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Systematics of Fission Based on the Exponential Mass Formula

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Abstract

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A simple function of the fissionability parameter Z^2/A is found which enables fission thresholds to be calculated relative to the exponential mass formula reference surface (with shell and pairing corrections omitted). From these thresholds, systematics of spontaneous fission rates and of the ratios of neutron to fission widths are developed. A discussion is given of multiple neutron capture experiments in the light of these fission characteristics. author

Introduction

The recent renewal of studies of heavy isotopes produced by multiple neutron capture in underground thermonuclear explosions has raised a variety of questions concerning the properties of the nuclei formed. I had interpreted the yields from the original Mike explosion as indicating that conventional atomic mass formulas gave incorrect predictions of neutron binding energies far from the valley of beta stability (Cameron 1957). This led to the development of a new form of mass formula, the "exponential" mass formula (Cameron and Elkin 1965), in which neutron binding energies are larger in the neutron-rich region than given by conventional mass formulas. The fission properties of such nuclei are also of interest, and hence in this paper the systematics of several fission properties are developed, based on the exponential mass formula.

It has been recognized for many years that there is a correlation between spontaneous fission half-lives and the departures of actual masses of heavy nuclei from a smooth mass surface owing to shell effects (Seaborg 1952; Swiatecki 1955; Foreman and Seaborg 1958; Johansson 1959; Dorn 1961). The present approach preserves much of the basic spirit of the above papers, but the chain of reasoning presented here is somewhat different, and the resulting relationships tend to be simpler.

Fission Thresholds

Since fission is an exoergic process, strictly speaking there is no energy threshold. However, there is a fission Coulomb barrier whose penetration probability is very small except very near the top. Hence the top of the Coulomb barrier may be defined as an effective threshold (Cameron 1956). Most bombardment processes leading to fission result in a cross section plateau some distance above the top of the barrier. There are good practical and theoretical reasons for taking the top of the barrier to be the energy at which the cross section reaches half of the plateau value; the determination of this energy is somewhat subjective, but different people would probably choose an energy within a range of about 200 kev. I have adopted the energy thresholds given by Hyde (1964), since spot checks indicated that they had been chosen consistent with this determination. In addition, from (d,p) measurements of Northrup, Stokes, and Boyer (1959), I have taken the fission thresholds for the target nuclei U^{233}, U^{235}, and Pu^{239} to correspond to neutron bombarding energies of - 1.0, - 0.6, and - 1.0 Mev.

Swiatecki (1955) showed that spontaneous fission half-lives correlated well with the fissionability parameter Z^2/A if one assumed that the fission thresholds for even-even, odd-A, and odd-odd nuclei fell on different "mass" surfaces. Apart from shell effects, the ground state masses of these classes of nuclei also fall on three separate and smooth mass surfaces. Thus it appeared to me that it would be useful to see if the separations of the fission threshold mass surfaces should also correspond to nucleon pairing energies P(Z,N) which were determined in the development of the exponential mass formula (Cameron and Elkin 1965).

For the purpose of the present paper we may write the essence of the exponential mass formula as

$$(M-A)_{form} = (M-A)_{ref} + S(Z,N) + P(Z,N) \qquad (1)$$

where S(Z,N) is the shell correction, P(Z,N) is the pairing correction, $(M - A)_{ref}$ is the smoothly-varying mass excess which contains the principal terms of the mass formula, and $(M - A)_{form}$ is the mass excess predicted for a nucleus. These quantities are further defined in Cameron and Elkin (1965) and are tabulated in an Institute for Space Studies report.

Let us define the height of a fission threshold mass surface above the reference surface as

$$\Delta\ (M - A)_f = (M - A)_{exp} + E_f - (M - A)_{ref} \qquad (2)$$

where $(M - A)_{exp}$ is the experimental mass excess of a nucleus and E_f is the observed fission threshold. Let us further form the quantity

$$\epsilon = \Delta\ (M - A)_f - P\ (Z,N) \qquad (3)$$

where we recall that the pairing energy P (Z,N) is intrinsically negative. If the hypothesis, that the energy differences between the fission threshold mass surfaces are the same as the pairing energy differences between ground state mass surfaces, is correct, then ϵ should define a universal fission threshold mass surface coincident with that for odd-odd nuclei.

A plot of ϵ versus the fissionability parameter Z^2/A is shown in Figure 1. It may be seen that an excellent straight-line empirical relationship is obtained. There is no indication of any systematic deviation of the points corresponding to one type of nuclear species or another. Thus it is clear that the above hypothesis is satisfied sufficiently closely to form the basis of an empirical method for calculating fission thresholds. The straight line of Figure 1 is described by the relation

$$\epsilon = 57.35 - \frac{4.3Z^2}{3A} \qquad (4)$$

If an experimental mass excess is known, then fission thresholds can be calculated from

$$E_f = (M - A)_{ref} + \epsilon + P\ (Z,N) - (M - A)_{exp} \qquad (5)$$

However, if the experimental mass excess is not known, then $(M - A)_{form}$ may be substituted for $(M - A)_{exp}$ in equation (5), which then reduces to

$$E_f = \epsilon - S\ (Z,N) \qquad (6)$$

Equation (6) demonstrates that fission thresholds do not exhibit an odd-even fluctuation due to pairing energy effects.

Spontaneous Fission Lifetimes

It is evident that the empirical relationship based on the data shown in Figure 1 does not extend over a very large range of Z^2/A. One way to test the validity of extrapolations of this relationship to other values of Z^2/A is to see if a consistent systematics of spontaneous fission half-lives can be based upon it.

Swiatecki (1955) noted that every millimass unit of extra ground state depression relative to the fission threshold resulted in about a factor 10^5 increase in spontaneous fission lifetime. Earlier, Frankel and Metropolis (1947) had suggested that spontaneous fission half-lives were of the form

$t_{\frac{1}{2}} = 10^{-21} \times 10^{7.85 E_f}$ seconds, where $10^{-7.85 E_f}$ is the barrier penetrability, and the nucleus is assumed to fission in 10^{-21} seconds when above the barrier. Their coefficient for the barrier penetration was based on machine calculations and it should be considered only to give the order of magnitude.

property may be deduced from the fact that the points corresponding to measured values of both λ_f and E_f partake of it. Presumably this scattering is to be attributed to variations in the reduced fission width of the ground state. A greater scattering is observed for the cases in which E_f was calculated from equation (6); presumably this represents additional uncertainties in the fission threshold energy. However, there is no indication of a variation of the mean value of c with Z^2/A. Hence equation (7) may be considered to give a suitable prediction for λ_f, but nevertheless a prediction which will be uncertain by as much as a factor 10^5. There is some indication that there is less scatter in the calculated values of c for a given value of Z, thus suggesting that variations in fission reduced widths depend in part on the proton structure. Thus for predictive purposes equation (7) becomes

$$\lambda_f = 10^{14-6.6 E_f} \tag{7a}$$

Hence the study of spontaneous fission lifetimes gives a correlation which is consistent with the validity of equation (4) through the entire range of Z^2/A for which spontaneous fission half-lives have been measured. However, it is clear that such a simple relationship must be used with caution outside the region for which it has been tested; we have no

The picture of a barrier-free nucleus fissioning in 10^{-21} seconds is clearly not in accord with modern data. It is well known that when neutron-fissile isotopes are bombarded with resonance-region neutrons, the resulting fission cross sections are comparable to or slightly greater than capture cross sections. For such nuclei radiation widths are about 10^{-1} ev, corresponding to a lifetime of 10^{-14} sec. Therefore I have assumed that nuclei at the fission threshold will have a fission rate of 10^{14} sec^{-1}. Hence in general the spontaneous fission rate can be written in the form

$$\lambda_f = 10^{14-cE_f} \tag{7}$$

Here c is a constant coefficient which must be determined.

The coefficient c was calculated for those nuclei with measured spontaneous fission half-lives (Hyde 1964). Experimental values of the fission threshold E_f were used if they were available. If they were not available, E_f was calculated preferably from equation (5), but if necessary from equation (6). The resulting values of the lifetime exponent factor c are plotted as a function of Z^2/A in Figure 2.

It may be seen in Figure 2 that the derived values of c vary by about $\pm$ 1 or a little more on either side of a mean value of about 6.6. That this scatter is an intrinsic nuclear assurance that it would work well for two nuclei with the same value of Z^2/A but differing appreciably in mass number A.

Neutron to Fission Width Ratios

The competition between fission and neutron emission at energies significantly above the fission and neutron thresholds has been studied by Vandenbosch and Huizenga (1958), who found that the ratio of widths Γ_n/Γ_f is correlated with the difference of the fission and neutron emission thresholds, $E_f - E_n$, after some adjustments have been made for nuclear type. It seems appropriate to seek similar correlations with the present approach to determination of E_f.

Vandenbosch and Huizenga adjusted $E_f - E_n$ by the neutron pairing energy, P(N), and by an assumed energy gap in the effective levels of the fissioning nucleus at the saddle point. However, from energy level density considerations (Newton 1956) it seems more appropriate to raise the neutron binding energy by the pairing energy of the residual nucleus, P(Z,N - 1). Hence we form the quantity

$$E_f - E_n + P(Z,N - 1) + \Delta_f$$

Experimental values of Γ_n/Γ_f must be correlated with this quantity. The term Δ_f is related to the assumed fission energy gap at the saddle point.

The data chosen for this correlation were those given by Vandenbosch and Huizenga for 3-Mev neutron fission and for photofission, supplemented by a somewhat lower energy neutron bombardment of Pu^{242} (Butler 1960). Examination of this data suggested the following values for Δ_f:

$$\Delta_f = \begin{cases} 0, \text{ even-even nuclei} \\ -\ 0.57 \text{ Mev, odd-A nuclei} \\ -\ 0.88 \text{ Mev, odd-odd nuclei} \end{cases}$$

It must be emphasized that these values of Δ_f are not reliable to more than one significant figure.

The resulting correlation is shown in Figure 3. It may be seen that the majority of the points are consistent with a straight-line relationship which will have some value for predictive purposes. This relationship is represented by

$$\frac{\Gamma_n}{\Gamma_f} = 8.66 \exp 1.42 \left[E_f - E_n + P(Z,N - 1) + \Delta_f\right] \qquad (8)$$

It may be seen that three points are displaced parallel to the correlation line of Figure 3 by approximately a factor 3. These points all correspond to isotopes of thorium, which apparently have intrinsically smaller fission widths. It is interesting to note that thorium isotopes also have unusually long spontaneous fission lifetimes.

Multiple Neutron Capture Experiments

In Table 1 are given values of E_f and E_n for a wide range of neutron-rich nuclei which may be of interest in multiple neutron capture experiments. All values have been calculated from the exponential mass formula. Several interesting conclusions follow from an examination of these numbers.

The yields formed by multiple neutron capture on a U^{238} target have now been published for several different experiments. These yields are characterized by a progressively diminishing abundance with mass number upon which is superposed an odd-even variation with successive mass numbers. This odd-even superposition changes sign beyond about $A = 250$.

Dorn and Hoff (1965) have interpreted this reversal of the odd-even effect as resulting from spontaneous fission of the heavy isotopes competing with beta decay. According to the relations found in this paper, this would require that for such nuclei $E_f < 3$ Mev. This requirement is in conflict with the numbers derived for E_f in Table 1.

However, this interpretation of Dorn and Hoff is not accepted by Bell (1965) or by some of his colleagues. Bell analyzes a simplified model of a contemporary experiment as follows. He assumes that a deuterium-tritium mixture has intimately intermixed with it a U^{238} target. There is an initial irradiation of the target with 14 - Mev neutrons, which will induce not only (n, 2n), (n, 3n), and (n, f) reactions, but also some (n, p) and (d, n) reactions, the latter resulting from knocked-on deuterons. The latter two types of reactions produce small amounts of Pa and Np target nuclei. Multiple capture of relatively low energy neutrons follows. Because of the higher capture cross sections of the Pa and Np nuclei, the yields at the highest mass numbers result from neutron capture in odd-Z nuclei, thus producing the reversal of the odd-even effect.

One of the principal objectives of the multiple neutron capture experiments is to produce the greatest possible yields of the highest possible mass numbers. One apparently attractive way to do this is to irradiate targets with the largest possible values of Z and A. But Table 1 indicates that caution is necessary. In order to minimize competition of slow neutron fission with neutron capture, it is necessary to choose the most massive reasonable isotope of a given element. Thus fission competition with capture is probably not prohibitive beyond a Pu^{242} target, but it would be serious beyond any practical curium or californium targets.

Fast neutron fission of the initial target nucleus can also become very serious; in fact it would already be serious in a Pu^{242} target. When bombarded by 14 - Mev neutrons, a heavy nucleus will have two or three chances at fission in competition with neutron emission; the Γ_n/Γ_f ratio for neutron bombardment of Pu^{242} is smaller than for U^{238}.

Moreover, after losing typically two neutrons, forming Pu^{240}, the subsequent slow neutron induced fission in Pu^{241} and Pu^{243} will lead to much more target depletion than will the corresponding neutron loss to U^{238}. Thus with plutonium or heavier targets it must be expected that the yield of heavy elements per unit mass of target will be considerably reduced, and the increased capture cross section due to fission products will require that the total mass of the target should also be reduced in order to maintain a large integrated neutron flux available for capture in the target material.

The fission parameters exhibited in Table 1 are also of interest for another type of multiple neutron capture: that process in nucleosynthesis in which neutrons are captured rapidly. The capture paths lie well on the neutron-rich side of the valley of beta stability. From Table 1 it may be predicted that such rapid capture will not be terminated by fission until far beyond the range of tested validity of either the exponential mass formula or the fission systematics of this paper.

TABLE 1. Fission Thresholds and Neutron Binding Energies (Mev.)

Mass	Pa		U		Np		Pu		Am		Cm		Bk		Cf	
Number	E_f	E_n	E_f	E_n	E_f	E_n	E_f	E_n	E_f	E_n	E_f	E_n	E_f	E_n	E_f	E_n
238	6.82	4.87	6.29	6.03	5.76	5.56	5.10	6.87								
239	6.90	5.74	6.36	5.05	5.85	6.22	5.33	5.74								
240	7.13	4.77	6.45	5.92	5.90	5.23	5.41	6.40								
241	7.40	5.61	6.74	4.94	6.00	6.09	5.48	5.41								
242	7.56	4.61	6.95	5.78	6.23	5.12	5.58	6.28	5.01	5.60						
243	7.69	5.79	7.11	4.78	6.51	5.95	5.82	5.30	5.13	6.46						
244	7.33	3.41	7.24	5.96	6.67	4.95	6.09	6.13	5.36	5.48						
245	7.17	5.28	6.89	3.57	6.81	6.13	6.25	5.13	5.64	6.31						
246	7.13	3.58	6.73	5.44	6.45	3.74	6.39	6.30	5.80	5.30	5.21	6.49	4.36	5.84		
247	7.17	5.12	6.59	3.74	6.30	5.61	6.05	3.91	5.95	6.47	5.38	5.48	4.65	6.67		
248	7.37	3.85	6.74	5.28	6.26	3.90	5.89	5.77	5.61	4.08	5.52	6.65	4.82	5.66		
249	7.56	5.10	6.94	4.00	6.31	5.44	5.85	4.07	5.46	5.91	5.18	4.25	4.97	6.83		
250	7.75	3.70	7.13	5.25	6.51	4.16	5.91	5.60	5.41	4.23	5.03	6.11	4.62	4.43	4.60	7.01
251	7.94	4.93	7.32	3.85	6.71	5.40	6.11	4.32	5.47	5.77	5.01	4.40	4.48	6.29	4.27	4.61
252	8.12	3.55	7.51	5.07	6.90	4.00	6.31	5.56	5.67	4.48	5.06	5.93	4.45	4.57	4.13	6.46
253	8.31	4.76	7.71	3.69	7.10	5.22	6.51	4.15	5.88	5.72	5.28	4.65	4.51	6.10	4.10	4.75
254	8.49	3.41	7.90	4.91	7.29	3.84	6.70	5.38	6.08	4.31	5.48	5.88	4.73	4.81	4.16	6.27
255	8.68	4.61	8.09	3.55	7.48	5.05	6.90	3.99	6.28	5.53	5.68	4.47	4.93	6.05	4.38	4.98
256	8.86	3.27	8.27	4.75	7.67	3.69	7.09	5.20	6.48	4.15	5.89	5.69	5.14	4.63	4.59	6.21
257	9.04	4.46	8.45	3.41	7.86	4.89	7.29	3.84	6.67	5.35	6.09	4.30	5.34	5.85	4.80	4.80
258	9.21	3.14	8.64	4.59	8.05	3.55	7.48	5.03	6.87	3.99	6.29	5.50	5.54	4.46	5.01	6.01
259	9.39	4.32	8.82	3.28	8.24	4.73	7.67	3.69	7.06	5.18	6.48	4.14	5.75	5.66	5.21	4.62
260	9.57	3.02	9.01	4.45	8.42	3.41	7.86	4.87	7.25	3.83	6.68	5.33	5.95	4.29	5.41	5.82
261															5.61	4.45
262															5.81	5.63

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Figure Captions

Figure 1. The quantity ϵ, defined by equation (3), plotted against the fissionability parameter Z^2/A. The straight line is given by equation (4).

Figure 2. The spontaneous fission lifetime exponent factor c a as calculated from equation (7) is plotted against the fissionability parameter Z^2/A.

Figure 3. The ratios of neutron and fission widths plotted against the adjusted threshold energy differences. The straight line is given by equation (8).