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KINETIC-ENERGY DISTRIBUTION FOR SYMMETRIC FISSION OF $^{236}\text{U}^\dagger$

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Abstract

KINETIC-ENERGY DISTRIBUTION FOR SYMMETRIC FISSION OF ^{236}U .

Fission fragment kinetic-energy distributions have been measured at the Grenoble high-flux reactor with the Lohengrin facility. Spurious events were eliminated in the symmetric region by a coherence test based on a time-of-flight measurement of fragment velocities. A Monte-Carlo calculation is then performed to correct the experimental data for neutron evaporation. The difference between the most probable kinetic energy in symmetric fission and the fission in which the heavy fragment is 'magic' ($Z_H = 50$) is found to be $\cong 30$ MeV. The results suggest that for the symmetric case the total excitation energy available at scission is shared equally among the fragments.

1. INTRODUCTION

A striking feature of the fission process is the fragments mass-asymmetry observed for low-energy fission of nuclei in the Actinides region. The liquid drop model cannot account for this result, which is commonly attributed to shell structure effects favoring, at an appropriate stage of the fission process, deformations deviating from reflection symmetry [1].

It is also well known that for fissioning nuclei close to Uranium, the kinetic energy release shows a pronounced dip near the symmetric mass division together with a greater number of evaporated neutrons. The absolute value of the energy difference between symmetric and asymmetric fission has been measured in several experiments [2,3,4] but the results are in conflict between each other. All these data are obtained by the double fragment kinetic energy method which suffers from a poor mass resolution (f.w.h.m. $\approx 3 - 4$ a.m.u.) and for which the contamination by spurious high yields events is very difficult to overcome.

In the case of fissioning nuclei around the Actinium, the mass distribution shows triple humps and this feature is considered as an evidence for two different fission components: a symmetric fission behaving like a liquid drop and the asymmetric component which is considerably dominant for the Uranium fission.

[†] Experiments carried out at Institut Laue-Langevin, Grenoble, France.

* And USM/Grenoble, France.

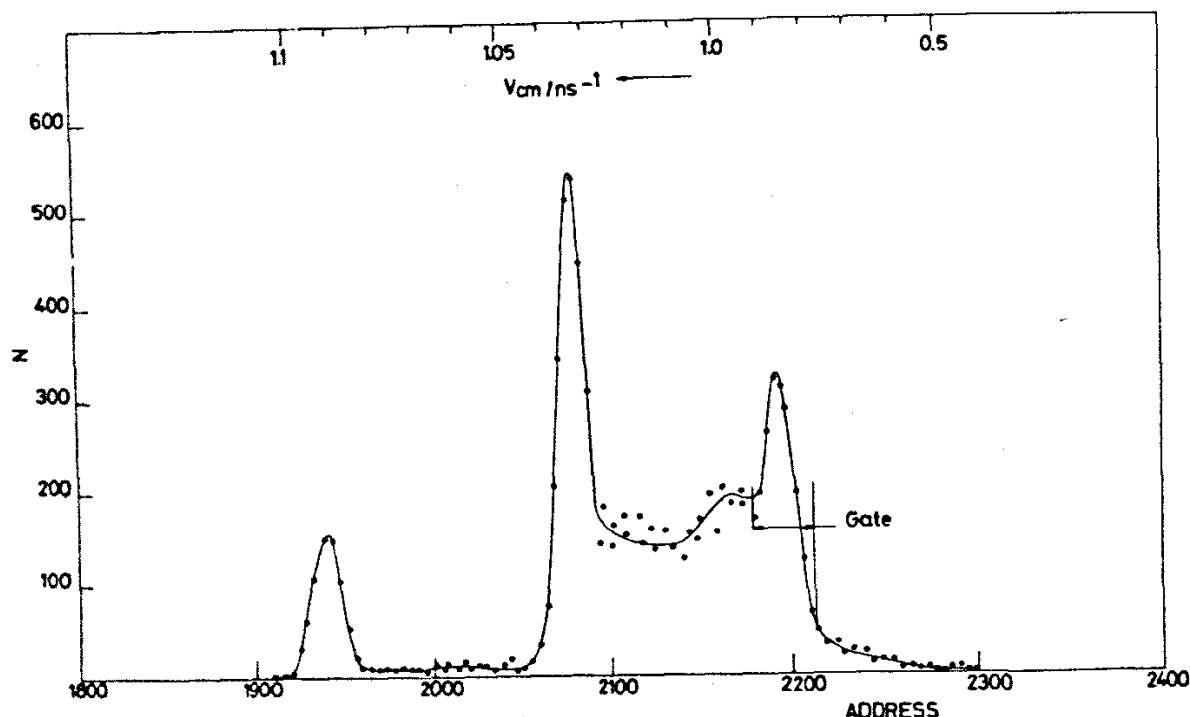


FIG.1. Time-of-flight or velocity spectrum measured for mass 118/19 at Lohengrin. The fragments with the normal speed (corresponding to the field settings) fall inside the gate and represent only a small fraction of the total spectrum.

There is still another open question for Uranium symmetric fission : do the nearly symmetric masses correspond or not to symmetric deformations of the two fragments ? In order to answer to such problems the kinetic energy distribution of masses around symmetry has been studied taking advantage of the high mass and energy resolution provided by the separator Lohengrin. Furthermore the width of the kinetic energy distributions, which is directly correlated to the distribution of the scission shapes, represents a very crucial test of changes in the scission configurations.

2. EXPERIMENTAL METHOD

2.1. Technical details

The separator Lohengrin (installed at the Institute Laue-Langevin) provides fission fragments of a given A/q (mass over ionic charge), at the collector of the instrument. Along the collector the fission fragments are distributed according to their velocity.

Unfortunately, the kinetic energy distribution measurement is not straightforward, when one is interested in low counting rate events, due to the fission fragments background of the instrument. The surroundings of the target are coated with uranium and thus emit fission fragments which are collected with a different velocity. This background becomes critical for the very low counting rates characteristic of symmetric fission in U^{236} .

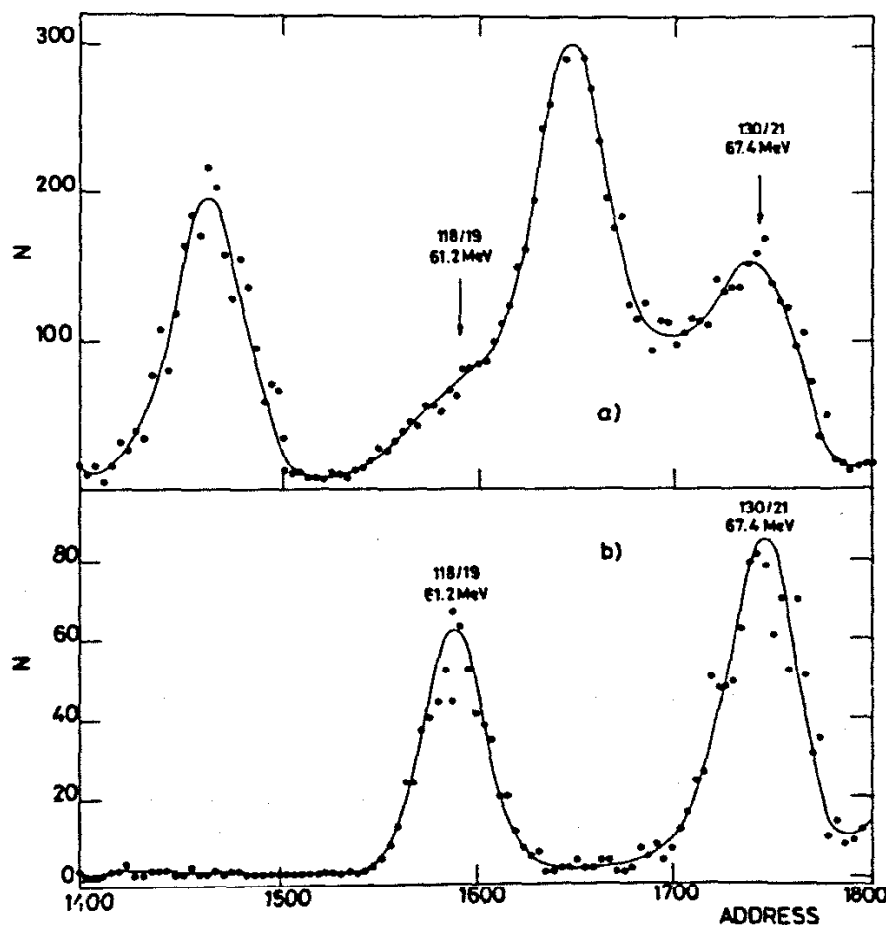


FIG.2. a) Ungated energy spectrum from the solid-state detector showing that the expected peak for mass 118/19 is completely hidden by the background;
 b) energy spectrum gated by the time-of-flight (Fig.1) showing the expected peaks for masses 118/19 and 130/21 (nearly the same A/q).

In order to eliminate these spurious events, the speed of the fragments was analysed by a time-of-flight system. The secondary electrons emitted by a carbon-foil, located in the focal plane of the instrument, were collected on channel-plates, thus providing a very fast "start" signal (80 psec.). After a flight path of approximately 30 cm, the ions were stopped into a silicon surface barrier detector, which was giving the "stop" signal and the kinetic energy measurement of the fission fragment. The two parameters (time T and energy E) events are then recorded on a magnetic tape correlated to a M-20 (Intertechnique) data acquisition device. The final resolution of the time-of-flight was around 250 p.s. due to the geometrical arrangement of the carbon foil (45° angle with the beam direction). A typical time-of-flight spectrum is shown in Fig.1, for which the spurious events proportion is very large. The corresponding ungated energy spectrum is shown Fig.2 a) together with the same spectrum, b) gated by a time window. The expected peak for mass 118/19 which is completely hidden by the contamination in the ungated spectrum, shows up very unambiguously in the gated one. Some other peaks are present in the gated spectrum but they correspond to a normal interference from masses for which the A/q ratio is

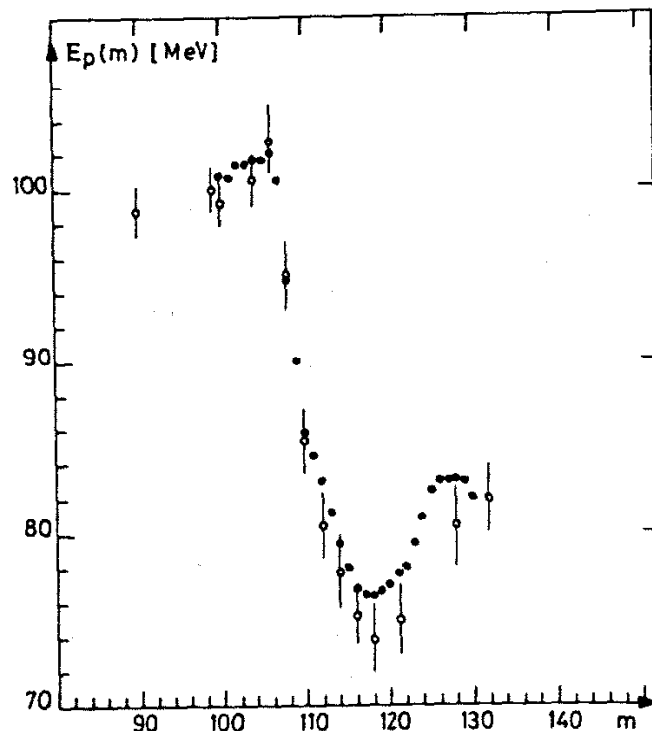


FIG.3. Most probable kinetic energy E_p as a function of the final mass m :

○ experimental data;

● Monte-Carlo simulation.

equal or very close to 118/19 (87/14, 93/15, 130/21). The correct assignment of all peaks present in the gated spectrum, represents a very good crosscheck of the efficiency of the background reduction.

2.2. Target thickness correction

The target thickness was $100 \mu\text{g}/\text{cm}^2$ of UO_2 , coated with a thin Tantalum film to slow down the burn up of the target. The average energy loss in the target, together with the dispersion of the distribution have been measured for two approximately complementary (90-144) (high yield masses). The energy loss and dispersion corrections can thus be tested with the conservation laws. The measurements of Wohlfarth [5] at Lohengrin have been used to calculate the average energy loss: $4 \pm 1 \text{ MeV}$. This result can reasonably [6] be considered as independent of the charge and energy at least for the values characterizing the fission products. The dispersion introduced by this energy loss corresponds to an additional variance of 6 MeV^2 for mass 144. This variance depends upon the mass and energy according to the law :

$$\sigma_{\text{target}}^2(\text{Ai}) \approx \sigma_{\text{target}}^2(144) \times \frac{E(\text{Ai})}{E(144)}$$

and the true variance σ_{th}^2 of a given mass Ai is calculated from the measured one σ_{exp}^2 by the relationship :

$$\sigma_{\text{th}}^2(\text{Ai}) = \sigma_{\text{exp}}^2(\text{Ai}) - \sigma_{\text{target}}^2(\text{Ai})$$

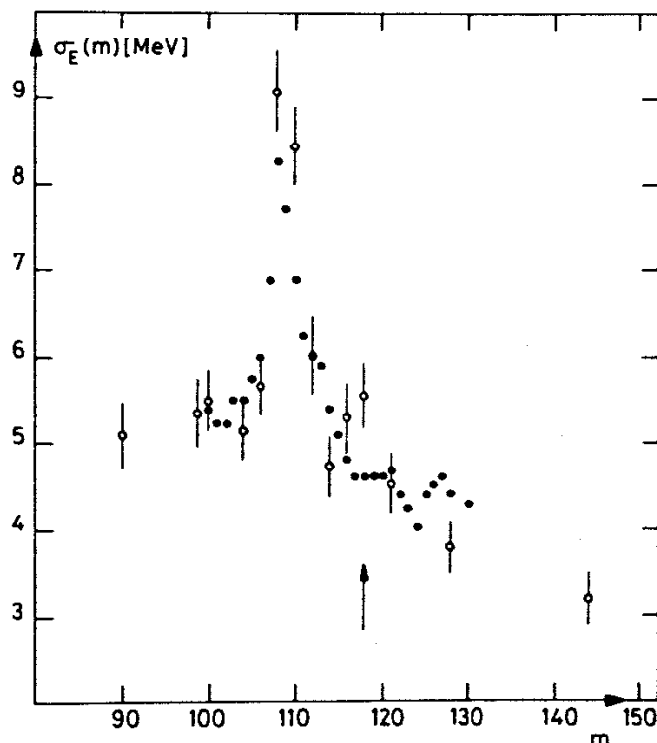


FIG.4. Root-mean-square width of kinetic energy distribution as measured at Lohengrin (m represents the final mass). Notice the strong increase of σ for $m = 108$.

○ experimental data;

● Monte-Carlo simulation.

3. EXPERIMENTAL RESULTS

3.1. Most probable kinetic energy and distribution variances

The most probable kinetic energy E_p is plotted in Fig.3 as a function of the mass m of the final fragment (these experimental values are not corrected for the neutrons evaporation). Two striking features emerge from these results :

- the sudden fall of the most probable kinetic energy when going towards symmetry.
- the low energy obtained for symmetric masses.

The root-mean square width σ of the kinetic energy distributions are indicated in Fig.4 as a function of m . The average value of σ for symmetric fission is 5.3 MeV, but very large values have been obtained for masses 108-110, corresponding to fwhm values around 20-22 MeV.

3.2. Monte-Carlo simulation

If one wants to deal with the initial values of the measured quantities (mass, energy, variance), the neutron evaporation must be taken into account. Therefore the neutron evaporation has been simulated using a Monte-Carlo code, in order to compare our results to other measurements (obtained by a double energy method for instance), for which the effect of

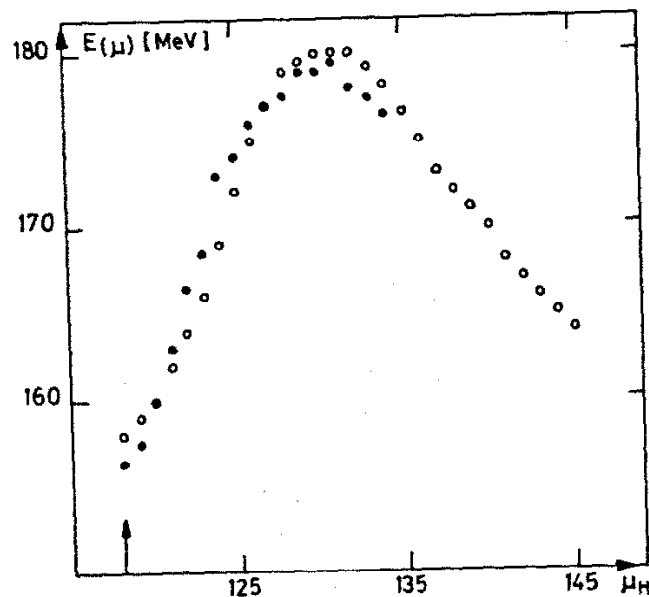


FIG.5. Most probable kinetic energy as a function of the pseudo-mass. The experimental data are taken from Ref.[3]

- experimental data;
- Monte-Carlo simulation.

neutron evaporation might be completely different. The data inputs used for the simulation were as follow :

- the total kinetic energy of both fragments as a function of their initial masses : $E_T(M_1, M_2) = E_1 + E_2$
- the dependence of the root-mean square width $\sigma_{E_T}(M_1, M_2)$ with initial mass.
- the average number of evaporated neutrons as a function of initial mass [7]
- the primary yields for the initial masse $Y(M)$.

The results of the procedure was then written on a magnetic tape event by event, each of them beeing characterized by :

- initial masses and total energy : M_1, M_2, E_T
- final masses and energies : $m_1, m_2, \epsilon_1, \epsilon_2$

In a first step, the simulated events have been selected according to their final masses, in order to check the compatibility of the input data with the energy distribution of final masses as measured at Lohengrin. During a second step, the data analysis procedure which is commonly used in the double energy method [7] has been applied to the simulated events. Thus for a fission event simulated with masses M_1, M_2 and final energies ϵ_1, ϵ_2 the pseudo-masses μ_1 and μ_2 are calculated by : $\epsilon_1 \mu_1 = \epsilon_2 \mu_2$ ($\mu_1 + \mu_2 = 236$).

Finally the simulation is considered to be satisfactory when the initial energy distribution $E_T(M_1, M_2)$ reproduces at one and the same time the experimental results obtained at Lohengrin, those resulting from the double energy method and the final masses distribution $y(m)$.

The agreement of the Monte-Carlo simulation with the distribution of energy measured at Lohengrin is shown in Fig.3 and with the recent results of Asghar and al. [8,9] in Fig.5 (double energy method). To perform this last comparison the correction for the resolution of the

detector had to be introduced at the end of the simulation since it was affecting the slope of the experimental curve. The difference between simulation and experience remains in all cases smaller than 2 MeV for total energy.

Another interesting point is the peak observed in the final σ_E values obtained at Lohengrin, for masses 108-110 (Fig.4). The simulation reproduces the peak although the initial dependence $\sigma_{ET}(M_1, M_2)$ was a smooth curve. This sudden rise of the final width of the kinetic energy distribution is then originating exclusively from the neutron evaporation process. The region around masses 108-110 is indeed characterized by :

- a sudden fall of the kinetic energy with mass number,
- a large number of evaporated neutrons. Therefore a given final mass m results from several contributions of initial masses $M, M+1, M+2, M+3$, each of them corresponding to a very different value of the most probable kinetic energy, thus broadening considerably the final distribution.

3.3. Total kinetic energy

For a given mass division the total kinetic energy $E_T(M_1, M_2)$, necessary to calculate the total excitation energy, has been obtained from the simulation, together with the variance σ_{ET} , as described in the preceding section. The dependence of $E_T(M_1, M_2)$ and σ_{ET} with the heavy fragment mass is represented in Fig.6 (full circles). For comparison the total kinetic energy as deduced directly from the measurement of the final energy of the light fragment (open circles) and of the heavy ones (open triangles) are also indicated. As a matter of fact the total energy is commonly calculated by the relationship :

$$E_T(m_H + \nu(m_H)) = \epsilon_H \left(1 + \frac{\nu_H}{m_L} \right) \frac{M_0}{m_0 - m_H - \nu_H} \quad (1)$$

and a similar one for the light fragment.

As can be seen in Fig.6 these last evaluations do not agree with each other, the main discrepancy arising around mass 129 (about 10 MeV of energy difference). The reason for these differences can be understood easily : the final mass 128 is located in a region where the mass yield is increasing very rapidly, therefore the initial masses 129 and 130 will contribute significantly to the final mass 128. It is necessary that masses 129 and 130 evaporate respectively one and two neutrons, though their most probable excitation correspond to about 0.3 neutron evaporated in the average. Thus the fragments of mass 129 and 130 which contribute to mass 128 will have an excitation energy higher than the average. The kinetic energy measured for the final mass 128 is then lower than the average due to the important contribution of masses 129 and 130. The same effect works in the opposite direction for the final mass 106 to which, due to their very low yields, masses 107 and 108 contribute very little. The events of mass 106 which do not evaporate neutrons correspond, thus, to lower than the average excitation energy or higher kinetic energy. Therefore, equation (1) cannot be used in those regions where the mass yields vary rapidly; it would lead to an underestimation of the total energy if the heavy mass energy is used and the contrary for the light mass.

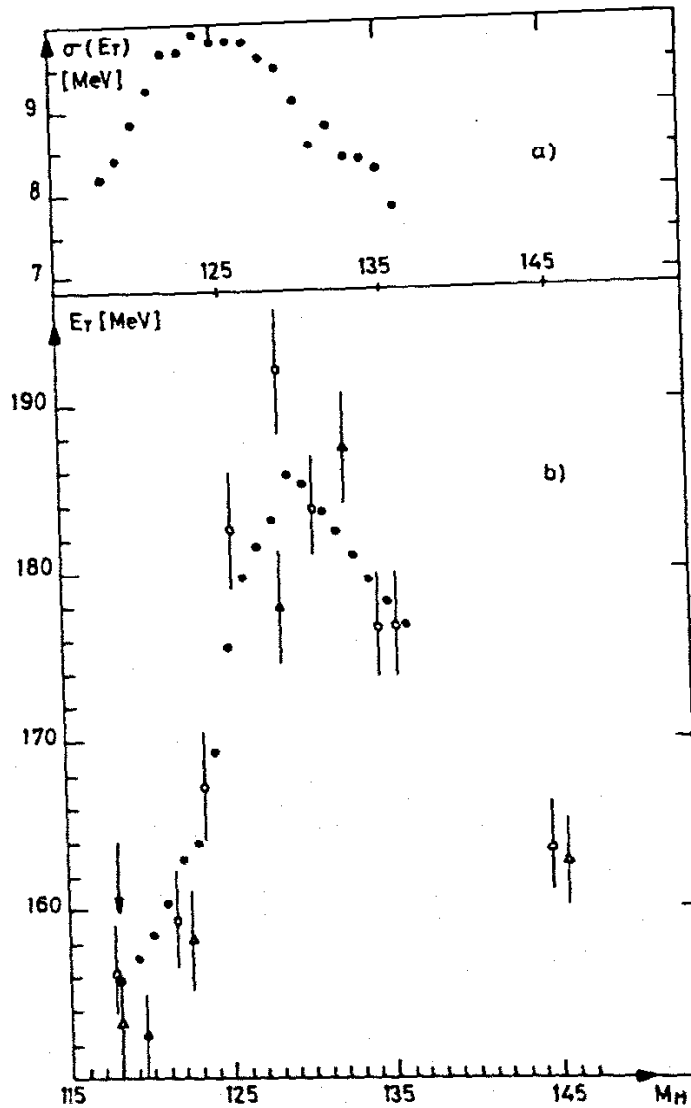


FIG.6. a) The root-mean-square widths of the total kinetic energy introduced into the Monte-Carlo simulation.

b) Total pre-neutron kinetic energy E_T as a function of heavy-fragment mass. The vertical arrow indicates symmetric fission.

○ E_T calculated from the light fragment kinetic energy measurement (relation (1));

△ E_T calculated from the heavy-fragment kinetic energy;

● initial values of the Monte-Carlo simulation.

4. DISCUSSION

4.1. Evidence for symmetrical shapes in symmetrical fission

The absolute value of the dip in the total kinetic energy was until now estimated to 22 MeV [8,9]. The result of the Monte-Carlo simulation corresponds to a $\Delta E = 30 \pm 4$ MeV difference between the total energy for the mass 129 and for the mass 118. This apparent discrepancy, can be attributed for an important part to mass resolution effects which lower the maximum around mass 129 and raise the minimum at symmetry.

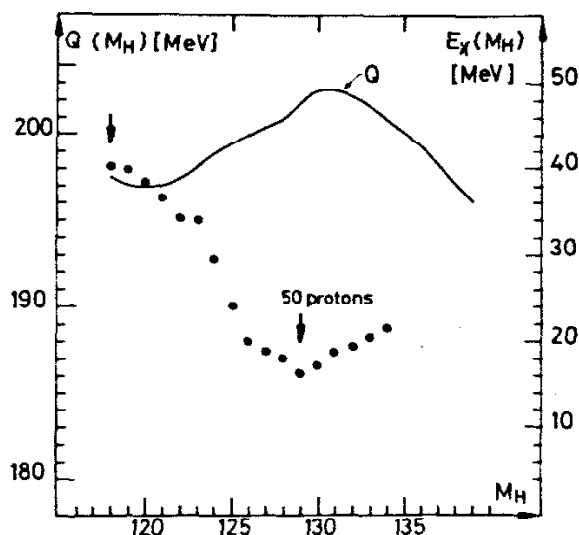


FIG.7. Energy release Q and total excitation energy of the two fragments E_x .

Nevertheless the kinetic energy for asymmetric fission is very low indicating a symmetrical share of the excitation energy among the fragments. This assumption is supported as follows :

The observed kinetic energy corresponds to a distance of 19.5 fm between the fragment centers at scission (assuming punctual charges and a purely Coulombian energy), therefore each fragment should have a deformation corresponding to $\beta > 0.8$. As the spherical radius for mass 118 is only around 6.5 fm, thus it seems unrealistic to attribute the whole deformation to one fragment alone.

Another point leads to the same conclusion : the values of σ_{ET} (Fig.6) are maximum around masses 124-125, corresponding to the maximum change of the kinetic energy with mass. This mass region represents then a shape transition from the asymmetric region around mass 129 to the symmetric shapes around mass 118 and the large distribution of shapes explains the maximum of σ_{ET} .

4.2. Excitation energy : influence of the 50 protons shell

The total excitation energy E_x corresponding to a given fission event can be deduced from the total energy release by :

$$E_x = Q - E_T \text{ (kinetic)}$$

The result of this calculation (Fig.7) shows a very large value (~ 40 MeV) for the excitation energy of symmetric fission, in agreement with the estimated value of Wahl [7] for the number of neutrons evaporated at symmetry ($\nu_T(118) \geq 4$).

The minimum value for the excitation energy is obtained at mass 129 rather than mass 132, in coincidence with the 50 protons shell in the heavy fragment (the influence of the 82 neutrons shell would occur around mass 134). The lack of completion of the 50 protons shell is also responsible for the sudden fall in the kinetic energy around mass 106 (Fig.3), since for masses higher than 106 the complementary fragment is no longer spherical.

5. CONCLUSIONS

The kinetic energy distributions for symmetric fission, together with the simulation of the neutron evaporation process lead to the following conclusions.

The kinetic energy dip for symmetric fission is larger than previously measured ($\Delta E \sim 30$ MeV).

The mass dependence of the kinetic energy exhibits a very sharp fall near symmetric fission.

The Monte-Carlo calculation can explain the peak in the widths of the experimental distributions measured at Lohengrin and has shown that it was not possible to calculate in a straightforward manner the initial kinetic energy (see section 3.3.). This work has also shown the strong influence of the 50 protons shell in the kinetic energy distributions.

Finally, the assertion that symmetric fission corresponds to a symmetric share of the excitation energy seems reasonable and could be strongly supported if one could measure an isotopic distribution in the neighbourhood of charge 46.

We are grateful to P. Bisenius, M. Bolore, J. Girard, R. Joly, C. Mazur, M. Ribrag, C. Signarbieux (DPh-N/MF, Saclay), for their help in the initial stage of the experiment.

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DISCUSSION

H.G. CLERC: I am glad to see that, according to your data, the $Z = 50$ shell produces the largest average total fragment kinetic energy. This tallies well with the data given in the paper which I presented today (see SM-241/F2 of these Proceedings).

My question is, which isobaric Z distribution did you assume in order to be able to calculate the Q value and average fragment excitation energies?

R. BRISSOT: The mass values A_L and A_H result from the superposition of different Z values close to the $2p$ value of the charge distribution. Hence to obtain the most probable Q value for a given mass split we have to obtain a weighted average of the masses of the different elements on the basis of their estimated independent yields.

M. ASGHAR: Do you consider that E_K around mass 130 can be reduced by more than 5 MeV merely by resolution effects? Furthermore, do you feel that the ^{235}U Monte-Carlo calculations can explain the $\sigma_{E_K}(\mu_H)$ results for other nuclei, such as ^{241}Am , ^{243}Am or ^{245}Cm ?

M. MONTOYA: E_K is reduced not only by the energy resolution effect but also by the $Y(M)$, $\nu(M)$ and $E_K(M)$ curve effects.

We have carried out Monte-Carlo calculations for ^{233}U , ^{235}U , ^{239}U and ^{252}Cf . For these nuclei the results can explain the $\sigma_{E_K}(\mu_H)$ curves. I feel that we can also explain the $\sigma_{E_K}(\mu)$ results for other nuclei.

H.A. NIFENECKER: Further to this question, I should point out that mass resolution effects not only attenuate peaks in the average kinetic energy curve, but also interfere in events with masses corresponding to low kinetic energy but high yields. This is why the difference in maximum average kinetic energy amounts to 5 MeV in the Monte-Carlo calculation and the double energy measurement.

H.J. SPECHT: The surprising fact that we have come back to a difference of 30 MeV kinetic energy between the symmetric and highly asymmetric fission of ^{236}U should delight Dr. Fraser and Dr. Milton at Chalk River. Their early double-velocity experiments yielded a figure of that order, but it fell into disfavour after the semi-conductor detector groups had discussed the point at great length during the Salzburg Symposium in 1965 and concluded that the figure was an overestimate.

P. ARMBRUSTER (*Chairman*): Dr. Brissot, what is the accuracy of the procedure used to relate primary and secondary masses?

R. BRISSOT: I think that Dr. Montoya can best answer that question.

M. MONTOYA: The accuracy depends on the number of neutrons emitted and the $Y(M)$ shapes. So it is not the same in all mass regions. In the region of most probable mass the figure is ~ 0.5 mass units.

There is, however, a general comment I would like to make. The Monte-Carlo calculations performed at Saclay (MONTTOYA, M., SIGNARBIEUX, C., DAKOWSKI, N., to be published.) and presented by Dr. Brissot require, as the input data, the total energy E_K , mass yield Y , neutron multiplicity $\bar{\nu}$ and assumed energy dispersion σ_{E_K} as functions of the primary mass M . The results of subsequent neutron evaporation are not very sensitive to the details of σ_{E_K} . We have shown that for different fissioning nuclei, even those with a constant $\sigma_{E_K}(M)$, the interplay of input functions is such that we produce high peaks and valleys in σ_{E_K} selected as a function of pseudo-masses μ or secondary masses m .

These peaks should not be confused, on account of neutron evaporation, with possible peaks indicating shell effects.

M. ASGHAR: In view of the fact that your experimental data show E_K as derived from E_L and E_H to be quite different, I should like to ask Dr. Clerc, who obtained E_K from only the E_L experimental values, how they would be altered by this difference.

H.G. CLERC: The different average TKE values obtained by Dr. Brissot for E_L and E_H may mean that the average neutron numbers $\bar{\nu}_L$ and $\bar{\nu}_H$ emitted per fission product and used to calculate the TKE are not quite correct.