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Some Highlights in the Evaluations of the Thermal Cross Sections and Resonance Parameters of the Actinides*

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ABSTRACT

The resonance parameters and thermal cross sections of ^{235,238}U and $^{239-242}$Pu are reevaluated by considering the measurements carried out since 1973. Capture, scattering, fission cross sections as well as resonance integrals are calculated from the parameters and are compared with experimental values with the objective of achieving consistency between calculations and measurements. The Dyson-Mehta Δ_3 statistical analysis was applied in order to calculate average level spacings. Calculations of average radiative widths based on systematics are carried out and are compared with experimental values as well as with Moore's and Lynn's estimates.

1. INTRODUCTION

An accurate knowledge of the individual as well as average resonance parameters and thermal neutron cross sections of the actinides is important in the design of thermal and fast reactors, in doppler coefficient studies, and optical model and systematic investigations. Since 1973, several data sets became available in the open literature and via private communications which warranted a new reevaluation of these parameters. The final results will be published shortly.[1]

2. THE EVALUATION PROCEDURE

We briefly describe the steps adopted in the evaluation of the resonance parameters. The evaluation of the thermal cross sections was discussed previously in detail.[2]

(1) The first step in the evaluation of the resonance parameters is compilation of the data. A complete and correct documented data base is imperative. The National Nuclear Data Center CSISRS Library was used for this purpose. A computerized resonance parameter file supplemented by the most recent data which was obtained via private communications was created.

(2) The second step is a reduction of the various data sets to a standard form such as $g\Gamma_n$ values.

(3) This is followed by grouping of the various data sets according to resonance energy and $g\Gamma_n$ values, taking the weighted averages, and calculating the internal and external errors for the parameters of each resonance.

(4) Examination of the results of the previous step and making necessary adjustments.

(5) The computerized recommended resonance parameter file is subsequently transformed into an ENDF-type format. Then physics checking computer codes partially based on ENDF codes operate on this file to calculate capture, fission, scattering (coherent and incoherent amplitudes) cross sections as well as resonance integrals and strength functions.

(6) In addition, staircase plots of the cumulative number of resonances and reduced neutrons widths are produced. Values of the Δ_3 statistics are calculated and are compared with experimental values to check on the possible missing or misassigning of the ℓ and J values of resonances.

(7) Consistentcy checks between differential and integral measurements are made. Possible adjustments in the parameters of the low-lying resonances are made to achieve this objective and/or negative energy resonances are postulated.

It may be necessary that several iterations of these steps are required before a satisfactory recommended set of resonance parameters is obtained.

To estimate reliable average level spacings tests other than the Porter-Thomas distribution are required for estimating whether any levels have been missed. The Dyson-Mehta Δ_3 statistics[3] can provide an independent test.

Both Dyson's ensemble theory and Wigner's random matrix theory[4] predict high correlation of single population of levels resulting in approximately equal spacings. This means that the levels are highly ordered rather than randomly distributed.

The Δ_3 statistics is sensitive to the position of individual levels and is given by

$$\Delta_3 = \min_{A,B} \frac{1}{\Delta E} \int_0^{\Delta E} [N(E) - AE-B]^2 \, dE$$

$$\langle\Delta_3\rangle = \frac{1}{\pi^2} [\ln(N) - 0.0687]$$

$$\text{Error } (\langle\Delta_3\rangle) = 0.11$$

Based on Monte Carlo calculations Georgopulos and Camarda[5] have provided graphs which help in determining the number of missed levels by making use of the Δ_3 statistics calculated for the observed single population sequence. The average level spacings were evaluated by applying the Δ_3 statistics in combination with staircase plots of the cumulative number of resonances versus neutron energy as shown in Fig. 1.

A brief description of the results of the evaluation follows.

3. RESULTS

^{235}U:

^{235}U is an important material in connection with power reactors and related applications. The need for an evaluated set of resonance parameters for this material need not be emphasized.

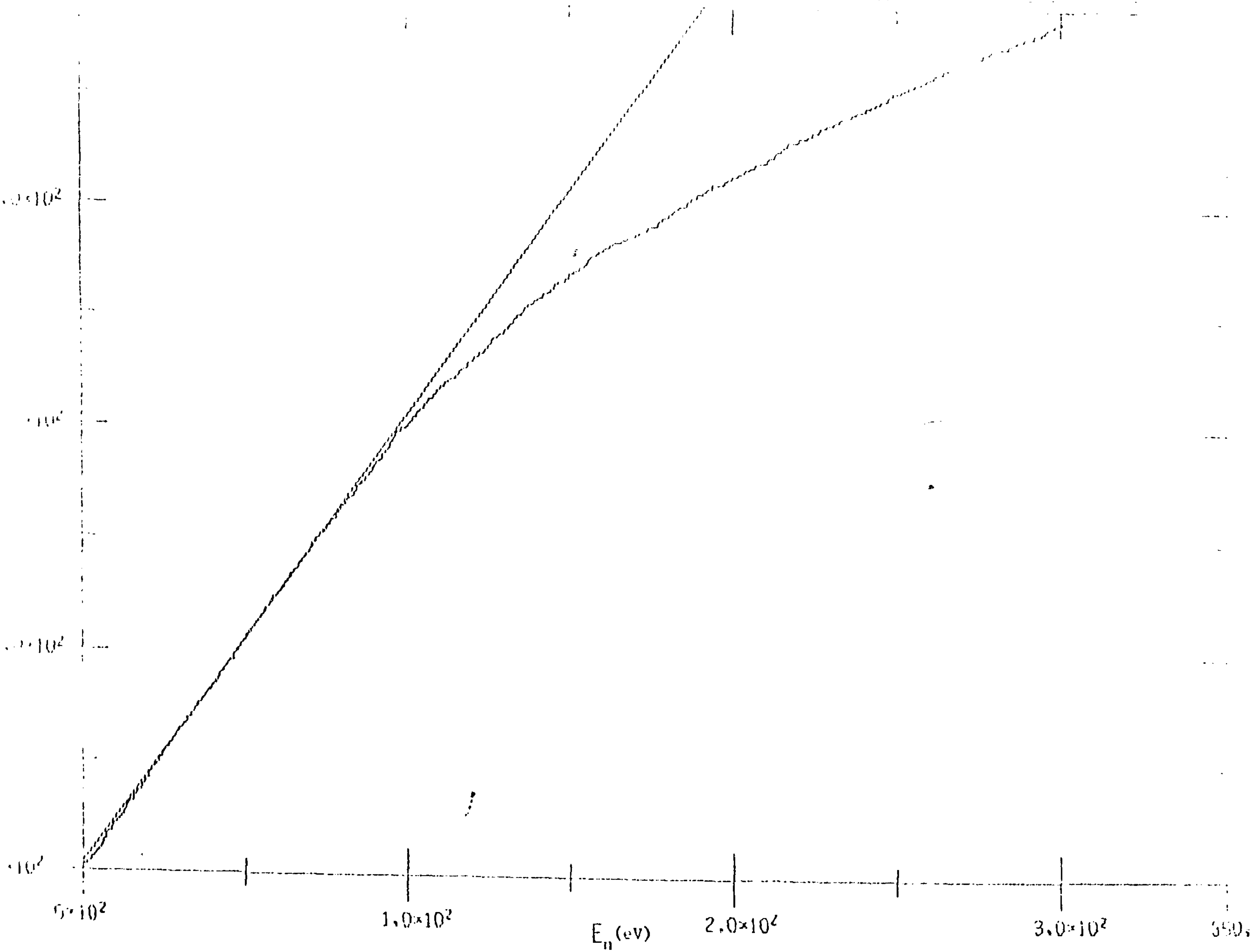

Fig. 1. Staircase plot of ^{235}U resonances. The average spacing D_0 obtained from the straight line least squares fit yields 0.496 eV is consistent with the value of 0.49 derived from Δ_3 statistics.

Some of the problems associated with the ^{235}U resonance parameters have been discussed by Keyworth and Moore[6] and Moore et al.[7] Since 1973, no new measurements of the widths of resonances of ^{235}U have been made.

In addition to Keyworth's[8] polarization measurements Michaudon et al.,[9] and Blons et al.,[10] reported total and fission cross-section measurements which extended up to 150 eV. Also three other data sets by Ryabov et al.,[11], Corvi et al.,[12] and Felvinci[13] were considered to arrive at a recommended resonance parameter set for ^{235}U. Other data sets available at the NNDC (CSISRS Library) were also surveyed for the purpose of ^{235}U resonance parameter evaluation.

In evaluating the resonance parameters an attempt was made to conserve fission areas with proper $2g\Gamma_n$, Γ_f and Γ_γ ($\Gamma \sim \Gamma_f + \Gamma_\gamma$; $\Gamma_n \ll \Gamma$) and these parameters were mainly derived from Moore et al.,[14], Michaudon et al.,[9] and Blons et al.,[10] combined set, and Ryabov et al.,[11] data set.

The spin assignments made by Keyworth et al.,[8] based on their polarization measurements were adopted up to 300 eV. However, the resonance parameters $2g\Gamma_n$, Γ_f, Γ_γ, and $\sigma_o\Gamma_f$ extend up to about 150 eV, beyond which only the spins are given extending up to 300 eV. More precise capture measurements would reduce the errors on Γ_γ values and would provide a further check on the total widths required to describe resonances. The total widths derived by Michaudon et al.,[9] are not always compatible with the fission widths and areas reported by Moore et al.[14] The recommended parameters reflect the single-level aspect of the Breit-Wigner formalism. These should not be compared with any multi-level parameters for ^{235}U.

A calculation of the thermal capture and fission* cross-sections indicates that bound levels are required to describe these cross-sections and the capture and fission resonance integrals as well. Table I presents the recommended average resonance parameters. For comparison Moore's[15] and Lynn's[16] estimates of average capture widths are included along with our own estimation based on the expression derived by Malecky[17] and which was derived from systematics.

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* A reevaluation of the ^{235}U thermal parameters and resonance integrals is in progress, as such consistency check of the differential and itegral measurement was not undertaken yet.

Table I

Average Resonance Parameters

Target	B_n(KeV)	J^π	D_J(eV)	S_o	S_1	Γ_γ(meV)	Γ_γ(meV)[a]	Γ_γ(meV)[b]	Γ_γ(meV)[c]
^{235}U	6550	3^-	1.10±0.06	1.03±0.10	1.8±0.3	39±3	37	37	35.4±1.9
		4^-	0.88±0.05						34.2±2.0
^{238}U	4806	$1/2^+$	22.5±1.4	1.0±0.1	1.7±0.3	23.2±0.3	34	25	21.5±1.4
^{239}Pu	6534	0^+	7.9±0.8	1.3±0.1		42±3	43	48	36.0±2.4
		1^+	3.4±0.2						35.9±2.5
^{240}Pu	5241	$1/2^+$	11.5±1.9	0.93±0.08		31±2	38	36	29.7±1.5
^{241}Pu	6309	2^+	2.7±0.3	1.5±0.3		38±3	38	33	35.9±1.0
		3^+	2.7±0.3						35.6±1.0
^{242}Pu	5034	$1/2^+$	15.6±1.7	0.90±0.16		27±2	36	26	27.0±1.3

(a) Present calculations based on systematics (ref. 17).
(b) Lynn calculations based on giant dipole resonance model (ref. 16).
(c) Moore calculations based on giant dipole resonance model (ref. 15).

Since the resonance spins are uniquely assigned ($J^{\pi}=3^{-}$ and 4^{-}) for s-wave resonances, ^{235}U provides a very good example for testing the Δ_3 statistics on a target with nonzero spin: Each (ℓ,J) sequence of resonances may be considered as a pure sample for statistical purposes. The Δ_3 values calculated for the observed sequence of levels for J=3 and 4 are listed in Table II at E_n=60 eV, 70 eV and the maximum energy considered for evaluating the average level spacing for each set. The corresponding theoretical (Δ_3) values are also included with 0.11 as uncertainty.

In the case of J=3 resonances the experimental Δ_3 value agrees with the theoretical value up to about 100 eV. Based on the Δ_3 value of 0.366 one could estimate the number of missed levels with the help of probability curves provided by Georgopulos and Camarda[5]. The number of missed J=3 levels seems to be about 5. Hence the recommended average level spacing for spin 3 resonances is $1.10 \pm {}^{0.00}_{0.06}$ eV.

In the case of J=4 resonances it is estimated that 6 levels are missed in an energy region up to 73 eV. Thus the average level spacing is $0.86 \pm {}^{0.00}_{0.05}$ eV. The quoted errors reflect only the missed level information.

It is interesting to note the comparison between experimental and theoretical Δ_3 values for J=3 and J=4 resonances. A sudden increase in the experimental Δ_3 value indicates possible missing of levels (Fig. 2) or the presence of spurious levels.

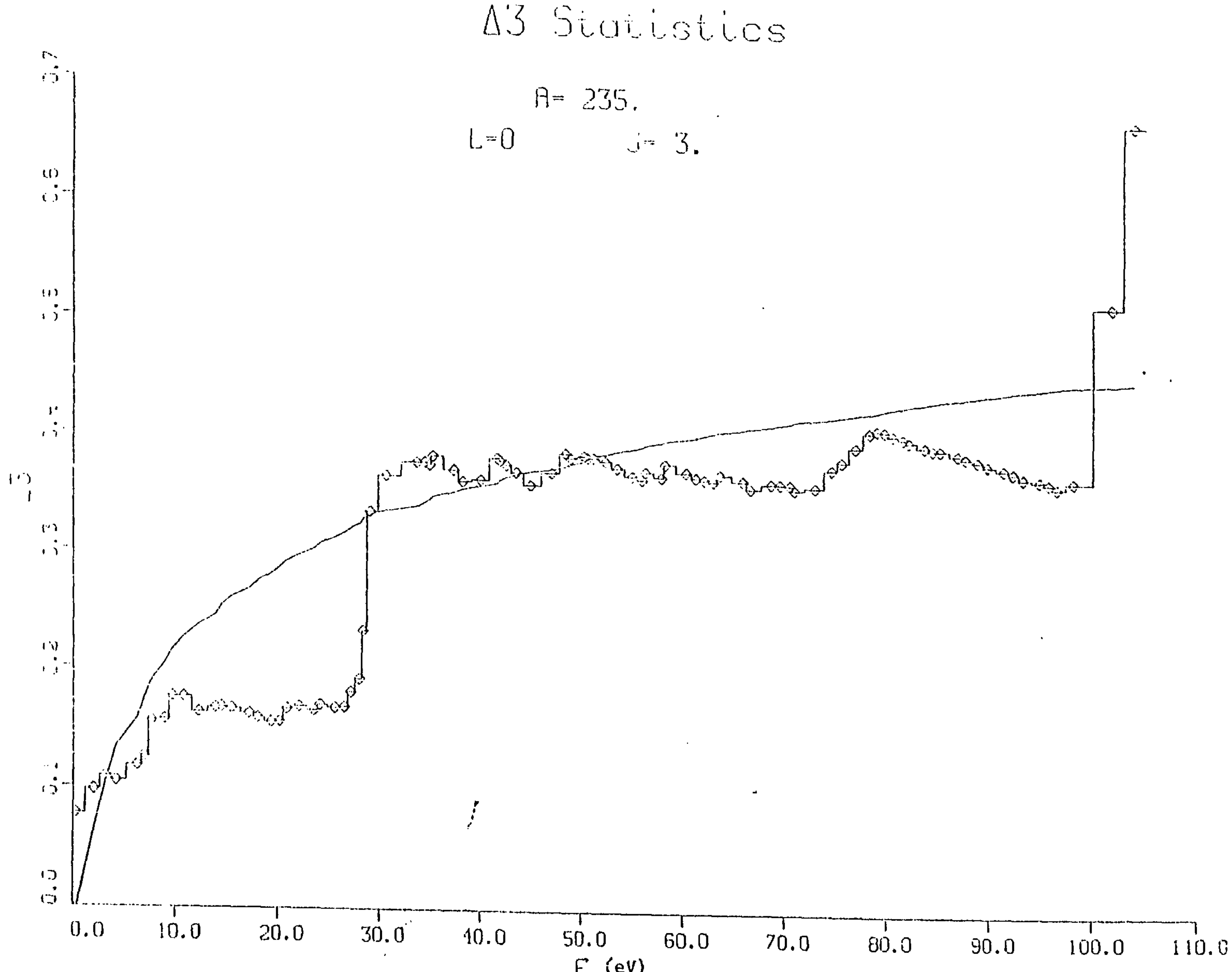

Fig. 2a. Comparison of experimental and theoretical Δ_3 values for ^{235}U J=3^- resonances.

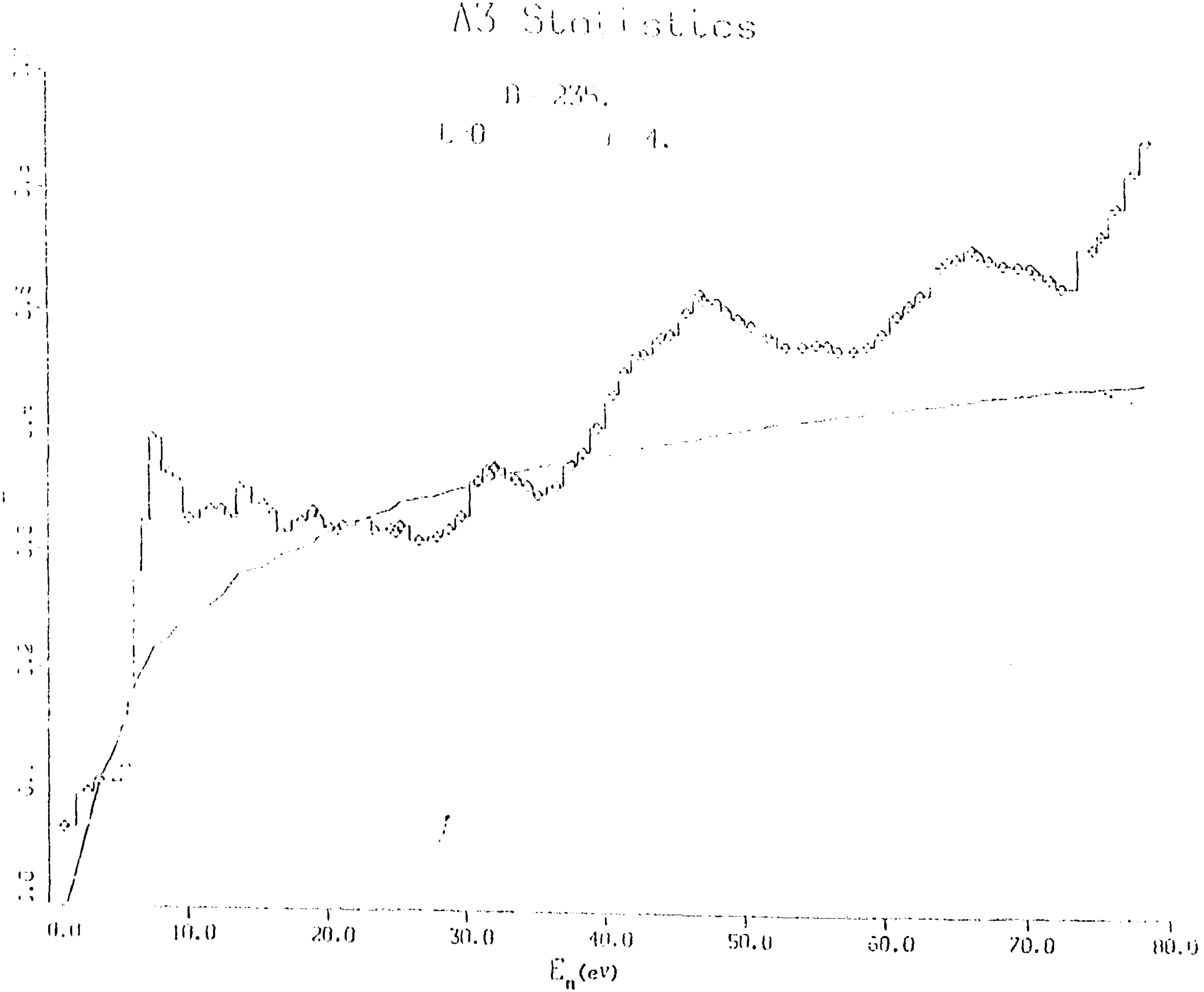

Fig. 2b. Comparison of experimental and theoretical Δ_3 values for ^{235}U $J=4^-$ resonances.

Table II Comparison between Theoretical and Experimental Δ_3 Values

J^π	E_n(eV)	Δ_3 Experimental	Δ_3 Theoretical	No. of Resonances	D_J(eV)
3^-	60	0.370	0.399 ± 0.11	54	1.091 ± 0.077*
	70	0.362	0.413 ± 0.11	63	1.090 ± 0.071
	100	0.366	0.446 ± 0.11	89	1.090 ± 0.060
4^-	60	0.487	0.422 ± 0.11	68	0.854 ± 0.053
	70	0.545	0.437 ± 0.11	79	0.862 ± 0.050
	73	0.523	0.442 ± 0.11	88	0.863 ± 0.049

* The errors are estimated with the help of the equation $\Delta\langle D\rangle = 0.52\langle D\rangle N^{-1/2}$.

^{238}U:

Several measurements[16-24] to determine more accurate values of the resonance parameters of ^{238}U have been carried out. These include transmission, scattering, capture, and self indication measurements. The major emphasis was placed on the determination of the radiative widths of the low energy s-wave resonances. The results of these investigations are the decrease of the value of the radiative width of the 6.67 eV resonance from 26 ± 2 meV to 22.8 ± 0.6 meV and the increase of the scattering width of the 20.9 eV resonance from 8.7 ± 0.5 meV to 10.0 ± 0.2 meV.

In addition, the parity assignments of resonances are at present placed on a firm basis because a new technique to establish the l values of ^{238}U resonances was developed by Corvi et al.[25] This is based on the gamma ray spectra differences between s and p-wave neutron capture.

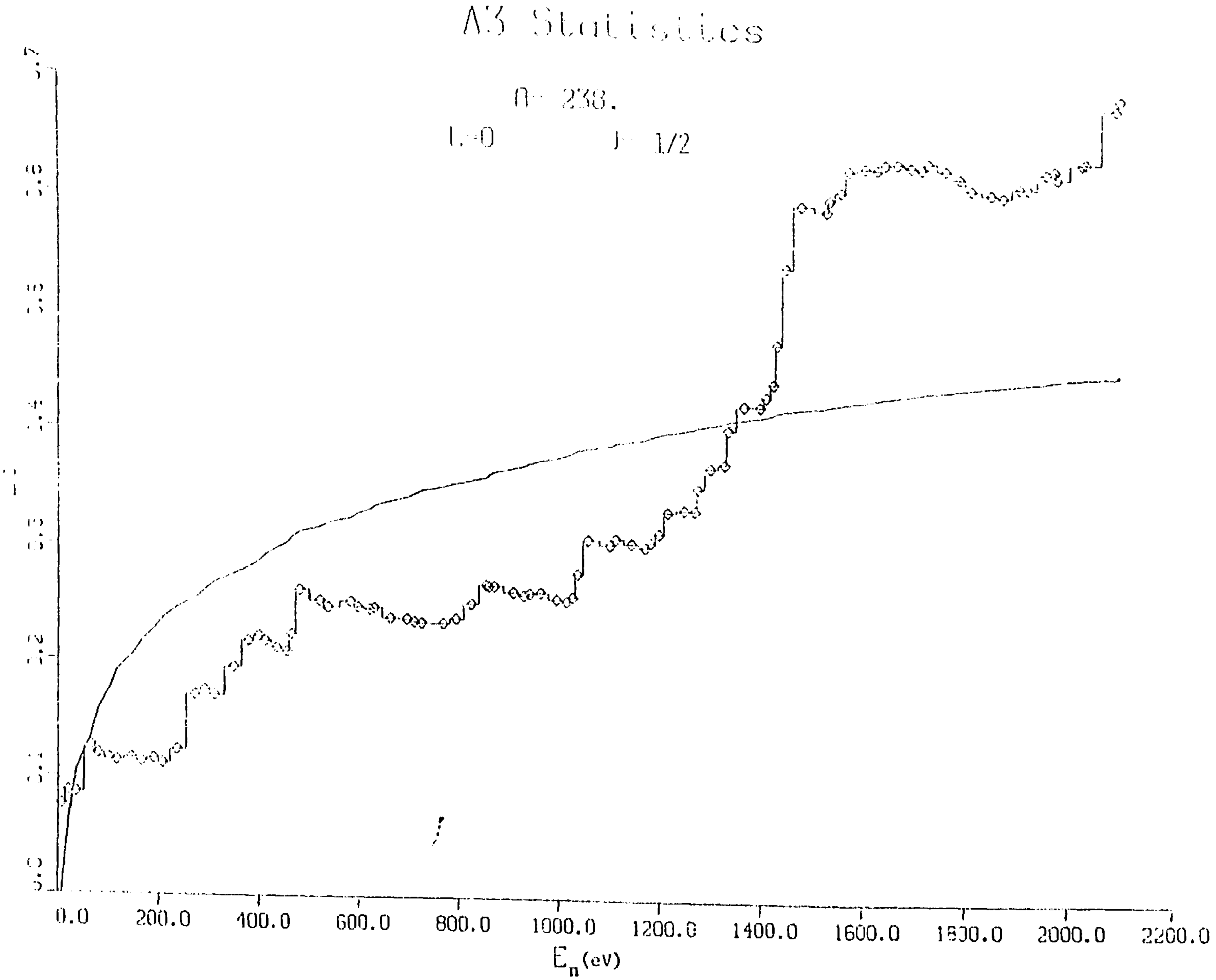

Fig. 3. Comparison of experimental and theoretical Δ_3 values for ^{238}U s-wav

With the aid of this technique, Corvi et al.,[25] were able to identify the p-wave resonances of ^{238}U in the energy range 63-1548 eV. More recently Moore et al., [26] successfully extended these measurements up to an energy of 3341 eV.

The variation of the theoretical and experimental Δ_3 values calculated for the s-wave resonances is illustrated in Fig. 3. As shown, the sudden increase of the experimental Δ_3 value at a neutron energy of 1400 eV indicates that s-wave levels are either missed or possibly misassigned as p-wave levels. Below this energy, the s-wave average level spacing is determined to be 22.5 ± 1.4 eV. The recommeded average resonance parameters of ^{238}U are shown in Table I.

The recommended positive energy s-wave resonances contribute 2.35b to the 2200 m/sec capture cross section. This is to be compared with the recent measurement of Poenitz et al.,[27] who reports $\sigma_\gamma = 2.68 \pm 0.019$ b. Calculations of the direct capture component in the framework of the Lane and Lynn[31] theory following the Mughabghab [28] approach indicate that the nonresonant contribution to the thermal capture cross section is 0.08 b. Therefore, a negative energy resonance is postulated in order to take into account the difference between the calculations and the measurement and to describe the coherent scattering length. The parameters of this resonance are shown in Table III.

^{239}Pu:

No major changes have taken place in the recommended resonance parameters of ^{239}Pu because no new measurements have been carried out as yet.

Table III. The Recommended Thermal Cross Sections, Average Resonance Parameters and the Low Energy Resonances of ^{238}U.

THERMAL CROSS SECTIONS

σ_γ^0 = 2.680±0.019 b
σ_s = 9.38±0.09 b
σ_α = 0.0013±0.0000 mb
σ_f^c = 0.047 mb
ν_p = 1.98±0.03
b_{coh} = 8.55±0.04 fm
R' = 9.6±0.1 fm

RESONANCE PROPERTIES

$<\Gamma_{\gamma 0}>$ = 0.0232±0.0003 eV
D_0 = 22.5±1.4 eV
D_1 = 9.7±1.3 eV
S_0 = 1.0±0.1
S_1 = 1.7±0.3
$S_{\gamma 0}$ = 10.3±0.8
I_γ = 277±3 b
I_f^c = 1.50±0.15 mb

RESONANCE PARAMETERS

J^π = 0$^+$
$\sigma_\gamma(+)$ = 2.35 b
$\sigma_\gamma(B)$ = 0.33 b
S_n = 4806.2±0.4 keV
$\sigma_\gamma(D)$ = 0.00 b

E_0 (eV)	J	ℓ	Γ (meV)	$g\Gamma_n$ (meV)	Γ_γ (meV)	$g\Gamma_n^0$ (meV)	$g\Gamma_n^1$ (meV)	Γ_f (meV)
-137					(23.2)	84.4		
4.409±0.004		1		0.00011± 0.00001			3.5 ±0.3	
*6.671±0.002	1/2	0	24.3 ± 0.6	1.50 ± 0.02	22.8±0.8	0.581 ±0.008		0.0000097±0.000001
*10.237±0.003	3/2	1		0.00165± 0.00005			14.9 ±0.5	
11.309±0.008		1		0.00039± 0.00003			3.0 ±0.2	
19.529±0.008		1		0.0013 ± 0.0001			4.5 ±0.3	
*20.872±0.008	1/2	0	33.54± 0.50	10.04 ± 0.20	23.5±0.6	2.198 ±0.044		0.000058 ±0.000001
*36.680±0.011	1/2	0	57.5 ± 0.5	34.0 ± 0.4	23.0±0.3	5.61 ±0.07		0.0000098±0.000001
45.17 ±0.04		1		0.0019 ± 0.0005			1.9 ±0.5	
49.62 ±0.07		1		0.0012 ± 0.0006			1.0 ±0.5	

$^{238}_{92}$U
[4.468 · 10^9 y]

^{240}Pu:

The importance of the parameters of the 1.056 eV resonance has been recently emphasized by Weston.[29] It was suggested[30] that the parameters of this resonance may provide a solution to the discrepancy between the microscopic differential and integral data. Since this resonance contributes 98% to the 2200 m/sec capture cross section, the highly accurate measurement of the thermal capture cross section by Lounsbury et al.,[31] imposes a constraint on the product $\Gamma_n \Gamma_\gamma$. Recent transmission and capture measurements carried out by Liou and Chrien[32] reveal that the capture width of this resonance is $\Gamma_\gamma = 32.4 \pm 1.0$ meV and the scattering width is $\Gamma_n = 2.32 \pm 0.06$ meV. These are more accurate than, but consistent with, the previously recommended parameters[33] : $\Gamma_n = 2.30 \pm 0.15$ meV and $\Gamma_\gamma = 31 \pm 3$ meV. Other recent measurements on the fission cross section by Auchampaugh and Weston[34] are concerned with the study of the subthreshold fission widths and hence the fission reaction mechanism. Previous similar investigations were made by Migneco and Theobald[35] .

The recommended resonance parameters of ^{240}Pu cover the energy range from thermal to 5.692 keV and are basically based on the measurements of Kolar and Bockoff[36] , Weigmann and Theobald[37] , Weigmann and Schmid[38] , and Hockenbury et al.[39] . The parameters of the 1.056 eV resonance are based on the very recent measurements of Liou and Chrien[32] . The highly accurate values of both the thermal capture cross section (σ_γ^0 = 289.5 ± 1.4 b) and the coherent scattering length (b = 3.5 ± 0.1 fm at E_n = 0.08 eV) indicate that a bound level is required to fit the thermal region. The parameters of this negative energy resonance are derived and are included in the evaluation. The position of this resonance is determined to be − 9.8 eV which is comparable with an average level spacing of 11.5 ± 1.9 eV.

The latter is obtained with the aid of the Δ_3 statistics. It is interesting to remark here that a calculation of the average radiative width on the basis of systematics does not reproduce the experimental value $\langle \Gamma_\gamma \rangle = 31 \pm 2$ meV. The same trend seems to be true for the other even-even target nuclei ^{238}U and ^{242}Pu. The recommended average resonance parameters along with the individual parameters of resonances up to 135.3 eV are shown in Table IV.

^{241}Pu:

Since, 1973, only one resonance parameter data set by Blons and Dirrien[40] is reported. The Reich-Moore formalism was applied to the fission cross section measurement which was carried out on a sample cooled down to 77K to reduce the Doppler effect. The neutron energy region covered in their measurements is from 0.26 eV to 103.66 eV. These parameters were adopted in the evaluation.

In addition, the average radiative width is calculated on the basis of the systematics[17] and is determined to be 38 meV. This is to be compared with values of 33 meV and 35.9 ± 1.0 meV ($J^\pi = 2^+$) and 35.6 ± 1.0 meV ($J^\pi = 3^+$) calculated respectively by Lynn[16] and Moore[15] on the basis of the giant dipole resonance model. The average level spacing for both spin states is determined as 1.3 ± 0.1 eV.

^{242}Pu:

The recommended thermal capture cross section at 0.0253 eV is evaluated as $\sigma_\gamma^0 = 18.46 \pm 0.49$ b, which is based on the measurements of Young et al.[41] Durham et al.,[42] and Butler et al.[43] The scattering cross section,

Table IV. The Recommended Thermal Cross Sections, Average Resonance Parameters and the Low-Energy Resonances of ^{240}Pu.

THERMAL CROSS SECTIONS

σ_γ^o = 289.5±1.4 b
σ_s = 1.64±0.06 b
σ_f = 0.056±0.030 b
ν_{sp} = 2.17±0.01
b_{coh} = 3.5±0.1 fm
R' = 9.6±0.2 fm

RESONANCE PROPERTIES

$<\Gamma_{\gamma 0}>$ = 0.031±0.002 eV
D_0 = 11.5±1.0 eV
S_0 = 0.93±0.08
$S_{\gamma 0}$ = 27±5
I_γ ≈ 8400±200 b
I_f^c = 1.0 b

RESONANCE PARAMETERS

$J^\pi = 0^+$
$\sigma_\gamma(+)$ = 288.3 b

$\sigma_f(B)$ = 3.2 b

S_n = 5241.3±0.7 keV

E_0 (eV)	J	ℓ	$g\Gamma_n$ (meV)	Γ_γ (meV)	$g\Gamma_n^0$ (meV)	Γ_f (meV)
-0.81				(32.4)	3.84	
1.057±0.002	1/2	0	2.32± 0.08	32.4±1.0	2.258 ±0.080	0.008
20.45 ±0.01			2.65± 0.07	32.2±2.4	0.588 ±0.015	
38.32 ±0.02	1/2	0	17.3 ± 0.5	29.4±2.8	2.79 ±0.08	
41.64 ±0.02			16.7 ± 0.6		2.59 ±0.09	
66.20 ±0.01			54 ± 1	31.0±1.7	6.8 ±0.1	
72.77 ±0.04			21.4 ± 0.7	28.5±1.5	2.51 ±0.08	
90.78 0.08			12.8 ± 0.3	27.5±3.0	1.34 ±0.03	
92.51 ±0.06			3.1 ± 0.1		0.32 ±0.01	
105.08 ±0.07			44.0 ± 1.5	35.6±2.0	4.29 ±0.15	
121.6 ±0.1			14.2 ± 0.8	31 ±4	1.29 ±0.05	
130.7 ±0.1			0.17± 0.03		0.015 ±0.003	
135.3 ±0.1			18.7 ± 1.5	31.5±2.5	1.61 ±0.13	

$^{240}_{94}$Pu [6.563·10³ yr]

$^{240}_{94}$Pu [6.563·10³ yr]

$\sigma_s = 8.24 \pm 0.2$ b, is determined from the coherent scattering length a = 8.1 ± 0.1 fm. These thermal constants provide constraints on the parameters of a bound level and the first resonance at 2.68 eV.

Recent measurements of the resonance parameters of ^{242}Pu have been carried out by Poortmans et al.,[44] (σ_t, σ_γ, σ_s), Harvey et al.,[45] (σ_t) , and Auchampaugh and Weston[46] (σ_f) . In addition, fission areas reported by Auchampaugh et al.,[44] were used in deriving subthreshold fission widths for ^{242}Pu. There is general agreement between the various Γ_n values reported by these authors. The average radiative width determined by Poortmans et al.,[44] is 21.9 ± 1.1 meV which is slightly lower than the radiative width[48] of the 2.675 ± 0.002 eV resonance. A coherent scattering length of 8.1 ± 0.1 fm combined with a potential scattering length of 9.6 ± 0.2 fm obtained from the systematics and from neighboring nuclides indicate the presence of a nearby negative energy resonance. To obtain acceptable values for the parameters of this bound level, it was necessary to adopt the lower limit of the capture width (Γ_γ = 25.0 ± 1.5 meV) of the 2.68 eV resonance which was derived by Young and Reeder[48] who used a shape fit analysis adopting the Reich-Moore multilevel formula. It is interesting to point out here that the accurate position of the resonance energy, E_o = 2.676 ± 0.002, as determined by Schrack[49] and Harvey[45] can be used as a neutron energy standard.

The fission cross section at 0.0253 eV is calculated as 0.21 b from the evaluated resonance parameters. In addition, the parameters indicate that the capture and fission resonance integrals are 1107 b and 6.4 b respectively. With the aid of the Δ_3 statistics, the average level spacing is determined as D_o =15.6 ± 1.7 eV.

4. CONCLUSION AND SUMMARY

The current status of the resonance parameters of the actinides $^{235,238}U$ and $^{239-241}Pu$ is briefly described. Average resonance parameters are derived and in particular the Dyson-Mehta Δ_3 statistics was applied in conjunction with the staircase plots to arrive at average level spacings. Average radiative widths are calculated here on the basis of systematics as derived by Malecky et al.,[17] and are compared with Moore's[15] and Lynn's[16] estimates. It is interesting to point out that the calculations on the basis of systematics are in reasonable agreement with the experimental values for the even-odd target nuclides ^{235}U and $^{239,241}Pu$ but not for the even-even target nuclides ^{238}U and $^{240,242}Pu$. The radiative widths of the low energy neutron resonances of ^{238}U are at present well established thus resolving a previous discrepancy between integral and differential measurements. Additional spin assignments for ^{235}U have been made using polarization measurement. It seems that the (2J+1) law is obeyed in this case. It will be of great interest to apply the same experimental technique to determine the spins of ^{241}Pu resonances. At present these were determined by the method of interference in the fission channel which does not give unambiguous assignments.

The scattering and radiative widths of the ^{240}Pu resonance at 1.057 eV are determined[32] to a higher accuracy. The result of this measurement indicates that a re-examination of the integral measurements for ^{240}Pu is necessary.

The recommended resonance parameters of "BNL-325" which can be presented in an ENDF-type format are available on request from the National Nuclear Data Center.

Acknowledgments

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