Measurement and simulation of prompt fission $\gamma$-rays from the (d,p)-induced fission of $^{241}\text{Pu}$

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    Se hva den lille jenta di har fått til
Abstract

In this thesis, the prompt fission $\gamma$-rays (PFGs) from the fission of $^{241}\text{Pu}^*$ are studied, with the goal of learning more about how the emission of these photons unfold. The PFG characteristics are obtained both experimentally and through model predictions, and the characteristics investigated are the total photon energy per fission $E_{\text{tot}}$, the average photon multiplicity per fission $M_g$, and the average photon energy $E_{g}$. The photon spectra are also found.

The PFGs were measured at the Oslo Cyclotron Laboratory, where a $^{240}\text{Pu}$ target was bombarded with deuterons of 12.5 MeV. OSCAR, the new LaBr$_3$-detector array at OCL, was used for PFG measurements for the first time. The PFGs were selected by gating on $\gamma$-rays that arrived in coincidence with both a proton and a fission fragment. By using the (d,p) reaction, the PFG characteristics can be extracted as a function of compound nucleus $^{241}\text{Pu}^*$ excitation energy $E_x$. In the present work, the range $E_x \in [5.5, 8.5]$ MeV is studied.

Furthermore, model predictions of the $^{240}\text{Pu}(n,f)$ reaction were calculated using the event-by-event fission model FREYA (Fission Reaction Event Yield Algorithm) [1]. As FREYA provides a complete description of fission, where all physical quantities are conserved, comparing its predictions to experimental results can give indications on whether the photon emission process is well understood.

No change in $E_{\text{tot}}$, $M_g$, and $E_g$ was observed as a function of $^{241}\text{Pu}^*$ excitation energy. The measured value for $E_{\text{tot}} \approx 6.5$ MeV is as expected compared to PFG characteristics extracted from other actinides, while $M_g \approx 5.5$ is lower. This discrepancy might result from an insufficient detector response function for photon energies below 0.5 MeV. FREYA calculations reproduce the experimental photon spectrum above this energy. OSCAR has a better time resolution and lower detector threshold compared to the previous photon detector array at OCL, which improve the quality of PFG measurements.

Previous PFG measurements have reported an increase in $E_{\text{tot}}$ as a function of $E_x$ [2]. Several recent experiments [3]–[5], including the present work, could not validate this dependence on compound nucleus excitation energy. This suggests that the current description of photon emission from the fission fragments needs improvement.
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Chapter 1

Introduction

*I solemnly swear that I am up to no good.*

— J.K. Rowling, Harry Potter and the Prisoner of Azkaban

In 2019 we celebrate 80 years since the discovery of fission [6, 7]. For such an old discovery, it might be surprising that aspects of the fission process remain unexplained. Both nuclear reactors and nuclear weapons are relatively old inventions, and knowledge of fission is essential for these applications. How could there still be questions about the fission process?

In basic science, scientists are driven by the urge for always wanting to know more about a given topic. The widths and depths of the topic are explored; inventing applications of the research is often considered convenient, but not essential. This was not the case with the study of fission in the 1940s. Once it was clear that truly frightful amounts of energy could be released in nuclear fission: a million times the energy of any chemical reaction [8, pg. 161], the minds of the scientists immediately sprung to what could be done with so much energy at hand [9]. Thus, the term “nuclear fission” was barely born before the study of it was steered sharply towards applications.

During the Second World War, understanding fission quickly became synonymous with winning the War. Whoever controlled nuclear weapons was thought to dominate the world [10]. This continued through the Cold War when the Superpowers competed to have the most extensive array of nuclear weapons. They put large sums of money into research on fission, focusing on creating better weapons, more effective means of producing fissile materials or how to get the largest energy output from a single device.

With such a focus on the applications of fission, we quickly learned what output fission produces. The question remains: why does fission unfold in this
specific way? Over the last eight decades, pioneers have produced a steady stream of theoretical and experimental work, focusing on both describing the fission process, in addition to improving the application of it. However, as fission is a many-body quantum mechanical problem, it is exceptionally difficult to describe. Fission is therefore still an active field of research [11], where a lot of questions are waiting to be answered.

One of the least studied aspects of fission is the emission of prompt fission $\gamma$-rays (PFGs) [12], which are photons originating from the de-excitation of the highly excited fission fragments. After the first measurements of PFGs were conducted in the 1970s [13, 14], the study of them was latent for four decades. PFGs might not have been regarded as significant in an application point of view, as just 4% of the energy released in fission is given to them [15]. However, while the fission fragments lose their energy over a short distance, $\gamma$-rays can deposit energy far from where they are emitted. It is thus crucial to have good estimates of these prompt fission $\gamma$-rays when designing the next generation of nuclear reactors [16]. Especially important is the fast-neutron region, where the characteristics of the PFGs are the least studied [5]. Therefore, in 2012, there was a call for more precise measurements of PFGs in order to obtain more accurate reactor simulations [15]. Following this call, researches have gathered PFG characteristics on both fissile and non-fissile nuclei, see for example Ref. [17–19].

Besides the reactor application of PFG measurements, information about these photons is a tool for scientists when probing the fission process. PFGs carry information on angular momentum and energy of the fission fragments, and also on the competition between neutron and photon emission in the de-excitation of the fission fragments [4]. Determining the characteristics of these PFGs can thus contribute to assembling a complete picture of the fission process. Furthermore, these experimental measurements can be compared to model predictions, which indicates if various features of fission are described correctly or not. This comparison guides us as we try to put the pieces of the fission puzzle together. Several recent papers have improved our understanding of PFGs, see for example Ref. [12, 20–22].

In order to contribute to the work on the PFGs, this thesis has two aspects: the PFGs from $^{240}\text{Pu}(dp,f)$ reaction have been measured experimentally and been simulated with the event-by-event fission model FREYA [1]. This investigation of the PFG behaviour and the evaluation of FREYA’s ability to reproduce experimental results may provide useful insights as we attempt to describe the fission process.

The layout of this thesis is as follows: after this introduction and motivation, chapter 2 summarizes some main concepts of the fission process. It is followed by chapter 3 on FREYA, where the model is presented and the simulation results are shown and discussed. Chapter 4 and 5 explain how the experimental data was obtained and how the PFG characteristics were extracted. The ex-
Experimental results are discussed in chapter 6 and compared to previous studies of PFGs. Here, the FREYA predictions are also examined in the light of the experimental results. The thesis is concluded with a summary and outlook in chapter 7.
CHAPTER 1. INTRODUCTION
Chapter 2

Nuclear theory: fission

Oh, what idiots we all have been! Oh, but it is wonderful! This is just as it must be! Have you and Lise Meitner written a paper about it?
— Niels Bohr to Otto Frisch about the discovery of fission

This chapter covers the basic properties of nuclear fission, including what fission is and why some nuclei are able to fission. The concept of prompt fission $\gamma$-rays is also introduced, and the experiments that shape our current understanding of these PFGs are recounted. As an introduction, the chapter starts by giving a brief retelling of the discovery of fission.

2.1 The discovery of fission

Following the discovery of the neutron by James Chadwick in 1932 [23], scientists wondered what might happen when known elements are bombarded with this newly discovered, neutral particle. In Rome, Enrico Fermi and his coworkers systematically irradiated elements of increasing atomic numbers, and they learned by identifying the reaction products that the reactions $(n,\alpha)$, $(n,p)$ and $(n,\gamma)$ exist [24]. However, when they started irradiating uranium in 1934, trouble arose when trying to identify the reaction products. Some reaction products could not be recognized as any of the closest neighbours of uranium in the periodic table. Fermi thought these unknown products were so-called transuranic elements [11], elements with a higher atomic number than uranium. He was criticized by Ida Noddack for not investigating if the unknown products could be other known elements from the periodic table. Noddack proposed that uranium might have broken up in larger fragments [25] as cited in Ref. [11], but her proposal was rejected and forgotten [11]. Fermi received the 1938 Nobel Prize in
physics for his work on nuclear reactions, and the discovery of new radioactive elements [20].

Just after the prize was announced, two scientists were able to identify one of the unknown reaction products, which would discredit the transuranic explanation. Otto Von Hahn and Fritz Strassmann were able to prove the existence of barium among the reaction products [6] as cited in Ref. [11], an element with atomic number 56, and thus far out of reach of any transfer reaction on uranium. The somewhat reluctant conclusion was, as Noddack had suggested, that the barium originated from splitting uranium. Right after this discovery, Lise Meitner and her nephew Otto Frisch proposed that the splitting of uranium could be modelled as a liquid drop that vibrated and then separated in two smaller drops [7, 24]. Thus, fission was discovered.

2.2 The fission process

Equation 2.1 shows the semi-empirical binding energy formula. This formula uses the liquid drop model as a basis, as it successfully describes collective features of the nucleus. It also includes basic shell model corrections.

\[
B = a_v A - a_s A^{2/3} - a_C Z (Z - 1) A^{-1/3} - a_{\text{sym}} \frac{(A - 2Z)^2}{A} + \delta
\]  

(2.1)

In the semi-empirical binding energy formula, \(B\) is the binding energy, \(\delta\) is the pairing term, and \(a_v, a_s, a_C, \text{ and } a_{\text{sym}}\) are empirical weighting factors [27, pg. 68]. This equation describes how much binding energy there is in a given nucleus, or in other words, what the difference is between the mass of a nucleus and the mass of the free nucleons. From this equation, we can observe that the binding energy increases with \(A\), while it decreases with \(Z^2\). When adding a proton to a heavy nucleus, several more neutrons must be added in order to keep the nucleus stable, which indicates that there may be a choice of \(A\) and \(Z\) which maximizes the binding energy. This is easily observed when plotting the average binding energy per nucleon, shown in figure 2.1 For \(A = 56\) the binding energy per nucleon peaks, and then starts to decrease for increasing \(A\).

The principle behind fission is that elements situated to the far right of the peak of \(A = 56\) release binding energy when they split into two fission fragments, as the average binding energy per nucleon increases. The two resulting fission fragments are more tightly bound than the original nucleus. This is why Haan and Strassmann discovered barium in the irradiated uranium sample: the uranium had divided into an isotope of barium [7], along with another, lighter fission fragment.

Figure 2.2 illustrates the fission process, where an unstable nucleus splits into two lighter, tighter bound nuclei. The difference in binding energy is released, and the fission fragments are driven apart due to their Coulomb repulsion [27].
Figure 2.1: Average binding energy per nucleon $B/A$, as a function of mass number $A$. Figure adapted from Ref. [27 pg. 67].
Sometimes a third, light particle is also emitted along with the fission fragments, in a process referred to as ternary fission [28, pg. 304-305].

As the fission fragments are neutron-rich [11], they will immediately after fission emit neutrons and $\gamma$-rays in order to de-excite. These are referred to as prompt fission neutrons (PFN) and prompt fission $\gamma$-rays (PFG) respectively. Most of the energy released in fission is carried away as kinetic energy of fission fragments, but a portion also goes to this neutron and $\gamma$-ray emission [27, pg. 491-492]. If the fission fragment undergoes $\beta$-decay, the radiation following is referred to as $\beta$-delayed neutron and $\gamma$-ray radiation. [28, pg. 535].

![Illustration of the fission process. The unstable, heavy nucleus splits into two fission fragments, and the fragments emit prompt neutrons (PFNs) and prompt $\gamma$-rays (PFGs).](image)

Figure 2.2: Illustration of the fission process. The unstable, heavy nucleus splits into two fission fragments, and the fragments emit prompt neutrons (PFNs) and prompt $\gamma$-rays (PFGs).

### 2.2.1 The fission barrier

By looking at the binding energy curve in figure 2.1, one observes that plenty of elements are energetically allowed to fission. However, only a handful of them have spontaneous fission as their main decay mode, and these elements are generally heavy elements far from the valley of stability. This can be understood, as Meitner and Frisch explained in Ref. [7], by looking at the nucleus as a charged, liquid drop. In order to split, the nucleus must undergo a change of shape. Here, the repulsive force of the Coulomb interaction competes with the restoring force of the surface energy, which is proportional to the surface area of the nucleus [28, pg. 8]. For most intermediate-mass nuclei, the surface term dominates, and the nucleus does not want to change its shape. Plenty of energy must be added in order for these nuclei to fission, and we say that they have a large fission barrier, $B_f$. For some heavy nuclei, the fission barrier is small, which opens up for the possibility of the nucleus tunnelling through it. These nuclei can thus undergo spontaneous fission. Other nuclei, again, have a fission barrier that is somewhere in between. Their fission barriers are too large for the nucleus to tunnel through by itself, but only a bit of extra energy is required to
overcome it. Such a nudge might come from the absorption of a particle or a photon. When the nucleus is provided with the extra energy it needs to fission through a nuclear reaction, it is referred to as induced fission [27, pg. 481].

From the liquid drop model, it might seem like the nucleus is spherical and the fission barrier is smooth, like the one marked with a dashed line in figure 2.3. However, the liquid drop model is not a sufficient description of fission [28, pg. 10]. When including shell effects, it turns out that deformed shapes can be more stable than spherical ones [29, pg. 390]. This is particularly evident for heavy nuclei. Such a stable deformation-configuration is found when the nucleus is halfway to fissioning. The nucleus reaches a deformation where it is more stable, and it can thus linger in this stretched shape. We say that the nucleus is superdeformed [30]. This results in the fission barrier taking the double-humped shape drawn in a full line in figure 2.3. When a heavy nucleus is excited from its ground state, it can pass the first hump in the barrier, and exist as a superdeformed fission isomer in the second potential well. From here, it can either pass the second barrier and fission or travel back to the first potential well. The two humps might have different heights [31], and they are referred to here as $B_{f,A}$ and $B_{f,B}$ for the inner and outer hump respectively.

The two-dimensional, double-humped fission barrier is a useful model, but it has some shortcomings. For example, it cannot explain the asymmetric mass distribution of the fission fragments. Therefore, fission is often illustrated as the compound nucleus traveling on a potential energy surface. The surface is a function of nuclear deformations [32, pg. 147], and some paths through this landscape are more favourable than others.
2.2.2 Induced fission

As explained above, when the nucleus by itself cannot overcome the fission barrier, fission can be induced by supplying the extra energy needed. If the nucleus absorbs a photon and then fissions, the ($\gamma$,f) process is called photofission [28, pg. 104-105]. Fission can also be neutron-induced, where the target nucleus absorbs a neutron. The neutron separation energy $S_n$ is added to the system along with the kinetic energy of the neutron, $E_n$, which can push the nucleus over the fission barrier. As the neutrons released in fission further can induce fission, this can create a chain reaction [27, pg. 501]. It is this property that is employed in nuclear reactors. In neutron-induced fission, it is not the target nucleus $^A_Z$ that fissions, but rather the reaction product $^{A+1}_Z^*$. The star emphasizes that the compound nucleus is in an excited state, with a compound nucleus excitation energy $E_x$. Thus, in the reaction $^{239}\text{Pu}(n,f)$, it is $^{240}\text{Pu}^*$ that fissions, which is the notation used in this thesis.

2.3 Prompt fission $\gamma$-rays

This thesis aims to extract the prompt fission $\gamma$-rays from the fission of $^{241}\text{Pu}^*$, and chapter 1 explains our motivation for measuring them. The PFGs are characterised by the average PFG multiplicity per fission $M_g$, the total PFG energy per fission, $E_{\text{tot}}$, and the shape of the photon spectrum [15]. The average PFG energy $E_g = E_{\text{tot}}/M_g$ is also given [13-14].

In section 2.2, it is described how prompt neutrons and $\gamma$-rays are emitted when the fission fragments de-excite. How much energy the photons are given thus heavily depends on the competition between neutron and photon emission in the fission fragments. In the simplest model, the fission fragments send out neutrons as long as they are energetically allowed to do so, that is, their excitation energy is larger than the neutron separation energy $S_n$. This would leave the PFGs with an $E_{\text{tot}}$ of approximately one $S_n$ on average [28, pg. 531]. If the excitation energy of the fission fragments increased, the neutrons would get the extra energy. In this model, the upper limit of $E_{\text{tot}}$ is thus the average $S_n$ among the fission products. As the $E_{\text{tot}}$ measured is higher than $S_n$ [15], this is considered proof that there is competition between neutron and photon emission above $S_n$ [33] [54]. However, how this competition changes as a function compound nucleus $E_x$ has not yet been fully studied [15]. We can thus obtain useful information on the fission process when comparing measured PFG characteristics as a function of $E_x$ to results from fission simulation codes. Still, most of the recent measurements of the PFGs through the (n,f) reaction are done with a single or a few incident neutron energies [5] [55]. The result is that several experiments must be conducted for a given nucleus to get a picture of how the PFGs depend on $E_x$. 
2.3.1 Current understanding of PFG emission

One of the most extensive studies of PFGs behaviour as a function of compound nucleus excitation energy $E_x$, was conducted decades ago. Frehaut et al. in Ref. [2] studied $E_{\text{tot}}$ from the $^{232}\text{Th}$, $^{235}\text{U}$, $^{237}\text{Np}(n,f)$ reactions up to $E_n = 15$ MeV. The results for $^{237}\text{Np}(n,f)$ are shown in figure [2.4] where the increase of $E_{\text{tot}}$ with $E_n$ is evident. The resulting dependence reported showed similar characteristics for all three nuclei. The plateau observed at $E_n \approx 6-7$ MeV was interpreted as the onset of second-chance fission, where the energy available in the system decreases.

Nifenecker et al. in Ref. [33] studied the PFGs from the spontaneous fission of $^{252}\text{Cf}$. They thus looked at $E_{\text{tot}}$ as a function of the fission fragment excitation energy, which yields a mass dependence of the PFG characteristics which is not present in the study by Frehaut et al.

Both studies noted a positive, linear dependence of $E_{\text{tot}}$ on the average neutron multiplicity $\bar{\nu}$. This is a clear indication of the competition between neutron and $\gamma$-emission, as not all the excess energy is given to the neutrons. Note that Frehaut et al. studied $E_{\text{tot}}$ as a function of compound nucleus excitation energy, while Nifenecker et al. studied $E_{\text{tot}}$ as a function of fission fragment excitation energy. Both studies suggested that the increase in $E_{\text{tot}}$ is due to the angular momentum of the fragments increasing with their excitation energy. As the neutrons carry almost no angular momentum out of the system [28, pg. 532], the competition with $\gamma$-emission thus becomes important. The book “The Nuclear Fission Process” by C. Wagemans [28] gave this description of PFG emission as the latest understanding of the process.

At this point, it might seem like we understand the process of PFG emission from the fission fragments. However, Frehaut re-examined the experimental data some years later, and new thoughts were presented in Ref. [34]. He observed that the increase in $E_{\text{tot}}$ was due to each photon carrying more energy, as the photon multiplicity barely changed. This is not in agreement with the explanation that the total angular momentum in the fragments has increased, as more angular momentum in the fission fragments is expected to give a larger number of emitted $\gamma$-rays. Still, this does not contradict the Nifenecker et al. results. In Nifenecker et al., the total energy in the fissioning system was constant, as they studied spontaneous fission. Thus the average angular momentum among the fragments was also constant. Their conclusion was that increased fragment excitation energy gives larger angular momentum of that fragment. In contrast, the study by Frehaut in Ref. [34] stated that they found no increase in the average angular momentum as the excitation energy of the fissioning nucleus increased. Therefore, the two studies did not contradict each other, but the description of the PFG emission given in C. Wagemans [28, pg. 531-532] no longer fitted with Frehaut’s conclusions in Ref. [34]. Still, after the paper by Frehaut was published in 1989, few studies on the competition between neutron and photon emission were conducted. The re-examination of the data by Frehaut
Figure 2.4: Measurements of total PFG energy per fission $E_{\text{tot}}$ and average neutron multiplicity $\bar{\nu}$ from the reaction $^{237}\text{Np}(n,f)$ by Frehaut et al. [2]. The average neutron multiplicity $\bar{\nu}$ is marked in pink, and uses the left $y$-axis. The total photon energy $E_{\text{tot}}$ is marked in blue, uses the right $y$-axis, and is given relative to $E_{\text{tot}}$ from $^{252}\text{Cf}(sf)$, where $E_{\text{tot}}(^{252}\text{Cf}(sf)) = 7.01$ MeV. Figure adapted from Ref. [34].
might have been largely unnoticed. If the description of PFG emission summarized by Wagenmans was given too much gravity, the question on neutron-photon competition could have been considered answered. This can explain why the Frehaut et al. experiments in Ref. [2] were not further revisited.

New results have added to the suspicion that the PFG emission from fission fragments is not well understood. As seen in figure 2.4, Frehaut et al. reported an increase of \( E_{\text{tot}} \) with \( E_x \). Recent studies could not validate this behaviour. In Ref. [3], Rose et al. found no apparent increase in \( E_{\text{tot}} \) with \( E_x \), and their results fitted well with model calculations using constant angular momentum of the fission fragments. This is supported by Lebois et al. in Ref. [4], where the PFGs emitted from both thermal and fast neutron-induced fission of \( ^{236}\text{U}^* \) were compared, and the conclusion was that the extra energy does not contribute significantly to the photon emission. Furthermore, Qi et al. measured PFGs from the reaction \( ^{238}\text{U}(n,f) \) using the two incoming neutron energies \( E_n = 1.9 \) and 4.8 MeV, and observed no significant dependence of the PFG characteristics on incident neutron energy. All three studies are well below the threshold for second-chance fission, and thus cannot be measurements of the plateau in \( E_{\text{tot}} \) shown in figure 2.4.

No direct comparisons between the results of Rose et al., Qi et al. and Frehaut et al. can be made, as different nuclei were studied, but rough estimates give that Frehaut et al. observed an increase in \( E_{\text{tot}} \) of 70-100 keV per MeV added excitation energy of the compound nucleus. In Rose et al., PFGs from \( ^{234}\text{U}^* \) and \( ^{240}\text{Pu}^* \) were studied over a range of 2.5-3 MeV excitation energy above the fission barrier. Though no increase in \( E_{\text{tot}} \) with \( E_x \) was found in Rose et al., larger excitation energy regions should be studied before dismissing the possibility of an increase. Furthermore, Qi et al. reported an \( E_{\text{tot}} \) of 5.25 ± 0.20 and 6.18 ± 0.65 for \( E_n = 1.9 \) and 4.8 MeV respectively, where no conclusions of an increase could be drawn due to few data points and significant error bars. In addition to this, Rose et al. in Ref. [3] reported a constant average \( \gamma \)-ray energy, contrary to what was reported in Ref. [34]. Lebois et al. did not state values for \( E_{\text{tot}} \) for the thermal and fast neutron-induced fission of \( ^{236}\text{U}^* \), and therefore their results cannot be compared to the values of Frehaut et al.

In order to get a better understanding of the de-excitation of the fission fragments, a handful of studies have conducted simultaneous measurements of the prompt photons and neutrons from fission fragments. These studies mostly look at \( ^{252}\text{Cf}(sf) \), yielding the same mass dependence as in Nifenecker et al. However, the conclusions drawn by these studies of \( ^{252}\text{Cf}(sf) \) differ significantly. Marcath et al. [36] summarized the varying conclusions of the neutron-photon competition studies:

“One [33] shows a positive correlation, another observes a complex fragment-dependent correlation [37], a third reports a negative correlation [38], while a fourth found no evidence of correlated emission from specific fragment pairs [39].”
Marcath et al. themselves observes a weak negative neutron-photon competition.

Based on these results, it is safe to say that we do not fully understand the competition between neutron and photon emission from the fission fragments. Both the discrepancies between the observations of old and new experiments of the PFG behavior as a function of $E_x$, and the disagreement between different studies of $^{252}$Cf(sf) leave the theoreticians blindfolded. It is unclear what experimental data a de-excitation model should reproduce, and it is essential that these discrepancies are sorted out in order for a proper model for neutron-photon-competition in the fission fragments to be established. New experimental data, where more nuclei are examined, a wider range of $E_x$s are studied and where older experiments are reconstructed, will guide us as we try to understand how the PFG emission from the fission fragments unfold.

2.4 Using (d,pf) as a surrogate reaction for (n,f)

For measuring the PFGs, the (n,f) reaction is most commonly employed, as this is the one taking place in nuclear reactors [28, pg. 64]. However, monochromatic neutron beams are difficult to produce, and cannot span a large energy range as easily as a charged particle beam. Therefore, the (d,pf) reaction was suggested as a surrogate to the (n,f) as the same compound nucleus is produced in both reactions. The (d,pf) reaction also has the advantage that, when a particle detector is present in addition to the fission fragment and $\gamma$ detectors, the excitation energy $E_x$ of the compound nucleus can be calculated. Thus the PFG characteristics can be obtained as a function of the compound nucleus excitation energy. The (d,pf) reaction is also useful for studying nuclei that have neutron separation energies $S_n$ larger than their fission barrier $B_f$ [28, pg. 205]. As the proton carries energy out of the system, it might leave the compound nucleus in a state where its excitation energy $E_x$ is above $B_f$, but below $S_n$. This opens for examining fission barriers that cannot be studied through neutron-induced fission.

A drawback with this surrogate reaction is that one must assume that the same compound nucleus is produced through both the (d,pf) and (n,f) reactions. This assumption is not completely valid, as quantities like the angular momentum transfer are different in the (d,pf) reaction compared to (n,f) [28, pg. 200]. The question is whether the assumption is approximately valid, and to what degree one can say that the fission observables produced in the two reactions are identical. This can be studied by comparing data measured from both the (n,f) and (d,pf) reactions.

Therefore, PFG characteristics obtained from the (n,f) and (d,pf) reactions should be compared to understand if two reactions are equivalent. However, the experimental data for conducting such a comparison is limited. As no experi-
To our knowledge, the only work that has previously extracted PFG characteristics from the (d, pf) reaction were Rose et al. in Ref. [3]. The data for comparing the (n, f) and (d, pf) reactions are therefore limited to the cases presented in the Rose et al. article, which are the $^{239}\text{Pu}(d, pf)$ and $^{233}\text{U}(d, pf)$ reactions. For the study of $^{239}\text{Pu}(d, pf)$, they observed an excess of PFGs compared to those measured from the (n, f) reaction, presented in Ref. [14], a deviation attributed to the larger angular momentum in the (d, pf) reaction. This is supported by calculations done by the fission simulation code GEF [40]. Still, Rose et al. struggled with both the bad time resolution of the sodium-iodine-detector array CACTUS and a rather high detector threshold at $E_\gamma = 450$ keV. This resulted in both the measured photon spectrum having to be corrected for the assumed neutron contribution, as well as assumptions had to be made for the PFGs with energies below 450 keV. Therefore, more measurements of PFGs from (d, pf) experiments should be conducted where (n, f) data also is available, in order to get a better understanding of the impact on the PFGs of deuteron versus neutron-induced fission.
Chapter 3

FREYA

Remember that all models are wrong; the practical question is how wrong do they have to be to not be useful.
— George Box

FREYA, an acronym for Fission Reaction Event Yield Algorithm, is a Monte-Carlo based computer code that generates nuclear fission events. In the FREYA code, the energy, linear momentum and angular momentum are conserved in a fission event. All the kinematic information on both the fission fragments and the emitted particles are available, and it is thus possible to look at different aspects of the fission process using the same model. FREYA needs experimental data for the fission fragment mass and kinetic energy distributions as input, and it can model spontaneous fission, photofission and neutron-induced fission of some nuclei, see table 3.1.

In this chapter, the simulation of the prompt fission $\gamma$-rays (PFGs) from the (d,p)-induced fission of $^{241}$Pu$^*$ is presented. The chapter begins by giving an overview of the fission events in FREYA in section 3.1 focusing on the simulation of photon emission. Furthermore, the approach to and execution of the simulation is presented in section 3.2 and the results are presented and discussed in sections 3.3 and 3.4.

3.1 Fission events

The simulation of a fission event in FREYA proceeds in several steps: pre-fission, during fission and post-fission. In this section, a general overview of the fission process in FREYA is given. As this work focuses on the prompt fission $\gamma$-rays that FREYA simulates, a thorough description of the photon emission is also included.
Table 3.1: The fissionable isotopes and their respective fission reactions included in FREYA. The maximum incoming neutron energy for neutron-induced fission is $E_n = 20$ MeV \[41\]. Also note that for neutron-induced fission, the target isotope is listed, and not the fissioning isotope.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Neutron-induced (n,f)</th>
<th>Spontaneous (sf)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{233}\text{U}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>X</td>
<td></td>
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<tr>
<td>$^{239}\text{Pu}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{242}\text{Pu}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{244}\text{Cm}$</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>$^{252}\text{Cf}$</td>
<td>X</td>
<td></td>
</tr>
</tbody>
</table>

FREYA only considers fission events, and it does not take into account the $\beta$-decay of the fission products. First, FREYA considers the pre-equilibrium nucleus, and the chance of a neutron being emitted before the nucleus reaches equilibrium. This is called pre-equilibrium neutron emission and is only significant for high energies \[41\]. For details of how pre-equilibrium neutron emission is treated, see Ref \[42\]. Next, when the compound nucleus is in equilibrium, the code examines the competition between pre-fission neutron evaporation and fission. The code evaluates the ratio between the radiative widths of neutron emission and fission, using the method presented in Ref. \[43\]. If a neutron is emitted, the resulting $(A-1)$-nucleus can either fission or emit another neutron. As each resulting nucleus can either emit a pre-fission neutron or undergo fission, this process is called multichance fission. Pre-fission neutron evaporation is possible as long as the excitation energy of the nucleus is above the neutron separation energy $S_n$. However, if the resulting excitation energy of the daughter nucleus is below the fission barrier $B_f$, then the event is abandoned by FREYA as prompt fission cannot occur.

After pre-fission radiation, FREYA proceeds to the fission process. FREYA considers only binary fission, where the compound nucleus splits into two fragments, one lighter and one heavier. Ternary fission, where a third light particle is emitted along with the fission fragments \[28\] pg. 304-305, is thus disregarded. Using an energy-dependent fission fragment mass distribution that is based on experimental data, the mass numbers of the two fragments are selected.
The charge is then split in accordance with a Gaussian probability distribution, where the peak corresponds to the fragments having the same charge-to-mass ratio as the compound nucleus \[41\]. The total mass number \( A \) and charge \( Z \) are conserved in the fission simulation.

The next step is to determine the energies of the fission fragments. FREYA relies on experimental data as input for the mass dependence of the average total kinetic energy \( \text{TKE}(A) \) of the fission fragments, for a given compound nucleus excitation energy \( E_x \) \[41\]. By calculating the Q-value of the fission reaction and subtracting \( \text{TKE}(A) \), the energy available for excitation of the two fragments is found. This energy is split between rotational \( E_{\text{rot}} \) and statistical \( E_{\text{stat}} \) excitation energy. \( E_{\text{rot}} \) is due to the total angular momentum \( J \) of each fragment and is given by \[27\] pg. 144):

\[
E_{\text{rot}} = \frac{\hbar^2 J(J+1)}{\mathcal{J}},
\]

where \( \mathcal{J} \) is the moment of inertia. The rest of the nuclear excitation energy is assigned to \( E_{\text{stat}} \). The excitation energy is then shared between the two fission fragments. First, a tentative energy division is made based on the two heat capacities of the fragments. In order to achieve a better reproduction of the mass-dependent neutron multiplicity, the energy is thereafter shifted slightly in favour of the light fragment. The total energy of the system is conserved.

The last step in the generation of a fission event in FREYA is the simulation of the post-fission radiation, which consists of neutron and photon emission from the fission fragments. First, the fission fragments evaporate neutrons until their statistical excitation energy is below the neutron separation energy \( S_n \) and neutron emission is no longer possible. Even though the process is known to be more complex, as discussed in chapter \[2\] this simplified de-excitation model gives a rapid simulation and is therefore employed.

### 3.1.1 Photon emission

When neutron emission is no longer possible, the photon emission starts as a statistical cascade of photons. Figure \[3.1\] shows how FREYA models photon emission. The nucleus starts with the statistical excitation energy \( E_{\text{stat}} \) and the rotation excitation energy \( E_{\text{rot}} \), which is left after neutron emission.

First, the statistical excitation energy \( E_{\text{stat}} \) of the product nucleus is disposed of by a \( \gamma \)-cascade down to the yrast line. The \( \gamma \)-rays are sampled from a black-body spectrum, modified by a giant dipole resonance form factor, and are emitted isotropically in the rest frame of the nucleus \[44\]. It is assumed that this radiation is purely \( E1 \) and \( M1 \) radiation, such that each photon removes \( 1\hbar \) of angular momentum from the nucleus \[41\]. This is illustrated in figure \[3.1\] by the orange arrows, each representing a photon being emitted in a statistical cascade.
CHAPTER 3. FREYA

Figure 3.1: Visual representation of photon emission from fission fragments in FREYA. The $x$-axis is the total angular momentum of the nucleus, $J$, and the $y$-axis is the excitation energy of the nucleus.

When the statistical excitation energy $E_{\text{stat}}$ drops below a given energy limit $E = g_{\text{min}}$ as illustrated in figure 3.1, $E_{\text{stat}}$ is considered depleted. The emission of photons then continues by exhausting the rotational energy $E_{\text{rot}}$. These $\gamma$-rays are referred to as collective $\gamma$-rays. The photon emission follows the yrast line down through $E/2$ photon emissions [45]. The energy of each photon is calculated as $E = E_{\text{rot}}(J) - E_{\text{rot}}(J-2)$ from equation [3.1] until the total angular momentum drops below $2h$ [45]. Finally, when $J < 2$, the remaining energy is given to a single $\gamma$-ray [44].

For some product nuclei, information on the lowest-lying states in the nucleus is either available in the RIPL-3 library [46], or can be constructed [20]. For these nuclei, FREYA uses these for discrete photon transitions whenever the excitation energy is within the RIPL-3 range. This is continued until either the excitation energy is below $g_{\text{min}}$, or the half-life of the state is longer than a given time $t > t_{\text{max}}$ [44]. The parameters $g_{\text{min}}$ and $t_{\text{max}}$ reflect detector properties, and should be chosen in accordance with the time resolution and energy threshold of the detectors.
3.2 Simulation of $^{240}\text{Pu}(d,pf)$

3.2.1 Simulation approach and assumptions

FREYA simulates fission of a limited number of isotopes, and three types of fission: spontaneous fission (sf), photofission ($\gamma,f$) and neutron-induced fission (n,f) [41]. To compare FREYA simulations with the experimental data from the $^{240}\text{Pu}(d, pf)$ experiment, we must assume that Bohr’s hypothesis of compound nuclei [47] holds. The hypothesis says that how the compound, excited nucleus $^{241}\text{Pu}^*$ is formed is of no consequence for how it decays. We must also assume that compound nuclei produced through the two reactions are identical, which might not be true, due to the higher angular momentum transfer in (d,pf) reactions. For a discussion of the differences between the (n,f) and (d,pf) reactions, see section 2.4. Based on these assumptions, we can simulate the reaction $^{240}\text{Pu}(n,f)$ in FREYA, as both the (n,f) and the (d,pf) reactions create the compound nucleus $^{241}\text{Pu}^*$.

The neutron separation energy $S_n$ for $^{241}\text{Pu}$ is 5.2 MeV [48]. The lowest compound nucleus excitation we can get through simulating $^{240}\text{Pu}(n,f)$, is when the neutron carries no kinetic energy, and the compound nucleus excitation energy $E_x$ then is equal to $S_n$. The height of the double-humped fission barrier of $^{241}\text{Pu}$ is larger than $S_n$, 6.1 and 5.4 MeV for the first and second barriers respectively [31]. When the neutron carries little kinetic energy, the compound nucleus is then left with an $E_x$ below the fission barrier. This should in principle not create fission events in FREYA, as tunnelling is not considered. In this simulation, FREYA is forced to fission with the incoming neutron energies $E_x = 0, 0.5$ MeV. This is convenient when comparing to experimental data, as events where the nucleus tunnels through the fission barrier are possible.

3.2.2 Implementation of fission of $^{241}\text{Pu}$

From recent publications [41][44], one can see that fission of $^{241}\text{Pu}$ is not covered in version 2.0.3 of FREYA. For the use in this thesis, the $^{240}\text{Pu}(n,f)$ reaction had to be implemented.

For modelling fission, FREYA needs the fragment mass and kinetic energy distributions [41], which are usually obtained from experimental data. In the $^{241}\text{Pu}$-case, no experimental data is yet available, and phenomenological models were therefore employed.

To obtain a fission fragment mass distribution $Y(A)$ and the kinetic energy of the fragments $\text{TKE}(A)$, the fission modelling code GEF (General Fission model) [40] was used. It was assumed that the energy dependence of these mass distributions is the same as for the $^{239}\text{Pu}(n,f)$ and $^{241}\text{Pu}(n,f)$ reactions already implemented in FREYA. A five-Gaussian fit to the mass distribution was

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1A great thank you to Ramona Vogt and Jørgen Randrup for implementing this!
conducted, and the energy dependence was introduced in the fit parameters. The details of the fit are described in Ref. [42].

The parameters used in the FREYA simulation, which are described in Ref. [41], were set to the same values as in the $^{239}\text{Pu}(n,f)$ case. The parameter describing the shift of the total kinetic energy, $d\text{TKE}$, was chosen such that the calculated average neutron multiplicity reproduced the ENDF evaluation [49].

### 3.2.3 Modified FREYA 2.0.3

We base the simulations shown in this thesis on a modified version 2.0.3 of FREYA. The original code is available for download at Ref. [1], while the modified version can be inspected at Ref. [50]. Here follows a brief overview of the code and the changes done to it.

Figure 3.2 gives an overview of the FREYA code structure. The user inputs the fissioning nucleus, along with the fission type (spontaneous, neutron-induced or photofission), the energy of the reaction, and the number of fission events to be simulated. FREYA then collects the needed input data of the fissioning nucleus from a collection of information in the folder “data FREYA”, and finally writes generated fission events to an output file. The output file has one entry per fission event and includes information about the fission products and the emitted neutrons and photons. It must be analysed in order to extract the fission characteristics.

The necessary data files for the simulation of neutron-induced fission of
$^{241}$Pu, named $\text{Pu241.xs}$, $\text{Pu241.PreEq}$ and $\text{Pu241.TKE-Af}$ were added to the “data FREYA” directory. In the files $\text{react.dat}$ and $\text{inputparameters.dat}$ from the same folder, a line was added for photon- and neutron-induced fission of $^{241}$Pu, where the physical parameters used in the simulation are specified. The value of the time parameter $t_{\text{max}}$ is also specified in the fission routine. The meaning of this parameter is explained in section 3.2.4. No further changes were implemented in FREYA version 2.0.3.

3.2.4 Choice of $g_{\text{min}}$ and $t_{\text{max}}$

In section 3.1.1 it is explained how the energies of the $\gamma$-rays and the allowed half-life of states simulated in FREYA are constricted by the two detector parameters $g_{\text{min}}$ and $t_{\text{max}}$. Photons with lower energies than $g_{\text{min}}$ or that come from states with half-lives longer than $t_{\text{max}}$, are not considered. These values can be specified by the user and should be chosen to reflect the experimental setup. For simulating the Oslo experiment, the parameters were chosen in accordance with the photon energy range considered and the specified time limit of the observed $\gamma$-rays. The parameter $g_{\text{min}}$ is specified in the file $\text{inputparameters.dat}$, while $t_{\text{max}}$ is found in the FREYA file $\text{msFREYA\_decayS.F90}$. The photon energy range included in the experiment was, as discussed in section 5.1.3, 122 keV to 10 MeV. Thus the limit $g_{\text{min}}$ was 122 keV. Furthermore, in the experiment, we wished to only include $\gamma$-rays that were considered prompt. What prompt means from the experimental point of view is explained in section 4.5.1. In our experiment, the time limit for arrivals of PFG’s was set to $\pm 3$ ns, see section 4.5.2 for the arguments for this choice. The time gate in the experiment is the time after scission, while $t_{\text{max}}$ is the lifetime of states in the fission fragments. This yields that the upper limit of $t_{\text{max}}$ is 3 ns, and $t_{\text{max}} = 3$ ns was chosen in the simulation. For a thorough investigation of how the total photon energy $E_{\text{tot}}$ and the average photon multiplicity $M_{g}$ is affected by the choice of $g_{\text{min}}$ and $t_{\text{max}}$, see Ref. [20].

3.2.5 Simulation

The reaction $^{240}\text{Pu}(d,pf)$ was simulated in FREYA for a range of incoming neutron energies $E_{n} \in [0, 7.25]$ MeV, with $10^6$ events per run. The output files were analysed with the script $\text{freya\_root\_analysr.C}$, available at Ref. [50].

3.2.6 Uncertainty

As there is no experimental uncertainty in FREYA, the sources of uncertainties are:

- FREYA model uncertainty
- Input parameter uncertainty
• Statistical uncertainty

The FREYA output values were calculated from $10^6$ events per run, which renders the statistical uncertainty insignificant. The main sources of uncertainties are therefore the model and input parameter uncertainties, both of which are out of the scope of this thesis to calculate. As estimates of the statistical uncertainty would not have provided an understanding of how large the true uncertainties are, it was decided to give the FREYA simulation results with no uncertainty attached. Calculations of the model uncertainty in FREYA related to the input parameter uncertainty are discussed in Ref. [51].

3.3 Results

The results of the simulation of the PFGs from neutron-induced fission reaction $^{240}\text{Pu}(n,f)$ are presented. Figure 3.3 shows the average photon multiplicity per fission $M_g$, figure 3.4 shows the total $\gamma$ energy released per fission $E_{tot}$, and figure 3.5 shows the average $\gamma$-ray energy $E_g$. The figures show the calculated PFG characteristics, as well as the contributions from first- and second-chance fission. In figure 3.6, the fractions of first-, second- and third-chance fissions are shown. All figures are plotted as a function of compound nucleus $^{241}\text{Pu}^*$ excitation energy $E_x = S_n + E_n$.

3.4 Discussion

From figures 3.3, 3.4 and 3.5, we see that the calculated PFG characteristics $M_g$, $E_{tot}$ and $E_g$ marked as “total” increase slightly with increasing $E_x$ in the region $E_x \in [5, 11]$ MeV. As more energy is available in the compound nucleus, more energy is given to the fission fragments. If a fission fragment is given enough excitation energy $E_x$ so that $E_x > S_n$, another neutron is evaporated. The bulk of the extra excitation energy available goes thus to emitting more neutrons with higher energies, which can be seen from figures A.2 and A.3 in appendix A. This is as expected from the simplified de-excitation model used by FREYA. Photon emission begins largely only after $E_x$ has fallen below $S_n$, and is why the change in $E_{tot}$ and $M_g$ as a function of $E_x$ generally is small.

To study the behaviour of the PFGs as a function of excitation energy of the compound nucleus, we must know if the initial de-excitation happens in the same fission fragments. It is known that the fission fragment mass distribution changes as a function of $E_x$ [28, pg. 228]. For a change in this distribution, the neutrons would be emitted from different primary fragments, which would affect the PFG characteristics. As seen in figure A.1 in appendix A, the average masses of the two fission fragments hardly change in this range of $E_x$. This means that the de-excitation through prompt neutron emission begins from the same fragments in the excitation energy range we consider. Only at the
Figure 3.3: Average photon multiplicity per fission $M_g$ from the reaction $^{240}$Pu(n,f), calculated as a function of compound nucleus excitation energy $E_x = S_n + E_n$. The same data is shown in the two plots, with different scales on the $y$-axes. The smaller scale shows more clearly the behaviour of $M_g$ with $E_x$, while the larger scale plot is easier to compare to the experimental results in chapter 6. Lines connect the data points to guide the eye. The blue curve shows the calculated $M_g$ for the reaction, while the red and black curves show $M_g$ calculated for first- and second-chance fission separately.
Figure 3.4: Total photon energy per fission $E_{\text{tot}}$ from the reaction $^{240}$Pu(n,f), calculated as a function of compound nucleus excitation energy $E_x = S_n + E_n$. The same data is shown in the two plots, with different scales on the $y$-axes. The smaller scale shows more clearly the behaviour of $E_{\text{tot}}$ with $E_x$, while the larger scale plot is easier to compare to the experimental results in chapter 6. Lines connect the data points to guide the eye. The blue curve shows the calculated $E_{\text{tot}}$ for the reaction, while the red and black curves show $E_{\text{tot}}$ calculated for first- and second-chance fission separately.
Figure 3.5: Average photon energy $E_g$ from the reaction $^{240}{\text{Pu}}(n,f)$, calculated as a function of compound nucleus excitation energy $E_x = S_n + E_n$. The same data is shown in the two plots, with different scales on the $y$-axes. The smaller scale shows more clearly the behaviour of $E_g$ with $E_x$, while the larger scale plot is easier to compare to the experimental results in chapter 6. Lines connect the data points to guide the eye. The blue curve shows the calculated $E_g$ for the reaction, while the red and black curves show $E_g$ calculated for first- and second-chance fission separately.
threshold of second-chance fission around 11 MeV, where a neutron is emitted prior to fission, do we see a change in the fragment distribution of about half a nucleon. Therefore, a changing initial fragment mass distribution has little impact on the $M_g$, $E_{\text{tot}}$ and $E_g$ calculated here.

Even if the increase in $E_{\text{tot}}$ with $E_x$ is small, it is visible. This is partly explained by the fact that $S_n$ is not one fixed value. As explained in section 2.3 where the simplified de-excitation model is presented, the upper limit for how much energy can be given to the PFGs from one fission product is the $S_n$ of that nucleus. The fission products, which are the fission fragments after the neutron emission has ceased, have a range of different $S_n$s. As $S_n$ is different in each product nucleus, the average limit on $E_{\text{tot}}$ is thus the average $S_n$ among the fission products. When $E_x$ increases, more neutrons are emitted on average. The result is that the fission product distribution changes and thus the average $S_n$ changes as well. As the product nuclei are then less neutron-rich with increasing $E_x$, the $S_n$ values tend to increase, and this is observed in the FREYA simulations presented here. This can partly account for the increase in $E_{\text{tot}}$ as a function of $E_x$.

Furthermore, the calculated $E_{\text{tot}}$ exceeds the average $S_n \approx 6$ MeV by about 1 MeV, where the weighted average $S_n$ was calculated using the FREYA tabulated masses. This violates the expectation of the model presented in section

Figure 3.6: Share of first-, second- and third-chance fissions from the reaction $^{240}\text{Pu}(n,f)$, plotted as a function of $E_x = S_n + E_n$. 

![Graph showing share of first-, second- and third-chance fissions](image-url)
where it is stated that $S_n$ is the upper limit on $E_{\text{tot}}$ in this simplified de-excitation model. This is explained by the FREYA de-excitation model being more refined than the simplified picture presented in section 2.3. With an increase in compound nucleus excitation energy, the fission fragments gain more angular momentum. As the neutrons do not carry away any significant angular momentum in FREYA, the angular momentum of the nucleus is practically conserved during neutron emission. The rotational energy is thus reserved for the photons, which is why $E_{\text{tot}}$ is larger than the average $S_n$ among the fission products. As the angular momentum increases with $E_x$, this also contributes to an increase in $E_{\text{tot}}$ as a function of $E_x$. Note that this is the same de-excitation model that was proposed in Ref. [2] and [33], which recent studies have challenged, see section 2.3.1.

When looking at the energy that the neutrons carry out of the system in the region $E_x \in [6, 9]$ MeV, we observe that the total energy given to the neutrons increases more than the increase in $E_x$. In order for energy to be conserved, the average kinetic energies of the fission fragments decrease, which can be seen in figure A.4 in appendix A. This decrease in fission fragment kinetic energy at higher excitation energies has been observed experimentally for asymmetric fission [28, pg. 365-368].

In the excitation energy region $E_x \in [11, 12]$ MeV, we observe a discontinuity in the plots for the calculated PFG characteristics. To understand why this occurs, we first take a look at figure 3.6 showing the share of first- and second-chance fissions as a function of $E_x$. Below 11 MeV first-chance fission dominates, because the compound nucleus is not excited enough to first emit a neutron and still be excited above the fission barrier $B_f$. The few events marked as second-chance fissions that are observed here originate from pre-equilibrium neutron emission, which is slightly different from second-chance fission as the neutron removed is not equilibrated. As the nucleus is not in equilibrium when the neutron emission takes place, $S_n$ is not the same as for the compound nucleus. After pre-equilibrium neutron emission, FREYA does not check if the excitation energy is above the fission barrier. This is an inconsistency in FREYA and can lead to unphysical fission events being accepted. Second-chance events are thus seen below the threshold in the simulation. At $\approx 11.25$ MeV, the threshold for second-chance fission $E_x = S_n(^{241}\text{Pu}) + B_f(^{240}\text{Pu})$ is passed. Second-chance fission then becomes the dominant process.

In this FREYA simulation of the $^{240}\text{Pu}(n,f)$ reaction, the transition between first- and second-chance fission occurs rapidly, as seen in figure 3.6. In a previous FREYA simulation of multichance fission in the reaction $^{239}\text{Pu}(n,f)$ found in Ref. [42], a more gradual transition between first- and second-chance fission is observed. This difference can be understood from the different relations between the neutron separation energies and the fission barriers in the two cases. In the $^{240}\text{Pu}(n,f)$ case, the neutron separation energy is rather low, while the fission barrier of $^{241}\text{Pu}$ and $^{240}\text{Pu}$ is about the same, at slightly above 6 MeV. The
cross section for $^{241}$Pu to emit a neutron is high compared to the fission cross section, due to the low $S_n$. If the daughter nucleus has $E_x < B_f$ after neutron emission, the event is discarded by FREYA. Thus lots of events are discarded at low excitation energies. Once the threshold for second-chance fission is passed, these events are included, leading to the sudden dominance of second-chance fission. For the $^{239}$Pu(n,f) reaction in Ref. [42], the neutron separation energy is higher. As the fission barrier is about the same as for $^{241}$Pu, this leads to a lower neutron emission probability and a smoother transition between first- and second-chance fission.

We can now understand the sudden change in the calculated $E_{\text{tot}}$, $M_g$ and $E_g$ at $E_x \approx 11.25$ MeV by looking at the PFG characteristics calculated for first- and second-chance fission separately. This is shown in figures 3.3, 3.4 and 3.5 as the red and black curves. In second-chance fission, the values for $E_{\text{tot}}$ and $M_g$ are lower than in first-chance fission, because the fissioning nucleus is less excited. In combination with the sudden dominance of second-chance fission, this leads to the drop in $E_{\text{tot}}$ and $M_g$ at $E_x \approx 11.25$ MeV. The average $\gamma$-ray energy is higher in second-chance fission, and thus $E_g$ abruptly increases. This is because the number of $\gamma$-rays emitted from the statistical cascade decreases, while the number of collective $\gamma$-rays is about constant. As the collective $\gamma$-rays tend to be less energetic compared to the statistical photons, one might think that $E_g$ should drop. However, the multiplicity of the statistical photons drops more than the energy released through statistical photon emission, resulting in an increase in average photon energy per statistical photon, again giving an increase in $E_g$. 
Chapter 4

Experiment and data collection

Remember, kids, the only difference between screwing around and science is writing it down.
— Adam Savage, quoting Alex Johnson

When conducting experiments in nuclear physics, the process of obtaining data can be viewed as a two-step procedure. The first part is the raw data collection: the detectors that have to be calibrated, the electronics, which must connect the detectors to the data acquisition, and the experimental setup, that is optimized for the reaction being studied. In this first step, the raw data is collected and stored, resulting in a bunch of data where only a fraction is of interest for those conducting the experiment. Therefore, in the second part, the raw data is refined, and specific detector events are filtered out from the array of reactions occurring in the target chamber. Thus the second part sorts out the data that will be brought further on to the data analysis. In this chapter this process of conducting the experiment and sorting the data is explained for the $^{240}$Pu(d,p) experiment that ran in Oslo in April 2018.

4.1 Experimental setup

4.1.1 General setup at OCL

The experiment presented in this thesis was conducted at the Oslo Cyclotron Laboratory (OCL), located in the cellar of the Physics building at the University of Oslo. The MC-35 Scanditronix cyclotron can deliver a pulsed beam of protons up to an energy of 35 MeV.
In an experiment at OCL, a suitable beam and beam energy is chosen, based on calculations run beforehand using reaction simulation tools like Qkinz [52]. The properties of Qkinz are explained in section 4.3.1. After the accelerated ions leave the cyclotron, the beam passes several magnetic dipoles $D$, used for bending the beam, and quadrupoles $Q$, used to focus the beam. The beam is transported to the experimental hall where it hits the target. The detector arrays OSCAR and SiRi, placed around the target, are then used to detect outgoing radiation. The layout of the experimental hall at OCL is shown in figure 4.1.

The Oslo Scintillator Array, OSCAR for short, is the name of the new $\gamma$-ray detector array at the Oslo Cyclotron Laboratory. It consists of 30 LaBr$_3$ scintillator detectors, giving the OCL better energy resolution and better timing than the previous setup [54]. The detectors are cylinders, and each measure 3.5 $\times$ 8 inches. The OSCAR-detectors are inorganic cerium activated LaBr$_3$ scintillator detectors. The energy resolution for these detectors is about twice as good as the resolution for NaI(Tl)-detectors, with $\approx 2.8 - 4.0\%$ resolution for 662 keV $\gamma$-rays, compared to $\approx 7.0\%$ for NaI(Tl) [55]. The decay time is also about one-tenth of the decay time of NaI(Tl). As the previous detector setup
at OCL consisted of NaI(Tl)-detectors, this is a major improvement and opens for a lot of new experiments to be performed.

Inside OSCAR, a silicone detector ring named SiRi is placed. SiRi is a ∆E-E detector, which is used to identify light ions and their energies. For an incoming particle, the energy deposited in a thin (130 µm) silicone strip called the ∆E-detector is measured first. The particle is not stopped in the ∆E-detector, but continues into a 1550 µm thick silicone detector called the E-detector, where the rest of the particle’s energy is deposited. The fraction of energy deposited in the ∆E and E detectors is characteristic for a given charged particle. How this fraction can be used to identify ions and their energies, is explained in section 4.3.1. SiRi consists of eight pads placed in a circle, shown in figure 4.2a, each pad containing 8 ∆E-detectors backed by one E-detector, as shown in figure 4.2b. By observing which ∆E-E-detector that fired, we can determine the angle of the outgoing particle.

4.1.2 The $^{240}$Pu(d,p) experiment

For the $^{240}$Pu(d,p) experiment, a deuterium beam of 13.5 MeV was chosen, with an intensity of about 1.5 µA. The target consisted of 0.4 mg/cm² thick $^{240}$Pu, placed on a backing of 2.3 mg/cm² $^9$Be for structural support, and was oriented such that the backing faced the beam. The target was purified and assembled using an anion-exchange resin column procedure and then electro-plating, as described by Ref. [57]. At a distance of 5 cm behind the target, the fission fragment detectors PPACs (Parallel Plate Avalanche Counter) were situated. The PPACs were used to select fission events, that is, to check if particles or γ-rays arrived at the same time as a fission fragment. SiRi was placed in backward angles, 5 cm from the target, and thus covering angles 126° – 140° in the azimuthal direction. SiRi, the target and the PPACs were all placed inside a vacuum tube, which in turn was enclosed by OSCAR. The distance between OSCAR and the target was chosen to be 20 cm for most of the detectors. This was a trade-off between covering as much of 4π as possible, while still being able to discriminate between neutrons and γ-rays based on time-of-flight. One detector was pulled back to 40 cm, to further test the ability to discriminate between neutrons and γ-rays. In total 28 of the 30 LaBr₃-detectors were installed. The setup is illustrated in figure 4.3 and a picture of the detector setup is shown in figure 4.4a.

SiRi was chosen to be placed in the backward position. This was because we wanted to obtain fission events that were as similar to $^{240}$Pu(n,f) as possible, where the compound nucleus $^{241}$Pu* was formed in an excited state. It is known that direct reaction products are preferably emitted in the forward angles, while compound reaction products are emitted more isotropically [27, pg. 420]. We wanted to study compound (d,pf) reactions instead of the direct reaction. The ratio of protons from the compound reaction compared to the direct reactions is thus larger in the backward angles. Therefore, the particle detector SiRi was
Figure 4.2: a) The SiRi particle detector, showing the eight pads it consists of, and b) the layout of one pad in SiRi: eight $\Delta E$-strips in the front, each covering an azimuthal angle range, backed by an E-detector. Both images reproduced from Ref. [56], the latter changed to include the backward angles.
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Figure 4.3: Experimental setup, showing the placement of SiRi, PPACs and OSCAR relative to the target. Only one of the LaBr$_3$-detectors are shown. The figure is not to scale.

Figure 4.4: In a), OSCAR is shown in the open position, with the vacuum tube containing SiRi and the PPACs visible. The fission fragment detectors, four parallel plate avalanche counters (PPACs) called NIFF, are shown in b).
placed in the backward angle.

The PPACs used to select fission events are gas filled, low-pressure detectors \[58\]. At an optimal gas pressure, the PPACs are sensitive to heavy ions only, and do not fire when hit by scattered beam particles or light ejectiles resulting from other nuclear reactions. The PPACs have no energy resolution, and cannot be used to identify masses or charges of fission fragments, but the time resolution is good. These properties make the PPACs excellent for fission selection. The detector used in this experiment, NIFF (Nuclear Instrument for Fission Fragments), consisted of four separate detectors, as shown in figure 4.4b. These PPACs were placed in the forward direction as fission fragments are easily stopped, and could not travel through the thick backing of the target. As the fission fragments are sent out more or less back-to-back, and only one fragment has to be detected in order to detect a fission event, this resulted in the PPAC being highly effective, even though the detectors covered less than 50\% of 2\pi \[58\]. For the fission fragments to be able to reach the fission counters, the setup was placed in a vacuum. The user had to be careful with the pressure in the detectors, as the thin aluminized Mylar foil separating the gas and the vacuum in these PPACs \[58\] easily broke.

By using PPACs in combination with SiRi and OSCAR, we got a very useful setup for studying γ-rays emitted after fission. SiRi was able to detect the proton from the (d,p) reaction, while OSCAR detected the γ-rays. We could thus select events where the γ-ray arrived at the same time as both a proton and a fission fragment, and this was the fingerprint of the prompt fission γ-rays from the (d,pf) reaction.

4.2 Electronics and data acquisition at OCL

The data acquisition is crucial for the success of an experiment. The Oslo Cyclotron Laboratory recently got new digital electronics replacing the old analog system, and the experiment described in this thesis was one of the first experiments conducted with the new electronics. A brief description of the signal treatment will follow here.

In the digital electronic setup, all the detectors are connected to the Multichannel Digital Gamma Finder (DGF) of the type PIXIE-16, produced by XIA. Here, each detector has its own channel: for the ∆E-detectors there are 64 channels, 8 channels are for the E-detectors, and 30 channels are dedicated to the full OSCAR array of 30 LaBr₃-detectors. In the present experiment, the PPAC signals were passed through a fast amplifier and a constant fraction discriminator (CDF) before entering the DGF. In the DGF, all events arriving in the detector are stamped with the energy and time of the signal. The ∆E- and E-detectors are sampled with a frequency of 250 MHz, while the LaBr₃-detectors are sampled with 500 MHz. In this experiment, the PPACs were sampled with
Figure 4.5: The new DAQ at OCL, where Digital Gamma Finders (DGFs) sample the detector signals, stamp them with the time and energy values, create events, and send the events to be stored on disk. The DAQ for the PPACs in the present experiment is also included.

In theory, all the incoming data could be saved to disk, but it would result in vast amounts of data that we are not necessarily interested in. Instead, the DGFs include a validator, that accepts and rejects events. The firing of an E-detector is the start signal of a good event. As the E-detector is rather slow with respect to the other detectors, the signals from the LaBr$_3$- and PPAC-detectors are digitally delayed so that they arrive after the E-detector signal. All signals that arrive within 4 $\mu$s of the E-detector are stored with the E-detector signal. They are sent through an optic cable and stored on a disk. For an illustration of the data acquisition (DAQ), see figure 4.5.

4.3 Energy calibration

4.3.1 Particle detector SiRi

A $\Delta E$-E particle detector, like SiRi at the OCL, takes advantage of the fact that particles of different mass, velocity and charge deposit distinct amounts of energy $E$ per distance $x$ when they travel through matter. This is summed up by the Bethe-Block formula, which described the average energy loss per
distance $-\frac{dE}{dx}$ [59]:

$$-\frac{dE}{dx} = 2\pi N_a r_e^2 m_e c^2 \rho Z \frac{z^2}{A\beta^2} \left[ \ln \left( \frac{2m_e\gamma^2 v^2 W_{max}}{I^2} \right) - 2\beta^2 - \delta - \frac{2C}{Z} \right], \quad (4.1)$$

where

$Z$: atomic number of matter
$A$: atomic weight of matter
$z$: charge of incoming particle
$r_e$: classical electron radius
$m_e$: electron mass
$N_a$: Avogadro’s number
$I$: mean excitation potential
$\rho$: density of matter
$\beta$: $\frac{v}{c}$ for incoming particle
$\gamma$: $\frac{1}{\sqrt{1-\beta^2}}$
$\delta$: density correction
$C$: shell correction
$W_{max}$: maximum energy transfer in a single collision

In a ΔE-E detector, an incoming charged particle will first deposit some of its energy in the front-facing ΔE-detector, before it is stopped in the E-detector. The beam energy must be chosen such that the particles actually are stopped in the E-detector. If this is not the case, we say we have punch-through where only a portion of the particle’s original energy has been measured.

As described by the Bethe-Block formula, the energy deposited in the ΔE- and E-detectors is characteristic depending on the particle. When plotting the deposited E-energy to the ΔE-energy, one obtains plots like the one shown in figure [4.6]. In these so-called banana or ΔE-E plots, the different particles are easily separated. The proton, being the lightest of the $Z = 1$ particles, loses the least energy in the ΔE-detector. The bottommost banana thus shows the protons. Above it, we see the deuterons from the elastic collision (d,d’), and above that again, even some (d,t) reactions.

The most energetic protons resulting from the $^{240}$Pu(d,p) reaction are the ones where $^{241}$Pu is left in the ground state. Little energy is deposited in the ΔE-detector, and the largest fraction is lost in the E-detector. These protons appear in the rightmost part of the proton banana, as shown in figure [4.6]. For a slightly slower proton coming from an excited state, this fraction will be different. We can thus observe the populated states in $^{241}$Pu as proton “blobs” in the ΔE-E-plot until the spacing between states becomes too narrow for the energy resolution of the particle detector.

In order to calibrate the particle detector, we had to know how much energy was deposited by the incoming protons in the ΔE- and E-detectors, respectively. For this, the reaction kinematics calculator Qkinz [52] was employed. Using the thickness of the target and the backing, along with the incoming beam energy, Qkinz calculates the energy the ejected particle will deposit in the ΔE- and E-detectors for different states in the target nucleus. As this calculation is angle dependent, the Qkinz calculations must be done for each separate strip in the
Figure 4.6: ∆E-E energy spectrum obtained from running a 13.5 MeV deuterium beam on a $^{240}$Pu target, where the $x$-axis shows the energy deposited in the E-detector, the $y$-axis shows the energy deposited in the ∆E-detector, and the $z$-axis shows number of particles registered per bin. The red circle shows the position of the detected protons with the highest energies.
SiRi detector. Even though Qkinz includes the recoil of the target nucleus, the unknown direction of the recoil leads to uncertainties in the case of light nuclei. For calibrating the \( \Delta E-E \) detector, reactions with heavy nuclei are therefore preferred.

We assumed that the response of the \( \Delta E-E \)-detector is linear, such that the channel number \( ch \) in a detector relates to the energy \( E \) in the following way

\[
E = a \cdot ch + b, \quad (4.2)
\]

where \( a \) and \( b \) are referred to as the gain and shift of the detector. With a linear calibration, only two calibration points are needed in order to create a calibration. Thus, two plutonium states had to be identified from the banana plot in figure 4.6.

Not all states in a nucleus are populated in a given reaction, so level population schemes for the \( ^{240}\text{Pu}(d,p) \) and \( ^{240}\text{Pu}(d,d') \) reactions at similar angles and beam energies were found in Ref. [60] and [61]. From figure B.1a in appendix B, we see that the ground state in \( ^{241}\text{Pu} \) is not strongly populated in the \( (d,p) \) reaction, and there are several states close to the ground state. A Gaussian smoothing of the peaks yielded that the resulting peak should be observed at \( \approx 100 \text{ keV} \). In contrast, as we see from figure B.1b the ground state in \( ^{240}\text{Pu} \) is highly populated through \( (d,d') \), and we could thus be sure that the strong peak to the right in the deuterium banana indeed was the ground state of \( ^{240}\text{Pu} \). After consulting figure B.1a and comparing it to figure 4.6, it was found that the rightmost tip of the proton banana did not result from \( ^{241}\text{Pu} \). The ground state in \( ^{241}\text{Pu} \) is marked in figure 4.7, and the counts to the right of this probably resulted from contaminants, as has been observed at previous actinide experiments done at OCL [62].

With states in both \( ^{240}\text{Pu} \) and \( ^{241}\text{Pu} \) identified, we had the two plutonium calibration points needed in order to create the linear calibration. Gains and shifts were calculated for each of the 64 \( \Delta E \). As there were intrinsic variations in the E-detectors, each E was calibrated with each \( \Delta E \) related to it, resulting in 64 calibrations for the E-detectors as well.

After the particle detector was calibrated, particle identification was done by calculating the so-called apparent thickness of the \( \Delta E \)-detector. By assuming that all the incoming particles are protons, and looking at the energy lost in the \( \Delta E \)-detector, one can calculate how thick the \( \Delta E \)-detector seems for this particle. This is called the apparent thickness of the \( \Delta E \)-detector. If the incoming particle is a proton, the calculation will give the true thickness of the \( \Delta E \)-detector. Otherwise, the thickness is overestimated. By plotting the apparent thickness of all the particles detected, we obtained the spectrum shown in figure 4.8. We could separate the protons, which gave us the true thickness of \( \Delta E \)-detector at about 130 \( \mu \text{m} \), from the deuterons and tritons. By putting a gate on the apparent thickness, we could select one type of particle. As this
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4.3.2 Photon detector OSCAR

In order to calibrate the LaBr$_3$-detectors in OSCAR, one needs a set of distinct $\gamma$-rays spanning a wide energy range. For this experiment, this was obtained by running the deuterium beam on a target of $^{28}$Si. As shown in figure B.2 in appendix B, both $^{28}$Si and $^{29}$Si have easily recognizable $\gamma$-transitions as their first excited states, which could be used to calibrate the LaBr$_3$-detectors.

The $\Delta E$-$E$-plot and the ungated $\gamma$-spectrum from the silicone run is shown in figure B.3a and B.3b. In order to pair the $\gamma$-rays with the correct transitions, gates were put on excited states in the $\Delta E$-$E$-plot and it was observed which $\gamma$-rays that arrived together with these particles. For example, by gating on the first excited state in $^{29}$Si, one $\gamma$-line was observed, as seen in figure B.4. This identified the transition as the 1273.4 keV line. This way, several transitions were recognized and used to create a linear calibration of the LaBr$_3$-detectors.

Contrary to the particle detectors, the response of the LaBr$_3$-detectors is known to be non-linear. A quadratic calibration is a better fit, where the calibration takes the form:

$$E = p_2 \cdot ch^2 + p_1 \cdot ch + p_0,$$

(4.3)

where $ch$ is the channel number for a given detector, $E$ is the energy, and $p_0, p_1$ and $p_2$ are the calibration coefficients. However, for $\gamma$-rays in the low-
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Figure 4.8: Apparent thickness of the ∆E detector for the detected protons, deuterons and tritons. The red lines show the particle gate on protons.

to-medium energy range, the quadratic calibration may be approximated with a linear one \[63\], taking the same form as equation 4.2. As previous studies of PFGs show that the majority of them have energies \(< 4 \text{ Mev} \[3, 18, 19\], it was decided that a linear calibration would suffice.

4.4 Aligning detector times

In section 4.2, the need for delaying detector signals is described. In the experiment, there were additionally small variations between the signals from the different detectors, e.g. due to different cable lengths connecting the detectors to the data acquisition. The result was that the times of the detectors were not aligned. Because of this, what one detector referred to as “time 0” did not match another detector’s “time 0”. In order to look at the times between events in different detectors, all the detector times had to be shifted such that they were aligned.

When aligning the detector times, this was done with respect to another detector. Therefore, the detectors with the best time resolution, i.e. the smallest FWHM in the time spectrum, were used to align the slower detectors. The ∆E- and LaBr$_3$-detectors were found to have similar time resolution of about FWHM $= 3.1603 \pm 0.0006 \text{ ns}$, while the PPACs had the worst time resolution of FWHM $= 5.293 \pm 0.001 \text{ ns}$. Consequently, the ∆E-detectors were first aligned with respect to the first LaBr$_3$-detector, and then the LaBr$_3$-detectors with respect
4.5 Coincidences and background subtraction

4.5.1 Coincidences

In the data acquisition during the experiment, all signals arriving within 4 µs of an E-detector were kept. This vast over-collection of data was corrected for in the data sorting routine, in order to find events we were interested in. We were looking for events where the detected reaction products originated from the same reaction event. This is called a true coincidence, and only true coincidences give us the information we want about a reaction.

In contrast to the DAQ, the sorting routine used the arrival of a particle in the ∆E-detector as the start of an event. This is because the ∆E-detectors are much quicker than the E-detectors, as the particle has to travel through the ∆E-detector before reaching the E-detector. To the first order, events where a LaBr$_3$- or PPAC-signal arrived within ±1.5 µs of the ∆E-signal were filled into the time spectra.

After taking the detector time resolution into account, the time scale of true coincidences in (d,p$\gamma$) reactions is typically in the order of ns. A 3 µs time interval will hence include several events that are not true coincidences. For example, there is one new beam burst from the cyclotron every ≈48 ns, and a time interval of 3 µs will include several beam bursts. This means that an event in one detector can be linked to an event from a previous beam burst. The result is weak lines in the time spectrum, called the background beam bursts, as seen in figure 4.3 in appendix B. Random coincidences also occur. The true coincidences were found by putting the so-called prompt time gates in the time spectrum. This prompt time gate put a harsher limit on the time between the ∆E-signal and the signal of either the PPACs or the LaBr$_3$-detectors, such that two events from different detectors that could not be a true coincidence were discarded. The background beam bursts were rejected through this time gate. There would still be random coincidences present, where two events happen to arrive within the prompt time gate, and these were removed from the spectrum through background subtraction.
Figure 4.9: The time spectrum measured in the LaBr$_3$-detectors, showing the time difference $\Delta t$ between the arrival of events in the LaBr$_3$- and $\Delta E$-detectors.

4.5.2 (p$\gamma$)-coincidences: Gating on the prompt peak in the $\gamma$ time spectrum

The particle-$\gamma$-coincidences were chosen by putting a gate on the prompt peak in the LaBr$_3$ vs $\Delta E$ time spectrum, as explained above. The prompt time cut in the LaBr$_3$-detectors was chosen to be $\pm 3$ ns. The background of false events was subtracted by setting an equally large time gate around one of the background beam bursts and subtracting these events from the prompt peak. How these gates were placed, is shown in figure 4.9.

As stated above, the time gate for the prompt $\gamma$-rays in this experiment was set to $\pm 3$ ns. In experiments measuring PFGs, the time gate for what is defined as prompt varies, see for example Ref. [3, 17, 18]. One reason why $\pm 3$ ns was chosen here is that about 95 % of the PFG’s are considered included in this time interval [21].

4.5.2.1 Neutron-$\gamma$-separation

Furthermore, when considering what the prompt gate on the LaBr$_3$-detectors should be, there is also the issue of separating neutrons and $\gamma$-rays. The prompt neutrons released in the fission process can create $\gamma$-resembling signals in the LaBr$_3$-detectors, which will create false p$\gamma$-events. The neutrons can be separated from the $\gamma$-rays based on time-of-flight, and the prompt time gate should
Table 4.1: Neutron time-of-flight in ns, for different neutron kinetic energies $E_n$ and target-LaBr$_3$-detector distances $D$.

<table>
<thead>
<tr>
<th>$E_n$ [MeV]</th>
<th>$D = 20$ cm</th>
<th>$D = 40$ cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>14.5</td>
<td>28.9</td>
</tr>
<tr>
<td>2.1</td>
<td>10.0</td>
<td>20.0</td>
</tr>
<tr>
<td>7.6</td>
<td>5.3</td>
<td>10.6</td>
</tr>
</tbody>
</table>

be set such that it includes as few neutrons as possible. From earlier measurements, it was found that the average prompt neutron energy from the fission reaction $^{240}$Pu(n,f) is around 2.1 MeV [64], and prompt neutron energies in the range 1.0 MeV - 7.6 MeV were observed. The relation between a particle’s velocity $v$, momentum $p$ and mass $m_0$ is known to be:

$$v = c \cdot \frac{pc}{\sqrt{(m_0c^2)^2 + (pc)^2}}, \quad (4.4)$$

where $c$ is the speed of light in vacuum. The energy of the beam given in experiments is the kinetic energy $K$ of the particle, which relates to the momentum $p$ in the following way:

$$K = \frac{p^2}{2m}, \quad (4.5)$$

The expected flight time of the prompt neutrons with different energies can thus be calculated in advance of the experiment. As explained in section 4.1.2, most of the target-LaBr$_3$-detector distances were 20 cm, while one detector was pulled back to 40 cm. The time-of-flight (TOF) of the neutrons for different energies is shown in table 4.1, and as we can see, the quickest neutrons observed by Ref. [64] have a flight time of longer than 5 ns. More energetic prompt neutrons are thus improbable, and this confirms that the prompt $\gamma$-ray gate at $\pm 3$ ns should exclude the majority of the prompt neutrons.

4.5.3 (pf)-coincidences: Gating on the prompt peak in the fission time spectrum

The goal of this thesis is to extract the total $\gamma$ energy and the number of $\gamma$-rays released per fission. These values are heavily dependent on the correct counting of the number of fissions. For the same reasons as for the LaBr$_3$-detectors, a prompt time cut was placed in the PPAC time spectrum. Due to the slower time response of the PPACs, the time cut was set to $\pm 5$ ns, so $\Delta E$-PPAC events that were within this time interval was counted as particle-fission-events. Fission events not in coincidence with a particle were not of interest, as one could not be sure that the compound nucleus $^{241}$Pu* had been formed. A background time cut was also put around the previous beam burst, and fissions events taking place here were subtracted as background. As can be seen from
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Figure 4.10: $\Delta E$-E energy spectrum, after putting an energy-dependent time gate in the $\Delta E$-E-detector time spectrum. The $x$-axis shows the energy deposited in the E-detector, the $y$-axis shows the energy deposited in the $\Delta E$-detector, and the $z$-axis shows number of particles registered per bin.

In figure 4.11a there were few background events in the PPACs, compared to the LaBr$_3$-detectors. This is understandable, as the fission fragments do not scatter around in the detector chamber as the $\gamma$-rays do, in addition to the fact that much of the $\gamma$-ray background is from the intrinsic radioactivity in the LaBr$_3$-detectors. The resulting number of fissions, plotted as a function of excitation energy of the compound nucleus $^{241}$Pu*, is shown in figure 4.11b.

4.5.4 Time gate between the $\Delta E$- and E-detectors

In addition to putting time gates in the LaBr$_3$- and PPAC-time spectra, the time between the $\Delta E$-detector and E-detector events should also be considered. The E-detectors are much slower than the $\Delta E$-detectors, and in addition, low-energy particles take some time to penetrate the $\Delta E$-detector before reaching the E-detector. Therefore, the time differences between the firing of the $\Delta E$- and E-detectors were calculated, and an energy-dependent time gate was put on it. This was in order to cut random $\Delta E$-E coincidences. A loose gate was chosen, as a harsh gate might have discarded good events. The lowest-energy particles could arrive in the E up to 200 ns after the $\Delta E$ fired and still be counted. The resulting $\Delta E$-E plot after this gate is shown in figure 4.10.

4.5.5 True (d,pf$\gamma$)-coincidences

So far, only events where two detectors fired in coincidence have been discussed, either $\Delta E$-LaBr$_3$ for particle-$\gamma$, or $\Delta E$-PPAC for particle-fission, and the back-
Figure 4.11: a), the time difference $\Delta t$ between the PPACs and $\Delta E$, and b), the numbers of fissions counted as a function of $^{241}\text{Pu}^*$ excitation energy.
grounds in these spectra were trivial to subtract. However, the events of interest for this thesis are (pfγ)-events, where three detectors must be considered. The prompt events were easily counted, as they were marked with both the LaBr3-detectors and the PPACs being within their respective prompt time cuts. The background events, those that were subtracted from the prompt spectrum, were decided to be the ones where the γ-ray or the fission fragment arrived in the background time cut, including the events where both were in the background time cut.

As a summary, the detection of true coincidences was as follows: the ∆E-detectors were used as the event trigger. There was a particle gate put on the protons, as explained in section 4.3.1. If a γ-ray arrived within ±3 ns of the trigger, the γ-ray was considered prompt. If there, in addition, was detected a fission fragment in the PPAC within ±5 ns of the trigger, then it was a prompt-fission-γ-event, (d,pfγ). If either the LaBr3-detector and/or the PPACs of the event was within their respective background time cuts, the event was subtracted as background. If no fission fragment was detected and the proton was prompt, the event originated from the reaction 240Pu(d,pγ) instead. This is data that will be used in another analysis.

### 4.6 Raw coincidence matrix

All the γ-rays originating from a (d,pfγ)-event were filled in a so-called coincidence matrix, where the γ energy is plotted against the excitation energy of the compound nucleus 241Pu*. The matrix is shown in figure 4.12.

The excitation energy was calculated from the energy deposited by the proton in ∆E-E by using Qkinz-calculated conversion coefficients. Tabulated values for the stopping powers are used for Z < 93 in the Qkinz-calculations, while for heavier elements, the Bethe-Block formula is employed. This can result in the conversion coefficients being imprecise. It was observed in this experiment that the excited states in 241Pu* known from figure B.1a were found at slightly wrong excitation energies. A linear shift in the excitation energy was therefore introduced to correct for this.

In the coincidence matrix, we expected to observe no γ-rays well below the fission barrier. The integral of the number of points below $E_x = 4$ MeV is close to 0, which is another indication that the background is properly subtracted. Closer to the fission barrier, fission can still occur due to tunnelling.
Figure 4.12: The raw coincidence matrix, showing the energies $E_\gamma$ of $\gamma$-rays from (d,pf\(\gamma\))-events on the \(x\)-axis, plotted against the excitation energy $E_x$ of $^{241}\text{Pu}^*$ on the \(y\)-axis. The \(z\)-axis shows the number of photons per bin. The red lines represent the values for the double-humped fission barrier, as reported by Ref. [31].
CHAPTER 4. EXPERIMENT AND DATA COLLECTION
Chapter 5

Data analysis

All science is either physics or stamp collecting.
— Ernest Rutherford

The raw coincidence matrix in figure 4.12 shows all the \( \gamma \)-ray energies as measured in the LaBr\(_3\) detectors, as a function of \(^{241}\)Pu* excitation energy. This, however, is not the same as the original \( \gamma \) energies. The difference between the two originates from three factors: the ability of a detector to detect \( \gamma \)-rays of different energies, how photons interact with matter, and the experimental setup. This chapter explains the process of unfolding the raw spectrum to correct for the detector response, and how the prompt fission \( \gamma \)-ray characteristics are extracted from this spectrum. A discussion of the uncertainty in the data is also included.

5.1 Unfolding

5.1.1 Detector response

A detector’s ability to detect \( \gamma \)-rays of different energies is a property that has to be considered when choosing detectors for an experiment. Some detectors have a wide energy range where they are able to identify \( \gamma \)-rays, while others are limited to for example only high or low energies. In the \(^{240}\)Pu(d,p) experiment it is the low-energy \( \gamma \)-rays that are crucial, as it is known that most of the PFGs have energies < 4 MeV [17, 18].

If a \( \gamma \)-ray was to enter the detector, it has to interact with the matter in the detector in order to be detected. Photons interact with matter in three main ways which are dominant for different photon energies: the photoelectric
CHAPTER 5. DATA ANALYSIS

effect, Compton scattering, and pair creation. As the cross section of photon interaction is dependent on the absorber material and the photon energy \(^{[59]}[\text{pg. 50-55}]\), what energies the detector is able to detect is only another aspect of how the photon interacts with matter.

In the photoelectric effect, common for low-energy photons, the photon hits an atomic electron, and all the energy goes to kicking the electron loose from its shell and giving it kinetic energy. The electron carries information about the total energy of the photon, and the energy of the original photon can thus be detected.

The Compton effect is also an interaction process of a photon with an atomic electron. This is a scattering process where only a portion of the photon energy is given to the electron. A photon with lower energy continues to travel through the detector, and if it escapes, the information of the photon’s original energy is lost. The measured energy from the electron will create so-called Compton background in the \(\gamma\) spectrum.

For photons with energy above two electron masses \(2m_e = 1022\) keV, a third option called pair production is possible. Here, the photon spontaneously transforms into an electron-positron pair in the presence of a nucleus. The electron can be detected directly, while the positron will annihilate with another electron, creating a pair of 511 keV photons. These photons can, in turn, either interact with the detector through the photoelectric effect or Compton scattering, or one or both can escape from the detector. If both are detected, then the information about the full energy of the original photon is kept. If one 511 keV photon disappears, then a peak shifted with 511 keV with regard to the full energy appears, called the single escape peak. If both escapes, then the double-escape peak appears in the spectrum, shifted with 1022 keV with respect to the full energy peak. Should one of the 511 keV photons escape from one detector, only to be detected in a neighbouring detector, the 511 keV backscattering peak emerges.

The combined effect of these three interactions are illustrated in figure \(5.1\), where the single \(\gamma\)-line in figure \(5.1\) is smeared out due to photon interaction with matter, and becomes the spectrum shown in \(5.1\).

So far, properties of a single detector have been discussed. However, the experimental setup will also alter the whole detector array’s response. For example, with few detectors that stand far apart, Compton scattering from one detector and into another will be minimized. The treatment of the detector signals in the electronics also alters the way the measured photon spectrum looks. It is necessary to not only know the properties of the single detectors, but also the behaviour of the whole experimental setup.

All factors that separate the incoming \(\gamma\) spectrum from the measured spectrum, are described by the detector array’s response function, \(F\), which must
Figure 5.1: a) illustrates the incoming photon spectrum from a single photon transition, and b) illustrates the photon spectrum from a single photon transition as measured in a detector.
be experimentally measured for a given experimental setup.

### 5.1.2 Iterative unfolding procedure

As explained above, when measuring $\gamma$-rays, a raw spectrum like the one in figure 5.1b is measured. The process of unfolding is to go backward, and based on the raw spectrum and the response function of the detectors, to calculate what the original spectrum looked like. The relation between the response function $F$, the folded spectrum $f$ and the unfolded spectrum $u$ is described by [65]:

$$ f = Fu, \quad (5.1) $$

where $f$ reproduces the raw spectrum $r$ within the uncertainties. As the response function $F$ is not necessarily invertible, this equation is solved for $u$ by a numerical iterative method. We do not know $u$, so we start with an initial guess:

$$ u^0 = r. \quad (5.2) $$

We know that when $r$ is sufficiently close to $f^n = Fu^n$, the true unfolded matrix is $u^n$. The difference between $r$ and $f^i$ is therefore the deviation in $u^i$, and must be subtracted in the next guess for the unfolded spectrum, $u^{i+1}$:

$$ u^{i+1} = u^i + (r - f^i). \quad (5.3) $$

This way, after $i = n$ iterations, we have an unfolded spectrum $u \approx u^n$.

### 5.1.3 Note on detector response function for low photon energies

It is known that a significant amount of the PFGs has energies below 1 MeV [17, 18], and the handling of the low-energy $\gamma$-rays can have a great impact on the calculated PFG characteristics. The efficiency of the photon detector varies with the energy of the incoming $\gamma$-ray. From earlier studies done on the PFG’s, scientists have therefore either chosen to simulate the low-energy photons based on the photon distribution, as done in Ref. [3], or given the energy range of which $\gamma$ energies they consider, like in Ref. [18]. The experimental results from this thesis will be compared to the fission simulation code FREYA, which has a $\gamma$ energy cutoff parameter. It was therefore chosen to give the energy range of the detected $\gamma$-rays. The lower energy cutoff was chosen to be 122 keV, below which the LaBr$_3$-detector efficiency drops rapidly. The upper limit was 10 MeV, as practically no $\gamma$-rays with larger energies were detected.

As stated in section 5.1.1, the response function $F$ must be known for a given experimental setup. Geant4 [66] has been used to simulate a preliminary response function of OSCAR. For photon energies below 0.5 MeV, the simulated response have not yet been able to reproduce experimental results.
The unfolded coincidence matrix, showing the energies $E_\gamma$ of $\gamma$-rays from $(d,pf\gamma)$-events on the $x$-axis, plotted against the excitation energy $E_x$ of $^{241}\text{Pu}$ on the $y$-axis. The $z$-axis shows the number of photons per bin. The red lines represent the values for the double-humped fission barrier, as reported by Ref. [31].

The response function for these low $\gamma$-ray energies might therefore not yield the correct unfolded spectrum.

### 5.2 The Compton subtraction method

After each iteration in the unfolding method, unphysical fluctuations have appeared in the resulting matrix. A method for smoothing these fluctuations was therefore presented in Ref. [65], called the Compton subtraction method. By assuming that the Compton background should be a slowly varying function of energy, the Compton background can be smoothed and removed from the raw spectrum. This way, the statistical fluctuations in the unfolded spectrum will be the same as in the raw one. This method is applied when unfolding the coincidence matrix.

After applying the iterative unfolding and the Compton subtraction methods to the raw coincidence matrix, as well as rebinning the matrix, the final coincidence matrix is shown in figure 5.2. The total efficiency of the detector is corrected for later in the analysis.
5.3 Determining the PFG characteristics

After the unfolded coincidence matrix is found, the extraction of the total PFG energy $E_{\text{tot}}$ and multiplicity $M_g$ per fission can begin. The values are extracted as a function of $^{241}\text{Pu}^*$ excitation energy bin $E_{\text{x},i}$, by looping over all the $\gamma$ energy bins $j$.

The total $\gamma$ energy in each excitation energy bin, $E'_{\text{tot},i}$, is found by multiplying the number of $\gamma$-rays in a given bin, $M_{ij}$, with the $\gamma$ energy of that $j$-bin, $E_j$:

$$E'_{\text{tot},i} = \sum_j M_{ij} E_j. \quad (5.4)$$

Similarly, the $\gamma$ multiplicity per excitation energy bin is found by counting the number of $\gamma$-rays in that bin:

$$M'_{g,i} = \sum_j M_{ij}. \quad (5.5)$$

The LaBr$_3$-detector array OSCAR do not cover the whole unit sphere, nor are the detectors 100% effective for detecting incoming radiation. The result is that a large portion of the $\gamma$-rays are lost. In the unfolding routine, the matrix is unfolded relative to the detector efficiency at 1.33 MeV, $\epsilon_\gamma$. In order to obtain the true spectrum, we therefore must divide the unfolded spectrum by $\epsilon_\gamma = 0.27 \pm 0.01$ in the calculations of $E'_{\text{tot},i}$ and $M'_{g,i}$. The value for $\epsilon_\gamma$ is based on preliminary measurements for the OSCAR efficiency.

Thus the equations for calculating the corrected $E'_{\text{tot},i,\text{corr}}$ and $M'_{g,i,\text{corr}}$ are as given in equation 5.6 and 5.7.

$$E'_{\text{tot},i,\text{corr}} = \frac{\sum_j M_{ij} E_j}{\epsilon_\gamma} \quad (5.6)$$

$$M'_{g,i,\text{corr}} = \frac{\sum_j M_{ij}}{\epsilon_\gamma} \quad (5.7)$$

These values must now be divided by the number of fissions per excitation energy bin, $F_i$, such that they take the forms given in equation 5.8 and 5.9. $F_i$ have already been found in section 4.5.3. As the PPACs have no energy response, and only $\gamma$-rays that arrive in coincidence with a fission fragment are counted, we need not unfold or correct for the detector efficiency in $F_i$.

$$E_{\text{tot},i} = \frac{E'_{\text{tot},i,\text{corr}}}{F_i} = \frac{\sum_j M_{ij} E_j}{F_i \epsilon_\gamma} \quad (5.8)$$

$$M_{g,i} = \frac{M'_{g,i,\text{corr}}}{F_i} = \frac{\sum_j M_{ij}}{F_i \epsilon_\gamma} \quad (5.9)$$

The uncertainty given for $\epsilon_\gamma$ is very preliminarily determined.
The average energy per $\gamma$-ray as a function of excitation energy, $E_{g,i}$, is given by

$$E_{g,i} = \frac{E_{\text{tot},i}}{M_{g,i}}.$$  \hfill (5.10)

From here, the total photon energy per fission, photon multiplicity per fission and average photon energy for a given excitation energy bin $i$, are referred to as $E_{\text{tot},i}$, $M_{g,i}$ and $E_{g,i}$ respectively.

5.3.1 Uncertainties

Contrary to the simulated measurements in FREYA, discussed in section 3.2.6, the measurement of the PFGs from the $^{240}$Pu(d,pf) reaction have experimental uncertainties. In principle this means that both systematic and statistical contributions should be included when calculating the total uncertainty. A source of systematic uncertainty is the determination of the $\gamma$ energies $E_j$ in equation 5.8 and the compound nucleus excitation energy $E_{x,i}$. The $E_{x,i}$-bins have a width $\approx 500$ keV, and including a small systematic uncertainty seems unlikely to change the result. Furthermore, an error in $E_j$ would shift all the calculated values up or down, but not change the shape of the photon distribution. We have therefore chosen to disregard this source of uncertainty.

This way, the evaluation of the uncertainty is limited to the statistical uncertainties in $M_{ij}$ and $F_i$, and the systematic uncertainty in $\epsilon_{\gamma}$. The systematic uncertainty in $\epsilon_{\gamma}$ is estimated to be $\sigma(\epsilon_{\gamma}) = 0.01$. As explained in section 4.5, background has been subtracted from the measured $M_{ij}$-spectrum in order to correct for random coincidences. Thereafter, the resulting spectrum was unfolded to obtain the true spectrum. The value $M_{ij}$ is therefore a result of subtracting the background $M_{ij,\text{BG}}$ from the measured spectrum $M_{ij,\text{all}}$, and then unfolding the spectrum. The sources of the statistical uncertainties in $M_{ij}$ are from $M_{ij,\text{all}}$ and $M_{ij,\text{BG}}$ [67, pg. 87] and they are $\sqrt{M_{ij,\text{all}}}$ and $\sqrt{M_{ij,\text{BG}}}$ respectively. How the uncertainty in $M_{ij}$ propagates though the unfolding routine is not known, and it is an active research topic in the Oslo group to find out how this should be handled properly. In this thesis, it is assumed that the uncertainties are left unchanged by the unfolding routine.

It is known [67, pg. 87] that the uncertainty $\sigma_f$ in a value $f(x_1, x_2, ...)$ which is dependent on independent variables $x_i$ with uncertainties $\sigma_{x_i}$, is given by

$$\sigma = \sqrt{\sum_{i} \left( \frac{\partial f}{\partial x_i} \sigma_{x_i} \right)^2}.$$ \hfill (5.11)

As we measure $M_{ij}$ and $F_i$ individually, they are independent variables. By applying equation 5.11 on equation 5.8, 5.9 and 5.10, the total uncertainties on

\footnote{Thank you Amanda Lewis, for the enlightenment on uncertainty calculation!}
$E_{\text{tot},i}$, $M_{g,i}$ and $E_{g,i}$ can be calculated. For the derivation of the formulas for the uncertainties, see section C.1.
Chapter 6

Results and discussion

I never am really satisfied that I understand anything; because, understand it well as I may, my comprehension can only be an infinitesimal fraction of all I want to understand about the many connections and relations which occur to me.
— Ada Lovelace

The measured PFG characteristics from the fission of $^{241}$Pu$^*$ are presented and discussed in this chapter. Sources of uncertainties in the experimental results are discussed, and the results are interpreted in the light of previous PFG experiments. Finally, the experimental results are compared to the results of the FREYA simulation.

6.1 Results

The measured and simulated characteristics of the PFGs from $^{241}$Pu$^*$ are presented. Figures 6.1, 6.2 and 6.3 show the total PFG energy per fission $E_{\text{tot}}$, average PFG multiplicity per fission $M_g$ and average PFG energy $E_g$, plotted as a function of $^{241}$Pu$^*$ excitation energy. Note that the displayed uncertainty only include the statistical uncertainty and the uncertainty in the OSCAR efficiency at 1.33 MeV $\epsilon_{\gamma}$, where $\epsilon_{\gamma} = 0.27 \pm 0.01$. In figure 6.4, the measured photon spectrum is plotted for different excitation energy bins, along with the FREYA photon spectrum for $E_x = 6.75$ MeV. The raw number of (d,pf) and (d,pf$\gamma$) events recorded in the experiment as a function of $^{241}$Pu$^*$ excitation energy $E_x$ is shown in figure 6.5.
CHAPTER 6. RESULTS AND DISCUSSION

Figure 6.1: Total photon energy per fission $E_{\text{tot}}$ from the reaction $^{240}\text{Pu}(d,\text{pf})$, plotted as a function of compound nucleus excitation energy.

Figure 6.2: Average photon multiplicity per fission $M_g$ from the reaction $^{240}\text{Pu}(d,\text{pf})$, plotted as a function of compound nucleus excitation energy.
CHAPTER 6. RESULTS AND DISCUSSION

Figure 6.3: Average photon energy $E_g$ from the reaction $^{240}\text{Pu}(d,pf)$, plotted as a function of compound nucleus excitation energy.

Figure 6.4: Measured spectra of the PFGs from $^{241}\text{Pu}$ for different excitation energy bins plotted against FREYA photon spectrum for $E_x = 6.75$ MeV. The spectra are normalized to photons per fission and MeV.
Figure 6.5: The raw number of $^{241}\text{Pu}\,(d,pf)$ and $^{241}\text{Pu}\,(d,pf\gamma)$ events recorded, plotted as a function of compound nucleus $^{241}\text{Pu}^*$ excitation energy $E_x$. The inner and outer fission barrier, $B_{f,A} = 6.1$ MeV and $B_{f,B} = 5.4$ MeV [31] are also shown.
CHAPTER 6. RESULTS AND DISCUSSION

Table 6.1: The PFG characteristics $E_{\text{tot}}$, $M_g$ and $E_g$ obtained for different actinides by a selection of previous experiments.

<table>
<thead>
<tr>
<th>Ref</th>
<th>Reaction</th>
<th>Energy</th>
<th>$E_{\text{tot}}$ [MeV/fission]</th>
<th>$M_g$ [photons/fission]</th>
<th>$E_g$ [MeV/photon]</th>
</tr>
</thead>
<tbody>
<tr>
<td>14</td>
<td>$^{239}$Pu(n,f)</td>
<td>Thermal</td>
<td>$6.81 \pm 0.30$</td>
<td>$7.23 \pm 0.22$</td>
<td>$0.94 \pm 0.05$</td>
</tr>
<tr>
<td>17</td>
<td>$^{240}$Pu(sf)</td>
<td>SF</td>
<td>$6.6 \pm 0.5$</td>
<td>$8.2 \pm 0.4$</td>
<td>$0.80 \pm 0.07$</td>
</tr>
<tr>
<td>13</td>
<td>$^{233}$U(n,f)</td>
<td>Thermal</td>
<td>$6.69 \pm 0.3$</td>
<td>$6.31 \pm 0.3$</td>
<td>$1.06 \pm 0.07$</td>
</tr>
<tr>
<td>5</td>
<td>$^{235}$U(n,f)</td>
<td>$E_n = 1.9$ MeV</td>
<td>$5.25 \pm 0.20$</td>
<td>$6.54 \pm 0.19$</td>
<td>$0.80 \pm 0.04$</td>
</tr>
<tr>
<td>5</td>
<td>$^{235}$U(n,f)</td>
<td>$E_n = 4.8$ MeV</td>
<td>$6.18 \pm 0.65$</td>
<td>$7.31 \pm 0.46$</td>
<td>$0.84 \pm 0.11$</td>
</tr>
</tbody>
</table>

6.2 Discussion

6.2.1 Overview of current results

We observe from the dark blue lines in figures 6.1, 6.2 and 6.3 that no significant dependence of $E_{\text{tot}}$, $M_g$ and $E_g$ on the fissioning nucleus excitation energy $E_x$ is observed in this experiment. This is further discussed in section 6.2.4.

No experimental measurements of the PFGs from the fission of $^{241}$Pu* have previously been conducted. Rough comparisons to previous experiments can still be made by looking at the PFG characteristics obtained from other actinides in table 6.1, as we do not expect there to be large variations between the actinides. The values for $E_{\text{tot}}$ are about the same compared to those one generally found for actinides. Note that the measured values for $E_{\text{tot}}$ are larger than the average neutron separation energy $S_n$ of the fission fragments, which in chapter 3 was calculated to be around 6 MeV. As explained in section 2.3 this is as expected, and indicates a competition between neutron and photon emission, as neutrons are not always emitted when energetically possible.

However, there is a major discrepancy when it comes to the values for $M_g$ and $E_g$, compared to the previous measurements. While the previous studies generally have average photon energies of below 1 MeV and average multiplicities of about 6–8, the current results show a lower multiplicity at about 5.5 and higher average energy, where the average prompt photon has an energy of about 1.2 MeV. Potential origins for this discrepancy are discussed in section 6.2.2.

For $^{241}$Pu, the inner fission barrier $B_{f,A}$ is larger than the outer fission barrier $B_{f,B}$, as shown in figure 6.5. Sub-barrier fission is observed, mostly when $B_{f,B} < E_x < B_{f,A}$ where the nucleus tunnels through the inner barrier. Some events where $E_x < B_{f,B}$ are seen as well.

The photon spectrum in figure 6.4 shows little change as a function of $E_x$, a behaviour that has been observed previously [3, 4]. For $\gamma$-ray energies below 0.5 MeV, a dip in the photon spectrum is observed. A further discussion of this is included in section 6.2.2.
6.2.2 Systematic errors

In order to obtain reliable experimental results, an analysis of potential systematic errors in the data should be included. In this section, we address what we regard as the main sources of systematic errors in the experiment measuring the prompt fission $\gamma$-rays from $^{241}$Pu$^*$. 

The need for an accurate detector response function is explained in chapter 5. Still, the preliminary analysis of the OSCAR response function shows a deviation from the current Geant4-simulations [66] for the low-energy part of the photon spectrum. Thus the response function is incorrect. This might severely affect the calculated PFG characteristics, and it is also challenging to include estimations of the error this imposes on the results. For future PFG experiments, a simple way to test the detector response is to measure the photons from the spontaneous fission of $^{252}$Cf. As $^{252}$Cf is a well-known source, it acts as a benchmark for the detector characteristics.

The relative detector response at 1.33 MeV, described by $\epsilon_\gamma$, is solely determined preliminarily. Contrary to the detector response function, a variation in $\epsilon_\gamma$ can easily be included in the uncertainty estimation. An uncertainty in $\epsilon_\gamma$ is accounted for in the calculated uncertainties of the PFG characteristics, as described in section 5.3.1.

Another source of error, which is also not included in the error bars displayed in the plots in section 6.1, is contamination in the photon and fission spectra. We discovered that the PPACs had triggered on noise during the experiment, and this resulted in fission triggers being associated with strong $\gamma$-lines from oxygen and beryllium. Thus the raw fission spectrum included oxygen and beryllium contribution. As written in section 4.5.5, the background was subtracted from the photon spectrum when the PPACs and/or the LaBr$_3$-detectors were within their background time cuts, and this cut was chosen to remove as much of the contamination as possible. It was not possible to remove it completely, and the contamination of O and Be is particularly large in the high and low excitation energy regions, which affects the calculated PFG characteristics. Oxygen and beryllium de-excite by fewer $\gamma$-rays of higher energies, which is why in figure 6.5, the ratio between (d,pf$\gamma$) and (d,pf) events is small in the high and low excitation energy regions. This does not reflect the PFG behaviour, and we analyse the PFG characteristics in an excitation energy region where the O/Be contamination is negligible, $E_x \in [5.5, 8.5]$ MeV.

Even within the excitation energy region $E_x \in [5.5, 8.5]$ MeV, the current result show a higher average photon energy $E_\gamma$ and a lower photon multiplicity $M_\gamma$ than previous PFG studies of other actinides. Contamination of O/Be could explain the deviation in our results. However, strong $\gamma$-lines would then have been visible in the photon spectrum in figure 6.4, which is not the case. The observed dip can also be interpreted as too many photons below 0.5 MeV are lost during the processing of the data. This dip is less prominent in other
measurements of the PFGs and occurs at lower photon energies [14, 17]. Here, the unfolding is a likely source of the error, as we know the response function is insufficient for low-energy photons. Furthermore, the cutoff value of 122 keV chosen in section 5.1.3 could result in too many low-energy photons being cut from the data. Both these options might result in too few low-energy photons in the photon spectrum, compared to the spectra of previous experiments.

Moreover, the OSCAR array covers a large fraction of the unit sphere, and thus the detectors are closely spaced. This results in potential crosstalk-events, where a photon Compton-scatters in one detector, escapes and is captured by the neighbouring detector. As crosstalk has not been corrected for in the experimental data, this may result in the photon multiplicity being artificially high. This is not what we observe in our experimental data, but we cannot exclude such a contribution.

### 6.2.3 Comparison to Rose et al. (2017)

One of the most anticipated results from the $^{240}\text{Pu}(d,pf)$ experiment is how the properties of the new LaBr$_3$ scintillation array OSCAR improve the quality of the experimental results. As the first PFG measurements conducted at OCL were run using the old NaI-detector array CACTUS, we can directly compare the properties of the two arrays. In this section, we address the detector challenges faced by Rose et al. in Ref. [3] and present how OSCAR has improved our ability to measure PFGs.

A central question for Rose et al. was how to treat the contribution from the prompt fission neutrons. As explained in section 4.5.2.1, this neutron contribution must be removed from the measured spectrum in order to obtain information about the prompt fission $\gamma$-rays. One way of discriminating the two is by time-of-flight (TOF). With a reported time resolution of 20 ns and a target-detector-distance of 22 cm, the majority of the prompt neutrons were still included in the prompt time cut of Rose et al. Distinguishing photons from neutrons based on the signal produced in the NaI, so-called pulse shape discrimination (PSD), was also not possible. Therefore, the neutron contribution had to be estimated based on previous experimental data and then subtracted.

In the present experiment, the total time resolution was $\approx 4.5$ ns. As shown in section 4.5.2.1, neutrons with energies up to around 8 MeV could be separated from photons based on time-of-flight, and it is improbable to have fission neutrons with higher energies. Consequently, the photon spectrum should in the present experiment not be contaminated with neutron contribution, which significantly increases the quality of the results. Neutrons with energies equal to the average PFN energy of 2.1 MeV are found in the second bump in the time spectrum in figure 4.9 at about 10 ns. This spectrum also includes neutron contribution from deuterium breakup, which are also delayed due to the breakup happening within the potential of the target.
A major advantage with OSCAR compared to CACTUS is thus the simple separation of the photons from the neutrons. This is especially useful when the fissioning nucleus is not among the most studied. In Rose et al. the average $\gamma$-ray multiplicity had to be known in advance in order to subtract the neutrons. When studying the PFGs from a nucleus for the first time, like in this thesis, this information is not available. Without the superior time-resolution of OSCAR, we may thus not have been able to extract valid PFG data from previously unstudied nuclei.

Another source of error in Rose et al. was the high photon detector threshold of 450 keV, where no $\gamma$-rays of lower energies could be detected. In order to compare their experimental results to those presented in Verbinski et al. [14], the photons with lower energies were included by extrapolation, by assuming that the photon spectrum held a constant value for these energies. Rose et al. named the assumed photon spectrum their “dominant source of uncertainty” [3], and this was accounted for by including large error bars. In the present experiment, we operate with a cutoff at 122 keV as discussed in section 5.1.3, but photons of lower energies were observed. No assumption about the photon spectrum had to be made, and thus the quality of the experimental results are significantly improved in the present case compared to those of Rose et al.

At the time this thesis was written, the nucleus $^{240}$Pu$^*$ was the only case where PFG characteristics had been extracted both from the (n,$f\gamma$) and (d,$pf\gamma$) reactions, by Verbinski et al. [14] and Rose et al. [3] respectively. As explained in section 2.4, the impact of the (d,$pf$) reaction on the PFG characteristics compared to the (n,$f$) reaction should be investigated. Even though the spectral characteristics of the PFGs measured from $^{240}$Pu$^*$ differ in the Rose et al. and Verbinski et al. cases, there are other reasons for this deviation, as discussed above. The discrepancy between the Rose et al. results and other measurements of PFGs from $^{240}$Pu$^*$ cannot thus be considered definitive proof that the assumptions about the PFGs from (d,$pf$) and (n,$f$) reactions presented in 2.4 are not valid.

6.2.4 Comparison to PFG behaviour of previous experiments

In section 2.3.1, the current understanding of the PFG behaviour is presented. It is noticeable that the experiment by Frehaut et al. in [2] from the 1980s report a different PFG behaviour as a function of compound nucleus excitation energy $E_x$ compared to the more recent experiments of Rose et al. [3], Lebois et al. [4] and Qi et al. [5]. The results from this thesis in figures 6.1, 6.2 and 6.3 show the same independence of the PFG characteristics on $E_x$ as observed by Rose et al. and Qi et al., and thus do not see the dependence on $E_x$ reported by Frehaut et al. The photon spectrum in figure 6.4 also does not show any evident changes with different $E_x$, as is also reported by Lebois et al. As the results from Frehaut et al. contributed to the model for PFG emission presented in Wagemans [28]
An explanation that might account for the different conclusions between Frehaut et al. in Ref. [2] and the recent experiments, is the range of excitation energies studied. Frehaut et al. studied the range $E_n \in [1, 15]$ MeV, while the range of both Rose et al. and the present work is 3-4 MeV. In Lebois et al., PFGs from the (n,f) reaction was measured using neutron energies $E_n \approx 0$ and 1.7 MeV. Furthermore, Qi et al. measured $E_{\text{tot}}$ for 1.9 and 4.8 MeV. As Frehaut et al. do not report strictly increasing values for $E_{\text{tot}}$, it is unclear how large the discrepancy is. More studies where larger excitation energy regions are covered should be conducted, in order to get a better understanding of the PFGs.

A second potential source of the deviation is systematic errors in the experiments by Frehaut et al. in Ref. [2]. The prompt neutrons and $\gamma$-rays were separated by pulse shape discrimination. If this discrimination was slightly inaccurate, neutrons might have been included in the PFG measurement. More neutrons with higher energies are emitted as a function of $E_x$, and if some of them were counted as $\gamma$-rays, this could explain why $E_{\text{tot}}$ and $E_\gamma$ increase. However, this would give an increase in $M_\gamma$, which is not the case.

### 6.2.5 FREYA results comparison to experiment

A key issue of interest for this thesis is not only what the measured PFGs from $^{234}$Pu look like, but also how they compare to results from the fission code FREYA. As explained in the introduction, by comparing the experimental results to FREYA prediction, we get an insight into whether the FREYA modelling of PFGs is accurate. It can also contribute to understanding critical aspects of these photons.

As discussed in chapter 3, the photon de-excitation model employed by FREYA yields increasing $E_{\text{tot}}$ and $M_\gamma$ with increased compound nucleus excitation energy. Whether this increase should be present in models is discussed in section 6.2.4. As no conclusions can be drawn about this PFG behaviour as function of compound nucleus excitation energy $E_x$, we cannot say if the FREYA model should display this behaviour.

A trend which is obvious when inspecting figures 6.1, 6.2 and 6.3, is that the red lines representing the FREYA calculated results from chapter 3 deviates from the experimental calculations. While the discrepancy in the total PFG energy $E_{\text{tot}}$ is less than 0.5 MeV, the FREYA calculations favour the emission of more low energy photons, and the experimental measurements indicate that fewer photons should be emitted, but each photon should carry more energy on average. The reason behind this disagreement is evident when looking at the photon spectrum in figure 6.4, where the experimentally measured number of photons drops for $\gamma$-ray energies below 0.5 MeV. As discussed in section 6.2.2, this decrease might be due to challenges with the unfolding procedure. In the photon spectrum, FREYA reproduces the experimental photon spectrum for
photon energies \( > 0.5 \) MeV. This raises the question if the only discrepancy between FREYA and the experimental results is due to the photons with energies below 0.5 MeV. We therefore decided to try to fit the low-energy section of the FREYA calculations to the experimental results with an efficiency correction. The number of fissions in FREYA is a variable not determined from the photon spectrum. Therefore, even if such a fit was made, it would not be obvious that FREYA then should reproduce the experimental PFG characteristics. The fit was introduced, and the resulting FREYA calculations are marked “FREYA scaled” in figures 6.1, 6.2 and 6.3.

When including the efficiency correction in the FREYA calculations, the experimental PFG characteristics are reproduced, which means either that the FREYA simulation of photons is wrong for \( E_\gamma < 0.5 \) MeV, or that the experiment has not measured these photons correctly. The fact that the experimental and simulated photon spectrum match for photon energies above 0.5 MeV implies that the simple treatment of the photons employed by FREYA generally is a good description of how the photon emission from the fission fragments takes place, even if it is unclear if there should be a dependence on \( E_x \). This might be surprising, as the FREYA treatment of photons is rather simplified. Individual fragment properties like the fragment shape are not included in FREYA, but as we average over all fragments in this work, the effect of these properties is washed out. Fission codes that account for individual fragment properties exist, like the Monte Carlo code CGMF \[36\], which is based on statistical Hauser-Feshbach nuclear reaction theory. As seen in Ref. \[36\], this elaborate treatment does not necessarily improve the reproduction of experimental photon observables. As the modelling of photons currently relies on experimental data as guidance, more experimental data on photon observables from fission must be available before we can expect our models for photon emission to further improve.
Chapter 7

Summary and outlook

Mischief managed.
— J.K. Rowling, Harry Potter and the Prisoner of Azkaban

7.1 Summary

In this thesis, the prompt fission $\gamma$-rays from the fission of $^{241}\text{Pu}^*$ have been studied, both experimentally and with a state-of-the-art fission simulation code. The aim was to compare the simulation to the experiment, and use it as a benchmark to whether the prompt photon emission process in fission is well understood.

In April 2018, the PFG characteristics of $^{241}\text{Pu}^*$ were measured at the Oslo Cyclotron laboratory, for the first time using the new LaBr$_3$-detector array OSCAR for PFG measurements. The $(d,p)$ reaction, a surrogate reaction for neutron absorption, was applied to a $^{240}\text{Pu}$ target to induce fission. The prompt $\gamma$-rays were measured by observing a proton in the particle detector in coincidence with the firing of the fission fragment and photon detectors. The advantage of this reaction is that the excitation energy $E_x$ of the compound nucleus $^{241}\text{Pu}^*$ can be calculated, giving the PFG characteristics as a function of $E_x$. A drawback, however, is that the $(d,pf)$ reaction is known to give a different angular momentum transfer compared to $(n,f)$ \[28\] pg. 205. More thorough studies of the difference between the $(d,pf)$ and $(n,f)$ reactions and their impact on the PFG characteristics should be conducted in order to determine the difference between the two reactions.

The properties of OSCAR yielded a huge improvement in the experimental results compared to previous PFG measurements conducted with the NaI-
detector array CACTUS. Due to the superior time resolution, prompt neutrons up to $E_n = 8$ MeV could be cut out from the photon spectrum based on time-of-flight. Even though this thesis uses a photon energy cutoff at 122 keV, photons with lower energies could be detected, which is a large contrast to the CACTUS cutoff at $E_{\gamma} = 450$ keV. This increases the quality of the obtained PFG characteristics significantly.

Furthermore, the $^{240}$Pu(n,f) was simulated using the Monte-Carlo based fission code FREYA. The behaviour of the FREYA simulation results was interpreted in the light of FREYA’s treatment of photons, a treatment which largely reflects how prompt photon emission in fission is thought to unfold.

This work observed that the values of the total photon energy per fission $E_{\text{tot}}$, the average photon multiplicity per fission $M_\gamma$ and average photon energy $E_\gamma$ were independent of $^{241}$Pu$^*$ excitation energy, and the values were found to be $E_{\text{tot}} \approx 6.5$ MeV, $M_\gamma \approx 1.2$ and $E_\gamma \approx 1.2$ MeV.

The measured spectral characteristics of the prompt fission $\gamma$-rays both agreed with and violated expectations. The total photon energy $E_{\text{tot}}$ measured was about the same compared to those measured from the fission of other actinides: a little more than the average neutron separation energy among the fission fragments. This is as expected from previous experiments [28, pg. 531], and is a fingerprint of competition between neutron and photon emission in the de-excitation of the fission fragments. However, previous experiments [2] observed an increase in $E_{\text{tot}}$ with an increase in $E_x$. This lead to the interpretation that the fission fragments have more angular momentum when the fissioning nucleus is more excited, and that this angular momentum can solely be disposed of by photon emission. Several recent studies of PFGs could not validate this increase of $E_{\text{tot}}$ with $E_x$ [3–5]. Furthermore, studies of the PFNs and PFGs from $^{252}$Cf(sf) have arrived at different conclusions regarding the relation between neutron and photon emission from fission fragments, which raises questions on whether photon emission in fission fragments is correctly described. By carrying out more experiments where the compound nucleus excitation energy spans a wider range, or where both neutrons and photons are measured simultaneously, the photon emission mechanism in fission can be examined more thoroughly.

At first glance, the FREYA simulations did not seem to reproduce the experimental results. Further examination showed that in the energy range $> 0.5$ MeV, the FREYA results reproduced the measured photon spectrum remarkably well. The only deviation was for photons with lower energies, where FREYA predicted photons that the experiment did not detect. Consequently, the photon multiplicity $M_\gamma$ in the FREYA results was too high, while the total and average photon energy $E_{\text{tot}}$ and $E_\gamma$ were too low compared to the experimental results. This deviation might originate from an incorrect detector response function for those energies, a claim that is supported by how the photon multiplicity measured in this experiment is low, compared to reported results from other PFG measurements from the fission of actinides. When the FREYA sim-
ulation results were fitted to experimental data at low energies, the calculated PFG characteristics reproduced experimental values, though the calculated total energy and multiplicity in FREYA do increase as a function of $E_x$. A more accurate response function of the OSCAR detector array is in progress, and will hopefully lead to conclusions of the accuracy of the FREYA calculations.

### 7.2 Outlook

In this work, our motivation for studying the PFGs was to get a better understanding of the fission process and to test the accuracy of a fission simulation code. Several experiments measuring PFGs are planned in the close future, both using the (n,f) and (d,pf) reactions. More data spanning a larger excitation energy range of the fissioning nucleus will certainly contribute to understanding the differences between the two reactions, as well as clear up the observed discrepancy with older experimental results.

In order to get a better understanding of the PFG behaviour, the competition between neutrons and photons in the de-excitation of the fission process must be investigated. Simultaneous measurements of the prompt photons and prompt neutrons are therefore needed. Few studies have looked at the photon-neutron-correlation from fission fragments, and these studies are conducted almost exclusively for $^{252}\text{Cf}(sf)$ [36]. The current setup at OCL is not suitable for neutron detection, but hopefully, another detector array will soon be underway. Dr. A. C. Larsen from the Nuclear Physics group in Oslo has submitted an application for an ERC Consolidator grant, which includes the building of the detector array StarLight. StarLight will consist of a new type of NaI-detectors, with the ability to detect both neutrons and photons and separate them based on pulse shape discrimination. Though originally intended to be used in experiments for nuclear astrophysics, this detector array’s properties make it ideal for fission experiments where neutrons and photons are measured simultaneously. In addition, the OCL team is currently looking for new fission detectors, and is considering acquiring fission detectors with mass resolution. A future setup of both StarLight and new fission fragment detectors opens up for a range of new fission experiments, where both the neutrons, photons and the fission fragments can be determined. If so, this would give the Oslo group an exceptional opportunity to provide more insights into the de-excitation of the fission fragments.

Just a few months after the work on this thesis started, another motivation for studying fission suddenly emerged with the observation of a binary neutron star mergers by LIGO [68]. Fission recycling, where seed nuclei repeatedly absorb neutrons and then fission, is considered essential for explaining the observed distribution of heavy elements in the solar system [69]. The nuclei of interest here are far more neutron-rich than those we can currently study experimentally. Accurate fission models are therefore needed in nucleosynthesis calculations, which gives fission a renewed interest amongst a wide range of scientists. We expect this revived interest in fission to contribute to more ex-
experiments measuring fission observables being scheduled and a sharper focus on modelling fission, shedding even more light on different aspects of the process.

In this thesis, we have shown that fission is still an active field of research. Countless questions about the process are yet to be answered, and we have contributed with a tiny piece to an exceptionally complex puzzle. Therefore, even though we celebrate 80 years since the discovery of fission in 2019, we are certain that the splitting of heavy nuclei will continue to fascinate scientists for several decades to come.
Appendix A

FREYA

Additional figures used in chapter 3 are given here. Figures A.1 and A.4 show the average mass and kinetic energy of the fission fragments as a function of $^{241}$Pu* excitation energy $E_x$. The prompt fission neutron behaviour as a function of $E_x$ are shown in figures A.2 and A.3 showing the average multiplicity and energy per fission.
APPENDIX A. FREYA

Figure A.1: Average mass of the two fission fragments from the reaction $^{240}\text{Pu}(n,f)$, prior to neutron and $\gamma$-ray emission, plotted as a function of $^{241}\text{Pu}^*$ excitation energy $E_x = S_n + E_n$. Note the broken $y$-axis.

Figure A.2: Average neutron multiplicity from the reaction $^{240}\text{Pu}(n,f)$ as a function of $^{241}\text{Pu}^*$ excitation energy $E_x = S_n + E_n$. The blue curve shows the calculated neutron multiplicity for the reaction, while the red and black curves show the contributions from first- and second-chance fission.
Figure A.3: Average neutron energy from the reaction $^{240}\text{Pu}(n,f)$ as a function of $^{241}\text{Pu}^*$ excitation energy $E_x = S_n + E_n$. The blue curve shows the calculated average neutron energy for the reaction, while the red and black curves show the contributions from first- and second chance-fission.

Figure A.4: Average kinetic energy of the two fission fragments from the reaction $^{240}\text{Pu}(n,f)$, plotted as a function of $^{241}\text{Pu}^*$ excitation energy $E_x = S_n + E_n$. Note the broken $y$-axis.
Appendix B

Experiment and data collection

Here supplementary figures to chapter 4 are given. Figure B.1 shows known levels populated in the $^{240}\text{Pu}(d,p)$ and $^{240}\text{Pu}(d,d')$ reactions, while figure B.2 shows states in $^{28,29}\text{Si}$. In figure B.3 data from the silicone calibration run for the LaBr$_3$-detectors and E-∆E-detectors are shown, and figure B.4 shows the photon spectrum when gating on the first excited state in $^{29}\text{Si}$. The effect of the time alignment is shown in figure B.5.
Figure B.1: a) and b) shows the proton spectra resulting from the reactions $^{240}\text{Pu}(d,p)$ and $^{240}\text{Pu}(d,d')$, measured at the angle of 140° and 125° respectively. Reproduced from Ref. [60] and [61], where the values of the peaks also can be found.
Figure B.2: Level scheme of first excited states in a) $^{28}$Si and b) $^{29}$Si, information reproduced from Ref. [48].
Figure B.3: a) shows the uncalibrated $\Delta E$-$E$ particle spectrum and b) shows the uncalibrated $\gamma$ energy spectrum from LaBr$_3$-detectors, both for the silicone calibration run.
Figure B.4: Uncalibrated $\gamma$ spectrum obtained by gating on the first excited state in $^{29}\text{Si}$. 
Figure B.5: The time difference between the ∆E-detector and one LaBr₃-detector plotted for the different ∆E-detectors, (a) before and (b) after the time alignment.
Appendix C

Data analysis

C.1 Uncertainty

We want to calculate what the uncertainties in the total photon energy per fission \( E_{\text{tot},i} \), the average multiplicity per fission \( M_{g,i} \) and average energy per photon \( E_{g,i} \) in a given \(^{234}\text{Pu}\) excitation energy bin \( i \), based on the assumptions made in section 5.3.1. \( F_i \) is the number of fissions per excitation energy bin, and its statistical uncertainty originate from the number of counts measured for that excitation energy \( F_{i,\text{all}} \) and the number of counts in the background spectrum for the same excitation energy \( F_{i,BG} \).

As \( F_i = F_{i,\text{all}} - F_{i,BG} \), equation \[5.11\] gives that \( \sigma(F_i) = \sqrt{F_{i,\text{all}} + F_{i,BG}} \).

To calculate the uncertainty in \( E_{\text{tot},i} \), we first calculate the uncertainty in \( E'_{\text{tot},i,\text{corr}} \): the total photon energy in an excitation energy bin, corrected for detector response, but not averaged over the number of fissions. We apply equation \[5.11\] on equation \[5.6\] and we obtain that the uncertainty in \( E'_{\text{tot},i,\text{corr}} \) is:

\[
\sigma(E'_{\text{tot},i,\text{corr}}) = \sqrt{\sum_j \left( (M_{ij,\text{all}} + M_{ij,BG}) \frac{E^2_j}{\epsilon^2} + \frac{E^2_j \sigma(\epsilon_j)^2}{\epsilon^4} (M_{ij,\text{all}} + M_{ij,BG})^2 \right)}. \tag{C.1}
\]

By applying the same equation on the expression for \( E_{\text{tot},i} \) in equation \[5.8\] we get that the uncertainty in \( E_{\text{tot},i} \) is:

\[
\sigma(E_{\text{tot},i}) = \sqrt{\left( \frac{\sigma(E'_{\text{tot},i,\text{corr}})}{F_i} \right)^2 + \left( \frac{E'_{\text{tot},i,\text{corr}} \sigma(F_i)}{F_i^2} \right)^2}. \tag{C.2}
\]

Similarly, the uncertainty in \( M'_{g,i,\text{corr}} \), the munpliplicity for an excitation energy bin, corrected for detector response, but not averaged over the number of fissions, is:
\[
\sigma(M_{g,i,\text{corr}}) = \sqrt{\sum_j \left[ (M_{ij,\text{all}} + M_{ij,BG}) \frac{1}{\epsilon^2} + \frac{\sigma(\epsilon)}{\epsilon} (M_{ij,\text{all}} + M_{ij,BG})^2 \right]}. \quad (C.3)
\]

Including the uncertainty in the number of fissions gives:

\[
\sigma(M_{g,i}) = \sqrt{\left( \frac{\sigma(M_{g,i,\text{corr}})}{F_i} \right)^2 + \left( \frac{M_{g,i,\text{corr}} \sigma(F_i)}{F_i^2} \right)^2}. \quad (C.4)
\]

From the equation for \( E_{g,i} \) in equation 5.10 we get that its uncertainty is:

\[
\sigma(E_{g,i}) = \sqrt{\left( \frac{\sigma(E_{\text{tot,i}})}{M_{g,i}} \right)^2 + \left( \frac{E_{\text{tot,i}} \cdot \sigma(M_{g,i})}{M_{g,i}} \right)^2}. \quad (C.5)
\]
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