

Fission of Heavy Nuclei by Slow  $\pi^-$  Mesons\*

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Experimental data are given showing that on capture of slow  $\pi^-$  mesons by nuclei of U, Pb and W, fission of these nuclei occurs. Of the three nuclei studied, the fission probability is greatest for uranium, for which more than 300 cases of fission by slow  $\pi^-$  mesons were found. The distribution in range of the fission fragments and the existence of cases of fission with emission of fast charged particles (principally protons) show that in the end result the capture of a slow  $\pi^-$  meson by a nucleus is similar to the action on it of a particle with high ( $\sim 100$  mev) energy.

## 1. INTRODUCTION

WITH the production of  $\pi^+$  and  $\pi^-$  mesons under laboratory conditions great scientific interest was aroused in investigating the character of the interaction of these new particles with nuclei. In the capture of  $\pi^-$  mesons by elements of low and medium atomic weight (observations in photo-emulsions) the nucleus emits various kinds of particles: protons, neutrons,  $\alpha$ -particles and deuterons. The number of tracks of charged particles observed in the stars from the disintegration of nuclei by  $\pi^-$  mesons reaches 5 or 6. These data indicate a strong excitation of the nuclei on capture of  $\pi^-$  mesons.

On capture of  $\pi^-$  mesons by heavy nuclei it was expected that fission would occur and that it would be possible to arrive at some conclusions as

\* The experiments which form the basis of the report were begun in January, 1950, and extended over 2½ years. The results of the investigation were published in references 1-5. The first reference, in which the documentary photographs are given showing the fission of U and W nuclei, received its final form in March, 1950. The most important results of all the reports were selected for publication in the present paper.

Independently of us and at almost the same time, the fission of the uranium nucleus by slow  $\pi^-$  mesons was observed by Belovitskii and Frank (Report Phys. Inst., Acad. Sci., USSR).

<sup>1</sup> N. A. Perfilov and N. S. Ivanova, Otchet RIAN (Report of Radium Inst.), Acad. Sci., USSR, March, 1950.

<sup>2</sup> N. A. Perfilov and N. S. Ivanova, Otchet RIAN (Report of Radium Inst.), Acad. Sci., USSR, October, 1950.

<sup>3</sup> N. S. Ivanova and N. A. Perfilov, Otchet RIAN (Report of Radium Inst.), Acad. Sci., USSR, June, 1951.

<sup>4</sup> D. V. Viktorov, N. S. Ivanova and N. A. Perfilov, Otchet RIAN (Report of Radium Inst.), Acad. Sci., USSR, January, 1952.

<sup>5</sup> N. S. Ivanova and N. A. Perfilov, Otchet RIAN (Report of Radium Inst.), Acad. Sci., USSR, June, 1952.

to the mechanism of the interaction of mesons with nuclei from the character of the distribution in the range of the fragments, and a comparison with the data on the fission of nuclei by other particles.

In the present paper results are given of experiments on the interaction of slow  $\pi^-$  mesons principally with uranium nuclei. For Pb and W, only photographs are given; these show the possibility of fission of these nuclei on capture of slow  $\pi^-$  mesons.

## 2. OBSERVATION OF FISSION OF U, Pb AND W NUCLEI

Thick emulsion photographic plates with uranium, lead and tungsten introduced into the emulsion in a special holder were irradiated by  $\pi^-$  mesons. Mesons of a definite energy, which had been separated by a magnetic field, were moderated before entering into the emulsion in a wedge-shaped filter to such energies that the majority ended their path in the emulsion.

The photographic plates were impregnated with the substance to be studied by boiling water solutions of salts of the above elements in suitable concentration. For impregnation with uranium we used the salt  $\text{UO}_2\text{Na}(\text{C}_2\text{H}_3\text{O}_2)_3$ . This salt can be used in water solution up to a concentration of 4% without noticeable effect on the sensitivity of the emulsion. For impregnation with lead and tungsten the plates were bathed in water solutions of lead acetate and sodium tungstenate at concentrations of 2 and 12%, respectively. After 30 minutes bathing, there remains in the emulsion about 0.3 and 2 mg/cm<sup>2</sup> of the salts of Pb and W, respectively (determined by weighing the dry plates before and after bathing). The quantity of uranium was determined from a count of the number of disintegrations (number of  $\alpha$ -particles) in a definite time interval (from the moment of impregnating with uranium to the moment of developing the plates). The measured number of  $\alpha$ -

particles per unit volume permitted a calculation of the number of uranium nuclei introduced into this volume, since it is known<sup>6</sup> that 1 mg of the natural mixture of uranium emits 1500  $\alpha$ -particles per minute.

After irradiation with mesons and developing, the plates were examined under the microscope. In the observation the following were noted: a) disintegrations with formation of stars, b) fission events. Fission events were recognized as being those in which traces of multiply charged particles of the type of fission fragments were observed diverging in approximately opposite directions at the end of a meson track.

In the first experiments we observed a very small number of fission events in lead and tungsten. Only three cases were observed in lead (Fig. 1, photomicrograph 2), but they may be considered reliable, since the fact of fission was later established in a neighboring element, namely, bismuth. As for the tungsten nucleus (Fig. 1, microphotograph 3) it appeared subsequently that the fission probability of this element is very small for slow  $\pi^-$  mesons.

For uranium, quite a large number of fission events were found; from their examination it was possible to make a number of deductions and to draw some conclusion as to the mechanism of the interaction of this nucleus with slow  $\pi^-$  mesons.

### 3. DISTRIBUTION IN RANGE OF THE FISSION PRODUCTS OF THE URANIUM NUCLEUS

In all we observed 356 fissions of uranium nuclei from capture of slow  $\pi^-$  mesons. Of these the greater part (87%) were events with emission at the place of absorption of the  $\pi^-$  meson of two heavy fragments in almost opposite directions (Fig. 1, photomicrograph 4). In addition to the fissions in which only two heavy fragments were visible, fissions of uranium nuclei were observed accompanied by the emission of a light charged particle (Fig. 1, photomicrograph 1). Thirty-five such cases were found, or 10% of the total\*.

In what follows we shall designate as double fissions those in which tracks of two heavy particles are observed. If, however, along with the fragments, tracks of light charged particles are observed, we shall provisionally call these events "complex fissions".

The considerable number of fissions found makes it possible to get a certain amount of statistical regularity.

\* In addition to these, one fission event was found with emission of a light charged particle from the end of the fragment.

<sup>6</sup> C. A. Kienberger, Phys. Rev. 76, 1561 (1949).

As is well known, one of the essential characteristics of the fission process is the distribution of individual fragments in range (from the point of fission to the end of the track). For all our cases the tracks of every fragment could be measured individually, since the center of fission can be well determined from the end of the track of the slow  $\pi^-$  meson.

The distribution in range of the individual fragments is presented in Fig. 2. for double fissions, and in Fig. 3 for complex fissions. Both curves are similar within the limits of error.

The most essential fact to be noticed in these curves is the existence of one sharply defined maximum, indicating a considerable percentage of fissions into approximately equal masses. The same phenomenon, as is well known, is observed in the fission of heavy nuclei by high energy particles (> 70 mev) as distinguished from the fission of uranium by slow neutrons, where the fission is principally into fragments of unequal mass. In the distribution in range, this asymmetry of fission is expressed in the appearance of a saddle-shaped curve for the individual fragments.

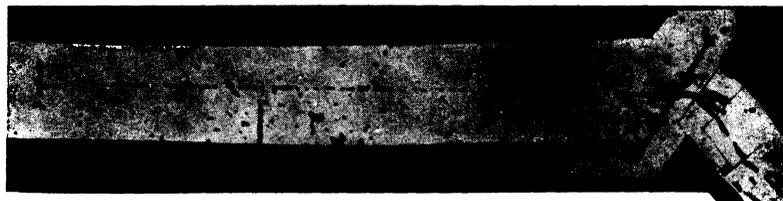
Figures 4 and 5 show the distributions of the total range of the two fragments in each fission event for double and complex fissions of uranium nuclei. Both curves with one broad maximum correspond to the type of the similar curve for the fission of uranium by slow neutrons.

The mean total range of the two fragments for double fission is equal to 27.3  $\mu$ , which agrees sufficiently well with the mean total range (in the same emulsion) for fission by slow neutrons (24  $\mu$ ). This shows, as is to be expected, that the energy released from the rest mass of the  $\pi^-$  meson is not realized in the form of kinetic energy of the fragments.

The mean total range of the heavy fragments from complex fission is somewhat less (21.7  $\mu$ ). However, this difference lies within the limits of error, which is fairly large in view of the small statistical accuracy of the curve (Fig. 5).

### 4. NATURE, ENERGY AND ANGULAR DISTRIBUTION OF THE LIGHT CHARGED PARTICLES EMITTED FROM FISSION OF URANIUM NUCLEI BY SLOW $\pi^-$ MESONS

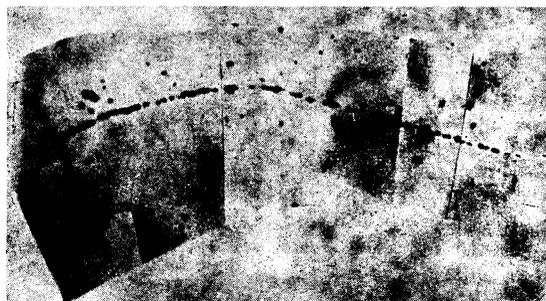
To explain the process of the fission of uranium by slow  $\pi^-$  mesons it is essential to know the properties of the light charged particles accompanying the fission. Under the conditions of our experiment, two methods can be employed for estimating the mass and energy of these particles.



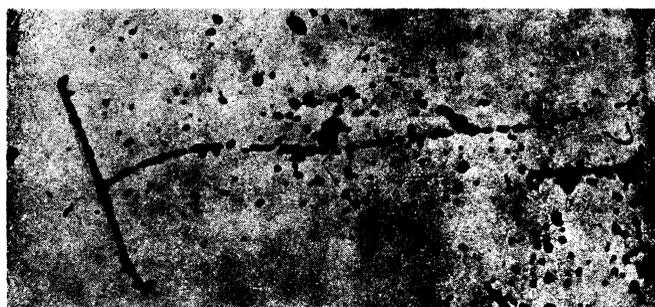
1



2



3



4

FIG. 1. Photomicrograph 1: Fission of the uranium nucleus on capture of a slow  $\pi^-$  meson with emission of a proton; Photomicrograph 2: Fission of the Pb nucleus on capture of a slow  $\pi^-$  meson; Photomicrograph 3: Fission of the W nucleus on capture of a slow  $\pi^-$  meson; Photomicrograph 4: Fission of the uranium nucleus on capture of a slow  $\pi^-$  meson.

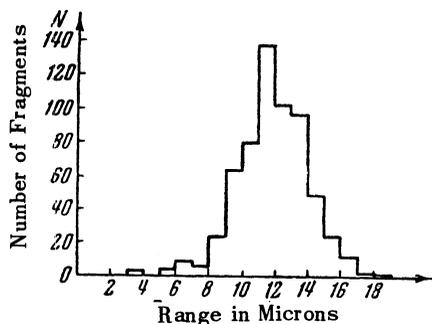


FIG. 2. Distribution in range of the individual fragments from double fission of the uranium nucleus by slow  $\pi^-$  mesons.

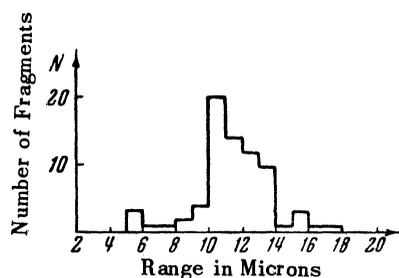


FIG. 3. Distribution in range of the individual fragments from complex fission of the uranium nucleus by slow  $\pi^-$  mesons.

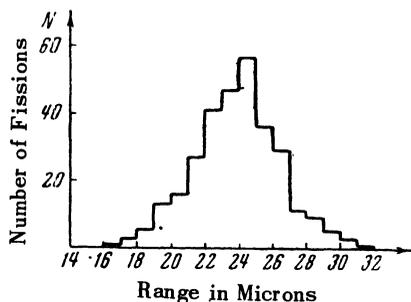


FIG. 4. Distribution of the total range of the two fragments from double fission of the uranium nucleus by slow  $\pi^-$  mesons.

a) For particles finishing their track in the emulsion--by a grain count as a function of the residual range.

b) For particles which leave the emulsion, but have a sufficiently long track--by measuring the mean scattering angle and the grain density at corresponding parts of the track.

It was possible to study only two particles by the first method, since only two tracks out of 35

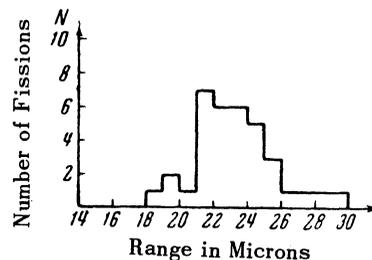


FIG. 5. Distribution of the total range of the two fragments from complex fission of the uranium nucleus by slow  $\pi^-$  mesons.

observed ended in the emulsion, which had a considerable range. The rest left the emulsion. One of the particles had a range of  $3800 \mu$ , the other, of  $480 \mu$ .

The determination of mass from a grain count as a function of the residual range  $R$  is possible, as is well known, if there exists an analogous curve of  $N$  as a function of  $R$  for particles of known mass. Such particles in our case were the  $\pi^-$  mesons, which have a long track and stay in the emulsion. The curve for these was taken as the mean of the grain count of ten  $\pi^-$  mesons. The mass of the particles being studied, determined by this method from the dependence of  $\log N$  on  $\log R$ , was found to be equal to the mass of the proton within the limits of experimental error. The energy of these protons, determined from the whole range in the emulsion, is 30 and 9.3 mev, respectively.

Using the second method, an estimate of mass and energy could be carried out for eight particles (with a range greater than  $350 \mu$ )\*. These particles were also shown to be protons.

The results of the measurements are given in Fig. 6. In this Figure the solid lines give the derivative  $dN/dR$  of the grain density as a function of the mean scattering angle  $\bar{\alpha}$  for protons and  $\pi^-$  mesons. For calibration of the protons, in order to be sure of the absence of regression in determining the grain density, tracks of particles ending in the emulsion were selected from a number of stars formed by  $\pi^-$  mesons. The mass of these particles, equal to the proton mass, was determined by a grain count as a function of the residual range. For orientation, the analogous curve of  $dN/dR$  for high energy  $\alpha$ -particles is

\* Graduate student D. V. Viktorov took part in the mass determination by the method of scattering.

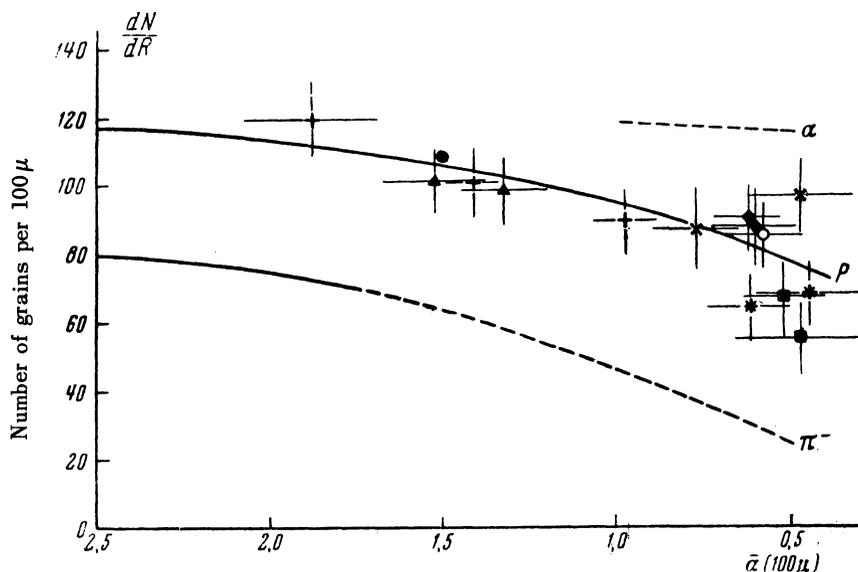


FIG. 6. Dependence of grain density in the track on mean scattering angle for  $p$ ,  $\pi^-$  and  $\alpha$ ; +, case 1;  $\blacktriangle$ , case 2;  $\circ$ , case 3;  $\blacksquare$ , case 4;  $\bullet$ , case 5 (stops in emulsion-proton);  $\times$ , case 6;  $*$ , case 7;  $\blacklozenge$ , case 8.

shown dotted in the same Figure\*\*.

The experimental points for the particles being studied are also given in Fig. 6. Some tracks, in view of their length, can be divided into several parts (2-3). For this reason we have two or three points designated by identical symbols for one and the same track.

From Fig. 6, it is seen that all the experimental points are grouped fairly well around the proton curve, which gives a basis for considering these particles to be protons.

As a check, a particle (whose track stopped in the emulsion) was selected from the group of particles studied by the method of scattering, whose mass and energy had already been determined by the first method. We see that the point for this curve, designated  $\bullet$ , fits the proton curve well. The energy, estimated from the mean scattering angle, and equal to 9.9 mev, agrees within experimental error with the energy determined from the residual range (9.3 mev).

Our data do not make it possible to distinguish protons from deuterons, but the probability of appearance of the latter should not be large in such processes.

\*\* The data for the construction of the curve were taken from reference 7, combining the curves for protons and  $\pi^-$  mesons with our curves.

7 P. H. Fowler, Phil. Mag. 41, 169 (1950).

Twenty-one additional particles could be identified as protons from the low grain density in the residual tracks on the basis of a comparison of the curves of density vs. range for protons and alpha particles. Only 5 particles with a high grain density could be considered alpha particles (photomicrograph, Fig. 7). Consequently, it is possible to say that the light charged particles emitted in the fission of uranium by  $\pi^-$  mesons are principally protons. It is well known that the light charged particles, which in rare cases ( $\sim 1/100$ ) accompany the fission of uranium by slow neutrons, are always alpha particles.

The energy of the protons emitted in fission could be estimated in 17 cases. The histogram of the energy distribution of the particles is given in Fig. 8. The spectrum shows a relatively large number of fast protons with energy  $> 20$  mev.

The distribution in angle of the emitted protons, relative to the line of ejection of the heavy fragments, is shown in Fig. 9, and is nearly isotropic. This isotropic distribution emphasizes even more strongly the difference in character between fission by  $\pi^-$  mesons and fission by slow neutrons. The latter case is characterized by the emission of alpha particles in a direction perpendicular to the line of emission of the fragments.

In conclusion, we would like to note one more special case in which the track of a charged



FIG. 7. Fission of the uranium nucleus on capture of a  $\pi^-$  meson with emission of an  $\alpha$ -particle.

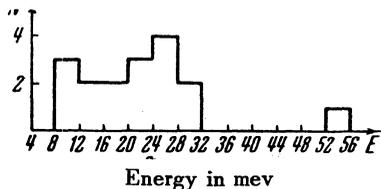


FIG. 8. Distribution in energy of the protons emitted in fission of the uranium nucleus by slow  $\pi^-$  mesons.

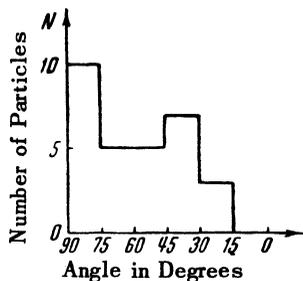


FIG. 9. Angular distribution of the light particles accompanying fission of the uranium nucleus by slow  $\pi^-$  mesons.

particle starts from the end of a heavy fragment (photomicrograph, Fig. 10). The track of this particle ends in the emulsion. The relatively high density and its small change with range, give some reason to believe that this is an alpha particle. We have only one such event. Unless this is a chance superposition, the observed phenomenon may be explained as an alpha decay after a preceding beta decay.

##### 5. FISSION PROBABILITY OF THE URANIUM NUCLEUS FOR SLOW $\pi^-$ MESONS

The value  $P_F$  of the probability of fission of the uranium nucleus on capture of a  $\pi^-$  meson can be found from our experimental data according to the relation:

$$P = P_U P_F \quad (1)$$

where  $P = N_F/N_\pi$  is the observed ratio of the number of fissions,  $N_F$ , to the number of  $\pi^-$  mesons,  $N_\pi$ , stopped in the same volume of emulsion;  $P_U$  is the probability of meson capture by a uranium nucleus in the emulsion and  $P_F$  is the probability of fission of the uranium nucleus after having captured the  $\pi^-$  meson.

The probability  $P_U$  can be evaluated from a relation proposed by Fermi<sup>8</sup>, that the probability of capture of a slow  $\pi^-$  meson in the orbit of a nucleus is proportional to the  $Z$  of the nucleus:

$$P_U = \frac{N_U Z_U}{\sum N_i Z_i + N_U Z_U}, \quad (2)$$

where  $N_U$  is the number of uranium nuclei per unit volume of emulsion;  $Z_U$  is the atomic number of uranium;  $N_i$  and  $Z_i$  are the number of nuclei per unit volume and the atomic number of the elements entering into the composition of the emulsion. The probability  $P_U$  can be calculated if  $N_U$  is known, i.e., if the number of uranium nuclei introduced, and the composition of the emulsion are known. The value of  $N_U$ , the number of uranium nuclei per  $\text{cm}^2$  of an emulsion  $100 \mu$  in thickness, determined (as described above) from the number of alpha decays, was  $(3.6 \pm 0.3) \times 10^{17}$  for our plates. The value of  $N_\pi$ , the mean number of mesons stopped per  $\text{cm}^2$  of a  $100 \mu$  thick emulsion, was found equal to  $3260 \pm 400$  in our experiment. The comparatively large error in the determination of the mean number of mesons stopped per  $\text{cm}^2$  is explained by the variation in current from one end of the plate to the other. The current of mesons being stopped is distributed uniformly along the thickness of the layer of emulsion. The value of  $N_F$ , the mean number of fissions from  $\pi^-$  mesons per  $\text{cm}^2$  of  $100 \mu$  thick emulsion, was equal in our case to  $9.1 \pm 1$ . The composition of the Ilford C-3 emulsion is known.

<sup>8</sup> E. Fermi and E. Teller, Phys. Rev. **72**, 399 (1947).

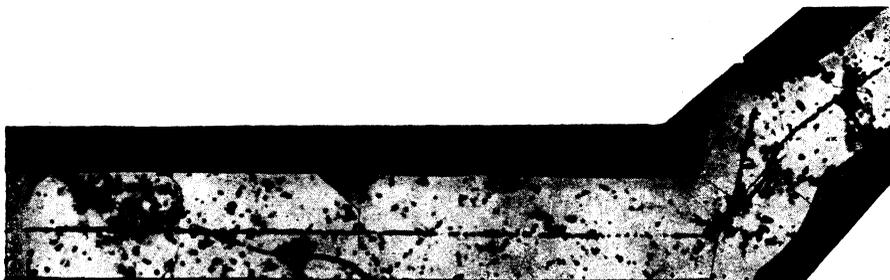


FIG. 10. Fission of the uranium nucleus with emission of an  $\alpha$ -particle from the end of a fission fragment.

Using the above data, we can calculate a value for the fission probability by  $\pi^-$  mesons:

$$P_F = 0.87 \pm 0.27,$$

i.e., we get a probability near unity, from which it follows that almost every uranium nucleus, which captures a  $\pi^-$  meson, undergoes fission.

It should be said that the probability calculations given above were made under the assumption that the uranium nuclei are mixed uniformly with all the elements forming the emulsion. In a recent report<sup>9</sup>, the probability of fission of the uranium nucleus by  $\pi^-$  mesons, evaluated by this same method, has a somewhat different value, equal to  $0.37 \pm 0.13$ . However, this value was obtained on the basis of a relatively small number of events. In addition, the authors of this report felt that the uranium nuclei might not be distributed uniformly throughout the emulsion, that is, that all the uranium might be in the gelatine, without penetrating the crystals of AgBr. In this case the fission probability from the data of reference 9 becomes  $0.18 \pm 0.06$ .

The question of the distribution of the uranium nuclei through the elements forming the emulsion was solved experimentally by Lozhkin and Shamov<sup>10</sup>. They showed that in an emulsion immersed in a water solution of a uranium salt, as used in our experiments, all the uranium nuclei are located in the gelatine, and none are observed in the crystals. We can take account of this non-uniformity of distribution of the uranium nuclei among the elements forming the emulsion in calculating the fission probability. For this it is

necessary in calculating  $P_U$ , the probability of capture of a meson by the uranium nuclei, to consider the latter distributed in the gelatine and for  $N_\pi$  to take the number of mesons stopped in the gelatine only (which constitutes 39% of the total number of stopped mesons<sup>11</sup>).

Then for the fission probability we get the value

$$P_F = 0.42 \pm 0.15,$$

that is, of all the uranium nuclei which interact with slow  $\pi^-$  mesons, only  $\sim 40\%$  experience fission. For the rest it is necessary to assume that they lose their excitation by boiling off particles.

On the other hand, the assumption can be made that the probability of fission of a uranium nucleus which has received a considerable excitation by interaction with a  $\pi^-$  meson is nevertheless near unity. Then it can be said that the formula we took, according to which the capture probability of a  $\pi^-$  meson is proportional to the  $Z$  of the element, does not entirely correspond with reality. Hence, taking  $P_F = 1$ , our experimental data give a formula proportional to  $Z^a$ , where  $a = 2/3$ .

## CONCLUSIONS

1. On capture of slow  $\pi^-$  mesons by nuclei of elements at the upper end of the periodic table, there is a definite probability of fission. For uranium the fission probability is not greatly different from unity.

2. For uranium nuclei, the distribution in range of the individual fission fragments from interaction with slow  $\pi^-$  mesons gives one sharply defined

<sup>9</sup> W. John and W. F. Fry, Phys. Rev. 91, 1234 (1953).

<sup>10</sup> O. V. Lozhkin and V. P. Shamov, Otchet RIAN (Report of Radium Inst.), Acad. Sci., USSR, January, 1954.

<sup>11</sup> S. Tamor, Phys. Rev. 77, 412 (1950).

maximum, which shows a considerable number of cases of fission into fragments of approximately equal mass.

3. Along with the fissions of the uranium nucleus which are binary in nature, giving only two heavy particles, in ten cases out of a hundred the fission is accompanied by the emission of light charged particles. A special investigation of the nature of these light particles has shown them to be protons in an overwhelming majority. Their angular distribution, measured from the line of ejection of the fragments, is isotropic. These data show the marked difference between the fission process on capture of  $\pi^-$  mesons and the fissions caused by slow neutrons, and correspond more nearly with the fission of uranium nuclei by fast particles.

4. The sum total of the facts adduced does not contradict the following conception of the mechanism of the process. A slow  $\pi^-$  meson, captured in one of the Bohr meson orbits of a uranium atom, interacts with a pair of nuclear particles ( $np$ ) or ( $pp$ ), to which it transfers the energy of its rest mass and charge. As a result, two fast particles are produced, with energies of about 70 mev each<sup>12</sup>. Fast particles of this energy, passing through the nucleus, can knock out one or two nucleons by collision and then escape (if they have enough energy), leaving the nucleus in a strongly excited state. The excited uranium nucleus then loses the energy of excitation by boiling off nucleons, undergoing fission at some stage of the excitation.

<sup>12</sup> H. Morinaga and W. F. Fry, *Nuovo Cim.* **10**, 308 (1953).

If the probability of fission of the uranium nucleus on capture of a  $\pi^-$  meson is equal to unity, then, in our view, it follows that at some (possibly very considerable) energy of excitation, the probability of fission of the uranium nucleus and the probability of the nucleus boiling off a neutron are comparable to one another. A value for the fission probability less than unity apparently means that in a number of cases all the energy of excitation may be lost only by boiling off nucleons.

Starting with the above proposed mechanism of the interaction it is possible to explain double fission as fission with emission of neutrons (primary or secondary), and the difference between this and complex fission consists merely in the fact that in the latter, primary or secondary protons are emitted along with the neutrons. The large number of protons with energies  $> 20$  mev that accompany fission indicates in our view that a considerable part of the protons observed in fission must be charged to knock-out and not to boiling.

The nearly isotropic angular distribution of the emitted protons, which was observed, follows naturally from the above.

The authors are grateful to M. G. Meshcheriakov and the staff of co-workers in his laboratory for their cooperation in carrying out the work on the study of the interaction of slow  $\pi^-$  mesons with nuclei.

The authors recall with gratitude the late Prof. P. I. Lukirski, who showed a constant interest in this work.

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(both of our own and of JETP)

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2	434	2	22	27.3 $\mu$	23.7 $\mu$
2	557	Fig. 10			On the right hand side, abscissa values should read 0, 200, 400, 600, 800, 1000.
2	591	2	7	$A = \frac{e^2 H_{00}^2 \delta_{00}^2}{mc^2}$	$A = \frac{e^2 H_{00}^2 \delta_{00}^2}{mc^2}$
2	754	1	3 ff.	_____	<sup>14</sup> B. B. Kinsey and G. A. Bartholomew, Phys. Rev. <b>82</b> , 380 (1951). <sup>15</sup> B. B. Kinsey and G. A. Bartholomew, Phys. Rev. <b>83</b> , 234 (1951).
2	771	1	10	Intermediate State	Intermediate State of Tin
	771	1	19	sphere of lead	sphere of tin
3	145	1	1	$R = 10 ec$	$R = 1/ec$