could not be achieved, a more complete and systematic study might prove this preliminary conclusion incorrect. In fact, it has been demonstrated[14] that the need for a six-parameter potential is not fully realized unless the elastic data is fit over a wide angular range. Moreover, the use of a six-parameter potential (i.e., real and imaginary wells of different shapes) admits an extension to DW calculations with complex coupling. This has been reported[14] to yield improved fits to the inelastic data. Consequently any definitive investigation involving either of the above extensions requires experimental cross sections which extend over a large angular range.

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ACKNOWLEDGMENT

The authors would like to express their gratitude to Dr. Howard C. Volkin of National Aeronautic and Space Administration, Lewis Research Center, for making available the optical-model code from which the one used in the present investigation was modeled.

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Symmetry of Neutron-Induced ^{239}Pu Fission at Individual Resonances*

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Neutrons resolved in energy by time-of-flight from an underground nuclear explosion ("Petrel," 11 June 1965) have been used to produce resonance fissions in ^{239}Pu in sufficient numbers to permit radiochemical measurements of the symmetry of fission at individual levels in the energy region 15 to 82 eV. About two-thirds of the levels examined are characterized by fission less symmetric than thermal-neutron fission. At these resonances the modal yield of the nearly symmetric fission product ^{115}Cd is approximately $\frac{1}{3}$ the thermal yield. For the remainder of the levels, fission is more symmetric; the modal yield of ^{115}Cd is approximately $\frac{3}{2}$ the thermal yield. There is correspondence between groupings by enhanced or decreased fission symmetry, level widths ("broad" or "narrow"), and some recently measured J values. Based on relative fission symmetry, spin assignments are proposed for 22 levels. It is suggested that those levels for which the ^{115}Cd yield is low represent 1+ states; the higher ^{115}Cd yields presumably occur at levels which represent 0+ states. The 0+ levels are broad compared to the more readily recognized and categorized 1+ levels. The proposed assignments are: for the $J=1$ group, resonances at 17.7, 22.3, 26.3, 41.5, 44.6, 47.7, 50.2, 52.7, 55.8, 59.4, 66.0, 75.2, and, with less confidence, 15.4 eV; for the $J=0$ group, resonances at 49.8, 57.6–58, 61.1, 66.8, 81.7, and, with less confidence, 32.4 eV. In addition, from other published data, we suggest $J=1$ for the 0.3-eV level and $J=0$ for the first negative-energy level.

INTRODUCTION

It has frequently been suggested that symmetry of fission can vary with spin and parity and might, therefore, change at resonances. Experiments to date indicate that changes do occur in yields of symmetric fission products at resonances in ^{239}Pu, ^{235}U, and ^{233}U fission,[1–6] but they have not been directly related to known values of spin or to changes in parity. A decrease of a factor of 3 in the ^{115}Cd yield has been measured at the 0.3-eV resonance in ^{239}Pu.[6] We have previously reported mass symmetry measurements for ^{239}Pu fission in which significant variations in the ^{115}Cd yield were observed in the neutron-energy region 200–500 eV but the resolution of the experiment did not permit identification of these variations with individual resonances.[5] The present experiment extends these ^{239}Pu measurements to lower neutron energies and higher resolution.

In the ground state ^{239}Pu has spin and parity $\frac{1}{2}+$ and accordingly, for s-wave neutron absorption, the possible compound nuclear states in ^{240}Pu are 0+ and 1+. Examples of both J values have been observed among the resonances.[7–9] It has also been noted that

* Work done under the auspices of the U. S. Atomic Energy Commission.

[1] Los Alamos Radiochemistry Group, Phys. Rev. **107**, 325 (1957).
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[5] G. A. Cowan, B. P. Bayhurst, R. J. Prestwood, J. S. Gilmore, and G. W. Knobeloch, *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, March 1965*, (International Atomic Energy Agency, Vienna, 1965) Vol. I, p. 347.
[6] R. B. Regier, W. H. Burgus, R. L. Tromp, and B. H. Sorensen, Phys. Rev. **119**, 2017 (1960).
[7] J. S. Fraser and R. B. Schwartz, Nucl. Phys. **30**, 269 (1962).
[8] L. M. Bollinger, R. E. Cote, and G. E. Thomas, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 127.
[9] G. D. Sauter and C. D. Bowman, Phys. Rev. Letters **15**, 761 (1965).

some very broad levels exist in the ^{239}Pu total cross section and that these broad levels may be predominantly 0+ since channel theory predicts this is the lowest lying state through which fission of the ^{240}Pu nucleus can proceed.[10,11]

EXPERIMENTAL PROCEDURES AND RESULTS

The general details of this experimental approach have been described previously[3,4]; we refer to the method as the "wheel" experiment. In the present version the flight path was 181.6-m long; the collimating slit (which defines a neutron acceptance time on a moving wheel) was 0.2-cm wide at the rim and 10-cm long, terminating at an outer radius of 27.94 cm; the ^{239}Pu metal target (99.42% pure Pu, 94.2% 239, 5.4% 240, 0.4% 241) was 0.384-mm thick; the wheel speed was 8775 cm/sec at the rim; and the exposure time for a given piece of target was 22.79 μsec, corresponding to an energy resolution of 0.125 μsec/m. Neutrons were moderated to a broad epithermal distribution peaking at $\sim 3\times 10^{10}$ neutrons/cm^2 in 22.79 μsec in the 50- to 80-eV energy region. The spectral distribution

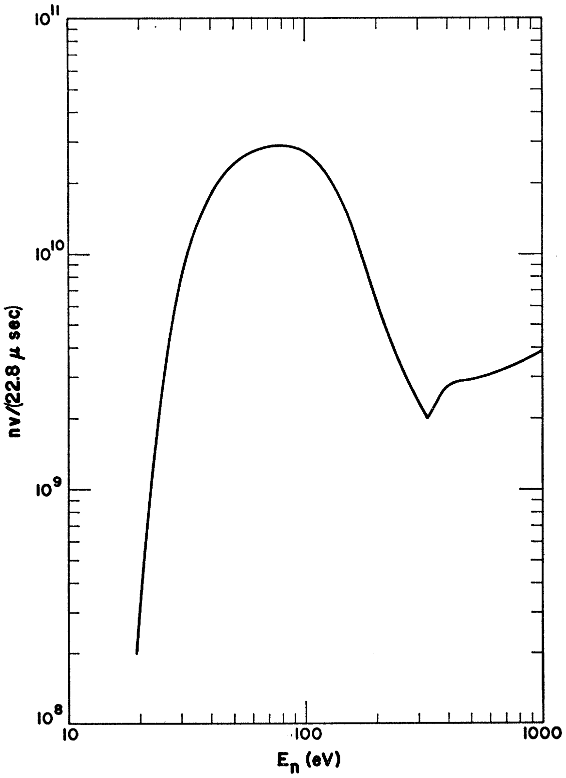

FIG. 1. Neutron flux at ^{239}Pu target in time interval defined by collimating slit and wheel speed.

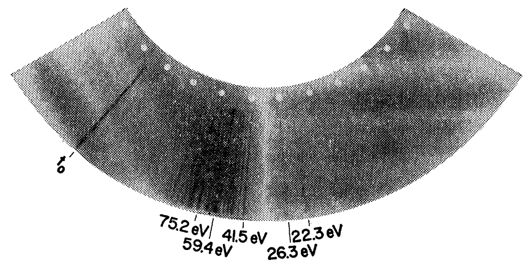

FIG. 2. Autoradiograph of plutonium metal produced by contact exposure of x-ray film to fission products in target. An energy scale is provided by identification of lines at t_0 (gamma-ray- and fast-neutron-induced fission) and a few strong resonances.

is plotted in Fig. 1.[12] The lead and polyethylene moderator, which was heated by neutrons and then accelerated by exploding debris to a velocity of $\sim$5.5 cm/μsec, cut off neutrons below 15 eV. Exposure of the wheel was terminated by closure of the line-of-sight near the origin and by the closure of a cadmium-coated shutter near the wheel at approximately 15 msec after t_0. The sled-mounted wheel was immediately pulled several hundred feet from the line-of-sight position and was dismounted thirty minutes after t_0. Autoradiographs of the target sectors were completed within two hours.

The autoradiographs of the two 60^0-^{239}Pu sectors on which all the exposure occurred are shown in Fig. 2. Dark bands caused by resonance fission are observed at each of the known strong levels in the epithermal region above 15 eV. These bands of fission products were cut from the metal in strips of 0.5- to 1.5-g weight, dissolved, and analyzed for ^{111}Ag, ^{115}Cd, and ^{99}Mo. The ratio of 2.3-day ^{115}Cd activity to 2.7-day ^{99}Mo activity, defined as $r_{Cd/Mo}$, is taken as an index of symmetry. We assume that the yield of ^{99}Mo is constant and that changes in $r_{Cd/Mo}$ are a measure of the variable yield of ^{115}Cd. The uncorrected results of analyses for ^{99}Mo and ^{115}Cd on samples taken from the 15-eV to 200-eV energy region are plotted in Fig. 3, together with an approximate smoothed background calculated from fissions observed in valleys between well-separated resonances.

The background consists chiefly of neutrons scattered from the tapered stainless steel wheel which varies in thickness from 0.8 to 1.0 cm immediately behind the target. An additional smaller correction arises from fission neutrons produced in the target. Since most of the background is due to neutrons only slightly degraded in energy from the incident energy, the level of the background is affected by the presence of nearby resonances. The smoothed background curve is, therefore, uncertain to 10–15%. Since we are interested in the yield of ^{115}Cd relative to ^{99}Mo rather than in an absolute yield, the background uncertainty does not add significantly to errors in the measurement of $r_{Cd/Mo}$ except where two adjoining levels with different

[10] J. Blons, H. Derrien, A. Michaudon, P. Ribon, G. de Saussure, Conference on Study of Nuclear Structure with Neutrons, Antwerp, Belgium, 1965 (unpublished).

[11] C. A. Uttley, Conference on Study of Nuclear Structure with Neutrons, Antwerp, Belgium, 1965 (unpublished).

[12] P. Seeger (private communication).

TABLE I. ^{99}Mo and ^{115}Cd data to 204 eV and related resonance parameters.

Energy interval E_1 (eV	Energy interval E_2 (eV)	Corrected $A_{^{99}Mo}$ (counts/min-g)	Corrected 100 $r_{Cd/Mo}$	Resonance energy[a,b]	Γ (meV)[a]
15.8	16.1	2006	0.78[c]	*15.42*	700
17.0	17.4	610	1.39		
17.4	17.7	1207	0.89	*17.66*	74
17.7	18.0	343	1.47		
22.0	22.4	3150	1.01	*22.28*	90
22.4	23.0	664	0.77		
23.5	23.9	(483)[d]	(1.34)	23.93	65
25.7	26.2	3931	0.74		
26.2	26.9	769	0.58	*26.29*	83
30.7	32.0	(467)[d]	(4.7[c])	*32.38*	165
41.1	42.0	9052	0.69	*41.52–41.76*	52–105
42.0	42.6	2072	0.72		
44.1	44.7	4752	1.18	*44.59*	58
44.7	45.7	(1008)[d]	(1.11)		
46.2	47.2	3984	0.73		
47.2	47.9	19322	0.8	*47.74*	300
47.9	48.7	11059	0.69		
48.7	49.5	6924	1.86		
49.5	50.3	15366	1.55	*49.85–50.22*	810–57
50.3	51.3	4238	1.74		
52.0	53.0	9564	1.19	*52.74*	68
53.0	53.6	3597	1.44		
54.7	55.3	2341	1.35		
55.3	56.4	8019	1.34	*55.79*	~50
56.4	57.4	18461	2.64		
57.4	58.1	47945	2.75	*57.6–58*	*593–850*
58.1	58.8	38084	2.50		
58.8	59.9	43922	1.61	*59.39*	190
59.9	60.7	20774	2.38		
61.3	62.5	12565	2.94	*61.1*	>2000
62.5	63.8	12133	2.30	63.4	*n*[e]
63.8	65.0	8662	2.28		
65.0	66.5	34034	1.96	*65.96*	137
66.5	67.5	9600	2.60	*66.83*	*b*[f]
67.5	68.9	4335	2.76		
68.9	70.3	2482	2.18		
72.3	73.8	11416	0.65		
73.8	75.5	40873	0.72	*74.31–75.21*	71–162
75.5	76.9	6470	1.25		
76.9	78.6	2388	1.83		
78.6	80.5	4191	2.18		
80.5	82.4	12652	2.85	*81.7*	>1500
82.4	84.3	21678	2.36	*83.3*	?
84.3	85.9	31319	2.12	*84.9*–85.6	≥2000
85.9	89.2	9163	2.27	*86.0*–87.9	?–?
89.2	91.4	5346	1.40	*90.9*	157
91.4	95	3311	2.13	*92.6*	*n*[e]
95	97	7987	2.36	95.5–*96.7*–97.6	270–>1400–400
106	108	6401	1.83	*106.7*	80
114	117	1970	1.52	*116*	220
117	120	3630	1.17	*119*	140
130	132	3674	2.09	*132*	3800
132	137	2375	1.59	134–*135*–*137*	?–?–70
137	140	1086	0.47		
140	143	1776	0.69	140–*143*	?–?
143	146	1642	0.94	146	200
155	158	2708	1.91	*157*	760
158	163	1199	0.81	*161*	550
163	167	1797	0.34	*164*–167	90–95
181	188	(902)[d]	(2.59)	183–*185*	1800
188	193	1061	2.03	188–190	?–160
193	198	2273	1.80	*195*–196	400
198	204	2526	1.52	199–204	160–365

[a] Resonance energies and widths are taken from Refs. 10, 11, 13, and 14.
[b] Italicized energies indicate more significant levels.
[c] Not on resonance.
[d] Background correction is more than 80% of observed activity.
[e] *n* indicates that the apparent Γ <0.4 eV.
[f] *b* indicates that the apparent Γ >0.4 eV.

mass symmetry characteristics produce a "mixed" background, thereby degrading the resolution for the separate levels. Wherever possible, background corrections were estimated from adjoining samples rather than from the smoothed curve.

The values of $r_{Cd/Mo}$ up to 200 eV, corrected for background, are listed in Table I. Where the background amounts to 85% or more of the total for both ^{99}Mo and ^{115}Cd activity, the results are not included in the tabulation. Where the background is larger than 80% of either activity, the corrected results are bracketed. Table I also includes all resonance energies

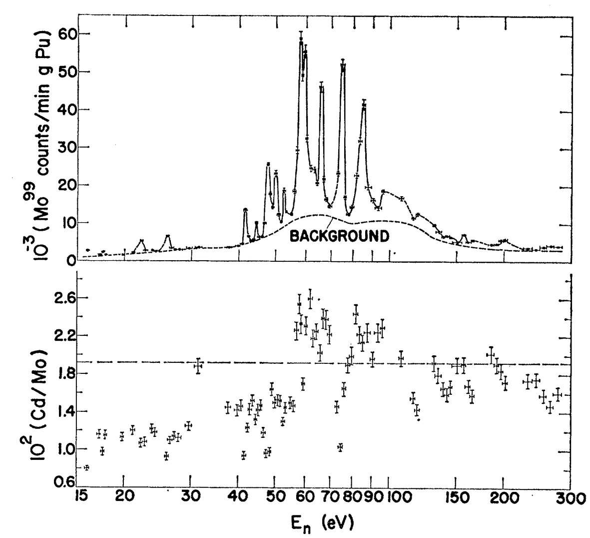

FIG. 3. Experimental results on fission density, represented by ^{99}Mo activity, and $^{115}Cd/^{99}Mo$ activity ratios from 15–300 eV. Horizontal lines represent width of metal cuts. Vertical lines represent estimated errors. The dashed background line represents fissions induced by sources other than collimated neutrons. The dashed line in the Cd/Mo plot represents the thermal fission value of this ratio (1.92×10^{-2}).

reported by Saclay and Harwell[10,11,13,14] between 15 and 200 eV and, where suitable data are available, total widths are listed for each of these levels. If widths are not known, we have attempted where possible to qualitatively categorize the levels as "broad" or "narrow" ("*b*" for apparent $\Gamma > 0.4$ eV; otherwise "*n*"). Values for $r_{Ag/Mo}$ are not presented. They correlate closely with the $r_{Cd/Mo}$ values; however, the relative change from resonance to resonance is only 0.24 ± 0.01 as large as for $r_{Cd/Mo}$.

Up to 82 eV it is possible to estimate individual contributions of two closely neighboring levels, where they occur in the same sample, by use of a single-level formula calculation incorporating published or, if necessary, assumed resonance parameters together with resolution corrections for an acceptance time of 22.8 μsec. In Table II there is listed a "best" value of $r_{Cd/Mo}$ for each strong resonance up to 81.7 eV. Resolution of neighboring levels by application of single-level formula corrections has been attempted at 49.8, 50.2, 52.7, 55.8, 57.6-58, 59.4, 61.1, 66.0, and 66.8 eV. The distribution of all values for $r_{Cd/Mo}$ is plotted in Fig. 4. Two groups are apparent with modal values of 0.7×10^{-2} (Group I) and 2.9×10^{-2} (Group II).

Probable errors can be estimated with some confidence for measurements where relatively little interference exists from neighboring levels and where nearby

[13] G. de Saussure, J. Blons, C. Jousseaume, A. Michaudon, and Y. Pranal, Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965 (unpublished), Vol. I, p. 205.

[14] G. D. James, Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, March, 1965 (unpublished), Vol. I, p. 235.

TABLE II. Values of $r_{\mathrm{Cd/Mo}}$ at resonances, corrected for neighboring levels.

E_0 (eV)	100 $r_{\mathrm{Cd/Mo}}$	*J* Ref. 7	Ref. 8	Ref. 9	Proposed (this paper)
15.42	0.78 ± 0.20^{a}		0	0	1^{a}
17.66	0.50 ± 0.20	1	1	1	1
22.28	1.05 ± 0.19	0		1	1
26.29	0.75 ± 0.09				1
32.38	$(4.7^{a})^{b}$			0	(0^{a})
41.52	0.67 ± 0.07	1	0	1	1
44.59	1.14 ± 0.22	0	1	1	1
47.74	0.69 ± 0.09			0	1
49.85	$\lvert 2.33\rvert^{c}$				0
50.22	0.7^{d}				1
52.74	1.03 ± 0.17		1	1	1
55.79	$\lvert 0.4\rvert^{c}$				1
{57.6	2.83 ± 0.30				0
58					0
59.39	$\lvert 1.36\rvert^{c}$				1
61.10	3.02 ± 0.45				0
65.96	$\lvert 1.94\rvert^{c}$				1
66.83	2.79 ± 0.60				0
75.21	0.68 ± 0.17			1	1
81.7	2.85 ± 0.43				0

[a] Not on resonance.
[b] Background correction is more than 80% of observed activity.
[c] Poorly resolved from neighbor of apparently different symmetry characteristics.
[d] Assumed value; group assignment relatively certain.

background measurements exist. An error analysis applied to the results at 17.7, 22.3, 26.3, 41.5, 44.6, 47.7, 52.7, and 75.2 eV indicates that a constant value for $r_{\mathrm{Cd/Mo}}$ of $(0.73\pm0.04)\times10^{-2}$ for all levels in Group I is consistent with the individual experimental errors quoted for values in Table II. For resonances at 57.6–58, 61.1, 66.8, and 81.7 eV, neighboring levels offer little interference and background uncertainties contribute most of the quoted error. Uncertainties in the background have been estimated on the high side ($\pm15\%$). For these levels a constant value of $r_{\mathrm{Cd/Mo}}$ of $(2.9\pm0.2)\times10^{-2}$ is consistent with the individual estimated over-all errors.

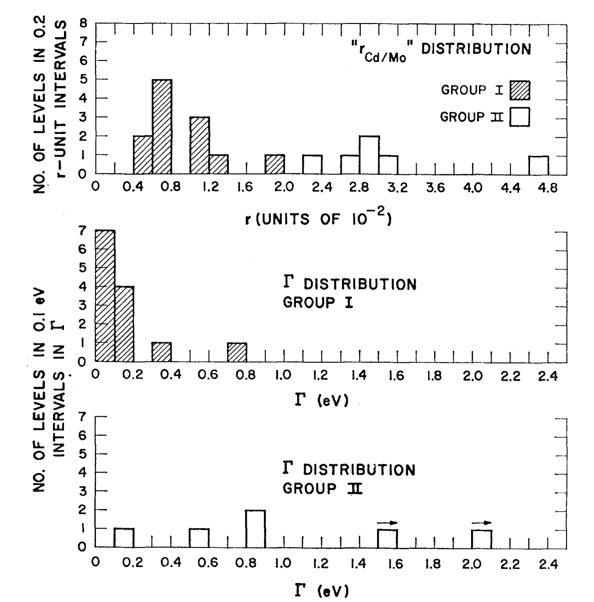

FIG. 4. The frequency distributions in $r_{\mathrm{Cd/Mo}}$ at resonances and in associated resonance widths. The distribution in $r_{\mathrm{Cd/Mo}}$ is taken from the data presented in Table II. The corresponding distributions in Γ are taken from the Γ values presented in Table I. Bars with arrows indicate lower limits on Γ.

These results are in agreement with the value reported by the MTR investigators[6] for $r_{\mathrm{Cd/Mo}}$ at 0.3 eV and the inferred value for the negative energy level; converted to comparable units, their reported values are 0.6×10^{-2} and 3×10^{-2}, respectively.

Inspection of Table II indicates a correlation between r values and level widths. This relationship is indicated in Fig. 4 which shows the distribution in Γ where measurements exist for levels in Group I and Group II. It seems reasonably clear that narrow levels fall predominantly in Group I and broad levels predominantly in Group II. Although the data are not included in Fig. 4 because it can not be demonstrated that individual levels are well-resolved above 82 eV, there is further qualitative correspondence between apparently very broad levels and higher values of $r_{\mathrm{Cd/Mo}}$ at 96, 132, 155, and 185 eV and between narrow levels and lower values of $r_{\mathrm{Cd/Mo}}$ at 119, 137, and 164 eV.

Table II also contains J values wherever measurements exist for these levels.[7–9] There is agreement between Group I and the Sauter-Bowman group of 1+ levels.[9] A clear-cut disagreement exists only at 47.6 eV.

The relative populations in Group I and Group II are 65% and 35%, respectively. If all levels in the Saclay reports up to 82 eV are counted and assigned to either the "narrow" or "broad" category, the relative populations are 72% and 28%, respectively, suggestive of a $2J+1$ distribution (75%-25%). Levels counted as "broad" were at 11.5, 15.4, 23.9, 49.8, 57.6, 58, 61.1, 66.8, and 81.7 eV. The relative areas for fission are approximately 30% under "broad" resonances and 70% under "narrow" resonances.

DISCUSSION

We suggest that $J=1$ for levels in Group I and $J=0$ for levels in Group II. This assignment would be consistent with the following considerations based on the channel theory of fission[15,16]:

1. Such theory predicts that 0+ states have a higher probability of symmetric fission than 1+ states.

2. The theory predicts that 0+ states will have lower fission thresholds and, consequently, broader widths than the 1+ states.

3. The apparent paucity of 0+ levels of narrow

[15] A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956) Vol. 2, p. 151.

[16] J. A. Wheeler, in *Fast Neutron Physics*, edited by J. B. Marion and J. L. Fowler (Interscience Publishers, Inc., New York, 1963), pp. 2051–2184.

width suggests, from Porter-Thomas arguments,[17] that several channels are available for 0+ fission, in qualitative agreement with channel theory prediction of lower thresholds for 0+ fission.

The thermal value of $r_{Cd/Mo}$ (1.92×10^{-2}) is apparently due to a mixture of contributions of fission from the 0.3-eV level (lower symmetry[6]) and the negative energy level or levels. For these levels we suggest $J=1+$ and $0+$, respectively. From the characteristic values of $r_{Cd/Mo}$ at 0+ and 1+ levels, it can be calculated that the 0.3-eV level contributes 45% and the negative energy level 55% of observed fissions, values in good agreement with Regier *et al.*[6] who used the Breit-Wigner single-level formula and published resonance parameters to estimate contributions of 48% and 52%, respectively.

The present analysis has features similar to the interpretation of cross section data for ^{233}U by Moore *et al.* based on multi-level analysis[18,19]; they hypothesized that two groups of levels of different width exist in ^{233}U, that these groups can be explained as $J=2+$ (broad) and $J=3+$ (narrow), and that the 2+ and 3+ levels are characterized by higher and lower fission symmetry, respectively, compared to thermal fission. Moore *et al.*[20] have also proposed the existence of two groups of levels of different average width for ^{241}Pu. Similar suggestions have been made for ^{235}U and ^{239}Pu.[9,21,22]

Our previously published ^{235}U data[3,4] contain some fragmentary evidence for groups of broad and narrow levels. Total widths for seven levels exhibiting decreased fission symmetry vary from 0.037 to 0.14 eV, with an average value of 0.085 eV[23]; increased fission symmetry is observed at 14, 25.6, 35.2, and 39.4 eV with measured or apparent level widths which are all broader than 0.085 eV. If the indications of the "wheel" experiment and the prediction of channel theory concerning symmetry of fission for ^{235}U are correct,[16] then the narrower group contains 3− states and the broader group 4− states. However, channel theory also predicts the opposite assignments, 4− and 3−, for "narrow" and "broad" groups, respectively, corresponding to the presumed relative energy requirements for fission through these channels.[16] One possible explanation for this apparent contradiction is that mass symmetry, which is known to increase with excitation energy, may depend more importantly on the amount of excess energy at the fission barrier than on the spin and parity of the channel. The lowest lying available channels would then presumably always be characterized by larger widths and by greater mass symmetry in fission than channels with a higher barrier against fission.

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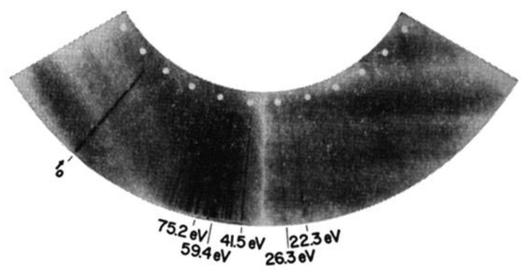
[18] M. S. Moore, L. G. Miller, and O. D. Simpson, Phys. Rev. **118**, 714 (1960).
[19] M. S. Moore and C. W. Reich, Phys. Rev. **118**, 760 (1960).
[20] M. S. Moore, O. D. Simpson, T. Watanabe, J. E. Russell, and R. W. Hackenbury, Phys. Rev., **135**, B945 (1964).
[21] I. V. Kirpichnikov, K. G. Ignatyev, and S. I. Sukhoruchkin, Soviet J. At. Energy **16**, 211 (1964).
[22] K. G. Ignatyev, I. V. Kirpichnikov, and S. I. Sukhoruchkin, Soviet J. At. Energy **16**, 110 (1964).
[23] A. Michaudon, thesis, University of Paris, Report No. CEA-R 2552, 1964 (unpublished), Fig. D. 3.1.



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