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ABSTRACT

The fission fragments energies and masses characteristics and their correlations have been studied for the fast neutrons induced fission of U-235,236,238, Np-237 and Am-243 (En=16.5 MeV) and cold-hot fragmentation of U-235 (thermal neutrons and 1 MeV), U-236 (1MeV). New results for reaction Np-237(n,f) with neutrons from 0.3 to 1.3 MeV (near the barrier top) are presented.

INTRODUCTION

In the frame of a systematic study of the fission yields characteristics, the induced fission of $^{235,236,238}U$, ^{237}Np and ^{243}Am by neutrons of 16.5 MeV has been investigated. The measurements were performed with so-called double-energy method. In this method the fission fragments are detected by two collinear surface barrier detectors. The analysis was based on the mass and momentum conservation relations and the Schmitt-calibration procedure, in wich the detector constants can be determined from the pulse-height spectra of the $^{235}U(n_{th},f)$- reaction (the mean total kinetic energy to be 170.6 MeV).

For the $^{235,236}U$ cold fragmentation investigations a twin ionization chamber with Frish-grids was used.

Nuclear reaction $^3H(d,n)^4He$ was used as a source of the neutrons with the energy 16.5 MeV (the 3-d fission cross-section chance region), and $^3H(p,n)^3He$ - for 1 MeV neutrons.

1. URANIUM-235,236 COLD FRAGMENTATION BY NEUTRONS.

The binary nuclear fission is specified by a wide spectrum of fragment mass-energy, that makes the study of this complex and interesting phenomenon considerably difficult. The reaction energy Q consists of the Ek- fragments kinetic energy and their excitation E^*, removed by the neutron and γ-emission after the nuclear rupture. Particular interest is generated by the study of so called cold fragmentation for which Ek is near or equel to Q. In this case the neutron emission is energetically forbidden, and the mass spectrum shows a bright structure due to shell and odd-even effects.

Detailed investigations of the cold fission have been performed only the last ten years using combined detectors, which allowe to obtained not only mass-energy, but also charge spectra [1]. The obtained data reflected all the essential properties of cold compact fission. However, one fundamental question has been still unclear- fate of excitation energy collected by nucleus in the saddle point of fission barrier. In this work it is proposed to compare the cold fragmentation properties (yields, spectra) of U-235 irradiated by thermal and fast (En= 1 MeV) neutrons. In addition, a recent results of cold fission of odd uranium isotope in the reaction $^{236}U(n,f)$ are presented.

The work has been done at the accelerator KG-2.5 IPPE. As a fast neutron source the reaction $T(p,n)^3He$ was used. The thickness of the target (T-Ti) was 1mg/cm2 (water cooled copper backing). Thermal neutrons were obtained by fast neutron moderation in a polyethylene unit (20 cm thick).

Multidimensional fission fragment spectrometer has been built using twin ionization chamber with Frish-grids [2]. The uranium target consisted of a thin uranium tetrafluoride layer (50 mkg/cm^2) with 3 cm in diameter. To obtain the cathode conductivity the backing and the layer were sprayed with gold (0,03 mg/cm^2) As an operating gas the mixture of 90%Ar+ 10% CH_4 was used. The ionization chamber operated in gas passage conditions (with the intensity of 3 L/H). A special device maintained the gas mass in the chamber volume with the stability of 0.3% under the pressure of 1.05*E5 Pa. The bias voltage (-4.7 kV) was applied to the cathode (uranium target) and through resistor dividers to the Frish grids [2]. Experimental data evaluation was realized using the method analogous to [3].

Fig.1 gives a typical fragment mass spectra for different values of light fragment kinetic energy E_L (in MeV). Fission events have been selected from all the array. They conform to a fragment yield angle within the limits $\leq 16^o$ relative to target normal (for good resolution). Primarily, attention is drawn to the fact that for $E_L \geq$ 112 MeV the spectrum is formed by four essential components, grouping near M_L = 102,96,90,84 a.m.u.(Z_L = 40,38,36 and 34 respectively [1]). Contour maps Y-M-E_L (fig.2) demonstrate this effect in detail. Every component is defined by its values of the reaction energy Q and by total kinetic energy TKE, that is why the mass spectra in question are modulated by individual excitation energy in the scission point E^*. With the increase of E_L (or TKE) E^* is decreasing giving rise to cold fragments spectrum changes. So, the fraction M_L = 84 a.m.u. increases progressively and for E_L = 116 MeV it dominates in the spectrum (see fig.1 and.2 on the left). The

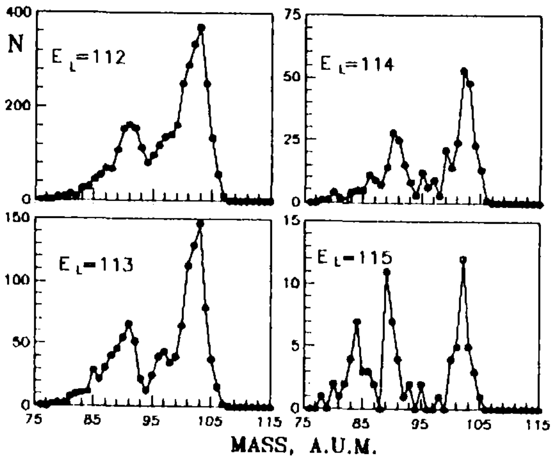


Fig.1.
Mass-spectra of U-235 thermal neutron induced cold fission for different E_L (in MeV) ($\vartheta \leqslant 16^\circ$)

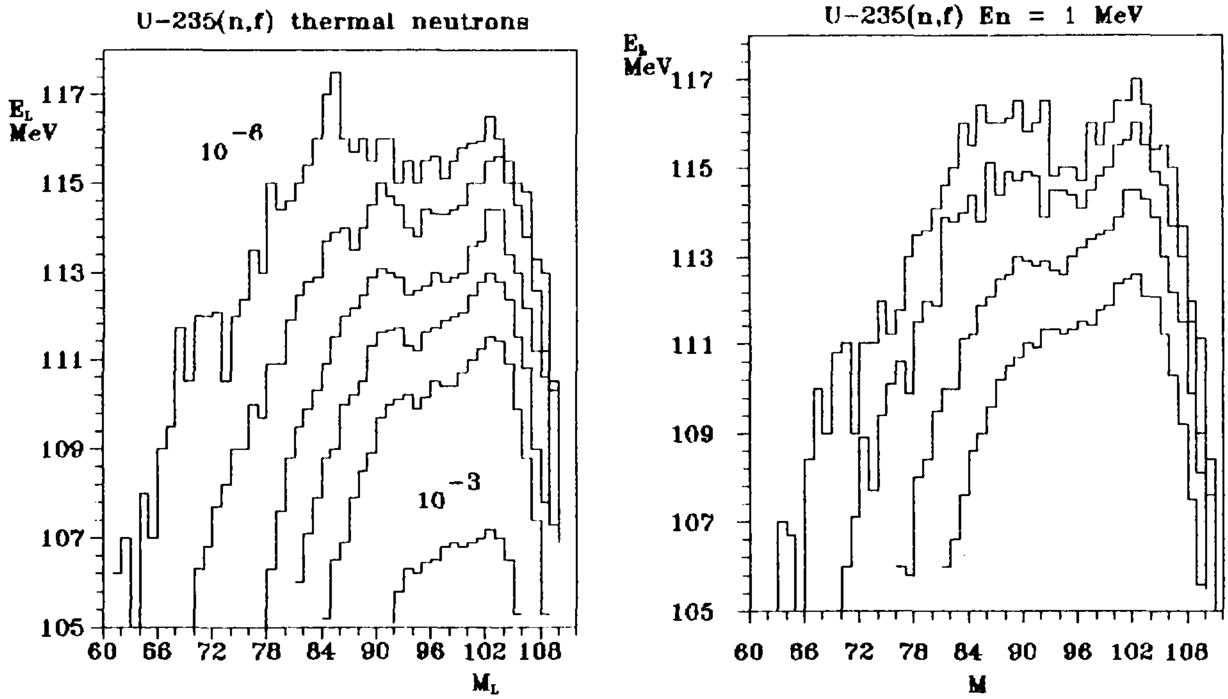


Fig.2. Contour maps E_L-M_L-Y for the two experiments. Y is from 10^{-3} to 10^{-6} yield levels

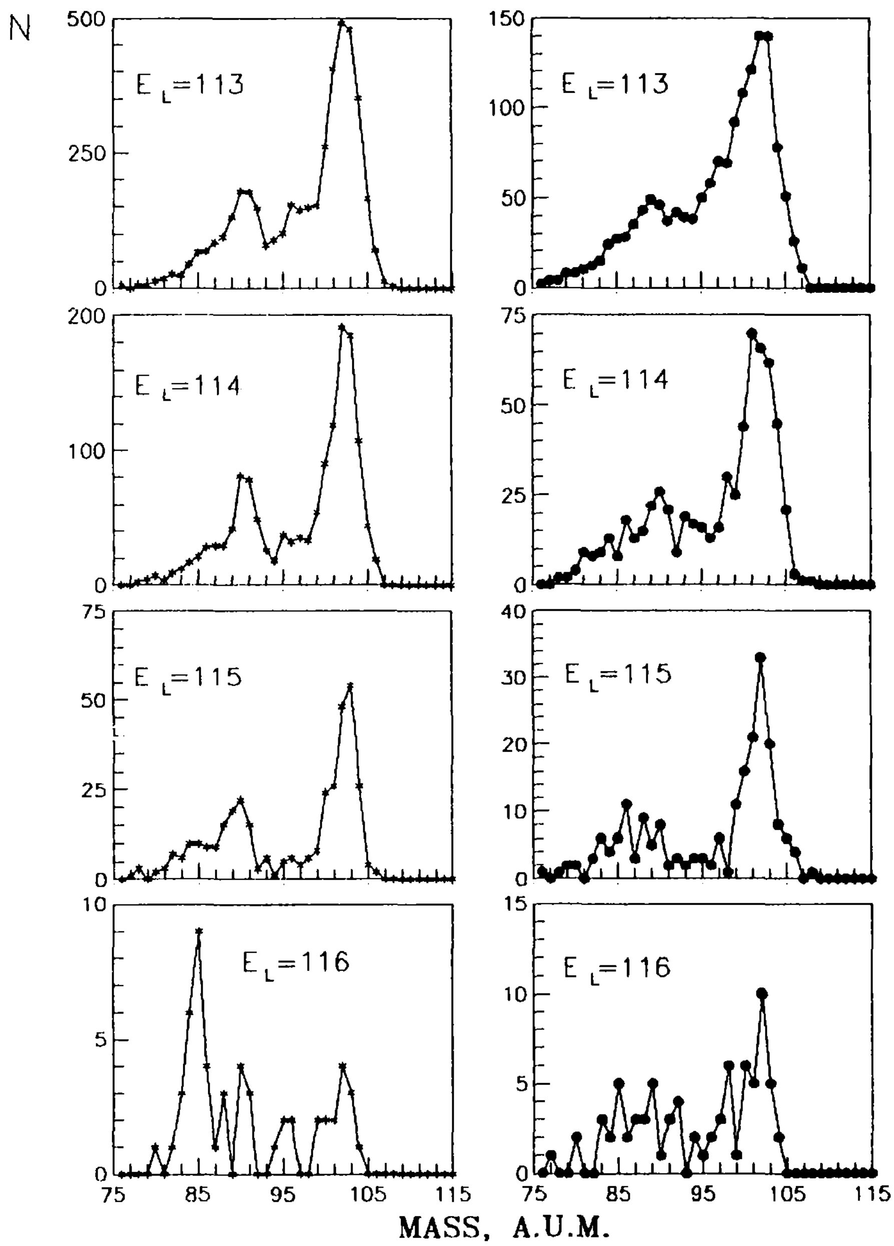


Fig.3. Mass spectra for fast (right) and thermal (left) neutron induced fission of U-235.

situation is opposite for the group M_L = 96 a.m.u.; with E_L = 115 MeV already its contribution is statistically negligible. Figure 4 demonstrates spectrum peculiarities more visually. This figure presents relative group yields for M_L = 96, 90 and 84 a.m.u. as a function of E_L. As a bench mark the component M_L = 102 a.m.u. has been taken. Mass spectra were described by four Gaussian curves. First moments were given precisely (102,96,90,84) and were not varied.

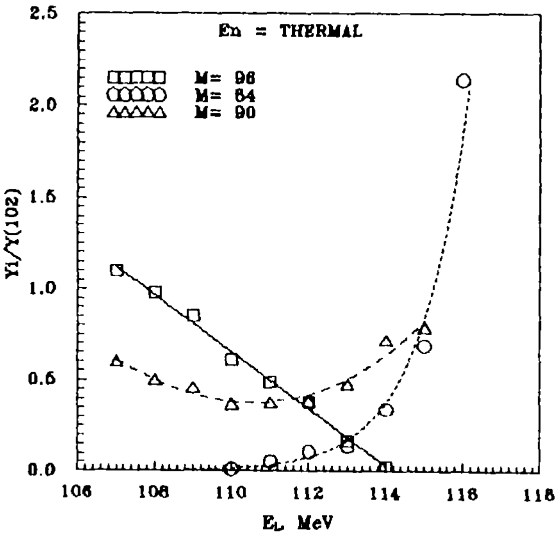


Fig.4. Yields of different mass components of the spectra relative to the component M_L= 102 a.m.u.

On the background of a sufficiently smooth linear reduction of ML=96 relative contribution and of an approximative constancy for ML=90 a.m.u. in all region of EL studied, the fragment addition around the mass 84 a.m.u. increases exponentionally. The explanation of this effect can be found from Q-values calculation in comparison with the TKE [2].

One can see from fig.5, which represents CF-yields for thermal and fast neutrons, that additional excitation contributed to the nucleus by 1-MeV-neutron is conserved up to scission point. CF-spectra (fig.3 on the right) confirm this. Side by side with already known four-component spectra , there exists qualitative and essential difference from the situation for fission by thermal netrons: for EL = 116 MeV the condition of the 102-mode prohibition is not yet been reached, that is one can observe some delay of a

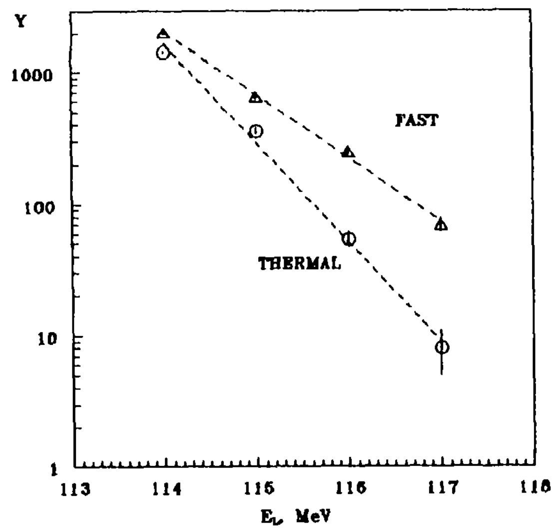


Fig.5. Integral yields of U-235 fission fragments for fast and thermal neutrons.

CF spectrum transformation in favour of the component M_L = 84 a.m.u.

On returning to the fig.5 it is necessary to make some important remarks. First, during the U-235 fission by fast neutrons the mean total kinetic energy of fragments, averaged over all fission modes increases relative to TKE in the thermal point only by 0,2 MeV [3] and can not influence the effects in question in a cold fraction region. Second, the δE_L- difference on the E_L axis of Y yields increases in the region of $E_L \geqslant 112$ MeV. Taking into account that the minimum neutron binding energy in light fragments group is approximately 3-4 MeV [4] we can explain the δE_L behaviour as a neutron emission manifestation and fix for the uranium the limit of the fragmentation without neutron emission (in all the region of masses from 80 to 105 a.m.u.), where E_L = 115 MeV. This limit refers first of all to the fraction M_L = 84 a.m.u., naturally. Unfortunately, in the frame of statistics obtained during the uranium fission by fast neutrons one can not realize a spectrum decomposition to components and trace the δE_L behaviour for each fragment group.

The data evaluation of cold uranium fragmentation by neutrons with the energy of 1 MeV permits to draw a conclusion that a nucleus excitation energy in a saddle point conserves up to the moment of the system rupture. It means that it is impossible to observe a true cold (TKE=Q) uranium-235 fragmentation process either with fast neutrons (E^*=2 MeV) or with thermal neutrons (E^*=1 MeV). In order to make this it is necessary to obtain the condition of subbarrier fission, which is connected with significant methodical difficulties. This situation can be reached for odd nucleus ^{237}U in the reaction $^{236}U(n,f)$ in the neutron energy range near 1 MeV. The mass- energy spectra for a cold fragmentation of ^{237}U have been measured. Experimental results are presented in fig.6,7.

In comparison with the data for ^{235}U fragmentation a main properties of the mass-energy spectra in the last case are quite similar: there are three pronounced components - 91/146, 97/140, and 103/134 a.m.u. Experimental yield-curve can be fitted by three Gaussian lines in detail (see fig.8). The gross-structure of the mass spectrum has just the same as for ^{235}U period - 6 a.m.u. On the other hand, one can conclude, that one unpairing neutron of the compound system ^{237}U is added to the light fragment during the fission process.

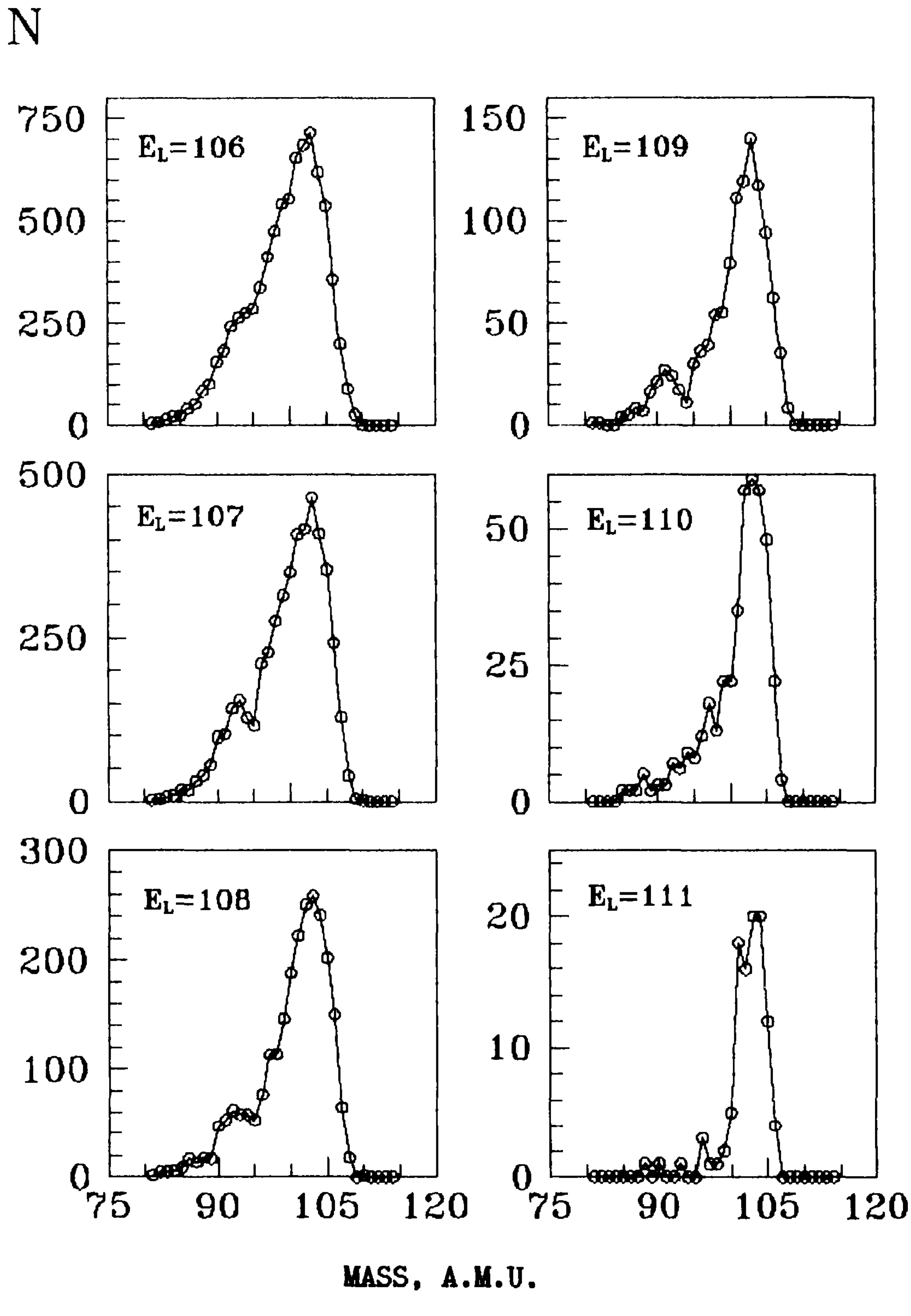


Fig.6. Fragment mass spectra for $^{236}U(n,f)$ reaction (E_L- in MeV) for COS$\vartheta \geqslant 0.75$; ϑ is the angle between fragment and incident neutron directions.

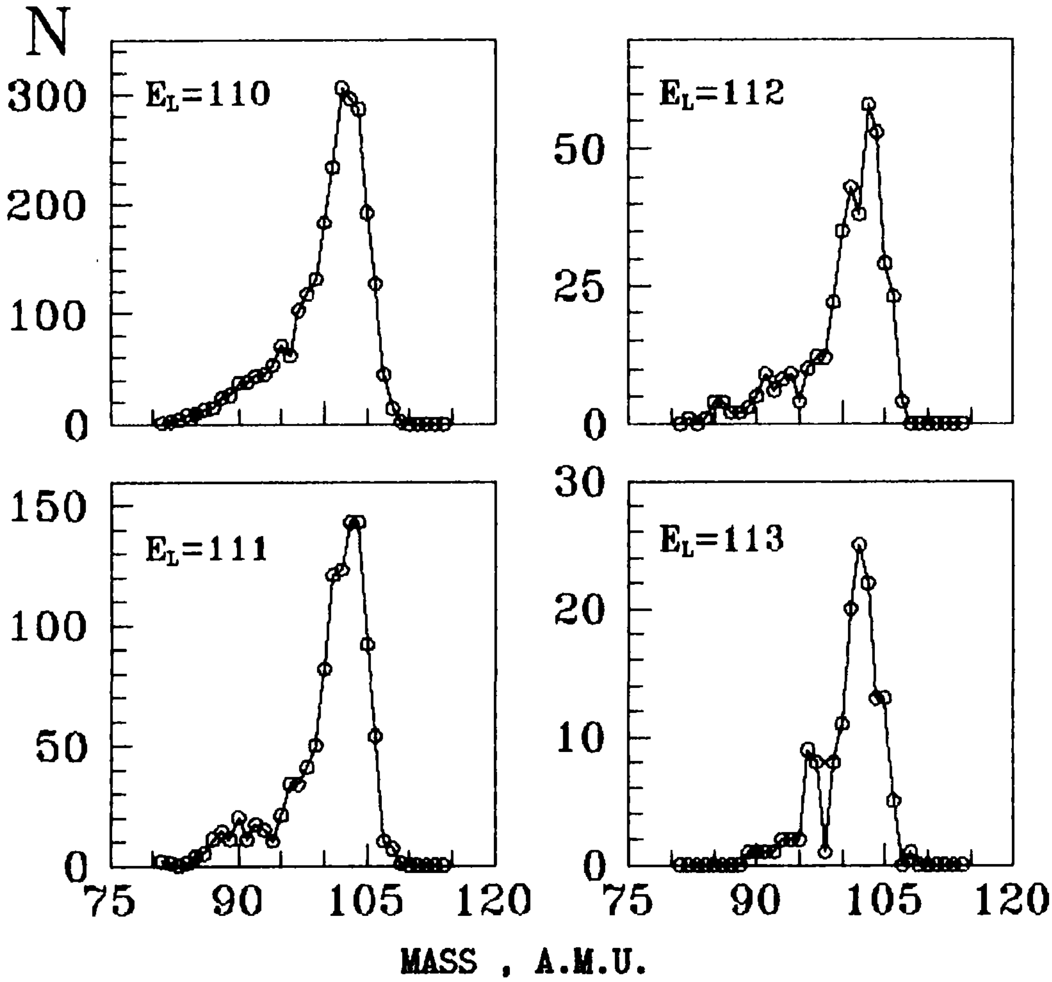


Fig.7. as in fig.6, but COSϑ ⩾ 0.5.

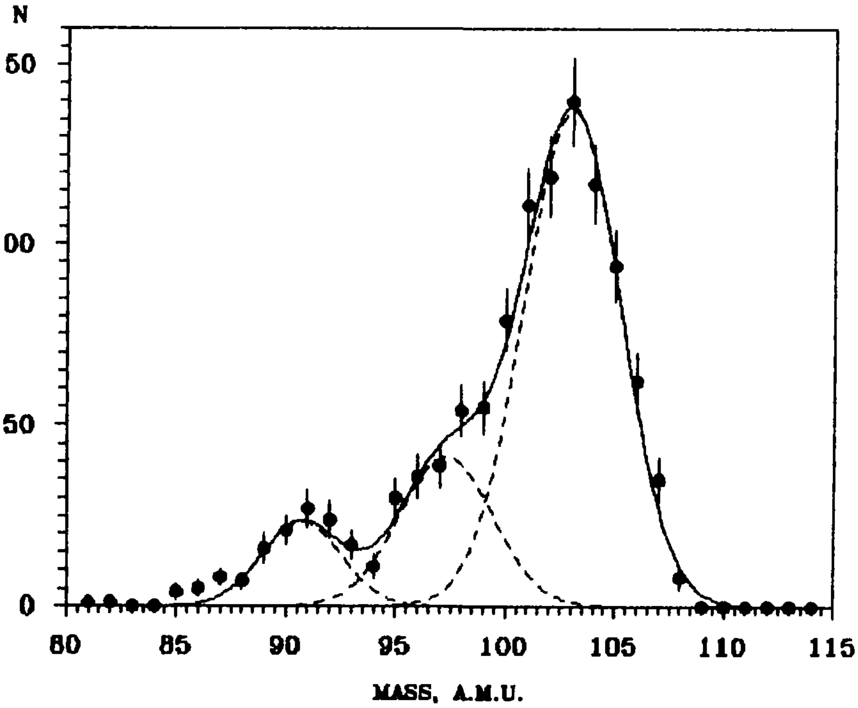


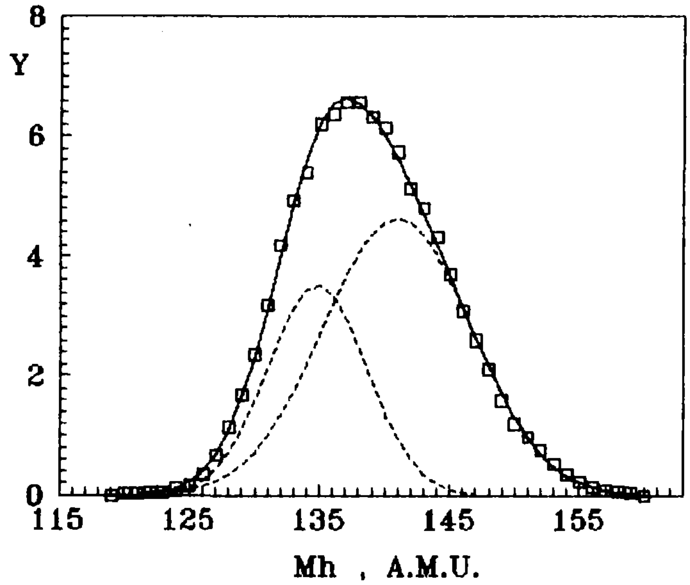
Fig.8. Main components of the mass-spectrum for E_L= 109 MeV, COSϑ ⩾ 0.75, reaction U-236(n,f).

Looking at the cold fraction of fission products having high kinetic energies in fission of ^{235}U or ^{236}U we find the mass distribution with a predominant yields at M_H = 134 and 140 a.m.u. The fine structure of the mass-distributions can be observed in wide region of free energy - S=(Q - TKE) - ≃ 7 - 8 MeV (see fig.2, left). For the high S-value connected with the final stage of the descent from saddle point up to scission , mutual overlapping of different components in question leds to the strong damping of the mass spectrum. However, the first moments of the components stay the same: 134 and 140 a.m.u. It means that we have separate mass channels -in good agreement with the Brosa-model of fission [5]. The period of 6 a.m.u. can be understood as a reflection of the proton-pairing effects in co-operation with the double neutron effect (2p+4n=6 a.m.u.). Starting point of the spectrum clasterization to be M_H = 134 a.m.u. influenced by strong shell-effects.

2. PROPERTIES OF THE FRAGMENTS FOR $^{237}Np(n,f)$ REACTION NEAR THE THRESHOLD.

Mass and kinetic energy distributions of fragments in a fission of compound system ^{238}Np have been measured using Si(Au) detectors to investigate a role of the top of fission barrier in the formation of the fragment properties. All experimental data obtained for 10 neutron energies from 0.28 to 1.28 MeV(fission threshold is equil to 0.7 MeV) were analyzed along the lines of the multi-exit-channel model of fission elaborated by Brosa, Grossman and Muller [5]. In the present work we assume the mass distributions of all fission channels to be symmetric and of Gaussian shape, and have used fitting method analogous to [6]. Fig.9 and fig.10 represent one example of fit of the mass and kinetic energy distributions respectively. The full line through the experimental points represents the result of the fit and is a superposition of the three fission channels.

Comparison of different components of mass spectrum for neutron energies 0.28 MeV and 1.28 MeV is represented on fig.11. One can see quite pronounced fluctuations of relative yields of the components during increasing of E_n - from subbarrier fission to above-barrier one. These fluctuations can be determined by the specific structure of the potential energy surface near the top of neptunium fission



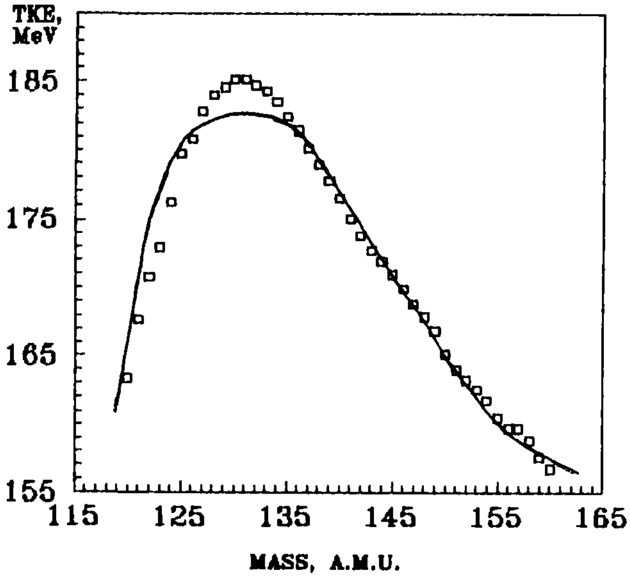


Fig.9. Mass spectrum of fragments for E_n = 1.09 MeV; lines - results of model fit (see text)

Fig.10. as in fig.9, but for kinetic energy ($^{237}Np(n,f)$).

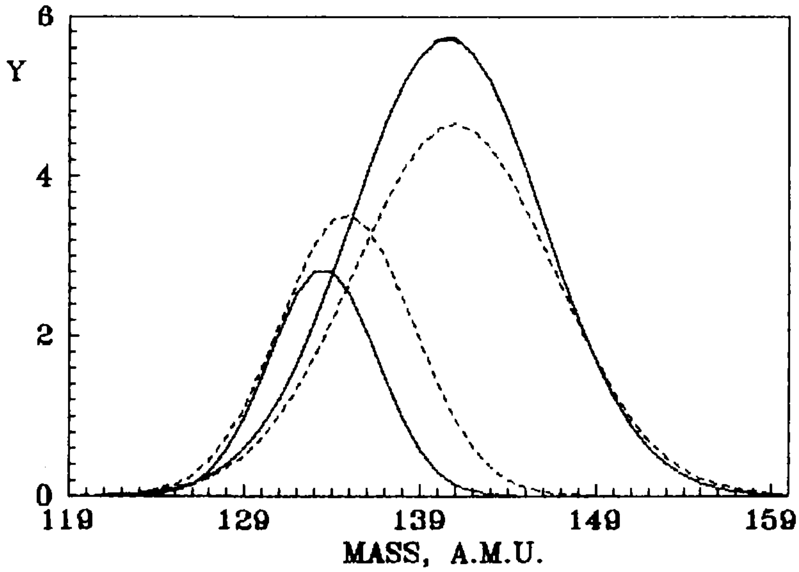


Fig.11. Fitting mass components for E_n= 0.28 MeV (full line) and 1.28 MeV (dashed line). $^{237}Np(n,f)$.

barrier - or more definitely - saddle points for separate mass channels.

The strong transformation of the mass distributions observed leds to the visible increasing of the mean total kinetic energy of the fission fragments of ^{238}Np compound-nucleus (fig.12). One can conclude that in the deep sub-barrier fission of ^{237}Np around the resolved resonances of fission cross section it is very possible to observe the strong TKE fluctuations.

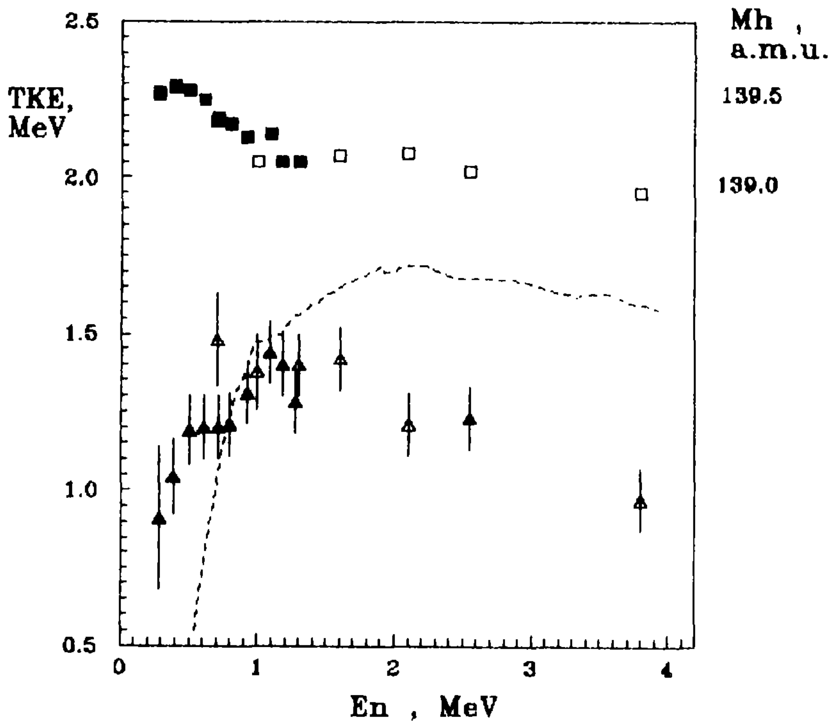


Fig.12. Mean values of TKE, M_H as a function of the neutron energy; line represents the fission cross section (arb. units); (□ △) - the data of [7].

The peak to valley ratio, P/V, between the yield of the 140/98 mass split and the average yield of the splits 118/120, 119/119 and 120/118 is found to decrease with increasing neutron energy. It is well known situation for induced fission of heavy nuclei. Around the neutron energy 1 MeV (from 0.71 to 1.09 MeV) the measured P/V ratio is 460 ± 82 - in agreement with the data of radiochemical measurements : 670 ± 70 [8]. The radiochemical method do not suffer from resolution effects, as can be the case in a 2E-method. The Table 1 shows, that P/V of ref.[10] is too small to be true. It is clear, that it is necessary to repeat measurements for the thermal neutrons.

TABLE 1. $^{237}Np(n,f)$,P/V-data.

METHOD	P/V	E_n	REF.
2E	460 ± 82	1 MeV	present
2E	420 ± 100	1.3 MeV	[7]
TOF	650 ± 125	0.8 MeV	[9]
RCHEM	670 ± 70	0.8 MeV	[8]
2E	129 ± 3	thermal	[10]

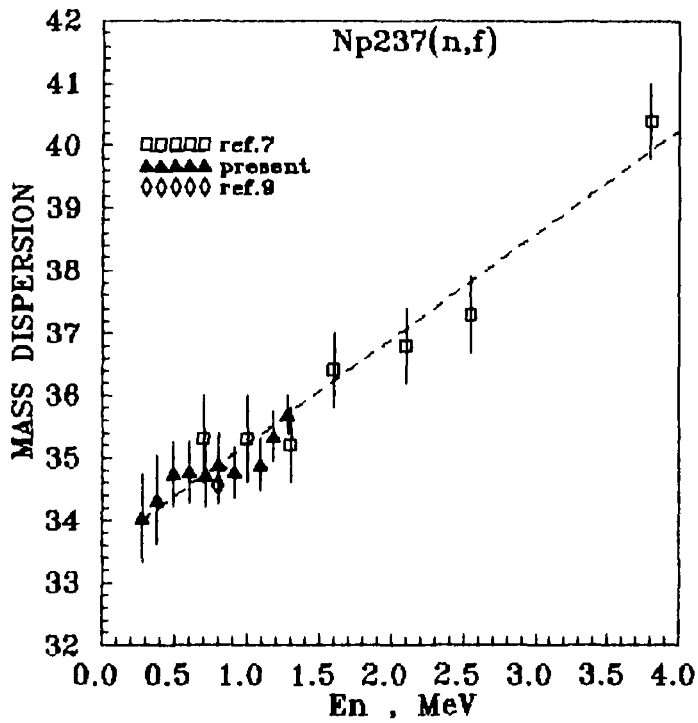


Fig.13. Energy dependence of σ_M^2 for the $^{237}Np(n,f)$-reaction.

Very interesting information can be subtracted from the mass distribution width as a function of incident neutron energy (or compound nucleus excitation). It's well known that inner hump of the barrier is higher than the outer one for ^{238}Np; it defines the fission cross section (fission probability) [11]. Fig.13 represents our data. We can see the smooth linear dependence $\sigma_M^2(E_n)$ in all region of neutron energy up to 0.28 MeV which is close upon the outer saddle point [11]. This result is in good agreement with our hypothesis elaborated in ref.[12] in connection with the problem of the energy dissipation as a function of system alongation after the outer saddle point of the fission barrier (details see in ref[12]).

3.MASS DISTRIBUTIONS OF FRAGMENTS IN THE FISSION INDUCED BY 16.5 MeV NEUTRONS.

The compound nucleus will be more excited as the incident neutron energy increases. The shell and pairing corrections will be partly washed out, and the fission process will approach a situation that can be described by liquid drop model. It would be expected to find an increase in mass-symmetric fission, and a broadening of the asymmetric mass yield peaks. Fig.14 shows the mass distributions of the fragments in the fission of ^{238}U by neutrons with the energies 1.22 MeV (sub-barrier fission) and 16.5 MeV (3-d chance fission).

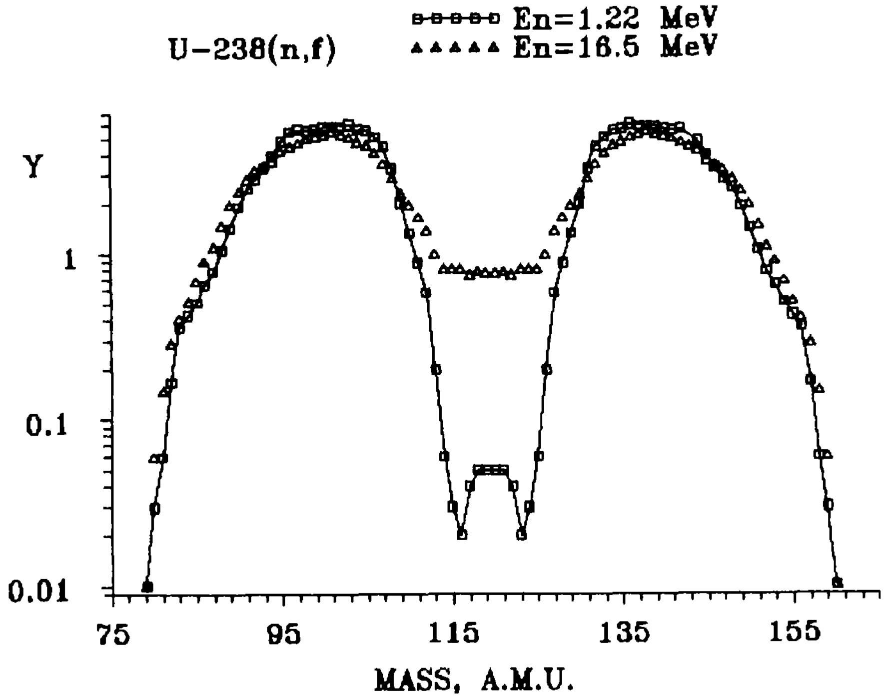


Fig.14. Mass spectra for $^{238}U(n,f)$-reaction with different neutrons.

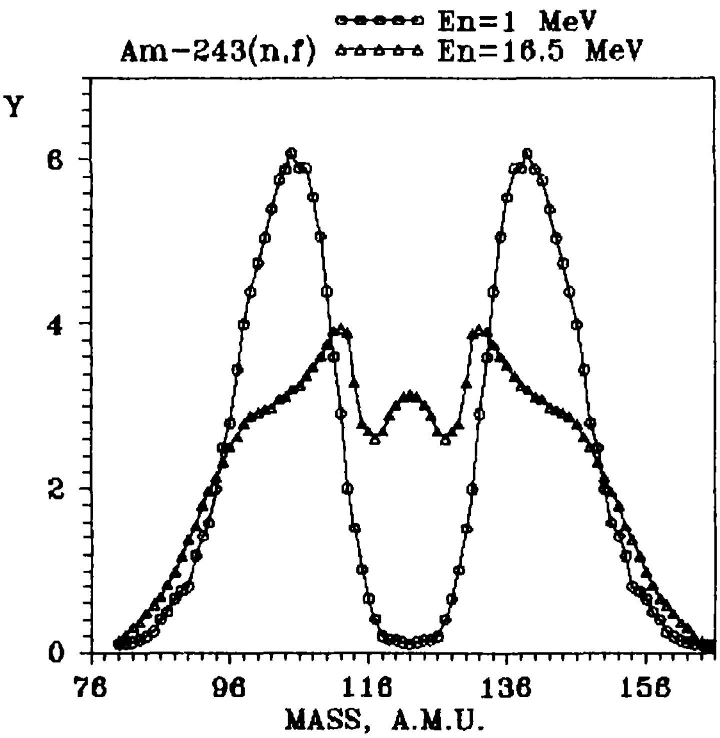


Fig.15.as in fig.14, but for $^{243}Am(n,f)$.

One can see the small but pronounced component of the spectrum near the mass symmetry for fission by 1 MeV-neutrons, because of large distance between asymmetric peaks. On the other hand, for very fast neutrons mass-symmetry region is structureless due to overlapping with the asymmetric peaks and has relatively high contribution to the spectrum. The inverse situation can be observed in the fission of ^{243}Am. Fig.15 represents the data for fissile system ^{244}Am only. Fig.16 demonstrates the partial contributions of separate channels to the total fission cross-section of $^{243}Am(n,xnf)$ reaction. The analogous quantities for ^{238}U are quite defferent because the first chance of fission cross-section for ^{238}U is relatively small. The same situation can be pointed out for $^{235,236}U$ and ^{237}Np. Therefore, experimental mass distributions have been evaluated as a mixture of different fission channels in the cross section by means of fit using Gaussian representation (see part 2). The example for ^{236}U is represented in fig.17.

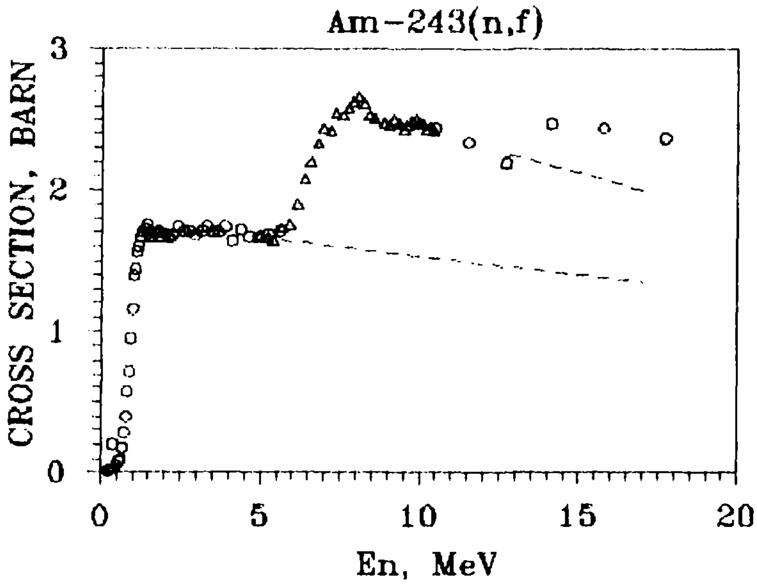


Fig.16. ^{243}Am fission cross section:
(o)- [13];
(Δ)- [14];
- - - fission chance contribution evaluation.

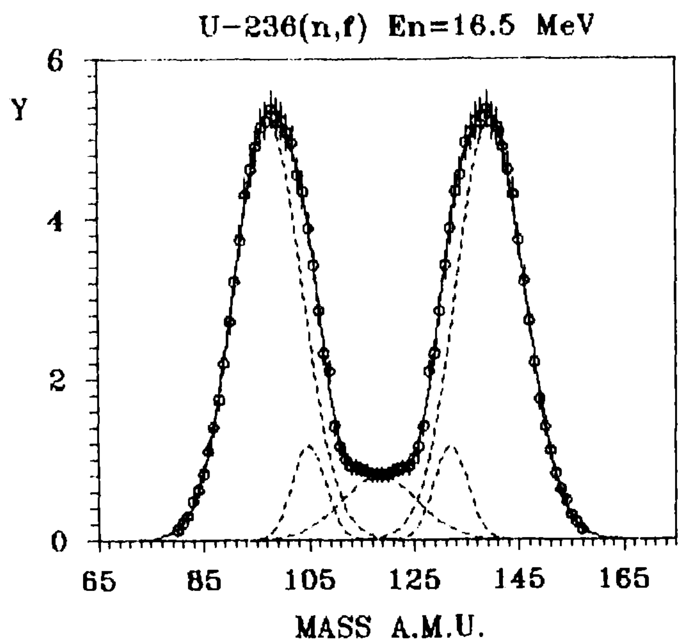


Fig.17. Mass distribution and its fit for $^{236}U(n,f)$ reaction.

The fitting parameters are presented in Table 2 , where: Wi, σi, Mi are the wight, width and first moment of Gaussian component i. The analagous parameters for separate reactions quite differ from each other. This is a reflection of the complex structure of fission cross-section, and consequently depends on the spectrum of fissile nuclei available after the neutron emission from the compound-system. Moreover, mass-spectra measured using 2E-method are very influenced by ν_p-values unknown as a rule for the separate fragments for high incident neutron energies E_n.

TABLE 2. Fitting parameters (E_n=16.5 MeV).

	^{243}Am(n,f)	^{237}Np(n,f)	^{236}U(n,f)
W1	9.9 ± 2.1	16.7 ± 3.1	14.1 ± 3.7
σ1	3.1 ± 0.5	11.7 ± 2.5	6.5 ± 1.7
M1	122	119	118.5
W2	10.5 ± 3.2	2.4 ± 0.7	8.2 ± 2.0
σ2	3.0 ± 0.4	2.3 ± 0.3	3.1 ± 0.6
M2	132.6 ± 0.2	134.2 ± 0.4	131.9 ± 0.5
W3	87.7 ± 4.7	85.4 ± 5.0	81.7 ± 5.0
σ3	8.9 ± 0.2	7.3 ± 0.2	6.6 ± 0.2
M3	139.7 ± 0.6	138.2 ± 0.6	139.4 ± 0.4

	^{238}U(n,f)	^{235}U(n,f)
W1	6.7 ± 2.0	7.6 ± 2.1
σ1	3.8 ± 0.5	3.8 ± 2.1
M1	119.5	118
W2	79.3 ± 5.2	65.2 ± 4.3
σ2	6.1 ± 1.5	5.9 ± 1.4
M2	137.5 ± 0.4	134.8 ± 0.5
W3	18.1 ± 5.2	31.3 ± 6.0
σ3	4.4 ± 0.4	4.7 ± 0.4
M3	147.4 ± 0.7	143.3 ± 0.5

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