

SADDLE AND SCISSION STATES EXPECTED FROM FINE STRUCTURE OF NUCLEAR CHARGE DISTRIBUTION BASED ON SEMI-EMPIRICAL THEORY

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ABSTRACT: The nuclear charge distribution function is derived from statistical theory as a function of the most probable charge transfer variable defined by the most stable charges of fission fragments. The fine structures observed in the charge distribution width, skewness and excess can be described by the present formula in terms of charge shift due to pairing effects, as well as in asymmetric terms of nuclear mass. According to an investigation on the fine structure by a semi-empirical method, some remarks are given for the saddle and scission states.

[Charge distribution, Saddle and scission states, fine structure]

I. INTRODUCTION

Nuclear charge distribution is of interest for better understanding fission mechanisms in nuclear physics, as well as being fundamental for estimating isotopic fission yield in the nuclear chemistry. The fractional independent yield based on Gaussian distribution function empirically obtained by Wahl et al.¹⁾, without the correction factor for pairing effect, has been used for a long time. The fractional yields estimated by this function are consistent with the observed yields in the gross structure. The even-odd effect, however, which was reported by Amiel and Feldstein²⁾, and the measurements of fission -yield by Clerc et al.³⁾ and Siegert et al.⁴⁾ with the high resolution mass separator, implied that the charge distribution width, as well as other fine structure constants; skewness and excess, are functions of mass number.

The present work aims at deriving the charge distribution function and its fine structure constants based on statistical theory, and then drawing some conclusion about the saddle and scission states from the experimental evidences, with the help of the present theory.

II. CHARGE DISTRIBUTION FUNCTION

Fission probability for a pair of fission fragments, denoted by subscripts 1 and 2, is expressed by the statistical model

$$P = C_p \int_0^Q T(E) \int_0^{Q-E} \rho_1(E_1^*) \rho_2(Q-E-E_1^*) dE_1^* dE \quad (1)$$

under the assumption that the angular distribution of outgoing fragments is uniform, where E_1^* means the excitation energy of the fission fragment denoted by subscript 1, and Q is the total energy release in the fission. The level density function ρ_i ($i=1,2$) is approximated by the constant temperature model⁵⁾

$$\rho_i^{(c)} = C \exp[(E_1^* - E_{0i})/T_i] / T_i \quad (2)$$

with the nuclear temperature T_i and energy shift E_{0i} due to the pairing effect. The temperature T_i ($i=1,2$), however, is estimated from the equation combining the level densities $\rho_i^{(c)}$ based on constant temperature

model with that based on Fermi gas model⁶⁾ with a_i and $U_{\pi i}$ defined by

$$\frac{1}{T_i} = \left(\frac{a_i}{U_{\pi i}} \right)^{1/2} - \frac{3}{2U_{\pi i}} \quad (3)$$

where the constant a_i is obtained from the empirical formula of Gilbert and Cameron⁷⁾ as a function of shell energy term S . After the derivation of the present charge distribution function, the shell energy $S(N,Z)$ used for the level density parameter a_i is replaced by the shell energy expression of Myers and Swiatecki⁸⁾ in order to take into account the deformation effect. The deformation parameter (θ)⁹⁾ is obtained by simplex method applied to the most stable nucleus in the present work.

The transmission coefficient $T(E)$ defined by Ericson⁷⁾ is used for the present work;

$$T(E) = [1 + \exp(\alpha(E_0 - E))]^{-1} \quad (4)$$

where E_0 , E and α mean the Coulomb energy, kinetic energy and the curvature of the Coulomb barrier, respectively.

The fission probability defined by eq.(1) can be expressed by a pair of symmetric terms with respect to an average excitation energy Q_a as,

$$P = C_p \int_0^Q T(E) \int_{-Q_a}^{+Q_a} \rho_1(Q_a + \eta) \rho_2(Q_a - \eta) d\eta dE, \quad (5)$$

$$\text{with } Q_a = (Q-E)/2. \quad (6)$$

This is separated into two components for the fission fragments denoted by subscripts 1 and 2 as,

$$P = C_{\text{asym}} \cdot (P_2 - P_1) \quad (7)$$

where C_{asym} means normalization constant.

The partial probability P_i ($i=1,2$) has the following compact form:

$$P_i(Q) = \int_0^Q T(E) \exp[2\zeta_i(Q - \lambda_i E_0) + \eta_i] dE \quad (8)$$

where ζ_i , λ_i and η_i are constants determined by the nuclear temperatures, the coefficient of Coulomb energy and pairing energies for level density.

The integration eq.(8) cannot be analytically performed since the constant τ_i

is not an integer value, in general, but by introducing the intermediate function $J(\tau_1, x_0)$, it can be expressed by a Gaussian hyper-geometric function ${}_2F_1(\alpha; \beta; \gamma; x)$ as shown below;

$$J(\tau_1, x_0) = \int_0^{x_0} \frac{\exp(\tau_1 x)}{1 + \exp(x)} dx \quad (9a)$$

$$= \frac{1}{x_0} \left[\left(\frac{\exp(x_0)}{1 + \exp(x_0)} \right) \right]$$

$${}_2F_1[1; 1; \tau_1 + 1; \left(\frac{\exp(x_0)}{1 + \exp(x_0)} \right)] \quad (9b)$$

with

$${}_2F_1(\alpha, \beta, \gamma, x) = \sum_{k=0}^{\infty} \frac{(\alpha)_k (\beta)_k}{(\gamma)_k} \frac{x^k}{k!} \quad (10)$$

$$(z)_k = \Gamma(z+k)/\Gamma(z) \quad (11), \quad (z)_0 = 1 \quad (12)$$

Consequently, the resultant charge distribution function becomes

$$P = C \exp \left[\frac{1}{2\tau} (Q - 2\alpha\tau E_0) \right] \sinh \left[\frac{Q}{2\epsilon} \right] \quad (13)$$

where the constants τ and ϵ^* are defined by the nuclear temperatures T_1 's as

$$1/\tau = 1/T_2 + 1/T_1, \quad 1/\epsilon = 1/T_2 - 1/T_1$$

The total energy release Q is approximately expressed in terms of the charge transfer variable ξ and constant term g by taking the first two terms of a Taylor expansion of the mass $M(A, Z)$ around the most stable charge Z_{A1} , i.e.,

$$Q = -K_{A1} (\xi - \xi_p)^2 + g \quad (14)$$

where

$$K_{A1} = \frac{1}{2} \left(\frac{\partial^2 M}{\partial Z^2} \right) \Big|_{Z=Z_{A1}} \quad (15)$$

$$K_{A1}^* = K_{A1} - \alpha\tau e^2 / r_0 D_{12} \quad (16a)$$

$$K_{A1}^* = K_{A1} + K_{A2}^* \quad (16b)$$

$$\xi = (Z_2 - Z_1) / 2 = Z_F / 2 - Z_1 = Z_2 - Z_F / 2 \quad (17)$$

where $e^2 / r_0 D_{12}$ means the coefficient of Coulomb interaction energy. The most probable charge transfer ξ_p introduced in eq.(17) is defined by the solution of the following equation;

$$\left(\frac{\partial P}{\partial \xi} \right) \Big|_{\xi = \xi_p} = 0 \quad (18)$$

The resultant solution for the most probable charge transfer and the most probable charge Z_{P1} and Z_{P2} are given by

$$\xi_p = [K_{A1}^* (0.5Z_F - Z_{A1}) - K_{A2}^* (0.5Z_F - Z_{A2})] / K_{A1}^* \quad (19)$$

$$Z_{P1} = [Z_{A2}^* Z_F - (K_{A2}^* Z_{A2} - K_{A1}^* Z_{A1})] / K_{A1}^* \quad (20)$$

$$Z_{P2} = [Z_{A2}^* Z_F + (K_{A2}^* Z_{A2} - K_{A1}^* Z_{A1})] / K_{A1}^* \quad (21)$$

As a special case, if the curvatures K_{A1}^* and K_{A2}^* are equal to one another, the Z_P -values tend to equal the result based on the ECD postulate,

$$Z_{P1} = [Z_F - (Z_{A2} - Z_{A1})] / 2 \quad (22a)$$

$$Z_{P2} = [Z_F + (Z_{A2} - Z_{A1})] / 2 \quad (22b)$$

Then, the probability P can be expressed by the Gaussian type function as

$$P = CN \exp[-\Omega(\xi - \xi_p)^2] \sinh[\Lambda(\xi - \xi_p) + \phi] \quad (23)$$

where CN means the normalization constant, and Ω and Λ indicate the constants giving the charge distribution width and the argument of the auxiliary function applied to the normal distribution, respectively.

III. FINE STRUCTURE AND FISSION STATES

The present charge distribution function as well as the fine structure constants, introduced by Siegert et al.⁴³, are compared with the experimental data obtained by high resolution mass separator. The observed fine structure constants are charge distribution width σ_A , the skewness S and the excess E , which are evaluated from the experimental fractional independent yield $P(A, Z)$ by using the following definitions:

$$\text{average charge: } Z_m = \int_0^{\infty} Z P(A, Z) dZ \quad (24a)$$

$$\text{width: } \sigma_A = \sqrt{\int_0^{\infty} (Z - Z_m)^2 P(A, Z) dZ} \quad (24b)$$

$$\text{skewness: } S = \frac{\int_0^{\infty} (Z - Z_m)^3 P(A, Z) dZ}{\sigma_A^3} \quad (24c)$$

$$\text{excess: } E = \frac{\int_0^{\infty} (Z - Z_m)^4 P(A, Z) dZ}{\sigma_A^4} - 3.0 \quad (24d)$$

The last two terms; S and E , indicate how consistent the the measured distribution function is with the normal distribution; i.e., if the interested charge distribution function belongs to a normal distribution with the center at the mean value Z_m , the skewness S vanishes, but if the center is not on the Z_m , the skewness has a non-vanishing value for which the sign depends upon the shift from the central value Z_m . The excess E indicates the intermediate shape parameter whose magnitude becomes zero if it is a completely normal distribution. Finally, the fractional charge distribution function can be expressed by these terms as

$$P = (1 + \epsilon) P_0 \quad (25)$$

where the deviation factor ϵ , usually called the pairing correction, can be defined by

$$\epsilon = \frac{S}{2\sqrt{2 \cdot 3!}} H_3(\Xi) + \frac{E}{96} H_4(\Xi) \quad (26)$$

$$\Xi = (Z - Z_m) / \sqrt{2} \sigma_A$$

and P_0 means the conventional charge distribution expressed by a Gaussian type function, and H_3 and H_4 are Hermite functions of the third and fourth orders, respectively.

The fine structure constants based on the present theory can be obtained by using the charge distribution function defined by eq.(23). The results are shown below.

$$\sigma_A = \left[\frac{1}{2\omega} + (\Delta Z_p)^2 \right]^{1/2} \quad (27a)$$

$$S = -3 \left(\frac{\omega}{2} \right)^{1/2} \frac{\Delta Z_p [1 + 2\omega (\Delta Z_p)^2 / 3]}{[1 + 2\omega (\Delta Z_p)^2]^{3/2}} \exp(\theta / 2) \quad (27b)$$

$$E = 3 \left[2 \frac{[1 + 4\omega (\Delta Z_p)^2 + 4\omega^2 (\Delta Z)^4 / 3]}{[1 + 2\omega (\Delta Z_p)^2]^2} \exp(\theta) - 1 \right] \quad (27c)$$

with $\omega = K_A/2\tau$ where θ indicates the normalization constant and ΔZ_p means the charge shift by a pairing energy effect.

To make a systematical investigation on the overall range of fission products, including the heavy fragments whose experimental data are not available in the literature, the most probable and stable charges are derived from the measured values for light-fragments. An outline of the "semi-empirical method" for derivation of the most probable and stable charges is shown below.

The most probable charge can be directly obtained from the charge conservation law; $Z_{p1} + Z_{p2} = Z_F$. Therefore, the most stable charges as the key parameters for the present theory are emphasized. The most stable charge Z_{A1} for the i -th fission fragment is the solution of the following equation²⁾

$$2K_{A1} (Z_{A1} - Z_{A1}^*) - \frac{5}{3} \mu_1 \{ Z_{A1}^{2/3} - (A_1 - Z_{A1})^{2/3} \} = 0 \quad (28)$$

where μ means the dumping factor for shell energy due to nuclear deformation as seen in the mass formula of Myers and Swiatecki³⁾. Besides, by taking into account the mass conservation law, a transcendental equation coupling two fragments is obtained as

$$\sum_{i=1}^2 [Z_{A1} + (Z_{A1}^{2/3} - \frac{6}{5\mu_1} K_{A1} (Z_{A1} - Z_{A1}^*))^{3/2}] = A_F - \nu \quad (29)$$

where A_F and Z_F mean the mass and the atomic number of the compound nucleus, and ν is the total number of prompt neutrons emitted from two fragments. By using the charge conservation law, the most stable charge Z_{A2} without any experimental data for Z_{p2} can be obtained from the experimental value $Z_{p1,exp}$ for the complementary nucleus as

$$Z_{A2} = \frac{K_{A2} Z_F + K_{A1} Z_{A1} - K_{A1} + K_{A2}}{K_{A2}} (Z_F - Z_{p1,exp}) \quad (30)$$

as the result of the relation between Z_A and Z_p as shown by eqs.(20) and (21). Therefore, the most stable charges Z_{A1} and Z_{A2} are obtained as the solution of simultaneous transcendental equations (29) and (30).

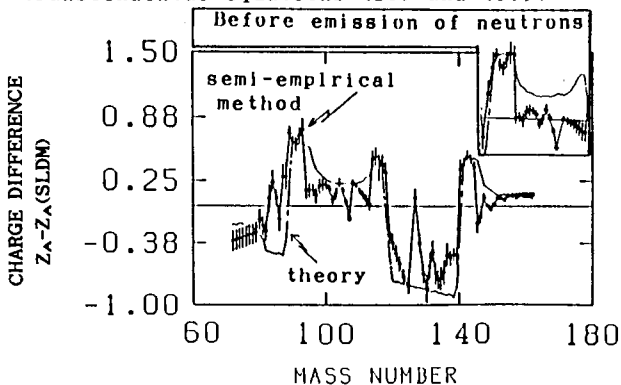


Fig. 1. The Most Stable Charge Z_A vs. Mass Number.

The semi-empirical and theoretical values of the most stable charge Z_A 's are shown in Fig. 1 as the deviation from those based on the simple liquid drop model (SLDM). In order to cover the overall mass-range of interest in reactor physics, some radiochemical data are supplemented in the mass region from

72 to 78 and 101 to 118 because of no available data obtained by mass separator.

As evident from Fig. 1, the gross structure of the most stable charge Z_A vs. mass number A curve due to shell and deformation effects are well traced by the theory. The fine structure, however, does not appear in the present result as a consequence of the missing the pairing energy terms in the Q -value estimation.

Theoretical and semi-empirical Z_A -values are significantly affected by the shell effect, especially in the neutron $N=50$ shell where shell and deformation effects show a concave trend. The $Z=50$ shell has no deformation effect since shell energy does not exceed the critical energy⁴⁾ and thus gives a linear trend.

Comparing the theoretical and semi-empirical Z_A -curves, the latter one, based on the experimental Z_F -value, shows a significant deformation effect, sharply cutting the line downward and then moving close to the simple liquid drop model (SLDM) baseline. This fact implies that the deformation effect on charge division in the scission state of fission seems to be impulsive rather than expected from $\exp(\theta^2)$ based on Myers and Swiatecki mass formula³⁾ or γ deformation neglected in their formula may be possible reason for the disagreement.

Before emission of neutrons Z_p -values are close to SLDM-value without shell effects, but after emission, the shell effects are emphasized. This seems to intimate that before emission of neutrons, maybe after saddle to scission states, the fissioning nucleus is "hot" and significantly deformed; consequently the effect of the shell structure is smeared.

The theoretical Z_p -curve, as shown in Fig. 2, is similar in shape to that based on the maximum energy release theory, but by using the semi-empirical Z_A -values, the predicted values are quite close to the experimental result.

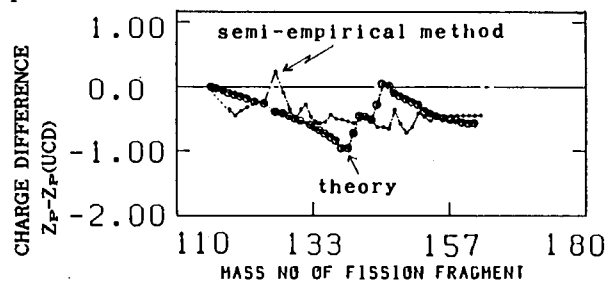


Fig. 2. The Most Probable Charge Z_p vs. Mass Number.

In order to predict the fine structure of the charge distribution by the present formula defined by eqs.(27a) to (27c), the term of the charge shift ΔZ_p is related with the reaction Q -value as suggested by Thomas and Vandenbosch⁵⁾. The Q -values for even Z nuclei and odd- A ones are shown in Fig.3 along with their differences denoted by δQ , which are evaluated from the mass table by Garvey et al.¹⁰⁾. As is well known, the Q -values for even- Z nuclei have local maxima and they are emphasized in δQ vs. mass number A curves, which are due to asymmetric terms in the

nuclear mass formula. The δQ -values as a function of mass number show a similar trend to that of the observed fine structure of charge distribution. Although at present a complete interpretation from a physical viewpoint between δQ and the charge shift parameter ΔZ_p has not yet established, the following assumption is made ;

$$\Delta Z = \frac{1}{1.55} \sqrt{1/\delta Q} \quad (31)$$

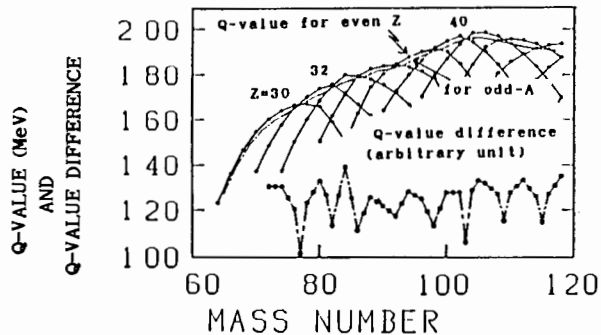


Fig 3. Q-value and The Deviation from Odd-mass Values as Function of Mass Number.

The fine structure constants evaluated by the present formula are shown in Figs. 4 to 6. The overall agreements are fairly well for three kinds of fine structure constants. For skewness S, however, the signs of ΔZ 's have to be changed at mass numbers 84, 88, 89, 93 and 94.

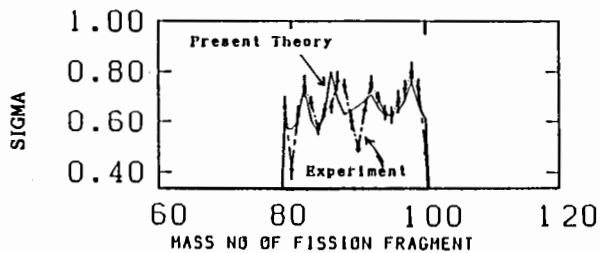


Fig. 4. Charge Distribution Width vs. Mass Number.

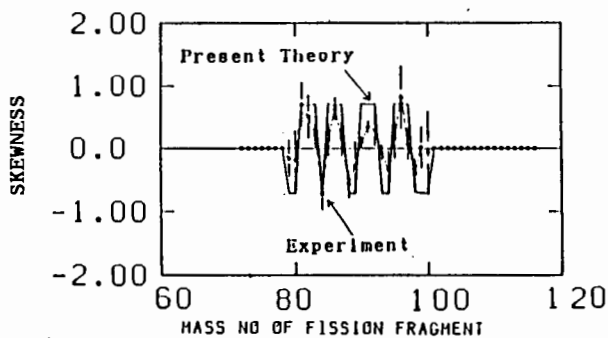


Fig. 5. Skewness vs. Mass Number.

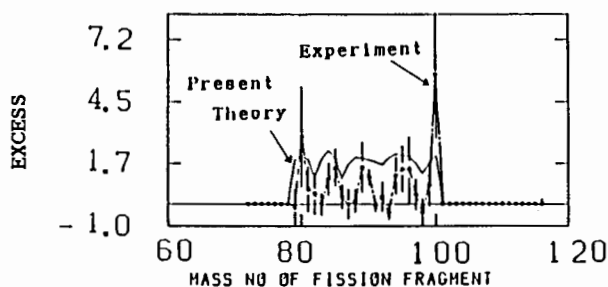


Fig. 6. Excess vs. Mass Number.

If the term for charge shift parameter ΔZ_p in the eq.(31) is missed, the charge distribution width σ_Λ -value is about 0.44 which is consistent with the X-ray measurement^{11,12}. The "hot" nucleus as the source of X-ray emissions has a "charge fluctuation" denoted by ΔZ_p due to pairing effects as well as asymmetric properties .

The semi-empirical result indicates that the pairing effect is not affected by deformation and this one seems to be characterized by the properties of the scission state.

Similarly, if the charge shift term is ignored, the other fine structure constants can not be interpreted since the nuclear temperatures are slowly varying variables as well as Fermi gas model parameters.

In the present theory, the physical properties joining the "forward process" of actual fission phenomena to the inverse one is the potential term and reaction Q-values. The latter one has been used for present work, but no through discussion has been made for the potential term including the saddle point. Further investigations are in progress in order to interpret the saddle point by using a modified Myers and Swiatecki Mass formula based on the present work.

IV. CONCLUDING REMARKS

The charge distribution function, based on the statistical theory, has been derived by using a parabolic approximation of the reaction Q-value as a function of the most probable charge transfer or the most stable charge.

The fine structure of the charge distribution obtained by a mass separator with high resolution was interpreted by the charge shift expected in the scission state. The constants for the fine structure were related to the pairing energy term used for the conventional Gaussian charge distribution function for which the width could be fairly well reproduced by present theory.

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