Theoretical evaluations of the fission cross section of the 77 eV isomer of ^{235}U

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We have developed models of the fission barrier (barrier heights and transition state spectra) that reproduce reasonably well the measured fission cross section of ^{235}U from a neutron energy of from 1 keV to 2 MeV. From these models we have calculated the fission cross section of the 77 eV isomer of ^{235}U over the same energy range. We find that the ratio of the isomer cross section to that of the ground state lies between about 0.45 and 0.55 at low neutron energies. The cross sections become approximately equal above 1 MeV. The ratio of the neutron capture cross section to the fission cross section for the isomer is predicted to be about a factor of 3 larger for the isomer than for the ground state of ^{235}U at keV neutron energies. We have also calculated the cross section for the population of the isomer by inelastic neutron scattering from the ^{235}U ground state. We find that the isomer is strongly populated, and for $E_n=1$ MeV the $(n,n'\gamma)$ cross section leading to the population of the isomer is of the order of 0.5 b. Thus, neutron reaction network calculations involving the uranium isotopes in a high neutron fluence are likely to be affected by the 77 eV isomer of ^{235}U. With these same models the fission cross sections of ^{233}U and ^{237}U can be reproduced approximately using only minor adjustments to the barrier heights. With the significant lowering of the outer barrier that is expected the general behavior of the fission cross section of ^{239}Pu can also be reproduced.

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I. INTRODUCTION

In environments of very high neutron flux the ultimate yields from the chains of nuclear reactions depend not only on the cross sections of nuclei in their ground states but also, to a lesser or greater degree, on the cross sections of excited states. In stars at high temperatures, the states involved will depend on the Boltzmann distribution of excitation while in more transient situations, such as nuclear explosions, the longer-lived isomeric states will play key roles. Long-lived isomers are difficult to populate electromagnetically in a hot dense plasma. However, they can be populated strongly via neutron capture and, if the neutron energies are high enough, by inelastic neutron scattering. An especially interesting example is that of ^{235}U, which has an isomer at only 77 eV excitation with a half-life of 26 m. The thermal neutron fission cross section of this isomer has been measured [1] and found to be about twice the value of the nucleus in its ground state. This has led to the speculation that the cross section may also be higher for fast neutrons, thus enhancing the fission yield in a transient, extremely high neutron flux. Equally, a lower cross section would diminish the yield. Since there is no method available at present to measure the fast neutron cross section of the isomer, a theoretical evaluation is required to answer this question.

The purpose of this paper is to perform detailed calculations of the fission cross sections for the ground state and isomeric state of ^{235}U. The ground state of ^{235}U has spin and parity $7/2^-$, while the isomer is $1/2^+$. The latter is the same as the ground state of ^{239}Pu, which has a gross fission barrier height (relative to the neutron separation energy) very similar to that of ^{235}U. It is therefore pertinent as part of this investigation to determine the main physics distinguishing the magnitude and energy dependence of the fission cross sections of ^{235}U and ^{239}Pu.

The task involves the assessment of the main features of the double-humped fission barrier (barrier heights and penetrability parameters) from available data relating to the fission of the compound nucleus ^{236}U. These parameters are somewhat dependent on models of the transition states at the barrier peaks. Therefore, a range of models was considered, adjustments made to obtain reasonable agreement with the measured fission cross section of ^{235}U, and calculations made of the corresponding cross section of the isomeric state. For the models investigated, the fission cross section of the isomer is calculated to be substantially lower (by about 50%) than that of the ground state over a significant part of the neutron energy range (from 0 to $\sim$ 0.5 MeV). This work is described in Sec. II.

Using analysis of the same kinds of data as for the uranium isotope, barrier parameters were established for plausible models of the barrier transition states of the compound nucleus ^{240}Pu, and from these parameters the fission cross section of ^{239}Pu was calculated. This was found to agree well with the measured cross section. Similar calculations were carried out for the cross sections of ^{233}U and ^{237}U. These odd-mass neighbors of ^{235}U are expected to have similar barrier properties, while ^{237}U, like ^{239}Pu, has the same spin and parity as the ^{235}U isomer. These studies help give confidence that we have a sound understanding of the barrier and transition state systematics of this whole group of nuclei. This work is described in Sec. III.

II. MODELS AND CALCULATIONS OF THE FISSION CROSS SECTION OF ^{235}U

A. General remarks

1. Cross-section theory

At moderately high excitation energies (up to the order of 10 MeV) Hauser-Feshbach theory [2] is used for calculating cross sections. In this, the cross section is separated into its components of total angular momentum and parity, and each component is proportional to a spin-weighting factor multiplied by the ratio of the product of transmission coefficients T for the entrance and exit channels and the sum over transmission coefficients for all channels. In its more sophisticated form extra factors have to be included to account for the statistical fluctuations of the partial widths of the underlying compound nucleus levels:

$$\sigma_{CC'}=\pi\frac{\lambda^2\sum_{c(e)} g_e T_{c(e)}\sum_{c'(o)} T_{c'(o)}S_{c(e),c'(o)}}{\sum_{c''} T_{c''}}. \qquad (1)$$

Labels $c(e)$ refer to the entrance channel c in its different possible spin combinations, $c'(o)$ to the outgoing channel c' in spin combinations o, λ is the de Broglie wavelength divided by 2π, and $S_{c(e),c'(o)}$ is the fluctuation averaging factor. Dresner [3] has derived a numerical integral that can be used for the evaluation of S in the general case when one channel (or, in practice, a large group of channels such as those accounting for radiative capture) is constant for individual compound nucleus levels while for each of the remaining channels the statistical distribution function over levels is a member of the χ^2 family.

For neutrons with energies up to about 1 MeV bombarding very long-lived and well-studied actinide target nuclei such as ^{235}U, the transmission coefficients for the entrance neutron channel and individual inelastic channels can be calculated with considerable confidence. Alternatively—and this is the procedure adopted here—we can use the experimental information on *s*-wave and *p*-wave neutron strength functions. The radiation widths are known with reasonable accuracy in the slow neutron resonance region and can be extrapolated using simple statistical models of the radiation process. The chief difficulty in applying the Hauser-Feshbach theory to the fissionable nuclei lies in the nature of the fission process.

It is well known that the fission barriers of the actinide group of nuclides are double humped in their functional dependence on deformation. This contrasts with the single hump or maximum in the potential energy given by the liquid-drop model of fission. A double-humped barrier has many consequences on the fission cross section as a result of the subtle interplay of the two maxima and makes analysis of the data a complicated and not always unambiguous one. It is not only in the one variable ("prolate" deformation towards elongation and division) that the potential energy departs from the classical liquid-drop form. It is well established that at the outer barrier of the fission path the nucleus is unstable to octupole deformations and the saddle point here is at a mass-asymmetric shape. There is also strong, though indirect, evidence that at the inner barrier the nucleus is unstable to axial deformations. These different shapes affect the energies of the transition states (also known as Bohr channels, after the introduction of the concept by Bohr [4]), the states of collective and quasiparticle excitation in which the nucleus passes over the barrier saddle point. Once the transition states are established, by either experimental evidence or hypothesis, the transmission coefficients for each barrier can be calculated, and from these we can calculate the overall fission transmission coefficient.

At excitation energies well above the barriers, the fission transmission coefficient T_F has the Strutinsky statistical form [5] that simply relates the overall transmission to the separate transmission coefficients for crossing the inner and outer barriers, T_A, T_B, respectively:

$$T_F=\frac{T_A T_B}{T_A+T_B}. \qquad (2)$$

The coefficients T_A, T_B are given by the Bohr and Wheeler prescriptions of the sum of transition states [6], each multiplied by a barrier penetrability factor:

$$T_A=\sum_f \frac{1}{1+\exp[(V_A+E_{f,A}-E)/\hbar\omega_A]} \qquad (3)$$

(and similarly for T_B). In Eq. (3) the penetrability factor is the Hill-Wheeler formula [7] for a barrier with parabolic form equivalent to an inverted harmonic oscillator with circular frequency ω, E is the excitation energy, V_A is the inner barrier height, and the $E_{f,A}$ are the energies of the transition states f above the inner barrier. A similar equation can be written for the outer barrier. It is possible to define transmission coefficients for individual transition states f:

$$T_f=\frac{T_A T_{B,f}}{(T_A+T_B)}. \qquad (4)$$

These, with their appropriate quantum numbers, are important for calculating cross sections for more specific properties of the fission reaction, such as fission product angular distributions.

At lower energies the intermediate structure due to compound-nucleus-type levels (class-II states [8]) associated with the deformation of the secondary well between the inner and outer barrier peaks must be taken into account. By concentrating the fission strength into narrow energy regions these lower the average fission probability [9]. Also the effect of Porter-Thomas fluctuations both in the fine-structure compound-nucleus levels (class-I levels) and class-II levels must be taken into account. These fluctuation effects are discussed in Ref. [10], where analytic and numerical results have been established for a few limiting cases.

In the present work fluctuation averaging has been studied much more extensively. From the transmission coefficients $T_A, T_{B,f}$ the mean coupling and fission widths $\langle\Gamma_{II(c)}\rangle, \langle\Gamma_{II(f)}\rangle$, respectively, of the class-II levels are obtained:

$$\langle\Gamma_{II(c)}\rangle = D_{II}T_A/2\pi,$$

$$\langle\Gamma_{II(f)}\rangle = D_{II}T_{Bf}/2\pi,$$

where D_{II} is the mean class-II level spacing. Monte Carlo techniques have been used for selection of the parameters (coupling widths, fission widths, and individual spacings) of the individual class-II levels from Porter-Thomas distributions and, using the selected class-II coupling width values, the coupling matrix elements with the class-I levels (for which spacings and reduced neutron widths were also selected using pseudorandom numbers) were drawn from zero-mean Gaussian distributions. Solution of the eigenvalue problem then gave the parameters for the resonance fine structure, from which the detailed cross section could be computed and averaged.

Comparison of these results with the formula [9] based on a uniform picket fence model of the intermediate and fine structure gives the fluctuation averaging factor. The uniform picket fence formula for the fission probability is

$$P_F=\frac{1}{\{1+R^2+2R\coth[\pi(\Gamma_{II(c)}+\Gamma_{II(f)})/D_{II}]\}^{1/2}}, \quad (5)$$

where

$$R=\frac{\Gamma_I(\Gamma_{II(c)}+\Gamma_{II(f)})D_{II}}{\Gamma_{II(c)}\Gamma_{II(f)}D_I} \quad (6)$$

and Γ_I is the mean total width of the class-I levels. When the mean class-II width is much less than the class-II level spacing, Eq. (8) gives a fission probability up to an order of magnitude lower than the value deduced from the Hauser-Feshbach formula with the statistical expression, Eq. (5), for the fission transmission coefficient. Inclusion of the fluctuation averaging factor can reduce the fission probability by up to another factor of 3 of more.

In the case of overlapping intermediate resonances ($T_A+T_B\gg 1$), Eq. (8) gives the statistical result. Even in this case, however, when the intermediate structure is washed out the class-II level fluctuations should be taken into account in the evaluation of the fission transmission coefficients of Eqs. (3) and (5). This is done here using the Dresner numerical integral technique applied to Eq. (5). The individual transition state components of T_B are governed by independent Porter-Thomas distributions. Although the magnitude of T_A is governed by the transition states across the inner barrier, its fluctuation properties are governed by the degree of overlap of the class-II resonances. The frequency distribution is assumed to be a member of the chi-squared family with ν degrees of freedom. The value of ν is evaluated from a picket-fence model of the class-II states. For large T_A the value of ν is $T_A/2$.

The Monte Carlo method for calculating the overall fluctuation averaging factor in the intermediate structure case is too time consuming to apply in a full calculation of the neutron cross sections. However, we find that if we apply the product of the separate fluctuation factors for the fission transmission coefficient, Eq. (5), and for the fine structure in the Hauser-Feshbach formula to the intermediate structure fission probability, Eq. (8), we obtain a quite good approximation to the Monte Carlo result. It is this approximation that we use in general in calculating neutron cross sections, although we have frequently used the more exact Monte Carlo method for calculating the fission probability below the neutron separation energy in the analysis of (d,pf) and (t,pf) data for deduction of fission barrier heights.

2. Fission barrier properties

The overall systematics of the fission barrier parameters of the actinides were established in a review by Bjornholm and Lynn [10]. Since that work, variations on the detailed parameters of specific nuclides have been published by other authors, but our understanding of the broad trends remains unchanged. Inner barrier heights (denoted by V_A) vary little over the range from Th to Cf for a given parity class. For the double-even parity class the barrier height for the uranium isotopes and their neighbors is about 5.5 MeV. For even-odd (or odd-even) and double-odd nuclides the inner barrier is about 0.5 and 1 MeV higher, respectively. Outer barrier heights (denoted by V_B) vary strongly with proton number. For the uranium nuclides they are about the same as the inner barriers, whereas for plutonium they are about 0.5 MeV lower. From a gross point of view the overall barrier heights of the compound nuclei ^{236}U and ^{240}Pu are very similar, as are their neutron separation energies, leading to the simple expectation that the neutron-induced fission cross sections of ^{235}U and ^{239}Pu should be similar. In fact the different relative heights of the inner and outer barriers lead to considerable differences in the cross sections of the two nuclides.

The energies, total angular momenta, and parities I^π of the transition states are as important as the barrier heights in the fission process. These are largely extrapolated from the nuclear spectroscopy known for the ground state deformation. For double-even fissioning nuclides the lowest transition state, at both barriers, is, of course, the "ground state" at the barrier deformation, with excitation energy zero and $I^\pi=0^+$. Built on this is a rotational band with $I^\pi=2^+,4^+,6^+$, etc., with rotational moment of inertia $\mathfrak{I}$ inferred to be about twice, for the inner, and thrice, for the outer barrier, of that of the ground state. Thus, the transition state energies are

$$E_f=E_I=I(I+1)\hbar^2/2\mathfrak{I}, \quad (7)$$

with respect to the barrier height. Above the "ground" transition state there is, in even nuclides, an energy gap, which could be significantly larger than 1 MeV—that is, devoid of quasiparticle excitations. In this energy gap, however, it is expected that there will be collective vibrations each with its own rotational band. The beta vibrations are the best known of these from nuclear spectroscopy, but being vibrations in the prolate deformation variable, which becomes largely the fission degree of freedom, they do not enter into consideration of the transition states.

Apart from the beta vibrations, there is strong spectroscopic evidence for gamma vibrations, which are vibrations about axial symmetry, with spin projection along the prolate deformation symmetry axis and parity $K^\pi=2^+$. The gamma-vibration energy is about 0.8 MeV in the actinides; its energy at the barrier deformations is unknown, and this is one of the quantities that is varied in the modeling process discussed below. In particular, if the nucleus is stable but soft to gamma deformation, then the gamma-vibration energy would be expected to be much lower than 0.8 MeV. Another possibility is that the nucleus at the barrier is stable for a certain degree of nonaxial symmetry, in which case extra bands for rotation about the major deformation axis occur. These possibilities have to be considered especially for the inner barrier.

Odd-parity octupole vibrations have a special role among the transition states; they provide the principal means for odd-parity states of the compound nucleus to decay through fission. Nuclear spectroscopy provides evidence for two of these. At the lower energy, generally about 0.5–0.8 MeV in the actinides, is the $K^{\pi}=0^{-}$ vibration, the ''mass asymmetry'' vibration with rotational band members $I^{\pi}=1^{-},3^{-},5^{-}$, etc. It is assumed to lie at about the same energy at the inner barrier, but probably much lower at the outer barrier where the saddle point has a mass-asymmetric shape [11], and the vibration is a low frequency reflection of the nuclear shape through the potential hill at zero octupole deformation. The higher energy vibration, often known as the ''bending'' vibration, has $K^{\pi}=1^{-}$ with rotational band members $I^{\pi}=1^{-},2^{-},3^{-}$, etc., and is usually found above 0.9 MeV at normal deformation. In most of our detailed modeling, the mass asymmetry vibration is assumed to be at 0.7 MeV at the inner barrier and 0.1 MeV at the outer barrier, while the bending vibration is taken to be 0.8 MeV and 0.6 MeV, respectively.

3. Statistical representations of barrier transition states

The collective states described above are those expected in the energy gap before the appearance of the quasiparticle excitations that result from breaking the pairing energy. The energy gap is well known in the spectra at normal deformation and is a little greater than 1 MeV in the actinides. Above this energy the levels are normally described by a statistical level density function. The simplest form that is used is an exponential with constant temperature:

$$\rho_{A,B}(E,J)=C_{A,B}(2J+1)\exp[-J(J+1)/2\sigma^2]\times\exp[(E-V_{A,B})/\theta], \tag{8}$$

where $\rho(E,J)$ is the density of transition states with zero angular momentum and single parity at excitation energy E and total angular momentum J, σ is a spin dispersion constant and θ is the temperature parameter. The subscripts A,B label the inner and outer barriers, respectively. At excitation energies of several MeV a Fermi-gas (independent particle) form is more appropriate. For such a composite model Gilbert and Cameron [12] have given tables of parameters that fit the level density data. For the actinides these parameters have been readjusted in Ref. [10]. It is found that the temperature parameter is about 0.5 MeV. Spin dispersion constants are in the range of approximately 5–6.

For a statistical representation of the transition states above the energy gap at the barrier deformations, theory suggests that the energy gaps are somewhat higher and the temperatures somewhat lower than those at stable deformation [10]. The level density constants $C_{A,B}$ are also expected to be greater than at stable deformation by a factor dependent on the symmetry of the barrier shape [13]. The fission cross section at excitation energies considerably below the barrier energy gaps is affected by these ''continuum'' transition states because of their high density and the Hill-Wheeler tunneling effect. The level density parameters can be adjusted in calculating the neutron-induced fission cross section up to about 2 MeV neutron energy.

B. Barrier parameters of ^{236}U and calculated fission cross sections

1. Axially symmetric inner barrier model

The ^{235}U(d,p) and ^{234}U(t,p) reactions can reach excitation energies in the compound nucleus well below the neutron separation energy ($S_n=6.53$ MeV for ^{236}U) and thus can explore the fission probability well below the fission barrier. These reactions give the most direct information on barrier heights. Measurements of the (d,pf) and (t,pf) reactions have been made by Back *et al.* [14,15]. Both these reactions excite compound-nucleus states with a wide range of total angular momenta. The results of the calculations of the relative cross sections for spin and parity are given in these references, and these have been used in our fits to the data. The error of measurement assessed in the above references includes a 20% systematic error in magnitude. Therefore only the shape of the fission probability curve in the region of the barrier gives useful information. In this energy region only competition between fission and radiation has to be considered.

The assumption of stiff axial symmetry at the inner barrier implies a high energy for the gamma phonon band transition states. We have assumed its value to be 0.8 MeV, similar to the observed value at normal deformation. With this transition state model, it is found that the fission probability data on the (d,pf) reaction are quite well reproduced with inner and outer barrier parameters:

$$V_A=5.2\ \text{MeV},\quad \hbar\omega_A=1.05\ \text{MeV},$$

$$V_B=5.7\ \text{MeV},\quad \hbar\omega_B=0.6\ \text{MeV}.$$

The energy variation of the (t,pf) data is also quite well reproduced by these parameters although the magnitude above the barrier is not in agreement. In this respect the (d,pf) and (t,pf) data seem inconsistent.

Using these barrier parameters and the model of individual transition states described above, the statistical level density parameters can be adjusted to obtain reasonable agreement between calculation and the measured fission cross section of ^{235}U up to about 1.2 MeV. This is about the value of the energy gap in the target nucleus ^{235}U, and the individual levels up to this energy seem to be quite completely known, thus accounting almost fully for the expected inelastic scattering. Adjusted parameters for the barrier ''continuum'' states are

$$C_A=0.20\ \text{MeV}^{-1},\quad C_B=0.05\ \text{MeV}^{-1},$$

$$\theta_A = \theta_B = 0.42 \text{ MeV},$$

with an energy gap of 1.65 MeV above the inner barrier and 1.03 MeV above the outer barrier.

For neutron energies above 1.2 MeV we also need to describe the states of the residual nucleus for inelastic scattering by means of a level density formula. If we retain the above parameters for the barrier state density, we obtain, by least squares fitting,

$$C_R = 0.194 \pm 0.045 \text{ MeV}^{-1}, \quad \theta_R = 0.54 \pm 0.05 \text{ MeV}.$$

This is a considerably lower density than recommended in Ref. [12]—namely, $C_R = 0.9 \text{ MeV}^{-1}$, $\theta_R = 0.5$ MeV. If we are to retain the parameters of Ref. [10], we must assume that the barrier densities change at 2.5 MeV and 2.0 MeV above the inner and outer barriers, respectively. Then the new barrier density parameters for this higher energy region are

$$C_A = 0.76 \text{ MeV}^{-1}, \quad C_B = 0.19 \text{ MeV}^{-1},$$

$$\theta_A = \theta_B = 0.40 \text{ MeV}.$$

The ground state spin and parity of ^{235}U are $I^\pi = 7/2^-$. At low energies (up to a few tens of keV) *s*-wave neutron absorption is predominant. Compound-nucleus states of spin and parity $J^\pi = 3^-, 4^-$ are formed. Transition states with these quantum numbers are fully open for both barriers at the neutron separation energy. At neutron energies of 50 keV *p*-wave neutron absorption has become comparable with *s*-wave absorption. The resulting compound-nucleus states with J^π ranging from 2^+ to 5^+ access transition states that are fully open over the inner barrier and the even spins are fully open over the outer barrier while the odd spins are about half open there. The *d* waves (exciting J^π ranging from 1^- to 6^-) become significant, but not dominant, at about 0.5 MeV neutron energy. The corresponding transition states are essentially fully open. By contrast the 77 eV isomer has spin $I^\pi = 1/2^+$. The *s*-wave compound-nucleus states have $J^\pi = 0^+, 1^+$, the latter carrying three-quarters of the compound-nucleus formation cross section. The $J^\pi = 0^+$ transition state is open at the neutron separation energy for both barriers, but the more important 1^+ state (a bending plus mass asymmetry combination) is about 0.3 MeV higher at the inner barrier and perhaps about equal to the neutron separation energy at the outer barrier. For this reason the low energy fission cross section of the isomer is calculated to be considerably lower than that of the ground state. The *p*-wave neutron absorption excites compound-nucleus states with $J^\pi = 0^-$, 1^- and 2^-. The lowest 0^- transition is not believed to exist within the energy gap (which is at approximately 7 MeV excitation for both barriers). Fission of the 0^- compound states (1/12th of the compound-nucleus formation cross section) is thus suppressed at low neutron energies.

The fission cross section calculated for the ground state from this barrier model, which we call model 1, is shown in Fig. 1, where the experimental data [16–18] are also plotted. It is in fair agreement with the experimental data up to nearly 2 MeV. The ratio of the fission cross section calculated for the isomer to the calculated cross section for the ground state is shown in Fig. 2.

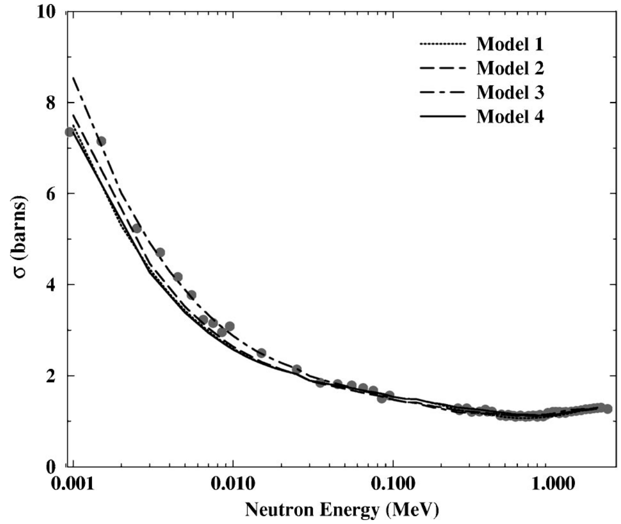

FIG. 1. The fission cross section of the ^{235}U ground state for models 1, 2, and 3.

2. Axially asymmetric inner barrier: Rigid rotator, $\gamma = 11°$

The degree of axial asymmetry of a rigid rotator is expressed by the conventional γ parameter, in which $\gamma = 0$ describes a prolate spheroid and $\gamma = 30°$ describes the maximum axial asymmetry. This model assumes $\gamma = 11°$, a moderate degree of axial asymmetry. With the ground state rotational band inertial constant taken as $\hbar^2/2\mathfrak{I} = 3.33$ keV (giving the first 2^+ rotational state at ≈ 20 keV) the first $2^+, 3^+, 4^+$, etc., band (which can be thought of approximately as a gamma rotational band) occurs at about 250 keV, while a $4^+, 5^+, 6^+$, etc., band ("2 gamma" rotational) starts at about 1 MeV [19]. Apart from higher bands involving combinations with the gamma bands, other transition states are similar to those in model 1.

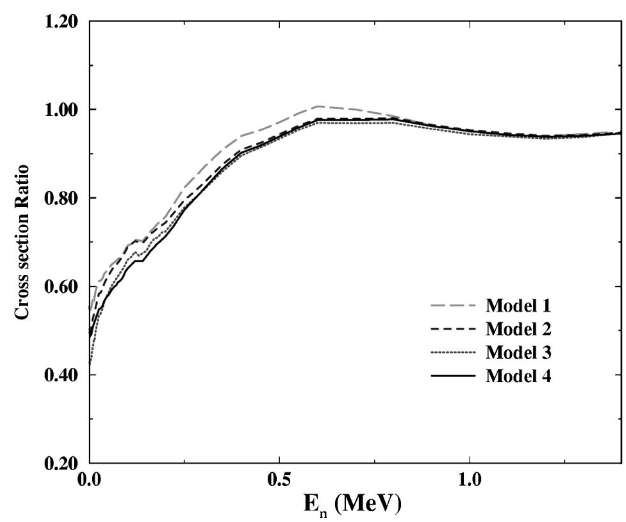

FIG. 2. Ratio of calculated cross section for the 77 eV isomer of ^{235}U to that of the ground state.

The $^{235}U(d,pf)$ fission probability data are quite well reproduced with inner and outer barrier parameters:

$$V_A = 5.53 \text{ MeV}, \quad \hbar\omega_A = 1.05 \text{ MeV},$$

$$V_B = 5.53 \text{ MeV}, \quad \hbar\omega_B = 0.6 \text{ MeV}.$$

Using these barrier parameters and the model of individual transition states described above to calculate the neutron fission cross section up to 1.2 MeV, the statistical level density parameters can be adjusted as in model 1 to obtain

$$C_A = 0.34 \pm 0.07 \text{ MeV}^{-1}, \quad C_B = 0.07 \pm 0.02 \text{ MeV}^{-1},$$

$$\theta_A = \theta_B = 0.475 \pm 0.03 \text{ MeV},$$

with an energy gap of 1.25 MeV above the inner barrier and 1.15 MeV above the outer barrier. Above 1.2 MeV neutron energy, the residual nucleus level density is described with parameters $C_R = 0.212 \pm 0.05$ MeV, $\theta_R = 0.566 \pm 0.04$ MeV relative to the barrier state density.

The effect of model 2 on the 77 eV isomer is to lower the *s*-wave contribution to the cross section even more than in model 1. The fission cross sections calculated with this barrier model are also given in Figs. 1 and 2.

3. Axially asymmetric inner barrier: Rigid rotator, $\gamma = 30°$

The ground state rotational band inertial constant is again taken as $\hbar^2/2\Im = 3.33$ keV. The higher bands, based on 2^+ and 4^+ states, deviate more from the rotational form, but the rotational relations can be used approximately with an inertial constant of 5.9 keV. The lower band starts at 0.06 MeV and the higher band at 0.2 MeV. There is also a "3 gamma" band starting with $J^\pi = 6^+$ at 0.4 MeV.

The $^{235}U(d,pf)$ fission probability data are reproduced with the same inner and outer barrier parameters as in model 2. Using these barrier parameters and the model of individual transition states described above, the statistical level density parameters can be adjusted to obtain agreement between the model 3 calculation and the neutron fission cross section up to 1.2 MeV to obtain

$$C_A = 0.34 \text{ MeV}^{-1}, \quad C_B = 0.07 \text{ MeV}^{-1},$$

$$\theta_A = \theta_B = 0.46 \text{ MeV},$$

with an energy gap of 1.32 MeV above the inner barrier and 1.22 MeV above the outer barrier. Above 1.2 MeV the parameters of the residual nucleus level density are $C_R = 0.22 \pm 0.05 \text{ MeV}^{-1}$, $\theta_R = 0.504 \pm 0.05$ MeV relative to the barrier state density.

The fission cross sections calculated with this barrier model are shown in Figs. 1 and 2. The low energy fission cross section for the ground state is higher than in model 2, but the ratio of the isomer and ground state cross sections is about the same.

4. Axially soft inner barrier

The assumption that the nucleus at its inner barrier prolate deformation is soft to axially asymmetric distortions will

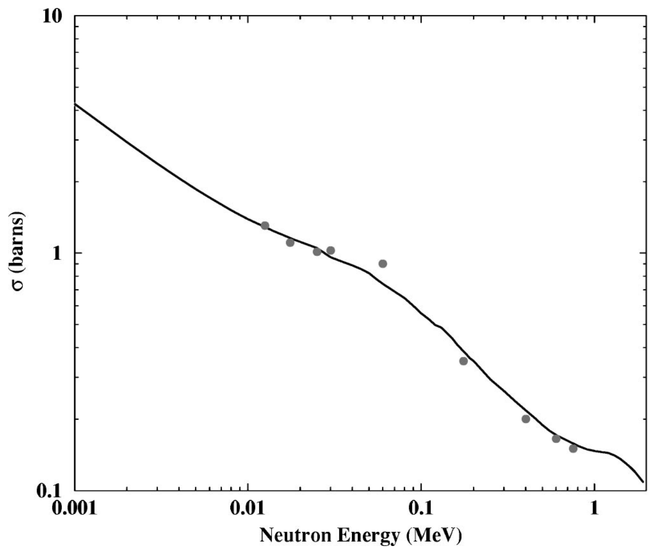

FIG. 3. The capture cross section of the ^{235}U ground state for model 4. As can be seen from Fig. 8, the capture cross section for the isomer is predicted to be significantly larger than that for the ground state below 0.5 MeV.

lower the estimates of the gamma vibrational energy. We assume for our calculations with this model that the gamma phonon energy is 0.25 MeV and that these vibrations are harmonic. The rotational inertial constant is assumed to be 3.33 keV for all bands.

Again, the $^{235}U(d,pf)$ fission probability data can be reproduced with inner and outer barrier parameters as in model 2.

Using these barrier parameters and this model of individual transition states, the statistical level density parameters can be adjusted as in previous models to obtain

$$C_A = 0.34 \text{ MeV}^{-1}, \quad C_B = 0.07 \text{ MeV}^{-1},$$

$$\theta_A = \theta_B = 0.47 \text{ MeV},$$

with an energy gap of 1.1 MeV above the inner barrier and 1.15 MeV above the outer barrier. The residual nucleus level density parameters $C_R = 0.19 \pm 0.05 \text{ MeV}^{-1}$, $\theta_R = 0.546 \pm 0.05$ MeV relative to the barrier state density [20–22].

The fission cross section calculated with this barrier model is shown in Fig. 1, while the ratio of the isomer cross section to the ground state cross section is shown in Fig. 2.

Graphs of the cross sections of the competing reactions are given in Figs. 3 and 4 for models 3 and 4, respectively. All cross sections have been corrected for the $(n,\gamma n')$ and $(n,\gamma f)$ reactions. Some calculations (based on model 3) have also been made of the cross section for populating the isomer from the ground state by the $(n,n'\gamma)$ reaction. Different results are obtained depending on the assumption that *K*-quantum-number selection rules apply to the cascading gamma transitions or not. However, in all cases the isomer is predicted to be strongly populated via the $(n,n'\gamma)$ reaction on the ^{235}U, and the cross section leading to the isomer at neutron energies ~ 1 MeV is of the order of 0.5 b. These results are shown in Tables I and II.

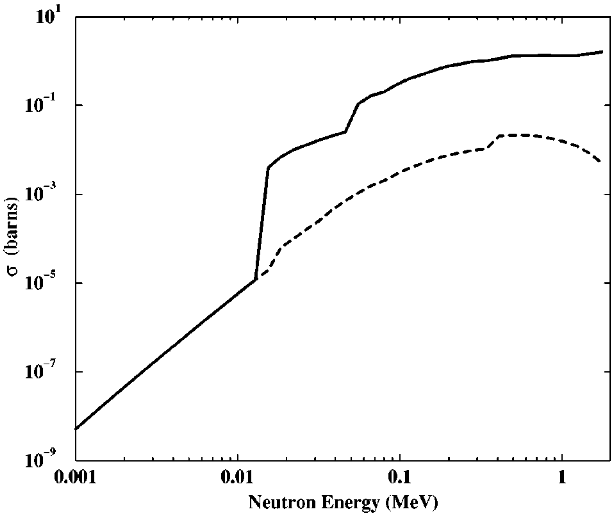

FIG. 4. The total inelastic cross section of ^{235}U. The dotted line shows the predicted cross section for population of the isomer by the (n,n') reaction.

III. CALCULATIONS OF THE CROSS SECTIONS OF RELATED ACTINIDES

A. Neutron-induced fission cross section of ^{233}U

In this calculation we consider only model 4 of Sec. II. The spin and parity of the target nucleus is $5/2^{+}$ and the neutron separation energy of the compound nucleus is 6.84 MeV. Fitting to the ^{233}U(d,pf) fission probability data suggests

$$V_A = 5.83 \text{ MeV}, \quad \hbar\omega_A = 1.05 \text{ MeV},$$

$$V_B = 5.83 \text{ MeV}, \quad \hbar\omega_B = 0.7 \text{ MeV}.$$

Using these barrier parameters and transition state model 4, and barrier state density parameters that are close to those deduced for the ^{235}U(n,f) models ($C_A = 0.34$ MeV^{-1}, $C_B = 0.157$ MeV^{-1}, $\theta_a = \theta_B = 0.45$ MeV, $C_R = 0.23 \pm 0.05$ MeV^{-1}, $\theta_R = 0.465 \pm .05$ MeV relative to the barrier state density), we calculate neutron fission cross sections in good agreement with the data [23,24]. We show the comparison in Fig. 5.

TABLE I. Calculated cross sections for inelastic scattering from the ground state leading to population of the isomer and the ground state (the ground state cross section includes the compound elastic scattering). The assumption is made that all states below 2 MeV in ^{235}U have good *K*-numbers.

Neutron energy (MeV)	$\sigma(n,n'\gamma \rightarrow is)$ (b)	$\sigma(n,n' \rightarrow gd)$ (b)
0.1	0.21	0.14
0.3	0.44	0.54
0.5	0.46	0.71
0.7	0.47	0.90
0.9	0.43	0.94
1.1	0.42	0.95
1.3	0.54	0.83
1.5	0.71	0.66
1.7	0.83	0.55

TABLE II. Calculated cross sections for inelastic scattering from the ground state leading to population of the isomer and the ground state. The assumption is made that there is complete *K* mixing.

Neutron energy (MeV)	$\sigma(n,n'\gamma \rightarrow is)$ (b)	$\sigma(n,n'\gamma \rightarrow is)$ (b)
0.1	0.21	0.14
0.3	0.60	0.38
0.5	0.77	0.41
0.7	0.88	0.49
0.9	0.89	0.48
1.1	0.88	0.50
1.3	0.83	0.53
1.5	0.80	0.57
1.7	0.78	0.60

B. Neutron-induced fission cross section of ^{237}U

Again we consider only model 4 of Sec. II. Like the isomeric state of ^{235}U the spin and parity of the target nucleus are $1/2^{+}$ and the barrier transition states governing the fission cross section should therefore be very similar with the two nuclides differing by only two neutrons. However, the neutron separation energy of the compound nucleus is lower: 6.15 MeV. Fitting to the ^{236}U(t,pf) fission probability data suggests

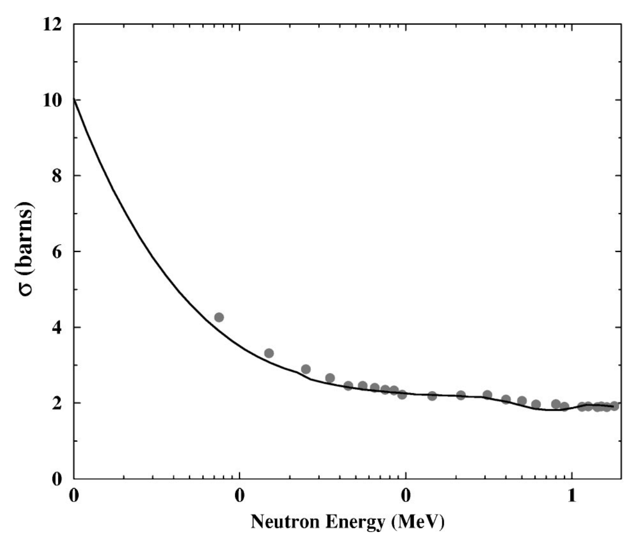

FIG. 5. The fission cross section of ^{233}U using the barrier parameters and transitions states of model 4. The spin and parity of the target nucleus are $5/2^{+}$.

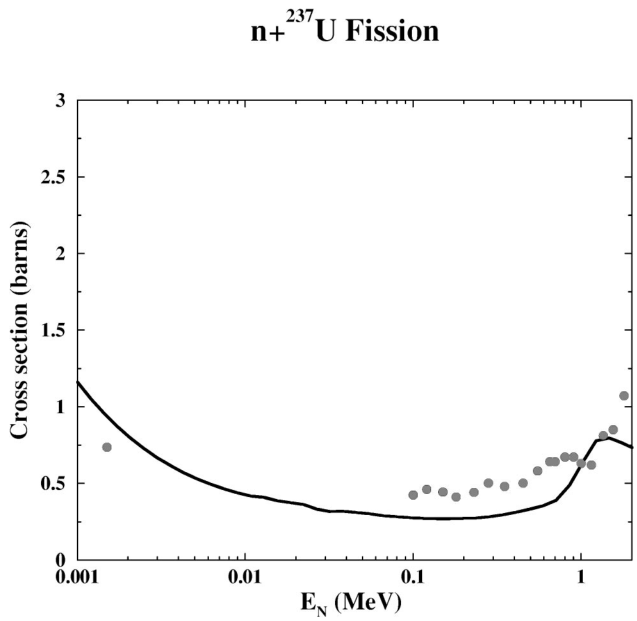

FIG. 6. The fission cross section of ^{237}U. The data of Ref. [25] are reduced by a factor 0.63 to agree with critical assembly measurement of the ratio of ^{237}U and ^{235}U fission cross sections [26].

$$V_A = 5.73 \text{ MeV}, \quad \hbar\omega_A = 1.05 \text{ MeV},$$

$$V_B = 5.83 \text{ MeV}, \quad \hbar\omega_B = 0.7 \text{ MeV}.$$

Using these barrier parameters and transition state model 4, we calculate neutron fission cross sections that are in fair agreement with the rather sparse data; these are limited to a single one-pulse time-of-flight measurement on the Pommard shot [25] and a ratio measurement relative to the ^{235}U fission cross section in a critical assembly [26]. We use this ratio as a normalization factor on the differential data. The comparison between calculation and adjusted data is shown in Fig. 6. The single point at 1.5 keV neutron energy is extrapolated from the resonance information measured in Ref. [25]. The generally lower trend of the calculation compared with the data suggests that the barrier heights are too high. However, to achieve agreement with the data in the range from 100 keV to 1 MeV calculations show that the barriers would have to be reduced by about 200 keV, which is incompatible with the analysis of the (t,pf) and resonance parameter data.

C. Neutron-induced fission cross section of ^{239}Pu

Again we consider only model 4 of Sec. II. Like the isomer of ^{235}U, the spin and parity of ^{239}Pu are $1/2^+$ and the relevant barrier transition states should be similar, as is the neutron separation energy of the compound nucleus: 6.53 MeV. However, the known general systematic trends of the double-humped fission barrier heights suggest that while the inner barrier height will be similar to that of the uranium nuclides we have studied above, the outer barrier may be about 0.5 MeV lower. This is confirmed by fitting to the ^{239}Pu(d,pf) fission probability data, which agrees with

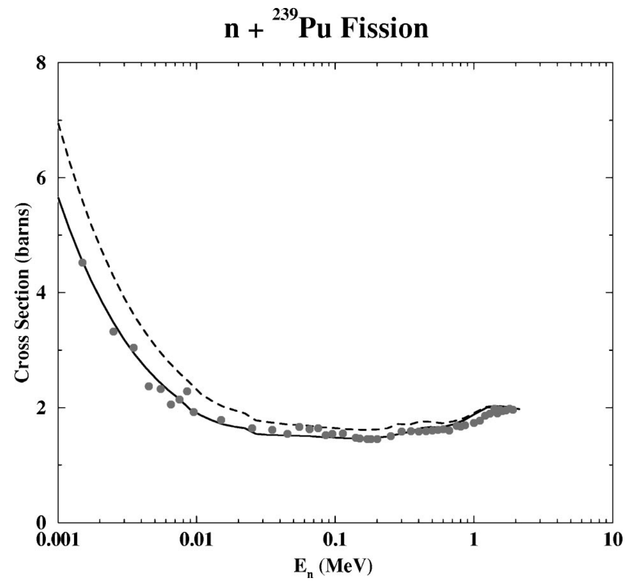

FIG. 7. The calculated fission cross section of ^{239}Pu compared with the experimental data. The solid curve has been calculated from model 4, while the dashed curve has been calculated with the barrier energy gaps increased by 0.1 MeV.

$$V_A = 5.63 \text{ MeV}, \quad \hbar\omega_A = 1.05 \text{ MeV},$$

$$V_B = 5.13 \text{ MeV}, \quad \hbar\omega_B = 0.7 \text{ MeV}.$$

Using these barrier parameters and transition state model 4, we calculate neutron fission cross sections that are in fairly good agreement with the data [24,27]. The comparison is shown in Fig. 7. The dashed curve has been calculated with the same barrier state densities as in model 4 of the ^{235}U(n,f) case. For the bold curve the energy gaps at the barriers have been raised by 0.1 MeV. The low energy fission cross section is considerably lower than that of the ground state of ^{235}U (even though the fast neutron cross section is considerably higher) because of the high energy of the 1^+ transition state at the inner barrier. Above 0.1 MeV where the *p*-wave absorption predominates, the cross section is higher than that of ^{235}U because of the considerably lower outer barrier.

IV. CONCLUSIONS

We have derived double-humped fission barrier parameters and transition state spectra of the compound nucleus ^{236}U that are consistent with the known physics of the fission process and agree with data on the fission probability extending into the sub-barrier energy region and with the neutron fission cross section of the ground state of ^{235}U. With these fission barrier properties we have calculated the fission cross section of the 77 eV isomer of ^{235}U. The key transition state in this calculation is the $J^\pi = 1^+$ state. Both theoretical and experimental evidence suggest that this is at a high energy, especially at the inner barrier. This is the most important transition state for *s*-wave neutron induced fission and, therefore, causes a considerable lowering of the low energy part of the cross section relative to the ground state cross section (∼45%–55% in the models studied). The isomer cross section does not reach near equality with the ground state cross section until the neutron energy is well above 0.5 MeV. The two cross sections then remain nearly equal until at least 2 MeV.

The predicted ratio of the neutron capture cross section to the fission cross section for the isomer is particularly striking. For model 3, for example, this ratio is predicted to be a factor of about 3.4 (2.5) times larger than for the ^{235}U ground state at 1 keV (10 keV). Figure 8 shows these predicted ratios, and the isomer capture to fission ratio remains larger up to a neutron energy of about 0.5 MeV. As discussed earlier and shown in Tables I and II, the isomer is strongly populated by inelastic neutron scattering on the ^{235}U ground state. Thus, neutron reaction network calculations involving the uranium isotopes in a high neutron fluence are likely to be affected by the 77 eV isomer of ^{235}U.

The high value of the thermal neutron fission cross section of the ^{235}U isomer is not necessarily in conflict with the main conclusions of this paper. The thermal neutron fission cross section has a very stochastic nature. It depends on quantities such as the proximity of the nearest resonance level (this has a roughly exponential dependence about a mean value related to the mean level spacing), the neutron width of this level (Porter-Thomas distribution), and its fission width (Porter-Thomas distribution or chi-squared distribution with a low degree of freedom). The resonance level responsible for the neutron fission cross section probably has total angular momentum and parity 0^+, which implies that it could have a large fission width

With one of these models (model 4), which is intermediate in properties among the set studied, the calculations of the fission cross sections are in good agreement with the measured cross sections for ^{233}U, ^{237}U, and ^{239}Pu. Some minor adjustments in barrier heights have to be made for the two uranium isotopes (to agree with the sub-barrier fission probability data). The agreement for ^{237}U is most significant, because this has, like the ^{235}U isomer, spin and parity $I^\pi = 1/2+$. For the ^{239}Pu case the outer barrier has to be reduced by about 0.5 MeV to obtain agreement with the sub-barrier data, but with this change and the same transition state model all the major differences between the fission cross sections of ^{235}U and ^{239}Pu can be explained.

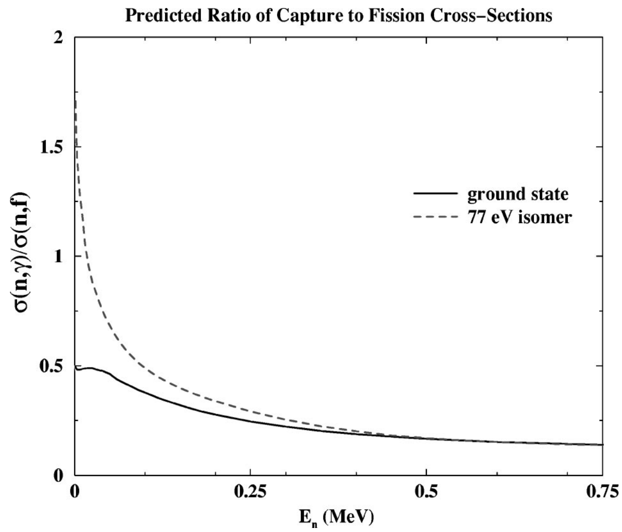

FIG. 8. The calculated ratio of the neutron capture cross section to the fission cross section for the isomer and ground state of ^{235}U. The ratio of these cross sections is 3.4 times larger from the isomer at 1 keV and 2.5 times larger at 10 keV.

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