Fine structure in nuclear fission charge distribution yields and the odd-even charge effects as the dynamical fragmentation process

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The fragmentation theory is used to study the fine structure effects in nuclear fission charge distribution yields. The odd-even proton effects as well as the resulting non-Gaussian charge distributions are shown to be given satisfactorily, without use of any free parameter. Calculations are made for light mass products 98, 100, 102, and 104 due to thermal neutron induced fission of ^{235}U.

NUCLEAR REACTIONS, FISSION Charge distributions, non-Gaussian, fine structure, odd-even proton effects, post saddle shape, fragmentation dynamics, light mass chains, ^{236}U.

The odd-even effects due to pairing were first observed by Thomas and Vandenbosch[1] in the fine structure of the low energy fission fragment (primary fragments) yields derived from the fission fragment kinetic energy spectra. The observed structure is correlated with the structure in the energy release calculated on a semiempirical mass formula. These authors also observed a persistence of some of this structure even at high excitation energies, though the small energy differences should then have less influence on the fragment yield. This observation is taken to suggest a preference for the formation of even-even nuclei as primary fission fragments. Recently, a detailed experimental study of the odd-even effects in fission yields has been made[2–6] for the thermal neutron induced fission of ^{233,235}U, ^{229}Th, and ^{239,241}Pu, the spontaneous fission of ^{252}Cf and 3 MeV neutrons on ^{235}U. The odd-even effects are found to be present even at high excitation energies, though with a drastically decreased amplitude. These effects are now considered[6] as one of the spectacular characteristics of low energy fission that can be used to put strong constraints on the various fission theories.

Thomas and Vandenbosch[1] had observed and Mariolopoulos *et al.*[6] have shown that the statistical theories such as the Fong's model[7] do not predict any odd-even effects on charge distribution yields. Fong's model is a scission-point model that includes no pairing term in the potential at the scission. These authors[6] have also shown that another scission-point model due to Wilkins, Steinberg, and Chasman,[8] which uses the deformation and intrinsic temperature (the single-particle excitation) dependent pairing and shell corrections, also does not account for the observed amplitudes and the variations with excitation energies of the odd-even effects for a reasonable choice of the parameters of this model. This model is also a statistical model, assuming a quasiequilibrium among collective degrees of freedom. Fong assumes a complete statistical equilibrium between the two nascent fragments. The sharp decrease of odd-even effects when the excitation energy is increased can, however, be qualitatively explained on another version of the quasiequilibrium statistical model[9] by allowing four-quasiparticle excitations in the case of increased excitation energy (3 MeV neutron on ^{235}U) above the saddle point. The model assumes a conservation of the number of two-quasiparticle excitations during the transition from saddle to scission point. In other words, this model predicts a very large probability of at least one proton pair breaking for 3 MeV neutron excitation compared with a vanishing small value for thermal neutron fission. A careful analysis[6] of the experimental data, however, shows that the probability of pair breaking is rather small and, if present, should occur at a rather late stage past the saddle point (almost near the scission point). Actually, a fission of a completely proton paired system is observed in a significant fraction of the cases.

In this paper we give an alternative description of the odd-even charge effects in terms of the fragmentation theory of one of the authors, Scheid, and Greiner,[10] which treats the charge asymmetry between the heavy (H) and light (L) fragments

$$\eta_Z=\frac{Z_H-Z_L}{Z_H+Z_L};\quad Z=Z_H+Z_L \tag{1}$$

as a dynamical collective coordinate of the fissioning dinuclear system. It is shown that odd-even effects are obtained in a natural process of fragmentation at a point just past the saddle point[10,11] and without including any pair breaking effects explicitly. This is the first time that a theory is shown to give non-Gaussian charge distributions that compare with experiments nicely without any free parameter fitting.

In the fragmentation theory,[10] the nuclear shape (see Fig. 1 of Ref. 10) is described in terms of the parameters of the asymmetric two-center shell model (ATCSM) and the stationary Schrödinger equation in η_Z coordinate,

$$\left\{-\frac{\hbar^2}{2(B_{\eta_Z\eta_Z})^{1/2}}\frac{\partial}{\partial\eta_Z}\frac{1}{(B_{\eta_Z\eta_Z})^{1/2}}\frac{\partial}{\partial\eta_Z}+V(\eta_Z)\right\}\psi^{(\nu)}_{\lambda\eta_A}(\eta_Z)=E^{(\nu)}_{\lambda\eta_A}\,\psi^{(\nu)}_{\lambda\eta_A}(\eta_Z)\ , \tag{2}$$

is solved numerically at the fixed elongation λ and mass asymmetry

$$\eta_A=(A_H-A_L)/(A_H+A_L);\quad A=A_H+A_L\ .$$

The main behavior of the mass and charge distributions is fixed at λ value just after the barrier penetration has occurred (see also Refs. 11 and 12). The potential $V(\eta_Z)$ is obtained under adiabatic approximations from the ATCSM in the Strutinsky method.[13] The mass parameters B_{ij}, defining the kinetic energy part of the Hamiltonian, are obtained consistently from the Cranking formula in BCS formalism.[14] The protons and neutrons are taken to move in two separate single-particle potentials of the ATCSM, such that the coupling mass $B_{\eta_Z \eta_N}=0$, where

$$\eta_N=\frac{N_H-N_L}{N_H+N_L};\ N_H+N_L=N \ .$$

The solution of Eq. (2) gives the probability $|\psi_{\lambda\eta_A}(\eta_Z)|^2$ of finding a charge fragmentation η_Z at the fixed position λ of the fission path with fixed mass asymmetry η_A. This probability, when scaled to a percentage charge yield Y at a charge number, say, Z_L of one fragment $(d\eta_Z=2/Z)$, gives

$$Y(Z_L)=|\psi_{\lambda\eta_A}[\eta_Z(Z_L)]|^2(B_{\eta_Z\eta_Z})^{1/2}\frac{200}{Z} \ . \quad (3)$$

The states $\psi_{\lambda\eta_A}^{(\nu)}$ are the vibrational states in the potential V, counted by the quantum number $\nu=0,1,2,\ldots$. In spontaneous fission, for complete adiabaticity and starting from the nuclear ground state, only the lowest vibrational state $\nu=0$ may be occupied. However, for fission from excited states or due to interaction of η_Z with other degrees of freedom, the effect of higher excited states may be important. For a Boltzmann-type occupation of excited states,

$$|\psi_{\lambda\eta_A}|^2=\left|\sum_{\nu=0}^{\infty}\psi_{\lambda\eta_A}^{(\nu)}(\eta_Z)\right|^2\exp(-E_{\lambda\eta_A}^{(\nu)}/\theta) \ , \quad (4)$$

the charge distribution yields are shown[10] to be independent of nuclear temperature $\theta \leq 7$ MeV. Apparently, the present experimental data,[3,6] showing sharp decrease of odd-even effects with increase of excitation energy, require some other form of temperature dependence. One possibility is to use the temperature dependent potential and mass parameters[15] that is considered here for estimating its importance for the odd-even effects.

We have chosen to apply this theory to the cases of light mass products $A_L=98$, 100, 102, and 104 obtained in the thermal neutron induced fission of ^{235}U. The measured charge distributions for these mass chains could not be described[2] by Gaussian functions. The mean values $\bar{Z}$, defined as the average nuclear charge, are always larger than the UCD (the unchanged charge distribution) estimates, $Z_{\rm UCD}$. The variances σ, determined as the square root of the second moment, oscillate around the average value 0.55, and this oscillation is shown to be closely related to the distance $|Z_e-\bar{Z}|$, where Z_e is the neighboring even integer charge number. The proton pairing or the odd-even proton effects are clearly indicated since the charge distribution for $A_L=100$ is very narrow and symmetric around $\bar{Z}=40$ and becomes strongly asymmetric as one moves down or up by only two units of mass to $A_L=98$ or 102. The charge distribution for $A_L=98$ extends towards the low average nuclear charge ($\bar{Z}<40$) and for $A_L=102$ towards high $\bar{Z}(>40)$. For $A_L=104$, the charge distribution is again asymmetric with $\bar{Z}<42$. We shall see in the following that all these fine structure effects of the charge distributions are given within the fragmentation theory.

Figures 1 and 2 give, respectively, the charge dispersion potentials $V(Z_L)$ and the mass parameters $B_{\eta_Z\eta_Z}(Z_L)$ for the light mass products $A_L=98$, 100, 102, and 104 of ^{236}U

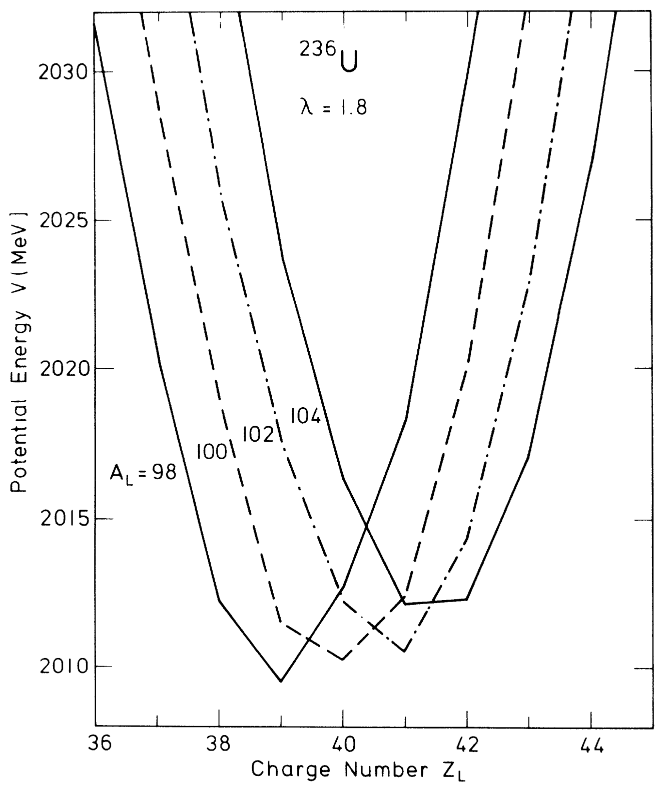

FIG. 1. Charge dispersion potentials for light mass fission products $A_L=98$, 100, 102, and 104 from ^{236}U at the length $\lambda=1.8$, calculated on the asymmetric two-center shell model. For actual calculations, the product mass is increased by the number of neutrons evaporated, as per Eq. (6).

calculated for $\lambda=1.8$ at an interval of $\Delta\eta_z=2/Z\approx 0.02$. This value of $\Delta\eta_z$ is small enough to consider η_z as a continuous variable. The nuclear shape at this length of the fissioning nucleus is illustrated in Fig. 1 of Ref. 10. The effect of neutron evaporation is included by increasing the product mass A_L by the average number ν_L of neutrons emitted. The primary fragment mass A_L' is then

$$A_L'=A_L+\nu_L \ , \quad (5)$$

where ν_L is estimated from the following functional relation:[16]

$$\nu_L=0.531\nu_T+0.062(A_L+143-A) \ , \quad (6)$$

constructed to represent the composite data for the average number of neutrons emitted per fragment in the thermal neutron fission of ^{233,235}U and the spontaneous fission of ^{252}Cf. ν_T is the total number of neutrons emitted per fission. For thermal neutron induced fission of ^{235}U, $\nu_T=2.5$ on the average.

We notice from Fig. 1 that the potential energy surfaces (PES) for product masses $A_L=98$, 100, and 102 are nearly symmetric at $Z_L=39$, 40, and 41, respectively, and asymmetric for $A_L=104$. Thus this structure of PES compares with the observed structure of charge distribution yields only for $A_L=100$ and 104. This interpretation is similar to that used by Thomas and Vandenbosch.[1] We also notice in Fig. 2 that the mass parameters $B_{\eta_Z\eta_Z}$ oscillate with Z_L and some of the points, marked crosses (×), could not be

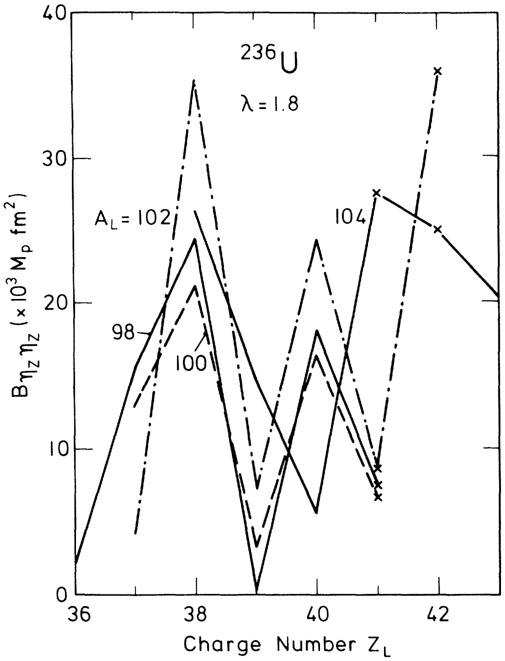

FIG. 2. Cranking mass parameters $B_{\eta_Z \eta_Z}$ for the fission products $A_L = 98$, 100, 102, and 104 from ^{236}U at $\lambda = 1.8$. The crosses (×) indicate the empirically fitted values that were otherwise obtained to be zero.

evaluated from the Cranking formula. The Cranking formula resulted in zero values due to the improper labeling of the single-particle states in the ATCSM. This happens at elongations where quasicrossings of the adiabatic single-particle levels occur and the Landau-Zener effect becomes important. In that case one actually requires to choose a diabatic single-particle basis where the quasicrossings become the real crossings.[17] Within the adiabatic basis, however, we have obtained the mass parameters at these points empirically by starting with a value close to the average value. The result of taking an average value for the mass parameter $\bar{B}_{\eta_Z \eta_Z}$ is also studied. It may be mentioned that, in contrast to the observed oscillations in Fig. 2, the diabatic Cranking masses[18] are smooth functions of Z_L.

Figure 3 gives the calculated percentage fission yields $Y(Z_L)$, compared with experimental data,[2] for all the four mass chains. The experimental data[2] are for the thermal neutron induced fission of ^{235}U, measured at the most probable kinetic energy of the fission products. The effect of in-

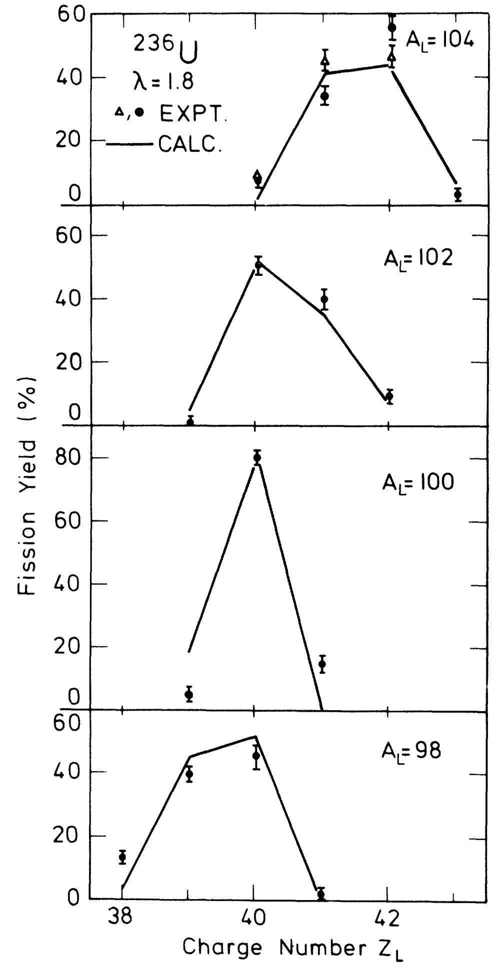

FIG. 3. Calculated percentage yields compared with the experimental data. The data indicated as circles (O) are for the most probable kinetic energy in each case and as triangles (Δ) in the case of $A_L = 104$ are for a higher kinetic energy.

crease of kinetic energy (i.e., decrease of excitation energy) on the data[3] is also indicated in Fig. 3 for the illustrative case of $A_L = 104$. It is interesting to observe that the fine structures of the fission yields in all the four cases are nicely reproduced. The agreement is shown to improve for the high kinetic energy data. This is expected since our calculations are for the ground state. We obtain the symmetric as well as the asymmetric charge distributions without fitting

TABLE I. Theoretical and the experimental (Ref. 2) parameters of charge distributions for light mass products A_L.

		$\bar{Z}$		σ		$Z_{UCD} - \bar{Z}$[a]		$\|Z_e - \bar{Z}\|$	
A_L	A_L'	Expt[b]	Theory	Expt[c]	Theory	Expt	Theory	Expt	Theory
98	99.64	39.36 ± 0.10	39.49	0.73 ± 0.02	0.56	−0.52	−0.65	0.64	0.51
100	101.76	40.10 ± 0.07	39.82	0.43 ± 0.03	0.39	−0.43	−0.15	0.10	0.18
102	103.89	40.59 ± 0.08	40.44	0.66 ± 0.02	0.70	−0.09	0.06	0.41	0.44
104	106.01	41.56 ± 0.10	41.67	0.66 ± 0.04	0.71	−0.23	−0.34	0.44	0.33

[a] Z_{UCD} is defined as $Z/A = Z_{UCD}/A_L'$.
[b] At a higher kinetic energy, slightly smaller values are obtained (see Ref. 3).
[c] At a higher kinetic energy, slightly larger values are obtained for $A_L = 98$ and 100 but smaller for $A_L = 102$ and 104 (see Ref. 3).

any parameter. Thus, the role of mass parameters is clearly depicted. In other words, the fine structure in fission yields is a dynamical effect of the fragmentation process. The element having an even proton number is produced more abundantly in comparison with its two neighbors having odd proton numbers. Correspondingly, for product masses with an odd mean Z value, the charge distribution is broad, and for masses with even $\bar{Z}$, it is narrow. Thus the odd-even effect leads to a non-Gaussian form of the charge distribution.

The calculated mean values $\bar{Z}$ (the first moment), the widths σ (the square root of the second moment), and their deviations from $Z_{\rm UCD}$ and Z_e values are compared in Table I with the corresponding numbers obtained from the experimental data.[2] The agreement is reasonably good. The observed oscillations of σ, related with the distance $|Z_e-\bar{Z}|$, are also supported by our calculations, as shown in Fig. 4. A more realistic comparison will, however, be possible only after the parameters for odd product masses are also calculated.

We have also estimated the summed yields of even-Z elements to the average summed yields $[\sum_e/\frac{1}{2}(\sum_e+\sum_0)]$ and the corresponding quantity for the odd-Z elements $[\sum_0/\frac{1}{2}(\sum_e+\sum_0)]$, that give a quantitative measure of odd-even charge effect. Our values, calculated for the interval $38\leq Z_L\leq 43$, are

$$\frac{\sum_e}{\frac{1}{2}(\sum_e+\sum_0)}=1.21,\quad \frac{\sum_0}{\frac{1}{2}(\sum_e+\sum_0)}=0.79\ , \tag{7}$$

that compare almost exactly with the experimental numbers 1.22 ± 0.01 and 0.78 ± 0.01, respectively, of Clerc *et al.*[2] In order to see the effect of temperature, we have also estimated these quantities for the case of complete washing away of the shell corrections (i.e., taking the liquid drop potential only). The corresponding numbers are 1.10 and 0.90, showing a considerable decrease in the odd-even effect. The odd-even effect is found to decrease further if the mass parameters are allowed to vary (e.g., taking almost smoothed function) or an average mass parameter $\bar{B}_{\eta_Z\eta_Z}$ is used.

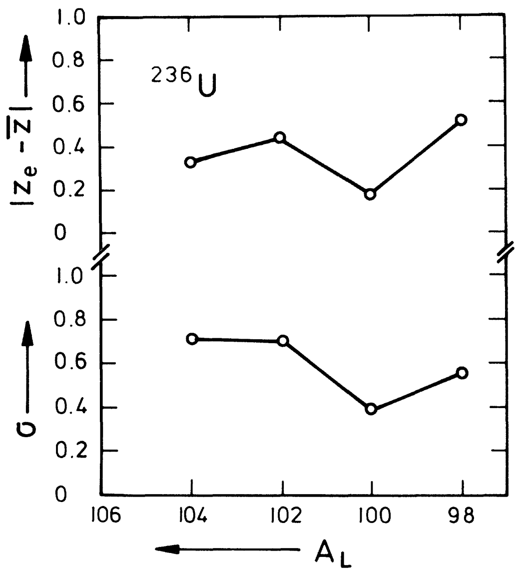

FIG. 4. The distance of the mean value $\bar{Z}$ from the neighboring even number Z_e and the variance σ calculated as a square root of the second moment, plotted as a function of the fission product mass A_L.

Concluding, we have shown that the fine structure in nuclear fission charge distribution yields, resulting in odd-even proton effect, is nicely reproduced within the fragmentation theory. There is no free parameter to be fitted. The odd-even proton effect that leads to non-Gaussian charge distributions is shown to be a dynamical fragmentation process fixed at a "post saddle point," just after the barrier penetration.

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