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# A statistical model for excitation energy distributions in actinide fission

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Abstract. The last stage of the fission process has been studied on the basis of a statistical model. Two fundamental hypotheses are made: (i) the internal degrees corresponding to the fragment nuclear structure are in conditions of statistical equilibrium and (ii) the freedom degrees related to the fragment motion at the scission point are not in statistical equilibrium with the internal degrees. The proposed statistical model reproduces the general properties of the excitation energies of the fission fragments in actinide fission.

# 1. Introduction

In recent years many aspects of the fission mechanism of heavy nuclei have begun to be understood; in particular, the properties of deformation energies, such as the basic discovery of the second minimum in the deformation energies, the properties of fission isomers, the asymmetric shapes of the nucleus at the second saddle and the description of fission widths and their behaviour (Brack *et al* 1972, Moeller and Nilsson 1970, Bolsterli *et al* 1972, Pauli *et al* 1971), have all been thoroughly investigated and clarified.

The last stage of the process, that is, the transition of the system from the saddle to the scission point, has not, however, been made clear up to now. The purpose of the present paper is to discuss this stage and in particular the energy properties of fission fragments.

In a recent paper Swiatecki and Bjørnholm (1972) present the stage currently reached by research on the nuclear system in the last fission path. Two extreme hypotheses have been considered in the meantime.

(i) A dynamical treatment of the scission process, when the nucleus is considered as a non-viscous irrotational fluid, has been studied by Nix (1968). His model gives correct results for fragment mass and energy spectra in the case of high-energy fission of light nuclei like bismuth but does not seem satisfactory for low-energy fission of actinides.

The most interesting result obtained in this analysis shows how the two fragments display a large amount of kinetic energy (20-40 MeV) at scission. This prediction agrees with the results obtained from the detailed analysis of ternary fission by various authors, such as Halpern (1963) and Feather (1969).

(ii) Starting from the opposite point of view, Fong (1953, 1956) assumed that the last stage of the deformation process occurs with strong viscosity so that the fragment kinetic energy at scission point is practically negligible. He then assumed that the probability of formation for a given pair of fragments is proportional to the density levels of the fragments at the scission point. With this model Fong reproduced the shape

of the mass distribution in  $^{235}$ U fission induced by thermal neutrons; this was done with the rather arbitrary use of numerous parameters<sup>†</sup>.

The analysis given below is based on a hypothesis that lies halfway between Fong's model of strong viscosity and Nix's approach, where viscosity is not taken into account.

We assume in fact that during the deformation motion from saddle to scission, a part of the available energy is converted into the internal excitation energy of the fragment and a part of it turns into the kinetic energy of the fragments.

The aim of this analysis is to show how a simple statistical description of the energy properties of the fission fragments can be given.

In § 2 the basic statistical hypotheses and formulae are discussed. In § 3 a comparison is made between the analysis predictions and the experimental results in  ${}^{252}$ Cf spontaneous fission (on which most experimental data are available), in  ${}^{235}$ U fission induced by thermal neutrons and in  ${}^{233}$ U fission at intermediate energies.

## 2. Basic hypotheses and formulae

#### 2.1. The energy at scission point

The energy balance of the nucleon system at scission point is achieved by considering the two nascent fragments  $A_1$ ,  $Z_1$  and  $A_2$ ,  $Z_2$  ( $A_1$ ,  $A_2$  being the mass numbers and  $Z_1$ ,  $Z_2$  the proton numbers of the two fragments) with a given distance  $r_s$  between their centres and with a total kinetic energy  $\mathscr{E}_c$ .

We have no direct information about the shape of the fragments at scission. We assume as a first approximation, that the fragments already have their final shape at scission. Therefore we do not consider the deformation energies of the fragments at the scission point as playing an important role. In § 4 we make a few remarks on this point.

The total initial energy of the fissile nucleus is

$$E_0 = m_0 c^2 + U_0 \tag{1}$$

where  $m_0$  is the rest mass of the nucleus in the ground state and  $U_0$  its initial excitation energy. The energy conservation at scission is written

$$E_0 = m_1 c^2 + m_2 c^2 + U_1 + U_2 + \mathscr{E}_c + V(r_s)$$
<sup>(2)</sup>

where  $m_1c^2$  and  $m_2c^2$  indicate the ground state energies of the two fragments  $(A_1, Z_1)$ ,  $(A_2, Z_2)$  respectively,  $U_1$  and  $U_2$  indicate the single fragment excitation energies at both the scission point as well as later on before neutron emission.  $V(r_s)$  is the coulombic repulsive potential at radius  $r_s$ .

As previously mentioned, the energy available at the scission point is divided between the internal excitation energy  $U = U_1 + U_2$  and the collective kinetic energy  $\mathscr{E}_c$  of the fragments.

The final total kinetic energy  $\mathcal{E}_{k}$  of the fragment is given by

$$\mathscr{E}_{\mathbf{k}} = V(r_{\mathbf{s}}) + \mathscr{E}_{\mathbf{c}},\tag{3}$$

† A different statistical model was developed by Newton (1956) and by Ericson (1960). Ericson's formulation takes into account the principle of detailed balance between fission and the inverse fusion process. Starting from this model Erba *et al* (1966) made an attempt to explain the properties of the fission fragments. It was shown recently by Swiatecki and Bjørnholm (1972) that fusion and scission cannot be considered the reverse of each other.

we then have the relation

$$\mathscr{E}_{\mathbf{k}} + U = Q_{\mathbf{t}} \tag{4}$$

where  $Q_1$  indicates the total fission energy.

The scission point is not sharply defined, being instead somewhat spread over the scission region. In this case the scission radius  $r_s$  must be considered an average value.

We can deduce an upper limit for  $V(r_s)$ . The maximum value of U is in fact limited by the energy  $Q_t - V(r_s)$  available at the scission point;  $V(r_s) \leq Q_t - U_{max}$ ; taking into account the values of  $Q_t$  and  $U_{max}$ , for instance in low energy fissions,  $V(r_s)$  is found to be smaller than 140 MeV and the corresponding value of  $r_s$  is higher than 18-20 fm.

#### 2.2. Statistical independence of $U_1$ and $U_2$

A fundamental property of excitation energies,  $U_1$  and  $U_2$ , of the fragments has been recently observed by Signarbieux *et al* (1972). Denoting  $v_1$  and  $v_2$  as the number of neutrons emitted by fragments  $A_1$ ,  $Z_1$  and  $A_2$ ,  $Z_2$ , respectively, these authors found experimentally that the variance of the sum  $v_1 + v_2$  is approximately equal to the sum of the separate experimental variances of  $v_1$  and  $v_2$ . Since the emitted neutron numbers are linearly dependent on the excitation energies, this means, by taking into account the well known properties of variances, that

$$\operatorname{cov}(U_1 U_2) = \int_0^\infty \int_0^\infty (U_1 - \overline{U}_1)(U_2 - \overline{U}_2)p(U_1 U_2) \, \mathrm{d}U_1 \, \mathrm{d}U_2 \simeq 0 \tag{5}$$

where  $\overline{U}_1$  and  $\overline{U}_2$  indicate the average values of fragment excitation energies and  $p(U_1U_2) dU_1 dU_2$  is the probability of finding fragment  $A_1, Z_1$  with excitation energy between  $U_1$  and  $U_1 + dU_1$  and fragment  $A_2, Z_2$  with excitation energy between  $U_2$  and  $U_2 + dU_2$ .

As a consequence of equation (5), the function  $p(U_1, U_2)$  turns out to be the product of two functions, one of  $U_1$  and one of  $U_2$ .

# 2.3. Scission states

Each particular quantum state of the nucleus at scission point is characterized first by the way the nucleons split into two fragments  $A_1$ ,  $Z_1$  and  $A_2$ ,  $Z_2$  and then by the particular configuration the nucleons assume in each fragment. We indicate the internal states of fragments  $A_1, Z_1$  and  $A_2, Z_2$ , respectively, by the symbols  $s_{1a}, s_{2b}$  where  $a = 1, 2, 3, \ldots; b = 1, 2, 3, \ldots$  The combined states of the system formed by fragments  $A_1, Z_1, A_2, Z_2$  in states  $s_{1a}$  and  $s_{2b}$  respectively, are indicated in the following simply by s(y). For our purpose it is important to point out that the system can assume an extremely large number of configurations; the number of fragment pairs is of the order of a few hundred and at excitation energies of 10–30 MeV the number of excited levels of fragments ranges more or less from  $e^{20}-e^{40}$ .

Each system state corresponds to a given value of the collective kinetic energy  $\mathscr{E}_{c}$ . The internal or structural energy E(y) of the scission system in s(y) state is given by

$$E(y) = m_1 c^2 + m_2 c^2 + V(r_s) + U_{1a} + U_{2b}$$
(6)

where  $U_{1a}$  and  $U_{2b}$  are the excitation energies of fragments  $A_1$ ,  $Z_1$  and  $A_2$ ,  $Z_2$  when they are excited in  $s_{1a}$  and  $s_{2b}$  states, respectively.

Our statistical description of the scission process is based on two fundamental hypotheses.

(i) The internal freedom degrees corresponding to the fragment nuclear structure and characterized by the quantities  $A_1, Z_1; A_2, Z_2; s_{1a}, s_{2b}$  are in a condition of statistical equilibrium.

(ii) The freedom degrees related to the fragment motion at scission point are not in statistical equilibrium with the internal degrees. This hypothesis is suggested by the fact that the fragment kinetic energy  $\mathscr{E}_{c}$  represents a collective motion of the nucleons which we consider not strongly coupled with the internal freedom degrees.

With a fixed  $r_s$  value, for each fragment pair we have

$$0 \leq \mathscr{E}_{c} \leq Q_{t} - V(r_{s}) \tag{7}$$

and the corresponding condition for E(y)

$$E_0 - (Q_t - V(r_s)) \leqslant E(y) \leqslant E_0.$$
(8)

We also have the conservation relations

$$A_1 + A_2 = A_0 (9)$$

$$Z_1 + Z_2 = Z_0 (10)$$

 $A_0, Z_0$  being the mass number and the proton number of the fissioning nucleus.

#### 2.4. Probability of formation of fragment states

We denote by p(y) the probability of finding the nucleon system at scission point in a given s(y) state. We assume that the p(y) probabilities are statistically distributed over all the available states of the fragment system.

In order to apply statistical considerations to a fissile nucleus  $A_0$ ,  $Z_0$  at scission, let us now consider an ensemble consisting of a very large number n of distinguishable systems, each one a replica of the system of interest.

As is usual in statistical mechanics (see for example Davidson 1962), we assume that the average properties of the fissile nucleus  $(A_0, Z_0)$  at the scission point can be obtained by ensemble averaging of the properties of the system.

We remark, incidentally, that this condition is equivalent to the hypothesis of statistical equilibrium among the scission fragment states<sup>†</sup>.

We can now consider the ensemble of n systems in a particular ensemble state, which can be described as follows:  $n_1$  systems of the ensemble are in state s(1),  $n_2$  in state s(2),  $n_3$  in state s(3) and so on. There are a number of individual distinguishable ensemble states, which correspond to the distribution  $D(n_1, n_2, n_3, ...)$ . The general hypothesis is made that any 'ensemble state' is *a priori* equiprobable.  $t_D$  is the symbol we use for this number of individual distinguishable ensemble states for this particular D distribution. We have

$$t_D = \frac{n!}{\prod_v n_v!}.\tag{11}$$

The possible distributions D are limited by the two conditions

$$\sum_{y} n_{y} = n \tag{12}$$

† This overcomes the difficulties presented by the fact that scission is a rapid irreversible process.

which corresponds to the conservation of the total number of ensemble systems and by

$$\sum_{y} n_{y} E(y) = \bar{E}n, \tag{13}$$

 $\overline{E}$  being the average internal energy per system. The average entropy of a nucleon system in the ensemble is defined by

$$\bar{S} = \ln t_{\rm tot}/n \tag{14}$$

 $t_{\rm tot}$  being the total number of states in which the ensemble occurs:

$$t_{\text{tot}} = \sum_{D} t_{D} \tag{15}$$

the sum being extended to all distributions which satisfy the relations (12) and (13).

Considering equations (11), (15) and their related conditions, we reach the conclusion that the statistical properties of the fragment system at scission point can be described by the well known canonical description of particle systems in thermal equilibrium (see, for instance, Jackson 1968 and Davidson 1962).

According to the usual procedure the properties of the system of interest are, to a high degree of approximation, the properties of the most probable distribution. Nearly all the ensemble states correspond in fact to the most probable distribution, which maximizes  $t_D$  subject to the conditions (12) and (13). Applying the standard procedure for optimizing  $t_D$  from equation (11) with conditions (12) and (13), one obtains the probability  $p(y) = n_y/n$  of finding the system of interest in the yth state

$$p(y) = \mathscr{Z}_{f}^{-1} e^{-\beta_{0} E(y)}$$
(16)

 $\mathscr{Z}_{f}$  is the canonical partition function of the system and is given by

$$\mathscr{Z}_{f} = \sum_{y} e^{-\beta_{0} E(y)}$$
(17)

where the sum includes all system states which satisfy the relations (8)–(10). The constant  $\beta_0$  value is usually represented as the inverse of the statistical  $t_0$  temperature of the system

$$\beta_0^{-1} = t_0. \tag{18}$$

The constant  $\beta_0$  value is related to the average value  $\overline{E}$  of the internal or structural energy of the system by the well known relation

$$\bar{E} = -\frac{\partial (\ln \mathscr{L}_{f})}{\partial \beta_{0}}.$$
(19)

It should be pointed out that the model does not predict the value of  $\overline{E}$  but gives the probability distribution law around the average value. We can finally give the expression for the entropy

$$\bar{S} = \beta_0 \bar{E} + \ln \mathscr{Z}_{\rm f} \tag{20}$$

which refers to the canonical systems.

We shall not attempt to prove the validity of the assumptions made, for instance by a general discussion of the viscous motion of the system; our purpose is to give a statistical interpretation of fragment properties.

In particular, in this paper we are interested in the energy properties of fragment pairs, so we apply equation (16) to examine the distributions of excitation energy for any given fragment pair. Then considering mass and potential energies constant, equation (16) for each given fragment pair becomes

$$p(s_{1a}, s_{2b}) = \mathscr{Z}_{12}^{-1} \exp(-\beta_0 U_{1a} - \beta_0 U_{2b})$$
(21)

where  $p(s_{1a}, s_{2b})$  indicates the probability of finding the  $(A_1, Z_1)$ ,  $(A_2, Z_2)$  system with a fragment  $(A_1, Z_1)$  in state  $s_{1a}$  and a fragment  $A_2, Z_2$  in state  $s_{2b}$ .

 $\mathscr{Z}_{12}$  is the sum

$$\mathscr{Z}_{12} = \sum_{ab} \exp(-\beta_0 U_{1a} - \beta_0 U_{2b}).$$
(22)

This sum includes all the available internal states of fragments  $A_1, Z_1$  and  $A_2, Z_2$ .

Due to the fact that states  $s_{1a}$  are statistically independent of states  $s_{2b}$ , it is possible to split up the sum, obtaining

$$\mathscr{Z}_{12} = \left(\sum_{a} \exp(-\beta_0 U_{1a})\right) \left(\sum_{b} \exp(-\beta_0 U_{2b})\right).$$
(23)

#### 2.5. Excitation energy distributions

Regrouping the internal states corresponding to given values of excitation energies  $U_1$  and  $U_2$  and considering the U's as continuum variables we have from equation (21)

$$p(U_1U_2) dU_1 dU_2 = \mathscr{Z}_{12}^{-1} e^{-\beta_0 U_1} \rho_1(U_1) e^{-\beta_0 U_2} \rho_2(U_2) dU_1 dU_2.$$
(24)

 $\rho_1(U_1)$  and  $\rho_2(U_2)$  indicate the nuclear level densities of  $A_1$ ,  $Z_1$  and  $A_2$ ,  $Z_2$ , respectively. Function  $\mathscr{Z}_{12}$  has to be expressed with appropriate integral forms.

It is interesting to note that equation (24) agrees with the independence rule (5) obtained from the experimental results.

The excitation energies  $U_1$  and  $U_2$  fluctuate around given average values.

From formula (24) we get the energy distributions of  $U_1$  and  $U_2$  separately

$$p(U_i) \,\mathrm{d}U_i \equiv \mathrm{e}^{-\beta_0 U_i} \rho(U_i) \,\mathrm{d}U_i \qquad (i = 1, 2). \tag{25}$$

For level density we introduce the expression given considering the nucleons in the nucleus as arranged in equi-spaced or nearly equi-spaced single-particle states (Ericson 1960). We have

$$\rho(U_i) = \frac{6^{1/4}}{12} g_i^{-1/4} \frac{\exp(2\sqrt{a_i U_i})}{U_i^{5/4}}$$
(26)

 $g_i = (6/\pi^2)a_i$ ,  $g_i$  being the average density of nucleons at the Fermi top, and  $U'_i = U_i - \Delta_i$ , where  $\Delta_i$  is the pairing energy. We take  $\Delta_i = 11.5/A_i^{1/2}$  MeV (Wing and Varley 1964).

The nuclear temperature is given by

$$T^{-1} = \frac{\mathrm{d}\ln\rho}{\mathrm{d}U}.\tag{27}$$

Where T/U is small, equation (26) leads to the following approximate relation between excitation energy and temperature:

$$U_i' \simeq a_i T_i^2 - 2.5 T_i. \tag{28}$$

Equation (26) has been compared with experimental data on level densities by numerous authors and a values are given for a large number of nuclei (Erba *et al* 1961, Facchini and Saetta-Menichella 1968). These values show a regular rise with A values, while deep minima are found for magic and double-magic nuclei. The *a* values of fission fragments for actinides were obtained by Lang (1964) and Bishop *et al* (1970) at different excitation energies through analysis of the cascade of neutrons evaporated from excited fragments.

It is interesting to note that the *a* values obtained by Lang (1964) at low excitation energies show a deep minimum in the region of the double-magic nucleus  $A_2 = 132$ ,  $(N_2 = 82, Z_2 = 50)$ . The minimum is less pronounced at higher excitation energies (Bishop *et al* 1970). This becomes clearer if it is recalled that many experimental results and theoretical analyses have shown the 'anomalous behaviour' of  $\rho(U)$  in the nuclei before and after the magic nuclei, for instance in the case of lead (Z = 82, N = 126) and of tin (Z = 50). In the region of magic nuclei the level density expression is not given by equation (26) but  $\ln \rho(U)$  shows approximately a linear rise against U for U values which are not high (Marujama 1969, Ignatiuk *et al* 1970, Williams *et al* 1972).

It is, therefore, also possible to assume similar behaviour for fission fragments A = 132 with Z close to 50 and with N around 82.

We have the peak values  $U^*$  and  $T^*$  at the peak of energy distribution (25) when

$$\left(\frac{\mathrm{d}\ln\rho_i}{\mathrm{d}\,U_i'}\right)_{\mathrm{at\,max}} = \beta_0. \tag{29}$$

Equations (29) and (27) give equal nuclear temperatures for the two complementary fragments as the most probable:

$$T_1^* = T_2^*. (30)$$

These are equal to each other as well as to the statistical temperature  $t_0: T_1^* = T_2^* = t_0$ .

If distribution (25) is taken into account, it can be easily shown that the average values  $\overline{U}_1$ ,  $\overline{U}_2$  and  $\overline{T}_1$ ,  $\overline{T}_2$  are approximately equal to the most probable one, so that we have

$$\overline{T}_1 \simeq \overline{T}_2 \simeq t_0. \tag{31}$$

The canonical ensemble formalism (Jackson 1968, Davidson 1962) gives the following simple expression of variance

$$\sigma_{U_i}^2 = t_0^2 \frac{\mathrm{d}\,\overline{U}_i}{\mathrm{d}\,t_0} \qquad (i = 1, 2). \tag{32}$$

When considering the equi-spaced single-particle model (Lang 1954, Ericson 1960) we have:

$$\overline{U}_i' = a_i t_0^2. \tag{33}$$

From relation (32) we have then:

$$\sigma_{U_i}^2 = 2a_i t_0^3 \qquad (i = 1, 2)$$

and then

$$\sigma_{U_i}^2 = 2\overline{U}_i t_0 \qquad (i = 1, 2). \tag{34}$$

If the nuclear level density logarithm follows an approximate linear law, as in the magic fragment region, we should expect a larger value of  $d\overline{U}/dt_0$  and a corresponding increase of  $\sigma^2$ .

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It is interesting to consider the fractional fluctuation in energy (ie the  $\sigma_U/\overline{U}$  ratio). To this aim we introduce  $\bar{n}$ , which represents the average number of excited degrees of freedom, that is, excited nucleons and their corresponding holes involved in the building up of the excitation average energy  $\overline{U}$ . The  $\bar{n}$  expression is given by Ericson (1960) as follows

$$\bar{n} = 2gt_0 \ln 2. \tag{35}$$

Using the expressions  $\overline{U}' = at_0^2$  and  $\sigma_U^2 = 2\overline{U}'t_0$  leads to

$$\left(\frac{\sigma_U}{\overline{U}'}\right)^2 = \frac{2}{at_0} = \frac{12}{\pi^2 gt_0}$$

and hence from (35)

$$\frac{\sigma_U}{U'} \simeq \frac{1\cdot 3}{\sqrt{\bar{n}}}.$$
(36)

Taking into account the values of g and the experimental values of  $t_0$  (see §3), the values of  $\bar{n}$  turn out to be of the order of 10–20 and the ratio  $\sigma_U/\overline{U'}$  is of the order of 0.3. The excitation energies will show a marked fluctuation due to the low values of  $\bar{n}$ .

#### 2.6. Total excitation and kinetic energy distribution

It is interesting to analyse the distribution of the total excitation energies  $U = U_1 + U_2$  for the various fragment pairs.

The value of U is directly related to total kinetic energy  $\mathscr{E}_k$  as shown in equation (4), so that the distribution of U corresponds to the distribution of  $\mathscr{E}_k$ , which is known from experiments. The distribution of U is obtained from equation (24) by substituting for the variable  $U_2$  and integrating over  $U_1$ :

$$p(U) dU = \int_0^U p(U - U_1, U_1) dU_1 dU$$
(37)

and successively from equation (24)

$$p(U) dU = \mathscr{Z}_{12}^{-1} e^{-\beta_0 U} \int_0^U \rho_1(U_1) \rho_2(U - U_1) dU_1 dU.$$
(38)

The indicated integral  $\int_0^U \rho_1 \rho_2 dU_1$  was studied by Fong (1956) and is approximately given by

$$C' \exp[2\sqrt{(a_1 + a_2)U'}]$$
 (39)

where C' is nearly constant, and  $U' = U - (\Delta_1 + \Delta_2)$ . Finally we obtain

$$p(U) dU \equiv e^{-\beta_0 U} \exp[2\sqrt{(a_1 + a_2)U'}].$$
(40)

The distribution turns out to have average values of U and U' given by

$$\overline{U} \simeq \overline{U}_1 + \overline{U}_2; \qquad \overline{U}' = \overline{U} - (\Delta_1 + \Delta_2) \tag{41}$$

while the variance is given by

$$\sigma_U^2 = \sigma_{\mathscr{E}_k}^2 = \sigma_{U_1}^2 + \sigma_{U_2}^2 = 2\overline{U}' t_0.$$
(42)

If the system of the two fragments is considered at a given value of total kinetic energy  $\mathscr{E}_k$ , that is, at a fixed value of total excitation energy U, the division of excitation energy between the two fragments turns out to be determined by the product  $\rho_1 \rho_2$ :

$$p(U_1, U_2)_{(U_1 + U_2 = U)} dU_1 dU_2 \equiv e^{-\beta_0 U} \rho_1(U_1) \rho_2(U_2) dU_1 dU_2 \delta(U_1 + U_2 - U).$$
(43)

This distribution has a maximum value for

$$\left(\frac{\mathrm{d}\ln\rho_1(U_1)}{\mathrm{d}U_1}\right)_{\mathrm{at\,max}} = \left(\frac{\mathrm{d}\ln\rho_2(U_2)}{\mathrm{d}U_2}\right)_{\mathrm{at\,max}}$$

ie,

$$T_1^*(\mathscr{E}_k) = T_2^*(\mathscr{E}_k) \tag{44}$$

as  $T_1^*(\mathscr{E}_k)$  and  $T_2^*(\mathscr{E}_k)$  are the most probable nuclear temperatures of each fragment  $A_1, Z_1$  and  $A_2, Z_2$  for any given value of total kinetic energy  $\mathscr{E}_k$ . From the equality of the most probable temperatures, we also have approximate equality of their average values.

#### 3. Comparison with experimental results

#### 3.1. Temperatures and excitation energies

In 1964 Lang developed a statistical analysis of the properties of the cascade of neutrons emitted by fission fragments. We are interested in the relation between the average value of the kinetic energy of these neutrons  $\bar{\mathscr{E}}_n$  and the fragment temperatures. Denoting by  $\tau_i$  the nuclear temperature at excitation  $U_i - \frac{1}{2}B_{ni}$ , when  $U_i$  is the initial nuclear excitation energy and  $B_{ni}$  is the binding energy of the first emitted neutron, we have

$$\bar{\mathscr{E}}_{ni} = \frac{3}{4}\tau_i. \tag{45}$$

Taking into account equation (28), the average initial temperature  $\overline{T}_i$  of the fragment is approximately given by

$$\overline{T}_i \simeq \frac{3}{4} \bar{\mathscr{E}}_{ni} \left( \frac{\overline{U}'_i}{\overline{U}'_i - \frac{1}{2} B_{ni}} \right)^{1/2},\tag{46}$$

 $\overline{T_i}$  indicates the temperature corresponding to average  $\overline{U_i}$ . This formula can be applied when the average number of neutrons emitted by the considered fragment is more than one. In those cases where level density behaviour is not represented by equation (26), but approaches the constant temperature formula, the average kinetic energy of the neutrons has a value very close to  $2T_c$ ,  $T_c$  being the constant or approximately constant temperature of the nuclear cascade in a given energy region.

The values of  $U_i$  are usually obtained from the values of  $\bar{v}_i$  and of energy  $\bar{E}_{\gamma i}$  emitted as  $\gamma$  rays

$$\overline{U}_i = \overline{v}_i (\overline{B}_{ni} + \overline{\mathscr{E}}_{ni}) + \overline{E}_{\gamma i}. \tag{47}$$

In analysis of fission processes for which the energy  $\overline{E}_{yi}$  is not given as the result of experiments, one can approximately assume that

$$\bar{E}_{yi} = 0.75\bar{v} + 2. \tag{48}$$

For the purpose of the present analysis, equations (47) and (48) can be considered satisfactory; in fact, the values of  $\overline{E}_{\gamma i}$  prove to be slightly higher than the values  $\frac{1}{2}B_{ni}$  because of competition between  $\gamma$  rays and neutrons at the end of the neutron cascade due to the high value of the fragment spin (Nifenecker *et al* 1972).

The  $B_{ni}$  values were taken from the tables of Wing and Varley (1964) averaging for each fragment pair  $A_1, A_2$  out of different  $Z_1, Z_2$  proton numbers.  $\overline{B}_{ni}$  indicates the weighted average value of  $B_{ni}$  over the cascade neutrons.

# 3.2. <sup>252</sup>Cf spontaneous fission

The values of  $\overline{U}_i$  were calculated from equation (47) taking the  $\overline{\mathcal{E}}_n$  values from Bowman *et al* (1963), the  $\overline{v}_i$  values given recently by Signarbieux *et al* (1972) and the  $\overline{E}_{\gamma i}$  values as given by Nifenecker *et al* (1972). The values of  $\overline{T}_i$  obtained from equation (46) are given in figure 1. Two interesting points can be brought out; the initial temperatures  $\overline{T}_i$  of the fragments are almost equal for the two complementary fragments of any given pair and they are also almost constant throughout the mass spectrum, except in the symmetric mass region, where they show an increase. It must be pointed out, however, that the  $\overline{T}_i$  values in this region, as given by the equation (46), will turn out to be overestimated.



Figure 1. Average initial temperatures of the fragments in  ${}^{252}$ Cf spontaneous fission. The temperatures refer to the beginning of the neutron cascade and correspond to the temperatures of the nascent fragments at scission point. Errors in temperatures are not given: they are of the order of  $\pm 5\%$  and due to errors of  $\overline{\mathscr{S}}_n$  and to some uncertainty in application of equation (26) for the level density; in the magic nuclei region around  $Z \simeq 50$ ,  $A \simeq 132$ , the correction will reduce the given  $\overline{T}_i$  values to a factor ranging from 1 to 0.6.

When the uncertain values in the symmetric mass region are not considered, we obtain the value  $\langle \overline{T}_i \rangle = 1.12$  MeV, as the average value for all fragment pairs. This value approximately corresponds to the equilibrium temperature  $t_0$ .

In the case of <sup>252</sup>Cf spontaneous fission it is possible to make direct verification of equation (44) which predicts equality of the average initial temperature of fragments for fixed values of the total kinetic energy  $\mathscr{E}_k$ . In fact, figure 15 of Bowman *et al* (1963) shows the approximate equality of the average kinetic energies  $\overline{\mathscr{E}}_n$  of the cascade neutrons

at given values of total kinetic energy for most fragment pairs : it should be recalled that the temperature values are nearly proportional to  $\mathcal{E}_n$  (see relation (46)).

We now calculate the values of  $\sigma_{\mathscr{E}_k}$  as given by equation (42). The U' values can be calculated with equations (41) and (47) or, otherwise, by considering the energy balance equation

$$\overline{U}' = Q_{t} - \overline{\mathscr{E}}_{k} - (\Delta_{1} + \Delta_{2}) \tag{49}$$

where  $\mathcal{E}_{\mathbf{k}}$  indicates the average value of  $\mathcal{E}_{\mathbf{k}}$ .

The values of  $Q_t$  have been taken from the tables of Wing and Varley (1964), averaging for each fragment pair  $A_1$ ,  $A_2$  out of different proton numbers  $Z_1$ ,  $Z_2$ . In the case of <sup>252</sup>Cf spontaneous fission very accurate values of  $\overline{\mathscr{E}}_k$  (Signarbieux *et al* 1973, private communication) are available, so that we prefer to use equation (49). The value  $t_0$ has been assumed as  $t_0 = 1.12$  MeV.

The calculated values of  $\sigma_{\mathscr{E}_{k}}$  are given in figure 2 together with the experimental ones. The experimental values come from recent results of the Saclay group (Signarbieux *et al* 1973, private communication).



**Figure 2.** Calculated ( $\bigcirc$ ) and experimental ( $\bigcirc$ ) values of  $\sigma_{\delta_k}$ . Errors in calculated values are estimated of the order of a few per cent. Errors in experimental values are of the order of 0.05–0.2 MeV (Signarbieux *et al* 1973). In the symmetric mass region the calculated values  $\sigma_{\delta_k}$  are underestimated because of shell effects.

The agreement between the two sets of values is quite good, except in the symmetric fission region. The discrepancy in the symmetric mass region can be explained by taking into account that  $d\overline{U}/dT$  is larger than the value given by (28) for nuclei around the magic region.

In the framework of this simple approach to the problem, experiments and model predictions agree fairly well.

# 3.3. Thermal neutron induced fission of $^{235}U$

The  $\vec{e_n}$  values were taken from the measurements of Milton and Fraser (1965); it should be recalled that there is considerable uncertainty in these values and that for many fragments the  $\bar{v}$  values are smaller than one, thus preventing application of equation (45). Many experiments have given  $\bar{v}$  values; we have averaged out the values obtained by many researchers (Milton and Fraser 1965, Apalin *et al* 1964, 1965, Maslin *et al* 1967).

By using equations (46) and (47) we obtain the  $\overline{T}_i$  values given in figure 2. The  $\overline{T}_i$  values are not considered for fragments where the  $\overline{\nu}$  values are too low.



**Figure 3.** Average initial temperatures of the fragments in <sup>235</sup>U fission induced by thermal neutrons. The temperatures refer to the beginning of the neutron cascade and correspond to the temperatures of the nascent fragments at the scission point. Errors in temperature are not given: they are of the order of  $\pm 5\%$  and due to errors of  $\mathcal{E}_n$ —in the symmetric mass spectra errors are bigger, up to 20–30%—and to some uncertainty in application of equation (26) for the level density; in the magic nuclei region around  $Z \simeq 50$ ,  $A \simeq 132$ , the correction will reduce the given  $T_i$  values to a factor ranging from 1 to 0.6.

The  $\overline{T}_i$  values are approximately constant for all fragments considered. It is interesting to note that the average  $\langle \overline{T}_i \rangle$  value for the given fragments turns out to be 1.095 MeV which is approximately the same value as that obtained for <sup>252</sup>Cf spontaneous fission.

This is a very interesting point : the viscous motion produces nearly the same temperatures in the two nuclear processes under consideration. A consequence of temperature equality and constancy is the fact that the excitation energy and consequently the values of emitted neutron number of a given fragment remain almost the same even when different parent nuclei are considered. This rule (ie, the approximate equality of  $\bar{\nu}$ values for <sup>252</sup>Cf spontaneous fission and for <sup>235</sup>U fission induced by thermal neutrons) was first established by Terrell (1962).

For the calculation of the values of  $\sigma_{\mathscr{E}_k}$  we have used equations (42) and (49). The values of  $\mathscr{E}_k$  have been taken from Ribrag (1967) and the values of  $Q_1$ , as in the case of <sup>252</sup>Cf fission, from the tables of Wing and Varley (1964).

The calculated values  $\sigma_{e_k}$  are shown in table 1 together with the experimental results taken from the paper by Schmitt *et al* (1966). Agreement between the two sets of values is generally satisfactory.

Since in the region where  $Z \simeq 50$ , we are facing the same problem as in <sup>252</sup>Cf fission, it is reasonable to assume that the discrepancies are due to the same effects.

$\sigma_{\boldsymbol{\ell}_{\mathbf{k}}(\mathbf{exp})}$ (MeV)	
6.9	
7.0	
7.2	
7.6	
8.7	
10.1	
10.6	

**Table 1.** Calculated and experimental values of  $\sigma_{\theta_k}$  for <sup>235</sup>U + thermal neutron fission.

Errors in calculated  $\sigma_{e_k}$  values are estimated of the order of a few per cent. The errors in experimental  $\sigma_{e_k}$  values are discussed in the original paper by Schmitt *et al* (1966). The values in the symmetric mass region have been omitted either because of the large errors in the experimental data and because of the uncertainty of application of equation (42).

# 3.4. $^{233}U + 13 - 14 MeV$ protons

We have examined the experimental results obtained by Bishop *et al* (1970) and by Burnett *et al* (1971).

The values of  $\bar{v}$  were given by both groups of researchers; but the values of Bishop *et al* fit the energy balance better. The values of  $\mathcal{E}_n$ , the average kinetic energy of the cascade neutrons, were measured by Bishop *et al* (1970); these values correspond to the whole distribution of total kinetic energy. The  $\overline{T}_i$  values obtained are given in table 2. The given  $\overline{T}_i$  values are almost constant for all the mass spectrum; in particular they are equal for the two fragments of a pair, as obtained in the statistical analysis. The average  $\langle \overline{T}_i \rangle$  value turns out to be about 1.2 MeV, which approximately represents the equilibrium temperature  $t_0$ .

<i>A</i> <sub>1</sub>	A 2	$\overline{T}_1$ (MeV)	T <sub>2</sub> (MeV)	$\sigma_{{\cal E}_{f k}({ m calc})} \ ({ m MeV})$	$\sigma_{\mathcal{S}_{k}(exp)}^{\dagger}$ (MeV)
90	144	1.18	1.23	9.8	9.0
95	139	1.22	1.26	9.7	10.0
100	134	1.15	1.30	10.1	11.5
105	129	1.15	1.26	10.0	12.3
110	124	1.22	1.24	10.7	11.0
116	118	1.21	1.20	10.9	10.5

Table 2. Experimental and calculated results for  $^{233}U + 13-14$  MeV proton induced fission.

Errors in  $\overline{T}_i$  and in  $\sigma_{\sigma_k}$  are not given; they can be estimated to be of the order of a few per cent due to errors in the experimental data.

† Burnett et al (1971).

The  $\sigma_{\mathscr{E}_{k}}$  values we obtained from equation (42) are reported in table 2 where the experimental values (Burnett *et al* 1971) are also shown.

The values  $\overline{U}'$  have been obtained by adding the values  $\overline{U}'_1$  and  $\overline{U}'_2$  obtained from the cascade properties.

There is reasonable agreement between the two sets of values (see equation (47)).

Similar results can be obtained by considering the data on  $^{238}U + 11.5$  MeV protons given by Bishop *et al* (1970) and the analogous fission reactions studied by Cheifetz and Fraenkel (1968).

# 4. Conclusions

The main conclusion obtained from this analysis is that the general properties of excitation energies of fission fragments are explained within the framework of a statistical model. The model does not predict the value of the statistical temperature, but this value can be obtained directly from experimental data.

The widths of the spectra are well reproduced and correspond to the peculiar fluctuations of nuclear energies due to the small number of nucleons which play a role in the building up of the internal excitation of the fragments.

The values of the fragment energy distribution variances are approximately reproduced in all the fission processes that are considered.

Two remarks should be made: firstly, the results were obtained without introducing a particular deformation of the fragments at scission; secondly, we considered the different fragment pairs separately, so that the mass spectrum was not discussed. This point, however, will be clarified in a further analysis.

The first point can be briefly discussed from the consideration that the deformation energy at the scission point is unimportant, it follows that the equilibrium temperatures have been evaluated by taking into account the whole excitation energies of the fragments.

Under this hypothesis a nearly constant temperature has been obtained and the variances of excitation energy spectra are in agreement with experimental data. The peculiar behaviour of average excitation energies  $\overline{U}_i$  plotted against A (the saw-tooth shape) are a consequence of the critical dependence of the level density parameters, the g for instance, on different nuclei and in particular on closed shell nuclei and their neighbours.

It is known that a different viewpoint was expressed in the past (Vandenbosch 1962): the assumption was made that a good deal of the final fragment excitation comes out of the deformation energy at scission, weakly coupled with the internal excitation.

We should emphasize that the fragments at the scission point cannot be considered as exactly isolated nuclei with their given level densities and ground state energies.

Interaction between the fragments which is active just until scission and the equilibrium condition between the various fragment pairs will cause a perturbation of the level densities, at least at the Fermi top.

We must say, however, that, when a large deformation energy is introduced at scission, the results presented in this paper will be deeply modified and may lose their significance.

The agreement between experimental results and statistical analysis indicates, in principle, that deformation is not very important, but in the symmetric mass region the results are confused and different possibilities are open.

One should note, finally, that the deformation energy exceeding the ground state energy, when stored in the nuclei, can be statistically unfavoured in spite of the excitation energy, which corresponds to many more degrees of freedom of the nuclear system.

The predictions given by the deformation model are not definitely proved. The saw-tooth behaviour of  $\overline{U}_i$  is reproduced by assuming a critical deformability of the

nuclei which is somehow related to the level density properties; the model does not give a precise interpretation of variance  $\sigma_U^2$ .

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