Nuclear level density and the mass distribution of fission fragments

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Using the single particle energy spectrum and Bethe's formula for nuclear level density, the mass distribution of fission fragments is calculated for nuclei ^{236}U, ^{258}Fm, and ^{240}Pu, and also for the compound systems ^{84}Kr + ^{238}U and ^{129}Xe + ^{197}Au at different excitation energies. The results are in reasonable agreement with experimental yield curves.

[RADIOACTIVITY, FISSION Nuclear level density, mass and charge distribution of fission fragments.]

I. INTRODUCTION

The mass distribution of fission fragments has been a subject of great interest in the recent past. There have been two major approaches to the problem of nuclear fission. One is the statistical method due to Fong[1,2] and the other is the microscopic method developed by Greiner and his co-workers[3,4] using the two center shell model and Strutinsky's prescription for shell correction. It is shown in Ref. 5 that the shell corrections in the separated fragments dominate and play a decisive role in the fissioning mode. This had been realized earlier by Wilkins *et al.*,[6] who made an elaborate study of the nuclear fission process by calculating the total energy of the fissioning system at the scission point. According to them, the probability of finding a fragmentation with mass numbers A_1 and A_2 is given by

$$P(A_1,A_2)=\int\int \exp[-V(A_1,A_2,\beta_1,\beta_2,\tau)/T_{\rm col}]d\beta_1 d\beta_2 \quad . \tag{1}$$

In the above expression the total energy $V(A_1,A_2,\beta_1,\beta_2,\tau)$ includes the temperature (τ), dependent shell corrections,[7] and the deformation degrees of freedom (β_1,β_2) of the individual fragments, and $T_{\rm col}$ is taken as a constant equal to 1 MeV. However, it may be remarked that the thermodynamical probability for an ensemble is given by exp(S), where S is the entropy, and hence, it is proportional to the level density. This forms the basis for Fong's method.

According to the statistical theory of fission proposed by Fong, the probability of a fission mode depends upon the density of quantum states available for the fissioning nucleus at the point of scission. This density can be taken to be equal to the product of nuclear level densities of the two fragments (A_1,Z_1) and (A_2,Z_2) with excitation energies E_1 and E_2, respectively. To calculate the individual nuclear level densities, Fong has used an approximate expression which takes into account only the exponential dependence of the excitation energy. The total excitation nuclear energy $E(=E_1+E_2)$ has been calculated using the principle of conservation of energy

$$E=M^*(A,Z)-M(A_1,Z_1)-M(A_2,Z_2)-K-D \quad , \tag{2}$$

where $M^*(A,Z)$ is the mass of the excited compound nucleus undergoing fission and $M(A_1,Z_1)$ and $M(A_2,Z_2)$ are masses of the fission fragments in their ground states. K is the total kinetic energy of the fragments and D is the total deformation energy of the fragments. So, E is different for different mass divisions, different charge divisions, and different kinetic energy values. Correspondingly, the fissioning nucleus exhibits a mass distribution, charge distribution, and kinetic energy distribution of fission fragments.

Although Fong's theory was successful in reproducing the mass distribution curve for fissioning of the compound system ^{236}U, it failed in other cases.[8] For instance, it yielded a four-humped curve for ^{240}Pu instead of the observed two-humped behavior. Furthermore, Fong's theory does not yield the fine structure that is observed in the mass yield curve for fission of ^{235}U with thermal neutrons.

Fong's theory, in its original form, did not take into account the shell structure. In a later contribution,[9] he introduced the shell effects through the two-center shell model calculation, but there, too, the dependence of the single particle level density parameter a on shell structure and nuclear temperature was not included.[10]

Ignatyuk[11] tried to use the level density of Ericson[12] which includes the pairing effects, but the results obtained were not encouraging in the sense that they did not reproduce the experimentally observed most probable fission fragments. One reason for its failure may be that the thermal equilibrium which is the basic assumption in statistical fission theories has not been strictly observed in the calculations. Furthermore, the constant values of the Nilsson parameters used by the author for all shells could possibly have affected the calculations and lead to wrong results. These two deficiencies have been removed in the present calculations in which we use the well tested parameters of Seeger[13]; also the temperature of the fragments has been kept constant. These modifications have yielded good results which are in qualitative agreement with experimental values. For example, in the case of ^{236}U in which fine structures (see Ref. 17) are observed in the yield curve between masses 134 and 144, the present theory reproduces the fine structures well, although the yield of the most probable fission fragment is slightly enhanced. The temperatures used are less than 2 MeV, for which the shell effects remain pronounced. The excitation energies for different fragmentation vary within 10 MeV of the average value which is within the shell correction limits.

Bethe's formula for nuclear level density is used and the excitation energy is calculated from the microscopic single-particle energy spectrum. Thereby, the shell structure of the fragments is incorporated in the theory. Besides, the nuclear temperature serves as a convenient parameter to determine the total excitation energy as well as excitation energies of the individual fragments. With these modifications, the statistical theory yields the observed mass distribution curves for ^{236}U, ^{240}Pu, and ^{258}Fm. Also, the study of quasifission reaction compounds, such as ^{84}Kr + ^{238}U and ^{129}Xe + ^{197}Au, yields results in agreement with earlier theories and experiments.

II. THE METHOD

The probability of nuclear fission P in a particular mode yielding fragments (A_1,Z_1) and (A_2,Z_2) is proportional to the product of nuclear level densities ρ_1 and ρ_2 of the fission fragments

$$P \propto \rho_1\rho_2 \ . \tag{3}$$

The nuclear level density $\rho(E)$ in turn can be expressed as a function of excitation energy E by means of Bethe's formula

$$\rho(E) = \tfrac{1}{12}(\pi^2/a)^{1/4}E^{-5/4}\exp(2\sqrt{aE}\,) \ , \tag{4}$$

where a is the single-particle level density parameter given by[14]

$$a = (\pi^2/6)g_0 = E/T^2 \tag{5}$$

with

$$g_0 = g_p(\epsilon_F^p) + g_n(\epsilon_F^n) \ . \tag{6}$$

The nuclear temperature T should be the same for both fragments since they are in thermal equilibrium at the fissioning stage. g_p and g_n are the single-particle level densities for protons and neutrons, respectively, and ϵ_F^p and ϵ_F^n are the corresponding Fermi energies. The excitation energy E is given by

$$E = \sum_i n_i\epsilon_i - \sum_i^{N,Z} \epsilon_i \ , \tag{7}$$

where n_i is the fermion occupation probability of the single-particle state i with energy ϵ_i.

$$n_i = \{\, 1 + \exp[(\epsilon_i - \epsilon_F)/T]\,\}^{-1} \ . \tag{8}$$

The method of calculation is outlined below. The single-particle levels ϵ_i are generated using the Nilsson Hamiltonian as given in Refs. 15 and 16. The Hamiltonian is diagonalized using a cylindrical basis and Seeger parameters[11] are used for the $l \cdot s$ and l^2 terms. This allows the use of single-particle levels over a wide range of nuclei. Then the excitation energies E_1 and E_2 of the two fragments are calculated using expressions (7) and (8) at any given nuclear temperature T. The single-particle level density parameters a_1 and a_2 are obtained from Eq.

(5). The probability of binary fission is then calculated using (3) and (4) as a function of the mass numbers of the fission fragments under the usual assumption that the charge to mass ratio of the fission fragments is the same as that of the parent system,

$$\frac{Z_1}{A_1}=\frac{Z_2}{A_2}=\frac{Z_1+Z_2}{A_1+A_2}\ . \tag{9}$$

III. RESULTS AND DISCUSSION

The yield curves obtained in the present calculation are depicted in Figs. 1–4. For ^{236}U, the yield curve peaks at mass numbers 102 and 134, which are the most probable fission fragments obtained experimentally.[17,18] The peak to valley ratio in the yield curve is large but the other factors which have not been considered here will affect these minor details, and future calculations will show how an agreement can be best obtained. Besides, the present theory is successful in reproducing the pronounced single hump for ^{258}Fm and the double hump with side shoulders for ^{240}Pu. The finer fluctuations on the double hump in Fig. 3 appear at first glance to correspond to a four-humped yield curve, but on comparing the theoretical curve with

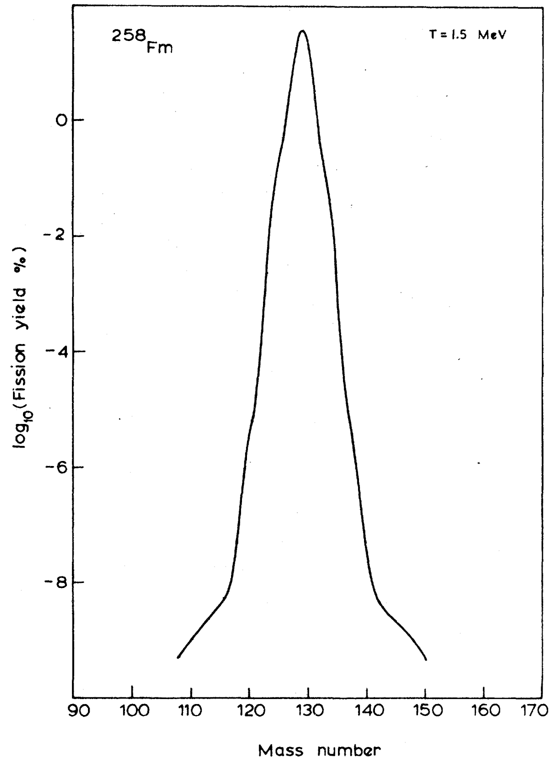

FIG. 2. The mass yield curve for ^{258}Fm at the nuclear temperature $T = 1.5$ MeV.

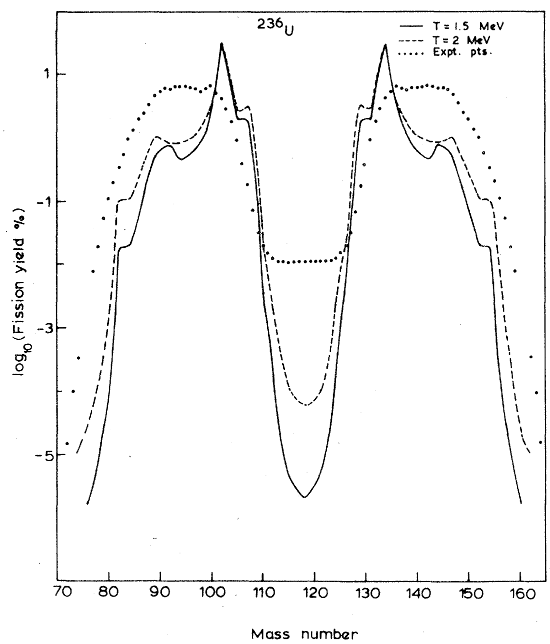

FIG. 1. The mass yield curve for ^{236}U at nuclear temperatures $T = 1.5$ and 2 MeV; the experimental points correspond to fission of ^{235}U by thermal neutrons and are taken from Ref. 17.

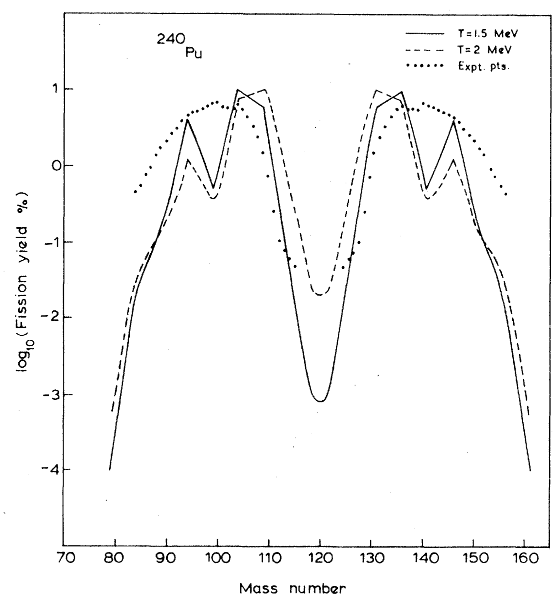

FIG. 3. The mass yield curve for ^{240}Pu at nuclear temperatures $T = 1.5$ and 2 MeV. The most probable fission fragment is 104 as reported in Ref. 19. The experimental points correspond to fission of ^{239}Pu by thermal neutrons.

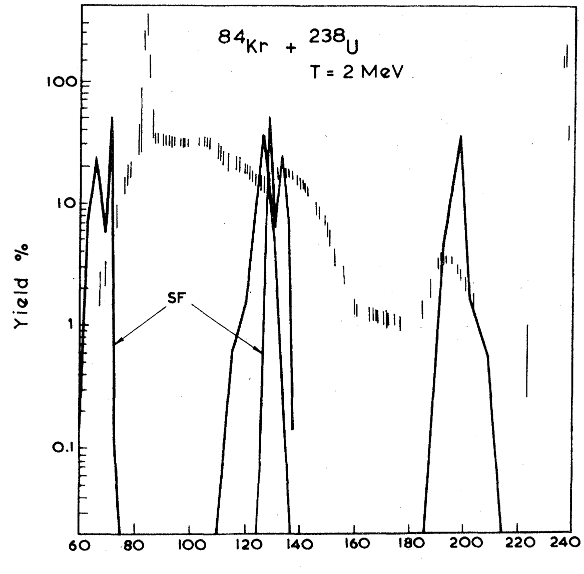

FIG. 4. The mass yield curve for the quasifission reaction $^{84}Kr + ^{238}U$ at a temperature $T = 2$ MeV. The sequential fission (SF) curve for the mass number $A = 198$ is also shown. The experimental data correspond to projectile energy $E_{\rm lab} = 600$ MeV and are taken from Ref. 20.

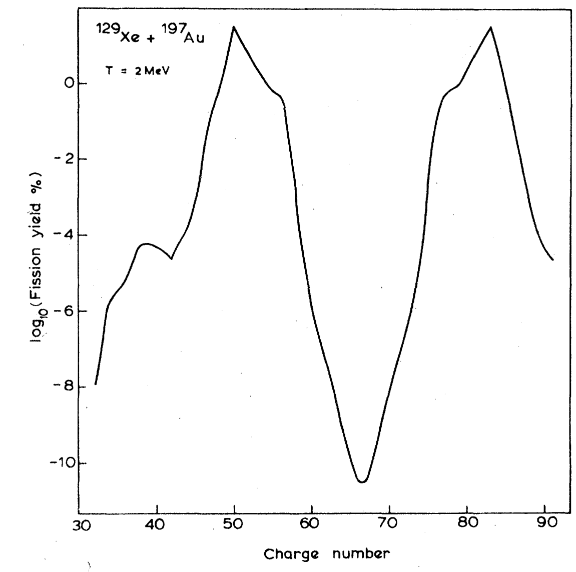

FIG. 5. The charge distribution curve for the quasifission reaction $^{129}Xe + ^{197}Au$ at a temperature $T = 2$ MeV. The most probable charge numbers of the fission fragments obtained are in agreement with the data reported in Ref. 20. The experimental data correspond to projectile energy $E_{\rm lab} = 761$ MeV.

the experimental yield curve,[19] one observes that they are only the finer details not fully observed in experiments. These fluctuations tend to vanish at slightly higher temperatures. On the other hand, the theoretical curve of excitation energy versus fragment mass ratio obtained in Ref. 8 shows four humps, well separated, implying disagreement with experiment.

The study has been extended to the quasifission reaction[20,21] $^{84}Kr + ^{238}U$. The theoretical mass yield curve in Fig. 4 shows two peaks, one at mass number 125 and the other at 197. If one takes into account the sequential fission of the heavier fragment, it is possible to realize the experimental curve. In Fig. 4, a specific case of sequential fission of the fragment with mass number 198 is shown. The charge distribution in the quasifission reaction of $^{129}Xe + ^{197}Au$ has been nicely reproduced[22] (see Fig. 5).

As is usually the practice in all fission calculations, the present theory assumes the ideal situation of binary fission and does not consider the prompt neutron emission. Although the inclusion of the emission of 2—3 prompt neutrons is beset with theoretical uncertainty, it may not significantly alter the yield curves but cause only small variations in the mass-yield distributions of light and heavy fragments as observed by Farrar and Tomlinson.[17]

The success of the modified statistical theory outlined here is essentially due to (1) the extraction of the single-particle level density parameter from the single-particle energy spectrum and (2) the inclusion of the dependence of the parameter on the nuclear temperature of the fragments. The latter incorporates into the theory in a natural way the dependence of the mass distribution of the fission fragments on the excitation energy of the fissioning nucleus.

Still the theory is a naive one and further refinements are possible. One may study the effect of inclusion of pairing[23] in the level density calculations and also the change in the probability of fission yield with respect to the deformation of the fragments. If one takes into account deformed fragments there is no *a priori* rule to suppose that the fissioning fragments are already in their equilibrium shape and all that can be done is to allow the deformation degree of freedom to vary for each fragment combination and sum the probabilities over the deformation parameters α_1 following Wilkins *et al.*[6] The yield will then be

$$Y(A_1,A_2) \propto \sum_{ij} \rho_1(A_1,\alpha_i)\rho_2(A_2,\alpha_j) \ . \tag{10}$$

These refinements are expected to modulate the yield curves only slightly but the main results of this paper will not be affected. However, as pointed out by Wigner,[24] there is a conceptual difficulty in understanding the nuclear level density of a deformed excited nucleus, since "in the excited state the nucleus will have different deformations." So, we have refrained from including the deformation degree of freedom in our calculation and the yield curves presented here are obtained by assuming the fragments to be spherical. Besides, the yield curves reported in this paper correspond to nuclear temperatures $T = 1.5$ and 2 MeV and consequently, the excitations of the nuclei are sufficiently large for the results to be affected by pairing correlations.

The study can also be extended to ternary fission and to rotating fragments. It is possible to introduce the angular momentum degree of freedom to the fissioning fragments by using the partition function of the rotating nuclei.[25]

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