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CHARGE POLARIZATION AND THE ELONGATION OF THE FISSIONING NUCLEUS AT SCISSION

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Abstract. The deviation $\Delta Z =< Z > -Z_{UCD}$, of the charge of a fission fragment with given mass number A_F, from the unchanged charge distribution is calculated by the minimization of the total macroscopic energy at scission. The scission configuration is approximated by two spherical fragments with masses and charges (A_1, Z_1) and (A_2, Z_2) separated by a distance d between their interior surfaces. An analytical formula for ΔZ is deduced and applied to the nucleus ^{236}U at different mass divisions. A qualitative agreement with experimental data for the $^{235}U(n_{th}, f)$ reaction is obtained for a wide range of d values (from 6 fm to 12 fm). When the generally accepted variation of the distance d with the mass asymmetry is introduced, the agreement becomes quantitative.

Key words: low-energy nuclear fission, scission configuration, fission fragment charge, charge polarization.

1. INTRODUCTION

As most detailed experimental and theoretical studies of nuclear fission, efforts to understand how the charge of a fissioning nucleus is devided between the two fission fragments [1, 2] started during the Manhattan Project. The fundamental question is: to what extent the spatial distribution of the proton and neutron densities is preserved during the transition from the ground state to the scission point. The answer, as we know it today, is an encouraging "quite well" meaning that the fissioning nucleus is keeping its identity until the neck connecting the nascent fragments cracks, *i.e.*, till the end. Fission is therefore a process of its own and not a replica of another phenomenon: a heavy cluster decay for instance.

The search for deviations from the unchanged charge distribution (UCD) in fission has been stimulated by even older studies [3, 4] on the effect of the Coulomb repusion of the protons in ground-state nuclei. The increase of the proton density in the surface region (accompanied by a decrease of the neutron density due to the incompresibility of the nuclear matter) is counterbalanced by an increase in the symmetry energy. Even so the result was that Z/N is 21% [3] to 36% [4] larger at the surface than in the center of the nucleus. If true, the light fission fragment should have a larger $< Z/N >$ ratio than the heavy one since it has a larger surface over volume ratio.

The deviation $\Delta Z =< Z > -Z_{UCD}$ for a fragment of given mass number A is well established. It was obtained by precise measurements: mass-spectroscopic [5] for the light fragments and radiochemical for the heavy fragments [6]. It increases slightly with the mass asymmetry of the primary fragments and has an average value of 0.5 charge units. Although small this deviation may have an observable effect on the number of β-decays of a given fragment and, as a result, also on the delayed-neutron multiplicity.

From theoretical side, the deviation ΔZ has been calculated exclusively by the minimization of the total energy of the two fragments (separated or in contact) [2, 7–10]. Only in the first model [2] the charge polarization was included through a charge density gradient. In all other models the charge density was taken constant inside each fragment but with different values in the light and in the heavy fragment.

A simple analytical model (as in Ref. [8]) is used in the present study. The scission configuration is approximated by two spherical fragments, with masses and charges (A_1, Z_1, A_2, Z_2), separated by a distance d between their inner tips. Three terms dependent on the fragment charge are considered in the minimization: the symmetry energy (volume term only), the self Coulomb energies of each fragment and their Coulomb interaction.

As a new element, we introduce here a variation of d with the mass asymmetry. Calculations of nuclear shapes at conditional saddle and scission points [11, 12] in the frame of the Finite Range Liquid Drop Model (FRLDM) [13] extended to reflection-asymmetric nuclei [14] show indeed a net decrease of d at large asymmetries. At the same result one arrives by applying Swiatecki's scaling rule [15] to find a lighter symmetric fissioning system equivalent (*i.e.*, having similar macroscopic behaviour) with a heavy asymmetric system.

Apparently, the $< \Delta Z >$ value doesn't depend on the excitation energy [16, 17] meaning that it is a macroscopic property rather than a shell effect. The Liquid Drop Model (LDM) is therefore the proper reference. It would be inconsistent to deduce the variation of d with mass asymmetry from the variation of total kinetic energy; the latter being a strong manifestation the double magic ^{132}Sn.

2. AN ANALYTICAL MODEL

2.1. FORMALISM

Here we approximate the scission configuration by two spherical fragments with masses, charges and radii (A_1, Z_1, R_1) and (A_2, Z_2, R_2) separated by a distance d between their interior surfaces. For such a system, we calculate the deviation $< \Delta Z >$ that minimizes its total macroscopic energy: $E_1 + E_2 + E_{12}$, where

$$E_{12} = \frac{Z_1 Z_2 e^2}{r_0 \left(A_1^{1/3} + A_2^{1/3}\right) + d}$$

is the fragments' Coulomb interaction.

The macroscopic energy of each fragment is given by the semi-empirical mass formula of Bethe [18] and Weizsacker [19]:

$$E_i \simeq -a_v A_i + a_s A_i^{2/3} + a_{sym} \frac{(N_i - Z_i)^2}{A_i} + \frac{3}{5} \frac{Z_i^2 e^2}{r_0 A_i^{1/3}} - \delta(A_i) \; , i = 1, 2, \quad (1)$$

where a_v, a_s, a_{sym} are positive constants and r_0 is the nuclear radius parameter. The term $\delta(A_i)$ is a shift that accounts for more (less) stability of even-even (odd-odd) isobars in even-A nuclei. Only the charge dependent 3rd and 4th terms were included in the minimization.

2.2. FORMULA FOR $< \Delta Z >$

If we denote by Z and A the charge and mass of the fissioning nucleus, $Z_i^{UCD} = \frac{Z}{A} A_i$. Let us consider that the fragments' charges deviate slightly from this value:

$$Z_1 = \left(\frac{Z}{A}\right) A_1 + \Delta Z$$

$$Z_2 = \left(\frac{Z}{A}\right) A_2 - \Delta Z.$$

From the incompresibility condition the total nuclear density has to stay constant. Therefore the neutron numbers of the two fragments must also deviate slightly from $N_i^{UCD} = \frac{N}{A} A_i$:

$$N_1 = \left(\frac{N}{A}\right) A_1 - \Delta Z$$

$$N_2 = \left(\frac{N}{A}\right) A_2 + \Delta Z.$$

We analyze separately the electrostatic and isospin terms to better see their roles. With the above notations the Coulomb energy becomes:

$$
\begin{aligned}
E_{Coul.} &= \frac{3}{5}\frac{\tilde{Z_1}^2 e^2}{r_0 A_1^{1/3}} + \frac{3}{5}\frac{\tilde{Z_2}^2 e^2}{r_0 A_2^{1/3}} + \frac{\tilde{Z}_1 \tilde{Z}_2 e^2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d} \\
&= \frac{3}{5}\frac{\left(\left(\frac{Z}{A}\right)A_1+\Delta Z\right)^2 e^2}{r_0 A_1^{1/3}} + \frac{3}{5}\frac{\left(\left(\frac{Z}{A}\right)A_2-\Delta Z\right)^2 e^2}{r_0 A_2^{1/3}} \\
&\quad + \frac{\left(\left(\frac{Z}{A}\right)A_1+\Delta Z\right)\left(\left(\frac{Z}{A}\right)A_2-\Delta Z\right)e^2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d} \\
&= \frac{3}{5}\frac{\left(\left(\frac{Z}{A}\right)A_1\right)^2 e^2}{r_0 A_1^{1/3}} + \frac{3}{5}\frac{\left(\left(\frac{Z}{A}\right)A_2\right)^2 e^2}{r_0 A_2^{1/3}} + \frac{\left(\frac{Z}{A}\right)^2 A_1 A_2 e^2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d} \\
&\quad + \left(\frac{Z}{A}\right)\left\{2\frac{3}{5r_0}\left(A_1^{2/3}-A_2^{2/3}\right) + \frac{-A_1+A_2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d}\right\} e^2 \Delta Z \\
&\quad + \left\{\frac{3}{5}\frac{1}{r_0}\left(A_1^{-1/3}+A_2^{-1/3}\right) - \frac{1}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d}\right\} e^2 (\Delta Z)^2,
\end{aligned}
$$

where r_0=1.2 fm and e^2= 1.4398 MeV$\times$fm. Its minimum value corresponds to

$$
\Delta Z = -\left(\frac{Z}{A}\right)\frac{\frac{3}{5}\left(A_1^{2/3}-A_2^{2/3}\right)+\frac{1}{2}\frac{-A_1+A_2}{\left(A_1^{1/3}+A_2^{1/3}\right)+d/r_0}}{\frac{3}{5}\left(A_1^{-1/3}+A_2^{-1/3}\right)-\frac{1}{\left(A_1^{1/3}+A_2^{1/3}\right)+d/r_0}} \tag{2}
$$

One notices that the terms in the numerator have oposite signs. Let us assume $A_1 < A_2$. The value of ΔZ is determined by the balance between the Coulomb self-energy that favors a more symmetric charge division ($\Delta Z > 0$) and the Coulomb interaction that favors a more asymmetric charge division ($\Delta Z < 0$). However, irrespective of the mass division the first term is dominant even at d=0 as it can be inferred from Eq. (2). Hence the sign of the experimentally observed deviation is by all means reproduced. Its magnitude is sensitive to the separation distance d. This process has therefore the potential to provide the elongation of the fissioning nucleus at scission.

In the same way, the symmetry energy can be written as

$$
\begin{aligned}
E_{sym.} &= a_{sym}\frac{\left(\tilde{N}_1-\tilde{Z}_1\right)^2}{A_1}+a_{sym}\frac{\left(\tilde{N}_2-\tilde{Z}_2\right)^2}{A_2} \\
&= a_{sym}\frac{\left(\left(\frac{N}{A}\right)A_1-\Delta Z-\left(\frac{Z}{A}\right)A_1-\Delta Z\right)^2}{A_1} \\
&\quad +a_{sym}\frac{\left(\left(\frac{N}{A}\right)A_2+\Delta Z-\left(\frac{Z}{A}\right)A_2+\Delta Z\right)^2}{A_2} \\
&= a_{sym}\frac{\left(\left(\frac{N}{A}\right)A_1-\left(\frac{Z}{A}\right)A_1\right)^2}{A_1}+a_{sym}\frac{\left(\left(\frac{N}{A}\right)A_2-\left(\frac{Z}{A}\right)A_2\right)^2}{A_2} \\
&\quad +a_{sym}\left(\frac{4}{A_1}+\frac{4}{A_2}\right)(\Delta Z)^2 \ ,
\end{aligned}
$$

where a_{sym}= 23.285 MeV [20]. The first two terms represent the asymmetry energy for fragments with UCD. Being only quadratic in ΔZ, the asymmetry energy is neutral with respect to the charge division: decreasing this energy in one of the fragments automatically increases it in the complementary fragment. So its minimum value corresponds to ΔZ=0. It doesn't mean that it has no effect. Holding on UCD it considerably diminuishes the ΔZ value predicted by the Coulomb energy (Eq. (2)).

The sum of the Coulomb and symmetry energies:

$$
\begin{aligned}
E_{Coul.}+E_{sym.} &= \left\{\frac{3}{5}e^2\left(A_1^{-1/3}+A_2^{-1/3}\right)-\frac{e^2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d}\right\}(\Delta Z)^2 \\
&\quad +a_{sym}\left(\frac{4}{A_1}+\frac{4}{A_2}\right)(\Delta Z)^2 \\
&\quad +\left(\frac{Z}{A}\right)\left\{2\frac{3}{5r_0}e^2\left(A_1^{2/3}-A_2^{2/3}\right)+\frac{e^2(-A_1+A_2)}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d}\right\}\Delta Z \\
&\quad +\frac{3}{5}e^2\left(\frac{Z}{A}\right)^2\left(A_1^{2/3}+A_2^{2/3}\right)+\frac{\left(\frac{Z}{A}\right)^2A_1A_2e^2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d} \\
&\quad +a_{sym}\frac{\left(\left(\frac{N}{A}\right)A_1-\left(\frac{Z}{A}\right)A_1\right)^2}{A_1}+a_{sym}\frac{\left(\left(\frac{N}{A}\right)A_2-\left(\frac{Z}{A}\right)A_2\right)^2}{A_2} \qquad (3)
\end{aligned}
$$

has a minimum at:

$$
\Delta Z=-\frac{1}{2}\frac{\left(\frac{Z}{A}\right)2a_c\left(A_1^{2/3}-A_2^{2/3}\right)+\left(\frac{Z}{A}\right)e^2\frac{-A_1+A_2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d}}{a_{sym}\left(\frac{4}{A_1}+\frac{4}{A_2}\right)+a_c\left(A_1^{-1/3}+A_2^{-1/3}\right)-\frac{e^2}{r_0\left(A_1^{1/3}+A_2^{1/3}\right)+d}}. \qquad (4)
$$

The above formula can be simplified using the reduced mass $m_R = A_1A_2/(A_1+A_2)$ and total distance $D = d + R_1 + R_2$

$$\Delta Z = -\frac{1}{2}\left(\frac{Z}{A}\right)\frac{2a_c\left(A_1^{2/3}-A_2^{2/3}\right)-(A_1-A_2)\frac{e^2}{D}}{\frac{4a_{sym}}{m_R}+a_c\left(\frac{1}{A_1^{1/3}}+\frac{1}{A_2^{1/3}}\right)-\frac{e^2}{D}}\,. \tag{5}$$

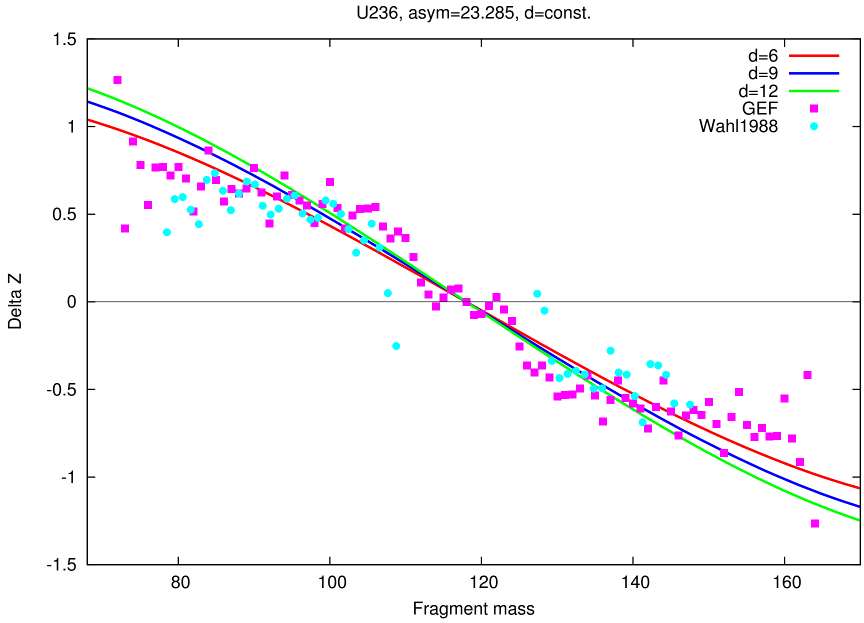

Fig. 1 – Charge deviations ΔZ as a function of fragment mass. Lines are calculations with a constant inter-fragment distance d. Full squares are generated by the GEF code [21]. Full circles are experimental data [22].

3. RESULTS FOR ^{236}U

In the previous Section, we derived a formula for the deviation of the fragment charges from the UCD value. Now we apply it for the reaction ^{235}U(n$_{th}, f$). In Fig. 1, the calculated charge deviations are shown as a function of mass asymmetry for three acceptable values of d. The red line is for the inter-fragment distance $d = 6$(fm). The green and blue lines are for $d = 9$(fm) and $d = 12$(fm), respectively. The full squares (magenta) are pre-neutron data for ^{236}U obtained with GEF code [21]. The full circles (cyan) are the corresponding experimentally determined ΔZ values [22]. The fluctuations in the data represent even-odd effects in the fission fragment yields. Our simple model doesn't contain such effects and

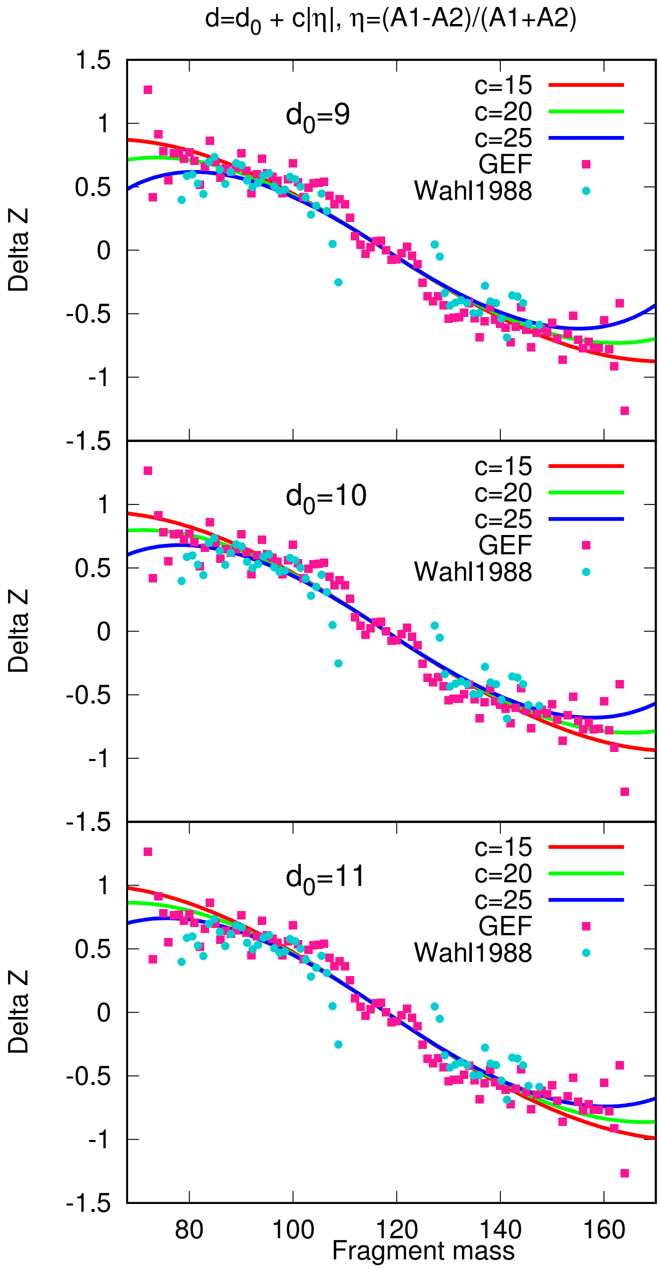

Fig. 2 – The same as in Fig. 1 but the lines are calculated with a distance d dependent on mass asymmetry.

hence we should compare only the average trend.

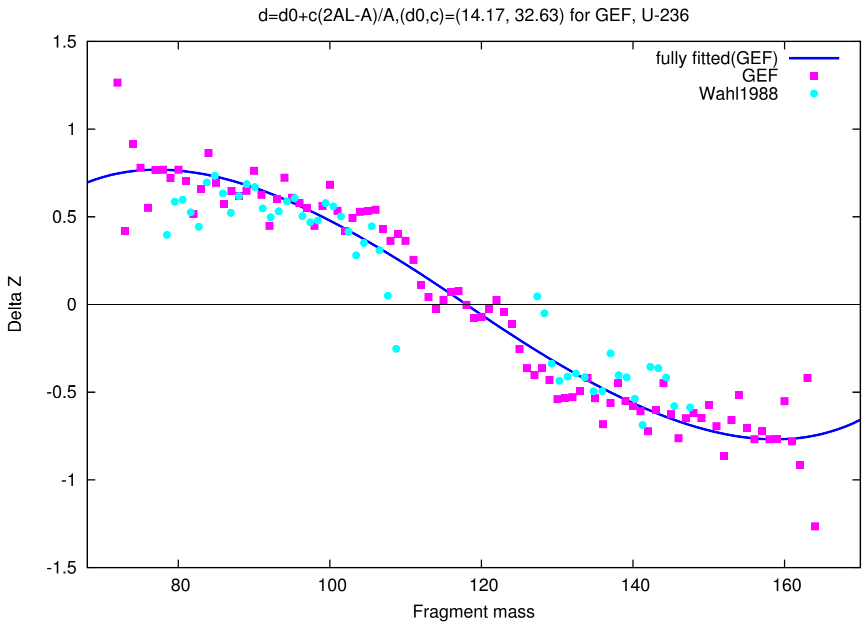

Fig. 3 – The fit of GEF data with Eq. (5) in which $d = d_0 - c \times |\eta|$.

We found that Eq. (5) can reproduce the observed behaviour especially in the region of the most probable masses (A_1= 90 to 105). Unfortunately the dependence on d is too weak in this domain to allow us to determine its value.

The disagreement at large asymmetries can be reduced if we consider a decrease of the distance d with the mass asymmetry parameter $\eta = \frac{A_1 - A_2}{A_1 + A_2}$, as suggested by macroscopic calculations of conditional saddle and scission points (see Introduction). Figure 2 shows results assuming a simple linear dependence: $d = d_0 - c \times |\eta|$. Different values of the parameters (c, d_0) that define this dependence are used, namely d_0 = 9,10,11 and c = 15,20,25. They are chosen so that the calculated curves stay inside the range of the data points.

Finally the best fit of GEF values is shown in Fig. 3. It is obtained with d_0 = 14.172 $\pm$ 3.835 and c = 32.631 $\pm$ 11.440. The large errors are due to the fine structure of the experimental data. Even so, the blue curve gives a satisfactory representation of the experimental data. To the extent to which the two-sphere approximation is justified, it means that the fissionning system is very elongated at scission (D_{cm}=25 fm) and that this elongation strongly depends on the mass asymmetry (d starts from 14 fm at symmetry and decreases to 1 fm at the largest measured asymmetry). Neither of these characteristics are generally accepted at present. The Wahl's data, being even more scattered, do not allow a reliable fit.

4. SUMMARY

The charge density is not perfectly conserved from the beginning to the end of the fission process. A simple analytical model was used to estimate the corresponding deviation ΔZ as a function of the fission-fragment mass ratio η at the scission point. It is essentially determined by the balance between the Coulomb self energy of the fragments and their Coulomb interaction. The first term is always dominant leading to $\Delta Z > 0$ for light fragments and $\Delta Z < 0$ for heavy fragments. The symmetry energy plays also a role: preferring an unchanged charge distribution, it reduces the absolute values of ΔZ predicted by the Coulomb term.

A semi-quantitative agreement with experimental data is obtained for a wide range of distances between the fission fragments at scission. When the generally accepted variation of this distance with the mass asymmetry is introduced, this agreement is improved. Since the general trend of the observed deviations can be explained without explicitely assuming charge polarization, the terminology generally used is, may be, inappropriate.

Two spherical separated fragments, used here, represents the simplest possible "immediately after scission" configuration. However the decision on charge division is taken "just before scission". Hence a more realistic calculation requires the minimization of the total energy of two nascent fragments connected by a thin neck (r_{neck}=2 fm) and this is our goal for the near future.

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REFERENCES

1. L. E. Glendenin, C. D. Coryell, R. R. Edwards, and M. H. Feldman, J. Am. Chem. Soc. **68**, 2411 (1946).
2. R. D. Present, Phys. Rev. **72**, 7 (1947).
3. E. Feenberg, Phys. Rev. **59**, 593 (1941).
4. E. Wigner, University of Pennsylvania Bicentennial Conference (1941).
5. H.-G. Clerc, W. Lang, H. Wohlfarth, H. Schrader, and K.-H. Schmidt, in Proc. 4th IAEA Symp. on Physics and Chemistry of Fission, Julich, 1979, Vol. II (IAEA, Vienna, 1980).
6. H. O. Denschlag, H. Brown, W. Faubel, G. Fischbach, H. Meixler G. Paffrath, W. Porsch, M. Weis, H. Schrader, G. Siegert, J. Blachot, Z. B. Alfassi, H. N. Erten, T. Izak-Biran, T. Tamai, A. C. Wahl, and K. Wolfsberg, in Proc. 4th IAEA Symp. on Physics and Chemistry of Fission, Julich, 1979, Vol. II (IAEA, Vienna, 1980).
7. H. M. Blann, University of California Radiation Laboratory Report No. UCRL-9190 (1960).
8. J. A. McHugh and M. C. Michel, Phys. Rev. **172**, 1160 (1968).
9. B. D. Wilkins, E. P. Steinberg, and R. R. Chasman, Phys. Rev. C **14**, 1832 (1976).
10. W. J. Swiatecki, J. Phys. (Paris) **33**, C5-45 (1972).
11. N. Carjan and J. M. Alexander, Phys. Rev. C **38**, 1692 (1988).
12. N. Carjan and M. Kaplan, Phys. Rev. C **45**, 2185 (1992).
13. H. J. Krappe, J. R. Nix, and A. J. Sierk, Phys. Rev. C **20**, 992 (1979).
14. K. T. R. Davies and A. J. Sierk, Phys. Rev. C **31**, 915 (1985).

15. W. J. Swiatecki, Phys. Scr. **24**, 113 (1981).
16. I. Nishinaka, H. Nakahara, and Y. Nagame, Eur. Phys. J. A **33**, 231 (2007).
17. I. Nishinaka, M. Tanikawa, Y. Nagame, and H. Nakahara, Eur. Phys. J. A **47**, 9 (2011).
18. H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 82 (1936).
19. C. F. von Weizsacker, Z. Phys. **96**, 431 (1935).
20. A. E. S. Green and N. A. Engler, Phys. Rev. **91**, 40 (1953).
21. K.-H. Schmidt, B. Jurado, and Ch. Amouroux, JEFF-Report 24, NEA/DB/DOC (2014).
22. A. C. Wahl, At. Data Nucl. Data Tables **39**, 1 (1988).