ZERO DEGREE NEUTRON YIELD FROM THE ^{7}Li(p, n)^{7}Be REACTION NEAR 2·2 MeV*

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Summary

A discrepancy between calculated and measured neutron spectra from the ^{7}Li(p, n)^{7}Be reaction is discussed. Results from a relative yield curve measurement with a long counter are compared with existing data.

Introduction

The ^{7}Li(p, n)^{7}Be reaction has been extensively used as a neutron source reaction. The sharp reaction threshold at 1·881 MeV is among the most accurately known accelerator energy calibration points (Nuclear Data Tables 1960, p. 167). The total cross section for neutron production as measured by Macklin and Gibbons (1958) with their 4π graphite moderated neutron counter is known absolutely to 4%, and the relative total yield as a function of proton energy is accurate to 1% from the same measurement. The differential yield at 0° as measured by Gabbard, Davis, and Bonner (1959, hereinafter referred to as GDB) and normalized by them to the 4π results through an angular distribution taken at a proton energy of 2·265 MeV was judged accurate to 5% by Marion (Nuclear Data Tables 1960, p. 38). Much other information on the reaction is summarized in a review by Gibbons and Newson (1960).

In a series of measurements at the University of Oregon of the neutron energy spectrum produced when this reaction occurs in a thick metal target, a persistent discrepancy appeared between the measured spectrum and that calculated from the yield curve. In view of the continuing importance of this reaction as an absolute neutron flux standard, an evaluation of existing data and the measurements reported here were undertaken in order to resolve the discrepancy.

The Problem

The continuous neutron energy spectrum resulting from production of neutrons to a single final state by protons slowing down in a thick target is given by

$$N(E_{\mathrm{n}},\theta)/N_{\mathrm{p}} = \{\sigma(\theta)/\epsilon\}\mathrm{d}E_{\mathrm{p}}/\mathrm{d}E_{\mathrm{n}} \qquad \text{neutrons proton}^{-1}\,\text{MeV}^{-1}\,\text{sr}^{-1}\,,$$

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where $\sigma(\theta)$ is the differential reaction cross section and ϵ is the compound or molecular stopping cross section per reactive atom in the target (Wylie 1969; Wylie, Bahnsen, and Lefevre 1969). For the ^{7}Li+p reaction, two terms contribute to the neutron spectrum when the proton energy is such that both ground state and first excited state neutrons can be produced.

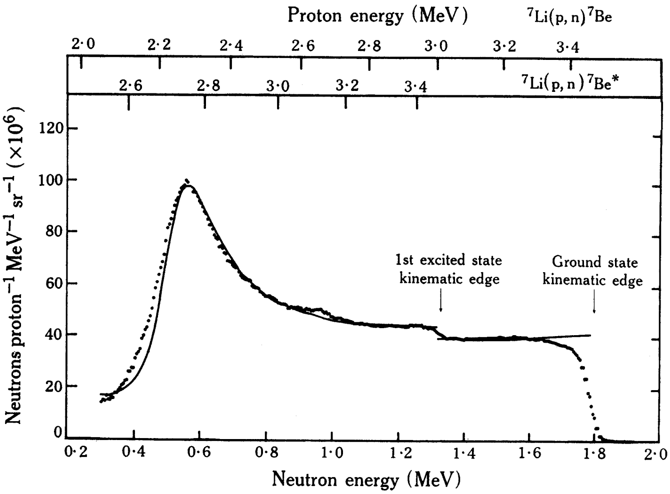

Fig. 1.—Calculated (solid curve) and measured neutron energy spectra produced by bombarding a thick natural lithium metal target with 3·45 MeV protons.

Figure 1 shows the neutron energy spectrum calculated from Marion's plot of the GDB results for $\sigma_{\text{gnd}}(0°)$, and a combination of those results with ratio measurements of Bevington, Rolland, and Lewis (1961) for $\sigma_{\text{1st}}(0°)$. It also shows the spectrum measured at Oregon. The measured spectrum was corrected semi-empirically for the energy variation of recoil proton scintillation counter efficiency ϕ using

$$\phi = \{1-\exp(-n\sigma l)\}(1-B/E)(a+bE),$$

where B, the detector bias, a, and b were used as adjustable parameters to best fit the calculated spectrum. The fit shown was obtained with $B = 0\cdot185$ MeV, $a = 1\cdot02$, and $b = 0\cdot068\,\text{MeV}^{-1}$. Figure 2 shows the ratio (experimental/calculated) of the spectra from Figure 1. Figure 3, which is similar to Figure 1, shows also the result of inserting a 3·6 cm lead filter between the lithium target and the neutron detector. The 525 keV total cross section resonance of lead (Neutron Cross Sections 1964) is prominent in the lower curve and verifies the accuracy of energy calibration of the spectrometer.

The peaks in the ratio shown in Figure 2 are certainly due in part to backscattering into the scintillator of neutrons from oxygen in the photomultiplier faceplate. They occur near neutron energies where ^{16}O has large neutron total cross section resonances (Neutron Cross Sections 1964). It is not possible, however, to explain the relative amplitude of the peaks in the ratio on the basis of scattering from oxygen. The general appearance of Figure 3 suggests that the experimental spectrum is simply displaced relative to the calculated one by about 20 keV neutron energy (18 keV proton energy).

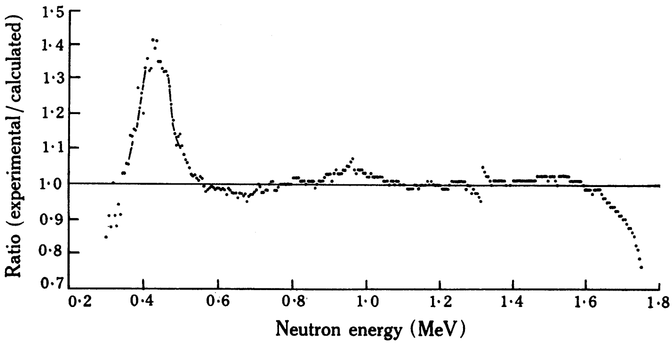

Fig. 2.—Ratio (experimental/calculated) of the spectra from Figure 1.

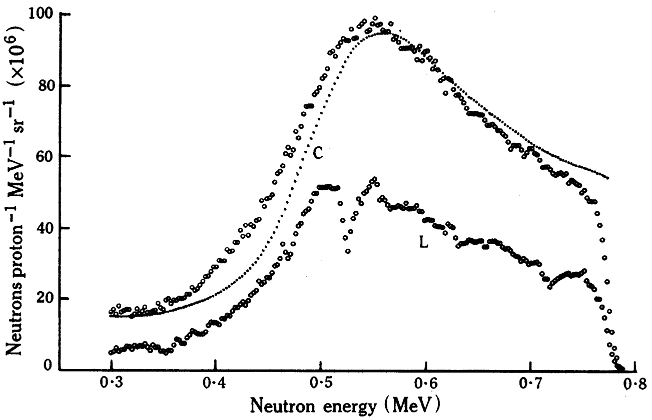

Fig. 3.—Calculated (dotted curve C) and measured neutron energy spectra produced by bombarding the lithium target with 2·52 MeV protons. The lower experimental spectrum (L) was obtained with a 3·6 cm lead filter between the target and the neutron detector.

Existing Data

Besides the classic measurements of Taschek and Hemmendinger (1948) and the GDB results, a careful set of measurements was made in 1962 by Austin (personal communication) using a long counter whose efficiency variation with neutron energy had been studied by Allen and Fergusson (1957). As well as a zero degree yield curve measurement, Austin measured the angular distribution of neutrons from the reaction at 10 bombarding energies and normalized each of these to the 4π results at those energies. He corrected his data for the energy-dependent parameters of the long counter and also spent some effort on evaluation of the relative numbers of direct and inscattered neutrons detected in his measurements.

No statement is made by GDB on either the cross section uncertainty or on the energy uncertainty of their data points. Austin's energies were believed accurate to ± 5 keV and his cross section values (relative ultimately to the neutron–proton cross section) were obtained with an accuracy of $\pm 3 \cdot 5\%$ or better. The normalized GDB angular distribution at $2 \cdot 265$ MeV yielded a $0°$ differential cross section of $154 \cdot 0$ mb sr^{-1}. Austin's measurement at $2 \cdot 270$ MeV gave $149 \cdot 0$ mb sr^{-1}. These results evidently agree within the limits of accuracy of the respective experiments.

Experimental Conditions

Protons were accelerated with the Australian Atomic Energy Commission's 3 MeV Van de Graaff accelerator at Lucas Heights. This accelerator is equipped with a High Voltage Engineering Corporation terminal pulser. While the pulser was not used for these measurements, its presence allowed the possibility of changes (possibly energy dependent) in the angle of injection of the proton beam into the accelerating tube. The magnetic energy analysing system was not equipped with object slits so changes in beam trajectory caused by changes in entrance angle would change the beam energy even though the analysing magnet field remained constant. For these reasons a thorough study of energy calibration and stability was necessary.

Magnetic field measurements in the analysing magnet were made with a proton n.m.r. spectrometer and frequency counter. The resonance stability of the system was better than 2 kHz. Three energy calibration points were used: the ^{7}Li(p, n) reaction threshold at $1880 \cdot 7 \pm 0 \cdot 4$ keV (Nuclear Data Tables 1960), the ^{65}Cu(p, n) reaction threshold at $2164 \cdot 6 \pm 0 \cdot 9$ keV (Marion and Kavanagh 1956), and the Pb(n, n) total cross section resonance at 525 keV mean neutron energy (Neutron Cross Sections 1964). Measurements were made during three two-day periods separated by about one month each. Changes of the r.f. ion source bottle made between the two-day periods resulted in significant changes in magnet calibration but within each two-day period calibration data were consistent within ± 2 keV. It was necessary to forego changes in terminal control settings during any set of runs in order to achieve this calibration stability. The yield data reported here were all obtained during the third two-day period. Eight lithium thresholds measured during that time fluctuated randomly by ± 1 keV from the mean. Two lead transmission measurements taken before and after yield measurements with the thinner of two targets showed transmission dips shifted by less than 4 kHz ($0 \cdot 8$ keV) from one another.

The two targets used were natural lithium fluoride of nominal thicknesses 45 and 90 μg cm^{-2} evaporated onto tantalum backings. Interference colouring indicated excellent uniformity. Yield data indicated a thickness ratio of $1 : 2 \cdot 08$ which was consistent with, but far more accurate than, the thickness ratio obtained by means of the rise curves at threshold. Carbon buildup on the targets was minimized by use of a liquid nitrogen cooled trap which surrounded the target (Bashkin and Ophel 1962). The same trap, biased to -300 V, was used for secondary electron suppression. Loss of target material was minimized by limiting beam currents to 1 μA. The neutron yield at a given bombarding energy decreased by less than 1% from beginning to end of the measurements. The mean proton energy loss in the targets was obtained from the relative shift in the yield curves from the two targets. Near incident proton energies of 2·200 MeV, smooth curves through normalized data from the two targets were 4 keV apart. The target thicknesses were therefore 8 and 16 keV respectively which was also consistent with the rise curves.

Neutrons were counted with a polythylene moderated long counter built according to De Pangher's (1961) design. A 2·54 cm diameter BF_3-filled proportional counter tube with a 0·6 cm thick paraffin jacket was used instead of the 3·81 cm diameter tube normally used with this type of moderator. The pulse height distribution from the tube was excellent. Bias was set at a pulse height one-fifth of that due to the first excited state α-particle group. Stability of the counter and electronics system as well as a truly statistical distribution of counting errors at the 1% level was demonstrated with an internal neutron source.

The general purpose target chamber used for these measurements was far from optimum from the point of view of minimizing scattering of neutrons in the vicinity of the target. The chamber was a 20 cm long brass tube 5 cm in diameter with 1 mm thick walls. The axis of the chamber was normal to the beam direction. The targets, backed with thin tantalum, were clipped to a 0·5 mm thick copper plate whose upper end (3 cm from the beam) was welded to a 1 cm diameter water-cooling manifold. Water-cooling lines entered the chamber through a Perspex plate at the upper end of the chamber. The thin walled, nitrogen-cooled trap was attached to the lower end of the chamber by a Perspex plate. A Perspex viewing port was located at the side of the chamber 4·6 cm from the beam spot. These three Perspex plates of thickness 15 mm and their associated brass flanges subtended a total solid angle of $4\pi\times0\cdot17$ sr near 90° to the beam direction. Since it was not possible to shadow the long counter without also removing a large fraction of inscattered neutrons from the target chamber, one of the largest uncertainties in this experiment arises from corrections for scattered neutrons.

Yield Measurements, Data Normalization, and Results

Yield measurements were carried out on each target with the long counter 1 m from the target where it subtended an angle of about $\pm6°$. Measurements were made in the region of threshold in 5 kHz steps in n.m.r. frequency ($\sim$ 1·2 keV) and were continued above 1900 keV with either 50 or 20 kHz steps. Counts were accumulated for a given charge on target, typically 30–60 μC, as determined with an Elcor model A309A current integrator. After completing a yield curve, the yields at the peak (2280 keV) and valley (2060 keV) were remeasured to check target stability. The threshold was then remeasured and, with the thinner target, the Pb(n, n) transmission resonance was observed.

The ratio of uncorrected yields at the peak and valley of the yield curve from both targets used in these measurements was 5·0. This ratio should be compared with Austin's corrected ratio of 6·2, and the GDB ratio of 5·4. This ratio is an indication of the relative numbers of direct and scattered neutrons detected, since

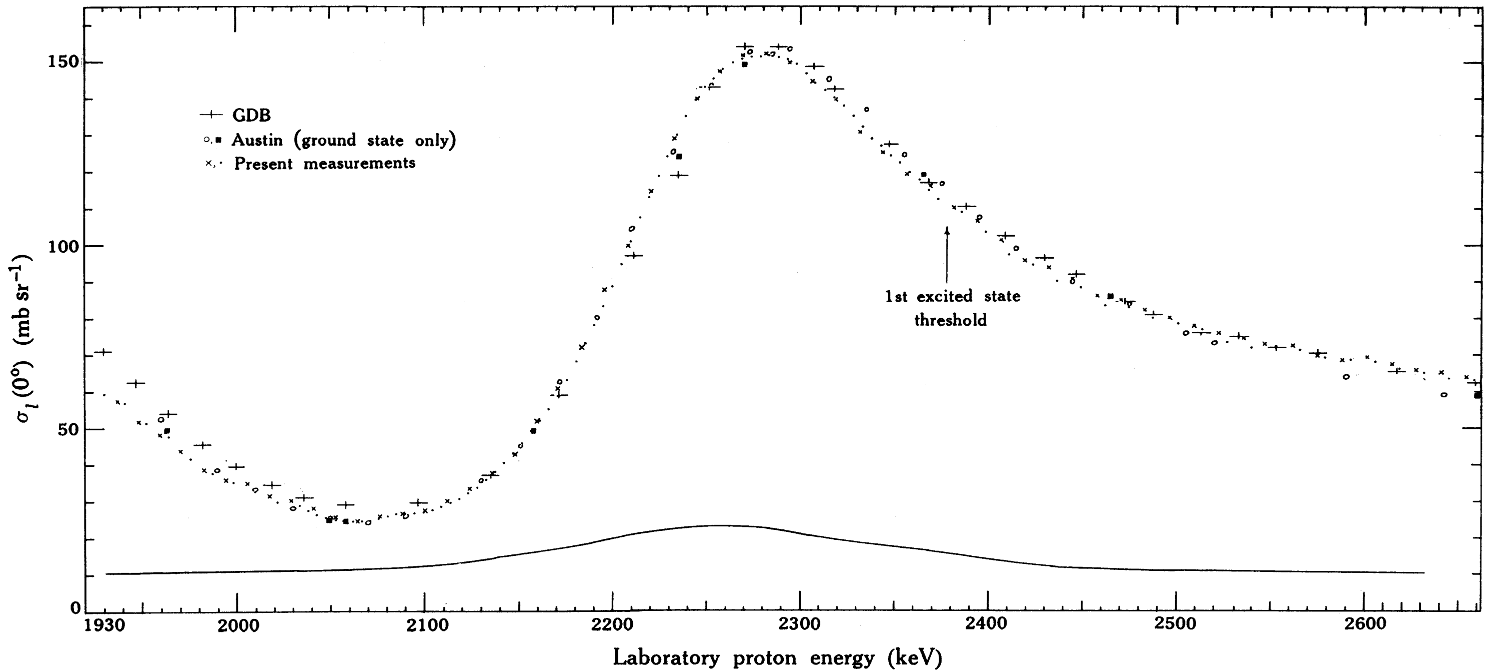

Fig. 4.—Yield curves for the $^7Li(p,n)^7Be$ reaction. The crosses are somewhat smaller than the data points presented in GDB. The solid squares are from Austin's angular distributions as normalized by him to the total reaction cross section from Macklin and Gibbons (1958), while the open circles are Austin's relative measurements as normalized by him to the solid squares. The dots and x's are from the 8 and 16 keV targets normalized to 24·5 mb sr^{-1} at 2060 keV and to 152 mb sr^{-1} at 2280 keV. The solid curve shows the scattering correction which was assumed proportional to the total reaction cross section. Between 2033 and 2373 keV, data from the 8 keV target have been smoothed by a sliding three-point average carried out twice.

the angular distribution of neutrons makes fractional inscattering at the valley much larger than that at the peak. A first-order correction for scattered neutrons was derived on the assumptions that (1) the number of scattered neutrons detected at a given bombarding energy varies only with the total number produced by the target, and (2) Austin's peak to valley ratio is correct. With these assumptions, and with the total reaction cross section it is possible to normalize the data at both the peak and valley. The normalization then gives the ratio of direct to scattered neutrons detected at each bombarding energy.

Figure 4 shows the results of these measurements with the normalization outlined above to $24\cdot5$ mb sr^{-1} at 2060 keV and to 152 mb sr^{-1} at 2280 keV, and the amount of inscattering that the normalization required. The target thickness correction to obtain mean proton energy in the target was made before normalization and consisted of a translation of the data points by half the target thickness at 2200 keV. No correction was made for the variation of long counter efficiency with energy since it is not obvious that De Pangher's (1961) moderator design has the same low energy efficiency variation that conventional long counters do. Our use of a $2\cdot54$ cm diameter BF_3 tube rather than the larger one called for by De Pangher also adds confusion.

For the measurements reported here, the uncertainty in mean proton energy due to calibration, stability, and target thickness corrections is ±2 keV. An unexplained shift in the yield curve by 3 keV between two runs with the thinner target leaves a slight suspicion that the energies reported are systematically high by something less than that amount. A machine shutdown caused by vacuum failure between runs caused discard of the earlier run even though the calibration data gave no particular reason for doing so.

The calibration frequencies from these measurements were (1) the ^{7}Li(p, n) threshold at 16715 ± 5 kHz; (2) the ^{65}Cu(p, n) threshold at 17945 ± 5 kHz; and (3) the Pb(n, n) 525 keV resonance at 18333 ± 5 kHz with the 8 keV target. Mean proton energies were calculated from

$$\langle E_{\mathrm{p}}\rangle = 6\cdot702\times10^{-6}f^2+8\cdot3\ \mathrm{keV}-\tfrac{1}{2}T,$$

where T was the target thickness in keV. In view of the increasing evidence that the ^{65}Cu(p, n) threshold is near 2168 keV (Overley, Parker, and Bromley 1969) rather than the earlier weighted average of $2164\cdot6$ keV (Marion and Kavanagh 1956), energies should probably have been obtained from

$$\langle E_{\mathrm{p}}\rangle = 6\cdot725\times10^{-6}f^2+1\cdot8\ \mathrm{keV}-\tfrac{1}{2}T.$$

This would raise the lowest tabulated energy in Table 1 from $2042\cdot6$ to $2043\cdot2$ keV, and the highest energy from $2362\cdot9$ to $2364\cdot5$ keV.

Evaluation

Since the scattering correction for this experiment was not verified, and since its required size for normalization to Austin's peak to valley ratio is surprisingly large, the cross sections obtained are correspondingly uncertain. An indication of the size of the uncertainty may be obtained by noting that if the scattering correction is not used and data are normalized only at 2280 keV, the cross section at 2060 keV increases from 24·5 to 32 mb sr^{-1} (31%), and at 2200 keV it increases from 89 to 98 mb sr^{-1} (10%). With the results constrained at 2060 and 2280 keV by normalization, the cross section at 2200 keV is probably accurate to better than 7% even if the actual amount of scattering was proportional to the total reaction cross section only to within 30% of the assumed local correction.

As a study of Figure 4 shows, the data sets agree rather well over an energy range substantially larger than that enclosed by the normalization points for this measurement. At the lowest energies, data from this experiment lie below the Austin and GDB results and probably indicate that the De Pangher moderator has a decreased efficiency at low neutron energy similar to (but perhaps smaller than) conventional moderators (Allen 1960).

Between 2030 and 2370 keV the data from the 8 keV target have been smoothed twice by application of a sliding three-point average. This smoothing function has a width comparable with the target thickness. As shown in Figure 4 the unsmoothed data from the 16 keV target agree well with the smoothed data. The smoothed results are contained in Table 1.

TABLE 1

SMOOTHED RESULTS FROM 8 keV TARGET

Only every other data point is tabulated. The scattering correction at 2259 keV was 23·0 and is proportional at other energies to the total reaction cross section

$\langle E_p \rangle$ (keV)	$\sigma_l(0°)$ (mb sr^{-1})	$\langle E_p \rangle$ (keV)	$\sigma_l(0°)$ (mb sr^{-1})	$\langle E_p \rangle$ (keV)	$\sigma_l(0°)$ (mb sr^{-1})	$\langle E_p \rangle$ (keV)	$\sigma_l(0°)$ (mb sr^{-1})
2042·6	26·2	2127·7	33·7	2204·7	94·9	2293·0	150·8
2052·0	24·9	2137·2	37·9	2214·4	107·6	2302·9	146·7
2061·4	24·4	2146·8	43·1	2224·2	118·9	2312·8	142·3
2070·8	24·6	2156·4	49·1	2233·9	130·1	2322·8	138·0
2080·2	25·7	2166·0	55·1	2243·7	139·7	2332·8	132·3
2089·7	26·4	2175·6	63·3	2253·5	145·5	2342·8	127·0
2099·1	26·8	2185·3	73·1	2263·4	149·8	2352·8	122·6
2108·6	28·4	2195·0	83·4	2273·2	151·3	2362·9	117·8
2118·1	30·6			2283·1	151·9		

While not demonstrated here, the differential cross sections contained in Table 1 improve the agreement between measured and calculated thick target neutron spectra substantially although some discrepancies remain. Figure 4 alone, however, indicates that the yield curve from Nuclear Data Tables (1960, p. 38) should not be used in this energy region.

Further efforts to improve the accuracy with which this standard cross section is known should probably await a theoretical analysis of Austin's angular distribution data since the guidance provided by such an analysis would be helpful.

Finally, it is perhaps worth noting that the scattering correction used in this experiment may be applied to the GDB experiment since those data were uncorrected for scattering. Those authors estimated that inscattering from their target chamber at $E_p = 2265$ keV and 0° was less than 5%. At 2060 keV their cross section is then as low as 26 mb sr^{-1}, in good agreement with Austin's value of 24·5 mb sr^{-1}.

Acknowledgments

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