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Symmetry of Neutron-Induced Pu^{239} Fission in Resonance Region

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REVISED ABSTRACT

Symmetry of fission produced in Pu^{239} by time-of-flight energy-resolved neutrons has been radiochemically measured in the resonance region by a neutron time-of-flight method, dubbed the "wheel" method, which has been described previously [1,2].

Data were obtained from 200 ev to 600 ev in energy intervals ranging from 10 ev to 25 ev in width, containing three to twelve resonances per sample. Both increases and decreases in symmetry of fission were observed as compared with thermal neutron fission of Pu^{239} . Measured by the ratio of Cd^{115} , a "valley" fission product, to Mo^{99} , a "peak" fission product, the symmetric yield decreased by 18% at 250 ± 4 ev, 29% at 307 ± 5 ev, and 33% at 395 ± 6 ev. The symmetric yield increased by 16% at 206 ev. The average symmetry of fission over the energy region 196 ev to 585 ev showed a decrease of 8% from the thermal value. Changes in the ratio $\text{Ag}^{111}/\text{Mo}^{99}$ appeared to be about one-third as great as changes in the $\text{Cd}^{115}/\text{Mo}^{99}$ ratio from level to level.

The ratio $\text{Ba}^{140}/\text{Mo}^{99}$ showed no systematic changes from the thermal value within analytical error and the average is within 1% of the thermal ratio, which permits the assumption that Mo^{99} and Ba^{140} yields are not variable for resonance fission and are the same as in thermal fission.

Neutrons from the explosion of a small nuclear device on April 29, 1964 have been used to examine the symmetry of Pu^{239} fission in the resonance region. The principles of the experimental technique used have been described in detail elsewhere [1,2]. Briefly, fissile material is revolved rapidly past a collimating slit which defines a beam of neutrons resolved in energy by flight through a long, evacuated pipe. Fissions in various parts of the wheel are caused by neutrons in a known energy group and can be examined by radiochemical means for changes in symmetry.

The isotope Pu^{239} has a ground state spin $1/2+$. For s+ wave neutron absorption, the compound nuclear states leading to fission are $0+$ and $1+$. It has been reported [3] that fission at the 0.297 ev resonance in Pu^{239} produces five-fold less symmetric fission than is observed in thermal fission. If this effect on symmetry is due to spin, it may be expected that resonances of the other possible spin, if they are large enough and sharp enough to be characterized as resonances, will have some other characteristic symmetry. At low excitations fission may occur through a predominantly $0+$ channel with a symmetric α_3 -vibrational wave function which permits a symmetric mode of fission. It may be possible to build a $1+$ state at low excitation energy, not involving a single particle state, for which symmetric fission should be forbidden [4]. The present investigation was undertaken principally in order to further investigate the applicability of channel analysis of fission in predicting symmetry of fission at individual resonances [5,6,7]. Other measurements with U^{235} [1,2] and U^{233} [8] have, in general, indicated that differences exist in

the frequency of symmetric fission at various resonances. The hypothesis that such differences are related to spin and might fall into two more or less well-defined groups, according to the spin of the nuclear state, has not yet been adequately tested.

1. Experimental Procedure

A 60 cm. diameter wheel with 0.038 cm. thick Pu^{239} metal fastened to the outer third of the wheel face was revolved past a tapered steel collimating slit 10 cm. long and 0.2 cm. wide at a distance of 2.630×10^4 cm. from the source. The velocity of the wheel at the outer edge of the beam was 8909 cm/sec. Under the conditions of this experiment the distance at which neutrons of a given energy fall measured along the rim of the Pu metal is given by the equation

$$S = 169.3/E^{\frac{1}{2}}$$

where S is in cm. and E is in ev.

The collimating slit width defined an exposure time or channel width of 22.4 μsec . The pulse width at the source was almost negligible compared to this interval. Thus the energy resolution corresponded to 0.09 $\mu\text{sec}/\text{m}$. At 100 ev and 200 ev for example, the time interval defined by the slit corresponds to resolutions of 2.4 ev and 6.7 ev, respectively. The level spacing in Pu^{239} is approximately 2 ev. A composite lead, iron, and polyethylene moderator was placed near the source. It was designed to produce $> 10^9$ neutrons/ cm^2 -ev below 100 ev at the wheel, adequate for the resolution of single levels.

The wheel was recovered immediately after the event and the location of the resonance band was established by contact autoradiography on X-ray film. The autoradiograph of the relevant portion of the wheel was fastened to the metal and the regions of maximum and minimum contrast were sheared in strips of ~ 0.2 cm. maximum width, corresponding to the collimating slit width. The metal was cleaned, dissolved, and analyzed for Mo^{99} and Ba^{140} , "peak" fission products, and Ag^{111} and Cd^{115} , "valley" fission products.

2. Experimental Results

2.1 Experimental data

The sample schedule and results are summarized in Table I. Background corrections are shown in Fig. 1 and discussed in 2.2. The ratios "r" indicate the ratio of Cd^{115} or Ag^{111} activity to Mo^{99} activity at a given neutron energy. The Pu^{239} thermal ratios, r_{th} , are 2.175×10^{-2} for $\text{Ag}^{111}/\text{Mo}^{99}$ and 1.911×10^{-2} for $\text{Cd}^{115}/\text{Mo}^{99}$. No data were obtained at energies below 200 ev due to an unexpectedly high acceleration of the moderator by nuclear shock, leading to a neutron spectrum in the laboratory frame-of-reference which peaked at about 300 ev and was cut off absolutely at 200 ev. Therefore, single levels were not resolved in this experiment. The probable number of levels included in each sample varied from 3-4 at 200 ev to 10-12 at 600 ev.

The fraction of Pu^{239} converted by fission versus neutron energy in the resonance region is presented in Fig. 2 together with the Pu^{239} fission cross section taken from the Karlsruhe compilation [9]. The corrected activity ratios for Cd^{115} to Mo^{99} normalized to the same ratio measured for thermal fission in Pu^{239} , i.e., $(r_{\text{corr}}/r_{\text{th}})_{\text{Cd/Mo}}$, versus neutron energy in the resonance region are presented in Fig. 3.

2.2 Background corrections

The background fissions in each sample were due to neutrons which can be characterized in three groups:

- (a) Scattered neutrons from the transmitted beam striking surrounding materials.
- (b) Fast neutrons from fissions induced in the Pu^{239} by resonance neutrons.
- (c) "Bad" neutrons from pipe walls, etc.

The effect of scattered neutrons which produce a general "fog" is responsible for the principal source of unwanted fissions. These neutrons appear to be in the resonance region. From measurements made on the portions of the wheel which were not exposed directly to any part of the

Sample Number	No ⁹⁹ obs c/m/g	S From middle of to line (cm.)	E _n (ev)	No ⁹⁹ D _{bgd} c/m/g	No ⁹⁹ D _{bgd} Res Part	No ⁹⁹ corr c/m/g	Ag ¹¹¹ obs c/m/g	Ag ¹¹¹ D _{bgd} c/m/g	Ag ¹¹¹ D _{bgd} Res Part	Ag ¹¹¹ corr c/m/g	F _{Ag¹¹¹/No⁹⁹} (units of 10 ⁻²)	Cd ¹¹⁵ obs c/m/g	Cd ¹¹⁵ D _{bgd} c/m/g	Cd ¹¹⁵ D _{bgd} Res Part	Cd ¹¹⁵ corr c/m/g	F _{Cd¹¹⁵/No⁹⁹} (units of 10 ⁻²)	($\frac{r_{corr}}{r_{th}}$) Ag/No	($\frac{r_{corr}}{r_{th}}$) Cd/No
1	20600	12.10	11.88	199	3350	360	16890	1100	66	329	1.95	428	58	13	357	2.11	0.90	1.10
2	23360	11.75	11.75	206	3380	390	19990	1408	66	370	1.89	508	58	15	435	2.22	0.87	1.16
3	23070	11.55	11.55	212	3410	450	19210	1490	67	409	2.14	493	59	17	377	1.96	0.98	1.05
4	27870	11.35	11.35	219	3440	540	23890	2445	67	461	1.93	509	59	20	430	1.800	0.88	1.03
5	32200	11.18	11.18	226	3470	650	28080	2824	68	536	1.916	636	60	24	552	1.886	0.88	1.05
6	40990	11.01	11.01	233	3490	810	36000	36000	69	785	2.017	769	60	30	679	1.736	0.93	0.91
7	46280	10.82	10.82	241	3530	950	42040	847	69	848	2.017	826	61	35	730	1.570	0.90	0.82
8	55740	10.65	10.65	249	3550	1120	51070	947	70	995	1.948	905	61	42	802	1.570	0.90	0.85
9	77880	10.51	10.51	256	3580	1470	72230	1455	71	1455	2.012	1267	62	55	1150	1.592	0.93	0.85
10	109210	10.35	10.35	264	3600	1890	97720	2105	71	2105	2.012	1821	62	70	1689	1.728	0.99	0.90
11	1134590	10.16	10.16	273	3630	2330	118430	2233	72	2233	2.065	2302	62	87	2353	1.857	0.95	0.96
12	116870	9.91	9.91	281	3680	2090	111100	2466	72	2466	2.079	2353	63	78	2212	1.994	0.96	1.04
13	73460	9.75	9.75	288	3700	1470	70310	1580	73	1580	1.869	1159	64	55	1044	1.552	0.86	0.81
14	72330	9.59	9.59	297	3730	1250	60990	1257	73	1257	1.938	999	64	47	828	1.512	0.89	0.71
15	65970	9.42	9.42	307	3760	990	44060	1174	74	1174	1.815	782	65	37	680	1.566	0.85	0.79
16	49710	9.28	9.28	317	3780	850	36740	858	74	858	2.008	682	65	31	586	1.608	0.90	0.84
17	41350	9.06	9.06	328	3820	710	31160	704	75	607	1.949	595	66	26	501	1.706	0.84	0.89
18	39570	8.90	8.90	341	3840	790	34940	740	75	640	1.892	691	66	29	596	1.957	0.86	1.01
19	42140	8.74	8.74	349	3870	840	37430	805	76	703	1.878	823	67	31	725	1.856	0.86	0.97
20	43350	8.61	8.61	355	3920	870	38570	819	76	720	1.867	848	67	32	746	1.884	0.77	0.67
21	47380	8.44	8.44	369	3960	940	42320	823	29	713	1.677	751	68	35	648	1.569	0.84	0.72
22	52230	8.25	8.25	395	3990	950	47330	970	30	865	1.823	791	69	35	699	1.785	0.91	0.95
23	52230	8.01	8.01	412	3990	730	33600	767	77	717	2.156	695	69	27	599	1.660	0.99	0.87
24	57980	7.89	7.89	434	4010	720	33250	818	79	717	2.033	721	69	27	625	1.897	0.93	0.99
25	57980	7.75	7.75	459	4030	720	32950	771	79	670	1.927	596	70	24	502	1.788	0.89	0.94
26	57980	7.52	7.52	496	4070	650	28080	641	80	541	1.970	509	71	21	417	1.767	0.91	0.82
27	57980	7.31	7.31	549	4110	560	23600	563	81	455	2.169	661	72	23	566	2.075	1.00	1.00
28	57980	7.14	7.14	574	4160	590	29230	734	82	634	2.405	608	73	22	513	2.130	1.11	1.08
29	57980	6.97	6.97	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
30	57980	6.80	6.80	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
31	57980	6.63	6.63	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
32	57980	6.46	6.46	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
33	57980	6.29	6.29	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
34	57980	6.12	6.12	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
35	57980	5.95	5.95	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
36	57980	5.78	5.78	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
37	57980	5.61	5.61	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
38	57980	5.44	5.44	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
39	57980	5.27	5.27	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
40	57980	5.10	5.10	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
41	57980	4.93	4.93	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
42	57980	4.76	4.76	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
43	57980	4.59	4.59	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
44	57980	4.42	4.42	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
45	57980	4.25	4.25	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
46	57980	4.08	4.08	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
47	57980	3.91	3.91	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
48	57980	3.74	3.74	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
49	57980	3.57	3.57	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
50	57980	3.40	3.40	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
51	57980	3.23	3.23	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
52	57980	3.06	3.06	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
53	57980	2.89	2.89	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
54	57980	2.72	2.72	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
55	57980	2.55	2.55	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
56	57980	2.38	2.38	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
57	57980	2.21	2.21	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
58	57980	2.04	2.04	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
59	57980	1.87	1.87	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
60	57980	1.70	1.70	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
61	57980	1.53	1.53	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
62	57980	1.36	1.36	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
63	57980	1.19	1.19	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
64	57980	1.02	1.02	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
65	57980	0.85	0.85	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
66	57980	0.68	0.68	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
67	57980	0.51	0.51	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
68	57980	0.34	0.34	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
69	57980	0.17	0.17	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
70	57980	0.00	0.00	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
71	57980	0.00	0.00	574	4160	590	29230	646	82	546	2.405	162	73	6	62	3.182	1.24	1.11
72	57980	0.00	0.00	574	4160	590	29230	646	82	546	2.405	162	73	6

Fig. 1. "General" background correction curve.

Fig. 2. Plot of fraction of Pu^{239} converted by fission in each sample vs. neutron energy. The fission excitation function is shown for comparison. Width of individual sample cut is shown by width of horizontal bars in the bar graph.

Fig. 3. The ratio of Cd^{115} activity to Mo^{99} activity, normalized to the same ratio for thermal fission in Pu^{239} , is plotted versus neutron energy. The horizontal bar indicates width of sample cut and the vertical bar indicates the estimated standard error in the measurement of the activity ratio.

neutron beam, a curve of "general" background versus position on the wheel was constructed (Fig. 1) and a correction was made for scattered neutrons originating chiefly in the steel wheel and adjacent hardware. The correction was made iteratively since it was necessary to estimate average yields for Ag^{111} and Cd^{115} for fission in the resonance energy region and to assume that resonance neutrons scattered back to the wheel would still be in the resonance region. Since the correction is no more than 10% of total activity observed in regions of greatest interest and a corresponding correction is made to observed Mo^{99} activity, the $\text{Ag}^{111}/\text{Mo}^{99}$ and $\text{Cd}^{115}/\text{Mo}^{99}$ corrected ratios are not very sensitive to the exact Ag^{111} and Cd^{115} yields chosen for background fission. The relative yields in the resonance region used for the correction were 0.9 of the Pu^{239} thermal fission values. The yield values used in the keV region were somewhat higher and are indicated in Fig. 2.

The effect of fast neutrons (for which the appropriate $(r_{\text{fast}}/r_{\text{th}})_{\text{Ag/Mo}}$ is taken as 1.43 and the appropriate $(r_{\text{fast}}/r_{\text{th}})_{\text{Cd/Mo}}$ is taken as 1.95) has been estimated by means of a Monte Carlo calculation. The calculation indicated the following correction applies:

$$F_{\text{sec}} = 1.52 \times 10^{-2} F_{\text{P1}} + 8.2 \times 10^{-4} (F_{\text{P2L}} + F_{\text{P2R}}) + 4.6 \times 10^{-4} (F_{\text{P3L}} + F_{\text{P3R}}) \\ + 3.2 \times 10^{-4} (F_{\text{P4L}} + F_{\text{P4R}}) + 2 \times 10^{-4} (F_{\text{P5L}} + F_{\text{P5R}})$$

where F_{P1} indicates resonance fissions in the sample itself, F_{P2L} and F_{P2R} indicate resonance fission on the immediately adjoining sample on the left and right, respectively; F_{P3L} and F_{P3R} indicate the next adjoining samples, etc.

The effect of non-collimated neutrons scattered from the walls of the pipe was minimized by baffles along the walls and no correction has been applied.

2.3 Ba^{140} data

In addition to the tabulated data, Ba^{140} analyses on all samples showed an average activity ratio to Mo^{99} of 0.365 in the resonance region compared to a thermal ratio of 0.373, with a spread no greater than the analytical

error. Based on this observation, the assumption is made that Mo^{99} and Ba^{140} yields are the same as in thermal fission; if their yields varied, it would be necessary to assume that they varied the same way and by the same amount which seems unlikely.

3. Discussion

Measured by the changes in the $\text{Cd}^{115}/\text{Mo}^{99}$ activity ratio, the symmetric yield decreased by 18% at 250 ± 4 ev, 29% at 307 ± 5 ev, and 33% at 395 ± 6 ev. In the energy region 200-600 ev the symmetric yield is significantly greater than the thermal yield only at 206 ev (16%).

The Ag^{111} yield over the resonance region is more nearly constant than is the Cd^{115} yield. In the region of minimum Cd^{115} yield the yield of Ag^{111} is also lower than its average resonance value by an amount which approximates one-third of the effect on Cd^{115} .

The $\text{Cd}^{115}/\text{Mo}^{99}$ activity ratio, averaged over all samples in the resonance region, is 0.92 of the Pu^{239} thermal fission ratio. The $\text{Ag}^{111}/\text{Mo}^{99}$ activity ratio, similarly averaged, is 0.91 of the thermal fission ratio.

If it is assumed that variations in the Cd^{115} yield are due to variations in the proportions in each sample of fission produced at two different kinds of resonances, attempts can be made to estimate the relative proportions of each of the two kinds of levels. One must further assume that each of the two kinds of levels has a characteristic value for $r_{\text{Cd}/\text{Mo}}$ which will be designated r_{min} and r_{max} , respectively. The value of $r_{\text{min}}/r_{\text{th}}$ may be as low as 0.2 at levels for which symmetric fission is forbidden [3] and, based on an extrapolation of the highest observed values, perhaps as high as 1.6 of the thermal value for levels at which symmetric fission is allowed. The fraction of fission, X , which occurs in the less symmetric mode in any sample will be given by

$$X = \frac{1 - (r_{\text{obs}}/r_{\text{max}})}{1 - (r_{\text{min}}/r_{\text{max}})}$$

If r_{max} and r_{min} are as assumed, X will have a maximum value of 0.67 at

395 ev in a cut which contains about eight levels and a minimum value of 0.32 at 206 ev in a cut which contains about three levels. The value of X averaged over the entire region from 200 to 600 ev is 0.5. Assumptions on $r_{\max}$ and $r_{\min}$ which result in values of X which are as large as 0.6 or as small as 0.4 are less consistent with observations of the average yield values in each cut. Therefore, it is reasonable to assume that these measurements indicate that the population of levels with the less symmetric mode of fission is about the same as the population with the more symmetric mode.

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