondingly large disagreements with the 14.1-MeV measurements of Poss *et al.*[47] and Coon *et al.*,[49] and the average between 13 and 15 MeV is 1.2% higher than the fit. Similarly, as we pointed out in Sec. II, our results are systematically low (averaging 4.2% or 1.4 standard deviations) below 3.5 MeV, where the subtraction of the cross section of carbon from that of CH_2, even though performed with a blank,[33] left traces of spurious structure and disagreement slightly outside of statistics between our two independent runs. Davis and Barschall[46] later revised the energies of several of the mea-

surements on which Gammel's fit is based, and substantially improved our agreement with the isolated point measured by the Columbia group[50] at 3.2 MeV, but we have not attempted to determine by how much the agreement over the entire region below 3.5 MeV has been affected by this readjustment.

The largest body of precise, recent data[53,54] with which we have been able to make detailed comparisons comes from the tandem accelerator at Wisconsin (entry 2). This work includes more than 4100 points within our energy range, distributed over 23 elements in the mass range from beryllium to bismuth, taken at higher resolution than ours throughout. The average disagreement is 1.0%, which is probably within the uncertainty of their inscattering correction (Table II shows a rather high inscattering index of 66×10^{-3}, which required corrections up to 8%), and only for praseodymium (where sample contamination is a persistent problem) does the disagreement average more than 2%. On closer examination, oxygen displays systematic disagreement in both energy scale and cross section. Quantitative comparison is hampered by the major difference in resolution, but if we minimize this effect by omitting the elements between carbon and sulfur, the rms difference is only 1.4 standard deviations. The Wisconsin data were taken point by point rather than simultaneously by time of flight, and eight of their cases span nearly our entire energy range. From this comparison we see no consistent indication of any energy-dependent systematic error greater than 1% that appears to be inherent in our continuous-spectrum time-of-flight method (other than the spurious fine structure above 11 MeV).

An additional verification, which is free of any question regarding large inscattering corrections, is provided by the 14.1-MeV measurement of Coon, Graves, and Barschall[8,49] at Los Alamos (entry 3), which provides comparisons on 58 elements taken at an inscattering index of only 3×10^{-3}. The average disagreement is 0.3%, and the rms disagreement is 0.94 standard deviations. This includes the 1.9-standard-deviation disagreement in hydrogen alluded to above, and a 3.5-standard-deviation disagreement in potassium, which is probably caused by difficulty in fabricating a reliable sample. It seems logical to add to this comparison the work of Conner[55] (also at Los Alamos) above 13.1 MeV, of which 40 points lie within our range (second line of entry 3). Coon *et al.* used a single energy which (as we pointed out in Sec. III) coincidentally was nearly immune to our spurious structure. Conner's broader sampling of the energy range, on the other hand, yields an rms disagreement of 1.6 standard deviations, although the average disagreement is only 0.6%.

As an example of a comparison with a large-accelerator time-of-flight system, we cite the 752 points from the Harwell synchrocyclotron[56,57] for Na, Ho, and U^{235} (entry 4), for which an average disagreement of 0.5% corresponds to an rms difference of 1.2 standard deviations.[58] An interesting hybrid system, used by the Padova group[59] to investigate fluctuations below 8.5 MeV (entry 5), combines the resolution and precision of a monoenergetic neutron source with the background rejection of a pulsed time-of-flight system. For 544 points[60] in Al, Si, P, S, and Th the average disagreement is 0.4%, and the 2.2-standard-deviations rms disagreement is caused almost entirely by the ability of their higher resolution to resolve the fluctuations still visible at these excitation energies.

We have shown at this point that a number of recent measurements, characterized by either very high precision, a large total number of points, coverage of a large number of elements, or some combination of these, are substantially in agreement with our results over large sections of the surface of cross section vs mass vs energy, and that the surface shows substantial internal consistency. Although the rms difference is completely within counting statistics only for the work of Coon *et al.*,[8,49] and much of the excess is attributable to differences in energy resolution or to our persistent false structure above 11 MeV, in almost every case there remain some additional systematic differences, which range up to approximately one half of the random error of an individual point. These are typically in the range of 1 to 2%, which is consistent with our expectation from internal evidence.

Comparison with Other Major Measurements

Many of the remaining measurements cited in Table IV were made with techniques and statistical precision comparable to those summarized in the first five entries. Although some of them are in good *average* agreement with our results, they display substantial systematic trends which mark them as lying off the surface defined by our results. Others have the same variation with energy but a substantial constant displacement from our surface (notably those in which high statistical precision was obtained at the expense of high inscattering).

We note in passing an overwhelming preoccupation, in previously published work, with the heavily structured cross sections of the light elements, rather than with the systematic properties of nuclear matter which can be seen consistently only

in the heavier elements. Within our energy range, almost one quarter of all the points we obtained from the Livermore compilation are devoted to just three light elements (carbon, magnesium, and aluminum), more than half are for $Z < 16$, and one fifth of the remainder are for iron and lead. Of the reasonably stable elements, we ourselves lack five noble gases. In addition to technetium and promethium, the existing literature lacks 10 stable elements altogether, five of which are rare earths, and offers only a single point at 14 MeV for U^{233} and three additional elements.

Groups engaged in compilation and evaluation of cross sections for practical applications have tended, in recent years, to discard total cross sections which were measured before 1950. In general we find this to be justified. The pioneering work of Ladenburg and Kanner[61] in 1937 actually lies on our reference surface within statistically acceptable limits, although the average error is 10%. The results of Aoki and Kikuchi[62] (entry 15) include five elements which lie more than 25% off the surface, and the rms disagreement is far outside statistics. Typical average errors remained 5 to 15%, with occasional excursions of more than 25%, until about 1946, when the Minnesota group[63] published measurements on hydrogen and deuterium up to 6 MeV which lie on the surface within about 1%, with statistically acceptable scatter. Their resolution was too broad to deal adequately with the highly structured cross sections of carbon and oxygen, however. After 1950 we find only three instances of a measurement which averages more than a 25% displacement from our surface; namely, the results of Lasday[64] on zirconium, Zubov, Lebedeva, and Morozov[65] on boron, and Winterhalter[66] on enriched Ca^{40}. Average departures greater than 10%, however, have continued to appear sporadically up to the present time, including (in all probability) our own measurement on Ca^{42}, as we pointed out in Sec. V.

We referred in Sec. I to the long-standing systematic disagreement between the results of Nereson and Darden,[10] BPS,[11] and Weil and Jones[16] (entries 6, 10, and 20 in Table IV, respectively). The intention of Nereson and Darden (entry 6) was quite similar to our own; namely, to survey the 3–14-MeV region without gaps in the energy scale by the use of a continuous spectrum, which they obtained from a reactor. Their results lie systematically below our surface between 7 and 13 MeV for elements heavier than iron, with an amplitude which reaches 8% in the neighborhood of lanthanum. In the heaviest elements this tendency is reinforced by an inscattering error of up to 4%,[67] corresponding to their inscattering index of 10×10^{-3}. Although their original counting statistics were of the order of a few percent, the final errors quoted included an overly generous allowance for just such systematic errors, so that the systematic departure from our surface is masked by an apparent rms deviation of only 0.78 standard deviations (0.70 if tungsten is omitted).

BPS's results[11] for isolated points between 7 and 15 MeV, on the other hand, display a complementary departure from our surface (entry 10) with a magnitude about one third of that exhibited by Nereson and Darden. From magnesium to palladium their results lie systematically above ours, while above palladium their results slope downward into ours, with the greatest discrepancy occurring between 7 and 10 MeV. Their inscattering index was small (2×10^{-3}) and easily corrected for. The systematic difference is again partly masked by a blanket allowance for systematic errors, which in this case was 0.5%, and the apparent rms difference drops from 1.53 to 1.41 standard deviations if just two elements (deuterium and tantalum) out of 40 are eliminated from the comparison. The largest discrepancy is +6.3% in deuterium, which is especially surprising inasmuch as our measurements were performed on the same sample as theirs, and there had been no apparent change in the weight or dimensions of the sample.

The results of Weil and Jones[16] lie systematically above ours (entry 20), as well as above nearly every other recent measurement. Weil and Jones used a uniquely thin scintillator to minimize sensitivity to γ rays, but we are unable to propose a mechanism which would connect this unique property with their systematically high cross sections.

The unpublished work[28] from Case Western Reserve (entry 11), on the other hand, apparently still contains a systematic effect which is discussed by the authors themselves. Their technique was very similar to our own in its use of $\mathrm{Li}(d,n)$ neutrons as a source for a time-of-flight system. In their initial work, the very high counting rates associated with the most energetic neutrons produced a severe dead-time distortion of the neutron spectrum which was dependent on counting rate, and led to a systematic upward slope in the cross section. Revisions in the electronic system overcame most of this problem, as they succeeded in demonstrating by a remeasurement of the cross section of iron, but their results still display a systematic positive slope relative to ours. We have noted above that there is some doubt about our energy scale for lead. Lead is the only example in which the Case results fail to agree with our energy scale.

The work of the Wisconsin group[6,68–70] below 3.4 MeV (entry 7) had a profound effect on the evolution of the optical model[9] of the nucleus, and was

directly responsible for the original appearance of the Barschall[5] surface. Most of this work was performed at energies below our range, and only a few isolated points overlap ours. The inscattering index was very large (187×10^{-3}), and Walt *et al.*[68] succeeded in measuring directly the strong inscattering from the forward diffraction peak in the elastic cross section. Unfortunately, however, the appropriate correction was not applied to the earlier parts of the Wisconsin work, and its absence is dramatically evident in the third line of entry 7 in Table IV. Vervier, DeConninck, and Martegani[67] showed in 1958 that a proper correction could be calculated for the Wisconsin results, which would remove most of the error, but the corrected results do not appear in the major compilations. The work of Bockelman *et al.*[69] on the lightest elements was done with a much higher density of points, and should be free from inscattering errors. Unfortunately, it also exhibits systematic departures from our surface, in addition to those caused by differences in energy scale and resolution.

The higher-energy extension of the early Wisconsin work (entry 14) exhibits a very large disagreement for nitrogen,[71] although we used the same compound (5-aminotetrazole)[34] for a sample that they did. Large rms differences in carbon and oxygen are again attributable mainly to our inferior resolution. Curiously, the energy scale for oxygen in part of this work[72] is in excellent agreement with ours, although the earlier[69] and later[54] Wisconsin work at lower and higher energies, respectively, is in disagreement. The fine structure in carbon and oxygen was subsequently confirmed by Fossan *et al.*[54]

The remaining work listed in Table IV includes many systematic disagreements which are real and unresolvable, as well as many which are caused primarily by differences in resolution. The latter accounts for much of the disagreement for silicon, sulfur, and chlorine in entry 9[73,74] and for carbon in entry 16.[12] Examples of the former include nitrogen, fluorine, iron, and Pb^{208} in entry 8,[75] W^{182} in entry 12[76] (which may be caused by the oxygen in our sample), all of entry 13[77] (carbon exhibits a shift in energy scale), the first line[12] of entry 16, most of entry 17[78] (where the main effect is scattering from the sample support above 4.5 MeV),[79] the first line[67,80] of entry 21 (which shows improbable curvature and slope for the 14-MeV region), titanium in entry 22,[81] potassium in entry 23,[82] and all of entry 24.[15] On the other hand, agreement almost within resolution and statistics marks nine elements[83,75] in entry 8, three isotopes in entry 12,[76] the second line[13] of entry 16, and three elements in entry 17.[78]

It is worth remembering that many of the measurements which we are scrutinizing here were made with special purposes in mind. For example, most of the early Wisconsin work[6,68-70] above 1.6 MeV (entry 7) was taken only at isolated high-resolution points, in order to obtain a general idea of the trend of the cross sections. The work of Nereson and Darden (entry 6)[10] was avowedly a low-resolution survey, for which a generous allowance for systematic errors was quoted. Much recent work, including that from Padova (entry 5),[59] Madrid (entry 17),[78] Catania (entry 18),[84] and the latest work from Wisconsin,[53] as well as others not included in Table IV, has been designed to obtain information about the fluctuations of the cross sections, for which neither accurate corrections for inscattering nor accurate knowledge of sample thickness was essential. In most instances the qualitative behavior of the fluctuations is reproduced by our measurements, within our limitations of resolution.

VII. CONCLUSIONS

Our primary effort in conducting these measurements was to cover the entire range of nuclear masses between 2.5 and 15 MeV under a single set of experimental conditions. Our hope in doing so was that the systematic properties of nuclei would be revealed more compellingly by such a blanket survey than by random and isolated measurements at better resolution and precision. We conclude this article with a summary of the properties so revealed.

We begin by comparing our measurements with the total cross section calculated from the optical model. Specifically, we have selected the phenomenological spherical nonlocal model of Perey and Buck,[85] using their original parameters (Set A). This model had the ambitious goal of describing all but the lightest nuclei over this entire energy range using a single set of parameters. In a more basic way, however, it represents a step towards the ultimate goal of calculating the interaction between neutrons and nuclei directly from the fundamental nucleon-nucleon interaction.

These calculations are discussed fully in a companion paper (II), and we shall only outline the conclusions. We recall first that Set A is based entirely upon fitting the differential elastic scattering cross section of Pb^{208} for neutrons of 7 and 14.5 MeV, subject to verification by comparison with the scattering from up to seven additional elements at 4.1, 7, 14.5, and 24 MeV. The calculations for our complete set of total cross sections confirm that Set A gives a remarkably accurate description of the total cross sections from 3 to 15 MeV for all spherical nuclei heavier than cal-

cium, but fails dramatically (>15%) for nonspherical nuclei by an amount which is approximately proportional to the deformation parameter. Most of the permanently deformed nuclei are concentrated in the latter part of the rare-earth region and in the actinides, neither of which regions was represented in the "verification" set of Perey and Buck. Furthermore, the calculations tend to cross our data for the rare earths near the particular energies (4 and 14 MeV) chosen for their comparisons, and likewise cross our data for the actinides near 7 MeV, so that the disagreement would have been obscured even if the data had been available at that time.[86] Although the basic problem lies in the use of a spherical model for a nonspherical nucleus, recent calculations[87] suggest that much of the discrepancy may be removed by a coupled-channel calculation which contains explicitly the interaction through the energy levels in the ground-state rotational band. Other possible modifications to the model include a new parameter search with the Perey-Buck formulation in an attempt to fit the total cross sections of deformed as well as spherical nuclei, or else an attempt to generalize the nonlocal-separable-potential formulation to an explicitly nonspherical region of interaction.

While it is inevitable that the qualitative features of the giant-resonance diffraction structure should be reproduced by a suitable optical model, it is not necessarily obvious that a single set of parameters should correctly describe the "optical" properties of nuclear matter. In other words, even the nonlocal parameters may vary from element to element for specific reasons. One looks naturally for such effects in the neighborhood of closed shells.

In Sec. V we have referred to an apparent step in the average ratio of the total cross section to the geometric cross section, $\bar{\Sigma} = \langle \sigma_T/\sigma_g \rangle$, which occurs between potassium and scandium in crossing the 20-proton and 20-neutron shells (Fig. 12). However, we observe no pronounced anomalies at the remaining closed proton shells, nor at the 28-neutron shell. Close examination of Figs. 5 and 6 reveals that the $n=3$ ridge (in Peterson's[19] notation) moves upward from 5 MeV in iron to 9 MeV in yttrium, with the $n=4$ valley apparently preparing to emerge from lower energies between nickel and gallium. At germanium, however, the incipient valley abruptly begins to disappear, and the diffraction structure is scarcely perceptible in selenium. It reappears between bromine and rubidium, and is well marked in the three elements (strontium, yttrium, and zirconium) in which the dominant isotope has a 50-neutron closed shell.

A similar anomaly occurs at the 82-neutron shell, although it is less conspicuous (we first observed it in preliminary data after dividing the total cross section by the geometric cross section and making a smoothed contour plot to enhance the diffraction structure). The bottom of the $n=4$ valley drops abruptly between barium and lanthanum (the first two elements dominated by the closed 82-neutron shell), and remains low through neodymium. It then rises abruptly in Pm^{147} and samarium, which are well off the closed shell.

It is difficult to relate any anomalies near the 82-proton/126-neutron closed shells directly to shell closure. Figure 12 shows a marked decrease in $\bar{\Sigma}$ between ytterbium and osmium, accompanying the return to spherical nuclei, followed by a steady rise up to bismuth, with no particular fluctuation on passing through the isotopes of lead. Close inspection of Fig. 11 will show that the crest of the $n=5$ ridge (just above 3 MeV) has a nearly constant value of approximately 7.7 b from mercury through the lead isotopes, and rises about 0.2 b in bismuth, but this effect again is scarcely visible except by comparison with the geometric cross section (or the nonlocal optical-model calculation described in II). The most conspicuous effects in this region probably occur below the lowest energy of our measurement.

We have pointed out in Sec. I that our resolution was inadequate to give a full account of the fluctuations which exist within our energy range. This is clearly documented by the results already obtained by the Karlsruhe group.[45] As mentioned in Sec. III, we have no results yet from a least-structure analysis[39,40] of our data, which could serve to remove the simulated fluctuations caused by covariance. For the present we regard with skepticism any fluctuation smaller than three standard deviations.

Nevertheless, one major conclusion regarding fluctuations can be derived from Figs. 3–5. Fossan *et al.*[54] observed that fine structure disappears rather abruptly, in the elements from beryllium through oxygen, at an excitation energy in the compound nucleus of 11 or 12 MeV. A similar abrupt disappearance of structure can be observed in our results from fluorine through copper, at a characteristic bombarding energy which differs from element to element by considerably more than the differences in binding energy. A closer study reveals that the greater the ground-state spin of the nucleus, the smaller is the amplitude of the fluctuations and the lower the neutron energy at which they were no longer resolved by our apparatus. Since both the Ericson[88] and Agodi[89] fluctuation models predict that the mean square variation of the total cross section is proportional to $(2I+1)^{-2}$, where I is the ground-state spin of the target nucleus, this observation does not serve to distinguish between

the two models, although it argues against the need to invoke doorway states[90] in order to explain the behavior of the fluctuations. We first observed the systematic behavior of the fluctuations as an odd-even variation as a function of atomic number. However, the sequence V^{51}-Cr^{52}-Cr^{53}-Mn^{55} (for which $I=\frac{7}{2}$, 0, $\frac{3}{2}$, and $\frac{5}{2}$, respectively) in Fig. 5 strongly suggests that I is the relevant paremeter.

Although our moderate resolution was not expected to yield any new information on the nuclear spectroscopy of the lightest elements, the particular choices of energy range, spacing, and resolution applied to lithium and its isotopes by previous authors[10, 14, 65, 69, 71, 82, 91] have caused them to overlook several interesting features which appear in our results. The curves from Fig. 3 are reproduced in Fig. 13, plotted together instead of in three dimensions, and with adjacent channels averaged together for greater legibility. We note first that the cross sections are indistinguishable above 7 MeV, and almost equal at 3.3 MeV, even though the radius of Li^7 predicted by Eq. (5) is more than 4% larger than the radius of Li^6. The cross section of Li^7 rises above that of Li^6 in four distinct structures. The excess below 3.3 MeV persists down to a crossover at approximately 1 MeV. Above 3.3 MeV there are broad peaks at 4.6 and 5.8 MeV ($\Gamma \sim 1.0$ and 0.4 MeV, respectively), and a narrow peak at 5.1 MeV ($\Gamma \sim 0.08$ MeV). The latter was barely resolved by our equipment, but appeared independently in our measurements on natural lithium and enriched Li^7. Since the measurement by Becker and Barschall[71] covered only the range from 4.3 to 5.5 MeV, it did not quite span the 4.6-MeV peak, which thus failed to be conspicuous, and their 5% statistical precision was insufficient to reveal the 5.1-MeV peak, which rises only about 5% above the side of the 4.6-MeV peak.

If the dineutron exists, it has been predicted that there would be a Wigner cusp in the total cross section of deuterium near 3 MeV. We have already discussed, in a previous publication,[3] an examination of our results for evidence of such a cusp, without success. Alfimenkov *et al.*[3] have shown conclusively that the original calculation of the magnitude of the cusp was made with the wrong set of s-wave scattering lengths, and the correct set yields a cusp which would be vanishingly small even if the dineutron is bound. This test is therefore inconclusive.

In a similar vein, Hrehuss and Czibók[92] have recently claimed to have discerned oscillatory fine structure in their own and previously published measurements of the total cross section of hydrogen, for which they proposed an explanation based on charge-state flipping. We have indicated above that our results for hydrogen contain some departures from the effective-range theory below 3.5 MeV, which include spurious structure related to the cross section of carbon. Nevertheless, the "best fit" to the charge-state-flipping theory predicts variations of up to 5%, which would be clearly distinguishable from our experimental spurious structure. While we have not performed a full statistical analysis, we find no evidence whatever for the presence of the proposed oscillations in our data, a conclusion supported by other recent work.[93] This serves to underline the importance of measuring the cross section of hydrogen over a range of several MeV using the same apparatus and techniques for each point, as we have done. We have emphasized in the preceding section that it is the rule, rather than the exception, for the work of different laboratories to exhibit systematic differences comparable with the random errors. When the results of one group are interspersed among those of other groups, therefore, it is easy to produce the appearance of a systematic oscillation. The H_2O-D_2O subtraction described by Hrehuss and Czibók contains the same kind of pitfall as our CH_2-C subtraction; namely, interference by the very highly structured cross section of oxygen, with the additional pitfall that if hydrogen can be shown to have an oscillatory cross section there remains no convincing reason for supposing that the cross section of deuterium would be smooth instead.

VIII. SUMMARY

We have completed here the description of an experimental program for the factory-scale measurement of total cross sections for neutrons between 2.5 and 15 MeV. We used a continuous-spectrum pulsed-source time-of-flight method to achieve resolution and precision in the few-percent range, with several points per resolution interval, and covered nearly all of the natural elements and a number of separated isotopes. Internal consistency and external comparisons imply that systematic errors should be less than 2% in almost all elements, in addition to a spurious fine structure above 11 MeV with an amplitude usually less than 1.5%. Extensive comparisons with almost all of the previous literature in this energy range reveal ubiquitous systematic disagreements outside of the quoted statistics, but agreement with a selected group of fairly recent major measurements is very good. We have presented the data themselves in the form of a three-dimensional graph, in order to emphasize the physical phenomena which the systematics of the total cross section reveal; the 24 000-point set of results itself is available through the international compilation system, and

this paper is designed to serve as its formal documentation. We have outlined some of the physical phenomena which such a systematic measurement reveals, but have not given a detailed discussion here.

IX. ACKNOWLEDGMENTS

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J. B. Marion and J. L. Fowler (John Wiley & Sons, Inc., New York, 1963), Sec. V.A.

[27]E. M. McMillan and D. C. Sewell, U. S. Atomic Energy Commission Report No. MDDC-1558, 1947 (unpublished).

[28]Galloway, III, and Shrader, Ref. 25.

[29]M. E. Rose and M. M. Shapiro, Phys. Rev. 74, 1853 (1948).

[30]The error is rising very sharply at 0.7 mfp (50% transmission), which is the thickness reported all too frequently in the literature. Even 70% transmission is not uncommon (Ref. 26).

[31]D. G. Foster, Jr., and D. W. Glasgow, Bull. Am. Phys. Soc. 8, 119 (1963).

[32]D. G. Foster, Jr., Hanford Laboratories Report No. HW-79054, 1963 (unpublished), p. 64.

[33]We were seriously misled by the term "reactor grade." Our sample of reactor-grade hafnium contained 6.2-at% zirconium, which was enough to justify the preparation of a zirconium blank. The situation with graphite is potentially more troublesome. In a private communication, T. L. Houk has informed us that he has observed radial density gradients in rods of reactor-grade graphite as high as 12%/cm, so that a beam which passes through substantially less than the entire area of a sample may encounter an appreciably different number of nuclei per cm^2 than is calculated from the average density of the sample. This problem can be avoided by using pyrolytic graphite.

[34]C. H. Johnson, B. Petree, and R. K. Adair, Phys. Rev. 84, 775 (1951).

[35]We completed work on these borrowed samples before we realized that small discrepancies in energy scale might prove disastrous to the algebraic correction. In retrospect, it would have been better to have used matched blanks of a well-defined oxide of an element with a slowly varying cross section (e.g., H_2O), and corrected for the additional element algebraically. A third possibility would have been blanks with a rapidly varying diluent (BeO, B_2O_3), for which a counter blank was added to the *sample*, as in the nitrogen sample of Ref. 34.

[36]Not surprisingly, all of these except strontium were powders.

[37]Since our aim was to bridge the gap between 3.2 and 14 MeV, we were unwilling simply to discard the data from the 11–14-MeV region, as other authors [e.g., Ref. 28 and J. Læroy, F. N. Berthelot, and E. Pomelas, J. Phys. (Paris) 24, 826 (1963)] have been compelled to do for similar reasons.

[38]In only one case, carbon, did the true fine structure [as shown by S. Cierjacks, P. Forti, D. Kopsch, L. Kropp, J. Nebe, and H. Unseld, Kernforschungszentrum Karlsruhe Report No. KFK-1000, 1968 (unpublished)] resemble the spurious structure closely enough to invalidate this procedure.

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[43]We find no internal evidence for such errors, however.

[44]CINDA 68 (ENEA Neutron Data Compilation Centre, Paris, 1968), and Suppl.

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[52]We emphasize that this ratio is referred to the standard deviation of a single point. It corresponds to $\chi^2 = 164$ for this 124-point comparison, which is a statistically significant disagreement. We shall use this convention consistently.

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