FISSION IN EXOTIC NUCLEI USING DENSITY FUNCTIONAL THEORY

By

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ABSTRACT

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This is my abstract.

Dedicated to I dunno

ACKNOWLEDGMENTS

I dunno who to acknowledge, either

TABLE OF CONTENTS

LIST OF FIGURES

Chapter 1

Introduction

1.1 History of Fission Theory

1.1.1 Liquid Drop Model

1.1.2 Strutinsky shell correction

1.1.3 Self-consistent models and the supercomputing era

[You should probably cite the fission discovery paper(s) by Hahn and Straßmann [20] and the subsequent qualitative explanation paper by Meitner and Frisch [26], not to mention the original liquid drop paper by Weizsäcker [44] and the paper in which Bohr and Wheeler invoked the liquid drop model to describe fission quantitatively in terms of bulk properties of nuclei [11]. Finally, I might also do well to mention the spontaneous fission discovery paper [17], which is actually just a letter to the editor of Physical Review that is only a paragraph long.] Models have grown increasingly sophisticated over time (the development of the Strutinsky shell correction to the LDM energy and the Funny Hills paper [40, 41, 12], which incorporated nuclear shell effects and explained the experimental observation that fragments were not equally-sized); however, the problem is incredibly complex. First of all, one must solve the nuclear many-body problem for a large (A>200) system. Then one must describe a transition from a single body to two. The way we do each of these things is described in Chapter 2.

If you're looking for a central narrative with which to tie together your thesis, you could, of course, use the whole "making things faster" angle you've been playing so far. But I think a more enriching, exciting, and satisfying approach would be to emphasize that you are doing fission calculations for *rare* nuclei. That's cool because you work in a facility for *rare* isotopes! And it's just one of those things that is interesting and fashionable in the field in general right now. The introduction to the platinum-178 paper has a good discussion about the importance of trying to understand fission in regions of exotic isospin ratios, and how simpler models tend to be less reliable in those regions. That covers both the platinum and the r-process project motivations (at least partially), and oganesson is just interesting because of how heavy it is.

It is an exciting time to study nuclear theory; major advances are now possible thanks to a groundwork lain of nuclear theory, paired with modern supercomputers fast enough for such complicated many-body problems to be solved.

It is an exciting time to study nuclear theory; for as we now enter the supercomputer age, we are able to implement the groundwork laid over the past several decades on modern, cutting-edge, high-performance computing centers. This allows

These advances in computing come simultaneously with advances in accelerator design and technology and other advances which allow experimental nuclear physics to reach far beyond what has been done before. For instance, the Facility for Rare Isotope Beams (FRIB) at Michigan State University is projected to be able to nearly double the number of isotopes that can be produced synthetically. Together, state-of-the-art facilities for experiment and high-performance computing for theory are expected to lead to rapid advancement in our understanding of atomic nuclei.

One process which has always been a driver of nuclear physics is nuclear fission, the process by which a heavy nucleus decays into two smaller nuclei of approximately equal mass. Nuclear fission has been applied by humans in the fields of energy generation and national defense, and it has been predicted to play a major role in astrophysical environments such as neutron star mergers. There is currently a great deal of interest in understanding "rare" isotopes, or isotopes which are highly-unstable, in order to better understand such exotic phenomena as neutron star mergers, as well as to better identify physical properties which we can use to better understand the nuclei which we regularly encounter on Earth.

There are a couple of ways for a nucleus to fission. One way is by imparting some excitation energy to a fissile nucleus, such as by bombarding a nucleus with neutrons (neutron-induced fission), by creating an excited nucleus as the decay product of another isotope (beta-delayed fission) or as a compound system of two collided nuclei. Owing to the randomness of quantum mechanics, another possibilty is for a nucleus in its ground state to spontaneously tunnel through a potential barrier and then emerge to form two distinct fragment (spontaneous fission). This dissertation will deal primarily/exclusively with the latter.

Fission, the fundamental process by which a single heavy nucleus splits into two smaller nuclei and a few emitted neutrons, is simple to understand qualitatively but remarkably difficult to explain quantitatively. One could argue that nuclear fission theory has leapt forward in three major waves. The first major wave of nuclear fission theory goes back to the very beginning of the nuclear age, when George Gamow proposed and Niels Bohr and John Archibald Wheeler developed the liquid drop model in the 1930s. This model was able to successfully describe nuclear binding energies and the energetics of nuclear fission. The second wave came with Strutinsky's microscopic correction in the late 1960s, which essentially amounted to adding a quantum mechanical correction to the liquid drop energy. This correction, based on the nuclear shell model, is added in order to better account for the added stability that occurs when a nucleus contains a “magic number” of protons and/or neutrons [40, 41, 12]. The third major wave is taking place now, heralded by the age of supercomputers. Now instead of using phenomenology and quantum corrections to describe heavy nuclei, we can use quantum many-body methods which were developed years or decades ago, but which were shelved until sufficiently-powerful computers came online in recent years.

Microscopic models (as they are called) are increasingly able to predict properties of fission fragments; however, a comprehensive description of fission fragments (including mass and fragment yields, excitation and kinetic energy distributions, angular dependence, spin, neutron emission) in a microscopic framework remains elusive. A major source of this elusiveness is due to the sheer difficulty of describing a smooth transition from one nucleus to two, a concept which is plagued with ambiguities. How can one precisely identify two distinct fragments when the wavefunctions of one fragment’s constituent nucleons may extend into the opposite fragment? And how do those correlations between nucleons affect the energeties of the resulting fragments? These are the questions to be addressed by this project.

Theoretically, making predictions about fission is challenging because, thanks to the large number of particles involved and the complex collective interactions which take place when one system deforms and becomes two, fission calculations have an ”inextinguishable thirst for computing power,” as stated in [39]. Historically, most fission calculations that have been done were based on empirical formulas or phenomenological models, most notably the “microscopic-macroscopic” family of models based on Bohr’s liquid-drop model (to model the bulk properties of the nucleus) with Strutinsky shell corrections (to account for quantum mechanical shell effects). These microscopic-macroscopic (“micmac”) fission models are computationally fairly inexpensive, and can achieve quite satisfactory results. However, since the model is based on a phenomenological description of what is actually a quantum mechanical system, its predictive power is limited, and there is no clear way of making systematic improvements.

A more reliable approach would be to consider the individual nucleon states using some kind of quantum many-body method. For large systems with many, many particles, density functional theory (DFT) is a way to recast the Schrödinger equation involving ∼200 particles into a simpler problem involving only a few densities and currents (see section 2.1.1). With DFT as a way of calculating nuclear properties quantum-mechanically, one can then use these self-consistent solutions to predict fission properties, such as lifetimes and fragment yields. Fortuntately, a great deal of work has been done to achieve exactly this (see the review article [39]). Some of the ideas which are used were inspired by lessons learned from micmac and other, simpler models; others are unique to DFT. Our approach is described in detail in chapter 2.

The challenge, now, is to do these calculations cheaply. In every theoretical calculation, one must ask oneself "What approximations can I safely make?" and "What are the important degrees of freedom for this problem?" One may also reduce the total time-to-answer via improvements to the computational workflow itself, such as better file handling and parallelization.

1.2 Goals of the project

By far the most commonly-studied region so far for fission has been the region of actinides near ^{235}U, which includes isotopes of uranium, plutonium, and thorium relevant for nuclear energy/reactor physics and stockpile stewardship/defense. Given the aforementioned recent interest in rare and exotic nuclei, we have applied our model to several exotic systems which undergo spontaneous fission in different regions of the nuclear chart. First in chapter 3 we discuss bimodal fission in the neutron-deficient isotope platinum-178, which until recently was expected to fission symmetrically. Then in chapter 4 we discuss cluster radioactivity in oganesson-294, the heaviest element ever produced by humans. In chapter 5 we move to the neutron-rich side of the nuclear chart to study ..., which are expected to play a major role in the astrophysical r-process. Finally, in chapter 6 we discuss the current state of the field, and, based on our experience, offer insights for guiding future developments in the field.

Chapter 2

Describing Fission Using Nuclear Density Functional Theory

(Nicolas gave a good annotated presentation in 2017 that describes some of the philosophy, as well as some of the outstanding challenges of spontaneous fission in an adiabatic framework: `https://t2.lanl.gov/fiesta2017/school/Schunck_NotesSlides.pdf`)

Fundamentally, the assumption of adiabaticity is the assumption that collective and intrinsic degrees of freedom can be decoupled, and that the time-scale associated with collective degrees of freedom is slow compared to the time-scale of intrinsic (i.e. single-particle) motions of the system.

Today there are 2 microscopic approaches to fission in common use: time-dependent and static (time-independent) (or 3 models: time-dependent, static, and statistical). Time-dependent approaches evolve the system in real-time. Since fission is an inherently time-dependent process, these methods offer great insight into the fission process and the characteristics of the fragments `\cite{Some TDHFB papers}`. However, they can only treat a single event at a time, making them impractical for estimating yields. Furthermore, despite efforts such as [14], there is currently no way to obtain a full yield distribution in a time-dependent framework.

On the other hand, static approaches assume that collective motion of the nucleus is slow compared to the motion of the intrinsic particles, and this assumption is used to create a potential energy surface in some space of collective shape coordinates. We can estimate fission yields in these approaches (see, for instance, [36] along with our forthcoming paper on ^{294}Og), but for practical reasons we are limited to describing complicated shapes in terms of just a few parameters, leading to uncertainty in the fragment properties. In particular, the part of the process at which the neck snaps and one nucleus becomes two, called scission, is not well-defined in static approaches.

This can be understood with an analogy: suppose we stretch a nucleus until a neck forms, and then we use a butcher knife to lop the two fragments apart. This works reasonably well for estimating fragment mass and charge, but it is very poor when it comes to estimating the relative energy of the fragments. To estimate fragment kinetic and excitation energies, one needs to carefully and delicately peel the interlocking fragments apart with a scalpel, or a proper accounting of entanglement and other many-body correlations.

In static approaches, scission is frequently characterized by a single number, corresponding to the number of particles in the neck. When that number falls below a certain predefined threshold, we say that the nucleus has scissioned. Fragments are identified and one can try to estimate the strength of the repulsive interaction forcing the fragments apart. Of course, as discussed by Younes and Gogny in [**?**, 46], wavefunctions corresponding to individual nucleons may extend into the spatial region of the opposite fragment.

Our approach, which combines the Hartree-Fock-Bogoliubov (HFB) mean-field approximation to the energy with a many-body method inspired by Kohn-Sham density functional theory (DFT), is described below.

Spontaneous nuclear fission is a type of quantum tunneling; consequently, it should be described using quantum mechanics.

2.1 Nuclear Density Functional Theory

Since nuclei are quantum mechanical systems, they can in principle be described using the Schrodinger equation. However, in practice one finds this type of description difficult or impossible, for two reasons:

- In order to use the Schrodinger equation, one needs to know how to describe the interaction between particles, such as between protons and neutrons. However, protons and neutrons are made up of quarks and gluons, which interact via the strong nuclear force. Consequently, an analytic expression for the nucleon-nucleon interaction analogous to the $\frac{1}{r}$ form of the Coulomb interaction is not available. Finding different mathematical expressions which can describe the interaction between nucleons continues to be an active area of research [**?**]

- Even when an interaction is known, nuclei are large systems made up of many protons and neutrons. Solving the Schrodinger equation directly quickly becomes computationally intractable as the number of nucleons increases.

2.1.1 Density Functional Theory

Kohn-Sham DFT is based on the Hohenberg-Kohn theorems

Let us define the nucleon density in the following way: suppose we have a system described in second quantization by a set of creation and annhilation operators $c_i, c_i^\dagger$ which act on the [Fock-space?] vacuum state $|\psi_0\rangle$. The first Kohn-Sham theorem says that the energy of the system is a uniquely-defined functional of the density. That means that if a system of interacting particles and a system of noninteracting particles give the same density, the energy of those systems will be the same. This gives us the freedom to try to describe our system using a mean-field method instead of having to describe the pairwise interactions between every particle in the system - a huge simplification to the problem!

The second Kohn-Sham theorem states that the functional which gives the energy of the system will give the ground state energy if, and only if, it acts on the true ground state density. Thus, given a particular functional, we can vary the input density to minimize the total energy and be assured that we are approaching the ground state energy of the system.

Suppose you have the density $\rho(\mathbf{r})$ of an interacting system of particles. There exists a unique noninteracting system with the same density Then I believe HFB is put on top of that to do the variation part. I think I (approximately) get it now! - So just to make sure, what would DFT look like without HF/HFB? And HF/HFB without DFT?

Rather than find the density of a system of interacting particles (which can be extremely complicated - as one particle moves, the force it exerts on neighboring particles causes them to move, which will in turn change the magnitude and direction of the net force acting on the original particle, and so on until an equilibrium configuration, if it exists, can be attained), Kohn-Sham allows us to find an equivalent density of fictional non-interacting particles. That is, instead of particles moving in a field generated by many interdependent neighboring particles, one may think of non-interacting particles moving about a mean-field, which is essentially an averaging over all other particles.

Together, the Hohenberg-Kohn theorems state that if one is able to find the true ground state density, regardless of where it comes from, then there exists a unique functional of the density which gives the ground state energy of the system. However, HK do not specify how this functional is to be obtained.

For a variety of reasons/complications (refs 73-78 of [39]), pure Kohn-Sham is not used in nuclear physics; however; in the spirit, we oftentimes switch to a representation involving densities (which are directly and exactly attainable from a many-body wavefunction) and energy density functionals (which are not known exactly). (Wait, but then what is the point of converting to densities? Why not just leave them as wavefunctions? Or maybe we do, but this representation just makes the math look nicer for papers)

The basic idea is to replace the single particle states $c_i^\dagger |\psi_0\rangle$ with the single-particle density, $\rho_{ij} = \langle\psi_0|c_j^\dagger c_i|\psi_0\rangle$

Because pairing interactions are of great importance to nuclear dynamics, we also construct an additional density $\kappa_{ij} = \langle\psi_0|c_j c_i|\psi_0\rangle$, which can be thought of as a coupling between the vacuum state and a state with two particles (in states i and j). Together with the single-particle density ρ we construct a generalized density

$$\mathcal{R} = \begin{pmatrix} \rho & \kappa \\ -\kappa^* & 1-\rho^* \end{pmatrix} \tag{2.1}$$

[These will probably need to be brought into a more compliant form; make sure you've got the same notation everywhere in the thesis] (with q=isospin and σ=spin/signature quantum number)

$$\rho_{q\sigma}(\mathbf{r}) = \sum_{\alpha\in q} v_\alpha^2 \left|\psi_\alpha(\mathbf{r}\sigma)\right|^2 \tag{2.2}$$

$$\tau_{q\sigma}(\mathbf{r}) = \sum_{\alpha\in q} v_\alpha^2 \left|\nabla\psi_\alpha(\mathbf{r}\sigma)\right|^2 \tag{2.3}$$

$$\mathbf{j}_{q\sigma}(\mathbf{r}) = \sum_{\alpha\in q} v_\alpha^2 \mathrm{Im}\left[\psi_\alpha^*(\mathbf{r}\sigma)\nabla\psi_\alpha(\mathbf{r}\sigma)\right] \tag{2.4}$$

$$\nabla\rho_{q\sigma}(\mathbf{r}) = 2\sum_{\alpha\in q} v_\alpha^2 \mathrm{Re}\left[\psi_\alpha^*(\mathbf{r}\sigma)\nabla\psi_\alpha(\mathbf{r}\sigma)\right] \tag{2.5}$$

$$\kappa = ??? \tag{2.6}$$

The total energy is a sum of several contributions:

$$E(\rho, \kappa) = E_{kin} + E_{Coul} + E_{nuc} + E_{pair} \tag{2.7}$$

where E_{kin} is the kinetic energy term, E_{Coul} contains the Coulomb ineraction between protons, E_{nuc} is a phenomenological nucleon-nucleon interaction term, and E_{pair} describes the tendency of nucleons to form pairs, which is smeared out in non-interacting mean-field models. Finding a good nucleon-nucleon interaction E_{nuc} (and to a lessert extent, E_{pair}) to use in calculations is an active topic of research in nuclear theory today (for one recent example, see [28]); two types of interactions which are commonly-used today are the Skyrme and Gogny families of interactions `\cite{???}`. We use primarily Skyrme-type interactions, which are described below.

2.1.1.1 Kinetic term

Defining τ_0 as in eqn 10 of HFODD-I, the kinetic energy contribution is

$$E_{kin} = \frac{\hbar^2}{2m}\left(1 - \frac{1}{A}\right)\int \tau(\vec{r})d^3\vec{r} \tag{2.8}$$

2.1.1.2 Coulomb interaction

The Coulomb interaction between protons is divided into a direct term and an exchange term, which is related to the Pauli exclusion principle.

$$E_{Coul} = E_{Coul,dir} + E_{Coul,exch} \tag{2.9}$$

$$E_{Coul,dir} = \frac{e^2}{2}\int \frac{\rho_p(\vec{r}_1)\rho_p(\vec{r}_2)}{|\vec{r}_1 - \vec{r}_2|}d^3\vec{r}_1 d^3\vec{r}_2 \tag{2.10}$$

$$E_{Coul,exch} = \frac{e^2}{2}\int \frac{\rho_p(\vec{r}_2,\vec{r}_1)\rho_p(\vec{r}_1,\vec{r}_2)}{|\vec{r}_1 - \vec{r}_2|}d^3\vec{r}_1 d^3\vec{r}_2 \tag{2.11}$$

Often the exchange term is computed in the Slater approximation `\cite{refs 27,28 of HFODD-I}`:

$$E_{Coul,exch} \approx -\frac{3e^2}{4}\left(\frac{3}{\pi}\right)^{\frac{1}{3}}\int \rho_p^{\frac{4}{3}}(\vec{r})d^3\vec{r} \tag{2.12}$$

2.1.1.3 Skyrme interaction

The total Skyrme interaction energy density is a sum of both time-even and time-odd terms:

$$E_{Skyrme} = \int \sum_{t=0,1}\left(\mathcal{H}_t^{even} + \mathcal{H}_t^{odd}\right)d^3\vec{r} \tag{2.13}$$

$$\mathcal{H}_t^{even} = C_t^{\rho}\rho_t^2 + C_t^{\Delta\rho}\rho_t\Delta\rho_t + C_t^{\tau}\rho_t\tau_t + C_t^{J}\mathsf{J}_t^2 + C_t^{\nabla J}\rho_t\nabla\cdot\vec{J}_t \tag{2.14}$$

$$\mathcal{H}_t^{odd} = C_t^{s}\vec{s}_t^{\,2} + C_t^{\Delta s}\vec{s}_t\Delta\vec{s}_t + C_t^{T}\vec{s}_t\cdot\vec{T}_t + C_t^{j}\mathsf{j}_t^2 + C_t^{\nabla j}\vec{s}_t\cdot(\nabla\times\vec{j}_t) \tag{2.15}$$

where τ_t is the kinetic energy density; J_t is the spin current density, with vector part given by $\vec{J}_{\kappa,t} = \sum_{\mu\nu} \epsilon_{\mu\nu\kappa} \mathsf{J}_{\mu\nu,t}$; $\vec{s}_t$ is the spin density, $\vec{T}_t$ is the spin kinetic density; and $\vec{j}_t$ is the momentum density (to see how these are related to ρ_t, see, e.g., [9]). The index $t = 0(1)$ refers to isoscalar(isovector) energy densities. Note that $\mathcal{H}_t^{even}$ depends only on time-even densities (and likewise for $\mathcal{H}_t^{odd}$).

Since this interaction is phenomenological, based on a zero-range contact interaction between nucleons, the coefficients are adjustable. There are dozens of Skyrme parameterizations on the market, each one optimized to a particular observable or set of observables. The parameter sets SkM* [8] and UNEDF1 [22] (along with its sister, $\text{UNEDF1}_{\text{HFB}}$ [38]) are optimized to datasets which include deformed nuclei, making them suitable for fission.

2.1.1.4 Pairing interaction

We use a density-dependent pairing interaction:

$$E_{pair} = V_0 \int \left(1 - \left(\frac{\rho(\vec{r})}{\rho_0}\right)^{\alpha}\right) d^3\vec{r} \tag{2.16}$$

As with the nuclear interaction term, the pairing interaction contains several adjustable parameters.

2.1.2 Bogoliubov transformation

In anticipation of the HFB formalism below, we define the so-called Bogoliubov transformation. The fundamental entity in the Bogoliubov transformed basis are ‘quasiparticle’ states, defined by quasiparticle creation and annhilation operators acting on a quasiparticle vacuum state $|\Phi_0\rangle$ (in contrast to the single particle operators from before). The creation and annhilation operators are given by

$$\beta_\mu = \sum_i U^*_{i\mu} c_i + \sum_i V^*_{i\mu} c^\dagger_i \tag{2.17}$$

$$\beta^\dagger_\mu = \sum_i U_{i\mu} c^\dagger_i + \sum_i V_{i\mu} c_i \tag{2.18}$$

or in block matrix notation,

$$\begin{pmatrix} \beta \\ \beta^\dagger \end{pmatrix} = \begin{pmatrix} U^\dagger & V^\dagger \\ V^T & U^T \end{pmatrix} \begin{pmatrix} c \\ c^\dagger \end{pmatrix} \equiv \mathcal{W}^\dagger \begin{pmatrix} c \\ c^\dagger \end{pmatrix} \tag{2.19}$$

where the transformation matrix $\mathcal{W}$ must be unitary to ensure that $\beta, \beta^\dagger$ obey the fermion commutation relations [33]. In this transformed basis, the density matrix takes the form

$$\mathsf{R} = \mathcal{W}^\dagger \mathcal{R} \mathcal{W} = \begin{pmatrix} \langle\Phi_0|\beta^\dagger_\mu \beta_\nu|\Phi_0\rangle & \langle\Phi_0|\beta_\mu \beta_\nu|\Phi_0\rangle \\ \langle\Phi_0|\beta^\dagger_\mu \beta^\dagger_\nu|\Phi_0\rangle & \langle\Phi_0|\beta_\mu \beta^\dagger_\nu|\Phi_0\rangle \end{pmatrix} = \begin{pmatrix} 0 & 0 \\ 0 & I_N \end{pmatrix} \tag{2.20}$$

... and the densities ρ and κ can be written ...

In general, E is a functional of the generalized density $\mathcal{R}$.

2.1.3 Hartree-Fock-Bogoliubov Equations

The ground state configuration of the system described by this particular energy density functional E is described by the density which minimizes $E(\mathcal{R})$. We can find this solution through the variational principle. We minimize the energy with respect to the generalized density, subject to the constraint that $\mathcal{R}^2 = \mathcal{R}$, or in other words, that the state remains a quasiparticle vacuum. Defining the HFB Hamiltonian $\mathcal{H}_{ba} \equiv 2\partial E/\partial \mathcal{R}_{ab}$, this variation leads to the result $[\mathcal{H}, \mathcal{R}] = 0$, which is called the Hartree-Fock-Bogoliubov equation. It is not typically solved in this form, but it can be recast into something more useful. Recalling that two Hermitian operators whose commutator is zero can be simultaneously diagonalized, we choose to diagonalize $\mathcal{H}$ using the same Bogoliubov transformation W which diagonalizes $\mathcal{R}$:

$$W^\dagger \mathcal{H} W \equiv \mathcal{E} \qquad \text{or} \qquad \mathcal{H} W = W \mathcal{E} \tag{2.21}$$

where

$$\mathcal{E} = \begin{pmatrix} E_\mu & 0 \\ 0 & -E_\mu \end{pmatrix} \tag{2.22}$$

is a matrix of quasiparticle energies.

Very often we will want to minimize the energy with the system subject to a particular constraint. Some common examples might be this simple form of particle number restoration (more complicated forms, such as Lipkin-Nogami `\cite{???}`, also exist)

$$\delta E' = \langle \Phi_0 | \hat{H} - \lambda \hat{N} | \Phi_0 \rangle \tag{2.23}$$

or shape, where we might use a particular multipole moment (or set of multipole moments)

$\hat{H} - \lambda \hat{N} - \lambda_i \hat{Q}_i$

Lipkin-Nogami is related to this, but I forget exactly how at the moment: [Note: 14 November 2017] It turns out I'm not quite right about this. The "pure HFB/static pairing case" refers to the case described by $H' = H - \lambda N$. There are still (so far as I understand) dynamical pairing fluctuations. Then, once you add another term $H'' = H' + \lambda_2 \Delta N^2$ you'll obtain even stronger pairing fluctuations

2.1.4 Collective inertia

Just as important to the fission dynamics aas the energy of the system is the collective inertia, which describes the tendency of the system to resist configuration changes (such as shape changes). The form of the collective inertia we use is the non-perturbative adiabatic time-dependent HFB (ATDHFB) inertia with cranking, which takes the form (see the full temperature-dependent derivation in Appendix ???

$$\mathcal{M} = \dots \tag{2.24}$$

A perturbative expression for the ATDHFB inertia also exists, which allows one to estimate the inertia without computing derivatives. It is computationally much faster and easier to implement, but it is less accurate and loses many of the important features of the inertia, as we shall see in Chapter ??? (294Og). Nevertheless, it is commonly-used in calculations and we shall use it later on.

Another common expression for the collective inertia comes from the Generator Coordinate Method (GCM). The GCM inertia also exists in two varieties: perturbative and non-perturbative [19]. Like the ATDHFB inertia, the perturbative GCM inertia is smoothed-out compared to the non-perturbative inertia. Both the perturbative and non-perturbative GCM inertias are found to be smaller in magnitude than their ATDHFB counterparts.

2.1.5 Nucleon localization function

One of the tools we will be using quite a bit in this thesis is the nucleon localization function (NLF), introduced in [47]. The NLF is defined using the single particle density in the following way (with q=isospin and σ=spin/signature quantum number):

$$\mathcal{C}_{q\sigma} = \left[1 + \left(\frac{\tau_{q\sigma}\rho_{q\sigma} - \frac{1}{4}|\nabla\rho_{q\sigma}|^2 - \mathbf{j}^2_{q\sigma}}{\rho_{q\sigma}\tau^{TF}_{q\sigma}} \right)^2 \right] \tag{2.25}$$

where $\tau^{TF}_{q\sigma} = \frac{3}{5}(6\pi^2)^{\frac{2}{3}}\rho^{\frac{5}{3}}_{q\sigma}$. A localization value $\mathcal{C} \approx 1$ means that nucleons are well-localized; that is, the probability of finding two nucleons of equal spin and isospin at the same location in space is low. A value of $\mathcal{C} = \frac{1}{2}$ corresponds to a Fermi gas of nucleons, as found in nuclear matter.

The NLF offers greater insight into the underlying shell structure of the system than, for instance, the single particle density. In particular, when applied to fission as in [37], it sometimes enables one to see the formation of well-defined prefragments whose shell structure is responsible for the peak of the fragment distribution.

2.2 Microscopic Description of Nuclear Fission

Should I describe these here, or in the sections when they actually get used?

With the nuclear physics somewhat under control, we now move onto the problem of using it to actually describe fission. For induced-fission, time-dependent density functional theory (TDDFT) allows one to calculate the time-evolution of a nucleus starting from some deformed initial configuration. So far, though, this approach has not been able to estimate a full yield for a fissioning nucleus; rather, the system propogates deterministically to a single scissioned configuration. Furthermore, and especially important for the case of spontaneous fission, the time-dependent approach does not allow for tunneling (why not?).

Nuclear fission is the fundamental physical process by which a heavy nucleus decays to two smaller nuclei with approximately equal masses, and a proper understanding of fission is critical for applications . It is a highly-collective process involving all the constituent nucleons of the system, and thus since its discovery it has been described via large shape deformations of an otherwise spherical "drop" of nucleons. In this framework, which is formalized by the adiabatic approximation, it falls upon theorists to describe many different nuclear shapes. In principle, one could describe any three-dimensional shape using an infinite basis such as the multipole expansion which is often encountered in eletrodynamics; however, for practical computations one must used a truncated set of only a few multipole moments (or, more generally, collective coordinates). Thus, an important challenge for researchers is to select the most relevant collective coordinates, ideally while demonstrating that others can be safely neglected.

Recently in [36], an approach based on this assumption was used to compute fragment yields from a potential energy surface (PES) that was computed self-consistently, using the WKB approximation to describe the tunneling and Langevin dynamics to describe post-scission dissipation. Now we test robustness of these results by exploring the impact of the energy density functional, the size of the collective space, and the calculation of the collective inertia on fragment yields.

2.2.1 Potential Energy Surfaces

Constrained HFB; map out many different constrained calculations and start to form a surface which resembles a topographical map. How do you decide which collective coordinates to use? Including pairing...

Physically, a nucleus can be thought of as a jumble of particles, each bouncing around in a potential well determined by the surrounding nucleons. Qunatum mechanics forbids us from determining the particle trajectories exactly; however, it allows us to estimate the probabilities of certain outcomes. In the case of fission, the most common approach has us thinking of nucleons grouping together collectively in a way which resembles a liquid drop (footnote: this idea was first proposed by Niels Bohr, I believe, and has proven to be a very fruitful way to describe fission). The collective shape is constrained to nearly-spherical shapes by a potential barrier; however, being a quantum mechanical system there is some nonzero tunneling probability, or a probability that the barrier will be penetrated, and the collective shape will stretch beyond the size fixed by the barrier. When this happens, the nucleus may remain in a long-term elongated state (called a fission isomer), or it may continue to deform until it separates into two fragments.

2.2.2 WKB Approximation

Adiabaticity: For fusion reactions, N,Z equilibrium reached in $\sim 10^{-21}$ seconds, then energy/thermal equilibrium in a similar time scale, then finally mass equilibrium in $\sim 10^{-19}$ - Yuri has a slide with these time scales from his talk Monday

Sort of an adiabatic approximation; useful because half-lives are long and therefore time-dependent approaches are impractical (they break down and/or become unstable or something after too many time steps, not to mention the amount of computing time). Wavefunction is assumed to be slowly-varying inside the potential barrier

Furthermore, TDHFB cannot tunnel

Consider a set of collective coordinates $\mathbf{q} \equiv (q_1, \ldots, q_N)$. In our implementation of the

WKB approximation, the most-probable tunneling path $L(s)|_{s_{\text{in}}}^{s_{\text{out}}}$ in the collective space is found via minimization of the collective action

$$S(L) = \frac{1}{\hbar} \int_{s_{\text{in}}}^{s_{\text{out}}} \sqrt{2\mathcal{M}(s)\left(V(s) - E_0\right)} ds, \tag{2.26}$$

where s is the curvilinear coordinate along the path L, $\mathcal{M}(s)$ is the collective inertia [35] and $V(s)$ is the potential energy along $L(s)$. E_0 stands for the collective ground-state energy. The dynamic programming method [6] is employed to determine the path $L(s)$. The calculation is repeated for different outer turning points, and each of these points is then assigned an exit probability $P(s_{\text{out}}) = [1 + \exp\{(2S)\}]^{-1}$ [4].

2.2.3 Langevin Dynamics

After emerging from the classically-forbidden region of the PES, fission trajectories begin from the outer turning line and then evolve along the PES according to the Langevin equations:

$$\frac{dp_i}{dt} = -\frac{p_j p_k}{2} \frac{\partial}{\partial q_i} \left(\mathcal{M}^{-1}\right)_{jk} - \frac{\partial V}{\partial q_i} - \eta_{ij} \left(\mathcal{M}^{-1}\right)_{jk} p_k + g_{ij} \Gamma_j(t)\,, \tag{2.27}$$

$$\frac{dq_i}{dt} = \left(\mathcal{M}^{-1}\right)_{ij} p_j\,, \tag{2.28}$$

where p_i is the collective momentum conjugate to q_i. The dissipation tensor η_{ij} is related to the random force strength g_{ij} via the fluctuation-dissipation theorem, and $\Gamma_j(t)$ is a Gaussian-distributed, time-dependent stochastic variable.

The fluctuation-dissipation theorem is given by the expression $\sum_k g_{ik} g_{jk} = \eta_{ij} k_B T$. It effectively couples the collective and intrinsic via the system temperature, given by $k_B T =$

$\sqrt{E^*/a}$ where $a = A/10\text{MeV}^{-1}$ parameterizes the level density and the excitation energy $E^* = V(s_{out}) - V(\mathbf{x}) - \frac{1}{2}\sum\left(\mathcal{M}^{-1}\right)_{ij} p_i p_j$.

It has been shown (Jhilam and Nicolas' paper) that fission yields are fairly robust with respect to the dissipation strength

Dissipation is treated in our work as a parameter, as a self-consistent description of dissipation is not yet known. However, work along this line has been started (maybe?) in refs 291-293 of [**?**] (see section 4.1.1 for the context).

Chapter 3

Two fission modes in ^{178}Pt

3.1 Asymmetric fission in the region of ^{180}Hg

As mentioned in the introduction, fission is most well-studied in the region of the actinides (Z=90 to Z=103), as many naturally-occurring isotopes in this region are fissile. Within this region, there is a characteristic tendency for fission fragment yields to be asymmetric (that is, one light fragment and one heavy fragment), with the heavy peak centered around $A \approx 140$. This has been understood as a manifestation of nuclear shell structure in the prefragments: doubly-magic ^{132}Sn drives the nucleus towards scission, and once the neck nucleons are divided up between the two fragments, we end up with the heavy fragment A=140 peak. As one moves to the lower-Z actinides, however, this tendency becomes less and less pronounced as yields tend to become more symmetric. Below thorium, it was generally believed until recently (though mostly not tested) that yields would continue to be symmetric as there was no doubly-magic nucleus candidate that could drive the system toward asymmetry as there is with actinides.

However, it was reported in a 2010 study [1] that neutron-deficient ^{180}Tl undergoes beta-delayed fission, leading to intermediate state $^{180}_{80}$Hg$_{100}$ which then decays into two fragments of unequal mass. This finding triggered a flurry of theoretical papers hoping to describe this new and unexpected phenomenon. A follow-up study using ^{178}Tl [23] further established this as a region of asymmetric fission, and not just a one-time occurence. Since then, other nuclei in the region have been studied, for instance using Coulex-induced fission reactions and compound nucleus (prompt?) fusion-fission reactions, and the finding is the same.

Nuclei in this region have a number of unique features which make them interesting for study, even aside from the unexpected fragment asymmetry. Predicted fission barrier heights in this region are relatively-low (of the order of 12 MeV), making them suitable for study using low-energy techniques such as β-delayed fission (maybe [2] and the work at ISOLDE at CERN?) or Coulex-induced fission (maybe [24] and the SOFIA (Studies On FIssion with Aladin) experiment/project/campaign). On the other hand, it has been found that compound nuclei formed in this region from particle-induced reactions tend to have high excitation energies, even for beam energies near the Coulomb barrier. This combination makes the region particularly well-suited for studies involving a variety of excitation energies.

Later experiments performed with isotopes in this region at different excitation energies have shown that, unlike the case of actinides where shell structure and fragment asymmetry is "washed out" at high excitation energies, mass asymmetric fragment distributions are a persistent feature of this mass region for various excitation energies. (A lot of good citations for this section can come from section 4.1.1 of [3]) An up-to-date (as of around 2016) overview of nuclei in the region of ${}^{180}_{80}\mathrm{Hg}_{100}$ which have since been experimentally studied, including the experimental technique used, is shown in Figure 3.1.

3.2 Multimode fission of ^{178}Pt

One particular follow-up experiment was performed investigating spontaneous fission of ${}^{178}_{78}\mathrm{Pt}_{100}$ [?], which differs from ${}^{180}_{80}\mathrm{Hg}_{100}$ by 2 protons. This system was studied at var-

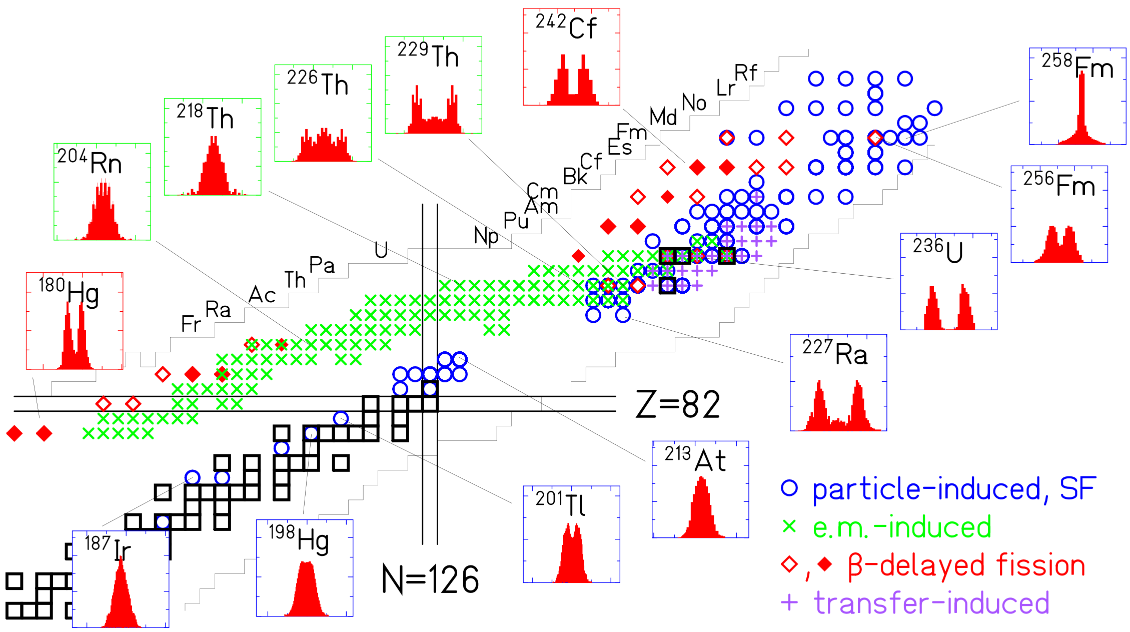

Figure 3.1: Fragment yields for several nuclei ranging from actinides, where primary fission yields tend to be asymmetric, down to near-thorium, where yields become more symmetric except in the region near neutron deficient $^{180}_{80}\text{Hg}_{100}$. Figure from [3].

ious excitation energies and found to fission consistently with a bimodal pattern, as shown in Figure 3.2. Of the nuclei which underwent spontaneous fission, roughly 1/3 were found to fission symmetrically while the other 2/3 fissioned asymmetrically with a light-to-heavy mass ratio of approximately 79/99. Furthermore, it was observed that symmetric fragments tended to have higher kinetic energies than non-symmetric fragments.

To better interpret the results of this experiment, DFT calculations were performed using the functionals UNEDF1$_{\text{HFB}}$ [38] and D1S [10]. These calculations involved computing a PES using the collective coordinates Q_{20} and Q_{30}. [Do I need to describe the calculations in detail here, or should I refer to the published papers?]

The UNEDF1$_{\text{HFB}}$ PES is shown in Figure 3.3, while the D1S PES is in Figure 3.4. A calculation with full Langevin dynamics was not performed; however, the static (minimum-energy) pathway shown in the figure corresponds to a fragment split $A_L/A_H \approx 80/98$.

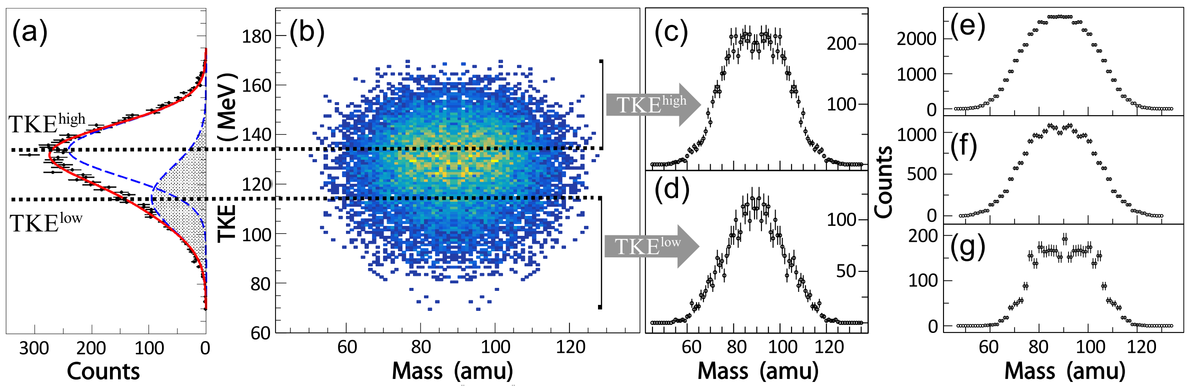

Figure 3.2: This figure contains the data from the $^{178}_{78}Pt_{100}$ experiment. I should go through and describe what all the individual boxes are for. Then I should cite out paper, once it's citeable

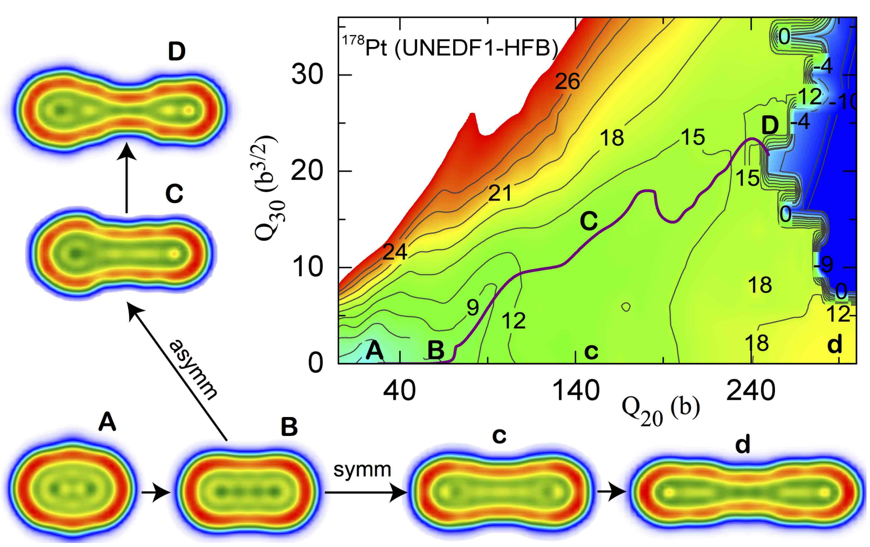

Figure 3.3: UNEDF1-HFB potential energy surface for ^{178}Pt. Note the two different trajectories ABCD and ABcd and their corresponding localizations.

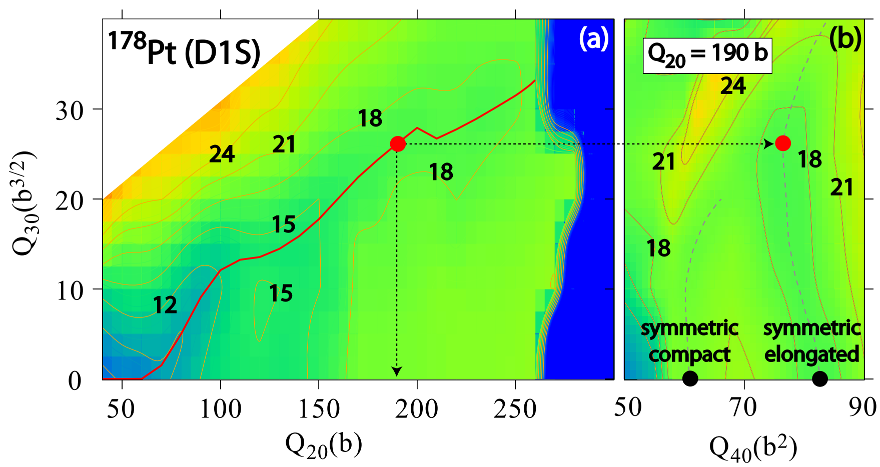

Figure 3.4: D1S potential energy surface for ^{178}Pt. Note also the additional information about the hexadecapole moment.

Also shown in Figure 3.3 are nucleon localization functions (recall Section) corresponding to various configurations in the PES. Along the symmetric path (ABcd in the figure), the fragments appear highly-elongated, with a rather large neck, even shortly before scission. Since elongation tends to minimize the Coulomb repulsion between fragments, then this configuration might be expected to lead to fragments with relatively low kinetic energies. On the other hand, compact fragments such as those in ABCD will tend to have a larger Coulomb repulsion, propelling the fragments away from one another with greater force and resulting in fragments with a higher kinetic energy. [We note that this is compatible with experiment]

Now consider the PES corresponding to the D1S functional in Figure 3.4. We note with some relief that, despite the inherent differences between the functionals, and despite the relative flatness of the surface with few discernible topological features, the overall topology of the PES is similar in both cases. The overall magnitude is different, but the static pathway follows a similar trajectory.

3.3 The physical origin of fragment asymmetry in the region of ^{180}Hg

Why is there a region of symmetric fission below thorium?

(These are notes from the 178Pt paper draft. Not mine, of course, but they have some good points to address): "Namely, the PES are predicetd to be flat and much less structure-less, and defined predominantly by the large liquid drop/macroscopic contribution, rather than by relatively small microscopic effects. Due to this, FFMDs exhibit failrly low dependenece.. [refer to 180Hg PLB, as one example].

"(this was an answer by Witek, when somebody asked a question to my talk at Tsukuba - why the lead region is less senstitive to temperaturte.. the answer was - there is no 'barrier' in a sennce, it's just flat/thick macroscopic surface, hardly influenced bu shell efffects.. so, even if one heats it up, tiny shell effects will be gone, but the main underlying macroscopic part will remain)."

Peter Moller argues in the concluding discussion of (https://link.aps.org/doi/10.1103/PhysRevC.85.0243 [27]) that we can't really use the fragment/prefragment shell structure arguments in this region, and thus that we have yet to identify all the essential physics which determines fragments. He says the yields are given (at least in this case) by subtle interplays in local regions of the potential energy surface.

Witek, Michal Warda, and Staszczak argue in Section IV. *Prescission Configurations* of [43] that 180Hg deforms as a molecular system consisting of 90Zr and 72Ge, with the remaining neck nucleons being distributed at scission to give the fragments they found in the experiment. Similarly, they make the same claim for 198Hg, except using 98Zr and 80Ge. The first one kind of makes sense to me since 90Zr is semi-magic, but 98Zr is not and neither is 80Ge. I wonder what might have happened had they tried to match up the densities of a different set of nearby nuclei (they used these because they had the same N/Z ratio as the fissioning parent nucleus). Then in the conclusions: "We conclude that the mass distribution of fission fragments in both nuclei is governed by shell structure of prescission configurations associated with molecular structures."

In the introduction to [25] it is stated as though conclusively that "the main factor determining the mass split in fission are shell effects at pre-scission configurations, i.e., between saddle and scission" (see also some additional references therein). I think the thing that is most selling it to me so far, though, is Fig. 3 from this paper, wherein they show the shell correction energy for each of the nuclei considered. Even though the PES itself is mostly flat in each of these cases, the magnitude of the shell correction is different whether you are looking at symmetric or asymmetric trajectories, and the one with the larger magnitude shell correction happens to be the one that wins out in the final fragment distribution. I'd also be curious to see what the collective inertia looks like, but this seems to at least give something. It's not like this shell correction gets added on top of the PES - the PES is still relatively-flat - but it at least gives an explanation for why our traditional physical intuition is not totally failing us here.

Interesting future work in this region might include calculations with full dynamics (including from nuclei with excitation energy), as suggested in the conclusions of [25]

Chapter 4

Cluster decay in ^{294}Og

4.1 Introduction

An exciting frontier in nuclear physics is the region of superheavy nuclei ($Z \geq 104$). The latest experiments are able to push the boundaries of the nuclear chart all the way to Z=118, and new ideas are being developed to increase production and improve measurements of superheavy elements [15, 29]. Due to the large number of nucleons, these nuclei push the limits of our nuclear structure models and are expected to highlight new aspects and phenomena of nuclear physics. Spontaneous fission, for example, will likely play an important role in governing the lifetimes of many of these new systems. Fission of superheavy elements may also play an important role in the astrophysical r-process, by placing an endpoint on neutron capture and starting fission cycling (see, e.g., [**?**]).

As these pioneering experimental efforts are made, theory plays a critical role by guiding and interpreting the results of those experiments, as well as by filling in gaps where experiment cannot reach. However, in these exotic regions it is especially important to use only the very best and most reliable predictive models. Recently, a great deal of work has been invested in building self-consistent microscopic models of spontaneous fission which are able to predict, for instance, half-lives and fragment yields [35, 34, 36, 37].

However, this success comes at a cost. In the adiabatic approaches that are often invoked to describe spontaneous fission, fission is described as a tunneling through a potential barrier in a multidimensional space of collective nuclear shape coordinates. Due to the large computational cost associated with calculations (their "inextinguishable thirst for computing power," as stated in [39]), this barrier is approximated using five or fewer shape coordinates in phenomenological microscopic-macroscopic models, and even fewer in mean-field approaches.

Of course, it is well-understood that some physics may be obscured in a limited collective space (see [16]). Thus, one's choice of collective coordinates is dependent on what physics are deemed important or relevant, and which aspects can be safely neglected. In mean-field models, the collective coordinates are typically chosen to be leading order terms in the shape multipole expansion: axial quadrupole moment, triaxial quadrupole moment, and axial octupole moment. Additionally, it was shown in a previous work [34] that pairing correlations have a strong impact on the half-lives calculated via action minimization, and should be taken as a collective coordinate equal in importance to multipole moments or other shape-based collective coordinates.

In the following sections we try to understand the role of the collective space on fission yield predictions. In section **??** we describe the microscopic framework used here to calculate fragment yields, and then in section **??** the model is applied to the superheavy element ^{294}Og, which is the heaviest element ever produced by humans. The paper then concludes with analysis and discussion of the results in section **??**.

4.2 ^{294}Og

Recent efforts to synthesize superheavy elements (SHE) have successfully produced the isotope ^{294}Og, which has been confirmed via its alpha-decay chain. In both experiments, the researchers found evidence of alpha decay, but both also noted the possible observation of decay via spontaneous fission. This suggests the possibility that ^{294}Og might have a similar decay time with respect to both alpha-decay and spontaneous fission.

While some authors [cite] have predicted that fission in the superheavies will proceed as with the actinides (that is, driven by the shell formation of ^{132}Sn in one of the prefragment) our calulations predict that the dominant fission mode will be highly-asymmetric and driven by ^{208}Pb (sometimes referred to in the literature as cluster emission).

There has been an expectation (for some reason?) that cluster emission (known also in the literature as cluster radioactivity, lead radioactivity, cluster decay, heavy-particle radioactivity, ???) might play an important role in the fission of superheavy elements, suggesting that even for such large nuclei (where the Coulomb repulsion is strong), shell structure of the prefragments still drives the determination of the fragments.

[31, 32] - In this paper they propose changing/extending the concept of Heavy Particle Radioactivity or Cluster Radioactivity. Also they apply some model to HPR/CR in SHE.

"A larger number of observed spontaneous fission activities enabled the establishment of a global dependency of spontaneous fission half-lives (T_{SF}) and the fissility of a nucleus, expressed by the ratio Z^2/A which had been realized already by Seaborg [111] and also by Whitehouse and Galbraith [112]. The data, available at that time indicated for even-even nuclei an exponential dependence of the fission half-lives from Z^2/A. From an extrapolation of the trend it was concluded, that a nucleus will become instantaneously unstable against nuclear fission at $Z^2/A \approx 47$, which was set in correspondence with a half-life of $\approx 10-20$ s. Interestingly, the heaviest nucleus reported to be synthesized so far, $^{294}118$ (^{294}Og) [65], has a value $Z^2/A \approx 47.36$. The half-life is given as $T_{1/2} = 0.69 + 0.640.22$ s. Up to now four α decays, but no spontaneous fission was observed [65]." - from [21] - Og is anomolous in that it violates this extrapolated trend (as would, I am sure, most SHE).

Whether or not this PES is able to reasonably describe the CN experiments which so far have produced ^{294}Og is uncertain, because such large compound nucleus expectation energies as appear in experiment may have quite a large effect on the topology of the PES [30]

On the theory side, there have been several attempts to compute spontaneous fission half-lives and alpha-decay half-lives for many superheavy nuclei, and in many cases it is predicted that the two lifetimes will be comparable [31, 32, 48] [Zhang was an application of several universal CR and alpha decay models to the SHE, in order to see if the predictions, too, were universal]. These previous works have tended to rely on phenomenological models which have been tuned to smaller, more stable nuclei. Thus, it is difficult or impossible to assess these models' predictive power in the region of SHE. Thus, a goal of this work is to bring the full predictive framework of self-consistent nuclear density functional theory to bear on the problem of spontaneous fission in the SHE ^{294}Og. This approach is relatively young in the world of nuclear fission models, but it is already producing quality results for a variety of nuclei in different regions of the nuclear chart (see, for instance, [25, 37, 36, **?**]). Some attempts in the region of SHE have already been made, using Skyrme and Gogny functionals in a 2D space [**?**, **?**, 42, 5].

Within these models, spontaneous fission lifetimes tend to be considerably larger than alpha decay lifetimes, ranging from $\frac{\tau_{SF}}{\tau_\alpha} \approx 10^{-10}$ in [5] and [**?**] to $\frac{\tau_{SF}}{\tau_\alpha} \approx 10^{-20}$ in [42].

However, it was shown in [34] that pairing correlations treated as a dynamical variable can have a substantial impact on spontaneous fission lifetimes. That is explored in the case of ^{294}Og here.

This was done in a 4D space consisting of the coordinates $(q_{20}, q_{22}, q_{30}, \lambda_2)$

A criticism that is sometimes leveraged against self-consistent mean-field-based approaches to fission is that, due to the large computational cost associated with calculations, typically only one or two collective coordinates are used. This is in contrast to microscopic-macroscopic methods, where up to five collective coordinates are often used. Those who use SCMF methods assert that the dominant characteristics of the nuclear collective motion necessary for understanding fission can be sufficiently described using perhaps the axial quadrupole moment and maybe one other multipole moment which depends on the specific system, often the axial octupole moment or triaxial quadrupole moment. Of course, it is well-understood that some physics may be obscured in a limited collective space (see [16]). Thus, one's choice of collective coordinates is dependent on what physics are deemed important or relevant, and which aspects can be safely neglected.

However, although various attempts have been made to demonstrate the validity of this assumption, our work represents the first published instance of a 4D potential energy surface calculated self-consistently. Furthermore, given the recent demonstration of the importance of pairing correlations as a collective "coordinate" of the system, ours will feature pairing as part of the collective space, and its impact compared to other collective coordinates will be evaluated.

We used 30 harmonic oscillator shells and 1500 states

4.2.1 Cluster Decay

Experimental instances of super-asymmetric fission: M. G. Itkis 1985, Z Phys A 320 - no assessment of the cause of highly-asymmetric fission, but likely related to 132Sn (there nuclei would tend to fission symmetrically, but with a slight bump around mass A=140-145) D. Rochmann Nucl Phys A 735 (2004) - driven by shell structure of lighter fragments I M Itkis, J Phys Conf Ser 515 (2014) 012008 - cluster radiation by another name

AKA "Lead Radioactivity" sometimes in the literature To predict cluster half-lives, some people take it as a very heavy alpha emission, and others a very asymmetric fission Warda looks at the N/Z ratio of known cluster emitters (or really of lead-208), and then extrapolates it out to SHEs. That's how he decided which superheavies to compute PRC 86 (2012) 014322 Nucl Phys A 944 (2015) 442 (with Baran and others)

4.2.2 Synthesis of Og

They found 3 (and possibly 4) instances in the original Dubna run. Then there was a secondary run at Oak Ridge that was about the same: something like 3 alpha events and a possible fission event. (Another Og paper is being prepared (Nathan Brewer, et al), which has a similar decay chain but a shorter half-life ($\sim$0.185 ms); Detected a 10.6 MeV recoil event, followed 78 microseconds later by a second decay event in the same pixel ($\sim$140 MeV), which is a candidate for SF)

4.2.3 Competition with Alpha Decay

Recent efforts to synthesize superheavy elements (SHE) have successfully produced the isotope ^{294}Og, which has been confirmed via its alpha-decay chain. In both experiments, the researchers found evidence of alpha decay, but both also noted the possible observation of decay via spontaneous fission. This suggests the possibility that ^{294}Og might have a similar decay time with respect to both alpha-decay and spontaneous fission.

There has been an expectation (for some reason?) that cluster emission (known also in the literature as cluster radioactivity, lead radioactivity, cluster decay, heavy-particle radioactivity, ???) might play an important role in the fission of superheavy elements, suggesting that even for such large nuclei (where the Coulomb repulsion is strong), shell structure of the prefragments still drives the determination of the fragments.

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“A larger number of observed spontaneous fission ac- tivities enabled the establishment of a global dependency of spontaneous fission half-lives (TSF) and the fissility of a nucleus, expressed by the ratio Z2/A which had been realized already by Seaborg [111] and also by Whitehouse and Galbraith [112]. The data, available at that time indicated for even-even nuclei an exponential dependence of the fission half-lives from Z2/A. From an extrapolation of the trend it was concluded, that a nucleus will become instantaneously unstable against nuclear fission at Z2/A 47, which was set in correspondence with a half-life of 10−20 s. Interestingly, the heaviest nucleus reported to be synthesized so far, 294118 (294Og) [65], has a value Z2/A 47.36. The half-life is given as T1/2 =0.69+0.64−0.22 s. Up to now four decays, but no spontaneous fission was observed [65].” - from [21] - Og is anomolous in that it violates this extrapolated trend (as would, I am sure, most SHE).

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[30]

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4.3 Method

Our calculations were performed within the framework of nuclear density functional theory using Skyrme and Gogny energy density functionals. In the Skyrme case, the parameterization UNEDF1-HFB [38] was used, and pairing correlations were described using a density dependent pairing interaction. To assure convergence despite the high density of states, the DFT solver HFODD was used with 30 harmonic oscillator shells and 1500 states allowed in the calculation. Calculations were performed in a 4D collective space consisting of 3 shape coordinates, (q_{20}, q_{30}, q_{22}), and, given the importance of dynamic pairing fluctuations demonstrated in [34], λ_2. To demonstrate model independence, another set of calculations was performed using the Gogny energy density functional D1M in the two-dimensional collective space described by coordinates (q_{20}, q_{30}).

It is seen in many models that introducing triaxiality as a degree of freedom can often be energetically-favorable, sometimes lowering saddle points by as much as 3 MeV; however, dynamic calculations in which the collective inertia is considered together with the potential energy surface have found that dynamical pathways usually tend to tunnel through barriers rather than break axial symmetry. This competition was explored for SHE in [18], with the conclusion that triaxiality plays a fairly insignificant role in determining the half-life of elements below $Z = 120$. However, another recent paper (https://arxiv.org/abs/1803.04616v2) suggests that triaxiality might significantly lower the second barrier. Regardless, we included q_{22} in our calculations. It may also be the case that isotopes which are oblate-deformed in their ground state may pass through triaxial configurations on their way to greater elongations.

The basis of the model is the assumption that spontaneous fission can be treated such that the lifetime is proportional to e^{-P}, where P is the transmission probability through some barrier.

The collective inertia of the system was computed using the nonperturbative ATDHFB cranking approximation in the Skyrme case, and perturbative ATDHFB with cranking and perturbative GCM with cranking in the Gogny case [7]. The tunneling is described using the WKB approximation, in which the tunneling path $L(s)$ was computed by using the dynamic programming method to minimize the collective action

$$S(L) = \int_{s_{in}}^{s_{out}} \frac{1}{\hbar} \sqrt{2\mathcal{M}_{eff}\left(V_{eff}(s) - E_0\right)} ds \tag{4.1}$$

where $\mathcal{M}_{eff}$ is the effective inertia and V_{eff} the effective potential energy along $L(s)$. Following the formalism of [35], the half-life is computed via $T_{\frac{1}{2}} = \ln 2/nP$, where $n = 10^{20.38} s^{-1}$ is the number of assaults on the fission barrier per unit time and the penetration probability P is given by

$$P = (1 + exp[2S(L)])^{-1} \tag{4.2}$$

Finally, after computing the action at many points along the outer turning line, the final fragment yields were determined by evolving the system many times via Langevin dynamics, following the work done in [36].

4.4 Langevin dynamics

4.5 Fragments and the Nucleon Localization Function

An improved scission criterion would go beyond simply counting the number of particles in the neck. To help with this, we have a tool at our disposal which helps us to understand correlations that affect fission dynamics. This is called the nucleon localization function, and it allows us to visualize the prefragment nuclear shell structure which largely determines the identity of fission fragments [47].

The nucleon localization function shows that some prefragments can be very well-formed even when the neck is large, while in another case the neck might be small but the prefragments, poorly-defined [37]. A better scission criterion should take into account, or at least be compatible with, the insights gained from the nucleon localization function. As noted in [45], fragment properties on either side of the scission line may differ drastically. This is because shell structure is not well-described geometrically. Our localization measure offers an alternative scheme for identifying fragments before the scission line (see [37]). Since it is based on the underlying quantum shells, it is less sensitive to fluctuations and particle rearrangements late in the evolution.

Chapter 5

R-process

I'll say some stuff about r-process nuclei here

Fission inputs to r-process network calculations

You should definitely cite the 2015 paper by Eichler et al (doi:10.1088/0004-637X/808/1/30), titled "THE ROLE OF FISSION IN NEUTRON STAR MERGERS AND ITS IMPACT ON THE r-PROCESS PEAKS". The region of largest variation (in their conclusions, at least) is in the region A=100-160, which makes sense because that's where most of your fission fragments are likely to lie. This is roughly-speaking also the region of that rare-earth peak everyone always talks about, along with the so-called second r-process peak.

Kilonova - the bright burst of gamma rays and other radiation that accompanies a compact object merger, presumably created mainly from the radioactive decay of unstable r-process nuclei (mainly via alpha and beta decay, but not fission - according to `https://www.nndc.bnl.gov/` see also the paper [49])

Light curves - show the magnitude/intensity of light/EM radiation as a function of time. For example, the light curve from the sun on the earth will be roughly sinusoidal; from an eclipse, you'll have a roughly straight line, then a dip, then a return to the original straight line; a pulsar will also be something regular and periodic. Each type of nova/supernova/kilonova/etc. has its own characteristic light curve, with an initial peak of varying sharpness, and then a gradual decay (though how gradual depends on the characteristics of the event).

Heating - Is exactly what it sounds like. When a nucleus decays, it loses energy. Some of that energy escapes in the form of neutrinos or photons, while other energy is absorbed elsewhere in the medium. A related concept is opacity. Heavier nuclei with a large level density tend to be more opaque because they can more readily absorb photons than smaller, less opaque nuclei. And the process by which all of this energy exchange takes place is called thermalization.

The idea behind this $^{254}_{98}\mathrm{Cf}_{156}$ calculation is related to [49]: There is some speculation that the heating from the NSM might be strongly-impacted by spontaneous fission of $^{254}_{98}\mathrm{Cf}_{156}$. A related idea that $^{254}_{98}\mathrm{Cf}_{156}$ may have been a large contributor to supernova (not kilonova) lightcurves dates back to 1956 (see references in Zhu's introduction), but perhaps fell out of favor once it was discovered that the decay chain of 56Ni was the primary contributor.

Mass and kinetic energy distributions of $^{254}_{98}\mathrm{Cf}_{156}$ were actually measured in [13]. For some reason, though, Zhu considers that measurement "sparse" and they do some dressing up of it in their paper.

Since $^{254}_{98}\mathrm{Cf}_{156}$ is heavier than actinides, detecting its presence in kilonovae would help solidify the location of r process nucleosynthesis. And if it has as large an impact on the heating and light curves as [49] says, it might well make the difference between "hard-to-observe" and "somewhat easier to observe."

So what was it about the fission of $^{254}_{98}\mathrm{Cf}_{156}$ that [49] needed in order to make this argument? And how did they zero-in on $^{254}_{98}\mathrm{Cf}_{156}$ to begin with? These are good questions that nobody knows the answer to. Nah, I'm just kidding. The answer is out there. I just need to find it. It is one of a handful of isotopes which *could* be produced in significant amounts during an r process scenario and which is known to undergo spontaneous fission, along with other californium and fermium isotopes. Furthermore, its half-life on the order of several days means that it could potentially make a significant impact on heating, especially at later times. The group of nuclei matching these criteria include ${}^{254}_{98}\mathrm{Cf}_{156}$, ${}^{257}\mathrm{Es}$, and ${}^{260}\mathrm{Md}$. Finally, according to the mass model and branching ratios they used in [49], the other two nuclei seem like maybe they're more likely to β-decay, so ${}^{254}_{98}\mathrm{Cf}_{156}$ it is. I talked to Samuel, and he says that everybody else predicts ${}^{254}_{98}\mathrm{Cf}_{156}$, too.

Will you not also want/need to mention your work on 290Fm? This ${}^{254}_{98}\mathrm{Cf}_{156}$ is not all that neutron rich, nor is it far from the region most-commonly studied. So perhaps you should say and do a bit more work on Fermium. Even just to say that triaxiality is not important here is something and not nothing. The reason you began to study this particular nucleus, though, is that based on Trevor Sprouse's r process network calculations (which utilized Peter Moller's fission calculations) and depending on the specifics of the astrophysical conditions, fission terminates the r process in a region of the N-Z plane. 290Fm falls into the range identified in one of these calculations (it would be good to cite it) and was selected by Trevor as one which may be particularly significant. However, looking back over my emails, it seems like this is not a robust finding. Other predictions (like the ones I have from Samuel and Marius Eichler) seem to perhaps indicate that I'd be better off looking in the region for which 280Fm is the northeast corner, perhaps.

Chapter 6

Outlook

6.1 Insights gained

6.1.1 Prefragment shell stsructure

A common theme in all of this has been the importance of the underlying shell structure of the prefragments. Shell energy corrections were found to be important in $^{178}_{78}\mathrm{Pt}_{100}$ and $^{180}_{80}\mathrm{Hg}_{100}$; cluster formation in $^{294}_{118}\mathrm{Og}_{176}$ was clearly influenced by the shell structure of the fragments; and the same may or may not be the case for $^{254}_{98}\mathrm{Cf}_{156}$. Let's discuss this.

We used localizations to visualize the internal/intrinsic shell structure inside nuclei, and we were able to see that this structure was sometimes intact early in the evolution, at times as far back as the outer turning line. And actually, this kind of makes sense. From just energetics alone, a nucleus on the outer turning line is just as happy (or just as stable, or just as settled) as a nucleus in the ground state. In some sense, it is formed. The difference now is just that the configuration it's in is now unstable due to Coulomb. The two halves, which are kind of maybe happy from a nuclear physics perspective, are pushing apart from the Coulomb repulsion. So that still has to be carried out, but the bulk of the physics might already be done at this point - though not necessarily. It *could* be that the fragments are well-formed and just pushing apart, but that may not be the case. It's like a divorce: sometimes the two have drifted so far apart, or are so well-defined and incompatible as individuals that the divorce is simple and relatively straightforward. Other times, it is a mess trying to sort out who gets what, and the two parties are fundamentally-changed by the proceedings.

I don't have any strong objections to Scamps and Simenel's octupole paper. In fact, to me it kind of makes sense: we've been saying, after all, that it's the shell structure of the deformed prefragments which determine scission, and not necessarily the final fragments themselves. That's really the whole idea behind the localization paper: we're seeing that, at least in some cases, the shell structure is pretty well intact early in the evolution, and that those prefragments drive the system to scission with some shuffling of the neck nucleons at scission. All they're saying is that those neck nucleons will effect the shell structure of the prefragments, and just based on the kinds of shapes that the system will take (small neck connecting two elongated or spherical fragments), the prefragments have a strong octupole moment (regardless of whether the fragments are elongated or spherical). So it shouldn't be the spherical magic numbers we worry about, but the deformed (in this case, octupole-deformed) magic numbers.

I feel like it shouldn't be too terrible to investigate this claim. What if we constrained the multipole moment(s) that correspond(s) to octupole-deformed fragments (perhaps Q_{50})? I think this parameter might be included in Peter Moller's model, but not in ours.

6.2 Review, outlook, and perspectives

In this chapter, it would be great to talk to everyone you know (Witek, Samuel, Jhilam, Nicolas, Michal, and so on) to get a better feel for what kinds of issues need to be addressed next. You've already got sort of a rudimentary understanding (see your Google Keep note for starters), but it might be good to get some outsider perspective. This will be especially important as you start looking for postdocs, and *especially* especially if you end up looking for postdocs in nuclear theory, but not necessarily nuclear fission.

As I said in chapter 1, “Finally, in chapter 6 we discuss the current state of the field, and, based on our experience, offer insights for guiding future developments in the field.”

At this stage, we have techniques to calculate half-lives and primary fragment distributions (I haven’t mentioned it yet, but there is also Nicolas’ method for the fragment yields that uses TD-GCM. Are there others? What about for half-lives?). Some methods (such as Walid’s, TDDFT, and possibly also this GCM method) are starting to estimate fragment energetics (kinetic and excitation energies). Down the line, there are others who try to predict neutron multiplicities and goodness knows what else using Hauser-Feshbach models and such (FREYA and more). These regions are still disconnected. Of course, these methods still need major refinements in order to better reflect experimental data. Some ideas currently in the pipeline for improving the models are:

- Improved EDFs (here you could mention the DME EDFs)
- Improved inertia tensor (such as automatic differentiation)
- Better/more collective coordinates (Walid’s D, ξ coordinates or whatever they were called [Technical Report LLNL-TR-586678 (2012) Fragment yields calculated in a time-dependent microscopic theory of fission]; continuity of the PES in ∞-dimensional space such like in David Regnier’s talks and papers)
- Fragment identification (our localization paper, Marc Verriere’s method; you might also mention that this is not an issue in TDDFT, but there you’ve only got one single fragment pair)

- Microscopic/self-consistent description for dissipation. This is the mechanism which exchanges between intrinsic and collective degrees of freedom, but we handle it in a very ad hoc way with parameters which are fitted instead of determined systematically through some theory. Solving this problem will probably help us with the energetics of fragments (TKE and E* at the same time!)

Furthermore, there are more experimental observables that we should try to predict (refer to Andreyev's review to see what other observables can currently be measured). These include energetics (TKE and E*, for we have only begun to scratch the surface here), angular momentum, prompt neutron multiplicities (is that within the scope of these self-consistent models?), prompt neutron and gamma energy spectra spectra (getting harder; these are usually handled via statistical models; see intro to [**?**] for some references), level densities?, and probably more but my mind is blanking. How to compute these in a self-consistent framework is still an open question. See also the outlook in Nicolas' review.

We definitely need a better handle on the inertia. The perturbative inertia is easy to compute, but not terribly reliable. The non-perturbative inertia can certainly do better, but as it is computed now (using finite differences) it is subject to numerical artifacts and instabilities (dependent on the level of convergence of the individual densities, the coefficient mutlipliers, different basis sizes) and actual physics, such as level crossings which manifest in projections from a higher-dimensional space.

UNEDF1 seems to underestimate fission barrier heights (artificial though the concept may be; the main impact is probably that lifetimes are underestimated). It also turns out to be a headache to work with, making convergence quite a challenge sometimes (any cases in particular, like for highly-deformed or heavy or octupole-deformed nuclei or something?).

Better functionals might hope to better capture the physics, and one can hope they are easier to work with.

APPENDIX

Appendix

Temperature-Dependent ATDHFB Collective Inertia

.1

Everything which was shown in this dissertation assumed that the system was maintained at temperature $T = 0$ and the nucleus behaved as a superfluid below the Fermi surface. However, in many environments (such as a neutron star merger or a nuclear blast) there may be quite a bit of excitation energy imparted to the system, which would raise the temperature above the Fermi surface. In this case, pairs may be broken and the topology of the potential energy surface may change (see, for instance, [25]). In this case, the collective inertia of the system is changed, too, as shown below.

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