Thermal-neutron fission cross section of 26.1-min $^{235}U^m$

W. L. Talbert, Jr., J. W. Starner, R. J. Estep, S. J. Balestrini, M. Attrep, Jr.,
D. W. Efurd, and F. R. Roensch
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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The thermal-neutron fission cross section of $^{235}U^m$ has been measured relative to the ground-state cross section. A rapid radiochemical separation procedure was developed to provide sizeable (10^{10} to 10^{11} atom) samples that were reasonably free of the parent ^{239}Pu. From a series of eight measurements, the value of 1.42 ± 0.04 was obtained for the ratio σ_m/σ_g.

I. INTRODUCTION

The 26.1-min isomeric state of ^{235}U has been studied extensively since its discovery and isolation in 1957.[1,2] Despite efforts to obtain samples large enough to make a measurement of its fission cross section, no such measurements were reported until 1984,[3] while the present work was in initial stages. While this work was in progress, we learned of another effort to measure the cross section at the Institut-Laue-Langevin (ILL).[4] Prior to this time, most of the experimental work on the properties of $^{235}U^m$ was devoted to a determination of the effects of environment on its decay constant[5–7] and energy.[8] This $\frac{1}{2}^+$ state decays to the $\frac{7}{2}^-$ ground state with a half-life of 26.1 min. The $E3$ transition proceeds almost entirely through the emission of very low-energy conversion electrons. The transition energy has been determined to be 76.8 ± 0.5 eV. We were attracted to a measurement of the fission cross section of this state by the realization that it is relatively easily populated in moderately "hot" environments.[9] To allow a determination of the average fission width and the level densities of the excited target nucleus, the measurement should extend to neutron energies of at least 10 keV; however, we chose to make a first attempt with thermal-energy neutrons, which are accessible in high intensity at the Los Alamos Omega West Reactor and for which the cross section could be expected to be high. We could then use this experience to develop the techniques required for the more stringent conditions imposed at higher neutron energies.

In past studies of $^{235}U^m$, the collection of samples of the isomeric state has relied on the fact that in the α decay of ^{239}Pu, more than 99.8% of the decays directly or indirectly populate the isomeric state of ^{235}U. Hence, collection of the 90-keV α-decay recoils has been the usual method of obtaining a sample of $^{235}U^m$. Samples of adequate size for studies of half-life and isomeric level energy can be obtained with reasonable areas of plutonium surface.[10] However, in order to obtain samples adequate for fission cross-section measurements, rather large surface areas are required; for example, the area used in the experiments of Ref. 3 was reported to be 1 m^2, and a 0.5-m^2 surface has been prepared for sample collection in experiments at ILL in Grenoble.[11]

Our first attempts to isolate samples of $^{235}U^m$ from the α decay of ^{239}Pu were based on capture of the recoiling $^{235}U^m$ nuclei from a 0.12-m^2 surface of plutonium by volatile aerosols suspended in helium flowing past the surface. The stream of helium was then entrained in a small-diameter tube and directed to a collection disk of platinum. By directing several helium streams to the same spot on the platinum disk, we could then concentrate the recoils collected from a large surface area of plutonium distributed over several plates. The resulting sample was then introduced into a small fission chamber for the cross-section measurement. To date we have been unsuccessful in the use of this so-called "He-jet" technique.[12] The samples produced had only a small fraction of the expected $^{235}U^m$, although we could easily detect and multiscale (i.e., sequentially count to determine the time dependence of the decay) the conversion electrons given off in the isomeric decay, and contained unexpectedly large amounts of ^{239}Pu (determined by α counting). Inspection of the plutonium coatings on the plates used as recoil sources revealed that the plutonium was badly corroded and had not adhered well to the plates. We will continue to explore using this technique because it has the advantage that it is easily scaled up to provide the larger samples of $^{235}U^m$ required for energy-dependent cross-section measurements.

II. EXPERIMENTAL APPROACH

There were two developmental efforts required to pursue the fission cross-section measurement for $^{235}U^m$. These were to develop a low-background fission chamber that could tolerate the severe conditions in the reactor thermal column (large γ-ray and thermal-neutron fluxes), and to develop a procedure, possibly radiochemical, for the separation of adequate samples of $^{235}U^m$ (10^{10}–10^{11} atoms) from a plutonium source. As mentioned above, our attempts to use the He-jet technique for source preparation have not been successful, so we turned to a radiochemical approach.

A. Rapid radiochemical separation of $^{235}U^m$ from ^{239}Pu

We have developed a rapid radiochemical procedure for the separation of $^{235}U^m$ from milligram quantities of

^{239}Pu. The separation procedure provides a suitable $^{235}U^{m}$ sample in a time of 25–35 min that is basically plutonium free. The radiochemical separation of uranium from plutonium uses a simple selective anion exchange step: uranium is absorbed onto AG-MP1 resin from a 47% hydrobromic acid (HBr) solution but plutonium is not.[13] The uranium is then easily eluted from the column with a very dilute hydrofluoric acid (HF) solution.[14] A detailed description of the procedure will be published elsewhere.

1. Reagents and equipment

All equipment used in these procedures was new and not used for any other procedure. The containers were precleaned with 4*M* nitric acid $(HNO)_3$, distilled water and finally with Milli-Q water. Reagents included Merck Suprapure 47% HBr, and 0.006 M HF prepared from ultrapure Seastar HF. For each sample preparation three polyethylene dropping bottles of each reagent were prepared. These were changed at various stages of the procedure to minimize the cross contamination from one stage to another. The columns were prepared in blue Eppendorf plastic pipette tips which were plugged with prewashed quartz fiber and filled with pretreated AG-MP1 50–100 mesh resin. We prepared 2-cm columns to be used for the initial cleanup steps and 1-cm columns which had faster flow rates to be used for the actual separation. All columns were preequilibrated with HBr immediately prior to use.

2. Radiochemical separation

The plutonium (70–250 mg) was taken up in a minimum amount of HBr and stored in a Teflon bottle until used for a separation. The initial steps were performed in a portable plastic glove box positioned in a fume hood. We removed the uranium that had grown in from the plutonium solution since the previous separation by passing the plutonium solution through a five-column (2-cm length) clean-up series. The plutonium solution was passed through a column, washed with a few drops of HBr and collected in a Teflon beaker. The column was discarded in each case. During the use of the fifth column, time was marked as the beginning of the ingrowth period.

After an approximate 15-min ingrowth period, the plutonium solution containing the freshly grown-in uranium was passed though a 1-cm column marking the end of ingrowth. The uranium was absorbed on the column. The column was then washed three times by filling it with HBr and forcing the HBr through the column with a syringe fitted with a rubber stopper. The uranium was removed from the column with 1–2 ml HF solution and was collected in a Teflon beaker. Four to five ml HBr were added to the collected solution containing the uranium, mixed and passed through another column. The absorbed uranium was again washed three times with HBr and the column was removed from the glove box. Following the removal of the uranium from this column, the 4–5 ml of HBr was added to the solution which, after mixing, was passed through a final column, washed three times with HBr, and eluted with HF into a warm Teflon beaker. The final sample was carefully evaporated to a final volume of 0.1–0.2 ml on a hot plate with the aid of an air jet gently passed over the surface. The final drop was transferred to a cold 9.5-mm platinum disk placed on a Teflon-covered hot plate under a heat lamp, both of which were turned on after delivery. Following complete evaporation aided by air jet, the disk was flamed and delivered for irradiation and counting.

We conducted several separation blanks to ensure that reagents, equipment, or environment of the procedure did not introduce fissionable materials into the final sample. We first performed these blanks in clean room conditions and then repeated them at the location where the experiment separations were to take place. All blanks were satisfactory, containing no appreciable fissionable materials, with the ^{235}U level in the blanks determined to be lower than 10^{7} atoms.

We were able to attain a separation factor of uranium from plutonium of approximately 10^{12}. The number of atoms delivered for the cross-section measurements ranged between 6×10^{10} and 3×10^{11} atoms of $^{235}U^{m}$.

B. Fission chamber development and data analysis

We constructed a fission chamber using titanium (because of its low thermal-neutron activation and low uranium impurity level) and polyethylene insulators. The chamber, a cylinder 19 mm in diameter and 9.5 mm long, has plate-like electrodes at both ends. The $^{235}U^{m}$ sample, deposited on a 9.5-mm diameter platinum disk, forms part of one electrode and is accessible by a screw-top flange at one end of the chamber. A flow of P-10 proportional counter gas (about 0.5 cm^3/s) was provided to the chamber at a gauge pressure of about 200 kPa.

A potential of 600 V was applied between the two electrodes, and the signals picked up at each electrode by high-speed preamplifiers were analyzed for coincidence to gate a multichannel analyzer for spectrum analysis. The gated linear signal was then multiscaled with a threshold level of approximately 40 MeV. Multiscaling was typically carried out for 180 min in 1-min intervals. The background rate of the fission chamber in a thermal neutron flux of $\sim5\times10^{11}$ n/cm^2 s was about 100 counts/min, less than 10% of the typical $^{235}U^{m}$ sample counting rate.

The $^{235}U^{m}$ sample provided from the rapid radiochemical separation was loaded into the fission chamber and inserted into the thermal column of the Omega West Reactor as quickly as possible (usually within 2 min). The fission chamber signals were multiscaled along with a ^{235}U-flux-monitor signal. The flux monitor was positioned near the fission chamber at the same elevation in the thermal column, but far enough away that the neutrons produced in the flux monitor would not perturb the thermal spectrum at the fission chamber location. The activation cadmium ratios in the fission chamber at the thermal column position are 117 for ^{197}Au, 100 for ^{115}In, and 163 for ^{175}Lu. The neutron spectrum is approximated by a Maxwellian distribution of temperature

350 K (75.5 meV is the energy of the most probable flux in the wavelength representation of the Maxwellian flux).[15]

The data obtained in multiscaling the fission chamber signals and the flux monitor were analyzed as follows. The fission chamber multiscale spectrum was divided, channel by channel, by the flux monitor multiscale spectrum. The resulting spectrum, typically 180 channels in length, was then fitted to the function $\mathbf{a}+\mathbf{b}e^{-\lambda t}$, where λ is the isomeric state decay constant (0.026 56 min^{-1}) and t is the time elapsed since the end of the grow-in period during chemical separation. The value for **a** is the normalized fission rate for $^{235}U^{g}$ in the deposit (or, equivalently, the fission rate at long times), while **b** is related to the ratio of the isomeric-to-ground-state cross sections, $R=\sigma_m/\sigma_g$, by the following relation:

$$R-1=\mathbf{b}/\{[(1-e^{-\lambda})/\lambda][(1-e^{-\lambda t_0})/\lambda t_0] \times(\mathbf{a}-B-\text{"Pu"})\} ,$$

where **a** and **b** are the coefficients of the above function, the multiscale interval is 1 min, t_0 is the grow-in time, B is the background rate for the platinum disk used (there were significant, but small, variations noted from disk to disk), and "Pu" is a correction term for the fissions due to any plutonium found in the deposit by later α counting the disk (to a sensitivity of less than 10^8 atoms of plutonium).

III. EXPERIMENTAL RESULTS

Over the period of a year, many experimental measurements were made. Those conducted during the first and last portions of the year period appeared not to be subject to difficulties with the chemistry (an involved procedure, and especially taxing to the person performing the first few steps in a glove box) which, for example, would result in an incomplete purging of the uranium grown-in since the last experiment, difficulties in depositing the sample on a platinum disk, or a carryover of too much plutonium. A total of eight samples combined the qualities of "good" sample size (consistent with that expected for the plutonium sample size and grow-in period), adequate fission counting rate, adequate multiscale length (some experiments were cut short by power failures), and low plutonium content.

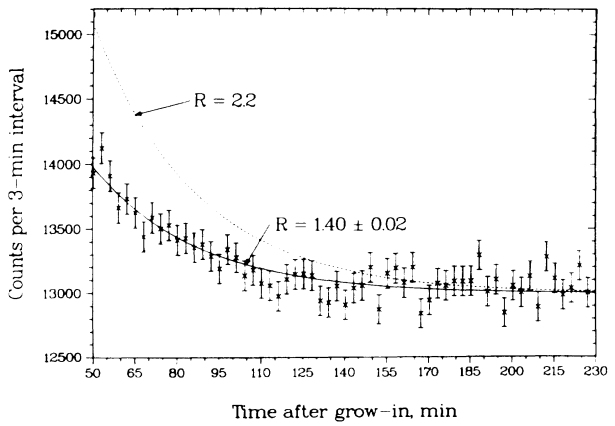

FIG. 1. Multiscale spectrum of fissions in a $^{235}U^{m}$ sample. The data have been condensed to 3-min intervals for convenience in plotting. The expected fit for the $R=\sigma_m/\sigma_g$ value reported in Ref. 3 is shown as the dashed line.

TABLE I. Summary of individual results.

Date of run	Atoms of ^{235}U	Atoms of ^{239}Pu	$(R-1)$
July 1, 1986	6.39×10^{10}	5.56×10^{8}	0.419±0.064
July 2, 1986	3.68×10^{11}	4.32×10^{9}	0.403±0.022
May 19, 1987	1.78×10^{11}		0.437±0.022
May 20, 1987	1.02×10^{11}	1.02×10^{9}	0.374±0.037
May 22, 1987	6.96×10^{10}		0.466±0.041
June 2, 1987	1.76×10^{11}		0.478±0.022
June 3, 1987	8.05×10^{10}	6.21×10^{8}	0.392±0.032
June 4, 1987	1.35×10^{11}	6.78×10^{8}	0.386±0.022

A typical multiscale spectrum is shown in Fig. 1, showing also the fit to be expected for the value $R=2.2\pm0.4$ reported in Ref. 3. The value for R shown as a fit to the data in Fig. 1 is an individual value, and the uncertainty is statistical. It is clear from the figure that we do not measure a time-dependent fission rate consistent with the result obtained in Ref. 3. Although Mostovoi and Ustroiev[3] report their value on the basis of 15 independent measurements (not reported individually), their large uncertainty must reflect considerable scatter in the individual measurements.

The data for the eight samples used in arriving at a final value for R are displayed in Table I. The weighted average value for R is $R=1.42\pm0.44$, where the uncertainty is one standard deviation (the values for $R-1$ were used in the averaging).

We very recently learned of preliminary results obtained for experiments performed at ILL.[4] The Mol/Ghent/ILL/LANL collaboration has reported $R=1.61\pm0.44$ for 5-meV (most probable energy) neutrons and 2.47 ± 0.45 for 70-meV (most probable energy) neutrons. These values show a trend for increasing isomer cross section with increasing neutron energy but are not in agreement with our value of 1.42 ± 0.04 for 75.5-meV (most probable energy) neutrons. These "most probable energy" values correspond to the energy of the most probable flux when plotted as a function of neutron wavelength, a common representation used with neutron beams available for diffraction studies.

IV. DISCUSSION AND FUTURE PLANS

The isomeric state of ^{235}U has a nuclear structure character similar to that of the ground state of ^{239}Pu. The measurement of the fission cross section induced by thermal-energy neutrons does not provide enough information to deduce any basic fission process parameters. However, the fact that the ratio of the isomeric and ground-state cross sections at thermal energies is roughly the same as the ratio of the ^{239}Pu ground-state to ^{235}U

ground-state cross sections allows one, as stated in Ref. 3, to postulate that thermal fission of $^{235}U^{m}$ likely proceeds by a level with spin zero in the compound nucleus (by analogy with ^{239}Pu). The trend of the cross section as a function of neutron energy from Ref. 4 (the neutron spectrum for Ref. 3 was not specified), may indicate the possible presence of a strong low-energy resonance in the $^{235}U^{m}(n,f)$ cross section, suggesting additional similarity to that for ^{239}Pu.

It is important to extend this measurement to higher neutron energies to test the possible cross-section indications stated above. The Los Alamos Neutron Scattering Center (LANSCE) provides an intense "white" neutron source for time-of-flight correlated measurements of neutron-induced processes at neutron energies up to many MeV. Preliminary estimates of sample sizes needed for an experiment at LANSCE lead to a plutonium "cow" size of 200–400 g to enable measurements of the fission cross section at neutron energies up to 100 keV. Chemical separation techniques may still be possible with such a size, but if the He-jet approach can be successfully developed, the scaling to plutonium source sizes of hundreds of grams would be direct. Toward this end, we are continuing our development efforts to employ the He-jet as a combined separation and concentrating technique for large source surface areas. The He-jet technique has been used successfully to transport recoiling activities with moderately high efficiency at a number of laboratories, including our own,[16] and should be capable of handling source areas of several m^2. We are also exploring the possibility of performing automated chemical separation on large plutonium samples.

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