Fission dynamics within time-dependent Hartree-Fock: deformation-induced fission

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Background: Nuclear fission is a complex large-amplitude collective decay mode in heavy nuclei. Microscopic density functional studies of fission have previously concentrated on adiabatic approaches based on constrained static calculations ignoring dynamical excitations of the fissioning nucleus, and the daughter products.

Purpose: To explore the ability of dynamic mean-field methods to describe fast fission processes beyond the fission barrier, using the nuclide $^{240}$Pu as an example.

Methods: Time-dependent Hartree-Fock calculations based on the Skyrme interaction are used to calculate non-adiabatic fission paths, beginning from static constrained Hartree-Fock calculations. The properties of the dynamic states are interpreted in terms of the nature of their collective motion. Fission product properties are compared to data.

Results: Parent nuclei constrained to begin dynamic evolution with a deformation less than the fission barrier exhibit giant-resonance-type behaviour. Those beginning just beyond the barrier explore large amplitude motion but do not fission, whereas those beginning beyond the two-fragment pathway crossing fission to final states which differ according to the exact initial deformation.

Conclusions: Time-dependent Hartree-Fock is able to give a good qualitative and quantitative description of fast fission, provided one begins from a sufficiently deformed state.

I. INTRODUCTION

Studies of nuclear fission have been ongoing since the discovery of the process in 1938 by Hahn and Strassmann [1]. The actinide nuclide $^{240}$Pu has long been a case of interest, as spontaneous fission presents itself as a decay mechanism with significant probability, relative to other isotopes in the actinide region. This allows for quantitative comparisons between spontaneous and induced fission [2–5]. Experimentally, fission can be induced by a variety of techniques, including neutron-induced fission; fission induced by more complex projectiles, and photofission [2, 6]. Recent experimental campaigns have investigated beta-delayed fission [7].

Theoretically, microscopic studies have focussed upon the role of the quadrupole degree of freedom in forming the fission pathway, as exemplified by constrained mean-field calculations [8, 9]. The typical observed behaviour in actinide nuclei for the binding energy as a function of increasing quadrupole deformation is to follow a multi-humped pathway (see Fig. 1). When considering the potential energy surface (PES), starting from the ground state then increasing the quadrupole deformation will result in a first fission barrier. By increasing the deformation further, a secondary minimum, corresponding to an isomer, is found. Beyond this minimum, a second fission barrier is encountered, and past this barrier, the general consensus is that it becomes more energetically favourable for the nucleus to fission. The energies $E_A$, $E_B$ and $E_{II}$ presented in Fig. 1 correspond to those defined in Ref. [10]: the energy difference between the ground state and the peak of the first fission barrier; the difference between the ground state and the peak of the second fission barrier, and the difference between the ground state and fission isomer, respectively. In some exotic cases, triple-humped potential surfaces are expected [11–14]. Although the multi-humped behaviour of the energy surface cannot be measured directly, experimental evidence points towards this characteristic structure [13, 15].

For fission studies, the quadrupole degree of freedom is of capital importance, as it describes the elongation of the nucleus [16]. Additionally, as many nuclei are ob-
served to fission with asymmetric mass distributions, the octupole degree of freedom is vital to describe any mass reflection asymmetry. Modern DFT solvers are able to perform symmetry unrestricted calculations which allow, in principle, any and multiple degrees of freedom to be explored [17]. As well as considering deformation degrees of freedom, constraints can be imposed from alternate perspectives to study fission in static calculations. Some studies, for instance, assume a priori knowledge of the fission fragments and constrain the distance between the two centres of mass to generate initial states before investigating time evolution [18, 19].

The approach of calculating the PES to describe fission, regardless of the number of particular degrees of freedom constrained, is limited to producing a series of static solutions which attempt to describe a dynamic process, resulting in an effectively adiabatic approximation. Shape-constrained DFT calculations produce Slater determinants which contain no internal excitations. Some attempts have been made to account for finite temperature effects [20], but as fission is a dynamic process, translational motion and collective excitations will most likely be present. Time-dependent techniques coupled with constrained calculations may therefore yield new, insightful results, as they describe the dynamics of a fissioning system. Time-dependent Hartree-Fock (TDHF) [21] presents itself as a candidate method, as it is able to temporally evolve Slater determinants which begin as a solution to the constrained static Hartree-Fock equations.

The calculational basis for this study is the so-called TDHF technique. We note, however, that nowadays this should probably be called “time-dependent density functional”, since it is based upon a nuclear energy density functional. The slightly less correct TDHF moniker has stuck within the nuclear physics literature, though, and we keep this convention in the present work.

TDHF is the basic lowest-order microscopic dynamical mean-field theory, first proposed by Dirac [21], and later on applied to more or less realistic nuclear systems in the 1970s-1980s [22–26]. A practical implementation involves beginning from an energy density functional, and using the variational principle to obtain Hartree-Fock-like equations for the static, initial state. The time evolution equations, based on the same energy density functional, are then run on this initial state. We use the Skyrme energy density functional, depending on the local densities and currents,

$$E = E_{\text{sky}}(\rho, J, \tau, s, j, \xi)$$  

(1)

where $\rho$ is the particle density, $J$ is the vector part of the spin-current tensor, $\tau$ is the kinetic density, $s$ is the spin density, $j$ is the particle current and $\xi$ is the pairing density [27]. These densities and currents include time-odd fields ($s$ and $j$), which are only active in the dynamic part of the calculation. Only those time-odd fields that couple to the necessary time-even fields through Galilean invariance are included in the present calculation. Full details of the practical aspects of solving the static and dynamic Hartree-Fock equations, along with the explicit details of the density functional are contained in the documentation of the Sky3D code [28].

There have been several historic attempts to describe fission dynamics using TDHF. The very first studies, in the late 1970s and early 1980s, suffered from computational power limitations, and were hindered by axial symmetry and restricted forms of the nucleon-nucleon interaction [24, 29, 30]. With a new generation of TDHF solvers able to perform symmetry-unrestricted, three dimensional calculations [28, 31, 32], modern TDHF studies have begun to take a renewed interest in fission [18, 33, 34]. Here, we will use a modified version of the recently published code Sky3D [28], with shape constraints explicitly included. We will then solve the Hartree-Fock + BCS equations in a three-dimensional Cartesian basis, and evolve the calculated states using TDHF.

Spontaneous fission cannot be accessed directly within TDHF. To reach a fissioned configuration from the ground or isomeric state, the nucleus must tunnel through the barriers in the potential energy surface. While TDHF allows a quantum mechanical description of single-particle wave functions, the collective motion is semi-classical, hence forbidding tunnelling in collective coordinates [35]. In contrast, TDHF is suitable for exploring the dynamics of induced fission. The potential challenging issue is the incorporation of the fissioning mechanism within the TDHF framework. Here, we follow the strategy of finding constrained Hartree-Fock (CHF) states and use them as initial conditions in a THDF calculation. Our aim is to investigate how the underlying deformed structure pushes the parent nucleus towards fissioning paths, if it does so at all.

We will also run TDHF simulations from initial states that lie past the fission barrier. At that stage, one is somehow mimicking spontaneous decay, in the sense that these represent the states right after tunnelling. Tunnelling would have populated randomly these points. Our dynamical calculations only take a handful of initial states, but consider the dynamics afterwards.

The paper is organised as follows. In Section II, the static constrained starting points are discussed extensively. Section III explores the dynamics of the fissioning nucleus, while section IV looks in more detail at the dynamics of the fissioning nuclei. Some concluding remarks are given in Section V.

II. STATIC CONFIGURATIONS IN $^{240}$Pu

We begin the investigation of fission by examining the potential energy surfaces for a nucleus of interest. The reproduction of the double-humped fission barriers of actinide nuclei are often used as a benchmark test for nuclear models [36, 37]. Due to the wealth of data available from experimental [2–5] and theoretical [9, 10, 38, 39] studies, $^{240}$Pu presents itself as a strong candidate for a
benchmark test of TDHF to investigate induced nuclear fission [37]. We note, in particular, that configuration mixing plays a relatively small effect in this isotope [40]. The static quadrupole constrained PES will firstly be calculated to provide a selection of initial states for time evolution. These are obtained by performing an energy minimisation with respect to constraints imposed upon the quadrupole shape degree of freedom.

Modern symmetry unrestricted DFT solvers have extended the PES for multiple constraints, for example simultaneously constraining the quadrupole and octupole degrees of freedom to explore two-dimensional deformation surfaces [41]. The approach of calculating a multi-dimensional PES to describe fission within a microscopic framework has enjoyed much recent attention [14, 20, 41–43]. Alternatively, one can explore fission pathways via shell-corrected macroscopic liquid drop models. This technique has been applied to perform exhaustive topographical surveys of deformation space to deduce fission properties of static configurations [10, 44–46]. However, this approach, regardless of the number of dimensions, is limited to producing a series of static solutions to describe a dynamic process, thus being equivalent to adiabatic motion. Time-dependent techniques can yield new, insightful results as they can describe the dynamics of a fissioning system allowing for internal excitation. This is important for the final, fast stages of fission, which we explore in the present work.

To build shape-constrained ground states, we begin from an arbitrarily deformed state and use the augmented Lagrangian method [41, 47] to constrain the quadrupole deformation. Our purpose is not to pursue an in-depth investigation of multiple shape-constraints in Hartree-Fock calculations, but rather to use the technique to produce initial states to then investigate their time evolution. All other degrees of freedom are assumed to settle into the configuration of minimum energy [35]. The constrained energy minimisation on quadrupole degrees of freedom requires the definition of a principal axis. In our calculations, the principal axis of the nuclear ground state always aligns with one of the Cartesian axes of our box. We describe this alignment procedure, which is necessary in the calculations, in Appendix A.

For the results presented here, where only one constraint is applied upon the quadrupole degree of freedom, a masking procedure has also been adopted. This limits the space which the nuclear wave functions can explore, allowing a single fission pathway to be explored where the nuclear shape gradually evolves, rather than abruptly jumping between competing energy minima. Details of the masking procedure are discussed in Appendix B. Our emphasis is on the fast fission dynamics beyond the barrier, but it may yet be fruitful to use other constraints to generate more starting points.

The ground state and constrained Hartree-Fock calculations are performed using the SkM* effective interaction. The fission barrier properties of $^{240}$Pu were considered when fitting the SkM* effective interaction [48].

### Table I. Summary of ground state and isomer properties

| Nucleus | Binding Energy [MeV] | rms Radius [fm] | $\beta_{20}$ | $\beta_{30}$ | $\beta_{40}$ |
|---------|----------------------|-----------------|-------------|-------------|-------------|
| $^{240}$Pu | -1781.95 | 5.941 | 0.280 | 0.000 | 0.255 |
| $^{240}$Pu* | -1778.91 | 6.418 | 0.682 | 0.000 | 0.547 |

We perform our static calculations in a regularly spaced Cartesian grid of $40 \times 40 \times 40$ points, ranging from $-19.5$ fm to 19.5 fm in the $x$, $y$, and $z$ directions. BCS pairing is included within the static calculation, using the Volume-Delta interaction [28], with 184 neutron and 126 proton single-particle wave functions and pairing strengths $V_{0,n} = 258.96201$ MeV and $V_{0,p} = 270.08200$ MeV for neutrons and protons, respectively. The PES for $^{240}$Pu presents a prominent local minimum, corresponding to a fission isomer. If the initial test wave functions (harmonic oscillator states in the case of Sky3D) are chosen to be prolate with similar deformations, shape unconstrained static calculations converge directly into the isomeric state. This provides two initial points for the CHF calculations, starting at either the ground or the isomeric state. Some properties of the ground state and isomer are presented in Table I. These compare well with previous results in the literature [9].

One-dimensional projections of the quadrupole-constrained PES for $^{240}$Pu are shown in the top three panels of Fig. 2. The top left panel focuses on the dependence of the energy on the constrained quadrupole degree of freedom. The central and left panel give the corresponding octupole and hexadecapole coordinates associated to each quadrupole configuration. The color scheme is chosen to display changing quadrupoles, for guidance. Two fission barriers are found in the quadrupole degree of freedom (top left panel), peaking at $\beta_{20} \approx 0.50$ and 0.86 respectively. The ground and isomeric states correspond to the two minima in the PES next to these barriers.

Table II presents a comparison between various features of the PES calculated in this work and previous literature. The Table also contains measurements of the fission barrier heights and energy differences between the ground and isomeric state, as defined in Fig. 1. We compare our calculations to recent Hartree-Fock-Bogoliubov (HFB) calculations using the Gogny interaction [39]; to older Hartree-Fock calculations employing SIII [9]; and to macroscopic-microscopic calculations [10]. There is a general agreement between the barrier geometries in the theoretical methods. The older SIII calculation predict a large second barrier height, most likely because of the assumption of axial symmetry. HF calculations yield barriers higher than either the experimental or the macroscopic-microscopic predictions, perhaps caused by a lack of dynamical effects. The details of the barri-
FIG. 2. (Color online) Resulting PES for $^{240}$Pu following a constraint of the quadrupole deformation parameter $\beta_{20}$. The top panels display the dependence of the constrained energy on the quadrupole (left panel), octupole (middle panel) and hexadecapole (right panel) moments. Once the second barrier is overcome, a significant octupole deformation (corresponding to mass asymmetry) develops. The isolines in the 2D slices of the 3D density correspond to 0.05 particles/fm$^3$. It is interesting to note that, for states far beyond the second barrier, scission has not yet occurred.

ers shapes should be sufficiently well reproduced in the present calculations to deal with the fast fission dynamics beyond the second barrier, as well as to give a qualitative description of the between-barrier dynamics.

The top central panel of Fig. 2 shows a prominent octupole deformation setting in at the second fission barrier, as would be expected [36]. The relationship between the quadrupole and octupole deformation parameters for the configurations along the PES is explored in Fig. 3 (top panel). Although the calculations have been performed constraining only one deformation degree of freedom, the observed behaviour is typical for the optimum static fission pathway obtained in quadrupole-octupole constrained deformation surfaces calculated using DFT [14, 41]. Beyond the second fission barrier, octupole degrees of freedom are explored. Fig. 3 also displays the relationship between the quadrupole and hexadecapole deformation parameters (central panel). Near the peak of the first and second barrier, the hexadecapole deformation sharply drops, and recovers subsequently. This corresponds to a transitioning shape as the neck region of the nucleus thins.

The 3D calculations verify that triaxiality is explored at the first fission barrier. The bottom panel of Fig. 3 shows unambiguously a region of non-zero values of $\gamma$, indicating that quadrupole-constrained calculations explore other deformation multipoles. Access to these additional degrees of freedom lowers the calculated barrier height with respect to axially symmetric calculations [36]. We note that triaxiality is explored significantly in the range $0.36 \leq \beta_{20} \leq 0.59$, but it is virtually negligible elsewhere.

The slices of the density in the lower panels of Fig. 2 display an increasingly deformed shape as the quadrupole degree of freedom grows. Interestingly, the nucleus has not fissioned in the range of $\beta_{20}$ considered, even for states beyond the second fission barrier. Despite the emergence of a competing fission pathway, a large selection of increasingly deformed states have been obtained. These are useful as starting points for our time-dependent calculations. We note that our configurations explore a range of states, from configurations with a quadrupole deformation less than that of the global Hartree-Fock minimum, to configurations well beyond the second fission barrier.

When performing constrained Hartree-Fock calcula-
TABLE II. Comparison of properties of the fission barrier for $^{240}$Pu from different calculations (defined in Fig. 1). In addition to our work, we present the calculations of Flocard et al. [9], Rodriguez-Guzman and Robledo [39], Moller et al. [10] and the experimentally inferred data presented in Ref. [10].

|          | $E_A$ [MeV] | $E_B$ [MeV] | $E_{11}$ [MeV] | Method                                      | Reference                  |
|----------|-------------|-------------|---------------|--------------------------------------------|----------------------------|
| This work| 8.25        | 7.68        | 3.04          | HF+BCS, Skyrme SkM$^*$                       |                            |
|          | 8           | 13          | 4             | HF+BCS, Skyrme SkMII                       | Table 2 of Ref. [9]        |
|          | 9.30        | 8.40        | 3.10          | HFB, Gogny D1M                             | Fig. 5 of Ref. [39]        |
|          | 5.99        | 4.91        | 2.94          | Shell-Corrected FRLDM                      | Table I of Ref. [10]       |
|          | 6.1±0.3     | 6.0±0.50    | 2.1±0.6       | Experiment                                 | Fig. 27 of Ref. [10] (Madland) |
|          | 5.6±0.2     | 5.1±0.20    | 2.4±0.3       | Experiment                                 | Fig. 27 of Ref. [10] (Madland) |

FIG. 3. (Color online) Octupole deformation as a function of quadrupole deformation (top panel) and hexadecapole deformation as a function of quadrupole deformation (central panel) for the calculated PES of $^{240}$Pu. The octupole deformation, corresponding to a mass asymmetry, rapidly onsets after the second fission barrier is passed. The lowest panel displays the corresponding $\gamma$ deformation parameter.

tions, we noticed that beyond a quadrupole deformation of $\beta_{20} = 1.25$, the configuration jumped abruptly to a competing two-fragment fission pathway. This behaviour, due to the numerics as the calculations converged, proved to be unavoidable, even when adopting the masking procedure described in Appendix B. This can be explained by considering the density slices presented in Fig. 4. A mask around the one-fragment configuration does not inhibit a transition to the two-fragment configuration, as the two-fragment configuration fits inside the one-fragment masking region.

The competing two-fragment fission pathway was explored, starting from the state after the calculations jumped pathways. From this configuration, the deformation was incrementally reduced. This competing pathway is shown in Fig. 4 with (blue) solid squares, and may be compared to the original, one-fragment pathway in (red) solid circles. Once the quadrupole deformation parameter is reduced below $\beta_{20} = 1.01$, the Hartree-Fock minimum jumps back onto the original fission pathway.

The competing pathway, referred hereafter as the two-fragment pathway [39], displays remarkably different configurations to that of the one-fragment pathway. Even with identical quadrupole deformations, the octupole and hexadecapole deformations and total energy differ significantly. It is exactly this behaviour that the authors of Refs. [10] and [46] identify as a flaw when using CHF to explore the PES. Here, we exploit this feature to gain an insight on the competing fission pathway, without having to include a higher number of constraints in the CHF calculations.

The fragments in the two-fragment pathway do not have an integer particle number. For example, for the case of $\beta_{20} = 1.19$, the fragments have $A_1 = 107.14$, $Z_1 = 43.14$, and $A_2 = 132.85$, $Z_2 = 50.85$. We note, however, that all the fragments in the two-fragment pathway correspond, to the nearest integer particle number, to $^{107}$Tc and $^{133}$Sb. It would be instructive to project the individual fragments onto a good particle number [49], since this would give access to a mass distribution. Our focus here is on the dynamics, though, so we will postpone this for future work.

III. TIME EVOLUTION OF CONSTRAINED HARTREE-FOCK STATES

The time evolution of the CHF states obtained for $^{240}$Pu may be investigated using the TDHF method. This analysis will focus on the states on the one-fragment fission pathway, starting from configurations beyond the fission isomer (that is, those with $\beta_{20} > 0.68$). We will investigate the effect of releasing the imposed shape constraints and time evolving the constrained static states, to gain an insight of the deformation-induced fission (DIF) process in TDHF.

The Sky3D code provides the capability to time-evolve
FIG. 4. (Color online) One- (solid circles) and two-fragment (solid squares) fission pathways for $^{240}$Pu. The arrows in the top left panel show the direction in which the PES is explored. Beyond $\beta_{20} = 1.01$ the one-fragment pathway jumps into the two-fragment pathway, and this state is used as the initial configuration for investigating the latter pathway. Sample density slices on the competing pathways with the same $\beta_{20}$ are shown in the lower panels. The isolines are separated by 0.05 particles/fm$^3$.

the static Hartree-Fock states obtained in the previous Chapter. As the single-particle wave functions remain orthogonal, dynamic calculations can be performed efficiently using Message Passing Interface (MPI) on high performance computing facilities. At every time step, the wave functions can be allocated onto separate nodes, where the time iteration may be performed. After all the wave functions have been iterated one step, they can be collected to obtain the densities, which in turn define the time-dependent single particle Hamiltonian.

TDHF calculations may be performed in a larger grid than that used to calculate the initial (static) state. The Slater determinant, which is a solution to the CHF problem, is placed in a grid of dimension $(42^3)$ points, ranging from $-20.5$ to $20.5$ fm in the $x$, $y$ and $z$ directions, with the centre of mass at $(x, y, z) = (0, 0, 0)$. The wave functions are set to zero for all the space which exceeds the dimensions of the static solution. The wave functions are also re-orthonormalised before time evolution begins. The BCS occupations remain frozen throughout the dynamical simulation.

One important consideration is that excited states will decay by particle emission in TDHF. This corresponds to the dispersion of the wave functions as time evolves. The dimensions of the grid and the boundary conditions will have some effect upon the measurements of observables, as the single particle wave functions, especially those of emitted particles, are free to explore the entire space of the calculation. Spherical TDHF calculations can be performed in an analytic continuum [50, 51]. Some progress has been made to numerically reproduce continuum calculations in 3D, but the methods are computationally expensive and may not be suitable for large amplitude processes [52, 53].

When analysing nuclear dynamics, a masking procedure is adopted to define the nucleus. This eliminates contributions to the observables from the tails of the wave function and ensures that consistent results are obtained with different choices of grid dimensions. The mask has a Fermi function shape, in line with the procedure described in Appendix B for the static case. A generalisation becomes necessary in the dynamic case, as the fragments in a post-fissioned system propagate through space. The mask is then defined from a fixed distance from the centre of mass, which is not necessarily a grid point. This is in contrast to Eq. (B1), which is computed relative to discrete grid points. With this, we avoid artificial discontinuities in observables resulting from jumps of the central mask position.

The time evolution of the CHF solutions yields two general cases. When the nucleus is sufficiently deformed, the repulsive Coulomb interaction is able to drive the
configuration to fission. For a fissioning system, once the system splits into two fragments, we define a separate mask for each fragment. Details are discussed in more depth in Sec. III C. In contrast, less deformed states fail to fission. For the non-fissioning case, the mask is chosen so that the initial measurement of the integrated particle density for the deformed $^{240}$Pu state contains over 239,999 particles at time $t = 0$ fm/c. We now discuss the non-fissioning scenario.

### A. Non-Fissioning States

A selection of states along the one-fragment PES (Fig. 2), starting from just beyond the static fission isomer ($\beta_{20} > 0.682$), were evolved using TDHF up to 9,000 fm/c. The time increment is chosen to be $\Delta t = 0.25$ fm/c, and 8 terms are used in the expansion of the time evolution operator \[28\]. The non-fissioning static configurations which will be presented are in the range $\beta_{20} = 0.710$ to 1.07. Due to the different masking approach used in dynamic and static calculations, the initial $t = 0$ values of the dynamic multipole deformation parameters can differ slightly from the static ones.

The time evolution of the multipole deformation parameters for the non-fissioning states are compared in Fig. 5. The evolution of the quadrupole deformation parameter is unique for each state presented, that is, the lines in the top left panel never intersect. This suggests that either the TDHF wave functions for each configuration are exploring a separate local minimum in multidimensional deformation space, or that there is an inhibition in rearranging substantially the nuclear density while keeping the total energy constant.

We now explore the distinct features in the evolution of the non-fissioning states, which arise depending on whether the initial state is deformed below or beyond the second static fission barrier.

#### 1. Evolution of States Below the Static Fission Barrier

The time evolution of the multipole deformation parameters for three states with an initial quadrupole deformation below the second static fission barrier (which peaks at $\beta_{20} = 0.86$) is shown in Fig. 6. For all cases, the initial static solutions have an almost negligible octupole deformation. Upon time evolution, the elongation of the nucleus (measured by the quadrupole deformation parameter) reduces to a certain extent. This may be seen, for example in the top left panel of Fig. 6, where the quadrupole deformation reduces from $\approx 0.71$ to $\approx 0.695$.
where $\zeta$ associated to spurious nuclear motion need to be eliminated any drifts of the central values of the multipole moments. Further, the quadrupole moments must be centred around zero. Additionally, dynamical pairing is not included in the time-dependent calculations. The initial deviation of nuclear shape could be ascribed to the fixed occupation approximation adopted in the dynamic calculations. In other words, energy is being rearranged, in which case density oscillations are expected.

Following the initial decrease in elongation, a behaviour which resembles a collective giant resonance begins. This is characterised by small-amplitude, high-frequency vibrations in the quadrupole and hexadecapole deformation (left and right columns of Fig. 6). For all cases, the octupole deformation is negligible throughout the time evolution. This is unsurprising, as the initial state displays mass symmetry between the upper and lower halves of the nucleus. As no external field is applied which breaks this symmetry, no octupole deformation will develop during the TDHF calculation [35]. In principle, however, odd and even-$l$ vibrational modes may couple in non-spherical nuclei [54, 55].

For the state with initial deformation $\beta_{20} = 0.83$ (bottom row of Fig. 6), the octupole parameter has an initial value very close to, but not identically, zero. Upon time evolution a dramatic evolution of the nuclear shape occurs (see middle panel of Fig. 5). The nuclear shape explores a slowly alternating mass asymmetry, oscillating about zero octupole deformation. The quadrupole deformation displays a drift due to the moving centre of mass of the system. This effect is not visible in the evolution of the hexadecapole deformation parameter, as the higher power terms in the definition of the parameter (proportional to $z^4$) drown it out. The initial configuration is close to the peak of the static fission barrier (see Fig. 2), and the dynamics demonstrate that mass asymmetry begins to be explored significantly at this point.

An instructive way to examine the excitation modes of a nucleus undergoing a small amplitude collective excitation is to perform Fourier analysis. This transforms the signal from the time into the frequency (and therefore energy) domain, allowing a decomposition of the dominant collective excitation modes corresponding to the oscillations of the nuclear shape. Historically, some studies of Giant Resonances have been performed by calculating a constrained Hartree-Fock minimum, and observing the behaviour upon releasing the constraint and time-evolving the wave functions [25]. In the case investigated here, we make use of the spectral power function

$$P(\omega) = |\text{Re} \zeta(\omega)|^2 + |\text{Im} \zeta(\omega)|^2,$$

where $\zeta(\omega)$ is the Fourier transform of the moment of interest.

To obtain the spectral power, the evolution of the multipole moments must be centred around zero. Further, any drifts of the central values of the multipole moments associated to spurious nuclear motion need to be eliminated. For the cases presented in Fig. 6, this is a simple procedure. We perform a linear (or a two gaussian) fit to the evolution of the deformation parameters. This fit is then subtracted from the evolution of the multipole moments [56].

A windowing technique is also used to force the signal to reduce to zero by the end of the calculation time, to suppress any artefacts [28]. The resolution of the spectra is defined by the measurement time, and is inversely proportional to the run time [57]. Some authors smooth power spectra to eliminate the fragmentation, which may be caused by boundary condition effects. This procedure may further be justified when comparing to experimental data, as detectors have a finite resolution. For the results presented, however, no such smoothing is applied, as we are interested more in comparing strength between different initial conditions than in a particular experimental result.

Figure 7 shows the resulting power spectra after the subtraction procedure is implemented. The qualitative features of the evolution of the multipole moments and their corresponding power spectra may be compared to one another. The quadrupole power spectra for the cases of $\beta_{20} = 0.71$ and 0.77 have in common a strong peak at $\omega \approx 4$ MeV (top left and middle left panels of Fig. 7, respectively). Additionally, both show a second well-defined peak. For the state with initial deformation $\beta_{20} = 0.71$, the second peak is at around 1 MeV (top left panel of Fig. 7), whereas for the evolution of the state with initial deformation $\beta_{20} = 0.77$ the second peak lies between 6-7 MeV (middle left panel of Fig. 7). For
Quadrupole and hexadecapole excitation modes.

For comparison, a benchmark calculation of the collective giant quadrupole resonance (GQR) of $^{240}$Pu has been performed, using standard methods within a TDHF framework [58]. Figure 8 shows the evolution of the deformation parameters for the isomeric state following an instantaneous, small-amplitude, isoscalar boost applied via a quadrupole field. The power spectrum is shown for direct comparison to Fig. 7. In both cases, the density remains reflection symmetric throughout the time evolution, therefore no octupole power spectrum is obtained.

For the GQR calculations built on the isomeric state, the quadrupole power spectrum has a dominant excitation mode between 4 and 5 MeV (Fig. 8, bottom right panel). This is directly comparable to the quadrupole power spectrum determined from the evolution of the deformed static states, shown in the left column of Fig. 7. The hexadecapole peak around 4 MeV is also reminiscent of the structures in the right panel of Fig. 7. The common peak in the spectra around the same energy could be interpreted as a GQR. Further, there are peaks in the spectrum built upon the excitation of the isomeric around 1, 6 and 8 MeV, corresponding to those seen in the the calculations starting from $\beta_{20} = 0.71$ and $\beta_{20} = 0.77$. The slightly different structure of the spectra starting from $\beta_{20} = 0.83$ reflects the dynamics altered by coupling to the octupole mode. This starting deformation takes the system sufficiently far away from the isomeric minimum to be out of the linear response regime.

2. Evolution of States Beyond the Static Fission Barrier

Beyond the peak of the static fission barrier, the time evolution of several increasingly deformed initial states still fails to display fission within 9000 fm/c. This is perhaps surprising, as naively one may have assumed that the static barrier corresponds to the threshold for fission. The evolution of the multipole deformation parameters for these states is presented in Fig. 9.

Qualitatively, one observes dramatically different behaviour in the time evolution of the multipole deformations for these states compared to Fig. 6. The elongation is seen to rapidly increase during the first 300 to 500 fm/c, corresponding to an increase of $\beta_{20}$. The most extreme case is seen in the bottom left panel of the Figure, where the quadrupole deformation increases from $\beta_{20} \approx 1.07$ to $\beta_{20} \approx 1.11$. This is in contrast to the evolution of states below the static fission barrier (left column of Fig. 6), where the initial quadrupole deformation was seen to decrease within the first 200 to 300 fm/c, and in the most extreme case the initial drop in $\beta_{20}$ was less than 0.02.

Beyond the initial increase in elongation, Fig. 9 displays slow, large amplitude oscillations setting in. Compared to the states below the static fission barrier (Fig. 6), these oscillations are substantially slower. They will therefore correspond to lower energy modes in a power spectrum. For the initial configurations with $\beta_{20} = 0.89$ and 0.95 (first and second rows from the top in Fig. 9), the behaviour of the quadrupole deformation is more complex than the other two cases ($\beta_{20} = 1.01$ and 1.07). The evolution of the quadrupole deformation for these cases ($\beta_{20} = 0.89$ and 0.95) shows a regions of rapid increase, then an oscillation about a plateau, then another rapid increase followed by another plateau.

An octupole deformation, depicted in the central column of Fig. 9, is observable in all cases. This is unsurprising in itself, as the initial configurations are significantly octupole deformed. However, an interesting feature is noticeable for the evolution of the states with initial deformation $\beta_{20} = 0.95$ and 1.01. The changes in octupole deformation are roughly in phase with either the evolution of the hexadecapole parameter, or both the quadrupole and hexadecapole parameters. This feature, in addition to the other differences observed, suggests that the mechanism driving the dynamics of the multipole deformations above and below the fission barrier is different.

Fourier analysis is applied here to examine the power spectra, with a subtraction procedure used to remove aperiodic motion, as in the previous section, and similar to a technique used to remove spurious motion from the study of the Giant Quadrupole Resonance [59].
FIG. 9. (Color online) Time evolution of quadrupole (left), octupole (centre) and hexadecapole (right panels) multipole parameters for initial states which are solutions to CHF calculations, with initial quadrupole deformation labelled on the right hand side. All of the initial states are deformed beyond the peak of the static fission barrier, $\beta_{20} > 0.86$.

The slow, large amplitude oscillatory behaviour of the multipole deformation parameters (second row from bottom of Fig. 9) suggests that, due to the Coulomb repulsion between the upper and lower lobes, the nucleus is attempting to fission. This is in line with the macroscopic model of Bohr and Wheeler [62], where the effect of the charge on an incompressible liquid drop is a crucial ingredient to describe the fissioning process. Within macroscopic liquid drop models, the surface term competes with the repulsive Coulomb force to inhibit fission; it costs energy to form an increasingly deformed shape. The TDHF calculations present a similar behaviour, but the mechanism arises microscopically.

Within the TDHF calculations, energy is transferred into collective motion as the nucleus attempts to rearrange into a configuration where it is able to fission (Fig. 11). Figure 12 displays the current vectors $j(r)$, corresponding to Fig. 11. The 2D vectors of the $x-z$ component of the current is a useful aid when interpreting the dynamics of the system. As the current vectors in the different panels of Fig. 12 are normalised to the same length, they may be directly compared to one another. They display a significant rearrangement of the particle density throughout the time evolution. The vectors indicate that the particle flows in the top and bottom lobes of the deformed nucleus are not moving in phase with one another, which suggests they are behaving like
FIG. 10. (Color online) Power spectra corresponding to Fig. 9. The inset panels display the magnitude of the spectra.

FIG. 11. (Color online) Slices of the total particle density for various times, starting from the static case with $\beta_{20} = 1.01$. Complex shape configurations are explored as the Coulomb force attempts to overcome the attractive nuclear force. The isolines are separated by 0.05 particles/fm$^3$. Separate, interacting, fragments rather than a single nucleus. At some points (such as $t = 8250$ fm/c, bottom
right panel), the magnitude of the current vectors is far greater, demonstrating translational motion as well as collective excitations as the nucleus rearranges.

We show in Fig. 13 the time evolution of the contribution to the total energy from the Coulomb term [28],

\[
E_C = \frac{e^2}{2} \int \int \frac{\rho_p(r) \rho_p(r')}{|r - r'|} \, dr \, dr' - \frac{3e^2}{4} \left( \frac{3}{\pi} \right)^{\frac{1}{2}} \int \rho_p^{4/3} \, dr,
\]

for the states with initial deformation \( \beta_{20} = 0.71 \) and \( \beta_{20} = 1.01 \). The former is below the static fission barrier and undergoes giant resonance-type behaviour (top panel), whereas the latter is beyond the static fission barrier (bottom panel) and displays slower and larger oscillations. Moreover, for this state the oscillation of the Coulomb energy coincides with the oscillations of the deformation parameters shown in Fig. 9. The Coulomb force thus increases the elongation, causing the total Coulomb energy to reduce as the charged lobes of the deformed nucleus move apart. The attractive terms in the energy functional then draw the nucleus back together, causing the Coulomb energy to increase once more as the elongation reduces. Due to this effect dominating over small amplitude resonances, all three deformation parameters are seen to follow large amplitude oscillations in phase with one another (second row from the bottom in Fig. 9). All in all, these results strengthen the picture that the dynamics of the nucleus in states above the fission barrier are a consequence of the competition between the repulsive Coulomb and the attractive terms in the energy density functional.

In contrast, for the case with \( \beta_{20} = 0.71 \) (top panel),...
the nuclear strong force drives the collective giant-resonance nuclear excitations. The fluctuations in the Coulomb term are small compared to an initial state beyond the fission barrier. For these giant resonance cases, the fluctuations in the Coulomb energy follow the small amplitude vibrations in the nuclear shape as the proton density rearranges. In contrast, for the cases beyond the barrier, it is the Coulomb term which is driving the large amplitude oscillations of the nuclear shape.

For illustrative purposes, the calculation evolving the state with initial \( \beta_{20} = 1.01 \) is repeated without the Coulomb interaction. The resulting evolution of the multipole deformation parameters is shown in Fig. 14. Unsurprisingly, the Figure shows a rapid decrease in elongation. The static configuration is calculated with the Coulomb interaction, the overall deformation of the nucleus “collapses” during the time evolution.

FIG. 14. (Color online) With no Coulomb interaction, the time evolution of the initial configuration \( \beta_{20} = 1.01 \) displays high-frequency, small amplitude vibrations in the quadrupole and hexadecapole degrees of freedom. As the initial static configuration is calculated with the Coulomb interaction, the overall deformation of the nucleus “collapses” during the time evolution.

Overall, there is significant evidence concerning the mechanism responsible for the slow, large amplitude oscillations of the nuclear shape observed on non-fissioning configurations beyond the static fission barrier. The oscillations are driven by a competition between the Coulomb force, trying to cause fission, and the attractive nuclear potential terms in the energy functional, countering this effect. When the states are evolved in time, they explore significant collective motion in an attempt to find a pathway towards fission. It can only be speculated as to whether, with a long enough time evolution, the states would eventually fission. The bottom left panel of Fig. 9 (evolution of a state from \( \beta_{20} = 1.07 \)) shows that the quadrupole deformation oscillates around a gradually increasing average. This suggests a final state that could eventually fission. The timescale for the quadrupole increase is very slow, however. As we have already mentioned, a TDHF calculation that explores the time evolution beyond \( t \approx 10,000 \text{ fm/c} \) may begin to encounter numerical instabilities.

B. Intersection of the One and Two-Fragment Fission Pathways

One striking characteristic of the static one- and two-fragment pathways is the intersection point that separates the initial states which fission upon time evolution from those which do not\[63\]. Figure 15 zooms into the area of the PES which is relevant for these differences. We divide the PES into three regimes. For configurations starting with a deformation below the static fission barrier (\( \beta_{20} < 0.86 \)), tunnelling is required to reach a fis-
sioned state. This is forbidden in TDHF calculations, in which collective coordinates behave semiclassically. We therefore define a forbidding region below the barrier. Beyond the barrier, in contrast, there is a region where fission is inhibited. It will shortly be demonstrated in Sec. III C that, beyond the intersection of the one and two-fragment pathways, fission is allowed within the considered time scales of the TDHF calculations. The line between \( \beta_{20} = 1.07 \) and 1.10 thus separates the inhibiting and allowed regions for fission, but it does not correspond to a threshold in the dynamic calculations. The bottom row of Fig. 9 indicates, for example, that the state with an initial deformation just below the separating line can evolve dynamically to a state with deformations beyond this very same line, but without fissioning.

An intuitive explanation may be given for the significance of the intersection point of the pathways on the PES with regard to fission occurring upon time evolution. In TDHF, the total energy is conserved. It is because of the inclusion of excitations (internal and translational) that nuclear configurations may change upon time evolution. For the states which undergo fission (allowed region in Figure 15, \( \beta_{20} > 1.085 \)), for a given value of \( \beta_{20} \), the two-fragment state is more bound than the one-fragment state. In consequence, the one-fragment state can evolve into a two-fragment configuration at a constant \( \beta_{20} \) by releasing nuclear potential energy into excitation energy. Of course, the picture is not really that simple, as the significance of the static PES becomes less clear in the dynamic case. In general, configuration which do not correspond to the static fission pathways will be explored. Further, a slight change of configuration will be required to move from the static one-fragment state to a fissioned configuration. In other words, the exact configurations on the two-fragment pathway cannot be reached dynamically from the one-fragment pathway, but an excited two-fragment configuration of a similar deformation can. The reasoning presented is that, as the static two-fragment state is more bound than the corresponding one-fragment state, the optimum TDHF trajectory is to evolve the one-fragment static state towards an excited fissioned configuration. By undergoing only a modest rearrangement of the nuclear shape.

In contrast, in the inhibited region of Fig. 15 (0.86 \( \leq \beta_{20} \leq 1.085 \)), for a given \( \beta_{20} \) in the one-fragment pathway, the two-fragment state with the same \( \beta_{20} \) is less bound. Due to energy conservation, the one-fragment state cannot move to the two-fragment state at the same \( \beta_{20} \). The only way to reach a two-fragment solution of equal binding energy (or an excited configuration with greater binding energy) is through a significant change in deformation and rearrangement of the nuclear state, which accounts for the inhibiting time scale for fission to occur. It may be that the presented calculations lack the degrees of freedom necessary to allow a fissioning path to be found in this window at all, and that a method beyond basic TDHF [34, 64] is needed to reach the fissioned configuration. Further investigation of the link between static and dynamic configurations applying Density Constrained TDHF [33, 65], would certainly be of interest. This method allows the dynamic configurations to be “frozen”, removing internal and collective excitations, thus bridging between static and dynamic configurations.

C. Fissioning States

For the static states with a quadrupole deformation at and beyond the threshold of \( \beta_{20} = 1.10 \), binary fission is seen to occur as the wave functions are evolved in time. The calculations to obtain the data for this Section were performed in a larger grid of size \( 48 \times 48 \times 160 \) points, corresponding to \( -79.5 \) to 79.5 fm in the \( z \) direction, and \(-23.5 \) to \( 23.5 \) fm in the \( x \) and \( y \) directions. The calculations were set to end once the separation of the centre of mass of the two fragments exceeded 100 fm. This cutoff avoids spurious effects associated to the fragments approaching the grid boundaries.

Figure 16 shows the typical time evolution of the particle density for the fissioning case by presenting 2D slices of the 3D density at various times for the state with initial deformation \( \beta_{20} = 1.19 \). It is difficult to establish exactly the scission point in a calculation involving quantum mechanical wave functions and densities. We take an operational approach and define “scission” as the point when the minimum density between the fragments along the principal axis of the system is less than 0.05 particles/fm\(^3\). As we shall see in the following, this also corresponds to the point where a sizeable collective energy develops as the fission products begin spatially separating. For the case where the initial quadrupole deformation is \( \beta_{20} = 1.19 \) (presented in Fig. 16), it takes between 775-800 fm/c for the density between the two fragments to drop below this threshold. Figure ?? displays sample current vectors corresponding to the particle density slices of Fig. 16. The current vectors display the system smoothly transitioning into a two-fragment configuration. Compared to the situation of the one-fragment pathway state in Fig. 12, there is no dramatic rearrangement of the density during time evolution. Throughout the calculation, the currents in the two preformed fragments are clearly distinguishable, and do not interact with one another. The central region has negligible current, and the two lobes stretch against each other. The magnitude of the current vectors in Fig. ?? gradually increases as the fission occurs. Beyond the point of scission, they increase rapidly as the fragments accelerate away from one another.

States with increasingly large initial deformations fission in a qualitatively similar way. We now proceed to compare their fission outcomes in terms of macroscopic observables. The states with static deformation \( \beta_{20} = 1.10, 1.13, 1.19 \) and 1.25 were evolved in time to investigate the fission of the different initial configurations. The time evolution of the multipole moments for these states are shown in Fig. 18. Different nuclear shapes are
FIG. 16. (Color online) Slices of the total particle density for various times, starting from the static case with $\beta_{20} = 1.19$. It takes between 775 and 800 fm/c for scission (as defined in the text) to occur. The isolines are separated by $0.05$ particles/fm$^3$.

explored as the nucleus evolves from the various static states. Other than the case with initial $\beta_{20} = 1.25$, we find that as $\beta_{20}$ and $\beta_{40}$ increase, the octupole deformation, as reflected in $\beta_{30}$, remains virtually constant. Here and in the following discussion, we sharply cut off the time evolution at the point of scission. An analysis of the post-scission fragments is presented in Sec. IV.

The chosen dynamic pathway towards fission, depending on how the particles rearrange during the time evolution, may have significant consequences upon the properties of the post-fission system. This will produce a range of fission fragments dependent on the initial configuration which is time evolved. Once again, this differs from the static case, where CHF calculations following the one-fragment fission pathway will only produce one resulting fissioned configuration. The distribution of fission products obtained with TDHF is in line with experimental investigations (see Sec. IV).

The time scale required for the initial configuration to fission varies. The least elongated case, with $\beta_{20} = 1.10$ takes $t_{\text{scission}} \approx 1250$ fm/c to scission to occur. Figure 18 shows the quadrupole deformation increasing rapidly for approximately 600 fm/c (solid line in the Figure). Between 600-1200 fm/c, the rate of increase in quadrupole deformation reduces as the nucleons rearrange out of the neck into the upper and lower fragments. Small oscillations in the octupole deformation can be seen as the system transitions into the preferred configuration. Beyond 1200 fm/c, the neck rapidly vanishes as the fragments take form and begin to separate, resulting in an acceleration in the increase of the quadrupole and hexadecapole parameters.

For more deformed initial states, the time taken to fission is significantly shorter. This can be explained as the initial configuration has fewer particles in the neck region. Upon time evolution, less rearrangement is required for the two fragments to take form, and the Coulomb interaction rapidly drives the configuration to fission. The most extreme case investigated here is that with initial deformation $\beta_{20} = 1.25$. The bottom right panel of Fig. 2 shows the initial density for this state. Two fragments are already taking form, connected only by a thin elongated neck. This narrow structure rapidly dissipates into the top and bottom fragments upon time evolution, and within $t_{\text{scission}} \approx 400$ fm/c the system fissions.

The evolution of the decomposed contributions to the energy density functional for the fissioning cases are presented in Fig. 19. The decomposed energy density functional for the entire two-fragment system is shown. The total energy of the two separate fission fragments will be analysed separately in Sec. IV. Figure 19 displays the evolution of the energy density functional up to and beyond the point of scission. The total energy, shown on the bottom panel of Fig. 19, is in principle conserved within the TDHF calculations. For these fission calculations, the fluctuations during the time evolution are less than 4 MeV.

The dynamic calculations allow for both translational motion and internal excitations. In the fissioning case, the nuclear binding energy is expected to be transformed...
mainly into the translational kinetic energy of the fragments. The nuclear collective kinetic energy is conventionally defined within TDHF as,

$$E_{\text{coll. kin.}} = \frac{\hbar^2}{2m} \int dr \left( \frac{j(r)^2}{\rho(r)} \right), \quad (5)$$

where \(\rho(r)\) is the particle density and \(j(r)\) is the current density [28]. This collective kinetic energy contains contributions from internal excitations of the nucleus, such as resonances, as well as the translational kinetic energy of the post-fission fragments. It is presented in the second panel from the bottom, right column of Fig. 19. The total kinetic energy is shown separately, in the adjacent panel. It is difficult to untangle the collective excitation energy attributed to the internal excitation of the fission fragments, from that attributed to translational motion. In fission reactions, the energy release is typically attributed to be \(\approx 80\%\) in the form of translational energy, and the other \(\approx 20\%\) is released in the form of \(\gamma\) rays, prompt neutron emission and radioactive decays of the fragments [66]. In our TDHF calculations, only part of these effects can be described. The degrees of freedom to allow fission fragments in hot resonance states to decay by particle emission, for instance, are included. We will demonstrate that the excitation energy of the fissioned system in the TDHF simulations is dominated by the translational kinetic energy, with a small contribution from internal collective excitation of the fragments. This will be discussed in Section IV C.

During the time evolution, the individual components of the energy functional may be examined separately. The physical interpretation of the evolution of each term of the integrated energy functional shown in Fig. 19 may not necessarily be simple. It is useful to identify which densities contribute to the separate terms to qualitatively explain the behaviour of the energy functional:

- **\(E_0\) and \(E_3\) terms:** These terms, proportional to the Skyrme parameters \(t_0\) and \(t_3\), are the central terms of the functional. They are attractive and repulsive respectively, providing a similar functional form with an expected cancelation between them. They depend upon the particle density. As the nucleus approaches scission, the \(E_0\) term is reduced in strength, thanks to the small, unfavourable density that briefly exists in the neck region. There is then a sudden increase in binding as separate, more stable fragments are formed. After this, the energy contributions oscillate with a smaller magnitude than the changes during the fission process, corresponding to the excited collective motion of the fragments.
• $E_1$ term: The $E_1$ term contains contributions from the kinetic, particle, and current densities. The time evolution of this term is qualitatively similar to the $E_0$ and $E_3$ contributions, with the same sign as the $E_3$ term but an absolute smaller scale. This suggests the that density that governs the $E_0$ and $E_3$ terms, the particle density, is also the most relevant contribution driving the $E_1$ term.

• $E_2$ term: The $E_2$ contribution contains the Laplacian of the particle density, and is commonly associated with a surface term. As the particles rearrange into the two fission fragments, this term increases in magnitude - i.e. gives an overall more repulsive contribution to the entire system. This can be explained by the two-fragment system having a combined surface fraction which is greater than that of the initial configuration. The gain in energy for this term up to the point of scission is dependent upon the deformation of the initial configuration. It can increase by as much as 45 MeV for the static configuration with $\beta_{20} = 1.10$.

• Coulomb term: The Coulomb energy is determined from the distribution of the charged protons. The magnitude of the repulsive Coulomb term slowly decreases as the nucleus elongates. The overall reduction is of the order of 200 MeV as the system evolves. At the point of scission, the rate at which the term reduces rapidly accelerates, as the two charged fragments separate from one another in coordinate space. At infinite fragment separation, the Coulomb term will reduce to the contributions of the Coulomb energy for each nucleus, without further interactions.

• Kinetic and Collective Kinetic terms: The kinetic energy can be determined from integrating the kinetic density. As mentioned above, the contribution to this energy from collective motion (assumed to be predominantly translational beyond scission, rather than internal collective excitation) can be decomposed according to Eq. (5). The collective energy is initially small, as it is only associated to the internal currents as the nucleus slowly rearranges into a fissioned configuration. The state with initial deformation $\beta_{20} = 1.10$ shows the most gradual transition to fission. An initial rapid increase and a saturation in the collective energy is seen before scission, which corresponds to the (previously discussed) rapid initial elongation, then extended rearrangement phase as the configuration evolves (see Fig. 18). In contrast, the state with $\beta_{20} = 1.25$ is already close to the point of scission,

![FIG. 18. (Color online) Time evolution of quadrupole (top), octupole (middle) and hexadecapole (bottom panel) deformation parameters for various static configurations observed to fission upon time evolution. The evolution is stopped at the scission point, as defined in the text.

![FIG. 19. Evolution of the decomposed energy density functional for the fissioning systems. The calculations are terminated when the fragments are separated by 100 fm. For reference, the vertical lines in the panels corresponding to the kinetic energies show the corresponding scission points. The total energy is conserved within fluctuations no greater than 4 MeV.](image)
so that the Coulomb interaction between the two lobes rapidly drives the configuration to the scission point (within the first few hundred fm/c), where translational motion rapidly accelerates once the neck ruptures. This shorter timescale could explain the more extreme behaviour observed in the evolution of the other terms in the energy functional as the particles in the neck have less time to rearrange into the two fragments. At the point of scission, the collective kinetic energy rapidly increases at a similar rate to the reduction in the Coulomb energy. The threshold collective kinetic energy associated with the scission point is between 6 and 8 MeV in all the cases presented. The vertical lines corresponding to the scission points are displayed in the panels corresponding to the kinetic and collective kinetic energies in Fig. 19. The definition adopted for scission [67] is justified by considering the rapid increase of collective kinetic energy at this point. The gain in the total kinetic energy beyond the scission point can be attributed to the gain in collective energy, and is of the order of 150 MeV in the time considered. This relates to the loss in Coulomb energy beyond the point of scission, as would be expected.

- Spin-Orbit term: The spin-orbit potential is important for the single-particle structure. Spin-orbit partner levels combine, when fully occupied, to give a total zero energy contribution, so the contributions reflect the changing of the single particle levels and the change of meaning of spin-orbit partners in the parent nucleus to the daughter nuclei. One therefore expects this contribution to be relevant in determining the structural details of the final fission fragments. The final approximately constant values observed following scission correspond to the sum of the two independent spin-orbit terms of the separate fragments. The notably different behaviour of the term for the state with initial deformation $\beta_{20} = 1.25$, compared to the others, suggests that different shell effects are acting. Indeed, the masses of the fission products are significantly different for this case (see Sec. IV A). The spin-orbit term also has significant contributions from time-odd densities, so that the evolution can explore configurations which may not be accessible on the static PES. Overall, the term varies by less than 30 MeV during time evolution.

Since increasing the dimensions of the numerical grid is extremely computationally expensive, we take an alternative perspective. We extrapolate the collective kinetic energies to large times to estimate the asymptotic value of the collective energy as the separation $r$ tends to $\infty$.

A simple approximation to the time-dependent behaviour can be made from classical mechanics. Let us assume two point-like fragments with charges $Z_u$ and $Z_l$, and masses $M_u$ and $M_l$. If the two fragments fission from a ground state because of the Coulomb force, and convert all this energy into translational kinetic energy, energy conservation implies:

$$\frac{1}{2} M_u v_u^2 + \frac{1}{2} M_l v_l^2 = \frac{Z_u Z_l}{r}.$$  \hspace{1cm} (6)

Here, $M_i$, $v_i$, and $Z_i$ are the mass, velocity and charge of each fragment ($i = u$ for upper and $i = l$ for lower fragment). The constant $\kappa$ is the Coulomb constant. As momentum must be conserved,

$$M_u v_u + M_l v_l = 0,$$  \hspace{1cm} (7)

Eq. (6) may be rewritten, substituting for $v_u$

$$v_l^2 \left( \frac{M_l^2}{M_u} + M_l \right) = 2 \kappa \frac{Z_u Z_l}{r}.$$  \hspace{1cm} (8)

For a given fissioned system, $M_u$, $M_l$, $Z_u$ and $Z_l$ are constant. A differential equation for $\frac{dv}{dt} (= v_l)$ can be formed

$$\frac{dr}{dt} = \sqrt{\Theta \over r},$$  \hspace{1cm} (9)

where all the constants are combined into $\Theta$. Performing the integration

$$\int_{r_0}^r r^{1/2} dr = \int_{t_0}^t \sqrt{\Theta} dt$$  \hspace{1cm} (10)

allows the solution

$$r^{3/2} = r_0^{3/2} + \frac{3}{2} \sqrt{\Theta}(t - t_0)$$  \hspace{1cm} (11)

to be written. According to this approximation, the distance between the two fission fragments, $r$, is approximately proportional to $t^{2/3}$. By assuming that the loss in Coulomb energy is equal to the gain in collective kinetic energy (that is , $E_{\text{Coul}} = E_{\text{coll. kin.}}$), a fit of the form

$$f(t) = a + \frac{b}{(t - c)^{3/2}}$$  \hspace{1cm} (12)

can be performed to interpolate the collective kinetic energy to asymptotically large values of $t$. Figure 20 shows a sample interpolation of the collective kinetic energy assuming the above form for the case of initial deformation $\beta_{20} = 1.10$. The fit is performed over three time ranges: once the centres of mass are separated beyond 30 fm, 50
FIG. 20. Fits to the obtained collective kinetic energy for the initial state with deformation $\beta_{20} = 1.10$. Fits are performed over three different ranges: from the point where the separation of the fragments exceeds 30 fm, 50 fm and 60 fm, respectively.

TABLE III. Interpolated total kinetic energy corresponding to different initial configurations. The fit of Eq. (12) has been performed once the fragment separation exceeds 30 fm, 50 fm and 60 fm.

| Static Deformation $\beta_{20}$ | Coll. KE (30 fm fit) [MeV] | Coll. KE (50 fm fit) [MeV] | Coll. KE (60 fm fit) [MeV] | Mean ± St. Dev. [MeV] |
|--------------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------|
| 1.10                           | 210.3                       | 206.5                       | 203.4                       | 206(4)                |
| 1.13                           | 210.8                       | 200.0                       | 193.7                       | 202(8)                |
| 1.19                           | 205.8                       | 196.8                       | 191.3                       | 198(9)                |
| 1.25                           | 193.4                       | 180.8                       | 176.3                       | 183(9)                |

fm and 60 fm, respectively. As the separation tends to $\infty$, the fit parameter $a$ can be interpreted as the final collective kinetic energy. Table III contains the values obtained for each of the fissioning cases with different distance fits. As a crude method to represent the uncertainty in the final collective energy value, we present the mean and the standard deviation of the three interpolation fits.

The values shown in Table III demonstrate that the resulting collective kinetic energy varies by about 10% depending upon the region of the data the fit is performed to. In this very crude model, fragment deformation, particle emission (discussed in the next Section) or tidal effects associated to the extended nature of the nuclei are not accounted for. This suggests that the results obtained from the interpolation method should serve only as illustrative values.

In comparison to the experimentally measured kinetic energy of the fissioning systems displayed in Table IV, the theoretical values presented in Table III are of a similar order of magnitude. The crude theoretical estimates are about 5 to 25 MeV larger in all cases. We note, however, that the experimental values correspond to an average kinetic energy. We only have access to a single fissioning event per static state, and a larger sample of theoretical results would be required to enable a quantitative comparison. Further discussion of methods to deduce the energy released by the fission reaction within TDHF will be presented in Sec. IV B.

TABLE IV. Measured total kinetic energies from various experiments. The measurements correspond to the pre-neutron emission fragment energies.

| Method                  | Kinetic energy [MeV] | Reference |
|-------------------------|----------------------|-----------|
| $^{240}$Pu(s.f)         | 178.85±0.30          | [2]       |
| $^{240}$Pu(s.f)         | 179.00±0.08          | [3]       |
| $^{239}$Pu($n_{th}$, f) | 177.69               | [2]       |
| $^{239}$Pu($n_{th}$, f) | 177.65±0.01          | [3]       |
| $^{240}$Pu($\gamma$, f) (12 MeV) | 176.39±0.24 | [2]       |
| $^{240}$Pu($\gamma$, f) (15 MeV) | 175.80±0.24 | [2]       |
| $^{240}$Pu($\gamma$, f) (20 MeV) | 175.15±0.24 | [2]       |
| $^{240}$Pu($\gamma$, f) (30 MeV) | 174.98±0.31 | [2]       |

IV. FRAGMENT ANALYSIS

Beyond the point of scission, we consider a two-fragment system. The published distribution of Sky3D has some capacity to analyse two-fragment dynamics [28], and a version has been modified further to investigate the fissioning system and extract some useful observables. The masking procedure used to investigate the non-fissioning dynamic case can be generalised to investigate the fissioning system. Details are provided in Appendix B.

A. Mass Distributions

As the post-fission fragments are excited, they may decay by particle emission. TDHF displays this decay by the spreading of the single particle wave functions from the region of central density (corresponding to the nucleus). When masking the region around the nucleus, this decay results in a reduction in the integrated particle density over time. For the cases of deformation-induced fission (DIF) examined in this paper, this decay is of the order of 0.1-0.2 particles during the whole post-scission time evolution.

In order to compare to experimental studies, we identify the number of particles in each fragment prior to any particle emission. This is done by integrating the total density in each half of the numerical grid separated by the dividing plane immediately after the scission point. The integral is performed without any masking. An uncertainty in the particle number of the fragments may
TABLE V. Fission fragments obtained from evolving initial static configurations from the one-fragment fission pathway. The uncertainties in the particle numbers are a conservative estimate related to the fluctuation in the particle number in the region of the grid corresponding to the separate fragments throughout time evolution. The result from the static two-fragment pathway for $\beta_{20} = 1.19$ is included for comparison.

| Static Deformation $\beta_{20}$ | Heavy Fragment $A,Z$ | Light Fragment $A,Z$ | Heavy Frag. (Integer) | Light Frag. (Integer) |
|----------------------------------|----------------------|----------------------|-----------------------|-----------------------|
| 1.10                             | 136.33(5), 52.78(5)  | 103.67(5), 41.23(5)  | $^{136}\text{I}$, $^{53}\text{I}$ | $^{104}\text{Nb}$ |
| 1.13                             | 135.02(5), 52.23(5)  | 104.98(5), 41.77(5)  | $^{135}\text{Te}$, $^{52}\text{Te}$ | $^{105}\text{Mo}$ |
| 1.19                             | 136.13(5), 52.70(5)  | 103.87(5), 41.30(5)  | $^{136}\text{I}$, $^{53}\text{I}$ | $^{104}\text{Nb}$ |
| 1.25                             | 143.70(5), 55.65(5)  | 96.30(5), 38.35(5)   | $^{134}\text{Cs}$, $^{55}\text{Cs}$ | $^{96}\text{Sr}$ |
| 1.19(2f)                         | 132.81, 50.84        | 107.04, 43.13        | $^{133}\text{Sb}$, $^{51}\text{Sb}$ | $^{107}\text{Tc}$ |

be associated with the fluctuation of this measurement throughout time evolution, which is less than 0.05 particles for all the considered cases. We are thus confident that the fission fragments that we provide have a good average mass number.

These fragment masses can be compared directly to experimental data. Table V displays the resulting mass distributions obtained from this theoretical study. The two-fragment static configuration is included for comparison. We note that this differs from the deformation-induced fragments. It produces more symmetric states by about 2 units of mass. The Table also includes the particle number rounded to the nearest integer. It would be of interest to project the individual fragments onto the particle number [49], to obtain a mass number distribution out of these results.

Further, Table VI contains experimental data taken from Refs. [2, 3], listing the most likely masses of the fission fragments. The references study various fission processes in $^{240}\text{Pu}$, including spontaneous fission, thermal neutron-induced fission, and various energy photon-induced fission. The spontaneous fission data has been included for completeness.

We emphasise that fission produces a range of masses. The values quoted in Table VI correspond only to the most likely fissioned configuration. This is a crude comparison of experimental data to theoretical calculations. A more meaningful comparison would go through a characterization of the full mass distribution, in line with the recently proposed method of Ref. [37]. While we cannot at present produce a full mass distribution from projection into particle numbers, we can use each of the 4 different fission cases to build a schematic fission fragment mass distribution. Referring to Fig. 21, which displays data for neutron-induced fission, the obtained theoretical values fall well within the experimentally obtained mass distribution. The different distributions shown in the Figure are for increasing energy in neutron-induced fission. As the energy increases, the experimental distribution of the fission fragments becomes, on average, more symmetric. For instance, the central region of the mass distribution on the bottom panel of Fig. 21 fills in. We note that the theoretical data has been binned and normalised to the maximum value of each experimental data set, so that visual comparisons may be drawn. With the limited data set available, the TDHF results seem to be consistent with the experimental data. Figure 21 also includes for comparison the adiabatic, constrained static result of the two-fission pathway.
TABLE VI. Experimentally measured average masses following the fission $^{240}$Pu. The measurements for neutron-induced fission were taken before neutron emission of the fissioned fragments.

| Method          | Heavy Fragment | Light Fragment | Reference |
|-----------------|----------------|----------------|----------|
| $^{240}$Pu(s.f) | 138.74±0.20    | 101.26±0.20    | [2]      |
| $^{240}$Pu(s.f) | 138.96±0.04    | 101.31±0.04    | [3]      |
| $^{238}$Pu(n,t, f) | 139.67 | 100.33 | [2] |
| $^{238}$Pu(n,t, f) | 139.73±0.01 | 100.27±0.01 | [3] |
| $^{240}$Pu(γ, f) (12 MeV) | 139.88±0.14 | 100.12±0.14 | [2] |
| $^{240}$Pu(γ, f) (15 MeV) | 139.92±0.09 | 100.08±0.09 | [2] |
| $^{240}$Pu(γ, f) (20 MeV) | 139.84±0.08 | 100.16±0.08 | [2] |
| $^{240}$Pu(γ, f) (30 MeV) | 139.71±0.14 | 100.29±0.14 | [2] |

B. Energy of Fission Fragments

By applying masks around the spatial regions of the fission fragments, the energy density functional corresponding to each individual fragment may be obtained. We note, however, that the interpretation of the results is not simple in the two-fragment case. The nuclear part of the energy density functional is short-ranged. We therefore expect that an integral in the spatial region corresponding to the individual fragments will be a faithful representation of the nuclear part of the energy density. The Coulomb interaction, however, is long-ranged. As well as the Coulomb interaction within the individual fragments, there is a contribution from the interaction with one another. This contribution is missing in the present two-fragment integral. Further, the fragments decay by particle emission, which imparts some time dependence upon the integrated energy corresponding to the individual fragments.

The time evolution of the total integrated energy functional corresponding to the heavy (upper) and light (lower) fission fragments is shown in Fig. 22. The time measurement at $t_{\text{sep}} = 0$ begins when the fragments are sufficiently separated such that the masks no longer overlap (see Fig. 33). To a good approximation, the fragment total energies are constant over time. A slight drift is observed in the time evolution, due to the long-range effects of the Coulomb interaction as well as particle decay. We only provide the total integrated energy of the post-fission fragments in Fig. 22. The evolution of the decomposed terms corresponding to the individual fragments shows no remarkable behaviour. Hereafter, the fragment energy at the cutoff time will be denoted as $E^*$. The time evolution of the total integrated energy functional corresponding to the heavy (upper) and light (lower) fission fragments is shown in Fig. 22. The time measurement at $t_{\text{sep}} = 0$ begins when the fragments are sufficiently separated such that the masks no longer overlap (see Fig. 33). To a good approximation, the fragment total energies are constant over time. A slight drift is observed in the time evolution, due to the long-range effects of the Coulomb interaction as well as particle decay. We only provide the total integrated energy of the post-fission fragments in Fig. 22. The evolution of the decomposed terms corresponding to the individual fragments shows no remarkable behaviour. Hereafter, the fragment energy at the cutoff time will be denoted as $E^*$. The time evolution of the total integrated energy functional corresponding to the heavy (upper) and light (lower) fission fragments is shown in Fig. 22. The time measurement at $t_{\text{sep}} = 0$ begins when the fragments are sufficiently separated such that the masks no longer overlap (see Fig. 33). To a good approximation, the fragment total energies are constant over time. A slight drift is observed in the time evolution, due to the long-range effects of the Coulomb interaction as well as particle decay. We only provide the total integrated energy of the post-fission fragments in Fig. 22. The evolution of the decomposed terms corresponding to the individual fragments shows no remarkable behaviour. Hereafter, the fragment energy at the cutoff time will be denoted as $E^*$.

The total energies of the two fragments provide an alternative way of computing their collective kinetic energies. The total excitation energies of the fragments is the sum of their translational and internal collective kinetic energies. If we subtract the total integrated energy to the corresponding ground state energy, we obtain a new estimate for the fragment excitation energy. This method complements the approach of interpolating the total collective kinetic energy of the system, as presented in Sec. III C. As long as the ground state and the fragment energies are qualitatively correct, this second method should produce comparable results.

The solver Sky3D has thus been applied to deduce the ground states of the fission fragments to the nearest integer particle number. Here, it is debatable whether the energy functional in Sky3D contains all the terms required to calculate odd-odd and odd-even nuclei. The full time-odd contribution is presented in Ref. [69], and the functional in Sky3D does not include all these terms. However, as the functional used for the static calculations is consistent with that applied to dynamic calculations, Galilean invariance is conserved. The functional used in Sky3D therefore satisfies all the invariance properties required to perform static calculations of odd-odd and odd-even nuclei, even if the functional is not in its most...
“complete” form. We also note that pairing has been neglected in these calculations. The total pairing contribution to the energy functional is typically of the order of 0 – 10 MeV, which is small compared to the excitation energies in the fissioning case.

We have access to two measurements of the total excitation energy of the systems, either by interpolating the evolution of the collective kinetic energy, or by comparing the ground state fragment energies to the excited fragment energies. Figure 23 displays the mean interpolated collective kinetic energies (circles) presented in Table III. We compare these to the total fragment excitation energy ($\Delta E_{\text{heavy frag.}} + \Delta E_{\text{light frag.}}$) for each fissioning case (triangles). The error bars in the values of $\Delta E$ display an uncertainty of 10 MeV, a conservative estimate for typical pairing correlations, possible deformation energies, and also that only nearest-integer nuclei are considered. For the interpolated collective energy, the values presented are the mean of the three interpolations performed at different fragment separations. We present a preliminary, rough estimate of the error of this calculation, obtained from the standard deviation of these 3 data points (see Table III).

Figure 23 shows that within the error bars, the results from the two techniques produce consistent values of the energy released in the fission process. A smaller error for both predictions could be achieved by performing the calculations up to the point where the Coulomb interaction is negligible, and obtaining ground state energies incorporating pairing correlations. As mentioned, the energies presented here are measures of the total excitation energy of the system. Section IV C will demonstrate a technique which may be used to decouple the translational kinetic energy from the internal collective excitation energy of the two separate fragments. These will provide further insight into the excitation mechanisms in TDHF fragment formation.

Figure 24 shows experimental measurements of the kinetic energy reproduced from Ref. [3] for thermal neutron-induced fission in $^{240}$Pu. The range of collective kinetic energies deduced in this Section are marked with a shaded box. By attributing the deduced total excitation energies solely to translational kinetic energy, this assumes that the internal collective excitation of the fragments are comparatively small. This will be demonstrated shortly. Despite the limited sample of theoretical data, the results agree with the experimental range of values. In particular, our results fall well within the experimental distribution.

C. Collective Excitation Modes of Fission Fragments

As mentioned, the excitation energy of the fission fragments is assumed to be dominated by translational kinetic energy. However, as well as translational motion, the fragments undergo collective vibrations because of internal excitation. The collective excitation modes of the fragments may be investigated using the same procedure which was applied to the excited non-fissioning nuclei, as discussed in Sec. III A. Unfortunately, due to the limitations in the numerical grid size (the $z$ direction spanned 160 grid points, ranging from -79.5 to 79.5 fm), a signal corresponding to the evolution of the multipole parameters of the individual fragments could only be measured for approximately 1000 fm/c before the grid boundaries were approached. For a signal of this length, the resolution of the calculated power spectrum is of the order $\hbar \omega = \frac{\pi}{T_{\text{obs}}} \approx 0.6$ MeV [57].

To extend the time evolution domain and consequently improve energy resolution, we adopted the following approach. Rather than performing the calculations in an unpractically large numerical grid, a Galilean transfor-
TABLE VII. Comparison of the fission fragment energies to the ground state energy calculated using the SkM* interaction. The fragment total energy at the cutoff time is denoted by $E^*$ (see Fig. 22), the ground state energy by $E_{gs}$, and the difference ($E^* - E_{gs}$) by $\Delta E$. See text for more details.

| $\beta_{20}$ | Heavy Frag. | $E^*$ [MeV] | $E_{gs}$ [MeV] | $\Delta E$ | Light Frag. | $E^*$ [MeV] | $E_{gs}$ [MeV] | $\Delta E$ |
|-------------|-------------|-------------|---------------|-----------|-------------|-------------|---------------|-----------|
| 1.10        | $^{196}$I   | -1029.22    | -1118.31      | 89.09     | $^{104}$Nb | -747.86     | -854.54       | 106.68    |
| 1.13        | $^{132}$Te | -1023.81    | -1110.24      | 86.43     | $^{105}$Mo | -757.38     | -865.71       | 108.33    |
| 1.19        | $^{190}$I   | -1034.23    | -1118.31      | 84.04     | $^{103}$Nb | -749.41     | -854.54       | 105.13    |
| 1.25        | $^{144}$Cs  | -1090.17    | -1162.47      | 72.30     | $^{96}$Sr  | -697.78     | -796.94       | 99.16     |

...This momentum is applied to each fission fragment. The boost momentum is chosen to cancel the corresponding linear momenta of each fragment. After boosting, the two fragments remain approximately still in the box, and their excitation modes can be studied for a much longer period. The evolution of the configuration with an initial $\beta_{20} = 1.25$ is presented as an example of this new method.

Inside the masked regions of space corresponding to the fragments, the linear momentum may be calculated by integrating the current density:

$$p_{\text{frag}} = \int dr \; j(r). \quad (13)$$

This momentum therefore has units of velocity [28]. The linear momentum of the fragments may then be instantaneously removed by applying the following Galilean boost to the single particle wave functions:

$$\varphi(r) = \exp \left( i \frac{p_{\text{frag}} \cdot r}{A_{\text{frag}}} \right) \varphi(r), \quad (14)$$

where $A_{\text{frag}}$ is the integrated particle density corresponding to the fragment, and $\varphi(r)$ are the single-particle wave functions. The Galilean transformation should be applied in the masked region of space with the corresponding momentum for each fragment. The effect of the transformation is to effectively boost the particles in the opposite direction with the exact momentum they are propagating with through the grid.

Figure 25 shows the decomposed energy functional for the fissioning case with initial $\beta_{20} = 1.25$. The Galilean transform was applied when the separation of the fragments reached 100 fm (corresponding to $t \approx 1900$ fm/c), and the calculation terminated at separation 105 fm. Upon application of the transformation, the (bottom) panel corresponding to the total energy displays a decrease of approximately 140 MeV. This mirrors the drop in collective kinetic energy by the same amount, which corresponds to an instantaneous removal of excitation energy due to translational motion. This demonstrates that the total excitation energy is dominated by contributions from translational motion, rather than internal collective excitations. As for the other energy contributions, the nuclear and Coulomb energies remain unaltered before and after the boost is applied around $t \approx 1900$ fm/c.

FIG. 25. (Color online) Decomposed contributions to the energy of the system for the case with initial deformation $\beta_{20} = 1.25$. A Galilean transformation has been applied to remove the linear momentum of the individual fragments once the separation between the fragment centre of mass reaches 100 fm. The calculation is terminated once the separation exceeds 105 fm.

This is to be expected, as these terms are all Galilean invariant.

The collective kinetic energy drops instantaneously to $\approx 1.1$ MeV following the transformation (see inset panel corresponding to collective kinetic energy in Fig. 25). As the translational energy has been removed at this point, the remaining collective energy can be interpreted as the sum of the internal excitation energy shared between the two fragments. Reference [18] discusses an alternative method to deduce the internal collective excitation energy of the fragments, but the method applied assumes a priori knowledge of the fission products of the system.

The internal collective excitation energy is very small.
compared to the total excitation energy released in the fission process, which is \( \approx 180 \text{ MeV} \) (see Fig. 23, \( \beta_{20} = 1.25 \)). This justifies the previous assumption that the final collective excitation energy deduced in TDHF is dominantly translational kinetic energy, so it may therefore be compared to the experimentally measured kinetic energies (Fig. 24). Further, the energy functional contributions (Fig. 25) may be compared to these in Fig. 19, where the calculation was terminated at the point where the transformation is applied in this case. Figure 25 demonstrates that the nuclear potential part of the energy functional is unaffected by the transformation. The calculation was performed in a grid of identical dimensions to those presented in Fig. 19 (\( 48 \times 48 \times 160 \) points), and the time elapsed has effectively doubled from those previous calculations. As the measurement time of the post-fission fragments has been elongated, the resolution of the resulting power spectra will be enhanced accordingly.

We note that the Coulomb interaction is long-ranged, so that even at a separation of 100 fm there is an interaction between the fragments. Translational motion resumes after the Galilean transformation is applied, and the translational kinetic energy slowly increases. This can be seen by the gradual increase of the collective kinetic energy in Fig. 25 following the transformation. Therefore, the centre of mass separation eventually reaches 105 fm and the calculation is terminated. We note that, in principle, one could re-apply the Galilean transformation at every iteration to extend the total time and improve energy resolution.

The evolution of the multipole deformation parameters for the two fragments are shown in Figs. 26 and 27, corresponding to the heavy and light fragment, respectively. The corresponding power spectra are shown in Figs. 28 and 29. Let us stress that the resolution of the spectra is \( \approx 0.4 \text{ MeV} \), a significant improvement with respect to that would be obtained without extending the measurement time.

The evolution of the multipole fragments is qualitatively similar for both fragments. The quadrupoles oscillations are centered around values of \( \beta_{20} \approx 0.18 \) and 0.35 for the heavy and light fragments, respectively. The corresponding hexadecapole moments oscillate in phase with the quadrupole deformation around central, non-zero values. The corresponding octupole deformations, in contrast, are modulated around a zero value, and it is difficult to ascribe a final octupole deformation for these (excited) fragments. While the specific deformations might not be particularly relevant, the excitation patterns of the multipoles provide information on the collective vibrations of both fragments. We note that, whereas a pattern of well defined, relatively rapid oscillations is found for the light fragment, the heavy fragment multipoles have a more erratic time evolution.

These features are reflected in the corresponding power spectra. We insist here that these can only be obtained with the required resolution after a boost has been performed on both fragments. Within the resulting spectra presented for the heavy fragment in Fig. 28, there is a well-defined peak for each multipole parameter between \( \approx 1 \) and 3 MeV. For the light fragment, in contrast, Fig.
FIG. 28. Power spectra corresponding to Fig. 26 (heavy fission fragment). The resolution is significantly improved due to the longer measurement time available with the use of Galilean transformations to remove the linear momentum of the fragments.

FIG. 29. Power spectra corresponding to Fig. 27 (light fission fragment).

V. CONCLUSIONS

We have presented an analysis of the fission process using TDHF techniques as implemented in Sky3D. Starting from the calculated one-fragment quadrupole-constrained PES, the dynamics of fission were investigated. Deformation-induced fission (DIF) was explored by releasing the quadrupole constraint and time-evolving a selection of states situated below, around and beyond the second static fission barrier. Three behaviours were observed. The states with a quadrupole deformation below the peak of the fission barrier undergo vibrations corresponding to a collective giant resonance. For these states, DIF is forbidden in TDHF, as a collective tunnelling through the barrier must occur to reach a fissioned configuration.

A different behaviour is observed for the evolution of states which are situated beyond the peak of the second static fission barrier, but before the critical point where the static one and two-fragment pathways intercept. Upon time evolution up to 9000 fm/c, these states also fail to fission, but the dynamics are not typical of collective giant resonant modes. The repulsive Coulomb force attempts to drive the configuration towards a fission point, but due to the competition with the attractive terms in the energy functional, scission does not occur. DIF is inhibited for these initial configurations, and it can only be speculated if these states would eventually fission with a longer time evolution.

For states with a static deformation exceeding the intersection of the one and two-fragment fission pathways, DIF was observed upon time evolution. We interpret that, as a static two-fragment configuration exists with greater binding energy than the one-fragment configuration, it becomes energetically possible for the one-fragment configurations to evolve to fission with only a modest rearrangement of the densities. The evolution of the pre-fission fragment displays a rearrangement of the densities up until around the point of scission. At this point, the Coulomb repulsion between the pre-formed fragments overpowers the nuclear potential, and translational motion sets in as the fission products rapidly accelerate away from one another. The timescale for DIF varies depending upon the deformation of the initial state. The least deformed configuration demonstrates a density rearrangement phase lasting approximately 1500 fm/c, whereas the most deformed configuration is initially close to the point of scission and the neck ruptures within 100-200 fm/c.

A selection of fission products was observed for the various initial configurations considered. When compared obtained for the post-fission fragments to standard calculations of resonances in the corresponding nuclei. This would give access to a microscopic understanding of the fragment excitation properties, including their temperatures and phonon structure.
to experimental measurements of neutron-induced fission processes, the agreement of the calculated fragment masses demonstrates promising results. The energy released is shown to be dominantly translational kinetic energy, and agreement between theory and experiment was found to be reasonable when comparing the calculated and measured kinetic energies of the post-fissioned systems. We have pioneered a method to remove translational kinetic energy of the post-scission fragments to provide details of their internal excitations.

This initial investigation into fission induced by deformation effects using time-dependent techniques has provided insightful results on the interplay between structure and dynamics in the fission process. We have also devised a set of useful computational analysis tools for the post-scission fragments. Deformation-induced fission, however, provides a limited amount of two-fragment configurations. Further extensions of the present research involving particle-number projection could provide access to more relevant mass distributions. Moreover, a refined step-by-step linear momentum removal could help extend the time length and energy resolution of the two-fragment excitation data.

The present analysis is relevant for a variety of fission processes. Spontaneous fission would presumably tunnel the system across the PES barrier into random states within the “allowed” region. These would subsequently decay into the two-fission pathway. One can in principle explore a wider landscape of fission fragments by exciting nuclei along their fission paths [56]. This theoretical approach can be linked more naturally to induced fission, where the energy deposited by external probes induces the fission process. We plan to explore this approach using TDHF techniques in the near future.

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Appendix A: Principal axis co-ordinate frame in 3D constrained Hartree-Fock calculations

The principal (or intrinsic) axis of the nucleus is an essential concept when performing static or time-dependent calculations in 3D. In unrestricted 3D calculations, there is no requirement that the orientation of the Slater determinant must align with the lab co-ordinate frame [28]. However, performing a constrained minimisation upon the $\beta_{20}$ parameter defined with respect to the fixed laboratory co-ordinate axis may cause the Slater determinant to rotate. This is because the Hartree-Fock minimisation seeks the most bound configuration, and it has no knowledge of the orientation of the nuclear state.

Figure 30 illustrates this point with a crude cartoon. For the case i), $\beta_{20}$ is defined with respect to the laboratory frame. The measurement of $\beta_{20}$ for case ii), also measured in the same frame, could yield the same value. Therefore, a Hartree-Fock minimisation constraining $\beta_{20}$ would converge upon whichever state has the greater binding energy. This can be problematic when a state like ii) is targeted in the minimisation procedure, as in general less deformed states are more bound.

To avoid this issue, we define a principal axis corresponding to the direction of the greatest elongation (see below for more details). In the case iii), the principal $z'$ axis corresponds to the lab $x$ axis shown in case i), as the shape is very slightly deformed. In case iv), the principal axis frame is again chosen to define $\beta_{20}$. Due to this choice, the value of the quadrupole deformation is well defined regardless of any rotations in co-ordinate space, allowing convergence upon the desired state.

Practically, the principal axis co-ordinate frame can be determined by first building the $(3 \times 3)$ quadrupole tensor measured in the laboratory frame, as defined by

$$Q_{ij} = \int d\rho(r) (3r_i r_j - r^2 \delta_{ij}) .$$  \hspace{1cm} (A1)

The tensor should take into account a centre of mass correction, since the origin of the co-ordinate system must be at the centre of mass of $\rho(r)$. The diagonalisation of the $Q$ matrix yields three eigenvalues, $Q_i$. These are ordered by absolute magnitude, and the largest $Q_{3} (= Q_{\text{princ}})$ is chosen to relate to $\beta_{20}$ as measured in the principal axis frame by [58]

$$\beta_{20} = \frac{4\pi}{5} \frac{Q_{\text{princ}}}{A(r^2)} .$$  \hspace{1cm} (A2)
The diagonalisation of $Q$ also produces a set of eigenvalues, $q_i$, which can be used to build a rotation from the laboratory frame to the principal axis frame. These eigenvectors make the components of the rotation matrix $R$,

\[
\begin{pmatrix}
  x' \\
  y' \\
  z'
\end{pmatrix} =
\begin{pmatrix}
  R_{11} & R_{12} & R_{13} \\
  R_{21} & R_{22} & R_{23} \\
  R_{31} & R_{32} & R_{33}
\end{pmatrix}
\begin{pmatrix}
  x \\
  y \\
  z
\end{pmatrix},
\]

where $r = (x, y, z)$ and $R_{ij} = (q_i)_j$. This rotation matrix can be used to determine the fixed axis and the single rotation around that axis which transforms from one frame to the other, allowing an intuitive picture of the alignment of the nucleus. By use of the Euler-Rodrigues formula for 3D rotations, the rotation angle $\theta$ can be defined as

\[
\theta = \arccos \left( \frac{1}{2} (\text{Tr} R - 1) \right),
\]

and the axis of rotation $\hat{n}$:

\[
\hat{n} = \frac{1}{\sqrt{(3 - \text{Tr} R) (1 + \text{Tr} R)}} \begin{pmatrix}
  R_{32} - R_{23} \\
  R_{13} - R_{31} \\
  R_{21} - R_{12}
\end{pmatrix}. \tag{A4}
\]

Special cases arise when $\text{Tr} R$ is equal to $-1$ or $+3$. In the case of an improper rotation, $\det R = -1$, and the rotation is followed by a reflection about the rotating axis.

The rotation matrix, which is determined by considering the quadrupole tensor, could be used to rotate the single-particle wave functions and thus align the laboratory and principal axis. Instead of this computationally demanding case, we compute the multipole deformation parameters in terms of the $(x', y', z')$ coordinates. This immediately yields multipoles that are aligned with respect to the principal axis reference frame.

### Appendix B: Masking procedure for calculation of deformation parameters

The masking function $\mathcal{M}(r)$ is a Fermi function which equals unity ‘inside’ the nuclear surface, and transitions smoothly to zero ‘outside’ of the nucleus. It is included when calculating the expectation value of the multipole deformation parameters. This effectively removes contributions from the wave functions which extend far beyond the nuclear surface when determining the expectation value of the deformation parameters. We note that this is particularly important for multipoles that increase with coordinates to a power of $l$. Any spurious strength at the box edges can translate into arbitrarily large multipoles. Moreover, the masking function must be chosen carefully to allow room for the density to change shape as the iterative procedure moves from one constrained configuration to the next.

A robust strategy to define $\mathcal{M}(r)$ uses an automatically updating mask (see Fig. 31), which has been applied successfully in the axially symmetric Hartree-Fock solver SKYAX [70]. At each iteration, the maximum value of the isoscalar density $\rho_{\text{max}}$ is determined. A grid point “inside” the nucleus is defined as one where $\rho(x, y, z) \geq \rho_{\text{max}}/10$, and all others are “outside”. A binary array corresponding to the density may then be stored, which is true for grid points inside the nucleus, and false outside.

The next step is to determine for every grid point the minimum distance to the nuclear surface. This is a costly process, requiring in principle $N_x N_y N_z^2$ operations at every time step. Instead, one can check if the nuclear surface has changed at each iteration. When it has not, recalculation of the mask is not necessary. With the minimum distance to the nuclear surface of each grid point, $D(x_i, y_i, z_i)$, stored in an array, the masking function can then be defined as:

\[
\mathcal{M}(x, y, z) = \frac{1}{1 + \exp\left(\frac{[D(x, y, z) - \alpha]}{\gamma}\right)}, \tag{B1}
\]

where

\[
D(x, y, z) = \begin{cases}
  +D(x, y, z), & D \text{ ‘outside’} \\
  -D(x, y, z), & D \text{ ‘inside’}
\end{cases}. \tag{B2}
\]

The parameters $\alpha$ and $\gamma$ modulate how rapidly the mask tails off at the nuclear surface. Typical values are $\alpha = 3.5$ and $\gamma = 0.5$ fm.

This masking technique is adjusted to the changing nuclear shape over a PES without manual input. However,
FIG. 32. Two-fragment dynamics may be investigated by defining the dividing plane of the system. The dividing plane is defined to be normal to the principal axis at the point of minimum particle density.

one must be aware that the definitions of the deformation parameters will change slightly every time the mask readjusts, which in some rare occasions caused convergence problems.

Due to the nature of the mask being fixed to grid points, this method is unsuitable in large amplitude TDHF calculations. In dynamic calculations, the definition of the mask is not as straightforward (see section III). The mask needs to follow the centre of mass of each fragment, which does not lie necessarily in a mesh point. For a two-fragment system, the published version of Sky3D measures the total quadrupole moment of the system and obtains the principal axis. The dividing plane is then defined as that which is perpendicular to the point of minimum density along the principal axis, as shown in Fig. 32. An interpolation is then performed to obtain this dividing plane, as the centre of masses of the two fragments may not necessarily lie on integer grid points. Observables relating to the two fragments, including their respective centre of masses, may then be deduced by integrating the densities either side of the dividing plane.

This process is then taken a step further by adding a mask around each fragment. The masks are of the form of a Fermi function (as used in all other cases), and are chosen to extend to a fixed distance from the centre of mass of the fragments. They are recalculated at every step to follow the movement of the fragment. One caveat of this method, however, is that the masks must not overlap when measurements are taken (see Fig. 33, left panel). In other words, the fragment analysis cannot begin at the point of scission: the particles must be separated sufficiently before observables may be calculated. Once the fragments are well separated, so that the masks do not overlap (Fig. 33, right panel), the masks may be used to integrate quantities of interest over the region defining the individual fragments. These quantities include, for example, the particle number, the energy density functional and the deformation parameters.

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