Calculations of Mass Distributions using the Balian-Vénéroni Variational Approach

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Abstract

Existing mean-field models, namely the Hartree-Fock (HF) and time-dependent Hartree-Fock (TDHF) approaches, can be used to determine the expectation values for one-body observables, such as fragment mass, in nuclear reactions and decays but are known to underestimate the fluctuations in these values. This is due to their assumption that each nucleon moves independently in a mean-field generated by the interactions between the nucleons neglecting important two-body correlations. Balian and Vénéroni considered the variational determination of expectation values and fluctuations and obtained an improved formula for these fluctuations which can be implemented using existing TDHF codes. This approach has previously been implemented in a small number of test cases but symmetries and simplified interactions were used due to computational limitations.

In this work we first review the Balian-Vénéroni approach. We then present calculations of the mass distributions for the decay of giant resonances in $^{32}$S, $^{40}$Ca and $^{132}$Sn and in deep-inelastic and fusion-evaporation reactions for $^{16}$O+$^{16}$O and $^{40}$Ca+$^{40}$Ca using a three-dimensional TDHF code with the full Skyrme interaction comparing with the previous calculations and/or experimental data as appropriate. We find that the Balian-Vénéroni approach consistently produces fluctuations that exceed the TDHF values but that the numerical problems inherent in running prolonged TDHF calculations, particularly due to emitted nucleons being reflected back from the boundaries of our spatial box, cause significant numerical difficulties for longer nuclear processes. We are consistently able to obtain converged results for giant resonance calculations but encounter difficulties for the deep-inelastic scattering reactions and are unable to obtain reliable results for the fusion-evaporation reactions. Our results differ from those obtained previously. We discuss the sources of these discrepancies.
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## Contents

**Abstract**  
i
**Acknowledgements**  
ii
**Table of Contents**  
iii
**List of Figures**  
v
**List of Tables**  
vi

### 1 Introduction
1.1 Mass Distributions  
1
1.2 Giant Resonances  
1
1.3 Overview of the HF and TDHF Approaches  
3
1.4 The Balian-Vénérioni Variational Method  
5
1.5 Previous Applications of the Balian-Vénérioni Approach  
7
1.6 Related Works  
9

### 2 Theory
2.1 Mass Distributions  
13
2.2 Expectation Values and Fluctuations in HF and TDHF  
13
2.3 The Balian-Vénérioni Approach  
15
  2.3.1 The Balian-Vénérioni Approach for Single-Particle Observables  
15
  2.3.2 Fluctuations and Correlations for Single-Particle Observables  
17
  2.3.3 Equations of Motion for a State and an Observable  
18
  2.3.4 The Characteristic Function  
20
  2.3.5 Optimised Equations of Motion for Single-Particle Observables  
21
  2.3.6 Equations of motion for \( m'(t), M'(t), V'(t) \) and \( L'(t) \)  
30
  2.3.7 Equations of Motion for \( p'(t) \) and \( \sigma'(t) \)  
30
  2.3.8 Expansion in terms of \( \xi' \)  
32
  2.3.9 Expectation Values for Single-Particle Operators  
33
  2.3.10 Correlations and Fluctuations for Single-Particle Operators  
34
  2.3.11 Features of this Result  
40
  2.4 The Balian-Vénérioni Approach applied to Mass Fluctuations  
40

### 3 Codes and Procedures
3.1 The OAK3D HF/TDHF Code  
42
  3.1.1 Implementation of the HF Approach within the OAK3D Code  
42
  3.1.2 Implementation of the TDHF Approach within the OAK3D Code  
43
  3.1.3 Interactions  
44
  3.1.4 Excitations of the System for Dynamic Calculations  
48
  3.2 Implementation of the Balian-Vénérioni Formula  
49

### 4 Code Validation: GDR in \(^{32}\text{S}\)
4.1 Hartree-Fock Calculation for \(^{32}\text{S}\)  
53
4.2 TDHF Calculation for a GDR in \(^{32}\text{S}\)  
54
4.3 Application of the BV approach to a GDR in \(^{32}\text{S}\)  
58
  4.3.1 Time Reversal in the OAK3D TDHF Code  
58
4.3.2 Results of the Balian-Vénéroni Approach for a $^{32}$S GDR .................... 61
4.3.3 Dependence on $R_c$ ............................. 64
4.3.4 The Effect of the Sharp Cutoff at $r = R_c$ ............................................ 66
4.3.5 Dependence on the size of the spatial box ........................................... 67
4.3.6 Dependence on $\Delta r$ ........................................ 67
4.3.7 Dependence on $t_1$ ........................................ 69
4.3.8 Dependence on $\Delta \tau$ ........................................ 69
4.3.9 Dependence on the Skyrme Parameterisation ................................. 71
4.4 Summary ........................................ 73

5 Resonance Calculations ................................. 74
5.1 Isoscalar Giant Monopole Resonance in $^{40}$Ca ................................. 74
5.2 Isovector Giant Dipole Resonance in $^{132}$Sn ........................................ 79

6 Collision Calculations ........................................ 84
6.1 $^{16}$O+$^{16}$O Collision ($E_{LAB} = 160$ MeV) ........................................ 84
   6.1.1 Time-dependent calculations for $b = 0.0$ fm ($\hbar \approx 0$) .................... 86
   6.1.2 Time-dependent calculations for $b = 2.7$ fm ($\hbar \approx 15$) .................... 89
   6.1.3 Time-dependent calculations for $b = 5.4$ fm ($\hbar \approx 30$) .................... 92
   6.1.4 Time-dependent calculations for $b = 8.1$ fm ($\hbar \approx 45$) .................... 92
   6.1.5 Discussion ........................................ 96
6.2 $^{40}$Ca+$^{40}$Ca Collisions ($E_{LAB} = 278$ MeV) .................................... 97

7 Conclusions and Future Work ........................................ 103

A The HF Equations ........................................ 105
A.1 The Hartree-Fock ground state energy ........................................ 106
A.2 The Variation of the Energy ........................................ 107

B The TDHF Equations ........................................ 110

C Associated Publications ........................................ 112
C.1 Papers ........................................ 112
C.2 Conference Proceedings ........................................ 112

Bibliography ........................................ 113
# List of Figures

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>A schematic view of giant resonances</td>
<td>2</td>
</tr>
<tr>
<td>1.2</td>
<td>Previous calculation of ΔNBV for 40Ca GMR by Troudet and Vautherin</td>
<td>8</td>
</tr>
<tr>
<td>1.3</td>
<td>Charge distributions for the deep-inelastic reaction 40Ca+40Ca (ELAB = 256)</td>
<td>9</td>
</tr>
<tr>
<td>4.1</td>
<td>The convergence of the HF calculation for 32S (SLy6)</td>
<td>55</td>
</tr>
<tr>
<td>4.2</td>
<td>⟨N⟩ and ΔNTDHF for the 32S GDR, plotted as a function of time</td>
<td>57</td>
</tr>
<tr>
<td>4.3</td>
<td>Dipole moments calculated during the decay of a GDR in 32S</td>
<td>58</td>
</tr>
<tr>
<td>4.4</td>
<td>Normalisation factors for φ and ψ to test the reversibility of the code</td>
<td>60</td>
</tr>
<tr>
<td>4.5</td>
<td>ΔNBV(ε) plotted as a function of ε and extrapolated back to ε = 0</td>
<td>61</td>
</tr>
<tr>
<td>4.6</td>
<td>ΔNBV(ε) plotted as a function of ε</td>
<td>62</td>
</tr>
<tr>
<td>4.7</td>
<td>Real and imaginary parts of the overlaps between ϕ(t₀) and ψ(t₀, ε = 0.0)</td>
<td>63</td>
</tr>
<tr>
<td>4.8</td>
<td>The effect of varying Rₜ on the results for the 32S GDR</td>
<td>65</td>
</tr>
<tr>
<td>4.9</td>
<td>A schematic view of the radial dependence of χ₁ and χ₂</td>
<td>66</td>
</tr>
<tr>
<td>4.10</td>
<td>⟨N⟩ plotted as a function of time and the size of the spatial box</td>
<td>68</td>
</tr>
<tr>
<td>4.11</td>
<td>⟨N⟩, ΔNTDHF and ΔNBV versus t₁ for a 32S GDR</td>
<td>70</td>
</tr>
<tr>
<td>5.1</td>
<td>The convergence of the HF calculation for 40Ca (SLy6)</td>
<td>76</td>
</tr>
<tr>
<td>5.2</td>
<td>⟨N⟩ and rms radii plotted against time during the decay of a GMR in 40Ca</td>
<td>77</td>
</tr>
<tr>
<td>5.3</td>
<td>The convergence of the HF calculation for 132Sn (SLy6)</td>
<td>80</td>
</tr>
<tr>
<td>5.4</td>
<td>The number of nucleons in the 132Sn GDR, plotted as a function of time</td>
<td>80</td>
</tr>
<tr>
<td>5.5</td>
<td>Dipole moments, Qₓ, Qᵧ and Qᶻ for a GDR in 132Sn</td>
<td>81</td>
</tr>
<tr>
<td>5.6</td>
<td>(ΔNBV(ε))² versus ε for 132Sn GDR at t₁ = 220 fm/c (SLy4)</td>
<td>83</td>
</tr>
<tr>
<td>6.1</td>
<td>The convergence of the HF calculation for 16O (SLy6)</td>
<td>85</td>
</tr>
<tr>
<td>6.2</td>
<td>Density contour plots for 16O-16O (head-on)</td>
<td>87</td>
</tr>
<tr>
<td>6.3</td>
<td>Energy in the COM Frame and fragment separation for 16O+16O at b = 0.0 fm</td>
<td>88</td>
</tr>
<tr>
<td>6.4</td>
<td>Density contour plots for 16O-16O (b = 2.7 fm, l ≈ 15ℏ)</td>
<td>90</td>
</tr>
<tr>
<td>6.5</td>
<td>Expectation values, fluctuations and rms radii versus t for (b = 2.7 fm)</td>
<td>91</td>
</tr>
<tr>
<td>6.6</td>
<td>Density contour plots for 16O-16O (b = 5.4 fm, l ≈ 30ℏ)</td>
<td>93</td>
</tr>
<tr>
<td>6.7</td>
<td>Expectation values, fluctuations and rms radii versus t for (b = 5.4 fm)</td>
<td>94</td>
</tr>
<tr>
<td>6.8</td>
<td>Density contour plots for 16O-16O (b = 8.1 fm, l ≈ 45ℏ)</td>
<td>95</td>
</tr>
<tr>
<td>6.9</td>
<td>Energy in the COM Frame and fragment separation for 16O+16O at b = 8.1 fm</td>
<td>96</td>
</tr>
<tr>
<td>6.10</td>
<td>Density contour plots for 40Ca-40Ca (b = 0 fm)</td>
<td>98</td>
</tr>
<tr>
<td>6.11</td>
<td>Energy in the COM Frame and fragment separation for 40Ca+40Ca at b = 2.6 fm</td>
<td>99</td>
</tr>
<tr>
<td>6.12</td>
<td>Density contour plots for 40Ca-40Ca (b = 2.6 fm)</td>
<td>101</td>
</tr>
<tr>
<td>6.13</td>
<td>Expectation values, fluctuations and RMS Radii versus t for (b = 2.6 fm)</td>
<td>102</td>
</tr>
</tbody>
</table>
## List of Tables

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>Summary of previous calculations of mass distributions using the BV approach</td>
</tr>
<tr>
<td>4.1</td>
<td>Properties of a $^{32}$S nucleus calculated with different Skyrme parameterisations</td>
</tr>
<tr>
<td>4.2</td>
<td>Parameters for the SLy4, SLy4d and SLy6 Skyrme parameterisations</td>
</tr>
<tr>
<td>4.3</td>
<td>Properties of a $^{32}$S nucleus calculated using the OAK3D HF code</td>
</tr>
<tr>
<td>4.4</td>
<td>Comparison of $\langle N \rangle$ and $\Delta N_{TDHF}$ to check the reversibility of the code</td>
</tr>
<tr>
<td>4.5</td>
<td>The dependence of $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ on $R_c$ for the $^{32}$S GDR</td>
</tr>
<tr>
<td>4.6</td>
<td>The dependence of $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ on the cutoff function</td>
</tr>
<tr>
<td>4.7</td>
<td>Properties of a $^{32}$S nucleus calculated for different values of $\Delta r$</td>
</tr>
<tr>
<td>4.8</td>
<td>Comparison of $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ for different Skyrme Parameterisations</td>
</tr>
<tr>
<td>5.1</td>
<td>Properties of a $^{40}$Ca nucleus calculated with different Skyrme parameterisations</td>
</tr>
<tr>
<td>5.2</td>
<td>Comparison of new and old results for $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ for a $^{40}$Ca GMR</td>
</tr>
<tr>
<td>5.3</td>
<td>Properties of a $^{132}$Sn nucleus calculated with different Skyrme parameterisations</td>
</tr>
<tr>
<td>5.4</td>
<td>Comparison $\Delta N_{TDHF}$ and $\Delta N_{BV}$ for a $^{132}$Sn GDR</td>
</tr>
<tr>
<td>6.1</td>
<td>$\langle N \rangle$ and $\Delta N$ for the $^{16}$O+$^{16}$O collisions at $E_{LAB} = 80$ MeV with $b = 0.0$ fm</td>
</tr>
<tr>
<td>6.2</td>
<td>$\langle N \rangle$ and $\Delta N$ for the $^{16}$O+$^{16}$O collisions at $E_{CM} = 80$ MeV with 8.1 fm</td>
</tr>
<tr>
<td>6.3</td>
<td>BV results for $^{40}$Ca+$^{40}$Ca collisions at $E_{LAB} = 278$ MeV</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

The atomic nucleus is a highly complex many-body system in which the behaviour of the individual constituents are governed by complicated many-body interactions. In any nuclear reaction or process there are a range of possible outcomes including, for the purposes of the current work, a range of possible final masses for the nuclei involved. A typical scattering reaction involving a projectile and target nucleus may lead to the break-up of one of the nuclei, the knock-out of nucleons from one of them or the transfer of nucleons between them or combinations thereof. The resulting nuclei may end up in an excited state the decay of which may involve the emission of nucleons. The nuclei may also fuse to form a hot compound nucleus which may subsequently fission or could itself decay through particle emission (a fusion-evaporation reaction).

1.1 Mass Distributions

Writing the mass of the fragment of interest as the expectation value of the operator $\hat{N}$, $\langle N \rangle$, we consider the standard deviation in the mass, $\Delta N$, given by

$$\langle \Delta N \rangle^2 = \langle (N)^2 \rangle - \langle N \rangle^2.$$  \hspace{1cm} (1.1)

In this work we perform calculations to determine the ranges of possible masses for the surviving nuclei/fragments following fusion-evaporation or deep-inelastic reactions. We also consider the case of giant resonances decaying by particle emission.

1.2 Giant Resonances

Giant resonances are the collective oscillations of the protons and neutrons in the nucleus discovered in the late 1940's [1] and now known to be a common feature in nuclei [2,3]. The decay, by particle emission, of a giant resonance is a simpler problem than a collision between two nuclei with a smaller range of possible outcomes and as such makes a good test case (although giant resonances are useful in their own right through providing information about the properties of nuclear matter.
1.2 Giant Resonances

Giant resonances decay through γ-ray or particle emission [4, 5]. Giant resonances are classified by the motion of the protons and neutrons. In this work we will consider giant monopole resonances (GMR) and giant dipole resonances (GDR). In a GMR the proton and neutron distributions in the nucleus oscillate radially either in phase (an isoscalar GMR, ISGMR) or out of phase (an isovector GMR, IVGMR), this is sometimes known as the breathing mode [6, 7]. In a GDR the centres-of-mass of the protons and neutrons oscillate about the centre-of-mass frame of the nucleus. These are all shown schematically in figure 1.1. This figure shows the motion of the protons and neutrons in the centre-of-mass of the nucleus and does not include an isoscalar GDR since in that case the protons and neutrons would be oscillating in phase which would be equivalent to the entire nucleus oscillating in space.

We will simulate these dynamic nuclear processes using self-consistent mean-field approaches, specifically the Hartree-Fock (HF) approach and its time-dependent extension, the time-dependent Hartree-Fock (TDHF) approach. These are microscopic models in that they explicitly consider all the components of the system and not just the bulk properties. In nuclear reactions the complex many-body nature of the processes and the number of possible decay paths makes it impossible to know in advance all of the relevant observables and reaction channels. Microscopic approaches are well suited to dealing with this sort of open-ended problem.

Figure 1.1: A schematic view of the collective oscillations in nuclei (adapted from [8]).
1.3 Overview of the HF and TDHF Approaches

The HF approach allows one to calculate the most energetically favourable (i.e. the lowest energy) states of a nucleus (although excited states can be considered if appropriate modifications are made to the method) whilst the TDHF approach is an extension of this providing an equation of motion which tells us how a particular state of the system will evolve in time, usually in response to an applied force. Although these approaches are now well developed and have been successfully used to make predictions for a range of observables there remain classes of observables, which include mass distributions, for which they are not able to produce accurate predictions.

Whilst it will be necessary re-visit this in more detail later it is useful to briefly introduce the essential ideas of the HF and TDHF approaches as well as their strengths and weaknesses so that we may refer to these later. These approaches were developed for modeling complex many body systems and processes where the number of interacting bodies and their complex interactions prevents the problem from being solved exactly.

The central assumption of these approaches is that each nucleon moves independently of the others in an average potential (the mean field), $H_{HF}$, generated by the interactions between them as described by some nucleon-nucleon force. Since the state of the system is determined from this mean-field, which itself depends upon the state of the system, this is a self-consistent approach requiring, in general, an iterative solution. The HF approach was originally used for studying electronic systems where the electronic interactions are well known [9] before being applied to nuclear systems where the more complex nucleon-nucleon interaction is not so well understood [10–13]. This means that we must use so-called effective interactions, the non-local Gogny force [14] or the local (i.e. zero-range) Skyrme force [15,16], whose functional forms were obtained by combining experimental observations and theory and which include a number of free parameters which are fitted to reproduce available experimental data.

The assumption of independent motion allows the many-body wavefunction of an $A$-nucleon system, $\Phi$, to be approximated to an anti-symmetrised (to satisfy the Pauli exclusion principle) product of $A$ single-particle wavefunctions, $\phi_k$, a Slater determinant [17,18]

$$\Phi(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_A) \rightarrow \Phi_{HF}(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_A) = \frac{1}{\sqrt{A!}} \begin{vmatrix} \phi_1 (\vec{r}_1) & \phi_2 (\vec{r}_1) & \ldots & \phi_A (\vec{r}_1) \\ \phi_1 (\vec{r}_2) & \phi_2 (\vec{r}_2) & \ldots & \phi_A (\vec{r}_2) \\ \vdots & \vdots & \ddots & \vdots \\ \phi_1 (\vec{r}_A) & \phi_2 (\vec{r}_A) & \ldots & \phi_A (\vec{r}_A) \end{vmatrix}.$$  \hspace{1cm} (1.2)

The vector $\vec{r}_i$ denotes both the spatial co-ordinates as well as the spin and the isospin co-ordinates (i.e. $\vec{r} \rightarrow (\vec{r}, \sigma, \tau)$). The numerical subscripts denote the relevant spatial, spin and isospin quantum numbers for the single particle states. In the TDHF approach these states all become
1.3 Overview of the HF and TDHF Approaches

time-dependent, $\phi_k (r_i) \rightarrow \phi_k (r_i, t)$.

It is common, and often notationally simpler, to work in terms of the one-body density matrix, $\rho$, satisfying $\rho^2 = \rho$. For a system described by the wavefunction $|\Phi (t)\rangle$ the elements of the single-particle density matrix are given by

$$\rho_{\mu \nu} (t) = \langle \Phi (t) | \hat{c}_\mu^\dagger \hat{c}_\nu | \Phi (t) \rangle , \quad (1.3)$$

where $\hat{c}_\mu^\dagger$ and $\hat{c}_\nu$ are creation and annihilation operators which create or destroy a nucleon in the single-particle states $|\phi_\mu (t)\rangle$ and $|\phi_\nu (t)\rangle$ respectively. In the HF approach the Slater determinant describing the state of the system is determined using a variational method in which the single-particle wavefunctions are varied to minimise the energy in the system (for a general review of variational techniques see [19]).

The TDHF approach is an extension of the HF approach which allows dynamical processes to be simulated through the use of an equation of motion describing how a system, starting from a known initial state, evolves with time [20]

$$i\hbar \frac{d\rho}{dt} = \left[ \hat{H}_{HF} (\rho) , \rho \right] , \quad (1.4)$$

where $\hat{H}_{HF} (\rho)$ is the HF Hamiltonian (see appendix A and specifically (A.24)) and depends, through $\rho$, upon the state of the system.

The HF and TDHF approaches are extremely successful methods which can be applied to any nucleus in the nuclear chart and to a wide range of dynamic situations (as discussed in an early review of these techniques [21]). However, it soon became apparent that the central assumption of the method, that the nucleons move independently of one another, whilst essential for reducing the complexity of the problem and making it tractable, means that important two-body effects are neglected.

This problem was first observed by Koonin et al. [22] who performed axially symmetric TDHF calculations of $^{16}\text{O}+^{16}\text{O}$ ($E_{LAB} = 192$ MeV) and $^{40}\text{Ca}+^{40}\text{Ca}$ ($E_{LAB} = 278$ MeV) collisions and found that, for the $^{40}\text{Ca}$ collision, they obtained charge distributions ($\approx 1.5 - 2$) that were less than half the experimental values ($\approx 4$) [23]. In later calculations Davies et al. [24, 25] performed a series of TDHF calculations for the heavy ion collisions $^{84}\text{Kr}+^{208}\text{Pb}$ ($E_{LAB} = 494$ MeV) and $^{84}\text{Kr}+^{209}\text{Bi}$ ($E_{LAB} = 600$ MeV), for which experimental data was available [26, 27], and found that they were able to reproduce the experimentally measured fragment energies, average masses and scattering angles but that their estimates for the full-width-half-maximum of the mass distribution, $\Delta N_{TDHF} \approx 3$, were consistently an order of magnitude lower than the experimental values. This was subsequently explained by Dasso et al. [28] who showed that the one-body nature of HF/TDHF
leads to an unphysical upper limit on the mass fluctuations that can be obtained from these approaches

\[(\Delta N_{\text{MAX}})^2 = \langle N \rangle^2 \left( 1 - \frac{1}{A} \langle N \rangle \right) \]  

(1.5)

We will derive this result in section 2.2. The consequences of this limitation are that the HF and TDHF approaches can correctly predict the expectation values for one-body operators, which depend solely upon the state of the individual nucleons, such as fragment mass, kinetic energy, angular momentum and scattering angle [29,30] but cannot, in general, predict expectation values which depend upon the states of multiple nucleons (two-body or many-body operators). To calculate a mass distribution according to (1.1) we must evaluate two expectation values; \( \langle N \rangle \), which is the expectation value of the mass of the nucleus of interest and is a one-body operator, and \( \langle N^2 \rangle \), which is the expectation value of a two-body operator and is the source of the errors. In practice it has been found that this deficiency is also seen in other observables such as momentum distributions [30] and the widths of giant resonances [28]. The one exception to this is the energy which is correctly predicted despite, in general, being a two-body observable (since the Hamiltonian is usually written as a sum of the one-body kinetic energy operator and the nucleon-nucleon interaction, a two-body term). This is because, in a HF calculation, the best approximation to the ground state is obtained by finding the state that minimises the energy in the system. We will see that taking an operator into account within the variational process is essential if we wish to get accurate results for operators which are not single-particle operators. Whilst the consequences of this problem have been slightly mitigated by the introduction of more sophisticated interactions (the inclusion, for example, of spin-orbit forces has been shown to result in increases in dissipation [31,32]) the fundamental problem remains.

Whilst the HF and TDHF mean-field approaches provide a very practical approach for the modeling of complex many-body systems the equations are still complicated, generally requiring a numerical solution. It is only recently that the rapid advances in computer power have allowed us to perform completely unrestricted three-dimensional calculations using the latest effective interactions [33–37]. Prior to this additional approximations and assumptions (both in the geometry (i.e. symmetries) and in the interactions) were necessary to make calculations feasible. These difficulties, coupled with the aforementioned weakness in the method [38] lead to a decline in interest in this approach at the end of the 1980’s, a decline which has been reversed as advances in computer power have removed some of these obstacles.

1.4 The Balian-Vénéroni Variational Method

In the 1980’s Balian and Vénéroni published a number of letters and papers [39–46] in which they investigate from first principles the best approaches for the variational determination of expectation values, correlations and fluctuations for one-body observables. Amongst these we will mainly refer to their three main papers.
1.4 The Balian-Vénéroni Variational Method

1. Time-Dependent Variational Principle for the Expectation Value of an Observable: Mean-Field Applications [44] (later reprinted as [47])

2. Static and Dynamic Variational Principles for Expectation values of Observables [45]

3. Correlations and Fluctuations in Static and Dynamic Mean-Field Approaches [46]

in which they laid out, in full, their new variational approach obtaining, amongst other results, an improved equation for determining the fluctuations of one-body observables. They found that if you wanted to calculate an observable other than a one-body observable (or the energy) then the quantity that you vary and minimise should depend upon both the energy and the observable that you are seeking to measure. They also showed that if the observable that you want to calculate is the expectation value of a single-particle operator, and you still want to describe the state of the system by a single Slater determinant, then their method provides the usual TDHF equations thus demonstrating that TDHF is one example of their more general variational approach. This approach will henceforth be referred to as the Balian-Vénéroni (BV) approach.

The derivation of the Balian-Vénéroni result is given in the next chapter but it is useful to present the final result now to help explain the strengths of this approach and highlight the links between this approach and the usual TDHF approach. They found that, given the state of a system described, at the time $t_0$, by the one-body density matrix, $\rho(t_0)$ (a Slater determinant satisfying $\rho^2 = \rho$), the fluctuation, $\Delta Q$, in a one-body observable, $\mathcal{Q}$, at some later time $t_1$, is given by [46]

$$\left(\Delta Q_{BV}\right)^2_{t_1} = \lim_{\epsilon \to 0} \frac{1}{2\epsilon^2} \text{Tr} \left[ (\rho(t_0) - \eta(t_0, \epsilon))^2 \right], \quad (1.6)$$

where $\eta(t, \epsilon)$ is a one-body density matrix related to $\rho(t)$ through the boundary condition

$$\eta(t_1, \epsilon) = \exp \left( i\epsilon \mathcal{Q} \right) \rho(t_1) \exp \left( -i\epsilon \mathcal{Q} \right), \quad (1.7)$$

and where, crucially, the time evolution of $\rho(t)$ and $\eta(t, \epsilon)$ is given by the usual TDHF equation. This equation differs from the standard TDHF result [43]

$$\left(\Delta Q_{TDHF}\right)^2_{t_1} = \text{Tr} \left[ \mathcal{Q} \rho(t_1) \mathcal{Q} (1 - \rho(t_1)) \right], \quad (1.8)$$

since (1.6) depends explicitly on the initial time, $t_0$, with the final time, $t_1$, entering only through the boundary condition (1.7). The other key feature of this result is that it contains, through (1.7), the observable $\mathcal{Q}$ such that this method is specifically tuned to the determination of the fluctuation of the observable of interest. Evaluating (1.6) requires a TDHF calculation to be run from $t_0 \to t_1$ to obtain $\rho(t_1)$ from $\rho(t_0)$ (the initial state of the system which is always assumed to be known). This is used to obtain $\eta(t_1, \epsilon)$ and then an additional TDHF calculation, running backwards $t_1 \to t_0$, must be carried out to obtain $\eta(t_0, \epsilon)$ with the latter steps being repeated for differing values of $\epsilon$ so as to evaluate the limit in (1.6).
One of the key points about this new approach is that the equations of motion which must be solved have the same form as the equations of motion in standard TDHF which means that existing TDHF codes (with their usual interactions) can be adapted to use the new formalism thereby avoiding the need to develop completely new codes. That said, evaluating (1.6) requires that a series of TDHF calculations be carried out for each observable which is being sought significantly increasing the computational cost. A small number of calculations were performed in the 1980’s to demonstrate the validity of this new approach [30,35,48] although limited computing power meant that these used symmetries (spherical or axial) and/or simplified nucleon-nucleon interactions (BKN) [30] to make the calculations more feasible.

1.5 Previous Applications of the Balian-Vénéroni Approach

The first realistic application of the Balian-Vénéroni method was the calculation of the mass distribution for a giant monopole resonance (GMR) in $^{40}\text{Ca}$ decaying by particle emission by Troudet and Vautherin in 1985 [48]. Their calculations were carried out using a spherically symmetric code and the BKN interaction as described in [49]. The final mass of the $^{40}\text{Ca}$ nucleus was $\langle N \rangle = 33.10$ with $\Delta N_{TDHF} = 2.30$. From this expectation value we obtain, through (2.11), the maximum mass distribution that can be obtained through TDHF, $\Delta N_{MAX} = 2.39$. They then performed a series of additional time-reversed TDHF calculations for various values of $\varepsilon$ to obtain estimates for $\Delta N_{BV}(\varepsilon)$ and perform the extrapolation $\varepsilon \to 0$. Their resulting graph (and extrapolation) is shown in figure 1.2 and gives $\Delta N_{BV} = 4.38$, which is nearly twice the TDHF value.

The first collisions calculations were by Bonche et al. [35,42] who performed calculations for the simple symmetric system $^{16}\text{O}+^{16}\text{O}$ ($E_{LAB} = 160$ MeV, impact parameter $l = 30\hbar$) using a three-dimensional code and BKN interaction and imposing spin-isospin symmetry to limit the number of states under consideration to four. This setup corresponds to an impact parameter just above fusion, chosen with the aim of maximising the mass distribution. This same calculation has also been carried out by Marston and Koonin [30] who used a two-dimensional code and the more realistic SkII interaction. They used the “clutching” model to perform collisions with non-zero impact parameters in which the moment of inertia of the two nuclei was assumed to be that of a rigid body once the density between the two nuclei had reached half the nuclear saturation (peak) density. As the reaction progressed the $z$-axis joining the two nuclei rotated in space whilst symmetry was maintained about that axis. They also performed this calculation for an impact parameter $l = 0\hbar$ as well as calculations for $^{40}\text{Ca}+^{40}\text{Ca}$ at $E_{LAB} = 278$ MeV and impact parameters of $l = 0$, $30\hbar$ where this latter case was chosen to allow a comparison with an experimental measurement from 1975 of $\Delta N_{EXP} \approx 4$ by Colombani et al. [23]. However, this experimental paper only shows charge distributions and not mass distributions. If we take this to be the charge distribution and not the mass distribution then this value would be consistent with
1.5 Previous Applications of the Balian-Vénéroni Approach

Figure 1.2: (Figure 1 from [48]) This is the first application of the Balian-Vénéroni approach by Troudet and Vautherin to a $^{40}$Ca GMR decaying by particle emission using a BKN interaction (in a spherically symmetric code). Their values of $\Delta N_{BV} (\varepsilon)$, plotted as a function of $\varepsilon$ are shown (dashed line) along with their extrapolation to $\varepsilon = 0$ (dot-dashed line).

the measurements of Royuette et al. (1977) who measured this reaction at $E_{LAB} = 256$ MeV [50] and obtained, averaging over all angles, a width of approximately 4 (see figure 1.3).

In a more recent experiment (1991) this reaction has been measured for $E_{LAB} = 197$ and 231 MeV and the full-width-half-maximum ($\Gamma_{FWHM}$) of the mass distributions was measured to be $\approx 26$ in both reactions [51]. If we assume, as in [52], that these mass distributions are normal (gaussian) distributions, such that $\Gamma_{FWHM} = \sqrt{8\ln(2)} \Delta N$, then this gives $\Delta N_{EXP} \approx 11$ which is significantly larger than the previous values despite being measured at lower energies. We would expect the mass distribution to be larger at higher energies so this gives us an approximate figure

<table>
<thead>
<tr>
<th>Code</th>
<th>Interaction</th>
<th>$E_{LAB}$ (MeV)</th>
<th>$l$ (h)</th>
<th>$\Delta N_{TDHF}$</th>
<th>$\Delta N_{BV}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{40}$Ca ISGMR [48]</td>
<td>1D</td>
<td>BKN</td>
<td></td>
<td></td>
<td>2.30</td>
</tr>
<tr>
<td>$^{16}$O+$^{16}$O [35, 42]</td>
<td>3D</td>
<td>BKN</td>
<td>160</td>
<td>30</td>
<td>0.81</td>
</tr>
<tr>
<td>$^{16}$O+$^{16}$O [30]</td>
<td>2D</td>
<td>SkII</td>
<td>160</td>
<td>0</td>
<td>0.574</td>
</tr>
<tr>
<td>$^{16}$O+$^{16}$O [30]</td>
<td>2D</td>
<td>SkII</td>
<td>160</td>
<td>30</td>
<td>0.495</td>
</tr>
<tr>
<td>$^{40}$Ca+$^{40}$Ca [30]</td>
<td>2D</td>
<td>SkII</td>
<td>278</td>
<td>0</td>
<td>1.62</td>
</tr>
<tr>
<td>$^{40}$Ca+$^{40}$Ca [30]</td>
<td>2D</td>
<td>SkII</td>
<td>278</td>
<td>30</td>
<td>1.26</td>
</tr>
</tbody>
</table>

Table 1.1: A summary of the results of all the previous calculations of mass distributions using the Balian-Vénéroni variational approach. All values are taken directly from the original publications.
1.6 Related Works

Almost all practical microscopic models rely on the assumption that the state of the system can be written as a Slater determinant. However, once that assumption is made there are still a large range of models, all based around different, but related, equations of motion for the microscopic or macroscopic properties of the system. The vast majority of these methods can be related back to Brownian motion as described by the Langevin equation [53,54]

\[ m \frac{dv(t)}{dt} = -\gamma v(t) + R(t), \]  

which describes the motion of a particle of mass ‘m’ in the presence of friction, described by the friction coefficient \( \gamma \), and a random force, \( R(t) \). These latter quantities are related by the so-called fluctuation-dissipation theorem which states that any dissipative process (i.e. friction) must be associated with fluctuations (the random force) and which also implies that you can’t have one
without the other [55]. Many of the difficulties in implementing this type of model in the nuclear case are associated with choosing $\gamma$ and $R(t)$ and then solving the resulting complicated, and sometimes coupled, equations. The inclusion of random forces makes this a stochastic approach and ensures that two independent calculations produce a different final state of the system. Repeated calculations produce a range (or ensemble) of final states and hence a broader distribution in the values of any given observable than can be obtained from any single calculation but are extremely demanding with large numbers of calculations being required (e.g. hundreds [56,57]) to obtain an accurate description of the evolution of the system.

In microscopic models the role of these previously omitted interactions is played by a collision term which can be related to the two-body correlations which are usually neglected in TDHF (Appendix B and particularly (B.9)). A theoretical description of the inclusion of collision terms in TDHF was given in (1984) by Reinhardt et al. [58,59]. Their work include similarities to the work of Balian and Vénéroni, with the use of statistical mechanics philosophies (the consideration of entropy as a measure of the amount of information/chaos in a system) and the inclusion of operators (such as those associated with conserved quantities) in the dynamic process. An example is the Uhling-Uhlenbeck collision term as implemented in the Vlasov equation and the Vlasov-Uhling-Uhlenbeck or Vlasov-Boltzmann (VUU) equations [60,61].

However, the Langevin equation tells us that any collision term should be balanced by a fluctuating (or random) force. The addition of this stochastic element in the collision term in time-dependent microscopic calculations was discussed by Ayik and Gregoire [60] (Erratum: [61]) where they included an additional “random” element (sometimes referred to as numerical noise [62]) in addition to retaining some of the previously neglected two-body correlations. There are many such models which include random and/or collision terms and which consider the time-evolution of either the state of the system (as described by density matrices) or, more simply, of particular attributes of the system, for example [60,61,63] where they consider the time-dependence of the momentum distributions in a two-dimensional Fermi gas.

There has also been some success with the more classical models of nuclear reactions such as the anti-symmetrised molecular dynamics model [64] with two-nucleon collision terms which was applied with some success to the $^{12}$C+$^{12}$C reaction at 28.7 MeV/nucleon although the emphasis in these models has tended to be on higher energy multi-fragmentation reactions looking at the production of light particles and intermediate mass fragments. Other examples of multi-fragmentation calculations are those of Bauer et al. [65] (1987) where they used a microscopic-macroscopic approach with collision terms to look at the production of light fragments in the reaction $^{20}$Ne+$^{20}$Ne at 100 MeV/nucleon or the more recent three-dimensional stochastic calculations of Xe+Sn at 50 MeV/A [56] (1996) or Au+Au at 100-400 MeV/A [66] (1997).
1.6 Related Works

Statistical models, such as the PACE4 simulations package [67] or LISE [68–70], are frequently used by experimentalists to predict the production yields of nuclei following a range of reactions using different statistical methods (e.g. Monte-Carlo methods) modelling each type of nuclear reaction (scattering, fission ...etc...) using a different statistical model. These types of models generally work by estimating decay probabilities based upon excitation energies, level densities, reaction rates and coupling coefficients (all of which have to be estimated using existing experimental/theoretical values for cross-sections ...etc...). The applicability of any statistical technique to many nuclear physics problems is complicated by the fact that, except in very low energy cases, the reaction processes can be too fast for one to be able to assume that the system has reached equilibrium as is often assumed [71].

Only recently have these stochastic methods been applied to cases that are of direct relevance to the current work, such as the recent calculations of the strength functions for resonances in $^{40}$Ca and other heavier nuclei by Lacroix et al. [72,73] using the small amplitude limit of Extended TDHF (TDHF with collision terms) as well as stochastic calculations of the rms radii [74] for a $^{40}$Ca decaying GMR. The formalism used in this later calculation remains too simplified for use in calculating fluctuations.

In another recent work (2008) Ayik [75] has presented a theoretical model, based upon stochastic TDHF, for calculating fluctuations within a mean-field framework, and particularly the fluctuations associated with one-body observables. They obtain, for these fluctuations some of the same results as are obtained using the Balian-Vénérioni approach (with equations that include a clear dependence upon the observable of interest) although they do not obtain Balian and Vénérioni's final result (which can be implemented using standard TDHF codes). They do not present any applications of their equations. The links between this work and that of Balian and Vénérioni serve to reinforce the view that many of these microscopic theories (with their myriad equations of motion) are related and can be obtained as different special cases of more general theories.

The use of stochastic models in nuclear physics is discussed at length in the 1996 review article of Abe et al. [54]. More recent reviews of microscopic models, including their descriptions of low-energy nuclear collisions, are given by Bonche [76] (2000) and by Bender et al. [77] (2003).
Chapter 2

Theory

In this chapter we start by introducing the operator for the mass of the nucleus, or nuclear fragment, of interest and hence the equations for determining the expectation value and fluctuation in the nucleus' mass. We introduce the equations used to calculate these quantities in the usual HF/TDHF approach and demonstrate why these approaches do not yield the correct results for mass fluctuations.

We then review the Balian-Vénéroni approach for the variational determination of the expectation values and fluctuations for the arbitrary one-body operator $\hat{Q}_i$. This method should be viewed as a more general variational approach than the usual HF and TDHF approaches rather than as an extension of these methods, which are just special cases of this more general theory. Several of the equations and results used or obtained during this derivation also appear in the derivations of the key equations for the HF and TDHF approaches which are included as appendices A and B respectively (although these are also available in any standard text e.g. [78, chap. 5, 12]). In the works of Balian and Vénéroni (principally [45-47]) they discuss the variational determination of the expectation values, $\langle Q_i \rangle$, fluctuations, $\Delta Q_i$, and correlations, $C_{ij}$, for the arbitrary set of single-particle operators $\hat{Q}_i$ where a fluctuation is a special case of the correlation between two operators $\hat{Q}_i$ and $\hat{Q}_j$

$$C_{ij} = \frac{1}{2} \left[ \langle Q_i Q_j \rangle + \langle Q_j Q_i \rangle - \langle Q_i \rangle \langle Q_j \rangle \right], \quad (2.1)$$

such that, for $\hat{Q}_i = \hat{Q}_j$, $C_{ii} = (\Delta Q_i)^2$. Thus, whilst our main interest is the fluctuations, $\Delta Q_i$, the theory presented in this thesis is also valid for calculating the correlations between single-particle observables with the final simplification for fluctuations being made at the end. Balian and Vénéroni's full works covered both static and dynamic mean-field models and as such are somewhat broader and more general than the current work (although many of the results they obtain, whilst theoretically interesting, continue to be of limited practical use due to the difficulties that would be encountered in their implementation).
2.1 Mass Distributions

We consider the mass distribution in a bounded region of space around a nucleus of interest after a dynamical process (a nuclear collision or the decay of a giant resonance through particle emission) and calculate the mass (number of nucleons) in the nucleus according to

$$\langle N(R_c, t) \rangle = \sum_m \int d\vec{r} |\phi_m(\vec{r}, t)|^2 \theta (R_c - |\vec{r} - \vec{r}_{CM}|),$$  \hspace{1cm} (2.2)

where $R_c$ is the cutoff radius used to define a bounded region of space about the centre-of-mass, $\vec{r}_{CM}$, of the nucleus of interest through the use of the theta function

$$\theta(x) = \begin{cases} 1 & x \geq 0, \\ 0 & x < 0, \end{cases}$$  \hspace{1cm} (2.3)

such that $\hat{N} = \theta (R_c - |\vec{r} - \vec{r}_{CM}|)$. The summation over $m$ runs over all occupied single-particle states. The fluctuation in the particle number, $\Delta N (R_c, t)$ is then given by

$$\langle \Delta N (R_c, t) \rangle^2 = \langle (\langle N (R_c, t) \rangle)^2 \rangle - \langle N (R_c, t) \rangle^2.$$

\hspace{1cm} (2.4)

2.2 Expectation Values and Fluctuations in HF and TDHF

In HF and TDHF it is assumed that the state of the system is described at all times by an anti-symmetrised product of single-particle states, $\phi(\vec{r}_i, t)$, a Slater determinant (1.2), which is used to determine all the desired properties of the system.

The expectation value for an arbitrary single-particle operator, $\hat{Q}_i$, at the time $t$ is then given by

$$\langle \hat{Q}_i \rangle |_t = \text{Tr} \left[ \hat{Q}_i \rho (t) \right].$$  \hspace{1cm} (2.5)

The fluctuation in $\hat{Q}_i$ is calculated using the usual expression for the standard deviation, (1.1) which, in TDHF becomes, using Wick's theorem (see, for example, [78, app. C.4])

$$\langle (\Delta Q^{TDHF}_i)^2 \rangle |_t = \text{Tr} \left[ \hat{Q}_i \rho (t) \hat{Q}_i (1 - \rho (t)) \right].$$  \hspace{1cm} (2.6)

As previously discussed the shortcomings of this result were originally observed by Koonin et al. [22] (who performed axially symmetric TDHF calculations of $^{16}O+^{16}O$ ($E_{LAB} = 192$ MeV) and $^{40}Ca+^{40}Ca$ ($E_{LAB} = 278$ MeV) collisions) and Davies et al. [24, 25] (who performed calculations for the heavy ion collisions $^{84}Kr+^{208}Pb$ ($E_{LAB} = 494$ MeV) and $^{84}Kr+^{209}Bi$ ($E_{LAB} = 600$ MeV)) and found that the charge/mass distributions given by (2.6) were significantly smaller than those measured experimentally [23, 26, 27]. This was subsequently explained by Dasso et al. [28] who showed that calculating $\Delta Q_i$ according to (2.6), where $\rho (t)$ is a Slater determinant, artificially
limits the mass distributions that can be obtained using this approach.

To demonstrate this we consider a system consisting of two colliding nuclei with a combined mass of \( A \) nucleons and where the objective is to measure the fluctuations in the mass of one of the nuclei after the collision. It is assumed that the state of the system is given, at the time \( t \), by a Slater determinant constructed using the set of single-particle states, \( \phi_i (\vec{r}, t) \), defined within some spatial box. A region-of-interest, \( R \), is defined within this spatial box such that this region encloses only the nucleus of interest. The expectation value for the number of nucleons within this nucleus at the time \( t \), \( \langle N \rangle_t \), is given by

\[
\langle N \rangle_t = \sum_i \langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R ,
\]

(2.7)

where the summation runs over all occupied single-particle states and the overlaps are given by

\[
\langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R = \int_R d\vec{r} \phi_i^\ast (\vec{r}, t) \phi_i (\vec{r}, t) .
\]

(2.8)

The fluctuation in \( \langle N \rangle_t \), \( \Delta N \), is the given (using (2.6)) by

\[
(\Delta N)^2 \bigg|_t = \langle N^2 \rangle_t - \langle N \rangle_t^2 = \sum_i \langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R - \sum_{ij} \langle \phi_i (\vec{r}, t) | \phi_j (\vec{r}, t) \rangle_R |_{ij}^2 .
\]

(2.9)

These equations are valid for any arbitrary single-particle basis so we now perform a unitary transformation into a basis in which the matrix elements \( \langle \phi_i (\vec{r}, t) | \phi_j (\vec{r}, t) \rangle_R \) are the elements of a diagonal matrix (but are not the elements of the single-particle density matrix since the integrations are not performed over all space). We then obtain

\[
(\Delta N)^2 \bigg|_t = \sum_i \langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R (1 - \langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R ) .
\]

(2.10)

This formula provides two notable limiting cases. Consider first if all of the nucleons in the system are fully bound within their parent nucleus (i.e. as the nuclei approach each other but before they are close enough to interact). The region \( R \) contains only the nucleus of interest so the overlaps, \( \langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R \), are unity for a nucleon within that nucleus and zero otherwise. This immediately provides the expected and trivial result \( \Delta N \big|_t = 0 \).

Alternatively, consider if all of the single-particle wavefunctions are fully dispersed throughout the spatial box. In this case the probabilities for finding any arbitrary nucleon at any arbitrary point within the spatial box are all equal. The overlaps \( \langle \phi_i (\vec{r}, t) | \phi_i (\vec{r}, t) \rangle_R \) are then determined by the size of the region \( R \) as a fraction of the size of the entire spatial box which leads directly to the result

\[
(\Delta N_{\text{MAX}})^2 \bigg|_t = \langle N \rangle_t \left( 1 - \frac{\langle N \rangle_t}{A} \right) .
\]

(2.11)
which tells us that the square of the fluctuation in the mass of any one fragment cannot exceed the reduced mass of the system. For the simple case of a collision between two identical nuclei, with initial masses of $A/2$ nucleons, and approximating the number of nucleons in the nucleus of interest to the number of nucleons in the appropriate half of the spatial box (e.g. the left half) gives, in the extreme case of maximum dispersion, \( \langle N \rangle_R = A/2 \). The overlaps are then all equal, \( \langle \phi_i (r, t) | \phi_i (r, t) \rangle_R = A/2 \), leading to a mass fluctuation \( \Delta N^2 |_L = A/4 \).

In practice the single-particle wavefunctions are unlikely to be evenly distributed throughout the spatial box as assumed here but these results represent the theoretical upper limit on the mass fluctuations that can be obtained from (2.6) and in practice the calculated mass fluctuation will always be less than the value obtained from (2.11). This upper limit has no physical basis and is a consequence of using (2.6) and assuming that the state of the system can be approximated to a Slater determinant. Experimentally measured mass fluctuations can, and do, exceed this limit. This particular result only applies to the particle number operator (or, equivalently, the charge operator) due to the simple form of these operators and it can’t be suggested, based purely on these arguments, that a similarly simple result can be given for other arbitrary operators without further analysis. However, the very existence of this result should serve as a warning about the limitations of the HF/TDHF approaches and the range of observables for which they can confidently be used.

2.3 The Balian-Vénéroni Approach

2.3.1 The Balian-Vénéroni Approach for Single-Particle Observables

In the HF approach the state of the system (a Slater determinant) is determined by minimising the energy in the system and used to determine the expectation values, \( \langle Q_i \rangle \), correlations, \( C_{ij} \), and fluctuations, \( \Delta Q_i \), for all arbitrary (single-particle) operators, \( \hat{Q}_i \). Given that the true many-body state of the system has been approximated to a Slater determinant it is sensible to ask if the usual formulae still provide the most accurate results for the determination of some arbitrary observable now that this assumption has been made. In this section we follow the derivation of the Balian-Vénéroni equations from [44-46].

Before considering the variational determination of the fluctuations for single-particle operators it is helpful to start (as Balian and Vénéroni do) by asking how we should write the state of the system if it must accurately describe the system whilst simultaneously satisfying some set of additional conditions. These conditions are introduced by requiring that this state yield specific expectation values for the set of operators \( \hat{M}_i \) (these might, for example, be the operators associated with conserved quantities [79]). These are collectively introduced through the operator \( \hat{A} \) defined as

\[
\hat{A} = \sum_i \hat{M}_i,
\]  
(2.12)
where the only restriction upon this set of operators is that it includes a unit operator. The first operator in the series, $\hat{M}_0$, is therefore required to be a unit operator whilst the others ($i \geq 1$) remain arbitrary. In later sections these operators will become the variables whose expectation values are being sought but for the moment it will be assumed that the expectation values for these operators, $\langle \hat{M}_i \rangle$, are known and are related to $D$, the density matrix describing the state of the system, by

$$\langle \hat{M}_i \rangle = \frac{\text{Tr} [\hat{M}_i D]}{\text{Tr} [D]}.$$  \hspace{1cm} (2.13)

If the state $D$ is normalised then $\text{Tr} [D] = 1$ and the denominator disappears leaving the usual result $\langle \hat{M}_i \rangle = \text{Tr} [\hat{M}_i D]$.

It is known from information theory and statistical mechanics [58,80,81] that there will be a set of density matrices, $D$, each associated with a density operator $\hat{D}$, which will give the same results for the expectation values $\langle \hat{M}_i \rangle$ but that the most useful of these is the so-called "reduced" density matrix for which the entropy, $S'$, is maximum

$$S = -\text{Tr} [D \ln (D)],$$ \hspace{1cm} (2.14)

since this is the one which is "least biased". The entropy measures the amount of information in a state, regardless of how relevant this information is to any particular observable, so the state with the largest entropy will contain the least information. We require that the state contain the information necessary to yield the set of expectation values $\langle \hat{M}_i \rangle$ but we also want it to be as general (and therefore un-biased) as possible with regard to any other operators that we could choose to consider. This means that we want the state $D$ which contains the minimum amount of information necessary to provide the expectation values $\langle \hat{M}_i \rangle$ but no more.

Such a density matrix is known to be given by [80]

$$\hat{D} = \exp \left( -\sum_i \mu_i \hat{M}_i \right),$$ \hspace{1cm} (2.15)

where the $\mu_i$'s are Lagrange multipliers and, since $M_0$ is a unit operator, $\mu_0$ guarantees the normalisation of the state.

In this approach $D$ describes the state of the system but also depends upon one or more observables associated with the system. This is not entirely dissimilar from the usual HF approach where the state of the system is determined by minimising the energy in the system, however this idea is now generalised and extended so that $D$ depends upon all of the observables of interest.

The aim of the Balian-Vénéroni approach is to derive the set of equations that should be solved to
obtain the best possible result for whatever observable is being sought. Expectation values are still obtained using the usual form, \( \langle A \rangle = \frac{\text{Tr} [\hat{A} \hat{D}]}{\text{Tr} [\hat{D}]} \), but the state \( \hat{D} \) must now be optimised for the determination of \( \langle A \rangle \). This means that we should not think of \( \hat{D} \) as a density matrix describing the state of the system but as a tool for calculating particular expectation values.

### 2.3.2 Fluctuations and Correlations for Single-Particle Observables

Evaluating the fluctuations for the single-particle operator \( \hat{Q}_i \), \( \Delta Q_i \) requires the determination of the expectation values \( \langle Q_i \rangle \) and \( \langle Q_i^2 \rangle \), the latter of which is a two-body operator. In the case of the correlations between two (assumed non-commuting) single-particle operators, \( \hat{Q}_i \) and \( \hat{Q}_j \), we would need to determine the expectation values \( \langle Q_i \rangle \), \( \langle Q_j \rangle \), \( \langle Q_i Q_j \rangle \) and \( \langle Q_j Q_i \rangle \) where we have two two-body operators. It is possible to derive the sets of equations that must be solved to determine the state \( \hat{D} \) optimised to the determination of a one-body, e.g. \( \langle Q_i \rangle \), or a two-body, e.g. \( \langle Q_i^2 \rangle \), operator however solving these sets of equations can be a complex problem and finding the desired fluctuation or correlation then requires combining these results, each of which will have been determined independently and by the solution of a markedly different set of equations [47]. It is preferable to calculate the desired fluctuation or correlation as the result of a single calculation to avoid any inconsistencies.

To calculate the expectation values, correlations and fluctuations for a set of operators, \( \hat{Q}_i \), simultaneously we consider (in place of (2.12)) the operator

\[
\hat{A} = \exp \left( - \sum_i \varepsilon_i \hat{Q}_i \right),
\]

where the \( \varepsilon_i \)'s are a set of parameters, which are assumed to be numerically small. Furthermore, instead of the expectation value \( \langle A \rangle \), we consider its logarithm

\[
\ln \langle \langle A \rangle \rangle = \ln \left( \frac{\text{Tr} \left[ \exp \left( - \sum_i \varepsilon_i \hat{Q}_i \right) \hat{D} \right]}{\text{Tr} [\hat{D}]} \right),
\]

which we expand to second order in \( \varepsilon \) and rearrange to obtain

\[
= \ln \left( \text{Tr} [\hat{D}] \left( 1 - \sum_i \frac{\text{Tr} [\hat{Q}_i \hat{D}]}{\text{Tr} [\hat{D}]} + \frac{1}{2} \sum_{ij} \varepsilon_i \varepsilon_j \frac{\text{Tr} [\hat{Q}_i \hat{Q}_j \hat{D}]}{\text{Tr} [\hat{D}]} \right) \right) - \ln (\text{Tr} [\hat{D}]).
\]

The resulting \( \ln (\text{Tr} [\hat{D}]) \) terms can be either cancelled out or eliminated by requiring that the state
D be normalised. The remaining terms can be expanded and rearranged to leave

\[
\ln \langle A \rangle = - \sum_i \epsilon_i \langle Q_i \rangle + \frac{1}{2} \sum_{ij} \epsilon_i \epsilon_j \langle Q_i Q_j \rangle - \frac{1}{2} \sum_{ij} \epsilon_i \epsilon_j \langle Q_i \rangle \langle Q_j \rangle,
\]

\[
= - \sum_i \epsilon_i \langle Q_i \rangle + \frac{1}{2} \sum_{i,j} \epsilon_i \epsilon_j C_{ij},
\]

(2.19)

where \( C_{ij} = \frac{1}{2} (\langle Q_i Q_j \rangle + \langle Q_j Q_i \rangle) - \langle Q_i \rangle \langle Q_j \rangle \) and, for \( i = j \), \( C_{ii} = \Delta Q^2_i = \langle Q_i^2 \rangle - \langle Q_i \rangle^2 \). This result means that by writing the operators in the form (2.16) all expectation values, correlations and fluctuations for the set of arbitrary single-particle operators \( \hat{Q}_i \) can be found by investigating the dependence of \( \ln \langle A \rangle \) on the \( \epsilon_i \)'s.

### 2.3.3 Equations of Motion for a State and an Observable

Consider the determination of the expectation value of some (assumed to be time-dependent) observable, \( \hat{A}(t) \), at the time \( t_1 \)

\[
\langle A(t) \rangle_{t_1} = \text{Tr} \left[ \hat{A}(t_1) D(t_1) \right],
\]

(2.20)

where \( D(t_1) \) is the full many-body density matrix describing the state of the system at the time \( t_1 \) (and which is assumed to be normalised). It is assumed that the state of the system, at some earlier time \( t_0 \), is known and given by the (normalised) density matrix, \( D(t_0) \). These two conditions, the fixed nature of \( \hat{A}(t) \) at the time \( t_1 \) and \( D(t) \) at the time \( t_0 \) are our boundary conditions and will be referred to repeatedly throughout this derivation. The time evolution of any state (or operator) is given by the unitary time-evolution operator (we use \( \hbar = 1 \) throughout)

\[
\hat{U}(t_b, t_a) = \hat{U}^\dagger(t_a, t_b) = \exp \left( -i \int_{t_a}^{t_b} dt \hat{H}(t) \right),
\]

(2.21)

where \( \hat{H}(t) \) is the Hamiltonian for the system which may also be time-dependent. The evolution operator can be applied to a state (or operator), known at some time \( t_a \) (e.g. \( |m, t_a\rangle \)), to determine that state (or operator) at some other time \( t_b \).

\[
\hat{U}(t_b, t_a) |m, t_a\rangle = |m, t_b\rangle.
\]

(2.22)

The density matrix, at the time \( t_1 \), \( D(t_1) \), can be written in terms of the original density, \( D(t_0) \), and the evolution operator

\[
D(t_1) = \hat{U}(t_1, t_0) D(t_0) \hat{U}^\dagger(t_1, t_0).
\]

(2.23)
The Balian-Vénérioni Approach

The Forward Schrödinger Picture

The time-evolution of the density operator can be described by an equation of motion. The density matrix at some time, \( t \), is related to the density matrix describing the initial state of the system through

\[
D(t) = \hat{U}(t, t_0) D(t_0) \hat{U}^\dagger(t, t_0) .
\]

(2.24)

Differentiating with respect to time, noting that \( D(t_0) \) is independent of the time \( t \), and using (2.21) gives

\[
\frac{dD(t)}{dt} = \frac{d\hat{U}(t, t_0)}{dt} D(t_0) \hat{U}^\dagger(t, t_0) + \hat{U}(t, t_0) \frac{dD(t_0)}{dt} \hat{U}^\dagger(t, t_0) ,
\]

\[
= -i\hat{H}(t) D(t) + i D(t) \hat{H}(t) ,
\]

\[
= i \left[ D(t), \hat{H}(t) \right] ,
\]

(2.25)

where we recognise the Liouville-von Neumann equation [47] describing the time evolution of the density operator. This equation has a similar functional form to the TDHF equation (B.11) but differs in that it contains the full many-body Hamiltonian and because \( D(t) \) is not assumed to be a Slater determinant.

The Backward Heisenberg Picture

Writing \( A(t) \) in terms of the state of the system at the final time, \( D(t_1) \), is a use of the forward Schrödinger picture where an evolution operator is applied in the forward direction to progress the density matrix from \( t_0 \) to \( t_1 \). It is entirely equivalent to evolve the operator for the observable backwards through time from \( t_1 \) to \( t_0 \)

\[
\hat{A}(t_0) = \hat{U}(t_0, t_1) \hat{A}(t_1) \hat{U}^\dagger(t_1, t_0) . 
\]

(2.26)

This can easily be demonstrated from (2.20) using the time-evolution operator (2.21) and the cyclic properties of the trace

\[
\langle A(t) \rangle_{t_1} = \text{Tr} \left[ \hat{A}(t_1) D(t_1) \right] = \text{Tr} \left[ \hat{A}(t_1) \hat{U}(t_1, t_0) D(t_0) \hat{U}^\dagger(t_1, t_0) \right] ,
\]

\[
= \text{Tr} \left[ \hat{U}(t_0, t_1) \hat{A}(t_1) \hat{U}^\dagger(t_0, t_1) D(t_0) \right] = \text{Tr} \left[ \hat{A}(t_0) D(t_0) \right] .
\]

(2.27)

This is the backward Heisenberg picture in which time flows backwards from the time \( t_1 \). An equation of motion for \( A(t) \) can be derived using the same method as was used to find (2.25). This gives

\[
\frac{d\hat{A}(t)}{dt} = i \left[ \hat{A}(t), \hat{H}(t) \right] .
\]

(2.28)

which is known as the Heisenberg equation of motion.
2.3.4 The Characteristic Function

Characteristic functions are useful for re-defining a problem to make it easier to solve or to introduce constraints that the solution of the problem must satisfy. In the Balian-Veneroni approach we determine the expectation value of the operator $\hat{A}(t)$ by finding the stationary value of the characteristic function, $J$, chosen so that its stationary value is the same as the expectation value being sought and which, in dynamic problems, takes the form

$$J = \text{Tr} \left[ \hat{A}(t) \hat{D}(t) \right] - \int_{t_0}^{t_1} \text{d}t \left( \text{Tr} \left[ \hat{A}(t) \frac{d\hat{D}(t)}{dt} \right] - h \left( \hat{A}(t), \hat{D}(t) \right) \right),$$

(2.29)

where, as before, $\hat{A}(t)$ and $\hat{D}(t)$ denote the operator of interest and the density matrix and $t_0$ and $t_1$ are the initial and final times. The expectation value of interest appears as the first term whilst the second term brings in the time-dependence of the state of the system and the observable.

$h \left( \hat{A}(t), \hat{D}(t) \right)$ is the pseudo-Hamiltonian defined as

$$h \left( \hat{A}(t), \hat{D}(t) \right) = -i \text{Tr} \left[ \hat{A}(t) \left[ \hat{H}(t), \hat{D}(t) \right] \right],$$

(2.30)

$$= -i \text{Tr} \left[ \left[ \hat{A}(t), \hat{H}(t) \right] \hat{D}(t) \right],$$

(2.31)

where $\hat{H}(t)$ is the Hamiltonian for the system under consideration. To obtain the stationary value for the characteristic function, $J$ (and show that it is indeed the same as the expectation value being sought) we substitute in using (2.30) and obtain

$$J = \text{Tr} \left[ \hat{A}(t_1) \hat{D}(t_1) \right] - \int_{t_0}^{t_1} \text{d}t \text{Tr} \left[ \hat{A}(t) \left( \frac{d\hat{D}(t)}{dt} + i \left[ \hat{H}(t), \hat{D}(t) \right] \right) \right],$$

(2.32)

which contains, in the integral term, the Liouville-von Neumann equation, (2.25). Alternatively, using (2.31) instead of (2.30) and integrating the first term in the integral gives

$$J = \text{Tr} \left[ \hat{A}(t_0) \hat{D}(t_0) \right] + \int_{t_0}^{t_1} \text{d}t \text{Tr} \left[ \left( \frac{d\hat{A}(t)}{dt} - i \left[ \hat{A}(t), \hat{H}(t) \right] \right) \hat{D}(t) \right],$$

(2.33)

where we recognise the Heisenberg equation of motion and where the first term now depends solely on $t_0$. Using (2.33) and (2.32) respectively one obtains the change (or variation) in $J$, $\delta J$, due to a small change in either $\hat{D}(t)$, $\delta \hat{D}$, or $\hat{A}(t)$, $\delta \hat{A}$

$$\delta J_D = \text{Tr} \left[ \hat{A}(t_0) \delta \hat{D}(t_0) \right] + \int_{t_0}^{t_1} \text{d}t \text{Tr} \left[ \left( \frac{d\hat{A}(t)}{dt} - i \left[ \hat{A}(t), \hat{H}(t) \right] \right) \delta \hat{D}(t) \right],$$

(2.34)

$$\delta J_A = \text{Tr} \left[ \delta \hat{A}(t_1) \hat{D}(t_1) \right] - \int_{t_0}^{t_1} \text{d}t \text{Tr} \left[ \delta \hat{A}(t) \left( \frac{d\hat{D}(t)}{dt} + i \left[ \hat{H}(t), \hat{D}(t) \right] \right) \right].$$

(2.35)
The total variation in $J$ due to arbitrary variations in $A(t)$ and $D(t)$ is given by

$$\delta J = \delta J_D + \delta J_A$$

where

$$\begin{align*}
\delta J &= \text{Tr} \left[ \hat{A}(t_0) \delta D(t_0) \right] + \text{Tr} \left[ \delta \hat{A}(t_1) D(t_1) \right] \\
&+ \int_{t_0}^{t_1} dt \text{Tr} \left[ \left( \frac{d\hat{A}(t)}{dt} - i \left[ \hat{A}(t), \hat{H}(t) \right] \right) \delta D(t) \right] \\
&- \delta \hat{A}(t) \left( \frac{d\hat{D}(t)}{dt} + i \left[ \hat{H}(t), D(t) \right] \right). 
\end{align*}$$

(2.36)

The Stationary Value of the Characteristic Function

The boundary conditions for the problem require that $D(t)$ is fixed at the initial time $t_0$ and that $\hat{A}(t)$ is fixed at $t_1$. Thus, although the variations $\delta \hat{A}(t)$ and $\delta D(t)$ are arbitrary in general it is required that

$$\delta D(t_0) = \delta \hat{A}(t_1) = 0.$$  

(2.37)

These conditions, combined with the equations of motion for $\hat{A}(t)$ and $D(t)$ (2.25, 2.28) ensure that $\delta J$ is zero and provide the stationary value of $J$, $J_{st}$

$$J_{st} = \text{Tr} \left[ \hat{A}(t_1) D(t_1) \right] = \text{Tr} \left[ \hat{A}(t_0) D(t_0) \right].$$ 

(2.38)

The stationary value of the characteristic function is the same as the expectation value of the operator $\hat{A}(t)$ evaluated at the time $t_1$. Thus the problem of finding the expectation value for $\hat{A}(t)$ has been converted into the problem of finding the stationary value of $J$. This result also highlights the links between the forward Schrödinger and the backward Heisenberg pictures (2.27).

We are also free to write that

$$J_{st} = \text{Tr} \left[ \hat{A}(t) D(t) \right],$$ 

(2.39)

where $t$ is some arbitrary time. It is important to recognise that no assumptions have been made about $D(t)$ and $\hat{A}(t)$ and their allowed variations (except that they must satisfy the known boundary conditions). Furthermore, whilst the aim is to find the expectation value of $\hat{A}(t)$ at the time $t_1$ it is now clear that, by treating both the state of the system and the operator $\hat{A}(t)$ as time dependent, it is possible to write the result in terms of any arbitrary time with the times $t_0$ and $t_1$ entering the problem only through the boundary conditions.

2.3.5 Optimised Equations of Motion for Single-Particle Observables

We wish to consider the expectation values, correlations and fluctuations associated with a set of single-particle operators, $\hat{Q}_i$, evaluated at the time $t_1$ (2.16)

$$\hat{A} = \exp \left( - \sum_i c_i \hat{Q}_i \right),$$

(2.40)
where the operators $Q_{i}$ are written as the combination of a zero-body and a one-body operator.

\[ Q_{i} = q_{i} + Q_{i}, \]
\[ = q_{i} + \sum_{\alpha\beta} (Q_{i})_{\alpha\beta} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\beta}, \]  
(2.41)

and where $\hat{c}_{\alpha}$ and $\hat{c}_{\beta}$ are creation and annihilation operators in some single-particle basis.

As discussed previously it is now necessary to treat the operator $\hat{A}$ as time dependent and as such the derivations that follow will be carried out in terms of the time-dependent operator

\[ \hat{A}(t) = \exp \left( -\hat{L}(t) \right), \]  
(2.42)

where $\hat{L}(t)$ is a time-dependent single-particle operator of the form (2.41)

\[ \hat{L}(t) = l(t) + L(t), \]
\[ = l(t) + \sum_{\alpha\beta} L(t)_{\alpha\beta} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\beta}, \]  
(2.43)

with the operators of interest, $Q_{i}$, entering through the boundary conditions

\[ \hat{L}(t_{1}) = l(t_{1}) + \sum_{\alpha\beta} L(t_{1})_{\alpha\beta} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\beta}, \]
\[ = \sum_{i} \varepsilon_{i} q_{i} + \sum_{\alpha\beta} \sum_{i} \varepsilon_{i} (Q_{i})_{\alpha\beta} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\beta}, \]  
(2.44)

and where, due to these boundary conditions, $\hat{L}$ includes a built-in dependence upon the $\varepsilon$'s (which is linear at the time $t_{1}$). The other ingredient in our problem is the density matrix $D(t)$.

As discussed at the start of section 2.3 we are interested in single-particle operators so the density matrix is written as the exponential of a single-particle operator

\[ D(t) = \exp \left( -\hat{M}(t) \right), \]  
(2.45)

where $\hat{M}(t)$ is now a single-particle operator of the form (2.43)

\[ \hat{M}(t) = m(t) + M(t), \]  
(2.46)

but includes the Lagrange multipliers, $\mu_{i}$, of (2.15) and an $\varepsilon_{i}$ dependence since the density must now include a dependence upon the operators of interest (compare (2.12) and (2.15)). As is now customary it is assumed that the state of system at the initial time, $D(t_{0})$, is known.

In this notation the variational parameters are the variables $l(t)$ and $m(t)$ and the elements of the matrices $L(t)$ and $M(t)$. Whilst the relationship between $\hat{L}(t)$ and $\hat{Q}$ makes it useful to be able
to work in terms of these parameters it is also clear that the boundary conditions on the state of the system are more directly related to $D(t)$ than to $m(t)$ and $M(t)$.

$D(t)$ is the full many-body density matrix. However, in any realistic case it would be impossible to work directly in terms of $D(t)$ so we must use some simpler approximated form [41]. In practice we choose to work in terms of one-body density matrices. Following [47] we define the elements of the matrix of contractions (the one-body density matrix, (A.6)), $\rho(t)$

$$\rho_{\alpha\beta}(t) = \frac{1}{z(t)} \text{Tr} \left[ D(t) \delta_{\alpha\beta} \right],$$  

(2.47)

with the normalisation factor

$$z(t) = \text{Tr} [D(t)].$$

(2.48)

The boundary conditions now become

$$\rho_{\alpha\beta}(t_0) = \text{Tr} \left[ D(t_0) \delta_{\alpha\beta} \right],$$  

(2.49)

$$z(t_0) = 1,$$  

(2.50)

since it is assumed that the initial state, $D(t_0)$, is normalised. However, using a single-particle density matrix to describe the state of the system does not mean that the current approach reduces straight away to the TDHF approach since it is not assumed that the expectation values and fluctuations for the single-particle operators, $\hat{q}_i$, are given by the usual formulae (2.5, 2.6). We also do not assume that $\rho(t)$ is a Slater determinant.

The equations (2.47) and (2.48) can be written in terms of $m(t)$ and $M(t)$ [47]

$$z(t) = \exp \left( -m(t) + \text{Tr} \left[ \ln \left( 1 + e^{-M(t)} \right) \right] \right),$$  

(2.51)

$$\rho(t) = \frac{1}{e^{M(t)} + 1},$$  

(2.52)

with the inverse relationships

$$m(t) = -\ln(z(t)) - \text{Tr} \left[ \ln(1 - \rho(t)) \right],$$  

(2.53)

$$e^{-M(t)} = \rho(t) \frac{1}{1 - \rho(t)} = \frac{1}{1 - \rho(t)^2} \rho(t).$$  

(2.54)

Similarly, it is useful from a symmetry perspective to define, in addition to $l(t)$ and $L(t)$, the single-particle matrix

$$\sigma(t) = \frac{1}{e^{L(t)} + 1},$$  

(2.55)

and the normalisation factor

$$y(t) = \exp \left( -l(t) + \text{Tr} \left[ \ln \left( 1 + e^{-L(t)} \right) \right] \right),$$  

(2.56)
with the inverse relationships (from (2.53) and (2.54))

\[
\begin{align*}
l(t) &= -\ln (y(t)) - \text{Tr} \ln (1 - \sigma(t)), \\
e^{-L(t)} &= \sigma(t) \frac{1}{1 - \sigma(t)} = \frac{1}{1 - \sigma(t)} \sigma(t).
\end{align*}
\] (2.57, 2.58)

Collecting these terms together gives us two equivalent sets of variational parameters. The parameters \(l(t), L(t), m(t), \) and \(M(t)\) are clearly of operator form and easy to relate to the operators of interest, \(Q_i\), through the boundary conditions (2.44). However, the matrices \(\rho(t)\) and \(\sigma(t)\), and the normalisation factors \(z(t)\) and \(y(t)\), are easier to associate with the state of the system described by the one-body density matrix \(\rho(t)\) and hence with the usual mean-field techniques.

The characteristic function, (2.32, 2.33), contains terms with the products \(\hat{A}(t)D(t)\) and \(D(t)\hat{A}(t)\). These are evaluated using the fact that the product of the exponentials of single-particle operators can be written as the exponential of another single-particle operator \([47]\)

\[
\begin{align*}
\hat{A}(t)D(t) &= e^{-L(t)}e^{-\hat{M}(t)} = e^{-L'(t)}, \\
D(t)\hat{A}(t) &= e^{-\hat{M}(t)}e^{-L(t)} = e^{-\hat{M}'(t)},
\end{align*}
\] (2.59, 2.60)

where \(\hat{L}'(t)\) and \(\hat{M}'(t)\) are two single-particle operators of the form (2.43) with

\[
\begin{align*}
l'(t) &= m'(t) = m(t) + l(t), \\
e^{-L'(t)} &= e^{-L(t)}e^{-\hat{M}(t)}, \\
e^{-\hat{M}'(t)} &= e^{-\hat{M}(t)}e^{-L(t)}.
\end{align*}
\] (2.61, 2.62, 2.63)

Finally, as before, there are the two sets of contractions

\[
\begin{align*}
\rho'_{\alpha\beta}(t) &= \frac{\text{Tr} \left[ D(t) \hat{A}(t) \delta_{\beta\alpha} \right]}{\text{Tr} \left[ D(t) \hat{A}(t) \right]}, \\
\rho'(t) &= \frac{1}{e^{\hat{M}'(t)} + 1} = \frac{1}{e^{\hat{L}(t)}e^{\hat{M}(t)} + 1},
\end{align*}
\] (2.64, 2.65)

\[
\begin{align*}
\sigma'_{\alpha\beta}(t) &= \frac{\text{Tr} \left[ \hat{A}(t)D(t) \delta_{\beta\alpha} \right]}{\text{Tr} \left[ \hat{A}(t)D(t) \right]}, \\
\sigma'(t) &= \frac{1}{e^{\hat{L}'(t)} + 1} = \frac{1}{e^{\hat{M}(t)}e^{\hat{L}(t)} + 1}.
\end{align*}
\] (2.66, 2.67)
2.3 The Balian-Vénéroni Approach

These are associated with the normalisation factor

\[ w(t) = \text{Tr} \left[ \hat{A}(t) D(t) \right] = \text{Tr} \left[ D(t) \hat{A}(t) \right], \tag{2.68} \]

\[ = \exp \left( -L'(t) + \text{Tr} \left[ \ln \left( 1 + e^{-L'(t)} \right) \right] \right), \tag{2.69} \]

\[ = \exp \left( -m'(t) + \text{Tr} \left[ \ln \left( 1 + e^{-M'(t)} \right) \right] \right). \tag{2.70} \]

The Equations of Motion

To derive the equations of motion (2.25, 2.28) in terms of the new variational parameters the characteristic function (2.32, 2.33) should be re-written in terms of them. The characteristic function (2.32) includes a term of the form \( \text{Tr} \left[ A(t) \frac{dD(t)}{dt} \right] \). To evaluate this \( w(t) \) is re-written in terms of \( z(t), \rho(t), y(t) \) and \( \sigma(t) \). Using the definition of \( w(t) \), (2.70), and substituting in using (2.53, 2.54), (2.57, 2.58) and (2.61, 2.63)

\[ w(t) = \exp \left[ \ln \left( z(t) \right) + \text{Tr} \left[ \ln \left( 1 - \rho(t) \right) \right] + \ln \left( y(t) \right) + \text{Tr} \left[ \ln \left( 1 - \sigma(t) \right) \right] \right) \]

\[ \times \exp \left( \text{Tr} \left[ \ln \left( 1 + \frac{1}{1 - \rho(t)} \right) \rho(t) \sigma(t) \left( \frac{1}{1 - \sigma(t)} \right) \right] \right), \]

\[ = z(t) y(t) \exp \left( \text{Tr} \left[ \ln \left( 1 - \rho(t) - \sigma(t) + 2 \rho(t) \sigma(t) \right) \right] \right). \tag{2.71} \]

or, equivalently, by starting from (2.69)

\[ w(t) = y(t) z(t) \exp \left( \text{Tr} \left[ \ln \left( 1 - \sigma(t) - \rho(t) + 2 \sigma(t) \rho(t) \right) \right] \right). \tag{2.72} \]

The term \( \text{Tr} \left[ A(t) \frac{dD(t)}{dt} \right] \) is obtained by differentiating (2.72) assuming \( y(t) \) and \( \sigma(t) \) to be constant.

\[ \text{Tr} \left[ \hat{A}(t) \frac{dD(t)}{dt} \right] = \frac{w(t)}{z(t)} \frac{dz(t)}{dt} + w(t) \text{Tr} \left[ \frac{d}{d\rho} \left( \ln \left( 1 - \sigma(t) + 2 \rho(t) \rho(t) \right) \right) \frac{d\rho(t)}{dt} \right], \]

\[ = \frac{w(t)}{z(t)} \frac{dz(t)}{dt} \]

\[ + w(t) \text{Tr} \left[ \left( \frac{1}{1 - \sigma(t) - \rho(t) + 2 \sigma(t) \rho(t)} \right)^2 \frac{d\rho(t)}{dt} \right]. \tag{2.73} \]

The remaining term in (2.32) is the term containing the commutator and the Hamiltonian.

\[ -i \text{Tr} \left[ A(t) \left[ \hat{H}(t), D(t) \right] \right] = -i \text{Tr} \left[ D(t) A(t) \hat{H}(t) \right] + i \text{Tr} \left[ A(t) D(t) \hat{H}(t) \right], \tag{2.74} \]

where the Hamiltonian, \( \hat{H} \), takes its usual form [78] with one-body and two-body terms (A.10)

\[ \hat{H} = \sum_{\alpha \beta} t_{\alpha \beta} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\beta} + \frac{1}{4} \sum_{\alpha \beta \gamma \delta} \bar{v}_{\alpha \beta \gamma \delta} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\gamma}^{\dagger} \hat{c}_{\beta} \hat{c}_{\delta}. \]

This is obtained by recalling that the energy of a system described by the general density matrix, \( D(t) \), with the Hamiltonian, \( \hat{H}(t) \) is given by \( E(t) = \text{Tr} \left[ D(t) \hat{H}(t) \right] / \text{Tr} \left[ D(t) \right] \). If we choose to work in terms of the one-body density matrix \( \rho(t) \) then we must use, instead of the full Hamiltonian,
2.3 The Balian-Vénéroni Approach

\( H \), the one-body HF Hamiltonian, \( H_{HF} \), and this becomes (A.16)

\[
E(\rho(t)) = \frac{\text{Tr} [\rho(t) H_{HF}]}{z(t)},
\]

\[
= \sum_{\alpha\beta} \epsilon_{\alpha\beta} \rho_{\alpha\beta} + \frac{1}{2} \sum_{\alpha\beta\gamma\delta} \bar{v}_{\alpha\beta\gamma\delta} \rho_{\alpha\beta} \rho_{\gamma\delta}.
\]

Thus

\[
\text{Tr} [\hat{D}(t) \hat{H}] \rightarrow \text{Tr} [\rho(t) \hat{H}_{HF}] = z(t) E(\rho(t)).
\] (2.77)

We’ve already used the fact that the product of the exponentials of single-particle operators is also
the exponential of a single-particle operator (2.62, 2.63). Thus, \( D(t) \hat{A}(t) \) and \( \hat{A}(t) D(t) \) are of
the same form as \( D(t) \) and can be interpreted as modified versions of the density matrix, \( D(t) \),
normalised by \( w(t) \) and associated with the one-body density matrices \( \rho'(t) \) and \( \sigma'(t) \) respectively.
By analogy with (2.77)

\[
\text{Tr} [D(t) \hat{A}(t) \hat{H}(t)] \rightarrow w(t) E(\rho'(t)),
\] (2.78)

\[
\text{Tr} [\hat{A}(t) D(t) \hat{H}(t)] \rightarrow w(t) E(\sigma'(t)).
\] (2.79)

Substituting (2.73), (2.78) and (2.79) into (2.32)

\[
J = w(t_1) - \int_{t_0}^{t_1} dt \left( w(t) \left\{ iE(\rho'(t)) - iE(\sigma'(t)) + \frac{1}{z(t)} \frac{dz(t)}{dt} \right\} + \text{Tr} \left[ \left( \frac{1}{1 - \sigma(t) - \rho(t) + 2\sigma(t) \rho(t) (2\sigma(t) - 1)} \frac{d\rho(t)}{dt} \right) \right] \right).
\] (2.80)

The equations of motion are obtained by requiring this to be stationary with respect to arbitrary
variations in any of the variational parameters.

The Equations of Motion for \( z(t) \) and \( \rho(t) \)

Equation (2.80) includes the expressions \( \frac{dz(t)}{dt} \) and \( \frac{d\rho(t)}{dt} \) describing the dependence of \( J \)
on \( z(t) \) and \( \rho(t) \). All of the \( y(t) \) dependence is contained in the \( w(t) \) term in the integrand, which
is itself linear in \( y(t) \).

\[
\frac{dw(t)}{dy(t)} = \frac{w(t)}{y(t)}.
\] (2.81)

The variation in \( J \) due to a variation in \( y(t) \), \( \delta J_y \), is hence given by

\[
\delta J_y = - \int_{t_0}^{t_1} dt \left( \frac{w(t)}{y(t)} \delta y(t) \left\{ iE(\rho'(t)) - iE(\sigma'(t)) + \frac{1}{z(t)} \frac{dz(t)}{dt} \right\} + \text{Tr} \left[ \left( \frac{1}{1 - \sigma(t) - \rho(t) + 2\sigma(t) \rho(t) (2\sigma(t) - 1)} \frac{d\rho(t)}{dt} \right) \right] \right).
\] (2.82)
The first term has disappeared since \( y(t) \) is a variational parameter associated with the observable, \( A(t) \), which is fixed at the time \( t_i \) by the boundary conditions. For this to be true for any arbitrary variation in \( y, \delta y(t) \), the terms within the curly brackets must be zero. This condition requires that

\[
0 = iE(\rho'(t)) - iE(\sigma'(t)) + \frac{1}{z(t)} \frac{dz(t)}{dt} + \text{Tr} \left[ \left( \frac{1}{1 - \sigma(t) - \rho(t) + 2\sigma(t)\rho(t)(2\sigma(t) - 1)} \right) \frac{d\rho(t)}{dt} \right].
\] (2.83)

The matrix \( \sigma(t) \) enters \( J \) through \( w(t) \), the trace term and the terms \( E(\rho'(t)) \) and \( E(\sigma'(t)) \). The \( w(t) \) term can be neglected since it has already been eliminated by the requirements that \( \delta J_y = 0 \).

We consider the trace term first and introduce the additional single-particle matrix

\[
\eta(t) = \frac{1}{1 - \sigma(t) - \rho(t) + 2\sigma(t)\rho(t)(2\sigma(t) - 1)},
\] (2.84)

which allows us to relate a change in \( \sigma(t) \), \( \delta\sigma(t) \) to a change in \( \eta(t) \), \( \delta\eta(t) \)

\[
\delta\eta(t) = \frac{d\eta(t)}{d\sigma(t)}\delta\sigma(t),
\] (2.85)

where the explicit form of \( d\eta(t)/d\sigma(t) \) will not be required. The \( E(\rho'(t)) \) and \( E(\sigma'(t)) \) terms also depend on \( \sigma(t) \) through \( \rho'(t) \) and \( \sigma'(t) \). The variation in \( E(\rho'(t)) \) and \( E(\sigma'(t)) \) is given, using (2.75), by

\[
\delta E(\rho'(t)) = \text{Tr}[H_{HF}(\rho'(t))\delta \rho'(t)],
\] (2.86)

\[
\delta E(\sigma'(t)) = \text{Tr}[H_{HF}(\sigma'(t))\delta \sigma'(t)],
\] (2.87)

\( \eta(t) \) can be re-written to contain an explicit dependence on \( \rho'(t) \) and \( \sigma'(t) \).

\[
\eta(t) = \rho(t)^{-1}(\rho'(t) - \rho(t))(1 - \rho(t))^{-1},
\] (2.88)

\[
= (1 - \rho(t))^{-1}(\sigma'(t) - \rho(t))\rho(t)^{-1},
\] (2.89)

where all of the \( \sigma(t) \) dependence is contained within the \( \rho'(t) \) and \( \sigma'(t) \) terms. These alternate forms of (2.84) can be demonstrated by writing

\[
\rho'(t) = \frac{1}{\sigma(t)(1 - \sigma(t))(1 - \rho(t))\rho(t)^{-1}}.
\] (2.90)

and, similarly

\[
\sigma'(t) = \sigma(t)\frac{1}{1 - \rho(t) - \sigma(t) + 2\rho(t)\sigma(t)},
\] (2.91)

and then substituting into (2.88) and (2.89) and re-arranging. A change in either \( \rho'(t) \) or \( \sigma'(t) \)
due to a change in $\eta(t)$ (which is itself related to a change in $\sigma(t)$ through (2.85)) is then given by

$$
\delta \rho'(t) = \rho(t) \delta \eta(t) (1 - \rho(t)), \quad (2.92)
$$

$$
\delta \sigma'(t) = (1 - \rho(t)) \delta \eta(t) \rho(t). \quad (2.93)
$$

The variation in (2.83) in due to a variation in $\sigma(t)$ is then given by

$$
\text{Tr} \left[ \left( \frac{d\eta(t)}{d\sigma(t)} \delta \sigma(t) \right) \frac{d\rho(t)}{dt} \right] + \text{i} \delta E(\rho'(t)) - \text{i} \delta E(\sigma'(t)) = 0, \quad (2.94)
$$

where the $z(t)$ term has disappeared since it is independent of $\sigma(t)$. Substituting in using (2.86, 2.87) and (2.92, 2.93) and writing $\delta \eta(t) = (d\eta(t)/d\sigma(t)) \delta \sigma(t)$ this becomes

$$
\text{Tr} \left[ \delta \eta(t) \left( \frac{d\rho(t)}{dt} \right) \right] + \text{i} \text{Tr} [H_{HF}(\rho'(t)) \rho(t) \delta \eta(t) (1 - \rho(t))] - \text{i} \text{Tr} [H_{HF}(\sigma'(t)) (1 - \rho(t)) \delta \eta(t) \rho(t)] = 0, \quad (2.95)
$$

which we re-arrange to be obtain

$$
\text{Tr} \left[ \delta \eta(t) \left( \frac{d\rho(t)}{dt} + \text{i} (1 - \rho(t)) H_{HF}(\rho'(t)) \rho(t) - \text{i} \rho(t) H_{HF}(\sigma'(t)) (1 - \rho(t)) \right) \right] = 0. \quad (2.96)
$$

For this to be zero for arbitrary $\delta \eta(t)$ we require

$$
\text{i} \frac{d\rho(t)}{dt} = (1 - \rho(t)) H_{HF}(\rho'(t)) \rho(t) - \rho(t) H_{HF}(\sigma'(t)) (1 - \rho(t)), \quad (2.97)
$$

which we identify as our equation of motion for $\rho(t)$. Substituting back into (2.83) we obtain

$$
\frac{1}{z(t)} \frac{dz(t)}{dt} + \text{Tr} \left[ \eta(t) (1 - \rho(t)) H_{HF}(\rho'(t)) \rho(t) - \rho(t) H_{HF}(\sigma'(t)) (1 - \rho(t)) \right]
- E(\rho'(t)) + E(\sigma'(t)) = 0,
$$

which, using (2.88, 2.89), yields the equation of motion for $z(t)$

$$
\frac{1}{z(t)} \frac{dz(t)}{dt} = E(\rho'(t)) - E(\sigma'(t))
- \text{Tr} [(\rho'(t) - \rho(t)) H_{HF}(\rho'(t))]
+ \text{Tr} [(\sigma'(t) - \rho(t)) H_{HF}(\sigma'(t))]. \quad (2.98)
$$

Equations of Motion for $y(t)$ and $\sigma(t)$

A full derivation of the equations of motion for $y(t)$ and $\sigma(t)$ would start from (2.33) instead of (2.32) and follow the same steps as above using the boundary conditions on the state $D(t)$ rather than on the observable $A(t)$. However, the symmetry between $A(t)$ and $D(t)$ enables us to easily write down the equations of motion for $y(t)$ and $\sigma(t)$. From (2.97) and (2.99) we obtain, through
the interchange of $\rho(t)$ and $\sigma(t)$, and $\rho'(t)$ and $\sigma'(t)$

\[
\frac{d\sigma(t)}{dt} = (1 - \sigma(t)) H_{HF} (\sigma'(t)) \sigma'(t) - \sigma(t) H_{HF} (\rho'(t)) (1 - \sigma(t)), \tag{2.100}
\]

\[
\frac{dy(t)}{y(t)} = E(\sigma'(t)) - E(\rho'(t)) - \text{Tr} [H_{HF}(\sigma'(t)) - H_{HF}(\rho'(t))]. \tag{2.101}
\]

This gives us a set of coupled differential equations that should be solved for $y(t)$, $z(t)$, $\sigma(t)$ and $\rho(t)$ subject to the boundary conditions (2.44), (2.49) and (2.50).

**Equations of Motion for $m(t)$, $l(t)$, $M(t)$ and $L(t)$**

We can also write down a set of equations of motion for the variational parameters $m(t)$, $l(t)$, $M(t)$ and $L(t)$. From (2.53) we obtain

\[
\frac{dm(t)}{dt} = -\frac{1}{z(t)} \frac{dz(t)}{dt} - \text{Tr} \left[ -\frac{1}{1 - \rho(t)} \frac{d\rho(t)}{dt} \right]. \tag{2.102}
\]

Substituting in using (2.97) and (2.99) we obtain

\[
\frac{dm(t)}{dt} = -E(\rho'(t)) + E(\sigma'(t)) + \text{Tr} [\rho'(t) H_{HF}(\sigma'(t))] - \text{Tr} [\rho(t) H_{HF}(\rho'(t))] - \text{Tr} [\rho'(t) H_{HF}(\sigma'(t))], \tag{2.103}
\]

Similarly

\[
\frac{dl(t)}{dt} = E(\rho'(t)) - E(\sigma'(t)) + \text{Tr} [\rho'(t) H_{HF}(\rho'(t))] + \text{Tr} [\rho(t) H_{HF}(\sigma'(t))]. \tag{2.104}
\]

Likewise from (2.54) we obtain

\[
\frac{de^{-M(t)}}{dt} = -\frac{1}{1 - \rho(t)} \frac{d\rho(t)}{dt} \frac{1}{1 - \rho(t)} \rho(t) + \frac{1}{1 - \rho(t)} \frac{d\rho(t)}{dt}, \tag{2.105}
\]

which gives

\[
\frac{de^{-M(t)}}{dt} = H_{HF}(\rho'(t)) \rho(t) \frac{1}{1 - \rho(t)} - \frac{1}{1 - \rho(t)} \rho(t) H_{HF}(\sigma'(t)), \tag{2.106}
\]

\[
= H_{HF}(\rho'(t)) e^{-M(t)} - e^{-M(t)} H_{HF}(\sigma'(t)), \tag{2.106}
\]
and similarly

\[ \frac{d e^{-L(t)}}{dt} = H_{HF}(\sigma'(t)) e^{-L(t)} - e^{-L(t)} H_{HF}(\rho'(t)). \]  

(2.107)

These equations of motion could be written wholly in terms of \( l(t), m(t), L(t) \) and \( M(t) \) using the appropriate substitutions if required.

### 2.3.6 Equations of motion for \( m'(t), M'(t), l'(t) \) and \( L'(t) \)

Using the original definitions of \( m'(t) \) and \( M'(t) \), (2.61, 2.63) we obtain,

\[ \frac{dm'(t)}{dt} = \frac{dm(t)}{dt} + \frac{dl(t)}{dt} = 0, \]  

(2.108)

and

\[ \frac{de^{-M'(t)}}{dt} = \frac{de^{-M(t)}}{dt} - e^{-L(t)} + e^{-M(t)} \frac{de^{-L(t)}}{dt}, \]

\[ = H_{HF}(\rho'(t)) e^{-M'(t)} - e^{-M'(t)} H_{HF}(\rho'(t)). \]  

(2.109)

Whilst we will not require them we note, for completeness, that, using (2.57) and (2.58)

\[ \frac{dl'(t)}{dt} = \frac{dm'(t)}{dt} = 0, \]  

(2.110)

\[ \frac{de^{-L'(t)}}{dt} = \frac{de^{-M'(t)}}{dt} = H_{HF}(\rho'(t)) e^{-M'(t)} - e^{-M'(t)} H_{HF}(\rho'(t)). \]  

(2.111)

It has already been shown that the stationary value of the characteristic function is given by (2.39) which we now write as

\[ J_{st} = \text{Tr} [A(t) D(t)] = w(t). \]  

(2.112)

This can be reaffirmed using the equations of motion for \( m(t), l(t), M(t) \) and \( L(t) \), (2.103), (2.104), (2.106) and (2.107) respectively. From the definition of \( w(t) \) (2.68)

\[ \frac{dw(t)}{dt} = -e^{-m'(t)} \frac{dm'(t)}{dt} \text{Tr} [\ln(1 + e^{-M'(t)})] \]

\[ + e^{-m'(t)} \text{Tr} [\ln(1 + e^{-M'(t)})] \text{Tr} \left[ \frac{1}{1 + e^{-M'(t)}} \frac{de^{-M'(t)}}{dt} \right], \]

\[ = e^{-m'(t)} \text{Tr} [\ln(1 + e^{-M'(t)})] \text{Tr} [H_{HF}(\rho'(t)) \rho'(t) - H_{HF}(\rho'(t)) \rho'(t)], \]

\[ = 0. \]  

(2.113)

### 2.3.7 Equations of Motion for \( \rho'(t) \) and \( \sigma'(t) \)

Whilst deriving these equations of motion it was convenient to work in terms of the variational parameters, \( y(t), z(t), \sigma(t) \) and \( \rho(t) \). However, to make clearer the links with the physical problem it is now advantageous to work with, instead of \( y(t) \) and \( \sigma(t) \), \( l(t) \) and \( L(t) \) since these are more obviously related to our observables of interest through the boundary conditions (2.44). For the
same reason we will continue to use the parameters $z(t)$ and $\rho(t)$, and not $m(t)$ and $M(t)$, since they are more directly associated with the state of the system and the boundary conditions (2.49, 2.50). Written in terms of these parameters $w(t)$ becomes, instead of (2.68)

$$w(t) = e^{-M(t)} e^{-m(t)} \exp \left( \operatorname{Tr} \left[ \ln \left( 1 + e^{-M(t)} e^{-L(t)} \right) \right] \right),$$

using (2.65,2.67). Similarly

$$\sigma'(t) = \frac{1}{e^{M(t)} e^{L(t)} + 1},$$

From (2.65) we obtain

$$1 - \rho'(t) = \frac{1}{e^{-M'(t)} + 1},$$

such that

$$\frac{1}{dt} \rho'(t) = (-1) \frac{1}{e^{-M'(t)} + 1} \frac{de^{-M'(t)}}{dt} \frac{1}{e^{-M'(t)} + 1},$$

using (2.109) and

$$e^{-M'(t)} = \rho'(t) \frac{1}{1 - \rho'(t)} \frac{1}{1 - \rho'(t)} \rho'(t).$$

Similarly, from (2.115) we obtain

$$\frac{1}{dt} \sigma'(t) = \frac{1}{dt} \rho'(t) e^{L(t)} + i e^{-L(t)} \frac{d}{dt} \rho'(t) e^{L(t)} + i e^{-L(t)} \rho'(t) \frac{d}{dt} \left( \frac{1}{e^{-L(t)}} \right),$$

using (2.107) and (2.116). We note that these equations both have the same form as the TDHF equation (B.11) although they depend on both the observables of interest and the state of the system and must therefore satisfy the mixed boundary conditions (2.44, 2.49, 2.50).
2.3 The Balian-Vénéroni Approach

2.3.8 Expansion in terms of $\varepsilon_i$

It has previously been shown in section 2.3.4 that there are advantages to considering the second characteristic function, $\ln(\psi_{st})$, whose stationary value, $\ln(\psi_{st})$, is given by

$$\psi_{st} = \ln(\psi_{st}) = \ln (\omega(t)),$$

$$= -l(t) + \ln(z(t)) + \text{Tr} \left[ \ln \left( 1 - \rho(t) \left( 1 - e^{-L(t)} \right) \right) \right], \quad (2.119)$$

where we now define $\psi_{st} = \ln(\psi_{st})$. Further simplifications can be obtained by considering, instead of the characteristic function, its derivative with respect to one of the parameters, $\varepsilon_i$. In this case (2.19), which relates $\psi_{st}$ to the expectation values, fluctuations and correlations of the set of single-particle observables $\hat{Q}_i$ (and the parameters $\varepsilon_i$), reduces to

$$\frac{d\psi_{st}}{d\varepsilon_i} = -\langle \hat{Q}_i \rangle + \sum_m \varepsilon_mC_{im} + \ldots, \quad (2.120)$$

where $C_{im}$ is the correlation between the two single-particle observables $\hat{Q}_i$ and $\hat{Q}_m$, or, for $\hat{Q}_i = \hat{Q}_m$, the fluctuation in this single observable. To compare (2.119) and (2.120) we differentiate (2.119) with respect to $\varepsilon_i$. The $\varepsilon_i$ dependence in $\psi_{st}$ enters through $l(t)$ and $L(t)$

$$\frac{dl(t)}{d\varepsilon_i} + \text{Tr} \left[ \frac{1}{1 - \rho(t) + \rho(t)e^{-L(t)}\rho(t)} \frac{de^{-L(t)}}{d\varepsilon_i} \right],$$

$$= -\frac{dl(t)}{d\varepsilon_i} + \text{Tr} \left[ \left( \frac{1}{\rho(t)} \right) \left( 1 - \rho(t) + \rho(t) e^{-L(t)} \right) \right]^{-1} \frac{de^{-L(t)}}{d\varepsilon_i},$$

$$= -\frac{dl(t)}{d\varepsilon_i} + \text{Tr} \left[ \rho(t) e^{-L(t)} \frac{de^{-L(t)}}{d\varepsilon_i} \right]. \quad (2.121)$$

In the current approach the state of the system includes a dependence on the observables of interest, and hence on $\varepsilon$. We therefore expand $\rho(t)$ in terms of $\varepsilon$ and write

$$\rho(t) = \rho^{(0)}(t) + \rho^{(1)}(t) + \ldots, \quad (2.122)$$

where the bracketed superscript denotes the power of $\varepsilon$ in the expansion. Similarly

$$l(t) = l^{(1)}(t) + \ldots, \quad (2.123)$$

$$L(t) = L^{(1)}(t) + \ldots, \quad (2.124)$$

where $l(t)$ and $L(t)$ have no component independent of $\varepsilon$ as a consequence of their boundary conditions (2.44). We use these expansions of $l(t)$, $L(t)$ and $\rho(t)$ to expand $\rho'(t)$ in terms of $\varepsilon$ using (2.65) and keeping terms up to first order in $\varepsilon$ we obtain

$$\rho'(t) = \left( 1 + L^{(1)}(t) \right) \left( 1 - \left( \rho^{(0)}(t) + \rho^{(1)}(t) \right) \right) \frac{1}{\rho^{(0)}(t) + \rho^{(1)}(t)} + 1 \left( 1 - \rho^{(0)}(t) \right), \quad (2.125)$$
which we separate into terms which are independent of $\epsilon$ and terms which are linearly dependent upon $\epsilon$

\[
\rho' (t) = \rho'^{(0)} (t) + \rho'^{(1)} (t),
\]
\[
\rho'^{(0)} (t) = \rho^{(0)} (t),
\]
\[
\rho'^{(1)} (t) = \rho^{(1)} (t) - \rho^{(0)} (t) L^{(1)} (t) \left( 1 - \rho^{(0)} (t) \right).
\]

Performing a similar expansion on $\sigma' (t)$ using (2.67) we obtain

\[
\sigma' (t) = \left( \frac{1}{\rho^{(0)} (t) + \rho^{(1)} (t)} \right) \left( 1 - \left( \rho^{(0)} (t) + \rho^{(1)} (t) \right) \right) \left( 1 + L^{(1)} (t) \right) + 1 \right)^{-1},
\]
\[
= \rho^{(0)} (t) + \rho^{(1)} (t) - \left( 1 - \rho^{(0)} (t) \right) L^{(1)} (t) \rho^{(0)} (t),
\]

such that

\[
\sigma' (t) = \sigma'^{(0)} (t) + \sigma'^{(1)} (t),
\]
\[
\sigma'^{(0)} (t) = \rho^{(0)} (t) = \rho'^{(0)} (t),
\]
\[
\sigma'^{(1)} (t) = \rho^{(1)} (t) - \left( 1 - \rho^{(0)} (t) \right) L^{(1)} (t) \rho^{(0)} (t).
\]

2.3.9 Expectation Values for Single-Particle Operators

We now expand (2.121) at the time $t_i$ using the boundary conditions (2.44). Keeping only those terms which are independent or linearly dependent upon $\epsilon$ we obtain

\[
\frac{d\psi_{st}}{dt} \bigg|_{t = t_i} = -q_i + \text{Tr} \left[ \left( \rho^{(0)} (t_i) + \rho^{(1)} (t_i) - \rho^{(0)} (t_i) \left( \sum_p \epsilon_p Q_p \right) \left( 1 - \rho^{(0)} (t_i) \right) \right) \times \left( 1 + \sum_n \epsilon_n Q_n \right) \left( -Q_i + \frac{1}{2} \sum_m \epsilon_m (Q_i Q_m + Q_m Q_i) \right) \right].
\]

We now see the benefits of considering the derivative of $\psi_{st}$ since the correlations, $C_{im}$, which were originally dependent on $\epsilon$ to second order, can now be calculated using the single-particle matrix $\rho^{(1)} (t)$, which is only first order in $\epsilon$.

Extracting only those terms which are independent of $\epsilon$ and comparing with (2.120) we obtain,

\[
\langle \hat{Q}_i \rangle = q_i + \text{Tr} \left[ \rho^{(0)} (t_i) Q_i \right],
\]

where we recognise the usual form for the expectation value and which depends solely on the $\epsilon$ independent contribution to the single-particle density, $\rho^{(0)} (t_i)$. Performing a similar expansion on (2.116) and keeping only those terms which are independent of $\epsilon$ we obtain the equation of motion for $\rho^{(0)} (t_i)$

\[
\frac{i}{\hbar} \frac{d\rho^{(0)} (t)}{dt} = \left[ H_{HF}, \rho^{(0)} (t) \right].
\]
2.3 The Balian-Vénéroni Approach

Whilst the equations of motion for \( \rho(t) \) and \( \sigma(t) \), (2.97) and (2.100), were coupled equations through the mixing of \( \rho(t) \) and \( \sigma(t) \) in \( \rho'(t) \) and \( \sigma'(t) \) this equation is totally independent of the observables of interest and depends solely on the state of the system as described by the single-particle density matrix \( \rho^{(0)}(t) \). If it is further assumed that the single-particle matrix, \( \rho^{(0)}(t) \), must be a Slater determinant then this equation becomes the standard TDHF equation (B.11). This shows that, subject to the assumption that the state of the system can, at all times, be described by a Slater determinant, the expectation value for a single-particle operator should be evaluated using the usual expression for an expectation value where the time-dependence of the Slater determinant describing the state of the system is given by the standard TDHF equation. The usual TDHF approach has been recovered as a special case of our more general theory.

2.3.10 Correlations and Fluctuations for Single-Particle Operators

Equation (2.120) tells us that the correlations and fluctuations for the single-particle operators, \( Q_i \), are obtained by looking at the terms in (2.131) which depend linearly upon \( \varepsilon \). From (2.131) we obtain

\[
\sum_m \varepsilon_m C_{im} = \text{Tr} \left[ \rho^{(0)}(t_1) \left( \sum_m \varepsilon_m Q_m \right) (-Q_i) \right] + \text{Tr} \left[ \rho^{(0)}(t_1) \left( \frac{1}{2} \sum_m \varepsilon_m (Q_i Q_m + Q_m Q_i) \right) \right] + \text{Tr} \left[ \rho^{(1)}(t_1) (-Q_i) \right] \]

(2.134)

where we've changed some of the subscripts to ensure consistency between the terms. This can be rearranged to obtain

\[
\sum_m \varepsilon_m C_{im} = \sum_m \varepsilon_m \text{Tr} \left[ \frac{1}{2} \rho^{(0)}(t_1) (Q_i Q_m - Q_m Q_i) + \rho^{(0)}(t_1) Q_m \left( 1 - \rho^{(0)}(t_1) \right) \right] - \text{Tr} \left[ \rho^{(1)}(t_1) Q_i \right] + \text{Tr} \left[ \rho^{(0)}(t_1) Q_m \left( 1 - \rho^{(0)}(t_1) \right) \right]
\]

(2.135)

The single-particle matrix \( \rho^{(1)}(t) \) is the contribution to \( \rho(t) \) which is linearly dependent on the \( \varepsilon \) parameters however we can further expand this into the contributions due to each individual \( \varepsilon_m \)

\[
\rho^{(1)}(t) = \sum_m \varepsilon_m \rho^{(1)}_m (t),
\]

(2.136)

which allows us to re-write (2.135) as the standard TDHF result with an extra correction term

\[
\sum_m \varepsilon_m C_{im} = \sum_m \varepsilon_m \left[ (C_{im})_{TDHF} - \text{Tr} \left[ \rho^{(1)}_m (t_1) Q_i \right] \right].
\]

(2.137)
This is easier to recognise if we consider the fluctuation in a single observable, \( Q_m = Q_i \). In this case the commutator term in (2.135) disappears and we are left with

\[
C_{ii} = \text{Tr} \left[ Q_i \rho^{(0)} (t_1) Q_i \left( 1 - \rho^{(0)} (t_1) \right) \right] - \text{Tr} \left[ \rho_i^{(1)} (t_1) Q_i \right],
\]

\[
= (\Delta Q_i)_{BV}^2 = (\Delta Q_i)_{TDHF}^2 - \text{Tr} \left[ \rho_i^{(1)} (t_1) Q_i \right], \tag{2.138}
\]

which we recognise as the standard TDHF result (2.6) with a correction term which depends upon the observable of interest, \( Q_i \), and, linearly, on \( \epsilon_i \) through \( \rho_i^{(1)} (t_1) \). To evaluate fluctuation, \((\Delta Q_i)_{BV}\), using (2.138) we would still need to calculate \( \rho_i^{(1)} (t_1) \) which would not be trivial and would certainly require the solution of coupled equations. Instead, we can look for a way to re-write (2.138) so that it is easier to implement.

For consistency with (2.136) we define the additional expansion

\[
L_{m}^{(1)} (t) = \sum_{m} \epsilon_m L_m^{(1)} (t), \tag{2.139}
\]

where the boundary condition (2.44) provides \( L_m^{(1)} (t_1) = Q_m \). Similarly

\[
\rho_m^{(1)} (t) = \sum_{m} \epsilon_m \rho_m^{(1)} (t), \tag{2.140}
\]

and

\[
\sigma_m^{(1)} (t) = \sum_{m} \epsilon_m \sigma_m^{(1)} (t). \tag{2.141}
\]

Using these equation we write, from (2.127)

\[
\rho_m^{(1)} (t) = \rho_m^{(1)} (t) - \rho^{(0)} (t) L_m^{(1)} (t) \left( 1 - \rho^{(0)} (t) \right), \tag{2.142}
\]

whilst (2.130) gives us

\[
\sigma_m^{(1)} (t) = \sigma_m^{(1)} (t) - \left( 1 - \rho^{(0)} (t) \right) L_m^{(1)} (t) \rho^{(0)} (t). \tag{2.143}
\]

The result (2.135) is true at the time \( t_1 \) but we can also write more generally that

\[
F_{m} (t) = \frac{1}{2} \text{Tr} \left[ \rho^{(0)} (t) \left[ L_m^{(1)} (t), L_m^{(1)} (t) \right] \right] + \text{Tr} \left[ L_i^{(1)} (t) \rho^{(0)} (t) L_m^{(1)} (t) \left( 1 - \rho^{(0)} (t) \right) \right]
- \text{Tr} \left[ \rho_m^{(1)} (t) L_i^{(1)} (t) \right], \tag{2.144}
\]
such that \( F_{im} (t_1) = C_{im} \) and, using (2.142) and (2.143)

\[
F_{im} (t) = \frac{1}{2} \text{Tr} \left[ \rho^{(0)} (t) L_i^{(1)} (t) L_m^{(1)} (t) - \rho^{(0)} (t) L_m^{(1)} (t) L_i^{(1)} (t) + 2 \rho^{(0)} (t) L_m^{(1)} (t) L_i^{(1)} (t) \right] - 2 \rho^{(0)} (t) L_i^{(1)} (t) \rho^{(0)} (t) L_m^{(1)} (t) - 2 \rho^{(1)} (t) L_i^{(1)} (t) \\
= \frac{1}{2} \text{Tr} \left[ -L_i^{(1)} \left( \rho^{(1)} (t) - L_m^{(1)} (t) \rho^{(0)} (t) L_m^{(1)} (t) \rho^{(0)} + \rho^{(1)} (t) - \rho^{(0)} (t) L_m^{(1)} (t) + \rho^{(0)} (t) L_m^{(1)} (t) \rho^{(0)} \right) \right] \\
= - \frac{1}{2} \text{Tr} \left[ L_i^{(1)} (t) \left( \sigma^{(1)} (t) + \rho^{(1)} (t) \right) \right].
\] (2.145)

Using this simplified form we now consider the time dependence of \( F_{im} (t) \).

We consider the time dependence of \( \rho^{(1)} (t) \) first starting from (2.116)

\[
i \frac{d \rho^{(1)} (t)}{dt} = [H_{HF} (\rho^{(0)} (t)) , \rho^{(1)} (t)] ,
\]

where \( H_{HF} \) is the usual HF Hamiltonian given by (A.24). We now substitute in using the expansion \( \rho^{(0)} (t) = \rho^{(0)} (t) + \rho^{(1)} (t) \), where \( \rho^{(0)} (t) = \rho^{(0)} (t) \) (2.126), and expanding to first order in \( \varepsilon \) we obtain

\[
i \frac{d \rho^{(0)} (t)}{dt} + i \frac{d \rho^{(1)} (t)}{dt} = \left[ K + \text{Tr}_2 \left[ \bar{\eta} \rho^{(0)} \right] + \text{Tr}_2 \left[ \bar{\eta} \rho^{(1)} \right] , \rho^{(0)} + \rho^{(1)} \right] \\
= \left[ K + \text{Tr}_2 \left[ \bar{\eta} \rho^{(0)} \right] , \rho^{(0)} \right] + \left[ \text{Tr}_2 \left[ \bar{\eta} \rho^{(1)} \right] , \rho^{(0)} \right] + \left[ K + \text{Tr}_2 \left[ \bar{\eta} \rho^{(0)} \right] , \rho^{(1)} \right] \\
\]

\[
i \frac{d \rho^{(1)} (t)}{dt} = \left[ H_{HF} (\rho^{(0)} ) , \rho^{(1)} \right] + \left[ \text{Tr}_2 \left[ \bar{\eta} \rho^{(1)} \right] , \rho^{(0)} \right] 
\]

(2.146)

where \( \text{Tr}_2 [\ldots] \) denotes a partial trace on the second particle. Finally, using (2.140) we obtain

\[
i \frac{d \rho^{(1)} (t)}{dt} = \left[ H_{HF} (\rho^{(0)} ) , \rho^{(1)} \right] + \left[ \text{Tr}_2 \left[ \bar{\eta} \rho^{(1)} \right] , \rho^{(0)} \right] 
\]

(2.147)

which we recognise from RPA linear response theory [78, chap. 8.5] as having the same form as the equation of motion for a small, time-dependent, first order correction, \( \Delta \rho \) to a constant density, \( \rho \) due to a small external force.

We repeat this for \( \sigma^{(1)} (t) \) starting from (2.118) and using (2.129)

\[
i \frac{d \sigma^{(1)} (t)}{dt} = [H_{HF} (\sigma^{(0)} ) , \sigma^{(1)} ] ,
\]

we obtain

\[
i \frac{d \sigma^{(1)} (t)}{dt} = \left[ H_{HF} (\rho^{(0)} ) , \sigma^{(1)} \right] + \left[ \text{Tr}_2 \left[ \bar{\eta} \sigma^{(1)} \right] , \rho^{(0)} \right] ,
\]

(2.148)

which becomes, using (2.141)

\[
i \frac{d \sigma^{(1)} (t)}{dt} = \left[ H_{HF} (\rho^{(0)} ) , \sigma^{(1)} \right] + \left[ \text{Tr}_2 \left[ \bar{\eta} \sigma^{(1)} \right] , \rho^{(0)} \right] ,
\]

(2.149)
which is identical in form to (2.147). Finally we must consider the time-dependence of \( L_i^{(1)}(t) \). We start from (2.107), which we re-write using (2.115)

\[
\begin{align*}
\frac{i}{2} \frac{d e^{-L}}{dt} & = H_{HF}(\sigma') e^{-L} - e^{-L} H_{HF}(\rho') = -ie^{-L} \frac{dL}{dt}, \\
\frac{i}{2} \frac{dL}{dt} & = H_{HF}(\rho') - e^{L} H_{HF}(\sigma') e^{-L}. \tag{2.150}
\end{align*}
\]

We expand this keeping only those terms which are linear in \( \epsilon \)

\[
\frac{d L_i^{(1)}}{dt} = H_{HF}\left(\rho^{(0)} + \rho^{(1)}\right) - \left(1 + L^{(1)}\right) \left[ K + Tr_2 \left[ \tilde{\theta} \left(1 - L^{(1)}\right) \left(\rho^{(0)} + \rho^{(1)}\right) \left(1 + L^{(1)}\right)\right] \right] \left(1 - L^{(1)}\right),
\]

\[
= -L^{(1)} H_{HF}(\rho^{(0)}) + H_{HF}(\rho^{(0)}) L^{(1)} + Tr_2 \left[ \tilde{\theta} L^{(1)}(\rho^{(0)}) \right] - Tr_2 \left[ \tilde{\theta} \rho^{(0)} L^{(1)} \right],
\]

\[
= \left[ H_{HF}(\rho^{(0)}), L^{(1)} \right] + Tr_2 \left[ \tilde{\theta} \left[ L^{(1)}, \rho^{(0)} \right] \right]. \tag{2.151}
\]

Substituting in using (2.139) we then obtain

\[
\frac{d L_i^{(1)}}{dt} = \left[ H_{HF}(\rho^{(0)}), L_i^{(1)} \right] + Tr_2 \left[ \tilde{\theta} \left[ L_i^{(1)}, \rho^{(0)} \right] \right]. \tag{2.152}
\]

We now use these equations to consider the time dependence of \( F_{im}(t) \). We write

\[
\frac{d F_{im}(t)}{dt} = -\frac{1}{2} Tr_2 \left[ \frac{d L_i^{(0)}}{dt} \rho_m^{(1)} + L_i^{(1)} \frac{d \rho_m^{(1)}}{dt} \right] - \frac{1}{2} Tr_2 \left[ \frac{d L_i^{(0)}}{dt} \sigma_m^{(1)} + L_i^{(1)} \frac{d \sigma_m^{(1)}}{dt} \right]. \tag{2.153}
\]

Since equations (2.147) and (2.149) are identical in form we only need to consider the first term since the second term can be obtained by replacing \( \rho_m^{(1)} \) by \( \sigma_m^{(1)} \).

\[
\frac{d}{dt} \left( Tr \left[ L_i^{(1)} \rho_m^{(1)} \right] \right) = Tr \left[ \left[ H_{HF}(\rho^{(0)}), L_i^{(1)} \right] \rho_m^{(1)} + Tr_2 \left[ \tilde{\theta} \left[ L_i^{(1)}, \rho^{(0)} \right] \right] \rho_m^{(1)} \right. \\
\left. + L_i^{(1)} \left[ H_{HF}(\rho^{(0)}), \rho_m^{(1)} \right] + L_i^{(1)} \left[ Tr_2 \left[ \tilde{\theta} \rho_m^{(1)} \right] \rho^{(0)} \right] \right]. \tag{2.154}
\]

The first and third terms (with the explicit dependence on \( H_{HF} \)) cancel each other out leaving

\[
\frac{d}{dt} \left( Tr \left[ L_i^{(1)} \rho_m^{(1)} \right] \right) = Tr \left[ Tr_2 \left[ \tilde{\theta} \left( L_i^{(1)} (\rho^{(0)}) - \rho^{(0)} L_i^{(1)} \right) \right] \rho_m^{(1)} \right. \\
\left. + \left( L_i^{(1)} \left[ Tr_2 \left[ \tilde{\theta} \rho_m^{(1)} \right] \rho^{(0)} \right] - \rho^{(0)} Tr_2 \left[ \tilde{\theta} \rho_m^{(1)} \right] \right) \right], \tag{2.155}
\]

which also cancel out as can be seen by writing out these terms in full in terms of the matrix elements of \( \tilde{\theta}, L_i^{(1)}, \rho^{(0)} \) and \( \rho_m^{(1)} \). The first term becomes

\[
Tr \left[ Tr_2 \left[ \tilde{\theta} \left( L_i^{(1)} (\rho^{(0)}) - \rho^{(0)} L_i^{(1)} \right) \right] \rho_m^{(1)} \right] = \sum_{abde} \tilde{v}_{cabd} \left( L_i^{(1)} \right)_{de} \left( \rho^{(0)} \right)_{ec} \left( \rho_m^{(1)} \right)_{ba} \\
- \sum_{abde} \tilde{v}_{cabd} \left( \rho^{(0)} \right)_{de} \left( L_i^{(1)} \right)_{ec} \left( \rho_m^{(1)} \right)_{ba}. \tag{2.156}
\]
whilst the second term becomes

\[
\text{Tr} \left[ L_i^{(1)} \left( \text{Tr}_2 \left[ \delta \rho_m^{(1)} \rho^{(0)} - \rho^{(0)} \text{Tr}_2 \left[ \delta \rho_m^{(1)} \right] \right] \right) \right] = \sum_{abcde} \left( L_i^{(1)} \right)_{ea} \delta_{coab} \left( \rho_m^{(1)} \right)_{dc} \left( \rho^{(0)} \right)_{be} - \sum_{abcde} \left( L_i^{(1)} \right)_{be} \left( \rho^{(0)} \right)_{ea} \delta_{coab} \left( \rho_m^{(1)} \right)_{dc}
\]

It is now possible to see that, using \( \delta_{coab} = \delta_{coab} \), these terms cancel each other out. If we now take into account the fact that the equation of motion for \( \sigma_m^{(1)} \) has exactly the same form as the equation of motion for \( \rho_m^{(1)} \), then we see that

\[
\frac{dF_{im}(t)}{dt} = 0.
\]

This means that \( F_{im}(t) \) is constant in time and can be evaluated at any time. We can therefore choose to evaluate it at the time \( t_0 \) where our boundary conditions require that the state of the system is known (and independent of \( \varepsilon \) such that \( \rho_m^{(1)}(t_0) = 0 \)). This eliminates the last term from (2.144) leaving

\[
C_{im} = \frac{1}{2} \left[ \rho^{(0)} \left( L_i^{(1)} \right) - 2 \rho^{(0)} L_i^{(1)} \rho^{(0)} L_m^{(1)} \right],
\]

If we now make the additional assumption that the initial state, \( \rho(t_0) \), is a Slater determinant (satisfying \((\rho^{(0)})^2 = \rho^{(0)}\)) then this can be re-written as

\[
C_{im} = -\frac{1}{2} \left[ \rho^{(0)} L_i^{(1)} L_m^{(1)} - \rho^{(0)} L_i^{(1)} \rho^{(0)} L_m^{(1)} + L_i^{(1)} \rho^{(0)} L_i^{(1)} L_m^{(1)} - \rho^{(0)} L_i^{(1)} \rho^{(0)} L_m^{(1)} \right],
\]

From the original earlier definitions of \( \rho_m^{(1)} \) and \( \sigma_m^{(1)} \) (2.127, 2.130) we know that

\[
\rho_m^{(1)} - \sigma_m^{(1)} = \left[ L_m^{(1)} , \rho^{(0)} \right].
\]

This tells us that the single-particle matrix \( \left[ L_m^{(1)} , \rho^{(0)} \right] \) satisfies an equation of motion with the same form as those satisfied by \( \rho_m^{(1)} \) and \( \sigma_m^{(1)} \) (2.147) and (2.149) respectively). \( \rho_m^{(1)} \) and \( \sigma_m^{(1)} \) are the first order (in \( \varepsilon_m \)) contributions to the single-particle matrices \( \rho'(t) \) and \( \sigma'(t) \) which means that \( \left[ L_m^{(1)}(t) , \rho^{(0)}(t) \right] \) can be regarded as the first order contribution to the new single-particle matrix, \( \eta_m(t, \varepsilon_m) \), defined as

\[
\eta_m(t, \varepsilon_m) = \rho^{(0)}(t) + \varepsilon_m \left[ L_m^{(1)}(t) , \rho^{(0)}(t) \right],
\]

where, as with \( \rho'(t) \) and \( \sigma'(t) \), the zeroth order contribution is \( \rho^{(0)}(t) \). Both the zeroth and first order terms in \( \eta_m(t, \varepsilon_m) \) obey the same equations of motion as the relevant components in \( \rho'(t) \) and \( \sigma'(t) \). The time evolution of \( \eta_m(t, \varepsilon_m) \) must be given by an equation of motion with the same
form as the equations of motion for \( \rho'(t) \) and \( \sigma'(t) \). This equation has the same form as the TDHF equation.

For small values of \( \varepsilon_m \) (2.162) can be written as

\[
\eta_m(t, \varepsilon_m) \approx \exp(i\varepsilon_m L_m(t)) \rho^{(0)}(t) \exp(-i\varepsilon_m L_m(t)),
\]

(2.163)

from which we see that, because \( \rho^{(0)}(t) \) is a Slater determinant, and because of the \( "i" \) constant in (2.162), the single-particle matrix \( \eta_m(t, \varepsilon_m) \) must also be a Slater determinant.

In linear response theory we consider the response of the system (change in the state, \( \rho \)) due to an infinitely small external force [78, pg. 317]. This is analogous to the current problem in which we are interested in the contributions to the single-particle matrix \( \eta_m(t, \varepsilon_m) \), \( \left[ L_m^{(1)}(t), \rho^{(0)}(t) \right] \), due to the operator of interest. We can already conclude that these contributions must be small due to the repeated use throughout this derivation of the assumption that the \( \varepsilon \) parameters will be numerically small (allowing us to terminate our many expansions at first order in \( \varepsilon \)). We’ve also shown that both small contributions, the small change in the state of the system in the case of linear response theory, or the small change in \( \rho_m(t, \varepsilon_m) \), obey the same equation of motion.

From (2.162) we write

\[
\left[ L_m^{(1)}(t), \rho^{(0)}(t) \right] = \frac{\eta_m(t, \varepsilon_m) - \rho^{(0)}(t)}{i\varepsilon_m}.
\]

(2.164)

Finally, taking a cue from linear response theory by taking the limit that the influence of the observable upon \( \eta_m(t, \varepsilon_m) \) is minimal (which enters the problem by taking the limit \( \varepsilon \to 0 \)) we obtain our main result

\[
C_{lm} = \lim_{\varepsilon_i, \varepsilon_m \to 0} \frac{1}{2\varepsilon_i \varepsilon_m} \text{Tr} \left[ \left( \rho^{(0)}(t_0) - \eta_m(t_0, \varepsilon_i) \right) \left( \rho^{(0)}(t_0) - \eta_m(t_0, \varepsilon_m) \right) \right].
\]

(2.165)

Whilst \( \rho^{(0)}(t_0) \) is provided by the boundary conditions on \( \rho \) the single-particle matrix \( \eta_m(t, \varepsilon_m) \) includes a dependence on the single-particle operator \( L_m \). However, \( \eta_m(t, \varepsilon_m) \) obeys the TDHF equation of motion so \( \eta_m(t_0, \varepsilon_m) \) can be obtained by running a TDHF calculation backwards using the boundary condition

\[
\eta_m(t_1, \varepsilon_m) = \exp(i\varepsilon_m Q_m) \rho^{(0)}(t_1) \exp(-i\varepsilon_m Q_m),
\]

(2.166)

as opposed to a normal TDHF calculation which is run forwards in time starting from a known initial density \( \rho^{(0)}(t_0) \) (although this forwards calculation is still required to solve (2.165) since it is needed to obtain \( \rho^{(0)}(t_1) \) and hence evaluate the boundary condition (2.166)).
If we are considering the fluctuation in a single operator then \( Q_t = Q_m \) and this reduces to

\[
(\Delta (Q_t)_{BV})^2 = \lim_{\epsilon, \gamma \to 0} \frac{1}{2\epsilon^2} \text{Tr} \left[ (\rho^{(0)}(t_0) - \eta_i(t_0, \epsilon))^{-2} \right].
\] (2.167)

### 2.3.11 Features of this Result

If we consider the single-particle operator, \( Q \), for a conserved quantity then the TDHF approach correctly predicts that the expectation value for that operator, \( \langle Q \rangle \), should remain constant however the fluctuations, \( \Delta Q \), given by (2.6) where the time dependence of \( \rho(t) \) is given by the TDHF equation, are not required to be constant in time. In the Balian-Vénéroni approach we have, for conserved observables \( [H, \hat{A}(t)] = 0 \) such that \( \hat{A}(t) = \exp(-\hat{L}(t)) = \exp(-\varepsilon \hat{Q}) \). The stationary value, \( \psi_{st} \), is then given by \( \psi_{st} = \ln(\exp(-\varepsilon \hat{Q}) \mathcal{D}(t)) \) which is independent of \( t_1 \) (which previously entered the problem through the boundary conditions on \( \hat{L}(t) \)) such that both \( \langle Q \rangle \) and \( \Delta Q \) are independent of the time when they are measured.

We can also consider whether we can use this result to make any observations about its predictions regarding the evolution of a nuclear system (and whether they differ to those of the usual TDHF approach).

A nucleus localised in space has associated with it both a momentum, \( p \), and a distribution of momentums, \( \Delta p \), but, in TDHF, in the absence of any external forces, it follows a fixed trajectory such that the distribution of possible positions of the nucleus, \( \Delta r \), remains constant in time. We say that, in TDHF, the nucleus is represented by a non-spreading wave packet [82]. However, in practice this distribution of possible momentums, \( \Delta p \) should lead to an widening distribution of possible positions, and hence an increasing \( \Delta r \), as a function of time according to [42]

\[
\Delta r^2(t) = \Delta r^2(t_0) + (t - t_0)\frac{\Delta p^2}{\hbar^2}.
\] (2.168)

This limitation of TDHF is not present in the Balian-Vénéroni approach where the state of the system evolves according to the more complicated equation of motion (2.97) which includes an inherent dependence on the operators of interest (through \( \rho' \) and \( \sigma' \)) such that, for observables which are not conserved (e.g. position), the fluctuation has a built-in dependence upon the time of measurement.

### 2.4 The Balian-Vénéroni Approach applied to Mass Fluctuations

To apply the Balian-Vénéroni approach to the calculation of mass fluctuations we consider the operator (from (2.2))

\[
\hat{N} = \theta (R_c - |\vec{r} - \vec{r}_{CM}|),
\] (2.169)
such that, from (2.167)

\[(\Delta N_{B(V)})^2 = \lim_{\varepsilon \to 0} \frac{1}{2\varepsilon^2} \text{Tr} \left[ \left( \rho^{(0)}(t_0) - \eta(t_0, \varepsilon) \right)^2 \right] , \quad (2.170)\]

with

\[\eta(t_1, \varepsilon) = \exp \left( i\varepsilon \theta (R_c - |\vec{r} - \vec{r}_{CM}|) \right) \rho^{(0)}(t_1) \exp \left( -i\varepsilon \theta (R_c - |\vec{r} - \vec{r}_{CM}|) \right) . \quad (2.171)\]

In the next section we describe our implementation of the Balian-Vénéroni approach using the OAK3D TDHF code.
Chapter 3

Codes and Procedures

3.1 The OAK3D HF/TDHF Code

The HF and TDHF calculations presented in this work were carried out using the OAK3D HF/TDHF code. The OAK3D code can be used to perform HF calculations of the ground state of any nucleus and has previously been used to carry out TDHF calculations for a single nucleus (for the simulation of resonances, e.g. [83,84]) or two nuclei (for simulating collisions, e.g. [38,85]). The OAK3D code is written in the Fortran 95 programming language and can (for dynamic calculations) be run parallelised to reduce the duration of the calculations. Calculations are carried out within a three-dimensional cartesian spatial box where the interactions between the nucleons are given by the full Skyrme interaction (including time-even and time-odd terms and the extended spin-orbit force) [6,15,16] and the Coulomb interaction with the exchange term in the Slater approximation. This approach is frequently referred, in the context of static calculations for determining nuclear ground state properties, to as the Skyrme-Hartree-Fock approach [86].

3.1.1 Implementation of the HF Approach within the OAK3D Code

The OAK3D code uses, as initial states, a set of three-dimensional harmonic oscillator wavefunctions where the size and shape of the simple harmonic oscillator potential is controlled by a set of user-defined parameters. This allows the user to use either a spherical starting point or an axially, or tri-axially, deformed one so as to maximize the chance of the code producing a deformed ground state, should one exist.

A HF calculation (for a nucleus with mass \( N \)) proceeds as outlined below [87].

1. An initial set of \( N \) harmonic oscillator states, \( \phi_i \), were generated by multiplying a gaussian by a set of Hermite polynomials [88, Complement Bv]. The width of the gaussian is controlled through the three size parameters, \( R_x \), \( R_y \) and \( R_z \) (all specified in fm).
2. The single-particle states, \( \phi_i \), are used to construct the one-body density matrix, \( \rho \), as well as all the other densities required by the Skyrme interaction (see section 3.1.3) and, as needed,
3.1.2 Implementation of the TDHF Approach within the OAK3D Code

In a TDHF calculation it is assumed that the initial states of all of the nuclei taking part in the calculation are known and were obtained from a previous HF calculation. To maintain consistency it is essential that the HF calculations for these single-particle wavefunctions use the same nucleon-nucleon interaction and spatial discretisation ($\Delta r$), but not the same sized spatial box, as are to be used in the dynamic calculation. This ensures that they describe a state which is a solution of the HF equations for both the static and dynamic problems and, in the absence of any external boost, remain stationary and stable during a time-dependent calculation. A resonance or collision calculation is performed by boosting the single-particle wavefunctions at the start of the dynamic calculation so as to excite the nucleus and/or set it in motion. The system is then allowed to evolve according to the TDHF equation of motion (B.11).

To perform a TDHF calculation the user must provide the set of single-particle wavefunctions describing the initial state of any nucleus which is to be included in the calculation and specify the desired initial position for each nucleus and the details of any boost which is to be applied to the system. The time-evolution of the $n^{th}$ single-particle state is then given by [91]

$$
\phi_n (\vec{r}, \sigma, t + \Delta t) = \exp \left( - \frac{i \Delta t \left( \hat{H}_{HF} \right)_n}{\hbar} \right) \phi_n (\vec{r}, \sigma, t),
$$

(3.2)

where $\left( \hat{H}_{HF} \right)_n$ is the single-particle Hamiltonian for the $n^{th}$ single-particle state (A.3) and $\Delta t$ is the timestep. The single-particle Hamiltonian is used in calculating the new single-particle state but itself depends on the state of the system and hence the single-particle wavefunctions. The action of the exponentiated operator on the single-particle wavefunction is obtained by a series expansion of the exponential (which is guaranteed to converge for small $\Delta t$) and the modified
Euler method is used to progress the single-particle wavefunctions from the initial time \( t_0 \) to the final time \( t_1 \) in steps \( \Delta t \). The essential procedure for each timestep is given below.

1. The single-particle Hamiltonian is evaluated using the current state (we denote the current time by \( t \)).
2. Hence estimate the state of the system at the time \( t + \Delta t/2 \) and the single-particle Hamiltonian at this later time.
3. Use this new single-particle Hamiltonian to determine the new state of the system at the time \( t + \Delta t \) using the original state of the system (known at the time \( t \)).

A resonance calculation is carried out using a single nucleus positioned at the origin of the spatial box and by applying a monopole or dipole boost (see section 3.1.4) after which the nucleus decays by particle emission. A collision is simulated by specifying the initial positions for the two nuclei (which must be in the \( x-z \) plane) as well as the centre-of-mass kinetic energy, \( E_{CM} \), (in MeV) and the impact parameter, \( b \), (in fm).

All of the TDHF calculations carried out with the OAK3D code used Dirichlet boundary conditions [89, pg. 496] in which any flux incident on the perimeter of the spatial box during the TDHF calculation is reflected back into the box. This flux may subsequently interact unphysically with any nuclei in the spatial box so this must be taken into account in the TDHF calculations [85]. This problem can be eliminated through the use of absorbing boundary conditions which remove any flux incident on the boundaries from the spatial box [33,92,93]. However, these can be computationally demanding and we also have an additional requirement in that the evaluation of (2.167) requires that the calculations be fully reversible. For these reasons we have not attempted to use absorbing boundary conditions in these calculations.

TDHF calculations are far more computationally demanding than HF calculations so the OAK3D code allows the length of the calculations to be reduced through parallelisation. This is most effective if the number of nucleons in the system, \( A \), is an integer number (e.g. \( n \)) times the number of processors used such that each processor calculates the time-evolution for \( N \) nucleons however in practice there is a law of diminishing returns such that (for a given availability of processors) there an advantage to be gained through running multiple independent calculations (using a smaller number of processors for each calculation) simultaneously rather than sequentially. This had to be taken into account in the current work where the evaluation of the limit in (2.167) means that large numbers of TDHF calculations must be carried out.

### 3.1.3 Interactions

The nucleon-nucleon interactions used in the OAK3D code are all based on the Skyrme interaction. Coulomb interactions are included but pairing interactions were not used in the current work. Pairing interactions are usually introduced into mean field calculations using the Barden-Cooper-
3.1 The OAK3D HF/TDHF Code

Schrieffer (BCS) or Hartree-Fock-Bogoliubov (HFB) approaches [78, Chap. 6 & 7] both of which change the fundamental equations which are solved and as such are not compatible with the Balian-Vénéroni approach in its current form. Balian et al. have published a more recent paper [94] in which they discuss variational methods for calculating expectation values, fluctuations and correlations including pairing, and make links between their equations and the BCS and HFB approaches, however this paper only considers static and not dynamic problems.

The Skyrme Interaction

In the OAK3D code the nucleon-nucleon interactions are given by the Skyrme interaction [15,16]. The Skyrme interaction is an effective interaction whose functional form was obtained by combining experimental observations with existing theories. Effective interactions include a number of parameters which are fitted to reproduce available experimental data (usually the bulk properties of the ground states of magic number nuclei such as their binding energies and charge radii). There are a number of parameterisations for the Skyrme interaction each fitted to reproduce slightly different sets of experimental observables.

Most Skyrme parameterisations are fitted assuming that centre-of-mass corrections will be used in the calculation [82,95]. Centre-of-mass corrections should be included in HF calculations since the nuclear mean-field is not translationally invariant, however they should not be included in TDHF calculations where the nuclei may be in motion. If we include centre-of-mass corrections in a HF calculation and use the resulting solution as the starting point for a dynamic calculation (without centre-of-mass corrections) then that state will not be a stable solution of the TDHF equations. To ensure consistency the centre-of-mass corrections must be neglected during the HF calculations. This will be a source of error when we compare the results of our HF calculations with experimental measurements. The SLy4d parameterisation is an example of a Skyrme force parameterisation fitted in the absence of centre-of-mass corrections [96].

The standard form of the Skyrme force is [7]

\[
V(\vec{r}_1, \vec{r}_2) = t_0 \left( 1 + x_1 \hat{P}_1 \right) \delta (\vec{r}_1 - \vec{r}_2) \\
+ \frac{1}{2} t_1 \left( 1 + x_1 \hat{P}_1 \right) (\vec{k}^2 \delta (\vec{r}_1 - \vec{r}_2) + \delta (\vec{r}_1 - \vec{r}_2) \vec{k}^2) \\
+ t_2 \left( 1 + x_2 \hat{P}_2 \right) \vec{k} \cdot \delta (\vec{r}_1 - \vec{r}_2) \vec{k} \\
+ \frac{1}{6} t_3 \left( 1 + x_3 \hat{P}_3 \right) \left( \rho \left( \frac{\vec{r}_1 + \vec{r}_2}{2} \right) \right)^{\alpha} \delta (\vec{r}_1 - \vec{r}_2) \\
+ ib_4 \left( \vec{\sigma}_1 \cdot \vec{\sigma}_2 \right) \cdot (\vec{k} \times \vec{k}), \tag{3.3}
\]

where \( t_1-t_4, x_1-x_4, \alpha \) and \( b_4 \) are parameters controlling the strengths of the interaction, fitted to
3.1 The OAK3D HF/TDHF Code

reproduce experimental data. \( P_\sigma \) is the spin exchange operator

\[
P_\sigma = \frac{1}{2} (1 + \vec{\sigma}_1 \cdot \vec{\sigma}_2),
\]  

(3.4)

where

\[
\vec{\sigma} = i\vec{\sigma}_x + j\vec{\sigma}_y + k\vec{\sigma}_z,
\]  

(3.5)

and the operator \( k \) is given by

\[
k = \frac{1}{2i} (\nabla_1 - \nabla_2),
\]  

(3.6)

and acts to the right whilst \( k' \) is the complex conjugate of \( k \) and acts to the left. The \( t_0 \) term is a central potential term, the \( t_1 \) and \( t_2 \) terms are the non-local terms and the \( t_3 \) term is a density dependent two-body interaction (or, for the special case of \( \alpha = 1 \), a three-body interaction). The \( b_4 \) term is the spin-orbit force. The Skyrme force is a zero-range (or local) force which makes it extremely well suited to use in computationally complex and demanding simulations. The development of the Skyrme force is discussed at some length in [6,77].

We write the Skyrme energy functional [97–99]

\[
E = \int d^3r \mathcal{H} (\vec{r}),
\]  

(3.7)

where \( \mathcal{H} (\vec{r}) \) is the energy density and is written in terms of a set of six densities [6]; the particle density, \( \rho (\vec{r}) \), and the spin density, \( \sigma (\vec{r}) \)

\[
\rho_q (\vec{r}) = \sum_m \sum_\sigma |\phi^q_m (\vec{r}, \sigma)|^2,
\]  

(3.8)

\[
\sigma_q (\vec{r}) = \sum_m \sum_\sigma \phi^q_m (\vec{r}, \sigma) \delta \phi^q_m (\vec{r}, \sigma),
\]  

(3.9)

where \( q \) denotes the isospin, \( \sigma \) the spin co-ordinate, and the summation over \( \text{"m"} \) runs over all occupied single-particle states with isospin \( q \). The kinetic energy density, \( \tau_q (\vec{r}) \), and the vector kinetic energy density, \( \mathcal{T}_q (\vec{r}) \) are given by

\[
\tau_q (\vec{r}) = \sum_{m \sigma} \sum \left| \nabla \phi^q_m (\vec{r}, \sigma) \right|^2,
\]  

(3.10)

\[
\mathcal{T}_q (\vec{r}) = \sum_m \sum_\sigma \sum_{i=x,y,z} \left( \nabla_i \phi^q_m (\vec{r}, \sigma) \right) \delta \left( \nabla_i \phi^q_m (\vec{r}, \sigma) \right).
\]  

(3.11)

The momentum density, \( \mathcal{J}_q (\vec{r}) \), and the spin-orbit density \( \mathcal{J}_q (\vec{r}) \) are

\[
\mathcal{J}_q (\vec{r}) = \frac{1}{2i} \sum_{m \sigma} \left( \phi^q_m (\vec{r}, \sigma) \left( \nabla \phi^q_m (\vec{r}, \sigma) \right) \right) \left( \nabla \phi^q_m (\vec{r}, \sigma) \right) - \left( \nabla \phi^q_m (\vec{r}, \sigma) \right) \phi^q_m (\vec{r}, \sigma),
\]  

(3.12)

\[
\mathcal{J}_q (\vec{r}) = -i \sum_{m \sigma} \phi^q_m (\vec{r}, \sigma) \left( \nabla \times \delta \right) \phi^q_m (\vec{r}, \sigma).
\]  

(3.13)
We write the Skyrme energy functional \([97-99]\) as the sum of a series of energy densities

\[
E = \int df(\mathcal{E}_{\text{kin}} + \mathcal{E}_{\text{Skyrme-even}} + \mathcal{E}_{\text{Skyrme-odd}} + \mathcal{E}_{\text{Coulomb}}).
\]  

(3.14)

\(\mathcal{E}_{\text{Skyrme-even}}\) and \(\mathcal{E}_{\text{Skyrme-odd}}\) are the time-even and time-odd contributions to the Skyrme energy functional. In general we should also include contributions due to pairing, \(\mathcal{E}_{\text{pair}}\), and, for static problems, centre-of-mass corrections, \(\mathcal{E}_{\text{CM}}\), however these have been omitted since they were not used in the current work. In dynamic problems all of these single-particle states and densities become time-dependent.

The proton and neutron densities are frequently combined to obtain the so-called isoscalar density, \(\rho = \rho_p + \rho_n\), and the isovector density, \(\bar{\rho} = \rho_p - \rho_n\), and similarly for the other densities \([6]\). The energy density can be written in terms of these six densities as

\[
\mathcal{E}_{\text{kin}} = \frac{\hbar^2}{2m}\tau,
\]  

(3.15)

\[
\mathcal{E}_{\text{Skyrme-even}} = \frac{B_0 + B_3 \rho^2}{2} \rho^2 - \frac{B_0' + B_3' \rho^2}{2} \bar{\rho}^2 + B_1 \rho T - B_1' \bar{\rho} T 
- \frac{B_2}{2} \rho (\nabla^2 \rho) + \frac{B_2}{2} \bar{\rho} (\nabla^2 \bar{\rho}) - B_4 \rho (\nabla \cdot J) - (B_4 + B_4') \bar{\rho} (\nabla \cdot J)
+ \frac{C_1}{2} \rho^2 - \frac{C_1}{2} \bar{\rho}^2,
\]  

(3.16)

\[
\mathcal{E}_{\text{Skyrme-odd}} = -B_1 \rho + B_1' \bar{\rho} - \frac{C_0 + C_3 \rho^2}{2} \rho^2 + \frac{C_0' + C_3' \rho^2}{2} \bar{\rho}^2 
+ \frac{C_2}{2} \cdot (\nabla^2 \rho) - \frac{C_2}{2} \cdot (\nabla^2 \bar{\rho}) - C_1 \rho \cdot T + C_1' \bar{\rho} \cdot T 
- B_4 \rho \cdot (\nabla \times j) - (B_4 + B_4') \bar{\rho} \cdot (\nabla \times j),
\]  

(3.17)

where the \(B, B', C\) and \(C'\) constants are related to the parameters of (3.3) according to \([6]\)

\[
B_0 = \frac{3}{2} t_0, \quad B_1 = \frac{1}{18} t_1 + \frac{8}{18} t_2 + \frac{1}{18} t_4 x_2, \quad B_2 = \frac{2}{9} t_2 - \frac{8}{9} t_3 + \frac{1}{9} t_4 x_2, \\
B_3 = \frac{4}{18} t_3, \quad B_4 = b_4 - \frac{1}{2} b_4, \quad B_0' = \frac{3}{2} t_0, \quad B_1' = \frac{1}{18} t_1 + \frac{8}{18} t_2 + \frac{1}{18} t_4 x_2, \\
B_2' = \frac{2}{9} t_2 - \frac{8}{9} t_3 + \frac{1}{9} t_4 x_2, \quad B_3' = \frac{4}{18} t_3, \quad B_4' = \frac{3}{2} b_4,
\]  

(3.18)

\[
C_0 = -\frac{1}{2} t_0 \left( \frac{1}{3} - x_0 \right), \quad C_0' = \frac{3}{2} t_0, \\
C_1 = \frac{1}{2} \left[ t_1 \left( \frac{1}{3} - x_1 \right) - t_2 \left( \frac{1}{3} + x_2 \right) \right], \quad C_1' = -\frac{1}{18} t_1 - t_2, \\
C_2 = -\frac{1}{18} \left[ 3 t_1 \left( \frac{1}{3} - x_1 \right) + t_2 \left( \frac{1}{3} + x_2 \right) \right], \quad C_2' = \frac{1}{18} \left( 3 t_1 + t_2 \right), \\
C_3 = -\frac{1}{6} t_3 \left( \frac{1}{3} - x_3 \right), \quad C_3' = \frac{1}{6} t_3,
\]  

(3.19)

and where, as an additional modification to the standard form of the Skyrme force, we have an
3.1 The OAK3D HF/TDHF Code

additional parameter, $b_4$, in the spin-orbit term which allows the strengths of the isoscalar and isovector components of the spin-orbit force to be tuned independently (this is the extended spin-orbit force [38,86]).

The Coulomb Interaction

The Coulomb interaction is a long-range interaction and as such is very different from the short-range nuclear forces taken into account by the zero-range Skyrme interaction. The Coulomb force must be included as an additional term in the interaction where, in the OAK3D code, this is accomplished using the Fourier Analysis with Long Range Forces approach described in [100] in which the short-range and the long-range components of the Coulomb force are separated out and treated separately.

3.1.4 Excitations of the System for Dynamic Calculations

Resonance Boost

To model a giant monopole resonance (GMR) in the OAK3D code the single-particle wavefunctions obtained from the HF calculation were boosted at the start of the TDHF calculation by multiplying them by the phase factor [84]

$$B_M(x, y, z) = \exp \left(-iF \frac{1}{4\pi} \frac{A_m \times (x^2 + y^2 + z^2)}{1 + \exp (x^2 + y^2 + z^2)} \right),$$  \hspace{1cm} (3.20)

where "$A_m$" is a parameter which fixes the strength of the boost (and determines the amount of energy given to the system and the amount of mass emitted as the nucleus decays). A giant dipole resonance (GDR) is obtained using the phase factor

$$B_D(x, y, z) = \exp \left( iF \frac{5}{4\pi} \frac{1}{1 + \exp \left( \frac{1}{\sqrt{x^2 + y^2 + z^2}} \right)} (A_x x + A_y y + A_z z) \right),$$  \hspace{1cm} (3.21)

where "$A_x$", "$A_y$" and "$A_z$" determine the strength of the boost.

The $F$ factor takes different values depending on whether an isoscalar or an isovector boost is being applied. For an isovector resonance $F = -1/(A - Z)$ for neutron states and $1/Z$ for proton states whilst for an isoscalar boost it is always 1.0 (where $A$ is the number of nucleons in the nucleus and $Z$ is the number of protons). This difference is required to ensure that the boost is applied equally to the protons and neutrons when an isovector boost is applied to generate a giant dipole resonance in nuclei with differing numbers of protons and neutrons. This ensures that the centre-of-mass of the nucleus is not shifted when applying an isovector giant dipole boost. The boost is position dependent and is focused upon the nucleus (assumed in the above formulae to be at (0,0,0,0,0)) with a reduced effect on the extended tails of the single-particle wavefunctions.
Collisions Boost

A collision calculation is performed by assigning initial positions to the nuclei and specifying the centre-of-mass kinetic energy (in MeV), and impact parameter (in fm) for the collision. The OAK3D code assumes that the reaction takes place in the $x$-$z$ plane and computes the kinetic energy boost required in the $x$- and $z$-directions based on the centre-of-mass kinetic energy and impact parameter. The centre-of-mass kinetic energy and impact factor are specified assuming that the nuclei start an infinite distance apart where they will not interact and are adjusted to take into account any initial Coulomb repulsion between the nuclei such that they approach each other along trajectories dictated by the Coulomb interactions between them. The boost is applied by multiplying the single-particle wavefunctions in each nucleus by the boost factor

$$ B_C = \exp \left( i \mathbf{k} \cdot \mathbf{R} \right), $$

where $\mathbf{k}$ is the wavevector specifying the strength of the boost to be applied in each direction and $\mathbf{R}$ is the position of the centre of mass of the nucleus being boosted.

$$ R_j = \frac{1}{A_j} \sum_{i=1}^{A_j} \mathbf{r}_i, \quad \text{where} \quad j = T, P $$

denotes the target and projectile nuclei so that the summation runs over all the single-particle wavefunctions associated with one of these nuclei. The $\mathbf{r}_i$ are the positions of the centre's of mass of the individual nucleons. $\mathbf{R}_j$ is defined such that

$$ A_T \mathbf{R}_T + A_P \mathbf{R}_P = 0, $$

which ensures that the total momentum in the centre-of-mass frame is zero.

3.2 Implementation of the Balian-Vénérioni Formula

To determine the fluctuation in a single-particle observable, $\hat{Q}$, using the Balian-Vénérioni Method we must evaluate

$$ (\Delta \hat{Q})^2 = \lim_{\varepsilon \to 0} \frac{1}{2 \varepsilon^2} \text{Tr} \left[ (\rho(t_0) - \eta(t_0, \varepsilon))^2 \right], $$

where $\rho(t)$ and $\eta(t, \varepsilon)$ are both one-body density matrices and $\eta(t, \varepsilon)$ is defined such that

$$ \eta(t_1, \varepsilon) = e^{i \varepsilon \hat{Q}} \rho(t_1) e^{-i \varepsilon \hat{Q}}. $$

The OAK3D code works in terms of the single-particle wavefunctions and not the single-particle density matrix. Written in terms of the single-particle wavefunctions, and substituting in $\hat{Q}$ =
3.2 Implementation of the Balian-Vénéroni Formula

\( \theta (R_c - |\vec{r} - \vec{r}_{CM}|) \) from (2.2), this becomes

\[
\psi_n (\vec{r}, \sigma, t_1, \epsilon) = \exp (i\epsilon \theta (R_c - |\vec{r} - \vec{r}_{CM}|)) \phi_n (\vec{r}, \sigma, t_1), \tag{3.27}
\]

where we use \( \phi_n (\vec{r}, \sigma, t) \) to represent the single-particle wavefunction associated with the \( n^{th} \) nucleon in the standard (time running forwards) calculations and \( \psi_n (\vec{r}, \sigma, t, \epsilon) \) to represent this single-particle wavefunction in the time-reversed calculations.

Evaluating (3.25) requires that the TDHF code be run forwards, \( t_0 \rightarrow t_1 \), and then backwards, \( t_1 \rightarrow t_0 \), many times for differing values of \( \epsilon \). The time-reversed calculations are carried out by changing the timestep, \( \Delta t \), to be negative. The OAK3D code includes the ability to generate a set of restart files which allows the calculation to be resumed at a later date. To minimise the modifications to the existing OAK3D code a modular approach was used in which each TDHF calculation (the starting forward calculations and the backwards calculations for varying values of \( \epsilon \)) were performed as separate calculations whilst additional independent programs and scripts were used to modify the wavefunctions in the restart files according to the transformation (3.27) as needed and to compute the fluctuations, \( \Delta N_{BV} (\epsilon) \), using the wavefunctions from two sets of restart files.

The density matrix \( \rho (t) \) is evaluated using the single-particle wavefunctions which describe the initial state of the system whilst the density matrix \( \eta (t, \epsilon) \) is evaluated using the final state wavefunctions obtained at the end of the time-reversed TDHF calculation. To evaluate (3.25) both \( \rho (t) \) and \( \eta (t, \epsilon) \) must be calculated in the same basis. If we choose to work in a basis of single-particle states then it is natural to write \( \rho (t) \) in the basis of the states \( \psi_n \) and \( \eta (t, \epsilon) \) in the basis of the states \( \phi_n \). A transformation matrix, \( D \), must then be used to convert one of the density matrices into the preferred basis of the other density matrix. In this case equation (3.25) would be written as,

\[
\left( \Delta \hat{Q} \right)^2 = \lim_{\epsilon \rightarrow 0} \frac{1}{2\epsilon^2} \text{Tr} \left[ (\rho (t_0) - D^\dagger \eta (t_0, \epsilon) D)^2 \right]. \tag{3.28}
\]

In practice we only compute the occupied single-particle states so the sets of single-particle states are not complete sets and we cannot calculate \( D \) with sufficient accuracy. The solution is to work in the basis of position since then we need no transformation since both sets of single-particle states can be simply defined in that same model space. The problem with working in the basis of position is that density matrices become unmanageably large. In the basis of occupied single-particle states the density matrices are \( N \times N \) (for an \( N \)-particle system) and can be calculated and used in (3.25) but in the basis of position these matrices (particularly for our 3D calculations) become huge. This means that (3.25) must be written and evaluated in terms of the single-particle wavefunctions.

In the OAK3D code the wavefunctions are calculated in terms of position, \( \vec{r} \), and spin, \( \sigma \), such
that, in the basis of position and spin, the elements of $\rho$ and $\eta$ are given by

$$\rho(\vec{r}\sigma, \vec{r}'\sigma') = \sum_n \phi^*_n(\vec{r}', \sigma') \phi_n(\vec{r}, \sigma),$$  \hspace{1cm} (3.29)

$$\eta(\vec{r}\sigma, \vec{r}'\sigma') = \sum_n \psi^*_n(\vec{r}', \sigma') \psi_n(\vec{r}, \sigma),$$  \hspace{1cm} (3.30)

where the summation is over all occupied single-particle states and we omit all $t$ and $\varepsilon$ labels in the interests of compactness. To write (3.25) in terms of the single-particle wavefunctions, $\phi_n$ and $\psi_n$ we first write

$$\text{Tr}[(\rho - \eta)^2] = \sum_{\sigma\sigma'} \int d\vec{r} \int d\vec{r}' \rho(\vec{r}\sigma, \vec{r}'\sigma') - \eta(\vec{r}\sigma, \vec{r}'\sigma') \rho(\vec{r}'\sigma', \vec{r}\sigma) - \eta(\vec{r}'\sigma', \vec{r}\sigma) \rho(\vec{r}\sigma, \vec{r}'\sigma') + \eta(\vec{r}\sigma, \vec{r}'\sigma') \eta(\vec{r}'\sigma', \vec{r}\sigma),$$  \hspace{1cm} (3.31)

Substituting in for $\rho$ and $\eta$ we obtain

$$\text{Tr}[(\rho - \eta)^2] = \sum_{\sigma\sigma'} \int d\vec{r} \int d\vec{r}' \sum_{nm} \phi^*_n(\vec{r}', \sigma') \phi_n(\vec{r}, \sigma) \phi^*_m(\vec{r}', \sigma') \phi_m(\vec{r}', \sigma')$$

$$- \phi^*_n(\vec{r}', \sigma') \phi_n(\vec{r}, \sigma) \psi^*_m(\vec{r}, \sigma) \psi_m(\vec{r}', \sigma')$$

$$- \psi^*_n(\vec{r}', \sigma') \psi_n(\vec{r}, \sigma) \phi^*_m(\vec{r}, \sigma) \phi_m(\vec{r}', \sigma')$$

$$+ \psi^*_n(\vec{r}', \sigma') \psi_n(\vec{r}, \sigma) \psi^*_m(\vec{r}, \sigma) \psi_m(\vec{r}', \sigma'),$$  \hspace{1cm} (3.32)

which we now re-write as

$$\text{Tr}[(\rho - \eta)^2] = \sum_{nm} \left[ \left( \sum_\sigma \int d\vec{r} \phi^*_m(\vec{r}, \sigma) \phi_n(\vec{r}, \sigma) \right) \left( \sum_\sigma \int d\vec{r}' \phi^*_n(\vec{r}', \sigma') \phi_m(\vec{r}', \sigma') \right) \right.$$

$$- \left( \sum_\sigma \int d\vec{r} \phi^*_m(\vec{r}, \sigma) \phi_n(\vec{r}, \sigma) \right) \left( \sum_\sigma \int d\vec{r}' \phi^*_n(\vec{r}', \sigma') \psi_m(\vec{r}', \sigma') \right)$$

$$- \left( \sum_\sigma \int d\vec{r} \psi^*_m(\vec{r}, \sigma) \psi_n(\vec{r}, \sigma) \right) \left( \sum_\sigma \int d\vec{r}' \phi^*_n(\vec{r}', \sigma') \phi_m(\vec{r}', \sigma') \right)$$

$$+ \left( \sum_\sigma \int d\vec{r} \psi^*_m(\vec{r}, \sigma) \psi_n(\vec{r}, \sigma) \right) \left( \sum_\sigma \int d\vec{r}' \psi^*_n(\vec{r}', \sigma') \psi_m(\vec{r}', \sigma') \right) \left. \right] \hspace{1cm} (3.33)$$

We now recognise that some of the terms in the brackets are just the elements of the one-body density matrices $\rho$ and $\eta$ written in the basis of single-particle states

$$\rho_{mn} = \sum_\sigma \int d\vec{r} \phi^*_m(\vec{r}, \sigma) \phi_n(\vec{r}, \sigma),$$  \hspace{1cm} (3.34)

$$\eta_{mn} = \sum_\sigma \int d\vec{r} \psi^*_m(\vec{r}, \sigma) \psi_n(\vec{r}, \sigma).$$  \hspace{1cm} (3.35)
We also introduce a new matrix, $\beta$ defined as

$$\beta_{mn} = \sum_{\sigma} \int d\vec{r} \psi_n^* (\vec{r}, \sigma) \phi_m (\vec{r}, \sigma),$$  \hspace{1cm} (3.36)$$

where this matrix is similar to the transformation matrix $D$ which transforms between the initial and final sets of single-particle states except that it does not require that the sets of single-particle wavefunctions be complete sets (it is the transformation matrix but only calculated using occupied states). Written in terms of these matrices (3.33) becomes

$$\text{Tr} \left[ (\rho - \eta)^2 \right] = \sum_{nm} \rho_{nm} \rho_{mn} - \sum_{nm} \beta_{nm} \beta_{nm}^* - \sum_{nm} \beta_{mn} \beta_{mn} - \sum_{nm} \eta_{nm} \eta_{mn},$$

which gives us our final result

$$\Delta N^2 = \lim_{\varepsilon \to 0} \frac{1}{2\varepsilon^2} \left( \text{Tr} \left[ \rho^2 \right] + \text{Tr} \left[ \eta^2 \right] - 2 \sum_{nm} \beta_{nm} \beta_{nm}^* \right).$$  \hspace{1cm} (3.38)$$

The three matrices, $\rho$, $\eta$ and $\beta$ are all defined in the basis of single-particle states which means that they are small and can be easily computed for use in this formula. Using this formula avoids the need to calculate the large density matrices or the need to perform double integrations over position and spin which would otherwise make these calculations extremely lengthy.
Chapter 4

Code Validation: GDR in $^{32}\text{S}$

The Balian-Vénéroni variational approach, implemented as discussed in the previous chapter, has been applied, as a simple test case, to a giant dipole resonance in $^{32}\text{S}$ decaying by particle emission. The real testing ground for this method will be heavy-ion collisions (where there is experimental data available for comparisons). However, a simpler and lighter system is preferable as a test case for reasons of computational speed.

4.1 Hartree-Fock Calculation for $^{32}\text{S}$

The OAK3D HF code was used to perform a set of HF calculations to determine the ground state wavefunctions for a $^{32}\text{S}$ nucleus using a selection of common Skyrme parameterisations. The initial simple harmonic oscillator potential was chosen to be triaxially deformed so as to maximise the chance of the code finding a deformed solution should one exist. The size of the simple harmonic oscillator potential was set to 2.9 fm in the $x$-direction, 3.0 fm in the $y$-direction and 3.1 fm in the $z$-direction. This is equivalent to a starting tri-axial deformation of $\beta_2 = 0.13, \gamma = 49^\circ$. The calculation was carried out in a cubic spatial box with the dimensions $-9.5 \rightarrow 9.5$ fm and points every 1.0 fm for a total of $20 \times 20 \times 20 = 8,000$ points. Calculations were carried out using the SkM$^*$ [101,102], SLy4 [7,103], SLy4d [96] and SLy6 [7,103] Skyrme parameterisations. The parameters for these different Skyrme parameterisations are shown in table 4.2. Some of the bulk properties of the calculated $^{32}\text{S}$ ground states are also shown in table 4.1. Skyrme parameterisations are typically fitted to reproduce the ground state bulk properties of magic number nuclei which means that these calculations for a mid-shell nucleus may be regarded as "parameter free". The differences between the calculated and experimental values are because we do not include centre-of-mass corrections since we will be conducting time-dependent calculations. All of the Skyrme force parameterisations used (except SLy4d which was specifically designed for use in dynamic calculations) were fitted using the assumption that they would be used in combination with centre-of-mass corrections. Using these forces without centre-of-mass corrections and without refitting the Skyrme parameters to compensate for this introduces errors which are reflected here in the
4.2 TDHF Calculation for a GDR in $^{32}$S

<table>
<thead>
<tr>
<th>Parameter Set</th>
<th>BE</th>
<th>RMS Radius (fm)</th>
<th>$\beta_n$</th>
<th>$\gamma$ (°)</th>
<th>S (P)</th>
<th>S (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SkM$^*$</td>
<td>275.97</td>
<td>3.137</td>
<td>0.005</td>
<td>3.1</td>
<td>7.465</td>
<td>13.202</td>
</tr>
<tr>
<td>SLy4</td>
<td>272.22</td>
<td>3.159</td>
<td>0.051</td>
<td>1.7</td>
<td>7.643</td>
<td>13.434</td>
</tr>
<tr>
<td>SLy4d</td>
<td>302.51</td>
<td>3.083</td>
<td>0.001</td>
<td>3.3</td>
<td>7.703</td>
<td>13.557</td>
</tr>
<tr>
<td>SLy6</td>
<td>260.36</td>
<td>3.175</td>
<td>0.111</td>
<td>2.3</td>
<td>7.354</td>
<td>13.086</td>
</tr>
<tr>
<td>Expt. [104–106]</td>
<td>271.78</td>
<td>3.248(11)</td>
<td>0.249(8)</td>
<td>-</td>
<td>8.864</td>
<td>15.042(8)</td>
</tr>
</tbody>
</table>

Table 4.1: Some bulk properties of a $^{32}$S nucleus calculated using the OAK3D HF code with different parameterisations of the Skyrme interaction and compared to the experimental values. $S (P)$ and $S (N)$ are the proton and neutron separation energies. All energies are in MeV.

differences between the calculated and experimental ground state energies [95]. This is the main source of the large differences between our calculated binding energies and the experimental values.

It might by a little surprising that, of all the Skyrme forces used, the SLy4d produced the least accurate ground state energy despite the expectation that this force would perform better due to it being fitted in the absence of centre-of-mass corrections. However, it should be remembered that the SLy4d parameterisation was designed for use in dynamic calculations and as such might not be expected to perform as well in a static calculation as the other Skyrme forces which are fitted more for use in static calculations.

From these results we chose (somewhat arbitrarily but also due to it having the largest deformation) to use the SLy6 Skyrme parameterisation in our time-dependent calculations. Table 4.3 shows some additional bulk properties for the calculated $^{32}$S ground state. The prolate shape of the nucleus (with $x$ the long-axis) can be seen in the values of $\langle x^2 \rangle$, $\langle y^2 \rangle$ and $\langle z^2 \rangle$.

The convergence of the HF code is measured by looking at the average fluctuation in the single-particle energies, $\Delta H_{HF}$. Figure 4.1 shows the total energy in the system, $E$, the change in the energy (as a fraction of the energy in the system), $\Delta E/E$, and $\Delta H_{HF}$ plotted as a function of iteration number to demonstrate the convergence of these quantities.

4.2 TDHF Calculation for a GDR in $^{32}$S

To validate the single-particle wavefunctions from the static calculation for use in the time-dependent calculations we calculated $\langle N \rangle$ and $\Delta N_{TDHF}$ using the single-particle wavefunctions from the HF solution and integrating over the entire spatial box. We obtained $\langle N \rangle = 32.0000$ and $\Delta N_{TDHF} = 0.0000$ as we would expect. To determine how well localised the nucleus is these observables were evaluated but integrating only over those points within 8.0 fm of the centre-of-mass of the nucleus, assumed to be at (0.0,0.0,0.0). In this case we obtained $\langle N \rangle = 31.9888$ and $\Delta N_{TDHF} = 0.1058$ which indicated that the nucleus was well localised. This value for the
4.2 TDHF Calculation for a GDR in $^{32}\text{S}$

<table>
<thead>
<tr>
<th>Parameters</th>
<th>$t_0$ (MeV fm$^3$)</th>
<th>$t_1$ (MeV fm$^5$)</th>
<th>$t_2$ (MeV fm$^5$)</th>
<th>$t_3$ (MeV fm$^{3+3\alpha}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{SkM}^*$</td>
<td>-2645.000</td>
<td>410.000</td>
<td>-135.000</td>
<td>15595.000</td>
</tr>
<tr>
<td>SLy4</td>
<td>-2488.913</td>
<td>486.818</td>
<td>-546.395</td>
<td>13777.000</td>
</tr>
<tr>
<td>SLy4d</td>
<td>-2497.662</td>
<td>473.216</td>
<td>-333.654</td>
<td>13487.000</td>
</tr>
<tr>
<td>SLy6</td>
<td>-2479.500</td>
<td>462.180</td>
<td>-448.610</td>
<td>13673.000</td>
</tr>
<tr>
<td>$x_0$</td>
<td>0.090</td>
<td>0.834</td>
<td>0.812</td>
<td>0.825</td>
</tr>
<tr>
<td>$x_1$</td>
<td>0.000</td>
<td>-0.344</td>
<td>-0.723</td>
<td>-0.465</td>
</tr>
<tr>
<td>$x_2$</td>
<td>0.000</td>
<td>-1.000</td>
<td>-1.000</td>
<td>-1.000</td>
</tr>
<tr>
<td>$x_3$</td>
<td>0.000</td>
<td>1.354</td>
<td>1.398</td>
<td>1.355</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>0.16667</td>
<td>0.16667</td>
<td>0.16667</td>
<td>0.16667</td>
</tr>
<tr>
<td>$b_4$ (MeV fm$^5$)</td>
<td>130.000</td>
<td>123.000</td>
<td>128.000</td>
<td>122.000</td>
</tr>
<tr>
<td>$b_4'$ (MeV fm$^5$)</td>
<td>65.000</td>
<td>61.500</td>
<td>128.000</td>
<td>61.000</td>
</tr>
</tbody>
</table>

Table 4.2: The Skyrme force parameters for the SkM* [101,102], SLy4 and SLy6 [7,103] and the SLy4d [96] parameterisations.

Figure 4.1: The binding energy, $E$, $\Delta E/E$ and the average fluctuation in the single-particle energies, $\Delta H_{HF}$, plotted as a function of iteration number to demonstrate the convergence of the HF calculation for $^{32}\text{S}$ (using the SLy6 parameterisation of the Skyrme interaction).
4.2 TDHF Calculation for a GDR in $^{32}$S

<table>
<thead>
<tr>
<th>No. Nucleons</th>
<th>Protons</th>
<th>Neutrons</th>
<th>All Nucleons</th>
</tr>
</thead>
<tbody>
<tr>
<td>RMS Radius (fm)</td>
<td>3.1957</td>
<td>3.1538</td>
<td>3.1748</td>
</tr>
<tr>
<td>$g_{20}$</td>
<td>7.2510</td>
<td>6.9586</td>
<td>14.210</td>
</tr>
<tr>
<td>$\langle x^2 \rangle$ (fm$^2$)</td>
<td>3.8831</td>
<td>3.7751</td>
<td>3.8291</td>
</tr>
<tr>
<td>$\langle y^2 \rangle$ (fm$^2$)</td>
<td>3.1813</td>
<td>3.1016</td>
<td>3.1414</td>
</tr>
<tr>
<td>$\langle z^2 \rangle$ (fm$^2$)</td>
<td>3.1479</td>
<td>3.0696</td>
<td>3.1088</td>
</tr>
</tbody>
</table>

Table 4.3: Some additional bulk properties of a $^{32}$S nucleus calculated using the OAK3D HF code with the SLy6 parameterisation of the Skyrme interaction.

cutoff radius is the same as that used in the earlier calculations of Troudet and Vautherin [48] and defines a volume large enough to completely envelope the nucleus whilst excluding the majority of the spatial box. The nuclear matter density at the boundary of the region of interest was 5 orders of magnitude lower than it was at the centre of the nucleus.

At the start of the dynamic calculation the single-particle wavefunctions from the HF solution were given an isovector boost in accordance with (3.21) and with $A_x = A_y = A_z = 112.5$ fm$^{-1}$. The dynamic calculation was carried out in an enlarged cubic spatial box with the dimensions $-15.5 \rightarrow 15.5$ fm and points every 1.0 fm.

The code was run from $t = 0$ fm/c to $t_1 = 250$ fm/c in steps of 0.2 fm/c (a total of 1250 timesteps). The emitted nucleons are reflected back from the boundary of the box and, if the simulation is allowed to run for long enough, re-enter the region occupied by the de-exciting nucleus causing additional unphysical interactions (one symptom of which would be an unphysical increase in the mass of the nucleus, $(N)$. An analysis of the density, and of $(N)$ as a function of time, was used to verify that the number of nucleons in the nucleus had stabilised well in advance of the time $t_1$ and to check for evidence of reflected nucleons re-interacting with the nucleus. The number of nucleons in the nucleus and the associated fluctuation, $\Delta N_{TDHF}$, plotted as a function of time, are shown in figure 4.2.

The dipole moments, $Q_x$, $Q_y$ and $Q_z$, are given by [34]

$$Q_i = \frac{(A - Z) Z}{A} \left( \langle x_i^p \rangle - \langle x_i^n \rangle \right), \quad (4.1)$$

where $i = 1, 2, 3$ denotes $x$, $y$ and $z$ and $\langle x_i^p \rangle$ and $\langle x_i^n \rangle$ are the expectation values for position calculated using the proton and neutron single particle states respectively. These are shown in figure 4.3. Due to the prolate deformation of the $^{32}$S nucleus (with $x$ the long axis), the $Q_y$ and $Q_z$ values are identical and differ from the $Q_x$ values. If it is assumed that the oscillations of the protons and neutrons are harmonic then the periodicity, $T$, of $Q_x$, $Q_y$ and $Q_z$ allow estimates for
the excitation energies of the oscillations along each of these three primary axes to be estimated according to

\[ E = \hbar \omega = \frac{2\pi \hbar}{T}. \]  

(4.2)

The assumption of harmonic motion means that we are assuming that the nucleus oscillates at a single fixed frequency (such that the strength function would be a delta function) and that the frequency of the oscillations is independent of their amplitude. The validity of these assumptions was investigated by Reinhard et al. [84] who showed that this is a better assumption in heavier nuclei, however the exercise still provides a useful estimate of the energy of the dominant frequency component. In this instance we obtain, for \( Q_x \), a period of \( \approx 71 \text{ fm/c} \) giving an excitation energy \( E_x \approx 17.5 \text{ MeV} \) and, for \( Q_y \) and \( Q_z \), a period of \( \approx 68 \text{ fm/c} \) giving an excitation energy \( E_y \approx E_z \approx 18.3 \text{ MeV} \). The energy of a giant dipole resonance is approximately given by [107]

\[ E_{\text{GDR}} = 31.2A^{-\frac{1}{3}} + 20.6A^{-\frac{1}{3}}, \]

(4.3)

which, for \(^{32}\text{S}\) gives \( E_{\text{GDR}} = 21.4 \text{ MeV} \) which confirms that our rough estimates are reasonable.

At the end of the calculation the mass of the nucleus was \( \langle N \rangle = 26.6366 \) (13.2884 protons and 13.3482 neutrons) which represents the emission of \( \approx 5 \) nucleons. The fluctuation, calculated according to the TDHF result, was \( \Delta N_{\text{TDHF}} = 2.0218 \). The large decrease in the mass of the nucleus in this calculation is a consequence of the large boost. These expectation values and

Figure 4.2: The number of nucleons in the \(^{32}\text{S}\) nucleus, \( \langle N \rangle \), and the mass fluctuation, \( \Delta N_{\text{TDHF}} \), calculated by integrating over all points within \( R_c = 8.0 \text{ fm} \) of the centre-of-mass of the nucleus and plotted against time during the decay of the giant dipole resonance.
4.3 Application of the BV approach to a GDR in $^{32}$S

4.3.1 Time Reversal in the OAK3D TDHF Code

Time-reversed calculations were carried out in the OAK3D code by starting from the set of single-particle wavefunctions extracted at the end of a previous TDHF calculation (as opposed to a set of wavefunctions obtained from a HF calculation) and replacing the existing timestep, $\Delta t$, by $-\Delta t$. Evaluating fluctuations using the Balian-Vénérion approach requires two sets of single-particle wavefunctions calculated at the initial time $t_0$. In practice the nuclei were boosted at the start...
4.3 Application of the BV approach to a GDR in $^{32}$S

$R_c = \infty \text{ fm}$

|        | $\langle N \rangle_{t_0}$ | $\Delta N_{TDHF}|_{t_0}$ | $\langle N \rangle_{t_0}$ | $\Delta N_{TDHF}|_{t_0}$ |
|--------|---------------------------|---------------------------|---------------------------|---------------------------|
| HF     | 32.0000                   | 0.0000                    | 31.9888                   | 0.1058                    |
| TDHF $t_0$ | 32.0015                   | 0.0394                    | 31.9902                   | 0.0987                    |
| TDHF $t_0 \rightarrow t_1 \rightarrow t_0$ | 32.0003                   | 0.0163                    | 31.9887                   | 0.1061                    |

$R_c = 8.0 \text{ fm}$

Table 4.4: Comparison of $\langle N \rangle$ and $\Delta N_{TDHF}$ to test the time reversal and the reversibility of the OAK3D code. The results labelled “HF” were obtained using the single-particle wavefunctions from the HF calculation for the $^{32}$S nucleus. The “TDHF $t_0$” results were obtained from a TDHF calculation after the nucleus had been boosted and a single iteration performed. The final set of results were obtained after a time-reversed TDHF calculation to assess the reversibility of the code.

of the dynamic calculation so the “starting” wavefunctions were the single-particle wavefunctions after the completion of the first dynamic timestep (such that $t_0 = \Delta t \text{ fm/c}$).

We have already tested the wavefunctions from the HF calculation to confirm that they describe a well localised system with exactly 32.0 nucleons and zero fluctuation in the particle number. Ideally $\langle N \rangle$ and $\Delta N$ should still have these values at the time $t_0$ however the mapping of the single-particle wavefunctions into the larger spatial box (including the re-orthogonalisation of the single-particle wavefunctions), the application of the dipole boost and the first iteration mean that we actually obtain $\langle N \rangle = 32.0015$ and $\Delta N_{TDHF} = 0.0394$.

To test the reversibility of the TDHF calculations the single-particle wavefunctions, taken at the time $t_1 = 250 \text{ fm/c}$ (i.e. after 1250 iterations), were used (unaltered) as the starting point for a second calculation running backwards to the time $t_0$. As in section 3.2 we will consistently use $\phi_i(t)$ to denote a single-particle state associated with a standard TDHF calculation ($\Delta t$ positive) and $\psi_i(t, \epsilon)$ to denote a single-particle wavefunction associated with a time-reversed time-dependent HF calculation ($\Delta t$ negative), omitting now any explicit position or spin ($\tau$ or $\sigma$) dependence. The $\epsilon$ label indicates the strength of the transformation applied to the single-particle wavefunctions $\psi_i(t, \epsilon)$ before the start of the time-reversed calculation (such that $\epsilon = 0.0$ means that the wavefunctions were left unaltered).

After performing the time-reversed calculations we find, using the single particle states $\psi_i(t_0, \epsilon = 0.0)$, $\langle N \rangle = 32.0003$ and $\Delta N = 0.0163$. These checks were all repeated limiting the integrations to those points within $R_c = 8.0 \text{ fm}$ of the centre of mass of the nucleus (assumed to be at the origin) and these results are summarised in table 4.3.1. These values are useful since they provide an estimate of the numerical errors inherent in our TDHF calculations. Looking more closely we plot in figure 4.4 the normalisation constants and overlaps for the two sets of single-particle states, $\phi_i(t_0)$ and $\psi_i(t_0, \epsilon = 0.0)$, at the time $t_0$. We can clearly see that the deviations from unity are much larger for the single-particle wavefunctions $\psi_i(t_0, \epsilon = 0.0)$.
Figure 4.4: This graph shows the normalisation constants for the proton (states 1-16) and neutron (states 17-32) single-particle states to demonstrate the numerical errors in the TDHF code and to assess in particular the reversability of the code. The states $\phi_i(t_0)$ were extracted at the time $t_0$ after the first iteration of the TDHF code and the states $\psi_i(t_0, \varepsilon = 0.0)$ were obtained after running the code forwards and backwards, $t_0 \rightarrow t_1 \rightarrow t_0$, whilst leaving the single-particle wavefunctions unchanged at $t_1$. 
4.3 Application of the BV approach to a GDR in $^{32}\text{S}$

4.3.2 Results of the Balian-Vénéroni Approach for a $^{32}\text{S}$ GDR

The single-particle wavefunctions from TDHF calculation were transformed according to (3.27) and time-reversed TDHF calculations were carried out for $\epsilon$ values of the order $10^{-1}$. For each value of $\epsilon$ $(\Delta N_{\text{BV}}(\epsilon))^2$ was calculated using (3.38) and the results obtained are plotted in figure 4.5. Extrapolating the linear portion of the curve back to the $y$-axis we obtain $(\Delta N_{\text{BV}})^2 \approx 5.92$ which is significantly larger than the TDHF result $(\Delta N_{\text{TDHF}})^2 = 4.09$ and suggests a $20\%$ increase in $\Delta N$. However, whilst this figure is consistent with the results of Troudet and Vautherin [48] for the calculation of the mass distribution for a giant monopole resonance in $^{40}\text{Ca}$ (see figure 1.2) it is not entirely consistent with the other two published results that used the Balian-Vénéroni approach, namely those of Marston and Koonin [30] and Bonche and Flocard [35]. These authors do not provide graphical evidence of the convergence of their results (and performed collisions and not resonance calculations) but indicate a use of $\epsilon$ values far smaller than those used in this calculation and in the calculations of Troudet and Vautherin ($\epsilon << 0.1$ in [30] and $\epsilon \approx 10^{-3} - 10^{-4}$ in the case of [35]).

An additional time-reversed calculation was carried out for $\epsilon = 0$ and the single-particle wavefunctions, $\phi_i(t_0)$, were replaced by the newly calculated single-particle wavefunctions $\psi_i(t_0, \epsilon = 0.0)$ in the evaluation of (3.38). In principle we would expect these sets of wavefunctions to be identical (we have already shown that they are not) in which case we would get the same results as before however in practice we find that we get very different results as shown in figure 4.6 where we...
4.3 Application of the BV approach to a GDR in $^{32}$S

Figure 4.6: $\Delta N_{BV}^2(\varepsilon)$ plotted as a function of $\varepsilon$ for $\varepsilon$ values in the range $10^{-7} \rightarrow 10^{-1}$ (note the logarithmic scale on the x-axis). This graph shows two sets of results where the "Improved" results were obtained by replacing $\phi_i(t_0)$ by $\psi_i(t_0, \varepsilon = 0.0)$ to eliminate systematic errors. The standard TDHF result (calculated at $t_1$ and independent of $\varepsilon$) is shown for reference.

show results for $\varepsilon$ values down to $1 \times 10^{-7}$. Using this second set of wavefunctions means that all of the wavefunctions used in evaluating (3.38) are the product of the same number of iterations of the TDHF code helping to eliminate any systematic errors from within the code. From these results we easily obtain $\Delta N_{BV}^2 = 5.5258$ and hence $\Delta N_{BV} = 2.3507(1)$ where we have averaged the results obtained for 9 values of $\varepsilon$ in the range $10^{-5} \rightarrow 10^{-3}$. The error is the standard error (calculated as the standard deviation of the set of values for $\Delta N_{BV}(\varepsilon)$ divided by the root of the number of results obtained [108, pg. 733]). This more accurate approach was also used by Bonche and Flocard [35, 42].

Figure 4.7 shows the real and imaginary parts of the diagonal elements of the matrix of overlaps, $\beta$ (see (3.38)), calculated using different combinations of the wavefunctions $\phi_i(t_0)$ and $\psi_i(t_0, \varepsilon)$ for the indicated values of $\varepsilon$. In the case of the real parts of the diagonal elements we plot their deviations from unity rather than their absolute values. The real parts of the overlaps are all almost exactly identical regardless of the value of $\varepsilon$ used in the transformation except for the first result where we used the the single-particle wavefunctions, $\phi_i(t_0)$. In this case we used, as the second set of single-particle wavefunctions, the set $\psi_i(t_0, \varepsilon = 0.0)$ such that these wavefunctions should be identical and we would expect $\text{Re} [\phi_i^*(t_0) \psi_i(t_0, \varepsilon = 0.0)] - 1.0 = 0.0$ and any deviations from zero represents numerical errors within the code. The fact that the other lines are all essentially identical indicates that the transformation (3.27) has only a small effect on the single-particle wavefunctions and that the code is deterministic to a high degree of accuracy. The difference between these lines
4.3 Application of the BV approach to a GDR in $^{32}$S

Figure 4.7: The real (top) and imaginary (bottom) parts of the single-particle wavefunction overlaps for the proton (states 1-16) and neutron (states 17-32) states calculated using several different sets of single-particle states. The states $\phi_i(t_0)$ were extracted at the time $t_0$ at the start of the first TDHF calculation whilst the states $\psi_i(t_0, \varepsilon)$ were obtained by performing a transformation at $t_1$ using the indicated value of $\varepsilon$ and running the code backwards to $t_0$ ($\varepsilon = 0.0$ is equivalent to omitting the transformation at $t_1$).
and the first lines indicates that the main errors in the code take the form of a numerical “drift”. Looking at the imaginary parts (where we would again expect \( \text{Im} [\phi^*_i (t_0) \psi_i (t_0, \epsilon = 0.0)] = 0.0 \)) we find that these values are non-zero and are an order of magnitude larger than the other results. This is unphysical, is a consequence of this numerical “drift” and, by comparison with the other results shown, demonstrates why the wavefunctions \( \phi_i (t_0) \) should not be used in evaluating (3.38). We also see that the other results all differ and scale with \( \epsilon \). The differences between these latter results demonstrates the physically significant consequences of the transformation (3.27) and, when (3.38) is evaluated, are responsible for the final, constant, value of \( \Delta N_{BV} \).

In the previous calculations of Marston and Koonin [30] the final value of \( (\Delta N_{BV} (\epsilon))^2 \) was determined by evaluating equation (2.167) whilst omitting the division by \( \epsilon^2 \) for a range of values of \( \epsilon (\epsilon < 10^{-1}) \). The results were plotted and fitted with a curve of the form \( f (\epsilon) = C_0 + C_1 \epsilon + C_2 \epsilon^2 \) such that \( C_2 = (\Delta N_{BV})^2 \) and any deviations from zero in \( C_0 \) and \( C_1 \) represented numerical noise. A similar quadratic regression has been performed using our new results and, using the results for \( \epsilon = 10^{-5} \rightarrow 10^{-3} \), we obtained \( C_0 = 0.0000, C_1 = 0.0000 \) and \( C_2 = 5.5274 \) which gives \( \Delta N_{BV} = 2.3510 \). The R-squared coefficient of determination for the fit was \( R^2 = 1.0000 \) reaffirming (as expected from figure 4.6) that these values provide an excellent fit to the data.

This is quite surprising both in terms of the difference between the two results (figures 4.5 and 4.6) and also the numerical stability of \( \Delta N_{BV} (\epsilon) \) across several orders of magnitude in \( \epsilon \). In particular for \( \epsilon \) values of order \( 10^{-4} \) we are looking at the 8th decimal place in the numerator in (3.38). Given that the deviations from orthonormality of the single-particle particle wavefunctions are typically of the order \( 10^{-3} \rightarrow 10^{-4} \) it is encouraging that this level of consistency has been obtained. These results also indicate that an accurate result can be obtained by only calculating \( \Delta N_{BV} (\epsilon) \) for several suitable values of \( \epsilon \) (of order \( 10^{-4} \)) rather than performing calculations for a much larger and wider range of \( \epsilon \) values (as would be needed if it was always necessary to obtain \( \Delta N_{BV} \) using a quadratic regression).

In the sections which follow we investigate the sensitivity of our results to the different parameters of our model. This will allow us to draw conclusions about the robustness of our implementation and identify those parameters which have the greatest impact upon our results.

4.3.3 Dependence on \( R_c \)

The calculations for the giant dipole resonance in \( ^{32}\text{S} \) have been repeated for the cutoff radii \( R_c = 8.5 \text{ fm} \) and \( R_c = 9.0 \text{ fm} \) to verify that the precise value of \( R_c \) does not significantly affect the results provided a sensible value is chosen, large enough to fully enclose the decaying nucleus but small enough to exclude most of the spatial box containing the emitted nucleons. The results are shown in figure 4.8 and summarised in table 4.5. In the previous calculations the results were given to a large number of significant figures mainly to demonstrate the consistency in the calculated
4.3 Application of the BV approach to a GDR in $^{32}$S

| $R_c$ (fm) | $\langle N \rangle |_{t_1}$ | $\Delta N_{TDHF} |_{t_1}$ | $(\Delta N_{TDHF})_{\text{MAX}} |_{t_1}$ | $\Delta N_{BV} |_{t_1}$ | Change |
|------------|-----------------|-----------------|-----------------|-----------------|------|
| 8.0        | 26.64           | 2.02            | 2.11            | 2.35            | +16% |
| 8.5        | 26.73           | 2.01            | 2.10            | 2.33            | +16% |
| 9.0        | 26.90           | 1.99            | 2.07            | 2.29            | +15% |

Table 4.5: The dependence of $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ on $R_c$ for the $^{32}$S giant dipole resonance calculations.

Figure 4.8: $(\Delta N_{BV})^2$ plotted as a function of $\varepsilon$ for different values of $R_c$. The TDHF results (calculated at $t_1$ and independent of $\varepsilon$) are shown for comparison.

values of $\Delta N_{BV}$ ($\varepsilon$) and to allow errors to be shown. In this, and future, sections the errors will only be given where they are comparable in size to the precision of the results. There are some small differences in $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ however, as shown in figure 4.8, the essential behaviour and trends remain unchanged and the percentage increase between $\Delta N_{BV}$ and $\Delta N_{TDHF}$ remains essentially constant. The trends in $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ are also consistent with what we would expect since as we increase $R_c$ we take in increasing amounts of the tails of the wavefunctions (and unavoidably pick up additional parts of the emitted flux). This explains the increasing values of $\langle N \rangle$ whilst the fluctuations, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ both decrease since, as $R_c \to \infty$, we require $\Delta N_{TDHF}, \Delta N_{BV} \to 0$. For $R_c = \infty$ the transformation (3.27) becomes a phase factor and as such would have no affect on the evolution of the system leading directly to $\Delta N_{BV} = 0$. 
4.3 Application of the BV approach to a GDR in $^{32}$S

4.3.4 The Effect of the Sharp Cutoff at $r = R_c$

The mass of the nucleus of interest is obtained by integrating the single-particle wavefunctions over a small spherical volume centred on the nucleus. Non-zero components of the single-particle wavefunctions within that region contribute to the mass of the nucleus. In the transformation (3.27) this region is defined by the theta function $\theta(R_c - |r - \bar{r}_{CM}|)$, which provides a sharp cutoff at the edge of this region. This is perfectly acceptable when determining the expectation value for the mass of the nucleus (or the mass dispersion) using the standard TDHF approach, however, with the Balian-Vénéroni approach care must be taken to ensure that this sharp cutoff does not cause numerical problems since it determines the region in which the transformation (3.27) is applied. This transformation could potentially introduce discontinuities into the single-particle wavefunctions and the densities (and their derivatives) across this boundary which would then adversely affect the time-reversed calculations. If $R_c$ is sufficiently large then the single-particle wavefunctions at this distance from the nucleus should be minimal and any discontinuities introduced through the use of the sharp cutoff should also be negligible.

To demonstrate this additional calculations were performed in which the theta function was replaced by either a stepping function, $\chi_1(|r - \bar{r}_{CM}|, R_c, R_t)$, or by a linearly decreasing function, $\chi_2(|r - \bar{r}_{CM}|, R_c, R_t)$. These functions are shown schematically in figure 4.9.

The results of these calculations are shown in table 4.6. These show results that changing the
### Table 4.6: The dependence of $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ on the form of the spatial cutoff function (and on $R_c$ and $R_t$). The function $\theta (R_c - |r - r_{CM}|)$ provides a sharp cutoff at $r = R_c$. The function $\chi_1 (r, R_c, R_t)$ provides a cutoff which decreases in steps whilst the function $\chi_2 (r, R_c, R_t)$ provides a linear cutoff. $R_t$ controls the sharpness of the cutoff functions $\chi_1 (r, R_c, R_t)$ and $\chi_2 (r, R_c, R_t)$ as shown schematically in figure 4.9. These results values were all calculated for $t_1 = 250$ fm/c using the SLy6 parameterisation of the Skyrme force.

| Function | $R_c$ (fm) | $R_t$ (fm) | $\langle N \rangle|_{t_1}$ | $\Delta N_{TDHF}|_{t_1}$ | $\Delta N_{BV}|_{t_1}$ | Change |
|----------|------------|------------|----------------|----------------|----------------|--------|
| $\theta (R_c - r)$ | 8.0 | - | 26.64 | 2.02 | 2.35 | +16% |
| $\chi_1 (r, R_c, R_t)$ | 8.0 | 0.25 | 26.62 | 2.02 | 2.33 | +15% |
| $\chi_2 (r, R_c, R_t)$ | 8.0 | 0.25 | 26.63 | 2.02 | 2.34 | +16% |
| $\theta (R_c - r)$ | 9.0 | - | 26.90 | 1.99 | 2.29 | +15% |
| $\chi_1 (r, R_c, R_t)$ | 9.0 | 0.25 | 26.90 | 1.99 | 2.27 | +14% |
| $\chi_2 (r, R_c, R_t)$ | 9.0 | 0.25 | 26.90 | 1.99 | 2.28 | +14% |

4.3.5 Dependence on the size of the spatial box

To assess the effect of the reflected flux on our results a set of calculations have been carried out using an enlarged spatial box, $40 \times 40 \times 40$ fm instead of $32 \times 32 \times 32$ fm. This increase doubles the volume of the spatial box. All of the other parameters of the model were left unchanged ($R_c = 8.0$ fm, $t_1 = 250$ fm/c ...etc...). This calculation yielded $\langle N \rangle = 26.45$ and $\Delta N_{TDHF} = 2.05$ (compared with $\langle N \rangle = 26.64$ and $\Delta N_{TDHF} = 2.02$ for the smaller spatial box). Since the only difference between these calculations was the size of the spatial box the difference between these two values of $\langle N \rangle$ provides evidence that a small amount of mass has re-entered the region of interest after reflection from the boundary and also provides a measure of that amount. Figure 4.10 shows the time evolution of $\langle N \rangle$ for both sizes of spatial box. A series of time-reversed calculations were performed and we obtained $\Delta N_{BV} = 2.39$ (compared with $\Delta N_{BV} = 2.35$ for the smaller spatial box) which represents an increase of 17% and is comparable to the 16% increase obtained using the smaller spatial box. This means that whilst a small amount of mass did re-enter the region of interest, leading to a small change in the calculated value of $\Delta N_{BV}$, this did not affect the convergence of $\Delta N_{BV} (\epsilon)$ with $\epsilon$ and there was no evidence of numerical instabilities.

4.3.6 Dependence on $\Delta r$

A set of calculations have also been carried out for $\Delta r = 0.8$ fm. These calculations were carried out using the same parameters as in the previous calculations ($R_c = 8.0$ fm, $t_1 = 1250$ fm/c, SLy6 Skyrme parameterisation). To accommodate the new value of $\Delta r$ the dimensions of the spatial
4.3 Application of the BV approach to a GDR in $^{32}$S

Figure 4.10: The number of nucleons in the $^{32}$S nucleus, calculated by integrating over all points within $R_c = 8.0$ fm of the centre-of-mass of the nucleus and plotted against time using two different sizes of spatial box.

<table>
<thead>
<tr>
<th>$\Delta r$ (fm)</th>
<th>BE</th>
<th>RMS Radius (fm)</th>
<th>$\beta_2$</th>
<th>$\gamma$ (°)</th>
<th>S (P)</th>
<th>S (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>260.36</td>
<td>3.175</td>
<td>0.111</td>
<td>2.3</td>
<td>7.354</td>
<td>13.086</td>
</tr>
<tr>
<td>0.8</td>
<td>259.60</td>
<td>3.175</td>
<td>0.109</td>
<td>1.9</td>
<td>7.316</td>
<td>13.101</td>
</tr>
<tr>
<td>Expt. [104–106]</td>
<td>271.78</td>
<td>3.248(11)</td>
<td>0.249(8)</td>
<td>-</td>
<td>8.864</td>
<td>15.042(2)</td>
</tr>
</tbody>
</table>

Table 4.7: The bulk properties of a $^{32}$S nucleus calculated using the OAK3D HF code with the SLy6 Skyrme parameterisation and different values of the grid spacing, $\Delta r$, and compared to the experimental values. $S(P)$ and $S(N)$ are the proton and neutron separation energies. All energies are in MeV.

grid for the dynamic calculation were changed to $-15.6 \rightarrow 15.6$ fm (40 nodes) instead of the usual $-15.5 \rightarrow 15.5$ fm (32 nodes). This represents a 95% increase in the number of nodes in the spatial box. For this calculation it was also necessary to repeat the static HF calculation to obtain a set of starting wavefunctions compatible with the smaller grid spacing. For the purposes of this static calculation the dimensions of the spatial box were set to $-9.2 \rightarrow 9.2$ fm (24 nodes) compared to the previous $-9.5 \rightarrow 9.5$ fm (20 nodes), which represents a 73% increase in the number a nodes in the spatial box. All other parameters were left unchanged. Table 4.7 compares the bulk properties of the $^{32}$S ground state for $\Delta r = 0.8$ fm and $\Delta r = 1.0$ fm to demonstrate that they were not significantly altered by the reduction in the grid spacing.

After 250 fm/c the nucleus contained 26.64 nucleons with $\Delta N_{TDHF} = 2.02$.
4.3 Application of the BV approach to a GDR in $^{32}\text{S}$

a series of time-reversed calculations we obtained $\Delta N_{BV} = 2.35$ These results are the same as those obtained for $\Delta r = 1.0$ fm and demonstrate that a step size of $\Delta r = 1.0$ fm provides us with sufficient accuracy.

4.3.7 Dependence on $t_1$

Once the excitation energy of the nucleus drops below the threshold for further particle emission we would expect the nucleus to remain stable and constant and as such we would expect $\langle N \rangle$ and $\Delta N$ for the nucleus to then remain constant with time. However, as previously discussed, in practice the emitted nucleons are reflected from the boundaries of the box and, after a time, interact again with the decayed nucleus. To investigate the effect of varying $t_1$ on our results a series of calculations have been carried out for a range of values of $t_1$ up to 2000 fm/c.

Figure 4.11 shows how $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ varies with $t_1$ from very small values of $t_1$ up until $t_1 = 2000$ fm/c. We see the first clear evidence of reflected nucleons just before $t = 400$ fm/c where we see a temporary increase in the mass of the nucleus. This is unphysical and must be due to nucleons having been reflected back from the boundary of the spatial box. These reflected nucleons will interact with the nucleus so that any results after this time will include an error due to these interactions. This is particularly important in the Balian-Vénetroni method where we expect to be more sensitive to numerical errors and noise.

This graph shows how for very small values of $t_1$, both $\Delta N_{TDHF}$ and $\Delta N_{BV}$ converge towards zero as $t_1 \to 0$ as we would expect. Once the nucleus has decayed, the values for $\Delta N_{TDHF}$ remain constant as a function of $t_1$ whereas the values of $\Delta N_{BV}$ show a much greater dependence on $t_1$, becoming increasingly erratic for larger values. Looking closely we see that just before 400 fm/c, where the mass of the nucleus shows an unphysical increase, $\Delta N_{BV}$ also increases. It is clear that the calculated values of $\Delta N_{BV}$ should not be trusted after this point.

4.3.8 Dependence on $\Delta t$

As an additional check of the accuracy of the current calculations a set of calculations have been carried out with a timestep of 0.1 fm/c, as opposed to the 0.2 fm/c timestep used in most of these calculations. These calculations were carried out with all the other calculation parameters (SLy6 Skyrme parameterisation, $R_c = 8.0$ fm ...etc...) left unchanged. After 250 fm/c the $^{32}\text{S}$ nucleus contained 26.64 nucleons with $\Delta N_{TDHF} = 2.02$ These are the same as the results obtained for $\Delta t = 0.2$ fm/c. Sets of time-reversed calculations were carried out as before and provided $\Delta N_{BV} = 2.36$ (compared with 2.35 for $\Delta t = 0.2$ fm/c). The differences in this value is less than 1% indicating that the current timestep $\Delta t = 0.2$ fm/c is small enough to provide stable results for $\Delta N_{BV}$. 
Figure 4.11: The expectation value $\langle N \rangle$ and the fluctuations, $\Delta N_{T_{\text{DHF}}}$ and $\Delta N_{\text{BV}}$, calculated using a cutoff radius $R_c = 8.0$ fm and plotted against $t_1$ for a $^{32}$S GDR for a large range of values of $t_1$. In principle the fluctuations should initially increase as the nucleus decays and then stabilise remaining constant thereafter. Any further features are a consequence of numerical instabilities and the interactions between the nucleus and the emitted nucleons that have been reflected back from the boundary of the spatial box.
4.3.9 Dependence on the Skyrme Parameterisation

All the calculations carried out in this work up until this point have been carried out using the SLy6 parameterisation of the Skyrme interaction. However, over the years a number of different parameterisations have been proposed and the choice of parameterisation does have an impact upon the results, as has already been seen in the range of values for the bulk properties of the $^{32}$S ground state (table 4.1). We have already investigated the numerical stability of the Balian-Vénéroni approach, implemented using the OAK3D code, by varying the various parameters of the model ($R_c$, $t_1$, ...etc...). Any additional differences in the results due to using a different Skyrme parameterisation (beyond those that can be attributed to numerical errors) must be a consequence of the different physics introduced through the use of that parameterisation. This makes it important to investigate the effect on our results of using a different Skyrme parameterisation in our calculations.

The calculations carried out thus far have shown that the model parameters most likely to affect our results are those that can be clearly linked to the problem of the reflection of emitted nucleons from the boundary of the spatial box. This problem can be investigated by varying either the size of the spatial box or $t_1$ (since the problem is reduced by either reducing $t_1$ or by increasing the size of the box). It is more efficient to vary $t_1$ since varying the size of the spatial box means performing a larger number of time-dependent calculations.

Sets of calculations have been carried out using the SkM*, SLy4 and SLy4d Skyrme parameterisations, the parameters for which were given in table 4.2. The bulk properties of the $^{32}$S ground state, calculated using these interactions, were given in table 4.1. All of the other parameters of the model took the values that have already been found to give sensible results when using the SLy6 parameterisation (a sharp cutoff at $|\rho| = R_c$, $R_c = 8.0$ fm, ...etc...).

Table 4.8 shows the expectation values and fluctuations (as well as the appropriate results from the SLy6 parameterisation for comparison) calculated for $t_1 = 220$, $250$ and $280$ fm/c. From these results we can see that our values for $\Delta N_{BV}$ appear to be far more sensitive to the choice of Skyrme parameterisation than they are to any of the other parameters of our model, the SkM* parameterisation in particular predicting less than half the increase in $\Delta N$ compared with the other parameterisations. This is important because it indicates that our calculations are sensitive to the subtly different physics included within the different Skyrme parameterisations (as a consequence of the different sets of data that were used to fit each parameter set) which could one day allow calculations of fluctuations to be used to fit and/or choose between different parameterisations.
### Table 4.8: A comparison of $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ using different parameterisations of the Skyrme interaction. These calculations were all carried out using $R_c = 8.0$ fm. In each calculation $\Delta N_{BV}(\varepsilon)$ was calculated for $\varepsilon = 0.0001$, 0.0004 and 0.0007 and the results averaged to obtain a final result and error.

<table>
<thead>
<tr>
<th>Parameterisation</th>
<th>$t_1$ (fm/c)</th>
<th>$\langle N \rangle$</th>
<th>$\Delta N_{TDHF}\mid_{t_1}$</th>
<th>$\Delta N_{BV}\mid_{t_2}$</th>
<th>Change</th>
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<tbody>
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<tr>
<td></td>
<td>250</td>
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<td>2.10</td>
<td>2.29</td>
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</tr>
<tr>
<td></td>
<td>280</td>
<td>26.18</td>
<td>2.10</td>
<td>2.28</td>
<td>+9%</td>
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<tr>
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<tr>
<td></td>
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<td>2.54</td>
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<tr>
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<td>2.02</td>
<td>2.35(1)</td>
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</table>
4.4 Summary

Averaging over all of the results that we have obtained using the SLy6 parameterisation we get $\Delta N_{BV} = 2.33$ (we exclude only those results obtained using parameters that differed significantly from those that have been identified as giving sensible results; i.e. exceptionally small or large values of $t_1$) where this result was obtained by averaging 66 individual results. Performing a similar averaging for $\langle N \rangle$ and $\Delta N_{TDHF}$ we obtain, after averaging 12 sets of results, $\langle N \rangle = 26.70(4)$ and $\Delta N_{TDHF} = 2.01(1)$. These results provide the best estimates for $\langle N \rangle$ and $\Delta N_{TDHF}$ however each calculation included in this average was used as the starting point for between 3 and 9 Balian-Vénérioni calculations which then contributed to the final value of $\Delta N_{BV}$. A fairer comparison with $\Delta N_{BV}$ is obtained by weighting the values of $\Delta N_{TDHF}$ to take this into account. In this case we obtain $\Delta N_{TDHF} = 2.01$ where the reduced error is a consequence of the weighting.

To perform a final comparison with the TDHF results and obtain a final answer for $\Delta N$ in this case we have averaged all of our results for the $^{32}$S giant dipole resonance (including those calculated with different Skyrme parameterisations) and we obtain a final mass for the nucleus of $\langle N \rangle = 26.54(5)$ with a mass fluctuation of $\Delta N_{BV} = 2.36(1)$ which is an average of 93 values and represents a 16% increase in $\Delta N$ when compared with the (weighted) TDHF average, $\Delta N_{TDHF} = 2.03$.

The Balian-Vénérioni approach has been implemented using the OAK3D code and a large number of calculations of the mass fluctuations for a giant resonance in $^{32}$S decaying by particle emission have been carried out. We have investigated the dependence of these results upon all of the parameters of our model and found that our code is accurate enough to obtain a reliable result using the Balian-Vénérioni approach and that these results are consistently larger than the equivalent TDHF results. We have found that the most important parameter in our model is the length of the calculations, $t_1$ (or, equivalently, the size of the box) due to the need to minimise the impact of reflected nucleons on our results. Varying all of the other model parameters, within sensible limits, leads only to small (and in our opinion acceptable) variations in the results and does not cause any instabilities or other numerical problems. We have also found that our results are sensitive to the different physics introduced through the use of different Skyrme parameterisations which is important since it could one day allow fluctuations to be used to tune and/or discriminate between different parameterisations.
Chapter 5

Resonance Calculations

5.1 Isoscalar Giant Monopole Resonance in $^{40}\text{Ca}$

A set of calculations have been carried out to calculate the mass distribution following the decay of an isoscalar giant monopole resonance in $^{40}\text{Ca}$. This case has been chosen to allow a direct comparison with the first implementation of the Balian-Vénérioni approach by Troudet and Vautherin [48]. As discussed in the last chapter there is an inconsistency in that this calculation only used $\epsilon$ values of order $10^{-1}$ whereas in the other implementations of the Balian-Vénérioni approach, and in the current implementation, much smaller values of $\epsilon$ ($10^{-3} - 10^{-4}$) were needed. This leads us to the conclusion that the results presented previously were subject to large numerical errors and should be re-checked.

Their calculation was carried out using a spherically symmetric TDHF code (although this shouldn’t be a limiting factor for this resonance and nucleus) and a BKN interaction [49]. The BKN force may be regarded as a simplified Skyrme force in that it’s based on the Skyrme functional but only the $t_0$ and $t_3$ parameters are non-zero. In their calculations the nucleus was excited by including an extra term in the mean-field potential (of the form $\lambda r^2$ with $\lambda = 10 \text{ MeV/fm}^2$) during the HF calculation which had the effect of squeezing the nucleus. This additional contribution to the potential was removed at the start of the TDHF calculation. In Troudet and Vautherin’s calculation the state of the system was calculated every $0.75 \text{ fm/c}$ until $t_1 = 60 \text{ fm/c}$ and they obtained (using a cutoff radius, $R_c = 8.0 \text{ fm}$) $\langle \tilde{N} \rangle = 33.10$, $\Delta N_{TDHF} = 2.30$ and $\Delta N_{BV} = 4.28$.

As before a set of HF calculations have been carried out to determine the ground state for the $^{40}\text{Ca}$ nucleus using several common Skyrme parameterisations. Whilst we would expect the ground state for this even-even doubly magic nucleus to be spherical we still used a tri-axial starting set of harmonic oscillator states. These states were described by $R_x = 3.0 \text{ fm}$, $R_y = 3.1 \text{ fm}$ and $R_z = 3.2 \text{ fm}$ which results in a starting deformation of $\beta_2 = 0.059$ and $\gamma = 29.47^\circ$. The spatial box was defined to be cubic with the dimensions $-12.5 \rightarrow 12.5 \text{ fm}$ and with points every $1 \text{ fm}$ for a total
5.1 Isoscalar Giant Monopole Resonance in $^{40}$Ca

<table>
<thead>
<tr>
<th>Parameter Set</th>
<th>BE</th>
<th>RMS Radius</th>
<th>$\beta_2$</th>
<th>S (P)</th>
<th>S (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SkM*</td>
<td>342.619</td>
<td>3.398</td>
<td>$0.4473 \times 10^{-10}$</td>
<td>7.591</td>
<td>14.290</td>
</tr>
<tr>
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<td>$0.1204 \times 10^{-9}$</td>
<td>8.483</td>
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</tr>
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<td>$0.1079 \times 10^{-9}$</td>
<td>7.953</td>
<td>14.735</td>
</tr>
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<td>SLy6</td>
<td>336.249</td>
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<td>$0.7312 \times 10^{-10}$</td>
<td>8.385</td>
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<td>3.476(1)</td>
<td>-</td>
<td>8.328(0)</td>
<td>15.643(2)</td>
</tr>
</tbody>
</table>

Table 5.1: Some bulk properties of a $^{40}$Ca nucleus calculated using the OAK3D HF code and different parameterisations of the Skyrme interaction and compared to the experimental values. The rms radii are given in fm. $S (P)$ and $S (N)$ are the proton and neutron separation energies. All energies are in MeV.

Some of the bulk properties for the ground state of the $^{40}$Ca nucleus are given in table 5.1. As before we see a large spread in the predicted binding energies with the SLy4d parameterisation doing particularly badly. Skyrme parameterisations are generally fitted to reproduce the ground state properties of magic number nuclei so we would expect them to do well for the doubly magic nucleus $^{40}$Ca. This spread in the ground state binding energies is a consequence of the absence of the centre-of-mass corrections. All of the Skyrme forces produced a spherical ground state. Figure 5.1 shows the variation in the ground state energy, $\Delta E/E$ and the average fluctuation in the single-particle energies, $\Delta H_{HF}$, plotted as a function of iteration number for the SLy6 parameterisation to demonstrate the convergence of the calculation. We can see that this calculation converged much faster than the previous calculation for $^{32}$S (figure 4.1) which is another consequence of the fact that this is a doubly magic nucleus. This graph is similar to those obtained using the other parameterisations.

A dynamic calculation was carried out to simulate an isoscalar giant monopole resonance in $^{40}$Ca. The HF solution obtained using the SLy6 Skyrme interaction was used as the initial state of the $^{40}$Ca nucleus (which was positioned at the origin). For the dynamic calculation the spatial box was expanded to $-19.5 \rightarrow 19.5$ fm with points every 1.0 fm. The system was perturbed with an initial isoscalar monopole boost and then allowed to evolve with the state of the system being calculated every 0.2 fm/c for a total of 1400 timesteps (or 280 fm/c). The single-particle wavefunctions were written to sets of restart files after 220, 250 and 280 fm/c. Several TDHF calculations were performed and the strength of the monopole boost was tuned to reproduce the values for $\langle N \rangle$ and $\Delta N_{TDHF}$ obtained by Troudet and Vautherin [48].

Using a boost given by $A_m = 16.5 \text{ fm}^{-2}$ we obtained a final mass of 32.63 nucleons (16.63 neutrons and 16.00 protons) which is close to the value of 33.10 from [48]. The mass dispersion, calculated using the standard THDF result (2.6), was 2.29 and is virtually identical to the mass fluctuation...
5.1 Isoscalar Giant Monopole Resonance in $^{40}\text{Ca}$

Figure 5.1: The binding energy, $E$, $\Delta E/E$ and the average fluctuation, $\Delta H_{HF}$, in the single-particle energies plotted as a function of iteration number to demonstrate the convergence of the HF calculation for $^{40}\text{Ca}$ (using the SLy6 parameterisation of the Skyrme interaction). The experimental value of the ground state energy is shown as a comparison.

in [48], 2.30. Figure 5.2 shows the number of nucleons and the rms radii for the protons and the neutrons as well as the total mass of the nucleus plotted as a function of time during the decay of the resonance. These values were obtained using a cutoff radius $R_c = 8.0$ fm as in our previous calculations and the calculations of Troudet and Vautherin. Comparing this graph with the equivalent graph for the $^{32}\text{S}$ giant dipole resonance, figure 4.2, we see that the decay of the giant monopole resonance is much slower than the decay of the giant dipole resonance. This means that we must balance the need to run the calculations for long enough to allow the nucleus to decay against the need to guard against instability and errors caused by the reflection of emitted nucleons from the boundary. The rms radii show the initial expansion of the nucleus in response to the initial boost and the subsequent oscillations in the size of the nucleus during its decay. The periodicity of the rms radii ($\approx 70$ fm/c) suggests, if we assume harmonic motion, an excitation energy of $\approx 17.7$ MeV. This is consistent with the work of Kohl et al. who suggested a centroid energy of 17.5(1.0) MeV after looking at the inelastic scattering of electrons off $^{40}\text{Ca}$ and studying the angular distributions when the excited $^{40}\text{Ca}$ nucleus decayed by the emission of an $\alpha$-particle [109] (and also the work of Youngblood et al. who looked at the small angle inelastic scattering of $\alpha$-particles and estimated the peak energy to be 18.9(4) MeV [110]).

Standard ($\Delta t$ positive) calculations were also performed with the SkM*, SLy4 and SLy4d Skyrme parameterisations and the results for all of these calculations are shown in table 5.2. Looking at all of these results we see that, for the SkM*, SLy4 and SLy6 Skyrme interactions the mass of the
5.1 Isoscalar Giant Monopole Resonance in $^{40}$Ca

Figure 5.2: The number of protons, $\langle N_p \rangle$, and neutrons, $\langle N_N \rangle$, and the rms radii for the protons and the neutrons (defined by the cutoff radius $R_c = 8.0$ fm) plotted as a function of time during the decay of an isoscalar GMR in $^{40}$Ca.

Figure 5.2: The number of protons, $\langle N_p \rangle$, and neutrons, $\langle N_N \rangle$, and the rms radii for the protons and the neutrons (defined by the cutoff radius $R_c = 8.0$ fm) plotted as a function of time during the decay of an isoscalar GMR in $^{40}$Ca.

nucleus decreases going from $220 \rightarrow 250$ fm/c but, unphysically, increases going from $250 \rightarrow 280$ fm/c. This indicates that we are already running for as long as is safe and, to run for longer values of $t_1$, we must increase the size of the spatial box. The values for $\Delta N_{TDHF}$ also show a change in behaviour at this point having been continually increasing up until this point as the emitted nucleons spread out throughout the box.

Given the high degree of consistency obtained during the previous calculations for the $^{32}$S GDR calculations we will now (and in all future calculations unless indicated otherwise) limit ourselves to only calculating $\Delta N_{BV} (\varepsilon)$ for $\varepsilon = 0.0001, 0.0004$ and $0.0007$, calculating for additional values only where these results do not provide sufficient consistency. Sets of time-reversed calculations were performed for these values of $\varepsilon$, for each Skyrme force, and for values of $t_1$ of $220, 250$ and $280$ fm/c and these results are also shown in table 5.2. These results are far more consistent than the results for the $^{32}$S giant dipole resonance which is another consequence of the fact that the Skyrme forces were mostly fitted to data for magic number nuclei so this is less of a test of their predictive power. In particular we see that the results from the SkM* force, the oldest of the Skyrme forces in use, are consistent with the results from the other forces (probably due to it having been fitted to reproduce the properties of giant monopole resonances [103]) despite producing noticeably smaller fluctuations for the $^{32}$S GDR. We also see from the errors associated with these results that the variations in the results as a function of $t_1$, or due to varying the Skyrme force, far exceed the variations due to $\varepsilon$. Averaging all of these results we find $\langle N \rangle = 32.84(8)$ with $\Delta N_{BV} = 2.96(1)$ compared with $\Delta N_{TDHF} = 2.28(1)$ (which represents a 30% increase in $\Delta N$).
5.1 Isoscalar Giant Monopole Resonance in $^{40}\text{Ca}$

|       | $t_1$ (fm/c) | $\langle N \rangle$ | $\Delta N_{TDHF}|_{t_1}$ | $\Delta N_{BV}|_{t_1}$ | Change  |
|-------|--------------|----------------------|---------------------------|------------------------|---------|
| SkM*  | 220          | 32.90                | 2.28                      | 2.90                   | +27%    |
|       | 250          | 32.77                | 2.30                      | 2.98                   | +30%    |
|       | 280          | 32.78                | 2.29                      | 2.98                   | +30%    |
|       | Average:     | 32.81(4)             | 2.29(1)                   | 2.95(1)                |         |
| SLy4  | 220          | 32.66                | 2.30                      | 2.98                   | +30%    |
|       | 250          | 32.59                | 2.31                      | 3.07                   | +33%    |
|       | 280          | 32.61                | 2.31                      | 3.04                   | +32%    |
|       | Average:     | 32.62(2)             | 2.31                      | 3.03(1)                |         |
| SLy4d | 220          | 33.34                | 2.21                      | 2.82                   | +27%    |
|       | 250          | 33.26                | 2.22                      | 2.87                   | +30%    |
|       | 280          | 33.19                | 2.23                      | 2.89                   | +30%    |
|       | Average:     | 33.26(4)             | 2.22                      | 2.86(1)                |         |
| SLy6  | 220          | 32.68                | 2.30                      | 2.96                   | +29%    |
|       | 250          | 32.62                | 2.31                      | 3.06                   | +33%    |
|       | 280          | 32.63                | 2.30                      | 3.02                   | +31%    |
|       | Average:     | 32.65(2)             | 2.30                      | 3.02(1)                |         |

| Troudet and Vautherin [48] | 60 | 33.10 | 2.296 | 4.377 | +91% |

Table 5.2: Comparison of old and new results for $\langle N \rangle$, $\Delta N_{TDHF}$ and $\Delta N_{BV}$ for a $^{40}\text{Ca}$ GMR.
5.2 Isovector Giant Dipole Resonance in $^{132}$Sn

As a final example of a resonance calculation we consider an isovector giant dipole resonance in the heavier doubly magic but neutron rich nucleus $^{132}$Sn. As a doubly magic nucleus this may be regarded as a computationally simpler problem than an open-shell nucleus and as such this nucleus has been included in previous calculations of the strength functions of giant resonances [84, 111] and we can expect our results to be more consistent and reliable than for a mid-shell nucleus. A Hartree-Fock calculation was carried out as before using a range of Skyrme forces and, as before, we will, in discussion, focus on the results from the SLy6 parameterisation whilst also presenting results for the other Skyrme parameterisations. The resulting ground states were spherical with a small range in the calculated ground state binding energies and a particularly poor result from the SLy4d parameterisation. The convergence of the calculation for the SLy6 parameterisation is shown in figure 5.3.

A time-dependent calculation was carried out storing the single-particle wavefunctions after $t = 220, 250$ and $280$ fm/c. A strong boost (defined by $A_x = A_y = A_z = 600$ fm$^{-1}$) was applied at the start of this TDHF calculation. After $280$ fm/c the nucleus contained, using $R_c = 8.0$ fm, $(N) = 121.17$ nucleons (75.69 neutrons and 45.48 protons) representing the emission of $\approx 4.5$ neutrons and $\approx 4.5$ protons.

Figure 5.4 shows the time-dependence of the mass of the $^{132}$Sn nucleus during its decay whilst the dipole moments are shown in figure 5.5. The graph shows $Q_x$, $Q_y$ and $Q_z$ to be identical as expected for a spherical nucleus. This graph also displays a clear shoulder at around 40 fm/c which is a consequence of the 8.0 fm cutoff radius. Figure 5.4 shows a matching discrepancy at this point (an apparent oscillation in the mass of the nucleus). The periodicity of the dipole moments is $\approx 88$ fm/c which corresponds to a resonance peak energy of $\approx 14.1$ MeV. This is close to the experimentally measured value of 16.1(7) MeV [112] and is the same as the value given in [84]. This is to be expected since those calculations were carried out using the same TDHF code and Skyrme

<table>
<thead>
<tr>
<th>Parameter Set</th>
<th>BE</th>
<th>RMS Radius</th>
<th>$\beta_2$</th>
<th>$S(P)$</th>
<th>$S(N)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SkM*</td>
<td>1113.56</td>
<td>4.8030</td>
<td>$0.312 \times 10^{-3}$</td>
<td>15.771</td>
<td>8.446</td>
</tr>
<tr>
<td>SLy4</td>
<td>1106.53</td>
<td>4.8056</td>
<td>$0.285 \times 10^{-3}$</td>
<td>15.739</td>
<td>7.640</td>
</tr>
<tr>
<td>SLy4d</td>
<td>1238.70</td>
<td>4.7462</td>
<td>$0.822 \times 10^{-5}$</td>
<td>18.433</td>
<td>8.158</td>
</tr>
<tr>
<td>SLy6</td>
<td>1099.71</td>
<td>4.8047</td>
<td>$0.235 \times 10^{-3}$</td>
<td>15.781</td>
<td>7.558</td>
</tr>
<tr>
<td>Expt. [104]</td>
<td>1102.85(1)</td>
<td>-</td>
<td>-</td>
<td>15.710(30)</td>
<td>7.311(25)</td>
</tr>
</tbody>
</table>

Table 5.3: Some bulk properties of a $^{132}$Sn nucleus calculated using the OAK3D HF code and different parameterisations of the Skyrme interaction and compared to the experimental values. The RMS Radii are given in fm. $S(P)$ and $S(N)$ are the proton and neutron separation energies. All energies are in MeV.
5.2 Isovector Giant Dipole Resonance in $^{132}$Sn

Figure 5.3: The binding energy, $E$, $\Delta E/E$ and the average fluctuation in the single-particle energies, $\Delta H_{HF}$, plotted as a function of iteration number to demonstrate the convergence of the HF calculation for $^{132}$Sn (using the SLy6 parameterisation of the Skyrme interaction).

Figure 5.4: The number of nucleons in the $^{132}$Sn nucleus, calculated by integrating the wavefunctions over all points with $R_c = 8.0$ fm of the centre-of-mass of the nucleus and plotted against time during the decay of the giant dipole resonance.
5.2 Isovector Giant Dipole Resonance in $^{132}$Sn

Figure 5.5: Dipole moments, $Q_x$, $Q_y$ and $Q_z$ for a GDR in $^{132}$Sn. The data points are marked with the crosses whilst smoothed curves have been added using cubic-splines to make it easier to estimate the periodicity of the dipole moments (for use in estimating the energy of the resonance).

A series of time-reversed calculations were carried out as before and the results are shown in table 5.4. Initially, significantly increased errors were obtained for the SLy6 $t_1 = 280$ fm/c calculation (for $\varepsilon = 0.0001, 0.0004$ and $0.0007$ we obtained $\Delta N_{BV} = 3.43(1)$). This result was rechecked to confirm that it was not a consequence of an inconsistency in an input file or caused by some other obvious error. The increased error was due to the values of $\Delta N_{BV} (\varepsilon)$ increasing as $\varepsilon$ decreased (previously, for the $^{32}$S GDR, this numerical breakdown did not appear until $\varepsilon \approx 10^{-6}$, see figure 4.6). Additional calculations were performed for $\varepsilon = 0.001, 0.004$ and $0.007$ producing far more consistent results as a function of $\varepsilon$. It is this improved result that is shown in table 5.4. However, even this updated result is not consistent with the results for $t_1 = 220, 250$ fm/c but we also see from table 5.4 that the values of $(N)$ start increasing after $t = 250$ fm/c which we know to be unphysical and a consequence of emitted nucleons being reflected back from the boundary of the spatial box. For these reasons additional calculations were performed for $t_1 = 240$ and 260 fm/c (the results of which are also shown in table 5.4) to better investigate the consistency and stability of these results as a function of $t_1$.

Calculations have also been carried out for the SkM*, SLy4 and SLy4d parameterisations for $t_1 = 220, 235, 250, 265$ and $280$ fm/c and the results of these calculations are also shown in
[Table 5.4: A comparison of $\Delta N_{TDHF}$ and $\Delta N_{BV}$ for a $^{132}\text{Sn}$ GDR. These values were calculated using $R_c = 8.0$ fm. The results marked with a $^+$ were obtained using $\varepsilon$ values of the order $10^{-3}$ instead of $10^{-4}$ and are discussed in the text.]

| Parameterisation | $t_1$ (fm/c) | $\langle N \rangle$ | $\Delta N_{TDHF}|_{t_1}$ | $\Delta N_{BV}|_{t_1}$ | Change |
|------------------|--------------|-------------------|-----------------|------------------|--------|
| SkM*             | 220          | 120.20            | 3.09            | 3.24             | +5%    |
|                  | 235          | 120.25            | 3.09            | 3.29             | +6%    |
|                  | 250          | 120.26            | 3.09            | 3.26             | +5%    |
|                  | 255          | 120.18            | 3.10            | 3.20             | +3%    |
|                  | 280          | 120.10            | 3.11            | 3.23             | +4%    |
| Average:         | 120.20(3)    | 3.09              | 3.24(1)         |                  |        |
| SLy4             | 220          | 120.49            | 2.99            | 3.44†            | +15%   |
|                  | 235          | 120.45            | 3.00            | 3.52             | +17%   |
|                  | 250          | 120.41            | 3.00            | 3.50             | +16%   |
|                  | 255          | 120.39            | 3.00            | 3.51             | +17%   |
|                  | 280          | 120.46            | 3.00            | 3.54             | +18%   |
| Average:         | 120.44(2)    | 3.00              | 3.51(1)         |                  |        |
| SLy4d            | 220          | 121.04            | 2.94            | 3.35             | +14%   |
|                  | 235          | 121.04            | 2.94            | 3.34             | +14%   |
|                  | 250          | 121.01            | 2.94            | 3.37             | +15%   |
|                  | 255          | 121.00            | 2.94            | 3.43             | +17%   |
|                  | 280          | 121.00            | 2.94            | 3.39             | +15%   |
| Average:         | 121.02(1)    | 2.94              | 3.38(1)         |                  |        |
| SLy6             | 220          | 121.10            | 2.94            | 3.34             | +14%   |
|                  | 240          | 121.06            | 2.94            | 3.37             | +15%   |
|                  | 250          | 121.02            | 2.94            | 3.36             | +14%   |
|                  | 260          | 121.11            | 2.94            | 3.36             | +14%   |
|                  | 280          | 121.17            | 2.93            | 3.42†            | +17%   |
| Average:         | 121.09(3)    | 2.94              | 3.37(1)         |                  |        |
5.2 Isovector Giant Dipole Resonance in $^{132}$Sn

Figure 5.6: $\Delta N_{BV}$ plotted against $\varepsilon$ for $^{132}$Sn GDR for $t_1 = 220$ fm/c and using the SLy4 parameterisation. The TDHF result, $\Delta N_{TDHF}$, calculated at $t_1$ and independent of $\varepsilon$ is shown for reference.

table 5.4. For the SLy4 $t_1 = 220$ fm/c calculation we initially obtained a very poor result using $\varepsilon$ values of order $10^{-4}$ due the $\Delta N_{BV}(\varepsilon)$ values dramatically increasing as $\varepsilon$ decreased (leading to $\Delta N_{BV} = 4.39(81)$). As before additional calculations were performed for $\varepsilon = 0.001, 0.004$ and 0.007 and we obtained a far more consistent result as shown in figure 5.6 which shows $\Delta N_{BV}(\varepsilon)$ plotted against $\varepsilon$ for this case. This problem was seen to a far lesser degree in the SLy4 $t_1 = 235$ fm/c result although this calculation was not repeated.

When we average all of these results we find $\langle N \rangle = 120.69(9)$ and $\Delta N_{BV} = 3.37(1)$ compared to $\Delta N_{TDHF} = 2.99(1)$, which represents a 13% increase in $\Delta N$. We find again that we get noticeable, but manageable, variations in our results as a function of $t_1$ and the choice of the Skyrme parameterisation but that we are able to obtain converged results for each parameter set (although this requires more care and work than in the previous cases). The SLy4, SLy4d and SLy6 parameterisations show a consistent increase when comparing $\Delta N_{TDHF}$ and $\Delta N_{BV}$ whilst the SkM* parameterisation produces a much smaller increase in $\Delta N_{BV}$. These results are consistent with those obtained for the $^{32}$S GDR (table 4.8). We find, for this heavier system, that we encounter more problems with numerical stability than we did in the earlier calculations.
Chapter 6

Collision Calculations

6.1 $^{16}\text{O}+^{16}\text{O}$ Collision ($E_{\text{LAB}} = 160 \text{ MeV}$)

We consider first a symmetric collision between two $^{16}\text{O}$ nuclei at $E_{\text{LAB}} = 160 \text{ MeV}$ as considered previously by Bonche et al. [35,42] (for $I = 30\hbar$) and Marston and Koonin (for $I = 0\hbar$ and $30\hbar$) [30]. The calculations of Bonche et al. were carried out using a three-dimensional code but the simple BKN interaction and assumed spin-isospin symmetry. The calculations of Marston and Koonin were carried out using the more realistic SkII interaction but a two-dimensional code with the "clutching" method to allow calculations to be performed for non-zero impact parameters. In both calculations a split spatial box approximation was made in that they considered the mass (and its associated fluctuation) for all nuclear matter in one half of their spatial box whereas in these calculations we will continue to consider a spherical region centred on the centre-of-mass of the fragment of interest. The results from these calculations were shown in table 1.1. As mentioned in the introduction the two calculations for $^{16}\text{O}+^{16}\text{O}$ ($I \approx 30\hbar$) produced significantly different results.

Bonche et al. implemented their time-reversed calculations by applying the time-reversal operator (complex conjugation of the single-particle states) to the single-particle states obtained at $t_0$ and then continuing the calculations until $2t_1$. They used $t_1 = t_0 + 18 \times 10^{-22}$ seconds with $\Delta t = 4 \times 10^{-24}$ seconds (which equates to $t_1 = t_0 + 540 \text{ fm/c}$ and $\Delta t = 1.2 \text{ fm/c}$). Systematic numerical errors were reduced by replacing the initial single-particle wavefunctions (taken from the original TDHF calculation at $t_0$) by those obtained by running a second calculation (with the time-reversed states) until $2t_1$ and applying the time-reversal operator but omitting the transformation (1.7). This more accurate approach is also used in the current work.

In the calculations of Marston and Koonin the calculations were terminated once the fragments had separated by about 10 fm. Whilst they indicated that their results were obtained for $\varepsilon < 0.1$ they gave no indication that they used the more accurate approach used by Bonche et al. and which we find to be essential. As previously discussed, Marston and Koonin obtained $\Delta N_{\text{BV}}$ by
6.1 $^{16}$O+$^{16}$O Collision ($E_{LAB} = 160$ MeV)

Figure 6.1: The binding energy, $E$, $\Delta E/E$ and the average fluctuation in the single-particle energies, $\Delta H_{HF}$, plotted as a function of iteration number to demonstrate the convergence of the HF calculation for $^{16}$O (using the SLy6 parameterisation of the Skyrme interaction).

Calculating the numerator of (3.38), dividing by two, and fitting the resulting points with a curve of the form $f(\varepsilon) = C_0 + C_1 \varepsilon + C_2 \varepsilon^2$. They only provide their final values of $\Delta N_{BV}$ and not the values of $C_0$ and $C_1$ from their fits, which would have provided a measure of the numerical errors present in their results. Given the extremely accurate nature of the results we have generally obtained to date this could be regarded as indicating that they did not use the more accurate approach used by Bonche et al. and the present author. The paper [30] does not provide enough details to be sure however if the more accurate approach was not used. This might explain how these calculations produced results with such a large variation in the percentage increases in $\Delta N_{BV}$ compared to $\Delta N_{TDHF}$ (e.g. +31% for the head-on $^{16}$O collision compared with +405% for the off-axis collision).

The ground state for the $^{16}$O nucleus was obtained from a HF calculation using the SLy6 Skyrme parameterisation and a starting set of tri-axial harmonic oscillator states as described previously. The calculated ground state binding energy was 118.2291 MeV (compared with the experimental value of 127.6193(0) MeV [104]) whilst the proton and neutron separation energies were 10.877 MeV and 14.073 MeV respectively (compared with the experimental values of 12.12741(0) MeV and 15.6639(5) MeV [104]). The calculated ground state was spherical ($\beta_2 = 0.18 \times 10^{-5}$) with an rms radius of 2.6915 fm. Figure 6.1 shows the evolution of the ground state binding energy, $E$, $\Delta E/E$ and the average fluctuation in the single particle energies, $\Delta H_{HF}$, to demonstrate that the ground state energy had converged before the limits of numerical accuracy were reached.

A series of TDHF calculations were carried out for $E_{LAB} = 160$ MeV ($E_{CM} = 80$ MeV) where the
two $^{16}$O nuclei were initially placed at $(\mp 10, 0, \pm b/2)$ and $b$ is the impact parameter (in fm) and took the values 0.0, 2.7, 5.4 and 8.1 fm. These values were chosen to enable a comparison with the previous calculations of Bonche and Flocard [35,42] and Marston and Koonin [30] (see table 1.1). They both performed calculations using the Balian-Vénéróni approach for this reaction and an impact parameter of $l = 30\hbar$ which, using [55]

$$l = b(2\mu E_{CM})^{1/3},$$

(6.1)

where $\mu$ is the reduced mass, is approximately equivalent to $b = 5.4$ fm. These initial positions ensure that the nuclei start far enough apart that they only interact weakly through long range Coulomb interactions. For the head-on collision the initial kinetic energies of the fragments were 37.663 MeV with a Coulomb repulsion of 4.614 MeV and the density at the origin was less that $10^{-6}$ times the density at the centre of the nuclei. For these dynamic calculations the spatial box was $-31.5 \rightarrow 31.5$ fm in the $x$-direction and $-15.5 \rightarrow 15.5$ fm in the $y$- and $z$-directions (with points every 1.0 fm).

The TDHF calculations for $b = 0.0$ fm and $b = 8.1$ fm resulted in two clearly separated fragments whilst for the collisions with $b = 2.7$ fm and $b = 5.4$ fm the projectile and target nuclei fused to produce an excited compound nucleus which then decayed by particle emission. The TDHF calculations for $b = 0.0$ and $b = 8.1$ fm were terminated shortly before the nuclei impacted upon the edge of the spatial box whilst the other two calculations were allowed to run for extended periods to allow the compound nucleus to decay and to ensure that it did not undergo fission. At the end of each calculation the number of nuclei in one of the fragments (the nucleus in the right hand side of the spatial box for the $b = 0.0$ fm and $b = 8.1$ fm collisions) was obtained. For the $b = 0.0$ fm and $b = 8.1$ fm calculations we used $R_c = 8.0$ fm whilst in the other calculations we used $R_c = 10.0$ fm to fully enclose the larger (deformed) compound nucleus.

**6.1.1 Time-dependent calculations for $b = 0.0$ fm ($l = 0\hbar$)**

For the $b = 0.0$ fm collision the simulation was allowed to run for 600 fm/c until the centres-of-mass of the two nuclei were separated by 23.824 fm and the nuclei were about to impact upon the boundary of the spatial box. At this time the kinetic energies of the two fragments were 3.613 MeV with a Coulomb repulsion of 3.867 MeV. A series of density plots showing how the nuclei evolved during this collision are shown in figure 6.2. Figure 6.3 shows the total fragment energy in centre-of-mass frame, made up from the kinetic energies associated with the motion of the two fragments with a contribution due to the Coulomb repulsion between them. The sharp reduction in this energy ($\approx 70$ MeV) between 100 and 200 fm/c is due to the conversion of this energy into excitation energy in the fragments ($\approx 35$ MeV each). We also show the time-dependence of the distance between the centres-of-mass of the fragments.
Figure 6.2: Density contour plots showing the density in the reaction (x-z) plane for a deep-inelastic head-on collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV. The arrows on the first figure show the directions of the boosts applied to the two nuclei.
Figure 6.3: The total energy in the centre-of-mass frame plotted as a function of time during the collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV and for $b = 0.0$ fm ($I = 0\hbar$). The total energy is the sum of the kinetic energies associated with the motion of the two primary fragments with a contribution due to the Coulomb repulsion between the fragments. These two contributions are also shown independently and as a function of time. The reduction in the centre-of-mass energy during the reaction is due to the conversion of the kinetic energy into excitation energy in the fragments. The shaded region indicates the period during which the nuclei were interacting were not clearly separated. No special meaning should be attached to any features or numerical artefacts appearing during this period. The separation between the centres-of-mass of the two nuclei is also plotted as a function of time.

<table>
<thead>
<tr>
<th>$t_1$ (fm/c)</th>
<th>$(N)$</th>
<th>$\Delta N_{TDHF}$</th>
<th>$\Delta N_{MAX}$</th>
<th>$\Delta N_{BV}$</th>
<th>Change</th>
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<td>+51%</td>
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<td>1.34</td>
<td>2.83</td>
<td>2.04</td>
<td>+52%</td>
</tr>
<tr>
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<td>1.31</td>
<td>2.83</td>
<td>1.88</td>
<td>+43%</td>
</tr>
</tbody>
</table>

$\text{Marston and Koonin [30]}$

\text{Table 6.1: Expectation values and fluctuations for the mass of one fragment following an $^{16}$O+$^{16}$O collision at $E_{LAB} = 160$ MeV and with $b = 0.0$ fm ($I = 0\hbar$).}$

\text{\textsuperscript{1}This final result was calculated by splitting the spatial box in half and assuming that everything in the right ($x > 0$) half of the box was part of the nucleus of interest (this approximation was used in the calculations of Marston and Koonin).}$

\text{\textsuperscript{*}This expectation value was not published.}
We calculated the mass of the nucleus and its associated fluctuations at $t_1 = 400, 450$ and $500$ fm/c (at which times the centres-of-mass of the two fragments were separated by 15.3, 17.4 and 19.5 fm respectively). These values of $t_1$ are comparable to the one used in the calculations of Bonche et al. (540 fm/c). The results of these calculations are shown in table 6.1. We also show, for $t_1 = 450$ fm/c, the results obtained if we use a split spatial box approximation as used in the calculations of Bonche et al. and Marston and Koonin which demonstrates that this has a noticeable but small effect on our results. In this calculation we would expect $\langle N \rangle = 16.0$ so the actual value $\langle N \rangle = 15.9971$, whilst correct to two decimal places, provides an indication of some of the numerical errors inherent in the calculation. In all of these calculations we obtained a good convergence in $\Delta N_{BV}(\epsilon)$ although we also note that the mass of the fragment increases between $t = 400$ and $450$ fm/c indicating that we are seeing the effects of reflected flux. We might expect this to cause less problems than previously due to the much smaller amount of dissipated mass, both as an absolute amount and as a fraction of the total mass in the system ($\approx 1$ nucleon per fragment), compared with the many nucleons seen in the previous calculations. We also see that our values for $\Delta N_{TDHF}$ and $\Delta N_{BV}$ are both significantly larger than the values obtained by Marston and Koonin and, whilst we did obtain converged and consistent results, the errors, whilst still small, are somewhat larger than were typically seen in the resonance calculations.

When we averaging the first three results (obtained for a spherical region centred on the fragment of interest) we obtain $\langle N \rangle = 15.17(11)$, $\Delta N_{TDHF} = 1.35(1)$ and $\Delta N_{BV} = 2.04(1)$ which represents a 52% increase in $\Delta N$. We defer any further discussion of these results until we have presented the results of the off-axis collisions.

### 6.1.2 Time-dependent calculations for $b = 2.7$ fm ($l \approx 15h$)

In this collision, and for $l \approx 30h$, the nuclei fused to form an excited compound nucleus at the origin (the position of the compound nucleus being guaranteed by the choice of initial positions and the fact that this is a symmetric reaction). In this collision the compound nucleus cannot impact upon the edge of the spatial box and these calculations were continued until 2000 fm/c to give the compound nucleus a chance to decay by particle emission or undergo fission. The final mass of the compound nucleus was 28.85 nucleons (14.14 protons and 14.72 neutrons).

A series of density plots showing the evolution of the system are shown in figure 6.4. Figure 6.5 shows the evolution of the mass of the compound nucleus (and also the rms radii for the protons and the neutrons) to demonstrate the decay of the excited compound nucleus and give an indication of the length of the decay process. The second plot in figure 6.5 shows $\Delta N_{TDHF}$ and $\Delta N_{BV}$ calculated for a range of values of $t_1$ as well as $\langle N \rangle$. Comparing this figure with those obtained previously for the resonance calculations (figure 4.2 for the $^{32}$S GDR, figure 5.2 for the case of the $^{40}$Ca GMR and figure 5.4 for the $^{132}$Sn GDR) we see that the decay of the compound nucleus takes place on a much longer time-scale than the decay of the resonances and has barely stabilised
Figure 6.4: Density contour plots showing the density in the reaction \((x-z)\) plane for a fusion-evaporation collision between two \(^{16}\text{O}\) nuclei at \(E_{\text{LAB}} = 160\text{ MeV}\) and for an impact parameter \(b = 2.7\text{ fm}\) (equivalent to \(t \approx 15\hbar\)). The arrows on the first figure show the directions of the boosts applied to the two nuclei.
Figure 6.5: Expectation values, fluctuations and rms radii plotted as a function of time during the collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV and for $b = 2.7$ fm ($l \approx 15\hbar$). These graphs were obtained by integrating over all points within 10.0 fm of the point $(0.0,0,0,0,0)$ to show the decay by particle emission of the excited compound nucleus formed during the collision. As such the data shown in this figure is only meaningful after the formation of the compound nucleus at $t \approx 125$ fm/c. The shaded region shows the period preceding the formation of the compound nucleus.
6.1 \(^{16}\text{O}+^{16}\text{O}\) Collision \((E_{\text{LAB}} = 160 \text{ MeV})\)

even after 2000 fm/c. The mass of the nucleus shows an unphysical increase after about 1700 fm/c which can only be due to the reflection of the emitted flux however \(\Delta N_{\text{TDFH}}\) only appears to stabilise after these long times. It has already been shown for the case of the \(^{32}\text{S}\) GDR that the results obtained using the Balian-Vénéron method can be adversely affected by the numerical problems associated with prolonged TDHF calculations (as was shown in figure 4.11). Whilst they are not really noticeable on this scale the points for \(\Delta N_{BV}\) include error bars to demonstrate that even though the different values of \(\Delta N_{BV}\) (for different \(t_1\)) do not agree with each other they were obtained from individual sets of calculations which converged (as a function of \(\varepsilon\)).

6.1.3 Time-dependent calculations for \(b = 5.4 \text{ fm} \ (1 \approx 30h)\)

A series of density plots showing the evolution of the system for this reaction are shown in figure 6.6 whilst figure 6.7 is analogous to figure 6.5 and shows the mass, fluctuations and rms radii plotted as a function of time for the compound nucleus. The final mass of the compound nucleus was 29.60 nucleons (14.48 protons and 15.13 neutrons). This particular calculation was performed to compare with the earlier calculations of Bonche et al. and Marston and Koonin however since our calculation produced a compound nucleus the results are not directly comparable. The mass of the nucleus shows an unphysical increase at about 1700 fm/c however it is only at around these times that \(\Delta N_{\text{TDFH}}\) seems to stabilise. It is clear that the values of \(\Delta N_{BV}\) are not at all consistent. Furthermore, it is very noticeable here that, unlike in the previous calculation, several of the points have large errors associated with them (where these errors are the standard errors after averaging the values of \(\Delta N_{BV} (\varepsilon)\) for \(\varepsilon = 0.0001, 0.0004\) and 0.0007). These errors are generally a consequence of one of the three values for \(\Delta N_{BV} (\varepsilon)\) differing significantly from the other two values although, unlike previously, we find here that the differing result is not consistently the result associated with the smallest value of \(\varepsilon\). Whilst these errors might be eliminated through re-checking some of these calculations and performing calculations for additional values of \(\varepsilon\) this would only be expected to produce a set of well converged values for \(\Delta N_{BV}\) and would not be expected to make these individual values of \(\Delta N_{BV}\) (for different \(t_1\)) agree with each other.

6.1.4 Time-dependent calculations for \(b = 8.1 \text{ fm} \ (1 \approx 45h)\)

In the \(b = 8.1 \text{ fm}\) collision the reaction was stopped after 400 fm/c as the nuclei scattered off each other and quickly approach the edges of the spatial box. A series of density plots showing the evolution of the nuclei during this collision are shown in figure 6.8. In this collision the nuclei scatter off each other with little interaction as demonstrated by figure 6.9 which shows the centre-of-mass energy and indicates that each nucleus was excited by less than 1.0 MeV. This is reflected in the results, shown in table 6.2, where we see that we get virtually no particle emission and very small fluctuations. The calculated values of \(\Delta N_{BV}\) are only slightly larger than the TDHF results but all the calculations converged and the results are also consistent (as a function of \(t_1\)). The very small amount of particle emission in this case may also be responsible for the small differences between
6.1 $^{16}$O + $^{16}$O Collision ($E_{LAB} = 160$ MeV)

Figure 6.6: Density contour plots showing the density in the reaction ($x$-$z$) plane for a fusion-evaporation collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV and for an impact parameter $b = 5.4$ fm. (equivalent to $l \approx 30\hbar$). The arrows on the first figure show the directions of the boosts applied to the two nuclei.
Figure 6.7: The expectation value $\langle N \rangle$ and rms radii plotted as a function of time during the collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV and for $b = 5.4$ fm ($l \approx 30\AA$). These graphs were obtained by integrating over all points within 10.0 fm of the point $(0,0,0,0,0)$ to show the decay by particle emission of the excited compound nucleus formed during the collision. As such the data shown in this figure is only meaningful after the formation of the compound nucleus at $t \approx 150$ fm/c. The shaded region shows the period preceding the formation of the compound nucleus.
Figure 6.8: Density contour plots showing the density in the reaction (x-z) plane for a deep-inelastic collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV for an impact parameter $b = 8.1$ fm. (equivalent to $l \approx 45\hbar$). The arrows on the first figure show the directions of the boosts applied to the two nuclei.
Figure 6.9: The total energy in the centre-of-mass frame plotted as a function of time during the collision between two $^{16}$O nuclei at $E_{LAB} = 160$ MeV and for $b = 8.1$ fm ($l \approx 45\hbar$). The total energy is the sum of the kinetic energies associated with the motion of the two primary fragments with a contribution due to the Coulomb repulsion between them. The slight reduction in the centre-of-mass energy during the reaction is due to the conversion of kinetic energy into excitation energy in the fragments. The shaded region indicates the period during which the nuclei were interacting. No special meaning should be attached to any features or numerical artefacts appearing during this period. The separation between the centres-of-mass of the two nuclei is also plotted as a function of time.

$\Delta N_{TDHF}$ and $\Delta N_{BV}$ since in the limit of no particle emission we expect $\Delta N_{TDHF} = \Delta N_{BV} = 0$.

6.1.5 Discussion

Comparing our results with the previous results (table 1.1) we find that our values for $\Delta N_{TDHF}$ are all significantly larger than the values from the previous calculations (all < 1). This is probably due to our use of a three-dimensional code combined with a modern interaction. The previous calculations of Bonche et al. were carried out with a three-dimensional code but their

<table>
<thead>
<tr>
<th>$t_1$ (fm/c)</th>
<th>$N$</th>
<th>$\Delta N_{TDHF}$</th>
<th>$\Delta N_{MAX}$</th>
<th>$\Delta N_{BV}$</th>
<th>Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>15.99</td>
<td>0.29</td>
<td>2.83</td>
<td>0.30</td>
<td>+4%</td>
</tr>
<tr>
<td>350</td>
<td>15.99</td>
<td>0.29</td>
<td>2.83</td>
<td>0.30</td>
<td>+5%</td>
</tr>
<tr>
<td>400</td>
<td>15.99</td>
<td>0.28</td>
<td>2.83</td>
<td>0.30</td>
<td>+5%</td>
</tr>
</tbody>
</table>

Table 6.2: Expectation values and fluctuations for the mass of one fragment following an $^{16}$O+$^{16}$O collision at $E_{LAB} = 160$ MeV and with 8.1 fm ($l \approx 45\hbar$).
6.2 \( ^{40}\text{Ca}+^{40}\text{Ca} \) Collisions \((E_{\text{LAB}} = 278 \text{ MeV})\)

BKN force included no spin-orbit forces which are known to result in increased dissipation in TDHF calculations \([31,38]\). This increased energy loss is associated with an increase in the energy threshold above which fusion does not occur \([32,113]\). The calculations of Marston and Koonin were performed with the more advanced SkII force which does include some spin-orbit interactions but with a two-dimensional code. In the fusion-evaporation reaction calculations \(\Delta N_{TDHF} \) takes a long time to reach its final value but this is not so clear in the deep-inelastic scattering reactions. In the scattering reactions we are limited in the range of values of \( t_1 \) that we can use. We must wait until after the nuclei have clearly separated, we generally require that the separation of their centres-of-mass be of order \( 2R_c \) or greater, but must also end the calculations before the nuclei reach the edge of the spatial box. For a spatial box of manageable size this only provides a small range of values of \( t_1 \) that can be used.

The mass emitted in these reactions, whilst less than in the resonances calculations, is consistent with the earlier calculations of Guo et al. \([85]\) for this system. They performed calculations of this reaction for \( E_{CM} = 25 \text{ MeV} \) \((b = 2.8 \text{ fm}, l \approx 8.6\hbar)\) and \( E_{CM} = 125 \text{ MeV} \) \((b = 2.8 \text{ fm}, l \approx 33\hbar)\) and saw 0.4 nucleons emitted in their lower energy collision and 1.7 nucleons in their higher energy collision. They investigated the conservation of angular momentum within the system and how this was affected when the emitted nucleons reached the edge of their box. They found that they could only run until about 250 fm/c for a spatial box of size 24 \( \times \) 32 \( \times \) 32 fm or 350 fm/c for a box of size 48 \( \times \) 64 \( \times \) 64 fm before they saw significant problems. Our spatial box is larger than their smaller spatial box but smaller than their larger one and we, in all cases, are running for far longer than they found to be safe. We have seen previously that the values of \( \Delta N_{TDHF} \) are quite robust and resistant to developing significant errors due to these reflected nucleons but the same cannot be said of \( \Delta N_{BV} \) (figure 4.11). In Guo's calculations it was found that the problems caused by the reflected nucleons could be managed with absorbing boundary conditions however, as previously mentioned, we cannot use these techniques (at least in their current forms) since we need our calculations to be reversible.

6.2 \( ^{40}\text{Ca}+^{40}\text{Ca} \) Collisions \((E_{LAB} = 278 \text{ MeV})\)

A series of calculations have also been carried out for the symmetric collision of two \( ^{40}\text{Ca} \) nuclei at \( E_{LAB} = 278 \text{ MeV} \). We consider the impact factors 0.0 fm and 2.6 fm (equivalent to \( l = 30.09276 \hbar \)) where this second impact factor was chosen to reproduce the orbital angular momentum of 30\hbar used in the previous calculations of Marston and Koonin. We use the \( ^{40}\text{Ca} \) ground state which we calculated previously using the SLy6 parameterisation and used in the \( ^{40}\text{Ca} \) GMR calculations. The properties of this ground state were summarised in table 5.1. In these dynamic calculations a large spatial box of size 80 \( \times \) 40 \( \times \) 40 fm, centred on the origin, was used. As before the nuclei were initially positioned at \((\mp 10, 0, \pm b/2)\). Density plots showing the dynamics of the collisions are shown in figures 6.10 (for the head-on collision) and 6.12 (for the off-axis collision). Figure
Figure 6.10: Density contour plots showing the density projected onto the reaction (x-z) plane for a deep-inelastic collision between two \(^{40}\text{Ca}\) nuclei at \(E_{LAB} = 278\ \text{MeV}\) and for a head-on collision. The arrows on the first figure show the directions of the boosts applied to the two nuclei.
Figure 6.11: The total energy in the centre-of-mass frame plotted as a function of time during a head-on collision between two $^{40}$Ca nuclei at $E_{LAB} = 278$ MeV. The total energy was obtained by summing the kinetic energies associated with the motion of the two primary fragments with a contribution due to the Coulomb repulsion between them. These two contributions are also shown as a function of time. The reduction in the centre-of-mass energy during the reaction is due to the conversion of kinetic energy into excitation energy in the fragments. The shaded region indicates the period during which the nuclei were interacting and were not clearly separated. No special meaning should be attached to any features or numerical artefacts appearing during this period. The separation between the centres-of-mass of the two nuclei is also plotted as a function of time.
### Table 6.3: Expectation values and fluctuations for the mass after the head-on and off-axis \(^{40}\text{Ca}+^{40}\text{Ca}\) collisions at \(E_{\text{LAB}} = 278\) MeV.

<table>
<thead>
<tr>
<th>(t_1) (fm/c)</th>
<th>(b) (fm)</th>
<th>(\langle N\rangle)</th>
<th>(\Delta N_{\text{TDHF}})</th>
<th>(\Delta N_{\text{BV}})</th>
<th>Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>900</td>
<td>0.0</td>
<td>38.97</td>
<td>1.55</td>
<td>3.47(1)</td>
<td>+124%</td>
</tr>
<tr>
<td>950</td>
<td>0.0</td>
<td>38.95</td>
<td>1.55</td>
<td>1.99(1)</td>
<td>+28%</td>
</tr>
<tr>
<td>1000</td>
<td>0.0</td>
<td>38.94</td>
<td>1.55</td>
<td>1.96</td>
<td>+26%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Marston and Koonin [30]</th>
</tr>
</thead>
<tbody>
<tr>
<td>800</td>
</tr>
<tr>
<td>900</td>
</tr>
<tr>
<td>1000</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Marston and Koonin [30]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.26</td>
</tr>
</tbody>
</table>

6.11 shows the total centre-of-mass energy and the separation between the fragments, plotted as a function of time, during the head-on collision. In the off-axis collision the two nuclei fused to form an excited compound nucleus with a final mass, after 2000 fm/c, of 76.8019 nucleons (37.7646 protons and 39.0372 neutrons). In the calculations for the head-on collision a cutoff radius of 8.0 fm was used whilst for the off-axis collision a cutoff radius of 10.0 fm was used to ensure that the deformed compound nucleus was fully contained within the region of interest. This is the first example where we see such a clear difference in the number of protons and neutrons emitted which is a consequence of the proton rich nature of the composite nucleus, \(^{80}\text{Zr}\) (the most abundant stable Zirconium isotope being \(^{90}\text{Zr}\)). Figure 6.13 shows the mass and \(\Delta N_{\text{TDHF}}\) for the composite nucleus formed during the off-axis collision as a function of time.

Calculations were carried out for several values of \(t_1\) and several values of \(\varepsilon\) and the results are shown in table 6.3 (the \(\Delta N_{\text{BV}}\) values for the off-axis collisions are also plotted, with error bars, in figure 6.13). As with the \(^{16}\text{O}\) collisions we find that the results have usually converged but that the errors are far larger than in the earlier calculations (but, it should be noted, are still less than 1%). The individual results obtained in these calculations were far more consistent with results which have converged (albeit with larger errors than previously) than with the unconverged results seen in the case of the \(^{132}\text{Sn}\) GDR. These errors are not large enough to justify the differences between the different values of \(\Delta N_{\text{BV}}\). In the head-on collision the mass of the nucleus and the values of \(\Delta N_{\text{TDHF}}\) remained approximately constant however the values of \(\Delta N_{\text{BV}}\) are clearly not consistent, the result for \(t_1 = 900\) fm/c in particular being very different from the other two results (this calculation was repeated but this did not alter the results). The values of \(\Delta N_{\text{TDHF}}\) are far more consistent with the previous values of Marston and Koonin than they were for the \(^{16}\text{O}\) collisions. In the off-axis collision it is clear that the decay of the excited composite nucleus...
Figure 6.12: Density contour plots showing the density projected onto the reaction (x-z) plane for a collision between two $^{40}\text{Ca}$ nuclei at $E_{LAB} = 278$ MeV and for an impact parameter of 2.6 fm. The arrows on the first figure show the directions of the boosts applied to the two nuclei.
takes place over an extended period of time although the percentage increase between $\Delta N_{TDHF}$ and $\Delta N_{BV}$ is surprisingly consistent when compared with the results from the fusion-evaporation reactions for $^{16}$O+$^{16}$O. We feel that it would be inappropriate to attach any particular significance to this without conducting many more calculations. It is known that the neglect of centre-of-mass corrections causes more problems (in terms of the conservation of energy and momentum) in prolonged TDHF calculations for light nuclei [82] but this is not enough to allow us to expect significantly more consistent results for the $^{40}$Ca+$^{40}$Ca reactions than we did for the $^{16}$O+$^{16}$O fusion-evaporation reactions. Since this reaction resulted in the formation of a composite nucleus these results cannot be directly compared with those of Marston and Koonin.

We feel that whilst our results show that the values of $\Delta N_{BV}$ are consistently larger than the associated TDHF values the increases we have seen are not consistent with the increases reported by Marston and Koonin (table 6.3) and we do not regard it as likely that additional calculations would reproduce the experimentally measured mass fluctuation, $\Delta N_{EXP} \approx 11$ [51].
Chapter 7

Conclusions and Future Work

The BV variational approach for calculating the fluctuations of single-particle operators has been implemented using a fully three-dimensional TDHF code with the full Skyrme interaction and applied to resonances, deep-inelastic collisions and fusion-evaporation reactions for a variety of systems.

The Balian-Vénéroni approach has been applied to GDR's in $^{32}$S and $^{132}$Sn and a GMR in $^{40}$Ca. It has been shown that the previous calculations for the $^{40}$Ca GMR were severely influenced by the numerical errors inherent in the TDHF calculations. We have shown that once these errors are eliminated the Balian-Vénéroni approach produces converged and consistent results. The fluctuations thus calculated are consistently larger than the equivalent TDHF values.

We have applied the Balian-Vénéroni method to $^{16}$O+$^{16}$O ($E_{LAB} = 160$ MeV) and $^{40}$Ca+$^{40}$Ca ($E_{LAB} = 278$ MeV) heavy-ion collisions to obtain results which can be compared with previous calculations and with experimental data and have found that we obtain results which are not consistent with the previous calculations or with the experimental data (for the $^{40}$Ca+$^{40}$Ca reaction). In part this is a consequence of our off-axis collisions resulting in fusion-evaporation reactions which cannot be directly compared with the previous calculations. This is thought to be due to our use of a three-dimensional code combined with more modern interactions. We encounter difficulties in obtaining both converged and consistent results in our deep-inelastic collisions. For the fusion-evaporation reactions the long decay times meant that, whilst we were able to obtain some converged results, these were not reliable. Whilst the values of $\Delta N_{BV}$ obtained were consistently larger than the TDHF values the increased values were still not consistent with the experimental values.

It has been determined that the fluctuations calculated using the Balian-Vénéroni approach are far more susceptible to numerical errors than those calculated using the usual TDHF approach. If we wish to be able to obtain converged and consistent results it is important to check that the Balian-Vénéroni approach is implemented for a system which is known to have fully decayed (which may
be indicated by consistent values for the expectation values and fluctuations calculated using the usual TDHF approach). This helps to ensure that, when repeated calculations are performed, any differences between the results are less attributable to the evolving state of the system allowing a more confident assessment of the associated errors. Prolonged TDHF calculations are inherently more unstable than shorter calculations (due to the omission of centre-of-mass corrections and, in deep-inelastic reactions, the coupling between the spatially separate fragments) and suffer from the dissipated nucleons being reflected around the spatial box causing spurious interactions with the nuclei.

The Balian-Vénérond approach as implemented may be used to calculate fluctuations for resonances decaying by particle emission and, with great care, some deep-inelastic scattering reactions. This model would benefit greatly from the addition of reversible boundary conditions since simply increasing the size of the spatial box to assess and/or avoid the problem of the reflected nucleons is not a very practical solution. Whilst the current work has focused on mass distributions (for which data is only available for some heavy-ion collisions) a better comparison with experimental data may be obtained by looking for a different observable where we could compare with data from giant resonances.
Appendix A

The HF Equations

In this section we follow the derivation of the HF equations from [78, Chap. 5]. The HF approach is a self-consistent mean-field approach in which the true many-body wavefunction, \(|\Phi\rangle\), of an \(A\) nucleon system is written as a Slater determinant (an anti-symmetrised product of \(A\) single-particle states), \(|\Phi_{HF}\rangle\) (1.2). In the notation of second quantisation this can be written as

\[
|\Phi(1, \ldots, A)\rangle \rightarrow |\Phi_{HF}(1, \ldots, A)\rangle = \prod_{i=1}^{A} \hat{\alpha}_{i}^{\dagger} |0\rangle,
\]

where \(|0\rangle\) is a vacuum state and \(\hat{\alpha}_{i}^{\dagger}\)'s the creation operator which creates a particle in the single-particle state \(|\phi_{i}\rangle\).

It is assumed that each nucleon moves independently in a potential generated by the interactions between the nucleons as described by a nucleon-nucleon interaction. The Hamiltonian for the system, \(\hat{H}\) is written as the sum of \(A\) single-particle Hamiltonians, \(\hat{h}_{i}\)

\[
\hat{H} = \sum_{i=1}^{A} \hat{h}_{i},
\]

whose lowest energy eigenfunctions are the single-particle states, \(|\phi_{i}\rangle\)

\[
\hat{h}_{i} |\phi_{i}\rangle = \epsilon_{i} |\phi_{i}\rangle
\]

and where \(\epsilon_{i}\) are the associated single-particle energies. In a HF calculation the Slater determinant describing the system is determined by finding, using a variational approach, the set of single-particle states which minimises the energy in the system, given by \(\langle \Phi_{HF} | \hat{H} | \Phi_{HF} \rangle\).

The single-particle states \(|\phi_{i}\rangle\) are generally not known so we start from a set of trial single-particle states, best guesses, \(|\chi_{i}\rangle\), which themselves form a complete orthonormal set. These are related to
the states $|\phi_i\rangle$ by the (unknown) unitary transformation matrix, $D$ (satisfying $D^\dagger D = DD^\dagger = I$).

$$|\phi_i\rangle = \sum_{j=1}^{\infty} D_{ji} |\chi_j\rangle, \quad |\chi_j\rangle = \sum_{i=1}^{\infty} D^\dagger_{ji} |\phi_i\rangle. \quad (A.4)$$

We can define creation and annihilation operators, $\hat{c}^+$ and $\hat{c}$, in the trial basis which are related to the $\hat{a}^+$ and $\hat{a}$ creation and annihilation operators through the same transformation matrix

$$\hat{a}_k^+ = \sum_{i=1}^{\infty} D_{ik} \hat{c}_i^+, \quad \hat{c}_i^+ = \sum_{k=1}^{\infty} D^\dagger_{ik} \hat{a}_k^+, \quad (A.5)$$

Instead of working directly with the single-particle states it is useful to work in terms of the one-body density matrix, $\rho$, defined, in the trial basis, as

$$\rho_{uv} = \langle \Phi | \hat{c}_u^+ \hat{c}_v | \Phi \rangle. \quad (A.6)$$

Using (A.5) and replacing $|\Phi\rangle$ by $|\Phi_{HF}\rangle$ we obtain

$$\rho_{uv} = \sum_{k, k'=1}^{\infty} D_{ik} D_{i'k'} \langle \Phi_{HF} | \hat{a}_k^+ \hat{a}_{k'} | \Phi_{HF} \rangle. \quad (A.7)$$

The orthonormality of the single-particle states eliminates those terms with $k' \neq k$ whilst the remaining creation and annihilation operators reduce to the number operator, $\hat{n}_k = \hat{a}_k^+ \hat{a}_k$, which returns unity for an occupied state and zero otherwise. Only occupied (particle) states contribute to this summation leaving

$$\rho_{uv} = \sum_{i=1}^A D_{ii} D_{i'j'}, \quad (A.8)$$

which is not automatically a unit matrix due to the restricted summation over $i$. Evaluated in the HF basis (the basis of the $\hat{a}_k^+$, $\hat{a}_i$ creation and annihilation operators) $\rho$ is diagonal with eigenvalues that are either zero or one such that, in this basis, it satisfies $\rho^2 = \rho$ and is a projector projecting out the occupied (hole) states. If $\rho$ satisfies $\rho^2 = \rho$ in the HF basis then this must also be satisfied in any other arbitrary basis.

### A.1 The Hartree-Fock ground state energy

The HF ground state energy, $E_{HF}^0$, is given by

$$E_{HF}^0 = \langle \Phi_{HF} | \hat{H} | \Phi_{HF} \rangle, \quad (A.9)$$
where \( \hat{H} \) is the full many-body Hamiltonian including one- and two-body interactions defined, in an arbitrary single-particle basis (chosen to be the trial basis), to be of the form

\[
\hat{H} = \sum_{ab} t_{ab} c_{a}^{\dagger} c_{b} + \frac{1}{4} \sum_{abcd} v_{abcd} c_{a}^{\dagger} c_{b}^{\dagger} c_{d} c_{c},
\]

where \( v_{abcd} = v_{abcd} - v_{abdc} \).

Substituting in using (A.5) and (A.8) we obtain, for the one-body term

\[
\sum_{ab} t_{ab} \langle \Phi_{HF} | c_{a}^{\dagger} c_{b} | \Phi_{HF} \rangle = \sum_{ab} t_{ab} \rho_{ba},
\]

(A.11)

Repeating for the second term we obtain

\[
\frac{1}{4} \sum_{abcd} v_{abcd} \langle \Phi_{HF} | c_{a}^{\dagger} c_{b}^{\dagger} c_{d} c_{c} | \Phi_{HF} \rangle = \frac{1}{4} \sum_{abcdmnop} v_{abcd} D_{ma}^{*} D_{nb} D_{oa} D_{pc} \langle \Phi_{HF} | \delta_{mn}^{+} \delta_{np}^{+} \delta_{oa} \delta_{pb} | \Phi_{HF} \rangle.
\]

(A.12)

We see by inspection that \( o, p \leq A \) since \( a_{o} |\Phi_{HF}\rangle = 0 \) for \( o > A \) (we can only depopulate an occupied state). Similarly, \( m, n \leq A \) due to the orthonormality of \( |\phi\rangle \) which means that we must populate and depopulate the same two states in \( |\Phi_{HF}\rangle \). We can also say that \( o \neq p \) and \( m \neq n \) since we can’t populate or depopulate the same state twice. Using the anti-commutation relations for the fermion creation and annihilation operators, \( \{ \delta_{m}^{+}, \delta_{n}^{+} \} = \delta_{mn} \), and \( \delta_{mn} |\Phi_{HF}\rangle = 0 \) for \( m \leq A \), we get

\[
\langle \Phi_{HF} | \delta_{mn}^{+} \delta_{np}^{+} \delta_{oa} \delta_{pb} | \Phi_{HF} \rangle = \delta_{no} \delta_{mp} - \delta_{mo} \delta_{np}
\]

(A.13)

so that the two-body term now reads

\[
\frac{1}{4} \sum_{abcdmnop} v_{abcd} \langle \Phi_{HF} | \delta_{mn}^{+} \delta_{np}^{+} \delta_{oa} \delta_{pb} | \Phi_{HF} \rangle.
\]

(A.14)

which, using \( \bar{v}_{abcd} = -v_{abcd} \), reduces to

\[
2\frac{1}{4} \sum_{abcdmnop} v_{abcd} \delta_{mn}^{+} \delta_{np}^{+} \delta_{oa} \delta_{pb} = \frac{1}{2} \sum_{abcd} \rho_{cb} \bar{v}_{abcd} \rho_{db}.
\]

(A.15)

Combining these two terms we obtain the Hartree-Fock ground state energy

\[
E_{HF}^{0} (\rho) = \sum_{ab} t_{ab} \rho_{ba} + \frac{1}{2} \sum_{abcd} \rho_{ca} \bar{v}_{abcd} \rho_{db}.
\]

(A.16)

**A.2 The Variation of the Energy**

The HF-basis is determined by finding the trial state (a Slater determinant) which minimises the energy in the system. We investigate the effect on the ground state energy of the system of making small changes to the one-body density matrix, \( \rho \rightarrow \rho + \delta \rho \) (equivalent to populating different
single-particle states), whilst requiring that, at all times, \( \rho \) satisfies \( \rho^2 = \rho \)

\[
\rho + \delta \rho = (\rho + \delta \rho)^2
\]
\[
= \rho^2 + \delta \rho \rho + \delta \rho^2 + \rho \delta \rho
\]
\[
(A.17)
\]

The last term can be ignored for small \( \delta \rho \). Using this and the requirement that \( \rho^2 = \rho \)

\[
\delta \rho = \rho \delta \rho + \delta \rho \rho
\]
\[
(\rho \delta \rho) = \rho^2 \delta \rho \rho + \rho \delta \rho \rho^2
\]
\[
0 = \rho \delta \rho \rho.
\]
\[
(A.19)
\]

\( \rho \) is diagonal in the HF basis so to satisfy \( \rho \delta \rho \rho = 0 \) we require that \( \delta \rho \) only has non-zero elements for particle-hole and hole-particle states (off-diagonal elements).

The HF energy, (A.16), is also given by the standard equation, \( \langle \hat{H}_{HF} \rangle = \text{Tr} [H_{HF} \rho] \) where \( H_{HF} \) is the one-body Hamiltonian and \( \rho \) is the one-body density matrix. A change in \( \rho, \delta \rho \), produces a change in \( E^0_{HF} \), \( \delta E^0_{HF} \)

\[
\delta E^0_{HF} (\rho) = E^0_{HF} (\rho + \delta \rho) - E^0_{HF} (\rho) = \text{Tr} [H_{HF} \delta \rho] = \sum_{kk'} (H_{HF})_{kk'} \delta \rho_{kk'} = 0.
\]
\[
(A.20)
\]

The matrix elements \( (H_{HF})_{kk'} \) of the Hermitian matrix \( H_{HF} \) relate the change in the HF ground state energy to a change in one element of the one-body density matrix. Thus, if we consider making a small change to just one element of \( \rho \) then all except one of the terms in the summation disappears and it is easy to see that

\[
(H_{HF})_{kk'} = \frac{\delta E^0_{HF} (\rho)}{\delta \rho_{kk'}}
\]
\[
(A.21)
\]

where we now take \( E^0_{HF} \) from (A.16). We consider the one-body interaction term first

\[
\sum_{ab} t_{ab} \frac{\delta \rho_{ba}}{\delta \rho_{kk'}} = \sum_{ab} t_{ab} \delta \rho_{kk'} = t_{kk'},
\]
\[
(A.22)
\]

and similarly

\[
\sum_{abcd} V_{abcd} \left[ \rho_{ca} \frac{\delta \rho_{db}}{\delta \rho_{kk'}} + \rho_{db} \frac{\delta \rho_{ca}}{\delta \rho_{kk'}} \right] = \sum_{ac} V_{acck'} \rho_{ca} + \sum_{bd} V_{kbb'} \rho_{db},
\]
\[
2 \sum_{ac} V_{kac} \rho_{ca}.
\]
\[
(A.23)
\]
Combining these gives

\[
(H_{HF})_{kk'} = t_{kk'} + \sum_{ac} \varphi_{baka c} \rho_{ca} \Gamma_{kk'}
\]

(A.24)

or \( H_{HF} = t + \Gamma \). This gives us the form of the one-body, density dependent, HF Hamiltonian (A.2)

\[
\hat{H}_{HF} = \sum_{kk'} (H_{HF})_{kk'} \hat{a}_k^{+} \hat{a}_{k'} = \sum_{kk'} (t + \Gamma)_{kk'} \hat{a}_k^{+} \hat{a}_{k'}
\]

(A.25)

We have already shown that \( \delta \rho \) only has non-zero hole-particle and particle-hole elements so to satisfy (A.20) we require the hole-particle and particle-hole elements of \( H_{HF} \) are zero. This requirement allows us to write

\[
[H_{HF}, \rho] = 0.
\]

(A.26)

\( H_{HF} \) and \( \rho \) commute so they can be diagonalised simultaneously. We have already stated that, in the HF basis, \( \rho \) is diagonal, but we now also require that \( H_{HF} \) is diagonal thus uniquely defining the HF basis as the one in which both \( H_{HF} \) and \( \rho \) are diagonal and where the hole-hole elements of \( H_{HF} \) are

\[
(H_{HF})_{kk'} = t_{kk'} + \sum_{i=1}^{A} \varphi_{ibk'i} = \epsilon_k \delta_{kk'}.
\]

(A.27)

In this basis the HF ground state energy becomes (A.16)

\[
\mathcal{E}_{HF}^{0}(\rho) = \sum_{a} \epsilon_a + \frac{1}{2} \sum_{ab} \varphi_{abab}
\]

(A.28)

or, equivalently, using (A.27)

\[
\mathcal{E}_{HF}^{0}(\rho) = \sum_{a} \epsilon_a - \frac{1}{2} \sum_{ab} \varphi_{abab}
\]

(A.29)

which relates the energies of the single-particle states to the energy in the system with a correction factor related to the strength of the two-body interaction.
Appendix B

The TDHF Equations

The time-dependent Hartree-Fock approach extends the HF approach to consider dynamical processes. We still assume that the wavefunction of the system can be approximated to a SD and now consider the time evolution of the single-particle wavefunctions or density matrix as described by the TDHF equations. The TDHF approach also ensures that the total energy and the number of particles in the system remain constant with time. In what follows we follow the derivation of the TDHF equations from [78].

To derive the TDHF equation describing the time evolution of the density matrix we start from the (full many-body) state of the system, \( \Phi(t) \), and consider the elements of the one-body density matrix according to (A.6)

\[
\rho_{kl}(t) = \langle \Phi(t) | \hat{c}_k^+ \hat{c}_l | \Phi(t) \rangle,
\]

(B.1)

The time dependence of \( \rho(t) \) is obtained by differentiating this equation with respect to time

\[
\dot{\rho}_{kl} = \langle \Phi(t) | \hat{c}_k^+ \hat{c}_l | \Phi(t) \rangle + \langle \Phi(t) | \hat{c}_l^+ \hat{c}_k | \Phi(t) \rangle.
\]

(B.2)

This can be re-written by substituting in using the Schrödinger equation \( \dot{\Phi}(t) = i\hbar \Phi(t) \)

\[
\dot{\rho}_{kl} = \langle \Phi(t) | \hat{H} \hat{c}_k^+ \hat{c}_l | \Phi(t) \rangle + \langle \Phi(t) | \hat{c}_l^+ \hat{c}_k \hat{H} | \Phi(t) \rangle,
\]

(B.3)

Substituting in using the standard form for a Hamiltonian with one- and two-body interactions (A.10) and using the anti-commutation relations for fermion creation and annihilation operators \( \{ \hat{c}_a, \hat{c}_b^+ \} = \delta_{ab} \) we obtain, for the one-body interaction term

\[
\langle \Phi(t) | \left[ \hat{c}_k^+ \hat{c}_l, \sum_{ab} t_{ab} \hat{c}_a^+ \hat{c}_b \right] | \Phi(t) \rangle = \sum_{a} (t_{ka} \rho_{kl} - \rho_{kl} t_{al}).
\]

(B.4)
whilst the two-body interaction yields

$$\langle \Phi(t) | \left[ \hat{c}^+_a \hat{c}_b, \sum_{abcd} \hat{\sigma}_{abcd} \hat{c}^+_d \hat{c}_d \right] | \Phi(t) \rangle = \frac{1}{2} \sum_{abc} \left( \sigma_{kabc} \rho_{bcla}^{(2)} - \rho_{kabc}^{(2)} \sigma_{bcla} \right), \tag{B.5}$$

where \( \rho_{abcd}^{(2)}(t) \) is the time-dependent two-body density matrix defined as

$$\rho_{abcd}^{(2)}(t) = \langle \Phi(t) | \hat{c}^+_a \hat{c}^+_b \hat{c}_d \hat{c}_c | \Phi(t) \rangle. \tag{B.6}$$

Combining these we get

$$i \hbar \dot{\rho}_{kl} = \sum_a (t_{ka} \rho_{al} - \rho_{ka} \Gamma_{al}) + \frac{1}{2} \sum_{abc} \left( \sigma_{kabc} \rho_{bcla}^{(2)} - \rho_{kabc}^{(2)} \sigma_{bcla} \right). \tag{B.7}$$

The two-body density matrix can be decomposed into a product of one-body density matrices and a correlations matrix \( g^{(2)} \) which incorporates all of the two-body correlations \[78\]

$$\rho_{abcd} = \rho_{ac} \rho_{bd} - \rho_{ad} \rho_{bc} + g_{abcd}. \tag{B.8}$$

We use this equation to substitute in for \( \rho^{(2)} \) and, after some rearrangement, obtain

$$i \hbar \dot{\rho}_{kl} = \sum_a (t_{ka} \rho_{al} - \rho_{ka} \Gamma_{al}) + \sum_a \left( \Gamma_{ka} \rho_{al} - \rho_{ka} \Gamma_{al} \right) + \frac{1}{2} \sum_{abc} \left( \sigma_{kabc} g_{bcla}^{(2)} - g_{kabc}^{(2)} \sigma_{bcla} \right), \tag{B.9}$$

where \( \Gamma \) was defined in (A.24). The TDHF approach assumes that the wavefunction of the system can be approximated to a Slater determinant at all times. This means that two-body correlations are ignored which enters this equation through the assumption that \( g^{(2)} = 0 \). This leads directly to the TDHF equation

$$i \hbar \dot{\phi} = \left[ \hat{H}_{HF}, \phi \right]_{kl}, \tag{B.10}$$

or, in its more common form

$$i \hbar \dot{\phi} = \left[ \hat{H}_{HF}, \phi \right]. \tag{B.11}$$

Equation (B.11) can also be derived \[114\] from

$$\delta \int dt \Phi^*(t) \left( i \hbar \frac{\partial}{\partial t} - \hat{H} \right) \Phi(t) = 0, \tag{B.12}$$

which states that the energy of the system should remain constant as the state of the system evolves in time.
Appendix C

Associated Publications

C.1 Papers

1. Mass dispersions from giant dipole resonances using the Balian-Vénéroni variational approach
   J. M. A. Broomfield and P. D. Stevenson

C.2 Conference Proceedings

1. Mass Distributions Beyond TDHF
   J. M. A. Broomfield and P. D. Stevenson
   (FUSION08: New Aspects of Heavy Ion Collisions Near the Coulomb Barrier,
   Chicago, USA, Sept 2008)

2. Mass Distributions in a Variational Model
   (XXXII Symposium on Nuclear Physics, Cocoyoc, Mexico, Jan 2009)
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