# Asymmetric fission of $^{180}$Hg and the role of hexadecapole moment

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## Abstract

In current work, the fission property of $^{180}$Hg is investigated based on the Skyrme density functional theory (DFT). The impact of the high-order hexadecapole moment ($q_{40}$) is found at large deformations. With the $q_{40}$ constraint, a smooth and continuous potential energy surfaces (PES) could be obtained. Especially, the hexadecapole moment constraint is essential to get proper scission configurations. The static fission path based on the PES supports the asymmetric fission of $^{180}$Hg. The asymmetric distribution of the fission yields of $^{180}$Hg is further reproduced by the time-dependent generator coordinate method (TDGCM), and agrees well with the experimental data.

## Full Text

## Preamble

### Asymmetric Fission of 180Hg and the Role of Hexadecapole Moment

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In this work, the fission properties of 180Hg are investigated using Skyrme density functional theory (DFT). The impact of the high-order hexadecapole moment (q40) is found to be significant at large deformations. With the q40 constraint, a smooth and continuous potential energy surface (PES) can be obtained. In particular, the hexadecapole moment constraint is essential for obtaining proper scission configurations. The static fission path based on the PES supports the asymmetric fission of 180Hg. The asymmetric distribution of fission yields for 180Hg is further reproduced by the time-dependent generator coordinate method (TDGCM), showing good agreement with experimental data.

**Keywords:** Nuclear fission, density functional theory, hexadecapole moment, potential energy surface, mass distribution

## INTRODUCTION

The asymmetric fission mode in neutron-deficient 180Hg was discovered in 2010 through the $\beta$ decay of 180Tl [1]. For the fission of 180Hg, its splitting into two 90Zr fragments with magic N = 50 and semimagic Z = 40 was initially believed to dominate the fission process. However, contrary to early theoretical predictions, 180Hg has been observed to fission asymmetrically, with heavy and light fragment mass distributions centered around A = 100 and 80 nucleons, respectively [1, 2].

This puzzling fission behavior of 180Hg has attracted considerable theoretical attention. For example, macroscopic-microscopic models [1, 3–5] and self-consistent microscopic approaches [6–8] have been used to analyze multidimensional potential energy surfaces (PESs), with the presence of an asymmetric saddle point and a rather high ridge between symmetric and asymmetric fission valleys explained as the main factor determining the mass split in fission.

Fission-fragment yields for 180Hg have also been calculated using various methods including the Brownian Metropolis shape-motion treatment [3, 5, 9], Langevin equation [10], scission-point model [4, 11, 12], and random neck rupture mechanism [13], based on either PESs or scission configurations. These results are in approximate agreement with experimental data, showing a deviation of ~4 nucleons for the peak positions. Several attempts have also been made to describe the fragment mass distribution of 180Hg in a fully microscopic way, namely through the time-dependent generator coordinate method (TDGCM) based on covariant density functional theory (CDFT) [8], which reproduces the asymmetric peaks very well while predicting a more asymmetric fission mode with A_H ~ 116 that was not observed experimentally.

In theoretical studies of nuclear fission, the PES is a crucial infrastructure that describes the evolution of nuclear energies with shape variations along the path from the initial configuration toward scission. In nuclear physics, there are generally two approaches to generate PES. One ranges from the historical liquid drop model [14–18] to the well-known macroscopic-microscopic model, using parametrization of the nuclear mean-field deformation [19–28]. The other is based on microscopic self-consistent methods [29–37] or the constrained relativistic mean-field method [8, 38–44].

In the macroscopic-microscopic method, a predefined class of nuclear shapes is defined uniquely in terms of selecting appropriate collective coordinates, and relatively smooth potential energy surfaces can be obtained. However, due to limitations in computing resources, microscopic calculations of PES can only be performed within a limited number of deformation degrees of freedom. In microscopic self-consistent methods, higher-order collective degrees of freedom are incorporated self-consistently based on the variational principle. In fission studies, quadrupole and octupole deformation (moments) constraints are the natural and most often used parameters for calculating microscopic PES.

However, several studies have shown that as a consequence of the absence of hexadecapole deformation (q40 or $\beta_4$), PES may exhibit discontinuities in the large deformation scission regions [45–49]. Ref. [50] investigated the role of hexadecapole deformation on the PES calculation of 240Pu by applying a perturbation on $\beta_4$. The results show that one can obtain a smooth 2-dimensional PES in ($\beta_2$, $\beta_3$) by parallel calculations with a suitable perturbation of hexadecapole deformation.

But for asymmetric fission of 180Hg, there have been no reports about the effect of q40 or $\beta_4$ on the PES of 180Hg. The self-consistent calculation in quadrupole and octupole deformation spaces indicated that the PES of 180Hg exhibits different behavior from that of 240Pu or 236U with increasing quadrupole moment [6, 8]. Thus it is interesting to examine the influence of hexadecapole moment on the PES of 180Hg at large deformation and also analyze some properties of scission configuration. In this paper, we extend two-dimensional (q20, q30) constraint calculations at large deformation region by adding q40 constraint on the microscopic PES calculation of 180Hg. The importance of q40 in the self-consistent calculation of PES for 180Hg at large deformation will be investigated. Moreover, the fission dynamics of 180Hg, the total kinetic energies, and the fragment mass yield distributions based on TDGCM [51] will be described and discussed.

## II. THEORETICAL FRAMEWORK

To study static fission properties, the PES was determined using Skyrme density functional theory (DFT). The dynamic process is further investigated within the framework of TDGCM. In this section, we briefly explain these two methods. Detailed descriptions of Skyrme DFT can be found in Ref. [52], and formulations of TDGCM can be found in Refs. [51, 53–55].

### A. Density Functional Theory

In the local density approximation of DFT, the total energy of finite nuclei can be calculated from the spatial integration of the Hamiltonian density H(r),

$$H(r) = \mathcal{T}(r) + \sum_{t=0,1} \mathcal{V}_t(r) + \tilde{\mathcal{V}}_t(r)$$

In the above equation, $\mathcal{T}(r)$, $\mathcal{V}_t(r)$, and $\tilde{\mathcal{V}}_t(r)$ stand for the kinetic energy density, potential energy density, and pairing energy density, respectively. The symbol $t = 0, 1$ denotes isoscalar or isovector, respectively [56].

The mean-field potential energy $\mathcal{V}_t(r)$ in Skyrme DFT has the general form:

$$\mathcal{V}_t(r) = C_t^{\rho\rho}\rho_t^2 + C_t^{\rho\Delta\rho}\rho_t\Delta\rho_t + C_t^{\rho\tau}\rho_t\tau_t + C_t^{J^2}J_t^2 + C_t^{\rho\nabla J}\rho_t\nabla\cdot J_t + C_t^{\rho\rho t0} + C_t^{\rho\tau t0} = C_t^{\rho\rho}\rho_t^{\gamma}$$

where the particle density $\rho_t$, kinetic density $\tau_t$, and spin current vector densities $J_t$ $(t = 0; 1)$ can be calculated from the density matrix $\rho_t(\mathbf{r}\sigma; \mathbf{r}'\sigma')$, with dependence on spatial $(\mathbf{r})$ and spin $(\sigma)$ coordinates. $C_t^{\rho\rho}$, etc., are coupling constants for different types of densities in the Hamiltonian density $H(r)$, which are usually real numbers. As an exception, $C_t^{\rho\rho}$ is the density-dependent term. The formulations relating the coupling constants to traditional Skyrme parameters can be found in Ref. [57]. For example, the spin-orbit force of the Skyrme interaction corresponds to the term $C_t^{\rho\nabla J}\rho_t\nabla\cdot J_t$.

Pairing correlations are often taken into account through the Hartree-Fock-Bogoliubov (HFB) approximation in DFT [52]. In the case of the Skyrme energy density functional, a commonly adopted pairing force is the density-dependent surface-volume, zero-range potential, as given in Refs. [35, 58]:

$$\hat{V}_{\text{pair}}(\mathbf{r}; \mathbf{r}') = V_0^{(n,p)}\left[1 - \frac{\rho(\mathbf{r})}{\rho_0}\right]\delta(\mathbf{r} - \mathbf{r}')$$

where $V_0^{(n,p)}$ is the pairing strength for neutrons $(n)$ and protons $(p)$, $\rho_0$ is the saturation density of nuclear matter fixed as 0.16 $\text{fm}^{-3}$, and $\rho(\mathbf{r})$ indicates the total density. As studied in Ref. [35], this type of pairing force is a suitable choice for nuclear fission studies.

The DFT solver HFBTHO(v3.00) [59] is used to generate the PESs, assuming axial symmetry. 26 major shells of the axial harmonic-oscillator single-particle basis are used, and the number of basis states is further truncated to 1140. In this work, the Skyrme DFT with SkM* parameters [60] is adopted, which is commonly used for fission studies. For the pairing strength, $v_0^{(n)} = -268.9$ $\text{MeV}\cdot\text{fm}^3$ and $v_0^{(p)} = -332.5$ $\text{MeV}\cdot\text{fm}^3$ are used for neutrons and protons, respectively, with a pairing window of $E_{\text{cut}} = 60$ MeV. This pairing strength together with the choice of SkM* force and model space has been adopted in Refs. [6, 61], in which the two-dimensional PES related to the fission of 180Hg has been studied.

### B. Time-Dependent Generator Coordinate Method

Nuclear fission is a large-amplitude collective motion that can be approximated as a slow adiabatic process driven by several collective degrees of freedom. In TDGCM, the many-body wave function of the fissioning system takes the generic form:

$$|\Psi(t)\rangle = \int f(q;t)|\Phi(q)\rangle dq$$

where $|\Phi(q)\rangle$ is composed of known many-body wave functions with the vector of continuous variables $q$. The $q$ are collections of variables chosen according to the physics problem. For fission studies, two collective variables—quadrupole moment $\hat{q}_{20}$ and octupole moment $\hat{q}_{30}$—are usually adopted.

In the above equation, $f(q;t)$ is a weight function determined by the time-dependent Schrödinger-like equation:

$$i\hbar \frac{\partial g(q;t)}{\partial t} = \hat{H}_{\text{coll}}(q) g(q;t)$$

in which the Gaussian overlap approximation (GOA) is used. $\hat{H}_{\text{coll}}(q)$ is the collective Hamiltonian:

$$\hat{H}_{\text{coll}}(q) = -\frac{\hbar^2}{2} \sum_{ij} \frac{\partial}{\partial q_i} B_{ij}(q) \frac{\partial}{\partial q_j} + V(q)$$

in which $V(q)$ is the collective potential, and $B_{ij}(q) = M_{ij}^{-1}(q)$ is the inertia tensor as the inverse of the mass tensor $M$. The potential and mass tensor are solved by Skyrme DFT in this work. $g(q;t)$ contains information about the dynamics of the fissioning nuclei and is the complex collective wave function with collective variables $q$.

To describe nuclear fission, the collective space has been divided into an inner region and an external region, corresponding to the nucleus staying as a whole and the nucleus separated into two fragments, respectively. The scission contour, which is a hyper-surface, separates these two regions. The flux of the probability current passing the scission contour can be used to evaluate the probability of observing two fission fragments at time $t$. For a surface element $S$ on the scission contour, the integrated flux $F(S;t)$ is calculated by:

$$F(S;t) = \int_S \mathbf{J}(q;t) \cdot d\mathbf{S}$$

as in Ref. [51], where $\mathbf{J}(q;t)$ is the current:

$$\mathbf{J}(q;t) = \mathbf{B}(q) \left[ g^*(q;t) \nabla g(q;t) - g(q;t) \nabla g^*(q;t) \right]$$

The yield of the fission product with mass number $A$ can be obtained by:

$$Y(A) = C \int_{S_A} F(S;t)$$

where $S_A$ denotes an ensemble of all surface elements on the scission contour containing the fragment with mass number $A$, and $C$ is a normalization factor ensuring the total yield is normalized to 200. In the same way, the yield of fission fragments with charge number $Z$ can also be obtained.

In this work, the computer code FELIX (version 2.0) [55] is used for describing the time evolution of nuclear fission in the TDGCM-GOA framework.

## III. RESULTS AND DISCUSSION

In the adiabatic approximation approach for fission dynamics, a precise multidimensional PES is the first and essential step toward a dynamical description of fission. Figure 1 displays the PES contour of 180Hg obtained by HFB calculation in the collective space of (q20, q30), where q20 ranges from -20 b to 300 b and q30 ranges from 0 b^(3/2) to 40 b^(3/2) with steps of Δq20 = 2 b and Δq30 = 2 b^(3/2). Overall, the pattern of PES obtained in this work based on the DFT solver HFBTHO with Skyrme SkM* functional is similar to that obtained using the symmetry-unrestricted DFT solver HFOOD [6] with the same functional and that obtained using covariant density functional theory with the relativistic PC-PK1 functional [8]. The static fission path starts from a nearly spherical ground state (q20 = 20 b, q30 = 0 b^(3/2)). A reflection-symmetric fission path can be found for small quadrupole deformations, and the reflection-asymmetric path branches away from the symmetric path at about q20 = 100 b. Unlike the PES of actinide nuclei, there is no valley toward scission for 180Hg; instead, it undergoes a continuous uphill process until reaching the mass-asymmetric scission point with high q30 asymmetry.

In (q20, q30)-constrained PES calculations by DFT, other deformation degrees of freedom are obtained based on the variational principle. In Refs. [49, 50], it has been shown that at given q20 and q30, there are two minima with different values of q40, and the minimum with larger q40 disappears when q20 is large enough, indicating the transition toward scission. The hexadecapole deformation is an important degree of freedom for describing PES at large deformation, and especially, a perturbation of hexadecapole deformation is required for a smooth and reasonable PES, as shown in Ref. [50]. Thus, in our work, at large quadrupole moments (i.e., larger than q20 200 b ($\beta_2$ 3.2)), a further constraint of hexadecapole moment q40 is introduced in a "perturbative" way. A smaller hexadecapole moment than the one obtained variationally is used as a further constraint in the first ten steps of DFT iterations, and it is then released to vary freely. Thus, a lower energy minimum with smaller q40 might be obtained. This treatment is used for the calculation of PES in Fig. 1 and is labeled "(B)" in Figs. 2–3. The calculation with only q20 and q30 constraints is labeled "(A)" in Figs. 2–3.

Figure 2 shows the energies of the static fission path as a function of quadrupole moment q20. The symmetric (q30 = 0 b^(3/2)) and asymmetric fission paths in 180Hg are given respectively. These energies increase steadily with q20. At around q20 100 b, the asymmetric fission path starts to be favored energetically compared to the symmetric path. The transitional valley that bridges the asymmetric and symmetric paths is also drawn in red in Fig. 2. Notably, this connection occurs at the deformation where the symmetric and asymmetric paths are nearly equivalent in energy. This characteristic of 180Hg has also been verified in Ref. [62] using the HFB-Gogny D1S interaction.

From Fig. 2, for case (A), one can see that the energy of 180Hg increases continuously with q20, and it is difficult to rupture even at very large elongation (e.g., q20 $\geq$ 300 b ($\beta_{20} \geq 4.8$)). As seen in case (B), with the inclusion of the q40 constraint, a gentle descent trend of the energy happens at q20 240 b, and a sudden drop in energy occurs at q20 280 b, indicating nuclear scission.

In Fig. 3, the hexadecapole moment (q40), the average particle number around the neck (qN), and the HFB energies are given as functions of q20 at given q30. To investigate the role of q40, only the region with large q20 is shown. From Fig. 3(a), one can see that q40 increases nearly linearly until very large q20 values, especially for case (A), in which q40 can become very large during elongation. After a "perturbative" constraint on q40, as in case (B), the q40 value has a sudden drop and then grows linearly. In nuclear fission studies, qN is often adopted as an indicator of nuclear scission. For example, qN = 4 has been used for determining the scission line of 240Pu in Refs. [35, 48, 63]. In Fig. 3(b), qN decreases gradually against q20. However, for case (A), the reduction of qN becomes rather slow with increasing q20. Especially at q20 340 b ($\beta_2$ 5.4), qN > 4 for q30 = 0 and 10 b^(3/2), and the total energies increase continuously at large q20, as seen in Fig. 3(c). After considering the q40 constraint, as shown in case (B) of Fig. 3(b) and (c), both qN and the total energy have a sudden drop at around q20 280 b ($\beta_2$ 4.5), indicating nuclear rupture. When qN $\leq$ 4, qN becomes close to zero with increasing q20.

To investigate the role of q40 on PES, the HFB energies and qN against q40 at given q20 and q30 are plotted in Fig. 4, obtained through exact constrained calculations of q20, q30, and q40. In the figure, q30 is constrained to 0 b^(3/2). Other q30 values were also tested, and the results are similar to those in Fig. 4. In Fig. 4(a), one can see that there are two local minima along the q40 degree of freedom, corresponding to distinct valleys on the multidimensional potential energy surface. In Ref. [50], a similar trend in 240Pu was found, and the minimum related to larger q40 disappears with increasing quadrupole deformation (at roughly $\beta_2$ 3.8), leading to a natural transition to the minimum with smaller q40. This transition causes the discontinuity and sudden energy drop in the two-dimensional PES of (q20, q30). However, for 180Hg, an extremely soft and relatively flat minimum with larger q40 value remains even at very large q20 values (e.g., at q20 = 340 b ($\beta_2$ 5.4)). In PES calculations with only (q20, q30) constraint, the q40 degree of freedom is obtained through variation of total energies. As seen for case (A) in Fig. 3, q40 after the variational calculation grows steadily even at very large q20, and no transition to the minimum with smaller q40 occurs. With only (q20, q30) constraint, it is difficult to find proper scission configuration, at least for 180Hg. After the "perturbative" inclusion of q40 constraint, as in case (B), such transition can occur at large q20. In Fig. 4(b), it is seen that qN increases with q40 in general. qN around the minimum with larger q40 is roughly larger than 4, and when q20 > 200 b its value around the minimum with smaller q40 is close to zero (numerically $10^{-3}$–$10^{-4}$, effectively near zero). For qN  0, the nucleus becomes well separated into two fragments. From this calculation, one can learn that introducing q40 constraint in the self-consistent calculation of PES can ensure continuity of the potential energy surface. q40 is essential in DFT calculations for fission studies, especially for the transition to scission.

Several results of (q20, q30, q40) constrained calculations are shown in Fig. 5 for the density distribution profiles of 180Hg. q20 is constrained to 240 b, and q40 changes from 140 $b^2$ to 60 $b^2$ for q30 = 0 b^(3/2) and q30 = 10 b^(3/2) in the upper and lower panels, respectively. The q40 degree of freedom influences the formation of the neck and scission configurations. From the figure, one can see that with large q40, there is no neck in the nucleus and the nucleus is simply stretched very long. For calculations with only (q20, q30) constraint as in case (A) of Figs. 2–3, q40 has a very large value with increasing q20 and thus the nucleus cannot undergo scission. With decreasing q40, the neck structure of the nucleus appears and becomes well separated when q40 has small values.

One of the most important quantities in induced fission is the total kinetic energy (TKE) carried by the fission fragments. In this work, the total kinetic energy of the two separated fragments at the scission point can be approximately estimated as the Coulomb repulsive interaction using the simple formula $e^2 Z_H Z_L / d_{\mathrm{ch}}$, where $e$ is the proton charge, $Z_H$ and $Z_L$ denote the charge numbers of the heavy and light fragments, respectively, and $d_{\mathrm{ch}}$ is the distance between the centers of charge of the two fragments at the scission point.

Figure 6 displays the distribution of calculated Coulomb repulsive energy based on the scission line shown by the purple circles in Fig. 1, compared with measured TKE [2]. The calculated results reproduce the trend of the measured TKE quite well, especially the dip at $A_H = 90$ and peak at $A_H = 94$, although the calculated results are generally overestimated by several MeV compared to the data, which might be caused by neglecting dissipation effects.

Finally, we performed TDGCM+GOA calculations to model the time evolution of the fission dynamics of 180Hg. Figure 7 shows the calculated mass distributions of the fission fragments of 180Hg, compared with experimental data [1, 2]. The theoretical result in the framework of covariant density functional theory (CDFT) using PES generated with the neck coordinate constraint qN from Ref. [8] is also given for comparison, denoted as "CDFT". As one of the most important microscopic inputs for fission dynamic calculations, the mass tensor is calculated by GCM or ATDHFB methods in the present work. The calculated mass distribution is generally similar using mass tensors from these two methods, with better agreement obtained using the GCM method for the height of asymmetric peaks and symmetric valley. Overall, the calculation reproduces experimental data well. The calculated peak position deviates by one unit from the experimental peak position. CDFT results show good asymmetric peak positions but overestimate the data and even predict a small peak for the symmetric valley. Moreover, a more asymmetric fission mode with $A_H \sim 116$–117 is predicted both in this work and in CDFT calculation. As explained in Ref. [8], this mode resulted from using an initial state with mixed angular momenta, while in the experiment only certain values occur due to selection rules of electron capture of 180Tl [1, 2]. In the current work, this discrepancy occurs because an initial state with mixed angular momenta is also applied.

## IV. SUMMARY

In this work, the static fission properties and fission dynamics of 180Hg were investigated using Skyrme DFT and TDGCM, respectively. During the calculation of multidimensional PES, it was found that the hexadecapole moment is crucial for obtaining a smooth PES and proper scission configurations, and thus it is essential for fission dynamic studies. For PES calculations with only q20 and q30 constraints, nuclear rupture does not occur even at very large q20. Through calculations with q20, q30, and q40 constraints, it was found that a rather soft and flat minimum with large hexadecapole moment still exists in the PES of 180Hg even with very elongated shapes, which hinders transition to the lower energy minimum with smaller q40. With the strategy of "perturbative" constraint of the collective freedom q40, transition to the minimum corresponding to nuclear rupture could occur naturally, and thus reasonable scission configurations can be obtained. From these scission configurations, the estimated distribution of TKE reproduces the trend of experimental data.

Based on static PES calculations, it was learned that the asymmetric fission channel is favored in 180Hg. Finally, fission fragment yields were calculated with TDGCM. The calculated mass distributions also support asymmetric fission for 180Hg, agreeing well with experimental data. Moreover, a more asymmetric peak with $A_H \sim 117$ is predicted, which was also predicted by covariant DFT with the PC-PK1 parameter set [8].

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*Note: Figure translations are in progress. See original paper for figures.*