# High-Resolution Neutron Transmission Measurements on $^{235}\mathrm{U}, ^{239}\mathrm{Pu},$ and $^{238}\mathrm{U}^*$

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Abstract: High-resolution transmission measurements have been made on three sample thicknesses of both <sup>235</sup>U and <sup>239</sup>Pu at liquid nitrogen temperature and also on three of <sup>238</sup>U at room temperature using neutrons from the water-moderated ORELA target. The data on <sup>235</sup>U and <sup>239</sup>Pu from 1 to 10,000 eV were obtained using <sup>6</sup>Li glass scintillation detectors at 17.909- and 80.394-m flight paths. The <sup>238</sup>U data from 1 to 100 keV were obtained using a new NE 110 proton-recoil scintillation detector at a 201.558-m flight path.

 $(^{235}\mathrm{U},\,^{239}\mathrm{Pu},\,^{238}\mathrm{U},\,\mathrm{high\text{-}resolution}$  transmission measurements)

#### Introduction

Accurate neutron transmission data for <sup>235</sup>U and <sup>239</sup>Pu are essential for determining accurate parameters of the neutron resonances. To improve the quality of the new ENDF/B evaluation of <sup>235</sup>U and <sup>239</sup>Pu (see paper of Derrien *et al.*<sup>1</sup> this conference) and to extend the resolved energy region to higher energies, new high-resolution, accurate data using cooled samples were desired. ORELA with its high neutron intensity and low backgrounds, is one of the best neutron sources to produce these data.

In 1982 a <sup>238</sup>U Task Force<sup>2</sup> was formed to deal with two neutron data problems for <sup>238</sup>U: (1) discrepancies in the neutron widths of the resolved resonances above 1.4 keV from several laboratories and (2) the capture cross section in the resolved and unresolved resonance region. This Task Force concluded that the prime reason for the discrepancy in neutron widths was that the resolution functions were wider and more complex than the experimenters had assumed, and that the representation of the unresolved region above ~4 keV neutron energy would best be solved by extending the resolved region to higher energies, say 10 to 20 keV. The Task Force recommended that "Higher resolution transmission measurements are desirable, using cooled samples on a  $\sim 200/300$  m flight path using a  $\sim 10$  ns burst width and a neutron detector which has an improved and wellknown resolution function." We have performed measurements at ORELA which meet these requirements except that for the present measurements the samples were at room temperature; however, cooled sample measurements are planned for next year.

## Experimental Details and Results for <sup>235</sup>U and <sup>239</sup>Pu

Two <sup>6</sup>Li glass scintillation detectors were used for these transmission measurements. For the low-energy data a 1-mm thick scintillator, 10 cm in diameter, mounted in a 0.025-mm thick, 15-cm diameter mylar reflecting cylinder between two RCA 8854 photomultiplier (PM) tubes was used at a 17.909-m flight path with a 1.5-mm cadmium filter and a pulse repetition rate of 347 pps of 20 ns pulses. For the higher energy data a 12.5-mm thick scintillator, 11.1 cm in diameter, mounted in a similar cylinder between two RCA 8854 PM tubes was

located at a 80.394-m flight path. A 0.15-gm/cm<sup>2</sup> <sup>10</sup>B filter and a repetition rate of 347 pps of 20-ns pulses was used for <sup>235</sup>U and 780 pps of 15-ns pulses for <sup>239</sup>Pu. A 2-cm thick Pb filter was used for the low-energy data and a 0.7-mm thick one for the higher energy data to reduce the background due to gamma rays from the target. The photomultiplier bases are gated to eliminate the gamma flash. In order to minimize background, a coincidence is made between fast-timing windows from each PM and a 40-ns smoothed sum signal from the two PMs.

The uranium samples (total weight of 24.3 gm) were enriched to 99.6% in  $^{235}\mathrm{U}$  and had sample thicknesses of uranium at room temperature of 0.03281, 0.002343 and 0.000575 atoms/barn. The plutonium samples (total weight of 71.9 gm) were enriched to 99.96% in  $^{239}\mathrm{Pu}$  and had sample thicknesses of plutonium at room temperature of 0.07375, 0.01803, and 0.00638 atoms/barn. All samples were cooled to liquid nitrogen temperature and were cycled with an open beam. These sample thicknesses are increased by factors of 1.004 for  $^{235}\mathrm{U}$  and 1.013 for  $^{239}\mathrm{Pu}$  in cooling to liquid nitrogen temperature

Both  $^{235}$ U and  $^{239}$ Pu data have been corrected for the deadtime (1104 ns) of the time digitizer and for several backgrounds which total  $\lesssim 3\%$  over most of the energy region studied. These backgrounds consist of a constant beam-independent background, a 17.6- $\mu$ s gammaray background from the capture of neutrons by the water moderator, overlap neutrons from the previous burst, and a time-dependent background arising from neutrons scattered from the <sup>6</sup>Li glass scintillator. Additional details on these backgrounds are given in published papers and reports. The for the  $^{235}$ U data, corrections were made up to 1500 eV for the contribution from a 0.385% tantalum impurity in two of the samples and for contributions from the  $^{234}$ U (0.033%),  $^{236}$ U (0.184%), and  $^{238}$ U (0.128%) in the three samples; these percentages were determined from the analyses of lowenergy resonances of these nuclides.

The energy resolution for these measurements is determined mainly by the moderation time in the target moderator which corresponds to a FWHM of  $\sim 25 \text{ mm}^5$  for the energy region below 1 keV. Hence, the energy resolution is less than Doppler broadening for neutron energies  $\lesssim 700 \text{ eV}$ . However, at the higher energies scattering in the thick <sup>6</sup>Li scintillator produces a 30% tail

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<sup>\*</sup> Research sponsored by the Office of Nuclear Physics, U.S. Department of Energy, under contract DE-AC05-84OR21400 with the Martin Marietta Energy Systems, Inc.

on the resolution function<sup>6</sup> which can be important in the analysis of resonances with low transmission. Analysis of these data are reported by Derrien  $et\ al.^1$  up to 110 eV for  $^{235}$ U and 1 keV for  $^{239}$ Pu. Examples of the data and the excellent fits for  $^{239}$ Pu are shown in the paper by Derrien  $et\ al.^1$ 

#### NE 110 Neutron Detector for <sup>238</sup>U Measurements

For the high-resolution transmission measurements  $^{238}\mathrm{U}$  we have developed a neutron detector  $^{7}$  with a narrow resolution function which is efficient for keV energy neutrons, has low background, and is quite insensitive to overlap neutrons (<200 eV) so the pulsed accelerator can be operated at a high repetition rate (800 pps). The detector consists of a 9- by 9-cm NE 110 scintillator, 1 cm thick, epoxy-coupled to two 12.5-cm diameter RCA 8854 photomultipliers. This bare (noncoated) scintillator is mounted in a 0.025-mm thick, 17.8-cm diameter mylar reflecting cylinder. Each PM is biased below the single photoelectron level, and a coincidence is required between the outputs of the two PMs to eliminate counts due to PM noise. The detector response has been studied down to a neutron energy of 50 eV. The observed signal-to-background ratio determined from blacking-out resonances using a 2.54-cm thick bismuth sample and 30-ns bursts was 300 at 12 keV and 30 at 2 keV. The detector has an efficiency similar to those of earlier detectors<sup>7</sup> developed at ORELA,  $\sim$ 40% at 15 keV (ten times that of a 1-cm-thick  $^6$ Li glass scintillation detector and equal to that of the <sup>6</sup>Li detector at 4 keV). The timing resolution of the detector is 7 ns for 10-keV neutrons, limited by the neutron flight time through the scintillator, and is less for higher energy neutrons. Each PM was gated off until 40  $\mu$ s after the gamma flash.

### Measurements and Results for <sup>238</sup>U

Transmission measurements from 1 to 100 keV have been made with this detector at an effective flight path<sup>9</sup> of 201.558 ± 0.004 m using neutrons from the water-moderated tantalum target at ORELA. The <sup>238</sup>U samples (99.92 and 99.99%) were the same ones used by Olsen et al., <sup>10</sup> but the surface oxide was removed before the present measurements were made. Measurements were made for three sample thicknesses, N=0.1748, 0.0396, and 0.01235 atoms/barn located in the 80-m flight station. The measurements on the thickest sample (36.2 mm) were made using 6-ns bursts at 800 pps, and on the 2.5- and 8.3-mm-thick samples using 15-ns bursts at 415 pps. The neutron energy resolution (0.03%) is less than the Doppler broadening below 15-keV neutron energy.

Eighty thousand 2-ns channels were used to cover the energy region from 5 to 100 keV, 30,000 4-ns channels from 2 to 5 keV, 10,000 10-ns channels from 1.2 to 2 keV, and 40-ns channels below 1.2 keV. Data were taken for four pulse-height windows to determine the backgrounds. A background due to 2.2-MeV gamma rays from the capture of neutrons in the water moderator is mainly in the highest window, and that arising from 478-keV gamma rays produced from neutrons scattered from the NE scintillator reacting with the boron in the pyrex of the phototubes is mostly in the second highest window.

Data for the open beam for 43 hours of operation at a repetition rate of 800 pps of 6-ns pulses are shown in Fig. 1. The upper curve is the open beam counts, the middle curve is the sum of all backgrounds, and the lower curve is the background due to  $^{10}$ B( $n, \alpha \gamma$ ) in the detector which is  $\sim 10\%$  of the total background for

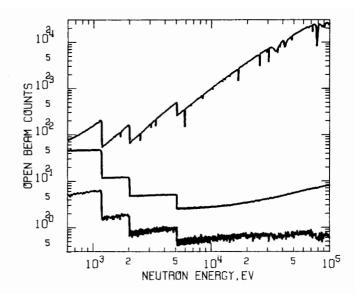


Figure 1. Open beam counts (upper curve) in 43 hours for 2-ns channels from 5 to 100 keV, 4-ns from 2 to 5 keV, 10 ns from 1.2 to 2 keV, and 40-ns below 1.2 keV. The middle curve is the sum of all backgrounds and the lower curve is the background due to  $^{10}$ B $(n, \alpha\gamma)$  in the detector.

these operating conditions. At low energies the background is mainly (90%) due to a constant time independent background. At high energies it is mostly due to 2.2-MeV gamma rays from the moderator ( $\sim$ 70% at 100 keV). For these operating conditions, the signal to background ratio for the open beam is greater than 1000 above 20 keV, 100 at 5 keV, 30 at 2.8 keV, and 10 at 1.8 keV. However, for the thickest <sup>238</sup>U sample with an average neutron transmission of only  $\sim$ 0.1, the signal to background is almost an order of magnitude poorer as shown in Fig. 2 (data averaged by 500), the background is mainly ( $\sim$ 97%) time independent which can be determined accurately ( $\sim$ 2%). A small energy region (only averaged by 2's) is shown in Fig. 3, where the signal to background is  $\sim$ 30 to 1 between resonances.

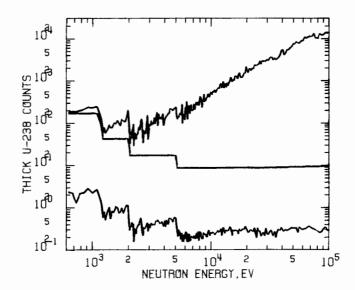


Figure 2. Counts (averaged by 500's, upper curve) with the thick  $^{238}\mathrm{U}$  sample (36.2 mm) in 179 hours for the same channel widths as Fig. 1. The middle curve is the sum of all backgrounds and the lower curve is the background due to  $^{10}\mathrm{B}(n,\alpha\gamma)$  in the detector.

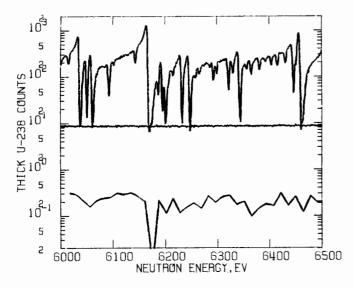


Figure 3. The observed counts with the thick  $^{238}$ U sample (upper curve, averaged by 2's), sum of all backgrounds (middle curve), and  $^{10}$ B $(n, \alpha\gamma)$  background (lower curve).

For low energies,  $\sim 2$  keV, the data are comparable to the earlier data of Olsen et al. <sup>10</sup> The energy resolution is somewhat better but the statistical uncertainties are somewhat greater as shown in Fig. 4 for the thick <sup>238</sup>U sample. At higher energies the statistical uncertainties are much less. The energy resolution is determined mainly by the moderation time in the target and is  $\sim 0.03\%$  at low energies and 0.05% at high energies. Several small energy regions for the thickest sample are shown in Fig. 5.

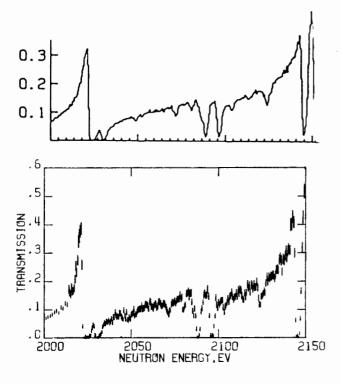


Figure 4. Comparison of thick  $^{238}$ U data from Olsen et al.  $^{10}$  (upper curve) and present data (lower curve) from 2.0 to 2.15 keV.

#### Conclusion

High-resolution transmission data on <sup>235</sup>U and <sup>239</sup>Pu have been produced for the next ENDF/B evaluation. Using a new NE 110 proton recoil scintillation detector, accurate, high-resolution transmission measurements have been made on <sup>238</sup>U which satisfy the recommendations of the <sup>238</sup>U Task Force, except that the samples were at room temperature and not cooled. It should be possible to extend the resolved resonance region for <sup>238</sup>U to 20 or 30 keV.

#### REFERENCES

- 1. H. Derrien et al., paper at this conference.
- M. Sowerby, "Summary of the Work of the NE-ANDC Task Force on U-238," Int. Conf. on Nuclear Data for Basic and Applied Science, May 13-17, 1985, Santa Fe, New Mexico.
- M.S. Pandey, J. B. Gary, and J. A. Harvey *Phys. Rev.* C15, 600 (1977); G. D. James *et al.*, *Phys. Rev.* C15, 2083 (1977).
- 4. D. C. Larson et al., ORNL/TM-5612, October 1976.
- C. Coceva, R. Simonini, and D. K. Olsen, Nucl. Instr. Meth. 211, 459 (1983); D. C. Larson, N. M. Larson, and J. A. Harvey, ORNL/TM-8880, June 1984; and F. G. Perey, private communication (1988).
- J. A. Harvey and N. W. Hill, Nucl. Instr. Meth. 162, 507 (1979).
- J. A. Harvey and N. W. Hill, Bull. Am. Phys. Soc. 33, 1078 1988).
- N. W. Hill et al., IEEE Transactions on Nuclear Science, Vol. NS-32, No. 1, February 1985.
- 9. D. C. Larson et al., ORNL/TM-9097, March 1985.
- D. K. Olsen et al., Nucl. Sci. Eng. 69, 202 (1979); also ORNL/TM-5915, October 1977.

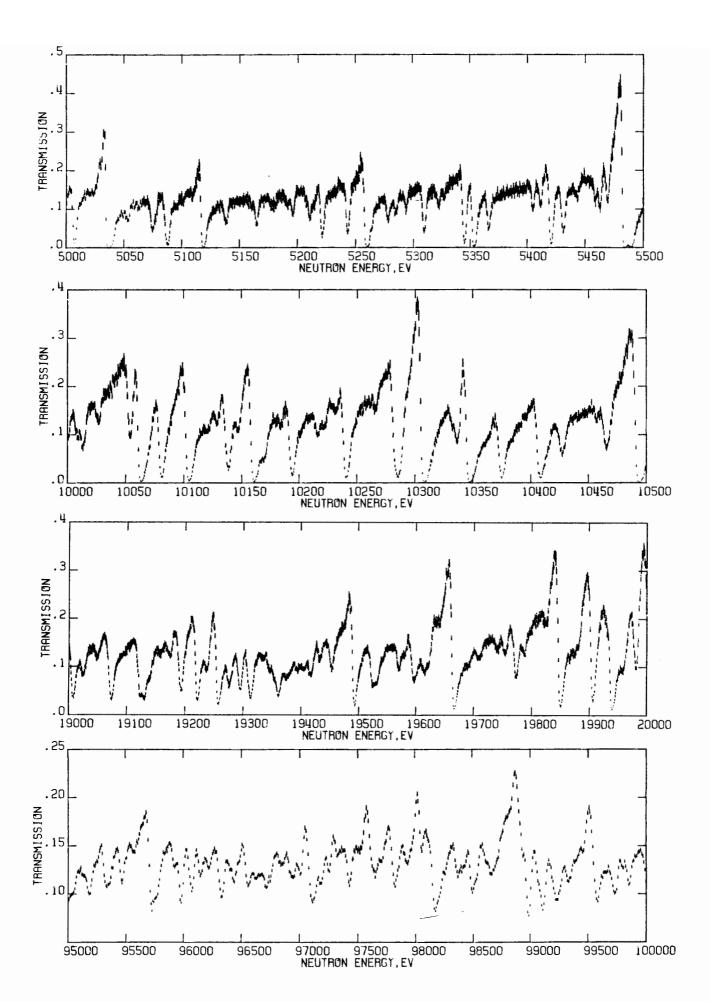


Figure 5. Transmission with the statistical uncertainties for the thick  $^{238}$ U sample for several energy regions.