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ANGULAR DISTRIBUTION OF FRAGMENTS FROM THE PHOTOFISSION OF U^{238}

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ABSTRACT

Ilford Dl and Eastman NTC nuclear emulsions have been loaded with a solution of Uranium acetate and then exposed to the X-ray beam of the Case betatron operated at 18 Mev. The developed plates have been searched with a Zeiss type microscope. A total number of 1062 fission tracks have been measured and analyzed and the angular distribution of the fission fragments in center of mass system has been obtained. The angular distribution has been fitted by the method of least squares to a function of the form

$$F(\theta) = a + b\sin^2\theta + c\sin^2 2\theta,$$

where $b\sin^2\theta$ is the dipole distribution and $c\sin^2 2\theta$ is the quadrupole distribution. The least squares solution gave the following ratios:

$$b/a = -\ 0.0097 \quad \text{and}$$

$$c/a = 0.0896$$

The negligible dipole component found in the track distribution is in agreement with other experiments at these energies.[1] The quadrupole coefficient, whose statistical significance we attempted to determine, has not hitherto been reported. Such a quadrupole term is to be expected when the wave length of the gamma rays is comparable to the nuclear diameter, and probably results from direct electromagnetic interaction between photons and the nuclear proton distribution.

(1) E. J. Winhold, "Photofission of Thorium and Uranium," Ph.D. Thesis Department of Physics, M.I.T., September, 1953.

TECHNICAL REPORT ON THE PHOTOFISSION OF U^{238}

I. INTRODUCTION

The general character of the photofission process resulting from the absorption of a photon by a nucleus is an electrodynamic interaction rather than a nuclear one. In the photofission of U^{238} study described in this report the principal competing reaction with photofission is the reaction U^{238} (γ,n) U^{237}. However, since the present work is performed with nuclear emulsions, there are no serious complications introduced by this latter process or other nuclear events which may occur. The photofission of U^{238} is characterized by a broad resonance with X-ray energy, as shown in Fig. 1, the peak of the resonance curve falling near 14 Mev where the cross section has a value of about 0.17 barns.(2)

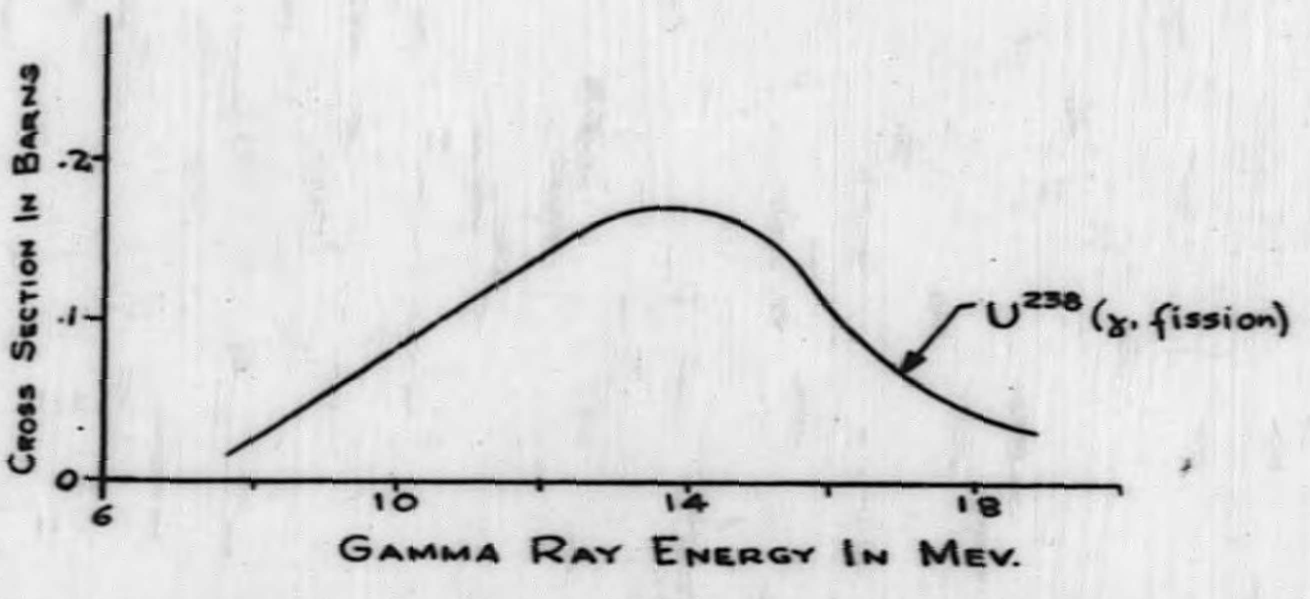


Fig. 1. The cross-section for reaction U^{238} (γ, fission) vs. the energy of the gamma ray.

(2) R. B. Duffield and J. R. Huizenga, Phys. Rev. 89; 1042 (1953)

When the process is investigated with the continuous bremsstrahlung spectrum from a betatron the most efficient maximum energy for operation of the betatron as shown by Fig. 2 is at 18 Mev.[3]

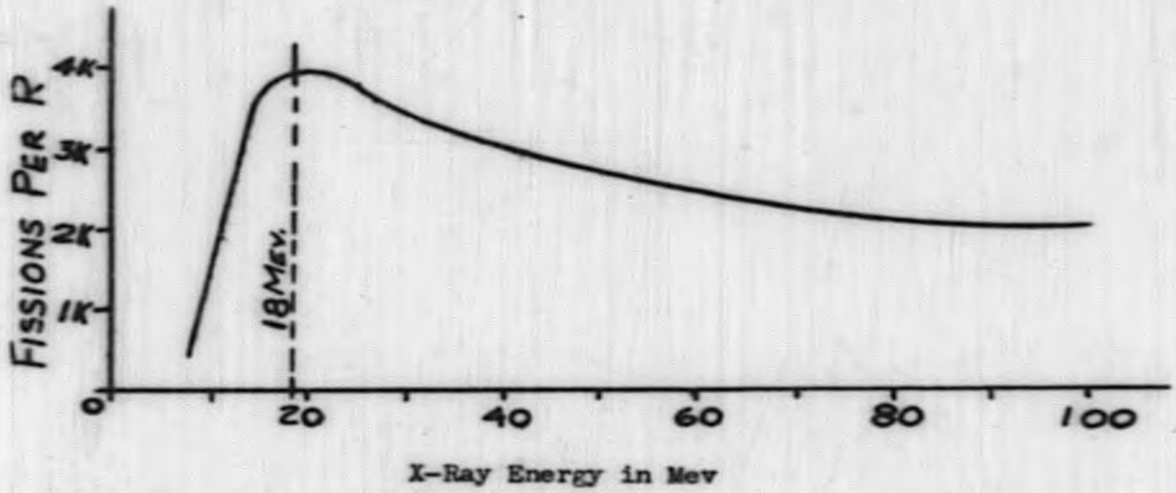


Fig. 2. X-Ray Fission Yield for Uranium.

Although the present investigation is concerned with binary fissions only, nevertheless, the plate scanning has revealed a number of tripartition fission events as noted and discussed below.

The angular distribution of the fission fragments shows a large isotropic component in center of mass system.[4] This is to be expected on the collective model theory of the atomic nucleus which assumes that the sharing of the energy of the X-Ray quantum takes place among many particles and modes of oscillation of the nucleus in which any angular relationship with respect to the initial X-Ray beam is entirely lost by the time fission occurs. If after absorption of the quantum of energy, a sufficient time elapses for the increased energy to be distributed fairly uniformly throughout the nucleus, the fission fragments should on the average be ejected isotropically with respect to the incident X-Ray beam.

(3) G. C. Baldwin and G. S. Klaiber, Phys. Rev. 71, 3 (1947)

(4) At the photon energies used in these experiments the corrections from laboratory to center of mass space are negligible.

On the simple collective model theory the time required for this energy distribution is very short and the predominant angular distribution of the fission fragments should therefore be isotropic.

In photonuclear reactions where a large change in the value of e/m results from the transformation (as in the $_3Li^6\ (\gamma,\ p)_2He^5$ reaction) the distribution of fragments should be that corresponding to an electric dipole interaction, with maxima at 90° to the incident X-ray beam. It was originally anticipated that this effect would be small in heavy nuclei such as U^{238}, for in these the strong exchange coupling would tend to preserve a uniform e/m ratio throughout the nucleus and thus suppress any electric dipole moment. This may alternatively be described by saying that the neutron-proton distribution in a nucleus is hard to change, and since the dipole effect would necessarily move protons with respect to the rest of the nucleus, and this process due to the strong coupling is hard to accomplish.

The dipole component has the form of the second term in $a + b\sin^2\theta$ where the angle θ is measured from the direction of the X-ray beam to the axis of the two fission fragments. The presence of a dipole moment is most directly explained on the independent particle model of the nucleus. When the interaction of an X-ray photon takes place with a single proton, as has been shown by Hill and Wheeler,(5) on the basis of this model, selective absorption takes place by protons in orbits of high angular momentum whose planes are perpendicular to the incident X-ray beam. The X-ray will be absorbed with a changed angular momentum of one unit ($\Delta m = \pm 1$) and with the plane of the proton's motion unchanged by the process. The subsequent fission in this case will occur predominantly with the fission fragments ejected at 90° to the X-ray beam. Due to the

(5) D. L. Hill and J. A. Wheeler, Phys. Rev. 89, 1102 (1953)

X-ray energies used in this experiment one should and does observe a very small dipole effect. This is consistent with other work.[(6)]

In addition to the isotropic and dipole distributions of fission fragments a quadrupole component in the distribution may also exist, giving fission fragment ejected at 45° to the incident X-ray beam in all quadrants. The quadrupole component would result from direct interaction of high energy X-ray photons with the nucleus. The X-rays must be of sufficient energy so that their wave lengths are comparable to the size of the nucleus. Under these circumstances a phase lag will occur in the interaction between the radiation and the nuclear protons giving rise to the quadrupole effect.

The present experiments verify the well established dipole component and its relationship to the isotropic component in the angular distribution of photofission fragments in the case of U^{238} and in addition in this study we have searched for a quadrupole component. The details of the experimental methods used and the results obtained are given below.

II. The Quadrupole Component in the Distribution of Fission Fragments.

If there is a direct and practically instantaneous interaction between the electric vector of the X-ray and protons in the Uranium nucleus a more complex distribution of the fission fragments would be anticipated. In particular, when the wave length of the X-rays is comparable to the nuclear diameter, then phase lags will exist in the electromagnetic forces acting on the protons throughout the nucleus. As a consequence the electromagnetic forces acting on the protons in the nucleus at a particular instant will be as shown in Fig. 3(a), with the result that protons on opposite sides of the nucleus experience transverse

(6) M.I.T. Nuclear Physics Report, May 31, 1954

forces in opposite directions, both perpendicular to the X-ray beam. This system of forces will produce not only a body force but also a shear and the latter will cause nuclear oscillations along a line making a 45° angle with the X-ray beam. This mode of oscillation of the nucleus shown in Fig. 3(b) is a fission mode in which the fission fragments would be ejected at 45° angles with respect to the incident X-ray beam. Exact estimates of nuclear diameters are somewhat uncertain, but on the basis of the recent high energy electron scattering experiments carried on at Stanford University the value 1.5×10^{-12} cm for the diameter of the Uranium nucleus seems probable. The wave length of the 18 Mev X-rays ($\lambda = 6.9 \times 10^{-12}$ cm) from the betatron used in these experiments is about 4 times this value, so it is expected that directional effects will be found if the quadrupole interaction is significant. It is expected at the energies of this experiment, that any magnetic dipole interaction between photon and nucleus is too small to be detected.

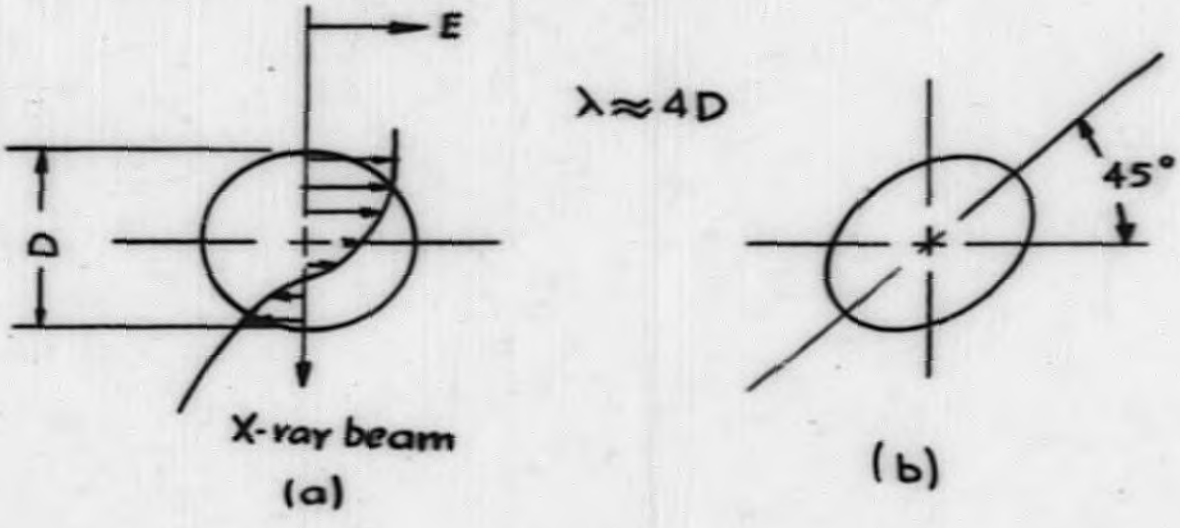


Fig. 3

III. Preparation and Exposure of Nuclear Emulsions.

The special types of nuclear emulsions used in this research were selected by trial with the following considerations in mind. It is especially important to use emulsions in which the background of alpha particle tracks is as low as possible. This criterion is satisfied by the Eastman Kodak nuclear plate type NTC, and by the Ilford plate type D1. A series of trials established the necessary conditions for the impregnation and processing of these emulsions so that they do not record any proton, deuteron, alpha particle, meson or electron tracks, but will give clear and useable tracks of the highly ionized fission fragments. Exposure to large amounts of alpha particles will of course cause eventual blackening, but even in this case these images are composed of a network of fog grains without definite track structures.

The schedule employed with the Kodak NTC and Ilford D1 plates were as follows. The plates were first dried, then accurately weighed, then soaked in a solution of uranium acetate for sixty minutes, after which they were washed with distilled water, and then again dried and weighed. The uptake of the uranium compound by this procedure is rather uniform. Weighing the plates before and after treatment showed that the average number of uranium atoms absorbed in the emulsion is about $1.8 \times 10^{20}/cm^3$.

After exposure of the plates in the X-ray beam of the betatron they were developed by the following procedure:

1. Place in distilled water at room temperature.
2. Cool to 5°C.
3. Add sufficient Kodak D 19 developer to obtain 1:4 solution. Allow to stand for one hour at 5°C.
4. Place in 1:20 D 19 solution for 45 minutes with mild hydrogen agitation.
5. Place in 2% acetic acid, stop at 5°C with mild hydrogen agitation.

6. 60% hypo solution at room temperature until clear plus 50% of time required to clear. Replenish hypo if necessary.
7. Place in acid fix for one hour.
8. Wash under running tap water at least six hours.
9. Wash in distilled water for one hour.
10. Dry in horizontal position.

The plates were exposed in the bremsstrahlung beam of the Case Betatron operated so that the maximum energy of the photons was 17 Mev. The nuclear plates as mounted in the X-ray beam were oriented with the plane of the emulsion set at 1° to the center line of the X-ray beam. This orientation is to prevent excessive absorption of the X-rays throughout the length of the emulsion. The plates were placed at about 18 inches from the target of the betatron. At this distance the betatron target subtends an angle of less than 1°, so that no collimating aperture was needed. No absorbers were placed in the X-ray beam except the thin copper discs used to monitor the beam as described below.

The exposure times of the plates in the betatron beam were of the order of ten minutes and at the position where the plates were mounted the intensity was approximately 15 roentgens per minute. To reduce the fission produced in U^{238} by secondary neutrons from the betatron, the plate holders were surrounded by sheets of cadmium metal. The exposure of the emulsions in the X-ray beam was determined both by readings of a Victoreen dosage meter and by the induced radioactivity in the above-mentioned thin copper discs placed in front of the plates. The time which the copper monitoring discs were exposed to the X-ray beam was accurately measured and then the induced radioactivity was immediately measured with standardized Geiger counter apparatus.

IV. Scanning of Nuclear Emulsions and Nature of Data.

When the plates were prepared and exposed as described above they were then scanned for fission tracks with Zeiss type BS 48/77 nuclear emulsion microscope whose magnification power was approximately 1000. The identification of fission tracks needed only the elimination of possible alpha particle tracks. These latter were identified as to appearance and number by examining several control plates which had been treated in the manner described above but had not been exposed to the betatron beam. After proper practice and training the eye could easily identify fission tracks as compared to alpha particle tracks. The fission tracks are considerably more dense, are more symmetrical about their central point and have a different average length from those of the alpha tracks. In explaining the quantities measured with the microscope the following terminology will be used (see Fig. 4):[7]

xy plane - the plane of the emulsion

z - the axis normal to the xy plane. Also, the observed length of the projection of a track on this axis.

L_z - the length of the projection of a track on the z axis corrected to the true value in the unprocessed dry plate.

L_{xy} - the length of the projection of the track in the xy plane.

L - the track length or range in the dry plate.

α - the angle of a track's dip into the emulsion measured from the xy plane (0 - 90°).

ψ - the projection in the xy plane of the smaller angle between a track and the beam (0 - 90°). Experimentally this angle was read 0-180°, but in calculations it was converted to 0 - 90°

θ - the space angle between forward directions of a track and the beam. (0 - 90°)

φ - the azimuth of a track about the beam measured from the xy plane. (0 - 180°) angle is obtained by calculations.

(7) V. E. Krohn and E. F. Shrader, "Use of D_2O Loaded Emulsions," AEC Report, March 16, 1951.

Also from Fig. 4 we can derive the following equations:

(1) $L = \sqrt{L_{xy}^2 + L_z^2}$

(2) $\text{Tan}\alpha = L_z/L_{xy}$

(3) $\text{Cos}\,\theta = \text{Cos}\,\alpha\ \text{Cos}\,\psi$

(4) $\text{Tan}\,\alpha = \text{Tan}\,\varphi\ \text{Sin}\,\psi$

(5) $\text{Sin}\,\alpha = \text{Sin}\theta\ \ \text{Sin}\,\varphi$

The following quantities were obtained directly from measurements with the microscope: (1) the projected angle (ψ) of the fission track in the plane of the emulsion with respect to the incident X-ray beam, (2) the projected length (L_{xy}) of the fission track in the xy plane, (3) the projected length (z) of the fission track normal to the xy plane. Also a sketch is made of the track projected in the xy and the yz plane. Now a track can be fully described by its range, L; its angle with the beam, θ; its azimuth, φ; and knowledge concerning the quadrant into which φ has been measured. All the quantities can be obtained by using the above formulae.

Owing to gelatin shrinkage the inclination of the track is altered, and the angle of dip as measured in the gelatin must be corrected before the track length is computed. The shrinkage is dependent on the volume of soluble materials removed during processing, and its value can be approximated from the volume of the silver halides on the assumption that the fixing process leaves no voids after the gelatin dries. The correction that is made is of the following form:

$$S = 1 + \frac{\text{Volume of Extractables}}{\text{Volume of Gelatin}}$$

Knowing the density of gelatin (1.31) and silver (6.47) and that the ratio of silver to gelatin by weight is 2.7 the shrinkage factor was found to be 1.54.

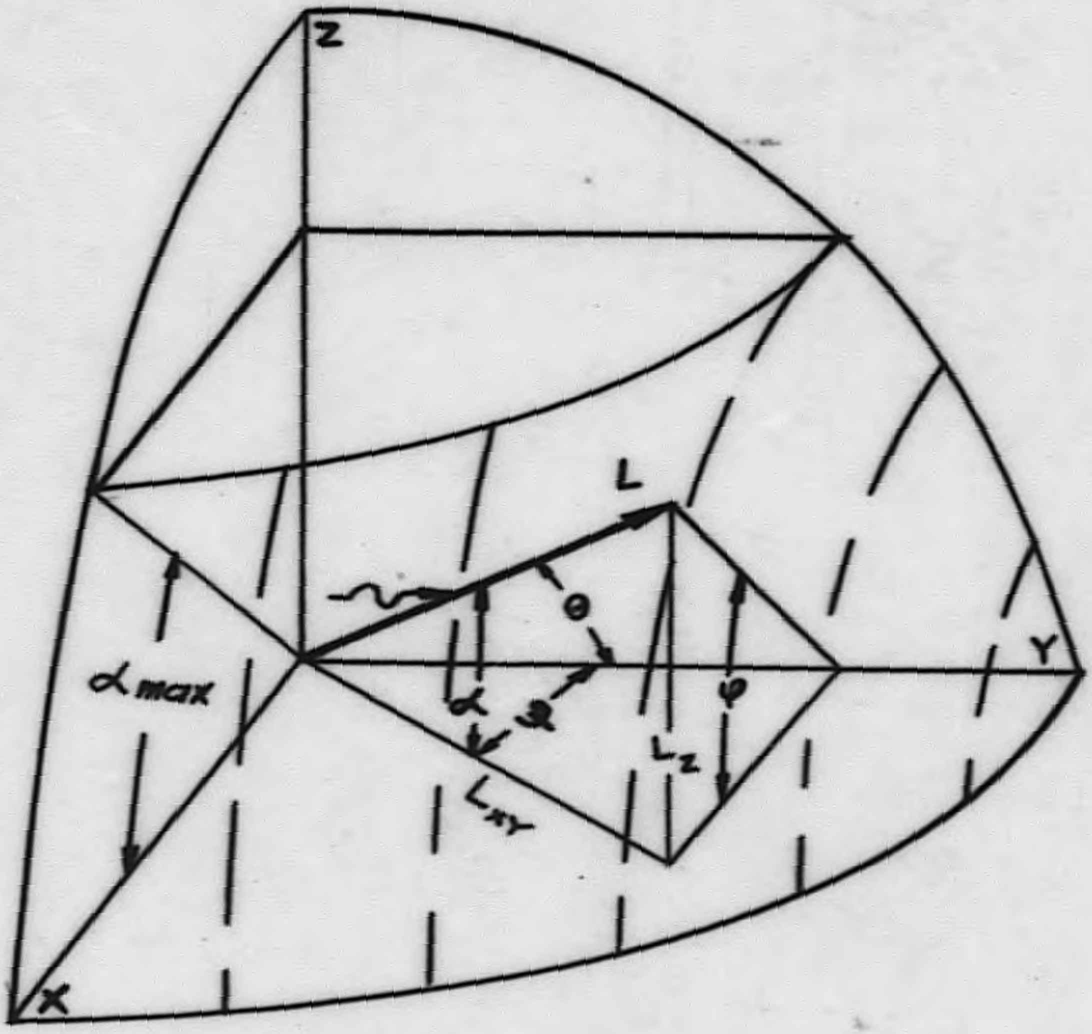


Fig. 4.

The observed quantities L_{xy}, L_z, and ψ , and the quantities desired to describe a fission track, L, θ, and φ , for the case where the gamma ray beam passes in the plane of the emulsion (the xy plane).

Besides the correction for the shrinkage one also has to correct for the errors in the distribution caused by the discrimination against the tracks with an angle of dip, α , in excess of an accepted maximum dip angle, α_{max}, and also the discrimination against the tracks which did not lie completely within the emulsion. The correction factor, W, is a θ - dependent number greater than one by which the number of tracks at a given angle θ must be multiplied by in order to render the data free of the aforementioned errors. W is derived from the following considerations. The requirement that the measured tracks have dip angles equal to or less than some α_{max} restricts the direction of tracks in that they must lie within the solid angle generated by the surface of a sphere of radius R and two parallel planes located at distances of $R \sin\alpha_{max}$ from the center of the sphere. A track can be pictured as having one end at the center of the sphere and the other end intersecting the sphere. Thus the point of intersection of the track and the sphere must lie between or on one of the two parallel planes. Let the two parallel planes be

$$Z = \pm R \sin\alpha_{max}$$

The tracks at a given space angle θ will lie in the differential solid angle of

$$d\Omega = 2\pi \sin\theta \, d\theta$$

However, if $\theta > \alpha_{max}$ only those tracks lying within the part of the solid angle between the two planes will be recorded. To correct for this discrimination the number of tracks dN recorded at a given space angle must be multiplied by the ratio

$$W' = 2\pi / 4\beta$$

where $4\beta/2\pi$ is the fraction of the differential solid angle lying between the two parallel planes.

$$\frac{1}{W'} = \frac{4\beta}{2\pi} = \frac{2}{\pi} \arcsin\left[\frac{R \sin\alpha_{max}}{R \sin\theta}\right]$$

$$= \frac{2}{\pi} \sin^{-1}\left[\frac{\sin\alpha_{max}}{\sin\theta}\right]$$

Since only those tracks lying completely within the emulsion were recorded some of the tracks satisfying the maximum dip angle requirements still were not recorded. This discrimination is also a space angle dependent function. The correction goes as follows: Assume that all of the tracks are of the same length, "l". Also assume that the fissions that caused the tracks took place at the midpoint of these respective tracks. These conditions are not satisfied exactly by the data, but it was felt that discrepancies so introduced would be of second order. Now going back to the considerations that led to the correction for the maximum dip angle criterion, it is clear that the two parallel planes which limit the dip angle are now at $z = \pm$ "l"/2 $\sin\alpha_{max}$ since R = "l"/2. It is also clear that for fissions that took place at a distance, D, from the surfaces of the emulsion greater than

$$D = Z = \pm \frac{l}{2} \sin\alpha_{max}$$

there was no discrimination of tracks other than for dip angle greater than α_{max}. However, for fissions which took place closer to the surface of the emulsion than $D = \frac{l}{2} \sin\alpha_{max}$ the maximum allowable dip angle, σ, that a track may have is defined by

$$\sin\sigma = 2D/\text{"l"} \qquad D < (\text{"l"}/2) \sin\alpha_{max}$$

This leads to a dip angle discrimination function of

$$\frac{1}{W'} = \frac{2}{\pi} \sin^{-1}\left[\frac{2D}{l \sin\theta}\right] \qquad D < 1/2 \sin\alpha_{max}$$

The total discrimination function for tracks throughout the emulsion can be found by averaging 1/W' over the total thickness, t, of the emulsion. Let D be the distance from one surface of the emulsion to the location of a fission. The total discrimination function 1/W is

$$\frac{1}{W} = \frac{2}{t} \int_0^{l/2 \sin\alpha_{max}} \frac{2}{\pi} \sin^{-1}\left[\frac{2D}{l \sin\theta}\right] dD + \frac{2}{t} \int_{l/2 \sin\alpha_{max}}^{t/2} \frac{2}{\pi} \sin^{-1}\left[\frac{\sin\alpha_{max}}{\sin\theta}\right] dD$$

Advantage has been taken of the symmetry about the center plane of the emulsion, that is the averaging was done only from $D = 0$ to $D = t/2$, thus simplifying the algebra considerably. Integrating and substituting in the limits we get

$$\frac{1}{W} = \frac{2}{\pi}\left[\sin^{-1}\left(\frac{\sin\alpha_{max}}{\sin\theta}\right) - \frac{1}{t}\left(\sin\theta - \sqrt{\sin^2\theta - \sin^2\alpha_{max}}\right)\right]$$

Fig. 5 gives the correction factor W for each θ as computed from the above equation. This curve was computed for a maximum angle of dip $\alpha_{max} = 23^{\circ}$.

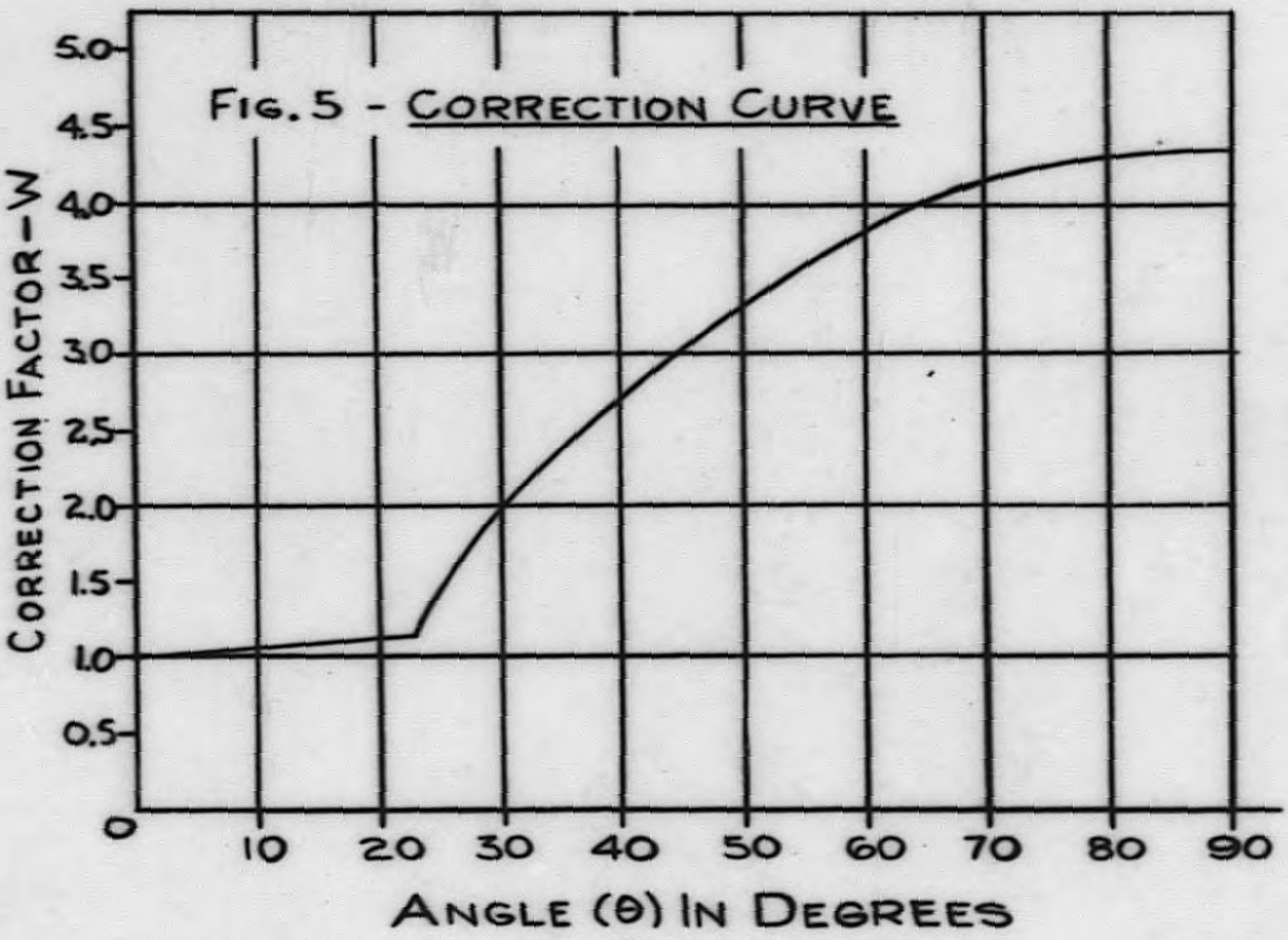


FIG. 5 - CORRECTION CURVE
CORRECTION FACTOR - W
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0
10
20
30
40
50
60
70
80
90
ANGLE (θ) IN DEGREES

V. The Experimental Results.

In order to analyze the data we have to separate the isotropic, dipole and quadrupole components from the angular distribution graph. This is accomplished in the following manner. Fig. 6 is a graph constructed by plotting as ordinate for each value of the angle θ as abcissa, the total number of fission tracks having θ values out to this angle. The ordinate points as plotted also include the weighting factor W as discussed above. Fig. 7 shows the envelope drawn through the points plotted in Fig. 6, and also the least squares fit for this curve.

The form of the differential distribution curve which the data was expected to fit was

$$dN/d\Omega = a + b\mathrm{Sin}^2\theta + c\mathrm{Sin}^2 2\theta$$

where the constants a, b, and c may be identified with the isotropic, dipole and quadrupole contributions respectively. Or expressing $d\Omega$ in terms of the space angle θ

$$dN/d\theta = 2\pi \left[a + b\mathrm{Sin}^2\theta + c\mathrm{Sin}^2 2\theta \right] \mathrm{Sin}\,\theta$$

Integrating this with respect to θ gives the integral distribution curve.

$$F(\theta) = 2\pi \int_0^\theta (a + b\mathrm{Sin}^2\theta + c\mathrm{Sin}^2 2\theta)\,\mathrm{Sin}\,\theta\, d\theta$$

$$F(\theta) = (a + b)\,(1 - \mathrm{Cos}\,\theta) - 1/3\,(b - 4c)\,(1 - \mathrm{Cos}^3\theta) - 4c/5\,(1 - \mathrm{Cos}^5\theta)$$

This curve is of the same form as the experimental curve of Fig. 6. The constants a, b, and c were determined by means of a least squares fitting of $F(\theta)$ to the experimental curve subject to the condition that $F(\pi/2)$ fitted the experimental curve exactly at $\pi/2$. These values then give Fig. 7 which shows the best fit of the experimental curve to the theoretical curve. The values obtained for a, b, and c by this method are

$$b/a = -0.0097$$

$$c/a = 0.0896$$

The value of b/a thus obtained can be compared with the values of b/a obtained by E. J. Winhold at M.I.T. using an entirely different technique. Their results

EXPERIMENTAL INTEGRAL DISTRIBUTION OF FISSION FRAGMENTS

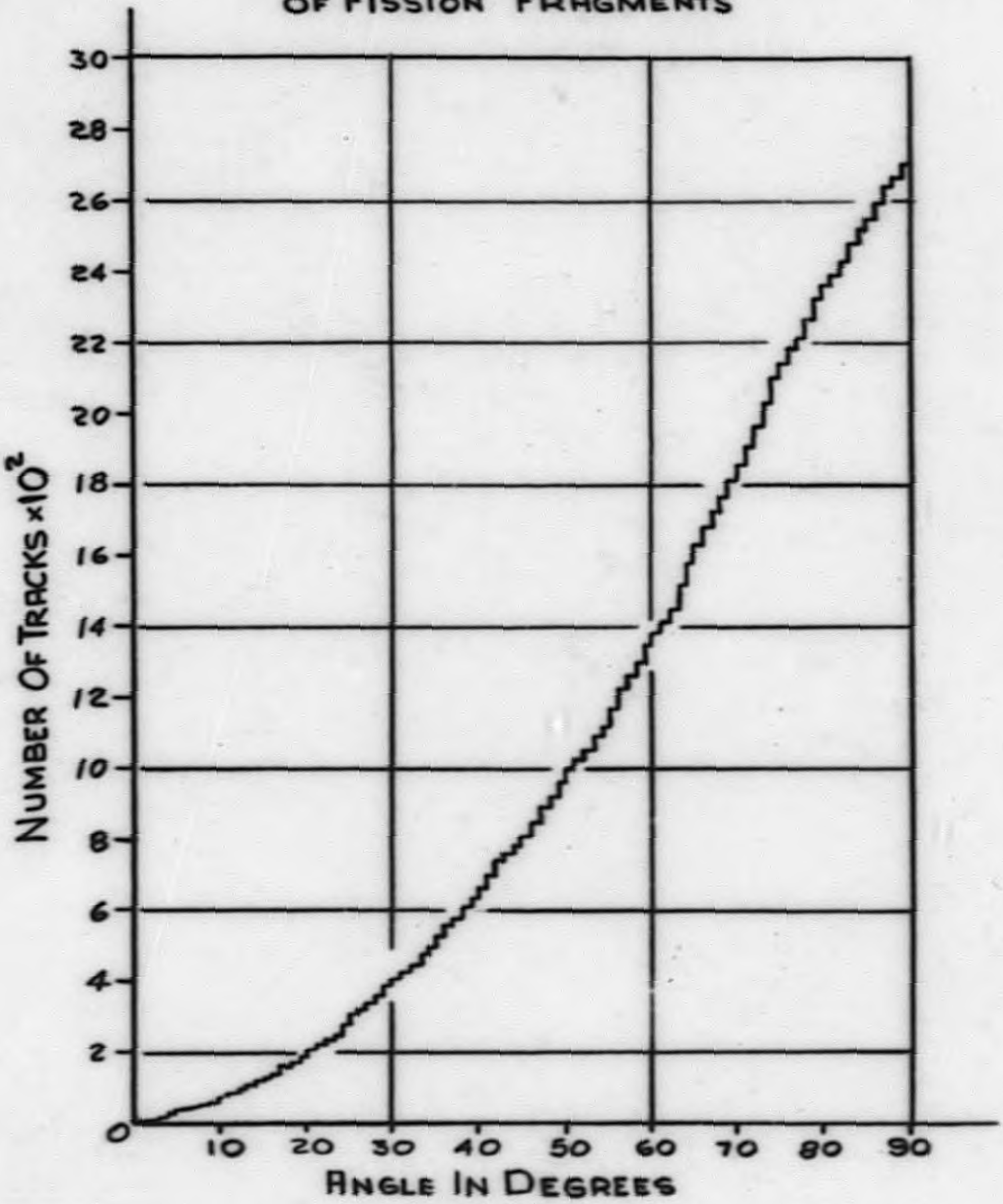


FIG. 6

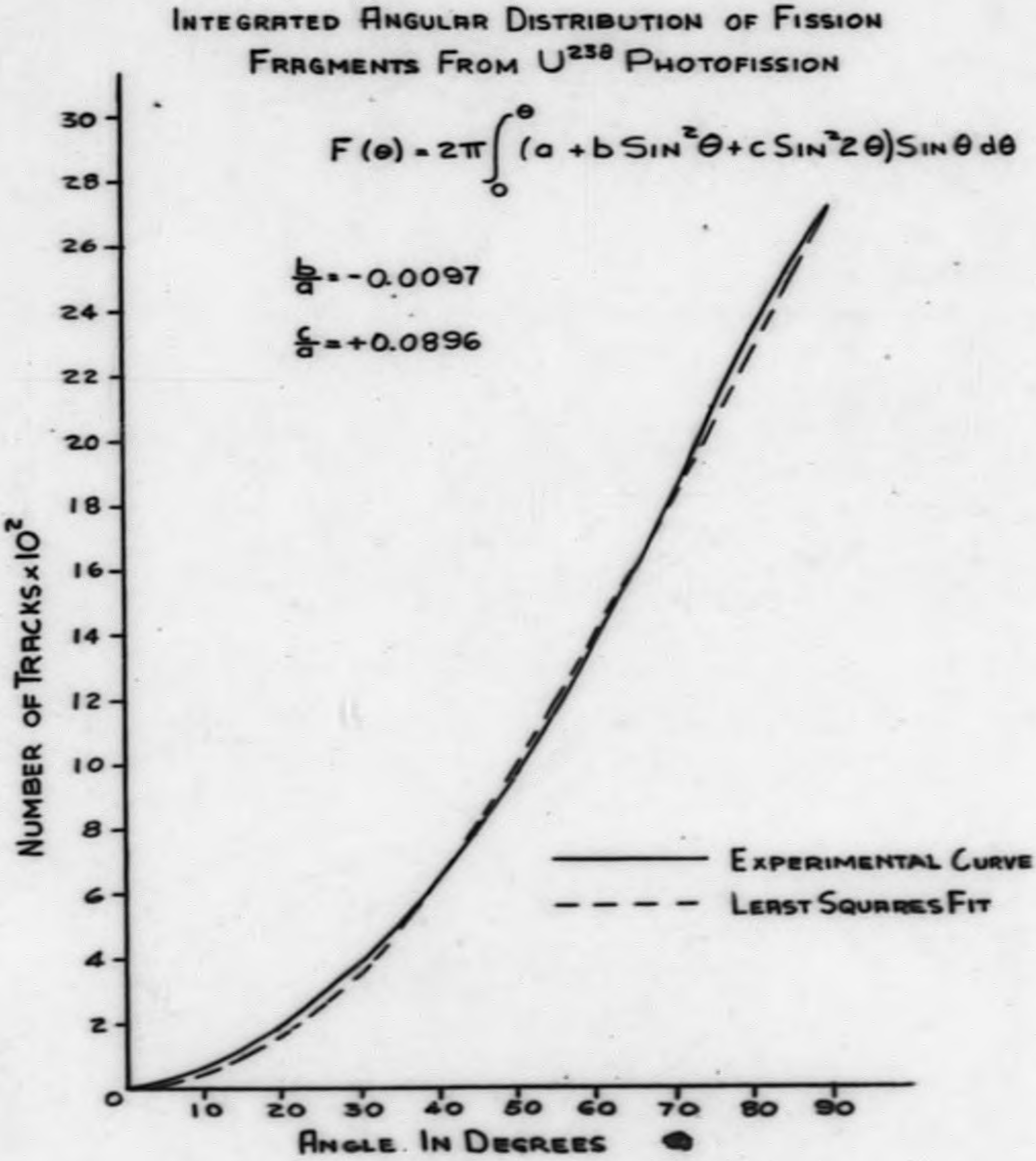

INTEGRATED ANGULAR DISTRIBUTION OF FISSION FRAGMENTS FROM U^{238} PHOTOFISSION
$F(\theta) = 2\pi \int_0^\theta (a + b \sin^2\theta + c \sin^2 2\theta) \sin\theta \, d\theta$
$\frac{b}{a} = -0.0097$
$\frac{c}{a} = +0.0896$
NUMBER OF TRACKS x 10^2
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
10
20
30
40
50
60
70
80
90
ANGLE IN DEGREES
EXPERIMENTAL CURVE
LEAST SQUARES FIT

FIG. 7

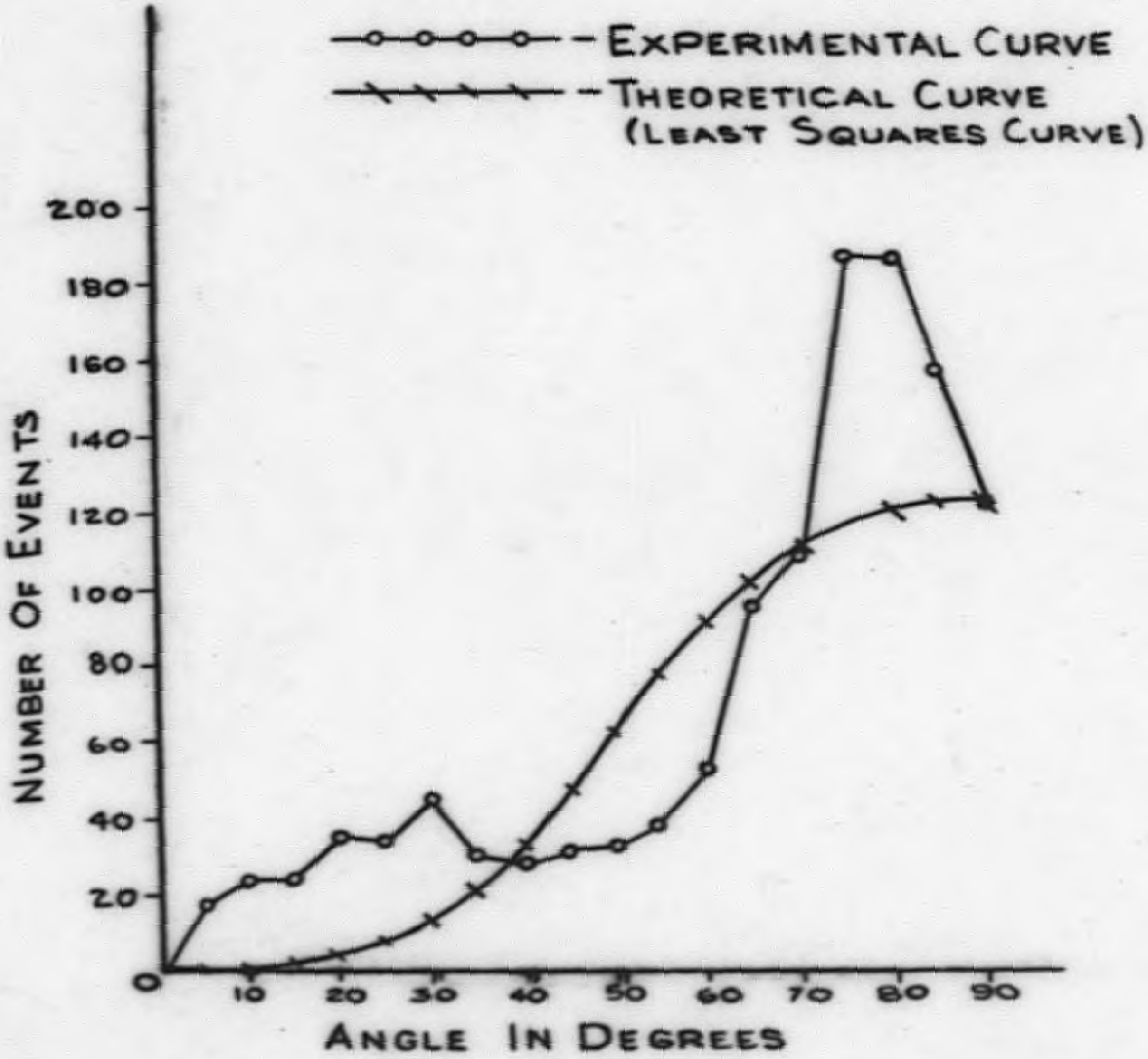
give b/a - 0.7 at 8 Mev, with a steady decrease in value of b/a to 0.1 at 13 Mev. It is also of interest to note that the M.I.T. experiments give b/a values for Th^{232} which are about twice the corresponding values for U^{238}, while for U^{235} the observed value of b/a is zero.

Now after having determined the necessary constants for the different angular distribution components, the isotropic and the dipole components are subtracted from the total distribution curve. This then gives Fig. 8. Also from the least squares value for the quadrupole component a second curve is plotted on Fig. 8. Comparing these two curves a tentative conclusion can be drawn that the quadrupole effect of 10% of the isotropic component as determined by this experiment has statistically very small significance, since we can change the values of C quite considerably without introducing a great change in the probable error.

The fission tracks in the emulsion have also been measured to give their total length. These average to 22.9 microns, but there is a considerable spread in the observed total length as shown in the distribution curve plotted in Fig. 9. It is not possible to identify with certainty the position where the uranium atom undergoes fission, but the total length as shown is directly related to the total energy of fission of the two fragments. The variation in track length is due in part to straggling in the photographic emulsion and also to the varying distribution in energy between the two fission fragments.

In scanning the nuclear plates a record has also been kept of the number of fission events in which three tracks occurred. In a total of 1062 fission events there have been observed a total of 9 tri-partitions. In two cases all three tracks were approximately equal length. In three cases there were the two usual long prongs plus a short prong at the position of the U^{238} nucleus. In four events, in addition to the two usual fission fragment tracks, there is a very long track, which is undoubtedly due to an alpha particle. Two examples of

FIG. 8 - QUADRUPOLE COMPONENT OF FISSION FRAGMENT DISTRIBUTION



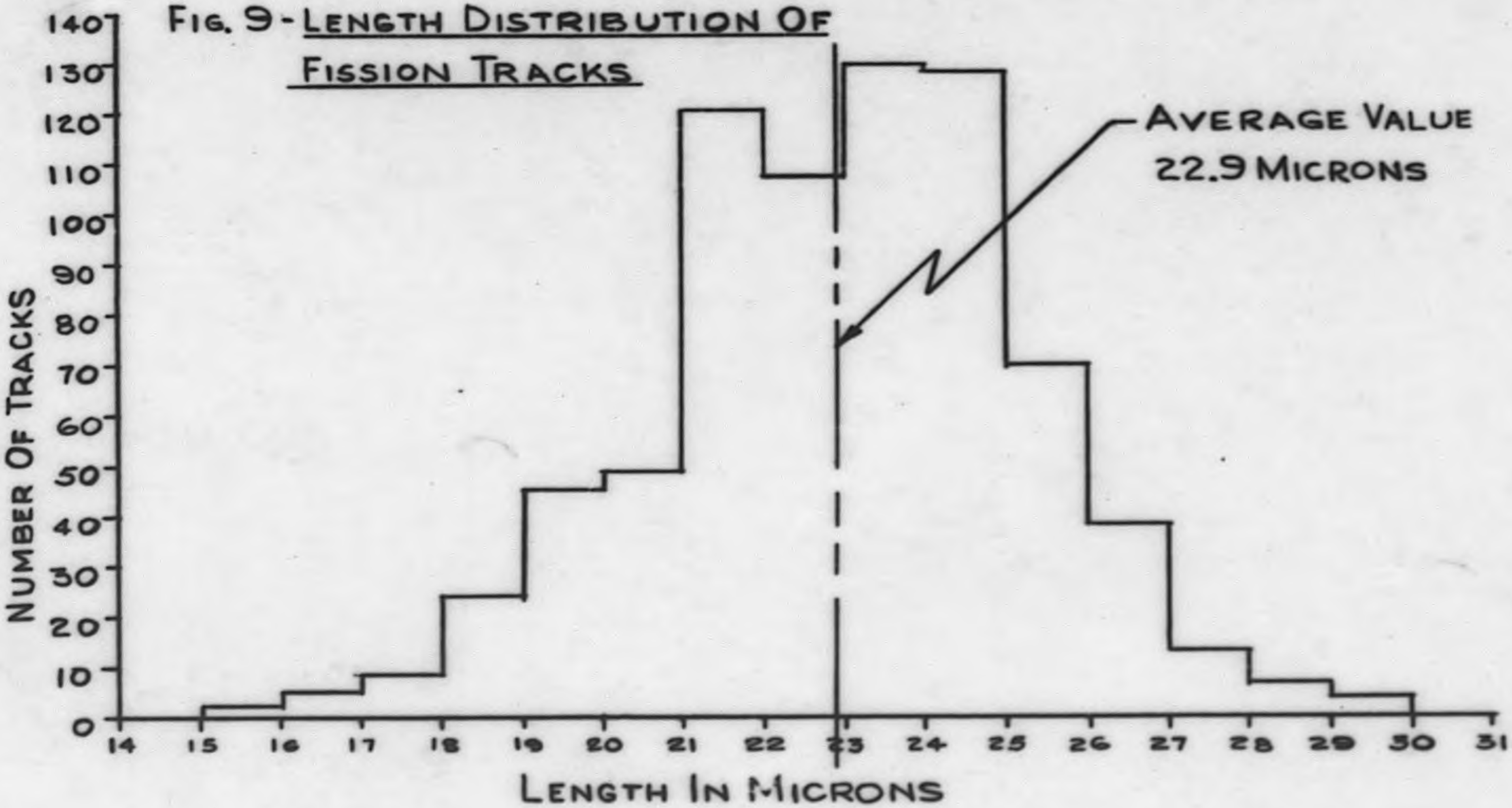


FIG. 9 - LENGTH DISTRIBUTION OF FISSION TRACKS

four-pronged stars have been observed. In one of these all four tracks were of comparable length and in one event there were three very short prongs and one long track. We also observed one multiple-pronged star in the plate, presumably due to a cosmic ray event. The total area of emulsion searched with the microscope was 4.5 square centimeters.