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Parity Assignment of the Pronounced Structure in the Radiative Capture of Neutrons by ^{238}U Below 100 keV.

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Abstract

Some years ago, Perez and de Saussure reported evidence for intermediate structure in the radiative capture cross section of ^{238}U. More recently, these and additional data, obtained by a different experimental technique but which showed the same non-statistical behavior, were analyzed by Perez et al. under the assumption that the structure could be attributed to doorway states in the $p^{3/2}$ neutron channel. In the present paper, we report the results of an experimental determination of the parity of the structure, using neutron capture-gamma ray spectroscopy. We find that much of the structure below 50 keV appears to be due to s-wave interactions. The magnitude of the fluctuations is much larger than can be calculated with the usual unresolved - resonance treatment unless the average neutron and radiative-capture widths are correlated. We show that such an apparent correlation can arise as a result of multiple-scattering enhancement of radiative capture in the samples used, and conclude that the evidence for intermediate structure in the capture of neutrons by ^{238}U is not yet firmly established.

I. Introduction

At the conference on Nuclear Cross Sections and Technology in 1975, Perez and de Saussure[1] noted that there are pronounced fluctuations in the average radiative capture cross section of ^{238}U that are much larger than would be expected from Porter-Thomas fluctuations in the neutron widths. They suggested that these fluctuations might constitute evidence for intermediate structure. In 1979, Perez et al.[2] addressed the problem in more detail. They incorporated additional data by Macklin, obtained by a different experimental technique[3] but which showed the same fluctuations, and applied a number of statistical tests that indicated the existence of non-statistical behavior. They then showed that a modulated strength function in the $p^{1/2}$ neutron channel could provide an explanation of the structure. These results have far-reaching implications. If intermediate structure exists and is important for (^{238}U + n), then, following Müller and Rohr[4] and Kerouac,[5] it should be taken into account for all the actinides, including the fissile species. If the structure is due to doorway states, then the channel-capture mechanism of Lane and Lynn[6] suggests that the neutron and radiative-capture widths may be correlated. It is thus of interest to establish the properties of the structure, and in particular to answer the following two questions: 1) Is the structure due to p-wave interactions, which are responsible for about 2/3 of the capture at 40 keV? 2) Does the structure imply correlated widths?

We addressed this second question as part of a study of practical implications of intermediate structure in ^{235}U and ^{238}U.[7] We concluded that, using the usual statistical treatment of unresolved resonances, the structure in ^{238}U capture seems to require that the neutron and radiative-capture widths be correlated. However, in the present study, we show that such an apparent correlation may be due to the inadequacy of multiple-scattering corrections to the data.

II. Experimental Method and Analysis

Noting that all the lowest-lying levels in ^{239}U have even parity, Corvi et al.[8] 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, 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 (GELINA) at the Central Bureau for Nuclear Measurements at Geel, Belgium. Both runs were carried out under rather similar experimental conditions: a sample of ^{238}metal 2mm thick (changed to 3mm thick in the second run) was placed in the neutron beam at a flight path of 30m. The sample was viewed by a 7 x 6 in D NaI(Tl) gamma-ray spectrometer placed 20-30 cm away from the sample, outside the neutron beam, and shielded from scattered neutrons by at least 10 cm of borated polyethylene and borated paraffin. Capture gamma spectra, appropriately binned, were collected as a function of neutron time of flight. The spectra that were analyzed consisted of three gamma-ray energy bins: 1.5 - 3.5 MeV (representative of transitions from resonances of both parities), 4.3 - 5.2 MeV (representative of transitions from p-wave structure), and > 5.2 MeV (to give the scattered-neutron background.)

The scattered-neutron background was found to be essentially featureless. A fluctuation analysis of the other data was carried out by binning the time-of-flight spectra in 400 eV bins, and subtracting from each point a running eleven-bin average. Parity assignments were made by the method of Corvi et al. by comparing the relative intensity of primary transitions to levels near the ground state of ^{239}U. The first run was a relatively short survey run; the second was done to obtain improved statistical accuracy.

As a further check, we were able to confirm most of the parity assignments made by Corvi et al. in the resonance region as shown in Table I, even though the present data appear to be statistically inferior to those obtained earlier (because the sample is so much thinner). We were also able to extend the range of resolved resonances over which parity assignments could be made to 4 keV.

In the region between 5 and 100 keV, the first run confirmed all the structure of Perez et al. and suggested that at the lowest energies, the most prominent peak at $\sim$ 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.

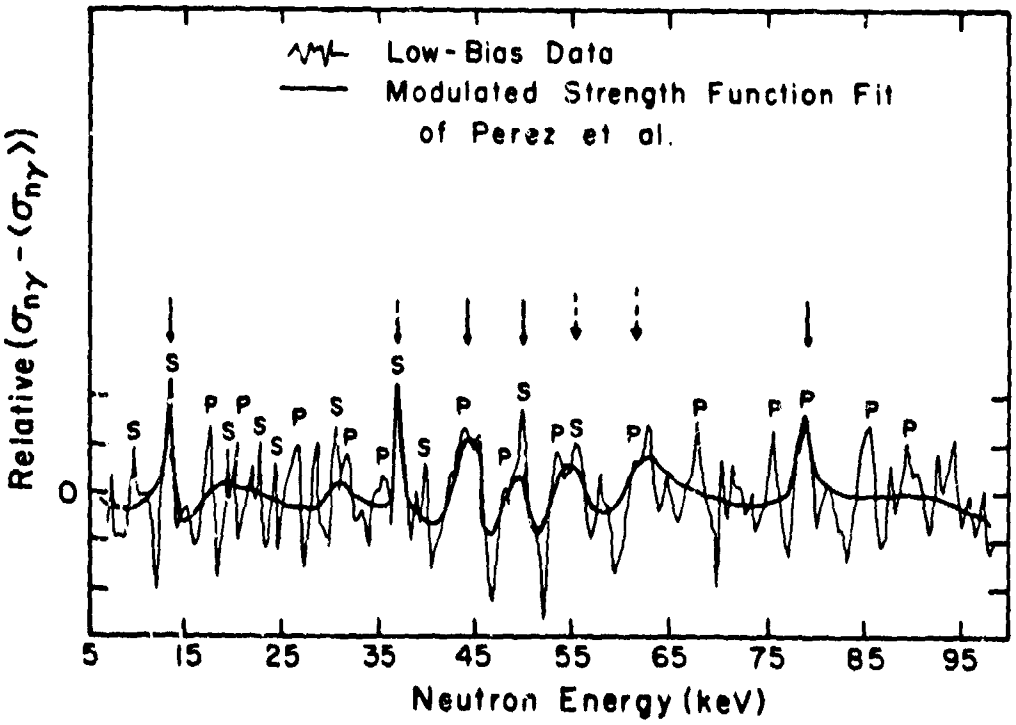

Fig. 1 Parity assignments of relative fluctuations in the capture cross section of (^{238}U + n). The fluctuations were determined by binning the data in 400 eV bins, and subtracting from each point a running eleven-bin average. Parity assignments were made by the method of Corvi et al. by the relative intensity of primary transitions to levels near the ground state of ^{239}U as a p-wave signature. The smooth curve shows a schematic representation of the intermediate structure proposed by Perez et al. described in the text.

III. Multiple Scattering Enhancement of the Capture Yield

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. We carried out a series of calculations of the observed fluctuations,[7] as noted above, using the prescribed unresolved resonance treatment of ENDF/B that allows an energy-dependent average neutron width but assumes a Porter-Thomas distribution within the averaging interval. We found that there is no way to obtain a consistent fit to the fluctuations in the capture cross section and to the variation in the total cross section measured by Olsen et al.,[9] unless the neutron and radiative-capture widths are correlated. Such an apparent correlation could arise from a purely experimental effect: multiple scattering enhancement of the observed capture, which is particularly important for s-wave resonances that are strongly asymmetric in scattering.

It should be noted that multiple scattering corrections in the resolved and unresolved resonance regions are formally somewhat different. In the resolved region, one uses initial values of the resonance parameters to calculate energy-dependent Doppler-broadened cross sections from which the relative interaction probabilities can be calculated as a function of energy and scattering angle, the final resonance parameters being determined by an iterative process. In the unresolved range, a Porter-Thomas distribution of neutron width about the average is generally assumed, and the width-fluctuation-corrected interaction probabilities are used to calculate a multiple-scattering correction that varies smoothly with neutron energy.

In order to determine whether these differences in approach give significantly different estimates for the multiple-scattering enhancement, we chose one particularly strong s-wave resonance clump, that at 37 keV, for further study. The high-resolution total-cross-section measurements of Olsen et al.[9] appear to confirm our conclusion that this region is dominated by several particularly strong s-wave resonances. We carried out an R-function fit to the region between 36.5 and 38.0 keV, using the MULTI code developed by Auchampaugh,[10] in order to obtain a set of typical resonance parameters that would describe the data. The typical parameters are listed in Table II. The fits we obtained are shown in Figs. 2 and 3. Fig. 2 gives the fit to the total cross section of Olsen et al. as measured; Fig. 3 shows the 300K Doppler broadened cross section that is appropriate for the multiple scattering calculations.

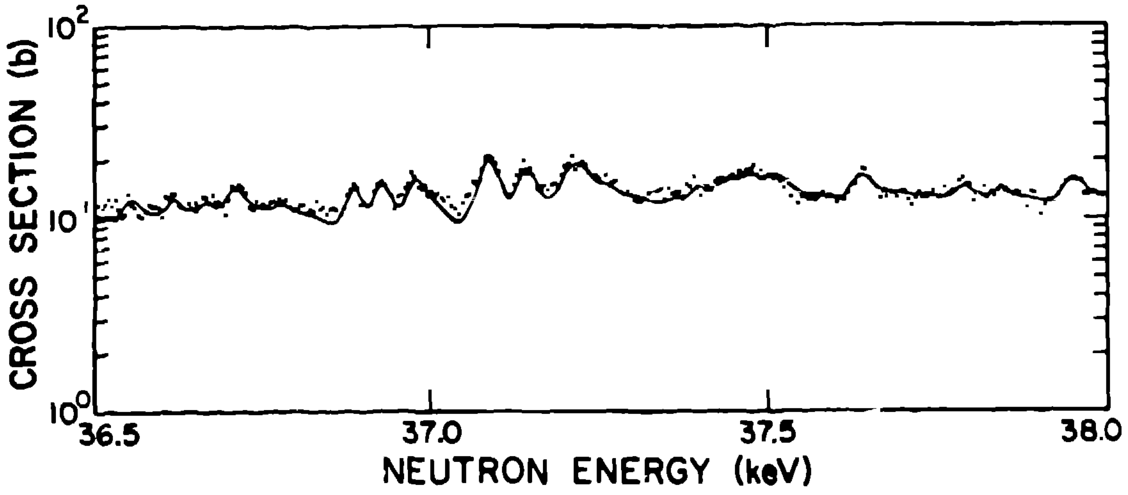

Fig. 2 Representation of the total cross section of (^{238}U + n) measured by Olsen et al. between 36.5 and 38.0 keV. The smooth curve is a resolution-broadened least-squares R-function fit to the data, which gave the parameters listed in Table II.

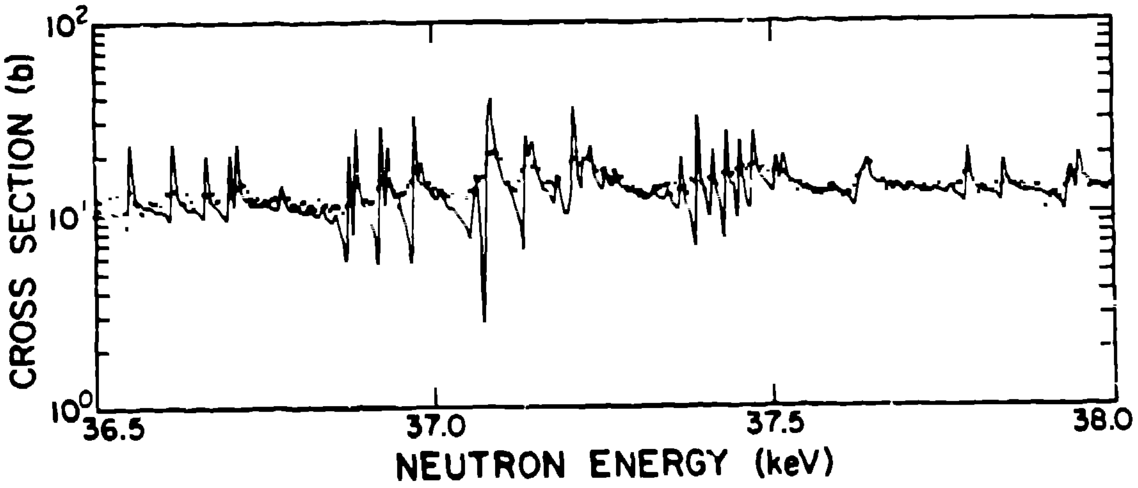

Fig. 3 Representation of the total cross section of (^{238}U + n) measured by Olsen et al. between 36.5 and 38.0 keV. The smooth curve shows the R-function calculated energy dependence that would be observed with perfect resolution using the parameters of Table II.

We then carried out a calculation of the energy-dependent capture yield in this energy region using a hybrid code in which the "resolved-resonance parameters" of Table II were used to describe the s-wave interactions and the usual unresolved resonance treatment described above was used for p-wave interactions. In this calculation, the capture yield with and without multiple-scattering enhancement was tabulated, in order to determine the magnitude of the effect, for various sample thicknesses used in the measurements.

The results of this exercise showed that a surprisingly strong energy dependence of the multiple-scattering enhancement can be expected. One of the data sets considered by Perez et al.[1,2] is that of de Saussure et al.,[11] who calculated a correction of 3.8% for multiple scattering and self screening for their thickest sample between 30 and 40 keV. This is in good agreement with the value we obtain at energies far away from the 37 keV clump, yet within the clump, the calculated multiple-scattering enhancement of the capture yield can be as large as 10% for this same sample thickness. For the 2-mm thick sample we used in the parity determination, the calculated energy-dependent multiple-scattering enhancement within the 37 keV clump ranges from 16 to 24%, compared to ∿ 10% at energies away from the clump.

While the results of this study do not preclude the existence of intermediate structure in the capture cross section of ^{238}U, they do suggest that further study may be needed. Before one applies statistical tests to determine whether the magnitude of the fluctuations is outside the range expected from statistical theory, either an improved multiple-scattering treatment is required, or the data used should have been obtained only with samples so thin that this effect is negligible.

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Table I. Energies, in eV, of resolved resonances in ($^{238}U + n$) assigned as p-wave from the present study and from Corvi et al. (shown with an asterisk). Assignments of Corvi et al. that were not confirmed in the present study are designated by a double asterisk.

63.51**
83.68*
89.29*
98.17
124.98*

158.95
200.71*
203.11*
214.85*
218.32*

224.97
242.71*
253.89*
263.93*
275.11*

282.43*
322.86
337.25*
351.86*
372.84*

439.74*
448.36
498.88*
523.33*
542.71*

550.98*
556.24*
560.12
584.46
615.75*

624.20*
668.41*
677.74*
698.21*
710.59*

713.77*
732.46*
743.14
779.31*
787.33*

828.75*
940.94
964.45*
977.36**
1029.1*

1047.3*
1067.7*
1074.1*
1081.7*
1095.2*

1102.9*
1131.4*
1152.7*
1155.1*
1184.8

1201.4
1219.9*
1230.1*
1252.0**
1277.0*

1289.3
1317.0*
1332.0**
1387.1**
1417.5*

1454.8
1486.8
1510.6*
1534.9*
1550.6**

1568.5
1672.7
1745.7
1768.6
1797.5

1834.2
1893.9
1925.4
1990.0
2000.7

2049.0
2063.3
2071.4
2215.4
2294.0

2296.5
2397.8
2401.8
2527.1
2606.6

2658.6
2682.8
2945.3
3043.8
3072.3

3081.1
3169.8
3264.1
3267.5
3341.2

3378.3
3383.8
3522.3
3528.5
3636.9

3654.2
3683.2
3724.7
3791.1
3809.2

3825.7
3927.9

Table II. A typical set of "resolved-resonance parameters" that can be used to describe the unresolved s-wave structure in the total cross section of (^{238}U + n) near 37 keV. The radiative capture width is assumed to be 0.02 eV for all resonances.

E (keV)	Γ (eV)	E (keV)	Γ (eV)	E (keV)	Γ (eV)	E (keV)	Γ (eV)
36.510	0.004	36.910	0.004	37.222	0.555	37.605	0.004
36.530	0.004	36.920	0.941	37.232	0.105	37.625	0.445
36.550	0.462	36.930	0.512	37.250	0.171	37.635	0.527
36.570	0.004	36.947	0.004	37.265	0.135	37.662	0.004
36.590	0.004	36.970	1.332	37.285	0.004	37.682	0.004
36.615	0.375	36.980	0.232	37.305	0.004	37.700	0.040
36.635	0.004	36.994	0.004	37.325	0.004	37.720	0.004
36.665	0.285	37.010	0.040	37.345	0.004	37.740	0.004
36.685	0.004	37.030	0.004	37.365	0.166	37.760	0.004
36.700	0.285	37.060	0.345	37.390	0.932	37.785	0.324
36.710	0.347	37.065	0.206	37.410	0.475	37.805	0.004
36.732	0.004	37.080	2.964	37.430	0.846	37.825	0.004
36.752	0.004	37.090	0.154	37.440	0.077	37.838	0.303
36.777	0.040	37.110	0.004	37.450	0.697	37.868	0.004
36.797	0.004	37.135	1.356	37.470	1.049	37.888	0.004
36.817	0.004	37.145	0.430	37.505	0.241	37.908	0.004
36.837	0.004	37.160	0.004	37.515	0.298	37.933	0.628
36.857	0.004	37.180	0.214	37.545	0.004	37.948	0.418
36.875	0.582	37.198	0.402	37.565	0.004	37.968	0.004
36.885	0.795	37.205	1.062	37.585	0.040	37.988	0.004