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AUSTRALIAN ATOMIC ENERGY COMMISSION
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LUCAS HEIGHTS

ANGULAR MEASUREMENT OF THE ENERGY DISTRIBUTION OF
NEUTRONS FROM THE THICK TARGET ${}^7\text{Li}(p,n){}^7\text{Be}$ SOURCE

by

A. ROSE

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ABSTRACT

The energy spectrum of neutrons emitted from a thick lithium target bombarded by protons has been measured as a function of neutron angle of emission. The measurements were done at proton energies up to 2.8 MeV and at 30° intervals in the range 0 to 120° , using proportional detectors with gas fillings of hydrogen and methane. A review is given of papers published on the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction at 0° ; where applicable, comparisons are made with the present results.

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LITHIUM 7 TARGET; PROTON REACTIONS; NEUTRONS; BERYLLIUM 7; MEV RANGE 01-10;
ENERGY SPECTRA; ANGULAR DISTRIBUTION

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1. INTRODUCTION

The ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction using thin targets is a widely used source of near monoenergetic neutrons and has been studied in detail [Taschek and Hemmendinger 1948; Gabbard et al. 1959; Bevington et al. 1961]. However, the thick target reaction, which is used as a source of energetic neutrons, has had less attention. Palmer et al. [1966] have measured total neutron yields using protons with energies close to the lithium threshold, Scott [1971] has measured total yields using protons in the range 3-10 MeV, and Lefevre and Din [1969] have published the results of earlier unpublished data from the bombardment of thick lithium metal with 2.52 and 3.45 MeV protons. The only thick target measurements of angular spectra cited in the literature were made recently by Lone et al. [1977] at bombarding energies between 14 and 21 MeV. Ritchie [1976] has used thin target data to calculate thick target yields and angle dependent spectra.

The above measurements used BF_3 or ${}^3\text{He}$ filled detectors, crystal scintillators, or time-of-flight techniques. The measurements described here are made with proportional counters filled with hydrogen or methane to detect the neutrons and determine their energy. As proportionality in a proportional counter can be maintained down to a single ion pair of initial ionisation [Curran et al. 1949], it is theoretically possible to detect recoiling gas atoms of 100 eV. However, the practical lower limit is about 20 keV. Above 3 MeV, relatively high pressures or large counters are needed if the range of recoiling atoms is to be much less than the counter diameter. When filled with a suitable hydrogenous gas, the counters described below had an effective energy range from 50 keV to 1.2 MeV.

In the present measurements, neutrons in the range 0-1.3 MeV were produced from the bombardment of thick lithium metal by protons with energies up to 2.8 MeV. As this energy range is particularly interesting in fast reactor studies and is very close to the effective range of gas proportional proton recoil spectrometers used to measure neutron energy spectra in fast reactors, an experiment to measure the angular neutron spectra from this reaction would serve two purposes. A gap in the experimental data would be filled and, because the source has few gamma-rays and none of the high energy neutrons present in reactor spectra, the measurement would also provide an effective test of the accuracy and reliability of the proton recoil method.

In the present work, three spherical proportional proton recoil detectors are used to measure the angle dependent spectra of neutrons from a thick

lithium metal target. Comparisons are made with thin target yield measurements and thick target calculations.

2. EXPERIMENTAL METHOD

2.1 Detectors

Three SP2 spherical detectors, of the type developed by Benjamin et al. [1968] and manufactured by 20th Century Electronics Ltd, were used (Figure 1). Each detector was 39.4 mm in diameter and had a stainless steel wall of thickness 0.51 mm. The anode was made of 22 μm tungsten wire. The detector chambers were filled with 1 and 3 atmospheres of hydrogen (H_2) and 3 atmospheres of methane (CH_4), respectively; in the first of these a small ^{241}Pu source was deposited over ~ 1 mm of the anode, at the centre, to enable energy calibration. Before filling with gas, the detectors were evacuated to a pressure below 10^{-6} mm Hg and allowed to outgas for several hours. Hydrogen for the 1 atmosphere filling was purified by passing it through the wall of a silver-palladium tube. The 3 atmosphere hydrogen filling was also passed through the tube wall, but beforehand approximately 100 mm of purified nitrogen was added. The methane filling for the third detector had been specially prepared by the manufacturer and contained 4% nitrogen. Initially, this gas was very satisfactory, but in later fillings it was necessary to remove impurities, mainly water vapour, from the methane.

Nitrogen is required in 3 atmosphere fillings to allow calibration of the energy scale in a thermal neutron flux using the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction. The resolution of detectors, referred to the nitrogen calibration peak, was used to check the adequacy of the detector fillings. Typical resolution values were 4.8% for 3 atmosphere hydrogen and 8% for methane. The shape of the response functions, measured when the detectors were exposed to near mono-energetic neutrons, also indicated the quality of the detector fillings.

The three types of gas filling allowed measurements of neutron spectra to be made in overlapping ranges from 50 to 400 keV, 100 to 600 keV and 400 to 1200 keV (see Figure 2a). Reproducibility of detector fillings, checked by response function measurements at fixed proton bombarding energies, was better than 2%.

2.2 Energy Calibration

The $^{14}\text{N}(n,p)^{14}\text{C}$ reaction was used to calibrate the energy scale for the counters with added nitrogen. This reaction gives rise to 615 keV protons when the detector is exposed to a thermal neutron flux. The $^{14}\text{N}(p,n)^{14}\text{C}$ reaction

was chosen in preference to the ${}^3\text{He}(p,n){}^3\text{H}$ reaction because of uncertainty about the effective energy deposited by the ${}^3\text{He}$ reaction.

The energy calibration for the 1 atmosphere hydrogen-filled detector was provided by the energy of the alpha particles emitted by the ${}^{241}\text{Pu}$ on the detector anode wire. The dominant alpha peak was first calibrated using known neutron energies; those near mono-energetic neutrons were produced by bombarding a thin lithium target (6 keV) with 2.04 MeV protons. Once the plutonium peak had been calibrated it was then used as the standard. The deposited energy from the main alpha peak was determined as 366 ± 10 keV. A typical measured response function and the plutonium alpha peak are shown in Figure 3.

2.3 Targets and Target Assembly

Details of the target assembly are shown in Figure 4. A thick or thin lithium target could be chosen by rotating an outer ring. This enabled response functions, resolution checks and spectrum measurements to be made with minimum change to beam parameters, thus ensuring maximum reproducibility of the system.

A number of targets were used for these measurements. The thin targets were about 10 keV thick and produced by vacuum evaporation of lithium metal onto a 0.127 mm thick tantalum backing. As the thickness of lithium needed to slow a proton from 3 MeV to 1.88 MeV is ~ 0.14 mm, it was impractical to produce thick targets by evaporation. Targets were therefore made by pressing a lithium metal pellet into a machined brass disc (see Figure 5). These targets were about 2 mm thick, thus ensuring that the minimum thickness of 0.14 mm was achieved, even in the presence of surface irregularities. All targets were transported to and installed in the target assembly under an argon atmosphere so that the lithium surface remained clean and bright. Since the target was cooled by blowing a jet of air onto the target backing, it was necessary, because of the poor thermal conductivity of lithium, to keep beam currents below 3 μA to avoid target damage.

3. EXPERIMENTAL PROCEDURES

Using the Commission's 3 MeV Van de Graaff accelerator at Lucas Heights, thick lithium metal targets were bombarded with protons in the range 2 to 2.8 MeV; spherical detectors then measured the resulting neutron spectrum in 30° intervals over the range 0 to 120° . A 2.5 cm diameter BF_3 counter acted as a monitor for all the runs.

Since the maximum neutron energies are a function of proton bombarding energy and measuring angle, this was the basis for the choice of detector gas filling for the various measuring positions (Table 1). For example, at a proton energy of 2.4 MeV the neutrons will have energies up to 0.68 MeV.

From Figure 2a, it is clear that this is outside the energy range of the 1 atmosphere filling; the proton energies and angles not covered by a particular filling are indicated in Table 1 by dashes.

As the maximum proton energy used in these measurements was 2.8 MeV, both the ${}^7\text{Li}(p,n){}^7\text{Be}$ (ground state transition) and the ${}^7\text{Li}(p,n){}^7\text{Be}^*$ (first excited state transition) contribute to the neutron yield. The yields from both states have been calculated as a function of proton energy and neutron energy by Ritchie [1976] and, as an example, the relative yields at 0° are shown in Figures 2a and b. Figure 2a shows the 0° ground and excited state yields as a function of neutron energy and Figure 2b shows the yields as a function of proton energy. These figures were also used as a guide to the choice of gas and pressure.

To reduce the effect of floor- and wall-scattered neutrons, the experiment was conducted on a tower 5 m above the floor and 6 m from the nearest concrete shield wall.

The voltages for the three detectors were 1500, 2500 and 3000 V respectively. The detector output was fed via an Ortec preamplifier to a main amplifier operated at long time constants (10 μs) to accommodate the detector pulses with the longest rise times. Output from the amplifier was connected to the analogue-to-digital converter of a PDP7 computer. The variation in target output was monitored with a 25 mm BF_3 detector. The relative position of detector to target was controlled by the computer, the positioning mechanism being mounted on the tower. This mechanism fixed the detector position 50 m from and in the same horizontal plane as the target for all experiments (see Figure 6).

As the target current was limited to $<3 \mu\text{A}$, running times were quite long. At angles 0 and 30° (see Figure 6) two hours were required, whereas at the other angles (60 , 90 and 120°) the running time was four hours. Before each set of measurements, the pulse height scale for the detector in use was calibrated. The detector containing the 1 atmosphere H_2 filling was calibrated against the inbuilt ${}^{241}\text{Pu}$ source. The pulse height scale of the other two detectors was calibrated using 615 keV protons from the ${}^{14}\text{N}(n,p){}^{14}\text{C}$ reaction which has a high cross section for thermal neutrons. A relatively high thermal flux was achieved by interposing a block of paraffin wax between the detector and a lithium target bombarded with 2.0 MeV protons. At this energy, the neutrons emitted at 0° have a maximum energy of 230 keV and are rapidly slowed in a few centimetres of paraffin wax.

4. DATA ANALYSIS AND RESULTS

Data were collected in the PDP7 computer which was run in the pulse height

analysis mode. Four regions, each with 1024 channels, were used to store the measured proton recoil spectra and monitor counts and time were recorded on scalars. This information was automatically transferred to paper tape and later to magnetic tape, together with relevant information for each run.

The analysis program of Benjamin et al. [1968] was used to calculate the neutron spectra from the proton recoil spectra. All spectra were normalised using counts from the BF_3 detector and run times. Intercomparisons were made of the neutron spectra in the energy ranges for each detector filling and they were then summed to produce a single curve for each angle. Finally, a composite curve for each angle was obtained using scale factors produced from the overlapping energy regions. Although no attempt was made to measure absolute yields, the counts from the BF_3 detector ensured that, after normalising to the 0° measurement, the other angular measurements were correctly related to each other. These spectra are shown in Figures 7 and 8.

Intercomparisons of the raw proton data were made between symmetrical positions (e.g. 30° left and right) and between repeated measurements to check their reproducibility; this was better than 2% for positions 0° , 30° and 120° . However, comparative checks on the analysed data revealed that the errors were propagated within the unfolding program and the 2% errors became 3%. In the case of the 60° and 90° positions, the reproducibility of the raw data was nearer 3% and, after unfolding, $\sim 5\%$. These discrepancies are attributed to systematic effects due to proton beam misalignment and neutron scattering ($\sim 1\%$ at 60° and 90°) in the target assembly.

5. REVIEW OF ANGULAR YIELD MEASUREMENTS

A number of measurements of the 0° neutron yield from the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction were made using thin targets and monoenergetic protons. A summary of $\text{Li}(p,n)$ yield measurements is given in Table 2. The earliest measurements of neutron yields from this reaction were done by Taschek and Hemmendinger [1948] who used the activity induced in a manganese bath to determine neutron yields for proton bombarding energies between 1.86 and 2.5 MeV. Angular measurements were made with a long counter. Bair et al. [1952] bombarded an 18 keV lithium metal target with monoenergetic protons in the range 1.8 to 5.3 MeV and measured the neutron yield in the forward (0°) direction with a BF_3 long counter. Using a 40 keV LiF target and a ${}^3\text{He}$ fast neutron spectrometer, Batchelor [1955] measured the 0° yield of neutrons from the ${}^7\text{Li}(p,n){}^7\text{Be}$ excited state reaction for proton energies in the range 2.5 to 2.9 MeV.

Using a LiF target 10 keV thick and protons in the range 1.9 to 3.0 MeV, Gabbard et al. [1958] measured the neutron yield at 0° , 60° and 120° with a

crystal scintillator positioned at each angle. A time-of-flight system and a thin metallic lithium target was used by Bevington et al. [1961] to measure angular neutron spectra at 30° intervals between 0° and 150° for proton energies in the range 2.6 to 4.1 MeV. They normalised their results to the 0° results of Gabbard et al. up to 3.0 MeV; for higher proton energies they used the measurements of Bair et al. normalised to those of Gabbard et al. The neutron detection threshold was 300 keV. Similarly, Borchers and Poppe [1963] measured the production of neutrons from protons with bombarding energies in the range 3 to 13 MeV.

Bergström et al. [1967] have measured the angular distribution of neutrons from a 6 keV lithium metal target over the proton range threshold to 2.36 MeV. Angular distributions were also measured by Buccino et al. [1964], using an 8 keV LiF target with protons in the range 2.4 to 3.0 MeV. More recently, Lefevre and Din [1969], using 8 and 16 keV evaporated lithium fluoride targets and a long counting neutron detector, measured the yield at 0° with protons in the range threshold to 2.65 MeV.

Elbakr et al. [1971] measured neutron angular distributions by time-of-flight using proton energies from 2.27 to 5.5 MeV in steps of 200 keV. Apparently they were unaware of the measurements made by Buccino et al. and Bergström et al.; even so, they made comparisons and claimed good agreement with the work of Bevington et al. [1961] and Borchers and Poppe [1963]. Meadows and Smith [1972] generated the 0° neutron yield curve over the range 1.88 to 7.7 MeV. They compared the data of Gabbard et al. [1958], Bevington et al. [1961] and Lefevre and Din [1969] over the range 1.88 to 3.2 MeV, their own measurements for $E_p = 3.2-7.0$ MeV, and a combination of their own data and those of Borchers and Poppe [1963] over the range 7.0 to 7.7 MeV.

This is the bulk of the measurements made using thin targets. Surprisingly few measurements have been made with thick lithium targets. Barnard et al. [1965] used thick lithium targets bombarded with protons to produce neutrons for elastic scattering measurements on Bi, Al, Fe and Ni. However, they found it impractical to measure the neutron spectrum emitted by the target and used a ratio technique for their cross section determination. Palmer et al. [1966] measured the total neutron yield near threshold and Scott [1970] measured the total yield over the proton bombarding energy range 3 to 10 MeV. The measurements made by Lefevre and Din [1969] were undertaken to resolve a discrepancy between a measured 0° neutron energy spectrum produced from a thick metal target and the calculated thick target neutron energy spectrum. Thick target measurements were also made at the University of Oregon but appear to be

unpublished. However, the spectrum obtained at Oregon by bombarding a thick lithium target with protons of 2.5 MeV energy and that obtained with 3.45 MeV protons are cited by Lefevre and Din [1969]. Lone et al. [1977] measured the neutron yields and spectral distributions of neutrons produced by bombarding thick metallic ${}^7\text{Li}$ targets with 14.8, 18.0 and 23.0 MeV protons at five angles between 0 and 40° . These are the only thick target measurements found in the literature.

There have been a number of calculations of thick target spectra. In their review of the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction, Gibbons and Newson [1960] give tables of neutron energy as a function of proton energy and emission angle for both the ground and excited state. Energy distributions have been calculated by Palmer [1966] for targets up to 1900 Å thick at 0, 45, 90, 135 and 180° . Theobald et al. [1971] have calculated neutron intensities from proton-bombarded thick lithium targets as a function of angle for the ground state of proton energies between 2.05 and 2.45 MeV and the excited state between 2.89 and 3.47 MeV.

Ritchie [1976] used the experimental values of Bergström et al. [1967] and Buccino et al. [1964] to calculate the thick target angular spectra from the $\text{Li}(p,n)$ source. The results were produced as a function of neutron energy and hence can be compared directly with the present measurements.

Rainbow et al. [1977] describe a method of estimating angular yields of neutrons from thick lithium targets using various experimental and theoretical estimates of the angular cross section for the $\text{Li}(p,n)$ reaction up to proton energies of 3 MeV.

In the case of neutron scattering, Ryves [1973] used the Monte Carlo method to calculate angular distributions and energy spectra of keV neutrons scattered from a typical target assembly. In one analysis, only 1% of the neutrons produced from 2.265 MeV protons on thin lithium, with a silver target backing 0.025 cm thick, underwent scattering in the target assembly. However, with thicker target backings, the component of scattered neutrons at 0° was reported to be as high as 10%.

6. COMPARISON OF RESULTS

The present results have been compared with the thick target calculations of Ritchie [1976] for all the angles measured. Absolute yields for the present measurements were not determined, hence to permit comparison they have been normalised to the results of Ritchie, allowing equal weight to all the measured points (see Figure 9).

The derived angular neutron spectra of Ritchie [1976] were accurate to 10 to 15%. Agreement with the present results is within this range for 0, 30

and 60° , although at the lower energies there is a discrepancy. The 90° and 120° results do not agree so well. At 120° , there is similarity of shape but the neutron numbers are clearly different. There seems to be an energy displacement in the 90° comparisons. As was stated earlier, these two angles may be sensitive to scattered neutrons; the degree of scattering is thought to be small (1.3%), however Ryves [1973] calculated that, in some cases for thick target backing, the scattered component can be as high as 10%.

The 0° measurements of a number of workers, including the present ones at 0° , are compared in Figure 10, all results being normalised to those of Ritchie [1976]. The scaling factors were again derived by fitting all points. In the case of the results of Bair et al. [1952], the lowest energy point was excluded during the scaling process and the results then scaled and the factor determined from the remaining points. It should be noted that all the experimental results in Figure 10, except for the present results and those achieved at Oregon, were made using thin targets and at near monoenergetic protons. A correction for the excited state neutron contribution has not been made for the thin target results. However, as this correction for the 0° angle is less than the experimental errors (see Figure 2a), the comparison between the thick and thin measurement is relevant. Agreement in all cases is reasonable except for the results of Elbakr et al. [1971] which are well outside the error range. The Meadows and Smith [1972] results over the energy range in Figure 2a are merely the data taken from Gabbard et al. [1959], Bevington et al. [1961] and Lefevre and Din [1969]. Similarly, the results of Ritchie [1976] were based on the data of Bergström et al. [1967] and Buccino et al. [1964].

7. CONCLUSIONS

With proton energies around 3 MeV, the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction in thick lithium targets offers a neutron source which is ideal for testing proton recoil detectors and the necessary unfolding codes. The present measurements show that the detectors used here will adequately cover the neutron energy range 50 keV to 1.2 MeV and should be capable of measuring neutrons in the range 20 keV to 3.0 MeV. The resolution of the detectors is dependent on the filling gas and the filling techniques and, for these measurements, varied from $\sim 5\%$ with hydrogen to $\sim 10\%$ with methane.

The reproducibility of the technique is good and, in most cases, better than 3%. As the composite spectra for the different angles were obtained by combining the energy regions covered by the different gas fillings, the reproducibility of the overlap of these energy regions is important. Again it was better than 3%.

The comparison of these measurements with the calculations of Ritchie [1976], although not in complete agreement, gives confidence in the reliability of this technique to measure the ${}^7\text{Li}(p,n){}^7\text{Be}$ neutron spectrum. This confidence is enhanced when the 0° measurements of this reaction by other workers are compared with the present results. With the exception of the Elbakr et al. [1971] results, agreement is good. Therefore, the use of the proton recoil technique for the measurement of neutron spectra produced by target bombardment, although limited to a specific energy range, compares favourably with other techniques and should complement methods used to measure reactor spectra.

8. ACKNOWLEDGEMENTS

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TABLE 1
CALCULATED MAXIMUM NEUTRON ENERGIES (MeV) EXPECTED FROM
THE ${}^7\text{Li}(p,n){}^7\text{Be}$ REACTION

Proton Energy (MeV) \ Angle (degrees)	0	30	60	90	120	Detector Filling (1)
2.0	0.230	0.202	-	-	-	1 at H ₂
2.15	0.407	0.371	0.267	-	-	1 at H ₂
2.31	-	-	0.414	0.292	-	1 at H ₂
2.5	-	-	-	0.433	0.320	1 at H ₂
2.4	0.680	0.626	0.500	0.359	0.260	3 at H ₂
2.8	-	-	-	0.658	0.510	3 at H ₂
2.8	1.099	1.027	0.853	0.658	0.510	3 at CH ₄

(1) at = atmosphere

TABLE 2

SUMMARY OF Li (p,n) YIELD MEASUREMENTS

Date	Authors	Proton Energy (MeV)	Measurement Type	Target Details
1948	Taschek & Hemmendinger	1.86 to 2.5	Mn bath and BF ₃	Thin Li metal
1952	Bair et al.	1.8 to 5.3	BF ₃	Thin Li metal
1955	Batchelor	2.5 to 2.9*	³ He	Thin LiF
1958	Gabbard et al.	1.9 to 3.0	Crystal scintillator	Thin LiF
1960	Gibbons & Newson	1.9 to 9.0	Calculation	Thick Li metal
1961	Bevington et al.	2.6 to 4.1	Time-of-flight	Thin Li metal
1963	Borchers & Poppe	3 to 13	Time-of-flight	Thin Li metal
1963	Buccino et al.	2.4 to 3.0*	Time-of-flight	Thin LiF
1965	Barnard et al.	-	-	Thick Li metal
1966	Palmer	Threshold	Calculation	-
1966	Palmer et al.	Threshold		Thick Li metal
1967	Bergström et al.	1.88 to 2.36	Time-of-flight	Thin Li metal
1968	University of Oregon	2.5 and 3.45	BF ₃	Thick Li metal
1969	Lefevre & Din	1.88 to 2.65	BF ₃	Thin LiF
1970	Scott	3.0 to 10.0	Mn bath	Thick Li metal
1971	Elbakr et al.	2.27 to 5.5	Time-of-flight	-
1971	Theobald	2.05 to 2.45 2.89 to 3.47*	Calculation	Thick Li metal
1972	Meadows & Smith	1.88 to 7.7	Time-of-flight	Thin Li metal
1976	Ritchie	1.88 to 3.0	Calculation	Thick Li metal
1977	Lone et al.	14.8, 18, 23	Time-of-flight	Thick Li metal
1977	Rainbow	1.88 to 3.0	Calculation	Thick Li metal
1978	Rose	1.88 to 2.8	Proton recoil	Thick Li metal

*⁷Li (p,n) ⁷Be excited state reaction only

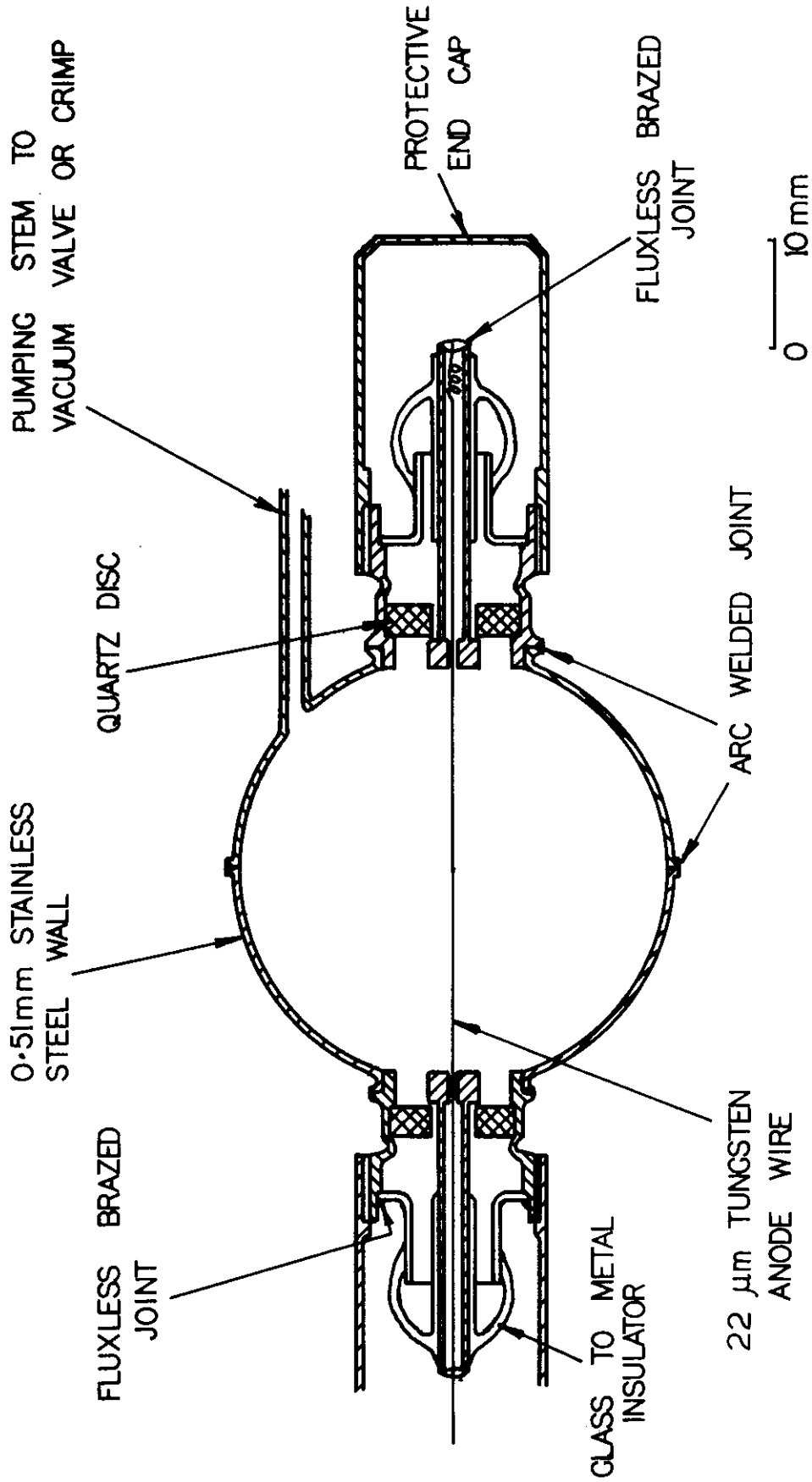


FIGURE 1. DETECTOR DETAILS

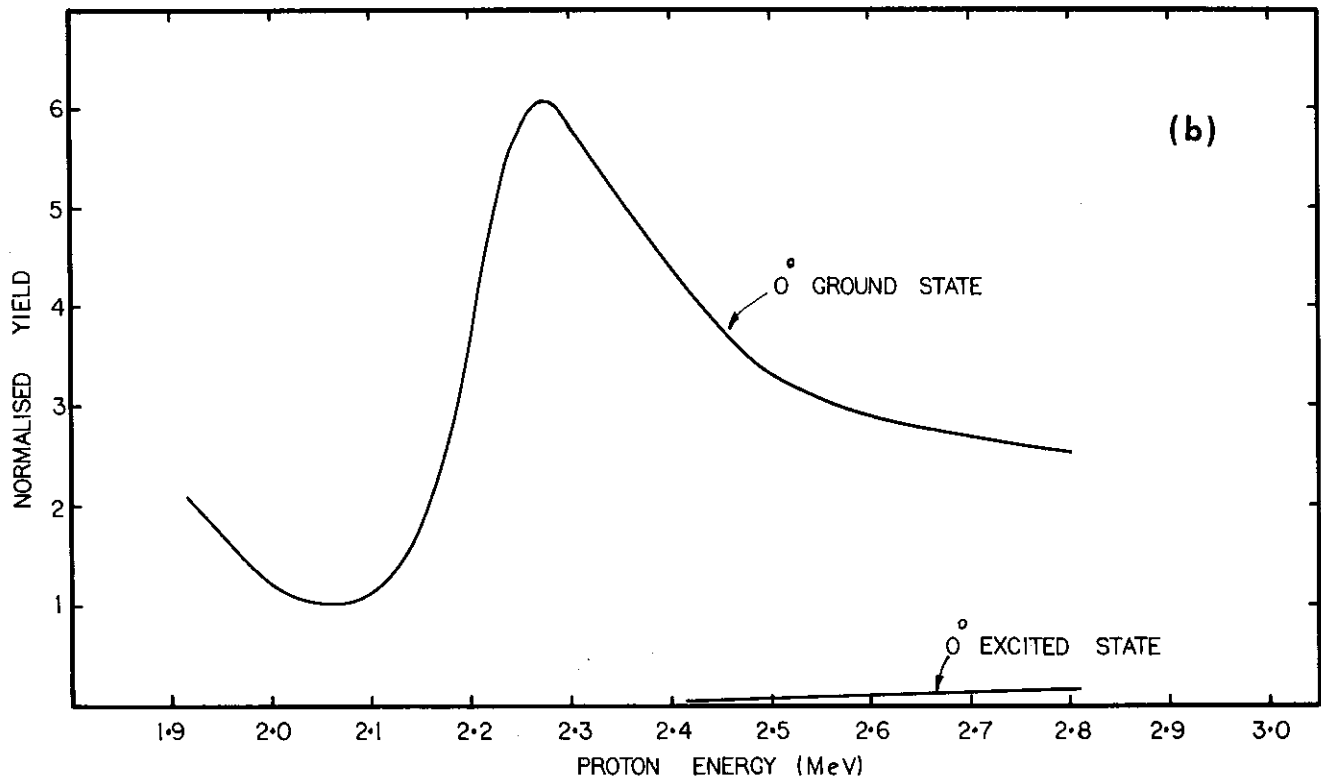
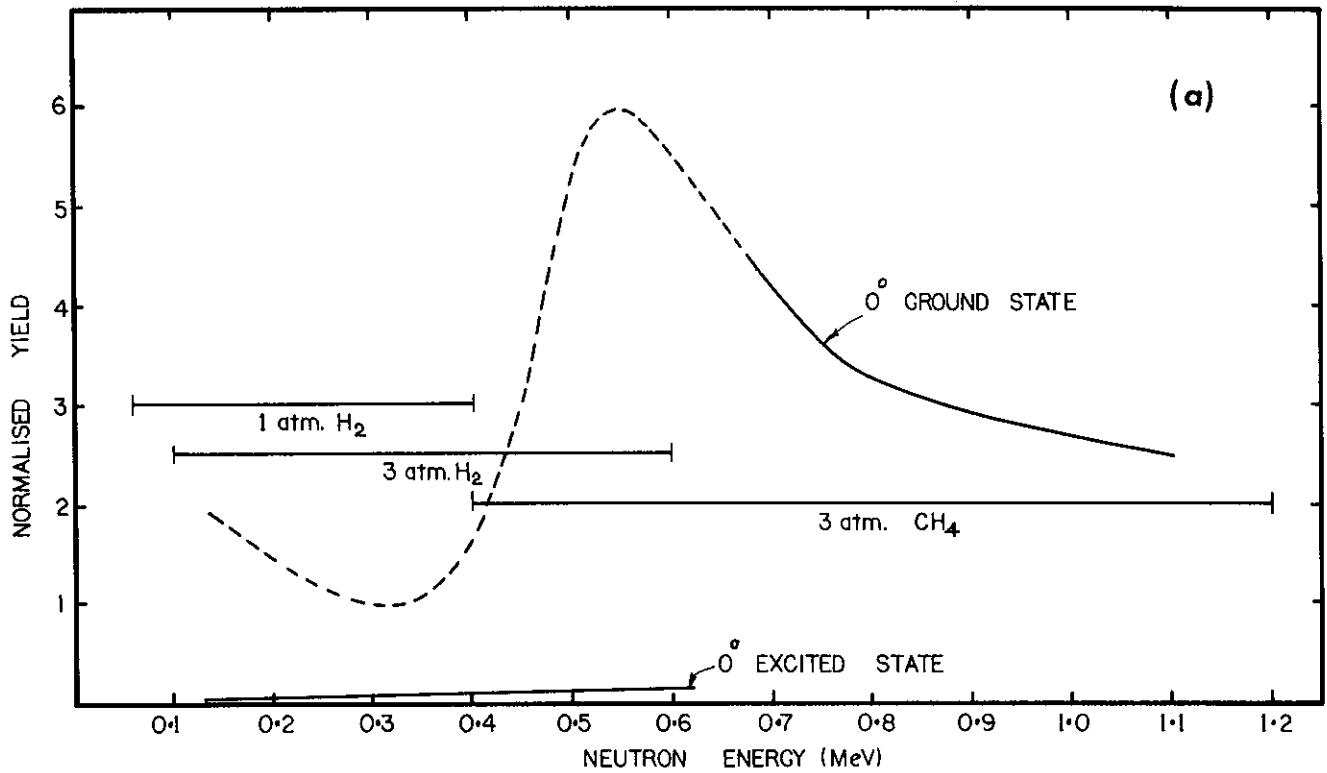


FIGURE 2. 0° GROUND AND EXCITED STATES AS A FUNCTION OF NEUTRON ENERGY AND PROTON ENERGY

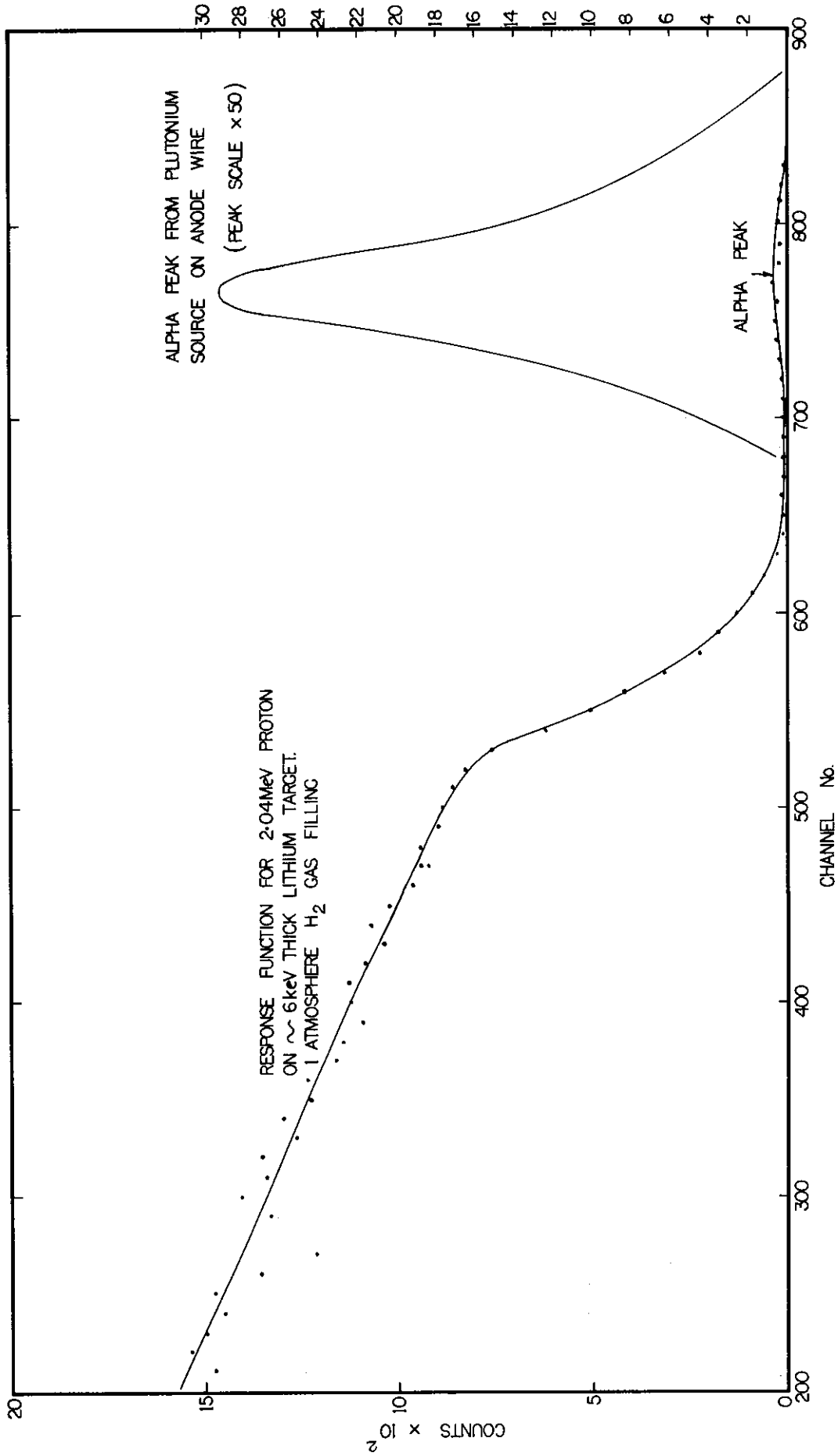


FIGURE 3. COMBINED MEASUREMENT OF 1 ATMOSPHERE HYDROGEN FILLING
RESPONSE FUNCTION AND ALPHA SPECTRUM

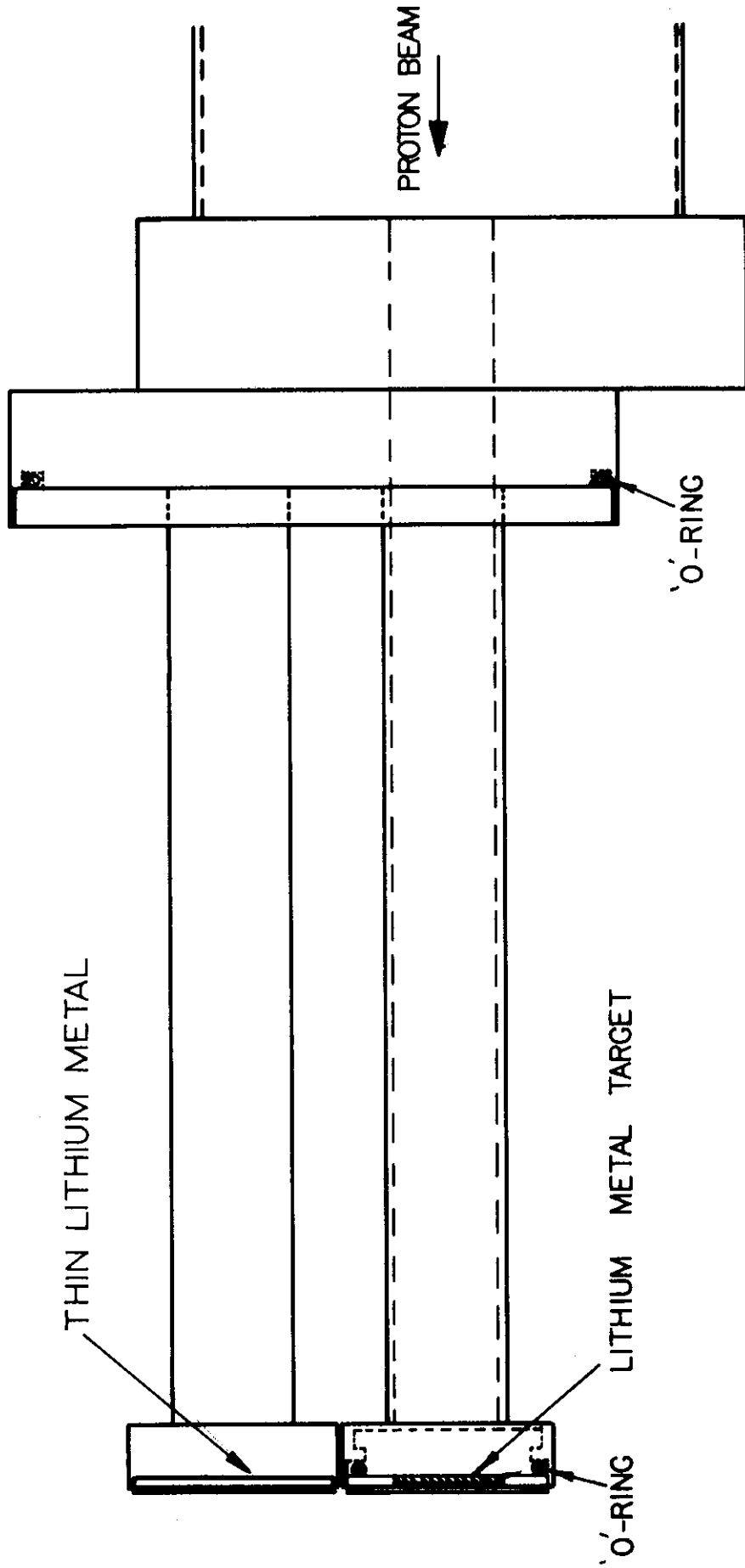


FIGURE 4. TARGET ASSEMBLY

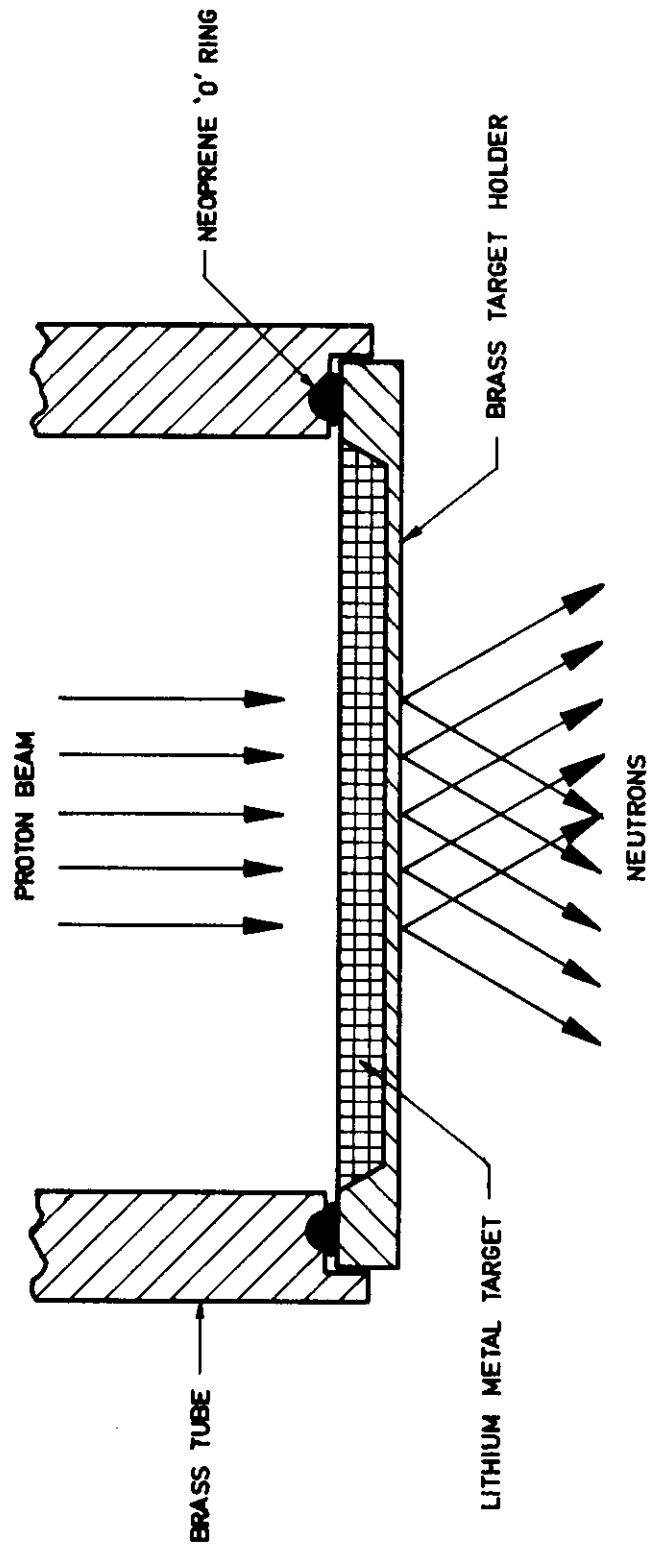


FIGURE 5. TARGET HOLDER

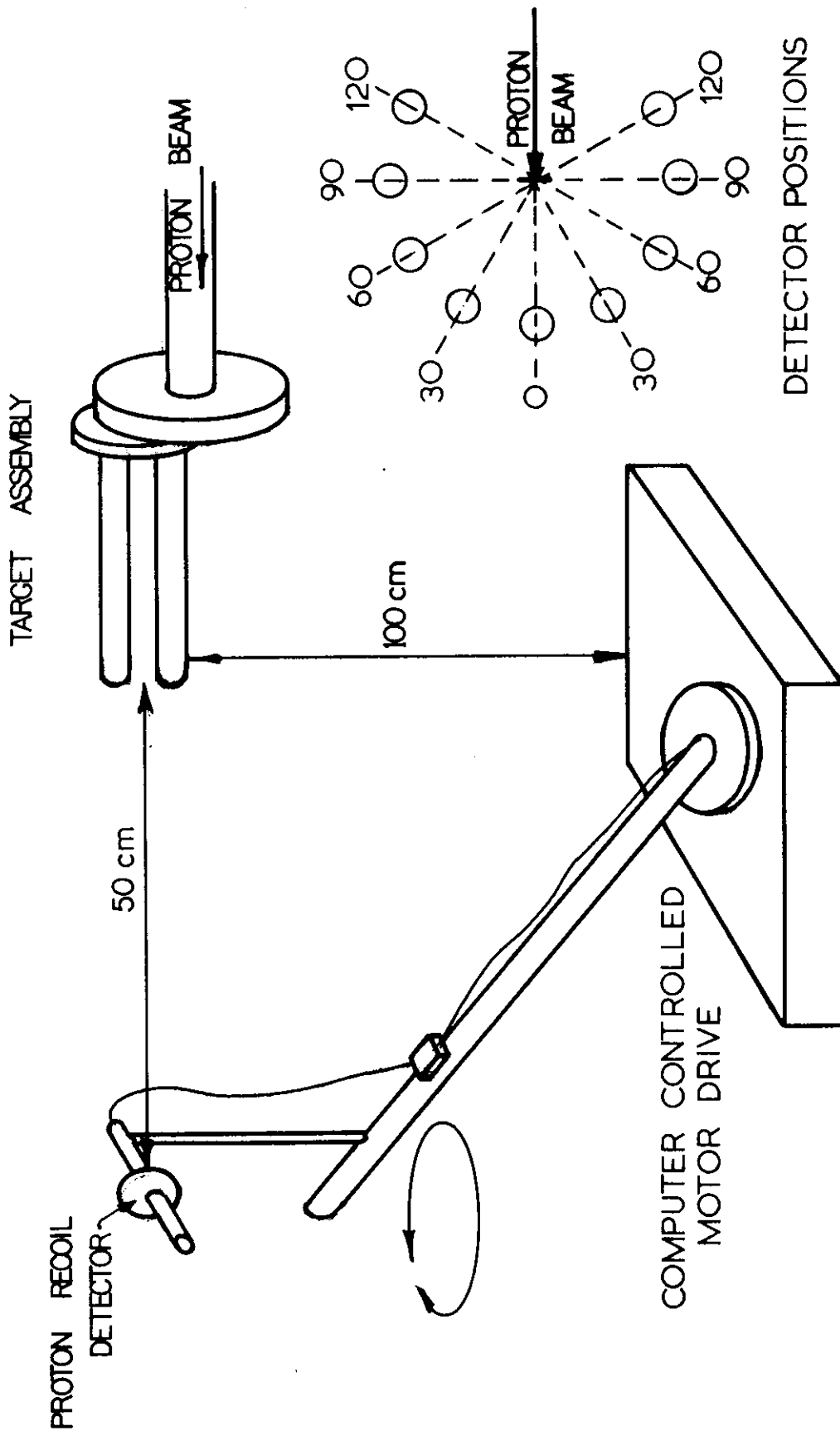


FIGURE 6. DETECTOR POSITIONING ARRANGEMENT

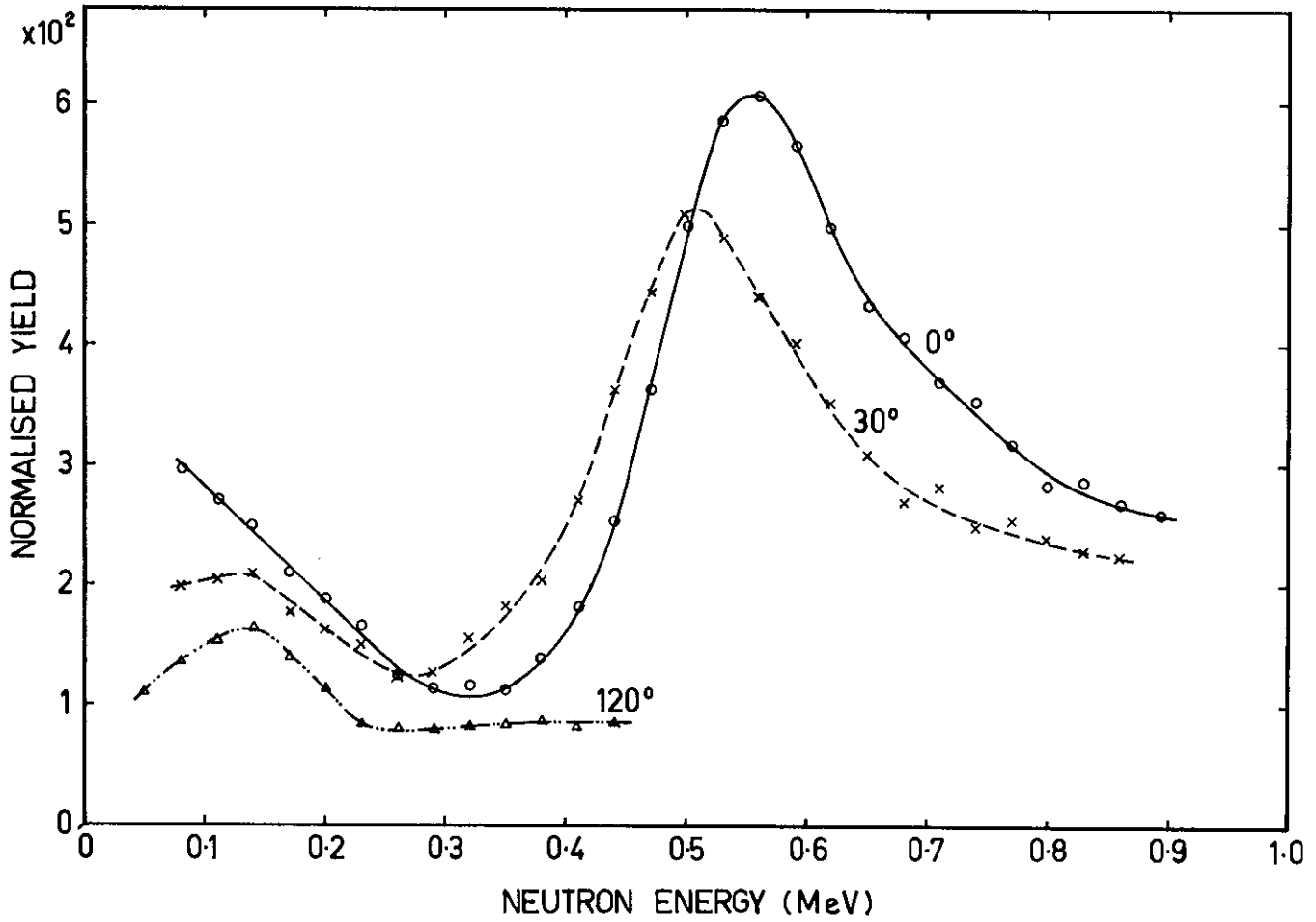


FIGURE 7. LITHIUM (p,n) SPECTRA AT 0, 30 AND 120°

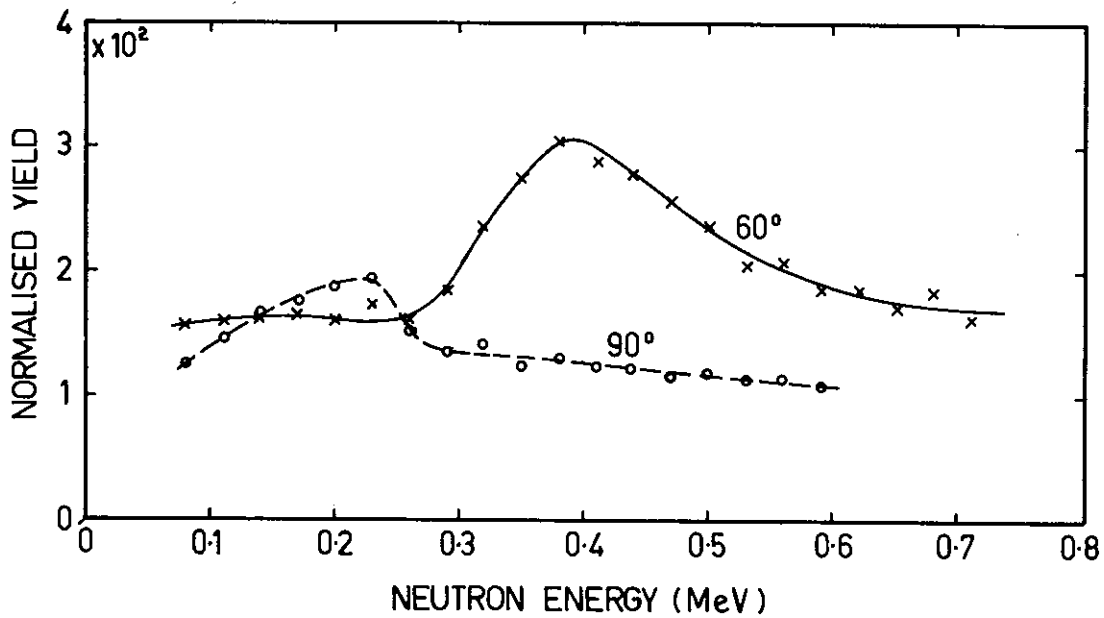


FIGURE 8. LITHIUM (p,n) SPECTRA AT 60 AND 90°

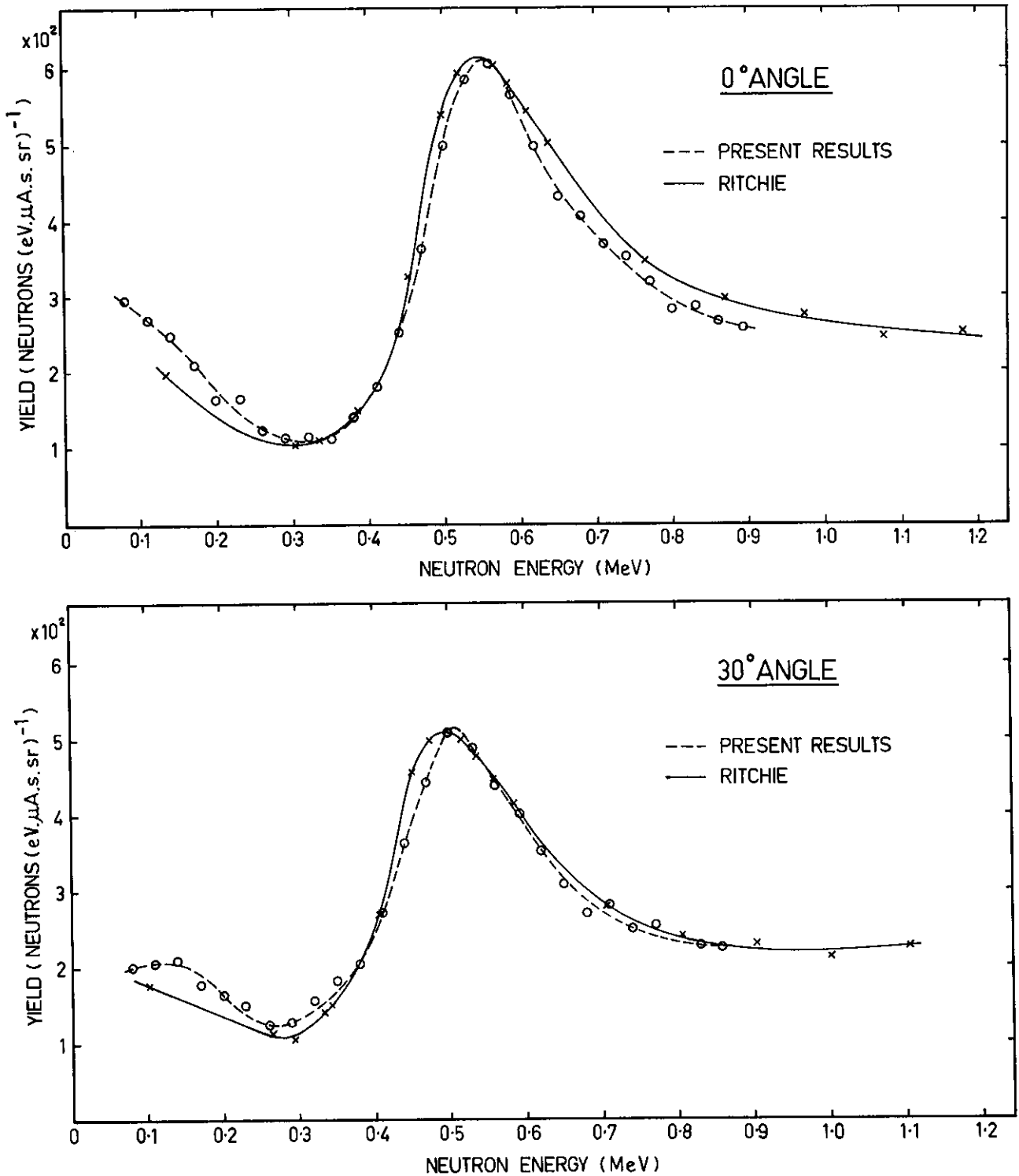


FIGURE 9. COMPARISON OF MEASURED AND CALCULATED SPECTRA

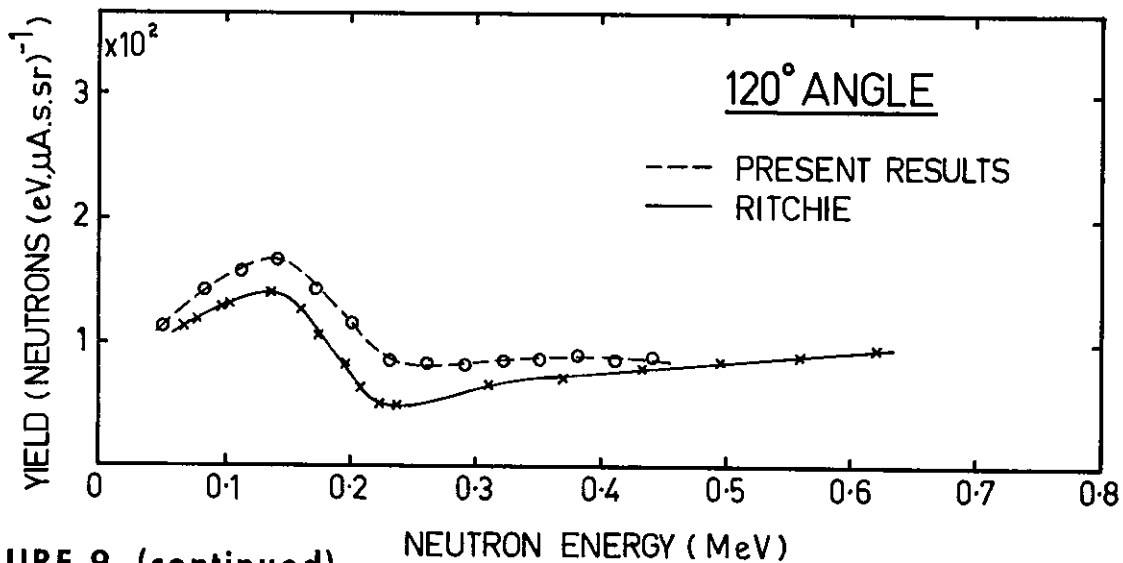
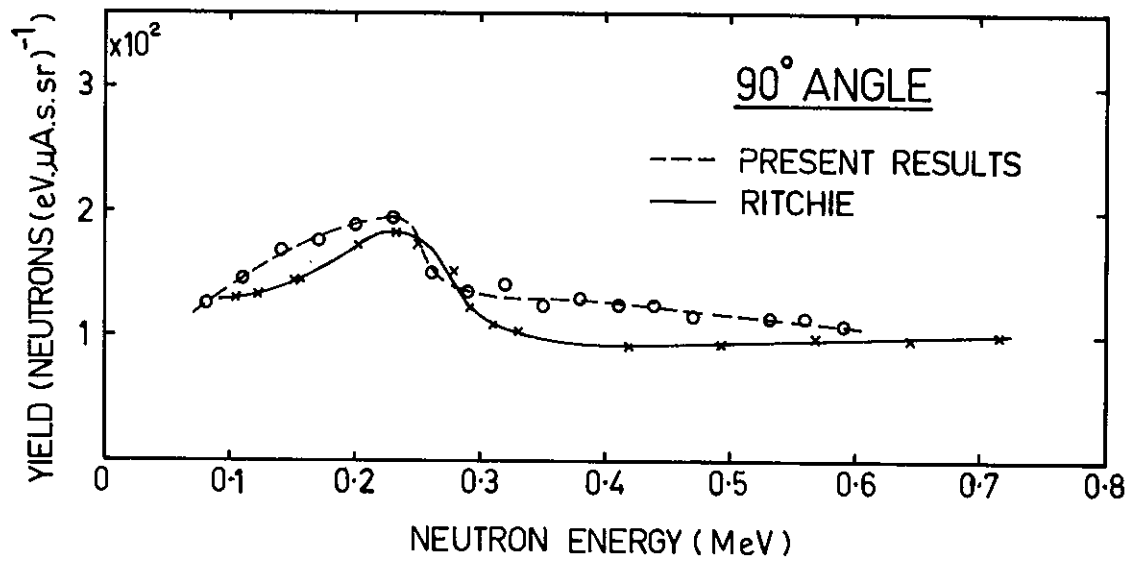
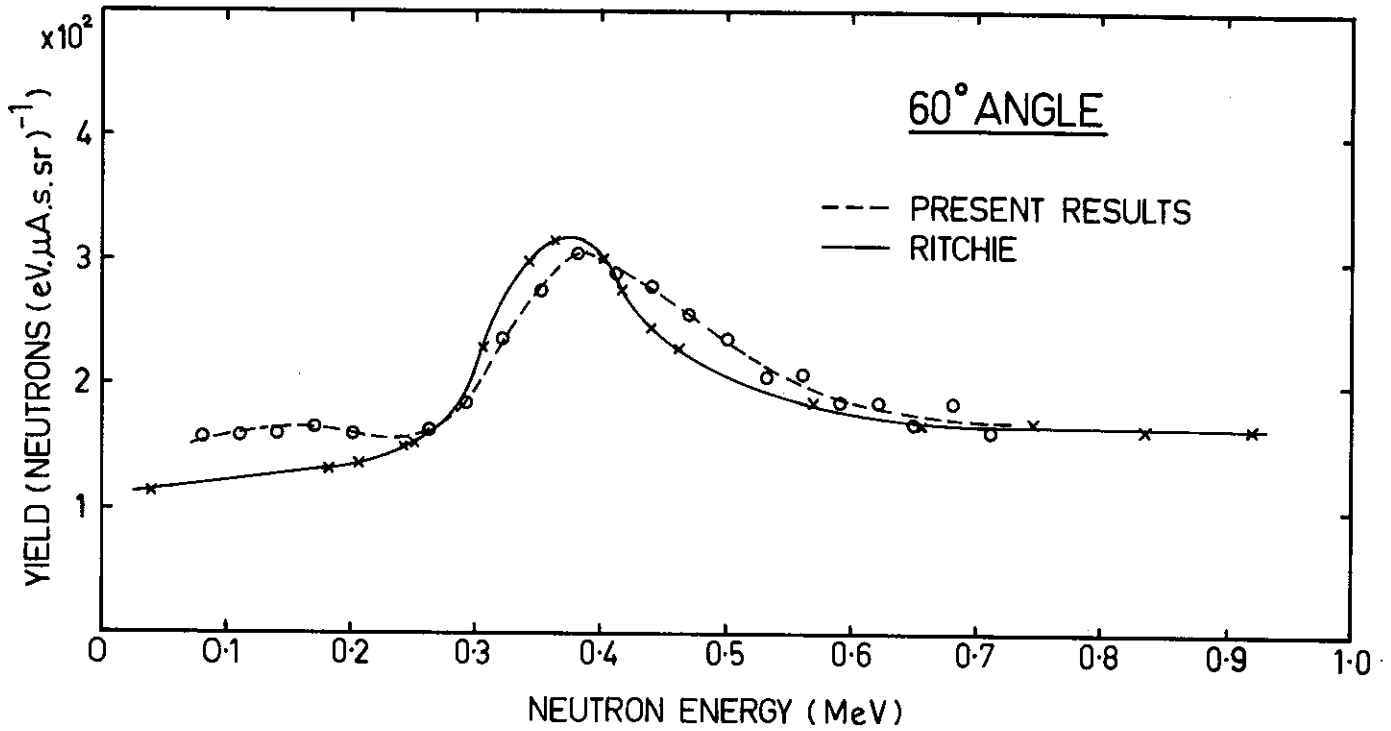


FIGURE 9. (continued)

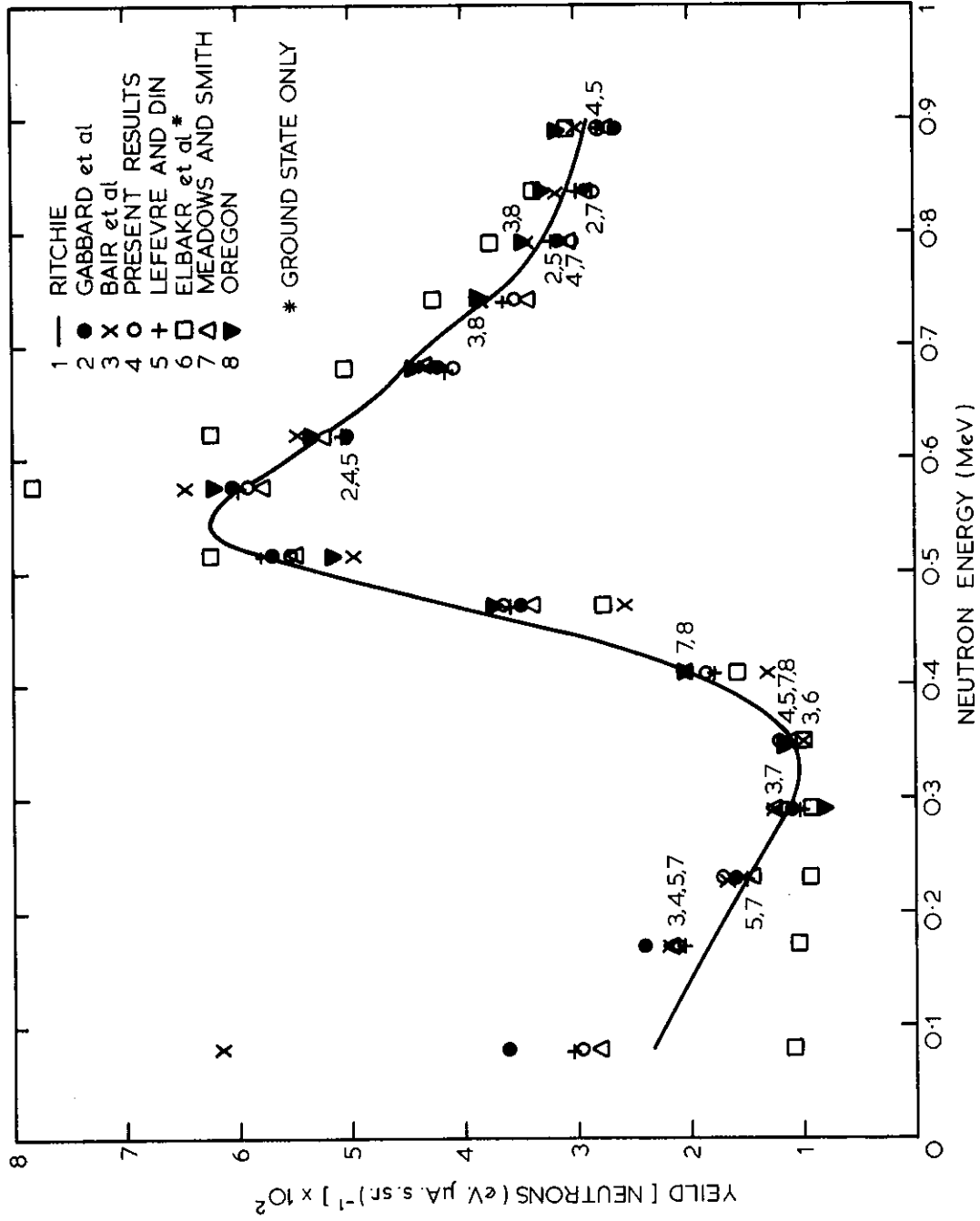


FIGURE 10.0° MEASUREMENTS COMPARISON