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Nucleon pair breaking in the thermal neutron induced fission of $^{233}\text{U}$ and $^{235}\text{U}$

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Résumé. — L'absence d'effet pair-impair systématique, récemment observé dans la distribution des masses des fragments de fission primaires, est discutée en termes de rupture de paires de nucléons entre le point selle et le point de scission. Il est conclu que les données expérimentales existantes sont compatibles avec le déroulement d'un processus où est brisée au moins une paire de nucléons.

Abstract. — The absence of a systematic even-odd effect in the primary fragment mass distributions, which has been recently observed, is discussed in terms of nucleon pair breaking during the saddle to scission transition. It is concluded that the existing experimental data are compatible with the assumption of a process where at least one nucleon pair is broken.

It is well known that thermal induced fission of $^{233}\text{U}$ and $^{235}\text{U}$ proceeds through a very small number of exit channels, which, according to A. Bohr [1], are associated with transition states of the fissioning nucleus at the saddle point. The available energy of the fissioning nucleus at saddle point lies below the expected value of the pairing gap [2]. Therefore the contribution of channels lying above this gap is hindered. The dominant channels are expected to be collective states built on the fundamental state of the even-even nucleus $^{234}\text{U}$ or $^{236}\text{U}$. In other words, when the fissioning nucleus starts its descent towards scission, it would be in a superfluid state where both neutrons and protons always come in pairs. Consequently, the problem of viscosity of nuclear matter (quasi particle excitation) during the saddle to scission transition could be approached through the evaluation of the number of nucleon pairs broken at scission, reflected in the even-odd effects on the primary fragment mass and charge distributions.

The problem of odd-even effects in fission has been very much studied until today. Odd-even effects on total charge distribution [3-6] and on average fragments kinetic energy [4, 5, 7] were found. These effects were related to the number of broken proton pairs [4, 7-13]. The notation utilized in this paper is inspired from the above indicated references.

In recent publications [14, 15], it has been shown that, for events with high kinetic fragment energy, fragmentation into two odd primary masses are just as probable as fragmentation into two even primary masses. Even more, the maximal kinetic energy, as a function of the mass ratio, does not exhibit any even-odd effect. These experimental results were interpreted as a proof that, between the saddle and the scission points, at least one pair of nucleons must be broken whatever the particular mass fragmentation.

However, Nifenecker et al. [12] having reanalysed the same experimental results, came to the conclusion that very late nucleon pair breaking is actually the predominant mechanism.

In this paper, we shall therefore argue and show that first, Nifenecker et al. have misinterpreted the experimental data taken from references [13] and [14], and second, that the hypothesis of early breaking of at least one pair is compatible with all verifiable experimental facts gathered so far.

The relative importance of an « even-odd-effect » on the mass distributions of the fission fragments can best be assessed by means of a parameter, $\delta A$, defined by:

$$\delta A = \frac{AY_e - AY_o}{AY_e + AY_o}$$  \(1\)

where $AY_e$ is a pure number representing the sum of all the even-mass fragment-yields, and $AY_o$ the sum

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of all the odd-mass fragment yields. The superscript $^{A}$ merely indicates that we are dealing with fragment masses only and that the evenness or oddness of the associated neutron and proton numbers, are not considered here.

The definition (1) is a generalization of the odd-even effects on protons and neutrons

$$\delta Z = \frac{Z_e - Z_o}{Z_e + Z_o} \quad \text{and} \quad \delta N = \frac{N_e - N_o}{N_e + N_o}. \quad (2)$$

As detailed in appendix A, one can then deduce that the ratio $P_{oo}$ of odd $Z$, odd $N$ fragments to the total number of fragments can be written as:

$$P_{oo} = \frac{1}{4} [1 + \delta A - \delta N - \delta Z]. \quad (3)$$

Data published in references [14] and [15] refer only to fissions resulting in fragments with high total kinetic energies, $TKE$. We use $K_{EL} = TKE - KEH$ as a parameter, where $K_{EL}$ and $KEH$ are the light and heavy fragment kinetic energies respectively. For the case of $K_{EL} = 115$ MeV, a simple energy balance between the entrance and exit channels then shows that there simply remains not enough excitation energy for two nucleon pairs to be broken. This, in turn, means that one cannot have odd-odd mass fragments and hence

$$P_{oo} = 0. \quad (4)$$

But, one also knows from our experimental data [14, 15] that

$$\delta A = 0 \quad (5)$$

since the number of experimentally observed even and odd fragment masses is indeed equal.

In their interpretation of these results, Nifenecker et al. now assume $\delta N = 0$ a non justified ansatz which, we believe, subsequently leads them to the wrong conclusions. Combining equations (3), (4) and (5) one obtains

$$\delta Z + \delta N = 1. \quad (6a)$$

Nifenecker et al. have deduced in a combinatorial analysis [13] the relation

$$\delta Z_{1/n} + \delta N_{1/n} = 1 + \delta A_{1/n} \quad (6b)$$

where $n$ is the maximum number of pairs broken. In any case the interpretation $\delta N = 0$ is not justified.

Let us now consider somewhat lower kinetic energies: $KE_{L} = 112$ MeV. It was shown by Quade et al. [16] that the fission of $^{233}$U then leads to $\delta Z = 0.46$ and $\delta N = 0.11$, whereas for the same reaction at the same $KE_{L}$, we [14] obtained $\delta A = 0$. One notes here that at this energy more than one pair can be broken. Moreover, application of equation (3) gives $P_{oo} = 0.11$, which means that, in 11% of all cases, one has broken at least one neutron and one proton pair. If one assumes that the probability to break a neutron or proton pair is proportional to the number of neutrons or protons present, and if one evaluates the probability for precisely one proton and one neutron pair to be broken, one finds that $P_{oo} = 0.12$, a value which is in good agreement with the experimental result.

The above-mentioned $\delta A = 0$ experimental results have recently been qualitatively confirmed by Quade [17] who found that odd-odd fragment masses were as numerous as even-even ones.

It thus appears as if, in a nucleus undergoing fission, the excitation energy available between the saddle and scission points, is primarily and essentially dissipated in a nucleon pair-breaking mechanism. If, for instance, the excitation energy is high enough to break two nucleon pairs, then two pairs would be broken. It will be shown that the hypothesis that at least one pair of nucleons is broken is not in contradiction with the following set of experimental data:

1. the even-odd charge effect, $\delta Z$,
2. the even-odd effect on the average kinetic energy, $\delta E_{k}$,
3. the structure observed on fragment yield versus atomic mass $A$ plots, obtained in several experiments.

**1. The observed $\delta Z$ effect.**

It was shown above that $\delta Z \neq 0$ is to be expected for high kinetic energies and one can show that this remains true even for cases where more than one nucleon pair can be broken.

Let $Q_{kl}$ be the probability that a fission event will result in $k$ broken proton pairs and $l$ broken neutron pairs. One can then write these probabilities in an « easy to remember » matrix form namely:

$$Q = \begin{bmatrix}
Q_{00} & Q_{01} & \cdots & Q_{0l} \\
Q_{10} & Q_{11} & \cdots & Q_{1l} \\
\vdots & \vdots & \ddots & \vdots \\
Q_{k0} & \ldots & \ldots & Q_{kl}
\end{bmatrix}.$$ 

We define $p$ = probability that the broken pair nucleons end up in different fragments [12]. If pair-breaking occurs well before scission, then the separated nucleons, moving independently, can end up in any of the fragments, leading to $p = \frac{1}{2}$. But if pair-breaking occurs at scission, then the separated nucleons must end in different fragments, leading to $p = 1$.

The $\delta Z$, $\delta N$ and $\delta A$ values depend of kinetic energy and mass region. The associated $p$ and $Q$ values will be referred to the same region where those odd-even effects are measured.

As shown in equation (B.4), of appendix B, one obtains for $p = \frac{1}{2}$:

$$\delta Z = \sum_{l} Q_{0l},$$
$$\delta N = \sum_{l} Q_{k0},$$
$$\delta A = Q_{00}.$$ 

Clearly, any $Q_{0l} \neq 0$ value, which represents the probability of breaking any number, $l$, of neutron pairs but no proton pair, will result in an even-odd
fragment charge effect $\delta Z$. Also, any $Q_{0i} \neq 0$ value, will result in an even-odd effect on the neutron number $\delta N$.

2. The even-odd effect $\delta \overline{E}$ on the average kinetic energy.

Let us define by $\overline{E}$ the average kinetic energy of the fragments in the case of no nucleon pairs broken at all. One can then show (appendix, Eq. (B.8)) that:

$$\delta \overline{E} = \frac{2 \Delta E}{1 - \left( \sum_i Q_{0i} \right)^2} \times \left[ \sum_i Q_{0i} \sum_{l}(k + l) Q_{kl} - \sum_l lQ_{0l} \right]$$

where $\Delta E$ represents the energy required to break a single nucleon pair, to be subtracted from $\overline{E}$. The above formula is equivalent to the corresponding more general case deduced by Nifenecker et al. [13].

Consider, for example, the case represented by the following $Q$ matrix:

$$Q = \frac{1}{10} \begin{bmatrix} 0 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 \\ 0 & 1 & 1 & 1 \\ 0 & 0 & 0 & 0 \end{bmatrix}$$

Here one has $Q_{00} = 0$, which means that the probability that no nucleon pair be broken is zero. This $Q$ matrix leads to the following results:

$$\delta \overline{E} = 1.3 \Delta E$$
$$\delta Z = 0.3$$
$$\delta N = 0.1$$
$$\delta A = 0.$$  

One notices that, even when at least one pair is broken, observable even-odd effects on the average kinetic energy $\overline{E}$, the proton number and the neutron number ought to exist.


The earliest measurements of the mass yield of the fission fragments, made at very high kinetic energy $KE_L$, show fluctuations with an average recurrence rate of roughly 5 a.m.u. [18]. These experimental data were then interpreted by some authors as being proof of the predominance of the even-even over the odd mass fragment yields, and this in spite of the fact that the experimental fragment-mass resolution was not capable of separating adjacent fragment masses. Our experimental results [14], however, show that no even-odd mass effects can be detected if the experimental mass resolution is capable of separating 1 a.m.u. at high $TKE$ values, even though the even-odd effect is predicted by the viscous theory to be maximum at such high $TKE$. However, certain mass-yield fluctuations can be induced by either lowering the $KE_L$ window and (or) diminishing artificially the mass reso-

Fig. 1. — Fission fragment mass yield in the $^{233}\text{U}(n, f)$ reaction for different windows set on the light fragment kinetic energy $KE_L$

![Graph 1](image1)

Fig. 2. — Same as figure 1 for the $^{235}\text{U}(n, f)$ reaction.

![Graph 2](image2)
values. Within the framework of such a model, the scission configuration consists of two ellipsoidal fission fragments separated by a distance, $d$, and has a total potential energy $PE$ defined by

$$ PE = DE_L + DE_H + CE $$

(7)

where $DE_L$ and $DE_H$ represent the deformation energy of the light and heavy fragments respectively and $CE$ the Coulomb energy between the two fragments. Each of the terms of equation (7) is then evaluated as a function of the deformation of the fragments.

According to Strutinsky [19], one can calculate the deformation energies, $DE_L$ and $DE_H$, by adding a shell term [20] and a pairing term [22] to the basic liquid drop [21] deformation energy. The Coulomb energy, $CE$, is obtained by numerical integration, the inter-fragment distance, $d$, being fixed at 2 fm. This $d = 2$ fm value can be deduced from the experimental fact that, for the fragment with $A = 102$ and $Z = 42$, the highest measured kinetic energy, $TKE$, is equal to the $Q$ value of the reaction [23].

For each light fragment mass and charge, one can then obtain certain fragment deformations corresponding to a maximum of the associated Coulomb energy $CE$, with the restraint $EP < Q$. These specific fragment deformations are referred to as « most compact configurations » and we assume that they are to be associated with the highest possible $TKE$ values. A comparison of our calculated $CE_{\text{max}}$ values with the maximum value of the observed total kinetic energy, $TKE_{\text{max}}$, as a function of the light fragment mass, $A_L$, is shown in figures 3 and 4 respectively. For both cases, the $CE_{\text{max}}$ and $TKE_{\text{max}}$ results are rather similar.

Certain fluctuations in the measured $TKE_{\text{max}}$ data, separated by approximately 5 a.m.u., can be discerned in the figures. The figures also show $TKE$ as a function of $A_L$ for a fixed $KE_L$ value, represented in both cases by a nearly straight line. It should be emphasized here that it is precisely the fluctuation in the difference between these calculated straight line results and the observed corresponding $TKE_{\text{max}}$ values as a function of $A_L$, which is responsible for the so-called experimentally « observed » even-odd fragment mass fluctuations, mentioned above.

One also notices a pronounced even-odd mass effect in the calculated $CE_{\text{max}}$ values for $A_L > 100$ a.m.u., shown in figures 3 and 4. These fluctuations are generated in the calculation itself because we suppose that all fragments are in their respective ground states which implies that no nucleon pairs can be broken. The corresponding experimental $TKE_{\text{max}}$ data, also shown in these figures, do of course not exhibit such fluctuations since we have shown that in all cases at least one nucleon pair must be broken. Such $CE_{\text{max}}$ structure is also present for $A_L \leq 100$ a.m.u., but cannot be discerned due to the steepness of the slope.

4. Conclusion.

We have shown in the above, that the experimental information is compatible with a process in which at least one pair of nucleons is broken.

Most of the previous interpretations of the observed even-odd effects which require the contribution of a non broken pairs component, are likely to lead to unreliable conclusions. Indeed, most if not all of these analyses were performed by considering any of the even-odd effects, $\delta A$, $\delta Z$, $\delta N$ and $\delta E$ separately and without caring for a possible interdependence, which we have shown to exist. In some case [12], it was also assumed that $\delta A = 0$ implies $\delta N = 0$, a proposition which is not right in the high kinetic energy region. The supposition that only a single nucleon pair is broken could be acceptable in that region but not in the lower ones.

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Appendix A.

Let \( Y_{pn} \) be the sum of the partial fragment yields containing \( p \) protons and \( n \) neutrons, where \( p \) and \( n \) can only take on two possible values namely \( e = \) even or \( o = \) odd. From equations (1) and (2) one thus obtains:

\[
\begin{align*}
\delta A &= \frac{4 Y_e - 4 Y_o}{4 Y_e + 4 Y_o} = \frac{(Y_{ee} + Y_{eo}) - (Y_{oe} + Y_{oo})}{Y_{ee} + Y_{eo} + Y_{oe} + Y_{oo}}, \\
\delta Z &= \frac{2 Y_e - 2 Y_o}{2 Y_e + 2 Y_o} = \frac{(Y_{ee} + Y_{eo}) - (Y_{oe} + Y_{oo})}{Y_{ee} + Y_{eo} + Y_{oe} + Y_{oo}}, \\
\delta N &= \frac{N Y_e - N Y_o}{N Y_e + N Y_o} = \frac{(Y_{ee} + Y_{eo}) - (Y_{oe} + Y_{oo})}{Y_{ee} + Y_{eo} + Y_{oe} + Y_{oo}}.
\end{align*}
\]

(A.1)

It can easily be shown that:

\[
1 + \delta A - \delta Z - \delta N = \frac{4 Y_{oo}}{Y_{ee} + Y_{eo} + Y_{oe} + Y_{oo}} \quad \text{(A.2)}
\]

and it follows that the ratio of odd \( Z \)-odd \( N \) fragments to the total fragment number can be written as:

\[
P_{oo} = \frac{Y_{oo}}{(Y_{ee} + Y_{eo} + Y_{oe} + Y_{oo})} = \frac{1}{4} (1 + \delta A - \delta Z - \delta N). \quad \text{(A.3)}
\]

Note: The above \( Y_{pn} \) notation is only used to demonstrate the validity of equation (3) in the text and appears nowhere else in this paper.

Appendix B.

In the text, \( ^4Y_e \) is defined as the sum of the even fragment yields when considering the mass \( (A) \) of the fragments in question. However, it turns out that a somewhat different terminology is much easier to handle.

Let us consider fragmentation processes in which exactly \( m \) nucleon pairs are broken. Then one could define the sum of the even fragment yields as \( ^4Y_e \) and the corresponding odd fragment yields as \( ^4Y_o \). Suppose now that one breaks the \((m + 1)\)-th nucleon pair. Then one can write the yields as:

\[
\begin{align*}
^4Y_e^{(m+1)} &= ^4Y_e^m (1 - p) + ^4Y_o^m p, \\
^4Y_o^{(m+1)} &= ^4Y_o^m p + ^4Y_e^m (1 - p),
\end{align*}
\]

where \( p \) is the probability that the two nucleons of the \((m + 1)\)-th broken pair end up in different fragments.

The even-odd effect can then be related as follows:

\[
\delta A^{(m+1)} = \frac{^4Y_e^{(m+1)} - ^4Y_o^{(m+1)}}{^4Y_e^{(m+1)} + ^4Y_o^{(m+1)}} = (1 - 2p)\frac{^4Y_e^m - ^4Y_o^m}{^4Y_e^m + ^4Y_o^m}
\]

which leads to the recurrence relation:

\[
\delta A^{(m+1)} = \delta A^{(m)} (1 - 2p).
\]

If one assumes that no nucleon pair is broken \((m = 0)\) in an even \( N \), even \( Z \) fissioning system, then one cannot obtain odd fragments and one gets

\[
\delta A^{(0)} = 1
\]

it follows that

\[
\delta A^{(m)} = (1 - 2p)^m. \quad \text{(B.1)}
\]

The global even-odd effect can then be written in the form

\[
\delta A = \sum_m (\frac{\delta A^{(m)} Y_e^m}{Y_o^m} + \frac{\delta A^{(m)} Y_o^m}{Y_e^m}) = \sum_m \sum_m (\frac{\delta A^{(m)} Y_e^m}{Y_o^m} + \frac{\delta A^{(m)} Y_o^m}{Y_e^m})
\]

which can be rewritten as:

\[
\delta A = \sum_m Q_m \delta A^m \quad \text{(B.2)}
\]

where \( Q_m = \frac{^4Y_e^m + ^4Y_o^m}{\sum_m (^4Y_e^m + ^4Y_o^m)} \) represents the probability for breaking exactly \( m \) nucleon pairs. An equivalent set of relations can be deduced for the even-odd effects in neutron number \( \Delta N \) and proton number \( \Delta Z \).

Let us now become more specific and write \( Q_m \) instead of \( Q_{kl} \) where \( k \) and \( l \) are the number of broken proton and neutron pairs respectively. With this new notation, the preceding relation can be rewritten in the form

\[
\delta A = \sum_{kl} Q_{kl} (1 - 2p)^{k+l} \quad \text{(B.3)}
\]

and similarly

\[
\delta Z = \sum_{kl} Q_{kl} (1 - 2p)^k \quad \text{and} \quad \delta N = \sum_{kl} Q_{kl} (1 - 2p)^l
\]

which for \( p = 1/2 \) reduces to

\[
\delta A = Q_{00}
\]

and

\[
\delta Z = \sum_k Q_{0k} \quad \text{and} \quad \delta N = \sum_k Q_{00}
\]

Let the average kinetic energy of the fragments in which no nucleon pair has been broken, be given by \( \bar{E} \), and let us suppose that it takes, on the average, an energy \( \Delta E \) to break any given nucleon pair. If, for instance, no proton-pair but only \( l \) neutron pairs have been broken in the fission process, one will then obtain
even Z fragments, with a probability $Q_{0l}$ and with a total kinetic energy of $T KE_{0l} = \bar{E} - l \Delta E$.

If one has a configuration in which at least one proton pair has been broken ($k \neq 0$) and if we assume $p = 1/2$, then one can obtain either an even or an odd Z fragment, with probabilities $(Q_{kl})^1/2$ and $T KE_{kl} = \bar{E} - (k + l) \Delta E$. It follows that the even Z fragments will have an average kinetic energy given by

$$\bar{E}_e = \bar{E} - \Delta E \left[ \frac{\sum_i Q_{0i} + \sum_{k \neq 0} \frac{(k + l) Q_{kl}}{2}}{\sum_i Q_{0i} + \sum_{k \neq 0} Q_{kl}} \right]. \quad (B.5)$$

The corresponding average kinetic energy for the odd Z fragments will be

$$\bar{E}_o = \bar{E} - \Delta E \left[ \frac{\sum_i Q_{0i} + \sum_{k \neq 0} Q_{kl}}{1 + \sum_i Q_{0i}} \right]. \quad (B.6)$$

By using :

$$\sum_i Q_{0i} + \sum_{k \neq 0} (k + l) Q_{kl} = \sum_{k \neq 0} (k + l) Q_{kl},$$

and

$$\sum_i Q_{0i} + \sum_{k \neq 0} Q_{kl} = \sum_{k \neq 0} Q_{kl} = 1,$$

one can rewrite equations (B.5) and (B.6) as follows :

$$\bar{E}_e = \bar{E} - \Delta E \left[ \frac{\sum_{k \neq 0} (k + l) Q_{kl} + \sum_i Q_{0i}}{1 + \sum_i Q_{0i}} \right]. \quad (B.7)$$

$$\bar{E}_o = \bar{E} - \Delta E \left[ \frac{\sum_{k \neq 0} (k + l) Q_{kl} - \sum_i Q_{0i}}{1 + \sum_i Q_{0i}} \right].$$

The even-odd effect on the average kinetic energy of the fragments being usually defined by :

$$\delta \bar{E} = \bar{E}_e - \bar{E}_o,$$

one can then rewrite this expression in terms of our above terminology as :

$$\delta \bar{E} = \frac{2 \Delta E}{1 - (\sum_i Q_{0i})^2} \left[ \sum_{i} Q_{0i} \sum_{k \neq 0} (k + l) Q_{kl} - \sum_{i} Q_{0i} \right]. \quad (B.8)$$

References


