# ANGULAR CORRELATION BETWEEN FRAGMENTS AND CHARGED PARTICLES EMITTED IN URANIUM FISSION

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Fission events induced by 660-Mev protons in uranium nuclei, involving the emission of charged particles, have been observed in P-9 nuclear emulsions. The angular distribution of these particles, with respect to the direction of the incident beam and the fragment line of flight, has been measured. It has been established that the protons do not exhibit an angular correlation with the fragments; however, the possibility has not been excluded that some of the protons are emitted by the fission fragments. The  $\alpha$  particles and multiply-charged particles have a tendency to be emitted at large angles with respect to the fragments. Possible factors which may be responsible for the observed correlation between the fragments and  $\alpha$  particles are discussed.

## INTRODUCTION

EXPERIMENTS on fission in heavy nuclei induced by high-energy nucleons, performed with nuclear emulsions, indicate that fission is frequently accompanied by the emission of protons,  $\alpha$  particles, and heavier particles, which are emitted from the fission site. In a photoemulsion, such a disintegration event looks like a star containing two additional dense fragment tracks ("star" fission). $1^{-4}$  The existence of an anisotropy with respect to the beam direction (approximately 60 percent of the particles are emitted in the forward hemisphere) leads us to assume that some of these particles are emitted by the nucleus in the first or cascade stage of the disintegration process. An estimate of the fraction of cascade protons, based on the magnitude of this anisotropy, is in agreement with the results of a Monte Carlo calculation. The isotropic distribution of low-energy charged particles can, in most cases, be associated either with evaporation from the nucleus prior to fission or emission from excited fragments. Comparing the number of charged particles observed in fission of heavy nuclei with the theoretical number expected on the basis of a model in which there is preliminary emission by the fissioning nucleus, Shamov<sup>3</sup> concludes that the  $\alpha$  particles and protons are evaporated from the nucleus prior to fission. In a paper by Ivanova and P'ianov,<sup>5</sup> however, it is indicated that a comparison of this kind does not allow us to distinguish emissive uranium fission from a fission which occurs in one of the intermediate levels of an excited nucleus. A calculation carried out by these authors has shown that the sum of the number of charged particles emitted by the uranium nucleus in cooling to approximately half the initial excitation energy, plus those emitted by the fragments, is equal to the number of particles which would be emitted from the urnaium nucleus before it is completely cooled prior to fission.

One of the factors that permit appraisal of the validity of various fission models at high excitation energies is the existence or absence of an angular correlation between the paths of the evaporated particles and the fission fragments.<sup>6</sup> An experiment on correlation of neutrons and fragments, similar to the well-known Fraser experiment on thermal neutrons, was carried out by Harding and Farley, who investigated uranium fission induced by 147-Mev protons.<sup>7</sup> Harding and Farley concluded that the majority of neutrons (approximately 80 percent) are emitted by the nuclei prior to fission. However, this result does not exclude the possibility that an increase in the energy of the bombarding particles to 500 - 700 Mev might result in the development of an intranuclear cascade and that the increased excitation energy might lead to a situation in which some of the highly-excited nuclei undergo fission from one of the higher levels.

At the present time, there is apparently only one paper on the angular distribution of light charged particles with respect to the fragments in fission induced by fast nucleons.<sup>4</sup> In a paper by Belovitskii et al.<sup>4</sup> a study was made of uranium fission induced by neutrons obtained by charge-exchange from 480-Mev protons. It was found that protons were emitted isotropically, whereas the  $\alpha$  particles have a tendency to be emitted at right angles to the fragment line of flight. Unfortunately, because of poor statistics (27  $\alpha$  particles) this result was not very conclusive.

In the present work we have undertaken to investigate, with better statistics, the angular distribution of charged particles with respect to fragments in uranium fission by high-energy protons. Since an angular correlation is to be expected only for particles of evaporation origin, the investigation was carried out with a low-sensitivity emulsion which could be exposed to a higher flux of bombarding protons.

#### EXPERIMENTAL ARRANGEMENT

In this work use was made of plates with P-9 fine-grain nuclear emulsion prepared in the laboratory of Professor N. A. Perfilov. The thickness of the sensitive layers was 100 and  $200\mu$ . The emulsions were sensitive to 30-Mev protons. The uranium was introduced by impregnating the emulsion in a water solution of NaUO<sub>2</sub>(C<sub>2</sub>H<sub>3</sub>O<sub>2</sub>)<sub>3</sub>.

The plates were exposed in the extracted 660-Mev proton beam from the synchrocyclotron of the Joint Institute for Nuclear Research. Some of the plates (first series) were exposed in a proton flux which was parallel to the plane of the emulsion; the second group was exposed perpendicularly to the beam. For protection against slow neutrons the plates were placed in a cadmium cover with double walls, between which borax was placed (a layer 1 cm thick). The emulsions were processed in the usual way, using temperature development. The scanning was carried out with a MBI-3 microscope with a magnification of  $5 \times 90 \times 1.5$ ; the measurements were performed at maximum magnification. The angles were measured with an ocular angle gauge accurate to  $1^{\circ} - 2^{\circ}$ ; the accuracy of the length measurements in the vertical and horizontal projections was approximately  $0.5\mu$ .

The first series of plates yielded the spatial angular distribution of the particles with respect to the fragments. For this purpose measurements were made of the inclination of each track to the plane of the emulsion and the angle between the projections of the tracks on this plane. The shrinkage factor for the emulsion was determined as by Vigneron<sup>8</sup> and Gramenitskii and Podgoretskii<sup>9</sup> from the observation of the  $\alpha$  particles due to the natural radioactivity of the uranium in the same plates. The shrinkage factor was found to be  $1.85 \pm 0.15$ . The same value was obtained by direct measurement of the emulsion thickness before and after development, using other plates under analogous experimental conditions.

In the second series of plates, which were exposed perpendicularly to the beam, only the angular distribution in projection was taken. In fission induced by high-energy particles the fragments frequently fly apart at angles less than 180° so that in these cases the angles could be read by using the lines connecting the ends of the fragment tracks.

The identification of the tracks of charged particles in "star" fission events was carried out visually. The reliability of this method of distinguishing singly and doubly charged particles (with preliminary training of the observers) has been indicated by Serebrennikov<sup>10</sup> and was verified in the present work. The error in assigning tracks to one or the other of the groups was less than 5 percent. Doubtful cases were not included in the experimental statistics.

### EXPERIMENTAL RESULTS

In all, 3201 cases of uranium fission were recorded, including 1156 in the plates of the second series. The prong distribution of the fissions (i.e., the distribution over the number of light charged particles emitted in fission) was as follows: 0 prongs -43.7%, 1 prong -25.3%, 2 prongs -15.7%, 3 prongs -8.5%, 4 prongs -4.2%, 5 prongs -1.5%, 6 prongs -0.8%, 7 and more prongs -0.3%. The ratio of the number of  $\alpha$  particles to the number of protons was  $0.35 \pm 0.02$ . The angular distributions of these particles with respect to the direction of the proton beam are shown in Figs. 1 and 2. The proton distribution is in agreement with the results obtained by Shamov<sup>3</sup> as far as the front-back ratio is concerned; however,  $\alpha$ -particle distribution obtained in Ref. 3 is isotropic, the present data indicate that a considerable fraction of the  $\alpha$  particles are to be associated with an anisotropic emission process.

The table lists the particle distribution over the angle formed by the particle path and fragment path (projected on the plane of the emulsion), under various exposure conditions. The last row of this table contains the anisotropy coefficient, defined as the ratio of the number of tracks lying in the interval  $60 - 90^{\circ}$  to the number of tracks at angles  $0^{\circ} - 30^{\circ}$  with respect to the fission line. Statistical errors are indi-

Projected fragment angle (degrees)	Parrallel exposure		Perpendicular exposure		Total number of particles		
	Protons	$\alpha$ particles	Protons	$\alpha$ particles	Protons	$\alpha$ particles	Protons with $Z \ge 3$
$\begin{array}{c} 0-15\\ 15-30\\ 30-45\\ 45-60\\ 60-75\\ 75-90\end{array}$	$233 \\ 242 \\ 274 \\ 260 \\ 264 \\ 245$	85 81 87 87 108 97	230 210 234 213 210 214	67 61 77 78 86 97	463 452 508 473 474 459	152 142 164 165 194 194	4 2 4 2 7 8
$\frac{N_{60-90^{\circ}}}{N_{0-30^{\circ}}}$	$\begin{vmatrix} 1.06 \pm \\ \pm 0.09 \end{vmatrix}$	$\begin{vmatrix} 1.23 \pm \\ \pm 0.18 \end{vmatrix}$	$0.96 \pm \pm 0.09$	$\begin{array}{c} 1.43\pm\\\pm0.24\end{array}$	$^{1,02\pm}_{\pm^{0,07}}$	$\begin{vmatrix} 1.32 \pm \\ \pm 0.14 \end{vmatrix}$	$\begin{array}{c} 2.5 \pm \\ \pm 1.6 \end{array}$



FIG. 1. Angular distribution of protons with respect to the beam in star fissions (projection).

FIG. 2. Angular distribution of  $\alpha$  particles with respect to the beam in star fissions (projection).



FIG. 3. Angular distribution of protons with respect to fragments, reduced to unit solid angle: the dashed line is the measured distribution; the solid line is the corrected distribution; the smooth curve is calculated from the barrier-fission model.

FIG. 4. Angular distribution of  $\alpha$  particles with respect to fragments, reduced to unit solid angle: the dashed line is the measured distribution; the solid line is the corrected distribution; the smooth curve is calculated from the barrier-fission model. cated. In the last column are given data on particles with charge greater than 2. Some of the tracks assigned to the triply charged particles may actually be those of  $\alpha$  particles, mistaken because of the inadequate visual resolution between these particles. In Figs. 3 and 4 are given the spatial angular distributions of the particles, as obtained by measuring tracks in plates of the first series and converting to unit solid angle.

## DISCUSSION OF THE RESULTS

A comparison of the data of the table and the distributions shown in Figs. 3 and 4 by the dotted lines indicates a certain discrepancy. Figure 3 seems to indicate a preference for emission of protons in the direction of the fragments, whereas the measurements of angles in projection on the same plates do not corroborate this finding. The origin of this discrepancy may be in errors arising in taking the spatial distribution or errors in the shrinkage factor. In order to establish the effect of the shrinkage factor on the shape of the spatial distribution the latter was plotted a second time, using 400 arbitrarily chosen tracks of protons from star fissions and taking a new value of the shrinkage factor, 2.2. No noticeable systematic change in this distribution was found. There is still one other factor which has a bearing on the shape of the angular distribution of the tracks. The spatial distributions of the fragments and the  $\alpha$  particles due to natural radioactivity observed under the same conditions indicate that some of the tracks (approximately 25 - 30percent) are at large angles (60°  $-90^{\circ}$ ) with respect to the plane of the field of view and are lost

in scanning the plates. Since the light charged particles have a certain directivity in the forward direction, even in the absence of any physical correlation between these particles and the fragments the experimentally determined distribution will be anisotropic with respect to the fragment line of flight. It is easily demonstrated that there will be an excess of cases exhibiting small angles between the track of the charged particles and the fragment. An estimate based on the data of Figs. 1 and 2 and on the observed fragment angular distribution with respect to the beam shows that the coefficient for this expected parallel anisotropy is about 1.05.

A comparison of the angular dependence for particles emitted in the given hemisphere, in which the directivity is less pronounced, with the corresponding dependence in the backward hemisphere corroborates the "independence" effect. In the observation of angles in the plane of the field of view this effect vanishes since the angular distribution of the acceptable cases is isotropic.

It may be assumed that the presence of cascade particles, directed predominantly along the beam, and the perpendicular anisotropy for uranium fission fragments with respect to this direction, also lead to a certain angular (perpendicular) correlation between the light particles and the fragments. Using the data of the present work (Figs. 1 and 2) and that of Ref. 11, in which the fragment angular distribution was obtained, we have computed the correlation associated with the superposition of the two distributions. The calculation was carried out numerically using the relation

$$\Phi(\varphi) = \int_{-\pi/2}^{\pi/2} f(\alpha) F(\varphi - \alpha) d\alpha,$$

where  $\Phi(\varphi)$  is the angular distribution of the prongs with respect to the fragments,  $f(\alpha)$  and  $F(\varphi - \alpha)$  are the distribution functions for the fragments and charged particles respectively with respect to the beam direction; the first function is taken from Ref. 11 while the second is based on the data in Figs. 1 and 2.

It would appear that for protons the perpendicular anisotropy factor is not greater than 1.03; for  $\alpha$  particles it is not greater than 1.02. It is obvious that this effect should not be observed in the perpendicularexposure experiment. This coincidental correlation was actually not observed for protons in the plates of the second series; for  $\alpha$  particles, however, the perpendicular-directivity effect was observed.

In Figs. 3. and 4 the solid lines show the spatial distributions with the above-mentioned factors taken into account. A comparison was made of the experimental distribution and that to be expected on the barrier-fission model. In Figs. 3 and 4 the solid curves show the calculated results (normalized to the number of experimental tracks). In carrying out the calculation it was assumed that fission occurs symmetrically starting at an initial excitation energy of 150 Mev which is divided equally between the two fragments. The mean fragment temperature at which charged-particle emission occurs is 2 Mev. The particle velocities were determined from the energy spectra, which were plotted taking account of the thermal expansion of the nucleus and barrier penetrability.<sup>12</sup> The kinetic energy of the fragments was taken as 80 Mev. The deviations from 180° in the direction of flight were not taken into account in the calculation.

Although the distributions shown in Fig. 3 and the results of the observations of the plates in the second series indicate the existence of a certain parallel anisotropy for the protons with respect to the fragments, it is felt that this effect cannot be attributed to the barrier mechanism of uranium fission since the departures from isotropy are within the limits of the statistical fluctuations. It is possible that a certain number of protons are emitted by the fragments, this is not inconsistent with the available data on the energy spectrum. As far as the  $\alpha$  particles are concerned, the picture is somewhat more definite, since emission of these particles by fragments is not very probable and a perpendicular anisotropy is observed.

In experiments on uranium fission by slow neutrons it has been established that  $\alpha$  particles with energies on the order of 20 Mev are emitted in a direction close to 90° with respect to the fragment line of flight with a probability of 1 in 400.<sup>13 - 14</sup> With an increase of the bombarding-neutron energy to 14 Mev the emission of  $\alpha$  particles becomes still less probable<sup>4</sup> as an explanation for the correlation observed in the present work. It is possible that in fission induced by high-energy protons there is an effect analogous to the emission of  $\alpha$  particles in fission at low excitation energies, with the sole difference that we are not dealing with the effect due to two individual fragments of the fissioned nucleus which are still fairly close, but rather two parts of a highly deformed elongated nucleus which is in a pre-fission stage. The effect of the electric field of these parts is of greater importance for particles with higher charge and hence the perpendicular directivity is more pronounced for  $\alpha$  particles than for protons. This mechanism applies only for part of the particles emitted prior to fission because some of the particles are emitted at an instant when the nucleus is not deformed; these particles are not correlated with the oscillation directions of the nucleus. In conclusion we may note that the observation of an angular correlation of this type is most conveniently carried out with perpendicular exposure since in this case the spurious factors associated with the directivity of the cascade particles are minimized.

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