ASYMMETRY IN NUCLEAR FISSION ‡

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Received 17 December 1970

The deformation energies for fissioning nuclei in the actinide region are calculated using the Strutinsky shell-correction method. Instability of the second barriers against an asymmetric shape degree of freedom is found in agreement with Möller and Nilsson. The inclusion of these asymmetric deformations improves the agreement between calculated and experimental barrier heights. The asymmetries found have the correct magnitude in order to explain the experimental mass ratios of the fission fragments.

Strutinsky's suggestion [1] to split the nuclear binding energy into a smooth average and a rapidly varying shell correction energy provides a powerful tool for the calculation of nuclear ground state and deformation energies. The fact, that the shell correction energy defined in this method depends mainly on the single-particle levels near the Fermi energy, allows the use of relatively simple single-particle models. Such calculations have been performed by several groups using quite different potentials [2-6]. The ground state deformations of the nuclei in the regions of the lanthanides and actinides calculated by the different groups agree quite well and also reproduce the experimental results in a satisfactory way. For larger deformations, the results agree qualitatively in as much as all authors obtain a second local minimum in the deformation energy for the nuclei, of which a fission isomer has been observed. In some cases, however, the second barrier turned out to be too high as compared to the first barrier, when comparison was made to experiments [7].

So far, all calculations mentioned above [2-6] have been done for shapes of the nucleus, which are symmetric under reflection at a plane perpendicular to the fission axis. Recently, Möller and Nilsson [8] have reported on calculations with reflection-asymmetric shapes. They found for most of the actinide nuclei the outer saddle point of the energy surface to be unstable against a suitable combination of P_3- and P_5-deformations, while the inner barrier as well as the second minimum were stable against these deformations.

In the present letter, we present some similar and more complete calculations which support the results obtained by Möller and Nilsson [8].

We calculate the deformation energy W of a nucleus according to the prescription of Strutinsky [1] as

$$W = E_{\mathrm{LD}} + \delta U + \delta P. \tag{1}$$

E_{LD} is the liquid drop energy of the deformed nucleus - normalized to be zero at spherical shape - and δU and δP are the energy shell correction and the BCS-pairing correction, respectively, both being sums of proton- and neutron contributions. The single particle energy levels needed for the calculation of δU and δP were obtained by shell model calculations with a deformed Woods-Saxon potential [9]. For details of the liquid drop model and for some changes of the method described in ref. [9] for the calculation of the single-particle levels, we refer to a larger, forthcoming publication which collects and reports the results of the work done by the group in Copenhagen during the last two years [10].

The shape of the nuclear surface, which is supposed to be axially symmetric around the fission axis (z-axis), is described by the equation

$$\rho^2 = (C^2 - Z^2)[A + B(Z/C)^2 + \alpha(Z/C)] \tag{2}$$

is the usual cylindrical co-ordinates ρ, z, φ; ρ, z and c are measured in units of the nuclear radius $R_0 = r_0 A^{1/3}$. For $\alpha = 0$, the parameters A, B, c describe shapes which are symmetric under reflection at the plane $z = 0$, $2cR_0$ being the length of the nucleus along the z-axis. If $\alpha = B = 0$, eq. (2)

‡ Work supported by the Swiss National Foundation.

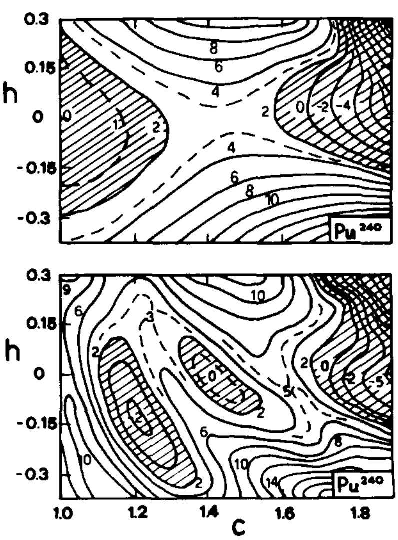

Fig.1. Potential energy surfaces of ^{240}Pu in the (c, h)-plane, calculated for symmetric shapes ($\alpha = 0$). Above: Liquid drop energy E_{LD}, normalized to zero at spherical shape ($c = 1$, $h = 0$). Below: Total deformation energy W [eq. (1)]. The equidistance of the solid lines is 2 MeV, regions below +2 MeV are shadowed.

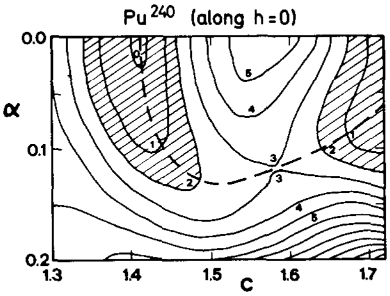

Fig.2. Deformation energy W of ^{240}Pu in the (c, α)-plane, calculated for h=0. The equidistance of the contour lines is 1 MeV. The dashed line shows the path, along which the energy is minimal in the α-direction.

describes pure ellipsoidal shapes with the two half-axis cR_0 and $cR_0\sqrt{A}$. $B > 0$ leads to necked-in and $B < 0$ to lemon-like shapes. Finally, for $A \leq 0$ and $B > 0$, the nucleus is separated into two fragments. The parameter α describes reflection-asymmetric shapes. The volume conservation condition reduces the number of free shape parameters from four to three by the relation

$$1/C^3 = A + \tfrac{1}{5}B \ . \qquad (3)$$

As the three free shape parameters we choose the elongation parameter c, the neck parameter h, defined by

$$h = \tfrac{1}{2}B - \tfrac{1}{4}(C - 1) \qquad (4)$$

and the asymmetry parameter α. The neckparameter h (4) is chosen in such a way, that the line $h = 0$ in the (c, h)-plane approximately fits the so-called liquid drop valley for the nuclei in the actinide region [10]. This can be seen in fig. 1, above, where we show the liquid drop energy surface of ^{240}Pu, calculated for symmetric shapes ($\alpha = 0$)

The lower map in fig. 1 shows the total deformation energy of the same nucleus, including the shell corrections according to eq. (1). One recognizes the ground state region with a local minimum of -2.5 MeV, the isomer minimum at ≈0.0 MeV and the two barriers with saddlepoints at +3 and +5 MeV. The barrier heights measured from the ground state are thus 5.5 MeV and 7.5 MeV for the inner and outer barrier, respectively, whereas an analysis of the experimental results by Bjørnholm [7] claims 5.8 MeV and 5.4 MeV, thus the outer barrier being even somewhat smaller than the inner one. This discrepancy results for most of the calculated actinide nuclei: the outer barrier is systematically too high. It should be noticed, that the barrier heights are mainly determined by the shell corrections, the liquid drop energy only amounting to about one third or less of the total fission thresholds. Therefore, the discrepancy mentioned above cannot be removed above cannot be removed by a new fit of the liquid drop model parameters.

The picture changes appreciably, if the reflection-asymmetric deformations ($\alpha \neq 0$) are also taken into account. In fig. 2 we show the contour map of the energy surface in the (c, α)-plane, calculated for a constant value of the neck parameter ($h = 0$). It can be seen that the shapes with $c < 1.4$ are stable against asymmetry (lowest energy at $\alpha = 0$). At $c \approx 1.4$, corresponding to a point near the second minimum, the instability onsets and increases strongly on the way up to the second barrier. The dashed path in the landscape of fig. 2 shows the locus of minimal deformation energies. It leads around the second saddle point, lowering the maximum by about 2.5 MeV. This result is qualitatively in agreement with the report by Möller and Nilsson [8].

It is, however, not certain at all, that the position of the second saddle point in the subspace of symmetric shape degrees of freedom remains the same when the asymmetric degree

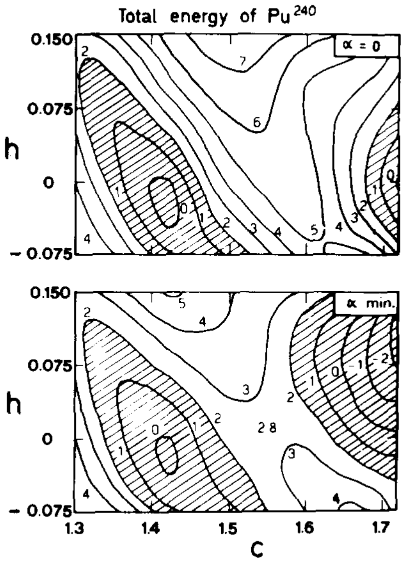

Fig.3. Deformation energy of ^{240}Pu in the (c, h)-plane. Above: Part of the energy surface shown in fig.1 (below) for symmetric shapes. Below: The same part of the surface, but the energy is minimized in each point (c, h) with respect to α. Equidistance of the contour lines: 1 MeV. The outer saddle point is lowered by the asymmetry by 2 MeV and shifted in the (c, h)-plane from the point (1.62, -0.05) to the point (1.55, 0.02).

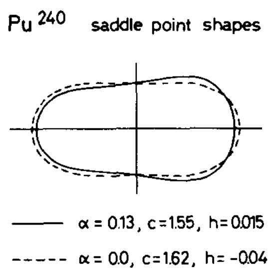

Fig.4. Shape of the nucleus ^{240}Pu at the second saddle point. Full curve: calculated for asymmetric deformations; dashed curve: for symmetric deformations. The values of the deformation parameters are given in the figure.

is taken into account. Thus one has to perform the same calculations for all values of the neck parameter h within the region of interest, i.e., one has to minimize the energy with respect to α in all points of the plane (c, h) of symmetrical deformations. In fig. 3 we present the result of such a calculation for ^{240}Pu. The upper map shows a part of the energy surface for symmetric shapes ($\alpha = 0$). A region of deformation is chosen which contains the second minimum and the outer barrier. The lower part of fig. 3 shows the same part of the energy surface, but here the energy is minimized with respect to α in each point (c, h). The minimum region is not affected by the assymmetry, i.e., it is stable. At larger deformations, however, the energy is lowered by several MeV. Due to the asymmetry of the shape, the outer saddle point decreases by about 2 MeV and has now - with 2.8 MeV - about the same energy as the inner saddle point, which is in close agreement with ref. [7]. It should also be realized that the position of the outer saddle point in the (c, h)-plane is shifted by the asymmetry towards a deformation with smaller elongation and smaller neck radius.

In fig. 4 we present the two shapes at the second saddle point of the ^{240}Pu nucleus, as calculated for symmetric (dashed line) and asymmetric (full line) deformations, respectively.

We did the same calculations for a series of other nuclei in the actinide region. Summing up the results, we can state the following points:

(i) The symmetric shapes in the whole region from the ground state up to the isomer state (second minimum) are stable against our asymmetric deformations for all actinides.

(ii) The shapes in the region of the second barrier and beyond it (scission region) are unstable against asymmetric deformations for all actinides heavier than ^{228}Ra. The saddle point energy is lowered by up to 3 MeV. The position of the outer saddle point in the (c, h)-plane is, throughout the actinide region, shifted towards a deformation with smaller elongation (c) and smaller neck radius (larger h).

(iii) The relative heights of the two barriers are now in much better agreement with experiment than for symmetric shapes alone.

(iv) For ^{228}Ra, the region of ascent between the second minimum and the second barrier is slightly unstable against asymmetry, but the second saddle point is almost and the region beyond it is completely symmetric. The same is the case for ^{210}Po, but there the second saddle point is completely stable.

(v) The value of the asymmetry parameter α, at which the energy is minimal, is in all cases not larger than $\alpha = 0.2$.

It should be noted, that the instability of the outer barrier for the actinides, found in our calculations as well as in ref. [8], is due only to shell effects. It is a well-known feature of the liquid drop model to favour reflection-symmetric shapes (see, e.g., refs. [11, 12]).

It is an important point to investigate, how these results are related to the mass ratios of the fission fragments. A rigorous answer to this question can, of course, only be given in a dynamical treatment of the fission process.

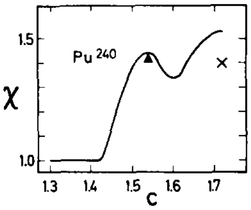

Fig.5. Estimated mass ratio χ [eq. (5)] of ^{240}Pu. plotted against c along a straight line from the second minimum through the outer saddle in the minimized energy surface (lower map in fig. 3). A triangle marks the saddle point; the cross on the right-hand side shows the experimental mass ratio $(m_H/m_L)_{exp}$ of the fission fragments.

Nevertheless, one can try to estimate roughly the mass ratios using our results of the potential deformation energy only. There are reasons to believe that scission of the nucleus occurs at deformations with an elongation $c \approx 1.6 - 1.7$ [10,12]. For these shapes, the mass ratio of the forming fragments may roughly be approximated by the quantity

$$\chi = \int_{-c}^{0} \rho^2(Z)\,dZ \Big/ \int_{0}^{c} \rho^2(Z)\,dZ = (1+\tfrac{3}{8}\alpha c^3)/(1-\tfrac{3}{8}\alpha c^3),$$

which is defined as the ratio of the two parts of the nucleus obtained by intersecting it by the plane $z = 0$. Fig. 5 displays χ as a function of c, evaluated along a straight line in the asymmetric energy surface of ^{240}Pu (lower map in fig. 3), which connects the second minimum with the outer saddle point. The curve rises steeply from its initial value $\chi = 1.0$ to the value at the saddle point - marked by a triangle - and then fluctuates weakly around a value of $\chi = 1.43$, being in reasonable agreement with the experimental value of (m_H/m_L), marked by a cross. Similar calculations were done for 10 other fissioning nuclei. The results are compiled in table 1. The second column shows the values of χ found at the outer saddle point, the third column contains the average values of χ obtained in the scission region ($1.6 \lesssim c \lesssim 1.7$). The fluctuations around these values, due to variations of the trajectories chosen in the energy surface, are indicated by the error limits. The experimental values in column 4, taken from ref. [13], lie within these limits for all the investigated nuclei except ^{232}Th. However, the absolute values of the estimated mass ratios may be less significant than their systematics: The sudden transition from symmetry to asymmetry around ^{228}Ra and the slow decrease of the mass ratios of the

Table 1

Nucleus	χ_{saddle}	$\chi_{scission}$	$(m_H/m_L)_{exp}$
^{210}Po	1.0	1.0	1.0
^{228}Ra	1.11	1.0	1.0/1.5
^{232}Th	1.40	1.37 ± 0.06	1.46
^{236}U	1.39	1.43 ± 0.09	1.46
^{240}Pu	1.44	1.43 ± 0.09	1.40
^{244}Cm	1.42	1.39 ± 0.07	1.32
^{248}Cf	1.37	1.37 ± 0.06	1.31
^{252}Cf	1.33	1.33 ± 0.03	1.33
^{252}Em	1.20	1.34 ± 0.05	1.29
^{256}No	1.10	1.32 ± 0.04	-
^{260}Ku	1.0	1.32 ± 0.04	-

Theoretical estimates and experimental values of the mass ratios of fission fragments in the actinide region. Column 2: Values of χ [eq. (5)] evaluated at the second saddle point in the minimized energy surface. Column 3: Average values of χ in the scission region (see text). Column 4: Experimental peak-to-peak ratios of the fragment mass distributions, taken from ref. [13]. (No difference has been made between pre- and post-neutron emission measurements.)

nuclei heavier than ^{240}Pu are well reflected in these results.

As a conclusion, we can say, that there is a clear shell structure in deformation energy also with respect to reflection-asymmetric shape distortions. Including this effect, the calculated heights of the fission barriers in the actinide region can be appreciably improved. An estimate shows, that the asymmetrics found have also the correct magnitude in order to explain the experimental mass ratios of the fission fragments.

Two of us (H.C.P. and M.B.) are greatful to Professor V. Strutinsky for many stimulating discussions during their stay at the Niels Bohr Institute in Copenhagen. We thank the SANDOZ A.G., Basel, for the grant of computational time on an UNIVAC 1108.

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