The Yield of Fission and Star Formation Processes in the Capture of π^--mesons by Nuclei of U, Bi and W[1]

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The interaction of slow π^--mesons with the nuclei of U, Bi and W has been studied by the method of thick photographic emulsions in which the element to be investigated is placed in the middle layer of the emulsion in a form which is insoluble during development and fixing. The ratio between the yield for fission and for star formation in these elements as a result of their capture of π^--mesons is obtained. The probability of nuclear fission as a result of the capture of π^--mesons strongly decreases with decreasing Z: for uranium the probability for fission is ~ 0.3, for bismuth ~ 0.02 and for wolfram, perhaps less than 0.002. The other processes of interaction of π^--mesons with heavy nuclei lead primarily to the formation of "no-prong" and "one-prong" stars. A possible mechanism for the fission of nuclei resulting from the capture of π^--mesons is discussed.

1. INTRODUCTION

IT was shown[2] by Perfilov and Ivanov in 1952 that the nuclei of U, Pb and W can undergo fission as the result of π^--capture. The yield of the fission process in π^--meson capture was estimated very approximately, because of the small number of observed events.

In this work[2], the following method of investigation was used. Samples of U, Pb and W were introduced in a thick photographic emulsion, sensitive to mesons, by soaking the plates in water solutions of the salts of these elements. The plates were irradiated by π^--mesons, were developed and then, during scanning, were searched for events where tracks of mesons ended, and tracks of two strongly ionizing particles, emitted in opposite directions, appeared.

However, when using such a method of investigation, it is not possible to draw any conclusion about the ratio of the yields of the two processes of fission and star formation resulting from the capture of mesons by nuclei. This is due to the fact that star formation in photographic emulsions can take place in a number of elements which make up the emulsion (Ag, Br, C,N,O,S). The problem of the determination of this ratio, which holds great interest for the understanding of the interaction mechanism between π^--mesons with a nucleus, can be solved by observing the process of meson capture in particular nuclei.

In this present work an attempt is made to come closer to the solution of this problem by the method of introducing into the photographic emulsion finely powdered materials which do not dissolve during its processing.

2. METHOD OF INVESTIGATION

Three-layered photographic plates, the method of preparation of which was developed earlier in this laboratory, were used in this investigation.

The elements U, Bi and W were introduced in the form of finely ground oxides U_3O_8, Bi_2O_3 and WO_3 in the middle layer, whose thickness after development and fixing was less than the depth of focus of the 90x objective of the microscope. Therefore, the layer can be scanned solely through the horizontal displacement of the plate. This shortens appreciably the scanning time in comparison with the case when the loaded material is dispersed throughout the whole volume of the photographic emulsion. Moreover, the location of the element being investigated in the middle of the photographic emulsion always ensures an appreciable path length in the emulsion for the nuclear fission products. The necessary dimensions of the grains of the material introduced are obtained by means of successive elutriation of oxide powders shaken in water. In our investigation, the dimensions of the crystals were about 3μ. By means of special experiments it was shown that during the development and fixing of the photographic emulsion, the dimensions of the crystals do not change, i.e., the chosen materials do not dissolve observably under the conditions of the experiment.

The material loaded in the middle layer of the emulsion, which does not dissolve during the processing of the photographic plate, does to some extent impede the diffusion of the developer to the lower layer. Because of this effect, a three-layered

[1] N. A. Perfilov, O. V. Lozhkin and V. P. Shamov, Report RIAN (1952)

[2] N. A. Perfilov and N. S. Ivanova, Report RIAN (1950)

plate requires some modification of the development procedure, designed to develop the lower layer sufficiently without overdeveloping the upper layer. Satisfactory results are given by the method of temperature development.

When a metal oxide is introduced in a photographic emulsion, there are two elements with which interactions may take place. However, on the basis of existing experimental data[3] and theoretical calculations[4], one expects that meson capture will occur chiefly for nuclei of high Z, if the elements are in a bound state. In our experiments oxygen was bound as an oxide of a heavy element having a Z 9 to 11 times greater than that of oxygen. Therefore, one could expect that the effect due to oxygen would be negligibly small. Our experimental data are in agreement with this conclusion.

Three-layered plates irradiated by negative π mesons slowed down by aluminum layers of suitable thickness were developed and then scanned through a microscope. Only the middle layer was examined for events in which a meson stopped in the region occupied by a crystal of the loading substance.

3. SCANNING RESULTS

In the Table below all the cases in which a π^-- meson stopped in the region occupied by a crystal are divided into "stars" and fission

TABLE I

Loaded material	No. of stars observed	No. of fission events
U_3O_8	210	10
Bi_2O_3	250	2
WO_3	600	1
Unloaded emulsion (AgBr + gelatin)	10 000	Not found

events. Fission events are identified by the appearance of two short, thick tracks, issuing in opposite directions from the places where the π^--meson stopped. "Star" events refer not only to those cases when a meson is captured and several ionizing charged particles, leaving tracks in the emulsion, are emitted, but also to those events when the capture of a meson does not result in the emission of charged particles (the so-called "no-prong stars"). The emulsion used (prepared by ourselves) was capable of registering π^-- mesons with energies up to 5 mev.

Table 1 shows the results of the scanning of plates (with the middle layer loaded with U_3O_8, Bi_2O_3 and WO_3) irradiated with slow negative π^-- mesons.

Table 2 shows the prong distribution of the stars as a function of the loaded material, and also the prong distribution of stars caused by π^-- mesons in an unloaded emulsion.

TABLE II

No. of prongs in star	No. of stars (%) for given material			
	Crystal WO_3	Crystal Bi_2O_3	Crystal U_3O_8	Unloaded emulsion
0	47	57	34	32
1	24	20	22	26
2	12	10	19	20
3	9	9	18	14
4	8	4	6	7
5	—	—	1	1

4. ANALYSIS OF THE OBTAINED DATA

The elements investigated were introduced into the photographic emulsion in the form of an oxide. Therefore, when a meson stopped in the crystal of the introduced substance, the interaction could take place either with the heavy nucleus or with the oxygen. Since the interaction of a π^-- meson with oxygen cannot result in ionizing particles similar to the fragments due to the fission of heavy nuclei, the observation of events in which two fragments issue from the crystal can be assigned with complete assurance to the fission of the nucleus being investigated as a result of its capture of a π^--meson. A decision can be made on the number of interactions between the π^--mesons and the nuclei being investigated which result in the emission of only light particles (star formation) if one can separate the following effects in the stars observed to originate in crystals:

1) The capture of mesons by oxygen resulting in stars in the oxides U_3O_8, Bi_2O_3 and WO_3, from the capture by the nuclei U, Bi and W.

2) The capture of mesons resulting in stars in the region of the emulsion near the crystals, from stars originating in the crystals.

Let us clarify the importance of the second effect in determining the true number of cases of the stopping of a meson in the crystal of the loaded substance. Let us assume that we have the case of a star formed in the photographic emulsion in a

[3] W. K. H. Panofsky, Phys. Rev. **78**, 825 (1950)

[4] E. Fermi and E. Teller, Phys. Rev. **72**, 399 (1947)

region a few grains above the loaded substance or else below it, also within a short distance. During the fixing and drying of the photographic emulsion the emulsion will shrink as a result of the removal of the silver bromide, and therefore the thickness will decrease by a factor k (k is the deformation factor. For the emulsions we used $k = 2.5$). The center of the star therefore comes nearer to the crystal of the loaded substance, and sufficiently so that during the study of such a star it will be counted as originating in the crystal.

Since the deformation of the emulsion is only in one direction, during the scanning all stars (approximately) within an ellipsoid of revolution will be counted as originating in the crystals of the loaded substance.

If one assumes that if there is no deformation, stars formed in the emulsion and in the crystal could be distinguished if the distance is the size of a developed grain of silver, then the axis of the ellipsoid would have the values:

$$a = R + d, \quad b = R + kd.$$

Here a is the semi-axis parallel to the surface of the plate, R is the crystal radius, d the diameter of the developed grain in the emulsion. Obviously, the smaller the crystals, the greater will be the number of stars which will appear to be originating in the crystals (when in reality they are due to mesons stopping in the photographic layer in the vicinity of the crystal).

Let us call the neighboring layer bounded in this way the "undistinguishable zone". We will then find the true number of stars originating in the crystals by multiplying the total number of stars observed at crystal locations by the probability, W_c, that a meson will stop in the crystal, or, since $W_c + W_{uz} = 1$ (where W_{uz} is the probability that it will stop in the neighboring layer), we will have the following expression for the true number of events of mesons stopping in the crystals:

$$N_{\text{true}} = N_{\text{obs}} \frac{W_c}{W_c + W_{\text{u.z.}}}$$

$$= N_{\text{obs}} \frac{S_c V_c}{S_c V_c + S_{\text{em}} V_{\text{em}}}.$$

Here V_{em} and V_c are the volumes of the undistinguishable zone and of the crystal, S_{em} and S_c are the stopping power for mesons in the photographic emulsion and in the crystal.

If the total number of stars in the whole layer containing the loaded substance is counted, then the true number of stars in crystals is obtained by substituting in the denominator of the expression the full volume of the scanned emulsion.

One can estimate the true number of stars in crystals of the loaded substance in yet another way. Using the method of the potential barrier[5], one can distinguish between stars resulting from the interaction with light and with heavy nuclei. Then, from the obtained total number of stars, under given assumptions one can deduce the number of stars due to the elements being investigated.

The calculation of the undistinguishable zone and the estimate of the effect on the element being investigated are discussed in the Appendix to this paper. The results of the analysis of the experimental data shown in Table 1 are given in Table 3.

In this way, even from these first results one could estimate approximately the probability of fission of the nuclei of U, Bi and W after absorption of negative π mesons. For uranium this probability lies in the interval from 1.0 to 0.30, for bismuth it is ~ 0.02 and for wolfram ~ 0.007.

Later, in order to improve the data on the fission probability (as the result of π^--meson capture) of the nuclei of U and W, supplementary experiments were performed in which A. V. Pyrkin took an appreciable part. It was shown as a result that the probability for fission of the uranium nucleus for π^- meson capture was 0.3 ± 0.1[6], and that the fission probability for wolfram nuclei is approximately 0.002[7].

In addition to clearing up the question of the probability of the fission of the nuclei of U, Bi and W after π^--meson capture, the character of the stars originating as a result of the capture of mesons by heavy nuclei was also determined. The analysis of the data in Table 2 has shown that for elements with $Z \geqslant 74$, most of the π^--meson captures by nuclei result in the formation of no-prong and one-prong stars. The results of the analysis are shown in Table 4.

5. DISCUSSION OF THE RESULTS

From the experimental data on the prong distribution stars in wolfram and bismuth, one can obtain analogous information for elements with a high Z.

The direct determination of the prong distribution of stars formed through π^--meson capture by uranium nuclei was made difficult in our experiment

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[5] M. G. K. Menon et al, Phil Mag. **41**, 583 (1950)

[6] O. V. Lozhkin and V. P. Shamov, Report RIAN, (January, 1955)

[7] N. A. Perfilov and A. V. Pyrkin, Report RIAN (1953)

TABLE III

Substance	Average size of crystal (μ)	True number of stars in crystals N_{true}	Number of stars in heavy element	Number of stars for each fission event
U_3O_8 . .	1.6	0 ÷ 30	0 ÷ 24	0 ÷ 2.4
Bi_2O_3 . .	5	130	114	57
WO_3 . .	3	176	133	133

TABLE IV

Number of charged particles in the star	No. of stars (%) as a function of the number of prongs		
	Bi	W	AgBr
0	86.2	85.5	42
1	13.8	14.5	29

because of the fact that, due to the small dimensions of the U_3O_8 crystals (approximately 1.6 μ), the number of events which had to be assigned to the emulsion in the undistinguishable zone accounted for 85 - 90% of the total number of stars observed on crystals. In addition, U_3O_8 crystals often cluster, which renders more difficult the measurement of the undistinguishable zone.

Figure 1 shows the dependence of the number of no-prong and one-prong stars on the Z of the nucleus capturing the π^--meson. If the curves are extrapolated to $Z = 92$, it is possible to estimate the average number of charged particles originating in the capture of one π^--meson by a uranium nucleus. We get an approximate value of 0.12. Thus 12% of the interactions of π^--mesons with uranium nuclei lead to the emission of one charged particle. In the other cases the uranium nuclei emit neutrons only, and together with this can undergo fission.

The number of stars corresponding to one fission of a uranium nucleus having captured a π^--meson are given in Table 3. In the Table the limits are indicated to lie between 0 and 24; these were obtained from the following considerations. The stars observed on the U_3O_8 crystals, separated by the method of the potential barrier, show that of the 122 stars which were analyzed,46 must be assigned to light nuclei. Knowing the relative number of stars formed by π^--meson capture by the AgBr, and the C, N and O nuclei in the emulsion, it is possible to determine the number of stars originating in uranium nuclei (neglecting a dif-

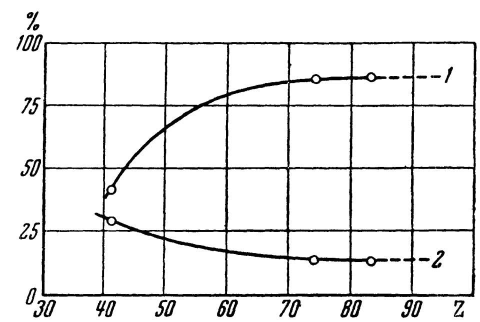

FIG. 1. Dependence of the relative number: 1 - of no-prong and 2 - of one-prong, stars on the charge of the nucleus which has a slow π^--meson.

ference in star formation by oxygen in an oxide) from the expression:

$$\chi = N_H - qN_L,$$

where N_H is the number of stars formed by π^--mesons in heavy nuclei (U,Ag,Br), N_L is the number of stars in light nuceli (C,N,O), and q is the relative number of stars in AgBr and stars in C,N and O in the emulsion.

Taking the ratio $q = 2$ from the work of Menon et al[5], one can come to the conclusion that,when uranium nuclei capture π^--mesons, stars are not produced*, i.e., only fission occurs (the lower

* If one uses the more reasonable value $q = 1.4$ in this calculation (this value was obtained by us after the completion of this work) the lower value of the number of stars corresponding to one fission will be 1.7.

limit to the number of stars corresponding to one fission is given in Table 3).

The upper limit to the number of stars for each fission of the uranium nucleus, equal to 2.4, was obtained from the calculation described in the Appendix. Later work by us[6] on the determination of the probability of fission of the uranium nuclei from π^--meson capture led to the value 0.3 ± 0.1 for the probability of fission. This corresponds to 2.3 stars for each uranium fission.

In such a way the following conclusions can be made on the basis of the results of the experiments described:

1. Fission of nuclei may occur when π^--mesons are captured by nuclei with $Z < 74$. The probability of fission decreases from 0.3 for U to perhaps less than 0.002 for W.

2. The other cases of interaction of π^--mesons with heavy nuclei lead mostly to the formation of one-prong and no-prong stars.

The data presented permit certain conclusions on the mechanism of the fission process. As the result of the interaction of a slow π^--meson with the nucleons of a heavy nucleus, this nucleus is excited to an energy of ~ 100 mev[5,8].

There are at least three ways in which an excited nucleus can go to a state of minimum energy: (a) by particle emission from the nucleus, (b) by emitting γ-quanta, and (c) by fission.

The similarity in the average number of charged particles emitted in the capture of one π^--meson by a uranium nucleus (0.12) and in the number of complex fissions of a uranium nucleus as a result of the capture of a π^--meson (0.1) permits one to think that for such an excitation energy the process of particle emission (chiefly neutrons) from the nucleus will take place with the greatest probability**.

If the excitation energy is decreased, the relative probability of two other processes increases in the neutron emission process. At the same time the absolute magnitude of the probability of fission increases because of the increase in Z^2/A. For uranium capturing a π^--meson, after emission of 8 or 9 neutrons, the probability of fission appears to be predominant.

For nuclei with smaller Z, when the excitation energy is decreased, by the process of particle emission, to the value of activation energy (which determines the threshold for fission for a given Z^2/A). The sum of the probability of particle emission and of γ-quanta emission will always be greater than the probability of fission. This difference will increase with decreasing Z. As a result, for nuclei in the neighborhood of Bi, the first two processes will predominate: particle emission for high excitation energies and γ-quanta emission for low excitation energies.

The fission of uranium with the emission of light, charged particles as the result of slow π^--meson capture can be explained in a natural way as fission arising after the emission of one charged particle and several neutrons. The fission of bismuth as the result of π^--meson capture is not in disagreement with the emission hypothesis: After the emission of 7 or 8 neutrons, the threshold for fission decreases to a value ~ 7 mev, beyond which the nucleus has an ascertainable fission probability.

The calculation for wolfram shows that for a decrease in the fission threshold to a value of the order of the binding energy of a neutron in the nucleus, the preliminary emission of 15 to 16 neutrons is necessary, which requires an excitation energy ~ 280 mev. This value is two times greater than the maximum possible excitation energy of the nucleus as the result of π^--meson capture. If one assumes that the fission of wolfram as the result of the capture of slow π^--mesons is established, it follows, from the statements above, that wolfram in this case is in an excited state which lies appreciably higher than the state corresponding to fission according to the emission hypothesis.

The basic results of this work were obtained in December, 1951. At this time, the authors of two other works[10,11] have estimated the probability for fission of the uranium nucleus as the result of slow π^--meson capture. In the work by S. G. Al-Salam[10] the probability for fission is given as ~ 1 and in the work by John and Fry[11] it is given as 0.18 ± 0.06.

In conclusion the authors express their appreciation to M. G. Meshcheriakov and his collaborators for their assistance in this work.

The authors recall with profound gratitude the now deceased Acad. P. I. Lukirskii who showed a constant interest in this work and who participated in the discussion of the results.

** From the data of Perfilov and Ivanova[9], 10% of the fissions of uranium nuclei (as the result of π^--meson capture) are accompanied by the emission of light charged particles (usually protons).

[8] D. H. Perkins, Phil. Mag. **40**, 601 (1949)

[9] N. A. Perfilov and N. S. Ivanova, Report RIAN (1951)

[10] S. G. Al-Salam, Phys. Rev. **84**, 254 (1951)

[11] W. John and W. Fry, Phys. Rev. **91**, 1234 (1953)

APPENDIX

1. THE SEPARATION OF THE PROCESS OF STAR FORMATION IN THE EMULSION LAYER ADJACENT TO THE CRYSTALS LOADED IN THE EMULSION

Estimate of the undistinguishable zone. When the method of loading the emulsion with finely ground substances is used, because of the appreciable deformation of the emulsion after processing, and because of the appreciable dimensions of the Ag grains, there exists (within the emulsion) a region around the loaded crystals which influences the results of the observations. The calculation of this region (the so-called undistinguishable zone) carried out for WO_3 crystals, of average dimensions 3μ, assumed that they have a spherical shape. The average size of the developed Ag grains was $\sim 0.5\mu$ in the emulsion used in this work, and the deformation coefficient of the emulsion $k = 2.5$.

In this work, using a microscope with a 90 x objective, it was decided that the event takes place in the crystal if the center of the star is located inside the crystal or on its surface. In this way, if the event takes place within the emulsion volume bounded by the surface of the ellipsoid of revolution (approximately), then, after processing, the center of the star falls into in a spherical layer of 0.5 μ thickness, and it will be recorded by us as an event occurring within the WO_3 crystal. This is because of the juxtaposition of the Ag grain (which was the center of the star) and the surface of the crystal. Besides the deformation of the emulsion in the calculation of the undistinguishable zone, it is necessary to consider the region under the crystal, which is not investigated because of the opaqueness of the crystal. Figure 2 shows schematically the dimensions of the undistinguishable zone.

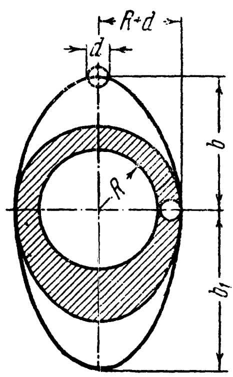

FIG. 2. Schematic representation of the undistinguishable zone. R is the radius of the crystal loaded in the emulsion; d is the diameter of the developed Ag grains. The shaded part shows the dimensions of the undistinguishable zone in the developed emulsion.

If one takes: R – the radius of the crystal (1.5 μ), d – the average size of developed Ag grains (0.5 μ), b, b_1 – the upper and lower semi-axis of the ellipsoid of revolution, then the volume of the undistinguishable zone will be

$$V_{u.z.} = {}^2/_3\,\pi(R+d)^2\,(b+b_1) - {}^4/_3\,\pi R^3 = 42.4\,\mu^3,$$

which is a value three times greater than the volume of the crystal having a radius of 1.5 μ.

2. SEPARATION OF THE EFFECT ON WOLFRAM FROM THE TOTAL NUMBER OF STARS RELATED TO WO_3 CRYSTALS

One can assume the ratio of the probability of capture of the meson in the undistinguishable zone of the emulsion and in the crystal is equal to

$$\frac{W_{em}}{W_{WO_3}} = \frac{V_{em}}{V_{WO_3}}\,\frac{S_{em}}{S_{WO_3}},$$

where V_{em}, V_{WO_3} are the volumes of the emulsion in the undistinguishable zone and of the crystal, S_{em}, S_{WO_3} are the stopping powers of the meson in the emulsion and in the WO_3 oxide. The stopping power for the meson can be written analogously to the stopping cross section for heavy charged particles:

$$S = \frac{4\pi NZe^4}{mv^2}\ln\frac{2mv^2}{\bar{I}}.$$

From this,

$$\frac{W_{em}}{W_{WO_3}} = \frac{V_{em}}{V_{WO_3}}\,\frac{N_{em}\bar{Z}_{em}}{N_{WO_3}\bar{Z}_{WO_3}}\left(\ln\frac{2mv^2}{\bar{I}_{em}}\Big/\ln\frac{2mv^2}{\bar{I}_{WO_3}}\right).$$

The velocity of the π^--meson for the calculation is chosen to be such that, encountering a crystal having a dimension of $\sim 3\mu$, it will stop in it; i.e., a meson having such an energy will have a path in the WO_3 oxide not greater than the diameter of the crystal. This condition will be observed for meson energies less than 100 kev, i.e., for velocities less than 1.13×10^9 cm/sec.

Substituting in the formula $\bar{Z}_{em} = 13.5$, $\bar{Z}_{WO_3} = 24.5$, $N_{em} = 8.2 \times 10^{22} cm^3$, $N_{WO_3} = 7.5 \times 10^{22} cm^3$, we obtain***

$$W_{em}/W_{WO_3} = 2.49.$$

*** The average ionization potential of atoms with stopping power $\bar{I}$ is taken, from Bloch's formula, to be:

$$\bar{I} = 11.5 \cdot \bar{Z}\,ev$$

Since the number of stars originating in the WO_3 oxide is determined from the formula

$$N_{\text{true}} = N_{\text{obs}} \cdot \frac{W_{WO_3}}{W_{WO_3} + W_{\text{em}}},$$

of the 600 stars observed on crystals, only 172 could be counted as originating at the WO_3 oxide, the others being due to the emulsion in the undistinguishable zone. From this it is possible to estimate the number of interaction events with wolfram nuclei, assuming that the probability of capture of the π^--meson in the oxide is proportional to the Z of the nucleus.

In this way we find that approximately 130 stars of the 172 originating at the WO_3 are due to interactions with wolfram nuclei.

An approximate expression for the number of π^--meson capture events by wolfram nuclei is obtained, by using the potential barrier method for separating the stars into stars originating in the heavy nuclei, N_H, and stars originating in the light nuclei, N_L. Then, from the expression

$$N_W = \frac{N_H - qN_L}{1 - qp},$$

one can determine the number of stars originating in the nuclei W. Here p is the probability of π^--meson capture by oxygen nuclei in the oxide, q is the ratio of the number of stars originating in AgBr and the number of stars originating in the C, N, and O of the emulsion.

Analogous calculations were carried out for bismuth and uranium.

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