PHYSICAL REVIEW C VOLUME 10, NUMBER 2 AUGUST 1974

Structure of the fission transition nucleus ^{227}Ra†

H. R. Groening* and W. D. Loveland

Department of Chemistry, Oregon State University, Corvallis, Oregon 97331

(Received 22 April 1974)

The height of the fission barrier and the character of the low lying single particle levels have been determined for the fission transition nucleus ^{227}Ra by fitting the energy variation of the previously reported fission cross section and fragment angular distributions in the neutron energy range $3.6 < E_n < 4.1$ MeV using a Hauser-Feshbach formalism. The best fit to the experimental data gave a fission barrier height $E_f = 8.2 \pm 0.1$ MeV and a single particle state sequence at the barrier of $\frac{3}{2}$, $\frac{1}{2}$, $\frac{5}{2}$, $\frac{1}{2}$ which agrees very well with the theoretical predictions of Nix and Möller. An extension of the calculations to higher neutron energies ($4.7 < E_n < 9.0$) allowed deduction of K_0^2 and the transition nucleus level density as a function of excitation energy. A discontinuity in the K_0^2 values suggests a value of the pairing gap parameter ($2\Delta_f = 2.7$ MeV) that is considerably larger than the corresponding one at the equilibrium deformation ($2\Delta_0 = 1.7$ MeV). The transition nucleus and residual nucleus level densities were compared with theoretical calculations and support the view that a collective rotational and vibrational enhancement of the intrinsic nuclear level density is necessary to account for the experimental data.

[NUCLEAR REACTIONS, FISSION ^{226}Ra(n,f), $E=3.6$–9.0 MeV; ^{227}Ra* deduced levels, K, π, K_0^2, and level density.]

I. INTRODUCTION

Recently, there has been a great deal of interest in and success in calculating various features of the fission barrier structure in heavy nuclei using the Strutinsky shell-correction method.[1] Recent calculations involving the use of P_3 and P_5 nuclear deformations have suggested that the fission mass distributions may be determined by asymmetric distortions at or near the fission saddle point.[2] While these fission barrier calculations appear to describe reasonably well the experimental data on spontaneously fissioning isomeric states and fission barrier heights in the transuranic elements, they have not been rigorously tested as to how well they predict the low energy single particle level spacings at the saddle point, and few tests have been made concerning predictions of fission barrier structure in nuclei with $Z<90$. In particular, the calculated barrier heights agree within ± 1–2 MeV of the experimental values for uranium and heavier nuclei, but the calculated heights of the barriers disagree by several MeV with the experimental values for Th. In view of this situation, we felt it was highly desirable to gather further information about the fission barrier and the low lying single particle levels in a nucleus with $Z<90$. Accordingly, we report in this paper the determination of the height of the fission barrier and the character of the low lying single particle levels for the ^{227}Ra fission transition nucleus by fitting the energy variation of the fission cross section and fragment angular distributions in the ^{226}Ra(n,f) reaction using a Hauser-Feshbach formalism.

Recently, Bjørnholm, Bohr, and Mottelson[3] have revived interest in the question of "collective enhancement effects" in nuclear level densities of deformed nuclei. We have explored the need for this enhancement and its nature by comparing theoretical calculations of the intrinsic level densities and experimental values of the level density derived from evaporation and neutron resonance data and from a fit to the energy variation of the fission cross section and angular distribution for the ^{226}Ra(n,f) reaction for the neutron energy range $4.7 \leq E_n \leq 9.0$ MeV.

Britt and Huizenga[4] have recently reevaluated evidence for an increased pairing gap at the fission saddle point and found no evidence for such an increase in ^{236}U or ^{240}Pu. In view of the increased likelihood of seeing such an increase in the pairing gap (if the pairing gap depends upon the nuclear surface area) in the lighter fissioning nuclei and the controversy surrounding the experimental data analysis in this region, we have carefully reanalyzed the energy dependence of K_0^2 in the neutron energy region $4.7 \leq E_n \leq 9.0$ MeV for the ^{227}Ra fissioning system. From this data, we infer a value of $2\Delta_f$, the pairing gap at the saddle point, which is much larger than the equilibrium deformation pairing gap, $2\Delta_0$.

Why did we pick the ^{227}Ra transition nucleus as a place to study these effects? Firstly, calculations[2,5-10] predict that, for all intents and purposes, the fission barrier is effectively single-humped for this system. (For example, the height of the inner barrier for ^{228}Ra has been estimated to be 2.4 MeV compared to an outer barrier height of 8.2 MeV.[8] See Sec. II for further discussion of this point.) Thus, deduction of the parameters describing the barrier shape is much easier and more meaningful than similar attempts for heavier nuclei with double-humped fission barriers. Secondly, the fission of nuclei in the Ra region has shown many unusual features, particularly with regard to the occurrence of triple-humped mass distributions and sharp changes in the mass distribution with small changes in excitation energy.[11–14] In particular, Konecny, Specht, and Weber[12] have reported seeing separate symmetric and asymmetric fission thresholds for the ^{226}Ra-(d,pf) reaction. It would be interesting to see how this might affect the fragment angular distributions.

Thirdly, Babenko and co-workers[15–17] have reported some very unusual data concerning the ^{227}Ra transition nucleus. Their published data on the energy variation of the fission cross section and angular distributions is shown in Figs. 1, 2, and 3. Note the sharp changes in the fission fragment angular distribution during the first smooth rise in the fission cross section from 3.5 to 4.1 MeV neutron bombarding energy. Also of interest is the plateau in the fission cross section between 4.0 and 4.7 MeV while there appears to be a violent change in the fragment anisotropy in this region. An additional interesting feature is the further rise in the fission cross section beyond 4.7 MeV. Since the neutron binding energy in ^{227}Ra is ~4.5 MeV and the fission barrier is ~8–9 MeV high, we can safely deduce that none of the low energy structure is due to second chance fission effects. For reference, we should also note that the asymmetric and symmetric fission thresholds observed by Konecny *et al.*[12] differ by about 1 MeV.

In Sec. II of this paper, we discuss the theoretical framework of the calculations while Secs. III and IV are devoted to the presentation and discussion of the calculational results for the neutron energy $3.6 \leq E_n \leq 4.1$ MeV and $4.1 \leq E_n \leq 9.0$ MeV, respectively. In Sec. V, we state the main conclusions of the paper.

II. THEORETICAL FRAMEWORK OF CALCULATIONS

The general theoretical formalism used in the calculations was the same as that outlined by Huizenga, Behkami, and Roberts.[18] The cross section for neutron-induced fission of an even-even nucleus through a specific state of the

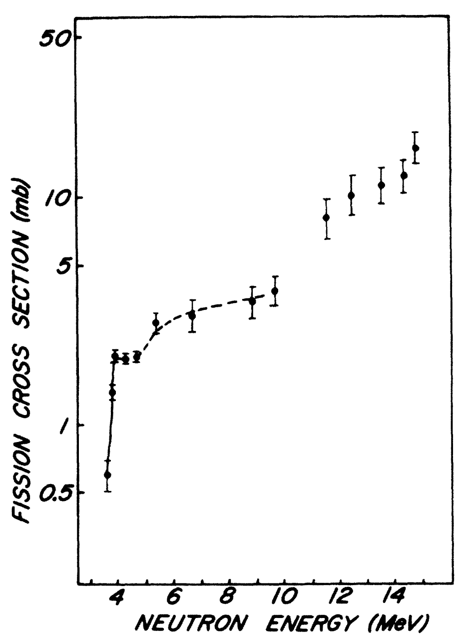

FIG. 1. The fission cross section vs neutron bombarding energy for the ^{226}Ra(n,f) reaction. Data taken from Refs. 15–17. The solid line shows are "best fit" calculation using discrete single particle levels in the transition nucleus while the dashed line represents a statistical description of the transition nucleus.

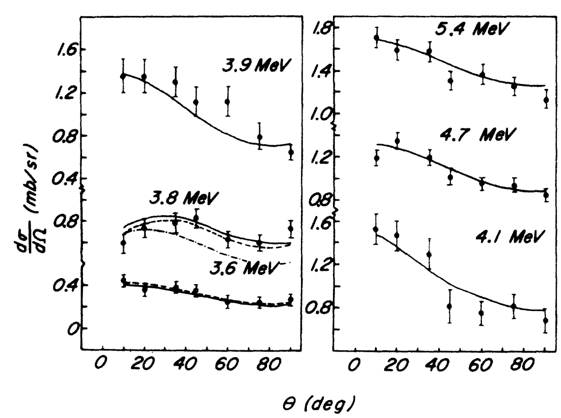

FIG. 2. The fission fragment angular distributions for the ^{226}Ra(n,f) reactions for incident neutron energies of 3.6, 3.8, 3.9, 4.1, 4.7, and 5.4 MeV. Data from Refs. 15–17. For neutron energies of 3.6, 3.8, and 3.9 MeV, the solid curves represent our "best fit" to all of the angular distributions using a symmetric saddle point shape and the dashed curves show the best fit using an asymmetric saddle point shape. The dot-dash curve for the 3.8 MeV data represents the best fit omitting $\frac{5}{2}$ states from the single particle spectrum. For the higher neutron energies, the solid curves represent our best fit using a statistical description of the transition nucleus.

transition nucleus of given (K, J, π) is

$$\sigma_f(K,J,\pi)=\pi\lambda\!\!\!^{-2}\,\frac{(2J+1)}{2}\,T_{lJ}(E_n)\,\frac{2T_f(K,J,\pi)}{\sum_K 2T_f(K,J,\pi)+T_\gamma(E,J,\pi)+\sum_{E'}\sum_{l',J'}T_{l',J'}(E')}\,,$$

where E_n is the incident neutron energy, $\lambda\!\!\!^{-}$ is the reduced wave length of the neutron, T_f, T_γ, and T_{lJ} are the transmission coefficients for fission, γ-ray emission, and neutrons of orbital angular momentum l populating a state of total angular momentum J, respectively. The factor of 2 multiplies T_f because of the double degeneracy of all $K \neq 0$ states. The total fission cross section, σ_f^{total}, is then given as

$$\sigma_f^{\text{total}}=\sum_{K,J,\pi}\sigma_f(K,J,\pi)$$

The fragment angular distributions were given then by the expression

$$\frac{d\sigma_f}{d\Omega}(\theta)=\sum_{J,K,\pi}\frac{\sigma_f(J,K,\pi)W^J_{KM}(\theta)}{2\pi}\,,$$

where the fragment angular distribution associated with fission through a given transition state $W^J_{KM}(\theta)$ has been described previously.[18]

For $E_n \leqslant 3.9$ MeV, the fission transmission coefficients T_f were calculated from the Hill-Wheeler expression for the penetrability of an inverted parabolic fission barrier as

$$T_f(J,K,\pi,E)=\left(1+\exp\{2\pi[E_f(J,K,\pi)-E_n]/\hbar\omega\}\right)^{-1},$$

where $E_f(J,K,\pi)$ is the fission barrier height (relative to the neutron binding energy) associated with the state (J,K,π) of the transition nucleus, and $\hbar\omega$ is the barrier curvature.

The barrier height $E_f(J,K,\pi)$ was calculated using the expression

$$E_f(J,K,\pi)=E_0+(\hbar^2/2I_\perp)\times[J(J+1)-\alpha(-1)^{J+1/2}(J+\tfrac{1}{2})\delta_{K,1/2}]\,,$$

where E_0 is a constant corresponding to the base of the rotational band, $I_\perp$ is the effective moment of inertia about an axis of rotation perpendicular to the nuclear symmetry axis, α is the familiar decoupling constant for the $K=\frac{1}{2}$ band, and $\delta_{K,1/2}$ is the Kronecker δ. The values of K, π chosen for each state of the transition nucleus govern the allowed values of J in the rotational band and the allowed values of l, the orbital angular momentum, to reach a given J.

The assumption of a single-humped fission barrier for the calculations deserves further discussion. As pointed out in Sec. I, all modern theoretical calculations predict a very small inner barrier for nuclei near ^{227}Ra (also see Table II). The theoretical calculations which have worked so well for U and heavier nuclei would have to be in error by several MeV for the height of the inner and outer barriers to be such that one would have to use a double-humped barrier description. On the other hand, the theoretical calculations do fail in the Th isotopes and Britt and Huizenga[4] have suggested that the structure in the fission cross section from 3.6 to 4.7 MeV may be suggestive of a sub-barrier resonance. We find several difficulties with the hypothesis of a sub-barrier resonance. They are: (a) Attempts to find the spontaneously fissioning isomers which might accompany such a sub-barrier resonance have been notably unsuccessful.[19] (b) The "resonance" would have to be unreasonably broad (~1.1 MeV) and the repeated, very careful, measurements which were closely spaced in energy of the fission cross section do not show the characteristic "rise and fall" behavior of a resonance, only a rise and a plateau. (c) As we shall show in Secs. III and IV, the data from 3.6 to 4.7 MeV can be reasonably explained in terms of a single-humped fission

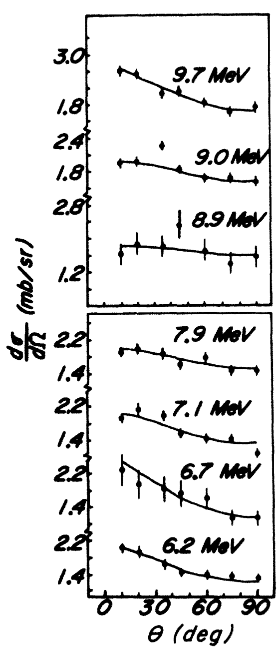

FIG. 3. The fission fragment angular distributions for the ^{226}Ra(n,f) reaction for incident neutron energies of 6.2, 6.7, 7.1, 7.9, 8.9, 9.0, and 9.7 MeV. Data from Refs. 15–17. The solid curves represent our "best fit" calculations.

barrier. We therefore chose to use an effective single-humped fission barrier for our calculations. We wish to emphasize, however, that this choice is a special assumption for the ^{227}Ra nucleus and would not, in general, be expected to be valid for heavier nuclei.

As recently pointed out by Vandenbosch,[20] Ericson,[40] and Bjørnholm, Bohr, and Mottelson,[3] the choice of the values of (J, π) in the rotational band associated with a given K state is somewhat ambiguous. If the transition nucleus has a reflection symmetric shape, then one would expect the usual form of the allowed values of (J, π) for a rotational band, i.e., 0+, 2+, 4+, 6+, ... or $\frac{3}{2}^-$, $\frac{5}{2}^-$, $\frac{7}{2}^-$, etc. However, if, as predicted by calculations,[2,8-10] the transition nucleus ^{227}Ra has an asymmetric shape, then the number of levels in the rotational bands are doubled. For example, asymmetric *e-e* nucleus rotational bands have form 0+, 1−, 2+, 3−, 4+, 5−, etc., while odd A-nucleus rotational bands have the form $\frac{3}{2}\pm$, $\frac{5}{2}\pm$, $\frac{7}{2}\pm$, etc. In the odd A case, levels of both parity are degenerate due to the lack of reflection symmetry in the nucleus. Thus, each (K, J) level is fourfold degenerate, i.e., ±parity and $\pm K$ value. Although some theoretical predictions favor an asymmetric shape for the ^{227}Ra transition nucleus, we do not feel that we can *a priori* rule out the symmetric transition nucleus. We have, therefore, carried out calculations using both assumptions for $E_n \leqslant 3.9$ MeV.

For $E_n \geqslant 4.1$ MeV, insufficient experimental data is available to allow a statistically significant specification of the single particle states of the transition nucleus. For these energies, a statistical description of the level density of the transition nucleus was used. Specifically, one calculated T_f as

$$T_f(K, J, \pi, E) = \frac{\int_0^{U-B_f} \rho_f(E, K, J, \pi) T_f'(E) dE}{\sum_{K=-I_{\max}}^{I_{\max}} \exp[-K^2/2K_0^2]},$$

where $\rho_f(E, K, J)$ is the density of levels with quantum numbers K, J, and π at energy E. The compound nucleus excitation energy is U and the fission barrier height is B_f. $T_f'(E)$ represents a Hill-Wheeler penetrability factor. The level density was parametrized in spherical nucleus Fermi gas form as

$$\rho_f(E, K, J) = \frac{1}{24\sqrt{2}} \frac{(2J+1)}{a_f^{1/4} E^{5/4} \sigma^3} \times \exp\left[2\sqrt{aE} - \frac{(J+\frac{1}{2})^2}{2\sigma^2} - \frac{K^2}{2K_0^2}\right],$$

where a_f, the level density parameter, and K_0^2 are varied as free parameters to give the best fit simultaneously to the energy variation of the fission cross section and angular distribution. The total level density $\rho_f(E, K, J)$ was assumed to consist of equal numbers of positive and negative parity levels.

In specifying the parameters to be used in the above equations, $\hbar\omega$ was assumed to be 0.4 MeV, while the spin cutoff parameter σ was calculated from the expression

$$\sigma^2 = I_\perp T/\hbar^2,$$

where $I_\perp$ was taken from the estimates of Brack *et al.*[8] ($\hbar^2/2I \simeq 2$ keV). The nuclear temperature T was calculated using the expression

$$\frac{1}{T} = \frac{\partial \ln[\omega_f(E)]}{\partial E}$$

where $\omega_f(E)$ was taken to be a function of E and $a_f(E)$.

After the conclusion of most of the calculations reported in this work, it was pointed out[3,21,22] that one may question the choice of a "spherical nucleus form" of the nuclear level density for the deformed ^{227}Ra transition nucleus since the level density expressions for a deformed nucleus should differ from those used for spherical nuclei. However, since we shall be primarily concerned with determining the absolute values of the transition nucleus level density and its energy dependence by fitting the data, and since the energy dependence of the "deformed" and "spherical" nucleus level density expressions is very similar, the choice of an incorrect functional form for the level density should not affect our conclusions. While the absolute values of a_f deduced in our work will not be correct, the level densities should be, and therefore we did not feel it to be worth the effort and expense of repeating the calculations with a deformed nucleus formalism.

The γ-ray transmission coefficients, $T_\gamma(E, J, \pi)$, for γ-ray decay of the compound nuclear state with total angular momentum J, parity π, and excitation energy U, were calculated using the formalism outlined previously by others (Refs. 18, 23).

The procedure which we decided to follow in order to account for the residual nucleus is to treat these residual levels in a nondiscrete, statistical manner, accounting for every $J\pi$ value individually. The over-all effect of this assumption is that outgoing neutron transmission coefficients are now replaced by "compound transmission coefficients." The neutron channel summation in the denominator of the Hauser-Feshbach expression would then be replaced in the following way:

$$\sum_{E'} \sum_{l'j'} T_{l'j'}(E') \to \sum_{J\pi} \sum_{l'j'} \int_0^{E_n} T_{l'j'}(E_k)\rho_{I\pi}(E_n - E_k)dE_k ,$$

where $\rho(E_n - E_k)$ is the level density at an excitation energy equal to the incident neutron kinetic energy minus that of the outgoing neutron.

No attempt was made in the calculations to account for level width fluctuation effects[23] because to do so would be inconsistent with the use of neutron transmission coefficients based on optical model search codes not incorporating the Moldauer theory. Individual neutron transmission coefficients were taken from the compilation by Meldner and Lindner[24] corresponding to $A = 232$, the only tabulated value in this region. A check upon the appropriateness of these transmission coefficients was made by using them to calculate the energy variation of the total reaction cross section for ^{232}Th $+n$. Quite good agreement was obtained between the calculations and the experimental data of Batchelor, Gilboy, and Townle.[25] Level densities for the residual nucleus were calculated using the

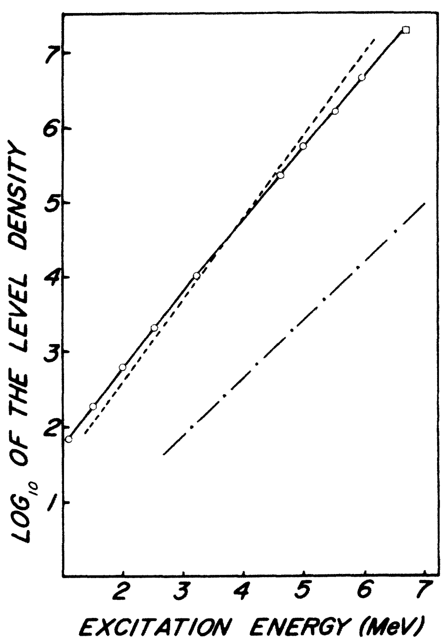

FIG. 4. The total level density of the residual nucleus ^{226}Ra as a function of excitation energy. The solid line represents our best fit to the experimental data (circles) for ^{230}Th, the dashed line represents the Gilbert and Cameron (Ref. 26) prediction. The dot-dash line represents our calculation of the intrinsic single-particle level density.

Gilbert and Cameron constants E_0 and T, in the constant temperature portion of the level density expression, and the level density parameter a in the Fermi gas portion of the level density expression was determined in a fit to the experimental data[25,27] on level densities for ^{232}Th. Furthermore, the transition between the constant temperature and Fermi gas forms of the level density was assumed to take place at 3.0 MeV excitation energy. A spin dependence was added to the level density expression below 3.0 MeV. The fit of the modified Gilbert and Cameron formula to the experimental level density data is shown in Fig. 4 and was found to be quite good. The best values for the other constants were determined to be $T = 0.422$ MeV and $E_0 = -0.397$ MeV. The calculated values of the (n, γ) cross section agreed well with experimental values of this cross section[28] in the energy range tested, $3.6 \leq E_n \leq 6.4$ MeV.

III. RESULTS AND DISCUSSION–$3.6 \leq E_n \leq 4.1$ MeV

We have attempted to fit the energy variation of the total fission cross section and fragment angular distributions in the energy region from $E_n = 3.6$ MeV to $E_n = 4.1$ MeV. Using the theory described above and after an extensive search of the possible number of accessible states of the transition nucleus and the possible values of the free parameters, K, π, E_0, and $\hbar\omega$ for each state, we have concluded that the experimental data in the energy region from $E_n = 3.6$ MeV to $E_n = 3.9$ MeV can be fitted by assuming the ^{227}Ra transition nucleus single particle spectrum shown in Fig. 5 and Table I. The best fits to the data in this region are shown in Figs. 1 and 2.

To guide us in a quantitative evaluation of the agreement between theory and experiment, we used the χ^2 criterion to reject unsatisfactory hypotheses. Each hypothesis tested consisted of two parts, the calculational framework described above, and a particular choice of the free parameters K, E_0, $\hbar\omega$, and π. Unsatisfactory hypotheses were rejected at the 0.05 level of significance. Although we reached reasonable choices of K, E_0, $\hbar\omega$, and π, we made only a limited search of different forms of the calculational framework. In particular, we found that a $\pm 10\%$ variation in ρ_n, a factor of 2 change in the decoupling constant α, and the rotational constant $\hbar^2/2I_\perp$ had a negligible effect upon the calculated transition state spectrum. Therefore, we are saying that *using the theoretical approximations described above as a basis for calculation*, we can reject all unsatisfactory values of the free parameters with only 1 chance in 20 of being in error.

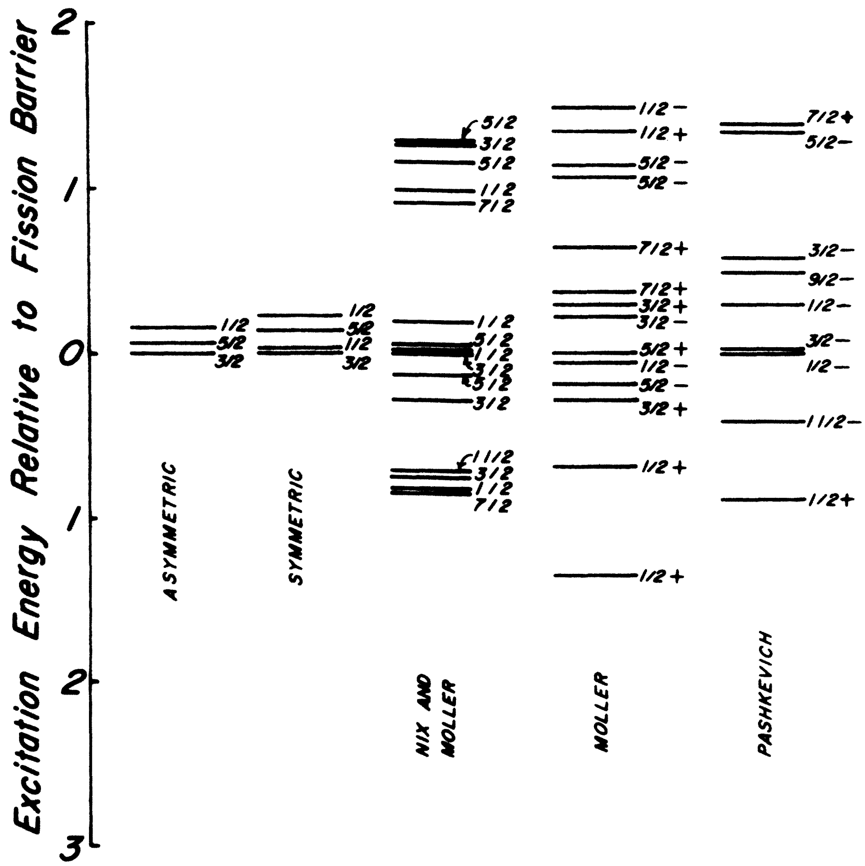

FIG. 5. A comparison of the ^{227}Ra transition nucleus single particle level spectrum for asymmetric and symmetric saddle point shapes deduced in this work with the calculations of Nix and Möller (Ref. 10), Möller (Ref. 2), and Pashkevich (Ref. 7).

In making our search for acceptable hypotheses to describe the data, we have assumed that we should use the minimum number of accessible states of the transition nucleus at any given energy. This assumption, made for simplicity and precision in the determination of the free parameters, means that there may be many hypotheses involving weakly excited states (i.e., high spin states) which will fit the data. We simply cannot say anything about them.

Examining the data in Fig. 1 and 2, we can see qualitatively the 3.6 MeV angular distribution shows the characteristic pattern of fission through a $K=\frac{1}{2}$ band. The parity of the $K=\frac{1}{2}$ state was found to be + rather than − because the required $K=\frac{3}{2}$ and $K=\frac{5}{2}$ strength needed to fit the 3.8 MeV data would not be achieved if a very strongly excited $K=\frac{1}{2}^-$ state were present. The 3.8 MeV angular distribution shows intermediate angle peaking characteristic of $K>\frac{1}{2}$ states. The peak at ~45° in the angular distribution requires a K values higher than $\frac{3}{2}$ to reproduce it. Significant $K=\frac{5}{2}$ strength must be present to cause peaking at such angles as shown in Fig. 2. The parity of the $K=\frac{3}{2}$ state was chosen to be negative to allow the necessary strength for this fission channel to fit the angular distribution and cross section data. It was found that both $K=\frac{5}{2}^-$ and $K=\frac{5}{2}^+$ states would allow statistically significant fits to the data so the parity of the $K=\frac{5}{2}$ state

TABLE I. Parameters describing the low lying single particle states in the ^{227}Ra transition nucleus.

State no.	(K,π)	E_0 (MeV)	$\hbar\omega$
Symmetric saddle point deformation			
1	$\frac{3}{2}^-$	3.65	0.4
2	$\frac{1}{2}^+$	3.67	0.75
3	$\frac{5}{2}^{\pm}$	3.7	0.4
4	$\frac{1}{2}^{\pm}$	3.88	~0.15
Asymmetric saddle point deformation			
State no.	K	E_0 (MeV)	$\hbar\omega$
1	$\frac{3}{2}$	3.76	0.6
2	$\frac{5}{2}$	3.83	0.4
3	$\frac{1}{2}$	3.92	1.0

has not been determined. The 3.9 MeV angular distribution data show the signature of a $K=\frac{1}{2}$ state whose parity could not be determined in a statistically significant manner. For $E_n \geq 4.1$ MeV, the quantity and quality of the data is not sufficient to sustain statistically significant further analysis in terms of the single particle levels of the ^{227}Ra nucleus.

Many detailed searches for best fits to the data have indicated that the positions of the single particle states given in Table I should be regarded as uncertain to $< \pm 0.1$ MeV for the symmetric case and $< \pm 0.05$ MeV for the asymmetrically deformed case. The values of the barrier curvature $\hbar\omega$ should be regarded as uncertain to ± 0.2 MeV. Assuming a neutron binding energy of 4.5 MeV for ^{227}Ra, this calculation places an upper limit on the fission barrier of ~8.2 MeV.

The available data on fission barrier heights for the lighter fissioning elements is summarized in Table II along with many calculations of these quantities. The fission barrier height of 8.2 ± 0.1 MeV deduced for the ^{227}Ra transition nucleus agrees well with the value of 8.5 ± 0.5 MeV found by Zhagrov *et al.*[22] for ^{226}Ra in photofission studies. (Throughout this discussion, we shall assume that odd-even effects, i.e., "specialization energies," are small for these nuclei and will thus compare *e*-*e* and odd *A* nuclei.) The value of 8.2 ± 0.1 MeV for the ^{227}Ra fission barrier height is only ~1–2 MeV lower than most calculations of fission barrier heights in this region (with the exception of the calculations of Nix and Möller, who predict an outer barrier height of 8.1 MeV for ^{226}Ra and Brack *et al.*, who predict a barrier height of 8.2 MeV for ^{228}Ra). Thus, it seems that the theoretical calculations of the outer barrier heights appear to be much better for ^{227}Ra than for ^{230}Th.

Figure 5 shows the spectrum of single particle states found for the ^{227}Ra transition along with various theoretical predictions of the neutron single particle level ordering in ^{227}Ra. No pairing corrections have been made to any of the levels except those of Nix and Möller. The levels shown in Fig. 5 represent our best attempt to interpret the single particle level schemes shown in Refs. 2 and 7. Clearly, the best agreement between the deduced single particle levels and those predicted by various authors occurs with the single particle level scheme of Nix and Möller.[10] In fact, the Nix-Möller calculations show truly remarkable agreement with experimental data in their predictions of the fission barrier height and single particle level schemes.

IV. RESULTS AND DISCUSSION—$4.1 \leq E_n \leq 9.0$ MeV

A. Energy region $3.9 \leq E_n \leq 4.7$ MeV

The energy region of 3.9–4.7 MeV is the region in which there is a plateau in the fission cross section and is a difficult region to analyze because the number of levels in the transition nucleus is (a) too large to allow a statistically significant microscopic calculation and (b) too small to allow

TABLE II. Calculated and experimental values of fission barrier heights for the lighter fissioning elements.

Nucleus	Barrier	Outer barrier (symmetric)	Outer barrier (asymmetric)	Experimental	Reference
^{226}Ra	···	···	···	8.5 ± 0.5	Zhagrov, Nemilov, and Selitskii (29)
	~4.5	~10	~10	···	Adeev, Gamalya, and Cherdantsev (6)
	4.2	10.5	9.0	···	Möller (2)
	3.7	10.7	···	···	Mosel and Schmitt (30)
		10.2	10.2	···	Pauli (9)
	···	···	8.12	···	Nix and Möller (10)
^{227}Ra	···	···	···	8.2 ± 0.1	This work
^{228}Ra	2.4	···	8.2		Brack *et al.* (8)
	4.2	11.3	8.7	···	Möller (2)
^{230}Th	···	···	···	6.0 inner 6.0 outer	Bjørnholm (3)
	4.0	6.9	9.4	···	Möller (2)
^{232}Th	···	···	···	5.9 inner 6.1 outer	Bjørnholm (3)
	3.9	···	6.8	···	Brack *et al.* (8)
	4.6	10.1	6.7	···	Möller (2)
	3.4	···	6.6	···	Pauli (9)

a meaningful statistical model for the transition nucleus level density to be used. To treat this region, we have assumed that the Nix-Möller single particle level scheme is correct, fed these discrete levels into our calculation, and calculated the fission cross section. The results, shown in Fig. 1, demonstrate that the plateau in the fission cross section is due to locally low density of low spin single particle levels in this region as shown in Fig. 5.

One might speculate further that the reason that this plateau in the fission cross section appears to be "washed out" or absent in the ^{226}Ra(d, pf) work of Konecny *et al.*[12] is that the higher angular momentum brought in by the (d, pf) reaction as compared to the (n, f) reaction has allowed population of higher lying members of the rotational bands built upon the single particle states and some higher spin single particle states.

B. Energy region $4.7 \leq E_n \leq 9.0$ MeV

The analysis of the higher energy data was carried out using the statistical description of the transition state nucleus described in Sec. II. The fission level density parameter a_f and the K_0^2 parameter were determined for each energy by determining the best fit to the available cross section and angular distribution data. Once again, a χ^2 criterion was used to judge the statistical significance of the results and to assign uncertainties to the K_0^2 values. The best fits to the cross section and angular distribution data are shown in Figs. 1, 2, and 3. The values of the transition nucleus level density determined in the fitting are shown in Fig. 6, while the values of K_0^2 are shown in Fig. 7. Some checks were made of the sensitivity of the calculated values of a_f and K_0^2 to changes in the barrier curvatures $\hbar\omega$ and the spin cutoff parameter σ. The effect of a 0.2 MeV change in $\hbar\omega$ was to produce a 0.5% change in a_f, while a 50% change in σ^2 produced a ~13% change in a_f and no change in K_0^2.

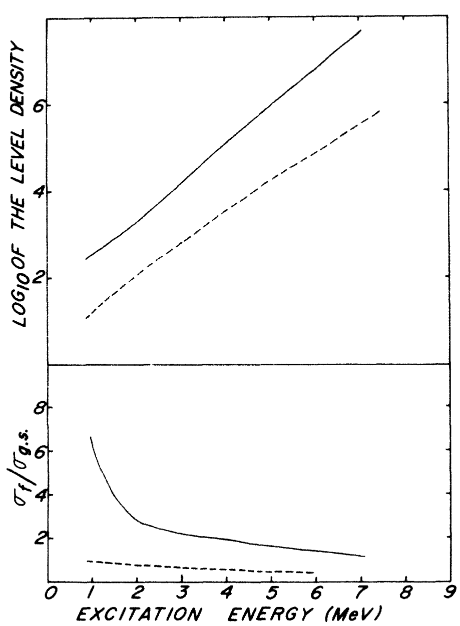

FIG. 6. (a) The total nuclear level density for the transition nucleus ^{227}Ra vs excitation energy. The solid curve represents our best fit to the experimental data from the ^{226}Ra(n,f) reaction while the dashed curve represents our calculation of the intrinsic single particle level density. (b) The ratio of the transition nucleus level density ρ_f to the ground state deformation level density $\rho_{g.s.}$. The solid curve shows $\rho_f/\rho_{g.s.}$ vs the true excitation energy while the dashed curve shows $\rho_f/\rho_{g.s.}$ vs pseudo-excitation energy (see text for details).

K_0^2 and the nuclear pairing gap

Figure 7 shows the values of K_0^2 deduced in this work compared with the values of K_0^2 calculated by Ippolitov *et al.*[31] based upon single particle level schemes. The agreement between theory and experiment seems to be good. The rise in the theoretically calculated value of K_0^2 at an excitation energy relative to the fission barrier of ~3.3 MeV is due to the formation of the three quasi-particle state. (For $U - B_f \simeq 1$–3 MeV, $K_0^2 \simeq 8$, while $K_0^2 \simeq 24$ for $U - B_f = 4$–5 MeV.) From this rise in K_0^2 which appears in both theory and experiment, one can infer a value of the energy gap parameter for the ^{227}Ra transition nucleus, $2\Delta_f = 2.7 \pm 0.4$ MeV. Table III shows the results of calculations using two different models to predict $2\Delta_f$ for various fissioning nuclei. Both models assume pairing is predominantly a surface effect. In the slab model of Kennedy, Wilets, and Henley[32] the gap parameter Δ is proportional to $S^{3/2}$ where S is the nuclear surface area. In the calculations of Stepien and Szymanski, Δ is assumed propor-

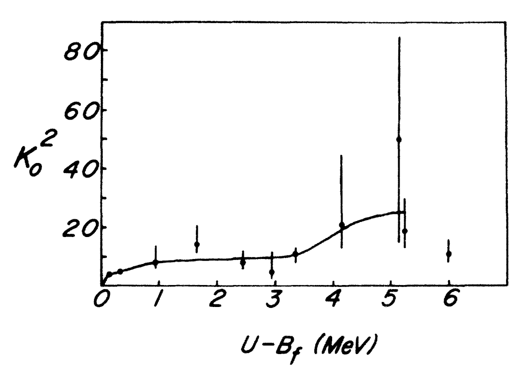

FIG. 7. The deduced values of K_0^2 vs excitation energy above the fission barrier ($B_f = 8.2$ MeV) for ^{227}Ra. Solid curve represents calculation of Ippolitov *et al.* (Ref. 31).

TABLE III. Estimates of the pairing gap at the fission saddle point.

Nuclide	$2\Delta_0$ [a]	(S/S_0) [b]	$2\Delta_f^{\mathrm{exp}}$ [c]	$2\Delta_f$ (Slab model)	$2\Delta_f$ [d] $(\Delta \sim S^3)$
^{227}Ra	1.66	1.243	2.7 ± 0.4	2.3	3.0
^{236}U	1.63	1.185	1.7 ± 0.3	2.1	2.6
^{240}Pu	1.34	1.162	1.6 ± 0.3	1.7	2.0

[a] A. Sobiczewski, S. Bjørnholm, and K. Pomorskii, Nucl. Phys. A202, 274 (1973).
[b] J. R. Nix, Nucl. Phys. A130, 241 (1969).
[c] Values for ^{236}U and ^{240}Pu from Ref. 4.
[d] Estimates of W. Stepien and Z. Szymanski, Phys. Lett. 26B, 181 (1968) as calculated by L. G. Moretto *et al.*, Phys. Rev. 178, 1853 (1969).

tional to S^3. The best fit (and a reasonable one) to the data is obtained with the assumption that $\Delta \sim S^{3/2}$. We conclude that this is firm evidence for the pairing strength being proportional to the surface area, in particular that $\Delta \sim S^{3/2}$.

This point is surrounded by some controversy. Britt and Huizenga[4] in their analysis of $2\Delta_f$ for ^{240}Pu and ^{236}U did not find conclusive evidence that the pairing strength is a strong function of deformation. We merely argue that the changes in nuclear surface area explored in their analysis were small and so it was difficult for them to identify the $\Delta \sim S^{3/2}$ dependence. The same authors allude to a possible sub-barrier fission resonance in ^{227}Ra as an explanation of the increase in $2\Delta_f$ for this system; however, we have shown previously that the ^{226}Ra(n,f) data is well described without such an assumption, and, in fact, there are some arguments against this assumption. Vandenbosch and Huizenga[33] found no evidence for a dependence of the pairing strength upon the nuclear surface area in their analysis of spontaneous fission lifetimes and fission barrier systematics for $Z > 90$. It would appear that because they have dealt with heavier nuclei where the saddle point and equilibrium deformations are not as different as in the lighter nuclei and the apparent weaker dependence of $\Delta \sim S^{3/2}$, the effect might have been missed. In this regard, it would be extremely interesting if one could reduce the uncertainty in estimates of the fission barrier height for ^{210}Po where (S/S_0) is large and $2\Delta_f$ has been reported[34] to be 4 MeV.

An alternative explanation of the rise in K_0^2 at $U\text{-}B_f \sim 3$ MeV would be that this is the consequence of symmetric fission channels becoming available in the transition nucleus. Both Zhagrov *et al.*[14] and Konecny *et al.*[12] have shown that in the energy region 3 MeV $\leq U_f - B_f \leq 4$ MeV, the asymmetric/symmetric fission ratio increases dramatically for the ^{227}Ra transition nucleus. [In particular, for the ^{226}Ra(n,f) reaction $\sigma_{\mathrm{sym}}/\sigma_{\mathrm{asym}}$ goes from 0.09 to 0.24, while $\sigma_{\mathrm{sym}}/\sigma_{\mathrm{asym}}$ goes from 0.19 to 0.54 for the ^{226}Ra(d,pf) reaction.] If, as suggested by the data of Konecny *et al.* and recent calculations, there are separate symmetric and asymmetric fission barriers, and we assume that K_0^2 is larger for the symmetric barrier states as suggested by the greater symmetric fission yields in the (d,pf) as compared to the (n,f) reactions at the same excitation energy, then the rise in K_0^2 near $U\text{-}B_f = 3$ MeV might be due to the onset of symmetric fission in the transition nucleus. It would be interesting to examine the angular distributions of each "component" of the mass yield curve in this region. However, there is a strong argument in favor of interpreting the break in K_0^2 as due to a quasiparticle excitation, i.e., the values of K_0^2 go from the predicted one-quasiparticle values (~ 8) to the three-quasiparticle values (~ 24). Furthermore, there is some theoretical evidence[35] that the mass yield distributions are determined at a later stage in the fission process.

Nuclear level densities

Figure 4 shows the experimentally deduced level density for the ^{230}Th nucleus in its equilibrium deformation (that was used in this work to simulate the ^{226}Ra equilibrium level density). Also shown in Fig. 4 are the results of a calculation of the intrinsic single particle level density using a computer code written by Bolsterli[37] and based upon the formalism of Decowski *et al.*[36] The input parameters for this calculation were the single particle levels and pairing strengths calculated by Nix and Möller.[10] Clearly there is a large discrepancy between the theoretically calculated intrinsic single particle level density and the experimental level density.

Several authors[3,21,22] have pointed out the origin of this discrepancy recently. One must add collective rotational and vibrational levels to the intrinsic single particle level density when the nucleus is deformed as it is in this case. It can be shown[5,21,22] that the "rotational enhancement" of the level density due to collective rotations in the high temperature limit (i.e., the rotational energy is small compared to the nuclear temperature) is of the order of rotational partition function $Z_{\mathrm{rot}} = 2\sigma^2/S$, where σ^2 is the spin cutoff parameter and S is a nuclear symmetry factor. ($S = 1$ for a reflection-asymmetric shape while, $S = 2$ for a nucleus with reflection symmetry.) Similarly the, "vibrational enhancement" of the level density due to collective vibrations is taken in the low temperature limit as the vibrational partition function, i.e.,

$$Z_{\text{vib}}(T)=[1-\exp(-\hbar\omega/T)]^{-(2\lambda+1)} ,$$

where $\hbar\omega$ is the vibrational energy, T is the temperature, and λ is the multipole order of the vibration.

Table IV shows a quantitative comparison of various predictions which include collective enhancement effects of the density of 2+ and 3+ levels at the neutron binding energy. All the predictions of the total level density do agree quite well with experimental data, thus indicating the importance of collective enhancement effects. However, the predictions of Huizenga *et al.*[21] and Dössing and Jensen[22] do not include a collective vibrational enhancement factor corresponding to the well-known softness of nuclei in this region to octupole vibrations ($\hbar\omega/T \sim 0.93$).

To shed further light upon the relative importance of collective vibrational and rotational effects upon the nuclear level density, let us examine the energy dependence of the collective enhancement of the level density. In going from an excitation energy E^* of 1.65 to $E^* = 5.60$ MeV, Fig. 4 shows that the logarithm of the ratio of the experimental level density, ρ_{exp}, to the intrinsic single particle level density ρ_{intr} goes from 1.86 to 2.35, i.e., a factor of 3.1 increase in the collective enhancement factor. We can estimate how our collective enhancement factor $Z_{\text{vib}} Z_{\text{rot}}$ changes in this energy region and get the result that Z_{vib} changes a factor 2.5 while Z_{rot} only changes a factor of 1.37, giving a total change in $Z_{\text{vib}} Z_{\text{rot}}$ of 3.42. Given the crudeness of the approximations in these estimates, we find this agreement between $Z_{\text{vib}} Z_{\text{rot}}$ and $\rho_{\text{exp}}\rho_{\text{int}}$ to be quite satisfactory. We feel that this little calculation demonstrates that the collective rotational enhancement effect does not increase fast enough with energy to account for the experimental data because one is at the high temperature limit of the rotational effects. However, since one is still in the low temperature region for vibrational effects, these effects are important for the energy dependence of ρ for this nucleus.

TABLE IV. Comparison of predictions of the level density of ^{230}Th.

Reference	Intrinsic level density (MeV^{-1}) $\rho_{\text{int}}(2+,3+)$	Collective enhancement factor	Total nuclear level density (MeV^{-1}) $\rho_{\text{tot}}(2+,3+)$
Huizenga *et al.* (21)	...	...	1.79×10^6
Dössing and Jensen (22)	2.56×10^4	26	0.67×10^6
This work	4.07×10^3	660	2.69×10^6
Experimental			2.44×10^6 [a]
			1.72×10^6 [b]
			1.67×10^6 [c]
			1.37×10^6 [d]

[a] J. E. Lynn, *The Theory of Neutron Resonance Reactions* (Clarendon, Oxford, 1968); S. Björnholm and J. E. Lynn, Rev. Mod. Phys. (to be published).

[b] H. Baba, Nucl. Phys. A159, 625 (1970).

[c] W. Dily, W. Schantl, H. Vonach, and M. Uhl, private communication.

[d] P. E. Vorotnikov, Yad. Fiz. 9, 303 (1969) [transl: Sov. J. Nucl. Phys. 9, 179 (1969)].

Figure 6 depicts the situation for the transition nucleus ^{227}Ra. Once again, there is a significant discrepancy between the transition nucleus level density deduced in this work and that calculated from the single particle levels of Nix and Möller.[10] Interestingly enough, the collective enhancement factors appear to be less for the transition nucleus ^{227}Ra than for the equilibrium deformation. The collective rotational enhancement should be larger ($\sim 4\times$) at a given excitation energy for the transition nucleus with its larger moment of inertia and its theoretically predicted lack of reflection symmetry compared to the nucleus at its equilibrium deformation. But the collective vibrational enhancement should be considerably less for the deformed transition nucleus due to the increased vibrational quantum energy $\hbar\omega$. [If $(\hbar\omega/T) \sim 2$ at the saddle point,[38] $(Z_{\text{vib}}^{\text{saddle}}/Z_{\text{vib}}^{\text{g.s.}}) \sim 0.11$].

Figure 6 also shows $(\rho_{\text{saddle}}^{\text{expt'l}}/\rho_{\text{g.s.}}^{\text{expt}})$ as a function of excitation energy relative to the true ground state or saddle point energy surface. The ratio $(\rho_{\text{saddle}}^{\text{expt'l}}/\rho_{\text{g.s.}}^{\text{expt'l}})$ is high at low excitation energies and decreases to near unity as the excitation energy increases. Ordinarily this might be interpreted[39] as being due to a lower than average single particle density for the ground state deformation (associated with a negative shell correction) and a higher than average single particle level density for the saddle point deformation (associated with a positive shell correction). However, Nix and Möller's calculations[10] show shell corrections for the ground state and saddle point deformations for ^{226}Ra of -2.4 and -3.5 MeV, respectively. It would appear, therefore, that in this case several factors are acting to influence the behavior of $\rho_f/\rho_{\text{g.s.}}$. Firstly, the level densities $\rho_f(E^*)$ and $\rho_{\text{g.s.}}(E^*)$ are taken relative to the true ground state and saddle point energy surfaces. The transition nucleus is a even-odd nucleus while the ground state nucleus in an even-even nucleus. At the same value of the excitation energy, one would expect a larger level density for the even-odd nucleus compared to the even-even nucleus. To remove these pairing effects, we shift the saddle point excitation energies down by an amount corresponding to Δ_f and shift the ground state excitation energies down by the amount $2\Delta_0$, thus arriving at the dashed curve shown in Fig. 6. Here we see the greater ground state deformation level densities at a given pseudo-excitation energy primarily due to the greater collective enhancement effects for the ground state nucleus. The decrease in $\rho_f/\rho_{g.s.}$ as E^* increases can be attributed to a lessening of the collective enhancement effects with energy.

V. CONCLUSIONS

What have we learned from this study? We have found:

(a) The fission barrier height and low energy single particle level spacings for ^{227}Ra are in good agreement with the theoretical calculations of Nix and Möller. There appears to be no systematic trend of these calculations to fail to reproduce the fission barrier structure as the (Z,A) of the fissioning system decrease.

(b) There is a break in the variation of K_0^2 vs E^* at ~ 3 MeV above the fission barrier which can be taken as evidence that the nuclear pairing gap at the saddle point, $2\Delta_f$, is larger ($2\Delta_f \sim 2.7 \pm 0.4$ MeV) than the corresponding quantity for the equilibrium deformation ($2\Delta_0 \simeq 1.7$ MeV). A further examination of the data seems to support the idea that $\Delta \sim S^{3/2}$, where S is the nuclear surface area.

(c) The level densities for the transition nucleus and the nucleus at its equilibrium deformation can only be understood in terms of a collective enhancement of the intrinsic single particle level densities. Furthermore, these level densities can only be explained in terms of both a collective rotational and a collective rotational and a collective vibrational enhancement playing important roles.

ACKNOWLEDGMENTS

We wish to thank Dr. J. R. Nix and Dr. P. Möller for allowing us to use their ^{226}Ra calculations before publication and Dr. M. Bolsterli for allowing us to use his level density computer code. Helpful discussions with R. Vandenbosch, H. C. Britt, J. R. Huizenga, and T. D. Thomas are gratefully acknowledged. We wish to express our gratitude to the National Environmental Research Laboratory and the Bonneville Power Administration for allowing us to use their computer facilities in this study. One of us (HRG) wishes to thank the International Atomic Energy Agency and the National Research Council for financial support during the course of this work.

---

†Work supported in part by the U.S. Atomic Energy Commission.

*Present address: Departamento de Fisica, Universidad Simon Bolivar, Caracas, Venezuela.

[1]P. Möller and J. R. Nix, in Proceedings of the Third International Atomic Energy Symposium on the Physics and Chemistry of Fission, August, 1973 (to be published), Paper IAEA-SM-174/202.

[2]P. Möller, Nucl. Phys. A192, 529 (1972).

[3]S. Bjørnholm, A. Bohr, and B. R. Mottelson, in Proceedings of the Third International Atomic Energy Symposium on the Physics and Chemistry of Fission, August, 1973 (to be published), Paper IAEA-SM-174/205.

[4]H. C. Britt and J. R. Huizenga, Phys. Rev. C 9, 435 (1974).

[5]P. Möller and S. G. Nilsson, Phys. Lett. 31B, 283 (1970).

[6]G. D. Adeev, I. A. Gamalya, and P. A. Cherdantsev, Sov. J. Nucl. Phys. 14, 637 (1972).

[7]V. V. Pashkevich, in Proceedings of the Twentieth Annual Conference on Nuclear Spectroscopy, Leningrad, 1970 (unpublished).

[8]M. Brack, J. Damgaard, A. Jensen, H. Pauli, V. Strutinsky, and C. Wong, Rev. Mod. Phys. 44, 320 (1972).

[9]H. Pauli, Phys. Rev. 7, 35 (1973).

[10]J. R. Nix and P. Möller, private communication.

[11]D. G. Perry and A. W. Fairhall, Phys. Rev. C 4, 977 (1971).

[12]E. Konecny, J. G. Specht, and J. Weber, Phys. Lett. 45B, 329 (1973); in Proceedings of the Third International Atomic Energy Symposium on the Physics and Chemistry of Fission, August 1973 (to be published), Paper IAEA-SM-174/20.

[13]R. A. Nobles and R. B. Leachman, Nucl. Phys. 5, 211 (1958).

[14]E. A. Zhagrov, I. M. Kuks, Y. A. Nemilov, Y. A. Selitskii, and V. B. Funshtein, Nucl. Phys. A213, 436 (1973).

[15]Y. A. Babenko, Y A. Nemilov, Y. A. Selitskii, and V. B. Funshtein, Yad. Fiz. 7, 269 (1968) [transl.: Sov. J. Nucl. Phys. 7, 186 (1968)].

[16]Y. A. Babenko, V. T. Ippolitov, Y. A. Nemilov, Y. A. Selitskii, and V. B. Funshtein, Yad. Fiz. 10, 233 (1969) [transl.: Sov. J. Nucl. Phys. 10, 133 (1969)].

[17]Y. A. Babenko, Y. A. Nemilov, L. A. Pleskachevskii, Y. A. Selitskii, and V. B. Funshtein, Yad. Fiz. 11, 1106 (1970) [transl.: Sov. J. Nucl. Phys. 11, 560 (1970)].

[18]J. R. Huizenga, A. N. Behkami, and J. H. Roberts, in *Proceedings of the Second International Atomic Energy Symposium on Physics and Chemistry of Fission, Vienna, Austria, 1969* (International Atomic Energy Agency, Vienna, 1969), p. 403.

[19]I. M. Kuks, V. I. Matvienko, Y. A. Nemilov, Y. A. Selitskii, and V. B. Funshtein, Yad. Fiz. 16, 438 (1972) [transl.: Sov. J. Nucl. Phys. 16, 244 (1973)].

[20]R. Vandenbosch, private communication.

[21]J. R. Huizenga, A. N. Behkami, R. W. Atcher, J. S. Sventek, H. C. Britt, and H. Freiesleben, USAEC Report No. LBL-2316, October, 1973 (unpublished).

[22]T. Dössing and A. S. Jensen, to be published.

[23]W. Loveland, J. R. Huizenga, A. N. Behkami, and J. H. Roberts, Phys. Lett. 24B, 666 (1967).
[24]H. Meldner and A. Z. Lindner, Z. Phys. 180, 302 (1964).
[25]R. Batchelor, W. B. Gilboy, and J. H. Townle, Nucl. Phys. 65, 236 (1965).
[26]A. Gilbert and A. G. W. Cameron, Can. J. Phys. 43, 1446 (1965).
[27]P. E. Vorotnikov, Yad. Fiz. 9, 303 (1969) [transl.: Sov. J. Nucl. Phys. 9, 179 (1969)].
[28]*Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report No. BNL-325 (U.S. Government Printing Office, Washington, D.C., 1958), 2nd ed.
[29]E. A. Zhagrov, Y. A. Nemilov, and Y. A. Selitskii, Yad. Fiz. 7, 264 (1968) [transl.: Sov. J. Nucl. Phys. 7, 183 (1969)].
[30]U. Mosel and H. W. Schmitt, Nucl. Phys. A165, 73 (1971).
[31]V. T. Ippolitov, Y. A. Nemilov, Y. A. Selitskii, and V. B. Funshtein, Yad. Fiz. 14, 939 (1971) [transl.: Sov. J. Nucl. Phys. 14, 526 (1972)].
[32]R. C. Kennedy, L. Wilets, and E. M. Henley, Phys. Rev. Lett. 12, 56 (1964).
[33]R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (Academic, New York, 1973), pp. 39–42, 50–51.
[34]L. G. Moretto, R. C. Gatti, S. G. Thompson, J. R. Huizenga, and J. O. Rasmussen, Phys. Rev. 178, 1845 (1969).
[35]J. Maruhn and W. Greiner, Phys. Rev. Lett. 32, 548 (1974).
[36]P. Decowski, W. Grochulski, A. Marcinkowski, K. Swjek, and Z. Wilhelmi, Nucl. Phys. A110, 129 (1968).
[37]M. Bolsterli, private communication.
[38]J. R. Nix, Nucl. Phys. 71, 1 (1965).
[39]R. Vandenbosch and U. Mosel, Phys. Rev. Lett. 28, 1726 (1972).
[40]T. Ericson, Nucl. Phys. 6, 62 (1958).