

0-35

9/IX-68

ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

Дубна



E7 - 3942

Ju. Tz. Oganessian

FISSION OF EXITED NUCLEI

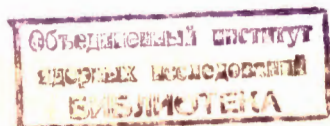
ЛАБОРАТОРИЯ ЯДЕРНЫХ РЕАКЦИЙ

1968

E7 - 3942

Ju.Tz.Oganessian

FISSION OF EXITED NUCLEI



Since the discovery of the uranium fission, the study of the fission mechanism is one of the important and interesting problems of nuclear physics. A lot of work is devoted to the investigation of this phenomenon; to explain experimental data various nuclear structure models are used: liquid drop model, shell model, statistical theory etc. It is natural that the range of application of a model is very limited and there is as yet, no consistent theoretical interpretation of this complicated nuclear process.

The well known mass distribution of fission fragments of the ^{235}U with thermal neutrons is sometimes explained from positions little compatible with each other. It is probably due to this circumstance, that a great deal of effort is taken to explain the mechanism of low-energy or spontaneous nuclear fission.

However it was established by experiments that with increasing nuclear temperature the picture changes: at an excitation energy higher than 50 Mev, fission becomes actually symmetrical $/1,2/$. It may be supposed that this energy increase does not radically alter the mechanism of this complicated nuclear transformation. At the same time the field of the experimental study of nuclear fission is considerably extended.

Indeed, for the investigation of spontaneous fission or fission on thermal neutrons, only nuclei from uranium to californium can be used while in the high excited state all nuclei with $\frac{Z^2}{A} > 25$ undergo

fission ^{/3/}. Of course, data obtained for a heated nucleus can not be directly used for explaining all fission regularities near the threshold. However the facets such as nuclear shape at the saddle point, mechanism of nuclear motion from saddle-to-scission point, character of separation etc. are common and important for the fission problem as a whole.

So, in what follows we shall consider nuclei at an excitation energy higher than 50 Mev, based on the fact that at these energies the structure features of low-energy fission are completely absent.

We note however, that the quantitative study of the excited nucleus fission, meets serious difficulties. When the projectile energy increases the number of reaction channels also increases and the initial state of a fissioning system is uncertain. Experiments with relativistic protons may serve as an example of this; here, after the fast cascade emission of nucleons quite a number of nuclei with a wide excitation spectrum undergo fission ^{/4,5/}.

From this point of view, heavy ions have some advantages since at an energy lower than 10 Mev per atomic mass units, the main mode of interaction of complex nuclei is the complete fusion with the production of a high excited compound nucleus. On one hand, this defines unambiguously the nuclear state just before fission, on the other hand, it allows a large number of nuclei in a wide range $\frac{Z^2}{A}$ to be investigated.

Below we shall discuss the experimental data on fission induced by heavy ions from ¹²C to ⁴⁰Ar .

According to modern considerations in the fission process the nucleus passes through a transition state—the saddle point, which corresponds to the fission barrier. It is assumed that at the saddle point the nucleus is in thermal equilibrium. This state is essentially initial during all further fission processes.

If such a situation takes place, then according to the statistical theory, the angular distribution of the fragments is unambiguously related to the effective moment of inertia J_{eff} which characterizes the nuclear shape at the saddle point ^{/7-9/}.

$$\frac{1}{J_{\text{off}}} = \frac{1}{J_{\parallel}} + \frac{1}{J_{\perp}}$$

where J_{\perp} and J_{\parallel} are the moments of inertia of the transition state nucleus at the saddle deformation about an axis perpendicular and parallel to the axis of symmetry, respectively.

The experimental data on J_{off} calculated from the angular anisotropies for heavy ion-induced fission in various target nuclides is given in Fig.1.

It should be noted that within the experimental accuracy J_{off} does not depend on the mode of production, the excitation energy and the angular momentum of a fissioning nucleus ^{/10,12/} but it is only a function of $\frac{Z^2}{A}$.

Now it is interesting to compare the experimental data with the calculation results for J_{off} using the usual liquid drop model^{/10,11/}. A noticeable disagreement for large $\frac{Z^2}{A}$ values is seen in Fig.1 which points out that the deformation at the saddle point is small as compared with the shape obtained by means of the liquid drop model.

However, this disagreement is practically eliminated if, as before in the framework of the liquid drop model, the distribution of the nucleon density of the nucleus is correctly taken into account and the surface tension as a function of the curvature of the effective surface is introduced in calculating J_{off} ^{/13/}.

It is very important that the calculation results agree with the experimental ones.

This provides evidence for the fact that the liquid drop model is a good approximation in calculating the potential energy and the nuclear shape at the saddle point. At the same time the assumption of the statistical nuclear equilibrium on the top of the barrier is valid with great accuracy for all investigated nuclei.

Hence, it follows that the deformed nucleus really for a long time is in an intermediate state corresponding to the saddle point.

Note that the extreme point at $\frac{Z^2}{A} = 43.5$ (Fig.1) obtained from the angular distribution of ¹⁴⁰La nuclei in the fission of ²³⁸U by

the ^{40}Ar ions corresponds to $\frac{J_{\text{app}}}{J_{\text{eff}}} = 0.1$. Due to this it is possible to determine accurately from experiments the main parameter of the liquid drop model $(\frac{Z^2}{A})_{\text{crit}}$ which defines the stability limit of nuclear matter against fission. The extrapolation $1/J_{\text{eff}} \rightarrow 0$ (nuclei with spherical shape at the saddle point) gives $(\frac{Z^2}{A})_{\text{crit}} = 45.1$.

It is shown above that the main characteristics of the nucleus in the quasistationary state near the barrier top can be determined fairly accurately. Further motion of the nucleus from saddle-to-scission point is very complicated whereas the appropriate information may be obtained only at the initial moment (the barrier top) and in the very final stage (division into two fragments). Thus, the charge and mass distributions of fragments, the total kinetic energy and fluctuations of the fragment kinetic energy are essentially the only data which reflect the fission mechanism.

It is obvious that this information is explicitly insufficient for a complicated nuclear system with many degrees of freedom to be described. Therefore, to describe the phenomenon one or other of the models is used which supposes a priori a certain regularity in the development of the process.

When a fissioning nucleus is highly excited it is assumed that the energy exchange between different degrees of freedom proceeds far more rapidly, compared to the rate of the deformation change. This allows one to consider the nucleus as an isolated system in thermodynamic equilibrium in all the stages of the motion from saddle-to-scission point ^[14-17]. Thus, all the process may be considered in the very final stage, that is at the scission point, and the statistical theory may be applied to describe the main fission regularities.

This approach seems to be justified to fission from excited states for general reasons ^[14,18], therefore an experimental check of the statistical theory applicability in this case is important.

It should be noted that the statistical model has been used for explaining much experimental data ^[19-21] on fission of excited nuclei. However until recently, only a small number of light nuclei with $\frac{Z^2}{A}$ from 31 to 35 have been systematically investigated.

Below we give the results of the measurements of the mass and charge distributions of fission fragments /22,23/ for nuclei with $\frac{Z^2}{A}$ from 37.5 to 43.5.

When bombarding ^{209}Bi and ^{238}U thin targets with the ^{16}O , ^{20}Ne and ^{40}Ar ions, fragments of the rare-earth group and, in some cases, heavy fragments from gold to polonium were separated by the radiochemical method /24/.

Further, using a Ge(Li) -gamma spectrometer, the gamma radioactivity of fragments was measured and the obtained spectrum was analysed with the aim of identifying the isotopes and determining their yield. This technique was fairly reliable and the yield was determined with an error of not greater than 15%.

To plot the mass distributions, assumptions were made about the charge distribution of fragments which were then checked experimentally.

1. For each fragment mass A_f the yield of isobars with Z differing from the most probable value of Z_p is described by the Gaussian distribution:

$$W(Z - Z_p) = \frac{1}{(\pi c)^{1/2}} \exp - \frac{(Z - Z_p)^2}{c}$$

2. The dependence of Z_p on A_f was calculated under the following hypothesis:

- i) constant charge density of fragments;
- ii) equal charge displacement /25/;
- iii) the charge distribution from the minimum of the potential energy of fragments /16/.

It was also assumed that neutrons were evaporated from fragments /25/. ($\Gamma_n / \Gamma_f \ll 1$) in a number proportional to the fragment mass $\nu_f = \frac{\nu}{A_0} A_f$.

With the aid of an electronic computer, using the least square method, the parameters ν and c were chosen for different dependences $Z_p(A_f)$ giving the smallest deflection of the experimental points from the smooth curve which, as expected, was well described by the Gaussian distribution :

$$P(A_f) = \frac{1}{(\pi\sigma^2)^{1/2}} \cdot \exp - \frac{(A_f - \frac{A_0}{2})^2}{\sigma^2}$$

with one parameter σ .

Using such a method, preference is given to no one hypothesis about the charge distribution of fragments, the latter follows from the best fit of the experimental points on the mass curve.

The experimental data is given in Table 1. It was found for all reactions that the worst agreement is obtained when the charge is assumed to be proportional to the fragment mass (Fig.2) while for the two other cases the results coincide practically.

The mass spectra for fission fragments are given in Fig.3. Using the dependence of the mass distributions width on the nuclear temperature we obtain as a function of the parameter $\frac{Z^2}{A}$ for a fixed excitation energy. As is seen from Fig.4 at $\frac{Z^2}{A} > 38$ a strong enlargement of the mass curve is observed.

Now it is interesting to compare the experimental data with the calculations by the statistical theory. Several methods of such calculations are known. Here we use the method suggested in ref. /16/ which is the most convenient and accurate.

In the framework of the statistical theory the fission probability for a given ratio of fragment masses is

$$P(A) = \exp \left[- \frac{(A - \bar{A})^2}{\bar{A}^2 \langle \Delta \delta \rangle^2} \right]$$

where

$$\bar{A} = \frac{A_0}{2} \quad \delta = \frac{1}{2} \left(1 - \frac{2A_f}{A_0} \right) \quad \langle \Delta \delta \rangle^2 = T \left[\frac{1}{2} \frac{\partial^2 W}{\partial \delta^2} \Big|_{\delta=\bar{\delta}} \right]^{-1}$$

Here T is the nucleus temperature.

$\frac{\partial^2 W}{\partial \delta^2}$ is the second derivative of the total potential nucleus energy which, according to ref. /16/, can be presented as:

$$\frac{\partial^2 W}{\partial \delta^2} = \alpha_1 \Delta E_s + \alpha_2 \Delta E_o - \alpha_3 \bar{E} - \alpha_4 E_{ds} + \alpha_5 E_{do} - \alpha_6 A_0^{7/8}$$

where a_1, a_2, \dots, a_8 are the constant coefficients, ΔE_s and ΔE_c is the difference of the surface and Coulomb energy for the initial (spherical) nucleus and two (spherical) fragments E_{ds} , E_{dc} are the surface and Coulomb energies of the fragment deformation at the scission moment.

E is the energy for the Coulomb interaction of fragments which is equal to the total kinetic energy, the latter being determined accurately from the experimental dependence ²⁷.

$$\bar{E} = 0.1065 \frac{Z_0^2}{A_0^{1/3}} + 20.1 \text{ MeV}$$

In calculating the potential nucleus energy different mass formulas were used [25,28,29]. The comparison of the theoretical and experimental data in Fig.5 shows that agreement is obtained only for a very narrow region of light nuclei, while for $\frac{Z^2}{A} > 38$ there is an essential disagreement considerably exceeding the errors. Thus, the large disagreement can be eliminated by not varying the model parameters within reasonable limits.

It is also seen from experiments that the dependence $\langle \sigma \rangle^2$ on the excitation energy is stronger than the theory it predicts.

However, it seems to us that the most strict criterion of the validity of the statistical approach, is the fluctuation of the fragment charge for a given mass ratio (or the mass fluctuation for a given charge ratio).

Indeed, according to ref. [16], in calculating the isobar or isotopic curves the well-known term of the mass formula $= \frac{(A - 2Z)^2}{A}$ alone is used which takes into account the affinity of the protons and neutrons in the nucleus.

In this connection, we have measured the isotopic distributions of fragments near the maximum of the mass curve for nuclei with different $\frac{Z^2}{A}$. It is seen from Fig.5 that the charge (or isotopic) curves are well described by the Gaussian distribution and the parameter c agrees with the data of Table 1.

The experimental dependence of the charge distribution width c on $\frac{Z^2}{A}$ (Fig.6) agrees with the theoretical one, only in the range of light nuclei and disagrees strongly for $\frac{Z^2}{A} > 38$.

The isotopic distributions in different places of the mass curve were measured for the reaction $^{238}\text{U}(\text{}^{40}\text{Ar}, f)$. The c values, obtained from this data as a function of the mass asymmetry (Fig.7), indicate that the charge distribution width is not constant for all the masses, as is usually assumed when calculating the mass distributions of fragments.

The main cause of the disagreement between the experimental data and the theoretical predictions of the statistical model, lie, as it seems to us, in that, unjustified assumptions on the fission mechanism have been made.

The existence of thermodynamic equilibrium at the scission point, which is used for the calculation of the nuclear potential energy and of all other quantities, seems to be doubtful for the following reasons.

The statistical approach is valid only in the case when the relaxation time of the nucleus is far shorter than the time of the deformation change. However, the rate of the deformation change is minimum at the barrier and maximum at the scission point. In the latter case it may be the same as or even larger than the rate of the nucleus relaxation [30].

This fact contradicts the assumptions of the statistical theory.

The correct description of the phenomenon may be obtained only with the account of the dynamics of the process which needs additional assumption of the kinetic properties of the fission process.

Let us try to explain qualitatively the experimental data.

For different $\frac{Z^2}{A}$ the starting figures (saddle point) may be essentially different (Fig.1), therefore the number of configurations along the whole path up to the scission point is also different. Taking into account the accelerative character of the motion, it should be assumed that during the main time, the nucleus has a deformation which differs strongly from the figure at the scission point. This means that the fate of the separation was at stake, long before the

moment when the nucleus was ready to assume the shape of two separate fragments. In the process of charge of deformation, each figure has a certain stability against the asymmetrical variations of the shape /31/.

For such an approach the fluctuation of the mass and charge of fragments depends on the rate of the deformation change and on the initial shape of the nucleus.

However the exact calculation is a matter of some difficulty but with some simplifications of the problem, the dependence of the mass distribution width $\langle \sigma \rangle^2$ may be obtained to be close to the experimental one.

It should be noted that the good agreement of the experimental points (Fig.4,6) with the calculations by the statistical theory in the range of light nuclei ($\frac{Z^2}{A} = 31 - 34$) is accounted for by the fact that the nuclear shapes at the scission moment and at the saddle point differ little from each other. Since the second stage of the process is practically absent, the assumption of thermodynamic equilibrium at the scission point is essentially related to the saddle point which is quite justified and agrees well with experimental results.

A noticeable enlargement of the mass curve with increasing $\frac{Z^2}{A}$ gives rise to an interesting phenomenon. "cascade fission" of the nucleus with production of three fragments of about equal masses /32/

The essence of this phenomenon is that an excited compound nucleus undergoes in some cases a strong asymmetrical fission.

If the excitation energy of a heavy fragment is higher than its fission barrier, then it can disintegrate once again into two parts. The cross section of the process depends on the mass distribution in the first stage of fission (the yield of large masses) and on the fission probability for the heavy fragment. This mechanism explains clearly the experimental data on fission of nuclei into three fragments in heavy-ion induced reactions /33,34/.

Note also that the fission of heavy excited nuclei may be a good method for synthesizing new isotopes /35/. It seems to us that fission induced by ions heavier than argon is very promising for obtaining nuclei far off the stability line /36/.

References

1. Colby L.J., Cobble J.W., *Phys. Rev.*, 121, 1410 (1961).
2. Unik J.P., Huizenga J.R., *Phys. Rev.*, 134, B90 (1964).
3. Obukhov A.I., Perfilov N.A., *Jadernaya Fizika* 3, 979 (1966).
4. Rudstam G., Sørensen G., *J. Inorg. Chem.*, 28, 771 (1965).
5. Hagebø E. J. *Inorg. Nucl. Chem.*, 29, 2515 (1967).
6. Bohr A., *Proceedings of the First United Nations International Conference on the Peaceful Uses of Atomic Energy (U.N. N.Y. 19 1955)* 2, 151.
7. Strutinski V.M., *JETP* 30, 606 (1956).
8. Strutinsky V.M., *Atomnaya Energia* 2, 508 (1957).
9. Halpern I., Strutinski V.M., *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy (U.N. Geneva 1958)* 15, 408.
10. Chaudhry R., Vandenbosch R., Huizenga J.R., *Phys. Rev.*, 126, 220 (1962).
11. Bate G.L., Chaudry R., Huizenga J.R., *Phys. Rev.*, 131, 722(1963).
12. Karamyan S.A., Kuznetsov I.V., Muzycka Yu.A., Oganessian Yu.Tz. Penionzkevich Yu.E., Pustyl'nik B.I., *Jadernaya Fizika* 6, 494(1967).
13. Strutinski V.M., *Jadernaya Fizika* 2, 821 (1965).
14. Fong P., *Phys. Rev.*, 102, 434 (1956).
15. Newton T.D. *Proceedings of the Symposium on the Physics of Fission held at Chalk River (Ontario, 1956)*.
16. Pik-Pichak G.A., Strutinski V.M., *Nuclear Fission (Atomizdat, Moscow 1962)*.
17. Mix J.R., *Preprint UCRL-11338, 1964*.
18. Hill D.L., Wheeler J.A., *Phys. Rev.*, 89, 1102 (1953).
19. Burnett D.S., Gatti R.C., Plasil F., Price R.B., Swiatecki W.J., Thompson S.G., *PReport UCRL-11079, 1963*.
20. Viola V.E., Blann H.M., Thomas T.D., *Proceedings of the Second Conference on Reaction between Complex Nuclei (N.Y. 1960) Report UCRL-11193, 1963*.
21. Plasil F. *Report UCRL-11193, 1963*.

22. Karamyan S.A., Liptac I., Oganessian Yu.Tz., Penionzkevich Yu.E., Urbanetz I., *Atom. Ener. (USSR)* 23, 564 (1967).
23. Karamyan S.A., Normuratov F., Oganessian Yu.Tz., Penionzkevich Yu.E., Pustynnik B.I., Flerov G.N., *Preprint JINR P7-3732*, 1968.
24. Kuznetsov I.V., Maltseva N.S., Oganessian Yu.Tz., Sukhov A.M., Shohegolev V.A., *Preprint JINR P7-3710*, 1968.
25. Green A.E.S., *Phys. Rev.*, 95, 1006 (1954).
26. Sikkeland T., *Preprint UCRL-16348*, 1965.
27. Terrell J., *Phys. Rev.*, 113, 527 (1959).
28. Cameron Q.G.W. *Canad.J.Phys.*, 35, 1021 (1957).
29. Myers W.D., Swiatecki W.J., *Preprint UCRL-11980*, 1965.
30. Nix J.R., *Contributions of International Conference on Nuclear Structure (Tokyo, 1967)*.
31. Strutinski V.M., *JETP* 45, 1900 (1963).
32. Muzycka Yu.A., Oganessian Yu.Tz., Pustynnik B.I., Flerov G.N., *Jadernaya Fizika* 6, 306 (1968).
33. Price R.B., Fleischer R.L., Walker R.M., *Proceeding of the Third Conference on Reactions Between Complex Nuclei (University of California Press USA 1963)*.
34. Karamyan S.A., Kuznetsov I.V., Oganessian Yu.Tz., Penionzkevich Yu.E., *Jadernaya Fizika* 5, 959 (1967).
35. Flerov G.N., *Nucleonica* 12, 1081 (1967).
36. Flerov G.N., *Preprint JINR D7-3623*, 1967.

Received by Publishing Department
on June 26, 1968.

Table 1. Experimental data on heavy-ion induced fission^[23]. The c_{exp} values are obtained for rare-earth fragments.

	$\frac{Z^2}{A}$	E_t (MeV)	σ	$\bar{\nu}$	c	c_{exp}
$^{209}\text{Bi} (^{20}\text{Ne}, f)$	37.7	100	710	10.8	0.56	
$^{288}\text{U} (^{12}\text{C}, f)$	38.4	42				0.7 ± 0.2
$^{288}\text{U} (^{16}\text{O}, f)$	39.4	81	1280	11.2	1.7	
$^{288}\text{U} (^{20}\text{Ne}, f)$	40.3	120	2280	12.6	3.3	
		95	1660	11.5	2.9	
		65	1130	8.9	2.7	2.5 ± 0.2
$^{209}\text{Bi} (^{40}\text{Ar}, f)$	41.0	115	2200	9.5	2.75	
$^{288}\text{U} (^{40}\text{Ar}, f)$	43.5	110	2790	13.3	3.0	
		75	1980	10.6	2.9	2.8 ± 0.2

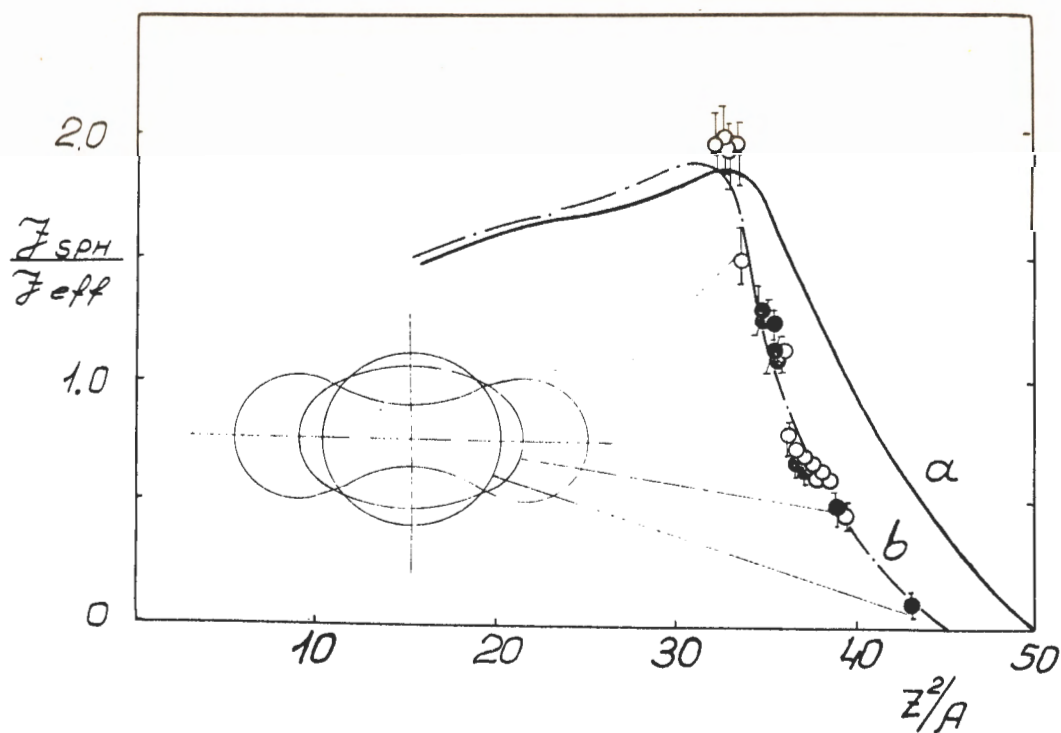


Fig.1. Dependence of the effective moment of inertia at the saddle point on the fission parameter $\frac{Z^2}{A}$. The curves are the calculation results obtained by the liquid drop model: a) for a nucleus with sharp edge, b) taking into account the nucleus density distribution in the surface layer. The experimental points: open circles are the data from experiments on deuterons and alpha particles ^[10,11], black points - on heavy ions. The figures correspond to the nucleus shape at the saddle point under the assumption of axial symmetry.

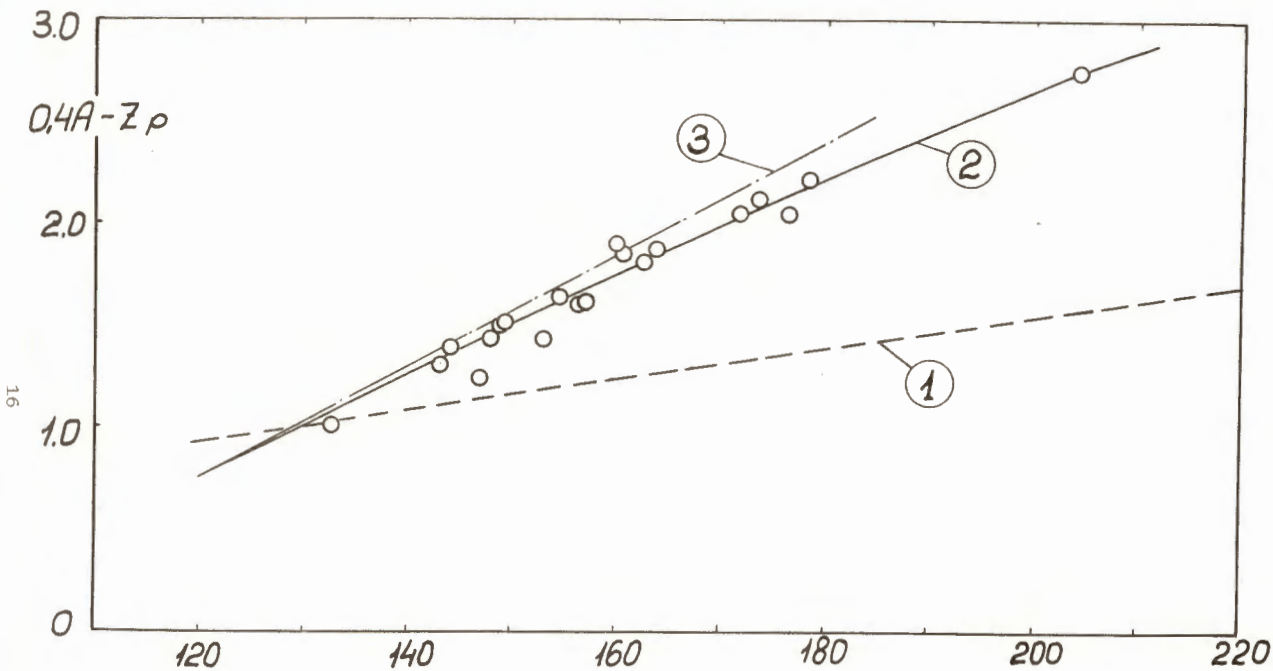


Fig.2. $(0.4A - Z\rho)$ as a function of the fragment mass for the reaction $^{238}\text{U} + ^{22}\text{Ne}, f$. The curves are the predictions of the hypothesis;

- 1, constant charge density of fragments, proportionality of the charge to the fragment mass,
- 2, equal charge displacement, 3, the charge distribution according to the minimum of potential energy at the scission point [16].

Af

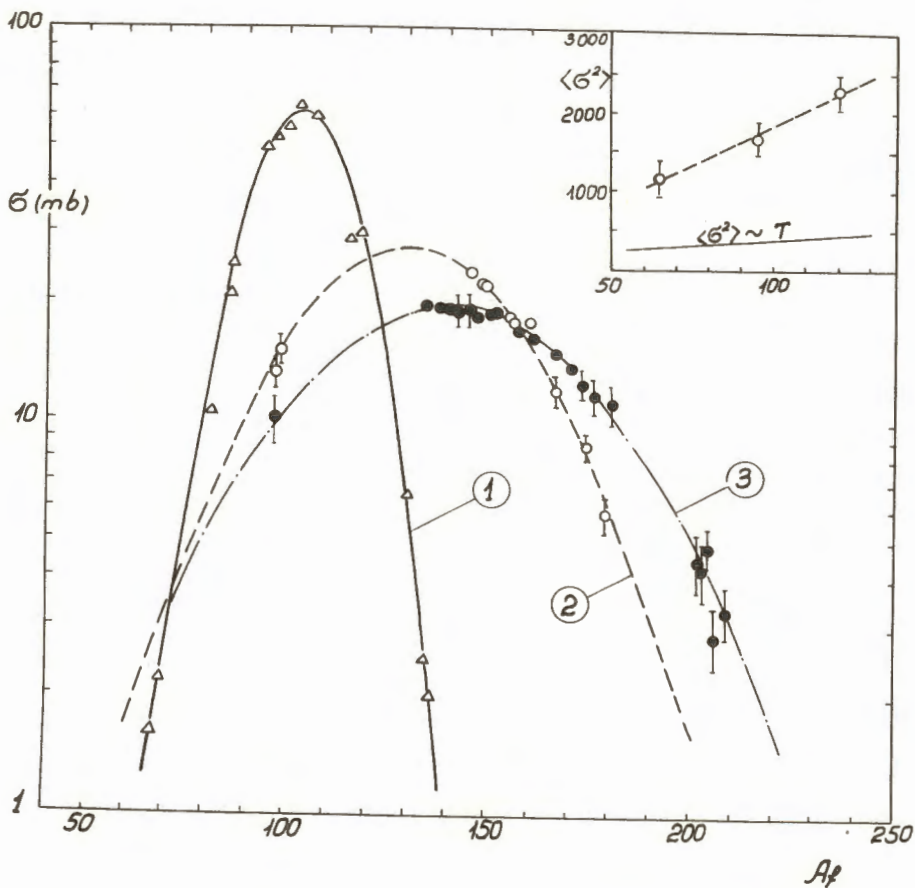


Fig.3. Mass distribution of the fission fragments: Curves 1 - $^{107}\text{Au}(^{12}\text{C},f)/20/$ excitation energy 105 Mev; 2 - $^{288}\text{U}(^{20}\text{Ne},f) /23/$ " " " 95 Mev; 3 - $^{288}\text{U}(^{40}\text{Ar},f) /23/$ " " " 110 Mev

The upper diagram is the dependence of σ^2 on the excitation energy of a fissioning nucleus $^{288}\text{U}(^{20}\text{Ne},f)$

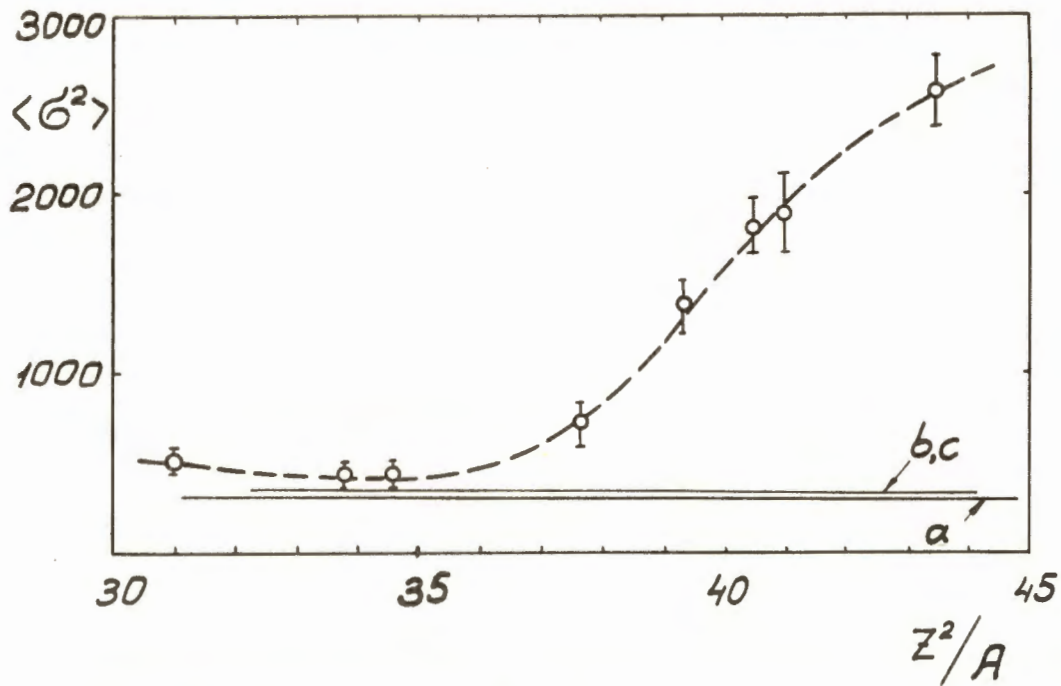


Fig.4. Mass distribution width σ^2 as a function of the parameter Z^2/A at an excitation energy of about 100 Mev. The dotted line is drawn through the experimental points. The continuous lines are the predictions of the statistical theory obtained for different mass formulas; a) Cameron^[28], b) Green^[25], c) Myers and Swiatecki^[29].

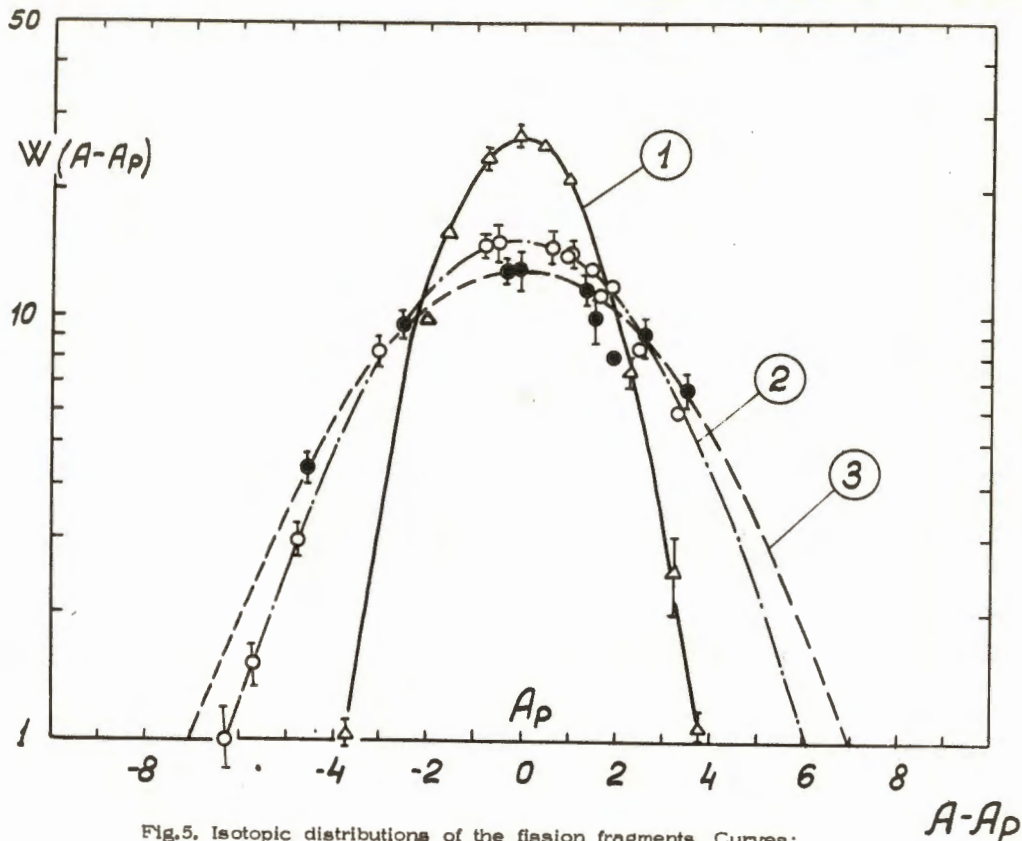


Fig.5. Isotopic distributions of the fission fragments. Curves:

1. $^{187}\text{Au} (^{19}\text{C}, f) /20/$
2. $^{238}\text{U} (^{20}\text{Ne}, f) /23/$
3. $^{238}\text{U} (^{40}\text{Ar}, f) /23/$

Parameter $\frac{\sigma}{\sigma_{\text{exp}}}$ is 34,5, 40,5 and 43,5 respectively. The charge distribution widths σ_{exp} for this reaction are presented in Table 1.

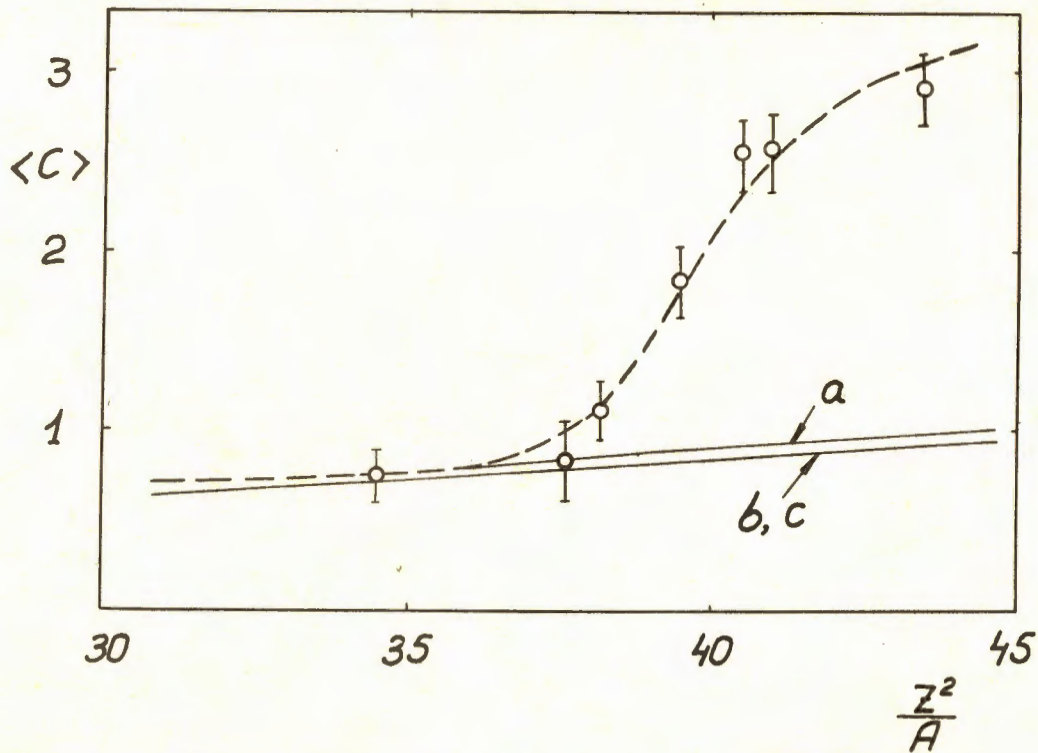


Fig.6. Charge distribution width as a function of the parameter $\frac{Z^2}{A}$ at an excitation energy of about 80 Mev. The dotted line is drawn through the experimental points. The curves a,b,c are the theoretical calculations (see Fig.4).