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Activation and Angular Distribution Measurements of $^7Li(p,n)^7Be(0.0 + 0.429$ MeV) for $E_p = 25$ to 45 MeV: A Technique for Absolute Neutron Yield Determination*

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Angular distributions of the combined $^7Li(p,n)^7Be$ reactions to the ground and first excited state of beryllium have been measured at proton energies of 24.8, 35.0, and 45.0 MeV with a typical accuracy of 5%. The total cross section was also obtained at eleven energies between 24 and 45 MeV by activation techniques. The reaction is a potentially convenient source for neutrons, and absolute neutron fluence can be determined independent of beam current and target thickness measurement if the amount of ^7Be ($\tau_{1/2} = 53.4d$) produced in the reaction is measured. Following the proton irradiation the amount of ^7Be produced is determined by observation of the 0.478 MeV gamma rays from a 10.4% branch of ^7Be and this information is combined with the total cross sections and angular distributions to give the neutron fluence at the time of irradiation.

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I. Introduction

The (p,n) reaction on ^7Li to the ground and first excited state of beryllium, $^7Li(p,n)^7Be$ (0.0 + 0.429 MeV), provides a potentially convenient source of 20-50 MeV neutrons for neutron scattering experiments and for efficiency calibration of neutron detectors. The cross sections are large and the emitted neutrons are well separated from lower-energy neutrons. Lithium is a ductile metal that is moderately convenient to handle as a target material. The ^7Li nucleus is light and there is a substantial kinematic shift with angle. This feature provides a range of neutron energies for a single proton energy and can be useful for neutron detector calibration.

Precise measurements of the $^7Li(p,n)^7Be$ (0.0 + 0.429 MeV) reaction are available up to 26 MeV)[1,2] but at higher energies measurements are more limited)[3,4] in both completeness and accuracy. We report measurements of the combined reactions, ground-state plus first-excited-state (0.429 MeV), in the proton energy range 25 to 45 MeV. Total cross sections for the combined reactions were obtained by an activation technique and were used for absolute normalization of angular distributions taken at 25, 35, and 45 MeV using neutron time-of-flight techniques.

The activation method takes advantage of the fact that the ground and 0.429-MeV states of ^7Be are the only particle-emission-stable states of ^7Be, so that the activation cross section for the production of ^7Be is due exclusively to reactions leading to these states. The 53.4-day half life of ^7Be is long enough that targets can be counted at a convenient time after activation. The production cross section can be measured by observing the 0.478-MeV gamma emission in ^7Li that follows the decay of ^7Be with a branching ratio of 10.4% ± 0.1%.

The availability of these cross sections permits simple measurements of the absolute neutron fluence from these reactions with better than 5% accuracy.

No measurements of beam currents or target thicknesses are required if gamma decay measurements of ^{7}Be are made.

II. Experimental Method

Angular distributions were obtained with the new beam swinger neutron time-of-flight facility at the Michigan State University Cyclotron Laboratory)[5]. This system uses rotating magnets to change the angle of the incident beam while the position of the detector remains fixed. Flight paths up to 32 meters are available.

The angular distributions were obtained with a 7.0-cm-diameter by 3.8-cm-thick glass-encapsulated scintillation detector containing NE213 liquid scintillator placed at flight paths of 4 to 5 meters. Energy thresholds were set with respect to the Compton edge of 2.61-MeV gamma rays from a ^{228}Th source and were 1.0, 2.0, and 3.0 times the Compton-edge energy for bombarding energies of 25, 35, and 45 MeV respectively. Pulse shape discrimination)[6] was employed to eliminate events caused by gamma rays and alpha particles. A NaI proton detector placed at a lab angle of 90° served as a monitor.

An updated version of R.J. Kurz's scintillation efficiency code)[7] was used to determine the detector efficiency. The Michigan State University version of this code employs a light curve measured at this laboratory for NE213 liquid scintillator. Edge-effect corrections were done separately following the procedure of Schuttler)[8]. The H(n,n) cross sections are well known and, as noted above, reactions on carbon leading to final-state alpha particles were eliminated by the pulse shape analysis. The cross sections of n+C reactions that lead to final-state protons are not as well known and must be considered with care. We have chosen to follow the recommendations of McNaughton et al.)[9], with the following modification. Rather than employing the -15.96-MeV Q-value of the ^{12}C(n,np)^{11}B reaction we have used the -12.59-MeV ^{12}C(n,p)^{12}B Q-value, smoothing the rise of the cross section with energy to compensate for the fact that the ^{12}C(n,np)^{11}B reaction contributes to the cross section for final state protons only above its threshold.

The relative efficiency calculation was checked by several methods. First, after re-introducing the n+^{12}C cross sections that produce alpha particles, the code was used to predict the experimentally calibrated efficiencies of the scintillation detector described by McNaughton)[9]. As a consistency check, angular distributions of the ^{7}Li(p,n)^{7}Be reaction taken at the same proton energy but at different detector light thresholds were compared. Finally, the absolute value of the integrated angular distributions obtained with the code were compared to the total cross sections obtained from the activation technique. These comparisons agreed within their respective experimental errors.

Activation experiments were carried out in the bottom of a Faraday cup of 1.3-cm diameter and 2.0-cm depth. A 0.6-cm-diameter collimator was placed 0.1 cm before the Faraday cup to ensure proper alignment of the beam and to monitor secondary electrons backscattered from the target. The charge collection of this arrangement was checked with a precision Faraday cup which could be inserted before the collimator with a pneumatic plunger. The current integrator was checked with reference current sources. Beam currents and current densities depended upon target thickness, target mounting technique, and beam energy, and were as low as 20 nAmp and 250 nAmp/cm^{2} respectively to minimize target heating. Aluminum foils 8 mg/cm^{2} thick were placed after each lithium target to stop any ^{7}Be nuclei recoiling from the Lithium target.

Lithium targets were activated both singly and in multiple-target stacks. The multiple lithium targets were separated by energy degrading shims and were placed in the Faraday cup in series for simultaneous irradiation to insure precise relative measurement of the activation cross sections. This procedure enabled targets of matched thicknesses to be irradiated at the same time and insured the same flux on each target (neglecting reactions in the target, which were less than a 1% effect for the present case). To remove dependence on theoretical stopping powers the mean proton energy in each of these targets was determined by observing the neutron time-of-flight spectrum at 0°. Figure 1 shows such a spectrum for a proton energy of 45 MeV. The neutron peaks from the five ^{7}Li targets in the stack are clearly resolved. Broadening of the lower-energy neutron groups is predominantly due to the non-linearity of time-of-flight as a function of energy, with an additional contribution from straggling.

Activated targets and catcher foils were counted with Ge(Li) detectors using United States National Bureau of Standards and International Atomic Energy Commission sources as efficiency references. The half life and branching ratios for ^{7}Be decay were taken from Ajzenberg-Selove and Lauritsen)[10].

Thick Li targets were made by pressing 99.99% enriched ^{7}Li metal with precisely machined stamps and molds. Thicknesses, measured by weighing, were in the range 20 to 60 mg/cm^2. Areas were determined from the diameter of the circular stamps used to cut the final target from the pressed sample. Secondary thickness measurements were done directly with a micrometer. Uniformity of thickness was checked by weighing small sections of targets removed with a small diameter tubular cutter as well as by repeatability of activation measurements of separate targets done under similar conditions. These measurements indicated that target thickness could be reproduced within ± 1%. All lithium targets were handled and stored under oil to minimize chemical reaction with the atmosphere. Measurements of the mass of targets before and after an experiment showed that no significant chemical reaction had taken place.

III. Results

Figure 2 shows the angular distributions at proton energies of 24.8, 35.0

and 45.0 MeV. The normalization has been determined by the activation measurements. For convenience, we also show cross sections from a separate high-resolution experiment)[11] which provided ratios of the cross sections for the 0.0- and 0.429-MeV states. Table 1 lists the combined (0.0 + 0.429 MeV) differential cross sections. Relative errors reflect contributions of counting statistics, uncertainties in the energy dependence of the neutron detector efficiency, uncertainties in the neutron attenuation corrections, and uncertainties in the scattering angle. The normalization error for the cross sections in Table 1 is an additional ± 3.6%. The ratios of the integrated cross sections for the excited ground states are 0.35 ± 0.01, 0.41 ± 0.01, 0.455 ± 0.01 for proton energies of 24.8, 35.0 and 45.0 MeV.

Total cross sections for the combined reactions obtained from the activation experiments are given in Figure 3 and Table 2. The relative error for each point is ± 2.0%. The major sources contributing to this error are counting statistics, uncertainties in beam energy and solid angle of gamma counting, change in beam flux due to reactions in the target stack, and random variations in target thicknesses. An additional systematic error of ± 3.0% in the absolute cross sections is due to the efficiency calibration of the Ge(Li) detector, target thickness, and the assumed shapes of the gamma peak and background.

IV. Comparison with other Measurements

The results of the zero-degree cross sections of the combined reactions for energies up to 100 MeV are shown in Figure 4. The measurements are in general agreement, the cross sections obtained in the present experiments being the most accurate available in the energy range above 25 MeV. Locard et al.)[12] measured the ^{7}Li(p,n)^{7}Be (0.429) total cross section between 20 and 55 MeV by observing the emitted 0.429 gamma decay during proton bombardment. Their results agree with the integrated cross sections for the excited state angular distributions at 25, 35, and 45 MeV shown in Figure 2.

The recent measurements of Poppe et al.)[2] agree within quoted errors with ours for the total cross sections of the ground and excited state reactions at 25 MeV. However, a comparison of the angular distributions for the individual reactions at 25 MeV shows a slight but systematic difference at back angles, with our results higher at $\theta > 45°$. A 15-20% disagreement is typical. The use of repeated measurements and a proton monitor in our work precludes systematic effects from target deterioration. Target deterioration is also unlikely to affect the measurements of Poppe et al. since many angles are taken simultaneously in their experiment. By comparing the detector efficiency used)[18] in the Livermore experiment)[2] with a calculation of the efficiency of their detector based on our code we can account for about 5% of the observed differences, but the remaining discrepancy remains unexplained.

V. Conclusions

The combination of a large 0° cross section and relatively convenient target preparation makes ^{7}Li(p,n)^{7}Be the source reaction of choice for many experiments in the neutron energy range above 20 MeV. However, there are limitations that must be borne in mind. The most important of these limitations are that source energy resolution is restricted to about 400 keV by the contribution from the 0.429-MeV state and that the source spectrum contains secondary structure. The three body continuum begins, in priniciple, at an equivalent excitation energy of 1.59 MeV, but this contribution is typically rather small. We are not aware of detailed careful studies of this point, but for example, at E_p = 28.0 MeV and θ = 0°, it is found to be less than 0.5%/MeV of the main source peak in our spectra. The first important secondary peak occurs at E_x = 4.57 MeV, the position of the second excited state of ^{7}Be.

The present results permit a prediction of fluxes from ^{7}Li(p,n)^{7}Be(0.0 + 0.429) in the E_n = 20 - 45 MeV range with an accuracy of about 5%, assuming that beam current and target thickness can be accurately measured. In addition, they allow one to determine the absolute total neutron fluence independent of current and target thickness measurements as follows. A fresh ^{7}Li target is used to produce neutrons at an angle θ. The number n_7 of ^{7}Be nuclei produced during the bombardment via the reaction ^{7}Li(p,n)^{7}Be(0.0 + 0.429 MeV) is then determined after irradiation by observing the 0.478-MeV gamma ray as was done in the present experiment. The neutron fluence F at a distance d from the target can then be obtained from the cross sections tabulated in Tables 1 and 2 according to:

$$F = \frac{n_7 \frac{d\sigma(\theta)}{d\Omega}}{d^2 \sigma_t}$$

Recently Brandenburger et al.)[19] have suggested another technique for determining the absolute flux form ^{7}Li(p,n)^{7}Be(0.429). This technique involves the determination with a gamma detector of the number of 0.429 MeV gamma rays from deexcitation of the first excited state of ^{7}Be at the time of irradiation. Since these gamma rays are distributed isotropically (in the rest frame of the recoiling ^{7}Be nucleus), such a measurement yields the number of formation of ^{7}Be(0.429), and if $\frac{d\sigma}{d\Omega}(\theta)$ is known for ^{7}Li(p,n)^{7}Be(0.429 MeV) it leads to an absolute measurement of flux. The procedure works well at low energies, but has certain disadvantages compared to the techniques suggested here at higher energies. The gamma rays will have a large Doppler broadening)[12] making accurate background subtractions difficult and inconveniently long flight paths of 20 meters or more may be required to resolve neutrons from the ground and first excited states.

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Table 1

θ_{lab} (deg)	$\frac{d\sigma}{d\Omega}\left(\frac{mb}{sr}\right) \pm \Delta$ (%)* 24.8 MeV		35.0 MeV		45.0 MeV	
0.0	25.4	4.9	29.1	2.9	31.6	3.0
10.0	19.2	3.9	25.0	2.7	24.6	2.7
15.0	14.2	3.8	17.2	2.7	17.0	2.9
20.0	9.47	3.5	11.5	2.7	10.7	3.0
25.0	6.74	2.9	7.60	2.7	6.47	2.8
30.0	5.29	2.3	5.25	2.6	4.42	2.2
35.0	4.82	2.0	3.83	1.7	3.27	2.8
40.0	4.85	5.5	3.31	1.9	2.48	2.6
45.0	4.64	1.4	2.84	1.8	1.98	2.4
50.0	4.45	1.4	2.56	2.9	1.51	4.5
55.0	4.00	1.7	2.00	2.1	1.22	3.1
60.0	3.31	5.4	1.55	2.4	0.829	3.0
65.0	2.73	5.5	1.10	2.4	0.531	5.8
70.0	2.03	5.6	0.862	2.7	0.414	6.0
75.0	1.60	5.7	0.653	2.9		
80.0	1.27	5.9	0.486	2.9	0.192	6.2
85.0	1.06	6.0	0.413	3.0		
90.0	1.02	6.0	0.330	3.3	0.122	6.4
95.0	1.00	6.1	0.269	3.5		
100.0	1.01	6.3	0.233	3.8	0.0727	7.7
110.0	0.939	6.8	0.174	4.0	0.0405	8.7
120.0	0.802	7.4	0.125	4.6	0.0358	9.9
130.0	0.490	8.0	0.0856	4.8		
140.0	0.391	8.5	0.0738	5.3	0.0266	8.3
150.0	0.382	9.1	0.0925	5.6	0.0327	9.2

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* Relative error is listed. Normalization error is an additional 3.6%.

Table 2

Proton Energy	Total Cross Section* $^{7}Li(p,n)^{7}Be$ (0 + 0.429)
24.7 ± 0.2 MeV	29.19 mb
27.3 ± 0.2	26.06
29.4 ± 0.2	24.03
29.9 ± 0.2	24.12
31.9 ± 0.2	22.48
34.5 ± 0.2	20.84
34.9 ± 0.1	20.53
36.6 ± 0.2	19.46
39.5 ± 0.2	18.03
40.7 ± 0.1	17.43
44.7 ± 0.1	16.44

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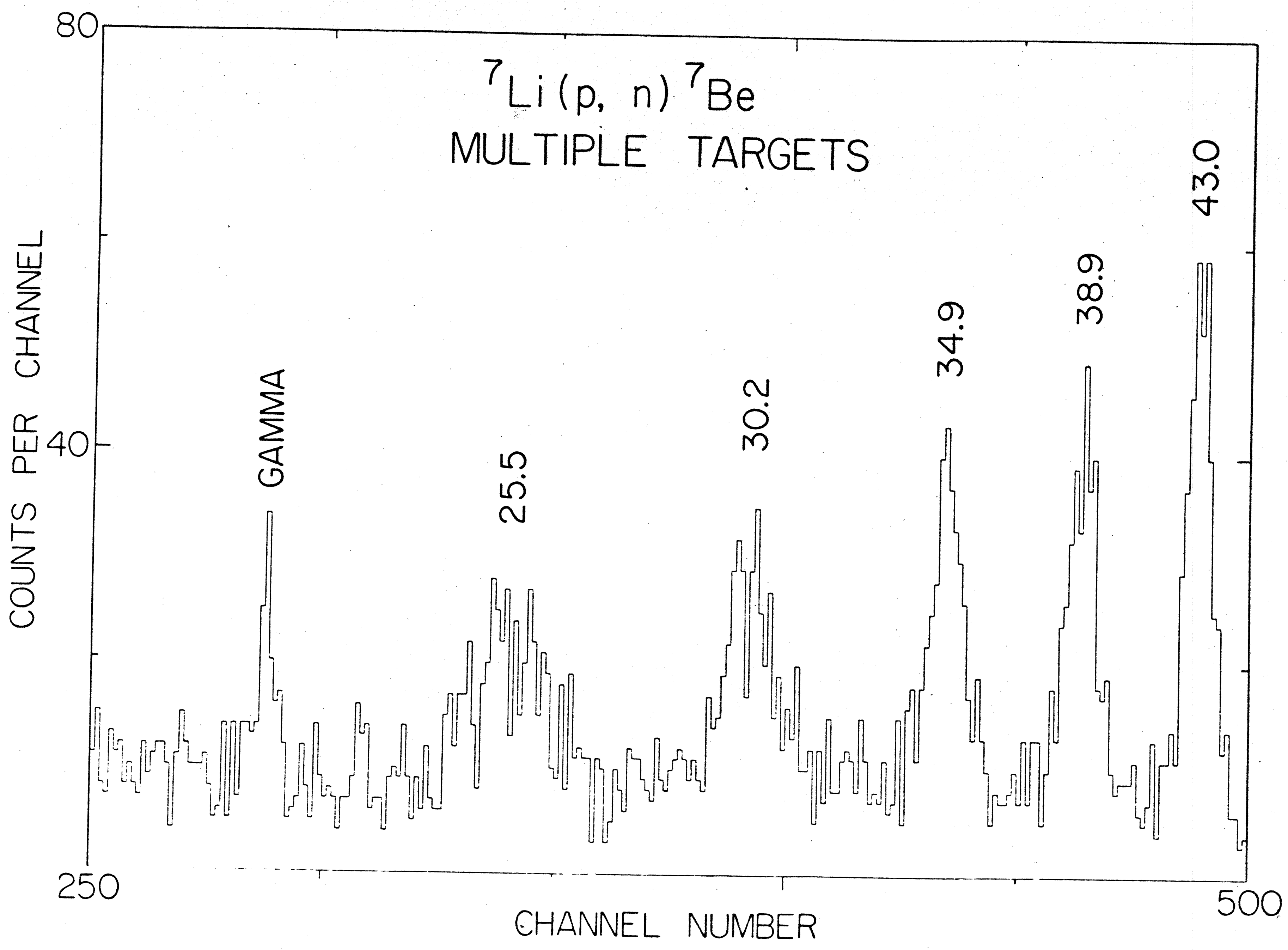
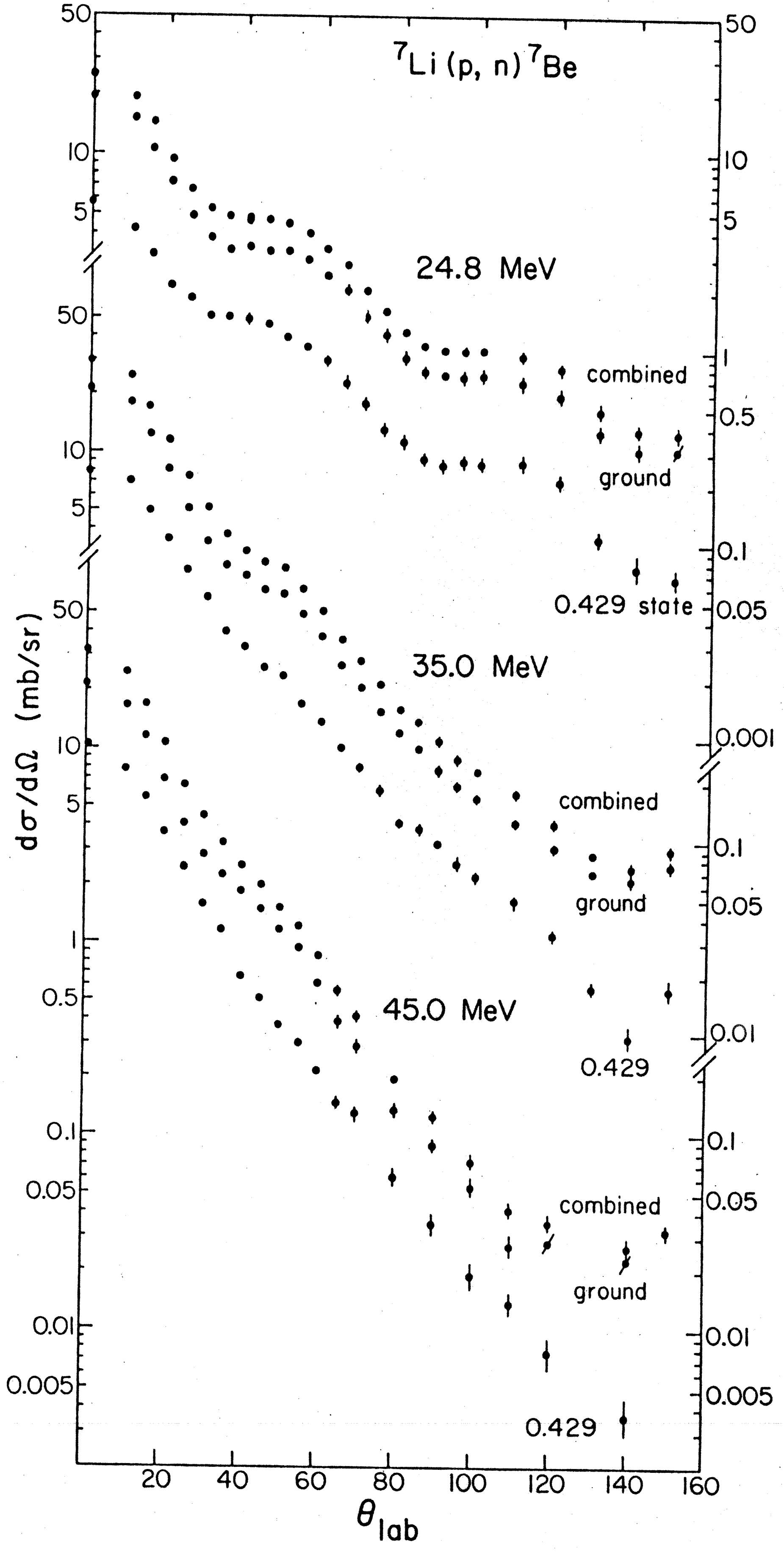
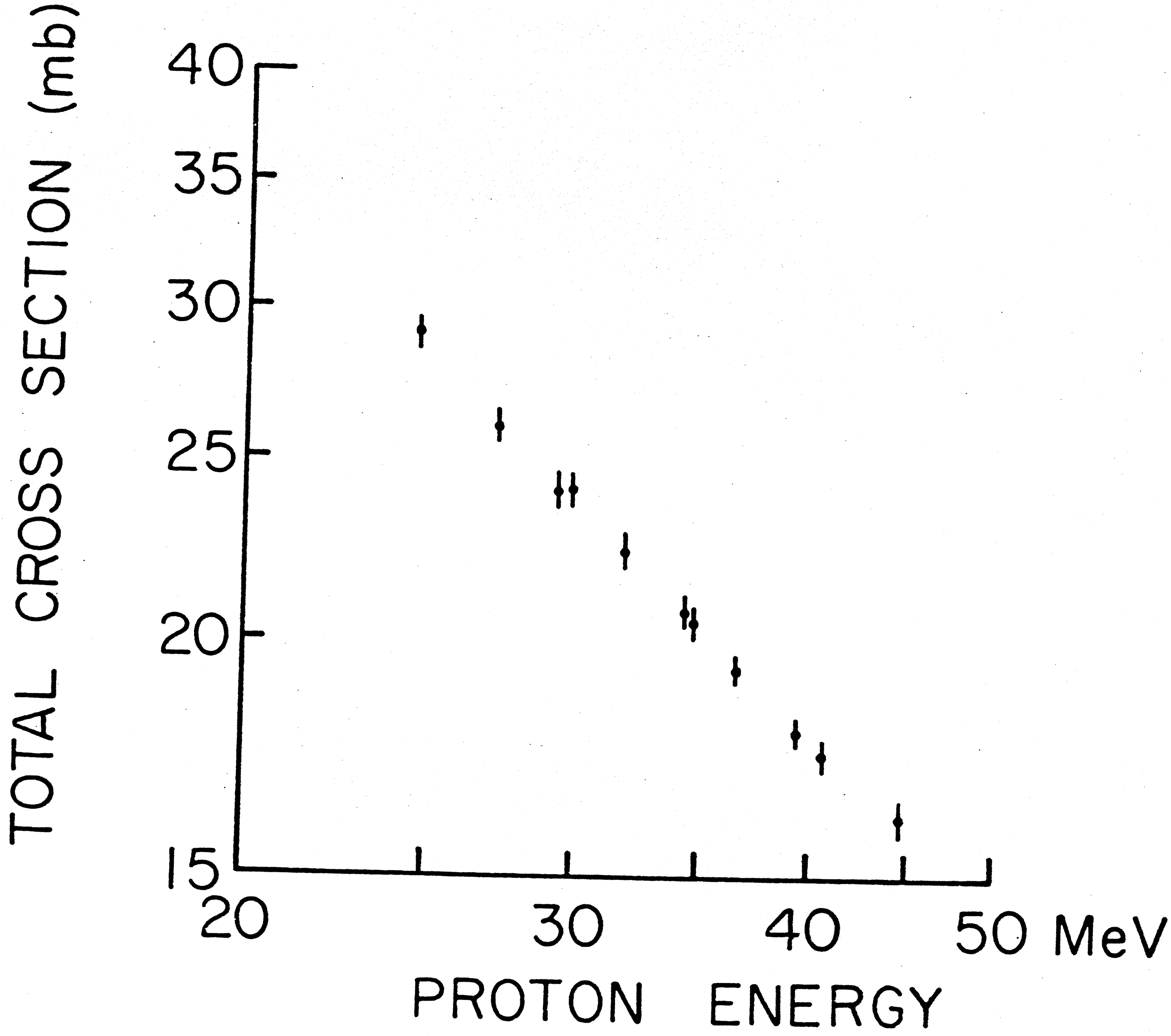
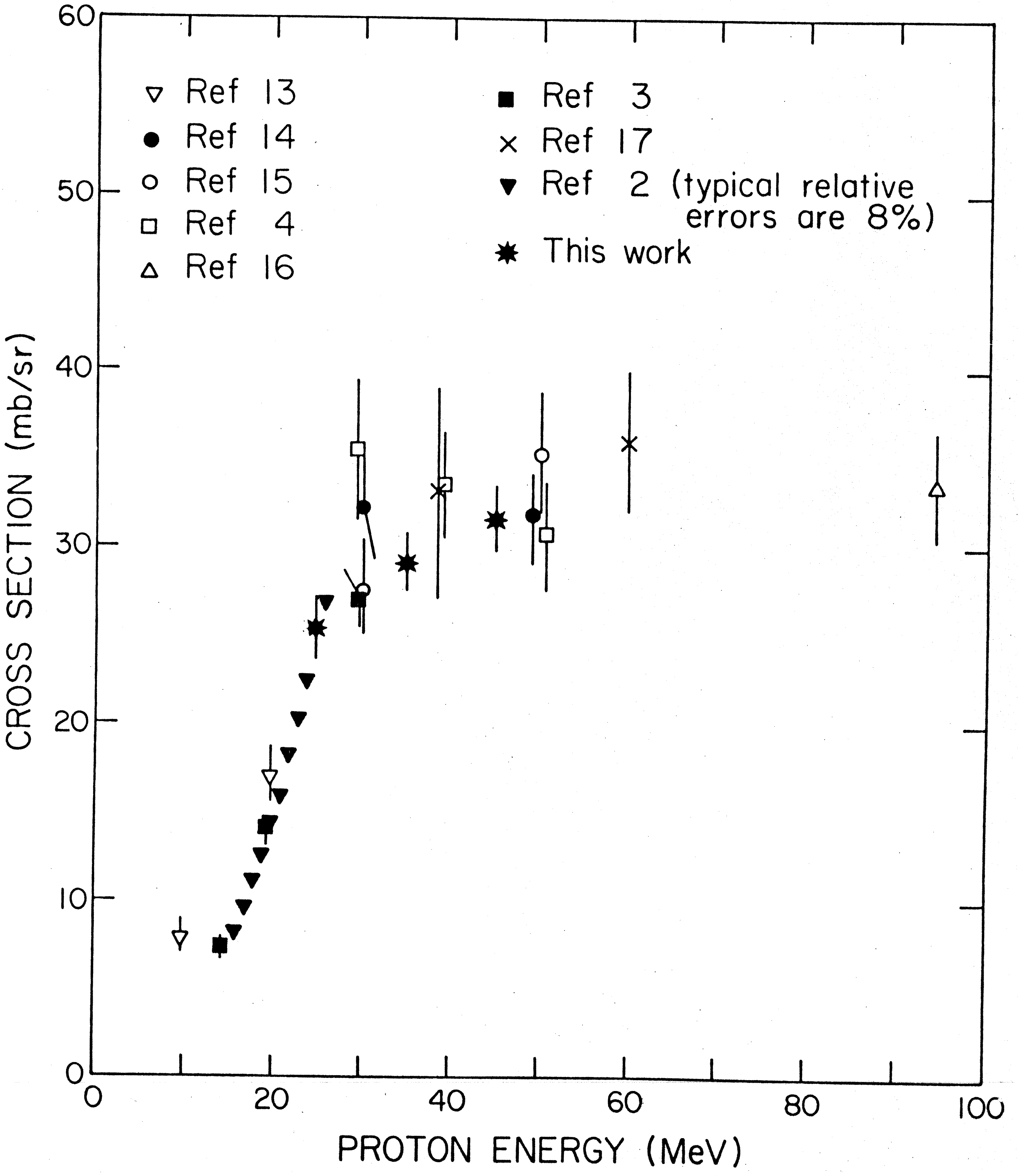
* Relative error for each cross section determined in the activation measurement is ±2%, the systematic error is ±3%, and the resulting total error (adding in quadrature) is ±3.6%.


^{7}Li(p, n)^{7}Be
MULTIPLE TARGETS
GAMMA
25.5
30.2
34.9
38.9
43.0
80
40
250
500
COUNTS PER CHANNEL
CHANNEL NUMBER


^{7}Li (p, n) ^{7}Be
24.8 MeV
35.0 MeV
45.0 MeV
combined
ground
0.429 state
combined
ground
0.429
combined
ground
0.429
dσ/dΩ (mb/sr)
θ_{lab}
20
40
60
80
100
120
140
160
50
10
5
50
10
5
50
10
5
1
0.5
0.1
0.05
0.01
0.005
50
10
5
1
0.5
0.1
0.05
0.001
0.1
0.05
0.01
0.1
0.05
0.01
0.005


TOTAL CROSS SECTION (mb)
40
35
30
25
20
15
20
30
40
50 MeV
PROTON ENERGY


CROSS SECTION (mb/sr)
PROTON ENERGY (MeV)
0
10
20
30
40
50
60
0
20
40
60
80
100
▽ Ref 13
● Ref 14
○ Ref 15
□ Ref 4
△ Ref 16
■ Ref 3
× Ref 17
▼ Ref 2 (typical relative errors are 8%)
✱ This work