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YIELD AND ENERGY DISTRIBUTION OF NEUTRONS RESULTING FROM THE INTERACTION OF 14-Mev NEUTRONS WITH Li^6 AND Li^7

Work done by:

F. L. Ribe
R. W. Davis
J. M. Holt

Report written by:

F. L. Ribe

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ABSTRACT

The yield and energy distribution of the lower-energy neutrons resulting from interaction of 14-Mev neutrons with Li^6 and Li^7 have been measured in the energy range between 4 and 12 Mev. A coincidence spectrometer was used to detect the neutrons from a scatterer in the form of a spherical shell placed around the neutron source. By use of the spectrometer as a threshold detector, transmission measurements were also made, from which could be determined the elastic cross section for scattered neutrons above 12 Mev. The result is in agreement with the measurement of Graves of the amount of small-angle elastic scattering. Using the known total cross section of Li^6, the known reaction cross sections, and the present measurements, a complete "audit" of the interacted neutrons is made. Qualitatively, one-half of the total cross section is due to small-angle elastic scattering, and all inelastic processes result in lower energy neutrons spread rather uniformly in the region from zero to 12 Mev. Measurement using a thin iron sphere gave a neutron spectrum in good agreement with previous results of Graves and Rosen.

ACKNOWLEDGMENT

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1. Introduction

When 14-Mev neutrons interact with Li^6, very few are lost by absorption processes which replace the neutrons by other light nuclear particles. Previous experiments have shown that the (n, α)[1, 2] and (n, p)[1, 3] reactions amount to only 0.026 and 0.007 barn, respectively, while the total cross section is 1.37 barns.[4] There is a fairly large cross section of about 0.20 barn due to the Li^6 (n, d)He^5 reaction,[1, 2] but each such process results in a neutron "given back" to the system by the breakup of He^5 into an alpha-particle and a neutron, or by a corresponding direct three-body reaction. A large amount of elastic scattering peaked in the forward neutron direction has been demonstrated by an experiment of Graves.[5] The object of the present experiment was to determine the energy spectrum of the lower energy degraded neutrons from large-angle elastic scattering, inelastic scattering, and (n, 2n) processes and to see if all interacted 14-Mev neutrons can be accounted for, in view of the known total cross section.

2. Method and Experimental Arrangement

A scatterer in the form of a spherical shell was placed around a D-T source of 14-Mev neutrons, and the scattered neutrons emitted into a small solid angle were analyzed by means of a coincidence spectrometer. In such an arrangement the spherical geometry effectively integrates over all angles between incident and secondary neutrons, and the energy spectrum $d\sigma/dE$ measured by the spectrometer represents the direction-averaged behavior of the scattering events. If the neutron source, of strength Q neutrons/sec, is isotropic then the cross section $\sigma(E)$ corresponding to a number $n(E)$ of spectrometer counts due to neutrons of energy E in time t is given by

$$\sigma(E) = n(E)\left[\frac{Qt}{4\pi R^2}\,\frac{N(a_1 - a_2)}{V}\,\beta(E)\right]^{-1} \qquad (1)$$

Here R is the distance from the neutron source at the center of the sphere to the spectrometer radiator, N the total number of atoms in the spherical shell, V its volume, a_1 and a_2 its outer and inner radii, and β the sensitivity of the spectrometer, defined by

$$\beta(E) = \frac{\text{spectrometer counts/sec}}{\text{flux of neutrons of energy E incident on radiator}} \qquad (2)$$

Such spherical scatterers, when used with a threshold detector, also provide a well-known means for measuring effective inelastic cross sections. If T is the transmission of

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the sphere measured with the threshold detector

$$T = \frac{\text{detector counts with sphere over source}}{\text{detector counts with no sphere}} \qquad (3)$$

then the effective inelastic cross section σ_i for degradation of neutrons to energies below that of the threshold of the detector and for loss by absorption is given by

$$T = e^{-\frac{N(a_1 - a_2)}{V}\sigma_i} \qquad (4)$$

Here the spherical geometry allows neutrons with energies above the threshold which are scattered away from the detector to be replaced by neutrons above threshold from other directions scattered into the detector.

It should be mentioned that equations (1) and (4) assume no multiple scattering of the neutrons in the spherical shell. The modifications of these equations necessitated by such effects have been studied in detail by Bethe, Beyster, and Carter.[6] The errors introduced into the present results by the use of equations (1) and (4) are discussed in the Appendix.

The source of 14-Mev neutrons was a ZrT target disc, bombarded on a circular area 0.52 cm in diameter by a beam of deuterons from the P-4 Cockcroft-Walton generator. The voltage of the accelerator was set at the relatively low value of 130 kv in order that the neutrons from the $T(d, n)He^4$ reaction should be emitted as nearly isotropically as possible. It was necessary to use a special target tube 0.300 inch in diameter which had no provision for monitoring the neutrons by counting alpha-particles. Therefore source monitoring was done by means of a long counter (with a neutron response which was approximately independent of energy). The multiplication (ratio of long counter counts with sphere on and off for constant Qt) of the lithium spheres used in this experiment has been determined by Graves[5] to be 1.00 ± 0.01. Therefore a 1% error is introduced into the results by using the long counter as a monitor. Figure 1 shows the target tube of the accelerator with a sphere in place. The lithium spheres were of solid metal, 2.00 inches in diameter with a cylindrical hole to the center for admitting the target tube of the accelerator. The distance from center of target tube to the radiator of the spectrometer was 13.6 cm.

3. The Coincidence Spectrometer

The coincidence spectrometer is shown in Fig. 1. It operates in the following manner. Neutrons in a direction along its axis impinge on a thin (12.2 mg/cm^2) polyethylene radiator, and the forward recoil protons travel through the two counters and strike the NaI crystal of

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the scintillation detector. The amplified pulses resulting from the ionization in the two proportional counters and from the scintillation of the sodium iodide crystal are fed to a coincidence circuit. The output pulse of the circuit indicates that a charged particle has traversed the spectrometer. The coincidence output pulse gates open an 18-channel pulse-height analyzer[7] which records in one of its channels the scintillation pulse. The voltage of this pulse determines the energy of the forward recoil proton and hence the energy of the neutron striking the radiator. Such a coincidence arrangement is necessary in order to observe the pulse-height spectrum of the proton scintillations since they occur in the presence of a large background produced by 14-Mev neutrons and gamma-rays striking the scintillator.

The radiator was mounted on a wafer in a vacuum-tight target assembly. At one end of the axle on which this wafer was mounted was a small bar magnet which allowed the wafer to be rotated by an external magnet to expose either the radiator or a platinum blank to the counters. The counters were filled with 17 cm of krypton and CO_2, with 2.5% of the latter, and operated in their proportional range at a multiplication of about 10. The sodium iodide crystal was in the form of a cylinder 1 inch in diameter and 2.5 mm thick with the edge polished to transparency in alcohol and toluene and the two flat surfaces freshly cleaved before insertion of the crystal into the scintillation detector. The crystal was mounted in a glass cup with a plane bottom which fitted into an aluminum light pipe leading to the photocathode of a DuMont type K1177 photomultiplier, as indicated in Fig. 1. With uncollimated plutonium alpha-particles the resolution (total width at half maximum) of the scintillator was 8.0%. The solid angle subtended at the radiator by the last diaphragm aperture before the crystal was 0.0144 std.

4. Tests of Spectrometer Operation

The spectrometer was first run with a bare T-D source, the strength of which was accurately measured by means of counted alpha-particles. The peaked spectrum from the source and room-scattered neutrons was measured, subtracting the background measured with the blank facing the counters. For the radiator used in the lithium measurements and the same source-to-radiator spacing, the resolution of the 14.1-Mev neutron group was 9.1% and the total background in the region of degraded neutrons above about 3 Mev was 11%. Of this, 7% was due to room scattering and 4% to disintegrations of the krypton gas in the first counter. The energy resolution at the lowest energy at which measurements were made (4 Mev) was 21%. This energy spread was due mainly to radiator thickness.

From the known incident flux of the source, the sensitivity factor β(14.1 Mev) of equation (2) was measured to be 1.32×10^{-5}. By using the known differential cross section for (n, p) scattering, the solid angle of the spectrometer, and the exposed radiator weight, the sensitivity factor was calculated to be 1.32×10^{-5}.

The linearity of the system of preamplifier, amplifier, and pulse-height analyzer for the scintillator was tested with a pulser and found to be exact to within 0.5% in absolute voltage and 5% in relative channel width.

It was determined that there were no accidental coincidences at the source strengths used for the lithium runs by operating the scintillator amplifier which fed the coincidence circuit (resolving time = 0.5 μsec) with a 1 μsec delay. This was further checked by observing that the sensitivity factor of the spectrometer did not vary with resolving time between the limits of 0.35 and 0.80 μsec.

To determine an operating point for the proportional counters, a bias curve was measured in which 14.1-Mev (n, p) spectrometer counts were plotted against bias voltage of the counter channels. By suitable choice of counter voltage this bias curve was made flat from zero to 50 volts, and an operating point of 20 volts was chosen. Evidently no counts would then be lost at lower neutron energies.

The spectrometer was mounted on a rotating stand and the angular distribution for (n, p) scattering was found to be isotropic in the center-of-mass system to within about 5% between zero and 90°. The energy variation of the recoil protons with angle was found to follow the required $\cos^2\theta$ relation with laboratory angle.

5. Procedure for Determining the Energy Spectrum of Scattered Neutrons

The following procedure was used in taking the data. The spectrometer was irradiated with the radiator "in" and the spherical scatterer over the D-T source, the total number of emitted source neutrons being monitored with the long counter. The same irradiation was then made with no sphere over the source. The radiator was then rotated to the "out" position and the irradiations with and without scatterer were repeated to give the first counter background. The background analyzer counts were subtracted from the counts with the radiator "in" to give net neutron counts with and without scatterer.

Having obtained the number of neutron counts per analyzer channel for the room background and the room background plus scattered neutrons, one could compute the integral counts above a given neutron energy by adding together all of the analyzer channel counts above a given voltage. From this the transmission as a function of threshold energy was computed and used to correct the background neutron counts to correspond to the fact that with the sphere on there were less 14-Mev neutrons to give rise to low energy room background. The correction amounted to about 5% of the background, while the background itself was for lithium about 0.4 times the background-plus-scattered-neutrons. The corrected

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background counts were then subtracted from the background-plus-scattered-neutron counts to give the net counts due to scattered neutrons.

Account was taken of the energy loss of the recoil protons in the radiator and counter gas to correct the scintillator spectrum to correspond to that of the original recoils. So that these energy losses should not be excessive, the lower energy limit of neutron energy investigated was set at 4 Mev. It was found necessary to apply a small correction to the counts in each channel to take account of its relative narrowing in changing from proton energy at the crystal to original recoil-proton energy. The resulting corrected counts n(E) per analyzer channel (channel width $\approx$0.65 Mev) could then be used in equation (1) to give the cross section per channel. The sensitivity factor $\beta(E)$ was obtained from the value β(14.1 Mev) by using the known energy variation of the (n, p) scattering cross section.[8]

6. Measurement of the Inelastic Neutron Spectrum of Iron

As a final test of the apparatus, the foregoing procedure was used to measure the spectrum of inelastic neutrons from iron. The iron spherical shell used had an outer diameter of 3.00 inches, a thickness of 0.25 inch, and a mass of 757 grams. Its inelastic transmission of about 93% was comparable with the transmissions of the lithium spheres for all inelastic processes. The long-counter multiplication of the iron sphere was 1.00 ± 0.01.

The results of the spectrum measurement are shown in Fig. 2 in which $d\sigma/dE$ is plotted as a function of neutron energy, using the solid circles. The results of Graves and Rosen,[9] who used photographic-plate techniques and a spherical scatterer, are plotted for comparison, and it is seen that there is good agreement between the two measurements.

At a neutron-source-to-radiator distance of 13.6 cm, which was necessary for sufficient inelastic neutron yield, a rather large angle was subtended at the radiator by the spherical shell. Because of the cos θ angular distribution of the proton recoils, neutrons from various parts of the scatterer affected the spectrometer with different sensitivities. This effect amounted to about 2% and a transmission measurement with an accuracy comparable to that of the spectrum measurement was therefore not possible.

7. Results of the Li^6 and Li^7 Measurements

Figure 3 is a graph of the data for the irradiations with and without the Li^6 sphere. The data shown are the analyzer channel readings corrected by subtracting the backgrounds taken with the radiator "out." The attenuation of the primary 14.1-Mev neutron group is apparent, and one can see that the neutrons lost from the primary group are spread, more or less evenly, in the lower-energy channels. In determining the energy spectra, care was taken not to attempt to use the data so close to the large primary peak that small drifts of the electronic apparatus would induce spurious differences in subtracting the no-sphere data from that taken with the sphere on. This precaution resulted in cutting off the spectra at an upper energy of about 12 Mev.

The final neutron spectra for Li^6 and Li^7 are plotted in Fig. 4. The enriched Li^6 sphere contained 11% Li^7, and the Li^7 sphere was the normal mixture containing 7.4% Li^6. A first-order correction was made for the smaller fraction in each case, and the results shown represent the behavior of the pure isotopes. The limits of error indicated on the graph give the statistical uncertainties.

The ordinates are labeled "effective $d\sigma/dE$" since the use of equation (1) implies that one neutron is emitted per inelastic event. Since some (n, 2n) processes may have occurred, one must qualify the term $d\sigma/dE$. However it is likely that most of the (n, 2n) neutrons have energies less than 4 Mev, and the ordinates are correspondingly fairly close approximations to the true differential cross sections, in addition to describing the energy distributions in a rigorous manner. The integrated cross sections for those processes which produce secondary neutrons with energies between 4.0 and 12.0 Mev are, subject to the above qualification, 0.33 ± 0.03 and 0.29 ± 0.02 barn for Li^6 and Li^7, respectively.

The corrected counts n(E) for each analyzer channel with and without scatterer were separately added as described earlier, and the corresponding quantities were divided to give the transmissions of the spheres as functions of threshold energy. The transmissions at a threshold energy of 12.0 Mev are of particular interest since this is the upper limit of the present neutron-spectrum measurements. For this threshold energy the transmissions of the enriched Li^6 sphere and the normal Li^7 sphere were 0.934 ± 0.017 and 0.940 ± 0.015, respectively. The corresponding effective inelastic cross sections, corrected to correspond to the pure isotopes Li^6 and Li^7, are 0.68 ± 0.17 and 0.59 ± 0.15 barn. Of more interest are the cross sections for elastic scattering of neutrons to energies between 12.0 and 14.1 Mev, obtained by subtracting these inelastic cross sections from the total cross sections. One obtains:

$$\sigma_{el}(12.0 \leq E \leq 14.1) = 0.69 \pm 0.17 \text{ for } Li^6$$

and

$$\sigma_{el}(12.0 \leq E \leq 14.1) = 0.86 \pm 0.15 \text{ for } Li^7.$$

8. Discussion of the Interaction of 14-Mev Neutrons with Li^6

A comparison can be made between the quantity $\sigma_{el}(12.0 \leq E \leq 14.1)$ of the last paragraph and the integrated cross section for forward elastic scattering measured by Graves.[5] In the latter experiment a direct measurement of the relative differential angular distribution

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of the elastically scattered neutrons was made for laboratory angles from zero to 38 degrees. In the absence of an accurate absolute scale for these measurements it is necessary to normalize the results. Such a normalization is provided by the following relation between the differential cross section for zero-degree scattering in the center-of-mass system, the center-of-mass incident neutron wavelength, and the total cross section:

$$\left(\frac{d\sigma_{el}}{d\Omega}\right)(0) \geqq \left(\frac{\sigma_T}{2\lambda}\right)^2 \qquad (5)$$

This relation, which gives a lower limit for the differential cross section, is due to Wick[10] and depends only upon quite general properties of the quantum-mechanical theory of scattering. Normalizing Graves' data at zero degrees to Wick's limit of 0.593 barn/steradian, one obtains

$$\sigma_{el}(12.0 \leq E \leq 14.1 \text{ Mev}) = 0.79 \pm 0.08 \text{ barn}$$

The agreement between this and the value 0.69 ± 0.17 barn obtained in the present experiment shows that the forward differential cross section cannot far exceed Wick's limit and allows the assignment of 0.08 barn as the probable error in the integrated cross section obtained from Graves' data.

In Fig. 5 a somewhat qualitative graph is given of the complete spectrum of secondary neutrons from the interaction of 14-Mev neutrons with Li^6. The solid curve from 12.7 to 14.1 Mev represents the normalized data of Graves with the experimental points, transformed to millibarns/Mev (laboratory coordinates), indicated. The dashed curve at these high energies is the corresponding spectrum as it would have appeared if measured with the 9% energy resolution of the present experiment. From this it can be seen that the upper end of the inelastic spectrum (dashed line at end of solid line from 4 to about 12 Mev) measured in the present experiment probably appears to rise because of elastic neutrons degraded somewhat by resolution.

The lower dashed curve between zero and 4 Mev represents the spectrum of neutrons from the (n, d) reaction deduced by Frye from the angular distribution of deuterons from this reaction and the dynamics of the process.

One can now draw up a "balance sheet" for the neutrons. The following processes: high-energy elastic scattering, medium-energy inelastic scattering, (n, d) processes giving rise to low energy neutrons, and the (n, α) and (n, p) reactions, account for 1.28 barns of the total cross section. One is left with 0.09 barn which must be assigned to the energy region between zero and 4 Mev. This assignment is made on the graph of Fig. 5 by arbitrarily adding one-fourth of this quantity per 1-Mev interval to the known energy distribution

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of the low-energy (n, d) neutrons. The resulting low-energy curve is seen to provide a reasonable continuation of the energy distribution measured above 4 Mev.

The Li^6 nucleus has known energy levels at 2.19 and 3.58 Mev.[11] From the present results it appears that any inelastic neutron scattering which leaves these levels excited does not give rise to strongly peaked neutron distributions such as that which occurs for the elastic scattering, but probably gives neutron groups spread in energy over a range comparable to that allowed by the dynamics of the reaction.

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APPENDIX

In the case of scattering of 14-Mev neutrons by lithium, multiple-scattering effects are lessened because of the fact that the elastic scattering is peaked strongly forward. The fact that the spherical shells used were thin was also favorable in this respect.

In calculating the error arising from the use of equation (4) it is sufficient, in view of the 94 per cent transmission of the lithium shells, to carry out the computations to second order in $\sigma_i x$ (x = atoms/cm^2 of spherical shell). A calculation of all inelastic processes including those following elastic scattering (using the elastic angular distribution shown in Fig. 5) showed a departure of the transmission from the exponential function of equation (4) by only 0.02 per cent. This corresponds to an error in the derived inelastic cross section of 0.3 per cent.

In calculating the error arising from the use of equation (1) one must also include in the second-order processes those events which follow inelastic scattering. The contribution of these events was calculated by assuming isotropic inelastic scattering and also assigning the energy distribution of Fig. 4 to elastic and inelastic processes for all neutron energies. I. e., it was assumed that for a neutron of any energy E the resulting energy distribution was divided equally between a group concentrated elastically at energy E and a uniform smear of neutrons extending from zero to E. The error arising in the inelastic yield at 4 Mev (the worst case) was about 6 per cent. This also represents approximately the error introduced into the results for the iron scatterer by the use of equation (1).

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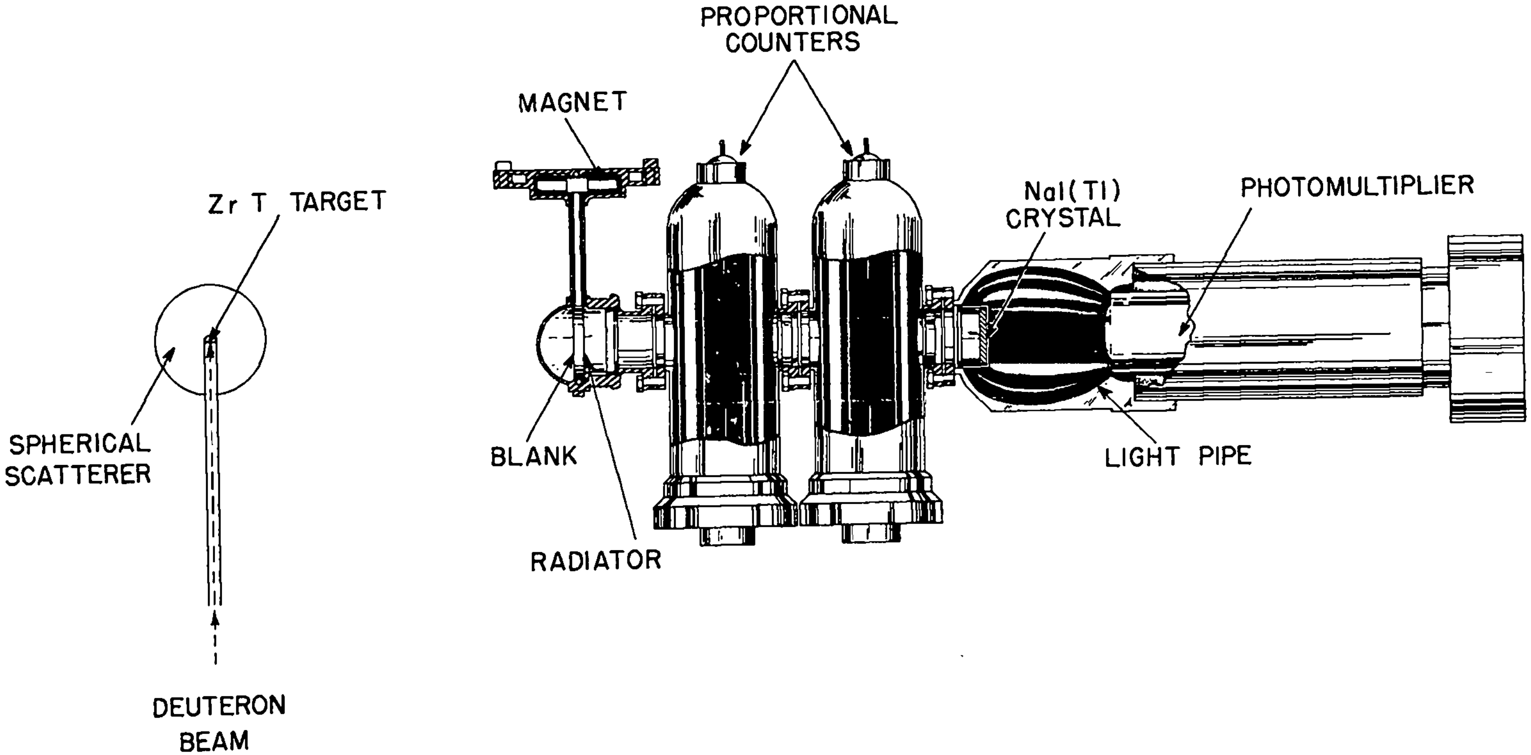

Fig. 1 Experimental arrangement showing coincidence spectrometer, 14-Mev neutron source, and spherical scatterer.

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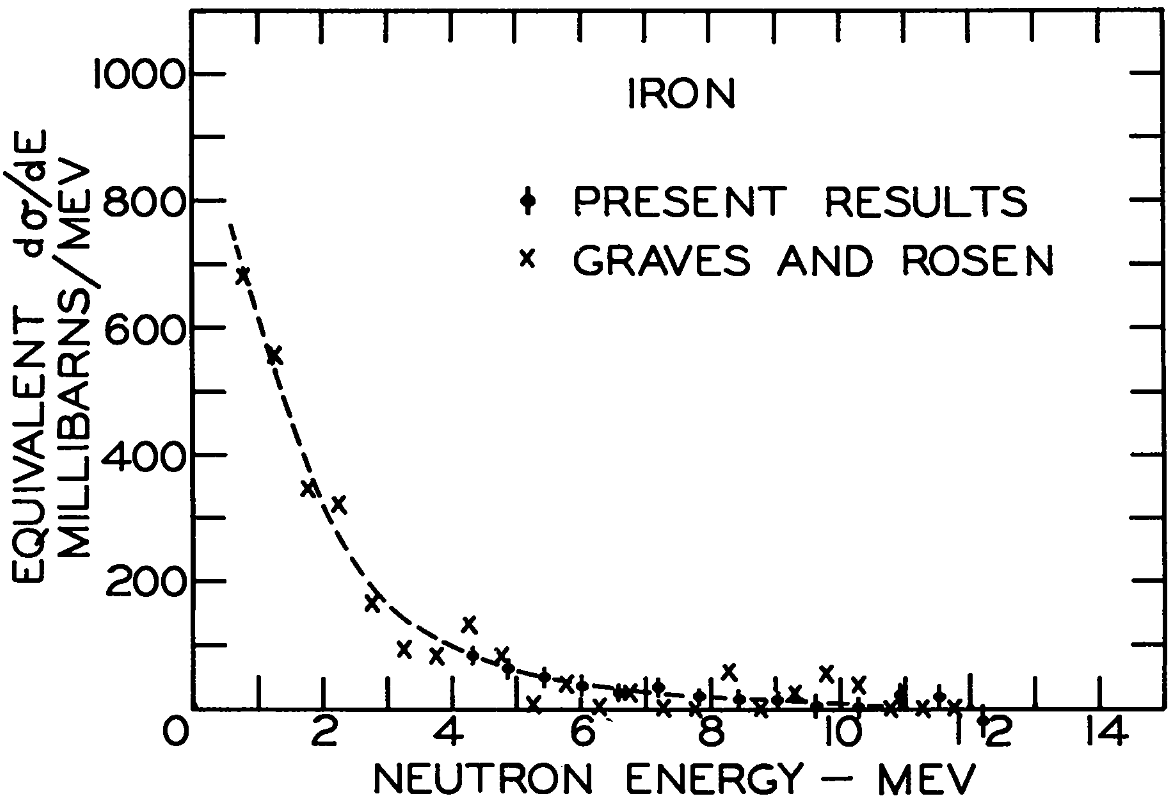

Fig. 2   Yield and energy distribution of inelastic neutrons resulting from the interaction of 14-Mev neutrons with iron.

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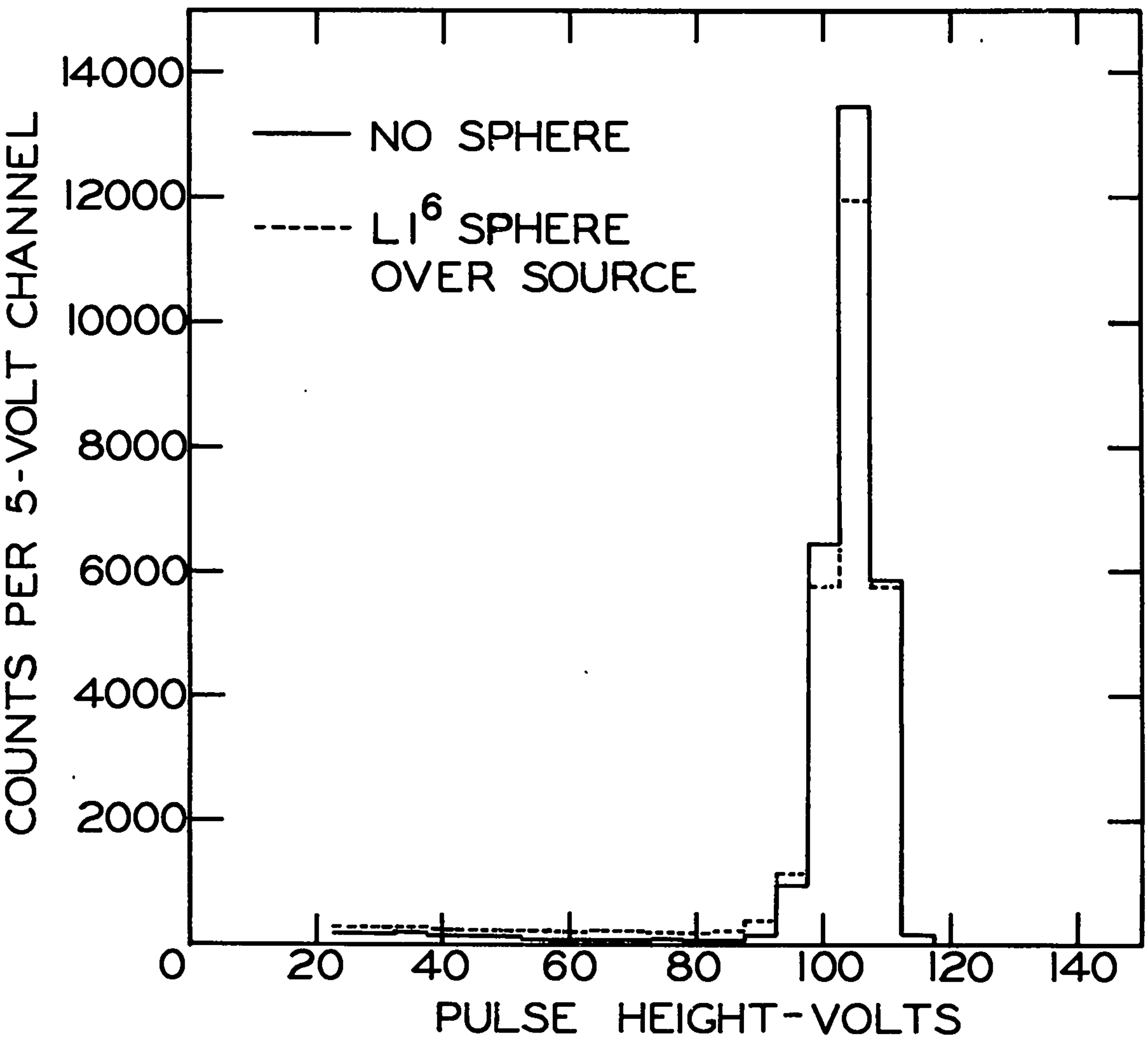

Fig. 3 Pulse-height distributions from irradiations of coincidence spectrometer by 14-Mev neutrons with and without spherical Li^6 scatterer.

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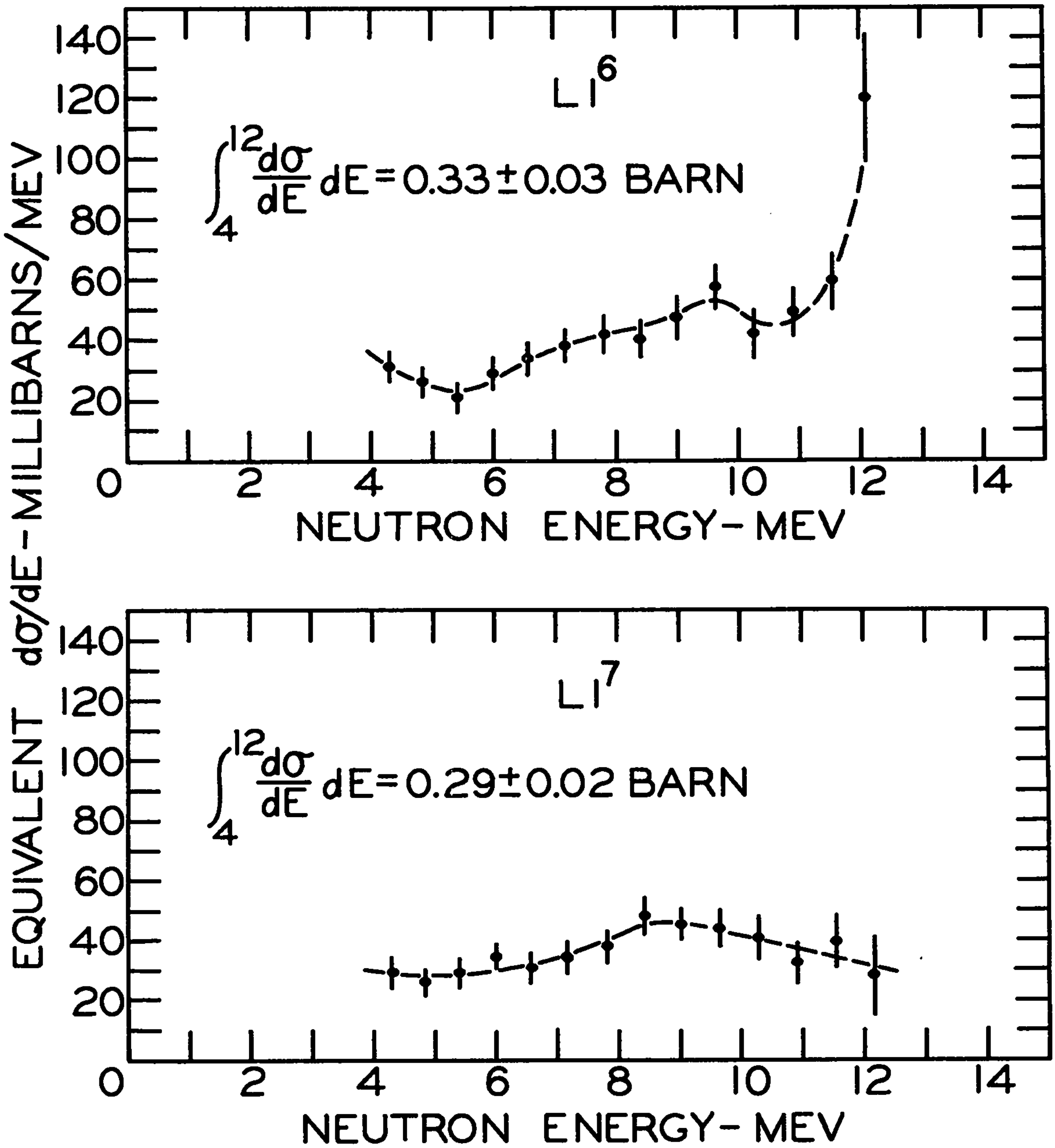

Fig. 4 Yields and energy distributions of neutrons in the energy range from 4 to 12 Mev resulting from the interaction of 14-Mev neutrons with Li^6 and Li^7.

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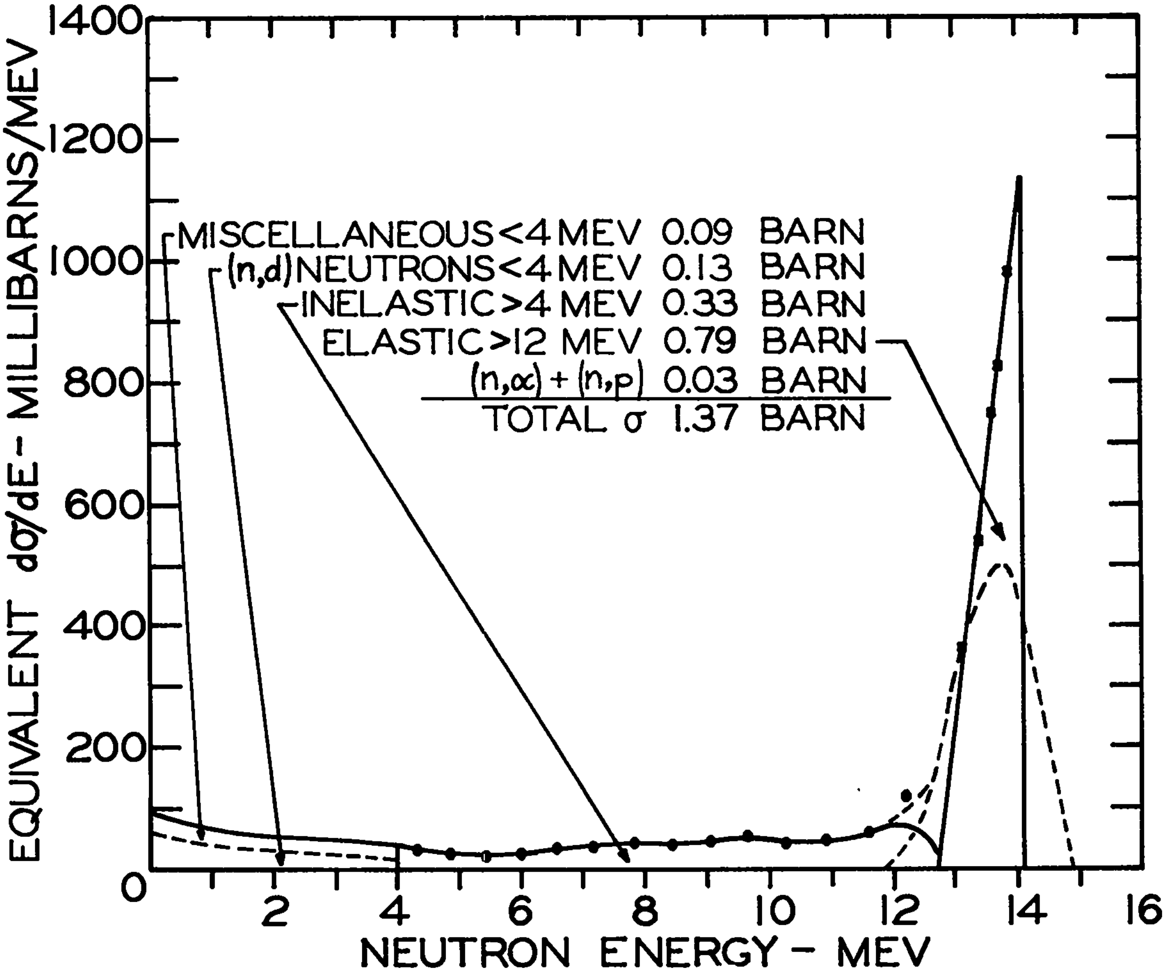

Fig. 5 Qualitative complete spectrum of neutrons resulting from the interaction of 14-Mev neutrons with Li^6. The "inelastic" group of neutrons in the energy range between 4 and 12 Mev includes large-angle elastically scattered neutrons.

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