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April, 1951

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Abstract

Several batches of plates loaded with uranyl acetate have been exposed to π^- mesons in the Berkeley cyclotron. The mesons were generated by the 350 Mev proton beam striking a carbon target and the plates were shielded from the direct beam, or positive particles generated at the target, by several inches of copper. They received negative mesons over a wide range of energies. About one in a hundred of the mesons observed to end in these plates, ended in a characteristic fission event. Twenty-two such fissions were observed. This was in rough qualitation agreement with what might be expected if it is assumed that the capture of a negative π meson by the uranium nucleus always produces fission. Three of these events showed 3 way fission, a light particle coming off at about 90° to the tracks of the heavy fission fragments.

MESON INDUCED FISSION

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I. Introduction

The capture of negative π mesons in nuclei is well understood in a qualitative sense. The π^- is captured on a single proton leading to a neutron and 140 Mev excitation energy. In accordance with the conservation laws the energy of the primary process can become available to a heavy particle only in the presence of at least an additional nucleon. On the basis of various models[1] the probability can be calculated that either the neutrons of the primary process or the "recoil nucleon" will escape the nucleus. This will lead to two experimental consequences: 1. There will be more than the statistically expected number of single high energy fragments in a star initiated by a σ^- (π^-) meson. 2. The excitation energy corresponding to the number of star fragments observed is less than 140 Mev; it is, in fact, not more than 100 Mev. These inferences agree with experimental observation.[2]

π^- capture in U^{238} can shed some additional light on the qualitative correctness of the above description. Since the excitation energy of the residual nucleus, even after initial loss of a fast particle is above the fission threshold one would expect fission to accompany essentially all cases of π^- capture in U^{238}. This should behave like ordinary "fast" fission; one might expect a large frequency of single fast particles accompanying the fission event; these would derive from the primary capture event. This paper deals with a study of this process.

The work reported here was interrupted before its normal completion. Results given are thus often incomplete and obvious improvements or extensions of method will suggest themselves to the reader. The work is reported here at this time since 1) the results are sufficient to indicate the qualitative correctness of the capture process as outlined above, and 2) the techniques used here might be of more general interest.

II. Experimental Set-up.

Mesons were produced in the internal target of the 184-inch cyclotron at Berkeley in the usual way.[3] The mesons were allowed to fall upon 200 μ nuclear plates previously treated with uranyl acetate. No channeling or specific energy selection of the mesons was used. The plates were exposed in stacks at 12" from the primary target.

III. Plate Loading Technique.

The technique of uranium soaked plates for the study of fission (especially when accompanied by alpha emission) has been previously used by Tsien San-Tsiang and coworkers,[4] Wollan, Moak and Sawyer,[5] Green and Livesey,[6] and recently by Leona Marshall.[7] E. Broda[8] has studied the effects of time, temperature, concentration, ρH , and presence of other ions on the uptake of the uranyl ion. This is an adsorption process, so that the emulsion acquires a higher concentration of uranium than the solution in which it is soaked. None of these investigators required or wanted sensitivity to such lowly ionizing particles as fast mesons.

The loading of photographic plates is generally discussed in Yagoda's book.[9] One must choose a soluble compound such that neither the anion or cation will have undesirable effects on the gelatin or the photographic sensitivity. Chlorides cause swelling and reticulation of the emulsion. Warmth, humidity, adverse ρH, and oxidizing agents such as nitrates will in time wipe out tracks that have been formed in a photographic emulsion, presumably by oxidation of the latent image. The time factor is ignored by too many writers when speaking of "limits" of concentration, or the "impossibility" of loading with certain compounds. Thus a plate which had been soaked in a 10 percent solution of lead acetate, exposed, and developed the next day was completely clear; but a plate soaked in a 20 percent solution, exposed, and developed within a few hours, showed good meson tracks. Of course, with this lead concentration it is necessary to remove the lead before developing, or the plate is rendered white and opaque by lead hydroxide.

These plates were loaded by soaking in a saturated solution (about 4 gm/100 cc) of uranyl acetate for 15 - 20 minutes after a presoak in distilled water, since wet plates were found to take up the uranyl ion faster than dry. The plates were then rinsed in water, and in 95 percent ethyl alcohol, and dried in warm air for about one-half hour. They were wrapped in black paper, exposed, and then developed as soon as practical, to prevent undue accumulation of the uranium alpha tracks and desensitization. Later batches of plates were rinsed 5 - 10 minutes with dilute acetic acid and water before developing, since some batches did not develop well except on the surface of the emulsion, and it was believed that the uranyl acetate might be interfering. The developer used was a 6:1 dilution of D19, and the time of development varied from 25 to 40 minutes depending on the type of emulsion (the thickness used was usually 200 μ).

The loading process does have a desensitizing action on the emulsion making it necessary to use a more sensitive emulsion than might otherwise be chosen. Kodak NTB plaes were found to be very useful; they have a low background grain density, and for some unknown reason the alphas and fissions have a characteristically different appearance, which is not true in the Ilford plates of corresponding sensitivity, C3. Later, electron sensitive emulsions, (Kodak NTB-3 and Ilford G-5) were used to avoid the possibility of missing any associated events.

IV. Technique of Observation.

The plates were scanned for mesons, rather than for fissions as first planned, because on most plates sensitive enough to show mesons well, alphas and fissions are not well distinguished, and the plates are full of uranium alpha tracks of a length (about 20 μ in the dry plate) comparable to the 25 μ overall length of the fission tracks. The more interesting or photogenic of the fissions were photographed by A. J. Oliver. (A fission is photogenic when it lies out flat on the plate rather than standing on end!) See Figure 1.

The task of counting all the mesons in a given area of a nuclear plate is not too simple. An inexperienced observer (the case here), using low power magnification for speed, can pick up most of the starforming mesons, but may miss non-starformers, or mistake a straggling proton for one, or not notice a single high energy star prong, or be unable to tell whether a meson track ends at the surface or outside the emulsion. Half of the mesons questionable for this last reason were counted. To avoid counting the same meson twice, it was necessary not to overlap on successive sweeps of the plate, or else record position and number of star prongs and thus weed out repetitions.

V. Determination of Uranium Concentration.

A reticule was placed in the eyepiece which marked out a square 63.5 x 63.5 μ on the plate (total diameter of field being about 150 μ). Alphas were counted from top to bottom of the emulsion, as long as over half the track lay in the square. The alpha tracks were recognizable by their characteristic density and length, although there was variation in both according as the track was formed when the plate was wet or dry. Also alphas that dived steeply appeared shorter and heavier and were difficult to distinguish; these were certainly not over 10 percent.

The thickness of the emulsion was assumed to be uniformly 200 μ.

The largest uncertainty so far as the determination of the amount of uranium in the plate is probably the time element. The time of soaking the plates in the uranium solution (15 - 20 minutes) and the time of development and washing

(35 - 50 minutes) are relatively large compared to the total time during which alphas are being formed (2 - 4 hours). In some cases the plates were washed with dilute acetic and water before developing in an attempt to remove the uranyl acetate, which was believed to interfere with developing. How effective this procedure was is not known, except that Broda[9] (using 25 μ emulsions) claimed it was very effective. In these cases development time was not counted as effective time for formation of alphas. In other case it was. Soaking time in uranyl acetate was counted because the uptake is at first rapid, dropping off exponentially with time.[9] Washing time after development was not counted; it is so small (10 minutes) as to make little difference.

As the firgures stand, (Table I) 4 - 8 x 10^6 alphas/cm^3/hour correspond to $\frac{4 - 8 \times 10^6}{9.0 \times 10^7} = .045 - .09$ gm U/cm^3. However, since the counts for the last batches of plates, on which most of the fissions were found, run between 4 and 5 x 10^6 alphas/cm^3/hour, we shall take 4.5 as a convenient figure giving .05 gm uranium per cm^3 emulsion, which is over twice the amount (about 0.02 gm/cm^3) in the saturated uranyl acetate solution.

Eastman Kodak data on the composition of the NTB emulsion was used, the composition of the G-5 was not available, but comparison of the variation in different types of Kodak emulsions would make one think the difference not significant, certainly for the accuracy here. The density of the dry emulsion is 3.64. Thus we have $\frac{.05}{3.64} = 1.4$ percent uranium by weight.

Table I

Plate	Type	Alphas Counted	Volume* Searched (cm^3)	Alphas per cm^3	Time in Minutes					Total eff. time (hours)	Alphas per cm^3 per hour
					Soaking	Handling	1st Washing	Developing	2nd Washing		
6392 (con.)	C-3	110	15.8×10^{-6}	7.0×10^{6}	15	45	20	25	10	1.33	5.3×10^{6}
6495 (con.)	C-3	142	7.9×10^{-6}	18×10^{6}	--	--	--	--	--	2.5-3	$6.0\text{-}7.2 \times 10^{6}$
8249	NTB	72	8.1×10^{-6}	8.9×10^{6}	15	160	0	25	10	3.33	6.7×10^{6}
8251	NTB	88	8.1×10^{-6}	11×10^{6}	15	160	0	25	10	3.33	8.3×10^{6}
9224	G-5	146	8.1×10^{-6}	18×10^{6}	20	200	15**	40	10	3.92	4.6×10^{6}
9235	NTB	35	3.2×10^{-6}	11×10^{6}	20	110	15**	40	11	2.42	4.5×10^{6}
9866	G-5	95	8.1×10^{-6}	11.7×10^{6}	25	150	10**	30	10	3.08	3.8×10^{6}
9868	G-5	270	23.5×10^{-6}	11.5×10^{6}	25	150	10	30	10	3.08	3.7×10^{6}
9876	G-5	113	7.3×10^{-6}	15.5×10^{6}	25	150	10	30	10	3.08	5×10^{6}

Table of data yielding information as to the uranium content of soaked plates exposed to mesons.

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*Volume searched equals area of field of view (7.9×10^{-5} cm^2 for first two plates and 4.04×10^{-5} cm^2 for the rest) times thickness of emulsion times number of fields counted.

**Washed in dilute acetic acid as well as water.

Table II

Element	Percent by weight	Comparative Z Ratio percent by wt. x Z	Comparative Atomic Ratio $\frac{\text{percent by wt.}}{\text{at. wt.}}$
Ag	47.1	20.5	.44
I	1.49	.62	.01
Br	33.9	14.8	.42
C	8.47	4.23	.705
H	1.17	1.17	1.17
N	3.06	1.53	.21
O	4.80	2.4	.30
U	1.4	.54	.05
	100	45.79	3.26

Tabulation of Atomic Composition of Loaded Plates

From Table II, number of atoms of uranium per atom emulsion is about 1 in 650, or less if the emulsion has picked up water. But since the probability of a meson being captured by the nucleus is usually assumed to be proportional to the charge,[10] we find after weighting with Z that 1 in 85 mesons might be expected to enter a uranium nucleus and possibly initiate fission. Note that the amount of water is no longer very important for this calculation.

VI. Results--Occurrence of Fission.

This observer found in the four successive batches of plates exposed in the fall of 1949: 1 fission among 72 mesons, 3 fissions among 405 mesons, 2 fissions among 226 mesons, 4 fissions among 443 mesons, totaling 10 fissions among 1146 mesons. 28 percent of the 820 mesons whose terminations were definitely recorded were non-starformers. This is the correct percentage for π^- mesons alone,[2,3] but low considering a possible 10 percent μ^- mesons. However, a few mesons with questionable terminations, omitted in this percentage calculation, were probably mostly non-starformers.

A second observer found 8 fissions among 915 mesons in the last (and best) batch of plates, and noted 40 percent of 590 mesons as non-starformers. 4 other fissions were found by two other observers in the second and third batches of plates.

This amounts to about 1 of every 100 observed mesons causing fission, which, considering the large uncertainties and the presence of μ^- as well as π^- mesons, is quite compatible with the assumption that all of π^- mesons entering uranium nuclei (calculated as 1 in 85) cause fission.

VII. Results--Characteristics of Meson Induced Fission.

Table III shows that the fission fragment tracks tended to be quite symmetrical, in agreement with observations of fission of U^{238}, as against the unsymmetrical fission of U^{235}, and of the length expected.

As to the reliability of the measurements, flat measurements can fairly readily be made with a well calibrated reticule in the eyepiece to within a micron, but measurements in depth depend upon the calibration of the fine adjustment screw on the microscope, the original and final thickness of the emulsion, and the uniformity of shrinkage. A 200 μ emulsion will shrink to about 85 μ when processed. This puts strains on the emulsion which may even cause it to peel away from the slide. If the slides are soaked in diluted glycerin they shrink less, but the shrinkage is unpredictable and the emulsion thickness will change with time. Thus the depth of a track as measured with the microscope is multiplied by the ratio

Table III

Data on Fissions

Date of Exposure	Plate	Flat Length	Depth Length	Total Length	Projected Range Ratio of Fission Fragments	Remarks
Aug. 28	7968	18±1	13±3	22±2	1.1	
Sept. 11	8249	20.5±1	17±2	26.5±2	1.0	
Sept. 11		9	24	25.5±?	2±?	Dives very steeply. Length and ratio unreliable.
Sept. 11	8250	26	3	26±1	1.2	
Sept. 11	8251	25.5	7	26.5	1.0	
Sept. 11	8252	17.5	23	29	1.1	Emergent alpha or proton at right angles.
Oct. 24	9224	25	10	26	1.25	
Oct. 24	9225	6	10	12	--	Measurements of 1 prong; other went out bottom.
Oct. 24	9231	23.5	6	24	1.1	
Oct. 24	9235	16	4	16.5	--	One end out top.
Dec. 11	9866	25	4	25	1.25	
Dec. 11	9866	18	17	25	1.25	Alpha or proton emerges at about 80°.
Dec. 11	9866	25	1-2	25	1.2	
Dec. 11	9866	6	33(26)	--	--	Dives sharply and one end out bottom. Also emulsion thinner at this point than in 3 places above making corrected length long.
Dec. 11	9865	25	14.5	28.5	1.4	
Dec. 11	9868			27.2		Alpha or proton emerges seemingly at different spot from meson entrance. Meson probably coincides with part of fission track

Table III (cont.)

Data on Fissions

Date of exposure	Plate	Flat length	Depth length	Total length	Projected Range Ratio of Fission Fragments	Remarks
Dec. 11	9873	24	7	25	1.0	
Dec. 11	9873	28	3.7	28	1.0	
Dec. 11	9876	18	14	23	1.4	
Dec. 11	9876	11	19.5	22.5	2	Length and ratio dubious because track dives steeply.
Dec. 11	9876	22.5	11	25	1.6	Track quite curved, perhaps emulsion distorted.
Dec. 11	9876	25.5	12.5	28.5	1.3	

These figures based on a standard ratio of 2.4 without measuring emulsion thickness, thus perhaps high.

On this last plate was also found a very interesting event which is the subject of another paper.[11]

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of 200 μ (assumed original thickness) to the measured thickness at that point (which varied from 70 to 150 μ). This assumes not only original thickness of 200 μ, but also uniform depth shrinkage.

It might also be noted that when the emulsion is wet the density (usually 3-1/2 - 4) is considerably lowered and the tracks fainter and longer. This was conspicuous in the case of the uranium alphas which were often about 1/4 longer than the 20 μ expected, but, of course, if the plates were even fairly damp, the cyclotron could not be operated. However, the moisture content on exposure was unknown.

The 22 events identified as fissions were in no way similar to usual meson stars. In this study no other stars were seen with 2 heavy prongs at 180°;

about one was seen with one heavy and one light prong at 180°. The only possible cause of confusion were two cases of one prong stars in which the meson track happened to lie over part of the one heavy prong, but this could be resolved with the oil immersion lens.

Three of these events showed a particle emerging at about 90° to the fission track. In two of these cases it was quite clear that the new particle emerged when the meson entered the fission; in the third case there seemed to be a short separation, but observation under high power made it seem probable that the meson track simply coincided with the fission track along this distance. The track was a light straight track, clearly a high energy alpha or proton which left the emulsion without ending, or scattering appreciably. One left after 200 μ corresponding to a proton of better than 5 Mev energy or an alpha of better than 25 Mev. This proton energy is far above that which could be caused by a knock-on, and the alpha energy higher than that found by L. Marshall for thermal U^{235} fission.[7] The frequency of this occurrence (1 in 7) is high compared to the frequency of alpha particle emission in thermal fission (1 in 250).

VIII. Discussion.

Despite the meager statistics obtained, these results confirm the prediction that π^- capture in U^{238} leads to fission in a large fraction of the cases. Also the occurrence of fast tracks in excess of those observed in neutron induced fission[7] is probably the phenomenon identical to the excess fast single tracks in π^- stars. Both these facts agree with the capture mechanism of Tamor.[1] Another fact of interest is the result that the energy of the fission fragments is the same as that in ordinary fission, despite the greater excitation. These results are in agreement with the results of Jungerman and Wright[2] on the energy release in fission induced by 90 Mev neutrons. Both their work and the data reported here confirm the idea that the fission fragment energy is derived from electrostatic forces alone and is independent of the excitation of the primary process.

It would also be interesting to expose plates loaded with other heavy elements to π^- mesons. Thorium and lead are practicable, but I did not succeed in loading plates with thorium. Mesons were counted in plates heavily loaded with lead, and no fissions were found where 2 - 5 might have been expected, if all π^- mesons captured in lead produced fission. This is not unreasonable, since the fission yield in lead could be quite low owing to the high excitation required.

I wish to express my thanks to Dr. W. K. H. Panofsky for suggesting and supervising the work, Dr. C. Richman for a suggestion as to method, Dr. H. Bradner for the use of the facilities of the film group, and for aid in exposing the plates, and to Miss Jocelyn Willat who spent many hours scanning the plates, and discovered 8 of the fissions observed.

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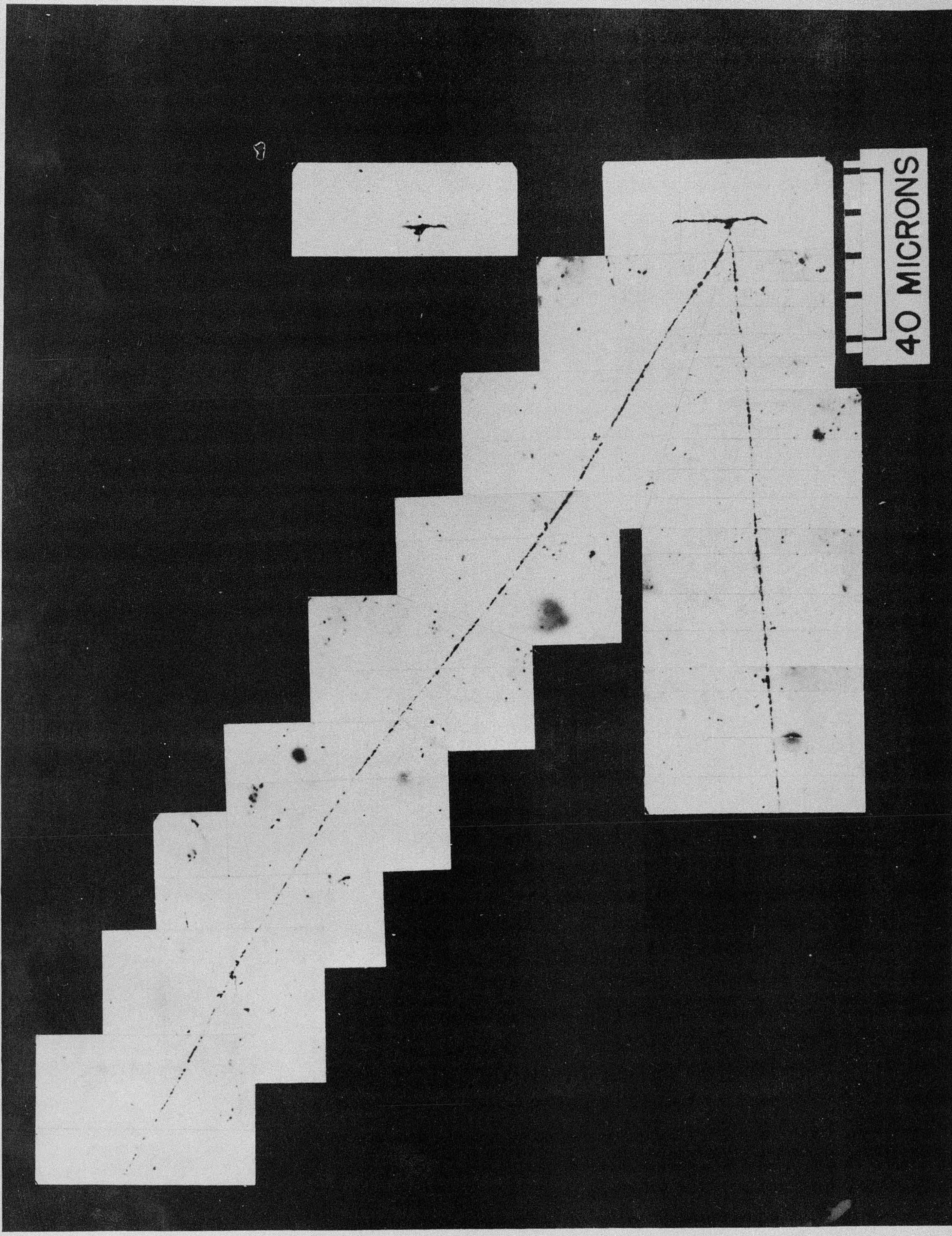
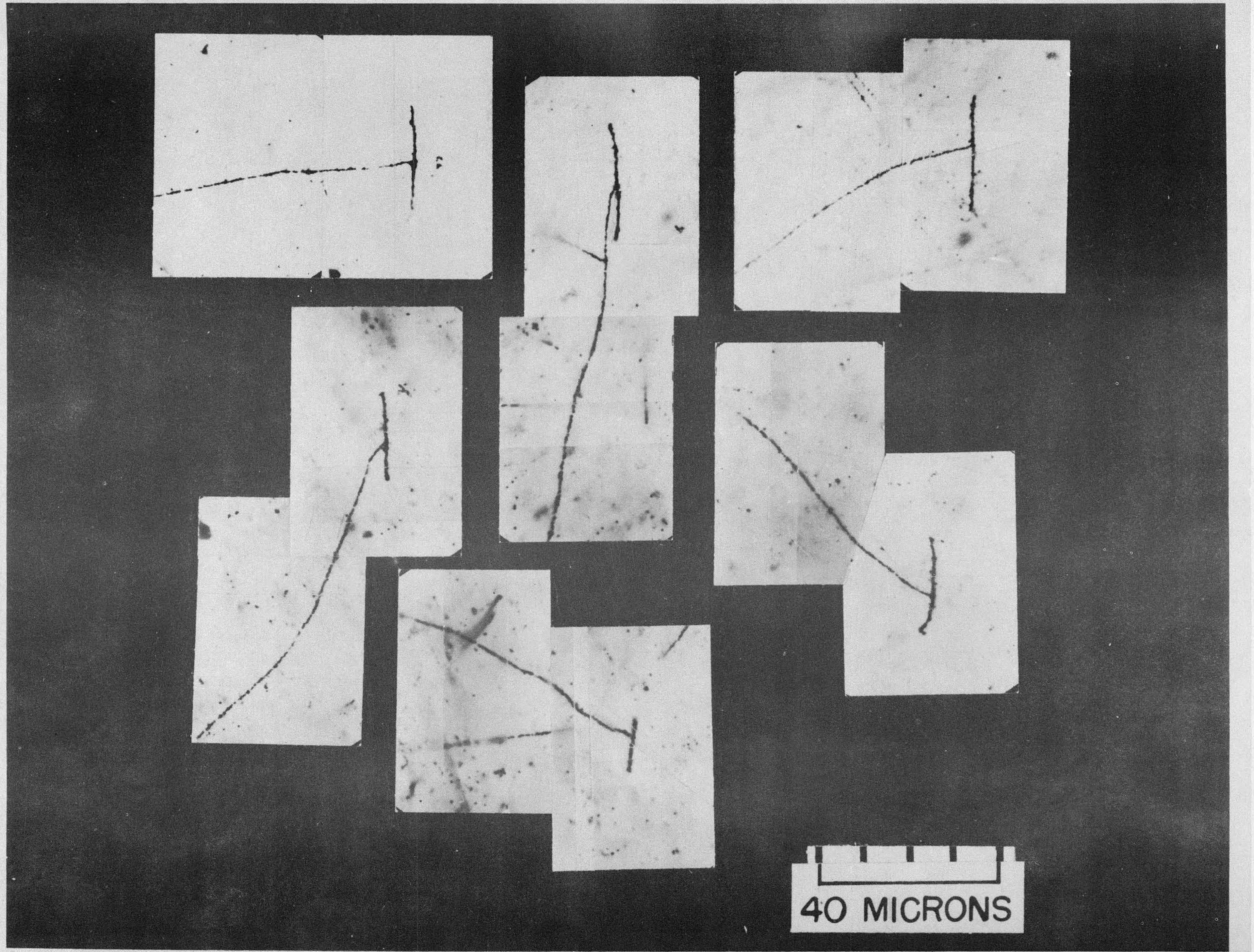
Figure Captions

Figure 1 a. Meson Induced Fissions in Uranyl Acetate Load G-5 Emulsion. (Mostly Plate 9876)

Figure 1 b. Meson Induced Fission with Third Particle. (Plate 9868)



ZN44



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