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EXPERIMENTAL RESULTS ON THE NUCLEAR FISSION INDUCED BY 600 MeV PROTONS *

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Summary. The fission phenomena induced by protons of about 600 MeV have been studied in several nuclei (^{238}U, ^{232}Th, ^{209}Bi, ^{194}W and natural silver) by means of loaded, low-sensitivity nuclear emulsions. A significant improvement was obtained in the knowledge of the relative cross-sections (Table I). We have studied the angular distribution for U and Th with particular regard given to the "non-collinear" tracks. No sign of anisotropy was found. For non-collinear events, a possible correlation was sought between the angle formed by the two tracks and the angle between the line connecting the two track extremities and the direction of the incident beam. No correlation was found. These results agree only partially with those obtained by other authors.

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1. Introduction.

The fission phenomena induced by the bombardment of heavy and medium-heavy nuclei with high energy (> 100 MeV) projectiles have been the object of several investigations[1].

However, partly because of the experimental difficulties, the existing data about this subject is rather poor. Indeed, the problem of discriminating the fissions of the heavy, highly-fissionable nuclei from the natural alpha-radioactivity phenomena must be solved, while on the other hand, non-radioactive nuclei often show very low fission cross-sections.

Besides, it is always necessary to discriminate the fission from the spallation phenomena. For increasing energies the further problem of separating fragmentation events[2] has also to be solved: this problem is particularly important for nuclei of rather low Z. For these reasons it seemed to us interesting to make an investigation about the high-energy fission events in an almost unexplored energy region (~ 600 MeV), using an already described technique[3,5] which seems to us one of the most suitable for this type of research. Low sensitivity pellicles loaded in a quantitative way with different elements were prepared in the Naples laboratory, starting from Ilford KO emulsion in gel form. The loading elements were: U, Th, Bi and W.

The loaded pellicles, together with some unloaded ones, were exposed to a (591 ± 3) MeV proton beam[6] from the CERN synchro-cyclotron. Some stacks were exposed at perpendicular, and some at grazing incidence.

Two topics were taken into consideration and form the object of the present paper: i) fission cross-section values, and ii) angular distribution of fragments with respect to each other and with respect to the incident beam; the correlation between these two distributions was also studied.

2. Cross-section measurements.

The monitoring of the beam was made by means of an ionization chamber which was reliable only for comparison between different exposures (within 5%). Its indications were estimated to be lower (by about a factor of 2) than the absolute proton flux at the stack location, so that we could not rely on it for absolute cross-section measurements.

However, by means of our precision loading technique (errors of 1%) [4,5] it was possible to obtain good measurements of the ratios $r = \sigma x/\sigma y$ between the cross-sections of any two, X and Y, of the considered elements. For this purpose, the stacks exposed at perpendicular incidence were used. Each stack consisted of 3 pellicles, 400 μm thick, each loaded with a different element, so that the relative cross-sections could be measured for any three of the investigated elements under identical exposure conditions. Different exposures were made in turn, with the same dose to within ± 5%. Each stack was so thin (< 0.5 g/cm^2, package included) that the secondary production could not give any appreciable contribution (~1%).

The pellicles were developed according to our low-sensitivity technique[3,5], mounted on glass and scanned according to a standard method (see below). The fission tracks were very clearly recorded, while any other track was not visible at all. The scanning efficiency was estimated by intercalibration to be constant and of the order of 95%.

For the Tungsten cross-section measurement, some difficulties were encountered due to the background arising from the emulsion nuclides. Indeed, for W, the fission cross-section (see Table I) is not so high as to overcome the high ratio (~150) between the numbers of Ag and W nuclei in the loaded emulsion. Therefore, a very accurate comparison between W loaded and unloaded pellicles was made and a reliable discrimination method was established, based essentially on range measurements; other experimental details on this point will be given elsewhere[7].

The results of all the described measurements of relative cross-sections and some other data of interest are collected in Table I, columns 1 to 4.

Of course, if one of the cross-sections were known, all the other ones as well as the absolute proton flux could be deduced[8]. But, in the 600 MeV energy region, all the cross-sections are rather poorly known. Therefore, in order to estimate the absolute cross-sections, we carefully considered all the known measurements of the cross-sections for fission of U, Th and Bi, induced by high-energy protons from 120 MeV on [9,10]. We made a cross-comparison between our measured ratios and the several reported cross-section values in the (120-660) MeV region. Besides, we took into account the fact that in the (9-20) GeV region the U and Th cross-sections are not much lower than in the 600 MeV region [11]. We thus reached the conclusion that our measured ratios agree quite well with the published data, if we take the value $\sigma(u) = (1.20 \pm 0.10)$ barns as best estimate of the cross-section for fission of U at 600 MeV. This has been used a normalization value to obtain the absolute cross-sections in the last column of Table I.

Table I. Experimental data of interest for the cross-section measurements.

Nuclides X	Loading density (nuclei/cm^2)	Observed events (N_1)	Relative cross-sections $r = \sigma X/\sigma U$	Cross-sections σX (cm^2)
^{238}U	$6.76 \; 10^{18}$	8 744	1.000	$(1.20 \pm 0.10) \; 10^{-24(*)}$
^{232}Th	$4.99 \; 10^{18}$	6 294	0.607 ± 0.016	$(0.73 \pm 0.08) \; 10^{-24}$
^{209}Bi	$4.94 \; 10^{18}$	780	0.184 ± 0.009	$(0.22 \pm 0.03) \; 10^{-24}$
^{184}W	$1.93 \; 10^{18}$	49	$(0.75 \pm 0.14) \; 10^{-2}$	$(9.0 \pm 2.2) \; 10^{-27}$
Ag	$3 \; 10^{20}$	166	$\leqslant (0.25 \pm 0.03) \; 10^{-3}$	$\leqslant (3.0 \pm 0.6) \; 10^{-28}$

(*) Normalization value.

In column 4 the overall experimental errors are given. They include, besides statistical errors, those affecting the loading and the exposure doses. In column 5 some of the errors are strongly enhanced by the uncertainty on the adopted normalization value.

However, we must state at once that the error indicated above could well be rather underestimated. Indeed, the errors on the published experimental values in the (400-600) MeV range are of the order of 30%, while small-error measurements (to ~10%) are available only at energies lower than 340 MeV [9].

From comparison of columns 4 and 5 of Table I, it is seen that the estimated error on $\sigma(U)$ gives a major contribution to the errors of several of the σ's. Therefore, the latter errors should be also affected if the $\sigma(U)$ error were to be somewhat increased.

The value of the cross-section for W is in agreement with the very scanty existing data. Such an agreement exists also regarding the figure for Ag, about which, however, a particular remark must be made. This value has been obtained by supposing that all the possible fissions observed in unloaded emulsions can be interpreted as fissions of Ag nuclei. The result agrees with the one by Shamov [12] at 660 MeV, obtained under the same assumption. However, there are serious reasons [10] to think that this assumption must be considered with great caution, because i) an appreciable contribution from Br cannot be excluded, and ii) a major fraction of the observed phenomena is suspected to be due to fission-like fragmentation tracks rather than to true fission events. That is why the Ag values in Table I are given as upper limits.

Finally, we must add that other considerations, involving the comparison between the fission probabilities of several nuclides under proton and γ-ray bombardment, would point to cross-section values somewhat higher ($\sim$10%) than those quoted in Table I, column 5.

A more extensive discussion of these points will be given elsewhere [8].

3. Angular distributions.

The Uranium-loaded pellicles exposed at grazing incident were used to investigate the angular distribution of fragments. This distribution was already studied by several authors, with rather intriguing results.

While at low energies (in the 100 MeV region) a "positive" anisotropy was observed (that is, more fragment tracks parallel to the beam direction than perpendicular to it [13], at the higher energies under consideration, a "negative" anisotropy was found by Lozhkin et al. [14] and by other authors [15,16]. A theoretical explanation of this effect was given by Halpern [17]. He assumed that one of the principal ways by which a high-energy nucleon transfers energy to a traversed nucleus is through "grazing" nucleon-nucleon collisions. After a grazing collision, one of the colliding nucleons flies away at a near-right angle, with a rather small energy. This low-energy nucleon should represent the principal vehicle for the transmission of excitation energy to the nucleus. Therefore, the effect in an original 600 MeV beam is in some way equivalent to that of a lower-energy beam at right angle with it. The negative anisotropy should therefore have the same origin (conservation of angular momentum) as the positive anisotropy observed at lower primary energies.

This theory found some support in recent experimental data by Faissner et al. on Th [16], as well as on other elements [18]. These authors studied the angular distribution by means of nuclear emulsions. For each event, they measured the space-angles θ and ε defined in Fig. 1 and divided the fissions in two classes, "A" and "B", according to the values of the angle ε between one of the tracks and the continuation of the other one. Class A includes only **those** events for which $\varepsilon > 4^\circ$, so that a rather large momentum (and energy) transfer from the primary particle to the fissioning nucleus can be safely assumed. Class B includes all other events. Only for the latter class they do find a negative anisotropy, as can be expected on the grounds of Halpern's theory. However, their angular distribution (of the form $1 + 0.36 \sin^2\theta$) does not agree quantitatively, neither with Lozhkin's [14] data, nor with Halpern's theory [17].

In order to settle this problem, bearing in mind the fact that the contraction factor of the emulsions is often a weak point for the measurement of an angular distribution we took advantage of the very large number of fissions observed in our U and Th-loaded pellicles, at grazing incidence, and selected only "flat" events, in which the maximum dip variation was 3 μm (corresponding to dip angles $< 10^\circ$). Instead of the space angles, θ and ε, we measured the corresponding "projected" angles φ and δ, obtained respectively in a similar way (Fig. 1) but after projection of the tracks on the emulsion plane. Due to the parallel incidence and to the strong selection of "flat" events, we have $\delta \sim \varepsilon$ and (with very good approximation) $\varphi \sim \theta$.

For the scanning of the emulsions each field of microcope observed was limited by a square engraved on an eyepiece micrometer. All the tracks which traversed, or touched the left and upper edges of the square (Fig. 2, type 1) were taken into consideration, while those traversing or touching the other two edges (type 2) were discarded. In this way, any scanning bias was avoided.

The bulk of our work was performed as follows. The φ-angles were measured for all the events, while the δ-angles were measured only for those events which appeared "visually" to be non collinear. The δ-angle distribution then obtained for Uranium is shown in Fig. 3a. It is seen that $\delta > 8°$ for all the 73 measured events.

In order to render possible a comparison with Faissner's results, a group of 50 "visually" collinear events was then studied more carefully, with the aim of detecting smaller δ-angles. Any possible angular point was scrutinized and any possible δ-angle was considered from 1° on. It was found that in several cases, -angles up to (4° - 5°) could be located in a few different positions along a given track. This happened almost always for $\sim (2° - 3°)$. Apart from this, a number of tracks showing definite angular points was found, of which 14 in the range $4° < \delta \leqslant 10°$ and 6 with $\delta > 10°$ (see Fig. 3b).

We conclude that a rough visual examination permits to detect non collinear events only for $\delta > \sim 10°$, while a careful measurement of any given event permits to detect with efficiency, only for events with $\delta > (4° - 5°)$.

The described experimental work allows to examine the following points: i) angular distribution of all events; ii) frequency of non collinear events, and iii) correlation between δ and φ angles.

As to the angular distribution, our results, obtained from a group of 1 044 flat tracks selected in the Uranium-loaded pellicles, are shown in the upper part of Fig. 4. No evidence appears of any kind of anisotropy; assuming an isotropic distribution, the χ^2 test gives a probability $P(\chi^2) \geqslant 95\%$. A least square adjustment of the coefficients a and b, under the assumption that the angular distribution function is of the form $f(\theta) = a + b \cdot \sin^2\theta$, gives $b/a = 0.04 \pm 0.1$.

Similar results were obtained for the Thorium (Fig. 4, lower hystogram). We found $b/a = 0.03 \pm 0.1$ and $P(\chi)^2 \geqslant 82\%$.

These results are in definite disagreement with those by Lozhkin et al. [14] (who gave $f(\theta) = 1 + 0.29 \sin^4(\theta)$ and with the quoted ones by Faissner et al. [16], even when allowance is made for the non negligible fraction of non collinear events ($\varepsilon > 4°$); these events, according to the latter authors, do not give a contribution to the negative anisotropy.

On the contrary, our results agree with a paper published by Obukhov and Perfilov [19] while our work was in progress. These authors find results consistent with isotropy for U and Bi. They discuss as a possible source of systematic errors, in the preceding paper by Lozhkin, Perfilov and Shamov [14], a scanning bias due to the fact that a bias free scanning rule as that shown by Fig. 2 has not been followed. We conclude that the negative anisotropy is much smaller than previously stated, if it exists at all.

The frequency of non collinear events will be of course different according to how it is defined. For $\delta > 10°$ ("visual" non collinearity), according to our results, the frequency is of $\sim 7\%$. If we consider as non collinear all events with $\delta > 4°$, we find a frequency of $(36 \pm 10)\%$, in fair agreement with Faissner's results.

A rather interesting conclusion can be reached by considering the correlation between δ and φ. While discussing the momentum and energy transfer to the residual nucleus, some authors (12, 16, 20) make the assumption that the momentum of the final fissioning nucleus is parallel to the line of flight of the incident particle. Under the additional assumption of symmetric fission, one obtains the formula (16):

$$\mathrm{tg}\,\epsilon = \frac{2u}{v} \sin\theta \qquad (1)$$

where u = velocity of the fissioning nucleus and v = velocity of a fission fragment in the c. m. system.

This relation shows that for a given u/v ratio, there is a strong correlation between θ and ε as can be expected under the quoted assumptions.

Such a correlation should be expected also if allowance is made for large variations of the u/v value and for non symmetric fissions, the only necessary assumption being that u/v is not bound by some physical relation with the direction of flight of the fission fragment (in the c.m. system).

Now, Fig. 5a shows the δ and φ values of all our "non collinear" Uranium events, each dot representing an event. Fig. 5b gives the angular distribution of the same non collinear events. It is seen that i) the frequency of non collinear events is very little (if at all) dependent on φ; ii) no variation of the mean δ-values with φ is observed (as shown by the horizontal least-square regression line shown in Fig. 5a); and iii) as a consequence, there is no appreciable correlation between δ and φ. Similar results are obtained for Thorium.

We are thus compelled to conclude that eq. (1) is by no means confirmed by the experimental data and that, in general, the fissioning nucleus does not move in a direction parallel to the incident beam, as the quoted authors assumed. On the contrary, following the nuclear cascade stage, the lines of flight of the excited residual nuclei which can undergo fission must have a rather flat angular distribution, which extends over a very large angle with respect to the primary direction, so that high δ-values are about equally frequent for any φ-value.

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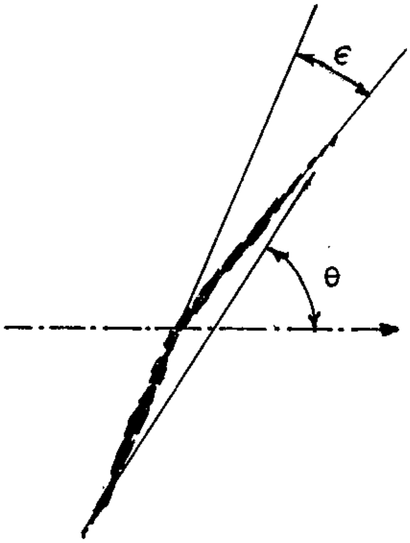

Fig 1.: Illustrating the definition of angles. When the projections of the tracks on the emulsions plane are considered instead of the real tracks, the angles obtained in a similar way are called, respectively, φ (instead of θ) and δ (instead of ϵ).

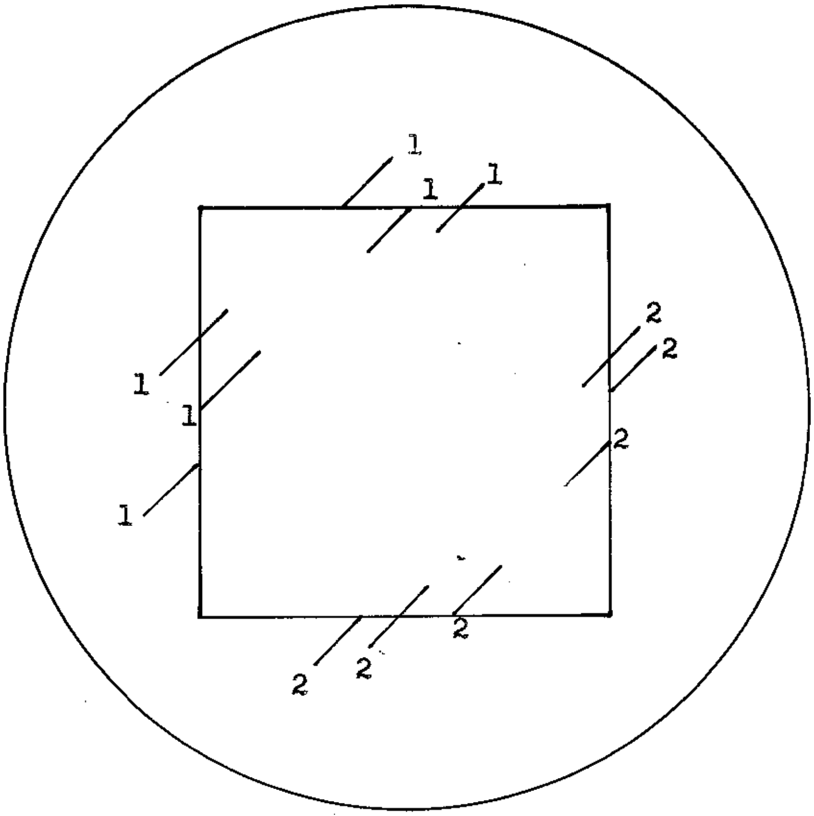

Fig. 2.: Illustrating the scanning rule.

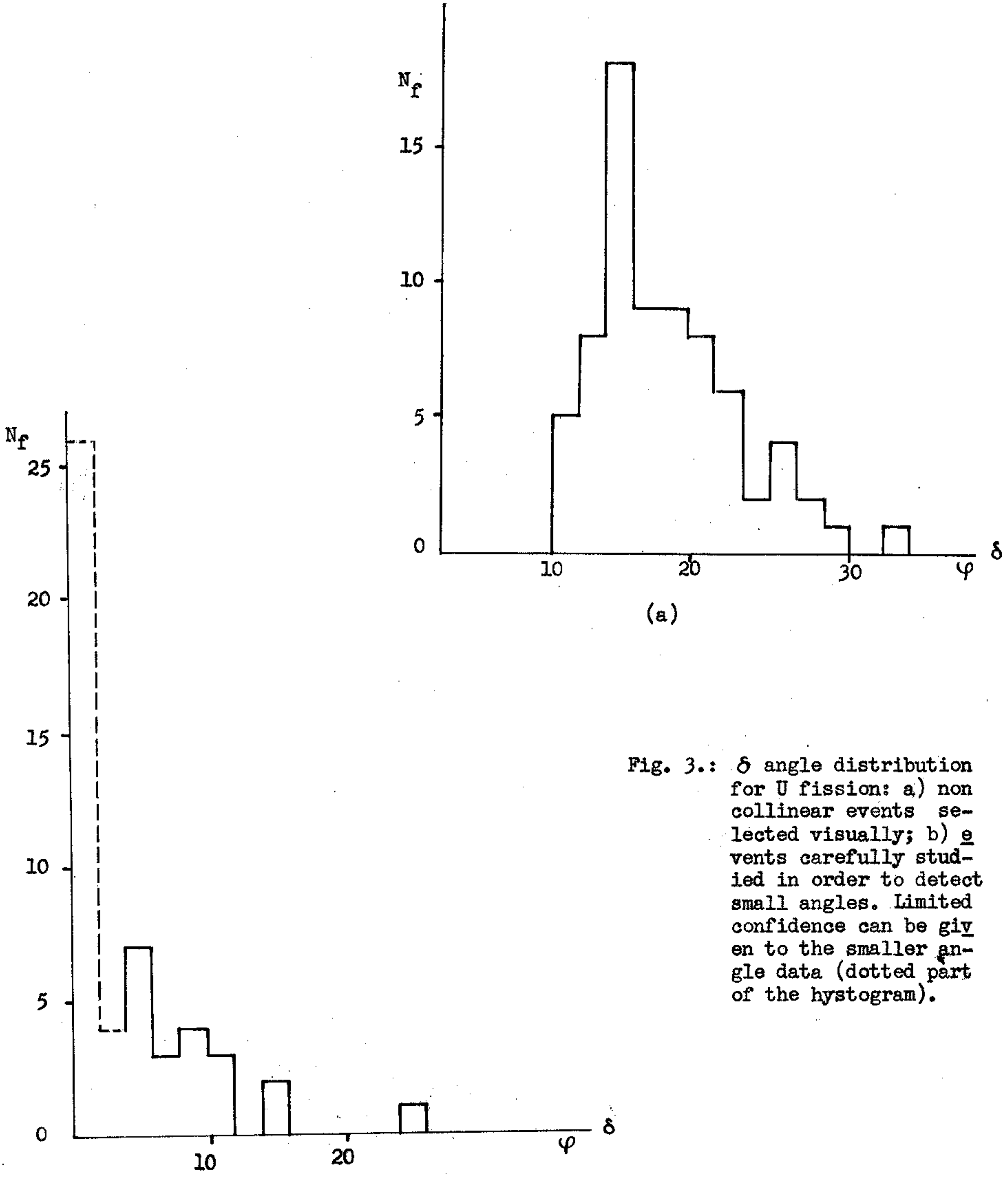

Fig. 3.: δ angle distribution for U fission: a) non collinear events selected visually; b) events carefully studied in order to detect small angles. Limited confidence can be given to the smaller angle data (dotted part of the hystogram).

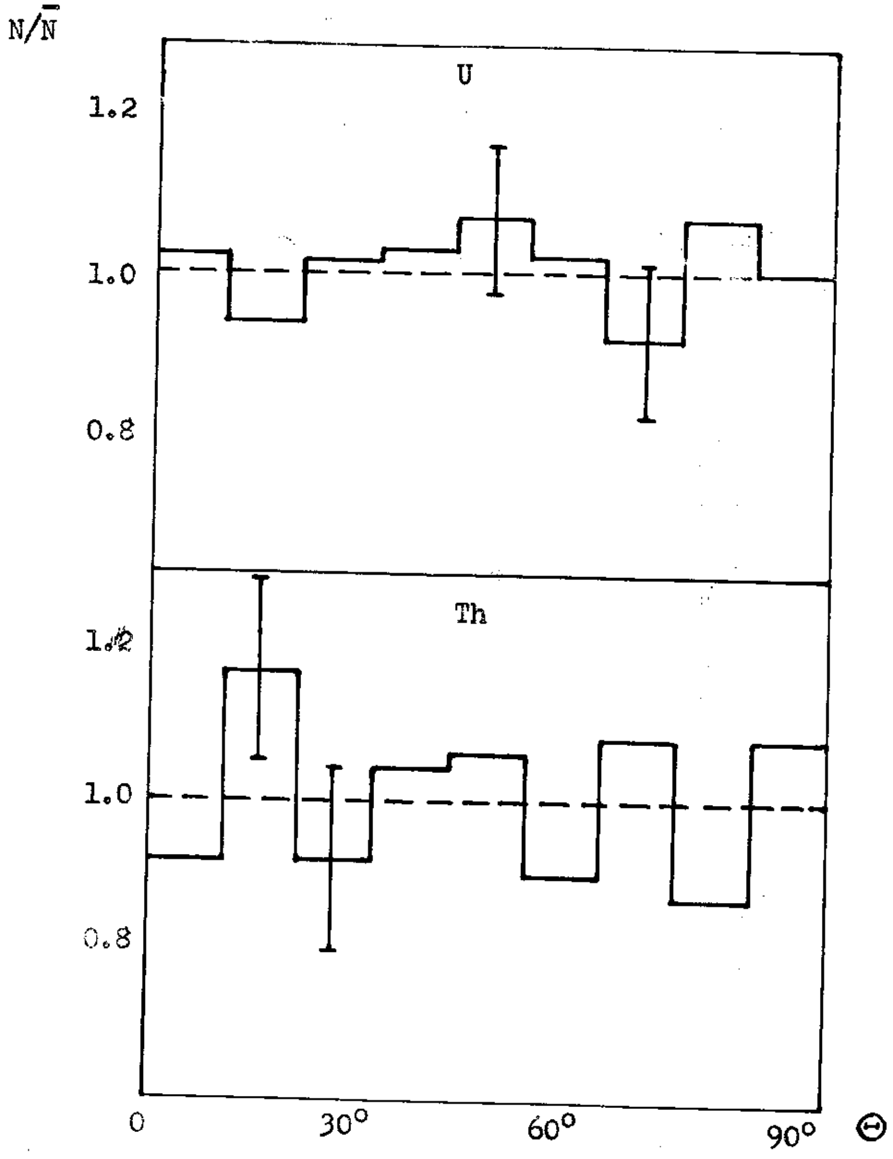

Fig. 4.: Normalized angular distributions of $\varphi \approx \theta$ angles for nearly flat events in U and Th plates. (A few statistical standard errors are shown as examples).

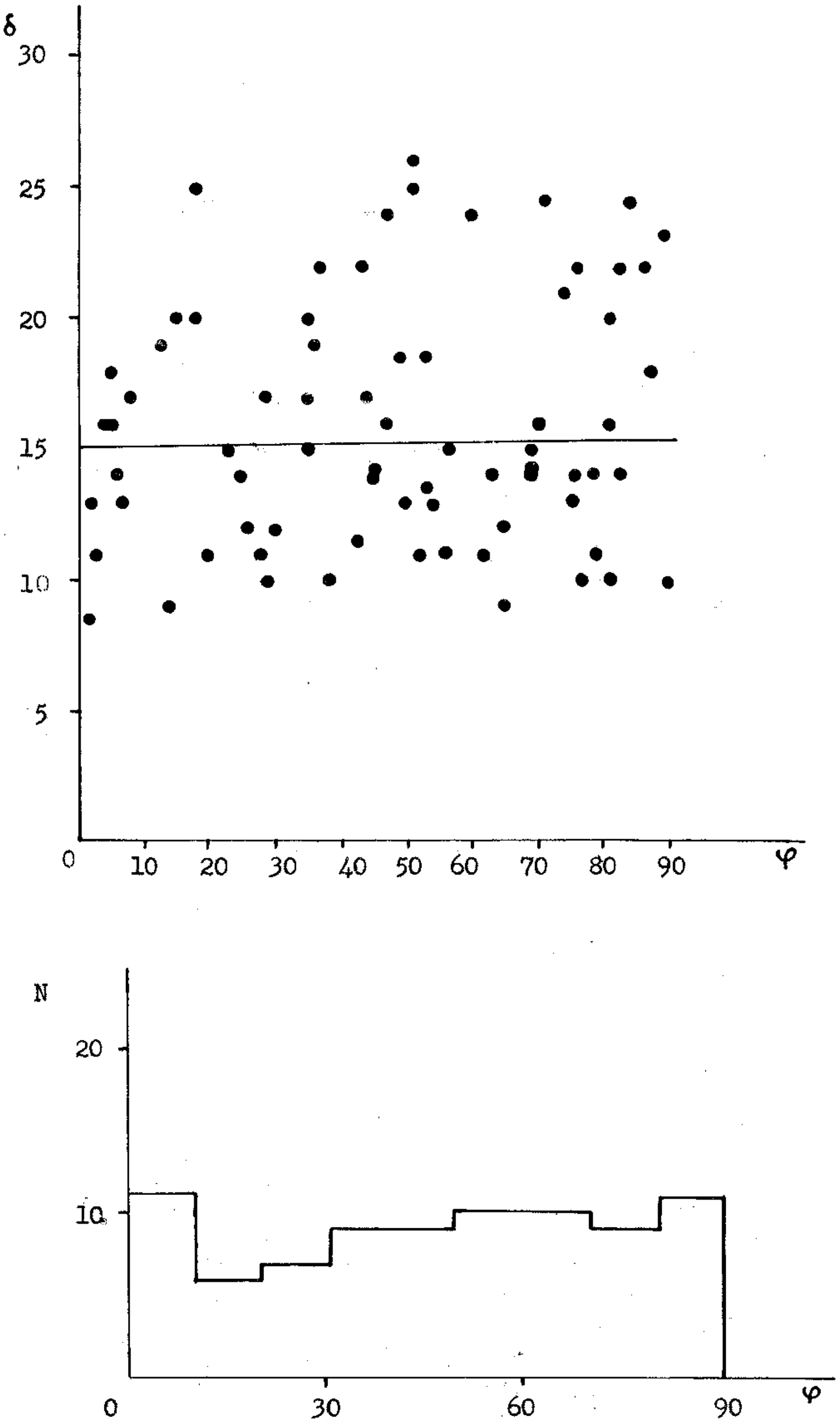

Fig. 5.: a) Illustrating the absence of correlation between δ and φ; the nearly horizontal line is the least-square regression straight line; b) angular distribution of non collinear events.