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SPONTANEOUS-FISSION INERTIAL-MASS FUNCTIONS IN THE ACTINIDE REGION

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Abstract

Recently obtained¹) deformation-energy surfaces, based on the droplet model²) and the modified-oscillator single-particle model³), have been used to establish spontaneous-fission potential barriers for even-even nuclei with Z between 92 and 106. Effects of axially asymmetric shapes in the deformation region around the first saddle point⁴) as well as of reflexion asymmetric shapes for large values of ε_2^{-1}) have been taken into account. Furthermore, the effect of P₆ deformations⁵) have been included in the ground-state energies. The recently discovered⁶) double-hump structure of the second barrier for U and Pu isotopes has also been taken into account.

Following the semi-empirical method introduced in ref. ⁷), we use these barriers in connection with the experimental spontaneous-fission half-lives⁸), to determine a smooth inertial-mass function. The established mass function reproduces the half-lives equally good as previously⁷) but it is shifted somewhat towards larger values.

As an alternative we have used theoretical cranking-model inertial-mass parameters⁹) to calculate the spontaneous-fission half-lives. It is found that when renormalized by a factor of around 0.60 these inertial masses give results similar to those corresponding to the smooth inertial function. This important finding supports the consistency of the theoretical approach and justifies the use of the method for predicting spontaneous-fission half-lives for so far unobserved nuclei.

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1. Introduction

Recently¹), improved deformation-energy surfaces have been obtained for nuclei in the actinide region. Our initial semi-empirical study of the spontaneous-fission inertial-mass functions⁷) was based on somewhat older material and it would be desirable to perform a similar analysis for these new results. As was pointed out in ref.⁷), the determined fission inertialmass functions may to some extent absorb possible overall errors in the theoretical barriers. Hence it is necessary to redetermine the inertias before making half-life predictions on the basis of the new barriers. Such a redetermination would also give an indication of the uncertainties associated with the semi-empirical mass functions and provide us with a check of our previous results. This is very important for the application of the method for making half-life predictions for more distant nuclear regions as for example the super-heavy elements. Furthermore, the analysis will constitute an important check of the established theoretical fission barriers and may indicate possible deficiencies in this material.

In section 2 we describe how the spontaneous-fission potential barriers are established from the available theoretical material. Subsequently, in section 3, we present the results of the numerical half-life calculations together with a discussion of the importance of the various possible types of corrections to the barriers. In section 4 we describe how a similar analysis may be carried through on the basis of theoretical cranking-model inertial-mass parameters. Finally, in section 5, the results of the investigation are summarized.

2. The Theoretical Fission Barriers

In fig. 1 we have indicated which nuclei we have considered. They form a group of 43 even-even nuclei with Z between 92 and 106. Among these, 34 nuclei have so far been observed and, except for the two Rf isotopes, rather accurate spontaneous-fission half-lives determined⁸). These half-lives are displayed in fig. 2.

The deformation-energy surfaces upon which this investigation is based are those obtained by $M\ddot{o}ller^1$) in the Spring of 1973. For the macroscopic part of the deformation energy the newly released droplet model²) has been used. The shape functionals B have been obtained by exact integration in the ε parametrization. The microscopic-correction energy is based on the modified-oscillator single-particle model³) with potential parameters corresponding to A = 242. The pairing strength is assumed independent of deformation, G \sim const.

According to the semi-empirical method introduced in ref. ') the fission barrier is established from the knowledge of the stationary points together with one or more additional points outside the last saddle point. Thus, we have extracted the following information.

I) Ground state: $\varepsilon_2^{I} \varepsilon_4^{I} v^{I}$

The ground-state energy v^{I} may be subject to a correction due to P_{6} deformations, as calculated by Möller⁵) for the A = 255 parameters. (This should be good enough.)

A) First saddle: $\varepsilon_2^A \varepsilon_4^A V^A$

The saddle-point energy v^A is subject to a correction due to γ deformations, as obtained by Larsson⁴) employing a three-dimensional variation in ($\varepsilon_2 \varepsilon_4 \gamma$) space. These calculations use the droplet model and G \sim const. For most (i.e. all but the Rf and 106 isotopes) of the nuclei considered there is also a secondary minimum; in that case we also extract the following three sets of information.

II) Second minimum: $\epsilon_2^{II} \epsilon_4^{II} v^{II}$

Since we have not in this investigation studied the isomeric half-lives it is not important to consider corrections to this point.

B) Second saddle: $\epsilon_2^B \ \epsilon_4^B \ v^B$

In the calculation of these values, reflexion asymmetric shapes have been taken into account by performing an ($\varepsilon_{24}^{}$, $\varepsilon_{35}^{}$) variation¹).

X) Exit region: $\varepsilon_2^x \varepsilon_4^x V^x$

In accordance with ref. ⁷) we choose $\varepsilon_2^{\mathbf{x}} = 1.0$, $\varepsilon_4^{\mathbf{x}} = 0.14$. The energy $V^{\mathbf{x}}$ is sensitive to the asymmetry ε_{35} and we have studied in particular the two possibilities of $\varepsilon_{35} = 0$ or ε_{35} equal to whatever yields the minimum value for $V^{\mathbf{x}}$ for ε_{24} fixed to the prescribed value.

For the (Rf and 106) nuclei without a secondary minimum we choose some (i.e. three) points along the ϵ_{24} path beyond the first saddle. The results are not very sensitive to the actual choice. Recent theoretical calculations⁶) indicate a more complicated structure of the second-barrier region for the U and lighter Pu isotopes. The modified-oscillator results¹), supported by the folded-Yukama model⁶), suggest that the second barrier for these nuclei has two humps separated by a shallow minimum. We have also taken this possibility into account in the established barriers.

As described in ref. ⁷) and illustrated in fig. 3, the one-dimensional fission-barrier potential is obtained by first transforming the ε points to the fission coordinate r (the fragment separation) and subsequently splining appropriate polynomials through the characteristic points described above. In the present investigation we have disregarded the influence of ε_4 distortions on the r coordinate, since it has proven of minor importance to the ground-state fission half-lives⁷). Before evaluating the penetration integral we have added a ground-state vibrational energy of $E_{vib} = 0.5$ MeV, as indicated in fig. 3.

3. The Numerical Calculations

Following ref. ⁷) we employ the following type of smooth trial inertial-mass function

$$B_{r} = \mu \left(1 + k \frac{17}{15} e^{-(r - \frac{3}{4} R_{o})/d} \right)$$
(1)

Here μ is the reduced mass of the final two-fragment system and d is the inertia associated with a rigid separation; R_0 is the nuclear radius, $R_0 = r_0 \cdot A^{1/3}$. Thus, we have two adjustable parameters, namely the amount k of non-rigid inertia and the width d of this part of the mass function. The irrotationalflow inertial function is obtained for k = c = 1, where $c = d/d_0$, $d_0 = R_0/2.452$ being the width of this function. We shall at first keep c fixed to this value of unity but will study the variation of this parameter as well later on. In fig. 4 we have shown B_r for the ensuing optimum value of the parameter set (k,c).

With the trial function corresponding to c=l we have calculated the spontaneous-fission half-lives for the considered group at nuclei, with particular attention to the effect of the various types of correction to the fission barriers. Figures 5-8 show the results of a stepwise inclusion of the various refinements.

First we have taken the barriers obtained when only refinements considered in ref.⁷) are included. This means ground state without P_6 correction, first saddle with γ correction, second saddle with P_{35} correction and exit energy without P_{35} correction. The results of this basic run are displayed in fig. 5. There is rather bad agreement with experiment, the average logarithmic deviation in the best case being $\Delta = 4.0$. The average behavior is somewhat wrong and the isotopic variation for the lighter elements U - Cf is very bad. The best inertial-mass function is obtained for k = 8.0 which is somewhat larger than the value of k = 6.5 obtained in ref.⁷). As already pointed out in that reference, such a renormalization may be expected when a new set of barriers is used in the fit.

Figure 6 shows the similar results after the exit energy V^{X} has been corrected for P₃₅ deformation. As is seen, this modification improves the fit considerably. Notice that the erroneous average behavior has improved and that especially the isotopic variation for Cm and Cf has improved. For the heavier nuclei, except 252,254,256 Fm, the penetration integral is unaffected by this modification. The best fit $\Delta = 2.8$ is obtained for k = 10.0. The considerable improvement of the fit (more than a factor of 15) indicates the importance of the reflexion asymmetry in the exit region. In the following, we have, therefore, kept this correction throughout.

The effect of the subsequent inclusion of P_6 ground-state corrections is shown in fig. 7. The fit improves to $\Delta = 2.4$ as obtained for k = 8.0. Notice the very good agreement for the light Fm isotopes (two of them, 244,246 Fm, were not included in our previous studies). It is seen that this modification does not influence the lighter elements very much, in particular it does not remove the bad isotopic variation. But there is an appreciable improvement and we shall keep the P_6 corrections from now on. Finally, the possibility of a triple-hump barrier has been considered. This concerns only the U and Pu isotopes (except for 244 Pu). We have modified the second barrier of these nuclei according to the results obtained by Moller¹) and the results are displayed in fig. 8. There is a remarkable improvement for all of these isotopes except 242 Pu. These isotopes are now in good agreement with experiment, and only the two isotopes 242,244 Pu are now badly reproduced. We shall return to this point later on. The best inertia is still k = 8.0 for which parameter we obtain $\Delta = 1.9$. This is a rather satisfactory agreement for the 32 nuclei included in the fit.

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For the barriers obtained with the modifications described above we have also studied the effect of varying the slope of the inertial-mass functions, as described by the width parameter c. We have established a contour plot of $\Delta(k,c)$ as was done in ref.^{7a}). It exhibits a very similar structure, with a narrow valley extending up-right. The whole structure is, however, displaced towards larger values of k and the valley bottom now follows a line through the points (5.0,0.6) and (8.0,1.0) (previously this minimum line was c = 0.15 k). As previously⁷) the optimal position of (k,c) in this valley is not very well determined, but this time the shallow minimum is positioned rather close to c = 1, the hydrodynamical slope. This supports the preference of this value as made in ref.^{7a}) on the basis of the isomeric half-lives. The optimal parameter set is thus very close to (k = 8.0, c = 1.0).

4. Microscopic Inertial-Mass Parameters

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As an alternative to the smooth inertial-mass function used above we have employed the microscopic inertial-mass parameters calculated by Sobiczewski⁹). That will also provide a check of the consistency of the whole theoretical approach. Sobiczewski⁹) has calculated the inertial-mass parameters B_{QQ} in a modified cranking model. The subsequent transformation to B_{EE} was done by using the microscopically calculated values for the quadrupole moments Q(E). This is more consistent than the usual assumption of a quadrupole moment corresponding to a uniform charge distribution. We use the results obtained for a constant pairing interaction, $G \sim \text{const.}$

These mass parameters were calculated along a path in the $(\varepsilon_2 \varepsilon_4)$ plane and are given in the form of a table with $\varepsilon_2 = 0.0(0.1)1.0$. From this material we generate continuous inertial-mass functions by linear inter(extra)polation. A problem arises, however, for $\varepsilon_2 > 1.0$, since such a prescription may lead to negative values of the inertia for large ε_2 . We have, therefore, introduced an arbitrary minimum inertial mass $B_{<}$ below which the inertial function is not permitted to come. As discussed below, we have investigated the sensitivity of the results of the value chosen for $B_{<}$.

The theoretical inertial-mass parameters are expected to yield correctly the variation with deformation and particle number, but overestimate the absolute values. We have, therefore, introduced one overall renormalization factor ρ for these mass parameters and subsequently fitted this one parameter to the experimental material. From our previous investigations⁷), we expect this factor to be around one half. In fig. 4 we have, as an illustration, shown the employed microscopic inertial-mass function for ²³⁶Pu corresponding to the obtained optimal renormalization factor. The transformation to the r coordinate is performed as described in ref.⁷). Figure 9 shows the results of this calculation. As is seen, the overall reproduction of the experimental half-lives is good ($\Delta = 2.06$). The best fit is obtained for a renormalization factor of $\rho \approx 0.6$. We have tried three different values for $B_{<}$: 0, 100 and 200 and the results are somewhat sensitive to the choice, especially for the lighter elements which have a considerable barrier extending into the involved region. The results are best for $B_{<} = 200$ which value also corresponds most to the obtained smooth inertia in this region, c.f. fig. 4. It seems as if the employed microscopic inertial functions are too steep, being unreasonably small for large values of r (in fact smaller than the irrotational or even the rigid inertias) and having a tremendous peak near the spherical shape. This latter peak does not come into play, however, because the employed zero-point energy of 0.5 MeV shifts the entrance value of r sufficiently outwards.

Comparing now these results with the ones for the smooth inertia, we find that on the whole the systematics of the deviations are quite similar to each other. This fact can be considered an indication of certain minor errors associated with the established theoretical fission barriers. In particular, we notice the same drastic discrepancy for 242,244 Pu, indicating that the theoretical barriers used for these two isotopes are incorrect; this discrepancy can probably be removed to a considerable extent when more accurate maps of the complicated second-barrier region for these nuclei are available. (The same type of discrepancy seems to be present for 244,246 Cm but to a much less extent.)

5. Conclusions

The analysis presented here supports the method introduced in ref. ⁷). That investigation concluded that the most reasonable smooth inertial-mass function is of the hydrodynamical type (i.e. c = 1), with a renormalization factor of k = 6.5. The present investigation supports the hydrodynamical-type mass function, but finds a somewhat larger overall value, k = 8.0. This is, of course, very important for the application to half-life predictions for distant nuclei, as for example the super-heavy region.

With the determined inertial-mass function it is possible to reproduce the 32 experimental spontaneous-fission half-lives to within a factor of 80 on the average. As explained in the analysis, there are indications of the barriers of 242,244 Pu being incorrect. If these two nuclei are disregarded the fit improves considerably, $\Delta' = 1.4$. This corresponds to a factor of 25 which is similar to our previous fit⁷). These half-lives are shown in fig. 2 together with those calculated for the so far unobserved nuclei we have studied.

As an interesting alternative, we have also calculated the spontaneousfission half-lives on the basis of completely theoretical fission inertial-mass parameters⁹), modified with one overall renormalization factor ρ . The best fit is obtained for $\rho = 0.60$ and the results are rather similar to those obtained with the smooth inertial-mass function. This finding is an important check of the consistency of the theoretical approach and justifies the application of the method to half-life predictions for so far unobserved nuclei.

Acknowledgments

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References

1.	P. Moller, private communication (1973), to be published.
2.	W. D. Myers and W. J. Swiatecki, LBL-1957 (1973), to be published
3.	S. G. Nilsson, C. F. Tsang, A. Sobiczewski, Z. Szymanski, S. Wyeech,
	C. Gustafson, I. L. Lamm, P. Moller and B. Nilsson, Nucl. Phys. A131 (1969) 1
4.	S. E. Larsson, Physics and Chemistry of Fission, IAEA, Rochester, 1973
5.	P. Möller, private communication (1972), not published
6.	P. Moller and J. R. Nix, Physics and Chemistry of Fission, IAEA, Rochester,
	1973
7.	a) J. Randrup, Lawrence Berkeley Laboratory Preprint, LBL-1699 (1973);
	b) J. Randrup, C. F. Tsang, P. Moller, S. G. Nilsson and S. E. Larsson,
	Nucl. Phys. A217 (1973) 221
8.	Compilations by K. Hulet, E. Hyde, K. Vandenbesch and V. Viola and
,	S. V. Jackson, private communications (1972).

9. A. Sobiczewski, private communication (1973), to be published

Figure Captions

- Fig. 1. The even-even nuclei considered in this investigation. The so far observed nuclei are shaded.
- Fig. 2. Spontaneous-fission half-lives. Full circles: experimental values ⁸). Open circles: calculated with the determined smooth inertial-mass function (k = 8.0, c = 1.0).
- Fig. 3. Illustration of the reduction from the multi-dimensional fission problem to a one-dimensional barrier penetration.
- Fig. 4. Inertial-mass functions for 236 Pu. The smooth curve is the determined mass function corresponding to (k = 8.0, c = 1.0) while the kinked curve represents the optimally renormalized cranking-model mass function (ρ = 0.60). The various choices of the minimum inertia B_< are shown and the entrance and exit values r_< and r_> are indicated.
- Fig. 5. Results of the basic run when no barrier modifications have been made apart from what was done in ref. ⁷). The figure is a logarithmic plot of the differences between calculated and experimental spontaneous-fission half-lives. The mass function used corresponds to k = 8.0 (and c = 1 as in the following three figures).
- Fig. 6. As fig. 5 but including reflexion-asymmetry corrections of the exit energies V^{X} . For the best value k = 10.0.
- Fig. 7. As fig. 6 but including also P_6 corrections of the ground-state energies V^{I} . For the best value k = 8.0.
- Fig. 8. As fig. 7 but including also triple-hump barriers for the U and Pu isotopes, except for 244 Pu. For the best value k = 8.0.
- Fig. 9. As fig. 8 but using instead the renormalized cranking-model mass functions corresponding to the best choice $\rho = 0.60$, $B_s = 200$.

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Fig. 1

XBL 743-2535

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XBL 743-2534







XBL738-3679

Fig. 3

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XBL 743-2533

Fig. 4





XBL 743-2532

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LBL-2385



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XBL 743-2530

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Fig. 7

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Fig. 8

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XBL 743-2528

Fig. 9

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