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A SEMI-EMPIRICAL APPROACH TO FISSION INERTIAS AND FISSION HALF-LIVES*

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May 1973

Abstract

The problem of determining the inertia associated with the nuclear spontaneous-fission process and calculating the corresponding half-lives is approached by a semi-empirical method. For the purpose of determining an effective fission inertia a test group of (31) known even nuclei, ranging from U to Rf/Ku, is selected. For this group, appropriate fission paths, together with the fission-barrier potentials along these paths, are established on the basis of potential-energy surfaces which have been obtained by the macroscopic-microscopic method. The macroscopic part of the energy is provided by the liquid-drop model of Myers and Swiatecki, modified however to reproduce well the experimentally known second barriers in the actinide region. The microscopic correction-energy contribution to the potential-energy surfaces is based on the modified-oscillator single-particle model in which allowance has been made for mass-asymmetric (P_3 and P_5) as well as axial asymmetric (γ) distortions. Indications from existing hydrodynamical and microscopic calculations of the nuclear inertia as well as from the known isomeric half-lives are used to construct a simple one-parameter smooth trial inertia which is then fitted to the half-lives for the test group.

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The equivalent center-of-mass separation r (corresponding to ellipsoidal shapes and equal-mass fragments) proves to be a preferable fission-path coordinate. The inertia obtained in this way reproduces those half-lives to within a factor of 25 on the average which is superior to any other existing method. Furthermore, the general trend of the calculated half-lives supports the extrapolation of the established inertial function to so far unobserved nuclei. Some part of the deviations from the experimental results can be understood in terms of certain deficiences in the underlying potential-energy surfaces, and refinements of the approach (such as taking into consideration the multi-dimensionality of the fission path, or allowing for more freedom in the trial inertia) show no considerate improvement of the fit. Applications (reported elsewhere) have been made to several problems of current interest, as for example, the predictions of fission half-lives for isotopes of the element 106 (and heavier elements), the calculation of the hindrance factors associated with fission of odd isotopes, and the possibility of a neutron-capture path to the superheavy region.

1. Introduction

The calculation of spontaneous-fission half-lives is a central problem in the physics of heavy nuclei and has particular importance for the understanding of the synthesis of heavy elements. However, until recently, calculations[1-4]), have not been able to reproduce the experimentally known half-lives better than to within a factor of typically 10^5. In recent years, more refined calculations have been made of the fission potential barriers[5-7]) and this part of the problem seems by now rather well understood. In order to calculate the fission half-lives, however, also the knowledge of the associated inertial-mass function is required. Several theoretical calculations of the fission inertial masses have been carried out[2,3,8]), but the detailed and unrenormalized applications appear to yield rather erroneous half-lives. Here we shall attempt the inverse approach to determine an effective fission inertial-mass function from the by now rather rich experimental information in combination with the theoretical barriers. One may hope by such an approach to determine an inertial function with a simple distortion dependence which is able to reproduce well the main trends of the known half-lives. This would provide us with a basis for what seems a relatively reliable extrapolation to other nuclei.

The fission inertial masses are expected to exhibit odd-even variations and since furthermore the theoretical fission barriers for odd nuclei are associated with larger uncertainty we shall in this first approach restrict the considerations to even nuclei. We are particularly interested in the application to the heavy and super-heavy region, hence we shall choose the experimental test group of nuclei from the region of transuranium elements. From this region

we have chosen 31 even nuclei, ranging from ^{232}U up to $^{258}Ku/Rf$. Their measured spontaneous-fission half-lives[9] are displayed in fig. 1. As is seen from this figure the half-lives show large variations through the region, as well as from one element to another as within a single element. Thus, these nuclei seem to provide a rather crucial test group for the calculations.

The nuclear distortion energy forms a multi-dimensional surface in the space spanned by the distortions parameters. Hence, the proper treatment of the fission problem requires the knowledge of the corresponding complete inertial-mass tensor. However, we shall here assume that the situation may be reasonably well described as a one-dimensional barrier-penetration problem. Thus, we consider a one-dimensional fission path through the multi-dimensional distortion space and the associated effective inertia for motion along this path, as illustrated in fig. 2. Furthermore, we shall assume that the corresponding half-lives may be obtained within the framework of ordinary WKB theory. Thus, denoting the fission-path coordinate by s, the action integral along the fission path is

$$K = \frac{2}{\hbar} \int_{s_0}^{s_{exit}} \sqrt{2B(s)\,(V(s) - E_{vib})}\, ds \quad . \tag{1}$$

E_{vib} denotes the fission-mode vibrational energy in the initial nuclear state and $V(s) - E_{vib}$ is thus the potential barrier along the fission path; $B(s)$ is the corresponding inertial-mass function. In terms of the action integral K the half-life is simply given by[1]

$$t = \frac{\ln 2}{n}\,(1 + e^K) = 10^{-28.04}\,(1 + e^K) \text{ years} \quad . \tag{2}$$

Here n is the number of assaults per unit time and has been taken to correspond to the standard choice of the vibrational energy: $E_{vib} = 1$ MeV. Since n enters only as a multiplicative factor, its value is not critical in the present context.

It may be useful to keep in mind how errors in the potential and/or the inertia will affect the corresponding half-lives. We find from (2) the following expression

$$\delta \log t \simeq 1/2 \left[\frac{\delta(V - E_{vib})}{V - E_{vib}} + \frac{\delta B}{B} \right] (28.04 + \log t), \text{ t in years} \qquad (3)$$

from which such estimates can be made. Thus, for example, an error of 10% in either the barrier or the inertia will typically give rise to a factor-of-error of 10^2 in the half-life.

2. Fission Barriers

The fission potential barriers applied in the present investigations are based on the macroscopic-microscopic method in which the smooth part of the distortion energy is obtained from a macroscopic model while the fluctuating part is associated with the shell structure. It is obtained from the application of the Strutinsky smoothing procedure to the single-particle level scheme calculated for the nuclear shape in question.

The macroscopic model was chosen as the standard liquid-drop model of Myers and Swiatecki[10]), however with the surface-energy parameters modified to reproduce the experimentally known heights of the second barriers in the actinide region. Such a modification has been suggested by Pauli and Ledergerber[11]). Thus, the shape dependence of the macroscopic energy is assumed to be

$$E_{LD}\,(\text{shape}) = a_2\,(1 - \kappa_s I^2)\,A^{1/3}\,B_s(\text{shape}) + c_3\,\frac{Z^2}{A^{1/3}}\,B_c(\text{shape}) \qquad (4)$$

where the nuclear asymmetry is $I = (N - Z)/A$ and $c_3 = \frac{3}{5}\frac{e^2}{r_0}$. B_s and B_c represent the shape variations of the surface and Coulomb energies relative to a spherical shape. Here we have used the modified values of a_2 and κ_s obtained in ref. [12]):

$$\kappa_s = 4.02 \text{ MeV} \quad (\text{from } 1.68 \text{ MeV})$$

$$2a_2/c_3 = 57.99 \qquad (\text{from } 51.0)$$

$$c_3 = 0.720 \text{ MeV} \quad (\text{assumed unchanged}) \quad .$$

In the determination of these values the microscopic model described below has been used, and they yield an excellent fit to the experimental data, see Fig. 2 of ref. [12]). One should observe the large change in the surface symmetry-energy coefficient κ_s. It may be mentioned already here that the transition from the standard coefficients to the modified ones improves the half-live fit considerably, especially the isotopic variation.

For the microscopic model we have used the modified-oscillator single-particle model introduced by Nilsson et al.[13]). Basically we have initially only considered the $(\varepsilon_2\varepsilon_4)$ degrees of freedom, but, as will be described below, we have subsequently taken into account the essential effects of other significant distortions.

The fission potential has (usually in this region) a first and a second minimum, (I and II, respectively), separated from each other by the first saddle (A) and from the exit region (X) by the second saddle (B). The actual fission path through this multi-dimensional landscape is assumed to be that yielding the least action[2]) and is thus not well determined a priori. We expect, however, that the fission path passes in close vicinity to the mentioned stationary points. (One should bear in mind that only these stationary energies have a unique character, i.e. are invariant under a coordinate transformation). We shall therefore characterize the fission barrier by the four extremum points (I, A, II and B) together with a fifth point in the exit region (chosen to lie approximately on the liquid-drop fission path).

Having obtained these five points in the multi-dimensional distortion space we project onto the axis of spheroidal deformation in order to reduce the problem to one dimension. The fission-barrier potential curve is then subsequently generated from these points by a simple spline method as illustrated in fig. 3. However, the ε coordinate has a singular behaviour for large distortions and thus the corresponding method seems not ideally suited for a physically lucid description of the fission problem. Following a suggestion by Swiatecki[14]), we choose instead the equivalent center-of-mass separation r; obtained from ε by the simple transformation

$$r = \frac{3}{4} R_0 \left(\frac{1 + 1/3\varepsilon}{1 - 2/3\varepsilon}\right)^{2/3} , \qquad R_0 = r_0 A^{1/3} . \tag{6}$$

This formula is valid for purely ellipsoidal shapes and equal-mass fragments; the inclusion of other degrees of freedom in this transformation is straightforward but turns out to be unimportant for the normal fission half-lives. The r coordinate has a more intuitively appealing asymptotic behaviour. As can be seen from fig. 4, the transformation from ε to r gives rise to a stretching of the barrier, giving an appropriately larger weight to the region of large distortions. One might argue that an even better choice of metric is one in which the inertial mass is independent of the distortion.

The positions of the five characteristic points are obtained from a calculation in $(\varepsilon_2\varepsilon_4)$ space. Other degrees of freedom may then be taken reasonably well into account by just correcting the relevant characteristic points before the splining is performed. It is well-known that the second-saddle energy is lowered significantly when allowance is made for asymmetric

distortions (in this model described in terms of the ε_3 and ε_5 coordinates). The effect of these distortions has been studied extensively by P. Moller[6]), and we have used his results for correcting E_B. These corrections are of the order of 1 MeV, typically, and it should be mentioned that this lowering of the saddle energy was taken into account when the liquid-drop parameters were refitted. In the first-saddle region, axial asymmetric (γ) distortions are important. This phenomenon has been studied by S. E. Larsson[7,15]), and we have used his results for correcting E_A. The corrections are 0.5 - 1.5 MeV throughout the actinide region and exhibit strong shell fluctuations. Furthermore, the possibility of P_6 distortions in the ground state has been investigated, but was found to be less important (they are always smaller than 0.75 MeV[16]).

3. Fission Inertias

We now turn to the problem of determining an effective inertial-mass function by fitting some suitable trial inertia to the experimental half-lives of the nuclei in the test group.

Hydrodynamical calculations of the fission inertia[4,17]) (in terms of the r coordinate and under the assumption of y-family shapes) yield an inertial function which decreases from the value of $\frac{32}{15}\mu$ for spherical shapes to the asymptotic value μ throughout the barrier region (cf. fig. 4), μ being the reduced mass of the final two-fragment system. Since these calculations have been based on the assumption of irrotational flow of the nuclear matter, they underestimate considerably the true inertial mass. However, we expect that the suggested gross shape of the inertial function is more reliable.

More realistic indications of the absolute magnitude of the inertial mass are provided by microscopic (cranking-model) calculations[8,3,18,19]). Calculations of such type yield inertial masses exhibiting fluctuations with distortion, reflecting the specific single-particle structure of the particular nucleus under consideration. Since these wiggles are difficult to parametrize, and in addition may tend to cancel because of their relatively short period, we have confined ourselves to considering only a smooth trial inertial function. In fig. 4 we show an example of such microscopic inertias (obtained recently by Krumlinde, ref. [18]), together with the corresponding hydrodynamical inertia. Figure 5 also displays recently calculated values obtained by Sobiczewski[19]) in a quasi-self-consistent model. It should be observed how well these latter calculations agree with the semi-empirically determined magnitude of the inertial

mass. From these figures we get a clear indication of the general behaviour of the fission inertial mass: it is considerably larger than the irrotational mass, but has the same type of gross decrease. This common indication of a relatively small inertial mass in the second-barrier region is also supported by the shortness of the fission half-lives of the shape isomers in the actinide region.

These considerations lead us to consider a trial inertial function of the following type

$$B_r = B_r^{rigid} + k(B_r^{irrot} - B_r^{rigid}) \tag{7}$$

where $B_r^{rigid} = \mu$ is the mass corresponding to a rigid separation of the two fragments, and B_r^{irrot} is the mass corresponding to irrotational flow during the fission process. Thus k is an adjustable parameter describing the contribution of the inertial mass from the internal nuclear motion, k being unity for purely irrotational flow. As mentioned above, we expect from the microscopic calculations k to be considerably larger than that. This inertial function is of the same type as was used by Nix *et al.*[4]). For simplicity we shall here assume equal-mass fragments and furthermore approximate the difference multiplying k by an exponential. The explicit form of B_r thus becomes

$$B_r = \frac{M}{4}\left[1 + k\,\frac{17}{15}\,e^{-(r - 3/4\,R_0)/d}\right] \quad . \tag{8}$$

Here M is the mass of the fissioning nucleus and accounts for the general scaling property of the inertial mass. The fall-off parameter d is taken to be that of the irrotational inertia, $d = R_0/2.452$.

With this trial inertial function the half-lives for the nuclei in the chosen test group were calculated, varying k to obtain the best overall fit. The least mean logarithmic deviation, Δ, of the calculated half-lives from the experimental values was obtained for k = 6.5 (cf. fig. 4). For this value of k, Δ is 1.4 corresponding to a factor of 25. In fig. 6 we show these calculated half-lives together with the experimental results as function of N and in fig. 7 as function of Z. An alternative way of representing the results is provided by fig. 8 in which the individual deviations from the experimental values are plotted.

From these figures it is seen that the overall trend of the half-lives throughout the entire test region is well reproduced. However, some systematic deviations exist, as we shall now comment on. The most severe deviations occur for the U isotopes. Here the isotopic variation as well as the absolute magnitude of the results are wrong. (The same tendency is present for Pu, but to a much less extent.) In part, this discrepancy may be due to fact ignored in this treatment that the U barriers are really three-peaked (the second barrier contains a shallow ternary minimum for several of the Th and U isotopes[6]). In part it may be due to the larger sensitivity of the half-lives for these nuclei to the correct description of the exit region. Hence, it seems difficult to improve on this point until the nuclear models have been made reliable for larger distortions. In the center region of the test group, the half-lives are reproduced best; in particular we observe that the N = 152 kink comes out qualitatively correct. However, it is too smooth as compared with the experimental one. This is probably due to some systematic deficiencies in the underlying single-particle levels. One should

relate this to the fact that also the theoretical ground-state masses exhibit a less pronounced kink than was experimentally observed. Furthermore, the decline of the second barrier when going away from N = 152 proceeds too slowly. In particular this has the consequence that the large drop in half-life, experimentally observed for ^{258}Fm and explained as the decline below the ground-state energy[12]), does not show up in the calculations until for ^{260}Fm.

A special problem is constituted by the shape-isomeric nuclei. This special group of nuclei was not included in the sample employed when fitting the inertial-mass function. As is seen from fig. 9, the obtained semi-empirical inertial function yields isomeric half-lives being too long by six orders of magnitude on the average. However, the ε_4 degree of freedom is expected to have a relatively large influence on the isomeric fission. In fig. 9 we have displayed the isomeric half-lives when the ε_4 dependence of the r coordinate is taken into account. It is seen that indeed this brings the calculated values into much better agreement with experiment. It should be added that a consistent inclusion of this effect does not appreciably change the good overall fit to the ground-state half-lives. We shall, therefore, continue to disregard the influence from this and other degrees of freedom in the evaluation of the penetrability integral.

To allow for the possibility of the inertia having a slope different from that of the irrotational inertia we have tried also to vary the parameter d in (8). Figure 10 shows the contour plot of Δ corresponding to this two-parameter variation. We see that there is a slight preference for a less steep variation with r of the inertia, but the net gain in Δ is little

($\simeq$ 0.1). Actually the best two-parameter fits are obtained for k = 4.0, d = 0.6, and for comparison we show in fig. 8 the corresponding scatter plot of the half-lives. Thus, this analysis reveals an ambiguity as to where in the valley displayed in fig. 10 the inertia should be positioned. This is important to keep in mind when applying the inertia to heavier elements for which the barrier region is restricted to small deformations and the half-lives thus rather sensitive to the actual choice of k. However, the shortness of the isomeric half-lives points to a preference of the upper-right part of the valley, hence we shall assume the previously discussed best one-parameter inertia to represent rather well the effective inertia.

Thus, it seems as if the present level of accuracy cannot be improved within this simple approach. Rather, more precise calculations will require more accurate energy surfaces together with a more detailed knowledge about and treatment of the associated nuclear inertial tensor - including shell effect in the latter. Finally, possibly also the effect of the nuclear viscosity may have to be taken into account.

An estimate of the uncertainty associated with the inertial-mass function may be obtained as follows. One may for each individual nucleus in the test group determine that value of the free parameter k which reproduces exactly the experimental half-life for that particular nucleus. This procedure yields a group of k values scattered around the mean value. The shaded region in fig. 5 is the ensuing inertial band corresponding to the width in the mentioned k distribution and thus indicates the uncertainty associated with the obtained semi-empirical inertial-mass function. The uncertainty defined in this way is found to be around 20%.

Since the main trends of the calculated half-lives throughout the test region agree with experiment, the method seems rather reliable for extrapolations to neighbouring, so far unobserved nuclei. Such applications are in progress and will be reported elsewhere, see for example ref. [12]). Here we shall as an example shortly mention the results for the element 106, ref. [12]). In fig. 6 we have displayed the calculated half-lives for some even isotopes of this element. It is seen that the isotope with N = 152 is predicted to be the most stable one, having a half-life of around 100 μsec (which is only a factor of three shorter than that of ^{258}Fm). The strong isotopic variation of the half-lives for the lighter elements was mainly due to the gradual disappearance of the second barrier. For the displayed even isotopes of the element 106, there is no second barrier at all and consequently the isotopic variation is expected to be very much smaller, as it also appears in the figure. Thus, we may predict that there should be one or several even isotopes of this element with spontaneous-fission half-lives not much shorter than that of ^{258}Fm. Investigations[12]) of the hindrance of element 106 associated with an odd particle suggest that some odd isotopes might exist with a half-life larger than that by one or a few orders of magnitude.

4. Conclusion

We have introduced a simple scheme for obtaining realistic one-dimensional fission barrier potentials and calculating the corresponding spontaneous-fission half-lives. On the basis of indications from hydrodynamical and microscopic calculations it has proved possible to establish a fission inertial-mass function which reproduces the transuranium fission half-lives (for even nuclei) to within a factor of 25 on the average, although the deviations for the light isomers measure is considerably larger. The overall trends of the calculated half-lives agree with experiment and furthermore some part of the deviations from experiment may be associated with certain deficiencies in the underlying potential-energy surfaces, while some part reflects neglected shell structure contributions to the inertial mass. The described scheme for calculating fission half-lives seems the most reliable one available at present for extrapolation to neighbouring regions and applications have been made to various problems of current interest, as will be reported elsewhere. For example, it is predicted that certain even isotopes of the element 106 have spontaneous-fission half-lives of the order of 100 μsec, while some odd cases could be one or a few powers of ten longer.

Acknowledgments

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Figure Captions

Fig. 1. Experimental spontaneous-fission half-lives as taken from ref. [10]).

Fig. 2. Illustration of the reduction from the multi-dimensional fission problem to a one-dimensional barrier penetration.

Fig. 3. Illustration of the method for obtaining the fission barrier potential from the five characteristic points.

Fig. 4. Comparison of various inertial-mass functions B_r (here shown for some Fm isotopes). The lower curve represents the irrotational-flow calculation[22]), while the three kinked upper curves are the cranking-model results obtained by Krumlinde[18]). The smooth curve in between is the determined best one-parameter semi-empirical inertial-mass function; the shaded region indicates the associated uncertainty as estimated from the spread in theoretical values around the experimental ones. For comparison the corresponding ε values are shown on the upper scale.

Fig. 5. Comparison of various inertial-mass functions B_r (here shown for ^{254}Fm). The lower curve represents the irrotational-flow calculation[22]), while the kinked upper curves correspond to various microscopic models:

Upper dashed: Cranking model, G = constant[23])

Lower dashed: Quasi-self-consistent model, G = constant[29])

Dot-dashed: Quasi-self-consistent model, $G \sim S$ [29])

The smooth curve in between is the determined best one-parameter semi-empirical inertial-mass function (corresponds to k = 6.5 in eq. (5)).

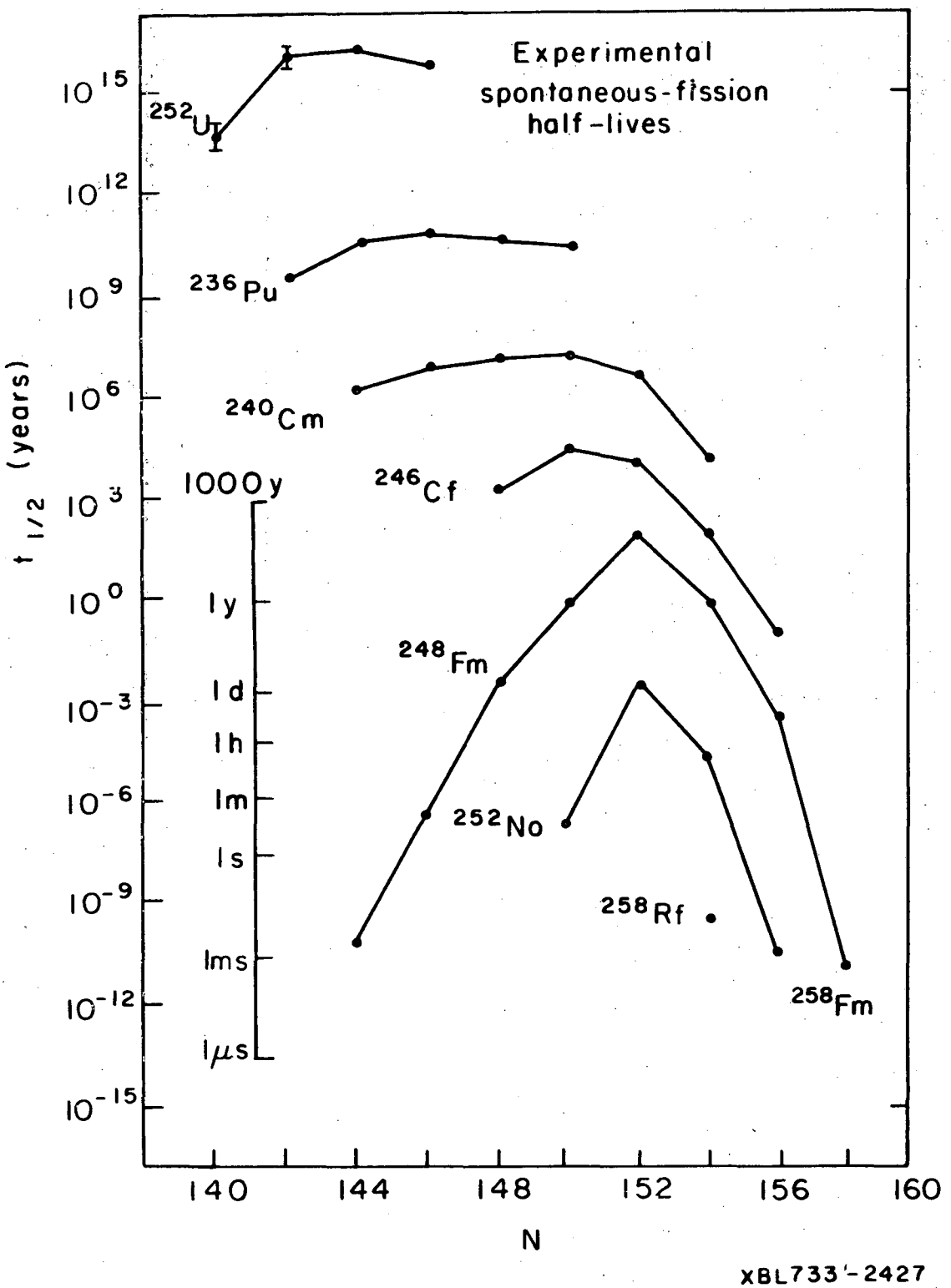
Fig. 6. Spontaneous-fission half-lives. Full circles: experimental values[9]). Open circles: calculated values with the determined semi-empirical inertia shown in fig. 4. The mean logarithmic deviation is 1.4. Also half-lives predicted for the element 106 are shown.

Fig. 7. Experimental and calculated spontaneous fission half-lives as a function of proton number Z for given values of neutron number N.

Fig. 8. Plot of the logarithmic deviations of the calculated half-lives from the experimental values. Full dots represent the best one-parameter inertial function, open dots the best two-parameter function (when also the slope is adjusted).

Fig. 9. Deviations of calculated half-lives from experimental values. In addition to the normal half-lives (full circles) also the results for some isomeric states are shown (open circles). The broken lines connect results obtained by including the effect of ε_4 on the r-coordinate, while all other points are calculated without this refinement.

Fig. 10. Contour map of the mean logarithmic deviation of calculated half-lives from experimental values, corresponding to the variation of as well magnitude k as slope c.


Experimental
spontaneous-fission
half-lives
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10^{12}
10^{9}
10^{6}
10^{3}
10^{0}
10^{-3}
10^{-6}
10^{-9}
10^{-12}
10^{-15}
1000y
1y
1d
1h
1m
1s
1ms
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XBL733-2427

Fig. 1

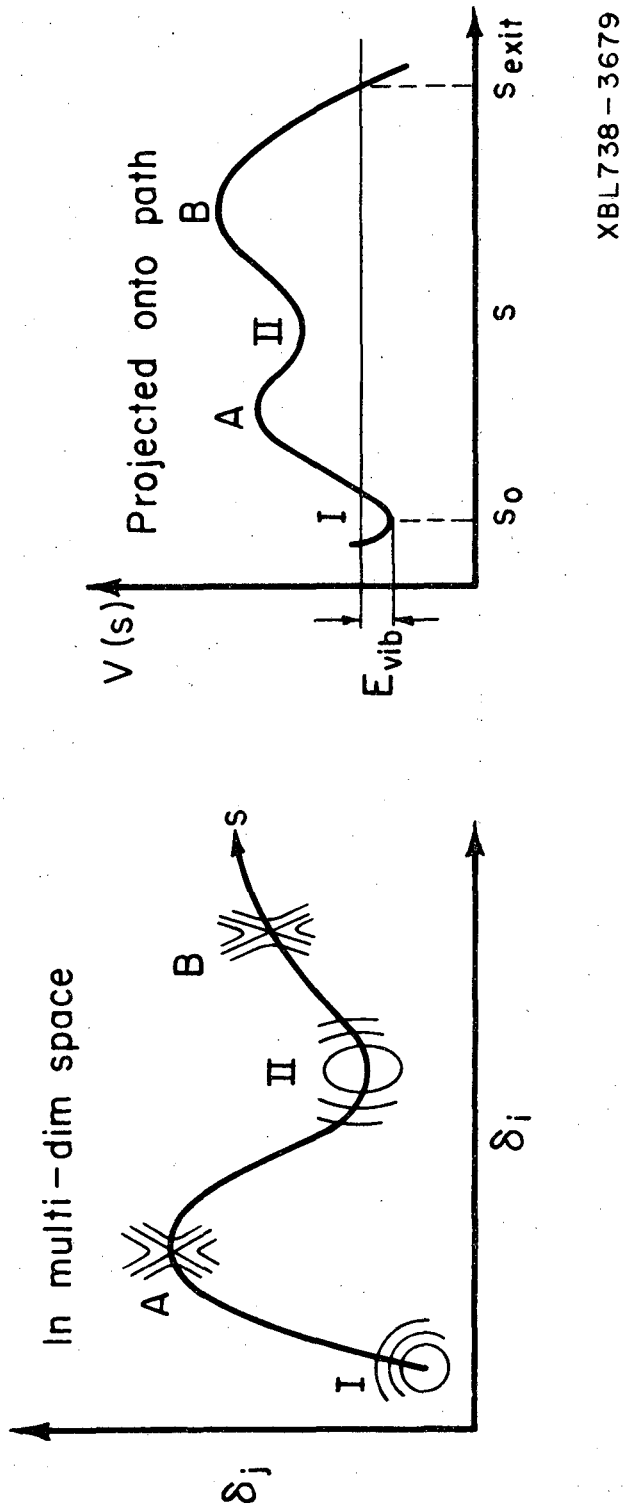

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Fig. 2

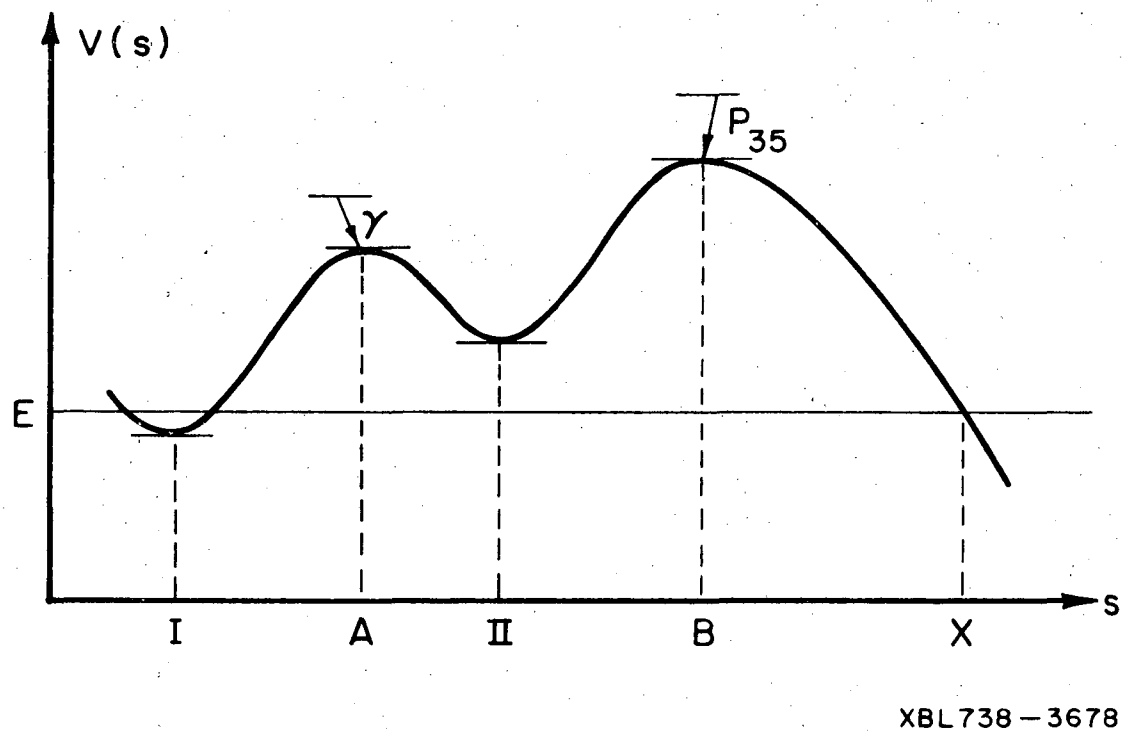

Fig. 3

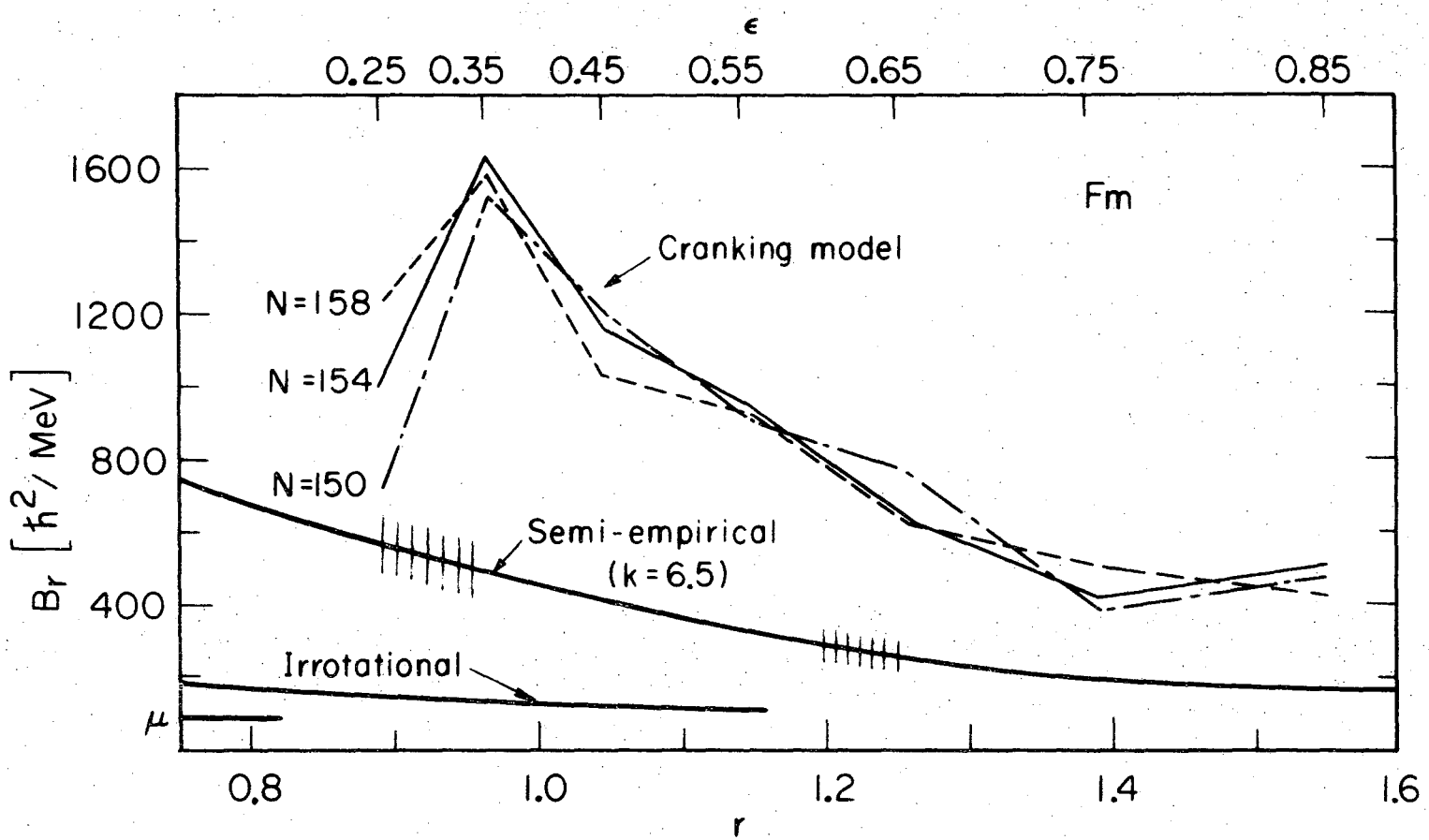

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Fig. 4

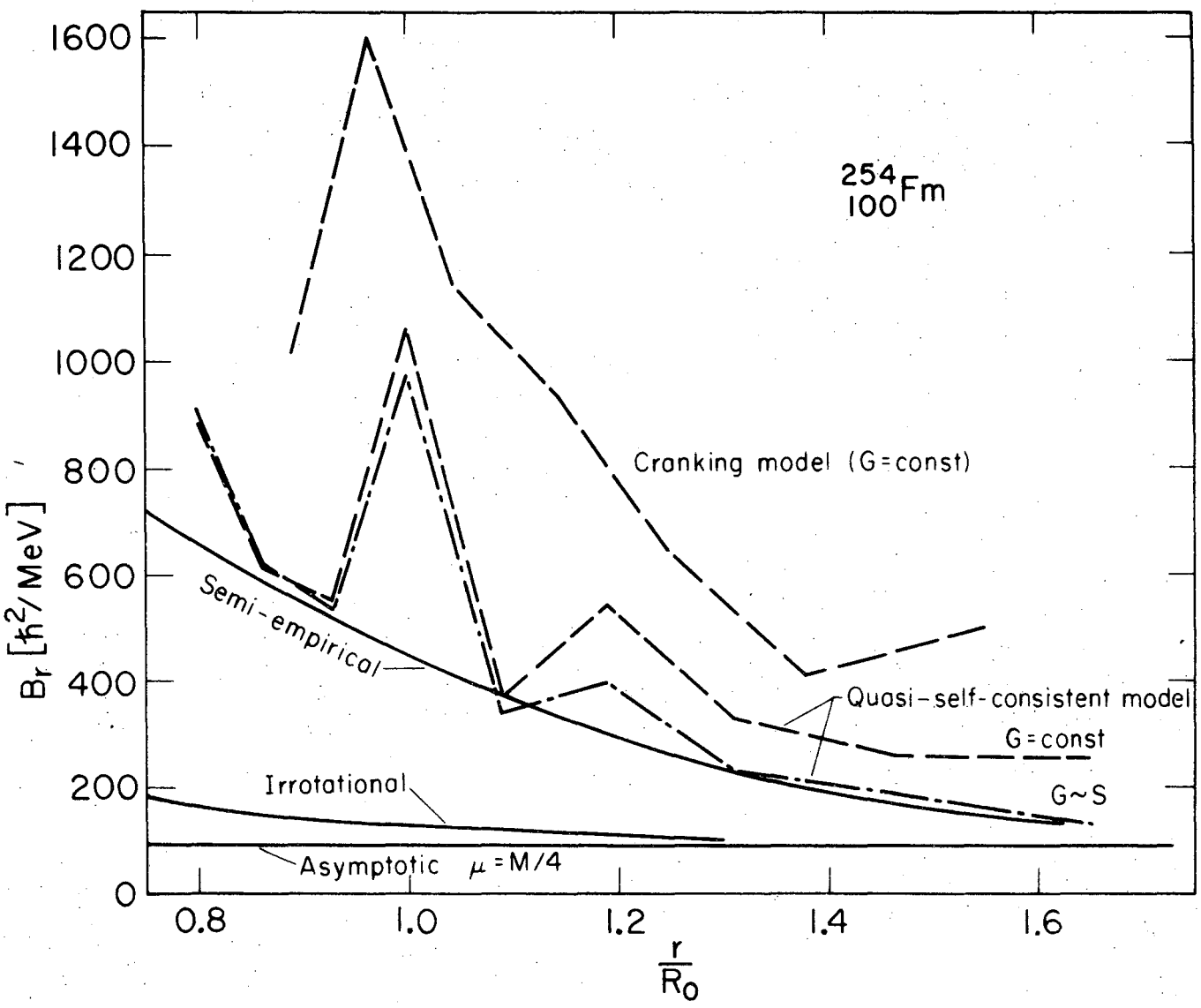

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Fig. 5

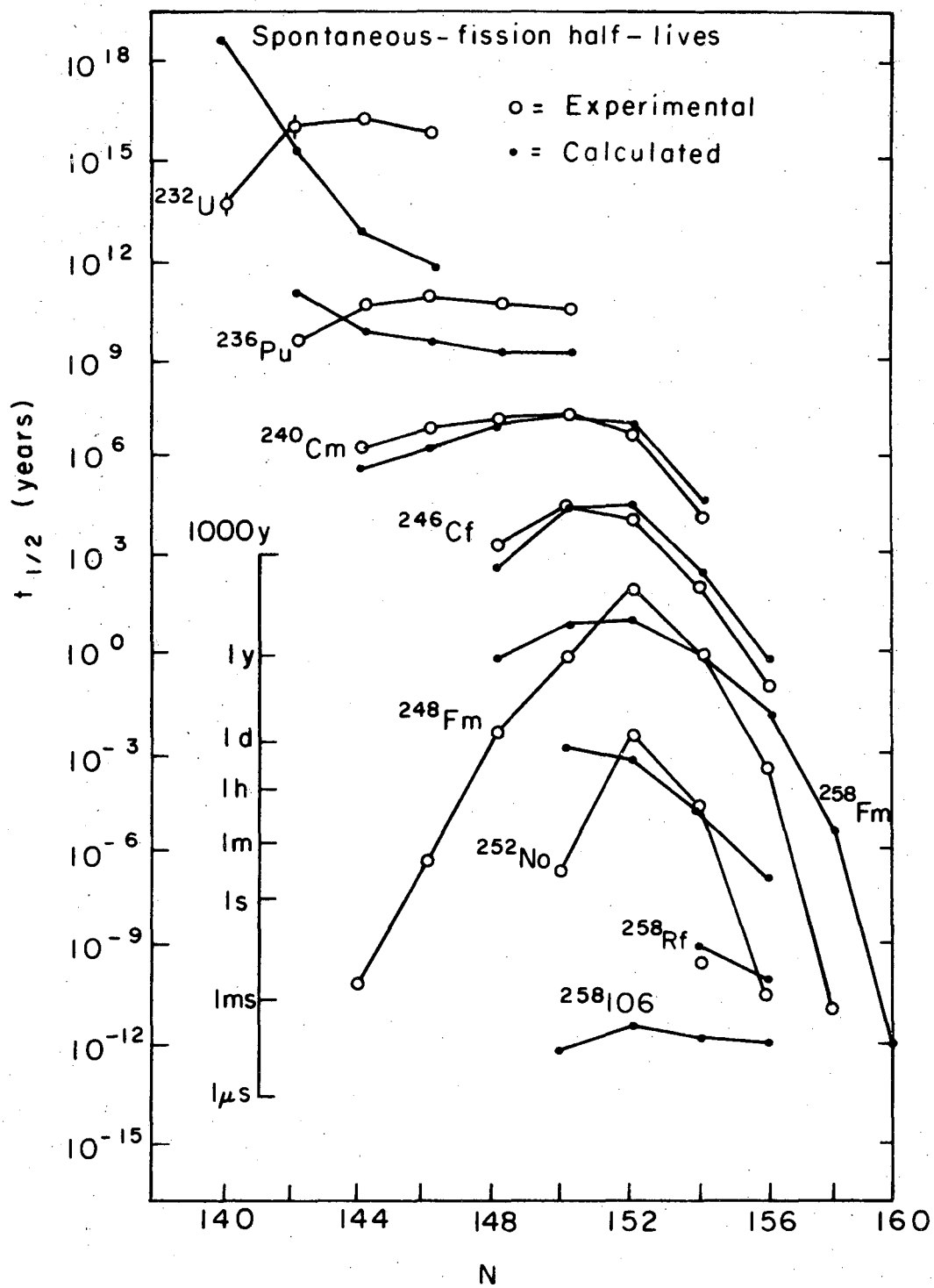

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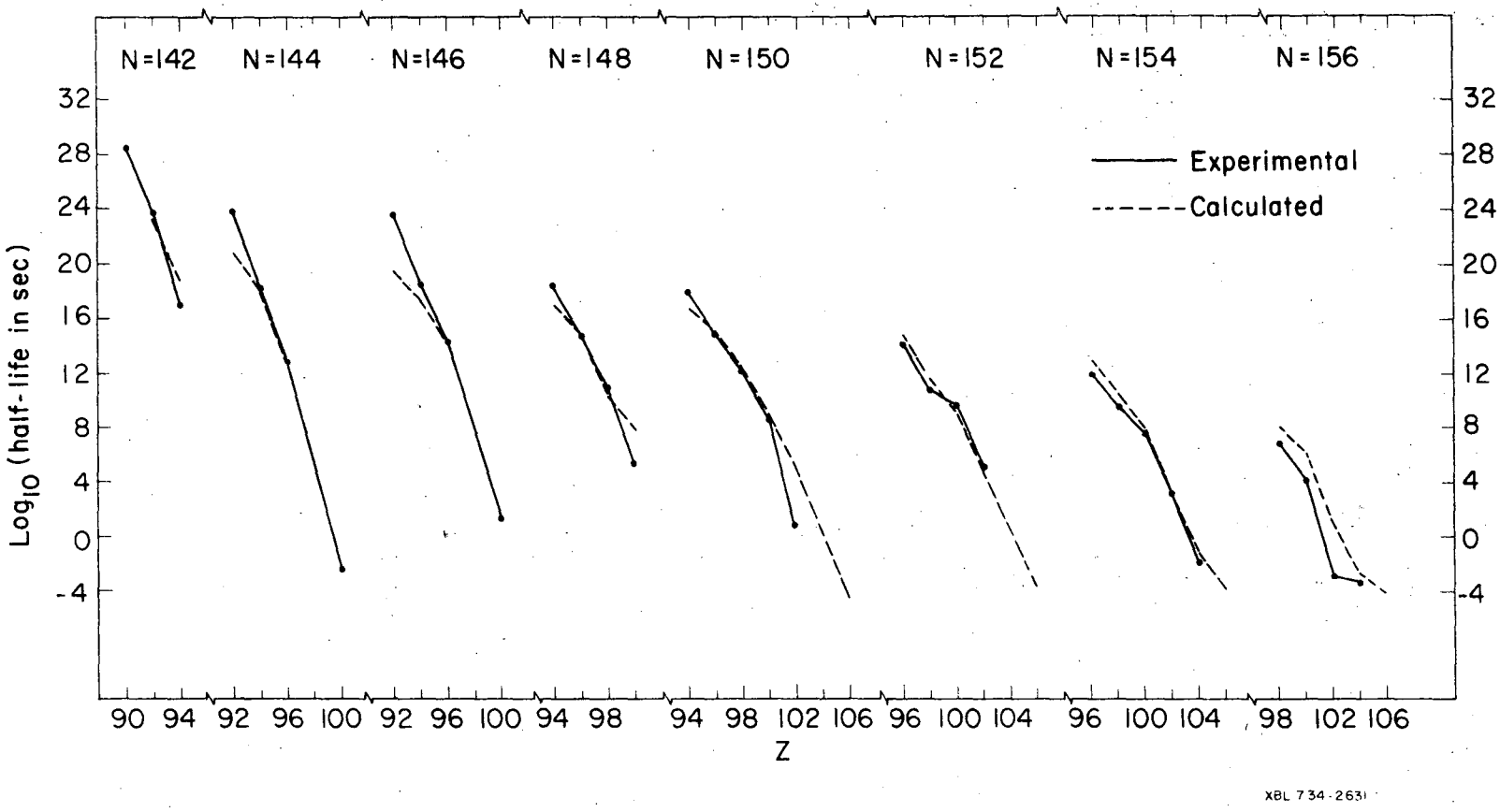
Fig. 6


N=142
N=144
N=146
N=148
N=150
N=152
N=154
N=156
Experimental
Calculated
Log10 (half-life in sec)
32
28
24
20
16
12
8
4
0
-4
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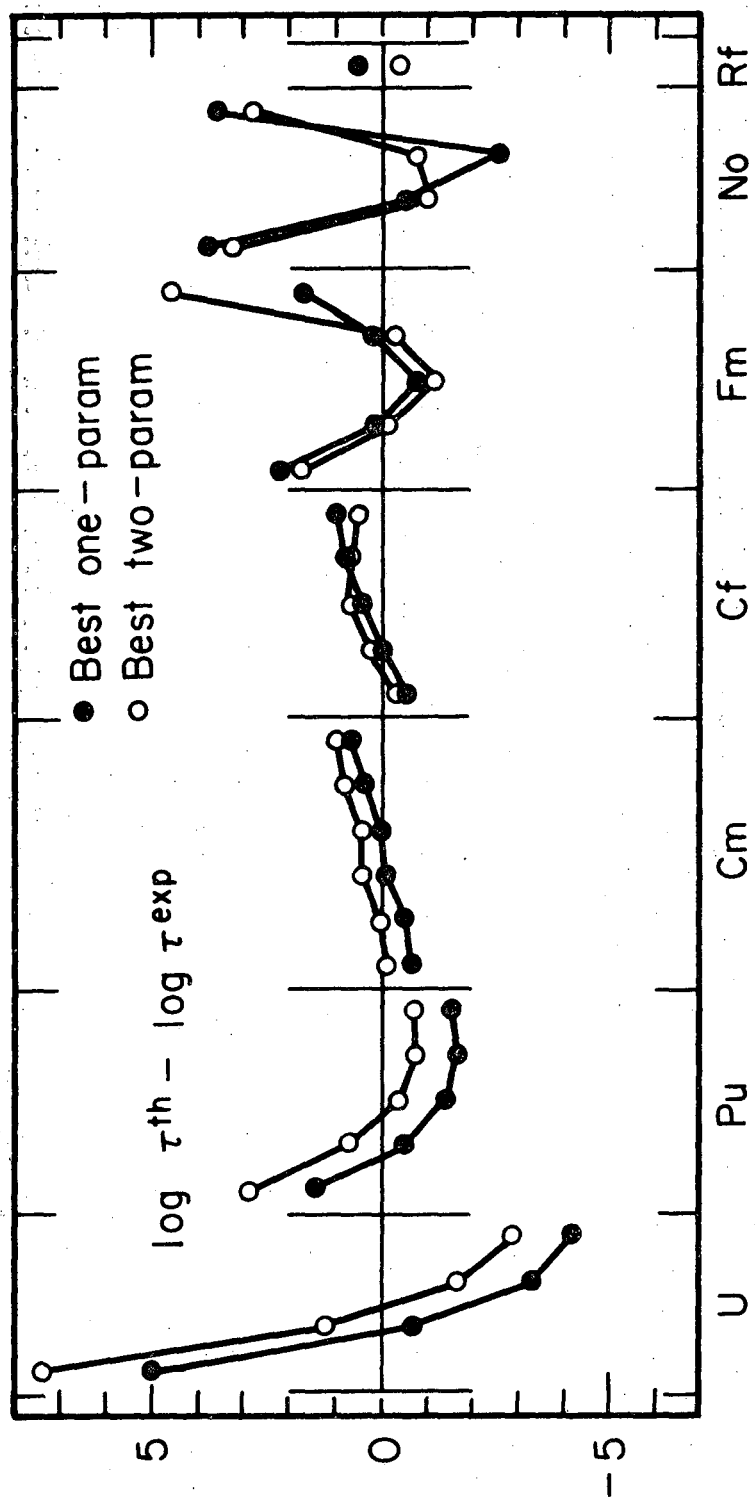
Z
XBL 734-2631

Fig. 7



XBL 738-3680

Fig. 8

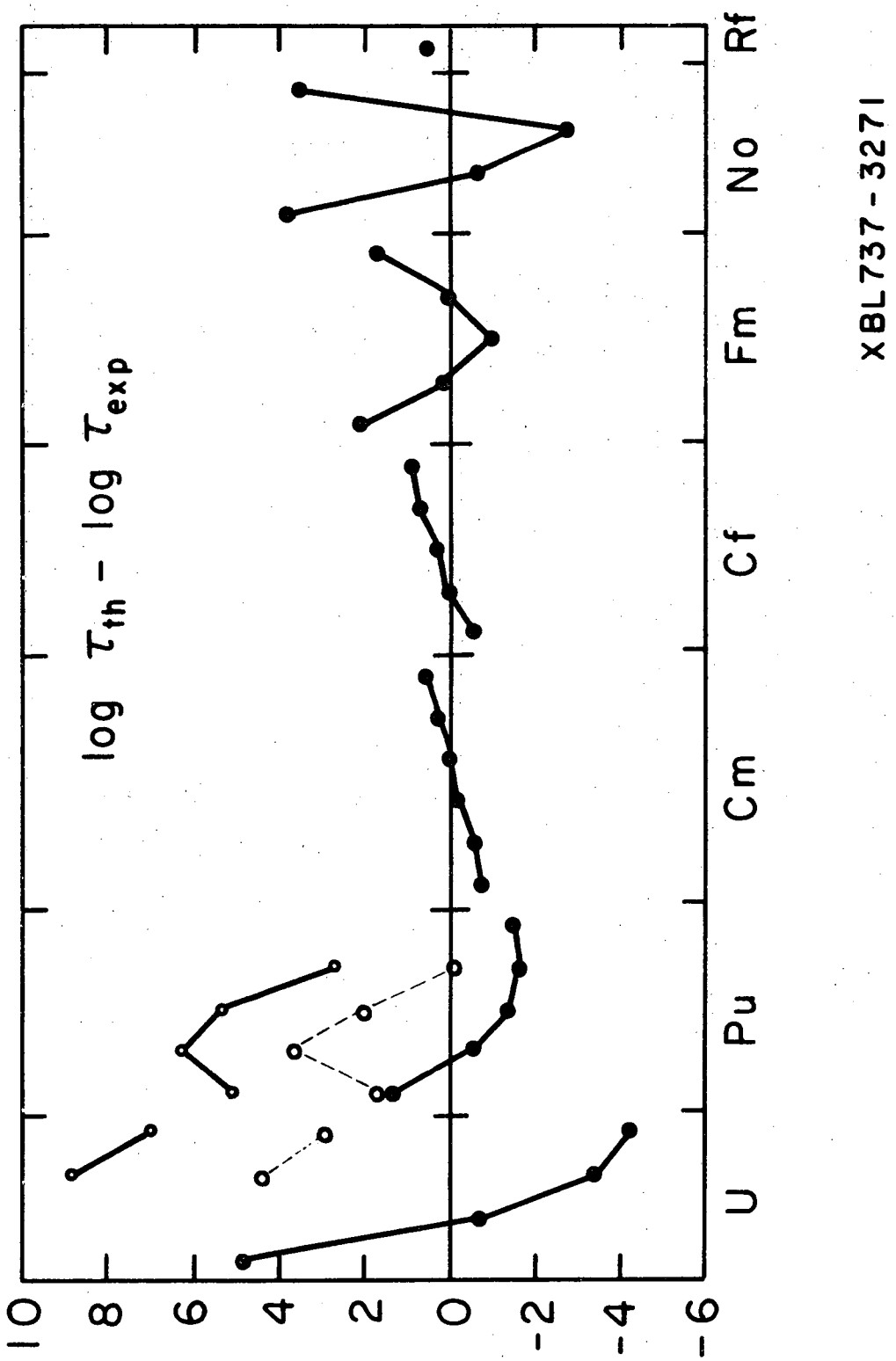
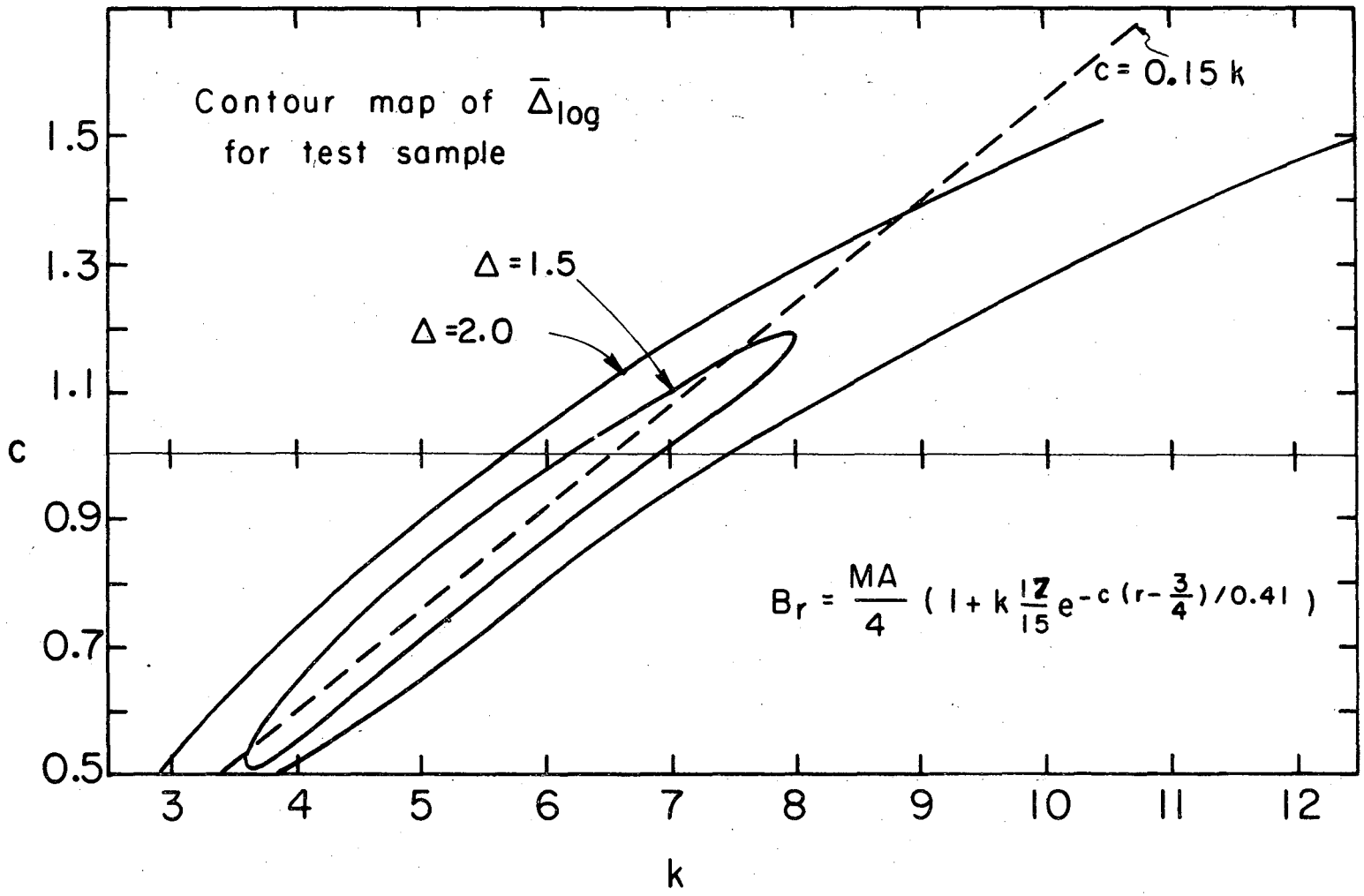

log τ_{th} - log τ_{exp}
10
8
6
4
2
0
-2
-4
-6
U
Pu
Cm
Cf
Fm
No
Rf
XBL737-3271

Fig. 9



XBL733-2426

Fig. 10

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