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ON TERNARY FISSION PRODUCED IN GOLD BISMUTH, THORIUM AND URANIUM WITH ARGON IONS

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1. *Introduction*

The investigation of nuclear fission into three fragments of comparable masses ("ternary fission") is a relatively new field in fission physics. It has been investigated under three essentially different conditions:

a) Ternary fission at low excitation energies (thermal neutron induced and spontaneous fission) is a very rare process with a probability of $\approx 10^{-6}$ as compared to binary fission/1/. One of the three fragments seems to have a rather low mass/1,2,3/, but might not be a radioactive nucleus/4,5/. The earlier work in this field has been reviewed by Hyde/6/.

b) Ternary fission induced by 20 GeV protons has been observed recently with the help of two kinds of solid-state track detectors. Muscovite mica detectors register essentially heavy fragments with $A \geq 30$. Using the mica-sandwich technique, the probability of ternary fission in uranium was determined to be 0.13 percent as compared to binary fission/7/. Using a polycarbonate solid-state track detector/8/, which is sensitive to particles with $A \geq 14$, this probability is ≈ 1 percent. So far only the existence of ternary fission has been proved, but no further characteristics of this process have been published.

c) Ternary fission *induced by heavy ions* has been studied to some extent. Price et al./9/ reported the frequency of this process in the interaction of argon ions with Bi and U as being some tenth of a percent up to 3 percent. They used mica and phosphate glasses as detectors. Both detectors are sensitive to bombarding particles. Kapuszik et al./10/ found that it is possible to detect in mica also to some extent such light particles as Ne . Therefore, an inelastically scattered particle accompanied by binary fission of the target nucleus can be misinterpreted as ternary fission. Karamian et al./11/ studied ternary fission using three semiconductor detectors placed at 120° to each other in a plane perpendicular to the beam direction. They bombarded Au , Bi and U with Ne and Ar and found a sharp increase of the probability of ternary fission with the energy of the bombarding particle.

The most convincing evidence for the existence of ternary fission was provided by Fleischer et al./12/. They irradiated Th in the form of artificial thorite crystals ($ThSiO_4$) with 414 MeV Ar . These crystals are insensitive to Ar - and Ca -ions. The frequency of ternary fission is 3.3 percent as compared to binary fission. The angular distribution of fragments is described.

Ternary fission has been investigated also from a theoretical point of view. Swiatecki/13.14/ and Strutinski et. al./15/ discussed the possibilities of multifragment break-up of a heavy nucleus using the liquid-drop model. Muzychka et al./16/ developed the hypothesis, that ternary fission occures in two binary fission steps ("cascade fission") and the work of Karamian et al./11/ is interpreted in this light.

Recently, it was discovered, that heating of mica for 6h at 420°C causes only the annealing of tracks of bombarding Ar particles. They can no longer be etched, but fission fragment tracks can still be developed with hydrofluoric acid /10/. It seemed useful to repeat some of the work of Price et al./9/. mentioned above. Previous attempts to employ the mica-sandwich technique (4π -geometry) in heavy-ion bombardments proved to be rather inconclusive, since only a small number of the actually produced ternary fission events are registered as such/17/. The technical problems could not be solved in this case. However, it was possible to succeed using more restricted geometrical conditions. Ternary fission is studied in this paper in the " 2π -forward" geometry for the interaction of Ar -ions with Au , Bi , Th and U . After a description of the experimental technique, the results will be presented and discussed.

2. Experimental

A thin layer (20-100 μ g/cm^2) of target material was prepared by evaporation of Au , Bi , ThF_4 or UF_4 on cleaved muscovite mica. The irradiations were carried out with 230-380 MeV Ar -ions produced in the 300 cm cyclotron of the JINR, Dubna, U.S.S.R.

The heavy ion beam was perpendicular to the target side of the detector. Due to the subsequent heat-annealing of the Ar -tracks, fluxes as high as 10^{11} particles/cm^2 could be tolerated. After irradiation, the heavy elements were dissolved and the mica was heated for 6h at 420°C. Afterwards it was etched in 48% HF for 20 min at 20°C. An additional experiment provided further information on the lower limit of Z values of recoils, the tracks of wich could no longer be annealing by this technique. In the bombardment of thin aluminium foils with 200 MeV Ne - ions 80 MeV compound nuclei of vanadium (Z =23) were produced. These V -ions impinged onto mica, but the tracks produced could be completely annealed by heating as mentioned above. Thus the lower limit for the detection of charged particles in this experiment was $Z \geq 24$. (Fleischer et. al./12/ estimated that $ThSiO_4$ is sensitive only to particles with $Z \geq 21$).

After annealing and etching, the mica was scanned at 900 x magnification in an optical microscope. The following types of events were observed (Fig. 1 a-c):

1) A dotted background caused by the interaction of Ar -ions with detector material.

2) Single tracks (called single events) originating from binary fission with only one fragment moving forward in the laboratory system. An event is considered a "single track" and not part of the "dotted background", when its length is about twice that of the "dotted background" events. It is admitted that this differentiation is somewhat arbitrary.

3) Correlated pairs of tracks (called binary events) arising from binary (or ternary) fission with two fragments moving forward in the laboratory system.

4) Three-pronged events (called ternary events) originating from ternary fission with all three fragments moving forward in the laboratory system. At ternary fission events only those are accepted, for which the distance between the points of entrance into the mica and the intersect of the projected direction is less than 2 μ m. It is interesting to note, that Debeauvais et al./8/ and Brandt et al. /7/ employed the sandwich technique and were forced to accept events with $d \leq 20$ μ m as ternary fission.

With such large distances d one has to prove, that those three-pronged events originated in ternary fission and not from an accidental overlap of a single track with a binary fission event. In this paper, however, it is not necessary to prove the observation of ternary fission explicitly, since d is considerably smaller. (Compare also with Fig.1a-c).

Results and Discussions

These experiments yielded two kinds of information:

I) Ratios of the observed numbers of single, binary and ternary events were determined. It is worthwhile to note, that more than 10^3 three-pronged events were observed.

II) Some details of the geometrical properties of ternary fission events, in particular track lengths and angles between tracks were obtained. Therefore, the angle ϕ between the projections of tracks in a plane parallel to the mica surface was measured as well as the length of these projections and the depth of track dipping.

ad I) The results of the determination of ratios of single, binary, and ternary events are given in Table 1. Due to the difficulty in definiting single events, the ratio of single to binary events can only be estimated. The numbers of binary and ternary events were actually counted event by event. From Table 1 is seen that on thick targets (≈ 100 $\mu g/cm^2$) the registration efficiency of ternary fission relative to binary one is about 30 -40% lower than for thin layers (20 - 30 μg / cm^2), The ratio of binary to ternary events given in Table 1 and Fig.2 holds only for the 2π -foreward geometry employed in this experiment.

This ratio is not equal to the true binary to ternary fission ratio in the 4π -geometry. It is not possible to calculate this true binary to ternary fission ratio from the experimental data reported here, since the angular distribution of ternary fission events in the 4π -laboratory system is generally not known. Nevertheless, Fig.2 contains also results obtained in 4π -geometry experiments .

In the experiments with Bi at energies $E < 300$ MeV, the ratio of ternary to binary events is $\approx 10^{-3}$. In this case it may appear possible, that the elastic scattering of a binary fission fragment by a nucleus of the target material can be misinterpreted as a ternary fission event. To check this. a thin Th -foil (1 μ m) was bomberded with Ar -ions. Fission fragments left the foil and impinged under 15° onto a mica plate covered with $50 \mu g/cm^2$ Bi . After the irradiation and corresponding processing, 10^4 single fission fragment tracks were found on the mica surface. Not a single event due to elastic scattering of a fission fragment on a Bi nucleus could be observed. Evidently, the upper limit for such a process to occur is sufficiently low.

It seems to be premature to compare the results of ternary to binary events obtained in the 2π -geometry of this work with theoretical calculations for the total ternary to binary fission ratio in the 4π -geometry. Still, Muzychka et al. calculated the ratio σ (ternary)/σ (binary) using a two-step "cascade" fission model$^{16)}$.

The experimental ratios of this work are considerably higher at low energies of the Ar -ions than those calculated theoretically. But the experimental ratio of ternary to binary events increases sharply with Z^2/A of the compound system for a given excitation energy (Fig.3) in agreement with those calculations. The extrapolation of this result into the region of still bigger values for Z^2/A

yields in the system ($^{241}Am + Ar$) a ternary fission probability twice as big as that in the system ($^{238}U + Ar$).

ad II) Some geometrical characteristics of three-pronged events were obtained for the following, representative cases: Th + 305 MeV Ar, U + 305 MeV Ar , Bi + 380 MeV Ar , U + 380 MeV Ar . The distribution of the projected angle ϕ in a plane parallel to the mica surface between pairs of fission fragment tracks are shown in Fig. 4 a-e .

All distributions have a maximum at $\phi = 120^{o}$. This indicates that ternary fission is a "symmetric" decay process with respect to ϕ . The measurement of this angle can be carried out with sufficient accuracy for tracks, whose angle θ with mica plane is $\lesssim 40^{o}$ (flat tracks). For steep tracks with $\theta > 40^{o}$, it is difficult to measure ϕ accurately, since the projection of the track onto the mica plane is not much longer than the apparent diameter of the track. Therefore, such events with steep tracks, being rare anyway, are omitted in Fig. 4 a-e.

For the system mentioned above, the track-lengths of binary and ternary events were calculated from the measured lengths of the projections and measured depths of the tracks (Fig. 5 a-e). All these distributions look rather similar, the mean values agree within the limits of error. However, it appeares that the distributions for ternary events are somewhat wider than those for binary events. Finally two attepmts to extract further information from these measurements are described.

Fifty events of the ternary fission of the compound nuclei in the Th + 305 MeV Ar reaction were studied. Fig. 6 shows the distribution of angles ψ between each pair of tracks in the center-of-mass system and ϕ angles between their projections on the plane parallel to the mica surface for these fifty tracks.

If we assume the compound nucleus is fissioned into three fragments of equal masses, no light particles accompany this fission and the summary kinetic energy releasing during the fission process is equal to 300 MeV, we may use the center-of-mass system. However, at the transition to the center-of-mass system the experimental error in the summary of the dipping angles of the tracks appeared to be very significant. This error is caused by the inaccuracy in the determination of the track depth ($\pm 1 \mu$ m). In Fig.6 is seen the distribution of the projection of the angles ϕ and center-of-mass system angles ψ among the fragments are similar.

Additionally, the true angle in space ϵ between pairs of tracks is calculated directly from the measured values of the projected track length, the angle ϕ and the depth of the track using standard geometrical considerations. This procedure is rather inaccurate but only directly measured quantities are used ($\Delta\epsilon \approx 10^{\circ}$ as compared to $\Delta\phi \approx 2^{\circ}$). Also the sum of the three true space angles

$$\phi = \epsilon_1 + \epsilon_2 + \epsilon_3 \qquad (1)$$

for ternary events were obtaine. Figs.7a and b show the results for the system(U + 305 MeV Ar). "Theoretical" values for the angles ϵ and ϕ , called ϵ_m and ϕ_m respectively, can be calculated using the following assumptions for the system (U + 305 MeV Ar).

i) ternary fission yields three equally large fragments and no further reaction partners.

ii) the kinetic energy of each fragments is 100 MeV in the laboratory system.

iii) ternary fission occures in a plane perpendicular to the direction of the beam.

The angles $\epsilon_m = 108^\circ$ and $\phi_m = 324^\circ$, calculated according to this model, agree surprisingly well with the observed maxima in Fig. 7a and b. This indicates that the observed three-pronged stars originate from ternary fission.

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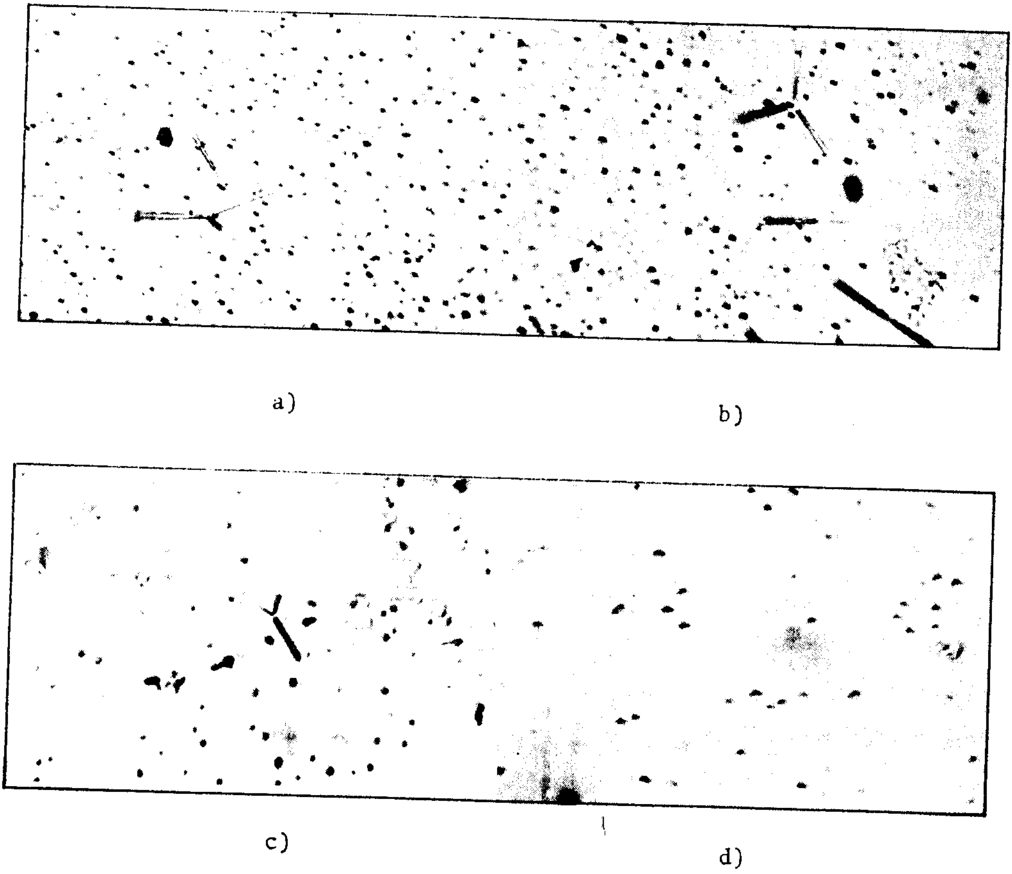
Table 1

Ratios of single to binary and binary to ternary events in the 2π - forward geometry

System	Estimated ratio of single to binary events [a)]	Observed binary events	Number of ternary events	Ratio of binary to ternary even
Au + 305 MeV Ar	6.3	50000	16	3130
Au + 380 MeV Ar	7.1	10220	10	1020
Bi + 260 MeV Ar	5.4	21306	6	3370
Bi + 305 MeV Ar	7.2	86740	79	1097
Bi + 380 MeV Ar	7.7	35100	86	408
Th +230 MeV Ar	8.0	43430	39	1130
Th + 260 MeV Ar	-	32106	77	416
Th + 305 MeV Ar	7.7	27777	197	141
Th + 305 MeV Ar [b)]	7.9	22321	127	176
U + 230 MeV Ar	8.2	10520	10	1050
U + 260 MeV Ar	-	20410	44	465
U + 305 MeV Ar	7.0	16250	117	139
U + 305 MeV Ar [b)]	7.6	6118	26	240
U + 330 MeV Ar	6.5	9820	83	119
U + 350 MeV Ar	6.8	18810	190	99
U + 380 MeV Ar	-	9364	137	72
U +380 MeV Ar [b)]	7.5	3500	339	103

a) The ratio of single to binary events can only be estimated due to experimental difficulties, as described in the text.

b) Thick targets were used.



a) b)

c) d)

Fig.1.Microphotographies of tracks as observed in mica. Fig. 1a, b and c are typical examples of ternary fission stars produced in the bombardment of U with Ar -ions. Fig. 1d shows the dotted background as produced in the interaction of Ar -ions with the detector material.

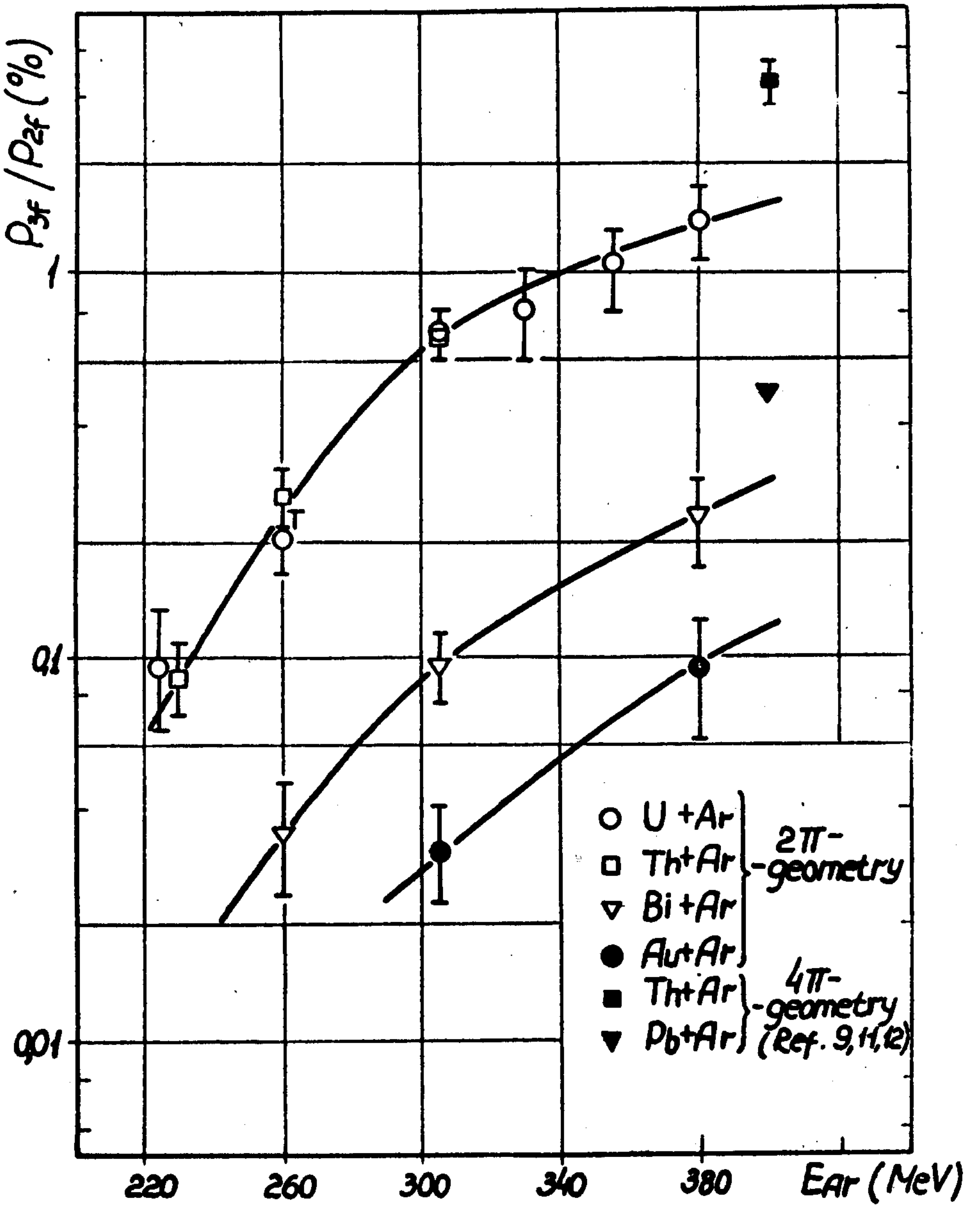


Fig.2. Ratios of ternary to binary fission P_{3f}/P_{2f} .

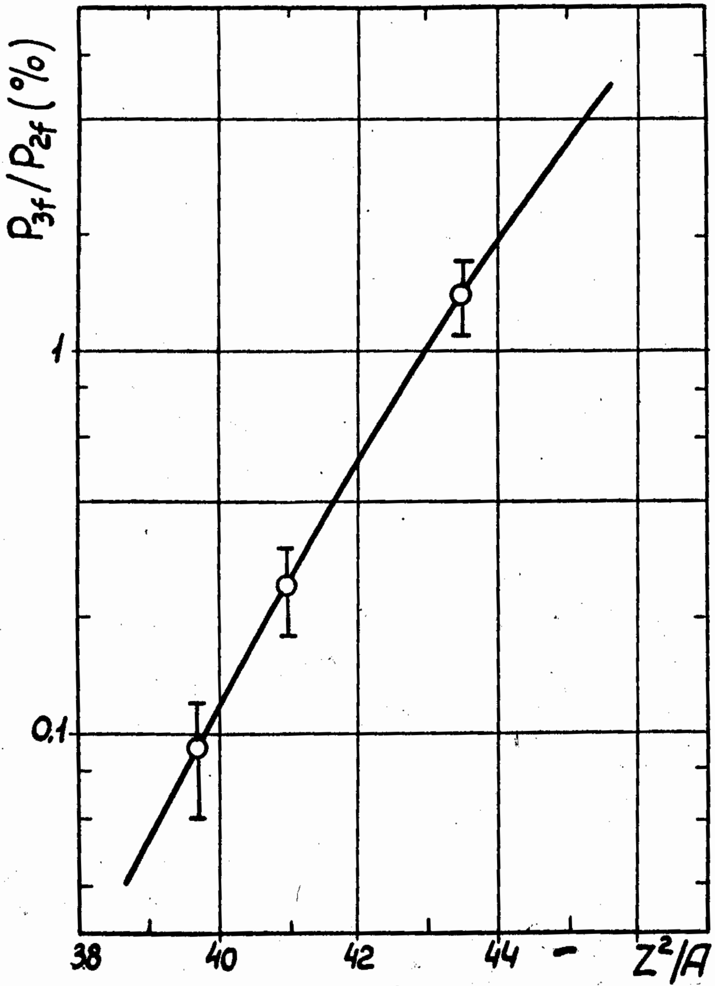
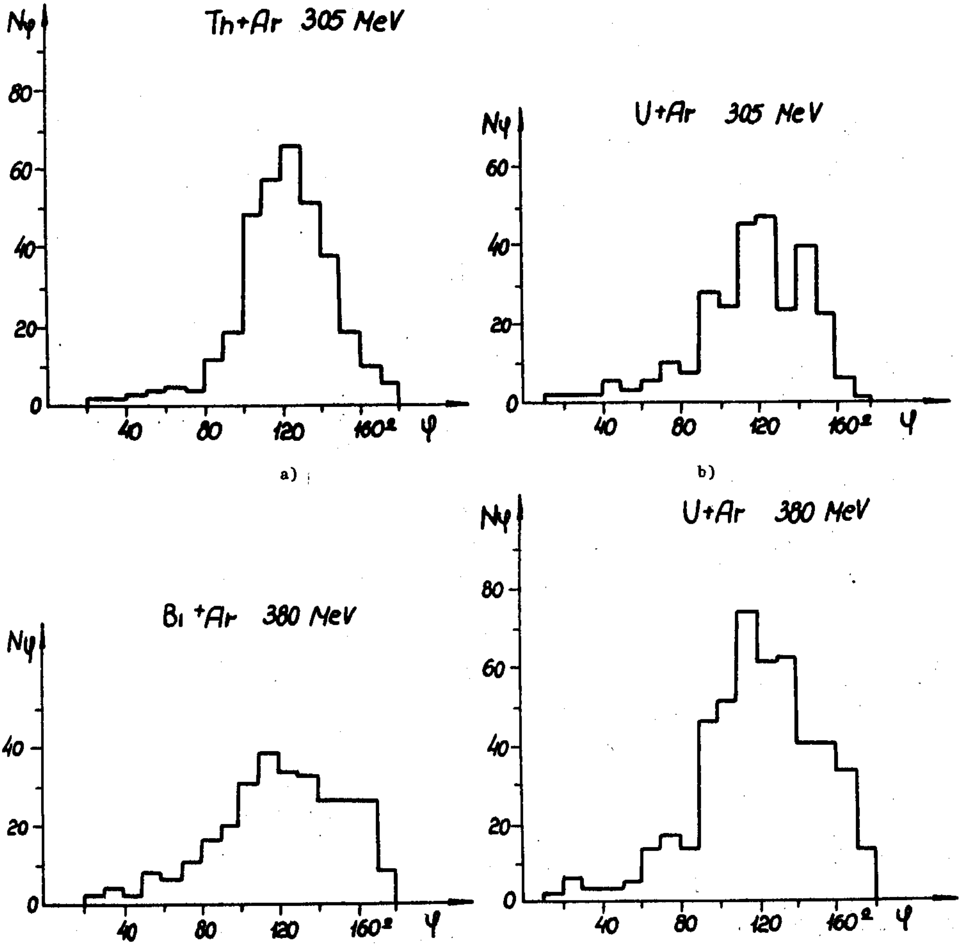
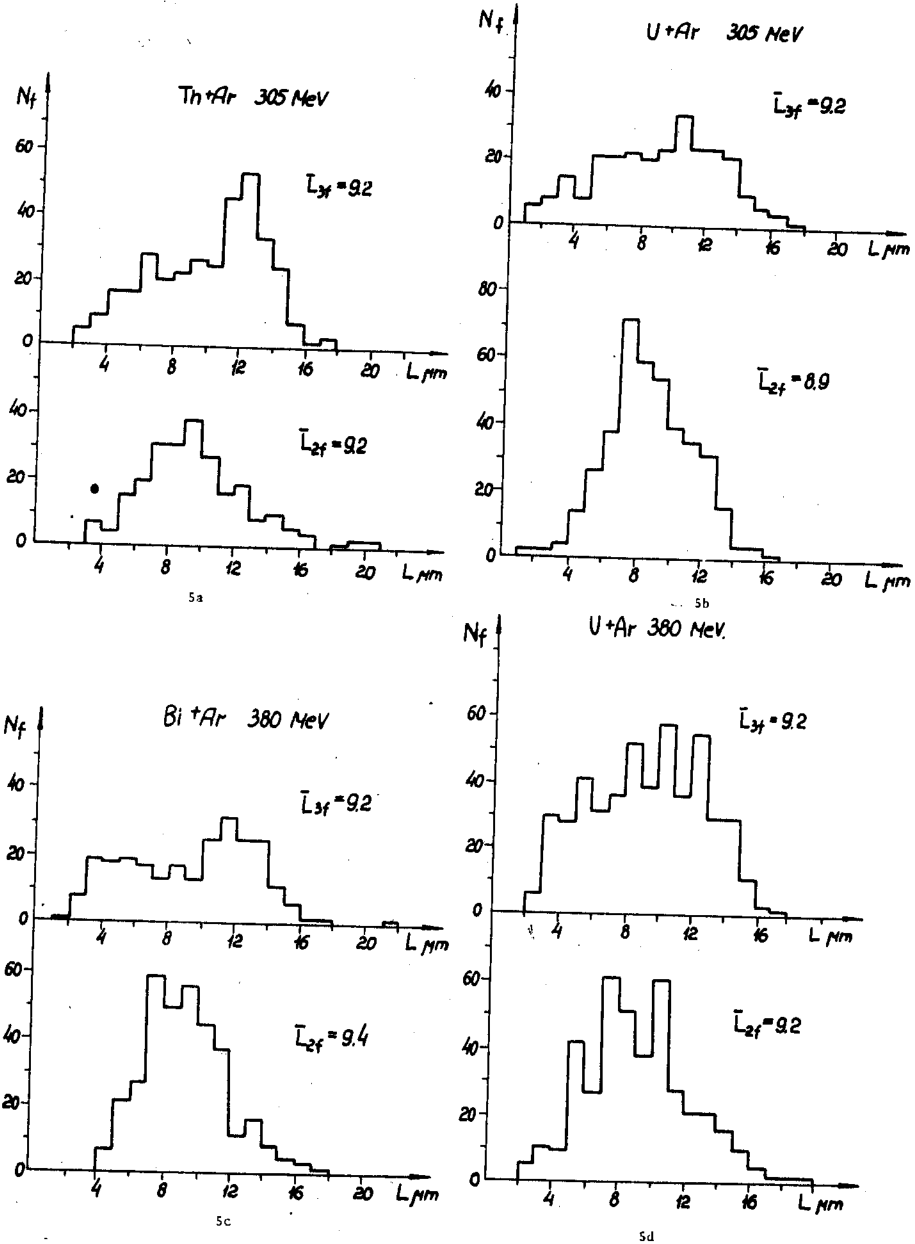


Fig.3. Dependence of the ratio of ternary to binary fission P_{3f}/P_{2f} on Z^2/A. The excitation energy of the compound nucleus is 190 MeV, Ar -ions are used as bombarding particles.



a) b) c) d)

Fig.4. The distribution of the angle ϕ between the projections of tracks in a plane parallel to the mica surface for the systems:
a) Th + 305 MeV Ar , b) U + 305 MeV Ar,
c) Bi + 305 MeV Ar d) U + 380 MeV Ar.



5a

5b

5c

5d

Fig.5. The distribution of tracks lengths for binary and ternary fission. The same system as in Fig.4 are investigated here.

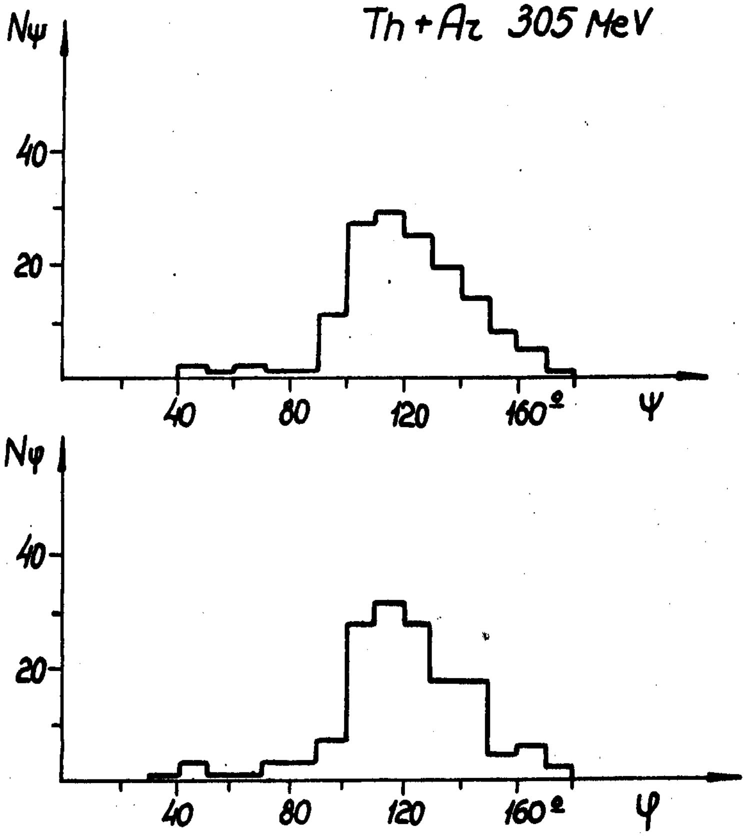
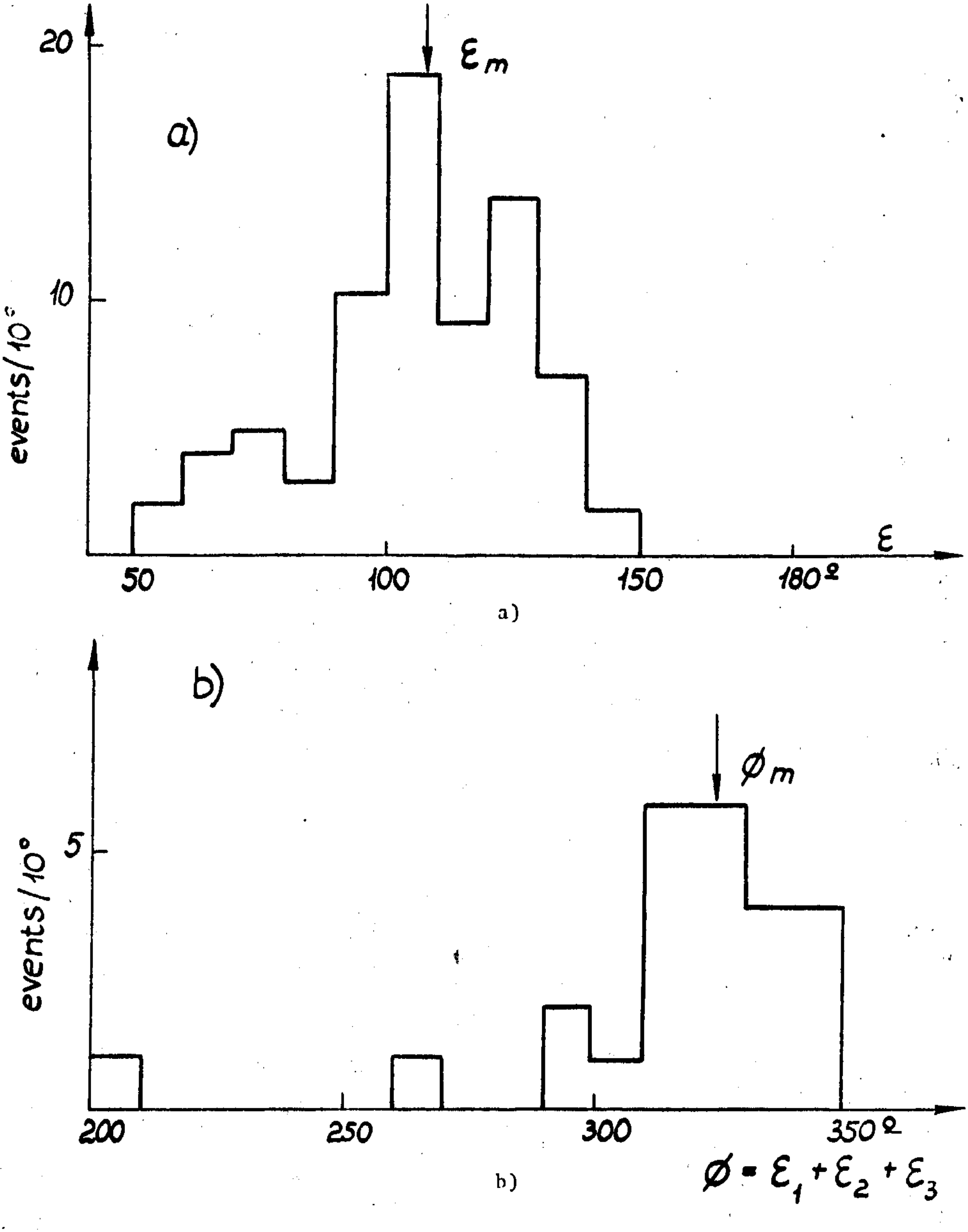


Fig.6. The distribution of the angle ψ between the fission fragments in the center-of-mass system and angle φ for 50 ternary events in the system (Th +305 MeV Ar).



a)

b)

Fig.7. The distribution of the angle ϵ between two tracks <u>in space</u> (Fig. 7a) and of the angle $\phi = \epsilon_1 + \epsilon_2 + \epsilon_3$ (Fig. 7b) in ternary fission in the system (U + 305MeV Ar). The angles ϕ_m and ϵ_m are "theoretical" values calculated according to a model described in the text.