

Elastic and Inelastic Scattering of Fast Neutrons from Th^{232} †

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The elastic and inelastic scattering of neutrons from thorium is studied throughout the incident neutron energy interval 0.3 to 1.6 Mev. Fast time-of-flight techniques are utilized in conjunction with an electromagnetic pulsing and bunching system for the production of short and very intense neutron bursts. The differential elastic cross section is determined as a function of incident neutron energy. The cross sections for inelastic scattering resulting in the excitation of residual nuclear levels at 50 ± 2 , 170 ± 10 , 720 ± 20 , 790 ± 20 , 820 ± 20 , 1050 ± 50 , and 1150 ± 50 kev are measured. The angular distribution of inelastically scattered neutrons resulting in the excitation of the lower two nuclear levels is determined. The measured individual inelastic excitation functions are combined to obtain the total inelastic scattering cross section as a function of incident neutron energy. The experimental results are compared with the results of other measurements.

INTRODUCTION

THE low energy nuclear structure of thorium is characteristic of that resulting from collective nuclear motions in heavy even-even nuclei.¹ The low-lying excited states of thorium have been studied utilizing Coulomb excitation techniques and alpha particle spectroscopy.²⁻⁴ These Coulomb excitations and alpha particle experiments are limited by available incident particles, energetics, and restrictive selection rules. Excitation of the lower energy levels in thorium by inelastic neutron scattering has been studied only indirectly or qualitatively.⁵⁻⁸ As a result, the respective inelastic scattering excitation functions are not well known. Several groups have experimentally examined elastic and total neutron scattering from thorium.^{9,10} However, the experimental techniques employed did not clearly distinguish between elastic neutron scattering and inelastic neutron scattering leading to the excitation of low-lying levels in thorium. As a result, the measured elastic scattering cross sections are contaminated with varying and/or uncertain contributions from the inelastic scattering processes.

Theoretical consideration of scattering processes in heavy nuclei, such as thorium, contributes to an understanding of the average properties of compound nuclear reactions.^{11,12} In addition, fast neutron scattering from thorium is of considerable applied interest, since this material is the fertile component of many breeding reactor concepts. Thus, the experimental study of fast

neutron scattering from thorium not only provides basic nuclear data which contributes to a better understanding of complex, heavy nuclei but also furnishes information of considerable applied value.

EXPERIMENTAL METHOD

This experiment utilized the pulsed beam fast neutron time-of-flight technique.¹³ The initial portion of the work was carried out with the experimental arrangement shown in Fig. 1(a). A Van de Graaff accelerator combined with a magnetic analyzing system provided an essentially mono-energetic proton beam.¹⁴ This beam passed through a pair of electrostatic deflection plates. A sinusoidal voltage applied to the deflection plates swept the beam across a small aperture several meters distant. The protons going through the aperture struck a lithium target, producing a burst of neutrons through the $\text{Li}^7(p,n)\text{Be}^7$ reaction. The duration of the burst was adjustable from 1 to 5 μsec . The lithium target was varied in thickness from 10 kev to 40 kev depending on the incident neutron energy spread desired in the particular measurements.

Some of the neutrons leaving the target struck the nearby sample and were scattered down the collimated flight path to the neutron detector. The cylindrical scattering samples were of such a size that the transmission was never less than 60% and usually $\sim 80\%$. The flight path from the sample to the detector was variable from 1 to a maximum of $2\frac{1}{2}$ m. The distance from the target to the sample was ~ 10 cm. The neutron detectors were plastic scintillators 1–2 cm thick coupled to RCA 7264 photo multiplier tubes.¹⁵ The phototubes were cooled to $\sim 20^\circ\text{F}$ in order to reduce the tube noise level. With this cooling procedure it was possible to detect neutrons with energies as low as 120 kev. The time between an electronic fiducial marker associated with the neutron burst and the arrival at the detector of

† This work supported by the U. S. Atomic Energy Commission.

¹ A. Bohr, Kgl. Danak Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 14 (1952).

² F. S. Stephens, Phys. Rev. Letters **3**, 435 (1959).

³ P. H. Stelson and F. K. McGowan (to be published). See also T. Huus, Nuclear Instr. and Methods **11**, 26 (1961).

⁴ I. Perlman and F. Asaro, Ann. Rev. Nuclear Sci. **4**, 157 (1954).

⁵ S. Berko, Bull. Am. Phys. Soc. **4**, 257 (1959).

⁶ R. Day (private communication).

⁷ R. Batchelor, Proc. Phys. Soc. (London) **A69**, 214 (1956).

⁸ J. E. Evans, Can. J. Phys. **37**, 396 (1959).

⁹ A. Langsdorf *et al.*, Argonne National Laboratory Report ANL-5567, (unpublished); Phys. Rev. **107**, 1077 (1957). Also, see R. O. Lane *et al.*, Ann. Phys. **12**, 135 (1961).

¹⁰ M. Walt and H. H. Barschall, Phys. Rev. **93**, 1062 (1954).

¹¹ A. Lane and R. Thomas, Revs. Modern Phys. **30**, 257 (1958).

¹² P. Moldauer, Phys. Rev. **123**, 968 (1961).

¹³ L. Cranberg, *Proceedings of the First International Conference on Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 4, p. 40.

¹⁴ High Voltage Engineering Corp., Type KN Accelerator, Burlington, Massachusetts.

¹⁵ Plastic Scintillator, Pilot B. Pilot Chemical Company, Waltham, Massachusetts.

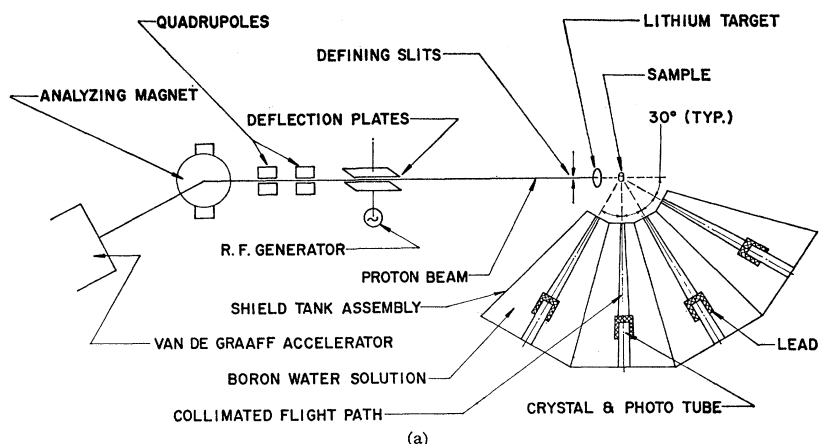
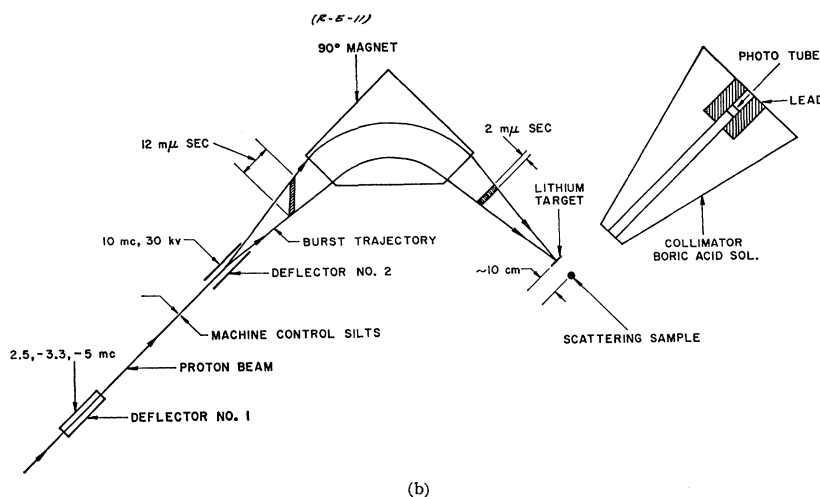


FIG. 1(a). The simple pulsed beam apparatus employed in the initial portions of the experiment. (b). The experimental apparatus including the electromagnetic bunching system.¹⁷⁻¹⁹



the scattered neutron was a measure of neutron velocity and thus energy. This time interval was converted in an analogous manner to a voltage pulse which was recorded in a large multichannel pulse height analyzer.¹⁶ Two detector arrangements were employed. In the first, two or more detectors were simultaneously operating situated at different scattering angles and flight paths. In the second mode of operation a single detector was rotated about the scattering sample. This latter arrangement was utilized in studies of the angular distribution of scattered neutrons.

In the latter part of the experiment an electromagnetic bunching system was employed to produce a more intense neutron burst.¹⁷⁻¹⁹ The experimental apparatus, including the bunching system, is shown in Fig. 1(b). A proton beam from the accelerator passed through two electrostatic deflection systems arranged in such a manner that the beam was periodically

deflected across the entrance face of a large magnet (focal length 60 in., particle radius 30 in.). The time between sweeps was adjustable from 100 μsec to 400 μsec . The sense of the sweep was such that protons entering the magnetic field first in time followed the longest trajectories within the field while those entering at successively later times proceeded along correspondingly shorter trajectories. The proton beam was magnetically deflected through 90 deg and focused, with unit magnification, at the lithium target. By proper adjustment of the instrument parameters, ions entering the magnet sequentially in time and with the same speed were made to arrive at the target essentially simultaneously. The system thus focused particles in both space and time. The angular spread of the protons arriving at the target resulted in a negligible neutron energy spread in comparison with the minimum 10 kev lithium target thickness. The particular apparatus used in this work could accept an 11 μsec sweep at the magnet entrance and produce a target burst of 1 μsec with a peak pulse current of 1.5 ma and a duty cycle of $\leq 1\%$.

The experimental apparatus was so arranged that

¹⁶ Both vacuum tube and transistorized time converters were employed, all designed within the laboratory.

¹⁷ R. C. Mobley, Phys. Rev. **88**, 360 (1952).

¹⁸ R. Holland, Nuclear Instr. and Methods (to be published).

¹⁹ The magnetic portion of the pulsing system supplied by High Voltage Engineering Corporation, Burlington, Massachusetts.

long counters monitored the target yield at all times.²⁰ The detector, collimator, and target assembly were constructed so that the lithium target could be placed at the collimator focus. Under these conditions, the plastic scintillator looked directly at the neutron-producing target. By monitoring the target yield and varying the incident proton and thus the neutron energy, it was possible to determine accurately the energy-dependent sensitivity of the scintillator relative to the response of a long counter. The sensitivity was normally determined at the beginning of each set of measurements and periodically checked as the work progressed.

EXPERIMENTAL METHOD AND INTERPRETATION

All of the measurements reported in this paper were made relative to the differential elastic scattering cross section of carbon as determined by Langsdorf *et al.*⁹ This cross section was experimentally verified at several selected incident neutron energies in the course of this work. In experimentally comparing the various thorium cross sections with that of carbon, the energy of the neutrons incident on the carbon sample was adjusted so as to obtain the same scattered neutron energy at the detector as that resulting from the thorium scattered neutron group of interest. In this

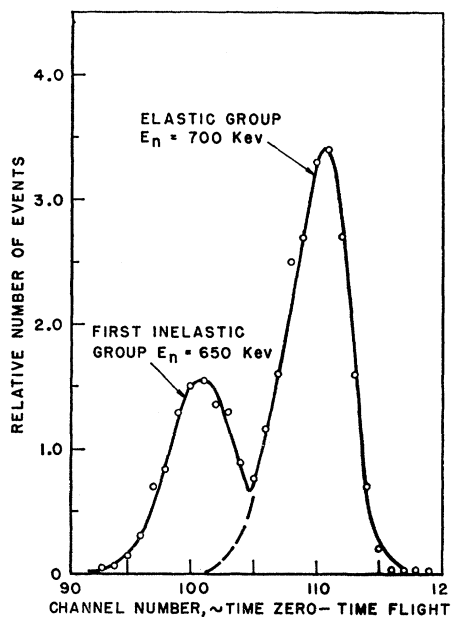


FIG. 2. The experimentally determined distribution in time of 700 kev neutrons scattered from thorium. The larger peak corresponds to the elastically scattered neutron group while the small peak is due to inelastically scattered neutrons leaving the residual nucleus in the first excited ($2+$) state at 50 kev. The dashed line is the extrapolated elastic peak obtained by comparison with the pure elastic scattering from bismuth.

²⁰ A. Hanson and J. McKibben, *Phys. Rev.* **72**, 673 (1947).

manner any errors due to uncertainties in the energy sensitivity of the neutron detectors were avoided.

All the measurements were corrected for incident beam attenuation and multiple scattering perturbations within the samples. The attenuation corrections were carried out in a straightforward manner. An iterative Monte-Carlo technique was used to obtain the multiple scattering correction factors for both elastic and inelastic scattering.²¹ The Monte-Carlo calculations considered elastic events and inelastic events leading to the excitation of a given residual nuclear level. The Monte-Carlo calculations also corrected for the small variation in scattering angle due to the finite angle subtended by the sample at the source. The largest multiple scattering corrections, corresponding to the maximum sample size and forward scattering angles, amounted to no more than 20%. The average correction was ~ 5 –10%. These correction factors are believed known to 5–10%.

At a given incident neutron energy, scattered neutrons with energies $\lesssim 200$ kev were not accepted for measurement. Although the neutron detector was sensitive to lower energy neutrons, it was felt that uncertainties in the detector sensitivity below 200 kev might lead to spurious results. It was estimated that neutron groups with total reaction cross section $\lesssim 50$ –100 mb would be difficult to observe. Only one neutron group, corresponding to the excitation of a level at ~ 950 kev, fell within this region of limited sensitivity. Its existence is questionable. Since the neutron source used throughout the experiment was the $\text{Li}^7(p,n)\text{Be}^7$ reaction, a second source neutron group was present above an incident proton energy of 2.4 Mev.²² This

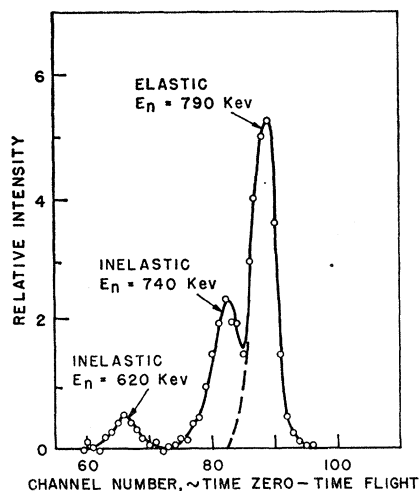


FIG. 3. Experimental time distribution of 790 kev neutrons scattered from thorium. The elastically scattered component and inelastically scattered groups leaving the residual nucleus in the 50 kev ($2+$) and 170 kev ($4+$) states are clearly evident.

²¹ For an outline of methods, see E. Cashwell and C. Everett, *A Practical Manual on the Monte-Carlo Method* (Pergamon Press, New York, 1959).

²² F. Ajzenberg and T. Lauritsen, *Revs. Modern Phys.* **27**, 77 (1955).

second group effectively created an experimental "blind" spot corresponding to a thorium residual nuclear level, at 470 ± 20 kev. Inelastic scattering from thorium resulting in the excitation of levels in this region would not be observed unless the respective differential cross section exceeded $\sim 20\%$ of the elastic cross section at the same angle.

EXPERIMENTAL RESULTS

Experimentally measured distributions of scattered neutron arrival times at the detector are shown in Figs. 2, 3, and 4. In Fig. 2 elastically scattered neutrons and inelastically scattered neutrons resulting in the excitation of the first excited state of 50 ± 2 kev are clearly resolved. These two components are distinguishable up to incident neutron energies of ~ 1.0 Mev. Figure 3 shows a typical result of an experimental measurement of inelastic scattering resulting in the excitation of the $4+$ level at 170 ± 10 kev. In this measurement the sample size, target thickness, and flight path were chosen to provide optimum sensitivity to this inelastic component. As a result, the inelastic group scattering to the first level at 50 kev is not as clearly resolved as in Fig. 2. Figure 4 clearly shows the triplet nature of the thorium level structure in the neighborhood of 800 kev. At this incident neutron energy of 1400 kev the residual levels at 720 ± 20 kev and 820 ± 20 kev are approximately equally excited, while the intermediate state at 790 ± 20 kev has approximately twice the probability of occurrence of either of the other two.

In addition to the inelastic neutron groups shown in Figs. 2-4, two groups corresponding to the excitation of levels in thorium at 1050 ± 50 and 1150 ± 50 were seen. The excited nuclear levels in thorium at 50 ± 2 , 170 ± 10 , 720 ± 20 , 790 ± 20 , 820 ± 20 , 1050 ± 50 , and 1150 ± 50 kev observed in this work are graphically summarized and compared with results of other studies in Fig. 5.

The elastic scattering cross section of thorium was measured at six different incident neutron energies

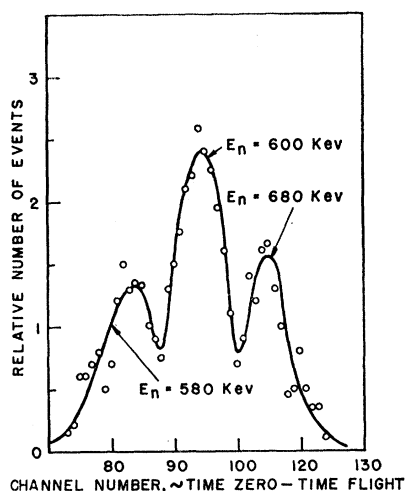


FIG. 4. Experimental measurement of inelastic scattering of 1.4 Mev neutrons from thorium resulting in the excitation of nuclear levels at 720, 790, and 820 kev. The intensity ratios are estimated at 1:2:1.

extending from 570 kev to 1.5 Mev. Some of the results are shown in Fig. 6. The measurements above an incident neutron energy of 1.0 Mev have been corrected for the contribution of the first inelastic group using the extrapolated inelastic cross section given in Fig. 10 (which will be discussed later). At lower incident energies the elastic component is experimentally resolved from all inelastic components. The solid curves in Fig. 6 are the results of a least-square fit of the following expression to the experimental data

$$\frac{d\sigma(\text{el})}{d\Omega} = \frac{\sigma(\text{total, el})}{4\pi} \times (1 + W_1 P_1 + W_2 P_2 + W_3 P_3 + W_4 P_4), \quad (1)$$

where $d\sigma(\text{el})/d\Omega$ is the differential elastic scattering cross section, W_i are energy dependent coefficients, and P_i are

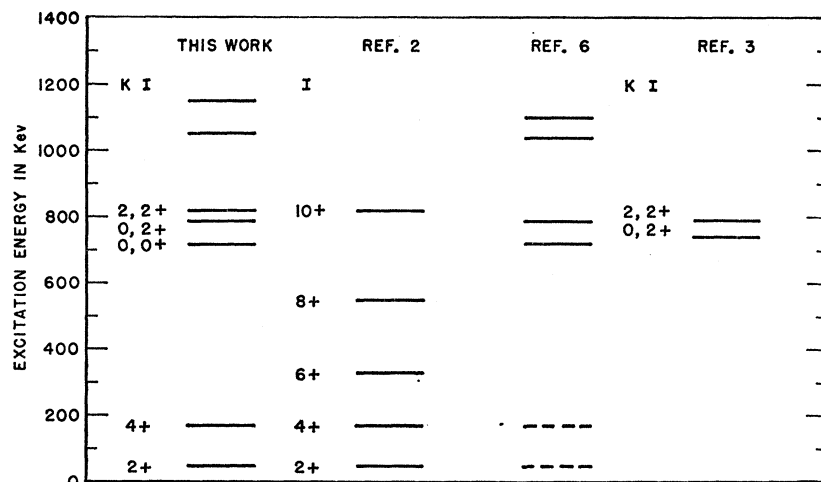


FIG. 5. Low energy nuclear levels in Th^{232} . The results of this experiment are compared with those obtained by other workers.^{2,3,6}

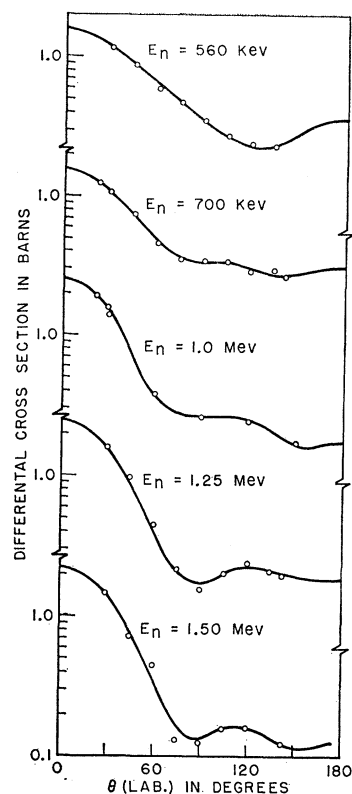


FIG. 6. The differential elastic scattering cross section of thorium as a function of incident neutron energy. Representative data points are shown.

the Legendre polynomials.²³ The energy dependence of $\sigma(\text{total, el})$ and the W_i is given in Fig. 7. The errors in $\sigma(\text{total, el})$ are estimated to be approximately 12%. This error includes an 8% uncertainty in the differential scattering cross section of carbon used as a standard. As indicated in Fig. 6, the individual angular measure-

ments extended from a forward angle of 22° to 145°. The average value of the cosine of the scattering angle

$$\langle \cos \theta \rangle_{\text{av}} = \bar{\mu} = \frac{1}{3} W_1, \quad (2)$$

and the elastic transport cross section²⁴

$$\sigma(\text{tr, el}) = \sigma(\text{el})(1 - \bar{\mu}) = (\sigma(\text{el}))(1 - \frac{1}{3} W_1), \quad (3)$$

are frequently used quantities in applied work. They follow directly from expression (1) and the experimental results of Fig. 7.

The cross sections for the excitation of individual residual nuclear levels in throrium by inelastic neutron scattering are given in Fig. 8 as a function of incident neutron energy. The errors shown in the figure represent estimates of the accuracies of individual measurements. They include the 8% error placed upon the differential cross section of carbon used as standard. The differential inelastic cross section for neutron scattering resulting in the excitation of each of the first two levels in the thorium was examined in detail. At incident neutron energies of 600 kev and 800 kev, inelastic scattering to the 170 kev level was found to be isotropic. At incident energies of 560 kev, 760 kev, and 1000 kev the angular distribution of inelastically scattered neutrons leaving the residual nucleus in the 50 kev 2+ state was non-isotropic, but it was symmetric about 90°. A typical measured differential cross section is shown in Fig. 9. The solid curve in the figure represents the expression

$$\frac{d\sigma(\text{inel})}{d\Omega} = \frac{1.1 \sigma(\text{total, inel})}{4\pi} (1 + b \cos^2 \theta), \quad (4)$$

where $b = -0.3$. $\sigma(\text{total, inel})$ in expression (4) is the inelastic cross section for scattering to the 2+ state as

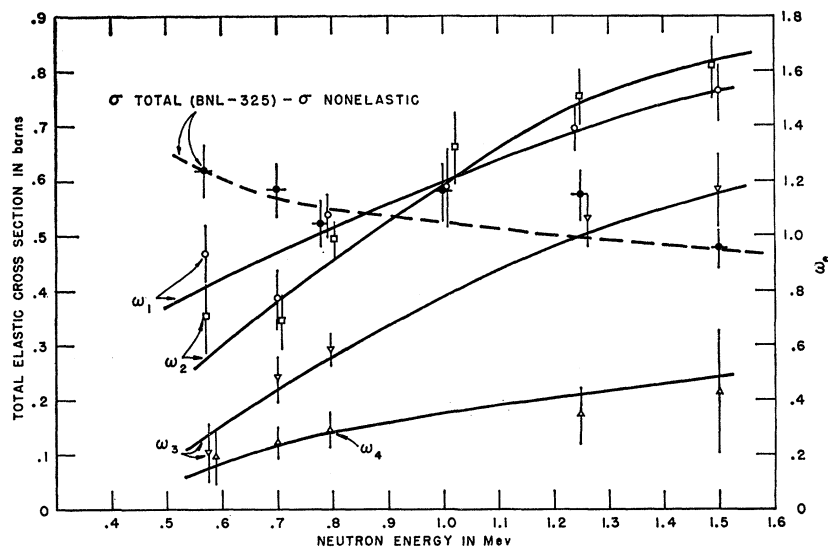


FIG. 7. The dependence of $\sigma(\text{total, el})$ and $W_i (i=1-4)$ on incident neutron energy. The thorium differential elastic cross section is given by

$$\frac{d\sigma(\text{el})}{d\Omega} = (4\pi)^{-1} \sigma(\text{total, el}) \times (1 + W_1 P_1 + W_2 P_2 + W_3 P_3 + W_4 P_4)$$

where P_i 's are the respective Legendre polynomials.

²³ I. S. Sokolnikoff and R. M. Pedheffer, *Mathematics of Physics and Modern Engineering* (McGraw-Hill Book Company, Inc., New York, 1958), p. 159.

²⁴ S. Yiftak et al., *Fast Reactor Cross Sections* (Pergamon Press, New York, 1960).

given in Fig. 8. The coefficient b in expression (4) is not measurably dependent on the incident neutron energy from 560 kev to 1.0 Mev.

The total inelastic scattering cross section of thorium is obtained directly from Fig. 8. The result is given in Fig. 10. Scattering to the triplet of states in the vicinity of 800 kev is experimentally difficult to study. The three components were separately resolved at only an incident energy of 1.4 Mev with the results shown in Fig. 4. Similarly, the doublet at approximately 1100 kev was experimentally resolved at 1.45 Mev only. As a result the triplet and doublet structures shown in Figs. 8 and 10 are represented as individual inelastic neutron groups resulting in the excitation of "average" nuclear levels at 800 and 1100 kev respectively.

DISCUSSION OF RESULTS

The level structure shown in Fig. 5 is characteristic of a heavy, even-even, deformed nucleus. Clearly evident from the measurements are the first two excited levels ($2+$, $4+$) of a rotational band based upon the ground state. The energy placement of these two levels is in good agreement with the theoretically predicted dependence on I given by

$$E = \frac{1}{2}AI(I+1) - BT^2(I+1)^2, \quad (5)$$

where I is the nuclear spin, B is a small ($\sim 10^{-3}$) vibrational distortion parameter, and A is proportional to the reciprocal of the moment of inertia.²⁵ This rotational sequence has been identified up to the $10+$ level in heavy ion Coulomb excitation experiments.² The higher levels of this rotational sequence are not measurably excited in this experiment due to the relatively small amount of incident angular momentum available. Alpha particle Coulomb excitation experiments have resulted in the identification of nuclear levels in

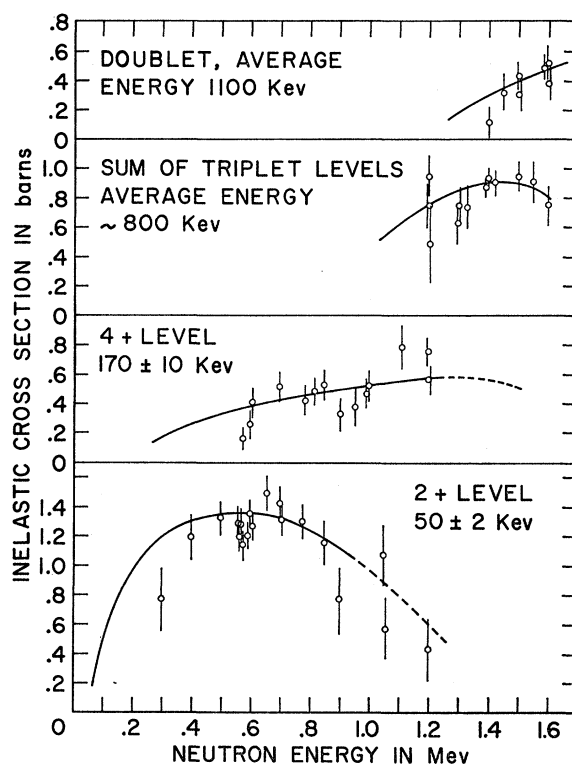


FIG. 8. Cross sections for the excitation of residual nuclear levels in thorium by inelastic neutron scattering as a function of incident neutron energy.

thorium at 760 and 790 kev.³ The measured branching ratios indicate that these states are, respectively, members of β and γ rotational bands and are characterized by $K=0$, $I=2+$, and $K=2$, $I=2+$ quantum parameters. Nuclear levels at 720 and 790 kev have been seen in studies of gamma-ray emission following inelastic scattering.^{6,26} It seems likely that the 720, 790, and 820

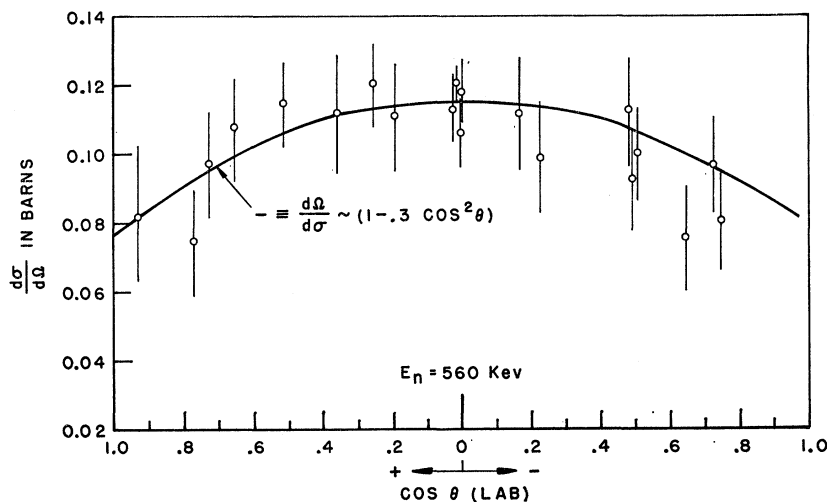


FIG. 9. The differential cross section for inelastic scattering from thorium resulting in the excitation of the first ($2+$) level at 50 kev.

²⁵ R. K. Sheline, Revs. Modern Phys. **32**, 1 (1960).

²⁶ L. Cranberg, Argonne National Laboratory Report ANL-6122, 1959 (unpublished).

TABLE I. A comparison of the total neutron cross section (in b) of thorium obtained in this experiment with the published values.^a

Neutron energy in Mev	A σ (total elastic) this exp.	B σ (capture) BNL-325	C σ (fission) BNL-325	D σ (total inelastic) this exp.	E σ (total) = A+B+C+D this exp.	F σ (total) BNL-325
0.570	6.18±0.80	0.160	...	1.40±0.15	7.740±0.82	7.70
0.700	5.92±0.70	0.155	...	1.50±0.15	7.575±0.80	7.40
0.790	5.17±0.70	0.145	...	1.54±0.15	6.860±0.80	7.20
1.000	5.87±0.80	0.125	...	1.70±0.16	7.697±0.83	7.00
1.250	5.80±0.90	0.100	0.015	1.90±0.20	7.817±0.92	6.65
1.500	4.77±0.70	0.090	0.070	2.00±0.20	6.930±0.74	6.60

^a See reference 27.

kev triplet observed in this experiment is due respectively to the 0, 0+, and 0, 2+ states of a β vibrational band and the first 2, 2+ level of a γ vibrational sequence. The 0, 2+ and 2, 2+ states correspond, within the respective experimental errors, with the states observed in the alpha particle Coulomb excitation studies. The 720 kev level would not be observed in the Coulomb excitation measurements due to its postulated 0+ configuration. Its existence is verified in studies of gamma rays emitted following inelastic neutron scattering.⁶ The 790 kev level reported in that work probably includes the contribution of the 820 kev level observed in this experiment. Such small differences as exist between the energies reported in the various experiments are easily within the experimental errors. The pair of levels seen in this experiment at 1050 and 1150 kev closely corresponds to similar structure observed in inelastic gamma-ray studies.⁶ The rapidly increasing

complexity of the thorium structure at higher energies combined with the decreasing resolution of the time-of-flight method makes the assignment of spins or parities to the 1050 and 1150 kev levels problematical.

The total elastic scattering cross sections given in Fig. 7 and the inelastic cross sections in Fig. 8 are compared with the published total cross sections of thorium in Table I.²⁷ The capture and fission cross sections in the energy range of interest are relatively small and have a correspondingly slight effect on the total cross sections. The total cross sections derived from this experiment are, within experimental error, in agreement with the results given in BNL-325.²⁷

Elastic scattering of 1.0 Mev neutrons from thorium has been studied using a biased counter system.¹⁰ Since it was difficult to essay the effect of inelastic scattering on that measurement, no comparison with the present work was made. Extensive studies of total scattering angular distributions from thorium have been reported.⁹ By making proper allowance for inelastic scattering, these total scattering angular distributions can be compared with the results of this experiment. When this is done, the W_1 , W_2 , and W_3 coefficients (see Fig. 7) resulting from the two experiments agree to within 10% although there appears to be a systematic trend toward slightly more forward peaking in this experiment. The W_4 term obtained in this work is consistently smaller than that derived from the total scattering distributions.

As described above and shown in Fig. 9, the angular distribution of neutrons scattered from the 2+ level was measured to be symmetric about 90° below an incident energy of 1.0 Mev. Scattering to the 4+ level was observed to be isotropic in all measured instances. Scattering to the other levels was examined at several angles with no measurable departure from isotropy. In all measured cases, the inelastic scattering was either isotropic or symmetric about 90°. This result is in accord with the Bohr theory of compound nucleus reactions.²⁸ In no instance do these measurements

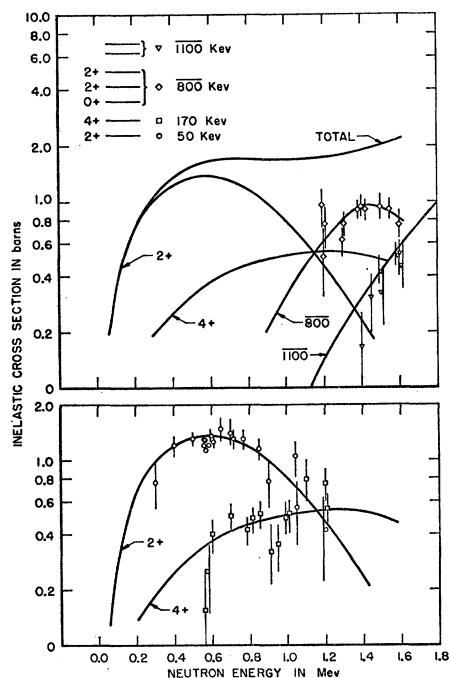


FIG. 10. The composite total inelastic cross section of thorium as a function of incident neutron energy.

²⁷ *Neutron Cross Sections*, compiled by D. J. Hughes and R. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd edition.

²⁸ J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952).

indicate any significant departure from symmetry about 90° that could be attributed to direct interactions.²⁹ The solid curves in Figs. 8 and 10, associated with the inelastic cross section for the excitation of the $2+$ and $4+$ residual nuclear levels, are in fair agreement with theoretical calculations of the Hauser-Feshbach type adjusted to fit the experimental data.^{12,30,31} Above an incident neutron energy of ~ 1.0 Mev the effect of nuclear levels in the vicinity of 800 kev becomes increasingly important. The spins and parities of these and higher levels are not certain, and it is not clear that nuclear reaction properties at these energies are fully analogous to those existing at lower excitations. However, estimates of the partial inelastic cross sections based upon the level assignments of Fig. 6 can be made.¹²

²⁹ D. Chase and L. Wilets, *Bull. Am. Phys. Soc.* **2**, 72 (1957).

³⁰ W. Hauser and H. Feshbach, *Phys. Rev.* **87**, 366 (1952).

³¹ P. Moldauer (to be published).

The results are in reasonable qualitative agreement with the empirical curves shown in Figs. 8 and 10.^{12,31}

This experiment has resulted in an increased understanding of scattering from thorium. The results are directly applicable to the design of fast reactive breeding systems. The measurements have given additional insight into the structure of the thorium nucleus. They also provide an experimental basis for theoretical understanding of fast neutron reactions in heavy nuclei.

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