Measurement of prompt gamma-ray energy distribution and multiplicity of $^{235}\text{U}$ following thermal fission using STEFF

A thesis submitted to The University of Manchester for the degree of Doctor of Philosophy in the Faculty of Engineering and Physical Sciences

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Abstract

ABSTRACT OF THESIS submitted to The University of Manchester by Miss Elizabeth Sarah Murray for the Degree of Doctor of Philosophy and entitled: Measurement of prompt gamma-ray energy distribution and multiplicity of $^{235}\text{U}$ following thermal fission using STEFF.

Date of submission: 28/03/2015

More accurate knowledge of gamma-ray heating in nuclear reactors has been listed as a high priority request by the NEA [23]. In response to this the SpecTrometer for Exotic Fission Fragments (STEFF), a 2-velocity, 2-energy spectrometer assembled by the Manchester Fission Group has been used to take measurements of prompt gamma-rays from thermal fission of $^{235}\text{U}$. Through the procedures discussed, the average total gamma-ray energy and average multiplicity were determined to be $8.40 \pm 0.26$ MeV and $7.74 \pm 0.12$, respectively. The single energy spectrum for each individual detector has been determined as well as the total energy distribution.

A new parallel plate avalanche counter has been tested for its potential to improve the timing resolution of the current STEFF stop detector. The timing resolution of the new detector is found to be 337 ps, an improvement of 75% on the previous stop detector.

The pulse shapes created by a fission fragment in an ionisation chamber have been investigated. The relationship with fragment atomic number has been tested by employing the Lohengrin spectrometer to separate fragments by mass so that their individual pulse shapes can be studied.

Evaluation work has been performed at the NNL, Cumbria, under the supervision of Dr Robert Mills. Experimental data determined from the spontaneous fission of $^{252}\text{Cf}$ has been extracted and evaluation techniques performed upon it so that the data can be considered for inclusion within an evaluated nuclear database.
Declaration

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School of Physics and Astronomy (or the Vice-President) and the Dean of the Faculty of Engineering and Physical Sciences.
I would like to thank my supervisor Dr Gavin Smith, for allowing me to join the STEFF collaboration. I appreciate his time and patience which has made my PhD experience as interesting and as enjoyable as possible. I am also extremely thankful to my secondary supervisor, Dr Robert Mills for his guidance and for inviting me to spend time working at the NNL in Cumbria.

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Chapter 1

Introduction

A high priority request has been set by the Nuclear Energy Agency (NEA) calling for more accurate knowledge on prompt gamma-ray heating in a nuclear reactor [1]. This chapter describes the importance of such measurements in the context of nuclear reactor safety. An overview of previous studies in this subject area is given here, describing the techniques employed and highlighting the current discrepancies in the associated measurements. Following this, an overview of nuclear fission theory is discussed against a brief background of nuclear structure.

1.1 Heating in nuclear reactors

The output power production of nuclear reactors is determined by the rate of heat removal from the reactor core. Pressurised Water Reactors (PWRs) heat water to ~300°C in the primary circuit, which is transferred to a secondary ‘cooling’ circuit. One gram of $^{235}$U inside a PWR will produce approximately one megawatt-day of heat energy [2].

The range over which this heat is dissipated inside the reactor is key to predicting material modification and subsequent reactor fuel failure mechanisms. It is depen-
<table>
<thead>
<tr>
<th>Form (kinetic energy)</th>
<th>Energy (MeV)</th>
<th>Range in reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission fragments</td>
<td>168</td>
<td>&lt; 1 mm</td>
</tr>
<tr>
<td>Prompt gamma-rays</td>
<td>7</td>
<td>10-100 cm</td>
</tr>
<tr>
<td>Prompt neutrons</td>
<td>5</td>
<td>10-100 cm</td>
</tr>
<tr>
<td>Capture gamma-rays</td>
<td>7</td>
<td>10-100 cm</td>
</tr>
<tr>
<td>Decay of fission products:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta$ particles</td>
<td>8</td>
<td>~ 1 mm</td>
</tr>
<tr>
<td>Neutrinos</td>
<td>12</td>
<td>$\infty$</td>
</tr>
</tbody>
</table>

Table 1.1: Estimated distribution of energy following thermal fission of $^{235}\text{U}$ fission and the associated range of heat distribution, adapted from [2].

dependent on the amount of energy released in fission, the surrounding material absorption properties and the associated geometry. A breakdown of the energy released in fission and the associated range of this energy dispersion is given in Table 1.1. From these predictions it is estimated that 180 MeV is deposited in the fuel, 8 MeV in the reactor structure, 2 MeV in the moderator and 10 MeV of energy is removed completely from the reactor by neutrinos [2].

In designs for early reactors, heat released within the nuclear reactor was assumed to be contained within the reactor fuel. Safety margins and safety limits are therefore likely to have been overestimated. More accurate knowledge is now in demand, so that localised heating per megawatt and the precise distribution of heat in a reactor can be be predicted.

A nuclear renaissance is currently underway in the UK, with a new fleet of Generation-III nuclear reactors, currently being built. New types of reactors, Generation-IV are currently being designed with the intention of implementation by 2050. These reactors utilise new materials and methods for which safeguards are as yet undecided.
Reactor codes such as WIMS [3] and PANTHER [4] predict localised heating within the core. The input parameters of these reactor codes rely heavily on data published in nuclear databases such as JEFF [5], JENDL [6] and ENDF [7], and the output of these simulations predict how close to the safety limit the reactor is running. Engineers designing these forthcoming reactors will refer to these nuclear databases for accurate nuclear data.

It is currently estimated that gamma radiation accounts for ~10% of the total heat energy released in the core of a thermal or fast reactor. In a conventional thermal reactor, 40% of this energy comes from the prompt gamma-rays released by fission of $^{235}$U and $^{239}$Pu [8]. However, Prompt gamma-ray energy and multiplicity distributions, which have been accepted into nuclear datasets are taken from measurements performed in the early 1970’s [9, 10]. In these papers fission fragment excitation energy and spin distribution values vary by up to 15%.

The OECD Nuclear Energy Agency (NEA) have issued a high priority request that discrepancies on gamma heating in a reactor are required to be within 7.5% variation [8]. One of the aims of STEFF is to improve the discrepancy surrounding prompt gamma heating, thereby going someway towards meeting this demand.

### 1.2 Previous studies

Interest in prompt gamma-ray fission spectra has been revived in the last few years due to the NEA high priority request discussed in Section 1.1. An outline of different studies in this area is given here and the significant results for each previous measurement and those included in the ENDF/B-VII database, are compared in Table 1.2. This includes average gamma-ray multiplicity per fission, $\bar{v}_\gamma$, average energy per gamma-ray, $\epsilon_\gamma$, and total gamma-ray energy, $E_{\gamma,\text{total}}$. Multiplicity in this publication
Table 1.2: Comparison of previous measurements of average gamma-ray multiplicity, $\bar{\nu}_\gamma$, average energy per gamma-ray, $\epsilon_\gamma$, and total gamma-ray energy, $E_{\gamma,\text{total}}$, following thermal emission of $^{235}\text{U}$. All results are given per fission.

<table>
<thead>
<tr>
<th></th>
<th>$\bar{\nu}_\gamma$</th>
<th>$\epsilon_\gamma$</th>
<th>$E_{\gamma,\text{total}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verbinski [10]</td>
<td>6.70 ± 0.30</td>
<td>0.97 ± 0.05</td>
<td>6.51 ± 0.30</td>
</tr>
<tr>
<td>Pleasonton [9]</td>
<td>6.51 ± 0.30</td>
<td>0.99 ± 0.07</td>
<td>6.43 ± 0.30</td>
</tr>
<tr>
<td>Peelle [11]</td>
<td>7.45 ± 0.35</td>
<td>0.96</td>
<td>7.18 ± 0.26</td>
</tr>
<tr>
<td>Oberstedt [12]</td>
<td>8.19 ± 0.11</td>
<td>0.85 ± 0.02</td>
<td>6.92 ± 0.09</td>
</tr>
<tr>
<td>ENDF/B-VII [7]</td>
<td>6.86</td>
<td>0.96</td>
<td>6.58</td>
</tr>
<tr>
<td>STEFF (this work)</td>
<td>7.74 ± 0.12</td>
<td>1.08 ± 0.03</td>
<td>8.40 ± 0.26</td>
</tr>
</tbody>
</table>

Initial detailed studies into thermal fission of $^{235}\text{U}$ were performed in the 1970’s [10, 9, 11]. A NaI(Tl) detector is utilised in [9] at a distance of 89 cm from the target and a surface-barrier detector is employed to detect fission fragments. A ‘Maier-Leibnitz’ weighting method is used to deduce average multiplicity and energy of prompt gamma-rays from measured pulse heights. Peelle [11] employed three NaI(Tl) scintillators and determined a different multiplicity measurement depending on the scintillator threshold level. The average multiplicity measurement, varied from $\bar{\nu}_\gamma = 8.13 \pm 0.35$ for a 10 keV threshold and $\bar{\nu}_\gamma = 7.45 \pm 0.32$ for a 140 keV threshold. Since these experiments, new detectors, digital acquisition systems and signal processing techniques have been developed, allowing for more accurate measurements.

Oed et al [13] in 1983, combines time-of-flight (TOF) and ionisation chamber energy-velocity to measure the velocity and kinetic energy of fission fragments. A timing resolution of 100 ps and a mass resolution of $\delta m/m = 0.6\%$ was achieved using
this method. For this experiment a section of Cossi Fan Tutti, a 2E2v spectrometer, is used at the ILL. The layout of this device is similar to STEFF as it employs two axial ionisation chambers in correlation with fragment’s TOF on a 107 m long flight path. This spectrometer, however, does not have the benefit of digital data acquisition. Cosi Fan Tutti also had a disadvantage in that it provided a narrow a angle of acceptance so that neutron evaporation caused fission fragments to be undetected.

Since 2013 another research group, Oberstedt et al [12], have contributed towards accurate measurements of prompt gamma-ray emission. The multiplicity and gamma emission has been measured using the fission fragment spectrometer, VERDI along with a variety of lanthanide-halide scintillation detectors located at 30 cm from the target. A double Frisch-grid ionisation chamber has been used by the group to supply the trigger for these experiments.

1.3 Nuclear models

There are a number of theoretical models employed to explain the complex, many-body system of nuclear fission. There is currently no single quantum mechanical or classical approach which gives a realistic result of the fission process, due to the large number of degrees of freedom involved. An overview of these models is given here, with a more detailed description of these ideas found in [14] and [15].

Nuclear fission was discovered by Hahn and Strassmen in 1938 [16]. When bombarding $^{235}$U with neutrons, instead of creating a heavier element the isotope $^{56}$Ba was produced. Meitner and Frisch coined the term ‘nuclear fission’ in 1939 [17] and theorised that the nucleus is analogous to a liquid drop. This analogy was extended 1939 by Bohr and Wheeler [18] and forms the basis of many approaches to fission theory.

The Liquid Drop Model (LDM) is centered on the concept that the binding energy
Figure 1.1: The average binding energy per nucleon as a function of atomic mass, illustrating the effect of the first four terms in the Weizsäcker’s formula [21].

The first three terms are described as having a macroscopic effect on the nuclear binding energy. They represent the nuclear volume, surface area and the Coulombic repulsion, respectively. The volume term is proportional to the number of nucleons, $A$ within the spherical drop when the nuclear radius is represented as $R(A) = r_0 A^{1/3}$ where $r_0 = 1.25$ fm. This accounts for the short range strong attractive nuclear force exerted by a nucleon on its nearest neighbours.

As nucleons at the surface of the nucleus have fewer neighbours than those in the...
interior of the nucleus but still exert the same strong force, surface tension is created. This results in the nucleus becoming spherical to minimise the potential energy on the surface. The surface term is therefore proportional to $A^{2/3}$.

The Coulombic term, proportional to $\frac{Z}{A^{1/3}}$, accounts for the charge density within the volume of the nucleus which exists due to long ranging electrostatic repulsion between the protons. The constants, $k_i$ are determined by fitting the SEMF to experimental data.

The binding energy per nucleon, calculated from the SEMF is plotted as a function of mass number in Figure 1.1. This illustrates that the first three terms alone are not sufficient to reach the experimentally determined ‘net binding energy’ curve.

Two additional corrections which are considered to have microscopic effect on the binding energy are also required. The asymmetry term accounts for stability of nuclei for which $N = Z$ and also accounts for the Pauli exclusion principle in paired nucleons. The pairing term, $\delta$, is small enough (~ 1 MeV) not to be included in the diagram in Figure 1.1 but is positive if $Z$ and $N$ are both even, and negative if $Z$ and $N$ are both odd. For odd-even nuclei, $\delta = 0$.

Fission is the decay of a large, parent nucleus, $X$ into two medium-mass fragments, $Y$ and $Z$,

$$X \rightarrow Y + Z. \quad (1.2)$$

A nucleus with $A > 56$, is more stable existing as two fragments and so it becomes energetically favourable for it to fission. The Q-value is the amount of energy released per fission and is calculated from the difference in the binding energy, of the parent nucleus and the binding energy, of the fragments,

$$Q = (M_x - M_y - M_z)c^2. \quad (1.3)$$
The maximum binding energy per nucleon occurs when \( A = 56 \), above which it gradually decreases. For a large nucleus, \( X \) with \( A > 120 \), the binding energy per nucleon is less than it is for two smaller, separate fission fragments, \( Y \) and \( Z \). This makes fission a likely decay pathway for heavy nuclei in order to increase their binding energy.

1.3.1 Distortions of the nucleus

As the nuclear volume increases, protons move apart lowering the Coulombic repulsion force. This movement is opposed by an increase in the surface tension force which returns the nucleus to its spherical shape. The interaction of these two mechanisms form a fission barrier in the potential energy of the system.

The shape of the distorted nucleus can be described by a Legendre polynomial expansion with the nucleus being represented as an axially symmetric distorted sphere of radius, \( R(\theta) \),

\[
R(\theta) = \frac{R_0}{\lambda} \left[ 1 + \sum_{n=1}^{\infty} \alpha_n P_n(\cos \theta) \right]. \tag{1.4}
\]

where \( \theta \) is the angle of the radius vector, \( \alpha_2 \) is a measure of the quadrupole distortion, \( R_0 \) is the radius of the spherical sphere prior to deformation. The parameter \( \lambda \), changes with the nuclear shape to ensure the volume remains constant, ensuring the incompressibility of the nucleus.

For larger deformations a higher order of polynomial is necessary but for relatively small contortions, the expansion can be limited to the second order. With this limitation, the surface tension, \( E_s \) and Coulomb energy, \( E_c \) are represented by,

\[
E_s = E_s^0(1 + \frac{2}{5} \alpha_2^2), \tag{1.5}
\]
Figure 1.2: Schematic illustrating the shape of nuclear deformation at the saddle point for various values of the fissility parameter, $x$ [22].

and

$$E_c = E_c^0 (1 - \frac{1}{5} \alpha_2^2),$$

(1.6)

where $E_s^0$ and $E_C^0$ are the surface and Coulombic energies before distortion. The fissility parameter, $x$ is defined as the ratio of these energies [18],

$$x = \frac{E_C}{2E_s} \propto \frac{Z^2}{A}.$$

(1.7)

Cohen and Swiatecki [22] calculated the radius vector of the nucleus shown in Equation 1.4 up to multipole order, $N = 18$. They employed 18 degrees of freedom to map the symmetry of the nucleus at the saddle point, and calculated the associated fissility parameter at each point. An example of the deformation symmetry using two degrees of freedom is illustrated in Figure 1.2. Nuclear fission will occur, when
the Coulombic repulsive forces overcome the short range surface tension force, when

\[ x > 1. \]

Nuclei for which \( x < 1 \) are considered metastable, and are bound only with

respect to small distortions.

The static path to fission as a function of deformation of the nucleus according to

the liquid drop model, is shown as the dotted line in Figure 1.3. The potential energy,

\( V \) increases with deformation up to a maximum ‘saddle point’. Following the breaking

of the neck between the fragments, at the scission point, the potential drops sharply
due to the loss of attractive surface tension. Coulombic repulsion will then accelerate

the separated fission fragments in opposite directions.

The main failure of the liquid drop model is that it does not include nuclear shell

effects. The model adequately describes the collective motions of a nucleus. It does

not produce, however, certain characteristics of nuclear structure. It does not explain

why the nucleus is stable at ‘magic’ nucleon shell numbers (2, 8, 20, 28, 50, 82 or 126).

The exclusion of shell structure in this model, means that certain features of fission
cannot be explained: It fails to predict lifetimes of spontaneous fission isomers, the
decay of heavy nuclei with varying excitation energies and cannot explain why fission

mass yield are symmetric.

### 1.4 Independent particle models

Significant refinements were made to the liquid drop model through the inclusion

of the single particle model introduced by Mayer and Jensen in 1955 [21]. The model
describes the interaction of a nucleon within a nucleus as the interaction of a single

particle with an average, static potential created by the rest of the nucleons within

the nucleus.

The total energy of the nucleus, according to the single particle model is described
Figure 1.3: The potential energy, $V$ of a nucleus on its path to fission as a function of deformation [14]. $A$ and $B$ indicate the heights of the first and second barrier, and $\beta_1$ and $\beta_2$ mark the positions of the saddle points. Class I and II show the excitation states at each stage of deformation. The schematic of the shape of the fissioning nucleus is shown above the diagram.
by,

\[ U = \sum_\nu 2n_\nu \epsilon_\nu, \quad (1.8) \]

where \( \epsilon_\nu \) are the energies of the single proton or neutron levels within a shell potential and \( n_\nu \) are the occupation number of the energy levels. The nucleon moves within levels which are arranged into ordered shells.

The arrangement of the energy levels are determined from the potentials in which the single nucleons exist. The mean field potential, \( V(r) \), in which the nucleons exist is described by,

\[ \left[ -\frac{\hbar^2}{2m} + V(r) \right] \psi(r) = E\psi(r). \quad (1.9) \]

The most commonly referred to potential models are the square well, simple harmonic oscillator (SHO) and the Woods-Saxon potential which are compared in Figure 1.4. The effect that each of these have on nucleon energy levels is shown in Figure 1.5. The square-well potential used to model the way in which nucleons move within the nucleus is derived from the Fermi gas model. This is improved by altering it so that the movement resembles that of a simple harmonic oscillator where,

\[ V(r) = \frac{1}{2}m\omega^2r^2 \quad (1.10) \]

in which \( \omega \) is the angular frequency and \( r \) is the distance from the centre of the nucleus to each nucleon.

Using the SHO to describe the potential does not reproduce experimentally observed magic numbers that are a result of shell structure. Two terms, \( l^2 \) and \( l \cdot s \), are added by Mayer [24] and Haxel [25] to correct for the experimentally observed shell gaps within the mean field. These terms account for coupling that occurs between the
nucleon’s intrinsic spin, $s$ and angular momentum, $l$ which contribute towards the total angular momentum, $j$ by the relationship,

\[ j = l + s. \tag{1.11} \]

The Woods-Saxon potential [26] is implemented as a better choice than the SHO for the mean field as it more closely reproduces the density distribution of nucleons for a spherical nucleus,

\[ V(r) = -\frac{V_0}{1 + \exp[(r - R)/a]}. \tag{1.12} \]

The skin thickness, $a$ represents the distance at which the density of charge falls from ninety to ten per cent of the value at the centre. In this relationship $r$ is the distance of the nucleons from the centre of the potential and $R$ is the nuclear radius. A spin-
orbit coupling term, $l \cdot s$ must also be added to the Woods-Saxon potential to account for correct magic numbers and reproduce the correct magic numbers. The effect of this spin-orbit term on the nucleon energy level ordering is shown in Figure 1.5.

The independent particle model is successful in predicting ground state masses and spin near closed magic shells. It also accounts for isomeric states, present when nuclear levels containing differing spins are found close to one another [27]. In this case orbitals in the shell structure lie close in energy to an intruder orbital from a different harmonic oscillator shell.

For nuclei with nucleons existing in orbits between magic numbers, which is the case for elements in the lanthanide and actinide series of the periodic table, it does not provide a good description. In these regions, the nucleus in the ground state is deformed into a prolate shape so that the movement of the nucleon in this potential has an axial, rather than spherical, symmetry.

### 1.4.1 Nuclear deformation

As described in Section 1.4.3, the parent nucleus undergoes deformation before fissioning. The level structure introduced by the independent particle model was extended by Bohr and Nilsson [19] [29] to account for deformed nuclei. For a deformed nucleus, the single particle Nilsson Hamiltonian, $H_{sp}(\varepsilon)$, is used to describe the potential,

$$H_{sp}(\varepsilon) = H_0 + C l \cdot s + D l^2,$$

where,

$$H_0 = -\frac{\hbar^2}{2M} \nabla^2 + \frac{1}{2} M\omega_0^2(\varepsilon)(x^2 + y^2)(1 + \frac{1}{3} \varepsilon)^2 + z^2(1 - \frac{2}{3} \varepsilon)^2.$$

(1.13)
Figure 1.5: Effect of different nuclear potentials on the nucleon energy level ordering: The simple harmonic oscillator, the Woods-Saxon and the Woods-Saxon with the spin orbit coupling term [28].
The first term in Equation 1.14 accounts for the kinetic energy of the single nucleon. The second term is the harmonic oscillator for which $x, y$ and $z$ are coordinates relative to the intrinsic axes and the oscillator frequency $\omega_0$ is a function of the deformation parameter, $\varepsilon$. The nucleus, in this potential can rotate around any axis. Two spin-orbit coupling terms $Dl^2$ and $Cl \cdot s$ are required in Equation 1.13 where parameters $C$ and $D$ determine the magnitude of these terms at zero deformation. More detail on the derivation of this is given in [29].

For the spherical nuclei, the energy levels do not change with the orientation of the nucleus but this is not true for deformed nuclei. For a non-spherical nucleus the degeneracy of the shell model orbitals is lifted so that the energy levels are altered by its deformation.

In the Nilsson diagram shown in Figure 1.6 positive parity is displayed with solid lines and negative parity by dashed lines. It shows the single particle energy levels for odd protons. Protons are plotted separately to neutrons, as protons have the extra Coulomb repulsion to account for.

1.4.2 Strutinsky model

The Strutinsky model is a hybrid of the LDM the and independent particle model [31]. It utilises a deformed Woods-Saxon potential to calculate the total energy, $E$, with the addition of shell $\delta U$ and pairing $\delta P$ corrections. These corrections are treated as a small deviation from the uniform single-particle energy level ordering,

$$ E = E_{LDM} + \sum (\delta U + \delta P), $$

where $E_{LDM}$ is the energy calculated from the liquid drop model. The pairing term reduces the effect of the shell correction as both terms reach a maximum and minimum at the same point of nuclear deformation but when the shell correction is
Figure 1.6: Nilsson diagram for odd protons $82 \leq Z \leq 126$. Positive parity is displayed with solid lines and negative parity by dashed lines [30].
positive, the pairing correction is negative, and vice versa.

Single particle corrections have been improved by various other methods, such as folding in a Yukawa potential into an existing nuclear shape [32], which corrects for the finite range of the nuclear force. A technical review of this method, and proof that the Stutinsky method is justified by the Hartree-Fock procedure is given in [33].

1.4.3 Double-humped barrier

Super-imposing shell corrections calculated using the Strutinsky procedure onto the path to fission as a function of nuclear deformation according to the LDM, produces a ‘double humped’ fission barrier, illustrated in Figure 1.3. A summary of the important features in this figure is given here. A more detailed discussion of how these features are derived from quantum mechanical processes are provided in [15] and [34].

The shape of the first and second minima, $\beta_1$ and $\beta_2$ is dependent on the deformation of the nucleus in the ground state and near the saddle point, respectively. Calculations from the Strutinsky model show that the second minimum is higher in energy than the first by $\sim 2$ MeV [15]. While existing in these minima, nuclear deformation is symmetric. Excitation levels within these minima are classed as, Class I and II states and it is the lowest of these in each class, the ground state, from which spontaneous fission occurs. At the second saddle point, $B$ the deformation becomes asymmetrical for high masses including, $^{235}$U.

The height of the first fission barrier, $A$ is determined from the Coulomb potential described in section 1.5.1 whereas the height of $B$ is entirely determined from shell corrections.

A nucleus existing in the second well is classed as meta-stable as there is a potential barrier on either side. From this position the nucleus can decay to its ground
state through gamma decay or via nuclear fission [35].

1.5 Fission of $^{235}\text{U}$

$^{235}\text{U}$ is a naturally occurring alpha-unstable isotope in the actinide series of the periodic table with a half life of 703 million years. When bombarded with thermal (0.025 eV) or fast neutrons $^{235}\text{U}$ nuclei absorbs a neutron, forming fissile $^{236}\text{U}$ in an excited state.

The excitation energy of the $^{236}\text{U}$ is given by the Q-value,

$$Q = [m_n + m(^{235}\text{U})]c^2 - [m(^{236}\text{U})]c^2 = 6.6\text{ MeV}$$  \hspace{1cm} (1.16)$$

This excitation energy is responsible for the high fission probability of $^{236}\text{U}$ and is high enough for the nucleus to overcome the fission activation barrier in the reaction,

$$^{235}\text{U} + n \rightarrow ^{236}\text{U}^* \rightarrow X + Y + \nu n$$  \hspace{1cm} (1.17)$$

where $\nu$ is the number of neutrons emitted per fission.

The ‘primary’ fission process refers to that which occurs within a few ns of the
scission point. This includes the initial decay into two products, accompanied by the release of prompt neutrons and prompt gamma-rays as the fragments cool from their initial excited state to a more stable ground state. A timeline of the processes that occur in fission is shown in Figure 1.7.

The ‘secondary’ process, refers to the period of time following fission and following prompt neutron evaporation, where the fragments are still neutron-rich and unstable. The fragments move towards stability via beta decay, which is also accompanied by the release of gamma radiation. The primary process may be further delayed by states with longer lived isomeric transitions.

1.5.1 Fission fragment kinetic energies

A simplified way to calculate the kinetic energy of the fission fragments is to model them as two spheres, initially in contact, being forced away from each other by Coulomb repulsion. The Coulombic potential energy is given by,

\[ V_C = \frac{Z_1 Z_2 e^2}{R_1 + R_2}. \]  

(1.18)

for spheres with radius, \( R_1 \) and \( R_2 \) and proton number \( Z_2 \) and \( Z_2 \). It can then be assumed, provided fission occurs symmetrically, that all of this Coulombic energy is transferred to the kinetic energy of the fission fragments i.e. not transferred to excitation of rotational or vibrational degrees of freedom.

By substituting the radius to be a function of mass number \( R = r_0 A^{1/3} \), the most probable kinetic energy of the fission fragment is found to be linearly dependent on \( \frac{Z^2}{A^{1/3}} \) [36],

\[ \langle E_k \rangle = 0.1071 \frac{Z^2}{A^{1/3}} + 22.2 \quad MeV. \]  

(1.19)
The constants are determined experimentally and vary with the excitation energy of
the fissioning nucleus as described in [37].

The amount of energy released in the thermal fission of a $^{235}\text{U}$ nucleus is approx-
imately 200 MeV of which 168 MeV is removed as fission fragment kinetic energy. A
break down of the approximate dispersion of the remaining released energy is given
in Table 1.1.

1.5.2 Mass of fragments

For thermal fission of $^{235}\text{U}$, the fission products consist of a light fragment with
average mass, $\langle A_l \rangle = 94$ and a heavy fragment, $\langle A_h \rangle = 139$. In contrast to that
predicted by the LDM, the fragment mass ratio is asymmetric with the heavy to light
ratio being $1.3 - 1.5$ [38].

For fission of various parent nuclei, the heavy fragment peak position remains
fixed and the change in fissioning mass is offset by shifts in the light fragment mass.
The lower boundary in the heavy peak is fixed at $A = 132$ as the heavy fragment is
doubly magic, and therefore highly stable with, $Z = 50$ and $N = 82$.

At higher excitation energies of the parent nucleus, nuclei are much more likely to
fission symmetrically [39]. Measuring independent and cumulative mass yields are
discussed in further detail in Section 6.2.

1.5.3 Charge of fragments

The charge distribution of the primary fission products follows the shape of the
mass distribution in that it is asymmetric and has a similar peak to valley ratio [15].
Conservation of charge requires that the charge of the parent nucleus $Z_{CN}$ is the
equivalent to the combination of the heavy fragment, $Z_h$ and light fragment, $Z_l$ [40].

The initial velocity of the recoiling fragments is high enough so that the outer elec-
trons from the parent nucleus are stripped away leaving the fission fragments in a
Figure 1.8: Independent fractional charge yield following a Gaussian distribution of fragments with $A = 91$. Adapted from [41].

For each fragment mass, $A$, produced from fission, the associated fractional independent yield $P(Z)$, of the fission product with atomic number $Z$ is assumed according to [41] to follow a Gaussian distribution determined by the relationship,

$$P(Z) = (c\pi)^{-1/2}\exp\left(\frac{(Z - Z_P)^2}{c}\right)$$

(1.20)

where $Z_P$ is the maximum atomic number value in the charge distribution curve and the parameter $c = 0.9$ is set by fitting this distribution to known experimental data. The independent fractional charge distribution for $A = 91$ is shown in Figure 1.8. The
proton to neutron ratio of each fragment also mirror that of the parent nucleus, the neutron to proton ratio of $^{236}\text{U}$ being 1.565 [38], making the fragments neutron rich.

1.5.4 Neutron emission

Neutron emission occurs at several stages during the thermal fission process and removes a large amount of excitation energy (~ 2 MeV) from the fission fragments. Some are emitted at the moment of scission as indicated in Figure 1.7, but the majority are evaporated from the accelerating fragments [15]. These prompt neutrons, of which 2.5 are emitted on average for $^{235}\text{U}$ there are within $10^{-9}$ ns as shown in Figure 1.7. The neutron energy distribution is discussed in detail in Section 3.2.1.

1.5.5 gamma-ray emission and angular momentum

Following scission, the fragments carry rotation, vibration and internal energy [42]. Immediately after neutron evaporation, fragments carry an average excitation value of 4 MeV and possess angular momentum ($6\hbar$) [43]. As the fragment cools, its angular momentum is reduced and excitation energy lowered by emission of prompt gamma-rays. gamma-rays that remove orbital angular moment are classed by multiple order as a function of quantum number, $L$ of which the majority are monopole, $L = 0$, dipole $L = 1$, or quadrupole $L = 2$.

Initially, statistical gamma-rays are emitted, within ~ 1 ps of scission, as a result of electric dipole ($E1$) transitions. The energy distribution of statistical gamma-rays is continuous but the quantity of statistical gamma-rays emitted per fission vary. A significant amount of energy is removed by these transitions (> 1 MeV) but they have a limited effect on reducing the fragments angular momentum as illustrated in the schematic in Figure 1.9. The result of statistical gamma-ray emission is that the fragment is shifted to the more structured yrast region which has a lower energy level density.
Figure 1.9: Schematic illustrating the decay of a primary fission fragment following prompt neutron evaporation by emission of statistical and yrast gamma-rays [44].
When the fragment has reached these lower excitation levels (2-2.5 MeV, spin $5\hbar$), discrete yrast gamma-rays due to quadrupole transitions ($E2$) are emitted [43]. Yrast decays do not remove as much energy as statistical decays but reduce the fragment’s angular momentum considerably.

A statistical model predicting the probability $P(J_i)$ of the fission fragment’s initial spin state, $J_i$, is described in detail in [45] and can be summarised by the expression,

$$P(J_i) \propto (2J + 1)\exp \left\{ -\frac{(J_i + 1/2)^2}{B^2} \right\},$$

(1.21)

where $B$ is proportional to the root mean square of the fission fragments angular momentum, $J_{rms}$. This model is utilised to calculate the gamma-ray multiplicity measured by STEFF, as described in more detail in Section 3.6.
Chapter 2

Experimental Setup

In June 2012, STEFF (SpecTrometer for Exotic Fission Fragments) ran for $2 \times 25$ day cycles in the PF1B pit at the Institut Laue-Langevin (ILL), Grenoble. A thermal neutron beam with a neutron flux density of $1.8 \times 10^{10}$ neutrons cm$^{-2}$s$^{-1}$ was used to induce fission on a $^{235}$U target. An overview of the experimental set-up is given here, followed by a detailed description of the different components that constitute STEFF and the corresponding experimental settings. Potential improvements to the stop detector design, by replacing it with a PPAC (Parallel Plate Avalanche Counter), have been investigated and are discussed in Section 2.9.

2.1 STEFF overview

STEFF is a 2-energy 2-velocity ($2E2v$) spectrometer designed and assembled by the ‘Fission Group’ collaboration at the University of Manchester. A central chamber containing a $^{235}$U target is surrounded by eleven sodium iodide detectors. Two fragment detection arms are positioned on either side of the central chamber so that the time-of-flight (TOF) of each fission fragment can be measured. A Bragg ionisation chamber is coupled to the end of both arms to measure the energy of each fragment. A
Figure 2.1: A cross sectional view showing the separate components of STEFF.
Figure 2.2: Photograph of STEFF in situ at the ILL, taken before installation of the NaI(Tl) array.

cross sectional view of STEFF, showing its separate components is given in Figure 2.1 and a photograph of the spectrometer in situ at PF1B is shown in Figure 2.2. When the velocity, \( v \) and the energy, \( E \) of the fragment are combined the fragment's mass, \( m \) is determined by,

\[
m = \frac{2E}{v^2}.
\] (2.1)

The velocity, \( v \) is calculated by \( v = l/t \), where \( t \) and \( l \) are the time and the distance between the start and stop detectors, respectively. The mass resolution, \( \Delta A \), which determines the accuracy of the mass measurement, is dependent on \( \Delta t \) and \( \Delta E \) by the relationship,

\[
\frac{\Delta A}{A} = \left[ \left( \frac{\Delta E}{E} \right)^2 + \left( \frac{2\Delta t}{t} \right)^2 \right]^{1/2},
\] (2.2)

which shows that the fractional error of the timing signals is the more important
Figure 2.3: 2D matrix of pulse height collected in the ionisation chamber against the fragment TOF.

contribution to the mass resolution than the energy measurement. The design objective is to achieve a fragment mass resolution of approximately 2% to compete with other ‘fragment identification methods’ described in Section 3.2. Further details about the development of STEFF are given in [46] and [47].

Converting the energy pulse height collected in the ionisation chambers and the fragment TOF to calibrated energy and fragment mass measurements requires the use of a transformation matrix calculated by Schmitt et al [48]. In this work, a ‘universal’ energy calibration is described whereby the fragment energy is determined using the pulse-height, $h$ and the fragment TOF, $t$,

$$E = (a + a't)h + b + b't, \quad (2.3)$$

where $a$, $a'$, $b$ and $b'$ are constants dependent on the experimental setup employed. Schmitt assumes that there is no mass dependence in the timing calibration such
that,

\[ t = c_t + a_t h_t^2 - b_t h_t, \quad (2.4) \]

where \( h_t \) is the timing pulse amplitude and \( a_t, b_t \) and \( c_t \) are calibration constants. A 2D matrix showing the pulse height collected in the ionisation chambers (IC) against the fragment TOF as measured by STEFF is shown in Figure 2.3. A routine has been implemented which makes a comparison with the transformed matrix and that measured by Schmitt [48] (called the ‘Target Matrix’). By optimising the fit between the measurements recorded by STEFF to those published in [48] for \(^{235}\text{U}\) thermal-neutron-induced fission, a calibrated energy and mass spectrum is produced. The result of this optimisation is the energy and mass projections shown in Figure 2.4 against the target matrix for reference.

Since the data from [48] have only mean kinetic energies quoted for each mass, the width of the kinetic energy distribution has been broadened with \( \sigma = 6.5 \text{ MeV} \), assuming a Gaussian distribution for each mass. The \( \chi^2 \) value associated with the fit is determined by comparison of the transformed data matrix and the target matrix, rather than just the energy and mass projections as was the case in [47]. The results show that although the mass peaks are in the correct place, the light fragment energy is higher than expected. In addition to this, the light and heavy peak intensities in the mass spectrum are not well reproduced which suggests that there are likely to be effects due to target thickness that have not been included in this approach. A \( \chi^2 \) value of 10.72 is associated with this fit.

### 2.2 Target

The 95% enriched \(^{235}\text{U}\) target shown in Figure 2.5 was prepared at the Bhabha
Figure 2.4: Projections of transformed matrix to Schmitt ‘target’ matrix [48] for fission fragment a) mass and b) energy.
Atomic Research Centre in Mumbai. The thickness of uranium is 100 mg cm\(^{-2}\), which for a neutron flux of \(10^5\) neutrons per second produces approximately \(10^4\) fissions per second per cm\(^{-2}\). It was manufactured on a 50 \(\mu g\) cm\(^{-2}\) nickel foil, supported by a thin copper plate. The copper was then etched away using sulphuric acid leaving behind the thin nickel backing, through which the fission fragments can pass with a minimal amount of energy loss. The target was placed in the target holder with the uranium facing the direction of Arm 1. The darker circle in Figure 2.5 shows that the thermal neutron beam incident on a paper target has a diameter of approximately 3 cm.

2.3 Start detector

The start signal is measured using a Secondary Electron Detector (SED) situated before the opening of Arm 2, 10 cm from the target inside the central chamber. A diagram of this is shown in Figure 2.6.

Fission fragments pass into the detector through the electrostatic mirror and exit
Figure 2.6: The SED start detector which relies upon MCPs to amplify the timing signal.
through the vertical grid. An indium coated foil, of 0.2 μm thickness, covers the vertical grid, providing an electrically conductive layer so that approximately nine secondary electrons are emitted each time a fission fragment passes through it [49]. A quarter of these electrons are back-scattered (travel in the opposite direction to the fission fragment), possessing several eV’s of kinetic energy. It is these which are measured, rather than electrons emitted in the same direction as the fragments, as the width of their energy distribution is smaller, therefore providing a more accurate timing signal.

The volume between the vertical grid and the upper microchannel plate is held at ground potential. The outer deflection grid is maintained at a high voltage (1.33 kV) so that when the backscattered electrons reach the gap between the inner and outer grids, they are deflected 90° downwards, in the direction of the upper multichannel plate. The electrical signals are then amplified using two Micro Channel Plates (MCPs) which sit below the electrostatic mirror. These plates amplify the number of electrons by a factor of ~ 10^5 by releasing more electrons with each interaction [49] [50]. For the start detector to work effectively, the pressure inside the central chamber must be kept at a high vacuum (at least below 2 × 10^{-5} mbar) to minimise noise on the timing signals.

An SED was chosen to detect the fragment’s start signals as it has several advantages over other timing detectors: A timing resolution of several hundred ps is possible and there is little distortion of the fragments’ trajectory due to the positioning of the MCPs below the fragment’s path. A limitation of this type of detector is the relatively small detection area. To account for this, the detector is placed close to the target to improve the acceptance angle for fission fragments.
2.4 Stop detector

Two SED stop detectors are positioned on either side of the central chamber. The detectors consist of an electrostatic mirror and a Multi Wire Proportional Counter (MWPC) as visible in the cross sectional diagram in Figure 2.7. A thin aluminised Mylar entrance window separates these components so that the electrostatic mirror can be maintained at a high vacuum, while the MWPC holds isobutane gas at a pressure of 10 mbar. Fragments enter into the electrostatic mirror through the aluminised thin foil releasing secondary electrons. These then deflected towards the MWPC by a strong electric field in a similar way as the start detector.

Inside the MWPC chamber, the cathode is divided into five copper segments of area 30 cm$^2$ as shown in Figure 2.8 with a high voltage supply to each strip via a resistor and decoupling capacitor. The anode is a mesh stretched over a copper frame, held at ground potential. A uniform electric field exists between the anode and cathodes so
that electrons produced from the ionisation of the gas drift towards the anode wires. In the near vicinity of the wire, electron avalanches occur. A discussion of electron avalanches is given in Section 2.9.2. Each timing signal is passed through an external fast preamplifier and then a Constant Fraction Discriminator (CFD), which picks off the timing signal, before being processed in a Timing to Amplitude Converter (TAC).

The stop detectors are considerably larger than the start detector in order to maintain the same 60 msr angle of detection. MWPCs have the advantage over other stop detectors in that the mirrors can be made to fit a wide area allowing for this large solid angle of acceptance [51]. Apart from the thin films, the bulk of the detector (MWPC) is positioned away from the path of the fragment, to reduce energy loss from straggling.

### 2.5 Sodium iodide detectors

An array of eleven Scionix NaI(Tl) detectors provide a large solid angle of acceptance for prompt gamma-rays. The size of the NaI(Tl) crystals is $5 \times 4$ inches and each are housed in 5 mm thick tungsten shielding to reduce the effects of Compton scattering between adjacent detectors. A timing and energy output signal is measured.
by each detector, processed through a built-in voltage divider and preamplifier before being fed into a TAC and an Analogue-to-Digital Converter (ADC), respectively.

The arrangement of detectors in STEFF is hemispherical, positioned at 60° to each other, with six detectors in the lower half at -45° to the horizontal axis, and five in the upper half at +45° to the horizontal axis. The interaction of gamma-rays with liquid scintillator detectors depends on the geometry of the detector array and the energy of the incident gamma-ray. A detailed discussion of gamma interaction with scintillator crystals are given in [52].

2.6 NE213 neutron detectors

Eight NE213 organic liquid scintillator counters are positioned around the central chamber to detect neutrons released by fission. The detectors consist of a photomultiplier tube coupled to a scintillation tube which are each supplied with ~1700 V. NE213’s are the preferred choice for prompt neutron detection from fission due to their ability to distinguish between prompt neutrons and gamma-rays when combined with offline pulse discrimination methods (PSD). Collection of the scintillation pulses include the time differences between prompt and delayed pulses which refer to the gamma-rays and neutrons, respectively. A full neutron energy spectrum can be drawn from the relative intensity of the detected pulses. Other advantages of using NE213s is that they have a high detection efficiency and resolution and that the scintillator has a fast decay time, avoiding signal pile up [53].

2.7 Bragg chambers

Two Bragg axial ionisation chambers are situated at the end of each arm of STEFF. The chambers in Arm 1 and Arm 2 are filled with isobutane gas and maintained at 70 and 105 mbar respectively. A cross sectional diagram of the inside of the Bragg
Figure 2.9: Cross sectional view of the Bragg chamber positioned at the end of Arm 2.

The entrance window to the chamber is made from aluminised, 1$\mu$m thick Mylar-film which is thin enough to allow the fragments to pass through, but allows the Bragg chamber and the stop chamber to be held at different pressures. The film must be as thin and evenly distributed over the window as possible, as the amount of energy the fragments lose as they pass through effects the energy resolution of the chamber. This caused many delays during the experiment as accidental rapid changes in pressure resulted in damage to the thin films. In these instances, the experiment was shut down, the chambers returned to atmospheric pressure and removed so that the films could be replaced.

The annular shaping rings visible in Figure 2.9 are held at 900 V and 1250 V inside
Arm 1 and Arm 2, respectively. These rings direct the fission fragments towards the anode by keeping the electric field parallel to the fragments trajectory and uniform throughout the chamber.

The Frisch grid is a set of parallel gold plated tungsten wires at 2 mm spacing situated at 15.2 cm from the entrance window and 1.2 cm in front of the anode. It’s function is to shield the anode from induced signals caused by the movement of ions towards the cathode. The anode should ideally only pick up signals, therefore when the electrons have passed through the Frisch grid. The distance from the entrance of the chamber to the Frisch grid is larger than the range of the most energetic fission fragments for that gas pressure, so that the pulse shape produced by the fragment is not cut off before it reaches a maximum.

The anode is separated into fifteen, $9.8 \times 4.8$ cm, segments. Each of these are held at high voltage (~1.25 kV) and are connected to individual preamplifiers through a 6800 pF capacitor. This set up allows sensitivity to the position of the fission fragments; the path of the fragment is longer if a fragment is traveling from the target towards an outer anode on the Bragg chamber than if it is traveling toward the central anode. Analysis of the pulse shapes collected from a Bragg chamber is described in Chapter 5.

2.8 Electronics and acquisition of data

The data acquisition system implemented in this experiment employs digital electronics collected by NIM and VME modules. An overview of the full electronic setup of STEFF is shown in Figure 2.10. The collected data are processed through the computer system MIDAS and stored on an external hard drive so that all noise filtering and pulse shape analysis is performed offline. The rate at which data are written to
Figure 2.10: Electronic component set-up of STEFF showing the layout of the signal processing modules which includes Constant Fraction Discriminators (CFDs), Gate and Delay Generators (GDGs), Multi Level Translators (MLUs), Analogue to Digital Converters, (ADCs), Time to Digital Converters, (TDCs), and gamma-ray Tracking 4 (GRT4) cards. Adapted from [47].
<table>
<thead>
<tr>
<th>Setting</th>
<th>Rate (per second)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_\gamma = 3$</td>
<td>460</td>
</tr>
<tr>
<td>$N_\gamma = 2$</td>
<td>4900</td>
</tr>
<tr>
<td>Start + ($N_\gamma = 2$)</td>
<td>29</td>
</tr>
<tr>
<td>Stop1 + ($N_\gamma = 2$)</td>
<td>28</td>
</tr>
<tr>
<td>Start +Stop1 + ($N_\gamma = 2$)</td>
<td>3.7</td>
</tr>
</tbody>
</table>

Table 2.1: Typical measurement rates recorded during the experiment. These rates varied throughout the experiment.

A trigger signal is produced when the output signals from the start and stop detectors occur in coincidence with a prompt gamma-ray signal. Three Ortec 556 Time-to-Amplitude Converters (TAC) receive a ‘Start & Stop1’, ‘Start & Stop2’ or ‘Stop 1 & Stop 2’ signal. This is combined with TAC signals from the NaI(Tl) detectors which are set to trigger on either $N_\gamma = 1, 2$ or $3$ gamma-rays of an energy determined by MIDAS. Typical measurement rates with various settings recorded during the experiment are displayed in Table 2.1. When a trigger signal is received, the GRT4 cards (Gamma-Ray Tracking 4) begin recording. There is then a window of 100 ns in which any timing signals received from the TAC cards are considered to be from the same fission fragment. As well as creating a trigger, the signals are also delayed and recorded to provide the timing measurement. A digitised pulse shape is collected by the 30 segmented anodes in the Bragg chambers (15 on each Bragg chamber). These are fed straight into the GRT4 card without alteration.
<table>
<thead>
<tr>
<th>Component</th>
<th>Pressure (mbar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bragg 1</td>
<td>80</td>
</tr>
<tr>
<td>Bragg 2</td>
<td>100</td>
</tr>
<tr>
<td>MWPC 1 and 2</td>
<td>10</td>
</tr>
<tr>
<td>Electrostatic mirror 1 and 2</td>
<td>$2 \times 10^{-5}$</td>
</tr>
<tr>
<td>Central chamber</td>
<td>$2 \times 10^{-5}$</td>
</tr>
</tbody>
</table>

Table 2.2: Typical pressure of each component of STEFF held throughout the experiment.

2.8.1 Gas flow

Before the Bragg chambers or the MWPC could be filled with isobutane gas, the whole of STEFF was evacuated for a minimum of 12 hours to remove any impurities and water vapour which could affect the measurements. This is achieved by a combination of turbo molecular pumps and rotary pumps. The rotary pump has the capability to reduce the pressure in the chamber to $\sim 10^{-2}$ mbar. Following this, to achieve the higher vacuum required in the central chamber and stop detectors, turbo pumps are utilised. Each element of STEFF is held at a different pressure throughout the experiment with the average pressure readings for each shown in Table 2.2. A combination of Penning and Pirani gauges were used to monitor the pressure.

Isobutane gas was circulated continually through each of the Bragg chambers and the MWPC detectors while STEFF was taking measurements to replace any radiation damaged gas inside the chambers. The gas flow rate into the Bragg and the MWPC chamber must be kept lower than 1 mbar per second to avoid damage to the thin foils inside the chambers.
2.9 PPAC development work

As discussed in Section 2.4, the timing resolution of STEFF has the potential to be improved by exchanging the current MWPC stop detector to a Parallel Plate Avalanche Counter (PPAC). The timing resolution of the MCP start detector is 100 ps compared to 1300 ps for the stop detector. Therefore it is the MWPC stop detector that is limiting the overall timing performance of STEFF. Operating conditions of the new PPAC have been tested to optimise its timing ability with the overall goal of improving the signal rise time from its current value of ~ 9 ns.

2.9.1 PPAC design

A photograph of the chamber used in this investigation is shown in Figure 2.11. The anode is a copper sheet consisting of five, 47 x 47 mm segments separated from the cathode by a 3 mm gap. The cathode is made of a stainless steel mesh, 0.46 mm thick screwed onto a PTFE plate. The cathode is grounded by a copper earthing strip. Air in the chamber is evacuated before isobutane is circulated through it for reasons described in Section 2.8.1. An $^{241}\text{Am}$ source, which decays through alpha emission to $^{237}\text{Np}$ was positioned on the floor of the chamber in the centre. The alpha particle causes primary ionisations of the gas molecules, liberating electrons. A high enough voltage is applied to the anode (~ 200 V), via a copper strip, so that the electric field is strong enough to cause secondary ionisations along the path of the alpha particle. These secondary electrons are accelerated by the strong electric field causing further collisions and ionisations resulting in an electron cascade known as a Townsend avalanche [54]. A Townsend avalanche amplifies the number of electrons by ~ $10^4$ before reaching the anode. The increase and can be quantified by the relationship,

$$N(r) = N_0 e^{\alpha r},$$

(2.5)
Figure 2.11: PPAC detector components (a) the side view from the outside (b) inside the chamber where the anode outputs are numbered from 1-5.
where $N(0)$ is the initial number of electrons and $N(r)$ is the number of secondary electrons at a distance along the electron path, $r$. The Townsend coefficient, $\alpha$ is the mean probability for ionisation per unit length. It is a function of the reduced electric field, $E(r)/P$, by the relationship,

$$
\alpha = A Pe^{-BP/E(r)}.
$$

where $P$ is the gas pressure, $E(r)$ is the electric field strength and $A$ and $B$ are experimentally determined constants dependent on the type of gas used, listed in [55]. The Townsend coefficient remains constant in a uniform electric field.

PPAC detectors are classed as proportional counters as the amount of energy lost by the alpha particle is proportional to the amount of ions that are produced [52]. Detailed discussions of PPAC detectors are given in Knoll [52], Kumagai [56] and Coffin and Engelstein [57]. The main advantages of a PPAC detector are its simple construction, good timing resolution ($\sim 100$ ps) and high counting rates ($10^4 s^{-1} mm^{-1}$). They also do not suffer radiation damage, unlike solid state detectors [58].

### 2.9.2 Anode signals

The signal must be collected quickly on the anode for accurate timing. This is...
quantified by the signal rise time which is defined as the length of time a signal takes
to rise from 10% to 90% of the total pulse height. It is dependent on the electron drift
velocity, \( v_e \) and the electron path length \( l \) such that,

\[
t_r = \frac{l}{v_e}
\]

where,

\[
\nu_e \propto \frac{E(r)}{P}.
\]  

(2.8)

The output signal on the anode has two components corresponding to the current
in the detector volume: one from electron and one from ion movement within the
chamber. Both of these currents induce a signal on the electrodes although, the signal
incurred by the electron motion rises ~ 1000 times faster than the signal induced by
the movement of ions. Using a fast preamplifier results in a sensitivity to the electron
motion only so that only the first peak is measured. The rejection of the second peak
is ensured by setting a short time constant, \( \tau \) in the corresponding charge-collection
circuit. This is given by,

\[
\tau = RC,
\]

(2.9)

where \( R \) and \( C \) are respectively, the resistance and capacitance of the anode circuit
including the input impedance of the preamplifier. There are also two inverted parallel
diodes between the preamplifier and the ground to prevent sparks in the detector
from damaging the preamplifier. A circuit diagram showing the electronic setup for
each anode is shown in Figure 2.11. The pulses are not combined until they have been
processed through the external preamplifier. This has the advantage of buffering the
impedance of each detector element so that the capacitance that determines the rise
Figure 2.13: The effect of the anode input voltage on the output signal rise time at various pressures. The trendline is determined at each pressure through quadratic regression analysis.

time is of one element and not the entire detector. The output signal on the anode is induced by the current in the detector volume as described in [59].

The results in Figure 2.13 confirm that the general trend is that the pulse rise time decreases as the voltage increases, as pressure decreases. The errors in this analysis are calculated from the average value of repeated readings and the trendline is fitted through quadratic regression analysis.

A similar test is done for the amplitude of the signal at different voltages as shown in Figure 2.15. As input voltage increases the amplitude of the pulse also increases, as expected. Previous studies using various PPAC detectors show that the amplitude
Figure 2.14: The effect of the anode reduced electric field, $E_r$ in the chamber on the output signal rise time at various pressures.
Figure 2.15: The effect of the anode input voltage on the output signal amplitude at various pressures. The trendline follows a power law determined at each pressure through regression analysis.
of the signal from the anode increases exponentially with increasing electrical field for a constant gas pressure [56][60] which is also true for this detector. The results are displayed on a log-linear plot as the trendlines are fitted with a power law determined through regression analysis. The relationship between amplitude and voltage is found to be,

\[ A \propto aV^b, \]

where \( a \ll 1 \) and \( b = 4.22 \pm 0.14, 3.96 \pm 0.20, 3.35 \pm 0.22 \) for pressures 12.5, 11.5 and 10.5 mbar, respectively, as shown in Figure 2.15. Figure 2.14 includes the relationship between signal rise time and the reduced electric field \( E(r) \), defined as electric field per unit of pressure, \( P \) as shown in Equation 2.10,

\[ E(r) = \frac{E}{P} \tag{2.10} \]

It is also true that the signal amplitude decreases with increasing pressure. If the pressure is too high, however, the output signal no longer represents the energy of the original particle as energy will be lost by scattering collisions as the electrons drift towards the anode. On the other hand, not enough primary ionisations will occur to produce a clear signal when the pressure of the isobutane gas is too low.

The timing resolution of the detector, \( \sigma_t \), is quantified by,

\[ \sigma_t = \frac{\sigma_A t_r}{A} \]

where \( \sigma_A \) is the uncertainty of the amplitude, \( A \), and is due to fluctuations in the signal strength and noise present in the preamplifier signal. The amount of low level noise is quantified by measuring the signal with no source in the chamber which
gives the amplitude of the noise at ~20 mV. The optimum ionizing conditions which provide the maximum signal-to-noise ratio and lowest rise time were determined to be when the pressure is \((10.3 \pm 0.05)\) mbar and input voltage \((650 \pm 0.5)\) V. Under these conditions, a pulse amplitude of \((85.25 \pm 0.25)\) mV is achieved with a rise time of \((1.44 \pm 0.024)\) ns. This gives a potential timing resolution of 337 ps, an improvement of \(~75\%\) on the current stop detector design. It is therefore worth further investigation so that this type of stop detector can be implemented in future STEFF experiments.

### 2.9.3 Summing anodes

A larger detector area can be achieved by summing together individual anode cells shown in Figure 2.12 in the PPAC detector. An \(^{241}\text{Am}\) source was placed under anode three and collimated so that theoretically, no radiation effects will be felt by the other anodes. This tests the extent to which the signals prior to entering the preamplifiers are isolated from each other.

The anode signal is passed through a preamplifier before being processed. To test the interaction of the preamplifiers the outputs from three and four, as shown in Figure 2.11 were connected to separate channels. Two pulses from anode three and four are shown in Figure 2.16. The signal in channel four lag just behind those in anode three by approximately 8 ns. It is also inverted suggesting that the real signal from ionization picked up in channel three is being induced into four. This result suggests that the measurements are sensitive to induced signals created according to the Shockley-Ramo theorem discussed in Section 5.2.1. The movement of charge is inducing a small signal in the detectors adjacent to anode three. This interference explains an unexpected result in that the rise time increases when output three and four were connected but decreases with any more addition outputs. This can be overcome by grounding the preamplifiers individually to reduce the amount of noise detected.
Figure 2.16: Anode output signals from anode three and four showing that the signals are not isolated when the preamplifiers are grounded collectively.

by the electronics. This is a significant finding which can be applied to the current STEFF set up, where the external preamplifiers on the current MWPC are grounded by clamping them to the outside of the chamber to reduce noise and ensure no signals are being induced into other channels.
Chapter 3

Improvements to single gamma spectra

3.1 Overview of corrections

Analysis and refinement of selected experimental data is performed offline using sortcodes. An overview of the correction procedure is given in Figure 3.1. In this diagram, blue arrows indicate stages of the process which have been iterated, resulting in small modifications to the data after each iteration. The following sections describe each correction in further detail.

3.2 Neutron background removal

Detection of neutrons by NaI(Tl) detectors can be difficult to distinguish from detection of gamma-rays. Discussed here is the response of scintillation detectors to neutrons, and how contamination from neutrons is avoided by separating signals via the difference in gamma-ray and neutron flight times.
Figure 3.1: Overview of corrections used to improve the gamma-ray energy spectra.
3.2.0.1 Neutron pulses

Detected pulses from neutrons are classed as either prompt or delayed. Prompt pulses occur within a few $\tau_{ns}$ of the neutron arriving in the detector due to inelastic scattering of the neutron in the scintillator crystal [52].

At least $\sim 100 \ \mu s$ exists before delayed pulses are detected due to the time taken to moderate fast neutrons to thermal energies before radiative capture by the material of the crystal. When this neutron-activated crystal goes onto decay, it is the corresponding capture gamma-rays which lead to pulse detection. If neutron capture results in a radioisotope which then decays, further delayed pulses are likely. For example, $^{24}\text{Na}$ has a half-life of 15 hours and $^{128}\text{I}$ of 25 minutes.

The intrinsic efficiency of neutron detection measured for a range of neutron ener-
Figure 3.3: Comparison of the three most commonly cited statistical models predicting prompt neutron energy spectra compared with experimental data [62].

...gies within a 7.6 x 7.6 cm NaI(Tl) scintillator is shown in Figure 3.2. In this example the most likely neutron energy to be emitted from fission is 1 MeV. At this energy, ~10% of the emitted neutrons will be detected by the NaI(Tl) detector.

3.2.1 Neutron emission

There are several statistical models implemented to predict the neutron energy distribution following fission which are compared in Figure 3.3. A detailed overview of the measured and calculated parameters involved in predicting neutron emission spectra from fission is given in [63] and [15]. The majority of neutrons are emitted during the evaporation of the fully accelerated fragments. The most frequently cited approximations for the neutron energy distribution $N_M$, are the Maxwellian approximations, outlined by Terrell [64],...
This approximation is for neutrons in the laboratory frame and is characterised by nuclear temperature $T_M$ which is determined by the average neutron energy, $E = \frac{3}{2}T_M$. A least squares fit method is employed to match the parameters in this distribution to experimental data. The assumption is made that all the fragments have the same kinetic energy per nucleon, and that the neutron evaporation spectrum, follows a Maxwellian distribution. In the laboratory frame of reference, the neutron energy can take the form of a Watt distribution [65],

$$N_m(E) = \frac{2\sqrt{E}}{\sqrt{\pi T_M^{3/2}}} \exp\left(\frac{-E}{T_M}\right), \quad (3.1)$$

where $A = 1/T_e$ and $B = 4E/T_e^2$ and the nucleus temperature following evaporation of one neutron is $T_e$. Watt theorises that the centre-of-mass frame of the neutron spectrum is approximately a Maxwellian spectrum, by also assuming that on average the fission fragments have the same kinetic energy per nucleon. Using accepted values, calculated from experimental data and statistical interpolation for the coefficients [62], Equation 3.2 can be condensed to give the probability distribution:

$$P(E) = a \sinh(\sqrt{2.29E})e^{-(bE)}MeV^{-1}, \quad (3.3)$$

where the probability $P(E)dE$ of a neutron following fission has an energy between $E$ and $E + dE$. Using accepted values $a = 0.4865$ and $b = 1.036$, which allow the distribution to fit well with experimental data, the approximation can be implemented
to describe the neutron energy distribution as shown in Figure 3.4.

3.2.2 Conversion to TOF distribution

A theoretical neutron timing spectrum which represents the time taken for an emitted neutron to travel from the fissioning fragment to the centre of the detector is derived from the neutron energy distribution described in Section 3.2.1.

Each scintillator detector face is positioned 20 cm from the centre of the detector array. By approximating that neutrons will travel this distance plus an additional 5.25 cm into the 10.5 cm long crystal then the total distance, $D$ combined with the kinetic energy of a single neutron given by Equation 3.3 gives the velocity, $V$,

$$V = \sqrt{\frac{2E}{m_n}}$$  \hspace{1cm} (3.4)

so that the of the neutron, $\text{TOF}_n$ is calculated from,
\[ \text{TOF}_n = \frac{D}{V} \]  

(3.5)

According to the Watt distribution a prompt neutron will have average energy 0.7 MeV and will take 21.8 ns to reach the NaI(Tl) detector. The highest energy neutrons, at 12 MeV will be detected at 5.27 ns, whereas gamma-rays will take 0.84 ns.

The energy probability distribution is then converted to a time-of-flight probability distribution by implementing the transform density function;

\[ g(y) = f(x(y)) \left| \frac{dx}{dy} \right| . \]  

(3.6)

The function \( f(x)y \) is substituted with the energy probability spectra \( P_E(E) \), given by Equation 3.3. \( \left| \frac{dx}{dy} \right| \) is the kinetic energy \( (E = \frac{1}{2}mv^2) \) derived as a function of \( t \) and rearranged to function of \( d, m_n \) and \( E \),

\[ \left| \frac{dE}{dt} \right| = \frac{1.59m_n^{\frac{2}{3}}E^{\frac{3}{2}}}{d}, \]  

(3.7)

so that the probability distribution as a function of time, \( P_t(t) \) is now,

\[ P_t(t) = P_E(E) \frac{dE}{dt}. \]  

(3.8)

### 3.2.2.1 Broadening of timing peak

The theoretical timing probability distribution must be broadened to account for the detector timing resolution. This is achieved by convolution of the calculated probability distribution \( P_t(t) \) given by Equation 3.8 and \( g(t, \mu, \sigma) \), with a normal distribution;
Figure 3.5: Theoretical neutron timing distribution before and after convolution with a Gaussian function. The spectra has been normalised so that the peak maxima are matched.

\[ g(t, \mu, \sigma) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{(t-\mu)^2}{2\sigma^2}} \]

where the mean, \( \mu = 0 \) and the standard deviation, \( \sigma \) corresponds to a FWHM of 3.29 ns, measured from the first half of the NaI(Tl) TAC peak i.e. the fastest timing signals. The two functions overlap and their areas combine to give a broadened version of \( P_t(t) \) described by,

\[ [f * g](t) = \int_0^t P(\tau)g(t - \tau)d\tau, \]

where \( \tau \) is the dummy integration variable in the convolution. The distribution before
and after convolution with a Gaussian is shown in Figure 3.5.

The prompt gamma-ray distribution is modeled as a Gaussian function with standard deviation, \( \sigma \) also matched to that measured in the TAC peak. The time scale on the NaI(Tl) TAC measurements is calibrated by inducing artificial pulses of a known frequency into the TAC channels during the experiment. The time delay between induced pulses is then set on the acquisition system so it is known that each detector is calibrated to 0.1 ns per channel.

The theoretical distributions are compared to experimental NaI(Tl) TAC signals in Figure 3.6. The theoretical summation of prompt gamma-ray and prompt neutron distributions is normalised and agrees well with the experimental TAC timing data. There is a tail present in the experimental spectra between 10 and 25 ns which is more pronounced than in the calculations. This is thought to be due to delayed isomer decays or delayed pulses from to inelastic scattering of neutrons. By determining the expected position of neutrons within the timing signal, a timing gate is now set which removes contamination from neutrons and contains only the signals from prompt gamma emission.

3.3 Polygonal timing gate and random background subtraction

The timing gate applied to the NaI(Tl) TAC to remove the neutron contamination, as discussed in the previous section, is dependent on the TAC’s response at different gamma-ray energies. The response is tested at different energies using a timing-energy 2D matrix which acts as a fragment-gamma timing signal. The efficiency of the TAC with various gamma-ray energies is tested by gating on energy and monitoring the shift in the position of the TAC peak. The results shown in Figure 3.8 confirms that the timing efficiency is not constant over a range of energies and that the low-
Figure 3.6: Comparison of theoretical prompt gamma and neutron TAC signals with experimental data for detector 0.

energy gamma-rays are detected later. This is the expected behaviour of leading-edge timing signal [52]. The position of the peak shifts by a maximum of 0.8% across the whole energy range and shows a similar trend for each detector.

The TAC peak distribution is fitted as an approximate Gaussian as shown in Figure 3.7 to and its sigma value calculated. The distribution is then re-fitted, with the lower and higher cut off point for which to make the Gaussian set as a fraction of sigma. For the lower gate this is $4\sigma$ from the peak and for the higher gate this is $3\sigma$ sigma. The second half if the peak requires a lower sigma value as the shape on this side is not quite Gaussian. This method ensures that the polygon gates are set to exclude the same fraction of the prompt data from each energy gate.

The efficiency curve signifies that the timing gate applied to exclude neutrons as described in Section 3.2.2 is more effective as a polygonal gate rather than rectan-
Figure 3.7: Timing peak corresponding to the 140 - 400 keV energy gate for detector 0. The centroid position is determined by fitting it to a Gaussian distribution with lower and upper limits set at $4\sigma$ and $3\sigma$, respectively.

gular. A gatemap is created for each detector, which contains the co-ordinates that follow the shape of the TAC efficiency curve presented in Figure 3.8. The array of gatemaps is introduced into the sortcode so that the energy versus NaI(Tl) TAC signal 2D matrix is now truncated to exactly that mapped by the polygons as Figure 3.9 displays. The timing gate is set between $4\sigma$ and $\frac{2}{3}\sigma$ on the left hand side of the TAC peak for each detector. Corresponding energy signal that falls within this enclosed region of the timing polygon will be accepted as coming from prompt gamma-rays, and any signals outside this will be rejected.

Random coincidences are subtracted by again using the energy-timing 2D Matrix. Limits are set on the TAC with the same timing limits described above. The channels away from the timing peak are present due to random coincidences or electronic noise are therefore excluded. The energy distribution corresponding to this background is then stripped away. This corresponds to a removal of 7% of the total energy distribution. Following random background subtraction and polygonal gating, 20% of the
Figure 3.8: Position of NaI(Tl) TAC peak as a function of gamma-ray energy for detectors 0, 2 and 4.
Figure 3.9: NaI(Tl) timing signal before and after polygon gate map gates have been applied for one detector (detector 7) and before random coincidence subtraction. Any corresponding energy signal that falls within $4\sigma$ and $\frac{2}{3}\sigma$ on the left hand side of the TAC peak is accepted.
Figure 3.10: Cumulative distribution of gamma-rays from fission used as the input distribution of gamma gun energies into Geant4. Each iteration has a modified fraction of multiple hits removed.

original gamma energy distribution will remain.

### 3.4 Multiple hit subtraction

A multiple hit is classed as a single NaI(Tl) pulse detection which is actually the summation of two or more gamma-rays. This will occur when two or more gamma-rays enter the detector within a small enough amount of time so that they cannot be distinguished from each other. The result is that the pulses pile up and energy is deposited in that detector that is indistinguishable from high-energy single gamma-ray events. The contribution from multiple hits is calculated as follows.
Geant4 simulates the interaction of the gamma-rays with the STEFF array. This includes gamma-rays scattering from one detector to another and scattering within the surrounding materials. Relatively large quantities of aluminum surround the detectors as it is used for the frame holding the NaI(Tl) detectors in place. The passive detector shielding is tungsten (W) to reduce Compton scattering between the NaI crystals. The steel frame acting as a base for STEFF is also incorporated into the Geant4 simulation as are the sodium and iodide crystal elements.

The default input into Geant4 in the current implementation is a range of mono-energetic gamma-rays, starting at 10 keV and incrementing by 10 keV up to 20 MeV. Although this provides a wide energy range of gamma-rays, it is not a realistic representation of the spectrum released during fission.

The input ‘gammagun’ energies into Geant4 are instead generated by the experimental neutron-excluded gamma-ray spectra as a more realistic representation of the emitted gamma-rays. This is converted to a cumulative probability distribution (CPD) shown in Figure 3.10, representing the probability of an individual gamma-ray energy entering the array. A random number between 0 and 1 is generated and the corresponding energy from the CPD is applied. The probability of the energy being selected is therefore dependent on the gradient of the curve, the gradient is much steeper at lower energies and is therefore more likely to be selected. This energy is incorporated into the Geant4 simulation, which will now call upon this distribution to generate the gamma-rays fired into the detector.

With this new input method, Geant4 is run with 10 million gamma-rays to produce response functions. The quantity of energy deposited in each detector, if any, for each fired gamma-ray is listed. From these response functions, the probability of detecting a ‘multiple hit’ is calculated.

For a certain number of gamma-ray histories, determined by the calculated mul-
Figure 3.11: Fraction of multiple hits in detector 0 calculated from response functions produced using Geant4 simulations for the original data then the first 3 iterations.
Multiplicity distribution described in Section 3.6, the number of multiple hits within a single detector is counted. The probability of two or more being counted within the defined number of histories is converted to a fraction of the total hits. This multiple hit fraction is displayed in Figure 3.11.

It can be concluded from this that the fraction of detection due to multiple hits increases as the input gamma-rays energy increases up to ~2 MeV where the multiple hit fraction remains at ~25%. This fraction is then removed from the fission spectra and the gamma-ray CPD, is re-calculated and input into Geant4.

One of the drawbacks of using this method, is that it is likely to cause an underestimation of the number of gamma-rays released per fission at lower energies, as 10% of gamma-rays are removed during multiple hit subtraction process. These are likely to be low energy gamma-rays that have been summed together and then removed so that every count classed as a multiple hit is actually the removal of two separate, low energy gamma-rays. To account for this the final spectra is re-scaled.

The results of this analysis explain the deviation of STEFF energy distribution data from previous measurements at high energies as discussed in later sections.

3.5 Deconvolution

A significant stage of the gamma-energy correction process is deconvolution. Emitted gamma-rays from the fissioning nucleus lose energy before detection by the NaI(Tl) scintillator due to absorption into the surrounding materials and by Compton scattering. By applying a deconvolution correction to account for these losses, a more realistic representation of the emitted energy spectra, rather than what is observed is determined.

Geant4 simulations are again utilised to determine the interaction of gamma-rays with STEFF. The output is a series of response functions, each containing 10 mil-
lion simulated gamma-rays of a specific energy between 0 to 20 MeV. Each response function contains the proportion of energy deposited in each detector.

The response functions are converted into a 2D response matrix, gamma energy distribution against absolute total efficiency as a function of energy, displayed in Figure 3.12. Matrix inversion is then performed to deconvolve the energy. The detected spectrum, $D(E)$ can be expressed as,

$$D(i) = \sum R(i,j).S(j),$$

where $S(j)$ is the emitted spectrum and $R(i,j)$ is the response function of the detector provided by Geant4 simulations. In matrix form, it is;

$$
\begin{pmatrix}
D_i \\
. \\
. \\
D_i
\end{pmatrix} = 
\begin{pmatrix}
0 & 0 & 0 \\
0 & 0 & . \\
0 & . & . \\
0 & R_{i2} & R_{ij}
\end{pmatrix} \times 
\begin{pmatrix}
S_j \\
. \\
. \\
S_j
\end{pmatrix}
$$

(3.12)

In order to transform the response matrix into triangular form, detector resolution and summing effects are not included in the Geant4 simulations. This ensures that all of the detected energies are less than or equal to the emitted energies. A Gauss-Jordon elimination is then used on the response matrix described in Equation 3.12, whereby the coefficients are treated as a series of simultaneous equations. A sequence of operations is then performed on them; either swapping two rows, multiplying by a non-zero or adding a multiple of one row to another. Following this process, the response matrix is inverted.

The inversion is optimised by three iterations. Following each iteration, a residual matrix is created which is the product of the original matrix, $R(j,k)$ and the inverse,
\( R^{-1}(i, j) \) multiplied by 1000,

\[
    r(i, k) = \sum_j \{ R^{-1}(i, j) R(j, k) \} \times 1000. \tag{3.13}
\]

The final residual matrix, \( r(i, k) \) following the final inversion iteration is therefore a diagonal in which all channels are zero with the exception of the diagonal have 1000 counts as shown in Figure 3.13. The residual matrices are tested, and the test is then iterated until the total \( \chi^2 \) deviation is minimised.

The uncertainties in this method are treated as Poisson within the raw spectrum and are carried through the inversion process. Where the energy is re-scaled, a randomisation function is used to prevent build up in certain bins. This method relies heavily on the array efficiency at all energies between 200 keV and 20 MeV predicted by the Geant4 simulations. These efficiency measurements, are validated with experimental data using \(^{137}\text{Cs} \), \(^{60}\text{Co} \), and \(^{241}\text{Am}^{9}\text{Be} \) sources which are discussed and
compared to Geant4 predictions in Chapter 4.

Figure 3.14 displays the measurements combined for all 11 NaI(Tl) detectors before and after deconvolution. The distribution agrees well with previous measurements for the mid-energy range (1-3 MeV) but there are discrepancies at high and very low energies.

3.6 Multiplicity and sum energy simulation

Monte Carlo (MC) simulations have been utilised to calculate the multiplicity and average sum energy. The detected fold is the number of gamma-rays detected, and the multiplicity is the number of gamma-rays emitted per fission event.

The multiplicity distribution shown in Figure 3.15(b) is based upon the statistical probability distribution used by Huizenga and Vandenbosch [45]. A MC simulation has been implemented which takes samples from this probability distribution to create a fission event. For a given input parameter, $B$, as described in Equation 1.21, a distribution for the fragments’ spin is generated.
Figure 3.14: Prompt gamma-ray energy distribution emitted from thermal fission of $^{235}\text{U}$. The raw experimental measurements are summed over 11 NaI(Tl) detectors and compared to the same distribution after the deconvolution process.
Figure 3.15: Distributions showing (a) the detected fold (b) gamma-ray multiplicity probability (c) detected sum energy and (d) emitted sum energy. In (a) and (c), STEFF measurements (black) are matched to Monte Carlo simulations (red).
A linear relationship is assumed to exist between the spin quantum number and the yrast multiplicity since yrast decays are mostly responsible for dissipating the spin of the fragment. Two independent geometrical probability distributions are then used to generate the number of statistical gamma-rays ($NS_1$ and $NS_2$) from each fragment and combined to give the total multiplicity value of $7.74 \pm 0.12$. More information about this process is given in Section 1.5.5 and [66].

The fold distribution shown in Figure 3.15(a) was produced by combining multiplicity with the results from a second Monte Carlo simulation that calculates the response of the array to a given multiplicity. The efficiency of the array, including scattering properties and peak-to-total ratios, has been obtained from measurements made with a $^{60}$Co calibration source within the STEFF array and from Geant4 simulations described in Chapter 4. A $\chi^2$ minimization is performed to fit the fold distribution to obtain, $B$ and the mean number of statistical photons, which is constrained to be the same for both fragments.

The detected sum energy shown in 3.15(c) is calculated using the Geant4 generated library of response functions described in Section 3.4. The probability distribution is assumed to be a skewed Gaussian, as shown in 3.15(d) with parameters varied to optimise the fit to that generated by MC methods. The detected fold and sum are compared to STEFF data in Figure 3.15(a) and (c), respectively. Through these procedures the average total emitted gamma-ray energy has been determined as $8.40 \pm 0.26$ MeV.
Chapter 4

Validation methods

A single NaI(Tl) detector from the STEFF detector array was tested to determine the absolute photopeak and total efficiency as a function of gamma-ray energy. The sources chosen for the efficiency measurements were $^{137}\text{Cs}$, $^{60}\text{Co}$ and $^{241}\text{Am}$, as they emit a range of gamma-ray energies. The interaction of the source gamma-rays with the detector was simulated in Geant4 to test the reliability of the measurements. Geant4 simulations were also implemented to verify the deconvolution process described in Section 3.5.

4.1 Source decay paths

The decay schemes of $^{137}\text{Cs}$ and $^{60}\text{Co}$ are shown in Figure 4.1. $^{137}\text{Cs}$ decays via two possible decay channels. The majority of decays (~ 95%) occur via $\beta$-emission to an $11/2^-$ isomeric state in $^{137}\text{Ba}$, which de-excites with a half-life of 2 minutes by emission of a 662 keV gamma-ray. It is reasonable to approximate that for each nuclear decay, one 662 keV gamma-ray is emitted.

$^{60}\text{Co}$ undergoes $\beta$-decay to a $4^+$ excited state in $^{60}\text{Ni}$ which then returns to the ground state by prompt emission of two gamma-rays of energies 1.17 and 1.33 MeV.
Figure 4.1: Decay scheme of $^{137}$Cs and $^{60}$Co showing the emission of gamma-rays of energy 0.662, 1.17 and 1.33 MeV.

The gamma-rays correspond to the de-excitation of $^{60}$Ni from the $4^+ \rightarrow 2^+$ state and then from the $2^+ \rightarrow 0^+$ state. From this dominant decay chain an intensity ratio of 1:1 is expected at these gamma-ray energies. The source strength of the $^{137}$Cs and $^{60}$Co was calculated from the date of manufacture to be (50000 ± 5200) Bq and (330 ± 10) kBq, respectively.

As the $^{241}$Am$^{9}$Be source is a combination of two isotopes, calculating the quantity of gamma emission per decay is a more convoluted process. $^{241}$Am decays by $\alpha$ emission as described by,

$$^{241}Am_{95} \rightarrow ^{237}Np_{93} + ^4\alpha_2. \quad (4.1)$$

The $\alpha$ particle carries discrete energies with ~ 84% carrying 5485 keV which subsequently causes an ($\alpha, n$) reaction in $^9$Be, depicted in Figure 4.2, producing an excited state of $^{12}$C,

$$^9Be_4 + ^4\alpha \rightarrow ^{12}C_0^* + ^1n_0. \quad (4.2)$$
Figure 4.2: Decay scheme depicting the \((\alpha, \gamma)\) reaction, resulting in emission of a 4.44 MeV gamma-ray, of \(^9\)Be and \(^{12}\)C, adapted from [67].

The de-excitation of \(^{12}\)C described in Equation 4.2 results in the emission of gamma-rays through a number of possible decay channels. However, 80% of these emissions, have energy 4.44 MeV [67]. As Equation 4.1 depicts, there is also an associated neutron flux emitted in this decay process.

According to the certificates produced by the manufacturers of the source, Amersham International plc, the \(^{241}\)Am\(^9\)Be source was measured to be \((100 \pm 10)\) mCi in 1970 and is producing neutrons at a rate of \(2.96 \times 10^5\) n/sec. For accurate gamma yields the gamma-ray to total neutron ratio, \(R\) is required,

\[
R = \frac{S_\gamma}{S_n},
\]

where \(S_\gamma\) and \(S_n\) is the number of gamma-rays and neutrons emitted per second, respectively. This ratio is dependent on the way in which the AmBe source is constructed and there is some discrepancy surrounding this characteristic in literature
A theoretical approximation of \( R \), is described by [70] as,

\[
R = \frac{S_{\gamma}}{S_n} = \int_0^{E_{\text{max}}} \chi(E_\alpha) \left[ \frac{\int_0^{E_\alpha} \sigma_{\gamma_1}(E)/(dE/dx)dE}{\int_0^{E_\alpha} \sigma_n(E)/(dE/dx)dE} \right] dE
\]  

(4.4)

where \( \chi(E_\alpha) \) is the fraction of \( \alpha \) emissions of energy \( E_\alpha \) from \( AmO_2 \), \( \sigma_{\gamma_1}(E) \) is the cross section of 4.44 MeV gamma-ray production, \( \sigma_n(E) \) is the cross section to produce a \( ^9Be(\alpha,n) \) reaction and \( dE/dx \) is the stopping power in the \( ^9Be \) for \( \alpha \) particles of energy \( E_\alpha \). Inputting these parameters gives a value of \( R = 0.57 \pm 0.06 \). By implementing this value into Equation 4.4 a gamma-ray emission rate of \( 1.68 \times 10^5 \) gamma-rays per second is determined. This gives a rate of \( S_{4.44} = 1.34 \times 10^5 \) s\(^{-1}\).

The source was placed \( (0.40 \pm 0.01) \) m from the detector. This is the nearest distance for which the detector dead time is <10% due to the high neutron flux present. As measurements for the other sources were made at 20 cm from the detector, to reflect that the target is 20 cm away from the detector in the STEFF array, the intensity of the photopeak is corrected using the relationship,

\[
I \propto \frac{1}{r^2},
\]  

(4.5)

where \( I \) is the measured intensity of the photopeak and \( r \) is the distance between the source and the detector.

### 4.1.1 Photopeak efficiency

The absolute photopeak efficiency, \( E_p \), for a gamma-ray of a specific energy is calculated by,

\[
E_p = \frac{N_c}{N_\gamma},
\]

(4.6)

where \( N_c \) is the number of counts in the photopeak and \( N_\gamma \) is the number of gamma-rays of that energy emitted by the source. The geometry of gamma-ray emission is
Figure 4.3: Photopeak measured by NaI(Tl) detector 0 compared to previous measurements [71].

not taken into account in these measurements. The results of the photopeak measurements for three energies of gamma-rays, at 662 keV, 1.33 MeV and 4.44 MeV, are compared with previous measurements and to a Geant4 simulation of this set-up in Figure 4.3.

The Geant4 simulation in this configuration consisted of a single NaI detector, with dimensions and materials defined to be an exact reproduction of the real NaI(Tl) detector. A hundred million monoenergetic gamma-rays were then fired into the detector at a distance of 40 cm for 4.44 MeV, and at 20 cm for 662 keV and 1.33 MeV. Further simulations were made at this distance for 2, 2.5, 3 and 3.5 MeV gamma-rays.
Figure 4.4: The absolute total efficiency of detector 0 as a function of energy.
The simulation cannot predict a quantitative photopeak efficiency measurement because many factors affecting the detector efficiency are not defined in the Geant4 simulation. For example, scattering of gamma-rays within the detector, the purity of the NaI crystal and purity of the Tl doping agent, the efficiency of the electronic components such as the preamplifier, and the temperature of the outside environment. Geant4 does provide, whilst not including these factors, an accurate relative efficiency measurement, which has been normalised to experimental data in Figure 4.3.

Figure 4.3 shows a comparison of the results measured with the STEFF NaI detector and similar measurements, made with a $4 \times 4$ NaI(Tl) detector with gamma-rays ranging from 25 keV to 2500 keV [71]. In this publication, photopeak efficiencies have been measured by positioning sources 46 cm away from the source, so the intensity of the photopeaks is corrected for the extra distance using Equation 4.5. Also included in Figure 4.3 is the photopeak efficiency measured using the sum-peak method which is described in Section 4.1.2.

The intrinsic total efficiency, the number of gamma-rays of all energies incident on the detector, is also measured and compared to the Geant4 simulation. It can be seen that in all of these measurements the photopeak and total efficiency falls exponentially as a function of increasing energy. The agreement between Geant4 and experimental data confirms that Geant4 is a suitable tool for measuring the efficiency of STEFF and can be legitimately implemented in the deconvolution process. The results measured by [71] are slightly lower than those measured by detector 0, which is to be expected for the smaller crystal size of the detectors used.

The total efficiency measured for AmBe has not been included in these results. This was because of the considerable background due to neutron induced gamma-rays in the surrounding materials. These gamma-rays were dominant in the lower energy ($< 2.7$ MeV) region due to the nature of excitation of the surrounding materials and
Figure 4.5: Geant4 simulation predicting the absolute total efficiency as a function of gamma-ray energy of the STEFF array.

will, therefore, not significantly affect the photopeak efficiency measurement at 4.44 MeV.

The absolute total efficiency prediction produced by Geant4, for the whole STEFF array is shown in 4.5. The total efficiency curve dips at ~ 2 MeV as detection from gamma-rays due to the photoelectric effect and Compton scattering is reduced and rises again where emission from pair production dominates. This is typical of an NaI(Tl) scintillator detector as explained in more detail in [52].

4.1.2 Efficiency of NaI(Tl) array

An alternative calculation, ‘the sum peak method’, to determine the photopeak
efficiency of the detectors was utilised when STEFF was in situ at PF1B. A $^{60}$Co source was placed at the centre of the detector array, but its strength was not known to a high enough degree of accuracy to implement the photopeak efficiency method described by Equation 4.6. The sum peak method eliminates the requirement of a known source strength. The energy spectrum measured from this source, is displayed in Figure 4.6 where measurements for each detector have been aligned using gain matching coefficients.

The efficiency of the detector at 1.33 MeV can be determined by measuring the intensity of the sum peak and the 1.17 MeV peaks. The intensity at 1.17 MeV, $T_{1.1}$ is given by,

$$T_{1.1} = N \varepsilon_{1.17}, \quad (4.7)$$

where $\varepsilon_{1.17}$ is the intrinsic photopeak efficiency at 1.17 MeV and $N$ is a normalisation factor which takes into account geometry and distance from the source to the detector. The intensity of the sum peak, $T_{sum}$ is dependent on the efficiency at 1.17 MeV and 1.33 MeV by the relationship,

$$T_{sum} = N \varepsilon_{1.17} \varepsilon_{1.33}. \quad (4.8)$$

The ratio of the intensities given in Equations 4.7 and 4.8 cancel the normalisation factor to give the intrinsic photopeak efficiency at 1.33 MeV,

$$\varepsilon_{1.33} = \frac{T_{sum}}{T_{1.17}}. \quad (4.9)$$

Compton background was subtracted before the intensities were measured. The measurement is done by fitting a Gaussian distribution to each peak and integrating
<table>
<thead>
<tr>
<th>Detector</th>
<th>$\varepsilon_{1,3}$</th>
<th>Peak-to-total</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.00990</td>
<td>0.1318</td>
</tr>
<tr>
<td>2</td>
<td>0.01118</td>
<td>0.1095</td>
</tr>
<tr>
<td>3</td>
<td>0.00552</td>
<td>0.1097</td>
</tr>
<tr>
<td>4</td>
<td>0.01332</td>
<td>0.1072</td>
</tr>
<tr>
<td>5</td>
<td>0.00632</td>
<td>0.1223</td>
</tr>
<tr>
<td>6</td>
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<td>0.0906</td>
</tr>
<tr>
<td>7</td>
<td>0.01550</td>
<td>0.1464</td>
</tr>
<tr>
<td>8</td>
<td>0.00835</td>
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</tr>
<tr>
<td>9</td>
<td>0.00809</td>
<td>0.1038</td>
</tr>
<tr>
<td>10</td>
<td>0.00628</td>
<td>0.1114</td>
</tr>
<tr>
<td>11</td>
<td>0.01322</td>
<td>0.1264</td>
</tr>
</tbody>
</table>

Table 4.1: The photopeak efficiency calculated using the sum peak method for each detector in the NaI(Tl) array and the associated peak-to-total ratio.
Figure 4.6: Calibration of the NaI(Tl) detectors performed in situ for the 11 detector array with $^{60}$Co using offline gain matching coefficients.

Table 4.1 shows the measured photopeak efficiency at 1.33 MeV using this method, and the peak-to-total ratio for each detector. The results show that the photopeak efficiency for each detector varies from 0.55% for detector 3, to 1.55% in detector 7 with the average value being 0.94%. This variation is expected, due to the differences in the individual detector electronics and crystal purities altering specific detector efficiencies. It may also be the case that the intensity of radiation emitted from the source is not uniform in all directions. The same is true for the peak-total ratio which
has an average value of 11.59%.

4.2 Validation of deconvolution

The effectiveness of the deconvolution method, described in Section 3.5, to remove Compton background for each of the calibration sources has been tested. Deconvolution of the $^{137}$Cs source is shown in Figure 4.7 where 82% of the Compton background has been removed. The sigma value and the intensity of the photopeak remains unchanged.

In the deconvolution of the $^{60}$Co source is displayed in Figure 4.8 and shows that 90% of the Compton background has been removed. The width and intensity of the 1.17 MeV peak has remained largely unchanged, other than undergoing a slight ~ 1% narrowing to account for the loss of Compton background in the first half of the peak. The 1.33 MeV peak, however is broadened, with its $\sigma$ value increasing by 3% and the intensity increasing by 11%. This accounts for the differences in detector efficiency at these two energies. As the efficiency is lower at increasing energies, the curve produced by Geant4 shown in Figure 4.3 compensates for this by replacing energy lost in the Compton background into the 1.33 MeV peak. After deconvolution, both peaks have the same intensity, as expected from the decay process. The result of a hundred million 4.44 MeV gamma-rays emitted from inside the centre of the STEFF array is shown in Figure 4.9. The detector resolution in this case is removed in order to show clearly the position of the photopeak as well as the 511 keV, single and double escape peaks. Deconvolution of the $^{241}$Am$^{9}$Be source is shown in Figure 4.10 and shows that 83% of the Compton background has again been successfully removed. This confirms that the deconvolution process is successful at high energies, at least up to 4.44 MeV.
Figure 4.7: The gamma-ray energy spectrum of the $^{137}$Cs calibration source measured using detector 0 before and after the deconvolution process.
Figure 4.8: The gamma-ray energy spectrum of the $^{60}$Co calibration source measured using detector 0 before and after deconvolution.
Figure 4.9: Geant4 simulation showing the interaction a 4.44 MeV gamma-ray with the STEFF array (excluding detector resolution).

Figure 4.10: The gamma-ray energy spectrum of the $^{241}$Am$^{9}$Be calibration source measured using detector 0 before and after the deconvolution process.
4.3 Correction to the TAC signal

The timing signal measured by the NaI(Tl) TAC units is dependent on the coincident TOF measurements. Figure 4.12 shows the 2D matrix of the TOF measured in Arm 2 of STEFF against the signal from an NaI(Tl) TAC. The TOF spectrum is divided into two clusters, the denser cluster corresponds to the light, fast fragments and the second, more distributed cluster to the heavy fragments. Gates were then set along the TOF spectrum, and the position of the NaI(Tl) TAC peak measured. For heavier fragments the signal detected by the NaI(Tl) is shifted to an earlier time as visible in Figure 4.11(a) and 4.12.

This shift is likely to be due to the energy lost by passing through foils between the chambers of STEFF and through the target backing. This effect will be more significant for the heavier fragments that the lighter fragments. The gradient of the matrix is determined and a linear correction a linear correction is introduced in the sortfile to account for this shift. For channels > 1180 the original TAC signal, TAC₀ is altered by,

\[ TAC_{corr} = TAC₀ \times t_{match} + 0.128 \times TOF - 843 \] (4.10)

where \( t_{match} \) is the gain matching coefficient and the \( T_{corr} \) is the corrected TAC signal. The correction is not necessary for channels < 1180 corresponding to lighter fragments as less energy is lost in the foil, due to their faster velocity. The position of TAC peak, after this correction, remains constant with varying TOFs as shown in Figure 4.12(b).
Figure 4.11: The shift TAC position as measured by NaI(Tl) detector 2 with gates on the TOF measured in Arm 2.

Figure 4.12: 2D matrix showing fragment TOF against NaI(Tl) timing signals (a) before and (b) after the linear correction is added to the sortcode.
Chapter 5

Lohengrin data analysis

In June 2011 the ionisation chamber FiFI (Fission Fragment Identifier) was coupled to the Lohengrin recoil mass spectrometer, located at the ILL, Grenoble. Lohengrin is an accurate mass spectrometer that provides a useful way to calibrate electronics and develop algorithms in the sort codes which can then be applied to data collected by the Bragg 1 and 2 ionisation chambers in STEFF (see Section 2.7). A description of the experimental set-up is given here and the relationship between the range of a fragment in an ionisation chamber and its atomic number is tested.

5.1 Lohengrin set-up

The Lohengrin recoil mass spectrometer, PN1, combines magnetic and electric fields to separate charged fission fragments along their 23 m flight path as illustrated in Figure 5.1. A $^{235}$U target is placed in the ‘target position’ on this diagram where it undergoes thermal neutron fission. The combination of these fields separate the fission fragments of the same velocity, into different parabolas defined by their atomic mass.
to charge ratio, $A/Q$. For every 1% difference in energy the fragments are separated by 7.3 cm along their direction of travel and for every 1% difference in mass the fragments are separated by 3.24 cm perpendicular to their direction of travel [72]. FiFI was coupled to the end of this flight path which consists of an SED start detector positioned at the entrance of a 30 cm long axial ionisation chamber, filled with isobutane gas and held at a pressure of 100 mbar. Full details about FiFI are given in [49].

For each experimental run an ionic mass to charge ratio, which is approximately atomic mass to charge ratio ($A/Q$), and an energy value is selected so that fragments within each run have the same velocity. Lohengrin is not able to isolate individual mass ratios when the ratios are similar so the output is a multiplet of $A/Q$ values. For example, in Run 103, shown in Figure 5.2, $A/Q = 5$ is selected. The peaks then correspond to $A = 100, 95, 90$ and 85 where $Q = 20, 19, 18$ or 17, respectively, visible in the energy spectra as peaks to the left of the $A/Q = 5$ peak.
5.2 Signal collection in ionisation chambers

When a fission fragment travels through a gas-filled ionisation chamber, the gas becomes ionised and the resultant electron and ion pairs drift towards the anode and cathode, respectively.

On entering the chamber, the fragment’s mean charge state is relatively high. The fragment then transfers energy to the isobutane gas by electron interaction with the gas molecules. The mean charge state of the fragment decreases along its path until nuclear, rather than electronic, collisions with the gas molecules dominate the energy transfer process [73].

The net effect of the collisions is to decelerate the fragment, at an increasing rate, until thermal energies are reached. The rate of energy loss per unit distance, is quan-
tified by the stopping power, $S$, described by the Bethe Bloch equation,

$$S = -\frac{dE}{dx} \propto Q_f^2 f(v_f),$$

(5.1)

where $Q_f$ is the charge of the fission fragment and $f(v_f)$ is a function of the gas ionisation energy and the fragment velocity. As isobutane gas is a compound C$_4$H$_{10}$, the stopping power of the hydrogen and carbon is added linearly.

As the fragment travels through the chamber, the ionic charge decreases due to electron collection [74], so that Equation 5.1 becomes,

$$S = -\frac{dE}{dx} \propto -(Z_f v_f v_0) \frac{2}{3} f(v_f).$$

(5.2)

where $Z_f$, is the atomic number of the fragment and $v_f$ is its Bohr velocity. The stopping power reaches a maximum, $dE/dx = 0$, when the fragment has gained sufficient electrons that it no longer possesses enough energy to ionise the gas molecules.

The signal pulse shape collected at the anode is a representation of the stopping power of the fragment in reverse [75]. This is because electrons liberated at the end of the fragment’s path will reach the anode more quickly than those liberated initially, due to the difference in path length. Therefore the raw pulse shapes, shown in Figure 5.3, correspond to the rate of charge collection on the anode, $dQ/dT$.

During this experiment, the selected charge state, $Q$ is achieved by stripping the fragment of electrons in the ‘charging unit’ of Lohengrin indicated in Figure 5.1. This characteristic has a weaker effect on the stopping ability of the fragment in FiFi than the atomic number, due to charge equilibration in the entrance window and in the gas of the ionisation chamber. The initial charge state, therefore has a negligible effect on the anode signal. For the following analysis, the charge state is kept constant at $Q = 20$. 

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Figure 5.3: Raw pulses collected on the anode of FiFI.
By collection of the charges, it is possible to calculate the energy of the fission fragment using the ionisation energy of the gas combined with the number of ionisations the fragment created along its path. This method, used to measure the energy and charge of heavy ions is called Bragg curve spectroscopy [76]. The fragment's initial energy is proportional to the area under the curve and the maximum height of the anode signal, is known as the Bragg peak.

To form a Bragg peak in an ionisation chamber the energy per unit mass of the ion must be a minimum of 1 MeV /amu [75]. Therefore low-energy fission fragments, such as those from thermal fission do not possess enough energy to form a Bragg peak. However, a relationship exists between the anode pulse shape which indicates the fragment’s range in the chamber, and the atomic number of the fragment [77]. The distance traveled by the incoming fragment in the ionisation chamber depends on its kinetic energy, mass and charge by the relationship,

$$R \propto \sqrt{AE / Z^b}. \quad (5.3)$$

where $b$ is an unknown parameter. This can be rearranged so that,

$$\frac{R}{A} \propto \frac{v}{Z^b}. \quad (5.4)$$

where $v$ is the fragment’s initial velocity. As Lohengrin runs are already gated on by velocity, the dependence on $Z$ is tested by utilising Equation 5.4.

For each fragment atomic mass, the most probable $Z$ value is determined from the Gaussian distribution of fragment charge states, published in the JEFF 3.2 [78].
5.2.1 Baseline corrections

For accurate range measurements the charge collected for each pulse must be separate from the charge produced by any low-frequency noise or the preceding fragment’s ionisation. For this to be the case the baseline must be reset to zero at the beginning of each pulse trace. Figure 5.4 focuses on the baseline of the raw pulses and illustrates that where the baseline should be flat, as no ionisations have yet occurred, the baseline is in fact rising by approximately 4% of the pulse height. This rise is visible in 5.4(a). The shift in baseline is thought to be due to the inefficiency of the Frisch grid. The Frisch grid should ideally screen all of the electric field caused by the moving ions from the anode, however it is expected that electric field will leak past the Frisch grid, affecting the signal. FiFI uses a small anode relative to the depth of the detector which makes it less sensitive to induced charges as described by the Shockley-Ramo theorem [52]. The electric field leakage rate, $E(x)$ can be approximated by the expression,

$$E(x) = \alpha e^{-\frac{x}{\kappa}}$$  \hspace{1cm} (5.5)

where $x$ is the distance of the moving ions away from the anode. Two parameters, therefore effect the strength of this field: $\kappa$, the rate by which the leaked electric field diminishes and $\alpha$, the initial strength of the leaked electric field. A correction which utilises this relationship is added to the sort code in which $\alpha$ and $\kappa$ are chosen to optimise the baseline restoration in the early part of the trace. The baseline after the Shockley-Ramo correction is shown in Figure 5.4(b), where the optimum constants to achieve this result are $\kappa = 100$ and $\alpha = 0.0008$.

The effect of this correction to the baseline on the energy resolution of each peak was also considered. The energy resolution, $E_r$ is derived from the normal distribution
Figure 5.4: Pulse shapes collected on anode (a) before and (b) after Shockley-Ramo corrections are applied to correct for rising baseline.
Figure 5.5: The effect of parameter, $\kappa$ that accounts for Frisch grid leakage, on the energy resolution of two energy peaks in Run 111 corresponding to $A = 102$ and $A = 97$.

of pulses collected for each mass using the relationship,

$$E_r = \frac{\text{FWHM}}{H_0},$$

(5.6)

where $H_0$ is the centroid peak and the Full Width at Half Maximum (FWHM) of the distribution is proportional to its standard deviation by, $\text{FWHM} = 2.35\sigma$. This provides a quantitative measure of $\kappa$ variation on the pulse shape resolution. The effect of this for $A = 97$ and $A = 102$ is given in Figure 5.5.

This result shows as the value of $\kappa$ increases, although it improves the rising baseline, it make the distribution of pulses for each mass slightly broader. Larger values
of $\kappa$, therefore slightly damage the energy resolution, and the magnitude of this effect is dependent on the mass of the fission fragment.

When $\kappa > 100$, the improved baseline is counteracted as the pulse baseline shifts below zero. $\alpha$ is also incremented in the same way and the optimum energy resolution is achieved when $\alpha = 0.0008$, although this parameter has a much smaller effect on the baseline position.

### 5.2.2 Pulse height defect

Before the ranges of the fragments inside the chamber can be analysed, the Pulse Height Defect (PHD) must be accounted for. The PHD is caused by the incoming fragment losing energy that does not result in charge collection at the anode. This can occur due to non-ionising elastic collisions between the incoming fragment and the isobutane gas atoms or charge recombination between the ion-electron pairs [52].

In this method, an energy gate was set at 100 MeV in each run as highlighted in Figure 5.2. With this constraint in place, an average pulse shape from 3000 pulses for each mass was created. The logarithm of the average pulse traces for the 100 MeV peak for Run 103-111 is shown in Figure 5.6(a). For each increase in selected atomic mass unit, the position of the peak shifts by approximately 5% to a lower energy and the charge collected decreases. This effect is visible in Figure 5.6(b). This is confirmed in Figure 5.7, where the maximum pulse height as a function of fission fragment $A$ is given. These results suggest that heavier fragments result in a higher amount of elastic collisions. For further analysis of fragment’s range, this defect is corrected for so that pulses for each mass have the same pulse height.

After this correction is applied, although the average traces will have the same amount of charge collected for all masses, the lighter fragment pulse shape rise more
Figure 5.6: Average pulse shapes for Run 103-111 where $A=88-102$ respectively with an energy gate at 100 MeV.
quickly. This is because they deposit their energy over a shorter distance than the slower, heavier fragments.

5.2.3 Range measurements

The fragment Range/A is plotted against the most probable Z value for that mass in Figure 5.8. The range of the velocity-gated fission fragments in Runs 103-111 is measured as 5-95% of the full pulse height.

As predicted by Equation 5.4 a correlation exists between Range/A with atomic number. A regression fit of a power law fits the data points well although the range of atomic number is relatively small. The most accurate fit is for Run 103 in Figure 5.2 as this contains the visible mass peaks and therefore the most data points. In this case parameter $b = -0.81 \pm 0.03$. 

Figure 5.7: Pulse Height Defect affecting signals at the anode as a function of fragment mass for E=100 MeV

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Figure 5.8: Range/A as a function of atomic number for various velocities.
The error on the range measurement, $\sigma$ is calculated from the discrepancy of detecting the channel at 5% of the pulse height given by the gradient at that point and the variation in the baseline from its the root mean square, rms before the mean value was taken,

$$\sigma = \frac{\sqrt{2} \text{grad}(5\%) \sqrt{\text{rms(baseline)}}}{\sqrt{\text{rms(baseline)}}}.$$ \hspace{1cm} (5.7)

### 5.2.4 Comparison with SRIM

Normalisation of the range measurement measurements made in FiFI, was possible by comparison to the ranges predicted by the simulation, SRIM (Stopping Range Ionisation Model).

SRIM calculations employ Monte Carlo simulations based upon the `Continuous Slowing Down Approximation (CSDA)` that assumes the particle’s trajectory is a straight line,

$$R_{CSDA} = \int_0^{TKE} \frac{dE}{dE/dx |_{\text{Total}}}.$$ \hspace{1cm} (5.8)

where $dE/dx |_{\text{Total}}$ is the stopping power, $TKE$ is the total initial kinetic energy of the fragment, and $R_{CSDA}$ the mean path length of the charged particle in the absorber. At each collision the ion will gain electrons depending on its $Z, v$ and $Q$ value at that particular collision. The effect of the charge state is based upon the Brandt-Kitagawa approximation [79] and the velocity effects are calculated using the Bethe-Bloch equation. SRIM calculations are semi-empirical as the simulation also employs a large database of experimental data of ranges to match the theoretical calculations to [80].

The input parameters into SRIM are the fragment’s energy (calculated from the velocity), the charge state (atomic number) and the density of the gas. This is derived from the ideal gas law from as the pressure in the chamber is kept at 70.6 mbar.
which gives a gas density of $4.6 \times 10^{-4}\text{gcm}^{-3}$. The most likely atomic number for each fragment mass is taken from the isobaric charge yields, published in the JEFF 3.1.2 [5].

The range outputs are compared to those calculated by the SRIM simulation in Figures 5.9-5.10. The output ranges FiFI are scaled down by a weighted average of the difference between the channels collected and the ranges predicted by SRIM.

The accuracy of ranges predicted by SRIM is quantified by the extent to which the theoretical calculations fit experimental data. For low-energy ions such as thermal fission fragments, it is stated in [80] that 78% will be are correct to within 10%, resulting in a relatively large margin of error. The reasons for this large error are due to some components which are not yet fully included in the SRIM calculations. The mean ionisation energy of the target is a vital part for calculation the stopping
power of the isobutane gas but SRIM loses accuracy when calculating the quantised energy levels, band gap or the shell corrections of the target gas [81]. It may be that an excluded shell correction gives rise to the unexpected jump between ranges shown in Figure 5.11.

The main factor affecting the range measurement is the amount of energy lost in the Mylar film. Fragment energy lost in the 0.8 µm thick Mylar film is deducted before the initial velocity of the fragment is calculated. This energy, was determined using SRIM and is most apparent in the 100 MeV peak where the same energy is selected. It is not the case, as would be expected, that the energy lost in the film is constant function of A and Z when calculated by SRIM. Instead, there is much more energy lost where $Z = 38$ which may again be due to missing atomic shell corrections and
Figure 5.11: Range measurements of pulses gating on 100 MeV peak for Runs 103-111.
therefore not accounted for in the simulations.

It may be that the more consistent values calculated from the average pulse shapes in the sort code provides a more accurate range measurement than the SRIM simulation for fission fragments in isobutane gas. The range measurements performed by these methods, although did not reflect the non-linearity of the SRIM predictions, all fell within the margin of error given by SRIM. These methods can legitimately be applied therefore to the pulse shapes collected in Bragg 1 and 2 of STEFF with an estimation that parameter in Equation 5.4, \( b = -0.81 \pm 0.03 \).
Chapter 6

$^{252}$Cf chain mass yield evaluation

Two modeling methods, GEF (GEneral model description of Fission observable) and the ‘Five-Gaussian Fit’ are tested against experimental data for their ability to determine the chain mass yield distribution of fission fragments from the spontaneous fission of $^{252}$Cf. This analysis was performed at the National Nuclear Laboratory (NNL), Cumbria under the supervision of Dr Robert Mills. Fission mass yields within nuclear databases are also discussed here, highlighting areas requiring improvement and future development work.

6.1 Nuclear databases

JEFF (The Joint Evaluated Fission and Fusion file), is an evaluated data library constructed by international collaborators and maintained by the Nuclear Energy Agency (NEA) [5]. New experimental data, such as that provided by STEFF, is volunteered and must undergo a review process and evaluation before it can be included in the database.

The process of evaluation is performed by an independent review group. Firstly, the relevant data must be extracted from the other information within the publica-
tion. Following this an assessment is made of the methods used, including analysis of any correction factors applied to the data and the associated uncertainties. One of the obstacles faced when inputting reliable information into a database is that data is obtained from published papers that have employed various experimental methods and used various analysis approaches before publishing final results.

Similar to the NEA high priority request for information on prompt gamma rays discussed in Section 1.1, there is also a requirement for accurate fission-product mass yields. This is because of the effect mass yields have in several stages of the nuclear fuel cycle; reactor design and operation, spent fuel reprocessing and calculating reactor burn-up rates [82]. Fission yields are included in all nuclear databases as these are an essential reference for reactor codes, such as those discussed in Section 1.1.

Modeling methods such as GEF and the ‘Five-Gaussian Fit method’ are especially useful in predicting mass yields for nuclei where experimental data is limited. One such example is the mass yield of $^{242}$Cm, produced as a fission product in a nuclear reactor, through consecutive neutron captures by isotopes of plutonium and americium. It dominates neutron emission and fission rates in spent nuclear fuel. Neutrons released from $^{242}$Cm result in neutron capture in $^{130}$I producing toxic $^{131}$I. Accurate mass yield calculations for this element are therefore a high priority to meet safety regulations. $^{242}$Cm undergoes spontaneous fission however with a short half-life (~163 days) making fission yield measurements difficult. As a result there is a shortage of data for this element.

Although $^{252}$Cf fission does not effect the nuclear fuel cycle directly it is often used as a standard for experimental calibrations. It is therefore useful as a test for analysis methods. Chain mass yield data from the spontaneous fission of $^{252}$Cf has been extracted from four published papers and compared to each other. By refining evaluation methods for a fissioning system where there is an abundance of experimental
data, the same techniques can be applied to systems where yield data is limited.

An overview of the techniques used and the output from each published paper is discussed. Currently, JEFF only includes data which has been measured from radio-chemical techniques, or those with $\Delta A < 1$ and $\Delta Z < 1$. Two of these papers, Schmitt [48] and Theirens [83] use ‘fragment identification’ methods and have therefore, not yet been included in an evaluated file. The aim of this analysis is to go someway towards a full evaluation for these papers, necessary for their eventual inclusion in JEFF-3.3.

6.2 Chain and independent yields

Fission yields are measured either as cumulative (chain) or independent yield. After the scission point, neutron rich primary fission fragments (pre-neutron fragments) are formed in an excited state, and quickly emit prompt neutrons to form secondary fragments. These then decay towards stability, principally by $\beta$-decay, forming stable or long-lived nuclides as illustrated in Figure 6.1.

The cumulative yield is measured after the fragment has completed the decay chain and reached its ground state. It is the total number of a specific nucleus produced per fission event. It includes all the fission fragments in a decay chain as well as delayed neutron emission rates [14]. As the cumulative yield includes products after the emission of delayed neutrons and the independent yield does not, the two yields have different values.

An independent fission yield is the number of atoms produced directly per fission event following the emission of prompt neutrons but before $\beta$ decays take place. It can be described as a summation of three factors,

$$I(A, Z, I) = Y(A), f(A, Z), R(A, Z, I),$$  \hspace{1cm} (6.1)
Figure 6.1: Schematic illustrating the primary and secondary fragments in the fission of $^{248}$Cm.
where the sum yield, $Y(A)$ is the total independent yield of all fission products of mass number, $A$. $F(A, Z)$ is the fractional independent yield of all products with atomic mass, $A$ and nuclear charge, $Z$. This term gives the most probable charge state for each fragment mass and follows a Gaussian distribution as described in Section 1.5.3. The isomeric yield, $R(A, Z, I)$ is the fraction of $A, Z$ produced directly from isomer, $I$. The lifetime of the isomer is determined by the fragment spin. The following analysis focuses on the first term, the sum yield, $Y(A)$ which has the largest effect on the independent mass yield.

Most nuclides far from stability decay quickly to become longer-lived nearer stability. Radiochemical methods are historically the first method used to determine yields. In this process, samples are irradiated and the final (or longest lived) radioisotope is isolated radiochemically and measured. These methods, however are not quick enough to capture short-lived nuclides, and measured yields must be corrected for partial decay of the measured nuclide and any undecayed precursors.

The path that the secondary fragments take down a $\beta$-decay chain can be traced by detecting the gamma rays emitted during $\beta$-emission. It is then possible to determine independent yields of the secondary (post-neutron) fragments by selecting and isolating a specific gamma energy, but only if the half-life of that isomer, or the energy of gamma emission is significantly different than those adjacent to it.

6.3 Comparison and evolution of experimental techniques

Nervik in 1960 used radiochemical methods to determine fission yields of $^{252}$Cf for $A=78-169$ with 2% accuracy [84]. The findings show that the heavy and light fragment distributions are not symmetrical around each respective maxima. The uncertainties are calculated from repeated measurements using three sources of different activity and calculating the average deviation from the mean. The accuracy of the results is
tested by assuming that the integration under the yield distribution should give a summation value of 200%. Calculating the integral of one peak gives 98.4%, quantifying the discrepancy at < 2%.

Schmitt followed this in 1965 using TOF and energy measurements in an ionisation chamber to determine fragment masses [48]. Although the resolution is not as high as from radiochemical methods, fine structures were observed that were previously unseen. In the mass region 138-145 there is an inconsistency in the peaks that is not evident in the other data sets which is thought to be due to an increase in prompt neutron emission in this mass region. Uncertainties in this experiment are given as 2.5% calculated from energy losses experienced by the fragment as it travels through foils and from timing errors.

Nine years later in 1974, Flynn added to this yield information, again using radiochemical techniques [85]. Some fragment yields such as $^{90}\text{Mo}$ and $^{111}\text{Ag}$ are determined using the independent yields which are given to 5% accuracy while the cumulative calculations are accurate to 10%. This accuracy is also confirmed by integration under the yield curve although certain mass yield values differ by 22% with those published by Nervik. The peak-to-valley ratio (probability for asymmetric to symmetric fission) is found to be > 370.

Theirens in 1976 [83] used a catcherfoil technique to measure the gamma spectra of the fission fragments in an attempt to improve the results of previous work, by measuring mass with low yields. Prior to this study, mass yields could not be measured unless the yield is > 1% of the peak value. The experiment focused on lower energy gamma-ray spectra of the fission products with energies from 80 keV up to 3 MeV. Fractional independent yields were also determined for $^{134}\text{I}$ and $^{135}\text{Xe}$.

The data sets differ slightly and have different uncertainties, largely due to systematic errors, however the yields follow the same broad trend, as shown in Figure
Figure 6.2: Chain mass yield data from the spontaneous fission of $^{252}\text{Cf}$, determined by various experimental methods and extracted from four published papers [48, 84, 85, 83]. The uncertainties of the experimental values are not included for visual purposes.
6.2. At the fine structure peaks at $A = 113$ and $A = 135$, Nervik explains that their results were more difficult to determine as there was confusion in resolving the specific decays in the long multi-component decay chain in cases where the half-lives and activities were similar.

6.4 Method 1: GEF

Schmidt and Jurudo [86] have created a new a simulation code, GEF to model fission yield information. The code predicts yields for fission of heavy elements for spontaneous fission and with excitation energies up to 100 MeV. The simulation is based on empirical data with constraints applied based on theoretical nuclear physics models.

The GEF model is designed to focus on small structural effects of a nucleus (its micro-structure), against a global background (macrostructure) of fundamental assumptions. The macrostructure background is based upon the Liquid Drop (LDM) model described in Section 1.3. Refinements of the basic droplet model are then added to the simulation due to shell effects, such as pairing correlations and spin-orbit coupling. The simulation assumes that the final properties of the fission fragments depend entirely on the initial state of the parent nucleus before it fissions and bases the model on a dynamical ‘freeze-out’ before scission. At this point the excitation states, kinetic energies, neutron number, atomic number, and the angular momentum of the fission fragments are determined.

An important assumption made by Schmidt is that his approach is basing the predictions on the ‘separability principle.’ This is that the macroscopic effects are defined by the fissioning system before the nucleus splits and the shell and other microscopic effects are defined after scission, along the fragments path. The result is that shell effects are the same for fragments produced from different fission systems.
Empirical data from different experiments can therefore have the same microscopic constraints applied.

A mass distribution simulated using GEF (version 2012/1.1) for post-neutron emission yields from spontaneous fission of $^{252}\text{Cf}$ is shown in Figure 6.3. The model includes the average number of neutrons emitted, $\bar{\nu} = 3.76$.

Figure 6.4 shows the GEF simulation compared to the combined experimental data to test its applicability to experiment. The theoretical mass distribution, simulated by GEF, fits the experimental data well except at the higher and lower mass peaks. At the lower mass peak the experimental values give a higher yield than what is predicted by GEF. Similarly for the heavier masses, the position of the peak is shifted so that the experimental data gives a higher mass value. A correction factor to the theoretical fit is required to account for these differences.

6.4.1 Correction factor

A correction factor can be applied to the GEF calculation to make it consistent with
Figure 6.4: GEF simulation predicting the mass yield of $^{252}$Cf fission compared to experimental values [48, 84, 85, 83].
Figure 6.5: Ratios of experimental to simulated data before and after the correction factor is applied.

The ratio between the yields increases as the mass increases and deviates furthest from the ideal value 1 (where the simulated matches the experimental data exactly) in the very low mass region and in the symmetric fission region. The correction is therefore linear in these regions. Fluctuations are reduced by taking the average yield for each mass value for every \( \pm 3 \) mass units. The simulated yield to experimental...
yield ratio was then re-calculated with this adjustment. The re-calculated ratio and the new average yield is included in Figure 6.6.

The new ratio still suggests that the largest difference occurs again in the region of symmetric fission. This is because yields are so much lower in the symmetric fission region making more difficult measurements and thus have larger uncertainties. The rest of the ratio points fluctuate close to one which suggests that the new experimental data agrees well with the adjusted Schmidt model.

6.5 Method 2: The Five-Gaussian method

James and Mills [87] have produced a library of independent and cumulative yield
measurements based upon the JEFF 3.0 database, released in 1995. In contrast to Schmidt’s code, they have used statistical analysis to fit mass yields and gaps in the databases are filled by extrapolation and interpolation. Experimental uncertainties have also been considered when publishing yields rather than relying on the raw experimental values. The method can be used to predict yields from systems where few measurements have been made.

The Five-Gaussian fit method breaks up the mass distribution into a small number of Gaussian curves: two for each of the heavy and light peaks and one for symmetrical mass region. The chain yield, \( Y(A) \) is constructed from a summation of these curves using,

\[
Y(A) = \frac{N_1}{\sigma_1 \sqrt{2\pi}} \left[ e^{-\frac{(A-\bar{A}-D_1)^2}{2\sigma_1^2}} + e^{-\frac{(A-\bar{A}+D_1)^2}{2\sigma_1^2}} \right] + \frac{N_2}{\sigma_2 \sqrt{2\pi}} \left[ e^{-\frac{(A-\bar{A}-D_2)^2}{2\sigma_2^2}} + e^{-\frac{(A-\bar{A}+D_2)^2}{2\sigma_2^2}} \right] + \frac{N_3}{\sigma_3 \sqrt{2\pi}} e^{-\frac{(A-\bar{A})^2}{2\sigma_3^2}}.
\]

In this equation, \( N_i \) is the coefficient of the \( i \)th Gaussian, which describes the probability of forming a particular split. The width of each peak is determined by \( \sigma_i \) and \( \bar{A} \) is the mean mass of the distribution. \( D_i \) is the separation of the \( i \)th peak from \( A \). By applying a constraint that the chain yield must sum to 2, the substitution,

\[
N_3 = 2(1 - N_2 - N_1),
\]

can be made resulting in a total of 8 parameters to define each yield calculation.

This Five-Gaussian model has been implemented to fill in gaps in the database where yield values are limited. By allowing only one parameter to vary each time, linear or quadratic fits are then made to each of the parameters as a function of mass and repeated until the fit is best matched with the experimental data. This technique
Table 6.1: Parameters which minimise the difference in the square of the yield values simulated by GEF and calculated by Five-Gaussian fit method.

<p>| | |</p>
<table>
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<tr>
<th></th>
<th></th>
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<tbody>
<tr>
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<tr>
<td>$N_2$</td>
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</tr>
<tr>
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</tr>
<tr>
<td>$D_2$</td>
<td>11.41854</td>
</tr>
</tbody>
</table>

has been applied to the experimental data extracted from the four papers described previously.

Input parameters into Equation 6.2 are initially estimated and a yield distribution plotted. This plot is then matched to the simulated yield generated by GEF by the least square fit method, whereby the difference of the square of the calculation and experimentally determined yields is systematically minimised by adjusting the input parameters, until the optimum values are determined. The optimum parameters are shown in Table 6.1 and a comparison of the matched simulated and calculated yields shown in Figure 6.7.

The results from the LSQ fit in Figure 6.7 show that the probability of $N_1$ is much higher than $N_2$ proving that a better match to yield data is achieved as the fit tends towards a single Gaussian distribution. The large width parameter of the third peak suggests a larger uncertainty on the mean heavy peak mass value. From this comparison and can be seen that the ‘Five-Gaussian method’ will not fit nuclear structure effects, instead predicting only the broader and smoother trends of the chain mass yield.
Figure 6.7: Simulated mass yield using GEF matched to the calculated yield using Five-Gaussian fit method using the least square fit method.
The analysis described here is typical of the evaluation work required of experimental data before it can be accepted into included into a nuclear database. It has been shown that the GEF code is giving a reasonable result for the mass distribution yields. The GEF code is still changing rapidly with updated versions being released on a monthly basis. When completed, the inclusion of this data will be published in JEFF-3.3, which is likely to use GEF as the principle model and a maximum likelihood method to adjust the model outputs.
Chapter 7

Conclusion

This work describes the analysis performed on data collected by STEFF following its first installment at the high flux research reactor at the ILL, Grenoble. The Spec- 
Trometer for Exotic Fission Fragments (STEFF), a 2-velocity, 2-energy spectrometer 
assembled by the Manchester Fission Group was used in PF1B to take measurements 
of prompt gamma-rays from thermal fission of $^{235}$U.

The motivation for this thesis has been to contribute to more accurate knowledge 
of gamma-ray heating in nuclear reactors as listed as a high priority request by the 
NEA [23]. Analysis of the timing, energy and gamma-ray signals have been performed 
and through the procedures discussed in Chapters 3 and 4, a single $^{235}$U energy spec- 
trum for each individual detector has been determined as well as the total energy distri- 
bution. The average total gamma-ray energy and average multiplicity have been 
determined as $8.4 \pm 0.26$ MeV and $7.74 \pm 0.12$, respectively.

To calibrate the energy signals collected by STEFF, the pulse shapes created by 
fission fragments in an ionisation chamber installed at the Lohengrin mass spectrom- 
eter, also at the ILL, have been investigated in Chapter 5. The relationship between 
the pulse shapes and fragment atomic number has been investigated.
A new Parallel Plate Avalanche Counter (PPAC) described in Chapter 2 has been tested for its potential to improve the timing resolution of the current STEFF stop detector. The timing resolution of the new PPAC is found to be 337 ps, a 75% improvement on the previous stop detector.

The evaluation work performed at the NNL, Cumbria, described in Chapter 6, is typical of the processes published experimental data goes through so that it can be included within an evaluated nuclear database. In this work it is the experimental data determined from the spontaneous fission of $^{252}\text{Cf}$ that is evaluated. The techniques described may be applied to the STEFF results so that STEFF data may be included in a future evaluated dataset.

### 7.1 Future work

STEFF has recently been disassembled and moved from the ILL to the neutron time of flight facility, n_TOF at CERN where instead of thermal neutrons, a ‘white’ neutron source, containing a range of neutron energies, from several MeV up to several GeV, will be used to induce fission in $^{235}\text{U}$. This experiment is expected to run in October 2015.

Two extra arms are in the manufacturing process and will be incorporated onto STEFF during this experiment to increase the solid angle available for fission fragment acceptance. These extra arms are Bragg detectors, so further TOF measurements will not be taken.

An improved PPAC stop detector is also in the development stages at Manchester with the intention of measuring faster timing signals, with higher efficiency, therefore improving the overall mass resolution.

The NaI(Tl) array geometry will remain as described in this thesis, however the electronics of the photomultiplier tube will be modified to improve the overall effi-
ciency. Beyond this experiment, it is likely that in future, STEFF will be employed using a $^{239}$Pu target. STEFF remains a versatile and unique spectrometer which will be utilised in future experiments to contribute towards accurate nuclear datasets.
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