

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

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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  $^{233}\text{U}(n_{\text{th}},\text{f})$ ,  $^{235}\text{U}(n_{\text{th}},\text{f})$  and  $^{239}\text{Pu}(n_{\text{th}},\text{f})$ , is confirmed in high kinetic energy as well as in low excitation energy windows, respectively. Data from reactions  $^{233}\text{U}(n_{\text{th}},\text{f})$ ,  $^{235}\text{U}(n_{\text{th}},\text{f})$ ,  $^{239}\text{Pu}(n_{\text{th}},\text{f})$  and  $^{252}\text{Cf}(\text{sf})$  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 low excitation energy windows, between two isobaric charge splits with different  $Q$ -values, the more asymmetrical fragmentations is preferred.

**Keywords:** Fission fragments; kinetic energy; charge; mass; cold fission.

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## 1. Introduction

Among the most studied properties of nuclear fission of actinides are the distributions of mass and kinetic energy associated to complementary fragments [1]. F. Pleasonton found that the highest total kinetic energy is around 190 MeV [2]. However, those distributions are disturbed by neutron emission. In order to describe one of the consequences of neutron emission, let's suppose that a nucleus with proton number  $Z_f$  and mass number  $A_f$  splits into complementary light (L) and heavy (H) fragments corresponding to primary mass numbers  $A_L$  and  $A_H$ , and proton numbers  $Z_L$  and  $Z_H$ , having kinetic energies  $E_L$  and  $E_H$ , respectively. After neutron emission, those fragments will end with mass numbers

$$m_L = A_L - n_L$$

and

$$m_H = A_H - n_H,$$

where  $n_L$  and  $n_H$  are the numbers of neutrons emitted by the light and heavy fragments, respectively. The corresponding final kinetic energies associated to those fragments will be

$$e_L \cong E_L \left( 1 - \frac{n_L}{A_L} \right)$$

and

$$e_H \cong E_H \left( 1 - \frac{n_H}{A_H} \right),$$

respectively.

In 1979, at the High Flux Reactor (HFR) of Laue-Langevin Institute (ILL), in order to avoid neutron emission effects, the cold fission regions, associated to the highest values of kinetic energy, consequently to the lowest values of total excitation energy, in reactions  $^{233}\text{U}(n_{\text{th}},\text{f})$ ,  $^{235}\text{U}(n_{\text{th}},\text{f})$  and  $^{239}\text{Pu}(n_{\text{th}},\text{f})$ , respectively, were studied by C. Signarbieux *et al.* [3, 4]. Using the difference of time of flight technique, with solid detectors to measure the fragment kinetic energy, they succeeded to separate neighbouring primary masses, which permitted them to measure the maximal

total kinetic energy as a function of primary fragment mass,  $K_{\max}(A_L)$  for light fragment masses from 80 to 108.

At the Lohengrin spectrometer in Grenoble, P. Armbruster *et al.* [5] measured the mass and charge distribution as function of light fragment kinetic energy from reactions  $^{233}\text{U}(n_{\text{th}},\text{f})$ . They measured light fragment mass from 80 to 105. Like C. Signarbieux *et al.* [3, 4] they interpreted the structure of mass yield for cold fragmentation as dominated by potential energy surface of the scission configuration which is influenced by shell effects. They also agree with Signarbieux *et al.* that superfluidity is at least partially destroyed. P. Armbruster *et al.* conclude that the lowest excitation energy occurs for masses around 92, while C. Signarbieux *et al.* [3] suggest that this phenomenon occurs to the fragment pair ( $^{104}\text{Mo}$ ,  $^{130}\text{Sn}$ ).

In order to interpret fluctuations in experimental  $K_{\max}(A_L)$  curves, M. Montoya *et al.* proposed the hypothesis that the maximal value of final total kinetic energy corresponds to fragments that began with a compact scission configuration with null free energy [4, 6, 7]. If this hypothesis, called Coulomb effect, is true 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  $K_{\max}$ , will be reached by the fragmentation with the lower light fragment charge ( $Z_L$ ).

In this paper the theoretical basis of the Coulomb effect hypothesis are firstly explained. Then several experimental results on charge yield in cold fragmentations, compatible with that hypothesis are presented.

## 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 the relation

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

where  $D$  is the total deformation energy of fragments,  $C$  is the Coulomb interaction energy between complementary fragments, and  $\mathcal{D}$  represents the elongation of the scission configuration, which may be defined as:

$$\mathcal{D} = \frac{c_H}{b_H} + \frac{c_L}{b_L},$$

where  $b$  and  $c$  are the ellipsoidal semi axes referred to heavy (H) and light (L) fragments, respectively (It is assumed that  $a_L = b_L$  and  $a_H = b_H$ ). See Fig. 1.

In general, at scission, the fragments have a free energy ( $E_{\text{free}}$ ) which is spent in pre-scission kinetic energy and intrinsic energy of fragments, respectively, obeying the relation

$$Q = P + E_{\text{free}}.$$

The most compact configuration obeys the relation  $E_{\text{free}} = 0$ , 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

$$C_{\text{sph}}^{Z_L} = \frac{1}{4\pi\epsilon_0} \Delta \frac{Z_L(Z_f - Z_L)e^2}{R_L + R_H + d},$$

where  $\epsilon_0$  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

$$\begin{aligned} \Delta C_{\text{sph}}(Z_L, Z_L - 1) &= C_{\text{sph}}^{Z_L} - C_{\text{sph}}^{Z_L-1} \\ &= \frac{(Z_f - 2Z_L + 1)}{Z_L(Z_f - Z_L)} C_{\text{sph}}^{Z_L}. \end{aligned}$$

Let's take two cases of charge splits from fission of nucleus  $^{236}\text{U}$  which has  $Z_f = 92$ . The first case corresponding to  $Z_L = 46$  for which the relative variation  $\Delta C_{\text{sph}}$  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  $\Delta C_{\text{sph}}$  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's assume that, for isobaric split  $A_L/A_H$ ,  $C^{Z_L}(\mathcal{D})$  is the

interaction Coulomb energy between the two complementary fragments corresponding to light charge  $Z_L$  and scission configuration shape  $\mathcal{D}$ , 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

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

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

$$C^{Z_L-1}(\mathcal{D}) < C^{Z_L}(\mathcal{D}).$$

In consequence, if one assumes that

$$D^{Z_L-1}(\mathcal{D}) = D^{Z_L}(\mathcal{D}),$$

one obtains relation

$$P^{Z_L-1}(\mathcal{D}) < P^{Z_L}(\mathcal{D}).$$

See Fig. 1.

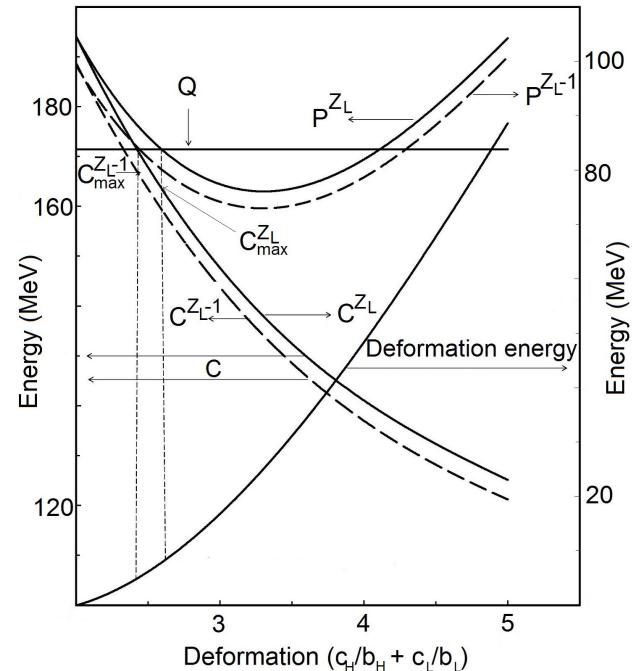


FIGURE 1. Low energy fission of actinides. Schematically, solid lines represent the total deformation energy ( $D$ ), the Coulomb interaction energy ( $C$ ) and the potential energy ( $P = D + C$ ) as a function of the elongation of a scission configuration, corresponding to light fragment charge  $Z_L$ . The space of deformation is limited by the total available energy ( $Q$ ). Dashed lines represent similar curves corresponding to the neighbouring more asymmetric charge split ( $Z_L - 1$ ) but having the same  $Q$ -value. One can see that the lower minimal deformation corresponds to the lower light fragment charge. As a result, the maximal Coulomb interaction energy, which will be converted in kinetic energy, corresponds to charge  $Z_L$ . This figure is based on a similar figure taken from Ref. 7).

### 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  $\mathcal{D}$ . 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 between two isobaric splits having similar  $Q$ -values, the more asymmetric charge split will reach a more compact configuration, which corresponds to a lower deformation energy, then 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, between two isobaric fragmentations, if

$$Q(Z1) \approx Q(Z2),$$

where  $Q(Z1)$  and  $Q(Z2)$  are the  $Q$ -values corresponding to the light fragment charges  $Z1$  and  $Z2$ , respectively, and  $Z1 > Z2$ , then

$$K_{\max}^{Z2} > K_{\max}^{Z1}.$$

Therefore, the higher yield will correspond to the more asymmetrical charge split. Data confirming the Coulomb effect hypothesis will be shown in the following paragraphs.

#### 4.1. $^{233}\text{U}(n_{\text{th}},\text{f})$

In 1986, H.-G. Clerc *et al.* [8] presented the charge and mass yield in light fragment kinetic energy windows in the reaction  $^{233}\text{U}(n_{\text{th}},\text{f})$ . The highest light fragment kinetic energy window studied was 110.55 MeV. In this energy window, between two isobaric charge splits, a preference for the light fragment charge corresponding to the highest  $Q$ -value is mostly observed. However, among the 27 measured light fragment masses, there are ten exceptions to that rule. For these exceptions, the highest probability corresponds to a

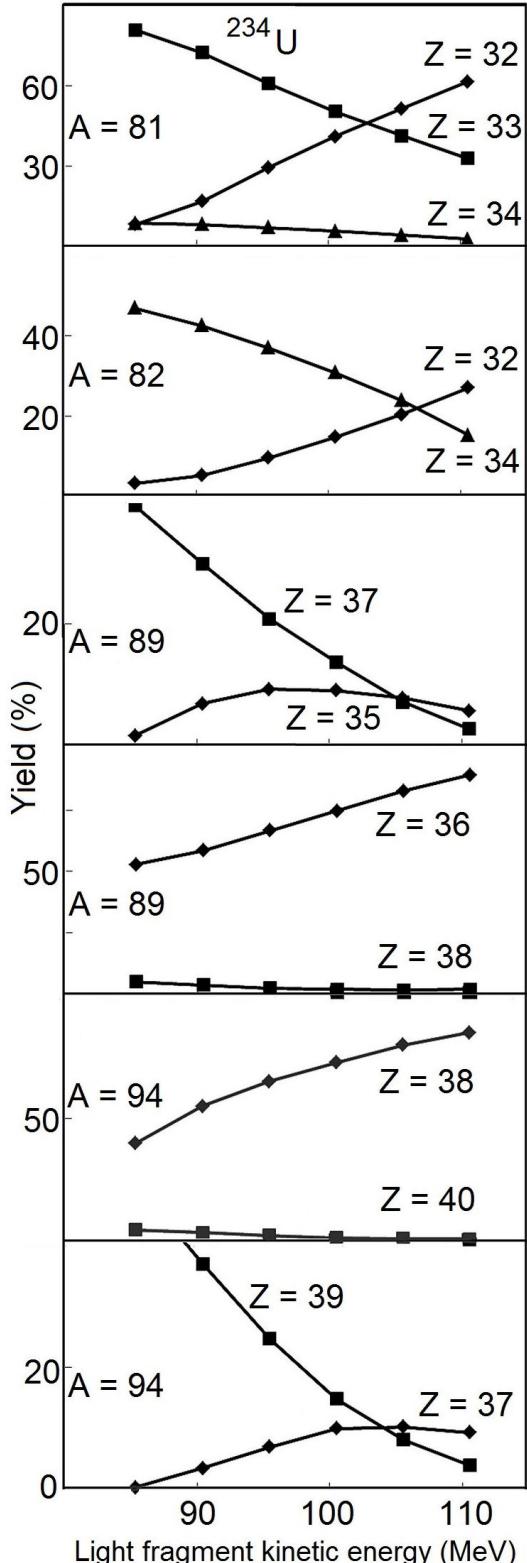


FIGURE 2. Experimental yield of charge, as a function of light fragment kinetic energy, corresponding to some isobaric splits with similar  $Q$ -values from reaction  $^{233}\text{U}(n_{\text{th}},\text{f})$ , as measured by U. Quade *et al.* [10]. If one compares yields of odd or even charges, respectively, at the highest measured kinetic energy ( $E_L = 110.55$  MeV), the higher yield corresponds to the lower light fragment charge.

TABLE I. Cold fission in reaction  $^{233}\text{U}(n_{\text{th}},\text{f})$ . From data referred to charge and mass yield of light fragments with 110.55 MeV, taken from Ref. 8, ten cases for which, as exceptions, the charge with the highest yield is not the corresponding to the highest  $Q$ -value are presented. For each mass ( $A$ ) the light fragment charges  $Z_1$  and  $Z_2$  correspond to  $Q_1$  and  $Q_2$ , respectively. Although  $Q_1 > Q_2$  the highest yield corresponds to  $Z_2$ . The  $Q$ -values are calculated using the atomic mass from Ref. 9.

$A$	$Z_1$	$Z_2$	$Q_1$ (MeV)	$Q_2$ (MeV)
		(Highest yield)		
81	33	32	179.1	178.6
82	34	33	181.6	180.7
85	35	34	184.3	184.1
86	36	34	186.2	185.9
87	36	35	186.2	185.6
91	37	36	190.4	190.2
92	38	37	198.3	197.5
101	41	40	197.3	192.8
102	42	40	205.1	201.2
103	42	41	203.1	202.2

a light fragment charge lower than the charge corresponding to the highest  $Q$ -value. See Fig. 2 and Table I. The  $Q$ -values are calculated using the atomic mass table of G. Audi *et al.* [9]. H.-G. 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  $R_{\text{int}}$  and tunneling through the barrier separating the fission valley from the valley corresponding to two separated fragments (fusion valley).

The above mentioned experimental results were confirmed in 1988 by U. Quade *et al.* [10]. Let's see some of these results.

For the mass split 82/152, the charges  $Z_1 = 34$  and  $Z_2 = 32$  correspond to  $Q$ -values equal to 181.6 MeV and 180.7 MeV, respectively; nevertheless, although the higher  $Q$ -value corresponds to  $Z_2 = 34$ , the higher probability is referred to the lower charge:  $Z_2 = 32$ . For the mass split 89/145, the charges  $Z_1 = 37$  and  $Z_2 = 35$  correspond to  $Q$ -values equal to 186.7 MeV and 186.5 MeV, respectively; nevertheless, although the higher  $Q$ -value corresponds to  $Z_1 = 37$ , the higher probability corresponds to the lower charge:  $Z_2 = 35$ . For the mass split 94/140, the charges  $Z_1 = 39$  and  $Z_2 = 37$  correspond to  $Q$ -values 191.6 MeV and 190.5 MeV, respectively; nevertheless, although the higher  $Q$ -value corresponds to  $Z_1 = 39$ , the higher probability corresponds to the lower charge:  $Z_2 = 37$ . For the mass split 81/153, the charges  $Z_1 = 33$  and  $Z_2 = 32$  correspond to  $Q$ -values, 179.2 MeV and 178.6 MeV, respectively; although the higher  $Q$ -value corresponds to  $Z_1 = 33$ , the higher probability corresponds to  $Z_2 = 32$ . It is interesting to notice

that in this last case the probability of an even charge split is higher than the corresponding to the odd charge split.

In 1991, to interpret those results, F. Gönnenwein and B. Börsig propose the “Tip model of cold fission” [11] which is a more elaborated version of the model proposed in 1984 by H.-G. Clerc *et al.* [8]. They include the deformation properties of nuclei in their ground states. They propose the concept of “true cold fission” which corresponds to a critical minimum tip distance. This distance, as derived from the theoretical deformations, is assumed to be 3.0 fm.

Confirming the Coulomb effect hypothesis, in 1994, W. Schwab *et al.* show that, in the reaction  $^{233}\text{U}(n_{\text{th}},\text{f})$ , definitely there is a clear trend to prefer more asymmetric charge split in cold fission [12]. They calculated the mass and charge yield as a function of excitation energy. Comparing cold isobaric fragmentations with charges with the same parity, in the region of low excitation energy, the highest yield corresponds to the lower light fragment charge. See Fig. 3.

#### 4.2. $^{235}\text{U}(n_{\text{th}},\text{f})$

In 1980, for the reaction  $^{235}\text{U}(n_{\text{th}},\text{f})$ , W. Lang *et al.* [13] presented experimental results about charge and mass yield in light fragment kinetic energy windows. In 1986, using those results, M. Montoya *et al.* [7] showed that, in the kinetic energy window of 108 MeV (the highest studied window) between two isobaric fragmentations with similar  $Q$ -values, the more asymmetric charge split is preferred.

In 1986, G. Simon *et al.* [14] measured the highest value of total kinetic energy as a function of light fragment charge and mass for reactions  $^{233}\text{U}(n_{\text{th}},\text{f})$ ,  $^{235}\text{U}(n_{\text{th}},\text{f})$  and  $^{239}\text{Pu}(n_{\text{th}},\text{f})$ , respectively. In the case of the reaction  $^{235}\text{U}(n_{\text{th}},\text{f})$  they observed that the maximal total kinetic energy are mostly reached by the charges maximizing the  $Q$ -value. The exceptions correspond to cases of similar  $Q$ -values, for which the highest values of total kinetic energy is reached by the more asymmetric charge split. See Tabla II.

In 1991 C. Signarbieux *et al.* [15] confirmed those results except for mass 88. For this mass, they found that the

TABLE II. Cold fission in reaction  $^{235}\text{U}(n_{\text{th}},\text{f})$ . Four cases of masses ( $A$ ) for which, as exceptions, the charge that reach the highest total kinetic energy is not  $Z_1$ , corresponding to the highest  $Q$ -value ( $Q_1$ ). For these exceptions the light fragment charge ( $Z_2$ ) that reach the highest kinetic energy is lower than ( $Z_1$ ). Data taken from Ref. 14. The  $Q$ -values are calculated using the atomic mass from Ref. 9.

$A$	$Z_1$	$Z_2$	$Q_1$ (MeV)	$Q_2$ (MeV)
		(Highest $K_{\max}$ )		
93	38	37	189.5	189.2
97	39	38	194.0	193.4
98	40	38	196.2	195.7
99	40	39	196.3	195.6

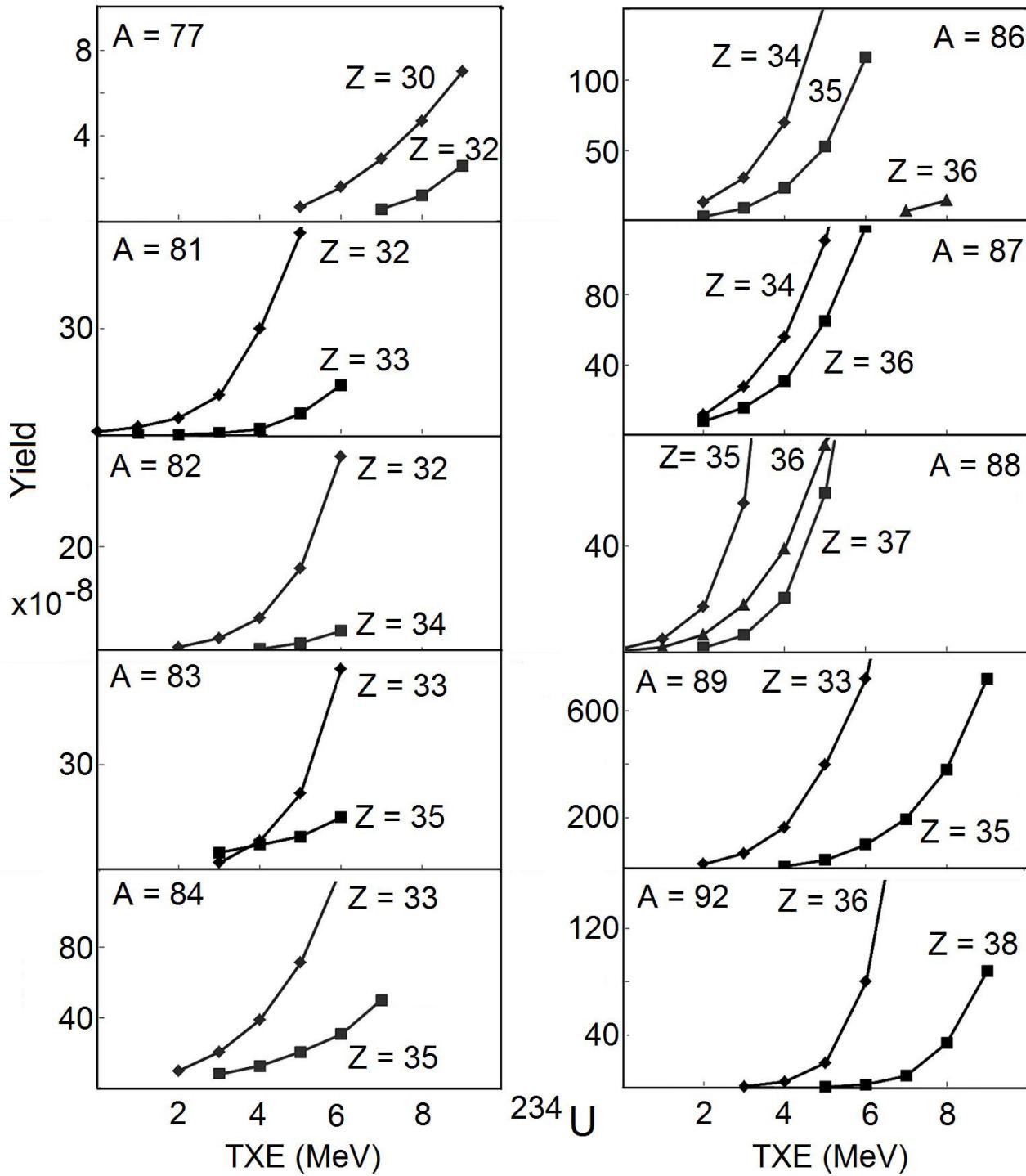


FIGURE 3. Experimental yield of charge, as a function of total excitation energy, from reaction  $^{233}\text{U}(n_{\text{th}},\text{f})$ , as measured by W. Schwab *et al.* [12]. If one compares yields of odd or even charges, respectively, at the lowest excitation energy, the higher yield corresponds to the lower light fragment charge.

maximal total kinetic energy is reached by the charge 36, corresponding to  $Q = 186.6$  MeV, instead of 35 suggested by G. Simon *et al.*, corresponding to  $Q = 182.8$  MeV. Nevertheless, one must notice that in this case the difference in  $Q$ -values corresponding to those charges is 3.8 MeV in favor of charge 36.

#### 4.3. $^{239}\text{Pu}(n_{\text{th}},\text{f})$

The results obtained by G. Simon *et al.* referred to the reaction  $^{239}\text{Pu}(n_{\text{th}},\text{f})$  show that, between isobaric charge splits, the maximal total kinetic energy is reached mostly by the charge that maximizes the corresponding  $Q$ -value. The ex-

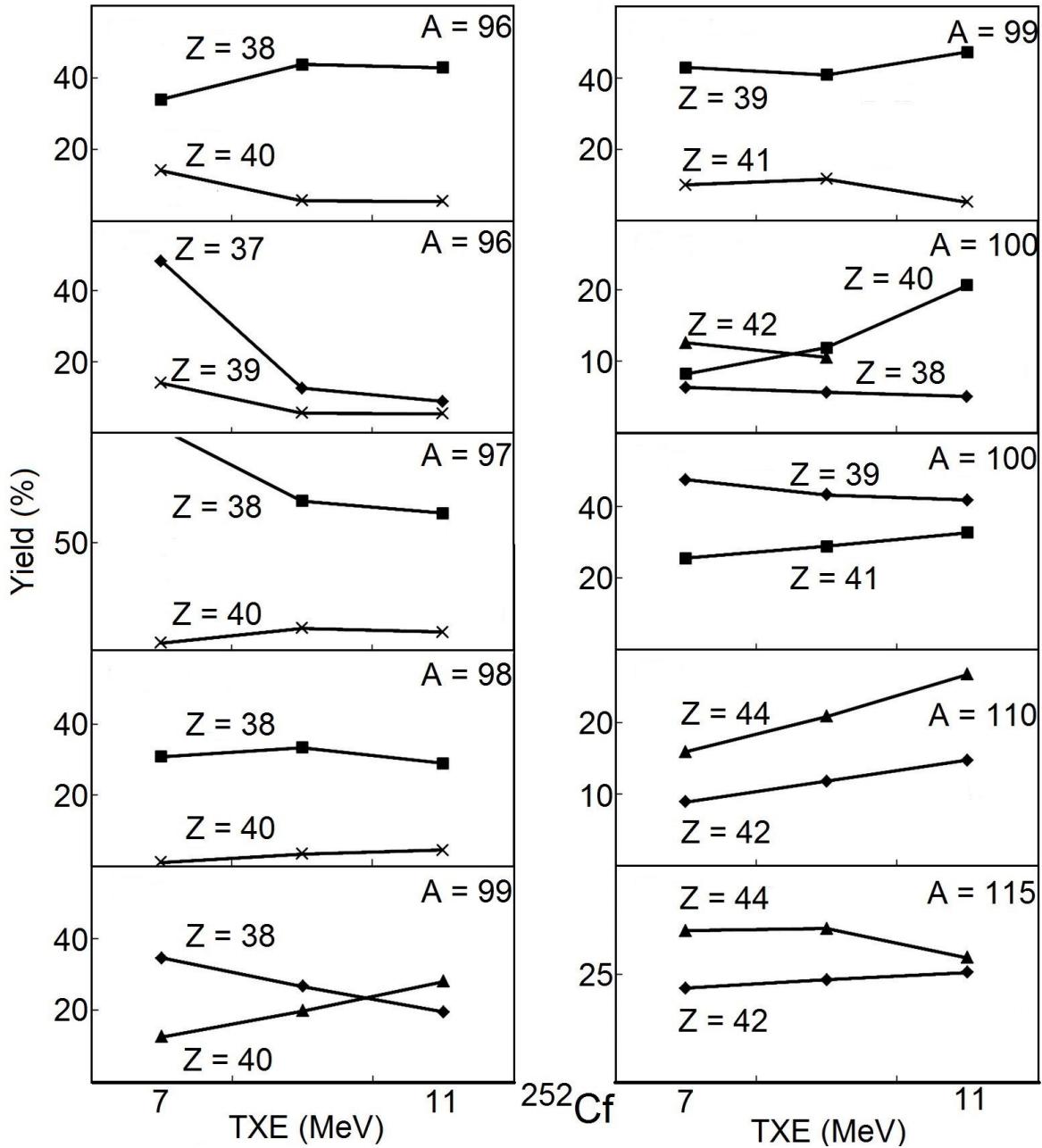


FIGURE 4. Experimental yield of charge, corresponding to total excitation energy values of 7, 9 and 11 MeV, respectively, from reaction  $^{252}\text{Cf(sf)}$ , as measured by F.-J. Hambach *et al.* [16]. If one compares yields of odd or even charges, respectively, at the lowest excitation energy, the higher yield corresponds to the lower light fragment charge. Exception is observed for mass 100 corresponding to transitional nuclei, whose deformabilities depends of neutron number.

ceptions are referred to cases for which the maximal total kinetic energy is reached by a more asymmetrical charge split. Those exceptions are presented in Table III.

#### 4.4. $^{252}\text{Cf(sf)}$

In 1993, F.-J. Hambach *et al.* [16] presented experimental data corresponding to spontaneous fission of  $^{252}\text{Cf}$ . They show the charge and mass yield referred to total excitation energy values equal to 7, 9 and 11 MeV, respectively. See

Fig. 4. One can observe that, in the coldest region, *i.e.* in the lowest excitation energy window, the highest yield corresponds to the lowest light fragment charge, with a few exceptions that we mention below.

The Coulomb effect is more evident in the region associated to the more asymmetric fragmentations. For light fragments heavier than 100 amu, other effects seem to be reflected on charge and mass yield. In the region corresponding to transitional fragments with mass number around 100 and

TABLE III. Cold fission in reaction  $^{239}\text{Pu}(n_{\text{th}},\text{f})$ . Two cases of masses ( $A$ ) for which, as exceptions, the charge that reach the highest total kinetic energy is not  $Z_1$ , corresponding to the highest  $Q$ -value ( $Q_1$ ). For these exceptions the light fragment charge ( $Z_2$ ) that reach the highest kinetic energy is lower than  $Z_1$ . Data taken from Ref. 14. The  $Q$ -values are calculated using the atomic mass from Ref. 9.

$A$	$Z_1$	$Z_2$	$Q_1$ (MeV)	$Q_2$ (MeV)
(Highest $K_{\max}$ )				
91	37	36	195.2	194.6
92	38	37	197.5	194.5

neutron numbers  $N \geq 58$  it is expected that deformabilities will be 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  $Z = 38$  and  $Z = 40$ . For the mass fragmentations 110/142 the yield of  $Z = 44$  ( $N = 66$ ) is higher than the 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

Evidence was shown that, in the coldest region of thermal neutron induced fission of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and spontaneous fission of  $^{252}\text{Cf}$ , respectively, in the asymmetric mass split region ( $A < 100$ ), between isobaric charge fragmentations with similar  $Q$ -values, 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” [4, 6, 7]: 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 then a higher maximal Coulomb interaction energy. The final result of that will be a higher maximal fragment kinetic energy. Even more, for charge splits with different  $Q$ -values, the more asymmetrical charge splits are associated to the lower minimal excitation energy.

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