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MEASUREMENTS OF GAMMA-RAY MULTIPLICITY SPECTRA AND THE ALPHA VALUE FOR ^{239}Pu IN GROUPS AND RESOLVED RESONANCES

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Introduction

The measurements of the multiplicity of gamma-quanta spectra for ^{239}Pu were performed in the resonance energy region by G.V.Muradyan et al., [1,2] to determine the value of $\alpha=\sigma_\gamma/\sigma_f$. These measurements have shown that the use of the multiplicity radiation spectrometry method results in lower methodical errors connected with the background and normalization to the reference values. Undoubtedly, investigations with multiplicity spectrometry in the region of resolved resonances for fissionable nuclei are of interest not only for the precision of the above-mentioned constants, but also for studying other characteristics of separate resonances. The positive peculiarity of this method is the possibility of simultaneously registering several processes of nuclei excitation in one experiment, which can be resolved. However, the method of radiation multiplicity spectrometry has not received wide acceptance because of the complexity and high cost of the equipment. This work is an attempt to apply radiation multiplicity spectrometry to ^{239}Pu nuclei excited by resonance neutrons to investigate the shape of γ-ray multiplicity spectra in fission and radiative capture, and also to improve the precision of some constants used in reactor calculations.

Experimental technique

The measurements of the ^{239}Pu multiplicity spectra were carried out at the IBR-30 pulsed neutron booster of the Joint Institute for Nuclear Research (JINR) in Dubna (Russia) on the 500 m flight path with a 16-section scintillation detector, consisting of NaJ(Tl) crystals, with a total volume of 36 litres [3]. Measurements with the IBR-30 pulsed neutron booster were made at a mean thermal power of 10 kW, neutron pulse repetition rate of 100 Hz; the pulse duration at a half-height uses 4 μsec. Metal discs 80 mm in diameter and 0.3 mm thick containing 22.7875 g of 99.9% ^{239}Pu were used as the sample-radiator. The samples were packed in stainless steel containers made of foil 0.12 mm thick. The samples were positioned at the center of the detector in a vacuum tube.

To remove the background of recycled neutrons, a B_4C (10 mm) filter was used at the beam. For determining the background components in the time-of-flight spectra, resonance filters of Al, Mn and Co were installed in the beam. For to reduce the background from fission neutrons and to register the process of neutron scattering by the sample, cylindrical converters of 35 mm thick borated polyethylene with 3.5% B_4C and 10 mm thick ^{10}B were placed

inside the detector. The discrimination threshold in each detector channel corresponded to an energy release of 100 keV. Beyond the linear signals summator from all 16 channels, the discrimination window separated pulses corresponding to the energy release range of 2-10 MeV. The discrimination window for neutrons scattered by the sample was chosen to be equal to 400-600 keV. The information in the form of 16 time-of-flight spectra, covering the neutron energy range of 15 eV - 10 MeV, was accumulated in the intermediate memory of the measuring module with a PC/AT-286 VANG. The first 12 spectra corresponded to a coincidence multiplicity from 1 to 12 for radiative capture and fission, spectra 13 and 14 contained information about the neutron scattering by the sample in the first and the second halves of the detector, spectra 15 and 16 were used for other purposes. Typical time-of-flight spectra of different multiplicities after the subtracting the backgrounds are presented in Figs.1-2.

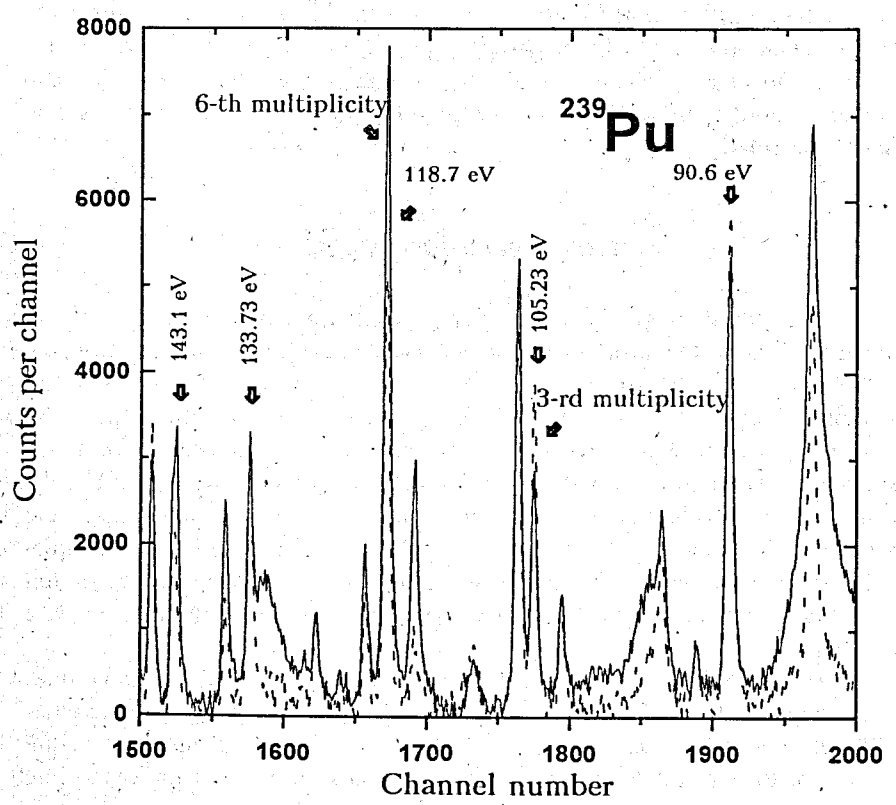

Fig.1 Time-of-flight spectra of gamma-ray coincidence of the 3rd and 6th multiplicities for ^{239}Pu. (The width of time channel -2 μsec)

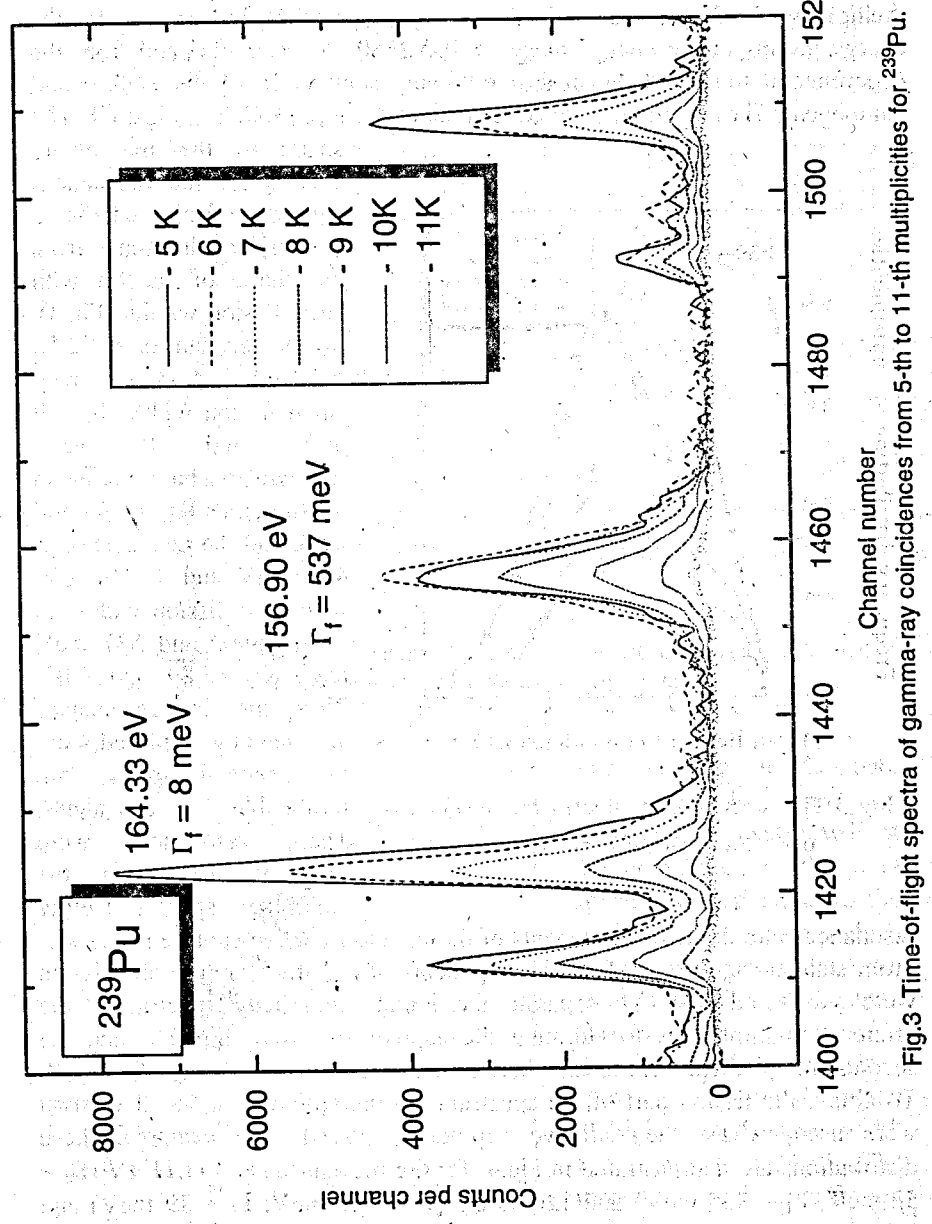

Fig.3 Time-of-flight spectra of gamma-ray coincidences from 5-th to 11-th multiplicities for ^{239}Pu.

Results of measurements

The coincidence multiplicity spectra from the first to the eleventh multiplicity for 72 resonances in the energy range of 21-313 eV and for the energy groups in the energy range of 100-2150 eV were obtained from the experimental time-of-flight spectra following subtraction of the background component. They were normalized to unity and are presented in Figs.3-4. The shape of the multiplicity spectra for the resonances with small fission widths is essentially different from the shape of spectra with large fission widths (Fig.3). So the resonances at 52.52 eV and 164.33 eV with small fission widths Γ_f = 9 meV and 8 meV, respectively, have the mean multiplicities K_m = 4.69 and 4.78, and the resonances, at 47.50 eV and 156.90 eV, with large fission widths Γ_f = 248 meV and 537 meV have K_m =5.89 and 6.07. Thus, the first resonances are actually captured, and the second ones are fissionable resonances. These resonances were used to separating the multiplicity spectra of other resonances into the two components of fission and radiative capture processes. From standard spectra, the small components of radiative capture and fission were subtracted first. To separate the initial multiplicity spectra, as the reference multiplicities for forming the captive spectrum, the first and the second multiplicities were used, where fission does not actually manifest itself. To obtain the fission part of the spectrum the multiplicities higher than sixth were used, where the radiative capture is absent. For example, these distributions are demonstrated in Fig.4 for the resonances at 195.11 eV (Γ_γ = 52 meV, Γ_f = 335 meV) and 120.88 eV (Γ_γ = 32 meV, Γ_f = 39 meV) and 269.51eV (Γ_γ = 40 meV, Γ_f = 28 meV).

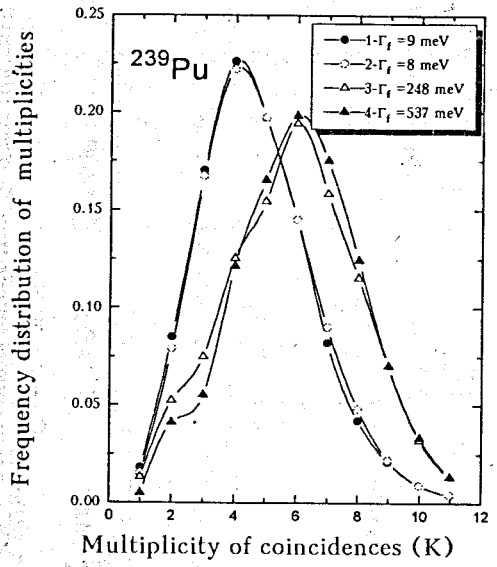

Fig.3 The multiplicity spectra for resonances of ^{239}Pu with different fission widths.

One may consider the change in the shape of the total multiplicity spectra as a function of the fission width of the resonances. The average multiplicity of the total spectra as a function of Γ_f is shown in Fig.5.

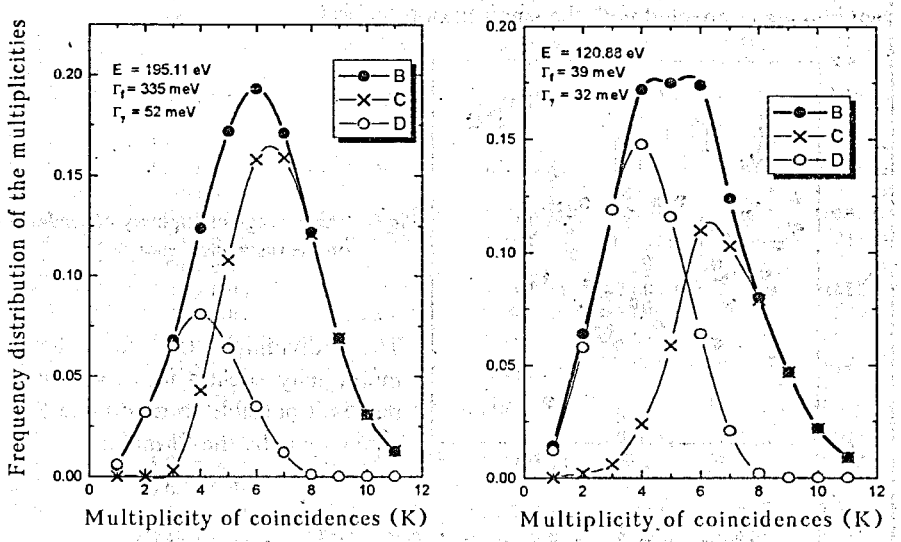

Fig.4 Examples of the multiplicity distribution spectra for different resonances (B- total spectrum, C- fission spectrum, D - radiative capture spectrum)

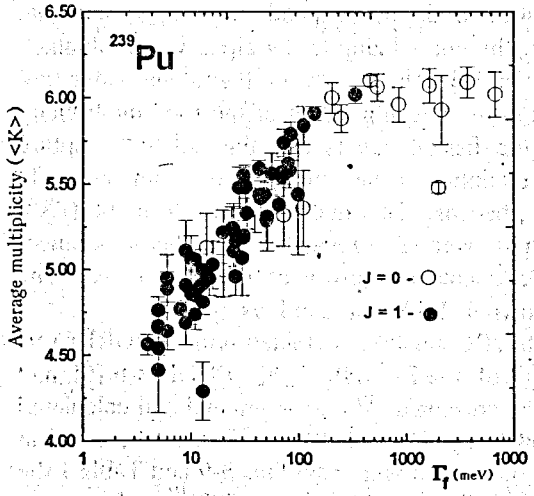

Fig.5 The dependence of the average multiplicity in resonances on their fission width.

The increase in K_m with increasing Γ_f can be explained by the growth of the component in the total spectra corresponding to the fission process, in which more gamma-quanta are emitted in comparison with the radiative capture. The dependence of the resonance spins of ^{239}Pu on the mean multiplicity and fission width Γ_f is shown in Fig.5. The resonances with large fission widths and large mean

multiplicities usually have the spin J = 0. One can see this effect in Figs.6 and 7. The energy dependence of the average multiplicity of the total spectra for 72 resonances of ^{239}Pu is presented in Fig.7. Figure 7 demonstrates the periodic functional dependence of K_m and the alpha value on the energy, and whose maxima are connected with the small fission width Γ_f.

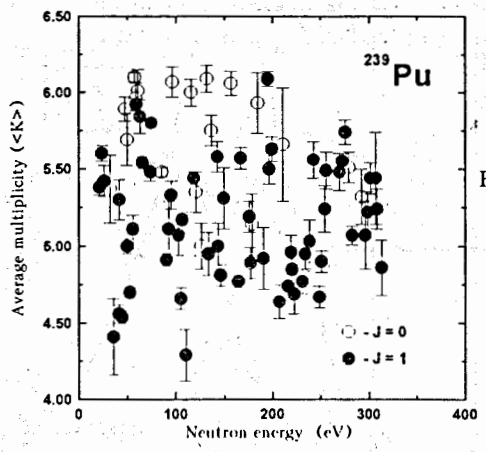

Fig.6 The average multiplicity dependence on the resonance energy.

The dividing of the total multiplicity spectra into two parts makes it possible to determine the alpha value by the formula:

$$\alpha = \sigma_\gamma/\sigma_f = N_\gamma\,\varepsilon_f / N_f\,\varepsilon_\gamma = A\,\Sigma K_{i\gamma}\,/\Sigma K_{if} \qquad (1)$$

where σ_γ, σ_f are the radiative capture and fission cross-sections; N_γ, N_f are the total number of counts of capture and fission gamma-quanta and $K_{i\gamma}$, K_{if} are the *i*-st multiplicity radiative capture and fission. Assuming that the efficiencies $(\varepsilon_\gamma, \varepsilon_f)$ of gamma-quantum registration do not depend on energy, one can determine the value $A = \varphi\,(\varepsilon_f\,\varepsilon_\gamma)$ by normalizing to the alpha value, obtained in the precision measurements in [2]. The errors in the alpha value are determined mainly by the errors in the decomposition of the total multiplicity spectra into components from the fission events and the radiative capture events. The errors from the uncertainties in the efficiency of gamma-quanta registration are considerably less, because the value A is close to unity. (As a result of the normalization of alpha over 13 resonances from this experiment and from [2], the ratio of the efficiencies is given as equal to unity). The experimental alpha values are shown in Table 1 and in Figs.7-9.

For comparison, the results of evaluations obtained with the GRUCON computer program [4] on the basis of the BROND-2 [5], ENDF/B-6 [6], and JENDL-3 [7] library data are also presented. The experimental and calculated alpha values were obtained in the energy intervals below 72 resonances and in the energy groups (see Table 2). As can be seen from Figs.8-9 and Table 1 the calculated alpha values of different libraries differ by 5-30%, and in some resonances by 50-100%. The experimental alpha values differ from the

Table 1.

E(eV)	J	K(m)	$E_1 - E_2$ (eV)	α (exp)	α ENDF/B-6	α JENDL-3	α BROND-2
1	2	3	4	5	6	7	8
21.09	1	5.38 ± 0.05	20.80-21.29	0.87±0.06	1.234	1.116	0.516
23.85	1	5.60 ± 0.05	23.52-24.16	0.70±0.06	1.090	1.037	0.643
26.22	1	5.42 ± 0.10	25.82-26.67	0.97±0.10	1.003	0.945	0.770
27.27	-	4.71 ± 0.23	-	-	-	-	-
32.27	0	5.37 ± 0.22	31.77-32.82	0.43±0.11	0.348	0.348	0.320
34.02	-	-	-	-	-	-	-
35.42	1	4.41 ± 0.25	35.12-35.80	-	-	-	-
41.38	1	4.57 ± 0.07	40.88-41.32	4.35±0.20	11.378	11.013	4.611
41.6	1	5.30 ± 0.13	-	-	-	-	-
44.42	1	4.57 ± 0.04	43.93-44.92	5.13±0.38	7.495	7.259	6.387
47.56	0	5.89 ± 0.09	46.98-48.07	0.23±0.06	0.161	0.160	0.244
49.70	0	5.69 ± 0.17	-	-	-	-	-
50.01	1	5.00 ± 0.05	49.93-50.44	2.19±0.11	2.503	2.421	1.881
52.52	1	4.69 ± 0.04	51.82-53.25	3.57±0.12	3.896	3.775	3.285
55.60	1	5.11 ± 0.09	55.01-55.92	1.35±0.08	1.065	1.014	0.815
57.48	0	6.11 ± 0.04	56.39-58.16	0.18±0.06	0.088	0.087	0.099
58.76	0	5.96 ± 0.11	-	-	-	-	-
59.16	1	5.92 ± 0.05	58.96-59.46	0.30±0.07	0.282	0.254	0.313
60.94	0	6.02 ± 0.14	60.59-61.60	0.15±0.10	0.033	0.031	0.043
63.07	1	5.85 ± 0.11	62.46-63.57	0.32±0.08	0.182	0.167	0.172
65.35	1	5.62 ± 0.09	-	-	-	-	-
65.70	1	5.54 ± 0.04	65.64-66.47	0.65±0.12	0.440	0.409	0.587
74.01	1	5.49 ± 0.06	73.31-74.08	0.98±0.09	0.437	0.409	0.721
74.84	1	5.80 ± 0.03	74.58-76.10	0.41±0.10	0.437	0.403	0.426
78.95	-	-	-	-	-	-	-
80.15	-	-	-	-	-	-	-
81.35	-	-	-	-	-	-	-
82.58	-	-	-	-	-	-	-
83.59	-	-	-	-	-	-	-
84.44	-	-	-	-	-	-	-
85.39	0	5.48 ± 0.04	85.13-85.74	0.76±0.12	0.926	0.899	0.867
86.27	1	-	-	-	-	-	-
90.60	1	4.91 ± 0.04	89.37-91.75	2.42±0.09	1.611	1.554	1.556
92.83	1	5.11 ± 0.18	92.24-93.43	1.30±0.12	1.260	1.220	2.203
95.36	1	5.34 ± 0.09	94.33-95.36	0.87±0.12	0.335	0.322	0.538
96.50	0	6.07 ± 0.11	96.60-97.88	0.18±0.07	0.035	0.034	0.033
98.63	-	-	-	-	-	-	-
100.06	-	-	-	-	-	-	-
101.96	-	-	-	-	-	-	-
102.88	1	5.07 ± 0.13	102.31-103.46	1.27±0.18	1.383	1.331	1.393
105.23	1	4.67 ± 0.07	104.28-105.47	3.54±0.24	2.423	2.338	2.456
106.68	1	5.17 ± 0.05	106.07-107.54	1.54±0.13	1.401	1.338	1.196
110.43	1	4.29 ± 0.17	109.67-111.34	6.24±0.43*	1.164	1.119	0.764
114.80	-	-	-	-	-	-	-
115.21	-	-	-	-	-	-	-
115.89	0	6.00 ± 0.09	115.48-116.73	0.29±0.08	0.177	0.172	0.186
118.70	1	5.44 ± 0.04	117.29-119.71	0.94±0.11	0.616	0.577	0.784
120.88	0	5.36 ± 0.16	120.44-121.62	0.84±0.13	0.908	0.857	0.670
123.42	1	-	-	-	-	-	-
126.20	0	5.01 ± 0.16	125.26-126.67	1.46±0.35	1.292	1.237	1.456
127.46	1	-	-	-	-	-	-
131.87	0	6.09 ± 0.10	129.56-131.87	0.20±0.08	0.017	0.017	0.018

1	2	3	4	5	6	7	8
133.73	1	4.96 ± 0.14	133.73-134.42	2.28±0.14	2.263	2.192	1.578
136.69	0	5.76 ± 0.11	135.64-137.58	0.39±0.07	0.251	0.250	0.230
139.20	-	-	-	-	-	-	-
142.91	1	5.59 ± 0.10	141.78-142.91	0.70±0.10	0.308	0.289	0.525
143.48	1	5.20 ± 0.16	143.48-144.24	1.05±0.08	1.125	1.064	1.044
146.18	1	4.81 ± 0.08	144.62-146.96	2.61±0.16	2.281	2.178	1.913
147.55	-	-	-	-	-	-	-
148.35	-	-	-	-	-	-	-
149.36	1	5.31 ± 0.21	148.95-150.17	0.94±0.12	0.851	0.808	0.828
155.60	-	-	-	-	-	-	-
156.90	0	6.10 ± 0.09	156.90-158.21	0.23±0.07	0.169	0.165	0.089
160.80	-	-	-	-	-	-	-
161.80	-	-	-	-	-	-	-
164.33	1	4.78 ± 0.04	163.40-165.03	3.40±0.19	4.506	4.363	3.776
166.92	1	5.58 ± 0.08	165.97-167.40	0.76±0.09	0.707	0.667	0.711
170.32	-	5.55 ± 0.17	168.85-171.06	0.66±0.10	0.433	0.408	0.262
171.06	-	-	-	-	-	-	-
174.58	-	-	-	-	-	-	-
175.86	1	5.19 ± 0.10	175.34-175.86	1.22±0.13	0.972	0.928	1.112
177.16	1	489 ± 0.10	176.64-177.94	2.58±0.19	3.499	3.379	3.331
178.73	0	5.14 ±	178.20-179.53	1.63±0.12	2.151	2.075	1.979
183.59	-	5.49 ± 0.12	181.68-183.59	0.66±0.11	0.222	0.213	0.292
184.69	0	5.94 ± 0.20	184.97-186.09	0.25±0.06	0.029	0.028	0.029
188.27	-	5.03 ± 0.42	-	-	-	-	-
190.67	1	4.92 ± 0.22	189.51-191.25	2.09±0.14	1.508	1.451	1.318
195.11	1	6.03 ± 0.06	193.02-195.72	0.25±0.07	0.109	0.108	0.144
196.63	1	5.50 ± 0.14	-	-	-	-	-
199.39	1	5.63 ± 0.08	197.55-200.34	0.60±0.10	0.449	0.420	0.395
203.20		5.97 ± 0.06	201.29-203.84	0.29±0.07	0.152	0.148	0.183
203.84	-	-	-	-	-	-	-
205.13	0	-	-	-	-	-	-
207.10	1	4.64 ± 0.12	206.44-208.09	3.84±0.35	3.491	3.370	2.639
211.09	0	5.66 ± 0.37	-	-	-	-	-
212.02	0	-	-	-	-	-	-
213.51	-	-	-	-	-	-	-
216.30	1	4.74 ± 0.09	215.60-217.00	3.30±0.34	1.893	1.831	2.236
219.50	1	4.96 ± 0.11	218.43-219.14	1.97±0.18	0.793	0.762	0.895
220.22	1	4.85 ± 0.14	220.22-220.95	2.31±0.18	2.474	2.382	1.865
222.78	1	4.69 ± 0.13	222.04-223.14	2.79±0.26	1.726	1.672	0.972
225.00	-	5.27 ± 0.20	-	-	-	-	-
226.50	0	-	-	-	-	-	-
227.64	-	-	-	-	-	-	-
231.10	1	4.77 ± 0.08	229.93-231.88	2.72±0.20	2.296	2.221	2.055
232.66	-	-	-	-	-	-	-
234.24	1	4.95 ± 0.11	233.45-235.43	1.97±0.16	1.854	1.792	1.952
238.66	1	5.03 ± 0.15	237.44-239.48	1.63±0.15	1.034	0.999	1.136
240.60	-	-	-	-	-	-	-
242.80	1	5.56 ± 0.13	241.54-244.06	0.69±0.08	0.365	0.341	0.360
247.50	1	-	-	-	-	-	-
248.76	1	4.67 ± 0.08	247.90-249.20	3.31±0.24	2.106	2.038	2.206
250.95	1	4.90 ± 0.07	250.51-252.27	2.43±0.14	2.301	2.221	2.228
254.51	1	5.25 ± 0.15	-	-	-	-	-
255.86	1	5.49 ± 0.12	255.41-257.22	0.83±0.09	0.948	0.915	0.591

1	2	3	4	5	6	7	8
258.60	-	5.95 ± 0.19	-	-	-	-	-
261.00	0	-	-	-	-	-	-
262.32	-	-	-	-	-	-	-
264.21	-	-	-	-	-	-	-
268.05	-	-	-	-	-	-	-
269.51	1	5.49 ± 0.13	268.05-269.51	0.79±0.10	0.222	0.207	0.221
272.48	1	5.56 ± 0.06	270.99-272.97	0.66±0.10	0.615	0.574	0.622
274.98	-	5.88 ± 0.07	273.98-274.98	0.35±0.09	0.255	0.242	0.191
275.49	1	5.74 ± 0.08	275.49-276.50	0.48±0.12	0.362	0.341	0.342
278.03	0	-	-	-	-	-	-
279.06	0	5.51 ± 0.10	278.55-280.10	0.71±0.09	0.279	0.263	0.215
282.71	1	5.07 ± 0.06	280.62-283.76	1.95±0.16	1.737	1.672	1.533
285.89	-	-	-	-	-	-	-
288.04	-	-	-	-	-	-	-
289.70	-	-	-	-	-	-	-
292.42	0	5.32 ± 0.19	291.32-292.97	0.96±0.11	0.225	0.216	0.153
296.33	1	5.07 ± 0.23	293.53-296.33	1.58±0.11	0.569	0.547	0.497
298.60	1	5.22 ± 0.13	297.46-299.17	1.39±0.13	1.546	1.483	1.398
301.48	1	5.45 ± 0.10	299.75-302.64	1.12±0.12	0.976	0.928	1.144
307.35	1	5.44 ± 0.35	-	-	-	-	-
308.55	1	5.24 ± 0.13	307.95-309.75	1.39±0.14	0.829	0.787	1.265
311.12	-	-	-	-	-	-	-
313.40	1	4.86 ± 0.18	312.80-314.64	2.56±0.29	1.472	1.422	1.264

* Ga isotope

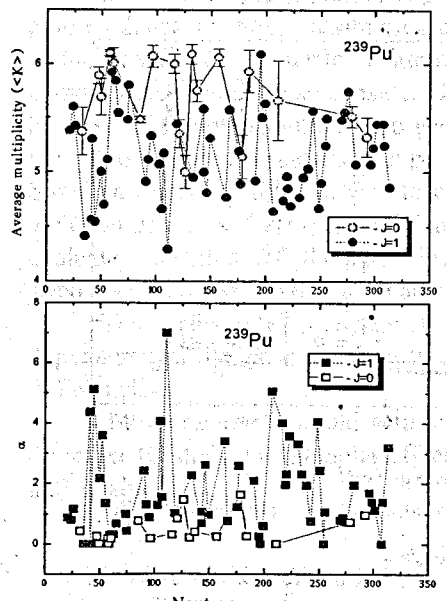

Fig.7 The average multiplicity and the alpha value dependence on the resonance energy.

calculated ones most significantly for small and large alpha values, when the contribution of the radiative capture or fission in the total multiplicity spectrum is small. The uncertainty increases greatly when the decomposing the spectrum into its component parts according to accepted standard spectra. These uncertainties decrease when determining the experimental and calculated alpha values in the wide energy groups (Table 1) and using of a large number of resonances. The experimental errors of the alpha values in some resonances amount to 2-12%, for large alpha values; for small alpha values, the errors may run to 60%.

In the energy groups, the errors are significantly less than 1-5%, but when compared with the experimental data of other papers, gives a

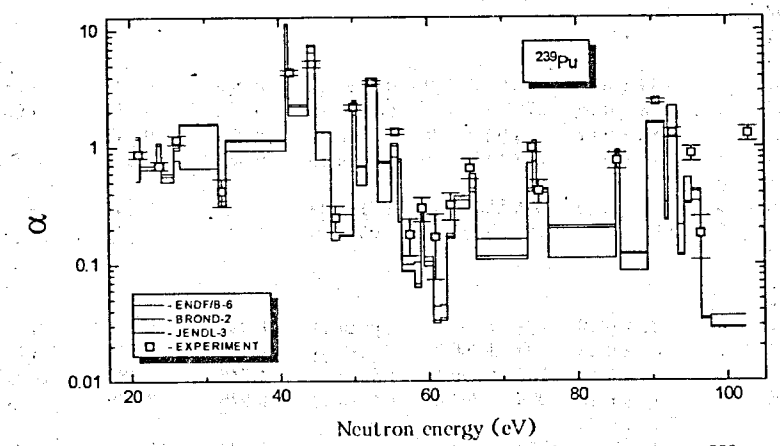

Fig.8 Experimental and calculated alpha values $\alpha=\sigma_\gamma/\sigma_f$ for ^{239}Pu.

discrepancy of more than 100% (see Table 1). The greatest disagreement are with the alpha values in the experimental data by Yu.V.Ryabov et al., [8] in the

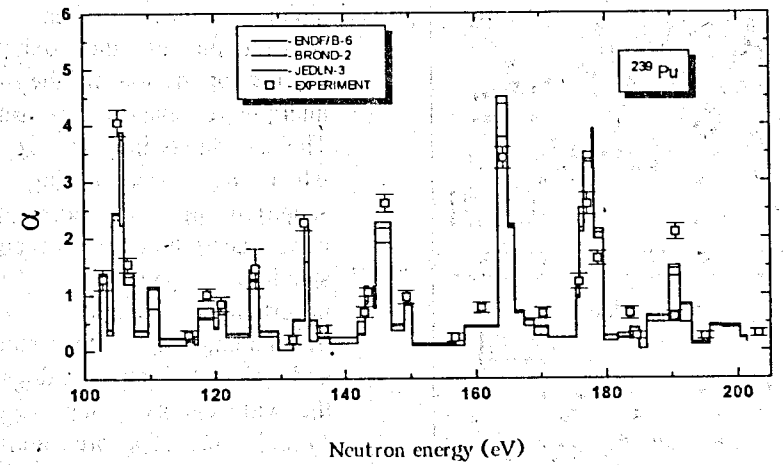

Fig.9 Experimental and calculated alpha values $\alpha=\sigma_\gamma/\sigma_f$ for ^{239} Pu.

0.8-2.0 keV groups and V.N.Kononov et al., [9] in the 0.2-0.3 keV, 0.6-0.7 keV, and 0.9-1.0 keV groups. The present work has a significant discrepancy, of about 30%, with the data by other authors, with the exception of J.A.Farrell

et al. [10], V.P.Bolotsky et al. [11] and G.V.Muradyan et al. [12] in the 0.4-0.5 keV group.

Table 2.

E (keV)	Gwin (1976)	Farrell (1970)	Derrien (1989)	Ryabov (1976)	Bolotsky (1977)	Kononov (1971)	Muradyan (1986)	<α> (*)	Present work
1	2	3	4	5	6	7	8	9	10
0.1-0.2	0.87	0.67	0.868	0.85	0.93	0.71	0.94	0.83	0.83
0.2-0.3	0.94	0.67	0.925	1.00	0.92	1.31	1.01	0.97	0.92
0.3-0.4	1.16	0.94	1.194	1.00	1.17	1.71	1.31	1.21	-
0.4-0.5	0.44	0.57	0.414	0.88	0.59	0.48	0.64	0.57	0.63
0.5-0.6	0.72	0.64	0.714	0.84	0.75	0.68	0.93	0.75	0.69
0.6-0.7	1.54	1.68	1.516	1.41	1.46	0.71		1.37	1.63
							1.25**		
0.7-0.8	0.97	0.85	0.904	1.31	1.00	1.03		1.04	1.24
0.8-0.9	0.82	0.79	0.746	1.15	0.78	0.68		0.83	0.72
							0.87**		
0.9-1.0	0.70	0.70	0.615	1.21	0.76	0.48		0.76	0.72
1.0-2.0	0.84	1.17	-	1.04	0.85	0.65	1.06	0.93	0.71

(*) - α average value calculated over data from columns 2 to 8

(**) - data covering two neighbouring energy ranges

In this way, the extensive experimental materials presented in this work give evidence of great possibilities of the radiation multiplicity spectrometry method for fissionable nuclei. Determination of the alpha value by this method is more precise than those considered in [1,2] and that have been used thus far because of better resolution. The general view of the obtained results shows the good agreement between experimental data and the evaluated values. Simultaneously, it might be well to point out the significant differences in the alpha values from the experiments and calculations made on the basis of the evaluated constants of the last library versions of BROND-2, ENDF/B-6, JENDL-3 for some isolated resonances (Table 1).

The results of the present work also verify the assumption about the spin dependence of the mean multiplicity of radiation for resolved resonances in the ^{239}Pu nucleus.

In the future, it would be advisable to continue the investigation in this direction to study the Doppler-effect and the resonance self-shelding in the alpha value.

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