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Resolution of the nature of the coupling in subthreshold fission in ^{238}U + n

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The analysis of a recent high-resolution neutron capture measurement at 152 m has provided the first evidence that the strong fission resonance at 721 eV is a class-II resonance. This conclusion is based on the measured capture width of 4.7 ± 0.6 meV, which is considerably smaller than the average capture width of 23.5 meV for the neighboring resonances. Furthermore, after analyzing the fission widths for the 721- and 1211-eV clusters, we conclude for the $J^{\pi} = \frac{1}{2}^{+}$ fission barrier in ^{239}U that the inner barrier is lower than the outer barrier by ~1.5 MeV.

I. INTRODUCTION

The existence of narrow, intermediate structures in the subthreshold region of the neutron-induced fission cross section of ^{238}U is well established.[1–7] The theoretical interpretation of the mechanism involved has been developed independently by Lynn[8] and Weigmann[9] in terms of a double-humped fission barrier. In this model, the two classes of states formed are distinguished by whether the square of the wave function is a maximum in the primary well (class-I states) or in the secondary well (class-II state) of the double-humped barrier. When a class-II state couples to neighboring class-I states through the inner barrier, the fission and capture widths of the resulting resonance depend on the fraction of the class-II component in the wave function for the resonance. We speak of a class-II resonance as one with a large fraction of the class-II wave function. Not only would this resonance have most of the fission width, but its capture width would also be predominately class II in character. Because the secondary well is ~2 MeV shallower than the primary well,[2,10] the capture width of this resonance would be smaller and the gamma-ray spectrum much softer than that for the normal class-I resonances. In ^{239}U, the coupling matrix element is known from experimental data[3,10] to be less than the spacing between the class-I states (D_I), a category described as very weak coupling by Bjørnholm and Lynn.[10] However, within this category there are two possibilities: (1) The transmission through the inner barrier (T_A) is much greater than that through the outer barrier (T_B), a case in which the class-II resonance would be observed in the total cross section, and (2) $T_B \gg T_A$, a case in which the class-II resonance would, most likely, not be observed in the total cross section. With total and fission cross-section data alone, we cannot distinguish between these two possibilities. However, a measurement of a direct property of the secondary well, such as the capture width or gamma-ray spectrum for the class-II resonance, could resolve the ambiguity. To date, the experimental evidence on the gamma-ray spectrum is conflicting. Browne[11] observed a much softer gamma-ray spectrum for the resonance at 721 eV, the purported lowest-energy class-II resonance in ^{239}U, than for the neighboring resonances, whereas Weigmann *et al.*[12] observed no difference in the gamma-ray spectrum for these resonances. Furthermore, until now, the statistical accuracy and resolution of a capture measurement[13] were insufficient to determine the capture width of the 721-eV resonance. This report presents the results of a neutron capture measurement of ^{238}U that confirm the 721.5-eV resonance as a class-II resonance and, therefore, that $T_A \gg T_B$, at least for the $J^{\pi} = 1/2^{+}$ fission barrier.

II. EXPERIMENTAL DETAILS

The neutron capture measurement on ^{238}U was performed at the Oak Ridge Electron Linear Accelerator (ORELA) using the 3000-ℓ ORELAST capture gamma-ray detector.[14] The ORELA was operated at 800 pps and 40 kW with a burst width (full-width at half maximum) of ~50 ns. A 0.64-mm sample of highly enriched ^{238}U (^{238}U isotopic enrichment > 99.99%), at room temperature, was placed in the center of the detector 151.9 m from the neutron source. We hoped that the detector would be sensitive enough to the gamma-ray spectrum that differences could be observed between the 721-eV resonance and the neighboring resonances. For this reason, time-of flight (TOF) spectra were taken at five different gamma-ray biases (B1—B5) corresponding, roughly, to gamma-ray energy intervals of 1.8 to 2.9 MeV, 2.9 to 4.0 MeV, 4.0 to 5.2 MeV, 5.2 to 6.3 MeV, and 6.3 to 8.6 MeV. However, we observed no statistically significant dependence of resonance energy on spectrum shape, probably because of the detector's poor (~70%) gamma-ray energy resolution.

The TOF data were accumulated over a five-day period. The B1 data were not analyzed because the large gamma-ray background obscured any measurable capture in the resonances of interest. Similarly, the B2 data were not included in the final analysis, although some capture could be observed in these resonances. Therefore, the final data consisted of the B3 to B5 data only.

III. ANALYSIS

A. General

The resonances of most interest for the lowest-energy class-II state are the large fission-area ones at 721.5 and 729.9 eV and the one at 730.4 eV, which has no fission area but a similar neutron width. These data are shown in Fig. 1, averaged over two 16 ns TOF channels. The raw areas are converted to capture areas by using those resonances between 650 and 850 eV with neutron widths less than 2 meV and assuming a value of 23.5 meV[13] for the capture width. This neutron width bias is chosen because the correction for multiple-scattering and self-shielding effects to these areas is small and, to first order, such effects cancel out in the normalization procedure.

To determine a capture width when $\Gamma_n \approx \Gamma_n\Gamma_\gamma/\Gamma$, it is important to measure accurately not only the capture area, but also the neutron width. For this reason, the 41- and 155-m total cross-section data[15] taken at ORELA were reanalyzed and particular attention given to the neutron widths for the three resonances (see Table I).

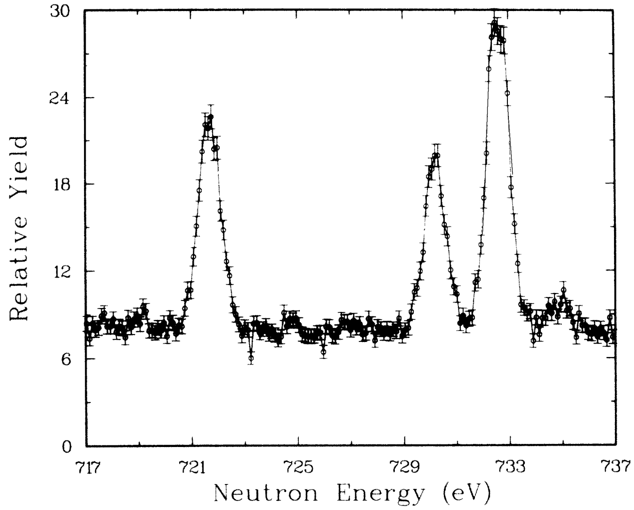

FIG. 1. Relative capture yield. Data have been averaged by two TOF channels. The solid line is a guide to the eye.

B. Capture width for the 721 eV-resonance

The capture width is calculated from the expression $\Gamma_\gamma = \Gamma_n(a_\gamma - \varepsilon a_f) / [\Gamma_n - a_\gamma + a_f(\varepsilon - 1)]$, where $a_f = \Gamma_n\Gamma_f/\Gamma$, $a_\gamma = \Gamma_n(\Gamma_\gamma + \varepsilon\Gamma_f)/\Gamma$, and ε is the contribution to the capture area from the fission process. The exact value of ε for the detector is not known, but it is certainly not zero (for which $\Gamma_\gamma = 6.6$ meV). The detector does not discriminate between fission and capture gamma rays and is probably more sensitive to fission gamma rays than to the low-energy gamma rays expected for the 721-eV resonance. Therefore, ε could be much greater than 1, in which case Γ_γ would be smaller (for example, $\Gamma_\gamma = 3.5$ meV for $\varepsilon = 2$). The capture width for the 721.5-eV resonance is not very sensitive to Γ_n and a_γ because Γ_n/a_γ is much greater than 1 (1.35 ± 0.05). However, for the other two resonances, a_γ is too close to Γ_n to obtain meaningful values for the capture widths. The error on

TABLE I. Parameters for the resonances near 721 eV.

E_r (eV)	Γ_n (meV)	a_γ (meV)	a_f (meV)	Γ_γ (meV)
721.5	1.794 ± 0.034	1.328 ± 0.036	0.1070 ± 0.0013	4.7 ± 0.6[a]
729.9	1.020 ± 0.029	1.019 ± 0.033	0.0063 ± 0.0003	---
732.4	1.985 ± 0.036	1.956 ± 0.043	0	---

[a] ε=1, see text for explanation.

the 721.5-eV capture width includes the uncertainties given in Table I for the other quantities in the expression for Γ_γ.

C. Capture width probability

A capture width of 4.7 meV for the 721-eV resonance implies that either this resonance is a normal class-I resonance with an anomalously small capture width, or that the resonance is special and may, indeed, be a class-II resonance. The probability of observing a capture width of less than Γ_{γ_T} from a normal frequency distribution is given by

$$P(\Gamma_{\gamma_T}) = \frac{1}{\sqrt{2\pi}\,\sigma}\int_{-\infty}^{\Gamma_{\gamma_T}} e^{-(\Gamma_\gamma-\bar{\Gamma}_{\gamma_T})^2/2\sigma^2}\, d\Gamma_\gamma \ . \qquad (1)$$

Using either the experimental value $\sigma = 1.96$ (Ref. 16) for the ^{238}U resonances or the theoretical estimate $\sigma = 0.47$ (Ref. 17) and $\bar{\Gamma}_{\gamma_T} = 23.5$ meV, $P(4.7) > 10^{-22}$. We conclude, therefore, that the 721-eV resonance is not a class-I resonance.

IV. THEORETICAL RADIATION WIDTH

If the 721-eV resonance is a class-II resonance, then is our value of Γ_γ consistent with theoretical estimates for the class-II radiative width? Bjørnholm and Lynn[10] give the following expression for the total capture width of an excited state E^*,

$$\Gamma_{\gamma_T} = Ke^{-E^*}\int_0^{E^*} \varepsilon_\gamma^{\,n}\, e^{(E^*-\varepsilon_\gamma)/\theta}\, d\varepsilon_\gamma$$

$$= K[n!\,\theta^{n+1} + O(e^{-E^*/\theta})] \ . \qquad (2)$$

Equation 2 is based on the assumptions that the gamma-ray strength function is given by the power law, $K\varepsilon_\gamma^{\,n}$, and that the level density is given by the constant temperature model, $C \exp(E^*/\theta)$. At excitation energies of more than a few MeV, the independent-particle model is more appropriate for the level density. However, Eq. 2 can still be used for a rough estimate of the class-II radiation width if the effective temperature is replaced by

$$2\sqrt{\frac{E^*}{a}} \ .$$

Equation 2 allows for a number of possibilities for the radiative width of the states in the primary and secondary wells. If the statistical behavior in the two wells is similar, then θ, n, and K are equal. Ignoring all but the leading term in Eq. 2, the radiative width for states in the secondary well ($\Gamma_{\lambda_{II}(\gamma)}$) is related to the radiative width for the states in the primary well ($\Gamma_{\lambda_I(\gamma)}$) by

$$\Gamma_{\lambda_{II}(\gamma)} = \left(\frac{E-\delta_{II}-E_{II}}{E-\delta_I}\right)^2 \frac{a_I}{a_{II}}\Gamma_{\lambda_I(\gamma)} \ , \qquad (3)$$

where δ_I, δ_{II} are the pairing energies for odd-mass nuclei, E_{II} is the height of the secondary well relative to that of the primary well, and a_I and a_{II} are the level density parameters for the two wells. For E equal to the neutron binding energy, $S_n = 4.807$ MeV (Ref. 18), $\delta_I = \delta_{II} = 0.69$ (Ref. 10), $a_I = a_{II} = 33.4$ MeV^{-1} (Ref. 19), and $E_{II} = 1.9$ MeV (Ref. 10), $\Gamma_{\lambda_{II}(\gamma)} = 6.9$ meV.

If the gamma-ray strength is derived from the tail of the giant resonance, then n may be as large as 5 (Ref. 10). In this case, the class-II radiative width is related to the third power of the ratio of effective excitation energies in the two wells, which results in an even smaller class-II radiative width (3.7 meV).

We have assumed in these estimates that the nuclear temperature θ (which is related to the level density parameter and pairing energy) is the same in both wells. However, it can be argued[20] that a lower temperature (a larger level-density parameter, for a given excitation energy) is indicated for the secondary well (at low-to-moderate effective excitation energies), consequently a smaller $\Gamma_{\lambda_{II}(\gamma)}$. Therefore, our value for the capture width of 4.7 meV is consistent with that for a class-II state in ^{239}U at the neutron-binding energy.

V. PARAMETERS FOR THE CLASS-II STATES AT 724 AND 1206 eV

The fact that we observe the class-II resonance at 721 eV implies that $T_A > T_B$ and, therefore, that the class-II coupling width $\Gamma_{\lambda_{II}(c)}$ is greater than the class-II fission width $\Gamma_{\lambda_{II}(f)}$. Furthermore, the fact that the fission strength is distributed among just a few resonances in the fission cross section indicates that $\Gamma_{\lambda_{II}(c)} \ll D_{II}$ (each class-II state can be considered independently). The eigenvalue problem[8] that describes the coupling ($H_{\lambda_I,\lambda_{II}}$) of a single class-II state ($E_{\lambda_{II}}$) to many class-I states (E_{λ_I}) can be solved exactly if the coupling to the continuum is negligible, which we assume. The required fission widths and resonance energies, excluding the parameters for the 721- and 1211-eV resonances, are shown in Table II (Ref. 3). The fission width for the 721-eV resonance is calculated using $\Gamma_\gamma = 4.7$ meV. The fission width for the 1211-eV resonance is calculated using $\Gamma_\gamma = 6.6$ meV. We arrived at this value iteratively, assuming that the capture width is given by

$$\Gamma_\lambda = f \cdot \Gamma_{\lambda_{II}(\gamma)} + (1-f) \cdot \Gamma_{\lambda_I(\gamma)} \ ,$$

TABLE II. Eigenvalue parameters for the class-II states at 724 eV and 1206 eV.

E_λ (eV)	$\Gamma_{\lambda(f)}$ (μeV)	E_{λ_I} (eV)	$E_{\lambda_{II}}$ (eV)	$H^2_{\lambda_I\lambda_{II}}$ (eV)^2	$\Gamma_{\lambda_{II}(f)}$ (μeV)
595.0	1.02	595.2		27.5	
619.9	0.15	619.9		2.6	
708.3	33.83	709.1		11.7	
721.5	411.69				
			724.1		610.4
729.9	151.67	727.7		14.7	
765.1	7.70	764.6		22.1	
821.6	0.24	821.6		4.0	
851.0	2.68	850.4		78.6	
856.1	1.42	855.7		32.2	
			$\langle H^2_{\lambda_{II}}\rangle_{\lambda_I}$ =	18.4 eV^2	
1140.0	2.35	1140.8		47.7	
1168.0	15.70	1171.6		145.9	
1211.0	174.05				
			1206.6		192.1
			$\langle H^2_{\lambda_{II}}\rangle_{\lambda_I}$ =	72.7 eV^2	

where $f = \Gamma_f/\Gamma_{\lambda_{II}(f)}$, $\Gamma_{\lambda_I(\gamma)} = 23.5$ meV, and $\Gamma_{\lambda_{II}(\gamma)} = 4.7$ meV. The solutions to both the 721- and 1211-eV eigenvalue problems are given in Table II. The average coupling matrix element for each class-II state ($\langle H\mathrm{si}^2_{\lambda_{II}}\rangle_{\lambda_I}$) is calculated from the sum of the individual matrix elements divided by the number of class-I resonances in the interval over which the sum is taken. The average H^2 for both class-II resonances is 45.6 eV^2, which corresponds to a coupling width

$$\Gamma_{\lambda_{II}(c)} = 2\pi H^2/D_I = 11.6\ \mathrm{eV}$$

for $D_I = 24.8$ eV (Ref. 13). The average class-II fission width for the two resonances is 0.401 meV.

VI. RELATIVE BARRIER HEIGHTS

The average coupling and fission widths for the class II states can be related to the heights of the inner and outer barriers, respectively, relative to the neutron binding energy, in the following way:

$$(V_{A,B} - S_n)\Big/\hbar\omega_{A,B} = \frac{1}{2\pi}\ln\left(\frac{D_{II}}{2\pi\Gamma_{\lambda_{II}(c),(f)}} - 1\right). \quad (4)$$

The values from the present results for $D_{II} = 1.0 \pm 0.25$ keV (Ref. 3) are summarized in Table III along with those obtained from fits made to the fast neutron fission cross section by Bjørnholm and Lynn[10] and by B. B. Back *et al.*[21] The striking disagreement between our results and the fitted results is difficult to understand. The spin and parity of the 721-eV resonance, and therefore of the 724-eV class-II state, is known[13] to be $J^\pi = 1/2^+$. The threshold region of the fission cross section should correspond to the opening of the lowest angular momentum barrier, that is, $J^\pi = 1/2^+$. Therefore, the results should be in better agreement. A possible explanation is to postulate a special vibrational or single-particle state at somewhat higher energy that couples its strength to the class-II states. In this case, the class-II fission and coupling widths used in the barrier calculation would be smaller, with a corresponding increase in the rela-

TABLE III. Summary of ^{239}U barrier parameters.

Ref.	$\dfrac{V_A - S_n}{\hbar\,\omega_A}$	$\dfrac{V_B - S_n}{\hbar\,\omega_B}$
10	2.07	2.60
21	1.94	2.30
present	0.40	2.05

tive barrier heights. A strength-function plot of the fission widths given in Table III of Ref. 3 suggests such a state at about 40 keV with a damping width of about 25 keV. However, if we assume Lorentzian coupling to the class-II states, then the special state has very little effect on the class-II parameters. Therefore, we conclude for the $J^\pi = 1/2^+$ barrier that the inner barrier is substantially lower (~ 1.5 MeV) than the outer barrier, in contradiction to the results from fits to the fast fission cross section.

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