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PARAMETERS OF THE SUBTHRESHOLD FISSION STRUCTURE IN Pu^{242} *

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Abstract

The neutron total cross section of Pu^{242} has been measured from 600 eV to 81 keV using the new LLL Linac. Parameters were obtained for these resonances below 4 keV with significant fission strengths. These parameters were used to study the fission widths of the intermediate structure in Pu^{242} and the coupling matrix elements between the levels in the primary and secondary wells in the double-humped fission potential barrier. The results strongly suggest that the coupling in Pu^{242} is very weak and that the widths of the levels in the intermediate well are very narrow, the widths being less than the average spacing between levels in the primary well. The distribution of the fission widths of the levels in the primary well about their expected values is found to be consistent with a Porter-Thomas distribution. However, the distribution of the square of the coupling matrix element appears to require more than one degree of freedom. The Wigner distribution is shown to adequately describe the spacings between levels in the intermediate well.

Introduction

Measurements have been made on the subthreshold fission cross section of Pu^{242} by several authors[1-3]. These measurements have revealed pronounced clusters of resonances with relatively large fission strengths, separated by 500eV to 1 keV, below the neutron fission threshold. The intermediate structure seen in subthreshold fissioning isotopes has been explained by Lynn[4] and Weigmann[5] in terms of a second mimimum in the fission potential barrier which had been predicted by Strutinsky[6]. A lack of detailed information about the neutron strengths of the resonances in each cluster in Pu^{242} has prevented a study of the fission widths of these resonances and consequently any properties of the intermediate barrier or of the levels in the second minimum. For this reason a high resolution neutron time-of-flight measurement of the total cross section of Pu^{242} was undertaken at the new LLL Linac with emphasis on determining the neutron widths of those resonances in each cluster seen in the fission cross section.

Experimental Technique

The experimental details of the total cross-section measurement are given in Table I. Two iron-copper collimators, 0.51 meters long, with axial holes 2.74 cm in diameter and 2.29 cm in diameter, located at 2.3 and 5.3 meters, respectively, from a bare Ta target defined the neutron beam prior to intercepting the Pu samples. The water coolant for the neutron target served as the moderator. The sample changer was located just after the second collimator. It contained the B^{10} overlap filters, the two Pu samples, and a blank sample. The B^{10} powder filter was used during the thin sample measurement and the borated polyethylene filter during the thick sample measurement. The Pu metal was mixed with 0.977 wt % of aluminum, cast into disks, and encapsulated in aluminum cans with 0.0039 cm Al windows. An unfilled aluminum can was used as the blank sample. A B^{10}, 480-keV γ-ray detector located at 30 m from the Ta target along another flight tube oriented at 90° to the 253-m flight tube was used as an integral flux monitor. The events from this detector controlled the sample-in to sample-out time set at a ratio of 4 to 1. The neutron detector system consisted of four identical Li^{6}-loaded glass scintillators mounted on RCA 4525 photomultiplier tubes. The current from each detector was integrated, double delay time clipped, and passed through a zero-cross differential discriminator. The window of each discriminator was set to bracket the $Li^{6}(n,\alpha)H^{3}$ peak in the scintillator. The outputs from each discriminator were mixed and sent back to the control room where they were reshaped by a fast discriminator. These stop pulses were then fed into an EG&G TDC 100 time digitizer which had been started by a signal from a gamma sensitive detector located in the neutron target cell. The time digitizer was interfaced to a 16 K PDP-15 computer which served as the data acquisition system for this experiment. To cover the entire energy region with good resolution in one run it was necessary to divide each memory location into three channels, each channel storing a maximum of 63 events. The computer was programmed to provide a live display of the data, automatically cycle the samples after a preset number of B^{10} pulses had been recorded, and either dump memory onto DECTAPE after a sample change or onto IBM magnetic tape when a channel overflowed. Channel overflows occurred at a frequency of less than one every few hours due to the low counting rate ($<$ 300/second) in this experiment.

The characteristic of the thick and thin Pu^{242} samples used in this measurement are given in Table I. Each sample was cycled every 12 to 15 minutes with the blank sample. Approximately 40 hours of data were accumulated for each sample.

The neutron background present during the measurement was determined by "black" resonances in Pu^{242} and by the 5.906 keV aluminum resonance. The neutron beam passed through approximately 0.4 cm of aluminum as a result of an aluminum window at the end of the evacuated flight tube and the aluminum cans holding the Li^{6} glass scintillators onto the photomultiplier tubes. Six Pu^{242} resonances in the thick-sample run and three in the thin-sample run, along with the aluminum resonance, were used to determine the background. It was measured to be a constant 30% of the open beam spectrum up to 6 keV. Unfortunately time did not permit the investigation required to determine the source of the background and to eliminate it. The data were normalized by assuming a 10-barn potential scattering cross section between resonances below 6 keV.

Determination of resonance parameters

To extract resonance parameters from two independent sets of data, it is desirable to have both sets of data on the same energy scale. This is especially true when the sets of data are obtained with vastly different energy resolution and when the resonances of interest are not completely resolved. The fission cross section data of Auchampaugh et al.[1] were used along with the present data to extract values of Γ_n and Γ_f. Since the resolution of the fission measurement made with neutrons from an underground nuclear explosion was much poorer than that from the present measurement, care was taken to ensure that the energy scales of the two sets of data were properly aligned. For convenience the energy scale of the present measurement was adjusted to that of the fission measurement by requiring that all resonance energies in the cluster at 762.5 eV agree in both sets of data. The present measurement shows that this energy scale is incorrect and a new energy scale has been adopted which is correct for the 5.906 keV resonance in aluminum.

a) Class I resonance parameters

The results of the analysis of the total cross section data are given in Table II. The Harvey-Atta area analysis code[7] was used to extract values of Γ_n from the total data for those resonances in the fission clusters at 762.5, 1836, 2741, 3112, 3568, and 3670 eV. The neutron width for the 762.5 eV resonance of 3.8 ± 1.1 meV (superscript a) was determined from the value of the peak height of this resonance in the total cross-section data (the strong Doppler effect at this energy, the fact that the resonance was situated on the tail of a very large resonance, and the small size of the resonance prevented an accurate determination of its complete area). At 762 eV resolution broadening is negligible and the shape of a small resonance is strongly influenced by the Doppler effect. Under such conditions the peak height of a resonance is directly related to Γ_n and is independent of Γ at least to first order in the parameter $1/\theta = \Gamma/2\Delta$. Values of Γ_n with the superscript b were obtained from the data taken recently by F. Simpson et al[8] at ORNL. The statistical quality of their data is at least a factor of 4 better than ours and consequently has revealed some of the smaller resonances not definable in the present data. The neutron widths of the resonances at 1783, 1789, and 1836 eV which were not statistically definable in the present total data have been set less than the smallest neutron width measured in the 1836-eV cluster. A lower limit to Γ_n for those resonances where only an upper limit could be set can be obtained from the fission areas by setting $\Gamma_f = \Gamma$. These values of Γ_n are given with the superscript c.

The fission areas corrected for a small γ-ray contribution of magnitude 0.007 A_γ^1 (capture area, calculated from the neutron widths) and a value of $\Gamma_\gamma = 30 \pm 5$ meV were used to deduce the values of Γ_f. No values of Γ_f for the 762.5, 1836, 3112, and 3568-eV resonances were computed. For the 762.5-eV resonance the measured neutron width was

smaller than that obtained from the fission area with $\Gamma_f = \Gamma$.
For the other three resonances Γ_n was not measurable. The upper limits given are not much larger than the values obtained from the fission areas with $\Gamma_f = \Gamma$. Consequently any fission widths derived for these resonances would be subject to large uncertainties.

The fission widths for the 2741 and 3670-eV resonances were computed using a value of the capture width appropriate to the primary well for the Pu^{242} compound nucleus. However, it is quite possible that the strong fission resonances are the class II states in which case the appropriate capture width should be that for the intermediate well which, according to Lynn,[4] should be a few times smaller than the capture width in the primary well. This uncertainty in the correct value of Γ_γ makes it difficult to compute meaningful values for the class II fission widths.

b) Class II Resonance parameters

In a very weak coupling situation where the magnitude of the coupling matrix element, $|H_{\lambda'\lambda''}|$, is less than the average class I level spacing, D_I, the unperturbed class II fission width $\Gamma^{II}_{\lambda''f}$ to a very good approximation is just equal to the sum of the measured fission widths, $\Gamma^{II}_{\lambda''f} \approx \sum_{\lambda' \neq \lambda''} \Gamma_{\lambda' f} + \Gamma_{\lambda'' f}$, where $\Gamma_{\lambda'' f}$ is the perturbed Class II fission width. For those cases where it is not possible to accurately determine the fission width of the resonance making the largest contribution to the sum, an upper limit on $\Gamma^{II}_{\lambda''f}$ can be set using the following expression:[4]

$$\Gamma^{II}_{\lambda''f} \approx \left\{ \sum_{\lambda' \neq \lambda''} \sqrt{\Gamma_{\lambda' n}\, \Gamma_{\lambda' f} / \Gamma_{\lambda'' n}(o)\ II} \right\}^2 .$$

Upper limits to $\Gamma^{II}_{\lambda''f}$ were calculated from the above expression for the resonances at 762.5, 1836, 3112, and 3568 eV assuming that the values of Γ_n determined from the fission areas with $\Gamma_f = \Gamma$ represent lower limits to the class II neutron widths. These values of $\Gamma^{II}_{\lambda''f}$ are probably an overestimate to a realistic upper limit on $\Gamma^{II}_{\lambda''f}$. Applying the same equation to the states at 2741 and 3670 eV results in values of $\Gamma^{II}_{\lambda''f}$ an order of magnitude larger than that observed. The lower limits to $\Gamma^{II}_{\lambda''f}$ represent sums of the measured fission widths, excluding the fission widths for the strongest resonances, within each cluster.

Values of the product of the square of the coupling matrix element, $H^2_{\lambda'\lambda''}$, and the class II fission width were computed from the equation $H^2_{\lambda'\lambda''}\Gamma^{II}_{\lambda''f} = \Gamma_{\lambda'f}\left(E^I_{\lambda'} - E^{II}_{\lambda''}\right)^2$ and are given in Table II. The range of values of the average of the square of the coupling matrix element, $\overline{H^2_{\lambda''}}$, for each group of resonances were obtained from the upper and lower limits on $\Gamma^{II}_{\lambda''f}$ and the average values of $H^2_{\lambda'\lambda''}\Gamma^{II}_{\lambda''f}$.

Information on the intermediate structure

It is instructive to calculate, for the two class II states at 2741 and 3670 eV which have reasonably well-defined class II fission widths, just how large the class II neutron widths can be for these states. Lynn[4] gives the following expression for the expectation value of the reduced neutron width of a class II state:

$$\left\langle \Gamma^{II(o)}_{\lambda''n} \right\rangle \approx \sum_{\lambda' \neq \lambda''} \frac{\Gamma_{\lambda'f}\Gamma^{o}_{\lambda'n}}{\Gamma^{II}_{\lambda''f} - \Gamma_{\lambda'f}}$$

Applying this expression to the two cases gives 22 and 33 meV for the expectation value of the class II neutron widths for the states at 2741 and 3670 eV, respectively. The upper limits on $\Gamma^{II}_{\lambda'f}$ were used in these calculations. The measured values for these resonances are 3 to 4 times smaller. Therefore, one can expect to observe class II states directly and it may well be that those resonances observed in the fission data with the strong fission components are indeed the class II states.

The method of Wilets[9] was used to determine the number of degrees of freedom, ν_{eff}, of the chi-square distribution function governing the class I fission widths about their expected values, $\Gamma_{\lambda'f} - \langle\Gamma_f\rangle = \Gamma_{\lambda'f} - \overline{H^2_{\lambda''}\Gamma^{II}_{\lambda''f}} \Big/ \left(E^I_{\lambda'} - E^{II}_{\lambda''}\right)^2$ and that of the product of the square of the coupling matrix elements, and the fission width $H_{\lambda'\lambda''}{}^2\,\Gamma^{II}_{\lambda''f} = \Gamma_{\lambda'f} \Big/ \left(E^I_{\lambda'} - E^{II}_{\lambda''}\right)^2$, for each of the six groups of resonances. The results are given in Table III. The errors are computed from the expression for

$\delta\nu_{eff}/\nu_{eff}$ given by Wilets, assuming an average error of 40% for the quantities $H_{\lambda'\lambda''}{}^2\Gamma_{\lambda''f}^{II}$ and $\Gamma_{\lambda'f} - \langle\Gamma_f\rangle$. If all the resonances are considered as one group, weighting each $H^2_{\lambda'\lambda''}\Gamma_f^{II}$ and $\Gamma_{\lambda'f}-\langle\Gamma\rangle$ by their average values for each individual group, then $\nu_{eff}\left(H^2_{\lambda'\lambda''}\,\Gamma^{II}_{\lambda''f}\right) = 1.88 \pm 0.24$ and $\nu_{eff}\left(\Gamma_{\lambda'f} - \langle\Gamma_f\rangle\right) = 0.89 \pm 0.11$. These determinations did not include the very strong fission resonances nor the resonances at 755.6 eV and 3107 eV for which the quantities $H^2_{\lambda'\lambda''}\,\Gamma^{II}_{\lambda''f}$ and $\Gamma_{\lambda'f}$ are sensitive to the exact position of the class II level. The results for the quantity $\Gamma_{\lambda'f} - \langle\Gamma_f\rangle$ strongly suggest a Porter-Thomas[10] distribution. Whether the square of the coupling matrix elements or equivalently $H^2_{\lambda''\lambda'}\Gamma^{II}_{\lambda''f}$ for a given group obey a similar distribution is questionable in view of the low (< 1%) probability of observing a value of ν_{eff} greater than 1.88 for a set of 63 Porter-Thomas quantities. This probability was determined by randomly selecting 63 values from a Porter-Thomas distribution of mean 1, determining ν_{eff} for each set of 63, repeating the procedure a larger number of times and comparing the number of times ν_{eff} exceeded 1.9 to the total number of sets. The low probability for observing a ν_{eff} greater than 1.9 suggests that $H^2_{\lambda'\lambda''}$ is governed by a chi-square distribution of more than one degree of freedom.

The reported fission data[1] on Pu^{242} revealed about 53 narrow groups of resonances (class I) below 30 keV with an average spacing between groups of 553 eV . A detailed analysis of six of these groups below 4 keV believed to be levels (class II) in the intermediate well gives an average spacing of 553 eV, indicating that most of the groups observed below 30 keV are indeed levels in the second well. Since the target nucleus is even, the Wigner single spacing repulsion surmise[11] should act between these levels. To test this assumption a histogram of the number of levels in 100-eV bins versus their spacing has been plotted and compared with the Wigner frequency function. This comparison is made in Fig. 1. The solid curve is a Wigner distribution for 53 levels with an average spacing of 553 eV. The spacings between class II levels appear to be well represented by a single-spacing Wigner distribution within the finite size of this sample.

Summary

The present neutron total cross-section measurement on Pu^{242} has provided neutron width information on resonances in the vicinity of six class II levels. This has enabled more detailed study to be made of the properties of the class II states than has heretofore been possible on any of the subthreshold fissioning nuclei. The fact that the upper limits on $|\sqrt{<H^2>}|$ and Γ_f^{II} are much less than D_I strongly suggests that we are dealing with a situation where the class II states are well below the saddle barrier and are very weakly coupled to the class I states. These results have also shown that the distribution of the class I fission widths about their expected value is consistent with a Porter-Thomas distribution and that of the distribution of the square of the coupling matrix elements appear to require more than one degree of freedom. In addition, the fission data have provided a sufficiently large set of class II resonances to show that the class II level spacings are well described by the Wigner surmise.

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Table I

Experimental Details

Time-of-flight parameters

electron pulse width	~40 nsec
pulse rate	720 pps
beam power	~ 8 KW
flight path	253.63 ± 0.1 m
overlap filters	
B^{10} in CH_2	0.6 gm/cm^2 B^{10}
B^{10} powder	0.35gm/cm^2 B^{10}

Memory configuration

channels	channel width (nsec)	energy interval (eV)
0-4095	8	81209 - 35648
4096--34815	16	35648 - 907
34816-39935	32	907 - 594

Neutron detector

4 Li^6-load glass scintillators

type	NE 905
diameter	10 cm
thickness	1.3 cm

Pu^{242} Metal sample characteristics

Isotopic composition

Pu^{238}	0.188 %
Pu^{239}	0.178 %
Pu^{240}	1.352 %
Pu^{241}	0.146 %
Pu^{242}	98.136 %

Sample No.	Wt. of Sample	Sample diameter	Thickness
1	33.88 gm	2.54 cm	0.0167 at/b
2	7.68 gm		0.00376at/b

Table II

Pu^{242} resonance parameters.

E (eV)	Γ_n (meV)	Γ_f (meV)	E_{II} (eV)	Γ_f^{II} (meV)	$\Gamma_f^{II} H_{\lambda'\lambda''}^2$ (eV^3)	$\overline{H_{\lambda''}^2}$ (eV^2)
595.2	28.9 ± 2.9	0.026 ± 0.007			0.72	
600.1	7.9 ± 0.8	0.134 ± 0.043			3.53	
610.9	10.1 ± 1.9	0.084 ± 0.026			1.95	
639.1	4.2 ± 0.6	< 0.05			0.75	
665.0	2.7 ± 0.5	< 0.08			0.77	
669.5	12.5 ± 1.0	0.11 ± 0.04			0.93	
693.4	32.3 ± 3.0	0.28 ± 0.06			1.33	
711.9	100 ± 16	0.048 ± 0.017			0.12	
727.9	3.6 ± 0.8	< 0.06			0.08	
736.8	120 ± 17	0.61 ± 0.14			0.39	
755.6	115 ± 18	1.65 ± 0.39			0.08	
762.5	3.8 ± 1.1[a] > 4.3 ± 0.5[c]		~ 762.5	< 300 >4.9		<165 >2.7
788.5	100 ± 30	1.24 ± 0.49			0.83	
794.5	275 ± 30	0.043 ± 0.009			0.05	
825.2	7.3 ± 1.0	0.17 ± 0.06			0.68	
837.9	35.0 ± 4.2	0.049 ± 0.017			0.28	
856.7	31.2 ± 3.1	0.17 ± 0.06			1.51	
866.1	11.0 ± 1.5	0.048 ± 0.018			0.50	
879.1	50 ± 5.0	0.031 ± 0.012			0.43	
886.8	24.8 ± 2.5	0.015 ± 0.006			0.22	
923.4	46.8 ± 5.0	0.037 ± 0.011			0.96	

TABLE II, continued

E (eV)	Γ_n (meV)	Γ_f (meV)	E_{II} (eV)	Γ_f^{II} (meV)	$\Gamma_f^{II} H^2_{\lambda'\lambda''}$ (eV^3)	$\overline{H^2_{\lambda''}}$ (eV^2)
1696	39.1 ± 5.5	0.067 ± 0.020			1.33	
1708	93.2 ± 12.1	0.001 ± 0.0005			0.02	
1737 1739	9.1 ± 2.4 21.5 ± 3.6	0.047 ± 0.014			0.46	
1751	9.4 ± 2.8	0.51 ± 0.20			3.78	
1762	85.4 ± 10.2	0.017 ± 0.004			0.09	
1783	< 4 >0.022 ± 0.009[c]	> 0.18			0.50	
1789	< 4 >0.13 ± 0.01[c]	> 1			2.20	
1806	12.9 ± 3.7	0.13 ± 0.05			0.09	
1820	4.1 ± 3.8	0.38 ± 0.37			0.09	
1836	< 4 >3.0 ± 0.3[c]		~1836	< 116 > 3.8		< 257 > 8.4
1862	4.9 ± 3.6	0.49 ± 0.39			0.28	
1881	84.3 ± 10.[illegible]	0.056 ± 0.013			0.11	
1891	4.1 ± 4.6	0.96 ± 1.13			2.70	
2699	250 ± 38	0.11 ± 0.03			0.18	
2734	108 ± 15	0.094 ± 0.031			0.006	
2741	6.9 ± 2[b]	1.45 ± 0.63	~ 2741	< 2.6 > 1.1		< 243 > 103
2756	220 ± 31	0.031 ± 0.016			0.008	
2772	2.8 ± 1.0[b]	0.91 ± 0.42			0.87	
3107	< 3[b] > 0.28 ± 0.05	> 3.4			0.084	
3112	< 3[b] > 1.2 ± 0.1[c]		~ 3112	< 480 > 5.4		< 142 > 1.6
3135	226 ± 32					
3142	40.2 ± 12.1	1.09 ± 0.22			0.79	
3156	52.3 ± 11.9	0.1 ± 0.04			0.19	
3164	3 ± 1.5	0.8 ± 0.5			2.16	

TABLE II, continued

E (eV)	Γ_n (meV)	Γ_f (meV)	E_{II} (eV)	Γ_f^{II} (meV)	$\Gamma_f^{II} H_{\lambda'\lambda''}^2$ (eV^3)	$\overline{H_{\lambda''}^2}$ (eV^2)
3412	380 ± 61	0.09 ± 0.03			2.16	
3422	10.4 ±5[b]	< 0.3			<6.40	
3438	7.8 ± 4[b]	< 0.3			< 5.06	
3451	220 ± 29	0.13 ± 0.03			1.75	
3472	78 ± 17	< 0.1			< 0.88	
3485	102 ± 19	0.28 ± 0.08			1.79	
3496	122 ± 23	0.36 ± 0.10			1.76	
3521	95 ± 21	< 0.2			< 0.40	
	< 4[b]	> 3				
3532					3.89	
	< .32 ± .05[c]					
	<4[b]	> 1				
3558					0.10	
	< .12 ± .03					
	< 4[b]			< 662		< 306
3568			~3568			
	> 1.2 ± 0.1[c]			> 6.7		> 3.1
3581	143 ± 24	0.18 ± 0.05			0.04	
3588	8.2 ± 4[b]	< 0.8			0.35	
3620	13.7 ± 5[b]	0.29 ± 0.16			0.72	
3629	5.5 ± 3[b]	< 1.9			3.19	
3651	155 ± 25	0.67 ± 0.18			0.22	
3670	13.9 ± 5[b]	7.1 ± 3.2	~3670	< 12 > 4.7		< 278 > 109
3698	13.1 ± 5[b]	< 0.3			< 0.23	
3712	12.2 ± 5[b]	0.81 ± 0.38			1.43	
3721	940 ± 200	0.05 ± 0.03			0.13	
3734	9.4 ± 5[b]	0.19 ± 0.16			0.78	
3773	246 ± 44	0.21 ± 0.08			2.18	
3790	62 ± 22	0.07 ± 0.04			1.02	
3812	683 ± 130	0.15 ± 0.06			3.11	
3836	948 ± 161	0.05 ± 0.02			1.42	

[a] Calculated from total cross section peak height.

[b] Obtained from data of F. Simpson et al.[8]

[c] Determined from fission areas setting $\Gamma = \Gamma_f$

TABLE III

Statistical information on the intermediate structure. The two columns give the effective number of degrees of freedom for the quantity $H^2_{\lambda'\lambda''}\,\Gamma^{II}_{\lambda''f}$ and the quantity $\Gamma_{\lambda'f} - \langle\Gamma_f\rangle$, respectively.

E^{II} (eV)	$\nu_{eff}\left(H^2_{\lambda'\lambda''}\,\Gamma^{II}_{\lambda''f}\right)$	$\nu_{eff}\left(\Gamma_{\lambda'f} - \langle\Gamma_f\rangle\right)$
762.5	2.0 ± 0.5	0.89 ± 0.17
1836	1.3 ± 0.3	1.05 ± 0.30
2741	1.1 ± 0.5	1.47 ± 0.55
3112	1.9 ± 0.7	0.72 ± 0.40
3568	2.2 ± 0.5	0.59 ± 0.18
3670	3.2 ± 1.0	0.83 ± 0.23

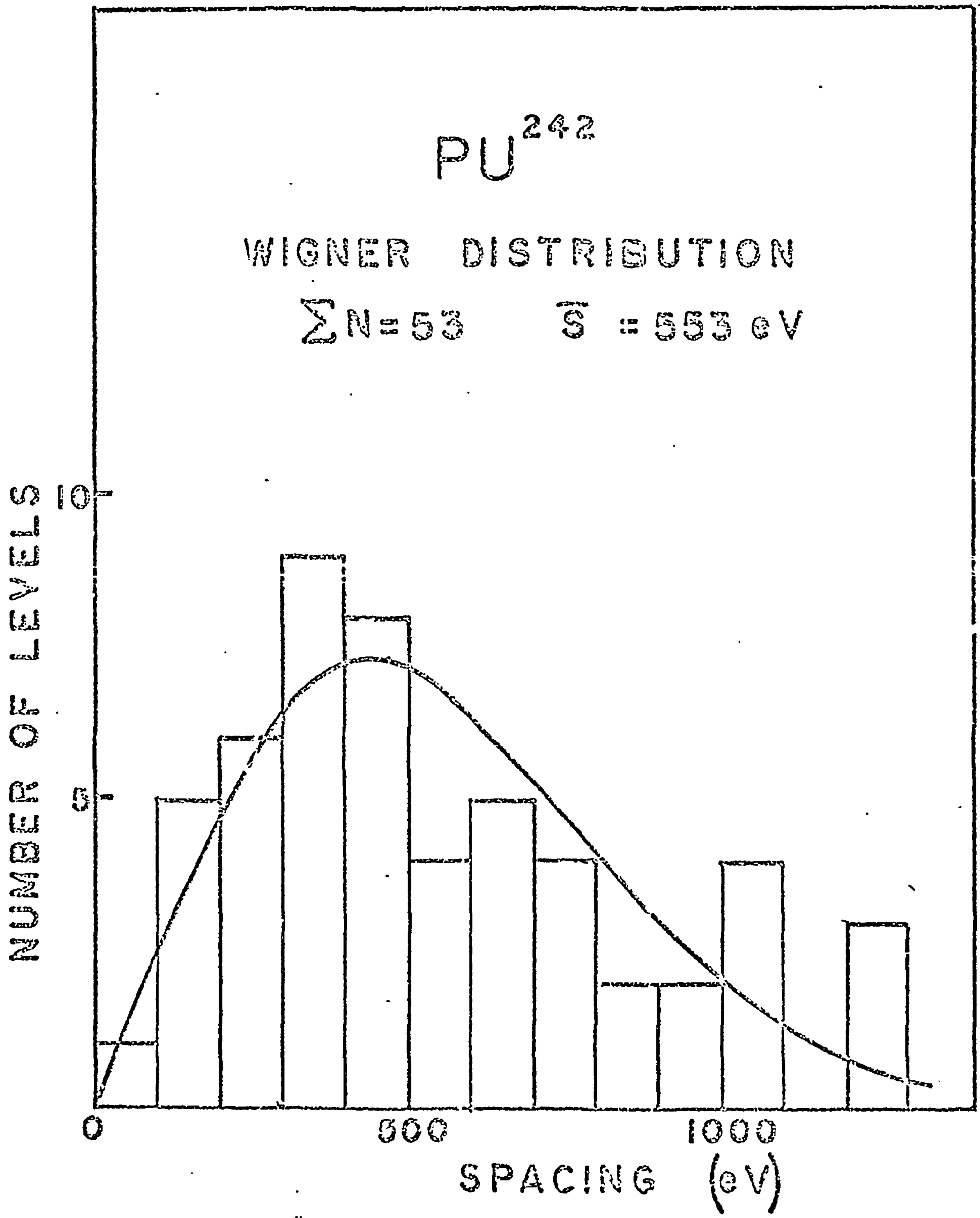
PU242
WIGNER DISTRIBUTION
ΣN=53 S̄ = 553 eV
NUMBER OF LEVELS
10
5
0
500
1000
SPACING (eV)

Figure 1