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INTERMEDIATE STRUCTURE IN UNRESOLVED RESONANCES OF FISSILE AND FERTILE NUCLIDES*

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ABSTRACT

Evidence for the existence of intermediate structure in the unresolved resonance fission and capture cross sections of ^{235}U and in the capture cross section of ^{238}U is reviewed. Certain of the statistical tests that have been used are known to be compromised by finite resolution when applied to unresolved resonance data, but we conclude that over most of the energy range the test results are valid. Parity assignments for the structure in ^{238}U(n,γ) suggest that s-wave neutron absorption is responsible for much of the structure. If this is the case, the cross section fluctuations are most likely due to fluctuations in the radiative capture width, rather than in the neutron width. Finally, the practical effect of the possible presence of width correlations in fertile and fissile nuclides is addressed.

INTRODUCTION

The discovery[1,2] of intermediate structure in neutron-induced fission of certain fissionable isotopes provided strong confirmation of the importance of shell corrections leading to a double-humped fission barrier.[3] In the case of fission occurring at excitations below the top of both humps in the barrier (subthreshold fission), the structure is quite pronounced. The problem was treated theoretically by Weigmann[4]

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and Lynn,[5] who provided a model of the fission process and a formalism to describe the structure. It is usually assumed that one can represent motion in the fission degree of freedom by the traversing of a two-humped one-dimensional potential barrier: the fissioning nucleus initially finds itself in one of the states in the first well (Class I states); these are coupled through the first barrier to states in the second well (Class II states), which are coupled to the continuum through the second barrier B. The fission components of the wave function correspond to the vibrations in the first and second wells. Class I states are expected to show relatively large neutron widths and small fission widths, while Class II states show small neutron widths and large fission widths. The coupling between the two gives rise to intermediate structure. Analysis of subthreshold fission data[6] has been carried out to provide information on the shape of the barrier: The magnitude of the fluctuations is related to the barrier coupling parameters, and the level density of the Class II or intermediate structure states allows one to infer the effective excitation in the second well. In this connection, the significant advantages of a polarized-neutron and polarized-target measurement in studying intermediate structure should be noted. For (^{237}Np + n), Keyworth et al.[7] showed that each resonance belonging to an intermediate-structure clump has the same spin. This implies that such polarization measurements can be used as an additional tool to reveal nonstatistical behavior.

EVIDENCE FOR INTERMEDIATE STRUCTURE IN FISSILE NUCLEI

Shortly after the discovery of intermediate structure in subthreshold fission, nonstatistical behavior was reported in the fission cross sections of fissile target nuclides, in particular ^{239}Pu and ^{235}U. The evidence was far less conclusive than that for the subthreshold-fissionable target species, however. This is primarily because the noise level of fluctuations arising from the expected statistical processes is relatively much larger for fissile targets. Certain of the statistical tests (e.g., the use of serial correlations that had seemed to indicate nonstatistical behavior) were found to be subject to misinterpretation because of end effects and finite sample size.[8] Only for (^{239}Pu + n) in the 1+ state (which is very nearly a case of subthreshold fission) was the evidence[9] for nonstatistical structure in fission strong enough to be generally accepted. Even here, the evidence is clouded because of the very marked qualitative difference in the sizes of the average 0+ and 1+ fission widths and the strong probability that spin assignments are not completely correct.[10]

For (^{235}U + n), a considerable amount of structure was observed in the fission cross section,[11-13] but total cross section measurements[14] showed that much of this structure could be explained as fluctuations in the entrance channel, and these were generally assumed to be of statistical origin. The question is not to be answered as simply as in the case of subthreshold-fissioning species; it is required that one extract the average fission widths. More nearly complete data are needed: either the total, scattering, or absorption (capture) cross sections in addition to fission.

One must also consider resolution effects. Dennis et al.[15] noted that tests based on runs distributions[16] and lengths of runs[17] are to be questioned if the energy step size is comparable to the coherence width (in unresolved resonance data analysis, the significant coherence width is the resolution width). We have carried out Monte Carlo simulation tests to study this effect on the Wald-Wolfowitz runs distribution test as it has been applied to unresolved resonance data on ^{235}U and ^{238}U. We find that if the bin size for averaging is comparable to the resolution width, the test results can be seriously compromised. However, if the resolution width is small compared to the averaging bin width (< 10%), the test results are approximately valid. For most of the data on ^{235}U and ^{238}U, the latter situation is the one that obtains.

Several studies[18-21] have suggested that the structure observed in $\langle\Gamma_f\rangle$ for (^{235}U + n) is nonstatistical, and again polarization measurements[22] showed that each of the anomalous structures has definite spin. Beer and Kappeler[23] recently analyzed the structure in $\langle\Gamma_f\rangle$ for (^{235}U + n) to infer properties of the deformation potential, under the assumption that the intermediate structure reflects the level spacing in the second well.

INTERMEDIATE STRUCTURE IN RADIATIVE CAPTURE BY ^{238}U

In 1975, Perez et al.[24] reported evidence for intermediate structure in the radiative capture cross section of ^{238}U. Recently, these and additional data obtained by a different experimental technique, but which showed the same nonstatistical behavior, were analyzed by Perez et al.[25,26] under the assumption that the structure could be attributed to doorway states in the $p^{3/2}$ neutron channel.

The conclusions reached have far-reaching implications. If intermediate structure exists and is important in the entrance neutron channel in (^{238}U + n), then, following Müller and Rohr[27] and Kerouac,[28] it should be taken into account for all the actinides, including the fissile species. The analysis[2?] of the polarized-neutron and polarized-target measurements on the fission of (^{235}U + n) did suggest a possibility of intermediate structure in the average reduced neutron widths, in that the fluctuations were found to be larger than expected, and the average fission and reduced neutron widths appear to be correlated, rather than slightly anticorrelated as expected from the models used to explain intermediate structure in subthreshold fission. It has long been recognized that correlated widths imply common doorway states: for fissile nuclei it might imply that the vibrational levels in the fission degree of freedom and certain of the few-quasiparticle or particle-vibration levels constituting entrance channel doorways have a large overlap. At subbarrier energies these first-well vibrations are ineffective as fission doorways, but at energies above the first barrier, this is not necessarily so; the large vibrational amplitudes in the first well carried by such a doorway may allow the possibility of overlap with entrance channel doorways, leading to correlated widths. According to such a model, the fission cross section would have a pre-equilibrium component.

PARITY DETERMINATION OF THE STRUCTURE IN (^{238}U + n) RADIATIVE CAPTURE

Noting that all the lowest-lying levels in ^{239}U have even parity, Corvi et al.[29] suggested that one could measure the intensity of primary transitions feeding these levels relative to transitions to all levels, and deduce the parity of p-wave resonances in (^{238}U + n), using the property that E1 transitions are on the average much more intense than M1 and E2. Corvi's method was used successfully in assigning 57 resonances as p-wave. The method cannot be used for assigning all resonances simply because of Porter-Thomas fluctuations in the partial widths for the few most energetic primary transitions. (Only two such transitions are possible for $p^{1/2}$ resonances, and four for $p^{3/2}$ resonances.)

For a determination of the parity of the intermediate structure reported by Perez et al.,[24-26] Corvi's method does not suffer from this problem. In a typical 400 eV energy bin, there are about twenty $s^{1/2}$ and $p^{1/2}$ resonances, and about forty $p^{3/2}$. If the structure is due to p-wave resonances in which the highest energy primary transitions occur with their expected intensity, the method should give a reliable estimate of the relative p-wave contribution. (One estimates the variance as 2/40 for $p^{1/2}$, 2/160 for $p^{3/2}$).

We used the method devised by Corvi to assign the parity of the structure reported by Perez et al. in two separate runs at the electron linear accelerator laboratory at the Central Bureau for Nuclear Measurements at Geel, Belgium.[30] The first run confirmed all the structure of Perez et al. and suggested that at the lowest energies, the most prominent peak at ~ 13.5 keV does not show the characteristic p-wave signature. The second run confirmed the results of the first, and gave the parity assignments shown in Fig. 1. The most prominent peaks below 50 keV appear to be due primarily to interactions that do not involve the highest energy transitions. We infer that these are s-wave interactions.

The mechanism for radiative capture in the energy region below 45 keV is rather different for s- and for p-wave neutrons. For p-wave neutrons, which account for roughly 2/3 of the capture, the radiation width is rather larger than the neutron width, and the cross section for radiative capture is roughly proportional to the neutron width. The s-wave neutron interactions are dominated mostly by elastic scattering. The radiation width is generally small compared to the neutron width; capture is roughly proportional to the radiation width, and the amount of capture that occurs is nearly independent of the neutron width. (Cf. Table II of the following section.) If s-wave neutron interactions are primarily responsible for the prominent observed intermediate structure resonances below 40 keV, it would appear that at least some of the observed variation is due to the radiation width, and possibly to correlations between the average neutron and radiation widths.

A SENSITIVITY ANALYSIS

For many years, one of the integral tests applied to check the adequacy of resolved and unresolved resonance evaluations of ^{235}U and

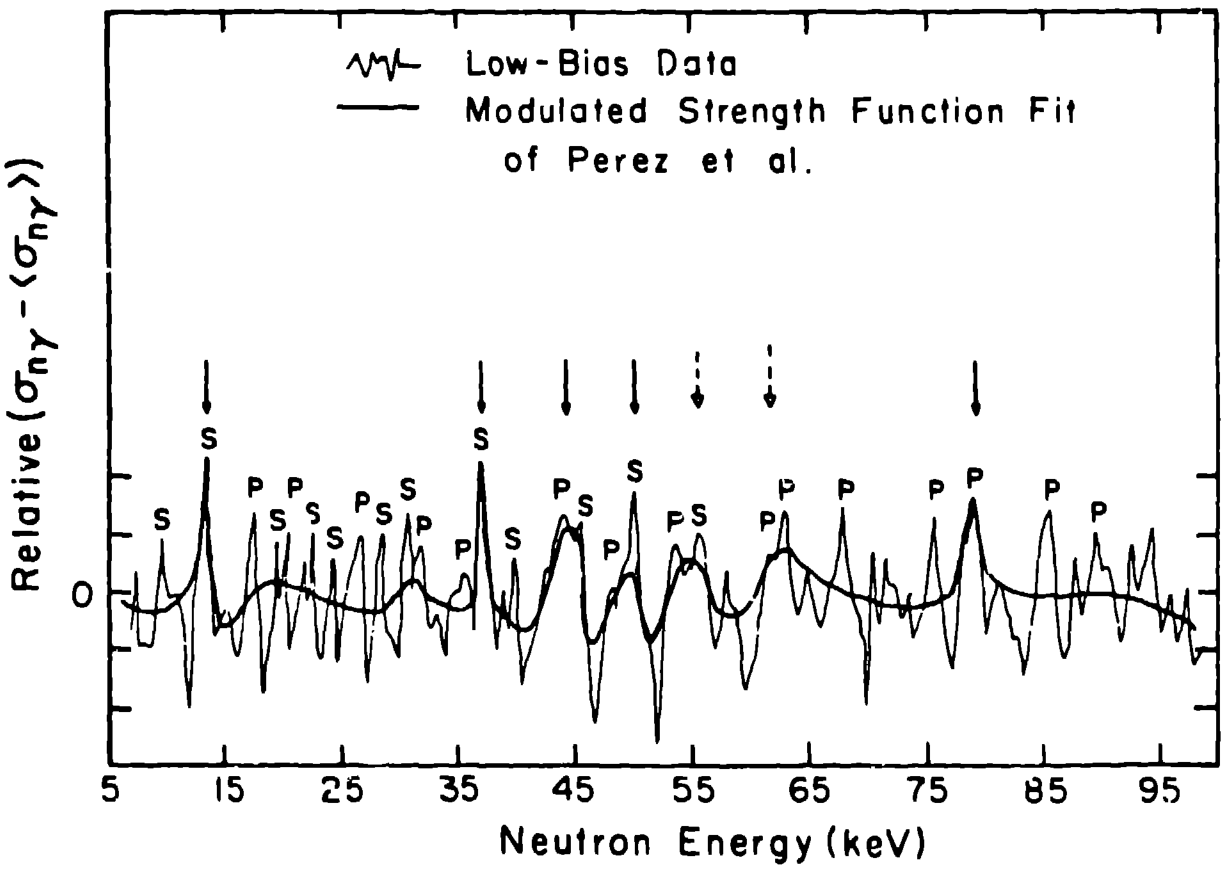

Fig. 1 Parity assignments of relative fluctuations in the capture cross section of (^{238}U + n). The fluctuations were determined by binning the data in 400 eV bins, and subtracting from each point a

^{239}Pu was to calculate the Bramblett-Czirr experiment,[31,32] in which resonance-self-shielded fission rates were measured as a function of absorber thickness in broad energy bins to neutron energies of a few keV. The first attempts to calculate the experiment were done to check the ENDF/B-III evaluation;[33,34] the results suggested that the ^{239}Pu evaluation could be considered to be satisfactory, but that up to 40% discrepancies existed between calculation and experiment for ^{235}U. Subsequent modified evaluations for ^{235}U gave little improvement, but recently Czirr[35] cleared up the discrepancy in a remeasurement of the experiment, concluding that the earlier results were not correct.

The Bramblett-Czirr experiment is remarkably sensitive to small changes in resonance absorption, and we have chosen to carry out a sensitivity study of the effects of varying the data sets of ^{235}U and ^{238}U by calculating a hypothetical Bramblett-Czirr measurement at three energies for ^{235}U (0.2, 2, and 20 keV) and at two energies for ^{238}U (13.5 and 37 keV). The calculation used an R-function representation (neglecting fission interference) with the basic average parameters shown in Table I. Individual resonance parameters for the ^{235}U study were selected by Monte Carlo sampling, using a Wigner distribution for the spacing and a Porter-Thomas distribution for the neutron and partial fission channels. The radiative widths were taken as constant. For the ^{238}U study, neutron widths were selected by Monte Carlo for two of the three channels studied and uniformly over the Porter-Thomas distribution for the third, to reduce the number of calculations required. Each of the variables studied was varied on the average by 10%. The results are given in Tables II and III. As expected, we found that the radiative capture cross section of ^{238}U is not sensitive to changes in $\langle \Gamma^0_n \rangle$; even a factor of two change leads to only a few percent in the capture cross section. However, if the s-wave neutron and radiative capture widths are correlated, one can expect changes of the size attributed by Perez et al. to intermediate structure. The effect of width correlations on the ^{235}U fission and capture cross sections is very pronounced. We find that if 10% of the fission width is neutron-width correlated for each resonance, this is equivalent to increasing the fission width by ~ 80%, decreasing the radiation width by ~ 10%, and gives rather different behavior in a strongly self-shielded environment.

It should be noted that similar strong effects can be expected in ^{238}U and other fertile materials above the threshold for inelastic scattering. Here one should observe an apparent enhancement of the inelastic width and decrease of the radiation width if the neutron elastic and inelastic widths are partially correlated.

SUMMARY AND CONCLUSIONS

Intermediate structure in the subthreshold fission resonances of fertile nuclides has been known for many years, and is thought to be well

TABLE I. Average Resonance Parameters Used in the Calculation of Temperature Dependent Resonance Self-Shielding Reaction Rates for (^{235}U + n) and (^{238}U + n) in the Unresolved Resonance Region.

Target	Resonance Spin (•10^{-4})	Strength Function (eV)	<D> (eV)	$\langle\Gamma_{f1}\rangle$ (eV)	$\langle\Gamma_{f2}\rangle$ (eV)	$\langle\Gamma\gamma\rangle$ (eV)	ka (•10^{-3})
^{235}U	3-	0.968	0.953	0.090	0.090	0.035	2.093
	4-	0.968	0.809	0.069	0.022	0.035	2.093
^{238}U	1/2+	1.134	20.9	-	-	0.0229	1.823
	1/2-	1.7	20.9	-	-	0.0229	1.823
	3/2-	1.7	11.1	-	-	0.0229	1.823

understood in terms of vibrational states in a double-humped potential barrier. This interpretation of intermediate structure in fission suggests that there should be a weak negative correlation of the apparent average fission width and the average neutron width; in the case of fissile, or suprathreshold fissioning species, this correlation should be so small as to be undetectible. The question of a possible correlation of neutron and radiation widths for fertile nuclides has also been considered over the years. It has generally been concluded that there is no reason to expect these widths to be correlated, even though a significant positive correlation is known to exist in the s-wave parameters for the resolved resonances in (^{238}U + n). Recently Perez et al.[25] have reported strong evidence for intermediate structure in the radiative capture cross section of ^{238}U in the energy region below 100 keV, and have fitted the structure with a doorway state model involving p-wave neutrons. Such a mechanism could lead to partial width correlations. Intermediate structure has also been reported in the fission cross sections of fissile nuclei, and in (^{235}U + n), the neutron and fission widths appear to show evidence of a positive correlation, suggestive of a common doorway and of a mechanism different from that for subthreshold fission.

Certain of the statistical tests that have been used to show the presence of intermediate structure, in particular the Wald-Wolfowitz runs and correlation tests, are known to be compromised when applied to unresolved resonance data analysis because of finite resolution in the measurements. We have studied this effect, and conclude that for most of

Table II. Resonance-self-shielded capture cross sections of ^{238}U at 13.5 and 37 keV calculated from the average parameters of Table I, at an assumed temperature of 300 K, and percent changes resulting from the following: A) multiplying the average s-wave radiation width by a factor of 1.1; B) multiplying the average s-wave neutron width by a factor of 1.1; C) multiplying both s-wave neutron and radiation widths by a factor of 1.1, and allowing 10% of the radiation width to be correlated; D) multiplying the average s-wave neutron width by a factor of 2, E) multiplying the average p 1/2 neutron width by a factor of 1.1; F) multiplying the average p 3/2 neutron width by a factor of 1.1; G) multiplying the average p 1/2 radiation width by a factor of 1.1; H) multiplying the average p 3/2 radiation width by a factor of 1.1; and J) multiplying the neutron and radiation widths for all p-wave levels by a factor of 1.1 with the increase in Γ_γ correlated to Γ_n.

E_n = 13.5 keV:

Absorber							
Thickness (g/cm^2):		0.0	0.48	1.86	3.64	8.94	17.96
$<\sigma_{n,\gamma}>$ (b):		0.666	0.632	0.546	0.453	0.262	0.106
% change	A:	+3.2	+3.2	+3.1	+3.0	+2.8	+2.5
	B:	+0.6	+0.5	+0.3	+0.1	−0.2	−0.6
	C:	+5.3	+5.2	+4.7	+4.1	+3.0	+1.8
	D:	+3.1	2.6	+1.0	−0.6	−4.0	−6.8
	E:	+0.7	+0.7	+0.7	+0.6	+0.6	+0.6
	F:	+2.5	+2.5	+2.5	+2.5	+2.4	+2.4
	G:	+1.3	+1.2	+1.2	+1.2	+1.2	+1.2
	H:	+ .9	+2.9	+2.9	+2.9	+2.8	+2.8
	J:	+6.9	+6.8	+6.5	+6.3	+5.7	+5.6

E_n = 37 keV:

Absorber							
Thickness (g/cm^2)		0.0	0.48	1.86	3.64	8.94	17.96
$<\sigma_{n,\gamma}>$ (b):		0.388	0.372	0.327	0.276	0.168	0.073
% change	A:	+2.0	+2.0	+2.0	+2.0	+1.9	+1.9
	B:	+0.2	+0.2	+0.1	+0.1	−0.1	−0.4
	C:	+3.0	+2.9	+2.8	+2.6	+2.3	+1.8
	D:	+1.6	+1.4	+0.8	0.0	−1.7	−3.7
	E:	+0.5	+0.5	+0.5	+0.5	+0.4	+0.4
	F:	+1.9	+1.9	+1.9	+1.9	+1.8	+1.6
	G:	+0.7	+0.7	+0.7	+0.7	+0.7	+0.7
	H:	+1.2	+1.2	+1.2	+1.2	+1.2	+1.2
	J:	+6.7	+6.6	+6.5	+6.3	+6.0	+5.9

Table IIIa. Typical calculated resonance-self-shielded average fission and capture cross sections of ^{235}U at 300 K, as a function of absorber thickness, for neutron energies of 0.2, 2, and 20 keV, using the average parameters of Table I.

Absorber Thickness (g/cm^2)	Energy = 0.2 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)	Energy = 2. keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)	Energy = 20 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)
0.0	22.43	13.78	6.81	3.99	1.86	1.01
0.47	20.75	12.59	6.61	3.87	1.83	1.00
1.83	16.63	9.77	6.05	3.54	1.74	0.94
3.59	12.70	7.19	5.40	3.15	1.62	0.88
8.83	6.19	3.22	3.86	2.25	1.33	0.72
17.74	2.25	1.06	2.20	1.28	0.94	0.51

Table IIIb. Calculation with the same parameters as part a, with the exception that 10% of the fission width is correlated with the neutron width.

Absorber Thickness (g/cm^2)	Energy = 0.2 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)	Energy = 2. keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)	Energy = 20 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)
0.0	26.78	9.87	8.14	3.01	2.24	0.83
0.47	24.73	9.10	7.89	2.92	2.20	0.81
1.83	19.75	7.21	7.22	2.67	2.09	0.77
3.59	14.99	5.43	6.45	2.39	1.95	0.72
8.83	7.17	2.55	4.60	1.71	1.60	0.59
17.74	2.51	0.87	2.61	0.98	1.13	0.42

Table IIIc. Calculation with the same parameters as part a, with the exception that Γ_f is multiplied by a factor of 1.8 and Γ_γ by a factor of 0.9.

Absorber Thickness (g/cm^2)	Energy = 0.2 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)	Energy = 2 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)	Energy = 20 keV $\langle\sigma_f\rangle$ (b)	$\langle\sigma_c\rangle$ (b)
0.0	26.12	10.14	8.02	2.92	2.27	0.75
0.47	24.21	9.25	7.78	2.83	2.23	0.73
1.83	19.54	7.16	7.13	2.59	2.11	0.70
3.59	15.02	5.25	7.37	2.31	1.97	0.65
8.83	7.41	2.33	4.56	1.65	1.61	0.53
17.74	2.68	0.75	2.60	0.93	1.14	0.38

the energy range for which the intermediate structure has been observed in ^{235}U and ^{238}U cross sections, the test results are valid. We have also carried out a study to assign the parity of the intermediate structure in $^{238}U(n,\gamma)$, using the method devised by Corvi et al.[29] for assigning resolved resonances in ^{238}U and ^{232}Th as s- or p-wave. Preliminary results of this study suggest that some of the intermediate structure reported by Perez et al. appears to be due to s-wave neutron absorption. If this is the case, then one must allow for the possibility that the observed fluctuations are due to fluctuations in the radiation width, which is considerably smaller than the average neutron width.

Finally, we address the practical effect of the possible presence of width correlations in fertile and fissile nuclides. Clearly, the usual unresolved resonance treatment with width-fluctuation corrections is not adequate, because in the usual treatment it is explicitly assumed that all partial widths are uncorrelated, and only the neutron width autocorrelation is taken into account in calculating the fluctuation correction. It is also clear that for an infinitely dilute configuration, present methods give correct results, because the changes in the effective cross sections that may arise from width correlations can easily be compensated by changes in the average partial widths. It is the distribution of cross section values in a probability table that will be wrong, not the averages, and this becomes important only in a strongly self-shielded configuration. It may also be noted that the existence of intermediate structure increases the uncertainty of extrapolating unresolved resonance parameters in cases where insufficient data exist.

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