SPONTANEOUS FISSION OF NUCLEI

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I. HISTORY OF THE DISCOVERY OF SPONTANEOUS FISSION

THE very first work on the investigation of natural radioactivity and nuclear reactions showed that there are enormous stores of energy hidden inside atomic nuclei. However, the use of this energy did not seem possible: radioactive elements are found in the earth's crust in negligible amounts; as for nuclear reactions, the energy liberated in them was many orders of magnitude smaller than the energy expended in producing them. For the practical use of the energy inside nuclei, it was first necessary to find a nuclear reaction which could be established essentially as a self-maintaining process, i.e., it was necessary to achieve a nuclear chain reaction. The search for such reactions was carried on quite intensively; as a result of the study of a large number of nuclear reactions, people succeeded in finding various possible schemes which would enable one to relate the energy liberated in stars with nuclear chain reaction processes. However, there was no hope of establishing a process of such type under terrestrial conditions.

In December, 1938, there appeared the first report of the discovery by Hahn and Strassmann of the fission of uranium nuclei by neutrons.[1] Further reports of the work of Frisch and Meitner[2] and of F. Joliot[3] and collaborators not only confirmed this discovery, but also showed that the fission of uranium by slow neutrons liberates an energy which is tens of times greater than in any other nuclear reaction. As a result of the fission, there should be produced excited nuclei which could emit neutrons with high probability. Just this feature of fission in principle opened the possibility for producing a nuclear chain reaction. It was natural that the physicists of all countries of the world began to study this phenomenon intensively.

In the Soviet Union at that time there was formed a powerful group of physicists — pupils and collaborators of I. V. Kurchatov, who worked successfully in the field of nuclear reactions and artificial radioactivity. The beginning of their work dates from 1933 when Kurchatov, who had finished a whole series of brilliant investigations of ferroelectricity, began working in the field of nuclear physics.

In the Leningrad Physico-Technical Institute, (LPTI), Kurchatov directed a laboratory which was closely associated with other scientific groups working in this same field: the Physics Section of the Radium Institute, the Pokrovsk Pedagogical Institute, and the Ukrainian Physico-Technical Institute.

In the LPTI, at the seminar on neutron physics which Kurchatov directed, there were discussions of practically all the important work carried on in the Soviet Union and in other countries. At this same seminar there were detailed discussions of proposed experiments, the results of experiments which had been carried out, and there were theoretical treatments of the phenomena under investigation.

In 1938 Kurchatov and his co-workers carried out many important studies. In a long series of experiments they developed and considerably supplemented the results of the studies of Fermi's group on the interaction of neutrons with nuclei.[4] They discovered the phenomenon of nuclear isomerism of artificially radioactive nuclei and showed that the main path of de-excitation of the nucleus from the isomeric level is by internal conversion.[5] In a series of experimental and theoretical papers they obtained results which could not be incorporated within the realm of the compound nucleus theory of N. Bohr, which dominated the field at that time.

These results were obtained with relatively simple experimental apparatus. The neutron sources were ampoules filled with a mixture of beryllium and radon, the detectors were silver and rhodium foils; all the electronic part of the apparatus was usually prepared by the experimenters themselves.

At the same time there were developed more powerful and complete experimental techniques. The first cyclotron in Europe was built at the Radium Institute. On the initiative of, and under the direction of Kurchatov and A. I. Alikhanov the construction of a still more powerful cyclotron was begun in the LPTI. At the Ukrainian Physico-Technical Institute, unique electrostatic generators were built. At the LPTI they successfully developed proportional counters with lithium layers and ionization chambers filled with BF_3 for counting neutrons. In collaboration with the Institute for Radio Reception and Acoustics, work was carried out on the production of a linear amplifier with a record value of amplification coefficient (for that time) of more than a million.

The enthusiastic work on scientific and technical problems in nuclear physics, the heated arguments at the seminar when new ideas and experiments were discussed — all this produced an atmosphere in which there was a very rapid rise in creative power and the formation of a brilliant scientific group able to solve difficult scientific problems.

It is impossible to overestimate the role of Kurchatov in all this. The most profound erudition, a knowledge of the whole range of experimental technique, and, most important, an exceptional scientific intuition enabled Kurchatov to choose the main paths for his investigations and to set before his group questions whose solution determined the answer to their problem.

Now, when we are separated by two decades from those romantic days of 1939 — 1940, when we first began to see the possible use of nuclear energy, it seems entirely logical that after the discovery of the fission of uranium all the efforts of the groups directed by or associated with Kurchatov immediately began a study of just these problems.

The first series of experiments in the new direction were related to the study of the most important features of nuclear fission. To estimate the possibility of a chain reaction in uranium, it was extremely important to find out whether (and how many), neutrons were emitted from fission, and also which of the uranium isotopes could be induced to fission by slow neutrons. As we now know, the answer to this question was independently found as a result of experiments carried out practically simultaneously in the laboratories of many countries.[6-9] The theoretical analysis of the conditions for the establishment of a self-maintaining nuclear chain reaction were given first in a fundamental paper by Ya. B. Zel'dovich and Yu. B. Khariton.[10]

On the basis of the experimental results and theoretical estimates obtained, Kurchatov already in those days clearly saw the basic way of using nuclear energy and pointed out the technical difficulties with which one has to struggle for the practical solution of this problem. Kurchatov presented his arguments in a well-known report[11] which is a brilliant example of profound analysis of a complicated scientific problem. This analysis showed that the establishment of a chain reaction by slow neutrons required a large amount of U-235. For the separation of such an amount of U^{235} from the natural mixture of isotopes there were no methods developed at that time. One could avoid this difficulty by using very large amounts of heavy water and helium, but this was likewise not practically feasible.

There remained an open question whether one could establish a chain reaction with fast neutrons using a large mass of natural metallic uranium. The answer to this question depended primarily on the value of the cross section for inelastic scattering of fast neutrons by uranium, since inelastic scattering is in this case a process competing with fission.

At that time our knowledge of these processes was by no means sufficient, so that experiments were thought of for studying inelastic scattering of neutrons by uranium and other elements.

In addition it was proposed to make precise measurements of the total cross section for fission of the natural mixture of uranium isotopes by photoneutrons with energies of 130, 220, and 860 kev. At the same time Kurchatov proposed an experiment which, without a detailed knowledge of the various constants and computations, should directly answer the question of the possibility of obtaining a chain reaction in a large mass of metallic uranium. The experiment consisted in observing the change in neutron flux produced by fission under conditions where, inside a massive sphere of metallic uranium, one placed neutron sources with various energy spectra. This was essentially an "experimentum crucis" which should directly answer the question.

To carry out all these experiments it was necessary to have detectors of the neutrons produced by uranium fission, with a sensitivity many tens of times greater than in the usually used fission chambers. The development of such a detector, a special fission chamber, made it possible on the one hand to answer the question of the possibility of a chain reaction in normal metallic uranium, and on the other led to the discovery of a new form of radioactivity — the spontaneous fission of the uranium nucleus.

A fission chamber of high sensitivity was developed at the beginning of 1940 as a result of numerous experiments with various varieties of construction. It was a flat, multi-layer condenser, with a total effective area of about 1,000 cm^2 (Figs. 1 — 3). The amount of uranium which could be put into this chamber was 30 — 50 times greater than in ordinary fission chambers. Later on in the same year of 1940 they succeeded in producing a multi-layer chamber with an operating area of ~ 6,000 cm^2 and a sensitivity approximately 200 times greater than for ordinary chambers.

They had to overcome many technical difficulties before they had complete confidence in the trustworthy

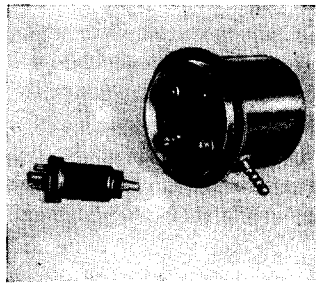

FIG. 1. Multi-layer fission ionization chamber by means of which the spontaneous fission of uranium was first recorded.[12]

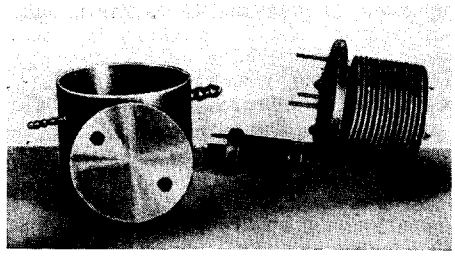

FIG. 2. Opened-up view of the chamber.

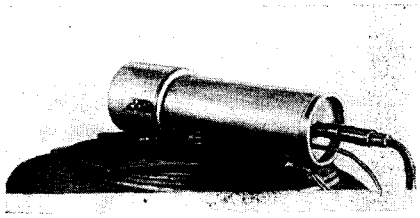

FIG. 3. Ionization chamber used in reference 12, in its operating position—on a double damping system of lead plates and rubber supports.

operation of the apparatus. The high sensitivity of the chamber for recording neutrons produced in fission made it possible quite easily to attack the problems for whose solution this apparatus had been made. In addition, in experiments for determining the background they made observations which, as already pointed out, led to the discovery of the spontaneous fission of uranium.

In the very first experiments for counting fission fragments, pulses were observed in the absence of a neutron source.[12] The number of these pulses was not large (on the average about six per hour), but their number and shape coincided with those observed for the fission fragments from fission of uranium by the action of neutrons. Careful experiments were undertaken to study the reason for the appearance of these "spontaneous" pulses. Kurchatov, carefully following the course of this work, took a very active part in a careful analysis of all these processes which might lead to the appearance of pulses of fission type. All the necessary control experiments were thought of and carried out.

Let us briefly describe a series of such experiments. First, it was assumed that the observed effect was not related to the presence of uranium in the ionization chamber, and was caused either as a result of some external vibrations or random pulses occurring in the electronic circuit. This assumption was not confirmed, since not a single pulse was recorded in a long experiment with the chamber without uranium oxide.

Furthermore, the possibility was not excluded that the "spontaneous" pulses occurred as a result of piling up of pulses from α particles. It is known that the number of such pile-ups depends very strongly on the α particle intensity. Therefore control experiments were made in which thorium emanation was introduced into an ionization chamber in such an amount that the ionization current produced by α particles associated with its decay was twice as great as that from the α particles of uranium itself. In this experiment no increase was observed in the effect, from which it follows that chance coincidences of α particles are excluded by the resolving power of the amplifier.

In addition, by special control experiments it was established that the "spontaneous" pulses of "fission" type could not be explained by the presence in any parts of the chamber of the conditions for gas amplification, or by chance discharges on the surface of the uranium oxide.

There was also carried out a series of experiments which confirmed the hypothesis that the recorded effect was related to the fission of uranium. First of all, they studied the dependence of the magnitude of the effect on the thickness of the layer of uranium oxide. Experiments were made with chambers in which the thickness of the uranium layer was 1.4, 2.2, and 5.7 mg/cm^2. It turned out that the intensities of the "spontaneous" pulses for these chambers were proportional to the counting rate of fission fragments when the chambers were irradiated by the same neutron flux. Such a result was to be expected if the effect is ascribed to fragments from uranium fission. The experimenters were even more confirmed in this conclusion when they succeeded in measuring the amplitude distribution of the mysterious pulses. In Fig. 4 is shown the distribution

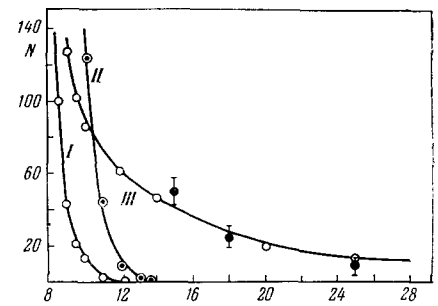

FIG. 4. Distribution of pulse heights in the ionization chamber. I, II – α particles, III – fission fragments.

in size of pulses from α particles (I), from fragments from fission by neutron bombardment (III) (along the abscissa are given the settings of an integral discriminator, along the ordinates a number proportional to the counting rate). Curve II was obtained by additional "loading" of the chamber by α particles from the decay products of thorium emanation. The distribution of pulse sizes of fission type in the absence of a neutron source (the black circles) practically coincides with curve III.

Using a chamber with a relatively thin layer of uranium, they succeeded in obtaining the differential pulse height distribution. In Fig. 5 is shown a comparison of

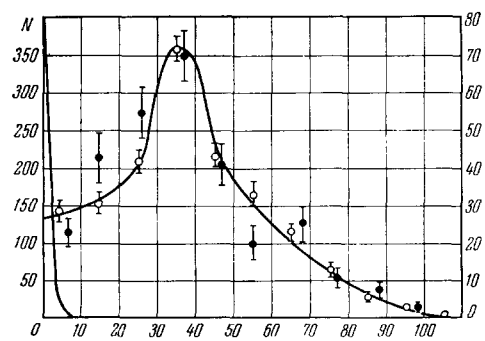

FIG. 5. Pulse height distribution of pulses in the ionization chamber. Open circles – induced fission of uranium; closed circles – spontaneous fission.

such a distribution for the "spontaneous" pulses (the closed circles) and for pulses from fragments from induced fission. The dependence of the number of pulses on size is the same in both cases.[13,14]

The whole set of results enabled one to draw the conclusion that the "spontaneous" pulses actually arose from fragments from the fission of uranium.

There remained only one point to explain: Is the observed "spontaneous" fission actually a spontaneous process, or does it have some external cause? For example, one might assume that the observed effect is produced by neutrons formed in the chamber as a result of the disintegration of light nuclei by the α particles from uranium. However, the known data concerning neutron yields from nuclear reactions showed that the intensity of the neutronic radiation which arose in the chamber was entirely insufficient to produce such an effect.

The appearance of a small number of fragments in the ionization chamber could be explained by the action on the uranium of cosmic rays: neutrons and mesons. The information which was available concerning the intensity of cosmic radiation and the cross section for its interaction with nuclei showed that this explanation was highly dubious. Nevertheless, to demonstrate experimentally the fact that the "spontaneous" fragments were not caused by cosmic rays, experiments were made in the shaft of the Moscow Subway under 50 meters of earth. Although the intensity of cosmic radiation at such a depth has been reduced approximately 40 times, the spontaneous fission effect still was the same as on the earth's surface.

All these numerous experiments and different types of control measurements led to the conclusion that uranium undergoes spontaneous fission.

It is necessary to point out that these experiments were not the first attempts to detect spontaneous fission. Bohr and Wheeler[63] first called attention to the possibility of such a process, but their estimates of the lifetime of uranium with respect to such a decay had the enormous value of 10^{22} years. In May of 1939 in the laboratory of the University of California, W. Libby made experiments for the purpose of determining the stability of uranium and thorium nuclei with respect to spontaneous fission.[15] He used two different methods, of which the first was radiochemical. Libby assumed that if there is spontaneous fission there will be produced the same groups of radioactive products as in the usual fission of nuclei under the action of neutrons. He made an attempt to separate the radioactive iodine and certain other fission products from a powder of uranium nitrate (approximately 200 grams). However, he did not succeed in detecting any residual radioactivity. Similar results were found in the corresponding operation with a large mass of thorium. The other method by means of which Libby attempted to find spontaneous fission of uranium and thorium was based on the search for secondary neutrons emitted in fission. As a neutron dectector he used a counter filled with BF_3 gas and surrounded by paraffin. When he placed uranium and thorium salts near the counter, he did not succeed in fixing any neutron activity. Starting from the negative results of these experiments, Libby gave as a lower limit for the lifetime of uranium and thorium with respect to spontaneous fission a value of 10^{14} years.

The success of the experiments carried out in Leningrad was the result of a substantial increase in sensitivity of the technique.

At this starting stage in the investigation, one could not state definitely which of the principal isotopes of uranium undergoes spontaneous fission. From the experimentally measured fission rate of a known amount of uranium, they calculated the half-lives for spontaneous fission under various assumptions concerning the mass number of the isotope.

As we now know, much later investigations showed that the observed effect of fission is associated with the heavy uranium isotope U^{238}. The value now accepted for the half-life[16] of uranium by spontaneous fission is 0.8×10^{16} years. The first value assigned to this quantity, which is indicated in the table, has been increased because of the inexact determination of the efficiency of the recording equipment for detecting the fragments.

Table I

Isotope	U^{238}	U^{235}	U^{234}
T_{sp}	$(4\pm1)\cdot10^{16}$	$(3\pm1)\cdot10^{14}$	$(2\pm0.5)\cdot10^{12}$

In the first work on spontaneous fission attempts were made to record also the spontaneous fission of thorium. No noticeable effect was observed with the thorium chamber and a lower limit of 10^{19} years was estimated for the half-life of Th^{232} with respect to spontaneous fission. Jumping a little ahead, we should remark that at the present time this limit has been raised by two more orders of magnitude: T_{sp} (Th^{232}) $> 10^{21}$ years,[17,18] which indicates the considerable successes which have been achieved in raising the sensitivity of the experiment in the course of the last 15 — 20 years.*

Because of the beginning of the Second World War, further experiments on the study of spontaneous fission had to be dropped for several years. In 1944 M. I. Pevzner and G. N. Flerov repeated the attempt of Libby to detect fragments from spontaneous fission

*In reference 89, the spontaneous fission of long-lived nuclei is treated in connection with the question of the stability of nucleons. The authors assume that the energy remaining in the nucleus after the decay of the nucleon is sufficient to give rise to fission with high probability. In reference 18, on the basis of the most recent data concerning T_{sp} for Th^{232}, it is stated that the lifetime of a bound nucleon with respect to decay into lighter particles is greater than 2×10^{23} years.

by radiochemical means.[19] Because of the higher sensitivity of the method compared to that of Libby, they succeeded in separating radioactive isotopes of iodine, I^{133} and I^{135}.

Experiments on the fission of nuclei by cosmic rays that were first carried out for determining the cosmic ray background in experiments on spontaneous fission have, in the postwar years, spilled over into a whole field devoted to the study of the fissioning component of cosmic rays. In 1945-1949 G. N. Flerov and V. I. Kalashnikova et al. carried out extensive studies of the fission of uranium and thorium by cosmic radiation.[20]

The first work in other countries confirming the results obtained in Leningrad in the laboratory of I. V. Kurchatov was that of G. Pose.[21] He also observed the effect of spontaneous fission of uranium by recording the neutrons accompanying this process. However, the results obtained by him for thorium ($T_{sp} = 1.7 \times 10^{17}$ years) are incorrect for the same reason as given above.

Right after the discovery of spontaneous fission it became clear that this phenomenon is interesting and important not just for nuclear physics. There were pointed out and worked out later a whole series of problems of chemistry, geology, and astrophysics for whose solution it was necessary to consider the process of spontaneous fission. Some of these aspects will be discussed in the following sections of this summary.

II. FURTHER DEVELOPMENT OF THE STUDY OF SPONTANEOUS FISSION

During the time which has elapsed since the discovery of spontaneous fission of uranium, investigations of 35 other nuclei have been made. For the majority of them, one has determined sufficiently accurately the values of the half-lives for spontaneous fission, while for some of them it has been possible only to give a lower limit for this quantity (cf. Petrzhak's survey[22]).

The experimental techniques used for measurements of periods of spontaneous fission of these nuclei were varied. In those cases where one investigated a nucleus with a relatively low α activity, one used either multi-layer ionization chambers or large proportional counters (30 – 40 cm diameter, 2 – 3 meters length[18]). For the study of spontaneous fission of nuclei with a high α activity, for example Am^{241}, they used fast gaseous scintillators,[23] which eliminated the danger of imitation of the "fission" pulses by pulses from pile-up of α particles.

The measured values of the half-lives are included in a wide interval: from a few seconds (102^{254}) to 10^{29} sec (Th^{232}).

As the experimental data have come forth, attempts have been made to systematize the information concerning the periods for spontaneous fission. The first steps in this direction were made in 1952 when it was established[24,25] that for the even-even nuclei studied up to that time, the spontaneous fission periods decreased approximately exponentially with increase of the parameter Z^2/A.

However, the appearance of new data showed that this dependence is only approximately true.

Further accumulation of experimental information led to the appearance of a more complicated dependence for the lifetime of nuclei with respect to spontaneous fission: a series of experiments gave a dependence of T_{sp} on A, N (neutron number),[26-29,32,33] $T_{sp}/T_\alpha = f(Z^2/A)$ (cf. references 30 and 31), etc. In Figs. 6 and 7 we show the dependence of T_{sp} on Z^2/A and on N for the even-even isotopes. One sees that the parameter Z^2/A does not uniquely determine the probability of spontaneous fission. For isotopes of a given element the probability of fission depends essentially on the mass number and has a "resonance" character. In Fig. 7 the crosses show T_{sp} for nuclei with odd Z and N. The probability of spontaneous fission of odd isotopes is much lower than for the neighboring even-even ones. From the most reliable data it follows that the reduction factor for the spontaneous fission of odd nuclei compared to even ones is $\sim 10^5$.

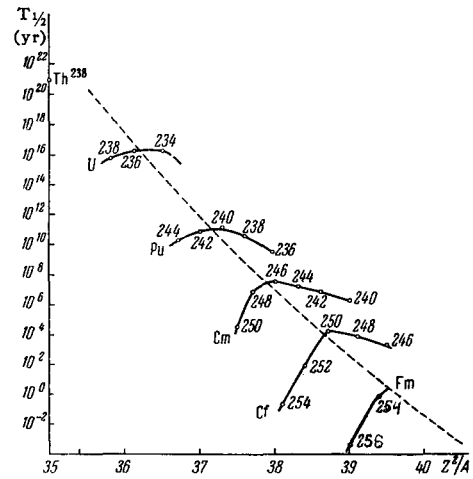

FIG. 6. Dependence of spontaneous fission half-life on Z^2/A (for even-even nuclei).

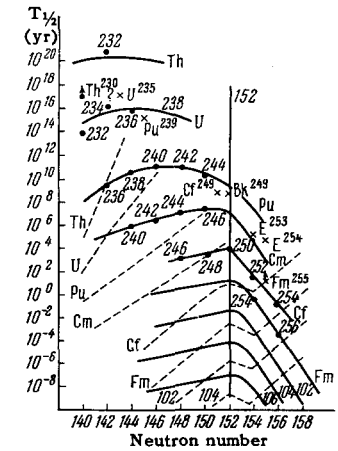

FIG. 7. Dependence of spontaneous fission half-life on neutron number. Dashed lines refer to α decay.

Possible theoretical explanations of this fact will be given in the section concerning the theory of spontaneous fission.

The first studies of the distribution of kinetic energy, ranges, and masses of fragments, using ionization chambers and the time of flight method, showed a marked asymmetry in the mass distribution of fragments.[34-39] A similar distribution was also obtained in the study of the yields of fission products by radiochemical methods.

In Fig. 8 we show experimental values[40-45] of the yields of fragments from the spontaneous fission of U^{238}, Cm^{242}, and Cf^{252}. For comparison, we give on the same figure a curve of the yield of fragments from the irradiation of Pu^{239} by slow neutrons. All of the curves have a similar character; namely they consist for the most part of two groups of fragments.

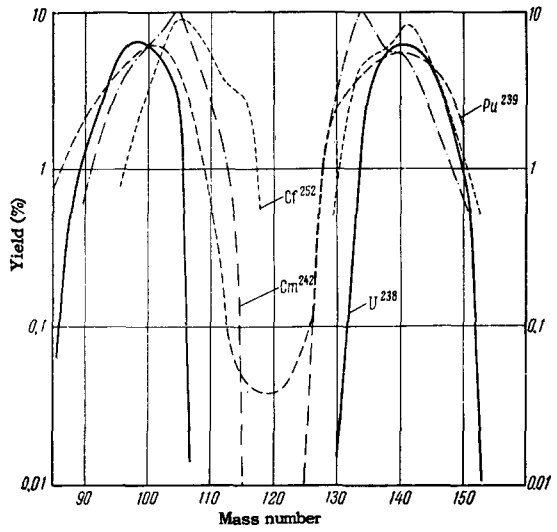

FIG. 8. Mass distribution of fragments from spontaneous fission of U^{238}, Cm^{242}, Cf^{252} and induced fission (by slow neutrons) of Pu^{239}.

The maximum yield for the light group is at A = 100, for the heavy group — at A = 140.

For the mass values 105 and 134 and their neighbors one observes an anomalous increase in fragment yield (the fine structure). This phenomenon is due to the increased probability of formation of heavy fragments with a filled shell at N = 82. It is obvious that the presence of a fine structure peak in the light group indicates the selective possibility of formation of fragments of the heavy group with a closed shell.

A certain difference between the curves for yield of fission fragments from spontaneous and induced fission occurs in the region of equal masses of fission fragments. This difference consists in the fact that the frequency of formation of fission fragments with approximately the same mass is considerably lower in the case of spontaneous fission.

The energy liberated in fission is distributed in kinetic energy and energy of excitation of the fragments. Bohr and Wheeler[63] already pointed out that the shape of the fragments immediately after their formation does not correspond to the equilibrium shape and that the energy of deformation after break-up is changed into energy of excitation of the fragments. The energy of excitation of the fragment is used in the emission of "prompt" neutrons and γ rays. The neutrons emitted in spontaneous fission were first observed by Fermi and collaborators in the construction of a uranium reactor.

Later on numerous investigations were made for the measurement of the average number of neutrons ($\bar{\nu}$) accompanying spontaneous fission. In Table II we give the known values of these numbers at the present time.[46-57]

Table II

Isotope	Average number of neutrons per fission	Isotope	Average number of neutrons per fission
U^{238}	2.4±0.2	Cm^{242}	2.65±0.09
Pu^{236}	2.3±0.19	Cm^{244}	2.84±0.09
Pu^{238}	2.33±0.08	Cf^{252}	3.82±0.12
Pu^{242}	2.18±0.019	Fm^{254}	4.05±0.19

The increase of $\bar{\nu}$ with increasing Z should be ascribed to a rise in energy of excitation of the fragments associated with an increase of total energy of the fissioning nucleus.

It is important to point out that in studying the dependence on Z of the average number of neutrons emitted in fission, spontaneous fission gives much more information than induced fission, since the range of elements which are included in the investigation of spontaneous fission is much greater.

Figure 9 gives the results of measurements of neutron number as a function of the ratio of fragment masses for the spontaneous fission of Cf^{252} and the fission of U^{235} by neutrons. For a mass ratio of 1.20 — 1.25 the main fraction of the neutrons is emitted

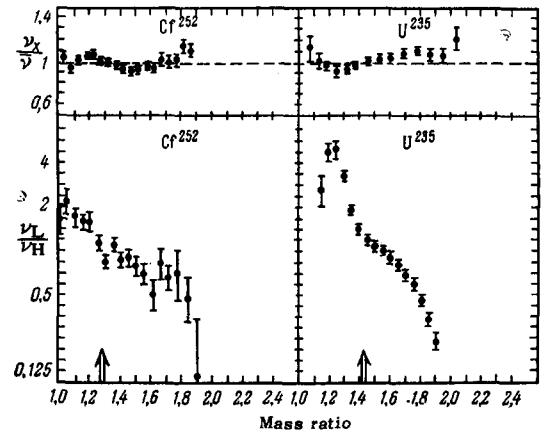

FIG. 9. Number of neutrons accompanying spontaneous fission of Cf^{252} and induced fission (by slow neutrons) of U^{235}, as a function of the ratio of masses of the fragments. ν_L, ν_H — number of neutrons emitted respectively from the light and heavy fragments, ν_x — total number of neutrons.

by the light fragment, while for greater asymmetry the main fraction comes from the heavy fragment. The total number of neutrons emitted by the pair of fragments depends only slightly on the mass ratio.

The study of neutron yield for a definite type of fission makes it possible to obtain information concerning the distribution of excitation energy over individual fragments and the degree of their deformation at the moment of formation. This is important for explaining details of the mechanism of fission.

The gamma radiation accompanying spontaneous fission was investigated[58] for the decay of Cf^{252}. It was shown that in each fission act there appear on the average 10 photons which take away a total energy equal to ~ 8 Mev. The energy spectrum of the γ quanta is concentrated mainly in the range up to 1 Mev, although a certain fraction of the radiation is in quanta with higher energy. The overall appearance of the γ-ray spectrum[59-61] is similar to that which was obtained in the study of the induced fission of U^{235}.

Numerous investigations with various nuclei carried out over the 20 years since the discovery of spontaneous fission have made it possible to establish various regularities for this phenomenon and to accumulate a rich store of factual material which has been briefly summarized above. However, despite the mass of experimental data one has still not succeeded in producing a quantitative theory of the phenomenon.

III. THEORETICAL PROBLEMS

The first discussions of the theory of nuclear fission were those of L. Meitner and O. Frisch.[2] They emphasized the analogy between the process of nuclear fission under the action of neutrons and the break-up of a liquid drop as a result of deformation produced by external action. In this connection they called attention to the fact that for the heaviest nuclei the electrostatic repulsion of the protons compensates to a large extent the short range attraction due to the nuclear forces. It was noted that the forces of attraction act like a surface tension which prevents an increase in surface. The theory of fission of nuclei based on the drop model was developed in papers by Ya. I. Frenkel'[62] and by N. Bohr and J. Wheeler.[63] If we assume that in the equilibrium state the nucleus is a drop of incompressible liquid of spherical shape, then an axially symmetric deviation from sphericity which is energetically more favorable from the point of view of the liquid drop model may be described by an equation for the surface of the nucleus of the form*

$$R(\vartheta) = R_0[1 + \sum_n \alpha_n P_n(\cos\vartheta)], \quad (1)$$

where R_0 is the radius of a sphere of the same volume.

The sum of surface energy and Coulomb energy of the nucleus as a function of the deformation parameters α_n (for small α_n) has the form

$$\varepsilon_{S+E} = U_0\left[1 + \frac{2}{5}\alpha_2^2 + \ldots + \frac{(n-1)(n+2)}{2(2n+1)}\alpha_n^2\right] + E_0\left[1 - \frac{1}{5}\alpha_2^2 - \ldots - \frac{5(n-1)}{(2n+1)^2}\alpha_n^2\right], \quad (2)$$

where $U_0 = 4\pi(r_0A^{1/3})^2\sigma$ is the surface energy for the spherical shape, and $E_0 = \frac{3}{5}(Ze)^2/r_0A^{1/3}$ is the energy of a uniformly charged sphere. Starting from these expressions one can show that, beginning from the value $(Z^2/A)_{cr} = 10\,\frac{4\pi}{3}r_0^3\sigma/e^2 \simeq 49$, the spherical shape becomes unstable with respect to the quadrupole deformations which are the principal ones (cf. the last footnote) Note that heavy nuclei are not spherical and have a certain equilibrium deformation ($\alpha_2 \sim 0.26 - 0.27$). In 1939 there were no data of this sort, so that in reference 63 they used the picture of a spherical nucleus. From analogous arguments applied to sufficiently large deformations they obtained a formula for the change in energy of a nucleus as a function of the parameter β (which we shall use from now on to denote the parameter α_2) (Fig. 10):

$$\frac{\varepsilon(x,\beta)}{U_0} = 2.178(1-x)y^2 - 4.09(1-0.645x)y^3 + 18.64(1-0.894x)y^4 - 13.33y^5, \quad (3)$$

where $x = \dfrac{Z^2/A}{(Z^2/A)_{cr}}$, $y = \frac{3}{7}\beta$. The maximum energy is reached at a value of $y = 1 - x$. For the energy of the fission threshold we find from (3) the expression

$$\frac{\varepsilon_{max}}{U_0} = 0.728(1-x)^3 + 0.661(1-x)^4 + 3.330(1-x)^5. \quad (4)$$

The corresponding critical shapes for unstable equilibrium[64] are shown in Fig. 11.

The results described above led the authors[63] to predict the possibility of spontaneous fission of nuclei. The point is that for heavy elements, $x \gtrsim 0.73$, the energy of the fission threshold amounts to a few Mev, so that there is a finite probability of spontaneous fission as a result of tunneling through the potential barrier. According to the general rules, the probability for such tunneling W is equal to

$$W \sim \omega_f \exp\left\{-\frac{2}{\hbar}\left|\int_{\beta_0}^{\beta_f} P_\beta\, d\beta\right|\right\}, \quad (5)$$

where ω_f is the frequency of quadrupole oscillation, β_0 is the value of the parameter β at the equilibrium position, β_f is the value at the point of "emergence under the barrier," P_β is the generalized momentum corresponding to the coordinate β: $P_\beta = \sqrt{2M_\beta[E - \epsilon(x,\beta)]}$, where M_β is the corresponding generalized mass, E is the total energy of the nucleus which, in the case in which we are interested of spontaneous fission, may

*The requirement of constancy of volume and unchanged position of the center of mass leads to extremely small values for the coefficients α_0 and α_1. The energetically most favorable deformations also correspond to small values of α_n for $n > 2$. Finally, n is limited above by the finite number of nucleons in the nucleus.

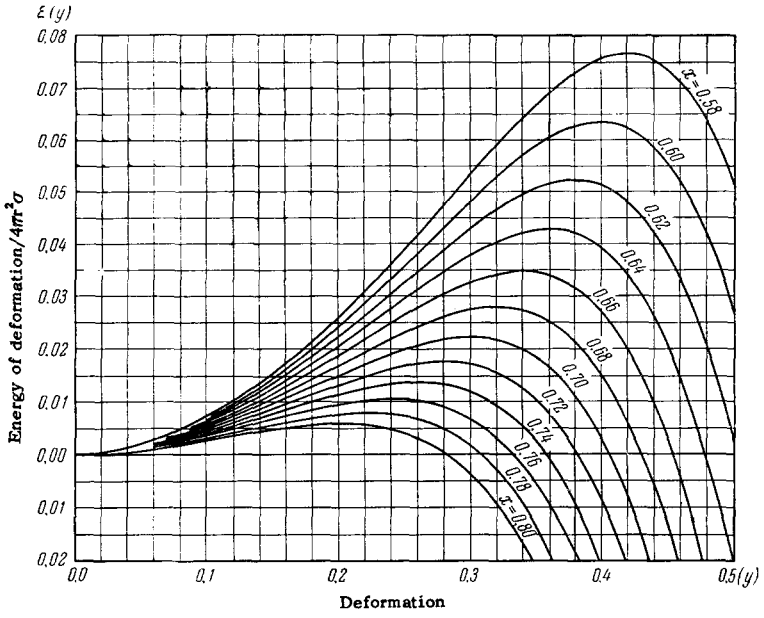

FIG. 10. Shape of potential curve for different x.

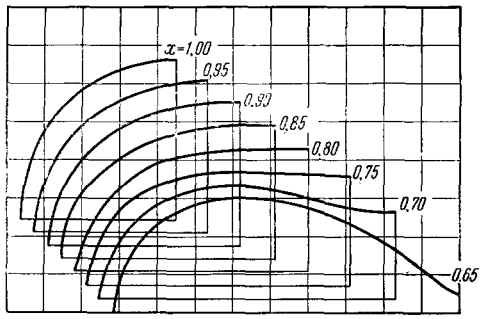

FIG. 11. Critical shapes of unstable equilibrium.

be set equal to zero. Thus formula (5) can be rewritten as

$$W \sim \omega_j \exp\left\{ -\frac{2}{\hbar}\int_{\beta_0}^{\beta_f} \sqrt{2M_\beta \varepsilon(x,\beta)}\, d\beta \right\}. \qquad (5a)$$

In reference 63 it was assumed, in analogy to α decay that

$$M_\beta = \sum^{A} m\left(\frac{dr_i}{d\beta}\right)^2. \qquad (6)$$

Making a qualitative estimate of the value of $(dr_i/d\beta)^2$, the authors found for U^{238} a lifetime $\tau = W^{-1} \sim 10^{22}$ yr. It should be pointed out that formula (6) is by no means a consistent treatment of the hydrodynamic theory of fission, since it clearly assumes the presence in the nucleus of individual particles. In this connection V. Berestetskii and A. Migdal[65] carried out a more consistent hydrodynamical calculation which, however, gave $\tau \sim 10^3$ sec, which is many orders of magnitude smaller than the experimental value. Thus there appeared the first difficulty in the path of an explanation of the phenomenon of spontaneous fission from the point of view of the liquid drop model: the theory gave too small an effective mass corresponding to the coordinate. This is not surprising since the nucleus is not actually a drop of incompressible ideal fluid, as we know at present from numerous experiments. Somewhat more strange is the fact that the single particle oscillator model also gives a small value for the effective mass. In this connection, there is still no further clarification of the question why, in spontaneously fissioning elements, there appear such large lifetimes.

If we take the value of the effective mass from experiment, formulas (3) and (5a) lead to a linear dependence of $\ln \tau$ on Z^2/A, which only very roughly describes the observed variation of τ as we move from nucleus to nucleus. The observed experimental dependence on Z and A of the lifetime of nuclei with respect to spontaneous fission is much more complicated than that obtained from the drop model (cf. Figs. 6 and 7). In contradiction to the predictions of the hydrodynamic model one observes an increase in probability of spontaneous fission for heavy even-even isotopes of elements; in addition, in the picture presented above of the phenomenon no distinction was made between even-even and odd nuclei, but we know that odd nuclei have anomalously large lifetimes.

To explain these features of the dependence of the probability of spontaneous fission on Z and A one has invoked the picture of the single-particle structure of the nucleus,[55-58] which has recently achieved a deserved recognition. These ideas were developed in more detail and fruitfully by Johansson.[69] He made the assumption that nucleons which are distributed in pairs (with antiparallel spins) on single-particle levels (cf. the Nilsson scheme[70]) can easily make a transition to free levels which cross them in the process of deformation (Figs. 12 and 13), if this is energetically favorable. Such a possibility arises because of the fact that the total spin of the nucleons is always equal to zero, and their parity is positive, so that such a transition is not forbidden. This in turn leads to the result that an even-

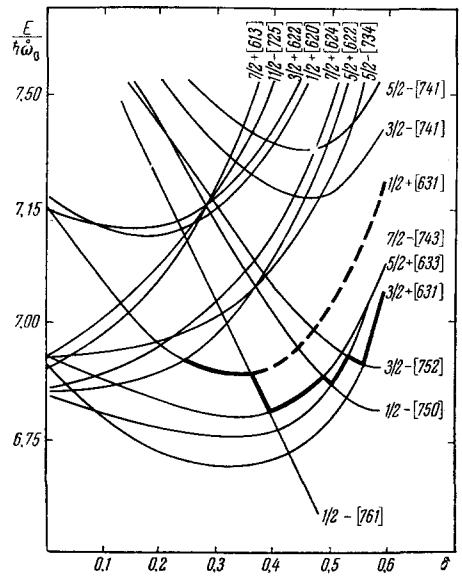

FIG. 12. Dependence of energy on parameter $\delta = 0.95\,\beta$ for the 146-th neutron (thick solid line) and the 145-th neutron, when it is the only one present in the level (thick dashed line).

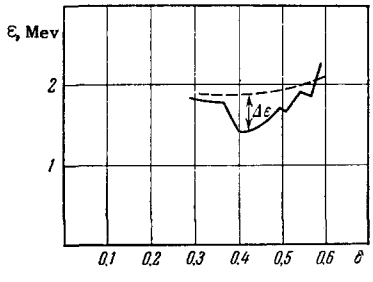

FIG. 13. Dependence of energy of 146-th neutron on deformation parameter δ. The dashed line shows the dependence calculated from the hydrodynamic model.

even nucleus in the process of changing shape is almost always in its lowest energy state. A completely different picture occurs in the case of odd nuclei. Since levels of the same symmetry do not cross[71] there is a strong selection rule forbidding the transition of the "last" odd nucleon to any other level. For this reason, if the level of the last odd nucleus does not go below in the process of increase of deformation, then there is formed between it and the even core a band of free levels, i.e., the nucleus is not in its lowest energy state.

These arguments led the author of reference 69 to the following results for the lifetime τ of a nucleus with respect to spontaneous fission:

$$\ln\tau = \ln\tau_{\text{hydr}} + \Delta\ln\tau,$$

$$\Delta\ln\tau = K\int_{\beta_0}^{\beta_f}\sqrt{\varepsilon_{\text{hydr}}(\beta)+\Delta\varepsilon(\beta)}-\sqrt{\varepsilon_{\text{hydr}}(\beta)}\cong\frac{K}{2}\times\int_{\beta_0}^{\beta_f}\frac{\Delta\varepsilon(\beta)}{\sqrt{\varepsilon_{\text{hydr}}(\beta)}}, \qquad (7)$$

where τ_{hydr} and $\epsilon_{\text{hydr}}(\beta)$ are the lifetime and barrier height calculated for the drop model, $\Delta\epsilon(\beta)$ is the correction to $\epsilon_{\text{hydr}}(\beta)$ (cf. Fig. 13). Here it is assumed that $|\Delta\epsilon| \ll \epsilon$ and $K = (2/\hbar)\sqrt{2M_\beta} = \text{const}$.

Thus, assuming a specific arrangement of levels, the author found the corrections ($\Delta\ln\tau$) to the lifetimes of nuclei calculated from the drop model. Using these corrections, he succeeded in getting the experimental points for even-even nuclei close to the theoretical line (cf. Figs. 6 and 14).

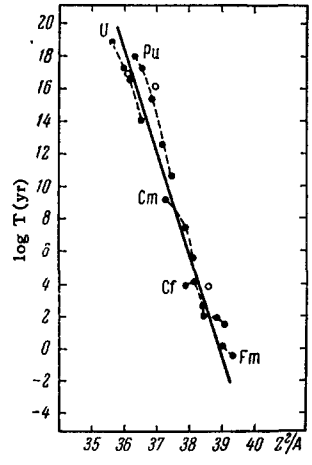

FIG. 14. Dependence of half-life for spontaneous fission on Z^2/A. The corrections shown in Fig. 6 have been made to the experimental points. The corrections are calculated in reference 69 using formula (7), which is valid on the assumption that $|\Delta\varepsilon| \ll \varepsilon$. The value of K is ≈ 230.

However, such an approach to the problem of lifetimes of even-even nuclei does not seem to us to be entirely valid. It is known at present that the presence of "pairing" at the Fermi surface leads to a smearing out of the surface. For this reason it seems to us to be meaningless to assume that near the Fermi surface the nucleons occupy definite energy levels; moreover, the Nilsson scheme which was used in reference 69 is very approximate and therefore it should be applied only to phenomena associated with single-particle properties of the nucleus (for example, to single-particle resonances, the effects of the last nucleon in odd nuclei, etc.).

The use of this scheme for describing phenomena associated with the behavior of a large number of particles can lead to sizable errors, since in this case one sums the uncertainties in the determination of the behavior of the individual levels. It should also be pointed out that the behavior of the levels when the deformation parameter is changed is to a considerable extent random. Therefore, from the picture developed by Johansson it turns out that the lifetime should fluctuate from one isotope to another. The experimental data are not in agreement with this conclusion: the probability of spontaneous fission varies smoothly when one changes the mass number of the isotope of a given element and shows a very definite regularity.

Let us now consider, following Johansson, the spontaneous fission of odd nuclei. In Fig. 12 as an example, we show the change in energy of the 145-th neutron when the deformation is increased. The energy of the odd nucleon increases much more rapidly than follows from a purely hydrodynamical treatment. This leads to an effective increase in the potential barrier for fission, and consequently to an increase in lifetime.

This qualitative explanation of the reason for the reduction in probability of fission in odd nuclei is extremely convincing. The "retardation coefficient" κ for an odd isotope with respect to its neighbor, for example a lighter even one, can be written, as one sees from formula (7), in the form

$$\ln \varkappa = \Delta \ln \tau_{odd} \cong \int_{\beta_0}^{\beta_f} \frac{\Delta \varepsilon_{odd}}{\sqrt{\varepsilon_{hydr}}} d\beta, \qquad (8)$$

where $\Delta\epsilon_{odd}$ is now the difference between the energy of the last odd nucleon as determined by the corresponding level in the single-particle model and the energy which it should have according to the drop model. (The latter is determined simply as the height of the barrier according to the drop model divided by the number of nucleons. We recall that the energy is measured from the energy for the equilibrium shape of the nucleus.)

Relation (8) leads to a whole series of conclusions, which can be the object of an experimental test. First of all, it is possible to have spontaneously fissioning isomers such that nuclei which have a higher energy live longer. For this it is sufficient that the level of the last nucleon in the nucleus, though it has higher energy, go up faster than the level on which the same nucleon is located in the nucleus with lower energy. Secondly, there should be a definite relation between the retardation coefficients of odd-odd and the corresponding odd-even and even-odd nuclei: if an odd-even nucleus (Z+1, N) has a coefficient κ_1, while the nucleus (Z, N+1) has a coefficient κ_2, then the odd-odd nucleus (Z+1, N+1) should have the coefficient $\kappa \sim \kappa_1\kappa_2$. Here it is important that this relation does not depend on the specific behavior of the levels. At present, we know the coefficient only for one odd-odd nucleus Es^{254}. It is approximately 5×10^4 and is too small, according to these considerations, since the average coefficient for even-odd nuclei (or odd-even nuclei) is of the order of 10^5. However, the experimental material is too sparse to permit us to say that the theory is incorrect. It should be pointed out that the detection of spontaneous fission with a large coefficient ($10^8 - 10^9$, as one should expect for odd-odd nuclei) is extremely difficult because of the predominating competition from other types of decay whose rates are many orders of magnitude greater. This also explains the absence of data for odd-odd nuclei. Thirdly, it is possible that with increasing x, i.e., in the region of the far transuranic elements, differences in lifetime between even-even and neighboring odd isotopes will decrease. In order to understand this we write the approximate expression for the logarithm of the retardation coefficient

$$\ln \varkappa = \frac{K}{2} \int_{\beta_0}^{f} \frac{\Delta \varepsilon}{\sqrt{\varepsilon}} d\beta \cong \frac{\overline{\Delta \varepsilon}}{\sqrt{\varepsilon}} \Delta\beta \frac{K}{2}. \qquad (7a)$$

With increasing x the size of the sub-barrier region $\Delta\beta = \beta_f - \beta_0$ decreases, $\overline{\Delta\epsilon}$ at least does not increase, while $\sqrt{\epsilon}$ in all probability decreases less rapidly than $\Delta\beta$. From this it follows that the value of κ should drop.

Table III

Isotope	Retardation coefficient	Isotope	Retardation coefficient
Pu^{239}	10^5	Es^{254}	$5 \cdot 10^4$
Am^{241}	$5 \cdot 10^5$	Es^{253}	10^4
Bk^{249}	$5 \cdot 10^4$	Fm^{255}	$5 \cdot 10^2$
Cf^{249}	10^5		

At the present time the theory of spontaneous fission still has a semi-qualitative character and is far from completion. Nevertheless it is in a position to make certain completely definite statements concerning experimental investigations which are needed for further development and tests.

IV. SPONTANEOUS FISSION AND SOME PROBLEMS OF ASTROPHYSICS AND GEOPHYSICS

Eruptions of novae and supernovae are some of the most interesting and important phenomena occurring in the universe. In particular, they serve as a source of cosmic rays and also are one of the processes in which chemical elements are formed. It is possible that the phenomenon of spontaneous fission plays an essential role in these processes. This applies in particular[79] to supernovae of type I. A characteristic feature of these stars is that after the initial period, which lasts 50 — 100 days, their luminosity falls exponentially as $J \sim \exp(-t/\tau)$, where $\tau \simeq 55$ days (Fig. 15). In

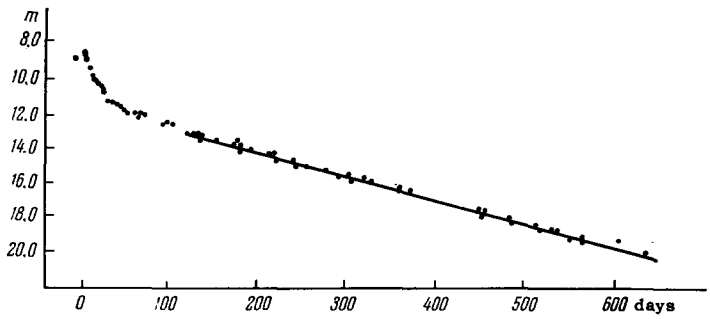

FIG. 15. Time dependence of the stellar magnitude of a supernova. The linear dependence in these coordinates corresponds to an exponential decrease of luminosity with time.

this connection it was suggested in reference 79 that after the initial period energy is liberated through spontaneous fission of Cf^{254}, which is formed from Fe^{56} by repeated capture of neutrons accompanied by the β decay of intermediate nuclei. In this way altogether 198 neutrons are captured. (It is assumed that the elements of the iron group are synthesized in the interior of the supernovae.[80]) The high neutron density resulting from thermonuclear reactions occurring in the initial stage makes such a rapid capture possible. Spontaneous fission of Cf^{254}, and no other nearby isotope,

is important because of the fact that its lifetime $\tau \sim 55$ days is several orders of magnitude lower than the lifetime of neighboring isotopes. As for the other far transuranic elements, they should decay in the first few days after the beginning of the flare. Burbidge et al.[79] have shown that to explain the luminosity of supernovae during the second period in this way requires entirely reasonable amounts of Cf^{254}.

Recently there has been proposed another mechanism for explaining the same phenomenon[81] in which it is assumed that the energy liberated during the second period is taken from the β decay of the isotope Fe^{59}, which has a lifetime $\tau \sim 45$ days and is formed as a result of capture of three neutrons by Fe^{56}. Theoretical treatment of these mechanisms explaining the fall-off in time of the luminosity of supernovae is extremely complicated since there are insufficient data at present. It seems to us that there is an experimental possibility for making a choice between these assumptions. The point is that the spontaneous fission of Cf^{254} and the β decay of Fe^{59} are accompanied by the emission of γ quanta whose spectra have characteristic shapes. A study of the γ radiation accompanying flashes of supernovae undoubtedly will give additional material in favor of one or the other of these hypotheses.

In addition to problems of astrophysics, one can point out various geophysical questions in which spontaneous fission of heavy elements plays an important role. Spontaneous fission of uranium has a noticeable influence on the distribution of various isotopes in the earth's crust, leading to an accumulation of isotopes of krypton, xenon, strontium, rubidium and cesium in uranium-bearing minerals. Of most interest is the isotope Xe^{136}. It has a small abundance in nature and at the same time is one of the most probable products of uranium fission. A measurement of the content of this isotope in uranium minerals is a good method for determining the origin of such minerals.

The spontaneous fission of transuranic elements can also affect the distribution of isotopes. It is not excluded that they existed at some time in approximately the same amounts as uranium and thorium. However, for the heavy isotopes of plutonium and curium — Pu^{244} and Cm^{248} — the probability of spontaneous fission is relatively high; while for the isotopes of californium and fermium — Cf^{254} and Fm^{256} — the spontaneous fission is the main type of radioactive decay. Therefore an effect associated with the spontaneous fission of these isotopes might be quite large.

In particular, one might assume that the distribution of isotopes of xenon in nature has been strongly influenced by the spontaneous fission of transuranic elements. The isotope Xe^{129} which has a wide distribution is the only exception to the rule according to which for three successive isotopes of a given element (two isotopes with even A and one with odd A) the abundance of the isotope with odd A is always smaller than the sum of the abundances of the even isotopes.

The formation of Xe^{129} cannot be attributed to spontaneous fission of uranium since in the spontaneous fission of U^{238} the yield of fragments with mass 129 is small. At the same time the study of the yield of fragments from spontaneous fission of Cm^{242} and Cf^{252} has shown that for these isotopes the probability of formation of fragments with mass 129 is considerably greater.[72,73]

Attempts have been made in various experiments to observe transuranic elements in thorium and uranium ores. However, one has succeeded in detecting only Pu^{239} in these various minerals. The amount of Pu^{239} is on the average $\sim 10^{-11}$ compared to the content of uranium and is explained entirely[74] by the effect of capture of neutrons by U^{239} nuclei.

The assumption has also been made that spontaneous fission of transuranic elements manifests itself in pleochroic rings in mica.[75] Pleochroic rings occur under the action of α particles when they impinge in mica on inclusions of uranium or thorium. Each group of α particles forms a ring with a diameter corresponding to the range of these particles. In certain samples rings have been detected corresponding to the ranges of fission fragments. They could not be explained quantitatively by spontaneous fission of uranium, and the assumption was made that these rings are related to the fission of transuranic elements. However, recently it has been found that for all the long-lived isotopes of the transuranic elements the probability for α decay is greater than for spontaneous fission.[76] Nevertheless in all samples of mica rings are found corresponding only to α particles from the known radioactive series. Therefore, it seems to us most probable that the fragments arose from induced fission of U^{235} by neutrons, which was 15 times more prevalent 3×10^9 years ago. The neutrons could be emitted in (α, n) reactions if in the ores from which the mica was taken there was contained a large amount of light elements — Li, Be.* Unfortunately, the available experimental material is too sparse and one cannot draw any definite conclusions on this basis. One should carry out more careful investigation of pleochroic rings in mica from older deposits in order to obtain quantitative data concerning the influence of these various effects.

It seems that the earth's crust should not contain any noticeable amounts of natural transuranic elements. The longest lived of the known transuranic elements are Pu^{244} ($T = 7 \times 10^7$ yr) and Cm^{247} ($T > 4 \times 10^7$ yr).[76]

*Another origin for fission fragments in mica is not excluded. It is possible that for one of the transuranic elements there is a spontaneously fissioning isomer whose lifetime exceeds the period of stabilization of the earth's crust. The decay of such an isomer would lead to the appearance of "fragmentary" pleochronic rings without a change in structure of the rings associated with α particles.

According to the systematics of radioactive nuclei, Cm^{247} practically does not undergo spontaneous fission, so that we shall not discuss methods for detecting it.* Since the time of existence of the earth's crust is not less than 3×10^9 years, there should remain no more than a fraction of 10^{-13} of the initial amount of Pu^{244}. If at the moment of formation of the elements, Pu^{244} was present to the same extent as uranium, then in 1 ton of uranium there should be $\sim 10^{-7}$ grams of Pu^{244}. Such an amount of isotope having a low specific activity would be very difficult to detect.

One could, however, try to detect products of spontaneous fission of Pu^{244}. The period of stabilization of the earth's core is estimated to be within the limits $4 \times 10^8 - 1.5 \times 10^9$ years.[77] If this time interval is close to the lower of these limits, then at the moment of formation of the minerals a quite large amount of Pu^{244} should have been retained. For this isotope the probability of spontaneous fission is 3×10^{-3} compared to α decay, i.e., it is 3,000 times greater than for U^{238}. Therefore the yield of fission products could be quite large. The most convenient for observation are the isotopes of xenon. The isotopic composition of xenon from fission of Pu^{244} should be the same as from spontaneous fission of U^{238}, since the Z/A ratio is almost the same for both. This fact essentially simplifies the separation of the effects associated with induced fission of U^{235}, since in the induced fission of U^{235} and the spontaneous fission of U^{238} the relative yields of the isotopes Xe^{136} and Xe^{129} are different by more than a factor of 20.[78]

It would be useful to make a search for xenon in older minerals containing rare-earth elements analogous to plutonium in their chemical properties, and with a small uranium impurity. The fundamental technical difficulties for such work are associated with the background occurring from spontaneous fission of uranium. Uranium has a greater capacity for migration, and therefore is present in small amounts in all minerals. However, the uranium which entered into the mineral, not at the time of formation of the minerals, but in the process of a long period of migration, is absorbed mainly on the surfaces of crystals. By dissolving away the outer layer of crystals to a depth equal to the range of the fragments, one could, presumably, considerably reduce the background due to uranium fission.

Thus it seems to us that the study of effects associated with spontaneous fission of heavy elements, after carrying out these appropriate experiments, might give an answer to various questions concerning the physics of the earth.

*To detect tracks of Cm^{247} in minerals one could make a neutron activation analysis. As a result of radiative capture of thermal neutrons, Cm^{247} is converted into Cm^{248} which, in 10% of the cases, decays by spontaneous fission. By recording the spontaneous fission of Cm^{248} one could reliably establish the presence of $\sim 10^{-12}$ Cm^{247} in the irradiated samples.

V. TRANSURANIC ELEMENTS. SPONTANEOUS FISSION AND THE LIMITS TO THE SYSTEM OF ELEMENTS

Spontaneous fission is energetically possible almost from the middle of the periodic table of elements, but the probability for this process is extremely small and it is practically negligible for elements lighter than uranium. As Z increases, the instability of nuclei with respect to spontaneous fission increases, and it begins to show itself in the background of other methods of decay, and then finally becomes one of the main types of decay. This regularity is shown by Fig. 16,

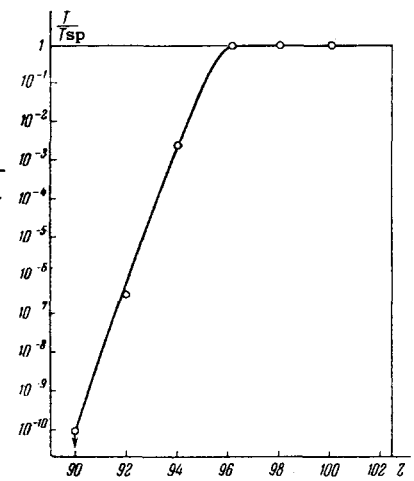

FIG. 16. Relative probability of spontaneous fission for the most easily fissionable even-even nuclei.

in which we show as a function of nuclear charge a quantity characterizing the relative probability of spontaneous fission T/T_{sp} (where T is the lifetime of the nucleus, T_{sp} is the partial lifetime for spontaneous fission). In constructing the graph we have used data for the most easily fissionable even-even nuclei.

Whatever the properties of the unknown transfermic elements ($Z > 100$), how fast will the rate of spontaneous fission increase with increasing charge? Since spontaneous fission is the fundamental process determining the stability of heavy nuclei, the problem of the rate of spontaneous fission of transfermic elements is closely associated with the question of the limit for the system of elements.

As already pointed out in section III, at present there is no satisfactory quantitative theory of spontaneous fission. It is therefore obvious that we have no purely theoretical approach to the problem of predicting properties of unknown heavy elements. The only possibility which here remains is the extrapolation of the experimental dependences and the use of semi-empirical relations. In the analysis of the dependence of observed half-lives for spontaneous fission on Z^2/A (cf. Fig. 6) or on number of neutrons (cf. Fig. 7), one sees that it is not possible to draw a smooth curve through the experimental points. There are essentially a whole series of "resonance" curves each of which corresponds to a definite element. The half-lives of

the known elements go through a maximum as A is varied. To each element there corresponds a certain value of $(Z^2/A)_0$ determining the most stable of the isotopes with respect to fission (which lies in the β stability region[27,28]). The smooth curve formed by the average of the lifetimes with respect to fission as a function of Z^2/A passes through these isotopes. In Fig. 17 we show the values of $(Z^2/A)_0$ as a function of atomic number of the element.

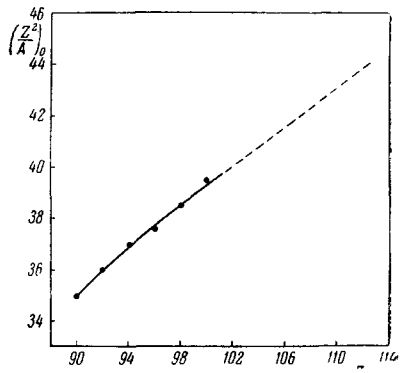

FIG. 17. Value of Z^2/A for the even-even nuclei which are most stable with respect to spontaneous fission.

To predict the probability of fission of various isotopes of unknown elements one must know how to draw the corresponding curve $T_{sp} = f(Z, A)$, i.e., to "guess" its shape and the location of the maximum. The further the element we are interested in is from the region of known nuclei, the greater the arbitrariness in shape and location of such a curve. Therefore predictions of the properties of individual isotopes can reasonably be made only for elements which directly neighbor on known elements. There is less arbitrariness in estimating the properties of the isotopes which are most stable against spontaneous fission, i.e., those isotopes which correspond to the maximum of the curve $T_{sp} = f(Z, A)$. Making such estimates, we extrapolate the dependence drawn in Fig. 17 and find for the element of interest to us the value of $(Z/A)_0$ and the approximate value of A_0 — the mass number of the most stable isotope. From the dashed curve of Fig. 6 extended into the region of unknown nuclei, one determines the corresponding lifetime. The results of such estimates for even-even isotopes are shown on Fig. 18. Since the precision of such predictions is extremely doubtful, we have not been willing to draw a line on this graph, but have drawn a band with a spread in values of the decay period of one order of magnitude. It is known that for the best studied nuclei with an odd number of nucleons the spontaneous fission rate is lower by $10^{5\pm1}$ than for neighboring even-even isotopes. In accordance with this the "band" of values for half-lives for spontaneous fission of the most "stable" odd isotopes is shifted toward larger times. In the figure we also give estimates of the half-lives for α decay of the isotopes which are most stable with respect to spontaneous fission. From an analysis of Fig. 18 we come to the following conclusions:

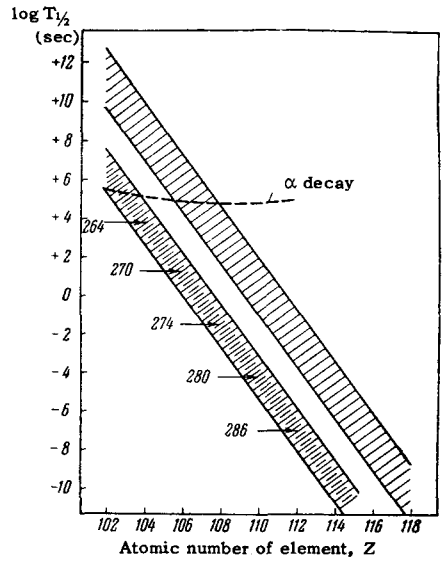

FIG. 18. Suggested values for half-lives with respect to spontaneous fission and α decay for the most stable isotopes of unknown elements (the double-dashed band refers to even-even nuclei, the simple dashed band to odd nuclei).

1. The lifetime of the most stable elements with $Z > 104$ will be determined by spontaneous fission, and not by α decay.

2. The instability of the elements increases rapidly with increasing nuclear charge. There is a very small probability of finding nuclei (even with an odd number of nucleons) with a charge exceeding 118, whose lifetime is greater than 10^{-8} sec.

3. If we take a value of 10^{-8} sec for the critical lifetime of an element, then roughly we may say that the system of elements is completed somewhere near $Z = 120$.

Unquestionably nuclei which live less than 10^{-8} sec are still of interest to investigators. Here it is possible to use methods of investigation which are special and specific for the super-heavy nuclei. S. M. Polikanov[82] has made the suggestion that in the decay of compound nuclei with $Z = 130 - 150$, which could be obtained in reactions using multi-charged ions, conversion transitions should occur with fair probability. The appearance of typical conversion electrons could be used as an indicator of the formation of a compound nucleus with a definite charge. Since for such heavy nuclei the K-shell electrons will be practically inside the nucleus, the study of the internal conversion process may become a method for investigating nuclear structure. This is still an assumption which requires detailed theoretical computation.

Another point of view concerning the problem of the limit of the system of elements was developed by J. Wheeler.[68] Wheeler estimated the lifetimes of superheavy nuclei with respect to spontaneous fission, α decay, and β^- decay, and came to the conclusion that it is possible for nuclei to exist with a charge greater than 140, and lifetimes exceeding 10^{-4} sec.

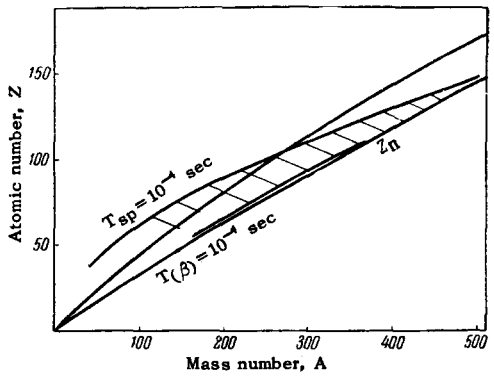

FIG. 19. Properties of superheavy nuclei, according to reference[88]. Inside the dashed region the lifetimes of the nuclei are greater than 10^{-4} sec. Z_n is the limit of neutron-stable nuclei.

In Fig. 19, taken from the paper of Werner and Wheeler,[88] we show the results of such estimates. These results are in essential disagreement with those given above, so that one must analyze the reason for the discrepancy. Since the stability of the far transfermic elements is determined by the spontaneous fission process, all differences in estimating the limits of "stable" nuclei are determined by the way one has computed the rate of spontaneous fission for the unknown elements. Wheeler also used the extrapolation of the dependence of T_{sp} on Z^2/A given in Fig. 6. However, he essentially assumed that the rate of spontaneous fission is uniquely determined by the value of Z^2/A, independent of the value of the nuclear charge. For a given Z he chose such a value of A that Z^2/A was sufficiently small, and from the smooth variation of T_{sp} with Z^2/A found a large value of T_{sp}. In such a calculation one does not take account of the "resonance" behavior of the dependence of the probability for spontaneous fission on Z^2/A. The estimates made in Wheeler's paper may be valid, if the dependence on A of the lifetime for spontaneous fission of a given element has the following form: the lifetime goes through a maximum with increasing A, then increases once more. This means that for isotopes of a given element there must exist some second region of stability with respect to spontaneous fission. Such an assumption has no basis and is highly improbable.

Returning to our estimates of the limit of the system of elements and the properties of heavy nuclei, we note that we have not considered the possibility of a different type of shell effect. The possibility is not excluded that, due to the filling of the nucleonic shells in the transfermic region of elements, one may find elements with increased stability. This undoubtedly would change the conclusions we have drawn.

Let us examine the methods for obtaining new elements.

a) Successive capture of neutrons. In using this method the initial material is U^{238}. As a result of radiative capture of a neutron, we form the β^--active isotope U^{239}. By two successive β^- decays this nucleus is converted to Pu^{239}, which in turn captures neutrons and leads to a chain of β^- decays increasing the charge of the nucleus. In Fig. 20 is shown the scheme for obtaining Fm^{254} from U^{238} by multiple capture of neutrons in a nuclear reactor. By this method, one has succeeded in synthesizing all the elements from Np to Fm. However, neither of the next two elements (Z = 101 and Z = 102) was obtained in this way. This is related to a difficulty which is characteristic for this method: all the β^- stable intermediate nuclei must live sufficiently long so that they can capture a neutron and be converted to a β^- active isotope. In synthesizing, for example, Mv^{259} by neutron capture in a reactor, the sequence of reactions is the following:

$$\ldots \mathrm{Fm}^{254}(n,\gamma)\mathrm{Fm}^{255}(n,\gamma)\mathrm{Fm}^{256}(n,\gamma)$$
$$\mathrm{Fm}^{257}(n,\gamma)\mathrm{Fm}^{258}(n,\gamma)\mathrm{Fm}^{259}$$
$$\downarrow\beta^-$$
$$\mathrm{Mv}^{259}.$$

The path to the β^--active isotope of fermium is through Fm^{256}, which fissions spontaneously with $T_{sp} \cong 160$ min, and Fm^{258} which probably fissions even faster. With the existing neutron densities in reactors, this time is too short for an effective formation of Fm^{259}, i.e., such a method for obtaining Mv^{259} is not feasible. These difficulties can be overcome to some extent by using neutron fluxes of such intensity that during a time interval which is significantly less than the life-

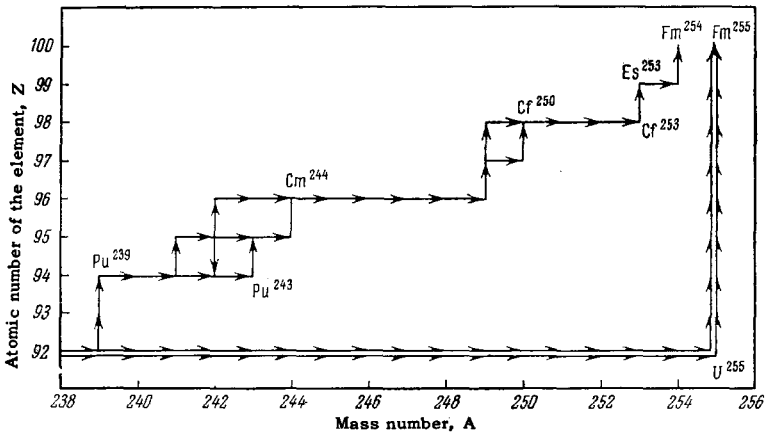

FIG. 20. Scheme of formation of transuranic elements by multiple neutron capture.

time of the intermediate nuclei a multiple capture of neutrons occurs. Such conditions are produced in nuclear or thermonuclear explosions. In Fig. 20 is shown the scheme for the formation of Fm^{255} by a powerful neutron irradiation of U^{238}. By such a method, as a result of a thermonuclear explosion, einsteinium and fermium (the 99-th and 100-th elements)[83] were first synthesized.

Further production of new elements ($Z > 102$) by using neutron fluxes accompanying nuclear explosions is probably not possible because of the fact that the short lifetimes of the elements require extremely rapid handling of the irradiated material. In all probability the method of multiple capture of neutrons has practically exhausted itself with respect to the question of directly obtaining new elements ($Z > 102$).

b) Reactions using charged particles. To obtain the transuranic elements from Np to Fm one has used in practically the same way the irradiation of U^{238} by neutrons and reactions produced by charged particles accelerated in suitable apparatus. It should be noted that of the eight transuranic elements from Np to Fm, four were first obtained by multiple radiative capture of neutrons and four in reactions using accelerated deutrons and α particles. The series of isotopes of these elements and the 102nd element were first obtained[84-87] using accelerated multiply-charged ions of C^{12}, N^{14}, and O^{16}. However, it should be emphasized that in reactions with charged particles, as a rule, the targets were materials obtained by irradiating uranium with neutrons. Thus the method described is essentially complex, unlike the preceding one. Such a combination of both methods will also be developed further in the future, and, as will be shown, makes it possible to overcome partially the difficulties arising in the use of charged particles in the synthesis of elements.

What are the fundamental advantages of this method compared with the method of multiple neutron capture, what are the prospects for its use in the future, and what difficulties does one encounter in using it?

In reactions produced by charged particles, the final nucleus has a charge Z_f, greater than the initial charge Z_i, by the amount of the charge of the bombarding particle Z. The new element is obtained directly from the initial one (Fig. 21), the properties of intermediate nuclei (in contrast to the preceding method) have no significance.

The total number of nuclei — reaction products — is determined by the integral flux of bombarding particles and does not depend on its intensity. The conditions of operation with beams of charged particles make it possible to analyze the reaction products right near the place where they are formed. This makes it possible to study the properties of new elements whose lifetimes are many orders of magnitude less than one second.

If we pose the problem of obtaining an element with atomic number Z_f, then we need a beam of nuclei with

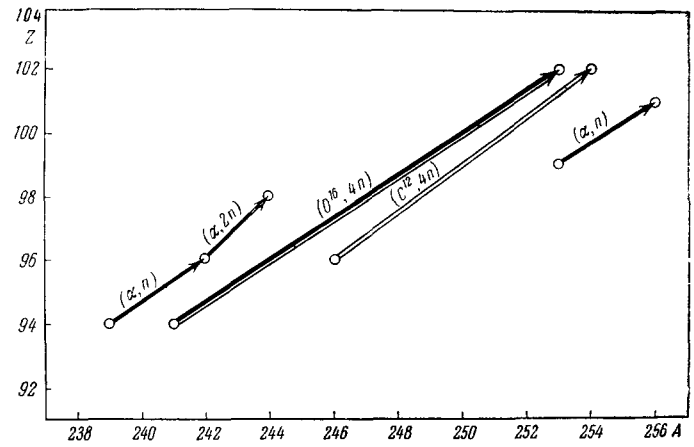

FIG. 21. Schematic picture of nuclear reactions produced by charged particles.

a charge at least equal to $Z = Z_f - Z_i$. When we are considering the synthesis of the elements from Np to Mv, it is possible to avoid the use of α particles, since the use of powerful reactors makes it possible to prepare targets of all the transuranic elements up to Es. However, the possibilities of preparing targets of even heavier elements is extremely limited, so that to obtain the 102nd element one already had to use bombarding particles with a charge greater than that of helium. We know that in the experiments for obtaining the 102nd element one used an irradiation of Cm^{246} by carbon:[86] $Cm^{246} + C^{12} \rightarrow 102^{254} + 4n$ and of Pu^{241} by oxygen:[87] $Pu^{241} + O^{16} \rightarrow 102^{253} + 4n$. The transition to synthesis of further transfermic elements requires that one use still heavier bombarding particles, and poses the problem of obtaining sufficiently intense beams of Ne, Al, P, and A of suitable energy. At the present time one has successfully obtained intense beams of carbon, nitrogen, and oxygen ions. One may hope that further development of the method of accelerating multi-charged ions will enrich the arsenal of experimental physicists with even heavier particles with the necessary intensity and the required energy.

The fundamental difficulty, in our opinion, which stands in the way of solving the problem of obtaining new elements by using heavy ions is related to the small value of the cross section for nuclear reactions and with a tendency for them to decrease when one goes to the synthesis of nuclei with high charge. The cross section for reactions produced by a given ion (C, O, Ne, etc.) with evaporation of x neutrons has the form

$$\sigma_{xn}(E) = \sigma_0(E) P_{xn}(E) \left(\frac{\overline{\Gamma_n}}{\Gamma}\right)^x,$$

$\sigma_0(E)$ is the cross section for formation of the compound nucleus for ions with energy E; $P_{xn}(E)$ is the probability of evaporation of x neutrons at the energy of excitation corresponding to the given energy of the ions; $(\overline{\Gamma_n}/\Gamma)^x$ is the average of the ratio of the neutron width of the level to the total width. As we know, the value of $\sigma_0(E)$ has the form of a threshold function,

because of the presence of the Coulomb barrier which prevents the fusion of the low energy ion with the target. This results in the fact that the energy of excitation of the compound nucleus in the region of transuranic elements is of the order of a few tens of Mev and the most probable value of x is 4 — 5. The factor $(\overline{\Gamma_n}/\Gamma)^x$ significantly lowers the cross section for fissioning nuclei: the compound nucleus in the overwhelming number of cases undergoes fission, and only in a negligible fraction of decays is the energy of excitation transferred only to neutrons. With increasing charge of the compound nucleus, the fissionability parameter Z^2/A increases, and the fission barrier is reduced, so that the quantity $(\overline{\Gamma_n}/\Gamma)$ is reduced and consequently the cross section falls. To illustrate this fact, we show in Fig. 22 the values of the cross sections for some of the reactions produced by multiply-charged ions as a function of the fissionability parameter of the compound nucleus.

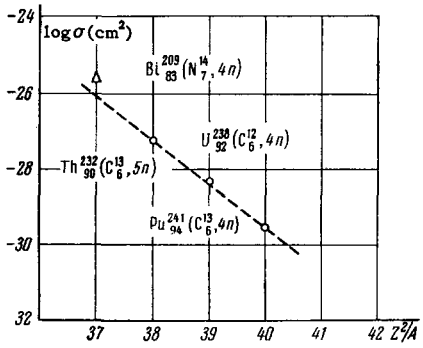

FIG. 22. Dependence of the cross section for certain nuclear reactions on the value of Z^2/A for the compound nucleus.

What are the possibilities for struggling with this catastrophic difficulty? Each new element can be obtained by several methods. If, for example, we choose an element with Z = 104, then for its synthesis we can use the following nuclear reactions:

$$_{92}U + {}_{12}Mg, \quad {}_{94}Pu + {}_{10}Ne, \quad {}_{96}Cm + {}_{8}O, \quad {}_{98}Cf + {}_{6}C.$$

Each of these possibilities in turn is not unique, since all the elements enumerated have several sufficiently stable isotopes. However, the various alternatives are not equally good since they lead to the formation of different isotopes of the 104-th element, and what is extremely important, the cross sections for the corresponding reactions may differ markedly. In order to deal with the most probable reaction, we must first of all use as bombarding particle a nucleus with the minimum charge, since this as a rule leads to the lowest excited compound nucleus. In addition, because of the influence of the parameter Z^2/A on the fissionability of the compound nucleus, the competition by fission will be less important for a nucleus with the maximum mass value A. Consequently, the reaction cross section will be optimal if, as our target nucleus, we choose the heaviest isotope of the element. Weighable amounts of heavy isotopes of transuranic elements are obtained by the use of nuclear reactors. Thus, we again arrive at the idea of the necessity for a close interaction of both methods for obtaining heavy elements. It may be hoped that the production of super-powerful reactors will make it possible to obtain heavy isotopes of curium, californium, and einsteinium in amounts sufficient for preparing targets.

To obtain further transuranic elements it may turn out to be favorable in our opinion to use reactions in which the main part of the energy transferred to the nucleus by the heavy ion is carried off as the result of a process which is not associated with the formation of a uniformly heated compound nucleus and consequently not fighting against competition from fission. One may, for example, imagine that as a result of a direct interaction occurring at the periphery of a nuclear target there will be ejected an energetic α particle or one or two neutrons which will take off a large fraction of the energy of the initial particle, while the residual, slightly excited nucleus will then eject one or two neutrons by evaporation. Competition from the fission process in this case will be considerably weaker in its effect on the probability for formation of the new nucleus.

The low value of the cross section for formation of new elements stimulates the development of such methods of identification as can be successfully used literally for counting atoms individually. Spontaneous fission is such a characteristic process that under complicated conditions of experiment it has an unquestionable advantage over α decay and K capture with regard to the simplicity and efficiency of counting. It is very important that in counting spontaneous fission there be practically no background. In Table IV we give a qualitative comparison of the efficiency of methods for recording spontaneous fission, α decay, and K capture. We give the minimum intensity of radiation needed for identification of the nucleus.

Table IV

Spontaneous fission	α-decay	K capture
0.211/hr	50/hr	5000/hr

The development of methods for counting fast spontaneous fission will apparently enable us to obtain more reliable methods for identification of new elements. These methods may be successfully applied despite the extremely small yield of products of nuclear reactions. The use of these methods will enable further progress in solving the problem of synthesis of new elements.

In this survey we have presented briefly the present state of investigations in one of the branches of nuclear physics whose development is closely associated with the name of Igor Vassil'evich Kurchatov. During the twenty years which have elapsed since the time of the

discovery of spontaneous fission, there has been accomplished a tremendous amount of work on the study of this phenomenon. However, it seems to us that both the theoretical and experimental investigations of spontaneous fission are still very far from completion. One may express the hope that in the next few years there will be new successes in this field of nuclear physics, especially with regard to the obtaining of new transuranic elements.

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