# SELECTED ASPECTS OF FISSION OF ATOMIC NUCLEI

**Goverdovski A.A.**
(Institute of Physics and Power Engineering, Obninsk, Russia
gaa@ippe.obninsk.ru)

## INTRODUCTION

Nuclear fission is one of most strange and fantastic in nuclear world where one heavy nucleus decays onto two fragments of comparable masses. There are two principal kinds of that nuclear transformation. First is spontaneous decay and second one is particle induced fission. Competition of coulomb and nuclear forces gives unstable relative fission equilibrium and small affecting the system outside can lead to scission of the nucleus. Fission phenomenon was found more than 60 year ago and therefore has its interesting and dramatic history. It can be found in practically all nuclear physics notebooks. Here we will observe only induced fission preferably by neutrons from points of view of modern knowledge on a basis of recent theoretical models. In addition main attention will be paid to unresolved problems to show scale and interesting aspects of future investigations.

Thorium-232, one of isotopes involved into the thorium cycle has a biggest attractive scientific interest, representing nuclear phenomenon named 20 years ago as a “thorium anomaly” [1]. The last connects with a possible additional splitting of the outer hump giving so-called triple-humped fission barrier and respectively long lived exotic hyper-deformed nuclear states. Recent large-scale both experimental and theoretical studies of nuclear fission did not solve the problem. Simultaneously it has opened some new principal questions about a mechanism and time feature of a nuclear fission process, role of shell and pairing effects, fine structure of potential energy surface at high and super-high (hyper) deformations in wide region of nuclear temperature. Nucleus $^{232}$Th allows us to combine in one experiment a possibility to fix initial properties (shape and quantum numbers) of fissioning nucleus just before descent from a saddle point to scission one with observable variables like mass, charge, kinetic energy and excitation energy (via prompt radiations). To do this, it’s necessary to populate nuclear levels at the second or third minimum of fission barrier (see below), which are giving so-called beta-vibrational resonances in fission cross section. Coming away from the barrier top, one can change temperature regime of the process and to study time-dependent dynamical effects in fast flow of nuclear matter. Again, all fission properties both statistical and dynamical should be investigated simultaneously, in one “complete” experiment. In this kind of approach many physical aspects could be studied in detail. Thorium-232 nuclei are not unique in fission physics at present time, because few other nuclei have similar potential energy surface, which leads to the some kind of complication of fission feature observing at an experiment. It’s interesting, that the main part of those nuclei is involved into thorium-uranium fuel cycle. There are $^{230}$Th, $^{231}$Pa, $^{233}$Pa, $^{234}$U and $^{236}$U. Here, the main attention is paid to physical aspects of detailed and high-resolution investigations of fission barrier via series of fission cross-sections measurements. It does not mean that the problem of practically important neutron data will be suppressed. In contrary, it will be shown that high-accurate nuclear data can give a key to understanding of the principal questions of so difficult large-scale collective motion of nuclear matter like fission of atomic nuclei.

## PHYSICS OF VIBRATIONAL RESONANCES.

Simple demonstration of one-dimensional fission barrier is presented in fig1. Deformation axis $\beta_2$ corresponds to elongation of fissile system through fission direction. Population of states inside intermediate well leads to the effect known as shape isomerism [2]. Resonance penetration of the potential barrier below saddle points will give narrow resonance-like structures, observable if energy resolution is good enough. From experimental point of view this problem means the use of neutrons in the energy range below 2 MeV for thorium cycle isotopes. For Th, Pa and U nuclei very pronounced vibrational resonances have been observed between 0.1 and 2 MeV where the energy resolution is better than 2-5 keV. Simultaneous accurate measurement of fission fragment angular distributions give full set of JK- quantum numbers after so-called channel analysis of the experimental data. It’s important, that both excitation functions and total angular spectra will be determined with the same high-resolution against incident neutron energy. It would be first in the World practice attempt to sufficiently isolate the individual peaks of the substructure of vibrational resonances and therefore to deduce the moment of inertia of the rotational bands as precisely as possible and to determine their quantum characteristics. These spectroscopic properties of the vibrartional resonances may throw some light on the question of the thorium anomaly not resolved since the first half of 70-th.

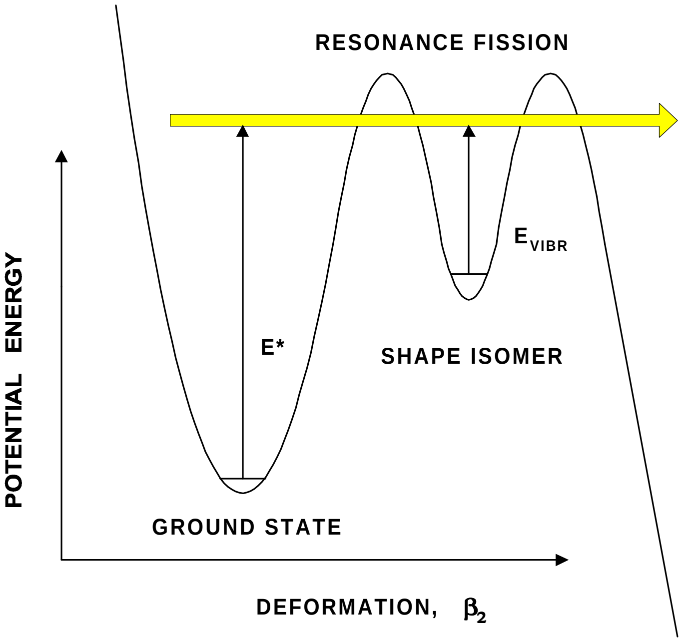

**Fig.1.** Schematic view of nuclear sub-barrier fission through β-vibrational state (thick arrow). $E_{VIBR}$ is characterized by depth and stiffness of the second minimum of fission barrier. $E^*$ - is excitation energy after neutron capture.

The theory of Strutinsky [3] introducing multi-humped fission barrier of the actinide nuclei has explained many resonance-like structures in fission cross-sections in fast neutron energy region. The weakly damped beta-vibrational states in secondary well of the potential barrier are responsible for those resonances and should be associated with a few-phonon state oscillating about the mean deformation of the secondary barrier well. The week damping due to the combination of the low excitation energy in the well and the inhibition provided by the first or intermediate barrier hump was observed at an experiment. Characteristic feature can be found in fig.2 where fission cross-sections selected for the thorium cycle isotopes are presented. Only near threshold part is shown. Usually observed dramatic changes in fission fragment angular distributions with changing excitation energy are due to dominance of separated vibrational resonances, each having definite spin-parity quantum numbers, and definite K numbers defined as projection of total angular momentum of deformed fissile nucleus onto fission axis. When there is no damping of the vibrational motion by the other degrees of freedom or it is not so high, the vibrational resonanse is rather pronounced since the respective transmission coefficient is close to unity with almost zero width. In reality only few resonances like this can be found. In fig.2 good examples of those resonances are seen for lowest resonances of $^{231}$Pa and $^{230}$Th (low damping). Usually damping is essential, but it has different magnitude for different fissile system. It depends on well's depth and excitation energy of nucleus.

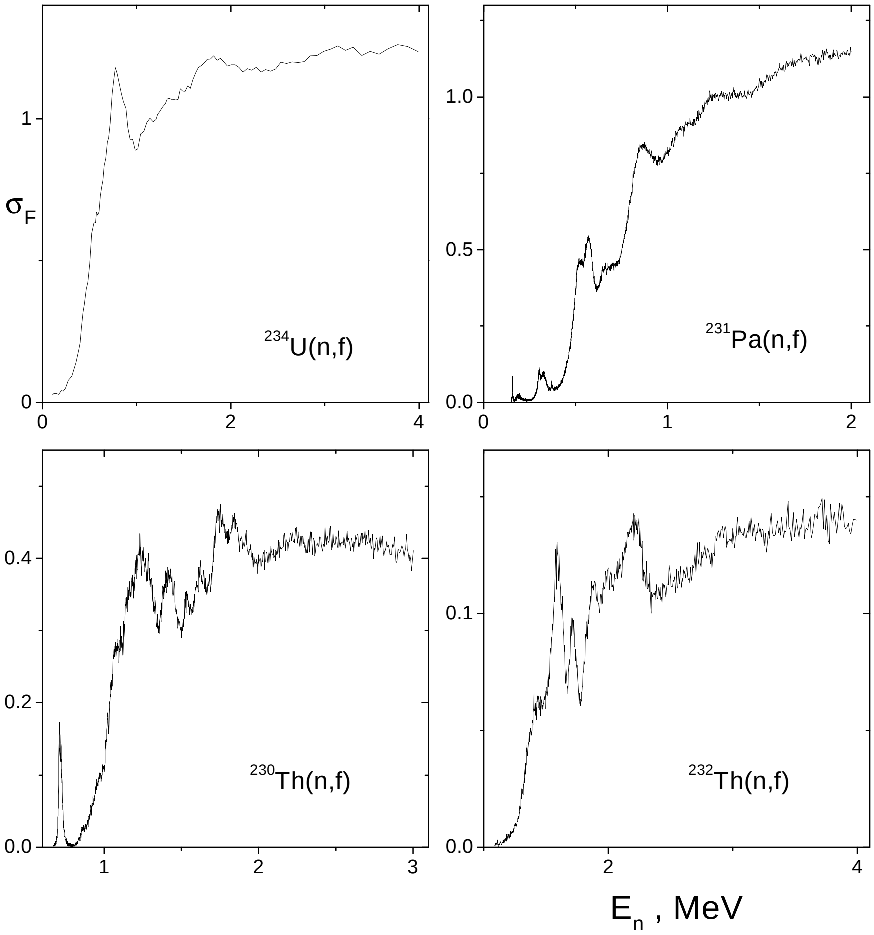

**Fig.2.** Vibrational resonances in fission cross-sections for thorium cycle isotopes. The data are taken from original works as follows: $^{234}$U(n,f) – [4]; $^{231}$Pa(n,f) – [5]; $^{230,232}$Th(n,f) – [6].

Large-scale experimental studies of near barrier fission probabilities by means of detailed high-energy resolution measurement of fission cross-sections with high enough statistical accuracy will give **new possibility for quantitative theoretical analysis of resonance structures** in wide region of excitation energy of the fissioning nuclei to determine precise positions of all of resonances associated with quasi-stationary states in the region of fission barrier top. Statistical fluctuations of the data available up to now as can be seen in fig.2 are too large to be neglected in the analysis and to come from hypothesis to self-consistent model.

Combining ideas of V.Strutinsky [3] and A.Bohr [7] with well-known Brosa-channels hypothesis [8], one can see, that the vibrational resonances have to appear in different intermediate barrier wells, if so-called bifurcation point of potential energy surface locates just before outer hump. Thus, physics of vibrational resonances is much more complex and difficult, than it was assumed in the late 80-th when intensive experimental and theoretical efforts were decelerated.

## DAMPED VIBRATIONAL RESONANCES. Reaction $^{234}$U(n,f).

The best studied example of a classical damped vibrational resonance was found in compound nucleus uranium-235 fissioning by fast neutrons in $^{234}$U(n,f) reaction [9] (see fig.3, upper part). The basic resonancelike feature at approximately 300 keV neutron energy was long believed to be explicable as competition between opening of fission and inelastic neutron scattering channels which was first assumed already in beginning of 60-th [11]. High-resolution studies were

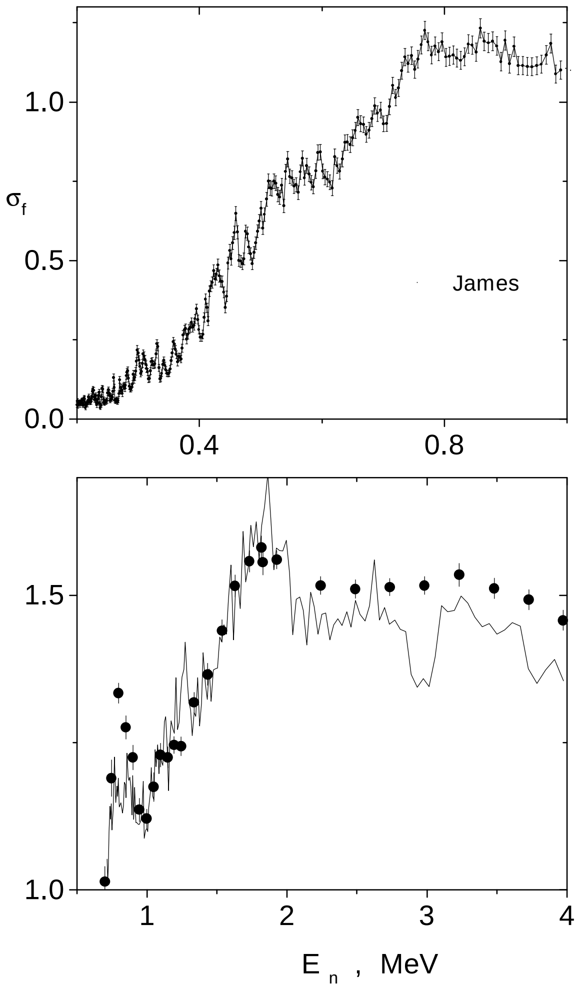

**Fig.3.** Upper part: fluctuations of data points of work by James e.a. [9] for $^{234}$U fission cross-section around "classical" damped vibrational resonance for neutron energy En = 330 keV. Bottom part: comparison with the data by Meadows [10] in the region of MeV-neutrons. One can see rather different resonance structures in both experiments, especially around well-known and good established resonance near neutron energy 780 keV.

performed first by James (1977) [9] to investigate so called class-II (associated with compound levels in the second well of fission barrier) intermediate structure within separate vibrational resonance. Due to relatively bad for this kind of structure energy resolution of 1 keV individual class-II structures could not be observed since the expected state spacing was about 0.2 keV. However, very considerable fluctuations of the data points through the vibrational resonance were observed. This was attributed to approximately 5 class-II states to be found in anyone resolution interval (see fig.3). The fluctuations of their strengths due to Porter-Thomas distribution expected to their partial fission widths were determined too. It was done taking into account intermediate structure of fission cross section for low energy neutrons. The barrier heights resulting from the combined analysis are different comparing with those required to ascribe fission cross section in higher energy region. Very probably, this is due to the unperfect and moreover not real behavior of vibrational resonance in both the coupling and fission widths of the class-II states in the parametrization adopted in the data analysis.

To solve the problem, one has to perform a set of additional measurements of fission cross section around energy 300 keV with much better neutron energy resolution. At the **CERN n_TOF** Facility for example the resolution at 200 m flight path will be approximately 0.06 keV ($\delta t$= 6 ns) which is good enough to observe separate class-II states. On the other hand, careful analysis of the data presented in fig.3 can give following conclusions. First of all, resonance-like fluctuations in data by James e.a. appear in whole energy region of incident neutrons including inter-resonance space (fig.3, upper part). Energy spacing of “resonances” is approximately constant in the region presented, and could be due to some kind of instabilities in fission fragment detection system, probably strongly influenced by accelerator driven neutron source. Anyway, the data must be confirmed in an independent experiment, and the “classical” damping resonance should be observed again or to be canceled. Next reason to repeat measurements of fission cross-section in $^{234}$U(n,f) reaction is obvious from bottom part of fig.3, where James's data are compared with results of the experiment by Meadows e.a. [10], done using the Van-de-Graaf accelerator with point-by-point measurement method. Lack of the pronounced resonance around the neutron energy 780 keV in data by James is rather symptomatic. Finally, fission cross section of uranium-234 should be measured with as higher as possible accuracy in wide region of excitation energy. Investigations of possible sub-structures of the resonances around 330 keV, 550 keV and 780 keV are needed for adequate explanation of fission barrier properties in uranium region, where the data from direct reactions give completely another feature of barrier shape.

## HYPER-DEFORMED NUCLEAR STATES. Reaction $^{231}$Pa(n,f).

One of the most interesting problems of classical low-energy nuclear physics is the searching for hyper-deformed nuclear shapes with a ratio of the long to the short axis of more than 3:1 and studying of their properties [1,12]. According to recent calculations the so-called third minimum of the potential barrier against fission appears in actinide region with deformation parameter $\beta_2 = 0.90$. One of the characteristic features of the hyper-deformed shapes of actinides, besides the large moment of inertia extracted from rotational bands bielt on beta-vibrational states, is the octupole bands connected with the respective kind of deformation around the third saddle point of fission barrier. Large quadrupole and octupole moments of the hyper-deformed states are reflected by existence of alternating parity bands with very high moment of inertia. This is rather convenient for experimental observations. Assuming overlapping of two rotational bands with the same moment of inertia (or rotational parameter), one can use relatively simple fitting procedure for resonance spectrum to derive the level scheme. Historically, first octupole deformed rotational bands were found and analyzed for fission of thorium isotopes in different reactions, both in neutron induced and in direct reactions. In further extensive studies the problem of the thorium anomaly was born. In addition, 20 years old idea to look for shape isomers in $^{232}$Th and neighboring nuclei was recently realized [13]. Expected decay by a strong gamma branch and relatively week fission was experimentally found in Grenoble. Detailed recent investigations with much improved equipment allowed to find third well in uranium nuclei directly excited in reactions with light charged particles. Fission probability of $^{234}$U was measured as a function of excitation energy in (d,pf) reaction. The rotational parameter was found to be $h^2/2\Theta = 2$ keV, which is characteristic for the hyper-deformed nuclear shape. Some evidences of analogous type of barrier parameters have been collected for plutonium. Further detailed studies with high-energy resolution of fission cross-sections around barrier top, and hence, fission probabilities for not heavy actinides can effectively contribute to the problem in question. Besides the thorium and uranium isotopes, double-odd nucleus protactinium-232 is interesting very much for the problem of hyper-deformation. Fission of this nucleus was explored in a series of measurements of the neutron interactions with $^{231}$Pa, including differential cross-sections or fission fragment angular distributions. Measurements of the fission cross section with high energy resolution have been made in several work using different neutron sources from nuclear explosion to LINAC driven neutron source (Muir e.a. [14], Plattard e.a. [5], and Sicre [15] – numerical data are not available). Considerable structures observed in fission cross section are presented in fig.2 and fig.4.

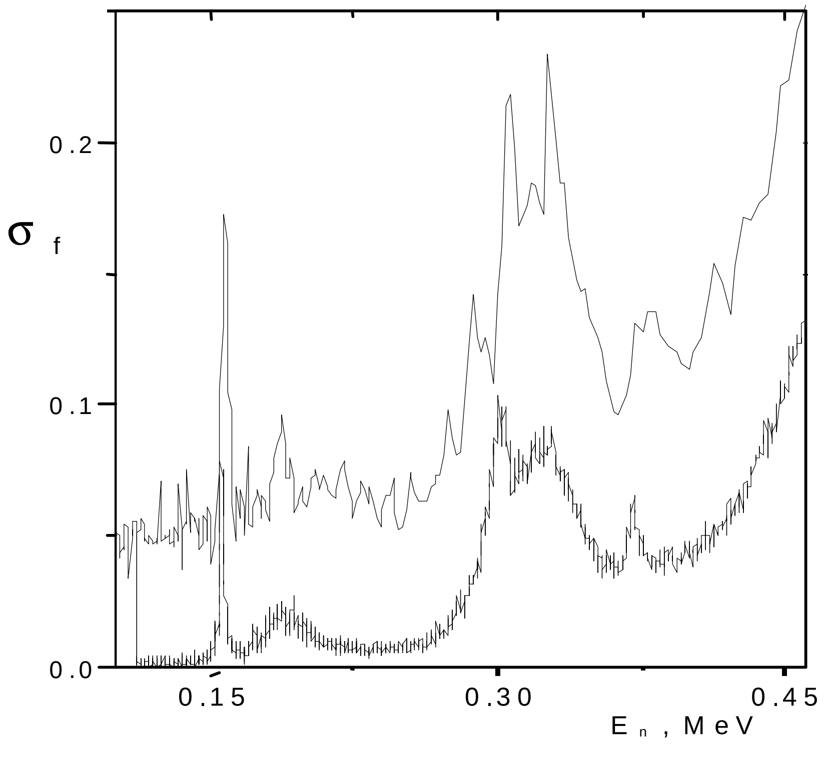

**Fig.4.** Comparison of fission cross-sections for $^{231}$Pa(n,f) reaction measured in two TOF experiments by Plattart [5] (bottom) and Muir e.a.[14] (top) for two vibrational resonances appearing around incident neutron energy 160 keV and 300 keV.

The analysis of the cross section and, more especially, angular distribution data giving quantum numbers of specific class-II states are complicated by the non zero spin of the target nucleus ($I^{\pi} = 3/2^{-}$). Hence the range of M quantum numbers extends from 2 to –2 and implies much less complex features in the angular distributions than in case of thorium fission, for example. At the same time, the rather low neutron energies for which the structures were observed limit the orbital angular momentum, probably for s- and p-neutrons, that could be effectively brought into the fissioning compound system. In spite of the noncomplex angular spectra it was found that the data could not be reproduced if assume the model based on a unique K-value for each vibrational resonance around 160 keV and 330 keV. Rotational bands based on $K^{\pi} = 2^-$ and $3^-$ for 160 keV resonance and $K^{\pi}= 0^+, 3^+$ for 330 keV were required. It's interesting, that the data were ascribed within the model of double-humped fission barrier. As usual , sufficient improvement of energy resolution up to 2 keV like in works by Plattard and Muir gave new look at the protactinium problem. The measurements reveal that the resonance around 160 keV does have in fact new weak substructure or narrow components with width and spacing considerably less than the experimental resolution of 2 keV. In addition to the main gross structure, observed early, this gave filling like in thorium. The overall picture for the odd-odd protactinium is consistent with a very shallow well (third well) of the potential barrier. According to groupings of intermediate resonances one can say that in a reflection symmetric fissioning system that could be excited by s- and p-neutrons the level density in the barrier well should be relatively high reaching 90 $MeV^{-1}$. Therefore, the superfine substructure should exist. In the experiment of Plattard e.a. [5] a non-Lorentzian behavior was found and this evidence would certainly be in favor of three-humped fission barrier hypothesis.

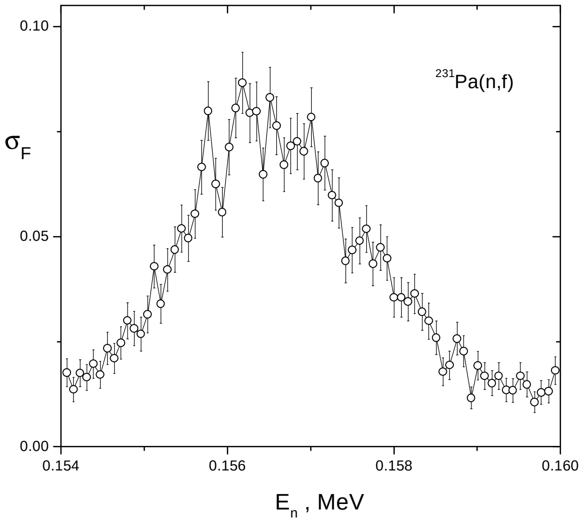

**Fig.5.** Reaction $^{231}$Pa(n,f). Detailed energy dependence of fission cross-section around the vibrational resonance En =156 keV which is in principle important for triple-humped fission barrier hypothesis (data by Plattard e.a. [5])

One should be stressed, that this conclusion is based on the one experimental work only, gross resonance data of which are in contradiction with another measurement as presented in fig4. First of all, the reproducing of fine structures between resonances is far from ideal. Muir e.a. [14] have reported existence of strong irregularities in fission fragment angular distributions. This is typical reflection of presence of resonances in the barrier penetration function. On the other hand, statistical uncertainties in work of Plattard, as can be seen in fig.5, are rather large to give definite conclusions about possible rotational band parameters. Analogous fluctuations of fission fragment yields, or total fission cross section, are obvious in whole sub-barrier energy range investigated (fig.2, right top, fig.4). Therefore no conclusive evidence for a triple-humped barrier emerges from the (n,f)-reaction data. Further detailed studies with the aim of establishing near degeneracy with states of opposite parity in the rotational bands will be productive and perspective. In addition, the problem of adequate interpretation and analysis of other vibrational resonances stays to be far from solving.

## PROTACTINIUM EFFECT. Reaction $^{233}$Pa(n,f).

Protactinium-233 is one of main isotopes of thorium-uranium fuel cycle. Its importance in operation of nuclear facilities like fission reactor or accelerator driven system (ADS) like Rubbia's "Energy Amplifier" (EA) connects with two aspects, known as a "protactinium effect". It means the following: first, decreasing of the reactivity of the reactor due to capture of neutrons and, second, increasing of the reactivity after reactor stop due to inventory transformation of $^{233}$Pa into $^{233}$U via β-decay with half-life equal to 27 days only. Therefore any knowledge of $^{233}$Pa amount which depends on fission and capture cross-sections is essential. As a rule, double-odd protactinium isotopes have neutron binding energy less than fission barrier height [1], and therefore fast neutrons can only give fission while radioactive neutron capture is effective for slow neutrons and negligible in fast (>0.1 MeV) region. Thus, both cross-sections – fission and capture – should be investigated separately and in different energy ranges.

Short lifetime of $^{233}$Pa determines extremely high radioactivity of the corresponding targets connected with principal limitations of experimental studies. Up to now one only measurement of fission cross-section was done with relative accuracy 26 % [16]. Thermal reactor was used as a neutron source with $^{233}$U converter of thermal neutrons into fission prompt neutrons with average energy equal to 1.5 MeV. Unfortunately thorium-232 was used as a neutron monitor. Rather developed resonance structure of $^{232}$Th cross-section determines observable fission yields with average cross-section less than 100 mb in neutron energy range 1 to 2 MeV where the main part of the incident neutron spectrum is localized. This is much lower than 142 mb taken as reference (monitoring) cross-section in the data processing. Very

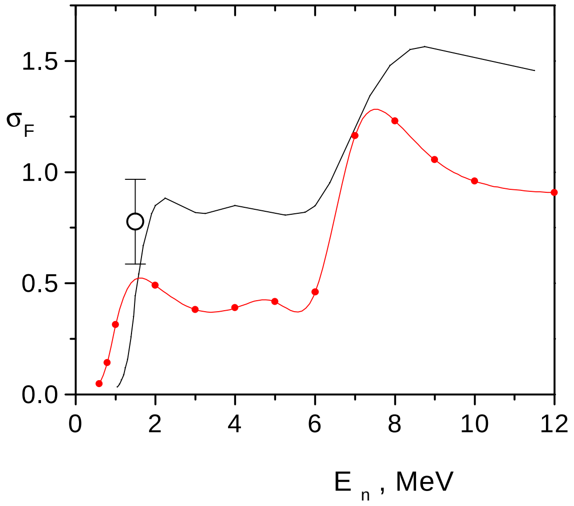

**Fig.6.** Fission cross-section data for the reaction $^{233}$Pa(n,f) [16] : O – experiment on Maxwellian spectrum of prompt fission neutrons of $^{233}$U. Curves are evaluated values from ENDF and JENDL-3.2 data libraries.

probably, that the value of cross-section should be changed for 40 %. Anyway, experimental data for fast neutron induced fission cross-section of $^{233}$Pa look so uncertain that they are not able to satisfy accuracy of 20% required for practical applications. Evaluated data available from the so good developed and adopted files like ENDF-B/VI and JENDL-3.2 distinguish to each other on a factor more than two. This situation is presented in fig.6. One can see differences in near-barrier region and especially above the threshold of penetration function. Detailed analysis of systematic trends in a fissility parameter (equal to the half-ratio of coulomb energy to surface energy) and fission probability of neighboring protactinium isotopes gives expected level of the cross-section magnitude in so-called first fission plateau somewhere between two evaluated lines. Again, one can conclude that both empirical and evaluated data for $^{233}$Pa(n,f) are not available with accuracy needed.

Experimental way to solve the problem of the "protactinium effect" is hardly perspective, hence some semi-empirical and theoretical approaches should be developed. This can be done on the basis of self-consistent model including detailed and careful determination of fission barrier parameters, level density, optical cross-sections etc. Fortunately, few fission data are available for 4 protactinium isotopes $^{230-233}$Pa derived from neutron-induced reactions $^{231,232}$Pa(n,f) [5,17], photo-fission $^{231}$Pa($\gamma$,f) [18], and charge particle induced reactions, like (3He,df), (3He,tf), (d,pf), (t,pf) [19,20]. Experiments cover excitation energy range of fissioning nuclei from 4 to mainly 11 MeV. Link from the nuclei and reactions indicated above to the reaction $^{233}$Pa(n,f) is not so complex, if fissile systems with all of possible nucleonic compositions will be ascribed for all reactions studied up to now. Good tested computational approach could give more accurate data than those available now. In this respect the question of fission barrier structure especially the problem of the barrier saddle point multiplicity is very important for so light actinide nucleus like protactinium. Obviously, success in getting the accurate data for the $^{233}$Pa(n,f) reaction strongly depends on new precise and high resolution data for $^{231}$Pa(n,f) reaction, which importance is underlined again.

## 2. BIFURCATION POINT OF MASS ASYMMETRIC FISSION VALLEY AND THORIUM ANOMALY PROBLEM.

Modern ideas of multi-valley landscape of fission barrier developed and used in the data analysis of the practically all experiments in fission physics for past 15 years, open completely new approach for an interpretation of observable data. If so-called bifurcation point where the mass-asymmetric fission valley is splitting into two independent parts really exists just before penetration of the barrier, it must be represented as a set of two separate fission barriers. Then, fission cross-section will be the sum of two components, associated with two standard fission modes with respective spectra of intrinsic states giving in exit channel of reaction probably different angular distributions. At the experiment it would be observed as pronounced fluctuations of shape of fission fragment mass distributions in correlation with FF angular spectra. Strong correlation or probably functional dependence of final properties of fissioning nucleus on initial conditions at latest pre-fission deformations of compound nucleus means an existence of some kind of nuclear memory.

Theoretical ideas of the character of heavy nucleus fission that were developed last two decades are based on the predicted multi-valley structure of the potential barrier [8]. The fission modes are considered to be due to the existence of well-separated valleys on the potential-energy surface of a fissile system. Motion over these valleys results in the structure of the kinetic-energy and mass distributions of fragments. There spectra are dominated by the components in the neighborhood of the heavy-fragment masses of $m_H$=134 and 140 amu. They are referred to as the first and second standard fission channels (modes), denoted as $C_1$ and $C_2$, respectively. Usual description of fission fragment mass distribution as a superposition of two or more Gaussians with centers around masses 134 and 140 a.m.u. is shown in fig.7 (heavy fragment parts of the spectra are shown for convenience; light parts are mirrow-reflected relative to M/2 axis). If each of valleys can be characterized by its own outer barrier, the yields of the standard components $C_1$ and $C_2$ must display variations in the near-threshold region, which leads to changes in the mean total kinetic energy. Angular correlations of fragments have also to exhibit a mass dependence. The first step along these lines was done by Hambsch e.a. [21], who studied the mass-energy distributions of fragments produced in $^{235}$U(n,f) reaction induced by resonance neutrons. A pronounced dependence of the $C_1$ and $C_2$ yields on the fission width of the resonances was observed. Scale of the effect was found to be approximately 10-20%. Recently these data were confirmed in Dubna by Furman e.a. [22]. Analogous characteristic redistribution of the $C_1$ and $C_2$ contributions to the total mass spectrum was revealed also in studying of $^{238}$Np [23] and $^{244}$Am [24] fission in the near-threshold region. For reaction $^{236}$U(n,f) it was found that the angular distributions of fission fragments have pronounced mass dependence; this is a compelling argument in favor of the Brosa model. Finally, it was found in a number of studies that, in fission of thorium and uranium isotopes, the mean total kinetic energy of fragments (TKE) fluctuates in the immediate vicinity of the reaction threshold. It was proven experimentally that the local variations of TKE are due to the β-vibrational barrier-penetrability resonance associated with a quasi-stationary state in fission barrier well [1], as discussed above. The present-day concept of the potential barrier requires [8,29] detailed analysis of the mass-

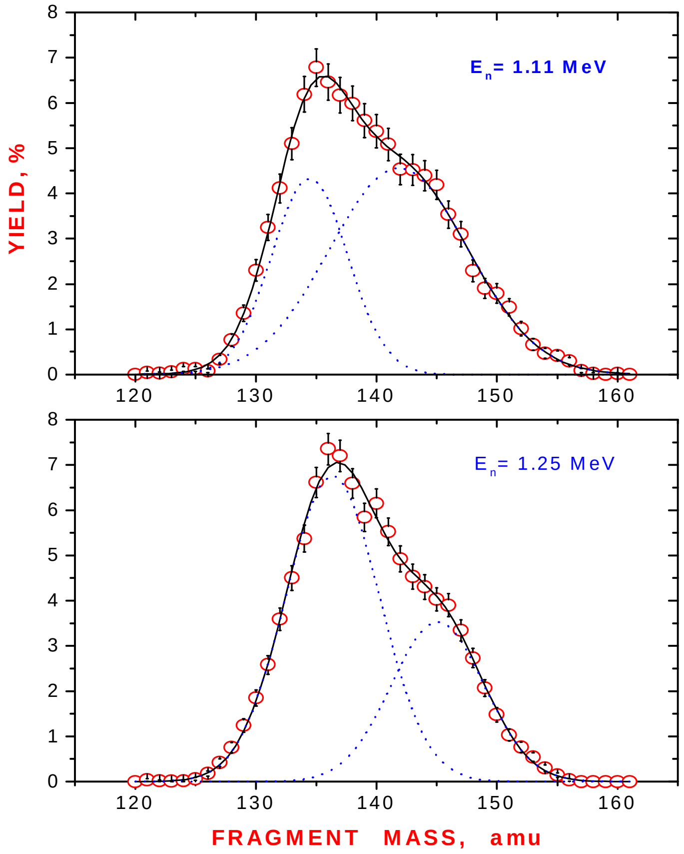

**Fig.7.** Fission fragment mass distributions in reaction $^{238}$U(n,f). The spectra were fitted with Gaussians using 5 free parameters - relative yield, 2 dispersions and 2 mean values. RMS analysis was done.

energy correlations when vibrational resonance plays main role in barrier penetration. Schematic view of fission through β-vibrational state (thick arrow) is shown in fig.1. Existence of the β-vibrational state in intermediate minimum (second like in uranium or third like probably in thorium) leads to resonance structure in fission probability. First successful attempt to solve this problem was made in Obninsk by observing strong – up to 50% - variations of $C_1$ from one resonance to another in fission of thorium-232 by fast neutrons. Probably, those resonances should be associated with vibrational states and corresponding rotational bands members of hypothetical third well of thorium fission barrier with extremely small depth. Energy resolution in experiment was rather bad and hence strong overlapping of the resonances gave in that case complex very much feature which was hard for theoretical analysis. Nevertheless, fluctuations of standard-I component yield were pronounced enough to extract partial fission probabilities for

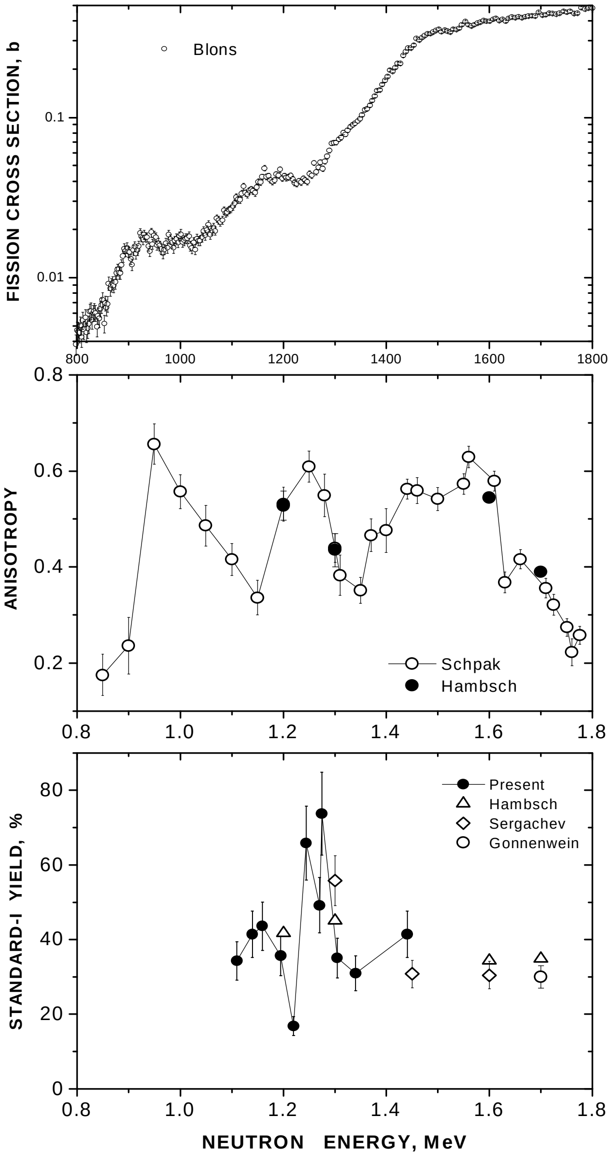

**Fig.8.** Yield of Standard-I mass channel in correlation with fission cross-section [31] and anisotropy of angular distributions [32,33] – {Y(0°)/Y(90°)-1}. Data for $C_1$ are taken from: Δ - [34], ◊ - [35], O –[36]. • - [30]. In work [35] semiconductor detectors were used and in works [34,36] – the Frisch grid ionization chamber with internal energy calibration.

both standard modes. Recently [30] $^{239}$U-fission in deep sub-barrier region of excitation energy was chosen to look for analogous to thorium effects for highly separated resonances associated with the states in second, relatively deep, well of the potential barrier. In that case the authors expected much higher scale of mass spectra variations than in previous experiments. In addition, large optimism was due to results of novel calculations of potential surface of compound nucleus $^{239}$U done in Geel by Hambsch and co-workers. It was shown, that the bifurcation point of the mass-asymmetric fission valley locates just before outer saddle.

Experimental mass spectra with decomposition onto Gaussian components are presented in fig.7 for two neutron energies. Differences in spectra are obvious. Analogous data processing was done for all spectra in 9 energy points. Results of determination of $C_1$ yield as a function of incident neutron energy is presented in fig.8 together with fission cross-section [31] and anisotropy of fission fragment angular distributions [32,33]. Fluctuations are observed on the level of 150 % (!). This means that the vibrational resonance around $E_n$ = 1.25 MeV is completely determined by Standard-I component. Dramatic change of mass spectrum around resonance is much more pronounced in fig.9 (right part) where the data are shown in wide region of excitation energy up to threshold of emissive fission (n,n'f). The scale of the resonance effect in mass distribution is much higher in the case of $^{238}$U than those for all other cases (one of them is shown in fig.9 on the left part for thorium-232) investigated and described above. It could be due to larger freedom in differences of outer barrier parameters connected with standard fission modes. Coming back to the experimental data, one should stress that the agreement between the data of different authors is practically perfect where the comparison can be done.

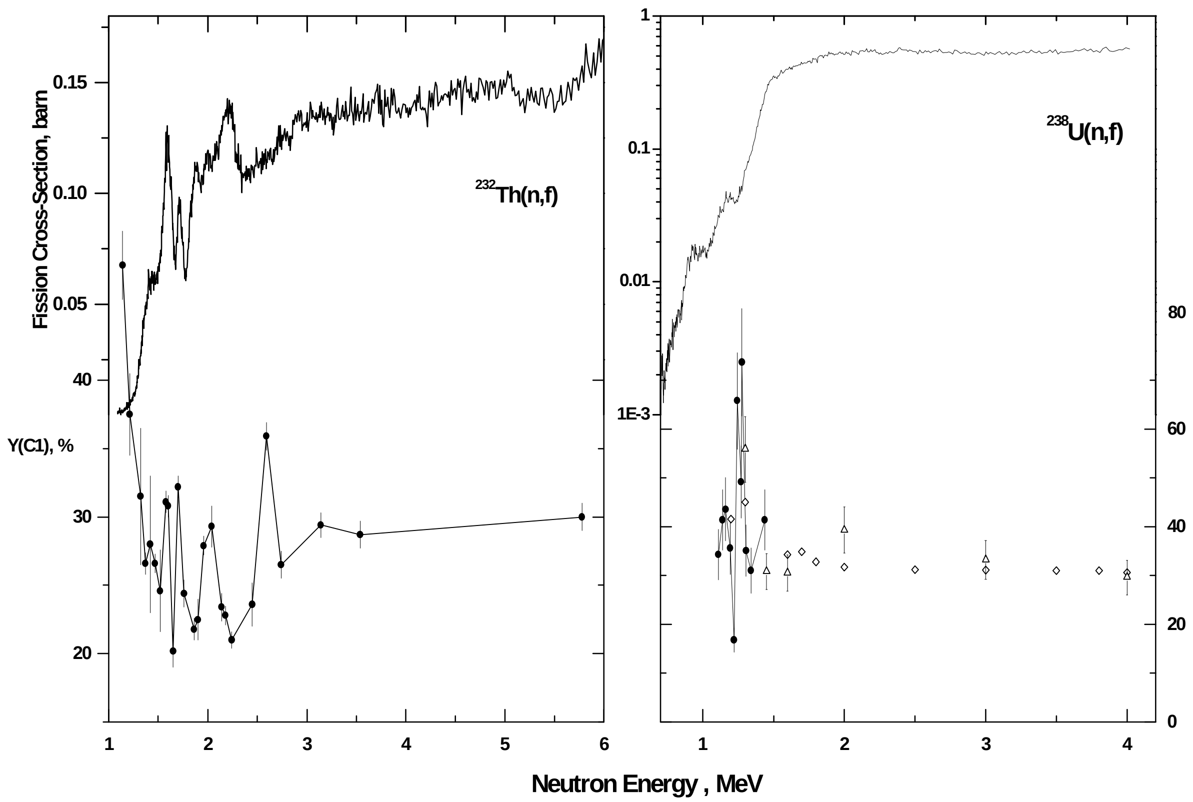

**Fig.9.** Same as in fig.8 but for wide region of neutron energy below and above fission barrier for two different reactions – $^{232}$Th(n,f) [37] (left) and $^{238}$U(n,f) [31] (right). See carefully scale of the variations in both cases.

The work performed for $^{238}$U(n,f) reaction in Tubingen [36] with ionization chamber was focused on the searching for mass-energy and emission angle correlations in analogy with the studies of $^{236}$U(n,f) reaction [37]. No mass or kinetic energy dependencies of fission fragment angular distributions were observed. Looking at the fig.8 and 9 one could explain that "negative" result. Starting energy point in Tubingen (1.4 MeV) was too far from vibrational resonance for which correlations take place. On the other hand, obvious and good established lack of the effect in the region where fission anisotropy is relatively high (fig.8) can be understood in the framework of Brosa-model by following way. Not impossible, it appears in the case of uranium-239 that the last β-vibrational state corresponding to incident neutron energy $E_n$= 1.45 MeV locates in both wells of barrier (Standard-I and Standard-II) with the same positions. If resonance around neutron energy $E_n$ = 1.25 MeV belongs to $C_1$ mass channel as we just presented in present work, it would be interesting very much to study in detail fission fragment mass distributions around next resonance $E_n$ = 0.95 MeV where the angular distributions are focused along $0^o$ relative to incident neutron direction [32]. Perhaps, main component there will be Standard-II with relatively low mean total kinetic energy. From experimental point of view, two last resonances in fission probability of compound nucleus $^{239}$U are unique objects of fission physics. For them full information like mass, kinetic energy and angular distributions should be obtained. Some special role can be played by difficult but at the same time very informative spectra of cold fragmentation of nuclei through vibrational resonances and far from them. Of course, Bragg ionization chamber is one of the most suitable detector for these purposes. In addition, detailed investigation of any fission properties near the low-laying sub-barrier resonances needs high enough energy resolution. **CERN n-TOF** Facility with expected resolution of 2 keV for 1 MeV neutrons looks the best as a neutron source.

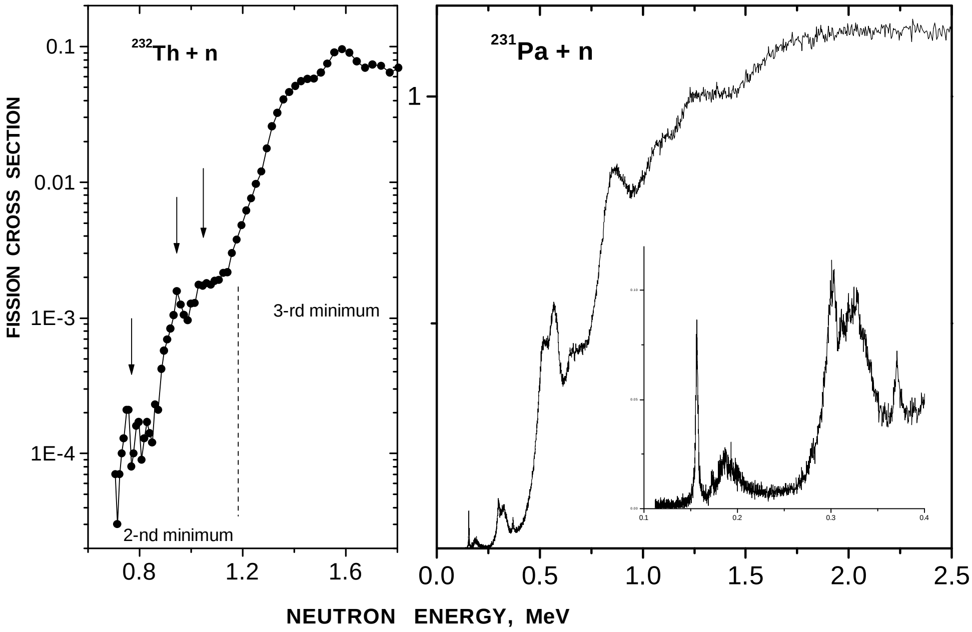

**Fig.10**. Left part: Fission cross-section of $^{232}$Th [38] in deep sub-barrier region of excitation energy. Possible class-II resonances are indicated with arrows (see text). Right part: fission cross-section of $^{231}$Pa [5]. Insert is for the data below 0.4 MeV

Physical prospects of investigations of multi-valley structure of potential energy surface of high and hyper deformed atomic nuclei and determination of their spectroscopic parameters could be found in an expanding of experimental program to other fission reactions, for example $^{234}$U(n,f) and $^{231}$Pa(n,f). As it was shown above in excitation function of compound system $^{235}$U fission the several good pronounced vibrational resonances with relatively high cross-sections up to 1 barn were observed for neutron energies 330 keV, 550 keV and 780 keV [9]. It's interesting, that corresponding angular distributions are in principle different for different resonances giving both positive and negative anisotropy [11]. Careful investigation of fission fragment mean total kinetic energy around those resonances gave possibility to conclude that the correlation between FF properties and fission barrier structure do exist [27]. In fission cross-section of protactinium fine structures and good separated resonances were observed by Plattard e.a. [5]. In fig.10 (right side) a set of those resonances are picted (like in fig.2, but simultaneously in all of interesting energy regions). Vibrational resonances are pronounced and convenient for investigation of multimodal fission. There are both good isolated and multiple resonances. This looks like overlapping of thorium and uranium resonance structures, but taking into account relatively high cross-section of protactinium it looks the best candidate for future studies. In addition, Sicre e.a. have reached in their work [15] the perfect theoretical description of all of low-laying resonances in assumption of double-humped structure of fission barrier (unfortunately data are not available in digital form). In general, perspectives to study peculiarities of fission modes (Brosa-modes) in resonance fission of odd-odd compound-system are promising very much.

The works with thorium-232 as well as uranium-236 (target nuclei) are not completed yet. As to thorium, one specific trend in data points for Y($C_1$) as a function of neutron energy can be seen in fig.9 when the energy is decreasing. Average amplitude of Standard-I yield's fluctuations is approximately 50 %. But for lowest energies Y($C_1$) increases very rapidly in the limits of more than 100% (!). This is scale of fissile nucleus $^{239}$U, in which the state in the second well of the fission barrier is populated. One can suggest that for thorium the resonance around neutron energy close to 1,1 MeV (fig.10, left side) should be attributed to the second but not to third well of the barrier. This hypothesis seems to be very helpful for solving of long-standing problem of thorium anomaly. Unfortunately, fission cross-sections are small and energy resolution is not perfect, and therefore the practical measurements need much efforts and patience. From this point of view, an additional source of information we are interesting in can be found in studies of the charge particle induced fission reactions like (d,pf) and (t,pf). Probably, they contain many new effects expected and unexpected. Another alternative and rather optimistic way can be found obviously in use of high-flux and high energy-resolution source of fast neutrons - **CERN n_TOF**- facility. In this case the problem of angular momentum as the problem of high efficient detection systems can be solved very easy due to the perfect neutron flux collimators.

## 3. RARE FISSION PROCESSES.

There are at least two most interesting and in principle different rare binary fission phenomena – cold fragmentation and mass symmetric fission. The first class of effects can be observed for maximum FF total kinetic energies (TKE) or small free energy of the system and therefore super-compact scission configurations. In contrary, super-elongated scission configurations for the fragments in long-lived extremely deformed states can only be found for anomalous low TKE. The problem of the energy transformation from the internal degrees of freedom to collective ones and back attracts deep interest for not only fission physics but also for general nuclear physics as a part of science. Total relative yield of cold fragments in binary fission is approximately $10^{-4}$ - $10^{-6}$, therefore experiments are as a rule long and need careful performance. Taking into account high flux of nuclear reactors and relatively large fission cross-sections, thermal neutron induced fission reactions were investigated [39]. Set of data on cold fragmentation of even-even and odd-odd compound nuclei is presented in fig.10. Mass spectra of heavy fragments are only shown [40]. For even-even compounds rather pronounced structures are seen good. Centers of the components resolved at the experiments are localized around the heavy masses near 134, 140, 146 and 152 a.m.u., for all of fissile systems, which is important. In contrary, odd-odd nuclei give spectra of cold fragments with much less pronounced structures, almost flat. Probably, proton and in less degree neutron pairing effects a responsible for these differences. Longstanding questions are: what is the mechanism of rare creation of large nuclear clasters in practically ground states in fission, when the dominant part of the fragments are deformed and excited ? What is a response of cold fragments to slow heating of the system by means of increasing the excitation energy of initial nucleus? It could be heating or population of other ground states on the potential energy surface, and hence creation of exotic shapes, unknown yet. This process depends on mechanism and magnitude of nuclear viscosity. Looking at the figure, one can conclude that cold fragmentation mass spectra are sensitive very much to profile of potential energy surface around the top of fission barrier. Therefore all resonance effects in the penetration function will be reflected in CF properties. Thus, cold fragmentation can give new tool for studies of many features of collective nuclear motion.

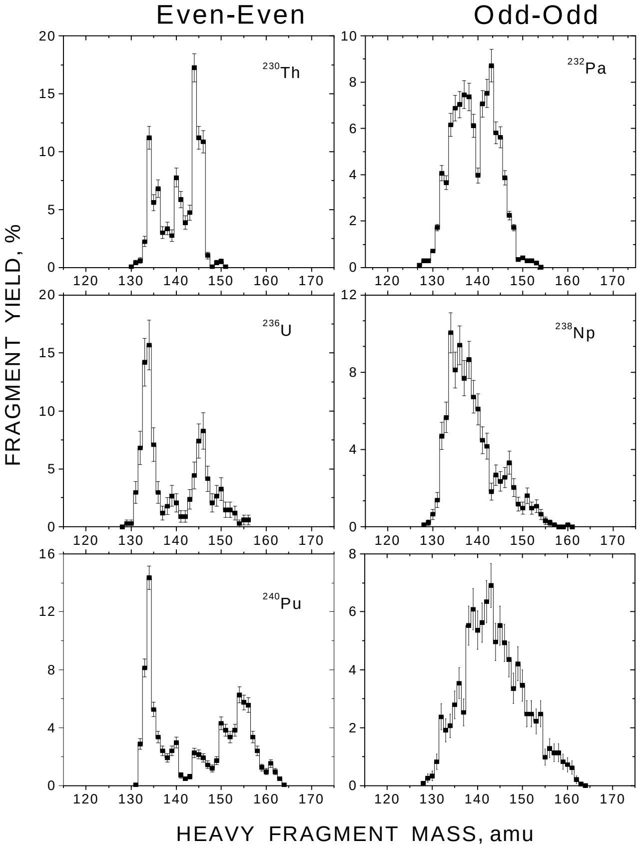

**Fig.11**. Mass spectra of cold fragmentation products, observed in thermal neutron induced reactions, giving double-even and double-odd initial compounds.

Completely rare phenomenon like so-called cold fragmentation or cold compact fission is a process when all of free energy collected in the system is transformed into kinetic energy of fission fragments. In this case two nuclei are in their ground states. More probably the process can exist for spherical nuclei or at least one of them with spherical shape. An example can be seen in fig.12 where novel data for neutron-induced fission of thorium-232 are presented. The work was done with Bragg ionization chamber to get nuclear charge spectra in addition to mass and kinetic energy. One can see very sharp decrease of yield of separated nuclear part $^{134}$Te-$^{99}$Sr with increasing of total kinetic energy. At some point corresponding to reaction Q-value the yield comes down almost to zero showing some kind of energy "wall". No events can exist on the right from Q-value. Experiment showed this.

Experimental possibilities available at modern neutron facilities like **CERN n_TOF** Facility or **S**pallation **N**eutron **S**ource (SNS) in Oak-Ridge allow to do a set of new works. Main goal from the point of view of problems in question is to observe cold fragmentation of excited nuclei, to determine extreme deformations of two-centers fissioning figures, for which the initial internal excitations of the compound system are steel spent for creation of nucleus's surface with almost spherical or other, perhaps exotic, shape. Another interesting and important effect is a fluctuation of CF yields around vibrational resonances. Possibility to observe K-quantum number non-conservation effect is more or less perspective. It could be interesting although from the point of view of testing of A.Bohr's model of intrinsic states. The best objects for studies are thorium cycle isotopes $^{229}$Th, $^{230}$Th, $^{232}$Th, $^{231}$Pa and $^{234}$U. CF spectra and total yields for some of them are unknown.

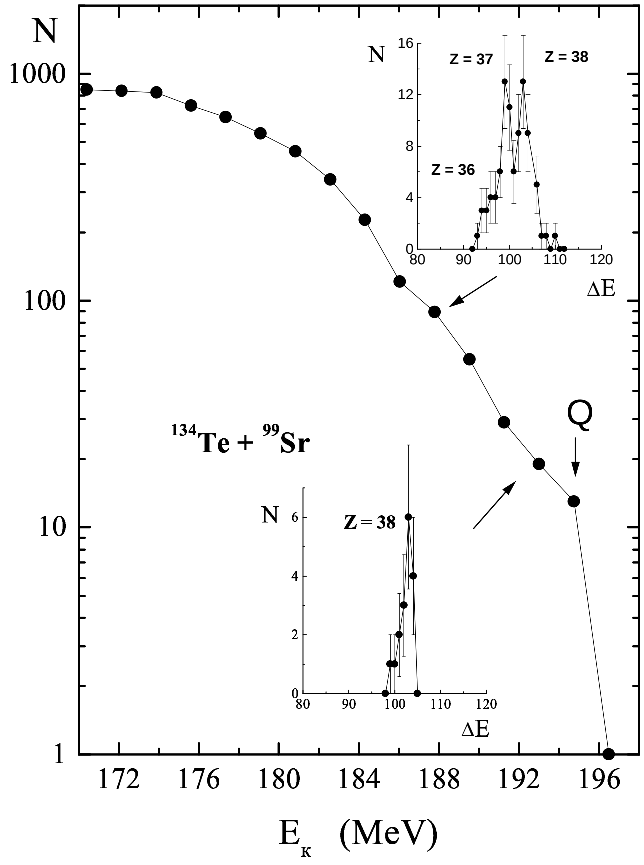

**Fig.12.** Observation of true cold fragmentation of thorium onto two completely not excited nuclei – Te-134 and Sr-99. Here Q – is reaction heat. Bragg analysis showed single nuclear charge in the vicinity of limited total kinetic energy.

Second class of rare fission processes of actinides - symmetric fission - is highly suppressed fission mode in the thorium region. Corresponding fission barrier is large comparing to asymmetric one. Hence the problem should be represented in following way – does this mode really exist in cold nucleus as well, and what is the mechanism of barrier penetration when the asymmetric barrier has dominant penetration?

Selected mass-spectra where symmetric components can be definitely extracted are presented in fig.13. Three reactions $^{229}$Th($n_{th}$,f) , $^{232}$Th(n,f) and $^{235}$U(n,f) are used. Small mass of the complementary light fragment determines relatively big distance between two asymmetric humps. In addition, asymmetric components are narrow enough. Therefore, symmetric part is not hidden by their overlapping. General trends in fission fragment mass distributions with excitation energy were intensively investigated for past four decades. But in only few experiments real symmetric fission was observed. Mostly there were radio-chemical studies. In fact it was known that during the heating of the fissile system two-humped mass curve transforms into single-humped distribution. In these conditions it was impossible to determine parameters of the mass-symmetric barrier, like height and width. Peak-to-valley ratio is completely not enough. Recent microscopic calculations performed by J.-F.Berger [29] showed that for thorium isotopes the barrier height is larger than 15 MeV. It means that symmetric fission should absolutely negligible relatively to asymmetric one for which barrier is not higher than 7 MeV. But nevertheless experiment gave observable effect on the relative level of $10^{-3}$. One can expect small width of symmetric barrier, or existence of alternative by-pass of the barrier. The last idea

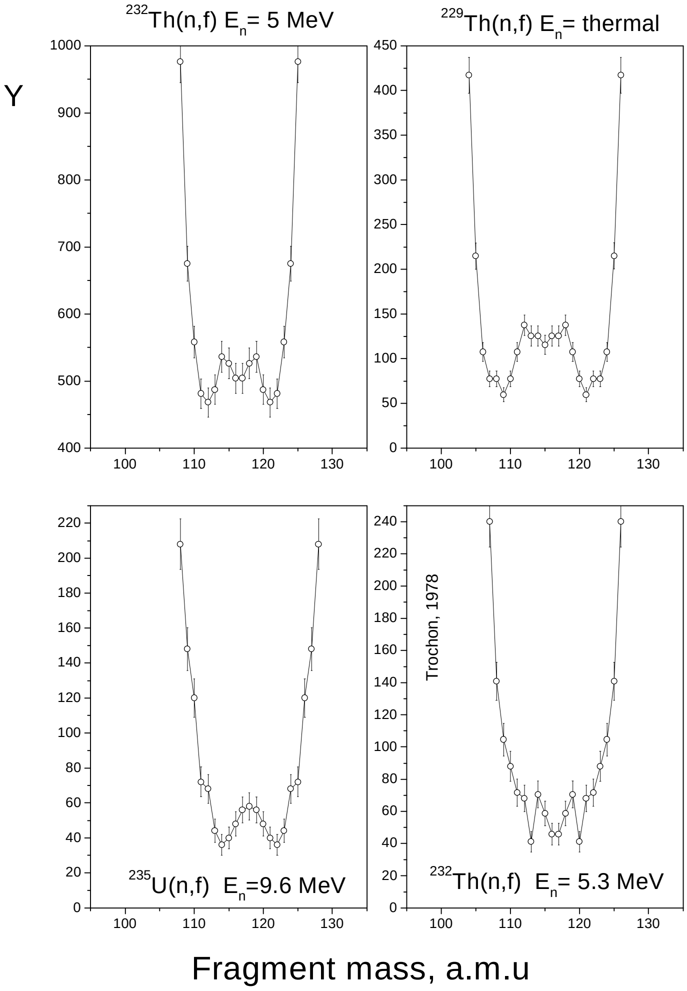

**Fig.13.** Pronounced mass-symmetric fission fragment components observed in few neutron- induced reactions. Fission fragment spectrometers were used in all cases.

was proposed recently by Pashkevich [41]. Anyway, the problem of mass symmetry in fission is now in the stage, when only experiment can give accurate answer. Measurements of differential fission cross-section of light actinides through mass-symmetric fission channel would be rather important in wide region of neutron energy. This range should cover area of vibrational resonances, where fluctuations of the symmetric component yield will show, is the corresponding barrier well analogous to those in asymmetric modes or not. In the region of higher excitations, let say up to 30 MeV, it's interesting very much to find the area where the symmetric fission barrier top is reached. Corresponding excitation function contains whole information about shape of the barrier. For neutron energy above 30-40 MeV one can expect, that open symmetric channel will play comparable with asymmetric channel role. Therefore, total fission cross-section will be superposition of two independent components characterized by separate barriers with different nuclear level densities, determined by the shape symmetry of the fissile compound nucleus together with excitation energy and deformation. This is important for description of fission cross-sections in wide region of neutron energy, even up to 200 MeV. It appears again, that the thorium cycle isotopes are most convenient for experimental studies and theoretical analysis mentioned above because their fission cross-sections do not reach maximum values in very wide region of energy and respective so-called multi-chance structure of the cross-section is good pronounced up to 30-40 MeV. Nuclei like thorium-232 and protactinium-231 are very representative in this respect.

Coming back to fig.13, the following additional specific feature can be observed. Mass-symmetric Gaussian is splitted into two components in fission of thorium and it is structure-less in the case of uranium. Spacing of that sub-structure is almost 6 a.m.u. The following hypothesis can be formulated. In low-energy nuclear fission pairing of nucleons, especially protons, is essential. It leads to well-established so-called local odd-even proton effect in fission fragments yields [42]. This means preferable formation of fission fragments with even number of protons comparing to odd number. While the excitation energy is increasing, odd-even effect is decreasing due to proton pairs breaking. Thus, magnitude of odd-even effect could be used as a tool for determination of nuclear temperature of fissile system. In thorium odd number (90/2=45) of protons is distributed to each fragment in symmetric region. Charge division 44/46 (in charge units) is much more probable due to even-odd local proton effect. Therefore structure in mass curve appears. Obviously, neutrons with the energies above 10 MeV will give fissile compounds heated just above the symmetric fission barrier, and structure in question will be eliminated.

One has to note that odd-Z fissile systems like Pa-231+n should be investigated in much more detail than it was done before. Very accurate recent studies of the nuclei of this class showed definite existence of local proton effects in far-asymmetric region of the mass curve and in cold fractions of fragments in neptunium and americium fission. The origin of odd-even effects in the fission fragment yields has been interpreted [42,43] by assuming a preservation of superfluidity in the fissioning system on its way from saddle point of the barrier top to scission one. Fissioning nuclei with odd-Z were not expected to show an odd-even effect because of the presence of an unpaired proton. Earlier experiments with $^{239}$Np in exotic reaction $^{237}$Np(2n,f) confirmed this expectation [44]. But later with much more high statistics the evidence for the proton odd-even effect was found in the fission of slightly excited americium $^{243}$Am$^{*}$ [45]. Novel investigations performed by large international group in Grenoble [46] complitely repeated in more detail that evidence. A preferential formation of light fragments with even nuclear charges was found in the very asymmetric mass region. The value of the odd-even effect for protons was found to increase with the asymmetry of the mass split up to 30%. It appears that the effect found is comparable to the one for even-Z nuclei like plutonium. The increase of the effect hints to a proportionality between the probability for the unpaired proton to end up in the heavy complementary fragment. Analogous effect was observed recently in Obninsk, one time in spectrum of cold fragmentation by fast neutrons of $^{238}$Np$^{*}$, and second time directly in charge spectra of cold fragments with moderate mass-asymmetry. The last data are represented in fig.14. There the local proton odd-even effect reaches 40%! In systematic large-scale studies in experiments with inverse kinematics made in Darmstadt [47] the idea was born that the attachment of the unpaired proton to one or to the other fragment is governed by the phase space available. This phase space is directly proportional to the number of nucleons contained in the fragment or to its mass. In very cold region when only few excited states are available the picture could be completely another. In addition, it is not so clear, why near-double-magic spherical nucleus-fragment around ${}^{134}$Sn must attach additional unpaired proton.

All questions mentioned above contribute to the field of interest to the fission physics when detailed and self-consistent studies can lead to observation of so exotic nuclear phenomena like current of nucleons inside hyper-elongated and practically cold nuclear system. Chaotic motion

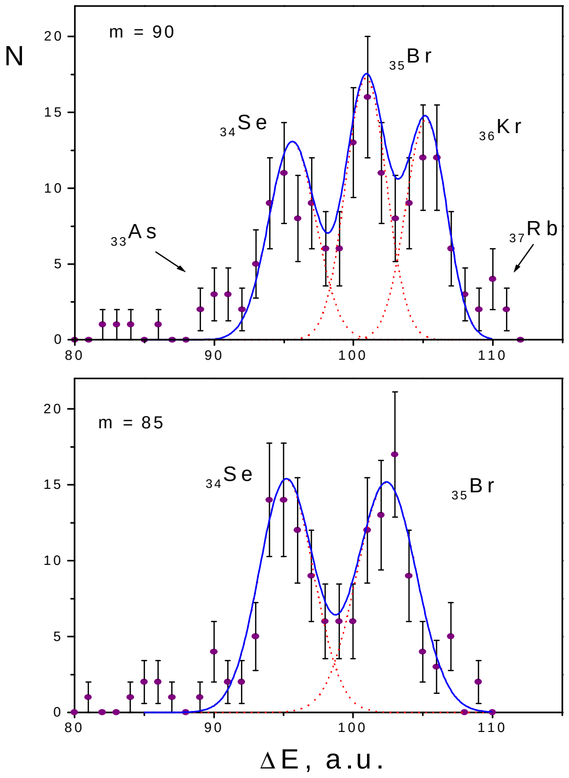

**Fig.14.** Fission fragment nuclear charges derived from Bragg-parameter distribution in ionization chamber filled with Ar+CO$_2$ – gas mixture. Cold fission of neptunium-237 by 1 MeV- neutrons was studied. Bottom – picture demonstrating zero odd-even effect expected for odd-Z fissioning nucleus. Top – observation of odd-even effect around maximum yield of fission fragment mass curve.

of nucleons of both types is transforming into order of at least part of them. In this respect the thorium-cycle isotope – ${}^{231}$Pa – looks like extremely interesting. The motivation of its detailed investigation is following. Neptunium and americium mentioned above, are the nuclei with rather long stage of descent from saddle point to the scission one. The excitation energy of the fissioning system in the scission point can be relatively large and in principle enough for breaking of proton pairs which increases an amount of free protons in addition to one proton, initially unpaired. The heating of the system is due to the dissipation of collective energy into internal degrees of freedom of the nucleus. That viscosity effect is roughly proportional to the descent length. For the protactinium this length is much shorter than for neptunium or americium. Therefore odd-even effects are expected to be much more pronounces. Changing neutron energy with one can look at the dynamics of nuclear charge formation in the fragments. This kind of observations is practically impossible at the mono-energetic neutron sources and spallation neutrons should be used.

## 4. DEEP DAMPING OF NEUTRON SHELLS WITH NUCLEAR TEMPERATURE.

Shell effects in fission fragments are responsible for mass asymmetry of fissile system at the scission point. On the earliest stages of descent just around the saddle area of the barrier, shells in compound system can already play some important role in structure of future fragments. Increasing excitation energy of compound nucleus a damping of those shells can be observed via detailed analysis of fission fragment shapes at scission when practically whole total kinetic energy of FF is influenced by their coulomb interaction. Corresponding experiments should be performed in the neutron energy range above 20 MeV up to 150 MeV. Thorium-232 is most convenient nucleus for these experiments having relatively narrow FF mass distribution in cold state. Frisch-gridded Bragg ionization chamber is most convenient spectrometer for this kind of studies.

## 5. VISCOSITY OF NUCLEAR MATTER AND NEUTRON CLOCK.

From the analysis of prompt neutron characteristics like angular distribution and energy spectrum in laboratory system one can deduce multiplicity and temperature of neutrons emitted after barrier penetration but before scission, or so-called pre-scission neutrons. They take place in fission due to dissipation of collective energy to internal degrees of freedom of nucleus, and therefore long duration of the descent from the barrier top. Their multiplicity gives the time by using model-dependent calculations of neutron emission width. In addition, local proton odd-even effect for different FF total kinetic energies reflects temperature of pear-like nucleus at different stages of elongation. All effects indicated above are sensitive to mechanism of nuclear viscosity unknown yet definitely and which should be established. From experimental point of view the work would be correlative analysis of FF mass-emission angle-kinetic energy-charge-prompt neutron multiplicity in one measurement cycle for different incident neutron energies. For Th-232 charge (o-e)-effect has biggest magnitude from all of actinides investigated, and therefore it can be used as a nuclear thermometer much more precisely. Again, thorium-232 is the best candidate for studies first.

## 6. PLACE OF FISSION IN GENERAL NUCLEAR PHYSICS.

One interesting demonstration of fission studies importance can be found in fig.15. There we prepared comparison of fragment mass spectra for two fissioning systems. One of them is superheavy element $^{116}296$ formated in complete fusion of $^{48}$Ca with target nuclei $^{248}$Cm. Another is uranium-238 irradiated with fast neutrons. Spectra are selected for one excitation energy of 35 MeV. One can see that light fragments of SHE are completely the same as heavy fragments of uranium. The width is the same too. It means that in principle fission process is unique for all of fissile nuclei and shells both proton and neutron are the same. Studying fission of usual nuclei we have good tool to get information and understanding of main properties of nuclei from “island of stability” which is closed to problems of nucleoproduction.

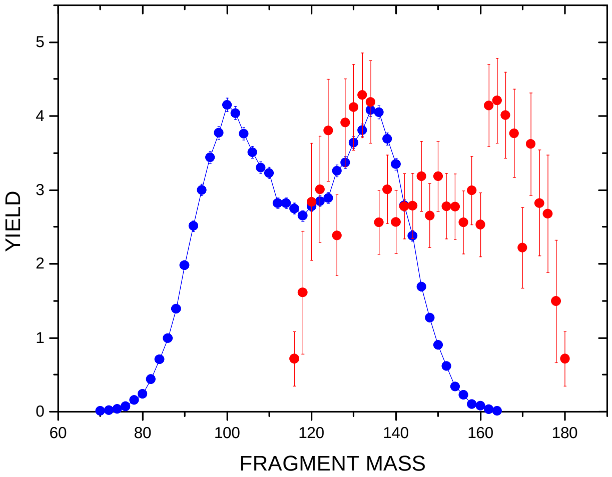

**Fig.15.** Fission of superheavy nuclei and uranium (see text )

# REFERENCES

1. Bjornholm S. and Lynn J.E. Rev.Mod.Phys. **52** (1980) 725
2. Polikanov S.M. e.a. ZETP **42** (1962) 1016
3. Strutinsky V.M. Nucl.Phys. **A95** (1967) 420
4. Behrens J.W. e.a. NSE **63** (1977) 250
5. Plattard S. e.a. PRL **46** (1981) 633
6. Blons J. e.a. Nucl.Phys. **A502** (1989) 121c
7. A.Bohr Peacefull Uses At. En., Geneve, 1955. UN, New York. **2** (1956) 220
8. Brosa U. Phys.Rep. **197** (1990) 167
9. James G.D. PR **C15** (1977) 2083
10. Meadows J.W. NSE **65** (1978) 171
11. Lamphere R.W. Nucl.Phys. **38** (1962) 561
12. Krasznahorkay A. e.a. Phys.Lett. **B461** (1999) 15
13. Oberstedt S. e.a. Nucl.Phys. **A564**(1998)
14. Muir D.W. e.a. Int.Conf.Nucl.Data. Knoxville.1971. Proc. V.1. P.292
15. Sicre A. e.a. Phys. Chem Fission. Julich. 1979. IAEA. 1980. V.I. P.187
16. H.R.vonGunten e.a. NSE **27** (1966) 85
17. Danon Y. e.a. NSE **124** (1996) 482
18. Soldatov A.S. e.a. Sov.J.Nucl.Phys. **46** (1987) 389
19. Gavron A. e.a. PR **C13** (1976) 2374
20. Back B.B. e.a. PR **C10** (1974) 1948
21. Hambsch F.-J. e.a. Nucl. Phys. **A491** (1989) 56
22. Furman W.I. Private communication. May 2000.
23. Goverdovski A.A. and Mitrofanov V.F. - Phys.At.Nucl. **55** (1992) 16
24. Goverdovski A.A. e.a. Phys.At. Nucl. **58** (1995) 1841
25. Steiper F. e.a. Nucl.Phys. **A563** (1993) 282
26. Goverdovski A.A. e.l. Doctoral Thesis. Dubna. 1995
27. Goverdovski A.A. e.l. Phys. At. Nucl. **44** (1986) 287; **46** (1987) 706
28. Trochon J. e.a. Proc. Int. Conf. on Nuclear Data, IAEA, Vienna. **1** (1982) 733.
29. Berger J.-F. e.a. Nucl.Phys. **A502** (1989) 85c

30. Goverdovski A.A. e.a. ISINN-8. Dubna. May 2000. In Press
31. Blons J. e.a. PRL **35** (1975) 1749
32. Schpak D.L. e.a. Sov.J.Nucl.Phys. **50** (1989) 922
33. Vives F. Ph.D. thesis. IRMM. Geel. Belgium. 1999
34. Vives F. e.a. Nucl.Phys. **A662** (2000) 63
35. Sergachev A.I. e.a. Sov.J.Nucl.Phys. **9** (1969) 296.
36. Kaufmann Ju. Dissertation. Universitat Tubingen Germany. 1996.
37. Goverdovski A.A. e.a. Phys.At. Nucl. **58** (1995) 188
38. Behrens J.W. e.a. NSE **81** (1982) 512
39. Gonnenwein F. In: Yhe nuclear fission process. Ed. Wagemans C. CRC Press. 1992
40. Goverdovski A.A. Report IPPE-262. Obninsk. 1999
41. Pashkevich V.V. Proc. 15th Nucl. Phys.Conf., Oganessian Y., Eds. (1995) 161
42. Niffenecker H.A. e.a. Phys. Chem Fission. Julich. 1979. IAEA. 1980. V.I. P.35
43. Bjornholm S. Phys. Scr. **10a** (1974) 110
44. Martinez G. e.a. Nucl.Phys. **A515** (1990) 433
45. Stumpf P. e.a. Report EUR 16295 EN. 1995
46. Tsekhanovich I. e.a. Nucl.Phys. **A658** (1999) 217
47. Steinhauser S. e.a. Nucl.Phys. **A634** (1998) 89