COULOMB EFFECTS IN COLD FISSION

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ABSTRACT

The structures in the mass and total kinetic energy (TKE) distributions in cold fission of ²³⁴U and ²³⁵U are interpreted in terms of the Coulomb interaction energy (C) between fragments at the scission point. The maximal value of C (Cmax) corresponding to the most compact scission configuration, is calculated for several mass fragmentations. It is shown that Cmax increases, if one increases the charge asymmetry for a given primary fragmentation and Q being constant. This dependence produces oscillations with a period of approximately 5 amu of C as a function of the light fragment mass which are correlated with the observed oscillations of the maximal value of TKE. Moreover, it follows that the yields of the more asymmetric charge fragmentation of the same system are increased, that is

RESUMEN

Se interpretan las estructuras de la distribución de masa y de energía cinética total (TKE), en la fisión fría de ²³⁴U y ²³⁶U, en términos de la energía de interacción coulombiana (C) en la escisión. El valor máximo de C (Cmax) correspondiente a la configuración más compacta, es interpretada para varias fragmentaciones de masa. Se muestra que Cmax aumenta si la asimetría de carga aumenta para una fragmentación primaria manteniendo el mismo valor de Q. Esta dependencia produce oscilaciones, con un período de 5 uma del valor máximo de C en función de la masa del fragmento liviano que están correlacionadas con las oscilaciones observadas en el valor máximo de TKE. Además, resulta que el rendimiento de las fragmentaciones más asimétricas, es decir por las configuraciones más compacta aumenta.

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1. INTRODUCTION

The structure in the mass and charge distributions in low energy fission of heavy nuclei have been interpreted as stemming from shell and pairing effects [1]. In the case of 234 U and 236 U produced in the capture of a thermal neutron by 233 U and 235 U, respectively, those structures are more pronounced at high kinetic energy, $\mathbf{E}_{\mathbf{L}}$, of the light fragments promoted the measurement of charge, mass and kinetic energy in regions with small excitation energy of fragments, that is, as close as possible to the region of cold fission, defined as fission with no neutron emission. In the region of cold fission mostly the systems $^{234}\mathrm{U}$ and $^{236}\mathrm{U}$ were studied. Several authors [2-4] measured with the "Lohengrin" recoil separator the light fragment mass (A_L) , and the light fragment charge (Z_{I.}) distributions in windows of light fragment kinetic energy (EL). A time of flight method has been used by other authors [5,6] to measure the total kinetic energy (TKE) and fragment mass distribution for the above mentioned fissioning systems. From those measurements the maximal value of TKE, TKE_{max} , as a function of A_{L} has been evaluated. A recent review of cold fission is given in Ref. [7].

The purpose of this paper is to disentangle the influence of the Coulomb energy from other effects. An increase of TKE_{max} is observed for increasing charge asymmetry for mass fragmentations with equal Q-values of the same members of the mass chain. The maximal value of the Coulomb interaction energy between the two fragments at scission, C_{max} , will be calculated in a static scission model and compared to the experimental values of TKE_{max} . The analysis allows to understand the yields of the surviving charges in the region of the highest values of the total kinetic energy.

2. THE MOST COMPACT SCISSION CONFIGURATION

At scission the potential energy of a two fragment system (P) is the sum of the total deformation energy (D) and the mutual Coulomb energy (C). The light and heavy fragments could have intrinsic excitation energy (X). In addition, the two fragments could have obtained a prescission total kinetic energy (TKE₀). Using these definitions, the energy balance, neglecting shell and pairing energies, at the scission configuration is given by the following equation:

$$Q = D + X + C + TKE_0, \tag{1}$$

where Q is the available energy for the fission process.

The most compact scission configuration corresponds, by definition, to the highest Coulomb interaction energy. It is assumed that this configuration consists of fragments in their ground states, without prescission kinetic energy. Then equation (1) reduces to:

$$Q = P_{max} = D + C . (2)$$

Note that this configuration corresponds to the maximal value of the potential energy (P_{max}) which cannot be higher than the Q-value. From relation (2) we see that the highest possible value of the mutual Coulomb energy (C_{max}) corresponds to the lowest possible deformation energy (D_{min}). In order to calculate the most compact scission configuration, the deformation energy and the Coulomb interaction energy as a function of the shape of the configuration is needed. The energy C_{max} can be taken as the maximum value of the TKE of the fragment pair. The energy D is transformed into excitation energy of the fragments.

2.1. COULOMB EFFECT

Let Z_L/Z_H and $(Z_L^{-1})/(Z_H^{+1})$ be two charge fragmentations for the same mass ratio A_L/A_H . Assuming that the corresponding scission configurations have the same deformations, the ratio between the corresponding C-values is the following:

$$C_{Z_L}/C_{Z_{L-1}} = Z_L Z_H/(Z_L^{-1})(Z_H^{+1}).$$
 (3)

The Coulomb interaction energy, at the most compact scission configuration, amounts to about 200 MeV. For the difference (${\rm C_{Z_L}}$ - ${\rm C_{Z_{L-1}}}$) a value of about 2 MeV is obtained. In Fig. 1, the curves C, corresponding to ${\rm Z_L}$ and (${\rm Z_{L-1}}$), respectively, are schematically presented as a function of fragment deformation. Let us assume that the deformation energy and the Q-values are the same for the two charge fragmentations. It is easy to see that the smallest value of the deformation energy (${\rm D_{min}}$) and the largest value of the maximal total kinetic energy (${\rm TKE_{max}}$) correspond to the most asymmetric charge fragmentation. The lower of the two light fragment charges with the higher value of the ${\rm TKE_{max}}$ has a higher fission yield. This observation we call the Coulomb effect in the independent yield. A consequence of this yield difference are oscillations in the mass dependence of ${\rm TKE_{max}}$.

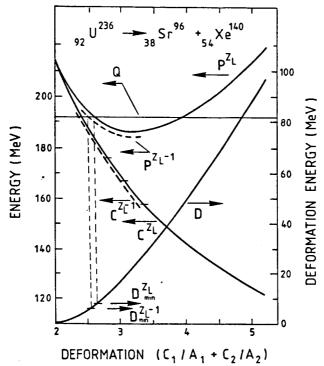


Fig. 1: Total deformation energy (D) and Coulomb interaction energy (C) of the scission configurations limited by the total available energy (Q) for the fragmentation $^{56}Sr/^{140}Xe$ (from Ref. [1]). The charge ratio Z_L/Z_H is equal to 38/54. If one assumes a second charge fragmentation $(Z_L^{-1})/(Z_H^{+1}) = 37/55$ having the same Q-value one will obtain a total kinetic energy TKE^ZL^{-1} higher than the one corresponding to the first fragmentation TKE^ZL .

2.2. Q_VALUE INFLUENCE

The Q-value limits the domain of fragment deformations at the scission configurations, as one can see in the schematic figure 1. Let us take two charge fragmentations corresponding to the Q-values Q' and Q", deformation energies D'_min and D'_min, and the total kinetic energy TKE'_max and TKE''_max, respectively. If Q' < Q", we see from Fig. 1 that D'_min > D''_min. The difference between the TKE_max values will be higher than the difference between the corresponding Q-values. It equals the sum of the differences of the Q-values and the deformation energies.

2.3. SHELL AND PAIRING INFLUENCE

The fragment deformabilities play an important role in the magnitude of the Coulomb effect. For example the TKE_{max} woul be constant if the fragment deformation would not depend on the deformation energy in the

domain of the most compact scission configuration. This means that, if the fragments are soft the difference in the TKE_{max} will vanish.

In order to estimate the role played by the deformation, one can calculate, for each fragment, the deformation energy, using the Strutinsky prescription [8]. The shell [9], δU , and the pairing [10], δP , terms are added to the liquid drop energy [11], W.

The relative variation of the deformation energy of a fragment is calculated by the difference

$$\theta = (\widetilde{W} + \delta U_N + \delta U_Z + \delta P_N + \delta P_Z) - \widetilde{W}_S, \tag{4}$$

where W and W $_S$ are the smoothed values, corresponding to deformed and spherical shapes of fragments, respectively. $\delta U_{N,\,Z}$ and $\delta P_{N,\,Z}$ are the shell and the pairing terms corresponding to neutrons and protons, respectively.

The maximal value of the Coulomb interaction energy, at the scission configuration, is calculated as the difference between the Q-values and the deformation energies of the fragments. As an example in Fig. 2 are

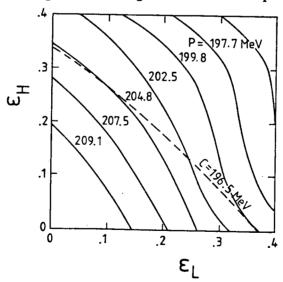


Fig. 2 Equipotential energy (_____) as a function of deformation at scission of the fragmentation $^{92}Sr/^{138}Xe$. The fragment deformations are represented by the Nilsson parameters ϵ . The dashed line is the equi-Coulomb interaction energy corresponding to the highest value permitted by the Q-value = 204.2 MeV. The most compact configuration correspond to $\epsilon_L = 0.4$ for and $\epsilon_L = .0$ for the light and the heavy fragment, respectively.

presented equipotential energy curves (P) as a function of the Nilsson deformation parameters (ϵ) at scission for the fragmentation $^{98}\text{Sr}/^{138}\text{Xe}$. For a Q-value of 199.8 MeV, the maximal value of the Coulomb energy is C_{max} = 196.5 MeV giving a total deformation energy of 3.3 MeV. The corresponding configuration is the heavy fragment in its spherical shape and the light fragment with a deformation ϵ = 0.37. Fig. 3 shows the fragmen-

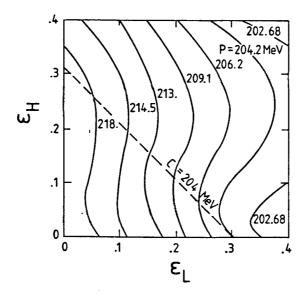
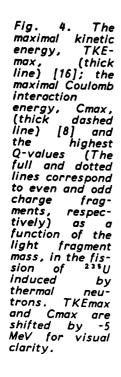


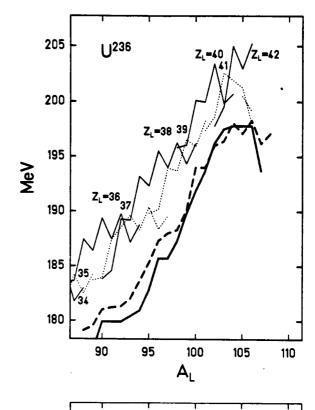
Fig. 3: Same as Fig. 2, but for the fragmentation $^{104}\text{Mo}/^{130}\text{Sn}$. The most compact scission configuration corresponds to the light fragment deformation ϵ_L = 0.3 (ground state) and the heavy fragment deformation ϵ_L = .0 (ground state).

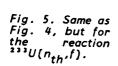
tation 104 Mo/ 130 Sn.Here C_{max} reaches the Q-value because the deformation energy of this fragmentation approaches zero. The C_{max}-values for the two fissioning systems 234 U and 236 U as a function of fragment mass are presented in Figs. 4 and 5.

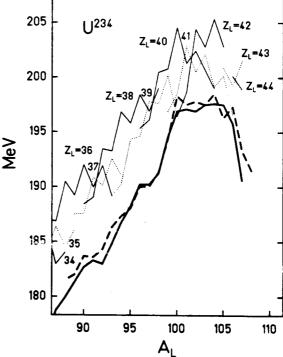
3. INTERPRETATION OF EXPERIMENTAL DATA

3.1. COULOMB EFFECTS









In tables 1 and 2 are shown yield data for the systems 236 U [4], and 234 U [3] for several pairs of isobaric fragmentations (with the same mass ratio) where the Coulomb effect can be observed. For each system, the light fragment kinetic energy ($^{\rm E}$ _L) is fixed. The fragmentations are indicated by the light fragment mass ($^{\rm A}$ _L), the corresponding total kinetic energy (TKE), the light fragment charge, $^{\rm Z}$ _L and $^{\rm Z}$ _L', the Q-values (from Ref. [12]), Q and Q', and the yield, Y and Y', respectively. For comparison the mass range A = 87-97 has been chosen which is not affected by shell effect.

Let us take, for example, the system 236 U. See table 1. Lang et al. [4] have measured the charge and the mass distribution for E_L equal to 108 MeV. This energy window corresponds to the TKE-line 10 MeV lower on the average than the maximal Q-line. Let us take the mass fragmenta-

Table 1: Reaction $^{235}\text{U(n}_{\text{th}}$, f). The yields Y and Y' of two charges Z_L and Z_L corresponding to very close fragment kinetic energies were fixed at E_L = 108 MeV. Q-values (Ref. [12]) Q and Q', respectively. (Taken from Ref. [4]).

$^{\mathtt{A}}_{\mathtt{L}}$	z_{L}/z_{H}	N_L/N_H	Z_{L}/Z_{H}	N_L/N_H	Q	Q'	Y(%)	Y'(%)
					(MeV)	(MeV)		
106*	43/49	63/81	44/48	62/82	199.2	199.2	5.7±3.2	0.
103		63/81		61/83		199.6		24.7±3.
102	39/53	53/81		61/83			39.0±2.4	4.9±1.
101	39/53	62/82		60/84		197.4		21.7±2.
100	41/51	59/85		60/84		189.7		0.7±0.
100	38/44	62/82		61/83		195.9		8.2±1.
99	39/53	60/84	40/52	59/85			45.0±3.1	47.2±3.
98		60/84		58/86			33.0±5.5	21.8±2.
97	38/54			58/86			54.8±3.3	39.1±3.
96	39/53			56/88			12.5±1.5	1.4±0.9
95	37/55			56/88			15.8±1.5	10:5±1.
93	37/55			55/89			64.2±3.7	21.8±3.
92	36/56			54/90			53.4±3.1	6.3±1.9
91	36/56			54/90			75.9±3.3	19.7±3.
90	37/55			54/90			4.4±2.6	.0
89	35/57		37/55				29.3±2.3	3.5±2.
88	34/48		35/57				10.4±1.4	50.7±2.
87	35/47			51/93			45.6±3.0	10.1±2.0
85	34/48	51/93	35/57	50/94	182.9	182.3	90.0±2.6	3.3±2.

^(*) For A_L = 106 the light fragment kinetic energy is taken equal to 98.9 MeV

Table 2: Reaction 233 U(n_{th}, f). The yield Y and Y' of two charges Z_L and Z_L' corresponding to very close Q-values (Ref. [12]) Q and Q', respectively. The light fragment kinetic energies were fixed at E_L = 110.55 MeV. (Taken from Ref. [3]).

$^{\mathtt{A}}_{\mathtt{L}}$	$z_{L}/z_{H} N_{L}/N_{H}$	$z_{L}/z_{H} N_{L}/N_{H}$	Q	Q'	Y(%)	Y'(%)
			(MeV)	(MeV)		
105 104 103 103 101 100	41/51 64/78 40/52 64/78 40/54 63/79 41/51 62/80 40/52 61/81 39/53 61/81	43/49 62/80 41/51 63 79 43/49 60/82 42/50 61/81 41/51 60/82 42/50 58/84	199.0 198.5 202.1 201.3	199.2 199.1 202.8 202.8	37.3±23. 5.4±2.6 16.0±6.2 47.7±10. 61.8±2.4 4.0±1.4	6.4±1.3 20.6±7.4 1.9±0.7 34.4±6.7 31.7±1.9 0.2±0.0
97 96 93 92 92 91 90 89 87 86 86 85 85	38/54 59/83 39/53 57/85 37/55 56/86 36/56 56/86 37/55 55/87 36/56 55/87 37/55 53/89 35/57 52/90 34/58 52/90 35/57 51/91 34/58 51/91 32/60 53/89	40/52 57/85 40/52 56/86 38/54 55/87 38/54 54/88 38/54 54/88 37/55 54/88 38/54 52/90 37/55 52/90 36/56 50/92 36/56 50/92 35/57 50/92 37/55 58/94	194.6 192.5 191.9 190.1 190.0 187.6 186.1 186.4 186.3 184.2	195.3 193.3 193.5 193.5 190.9 188.4 185.1 186.9 187.1 187.1	24.3±2.0 23.8±1.7 46.0±4.1 20.7±2.6 67.5±3.0 50.0±2.7 13.7±1.0 5.8±3.0 54.4±2.5 53.2±3.3 77.2±4.9 76.7±3.2 1.3±0.2	10.1±1.5 3.6±0.8 47.3±6.1 12.7±1.1 12.7±1.1 46.4±4.0 1.1±0.3 2.8±0.2 33.1±2.0 7.1±1.1 7.1±1.1 18.5±1.3

tion 91/135. The light fragment charges Z_L = 36 and 37 have the same Q-value (=187.5 MeV) but the charge Z_L = 36 has a yield (Y = 75.9 ±3.3) higher than Z_L = 37 (Y' = 19.7 ± 3.1). Trochon et al. [13] identified , in the case of 236 U, the surviving charge for the highest values of TKE. The charge corresponds, in most cases to the maximal Q-value. But the surviving charge, for the fragmentation A_L/A_L = 91/145 was again found to be Z_L/Z_L = 36/54 in contrast to the hypothesis of highest Q-value which correspond to the charge fragmentation 37/55.

For A_L = 93, the highest Q-value corresponds to Z_L = 38 (Q = 189.2 MeV). The Coulomb effect shifts for the most compact configuration the charges downward to Z_L = 37 (Q=189.6 MeV) as has been seen in the experimental data obtained by Lang et al. [4]. The charge Z_L = 37 has a higher yield (64.2 % ± 3.7) than for Z_L = 38 (21.8 % ± 3.5).

The mentioned experimental results exhibit Coulomb effects in the yields for fragmentations corresponding to Q-values other than the highest ones. For example, for A_L = 95, the maximal Q-value (192.3 MeV) cor-

respond to Z_L = 38. Nevertheless, one can compare two fragmentations in a pair corresponding to lower Q-values. For instance, the charge Z_L = 37, the corresponding Q-value which is 190.7 MeV, has a yield (Y = 15.8 % ± 1.5) higher than Z_L = 39 (Y' = 10.5 % ± 1.5) despite of their very close Q-value.

Let us now draw the consequences of the Coulomb effect on the high TKE-lines presented in Figs. 4 and 5 for the systems 236 U and 234 U, respectively. In both cases, shoulders in the TKE-lines are observed [14]. These shoulders could be interpreted by the Coulomb effect as follows.

Let us take, for example, the TKE-lines of 236 U, presented in Fig. 4. Assuming that the most compact scission configurations correspond to the maximal Q-value the oscillations in the TKE-lines can not be explained. In Sect. 2.1 we have learnt when two charges Z_L correspond to very close Q-values, for high TKE-values the Coulomb effect favours the yield of the lowest Z_L . This assumption allows to interprete the mentioned shoulders.

The TKE-lines with a slope of 2.5 MeV/amu have a very pronounced shoulder for the region from A_L = 93 - 90. This shoulder can be interpreted as follows: From A_L = 96 to 94 the maximal Q-values are very high relative to the corresponding second Q-values. Then , the charge corresponding to that maximal Q-values (Z_L = 38) will survive up to TKE_{max}. For A_L = 93, the charge Z_L = 37 has the Q-value = 189.63 MeV which is the maximal and higher than that for Z_L = 38 (Q = 189.18). Then the charge favoured by the Coulomb effect will be the lower one (Z_L = 37). The change of Z_L from 38 to 37 for the change of Z_L from 94 to 93 will produce an increase of TKE relative to the extrapolation from the region Z_L = 96 - 94.

For A_L = 92, the charge Z_L = 36 will survive up to TKE_{max} , because it is in competition with a higher Z_L (= 38). Then the change of Z_L from 37 to 36, for the transition from A_L = 93 to 92 will produce ,as before, a relative increase of TKE, making the mentioned shoulder more pronounced.

Let us take the system 234 U. See Fig. 5. The TKE-value corresponding to A_L = 91 is higher than the extrapolation from A_L > 91. The charge decreases from Z_L = 38 (A_L = 92) to Z_L = 37 (A_L = 91). Moreover, from A_L = 91 to 90, Z_L decreases from 37 to 36 producing, by the Coulomb effect the pronounced shoulder in this mass region.

3.2. INFLUENCE OF THE Q-VALUE

Let us consider the 234 U system. In order to separate the Q-value influence from the other effects one takes the fragmentations 104 Mo/ 130 Sn and 106 Mo/ 128 Sn, the corresponding Q-values of which are 205.3 MeV and 204.7 MeV. From the energy balance unique charges were attributed for those fragmentations [5,6]. In Fig. 5 one can see that the TKE_{max}-value corresponding to the mass fragmentation 104/130 is higher than the TKE_{max}-value corresponding to the mass fragmentation 106/128.

Let us consider the fragmentations 96 Sr/ 138 Xe and 94 Sr/ 140 Xe, Ceir Q-values being 198.4 MeV and 196.8 MeV, respectively. One has chosen those fragmentations because the Q-value of their neighbouring charge splits are relatively low making reasonable the hypothesis of pure charge fragmentations. One can observe that the difference between the TKE max values is higher than the difference between the corresponding Q-values, as expected and as explained by the influence of the Q-value (c.f. subsection 2.3).

Let us take the system 236 U. For A_L = 90 the maximal Q-value (189.4 MeV), corresponding to Z_L = 36 is much higher than the second Q-value (183.9 MeV) corresponding to Z_L = 37. Then, the surviving charge will be Z_L = 36. From this reasoning and from the discussion in subsection 3.1, one can say that for A_L = 92 -90 the charge fragmentation will be 36/56. The Q-value corresponding to A_L = 90 (Q = 189.4 MeV) is very close to the Q-value corresponding to A_L = 92 (Q = 189.8 MeV). This fact is the reason why the corresponding TKE_{max} -values are also very close. In this case the influence of the Q-value will enlarge the shoulder in the TKE-lines, which begins at A_L = 93 as it is observed in the Fig. 4 and has been mentioned in the Sect. 3.1.

3.3. SHELL AND PAIRING EFFECTS

Experimental results of fragment yields in the mass range A =98-106 cannot be explained by the Coulomb effect. The reason for that is the softness of the fragment which diminishes the Coulomb effect. Moreover, according to equation (3), the Coulomb effect decreases with increasing $\mathbf{Z_L}$, and the indicated mass range corresponds to the highest observed $\mathbf{Z_L}$ values.

Moreover, in the system 234 U at E_L = 110.55 MeV presented in table 2 the light fragment 104 Nb (Q = 199.2 MeV) is favoured as comparated to

 104 Zr (Q = 199.0 MeV). This result could be interpreted as a product of a nucleon pair breaking in the case of even-even fragments.

In subsection 2.3 has been presented the calculation of the maximal value of the Coulomb interaction energy at scission. For the fragmentation $^{104}\mathrm{Mo/^{130}}\mathrm{Sn}$ a TKE_max very close to the corresponding Q-value was found in agreement with the calculated $\mathrm{C_{max}}$ (see Fig. 4). The heavy fragments, neighbours of the doubly closed spherical shell nucleus $^{132}\mathrm{Sn}$ and the light fragments in the region $\mathrm{Z_L}$ = 40-42 and $\mathrm{N_L}$ = 60-64 corresponding to deformed nuclei [15,16] constitute the fragmentation with the lowest excitation energy. The deformation properties, caused by shell and pairing effects, could be also responsible for the structures in the TKE-lines. The $\mathrm{C_{max}}$ calculated by the method explained in subsection 2.2. for the cases $^{234}\mathrm{U}$ and $^{236}\mathrm{U}$ are in agreement with the experimental TKE-lines, see Figs. 4 and 5.

4. DISCUSSION

In order to observe the Coulomb effect on the charge yield one has to separate out other effects. One has to chose pairs of fragmentations having very close Q-values and negligible shell effects. One also has to take pairs of fragmentations with the same number of broken nucleon pairs. In order to exclude all the mentioned effects one can take the average charge as a function of light fragment mass for increasing values of the kinetic energy. Lang et al. [4] have obtained the deviation, ΔZ , of the average nuclear charge Z_L from the unchanged charge density value Z_{UCD} . Results corrected for neutron emission show that in the mass region A_L = 85 -98 Z_L decreases as a function of the light fragment kinetic energy as expected from the Coulomb effect.

In order to give more proofs for the Coulomb effect one can take the example of A_L = 92 and the charges Z_L = 36 and Z_L = 38. The liquid drop energy of spheroidal configurations and the Coulomb interaction energy are calculated and no shell effects in the fragment energies are taken into account. The distance between the fragment's tips is taken equal to 2.0 fm.

For the charge Z_L = 36 (Q = 189.8 MeV) the most compact configuration fulfilling equation (2) corresponds to a fragment deformation β_L = 0.40 and β_H = 0.30, respectively. For the charge Z_L = 38 (Q = 189.2 MeV) the most compact configuration corresponds to β_L = 0.35 and β_H = 0.50, respectively. The maximal Coulomb energy corresponding to Z_L = 36 (C_{max} = 180.6 MeV) is higher than Z_L = 38 (C_{max} = 176.8 MeV) despite of its lower Q-value. This result shows that Coulomb effects could explain

the reverse trend of the maximal kinetic energy with respect to the Q-values.

5. CONCLUSION

The experimental results on charge, mass and kinetic energy distribution of the systems $^{234}\mathrm{U}$ and $^{236}\mathrm{U}$ have been analysed. Our consideration leads to the conclusion that that the Coulomb interaction energy as a function the fragment deformation as well as the total fragment energy as a function of deformation influence the scission configuration with a similar importance as the role played by the Q-value. The oscillations in the TKE $_{max}$ lines as a function of the light fragment mass can be interpreted by the Coulomb effect, the influence of the Q-value and by shell effects. The Coulomb effect is higher for the most asymetric fragmentations and it is negligible for the soft fragments $\mathrm{A_{L}} \approx 100$.

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