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$^{242}Am^m$ fission cross section

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The neutron-induced fission cross section of $^{242}Am^m$ has been measured over the energy region from 10^{-3} eV to ~20 MeV in a series of experiments utilizing a linac-produced "white" neutron source and a monoenergetic source of 14.1 MeV neutrons. The cross section was measured relative to that of ^{235}U in the thermal (0.001 to ~3 eV) and high energy (1 keV to ~20 MeV) regions and normalized to the ENDF/B-V $^{235}U(n,f)$ evaluated cross section. In the resonance energy region (0.5 eV to 10 keV) the neutron flux was measured using thin lithium glass scintillators and the relative cross section thus obtained was normalized to the thermal energy measurement. This procedure allowed a consistency check between the thermal and high energy data. The cross section data have a statistical accuracy of ~0.5% at thermal energies and in the 1-MeV energy region, and a systematic uncertainty of ~5%. We confirmed that $^{242}Am^m$ has the largest thermal fission cross section known with a 2200 m/sec value of 6328 b. Results of a Breit-Wigner sum-of-single-levels analysis of 48 fission resonances up to 20 eV are presented and the connection of these resonance properties to the large thermal cross section is discussed. Our measurements are compared with previously reported results.

I. INTRODUCTION

The neutron-induced fission cross section of $^{242}Am^m$ is important from several aspects. $^{242}Am^m$ is the only odd-odd target nucleus whose half-life (152 yr) and relative availability make fission cross section measurements possible at high energies as well as at thermal energy. It has the largest measured thermal fission cross section of any nucleus and is the only one in which fission measurements can be performed on a target which is in an excited state. Analysis of fission resonances in the $^{242}Am^m(n,f)$ reaction in the eV energy region yields valuable information on level spacing. In this case, we should expect an increased level density compared to the more commonly studied even-odd targets.

Accurate high energy fission cross section measurements on transplutonic nuclei such as $^{242}Am^m$ yield data which can be compared with theoretical calculations[1,2] to arrive at systematic trends in fission barrier heights over a relatively wide range of actinide masses. $^{242}Am^m$ is also a secondary product in fission reactors and its cross section is useful in calculation of heavy actinide concentrations in reactor fuel elements. The cross section is also important for calculations involving production of heavy isotopes[3] and in actinide waste recycling schemes.[4]

Previous measurements of the fission cross section of $^{242}Am^m$ over limited energy regions and using samples with less isotopic purity have been made utilizing a variety of experimental techniques, including the use of a nuclear explosion as a neutron source. Until 1977, the only high energy fission cross section data on $^{242}Am^m$ were those of Seeger *et al.*[5] which spanned a neutron energy range from 20 eV to 1 MeV and those of Bowman *et al.*[6] which spanned the energy region from 0.02 eV to 6 MeV. Both of these measurements used samples whose $^{242}Am^m$ (isotopic) purity was about 20%, and corrections for the impurities, consisting mainly of ^{241}Am, were significant above 500 keV neutron energy. The more recent measurements of Fomushkin *et al.*[7] used a sample of $^{242}Am^m$ of ~86% isotopic purity and spanned an energy range of ~40 keV to 4.5 MeV and included a measurement at 14.8 MeV. Most recently, Dabbs *et al.*[8] published results using a high purity sample of $^{242}Am^m$ and which spanned an energy range from 0.005 eV to 20 MeV. Our series of measurements used isotopically pure (>99%) fission samples totaling ~1 mg and spanned a neutron energy range of 10^{-3} eV to ~20 MeV.

In Table I we outline our $^{242}Am^m(n,f)$ measurements which consisted of two series of electron linac measurements (1977 and 1979) taken at the Lawrence Livermore National Laboratory (LLNL) and a monoenergetic 14.1 MeV measurement (1980) taken at the LLNL Insulated Core Transformer (ICT) accelerator. Preliminary results of the 1977 linac measurement have been reported elsewhere.[9] In Sec. II below we discuss the experimental procedures and results of the 1979 linac measurements in the thermal, resonance, and high energy regions and compare these results with our 1977 measurements. A Breit-Wigner sum-of-single-levels analysis of the low energy fission resonances is presented and the connection of these resonances to the large thermal cross section is discussed. In Sec. III we review the experimental procedures and results of our 14.1 MeV $^{242}Am^m(n,f)$ measurement. Finally, in Sec. IV, we make comparisons of our fission cross section data with previously published results.

TABLE I. Summary of experimental measurements on the neutron-induced fission cross section of $^{242}Am^m$ at Livermore facilities since 1977.

Year	Facility	Energy range[a]	Neutron-production target	Neutron flight-path (m)	Sample[b] mass (μg)	Technique[c]
1977[d]	LLNL-linac[e]	0.01–100 eV	H_2O moderated Ta[f]	9.8	788.8±12.1	1
		0.5 eV–1 MeV	Ta[g]	7.4	788.8±12.1	1,2
		1 keV–20 MeV	Ta	13.5	788.8±12.1	1
1979	LLNL-linac	10^{-3}–3 eV	H_2O moderated Ta	3.82	207.3±2.1	1
		0.5 eV–10 keV	H_2O moderated Ta	9.86	207.3±2.1	2
		1 keV–20 MeV	Ta	13.42	207.3±2.1	1
1980	LLNL-ICT[h]	14.1 MeV	$^3H(d,n)^4He$	3.0	207.3±2.1	1

[a]The 1977 and 1979 linac measurements each were divided into three parts corresponding to the energy range measured in each part.
[b]Isotopic purity was >99% for all samples.
[c]Technique refers to the method used to reduce the data to absolute cross section: (1) $^{242}Am^m(n,f)$ measured relative to $^{235}U(n,f)$, (2) Li-glass detectors used to measure neutron flux; the relative cross section obtained from this flux shape and the TOF fission spectrum was then normalized to the thermal energy cross section.
[d]Preliminary results of this measurement are given in Ref. 9.
[e]Lawrence Livermore National Laboratory 100-MeV electron linac.
[f]This target consisted of water-cooled tantalum plates coupled to an H_2O moderator assembly—for thermal and low energy neutron production, see Ref. 10.
[g]Target consisted of water-cooled tantalum plates only—for high energy neutron production.
[h]Livermore Insulated Core Transformer accelerator.

II. LINAC MEASUREMENTS

A. Experimental procedure

Fission data were taken using the LLNL linac to produce 100-MeV electrons which were focused on either of two neutron-producing targets. Each of these targets was composed of a stack of water-cooled tantalum plates and one of them contained an integral H_2O moderator assembly of a type described by Camarda.[10] The target with the moderating assembly yielded an enhanced production of neutrons in the eV and keV energy regions while the other target produced a fast spectrum of neutrons with a mean energy of ~1.2 MeV. As shown in Fig. 1 these targets were arranged such that the neutron drift pipe, containing the $^{242}Am^m$ fission chamber at either of two precisely determined flight-path lengths, could view either of the two neutron-producing targets. The linac was run in a pulsed mode which permitted the accurate determination of incident neutron energies by the time-of-flight (TOF) technique. Neutrons from either target were collimated with a series of polyethylene, borated polyethylene, and lead collimators.

The fission sample was prepared by electroplating $^{242}Am^m$ onto a 0.05 mm thick hemispherically shaped nickel substrate. The hemispherical geometry was similar to that discussed in Ref. 8. This geometry and the ionization gas pressure used in the fission chamber were optimized to enhance the difference between the energy loss of alphas and fission fragments thereby minimizing the possibility of alpha pileup being mistaken for a fission fragment. The ionization gas consisted of an Ar-CO_2 mixture (96%–4%) at a pressure of ~100 kPa. The hemisphere

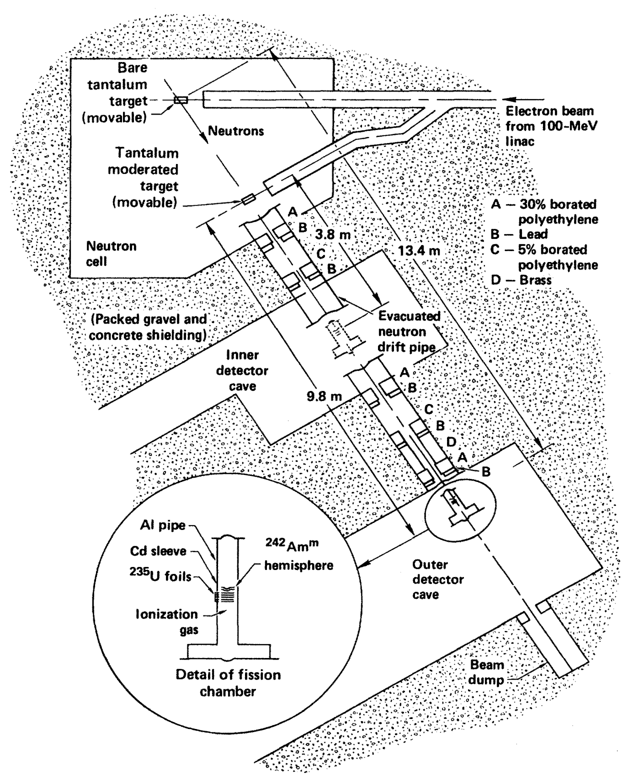

FIG. 1. The experimental geometry for the LLNL-linac measurements of the $^{242}Am^m(n,f)$ cross section. The figure shows the positions of the fission chamber in both the inner and outer detector caves as well as the low energy and high energy neutron-producing targets.

was biased at +400 V and was spaced 2.0 mm from its ground plate.

The $^{242}Am^m$ sample initially contained 99.21 wt. % $^{242}Am^m$, 0.79% ^{241}Am, and less than 0.007% ^{243}Am as determined by mass spectrometry. Separation from ^{242}Cm was accomplished by precipitation of $K_3AmO_2(CO_2)_2$ using lanthanum as a holdback carrier for the curium. After further chemical purification the americium was electroplated onto the nickel hemisphere from a solution consisting of 97% isopropyl alcohol and 3% 0.4 molar HCl and then fired as AmO_2 in a vacuum using radio frequency heating. The hemispherical area plated was defined by a rubber gasket 14.5 mm in diameter. The uniformity of the $^{242}Am^m$ plating on the hemispherical foil was checked by low geometry alpha counting using a solid state detector that was movable along a spherical surface. The uniformity was found to be within ±10%.

The masses of both the 1977 and 1979 $^{242}Am^m$ samples were determined in post-experiment destructive analyses by dissolving the hemispherical foils to recover all of the americium originally plated. The resultant samples were assayed by detection of the emitted alpha particles to determine the number of ^{241}Am and ^{243}Am atoms from which the number of $^{242}Am^m$ atoms could be determined using the $^{241}Am/^{242}Am^m$ and $^{243}Am/^{242}Am^m$ atom ratios which were measured by mass spectrometry. This technique was necessary due to the uncertainty in both the $^{242}Am^m$ half-life and the alpha branching ratio. The total mass of the 1977 $^{242}Am^m$ sample was determined to be 788.8 μg±2% and the 1979 sample 207.3 μg±1%.

It is important that ^{242}Cm be separated at the start of the experiment because $^{242}Am^m$ primarily decays by isomeric transition to its ground state which then can β decay ($T_{1/2}=16$ h) to ^{242}Cm. The ^{242}Cm can α decay ($T_{1/2}=163$ d) to ^{238}Pu, but it also has a spontaneous fission half-life of 6×10^6 yr. Therefore, the spontaneous fission rate of the $^{242}Am^m$ sample increases with time. Since the duration of each separate measurement was, in the longest run, only several weeks, we simply measured the spontaneous fission rate throughout the course of each measurement. However, the 18-month duration of the 1979–80 series of measurements described in this paper meant that over that time interval the spontaneous fission rate of the $^{242}Am^m$ sample grew from 0.05/sec to 3.2/sec.

Also included in the fission ionization chamber were planar foils containing ^{235}U. The ^{235}U was vapor deposited to a density of $\sim$500 μg/cm^2 on five 6-μm-thick titanium foils. The total mass of the ^{235}U foils was determined with reference to standard ^{235}U foils at the National Bureau of Standards (NBS). Our ^{235}U foils were positioned in a fission chamber between two standard ^{235}U foils each of whose masses was known to ±1%. The fission chamber was placed in a thermal neutron beam at the NBS reactor and the fission rates of all the foils compared. The neutron beam was checked for uniformity across the area of the ^{235}U deposits and for gradients along the length of the fission chamber. From this measurement, the mass of ^{235}U was determined to be 8485 μg with an uncertainty of ±2%. The larger mass of ^{235}U, compared to $^{242}Am^m$, was used so that the limiting statistics in our ratio measurements would not come from our standard reference. The greater uncertainty in the ^{235}U mass (±2% compared to ±1% for the mass of $^{242}Am^m$ used in the 1979 measurement) derives mainly from the fact that no post-experiment destructive analysis was done on our calibrated ^{235}U foils.

The efficiencies of the $^{242}Am^m$ hemispherical and ^{235}U parallel-plate ionization chambers were determined from fission pulse-height spectra acquired during the experiment and found to be 98.2±1.0% and 95.9±1.0%, respectively. Corrections for these efficiencies were included in the final cross section results. Signals from the fission ionization chambers were amplified in separate fast, current-sensitive preamplifiers before being transmitted to a counting area containing the signal processing electronics, time digitizers, and computers.

B. Experimental results

The experiment was divided into three parts categorized below by the energy range covered in each part. The energy ranges were the following: (1) thermal energy (10^{-3} eV to $\sim$3 eV), (2) resonance energy (0.5 eV to $\sim$10 keV), and (3) high energy (1 keV to $\sim$20 MeV). The experimental details, data reduction techniques, and results specific to each energy region are described in the following sections. In the various figures which follow we show only portions of the final data set which are pertinent to our resonance analysis or to comparisons with previously published results. The complete cross section data set, from 10^{-3} eV to $\sim$20 MeV is available from the National Nuclear Data Center.[11]

1. Thermal energy region

For the thermal energy region the water-moderated target was used and the fission chamber was placed in the inner detector cave (Fig. 1) at a neutron flight path of 3.82 m for a beam-on time of 20 h. The linac was pulsed at 30 pulses per second with a pulse width of 3 μsec. A 0.64 cm thick Pb filter was placed in the neutron beam to help suppress the gamma flash. The backgrounds, both constant and beam dependent, were determined in a separate 20-h run by placing transmission filters consisting of 1.0 mm of Cd and 0.23 mm of Rh in the neutron beam. With the Cd in the beam and at extremely low energies ($\sim10^{-4}$–10^{-2} eV), the constant number of counts/μsec in the $^{242}Am^m$ TOF fission spectrum yielded a background of 0.078 spontaneous fissions/sec which compared well with our beamoff averaged value of 0.074 spontaneous fissions/sec. This constant background corresponded to a 3% correction to the data at our lowest energy (0.001 eV) decreasing to a 0.03% or less correction at all energies above 0.01 eV. At approximately 0.07 eV, a background of $\sim$1 count/μsec, compared to $\sim$720 counts/μsec for the measurement without the Cd in the beam, implied a 0.14% beam-dependent background at this energy. At the Rh resonance (1.257 eV) the background was 0.3% compared to the measurement without Rh in the beam. The corresponding beam-dependent backgrounds for the ^{235}U chamber were 0.2% at 0.07 eV and 0.7% at 1.257 eV. Based upon these results no corrections for backgrounds were made to the low energy data except for subtracting the (constant) spontaneous fission background.

The data in the low energy region were measured relative to ^{235}U and reduced to an absolute fission cross section as a function of incident neutron energy $\sigma_f^{242}(E)$ by the following relation:

$$\sigma_f^{242}(E)=\sigma_{\rm std}^{235}(E)\frac{\text{U-mass}}{\text{Am-mass}}\frac{\text{at. wt(Am)}}{\text{at. wt(U)}}\frac{\text{Eff(U)}}{\text{Eff(Am)}}\times\left[\frac{F(E)-\text{FB}}{F^{235}(E)}\right], \quad (1)$$

where $\sigma_{\rm std}^{235}(E)$ is the ENDF/B-V $^{235}U(n,f)$ evaluated cross section,[12] U-mass is the mass of the ^{235}U fission sample, Am-mass is the mass of the $^{242}Am^m$ fission sample, at. wt(Am) is the atomic weight of $^{242}Am^m$, at. wt(U) is the atomic weight of ^{235}U, Eff(U) is the efficiency of the ^{235}U fission chamber, Eff(Am) is the efficiency of the $^{242}Am^m$ fission chamber, $F(E)$ is the raw fission TOF/energy spectrum of $^{242}Am^m$, FB is the spontaneous fission background of $^{242}Am^m$, and $F^{235}(E)$ is the raw fission TOF/energy spectrum of ^{235}U. Our measured value for the 2200 m/sec $^{242}Am^m(n,f)$ cross section is 6328 b with a statistical uncertainty of 0.5%. The known systematic errors evolve from uncertainties in the masses of ^{235}U and $^{242}Am^m$ and from uncertainties in efficiencies of the corresponding fission chambers [see Eq. (1)]. Adding these uncertainties yields a systematic error of ~5% relative to the ENDF/B-V $^{235}U(n,f)$ evaluated cross section[12] used to normalize the data. A summary of the statistical and systematic errors for our final $^{242}Am^m(n,f)$ data set is given in Table II.

In our 1977 experiment (see Table I), the neutron flux was measured from 10^{-2} eV to 10 keV with Li-glass scintillators, and the data reduction technique and preliminary 2200 m/sec fission cross section value have been reported in Ref. 9. In Fig. 2 we compare the 1977 and 1979 $^{242}Am^m(n,f)$ data sets (multiplied by $\sqrt{E}$) for the thermal energy region. The 1977 data showed an anomalous behavior below ~0.02 eV which could not be explained by a "near-zero" energy resonance with realistic parameters. This result inspired the remeasurement (1979) of these data down to $<10^{-3}$ eV. This remeasurement, when multiplied by the $\sqrt{E}$, yielded a constant value to $<10^{-3}$ eV, the limit of our measurement. This shape reflects the presence of the large 0.178 eV resonance and no other level close to zero energy. The preliminary 2200 m/sec ($E_n=0.0253$ eV) value of the fission cross section reported previously,[9] 7065±280 b, is high because of the anomalous behavior in the 1977 data below 0.02 eV as can be seen in Fig. 2. The shape agreement between the two measurements is excellent above 0.2 eV and the two independent data sets agree in absolute normalization to within 4%, well within our estimated uncertainty. Although we can find no error in the analysis of the 1977 thermal energy data, we believe the 1979 results to be correct. Our result of 6328±320 b for the 2200 m/sec $^{242}Am^m(n,f)$ cross section compares well with the value of 6600±300 b of Ref. 6 but is ~9% below the value of 6950±250 b reported in Ref. 8. However, the general shape of our 1979 data does agree with that of Ref. 8 in this region below 1 eV.

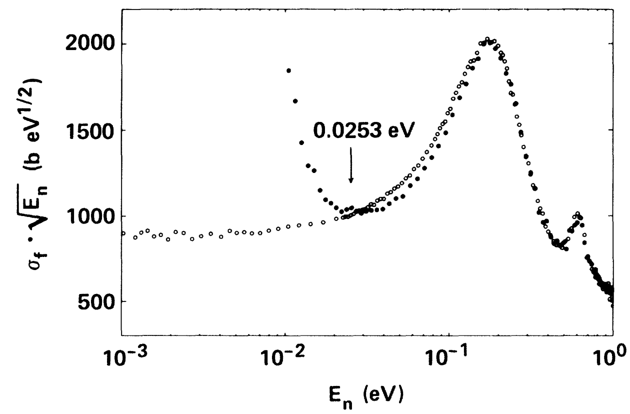

FIG. 2. Comparison of the 1979 LLNL-linac measurement of the $^{242}Am^m(n,f)$ cross section (open circles) vs the 1977 measurement (dark circles), both multiplied by $\sqrt{E_n}$, in the thermal neutron energy region (refer to Table I). The shape of the 1979 measurement, when multiplied by $\sqrt{E_n}$, is essentially constant below 10^{-2} eV. The difference between the 1979 and 1977 measurements below 0.2 eV is discussed in the text.

TABLE II. Summary of statistical and systematic errors for the 1979 $^{242}Am^m(n,f)$ cross section measurements at the Livermore linac.

Statistical error		Systematic error[a]		
Energy	$^{242}Am^m$		$^{242}Am^m$	^{235}U
Thermal	~0.5%	Mass	±1%	±2%
1 keV	~3.5%	Eff[b]	±1%	±1%
1 MeV	~0.5%			
14 MeV	~2.0%			

[a]The known systematic errors are associated with uncertainties in the measured masses of $^{242}Am^m$ and ^{235}U as well as from uncertainty in the efficiency of the corresponding fission chamber of each sample.

[b]Estimated uncertainty in efficiency from fission pulse-height distribution and calculation of percent of fission fragments lost in sample deposit.

2. *Resonance energy region*

The water-moderated target was used again for the resonance energy region (0.5 eV to ~10 keV) but the fission chamber was placed in the outer detector cave (Fig. 1) at a neutron flight path of 9.86 m. The linac was pulsed at 720 pulses per second with a pulse width of 0.5 μsec. With 6.4 mm of Pb and 1.0 mm of Cd placed in the neutron beam, data were acquired for 150 h. In this energy region the $^{242}Am^m(n,f)$ cross section cannot be measured relative to that of ^{235}U because of the abundance of resonance structure in the ^{235}U fission cross section. We therefore measured, in alternate runs with a thin lithium-glass-scintillator detector, the shape of the linac-produced neutron flux. This flux detector consisted of a 1 mm-thick piece of Nuclear Enterprises NE905 glass coupled on two opposite edges to two 5-cm diam photomultiplier tubes. The NE905 glass scintillator contained 6.6% lithium enriched in ^6Li to 95%.

A series of measurements was made with different transmission filters, including Rh, ^{238}U, Pt, Mn, Na, and Fe, placed in the neutron beam along with the Pb and Cd. From these measurements the beam-dependent background was determined and an accurate check of the energy calibration was made as well. The beam-dependent background for the fission data was determined to be less than 1% over this energy region. The beam-dependent background for the flux shape measurements ranged from 0.1% at 1 eV to 4.5% at 100 eV to 9% at 10 keV. Corrections were made to the flux shape for beam-dependent background and to the fission data for spontaneous fission background. The relative efficiency of the flux detector was unfolded using the $^6Li(n,\alpha)$ cross section shape, and the raw fission TOF spectrum was reduced to a relative cross section. This relative cross section was placed on an absolute scale by normalizing it to our measured thermal energy cross section between 0.7 and 1.7 eV.

To minimize the neutron inscattering in the resonance region, the mass of the aluminum fission chamber entrance window, hemispherical nickel substrates, and planar titanium foils was kept to a minimum. The ionization gas Ar-CO_2 was chosen over methane for the same reason. To verify that neutron inscattering was insignificant, we compare the results of our measured $^{235}U(n,f)$ cross section shape from 0.3 to 11 eV with the ENDF/B-V $^{235}U(n,f)$ cross section[12] in Fig. 3. Our $^{235}U(n,f)$ cross section shape was obtained in the same manner as the cross section shape of $^{242}Am^m(n,f)$ described above and normalized to the ENDF/B-V values. Significant inscattering would fill in the valleys between the resonances in our measured data. It is clear from Fig. 3 that our

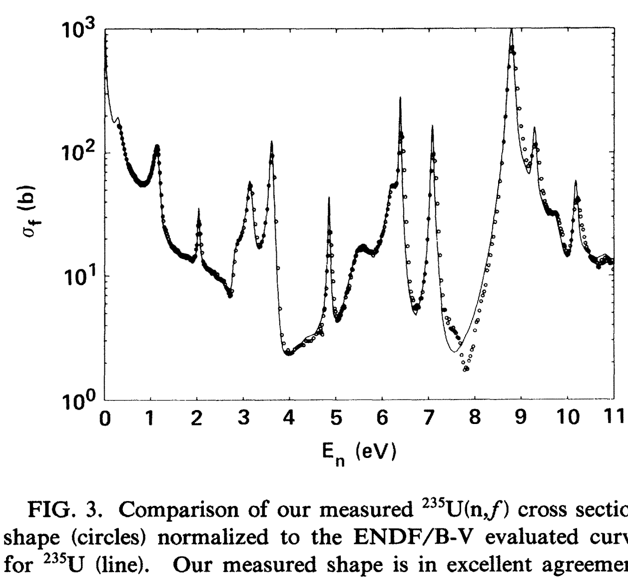

FIG. 3. Comparison of our measured $^{235}U(n,f)$ cross section shape (circles) normalized to the ENDF/B-V evaluated curve for ^{235}U (line). Our measured shape is in excellent agreement with the ENDF/B-V values except at 7.5 eV where a small resonance, seen also in the measurement of Ref. 8, has not yet been included in the ENDF/B-V evaluation. From this comparison we concluded that no significant neutron inscattering occurred in our experiment.

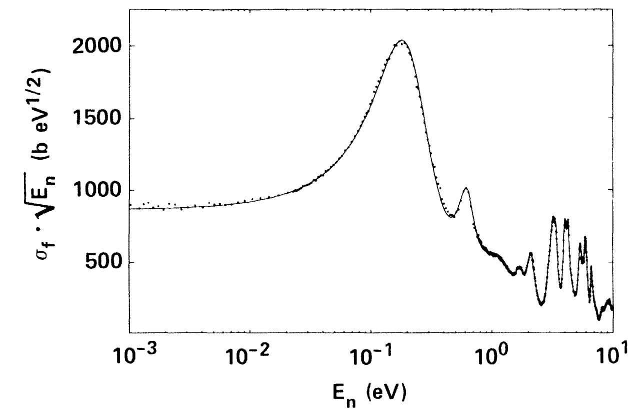

FIG. 4. Combined results of linac measurements of the $^{242}Am^m(n,f)$ cross section (multiplied by $\sqrt{E_n}$) from 0.001 to 10 eV. The influence of the first resonance at $E_n = 0.178$ eV on the magnitude of the 2200 m/sec (0.0235 eV) fission cross section is clearly evident. Our measured value for the 2200 m/sec cross section is 6328 b (±0.5% statistics). The line through the data represents a sum of Breit-Wigner resonances as discussed in Sec. II C.

measured $^{235}U(n,f)$ shape, when normalized to ENDF/B-V, is in excellent agreement with the ENDF/B-V values except at 7.5 eV where a small resonance, seen in our measurement as well as in the measurement of Ref. 8, has not yet been included in the ENDF/B-V evaluation. From these results we conclude that inscattering corrections are negligible.

Analysis of the 1977 and 1979 data sets in the 1–10 eV energy region yielded excellent agreement in energy calibration and normalization of the two data sets. Since the 1977 measurement used a fission sample of ~800 μg of $^{242}Am^m$ (Ref. 9), compared to ~200 μg in the 1979 measurement, the statistical quality of the earlier measurement is superior to that of the later data and, therefore, the earlier data are used in the final data set. In Fig. 4 our combined $^{242}Am^m(n,f)$ data are plotted (multiplied by $\sqrt{E}$) from 10^{-3} to 10 eV. The line through the data is a Breit-Wigner fit described in Sec. II C. Calculation of the fission resonance integral,

$$I_f = \int_{0.5\text{ eV}}^{20.0\text{ MeV}} \sigma_f(E)dE/E ,$$

using our results for $\sigma_f(E)$ yielded a value of 1553±78 b which agrees well with 1570±110 b of Ref. 6, but is 14% below the value of 1800±65 b reported in Ref. 8.

3. High energy region

For the high energy measurement (~1 keV to 20 MeV) the fission chamber remained in the same position as described in Sec. II B 2 but the bare Ta target was used in place of the previously used water-moderated target. The flight path distance was 13.42 m (Fig. 1). The linac was pulsed at 1440 pulses per second with a pulse width of 10 nsec and the data were accumulated for 306 h. In addition to 2.5 cm of Pb to help suppress the gamma flash, 0.2 mm of ^{10}B and 6.3 mm of Na were placed in the neutron beam to help determine the beam-dependent background. At very low energies (10—100 eV), the constant number of counts/μsec in the TOF fission spectrum implied a background of 0.246 spontaneous fission/sec which compared well with our off-line measured value of 0.250 spontaneous fission/sec. At the 2.85 keV Na resonance (transmission dip) we measured a beam-dependent background of less than 1% with respect to the raw fission TOF spectrum below the transmission dip. By interpolating from 2.85 keV to neutron energies greater than 60 MeV, where there existed essentially no neutron flux from the linac, we determined the beam-dependent background in the energy region from 10 keV to 20 MeV to be less than 1%.

The data in the high energy region were measured relative to ^{235}U and, after correction for spontaneous fission background, were reduced to an absolute fission cross section in the same manner as the thermal energy data [see Eq. (1)]. By independently measuring the thermal energy and high energy cross sections relative to ^{235}U, and normalizing the relative cross section in the resonance region to the thermal energy cross section, we can test the internal consistency and systematic error of the resonance and high energy cross sections. The overlap, in the energy region from 10^3 to 10^4 eV, of the resonance data and the high energy data yielded an agreement in both shape and normalization to within 2%.

The time of flight for the high energy region was determined from photofission events induced by the gamma flash (from electrons striking the tantalum plates in the neutron production targets). The flight path distance was determined by laser optical techniques. The time scale was verified to within ±2 nsec using carbon and lead filters to measure the location of the transmission dips of the 0.525±0.002 MeV resonance of ^{208}Pb and the 2.077±0.002 MeV resonance of ^{12}C in the fission data. The resultant uncertainty in the energy scale is ±4 keV at 1 MeV and ±0.37 MeV at 20 MeV.

In Fig. 5 we compare the results of the (averaged) high energy measurements of the $^{242}Am^m(n,f)$ cross section from the 1977 and 1979 experiments over the energy range from 10 keV to 10 MeV. These two experiments were independent measurements using different samples of $^{242}Am^m$. As can be observed from Fig. 5, there exists excellent agreement in these two data sets. Our final $^{242}Am^m(n,f)$ cross section data set includes, in this high energy region, a statistically weighted average of the 1977 and 1979 measurements.

C. Resonance analysis

Data in the energy region between 0.001 and 20 eV were fit with a sum of single level Breit-Wigner resonances allowing for Doppler broadening and resolution broadening. A 1-μsec wide square resolution function was used to fit the data in the 1—20 eV region. This width was appropriate for the width of the electron beam pulse used in the measurement of these fission data (1977). Fission data were also taken in this same energy range with electron beam pulse widths of 100 nsec and 12 nsec to determine the effect of resonance broadening due to the electron beam pulse width. Except for a few resonances, the beam pulse width did not affect the resolution of the data. In Figs. 4, 6, and 7 we show the detailed fit to the data in the 0.001—20 eV region. Resonance parameters of 48 fission resonances up to 20 eV are summarized in Table III. It is clear from the figures that these data show a complicated structure with many levels and a lack of any obvious interferences. This implies a large number of Bohr transition states[13] (fission channels) open for this nucleus since the level-level interference is essentially nonexistent in the case of many channels. This is further supported by the fact that the distribution of fission widths for these levels follows a chi-square distribution having at least ten degrees of freedom (see Fig. 8). Using the maximum likelihood method (see, for example, Ref. 14) to determine the number of degrees of freedom from the fis-

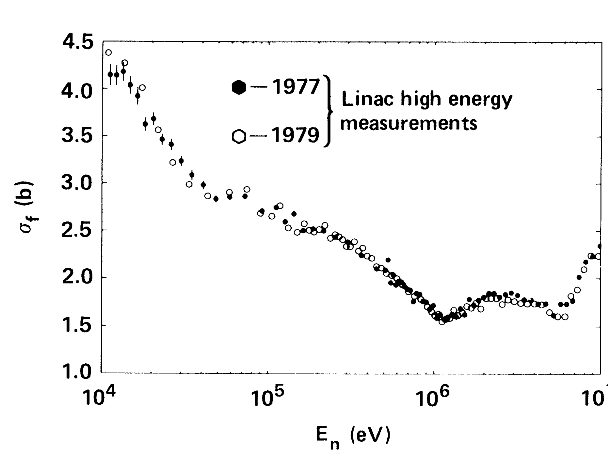

FIG. 5. Comparison of the averaged high energy $^{242}Am^m(n,f)$ cross section measurements from the 1977 and 1979 linac experiments (see Table I). Statistical errors are plotted for the 1977 data only where larger than the plotting symbols. The data in this figure are the result of two completely independent measurements using different samples of $^{242}Am^m$.

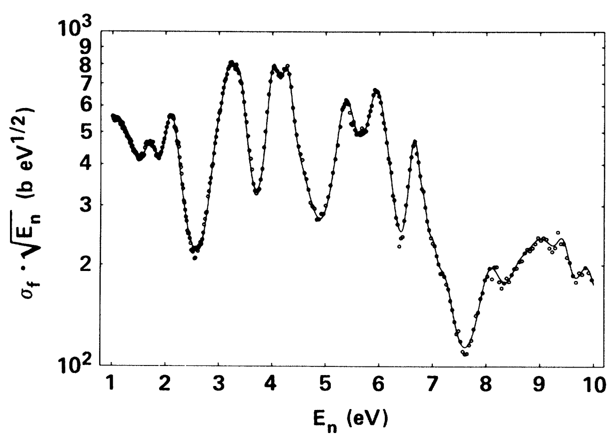

FIG. 6. Results of a Breit-Wigner sum-of-single-level fit to the $^{242}Am^m(n,f)$ cross section in the energy region from 1 to 10 eV. Several of the larger peaks are actually a sum of several levels and the complicated structure, lacking any significant interference effects, suggests a large number of fission channels open for the (compound) ^{243}Am fissioning system. Details of the resonance parameters are given in Table III.

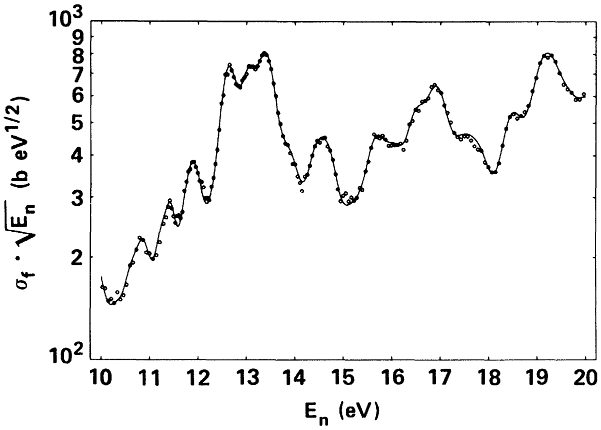

FIG. 7. Continuation of the Breit-Wigner fit to the $^{242}Am^m(n,f)$ cross section in the energy region from 10 to 20 eV. Above 20 eV the structure becomes too complicated to attempt a further fitting procedure. Details of the resonance parameters are given in Table III.

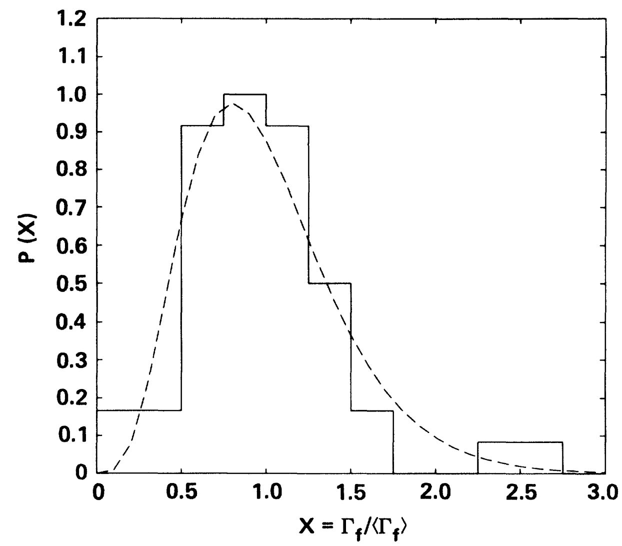

FIG. 8. Histogram of the experimental distribution of fission widths from a Breit-Wigner analysis of 48 fission resonances in the $^{242}Am^m(n,f)$ reaction up to 20 eV. The dashed line represents a chi-square distribution with $\nu=10$ degrees of freedom.

sion width data gives $\nu=10\pm2$. The effective number of fission channels, given by $N_{\text{eff}}=2\pi\langle\Gamma_f\rangle/\langle D\rangle$, is 5.7 as calculated from the parameters in Table III. It is important to distinguish between N_{eff} and ν. The quantity N_{eff} is related to the average fission width whereas ν is sensitive to the fluctuations of the fission widths and $N_{\text{eff}}\leq\nu$ (see, for example, Ref. 15). The average fission width of the 48 levels given in Table III is 363 meV.

Because of the large number of fission channels open for this nucleus it is more meaningful to fit these data with a sum of single level Breit-Wigner resonances than to utilize a multilevel (R-matrix) approach which can calculate the interference between levels. In the case where only a few fission channels are open, there sometimes ex-

TABLE III. Breit-Wigner resonance parameters (a value of $\Gamma_\gamma=50$ meV and an average g factor of 0.5 were used for all resonances) for $^{242}Am^m$ up to 20 eV.

E_0 (eV)	Γ_n (meV)	Γ_f (meV)	E_0 (eV)	Γ_n (meV)	Γ_f (meV)
0.178	0.213	240	10.30	0.025	250
0.615	0.108	200	10.62	0.105	400
1.10	0.385	999	10.87	0.120	300
1.71	0.060	250	11.25	0.110	225
2.11	0.196	330	11.43	0.090	100
2.95	0.075	230	11.79	0.140	325
3.18	0.298	310	11.92	0.230	325
3.39	0.220	260	12.62	0.790	360
4.013	0.290	220	13.04	0.470	300
4.27	0.255	215	13.41	0.940	400
4.55	0.210	600	13.90	0.280	400
5.37	0.470	422	14.42	0.280	350
5.70	0.040	250	14.68	0.375	400
5.95	0.440	350	15.15	0.100	350
6.15	0.065	300	15.67	0.680	550
6.65	0.220	170	16.06	0.195	300
6.84	0.050	70	16.48	0.470	380
7.00	0.035	80	16.92	0.950	500
7.21	0.100	380	17.50	0.475	525
8.07	0.155	500	17.82	0.285	400
8.60	0.080	500	18.47	0.610	400
9.03	0.375	850	19.07	0.800	500
9.43	0.070	220	19.31	0.900	550
9.88	0.140	420	19.70	0.450	450

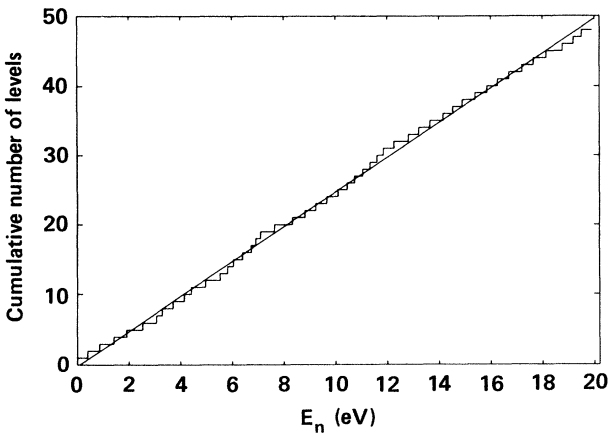

FIG. 9. Staircase plot of the cumulative number of fission resonances versus neutron energy in the $^{242}Am^m(n,f)$ reaction up to 20 eV. The straight line is a least-squares fit to the staircase and its slope gives an average level spacing of 0.4 eV.

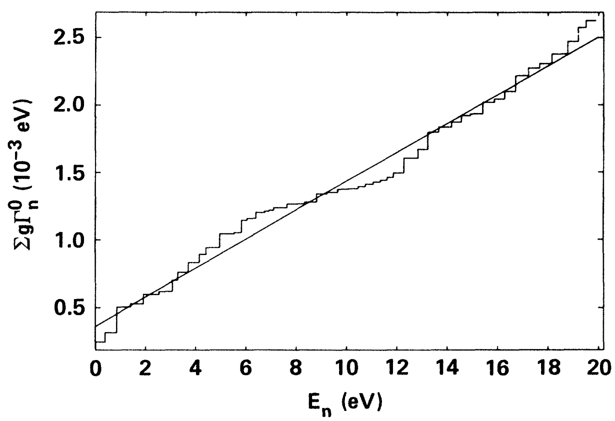

FIG. 11. Histogram of the sum of the reduced neutron widths $\Sigma g \Gamma_n^0$ as a function of neutron energy from a Breit-Wigner analysis of the fission resonances in the $^{242}Am^m(n,f)$ reaction. The slope of the least-squares fitted straight line gives the s-wave strength function, $S_0=(1.07\pm0.22)\times10^{-4}$, which is in reasonable agreement with the strength functions of neighboring nuclei.

ist interferences between resonances, and an R-matrix (or other multilevel) approach will describe this interference. In the present case where the resonance structure is so complicated, many channels would be required per level to fit these data with a multilevel formalism and no further information would be obtained than the total fission width Γ_f per level in any event. The complicated resonance structure in the $^{242}Am^m(n,f)$ cross section in some regions between 1 and 20 eV requires a "synthesis" of enough levels to fit the magnitude of the cross section while at the same time preserving its shape. In Fig. 9 we give a staircase plot of the number of levels versus neutron energy. A least-squares fit to these data yields an average level spacing $\langle D \rangle$ of 0.4 eV. This is a maximum

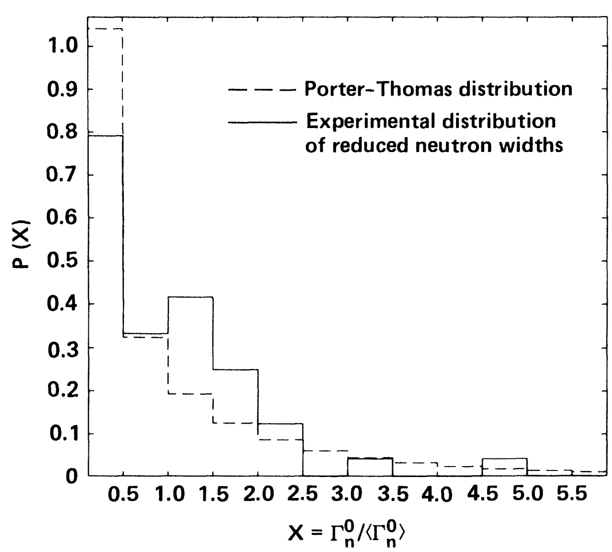

FIG. 10. The solid line is a histogram of the experimental distribution of reduced neutron widths from a Breit-Wigner analysis of 48 fission resonances in the $^{242}Am^m(n,f)$ reaction up to 20 eV. The dashed line is a Porter-Thomas distribution, i.e., a chi-square distribution with $\nu=1$ degree of freedom.

value since it can be seen from Figs. 6 and 7 that we have almost certainly missed small resonances in this region.

In Fig. 10 a comparison is made between the experimental distribution of reduced neutron widths for the 48 levels up to 20 eV and a Porter-Thomas distribution (i.e., a chi-square distribution with one degree of freedom). The average reduced neutron width is 0.109 meV. The histogram in Fig. 11 represents the variation of the sum of the reduced neutron widths $\Sigma g \Gamma_n^0$ as a function of neutron energy. The slope of the least-squares fitted straight line yields a strength function $S_0=(1.07\pm0.22)\times10^{-4}$, where the uncertainty S_0 is given by $\Delta S_0 \simeq \sqrt{2/N}\cdot S_0$. This strength function for $^{242}Am^m$ is in reasonable agreement with the strength functions of neighboring nuclei.

When comparing the level spacing of the odd-even ^{243}Am (compound) nucleus with even-even fissioning systems, a smaller average level spacing should be expected because the unpaired proton allows population of additional intrinsic excitations without the expenditure of energy necessary to first break a nucleon-nucleon pair. Because of this higher level density in the ^{243}Am compound nucleus, the probability of the first resonance occurring lower in neutron energy is more likely. The first observed resonance in the $^{242}Am^m(n,f)$ cross section (see Table III) occurs at $E_n=0.178$ eV. The low energy of the first resonance and the fact that it also happens to have a very large reduced neutron width are the reasons that $^{242}Am^m$ has the largest thermal fission cross section known.

III. 14.1-MeV MEASUREMENT

Above about 10 MeV the neutron flux produced by the bare Ta target discussed in the previous section decreases significantly, and data acquired at these energies must be averaged over wide energy bins to maintain reasonable statistics. Also, the high-gain current-sensitive preamplifiers used to drive the signal cables from the fission ionization chambers were, at the highest energies in this experiment, sensitive to radio-frequency energy (rf pickup) used to drive the linac. Although this problem was minimized by grounding and rf shielding, the data above 10 MeV are less reliable due to this effect when compared to the good accuracy of our data below 10 MeV. Therefore, we conducted a separate measurement with monoenergetic neutrons (14.1-MeV) at the LLNL-ICT accelerator using the same fission chamber described above.

A. Experimental procedure

At the ICT facility a 400 keV deuteron beam was directed to a thick tritiated titanium target to produce an intense flux of neutrons via the $^3H(d,n)^4He$ reaction. The neutron-production angle was 90° with respect to the incident deuteron beam, thus the neutron energy was 14.1 MeV with a minimum energy spread. The experimental geometry is shown in Fig. 12. The deuteron beam was pulsed at 5×10^5 pulses per second with a beam pulse width of 5 nsec; data were accumulated for 160 h. Neutrons from the source were collimated by a series of iron, polyethylene, and 30% boron-loaded polyethylene collimators to produce a 1.9-cm diam beam (Fig. 12). The neutron flight path from source to fission chamber was 3 m. Most of the massive shielding in this experiment was designed to prevent a highly efficient neutron detector, used for an accompanying fission neutron multiplicity experiment,[16] from being overwhelmed by source neutrons or those scattered from materials in the room. To further reduce background events from scattered neutrons, the entire shielded fission chamber assembly rested on a low-mass floor, $\sim$5 m above the true bottom of the experimental area. In this experiment, the TOF technique was employed to enhance the signal-to-background ratio.

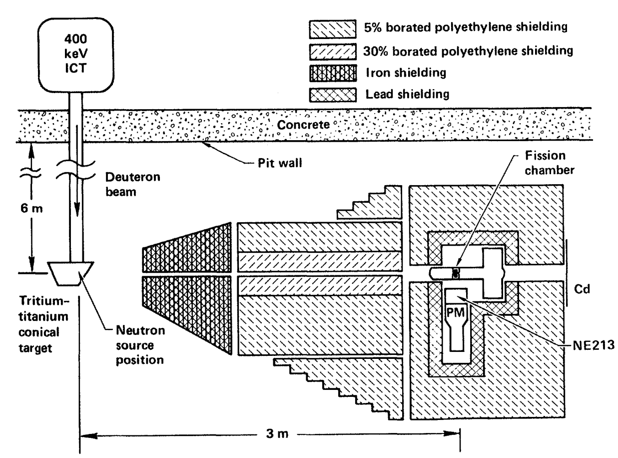

FIG. 12. Experimental geometry for LLNL-ICT 14.1-MeV $^{242}Am^m(n,f)$ measurement. A neutron detector consisting of the NE213 liquid scintillator coupled to a photomultiplier (PM) tube shown in this figure was used for an accompanying fission neutron multiplicity experiment. Most of the massive shielding was designed to prevent this neutron detector from being overwhelmed by scattered neutrons originating from the neutron-producing target assembly.

B. Experimental results

The 14.1 MeV $^{242}Am^m(n,f)$ cross section was measured relative to that of ^{235}U and reduced to an absolute cross section via Eq. (1). Because this measurement was taken a year later than the 1979 linac measurement (see Table I), the spontaneous fission rate in the $^{242}Am^m$ sample had increased to $\sim$3 spontaneous fissions per second. Even though we used the TOF technique to set a narrow (30 nsec) time window on the 14.1 MeV neutrons, which enhanced the signal-to-background ratio, the background correction was sizable. The spontaneous fission background was measured by turning off the beam and using a pulser to drive the electronics at 5×10^5 pulses per second for times roughly equivalent to the beam-on time. The spontaneous fission background was 4.5 times the neutron-induced fission signal and therefore statistical accuracy was limited to 5.1%.

The efficiencies of the $^{242}Am^m$ and ^{235}U fission chambers were obtained from fission pulse height spectra acquired during data acquisition in a manner similar to the measurements using the linac. The efficiencies were 93.1% and 92.6% for the $^{242}Am^m$ and ^{235}U chambers, respectively. Using Eq. (1) we obtain a $^{242}Am^m(n,f)$ to ^{235}U (n,f) ratio of 1.163. A systematic error of 5% is the same as given in Sec. II B 1 and Table II for the low energy linac measurement. Using the ENDF/B-V $^{235}U(n,f)$ cross section of 2.074 b (Ref. 12) at 14.1 MeV yields a $^{242}Am^m(n,f)$ cross section of 2.412 b.

IV. DATA COMPARISONS

In Fig. 13 we compare data from the present work with those of Dabbs *et al.*[8] in the energy region from 1 to 10 eV. To our knowledge, these represent the only measurements in the resonance energy region taken with both high resolution and high purity ($>$99%) samples of $^{242}Am^m$.

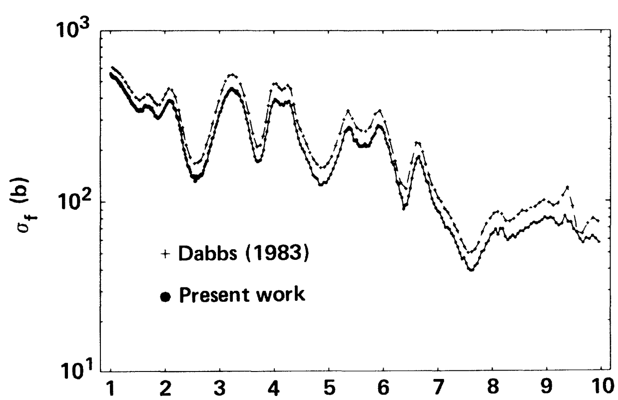

FIG. 13. Comparison of the $^{242}Am^m(n,f)$ cross section of the present work with that recently published by Dabbs *et al.* (Ref. 8) in the 1—10 eV energy region. The data points are connected with lines to help guide the eye. While the two data sets agree in energy calibration, our data average $\sim$18% lower in magnitude and there exist systematic variations about the average of several percent between 1 and 10 eV.

The two data sets agree in energy calibration and in general shape but our data average ~18% lower in magnitude than those of Ref. 8. There are also systematic variations about the average of several percent from 1 to 10 eV. In Fig. 14 we compare the same data sets as in Fig. 13 but from 100 keV to 20 MeV. The errors indicated are statistical and are shown only if larger than the plotting symbols. The statistical quality of the two data sets is significantly different and there also exists a shape difference between the two sets above 5 MeV. Again, our data average ~18% lower and systematic differences of a few percent around the average also occur over this energy region.

Other published $^{242}Am^m(n,f)$ high energy data are compared in Fig. 15 with the results of the present measurements in the energy region from 10 keV to 10 MeV. The data of Bowman *et al.*[6] were taken using a linac-produced neutron source and the data of Fomushkin *et al.*[7] above ~1 MeV were acquired using a monoenergetic neutron source [electrostatic proton accelerator utilizing the $^3H(p,n)^3He$ reaction]. The histograms in the energy region below ~1 MeV are the averaged fission cross section measurements of Seeger *et al.*[5] and Fomushkin *et al.*[7] both taken using nuclear explosions as a white source of neutrons. Except for the data of Bowman *et al.* where no errors are plotted, the errors indicated are statistical and are shown only if larger than the plotting symbols. In this region our data were acquired using a basic TOF channel width of 8 nsec. Each of our data points in Fig. 15 represents an average of ten TOF channels in the region from 370 keV to 2 MeV, of 5 channels to 5 MeV and of 2 channels to 10 MeV. From our results in the 1–5 MeV region we do not detect any "nonregularity" in the shape of the cross section or any particular structure in the cross section as suggested in Ref. 7.

At 14.1 MeV the results from the combined linac measurements yield a cross section of 2.53 b with a statistical uncertainty of 2.3%. However, this result is an average

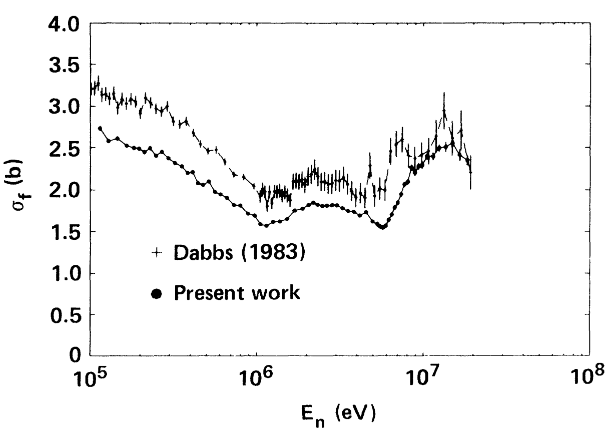

FIG. 14. Comparison of the same $^{242}Am^m(n,f)$ data sets as in Fig. 13 but from 100 keV to ~20 MeV. The uncertainties indicated are statistical and are shown only if larger than the plotting symbols. There exists a shape difference above 5 MeV as well as the normalization difference between these data sets seen also in Fig. 13.

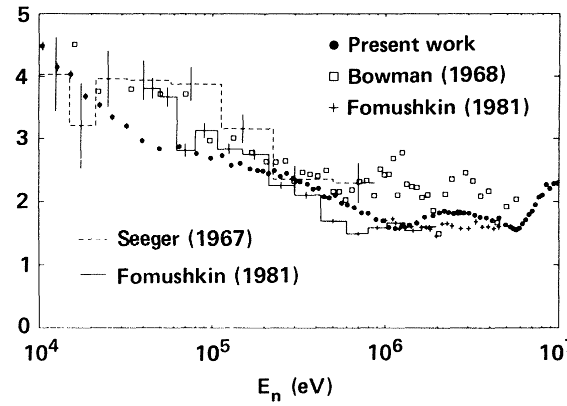

FIG. 15. Comparison of present work with previously published high energy $^{242}Am^m(n,f)$ cross section data. The filled circles are a weighted average of our 1977 and 1979 high energy data (see Fig. 5). The data of Bowman *et al.* (Ref. 6) were taken with a linac "white" neutron source and those of Fomushkin *et al.* (Ref. 7) above ~1 MeV with a monoenergetic source [$H^3(p,n)^3He$ reaction as a neutron source]. The histograms are averaged data from measurements using nuclear explosions as neutron sources: Seeger *et al.* (Ref. 5) and Fomushkin *et al.* (Ref. 7).

over ~1.3 Mev and has a systematic error larger than the 5% quoted for energies less than 10 MeV. The ICT measurement yielded 2.412 b (5% statistics) which may be compared with the recently published 14.8 MeV $^{242}Am^m(n,f)$ cross section of 2.305 b (2.2% statistics) of Fomushkin *et al.*[7] While our 14.1-MeV measurements (linac and ICT) agree within our stated uncertainty, we feel the cross section in this region is not as well established as for energies below 10 MeV. Our 14.1-MeV ICT measured value and that of Ref. 7 at 14.8 MeV probably represent best the cross section at these energies.

V. SUMMARY

We have measured the fission cross section of $^{242}Am^m$ over ten decades of neutron energy in a series of experiments using two neutron-producing facilities. We have achieved excellent statistical accuracy and small systematic uncertainty with submilligram but high isotopic purity samples of $^{242}Am^m$. Through a Breit-Wigner resonance analysis we have determined that at least ten fission channels are open at an excitation energy of 6.4 MeV ($E_n=0$) in the ^{243}Am compound fissioning system. The value for this odd-even system is several times larger than those for even-even fissioning systems. The large thermal fission cross section of $^{242}Am^m$ can be explained by the increased level density due to the odd-even nature of the compound system and the chance occurrence of the first fission resonance having a very large neutron width.

We have compared our measured $^{242}Am^m(n,f)$ cross section with previously published results and find sizable differences. The only other work which can be compared with the present series of measurements in terms of the energy range covered, the energy resolution with which the data were taken, and the sample purity used is that of Ref. 8. Our results are ~18% lower in magnitude than those. Reasons for this large normalization difference between the two works could include errors in the fission sample masses used in the experiments or in the efficiencies of detecting fission events in the fission ionization chambers. Our efficiencies were determined independently for the thermal energy and high energy measurements, were within $\sim 1\%$ in both cases, and were systematically higher (98.2% for $^{242}Am^m$ and 95.9% for ^{235}U) than the efficiencies reported in Ref. 8 (76% for $^{242}Am^m$ and 85.6% for ^{235}U). The normalization agreement between our thermal and high energy data as well as the agreement between our 1977 and 1979 measurements gives confidence in our efficiency determinations.

Since all the $^{242}Am^m$ samples used in our measurements as well as those of Ref. 8 were prepared by the same laboratory (LLNL), and we find excellent agreement between our own series of measurements using different samples, we believe the large normalization disagreement between our data and those of Ref. 8 is unlikely to be caused by large errors in the $^{242}Am^m$ sample mass determinations. The common link in our own series of experiments is the mass of ^{235}U against which we measured the $^{242}Am^m$ fission cross section. Our efforts to establish the ^{235}U mass accurately through measurements at the National Bureau of Standards are documented in this work.

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