Theory of Charge Dispersion in Nuclear Fission

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By introducing charge asymmetry as a new dynamical collective coordinate in the asymmetric two-center shell model, the nuclear charge dispersion in the fission of ^{236}U is calculated without using any free parameter. The agreement between theory and experiment is quite good.

For the distribution of nuclear charge in fission, a number of empirical hypotheses such as that of equal charge distribution, unchanged charge density, and minimum potential energy have been proposed in the past by various authors.[1] Only recently, Holub, Mustafa, and Schmitt[2] have calculated the potential energy surface for the charge vibration in ^{236}U by using the two-spheroid liquid-drop model of Nix and Swiatecki and including the shell effects calculated in the Strutinsky prescription.[3] The single-particle states used to calculate the shell correction are those of a one-center Nilsson-type oscillator. In this paper we develop a theory for the charge dispersion in nuclear fission by using the concept of the charge-asymmetry coordinate treated as a dynamical coordinate in the asymmetric two-center shell model[4] (ATCSM). The theory is applied to the fission of ^{236}U and there is no free parameter to be fitted.

We consider the protons and neutrons as moving in two separate single-particle potentials of the ATCSM (Fig. 1) and define the proton- and neutron-asymmetry coordinates, respectively, as

$$\xi_Z = \frac{Z_1 - Z_2}{Z_1 + Z_2}, \quad \xi_N = \frac{N_1 - N_2}{N_1 + N_2}. \tag{1}$$

Z_1, Z_2 and N_1, N_2 are the respective proton and neutron numbers of the fragments obtained from the geometrical sizes of the fragments. The total volumes occupied by the Z $(=Z_1 + Z_2)$ protons and the N $(=N_1 + N_2)$ neutrons are assumed to be the same. The coordinates ξ_Z and ξ_N are, however, related to the mass-asymmetry coordinate ξ,

$$\xi = \frac{A_1 - A_2}{A_1 + A_2} = \frac{Z}{A}\xi_Z + \frac{N}{A}\xi_N, \tag{2}$$

and thus any two of these three coordinates are sufficient for treating as dynamical coordinates in the ATCSM. The other four coordinates used[5-8] to define the nuclear shape and thus the parameters of our potential are the total length of the nucleus λ $(=l/2R_0)$, or the distance R between the centers of mass of the two fragments, the deformations β_1 and β_2, and the necking-in parameter

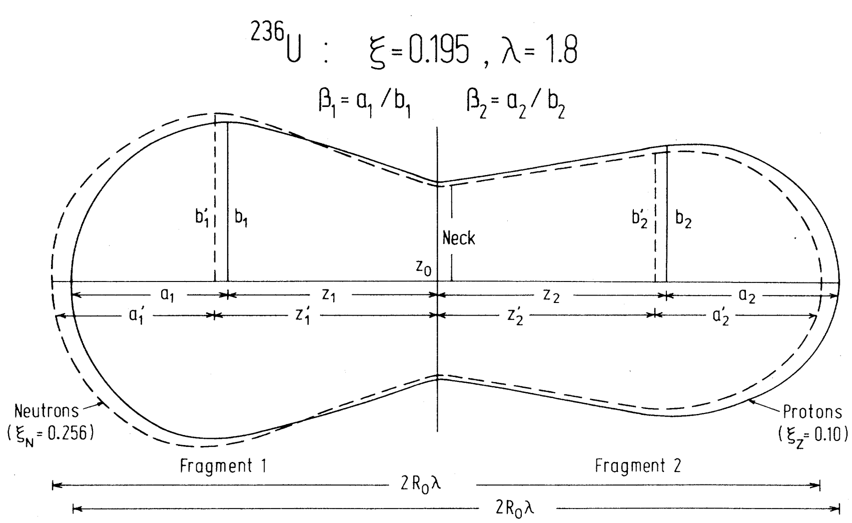

FIG. 1. Explanation of the parameters of the asymmetric two-center shell models for the protons and neutrons.

ϵ shown in Fig. 1.

The collective Hamiltonian is then written as

$$H=T(\lambda,\beta_1,\beta_2,\xi,\xi_Z,\dot\lambda,\dot\beta_1,\dot\beta_2,\dot\xi,\dot\xi_Z)+V(\lambda,\beta_1,\beta_2,\xi,\xi_Z). \tag{3}$$

We obtain the collective potential V from the single-particle levels $\epsilon_i(\lambda,\beta,\xi,\xi_Z)$ of the ATCSM by renormalizing the sum

$$\sum_{i=1}^{A}\epsilon_i$$

in the Strutinsky method[3] to the liquid-drop model of Myers and Swiatecki[9] with a modified surface-asymmetry constant.[10]

The mass parameters B_{ij} in the kinetic energy are consistently calculated by using the cranking formula in the BCS formalism[6-8]:

$$B_{x_ix_j}=2\hbar^2\sum_{\mu,\nu}\frac{\langle\mu|\partial H/\partial x_i|\nu\rangle\langle\nu|\partial H/\partial x_j|\mu\rangle}{(\tilde\epsilon_\mu+\tilde\epsilon_\nu)^3}(U_\mu V_\nu+U_\nu V_\mu)^2+P_{ij}, \tag{4}$$

where $\tilde\epsilon_\mu$ and $\tilde\epsilon_\nu$ are the quasiparticle energies and P_{ij} is a small correction term which accounts for the change of the Fermi surface and the energy gap due to deformation.[11]

Since in the fission process the motion in λ is at least approximately adiabatically slow compared to the motion in ϵ and β, and in order to simplify the problem, we minimize the potential in ϵ, β_1, and β_2 at each value of λ, ξ, and ξ_Z. By that we avoid a dynamical treatment in the coordinates ϵ, β_1, and β_2. Thus the collective energy can be written as

$$E=\tfrac12 B_{\lambda\lambda}\dot\lambda^2+\tfrac12 B_{\xi\xi}\dot\xi^2+\tfrac12 B_{\xi_Z\xi_Z}\dot\xi_Z^2+B_{\lambda\xi}\dot\lambda\dot\xi+B_{\lambda\xi_Z}\dot\lambda\dot\xi_Z+B_{\xi\xi_Z}\dot\xi\dot\xi_Z+V(\lambda,\xi,\xi_Z), \tag{5}$$

where the masses are functions of λ, ξ, and ξ_Z.

For the spontaneous fission and for the fission through the barrier, the motion in λ is slow after the system has tunneled through the barrier and has first begun to run down the Coulomb potential. Therefore, similarly as for the ξ motion,[8] we assume that the ξ_Z motion is fast compared to the λ motion. Furthermore, the potential has the characteristic that it remains nearly constant in its dependence on ξ and ξ_Z at later stages of λ, so that the main behavior of the distribution should be fixed at λ values just after the penetration of the barrier has occurred[6-8] (see also Mustafa, Mosel, and Schmitt[12]). Assuming complete adiabaticity, we can regard λ as a time-independent parameter. Also the coupling between the ξ and ξ_Z motion is weak, so that we can treat the ξ and ξ_Z motion as uncoupled in first approximation. The charge dispersion is then determined as a function of ξ_Z for the fixed λ and ξ values. The stationary Schrödinger equation, in which the coordinates λ and ξ enter only as parameters, is given by

$$\left(-\frac{\hbar^2}{2(B_{\xi_Z\xi_Z})^{1/2}}\frac{\partial}{\partial\xi_Z}\frac{1}{(B_{\xi_Z\xi_Z})^{1/2}}\frac{\partial}{\partial\xi_Z}+V(\lambda,\xi,\xi_Z)\right)\psi_{\lambda\xi}^{(\nu)}(\xi_Z)=E_{\lambda\xi}^{(\nu)}\psi_{\lambda\xi}^{(\nu)}(\xi_Z). \tag{6}$$

The λ and ξ dependences also enter through the mass parameters. The states $\psi_{\lambda\xi}^{(\nu)}$ are vibrational states in the potential V and are 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 because of interaction between λ, ξ, and ξ_Z degrees of freedom, higher states in ξ_Z will become excited. As a first study we consider the possible consequences of such excitations by assuming a Boltzmann-like occupation of excited states:

$$|\psi_{\lambda\xi}|^2=\sum_{\nu=0}^{\infty}|\psi_{\lambda\xi}^{(\nu)}(\xi_Z)|^2\exp(-E_{\lambda\xi}^{(\nu)}/\Theta), \tag{7}$$

where Θ is the nuclear temperature, related approximately to the excitation energy.[13] In a more complete treatment of the nuclear-temperature effects one has also to use a cranking formula for the mass parameters generalized for finite temperatures.

The probability for finding a certain charge fragmentation ξ_Z at the position λ and ξ on the fission charge-dispersion path is proportional to $|\psi_{\lambda\xi}(\xi_Z)|^2$. This probability is scaled to a fractional charge yield Y at a charge number Z_1 of one fragment ($d\xi_Z=2/Z$).

$$Y(Z_1)=|\psi_{\lambda\xi}(\xi_Z(Z_1))|^2[B_{\xi_Z\xi_Z}(Z_1)]^{1/2}2/Z \tag{8}$$

and can be compared directly with the experiments.

As a first application of the theory, we have made calculations for the charge distribution in the fission of ^{236}U for the mass fragmentions $|\xi| = 0.195$ and $|\xi| = 0.20$, which for the spontaneous fission refer to the mass chains $A_1 = 141$, $A_2 = 95$ and $A_1 = 142$, $A_2 = 94$, respectively.

Figure 2 gives the potential energy V and the mass parameters B_{ij} for ^{236}U as a function of ξ_Z and Z_1 for $\lambda = 1.8$ and $\xi = \pm 0.195$ minimized in the three-dimensional space of β_1, β_2, and ϵ. In these examples we obtain the interesting result that the potentials show single deep minima at $\xi_Z = \pm 0.195$. Hence Eq. (2) yields $\xi = \xi_Z = \xi_N$ at the minima. It should be remembered that protons and neutrons are considered to be moving in two separate ATCSM potentials, though we find that for a given ξ, the ξ_Z and the corresponding ξ_N result in shapes which are not very different. This is illustrated in Fig. 1 for $\xi_Z = 0.10$ and $\xi_N = 0.256$. These values refer to $\xi = 0.195$, for which the nuclear shape, not plotted in Fig. 1, lies in between these two surfaces. This result suggests that the charge-dispersion effects in fission are only of small orders, which is a known experimental fact.[1] Thus for comparison with experiments only a very small region of the potential and the mass parameters in the neighborhood of the potential minima play a significant role in the calculation of fractional charge yield. This reduces the importance of the otherwise large oscillations in the mass parameters shown in Fig. 2. Further, the coupling mass parameter $B_{\lambda \xi_Z}$ is small and the relation $B_{\lambda \xi_Z}{}^2 \ll B_{\lambda\lambda} B_{\xi_Z \xi_Z}$ holds well such that the coupling term proportional to $B_{\lambda \xi_Z}$ in (5) can be neglected like the terms proportional to $B_{\lambda \xi}$ and $B_{\xi \xi_Z}$. Calculations for $|\xi| = 0.20$ yield the same results.

The calculated fractional charge yield Y is plotted in Fig. 3 for both $|\xi| = 0.195$ and $|\xi| = 0.20$ and for several temperatures. The calculated charge-dispersion curves show Gaussian functional form, independent of the nuclear temperature. Both these results are in agreement with experiment: Empirically, the experimental data

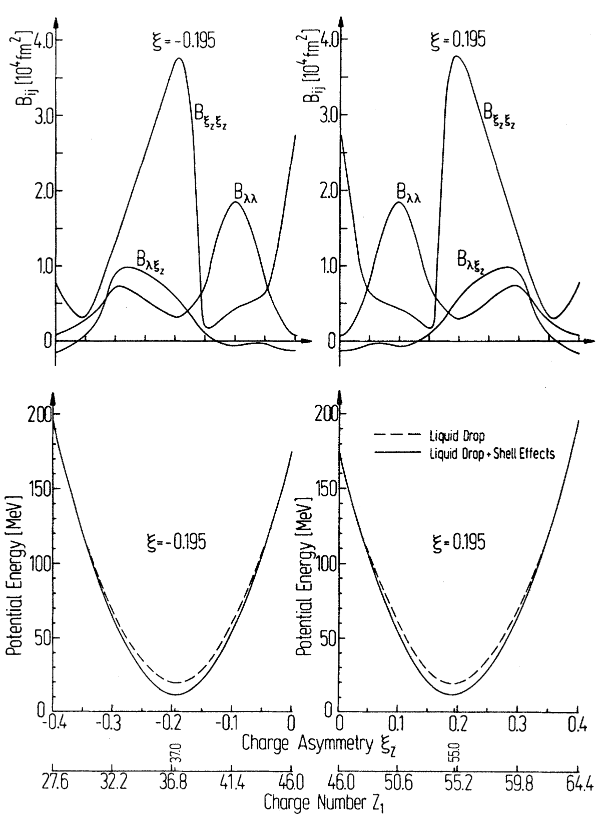

FIG. 2. Charge dispersion potential and the mass parameters in units of nucleon mass, for ^{236}U at elongation $\lambda = 1.8$ and mass asymmetries $\xi = \pm 0.195$.

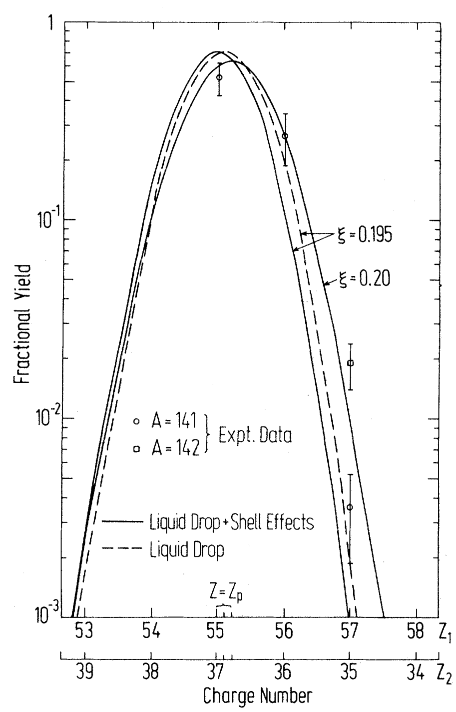

FIG. 3. Theoretical charge dispersion curves for the mass asymmetries $|\xi| = 0.195$ (mass chains 141 and 95) and $|\xi| = 0.20$ (mass chains 142 and 94) in the spontaneous fission of ^{236}U. Nuclear temperatures with $\Theta \leq 7$ MeV give no visible changes in the dispersion curves. The experimental data (Ref. 14) are plotted for the mass chains 141 and 142.

on independent fractional charge yields for a given mass chain are represented by a Gaussian function

$$P(Z) = (c\pi)^{-1/2} \exp[-(Z - Z_p)^2/c] , \quad (9)$$

which is characterized by the most-probable charge Z_p and the width c of the distribution, which is observed to be insensitive to the excitation energy for excitation energies less than about 40 MeV (Ref. 1). The experimental data[14] shown in Fig. 3 are for the mass chains $A_1 = 141$ and 142. For the lighter-mass chains $A_2 = 95$ and 94 the experimental data are not known. The empirical values[14] of Z_p for the mass chains 141 and 142 are, respectively, 54.97 and 55.36 and the width is $c = 0.9 \pm 0.1$ for both the chains. Our theoretical curves are peaked around $Z = 55$ and 55.2, respectively, fixed mainly by the potential, and have widths of the order of the experimental values.

In order to study the question of whether the charge dispersion is influenced by the shell effects (shell plus pairing corrections) or not, we have calculated the dispersion curve with the liquid-drop potential for $\xi = \pm 0.195$ (dashed curves of Fig. 2). The results for this calculation are shown by the dashed curve in Fig. 3, which presents a somewhat improved agreement with experiment. However, we might mention that the experimental data plotted are measured by the thermal-neutron fission of ^{235}U whereas our calculations refer to the spontaneous fission. We have also tested the effect of large oscillations in mass parameters on the charge dispersion and found that it is sensitive to the detailed oscillations of the mass parameters. The width of the peak is increased when $B_{\xi_Z \xi_Z}$ is replaced by a constant $\bar{B}_{\xi_Z \xi_Z}$ value. Hence, for a better quantitative comparison the calculations of particularly the mass parameters have to be carried out to a further accuracy.

In conclusion, firstly we notice that for all the examples of charge dispersion studied here, our calculations support the hypothesis of unchanged charge distribution where $\xi = \xi_Z = \xi_N$. This, however, should not be taken as a general result of our theory. Applications of our theory to other mass chains where the hypothesis of unchanged charge distribution shows deviations between the mass and charge asymmetries are in progress. Secondly, there will certainly be some dependence of our calculated charge dispersion on the time-dependent treatment of the elongation λ.

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