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VARIANCE OF THE ENERGY DISTRIBUTIONS OF FRAGMENTS FORMED BY LOW-ENERGY FISSION: EXPERIMENTAL DATA AND THEORETICAL PREDICTIONS

Yu. A. LAZAREV Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna, Union of Soviet Socialist Republics

ABSTRACT. Experimental data on the variance of the total number of prompt neutrons (σ_{ν}^2) and the variance of the total kinetic energy distribution of fission fragments $(\sigma_{\rm E_K}^2)$ resulting from low-energy fission of heavy nuclei are reviewed. An analysis of the dependences of the quantities σ_{ν}^2 and $\sigma_{\rm E_K}^2$ on the main parameters of the fissionable nucleus is presented: the mass number (in the range $230 \le A \le 258$), the number of protons in the nucleus (in the range $90 \le Z \le 102$), the fissility parameter x, and the excitation energy E* for E* = 0 (spontaneous fission) and E* $\cong 6.5$ MeV (thermal neutron fission). It is shown that these dependences are regular and that, despite certain contradictions, the experimental results of several dozen works concerning the variance of energy distributions are on the whole in fairly good agreement. There appears to be a strong exponential-like dependence of the variances on the number of protons in the nucleus or on the fissility parameter: an increase of 15% in x leads to an increase of several times in the fission fragment energy fluctuations. The experimental curves are compared with the predictions of the dynamic liquid-drop model of Nix and Swiatecki and the statistical theory of Fong, as well as with the results of other calculations performed on the basis of a statistical approach. It is shown that the theoretical calculations performed so far do not give a satisfactory description of the patterns observed, leaving open the question of the nature of fission fragment energy fluctuations, their magnitude and the dependence of the magnitude on Z, A and E* of the fissionable nucleus.

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1. INTRODUCTION

The nuclear fission reaction exhibits a remarkable feature: the number of final states of the reaction products – pairs of fragments – is enormous. According to estimates by Wilets [1] it is $\sim 10^7$. In experiments this feature of the fission reaction manifests itself in the fact that all the characteristics of the fragments – their masses, charges, energies, etc. – exhibit significant fluctuations

The present status of the problem of nuclear fission in general and of the question of the properties of fission products in particular is characterized by a large amount of experimental and theoretical material concerning the mean characteristics of fission fragments and the many correlations between them for spontaneous fission and for fission by widely different particles over a large range of nucleon compositions and excitation energies of the fissionable nucleus [2–5]. The main patterns of change of the mean quantities — masses, charges, kinetic energies, fragment excitation energies, etc. — are well known and the causes of these changes are fairly well understood.

The variations of these fission characteristics have not been investigated extensively. Both experimental and theoretical investigations of these variations are a much more complex problem. These variations appear to be of a quantum nature, perhaps a complex, dynamic characteristic of the fission process. They probably arise as a result of the enormous number of degrees of freedom of the fissioning system and, in principle, they reflect the diversity of the system's initial conditions at the saddle point, the diversity of the paths of evolution of the system from the saddle point to the scission point and the diversity of the outcomes of this evolution at the scission stage. At the same time, the variations of the characteristics of fission fragments may contain valuable additional information concerning the determination of the corresponding mean values, shedding light on the fission mechanism itself.

On the other hand, with the extensive accumulation of experimental and theoretical results over the past ten years there has been significant qualitative progress in the physics of fission, above all due to the experimental discovery of spontaneously fissioning isomers and of other phenomena and facts associated with nuclear shape isomerism. Strikingly unusual and interesting phenomena have been observed in the synthesis of spontaneously fissioning isotopes of the new elements — kurchatovium (Z = 104) and elements 106 and 107 — and in the investigation by comparatively traditional methods of low-energy fission of the heavy isotopes of fermium and of element 102. It is particularly worth noting the significant change in ideas concerning the systematics of the lifetimes for spontaneous fission of nuclei with Z > 102 (initiated by work done at Dubna [6-9]), the symmetric fission of the heavy isotopes of fermium, and the many other unusual properties of the fission fragments of fermium and element 102 [10-13].

The progress in the theory of fission has resulted above all from a substantial development and generalization of ideas about the shell structure of nuclei — especially strongly deformed nuclei — and from the construction of the theoretical apparatus necessary for meaningful calculations [5, 14, 15]. This in turn has led to a much deeper understanding of the complex structure of fission barriers and of the probability and asymmetry of the fission of heavy nuclei [16], and to attempts to predict some of the dynamic characteristics of fission — for example, lifetimes for spontaneous fission [17, 18].

However, the progress about which we have been speaking has at present only a potential bearing on the question of the variations of the fission characteristics, although these are actually one of its basic features. We do not even know reliably and quantitatively what the main factors are that determine the extent of the variations of fission fragment properties. Moreover, in many cases we are not even qualitatively certain about the patterns of change in the extent of such variations with respect to the main parameters (A, Z and E*) of a fissionable nucleus.

Despite the existence of some experimental information, one of the least studied questions is the variations of the kinetic and excitation energies of the fragments formed in low-energy fission of heavy nuclei¹. Unfortunately, the studies which have been made in this connection have covered only very narrow ranges of Z and A values of the fissionable nucleus, so that it has not been possible to discern any regularities in the changes of the width of the overall energy distributions of the fission fragments.

In the paper an attempt is made to analyse the experimental data on the variance of the total number of prompt neutrons $(\sigma_{E_k}^2)$ and the variance of the total kinetic energy of fragments $(\sigma_{E_k}^2)$ resulting from low-energy fission of nuclei ranging from thorium to element 102. We present the systematics of σ_{ν}^2 and $\sigma_{E_k}^2$ and, on this basis, determine the dependence of these quantities on the main parameters of the fissioning nucleus: A in the range $230 \le A \le 258$, Z in the range $90 \le Z \le 102$ and excitation energy $E^* = 0$ (spontaneous fission) and $E^* \cong 6.5$ MeV (thermal neutron fission). We compare the dependences of σ_{ν}^2 and $\sigma_{E_k}^2$ on the parameters in question and verify their mutual consistency. We also discuss the dependence of the variance

¹ Here and subsequently, unless otherwise indicated, by "low-energy fission" we mean exclusively spontaneous fission (SF) and thermal neutron fission (n, f); for example, ^{252}Cf (SF) and $^{235}U(n, f)$.

of the overall energy distributions of fission fragments on the fissility parameter x in the range $0.75 \le x \le 0.87$ for $E^* = 0$ and $E^* \cong 6.5$ MeV. The experimental curves are compared with predictions based on the dynamic liquid-drop model. The possibility is discussed of describing the experimental material in question with the help of Fong's statistical theory of fission.

The main purpose of the paper is the systematization and critical analysis of all experimental data published so far on the variations of the energies of fission fragments of heavy nuclei; we do not claim to explain fully or interpret in detail the experimental data in question.

VARIANCE OF THE TOTAL NUMBER OF NEUTRONS EMITTED DURING FISSION AND ITS DEPENDENCE ON THE INITIAL PARAMETERS OF THE FISSIONABLE NUCLEUS

It is known that the total energy Q released during fission consists of fission fragment kinetic energy E_k and fission fragment excitation energy E_x . As most of the fission fragment excitation energy is removed through neutron emission, one may assume that the variance of the number of neutrons is fairly positively correlated with the variance of the distribution of the total fission fragment excitation energy². Therefore we analyse the values of σ_{ν}^2 , which can be measured directly. By definition,

$$\sigma_{\nu}^{2} = \sum_{\nu=0}^{\nu=\nu_{\text{max}}} (\nu - \bar{\nu})^{2} P_{\nu}$$
 (1)

where $\overline{\nu}$ is the mean number of neutrons emitted per fission and P_{ν} the probability of emission of exactly ν neutrons during one fission event

$$\sum_{\nu=0}^{\nu=\nu_{\text{max}}} P_{\nu} = 1$$

The first attempt to analyse the (Z, A) dependence³ of the distribution of the number of neutrons produced during fission, P_{ν} , was made by Terrell [19]. He assumed that:

- (1) Neutrons can be emitted by fission fragments whenever this is energetically possible
- (2) The emission of any neutron from any fragment reduces the excitation energy of the fragment by a constant amount, E_0
- (3) The total excitation energy of the two primary fragments has, in the case of binary fission, a Gaussian distribution with a variance $\sigma^2 E_0^2$.

² As will be shown in section 6, this assumption is subsequently confirmed.

³ Here, and subsequently, "(Z, A) dependence" means the dependence of the quantity in question on Z and A of the fissioning nucleus; similarly, for one of the parameters we speak of "Z dependence" or "A dependence".

TABLE I. VARIANCE OF THE NUMBER OF NEUTRONS EMITTED IN THE FISSION OF HEAVY NUCLEI

Isotope	Reference	$\overline{ u}$ a	Γ_2	$\sigma_{ u}^2$
		Spontaneous fission		
²³⁸ U	[22]	1.98 ± 0.03	0.699 ± 0.038	0.80 ± 0.15
²³⁶ Pu	[23]	2.12 ± 0.13	0.809 ± 0.045	1.26 ± 0.20
²³⁸ Pu	[23]	2.21 ± 0.07	0.812 ± 0.010	1.29 ± 0.05
²⁴⁰ Pu	[23-30]	2.14 ± 0.01	0.822 ± 0.002	1.32 ± 0.01
²⁴² Pu	[23, 27, 28]	2.12 ± 0.01	0.821 ± 0.002	1.31 ± 0.01
²⁴² Cm	[23]	2.51 ± 0.06	0.793 ± 0.004	1.21 ± 0.03
²⁴⁴ Cm	[21, 23, 25]	2.69 ± 0.01	0.798 ± 0.007	1.23 ± 0.05
²⁴⁶ Cm	[21, 31]	2.94 ± 0.03	0.812 ± 0.003	1.31 ± 0.02
²⁴⁸ Cm	[28, 31]	3.10 ± 0.01	0.820 ± 0.001	1.37 ± 0.01
²⁴⁶ Cf	[32]	3.14 ± 0.09	0.850 ± 0.031	1.66 ± 0.31
²⁵⁰ Cf	[33]	3.53 ± 0.02	0.839 ± 0.002	1.52 ± 0.02
²⁵² Cf	[23,25,26,31,33-38]	3.735 ± 0.014	0.845 ± 0.001	1.57 ± 0.01
²⁵⁴ Fm	[39, 40]	3.96 ± 0.14	0.843 ± 0.012	1.50 ± 0.20
²⁵⁶ Fm	[40, 41]	3.73 ± 0.18	0.897 ± 0.047	2.30 ± 0.65
²⁵⁷ Fm	[36, 40, 42]	3.77 ± 0.02	0.910 ± 0.002	2.49 ± 0.06
²⁵² 102	[13]	4.15 ± 0.30	0.991 ± 0.075	4.0 ± 1.3
		Thermal neutron fission		
²³⁴ U	[34]	2.47 ± 0.01	0.793 ± 0.002	1.21 ± 0.01
²³⁶ U	[34]	2.39 ± 0.01	0.798 ± 0.002	1.24 ± 0.01
²⁴⁰ Pu	[34]	2.86 ± 0.01	0.822 ± 0.002	1.40 ± 0.01
²⁴² Pu	[34]	2.91 ± 0.01	0.819 ± 0.001	1.38 ± 0.01

a Data from Refs [13, 21-43].

With these assumptions, Terrell showed [19, 20] that the distribution P_{ν} in cumulative form is described approximately by the 'Gaussian' distribution

$$\sum_{n=0}^{n=\nu} P_n = (2\pi)^{-1/2} \int_{-\infty}^{(\nu - \overline{\nu} + 1/2 + b)/\sigma} \exp(-t^2/2) dt$$
 (2)

where b $< 10^{-2}$ is a constant introduced for the sake of obtaining better agreement with experiment and $\sigma \cong \sqrt{\sigma_{\nu}^2 - 1/12}$. It was found that almost all the experimental data on which Terrell had based himself could be satisfactorily described by distribution (2) with $\sigma = 1.08$; the one exception was 252 Cf(SF), for which it was necessary to take $\sigma = 1.21$. The estimate $E_0 = 6.7 \pm 0.7$ MeV was

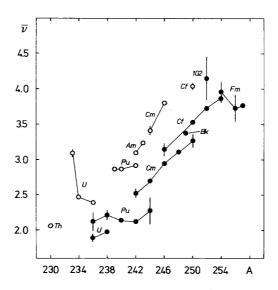


FIG.1. Dependence of the average number of neutrons per fission event $\overline{\nu}$ on Z and A of the fissioning nucleus: closed circles – spontaneous fission; open circles – thermal neutron fission. The continuous lines joining the experimental points indicate that the isotopes in question are of the same element.

found to be in reasonable agreement with the experimental data on the rate of change in $\overline{\nu}$ with increasing energy of the neutrons inducing fission, $d\overline{\nu}/dE_n\cong 1/E_0$, and the value of the variance of the distribution of the total fission fragment excitation energy, obtained in the form σ^2 E_0^2 , was found to be in agreement with the results of direct measurements of the variance of the distribution of the total fission fragment kinetic energy.

The experimental data on the distribution of the neutron number, P_{ν} , obtained over the past few years show that it is possible to refine many of Terrell's assumptions [19, 20], both essentially and quantitatively. This applies particularly to the (Z,A) dependence of the distribution of the number of neutrons and to the assertion that σ^2 E_0^2 is the variance of the total fission fragment excitation energy. A step in this direction was taken by us in Ref. [21], where the systematics of the values of σ_{ν}^2 was presented. Here we supplement this with the latest experimental data of importance for clarifying the (Z,A) dependence of σ_{ν}^2 and analyse this dependence in greater detail.

The experimental data on the variance of the number of neutrons in spontaneous fission and thermal neutron fission published so far are presented in Table I. The starting point in determining σ_p^2 was a quantity Γ_2 not dependent on the efficiency of neutron detection:

$$\Gamma_2 = \frac{\langle \nu^2 \rangle - \overline{\nu}}{\overline{\nu}^2} = \frac{\langle \mathbf{n}^2 \rangle - \overline{\mathbf{n}}}{\overline{\mathbf{n}}^2}$$

where \overline{n} is the mean number of neutrons detected per fission event and $\langle \nu^2 \rangle = \sum_{\nu=1}^{\nu=\nu_{\rm max.}} \nu^2 P_{\nu}$ (and similarly for $\langle n^2 \rangle$). For a given value of $\overline{\nu}$ the quantity Γ_2 is uniquely related with σ_{ν}^2 by the relation

$$\sigma_{\overline{\nu}}^2 = \overline{\nu} - \overline{\nu}^2 (1 - \Gamma_2) \tag{3}$$

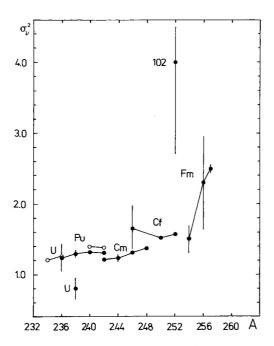


FIG.2. Dependence of the variance of the total number of neutrons σ_{ν}^2 on Z and A of the fissioning nucleus (see caption to Fig. 1 for notation).

Thus, Γ_2 serves as a measure of the deviation of the distribution P_{ν} from a Poisson distribution for which $\Gamma_2 \equiv 1$. Where there were several Γ_2 values for the same nucleus in the literature, they were averaged with allowance for the errors in the individual results and the variance was determined on the basis of the weighted mean value of Γ_2 . The values of $\overline{\nu}$ and Γ_2 used by us in determining σ_{ν}^2 and references to the original literature are given in Table I.

The (Z, A) dependences of $\overline{\nu}$ and $\sigma_{\overline{\nu}}^2$ for spontaneous fission and thermal neutron fission are shown in Figs 1 and 2, respectively. In contrast to the conclusions of Terrell [19, 20], the weak dependence of $\sigma_{\overline{\nu}}^2$ on Z and A for plutonium and curium isotopes is more an exception to the rule than the high value of $\sigma_{\overline{\nu}}^2$ for 252 Cf. It can be seen that, when one goes from 238 U to 252 102, the variance of the number of neutrons increases by a factor of 5, which significantly exceeds the scale of change in $\overline{\nu}$. Comparison of Fig. 1 with Fig. 2 shows that the behaviour of $\sigma_{\overline{\nu}}^2$ as the initial parameters of the fissioning nucleus change differs substantially from that of $\overline{\nu}$. On the whole, from the data in Table I and Fig. 2 it may be concluded that the variance of the number of neutrons depends to a much greater extent on Z than on the total number of nucleons in the fissioning nucleus. Conversely, the A dependence of $\overline{\nu}$ is just as strong as the Z dependence if one considers all nuclei with $90 \le Z \le 102$ investigated experimentally.

In order to trace more clearly the Z dependence of the variance of the number of neutrons, we have averaged the values of σ_p^2 with respect to A for each Z, treating spontaneous and induced fission separately; the result is presented in Fig. 3 (the continuous curves joining the experimental points only emphasizes the trend of σ_p^2 changing with Z). The dependence on Z is, on the whole, extremely sensitive, and it may well be of an exponential nature; only in the Z = 94–96 region

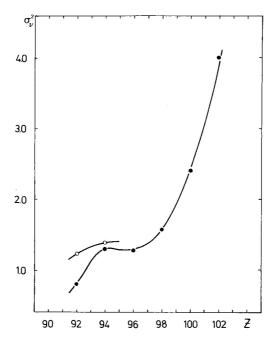


FIG.3. Z dependence of the variance of the total number of neutrons σ_v^2 : closed circles – spontaneous fission; open circles – thermal neutron fission.

an 'anomaly' is observed. Here the Z dependence of σ_{ν}^2 is weak and possibly not monotonic: the transition from Z = 94 to Z = 96 (spontaneous fission) is accompanied by a decrease in σ_{ν}^2 – a small one, but one that goes appreciably beyond the limits of the experimental errors (see Table I). Apparently, it is this 'anomaly' that is responsible for the opinion that the (Z, A, E*) dependence of σ_{ν}^2 is weak which has become established in the literature⁴ since Terrell's work was published [19, 20]. Unfortunately, the data on which Terrell based himself were confined to the Z = 92–96 region, with the exception of ²⁵²Cf, and the large experimental errors in the data meant that there was no point in distinguishing between spontaneous and induced fission. The conclusion regarding the weak (Z, A, E*) dependence of the distribution of the number of neutrons P_{ν} was thus the only one arrived at. Now it can be seen from Figs 1 and 2 that σ_{ν}^2 increases in the case of induced fission as compared with spontaneous fission. The increase in σ_{ν}^2 for Z = 94 is again small, but it is definitely established by the precision measurements of Boldeman [27, 28, 34].

As the experimental data on σ_{ν}^2 are still confined to spontaneous fission (with the exception of the four cases of thermal neutron fission of uranium and plutonium isotopes), it would be interesting to obtain quantitative information about the variance of the number of neutrons in the fission of nuclei with different Z values – for example, 235 U, 239 Pu and 249 Cf – by neutrons with energies ranging from zero to 14 MeV. This would permit reliable determination of the dependence of σ_{ν}^2 on the excitation energy of the fissioning nucleus. Apart from some fragmentary data that cannot be compared quantitatively [19, 25, 35, 44], there is no such information available [21].

⁴ Including quite recent reviews – see, for example, VANDENBOSCH, R., HUIZENGA, J.R., Nuclear Fission, Academic Press, New York and London (1973).

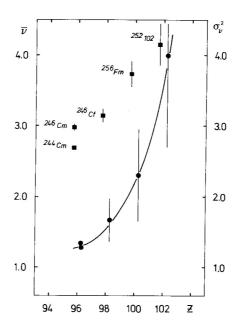


FIG.3a. Experimental data on $\overline{\nu}$ and σ_{ν}^2 for a number of heavy nuclei determined at the Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research [13, 21, 32, 40, 41].

Completing the discussion of the systematics of the values of σ_p^2 , we would like to point out that the extremely high value of σ_p^2 for spontaneous fission of $^{252}102$ that we have recently discovered [13], is very much in line with the tendency indicated by the results of the more exact measurements for nuclei with Z=96, 98 and 100. (The experimental values of $\overline{\nu}$ and σ_p^2 for a number of isotopes with Z=96-102 obtained at the Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research are presented in Fig. 3a.) Extrapolation of the Z dependence of the variance of the number of neutrons presented in Fig. 3 to the case of Z=104 gives $\sigma_p^2\approx 6$. Further extrapolation is, in our opinion, premature.

3. CORRELATION BETWEEN THE VARIANCE OF THE TOTAL NUMBER OF NEUTRONS AND THE STRUCTURE OF THE MASS DISTRIBUTION OF FISSION FRAGMENTS

The study of the experimental data on the variance of the number of neutrons presented in Fig. 2 and Table I shows that the greatest changes in σ_{ν}^2 are observed where the change in the mass distribution of the fragments is greatest.

It is well known that low-energy fission of heavy nuclei is very asymmetric; the mass distribution of the fission fragments is represented by a double-peaked curve with an extremely pronounced deep valley between the peaks. This curve is fairly complex, and its shape cannot be characterized by any single parameter. For a general qualitative description of the mass distribution, several quantities are used: the peak-to-valley ratio R, the asymmetry parameter χ characterizing the distance between the peaks, the width or variance of the peak σ_A^2 , etc.

TABLE II. CORRELATION BETWEEN THE VARIANCE OF THE TOTAL NUMBER OF NEUTRONS AND THE CHARACTERISTICS OF THE FISSION FRAGMENT MASS DISTRIBUTION

Isotope	$\sigma_{ u}^2$	R ^a	$\chi = A_2/A_1^{a}$	σ_{A}^{a}
		Spontaneou	s fission	
²³⁸ U	0.80 ± 0.15	> 600	1.48	12.8
²⁴⁰ Pu	1.32 ± 0.01	> 250	1.38	13
²⁴² Cm	1.21 ± 0.03	> 800	1.35	12
²⁴⁴ Cm	1.23 ± 0.05	≥ 5700	1.35	12
²⁵² Cf	1.57 ± 0.01	≥ 600	1.32	15
²⁵⁴ Fm	1.50 ± 0.20	60	1.30	_
²⁵⁶ Fm	2.30 ± 0.65	12	1.26	16
²⁵⁷ Fm	2.49 ± 0.06	≈ 1.5	-	-
		Thermal neu	tron fission	
²³⁴ U	1.21 ± 0.01	440	1.48	14.6
^{236}U	1.24 ± 0.01	620	1.45	15.0
²⁴⁰ Pu	1.40 ± 0.01	150	1.40	15.3
²⁴² Pu	1.38 ± 0.01	230	1.38	14.5

^a Radiochemical data from Refs [45-48].

The mass asymmetry of fragments is most pronounced in the low-energy fission of nuclei belonging to the region from U to Cf. Qualitatively, the mass distributions of the fission fragments in this region do not change very much, in keeping with the slight changes in the value of σ_{ν}^2 . At the same time, some quantitative changes in the parameters of the mass distribution curve are observed in this region also: for example, despite the high value of the peak-to-valley ratio for the spontaneous fission of 252 Cf and 240 Pu (R \geq 650 and R \geq 250 respectively), the peaks of the light and of the heavy fission fragments on the mass distribution curve for 252 Cf are significantly wider and closer together than in the case of 240 Pu; the values of σ_{ν}^2 for these two cases also differ appreciably. The mass distributions for 242 Cm(SF) and 244 Cm(SF) are in turn somewhat more asymmetric than in the case of 240 Pu, as pointed out in Refs [2, 45 and 46]. At the same time in these two cases also a difference in the value of σ_{ν}^2 exceeding the limits of the experimental errors is observed. The substantial difference in the fragment mass distributions for 235 U(n, f) and 238 U(SF) is matched by a significant difference in the variance of the number of neutrons, etc.

The strong increase in σ_{ν}^2 and the marked change toward symmetric mass distribution as one goes from Cf isotopes to Fm isotopes have been reliably established; the consistency of these changes hardly requires comment.

On the basis of reasoning along a similar line the idea of a correlation between the variance of the total number of neutrons and the fission fragment yield in the symmetric mass region was proposed in Ref. [21]. This correlation is brought out by Table II and demonstrated by the fact that the highest values of σ_p^2 correspond to the cases with the most pronounced symmetric fragment yields. Without repeating the factual material set forth in Ref. [21], here we should like to comment on the possible consequences of this correlation.

Using definition (1), it is not difficult to show that, in the general case,

$$\sigma_{\nu}^{2} = \sum_{\mathbf{A}_{1}} \left\{ \sigma_{\nu}^{2}(\mathbf{A}_{1}, \mathbf{A}_{2}) + [\overline{\nu} - \overline{\nu}(\mathbf{A}_{1}, \mathbf{A}_{2})]^{2} \right\} Y(\mathbf{A}_{1})$$
 (4)

where $\sigma_{\nu}^2(A_1,A_2)$ is the variance of the total number of neutrons emitted by two complementary fragments A_1 and A_2 (where $A_1+A_2=A$ is the mass of the fissioning nucleus); $\overline{\nu}(A_1,A_2)=\overline{\nu}(A_1)+\overline{\nu}(A_2)$ is the average number of neutrons emitted by the two complementary fragments for a given ratio of their masses; and $Y(A_1)$ is the initial (before neutron emission) mass distribution ($\sum_{A_1} Y(A_1)=1$).

As σ_v^2 increases with the yield of the symmetric fragments, the contribution of which to the absolute mass yield is small, the quantity

$$\Sigma_{\nu}^{2}(A_{1}, A_{2}) = \sigma_{\nu}^{2}(A_{1}, A_{2}) + [\overline{\nu} - \overline{\nu}(A_{1}, A_{2})]^{2}$$
(5)

must be substantially greater for $A_1 \approx A_2$ than its mean value with respect to the mass distribution, i.e. it must significantly exceed σ_p^2 . The variance $\sigma_p^2(A_1, A_2)$ may in turn depend on both the properties of the individual fragments A_1 and A_2 and on the sharing of the total excitation energy between them. On the assumption that the emission of a neutron by one fragment is statistically independent of the quantum state of the complementary fragment,

$$\sigma_{\nu}^{2}(A_{1}, A_{2}) = \sigma_{\nu}^{2}(A_{1}) + \sigma_{\nu}^{2}(A_{2}) + 2C_{\nu}(A_{1}, A_{2})$$
(6)

where $\sigma_{\nu}^{2}(A_{f})$ is the variance of the number of neutrons emitted by one fragment with mass A_{f} , (f = 1, 2), and $C_{\nu}(A_{1}, A_{2})$ is the covariance of the two distributions of the number of neutrons for the two complementary fragments. For a Gaussian distribution

$$C_{\nu}(A_1, A_2) = \rho(A_1, A_2) \cdot \sigma_{\nu}(A_1) \cdot \sigma_{\nu}(A_2)$$
 and $|\rho(A_1, A_2)| \le 1$

In Ref. [21] the correlation between the variance of the number of neutrons and the structure of the fission fragment mass distribution was analysed for the case $\overline{\nu}(A_1, A_2) = \overline{\nu} = \text{constant}$, which inevitably led to a high value of $\sigma_{\nu}^2(A_1, A_2)$ in the symmetric region. The condition $\overline{\nu}(A_1, A_2) = \text{constant}$ is fulfilled fairly well for $^{252}\text{Cf}(SF)$; however, in the general case, and particularly in the region of symmetric and also strongly asymmetric fission, $\overline{\nu}(A_1, A_2)$ may substantially differ from $\overline{\nu}$. We know [49, 50] that for $^{235}\text{U}(n, f)$, for example, the average number of neutrons $\overline{\nu}(A_1, A_2)$ reaches about 4.5 for $A_1 \approx A_2$, which is significantly greater than $\overline{\nu} = 2.4$, whereas for $^{257}\text{Fm}(SF)$ one has the opposite situation, with $\overline{\nu}(A_1, A_2) \approx 1$ in the symmetric region compared with $\overline{\nu} = 3.77$ [42]. Thus, the second term in the formula (5), which is always positive, can, in principle, increase $\Sigma_{\nu}^2(A_1, A_2)$ in the symmetric region. This fact reduces the requirements regarding the value of the variance of the number of neutrons $\sigma_{\nu}^2(A_1, A_2)$ in the symmetric region and makes the correlation of σ_{ν}^2 with the symmetric mass yield somewhat clearer.

Returning to formula (4), we would like to point out that the value of $\sum_{A_1} [\overline{\nu} - \overline{\nu}(A_1, A_2)]^2 \cdot Y(A_1)$ can be calculated fairly accurately on the basis of experimental data, at least for the fission of uranium and plutonium isotopes by thermal neutrons and for the spontaneous fission of ²⁵²Cf. This provides information about $\langle \sigma_{\nu}^2(A_1, A_2) \rangle$, the mean – with respect to mass distribution – variance of the number of neutrons $\sigma_{\nu}^2(A_1, A_2)$. On the other hand, the value of $\langle \sigma_{\nu}^2(A_1, A_2) \rangle$, which is not

burdened by a trivial addition due to the dependence of $\overline{\nu}(A_1, A_2) = \overline{\nu}(A_1) + \overline{\nu}(A_2)$ on the fragment mass ratio, is linked more directly than σ_{ν}^2 with the partition of the total excitation energy between the fragments and the mechanism of their de-excitation; just its dependence on the initial parameters of the fissioning nucleus would be of very great interest.

Such calculations have been performed by us for 235 U(n, f) and 256 Fm(SF). In the case of 235 U(n, f), we have used data on the dependence of $\overline{\nu}(A_1, A_2)$ from Refs [50–52]; in the case of 256 Fm(SF), the dependence of $\overline{\nu}(A_1, A_2)$ and the fragment mass distribution were taken from Ref. [48]. It was found that, in the 235 U(n, f) case, the value of $\sum_{A_1} [\overline{\nu} - \overline{\nu}(A_1, A_2)]^2 \cdot Y(A_1)$ is about 0.02, or 1.7% of σ_{ν}^2 ; in the 256 Fm case, it is about 0.19, or 8% of σ_{ν}^2 . Thus, the absolute value of $\sum_{A_1} [\overline{\nu} - \overline{\nu}(A_1, A_2)]^2 \cdot Y(A_1)$ is low and in the first approximation one can probably neglect it, assuming A_1

$$\sigma_{\nu}^{2} \approx \langle \sigma_{\nu}^{2}(\mathbf{A}_{1}, \mathbf{A}_{2}) \rangle = \sum_{\mathbf{A}_{1}} \sigma_{\nu}^{2}(\mathbf{A}_{1}, \mathbf{A}_{2}) \cdot \mathbf{Y}(\mathbf{A}_{1})$$
 (7)

For ²⁵²Cf (SF) and the neighbouring nuclei this approximation is even more justified.

Ideally, most detailed information is to be provided by direct measurements of the variance of the number of neutrons for fragments with fixed masses and kinetic energies. However, such experiments are very difficult; to obtain results, one requires a great deal of statistical data and one must introduce into the experimental data many corrections that are small in absolute value but complex and poorly defined. The variance of the number of neutrons for fragments of fixed masses has been measured directly only for 252 Cf (SF) [53, 54], and the range of nuclei investigated in that way can hardly be substantially extended, especially in the direction of heavier nuclei. An excellent review of these experiments and an analysis of the results are presented in Ref. [55]. Here we would like to call attention only to the most important experimental fact [53]: in the spontaneous fission of 252 Cf, the total excitation energy is distributed between the two complementary fragments in an uncorrelated manner — i.e. in the formula (6), $C_{\nu}(A_1, A_2) \approx 0$ for virtually all fragment mass ratios.

By analysing the values of σ_p^2 and determining their dependence on the initial parameters of the fissioning nucleus (Z, A and E*) and by ascertaining the correlations between σ_p^2 and the other fission characteristics for a wide range of Z and A values of the fissioning nucleus, it is possible, we think, to obtain information about the fission mechanism that considerably supplements results of detailed investigations of the variance of the number of neutrons for a particular nucleus taken individually.

The correlation between the variance of the total number of neutrons and the structure of the fragment mass distribution should not be regarded as an exhaustive explanation for the variations of σ_{ν}^2 with changing Z, A and E* of the fissioning nucleus. There may also be other factors determining the variations of σ_{ν}^2 — for example, a statistical type of dependence of σ_{ν}^2 on the mean excitation energy $(\bar{\nu})$ or on the mean initial temperature of the primary fission fragments. The correlation proposed by us emphasizes only one of the many complex interrelationships between the various fission characteristics. The reasons for this correlation are still not quite clear. On the other hand, one of the consequences of such a correlation (if it really exists and is not fortuitous) may be a significant increase in the variance of the number of neutrons in the symmetric fission region. Existing experimental data on the direct measurement of the variance of the number of neutrons for fragments of fixed masses do not preclude this possibility: according to the data of Signarbieux et al. [53] on 252 Cf, for example, σ_{ν}^2 (A₁, A₂) \approx 3 for A₁ = A₂ = 126, whereas σ_{ν}^2 = 1.57 (see Figs 2 and 4 in Ref. [53]).

Interesting results were recently obtained at Los Alamos [33] by measuring the distribution of the number of neutrons P_{ν} as a function of the total fragment kinetic energy E_k for the spontaneous

TABLE III. DEPENDENCE OF THE VARIANCE OF THE NUMBER OF NEUTRONS ON THE TOTAL FRAGMENT KINETIC ENERGY^a

	E _k (MeV)	Ēķ	>210	190 210	170 – 190	150 170	130 – 150
	<u>ر</u> مه	3.53 ± 0.02	1.70 ± 0.04	2.71 ± 0.01	3.77 ± 0.08	4.50 ± 0.02	4.06 ± 0.04
$^{250}{ m Cf}$, 63 9n	1.52 ± 0.02 0.839 ± 0.002	0.53 ± 0.07 0.595 ± 0.014	0.85 ± 0.02 0.747 ± 0.003	1.07 ± 0.02 0.810 ± 0.002	14.0% 1.57 ± 0.04 0.855 ± 0.002	3.3% 1.82 ± 0.10 0.864 ± 0.006
253	, A	3.735 ± 0.014 100%	2.16 ± 0.04 2.6%	3.13 ± 0.01 34.0%	4.07 ± 0.01 49.3%	4.98 ± 0.03 12.5%	4.62 ± 0.08 1.6%
5	$\Gamma_2^{\sigma_p^2}$	1.57 ± 0.01 0.845 ± 0.001	0.81 ± 0.03 0.711 ± 0.006	1.01 ± 0.03 0.784 ± 0.003	1.21 ± 0.03 0.827 ± 0.002	1.45 \pm 0.06 0.858 \pm 0.003	2.05 ± 0.20 0.880 ± 0.009
E _k (MeV)	E _k (MeV)	Bĸ	> 240	220 – 240	200 – 220	180 – 200	160 – 180
736	5 X	3.77 ± 0.02 100%	0.93 ± 0.09 4.6%	2.42 ± 0.06 14.3%	3.66 ± 0.05 32.5%	4.54 ± 0.05 32.3%	4.86 ± 0.08
m L	σ_{V}^{2}	2.49 ± 0.06 0.910 ± 0.002	0.93 ± 0.20 1.000 ± 0.230	1.30 \pm 0.15 0.809 \pm 0.026	1.55 \pm 0.11 0.842 \pm 0.008	1.83 \pm 0.14 0.869 \pm 0.007	2.29 ± 0.23 0.891 ± 0.010

Results from Ref. [33].
 Fraction of fission events with the total fragment kinetic energy in the range in question.
 Calculated by the author from experimental data presented in Ref. [33].

fission of 250 Cf, 252 Cf and 257 Fm; they are presented in Table III. These results cannot be used, however, for direct determination of the variance of the number of neutrons in the symmetric region or direct verification of the assumption concerning the correlation between σ_p^2 and the yield of symmetric fragments: fission modes with markedly different fragment mass ratios may contribute to the same experimental interval of E_k (in Ref. [33] the energy range of these intervals is 20 MeV). Moreover, if one considers not the variance but the relative width of the distribution of the number of neutrons.

$$\Gamma_2 = \frac{\langle v^2 \rangle - \overline{v}}{\overline{v}^2} = 1 - \frac{\overline{v} - \sigma_v^2}{\overline{v}^2} \tag{8}$$

it can be seen from the data in Ref. [33] for 257 Fm (SF) that Γ_2 = 1 for $E_k > 240$ MeV (symmetric region), whereas Γ_2 = 0.843 – for example – in the range $220 \le E_k \le 200$ MeV (see Table III).

In our opinion, the assumption concerning the existence of a correlation between the variance of the number of neutrons and the structure of the fragment mass distribution, and also the reasons for and consequences of this correlation, can be verified directly only through direct, quantitatively reliable measurements of the variance of the total number of neutrons as a function of the fission fragment mass ratio — especially in the symmetric fission region — for several fissioning systems differing appreciably in their σ_{ν}^2 values.

4. VARIANCE OF THE TOTAL KINETIC ENERGY OF FISSION FRAGMENTS

Let us now turn to the (Z,A) dependence of the variance of the total fragment kinetic energy $\sigma_{E_k}^2$ in spontaneous and thermal neutron fission. By definition

$$\sigma_{\overline{E}_k}^2 = \int_{(E_k)} (E_k - \overline{E}_k)^2 \cdot N(E_k) \cdot dE_k$$
(9)

where $\overline{E}_k = \int\limits_{(E_k)} E_k \cdot N(E_k) \, dE_k$ is the mean total fragment kinetic energy and $N(E)_k$ is the normalized, directly measured distribution of the total kinetic energy. In the first approximation, $N(E_k)$ is described fairly well by a Gaussian distribution; therefore for estimating $\sigma^2_{E_k}$, use is often made of the width of the experimental $N(E_k)$ curve at half-maximum (for a Gaussian distribution, it is $2.35 \, \sigma_{E_k}$).

The list of works on the investigation of fragment kinetic energy distributions contains dozens of references. However, most of them are of only limited interest for us. This is due to the following: the differences in the methods of energy calibration and of introducing corrections for neutron emission from fragments, the non-uniform nature of the sources, the differences in energy resolution and in the way of presenting the experimental data; all this makes it very difficult or virtually impossible to make quantitative comparisons. In comparing the systematics of the values of $\sigma_{\rm E_k}^2$ we therefore confine ourselves to well-known works [48, 56–69] performed mainly during the past decade and containing quantitative information on $\sigma_{\rm E_k}^2$ obtained by a single method, as a rule, for several cases of fission at the same time. Experimental values of $\sigma_{\rm E_k}^2$ are presented in Table IV for 21 cases of low-energy fission of heavy nuclei. Being interested primarily in the major features of the changes in $\sigma_{\rm E_k}^2$, we have not attempted to average the experimental data for cases of multiple measurements, and we have chosen a somewhat different way of constructing the systematics.

TABLE IV. VARIANCE OF THE TOTAL FRAGMENT KINETIC ENERGY IN FISSION OF HEAVY NUCLEI

Isotope	Reference	E _k (MeV)	$\sigma_{\rm E_k}^2$ (MeV ²)
	Spontane	ous fission	
²⁴⁴ Cm	[57, 58]	183.7	122.6
²⁴⁶ Cm	[48]	183.9	112.4
²⁴⁸ Cm	[48]	182.2	110.3
²⁵⁰ Cm	[59]	179.8ª	112.0
²⁵⁰ Cf	[48]	187.0	127.7
²⁵² Cf	[48]	185.9	134.6
²⁵⁴ Cf	[48]	186.9	139.2
²⁵³ Es	[61]	188.0	179.6
²⁵⁴ Fm	[61]	189.0	162.6
²⁵⁶ Fm	[48]	197.9	207,4
²⁵⁷ Fm	[62]	198.0	197.5
	Thermal neu	tron fission	
²³⁰ Th	[48]	163.6	67.2
²³⁴ U	[48]	172.1	98.0
²³⁶ U	[48]	171.8	106.1
²⁴⁰ Pu	[48]	177.1	132.3
²⁴² Pu	[56]	179.6	121.9
²⁴⁶ Cm	[48]	184.2	136.9
²⁵⁰ Cf	[48]	189.1	169.0
²⁵² Cf	[60]	182.1 ^b	205.0 ^b
²⁵⁵ Es	[48]	194.3	252.8
²⁵⁶ Fm	[60]	192.5 ^b	289.9 ^b

^a Most probable value of E_k .

We took data from Ref. [48], where the distributions of the total fragment kinetic energy were investigated for 13 fissioning systems ranging from $^{229}\mathrm{Th}\,(n,\,f)$ to $^{256}\mathrm{Fm}\,(SF).$ At the same time, we have included in Table IV values of $\sigma_{E_k}^2$ for eight other fissioning systems; these values were taken from Refs [56–62] and were introduced into Table IV after appropriate renormalization with allowance for the difference in $\sigma_{E_k}^2$ for $^{252}\mathrm{Cf}\,(SF),$ which served as a standard in the measurements performed in Refs [56–62]. It is hoped that this renormalization method takes into account most of the differences associated with the experimental procedure $^5.$

Values relating to fragments after neutron emission; the values of E_k are the most probable ones.

 $^{^5}$ The data in Refs [48 and 56-62] were obtained essentially in the same way - by recording the kinetic energies of two complementary fission fragments with Si(Au) surface-barrier detectors.

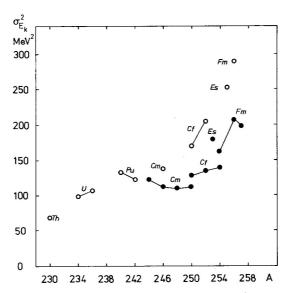


FIG.4. Dependence of the variance of the total fission fragment kinetic energy $\sigma_{E_k}^2$ on Z and A of the fissioning nucleus (see caption to Fig. 1 for notation).

Unfortunately, the accuracy of the experimental determination of $\sigma_{E_k}^2$ is not very high, being on the whole significantly less than the accuracy of the determination of $\sigma_{\mathcal{F}}^2$. It was therefore necessary to analyse in detail the quality of the data in Table IV. As the errors in the reported values of $\sigma_{E_k}^2$ are not indicated in any of the works (except Ref. [61]) from which the data in Table IV were taken, the only way of estimating them was to analyse the spread of the $\sigma_{E_k}^2$ values in cases of multiple measurements. Such an analysis of the experimental results from Refs [48 and 56-69] shows that on the average the spread of the absolute values of the quantities in question is 10-12%; it would seem reasonable to take this as the mean (for Table IV) error in the absolute values of $\sigma_{E_k}^2$. After renormalization and reducing all data to the single reference value $\sigma_{E_k}^2$ (252 Cf) = 135 MeV², the spread of the $\sigma_{E_k}^2$ values decreases by about a factor of two. It may then be assumed that on average (for Table IV) the accuracy of the relative values is not worse than 5-6%. In Table IV we also present values of the mean total fragment kinetic energy \overline{E}_k and references to the original literature.

The (Z, A) dependence of the variance of the total fission fragment kinetic energy is presented in Fig. 4. The scale of the changes is worth noting: as one moves from $^{230}{\rm Th}$ to $^{257}{\rm Fm}$ the variance increases by a factor of four, whereas the mean fragment kinetic energy changes by only 30%. Like the variance of the number of neutrons, $\sigma^2_{\rm E_k}$ depends much more strongly on the number of protons in the fissioning nucleus than on its mass number. Accordingly, we again average the values of $\sigma^2_{\rm E_k}$ with respect to A for each Z separately for spontaneous and induced fission. The result is shown in Fig. 5. It can be seen that both for spontaneous fission and for thermal neutron fission the dependence on Z is, on the whole, quite pronounced.

In the case of induced fission, weakening of the Z dependence is observed in the Z = 94-96 region, where it has a double kink. Unfortunately, information about $\sigma_{E_k}^2$ for the spontaneous fission of plutonium isotopes is not very reliable quantitatively. According to the results reported in

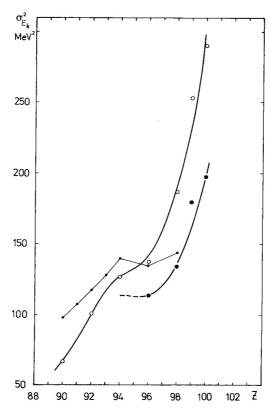


FIG.5. Z dependence of the variance of the total fission fragment kinetic energy $\sigma_{\rm Ek}^2$ (see caption to Fig. 3 for notation). The thin continuous line has been drawn through points taken from Ref. [73].

Refs [70, 71], the values of $\sigma_{E_k}^2$ for the induced and spontaneous fission of ²⁴²Pu [70] and ²⁴⁰Pu [71] differ by no more than 5–10 MeV², which clearly lies within the limits of the experimental errors. Ref. [72], on the other hand, gives for ²⁴⁰Pu (SF) an increase in $\sigma_{E_k}^2$ of 45 MeV² compared with ²³⁹Pu(n, f); however, this difference is based on the results of two experiments which were by no means identical from the methodological point of view. At the same time, comparison of the Z dependences of $\sigma_{E_k}^2$ and σ_{ν}^2 in Fig. 6 shows that the trend of the change in these quantities is virtually the same for both spontaneous and induced fission. It is therefore quite probable that the correct tendency – the constancy or even a very slight decrease of $\sigma_{E_k}^2$ as one moves from Z = 94 to Z = 96 and a very slight change as one goes from E* = 0 to E* = 6.4 MeV for Z = 94 — is given by measurements of the variance of the number of neutrons (see Table I and Figs 2 and 3); hence we construct the broken line in Fig. 5. Thus, analysis of all the experimental data available does not arouse doubts concerning the existence of 'anomalies' in the Z dependence of $\sigma_{E_k}^2$ for Z = 94. However, for a more detailed clarification of this dependence in the Z = 94–96 region one needs additional, quantitatively more reliable information about fragment kinetic energy distributions which will permit comparison with the data for ²⁵²Cf(SF) or ²³⁵U(n, f).

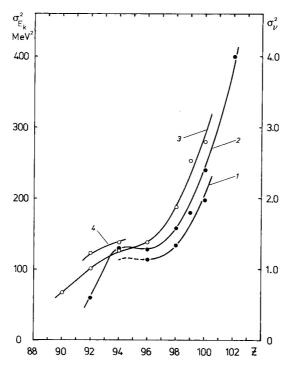


FIG.6. Comparison of the Z dependences of the variance of the total fragment kinetic energy $\sigma_{E_k}^2$ (curves 1 and 3) and the variance of the total number of fission neutrons σ_{ν}^2 (curves 2 and 4).

It should be noted that the Z dependence of the variance of the total fragment kinetic energy $\sigma_{E_k}^2$ in the range $90 \leqslant Z \leqslant 98$ has been analysed earlier [73] (see the thin continuous line in Fig. 5). The trend of the change of $\sigma_{E_k}^2$ in this Z range revealed by the data in Ref. [73] is very similar to that presented here. However, on the basis of the results for plutonium isotopes the authors of Ref. [73] maintained that a change of ≈ 6 MeV in excitation energy had virtually no effect on the value of $\sigma_{E_k}^2$ and did not separate the data for spontaneous fission, thermal neutron fission and fast neutron fission. It is probably this consideration that led to the 'levelling-off' of $\sigma_{E_k}^2$ for $Z \geqslant 94$. If one considers spontaneous and induced fission separately, on the other hand, the curves of the Z dependences of $\sigma_{E_k}^2$ for these two cases are clearly separated.

As the variance of the total fragment kinetic energy is an unusually strong and at the same time a fairly 'smooth' function of Z, it is worth examining its mathematical character more closely. An attempt at this has been made by us in Fig. 7(b), where the Z dependence of $\sigma_{E_k}^2$ is presented on a semi-logarithmic scale; the Z dependence of the variance of the number of neutrons is presented in the same form in Fig. 7(a). It follows from Fig. 7 that, except for the 'anomaly' region, all the experimental points for $\sigma_{E_k}^2$ and σ_{ν}^2 in the case of both spontaneous and induced fussion lie close to continuous straight lines drawn with exactly the same slope. This suggests that for heavy nuclei in the range considered by us $(90 \le Z \le 102)$ the dependence of $\sigma_{E_k}^2$ and σ_{ν}^2 on the number of protons of the fissioning nucleus is, on the whole, exponential. To be more precise, if our assumption is correct, with the exception of 93 < Z < 97

$$\sigma_{\rm E_k}^2 \sim a_{\rm E} \exp(bZ)$$
 (10a)

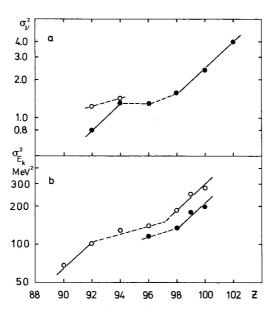


FIG. 7. Z dependence of σ_{ν}^{2} (a) and $\sigma_{\rm E_{k}}^{2}$ (b) on a semi-logarithmic scale.

and similarly

$$\sigma_{\nu}^2 \sim a_{\nu} \exp(bZ)$$
 (10b)

where b \approx 0.2, a_E and a_{ν} are constants.

5. MEAN VALUE OF THE VARIANCE OF THE TOTAL KINETIC ENERGY FOR INDIVIDUAL FISSION FRAGMENTS

In the preceding section it was shown that the variance of the total fragment kinetic energy $\sigma_{E_k}^2$ increases considerably as the number of protons in the fissionable nucleus increases. The value of $\sigma_{E_k}^2$ for a nucleus with a given Z, on the other hand, may be governed appreciably by the spread of the values for the average total kinetic energy $\overline{E}_k(A_1,A_2)$ for different pairs of fragments with differing mass ratios. It is therefore worth considering the extent to which the changes in $\sigma_{E_k}^2$ observed with changing Z, A and E* of a fissionable nucleus are associated with the average properties of the fragments and the extent to which they are associated with the dependence of the total kinetic energy of a pair of fragments on their mass ratio. Using expression (8), one can write

$$\sigma_{E_k}^2 = \sum_{A_1} \left\{ \sigma_{E_k}^2(A_1, A_2) + [\overline{E}_k(A_1, A_2) - \overline{E}_k]^2 \right\} \cdot Y(A_1)$$
 (11)

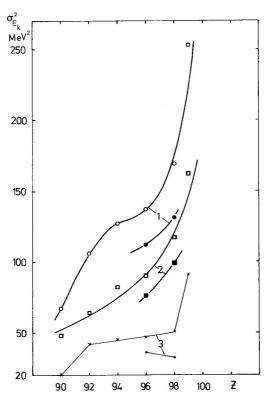


FIG.8. Z dependence of the mean value of the variance of the total kinetic energy for individual fission fragments $\langle \sigma_{\rm E_k}^2(A_1,A_2)\rangle$ (curve 2). Curves 1 and 3 – dependence of $\sigma_{\rm E_k}^2$ and $\langle \Delta \overline{E}_k^2(A_1,A_2)\rangle$ on Z of the fissioning nucleus. Open symbols – thermal neutron fission; solid symbols – spontaneous fission.

where $\sigma_{E_k}^2(A_1,A_2)$ is the variance of the distribution of the total kinetic energy in the fission of an initial nucleus into two fragments with masses A_1 and A_2 , and $\overline{E}_k(A_1,A_2)$ is the average value of the total kinetic energy of the pair of fragments with masses A_1 and A_2 . We introduce the notation

$$\langle \sigma_{E_k}^2(A_1, A_2) \rangle = \sum_{A_1} \sigma_{E_k}^2(A_1, A_2) \cdot Y(A_1)$$

$$\langle \Delta \overline{E}_k^2(A_1, A_2) \rangle = \sum_{A_1} [\overline{E}_k(A_1, A_2) - \overline{E}_k]^2 \cdot Y(A_1)$$

where the triangular brackets denote averaging of the quantity in question with respect to the mass distribution of the fragments – for example, $\overline{E}_k = \langle \overline{E}_k(A_1, A_2) \rangle$. The expression (11) can then be rewritten as follows:

$$\langle \sigma_{E_k}^2(A_1, A_2) \rangle = \sigma_{E_k}^2 - \langle \Delta \widetilde{E}_k^2(A_1, A_2) \rangle \tag{12}$$

As $\sigma_{E_k}^2$, \overline{E}_k and $\overline{E}_k(A_1,A_2)$, as well as the fragment mass distributions for many cases of fission, have been measured fairly reliably, using experimental data from Refs [48, 56, 66] and the formula (12), one can try to determine $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$ and trace its dependence on Z, A and excitation energy of the fissioning nucleus. Estimates have been made by us for the thermal neutron fission of 229 Th, 235 U, 239 Pu, 241 Pu, 245 Cm, 249 Cf and 254 Es and for the spontaneous fission of 246 Cm, 250 Cf and 252 Cf. The results of the calculations are presented in Fig. 8, where the Z dependences of $\sigma_{E_k}^2$ and $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$ for induced and spontaneous fission are compared; the Z dependence of $\langle \Delta \overline{E}_k^2(A_1,A_2) \rangle$ is also shown in the lower part of the figure.

It follows from Fig. 8 that $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$, like $\sigma_{E_k}^2$, undergoes substantial changes: as one moves from Z = 90 to Z = 99 it increases by more than a factor of 3. Calculations show that the dependence of $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$ on the mass number of the fissioning nucleus is weak. It is hard to say whether weakening of the Z dependence of $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$ is observed for Z = 94–96; owing to errors in its determination, this question cannot be answered directly.

At the same time, when one considers the Z dependence of $\langle \Delta \overline{E}_k^2(A_1,A_2) \rangle$ one sees that this quantity is virtually constant in the Z=92-98 region. This is understandable: neither the mass distributions nor the kinetic energy distributions $\overline{E}_k(A_1,A_2)$ exhibit substantial changes here as a function of the fragment mass ratio. On the other hand, a significant change in both these distributions is observed in the fission of nuclei with Z>98: the mass distributions become more symmetric and wider, and the fragment kinetic energy $\overline{E}_k(A_1,A_2)$ in the symmetric region begins to exceed substantially its mean value \overline{E}_k with respect to the mass distribution. We would also like to point out the considerable decrease in $\langle \Delta \overline{E}_k^2(A_1,A_2) \rangle$ for Z<92: here, the difference between $\sigma_{E_k}^2$ and $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$ is minimal, probably originating from the well-known changes in the mass and kinetic energy distributions of fission fragments as one approaches the region of actinium and radium.

The observed behaviour of $\langle \Delta \overline{E}_k^2(A_1, A_2) \rangle$ may make it somewhat easier to understand the reasons for the occurrence of 'anomalies' in the Z dependence of $\sigma_{E_k}^2$ in the region Z = 94-96: on $\langle \sigma_{E_k}^2(A_1, A_2) \rangle$, which increases as Z increases, is superposed an addition which is comparable in absolute value, but is constant in the Z = 92-98 region, associated with the change in the mean kinetic energy of a pair of fragments as their mass ratio changes.

Thus, the mean value — with respect to the mass distribution — of the variance of the total kinetic energy for individual fission fragments $\langle \sigma_{E_k}^2(A_1,A_2) \rangle$, like $\sigma_{E_k}^2$, is determined mainly by the charge on the fissionable nucleus and increases considerably when the latter increases. One can arrive at a similar conclusion by not considering the mean values of $\sigma_{E_k}^2(A_1,A_2)$ with respect to the mass distribution but the differential curves for $\sigma_{E_k}^2(A_1,A_2)$ as a function of the fragment mass ratio [48, 56–58, 63, 64, 66, 68, 69] for different fissioning systems.

6. BALANCE OF THE FLUCTUATIONS OF THE ENERGY RELEASED IN LOW-ENERGY FISSION OF HEAVY NUCLEI

The analysis of experimental data concerning the variance of the number of neutrons and the variance of the total fragment kinetic energy performed in the preceding sections shows that the nature of the change in these quantities when the nucleon composition of the fissioning nucleus varies is virtually the same. Let us analyse some of the consequences of this experimental fact.

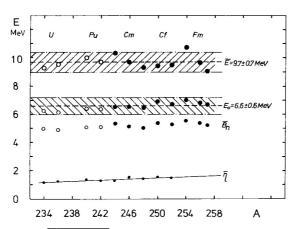


FIG.9. Dependence of $E = \sqrt{\sigma_{\rm Ek}^2/(\sigma_{\nu}^2 - 1/12)}$ on Z and A of the fissioning nucleus (for further details see main text).

Let Q be the total energy released in the fission process:

$$Q = E_k + E_x \tag{13}$$

It is well known that the quantities E_k and E_x entering into the energy balance (13) are anticorrelated, and Q experiences only relatively weak fluctuations, caused by fluctuations of the fission fragment masses and charges. This means that in the first approximation

$$\sigma_{\mathbf{Q}}^2 \cong \sigma_{\mathbf{E}_{\mathbf{k}}}^2 - \sigma_{\mathbf{E}_{\mathbf{x}}}^2 \tag{14}$$

where σ_Q^2 and $\sigma_{E_x}^2$ are the variance of the total amount of energy released in the reaction and the variance of the total fragment excitation energy respectively.

The rigorous correlation in the changes in $\sigma_{E_k}^2$ and σ_{P}^2 with respect to Z and A of the changing fissionable nucleus then means that the variance of the total number of neutrons σ_{P}^2 is fairly directly linked with the variance of the total excitation energy $\sigma_{E_x}^2$ of the fragments as a whole. Thus, the assumption made by us at the beginning of section 2 is fully justified. Further, taking into account Terrell's conclusion [19, 20] that $\sigma_{E_x}^2 = \sigma^2 E_0^2$, where $E_0 = 6.7 \pm 0.7$ MeV and $\sigma = \sqrt{\sigma_{P}^2 - 1/12}$, we analyse quantitatively the variations of

$$E = \sqrt{\frac{\sigma_{E_k}^2}{\sigma^2}} \tag{15}$$

on the basis of recent experimental data: from the results presented in Tables I and IV it is possible to determine values of E for 12 cases of fission.

We present the result in Fig. 9, from which it follows that E remains fairly constant at 9.7 ± 0.7 MeV on the average. This is significantly greater than the energy E_0 necessary, on the average, for the emission of one neutron from a fragment. For E_0 it is possible to write

$$E_0 = \overline{B}_n + \overline{\eta} \tag{16}$$

where \overline{B}_n is the energy necessary for separating a neutron from the fission fragments (neutron binding energy) averaged with respect to neutron cascade, fragment charge and fragment mass

distribution, and $\overline{\eta}$ is the mean energy of the fission neutron spectrum in the centre-of-mass system. Values of $\overline{\eta}$ have been measured with an accuracy of the order of 0.1 MeV for many nuclei in the $90 \le Z \le 98$ range [74]; they are shown in Fig. 9. We would like to point out, incidentally, that the variance of η is about 1.5 MeV². \overline{B}_n is not measured directly and therefore is more uncertain; however, it is possible to calculate it with a degree of accuracy sufficient for our purposes using tables of nuclear masses and experimental data on the mass and charge distributions of fission fragments. In Fig. 9 we present values of \overline{B}_n for those fissioning systems for which values of E are known; the values of \overline{B}_n were taken from Ref. [36] (the mass tables of Garvey and Kelson) and Ref. [61] (mass tables of Myers and Swiatecki), where they have been determined by the authors. It follows from Fig. 9 that the mean value of E_0 obtained in this way is 6.6 ± 0.7 MeV, which literally coincides with Terrell's estimate. However, his conclusions are clearly contradicted by the fact that

$$\sigma^2 E_0^2 \cong \sigma_{E_v}^2 < \sigma_{E_k}^2 \tag{17}$$

the strength of the inequality (17) being determined by the quantity

$$\sigma^{2}(E^{2}-E_{0}^{2})\approx 70 \text{ MeV}^{2}$$

The inequality (17) may have several causes. One of the most likely, in our opinion, is the fact that $\sigma^2 E_0^2$ is the variance only of that part of the total fragment excitation energy

$$E_{x} = E_{0}\nu + E_{\gamma}$$

– i.e. $E_0\nu$, which is removed through neutron emission. An appreciable contribution to the variance of the total excitation energy comes from the fluctuations of E_{γ} , the total energy removed by gamma-ray emission. On the basis of this assumption it is possible to write

$$\sigma_{E_x}^2 \cong \sigma^2 E_0^2 + \sigma_{E_\gamma}^2 \tag{18}$$

Secondly, when reconciling the fluctuation balance (14) it is necessary to take into consideration the variance of the total energy release, which may amount to several tens of MeV². Estimates of σ_Q^2 with semi-empirical data [56, 66] gives $\sigma_Q^2 = 20-30$ MeV² for the thermal neutron fission of uranium and plutonium isotopes. On the other hand, with fixed fragment masses the fluctuations of Q caused by charge variations are very small. Consequently, assuming $\sigma_Q^2(A_1, A_2) \approx 0$ it is possible to write

$$\sigma_{E_{\nu}}^{2}(A_{1}, A_{2}) \approx \sigma_{E_{\nu}}^{2}(A_{1}, A_{2})$$
 (19)

Then, taking the relations (14)–(19) into account, it is possible to obtain an estimate of the variance of the total gamma energy $\sigma_{E\gamma}^2$. For the thermal neutron fission of 235 U, 239 Pu and 241 Pu and for the spontaneous fission of 252 Cf such estimates lead, on the average, to $\sigma_{E\gamma}^2 \approx 30-35$ MeV². Naturally, to obtain a more rigorous estimate of $\sigma_{E\gamma}^2$ it is necessary to take into account other degrees of freedom as well: fluctuations of the total energy release associated with fragment charge variations, fluctuations of the energy E_0 expended for the emission of one neutron from a fragment, the possible existence of some small positive correlation between $E_0 \nu$ and E_γ [75], etc. Allowance for these factors will most likely lead to weakening of the inequality (17) and to the appearance of additional terms on the right-hand side of the balance (18), which may eventually change the estimate of $\sigma_{E\gamma}^2$ arrived at by us somewhat. It should be emphasized that this estimate was arrived at by taking the difference of two comparable and fairly large quantities, which were also burdened with experimental errors. It should therefore be considered rather as an upper limit and be used with care.

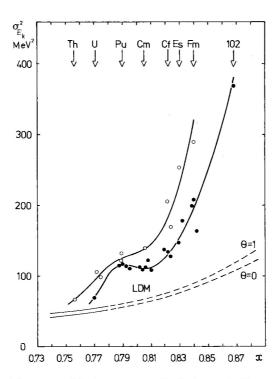


FIG.10. Dependence of the variance of the fragment energy distributions on fissility parameter x for spontaneous fission (solid circles) and thermal neutron fission (open circles). The curves in the lower part of the figure represent the results of calculations of $\sigma_{\rm E}^2$ based on the dynamic liquid-drop model [78].

From the literature we know of only one attempt to estimate the fluctuations of the rotational energy of the fragments or the associated E_{γ} ; with many assumptions and the fixed fragment mass ratio A_1/A_2 = 121/115, a value of $\sigma_{E_{\gamma}}^2 \approx 26$ MeV² was obtained for ²³⁵U(n, f) [76]. Thus, in the first approximation the balance of the fluctuations of the fragment energy in the

Thus, in the first approximation the balance of the fluctuations of the fragment energy in the fission reaction is reconciled. For a more careful study of the balance it would be interesting to make a direct experimental determination of $\sigma_{E_{\gamma}}^2$ or of the variance of the number of effective gammas, as is done for the distribution of the number of neutrons in fission. We know that the mean value of the total energy E_{γ} removed by fission gamma rays is virtually independent of A and Z of the fissioning nucleus in the entire region from U to Cf [10]. Given the similar nature of the (Z, A) dependences of $\sigma_{E_k}^2$ and σ_{ρ}^2 , one may assume that the variance of E_{γ} will probably depend on the fissioning nucleus A and Z only slightly.

7. DEPENDENCE OF THE VARIANCE OF THE ENERGY DISTRIBUTIONS OF FISSION FRAGMENTS ON THE FISSILITY PARAMETER AND THE EXCITATION ENERGY OF THE FISSIONING NUCLEUS: COMPARISON WITH THEORETICAL PREDICTIONS

As has been shown, the (Z, A) dependences of the variances of the total number of neutrons σ_{ν}^2 and the total fragment kinetic energy $\sigma_{E_k}^2$ are completely similar, and the ratio $\sigma_{E_k}^2/\sigma^2$ is virtually constant for all heavy nuclei. Hence, in the first approximation it is possible to consider changes

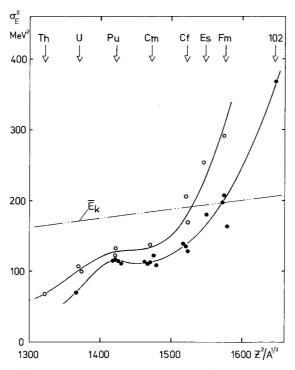


FIG.10a. Dependence of the variance of fragment energy distributions $\sigma_{\rm E}^2$ on the Coulomb parameter $Z^2/A^{1/3}$: open circles – thermal neutron fission; solid circles – spontaneous fission. The dash-dot line represents the dependence of the mean total fragment kinetic energy $\overline{E}_{\rm k}$ on $Z^2/A^{1/3}$.

in any one of these quantities (for example, $\sigma_{E_k}^2$) by supplementing where necessary its dependence on one or the other parameter by results obtained through measurements of the variance of the number of neutrons $(\sigma_{\nu}^2-1/12)$ $E^2\cong\sigma_{E_{\kappa}'}^2$. It is therefore possible to construct, for example, two curves representing the dependence of the variance of the fragment energy distribution $\sigma_E^2=\sigma_{E_{\kappa}}^2\cong\sigma_{E_{\kappa}'}^2$ on the nucleus fissility parameter x, one corresponding to a fissioning nucleus excitation energy $E^*=0$ (spontaneous fission) and the other to $E^*\cong 6.5$ MeV (thermal neutron fission). The dependence of σ_E^2 on the fissility parameter x — determined, as indicated above, through 20 measurements for $E^*=0$ and 11 measurements for $E^*\cong 6.5$ MeV in the range $0.75 \le x \le 0.87$ — is presented in Fig. 10, the fissility parameter x being chosen in the form [77-79]

$$x = \frac{Z^2/A}{50.88 \{1 - 1.7826 [(N - Z)/A]^2\}}$$
 (20)

It follows from Fig. 10 that a 15% increase in the fissility of nuclei leads to an increase of the fragment energy fluctuations several times for both spontaneous and induced fission. The dependence of σ_E^2 on x is very similar in character to the Z dependence of σ_P^2 (Fig. 3) and $\sigma_{E_k}^2$ (Fig. 5). On the

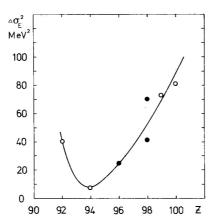


FIG.11. Increase in the variance of fragment energy distributions $\Delta o_{\rm L}^2$ associated with the transition from $E^*=0$ (spontaneous fission) to $E^*\cong 6.5$ MeV (thermal neutron fission) as a function of Z of the fissioning nucleus (see main text for further details).

whole it is similar to an exponent, and a weaker dependence is observed only in the x = 0.78-0.81 region. The continuous lines passing through the experimental points in Fig. 10 only emphasize the general trend of σ_E^2 changing with x. It can be seen that the experimental points are somewhat scattered in relation to these lines. This probably indicates weak dependence of σ_E^2 on A, which is not precluded by a switch from considering (Z, A) dependences to considering the dependence on x. The dependence of the variance of the fragment energy distributions σ_E^2 on the Coulomb parameter $Z^2/A^{1/3}$, constructed in the same way as the dependence of σ_E^2 on x, is shown in Fig. 10a.

It also follows from Figs 10 and 10a that, as the fissioning nucleus excitation energy increases from zero to 6.5 MeV, the variance of the total fragment energy distributions increases on the average by about 40%. To ascertain the extent of this increase more precisely, we determined the difference of $\Delta\sigma_E^2$ associated with the transition from $E^*=0$ to $E^*\cong 6.5$ MeV for different fissioning nuclei with fixed Z and A. The result is shown in Fig. 11. The closed circles denote cases of σ_E^2 for one and the same fissioning nucleus using data from one experiment; the open circles represent the result for $\Delta\sigma_E^2$ obtained on the basis of two different experiments or for cases where the fissioning nucleus for $E^*=0$ and $E^*\cong 6.5$ MeV do not have exactly the same mass. As Fig. 11 shows, $\Delta\sigma_E^2$ can assume a value from 40 to 80 MeV²; $\Delta\sigma_E^2$ probably increases with increasing Z of the fissionable nucleus. In the Z=94-96 region an 'anomaly' is again observed: for Z=94 the value of $\Delta\sigma_E^2$ tends to be at a minimum (5–10 MeV²).

Thus, the analysis of the available experimental data on the variance of fragment energy distributions in the low-energy fission of heavy nuclei throws light on the main features of the dependence of σ_E^2 on the fundamental parameters of the fissioning nucleus: the number of protons in the nucleus (Z), the total number of nucleons (A), the fissility parameter x and the excitation energy E*. We see that there are remarkable changes in σ_E^2 as a function of the number of protons in the fissioning nucleus and of its excitation energy, and these changes are of a regular nature. Altogether, the experimental data on σ_E^2 obtained from dozens of determinations of the multiplicity of prompt fission neutrons and from measurements of the distributions of the total kinetic energy of fragments are — despite certain contradictions — on the whole in good agreement. Analysis of the data suggests that the trends pointed out by us in the energy distribution variance are not a consequence of possible experimental errors; they go far beyond the limits of such errors, mainly

reflecting certain features characterizing the fission of heavy nuclei. In such a situation it is worth comparing the experimentally observed dependences of $\sigma_{\rm E}^2$ on the fissility parameter x and excitation energy E* with the theoretical predictions. However, there is at present no complete and consistent theory capable of describing quantitatively such complex fission process characteristics as fragment energy fluctuations. All we have are individual attempts to predict these quantities on the basis of very simple fission models.

Direct comparison of experimental data for the entire range of fissioning nucleus considered by us is possible only with predictions based on a very simple dynamic variant of the liquid-drop model [77–79]. Other calculations of fragment energy distributions for low excitation energies have been performed for only a few actual cases of fission — as a rule, for ²³⁵U(n, f) and ²⁵²Cf(SF) — with a fixed fragment mass ratio. It follows from sections 2 and 4 that the fragment energy distributions do not differ very much for these cases of fission or for the intermediate ones with respect to Z and A, so that it is not possible to verify effectively the predictions based on one or the other model.

Let us now turn to the comparison of experimental data with the predictions of the liquid-drop model [77–79]. In this model statistical equilibrium at the saddle point, which determines the initial conditions, is postulated and the development of the fissioning system from the saddle point to the scission point is studied. The general procedure for calculating the mass and energy distributions is to use the standard methods of statics, dynamics and statistical mechanics for the classical Hamiltonian of an idealized system. The Hamiltonian is formulated as the sum of the potential energy of an incompressible charged liquid drop, i.e. the surface and Coulomb energies, and the kinetic energy of the potential flow of an ideal non-viscous liquid. The results of the calculations of the variance of the fragment energy distributions σ_E^2 as a function of fissility parameter x obtained with the liquid-drop model [78] are shown in Fig. 10 for two values ($\theta = 0$ and $\theta = 1$ MeV) of the fissioning nucleus temperature at the saddle point; the temperature θ is determined by the expression

$$\mathbf{E}^{\mathbf{SP}} = \mathbf{a}\,\theta^2 - \theta \tag{21}$$

where E^{SP} is the internal excitation energy of the nucleus at the saddle point and a = A/8 (MeV⁻¹) is the level density parameter. As x increases beyond 0.78, the numerical accuracy of the calculations declines sharply, and this is indicated in Fig. 10 by broken lines.

Comparison of these predictions with the experimental data in Fig. 10 shows fairly clearly that the liquid-drop model does not enable one to describe — either quantitatively or qualitatively — the increase in the experimentally observed exponential-like curve σ_E^2 for heavy nuclei as the fissility parameter x increases or the substantial increase in σ_E^2 as the excitation energy of the fissionable nucleus increases. Thus ignoring the existence of shell effects in the fissioning nucleus, together with many other important features of the asymmetric low-energy fission of heavy nuclei, the liquid-drop model in the form in which it appears in Refs [77–79] does not enable one to explain even the existence of significant variations of the fragment kinetic energy and excitation energy.

An attempt to take shell effects into account within the framework of the dynamic liquid-drop model has been made by Hasse [81] in calculating the mass and energy distributions of fragments resulting from low-energy fission of heavy nuclei. In addition to the smooth liquid-drop part of the potential energy, a semi-phenomenological shell correction similar to the correction of Myers and Swiatecki [81] was introduced. The qualitatively most important result of Hasse's work is the reproduction of the asymmetry of the mass distribution in low-energy fission of heavy nuclei with a most probable heavy fragment mass of about 132; however, the quantitative description of the fragment mass distributions and kinetic energies remains unsatisfactory.

An alternative to the dynamic liquid-drop theory is the statistical theory of fission first proposed by Fong [82-85]. The basic assumption of this theory is that the development of a fissioning system from the saddle point to the scission point is such a slow process that statistical equilibrium is established at each moment along the path to scission, so that each individual quantum

state of the system has an equal a priori probability of materializing. The relative probability of observing one or the other mode of fission of the initial nucleus is therefore proportional to the density of the quantum states of the nuclear configuration at the moment immediately before scission. The condition for statistical equilibrium at the scission point enables one, in principle, to determine theoretically all the experimentally observed characteristics of the fission fragments and also to determine the dependence of these characteristics on Z, A, and the excitation energy of the fissionable nucleus.

Detailed formulas for the mass distributions, charges, kinetic energies and other characteristics of fission fragments have been derived by Fong on the basis of the statistical theory [85]. In particular, the following expression has been obtained for the width of the distribution of the total kinetic energy of fragments for a fixed ratio of their masses and charges⁶:

$$\Delta \cong \text{constant } (Z_1 \cdot Z_2)^{\frac{1}{2}} \left(\frac{E}{a_1 + a_2} \right)^{\frac{1}{4}} \left(1 - \frac{9}{8} \frac{1}{\sqrt{(a_1 + a_2)} E} \right) \text{MeV}$$
 (22)

where E is the total internal excitation energy of a system of two interacting fragments that are in thermal equilibrium and a_1 , a_2 are level density parameters; the dependence of level density ρ on excitation energy is determined by Fermi statistics for a degenerate nucleon gas, i.e. by an expression of the type $\rho \sim \exp(2aE)^{\frac{1}{2}}$.

Without performing detailed calculations, Fong arrived at a number of conclusions about Δ . It was noted that Δ is determined largely by the charges of the fission fragments, Z_1 and Z_2 , and consequently also by the charge of the fissioning nucleus $Z = Z_1 + Z_2$. It was also pointed out that formula (22) leads to a very weak dependence of Δ on the fragment mass ratio; this dependence is determined by the factor E^4 , which weakly reflects the mass distribution, and the factor $(Z_1 \cdot Z_2)^{\frac{1}{2}}$, which makes Δ decrease somewhat with increasing fission asymmetry. It also follows from expression (22) that Δ increases for a given nucleus with increasing excitation energy. As we go from one nucleus to another Z, A and excitation energy E change, which leads to corresponding changes in Δ . For example, E and E are greater for E change, which leads to correspondingly, formula (22) leads to a higher kinetic energy distribution width E for the former. At the same time, the values of E for E

Quantitative results were presented in Ref. [85] only for 235 U fission by thermal neutrons and neutrons with energies of 2.5 and 14 MeV. For the most probable ratio of the fragment masses in the thermal neutron fission of 235 U a value of $\Delta=5.8$ MeV was obtained; taking into account the fact that Δ is the 1/e-half-width of the distribution, we find the corresponding value $\sigma_{\rm E_k}^2(A_1,A_2)\approx 47$ MeV², which is 1.5 times lower than the experimental value of this quantity [66] (see Fig. 8). The predicted rate of increase in Δ with increasing energy of the neutrons causing fission according to equation (22) agrees with the experimental data [86, 87] only moderately well at most.

It should be noted that in the formula (22) the dependence of Δ on the fissioning nucleus Z is expressed in explicit form only by the factor $(\mathbf{Z}_1\cdot\mathbf{Z}_2)^{\frac{1}{2}}$. If the dependence of Δ on Z is limited to this, quantitative agreement of the formula (22) with all the experimental data presented in sections 2–5 of this paper is hardly possible: the experimentally observed dependence of the variance of the fragment energy distributions on the number of protons in the fissioning nuclei is probably much stronger than \mathbf{Z}^2 . The dependence of the variance $\sigma^2_{\mathbf{E}_k}(\mathbf{A}_1, \mathbf{A}_2)$ on the fragment mass ratio is also fairly strong and of a complex nature, especially in the range of fragment mass

⁶ Here we retain the notation adopted in the original work [85]: Δ is the $\frac{1}{e}$ -half-width of the fragment kinetic energy distribution.

ratios from the symmetric to the most probable one [48, 56-73, 86, 87]; this dependence is not exhausted by a factor of $(\mathbf{Z}_1 \cdot \mathbf{Z}_2)$.

At the same time, use of the statistical theory for calculating such complex fission characteristics as fragment energy variations is very encouraging. The statistical theory enables one to take into account an enormous number of degrees of freedom of the fissioning system and, in principle, provides detailed predictions for virtually all fission fragment properties. It can naturally include shell effects, which play a decisive part in many aspects of low-energy fission of heavy nuclei. It is possible that the statistical theory in the form in which it appears in Ref. [83] will not lead to a successful quantitative description of the variations of fragment characteristics and that it will therefore be necessary to take into account the dynamic aspects of the fission process; this is decisive, for example, for the fission of heavy nuclei at higher excitation energies or for the fusion of heavy nuclei [84]. Nevertheless, we think it is well worth performing detailed calculations of fragment energy variations on the basis of the statistical theory over the full range of fissioning nuclei investigated experimentally using the latest nuclear data and methods of calculating potential energy, the level density of strongly deformed nuclei, etc. With such calculations it may be possible to gain a clearer picture of the limits of applicability of the statistical theory of fission.

Together with the dynamic liquid-drop theory and the statistical theory of fission, a number of models has also been used for calculating fission fragment energy distributions. A common feature of them is the introduction of additional assumptions and various parameters which should be obtained from experimental data.

For example, applying the fundamental propositions of the general theory of nuclear reactions to fission [88], Ericson performed an extensive theoretical analysis of the statistical model of fission [89]. With this approach, the relative probability P_{12} of the fission of a nucleus into fragments having masses A_1 , A_2 and charges Z_1 , Z_2 with a given total kinetic energy of the fragment pair ϵ and excitation energies U_1 and U_2 is given by the expression

$$P_{12}(\epsilon, U_1, U_2) \approx T(\epsilon) \rho_1(U_1) \rho_2(U_2)$$
(23)

where $T(\epsilon)$ is the penetration factor of the barrier between two completely separated fragments, and ρ_1 and ρ_2 are the densities of states of the excited fragments. Thus, the statistical equilibrium assumption is applied here not to the scission point but to a system of two fragments separated by an infinite distance. The function $T(\epsilon)$ is calculated in the WKB approximation for the penetration of a parabolic barrier whose parameters are determined by the condition of best agreement with experiment or by an analysis of experimental data on reactions with heavy ions. The distribution of the total fragment kinetic energy $P_{12}(\epsilon)$ is in this case obtained by integrating expression (23) over the excitation energy.

Using Ericson's model, Erba et al. [90] calculated fragment mass and kinetic energy distributions for several cases of fission, including thermal neutron fission of 235 U and spontaneous fission of 252 Cf. For 235 U(n, f) they obtained $\sigma_{E_k}^2(A_1, A_2) = 52$ MeV² for $A_1/A_2 = 101/135$, whereas the experimental value of this quantity [66] is 102 MeV². Similarly, for 252 Cf the calculated value of $\langle \sigma_{E_k}^2(A_1, A_2) \rangle$ is 72 MeV², whereas the experimental value (see Fig. 8) is about 100 MeV². Erba et al. did not calculate the kinetic energy distributions for other cases of low-energy fission of nuclei with $Z \ge 90$.

Subsequently, the difficulties of the above-mentioned approach to the calculation of fission fragment energy distributions [88–90] were noted in Refs [91–93], the authors of which, taking the comments of Swiatecki and Bjørnholm [94] into account, pointed out that the processes of nuclear fission and fusion cannot be considered at all mutually reversible. They also emphasized that detailed calculations of the potential energy surface did not lead to the kind of sharp, high barrier between fragments encompassed in the model discussed in Refs [89, 90]. The authors of Refs [91–93] proposed a new variant of the statistical model for calculating the energy distributions of fragments formed by the fission of actinides. This model is based on two assumptions: (a) the

internal degrees of freedom corresponding to the structure and internal states of the fragments are in statistical equilibrium; (b) the collective degrees of freedom associated with fragment motion do not share statistical equilibrium with the internal degrees of freedom. With these assumptions and using the usual methods of statistical mechanics, they obtained the following expression for the variance of the total excitation energy $U_1 + U_2$ or for the variance of the total kinetic energy for a specific pair of fragments:

$$\sigma_{(\mathbf{U}_1 + \mathbf{U}_2)}^2 = \sigma_{\mathbf{E}_k}^2 = \sigma_{\mathbf{U}_1}^2 + \sigma_{\mathbf{U}_2}^2 = 2(\overline{\mathbf{U}}_1' + \overline{\mathbf{U}}_2') t_0$$
 (24)

where $\overline{U}'=\overline{U}-\Delta$, Δ being the pairing energy, and t_0 is the mean initial temperature of the fission fragments. Again calculations were performed for only two cases of low-energy fission: 235 U(n, f) and 252 Cf(SF). In the first case, the calculated value of $\sigma_{E_k}^2(A_1,A_2)$ for $A_1/A_2=97/139$ was approximately 50 MeV², which is 1.6 times less than the experimental value [63, 66]. A similar difference between calculated and experimental values of $\sigma_{E_k}^2(A_1,A_2)$ is found for the most probable mode of 252 Cf fission; the difference is significantly greater for symmetric and strongly asymmetric fission.

It should be noted that, in the calculations performed in Refs [91–93], the values of the excitation energy and the statistical temperature entering into the right-hand side of expression (24) were taken directly from experiments; nevertheless, the agreement between the calculated and experimental values of $\sigma_{E_k}^2(A_1, A_2)$ is quantitatively unsatisfactory. Moreover, the results of calculation based on formula (24) can hardly be reconciled with the unusually strong Z dependence of the variance of the number of neutrons and the variance of the fragment kinetic energy for a wide range of fissioning nucleus presented in Figs 3 and 5–7.

Finally, Schmitt [95] used a simple static two-spheroid model in which the most probable kinetic energy $E_k(A_1,A_2)$ as a function of the fragment mass ratio was calculated by minimizing the total potential energy of the system, and the variance of the kinetic energy distributions $\sigma_{E_k}^2(A_1,A_2)$ was obtained by considering the quantum-mechanical properties of the system in the harmonic-potential approximation. For thermal neutron fission of uranium and plutonium isotopes and spontaneous fission of 252 Cf, satisfactory agreement between the calculations presented in Ref. [95] and experiment was observed. However, the effective values of the parameters of fragment stiffness necessary for the calculation of $\sigma_{E_k}^2(A_1,A_2)$ and $E_k(A_1,A_2)$ were taken from experiments [56, 66] involving measurements of fragment kinetic energies in thermal neutron fission of 235 U, 239 Pu and 241 Pu.

Thus, comparison of experimental data on the variance of the energy distributions of fragments formed by fission of heavy nuclei with theoretical predictions and calculations of a phenomenological or semi-empirical character shows that the question concerning the nature of the energy variations of fission fragments, their magnitude and the reasons for the changes of this quantity with respect to Z, A and excitation energy of the fissionable nucleus is still open.

8. CONCLUSIONS

Analysis of experimental data on the variance of the mean total kinetic or excitation energy distribution of fragments formed in low-energy fission of heavy nuclei as a function of the initial parameters of the fissioning nucleus shows that these quantities can change over a wide range. The scale of change in the fragment energy variations significantly exceeds the scale of change in the mean values of quantities related to them, that is, the mean total excitation energy and total kinetic energy of the fragments.

The dependence on the number of protons in the fissioning nucleus is most pronounced. The variance of the energy distributions increases sharply - by a factor of about 5- with

increasing Z as one goes from Z = 90 to Z = 102. It is possible that this dependence is on the whole exponential in character.

The 93 < Z < 97 region is an exception: at its boundaries there is a kink in curve of the Z dependence of the variance, and within this region the variance does not change very much. For a detailed study of the dependence in the 93 < Z < 97 region more reliable measurements are necessary.

A correlation is observed between the variance of the fragment energy distributions and the characteristics of the fragment mass distribution. The sharpest increase in the variance of the total number of neutrons σ_{ν}^2 and the variance of the total fission fragment kinetic energy $\sigma_{E_k}^2$ occur where the fragment mass distribution changes abruptly, the highest values of σ_{ν}^2 and $\sigma_{E_k}^2$ corresponding to the cases of the most symmetric fragment mass distribution. It is probably such a correlation that leads to a weakening of the Z dependence of the variance of the total energy distributions in the 93 < Z < 97 region. The low-energy fission of nuclei in this region is characterized by the strongest and most pronounced fragment mass asymmetry; in addition, the mass distributions do not experience significant changes as one goes from one nucleus to another.

The nature of the changes in the variance of the total number of neutrons σ_{ν}^2 and the variance of the total fragment kinetic energy $\sigma_{E_k}^2$ is virtually the same for spontaneous fission and thermal neutron fission. Quantitative comparisons of $\sigma_{E_k}^2$ with $(\sigma_{\nu}^2-1/12)E_0^2$, where E_0 is the average energy expended for the emission of one neutron, show that the ratio of these quantities remains fairly constant over a wide range of fission nucleus Z and A values, and the difference between them is on the average about 70 MeV². The balance of the variations of the fragment energy in the fission reaction is self-consistent if – taking into account the variance of the total reaction energy release σ_{Q}^2 – one takes the variance of the total gamma-ray energy distribution in fission $\sigma_{E\gamma}^2$ to be of the order of 30 MeV². For a more careful balancing of the fragment energy variations it is desirable to make a direct experimental determination of $\sigma_{E\gamma}^2$.

The dependence of the variance of the energy distributions on fissility parameter x is very

The dependence of the variance of the energy distributions on fissility parameter x is very similar in nature to the Z dependence for both spontaneous fission and neutron-induced fission. As the excitation energy of the fissioning nucleus changes from zero to $\cong 6.5$ MeV, on the whole the variance increases substantially. The difference is about 40 MeV² for uranium and increases appreciably in the Cf-Fm region, reaching 50-80 MeV². It appears to be at a minimum (5-10 MeV²) for plutonium isotopes.

The theoretical calculations performed so far do not give a satisfactory account of the pattern of experimental results described in this paper. Comparison of the experimental data with the predictions of the dynamic liquid-drop model and of the statistical theory of fission and with the results of calculations of a phenomenological or semi-empirical character shows that the question concerning the extent of fragment energy variations and the dependence of the variance on Z, A and excitation energy of the fissionable nucleus is still open.

Experimental data on the variance of the fragment energy distribution for the fission of nuclei with Z > 102 are completely lacking; similarly there is a lack of data on the fission of nuclei with Z > 94 at higher excitation energies. Besides spontaneous and neutron-induced fission, a promising source of such data may come from the fission of nuclei by heavy ions, especially in such combinations as 208 Pb + 48 Ca [96], which lead to compound nuclei with $E^* \cong 18$ MeV and a comparatively low angular momentum.

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