

## Fission of Heavy Nuclei by High Energy Neutrons\*

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Results of experiments on the fission of various heavy nuclei in the region  $Z = 74-92$  by neutrons of nominal energy of 120 and 380 mev are presented. An evaluation of threshold for fission connected with previous neutron emission is based on a comparison of the binding and critical fission energies. The average number of neutrons emitted during the fission of a heavy nucleus is evaluated.

### 1. INTRODUCTION

**I**N the course of the experimental work of the past few years various peculiar characteristics of the fission of heavy nuclei by high energy nucleons have been discovered and investigated. These differed in many respects from the previous work on fission by slow neutrons.

The first stage of the interaction of a high energy nucleon with a complex nucleus is an internal cascade, including one or several nucleon collisions within the nucleus. Struck nucleons may either fly out of the nucleus without further collisions or may stick within the nucleus, expending their kinetic energy in further collisions. At the end of the internal cascade, the nucleus remains hot, i.e., in an excited state, and the cooling of such a nucleus may take the form of multiple emission of particles. In the case of heavy nuclei, the fission process may offer strong competition to the multiple emission of particles. One can imagine two extreme cases of fission of a highly excited nucleus: the fission takes place with the loss of an excitation energy that is far larger than the critical fission energy, the fissioning nucleus having been very highly heated (high temperature fission); or the fission is preceded by multiple emission of particles (especially neutrons), after which the remaining excitation energy of the nucleus barely exceeds the critical fission energy ("emissive fission").

Such a division into high temperature and emissive fission is of course arbitrary, since all intermediate cases can occur. However, this division is useful for understanding characteristics of nuclear fission caused by high energy nucleon bombardment.

During slow neutron induced fission, a characteristic double-humped fission fragment spectrum (in both mass and energy) is observed.

With increase in bombardment energy, the valley between the humps gradually smoothes out, and at high energies, for example in the fission of uranium by 380 mev  $\alpha$ -particles<sup>1</sup>, or in the fission of bismuth by 190 mev deuterons<sup>2</sup>, a strong single-humped fragment mass spectrum is observed. Chemical analysis of the fission fragments leads to the conclusion that the maximum of the mass spectrum is some 3-6 mass units lighter than half the sum of the nuclear and incident particle masses.

Such reduction of the fission fragment mass is connected with the emission of a large number of nucleons (chiefly neutrons) during fission of a heavy nucleus induced by a high energy particle. This emission may precede "emissive" fission, or may follow high temperature fission from an excited state; in this case neutrons are emitted by the fission fragments. The shape of the fission fragment mass spectrum for slow or fast nucleon induced fission has not been explained. Energetically, fission into fragments of equal mass is generally most efficient, i.e., a single-humped fission fragment mass spectrum. The occurrence of a two humped mass spectrum in low energy induced fission has been connected by several authors<sup>3</sup> with the fact that such a spectrum allows the formation of many fission fragments of magic neutron number ( $N = 50$  and  $82$ ). From this viewpoint a two-humped mass spectrum is indicative of fission of a heavy nucleus from a state of weak excitation. The radical change in character of the spectrum of the mass fragments with increase of the bombardment energy may then be considered as an argument in support of high temperature fission from a highly excited state. However, this conclusion is opposed by the appearance among high energy induced fission fragments of large numbers of

<sup>1</sup> P. O'Connor and G. Seaborg, Phys. Rev. **74**, 1189 (1948).

<sup>2</sup> R. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).

<sup>3</sup> D. Brunton, Phys. Rev. **76**, 1798 (1949); L. Meitner, Nature **165**, 561 (1950).

\*The experimental part of this work was completed in 1950-1951.

heavy nuclei of  $\beta^+$  or  $K$  active isotopes (especially for  $A \geq 125$  – in uranium fission). Appearance of such neutron-deficient fragments allows the conclusion that most of the neutron emission takes place before fission, that is, the process is “emissive” fission.

Experiments determining kinetic energy of the fragments in the fission of uranium and thorium by 90 mev neutrons<sup>4</sup>, as well as in the fission of uranium by stopping  $\pi$  - mesons<sup>5</sup> show that the total energy of the two fragments is not higher than is characteristic for slow neutron fission.

This fact also supports, although it does not prove, interpretation of the process as emissive fission, since neutron emission by highly excited fission fragments must be over in  $\tau \sim 10^{-15} - 10^{-16}$  second, and the remaining observable energy will

be determined solely by the coulomb repulsion.

Further evidence toward the emissive fission viewpoint is offered by the bombardment energy dependence of the fission cross sections for nuclei of  $Z = 74-83$ . As we know, the fission cross section of such nuclei as  $U^{238}$  and  $Th^{232}$  increases rapidly for excitation energies several mev higher than threshold. It reaches some tenths of a barn, and is fairly insensitive to further increase of energy. The partial level widths of these nuclei for fission and for neutron emission are of the same order of magnitude. Below are shown upper limits to the fission cross section of nuclei from W to Bi by 14 mev neutrons<sup>6</sup>, and the critical fission energy for the corresponding compound nuclei calculated from data<sup>7</sup>:

	Bi <sup>209</sup>	Pb <sup>208</sup>	Pb <sup>206</sup>	Pb <sup>207</sup>	Pb <sup>205</sup>	Hg <sup>200</sup>	Au <sup>197</sup>	Ir <sup>193</sup>	W <sup>184</sup>
$E_{crit} (mev)$	9.5	9.3	10.6	11.1	11.4	12.1	13.0	15.0	19.5
$\sigma_f \times 10^{29} (cm^2)$	1.4	130	8.2	8.6	3.7	0.67	1.2	1.5	1.2

In all the above cases, the excitation energy of the compound nucleus is greater than the fission threshold; for  $W^{184}$  it is greater by only 0.3 mev, while for  $Bi^{209}$  it is almost 9 mev greater. Nevertheless, the upper limits for the fission cross section of the above nuclei (with the exception of  $Pb^{204}$ ) are about  $10^{-29} cm^2$ , which points to the smallness of the fission widths compared to the neutron widths of these nuclei.

It follows from the statistical model that increase of excitation energy in a region considerably higher than the critical fission energy is accompanied by roughly equal increases of the neutron and fission widths (equal to within an exponential coefficient). It is therefore hard to imagine that the fission cross sections of the above nuclei could increase appreciably with higher energy in the case of high temperature fission. However, the fission cross section of bismuth with 84 mev neutrons<sup>8</sup> is at least 1300 times larger than at 14 mev. On the other hand, the ratio of fission cross section of  $Pb^{204}$  and  $Pb^{208}$  decreases with increasing bombardment energy, from 35 (neutrons, 14 mev) to 3.5

(deuterons<sup>2</sup>, 100 mev). It is thus natural to assume that the fission cross section at high excitations is determined, not by the fission width of the initial nucleus, but by that of a nucleus which has already emitted several neutrons.

Due to the above arguments, we based the analysis of our experimental data (completed in 1950-1951) on the emissive nature of fission. Seeking to determine the average number of neutrons emitted during a fission process, we proceeded from the assumption<sup>9</sup> that the neutron emission takes place until the critical fission energy becomes lower than the neutron binding energy. The calculation of the average number of neutrons on this assumption proved to be in satisfactory agreement with independent experimental determinations.

All the above facts served as a more or less indirect confirmation of the predominance of emissive fission over high temperature fission. A recent paper by Dauthett and Templeton<sup>10</sup> described an attempt to prove experimentally the emission of neutrons before fission.

<sup>4</sup> J. Jungerman and S. Wright, Phys. Rev. **76**, 1112 (1949).

<sup>5</sup> S. Al-Salam, Phys. Rev. **84**, 254 (1954); N. A. Perfilov and H. S. Ivanova, J. Exper. Theoret. Phys. USSR **29**, 551 (1955); Soviet Phys. JETP.

<sup>8</sup> E. Kelly and C. Wiegand, Phys. Rev. **73**, 1135 (1948).

<sup>6</sup> A. Phillips, L. Rosen and R. Taschek, Phys. Rev. **75**, 919 (1949).

<sup>7</sup> D. Hill and J. Wheeler, Progress of the Physical Sciences **52**, 83, 239 (1954).

<sup>9</sup> V. I. Gol'danskii, Report of Inst. Chem. Phys., Acad. Sci. USSR, 1950.

<sup>10</sup> E. Dauthett and D. Templeton, Phys. Rev. **94**, 128 (1954).

These workers compared the range of particular fission fragments (for example  $\text{Sr}^{89}$ ,  $\text{Ag}^{111}$ ,  $\text{Ba}^{140}$ ) in the fission of  $\text{U}^{238}$  by 18 mev deuterons and by 335 mev protons. It turned out that for fission by high energy protons the fragment energies were several mev lower. Such energy reduction is connected with mass reduction of the fragment pair by previous neutron emission. In these cases, the number of neutrons emitted before the  $\text{U}^{238}$  fission was 6-8 ( $\text{Ba}^{140}$  fragment) to 21-22 ( $\text{Sr}^{89}$  fragment). The quantitative results of reference 10 are evidently not sufficiently precise, but the qualitative demonstration of the emission nature of fission seems to be valid.

## 2. DESCRIPTION OF THE EXPERIMENTS

In our experiments, performed in 1950-1951 with the synchrocyclotron at the Institute of Nuclear Problems of the Academy of Sciences, USSR, we investigated the fission of  $\text{U}^{235}$  and  $\text{U}^{238}$  by neutrons of 120 and 380 mev, excitation functions for neutron fission of bismuth in the energy interval 120-380 mev, and the fission of Th, Pb, Tl, Au, Pt, and W by 380 mev neutrons.

The neutrons used in the 1950 experiments were obtained by stripping 280 mev deuterons in a 16 mm ( $14.2 \text{ g/cm}^2$ ) copper target. The energy spectrum of such neutrons is a bell shaped curve with a maximum and average energy of  $\sim 120$  mev and a half width  $\sim 40$  mev.

The neutrons used in the 1951 experiments were obtained by charge exchange of protons in a 25 mm ( $4.6 \text{ g/cm}^2$ ) beryllium target. The target was placed at various distances from the center of the magnet - 153, 195, and 220 cm, which corresponded to proton energies of 260, 400, and 490 mev. The charge exchange neutron energy spectrum is considerably more smeared out than that for stripping reaction neutrons. The energy spectrum of charge exchange neutrons at a proton energy of 490 mev is shown in reference 11. We are calling these (from the position of the spectrum maximum) 380 mev neutrons. Actually the average effective energy for such neutrons depends on the threshold and excitation function shape of the detector used. In the case of the bismuth fission the effective energy was close to 340 mev. The maxima of the charge exchange neutron spectra for proton energies of 260 and 400 mev, obtained by interpolation of various charge exchange spectra, were close to 210 and 315 mev respectively.

The fission process was observed by counting pulses due to fission fragments in ionization chambers. Figure 1 shows the ionization chamber used. Electrodes were aluminum rings to which  $20\mu$  aluminum foil was glued. A layer of the fissioning material of area 8.5 or  $12 \text{ cm}^2$  was deposited on both sides of the working electrodes. Collecting electrodes were less than 10 mm from the working electrodes, which is less than the range of fragments at atmospheric pressure. The chambers were filled with chemically pure argon at a pressure of about 800 mm Hg.

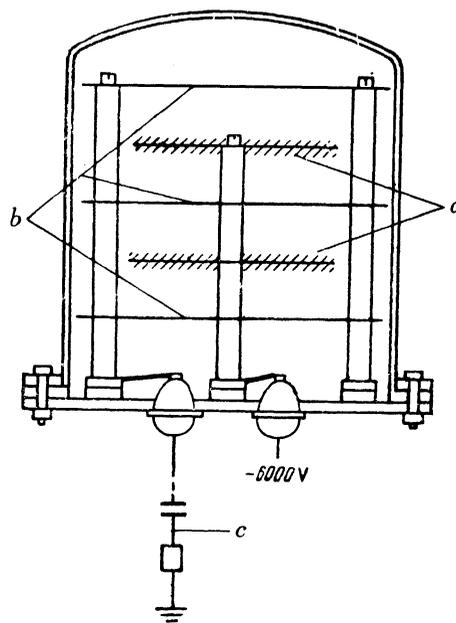


FIG. 1. Drawing of the ionization fission fragment chamber. *a* - the fissioning material; *b* - collecting electrodes; *c* - to amplifier grid.

Electronics consisted of a preamplifier, wide band amplifier, discriminator, scaler, and mechanical register.

The preamplifier was located in the synchrocyclotron room next to the ionization chamber. Signal was conducted out by a 40 meter armored coaxial cable to the amplifier.

Two types of linear amplifier were used in this work (LU-1 and LU-2). The upper frequency limit of LU-1 was 1.3 mc with an amplification factor 20,000 to 25,000.

The upper frequency limit of LU-2 was 8 mc, with an amplification factor of 8,000 to 10,000. Figure 2 shows the dependence of chamber count-

<sup>11</sup> V. P. Dzheleпов and Iu. M. Kazarinov, Dokl. Akad. Nauk SSSR 99, 939 (1954).

ing rate on the discriminator clamp voltage for the fission of U, Bi, and W. The noise level was some 50-60 times less than the smallest fission fragment pulse.  $\alpha$ -particle signals were up to 0.1 the size of the fission fragment pulse. The basic difficulty of measurement was connected with the necessity of cutting off possible registration of the frequently collected charged particles which originated in the chamber gas or walls and electrodes due to high energy neutron collisions. The difficulty depends to a large extent on the pulsed nature of the accelerator; it increases as the fission cross section of the investigated nucleus decreases, since in this case low neutron beam currents are convenient for removal of the background, but necessitate measurements of long duration. The necessity of removing the background demands the use of wide band amplifiers. Special experiments carried out in chambers without the fissioning layers showed that the background under typical working conditions was small (order of several percent) even compared to the fission effect of wolfram.

As can be seen from Fig. 2, in all cases investigated, there was observed a sufficiently wide plateau, where the counting rate varied weakly with the discriminator clamping level. Thus for uranium the plateau stretched from 5 to 12 volts, with a slope of 3% per volt. This slope was evidently connected with fringe effects, the effect of fissioning material layer thickness, and the emission of fragments at various angles. Measurements were carried out at the center of each plateau.

Thin layers of  $U^{235}$  and  $U^{238}$  were deposited on the aluminum cathode by electrolysis of a solution of uranium nitrate in absolute alcohol ( $\sim 1$  g/liter), using a platinum anode. With a current density of  $0.5$  ma/cm<sup>2</sup>, a firm yellow-green deposit of thickness  $\sim 0.5$  mg/cm<sup>2</sup> was formed in 10 minutes. The thickness of uranium on the working electrodes was  $0.15 - 0.5$  mg/cm<sup>2</sup>. In view of the impossibility of baking the aluminum foils (with consequent change of the uranium to higher oxides), we assumed that the composition of the fissioning layer was between  $UO_2$  (11.9% oxygen by weight) and  $U_2O_3$  (9.2% oxygen by weight), i.e., we assumed that the layer was 89.5% uranium by weight. The  $U^{235}$  content of the isotope mixture used was 92.6%, so that 82.8% of the fissioning layer was  $U^{235}$ . The  $U^{235}$  ionization chambers were shielded by boron carbide and cadmium to eliminate the background due to fission by thermal neutrons. A thorium layer of  $\sim 1$  mg/cm<sup>2</sup> was deposited on the aluminum foil in the form of a suspension of finely ground  $ThO_2$  in acetone with 1% (by weight)

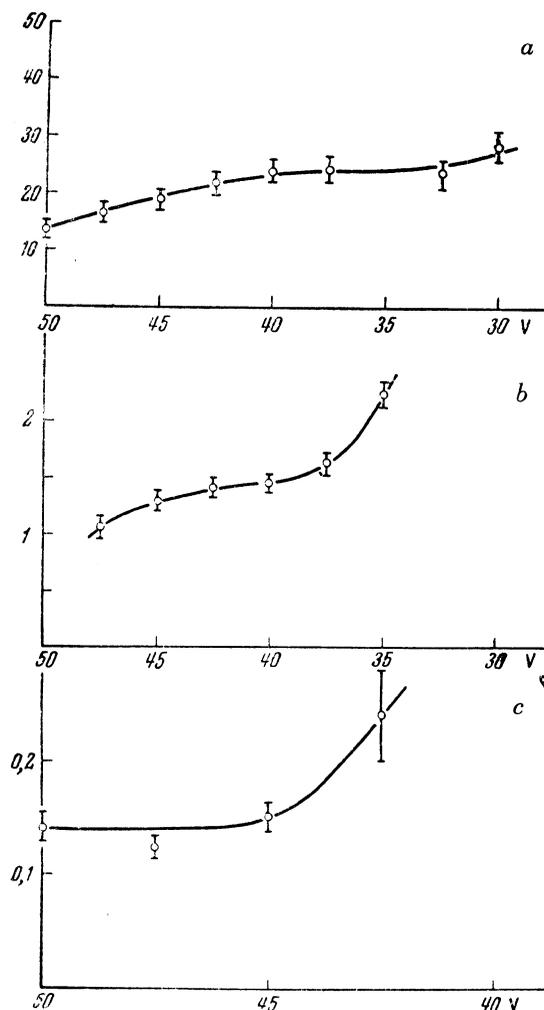


FIG. 2. Dependence of the relative fission counting rate on discriminator locking potential for a - uranium, b - bismuth and c - wolfram.

organic binder. Layers of bismuth, lead and thallium ( $1 - 1.5$  mg/cm<sup>2</sup>) were produced by vacuum evaporation onto the aluminum foil, while the gold and platinum layers ( $\sim 0.8$  mg/cm<sup>2</sup>) were produced by cathode sputtering.

Wolfram was electrolytically deposited onto a copper cathode, (with platinum anode) in a bath consisting of 125 gm  $H_2WO_4$  and 330 gm  $Na_2CO_3$  per liter of solution. With a current density of  $50$  ma/cm<sup>2</sup>, a silvery metallic deposit was formed in 30 minutes, with thickness  $\sim 0.8$  mg/cm<sup>2</sup>. From the data of reference 12 the composition by weight of wolfram in such a deposit is  $\sim 65\%$ .

<sup>12</sup> V. I. Lainer and N. G. Kudriavtsev, *Fundamentals of Electroplating*, ch. 2, ONTI, USSR, 1938.

To determine the cross section, the incident neutron current was measured with the aid of carbon activated in the  $C^{12}(n,2n)C^{11}$  reaction. The absolute detection efficiency of the activity of the product of this reaction – the  $C^{11}$  isotope (the count of the carbon detectors) was measured in special experiments. The cross section of  $C^{12}(n,2n)C^{11}$  at  $E_n = 90$  mev is  $0.022$  barns<sup>13</sup> and at 380 mev it is  $0.021$  barns<sup>14</sup>. The latter quantity was obtained in reference 14 by the comparison of  $np$  scattering data with the activation of carbon detectors, using our data for the count of these detectors. Therefore, possible errors in the absolute value of the activation cross section and in the count of the carbon detectors tend to cancel for  $E_n = 380$  mev.

In a series of the special experiments, during which good constancy of the high energy neutron current was maintained, the fission cross section of bismuth was obtained by comparison with the carbon detectors. For subsequent determinations of fission cross sections of other nuclei, a bismuth fission chamber was used to monitor the neutron beam. In such a comparison, fluctuations of the neutron intensity could not affect the measured quantities.

The accuracy of the absolute values of the fission cross sections given below is connected with the determination of the bombarding neutron current with the carbon detectors and with

statistical errors, and is taken as 20-30% for  $E_n = 120$  mev and 15-25% for  $E_n = 380$  mev. It must be noted that in the determination of the bombarding neutron current with carbon detectors, the magnitude of the current is lowered for the registration of U and Th fission and is raised for the lighter nuclei, the amount of rise increasing with the fission threshold. Therefore the fission cross sections indicated below are undoubtedly too high for U and Th, and too low for the other nuclei. Furthermore, the average effective neutron energy causing the fission is higher the greater the fission threshold and the faster the growth of the fission cross section with energy. The indicated errors are common to all work with high energy neutrons where it is impossible to define a narrow region in the neutron energy spectrum. Approximate evaluation of low energy neutron admixture in the outgoing spectrum shows that the increase of fission cross section of U and Th is not greater than 5-10%. Corrected values of the bismuth fission cross section, taking account of the spectrum shape and the excitation function, are shown in Fig. 3. Such a correction was not performed for the lighter nuclei because of insufficient data on the dependence of their fission cross section on energy.

Data on high energy neutron fission cross sections of nuclei from U to W from our work (no superscript) as well as from other authors<sup>8,15,16</sup> are given in Table 1.

TABLE I. Fission cross sections of heavy nuclei for high energy neutrons (in barns).

$E_n$ (mev)	U <sup>235</sup>	U <sup>238</sup>	Th	Bi	Pb	Tl	Au	Pt	Re	W
84	—	1.4 <sup>[8]</sup>	1 <sup>[8]</sup>	0.019 <sup>[8]</sup>	0.0055 <sup>[8]</sup>	0.0032 <sup>[8]</sup>	0.0021 <sup>[8]</sup>	0.00095 <sup>[8]</sup>	—	—
120	1.5	1.14	1 <sup>[15]</sup>	0.036	0.020 <sup>[16]</sup>	0.010 <sup>[16]</sup>	0.010 <sup>[16]</sup>	—	0.0017 <sup>[15]</sup>	0.0011 <sup>[15]</sup>
380	1.24	1.03	0.9	0.074	0.033	0.019	0.020	0.012	—	0.0038

Figure 3 shows the excitation function for the fission of bismuth, for a nominal neutron energy (curve 1) and corrected for the spectrum shape (curve 2). Values up to 84 mev and for 270 mev

are taken from the literature<sup>8,17</sup> the remaining points are from our work. The satisfactory agreement of all points confirms the determination of the absolute fission cross section.

<sup>13</sup> E. McMillan and H. York, Phys. Rev. 73, 262(1948).

<sup>14</sup> K. O. Oganessian, Report Inst. Nuclear Problems, Acad. Sci. USSR, 1953.

<sup>15</sup> V. P. Dzhelepov, B. M. Golovin, Iu. M. Kazarinov, Report Inst. Nuclear Problems, Acad. Sci. USSR, 1950.

<sup>16</sup> A. A. Reut, G. I. Celivanov and V. V. Iur'ev, Report Ins. Nuclear Problems, Acad. Sci. USSR, 1950.

<sup>17</sup> W. Knox, Phys. Rev. 81, 687 (1951).

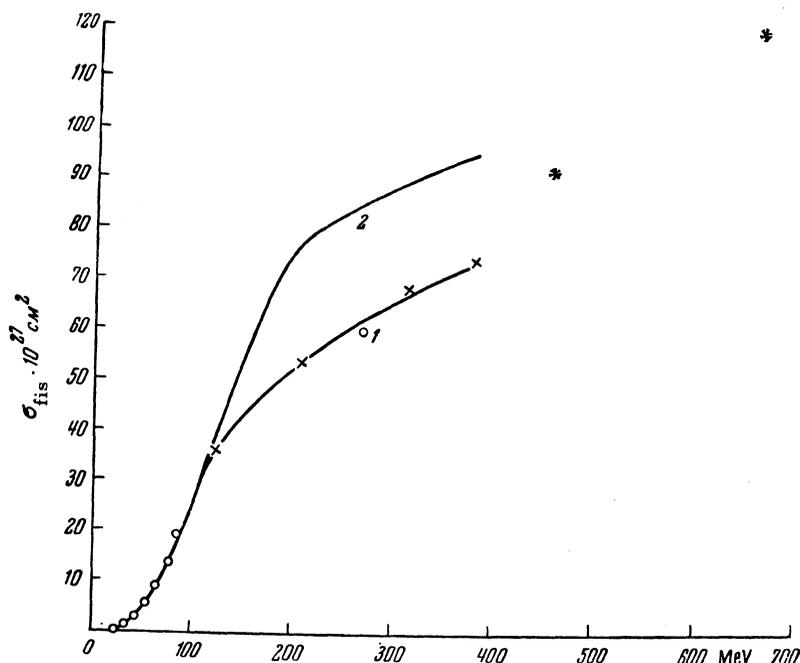


FIG. 3. Dependence of fission cross section of bismuth on neutron energy. 1 - without corrections for the energy spectrum of the incident neutrons; 2 - with such correction. The stars indicate data on the fission of bismuth by protons<sup>23</sup>.

### 3. EVALUATION OF THRESHOLD FOR EMISSIVE FISSION

During the excitation of a heavy nucleus to high energies, the most likely processes are apparently emission of neutrons and fission. It is clear from the facts given in the introduction that the fission widths for stable nuclei from Bi to W are much narrower than the corresponding neutron widths. Evidently according to the emission of neutrons, the ratio of the fission width to the total width, which determined the probability for fission, must increase with increase of the parameter  $Z^2/A$ . Since there are no qualitative data relating the fission probability and the neutron emission for a highly excited nucleus, we have, in treating our experimental data, made the assumption<sup>9</sup> that emissive fission does not take place earlier than the moment at which the binding energy of the next neutron (increasing with the neutron emission) becomes larger than the critical fission energy of the remaining nucleus (which decreases according to the number of emitted neutrons).

The neutron binding energy is calculated from the Weizsacker-Fermi formula:

$$E_{bn} = -5.32 - 8.68 A^{-1/3} + (Z^2/A)(77.4 A^{-1}) \quad (1)$$

$$+ 0.195 A^{-1/3} \pm 33.5 A^{-2/3}.$$

The last term is added for even-even and odd-even nuclei (i.e., for even  $N = A - Z$ ), and is subtracted for even-odd and odd-odd nuclei (i.e., with odd  $N$ ). To determine the binding energy for nuclei with odd  $A$ , the last term should be used with  $A - 1$  instead of  $A$ . For heavier, even  $N$  nuclei, whose neutron binding energy is higher, and for which, in general, the indicated conditions for emissive fission are satisfied,

$$E_{bn} \approx -6.16 + (Z^2/A)(77.4 A^{-1}) \quad (2)$$

$$+ 0.195 A^{-1/3}.$$

In the determination of the neutron binding energy for nuclei with  $A > 208$ , in agreement with reference 18, a correction term should be put in which reduces the binding energy by

<sup>18</sup> O. Stern, *Revs. Mod. Phys.* **21**, 316 (1949).

$$\Delta E = 1.97 \exp \{ -(A - 208) / 208 \} \text{ MeV.} \quad (3)$$

In determining the neutron binding energy from Eqs. (2) and (3) we are naturally not taking account

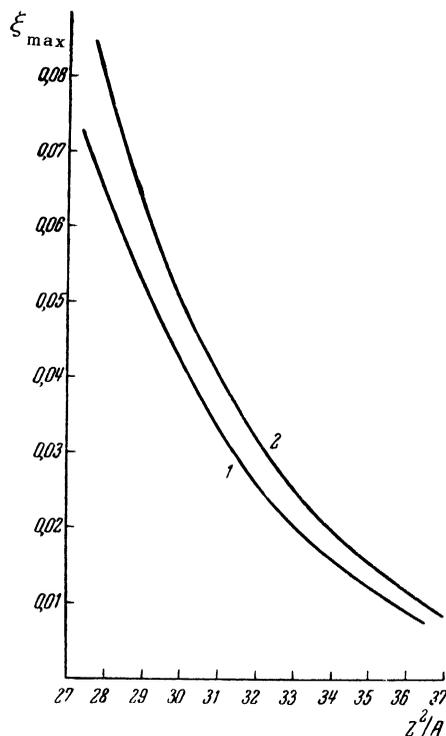


FIG. 4

FIG. 4. Dependence of the quantity  $\xi_{\max}$  in the critical energy expression on the parameter  $Z^2/A$ ; 1 - according to Hill and Wheeler<sup>7</sup> ( $E_{\text{fis}} = 13 A^{2/3} \xi_{\max}$ ); 2 - according to Frankel and Metropolis<sup>21</sup> ( $E_{\text{fis}} = 14 A^{2/3} \xi_{\max}$ ).

of the fluctuations in nuclear density connected with the filling of shells.

To calculate the critical fission energy, use was made in reference 9 of the results of the first paper of Bohr and Wheeler<sup>19</sup>; in our earlier paper<sup>20</sup> we worked from the results of Frankel and Metropolis<sup>21</sup>. However evaluation of the fission threshold from reference 21 produces values that are too high - for example the calculated threshold of  $\text{U}^{238}$  is higher by 1.7 mev than that experimentally observed, and for  $\text{Th}^{232}$  by 2.8 mev. Thus, our use<sup>20</sup> of the results of reference 21 for

the evaluation of the critical fission energy gave high values for the number of neutrons emitted before the neutron binding energy is comparable to the fission barrier\*.

Below we have made use of the results of a recent paper by Hill and Wheeler<sup>7</sup> to determine the critical fission energy. This gives

$$E_{\text{fis}} = 13 A^{2/3} \xi_{\max} \quad (4)$$

in good agreement with the experimental value. The quantity

<sup>19</sup> N. Bohr and J. Wheeler, Phys. Rev. **52**, 426 (1939).

<sup>20</sup> V. I. Gol'danskii, E. Z. Tarumov and V. S. Pen'kina, Dokl. Akad. Nauk SSSR **101**, 1027 (1955).

<sup>21</sup> S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).

\* It must be noted that a rough extrapolation was used in the series of values from reference 20 presented in Table I, and no account was taken of the difference in binding energy for nuclei with odd  $N$  and  $Z$ , so that these results are only of qualitative value.

$$\xi_{\max} = f(x) = 0,728(1-x)^3 - 0,661(1-x)^4 + 3,330(1-x)^5,$$

where  $x = (Z^2/A) / (Z^2/A)_{\text{initial}}$ , and  $(Z^2/A)_{\text{initial}} = 46.78$ . The dependence of  $\xi_{\max}$  on  $Z^2/A$  is shown by curve 1, Fig. 4. For comparison, curve 2 shows the corresponding dependence according to Frankel and Metropolis<sup>21</sup>, who give

$$E_{\text{fis}} = 14A^{2/3} \xi'_{\max} \quad (5)$$

Using (2), (3), and (4), we determined the mass number  $A_c$  at which the neutron binding energy becomes greater than the critical fission energy for various nuclei. The dependence of  $A_c$  on  $Z$  is shown in Fig. 5. The main results of the calculations are presented in Table 2.

The first column of this table gives the mass number of the most stable isotopes or the average mass number of the natural isotopic mixture. The second column shows calculated values (according to reference 7) of the critical fission energy for the initial nucleus, while column four shows this for a nucleus of mass number  $A_c$ , as given in column

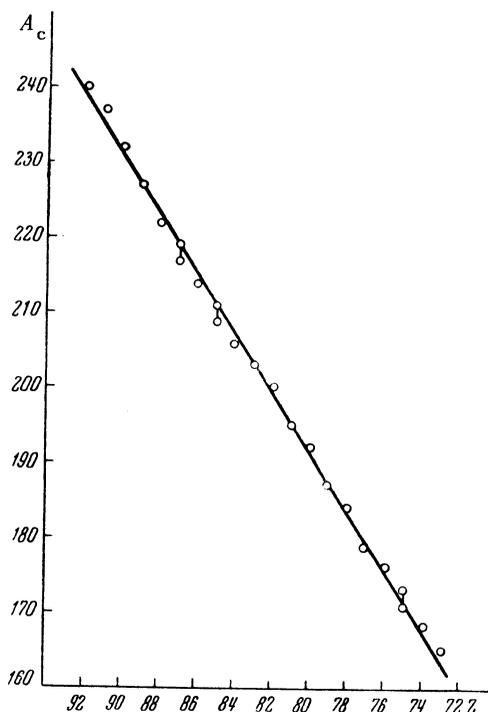


FIG. 5

FIG. 5. Dependence of the mass number  $A_c$  (at which the critical fission energy, calculated according to Hill and Wheeler<sup>7</sup>, becomes less than the neutron binding energy) on atomic number  $Z$ .

three. The fifth column gives the fission parameter  $Z^2/A_c$ . The next three columns give the minimum number of neutrons (not counting the incident neutron) which must be emitted by the nucleus before emissive fission, with the emission of only neutrons, or with the addition of one or two protons. The last three columns show the values of the energy spent in the case of emissive fission

with the emission of neutrons only, — the binding energy  $E_b$  of  $N_c = A - A_c$  neutrons, their average kinetic energy ( $E_{\text{kin}}$ ), and the average total energy of such a fission ( $E_{\text{tot}} = E_b + E_{\text{kin}} + E_c$ ). The average neutron kinetic energy was calculated from the relation  $\bar{E}_{\text{kin}} = 2T$ , where the nuclear temperature  $T$  is connected to the excitation energy  $U$  by the relation  $T = \alpha \sqrt{U}$ . For this calculation we

TABLE II. Evaluation of emissive fission thresholds.

Nucleus	$E_{\text{fis}}^{\text{c}}$ (mev)	$A_{\text{c}}$	$E_{\text{fis}}$ (mev)	$Z^2 / A_{\text{c}}$	Number of emitted neutrons			Binding energy of $N_{\text{c}}$ neutrons $E_{\text{b}}$ (mev)	Kinetic energy of $N_{\text{c}}$ neutrons $E_{\text{kin}}$ (mev)	Total fission energy $E_{\text{tot}}$ (mev)
					Only $n$ $N_{\text{c}}$	$1 p$	$2 p$			
$^{92}\text{U}^{235}$	4.4	(240)	(5.7)	(35.27)	0	0	1	—	—	4.4
$^{92}\text{U}^{238}$	5.2	(240)	(5.7)	(35.27)	0	0	4	—	—	5.2
$^{91}\text{Pa}^{231}$	4.6	(237)	(6.2)	(34.94)	0	0	2	—	—	4.6
$^{90}\text{Th}^{232}$	6.2	232	6.2	34.91	0	4	8	—	—	6.2
$^{89}\text{Ac}^{227}$	6.3	227	6.3	34.89	0	4	6 (8)	—	—	6.3
$^{88}\text{Ra}^{226}$	7.1	222	6.1	34.88	4	6 (8)	10	25	8	39
$^{87}\text{Fr}^{223}$	7.4	219	6.5 (6)	34.56	4 (6)	8	10 (12)	25 (38)	8 (12)	39 (56)
$^{86}\text{Rn}^{222}$	8.8	214	6.4	34.56	8	10 (12)	14	50	22	78
$^{85}\text{At}^{210}$	6.6	211	7 (6.3)	34.24	0 (1)	3	5	(6.4)	(1.1)	6.6
		(209)		(34.57)						(13.8)
$^{84}\text{Po}^{209}$	7.7	206	6.8	34.25	3	5	7	19	7	33
$^{83}\text{Bi}^{209}$	9.3	203	7.3	33.94	6	8	12	39	15	61
$^{82}\text{Pb}^{206}$	10.1	200	7.7	33.62	6	10	12	41	15	64
$^{81}\text{Tl}^{204}$	11.6	195	7.6	33.65	9	11	15	63	28	99
$^{80}\text{Hg}^{201}$	12.6	192	8.1	33.33	9	13	15	63	29	100
$^{79}\text{Au}^{197}$	12.4	187	7.9	33.37	10	12	16	74	35	117
$^{78}\text{Pt}^{195}$	13.6	184	8.3	33.07	11	15	17	82	42	132
$^{77}\text{Ir}^{192}$	15.5	179	8.3	33.12	13	15	17 (19)	98	53	159
$^{76}\text{Os}^{190}$	16.6	176	8.7	32.82	14	16 (18)	20	106	59	174
$^{75}\text{Re}^{186}$	16.9	173	9.1 (8.3)	32.51	13 (15)	17	19	100 (118)	54 (67)	163
		(171)		(32.89)						(193)
$^{74}\text{W}^{184}$	18.9	168	8.8	32.60	16	18	—	128	73	210
$^{73}\text{Ta}^{181}$	19.7	165	9.5	32.30	16	—	—	128	74	212

took the numerical value of  $\alpha$  directly from the model of a Fermi gas which gives  $\alpha = 2.92 A^{-1/3}$  (for a Fermi surface of 21 mev). The calculation of the critical fission energy according to Hill and Wheeler<sup>7</sup>, and the improvement of the calculation of  $A_{\text{c}}$  and the neutron binding energy taking account of whether the nucleus is even or odd, gives a significantly reduced value of  $N_{\text{c}}$  and the energy of emissive fission compared to reference 20, so that the results shown in Table 2 are close to our previous evaluations given in reference 9.

It follows from the above assumptions on the conditions for emissive fission that we may consider the fission section of each nucleus as the cross section of emission before fission of  $N > N_{\text{c}} = A - A_{\text{c}}$  neutrons. Then the fission cross section of the given nucleus is

$$\sigma_{N_{\text{c}}} = \sigma_0 \int_{N_{\text{c}}}^{\infty} w(n) dn,$$

where  $\sigma_0$  is the fission cross section of a nucleus which can divide without the emission of neutrons, and  $w(n)$  is the normalized probability of emission before fission of  $n$  neutrons. Comparing the fission

cross sections of various nuclei, it is possible to determine the function  $w(n)$ , i.e., distribution of the fission-accompanying neutron stars by the number of prongs, as well as to determine the average number of neutrons (for  $n > N_{\text{c}}$ ) emitted by the given nucleus before fission. It is necessary in such an evaluation to take account of the possibility that the excited nucleus will emit charged particles in addition to neutrons before fission. From their investigation of uranium fission in thick emulsions by 150 and 380 mev neutrons, the authors of reference 22 conclude that, in practice, the emission of one charged particle should be taken into account in each case of fission. We shall therefore work on the assumption that in each fission event one proton is emitted either during the internal cascade or in the subsequent evaporation process. Then we may, using the data of Table I, and taking the fission cross section of  $\text{U}^{238}$  for  $\sigma_0$ , determine the dependence of  $\sigma/\sigma_0$  on  $N_{\text{c}}$  shown in Fig. 6. From this dependence and the

<sup>22</sup> G. E. Belovitskii, T. A. Romanova, L. V. Sukhov and I. M. Frank, J. Exper. Theoret. Phys. USSR **29**, 537 (1955); Soviet Phys. JETP **2**, 493 (1956)

assumption that one proton is emitted before fission, we obtain the following values for the number of neutrons emitted before fission of  $U^{238}$  for various bombarding energies:  $E_n = 84$  mev,  $\bar{N} = 6.0$ ;  $E_n = 120$  mev,  $\bar{N} = 6.7$ ;  $E_n = 380$  mev,  $\bar{N} = 7.1$  (here the incident neutron is already taken into account). For lighter nuclei, the average number of neutrons emitted before fission turns out to be higher - for example in bismuth (from Fig. 5), normalizing to  $\sigma = \sigma_0$ , we obtain  $E_n = 84-380$  mev,  $\bar{N} = 11-11.5$  (counting the incident neutron).

The results obtained are in satisfactory agreement with purely radiochemical investigation of the fission products of uranium<sup>1</sup> and bismuth<sup>2</sup> in high energy reactions; the average number of neutrons emitted during a fission event was there found to be 6-12 for uranium and 12 for bismuth. In comparing our values of  $\bar{N}$  with those found by other methods, it should be borne in mind that the smeared out energy spectrum of the bombarding neutrons which raises the fission cross sections of U and Th, and lowers the fission cross sections of the lighter nuclei, has the effect of somewhat lowering the value of  $\bar{N}$ . The excitation energy necessary for emissive fission shown in Table II for nuclei heavier than Ta, and calculated by us for lighter nuclei in reference 9, is also in satisfactory agreement with the results of emulsion studies of emissive fission by Shamov<sup>23</sup>.

Such agreement with experiment seems to justify the simple qualitative interpretation we have made of the fission of heavy nuclei by high energy particles, based on equating the neutron binding energy with the critical fission energy of the nucleus after the emission of neutrons.

The comparisons between experiment and calculation can not aspire to be quantitatively exact, if only because the data represent averages over the isotopic composition of the elements and over the energy spectrum of the bombarding neutrons. However, even the qualitative observations allow some conclusions to be drawn about the general nature of fission of various nuclei by high energy particles, and allow the proposal of some experiments to check on the assumptions that were made above. For example, it follows from Fig. 6 that the ratio  $\sigma/\sigma_0 = f(N_c)$  falls off very rapidly for nuclei lighter than thorium. If we take into account the lowering of the fission cross sections of lighter nuclei by the smearing of the neutron energy spectrum, then the tails of the curves on Fig. 6 for  $z = 74-78$  are raised strongly. The dependence of  $\sigma/\sigma_0$  on  $N_c$  is then divided into

three regions - slow decrease (from U to Th, and for nuclei lighter than Au), and a region of fast decrease from Th to Au. The existence of three such regions is apparently connected with the fact that for nuclei from U to Th and lighter than Au in the present energy interval, fission may occur with the emission of less than  $N_c = A - A_c$  neutrons. This conclusion is in agreement with the theoretical results of Geilikman<sup>24</sup>, who examined the thermodynamics of excited and fissioning

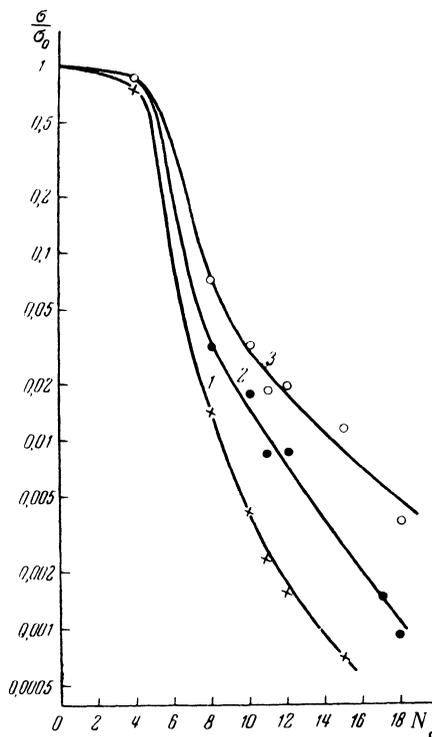


FIG. 6. Dependence of the fission cross section of nuclei on the minimum number of neutrons which must be emitted before fission (assuming that one proton is also emitted). 1 -  $E_n = 84$  mev; 2 -  $E_n = 120$  mev; 3 -  $E_n = 380$  mev.

nuclei. He concluded that high temperature fission from a highly excited state may compete with emissive fission for the heaviest nuclei where  $\sigma_{\text{fis}} \approx \sigma_{\text{geom}}$ , and for the relatively light nuclei, which must emit a very large number of neutrons for emissive function.

Similar conclusions can be drawn from recent

<sup>23</sup> V. P. Shamov, Dissertation, Radium Inst. Acad. Sci. (RIAN), 1955.

<sup>24</sup> B. T. Geilikman, Report of the Acad. Sciences USSR, 1950.

Soviet work on the fission of heavy nuclei by high energy particles using emulsions<sup>25</sup> as well as radiochemical methods<sup>26,27</sup>.

To check on the assumptions we have made it would be desirable to perform experiments with

<sup>25</sup> N. A. Perfilov, N. S. Ivanovna, O. V. Lozhkin, V. I. Ostroumov and V. P. Shamov, paper at the July 1955 meeting of the Academy of Sciences, USSR.

<sup>26</sup> A. P. Vinogradov, I. P. Alimarin, V. I. Baranov, T. V. Baranova, A. K. Lavrukina and F. I. Pavlotskaia, paper at the July 1955 meeting of the Academy of Sciences, USSR.

<sup>27</sup> B. V. Kurchatov, V. N. Mekhedov, M. Ia. Kuznetsova and L. N. Kurchatova, paper at the July 1955 meeting of the Academy of Sciences, USSR.

monoenergetic neutrons. It would be interesting to measure the fission cross section for separated isotopes - particularly those with neutron deficit, in which case fission does not require multiple emission of neutrons. The fission cross sections of some similar isotopes (for example At<sup>207-209</sup>) must be of the order of  $10^{-25}$  cm<sup>2</sup>. The combination of data on emissive fission of various isotopes at high energies may be used to determine the neutron widths and fission widths of strongly excited heavy isotopes with neutron deficits, which are not at all formed in experiments at low energies.

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### Investigation of the Structure of the Surface of Films of Copper Oxide on Different Faces of a Monocrystal of Copper, and the Determination of the Contact Potential Difference between These Surfaces

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The structure of the surfaces of films of copper oxide formed on various faces of a monocrystal of copper is investigated using the method of diffraction of slow electrons. It is found that on faces (100) and (111) of a monocrystal of copper there is formed a monocrystalline film of Cu<sub>2</sub>O with a (111) plane parallel to the base. On face (110) a monocrystalline film of Cu<sub>2</sub>O is also formed but with a less perfect crystalline structure and with a (110) surface plane. Investigation of the contact potential difference by the method of displacement of the volt-ampere characteristic curves demonstrated that films of Cu<sub>2</sub>O on faces (100) and (111) of a monocrystal of copper have the same work function (0.2V), and consequently, have the same crystalline orientation. The film of Cu<sub>2</sub>O on the (110) face of copper possesses a larger work function (0.3v), and also has stronger adsorptivity of residual gases than films of Cu<sub>2</sub>O on faces (100) and (111), which indicates a difference in the properties (orientations) of the film of Cu<sub>2</sub>O on face Cu (110) and of films of Cu<sub>2</sub>O on faces Cu (100) and (111).

**I**N order to clarify the results obtained during the investigation of secondary electron emission from the surfaces of various faces of a monocrystal of copper which were covered with films of copper oxide<sup>1</sup>, it was necessary to determine the

crystalline structure of these films and also the work function of the surface of these films. With this aim in mind, we carried out investigations of the surface structure of monocrystalline specimens by the method of diffraction of slow electrons and the contact potential difference between the surfaces of the investigated specimens.

Numerous investigations have demonstrated that after pure copper, polycrystalline or monocrystalline, is exposed to the atmosphere for a certain

<sup>1</sup> N. B. Gorny, J. Exper. Theoret. Phys. USSR **26**, 79, 327 (1954).