A Microscopic Study of Nuclear Fission using the Time-Dependent Hartree-Fock Method

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The animation presented in the bottom right corner of odd page numbers from 1-145 corresponds to particle density slices of a deformation induced fission process, starting from an initial state with $\beta_{20} = 1.19$. The time interval is 12.5 fm/c.

On the bottom left corner of even page numbers from 146-2, the animation corresponds to particle density slices of a boost induced fission process, starting from an initial state with $\beta_{20} = 0.89$. The boost provides 225 MeV of collective energy instantaneously in the form of a quadrupole excitation. The time interval is 25 fm/c. In both cases, the isolines are separated by 0.05 particles/fm$^3$. 
Abstract

This thesis is concerned with the application of the time-dependent Hartree-Fock method, adopting the Skyrme effective nuclear interaction, to investigate induced nuclear fission processes. Three-dimensional, symmetry-unrestricted constrained Hartree-Fock calculations of $^{240}$Pu are presented to obtain the static potential energy surface corresponding to an increasing quadrupole deformation. The time-dependent Hartree-Fock method is applied to evolve in time these deformed states. Different types of fission process are considered. Firstly, the evolution of static configurations which are deformed sufficiently such that they fission upon time evolution, without any external excitation, (`deformation-induced' fission) is investigated. The fragments smoothly form during a gradual evolution of the static state (taking up to $\approx 1500$ fm/c for scission to occur), and the resulting fission products agree well with experimental neutron-induced fission data. The kinetic energy released in the process is also shown to compare well to experimental results. Secondly, the effect of providing an external excitation field to static configurations which would not otherwise fission (`boost-induced' fission) is investigated. Upon application of an instantaneous excitation, the evolution of the densities displays a violent oscillatory behaviour as the state evolves to fission, in contrast what was seen for the case of deformation-induced fission. The resulting fission products lie at the edges of typical mass distributions obtained from experiment. The evolution of the densities following the application of a gradual excitation field demonstrates behaviour similar to deformation-induced fission, suggesting that the timescale for the energy deposition has consequences regarding the fission dynamics. The comparison to experimental results following the application of a gradual excitations field is improved compared to an instantaneous excitation. Overall, the results provide a significant exploratory investigation of fission treated as a dynamic process, and suggest that the time-dependent Hartree-Fock method has potential to be applied further to describe the dynamics of nuclear fission.
Acknowledgements

I would like to acknowledge and thank my supervisors, Paul Stevenson and Arnau Rios Huguet, for their guidance, motivation and patience over the past three years. They have made my PhD an enjoyable and satisfying experience, and their knowledge and advice has been invaluable.

I would also like to thank Stef Jones, not only for putting up with me (especially during the writing of this thesis) but also for her support and love.

My PhD was supported by STFC grant No. ST/J500768/1. This research made use of the DiRAC HPC cluster. DiRAC is the UK HPC facility for particle physics, astrophysics and cosmology and is supported by STFC and BIS. As this thesis proves, it can also be used for nuclear physics.
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Introduction

Theoretical nuclear structure physics is concerned with describing the properties of the many-body microscopic system of fermions known as the atomic nucleus. The nucleus is a complex, self-interacting (correlated) system comprised of positively-charged protons and uncharged neutrons, bound together by the nuclear strong interaction. Chemical elements are defined by their atomic number, $Z$, which corresponds to the number of protons in the nucleus, and for each element there can exist isotopes with varying numbers of neutrons, characterised by the neutron number $N$. The atomic mass of the nucleus $A$ is simply the sum $N + Z$.

There exist in the region of 300 stable isotopes which are found naturally on Earth; for light nuclei these are typically found in isotopes with $Z \approx N$, but as the nuclear systems grow heavier, stable isotopes typically have an abundance of neutrons. This is because the Coulomb interaction between protons is repulsive, so that proton-rich systems tend to be unstable. Observation of isotopes away from the region of stability is an ongoing area of research [1]. Knowledge of the properties of unstable nuclei allows physicists to test their understanding of nuclear synthesis and astrophysical processes, and further to verify the understanding of the behaviour of nuclear systems by providing experimental observations to compare to theories. Over 3000 isotopes have been experimentally observed, from $Z = 1$ to 118 [2].

Stable and unstable isotopes may be synthesised in the laboratory using techniques ranging from nucleon capture and stripping and transfer reactions at low and intermediate energies [3, 4], to extreme processes at high energies such as spallation and fragmentation [5, 6]. Modern radioactive beam facilities have opened up a new region of discovery for unstable isotopes [7], and results from future studies will provide key to verifying predictions made by theo-
retical models employed to describe the structure of nuclei. As many as 6000 isotopes have been predicted to exist [8].

For those unstable isotopes, a transition into a stable configuration will occur by undergoing one or more decay process. These transitions can be as simple as an excited state (an isomer) losing energy by the emission of a photon (γ-decay), and therefore the isotope will keep its identity following the process. Other decays include the emission of an α-particle, or processes such as β⁺ or β⁻ decay [3, 4]. Following decays of these types, the initial isotope will transition to a neighbouring configuration defined by a new N and Z. The aforementioned processes are well known, but not an exhaustive list of decay mechanisms. For example, exotic β decay processes are predicted to occur; neutrinoless double-beta decay is a topic of interest in modern nuclear physics [9].

Nuclear fission (see [4, 10, 11] for general introductory discussions and reviews) presents itself as perhaps the most dramatic decay process, where it becomes energetically favourable for the nucleus to split into two fragments. The mass difference between the parent nucleus and daughter nuclei is distributed between the daughters, mainly in the form of translational kinetic energy. The process can occur spontaneously in some cases [12, 13], and may be induced using a multitude of processes, typically through the absorption of particles or photons [11].

Harnessing the energy released in fission reactions is routinely performed in nuclear reactors for energy production. However, it is the application of fission in weaponry which has spurred intense research into the process since its discovery. The social and political implications of nuclear weaponry has ensured the profound impact that the discovery of fission has had upon modern history [14].

1.1 A Brief Account of the Experimental Discovery and Theoretical Descriptions of Nuclear Fission

Research led by Fermi in 1934 [15] demonstrated that following the capture of thermal neutrons by heavy elements, new radioactive isotopes could be formed. Fermi’s initial interpretation of the decays of these isotopes was through chains of α and β decays. Pursuing these investigations, in 1938 Hahn and Strassmann identified decay products of medium charges (which we know today to be fission products). Following communication with one of Hahn’s
collaborators, Meitner, Hahn and Strassmann published their results in 1939 [16]. The nuclear reaction was named ‘fission’ by Meitner and Frisch, based on the namesake process in cell biology. Meitner and Frisch described nuclear fission in terms of an incompressible charged liquid drop model (LDM), which demonstrated the nucleus splitting into two when subjected to extreme deformation. Following the publication by Meitner and Frisch [17], the basic theoretical understanding of nuclear fission was rapidly established. It was verified independently that a charged droplet becomes unstable to fission when the ratio of the Coulomb energy to twice the surface energy exceeds unity [18, 19]. The seminal paper by Bohr and Wheeler [20] established the concept of the surface energy in terms of deformation parameters.

Calculating the potential energy of a charged liquid drop as a function of increasing elongation (and therefore deformation) introduced the concepts of the potential energy surface (PES) and the fission barrier, shown in Fig. 1.1. For the PES corresponding to the liquid drop model (labelled LDM in the Figure), there exists a potential barrier of height $E_{\text{barrier}}$. Energy must be added to the system to overcome the barrier, thus allowing the nucleus to fission (the so-called activation energy [4]). Bohr and Wheeler presented calculations of the energy required to produce a critical deformation in their 1939 paper [20], and thus estimated fission barrier heights for various actinide nuclei.

The LDM provided an adequate explanation of the mechanism of the fission process, enough such that nuclear weaponry could be developed during the course of the 1940s. However, the liquid drop model was lacking with respect to several factors [21]. Firstly, the resulting fission fragments are always symmetric; nuclei often fission into asymmetric fragments. Additionally, the LDM lacks the inclusion of quantum mechanical ingredients, which are often essential to accurately describe nuclear structure effects.

It wasn’t until the late 1950s-1960s that a renaissance in the calculation of fission barriers began. In 1955, Swiatecki proposed a shell correction to be included in the LDM calculations [22], based upon the difference between LDM calculated masses and experimentally observed ground state masses. This allowed a more accurate calculation of experimentally observed spontaneous fission half-lives, based upon calculating the transmission of a quantum mechanical wave function through a potential barrier.
Figure 1.1: Illustration displaying the form of potential energy surfaces as a function of increasing elongation. The Liquid Drop Model (LDM) displays a single potential barrier. Bohr and Mottleson reasoned that if enough energy was applied so that the nucleus may become critically deformed beyond the peak of the barrier, fission may occur. Experimental results obtained in later years suggested that a double-humped barrier exists for actinide and heavy nuclei. Microscopic shell corrections allow the LDM to possess this structure, displaying a prominent isomeric state (IS) as well as the ground state (GS). See text for further discussion.

Perhaps the most significant breakthrough was in the mid-1960s when Strutinskysky proposed a method to calculate the shell correction as a function of increasing deformation [23, 24]. This began the era of ‘macroscopic-microscopic’ calculations, where some quantum-mechanical shell effects were incorporated into the LDM calculations, and produced ‘double-humped’ potential surfaces for actinide and heavy nuclei (see Fig. 1.1, PES labelled LDM + Shell Correction). These demonstrate two fission barriers beyond the global minimum of the PES (the ground state), and a prominent local minimum, which corresponds to a long-lived fission isomer. Experimental evidence had been obtained to verify this structure [25, 26]. For example, the excitation spectra and lifetimes of isomeric states could be measured [27]. Further, without the multiple barriers in the PES, theoretical calculation of spontaneous fission half-lives for heavy nuclei yielded values which were far too short.

As early as the 1970s Möller and Nix began pursuing a more thorough investigation of the PES by calculating the energies of hundreds of nuclear shapes with more complex deformations within the macroscopic-microscopic approach [28, 29]. The most modern calculations published by Möller determine the potential energies of millions of configurations using a five-dimensional parameterisation. Fission pathways are determined by analysing the topography.
1.1 A Brief Account of the Experimental Discovery and Theoretical Descriptions of Nuclear Fission

of the multi-dimensional PES [30, 31, 32, 33].

Parallel to the development of the macroscopic (and macroscopic-microscopic) approach for describing the properties of the atomic nucleus, with advances in computational abilities the treatment of the nucleus as a many-bodied quantum-mechanical system became possible. In the early 1970s Vautherin and Brink demonstrated that the Hartree-Fock method could be applied (with considerable success) to describe ground-state properties of nuclei when adopting an effective nucleon-nucleon interaction [34]. This method, analogous to the Kohn-Sham Density Functional Theory (DFT) [35, 36] allows the nucleus to be described using the concept of a ‘mean-field’. Each nucleon in the system interacts with the averaged effect of all the other nucleons, and this average mean-field is in turn determined by the presence of the nucleon, which leads to a self-consistent problem.

In contrast to this approach, the most fundamental modern models for describing properties of nuclear structure are ab initio techniques, which adopt a bare nuclear interaction [37]. Modern calculations include up to four-body interaction terms [38]. The form of the nuclear interaction is a key ingredient to many-body nuclear theory (and is not considered at all in purely macroscopic calculations). As early as 1935, Yukawa presented the concept of the bare nucleon-nucleon force being mediated by pion exchange [39]. In the 1980s and 1990s, the concepts of Quantum Chromodynamics and Effective Field Theory developed significantly, allowing a fundamental description of the nucleon-nucleon interaction [40].

Ab initio methods adopting bare nuclear interactions are limited to describing nuclei of light masses; perhaps the most advanced calculations are breaking into the $A$=50-100 mass regions [41]. For studies of nuclear fission, where actinide nuclei are typically of interest ($Z$=89-103), the application of an ab initio approach is unfeasible at present.

In contrast, mean-field calculations assuming effective interactions allow access to the entire nuclear chart [8]. Effective nuclear interactions are fitted phenomenologically to experimental data, and have been proven to be remarkably successful when adopted in Hartree-Fock calculations [21, 42]. The zero-ranged Skyrme effective interaction [43] has been used extensively in Hartree-Fock calculations since the work of Vautherin and Brink. Finite-range interactions, such as the Gogny interaction [44], are also frequently used.

The Hartree-Fock method using the Skyrme effective interaction has been applied to calculate fission barriers as early as 1973, where constrained Hartree-Fock calculations were
performed by Flocard *et. al* [45, 46] to determine the minimum energy configuration of nuclei subject to a fixed quadrupole deformation. The double-humped structure of the PES was obtained. This result was remarkable, considering that other than the choice of the Skyrme interaction, the structure was obtained without the inclusion of any other corrections or additional parameters. This confirmed that within such a framework, the shell effects needed to describe the structure of the fission barriers are intrinsically included.

The early calculations performed by Flocard included symmetry restrictions due to the limitations of computational ability. Modern Hartree-Fock solvers are capable of performing symmetry-unrestricted calculations, and have also increased the number of constraints to investigate a higher dimensional PES [47, 48, 49, 50, 51, 52]. Typically the octupole deformation is constrained simultaneously with the quadrupole deformation, so that the effects of mass asymmetry may be investigated.

Advances in the description of fission barriers has been impressive over the past 40-50 years, both from the perspective of the shell-corrected liquid-drop model and constrained Hartree-Fock. However, both approaches are lacking in the description of nuclear fission as a dynamic process. Using these models, fission is essentially analysed by calculating a series of static configurations to determine the optimum path from the ground state to fission, essentially describing an adiabatic process. This adiabatic assumption may be valid as a nuclear state gradually transitions towards a fissioned configuration, but around the point of scission the dynamics become non-adiabatic as the neck breaks and the fragments rapidly accelerate away from one another due to the Coulomb interaction [53].

The time-dependent Hartree-Fock (TDHF) method has been applied to describe the non-adiabatic dynamics of nuclear fission since the late 1970s. The method allows static Hartree-Fock states to be time-evolved within a microscopic framework. Negele *et al.* published one of the best known early applications of the method to nuclear fission in 1978 [54]. They analysed the dynamics of $^{236}\text{U}$, initialising their calculations from quadrupole-constrained static configurations either side of the peak of the second static fission barrier. Due to computational limitations, axial and reflection symmetry was assumed, and a simplified nuclear interaction was applied with no spin-orbit force. Time-dependent pairing was included in order to break the symmetries of the system to allow fission. A number of fission configurations were found when evolving the initial state with various pairing strengths. The authors con-
cluded that symmetry unrestricted calculations and the inclusion of a spin-orbit interaction were necessary to further investigate the dynamics of fission.

During the 1980s, several other investigations were published. For example, the work of Okołowicz et al. [55] in 1983. The approach of Ockołowicz was to apply a quadrupole excitation to a nuclear state either side of the second static fission barrier. The static states were created using either a single-centred or two-centred trial wave function. Pairing was not included in the calculations, and once again axial symmetry was imposed and no spin-orbit force included. The authors demonstrated that fission could be induced in axially symmetric calculations with the absence of a pairing force to break symmetries. They found applying a quadrupole excitation (or ‘boosting’) allowed the static configurations built from two-centred trial wave functions to fission. Those built from single-centred trial wave functions, however, failed to fission. They concluded that different angular momentum boosts should be explored in future work, suggesting that quadrupole shape deformations may not be the most appropriate degree of freedom to invoke for inducing fission dynamics.

Jung et al. [56] published work in 1988 investigating the effect of inducing fission either by instantaneously changing the ground state density, or applying a boost. In addition to quadrupole boosts, they investigated excitations with more exotic spatial profiles in an attempt to describe multi-fragmentation and single fragment emission, as well as fission. Their brief results presented for fission also investigated the the effect of bisecting the nucleus with a plane of zero density, and they established a limiting value of separation where the Coulomb repulsion would drive the two halves of the nucleus apart.

Application of TDHF for the investigation of fission all but stopped for several decades, presumably due to the symmetry restrictions imposed when performing the calculations. With advances in computation power, a new generation of three-dimensional, symmetry unrestricted TDHF codes has been developed (e.g. [57, 58, 59]). In 2010 Umar et al. [60] investigated fusion-fission of $^{240}\text{Pu}$ by colliding $^{100}\text{Zr}$ and $^{140}\text{Xe}$, and then fissioning the composite system by applying velocity boosts. By performing density-constrained TDHF calculations of the fissioning nuclei, the potential barriers of the system could be investigated. In 2014, Umar and Simenel [53] applied TDHF to investigate the formation and dynamics of fission fragments in the symmetric fission of $^{264}\text{Fm}$ during the non-adiabatic stages of the fission process.
1. INTRODUCTION

Few studies have been performed using modern TDHF solvers, and it is timely for these exploratory investigations to be expanded to determine the suitability of TDHF to investigate the dynamics of fission. A realistic description of fission as a dynamic process will complement and expand upon previous theoretical attempts to describe the process with static calculations, as done during the course of the past 80 years.

1.2 Scope of this Thesis

After appropriate modification to investigate the physics of interest, the Hartree-Fock and time-dependent Hartree-Fock solver Sky3D [57] will be applied to study the dynamic properties of nuclear fission. Chapter 2 will summarise the Hartree-Fock and time-dependent Hartree-Fock methods, adopting the Skyrme effective nuclear interaction.

The fission properties of the actinide nucleus $^{240}$Pu will be investigated, as this isotope is often used for benchmark calculations within the mean-field approximation [61]. The functionality to perform constrained static Hartree-Fock calculations will firstly be incorporated into Sky3D, so that the static PES corresponding to the quadrupole degree of freedom may be obtained. The results of the constrained Hartree-Fock calculations will be presented in Chapter 3, and the configurations obtained may then be used as starting points for time evolution.

Two types of dynamic fission process will be investigated using TDHF. In Chapter 4, deformation-induced fission will be presented. For this process, the initial states are those static configurations which are deformed sufficiently so that upon time evolution a fissioned configuration will be reached.

Chapter 5 will explore boost-induced fission, where a quadrupole excitation field either with an instantaneous or Gaussian temporal profile will be applied. The states to be investigated are those which are not deformed sufficiently to undergo deformation-induced fission. The results presented in this thesis, and an outlook for future work, will be summarised in Chapter 6.
2

Applications of Many-Body Theory to a Nuclear System

2.1 Microscopic Many-Body Systems

Non-relativistic microscopic many-bodied systems are described by the Schrödinger equation,

\[ \hat{H} \Psi = E \Psi, \]  

(2.1)

where \( \hat{H} \) is the Hamiltonian operator, \( \Psi \) the many-body wave function, and \( E \) is the energy. The many-body wave functions which are solutions to the Schrödinger equation contain the ground state and excitation spectrum of the system. A general Hamiltonian describing a nuclear system, \( \hat{H} \), consists of a one-body kinetic energy operator and an operator describing the nucleon-nucleon interaction. This term describes two-body, three-body, ..., \( N \)-body interactions. In practice, however, this contribution is often truncated at the level of two-body interactions due to the technical difficulty of solving the equations. Even effective field theories, which present themselves as cutting edge techniques capable of describing realistic interactions in a nuclear system, are limited to 4-body interactions [38].

Few cases exist where the exact many-body description of a nuclear system is known [62]. In general, simplifications are required to create tractable problems. These typically include restricting the Hilbert space of the many-body wave function, and making approximations in the form of the nucleon-nucleon interaction.
2. APPLICATIONS OF MANY-BODY THEORY TO A NUCLEAR SYSTEM

2.1.1 The Many-Body Wave Function and Slater Determinants

The many-body wave function is a mathematical construct postulated to describe all the available information of a microscopic quantum system. Within a quantum mechanical description of a nuclear system governed by the Schrödinger equation, one must adopt a many-body wave function describing an $N$-particle system

$$\Psi = \Psi(r_1, ..., r_N).$$  \hfill (2.2)

The co-ordinate $r$ is chosen to represent the space, spin and isospin degrees of freedom $(r, \sigma, q)$ for each particle, such that

$$\int dr = \sum_{\sigma, q} \int dr.$$  \hfill (2.3)

In this thesis, the nuclear wave function will be written as $\Psi(r)$, with the spin and isospin degrees of freedom left implicit for brevity.

The simplest way to build an antisymmetric many-body wave function out of $N$ single-particle states, $\varphi_i$, with $i = 1, ..., N$, is a Slater determinant

$$\Phi = \frac{1}{\sqrt{N!}} \left| \begin{array}{ccc} \varphi_1(r_1) & \ldots & \varphi_1(r_N) \\ \vdots & \ddots & \vdots \\ \varphi_N(r_1) & \ldots & \varphi_N(r_N) \end{array} \right|.$$  \hfill (2.4)

The consequence of the wave function being anti-symmetric is that the Pauli principle is enforced, which is a fundamental necessity for a system of indistinguishable fermions. The Slater determinant is convenient for expressing a many-body wave function, however it is not the most general form. At the price of allowing feasible calculations, the description of particle-particle correlations within the system is sacrificed (other than those intrinsically defined by the Pauli principle). In the context of this thesis, the most important correlation that needs to be explicitly accounted for is the short-range pairing interaction, which will be discussed in Sec. 2.5.1.

2.2 The Hartree-Fock Method

The Hartree-Fock approximation is used to describe a many-body system of fermions in terms of an effective single-particle problem. A starting point is to consider a non-relativistic Hamiltonian which contains only one-body kinetic terms and a two-body nucleon-nucleon
2.2 The Hartree-Fock Method

interaction $\hat{v}$:

$$ \hat{H} = \sum_{i=1}^{N} \left( -\frac{\hbar^2}{2m} \nabla_i^2 \right) + \frac{1}{2} \sum_{i \neq j}^{N} \hat{v}_{ij} \hat{\phi}_{ij}. $$

(2.5)

An effective nucleon-nucleon interaction is usually used in Hartree-Fock calculations [21, 42]. The Skyrme effective interaction is a popular choice, and will be discussed in Sec. 2.3.

The aim of the Hartree-Fock method is to approximate the two-body Hamiltonian operator as an effective single-particle potential. The many-body wave function that is a solution to the eigenvalue equation,

$$ \hat{H} \Phi = E_{HF} \Phi, $$

(2.6)

is a Slater determinant. The energy $E_{HF}$ is the Hartree-Fock energy, which is an approximation of the exact energy $E$. The single-particle wave functions $\varphi_i$, with corresponding eigenvalues $\epsilon_i$, can be expressed by

$$ \hat{h} \varphi_i(r) = \epsilon_i \varphi_i(r), $$

(2.7)

where the effective single-particle Hamiltonian is given by

$$ \hat{h} = \sum_{i=1}^{N} \left( -\frac{\hbar^2}{2m} \nabla_i^2 + \hat{v}_{HF}^{(i)} \right). $$

(2.8)

The operator $\hat{v}_{HF}$ acts as an effective single-particle potential. This Hartree-Fock potential, and therefore the Hartree-Fock energy, can be recovered by minimising the expectation value of the two-body Hamiltonian. This is analogous to the Kohn-Sham Density Functional Theory (DFT) [35, 36], with the philosophical difference that in DFT, only the densities have physical significance, and the wave functions are auxiliary objects. In DFT, the existence of an energy density functional is postulated in its most general form. An energy functional is provided and minimised to determine the ground state. From the Hartree-Fock perspective, the nucleon-nucleon interaction is provided, and the energy density functional is determined by minimising the energy of the system.

2.2.1 The Ritz Variational Principle

The energy functional for a system in a state $\Psi$ can be expressed by

$$ E[\Psi] = \frac{\langle \Psi | \hat{H} | \Psi \rangle}{\langle \Psi | \Psi \rangle}. $$

(2.9)
The Ritz variational principle is an important concept for variational calculations, which can be applied to quantum many-body theory. It states that the many-body wave function $\Psi$ that makes the energy functional $E[\Psi]$ stationary, where $\Psi$ is allowed to vary over the whole Hilbert space, is an eigenstate of the Hamiltonian $\hat{H}$ belonging to the eigenvalue $E$ \([63]\). It is this variational principle which can be used to derive the Hartree-Fock equation.

The Ritz variational principle may be demonstrated by considering an infinitesimal variation of $\Psi^*$. The corresponding variation of the functional is given by

$$\frac{\delta E[\Psi]}{\delta \Psi^*} = \frac{\delta}{\delta \Psi^*} \left( \frac{\langle \Psi | \hat{H} | \Psi \rangle}{\langle \Psi | \Psi \rangle} \right) = \frac{\langle \delta \Psi | \hat{H} - E | \Psi \rangle}{\langle \Psi | \Psi \rangle}. \quad (2.10)$$

Assuming the variation of $E[\Psi]$ vanishes for an infinitesimal variation $\delta \Psi^*$, the relation

$$\langle \delta \Psi | \hat{H} - E | \Psi \rangle = 0 \quad (2.11)$$

is recovered. If this relationship is satisfied for an arbitrary variation (that is, considering an unrestricted wave function), then the Schrödinger equation $\hat{H} | \Psi \rangle = E | \Psi \rangle$ is recovered \([21, 63]\). If the variation in Eq. (2.10) was taken with respect to $\Psi$ rather than $\Psi^*$, the Hermitian conjugate of the Schrödinger equation would be recovered.

The variational principle has powerful implications. The eigenfunctions which satisfy the variational principle correspond to the lowest energy many-body state within the restricted space of the wave function. A trial wave function, however, must be chosen such that the expectation value of the Hamiltonian (Eq. (2.9)) may be evaluated. The average value of the energy from trial wave functions will always be equal to or greater than the energy of the ground state:

$$E[\Psi] \geq E[\Psi_0]. \quad (2.12)$$

Therefore, the ground state serves as a lower bound for variational calculations.

In the case of the Hartree-Fock approximation, the Ritz variational principle is applied by constructing a trial wave function which is (and always remains) a Slater determinant $\Phi$. The wave function is then determined by making the expectation value of the energy stationary with respect to infinitesimal variations of the wave function when inserting a Hamiltonian containing one and two-body operators. Unless the ground state Hamiltonian belongs to the class of the Slater determinant wave functions, the state $\Phi$ which makes the energy functional stationary will not be a true eigenstate of the Hamiltonian.
2.2 The Hartree-Fock Method

The philosophy of the Hartree-Fock method, however, is that the Slater determinant $\Phi$ can be a very good approximation to the eigenstate of the Hamiltonian. One must be aware that $E_{\text{HF}}$ is not necessarily equivalent to the eigenvalue $E$ that would be obtained from solving the full many-body problem exactly. The variation of the energy functional is equated to zero to find a stationary value, and for the case of the Slater approximation it may be written

$$0 = \delta \left[ \langle \Phi | \hat{H} | \Phi \rangle - \sum_{i=1}^{N} \epsilon_i \int \varphi_i^*(r) \varphi_i(r) dr \right],$$

(2.13)

where $\epsilon_i$ is a Lagrange multiplier that insists upon the orthonormalisation of the single-particle wave functions in the subspace of the solution.

2.2.2 Derivation of the Hartree-Fock Equation from the Variational Principle

The Hartree-Fock equation may be derived by performing the variation of the general two-body Hamiltonian given by Eq. (2.5) in the restricted subspace of the Slater determinant. The variation can be performed separately with respect to $\varphi_\alpha(r)$ or $\varphi_\alpha^*(r)$, assuming the other is an independent variable. It sufficient to consider only one, as taking the variation with respect to the other produces the adjoint equation.

The expectation value of $\hat{H}$ may be written

$$\langle \Phi | \hat{H} | \Phi \rangle = \sum_{i=1}^{N} \int \varphi_i^*(r) \left( -\frac{\hbar^2}{2m} \nabla_i^2 \right) \varphi_i(r) \, dr$$

$$+ \frac{1}{2} \sum_{i,j=1}^{N} \int \int \varphi_i^*(r) \varphi_j^*(r') v(r,r') \varphi_i(r) \varphi_j(r') \, dr \, dr'$$

$$- \frac{1}{2} \sum_{i,j=1}^{N} \int \int \varphi_i^*(r) \varphi_j^*(r') v(r,r') \varphi_j(r) \varphi_i(r') \, dr \, dr'. \quad (2.14)$$
The variation given by Eq. (2.13) can be performed explicitly

\[
0 = \frac{\delta}{\delta \varphi^*_\alpha(x)} \left[ \langle \Phi | \hat{H} | \Phi \rangle - \sum_{i=1}^{N} \epsilon_i \int \varphi^*_i(r) \varphi_i(r) \, dr \right]
\]

\[
= \left( -\frac{\hbar^2}{2m} \nabla^2 \right) \varphi_\alpha(x) + \frac{1}{2} \sum_{j=1}^{N} \varphi^*_j(r') \varphi_j(x) \varphi_\alpha(x) \, dr' + \frac{1}{2} \sum_{j=1}^{N} \varphi^*_j(r') \varphi_j(x) \varphi_\alpha(x) \, dr' - \frac{1}{2} \sum_{i=1}^{N} \int \varphi^*_i(r) \varphi_i(x) \varphi_\alpha(x) \, dr - \epsilon_\alpha \varphi_\alpha(x)
\]

\[
= \left( -\frac{\hbar^2}{2m} \nabla^2 \right) \varphi_\alpha(x) + \sum_{j=1}^{N} \int \varphi^*_j(r) \varphi_j(x) \varphi_\alpha(x) \varphi_i(x) \, dr - \epsilon_\alpha \varphi_\alpha(x). \tag{2.15}
\]

The vector \( \mathbf{x} \) is introduced to represent the position, spin and isospin co-ordinates of the conjugate wave function \( \varphi^*_\alpha(x) \), which the variation is taken with respect to. The sums over the wave functions can be rewritten in terms of the one-body density matrix

\[
\rho(r, r') = \sum_{i=1}^{N} \varphi^*_i(r') \varphi_i(r). \tag{2.16}
\]

For the case where \( r \to r' \), the local density may be written

\[
\rho(r, r) \equiv \rho(r) = \sum_{i=1}^{N} \varphi^*_i(r) \varphi_i(r). \tag{2.17}
\]

This allows Eq. (2.15) to be written as the Hartree-Fock equation

\[
\epsilon_\alpha \varphi_\alpha(r) = \left[ -\frac{\hbar^2}{2m} \nabla^2 + \int \rho(r') v(r, r') \, dr' \right] \varphi_\alpha(r) - \int \rho(r, r') v(r, r') \varphi_\alpha(r') \, dr'. \tag{2.18}
\]

The single-particle energy of each orbital is now written in terms of a kinetic energy term and the one-body, non-local Hartree-Fock potential:

\[
v_{\text{HF}}(r) \varphi_i(r) = \int \rho(r') v(r, r') \, dr' \varphi_i(r) - \int \rho(r, r') v(r, r') \varphi_i(r') \, dr'. \tag{2.19}
\]

which may be inserted into Eq. (2.8) to define the Hartree-Fock single-particle Hamiltonian.
2.3 The Hartree-Fock Method with Skyrme’s Effective Interaction

Expressions for the Hartree-Fock energy $E_{HF}$ can be determined by inserting the Hartree-Fock equation (Eq. (2.18)) into the expectation value of the Hamiltonian (Eq. (2.14)) [64]. The Lagrange multipliers $\epsilon_\alpha$ may be interpreted using Koopman’s theorem as the particle ionisation energies, that is, the energy required to remove the particle in state $\alpha$ from the nucleus. One must be aware that the sum of the single-particle energies is not equal to the Hartree-Fock energy. As the interaction between particles $i$ and $j$ is identical to the interaction between particles $j$ and $i$, a double counting will occur when summing the single-particle energies, which must be corrected for. For the application of Hartree-Fock with effective nucleon-nucleon interactions, it will be shown in the following that the energy of the system may be determined by constructing and integrating a universal energy density functional which describes the nuclear system.

2.3 The Hartree-Fock Method with Skyrme’s Effective Interaction

Skyrme proposed an effective interaction in 1959 [43] to describe the effective nucleon-nucleon interaction in the particle-hole channel. The underlying assumption was that the interaction could be expressed by an expansion on the range of the interaction, starting from a zero-range and building up the inter-particle dependence in terms of derivatives. This interaction therefore contains a zero-range contact term, and momentum terms which arise due to higher orders in the expansion. These momentum terms allow some finite-range effects to be captured. Reference [65] instructively demonstrates that taking the zero-range limit of the finite-range Gogny interaction results in the same momentum dependence included in the Skyrme interaction.

The proposed interaction included both a two and three-body interaction term, and was successfully adopted by Vautherin and Brink in Hartree-Fock calculations for nuclei in the early 1970’s [34, 66]. The work of Vautherin and Brink demonstrated remarkably good results, reproducing experimentally observed binding energies and nuclear radii across the nuclear chart to a reasonable degree of precision, despite the simple mathematical form assumed for the nuclear interaction.

Use of the Skyrme interaction has remained popular in Hartree-Fock calculations for over 40 years, allowing descriptions both of properties of nuclear structure and nuclear matter.
This popularity has held due to the fact the zero-range interaction vastly reduces the complexity of calculations as both the direct and exchange terms have the same mathematical form \cite{42, 61, 67}. This allows the energy density functional of the system to be expressed solely in terms of local densities.

Effective interactions with finite ranges, such as the Gogny interaction \cite{21, 44} are argued to capture more ‘realistic’ physics, as intuitively one would expect a local interaction to be unable to reproduce the long or intermediate range properties of the nuclear interaction. In particular, when describing nucleon-nucleon pairing correlations, a finite-range interaction would be more desirable \cite{44}.

With increased computational power, recent efforts have begun utilising finite-range interaction forces in three dimensional, symmetry unrestricted calculations \cite{47}. However, due to the fact that the Skyrme interaction provides remarkably good results in comparison to calculations using finite-range effective interactions, with the added advantage of being considerably less demanding to use, it remains a popular, practical choice to date when pushing Skyrme Hartree-Fock (or equivalently, Density Functional Theory) calculations of nuclei to modern limits.

2.3.1 Functional Form

The Skyrme interaction \cite{43} contains a two and three-body part and describes the nucleon-nucleon interaction in terms of a zero-range expansion. The two-body part stems from the contact term and momentum dependence, and also includes spin exchange, spin-orbit \cite{68} and tensor effects. It can be written (adopting the notation from Ref. \cite{69}):

\[
\hat{v}^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = t_0 \left( 1 + x_0 \hat{P}_\sigma \right) \delta(\mathbf{r}_1 - \mathbf{r}_2) \\
+ \frac{t_1}{2} \left( 1 + x_1 \hat{P}_\sigma \right) \left[ \hat{k}^2 \delta(\mathbf{r}_1 - \mathbf{r}_2) + \delta(\mathbf{r}_1 - \mathbf{r}_2) \hat{k}^2 \right] \\
+ t_2 \left( 1 + x_2 \hat{P}_\sigma \right) \hat{k}\delta(\mathbf{r}_1 - \mathbf{r}_2) \hat{k} \\
+ iW_0 \hat{k}'\delta(\mathbf{r}_1 - \mathbf{r}_2) (\hat{\sigma}_1 + \hat{\sigma}_2) \times \hat{k} \\
+ \frac{t_e}{2} \left( 3(\hat{\sigma}_1 \cdot \hat{k}')(\hat{\sigma}_2 \cdot \hat{k}') - (\hat{\sigma}_1 \cdot \hat{\sigma}_2) \hat{k}'^2 \right) \delta(\mathbf{r}_1 - \mathbf{r}_2) \\
+ \frac{t_e}{2} \left[ \delta(\mathbf{r}_1 - \mathbf{r}_2) \left( 3(\hat{\sigma}_1 \cdot \hat{k}) (\hat{\sigma}_2 \cdot \hat{k}) - (\hat{\sigma}_1 \cdot \hat{\sigma}_2) \hat{k}^2 \right) \right] \\
+ \frac{t_o}{2} \left( 3(\hat{\sigma}_1 \cdot \hat{k}') (\hat{\sigma}_2 \cdot \hat{k}) - (\hat{\sigma}_1 \cdot \hat{\sigma}_2) \hat{k}'^2 \delta(\mathbf{r}_1 - \mathbf{r}_2) \hat{k}^2 \right). \tag{2.20}
\]
2.3 The Hartree-Fock Method with Skyrme’s Effective Interaction

Here, $\hat{k}$ and $\hat{k}'$ are the momentum operators, which are defined by

\[
\hat{k} = -\frac{i}{2}(\nabla_1 - \nabla_2)
\]
\[
\hat{k}' = \frac{i}{2}(\nabla'_1 - \nabla'_2).
\]  \hspace{1cm} (2.21)

The operator $\hat{k}$ acts to the right (upon the wave function), and $\hat{k}'$ to the left (upon its complex conjugate). The $\hat{\sigma}_1, \hat{\sigma}_2$ are the Pauli spin matrices operating on particle 1 and 2, respectively. The spin-exchange operator $\hat{P}_\sigma$ is defined by:

\[
\hat{P}_\sigma = \frac{1}{2}(1 + \hat{\sigma}_1 \cdot \hat{\sigma}_2).
\]  \hspace{1cm} (2.22)

The $t_0, t_1, t_2, x_0, x_1, x_2, W_0, t_e$ and $t_o$ are all free parameters, which define a unique Skyrme parametrization. They are typically fitted to nuclear structure data, nuclear matter properties, or equations of state. To date, over 200 parameterizations have been published by various authors [70]. The term proportional to $t_0$ corresponds to the contact interaction, and the $t_1$ and $t_2$ correspond to the momentum dependence which allows the description of some finite-range effects. The term proportional to $W_0$ is the spin-orbit part of the nucleon-nucleon interaction. The time-even and time-odd tensor terms, proportional to $t_e$ and $t_o$, were widely neglected for many years since the initial proposal by Skyrme, and hence are not included in many parameterizations. Recent studies of nuclear structure properties have been dedicated to investigating the effect of the re-inclusion of these terms [67, 69, 71, 72].

The three-body term is also assumed to have zero-range, and is formally equivalent to a density dependent two-body term [34]. It is included in the functional by the term proportional to $t_3$:

\[
\hat{v}^{(3)}(r_1, r_2) = \frac{t_3}{6} \left( 1 + x_3 \hat{P}_\sigma \right) \rho \left( \frac{r_1 + r_2}{2} \right)^\alpha \delta(r_1 - r_2).
\]  \hspace{1cm} (2.23)

The exponent $\alpha$ was taken in early parametrizations to be equal to unity [34, 66]. However, in later publications other values, $1/6 < \alpha < 1/3$, have been subsequently explored to improve the compressibility properties of nuclear matter [73, 74]. Taking the exponent to be other than unity disassociates the term from a zero-range three-body force. At that point, it should be considered as a density-dependent part of the interaction [75].

The full Skyrme interaction $\hat{v}_{\text{skyrmee}}$ can be written as the sum of Eqs. (2.20) and (2.23). The total binding energy of the system in the Skyrme Hartree-Fock (SHF) approach can be expressed as the sum of the kinetic energy, the potential energy (which is recovered from
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integrating the Skyrme energy density functional), and the Coulomb energy due to the charge of protons:

\[ E = E_{\text{kinetic}} + E_{\text{Skyrme}} + E_{\text{Coul}}. \]  

(2.24)

Each contribution can be defined in terms of local densities and currents, which are in turn obtained from the non-local one-body density and spin density matrices [69, 76, 77]

\[ \rho(r, r') = \sum_{\sigma, q} \rho_q(r, r', \sigma, \sigma') \xi_i^*(r', \sigma, \sigma') \xi_i(r, \sigma, q) \]  

(2.25)

\[ S(r, r') = \sum_{\sigma, \sigma', q} \rho_q(r, r', \sigma, \sigma') \langle \sigma | \sigma' \rangle = \sum_{\iota, \sigma, \sigma', q} \xi_i^*(r', \sigma, \sigma') \hat{\sigma}_{\sigma \sigma'} \xi_i(r, \sigma, q), \]  

(2.26)

where \( \sigma = \pm 1/2 \) is the spin projection, and \( q \) is the isospin projection, denoting proton or neutron wave functions or densities. The operator \( \hat{\sigma}_{\sigma \sigma'} \) denotes the elements of the Pauli spin matrices. Note here that the spin and isospin degrees of freedom are explicitly written, whereas they have often been implicitly assumed. The following local densities can be defined from the non-local densities:

\[ \rho(r) = \rho(r, r')|_{r=r'} \]

\[ \tau(r) = \langle \nabla \cdot \nabla' \rangle \rho(r, r')|_{r=r'} \]

\[ S_\mu(r) = S_\mu(r, r')|_{r=r'} \]

\[ T_\mu(r) = \langle \nabla_\mu \cdot \nabla'_\mu \rangle S_\mu(r, r')|_{r=r'} \]

\[ j_\mu(r) = -\frac{i}{2} (\nabla_\mu - \nabla'_\mu) \rho(r, r')|_{r=r'} \]

\[ J_{\mu \nu}(r) = -\frac{i}{2} (\nabla_\mu - \nabla'_\mu) S_\nu(r, r')|_{r=r'} \]

\[ F_\mu(r) = \frac{1}{2} \sum_{\nu=x} (\nabla_\mu \nabla'_\nu + \nabla'_\mu \nabla_\nu) S(r, r')|_{r=r'}. \]  

(2.27)

The local densities are the scalar particle density \( \rho(r) \) (time-even), the scalar kinetic density \( \tau(r) \) (time-even), the vector spin density \( S(r) \) (time-odd), the vector spin-kinetic density \( T(r) \) (time-odd), the vector current density \( j(r) \) (time-odd), the tensor spin-current density \( \vec{J}(r) \) (time-even), and the vector tensor-kinetic density \( F(r) \) (time-odd). The subscripts \( \mu, \nu \) indicate the components of the Cartesian co-ordinates. It can also be convenient to define a (time-even) spin-orbit density as the anti-symmetric part of \( J_{\mu \nu} \) [76, 78]

\[ J_\kappa(r) = \sum_{\mu \nu} \epsilon_{\kappa \mu \nu} J_{\mu \nu}(r). \]  

(2.28)
2.3 The Hartree-Fock Method with Skyrme's Effective Interaction

When performing static Hartree-Fock calculations for even-even nuclei, the time-odd densities have no effect upon the result, so are generally neglected [42]. This vastly simplifies the form of the energy density functional by assuming that if a state is occupied, so is its time-reversed partner. For time-dependent calculations of even-even nuclei, however, the minimum densities required to allow the energy density functional to conserve Galilean invariance under local gauge transformations include some time-odd terms [76] (also see Appendix A). The minimum densities required to investigate the dynamics of even-even nuclei within time-dependent Hartree-Fock are the particle density \( \rho(r) \), the spin density \( S(r) \), the current density \( j(r) \), the spin-orbit density \( J(r) \), and the kinetic density \( \tau(r) \) [57].

2.3.2 Constructing the Energy Density Functional

Each contribution to the energy density functional given by Eq. (2.24) can be written in terms of the local densities given by Eq. (2.27). The contribution from the kinetic term is given by:

\[
E_{\text{kinetic}} = \frac{\hbar^2}{2m} \int \tau(r) dr .
\] (2.29)

The Coulomb term depends upon the charge density of the system, although it is common practice to approximate the charge density with the proton density \( \rho_p(r) \). The Coulomb term contains a local part given by [57]

\[
E_{\text{Coul}}^{\text{direct}} = \frac{e^2}{2} \int \int \frac{\rho_p(r) \rho_p(r')}{|r - r'|} dr dr',
\] (2.30)

and an approximation of the non-local exchange part is given by [79]:

\[
E_{\text{Coul}}^{\text{exchange}} = -\frac{3e^2}{4} \left( \frac{3}{\pi} \right)^{\frac{1}{3}} \int \rho_p(r) \frac{4}{3} d^3 r .
\] (2.31)

The total Coulomb contribution to the energy density functional \( E_{\text{Coul}} \) is the sum of the direct and exchange Coulomb terms.

The Skyrme potential contains a direct and exchange term:

\[
\hat{v}_{\text{skyrme}} = \langle ij | \hat{v} | ij \rangle - \langle ij | \hat{v} | ji \rangle = \langle ij | \hat{v} (1 - P_{i\leftrightarrow j}) | ij \rangle .
\] (2.32)

The exchange \( P_{i\leftrightarrow j} \) is accounted for by inserting a term containing the exchange operators \( (1 - \hat{P}_M \hat{P}_P \hat{P}_q) \). The Majorana operator \( \hat{P}_M \) applies a spatial exchange, the isospin operator
$\hat{P}_q$ applies an exchange of isospin between the wave functions, and the spin exchange operator $\hat{P}_\sigma$ exchanges the spins (Eq. (2.22)). The Majorana operator has a value of $+1$ if the power of the momentum operator is even, or $-1$ if it is odd. The isospin operator $\hat{P}_q$ is equivalent to the term $\delta_{q_1q_2}$, $q \in \{ p, n \}$, forbidding mixing of isospin states. As an aside, some modern studies have been dedicated to the construction of an isospin invariant Energy Density Functional framework [80].

The contribution to the EDF from the Skyrme potential term can therefore be determined by:

$$E_{\text{skyrme}} = \frac{1}{2} \sum_{i,j} \int \varphi_i^*(r_1') \varphi_j^*(r_2') \hat{v}_{\text{skyrmc}}(r_1', r_2')(1 - \hat{P}_M \hat{P}_\sigma \hat{P}_q) \times \varphi_i(r_1) \varphi_j(r_2) \, dr_1 dr_2 dr_1' dr_2' \mid _{r_1=r_2=r_1'=r_2'}.$$  \tag{2.33}

Each term of the Skyrme interaction (Eqs. (2.20) and (2.23)) must be inserted and evaluated, and then the full energy contribution can be taken as the sum of the individual terms. The individual contributions must be evaluated before the limit is taken.

Taking into account only the minimum densities required to describe the ground state and dynamics of even-even systems, the Skyrme energy functional can be written explicitly. It is common practice [57, 61] to reformulate the resulting terms using the coefficients defined by

$$b_0 = t_0 \left( 1 + \frac{1}{2} x_0 \right) \quad b'_0 = t_0 \left( \frac{1}{2} + x_0 \right)$$

$$b_1 = \frac{1}{4} \left[ t_1 \left( 1 + \frac{1}{2} x_1 \right) + t_2 \left( 1 + \frac{1}{2} x_2 \right) \right] \quad b'_1 = \frac{1}{4} \left[ t_1 \left( \frac{1}{2} + x_1 \right) - t_2 \left( \frac{1}{2} + x_2 \right) \right]$$

$$b_2 = \frac{1}{8} \left[ 3t_1 \left( 1 + \frac{1}{2} x_1 \right) - t_2 \left( 1 + \frac{1}{2} x_2 \right) \right] \quad b'_2 = \frac{1}{8} \left[ 3t_1 \left( \frac{1}{2} + x_1 \right) + t_2 \left( \frac{1}{2} + x_2 \right) \right]$$

$$b_3 = \frac{1}{4} t_3 \left( 1 + \frac{1}{2} x_3 \right) \quad b'_3 = \frac{1}{4} t_3 \left( \frac{1}{2} + x_3 \right)$$

$$b_4 = \frac{1}{2} W_0 \quad b'_4 = \frac{1}{2} W'_0,$$  \tag{2.34}

which allows the contributions to the energy density functional to be separated into the following terms:

$$E_0 = \int \left( \frac{b_0}{2} \rho^2 - \frac{b'_0}{2} \sum_q \rho^2_q \right) \, dr,$$  \tag{2.35}

$$E_1 = \int \left( b_1 [\rho \tau - j^2] - b'_1 \sum_q [\rho_q \tau_q - j_q^2] \right) \, dr.$$  \tag{2.36}
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\begin{align*}
E_2 &= \int \left( -\frac{b_2}{2} \rho \Delta \rho + \frac{b'_2}{2} \sum_q \rho_q \Delta \rho_q \right) \, dr , \\
E_3 &= \int \left( \frac{b_3}{3} \rho^{\alpha+2} - \frac{b'_3}{3} \rho^\alpha \sum_q \rho^2_q \right) \, dr , \\
E_{ls} &= \int \left( -b_4 [\rho \nabla \cdot \mathbf{J} + \mathbf{S} \cdot (\nabla \times \mathbf{j})] - b'_4 \sum_q [\rho_q \nabla \cdot \mathbf{J}_q + \mathbf{S}_q \cdot (\nabla \times \mathbf{j}_q)] \right) \, dr .
\end{align*}

Some Skyrme forces add further refinements to the definition of the energy density functional, for example the SkI subset attempt to add isospin freedom to the spin-orbit term by giving different weightings to the proton and neutron states \cite{81} (i.e. \( b_4 \neq b'_4 \)).

The overall result of adopting the Skyrme interaction is that the total energy of the system as given in Eq. (2.24) can be expressed purely in terms of local densities, which leads to the apparent equivalence between SHF and DFT.

2.3.3 The Hartree-Fock Hamiltonian

The Hartree-Fock single-particle Hamiltonian can be defined from the variation \cite{57, 76}

\begin{equation}
\frac{\delta}{\delta \varphi_\alpha} E = \hat{h} \varphi_\alpha ,
\end{equation}

where \( E = E[\varphi] \) describes the energy density functional of the system. As the wave functions completely define the local densities of the system, it is customary to separate the single-particle Hamiltonian into the mean-field potentials that arise from variations of the energy functional with respect to the different densities \cite{34, 76}. Reference \cite{76} presents the resulting Hamiltonian for the case of inclusion of all the time-odd and time-even densities (not inclusive of tensor terms, see e.g. \cite{77}), whereas for the TDHF code Sky3D \cite{57} the energy density functional used is one which includes the minimum terms for the time evolution of even-even nuclei.

The single-particle Hamiltonian (used in Sky3D) can be written \cite{57}:

\begin{equation}
\hat{h}_q = U_q(r) - \nabla \cdot [B_q(r) \nabla] + i \tilde{W}_q(r) \cdot (\sigma \times \nabla) + \Sigma_q(r) \cdot \sigma - \frac{i}{2} \left[ \nabla \cdot \mathbf{I}_q(r) + \mathbf{I}_q(r) \cdot \nabla \right] .
\end{equation}
The first term, $U_q$, can be obtained by performing the variation of $E$ with respect to the particle density $\rho$ (dropping the argument $r$ of the densities here for brevity):

$$
U_q = b_0 \rho - b'_0 \rho_q + b_1 \tau - b'_1 \tau_q - b_2 \Delta \rho + b'_2 \Delta \rho_q \\
+ b_3 \frac{\alpha+2}{4} \rho^{\alpha+1} - b'_3 \frac{\alpha}{2} \rho^\alpha \rho_q - b_3 \rho^\alpha \sum_{q'} \rho_{q'}^2 \\
- b_4 \nabla \cdot J - b'_4 \nabla \cdot J_q .
$$

(2.42)

Next, by performing the variation with respect to the kinetic density $\tau$, the effective mass term $B_q$ can be written:

$$
B_q = \frac{\hbar^2}{2m_q} + b_1 \rho - b'_1 \rho_q.
$$

(2.43)

The variation with respect to the spin-orbit density $J$ yields:

$$
\hat{W}_q = b_4 \nabla \rho + b'_4 \nabla \rho_q .
$$

(2.44)

By considering the variation of the energy with respect to the time-odd current density $j$ the term

$$
I_q = -2b_1 j + 2b'_1 j_q - b_4 \nabla \times S - b'_4 \nabla \times S_q
$$

is given. Finally, the variation with respect to the spin density $S$ gives:

$$
\Sigma_q = -b_4 \nabla \times j - b'_4 \nabla \times j_q .
$$

(2.46)

### 2.3.4 Implementation of the Skyrme Hartree-Fock Method

The Skyrme Hartree-Fock equation boils down to a non-linear differential equation, and it is solved for the ground state Slater determinant. This Slater determinant is comprised of the single-particle wave functions $\varphi_i$ (with energy eigenvalues $\epsilon_i$). The single-particle wave functions in turn define the densities, which define the mean fields, which define the single-particle Hamiltonian. Therefore, the equation has to be solved in a self-consistent manner.

Typically, the starting point is a guess of the single-particle wave functions, often assuming the form of a harmonic oscillator. From this, the initial density and mean-field is defined. In the solver Sky3D, the Hartree-Fock equation is solved using a gradient step method accelerated by kinetic energy damping [82]. The wave functions are initialised in a 3D Cartesian grid at iteration $n = 0$, and then are driven to the ground state by the operation:

$$
\varphi^{(n+1)}_\alpha = \hat{\mathcal{O}} \left[ \varphi^{(n)}_\alpha - \frac{\delta}{\hat{T} + E_0} \left( \hat{h}^{(n)} - \langle \varphi^{(n)}_\alpha | \hat{h}^{(n)} | \varphi^{(n)}_\alpha \rangle \right) \varphi^{(n)}_\alpha \right] .
$$

(2.47)
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Here, $\tilde{O}$ is an orthogonalisation operation; at each iteration $n$ a Gram-Schmidt orthogonalisation is performed upon all the single-particle wave functions. The operator $\tilde{T}$ is the kinetic energy operator, $\delta$ is a parameter determining the step size (typically in the range 0.1 to 0.8) and $E_0$ is the damping regulator. The damping regulator should be of the order of the depth of the mean-field potential. The self-consistency of the problem is apparent here; $\varphi^{(n+1)}$ is defined by the Hamiltonian $\tilde{h}^{(n)}$, and from $\varphi^{(n+1)}$ new densities are built, which are used to calculate the new mean fields, which determine $\tilde{h}^{(n+1)}$.

The convergence of the scheme is tested numerically at each iteration step. Within Sky3D, the convergence is measured by considering the energy variance of the single-particle states, which is a way to test how ‘good’ the $\epsilon_i$ are as eigenvalues of the Hamiltonian. This criterion can be defined by

$$\Delta \epsilon = \sqrt{\frac{\sum_\alpha \Delta \epsilon^2_\alpha}{\sum_\alpha \langle \varphi_\alpha | \varphi_\alpha \rangle}}, \quad (2.48)$$

where

$$\Delta \epsilon^2_\alpha = \langle \varphi_\alpha | \tilde{h}^2 | \varphi_\alpha \rangle - \epsilon^2_\alpha$$
$$\epsilon_\alpha = \langle \varphi_\alpha | \tilde{h} | \varphi_\alpha \rangle. \quad (2.49)$$

Typical calculations converge with $\Delta \epsilon$ in the order of $10^{-4}$ to $10^{-5}$.

There are, of course, other ways to determine the ground state solution. The imaginary time method is another historically well used technique for Hartree-Fock iteration schemes [83]. Further techniques may be used to accelerate the convergence of the solution, from simply linearly mixing the densities from iteration $n$ to $n + 1$, to sophisticated mixing techniques, such as Broyden’s method [84]. As an aside, the Broyden method has been proven to be remarkably successful when performing calculations in a harmonic oscillator basis [47].

We have tried to implement this method within Sky3D. We have found that, when working in a 3D Cartesian basis, the method would not provide any additional speed-up. This is in contrast to our findings in a spherically-symmetric code. All in all, this suggests that the 3D geometry is incompatible with Broyden’s mixing. To further test this hypothesis, we used a basic 3D Hartree-Fock code using a simplified version of the Skyrme nucleon-nucleon interaction (containing only the $t_0$ and $t_3$ terms). Mixing either the densities, mean fields or both between iterations showed no improved convergence compared to the case of a simple linear mixing.
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As a final comment, Hartree-Fock minimisation schemes find local extrema; depending on the initial configuration of the harmonic oscillator guesses, the final solution may correspond to an isomeric state rather than the ground state solution. This may not be an issue if these states are of interest, but to determine the global minimum it is best to perform several Hartree-Fock calculations with the initial harmonic oscillator starting in an oblate or prolate configuration.

2.4 Constrained Hartree-Fock

There can be many instances where it may be desirable not to calculate the global Hartree-Fock ground state, but the Hartree-Fock minimum subject to certain conditions. For example, techniques such as the Generator Co-ordinate Method can be implemented upon a superposition of mean-field states with different shape degrees of freedom imposed via constraints [67]. Specifically to this work, investigating increasingly deformed Hartree-Fock states up to the point of fission is a reasonable method to obtain initial states for time-dependent investigations.

Constrained Hartree-Fock (CHF) is a constrained optimisation problem, where various methods can be applied to impose the auxiliary conditions to the minimisation in the iteration scheme. The simplest Linear Constraint Method (LCM) is the technique of Lagrange multipliers [48]. The minimisation of the expectation energy of the energy functional $E[\varphi]$ can be subjected to a generic constraint on the expectation value of an operator $\hat{\zeta}$. Therefore, the variation of

$$\bar{E} = E + \lambda (\langle \hat{\zeta} \rangle - \zeta^0)$$

(2.50)

can be considered. The Lagrange multiplier will drive the expectation value towards the desired value, $\zeta^0$, as the Hartree-Fock equation is solved.

The technique can fail to enforce constraints in nuclear Hartree-Fock calculations [48], requiring the use of refined methods. Another technique, as discussed in Refs. [85, 86] utilises the method of Lagrange multipliers, but allows the constraining conditions to be violated throughout the iterative procedure up until the final $\lambda$ is determined. The technique aims to reduce numerical instabilities and improve convergence upon the desired value of the constraint, and has been successful implemented in nuclear Hartree-Fock codes [87, 88].
Another historically well-used technique for implementing constraints in nuclear Hartree-Fock theory is the Quadratic Penalty Method (QPM) [21, 45, 48], which minimises the energy of the system subject to

$$\bar{E} = E + c (\langle \hat{\zeta} \rangle - \zeta^0)^2.$$  \hspace{1cm} (2.51)

The parameter $c(>0)$ is known as the penalty parameter. The energy of the system will be a minimum when $\langle \hat{\zeta} \rangle = \zeta^0$, and this is enforced as $c$ tends to $\infty$, allowing convergence on the exactly desired value of $\langle \hat{\zeta} \rangle$. Unfortunately, for very large values of $c$ the system can become numerically unstable; in many cases the QPM cannot enforce the exactly specified auxiliary conditions.

A third technique, the so-called Augmented Lagrangian Method (ALM) [89, 90] has been demonstrated in Ref. [48] to be a powerful method to enforce constraints in nuclear DFT calculations, both providing numerical stability and the ability to converge precisely upon the specified value of the constraint. This method will be summarised and its integration into the solver Sky3D discussed in the following Sections.

### 2.4.1 The Augmented Lagrangian Method

The Augmented Lagrangian Method, as presented in Ref. [48], can be viewed as a hybrid of the linear and quadratic constraint methods. The energy density functional is augmented to contain extra terms corresponding to the constraint upon the expectation of the operator $\hat{\zeta}$;

$$\bar{E} = E + \lambda (\langle \hat{\zeta} \rangle - \zeta^0) + c (\langle \hat{\zeta} \rangle - \zeta^0)^2 = \langle \Phi | \hat{h} | \Phi \rangle + \lambda \left( \langle \Phi | \hat{\zeta} | \Phi \rangle - \zeta^0 \right) + c \left( \langle \Phi | \hat{\zeta} | \Phi \rangle - \zeta^0 \right)^2,$$  \hspace{1cm} (2.52)

To compute the minimum energy subject to the desired value of the constraint, the variational derivative can be taken. This allows the modified single-particle Hamiltonian to be expressed in terms of the original ('unconstrained') Hamiltonian, and an additional part which drives the solution towards the desired value,

$$\frac{\delta}{\delta \varphi_a} \bar{E} = \hat{h}'|\varphi_a\rangle = \hat{h}|\varphi_a\rangle + \lambda \langle \hat{\zeta}|\varphi_a\rangle + 2c \left( \langle \varphi_a | \hat{\zeta} | \varphi_a \rangle - \zeta^0 \right) \hat{\zeta}|\varphi_a\rangle$$

$$= \hat{h}|\varphi_a\rangle + 2c (\zeta - \zeta^0(\lambda)) \hat{\zeta}|\varphi_a\rangle,$$  \hspace{1cm} (2.53)

where $\zeta^0(\lambda) = \zeta^0 - \lambda/2c$. The Lagrange multiplier $\lambda$ is updated iteratively as

$$\lambda^{k+1} = \lambda^k + 2c (\zeta - \zeta^0).$$  \hspace{1cm} (2.54)
Throughout the iterative procedure, the multiplier $\lambda^k$ will converge, resulting in a stationary value of the energy subject to the desired constraint. An initial value of $\lambda^0 = 0$ is often adopted. Reference [90] also contains an algorithm for updating the stiffness parameter $c$.

As with other constraint methods, in principle the method can be generalised to enforce an arbitrary number of auxiliary conditions, which can be added into the expression for the mean-field given in Eq. (2.53).

### 2.4.2 Implementation of the ALM in a Nuclear SHF Solver

The ALM can be implemented in a straightforward manner into a Hartree-Fock iteration scheme. The algorithms are presented in Refs. [48] and [90]. The ALM single-particle Hamiltonian $\hat{h}'$, as given in Eq. (2.53), can be substituted in place of the Hartree-Fock Hamiltonian into the iteration scheme (Eq. (2.47)). Once the Hamiltonian has been applied to all the single-particle wave functions for a single iteration, the Lagrange parameter is updated via Eq. (2.54). Convergence is defined by the simultaneous conditions that both the fluctuations in the single-particle energies and difference between the desired and current values of the constrained observable are sufficient small.

Initial values for $\lambda$ and $c$ can be chosen arbitrarily. The parameter $\lambda$ was always chosen to start from 0. The parameter $c$ requires a little more fine tuning: too large values of $c$ can result in numerical instabilities if the difference between the desired and current expectation value of the constrained observable is large. To overcome this, $c$ may be increased iteratively as the calculation converges upon the desired value. An algorithm for this purpose is detailed in Ref. [90].

Figure 2.1 shows an example of the evolution of various observables and parameters associated with enforcing a constraint upon the quadrupole deformation parameter $\beta_{20}$ (see Appendix B for the definition) for the nucleus $^{46}$Ti. The calculation starts from a state with $\beta_{20} = 1.21$, and the desired value is 1.24. An oscillatory behaviour is observed as the shape of the nucleus converges; this is due to linear mixing of the scalar densities between iterations. The panel i) shows the evolution of the expectation value of $\beta_{20}$ throughout the iterative procedure, and the panel ii) displays the absolute magnitude of the difference between the expectation and the desired value at each step. Panel iii) displays the convergence of the
2.5 Correlations Beyond the Static Mean-Field

The Hartree-Fock approach provides only the lowest order microscopic description of the nucleus. Due to the Slater determinant approximation of the many-body wave function, the method breaks symmetries and does not include various correlations that must be considered to truly describe the ground-state correlations of the atomic nucleus [61, 67]. This does not imply that the Hartree-Fock method is intrinsically flawed, but rather that the method can be considered as a starting point for methods that go beyond the mean-field approximation.

In the context of this work, static solutions to the Hartree-Fock equation are required for initial configurations to perform dynamic calculations. Therefore, the complex procedure of

Figure 2.1: Various parameters measured applying the ALM to constrain the quadrupole deformation parameter $\beta_{20}$ in a Hartree-Fock minimisation for the nucleus $^{46}$Ti. See text for more details.

Lagrange parameter $\lambda$ used to enforce the constraint, and panel iv) shows the value of the stiffness constant $c$ throughout the process, which starts from a value of 50.

The shape is observed to oscillate around the desired expectation value. These oscillations are quickly damped by the increase of the $c$ parameter, and a converged solution is obtained after about 900 iterations.
restoring symmetries and adding correlations to these static states will be neglected. The only correlation beyond the mean-field that will be considered for the static calculations is short-range pairing correlations.

2.5.1 Pairing Correlations

Pure Slater determinant states are appropriate for describing doubly-magic, spherical nuclei [61], where single-particle levels are filled below the Fermi energy, and empty above. For these nuclei, there is a large energy difference between the highest occupied state and the next empty one. However, when investigating nuclei away from these shell closures, the concept of quasiparticles is more appropriate: nucleons can in principle occupy any level. Occupation is determined as a probability of the single-particle state being a particle (filled) or hole (empty) state. Due to the fact the energy levels near the Fermi surface are nearly degenerate in energy, there can be an appreciable scattering of occupation probability, so pairing is required to obtain a unique ground state [21, 42, 61, 67]. Inclusion of pairing in a Hartree-Fock scheme therefore allows the inclusion of some particle-particle correlations, which the pure Slater determinant cannot describe.

The most general pairing mean-field scheme is Hartree-Fock Bogliubov (HFB), which introduces the concept of quasiparticle states which are related to the single-particle states by the Bogoliubov transformation [21, 42, 67]. This scheme intrinsically includes pairing effects, however the HFB method is computationally expensive to apply. The Bardeen-Cooper-Schrieffer (BCS) pairing scheme is an approximation of full HFB, and is a popular technique for modelling the short-range pairing correlations in deformed nuclei. The approach, as first described by Bardeen, Cooper and Schrieffer to describe superconductivity [91], was applied to nuclear structure physics with considerable success by Belyaev in 1959 [92]. The BCS approach simplifies the pairing scheme by forcing the pairing potential to be diagonal in the basis of the eigenstates of the mean-field Hamiltonian [67].

Recent investigations have shown calculations using the BCS approximation can quantitatively include most of the information HFB provides. Reference [93] demonstrates that BCS pairing with a finite-range pairing potential is a good substitution for HFB, and Ref. [94] discusses simply renormalising the BCS pairing field to capture most of the information given by HFB employed in the relativistic mean-field model.
Within the scope of this thesis, pairing has been described entirely within the BCS scheme, which will be discussed in the following Sections.

### 2.5.2 The BCS Pairing Model

As mentioned previously, the need to incorporate a pairing scheme arises as single-particle wave functions making up a pure Slater determinant are no longer an appropriate basis for the description of deformed nuclei. Within the BCS scheme, it is assumed that pairs of nucleons which are symmetric under time reversal (known as Kramers degenerate) can be coupled by a short-range pairing force into states of zero total angular momentum $j$. In other words, the nucleons pair into states within an angular momentum $j$ shell with projection quantum numbers $m_j$ and $-m_j$. The BCS state can be constructed from the vacuum by creating pairs denoted by $k$ and $\bar{k}$. These states are time reversed partners of one other, corresponding to the angular momentum projected partners $m_j$ and $-m_j$. The states $|k, \bar{k}\rangle$ make up the whole single-particle space:

$$|\text{BCS}\rangle = \prod_{k>0} (u_k + v_k \hat{a}_k \hat{a}^\dagger_k |0\rangle).$$

Here, $k$ is used to label the single-particle states, and $\bar{k}$ the time reversed partner. The product only runs over positive $k$, as for each $k > 0$ there exists the conjugate state $\bar{k} < 0$. The probability of occupation of the state $|k, \bar{k}\rangle$ can be interpreted as $|v_k|^2$, whereas $|u_k|^2$ is associated with the probability the state remains empty. This interpretation as a probability arises from the normalisation of the BCS state:

$$\langle \text{BCS} | \text{BCS} \rangle = \langle 0 | \prod_{k>0} (u_k + v_k \hat{a}_k \hat{a}^\dagger_k) \prod_{k'>0} (u_{k'} + v_{k'} \hat{a}_{k'} \hat{a}^\dagger_{k'}) | 0 \rangle = \prod_{k>0} (u_k^2 + v_k^2)$$

$$= \prod_{k>0} (u_k^2 + v_k^2) = 1.$$ 

The BCS state has some qualities that makes it different from a pure Slater determinant. Notably, the BCS wave function does not conserve particle number. This can be realised from considering the mean square deviation of the expectation value of the number, which can be defined by

$$\Delta(N)^2 = \langle \text{BCS} | \hat{N}^2 | \text{BCS} \rangle - \langle \text{BCS} | \hat{N} | \text{BCS} \rangle^2,$$
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and can be evaluated to $4 \sum_{k>0} u_k^2 v_k^2$ [42]. This uncertainty arises from partially occupied states, that is, states where $u_k$ or $v_k$ are not equal to 0 or 1, respectively.

Taking into account these properties of the BCS state, as with the discussion of the Hartree-Fock method in Sec. 2.2, where the many bodied wave function was a pure Slater determinant state, a Hamiltonian containing one and two-body interactions may be considered. It is customary to to write the Hamiltonian in terms of a single-particle part (following the Hartree-Fock approximation), and a pairing field which is described by a separate pairing interaction, $\hat{v}_{\text{pair}}$. In this case, the following expectation value of the BCS Hamiltonian can be written [21]:

$$\langle \text{BCS} | \hat{H}_{\text{BCS}} | \text{BCS} \rangle = \sum_{k \geq 0} \langle k | \hat{t} | k \rangle v_k^2 + \frac{1}{2} \sum_{k, k' \geq 0} \langle kk' | \hat{v} | k' k \rangle v_k^2 v_{k'}^2 + \sum_{k, k' > 0} \langle k \bar{k} | \hat{v}_{\text{pair}} | k' \bar{k}' \rangle u_k v_{k'} u_k v_{k'}.$$  \hspace{1cm} (2.59)

The first two terms run over all the configuration space, acting only upon particle states (which have occupation probability $v_k^2$). The last two-body term acts upon pairs, therefore the sum runs only over positive $k$. Considering the expectation value of the BCS Hamiltonian in this form allows the Hartree-Fock and BCS equations to be coupled and solved (commonly referred to as HF+BCS). A simple interaction is often assumed in the pairing channel for convenience when performing HF+BCS calculations [95].

As with the Slater determinant case, the variation of the expectation value of the BCS Hamiltonian can be taken to find the ground state energy. As the basis was seen to allow violations of particle number, a Lagrange multiplier $\lambda$ must be included in the BCS Hamiltonian to ensure particle number is conserved. The variation can be replaced by a partial derivative as the BCS wave function is defined by the $v_k$:

$$0 = \delta \langle \text{BCS} | \hat{H}_{\text{BCS}} - \lambda \hat{N} | \text{BCS} \rangle = \frac{\partial}{\partial v_k} \langle \text{BCS} | \hat{H}_{\text{BCS}} - \lambda \hat{N} | \text{BCS} \rangle.$$  \hspace{1cm} (2.60)

Explicitly performing the variation (see, e.g. Refs. [21, 42]) gives several results. Firstly, the BCS equation can be written:

$$2 \epsilon_k u_k v_k + \Delta_k (v_k^2 - u_k^2) = 0$$  \hspace{1cm} (2.61)
where the single-particle energies obtained from the Hartree-Fock scheme \( \epsilon_k^0 \) are modified:

\[
\epsilon_k = \epsilon_k^0 - \lambda. \tag{2.62}
\]

This allows the Lagrange parameter \( \lambda \) to be interpreted as the Fermi energy.

The gap parameter \( \Delta_k \) is given by:

\[
\Delta_k = -\sum_{k' > 0} \langle k\bar{k} | \hat{v}_{\text{pair}} | k'\bar{k}' \rangle u_{k'} v_{k'}. \tag{2.63}
\]

Finally, expressions for the probability of the pair state \((k, \bar{k})\) as either a particle or hole state may be written:

\[
v_k^2 = \frac{1}{2} \left( 1 - \frac{\epsilon_k}{\sqrt{\epsilon_k^2 + \Delta_k^2}} \right), \tag{2.64}
\]

\[
u_k^2 = \frac{1}{2} \left( 1 + \frac{\epsilon_k}{\sqrt{\epsilon_k^2 + \Delta_k^2}} \right). \tag{2.65}
\]

Figure 2.2: Occupation probabilities as a function of \( \epsilon_k \) for three different gap parameters. The non pairing case (\( \Delta = 0 \)) shows a sharp cutoff of occupation at the Fermi energy. The central and right panel present two cases of pairing interactions; as the pairing gap parameter is increased, the occupation of states about the Fermi energy transitions to zero more gradually.

Figure (2.2) shows an example of the occupation probability of a single-particle state as a function of the single-particle energy \( \epsilon_k \) for different pairing gaps, for the case of a simplified ‘pure pairing’ interaction \cite{21}. In this case, the pairing gap \( \Delta \) is fixed for each state \( k \). For the case of \( \Delta = 0 \), there is a sharp cutoff of occupation at the Fermi energy, which is equivalent to a pure Slater determinant state. The transition from particle to hole states is governed by the magnitude of \( \Delta \), requiring comparison to nuclear structure data to fit the strength of the pairing interaction. In the application of pairing in the work for this thesis, however, the pairing gap \( \Delta_k \) is solved explicitly for each \( k \) following Eq. (2.63).
2.5.3 Implementing BCS Pairing in a Nuclear SHF Solver

When implementing BCS pairing within the Hartree-Fock framework, the problem requires solutions to the BCS equation to determine the occupation amplitudes $v_k^2$ (Eq. (2.64)), the pairing gaps $\Delta_k$ (Eq. (2.63)) and the single-particle energies obtained from subtracting the Fermi energy $\lambda$ from the Hartree-Fock single-particle energies (Eq. (2.62)). These equations are non-linear, and can be solved by iteration [57] for proton and neutron states separately.

The densities needed to describe the energy density functional (as discussed in Sec. 2.3.1) in this basis can be modified to account for the pairing occupation $v_k^2$ of each single-particle state. For example, the particle density matrix becomes (the index $i$ here running over all single-particle states):

$$\rho(r, r') = \sum_{i} v_i^2 \phi^*_i(r') \phi_i(r).$$  \hspace{1cm} (2.66)

In practice, the sum cannot be performed over an infinite number of states, therefore the size of the pairing space must be chosen carefully. Using a realistic pairing interaction, the probability of occupation scattering into states well above the Fermi energy will reduce towards zero. This justifies the energy-dependent cutoff of level occupation adopted in some calculations [67, 96]. If using a simplified zero-range pairing interaction, as is often adopted, a cutoff of occupation is needed as scattering can occur over any energy range.

From an alternative perspective to an energy-dependent cutoff, the scattering of occupation across a shell closure will be greatly reduced due to the large energy differences of the levels above and below the closure. The size of the pairing space $N_{\text{max}}$ for proton and neutron states can be chosen reasonably from the approximate relation [57, 67]:

$$N_{\text{max},q} = N_q + \frac{5}{3} N_q^{2/3}. \hspace{1cm} (2.67)$$

Here, $N_q$ refers to the number of protons ($Z$) or neutrons ($N$) in the nucleus being investigated.

When including pairing effects, the energy contribution to the SHF energy density functional is dependent upon the form assumed for the pairing potential. A typical choice within the Skyrme Hartree-Fock framework is to use a zero-range pairing interaction, ensuring that the terms are of the same form of a Skyrme interaction with only $t_0$ and $t_3$ terms [61, 67, 95, 96]. The pairing interaction using the so-called density-dependent delta
interaction (DDDI) is a function of the nuclear density $\rho(r)$, and is given by

$$\hat{v}_{\text{pair}}(r_1, r_2) = \frac{V_{0,q}}{2} (1 - \hat{P}_\sigma) \left[ 1 - \left( \frac{\rho(r)}{\rho_0} \right) ^\gamma \right] \delta(r_1 - r_2),$$

(2.68)

where a different strength parameter $V_{0,q}$ can be used for particles of different isospin $q$, and may be adjusted for different locations throughout the nuclear landscape. The nuclear saturation density $\rho_0$ is typically taken to be 0.16 fm$^{-3}$, but in the case where $\rho_0 \to \infty$, the volume delta interaction (VDI) is recovered. The exponent $\gamma$ is usually taken to be equal to unity. Some modern Skyrme parametrizations propose use of a pairing interaction which is a mix of the DDDI and VDI interaction [97, 98, 99].

The energy contribution from the pairing term can be evaluated analogously to the energy contribution from the Skyrme interaction (Eq. (2.33)). By defining the pseudo-pairing density,

$$\chi(r, r') = \sum_{i=1}^{N_{\text{max},q}} u_i v_i \bar{\varphi}_i(r') \varphi_i(r),$$

(2.69)

where the sum is performed over time-reversed partners [57], and neglecting time-odd contributions to the energy density functional (as by definition the pairs are symmetric under time reversal), the energy contribution can be evaluated to [67, 96]:

$$E_{\text{pair}} = \frac{1}{4} \sum_{q=p,n} \int \chi_q^2(r) V_{0,q} \left[ 1 - \left( \frac{\rho(r)}{\rho_0} \right) ^\gamma \right] dr.$$  \hspace{1cm} (2.70)

where $\chi_q(r)$ is the local pairing density. When accounting for pairing correlations, this term must be added into the energy density functional given by Eq. (2.24).

### 2.6 Time-Dependent Hartree-Fock

In general, a non-relativistic system will obey the time-dependent Schrödinger equation as it evolves in time:

$$i\hbar \frac{d|\Psi(t)\rangle}{dt} = \hat{H}|\Psi(t)\rangle.$$  \hspace{1cm} (2.71)

The time evolution of a many-body wave function $\Psi$, which spans the entire Hilbert space, from $t = t_0$ to $t = t_1$ can therefore be deduced by applying the time evolution operator to the state $|\Psi(t = t_0)\rangle$:

$$|\Psi(t_1)\rangle = e^{-i\hat{H}t/\hbar}|\Psi(t_0)\rangle.$$  \hspace{1cm} (2.72)

The time-dependent Schrödinger equation can only be solved exactly for a few special cases, motivating the need for approximations. The time-dependent Hartree-Fock equation was
first written down by Dirac in the 1930s [100] to describe atoms by considering the time evolution of the self-consistent mean-field. The procedure was applied to nuclear dynamics in the 1970s [76], but early calculations suffered from computational limitations, requiring spatial symmetries to be assumed. Modern TDHF solvers can evolve Slater determinants in three dimensional, symmetry unrestricted frameworks [57, 58, 59].

As with the stationary Hartree-Fock equation, a fundamental assumption in time-dependent Hartree-Fock is that the many-body wave function is the Slater determinant \( \Phi(\mathbf{r}, t) \). The variational principle can be applied to obtain the equations of motion for the path of least action to describe the time evolution of such a system. The action can be written:

\[
S_{t_0, t_1}[\Phi(\mathbf{r}, t)] = \int_{t_0}^{t_1} dt \langle \Phi(\mathbf{r}, t) | \left( i\hbar \frac{d}{dt} - \hat{h} \right) | \Phi(\mathbf{r}, t) \rangle.
\] (2.73)

Due to the Slater determinant approximation, the two-body, three-body,..., \( N \)-body density matrices can be expressed in terms of the one-body density. For a Hamiltonian containing one and two-body terms, the two-body density can be written in terms of one-body density matrices, plus a correlation term which describes two-body collisions beyond the mean-field approximation. Within TDHF, by definition the two-body correlation term is equated to zero. The technique, however, does include some correlations that go beyond the concept of a mean-field. Although two-body collisions (two-body ‘friction’) are neglected in TDHF, TDHF does include what is known as one-body friction, that is, collisions between particles and the wall of the mean-field [21].

As with Hartree-Fock, time-dependent Hartree-Fock calculations may be performed using the Skyrme effective interaction [76, 101]. Analogously to the static case, one can draw an equivalence between time-dependent Hartree-Fock and time-dependent Density Functional Theory.

### 2.6.1 Derivation of the Time-Dependent Hartree-Fock Equation from the Variation Principle

The time-dependent Hartree-Fock equation may be recovered by minimising the action \( S \) defined by Eq. (2.73) (see, e.g. Ref. [101]). This is equivalent to performing the variation \( \delta S = 0 \), applied with the boundary conditions \( \delta \Phi(t_0) = \delta \Phi(t_1) = 0 \). The time-dependent Hartree-Fock equation will therefore yield deterministic trajectories, which will always follow the path of least action.
As with the static Hartree-Fock equation, the variation can be performed with respect to
the single-particle wave function \( \varphi_\alpha(x, t) \), or the conjugate \( \varphi_\alpha^*(x, t) \); the TDHF equation or
its conjugate will be recovered depending on the choice. The action can be written in the
form
\[
S = \int_{t_0}^{t_1} dt \left( i\hbar \sum_{i=1}^{N} \int d\mathbf{r} \varphi_i^*(\mathbf{r}, \tau) \frac{d}{d\tau} \varphi_i(\mathbf{r}, \tau) - E[\rho(\tau)] \right),
\]
(2.74)
where here the energy expectation \( E[\rho(\tau)] \) is written as a functional of the time-dependent
one-body density (which in turn is determined by the single-particle wave functions). The
time co-ordinate is denoted by \( \tau \). The variation of the action with respect to \( \varphi_\alpha^*(x, t) \) can be
performed:
\[
0 = \frac{\delta S}{\delta \varphi_\alpha^*(x, t)} = i\hbar \frac{d}{dt} \varphi_\alpha(x, t) - \int_{t_0}^{t_1} d\tau \frac{\delta E[\rho(\tau)]}{\delta \varphi_\alpha^*(x, t)}.
\]
(2.75)
The functional derivative of \( E[\rho(\tau)] \) can be rewritten using a change of variable,
\[
\frac{\delta E[\rho(\tau)]}{\delta \varphi_\alpha^*(x, t)} = \int \int d\mathbf{r} d\mathbf{r}' \frac{\delta E[\rho(\tau)]}{\delta \rho(\mathbf{r}, \mathbf{r}'; \tau)} \frac{\delta \rho(\mathbf{r}, \mathbf{r}'; \tau)}{\delta \varphi_\alpha^*(x, t)},
\]
(2.76)
and by writing the non-local density explicitly as
\[
\rho(\mathbf{r}, \mathbf{r}'; \tau) = \sum_{i=1}^{N} \varphi_i^*(\mathbf{r}', \tau) \varphi_i(\mathbf{r}, \tau),
\]
(2.77)
the variation of this density with respect to \( \varphi_\alpha^*(x, t) \) can be performed to yield
\[
\frac{\delta \rho(\mathbf{r}, \mathbf{r}'; \tau)}{\delta \varphi_\alpha^*(x, t)} = \varphi_\alpha(\mathbf{r}, t) \delta(\mathbf{r}' - \mathbf{x}) \delta(t - \tau).
\]
(2.78)
This results in Eq. (2.76) reducing to
\[
\frac{\delta E[\rho(\tau)]}{\delta \varphi_\alpha^*(x, t)} = \int d\mathbf{r} \frac{\delta E[\rho(t)]}{\delta \rho(\mathbf{r}, \mathbf{x}; t)} \varphi_\alpha(\mathbf{r}, t).
\]
(2.79)
The Hartree-Fock Hamiltonian can be substituted into the above relationship using:
\[
\hat{h}(\mathbf{r}, \mathbf{x}, t) = \frac{\delta E[\rho(t)]}{\delta \rho(\mathbf{r}, \mathbf{x}; t)}.
\]
(2.80)
This allows Eq. (2.75) to be written in the form of the time-dependent Hartree-Fock equation:
\[
i\hbar \frac{d}{dt} \varphi_\alpha(x, t) = \int d\mathbf{r} \hat{h}(\mathbf{r}, \mathbf{x}, t) \varphi_\alpha(\mathbf{r}, t).
\]
(2.81)
This equation describes the time evolution of the single-particle wave function \( \varphi_\alpha \) interacting
with the self-consistent mean-field. Adopting the zero-range Skyrme interaction allows the
TDHF equation to be written in terms of the local spatial co-ordinate \( \mathbf{r} \):
\[
i\hbar \frac{d}{dt} \varphi_\alpha(\mathbf{r}, t) = \hat{h}(t) \varphi_\alpha(\mathbf{r}, t).
\]
(2.82)
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Reference [21] details some properties of the TDHF equation. The total energy, orthogonality of the wave functions, product character (that is, the wave function is a Slater determinant) and the average expectation values of symmetry operators (such as particle number and linear and angular momentum) are conserved for all time. The fact that orthogonally is conserved is extremely convenient, allowing TDHF calculations to be performed using massively parallel computation techniques [57]. The TDHF equation produces deterministic trajectories as the wave functions evolve, and the wave functions display microscopic reversibility.

2.6.2 Time-Dependent Applications of Pairing Correlations

Several recent TDHF studies have been dedicated to investigating the effects due to the inclusion of time-dependent pairing correlations within time-dependent Hartree-Fock calculations. Full time-dependent Hartree-Fock-Bogoliubov (TDHFB) has been implemented for investigations of collective resonances [102, 103], whereas Canonical-basis time-dependent Hartree-Fock-Bogoliubov [104], or equivalently time-dependent Hartree-Fock coupled with time-dependent BCS pairing [105], has been implemented both for small (e.g. collective resonances [106, 107]) and large amplitude (e.g. fusion and particle transfer reactions [108, 109, 110]) TDHF calculations.

In this thesis, the fixed occupation amplitude (FOA) approximation has been assumed: the ground state is calculated using HF+BCS, then the occupations of the single-particle states are fixed at the value defined from the static solution throughout time evolution. The contribution to the energy functional from pairing is taken to be zero for the TDHF calculations.

The effects of time-dependent pairing upon the nuclear dynamics in comparison to adopting the FOA has been shown in some cases to be significant for large amplitude applications of TDHF. References [108, 109] show, for example, inclusion of time-dependent pairing reducing the fusion cross section in sample reactions. A study of the effect of time-dependent pairing upon fission dynamics within TDHF presents itself as a natural extension of this work.
2.6.3 Implementation of the Time-Dependent Hartree-Fock Method

The single-particle wave functions can be evolved in time via the TDHF equation (Eq. (2.81)) by writing it as the integral equation

\[ |\varphi_i(t + \Delta t)\rangle = \hat{T}(t, t + \Delta t)|\varphi_i(t)\rangle, \]

(2.83)

where \( \hat{T} \) is the time evolution operator:

\[ \hat{T}(t, t + \Delta t) = \exp\left( -\frac{i}{\hbar} \int_t^{t+\Delta t} \hat{h}(t) \, dt \right). \]

(2.84)

A small \( \Delta t \) is used to obviate the problems of time ordering due to the discrete time step. This operation can be performed efficiently on a discretised 3D grid using a Taylor expansion to represent the exponential (typically of the order of 4-6), and adopting a predictor-corrector strategy when evolving the wave functions [57].

In principle, the time evolution is unitary, therefore the single-particle orthonormalization and energies are conserved [21]. If the Slater determinant is a true eigenstate of the Hartree-Fock Hamiltonian, when time evolution begins, nothing will happen.

For this thesis, the time evolution of deformed initial configurations will be examined; many of these will not be eigenstates of the Hamiltonian. Therefore it may be of interest to simply evolve the Slater determinants in time to observe their behaviour. Other scenarios can of course be considered; it is common practice to apply time-dependent external fields or kinetic energy boosts to the system to investigate the physics of interest, ranging from small amplitude collective excitation modes [111, 112], to large amplitude fission or fusion studies [53, 60, 101, 113, 114]. Both approaches will be studied in time-dependent calculations of fission in Chapters 4 and 5.
2. APPLICATIONS OF MANY-BODY THEORY TO A NUCLEAR SYSTEM
Initial Conditions

Experimental studies of nuclear fission have been ongoing since the discovery of the process in 1938 by Hahn and Strassmann [16]. 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), allowing quantitative comparisons to be made between spontaneous and induced fission [115, 116, 117, 118]. Studies have demonstrated a multitude of techniques known to induce fission, including neutron-induced fission, fission induced by more complex projectiles, and photo-fission [115, 119]. Recent experimental campaigns have also been investigating the process of beta-delayed fission [120].

Investigations of spontaneous fission within TDHF are prohibitive. To reach a fissioned configuration from the ground or isomeric state, the nucleus must tunnel through the barriers in the potential energy surface (PES). TDHF allows a quantum mechanical description of single-particle wave functions, but the collective motion is semi-classical, forbidding tunnelling to occur. Therefore, TDHF is suitable for investigating induced, rather than spontaneous, fission processes.

To describe induced fission within TDHF certainly presents a challenge; namely choosing a method which will induce the process. We will follow two different strategies. Firstly, deformation-induced fission (DIF), where an initial state which is deformed such that fission is the most favourable way for the wave functions to evolve (thereby assuming the process inducing the reaction has already occurred). Secondly, boost-induced fission (BIF), where an excitation is provided during the time evolution that will allow the nucleus to fission.

The latter strategy can be justified by analogy to other investigations using TDHF. Collective giant resonances are often explored by calculating a ground state Slater determinant,
3. INITIAL CONDITIONS

then applying a small amount of energy in the form of a multipole boost to represent a physical process having occurred. For example, giant dipole excitation modes can be investigated by adding a small amount of energy in the form of a collective dipole operator, modelling the absorption of a photon \[111, 112\]. As the quadrupole degree of freedom is important when describing fissioning shapes, a large amplitude collective quadrupole excitation could be applied to a Slater determinant to mimic a process which induces nuclear fission \[55, 56\]. This will be investigated in Chapter 5.

A logical place to begin an investigation of fission within a microscopic framework is to examine the PES for the nucleus of interest. These are obtained by performing an energy minimisation with respect to constraints imposed upon the shape degrees of freedom. Early studies focussed upon constraining the quadrupole degree of freedom \[45, 46\]. 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. 3.1). Although the multi-humped behaviour of the energy surface cannot be measured directly, experimental evidence points towards the characteristic structure \[25, 26\]. Starting from the ground state, increasing the quadrupole deformation will result in encountering a first fission barrier. By increasing the deformation further, a secondary minimum, corresponding to an isomer, is found. Beyond this, a second fission barrier is encountered, and past this 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. 3.1 correspond to those defined in Ref. \[33\] as the energy difference between the ground state and the peak of the first fission barrier, the energy difference between the ground state and the peak of the second fission barrier, and the energy difference between the ground state and fission isomer, respectively. In some exotic cases, triple-humped potential surfaces are expected \[26, 50, 121, 122\].

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 \[48\]. The approach of calculating a multi-dimensional PES to describe fission within a microscopic framework has enjoyed much recent attention \[48, 49, 50, 51, 52\]. Another method used to explore fission pathways is to apply a shell-corrected macroscopic liquid drop model, which has been used to investigate five-dimensional deformation space. This technique has been applied to perform exhaustive
3.1 Shape Constraints

Figure 3.1: Schematic of typical potential energy surface obtained when increasing the elongation (which corresponds to a quadrupole deformation) of an actinide nucleus. The energies $E_A$, $E_B$ and $E_{II}$ relate to properties of the fission barriers, and correspond to the measurements defined in Ref. [33].

topographical surveys of deformation space to deduce fission properties of static configurations [30, 31, 32, 33].

However, this approach of calculating the PES to describe fission, regardless of the number of dimensions, is limited to producing a series of static solutions which attempt to describe a dynamic process. Performing shape-constrained DFT calculations produce Slater determinants which contain no internal excitation. Some attempts have been made to account for finite temperature effects [51], but as fission is a dynamic process, translational motion and collective excitations should be present. Time-dependent techniques may therefore yield new, insightful results as they can describe the dynamics of a fissioning system. Within the scope of this thesis, the PES will be investigated to produce static Slater determinants of varying deformation. The time evolution of these Slater determinants will then be investigated using TDHF.

3.1 Shape Constraints

Within the Hartree-Fock (or DFT) framework, the PES of a nucleus may be explored by performing a series of constrained Hartree-Fock (CHF) minimisations. As discussed in the previous Chapter, CHF techniques allow the calculation of the Slater determinant of minimum energy, subject to an auxiliary constraint. In all further discussions the Augmented
3. INITIAL CONDITIONS

Lagrangian Method (ALM) has been used to impose the auxiliary conditions (see Sec. 2.4.1).

3.1.1 Defining the Shape of a Nucleus

The starting point to perform CHF calculations of a nucleus is to choose an observable which may be constrained. In the context of an investigation of fission, the shape of the nucleus is a logical degree of freedom to consider. The nuclear shape may be defined by relation to the spherical harmonics. Figure (3.2) shows the spherical harmonics $Y_{l,\mu=0}$, $l = 1, \ldots, 4$ in comparison to a circle of fixed radius. The quadrupole ($l = 2$) degree of freedom provides a measurement of the elongation of the nucleus. The octupole ($l = 3$) deformation provides a quantification of deviations from mass reflection symmetry. Measurements of the hexadecapole ($l = 4$) deformation can be used to infer information regarding neck formation [123]. The mathematical expression of the $Y_{l0}$ is given explicitly in Appendix B.

![Spherical harmonics](image)

**Figure 3.2:** The spherical harmonics $Y_{l0}$, $l = 1, \ldots, 4$ with respect to a circle of constant radius. The shape of the nucleus will be defined with respect to these. Note that the $l = 1$ (dipole) case corresponds to a shift in the centre of mass, rather than a deformed shape.

The shape of standard nuclear states may be written in terms of an expansion of spherical harmonics; there is no reason why one should not include terms beyond $l = 4$, or restrict $\mu$ to zero. However, when performing CHF calculations or measuring the nuclear deformation, a small selection of relevant parameters is typically used to characterise the nuclear shape. In the context of this work, the deformation parameters $\beta_{l0}$, $l = 1, \ldots, 4$ have been defined as a measurement of the deformation, relating to the first four spherical harmonics. These deformation parameters are given explicitly in Appendix B.

Alternate useful parameterisations may also be defined: for example the Bohr-Mottelson deformation parameters describe the quadrupole degree of freedom and the corresponding...
3.1 Shape Constraints

triaxiality via the $\beta, \gamma$ representation [42]. In this representation, $\beta \geq 0$ and $0^\circ \geq \gamma \leq 60^\circ$ (see Appendix B). This parametrisation is advantageous as it gives a measure of triaxial deformation. For non-zero values of $\beta$, if $\gamma = 0^\circ$ the shape is purely prolate deformed. For $\gamma = 60^\circ$ the shape is purely oblate deformed. Other measures of $\gamma$ represent a triaxial shape.

3.1.2 Choice of Constraint for Studying Fission

For fission studies, undoubtedly the quadrupole degree of freedom is of importance, as it describes the elongation of the nucleus. Additionally, as many nuclei are observed to fission with asymmetric mass distributions, the octupole degree of freedom is vital to describe any mass reflection asymmetry. As previously mentioned, modern DFT solvers are able to perform symmetry unrestricted calculations which allow, in principle, any and multiple degrees of freedom to be explored.

Constraints can be imposed from alternate perspectives to study fission in static calculations. Some studies assume a priori knowledge of the fission fragments [53, 124], and constrain the distance between the two centres of mass to generate initial states before investigating time evolution.

Within the scope of this work, no a priori knowledge of the fission fragments is assumed. From a static perspective, fission is explored by gradually increasing the deformation of the nucleus from the ground state. Only the quadrupole deformation will be constrained; the purpose of this thesis 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 [21]. References [33] and [32] warn that this may not always be the case, arguing that a constrained minimisation will not necessarily follow the optimum fission pathway. They suggest that a complete topographical survey of the deformation space may be required to determine the static fission pathways. Such a survey may be possible within the shell-corrected macroscopic model employed in such studies, but is beyond the scope of current CHF calculations. It bears mention that the method applied in Ref. [33] can only explore the explicitly parameterised nuclear shapes, whereas Hartree-Fock calculations can in principle explore every shape degree of freedom, and determined the minimum energy configuration for a given constraint. It is purely down to the numerics of the calculation if
a state which is not the energy minimum configuration for a given constraint is converged upon.

### 3.1.3 The Principal Axis Co-ordinate Frame

The principal (or intrinsic) axis of the nucleus is an essential concept when performing static or time-dependent calculations in three dimensions: there is no requirement that the orientation of the Slater determinant must align with the lab co-ordinate frame \[^{[57]}\]. Simply performing a constrained minimisation upon the \(\beta_{20}\) parameter defined with respect to the fixed lab 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 3.3 illustrates this point with a crude cartoon.

For the case i), \(\beta_{20}\) is defined with respect to the lab frame. The measurement of \(\beta_{20}\) for case ii), also measured in the lab 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.

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. The principal axis corresponds to that of the greatest elongation. In the case iv), again the principal axis frame is 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 lab frame, as defined by

\[
Q_{ij} = \int dr \rho(r) (3r_i r_j - r^2 \delta_{ij}) .
\] (3.1)

The tensor should take into account a centre of mass correction (the origin of the co-ordinate system must be at the centre of mass of \(\rho(r)\)), and once built may be diagonalised. The diagonalisation 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 \[^{[111]}\]

\[
\beta_{20} = \frac{4\pi}{5} \frac{Q_{\text{princ}}}{A(r^2)} .
\] (3.2)
3.1 Shape Constraints

Figure 3.3: Cartoon displaying the comparison of multipole shapes in the lab $x - z$ frame [cases i) and ii)] vs. the principal axis $x' - z'$ frame [cases iii) and iv)]. When the Slater determinant is free to rotate in co-ordinate space, use of the principal axis co-ordinate frame is essential to define the nuclear shape via the $\beta_{ll}$ deformation parameters.

Diagonalising the quadrupole tensor also produces a set of eigenvalues, which can be used to map the rotation from the lab 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 $\mathbf{r} = (x, y, z)$. 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 [125], the rotation angle $\theta$ can be defined as

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

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

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

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.
3. INITIAL CONDITIONS

The rotation matrix, which is determined by considering the quadrupole tensor, may therefore be used to explicitly determine the multipole deformation parameters as measured in the principal axis frame.

3.1.4 Caveats When Performing Constrained Hartree-Fock Calculations

Three-dimensional, symmetry unrestricted calculations are preferable for describing the atomic nucleus, but performing such calculations does not come without difficulties. Several problems encountered when integrating the ALM to enforce shape constraints in the solver Sky3D will be briefly discussed.

3.1.4.1 Exploring The Space of Nuclear Configurations and Masking

Due to the choice of basis used in Sky3D, the wave functions are in principle free to explore the entire space within the numerical grid. As the Hartree-Fock minimisation will converge upon stationary points, instances may occur where multiple energy minima exist for a desired expectation of $\beta_{20}$ (they may have, for instance, different octupole or hexadecapole deformations). The CHF calculation will have no bias to converging in either minimum. Multiple minima may correspond to competing fission pathways, which can differ significantly in energy. When performing CHF calculations to obtain the quadrupole PES, it is preferable to explore fission pathways separately, rather than having configurations jumping between two or more minima. It is this jumping between fission pathways that is the main criticism Refs. [32, 33] present regarding the calculation of fission barriers using CHF.

Clearly, the most thorough way to explore the space of nuclear configurations is to enforce multi-dimensional constraints. For example the macro-micro calculations performed in Ref. [30, 32, 33] simultaneously constrain the elongation, mass asymmetry, left and right fragment deformation, and the neck thickness of the nucleus, resulting in the order of millions of possible configurations. As the number of constraints increases, the ambiguity in describing the nuclear shape will lessen. Unfortunately, calculating this many configurations within a self-consistent microscopic framework would require an unrealistic amount of time. Future modifications to Sky3D could reasonably increase the number of simultaneous constraints to two, in line with the solver HFODD [47].

For the results presented in this thesis, where only one constraint is applied upon the quadrupole degree of freedom, a masking procedure has 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.

The masking function $M(r)$ is a Fermi function which equals unity ‘inside’ the nuclear
surface, and transitions smoothly to zero ‘outside’ of the nucleus. It is included when cal-
culating 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. This 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.

![Figure 3.4: Masking procedure. Panel i) shows a slice of the particle density in the $x-z$ plane.
The binary mask shown in panel ii) is represented with a value of 1 if the grid point is inside
the nuclear surface, and 0 if the point is outside. The masking function shown in panel iii) is
then used to mask the particle density when calculating the expectation value of the deformation
parameters. See text for more details.](image)

A robust strategy to define $M(r)$ uses an autonomously updating mask (see Fig 3.4),
which has been applied successfully in the axially symmetric Hartree-Fock solver SKYAX
[88]. 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 $\rho(x, y, z) \geq \frac{\rho_{\text{max}}}{10}$, 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 the order of \((N_xN_yN_z)^2\) operations. However, one can check to see if the nuclear surface has changed at each iteration. If it has not, then 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:

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

where

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

The parameters \(\alpha\) and \(\gamma\) modulate how rapidly the mask tails off at the nuclear surface, and typical values were chosen to be 3.5 and 0.5 fm, respectively.

This masking technique can adjust to the changing nuclear shape over a PES without manual input. However, 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 was found to be unsuitable for use when measuring observables in large amplitude TDHF calculations, requiring an alternate strategy to be adopted. For dynamic calculations, the mask is defined relative to a fixed distance from the centre of mass (see Chapter 4). The masking strategy described in this Section generally proved to give good results for CHF calculations, and has been adopted for the results presented in this Chapter.

3.1.4.2 Defining Odd-\(l\) Deformation Parameters

When defining the principal axis co-ordinate system with respect to the quadrupole tensor, one must be aware that odd-\(l\) deformation parameters can only be determined up to an absolute sign. This is due to the unrestricted nature of the 3D calculations performed. Conversely, even-\(l\) deformation parameters are insensitive to mass asymmetry along the principal axis.

For example, consider an octupole deformed nucleus in the case where the principal axis \(z'\) aligns with the lab \(z\) axis. The measurement of \(\beta_{20}\) will be identical, regardless of whether
the positive $z'$ axis points in the direction of the positive or negative $z$ axis. However, as the definition of odd $l$ deformation parameters contain a factor of $z$ raised to an odd power, the measurement of $\beta_{30}$ will differ by an absolute sign depending on which direction the positive principal axis points along. A consistent definition of $\beta_{30}$ can be judged by examining the particle density.

3.2 Test Case: $^{46}$Ti

The nucleus $^{46}$Ti was chosen as a test case for implementing the ALM in Sky3D, selected as a sample nucleus expected to show a well-deformed global minimum. Due to the fact that it is a relatively light nucleus compared to those in the actinide region, calculations are less computationally demanding, and methods for investigating fission using Sky3D can be relatively quickly tested and verified before applying the framework to cases of greater interest.

Relatively few investigations of fission in this mass region have been performed. Light nuclei are stable to spontaneous fission, and require large excitations to induce the process. The fission of $^{44}$Ti has been studied via a 280 MeV fusion reaction of $^{32}S + ^{12}C \rightarrow ^{44}Ti^*$ [126, 127]; the composite system was excited enough to allow fission decays to become energetically possible. This leads to the expectation that the calculated fission barrier for $^{46}$Ti will be large. Additionally, as the nucleus is significantly lighter than those in the actinide region, the PES would not be expected to display the double-humped structure typical of actinides.

3.2.1 Specifics of the Calculation

The ground state and constrained Hartree-Fock calculations were performed using the SkM* effective interaction [128] in a regularly spaced Cartesian grid of $36 \times 36 \times 36$ points, ranging from $-17.5$ fm to $17.5$ fm in the $x, y,$ and $z$ directions. The interaction SkM* is fitted, amongst other nuclear structure properties, to fission barrier data, which makes it a logical choice to use in this investigation. Traditionally the interaction contains a centre of mass correction, which has been neglected.

Pairing was implemented using the BCS scheme, assuming a volume-delta interaction. The number of single-particle wave functions was chosen to span to 50 proton and 50 neutron states. Pairing strengths $V_0$ were 258.96201 and 279.08200 MeV for neutrons and protons,
3. INITIAL CONDITIONS

respectively [96]. The quadrupole deformation parameter $\beta_{20}$ was constrained in the range 0.150 to 1.40, incrementing in steps of 0.03. The ground state was calculated initially in an unconstrained Hartree-Fock minimisation to serve as a starting point for the calculation of the PES. Some ground state properties are listed in Table 3.1.

**Table 3.1:** Brief summary of ground state properties of $^{46}$Ti, calculated using the SkM$^*$ Skyrme effective interaction. Further details of the calculations are included in the text. The ground state is prolate deformed and axially symmetric.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Binding Energy [MeV]</th>
<th>rms Radius [fm]</th>
<th>$\beta_{20}$</th>
<th>$\beta_{30}$</th>
<th>$\beta_{40}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{46}$Ti</td>
<td>-379.68</td>
<td>3.555</td>
<td>0.171</td>
<td>0.000</td>
<td>0.100</td>
</tr>
</tbody>
</table>

The Lagrange multiplier $\lambda$ used to apply the constraint using the ALM was initially set to zero, and the stiffness constant $c$ was initially set to 50. The measurements of the density were masked when performing the calculations, as described in Sec. 3.1.4.1. Convergence was defined when the absolute difference between the desired and current expectation value of $\beta_{20}$ was less than $10^{-6}$, and the fluctuations in the single-particle energies were less than $10^{-4}$, averaged over the 30 most recent iterations.

Once a state was converged upon, the Lagrange multiplier was reset to zero, and the stiffness constant $c$ to 50. The desired value of $\beta_{20}$ was increased, and the process repeated for the next state, using the previous configuration as the starting point for the calculation.

### 3.2.2 Results

The quadrupole PES for $^{46}$Ti is shown in the top left panel of Fig. 3.5. As expected, the results imply that $^{46}$Ti is highly stable against fission decay, with a calculated potential barrier height in the order of 50 MeV. The system eventually fissions into two symmetric $^{23}$Na nuclei, where beyond this point the total binding energy of the system begins to increase once more as the two fragments separate. Throughout the PES, triaxial and octupole deformations are negligible. An interesting feature as the nucleus necks to the point of scission is a ‘back bending’ of the hexadecupole deformation parameter $\beta_{40}$ (top right panel of Fig. 3.5). This can be explained from the definition of the parameter (see Appendix B, and Fig. 3.2). As the neck vanishes, the hexadecupole deformation initially decreases. However, as the two fragments separate in space, the $z^4$ term dominates, causing $\beta_{40}$ to rapidly increase once more.
Overall, reasonable results have been obtained, verifying the implementation of the ALM in Sky3D. The method may be applied to investigate actinides or heavy nuclei within studies of nuclear fission.

3.3 Benchmark Case: $^{240}$Pu

The reproduction of the double-humped fission barriers of actinide nuclei are often used as a benchmark test for nuclear models [61]. Due to wealth of data available from experimental [115, 116, 117, 118] and theoretical [23, 33, 46, 129] studies of the nucleus, $^{240}$Pu presents itself as a strong candidate for a benchmark test of TDHF to investigate induced nuclear fission. The static quadrupole constrained PES will firstly be calculated to provide a selection of initial states for time evolution.
3. INITIAL CONDITIONS

3.3.1 Specifics of the Calculation

The ground state and constrained Hartree-Fock calculations were performed using the SkM* effective interaction 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. The fission barrier properties of $^{240}$Pu were considered when fitting the SkM* effective interaction [128]. Flocard et. al published the first self-consistent calculations of the barrier for the nucleus in 1974 using the SIII effective interaction [46]. The interaction SkM was applied to barrier calculations by Bartel in 1982 [128], and was further optimised, resulting in the SkM* interaction. Both the early studies imposed axial symmetry, and in the case of the calculations by Flocard, reflection symmetry perpendicular to the elongation of the nuclear was enforced. The double-humped structure of the PES was reproduced in both cases, and measurements of the barrier heights may be used for comparison to this work.

Pairing was once again included within the calculation; 184 neutron and 126 proton single-particle wave functions were used. The pairing strengths adopted were the same used as in the $^{44}$Ti case discussed previously. Due to the existence of a prominent local minimum in the PES (corresponding to the fission isomer), by choosing a highly prolate deformed initial harmonic oscillator width, Hartree-Fock calculations were found to converge in this minimum without imposing shape constraints.

Table 3.2: Brief summary of some ground state and isomer properties of $^{240}$Pu, calculated using the SkM* Skyrme effective interaction. Further details of the calculations are included in the text. Both the ground state and isomer are prolate deformed and axially symmetric.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Binding Energy [MeV]</th>
<th>rms Radius [fm]</th>
<th>$\beta_{20}$</th>
<th>$\beta_{30}$</th>
<th>$\beta_{40}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{240}$Pu</td>
<td>$-1781.95$</td>
<td>5.941</td>
<td>0.280</td>
<td>0.000</td>
<td>0.255</td>
</tr>
<tr>
<td>$^{240}$Pu*</td>
<td>$-1778.91$</td>
<td>6.418</td>
<td>0.682</td>
<td>0.000</td>
<td>0.547</td>
</tr>
</tbody>
</table>

This provided two initial points for the CHF calculations, starting at either the ground state or isomeric state to explore the PES. Some properties of the ground state and isomer are presented in Table 3.2. Flocard and Bartel reported ground states of greater binding energy in their work; $-1801.5$ and $-1822.6$ MeV, respectively. The differences may be attributed to several factors. Firstly, adoption of different Skyrme effective interactions may lead to significant differences. Additionally, the calculation performed in this work contained no
centre of mass correction. Pairing correlations were treated differently, as Flocard chose the pairing strength to be proportional to the surface of the nucleus, whereas Bartel adopted a constant pairing matrix element. Further, the method used to apply the constraints was different; we use the Augmented Lagrangian Method, whereas the Quadratic Penalty Method was used in early CHF investigations (see Sec. 2.4). The early calculations performed also enforced spatial symmetries. In principle, this should not have any effect as the ground state and isomer as they do not demonstrate any triaxial deformation or reflection asymmetries, but the enforced symmetry may have some effect upon the numerics of the calculation, and will certainly have an effect upon the calculated fission barriers.

3.3.2 Results

The PES for $^{240}$Pu is shown in Fig. 3.6. As the PES is far more complex than the case of $^{46}$Ti, a colour scheme has been imposed to correspond to measurements of the different multipole parameters for the same configuration. Two fission barriers are seen in the quadrupole degree of freedom (top left panel of Fig. 3.6), peaking at $\beta_2 = 0.50$ and 0.86 respectively. The ground state and isomeric state correspond to the two minima in the PES next to these barriers. Table 3.3 presents a comparison between various features of the PES calculated in this work to results presented in References [46], [128], [33] and [129]. The Table contains measurements of the fission barrier heights and energy difference between the ground and isomeric state, as defined in Fig. 3.1.

<table>
<thead>
<tr>
<th></th>
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</thead>
<tbody>
<tr>
<td>8.25</td>
<td>7.68</td>
<td>3.04</td>
<td>HF+BCS, Skyrme SkM$^\ast$</td>
<td>This work</td>
</tr>
<tr>
<td>9</td>
<td>13</td>
<td>4</td>
<td>HF+BCS, Skyrme SIII</td>
<td>Table 2 of Ref. [46]</td>
</tr>
<tr>
<td>9.54</td>
<td>8.62</td>
<td>1.08</td>
<td>HF+BCS, Skyrme SkM</td>
<td>Fig. 8 of Ref. [128]</td>
</tr>
<tr>
<td>9.30</td>
<td>8.40</td>
<td>3.10</td>
<td>HFB, Gogny D1M</td>
<td>Fig. 5 of Ref. [129]</td>
</tr>
<tr>
<td>5.99</td>
<td>4.91</td>
<td>2.94</td>
<td>Shell-Corrected FRLDM</td>
<td>Table 1 of Ref. [33]</td>
</tr>
<tr>
<td>6.1±0.3</td>
<td>6.0±0.50</td>
<td>2.1±0.6</td>
<td>Experiment</td>
<td>Fig. 27 of Ref. [33] (Madland)</td>
</tr>
<tr>
<td>5.6±0.2</td>
<td>5.1±0.20</td>
<td>2.4±0.3</td>
<td>Experiment</td>
<td>Fig. 27 of Ref. [33] (Madland)</td>
</tr>
</tbody>
</table>
3. INITIAL CONDITIONS

Figure 3.6: Resulting PES for $^{240}$Pu following a constraint of the quadrupole deformation parameter $\beta_{20}$. Once the second barrier has been passed, 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 even for states far beyond the second barrier, scission has not yet occurred.

When comparing the mean-field calculations of this work with that of [129] (Table 3.3), a difference in $E_A$ and $E_B$ of the order of 1 MeV is found. This could be attributed to the difference in effective nuclear interaction adopted, and further, the HFB calculations of [129], in principle, treat pairing correlations more completely than the CHF calculations of this work. The barrier calculations of Flocard [46] determine $E_A$ to be of a similar value to this work, which is somewhat surprising, as the axial symmetry enforced in the calculations would be expected to produce a barrier approximately 1-2 MeV higher. This demonstrates that there are other significant differences in the calculations compared to this work. The difference in effective interaction, the treatment of pairing and the centre of mass correction are factors which may contribute significantly. The second barrier height ($E_B$) of Ref. [46] is
almost double that of this work, which is unsurprising as reflection symmetry was enforced, removing the possibility of the calculations to explore octupole deformed shapes.

The barrier heights obtained by Bartel et al. are in agreement to within \( \approx 1 \text{ MeV} \) of this work, which is perhaps unsurprising due to the similarities between the interactions SkM and SkM*. The differences can likely be attributed to factors such as the treatment of pairing and any centre of mass corrections. As reflection asymmetry could be explored, \( E_B \) is significantly lower than the result presented by Flocard. The value obtained for \( E_{II} \) is notably lower than any of the other results, which is perhaps surprising.

Upon comparing the results of the mean-field calculations to those of the macroscopic-microscopic calculations (shell-corrected Finite-Range Liquid Drop Model), the values of \( E_A \) and \( E_B \) presented are 2-4 MeV lower for the macroscopic models. The differences likely stem from the ingredients of the different models; in the mean field calculations the only parameters are those defined in the Skyrme interaction and those which define the treatment of pairing. In the macroscopic-microscopic calculations, the energies are defined by the fixed five-dimensional parameterisation [33], whereas in principle the mean field calculations explore an infinite dimension deformation space. When comparing the calculated values to experimental data, the lower values of \( E_A \), \( E_B \) and \( E_{II} \) presented in Ref. [33] agree more closely to the experimental values. It must be emphasised, however, that the ‘experimental’ values are extracted in a model-dependent manner from fission cross section data. Assumptions made when extracting the barrier heights from this data will have implications upon the deduced values [26]. Therefore, assessing the merits of a PES calculation based solely upon reproducing these values may be inadvisable.

Figure 3.6 shows a prominent octupole deformation setting in at the second fission barrier (top middle panel), as would be expected [61]. The relationship between the quadrupole and octupole deformation parameters for the configurations along the PES is shown in Fig. 3.7 (top left panel). Although the calculations have been performed constraining only one deformation degree of freedom, the behaviour observed is typical for the optimum static fission pathway obtained in quadrupole-octupole constrained deformation surfaces calculated using DFT [48, 50].

Figure 3.7 also displays the relationship between the quadrupole and hexadecupole deformation parameters (top right panel). Near the peak of the first and second barrier, the
hexadecupole deformation is seen to sharply drop, then recover. This corresponds to a transitioning shape as the neck region of the nucleus thins. By increasing the quadrupole deformation beyond these points, the elongation of the state increases, which allows the value of $\beta_{40}$ to increase once more as the $z^4$ term in the hexadecupole operator dominates.

The 3D calculations verify that triaxiality is explored at the first fission barrier. This is demonstrated by observing a non-zero measurement of $\gamma$ when calculating the Bohr-Mottelson $\beta - \gamma$ deformation parameters (lower left panel of Fig. 3.7). This shows the barrier is not only defined for $\beta_{20}$. Being able to explore this degree of freedom lowers the calculated barrier height with respect to axially symmetric calculations [61]. Triaxiality is explored significantly in the range $0.36 \leq \beta_{20} \leq 0.59$. Other than the region corresponding to the first barrier, triaxiality is virtually negligible, therefore $\beta = \beta_{20}$. Even for the resulting states where $\gamma$ is non-zero, $\beta \approx \beta_{20}$.

One interesting comparison to the results of the calculations for Ti (Fig. 3.5) is that the Pu nucleus has not fissioned in the range of $\beta_{20}$ considered, even for states beyond the
second fission barrier. The slices of the density shown in Fig. 3.6 display an increasingly deformed shape rather than two separate fragments. Due to a competing fission pathway, the PES could not be explored all the way to the point of scission (see Sec. 3.3.3). Despite the emergence of this competing fission pathway, a large selection of increasingly deformed states have been obtained. These range from configurations with a quadrupole deformation less than that of the global Hartree-Fock minimum, to configurations well beyond the second fission barrier.

3.3.3 Competing Fission Pathways

When performing constrained Hartree-Fock calculations, it was noticed that once the quadrupole deformation was increased beyond $\beta_{20} = 1.25$, the configuration jumped abruptly to a competing fission pathway. This behaviour was due to the numerics as the calculations converged and proved to be unavoidable, even when adopting the masking procedure described in Sec. 3.1.4.1. This can be explained by considering the density slices presented in Fig. 3.8; a mask around the one-fragment configuration will not inhibit a transition to the two-fragment configuration, as the two-fragment configuration ‘fits’ in the one-fragment masking region.

The competing fission pathway was explored, starting from the state after the calculations jumped pathway. From this configuration, the deformation was incrementally reduced. This competing pathway is shown in Fig. 3.8, coloured in blue, and may be compared to the original pathway, which is coloured in red. Once the quadrupole deformation was reduced below 1.01, the Hartree-Fock minimum was observed to jump back onto the original fission pathway.

The competing pathway, referred to as the ‘two-fragment’ pathway in Ref. [129], displays remarkably different configurations to that of the ‘one-fragment’ pathway. Even with identical quadrupole deformations, the octupole and hexadecupole deformations, and total energy differ significantly. It is exactly this behaviour that the authors of Refs. [33] and [32] identify as a flaw when using CHF to explore the PES. However, this ‘flaw’ may be exploited in this instance to gain an insight of a competing fission pathway, without having to include a higher number of constraints in the CHF calculations.

Unlike the case of $^{46}$Ti, which cleanly splits upon fission in the CHF calculations (Sec. 3.2.2), the fragments in the two-fragment pathway do not have an integer particle number.
3. INITIAL CONDITIONS

Figure 3.8: One (red line) and two-fragment (blue line) fission pathways for $^{240}$Pu. The arrows in the top left panel show which direction the PES was explored in. Beyond $\beta_{20} = 1.25$ the one-fragment pathway jumps into the two-fragment pathway, and this state was used for 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$^3$.

For example, for the case of $\beta_{20} = 1.19$, the fragments have $A_1, Z_1 = 107.14, 43.14$, and $A_2, Z_2 = 132.85, 50.85$ (to two decimal places). 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 [131]. This would give access to a mass distribution, but such an analysis is beyond the scope of this work.

It warrants mention that the mass distributions of the two-fragment solutions differ significantly from the experimentally observed distributions: the most probable mass split for thermal neutron-induced or photon-induced fission in $^{240}$Pu is $A \approx 100$ and $A \approx 140$ [115, 116]. A discussion of the significance of the intercept of the one and two-fragment pathways in light of TDHF fission results will be presented in Sec. 4.1.4.

Overall, a large selection of constrained Hartree-Fock states have been determined for $^{240}$Pu. The calculations provide qualitatively consistent results to other modern mean-field investigations of $^{240}$Pu [129]. They present themselves as initial configurations to commence an investigation of induced fission using time-dependent Hartree-Fock.
Time Evolution of Constrained Hartree-Fock States

The time evolution of the constrained Hartree-Fock (CHF) states obtained for $^{240}$Pu (as described in Chapter 3) may be investigated using the time-dependent Hartree-Fock method.

The analysis will focus on the states on the one-fragment fission pathway, starting from configurations beyond the fission isomer (that is, those with $\beta_{20} > 0.68$). This Chapter 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 as described by time-dependent Hartree-Fock.

The Sky3D code provides the capability to time-evolve 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 [57]. 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 of dimension $(N_x N_y N_z)$ with its centre of mass at $(x, y, z) = (0, 0, 0)$. The wave functions are set (close) to zero for all the space which exceeds the dimensions of the static solution. The wave functions are then re-orthonormalised before time evolution begins.

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 dimen-
sions 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 a continuum [132, 133]. Some attempts have been made to reproduce continuum calculations in 3D, but the methods are computationally expensive and may not be suitable for large amplitude processes [134, 135].

When analysing the nuclear dynamics, a masking procedure is once more adopted to define the nucleus. This allows contributions to the observables from the tails of the wave functions to be eliminated. This ensures that consistent results are obtained with different choices of grid dimension. This mask was chosen to be of the form of a Fermi function, in line with the masking procedure described in Chapter 3 for the static case. A generalisation becomes necessary in the dynamic case as the fragments in a post-fissioned system will propagate through space. For dynamic calculations, the mask is defined by a fixed distance from the centre of mass, which is not necessarily a grid point. This is in contrast to the mask described by Eq. (3.5), which was computed relative to discrete grid points. If the mask is defined relative to grid points, as the fragments propagate through space, the mask will ‘jump’ between grid points, resulting in a slight discontinuity in the time evolution of any measurements performed.

For a fissioning system, once the system splits into two fragments, a separate mask is defined for each fragment. This will be discussed in more depth in Sec. 4.2. 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 239.9999 particles at time $t = 0$ fm/c.

The time evolution of the CHF solutions yielded two general cases. Firstly, where the nucleus is sufficiently deformed so that the repulsive Coulomb interaction is able to drive the configuration to fission. Secondly, states which fail to fission. The non-fissioning case will be discussed first.

### 4.1 Non-Fissioning States

A selection of states along the one-fragment PES (Fig. 3.6), starting from just beyond the static fission isomer ($\beta_{20} = 0.682$), were evolved using TDHF. For these calculations, the initial static Slater determinants were placed in a grid of $42 \times 42 \times 42$ points, ranging from
−20.5 to 20.5 fm in the $x$, $y$ and $z$ directions. The states were evolved in time up to 9000 fm/c. The time increment was chosen to be $\Delta t = 0.25$ fm/c, and 8 terms were used in the expansion when applying the time evolution operator (see Sec. 2.6).

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 calculations compared to the static calculations the initial values at $t = 0$ of the multipole deformation parameters differ slightly. As we shall see, differences in the time evolution due to, for instance, pairing neglect are no larger than than 1%.

![Graph of time evolution of multipole deformation parameters](image)

**Figure 4.1:** Time evolution of multipole deformation parameters from different initial states (the corresponding $\beta_{20}$ of the static state is labelled). Note that for the initial states with $\beta_{20} = 0.71$ and 0.77, the octupole deformation is negligible for the time elapsed in the calculation.

The time evolution of the multipole deformation parameters for the non-fissioning states are compared in Fig. 4.1. From a brief inspection of the Figure, it can be seen that the evolution of the quadrupole deformation parameter is unique for each state presented (that is, the lines in the top left panel never cross). This suggests that the TDHF wave functions for each configuration are exploring a different minimum in quadrupole deformation space.

The distinct differences 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, will be explored.
4. TIME EVOLUTION OF CONSTRAINED HARTREE-FOCK STATES

4.1.1 Time 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 $\beta_{20} = 0.860$) are shown in Fig. 4.2. 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. 4.2, where quadrupole deformation reduces from $\approx 0.71$ to $\approx 0.695$ within 200-300 fm/c. The initial drop in elongation corresponds to a rearrangement of the density: within time-dependent calculations energy is conserved, but collective motion and internal excitations are allowed. Additionally, pairing is not included in the time-dependent calculations. The initial deviation of nuclear shape could be due 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 the small-amplitude, high-frequency vibrations in

Figure 4.2: Time evolution of multipole parameters for initial states which were solutions to the CHF calculations (labelled on the right hand side). All of the initial states are deformed below the static fission barrier.

- $\beta_{20}$
- $\beta_{30}$
- $\beta_{40}$

Time [fm/c]
4.1 Non-Fissioning States

the quadrupole and hexadecupole deformation (left and right columns of Fig. 4.2).

For the case with static deformation $\beta_{20} = 0.71$ (top row of Fig. 4.2), the octupole deformation of the nucleus remains negligible throughout 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 [21]. In contrast, the state with initial deformation $\beta_{20} = 0.77$ (middle row of Fig. 4.2) shows evolution of the octupole deformation oscillating about zero. The amplitude of this vibration is small: the octupole deformation throughout time evolution is practically unnoticeable when examining the particle density. Additionally, the frequency of vibration is far slower than that of the quadrupole and hexadecupole parameters. Although in principle odd and even-$l$ vibrational modes may couple in non-spherical nuclei [136, 137], due to the differences in frequency it appears that the $l = 3$ mode is not coupled with the $l = 2$ or $l = 4$ mode.

For the state with initial deformation $\beta_{20} = 0.83$ (bottom row of Fig. 4.2), the octupole parameter has an initial value very close to zero, but upon time evolution a dramatic evolution of the nuclear shape occurs. 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 hexadecupole deformation parameter, as the higher order terms in the definition of the parameter (proportional to $z^4$) drown out this effect. The initial configuration is close to the peak of the static fission barrier (see Fig. 3.6), 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 [138].

Typically, when Fourier analysis is applied, it is to investigate the response of the nucleus to a specific excitation, delivered in the time profile $f(t)$, to obtain a strength function.
Practically, within TDHF calculations, this is determined by computing the expectation of an observable \( \langle \hat{\zeta} \rangle \) as a function of time

\[
\zeta(t) = \langle \Phi(t) | \hat{\zeta} | \Phi(t) \rangle. \tag{4.1}
\]

This is then Fourier transformed into the frequency domain:

\[
\tilde{\zeta}(\omega) = \int_{0}^{\infty} e^{i\omega t} \zeta(t) dt, \tag{4.2}
\]

as is the time profile of the function which provides the excitation, thus obtaining \( \tilde{f}(\omega) \). The strength function is then defined by:

\[
S_{\zeta}(\omega) = -\frac{1}{\pi} \text{Im} \left\{ \frac{\tilde{\zeta}(\omega)}{\tilde{f}(\omega)} \right\}. \tag{4.3}
\]

In the case investigated in this thesis, however, there is no well-defined strength function as no initial excitation is applied to determine the corresponding nuclear response. Instead, the spectral power function

\[
P_{\zeta}(\omega) = \left( \text{Re} \tilde{\zeta}(\omega) \right)^2 + \left( \text{Im} \tilde{\zeta}(\omega) \right)^2 \tag{4.4}
\]

is defined. As it will be shown in the following, this will serve to identify the dominant collective excitation energies of the nucleus.

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 a spurious nuclear motion need to be eliminated. For the cases presented in Fig. 4.2, this is a simple procedure which can be done by performing an average fit to the evolution of the deformation parameters. This fit is then subtracted from the evolution of the multipole moments. For the case of the quadrupole deformation parameter for the state with initial \( \beta_{20} = 0.83 \) (bottom left panel of Fig. 4.2), two Gaussian functions have been fitted and subtracted to remove the drift caused by the moving centre of mass.

From the results of this subtraction process (shown in Fig. 4.3), the spectral power can be obtained. Figure 4.4 shows the resulting power spectrum. For the case of the octupole power spectrum for initial \( \beta_{20} = 0.83 \), not enough signal was present to perform the Fourier transform (bottom centre panel of Fig. 4.2).

In the process to obtain the power spectra, a windowing technique was used to force the signal to reduce to zero by the end of the calculation time to suppress any artefacts.
4.1 Non-Fissioning States

Figure 4.3: The time evolution of the multipole moments (Fig. 4.2) have been centred around zero to obtain power spectra. See text for more details.

The resolution of the spectra is defined by the measurement time, and is of the order \( h\omega = \frac{\pi}{T_{\text{obs}}} \) [139]. Some authors smooth the power spectrum 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 will be applied, as it is not necessary when identifying the dominant excitation modes.

The qualitative features of the evolution of the multipole moments and their corresponding power spectrum presented so far 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 4 MeV (top left and middle left panels of Fig. 4.4, respectively). Additionally, both show a second well-defined peak. For evolution of the state with deformation \( \beta_{20} = 0.71 \) the second peak is around 1 MeV (top left panel of Fig. 4.4), whereas for evolution of the state with deformation \( \beta_{20} = 0.77 \) the second peak lies between 6-7 MeV (middle left panel of Fig. 4.4). For the case with initial deformation \( \beta_{20} = 0.83 \), the dominant quadrupole excitation mode lies slightly higher, between 4-6 MeV.

In all three cases, the dominant peaks in the hexadecupole spectra lie at the same ener-
Figure 4.4: Power spectra corresponding to Fig. 4.3. The signal was insufficient to build a power spectrum for the evolution of the octupole deformation for the case with initial deformation $\beta_{20} = 0.83$.

energies as those in the quadrupole spectra (comparing the left and right columns in Fig. 4.4). This feature is expected due to the strong coupling between quadrupole and hexadecupole excitation modes.

A low-energy peak is visible in the octupole power spectrum for the evolution of the state with initial deformation $\beta_{20}=0.77$ (middle centre panel in Fig. 4.4). The peak is sharp, corresponding to very well-defined mode. The peak lies away from the energies of any excitation modes attributed to quadrupole or hexadecupole vibrations, verifying that the octupole mode seen in Fig. 4.2 (middle row) is not coupled with the quadrupole or hexadecupole modes.

As mentioned, no power spectrum could be calculated, due to the lack of signal, for the evolution of the octupole deformation for the state with initial deformation $\beta_{20} = 0.83$. It can be seen in the bottom centre panel of Fig. 4.3 that the oscillation undergoes half a cycle in approximately 7000 fm/c, which would correspond to a collective excitation energy in the region of 0.1 MeV. This sits far away from any excitation energies corresponding to the quadrupole or hexadecupole modes for this state (bottom left and bottom right panels of Fig. 4.4).

For comparison, a benchmark calculation of the collective giant quadrupole resonance
4.1 Non-Fissioning States

(GQR) of $^{240}$Pu has been performed. Figures 4.5 and 4.6 show the evolution of the deformation parameters for the ground state and isomeric state following an instantaneous, small-amplitude, isoscalar boost applied via a quadrupole field. The power spectrum is shown, rather than the strength function, for direct comparison to Fig. 4.4. In both cases, the density remains mass symmetric throughout the time evolution, therefore no octupole power spectrum is obtained.

![Figure 4.5](image)

**Figure 4.5:** Evolution of quadrupole and hexadecupole deformation parameters for the ground state of $^{240}$Pu following a small amplitude, isoscalar, instantaneous quadrupole boost (top). The bottom panels show the corresponding power spectrum.

For the GQR calculations, in the quadrupole power spectrum both for the ground state and isomeric state, the dominant excitation mode lies between 4 and 5 MeV (Figs. 4.5 and 4.6, bottom left panels). This is directly comparable to the quadrupole power spectrum determined from the evolution of the deformed static states, shown in Fig. 4.4 (left column). The common peak in the spectra around the same energy could be interpreted as a GQR. The quadrupole and hexadecupole spectra for the ground state GQR spectra shows a secondary peak around 10 MeV (Fig. 4.5), which is not observed in any of the spectra in Fig. 4.4, or that for the isomer GQR (Fig. 4.6).

There are other dominant excitation modes in the quadrupole and hexadecupole spectra presented in Fig. 4.4 that do not appear in the spectra corresponding to the ground state GQR (Fig. 4.5). However, peaks are observed in the spectra corresponding to the isomer GQR.
Figure 4.6: Evolution of quadrupole and hexadecupole deformation parameters for the isomeric state of $^{240}$Pu following a small amplitude, isoscalar, instantaneous quadrupole boost (top). The bottom panels show the corresponding power spectrum. The peaks around 1, 6 and 8 MeV are not observed in the ground state spectra, but are seen in some cases in Fig. 4.4.

at these energies, as well as the dominant mode around 4 MeV (Fig. 4.6). In other words, for the three states investigated in this Section, the system oscillates collectively around a quadrupole minimum with similar properties to that of the isomer GQR, rather than that of the ground state GQR.

Qualitatively, a picture of the evolution of collective excitation modes with increasing deformation can be observed in Fig. 4.4. The quadrupole and hexadecupole excitation modes appear to couple strongly together in all cases, which is unsurprising when considering the shapes that the multipole deformations physically correspond to. In the only case where the octupole power spectrum could be extracted, the excitation frequency is significantly different to the quadrupole and hexadecupole modes, demonstrating that the $l = 3$ mode is not coupled strongly to the even-$l$ modes.

4.1.2 Evolution of the State at the Peak of the Static Fission Barrier

The time evolution of the static state at the peak of the second fission barrier ($\beta_{20} = 0.86$) is a case of interest. If the static barrier has physical implications regarding fission, the evolution of the states at the peak of the barrier would be expected to display behaviour different to those deformed below the barrier.
Figure 4.7 displays the evolution of the multipole deformation parameters for the state with initial deformation $\beta_{20} = 0.860$. Unfortunately, the calculation became numerically unstable beyond 4000 fm/c. As the non-fissioning states presented in the previous Section could be evolved safely up to 9000 fm/c (Fig. 4.2), it can be speculated that this is due to a non-linear behaviour as the configuration evolves, and the state is indeed a point of significance along the PES.

Some features of the evolution may be commented upon. The development of the octupole deformation is perhaps the most striking feature. Like the case evolved from the static state with $\beta_{20} = 0.83$ (Fig. 4.2, bottom row), which lay just below the peak of the barrier, a significant octupole deformation is explored. The initial state with $\beta_{20} = 0.86$ has a very small mass asymmetry ($\beta_{30} \approx -0.01$), which develops as the nucleus rearranges configuration upon time evolution to oscillate about $\beta_{30} \approx -0.2$.

In contrast to the state evolved from $\beta_{20} = 0.83$, the octupole deformation seems to begin to explore an energy minimum at a non-zero value, rather than slowly oscillating about zero. This implies that there is a more favourable configuration which may be explored in TDHF. Quadrupole and hexadecupole oscillations are also observed as the state is evolved in time, with a drift corresponding to the evolution of the octupole deformation as the centre of mass.
adjusts significantly (left column of Fig. 4.7). Not enough signal is present to calculate a power spectrum. Qualitatively, the oscillations in the quadrupole and hexadecupole mode correspond to a collective excitation energy of \(\approx 4\) MeV, which is similar to that observed for the excitation peak shared in common in the quadrupole and hexadecupole spectra for all three states in Fig. 4.4 (and also in the ground state and isomer GQR spectra (Figs. 4.5 and 4.6)). A transitionary behaviour is undergoing in the evolution of this state; as well as giant resonance-type behaviour occurring in the quadrupole and hexadecupole modes, the nucleus is undergoing a large amplitude rearrangement in the octupole degree of freedom.

Fission is not observed in the time scale for which the calculation was performed. An investigation of the time evolution of those initial states deformed beyond the static fission barrier (that is, initial \(\beta_{20} > 0.86\)) will be presented in the next Section.

### 4.1.3 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 failed 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. 4.8.

Qualitatively, one observes dramatically different behaviour in the time evolution of the multipole deformations for these states compared to Fig. 4.2. The elongation is seen to rapidly increase during the first 300-500 fm/c of time evolution (corresponding to an increase \(\beta_{20}\), left column of Fig. 4.8). 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 \(\beta_{20} \approx 1.11\) in this time. This is in contrast to that seen for the evolution of states below the static fission barrier (left column of Fig. 4.2), where the initial quadrupole deformation was seen to decrease within the first 200-300 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. 4.8 displays slow, large amplitude oscillations setting in. Compared to the states below the static fission barrier (Fig. 4.2), these oscillations are substantially slower. They will 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. 4.8), 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 region of rapid increase, then an oscillation about a plateau, then another rapid increase followed by another plateau.

An octupole deformation is also observable in all cases (centre column of Fig. 4.8). 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 hexadecupole parameter, or both the quadrupole and hexadecupole parameters. For the giant resonance cases observed previously (Fig. 4.2) the octupole modes did not seem to be coupled strongly with the quadrupole or hexadecupole modes. This fea-
4. TIME EVOLUTION OF CONSTRAINED HARTREE-FOCK STATES

ture, in addition to the other differences observed between the evolution of the multipole deformations above and below the fission barrier, could suggest that the mechanism driving the dynamics is different, and not typical of a giant resonance. This point will be elaborated shortly.

Figure 4.9: The time evolution of the multipole moments (Fig. 4.8) have been centred around zero, and will be used to obtain power spectra. The order of the polynomial fitted to the data has been noted. See text for more details.

Fourier analysis can be applied analogously to the procedure in the previous Section. The subtraction procedure to centre the oscillations about zero is far more challenging here. Figure 4.9 shows the evolution of the multipole deformation parameters following the subtraction. The polynomial order of the function fitted to the data and subsequently subtracted is stated on the Figure. These fits ranged from a constant fit (0\textsuperscript{th} order) to a 4\textsuperscript{th} order polynomial. A similar procedure has been used to eliminate spurious motion for systematic investigations of the GQR in Ref. [107].

The resulting power spectra are shown in Fig. 4.10. These differ drastically to those seen
for the evolution of states below the static fission barrier (Fig. 4.4). Qualitatively, the power spectra for all three deformation parameters show dominant excitation modes at low energies (below 2 MeV), and secondary excitation modes still at energies far below what was observed for the states below the fission barrier (Fig. 4.4). One must be cautious when interpreting the spectra presented in Fig. 4.10. Although great care has been taken to centre the evolution of the multipole moments about zero, any remaining drift in the original signal will cause low energy artefacts to appear in the power spectrum.

Several results have been obtained so far which suggest that the mechanism driving the evolution of the nuclear shape differs significantly when starting from a configuration below or beyond the barrier. The evolution of the state with initial deformation $\beta_{20} = 1.01$ will be used to illustrate the hypothesised mechanism which drives the dynamics for the non-fissioning states beyond the second fission barrier.

Figure 4.11 displays 2D slices of the 3D density at various times during the time evolution
4. TIME EVOLUTION OF CONSTRAINED HARTREE-FOCK STATES

Figure 4.11: 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 at 0.05 particles/fm$^3$.

The density can be seen to be rearranging during the time evolution, and a striking feature is seen beyond 6000 fm/c. The nucleus begins twisting and rotating; energy is being transferred from potential energy into collective kinetic and rotational energy$^1$. This behaviour has been observed in other studies [141]. The slow, large amplitude oscillatory behaviour of the multipole deformation parameters (second row from bottom of Fig. 4.8) 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 [20], 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,

\footnote{This observation should be investigate further to ensure that it is not caused by the numerics of the calculation.}
4.1 Non-Fissioning States

but the mechanism is microscopically and not phenomenologically included.

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. 4.11). Figure 4.12 displays the current vectors $\mathbf{j}(\mathbf{r})$, corresponding to Fig. 4.11. 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. 4.12 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 show a particle flow that demonstrates the top and bottom lobes of the deformed nucleus are not moving in phase with one another, which suggests they are behaving like separate, interacting, fragments rather than a single nucleus. At some points (such as 8250 fm/c), the magnitude of the current

\[ t = 0 \text{ fm/c} \quad t = 750 \text{ fm/c} \quad t = 1500 \text{ fm/c} \quad t = 2250 \text{ fm/c} \]

\[ t = 3000 \text{ fm/c} \quad t = 3750 \text{ fm/c} \quad t = 4500 \text{ fm/c} \quad t = 5250 \text{ fm/c} \]

\[ t = 6000 \text{ fm/c} \quad t = 6750 \text{ fm/c} \quad t = 7500 \text{ fm/c} \quad t = 8250 \text{ fm/c} \]
vectors is far greater, demonstrating translational motion as well as collective excitations as the nucleus rearranges. All in all, this further shows that the behaviour is not typical of a collective giant resonance, and that a different mechanism is driving the nuclear dynamics.

To strengthen the picture that the dynamics of the nucleus are due to competition between the repulsive Coulomb and attractive terms in the energy density functional, the contribution to the total energy of the system from the Coulomb energy (defined in Sec. 2.3.2) is shown in Fig. 4.13 throughout the time evolution for two cases. The Figure displays the evolution of the Coulomb energy for the state with deformation $\beta_{20}=0.71$, which is below the static fission barrier and undergoes giant resonance-type behaviour (left panel), and for the state with $\beta_{20} = 1.01$, which is beyond the static fission barrier (right panel).

![Figure 4.13](image)

**Figure 4.13:** Evolution of the Coulomb energy for the evolution of the state with static deformation $\beta_{20} = 0.71$ compared to that to a state with initial deformation $\beta_{20} = 1.01$. The less deformed state lies below the second static fission barrier, and the more deformed state beyond. See text for further discussion.

For the giant resonance cases observed where the state is initially deformed below the static fission barrier (Fig. 4.2), the strong force drives the collective nuclear excitations. For the state with initial deformation $\beta_{20} = 1.01$, the oscillation of the Coulomb energy coincides with the oscillations of the deformation parameters shown in Fig. 4.8. The Coulomb force 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 of the deformation parameters are seen to follow large amplitude oscillations in phase with one another (second row from the bottom in Fig. 4.8).
Figure 4.13 (left panel) displays that the fluctuations in the Coulomb energy for the evolution of the state with $\beta_{20} = 0.71$, which is undergoing a collective giant resonance. The fluctuations in the Coulomb term are small compared to where the initial state is deformed beyond the fission barrier (right panel of Fig. 4.13). 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. For the cases beyond the barrier, it is the Coulomb term which is driving the large amplitude oscillations of the nuclear shape.

**Figure 4.14:** 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 hexadecupole degrees of freedom. As the static configuration was calculated with the Coulomb interaction, the overall deformation of the nucleus ‘collapses’ as the Coulomb force played a significant part in defining the initial configuration.

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. 4.14. Unsurprisingly, the figure shows a rapid decrease in elongation. This is as the static configuration was calculated with the Coulomb interaction; removing this will cause the nucleus to collapse into a more stable configuration. Most importantly, on top of the rapidly decreasing quadrupole and hexadecupole deformations, a small amplitude, high-frequency oscillation can be observed (left column in Fig. 4.14), which was not present in the corresponding panels in Fig. 4.8. The frequency of these vibrations in
the quadrupole and hexadecupole modes are qualitatively similar to those seen for the giant resonance cases (Fig. 4.2).

Overall, there is significant evidence suggesting the origins of the mechanism responsible for the slow, large amplitude oscillations of the nuclear shape observed for non-fissioning configurations beyond the static fission barrier. It lies in the competition between the Coulomb force trying to cause fission, and attractive nuclear potential terms in the energy functional countering this effect. As the states are evolved in time, they begin to explore significant collective motion as the nucleus attempts to find a pathway towards fission. It can only be speculated as to whether with a long enough time evolution the states eventually fission. Figure 4.8 shows, for example, in the bottom left panel (evolved from $\beta_{20}=1.07$), that the quadrupole deformation oscillates around a gradually increasing average. This increase is very slow, and performing a TDHF calculation to explore time evolution beyond 10,000 fm/c will be computationally expensive, and may begin to encounter numerical instabilities.

For the evolution of the states below the static barrier (Sec. 4.1.1), the required tunnelling through the barrier implies a forbidding time scale for fission within TDHF. For the non-fissioning cases investigated in this Section, where the initial deformation exceeds that of the second static fission barrier, the time scale for fission is at the very least inhibiting when performing TDHF calculations.

4.1.4 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 those initial states which fission upon time evolution from those which do not (states undergoing deformation-induced fission will be investigated in the next Section). Figure 4.15 displays this. The separating line (drawn between $\beta_{20}=1.07$ and 1.10 on Fig. 4.15) between the inhibiting and allowed regions for fission does not correspond to a threshold in the dynamic calculations. Figure 4.8 displays, 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 (see Fig. 4.8, bottom row).

For configurations starting with a deformation below the static fission barrier ($\beta_{20} < 0.860$), tunnelling is required to reach a fissioned state. This is forbidden in TDHF calculations. Beyond the barrier, it has been seen that there exists a region where fission is inhibited,
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Figure 4.15: Static one and two-fragment fission pathways (red and blue lines, respectively). For TDHF calculations three regions may be defined when following the one-fragment pathway; one in which fission is forbidden within TDHF time scales, one in which the time scale is inhibiting, and one which fission is allowed. The dividing lines are drawn at $\beta_{20} = 0.860$ and $\beta_{20} = 1.085$. See text for more details.

and it will shortly be demonstrated that beyond the intersection of the one and two-fragment pathways, fission is allowed within the considered time scales of the TDHF calculations. An intuitive explanation may be given for the significance of this point of intersection on the PES with regard to fission occurring upon time evolution.

In TDHF, energy is conserved. It is due to 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 4.15, $\beta_{20} > 1.085$), for a given value of $\beta_{20}$, the two-fragment state is more bound than the one-fragment state. Therefore, for a one-fragment state to evolve into a two-fragment configuration at a constant $\beta_{20}$, energy must be transformed from potential energy into excitation energy, which is allowed. Of course, the picture is not really that simple as the significance of the static PES becomes less clear in the dynamic case. Other configurations may be explored which do not correspond to the static fission pathways. 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 intuitive 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 the inhibited region of Fig. 4.15 (0.86 ≤ β_{20} ≤ 1.085), for a given β_{20} in the one-fragment pathway, the two-fragment state with the same β_{20} is less bound. Due to energy conservation, the one-fragment state cannot move to the two-fragment state at the same β_{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.

Further investigation of the link between static and dynamic configurations using Density Constrained time-dependent Hartree-Fock [60, 85], 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. A study of this nature, however, is beyond the scope of this thesis.

4.2 Fissioning States

For the static states with a quadrupole deformation at and beyond the threshold of β_{20} = 1.10, binary fission was seen to occur as the wave functions were evolved in time. The calculations to obtain the data for this Section were performed in a larger grid of size 48 × 48 × 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 due to the fragments approaching the grid boundaries.

Figure 4.16 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 β_{20} = 1.19. The scission point is difficult to define in a calculation involving quantum mechanical wave functions and densities. We take an operational approach and define it as the time when the point of 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 is also the point were a sizeable collective energy develops as the fission products begin spatially separating. For the case where the initial quadrupole deformation is β_{20} = 1.19 (presented
4.2 Fissioning States

in Fig. 4.16), it takes between 775-800 fm/c for the density between the two fragments to drop below this threshold. Figure 4.17 displays sample current vectors corresponding to the particle density slices presented in Fig. 4.16. The current vectors display the system smoothly transitioning into a two-fragment configuration; compared to Fig. 4.12 there is no dramatic rearrangement of the particle density during time evolution. Throughout the calculation, the currents in 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. 4.17 gradually increases as the fission occurs; beyond the point of scission they will increase rapidly as the fragments accelerate away from one another.

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

Figure 4.16: Snapshots of the time-dependent density starting from an initial state with $\beta_{20} = 1.19$. The isolines are separated by 0.05 particles/fm$^3$. It takes between 775 and 800 fm/c for scission (as defined in the text) to occur for this case.


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Figure 4.17: Sample current vectors corresponding to the slices of the particle density presented in Fig. 4.16. The vectors have been normalised to the same scale in each panel, so may be directly compared to one another. Note that the dimensions of the grid exceed what is presented in these panels; the densities are not experiencing spurious boundary effects.

Multipole moments for these states are shown in Fig. 4.18. These measurements have been sharply cut off at the point of scission. An analysis of the post-scission fragments will be presented in Sec. 4.3. Figure 4.18 shows the evolution of the nuclear shape up to the point of fission for the considered initial configurations. Different nuclear shapes are explored as the nucleus evolves from the various static states. Other than the case with static $\beta_{20} = 1.25$, as $\beta_{20}$ and $\beta_{40}$ increase, $\beta_{30}$ remains virtually constant.

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, depending 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
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Figure 4.18: Time evolution of the multipole deformation parameters for various static configurations observed to fission upon time evolution. Scission has been defined at the point where the particle density between the two fragments is less than 0.05 particles/fm³ along the principal axis of the system, and the measurements have been cut off at this point.

The distribution of fission products obtained with TDHF is in line with experimental investigations (see Sec. 4.3).

The time scale required for the initial configuration to fission varies. The least elongated case, with $\beta_{20} = 1.10$ takes $\approx 1250$ fm/c for scission to occur. Figure 4.18 shows the quadrupole deformation increasing rapidly for approximately 600 fm/c (red 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 the rate of increase of the quadrupole and hexadecupole parameters to accelerate.

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 is

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that with initial deformation $\beta_{20} = 1.25$. Figure 3.6 shows the initial density for this state (bottom right panel Fig.), and it seems that two fragments are already taking form, connected only by a thin elongated neck which rapidly dissipates into the top and bottom fragments upon time evolution.

Figure 4.19: 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 scission point. The total energy is conserved within fluctuations no greater than 4 MeV.

The evolution of the decomposed contributions to the energy density functional for the fissioning cases are presented in Fig. 4.19 (see Sec. 2.3.2 for the definitions of the terms). The decomposed energy density functional for the entire system is shown; the fragments will
be analysed separately in Sec. 4.3. Figure 4.19 displays the evolution of the energy density functional up to and *beyond* the point of scission. Total energy is conserved within the TDHF calculations, within fluctuations of less than 4 MeV (shown on the bottom right panel of Fig. 2.3.2).

The dynamic calculations allow translational motion and internal excitations. For the energy released in the fissioning case, nuclear binding energy is expected to be transformed mainly into the translational kinetic energy of the fragments. The following definition is usually employed in calculating the nuclear collective kinetic energy [57],

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

(4.5)

from the particle density $\rho(r)$ and the current density $j(r)$. This collective kinetic energy contains contributions from internal excitations of the nucleus, such as resonant excitations, and the translational kinetic energy of the post-fission fragments. It is presented in Fig. 4.19 (second panel from the bottom, right column), separately from the total kinetic energy. It is difficult to untangle the collective excitation energy attributed to the internal excitation of the fission fragments, to that attributed to translational motion. In fission reactions the energy release is typically attributed to $\approx 80\%$ 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 [4]. In our TDHF calculations, these effects may not be described. Therefore, we will demonstrate that the excitation energy of the fissioned system is dominated by the translational kinetic energy, with a small contribution from internal collective excitation of the fragments. This will be discussed in Section 4.3.4.

During time evolution, the individual components of the energy functional may be separately examined. The physical interpretation of the evolution of each term of the integrated energy functional shown in Fig. 4.19 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 (see to Sec. 2.3.2):

- **The $E_0$ and $E_3$ terms:** Both these terms are derived from the particle density. Although the total particle number is conserved throughout time evolution, the local particle density varies significantly during the fission process. As particles rearrange from one into two fragments, a neck region of low density emerges, resulting in a decrease in
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magnitude of both terms. Upon scission, the magnitude of these contributions rapidly increases as two dense local regions, corresponding to the two fragments, emerge. For both the $E_0$ and $E_3$ terms, the energy varies over a range of approximately 400 MeV during the time evolution of the considered states.

- $E_1$ term: The $E_1$ term contains contributions from the kinetic, particle, and current densities. The term looks qualitatively similar to the $E_0$ and $E_3$ terms (although the $E_0$ term is negative). This suggests that the density that governs the $E_0$ and $E_3$ terms, the particle density, is also the most relevant contribution governing the $E_1$ term. In comparison to the $E_0$ and $E_3$ terms, the energy varies over a much smaller range of approximately 25 MeV during time evolution.

- $E_2$ term: The $E_2$ term 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. This can be intuitively explained as the two-fragment system will have a combined surface region 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 is seen to 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 Coulomb term slowly decreases as the nucleus elongates. At the point of scission, the rate at which the term reduces rapidly accelerates as two charged fragments separate from one another in co-ordinate space. At infinite fragment separation, the Coulomb term will reduce to the contributions of the Coulomb energy for each nucleus, without further interactions. The reduction in the term is of the order of 200 MeV as the system evolves.

- 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. (4.5). The collective energy is initially small, corresponding to the internal currents as the nucleus slowly rearranges into a fissioned configuration (see inset in collective energy panel).
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The state with initial deformation $\beta_{20} = 1.10$ shows the most gradual transition to fission. An initial increase then decrease in collective energy is seen before scission, which corresponds to the (previously discussed) rapid initial elongation, then extended rearrangement phase as the configuration evolved (see Fig. 4.18). In contrast, the state with $\beta_{20} = 1.25$ is already close to the point of scission, 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. 4.19. The definition adopted for scission\(^1\) 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 contribution contains complex contributions from different densities, making it hard to qualitatively identify how the different densities contribute to the term. It is typically associated with shell effects, and the oscillations observed may indicate the importance of the term in the dynamic evolution of the nuclear state. 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 different shell effects are acting. Indeed, the masses of the fission products are significantly different from the other cases (see Sec. 4.3.2). The term has significant contributions from time-odd densities, so that the

\(^{1}\)Time at which neck density is below 0.05 particles per fm\(^3\) along the principal axis of the system.
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evolution can explore configurations which may not be accessible on the static PES. The term varies by less than 30 MeV during time evolution.

In experimental studies of fission, it is customary to measure the kinetic energy of the fission fragments. We can find an analogous observable within TDHF making use of the collective kinetic energy, defined by Eq. (4.5), and assume the translational kinetic energy dominates this term. Referring to Fig. 4.19, the collective kinetic energy and Coulomb energy are both expected to plateau as the separation of the two fragments becomes large. Unfortunately, the Coulomb force is long-ranged; the energy due to the interaction of two charged fragments reduces proportionally to $\frac{1}{r}$, where $r$ is the separation. Increasing the dimensions of the numerical grid is extremely computationally expensive. Therefore it is preferable to interpolate the collective kinetic energies to a large time to estimate the value 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 due to the Coulomb force, and convert all this energy into translational kinetic energy, energy conservation implies:

$$\frac{1}{2}M_u v_u^2 + \frac{1}{2}M_l v_l^2 = \kappa \frac{Z_u Z_l}{r}.$$  \hfill (4.6)

Here, $M$ are the masses of the upper and lower fragments, $v$ the velocity, and $Z$ the charge. The constant $\kappa$ is the Coulomb constant. As momentum must be conserved

$$M_u v_u + M_l v_l = 0,$$  \hfill (4.7)

the Eq. (4.6) may be rewritten, substituting for $v_u$

$$v_l^2 \left( \frac{M_l^2}{M_u} + M_l \right) = 2\kappa \frac{Z_u Z_l}{r}.$$  \hfill (4.8)

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

$$\frac{dr}{dt} = \sqrt{\frac{\Theta}{r}},$$  \hfill (4.9)

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

$$\int_{r_0}^{r} r^{1/2} dr = \int_{t_0}^{t} \sqrt{\Theta} dt$$  \hfill (4.10)
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allows the solution

$$r^{3/2} = r_0^{3/2} + \frac{3}{2} \sqrt{\Theta}(t - t_0)$$  \hspace{1cm} (4.11)

to be written. According to this approximation, $r$ 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, $E_{\text{Coul}} = E_{\text{col}}$), a fit of the form

$$f(t) = a + \frac{b}{(t - c)^{3/2}}$$  \hspace{1cm} (4.12)

can be performed to interpolate the collective kinetic energy to larger values of $t$. Figure 4.20 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 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 4.1 contains the values obtained for each of the fissioning cases with different distance fits. As a crude method to represent the uncertainty in the value, the mean value of the interpolated fits has been presented with the standard deviation as an uncertainty.

Figure 4.20: 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.

The values shown in Table 4.1 demonstrate that the resulting collective kinetic energy varies depending upon the region of the data the fit was performed to. The fragment deformation and the effect of particle emission (discussed in the next Section) may have to be
accounted for. This suggests that the results obtained from the interpolation method should serve only as illustrative values, due to the simplified model assumed.

Table 4.1: Interpolated total kinetic energy corresponding to different initial configurations. The fit of Eq. (4.12) has been performed once the fragment separation exceeds 30 fm, 50 fm and 60 fm.

<table>
<thead>
<tr>
<th>Static Quadrupole Deformation $\beta_{20}$</th>
<th>Coll. KE (30 fm fit) [MeV]</th>
<th>Coll. KE (50 fm fit) [MeV]</th>
<th>Coll. KE (60 fm fit) [MeV]</th>
<th>Mean ± St. Dev. [MeV]</th>
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<tr>
<td>1.10</td>
<td>210.3</td>
<td>206.5</td>
<td>203.4</td>
<td>206(4)</td>
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<tr>
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<td>210.8</td>
<td>200.0</td>
<td>193.7</td>
<td>202(8)</td>
</tr>
<tr>
<td>1.19</td>
<td>205.8</td>
<td>196.8</td>
<td>191.3</td>
<td>198(8)</td>
</tr>
<tr>
<td>1.25</td>
<td>193.4</td>
<td>180.8</td>
<td>176.3</td>
<td>183(9)</td>
</tr>
</tbody>
</table>

Table 4.2: Measured total kinetic energies from various experiments. The measurements correspond to the pre-neutron emission fragment energies.

<table>
<thead>
<tr>
<th>Method</th>
<th>Kinetic energy [MeV]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{240}$Pu(s.f)</td>
<td>178.85±0.30</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu(s.f)</td>
<td>179.00±0.08</td>
<td>[116]</td>
</tr>
<tr>
<td>$^{239}$Pu($n_{th}$, f)</td>
<td>177.69</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{239}$Pu($n_{th}$, f)</td>
<td>177.65±0.01</td>
<td>[116]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma$, f) (12 MeV)</td>
<td>176.39±0.24</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma$, f) (15 MeV)</td>
<td>175.80±0.24</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma$, f) (20 MeV)</td>
<td>175.15±0.24</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma$, f) (30 MeV)</td>
<td>174.98±0.31</td>
<td>[115]</td>
</tr>
</tbody>
</table>

In comparison to the experimentally measured kinetic energy of the fissioning systems displayed in Table 4.2, the theoretical values presented in Table 4.1 seem rather too high in most cases. However, the experimental values correspond to an average kinetic energy. We only have access to one 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. 4.3.3.

4.3 Fragment Analysis

Beyond the point of scission, it is preferable to consider a two-fragment system. The published distribution of Sky3D has some capacity to analyse two-fragment dynamics [57], and a
version has been modified further to investigate the fissioning system and extract some useful observables.

4.3.1 Masking

The masking procedure used to investigate the non-fissioning dynamic case can be generalised to investigate the fissioning system. 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. 4.21. An interpolation is 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 may then be deduced by integrating the densities either side of the dividing plane.

![Diagram of principal axis and dividing plane](image)

**Figure 4.21:** 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.

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. 4.22, 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 separated sufficiently so that the masks do not overlap (Fig. 4.22, 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.

Figure 4.22: Demonstration of masking procedure used. The particle density is represented with red isolines separated by 0.05 particles/fm$^3$. The masking functions (blue) are spherical Fermi functions around the centre of mass of the fragments. The isolines for the mask are (from closest to the fragment outwards) 1, 0.1 and 0.01. Measurements of observables corresponding to the individual fragments are only valid when the masks do not overlap (right panel).

4.3.2 Mass Distributions

As the post-fission fragments are excited, they may decay by particle emission. TDHF displays this decay by the dispersion of the 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 Chapter, this decay is of the order of 0.1-0.2 particles during the time evolution.

The dispersion of the wave functions, in addition to the fragments not having a well-defined (i.e. integer) particle numbers, makes identification of the fission products difficult.
4.3 Fragment Analysis

In line with experimental studies, the pre-emission fragments should be identified. This may be done by integrating the total density in each half of the numerical grid separated by the dividing plane. For this process, the masking procedure is not applied. For consistency, the measurement is taken as soon as the system identifies as a two-fragment system, that is, when the density connecting the fragments along the principal axis drops below 0.05 particles/fm$^3$. An uncertainty in the particle number of the fragments may be associated with the fluctuation of this measurement throughout time evolution, which is less than 0.05 particles for the considered cases.

Table 4.3: 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 (see Sec. 3.3.3).

<table>
<thead>
<tr>
<th>Static Deformation $\beta_{20}$</th>
<th>Heavy Fragment $A, Z$</th>
<th>Light Fragment $A, Z$</th>
<th>Heavy Frag. (Integer)</th>
<th>Light Frag. (Integer)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.10</td>
<td>136.33(5) , 52.78(5)</td>
<td>103.67(5) , 41.23(5)</td>
<td>$^{136}$I $^{53}$</td>
<td>$^{104}$Nb $^{41}$</td>
</tr>
<tr>
<td>1.13</td>
<td>135.02(5) , 52.23(5)</td>
<td>104.98(5) , 41.77(5)</td>
<td>$^{135}$Te $^{52}$</td>
<td>$^{105}$Mo $^{42}$</td>
</tr>
<tr>
<td>1.19</td>
<td>136.13(5) , 52.70(5)</td>
<td>103.87(5) , 41.30(5)</td>
<td>$^{136}$T $^{53}$</td>
<td>$^{104}$Nb $^{41}$</td>
</tr>
<tr>
<td>1.25</td>
<td>143.70(5) , 55.65(5)</td>
<td>96.30(5) , 38.35(5)</td>
<td>$^{144}$Cs $^{55}$</td>
<td>$^{96}$Sr $^{38}$</td>
</tr>
<tr>
<td>1.19$^{(2f)}$</td>
<td>132.81 , 50.84</td>
<td>107.04 , 43.13</td>
<td>$^{133}$Sb $^{51}$</td>
<td>$^{167}$Tc $^{43}$</td>
</tr>
</tbody>
</table>

Table 4.4: Some experimentally measured average masses following the fission $^{240}$Pu. The measurements for neutron-induced fission were taken before neutron emission of the fissioned fragments.

<table>
<thead>
<tr>
<th>Method</th>
<th>Heavy Fragment</th>
<th>Light Fragment</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{240}$Pu(s.f)</td>
<td>138.74±0.20</td>
<td>101.26±0.20</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu(s.f)</td>
<td>138.96±0.04</td>
<td>101.31±0.04</td>
<td>[116]</td>
</tr>
<tr>
<td>$^{239}$Pu($n_{th}, f$)</td>
<td>139.67</td>
<td>100.33</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{239}$Pu($n_{th}, f$)</td>
<td>139.73±0.01</td>
<td>100.27±0.01</td>
<td>[116]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma, f$) (12 MeV)</td>
<td>139.88±0.14</td>
<td>100.12±0.14</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma, f$) (15 MeV)</td>
<td>139.92±0.09</td>
<td>100.08±0.09</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma, f$) (20 MeV)</td>
<td>139.84±0.08</td>
<td>100.16±0.08</td>
<td>[115]</td>
</tr>
<tr>
<td>$^{240}$Pu($\gamma, f$) (30 MeV)</td>
<td>139.71±0.14</td>
<td>100.29±0.14</td>
<td>[115]</td>
</tr>
</tbody>
</table>

These fragment masses can be compared directly to experimental data. Table 4.3 displays the resulting mass distributions obtained from this theoretical study. The two-fragment static configuration is included for comparison (see Sec. 3.3). The Table also includes the particle...
number rounded to the nearest integer: although it is beyond the scope of this thesis, it would be of interest to project the individual fragments onto the particle number \([131]\) to obtain a distribution.

Table 4.4 contains experimental data taken from Refs. \([115, 116]\) 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.

It must be emphasised that fission produces a range of masses; the values quoted in Table 4.4 correspond to the most likely fissioned configuration. Referring to Fig. 4.23, 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 various energy neutron-induced fission; as the energy increases the distribution of the fission fragments becomes, on average, more symmetric (the central region of the mass distribution on the right panel of Fig. 4.23 can be seen to be filling in). 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 in excellent agreement with the experimental data.

\[\text{Figure 4.23: Independent fission yields for various energy neutron-induced fission processes. The data is from Ref. [142]. The red bars correspond to the binned TDHF results, normalised to the experimental results. The blue bar corresponds to the static two-fragment mass split. See text for more details.}\]

The result of the two-fragment static solution is also displayed on Fig. 4.23 for qualitative comparison (see Sec. 3.3.3). Although the fission products lie within the mass region of the...
experimental data, for the various static two-fragment configurations the mass split was seen to be constant (to an integer nucleon number). Time evolution of these two-fragment static solutions will not produce a varying mass distribution as the fragments are already well defined. The constant configuration of the two-fragment solution corresponds to an optimum static two-fragment configuration. The variation (and therefore distribution) of post-fission fragment masses will only arise due to the dynamic evolution of one-fragment static solutions, allowing configurations which do not correspond to two-fragment static solutions to be explored.

### 4.3.3 Energy of Fission Fragments

By applying masks around the spatial regions of the fission fragments, the energy density functional corresponding to the individual fragments may be obtained. However, interpreting the results is not simple in the two-fragment case. The nuclear part of the energy density functional is short-ranged, allowing the functional to be integrated in the spatial region corresponding to the individual fragments. The Coulomb interaction, however, is long-ranged; as well as the Coulomb interaction within the individual fragments, there is a contribution from their interaction with one another. Further, the fragments were seen to decay by particle emission, which will also impart some time dependence upon the integrated energy corresponding to the individual fragments.

**Figure 4.24:** Summed energy density functionals for the region of space corresponding to the heavy and light fission fragments. The quadrupole deformation of the initial state is labelled. The drift in the energy can be attributed mainly to the Coulomb interaction between the two fragments. See text for more details.
The time evolution of the total integrated energy functional corresponding to the heavy (upper) and light (lower) fission fragments is shown in Fig. 4.24. The time measurement at $t_{\text{sep}} = 0$ begins when the fragments are sufficiently separated such that the masks no longer overlap (see Fig. 4.22). To a good approximation, the fragment total energies are constant, but a slight drift is observed over time due to the long-range effects of the Coulomb interaction and particle decay. Only the total integrated energy of the post-fission fragments is shown in Fig. 4.24; the evolution of the decomposed terms corresponding to the individual fragments showed no remarkable behaviour. The fragment energy at the cutoff time will be denoted $E^*$. 

The total excitation energies of the fragments (which is the sum of the translational and internal collective kinetic energies) may be examined in comparison to calculations of the corresponding ground states. This method will complement the approach of interpolating the total collective kinetic energy of the system, as presented in Sec. 4.2, and should produce comparable results.

The solver Sky3D has been applied to deduce the ground states of the fission fragments (to the nearest integer particle numbers). Here, it is debatable that the energy functional in Sky3D contains all the terms required to calculated odd-odd and odd-even nuclei. The full time-odd contribution is presented in Ref. [77], 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.

**Table 4.5:** 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. 4.24), the ground state energy by $E_{gs}$, and the difference ($E^* - E_{gs}$) by $\Delta E$. See text for more details.

<table>
<thead>
<tr>
<th>Static $\beta_{20}$</th>
<th>Heavy Frag.</th>
<th>$E^*$ [MeV]</th>
<th>$E_{gs}$ [MeV]</th>
<th>$\Delta E$ [MeV]</th>
<th>Light Frag.</th>
<th>$E^*$ [MeV]</th>
<th>$E_{gs}$ [MeV]</th>
<th>$\Delta E$ [MeV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.10</td>
<td>$^{136}\text{I}$</td>
<td>-1029.22</td>
<td>-1118.31</td>
<td>89.09</td>
<td>$^{104}\text{Nb}$</td>
<td>-747.86</td>
<td>-854.54</td>
<td>106.68</td>
</tr>
<tr>
<td>1.13</td>
<td>$^{135}\text{Te}$</td>
<td>-1023.81</td>
<td>-1110.24</td>
<td>86.43</td>
<td>$^{105}\text{Mo}$</td>
<td>-757.38</td>
<td>-865.71</td>
<td>108.33</td>
</tr>
<tr>
<td>1.19</td>
<td>$^{136}\text{I}$</td>
<td>-1034.23</td>
<td>-1118.31</td>
<td>84.04</td>
<td>$^{104}\text{Nb}$</td>
<td>-749.41</td>
<td>-854.54</td>
<td>105.13</td>
</tr>
<tr>
<td>1.25</td>
<td>$^{134}\text{Cs}$</td>
<td>-1090.17</td>
<td>-1162.47</td>
<td>72.30</td>
<td>$^{96}\text{Sr}$</td>
<td>-697.78</td>
<td>-796.94</td>
<td>99.16</td>
</tr>
</tbody>
</table>
4.3 Fragment Analysis

Pairing has been neglected in these calculations. For odd numbers of protons or neutrons, the blocking approximation [21] must be applied if pairing is to be included. This approximation removes one state from the pairing scheme. An element of trial and error is required to determine which state must be removed. However, the total pairing contribution to the energy functional will typically be of the order of 0 – 10 MeV, which is small compared to the excitation energies in the fissioning case. Therefore, for a qualitative comparison of the excited state energy to the ground state energy, it is not unreasonable to neglect this correlation.

Two measurements of the total excitation energy of the system are now performed, either by interpolating the evolution of the collective kinetic energy, or by comparing the ground state fragment energies to the excited fragment energies. Figure 4.25 displays the mean interpolated collective kinetic energies presented in Table 4.1, compared the total fragment excitation energy ($\Delta E_{\text{heavy frag.}} + \Delta E_{\text{light frag.}}$) for each fissioning case. The error bars in the values of $\Delta E$ display an uncertainty of 10 MeV, which serves to take into account that pairing was not included for the ground state calculations, 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, with the standard deviation taken for the error (see Table 4.1).

![Figure 4.25](image)

**Figure 4.25:** Comparison of the mean interpolated collective kinetic energy (Table 4.1) to the summed $\Delta E$ (Table 4.5) for each of the fissioning cases. Error bars have been allocated to each case, see text for details.

Figure 4.25 shows that within the error bars, the results from the two techniques produce
consistent values of the energy released in the fission process. It would be of interest to obtain these values to a greater accuracy, both by performing the calculations up to the point where the Coulomb interaction is negligible, and obtaining ground state energies incorporating pairing correlations. It is interesting to note that the measurement $\Delta E$ is always lower, which may indicate a systematic effect worth further investigation. As mentioned, these are both measures of the total excitation energy of the system. Section 4.3.4 will demonstrate a technique which may be used to decouple the translational kinetic energy from the internal collective excitation energy.

![Figure 4.26](image)

**Figure 4.26:** Experimental data taken from Ref. [116]. The shaded box corresponds to the extremes of the range of kinetic energy values displayed in Fig. 4.25.

Figure 4.26 shows experimental measurements of the kinetic energy reproduced from Ref. [116] for thermal neutron-induced fission in $^{240}\text{Pu}$. The range of collective kinetic energies deduced in this Chapter 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 well with the experimental range of values.

### 4.3.4 Collective Excitation Modes of Fission Fragments

As mentioned, the excitation energy of the fission fragments is assumed to be dominated translational kinetic energy. However, as well as translational motion, the fragments undergo collective vibrations due to 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. 4.1. 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 $\hbar \omega = \frac{\pi}{T_{\text{obs}}}$ [139], giving an energy resolution of approximately 1.5 MeV for a signal lasting 1000 fm/c.

A novel approach was developed to extend the measurement time. Rather than performing the calculations in an unpractically large numerical grid, a Galilean transformation will be applied to the fission fragments to remove their linear momentum. The evolution of the static configuration with $\beta_{20} = 1.25$ will be presented as an example.

Inside the masked regions of space corresponding to the fragments, the linear momentum may be calculated (not containing the nucleon mass) by integrating the current density:

$$p_{\text{frag}} = \int j(r) dr.$$  \hspace{1cm} (4.13)

This momentum therefore has units of velocity [57]. The linear momentum of the fragments may then be instantaneously removed by applying the following Galilean boost to the single particle wave functions:

$$\tilde{\varphi}(r) = \exp \left( \frac{i \left( p_{\text{frag}} \cdot r \right)}{A_{\text{frag}}} \right) \varphi(r),$$  \hspace{1cm} (4.14)

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.

Figure 4.27 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, and the calculation terminated at separation 105 fm. Upon application of the transformation, the panels corresponding to the total energy increases in absolute magnitude by approximately 140 MeV as the collective kinetic energy drops by the same amount. This
corresponds to the excitation energy of the system due to translational motion being instantaneously removed. This invariably demonstrates that the total excitation energy is dominated by contributions from translational motion, rather than internal collective excitations.

The collective kinetic energy drops instantaneously to \( \approx 1.1 \) MeV following the transformation (see inset panel corresponding to collective kinetic energy in Fig. 4.27). As the translational energy is removed at this point, this remaining collective energy is the sum of the internal excitation energy shared between the two fragments. Reference [53] discusses an
4.3 Fragment Analysis

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 small compared to the total excitation en-
ergy released in the fission process, deduced to be $\approx 180\text{ MeV}$ (see Fig. 4.25, $\beta_{20} = 1.25$). 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. 4.26).

The energy functional (Fig. 4.27) may be compared to that in Fig. 4.19, where the calculation was terminated at the point where the transformation is applied in this case. Figure 4.27 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. 4.19 ($48 \times 48 \times 160$ 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.

As the Coulomb interaction is long-ranged, even at 100 fm separation there is an interaction between the fragments. Translational motion resumes after the Galilean transformation is applied, and the translational kinetic energy slowly increases (can be seen by the gradual increase of the collective kinetic energy in Fig. 4.27 following the transformation). Therefore, the centre of mass separation eventually reaches 105 fm and the calculation is terminated.

In principle, one could re-apply the Galilean transformation at every iteration to extend the time of the calculations to an even greater extent.

The evolution of the multipole deformation parameters are shown in Figs. 4.28 and 4.30, corresponding to the heavy and light fragment, respectively. The calculated power spectra are shown in Figs. 4.29 and 4.31. The resolution of the spectra is approximately 0.6 MeV, which is a significant improvement of the resolution of 1.3 MeV that would be obtained without extending the measurement time.

Within the resulting spectra presented for the heavy fragment in Fig. 4.29, there is a well-defined peak for each multipole parameter between 1-3 MeV. For the light fragment, Fig. 4.31 shows a well-defined peak for each multipole parameter between 4-6 MeV. Another noteworthy feature of the spectra is that both Figs. 4.29 and 4.31 share a defined peak in the quadrupole spectra at 4 MeV. This suggests that the two fragments share an excitation...
4. TIME EVOLUTION OF CONSTRAINED HARTREE-FOCK STATES

Figure 4.28: Evolution of multipole deformation parameters for the heavy fission fragment. The initial deformation was $\beta_{20} = 1.25$. The measurement time is significantly extended by applying the Galilean transformation to remove the linear momentum of the fragments. See text for more details.

Figure 4.29: Power spectra corresponding to Fig. 4.28 (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.

mode, despite the significant differences in mass. It may be of interest in a future study to compare the excitation modes obtained for the post-fission fragments (undergoing ‘hot’ resonances) to those obtained for the ground state modes of the corresponding nuclei.
Figure 4.30: Evolution of multipole deformation parameters for the light fission fragment. The initial deformation is $\beta_{20} = 1.25$.

Figure 4.31: Power spectra corresponding to Fig. 4.30 (light fission fragment).

Overall, this Chapter has presented a variety of fission products resulting from deformation-induced fission processes, with different excitation characteristics. Within the results described by TDHF, DIF seems to be a slow, gradual process, where the collective energy is small up until around point of scission. At this point, the translational kinetic energy rapidly increases. This translational kinetic energy corresponds to the dominant form of en-
ergy release (> 99% in the case examined). The experimentally measured neutron-induced fission products and kinetic energies compare well with the results obtained using TDHF. This demonstrate the potential of applying time-dependent Hartree-Fock to describe fission phenomenon within a fully microscopic framework.
Boost-Induced Fission using Time-Dependent Hartree-Fock

This Chapter is concerned with investigating methods which induce fission for initial configurations where the process is either forbidden or inhibited within the time scale of time-dependent Hartree-Fock (see Fig. 4.15). This will be referred to as boost-induced fission (BIF), in contrast to the cases of deformation-induced fission (DIF) presented in the previous Chapter.

Large-amplitude collective motion may be induced by applying an external field to the system. A reasonable choice for this external field is one which will provide a quadrupole excitation, in line with References [55, 56]. The external excitation field may be applied instantaneously, or within a time-dependent profile. Both cases will be discussed.

5.1 Instantaneous Velocity Boosts

An external field may be instantaneously applied to the system at $t = 0$ by applying the gauge transformation $e^{i\phi(r)}$ to the single-particle wave functions. This corresponds to a velocity boost which carries the profile of $\nabla \phi(r)$. The spatial profile, $\phi(r)$, is chosen here to be proportional to a quadrupole field. Due to the gauge invariance of the Skyrme interaction, the energy added by this boost is purely in the form of collective kinetic energy [78, 143] (see Appendix A).

Two initial configurations will be investigated; the fission isomer of $^{240}$Pu ($\beta_{20} = 0.682$), and a state just beyond the peak of the second fission barrier, with static deformation $\beta_{20} = 0.890$. The isomer sits in a region where fission is forbidden within TDHF time.
scales, whereas the state with deformation $\beta_{20} = 0.890$ sits in the region where fission is inhibited within the timescale of feasible TDHF calculation (see Fig. 4.15).

5.1.1 Application of an Instantaneous Quadrupole Excitation Fields

To investigate BIF, an instantaneous quadrupole excitation field will firstly be applied to the static isomeric state. The current vectors $\mathbf{j}(\mathbf{r})$ resulting from the application of this velocity field at $t = 0$ are shown in Fig. 5.1, alongside the particle density.

![Particle Density (t=0) and Current Vectors (t=0)](image)

Figure 5.1: 2D slice of the 3D particle density and current vectors $\mathbf{j}(\mathbf{r})$ for a quadrupole velocity field applied instantaneously to the isomeric state ($\beta_{20} = 0.68$) at time $t = 0$. The current vectors have been normalised to a visually instructive length.

The energy added by the instantaneous excitation field may be calculated in a straightforward manner due to the simple form of the quadrupole operator (see Appendix A). The scaling factor $\eta$ may be chosen to provide an excitation of $\Delta E_{\text{kin}}$ using the relation

$$\eta = \sqrt{\frac{\Delta E_{\text{kin}}}{\beta^2 A \langle |\nabla \phi(\mathbf{r})|^2 \rangle}}, \quad (5.1)$$

where $A$ is the particle number and $\phi(\mathbf{r})$ the spatial profile of the excitation field, taken to be

$$\phi(\mathbf{r}) = 2z^2 - x^2 - y^2. \quad (5.2)$$

Figure 5.2 shows the evolution of the deformation parameters following quadrupole velocity boosts applying different amounts of energy to the isomeric state. In all cases an initial, rapid increase in quadrupole deformation may be seen within the first 50 fm/c (top left panel of Fig. 5.2). Following this, for an excitation below the threshold energy required to induce
fission, the nucleus draws back to its original quadrupole deformation, and then begins low frequency, large-amplitude vibrations (red, green and blue lines in Fig. 5.2). As the initial configuration is mass symmetric, and the excitation was of a pure quadrupole nature, no octupole deformation is induced as the mass symmetry is conserved (top right panel of Fig. 5.2).

In all cases, the evolution of the hexadecupole deformation demonstrates that the nucleus necks significantly between 100-150 fm/c, which can be seen by the characteristic drop in magnitude as the elongation increases (bottom left panel of Fig. 5.2). This was seen in the evolution of the static configurations for the constrained Hartree-Fock (CHF) calculations presented in Fig. 3.7. This behaviour was not observed in the DIF cases (Fig. 4.18), as the initial configurations were deformed such that they were already displaying significant necking.

Figure 5.2 shows that the threshold energy for inducing fission with this field is \(175 \leq E_{\text{thresh.}} \leq 200\) MeV. Scission occurs between 950 and 1000 fm/c for the 200 MeV boost.
system smoothly evolves to a fissioned configuration (see Fig. 4.18). These oscillations may be interpreted as an effect due to the large excitation energy applied, and will be commented upon further shortly.

Figure 5.3 displays slices of the 3D particle density for various times for the fissioning case (200 MeV excitation) presented in Fig. 5.2. The resulting mass symmetry of the fission fragments suggests that this process is not inducing fission in a manner comparable to DIF, where a range of fission products with asymmetric masses were observed, which agreed well with sample experimental results (see Fig. 4.23).

Figure 5.3: 2D slices of the 3D density at various times following an instantaneous 200 MeV quadrupole excitation upon the isomeric state. The isolines are separated by 0.05 particles/fm$^3$.

The results quash the naive assumption that the instantaneous quadrupole excitation will simply move the nucleus by the corresponding energy along the static potential energy surface (PES). The static PES fission barriers were seen to be $\approx 10$ MeV (see Fig. 3.6), and the energy required for BIF is an order of magnitude greater than this. All of the energy
5.1 Instantaneous Velocity Boosts

from the boost is imparted in the form of collective kinetic energy, and in both the static and
dynamic cases, the potential energy is identical at $t = 0$. Therefore, the instantaneous boost
causes the picture to depart from the static PES at $t = 0$, as the dynamic state now contains
considerable internal excitation, despite the initial shape being identical.

![Diagram of energy contributions]

**Figure 5.4:** Integrated contributions to the energy density functional following an instantaneous
excitation applied via a quadrupole field. Two cases are presented, one displaying BIF, and one
where the system fails to fission. Vertical lines in the panels displaying the total and collective
kinetic energies correspond to the point of scission for the 200 MeV case. The terms of the
functional are defined in Sec. 2.3.2. See text for more details.

The time evolution of the energy density functional is shown in Fig. 5.4, comparing a
non-fissioning and fissioning case following the application of an instantaneous quadrupole
velocity boost to the isomeric state. In both cases, the initial boost energy is imparted to the system as collective kinetic energy at $t = 0$. Figure 5.4 shows the resulting evolution of the energy functional for a 150 MeV instantaneous excitation (red lines) compared to a 200 MeV excitation (green lines), which demonstrates BIF. For both cases presented, other than the collective and total energy, all of the contributions to the energy functional are initially identical.

Looking at the evolution of the collective kinetic energy (second panel from the bottom, right column) in Fig. 5.4, the collective kinetic energy provided by the excitation field is rapidly absorbed into the nuclear terms of the energy functional. The bulk of the excitation energy is absorbed within the first 50-100 fm/c, and by 200-250 fm/c a roughly constant collective energy ($\approx 5$ MeV) remains for the fissioning case up to the point of scission (see inset panel). This $\approx 5$ MeV corresponds to internal currents induced by the boost as translational motion does not set in until after scission occurs. As it will be seen, this significant internal collective excitation energy corresponds to a dramatic behaviour as the densities rearrange into a fissioned configuration. For the DIF case (Fig. 4.19), the collective energy was orders of magnitude smaller up until around the point of scission, which emphasises that the process

Figure 5.5: 1D slices of the particle density along the principal axis, following the application of an instantaneous 200 MeV quadrupole excitation upon the isomeric state. The solid red lines represent the density slice at the specified time, and the dotted green line corresponds to the density slice in the previous panel, which has been included for visual comparison.
Figure 5.5 displays 1D slices of the particle density along the principal axis of the nucleus for different times following the application of the 200 MeV instantaneous excitation, which is instructive when examined in conjunction with the 2D density slices presented in Fig. 5.3. It can be seen in Fig. 5.5 that the particle density follows an initial rapid elongation (comparing the density at 0 fm/c to 50 fm/c). This corresponds to the initial drop in absolute magnitude of the terms in the EDF (Fig. 5.4). This is as the particle density is rapidly stretched and spread out over a larger volume. By considering the definitions of the terms in the energy functional (see Sec. 2.3.2), a stretched shape with a sparse local particle density will cause a decrease in magnitude in all the terms in the energy functional where the particle density contributes significantly.

Following the initial stretching of the nucleus between 0-50 fm/c, Fig. 5.5 displays the 1D density slice being drawn back sharply ($t=100$ fm/c in Fig. 5.5). The 1D slice of the particle density at this time displays a prominent dip around $z = 0$. The particle density is now spatially localised at the two ends of the nucleus, causing the terms in the energy functional to rapidly increase in magnitude (Fig. 5.4). It is following this drawing back that necking begins to develop, which can be seen when examining the particle density at $t = 100$ fm/c in Fig. 5.5. The current vectors corresponding to Fig. 5.5 are displayed in Fig. 5.6, and they provide a useful visual aid when examined in conjunction with the particle density slices. The current vectors at $t = 100$ fm/c demonstrate necking occurring as the particle flow draws in at the neck region. This coincides with the time at which the characteristic behaviour of the hexadecupole deformation parameter corresponding to necking occurs, as seen in the bottom left panel of Fig. 5.2.

By 250 fm/c, the density is seen to be more evenly distributed over the slice along the $z$ axis (Fig. 5.5), causing the amplitude of the oscillating terms in the energy functional (Fig. 5.4) to reduce somewhat in comparison to the behaviour at earlier times. The energy from the boost has been mostly absorbed into the nuclear potential part of the energy functional, and $\approx 5$ MeV of collective energy remains, corresponding to the currents which the boost has induced. The $E_2$ term of the energy functional (left column, second panel from top in Fig. 5.4) for the fissioning case levels off at approximately 375 MeV, in comparison to the non-fissioning case at 325 MeV. For the fissioning case, the extra energy in this term is due to the
nucleus necking as it fissions, which creates a greater surface compared to the non-fissioning system.

Beyond 250 fm/c, the density gradually transitions into a fissioned configuration (Fig. 5.3). Oscillations in the deformation can be seen (Fig. 5.2) as the shape evolves, this ‘sloshing’ is related to the internal current induced by the boost. They correspond to the oscillations seen in the evolution of the terms in the energy functional (Fig. 5.4) as the configuration varies. The current vectors presented in Fig. 5.6 complement the particle density slices presented in Fig. 5.3 to give further insight of the dynamics of the system. Compared to the sample slices of the current density presented for the DIF case (Fig. 4.17), the evolution of the current density is far more dramatic for instantaneous BIF. The vectors show that following the

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**Figure 5.6:** Current vectors corresponding to the particle density slices presented in Fig. 5.5. The normalisation factors for the vector arrows from $t = 250$ to $t = 900$ are the same ($=N$), and for other times the normalisation factor has been rescaled so that the panels are visually instructive. The normalisation factors are $N/10, 3N/10$ and $3N/10$ for $t = 0, t = 50$ and $t = 100$, respectively, and $4N/10$ and $3N/10$ for $t = 1000$ and $t = 1100$. 
5.1 Instantaneous Velocity Boosts

initial stretching phase ($t = 0$ to $t = 50$ fm/c), as the current vectors reverse direction and the drawing in phase begins (between $t = 50$ and $t = 250$ fm/c), a shockwave-type behaviour sets in. We define this shockwave-type behaviour to correspond to an area with well-defined current vectors moving in phase in opposite directions, leaving a static void behind. This is perhaps most clearly observable in the panel of Fig 5.6 corresponding to $t = 600$ fm/c.

The oscillatory nature of the deformation parameters (Fig. 5.2) and decomposed terms of the energy functional (Fig. 5.5) suggest that after the initial shockwave occurs (as the direction of the currents is reversed between 50 and 100 fm/c), the behaviour continues and another shockwave occurs when the particle flow hits the central region, and reverses direction once more. This corresponds to the sloshing effect seen in the evolution of the densities and the energy functional. This behaviour continues until 800-900 fm/c, where beyond this point the nucleus has rearranged such that it can evolve into a fissioned configuration.

In the evolution of the $E_0$, $E_1$ and $E_3$ terms for the BIF case (Fig. 5.4, green line), around the point of scission ($\approx 900$-1000 fm/c) there is a small increase in the average value which the oscillations are based around. By comparing the average of the oscillating values of the terms before and after fission, the $E_0$ and $E_3$ terms increases by 250-300 MeV, and the $E_1$ term by 15-25 MeV. The oscillatory nature of the evolution of these terms make these values approximate, but they may be compared to the typical changes in magnitude observed in the DIF case at the point of scission (Fig. 4.19) of $\approx 400$ MeV for the $E_0$ and $E_3$ terms, and $\approx 25$ MeV for the $E_1$ term. These differences may be attributed to the different final fission products for the BIF case considered here to the DIF cases considered previously, as it is the local particle densities within the fragments that will determine the post-scissioned values of the $E_0$ and $E_3$ terms.

A similar investigation of BIF using instantaneous excitation fields may be considered, starting from the static state with quadrupole deformation $\beta_{20} = 0.89$. The state lies just beyond the peak of the second static fission barrier, and was seen in the previous Chapter to fail to fission within a time evolution of 9000 fm/c (see Fig. 4.8). For this static state, mass asymmetry is present. Due to this, octupole degree of freedom may be explored. Figure 5.7 shows the evolution of the multipole parameters following quadrupole excitations of various energies. It can be seen in the top right panel of Fig. 5.7 that for the fissioning case the octupole deformation parameter increases in magnitude from $\beta_{30} = -0.3$ to $-0.7$ by the
Figure 5.7: Evolution of multipole moments for the initial state with $\beta_{20} = 0.89$, following an instantaneous quadrupole excitation. For the fissioning case following a 225 MeV boost, scission occurs at around 1700 fm/c, and the measurements of the multipole deformation parameters are cut off at this point.

point of scission. Figure 5.7 demonstrates that the threshold energy required to induce fission for this state is in the range $200 \leq E_{\text{thresh}} \leq 225$ MeV. Figure 5.8 shows the time evolution of the particle density following an instantaneous quadrupole excitation delivering 225 MeV of energy. The Figure demonstrates that asymmetric fission fragments are indeed produced within this BIF process, with scission occurring between 1700-1750 fm/c.

The threshold energy for BIF is greater in this case (225 MeV) compared to the 200 MeV boost required for the isomeric state (Fig. 5.2). This is a surprising result when considering the static PES. One may have assumed as the initial configuration is more deformed ($\beta_{20} = 0.89$) compared to the isomer ($\beta_{20} = 0.68$), that less energy should be required to induce fission. However, as previously mentioned, by applying an instantaneous boost the state is removed from the static configuration and the corresponding PES. At $t = 0$, despite the particle density being identical to the static configuration, the boosted state contains a large excitation in the form of collective kinetic energy. It can be reasonably assumed that this highly excited state does not correspond to the static counterpart and a violently energetic evolution will commence. Within the first few hundred fm/c, the bulk of the excitation energy is absorbed into the nuclear terms of the EDF, and the configurations encountered during this time (see Figs. 5.3 and 5.8, for example) do not resemble anything encountered on the static PES (Fig.
Figure 5.8: 2D slices of the 3D density at various times for the case of an instantaneous 225 MeV quadrupole excitation upon the state with initial deformation $\beta_{20} = 0.89$. The isolines are separated by 0.05 particles/fm$^3$.

Table 5.1: Threshold energies and masses obtained from applying instantaneous excitation fields to the fission isomer. The masses for the minimum energy case observed to induce fission are presented. The interpolated collective energy, corresponding to (mainly) translational kinetic energy, is performed using the same procedure detailed in Sec. 4.2

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<tbody>
<tr>
<td>Isomer</td>
<td>200</td>
<td>120.00(5)</td>
<td>120.00(5)</td>
<td>$^{120}$Ag</td>
<td>$^{120}$Ag</td>
<td>218(8)</td>
</tr>
<tr>
<td>$\beta_{20} = 0.89$</td>
<td>225</td>
<td>150.50(5)</td>
<td>89.49(5)</td>
<td>$^{151}$Pr</td>
<td>$^{89}$Br</td>
<td>189(6)</td>
</tr>
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Table 5.1 displays some of the properties of the fission fragments produced when applying
the threshold instantaneous boosts required to observe BIF for the isomer and the state with static deformation $\beta_{20}=0.89$. Application of the instantaneous excitation field to the state with initial deformation $\beta_{20}=0.89$ results in asymmetric fission products. The bulk of the excitation energy is in both cases absorbed into the nuclear terms of the energy functional within the first few hundred fm/c (Fig. 5.4 is representative), and the remaining excitation is on the form of internal collective kinetic energy as a current is induced. Scission does not occur until well after the boost is applied ($\approx 1000$-$2000$ fm/c, see Figs. 5.3 and 5.8), demonstrating that it is not the boost itself that directly induces fission, but rather that it provides the collective energy required for the densities to rearrange into a fissioned configuration. The final collective kinetic energies of the fissioned systems have been deduced using the interpolation procedure described in Sec. 4.2. As discussed in Sec. 4.3.3, the collective energy may be assumed to be dominated by the translational kinetic energies of the fragments. The mass distributions obtained for this state ($\beta_{20}=0.89$) by BIF will be compared to experimental data and DIF results in Sec. 5.2.5.

Table 5.2: Fission products obtained by BIF, applying an instantaneous quadrupole excitation of various energies to the state with initial deformation $\beta_{20}=0.89$. The collective kinetic energy, corresponding to the translational kinetic energy of the system, has been interpolated using the procedure detailed in Sec. 4.2.

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<tr>
<td>225</td>
<td>150.50(5), 58.78(5)</td>
<td>89.49(5), 35.23(5)</td>
<td>$^{151}\text{Pr}$</td>
<td>$^{89}\text{Br}$</td>
<td>189(6)</td>
</tr>
<tr>
<td>250</td>
<td>147.61(5), 57.71(5)</td>
<td>92.47(5), 36.28(5)</td>
<td>$^{148}\text{Ce}$</td>
<td>$^{92}\text{Kr}$</td>
<td>189(2)</td>
</tr>
<tr>
<td>300</td>
<td>147.06(5), 57.50(5)</td>
<td>92.92(5), 36.50(5)</td>
<td>$^{147}\text{Ce}$</td>
<td>$^{93}\text{Rb}$</td>
<td>188(4)</td>
</tr>
<tr>
<td>350</td>
<td>148.37(5), 58.10(5)</td>
<td>91.62(5), 35.90(5)</td>
<td>$^{148}\text{Ce}$</td>
<td>$^{92}\text{Kr}$</td>
<td>180(3)</td>
</tr>
<tr>
<td>400</td>
<td>150.61(5), 58.51(5)</td>
<td>89.37(5), 35.48(5)</td>
<td>$^{151}\text{Pr}$</td>
<td>$^{89}\text{Br}$</td>
<td>176(11)</td>
</tr>
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So far, only the threshold instantaneous BIF cases have been presented. Use of instantaneous quadrupole boosts which impart more energy than the deduced threshold are now briefly investigated for the configuration with static deformation $\beta_{20}=0.89$. Table 5.2 presents the fission products following different energy excitations. By interpolating the collective kinetic energy of the systems following the different boosts, the translational kinetic energy of the system may be deduced by assuming that the collective energy is dominated by translational kinetic energy. To verify that this assumption still stands, the interpolated
5.1 Instantaneous Velocity Boosts

collective energy for the 225 MeV excitation may be compared to the 400 MeV excitation, as the final fission fragments are the same. Therefore, due to the Coulomb interaction imparting the translational kinetic energy, the resulting values should agree within uncertainties if the contribution from internal collective excitations are small. The resulting interpolated energies agree within uncertainties, demonstrating that the energy released in BIF is still dominantly translational kinetic energy, even for boost energies beyond the threshold for fission. It can also be seen from Table 5.2 that the interpolated collective kinetic energies, which are interpreted as the energy release, agree within uncertainties for a charge difference of ±1 in the fission products.

The evolution of the energy functionals for the 225 and 400 MeV excitations applied to the state with $\beta_{20} = 0.89$ display two different time scales for fission (Fig. 5.9). Scission occurs within 450 fm/c for the 400 MeV excitation, and takes approximately 1650 fm/c for the 225 MeV excitation. The point of scission is marked on the panels corresponding to the kinetic and collective kinetic energies in Fig. 5.9. In both cases presented, the bulk of the initial excitation energy is absorbed within the first 0-100 fm/c (Fig. 5.9, right column, second panel from bottom). The period of the oscillations in the other terms of the energy functional are initially similar for both energy boosts (within the first 0-200 fm/c), although the amplitude of the oscillations for the 400 MeV case are larger.

To explain the differences in the fission timescales when applying the 225 and 400 MeV excitations to the state with initial deformation $\beta_{20} = 0.89$, it is helpful to examine 1D slices of the particle densities for both cases as the system evolves. These are displayed in Figs. 5.10 and 5.11 for the 225 and 400 MeV excitations, respectively. For the 225 MeV excitation, where a longer fission timescale is observed, the behaviour is similar to that where a 200 MeV excitation was applied to the isomer (Fig. 5.5). Following the application of the boost, the nucleus is stretched, and then draws sharply back in within the first 100 fm/c (Fig. 5.10). By 300 fm/c, virtually all of the excitation energy has been absorbed, and the remaining collective energy corresponds to the induced current (Fig. 5.9). The density has recovered in the central region (Fig. 5.10, $t = 300$), and it is here that the shockwave behaviour sets in, as seen previously for the isomer (most clearly when analysing the current vectors in Fig. 5.6). During this phase, the densities slosh around as the particle flow travels outwards, then sharply reverses direction, and continues oscillating in this manner. Beyond 1600 fm/c, the
particles in the neck have mostly transitioned into the two lobes, and the Coulomb repulsion drives the configuration to scission.

When applying the 400 MeV excitation, a much faster timescale is observed for fission (Fig. 5.9). Figure 5.11 displays the corresponding 1D density slices as the system evolves, and as is seen from the evolution of the energy functional (Fig. 5.9), the amplitude of the oscillations in the decomposed terms for the first 500 fm/c are much larger than the 225 MeV case. This corresponds to the more significant current induced by the excitation, and it
5.1 Instantaneous Velocity Boosts

can be seen by comparing the collective kinetic energy of the system that $\approx 20$ MeV of collective kinetic energy remains (400 MeV boost) in comparison to $\approx 10$ MeV (225 MeV boost) following the initial absorption phase (Fig. 5.9, inset panel). This suggests that the shockwaves in the evolution of the densities are more energetic, which can be verified by the larger amplitude of the oscillations of the terms in the energy functional within the first 500-750 fm/c (Fig. 5.9). This allows the configuration to rearrange more rapidly. Indeed, the initial state requires far fewer oscillations of the particle flow moving outwards then inwards before the nucleus rearranges such that the Coulomb repulsion drives the configuration into two fragments. Here, scission occurs between 400 and 500 fm/c.

It is interesting to observe the differences in the $E_0 - E_3$ and spin-orbit terms in Fig. 5.9 once the systems have fissioned. This suggests that the final fragments have different deformations (that is, the particle density is arranged differently), despite having the same $N$ and $Z$. This difference in shape configuration results from the trajectory followed to fission due to the differences in the energy deposited by the boosts and the currents induced.

Increasing the energy beyond that which was observed to produce the faster fission timescale has been observed to induce a different fission process entirely, producing ternary...
5. BOOST-INDUCED FISSION USING TIME-DEPENDENT HARTREE-FOCK

Figure 5.11: 1D slices of the particle density along the principal axis of the nucleus for different times, following an instantaneous quadrupole excitation field depositing 400 MeV of energy into the system. The red lines display the density slice at the marked time, and the dotted green line shows the density in the previous panel.

Figure 5.12: 1D slices of the particle density along the principal axis of the nucleus for different times, following an instantaneous quadrupole excitation field depositing 800 MeV of energy into the system. The red lines display the density slice at the marked time, and the dotted green line shows the density in the previous panel. Ternary fission is observed for this BIF process.
5.2 Time-Dependent External Fields

As it has been demonstrated, the threshold excitation to observe BIF with an instantaneous boost requires an excitation depositing energy of the order of 200 MeV for the cases considered. As the energy is all deposited at \( t = 0 \), the correspondence between the static configuration and the state which is time-evolved is distorted due to the instantaneous, high-energy excitation. This leads to a dramatic evolution of the state. Adding energy to the system gradually may allow the densities time to smoothly evolve into a fissioned configuration, in a manner comparative to DIF. In this Section, the external excitation will be applied gradually via a time-dependent profile. The isomer and configuration with \( \beta_{20} = 0.89 \) will be considered. The spatial form of the excitation will be kept as a quadrupole field, consistent to that considered in the previous Section (Eq. (5.2)).

The single-particle Hamiltonian \( \hat{h}_q \) acting on the proton and neutron states can be modified to include the time-dependent external field \( U_{\text{ext}, q}(r, t) \), which was chosen to be isoscalar, affecting both the proton and neutron states in an identical manner. The single particle Hamiltonian is augmented by [57]

\[
\hat{h}'_q(t) = \hat{h}_q(t) + U_{\text{ext}, q}(r, t).
\]  

(5.3)

Here, the external field \( U_{\text{ext}, q}(r, t) \) is given by

\[
U_{\text{ext}, q}(r, t) = \eta f(t) \phi_q(r).
\]  

(5.4)

The constant \( \eta \) must be tuned as it scales the amount of energy added to the system, and \( \phi_q(r) \) is the spatial profile of the field (chosen to be proportional to the quadrupole field). It is more difficult to tune \( \eta \) to add the desired energy to the system, due to the non-trivial way it enters the time evolution compared to the case of the instantaneous boost (Eq. (5.1)). The
temporal profile of the excitation field is characterised by $f(t)$, and in all cases considered it takes the Gaussian form:

$$f(t) = \exp \left( -\frac{(t - \tau_0)^2}{\Delta \tau^2} \right). \quad (5.5)$$

The profile is centred around $\tau_0$, and has a width $\Delta \tau$. Values of $\tau_0$ will be investigated in the region 150-800 fm/c and $\Delta \tau$ will be taken as approximately $\frac{\tau_0}{3}$.

5.2.1 Applying Time-Dependent External Fields to the Fission Isomer

Boost-induced fission can be investigated using a time-dependent quadrupole excitation field. As it was seen before, due to the mass symmetry of the initial state and the choice of excitation field, no octupole deformation is induced during the time evolution. An initial choice of $\tau_0 = 500$ fm/c and $\Delta t = 150$ fm/c was adopted to describe the temporal profile of the external field. Figure 5.13 displays the evolution of the multipole moments subject to an external field with different strengths $\eta$. For the fissioning case, (blue line on Fig. 5.13, $\eta = 0.0095$), the evolution of the multipole moments of the system have been sharply cut off at the point of scission. In those cases where the nucleus fails to fission, the quadrupole deformation reverts back to the original value once the external excitation ends (red and green lines in Fig. 5.13). Oscillations in the quadrupole and hexadecupole degrees of freedom (left column in Fig. 5.13) are visible beyond this.

Figure 5.14 displays 2D slices of the particle density for the fissioning case ($\eta = 0.0095$). Symmetric fission into two $^{120}$Ag fragments is observed, identical to the case where the instantaneous quadrupole excitation field was applied (Fig. 5.3). When applying the time-dependent excitation field (Fig. 5.14), visible deviations from the initial density are only visible after 300-400 fm/c, whereas Fig. 5.3 shows a dramatic immediate change in the nuclear configuration following the instantaneous excitation. The timescale for scission to occur is comparable the instantaneous BIF case (Fig. 5.3), requiring approximately 1050 fm/c for the time-dependent field and 950 fm/c for the instantaneous boost.

The current vectors corresponding to the particle density slices presented in Fig. 5.14 are shown in Fig. 5.15. The observed behaviour may be compared to the DIF example (Fig. 4.17) and the BIF case presented where an instantaneous boost was applied to the static isomer (Fig. 5.6). For the time-dependent BIF case, Fig. 5.15 shows current vectors where the flow of particles moves outwards into the two fission fragments, which gradually increase
in magnitude up to 800 fm/c. At this point the external field becomes negligible. Beyond 800 fm/c, a current has been induced, and the nucleus has reconfigured itself so that it evolves to fission without further influence from the excitation field. It will be shown that the collective energy corresponding to this current is small compared to the instantaneous BIF case. The system continues evolving into a two-fragment configuration without the flow of particles begin drawn back inwards, similarly to the behaviour seen for the DIF case (Fig. 4.17). In the DIF case, the currents in the two forming fragments point in opposite direction, with little contribution from the neck region. Here, for the time-dependent BIF case, there are initially far more particles in the neck (the initial configuration is less deformed), resulting in a significant particle flow in this region, especially around 600-700 fm/c (Fig. 5.15).

Compared to the instantaneous BIF case (Fig. 5.6), where a shockwave-type behaviour was seen in the time evolution of the current density, the behaviour observed as the configuration evolves when using a time-dependent external field (‘temporally extended’ BIF) is far less dramatic. Unlike instantaneous BIF, there is no sloshing or oscillations in the density during the time evolution. This suggests a physically different transition to the fissioned state is occurring for temporally extended BIF, similar to that seen for DIF.

It is instructive to examine the integrated contributions to the energy functional for all
three cases of the time-dependent excitation field (where one leads to BIF) presented in Fig. 5.13. The decomposed energy functionals are presented in Fig. 5.16. The Figure displays the temporal profile of the external field as a visual aid in the bottom left panel. The energy added to the system by the field for the different scaling parameters $\eta$ can be read off the panel corresponding to the total energy (bottom right). It is remarkable to observe that fission has been induced in the case where $\eta = 0.0095$ by adding 52 MeV of energy to the system. A threshold of $41 \leq E_{\text{thresh}} \leq 52$ may be deduced. This compares to $175 \leq E_{\text{thresh}} \leq 200$ MeV which was required for the instantaneous quadrupole boost (Fig. 5.2). When applying a time-dependent excitation field, energy is deposited into the system in a more gradual manner. In doing so, less excitation energy is needed to bring the system to fission. This suggests that not only energy deposition, but also the timescale for the energy deposition, matters in terms of fission dynamics.
Figure 5.15: Current vectors corresponding to the particle density slices presented in Fig. 5.14. The external field peaks at $\tau_0 = 500$ fm/c with width $\Delta \tau = 150$ fm/c, so the excitation field has reduced to a negligible magnitude by $t = 800$ fm/c. The vectors have been normalised to be visually instructive. The normalisation factor $(N)$ is the same in each panel, other than $t = 1100$ fm/c, where it is $3N/5$.

The time evolution of the contributions to the energy functional shown in Fig. 5.16 may be compared to the case of the instantaneous BIF presented in Fig. 5.4. Some parts remain similar; for example upon scission the Coulomb and collective kinetic energies display behaviour characteristic of two repulsively charged fragments accelerating away from one another (second and third panels from the bottom on the right of Figs. 5.16 and 5.4).

The evolution of the $E_0, E_1, E_2$ and $E_3$ terms are drastically different when comparing the cases of instantaneous and time-dependent BIF (top two rows in Figs. 5.4 and 5.16). For the case presented of instantaneous BIF applied to the isomer (Fig. 5.4), all four of these terms were seen to display a prompt reduction in magnitude during the first 50-100 fm/c as the configuration underwent rapid, dramatic elongation. Following this, the magnitude of the
5. BOOST-INDUCED FISSION USING TIME-DEPENDENT HARTREE-FOCK

Figure 5.16: Time evolution of the integrated contributions to the energy functional when applying a time-dependent external field to the isomeric state. The time profile of the external field is displayed in the bottom left panel. The case with scaling constant $\eta = 0.0095$ is seen to fission, which corresponds to an excitation of 52 MeV. For this case, the calculation is terminated once the fragment separation exceeds 100 fm. See text for more details.

terms recovered as the particle density ‘evened out’, and oscillated violently as shockwaves in the evolution of the densities set in.

For the time-dependent BIF case (blue line on Fig. 5.16), the evolution of the energy functional displays behaviour which is more qualitatively similar to the DIF cases (Fig. 4.19). The $E_0$, $E_1$ and $E_3$ terms for the time-dependent BIF case in Fig. 5.16 display a gradual decrease in magnitude, which upon scission recovers (at 900-1100 fm/c), and then undergoes
small oscillations about an approximately constant value. These small oscillations in the post-fissioned system correspond to the collective excitations of the fission fragments. For the case of the instantaneous boost (Fig. 5.4), the $E_2$ term shows an initial reduction in magnitude of approximately 100 MeV. In contrast to this, for the time-dependent excitation (Fig. 5.16), the evolution of the $E_2$ term shows only a dip of $\approx 5$ MeV in magnitude at around 500 fm/c as the shape configuration starts changing, before rapidly increasing by approximately 50 MeV as the surface of the nucleus increases.

The collective kinetic energy displayed in Fig. 5.16 is negligible up to $\approx 300$ fm/c, and displays an initial peak just beyond the maximum of the external field time profile (inset in second panel from the bottom, right column). It is interesting to observe that the collective kinetic energy reduces after the external field peaks (before scission occurs). At approximately 1000 fm/c for the fissioning case it rapidly increases once more as the system transitions into a fissioned configuration. This behaviour is similar to that seen for the DIF cases examined (Fig. 4.19). This suggests that the time-dependent external field has induced internal currents and gradually transitioned the nucleus into a configuration where fission becomes energetically favourable, but it is not the external field itself which forces the system to fission. In both BIF cases, where either an instantaneous or time-dependent excitation was applied, the excitation energy from the boost is dissipated mainly into the nuclear terms in the functional. The induced current, corresponding to the collective energy up until around the point of scission, is small in comparison to the total excitation energy added to the system. However, this current is much larger for instantaneous BIF: no shockwave-type behaviour is observed in the time-dependent BIF case as the energy is slowly released into the nucleus and a gradual evolution of the densities occurs.

The collective kinetic energy of the system shown in Fig. 5.16 may be interpolated to
5. BOOST-INDUCED FISSION USING TIME-DEPENDENT HARTREE-FOCK

a large time, and a value of 221(1) MeV is obtained to correspond to the energy released in the fission process (dominantly as translational kinetic energy of symmetric $^{120}_{47}$Ag fission fragments). This is in good agreement to the value obtained for the case of the instantaneous quadrupole boost (218(8) MeV, Table 5.1). This agreement is unsurprising considering the fission products are identical, and it is the Coulomb interaction which imparts most of the final collective energy to the system in the form of translational kinetic energy.

5.2.2 Fragment Excitation Modes Following Boost-Induced Fission

The excitation modes of the fission fragments following the time-dependent excitation applied to the isomer may be compared to those obtained with the 200 MeV instantaneous quadrupole excitation. Unlike those power spectra presented in Sec. 4.3.4, the measurements of the deformation parameters of the fragments have not been extended by applying Galilean transformations to remove translational motion. In future work, they would certainly benefit from this technique as it would allow for extended measurements.

![Figure 5.17:](image)

Figure 5.17: Comparison of the time evolution of the multipole deformation parameters from one of the (symmetric) fission fragments, following either an instantaneous boost or a time-dependent external field providing a quadrupole excitation.

Figure 5.17 displays the evolution of the multipole deformation parameters for one of the fragments (they are symmetric and display the same behaviour), and the corresponding power spectra are displayed in Fig. 5.18. The spectra corresponding to the quadrupole mode
in Fig. 5.18 displays a much broader peak for the 200 MeV instantaneous boost compared to the time-dependent field, although both are centred around 4 MeV. The time-dependent field also displays a noticeable secondary peak at 8 MeV. Insufficient signal was collected to obtain the power spectrum for the octupole deformation for the time-dependent excitation field. Referring to Fig. 5.17, a low frequency and large amplitude power spectrum would be expected in comparison to the octupole spectrum for the 200 MeV instantaneous boost. This is deduced by the signal oscillating far more slowly for the case of the time-dependent field (top right panel of Figure 5.17). Both cases display a fragmentation in the hexadecupole power spectra (bottom left panel of Fig. 5.18), and the time-dependent BIF case peaks at slightly higher excitation energies.

Ideally, with more signal, the power spectra would be presented with a greater resolution, and a higher quality comparison could be made. Even with the limited resolution available, despite the fission fragments being identical, it seems that their excitation modes differ depending on the method used to induce the fission process. The differences may relate to the fission induced by the instantaneous boost, which imparted a violent internal excitation as the nucleus evolved, compared to the fission process induced by the gradual excitation.
5.2.3 Variation of the Time-Dependent Excitation Profile

The effect upon the threshold energy for BIF when applying time-dependent excitation fields with different temporal profiles may be briefly analysed. Figure 5.19 presents the time evolution of the integrated energy functional for the minimum energy found to induce fission applied to the isomeric state for for three different fields.

![Figure 5.19: BIF using excitations with three different time profiles. The narrowest (green) has $\tau_0 = 150$ fm/c with width $\Delta \tau = 50$ fm/c (case A), the case in red has $\tau_0 = 500$ fm/c with width $\Delta \tau = 150$ fm/c (case B), and the widest (blue) has $\tau_0 = 800$ fm/c with width $\Delta \tau = 250$ fm/c (case C). These are the minimum energy cases which were found to induce fission.](image-url)

The shortest field, case A, (green line) has a time profile $f(t)$ (Eq. (5.5)) with $\tau_0 = 150$ fm/c and width $\Delta \tau = 50$ fm/c. The case B (red line) has $\tau_0 = 500$ fm/c and width $\Delta \tau = 150$ fm/c.
5.2 Time-Dependent External Fields

The widest case, C, (blue line) has $\tau_0 = 800$ fm/c and width $\Delta \tau = 250$ fm/c. Upon the limit $\Delta \tau \to 0$, instantaneous boosts are recovered. It may be seen that the energy functional (Fig. 5.19) for case A behaves more similarly to the instantaneous boosts used in the previous Section (see, for example, Fig. 5.4 and 5.9). The evolution of the energy functional for case A (Fig. 5.19) displays an initial sharp reduction in magnitude of the terms in the functional (other than the collective kinetic energy) at 150 fm/c, corresponding to the centroid of $f(t)$. Beyond this point, oscillations kick in, and the nucleus transitions to a fissioned configuration within 500 fm/c. The large oscillations in the terms of the energy functional suggests a shockwave-type behaviour, as seen for the cases of instantaneous BIF (Figs. 5.4 and 5.9), is occurring as the densities evolve. The initial fluctuation in the energy terms upon application of the external field A is of a much smaller magnitude than the case of the instantaneous boost, however. Taking the $E_0$ term for example, the initial spike shows a peak dropping in magnitude by $\approx 3000$ MeV from the starting point (Fig. 5.19), which compares to the $\approx 5000$ MeV seen for the instantaneous boost applied to the isomeric state (Fig. 5.4).

It is interesting to observe that the final values of the $E_0, E_1$ and $E_3$ terms are approximately equal for the two more gradual profiles (cases B and C). The case with the shortest temporal profile (A) plateaus with a magnitude approximately 1000 MeV less both for the $E_0$ and $E_3$ terms, and 15-20 MeV less for the $E_1$ term. The differences in the $E_0$ and $E_3$ terms when comparing case A to cases B and C suggests that the final fragments are less deformed, and the density is more compact, for the cases B and C.

For case A, a sharp drop in the magnitude of the $E_2$ case occurs initially, as was seen when applying instantaneous boosts (Fig. 5.4). Here, the drop in magnitude is approximately 30 MeV, which is much smaller than the drop of approximately 100 MeV seen for the application of the instantaneous boost to the isomer (Fig. 5.4). This suggests that with the short time profile a less extreme, but none the less similar, behaviour compared to the application of an instantaneous boost is occurring. This sharp drop in the $E_2$ term is not seen for the cases B and C, which suggests the process is much more similar to the gradual evolution seen for DIF (no drop in the $E_2$ term was seen in the time evolution for DIF, see Fig. 4.19).

The wider temporal profiles of cases B and C show a much more gradual transition in the energy functional as the system is evolved to fission. No large amplitude, rapid oscillations
are seen in the evolution of the terms of the energy functional (Fig. 5.19), and the behaviour
during the transition to fission is more reminiscent to that seen for the DIF cases before (Fig
4.19). This suggests that the shockwave-type behaviour in the evolution of the densities,
which was seen for excitations delivered in a shorter or instantaneous time profile, is not
occurring as the system smoothly evolves to fission. This observation points to a physically
different process occurring. The temporally extended BIF for cases B and C has similar
properties to DIF. In contrast, instantaneous BIF displays a violent oscillatory evolution of
the densities as the system fissions. This once more demonstrates that the timescale of the
energy deposition matters in terms of the resulting fission dynamics.

The initial drop in magnitude in the values of the $E_0$, $E_1$ and $E_3$ terms for case C are
similar to that of the case B (Fig. 5.19). Comparing the initial drop in magnitude of the $E_0$
term, a drop of approximately 1000 MeV is seen for both cases B and C, compared to the
drop of $\approx 3000$ MeV for case A. In fact, the main difference observed between cases B and
C is that the evolution of the $E_0$, $E_1$ and $E_3$ terms show a recovery in absolute magnitude
corresponding to scission, at a later time of $\approx 1250$ fm/c for case C, compared to around 950
fm/c for case B.

**Table 5.3:** Threshold scaling parameters and energies required to induce fission in the iso-
meric state when applying time-dependent external fields with different temporal profiles. The
final collective kinetic energy, corresponding to (mainly) translational kinetic energy has been
interpolated using the technique described in Sec. 4.2.

<table>
<thead>
<tr>
<th>Case</th>
<th>$\tau_0$ [fm/c]</th>
<th>$\Delta \tau$ [fm/c]</th>
<th>Threshold $\eta$</th>
<th>Threshold Energy [MeV]</th>
<th>Interpolated Coll. KE [MeV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>150</td>
<td>50</td>
<td>$0.0225 \leq \eta_{\text{thresh}} \leq 0.0250$</td>
<td>$99 \leq E_{\text{thresh}} \leq 110$</td>
<td>227(2)</td>
</tr>
<tr>
<td>B</td>
<td>500</td>
<td>150</td>
<td>$0.0090 \leq \eta_{\text{thresh}} \leq 0.0095$</td>
<td>$41 \leq E_{\text{thresh}} \leq 52$</td>
<td>221(1)</td>
</tr>
<tr>
<td>C</td>
<td>800</td>
<td>250</td>
<td>$0.0070 \leq \eta_{\text{thresh}} \leq 0.00725$</td>
<td>$33 \leq E_{\text{thresh}} \leq 45$</td>
<td>223(3)</td>
</tr>
</tbody>
</table>

Table 5.3 demonstrates the effect of varying the time-dependent profile $f(t)$ upon the
threshold energy required to induce fission, this can also be seen in the bottom right panel
of Fig. 5.19. In all cases, the final interpolated kinetic energies, corresponding to the energy
release, displayed in Table 5.3 agree within uncertainties. As all three cases produce identical
fission fragments, this demonstrates once more that the energy release is dominated by the
translational kinetic energy due to the Coulomb interaction between the fission products.

The application of a time-dependent excitation ensures that the static and dynamic states
at $t = 0$ are the same (unlike the application of an instantaneous boost, where a large excitation is deposited at $t = 0$). It can be seen that the shortest time profile (case A, green line in Fig. 5.19) requires significantly more energy to induce fission than the more gradual fields (cases B and C). The lowest energy observed to induce fission for case A was 110 MeV, which is approximately half of that which was required for the instantaneous boost (see Fig. 5.2). The wider fields (cases B and C) demonstrate a significant reduction in the required excitation energy compared to case A, with the lowest energies required found to be 52 and 45 MeV for cases B and C, respectively. As the temporal profile of the external field is widened, the energy required to induce fission is reduced. The comparative threshold energies for cases B and C suggest that an adiabatic limit may be approached when using an even more gradual temporal profile for the external field (up until the point of scission).

5.2.4 Applying Time-Dependent External Fields to the State with Initial Deformation $\beta_{20} = 0.89$

A time-dependent external quadrupole field may be applied to the state with initial deformation $\beta_{20} = 0.89$ to investigate the BIF process. Using the temporal profile described by Eq. (5.5), the parameters $\tau_0 = 500$ fm/c and $\Delta t = 150$ fm/c were used to specify the peak and width of the profile. Figure 5.20 displays the evolution of the multipole moments using various scaling parameters, $\eta$. For $\eta = 0.007$ the system is observed to fission, which is demonstrated by the rapid increase of elongation in the quadruple deformation following the application of the field, shown on the top left panel of Fig. 5.20. The octupole degree of freedom is also explored due to the initial mass asymmetry of the static configurations (top right panel of Fig. 5.20), and for the fissioning case increases in magnitude from $\beta_{30} = -0.3$ to the region of $-0.5$ at the point of scission.

The evolution of the particle density up to the point of scission is displayed in Fig. 5.21. The final fission products are asymmetric. Upon scission, the products are: $A_1, Z_1 = 145.05(5), 56.32(5)$ and $A_2, Z_2 = 95.02(5), 37.69(5)$. To the nearest integer particle number, this gives $^{145}_{56}$Ba and $^{95}_{35}$Sr. The mass distributions of this BIF process will be compared to DIF, as well as experimental results, in Section 5.2.5.

Figure 5.21 displays no visible changes in the particle density slices until 300-400 fm/c, unlike the case where an instantaneous boost was applied (Fig. 5.8). The evolution of the particle density to fission is seen to take longer for this excitation (2050 fm/c) than the
application of the threshold 225 MeV instantaneous boost to the same state, which took approximately 1700 fm/c (Fig. 5.8). The influence of the external field is negligible by 800 fm/c, and beyond this the neck region smoothly rearranges into the two fission fragments. Referring to Fig. 5.22, which displays the decomposed integrated energy functional for the system, the threshold energy required to induce BIF using the specified time-dependent excitation field is $32 \leq E_{\text{thresh}} \leq 40$ MeV. This energy compares to the 225 MeV required for the minimum energy case of instantaneous BIF for this state (Fig. 5.7). Once again it demonstrates that when applying a gradual evolution, a significantly lower threshold energy is required to induce fission, compared to the application of an instantaneous high-energy boost.

The evolution of the $E_0$, $E_1$ and $E_3$ terms of the energy functional for the fissioning case (blue line in Fig. 5.22) display that the nucleus moves to an elongated configuration in the first 500 fm/c (displayed by the drop in magnitude). During the time between 500 and 2000 fm/c, slight fluctuations are observed in the terms as the configuration rearranges due to the current induced by the excitation. The fluctuations are small; for example the $E_0$ term varies by less than 250 MeV whilst in this elongated configuration. Around 2000 fm/c, at the point of scission, the characteristic increase in magnitude of the $E_0$, $E_1$ and $E_3$ terms are
Figure 5.21: 2D slices of the 3D particle density starting from the state with deformation $\beta_{20}=0.89$, upon application of a time-dependent external field. The scaling constant $\eta$ is 0.007, and the field is centred around $\tau_0 = 500$ fm/c with width $\Delta t = 150$ fm/c. The isolines are separated by 0.05 particles/fm$^3$.

observed. The $E_2$ term (left column, second panel from top of Fig. 5.22) displays a gradual increase beyond an initial peak at 800 fm/c, increasing by approximately 20 MeV between $t = 800$ and $t = 2000$ fm/c as the two-fragment configuration gradually forms.

As with the other cases where a time-dependent external field has been considered (Fig. 5.19), the collective kinetic energy in Fig. 5.22 shows an initial peak near the centroid of the temporal profile of the field, which then reduces as the excitation field ends (inset in collective kinetic energy panel of Fig. 5.22). The collective kinetic energy for the fissioning case remains constant at around 1 MeV once the external field ends, which corresponds the current induced by the field. This collective energy is significantly less than that seen for instantaneous BIF for the 225 MeV boost (see Fig. 5.9) and is more comparable to that seen for DIF before scission (see Fig. 4.19). Beyond 2000 fm/c, a rapid increase in collective kinetic energy is
Figure 5.22: Time evolution of the integrated contributions to the energy functional following an application of a time-dependent external field. The time profile of the external field is shown in the bottom right panel. The scaling constant $\eta = 0.0070$ is seen to induce fission, corresponding to an excitation of 40 MeV. See text for more details.

seen around point of scission as translational motion sets in (Fig. 5.22).

As the system took longer to fission than the other BIF cases, the calculation was terminated at 3000 fm/c, where by this point the fragment separation was only 75 fm. By interpolating the collective kinetic energy from the points where the fragment separation was 40 fm and 50 fm, the average value obtained to correspond to translational energy at large times is 207(9) MeV. This value is reasonable when compared to experimental data, falling within the distribution of kinetic energies obtained from thermal neutron-induced fission.
displayed in Fig. 4.26.

Once again, it has been demonstrated that with the use of an external field with a gradual time-dependence, fission may be induced requiring energy an order of magnitude lower than an instantaneous boost. The evolution of the energy functional (Fig. 5.22) when applying the time-dependent excitation field shows a smoother, less dramatic transition as the one-fragment system evolves into a two-fragment system, compared to the application of an instantaneous or short excitation field (Figs 5.9 and 5.19).

### 5.2.5 Comparing the Masses Obtained using BIF to DIF

The mass distributions obtained from the DIF processes examined in the previous Chapter (Table 4.3) may be compared to those obtained from the BIF processes investigated. For BIF, only those cases where the static configuration had mass asymmetry will be considered. Due to the symmetric nature of the excitation fields applied, an initial configuration with no octupole deformation will be unable to explore this degree of freedom, therefore symmetric fission fragments will always be obtained.

Figure 5.23 displays a comparison between the masses obtained from the DIF cases presented in the previous Chapter (red bars in Figure), and the BIF cases examined in this Chapter. For the BIF cases, the asymmetric fission products obtained from the initial state with $\beta_{20} = 0.89$ have been included. The blue bars correspond to the masses obtained using instantaneous excitations (Table 5.2), and the green bar to the sample temporally extended time-dependent excitation field (Sec. 5.2.4).

As was presented in the previous Chapter, the DIF fission products fall well within the experimentally obtained mass distributions. This suggests that investigating DIF within TDHF corresponds to a process which is similar to the typical experimental methods used to induce fission (see, e.g. Ref. [115]). The instantaneous BIF cases investigated have displayed a different process. When applying instantaneous boosts to the static state with deformation $\beta_{20} = 0.89$, the resulting masses fall on the edge of the experimentally obtained results, displaying a mass distribution which is more asymmetric (blue bars in Fig. 5.23). This suggests, that although fission may be induced with such a technique, that the mechanism does not correspond to typical experimentally observed fission processes. It also bears mentioning that the threshold energy of 225 MeV required for BIF starting from the state with $\beta_{20} = 0.89$
is at least an order of magnitude larger than that required to induce fission in experimental studies. For example, photo-fission may be induced in $^{240}$Pu using a 12 MeV endpoint energy bremsstrahlung source [115] (although such a process corresponds to a dipole excitation, rather than the quadrupole excitation examined).

When performing BIF and applying the excitation field with a temporally extended time-dependent profile, the energy required to induce fission is significantly reduced. For the application of a field with a temporal profile characterised by a Gaussian peaking at $\tau_0 = 500$ fm/c with width $\Delta \tau = 150$ fm/c, the minimum energy found to induce fission was 40 MeV. The sample mass distribution (green bar in Figure 5.23) lies closer to the peak of the experimental distribution than the other BIF examples. This suggests that the process is more closely linked to the dynamic evolution observed for DIF. This assumption is strengthened by comparing the behaviours during time evolution; the time-dependent BIF case (Sec. 5.2.4)
displayed a small collective energy up until the point of scission and no shockwave-type behaviour as the densities evolved, which is in line to what is observed for the DIF processes (see Sec. 4.2). Comparing the results of DIF to instantaneous and temporally extended BIF, it could be concluded that the experimentally observed fission fragments are formed during this smooth, gradual evolution to fission, rather than in the violent, dramatic process seen to occur for instantaneous BIF. The comparison of instantaneous BIF to temporally extended BIF demonstrates that the timescale for the energy deposition has important consequences regarding the fission dynamics.

Overall, different approaches for inducing fission processes within TDHF have been presented and compared. It would be of interest to further investigate the connection between DIF and temporally extended BIF, as this smooth, gradual evolution to scission appears to have important consequences regarding the formation of the fission products.
5. BOOST-INDUCED FISSION USING TIME-DEPENDENT HARTREE-FOCK
Conclusions and Outlooks

In this thesis, the time-dependent Hartree-Fock (TDHF) method has been applied to investigate the dynamics of nuclear fission in $^{240}$Pu. The 3D Skyrme-Hartree-Fock code Sky3D was modified to include a constraint on the quadrupole deformation using the Augmented Lagrangian Method. The static potential energy surface (PES) was calculated with respect to this constraint. As well as the one-fragment static fission pathway, a competing pathway of two-fragment configurations was explored. The point at which these pathways met was interpreted to have significance in the dynamics upon the time evolution of the static states.

Starting from the calculated one-fragment PES, the dynamics of fission were investigated. Deformation-induced fission (DIF) was explored first, by releasing the quadrupole constraint and time-evolving a selection of states situated around and beyond the second static fission barrier. Three behaviours were observed. For those states with a quadrupole deformation below the peak of the fission barrier, vibrations corresponding to a collective giant resonance mode were seen. 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 was observed for the evolution of states which were situated beyond the peak of the second static fission barrier, but before the critical point where the static one and two-fragment pathways intercepted. Upon time evolution up to 9000 fm/c, these states also failed to fission, but the dynamics were not typical of collective giant resonant modes. The repulsive Coulomb force attempted to drive the configuration towards fission, but due to competition with the attractive terms in the energy functional, scission did 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.
6. CONCLUSIONS AND OUTLOOKS

For states with a static deformation exceeding the intersection of the one and two-fragment fission pathways, DIF was observed upon time evolution. It was interpreted that for these states, as a static two-fragment configuration exists with greater binding energy than the one-fragment configuration, that it was energetically possible for the one-fragment configurations to evolve to fission with only a modest rearrangement of the densities. The evolution from the static states to a fissioned configuration displayed a gradual rearrangement of the densities up until around the point of scission. At this point, the Coulomb repulsion between the fragments overpowered the nuclear potential, and translational motion set in as the fission products rapidly accelerated away from one another. The timescale for DIF varied depending upon the deformation of the initial state. The least deformed configuration demonstrated a phase where the densities rearranged lasting approximately 1500 fm/c before scission occurred, whereas the most deformed configuration was initially close to the point of scission and the neck ruptured within 100-200 fm/c.

A selection of fission products was observed for the various initial configurations considered. When compared to experimental measurements of neutron-induced fission processes, the agreement of the calculated fragment masses demonstrated promising results. The energy released was 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.

For those states where DIF failed, boost-induced fission (BIF) was investigated. For this process, a quadrupole excitation field was applied either instantaneously, or gradually with a Gaussian time profile. Two states were investigated: the fission isomer ($\beta_{20} = 0.68$) and a state just beyond the peak of the second barrier, with static deformation $\beta_{20} = 0.89$.

Minimum excitation energies of the order of 200 MeV were required when applying instantaneous boosts to the considered states to induced fission, which is an energy significantly greater than the calculated static fission barrier heights ($\approx 8$ MeV). In these processes the excitation energy, which was deposited as collective energy at $t = 0$, was absorbed within the first 100-200 fm/c into the nuclear potential terms of the energy density functional. A significant current was also induced, and a violent evolution of the state ensued, which was in contrast to the gradual rearrangement of the densities observed for DIF.
The fission products obtained when applying various instantaneous boosts to induce fission in the state with $\beta_{20} = 0.89$ fell on the edges of the considered experimental mass distributions, showing tendency to be more asymmetric. These results suggest that the instantaneous BIF process does not compare physically to the process of neutron-induced fission. The pre-scission states demonstrated rearrangements of both energy and density which was far more extreme than the DIF process. Different timescales for fission were observed when various energy instantaneous boosts were applied, ranging from hundreds to thousands of fm/c. Observation of ternary fission products using instantaneous excitations far beyond the minimum threshold to induce fission may warrant further investigation to examine the dynamics of these instantaneous BIF processes within TDHF.

When applying a quadrupole excitation field with a Gaussian time profile, the energy required to induce fission was found to reduce significantly. For example, when exciting the fission isomer, the minimum energy required to observe BIF with an instantaneous excitation was 200 MeV, in comparison to 45 MeV with an temporally extended excitation field centred around 800 fm/c with width 250 fm/c. Similarities in the evolution between temporally extended BIF and DIF were apparent, displaying a smooth, gradual evolution of the densities up to around the point of scission. When evolving the state using temporally extended BIF, the timescale for scission was approximately 2000 fm/c, during which the rearrangement of the energies and densities were far less extreme compared to the instantaneous BIF processes investigated. This timescale compares to the DIF case with the least deformed initial configuration.

In both types of BIF process (instantaneous and temporally extended), the evolution is such that the nucleus absorbs all the energy of the boost and rearranges substantially before fission. During this rearrangement process, currents build up and the nucleus splits into fragments. The length of this rearrangement phase is more or less independent of the type of boost, which suggests the density rearrangement is a robust mechanism for fission that does not carry memory of the initial excitation process.

For the state with $\beta_{20} = 0.89$, when applying a time-dependent external field centred around 500 fm/c with a width of 150 fm/c, BIF was observed requiring an excitation energy in the region of 40 MeV (compared to 225 MeV for the instantaneous excitation applied to the same state). The resulting fission products agreed better with the experimental results.
for neutron-induced fission. This improved agreement of the mass distributions between experiment and theory for non-violent fission processes (DIF and temporally extended BIF) suggests that the fragment formation is mostly determined an evolution of this type. Investigations of BIF could be extended to examine widening the time profile of the excitation even further, as the comparison between instantaneous BIF and temporally extended BIF demonstrated that the timescale for the energy deposition has important consequences regarding the fission dynamics.

In conclusion, with the advances of computational power and the development of 3D symmetry-unrestricted codes, time-dependent Hartree Fock presents itself as a promising tool for describing nuclear fission processes. In particular, the DIF and temporally extended BIF processes investigated agreed well with the experimental results of neutron-induced fission. TDHF allows the flexibility to apply external excitation fields with different spatial and temporal profiles, so that different types fission processes may be examined. The results presented in this thesis serve to justify future investigation of fission phenomenon using the time-dependent Hartree-Fock method.

Future work could investigate the inclusion of a number of effects. It would be of interest to determine whether time-dependent pairing would have any significant impact upon the results observed in this thesis. Such an extension to Sky3D is a feasible modification to bring its abilities in line with other modern studies using TDHF [104, 106, 107, 109]. The inclusion of a three-body analysis within Sky3D would be essential to further investigate the BIF process which was seen to lead to ternary fission fragments. Additionally, increasing the number of constraints when calculating static states [47] would allow a wider selection of initial configurations to investigate the dynamics of fission. Alternatively, existing Hartree-Fock solvers which are capable of enforcing multiple shape constraints could be modified to produce static configurations which may be time-evolved using Sky3D.

From another perspective, rather than collecting a larger sample of results using the techniques investigated in this thesis, the distribution of fission products could be computed explicitly using different methods. It would be of interest to project the resulting fission products onto a good particle number [131]. Further, a fluctuation of the particle number could be deduced by applying the Balian-Vénéroni variational principle [101, 144, 145, 146].
This would allow an enhanced mass distribution probability to be deduced, even with a
limited number of TDHF fission events.
6. CONCLUSIONS AND OUTLOOKS
Appendix A

Gauge Transformation of the Skyrme Energy Density Functional

A.1 Gauge Transformation properties of Local Densities

An instantaneous kinetic energy boost can be applied to the single particle wave functions by applying the gauge transformation [143],

\[ \tilde{\varphi}_i(r) = e^{i\phi(r)} \varphi_\alpha(r), \quad (A.1) \]

where \( \phi(r) \) is an arbitrary spatial profile. The Skyrme energy density functional must be gauge invariant (which implies that it is also invariant under Galilean transformations). To prove this, one must consider the effect the gauge transformation has upon the local densities which define the energy density functional of the system.

A.1.1 Particle Density

The (non-local) density matrix is defined in the Hartree-Fock picture:

\[ \rho(r, r') = \sum_\alpha \varphi^*_\alpha(r') \varphi_\alpha(r). \quad (A.2) \]

Taking the limit \( r' \to r \) gives:

\[ \rho(r) = \sum_\alpha |\varphi_\alpha(r)|^2. \quad (A.3) \]

The ‘boosted’ particle density following the gauge transformation may be defined by:

\[ \tilde{\rho}(r, r') = e^{i(\phi(r) - \phi(r'))} \sum_\alpha \varphi^*_\alpha(r') \varphi_\alpha(r) \]
\[ = e^{i(\phi(r) - \phi(r'))} \rho(r, r'). \quad (A.4) \]
A. GAUGE TRANSFORMATION OF THE SKYRME ENERGY DENSITY FUNCTIONAL

Taking the limit \( r' \rightarrow r \) gives:

\[
\bar{\rho}(r) = \rho(r). \tag{A.5}
\]

This demonstrates that the particle density is invariant under gauge transformations. Due to this, all terms in the energy density functional (Eq. (2.24)) that are solely dependent upon \( \rho(r) \) (the \( E_0, E_2, E_3 \) and \( E_{\text{Coul}} \) terms) are invariant under gauge transformations.

A.1.2 Spin Density

The spatial components \( \mu = x, y, z \) of the spin density \( S_\mu \) are defined by:

\[
S_\mu(r, r') = \sum_{\alpha, \sigma, \sigma'} \varphi^*_{\alpha \sigma'}(r') \hat{\sigma}^\mu_{\sigma \sigma'} \varphi_{\alpha \sigma}(r), \tag{A.6}
\]

where \( \hat{\sigma}^\mu_{\sigma \sigma'} \) are the components of the Pauli spin operator. At \( r' \rightarrow r \), one finds:

\[
S_\mu(r, r) = \sum_{\alpha, \sigma, \sigma'} \varphi^*_{\alpha \sigma'}(r) \hat{\sigma}^\mu_{\sigma \sigma'} \varphi_{\alpha \sigma}(r). \tag{A.7}
\]

The boosted spin density may be written:

\[
\bar{S}_\mu(r, r') = \sum_{\alpha, \sigma, \sigma'} e^{-i \phi(r')} \varphi^*_{\alpha \sigma'}(r') \hat{\sigma}^\mu_{\sigma \sigma'} e^{i \phi(r)} \varphi_{\alpha \sigma}(r)
= e^{i(\phi(r) - \phi(r'))} S_\mu(r, r'). \tag{A.8}
\]

The limit therefore yields

\[
\bar{S}_\mu(r) = S_\mu(r). \tag{A.9}
\]

This demonstrates the spin density is invariant under the gauge transformation \( e^{i \phi(r)} \).

A.1.3 Current Density

The components of the current density \( j_\mu \) are defined by:

\[
j_\mu(r, r') = -\frac{i}{2} (\nabla_\mu - \nabla'_\mu) \rho(r, r')
= -\frac{i}{2} \sum_\alpha (\nabla_\mu - \nabla'_\mu) \varphi^*_{\alpha}(r') \varphi_{\alpha}(r)
= -\frac{i}{2} \sum_\alpha \varphi^*_{\alpha}(r') \nabla_\mu \varphi_{\alpha}(r) - \nabla'_\mu \varphi^*_{\alpha}(r') \varphi_{\alpha}(r). \tag{A.10}
\]

At the limit \( r' \rightarrow r \)

\[
j_\mu(r) = -\frac{i}{2} \sum_\alpha \varphi^*_{\alpha}(r) \nabla_\mu \varphi_{\alpha}(r) - \nabla'_\mu \varphi^*_{\alpha}(r) \varphi_{\alpha}(r). \tag{A.11}
\]
A.1 Gauge Transformation properties of Local Densities

The boosted current density is defined as:

$$\tilde{\mathbf{j}}_{\mu}(r, r') = -\frac{i}{2} \left( \nabla_{\mu} - \nabla'_{\mu} \right) e^{i(\phi(r) - \phi(r'))} \rho(r, r')$$

$$= -\frac{i}{2} \sum_{\alpha} \left( (\nabla_{\mu} \phi(r)) \varphi_{\alpha}^*(r') \varphi_{\alpha}(r) + e^{i(\phi(r) - \phi(r'))} \varphi_{\alpha}^*(r') (\nabla_{\mu} \varphi_{\alpha}(r)) + \right.$$

$$+ i (\nabla'_{\mu} \phi(r')) e^{i(\phi(r) - \phi(r'))} \varphi_{\alpha}^*(r') \varphi_{\alpha}(r)$$

$$- e^{i(\phi(r) - \phi(r'))} (\nabla'_{\mu} \varphi_{\alpha}^*(r')) \varphi_{\alpha}(r) \right). \quad (A.12)$$

Taking the limit \( r' \to r \) gives:

$$\tilde{j}_{\mu}(r) = -\frac{i}{2} \sum_{\alpha} \left( 2i (\nabla_{\mu} \phi(r)) |\varphi_{\alpha}(r)|^2 + \varphi_{\alpha}^*(r) (\nabla_{\mu} \varphi_{\alpha}(r)) - (\nabla'_{\mu} \varphi_{\alpha}^*(r)) \varphi_{\alpha}(r) \right)$$

$$= \sum_{\alpha} \left( (\nabla_{\mu} \phi(r)) |\varphi_{\alpha}(r)|^2 - \frac{i}{2} \left( \varphi_{\alpha}^*(r) (\nabla_{\mu} \varphi_{\alpha}(r)) - (\nabla'_{\mu} \varphi_{\alpha}^*(r)) \varphi_{\alpha}(r) \right) \right)$$

$$= j_{\mu}(r) + \nabla_{\mu} \phi(r) \rho(r). \quad (A.13)$$

Consequently, the current density transforms under a gauge transformation as \( \tilde{j}_{\mu}(r) \to j_{\mu}(r) + \nabla_{\mu} \phi(r) \rho(r) \). The additional term is proportional to the gradient of the gauge angle.

A.1.4 Kinetic Density

The kinetic density \( \tau \) is defined by:

$$\tau(r, r') = \nabla \cdot \nabla' \rho(r, r')$$

$$= \sum_{\alpha} \nabla' \varphi_{\alpha}^*(r') \cdot \nabla \varphi_{\alpha}(r). \quad (A.14)$$

At the limit \( r' \to r \):

$$\tau(r) = \sum_{\alpha} \nabla \varphi_{\alpha}^*(r) \cdot \nabla \varphi_{\alpha}(r). \quad (A.15)$$

The boosted kinetic density is given by:

$$\tilde{\tau}(r, r') = \sum_{\alpha} \nabla \cdot \nabla' e^{i(\phi(r) - \phi(r'))} \varphi_{\alpha}^*(r') \varphi_{\alpha}(r)$$

$$= \sum_{\alpha} \left( -i (\nabla' \phi(r')) e^{i(\phi(r) - \phi(r'))} \varphi_{\alpha}^*(r') + e^{i(\phi(r) - \phi(r'))} (\nabla' \varphi_{\alpha}^*(r')) \right)$$

$$\cdot \left( i (\nabla \phi(r)) e^{i(\phi(r) - \phi(r'))} \varphi_{\alpha}(r) + e^{i(\phi(r) - \phi(r'))} (\nabla \varphi_{\alpha}(r)) \right). \quad (A.16)$$
Taking the limit \( r' \to r \):

\[
\bar{\tau}(r) = \sum_{\alpha} |\nabla \varphi(r)|^2 |\varphi_{\alpha}(r)|^2 \\
+ (\nabla \varphi^*_{\alpha}(r)) \cdot (\nabla \varphi_{\alpha}(r)) \\
+ i (\nabla \varphi(r)) \left( (\nabla \varphi^*_{\alpha}(r)) \varphi_{\alpha}(r) - \varphi^*_{\alpha}(r)(\nabla \varphi_{\alpha}(r)) \right) \\
= \tau(r) + |\nabla \varphi(r)|^2 \rho(r) + 2\nabla \varphi(r) \cdot \mathbf{j}(r). \tag{A.17}
\]

The current \( \mathbf{j}(r) \) is zero for a static state, so the kinetic density transforms under a gauge transformation as \( \tau(r) \to \tau(r) + |\nabla \varphi(r)|^2 \rho(r) \). The additional term, as seen for the current density, is proportional to the gradient of the gauge angle.

### A.1.5 Tensor Spin-Current Density

The components of the tensor spin-current density \( J_{\mu \nu} \) are defined by:

\[
J_{\mu \nu}(r, r') = -\frac{i}{2} (\nabla_{\mu} - \nabla_{\nu}') S_{\nu}(r, r') \\
= -\frac{i}{2} \sum_{\alpha, \sigma, \sigma'} \varphi_{\alpha \sigma'}(r') \delta_{\sigma, \sigma'} \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r) \right) - \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r') \right) \delta_{\sigma, \sigma'} \varphi_{\alpha \sigma}(r). \tag{A.18}
\]

Taking the limit \( r' \to r \) gives:

\[
J_{\mu \nu}(r) = -\frac{i}{2} \sum_{\alpha, \sigma, \sigma'} \varphi_{\alpha \sigma'}^{*}(r) \delta_{\sigma, \sigma'} \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r) \right) - \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r) \right) \delta_{\sigma, \sigma'} \varphi_{\alpha \sigma}(r) \tag{A.19}
\]

The boosted tensor spin-current density is given by:

\[
\tilde{J}_{\mu \nu}(r, r') = -\frac{i}{2} \sum_{\alpha, \sigma, \sigma'} e^{-i \phi(r')} \varphi_{\alpha \sigma'}^{*}(r') \delta_{\sigma, \sigma'} \nabla_{\mu} \left( e^{i \phi(r')} \varphi_{\alpha \sigma}(r) \right) \\
- \nabla_{\mu}' \left( e^{-i \phi(r')} \varphi_{\alpha \sigma}(r') \right) \delta_{\sigma, \sigma'} e^{i \phi(r')} \varphi_{\alpha \sigma}(r) \\
= -\frac{i}{2} \sum_{\alpha, \sigma, \sigma'} e^{-i \phi(r')} \varphi_{\alpha \sigma'}^{*}(r') \delta_{\sigma, \sigma'} \left( i \left( \nabla_{\mu} \varphi(r) \right) e^{i \phi(r)} \varphi_{\alpha \sigma}(r) + e^{i \phi(r)} \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r) \right) \right) \\
- \left( -i \left( \nabla_{\mu} \varphi(r) \right) e^{-i \phi(r')} \varphi_{\alpha \sigma}(r') \right) + e^{-i \phi(r')} \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r') \right) \delta_{\sigma, \sigma'} e^{i \phi(r')} \varphi_{\alpha \sigma}(r) \tag{A.20}
\]

Taking the limit \( r' \to r \):

\[
\tilde{J}_{\mu \nu}(r) = -\frac{i}{2} \sum_{\alpha, \sigma, \sigma'} \varphi_{\alpha \sigma'}^{*}(r) \delta_{\sigma, \sigma'} \left( i \left( \nabla_{\mu} \varphi(r) \right) \varphi_{\alpha \sigma}(r) \right) \\
+ \varphi_{\alpha \sigma'}^{*}(r) \delta_{\sigma, \sigma'} \left( \nabla_{\mu} \varphi_{\alpha \sigma}(r) \right) \\
- \left( -i \nabla_{\mu} \varphi(r) \right) \varphi_{\alpha \sigma'}(r) \delta_{\sigma, \sigma'} \varphi_{\alpha \sigma}(r) \right) \\
- \left( \nabla_{\mu} \varphi_{\alpha \sigma'}(r) \right) \delta_{\sigma, \sigma'} \varphi_{\alpha \sigma}(r) \\
= J_{\mu \nu}(r) + S_{\nu}(r) \nabla_{\mu} \varphi(r). \tag{A.21}
\]
The tensor spin-current density transforms under a gauge transformation as \( J_{\mu\nu}(r) \to J_{\mu\nu}(r) + S_\nu(r)\nabla_\mu \phi(r) \). The additional term, once again, is proportional to the gradient of the gauge angle.

### A.1.6 Spin-Orbit Density

The Cartesian components of the spin-orbit density \( J_\kappa(r) \) are defined from the tensor spin-current density as,

\[
J_\kappa(r) = \sum_{\mu\nu} \epsilon_{\kappa\mu\nu} J_{\mu\nu}(r),
\]

(A.22)

where \( \epsilon_{\kappa\mu\nu} \) is the Levi-Civita symbol, and \( \kappa, \mu, \nu \) tun over \( x, y, z \). The spin-orbit density may be written explicitly as

\[
J(r) = \begin{pmatrix}
J_{yz} - J_{zy} \\
J_{zx} - J_{xz} \\
J_{xy} - J_{yx}
\end{pmatrix}.
\]

(A.23)

The boosted spin-orbit density is defined analogously to the tensor spin-current density:

\[
\bar{J}_\kappa(r) = \sum_{\mu\nu} \epsilon_{\kappa\mu\nu} \left( J_{\mu\nu}(r) + S_\nu(r)\nabla_\mu \phi(r) \right),
\]

(A.24)

or explicitly as

\[
\bar{J}(r) = \begin{pmatrix}
( J_{yz}(r) + S_z(r)\nabla_y \phi(r) ) - ( J_{zy}(r) + S_y(r)\nabla_z \phi(r) ) \\
( J_{zx}(r) + S_x(r)\nabla_z \phi(r) ) - ( J_{xz}(r) + S_z(r)\nabla_x \phi(r) ) \\
( J_{xy}(r) + S_y(r)\nabla_x \phi(r) ) - ( J_{yx}(r) + S_x(r)\nabla_y \phi(r) )
\end{pmatrix},
\]

(A.25)

Or alternatively, in the more convenient form of

\[
\bar{J}(r) = J(r) - S(r) \times \nabla \phi(r).
\]

(A.26)

The spin-orbit density transforms under a gauge transformation as \( J(r) \to J(r) - S(r) \times \nabla \phi(r) \). For all the densities which are not invariant under the gauge transformation, the extra term is always proportional to the gradient of the gauge angle \( \phi(r) \). It will be shown shortly that due to the combinations of densities that appear by construction in the Skyrme energy density functional, the energy density functional as a whole is invariant under a gauge transformation. As a consequence of gauge invariance, the functional is also Galilean invariant.
A.2 Gauge Invariance of the Skyrme Energy Density Functional

The $E_0$, $E_2$ and $E_3$ terms of the Skyrme energy density functional only depend explicitly on the local particle density (see Sec. 2.3.2). As this density, $\rho(r)$, has been shown to be gauge invariant, the three terms in the energy density functional are also invariant. The remaining terms are the $E_1$ term and $E_{ls}$ terms, and it will be shown in the following that they are invariant under gauge transformations of the local densities.

A.2.1 $E_1$ Term

The $E_1$ term contains the combination of densities $\mathcal{E}_1$

$$\mathcal{E}_1 = \rho(r)\tau(r) - j(r)^2. \quad (A.27)$$

By replacing the densities by their gauge transformed versions, the gauge transformed density combination becomes

$$\bar{\mathcal{E}}_1 = \left(\rho(r)\right)\left(\tau(r) + |\nabla \phi(r)|^2 \rho(r) + 2\nabla \phi(r) \cdot j(r)\right) - \left(j(r) + \nabla \phi(r) \rho(r)\right)^2. \quad (A.28)$$

Expanding this expression gives

$$\bar{\mathcal{E}}_1 = |\nabla \phi(r)|^2 \rho(r)^2 + \tau(r)\rho(r) + 2\rho(r)\nabla \phi(r) \cdot j(r)$$

$$- |\nabla \phi(r)|^2 \rho(r)^2 - j(r)^2 - 2\rho(r)\nabla \phi(r) \cdot j(r)$$

$$= \mathcal{E}_1, \quad (A.29)$$

and hence the $E_1$ term is gauge invariant.

A.2.2 $E_{ls}$ Term

The remaining term in the Skyrme EDF is the spin orbit term $\mathcal{E}_{ls}$, which contains the combinations of densities $\mathcal{E}_{ls}$:

$$\mathcal{E}_{ls} = \rho(r)\nabla \cdot J(r) + S(r) \cdot (\nabla \times j(r)). \quad (A.30)$$

Appendix A of Ref. [76] demonstrates that this term may be rewritten:

$$\mathcal{E}_{ls} = \rho(r)\nabla \cdot J(r) + j(r) \cdot (\nabla \times S(r)). \quad (A.31)$$
A.3 Energy Added by Instantaneous Boosts

By replacing the densities by their gauge transformed versions, the combination of densities

\[ \bar{E}_{ls} = \rho(r) \nabla \cdot \left( J(r) - (S(r) \times \nabla \phi(r)) \right) + \left( j(r) + \rho(r) \nabla \phi(r) \right) \cdot (\nabla \times S(r)) \]  

(A.32)

is obtained. The expression may be expanded and written in the form

\[ \bar{E}_{ls} = \rho(r) \nabla \cdot J(r) + j(r) \cdot (\nabla \times S(r)) + \rho(r) \nabla \phi(r) \cdot (\nabla \times S(r)) - \rho(r) \nabla \cdot (S(r) \times \nabla \phi(r)) \]  

(A.33)

Making use of the identity

\[ \nabla \cdot (A \times B) = B \cdot (\nabla \times A) - A \cdot (\nabla \times B) \]  

(A.34)

the transformed density combination may be written

\[ \bar{E}_{ls} = \rho(r) \nabla \cdot J(r) + j(r) \cdot (\nabla \times S(r)) + \rho(r) \nabla \phi(r) \cdot (\nabla \times S(r)) + \rho(r) S(r) \cdot (\nabla \times \nabla \phi(r)) - \rho(r) \nabla \phi(r) \cdot (\nabla \times S(r)) = E_{ls} , \]  

(A.35)

making use of the relation

\[ \nabla \times \nabla \phi(r) = 0 \]  

(A.36)

This shows the combination of densities in the \( E_{ls} \) term is gauge invariant, and therefore it has been demonstrated the entire Skyrme energy density function is also invariant.

This is an important result when considering a dynamic system adopting the Skyrme energy density functional. First and foremost, it demonstrates that even for even-even nuclei, some time-odd densities are required to conserve Galilean invariance in the dynamic case. Additionally, it proves that velocity-dependent interactions induced by a gauge transformation will only affect the kinetic energy (see Eq. (2.24)). In contrast, the nuclear EDF and the Coulomb parts are invariant under gauge transformations.

A.3 Energy Added by Instantaneous Boosts

The adopted energy density function of the nuclear system is given by

\[ E = E_{\text{kin}} + E_{\text{skyrme}} + E_{\text{coul}} . \]  

(A.37)
As the contributions from the Skyrme and the Coulomb terms are gauge invariant, the only energy contribution to the system from an instantaneous velocity boost applied by the transformation \( \varphi(r) \rightarrow e^{i\varphi(r)} \varphi(r) \) is in the form of kinetic energy. The kinetic energy is given by:

\[
E_{\text{kin}} = \frac{\hbar^2}{2m} \int d^3r \sum_i \nabla \varphi_i^*(r) \cdot \nabla \varphi_i(r). \tag{A.38}
\]

Substituting in the gauge transformed expression for the kinetic density (Eq. (A.17), noting that \( j(r) \) for an initial static state is zero) gives:

\[
\tilde{E}_{\text{kin}} = \frac{\hbar^2}{2m} \int d^3r \sum_\alpha \nabla \varphi_\alpha^*(r) \cdot \nabla \varphi_\alpha(r) \]
\[
+ \frac{\hbar^2}{2m} \int d^3r |\nabla \varphi(r)|^2 \rho(r)
= E_{\text{kin}} + \Delta E_{\text{kin}}. \tag{A.39}
\]

The excitation energy, \( \Delta E_{\text{kin}} \), is equivalent to a collective kinetic energy as \( \nabla \varphi(r) \) may be interpreted as a velocity field [143]. The amount of energy imparted by the boost can be computed precisely. Defining \( \eta \) as a scaling factor for the boost field (\( \varphi(r) \rightarrow \eta \varphi(r) \)), the energy becomes

\[
\Delta E_{\text{kin}} = \frac{\hbar^2}{2m} A \eta^2 \langle |\nabla \varphi(r)|^2 \rangle, \tag{A.40}
\]

where \( A \) is the integrated particle density. As \( A \) is constant, the energy is proportional to the square of the boost strength \( \eta \). By rearranging for \( \eta \),

\[
\eta = \sqrt{\frac{\Delta E_{\text{kin}}}{\frac{\hbar^2}{2m} A \langle |\nabla \varphi(r)|^2 \rangle}}, \tag{A.41}
\]

the magnitude of the boost may be fixed for a given \( \Delta E_{\text{kin}} \).

### A.3.1 Quadrupole Boost

As an example, to excite the nucleus by adding collective kinetic energy via a quadrupole velocity field, one may consider

\[
\phi(r) = \eta(2z^2 - x^2 - y^2). \tag{A.42}
\]

Therefore,

\[
|\nabla \phi(r)|^2 = \eta^2(4x^2 + 4y^2 + 16z^2). \tag{A.43}
\]
Rearranging for $\eta$, the relationship is given:

$$\eta = \frac{1}{2} \sqrt{\frac{\Delta E_{\text{kin}}}{\frac{p^2}{2m} A(4x^2 + 4y^2 + 16z^2)}}.$$  \hspace{1cm} (A.44)

which may be used to add a precise amount of excitation energy to an initially static configuration at $t = 0$ in the form of a collective quadrupole velocity boost.
A. GAUGE TRANSFORMATION OF THE SKYRME ENERGY DENSITY FUNCTIONAL
Appendix B

Spherical Harmonics and Deformation Parameters

B.1 Spherical Harmonics

The spherical harmonics, $Y_{lm}(\theta, \varphi)$, are defined by

$$Y_{lm}(\theta, \varphi) = (-1)^m \sqrt{\frac{2l + 1}{4\pi} \frac{(l - m)!}{(l + m)!}} P_{lm}[\cos(\theta)] e^{im\varphi}. \quad (B.1)$$

Here, $P_{lm}[\cos(\theta)]$ are Lagrange polynomials. When expressing the spherical harmonics, $Y_{lm}$, in Cartesian co-ordinates, it is convenient to take into account the following conversions from spherical to the Cartesian co-ordinate system:

$$x = r \sin \theta \cos \varphi$$
$$y = r \sin \theta \sin \varphi$$
$$z = r \cos \theta. \quad (B.2)$$

Here, $\theta$ is the polar angle, and $\varphi$ the azimuthal angle. The distance $r$ may be defined as

$$r^2 = x^2 + y^2 + z^2. \quad (B.3)$$

The spherical harmonics $Y_{l0}$, $l = 1, ..., 4$, will be given explicitly in the following ($Y_{00}$ is constant ($= \frac{1}{2} \sqrt{\frac{1}{\pi}}$)). These are the spherical harmonics which are required to defined the deformation parameters $\beta_{lm}$ used in this thesis (see Appendix B.2).

B.1.1 $l = 1, m = 0$

$$Y_{10}(\theta, \varphi) = \frac{1}{2} \sqrt{\frac{3}{\pi}} \cos \theta$$
$$= \frac{1}{2} \sqrt{\frac{3}{\pi}} \frac{z}{r} \quad (B.4)$$
B. SPHERICAL HARMONICS AND DEFORMATION PARAMETERS

B.1.2 \( l = 2, m = 0 \)

\[
{Y_{20}(\theta, \varphi)} = \frac{1}{4} \sqrt{\frac{5}{\pi}} \left( 3 \cos^2 \theta - 1 \right) = \frac{1}{4} \sqrt{\frac{5}{\pi}} \left( \frac{2z^2 - x^2 - y^2}{r^2} \right)
\]

(B.5)

B.1.3 \( l = 3, m = 0 \)

\[
{Y_{30}(\theta, \varphi)} = \frac{1}{4} \sqrt{\frac{7}{\pi}} \left( 5 \cos^3 \theta - 3 \cos \theta \right) = \frac{1}{4} \sqrt{\frac{7}{\pi}} \left( \frac{2z^3 - 3x^2z - 3y^2z}{r^3} \right)
\]

(B.6)

B.1.4 \( l = 4, m = 0 \)

\[
{Y_{40}(\theta, \varphi)} = \frac{3}{16} \sqrt{\frac{1}{\pi}} \left( 35 \cos^4 \theta - 30 \cos^2 \theta + 3 \right) = \frac{3}{16} \sqrt{\frac{1}{\pi}} \left( \frac{8z^4 - 24x^2z^2 - 24y^2z^2 + 3(x^2 + y^2)^2}{r^4} \right).
\]

(B.7)

B.1.5 \( l = 2, m = 2 \)

The real part of the spherical harmonic \( Y_{22} \) is needed (as well as \( Y_{20} \)) to define the Bohr-Mottleson \( \beta, \gamma \) parametrisation. The spherical harmonic is given by

\[
{Y_{22}(\theta, \varphi)} = \frac{1}{4} \sqrt{\frac{15}{2\pi}} e^{2i\varphi} \sin^2 \theta = \frac{1}{4} \sqrt{\frac{15}{2\pi}} \frac{(x + iy)^2}{r^2},
\]

and the real part is

\[
{Y_{22}(\theta, \varphi)} = \frac{1}{4} \sqrt{\frac{15}{2\pi}} \frac{x^2 - y^2}{r^2}.
\]

(B.8)

(B.9)

B.2 Deformation Parameters

The multipole moment \( Q_{lm} \) is defined with respect to the centre of mass of the system by the integral [57]

\[
Q_{lm} = \int \rho(r) r^l Y_{lm} \, dr,
\]

(B.10)

where \( \rho(r) \) is the particle density and \( Y_{l0} \) the spherical harmonic defined previously. The dimensionless deformation parameter \( \beta_{20} \) is defined from the quadrupole moment \( Q_{20} \) [111]:

\[
\beta_{20} = \frac{4\pi}{5} \frac{Q_{20}}{A(r^2)}
\]

(B.11)
where here, \(\langle r^2 \rangle\) is the root mean square radius defined by

\[
\langle r^n \rangle = \frac{\int d^3r \, r^n \rho(r)}{A},
\]  \hspace{1cm} (B.12)

for the case \(n = 2\), and \(A\) is the particle number.

Following the definition of the quadrupole deformation parameter \(\beta_{20}\), deformation parameters of a different order are defined:

\[
\beta_{10} = \frac{4\pi}{5} Q_{10} A(r),
\]

\[
\beta_{30} = \frac{4\pi}{5} Q_{30} A(r^3),
\]

\[
\beta_{40} = \frac{4\pi}{5} Q_{40} A(r^4).
\]

The \(\frac{4\pi}{5}\) constant scaling factor is included in the definition of \(\beta_{20}\) in Ref. [111], so it has been adopted for consistency with the other deformation parameters.

The Bohr-Mottelson \(\beta, \gamma\) parameters may be defined also, which gives a measurement of the quadrupole deformation and the corresponding axial asymmetry [42]. In this case, the deformation parameter \(\beta_{22}\) must be additionally considered. The parameter \(\beta_{22}\) is defined from \(Q_{22}\), and is given by

\[
\beta_{22} = \frac{4\pi}{5} \frac{5}{96\pi} \frac{Q_{22}}{Ar^2}.
\]  \hspace{1cm} (B.13)

The parameter \(\beta\) is defined by combining \(\beta_{20}\) and \(\beta_{22}\)

\[
\beta = \sqrt{(\beta_{20})^2 + 2(\beta_{22})^2},
\]  \hspace{1cm} (B.14)

and \(\gamma\) is defined by

\[
\gamma = \text{atan} \left( \frac{\sqrt{2} \beta_{22}}{\beta_{20}} \right).
\]  \hspace{1cm} (B.15)

The principle value of the atan function is in the range \(-\pi \rightarrow \pi\), so \(2\pi\) must be added to negative values of \(\gamma\). The parameter \(\gamma\) can take a range of values from \(0^\circ \rightarrow 360^\circ\), but only the \(0^\circ \rightarrow 60^\circ\) range is of interest, as the other sectors correspond to equivalent configurations [42]. A value of \(\gamma = 0^\circ\) corresponds to an axially symmetric prolate deformed shape and a value of \(\gamma = 60^\circ\) corresponds to an axially symmetric oblate deformed shape. Any other values of \(\gamma\) correspond to a triaxial deformation.
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