MASS YIELDS FROM $^{235}U(n_{th}, f)$-REACTION IN THE SYMMETRY REGION

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Mass yields and kinetic energy distributions of fission fragments from thermal neutron induced fission of ^{235}U have been measured in the symmetry region with the mass separator Lohengrin of the Institute Laue-Langevin (Grenoble). The kinetic energy dip between asymmetric and symmetric fission amounts to 29 MeV, which agrees with previous data. A strong peak is observed in the widths of the kinetic energy distributions around the mass A=112. A presence of the second peak in the rms distribution for the heavy mass group does not unequivocally follow from the data measured. A comparison of the data to the libraries and to previous experimental data has been done. A strong deviation in the yields from the predicted values is found for the masses from A=113 to A=127. The origin of this deviation (asymmetry in the mass distribution) is discussed.

1 Introduction

The measurements of fission yields have been started right after the discovery of nuclear fission and are continuing, involving a large number of scientists and a large variety of experimental methods. Nowadays, one can distinguish two main areas of the use of fission yields: in fundamental physics, their significance lies in all aspects of the probability of fragment formation in the fission process, and in applied fields where they are needed for reactor design and its safe operation, waste management and nuclear material safeguards. Thus, the importance of having reliable data is well recognised nowadays by the scientific community.

However, in spite of many years of intensive research in this field, the experimental data set on fission is still not complete. This is first of all true for the valley of the mass yield curve where, for most of the fissile isotopes, even the mass yields are poorly known. Concerning isotopic yields, in this mass region there existed data up to recent time [1] on (mainly, cumulative) yields of only few isotopes (as ^{111}Ag or ^{115}Gd), which due to their relative long half-life times were easy to measure.

From the standpoint of fission process study, the symmetry region is most interesting because fission products undergo dramatic changes in the kinetic energy and in the neutron evaporation. This gives an important value to the information on fragment excitation and kinetic energy distributions.

One of the goals of our present research is to improve and to extend the available data set on the mass and isotopic yields in the region of symmetric fission of trans-uranium isotopes. First data from the fission of ^{245}Cm are already available [1]. In this paper, we present and analyze the results (mass yields and kinetic energy distribution) from thermal neutron induced fission of ^{235}U and compare with previous experimental data and compilations from libraries.

2 Experimental

The measurements were performed at the mass separator of unslowed fission fragments Lohengrin [2] installed at the high flux research reactor of the Institute Laue-Langevin in Grenoble. Specific fission products at Lohengrin are selected by a combination of a magnetic and an electric sector field, according to the mass over ionic charge (A/q) and energy over ionic charge (E/q) ratio. An use of an additional magnet (RED – Reverse Energy Dispersive – magnet) [3] added to the exit slit of the mass separator has provided an additional separation condition for fission products selected by Lohengrin and lead to practically background free measurements. A small ionization chamber with split anode [4] was used as a detector of separated fragments. Both, high efficiency of the ionization chamber and clean Lohengrin spectra allowed the measurements in the mass regions with low yield as it is the case in symmetric fission.

Kinetic energy distributions covering the energy interval of around 30 MeV (+15 MeV and –15 MeV from the most probable kinetic energy for each mass) were successively measured for 25 masses (from A=107 to A=131), each at mean ionic charge state. In each mass chain at least 8 ionic charge states at mean kinetic energies were measured. The evolution of the target intensity was followed as a function of exposure time, by daily measuring the reference spectra (kinetic energy distribution of A=100). The yield of mass A=100 was determined as well and used further as a normalization point for the data in symmetry. Its absolute value was taken from W. Lang et al. [5].

All the data measured were corrected for the experimental parameters. The detailed description for these corrections may be found in [6] or [7]. The difference to measurements in other mass regions is that in symmetry mass peaks are overlapping and have to be fitted with several gaussians (see Fig. 1). The final results obtained after all corrections and the integration over the kinetic energy are the absolute mass yields and parameters of kinetic energy distributions.

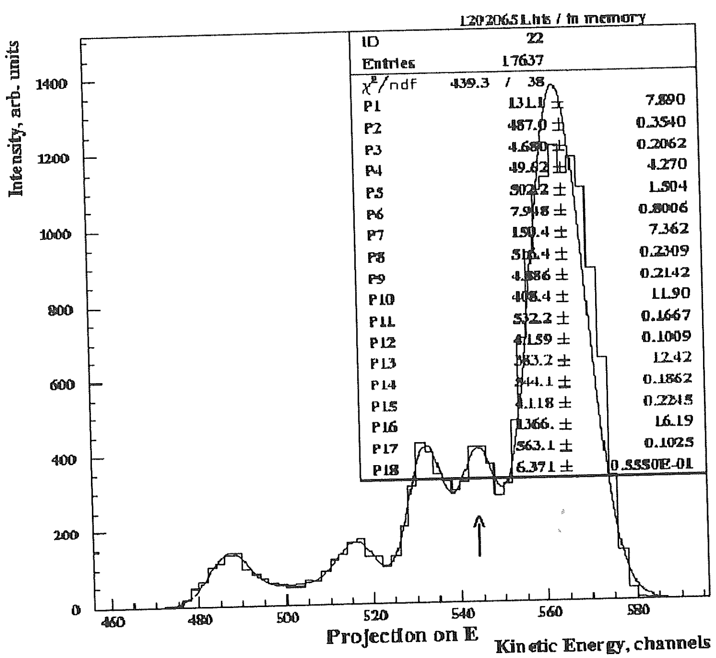

Figure 1. An example of the mass spectra in the symmetry region (projection on the total kinetic energy axis) measured at the separator settings $A/q=120/20$ and $E/q=65/20$ and fitted by a sum of 6 Gauss distributions. The numbers $P_1 - P_{18}$ given in the upper right corner form the triplets (P_{1+3*i}, P_{2+3*i}, P_{3+3*i}) with $i=0,1,\ldots,5$, which correspond to each peak parameters (the height, the position and the width, respectively). The mass of the interest is pointed with an arrow and the parameters of the peak are marked in red.

3 Results and discussion

3.1 *Kinetic energy distribution*

A distribution of the kinetic energy measured at Lohengrin usually shows a tailing to the low-energetic side, i.e. slightly deviates from the normal distribution. This deviation from the symmetrical shape is understood as due to effect of entrance window used in the experiment. To describe such a slightly asymmetric distribution in a proper way, an additional term was introduces into the normal distribution function:

$$I(E)=P_1*\exp(-0.5*[(E-P_2)/P_3]^2)*(1+P_4*[(E-P_2)/P_3]^3),$$

where parameters $\mathbf{P_1}$-$\mathbf{P_4}$ are to be found from the fit. The last parameter, $\mathbf{P_4}$, gives the deviation of the real distribution from the normal one. For symmetric shapes it converges to zero and the equation turns then back to the normal distribution.

In this way mean kinetic energies **Emean** (Fig. 2) and the widths (standard deviations) σ_E of the energy distributions were determined (parameters $\mathbf{P_2}$ and $\mathbf{P_3}$ in the equation above, respectively).

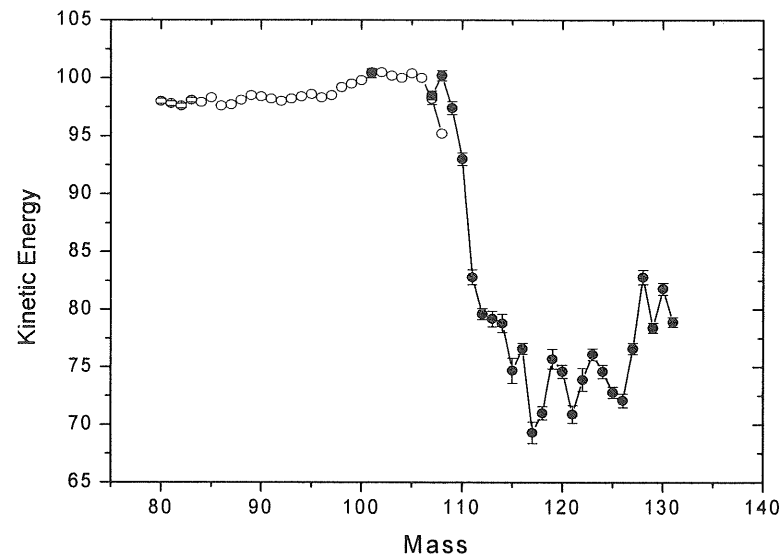

Figure 2. Mean kinetic energy of the kinetic energy distribution of the fission fragments as a function of their mass number. The values were corrected on the energy loss in the target material as described in [7]. For comparison the data from [5] (open circles) are taken.

To facilitate the numerical comparison with previous results from [8], the standard deviation values were converted to the root mean square (**rms-**) values (Fig. 3).

As seen from the Fig. 2 and 3, our data agree very well with the results obtained by W. Lang et al. [5].

From the Fig. 2 follows that the mean kinetic energy shows a very steep decrease in the vicinity of mass A=110. The energy dip between asymmetric and symmetric fission amounts to 29 MeV. This is in agreement with previous findings [8]. The sharp fall in the kinetic energy is associated to a large number of neutrons evaporated in this mass region. This is also reflected in Fig. 3 as a broadening of the width parameter of the energy distribution around the same mass numbers, which is caused by a mixing of several primary masses with strongly varying average kinetic energies, which contribute to the yield of the secondary masses.

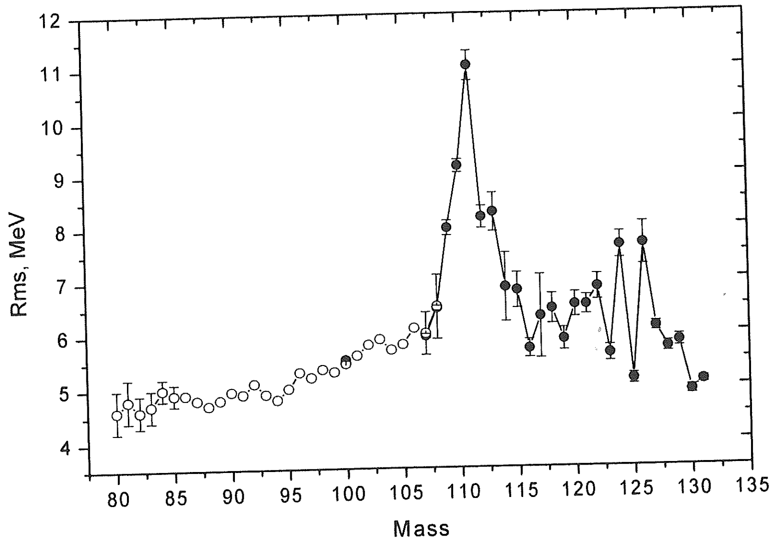

Figure 3. Rms width of the kinetic energy distribution of the fission fragments as a function of their mass number. Open circles are the data from [5] taken for comparison.

For the masses around A=120 we find that mean kinetic energies do not behave smoothly as it was shown in Ref. [8] but show some fine structure. A local maximum around the mass A=130, seen in the Fig. 2, was known. It was attributed to a compact scission configuration due to the shell closure in the heavy peak with higher Coulomb repulsion for nascent fission fragments. The excitation energy in this mass region is supposed to be very low, which results in very low neutron evaporation. This might allow the nuclear pairing effects to be seen in the kinetic energy distribution – what is indeed the case in the Fig. 2

The disappearance of neutron evaporation from the heaviest masses measured is well corroborated by the width behavior, which shows not only a steep decrease, but also some odd-even modulations for the masses around A=130 (Fig. 3).

An analysis of the widths of the kinetic energy distribution from Ref. [8] revealed a presence of a second hump around the mass A=125. Authors of [8] explained its presence from the point of view of potential energy surface

calculations in the fissioning nucleus showing a high deformability for different nuclear charge splits at a certain mass in this mass region.

The authors of [9] and [10] has given another interpretation for this structure. It was based on the Brosa model calculations, which interpreted this structure as a result of the coincidence of standard-II and superlong fission modes. In their calculations all fission modes were assumed to possess a symmetrical shape. Parameters of each mode (among them, widths and positions) were obtained from the fit of the mass distribution measured from the fission of ^{235}U induced by neutrons with incident kinetic energy from 0.006 eV to 130 eV. Since the mass distribution revealed certain asymmetry, this approach has not much predictable power for the other fissioning systems where reliable mass yields in symmetry region are still missing.

When studying fission process, systematics from the experimental data are useful. However, the available experimental data on the kinetic energy distribution in the valley region appear to be in some extent contradictory. The second hump in σ_E was not observed, neither for the $^{250}Cf^*$ [11] nor for $^{243}Am^*$ [12]. Data for $^{246}Cm^*$ taken from [13] point out this structure. Finally, the data from the present experiment, which were measured with a good accuracy, do not confirm the observation from [8]. The second peak in σ_E does not appear but, instead, we find a systematic, quite strong, oscillation of the observable around some average value (˜6.5 MeV) in the mass region of the interest.

The nature of this oscillation should be cleared, which requires further precise and systematic studies in this mass region. Since for the heavy fissioning systems like californium the first and the second maxima (or the oscillations, as found in this work) in width distribution can overlap and, due to this reason, be difficult to resolve, these data should be obtained from the fission of lighter trans-uranium elements.

3.2 *Mass yields*

The absolute values of the mass yields determined are presented in Fig. 4.

A striking feature here is a quite large discrepancy between theoretical and pure experimental data for the masses between A=119 and A=126. Yields of these masses exceed those given by libraries by a factor of two making the whole mass distribution asymmetric. It is interesting to note here that this asymmetry can be observed even in the exited fissioning system (up to 6 MeV of kinetic energy of incident neutrons) as it follows from the experimental results presented in [14].

Similar asymmetry in yields (even more sharply pronounced) was already observed before in the experiments with ^{245}Cm [13]. It can also be found in the ^{241}Am data [12] though for this nucleus not every mass chain in the symmetry region was measured and the statistical evidence of the data was considerably smaller than in the present experiment. The data of interest for ^{249}Cf can be found in [11]. From the first point of view, they reveal a perfect symmetry in the mass yields. A closer inspection however shows that this is nothing but an optical illusion: the yields are symmetric around the mass A=123 whereas the true symmetry point for ^{250}Cf* compound nucleus lies at mass A=125. These findings give a further evidence for our results.

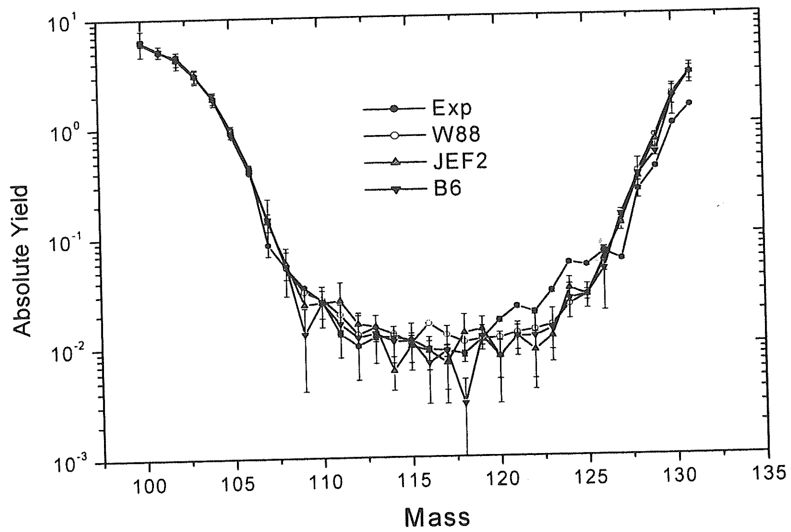

Figure 4. Absolute mass yields as a function of mass number: in black – present data (for fullness, yields of A=101-106 are taken from [5]); in red – semi-empirical data taken from A. Wahl [15]; in green – European library data (JEFF-2), in blue – American library data (ENDF/B6). The error bars given include all the uncertainties from the corrections done during the data evaluation plus statistical ones.

One further point to be considered is a fine structure in the mass yields. It is clearly seen that the yield of the masses A=124 and 126 lie higher then those of A=125 and 127 – unlike their complementary fragments where yields of successive masses drop very steeply. This odd-even staggering can be guessed as well in the yields of heavier masses where the number of evaporated neutrons should be very close to zero. It then naturally follows from our results that the region of low prompt neutron emission should be extended from the magic mass region to lighter masses (at least, down to A=124) – otherwise the fine structure in these yields would be washed out. One should however stress that this is exactly the region where masses show strong fluctuations in the parameters of the kinetic energy distribution. There probably exists certain dependence between these two effects, but it cannot be derived based only on present experimental results. Information on nuclear charge distribution in this mass region could spread a light on the origin of the fine structure in the mass yields. For this, some other experiments (i.e., gamma spectroscopy measurements) are to be done.

Due to low excitation energy at the mass A=124 and higher, it seems to be very unlikely that the enhancement found in the mass yields is due to neutron evaporation, which took place in the heavy fragment group. From the other hand, it is a well-known fact that neutron evaporation reaches its maximum in the vicinity of A=120 and drops rapidly down for the heavier masses.

We believe that the yields of masses from the heavy peak (i.e., of the masses measured in this experiment) are close to the primary yields whereas the yields of the heaviest masses from the light peak are strongly modified by the neutron evaporation process, which considerably lowers their values and is a reason for the mass yield asymmetry.

In fact, the asymmetry of the yield of the fission products (= of secondary masses) unequivocally follows from the neutron evaporation process – in experiment, one should never observe a symmetric distribution of fission products over the mass parameter. However, none of the libraries takes this evident statement into consideration. This leads to considerable discrepancies in the yields of some masses from evaluated data sets, as it is the case in Fig. 4, with our experiment.

4 Summary

The mass yield and kinetic energy distribution of fragments from thermal fission of ^{236}U* has been studied in the symmetry region at the mass separator Lohengrin. Main results of the study can be summarized in following statements:

1. The data on mass and kinetic energy distributions were precisely measured in the mass range from A=107 to A=131, which considerably improved and extended previous experimental results.
2. The oscillations of the width parameter of the kinetic energy distribution in the vicinity of the mass A=125 were observed. This is in contradiction with previous findings for ^{236}U* stating a presence of the second peak in σ_E distribution. A proper understanding of the origin of this peak (or of the oscillations) requires further precise studies in the symmetry mass range.
3. An asymmetry in mass yields was found and a discrepancy with the data from different libraries was stated. This was explained as a result of the neutron evaporation process, which reaches its maximum in symmetry and can strongly affect the yields of certain masses.
4. The discrepancy between theoretical and experimental data on secondary mass yields implies certain corrections for theoretical codes to be done.

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