Coulomb effects in cold fission from reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f), ^{239}Pu(n_{th},f) and ^{252}Cf(sf)

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The Coulomb effect hypothesis, formerly used to interpret fluctuations in the curve of maximal total kinetic energy as a function of light fragment mass in reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f), is confirmed by the experimental yield of charge in high kinetic energy as well as in low excitation energy windows, respectively. In order to distinguish Coulomb effect from other, one must choose fragmentations with similar mass and energetic characteristics. Data from reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f), ^{239}Pu(n_{th},f) and ^{252}Cf(sf) show that, between two isobaric fragmentations with similar Q-values, the more asymmetric charge split reaches the higher value of total kinetic energy. Moreover, in charge splits with different Q-values, similar preference for asymmetrical fragmentations is observed in low excitation energy windows.

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1. Introduction

In 1979, at the High Flux Reactor (HFR) of Laue-Langevin Institute (ILL), the reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f), respectively, were studied by C. Signarbieux *et al.* [1, 2]. They measured the maximal total kinetic energy as a function of primary fragment mass, $K_{max}(A_L)$ for light fragment masses from 80 to 108.

In a scission point model, assuming that the pre-scission kinetic energy is zero, the maximal total kinetic energy was interpreted as a result of the most compact configuration permitted by the interplay of the Coulomb interaction energy between the two complementary fragments and the total deformation energy limited by the available energy (Q-value) [1, 2].

The total deformation energy of two complementary fragments (D) and the Coulomb interaction energy between these fragments (C) are calculated. In the equipotential energy ($P = D + C$) line associated to the Q-value, the deformation corresponding to the maximal value of C is selected. With this constraint, the maximal value of C (C_{max}) is not higher than the Q-value.

One can easily show that between neighbouring mass fragmentations with similar Q-values, the higher maximal Coulomb interaction energy (C_{max}) and, consequently, the higher K_{max} value, will be reached by the fragmentation with the lower light fragment charge (Z_L). This named Coulomb effect reproduces the observed fluctuations in experimental $K_{max}(A_L)$ curves, with a period of 5 units of fragment mass, which is the average of the period of change in the fragment even charge that maximizes the Q-value [2, 3, 4].

2. Basics of Coulomb effects in cold fission

In a scission point model, the potential energy (P) of a scission configuration corresponding to a light fragment charge Z_L is given by

$$P^{Z_L}(\mathscr{D}) = D^{Z_L}(\mathscr{D}) + C^{Z_L}(\mathscr{D})$$

where D is the total deformation energy of fragments, C is the Coulomb interaction energy between complementary fragments, and $\mathscr{D}$ represents the deformed configuration shape. See Fig. 1. In general, at scission, the fragments have a free energy (E_{free}) which is spend in pre-scission kinetic and intrinsec energy of fragments, respectively, obeying the relation $Q = P + E_{free}$. The most compact configuration obeys the relation $E_{free} = 0$, then $Q = P$.

In order to explain the Coulomb effect in isobaric splits in cold fission, it matters first to show that, for the same shape configurations, the more asymmetric charge split has a lower Coulomb interaction energy. Let's take the case of two spherical fragments. The Coulomb interaction energy between two complementary hypothetical spherical fragments at scission configuration is given by

$$C_{sph}^{Z_L} = \frac{1}{4\pi\varepsilon_0} \frac{Z_L(Z_f - Z_L)e^2}{R_L + R_H + d}$$

where ε_0 is the electrical permittivity, e is the electron charge, R_L and R_H are the radii of light and heavy fragment, respectively, and d is the distance between surfaces of fragments. In this paper it is assumed that $d = 2$ fm. The nucleus radius for each fragment is given by the relation $R = 1.24A^{1/3}$

fm. Then, one can show that

$$\Delta C_{\text{sph}}\left(Z_L, Z_L - 1\right) = C_{\text{sph}}^{Z_L} - C_{\text{sph}}^{Z_L-1} = \frac{(Z_f - 2Z_L + 1)}{Z_L(Z_f - Z_L)} C_{\text{sph}}^{Z_L}.$$

Let us take two cases of charge splits from fission of nucleus ^{236}U which has $Z_F = 92$. The first case corresponding to $Z_L = 46$ for which the relative variation ΔC_{sph} produced by changing to $Z_L - 1 = 45$ will be nearly zero; and the second case, a much asymmetric charge split, corresponding to $Z_L = 30$, for which the variation ΔC_{sph} produced by changing to $Z_L - 1 = 29$ will be approximately 3.5 MeV. This gives an idea of how much the Coulomb effect increases with asymmetry of charge split.

In general, the Coulomb interaction energy between spherical fragments is higher than the Q−value. Therefore, in a scission configuration, fragments must be deformed. Let us assume that, for isobaric split A_L/A_H, $C^{Z_L}(\mathscr{D})$ is the interaction Coulomb energy between the two complementary fragments corresponding to light charge Z_L and scission configuration shape $\mathscr{D}$, with fragments nearly spherical. If one takes two isobaric splits with light fragment charges Z_L and $Z_L - 1$, respectively, one obtains the relation

Figure 1: Solid lines represent total deformation energy (*D*), Coulomb interaction energy (*C*) and potential energy (*P = D + C*) of scission configurations as a function of deformation, respectively, for the fragmentation $^{96}_{38}Sr/^{140}_{54}Xe$. The space of deformation is limited by the total available energy (*Q*). From Ref. [4].

$$C^{Z_L}\left(\mathscr{D}\right) - C^{Z_L-1}\left(\mathscr{D}\right) \cong \frac{Z_f - 2Z_L + 1}{Z_L(Z_f - Z_L)} C^{Z_L}\left(\mathscr{D}\right).$$

From this relation, for the same shape of scission configuration, one can show that

$$C^{Z_L-1}\left(\mathscr{D}\right) < C^{Z_L}\left(\mathscr{D}\right).$$

In consequence, if one assumes that

$$D^{Z_L-1}\left(\mathscr{D}\right) = D^{Z_L}\left(\mathscr{D}\right)$$

one can show that

$$P^{Z_L-1}\left(\mathscr{D}\right) < P^{Z_L}\left(\mathscr{D}\right).$$

3. The maximal value of total kinetic energy

The fragment deformation energy and Coulomb interaction energy between fragments are limited by the Q-value of the reaction. The maximal Coulomb interaction energy corresponding to
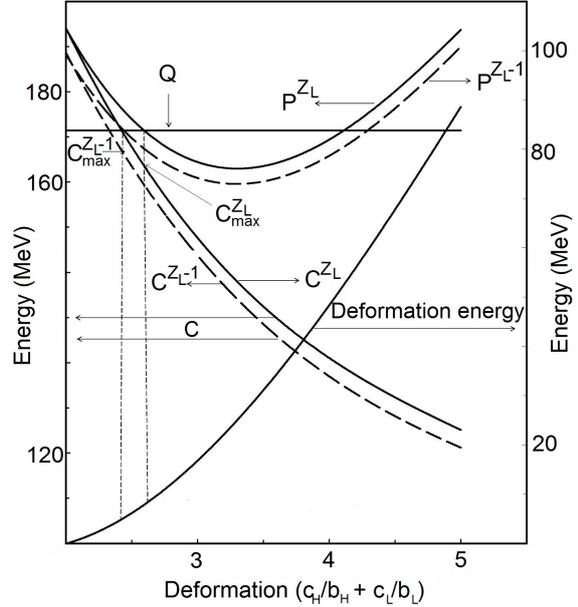
Z_L ($C_{max}^{Z_L}$) and the minimal value of deformation energy $D_{min}^{Z_L}$ obeys the relation

$$C_{max}^{Z_L} = Q - D_{min}^{Z_L}.$$

Similarly, the relation corresponding to fragmentation with light fragment charge $Z_L - 1$ will be

$$C_{max}^{Z_L-1} = Q - D_{min}^{Z_L-1}.$$

The deformation energy (D) increases with $\mathscr{D}$. Then the most compact configuration corresponding to $Z_L - 1$ has a lower deformation than the corresponding to Z_L. See Fig. 1. In consequence:

$$D_{min}^{Z_L-1} < D_{min}^{Z_L}.$$

From these three relations one deduces that

$$C_{max}^{Z_L-1} > C_{max}^{Z_L}.$$

Therefore, it is expected that among isobaric splits having similar Q- values, the more asymmetric charge split will reach a more compact configuration, which corresponds to a lower deformation energy, a higher Coulomb interaction energy and, therefore, a higher maximal total kinetic energy.

4. Experimental data confirming the Coulomb effect hypothesis

According to the Coulomb hypothesis, if the total kinetic energy is due to Coulomb interaction, in the asymmetrical fragmentation region (light fragment mass lower than 100), it is expected that

$$K_{max}^{Z_L-1} > K_{max}^{Z_L}.$$

Therefore, the higher yield will corresponds to the more asymmetrical charge split. Data confirming the Coulomb effect hypothesis will be shown in the following paragraphs. In order to exclude pairing and shell effects in the test of Coulomb hypothesis, one must only take into account charges with same parity and regions exempt of shell transitions.

Clerc *et al.* [5] measured the charge and mass yield in light fragment kinetic energy windows in reactions $^{233}U(n_{th},f)$ and $^{235}U(n_{th},f)$. The highest kinetic energies studied are 110.55 MeV for $^{233}U(n_{th},f)$ [6] and 108.0 MeV for $^{235}U(n_{th},f)$ [7], which correspond to total kinetic energies appreciably below the highest Q-values. They observe that the excitation energy of the fragments increases with increasing asymmetry of the mass split, result which agrees with the Coulomb effect hypothesis. In order to not exceed Q-values at least one fragment must be deformed, and that means deformation energy, excepts if the needed deformation corresponds to fragment ground state, as in the case of ^{104}Mo, observed by Montoya *et al.* [2, 3, 4]. However, Clerc *et al.* interpret this result playing with the distance between fragments centers (R_c) corresponding to a Coulomb energy equal to the Q-value, the interaction radius R_{int} and tunneling through the barrier separating the "fission valley" from the valley corresponding to two separated fragments ("fusion valley"). If one assumes that those variables have the same values for neighbouring mass and even or odd charge fragmentations, respectively, the Coulomb effect hypothesis still valid in the Clerc *et al.*' approach.

In 1985, Trochon *et al.* [8] presented the mass and charge yield corresponding to the highest values of $K_{max}(A_L)$ ($K \geq 194$ MeV) referred to reaction $^{235}U(n_{th},f)$. In this region, the masses 102 and 104 correspond to charges 40 and 42, respectively. Although the Q-value corresponding to charge 40 is 3 MeV lower than the corresponding to charge 42, their corresponding yields are similar. Neighbouring masses with different even or odd charges, respectively, with similar Q-values can be studied to test the Coulomb effect hypothesis. The fragment with mass 86 and charge 34 has a Q-value about 2 MeV lower than the corresponding to the fragment with mass 88 and charge 36 but its K_{max} is 1 MeV higher. Moreover, for the mass split 104/132 Trochon *et al.* observed that only the odd charge split 41/51 reaches a "true" cold fission ($K_{max} \cong Q$), while K_{max} referred to the magic charge split 42/50 is 3 MeV lower than the corresponding Q-value [8].

Figure 2: Experimental yield of charge, corresponding to total excitation energy values of 7, 9 and 11 MeV, respectively, from reaction $^{252}Cf(sf)$, as measured by F.-J. Hambsch *et al.* [13].

They also observed that, for isobaric fragmentation, the highest kinetic energy is reached by the charge corresponding to the highest Q-value; except for the mass 91 for which the charge that maximizes Q is 37, but the highest K is reached by the charge 36.

In 1986, G. Simon *et al.* [9] presented results on charge and mass distribution for cold fragmentation from reactions $^{233}U(n_{th},f)$, $^{235}U(n_{th},f)$ and $^{239}Pu(n_{th},f)$, respectively. Among other they arrive to the following conclusions: i) the maximal total kinetic energy are mostly reached by the charges maximizing the Q-value ii) for similar Q-values the more asymmetric charge split reach higher values of total kinetic energy.

According to Coulomb effect, a bump in K_{max} as a function of mass must be produced with a change of the charge that maximizes the Q-value, which occurs with a period of 5 or 6 amu. For the reaction $^{235}U(n_{th},f)$ the change of the light fragment charge that maximizes the Q-value occurs for masses 86, 90, 96 and 102, corresponding to charges 34, 36, 38 and 40, respectively. In 1991 C. Signarbieux *et al.* [10] showed that the total excitation energy present minimal values for those fragments.

In 1988 U. Quade *et al.* [6] studied cold fragmentation in the reaction $^{233}U(n_{th},f)$. For the mass split 82/152, although $Z_L = 32$ corresponds to a Q-value approximately 4 MeV lower than the corresponding to $Z_L = 34$, its probability is higher in the coldest region.

For the mass split 89/145, comparing two odd charge splits referred to $Z_L = 35$ and $Z_L = 37$, respectively, although $Z_L = 35$ corresponds to a Q-value 1 MeV lower than the corresponding to $Z_L = 37$, its probability is higher in the coldest region.

Similarly, for the mass split 94/140, between the two odd charge splits referred to $Z_L = 37$ and
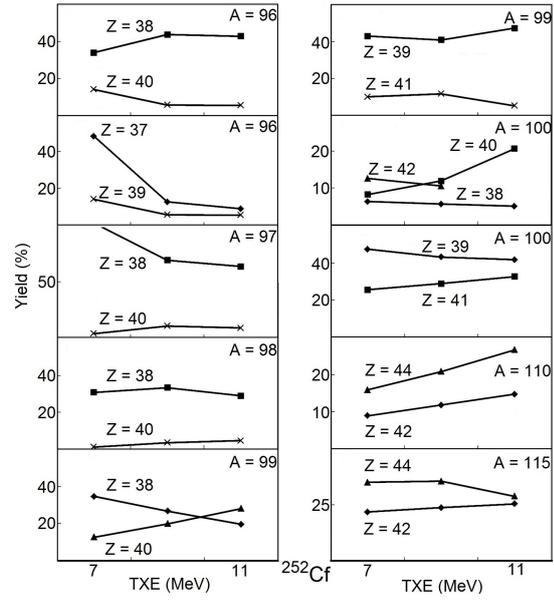
$Z_L = 39$, respectively, having a similar Q-value, in the cold fission region, the probability for $Z_L = 37$ is higher than the corresponding to $Z_L = 39$.

U. Quade *et al.* noticed, in cold fragmentations, the preferential formation of the element with the highest Q-value. However, among the 28 masses they found 10 exceptions. In these exceptions, the highest probability corresponds to a light fragment charge lower that the corresponding to the highest Q-value [11].

In 1991 Gönnenwein *et al.* propose the "Tip model of cold fission" [12] which is a more elaborated version of the model proposed in 1984 by Clerc *et al.* [5]. They include the deformation properties of nuclei in their ground states. They propose the concept of "true cold fission" which correspond to a critical minimum tip distance. This distance, as derived from the theoretical deformations, is assumed to be 3.0 fm.

In Fig. 2, experimental data corresponding to spontaneous fission of ^{252}Cf obtained by 1993 F.-J. Hambsch *et al.* [13] is presented. They presented the charge and mass yield referred to total excitation energy values equal to 7, 9 and 11 MeV, respectively. Taking into account the cases with three available excitation energies, corresponding to cold isobaric fragmentations with charge with the same parity, one can observe that the higher yield corresponds to the lower light fragment charge. In Fig. 3 experimental data obtained in 1994 by W. Schwab *et al.* referred to reaction $^{233}U(n_{th},f)$ are presented.

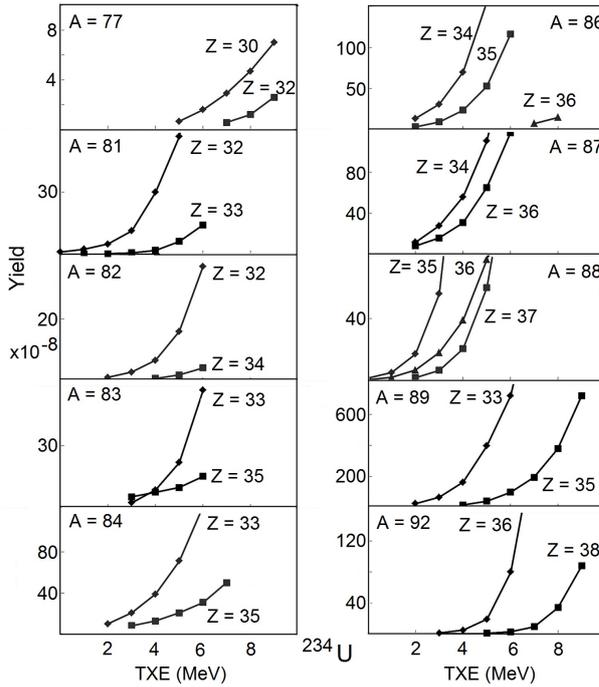Definitely there is a clear trend to prefer more asymmetric charge split in cold fission [14]. They calculated the mass and charge yied as a function of excitation energy. Comparing cold isobaric fragmentations with charge with the same parity, in the region of low excitation energy, the higher yield correspond to the lower light fragment charge.

The Coulomb effect is more evident in the region associated to the more asymmetric fragmentations. For light fragments heavier than 100 amu, other effects seems to be reflected on charge and mass yield. In the region corresponding to transitional fragments with mass number around 100 and neutron numbers $N \geq 58$ it is expected that deformabilities are reflected on the charge and mass yield. For instance, in the mass fragmentation 100/152 with 7 MeV of excitation energy, the charge $Z = 42$ is preferred to $Z = 38, 40$. In mass fragmentations 110/142 the yield of $Z = 44$ ($N = 66$) is higher than the

Figure 3: Experimental yield of charge, as a function of total excitation energy, from reaction $^{233}U(n_{th},f)$, as measured by W. Schwab *et al.* [14].

corresponding to $Z = 42$ ($N = 68$). For the mass fragmentation 115/137, the yield of charge $Z = 44$ ($N = 61$) is higher than the corresponding to $Z = 42$ ($N = 63$).

5. Conclusion

It was shown that, in the cold region of thermal neutron induced fission of ^{233}U, ^{235}U, ^{239}Pu and spontaneous fission of ^{252}Cf, respectively, between isobaric charge fragmentations in the asymmetric region ($A < 100$), with similar Q-values of the reaction, the more asymmetric charge fragmentation reaches the higher maximal total kinetic energy.

This results is interpreted, in a scission point model, as a "Coulomb effect" [2, 3, 4]: between charge splits with similar Q-value, a lower light fragment charge corresponds to a lower Coulomb repulsion, which will permit to reach a more compact configuration and, as a consequence, a lower minimal deformation energy, and a higher maximal Coulomb interaction energy. The final result of that will be a higher maximal fragment kinetic energy. For charge splits with different Q-values, the more asymmetrical charge splits are associated to the lower minimal excitation energy. The most compact configuration could be interpreted in terms of fragments shapes [2, 3, 4] or in terms of distance between fragments [12]: more compact configuration is equivalent to lower distance between complementary fragments.

In cold fission, fragment mass, charge and kinetic energy distribution can be interpreted in terms of shell effects, odd-even effects, deformabilities, fragment ground state deformation, Coulomb effects or other characteristis of the fission process. The contribution of this paper is to identify cases where Coulomb effect are a predominant effect.

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