Deformation Energy Minima at Finite Mass Asymmetry

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A very general saddle point nuclear shape may be found as a solution of an integro-differential equation without giving *apriori* any shape parametrization. By introducing phenomenological shell corrections one obtains minima of deformation energy for binary fission of parent nuclei at a finite (non-zero) mass asymmetry. Results are presented for reflection asymmetric saddle point shapes of thorium and uranium even-mass isotopes with $A = 226$-238 and $A = 230$-238 respectively.

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INTRODUCTION

One of the earliest features observed in nuclear fission was the preference for breakup into two fragments of unequal mass [1]. The asymmetric distribution of fragment masses from the spontaneous or low excitation energy induced fission was a longstanding puzzle of the theory [2]. In the framework of the liquid drop model (LDM), the mass distribution of fission fragments is symmetric. By adding shell corrections within Strutinsky's [3] macroscopic-microscopic method, it was shown [4, 5] that the outer barrier for asymmetric shapes is lower than for symmetric ones. In this way it was possible to explain qualitatively the fission asymmetry. Significant progress was achieved with the development of the asymmetric two center shell model by the Frankfurt school [6, 7]. The fragmentation theory was succesful in describing both regions of low and high mass asymmetry.

The shapes during a fission process from one parent nucleus to the final fragments, have been intensively studied either statically (looking for the minimimum of potential energy) [8, 9], or dynamically (by choosing a path with the smallest value of action integral) [10, 11]. Particularly important points on a potential energy surface are those corresponding to the ground state [12], saddle point(s) [8, 13] and scission point [14]. In a static approach, the equilibrium nuclear shapes are usually determined by minimizing the energy functional on a certain class of trial functions representing the surface equation. The required number of independent shape parameters may be as high as nine values [8]; they are discussed in ref. [13].

The purpose of this paper is to present a method allowing to obtain a very general reflection asymmetric saddle point shape as a solution of an integro-differential equation without a shape parametrization *apriori* introduced. This equation was derived by minimizing the potential energy with constraints (constant volume and given deformation parameter). The method [9, 15] allows to obtain straightforwardly the axially symmetric surface shape for which the liquid drop energy, $E_{LDM} = E_s + E_C$, is minimum. By adding the shell correction δE to the LDM deformation energy, $E_{def} = E_{LDM} + \delta E$, we can obtain minima at a finite value of the mass asymmetry parameter. Phenomenological shell corrections are used. Results for binary fission of parent nuclei $^{226-238}$Th and $^{230-238}$U are presented.

DEFORMATION ENERGY

One assumes cylindrical symmetry. The deformation parameter α is defined as the distance between centers of mass of the fragments lying at the left hand side and right hand side of the plane $z = 0$, respectively: $\alpha = |z_L^c| + |z_R^c|$. This definition allows to reach all intermediate stages of deformation from one parent nucleus to two fragments by a continuous variation of its value. The position of separation plane, $z = 0$, is given by the condition: $(d\rho/dz)_{z=0} = 0$.

We are looking for a nuclear surface equation $\rho = \rho(z)$ in cylindrical coordinates, which minimizes the potential energy of deformation with two constraints: volume conservation, and given deformation parameter, α, assumed to be an adiabatic variable.

Relative to a spherical shape, the deformation energy is defined by

$$E_{def}(\alpha) - E^0 = E_s^0[B_s - 1 + 2X(B_C - 1)] + \delta E - \delta E^0 \quad (1)$$

where $E_s^0 = a_s(1 - \kappa I^2)A^{2/3}$ and $E_C^0 = a_C Z^2 A^{-1/3}$ are energies corresponding to spherical shape and $I = (N - Z)/A$. The relative surface and Coulomb energies $B_s = E_s/E_s^0$, $B_C = E_C/E_C^0$ and the shell correction $\delta E(\alpha)$ are functions of the nuclear shape. The dependence on the neutron and proton numbers is contained in E_s^0, the fissility parameter $X = E_C^0/(2E_s^0) = [3Z^2e^2/(5R_0)]/2[a_s(1-\kappa I^2)A^{2/3}]$, as well as in shell correction energy δE^0 of the spherical nucleus. From a fit to experimental data on nuclear masses, quadrupole moments, and fission barriers, the following values of the parameters have been obtained [16]: $a_s = 17.9439$ MeV, $\kappa = 1.7826$, $a_C = 3e^2/(5r_0) = 0.7053$ MeV. The radius of spherical nucleus is $R_0 = r_0 A^{1/3}$ with $r_0 = 1.2249$ fm, and $e^2 = 1.44$ MeV·fm is the square of electron charge. The shape-dependent dimensionless surface term is proportional to the surface area, and the expression of Coulomb energy is a double integral. Both are computed [17] by Gauss-Legendre numerical quadratures.

To the LDM energy we add a *phenomenological shell correction* $\delta E(\alpha)$ by using a formula adapted after Reference [16]. At a given deformation we calculate the volumes of fragments and the corresponding numbers of nucleons $Z_i(\alpha)$, $N_i(\alpha)$ $(i = 1, 2)$, proportional to the volume of each fragment. Then we add for each fragment the contribution of protons and neutrons

$$\delta E(\alpha) = \sum_i \delta E_i(\alpha) = \sum_i [\delta E_{pi}(\alpha) + \delta E_{ni}(\alpha)] \qquad (2)$$

given by

$$\delta E_{pi} = Cs(Z_i); \quad \delta E_{ni} = Cs(N_i) \qquad (3)$$

where

$$s(Z) = Z^{-2/3} F(Z) - cZ^{1/3} \qquad (4)$$

and a similar equation for $s(N)$.

$$F(n) = \frac{3}{5}\left[\frac{N_i^{5/3} - N_{i-1}^{5/3}}{N_i - N_{i-1}}(n - N_{i-1}) - n^{5/3} + N_{i-1}^{5/3}\right] \qquad (5)$$

where $n \in (N_{i-1}, N_i)$ is the current number of protons (Z) or neutrons (N) and N_{i-1}, N_i are the nearest magic numbers. The parameters $c = 0.2$, $C = 6.2$ MeV were determined by fit to experimental masses and deformations. The dependence on deformation α [18] is given by

$$\delta E(\alpha) = \frac{C}{2}\left\{\sum_i [s(N_i) + s(Z_i)]\frac{L_i(\alpha)}{R_i}\right\} \qquad (6)$$

where $L_i(\alpha)$ are the lenghths of fragments along the symmetry axis. During the deformation process, the variation of separation distance between centers induces the variation of the geometrical quantities and of the corresponding nucleon numbers. Each time a proton or neutron number reaches a magic value, the correction energy passes through a minimum, and it has a maximum at midshell.

INTEGRO-DIFFERENTIAL EQUATION

As mentioned above, the nuclear surface equation with axial symmetry around z axis is expressed as $\rho = \rho(z)$ in cylindrical coordinates. We use the following relationships for the principal radii of curvature $\mathcal{R}_1 = \tau\rho$, ${\mathcal{R}_2}^{-1} = -\rho''/\tau^3$, in which $\tau^2 = 1 + \rho'^2$. In order to minimize the deformation energy the surface equation should be a solution of the following equation

$$\rho\rho'' - \rho'^2 - [\lambda_1 + \lambda_2|z| + 10XV_s(z,\rho)]\,\rho(1+\rho'^2)^{3/2} - 1 = 0 \qquad (7)$$

where $\rho' = d\rho/dz$, $\rho'' = d^2\rho/dz^2$, and V_s is the Coulomb potential on the nuclear surface. In this equation λ_1 and λ_2 are Lagrange multipliers corresponding to the constraints of volume conservation (or given mass asymmetry if the voulume is conserved in each "half" of the nucleus) and a determined value of the deformation parameter α. All lengths are given in units of R_0, Coulomb potential in units of Ze/R_0, and energy in units of the surface energy E_s^0. One can calculate for every value of α the deformation energy $E_{def}(\alpha)$. The particular value α_s for which $dE_{def}(\alpha_s)/d\alpha = 0$ corresponds to the extremum, i.e. the shape function describes the saddle point (or the ground state). The associated surface equation gives the unconditional extremum of the energy and corresponds to the fission barrier. The other surfaces (for $\alpha \neq \alpha_s$) are extrema only with condition $\alpha =$ constant.

The Coulomb potential on the surface depends on the function $\rho(z)$, hence eq 7 is an integro-differential one, as V_s is expressed by an integral on the nuclear volume. The integration method used to solve eq 7 is based on the weak dependence of Coulomb energy on the nuclear shape. It is invariant under subtraction from V_s of a linear function because λ_1 and λ_2 are arbitrary constants. The extremal surface depends on the quantity with which the Coulomb potential on the nuclear surface differs from the function $\lambda_1 + \lambda_2|z|$, where the constants λ_1, λ_2 could be chosen in a way to minimize this difference. In the next iterration one uses the solution $\rho(z)$ previously determined.

The following boundary conditions have to be fulfilled

$$\rho(z_1) = \rho(z_2) = 0 \qquad (8)$$

$$\lim_{z\to z_1} d\rho(z)/dz = \infty \ ; \ \lim_{z\to z_2} d\rho(z)/dz = -\infty \qquad (9)$$

where z_1 and z_2 are the intercepts with z axis at the two tips. Equations 9 are called transversality conditions. For *reflection symmetric shapes* $z_1 = -z_2 = -z_p$, hence one can consider only positive values of z in the range $(0, z_p)$. In order to get rid of singularities in eq 9 it is convenient to introduce a new function $u(v)$ instead of $\rho(z)$

$$u(v) = \mathcal{A}^2\rho^2(z(v)) \qquad (10)$$

where

$$z(v) = z_p - v/\mathcal{A} \qquad (11)$$

By substituting into eq 7 one has

$$u'' - 2 - \frac{1}{u}\left[u'^2 + \left(\frac{5XV_s}{2\mathcal{A}} + \frac{\lambda_1+\lambda_2 z_p}{4\mathcal{A}} - \frac{\lambda_2 v}{4\mathcal{A}^2}\right)\cdot (4u + u'^2)^{3/2}\right] = 0 \qquad (12)$$

Then we introduce a linear function of v by adding and subtracting $a+bv$ to $5XV_s/(2\mathcal{A})$ and defining V_{sd} as deviation of Coulomb potential at the nuclear surface from a linear function of v

$$V_{sd} = [5X/(2\mathcal{A})]V_s - a - vb \tag{13}$$

The linear term may be considered an external potential of deformation with $a = [5X/(2\mathcal{A})]V_s(v = 0)$ and $b = \{[5X/(2\mathcal{A})]V_s(v = v_p) - a\}/v_p$ leading to

$$u'' - 2 - \frac{1}{u}\left\{u'^2 + \left[\left(\frac{\lambda_1+\lambda_2 z_p}{4\mathcal{A}} + a\right) + v\left(b - \frac{\lambda_2}{4\mathcal{A}^2}\right) + V_{sd}\right](4u+u'^2)^{3/2}\right\} = 0 \tag{14}$$

Here we have new constants $\mathcal{A}$ and z_p related to eq 10, besides the previous ones λ_1 and λ_2. Nevertheless the solution is not dependent on each parameter; important are the linear coefficients in v of the binomial term within parantheses. By equating with 1 the coefficient of v, one can establish the following link between parameter $\mathcal{A}$ and the Lagrange multiplier λ_2

$$\mathcal{A}^2 = \lambda_2/4(b-1) \tag{15}$$

In this way $u(v)$ should be determined by equation

$$u'' = 2 + \frac{1}{u}[u'^2 + (v - d + V_{sd})(4u + u'^2)^{3/2}] \tag{16}$$

containing a single parameter d. At the limit

$$u(0) = 0, \;\; u'(0) = 1/d \tag{17}$$

and eq 9 is satisfied if $z_p = v_p/\mathcal{A}$ is obtained from

$$u'(v_{pn}) = 0 \tag{18}$$

The subscript n was introduced as a consequence of the fact that the number of points v_{pn} (depending on d and other parameters), satisfying eq 18 is larger than unity. In order to solve eq 16 one starts with given values of parameters d and n. Different classes of shapes solutions of eq. 16 are obtained by taking various values of n: for $n = 2$ there is one neck (binary fission), $n = 3$ gives two necks (ternary fission), etc. For reflection symmetric shapes $d_L = d_R$ and $n_L = n_R$. Although the parameter $\mathcal{A}$ is not present in this equation we have to know it in order to determine the shape function from eq 10. From the volume conservation one has

$$\mathcal{A} = \left\{\frac{3}{2}\int_0^{v_{pn}} u(v)dv\right\}^{1/3} \tag{19}$$

After solving the integro-differential equation one can find the deformation parameter $\alpha = z_L^c + z_R^c$, where

$$z_L^c = \int_{z_1}^{0} |z|\rho^2(z)dz \Big/ \int_{z_1}^{z_2} \rho^2(z)dz \tag{20}$$

$$z_L^c = \frac{3}{2}\mathcal{A}^{-4}\int_0^{v_p}(v_p - v)u(v)dv \tag{21}$$

depends on d. From $\alpha(d)$, one can obtain the inverse function $d = d(\alpha)$.

For *reflection asymmetrical shapes* we need to introduce another constraint: the asymmetry parameter, η, defined by

$$\eta = \frac{M_L - M_R}{M_L + M_R} = \frac{A_1 - A_2}{A_1 + A_2} \tag{22}$$

should remain constant during variation of the shape function $\rho(z)$. Consequently eq 16 should be written differently for left hand side and right hand side. Now d_L is different from d_R, and $\mathcal{A}_L \neq \mathcal{A}_R$. They have to fulfil matching conditions $\rho_L(z=0) = \rho_R(z=0)$ hence

$$u_L^{1/2}(v_p)/\mathcal{A}_L = u_R^{1/2}(v_p)/\mathcal{A}_R \tag{23}$$

The second derivative $\rho''(z)$ can have a discontinuity in $z = 0$ if $d_L \neq d_R$. The parameters $\mathcal{A}_L$ and $\mathcal{A}_R$ are expressed in terms of η, if we write eq 22 as

$$M_L = \frac{2\pi}{3}(1+\eta) = \pi\mathcal{A}_L^{-3}\int_0^{v_p} u_L(v)dv \tag{24}$$

$$M_R = \frac{2\pi}{3}(1-\eta) = \pi\mathcal{A}_R^{-3}\int_0^{v_p} u_R(v)dv \tag{25}$$

We assume that $M_L + M_R$ is equal to the mass of a sphere with $R = 1$. From eqs 24, 25 we obtain

$$\mathcal{A}_L = (1+\eta)^{-1/3}\mathcal{A}_{L0} \tag{26}$$

$$\mathcal{A}_R = (1-\eta)^{-1/3}\mathcal{A}_{R0} \tag{27}$$

where we introduced notations similar to eq 19

$$\mathcal{A}_{L0(R0)} = \left\{\frac{3}{2}\int_0^{v_p} u_{L(R)}(v)dv\right\}^{1/3} \tag{28}$$

The equation 16 is solved by successive approximations. In every iteration one uses the 2nd order Runge-Kutta numerical method with constant integration step. The initial value $u''(v=0)$ can be found straightforwardly by removing the indetermination in the point $v = 0$, $u''(0) = -2 + (1 - b + g)/2d^2$, where $g = [5X/(2\mathcal{A})][dV_s(v)/dv]_{v=0}$. The equation is integrated up to the point $v = v_{pn}$, in which the first derivative $u'(v_{pn})$ vanishes.

MASS ASYMMETRY

The variations of the saddle point energy with the mass asymmetry parameter $d_L - d_R$ (which is almost linear function of the mass asymmetry η) for some even-mass isotopes of Th and U are plotted in figures 1 and 2. The minima of the saddle-point energy occur at nonzero mass asymmetry parameters $d_L - d_R$ between about 0.04 and 0.085 for these nuclei. When the mass number of an isotope increases, the value of the mass asymmetry corresponding to the minimum of the SP energy decreases.

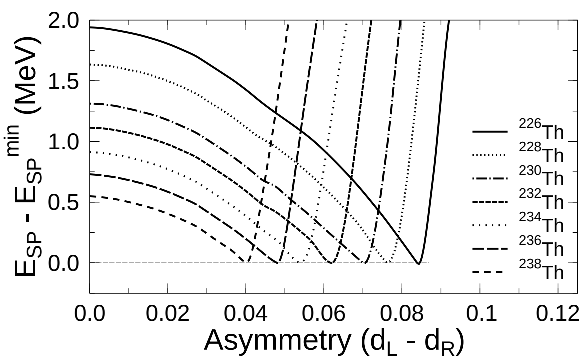

FIG. 1: Saddle-point deformation energy versus mass asymmetry parameter for the binary fission of some even-mass Th isotopes in the presence of shell corrections.

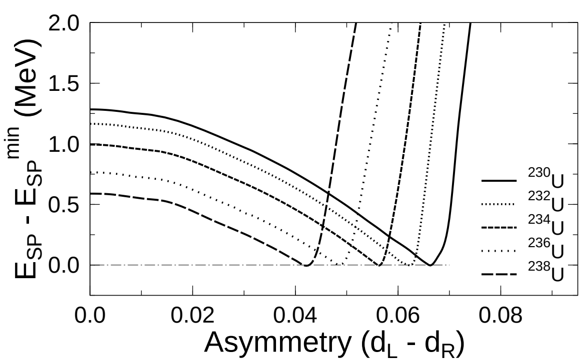

FIG. 2: Saddle-point deformation energy versus mass asymmetry parameter for the binary fission of some even-mass U isotopes in the presence of shell corrections.

From the saddle point energies E_{SP} of every nucleus we subtract the minimum E_{SP}^{min}, as in the figures 1 and 2. The equilibrium conditions are changed when the shell effects are taken into account. The minimum of the E_{SP} is produced by the negative values of the shell corrections $\delta E - \delta E^0$.

In conclusion, the liquid drop model saddle point shapes and energy barrier heights are well reproduced by the present method. By adding shell corrections to the LDM energy we succeded to obtain the minima shown in Figures 1 and 2 at a finite value of mass asymmetry for the binary fission of $^{226-238}$Th and $^{230-238}$U nuclei.

As mentioned by Wilkins et al. [19], calculations of PES for fissioning nuclei “qualitatively account for an asymmetric division of mass”. From the qualitative point of view the results displayed in Figures 1 and 2 proove the capability of the method to deal with fission mass and charge asymmetry.

The experimentally determined mass number of the most probable heavy fragment [20] for U isotopes ranges from 134 to 140. The corresponding values at the displayed minima in Figures 1 and 2 are very close to 125, which means a discrepancy between 6.7 % and 10.7 % for A_H. The inaccuracy in reproducing the experimental mass asymmetry is due to the contribution of the phenomenological shell corrections. In the absence of shell corrections the pure liquid drop model (LDM) reflection-symmetric saddle point shapes [8] are reproduced, and the barrier height increseas with an increased mass asymmetry. When the shell corrections are taken into account the LDM part behaves in the same manner (larger values at non-zero mass asymmetry). Only the contribution of shell effects can produce a minimum of the barrier height at a finite value of the mass asymmetry. One may hope to obtain a better agreement with experimental data by using a more realistic shell correction model.

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