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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. Yang Su,[1] Yong-Jing Chen,[1] Ze-Yu Li,[1] Li-Le Liu,[1] Guo-xiang Dong,[2] and Xiaobao Wang[2], † [1]China Nuclear Data Center, China Institute of Atomic Energy, Beijing 102413, China [2]School of Science, Huzhou University, Huzhou 313000, China

In the current work, the fission property of 180Hg is investigated based on the Skyrme density functional theory (DFT). The impact of the high-order hexadecapole moment (q40) is found at large deformations. With the q40 constraint, a smooth and continuous potential energy surface (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 180Hg. The asymmetric distribution of the fission yields of 180Hg is further reproduced by the time-dependent generator coordinate method (TDGCM), and agrees well with the 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 via the β decay of 180Tl [1]. For the fission of 180Hg, its splitting into two 90Zr fragments with magic N = 50 and semimagic Z = 40 was believed to dominate the fission process. However, unlike the initial theoretical prediction, 180Hg has been observed to fission asymmetrically, with heavy and light fragment mass distributions centered around A = 100 and 80 nucleons, respectively [1, 2].

A lot of theoretical research attention has been drawn to the puzzling fission behavior of 180Hg. For example, macroscopic-microscopic models [1, 3–6] and self-consistent microscopic approaches [7–9] were used to analyze the multidimensional potential energy surfaces (PESs), and the presence of an asymmetric saddle point with a rather high ridge between symmetric and asymmetric fission valleys was explained as the main factor determining the mass split in fission.

Calculations of fission-fragment yields have also been done for 180Hg by means of the Brownian Metropolis shape-motion treatment [3, 5, 10], Langevin equation [11], scission-point model [4, 12–14], the random neck rupture mechanism [15], based on the PESs or scission configurations. The results are in approximate agreement with the experimental data, with a deviation of 4 nucleons for the peak positions. There were also several attempts to describe fragment mass distribution of 180Hg in a fully microscopic way, i.e., the time-dependent generator coordinate method (TDGCM) based on covariant density functional theory (CDFT) [9], and the asymmetric peaks are reproduced very well, while a more asymmetric fission mode with AH 116 is predicted, which was not observed in experimental measurement.

In the theoretical study of nuclear fission, the PES is an important infrastructure, which describes the evolution of nuclear energies with shape variations on its way from the initial configuration towards scission. In nuclear physics, there are generally two approaches to generate PES. One ranges from the historical liquid drop model [16–22] to the well-known macroscopic-microscopic model, using parametrization of the nuclear mean-field deformation [23–33]. The other is based on microscopic self-consistent methods [34–43], or the constrained relativistic mean-field method [9, 44–50].

In the macroscopic-microscopic method, a predefined class of nuclear shapes is defined uniquely in terms of selecting appropriate collective coordinates, and the relatively smooth potential energy surface can be obtained. However, due to limitations in computing resources, the microscopic calculation 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, the quadrupole and octupole deformation (moments) constraints are the natural and most often used to calculate microscopic PES.

However, several studies have shown that as a consequence of the absence of hexadecapole deformation (q40 or $ $4), PES may exhibit discontinuities in the large deformation scission regions [51–55]. Ref. [56] investigated the role of hexadecapole deformation on the PES calculation of 240Pu by applying a disturbance on $ $4. $The results show that one can obtain a smooth 2-dimensional PES in$ ($2, $ $3) by parallel calculations with a suitable disturbance of hexadecapole deformation.

But for asymmetric fission of 180Hg, there have been no reports about the effect of q40 or $ $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 [7, 9]. Thus it is interesting to examine the influence of hexadecapole moment on PES of 180Hg at large deformation, and also analyze some properties of scission configuration. In this paper, we will 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 the TDGCM [57] will be described and discussed.

II. THEORETICAL FRAMEWORK

To study the static fission properties, the PES was determined by using the Skyrme density functional theory (DFT). The dynamic process is further investigated in the framework of TDGCM. Thus, in this section, we explain these two methods briefly. The detailed description of Skyrme DFT can be found in Ref. [58], and the formulations of TDGCM can be found in Refs. [57, 59–61].

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),

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

In the above equation, $\mathcal{T}(r)$, $\mathcal{V}_t(r)$ and $\tilde{\mathcal{V}}_t(r)$ stand for the density of the kinetic energy, the potential energy and the pairing energy respectively. The symbol $t = 0, 1$ denotes the isoscalar or isovector, respectively [62].

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

$$\mathcal{V}_t(r) = C_t^{\rho\rho}\rho_t^2 + C_t^{\rho\Delta\rho}\rho_t\Delta\rho_t + C_t^{\rho\tau}\rho_t\tau_t + C_t^{\rho\nabla J}\rho_t\nabla\cdot J_t + C_t^{J^2}J_t^2 + C_t^{\rho\rho}D\rho_0^\gamma$$

where the particle density ρ_t, kinetic density τ_t, and the spin current vector densities $J_t(t=0,1)$ can be calculated by the density matrix $\rho_t(\mathbf{r}\sigma;\mathbf{r}'\sigma')$, with the dependence of spatial ($\mathbf{r}$) and spin (σ) coordinates. $C_t^{\rho\rho}$, and etc. are coupling constants for different types of densities in the Hamiltonian density $H(r)$, which are usually real numbers. As an exception, $C_0^{\rho\rho}$ is the density-dependence term. The formulations of the relation of the coupling constants to traditional Skyrme parameters can be found in Ref. [63]. For example, spin-orbit force of the Skyrme interaction corresponds to the term $C_t^{\rho\nabla J}\rho_t\nabla\cdot J_t$.

The pairing correlation is often taken into account through the Hartree-Fock-Bogoliubov (HFB) approximation in DFT [58]. 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. [40, 64]:

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

where $V_0^{(n,p)}$ is the pairing strength for the neutron (n) and the proton (p), ρ_0 is the saturation density of nuclear matter fixed as 0.16 fm^{-3}, and $\rho(\mathbf{r})$ indicates the total density. As studied in Ref. [40], this type of pairing force is a suitable choice for nuclear fission study.

The DFT solver HFBTHO(V3.00) [65] is used to generate the PESs, in which axial symmetry is assumed. 26 major shells of the axial harmonic-oscillator single-particle basis are used, and the number of basis states are further truncated to be 1140. In this work, the Skyrme DFT with SkM* parameters [66] is adopted, which is commonly used for fission studies. For the strength of pairing, $v_0^{(n)} = -268.9$ MeV $\cdot$ fm^3 and $v_0^{(p)} = -332.5$ MeV $\cdot$ fm^3 are used for the neutron and the proton respectively, with the 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. [7, 67], 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, which could 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

$$|\Psi(t)\rangle = \int f(\mathbf{q};t)|\Phi(\mathbf{q})\rangle d\mathbf{q}$$

where $|\Phi(\mathbf{q})\rangle$ is composed of known many-body wave functions with the vector of continuous variables $\mathbf{q}$. The $\mathbf{q}$ 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, the $f(\mathbf{q};t)$ is a weighted function. It is determined by the time-dependent Schrödinger-like equation,

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

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

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

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

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

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

as in Ref. [57], in which $J(\mathbf{q};t)$ is the current

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

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

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

where S_A denotes an ensemble of all the surface elements on the scission contour containing the fragment with mass number A, and C is the normalization factor

to ensure that the total yield is normalized to be 200. In the same way, the yield of fission fragment with charge number Z can also be obtained.

In this work, the computer code FELIX(version 2.0) [61] 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, the precise multi-dimensional PES is the first and essential step toward the dynamical description of fission. Fig. 1 displays the PES contour of 180Hg obtained by the HFB calculation in the collective space of (q_{20}, q_{30}), in which q_{20} is from -20 b to 300 b and q_{30} is from 0 $b^{3/2}$ to 40 $b^{3/2}$ with the step of $\Delta q_{20} = 2$ b and $\Delta q_{30} = 2$ $b^{3/2}$. 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 HFBODD [7] with the same functional and that obtained using covariant density functional theory with the relativistic PC-PK1 functional [9]. The static fission path starts from a nearly spherical ground state ($q_{20} = 20$ b, $q_{30} = 0$ $b^{3/2}$), the reflection symmetric fission path can be found for small quadrupole deformations, and the reflection-asymmetric path branches away from the symmetric path about $q_{20} = 100$ b. One can see that unlike the PES of actinide nuclei, there is no valley towards scission for 180Hg; it undergoes a continuous uphill process until the mass asymmetric scission point with high q_{30} asymmetry.

In the (q_{20}, q_{30})-constrained PES calculations by DFT, the other degrees of deformation are obtained based on the variational principle. In Refs. [55, 56], it has been learned that at given q_{20} and q_{30}, there are two minima with different values of q_{40}, and the minimum with larger q_{40} disappears when q_{20} is large enough, which indicates the transition toward scission. The hexadecapole deformation is an important degree of freedom for the description of PES at large deformation, and especially, a disturbance of hexadecapole deformation is required for a smooth and reasonable PES, as shown in Ref. [56]. Thus, in our work, at large quadrupole moments, i.e., larger than $q_{20} \simeq 200$ b ($\beta_2 \simeq 3.2$), a further constraint of hexadecapole moment q_{40} is introduced. It is done 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 q_{40} might be obtained. This treatment is used for the calculation of PES in Fig. 1, and labeled "(B)" in Figs. 2-3. The calculation with the constraints of q_{20} and q_{30} only is labeled "(A)" in Figs. 2-3.

In Fig. 2, the energies of the static fission path as a function of quadrupole moment q_{20} are shown. The symmetric ($q_{30} = 0$ $b^{3/2}$) and asymmetric fission paths in 180Hg are given respectively. One can see clearly that these energies increase with q_{20} steadily. At around $q_{20} \sim 100$ b, the asymmetric fission path starts to be favored in energy compared to the symmetric fission 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. [68] using the HFB-Gogny D1S interaction.

From Fig. 2, for case (A), one can see that the energy of 180Hg increases continuously with q_{20}, and it is difficult to rupture even at very large elongation, e.g., $q_{20} \geq 300$ b ($\beta_{20} \geq 4.8$). As seen in case (B), with the inclusion of the q_{40} constraint, a gentle descent trend of the energy happens at $q_{20} \sim 240$ b, and a sudden drop in energy occurs at $q_{20} \sim 280$ b, indicating nuclear scission.

In Fig. 3, the hexadecapole moment (q_{40}), the average particle number around the neck (q_N) and the HFB energies are given as functions of q_{20} respectively, at given q_{30}. In order to investigate the role of q_{40}, only the region with large q_{20} is shown. From Fig. 3(a), one can see that the q_{40} increases nearly linearly until very large q_{20} values, especially for case (A), in which the q_{40} can become very large during elongation. After a "perturbative" constraint on q_{40}, as case (B) in the figure, the q_{40} value has a sudden drop and then grows linearly. In the study of nuclear fission, q_N is often adopted as the indicator of nuclear scission. For example, $q_N = 4$ has been used for the determination of scission line of 240Pu in Refs. [40, 54, 69]. In Fig. 3(b), q_N decreases gradually against q_{20}. However, for case (A), the reduction of q_N becomes rather slow with the increase of q_{20}. Especially, at $q_{20} \sim 340$ b ($\beta_2 \sim 5.4$), $q_N > 4$ for $q_{30} = 0$ and 10 $b^{3/2}$, and the total energies increase continuously at large q_{20}, as seen in Fig. 3(c). After considering the q_{40} constraint, as case (B) shown in Fig. 3(b) and (c), both q_N and the total energy have a sudden drop at around $q_{20} \sim 280$ b ($\beta_2 \sim 4.5$), indicating nuclear rupture. It is seen that when $q_N \leq 4$, q_N becomes close to zero with the increase of q_{20}.

To investigate the role of q_{40} on PES, the HFB energies and q_N against q_{40} at given q_{20} and q_{30} are plotted in Fig. 4, which are obtained through the exact constrained calculations of q_{20}, q_{30} and q_{40}. In the figure, q_{30} is constrained to be 0 $b^{3/2}$. Other q_{30} 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 q_{40} degree of freedom, which correspond to distinct valleys on the multi-dimensional potential energy surface. In Ref. [56], a similar trend in 240Pu has been found, and the minimum related to the larger q_{40} disappears with increasing quadrupole deformation (at roughly $\beta_2 \sim 3.8$), leading to a natural transition to the minimum with the smaller q_{40}. This transition is the cause of the discontinuity and the sudden drop of energy in the two-dimensional PES of (q_{20}, q_{30}). However, for 180Hg, there remains an extremely soft and relatively flat minimum with larger q_{40} value even at very large q_{20} values, e.g., at $q_{20} = 340$ b ($\beta_2 \sim 5.4$). In PES calculation with only the (q_{20}, q_{30}) constraint, the q_{40} degree of freedom is obtained by variations of the total energies. As seen for case (A) in Fig. 3, q_{40} after the variation calculation grows steadily even at very large q_{20}, and no transition to the minimum with smaller q_{40} happens. With

only (q_{20}, q_{30}) constraint, it is difficult to find the proper scission configuration, at least for 180Hg. After the "perturbative" inclusion of q_{40} constraint, as for case (B) in Fig. 3, such transition can occur at large q_{20}. In Fig. 4(b), it is seen that q_N increases with q_{40} in general. q_N around the minimum with larger q_{40} is roughly larger than 4, and when $q_{20} > 200$ b its value around the minimum with smaller q_{40} is close to zero (numerically 10^{-3}-10^{-4}, effectively near zero). For $q_N \sim 0$, the nucleus becomes well separated into two fragments. From this calculation, one can learn that the introduction of q_{40} constraint in the self-consistent calculation of PES can ensure the continuity of the potential energy surface. q_{40} is essential in the DFT calculation for fission study, especially for the transition to scission.

Several results of (q_{20}, q_{30}, q_{40}) constrained calculations have been shown in Fig. 5 for the density distribution profiles of 180Hg. q_{20} is constrained to be 240 b, and q_{40} changes from 140 b^2 to 60 b^2 for $q_{30} = 0$ $\mathrm{b}^{3/2}$ and $q_{30} = 10$ $\mathrm{b}^{3/2}$ in the upper and lower panels respectively. The q_{40} degree of freedom influences the formation of the neck and the scission configurations. From the figure, one can see that with large q_{40}, there is no neck in the nucleus and the nucleus is just stretched very long. For the calculation with only (q_{20}, q_{30}) constraint as case (A) in Figs. 2-3, q_{40} has a very large value with the increase of q_{20} and thus the nucleus cannot undergo scission. With the decrease of q_{40}, the neck structure of the nucleus appears and becomes well separated when q_{40} has small values.

One of the most important quantities in induced fission is the total kinetic energy (TKE) carried out by the fission fragments. In this work, the total kinetic energy of the two separated fragments at scission point can be approximately estimated as the Coulomb repulsive interaction by using a simple formula $e^{2Z}_{HZ}L/d_{\mathrm{ch}}$, where e stands for the proton charge, Z_H and Z_L denote the charge numbers of the heavy and light fragments respectively, and d_{ch} is the distance between the centers of charge of the two fragments at the scission point. Fig. 6 displays the distribution of calculated Coulomb repulsive energy based on the scission line shown by the purple circle in Fig. 1 and compared with the measured TKE [2]. It can be seen that the calculated results reproduce the trend of the measured TKE quite well, especially a dip at $A_H = 90$ and peak at $A_H = 94$, although the calculated results are generally overestimated by several MeV compared to data, which might be caused by the neglect of dissipation effects.

Finally, we performed the TDGCM+GOA calculation to model the time evolution of the fission dynamics of 180Hg. Fig. 7 shows the calculated mass distributions of the fission fragments of 180Hg, in comparison with the experimental data [1, 2]. The theoretical result in the framework of covariant density functional theory (CDFT) using PES generated with the neck coordinate constraint q_N from Ref. [9] is also given in the figure for comparison, denoted as "CDFT". As one of the most important microscopic inputs of fission dynamic calculations, the mass tensor is calculated by GCM or ATDHFB methods in the present work. The calculated mass distribution is generally similar when using the mass tensor by these two methods, and better agreement is obtained by us-

ing the GCM method for the height of asymmetric peaks and symmetric valley. Overall, the calculation reproduces the experimental data well. The calculated peak position deviates by one unit from the experimental peak position. The results of CDFT show good asymmetric peak positions, but overestimate the data and even predict a little peak for the symmetric valley. The deviation of the peak position from the data might be caused by the mean-field potential. In the analysis of the PES for 180Hg in Ref. [7], it was found that the mass of the optimum fission fragment at static scission point varies by two units when using either the Skyrme force or Gogny force. Here, in the dynamic calculation results, one also observes discrepancies in the peak positions between the results based on Skyrme-DFT and CDFT. The tail of the fission fragment distribution is sensitive to the approximation of the mass tensor in the dynamic calculations. The width of the distribution becomes slightly wider when using the GCM mass tensor.

Moreover, a more asymmetric fission mode with $A_H \sim$ 116-117 is predicted both in this work and CDFT calculation. As explained in Ref. [9], this mode resulted from the usage of the initial state with mixed angular momenta while in the experiment there are only certain values due to the selection rule of electron capture of 180Tl. In the current work, these discrepancies from the data still exist as the initial state with mixed angular momenta is also applied.

IV. SUMMARY

In this work, the static fission properties and the fission dynamics of 180Hg were investigated by the Skyrme DFT and TDGCM respectively. During the calculation of multidimensional PES, it was found that the hexadecapole moment is crucial to obtain a smooth PES and proper scission configurations, and thus it is essential for fission dynamic studies. For the calculation of PES with only the q_{20} and q_{30} constraints, nuclear rupture does not happen even at very large q_{20}. Through calculations with q_{20}, q_{30} and q_{40} constraints, it was found that a rather soft and flat minimum with large hexadecapole moment still exists in the PES of 180Hg even with a very elongated shape, which hinders the transition to the lower energy minimum with smaller q_{40}. With the strategy of "perturbative" constraint of the collective freedom q_{40}, the transition to the minimum corresponding to nuclear rupture could happen 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 the static PES calculation, it was learned that the asymmetric fission channel is favored in 180Hg. Finally, the fission fragment yields were calculated with the TDGCM. The calculated mass distributions also support the asymmetric fission for 180Hg. The calculation agrees well with the experimental data. Moreover, a more asymmetric peak with $A_H \sim 117$ is predicted, which is also predicted by the covariant DFT with PC-PK1 parameter set [9].

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