Multimodal fission and neutron evaporation

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The average multiplicities $\overline{v}(A)$ of prompt neutrons emitted in the spontaneous fission of ²⁵²Cf and ²⁵⁸Fm are derived. Two new features are predicted: A simple sawtooth for ²⁵⁸Fm and a triple one for ²⁵²Cf. Experiments to check these predictions should be feasible now.

The arguments to be given are based on two hypotheses: multimodal fission and scission at random positions on the neck. The term "multimodal fission" means that a nucleus may choose several paths to scission. Consequently a nucleus has several prescission shapes which are distinguished by total length and asymmetry. The decision on the actual path is taken at one of the bifurcation points which are reached much before scission takes place. Each of these prescission shapes is subject to fluctuations, both with respect to length and to asymmetry. Here we will be concerned only with the second kind, i.e., with "scission at random positions on the neck" or, in other words, with random neck rupture. Both hypotheses are not at all arbitrary. In the context of recent experimental results, multimodal fission is discussed by Itkis et al. for the preactinides¹ and by Hulet etal. for very heavy actinides.² Impressive evidence is due to Knitter and co-workers³ who showed that there are three modes in the neutron induced fission of ²³⁵U which are sizably repopulated when the kinetic energy of the incident neutron is changed by 2 eV. The discovery of a very asymmetric component in the spontaneous fission of ²⁵²Cf may also be considered as an indication of multimode fission.^{4,5} Furthermore much theoretical work confirms the existence of several fission paths, mostly for the heaviest actinides, $^{6-11}$ but also for the lighter ones. $^{6,12-14}$ Whetstone¹⁵ was probably the first to propose random neck rupture as a mechanism for nuclear fission. He introduced it to give a qualitative explanation of the sawtooth curve well known for the neutron multiplicities. Later on the same hypothesis was used by Karamyan, Oganesyan, and Pustylnik¹⁶ to understand the anomalously large variances of mass distributions measured in nuclear fission. These authors presented first quantitative comparisons. Still later, Knitter et al.¹⁷ obtained results for 235 U (n, f) in the same direction, but this time comparisons were much more detailed than in any previous work. A survey on publications concerned with random neck rupture including broad quantitative information can be found in Ref. 18.

The new idea is a synthesis of both hypotheses. Suppose there are three different prescision shapes: a symmetric very short one, a moderately asymmetric one of larger length, and a very asymmetric one which has about the same length as the second shape. These are the shapes shown in Figs. 1(a), 1(c), and 1(d); they are labeled "1", "2", and "3" in Table I. According to random neck



FIG. 1. The relations between prescission shapes, mass yields Y (dotted lines, right-hand-side scales), and neutron multiplicities $\bar{\nu}$ (solid lines, left-hand-side scales) as functions of mass number A of one of the fragments. Although the pictures were made for general illustration, they represent the fission components that should be relevant for ²⁵⁸Fm. In particular, (d) displays a very asymmetric fission component which, according to our calculations, should exist for ²⁵⁸Fm as it exists for ²⁵²Cf (Ref. 13). The lengths of the prescission shapes in Figs. 1(a)-1(c) can be found in Table I as l_1 , l_2 , and again l_2 , respectively. The mass yield in Fig. 1(c) consists of two asymmetric parts which are discriminated by the light dots.

TABLE I. Fission-mode probabilities p_c , scission lengths l_c , and asymmetries, expressed by the average mass numbers \overline{A}_c of the light fragments, as used for the preparation of Figs. 2 and 3. \overline{A}_{2a} and \overline{A}_{2b} refer to the variants displayed in Figs. 2(a) and 2(b). The "experimental" scission lengths were obtained from the measured average total kinetic energies using Eq. (14) of Ref. 20. Just for comparison, scission lengths and asymmetries taken from microscopic calculations are also presented. For these "theoretical" scission lengths the factor 1.15/1.2249 was applied to the numbers given in Refs. 8, 13, and 22 in order to account for different definitions of the nuclear radius constant.

Nucleus	Variable	Experimental	References	Theoretical	References
²⁵⁸ Fm	P ₁	0.56	2,19		
	p ₂	0.44	2,19		
	l_1/fm	28.0	19,20	27.0	8
	l_2/fm	33.5	19,20	34.9	8
	\overline{A}_1	129	2	129	8,22
	\overline{A}_{2a}	119	2	119	8,22
	\overline{A}_{2b}^{2a}	129	2		,
²⁵² Cf	p ₂	1	4,5		
	<i>p</i> ₃	10^{-5}	4,5		
	l_2/fm	35.4	5,20	34.4	13
	l_3/fm	35.1	5,20	29.5	13
	\overline{A}_{2}	109	5	112	13
	\overline{A}_{3}^{2}	65	4,5	68	13

rupture, a mass yield curve is a picture of the prescission shape because a short neck makes a narrow mass yield, whereas a long neck produces a broad yield [cf. Figs. 1(a) and 1(b)]. Furthermore, a symmetrical shape produces a symmetrical mass distribution, while an asymmetric shape gives an asymmetric yield [cf. Figs. 1(b), 1(c), and 1(d)]. Since the prescission shape may break at various positions, fragments with very different deformations are produced. This ends up with an average neutron multiplicity $\overline{\nu}(A)$ which increases monotonously with mass number A if the prescission shape is symmetric [cf. Fig. 1(a)].²⁰ A longer shape just lifts the neutron-multiplicity curve [see Fig. 1(b)] because it generates fragments with larger deformations. From an asymmetric prescission shape, however, a sawtooth-shaped multiplicity is obtained [Figs. 1(c) and 1(d)].¹⁸ Now, the multimodal hypothesis suggests that each of these prescission shapes is reached with some probability p_c

$$\sum_{c} p_{c} = 1 , \qquad (1)$$

where c is the number of the fission path (c = 1, 2, 3) in the present example). Each prescission shape generates a Gaussian-like mass yield $Y_c(A)$ with the customary normalization

$$\sum_{A=0}^{A_{cn}} Y_c(A) = 2 , \qquad (2)$$

where A_{cn} denotes the mass number of the compound nucleus, and a neutron multiplicity $\bar{\nu}_c(A)$. The measured mass yield and the neutron multiplicity should be then just superpositions

$$Y(A) = \sum_{c} p_{c} Y_{c}(A) , \qquad (3)$$

$$\overline{\nu}(A) = \sum_{c} p_{c} Y_{c}(A) \overline{\nu}_{c}(A) / \sum_{c} p_{c} Y_{c}(A) .$$
(4)

Some results for $\overline{\nu}(A)$ are displayed in Figs. 2 and 3.

The numbers used for the construction of Figs. 2 and 3 are listed in Table I. I proceeded exactly as described in Ref. 18, i.e., to determine these numbers from a fit to known experimental data: If all $Y_c(A)$ are represented by normalized Gaussians, the p_c 's give their relative



FIG. 2. Predicted neutron multiplicities \overline{v} in the spontaneous fission of ²⁵⁸Fm as functions of mass number A. (a) is a superposition of the components shown in Figs. 1(a) and 1(c) according to Eq. (4), (b) comes from Figs. 1(a) and 1(b). The dashed lines display what is thought to be the best indicator of an asymmetric fission mode (see text). The contribution of the super-asymmetric fission mode [cf. Fig. 1(d) and Fig. 3] is not shown because its experimental verification would probably take more than 10^8 counts.



FIG. 3. Predicted neutron multiplicities \overline{v} in the spontaneous fission of ²⁵²Cf as functions of mass number A. This figure results from a superposition of components similar to those shown in Figs. 1(c) and 1(d). The two branches of $\overline{v}(A)$ according to Fig. 1(c) are not seen because only the sum of the contributions from the light and the heavy fragments can be observed. I describe this fact by a straightforward extension of Eq. (4). The superlong fission mode, i.e., one that leads to a very long, nearly symmetric prescission shape (Ref. 13) does not essentially change this prediction because there is no range of mass numbers where this mode dwarfs the others.

heights, the average mass numbers \overline{A}_c of the light fragments are the centroids, and the lengths l_c are fixed by the average kinetic energies of the corresponding fission modes. However, most of the numbers needed for construction can now also be found from purely theoretical arguments, ¹³ and it is satisfactory to see that they come out in close agreement except for l_3 of ²⁵²Cf (see Table I).

One may doubt the applicability of random neck rupture if the neck is so short as, e.g., in Fig. 1(a). To answer the question, we performed dynamic liquid drop calculations²¹ and found that the variances in the mass distribution are prepared by an instability which shifts the location of the smallest diameter on the droplet either to the right or to the left. This instability is most effective when the nuclear shape changes from an oval to a necked-in configuration. In the actinides, the change happens close to the saddle, i.e., for a shape shorter than any of the shapes displayed in Fig. 1. The physical reason for this instability (not to be confused with the Rayleigh instability) is easily comprehensible: On the transition from oval to necked-in configurations the droplet becomes so flat that it takes actually zero energy to effect a large shift of that position where the nucleus will part into pieces. Furthermore, by dynamic calculations we found out that a shorter prescission shape limits the evolution of this instability so that a shorter prescission shape must result in a narrower distribution of mass. But this is contained in the algorithms explained in Ref. 18.

In order to test the reliability of the predictions, I have altered the prescriptions of Ref. 18 in a more or less arbitrary manner. It turned out that the neutron multiplicities depend only on the length and the asymmetry of the prescission shape. Further studies were done to establish that no reasonable change in the mode probabilities p_c or in the excitation energy of the prescission nucleus would essentially alter Figs. 2 or 3.

TABLE II. Predicted total neutron multiplicities \overline{v}_{tot} for various fission modes. The errors in these numbers should be smaller than ± 1 neutron. The fission modes are labeled in the same way as in Table I.

Nucleus	Mode	$\overline{v}_{ m tot}$
²⁵⁸ Fm	1	1.3
	2 <i>a</i>	4.3
	2 <i>b</i>	4.2
²⁵² Cf	2	4.6
	3	3.0

The results for ²⁵⁸Fm are presented in two variants [Figs. 2(a) and 2(b)]. Clearly, the fission of ²⁵⁸Fm produced a sharp mass peak at symmetry related to very high kinetic energies of the fragments. Moreover, there is yield at large asymmetry connected with moderate kinetic energies, but it cannot be decided from the data if this second component should be represented by an asymmetric $Y_2(A)$ with usual variance or by a symmetric one with prodigious width. Our theory clearly favors the first variant.^{8,22} In both cases we find a dip at symmetry caused by the events with high kinetic energy which leaves little excitation for evaporation. The main difference between the two variants is a large displacement between the wings of $\overline{v}(A)$ in Fig. 2(a) caused by an asymmetric prescission shape. This is the main message of this figure. It should be relevant for the following reason: Also with better counting rates a decision if the low-energy component in ²⁵⁸Fm is symmetric or not will be difficult when only the mass yield is measured. If, however, $\overline{v}(A)$ is also recorded, the question can be answered.

The surprising features in Fig. 3 are the sawteeth at $A \simeq 80$ and 172. They are both caused by an extremely asymmetric prescission shape which seems to be necessary for the interpretation of new experimental^{4,5} and theoretical¹³ results. We see here that a measurement of $\overline{v}(A)$ can serve as a powerful indicator of separate fission modes.

The predictions do not seem to be very risky. For the fission of ²²⁷At, which is two-mode fission according to Britt, Wegner, and Gursky,²³ a $\overline{v}(A)$ similar to that in Fig. 2(a) was observed (see Fig. 5 in Ref. 19), the only difference being that ²²⁷Ac has at symmetry a long fission path, leaving more energy for the evaporation of neutrons. Furthermore, in 1985 I predicted the total neutron multiplicities in the fission of ²⁶⁰Md, a system similar to ²⁵⁸Fm.²⁴ If found $\overline{v}_{tot}=1.0$ for the high-energy component and $\overline{v}_{tot}=5.1$ for the low-energy events. These multiplicities were now measured with the results $\overline{v}_{tot}=2.0\pm0.8$ and $\overline{v}_{tot}=4.4\pm1.4$, respectively.²⁵ Finally, it is clear that my $\overline{v}(A)$ for ²⁵²Cf (Fig. 3) is in reasonable agreement with the data as they were measured up to now (cf. Ref. 18).

It should be possible to check both predictions in the

near future: The production of ²⁵⁸Fm is on the verge of being sufficient; it is also possible to use neutron counters to cover a sufficiently large solid angle. The dip at symmetry can be measured with gadolinium-loaded scintillation tanks; for such experiments, the predictions given in Table II may be useful. In the case of ²⁵²Cf, existing high-statistics data already seems to give an indication.²⁶ However, the total neutron multiplicity of the superasymmetric component (mode 3 in Table II) has not yet been measured.

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ACKNOWLEDGMENTS

I am grateful to K. Sümmerer for suggesting the problem. He, D. Hilscher, and W. Westmeier examined the feasibility of the experiments which are proposed in this paper. I am deeply obliged to C. Budtz-Jørgensen and H.-H. Knitter for numerous discussions; recently they showed me their finally evaluated but not yet published $\overline{\nu}(A)$ of ²⁵²Cf (*sf*) (Ref. 26). This data clearly exhibits the triple sawtooth as predicted in Fig. 3.

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