NUCLEAR DATA AND MEASUREMENTS SERIES

ANL/NDM-29

Note on the 250 keV Resonance in the Total Neutron Cross Section of ^{6}Li

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A.B. Smith, P. Guenther, D. Havel, and J.F. Whalen

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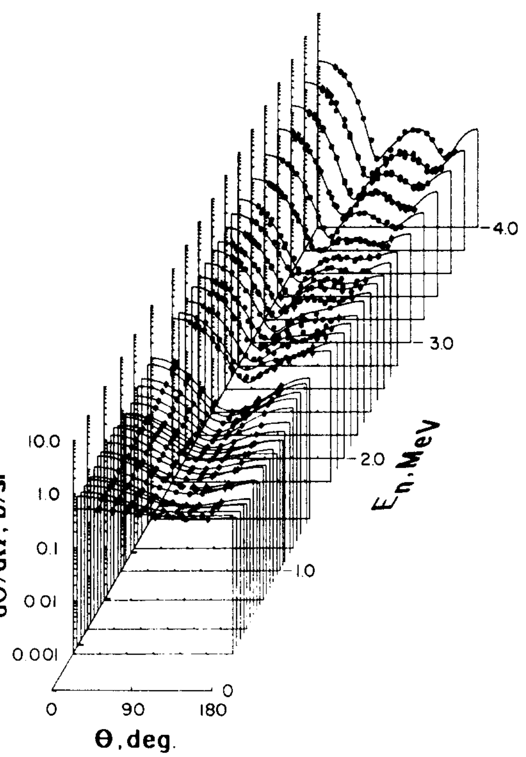

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In January 1975, the research and development functions of the former U.S. Atomic Energy Commission were incorporated into those of the U.S. Energy Research and Development Administration

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NUCLEAR DATA AND MEASUREMENTS SERIES

The Nuclear Data and Measurements Series presents results of studies in the field of microscopic nuclear data. The primary objective is the dissemination of information in the comprehensive form required for nuclear technology applications. This Series is devoted to: a) measured microscopic nuclear parameters, b) experimental techniques and facilities employed in measurements, c) the analysis, correlation and interpretation of nuclear data, and d) the evaluation of nuclear data. Contributions to this Series are reviewed to assure technical competence and, unless otherwise stated, the contents can be formally referenced. This Series does not supplant formal journal publication but it does provide the more extensive information required for technological applications (e.g., tabulated numerical data) in a timely manner.

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NOTE ON THE 250 keV RESONANCE IN THE TOTAL NEUTRON CROSS SECTION OF ^{6}Li*

by

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ABSTRACT

The energy of the observed maximum of the $\approx$ 250 keV resonance in the total neutron cross section of ^{6}Li is measured to be 244.5 ± 1.0 keV relative to the velocity of light. The observed peak magnitude is 11.20 ± 0.20 b.

*This work supported by the U.S. Energy Research and Development Administration.

I. INTRODUCTION

The $^{6}Li(n;\alpha)$ reaction is widely employed as a standard in neutron cross section measurements. Its primary value is at energies below $\gtrsim$ 100 keV and in this region the cross section is influenced by the detailed nature of the p-5/2 resonance at $\gtrsim$ 250 keV. The ^{7}Li system is relatively simple and extensive R-matrix interpretations have been used to correlate total and reaction cross sections to obtain optimum estimates of the $(n;\alpha)$ cross section (1). Such interpretations are sensitive to exact energy scales and cross section magnitudes, including those of the total cross section. Unfortunately, total cross-section energy scales can vary between measurements by several keV and there is some tendency for monoenergetic-source results to be higher in energy than those obtained using white-source techniques. Moreover, it is difficult to reconcile the observed total cross section magnitude of the resonance maximum with that of the $(n;\alpha)$ cross section as reported from a number of experiments. The present measurements had the objectives of: 1) determining the observed energy of the maximum of the total cross section resonance using a vernier technique combining advantages of both white- and monoenergetic-source techniques, and 2) obtaining a measure of the magnitude of the observed resonance maximum of the total cross section. Ancillary and concurrent measurements of iron, and carbon supported the validity of the lithium results.

II. EXPERIMENTAL METHOD

The present method employs a pseudo-monoenergetic neutron source, nanosecond time-of-flight techniques and a vernier time scaling to determine the energy scales relative to the velocity of light and a crystal-oscillator time base. Proton bursts of $\approx$ 1 nanosecond duration were incident upon a lithium target at a repetition rate of $\approx$ 1999.05 kHz. The lithium targets were vacuum evaporated to a thickness sufficient to provide a neutron energy spread of $\approx$ 100 to 200 keV centered about an incident energy of $\approx$ 250 keV. A fast scintillation detector was placed at selected positions between $\approx$ 4 and 14 m from the source at a zero-degree reaction angle in such a manner that gamma rays associated with the subsequent neutron bursts (multiples of $\approx$ 500 nsec. later) were approximately coincident with selected neutron energies in the range 200 to 300 keV. The neutron response of the detector was corrected for multiple-event effects using a Monte-Carlo procedure (3). The time-of-flight of the neutrons from the target to the detector was measured using conventional analog techniques. The timing system was calibrated to better than 1/2 percent accuracy using precision delay lines. This calibration and its associated uncertainty was applicable only over the few nanosecond vernier extrapolation from the gamma-ray flash to the neutron energy region of interest ($\lesssim$ 20 keV). Thus the timing precision was very largely determined by the accurately known and crystal-controlled pulse rate. The observed gamma-ray flash was of modest intensity and thus did not overload detection equipment but was clearly recognizable in the observed spectra. Flight paths were determined to ± 3 to 5 mm using a steel surveyor's tape. The uncertainty was largely associated with the determination of the effective center of the 2 cm thick liquid scintillator employed as a detector. A large shield with a meter long 1 cm diameter collimator was arranged about the neutron source with the collimator axis on a zero-degree reaction angle. The transmission samples were placed upon a wheel approximately 1.25 m from the source with neutrons incident on the bases of the right cylindrical transmission samples. The wheel rotated in a stepped motion alternately placing samples, empty containers and a void in the beam for periods of approximately 1/3 second. The sample position was correlated with detector response using an on-line digital computer system. A live-time clock was inserted into the data acquisition system to assure proper correction of small dead-time effects and to verify the equality of the time window of the sample wheel. Independent monitoring of source intensity was not required due to the rapid sample-changing sequence. Target thickness, flight paths, and timing scales were selected so as to clearly define time-uncorrelated backgrounds both before and after the detection of neutrons in the energy range of interest. The fidelity of the background interpretation was further examined using targets providing widely different energy spreads. No time correlated background could be identified in the energy region of interest.

III. SAMPLES

Five samples were measured concurrently; one iron, one carbon, one aluminum and two lithium. All samples were right cylinders with specifications defined in Table 1. The iron and aluminum samples were used as additional verifications of the energy scale. However, the observed resonances of the aluminum were not generally sharp enough to provide good check points. Carbon was used to test the fidelity of measured cross section magnitudes. The thicker ^{6}Li sample was obtained on loan from BCMN (Bureau Central de Mesures Nucleaires, Geel, BELGIUM) primarily for higher-energy cross section measurements. The thinner ^{6}Li sample was fabricated at ANL (Argonne National Laboratory) primarily for neutron scattering studies. The dimensionality of the ANL sample was not as well known as that of the BCMN sample as it was not precision machined after casting as was done for the BCMN sample. An identical empty container was available for both lithium samples (aluminum for the BCMN sample and stainless steel for the ANL sample). The iron and aluminum samples were fabricated from commercially available high-purity-material. The carbon sample was fabricated from pile-grade graphite and gave total cross sections agreeing to within $\lesssim$ 1 percent with those obtained using other and independent carbon samples. All the lithium samples contained $\approx$ 4.5 percent ^{7}Li and measured values were corrected for this heavier isotope using ^{7}Li cross sections given in ENDF/B-IV (2). The experimental results showed no evidence of chemical impurities, such as oxygen, and thus such impurities were assumed to be negligible as indicated by assay prior to fabrication. The samples were not destroyed in order to verify this assumption.

IV. EXPERIMENTAL RESULTS

The energy scales were independently deterimed in three measurements at $\approx$ 7 m and three at $\approx$ 14 m. They were tested by comparisons with prominent iron resonances in the range $\approx$ 170 to 270 keV as given by ENDF/B-IV (2). The measured iron cross sections were plotted on large scales and the resonance energies interpolated from the figures. The results are summarized in Table 2. The sets of data at 7 m and 14 m were obtained six months apart and yet are consistent both within themselves and between each other. Moreover, the results were consistent with those obtained at shorter ($\approx$ 4.5 m) flight paths. The latter short-flight-path measurements were used to determine the shape of the ^{6}Li resonance free of gamma-flash perturbations, not for energy scale determinations. The present results were generally consistent with resonance-energies of

ENDF/B-IV within the uncertainties of the measurements as outlined in Table 3. The present results were slightly biased toward higher energies than given in ENDF/B-IV possibly as a result of instrumental-resolution effects. This bias was small and a similar shift has been noted elsewhere (4). Precise observation of the gamma-ray flash was essential to the quality of the present results. It was assumed that the neutron and gamma-ray time responses of the detector system were identical except for the small correction for multiple-neutron events in the detector, noted above. Detector biases were set up so as to require the same detector pulse-height range for both gamma-rays and neutrons. The detector time response was tested by removing all electronic constraints on the gamma-ray detection; i.e. removing pulse-shape gamma-ray suppression circuitry and extending the maximum pulse-height selection range to a very wide interval. With this extreme variation in gamma-ray sensitivity, there was a shift in gamma-ray time response corresponding to a shift of $\lesssim$ 1 keV in neutron energy at 250 keV. In addition, a minor artifact was observed due to high-energy gamma-rays attributed to neutron capture about the target assembly and collimator entrance. These tests suggest that the time response of the detector was essentially identical for neutrons and gamma-rays. The above tests support the estimates of Table 3 giving an instrumental time-scale uncertainty of $\lesssim$ 1 keV from $\approx$ 150 to 300 keV. The internal consistancy of the measurements was a good deal better; e.g. $\approx$ 200 eV at $\approx$ 250 keV in the 14 m measurements.

The neutron-total-cross-section magnitudes were verified by concurrently determining the neutron total cross section of carbon at 245 keV. The carbon sample used in the measurements was selected to give a transmission similar to that of the thinner (ANL) ^{6}Li sample. It was one of a set of carbon samples that had been widely used in total cross section measurements at other energies and with other techniques with results that were generally consistent with published values to $\approx$ 1 percent. Many corrections, such as background effects, should have been similar for this carbon sample and the thinner ^{6}Li sample, thus a determination of the total cross section of carbon is a test of the validity of the accompanying ^{6}Li measurement. The measured carbon values obtained with flight paths of $\approx$ 450, 650 and 750 cm were averaged over the energy interval 240 to 250 keV with the results shown in Table 4. The results are consistent with the average to within much less than 1 percent. The corresponding ENDF/B-IV value is 1 percent larger. Similar comparisons were made with the experimental values of Whalen et al. (5), Perey et al. (6) and Uttley et al. (7) as indicated in Table 4. Two of these previously reported experimental values are within $\lesssim$ 1 percent of the results of the present measurements and the third differs by $\approx$ 3 percent. It was concluded that the present experiments resulted in total cross sections of carbon at 245 keV within $\lesssim$ 1 percent of the widely accepted value and that such precisions could reasonably be expected from the thinner ^{6}Li samples excepting uncertainties inherent to the ^{6}Li sample itself.

The ^{6}Li energy scales were established from the three sets of measurements near 7 m and the three near 14 m so arranged that the gamma-ray flash was very near the ^{6}Li resonance peak thus avoiding any appreciable dependance on the calibration of the time analyzers. Representative cross sections determined from such measurements are shown in Figs. 1 and 2. The latter figure is representative of results obtained with flight paths adjusted to place the gamma-flash very near the resonance peak (in this example a flight path of 699.96 cm and a pulse repetition period of 1004 nsec). The equivalent calculated-relativistic neutron energy is 245.1 keV. Inspection of Fig. 2 suggests that the gamma-ray equivalent neutron energy is slightly above the observed resonance energy. A number of numerical fitting procedures for the quantitative location of the energy of the resonanc maximum were examined including least-square fits of various order polynomials, logarithmic expansions and an R-matrix resonance description. Each method included a degree of subjective judgment that tended to be obscured by various numerical artifaces. Of these a simple quadratic power series limited to measured values near the resonance maximum gave the most consistent results and was chosen as the numerical fitting procedure. An alternate approach consisted of a linear segment fitted to either side of the resonance at $\approx$ 60, 70 and 80 percent of the peak height from which the peak center at each of the fractional heights was determined. The center values skewed slightly due to the asymmetry of the resonance but were smoothly extrapolated to the center value at full maximum with little uncertainty. Care was taken to avoid perturbations due to the gamma-ray flash. The above procedure was carried out independently by two scientiest with results generally agreeing to within $\approx$ 0.5 keV. In addition, the results obtained using the two procedures were consistent to within $\lesssim$ 1 keV. The results obtained at $\approx$ 7 m and $\approx$ 14 m are summarized in Table 5. The values of each set are consistent. The "Best Estimates" from the two sets are 242.6 ± 2.0 and 245.45 ± 1.0 keV. This value is compared with a number of previously reported results in Table 5. In a review of selected previously reported values, James quotes an average of 244 ± 0.5 keV (4). Obviously, the James average value is essentially identical to the weighted average of the present results. It is evident in Table 3 that uncertainties associated with the ^{6}Li resonance maximum are a major contribution to the overall uncertainty. The consistancy of the instrumental energy scales is generally much better than the definition of the resonance maximum. Thus it is probable that any significant improvements in the overall result will have to give primary attention to the definition of the resonance peak. That will be a tedious procedure and, possibly, not very rewarding.

The maximum cross section of the ^6Li resonance was determined using a shorter flight path of 422.1 cm which put the gamma-ray flash well away from the area of interest. Both BCMN and ANL ^6Li samples, as defined in Table 1, were used. The former was more precisely fabricated but relatively thick and, as a consequence, "dark gray" at the resonance maximum. The ANL sample was approximately 1/3 as thick thus more suitable at the resonance maximum but too thin for good statistical accuracies well off the resonance. A second ANL sample was also used but its density was suspect and thus the results were abandoned. Concurrently with the ^6Li measurements the iron, carbon and aluminum cross sections were determined with the samples of Table 1. Carbon results were consistent with previously reported values such as illustrated in Table 4 and discussed above. At this shorter flight path the gamma-ray calibration technique becomes more sensitive to the precise calibration of the time scales of the measuring equipment. Thus the initial energy scale was determined from the iron resonance values using the resonance energies of Table 2 which were determined in the present experiments at more optimum flight paths. Two runs were made to good statistical accuracies using somewhat different time-scale settings of the instruments. The cross section results were corrected for instrumental resolution (very small correction) and ^7Li sample content using ^7Li values as given by ENDF/B-IV.

Several alternate methods of background interpretation were used with essentially no effect on the resulting cross sections. This was not surprising as backgrounds were relatively small and very well determined on either side of the relatively narrow energy range of interest. The resulting cross sections obtained using both samples are shown in Figs. 3

and 4 differ in energy scale by about 1.0 keV. That shift is well within the resolution of the apparatus at this shorter flight. The actual resolution was estimated from the observed gamma-ray resolution and from comparisons of the observed iron cross sections with progressive averages of the much higher resolution iron values given in ENDF/B-IV. The latter method indicated resolutions of $\gtrsim$ 2 keV, of course, varying slightly with flight path and energy. A 4 keV average of the ENDF/B-IV iron results was very clearly a much broader resolution than observed experimentally at the shortest flight paths. The final cross section values were obtained by shifting the above results by small amounts so as to make the resonance peak energy coincident with the weighted average of "Best Estimates" given in Table 5. The shift was $\gtrsim$ 1.0 keV. These final results are shown in Figs. 5 and 6.

The maximum cross section values were determined from the results of Figs. 3 and 4 by averaging subjectively selected measured values near the extremum. The result of 11.2 ± 0.20 b is very consistent with the ENDF/B-IV value of 11.259 b. The present value is slightly larger than the 10.85 ± 0.10 given by Harvey and Hill (12), but the latter value may have been from a data set that still contained a very small percentage of ^{7}Li. Corrected data received from these authors gives a real value of 11.0 b; thus the present results and those of Ref. 12 appear consistent. The recent results of Knitter et al. (14) indicate a maximum value of approximately 11.15 b, very near the present result.

The resonance shapes of the present measurements are compared with those of Harvey and Hill (12) and Knitter et al. (14) in Figs. 5 and 6, respectively. The present results compare very favorably with those of Harvey and Hill (12). Such differences as may exist are well within the accuracies of the present measurements alone. The present results differ in energy scale from those of Ref. 14, as expected, since the latter are systematically somewhat higher in energy than the values of Ref. 12. The relative resonance shapes of the present work and that of Ref. 14 are similar. The primary difference is in energy scale but even there the two results are consistent within the respective energy uncertainties.

V. CONCLUDING COMMENT

The present results, obtained using a vernier method that is free from some uncertainties associated with other techniques, indicate that the observed maximum of the ^{6}Li resonance is at 244.5 ± 1 keV. Concurrent measurements of well defined resonances in iron in the energy range $\approx$ 150 to 300 keV result in an energy scale agreeing to within better than 1 keV with that given by ENDF/B-IV. Many of the possible systematic sources of energy error should be reflected in both iron and ^{6}Li measurements. The peak cross section value at the resonance was observed to be 11.2 ± 0.20 b. This value is contingent upon a knowledge of sample density and geometric factors to within $\approx$ 1 percent. This could not be verified without destructive assay of the samples. However, the two samples used are of very different size and come from completely independent sources.

ACKNOWLEDGEMENTS

The authors are indebted to Dr. J. Harvey for pointing out the importance of corrections for multiple-neutron-scattering events in the detector and to Dr. W. Poenitz for the calculation of the appropriate correction factors.

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TABLE 1. Sample Specifications

Fe	Diameter	2.54 cm
	Length	2.00 cm
	Atoms/cm^2	1.7029 × 10^{23}
C	Diameter	2.54 cm
	Length	3.00 cm
	Atoms/cm^2	2.358 (± 0.015) × 10^{23}
Aℓ	Diameter	2.00 cm
	Length	2.00 cm
	Atoms/cm^2	1.061 × 10^{23}
^{6}Li (BCMN)[a]	Diameter	3.504 cm
	Length	5.538 cm
	Atoms/cm^2	2.563 (≤ 0.01) × 10^{23}[b]
^{6}Li (ANL)[a]	Diameter	2.0 cm
	Length	5.538 cm
	Atoms/cm^2	0.8733 (± ≤ 0.008) × 10^{23}[b]

a. All "^{6}Li" samples were assumed to consist of 95.5% ^{6}Li and 4.5% ^{7}Li.

b. More realistic uncertainties may be 1%. The samples could not be destructively arrayed for verification.

TABLE 2. Observed Energies of Iron Resonances

	Resonances (keV)				
	1	2	3	4	5
A. ∿ 7 m Flight Paths					
Flight Path					
650.27 cm	169.7	188.8	222.2	246.7	278.7
699.96 cm	170.7	188.9	221.7	245.9	277.7
750.13 cm	171.7	190.4	222.8	246.7	278.5
Ave.	170.7	189.4	222.2	246.4	278.3
/Ave.-Deviation/	0.7	0.7	0.4	0.4	0.4
B. ∿ 14 m Flight Paths					
Flight Path					
1381.65 cm	-	-	-	245.7	-
1412.76 cm	-	-	-	246.2	-
1350.53 cm	-	-	-	246.0	-
Ave.	-	-	-	245.96	-
/Ave.-Deviation/	-	-	-	0.22	-
ENDF/B-IV (2)	169.4	188.5	220.8	245.0	278.1
Ave.—ENDF (keV)	+1.3	+0.9	+1.4	+1.4 or +0.96	+0.2

TABLE 3. Known Sources of Uncertainty

Source	Uncertainty	Contribution at 7 m	Contribution at 14 m
Flight Path	$\leq$ 0.5 cm	$\leq$ 0.36 keV	$\leq$ 0.18 keV
Frequency	$\lesssim$ 0.01 kHz	Negligible	
Time-Analyzer Calibration	< 1.0%	< 0.5 keV	< 0.25 keV
Gamma-ray Position	$\sim$ 2.0 nsec	$\sim$ 1.0 keV	$\sim$ 0.5 keV
Velocity of Light		Negligible	
Cumulative Energy-Scale Uncertainties		1.17 keV	0.75 keV
Uncertainties Associate with the the Location of the Maximum of the ^{6}Li Resonance		1.6 keV	0.7 keV
Combined Uncertainties Associated with the ^{6}Li Resonance		$\approx$ 2.0 keV	$\approx$ 1.0 keV

TABLE 4. Illustrative Neutron Total Cross Sections of Carbon Averaged Over the Interval 240 to 250 keV

Flight Path (cm)[b]	$\overline{E}$(keV)	$\overline{\sigma}$(b)
449.93	245.0	3.920
650.27	245.1	3.883
750.13	245.0	3.913
	AVE =	3.905 (at 245 keV)
	|AVE-DEV.| =	0.007

ENDF-IV (2)	=	3.945 (1%)[a]
Whalen et al. (5)	=	4.037 (3%)
Perey et al. (6)	=	3.921 (0.5%)
Uttley et al. (7)	=	3.947 (1%)

a. Indicated percentage variation from average of present results.

b. Flight-paths are significant to only 0.1 cm.

TABLE 5. Observed Energy of the ^{6}Li Resonance Maximum

	Samples	
	BCMN	ANL
A. ≈ 7 m Flight Paths		
750.13 cm[a]	244.2 keV	244.7 keV
650.27 cm	241.7	242.5
699.96 cm	240.7	241.5
Average	242.2	242.9
RMS Dev. From Ave.	1.5	1.3
"Best Estimate"	242.6 ± 2.0 keV	
B. ≈ 14 m Flight Paths		
1381.65 cm	245.2 keV	245.1 keV
1412.76 cm	246.1	246.2
1350.53 cm	243.9	246.1
Average	245.1	245.8
RMS Dev. From Ave.	0.9	0.5
"Best Estimate"	245.45 ± 1. keV	
C. Weighted Averages of "Best Estimates"	244.5 ± 1. keV	
D. Other Comparable Results Reported in the Literature		
Hibdon and Mooring (8)	246 keV	
Meadows and Whalen (9)	252.5	
Uttley (10)	243.5 ± 1	
James et al. (11)	242.71 ± 0.33	
Harvey et al. (12)	246 ± 1	
Böckhoff et al. (13)	245 ± 1	
Knitter et al. (14)	247 ± 3	
ENDF/B-IV (MAT-101) (2)	246	

a. Flight paths are significant to only 0.1 cm.

FIGURE CAPTIONS

Fig. 1. Illustrative ^{6}Li total cross sections observed at a flight path of 650.3 cm. Gamma-flash location is indicated. Gamma-ray period = 1004 nsec.

(ANL Neg. No. 116-77-12)

Fig. 2. Illustrative ^{6}Li total cross sections observed at a flight path of 670.0 cm. Gamma-flash is clearly evident. Gamma-ray period = 1004 nsec.

(ANL Neg. No. 116-77-9)

Fig. 3. First ^{6}Li total cross section set obtained at a flight path of 422.1 cm. ○ = thick sample results, + = thin sample results as specified in Table 1.

(ANL Neg. No. 116-77-14)

Fig. 4. Second ^{6}Li total cross section set obtained at a flight path of 422.1 cm. Symbolism is identical to that of Fig. 3.

(ANL Neg. No. 116-77-13)

Fig. 5. ^{6}Li total cross section results compared with the values of Harvey and Hill (12). ○ and + have the same meaning as in Fig. 4, △ denote values of ref. 6.

(ANL Neg. No. 116-77-452)

Fig. 6. ^{6}Li total cross section results compared with the values of Knitter et al. (14). ○ and + have the same meaning as in Fig. 4, □ denote values of ref. 14.

(ANL Neg. No. 116-77-453)

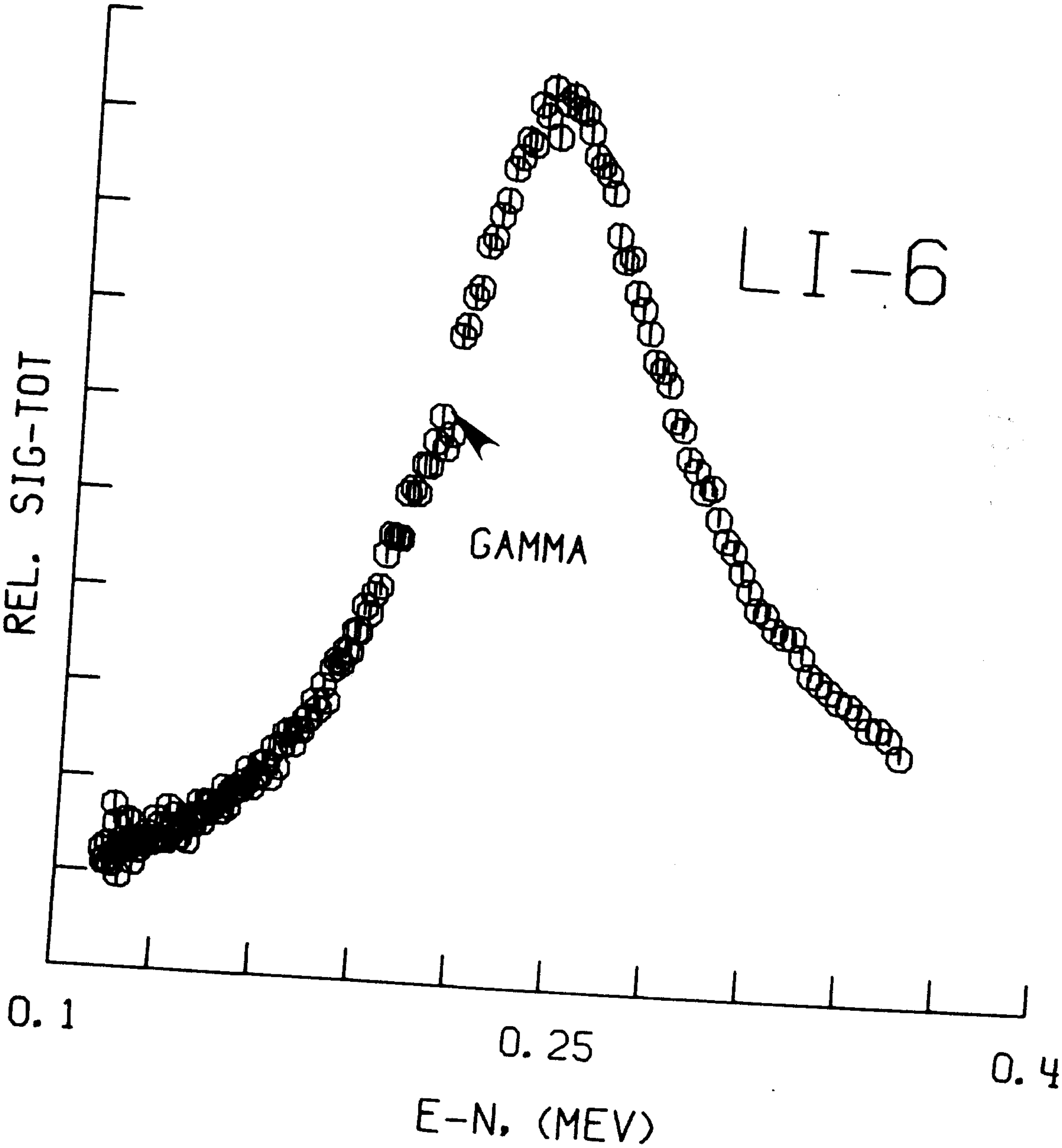

Fig.1

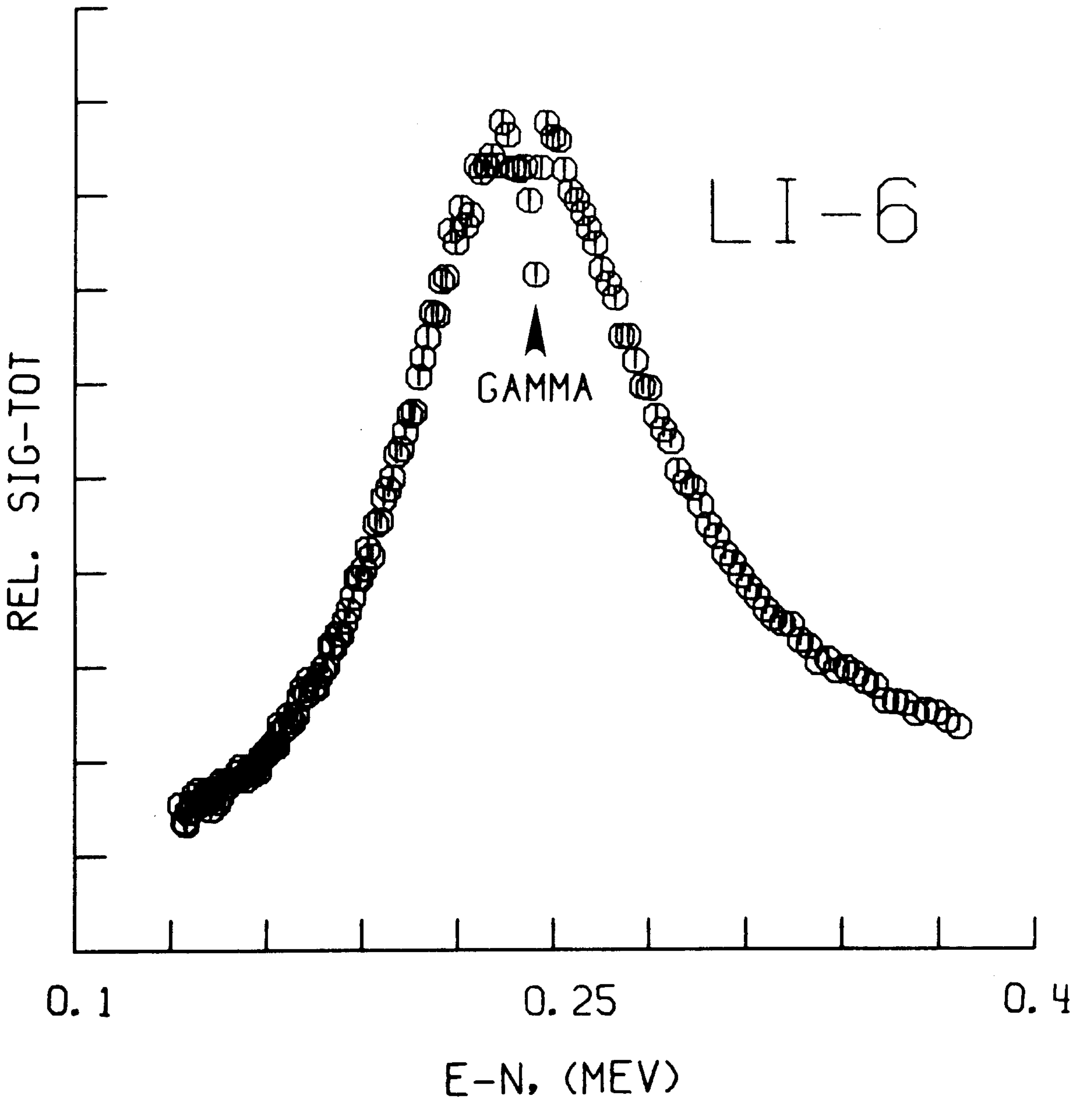

Fig. 2

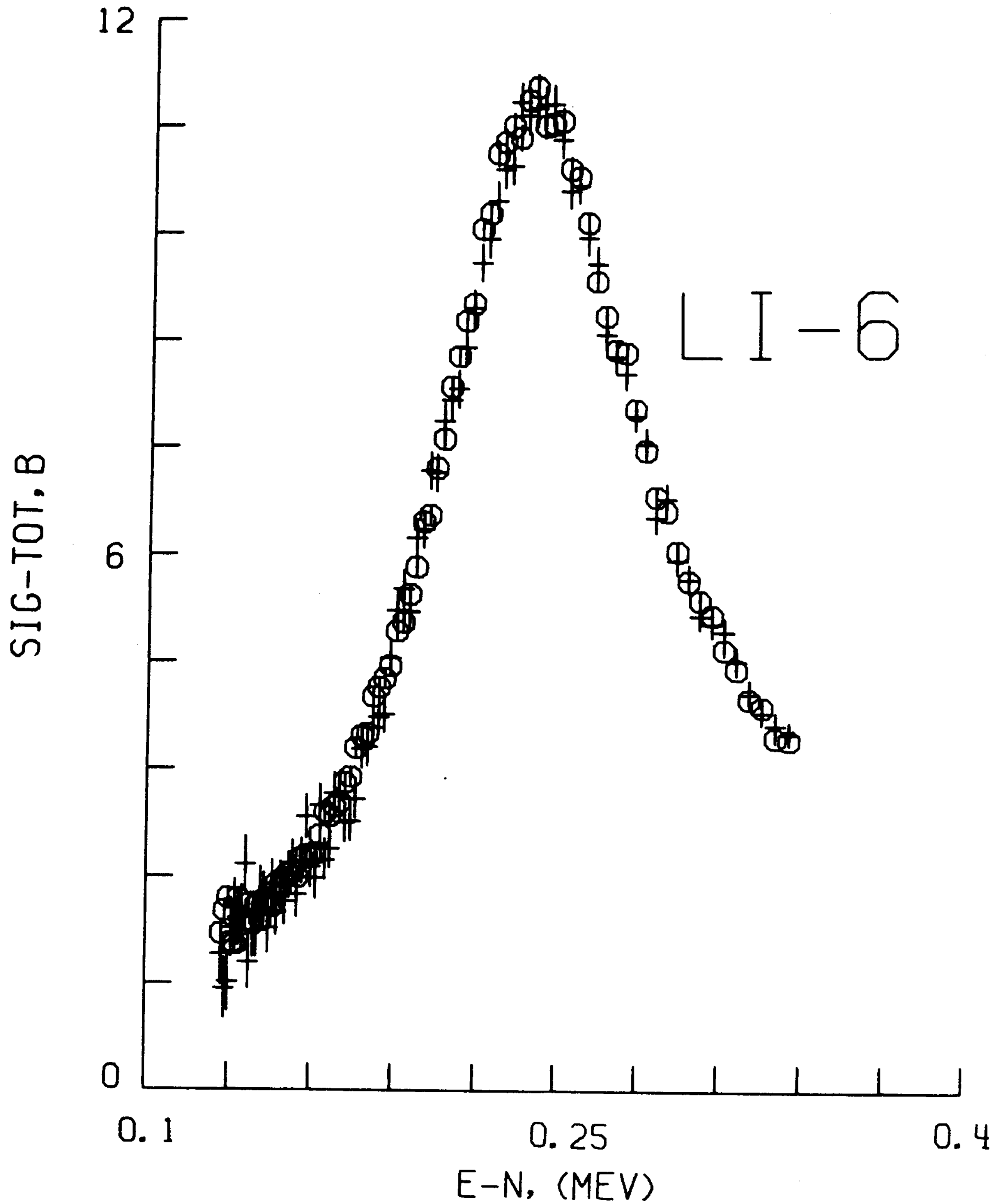

Fig. 3

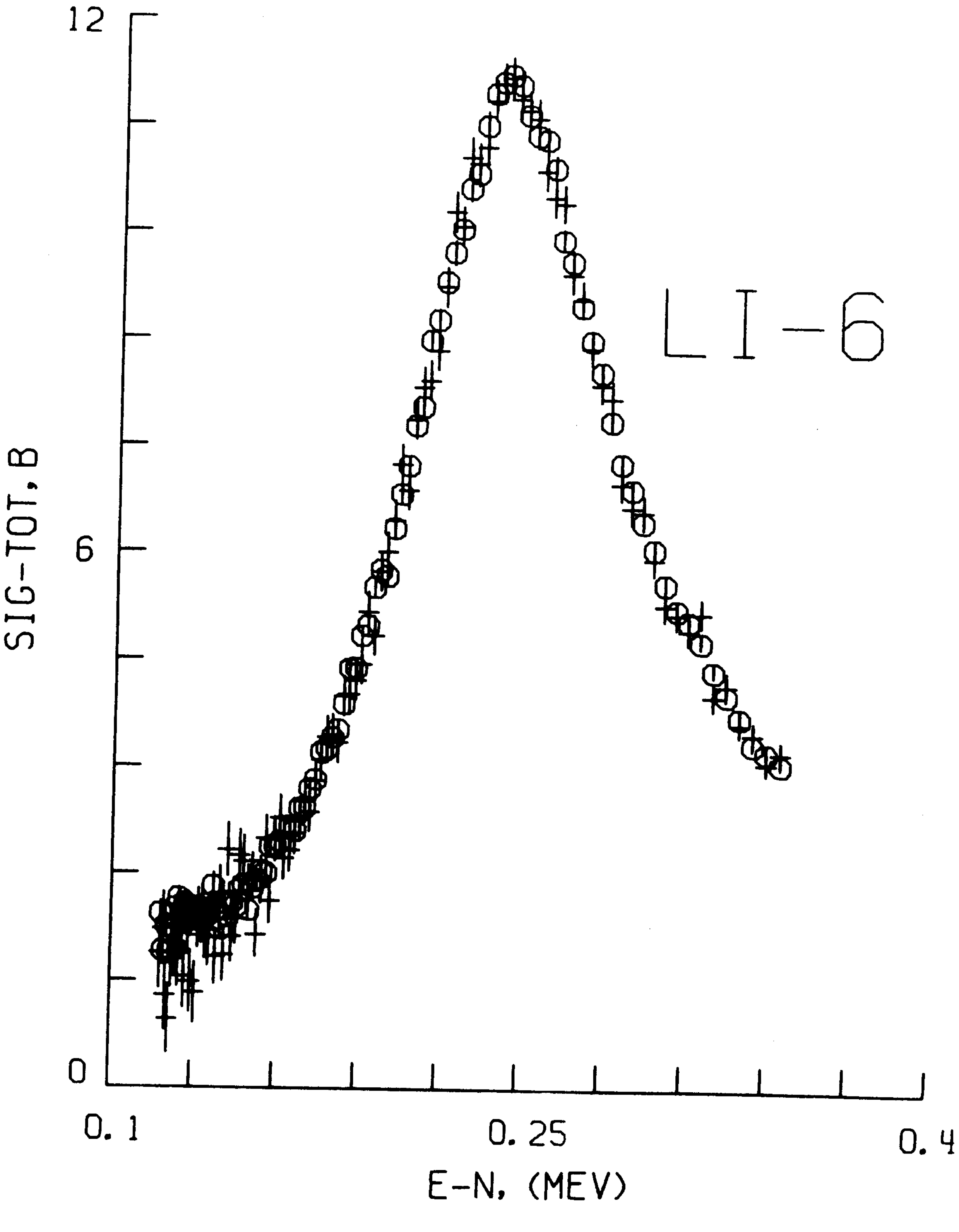

Fig. 4

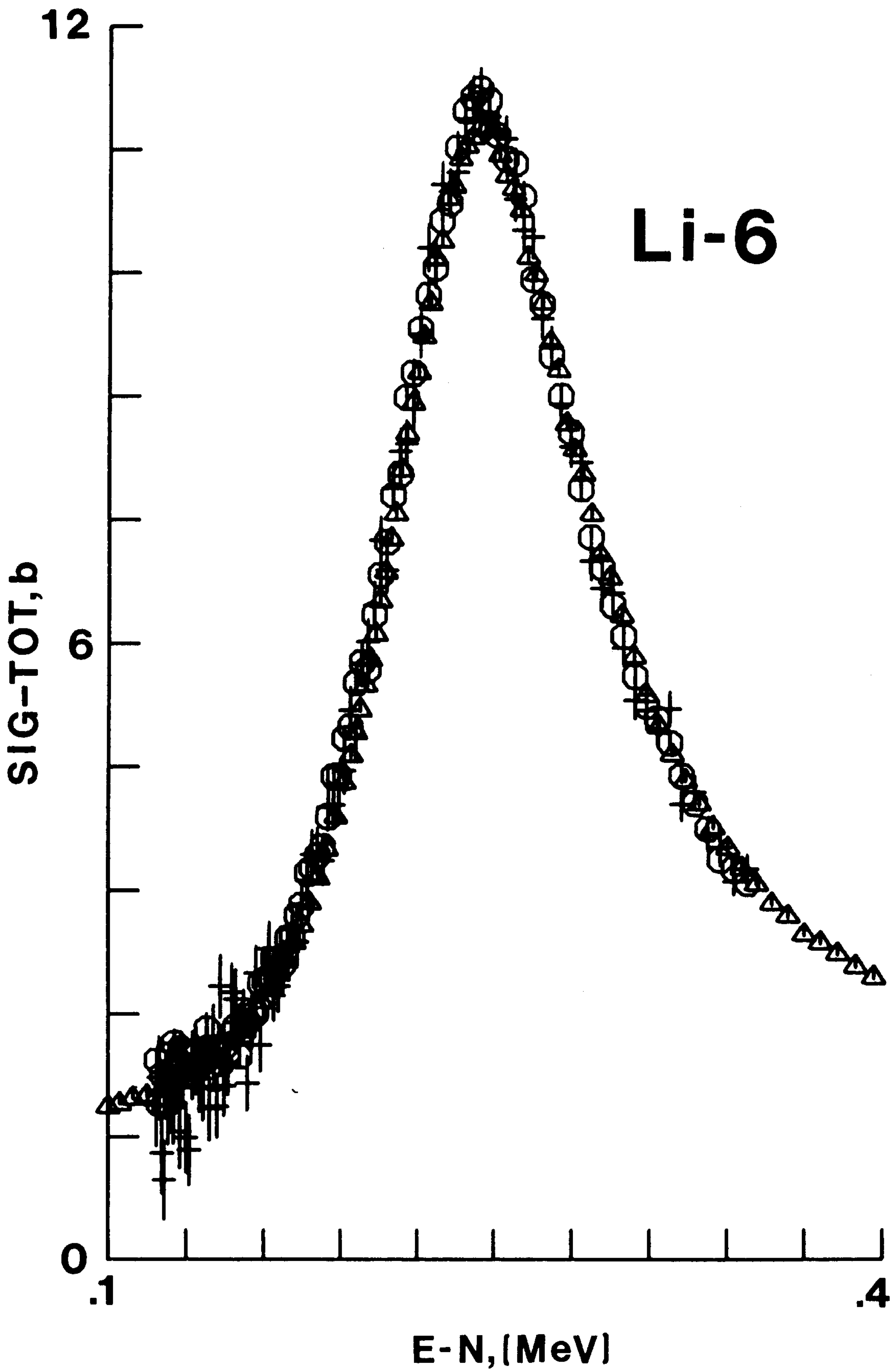

Fig. 5.

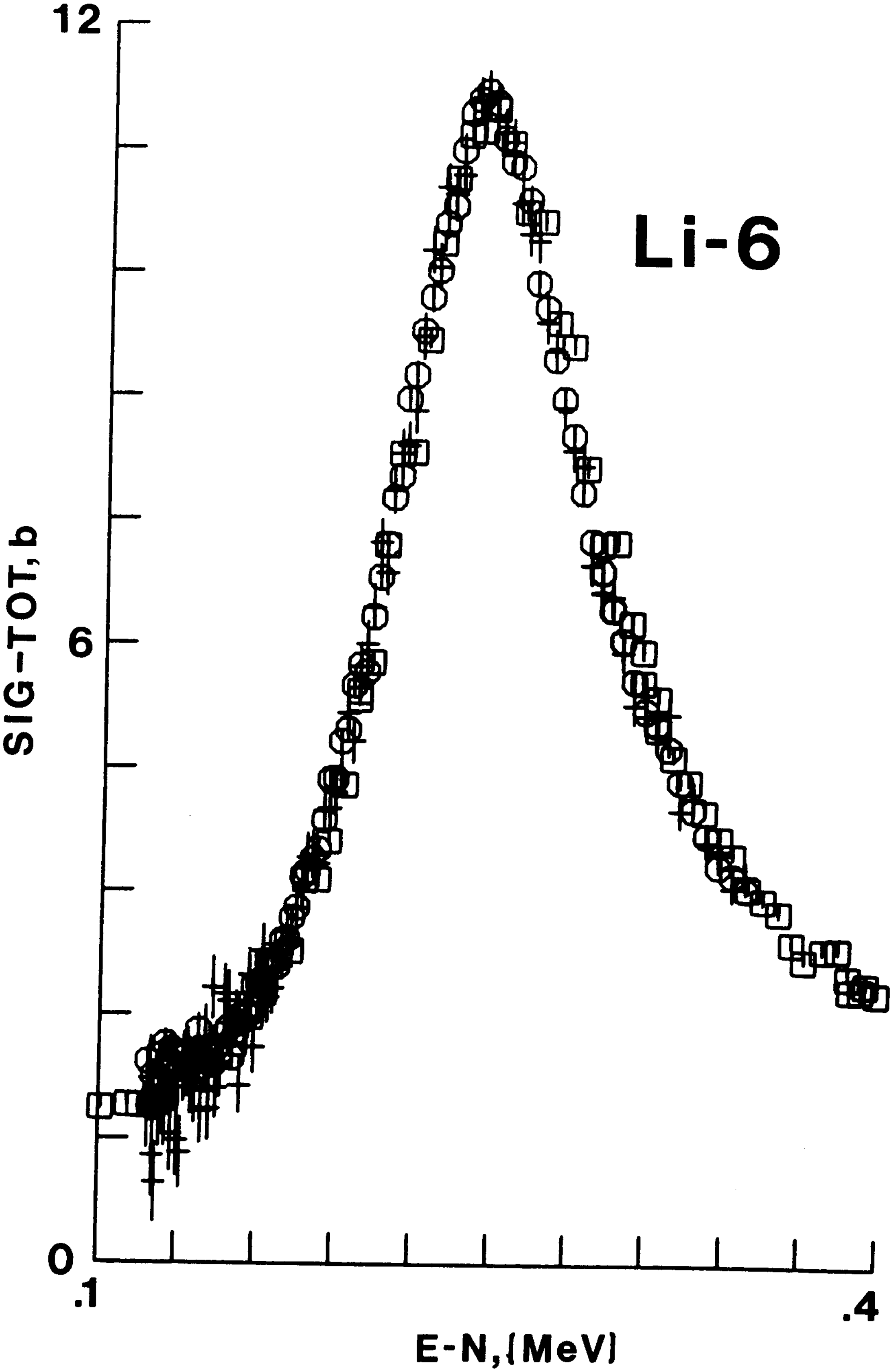

Fig. 6.