Characteristics of light charged particle emission in spontaneous and neutron induced fission of Cm and Cf isotopes
The measurements of this thesis were performed at the Institute for Reference Materials and Measurements (EC-JRC-IRMM) in Geel, Belgium and at the Institute Laue Langevin (ILL) in Grenoble, France.
Characteristics of light charged particle emission in spontaneous and neutron induced fission of Cm and Cf isotopes

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Introduction

Talking about fission, people think almost automatically about a nucleus that splits into two heavy pieces. However, in 1946 it has been discovered that once every 300 to 400 fission events a light charged particle accompanies the two fission fragments. Since that day, the so called ternary fission process has been extensively studied. Although its relative small probability, this interest can be explained by two reasons.

First, ternary fission data are of interest for nuclear physics since the ternary particles are emitted in space and time very close to the scission point. Hence they are expected to supply information on the scission point configuration. A good knowledge of their characteristics will also contribute to a better understanding of the emission mechanism of these particles. A second reason is that nuclear industry requests accurate data for ternary fission yields, more specifically of $^3$H (tritium) and $^4$He particles, since they are at the origin of the production of He gas and especially of radioactive tritium gas in the reactor. So a safe operation of the reactor and the handling and reprocessing of irradiated fuel elements require a good knowledge of their production.

The main goal of this work is a study of the emission probabilities and the characteristics of the energy distributions of ternary particles at different excitation energies. In the literature data are available for a whole series of isotopes. However, there exists a lack of data for the exotic curium and californium isotopes, since it is often difficult to get these isotopes with a sufficiently high enrichment. Some of the curium and californium isotopes are very active, which permits only very small amounts of material on the target. For the neutron induced fission measurements, performed at the ILL in Grenoble, this will be compensated by the large neutron flux.

Furthermore a comparison between the data available is not straightforward. Indeed, these results are provided by different research groups and the measurements are performed with different detection techniques. Therefore we want to perform a systematic study of different curium and californium isotopes under the same experimental conditions. This will permit to investigate the correlation of ternary fission characteristics with $(Z,N)$ of the fissioning system as well as with other parameters characteristic for the fission process.

A series of 'isotope couples' will be examined: the neutron induced fission of $^{243}$Cm and the spontaneous fission of $^{244}$Cm, as well as $^{245}$Cm(n,f) - $^{246}$Cm(SF), $^{247}$Cm(n,f) - $^{248}$Cm(SF), $^{249}$Cf(n,f) - $^{250}$Cf(SF) and $^{251}$Cf(n,f) - $^{252}$Cf(SF). These couples allow the study of the most probable ternary particles, namely $\alpha$-particles, tritons and $^6$He particles, of the fissioning systems $^{244,246,248}$Cm and $^{250,252}$Cf at an excitation energy of 0 MeV (SF) and at an excitation energy equal to the binding energy of the neutron ($\approx 6.5$ MeV) in the case...
of the (n,f) reactions. With all the new data a significant enlargement of the existing data base will be realised.

In the first chapter some general aspects of the fission phenomenon are discussed. Furthermore a brief overview of some theoretical fission models can be found here. Chapter 2 is dealing with ternary fission. The general characteristics of ternary fission are discussed, like e.g. emission probabilities, energy and angular distributions. Again some theoretical models to describe the mechanism of ternary fission are mentioned.

Chapter 3 concerns the detection of particles, since this is the basic technique used in our experiments. The possibility to identify particles is discussed and the working principle of $\Delta E - E$ detecting systems is explained. This has been used for the determination of the energy distributions of the ternary particles. In addition, ionisation chambers and semiconductor detectors are described more in detail, since they will be used to perform the measurements.

The next chapter (chapter 4) describes the experimental setup of the measurements. Both locations of the measurements, the IRMM in Geel (Belgium) for the spontaneous fission measurements and the ILL in Grenoble (France) for the neutron induced fission measurements, are presented here. Some words about the electronic setup used for the data acquisition can be read in this chapter. Furthermore the calculation of energy losses and the calibration of the detectors are explained here.

In chapter 5 the measurements of the different curium isotopes are presented with their results. The neutron induced fission of $^{243}$Cm and the spontaneous fission of $^{244}$Cm are discussed in detail, since they have been measured during the four years of this work. The parameters of the energy distributions of $\alpha$-particles, tritons and $^6$He particles obtained from a Gaussian fit to the experimental data can be found here, as well as the emission probabilities for these particles. For the other curium isotopes, measured by our research group in previous years, an overview of the results is given. This chapter is completed with a survey of the literature concerning these curium isotopes.

Chapter 6 deals with the measurements of the different californium isotopes and their results. The structure is the same as in the previous chapter. The neutron induced fission of $^{249,251}$Cf and the spontaneous fission of $^{250,252}$Cf are discussed in detail, completed with a literature survey for these californium isotopes.

In the last chapter of this work, chapter 7, all the obtained results are discussed and interpreted. For a coherent interpretation, the present results are combined with the results for all other isotopes measured by our research group. The average energy and FWHM of the energy distributions as well as the emission probabilities are investigated as a function of the fissility parameter $Z^2/A$. Also the correlation between the emission probabilities and different parameters characteristic for the fission process, like e.g. the coulomb parameter, are discussed. Furthermore the influence of excitation energy on emission probabilities is examined, introducing here the factor $S_{\alpha}$, which is called the alpha cluster preformation
probability factor. Finally a method presented by Lestone is discussed, to infer a nuclear temperature associated with low energy ternary fission using the yield of light charged particles as a function of the mass of the fission system.
Chapter 1

Fission

1.1 Introduction

In 1932 Chadwick discovered the neutron. As often happens, this was the start of the discovery of another interesting phenomenon, called fission. Fermi and co-workers irradiated natural uranium with neutrons, realizing this might lead to the production of elements with greater atomic numbers than uranium, by one or more successive β decays following neutron capture. Out of these experiments, many different radioactive species were produced, which were erroneously assigned to be transuranic elements. Hahn and Strassman (1939) [4] performed a series of experiments to prove the chemical identification of the species, showing that it were isotopes of elements with intermediate mass. Again in 1939, Meitner and Frisch [5] recognized that some nuclei could become unstable after capture of a neutron, and split into two fragments of comparable mass. From this moment on, neutron induced fission was identified. Still the same year, Bohr and Wheeler [6] pointed out the theoretical possibility of spontaneous fission, being the desintegration of the nucleus in two heavy fragments without external supply of energy. The first experimental evidence of spontaneous fission of uranium was obtained one year later by Petrzhak and Flerov [7].

The curve of the nuclear binding energy per nucleon (fig. 1.1) illustrates it is energetically favourable for a heavy nucleus to split into two lighter nuclei, hence releasing an energy of about 200 MeV.

Immediately after the discovery of fission, Meitner and Frisch gave a qualitative explanation of the process using the picture of a charged liquid drop. Today it is known that many details of the fission process cannot be understood without taking into account the shell structure of the nucleus.

1.2 The liquid drop model

The liquid drop model treats the nucleus macroscopically in analogy with a uniformly charged liquid drop. The idea came from the observation that nuclear forces exhibit saturation properties, which can be deduced from figure 1.1. Also, the nucleus presents a low

---

1This chapter is mainly based on references [1, 2, 3].
compressibility and hence a well defined nuclear structure.

The first and dominant term $E_v$ of the binding energy is proportional to the nuclear volume and expresses the fact that the nuclear binding energy is proportional to the number of nucleons ($A$):

$$BE(A, Z) = a_v A.$$  \hspace{1cm} (1.1)

The other terms in the liquid drop formula will reduce the binding energy to a value of about 8 MeV per nucleon.

The second term $E_s$ takes into account the fact that nucleons at, or close to, the nuclear surface will have a reduced binding energy since only partial surrounding with nucleons is possible. The correction is proportional to the nuclear surface area, and we obtain

$$BE(A, Z) = a_v A - a_s A^{2/3}$$  \hspace{1cm} (1.2)

as the new equation.

The third term is the Coulomb term due to the repulsive force between the protons present within the nuclear volume.

For a homogeneously charged liquid drop with radius $R$ and density

$$\rho_c = \frac{Ze}{\frac{4}{3}\pi R^3}$$  \hspace{1cm} (1.3)

one can evaluate the Coulomb contribution to the nuclear binding energy in the following way. The Coulomb energy needed to add a spherical shell, to the outside of the sphere with radius $r$, to give an increment $dr$ becomes

$$E_c' = \frac{1}{4\pi \varepsilon_0} \int_0^R \frac{4}{3}\pi r^3 \rho_c A_\pi r^2 \rho_c dr.$$  \hspace{1cm} (1.4)
1.3. **THE FISSLITY PARAMETER**

Using the above charge density, the integral becomes

$$ E'_c = \frac{3}{5} \frac{Z^2 e^2}{R} \frac{1}{4\pi \epsilon_0}. \quad (1.5) $$

In this way, the charge of $Z$ protons is smoothed out over the whole nucleus. Herewith a self-energy Coulomb interaction is counted, which is false and we should correct for the effect of $Z$ protons. Using the same method as before but now with the proton smeared charge density

$$ \rho_p = \frac{e}{\frac{4}{3} \pi R^3}, \quad (1.6) $$

and a self-Coulomb energy, for the $Z$ protons, as

$$ E''_c = \frac{3}{5} \frac{Z e^2}{R} \frac{1}{4\pi \epsilon_0}, \quad (1.7) $$

the total Coulomb energy correction becomes

$$ E_c = E'_c - E''_c = \frac{3}{5} \frac{Z (Z - 1)}{R} \frac{1}{4\pi \epsilon_0}. \quad (1.8) $$

Using the variables $A$ and $Z$, the following relation is obtained

$$ E_c = a_c Z (Z - 1) A^{-1/3}. \quad (1.9) $$

Adding this term, the binding energy can be written as

$$ BE(A, Z) = a_v A - a_s A^{2/3} - a_c Z (Z - 1) A^{-1/3}. \quad (1.10) $$

### 1.3 The fissility parameter

In application to the fission process, the dependence on deformation is important. For nuclei which deviate from a spherical shape, both the surface and Coulomb energy corrections will change. The deformation energy of the liquid drop model can be defined as

$$ E_{def}(\varepsilon) = E_s(\varepsilon) + E_c(\varepsilon) - E_s(0) - E_c(0) \quad (1.11) $$

with $\varepsilon$ a parameter representing the deformation.

If we limit to the case of an ellipsoidal deformation, the surface of the nucleus can be described by the multipole expansion

$$ R = R_0 (1 + \alpha_2 P_2 (\cos \theta) + \alpha_4 P_4 (\cos \theta)). \quad (1.12) $$

Taking into account the volume conservation, the major axis $a$ and the minor axis $b$ are expressed by

$$ a = R (1 + \varepsilon) \quad and \quad b = R (1 + \varepsilon)^{-1/2}. \quad (1.13) $$
The surface and Coulomb energy terms will become

\[ E_s = a_s A^{2/3}(1 + \frac{2}{5}\varepsilon^2) \]
\[ E_c = a_c Z(Z - 1)A^{-1/3}(1 - \frac{1}{5}\varepsilon^2). \]  
(1.14)

Hence the charged liquid drop will only be stable against small distortions if the decrease in Coulomb energy is smaller than the increase in surface energy. Consequently, the condition for stability of a spherical nucleus is

\[ \frac{a_c Z(Z - 1)A^{-1/3}\frac{1}{5}\varepsilon^2}{a_s A^{2/3}\frac{2}{5}\varepsilon^2} < 1. \]  
(1.15)

This has lead Bohr and Wheeler [6] to define the fissility parameter

\[ x = \frac{E_c(0)}{2E_s(0)}. \]  
(1.16)

We use the simplification \( Z(Z - 1) \rightarrow Z^2 \) and the best fit values for \( a_s \) and \( a_c \), 17.2 MeV and 0.70 MeV, respectively. The condition for stability against spontaneous fission, according to the liquid drop model is then the following

\[ x < 1 \quad \text{or} \quad Z^2/A < 49. \]  
(1.17)

In the context of this work, the value \( Z^2/A \) is noted as the fissility parameter, being a measure of the fissility of the fissioning system.

## 1.4 Shell corrections

Even though the nuclear binding energy systematics mimics the energy of a charged liquid drop to a large extent, the model was inadequate to explain some properties of actinide nuclei. First, the model predicts spherical ground states for all stable nuclei, in contrast to observation. Second, the liquid drop model cannot explain the predominantly asymmetric mass division in the fission of most actinides.

The nuclear shell model, describing microscopic single-particle effects, is responsible for this kind of effects. In the nuclear shell model, the interaction between the constituents of the nucleus is summarized by an average potential, called the shell model potential. Here the particles (protons and neutrons) are assumed to move individually and independently in this potential. The problem with this model is the approximate nature of the shell model potential.

The solution was found by Strutinsky [8] who combined the macroscopic aspect of the liquid drop model with the microscopic effect of the nuclear shell model. His model is called the shell correction model and combines the correct average energy of the liquid drop model with a realistic way of calculating fluctuations of the total energy due to single-particle effects.
The binding energy can now be expressed as

\[ \text{BE}(A, Z) = E_n - E_p - E_c + \delta U = E_{LDM} + \delta U. \]  \hspace{1cm} (1.18)

Besides this shell correction \( \delta U \), a correction \( \delta P \) has to be included which takes into account the pairing energy contribution. Nucleons preferentially form pairs (neutron pairs, proton pairs) in the nucleus under the influence of the short-range nucleon-nucleon attractive force. This effect can be illustrated by studying nucleon separation energies, with the separation energy defined as the energy needed to take a particle out of the nucleus. Plotting neutron and proton separation energies, \( S_n \) and \( S_p \) respectively, a specific saw-tooth figure results (figure 1.2 for \( S_n \) values in the Ce isotopes). This figure clearly expresses the fact that it costs more energy to separate a neutron from a nucleus with even neutron number, than for the adjacent odd-neutron number nuclei. This proves an odd-even effect showing that even-even nuclei are more bound than odd-even nuclei by an amount which we call \( \delta \). Proceeding to an odd-odd nucleus we have to break a pair, relative to the odd-even case and lose an amount \( \delta \) of binding energy. Taking the odd-even nucleus as a reference point we can express the extra pairing energy correction as

\[
\begin{align*}
\delta P &= \delta (e - e) \\
&= \delta (o - e) \\
&= -\delta (o - o).
\end{align*}
\]  \hspace{1cm} (1.19)

![Figure 1.2: Single-neutron separation energy \( S_n \) for the even-even and even-odd Ce (\( Z = 58 \)) nuclei.](image)

Combining all of the above results, we obtain a semi-empirical equation for the total binding energy of the nucleus

\[ \text{BE}(A, Z) = a_e A - a_s A^{2/3} - a_c Z (Z - 1) A^{-1/3} + \delta U + \delta P. \]  \hspace{1cm} (1.20)
1.5 The fission barrier

For heavy nuclei, the $Q$-value or reaction energy for nuclear fission ($Q_f$) is positive, so for these nuclei it is energetically possible to undergo spontaneous fission. Let us have a look to the example of $^{238}U$. If this nucleus splits into two fragments with equal mass, an energy of 214 MeV is released.

Basically nuclear fission is a consequence of the competition between the total binding energy of the nucleons and the repulsive Coulomb energy of the protons. In the case of $^{238}U$, a Coulomb barrier $V$ of about 250 MeV will hinder fission. The difference between $V$ and $Q_f$ is roughly equal to the height of the barrier that has to be overcome to make fission possible, therefore it is called the fission barrier (figure 1.3). It must be stressed this is an approximate calculation, since it is very rare that $^{238}U$ splits into two equal fragments, which will be in most cases deformed.

![Fission barrier diagram](image)

Figure 1.3: Schematic view of $^{238}U$ (as two $^{119}Pd$ nuclei), situated in his nuclear potential.

Figure 1.3 illustrates fission can happen spontaneously, as a natural decay process (by tunnelling through the barrier) or it can be induced by the absorption of a particle (e.g. a neutron) or a photon. In that case, the energy of the fissioning system can go up above the top of the barrier.

Now the idea of the fission barrier will be adapted to the liquid drop model. This model predicts spherical ground states for all stable nuclei, since for $x < 1$, the deformation energy has a local minimum for the spherical case. Most of the actinide nuclei have a value for $x$ between 0.68 and 0.76. For such $x$-values the deformation energy has a saddle point with positive energy relative to the ground state. A schematic picture of a liquid drop model calculation is shown in figure 1.4 [9]. In the upper part deformation energy contours are drawn as a function of the quadrupole and hexadecapole deformation parameters, showing the spherical minimum and the saddle point indicated as "col". The broken line indicates
the most favourable path to fission, and the fission barrier, which is the potential energy effective along this path, is shown in the lower part of the figure.

Figure 1.4: a) Schematic picture of deformation energy contours as a function of quadrupole and hexadecapole deformation parameters. b) The potential energy barrier along the path to fission indicated as the broken line in a.

Due to the shell correction model developed by Strutinsky, a new picture of the fission barrier is obtained. The deformation energy calculated for a typical actinide nucleus with the Strutinsky procedure is schematically illustrated in figure 1.5. The energy is lowest at a deformation corresponding to the known ground state quadrupole moments of actinide nuclei. At the same time a second minimum has appeared due to the strong negative shell correction which occurs at a deformation corresponding to the liquid drop fission barrier. The coordinate labeled "deformation" should be understood as a measure of progress along the energetically most favourable path to fission, the so-called static fission path, in the same sense as for the liquid drop fission barrier.

1.6 Barrier transmission

The probability of fission is mainly determined by the quantum mechanical penetrability $P_T(E)$ through the fission barrier. This penetrability is normally calculated along a one-dimensional path of least action, according to the WKB approximation. The general expression for such a one-dimensional penetration of the barrier, is given by
\[ P_F(E) = \exp \left\{ -2 \int_{\eta_1}^{\eta_2} d\eta \left[ \frac{2B_\eta(V(\eta) - E)}{\hbar^2} \right]^{1/2} \right\}. \] (1.21)

In the above equation \( V(\eta) \) is the potential energy as a function of a coordinate \( \eta \) describing deformation. \( B_\eta \) is the mass parameter of the system with respect to this degree of freedom, and \( E \) being an energy below the barrier top. \( \eta_1 \) and \( \eta_2 \) are the points on the \( \eta \)-axis for which \( V(\eta) = E \).

In the case of ground state spontaneous fission, \( E = E_0 = 1/2\hbar \omega_0 = 0.5 \text{ MeV} \) \cite{10} and \( P_F(E = E_0) = P_0 \). In the simplest case, namely under the assumption of a single barrier having the shape of an inverted parabola with maximum energy \( E_F \) and barrier curvature \( \hbar \omega_F \), equation (1.21) becomes much simplified:

\[ P_0 = \frac{1}{1 + \exp \left[ 2\pi E_F / \hbar \omega_F \right]} \] (1.22)

which is the so-called Hill-Wheeler expression \cite{11}.

If \( f_0 \) is the frequency of oscillations in the fission mode for the ground state in the first well, then the corresponding rate for spontaneous fission is given by the number of assaults to penetrate the barrier multiplied by the penetrability: \( f_0 P_0 \). Hence the mean life \( \tau \) is given by \( 1/f_0 P_0 \). The ground state spontaneous fission half-life \( T_{1/2}(SF) \) can now be expressed as

\[ T_{1/2}(SF) = \frac{\ln 2}{f_0 P_0}. \] (1.23)

For a vibrational frequency corresponding to \( \hbar \omega_0 = 1 \text{ MeV} \), one obtains a number of barrier assaults \( f_0 = 2.4 \times 10^{20} \) per second. Hence for an inverted parabola-shaped fission barrier,
1.7. THEORETICAL MODELS OF FISSION

$T_{1/2}(SF)$ is in a good approximation given by

$$T_{1/2}(SF) \approx 2.77 \times 10^{-21} \exp[2\pi E_F / \hbar \omega_F].$$

(1.24)

This expression shows that the spontaneous fission half-life is determined by the height and the curvature of the fission barrier. In the more realistic case of a double-humped fission barrier, the barrier can be simplified as being composed of two inverted parabolae. In this approximation, $T_{1/2}(SF)$ is given by

$$T_{1/2}(SF) \approx 2.77 \times 10^{-21} \exp \left\{ \frac{2\pi}{\hbar} \frac{E_A}{\omega_A} + \frac{E_B}{\omega_B} \right\}.$$ 

(1.25)

Until now, only ground state spontaneous fission was described. There exist also excited nuclear states, populated in a nuclear reaction and decaying by spontaneous fission. They are called fission isomers. Their half-lives are many orders of magnitude shorter than those of the corresponding ground state spontaneous fission. The excitation energy of the fission isomers is typically of the order of 2 to 3 MeV and these isomers are located in the second minimum of the fission barrier. Also the penetrability of the barrier for the fission isomers is larger, since the outer barrier height is lower.

1.7 Theoretical models of fission

In this section some models are mentioned that were and are important in the way to our understanding of the fission process.

A distinction between static and dynamic models is made.

1.7.1 Static models

The static models have a common property, namely they are almost completely based on potential energy calculations. In this way experimental fission barriers can be quite well reproduced.

The fission process can be described in a qualitative way, in the assumption that the path of the least action is followed by the nucleus. If one wants to perform quantitative calculations to determine e.g. the different distributions, additional assumptions concerning the evolution of the fission process have to be made. In these cases one should always be careful with the interpretation of the results.

Scission Point Model

The basic hypothesis of the Scission Point Model is that during the descent from saddle to scission an equilibrium among the collective degrees of freedom is kept by the fissioning system.

- Statistical theory of Fong [12]
The start of Fong's calculations are two touching fragments with axial asymmetric deformations as the only possible scission configurations. The relative probability to have a pair of complementary fission fragments with certain fission characteristics (mass, charge, deformation...) is calculated statistically as a function of the number of available excitations for the scission configuration. At the moment the model of Fong was developed, the Strutinsky model [8] was not known yet.

- Static model of Wilkins, Steinberg and Chasman [13]

The relative probabilities of formation of complementary fission fragment pairs are determined from the relative potential energies of a system of two nearly touching, coaxial spheroids with quadrupole deformations. The total potential energy of the system at the scission point is calculated as the sum of liquid drop and shell- and pairing-correction terms for each spheroid, and Coulomb and nuclear potential terms describing the interaction between them.

**Random neck rupture and fission channels: model of Broa [14]**

The shape of the nucleus just before scission is described in the model as two spheroids connected by a thick neck. This neck starts to appear shortly behind the last barrier. Now a neck rupture means the neck snaps when the nucleus stretches beyond the precission shape. Random neck rupture means it is not decided where the neck breaks. The probability for a certain mass split is calculated making use of the probability of a neck rupture at a certain position of the neck. The random neck rupture model can compute the mass distribution and his width and the average kinetic energy of the fragments.

Furthermore it has been found that several exit channels for the fissioning nuclei exist. These channels can be found by computing the potential energy as a function of the shape parameters. These calculations have demonstrated that even for extreme deformations shell corrections of the order of 10 MeV can occur. These shell corrections are caused by the occurrence of closed shells, which do not disappear with a certain change in deformation. The existence of such shell effects causes valleys in the potential energy surface, called fission channels.

The different channels correspond with partly different fission barriers and precission shapes. Nevertheless the original theory with only one precission shape was not so incorrect, since in many cases one precission shape is sufficient for more than 95% of all fission events. This shape is called Standard. For more precision at least three precission shapes are needed: Standard, Superlong and Supershort. Standard is slightly asymmetric and of normal length, while Superlong and Supershort are both almost symmetrical and respectively longer or shorter than Standard. Figure 1.6 shows the channel probabilities $p_c$ as a function of the mass of the fissioning system.

Due to intensive studies of Standard fission, two Standard channels have been discovered, namely Standard I and Standard II. They seem to occur in all pre-actinides and
actinides. Standard I mode is generally shorter and more symmetric than the Standard II mode. Therefore Standard I precession shapes make fragments with less deformation.

1.7.2 Dynamic models

The most exact method for describing a fissioning system of many nucleons is to solve simultaneously the equations of motion for each particle. Therefore the time-dependent Schrödinger equation has to be solved:

\[ i\hbar \frac{\partial \Psi(t)}{\partial t} = H \Psi(t) \]  \hspace{1cm} (1.26)

with \( H \) the Hamiltonian for all nucleons in interaction and \( \Psi(t) \) the wave-function of the fissioning nucleus. Due to the complexity of the computation involved, some simplifications can be introduced in order to allow microscopic calculations, like e.g. the Time Dependent Hartree-Fock approximation [2].

The availability of much more powerful computers nowadays has led to a revival of dynamical calculations. As an example, figure 1.7 [15] shows the fragment mass distributions obtained from a dynamical calculation (continuous curve) and a static one (dotted curve) compared to evaluated data from Wahl for 4 MeV neutron induced fission of \(^{238}\text{U}\) (dashed curve). Here we clearly see that maxima have the same location around mass 134 for the heavy fragment in both calculations. It means that the most probable fragmentation is just due to properties of the potential energy surface such as the well-known shell effects. On the contrary, the dynamical distribution is found to be two times broader than the static one, which indicates that the width of the peaks is mostly due to dynamical effects.
1.8 Mass distribution of the fission fragments

As we discussed before, nuclear fission is generally understood as a process where a heavy nucleus splits into two fragments of comparable mass. The daughter nuclei of the fissioning mother nucleus showing up right at scission are called the primary fission fragments. One calculates the total kinetic energy release $E_K$ from the energies $E_L$ and $E_H$ of the individual fragments as

$$E_K = E_L + E_H$$

(1.27)

where the indices $L$ and $H$ represent the light and heavy fragment, respectively. Furthermore, mass conservation for the primary fragments (i.e. before the emission of neutrons) reads

$$M_L^0 + M_H^0 = M_F$$

(1.28)

with $M_L^0$ and $M_H^0$ the masses of the primary fragments and $M_F$ the mass of the fissioning nucleus. From momentum conservation, supposing the fissioning nucleus in rest, one has

$$M_L^0 V_L^0 = M_H^0 V_H^0$$

(1.29)

where $V_L^0$ and $V_H^0$ are the velocities of the primary fragments. For the energies of the primary fragments, the following ratio can be given

$$E_L^0 / E_H^0 = M_H^0 / M_L^0.$$  

(1.30)
Hence the light fission fragments have the most energy.

One of the main characteristics of fission is the fragment mass distribution. A mass distribution is said to be symmetric whenever the fragment yield peaks at $A/2$, so at half the mass number $A$ of the fissioning nucleus. For asymmetric fission the highest yields are attained for two different but complementary fragment mass numbers, the sum being the mass of the fissioning nucleus $A$. Explained in another way, the yield curve, i.e. the yield $Y(A)$ as a function of the fragment mass $A$, is single-humped or double-humped in case of symmetric or asymmetric fission, respectively.

Mass distributions are observed to be symmetric for fissioning compound nuclei lighter than Th and then become asymmetric in the actinides starting from Th. In figure 1.8 some primary mass distributions for thermal neutron-induced fission are given as a function of fragment mass. Comparing these distributions, one observes that the position of the heavy mass group does not shift due to the stabilizing influence of the spherical $Z = 50$ and $N = 82$ shells and the deformed neutron shell $N \approx 87$, so all mass change is taken up by the light fragment. Hence the position of the light group moves to higher mass numbers as the compound nucleus mass is increased. For the superheavy elements on the contrary, the light mass is stabilized by shell effects and all mass change is taken up by the heavy fragments. This is nicely illustrated in figure 1.9 from Itkis et al. [17].

![Figure 1.8](image1.png)

Figure 1.8: Primary mass distributions for a number of isotopes. The mass of the heavy fission fragments remains more or less constant, while the mass of the light fragments increases with the mass of the compound fissioning nucleus [16].
Figure 1.9: Average fission fragment masses for fissioning systems with mass 224 to 305.
Chapter 2

Ternary fission

2.1 Introduction

As explained in the previous chapter, fission is generally a binary process, in which only two particles are formed when the fissioning nucleus splits. These primary fission fragments attain their full energy of motion within a time interval of $10^{-18}$ s, having separated by about $2 \times 10^{-11}$ m. Much less frequently, three particles are formed within $10^{-18}$ s of the instant of scission. This phenomenon is called ternary fission and was discovered in 1946 [18]. The definition of ternary fission covers the whole region of three-particle events, from one extreme mode in which a scission neutron accompanies two primary fragments to the other extreme mode in which three primary fragments of not very different mass are emitted. This last mode is called true ternary fission. The light charged particle (LCP) accompanied fission is situated in between both extremes, and mostly He (and H) isotopes are emitted, although particles up to mass 36 have been observed [19]. Ternary fission occurs once every 300 - 400 fission events.

Since its discovery ternary fission has been extensively studied, both for spontaneous fission decay and neutron induced fission reactions. There are two reasons for this interest. First, ternary fission data are of interest for nuclear physics in order to improve our understanding of ternary particle emission and of the fission process itself. Indeed, the ternary particles are emitted in space and time very close to the scission point. Hence they are expected to supply information on the scission point configuration. A second reason is that nuclear industry requests accurate data for ternary fission yields, more specifically of $^3$H (tritium) and $^4$He particles, since they are at the origin of the production of He gas and especially of radioactive tritium gas in the reactor. Tritium is indeed a $\beta^+$ emitter with a half-life of 12.33 ± 0.02 years, which can be easily absorbed into living tissue. So a safe operation of the reactor requires a good knowledge of their production [20]. At the end of the fuel cycle, several spontaneously fissioning isotopes are present in the fuel element, so even after stopping the reactor tritium is produced in the fuel element. The tritium fission yield is thus important for calculations concerned with handling and reprocessing of irradiated fuels, and for modeling of accident scenarios. The $^4$He is not significant radiologically but is important in terms of materials properties of nuclear fuels and is also used as a standard relative to which the tritium yield can be measured.
2.2 Binary versus ternary fission

Many experiments have been performed to compare the main characteristics of binary fission with those of ternary fission. The main conclusion from such comparisons is that ternary fission has all the essential characteristics of binary fission except those having to do with energetics.

A first observable to consider is the fission fragment kinetic energy. Data for $^{235}$U(n,f), $^{245}$Cm(n,f) and $^{252}$Cf(SF) all agree that the difference between the average total fission fragment kinetic energy in binary and in $\alpha$-accompanied fission is 12 to 14 MeV. This difference is smaller than the mean kinetic energy of the emitted alpha particle, which is about 16 MeV. This suggest that $\alpha$-particles are emitted in the fission events which would have given rise to fragments with less than average kinetic energy and correspondingly greater than average deformation energy. This idea will appear again in section 2.6.

The second quantity to observe is the fission fragment mass distribution. In figure 2.1, the mass distributions in the binary and $^3$H, $^4$He and $^6$He accompanied fission of $^{252}$Cf are compared [21].

![Figure 2.1: Fission fragment mass distributions in $^3$H, $^4$He and $^6$He accompanied fission of $^{252}$Cf. The solid lines show the Gaussian fit to mass spectra in binary fission.](image-url)
2.3 EMISSION PROBABILITIES

It can be observed that both the low- and high-mass peaks in ternary fission are shifted to lower values for the emission of LRA and $^6\text{He}$ particles, while the formation of $^3\text{H}$ seems mainly to result in lower heavy fission fragment masses. An important feature of the fragment mass spectra is that the observed widths of both the low- and high-mass peaks are always smaller in ternary fission. The picture of particle formation from neck nucleons with an accordingly reduced available energy for the formation of the fragments seems to be consistent with the observation of narrower mass peaks [22].

2.3 Emission probabilities

In the study of ternary fission, the emission probabilities of the ternary particles are one of the most important characteristics. First of all, a distinction should be made between absolute and relative emission probabilities. The most common ternary fission corresponds with the emission of an $\alpha$-particle, often called Long Range Alpha or LRA particle, in order to distinguish them from the less energetic particles emitted in the radioactive alpha decay. For the alpha particles absolute emission probabilities are given, noted as LRA/B, which indicates the ratio of the number of ternary alpha particles and the number of fission events. Absolute emission probabilities are also used to indicate the ratio of all ternary fission events (all kind of particles) to the total number of fissions, denoted as T/B.

As mentioned above, in ternary fission, not only $\alpha$-particles are released. The emission probabilities of the rarer particles are generally determined relative to the ternary alpha yield, for instance, the emission probability for the tritons is indicated as $t$/LRA.

A problem to determine the yields of the ternary particles is caused by the particles below the detection limit of the experiment. Since no experiment goes down to zero energy, the measured spectra should be extrapolated to lower energies. Especially one should pay attention to the $\alpha$-particles, as the lower part of the LRA energy distribution deviates from a Gaussian shape (see section 2.4).

Various experiments indicate that the influence of the excitation energy on the emission probability of the ternary particles is very small. For instance, there seems to be almost no difference between the ternary fission of $^{235}\text{U}$ with thermal neutrons and more energetic neutrons [2]. However, comparing the LRA/B values for spontaneous and neutron induced fission surprisingly shows that the number of ternary fissions is higher in the case of spontaneous fission. This will be discussed in more detail in chapter 7.

2.4 Energy distributions

The energy distributions of all ternary particles are Gaussian in shape, except for the $\alpha$-particles. The best explanation for these Gaussian shapes is found in the central limit theorem, which states that when a quantity depends on many uncorrelated parameters, as is the case for the ternary particle emission, its distribution tends to be Gaussian. The non-Gaussian low-energy tail in the ternary $\alpha$-energy distribution is illustrated in fig-
Figure 2.2. Here two partially overlapping experimental LRA energy distributions for $^{235}$U(n,f) are combined, together with a Gaussian fit through the data points above 12.5 MeV. It took some time to discover this phenomenon, since usually the detection limit is around 10 MeV or more. Therefore it is good to mention one should be careful with ternary particles where only the high energy part of the distribution is measured. It is not completely excluded that also other particles can have an asymmetric energy distribution.

![Graph](image_url)

Figure 2.2: Results of the energy distributions of the ternary $\alpha$-particles emitted in $^{235}$U(n,f), obtained by D'hondt et al. [25] and Cañuñoli [26]. The full line is a Gaussian fit through the data points above 12.5 MeV.

Of course, people were questioning the origin of the low-energy tailing of the $\alpha$-energy distribution. An acceptable explanation was found in the decay of the $^{5}$He nucleus. $^{5}$He has not been observed so far in ternary fission, which does not correspond with the yield calculated from the energy cost (see section 2.6.2). This can be explained by the fact that $^{5}$He is unstable ($T_{1/2} = 700 \times 10^{-24}$s) and decays into a neutron and an alpha particle. These decay alphas would be responsible for the observed low-energy tailing. In order to avoid the influence of the non-Gaussian low-energy tail of the $\alpha$-energy distribution, a Gaussian fit can be performed taking into account only the data above 12.5 MeV [23]. More recently, this low-energy tailing was also clearly established for $^{252}$Cf(SF) by Mutterer et al. [24].

To describe the energy distributions of the ternary particles (except if a low-energy tail is present), it is sufficient to determine the average energy $\langle E \rangle$, which corresponds to the most probable energy, and the width of the distribution. This width is indicated by the full width at half maximum (FWHM).
2.5 Angular distributions

Most of the ternary particles are emitted almost perpendicular to the fission axis, which is called equatorial emission. This indicates that most ternary particles originate from the neck region between both main fission fragments. Nevertheless, a small fraction of the ternary particles is emitted along the fission axis or under very small angles with respect to the fission axis and this is called polar emission.

It takes a long time to measure the angular distribution of the ternary particles with a good resolution, because of the low detection geometry. Therefore the information on angular distributions is mainly obtained for ternary alpha particles.

Figure 2.3 shows the angular distribution of the ternary α-particles emitted in the thermal neutron induced fission of $^{235}\text{U}$. In general, the angular distribution has approximately a Gaussian shape around the most probable angle of emission, which is about equal to 83° with respect to the direction of the light fragment.

![Angular distribution diagram](image)

Figure 2.3: The lower part shows the ternary α particle yields as a function of the emission angle (with respect to the direction of the light fragment). The upper part of the figure shows the corresponding kinetic energies [27].

From this figure it is clear that the polar particles have a much higher kinetic energy than the equatorial alphas and they seem to be preferentially emitted from (or near) the
light fragment.

2.6 Theoretical models

2.6.1 Trajectory calculations

[28] Although trajectory calculations are not really theoretical models, they are briefly mentioned here since they can describe what happens to the ternary particle after its emission. Different trajectory calculations have been reported in the literature, varying from very simple to more complicated ones, depending on the hypotheses adopted for the initial conditions. Indeed, in all trajectory calculations initial distributions have to be assumed for the various parameters defining the nuclear configuration at the moment of ternary particle emission. Considering the forces on the ternary particle and on both fission fragments, their trajectories can be calculated, leading to final distributions, which have to be compared with experimental data.

The earliest and most simple calculations use the three point charges approximation, in which the ternary particle and the fission fragments are represented by classical point charges interacting only by Coulomb forces. More recent trajectory calculations consider not only the Coulomb force but also nuclear forces, and demonstrate that factors such as the size of the α-particles and fission fragments and the deformation of the fragments have a strong impact. It has been shown that a unique determination of all the initial parameters is not possible.

2.6.2 Extended Halpern model

A first model to describe the emission mechanism of ternary fission was developed by Halpern [29, 30] in 1965. Soon after the discovery of ternary alpha particles two important conclusions could be drawn, based on their energy and angular distributions. First, the α-particle is emitted from a region between the two main fragments. Its final direction of motion can then be understood in terms of the effect of the Coulomb field of the two fragments. Second, the emission takes place while the fragments are still close together. Otherwise the Coulomb field of the fragment emitting the alpha particle would play a much stronger role than that of the other fragment. Hence the alpha particle must be produced at times close to that of the snapping of the neck that connects them.

A certain amount of energy is needed for the production and release of the ternary particles. Halpern tried to calculate the so-called average energy cost for this production and release of the various particles. His model gives an explanation for the relative yields of the different ternary particles.

Imagine a pair of fission fragments freshly divided. How much energy one would have to supply to remove a particular third particle from one of the fragments and place it midway between both fragments? In figure 2.4 the configuration of the nucleus just after scission is shown in the case of binary and ternary fission. In this illustration the third particle has been taken entirely from \( Z_2 \) and the main fragment \( (Z_2 - Z_3) \) is displaced so that the
center of mass of the entire system has not moved. The average energy cost is the energy required to obtain the ternary from the binary configuration and can be written as follows:

\[ E_c = B + \Delta V + K. \]  

(2.1)

Here \( B \) is the average binding energy of the third particle to its mother fragment, \( \Delta V \) is the average difference in Coulomb energy between the corresponding binary and ternary configurations and \( K \) is the average initial kinetic energy of the third particle. The \( E_c \) values calculated in this way give a minimum of 20 MeV for all ternary particles, which is comparable to the average total distortion energy. Hence, the energy required for the emission of the third particles is taken from the available deformation energy at the moment of scission. This suggests that ternary particles are released with greater chance in those events which normally would have given rise to fragments with less than average kinetic energy and correspondingly greater than average distortion energy. In other words, when the nucleus manages to stretch to a length greater than average before its neck snaps, there is an increase in the chance of emitting a third particle. Thus the emission of particles with \( E_c \) values larger than the average deformation energy must be associated with the fission events having a distortion energy greater than average. As a consequence, the yields of various ternary particles are expected to be a decreasing function of \( E_c \).

![Diagram](image)

Figure 2.4: A diagram to show the different configurations in binary and ternary fission.

Figure 2.5 [31] shows the relative yields of the ternary particles emitted in the ternary fission of \(^{233}\text{Am}^+\) as a function of the corresponding \( E_c \) values. The expected anticorrelation is demonstrated in this picture.

In 1996, a new concept has been introduced in the model of Halpern, which was called then extended Halpern model. Out of the mass spectrum (see section 1.8), it appears there
Figure 2.5: Experimental relative yields of the charged light particles emitted in the ternary fission of $^{243}\text{Am}$ as a function of the calculated energy cost needed to emit these particles. The straight full line is a linear fit.

are a lot of possible fission configurations. Wöstheinrich et al. [31] took into account all the different mass and charge splits of the fragments that are possible, each with their probability $\omega_i$. In this way, the average energy cost can be expressed like this:

$$E_c = \frac{\sum E_{c,i} \omega_i}{\sum \omega_i}. \quad (2.2)$$

For the calculations it was assumed that the ternary particles originate from the heavy fragment. At scission the neck of the nuclide snatches and snaps back to the fragments. The distance between the tips of the fragments directly after this has happened is called the tip-distance. This tip-distance could be evaluated from the total kinetic energy distributions of the fragments. The fragments with the smallest kinetic energy originate from scission configurations with the largest deformations and the largest distance between the fragments.

Figure 2.6 shows the comparison of the experimental and calculated yields for the thermal neutron induced fission of $^{235}\text{U}$. A linear dependence between the logarithm of the yields and the energy costs was assumed for the calculation. All ternary yields are normalized to the $^4\text{He}$ yield.

Due to the presence of the binding energy $B$ in formula 2.1, the even-odd effect illustrated in the experimental results can be explained.

2.6.3 Model of Cărjan [36, 37]

The model developed by Cărjan describes another possible mechanism for the emission of ternary alpha particles. It starts from the observation that all fissioning nuclei are
2.6. THEORETICAL MODELS

Figure 2.6: Comparison of the experimental yields and calculated yields for the thermal neutron induced fission of $^{235}\text{U}$.

alpha emitters in their ground state. This property is conserved along the path towards the scission point, however, the characteristics of the alpha emission being modified as a function of the varying shapes and energy balances associated with every point along this path. The emission points in his model are continuously distributed over the surface of the fissioning system with a preference, only at scission, for the neck region. In such a picture, ternary alpha emission would simply be due to the alpha decay of the fissioning nucleus in the last phase of the fission process. The same reasoning can be applied for the other charged light particles, which would be emitted according to a similar decay mechanism. The angular distribution and the polar emission of the ternary $\alpha$-particles can be explained qualitatively and the average energy and the emission probability of the ternary alpha particles can be reproduced in this model.

2.6.4 Double neck rupture model

Rubchenya and Yavshits [32] presented a new dynamic model of LCP formation in ternary fission. They assumed that ternary fission occurs when two random neck ruptures arise and these ruptures follow one another during a time interval $\Delta t \approx t_{sc}$, where $t_{sc}$ is the rupture time. The part of the neck between two ruptures is considered as the third light particle. The probability of ternary to binary fission is calculated out of the life time of the neck and the rupture time. Finally they obtain an expression for the probability of ternary to binary fission expressed through the characteristics of the fissioning nucleus during its
descent from saddle to scission point.

The LCP are formed from almost all the neck nucleons, since the rupture of the neck happens in a time much smaller than the time that is needed to rearrange the nucleons. This does not correspond to what is observed in experiments, therefore a nucleon exchange between the interacting nuclei is necessary. This is possible as the model describes a rather compact ternary fragment system. They calculated that the light particle is situated near the light fragment, so especially intense exchange must take place in this double system of light fragment + LCP because the overlapping of the density distribution tails leads to the formation of a wide interaction window between the light fragment and the LCP. As the asymmetry of the mass distribution increases the LCP moves towards the light fragment. Starting from some asymmetry the LCP and the light fragment are practically fused together, this is the limit above which no ternary fission can happen anymore.

The main result of the calculations of Rubchenya and Yavshits is a semi-empirical relation to obtain the relative yields of the light charged particles, based on the idea that the double nuclear system formed by light fission fragment and LCP is in equilibrium relative to the internal degrees of freedom. They demonstrated that the logarithm of the experimental LCP yields depend linearly on $Q_{qq} - V_{coul}$, except for the hydrogen isotopes. Here $Q_{qq}$ is the average energy needed to produce the light particle in its final state and $V_{coul}$ is the Coulomb energy in the contact point between the LCP and the light fission fragment.

2.6.5 Pik-Pichak model

The mechanism of ternary fission with emission of light nuclei proposed by Pik-Pichak [33] is based on some experimental facts. First, it appears that all characteristics of ternary fission are very similar to the corresponding characteristics of binary fission with some modifications concerning energetics. According to Pik-Pichak this testifies that the formation and emission of the third particle occur after the instant of separation of fragments in binary fission. Second, the probabilities of emission of various light nuclei in ternary fission depend on the total energy release, i.e. on the binding energies of the emitted nuclei. The higher the binding energy of the light nucleus, the higher the probability of its emission in ternary fission. Finally, the probability of emission of ternary particles seems to be hardly dependent on the excitation energy of the system (see section 2.3).

The light third particle $p$ is situated on the axis connecting the centers of mass of the fragments and the symmetry axis of the light particle is directed along this axis. The emission of a light particle in ternary fission proceeds adiabatically in this model, and occurs at the moment of retracting branches remaining from the neck into the fragments. The energy needed to transit from binary to ternary fission, is called the transition energy $\Delta \epsilon_\alpha$. This energy is connected mostly with the difference between the total energy releases in binary and ternary fission. The energy of transition attains its minimum value in the case of emission of an $\alpha$-particle. The energy of transitions with emission of all other particles can be considerably higher.

The probability of the transition to a state of ternary fission in first order perturbation $W_p$
2.6. THEORETICAL MODELS

theory is given by the expression:

$$W_p = c \exp\left(\frac{-\Delta \epsilon_p}{\Gamma}\right).$$  \hspace{1cm} (2.3)

Hence the probability of emission of a light charged particle $p$ for a given nucleus undergoing fission is determined by two quantities $c$ and $\Gamma$, which can be obtained from the experimental data. The quantity $\Gamma$ is found to have slightly different values for every fissionable nucleus. The value of $\Gamma$ approximately corresponds to the nuclear temperature (see section 7.2.1).

Pik-Pichak made some further assumptions. In the calculation of $\Delta \epsilon$, one should take into account shell effects. In this model no shell corrections are calculated and only the light fragment was supposed to be deformed. This is the case for nuclei which have a heavy fragment with mass number near the value $A_H = 132$, corresponding with the double magic nucleus $^{132}\text{Sn}$. This is for instance valid for $^{236}\text{U}$. A figure illustrating this phenomenon can be found in section 1.8 (figure 1.8).

The relative yields of the ternary particles can be obtained from:

$$W_{p\alpha} = \frac{Y_p}{Y_{\alpha}} = \exp\left(\frac{-\Delta \epsilon_p - \Delta \epsilon_{\alpha}}{\Gamma}\right)$$  \hspace{1cm} (2.4)

with $Y_p$ the yield for a certain ternary particle and $Y_{\alpha}$ the yield for the $\alpha$ particles. In this way, Pik-Pichak developed a model to calculate yields of ternary particles, relative to the alpha particle yield. In figure 2.7 the logarithm of the relative yields of ternary particles as a function of their mass number is shown. A comparison is made for $^{232}\text{Cf(SF)}$ between the experimental and calculated values.

Comparing figures 2.6 and 2.7, it can be concluded that both of them demonstrate the same odd-even effects. However, in figure 2.7 a better agreement between the calculated and experimental values is obtained, which is not surprisingly since the parameters $c$ and $\Gamma$ are extracted from experimental data.

2.6.6 Sudden approximation model

As discussed in 2.6.2, Halpern [29] proposed the idea of the sudden approximation as a model for the ternary particle emission mechanism. He suggested that a rapid transfer of the available deformation energy into release energy of the ternary particle takes place through a sudden snap of the neck stubs after scission. This collapse must be sudden since a slow thinning of the neck followed by a slow retraction of the neck stubs into the fragments would transform most of the deformation energy into excitation energy of the fragments. In that case the ternary particle would not receive enough energy to be released. The process of ternary particle release is therefore probably not adiabatic, but rather sudden. This model was only able to give a qualitative description of the phenomenon of ternary fission.

This idea was extended by Serot et al. [34, 35] for the emission of ternary alpha particles. Based on a study of the characteristics of LRA particles emitted during spontaneous fission of $^{238,240,242,244}\text{Pu}$ isotopes, they suggested the following emission process.
The LRA emission process occurs only if three conditions are fulfilled. First, the available energy should be larger than a minimum quantity that is related to the energy cost \( E_c \) (see section 2.6.2) which corresponds to the energy needed to put an alpha particle between two fragments. The \( E_c \) is supposed to be taken from the deformation energy of the scissioning nucleus. In the case of spontaneous fission, the total excitation energy (TXE) is mainly composed of the deformation energy. Hence, the probability \( P_{df} \) that the deformation energy is larger than \( E_c \) can be written as follows (\( Pr \) means probability):

\[
P_{df} = Pr(TXE > E_c).
\]  

The second condition asks that the alpha particle is already present in the fissioning nucleus \[36\]. In this context, the alpha cluster preformation probability \( S_\alpha \), also called spectroscopic factor, is calculated. It is supposed that \( S_\alpha \) is rather constant during the fission process \[37\] and the calculation is done in a semi-empirical way using the following formula:

\[
S_\alpha = b\lambda_c/\lambda_{WKB}
\]  

where \( b \) is the branching ratio for the ground state to ground state transition, \( \lambda_c \) is the experimental alpha decay constant and \( \lambda_{WKB} \) the alpha decay constant calculated from the WKB approximation (see section 1.6). The experimental values for \( b \) and \( \lambda_c \) can be found in \[38\].

The quantity \([\text{LRA/B}]/S_\alpha\) corresponds to the escape probability of an \( \alpha \)-particle from the scissioning nucleus.
2.6. THEORETICAL MODELS

Still a third condition has to be fulfilled, namely the minimum energy \( E_c \) needed should be effectively transferred to the LRA particle. The calculation of the probability of this energy transfer, \( P_{Tr} \), requires a mechanism by which this energy transfer occurs. The model that determines the values \( P_{Tr} \) is only realistic if it leads to the prediction that the calculated values are strongly enhanced with the nuclear elongation at scission and it should explain the characteristics of the LRA angular distribution.

Here the model of sudden approximation is used and treated in one dimension, namely the deformation axis. If the neck rupture time is very small compared to the typical period of motion for an \( \alpha \)-particle inside the scissoring nucleus, the process can be described as infinitely rapid. The potential changes immediately to the potential of just after scission, so that the particle has no time to follow this change. In this case, the \( \alpha \)-motion is not at all adiabatic, and the \( \alpha \)-particle can be described immediately after scission by the same wave function as just before scission.

With the knowledge of \( \psi_{LRA}^\alpha \), which describes the \( \alpha \)-particles escaping from the nucleus, the energy transfer probability \( P_{Tr} \) can be determined.

So far, the LRA emission probability can be expressed by the following relation:

\[
LRA/B = S_\alpha P_r(TXE > E_c) P_{Tr}.
\]

(2.7)

In figure 2.8 it is shown that \( P_{Tr} \) strongly depends on the \( Q \)-value of the LRA particle (\( Q_\alpha \)) at the scission point and on the elongation of the scissoring nucleus. This elongation is indicated by the parameter \( D_{em} \), which corresponds to the distance between the two centers of mass of both fission fragments. The strong increase of \( P_{Tr} \) with the elongation is in agreement with the experimental data.

![Graph showing the energy transfer probability \( P_{Tr} \) as a function of the \( Q_\alpha \)-scission value for five different elongations at scission.](image)
Indeed, experimental results for a set of four plutonium isotopes [34] allowed the investigation of the possible influence of the fission modes on the LRA emission probability. Strong variations of the fission mode components Standard I and Standard II are reported for these isotopes in [39]. The average total excitation energy \( \langle TXE \rangle \) for a certain isotope can be deduced:

\[
\langle TXE \rangle = W_I \langle TXE \rangle_I + W_{II} \langle TXE \rangle_{II}
\]

with \( \langle TXE \rangle_i \), the average value of the \( TXE \) distribution \( Y_{i=III}(TXE) \) and weighed by the corresponding fission mode components \( (W_i) \). Following this reasoning, formula 2.7 can be generalised taking into account the two fission modes:

\[
LRA/B = S_\alpha \sum_{i=I,II} P_r(TXE_i > E_c) P_r^i.
\]

Supposing that \( P_{Tr} \) is constant for all Pu-isotopes for a given fission mode, this formula can be rewritten as follows:

\[
\frac{[LRA/B]/S_\alpha}{W_I P_r(TXE_I > E_c)} = P_r^I + \frac{W_{II} P_r(TXE_{II} > E_c^{II})}{W_{II} P_r(TXE_{II} > E_c^{II})} P_r^{II}.
\]

![Figure 2.9: Correlation between LRA emission probability and TXE taking into account both alpha cluster preformation and fission modes probabilities.](image)

In figure 2.9 \( \frac{[LRA/B]/S_\alpha}{W_I P_r(TXE_I > E_c)} \) is plotted as a function of \( \frac{W_{II} P_r(TXE_{II} > E_c^{II})}{W_{II} P_r(TXE_{II} > E_c^{II})} \). In section 1.7.1, the Standard I mode is described to be not only less asymmetric than Standard II but also shorter. From Fig. 2.9 we can learn that the energy transfer probability is higher in the Standard II than in the Standard I fission mode since \( P_{Tr} \) is strongly favoured with
the elongation of the scissioning nucleus. Furthermore, since Standard II corresponds to a more deformed scissioning nucleus than the Standard I fission mode, there are more fission events with enough energy available in Standard II than in Standard I fission mode.

Within the same model, the LRA angular distribution can be obtained after trajectory calculations. Figure 2.10 shows the angular distribution for three different configurations, in order to study the influence of the mass asymmetry $R = M_R/M_L$ and the elongation $D_{cm}$:

- $D_{cm} = 18.2$ fm and $R = 1$
- $D_{cm} = 20.5$ fm and $R = 1$
- $D_{cm} = 20.5$ fm and $R = 1.4$

It is clearly demonstrated that the mechanism leads to both equatorial and polar emission of LRA particles. Comparing cases a and b it is observed that the elongation of the scissioning nucleus increases the polar distribution and the width of the equatorial distribution. Comparing cases b and c, an increase of the polar light contribution appears when the mass asymmetry increases.
Figure 2.10: LRA angular distribution (top of the figure), the shape immediately after scission is plotted on the bottom part, while $|\psi_{LRA}^\text{out}|^2$ (straight line) and $\theta_{\alpha L}$ (dashed line) are drawn in the middle part. All this is plotted for the three cases mentioned in the text above.
Chapter 3

Particle detection\textsuperscript{1}

Experiments related to fission are based on the detection of particles, for instance the fission fragments or the ternary particles that are released at the moment of scission. To detect these particles a large variety of detectors can be used. The main principle of the detection remains the interaction of the energetic particles with the material of the detector, hence generating a signal. In the present chapter we will discuss the functioning and the main characteristics of the detectors used in our experiments.

3.1 Radiation and matter

Different types of radiation are existing and they can be roughly classified based on the electric charge of the radiation. Two main groups can be distinguished, namely the charged particles on the one hand, and the uncharged particles and photons on the other hand. They will be discussed in more detail in the next sections.

3.1.1 Charged particles

The charged particles should be separated into two classes. The first group consists of the electrons and positrons and the second one contains the heavy particles, which includes for instance protons, $\alpha$-particles, other light nuclei and fission fragments.

Heavy charged particles

When heavy charged particles are going through matter, they lose energy due to inelastic collisions with the atomic electrons of the material. In these collisions, energy is transferred from the particle to the atom causing an ionisation or excitation of the latter. The number of collisions per unit path length is so large that the fluctuations in the total energy loss are very small. Therefore one can consider the average energy loss per unit path length. This quantity is often called stopping power and is denoted as $dE/dx$. The Bethe-Bloch formula is the basic expression used for energy loss calculations of charged particles:

\textsuperscript{1}This chapter is mainly based on references [40, 41].
\[- \frac{dE}{dx} = \frac{4\pi n e^4 q_{eff}^2}{m v^2} \left( \ln \left[ \frac{2 m v^2}{I (1 - \beta^2)} \right] - \beta^2 - S - D \right). \tag{3.1}\]

In this equation \( n \) is the number of electrons/cm\(^3\) of the absorber, \( e \) and \( m \) are the charge and the mass of the electron and \( I \) is the mean ionisation potential for the absorber. The properties of the incoming particle are described by \( q_{eff} \), the effective charge of the particle and \( v \), its velocity. Deduced from \( v \), there is also \( \beta = v/c \), with \( c \) the velocity of light. \( S \) is a shell correction for the fact that the electrons from different shells do not all equally participate in the ionisation process and \( D \) is a density correction.

**Electrons and positrons**

As the heavy charged particles, electrons and positrons lose energy when passing through matter due to inelastic collisions with the atomic electrons of the matter. However, because of their small mass, another mechanism for energy loss becomes important, which is called bremsstrahlung. This can be understood as the emission of electromagnetic radiation arising from the acceleration of the electron as it is deviated from its straight line trajectory by the electrical attraction of the nucleus.

Therefore the total energy loss of electrons and positrons is composed of two parts:

\[ \left( \frac{dE}{dx} \right)_{tot} = \left( \frac{dE}{dx} \right)_{rad} + \left( \frac{dE}{dx} \right)_{cell}. \tag{3.2} \]

**3.1.2 Uncharged particles and photons**

In comparison to the charged particles, the energy transfer in the case of uncharged particles happens in a different way, due to the absence of electric charge. Through collisions, charged particles will be created first and these will be stopped in the medium as described in section 3.1.1. The way the charged particles are created, differs for photons and neutrons.

**Photons**

The main interactions of photons with matter are the following:

- Photoelectric effect
- Compton scattering
- Electron-positron pair production

The much smaller cross section of the three processes relative to the inelastic electron collision explains why \( \gamma \)-rays are many times more penetrating in matter than charged particles.
3.2. PARTICLE IDENTIFICATION AND $\Delta E$ - E SYSTEMS

Neutrons

The principal means of interaction for the neutrons is through the strong force with nuclei. These reactions are rather rare because of the short range of this force. Neutrons must come within $\approx 10^{-13}$ cm of the nucleus before anything can happen, therefore it is not surprising that the neutron is observed to be a very penetrating particle.

The neutrons interact with the atomic nucleus through scattering or absorption. Absorption is most likely at low energies, since the interaction probability for neutron capture goes approximately as $1/v$, with $v$ the velocity of the neutron. Depending on the element, resonance peaks can appear superimposed on the $1/v$ dependence. In practice, the neutron will first lose energy by collisions with atomic nuclei before being absorbed. This leads to the formation of an excited compound nucleus, which will decay through emission of other particles or will fission.

3.2 Particle identification and $\Delta E$ - E systems

By using one detector to stop an incoming particle completely, only information on the total energy of the particle can be obtained. However, no information on the mass and the charge of the particle is available. A method to identify particles is developed to determine not only the energy $E$ of a particle, but also the mass $M$ and the charge $Z$ of the particle. To obtain $E$, $M$ and $Z$, three quantities should be measured that are independent functions of $Z$, $M$ and $E$. Several types of measurements depend on independent functional combinations of $M$, $Z$ and $E$, but no single measurement determines a unique set of these parameters.

The value $E$ is nearly always required in an experiment, but fortunately individual values for $M$ and $Z$ are not often required.

To make a difference between non-relativistic particles, it is sufficient to determine the quantity $MZ^2$. Indeed, $MZ^2$ assumes unique values for protons, deuterons, tritons, $^3$He, $^4$He and $^6$He particles of 1, 2, 3, 12, 16 and 24 respectively, so its value characterizes each of these isotopes unambiguously.

In a thin transmission detector, included in a detector telescope, a particle will lose only part of its energy when passing through it, providing a measure for the energy loss per unit path length. This thin detector is called the $\Delta E$ detector. Finally the particle is stopped in a second detector, which is called the E detector. The total energy of the particle $E_{tot}$ is equal to the sum of the energies deposited in both detectors.

In order to calculate $MZ^2$ of an ion, it is sufficient to know $E_{tot}$ and $dE/dx$, together with formula 3.1, under the assumption of a fully stripped ion, with $q_{eff} = Z$. Based on the $\Delta E$ and E signals from both detectors, a function can be generated whose value is characteristic for a particular type of ion and independent of its energy. Based on this value, data that correspond to a certain particle can be selected.

The Bethe-Bloch equation itself cannot be used directly, but after applying some approximations, a simple algorithm is obtained. First of all, the small corrections $S$ and $D$ will be neglected. Furthermore, in the case of nonrelativistic particles, $\beta \to 0$, so equation 3.1 can be reduced to a simpler form.

In figure 3.1 [41] the stopping power, expressed in terms of $(1/Z^2)dE/dx$, is illustrated
for various ions in aluminium. As can be seen in formula 3.1, the stopping power is basically dependent on the velocity \( v \). Taking into account that for nonrelativistic particles \( v^2 = 2E/M \), it is convenient to plot the curves as a function of \( E/M \).

\[
- \frac{1}{Z^2} \frac{dE}{dx} \sim \left( \frac{E}{M} \right)^n
\]  

(3.3)

where the value of \( n \) varies from about -0.75 for a proton to -0.5 for the heavier ions. This power law approximation can be explained by the \( 1/E \) behaviour of the main term in the right-hand side of equation 3.1, combined with the fact that \( \ln E \) can be represented by a \( E^{0.3} \) law in the region of interest. For instance, in the case of \( \alpha \)-particles, \( n = -0.73 \) in the energy region between 4 and 40 MeV, which is the important energy range for ternary fission.

For heavy ions also charge exchange processes are involved, modifying the energy dependence at low energies, resulting in a change of the exponent \( n \).

A simple method used for particle identification, based on a range function, starts from the simplified relation 3.3. For a given ion and over a limited energy range, the stopping power can be written as follows:

\[
- \frac{dE}{dx} = E^n/a
\]  

(3.4)

where the value of \( a \) will be approximately proportional to \( 1/M^{-n}Z^2 \), or more roughly to \( 1/MZ^2 \). The range \( R \) of an ion with energy \( E \) in an absorber can be calculated by integrating the incremental elements of the path corresponding to incremental energy losses.
3.3. GENERAL CHARACTERISTICS OF DETECTORS

Therefore one has:

\[ R = \int_{E_1}^{E} (dx/dE) dE + R_i \]  \hspace{1cm} (3.5)

where \( E_1 \) represents the minimum energy at which the \( dE/dx \) formula is valid, and \( R_i \) represents the remaining range at energy \( E_1 \). For the particles considered, \( R_i \ll R \) and \( E_1 \ll E \). Neglecting these terms, the expression for the range after integration becomes:

\[ R = aE^{1-n}. \]  \hspace{1cm} (3.6)

More generally, one may use an index \( b = 1 - n \) that depends on the type of the ion. Therefore

\[ R = aE^b. \]  \hspace{1cm} (3.7)

Suppose an ion loses an energy \( \Delta E \) in the first detector with thickness \( T \) and is stopped then in the second detector, depositing an energy \( E \). The range of a particle with energy \( E + \Delta E \) is \( T \) longer than the range of the same particle with energy \( E \). Based on equation 3.7, the following relation can be written:

\[ T/a = [(E + \Delta E)^b - E^b]. \]  \hspace{1cm} (3.8)

The left side of this equation is roughly proportional to \( MZ^2 \). The thickness \( T \) of the thin first detector is a known quantity, whereas \( E \) and \( \Delta E \) are determined by measuring the amplitude of the detector signals. Therefore equation 3.8 provides a direct determination of \( MZ^2 \) and can be used for particle identification. The relation above was used in the analysis of our measurements to determine the different ternary particles, namely \( \alpha \)-particles, tritons and \( ^6 \)He particles. For these particles, a value of -0.73 for \( n \) has been adopted, yielding \( b = 1.73 \).

Of course, the choice of the detectors is very important in order to obtain good results. The thickness of the \( \Delta E \) detector is depending on which particles one wants to measure. One should pay attention that the \( \Delta E \) detector is as thin as possible, since the thickness of this detector imposes a lower limit to the energy. The second detector should be chosen in a way that the particles are completely stopped. The dead zone in the detector should be as small as possible. A dead zone in a detector is a part of the detector where the particles pass through and lose energy, but without collection of the free charges. Consequently, the energy of the particles cannot be determined in a correct way.

3.3 General characteristics of detectors

3.3.1 Sensitivity

A first important property for a detector is its sensitivity. A detector should be able to produce a usable signal for a certain type of radiation in a given energy range. This detector sensitivity depends on several factors. The cross section for ionising reactions and the detector mass determine the probability that the incident radiation will convert (part
of) its energy in the detector into ionisation. As charged particles are highly ionising, already detectors of low density and small volume will have some ionisation produced. Even if ionisation is produced in the detector, there exists a lower limit of energy that is necessary in order for the signal to be usable. This is caused by the noise coming from the detector and the associated electronic chain. It is clear the ionisation signal must be larger than the average noise level. Another factor to be taken into account, is the material covering the entrance window of the detector in order to prevent it from radiation damage. Hence, part of the energy of the radiation will be lost in this layer, so it should be chosen as thin as possible.

3.3.2 Detector response

Except detecting the presence of radiation, most detectors are also capable to provide information on the energy of the radiation. This is a consequence of the fact that the amount of ionisation produced by radiation in a detector is proportional to the energy it loses in the sensitive volume. The amount of ionisation is reflected in the output signal, or more specific, in the integral of the pulse with respect to the time. If the shape of this pulse does not change, this integral is proportional to the amplitude or pulse height of the signal. The relation between the radiation energy and the pulse height of the output signal is called the response of the detector.

3.3.3 Energy resolution

When a monoenergetic beam of radiation is send into a detector, ideally, one would like to see a sharp delta-function peak. Unfortunately, this is not the case and one will observe a peak structure with a finite width, Gaussian in shape. This width arises since the interaction of radiation with matter is a statistic process, so there will be fluctuations in the number of ionisations and excitations produced. The width of this Gaussian curve is given in terms of the full width at half maximum of the peak, indicated as FWHM. Energies which are closer together than this interval, are considered unresolvable. This phenomenon is illustrated in figure 3.2.

![Figure 3.2: Illustration of the energy resolution.](image)
3.4. IONISATION CHAMBER

It is important to notice that not only the fluctuations in ionisation determine the energy resolution. In addition, a number of external factors can affect the resolution of the detector, for instance the noise and the properties of the electronics, as well as radiation damage.

3.3.4 The response function and response time

When a detector is bombarded by a monoenergetic beam of a given radiation, a pulse height spectrum will be observed. This pulse height spectrum is called the response function of the detector for this kind of radiation being detected. Up to now, a Gaussian curve is assumed for this response function. If the response of the detector is linear, the pulse heights measured from the detector correspond directly to the energy spectrum.

However, it is possible that a low energy tail will be produced, determined by the amount of energy loss due to scattering and bremsstrahlung.

Another important characteristic of a detector is the response time. This is the time which the detector needs to form the signal after the arrival of the radiation.

3.3.5 Detector efficiency and dead time

The total efficiency of a detector is defined as the ratio of events actually registered by the detector and the events emitted by the source. Both the intrinsic efficiency and the geometrical efficiency are important here. The intrinsic efficiency depends on the interaction cross section of the incident radiation on the detector medium and corresponds to the fraction of the events impinging on the detector which is registered. On the other hand, the geometric efficiency is that fraction of the source radiation which effectively reaches the detector. This depends entirely on the geometrical configuration of the detector and the source.

Related to the efficiency is the dead time of the detector. This is the finite time required by the detector to process an event. During this period, no second event can be accepted either because the detector is insensitive or because the second signal will be added to the amplitude of the first one, which is called pile up. This contributes to the dead time of the detector. Therefore the dead time imposes an upper limit to the counting rate of a measuring system.

3.4 Ionisation chamber

Gas filled ionisation chambers belong to the oldest detectors for ionising radiation. They are based on the direct collection of the ionisation electrons and ions produced in a gas by the passage of radiation. These kind of detectors were used for the detection of cosmic rays in 1911, as well as to demonstrate the existence of the neutron in 1932. In later years, the interest in ionisation chambers decreased with the rise of the semiconductor detectors. A big advantage of the latter detectors is their dimension, since they can be much smaller due to the higher density. However, it is very difficult to build a big semiconductor detector in
order to detect charged particles over large solid angles. Compared to this, the ionisation chamber has the advantage of the 2π detection geometry.

An ionisation chamber basically consists of a closed container, filled with an isolating gas. Inside there are two electrodes, a negatively charged cathode and a positively charged anode, in between which an electric field is applied. When a charged particle passes through the gas volume, it will lose its kinetic energy by interacting with the gas molecules until it is in thermal equilibrium with the gas. Hence electron-ion pairs are created along its track, in this way ionising the gas.

Under the influence of the applied electric field, the positive and negative charges drift to the cathode and the anode, respectively. This movement induces a current in the external measuring chain. This current has to be amplified before it is used for further handling. Analysis of this signal provides information about some characteristics of the ionising particles.

The electric field in the ionisation chamber is a superposition of two fields. First there is the externally applied field, secondly there is the field caused by the created ionisation charges.

Due to the movement of the charges in the gas, currents are flowing in the external chain, which can be considered as caused by a charge induced on the electrodes. The setup of the chamber can be chosen in a way that all electrons and positive ions are collected. In this way the observed signal will be independent of the place of ionisation and moreover it will be proportional with the amount of charge created. A chamber used like described above, is called an ion pulse ionisation chamber.

However there is one big disadvantage about this setup. Due to the applied field, the charged particles will drift through the gas. It is experimentally shown that positive ions move about 1000 times slower than electrons under the same circumstances. Therefore such a configuration is not suitable for high count rates. In order to avoid this problem, the chamber can be configured in a way that the electrons drift towards the anode in a time where the positive ions can be considered as static. Such a chamber is called an electron pulse ionisation chamber.

If the positive ions are considered to be static during the electron collection, they however will induce a charge on the electrodes. Therefore the final collected charge is the one coming from the electrons minus the induced one from the ions, and it is dependent on the place where the electron ion pair is created.

In this work, an ionisation chamber was used with the cathode and the anode mounted parallel to each other, and the sample was placed in the center of the cathode. The anode consists of a 60 μg/cm² thick polyimide foil covered with 50 μg/cm² of gold on both sides. The signal that will be registered, caused by the ionising particle can be expressed as (figure 3.3) [42]:

\[ V = \frac{eN_0}{C} \left( 1 - \frac{X(E)}{d} \cos \theta \right) \]  (3.9)

with \( e \) the electron charge, \( N_0 \) the number of created electron ion pairs, depending on the energy of the incoming particle, \( C \) the capacity of the ionisation chamber, \( X(E) \) the distance between the origin of the ionisation track and the center of the ionisation track, \( d \) the distance between the anode and the cathode and \( \theta \) the angle between the ionisation
3.4. IONISATION CHAMBER

track and the normal of the cathode.

![Diagram of ionisation chamber](image)

Figure 3.3: Schematic picture of the ionisation process caused by a charged particle. The ionisation chamber consists of two parallel plates with the sample mounted in the middle of the cathode.

For an isotropic angular distribution of the emitted particles, the pulse distribution $f(V)$ appears to be constant between $V_{\text{min}}$ in the case of $\theta = 0^\circ$ and $V_{\text{max}}$ for $\theta = 90^\circ$. The problem is that even monoenergetic particles will cause a broad rectangular distribution for $V$. Of course this is not convenient if one wants to determine exact energies of emitted particles.

A solution for the problem was found by Frisch. He suggested to place a third electrode, which is called the Frisch grid, in between the anode and the cathode, on a suitable potential. This grid (figure 3.4) consists of thin steel wires, placed equidistant and parallel to each other and it divides the chamber into two parts with independent electric fields. The anode, which collects the electrons, is located in the first part of the chamber. In the second part, the ionisation happens, so the positive ions are located there. In order to avoid positive ions entering the first part, one should take care that there is no ionisation between the grid and the anode. In this way the anode is shielded for the positive ions and all the created electrons pass through the same potential difference and contribute equally to the anode signal.

With this setup the amplitude of the anode signal is only dependent on the number of electron ion pairs created by the incoming particle and therefore it is proportional to the energy of this particle:

$$V_{\text{anode}} = \frac{eN_0}{C} \sim E.$$  \hspace{1cm} (3.10)

3.4.1 Tuning of the ionisation chamber

First of all, a suited detection gas should be chosen. Of course, the drift velocity of the charges in the gas should be as high as possible, in order to increase the detection velocity of
the electrons. The drift velocity \( v \) of electrons is dependent on the ratio of the electric field \( E \) and the gas pressure \( P \) [40]. For the measurements performed in this work, 99.995% pure methane gas was chosen as detector gas. In figure 3.5 the electron drift velocity in methane and an argon-methane mixture is illustrated. It shows that the electron drift velocity for methane has a saturation effect which permits to work with different combinations of \( E \) and \( P \) without increasing collection time.

This leads to a first requirement for the setup of the ionisation chamber, using \( CH_4 \):

\[
E/P \geq 650 \left[ \frac{V}{atm.cm} \right].
\]  

(3.11)

Furthermore, one should take into account that a gas pressure which is too high can lead to recombination of the electrons and ions, resulting in signal loss.

Another important thing is the Frisch grid. The introduction of this grid with wires of a certain thickness, causes a leak of some of the electrons via these wires. In order to reduce this effect as much as possible, no electric field lines should end on these wires. This is the case if the following condition is fulfilled [42]:

\[
\frac{E_A}{E_C} = \frac{V_A - V_C}{d_{AG}} \frac{d_{CG}}{V_C - V_A} \geq 1 + \frac{2\pi}{g} \frac{g}{2\pi} \approx 2
\]  

(3.12)

where \( E_A \) and \( E_C \) are the electric fields at the anode and the cathode, \( d_{AG} \) and \( d_{CG} \) are the distances between anode and grid and between cathode and grid, respectively, \( r \) is the radius of the wires and \( g \) is the distance between two wires. The Frisch grid used in this work was made of wires with a radius of 0.05 mm, and interval distances of 1 mm between each other.
3.4. IONISATION CHAMBER

Figure 3.5: The electron drift velocity as a function of $E/P$ [43].

The ionisation chamber used for the experiments in this work operates as a $\Delta E$ detector, which means that the charged particles may not be completely stopped in this detector. The stopping happens in the subsequent E detector, in our case a surface barrier detector (see figure 4.3). Therefore one should take into account the range of the different particles one wants to detect, the range being dependent on the gas pressure and the material used. The range of particles can be calculated with the computer program SRIM (Stopping and Range of Ions in Matter)[44], for instance in methane, but also in silicon. As a consequence of this $\Delta E$-E setup, the positive ions are shielded only between the cathode and the grid, and not between grid, anode and surface barrier detector. Therefore some of the $\Delta E$ signal is lost, for which a correction is necessary. This is applied in the analyzing program, based on the work of Pommé et al. [45].

It is clear that the final tuning of the ionisation chamber is always a compromise. The setup of the ionisation chamber used for the spontaneous fission measurement of $^{244}$Cm is shown in table 3.1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>distance cathode-grid</td>
<td>26 mm</td>
</tr>
<tr>
<td>distance grid-anode</td>
<td>5 mm</td>
</tr>
<tr>
<td>voltage cathode</td>
<td>-1240 V</td>
</tr>
<tr>
<td>voltage anode</td>
<td>850 V</td>
</tr>
<tr>
<td>grid</td>
<td>0 V</td>
</tr>
<tr>
<td>pressure gas</td>
<td>1 atm + 0.2 bar</td>
</tr>
</tbody>
</table>

Table 3.1: Setup of the ionisation chamber for the $^{244}$Cm(SF) measurement.
3.5 Semiconductor detectors

The most used materials for semiconductor detectors are silicon and germanium. Quickly
after the development of these instruments in the late 1950's, they were frequently used
for the detection and energy determination of charged particles. The basic principle of
semiconductor detectors is the same as for a gaseous ionisation chamber. Instead of a
gas however, the medium is now a solid semiconductor material. The passage of ionising
radiation creates electron-hole pairs, instead of electron-ion pairs, which are then collected
by an electric field. The advantage of the semiconductor is that the average energy needed
to create such an electron-hole pair is about 10 times smaller than that required for gas
ionisation. Therefore the amount of ionisation produced for a given energy is one order of
magnitude greater, which results in a better energy resolution. Furthermore, semiconductor
detectors have a greater density and as a consequence they are compact in size.
Unfortunately, there are also some disadvantages concerning the use of semiconductor
detectors. Most of them, except silicon, lose their qualities when they are used at a
temperature which is too high. Hence an additional cooling system is required. They also
have a greater sensitivity to radiation damage, which causes defaults in the lattice. This
phenomenon limits their long term use, and requires sometimes the use of protecting foils.

3.5.1 Basic properties

Energy band structure

The basic structure of a semiconductor consists of a valence band, a so-called forbidden
energy gap and a conduction band. These energy bands are regions of many discrete
levels which are so closely spaced that they may be considered as a continuum, while the
forbidden energy gap is a region where there are no available energy levels at all. In figure
3.6 the band configuration of a semiconductor is shown, as well as for conductors and
insulators.

The highest energy band is the conduction band, electrons in this region are detached
from their parent atoms and free to roam about the entire crystal. The electrons in the
valence band however, are more tightly bound and remain associated to their respective
lattice atoms.

In insulators, the energy gap between the conduction and valence band is large. Applying
an external electric field, there will be no movement of electrons through the crystal and
consequently no current. For conductors on the other hand, there is an overlap between
the two energy bands and there are always electrons present in the conduction band. When
an electric field is applied, a current will flow. In between these two, the semiconductor
is situated. Only a few electrons are excited into the conduction band by thermal energy.
When an electric field is applied, a small current is observed.

Electron-hole pairs

Both materials used for semiconductor detectors, namely silicon and germanium, have
four valence electrons so that four covalent bonds are formed. As explained earlier, at
normal temperatures, the action of thermal energy can excite a valence electron into the conduction band leaving a hole in its original position. Then it is easy for a neighboring valence electron to jump from its bond to fill the hole. This now leaves a new hole in the neighboring position. In this way the hole appears to move through the whole crystal acting like a positive charge carrier, and constituting an electric current. Hence not only the free electrons in the conduction band contribute to the electric current in a semiconductor.

Electrons do not stay forever in the conduction band. An electron may recombine with a hole by dropping from the conduction band into an open level in the valence band with the emission of a photon. This process is called direct recombination. Impurities and structural defects in the lattice perturb the energy band structure by adding additional levels to the middle of the forbidden energy gap. These states may capture an electron from the conduction band, and then release it again after a certain holding time or it may capture also a hole during the holding time, which annihilates with the trapped electron. It is clear that these effects can influence the charge collection.

Doped semiconductors

In a pure semiconductor crystal, the number of holes is equal to the number of electrons in the conduction band. This balance can be disturbed by introducing a small amount of impurities in the crystal. In this way so called doped semiconductors are created. If the impurity has only three valence electrons, there will be not enough electrons to fill the valence band. Consequently, an excess of holes is created in the lattice, so the (positive) holes become the majority charge carriers. Such materials are referred to as p-type semiconductors. On the contrary, if the impurity has five valence electrons, extra electrons are available to enhance the conductivity of the semiconductor and to fill up holes which are normally formed. In such materials the current is mainly due to the movement of electrons and they are called n-type semiconductors. Sometimes it is useful to add both n- and p-type impurities to a semiconductor. Then they
can compensate each other, and one obtains a semiconductor with the same properties as an intrinsic semiconductor.

3.5.2 np-junction and depletion zone

The functioning of the semiconductor detectors used nowadays depends on the formation of a semiconductor junction, which occurs if a p-type semiconductor material is in contact with a n-type material. Because of the difference in the concentration of holes and electrons between the two materials, there will be a diffusion of holes towards the n-region and a similar diffusion of electrons towards the p-region. This creates an electric field and finally the diffusion process stops and there arises an equilibrium between the two opposite effects. Due to the electric field, there is a potential difference across the junction, which is known as the contact potential, being generally of the order of 1 V.

The region of changing potential is known as the depletion zone and it has the special property that there are no free charge carriers. Moreover, all free charges created into this zone by energetic radiation will be swept out by the electric field. Hence, the current through the junction is proportional to the ionisation and will be a measure for the energy of the incoming radiation.

Unfortunately, in general the intrinsic electric field will not be sufficiently large to provide an efficient charge collection and the thickness of the depletion zone will be too thin. The depletion depth can be enlarged by applying a reverse bias voltage to the junction, like a positive voltage to the n-side. This will also provide a more efficient charge collection.

Leakage current

Although a reversed bias diode is ideally nonconducting, there always flows a small fluctuating current through semiconductor junctions when voltage is applied. This current appears as noise at the detector output and it sets a limit on the smallest signal pulse height which can be observed. This leakage current has several sources, for instance the movement of holes from the n-region which are attracted across the junction to the p-side and electrons from the p-region which are similarly attracted to the opposite side. Another, more important source of the leakage current is through surface channels. This depends on many factors including the surface chemistry, the surrounding atmosphere, etc. Radiation can damage the detector, and cause defects in the crystal. In this way, the leakage current will increase and an increase of the applied voltage will be necessary to compensate for the losses due to the leakage current.

3.5.3 Silicon detectors: surface barrier detectors

Silicon is the most widely used semiconductor material for charged particle detection. It has the advantage of possible working at room temperature and wide availability. One of the disadvantages is their relatively small size, limited to a few ten’s of square centimeters. The most common silicon detectors for charged particle measurements are surface barrier type detectors. The detectors rely on the junction between a semiconductor and certain
3.5. SEMICONDUCTOR DETECTORS

metals, usually n-type silicon with gold or p-type silicon with aluminium. Such junctions are called Schottky contacts and they have a depletion zone like np-junctions. This depletion zone arises since the Fermi levels of both materials are not equal, which causes a lowering of the band levels in the semiconductor near the contact surface, as illustrated in figure 3.7.

![Diagram of Schottky barrier junction and schematic of surface barrier detector](image)

Figure 3.7: Formation of a Schottky barrier junction (left) and a schematic diagram of a surface barrier detector (right).

It is simpler to fabricate surface barrier detectors (SBD) than diffused junctions. Indeed, it is sufficient to first etch the silicon surface and then depositing a thin layer of gold by evaporation. The junction is then mounted in an insulating ring with metallized surfaces for electrical contact (figure 3.7).

Surface barrier detectors can be made with varying thickness and depletion zones. If the detector is not too thick, a fully depleted detector is possible, which means the depletion zone extends through the entire thickness of the silicon wafer. Such detectors are very useful as transmission detectors in order to measure the energy deposition of passing charged particles.

A disadvantage of surface barrier detectors is their sensitivity to light. The thin gold covering is insufficient to stop ambient light. Therefore they should be used and stored in the dark. Moreover, the surface is very sensitive and touching should be avoided.
Chapter 4

Experimental setup

In order to investigate the ternary fission of curium and californium isotopes, two types of measurements are performed: neutron induced fission measurements and spontaneous fission measurements.

4.1 Location of the measurements

4.1.1 Neutron induced fission measurements

The neutron induced fission measurements were performed at the High Flux Reactor of the Institut Laue-Langevin (ILL) in Grenoble, France [46]. The High Flux Reactor, shown in figure 4.1, produces the most intense neutron flux in the world for research applications: $1.5 \times 10^{15}$ neutrons per second per cm$^2$, with a thermal power of 58.3 MW. There is only one fuel element in the reactor, located in the center of a tank containing the heavy water moderator. Cooling and moderation happens by heavy water circulation. The moderator reflects part of the thermal neutrons back towards the fuel element, in this way creating a critical reactor. The reactor normally works continuously for 50-day cycles, followed by a shut-down to change the fuel element. No electricity is produced at the ILL, the reactor is build only for scientific experiments with neutrons.

More specifically, our measurements were performed at the PF1b cold neutron guide. It is located in the neutron guide hall I, at the end of a 76 m long neutron guide [47]. This neutron guide is used to transport the cold neutrons from the neutron source to the instrument PF1b. Due to the small curvature of the neutron guide, essentially all fast neutrons and $\gamma$-rays produced in the reactor are removed, resulting in excellent background conditions. The mean neutron wavelength at this position is about 4.0 to 4.5 Å, with the average neutron energy being 5.4 meV. Depending on the setup for the different measuring campaigns, the neutron flux at the sample position varied from about $2 \times 10^9$ neutrons/s.cm$^2$ to $10^{10}$ neutrons/s.cm$^2$. 
4.1.2 Spontaneous fission measurements

For the measurements of the spontaneously fissioning nuclei, no neutron beam is required. In fact this type of measurements could be performed everywhere. However, due to the radioactivity of the samples used, sample handling and experiments should happen only in a controlled area. These facilities are available in the Institute for Reference Measurements and Materials (IRMM) in Geel, Belgium.

4.2 General setup

In principle, each measurement consists of two main parts:

1. the identification of the different ternary particles with an appropriate ΔE-E telescope detector and the subsequent determination of the energy distributions and counting rates of these particles.

2. the determination of the binary fission counting rate with the E detector.

During the measurements, the active side of the sample is always covered with a polyimide foil of about 30 μg/cm² thick in order to avoid contamination of the chamber. Ternary fission particles and binary fission fragments hardly lose energy in this foil, but the low-energetic recoil nuclei are stopped. It is necessary for the ternary fission measurements to cover the detectors facing the sample with a thin aluminium foil (25 or 30 μm) in order to stop the radioactive decay α’s and the fission fragments.

For a binary fission measurement the protecting aluminium foil and the ΔE detector are removed. In the measurement with the ionisation chamber the latter is realised by pumping the methane gas out of the chamber, which then becomes a vacuum chamber. In
the measurements with surface barrier detectors, the \( \Delta E \) detector is replaced by an empty frame, strictly maintaining the detection geometry.

### 4.2.1 Neutron induced fission measurements

For all neutron induced fission measurements, the sample was mounted in the center of a vacuum chamber, at an angle of 45° with respect to the incoming neutron beam. There is the possibility to install a \( \Delta E \)-E telescope detector, consisting of two surface barrier detectors, on each side of the sample. Hence, one telescope is facing the sample, the other one faces the backing of the sample. The detectors are positioned perpendicular to the incoming neutron beam, which is illustrated in figure 4.2.

![Diagram of neutron induced fission measurements](image)

Figure 4.2: Schematic view of the vacuum chamber used for the neutron induced fission measurements.

The whole of sample and detectors is placed in a vacuum chamber, otherwise the emitted particles would lose energy through ionisation of the air.

The chamber has two thin windows, made of aluminium. A lithium collimator with a diameter of 12 mm is installed in front of the entrance window in order to avoid neutrons to irradiate the detectors.

Around the chamber a shielding is placed, consisting of boron carbide and lead. The boron carbide is necessary in order to avoid activation of the lead. As \( ^6 \text{Li} \), \( ^{10} \text{B} \) has a large neutron absorption cross section in the concerned neutron energy region, the boron carbide stops the (scattered) neutrons and the lead shields the environment from \( (n,\gamma) \) rays produced along the beamline.

### 4.2.2 Spontaneous fission measurements

In the case of spontaneous fission measurements, no neutron beam is used, so the sample can be placed parallel to the detectors. This geometry is slightly better than the geometry in the neutron induced fission measurements, since now a smaller energy loss in the sample will occur. For the spontaneous fission measurements, two different setups were used. A
first one with the ionisation chamber as $\Delta E$ detector and a surface barrier detector as E detector [45], shown in figure 4.3. A continuous methane gas flow is going through the chamber (see section 3.4).

![Diagram of ionisation chamber](image)

Figure 4.3: Schematic view of the ionisation chamber used for the $^{244}\text{Cm}$ spontaneous fission measurement, with C = Cathode; G = Grid; A = Anode; SBD = Surface Barrier Detector; PI = Polyimide.

The other setup used is basically the same as for the neutron induced fission measurements, consisting of a vacuum chamber, however the sample is placed parallel to the telescope detectors each consisting of two surface barrier detectors, as becomes clear from figure 4.4.

![Diagram of vacuum chamber](image)

Figure 4.4: Schematic view of the vacuum chamber used for the spontaneous fission measurements.
4.3 Data acquisition

A scheme of the electronic setup is shown in figure 4.5. As explained in the previous chapter, a $\Delta E$-E telescope detector system is used, in order to be able to identify the different incoming particles. Both detectors are placed behind each other, so that a particle that passes through detector $\Delta E$, finally ends up in detector E.

The weak signals coming from both detectors first have to be amplified by a pre-amplifier (PA) and then they are sent through an amplifier in order to obtain signals between 0 and 10 Volt. This amplified signal of the E detector is sent to the Timing Single Channel Analyzer (TSCA) and the Analog to Digital Converter (ADC). The TSCA sorts incoming analog signals according to their amplitudes. It contains both lower and upper level thresholds, and only signals which fall between these two levels generate a pulse from the TSCA. The signal coming from the TSCA is send to both ADC’s where it opens a gate. The amplifier signal is positioned in the center of the corresponding gate by adjusting a time delay.

In some of the experiments a linear gate module was added to the $\Delta E$ chain (indicated by the dashed line in figure 4.5). Indeed, a problem can occur due to the high sample activity, resulting in high counting rates in the $\Delta E$ detector causing a dead time in the corresponding ADC. This could be avoided by adding the linear gate, which was steered by a signal from the E detector, rejecting the largest part of the radioactive decay events. Finally only signals accepted in the ADC’s are written and stored in a PC, using a Labview based program [48]. In this way, the analysis of the data can happen afterwards.

Figure 4.5: Schematic view of the electronic setup used in all ternary fission measurements. For some isotopes a linear gate was added to avoid ADC($\Delta E$) dead time (dashed line), with DAQ = Data Acquisition.
4.4 Calculation of energy losses

Ternary particles are emitted by the source and detected by a detector. However, before this detection, they lose part of their energy in the source itself, and also in additional protecting foils like the polyimide foil and the aluminium foil, or the backing of the source. In our measurements, the sources are very thin, so the energy loss in the source itself can be neglected.

The energy losses of the particles are calculated with SRIM [44]. As an example, the result of such a calculation is given in figure 4.6, which shows the energy loss of α-particles in an aluminium foil of 30 μm thickness. The spectrum of the α-particles needs to be corrected for this energy loss. Based on the values calculated by SRIM, the energy loss of a particle with an arbitrary energy can be calculated through linear interpolation between two calculated values.

![Energy loss graph](image)

Figure 4.6: The energy loss of α-particles in a 30 μm thick aluminium foil, calculated with SRIM.

The corrections for the energy losses of the ternary particles in the polyimide and aluminium foil are integrated in the software to analyse the data. Therefore, the method used is based on a relation similar to equation (3.8).

Suppose one has a particle with an initial energy $E_0$ and a foil with a known thickness $T_f$, characterized by the constant $a_f$, which is material and particle specific, as mentioned before. The emitted particle passes through the foil and then it is detected in the ΔE-E detection system. In that case, the initial energy of the particle can be determined, based on the ΔE and E signals and the values for $T_f$ and $a_f$.

$E + \Delta E$ is the energy of the particle after passing through the foil. Using formula 3.8 one can write:

$$E + \Delta E = \left( E^{0.73} + T/a \right)^{1/1.73} \quad (4.1)$$

with $T$ the thickness of the ΔE detector. If now also the foil is taken into account, the following relation is obtained:

$$E_0 = \left( (E + \Delta E)^{0.73} + T_f/a_f \right)^{1/1.73}. \quad (4.2)$$
4.5. DETECTOR CALIBRATION

A combination of equations 4.1 and 4.2 gives:

$$E_0 = \left[ E^{1.73} + T/a(1 + z) \right]^{1/1.73}, \text{with} \quad z = \frac{T_f/a_f}{T/a}. \quad (4.3)$$

It is possible that more than one foil is used, for instance an aluminium foil together with a polyimide foil. Applying this to formula 4.3, one obtains:

$$E_0 = \left[ E^{1.73} + \frac{T}{a} \left( 1 + \frac{T_{Pl}/a_{Pl}}{T/a} + \frac{T_{Al}/a_{Al}}{T/a} \right) \right]^{1/1.73}. \quad (4.4)$$

Values for $a$ for the different particles in the different materials have been calculated. This has been done calculating the range for different energies with SRIM. Based on relation 3.7 a value for $a$ is obtained. Figure 4.7 illustrates the range of different ternary particles in silicon, together with the values obtained for $a$.

![Graph showing the range of different ternary particles in silicon](image)

Figure 4.7: The range of different ternary particles in silicon, together with the values obtained for $a$.

4.5 Detector calibration

An incoming charged particle creates a pulse in a detector. This pulse is amplified and then converted into an integer number, which is called the channel number ($K$). Each ADC spectrum consists of 4096 channels. The energy of the incoming particle that is lost in the detector should be determined. Therefore the relation between the energy and the channel numbers has to be found. As mentioned in section 3.3, there exists a linear relation between the energy and the created pulse, so also between the energy and the channel number.

The energy calibration of the detectors is done with some sources, which emit $\alpha$-particles with well-known energies. In this way, the relation between the energy and the channel number is known for some points. A linear fit to these points leads to the calibration relation.
Neutron induced fission measurements

The energy calibration of all the detectors used for the neutron induced fission measurements was done based on the radioactive $\alpha$-decay energies of

- $^{237}$Np with $E_\alpha = 4.790$ MeV
- $^{241}$Am with $E_\alpha = 5.486$ MeV
- $^{244}$Cm with $E_\alpha = 5.805$ MeV

and on the well-known nuclear reactions

- $^{10}$B$(n,\alpha)^7$Li$_0$ and $^{10}$B$(n,\gamma\alpha_1)^7$Li$_1$ with $E_{Li1} = 0.830$ MeV, $E_{Li0} = 1.007$ MeV, $E_{\alpha1} = 1.483$ MeV, $E_{\alpha0} = 1.789$ MeV
- $^6$Li$(n,\alpha)t$ with $E_\alpha = 2.055$ MeV and $E_t = 2.720$ MeV.

In figure 4.8 the $^{10}$B$(n,\alpha)$ spectrum is shown for a $\Delta E$ detector with a thickness of 49.8 $\mu$m. The points used for the calibration are indicated with a cross. Combining these points with the three points of the $\alpha$-decay and the two points of the $^6$Li reaction, a nice linear fit can be made, which is illustrated in figure 4.9. In this way, a calibration relation of the following form is obtained:

$$ E = (a \pm \delta a) + (b \pm \delta b).K \ [MeV]. \quad (4.5) $$

It must be noted that not always all these points were available for the calibration of a certain detector. In principle it is sufficient to have two points to determine the calibration relation.

![Figure 4.8: $^{10}$B$(n,\alpha)^7$Li spectrum obtained with a $\Delta E$ detector. The points used for the calibration are indicated with a cross.](image-url)
Figure 4.9: Linear fit through 8 calibration points in order to obtain the calibration relation.

Spontaneous fission measurements

The energy calibration of all the detectors used for the spontaneous fission measurements was done based on the radioactive $\alpha$-decay energies mentioned above, together with those of

- $^{147}$Sm with $E_\alpha = 2.235$ MeV
- $^{238}$U with $E_\alpha = 4.198$ MeV.
Chapter 5
Cm isotopes

In this chapter the neutron induced ternary fission measurement of $^{243}\text{Cm}$ and the spontaneous ternary fission measurement of $^{244}\text{Cm}$ are presented in detail together with their results. An overview of the results of the other curium isotopes measured by our research group is given in section 5.3, together with a literature survey for the fissioning systems $^{241,246,248}\text{Cm}$.

5.1 $^{243}\text{Cm}(n,f)$

5.1.1 Characteristics of the sample

The $^{243}\text{Cm}$ sample used for the neutron induced fission measurement was prepared at the Russian Federal Nuclear Center (RFNC) in Arzamas, by using the technique of electrodeposition. A spot of curium oxide with a diameter of 15 mm was deposited on a 30 $\mu$m thick aluminium foil, which is called the backing of the sample. The $^{243}\text{Cm}$ sample has a mass of 2 $\mu$g and at the moment of preparation its activity was 3.7 MBq. Some nuclear characteristics of the $^{243}\text{Cm}$ isotope can be found in table 5.1.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}(\alpha)$[a][38]</th>
<th>$T_{1/2}(\text{SF})$[a][38]</th>
<th>$\sigma(n_{th}, f)$[b][49]</th>
<th>$S_n$ [MeV] [38]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{243}\text{Cm}$</td>
<td>29.1</td>
<td>5.50 E+11</td>
<td>613.51</td>
<td>6.801</td>
</tr>
<tr>
<td>$^{244}\text{Cm}$</td>
<td>18.1</td>
<td>1.32 E+7</td>
<td>1.038</td>
<td></td>
</tr>
<tr>
<td>$^{245}\text{Cm}$</td>
<td>8.50 E+3</td>
<td>1.40 E+12</td>
<td>2.14 E+3</td>
<td>6.458</td>
</tr>
<tr>
<td>$^{246}\text{Cm}$</td>
<td>4.73 E+3</td>
<td>1.81 E+7</td>
<td>0.144</td>
<td></td>
</tr>
<tr>
<td>$^{247}\text{Cm}$</td>
<td>1.56 E+7</td>
<td>(*)</td>
<td>111.29</td>
<td>6.213</td>
</tr>
<tr>
<td>$^{248}\text{Cm}$</td>
<td>3.70 E+5</td>
<td>4.05 E+6</td>
<td>0.087</td>
<td></td>
</tr>
</tbody>
</table>

(*) SF not observed

Table 5.1: Nuclear characteristics of the different curium isotopes.

$^{243}\text{Cm}$ undergoes radioactive $\alpha$-decay ($T_{1/2} = 29.1$ a), emitting $\alpha$-particles with a most probable energy of 5.785 MeV. Because of this $\alpha$-decay, $^{239}\text{Pu}$ is rapidly growing in the
sample material, but it was chemically removed before the sample preparation. Nevertheless, at the moment of the measurements, some ingrow of $^{239}$Pu is already present in the sample again, and a correction has to be made. Moreover, due to the radioactive $\alpha$-decay of $^{241}$Cm, a small amount of $^{239}$Pu is growing in the sample material. Details on the isotopic composition at the different times of the experiments can be found in table 5.2.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Isotopic analysis</th>
<th>Preparation sample</th>
<th>$1^{st}$ experiment</th>
<th>$2^{nd}$ experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Cm</td>
<td>98.35</td>
<td>98.42</td>
<td>93.63</td>
<td>90.85</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>1.59</td>
<td>1.51</td>
<td>1.40</td>
<td>1.33</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>0.06</td>
<td>0.07</td>
<td>0.07</td>
<td>0.07</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>0</td>
<td>0</td>
<td>4.79</td>
<td>7.57</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>0</td>
<td>0</td>
<td>0.11</td>
<td>0.18</td>
</tr>
</tbody>
</table>

Table 5.2: Isotopic composition (in atomic percent) of the $^{241}$Cm sample at the different times of the experiments.

### 5.1.2 Overview of the experiments

Two different measurement campaigns were performed at the PF1b neutron guide in order to examine the neutron induced fission of $^{241}$Cm. For the first measurement, the chamber was placed behind an additional neutron guide of 2.5 m length and the thermal equivalent neutron flux at the sample position was about $2 \times 10^9$ neutrons/s.cm$^2$. The second measurement was performed in another configuration of the PF1b beam. Here the chamber was installed behind an additional neutron guide of 4 m length, yielding a thermal equivalent neutron flux of about $10^{10}$ neutrons/s.cm$^2$.

For both measurements, two $\Delta E$-$E$ surface barrier telescope detectors were placed on both sides of the sample, perpendicular to the incoming neutron beam. Their characteristics can be found in table 5.3. With this setup the detection geometry was about 2%.

<table>
<thead>
<tr>
<th>$^{241}$Cm</th>
<th>$\Delta E$</th>
<th>$E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>telescope 1</td>
<td>300</td>
<td>29.8</td>
</tr>
<tr>
<td>telescope 2</td>
<td>300</td>
<td>49.8</td>
</tr>
<tr>
<td>telescope 3</td>
<td>300</td>
<td>31.0</td>
</tr>
<tr>
<td>telescope 4</td>
<td>300</td>
<td>41.0</td>
</tr>
</tbody>
</table>

Table 5.3: Characteristics of the surface barrier telescope detectors used for the $^{241}$Cm neutron induced fission measurements. The indicated resolution is for 5.5 MeV $\alpha$-particles.
5.1. $^{243}$Cm(N,F)

For the first measurement telescope 1 and 2 are used simultaneously, for the second one telescope 3 and 4. Telescope 1 (3) is facing the sample and is covered with a protecting aluminium foil of 30 $\mu$m for the ternary fission measurement. At this side a binary fission measurement can be performed. Telescope 2 is facing the backing of the sample, hence no protecting aluminium foil is needed. Therefore also no binary fission measurement is possible at this side, since all fission fragments are stopped by the backing foil.

The thickness of this aluminium backing foil is 30 $\mu$m. However, the angle between the sample with backing and the detector is 45$^\circ$. Hence the particles pass through 30 $\mu$m/cos 45$^\circ$ = 42.43 $\mu$m aluminium on the average, instead of 30 $\mu$m.

The telescope detectors 1 and 3 have the right characteristics to detect simultaneously $\alpha$-particles and $^6$He particles. These particles are stopped completely in the 500 $\mu$m E detector and a good separation between $\alpha$ and $^6$He particles is achieved.

On the other hand, telescope detectors 2 and 4 are chosen to detect tritons and $\alpha$-particles. To stop the tritons, an E detector of 1500 $\mu$m is needed. The thicker $\Delta E$ detector is necessary to obtain a reasonable separation between the tritons and the background.

5.1.3 Binary fission

As explained in section 4.2, two measurements are needed in order to determine the absolute emission probability of the ternary particles. First the binary fission spectrum of $^{243}$Cm has to be measured. This measurement is performed with the E detector from telescope 1 (table 5.3). The $\Delta E$ detector is replaced by an empty frame with exactly the same geometry.

The result of the measurement with open neutron beam is shown in figure 5.1. To be sure about the contribution of the background, an additional measurement with closed neutron beam was performed, which is illustrated in figure 5.2.

![Binary fission spectrum for $^{243}$Cm(n,f) with open neutron beam.](image)
Figure 5.2: Background binary fission spectrum for $^{243}\text{Cm}(n,f)$ with closed neutron beam.

The peak in the beginning of the spectrum is due to the radioactive $\alpha$-decay and appears when two or more $\alpha$-particles arrive at the same moment on the detector. This phenomenon is called alpha pile-up. It is clear these signals should be removed from the spectrum to obtain the right number of fission events. From figure 5.2 one can determine where to cut in the binary fission spectrum to be sure to remove this pile-up. Hence channel 425 is chosen as cut off. Due to this cut off an extrapolation of the remaining spectrum is necessary to obtain the correct fission yield. This is done by fitting an exponential function to part of the spectrum and the final extrapolated spectrum is illustrated in figure 5.3.

Figure 5.3: Extrapolated binary fission spectrum for the neutron induced fission measurement of $^{243}\text{Cm}$.
Now the number of binary fission fragments is obtained by integrating the area under the curve. Since in every fission event, two fission fragments are released, the number of binary fission events is equal to half of the detected counts. Taking into account the measuring time, the binary fission yield is obtained.

From figure 5.2 it is clear that the contribution due to spontaneous fission and electronic background was negligible.

The results of the measurement are given in table 5.4.

<table>
<thead>
<tr>
<th>ΔT beam off [s]</th>
<th>600</th>
</tr>
</thead>
<tbody>
<tr>
<td>ΔT beam on [s]</td>
<td>4850</td>
</tr>
<tr>
<td>number of fission fragments after extrapolation and dead time correction</td>
<td>2180540 ± 1477</td>
</tr>
<tr>
<td>number of fission events</td>
<td>2203469 ± 19788</td>
</tr>
<tr>
<td>binary fission yield [s⁻¹]</td>
<td>1101735 ± 9894</td>
</tr>
<tr>
<td></td>
<td>227.16 ± 2.04</td>
</tr>
</tbody>
</table>

Table 5.4: Results of the binary fission measurement of $^{243}$Cm ($\Delta T = \text{measuring time}$).

In table 5.4 the number of fission fragments is given after dead time correction. Indeed, during the binary fission measurement with open neutron beam, some dead time occurred. To correct for this, a measurement with a counter was performed in parallel, yielding a dead time of about 0.6%.

The uncertainties given in table 5.4 are a combination of statistical and systematical uncertainties:

- the statistical fluctuation on the number of counts $N$, which is equal to $\sqrt{N}$.
- the uncertainty due to the choice of the channel number where the cut off is made.
- the uncertainty due to the extrapolation that is made on the left side of the spectrum.
- the uncertainty due to the dead time correction.

### 5.1.4 Ternary fission

To continue with the neutron induced fission measurement of $^{243}$Cm, the different ternary particles have to be measured. The identification of the LRA particles, tritons and $^6$He particles happens with an appropriate $\Delta E$-$E$ telescope detector. Subsequently the parameters of the energy distributions and the counting rates are determined.

Similar to the binary fission measurement, a background measurement without neutron beam has been performed for the ternary fission measurements. Again it was concluded that the background was negligible.
Alpha particles

For every ternary fission event, two different values are registered in coincidence, namely the energy ΔE lost in the ΔE detector and the energy E deposited in the E detector. These spectra are always recorded and, as an example, they are illustrated in the upper part of figure 5.4, for the first measurement with telescope 1, suited for the detection of α-particles.

Figure 5.4: ΔE and E spectra for the $^{243}\text{Cm(n,f)}$ measurement with telescope 1. Upper part: total ΔE and E spectra, lower part: ΔE and E spectra with a selection window on the α-particles.

The energies lost in both detectors can be plotted in a 2-dimensional graphic, with ΔE on the Y-axis and E on the X-axis, as can be seen in figure 5.5 (left).

Out of the values for ΔE and E, a 1-dimensional spectrum of $T/a$ values can be build, which is shown in figure 5.6 (upper part, right). The $T/a$-values can also be plotted as a function of the total energy $E_{\text{tot}}$ (after correction for the energy losses) of the particle. Doing like this, figure 5.6 (upper part, left) is obtained for the measurement with the ΔE detector of 29.8 μm.

Due to the definition of $T/a$, this value remains constant on the average for a certain particle. In this way the different ternary particles can be easily identified by putting a window on the 2-dimensional spectrum $E_{\text{tot}} - T/a$ and isolate for example only the α-particles, which leads to the lower part of figures 5.4 and 5.6 and the right part of figure 5.5.

With the knowledge of the value for $a$ in the case of α-particles (see figure 4.7) and
the thickness of the ΔE detector from telescope 1, one can calculate a theoretical value for $T/a$:

$$T/a = 29.8/1.199 = 24.85. \quad (5.1)$$

Making a Gaussian fit to the $T/a$-spectrum obtained after selection of the α-particles, an experimental value of exactly 24.85 is obtained. Hence a perfect agreement exists between the theoretical and experimental $T/a$-value, which illustrates that the calibration of the detectors was done very well.

In figure 5.7, the total energy spectrum for the ternary α-particles is shown, after correction for the energy loss.

It is clear from this picture that not all the α-particles are detected. There is an energy threshold below which no particles can be detected. This lower limit is caused by the electronic noise of both detectors and the energy loss in the ΔE detector and the protecting foils. For telescope 1 this limit is about 10.5 MeV.

As described in section 2.4, a Gaussian fit to the data is performed in order to determine the parameters of the energy distribution and the LRA yield. For the α-particles the fit takes into account only data above 12.5 MeV, in order to avoid the influence of the non-Gaussian low energy tail. The result of this Gaussian fit is shown in figure 5.7 and the parameters of the curve are given in table 5.5.

As mentioned in section 5.1.1, a correction has to be made for the ingrow of $^{239}$Pu in the sample. This can be done in the following way. From table 5.2, it can be seen that at the moment of the first experiment there is about 4.79% of $^{239}$Pu present in the sample. The ternary particles that have been measured then, are in fact the sum of the ternary particles emitted by $^{243}$Cm and those emitted by $^{239}$Pu:

$$LRA = LRA_{Cm} + LRA_{Pu} \quad (5.2)$$

and the same is true for the binary fission events:

$$B = B_{Cm} + B_{Pu} \quad (5.3)$$
with

\[
\frac{B_{Cm}}{B_{Pu}} = \frac{N_{Cm}\sigma_{Cm}^{Pu}}{N_{Pu}\sigma_{Pu}^{Pu}}
\]  

(5.4)

where \(N\) is the number of atoms and \(\sigma_{n,\alpha}^{Pu}(5.4\,\text{meV}) = 1.58 \times 10^3\,\text{b}\) [49]. For \(^{243}\text{Cm}\) the cross section at 5.4 meV is \(1.33 \times 10^3\,\text{b}\) [49]. Taking into account these equations, the measured value LRA/B can be rewritten:

\[
LRA/B = \frac{(LRA/B)_{Cm}}{1 + \frac{B_{Pu}}{B_{Cm}}} + \frac{(LRA/B)_{Pu}}{B_{Pu} + 1}
\]  

(5.5)

where LRA/B = \((2.09 \pm 0.07)\times10^{-3}\) has been adopted for \(^{239}\text{Pu}(n_{th},f)\) [58]. Due to the very small difference between thermal energy (25.3 meV) and 5.4 meV, this value for LRA/B at thermal energy can be used without problems. In equation 5.5, the only remaining unknown quantity is \((LRA/B)_{Cm}\), which is exactly the corrected value of LRA/B we wanted to determine (see table 5.5).

The ingrow of \(^{239}\text{Pu}\) will also have a small influence on the results for the characteristics of the energy distribution: \(\langle E\rangle\) and FWHM. The values for \(^{239}\text{Pu}\) used to calculate this correction are \(\langle E\rangle = 15.9 \pm 0.1\,\text{MeV}\) and FWHM = \(10.3 \pm 0.2\,\text{MeV}\) [2]. The corrected values can be found in table 5.5.
5.1. $^{243}$CM($N,F$)

![Figure 5.7: Ternary $\alpha$ energy distribution and corresponding Gaussian fit for $^{243}$Cm(n,f) obtained with telescope 1.](image)

<table>
<thead>
<tr>
<th>$\Delta T$ [s]</th>
<th>number of ternary $\alpha$'s counting rate [s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>225616</td>
</tr>
<tr>
<td>before $^{239}$Pu correction</td>
<td>123317 ± 3700</td>
</tr>
<tr>
<td>after $^{239}$Pu correction</td>
<td>0.547 ± 0.016</td>
</tr>
<tr>
<td>LRA/B [$10^{-5}$]</td>
<td>2.41 ± 0.08</td>
</tr>
<tr>
<td>$\langle E\rangle$ [MeV]</td>
<td>16.14 ± 0.14</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.33 ± 0.20</td>
</tr>
</tbody>
</table>

Table 5.5: Overview of the results of the ternary $\alpha$ measurement of $^{243}$Cm with telescope 1.

The uncertainties given on the values in table 5.5 are a combination of statistical and systematical uncertainties:

- the statistical fluctuation on the number of counts $N$, which is equal to $\sqrt{N}$
- the uncertainties on the calibration relations
- the uncertainty due to the selection of the particles
- the uncertainty due to the Gaussian fit

During the second measurement campaign, the ternary $\alpha$-particles were detected again, this time with telescope 3. The parameters of this alpha spectrum are given in table 5.6, and the total energy spectrum with Gaussian fit, starting at 12.5 MeV, is shown in figure 5.8.

Again a correction for the ingrow of $^{239}$Pu has to be made, taking into account the increased
\[
\begin{array}{|c|c|c|}
\hline
\Delta T [s] & \text{number of ternary } \alpha \text{'s} & 269556 \\
& \text{counting rate } [\text{s}^{-1}] & 266538 \pm 4798 \\
& & 0.989 \pm 0.018 \\
\hline
\langle E \rangle \text{ [MeV]} & \text{before } ^{239}\text{Pu correction} & 16.11 \pm 0.14 \\
\text{FWHM [MeV]} & \text{after } ^{239}\text{Pu correction} & 16.13 \pm 0.14 \\
\hline
\end{array}
\]

Table 5.6: Overview of the results of the ternary \( \alpha \) measurement of \(^{243}\text{Cm}\) with telescope 3.

![Graph](graph.png)

Figure 5.8: Ternary \( \alpha \) energy distribution and corresponding Gaussian fit for \(^{243}\text{Cm}(n,f)\) obtained with telescope 3.

The amount of \(^{239}\text{Pu}\) as mentioned in table 5.2. The corrected values can be found in table 5.6 as well.

In order to obtain a general result for the characteristics of the energy distribution of the alpha particles, their average energy and FWHM is determined as a weighted average of both results from tables 5.5 and 5.6. The absolute emission probability LRA/B has been determined only once, in the first experiment, hence no weighted average has to be calculated. In table 5.7 the general results for ternary alpha particles in the neutron induced fission of \(^{243}\text{Cm}\) are shown.

**Tritons**

Telescope 2 and 4 were used for the detection of ternary triton particles. With these detectors no binary fission measurement was performed, hence a relative triton emission probability \( t/LRA \) has to be determined, together with the parameters of the energy distribution of the tritons.
Table 5.7: General results of the ternary $\alpha$, triton and $^6\text{He}$ neutron induced fission measurement of $^{243}\text{Cm}$.

The first measurement was done with telescope 2, with a thickness of 49.8 $\mu$m for the $\Delta E$ detector. The 2-dimensional $E_{\text{tot}} - T/a$ spectrum and the 1-dimensional $T/a$ spectrum are plotted in figure 5.9.

![Figure 5.9: 2-dimensional $E_{\text{tot}} - T/a$ spectrum and corresponding $T/a$ spectrum for the $^{243}\text{Cm}$ neutron induced fission measurement with telescope 2.](image)

A selection of the ternary alpha particles and tritons was made in the 2-dimensional $E_{\text{tot}} - T/a$ spectrum, based on their theoretical value for $T/a$:

$$T/a(\alpha) = 49.8/1.199 = 41.53$$

$$T/a(t) = 49.8/5.898 = 8.44.$$

These values are in good agreement with the experimentally determined values of $T/a$, namely:

$$T/a(\alpha) = 41.80$$

$$T/a(t) = 8.48.$$

Both selections are shown in figure 5.10. As can be seen in figure 5.9, there is a background contribution at the position of the tritons. This background consists of different contributions, like e.g. tails of the deuteron and proton distributions. In most of the cases, the
selection of the tritons happens in the 1-dimensional $T/a$ spectrum. Then two corrections can be needed: first a correction due to the background, which will decrease the number of tritons and on the other hand a possible correction due to an incomplete $T/a$ peak, which will increase the number of tritons. Since however in the case of $^{243}$Cm, the selection of the tritons is done in the 2-dimensional $E_{tot} - T/a$ spectrum in order to obtain a better total energy spectrum, the background could be eliminated already at that time. Therefore no correction on the ternary triton counting rate will be needed.

The total energy spectrum of the $\alpha$-particles is plotted in figure 5.11 (left). In this case the detection limit is about 14 MeV. Therefore the Gaussian fit is started from 14 MeV. The parameters of the fit can be found in table 5.8.

In principle only the alpha counting rate will be used to calculate $t/LRA$. Nevertheless, it is interesting to compare the values for the average energy and the FWHM obtained with telescope 2 with those measured with telescope 1 and 3 (table 5.7). As a conclusion one can say that within the uncertainties they are in good agreement with each other.
5.1. $^{243}\text{Cm}(N,F)$

<table>
<thead>
<tr>
<th></th>
<th>Telescope 2</th>
<th>Telescope 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>225616</td>
<td>160325</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>16.22 ± 0.13</td>
<td>15.99 ± 0.17</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.65 ± 0.32</td>
<td>10.53 ± 0.41</td>
</tr>
<tr>
<td>number of ternary $\alpha$’s</td>
<td>120778 ± 4590</td>
<td>153548 ± 4607</td>
</tr>
<tr>
<td>counting rate [s$^{-1}$]</td>
<td>0.535 ± 0.020</td>
<td>0.958 ± 0.029</td>
</tr>
</tbody>
</table>

Table 5.8: Overview of the results of the ternary $\alpha$ measurement of $^{243}\text{Cm}$ with telescopes 2 and 4.

The 49.8 $\mu$m thick $\Delta E$ was chosen especially to measure the tritons. A Gaussian fit is performed to all triton data points, starting at 6.5 MeV. The resulting $E_{\text{tot}}$ spectrum is shown in figure 5.12 (left). The fit yields an average energy and FWHM as well as the triton counting rate, noted in table 5.9.

![Figure 5.12: Ternary triton energy distributions for the $^{243}\text{Cm}(n,f)$ measurement obtained with telescope 2 (left) and telescope 4 (right).](image)

The second experiment to measure tritons, performed with telescope 4, can be described in an analogue way as the previous one.

After selection of the LRA particles, a Gaussian fit to all data points was performed in order to obtain the yield of the ternary $\alpha$-particles. The spectrum is plotted in figure 5.11 (right), and the results are given in table 5.8. Figure 5.12 (right) shows the energy distribution for the tritons, which have been selected in the 2-dimensional $E_{\text{tot}} - T/\alpha$ spectrum. A fit to all the data points yields an average energy and FWHM as well as a triton counting rate (table 5.10).

Again the results have to be corrected for the ingrow of $^{239}\text{Pu}$. The correction on the relative emission probability $t/LRA$ can be done in a similar way as explained for LRA/B. A value $t/LRA = (6.79 \pm 0.20)\%$ has been adopted for $^{239}\text{Pu}$ [58]. The correction on $t/LRA$ is done for both measurements with telescope 2 and 4, taking into account the corresponding amount of $^{239}\text{Pu}$ present in the sample. Also the parameters of the energy
\[ \begin{array}{|c|c|c|}
\hline
\Delta T [s] & 225616 \\
number of tritons counting rate [s\(^{-1}\)] & 9787 \pm 1107 \\
0.0434 \pm 0.0049 & \\
\hline
\hline
before \(^{239}\)Pu correction & after \(^{239}\)Pu correction \\
\hline
t/LRA [%] & 8.10 \pm 0.97 \\
\langle E \rangle [\text{MeV}] & 7.98 \pm 0.44 \\
\text{FWHM} [\text{MeV}] & 8.26 \pm 0.65 \\
\hline
\end{array} \]

Table 5.9: Overview of the results of the ternary triton measurement of \(^{243}\)Cm with telescope 2.

\[ \begin{array}{|c|c|c|}
\hline
\Delta T [s] & 160325 \\
number of tritons counting rate [s\(^{-1}\)] & 11629 \pm 814 \\
0.0725 \pm 0.0051 & \\
\hline
\hline
before \(^{239}\)Pu correction & after \(^{239}\)Pu correction \\
\hline
t/LRA [%] & 7.57 \pm 0.58 \\
\langle E \rangle [\text{MeV}] & 8.32 \pm 0.40 \\
\text{FWHM} [\text{MeV}] & 7.93 \pm 0.58 \\
\hline
\end{array} \]

Table 5.10: Overview of the results of the ternary triton measurement of \(^{243}\)Cm with telescope 4.

distribution are corrected, using \(\langle E \rangle_t = 8.4 \pm 0.2 \text{ MeV}\) and \(\text{FWHM}_t = 7.3 \pm 0.3 \text{ MeV}\) for \(^{239}\)Pu [2]. All the corrected values can be found in tables 5.9 and 5.10. To obtain a general result for the characteristics of the energy distribution and the relative emission probability \(t/LRA\), a weighted average is made of both results from tables 5.9 and 5.10. Now results for \(LRA/B\) and \(t/LRA\) are available, so a value for the absolute emission probability \(t/B\) can be determined. These results are shown in table 5.7.

\(^{6}\text{He particles}\)

The \(^{6}\text{He particles}\) were measured with telescope 1 and 3, which were also used to detect the \(LRA\) particles. The first measurement was performed with telescope 1. The 2-dimensional \(E_{\text{tot}} - T/a\) spectrum and corresponding \(T/a\) spectrum are shown in figure 5.6 (upper part). A selection of the \(^{6}\text{He particles}\) is done in the 2-dimensional spectrum, based on the \(T/a\) identification relation. A comparison between the theoretical and experimental values for \(T/a\) can be made:

\[
T/a(^{6}\text{He})_{\text{theo}} = 32.15 \\
T/a(^{6}\text{He})_{\text{exp}} = 32.50
\]

which illustrates a good agreement between both values. The selection of the \(^{6}\text{He particles}\) is shown in figure 5.13.
5.1. $^{243}$Cm($N,F$)

Figure 5.13: 2-dimensional $E_{\text{tot}} - T/a$ spectrum and corresponding $T/a$ spectrum with a selection window on the $^6$He particles for the $^{243}$Cm neutron induced fission measurement with telescope 1.

For the measurements of $^6$He particles it is important to find the best compromise between two different requirements. First of all a sufficient separation is needed between the LRA and $^6$He particles, and on the other hand the detection limit of the $^6$He particles should not be too high in order to still permit a Gaussian fit through the data points, starting before the top. For this measurement the $\Delta E$ detector had a thickness of 29.8 $\mu$m, which resulted in a detection limit of about 10 MeV. In figure 5.14 (left) the total energy spectrum after energy correction is plotted.

Figure 5.14: Ternary $^6$He energy distributions for the $^{243}$Cm(n,f) measurement obtained with telescope 1 (left) and telescope 3 (right).

A Gaussian fit to all data points yields an average energy and a value for the FWHM, as well as the $^6$He counting rate. Combining this counting rate with the counting rate for $\alpha$-particles determined in table 5.5, a value for the relative emission probability $^6$He/LRA is obtained. All these results can be found in table 5.11.

Another measurement of the $^6$He particles could be performed with telescope 3. The $^6$He particles are again selected from the $E_{\text{tot}} - T/a$ spectrum, based on the $T/a$ relation. Due to the thickness of the $\Delta E$ detector, which is 31 $\mu$m, a higher cut off appears in the
Table 5.11: Overview of the results of the ternary $^6$He measurement of $^{243}$Cm with telescope 1.

The total energy spectrum. Therefore the Gaussian fit was started at 10.5 MeV, as can be seen in figure 5.14 (right). Again values for $\langle E \rangle$, FWHM and the $^6$He counting rate are deduced (table 5.12).

Table 5.12: Overview of the results of the ternary $^6$He measurement of $^{243}$Cm with telescope 3.

Again all the results have to be corrected for the ingrow of $^{239}$Pu. The correction on the relative emission probability $^6$He/LRA can be done in a similar way as explained for LRA/B. A value $^6$He/LRA = (1.8 ± 0.1)% has been adopted for $^{239}$Pu [2]. The correction on $^6$He/LRA is done for both measurements with telescope 1 and 3, taking into account the corresponding amount of $^{239}$Pu present in the sample. Also the parameters of the energy distribution are corrected, using $\langle E \rangle_{^6He} = 11.3 ± 0.3$ MeV and FWHM$_{^6He} = (10.7 ± 0.4)$ MeV for $^{239}$Pu [2]. All the corrected values can be found in tables 5.11 and 5.12.

To obtain general results for the $^6$He particles, a weighted average of the results of both campaigns is made. These values are written in table 5.7. Now results for LRA/B and $^6$He/LRA are available, so a value for the absolute emission probability $^6$He/B can be determined as well, which is also given in table 5.7.
5.2 $^{244}$Cm(SF)

5.2.1 Characteristics of the sample

Like the $^{243}$Cm sample, also the $^{244}$Cm sample was prepared at the Russian Federal Nuclear Center in Arzamas, using the technique of electrodeposition. A spot of curium oxide with a diameter of 15 mm is deposited on an aluminium backing foil with a thickness of 30 µm. The $^{244}$Cm sample has a mass of 18.5 µg and at the moment of preparation its activity was 53.7 MBq. Details on the isotopic composition of the sample can be found in table 5.13.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Isotopic analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{244}$Cm</td>
<td>99.48</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>6.5 E-2</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>4.38 E-1</td>
</tr>
<tr>
<td>$^{247}$Cm</td>
<td>6 E-3</td>
</tr>
<tr>
<td>$^{248}$Cm</td>
<td>1.1 E-2</td>
</tr>
</tbody>
</table>

Table 5.13: Isotopic composition (in atomic percent) of the $^{244}$Cm sample.

$^{244}$Cm undergoes radioactive $\alpha$-decay, emitting $\alpha$-particles with a most probable energy of 5.805 MeV. Because of this $\alpha$-decay, $^{240}$Pu is growing in the sample material. However, this will not cause a problem, since $T_{1/2}^{(SF)} = 1.32 \times 10^7$ a.

Some characteristics of the isotope $^{244}$Cm are described in table 5.1. As can be seen in this table, the ratio between the half lives for spontaneous fission and radioactive $\alpha$-decay is unfavourable to measure spontaneous fission. Therefore a lot of time was needed to accumulate a statistically significant amount of data. Moreover, the activity of the source is quite high, which complicated the measurements.

5.2.2 Overview of the experiments

Four measurements with different telescope detectors were performed in order to examine the spontaneous fission of $^{244}$Cm:

1. The first telescope consisted of an ionisation chamber as $\Delta E$ detector, coupled to a surface barrier detector as E detector, as explained in section 4.2.2. In this way the ternary particles and the binary fission fragments can be measured consecutively in the same detection geometry.

The surface barrier detector has an active area of 2000 mm², a depletion depth of 1500 µm and an energy resolution of 21 keV. With this telescope a detection geometry of about 10% was realised.

2. In a second experiment, telescopes 1 and 2 (see table 5.14), each consisting of two surface barrier detectors optimised for ternary $\alpha$ detection, were mounted in a vacuum.
chamber, at a distance of about 15 mm of the $^{244}$Cm sample.

3. The third telescope also consisted of two surface barrier detectors (table 5.14), which were optimised for the detection of tritons.

4. The fourth telescope consisted of two surface barrier detectors with a smaller active area of 150 mm$^2$, optimised to detect $^6$He particles by limiting the influence of the high activity of the source.

With the smaller telescopes (1, 3) used in the vacuum chamber the detection geometry was only 3%.

<table>
<thead>
<tr>
<th>$^{244}$Cm</th>
<th>$\Delta E$</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td>area</td>
<td>thickness</td>
<td>resolution</td>
</tr>
<tr>
<td>[mm$^2$]</td>
<td>[µm]</td>
<td>[keV]</td>
</tr>
<tr>
<td>telescope 1</td>
<td>300</td>
<td>31.7</td>
</tr>
<tr>
<td>telescope 2</td>
<td>150</td>
<td>28.9</td>
</tr>
<tr>
<td>telescope 3</td>
<td>300</td>
<td>49.8</td>
</tr>
<tr>
<td>telescope 4</td>
<td>150</td>
<td>32.8</td>
</tr>
</tbody>
</table>

Table 5.14: Characteristics of the surface barrier telescope detectors used for the $^{244}$Cm spontaneous fission measurements. The indicated resolution is for 5.5 MeV $\alpha$-particles.

5.2.3 Binary fission

The set-up described in section 5.2.2(1) was used to determine the $^{244}$Cm(SF) absolute ternary $\alpha$ emission probability LRA/B. To measure the binary fission fragments, the $\Delta E$ detector is removed. This is realised by pumping the methane gas out of the chamber, which then becomes a vacuum chamber. The E detector was used to detect the fragments.

The determination of the binary fission counting rate was hampered by the high activity of the sample, which created a too strong $\alpha$ pile-up and dead time in the data acquisition system. This problem was solved by reducing the counting rate by putting a collimator with a diameter of 7 mm in front of the detector, without changing the internal distances. However, due to radiation damage of the detector, the pulse height spectrum was gradually shifting to the left, as is illustrated in figure 5.15. In total the measuring time was 130 hours. In the figure three different runs, each with a measuring time of 10 hours, are plotted together. From the picture, it is clear that every run had to be analysed separately.

Moreover, again a peak due to the radioactive $\alpha$-decay pile-up is visible in the beginning of the spectra. This should be removed and then a clean binary fission spectrum for every run is obtained like shown in figure 5.16.

The results of the binary fission measurement can be found in table 5.15. Since the measurement of the ternary particles is performed without the collimator, the scale factor for a measurement with and without the collimator has to be determined. Therefore the $^{244}$Cm sample was replaced by a $^{252}$Cf sample with the same dimensions.
### 5.2. $^{244}$CM(SF)

![Graph](image)

**Figure 5.15:** Partial measured binary fission spectra for $^{244}$Cm(SF), illustrating the shift of the spectrum due to radiation damage of the detector (black: first run, green: last run).

![Graph](image)

**Figure 5.16:** Partial binary fission spectrum for $^{244}$Cm(SF) after removing the $\alpha$ pile-up.

and the scale factor was obtained by counting the $^{252}$Cf radioactive decay $\alpha$’s with the E detector in both configurations, once with and once without collimator.

### 5.2.4 Ternary fission

**Alpha particles:** yield

The ternary fission measurement with the ionisation chamber was used to determine the yield of the ternary $\alpha$-particles in order to obtain a value for the absolute emission proba-
Table 5.15: Overview of the results of the binary fission measurement of $^{244}$Cm, as well as the determination of LRA/B.

Figure 5.17 shows the complete $E_{tot} - T/a$ spectrum, together with the projected $T/a$ spectrum obtained with the ionisation chamber. Again the $T/a$ identification relation was used to select the ternary $\alpha$ particles ($T/a \approx 18.5$). This spectrum shows also that the background in the region of the triton peak ($T/a \approx 3.7$) is too high to permit a reliable selection.

The ternary alpha counting rate is obtained from a Gaussian fit to the total energy spectrum, starting at 12.5 MeV (figure 5.18). The result is listed in table 5.15, together with the obtained value for the absolute emission probability LRA/B.

![Graph](image)

Figure 5.17: 2-dimensional $E_{tot} - T/a$ and corresponding $T/a$ spectrum for the $^{244}$Cm spontaneous fission measurement with the ionisation chamber.

The parameters of the energy distribution will be determined with the vacuum chamber. Indeed, the accuracy of the values obtained with the ionisation chamber can not be guaranteed, since a precise calibration of the $\Delta E$ detector was difficult to perform. Most of the particles used for the calibration had an energy too high to be stopped in the $\Delta E$ detector. Therefore only one calibration point ($^{147}$Sm with $E_\alpha = 2.235$ MeV, section 4.5) was available.
Figure 5.18: Ternary $\alpha$ energy distribution and corresponding Gaussian fit for $^{244}$Cm(SF) obtained with the ionisation chamber.

**Alpha particles: Energy distribution**

The set-up described in section 5.2.2(2) was used to measure the ternary alpha particles. Initially there was an attempt to measure also tritons, but due to the high activity of the sample also here the selection of the tritons was not satisfactory. Figure 5.19 shows the ternary $\alpha$ distributions obtained with both telescopes 1 and 2 after correction for the energy loss in the aluminium. As can be seen in table 5.16, the results of the fit to both spectra are in agreement and a Gaussian fit to the data points above 12.5 MeV of the sum spectrum yields the average energy and FWHM.

Figure 5.19: Ternary $\alpha$ energy distribution and corresponding Gaussian fit for $^{244}$Cm obtained with telescope 1 (right) and telescope 2 (left).
<table>
<thead>
<tr>
<th>$\Delta T$ [s]</th>
<th>Telescope 1</th>
<th>Telescope 2</th>
<th>Sum spectrum</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>91.05269</td>
<td>5710.436</td>
<td>15.99 ± 0.10</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>15.96 ± 0.13</td>
<td>16.04 ± 0.15</td>
<td>9.99 ± 0.29</td>
</tr>
</tbody>
</table>

Table 5.16: Overview of the results of the ternary $\alpha$ measurement of $^{244}$Cm with telescope 1 and 2, as well as the results obtained with the sum spectrum.

**Tritons**

In section 5.2.2(3), a set-up is described where a thicker $\Delta E$ detector is used, resulting in an enhanced energy loss of the ternary particles, thus a better separation of the particles in the $E_{\text{tot}} - T/\alpha$ spectrum and its projection (figure 5.20). A selection of the $\alpha$-particles and tritons is made in the 2-dimensional spectrum, based on the $T/\alpha$ identification relation ($T/\alpha(\alpha) \approx 41.53$, $T/\alpha(t) \approx 8.44$).

![2-dimensional $E_{\text{tot}} - T/\alpha$ and corresponding $T/\alpha$ spectrum for the spontaneous fission measurement of $^{244}$Cm with telescope 3.](image)

In this way, and after correction for the energy loss, the ternary $\alpha$ and triton total energy distributions could be obtained and they are shown in figure 5.21. A Gaussian fit to all the triton data points yields the average energy and FWHM given in table 5.17, as well as a counting rate for the tritons. Due to the selection of the tritons in the 2-dimensional spectrum, no background correction is needed.

In combination with the yield of the ternary $\alpha$-particles, this measurement gives a value for the relative triton emission probability $t/LRA$. Since a value for $LRA/B$ is determined with the ionisation chamber, $t/B$ can also be deduced (table 5.17).

**$^6$He particles**

As mentioned already before, $^{244}$Cm has a very unfavourable ratio between the half lives for spontaneous fission and radioactive $\alpha$-decay, which makes it really difficult to measure the more rare ternary particles like $^6$He particles. Therefore these measurements were
Figure 5.21: Ternary α (left) and triton (right) energy distributions for the $^{244}$Cm spontaneous fission measurement with telescope 3.

Table 5.17: Overview of the results of the ternary α and triton measurement of $^{244}$Cm with telescope 3, as well as triton emission probabilities.

<table>
<thead>
<tr>
<th></th>
<th>α-particles</th>
<th>tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>13533781</td>
<td>13533781</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>16.21 ± 0.15</td>
<td>8.05 ± 0.29</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.15 ± 0.35</td>
<td>7.89 ± 0.48</td>
</tr>
<tr>
<td>total number</td>
<td>84943 ± 1257</td>
<td>5250 ± 616</td>
</tr>
<tr>
<td>yield [h$^{-1}$]</td>
<td>22.59 ± 0.33</td>
<td>1.40 ± 0.16</td>
</tr>
<tr>
<td>t/LRA [%]</td>
<td>6.18 ± 0.73</td>
<td></td>
</tr>
<tr>
<td>t/B [10$^{-4}$]</td>
<td>1.89 ± 0.24</td>
<td></td>
</tr>
</tbody>
</table>

very time consuming and still statistics are not very satisfactory. In addition, due to the high activity of the source, it was complicated to eliminate the background. Two different measurements were performed.

First telescope 1, used for the detection of the LRA particles, was used. Making a selection of the $^6$He particles in the 2-dimensional $E_{tot} - T/a$ spectrum, the total energy spectrum is obtained after energy correction. This spectrum is shown in figure 5.22. A Gaussian fit starting at 9.5 MeV yields the parameters of the energy distribution as well as the total number of $^6$He particles. The results of this first measurement can be found in table 5.18.

A second measurement is performed with telescope 4, which consists of both a ΔE and E detector with an active area of 150 mm$^2$, in order to limit the hindrance due to the high activity of the source. However, due to the smaller active area of the detectors, the counting rate is even lower. Again a selection of the $^6$He particles is made in the 2-dimensional $E_{tot} - T/a$ spectrum. The total energy spectrum is shown in figure 5.23 and the results of the Gaussian fit, starting at 10 MeV, can be found in table 5.18.

In order to obtain a general result for the $^6$He particles, a weighted average of the results for the average energy, FWHM and $^6$He/LRA is calculated. These values are given in table 5.18.
CHAPTER 5. CM ISOTOPES

Figure 5.22: Ternary $^6$He energy distribution for the $^{244}$Cm spontaneous fission measurement with telescope 1.

<table>
<thead>
<tr>
<th></th>
<th>Telescope 1</th>
<th>Telescope 4</th>
<th>weighted average</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>9105269</td>
<td>21479862</td>
<td></td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>10.00 ± 0.69</td>
<td>10.57 ± 0.77</td>
<td>10.25 ± 0.51</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>9.64 ± 1.01</td>
<td>10.24 ± 1.21</td>
<td>9.89 ± 0.78</td>
</tr>
<tr>
<td>number of $^6$He particles</td>
<td>1206 ± 148</td>
<td>741 ± 113</td>
<td></td>
</tr>
<tr>
<td>number of $\alpha$-particles</td>
<td>59680 ± 657</td>
<td>46128 ± 572</td>
<td></td>
</tr>
<tr>
<td>$^6$He/LRA [%]</td>
<td>2.02 ± 0.25</td>
<td>1.61 ± 0.25</td>
<td>1.82 ± 0.18</td>
</tr>
<tr>
<td>$^6$He/B [$10^{-5}$]</td>
<td></td>
<td></td>
<td>5.75 ± 0.59</td>
</tr>
</tbody>
</table>

Table 5.18: Overview of the results of the ternary $^6$He measurements of $^{244}$Cm.

5.3 Literature survey and comments

As shown in table 5.1, six curium isotopes have a sufficiently long total half-life ($T_{1/2}$) to permit the preparation of samples. Three of these isotopes ($^{244,246,248}$Cm) have a sufficiently small half-life for spontaneous fission to permit ternary fission measurements, three others ($^{243,245,247}$Cm) have a large fission cross section with thermal neutrons and a negligible spontaneous fission decay.

To permit a coherent interpretation, the present results for the fissioning system $^{244}$Cm will be combined with similar data previously obtained by our research group for $^{246}$Cm [50, 51] and $^{248}$Cm [52, 53].

An overview of the characteristics of the measured energy distributions for the ternary $\alpha$’s and tritons for the different curium isotopes is given in table 5.19, while an overview of the measured emission probabilities (the uncertainties and the background correction for the tritons were in some cases slightly revised) for the ternary $\alpha$’s and tritons for the Cm isotopes can be found in table 5.20.
5.3. LITERATURE SURVEY AND COMMENTS

![Energy Distribution Graph](image)

Figure 5.23: Ternary $^6$He energy distribution for the $^{241}$Cm spontaneous fission measurement with telescope 4.

<table>
<thead>
<tr>
<th>Ternary alpha particles</th>
<th>Ternary tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>FWHM [MeV]</td>
</tr>
<tr>
<td>$^{243}$Cm(n,f)</td>
<td>16.14 ± 0.10</td>
</tr>
<tr>
<td>$^{245}$Cm(n,f)</td>
<td>16.35 ± 0.15</td>
</tr>
<tr>
<td>$^{247}$Cm(n,f)</td>
<td>16.01 ± 0.13</td>
</tr>
<tr>
<td>$^{241}$Cm(SF)</td>
<td>15.99 ± 0.10</td>
</tr>
<tr>
<td>$^{246}$Cm(SF)</td>
<td>16.41 ± 0.20</td>
</tr>
<tr>
<td>$^{248}$Cm(SF)</td>
<td>15.97 ± 0.12</td>
</tr>
</tbody>
</table>

Table 5.19: Overview of the characteristics of the measured energy distributions for the ternary $\alpha$’s and tritons for the different curium isotopes.

The $^6$He particles have not been measured for all six curium isotopes. In this work results were obtained for $^{243}$Cm and $^{244}$Cm, while before only in the case of $^{245}$Cm(n,f) $^6$He particles have been measured. An overview of the characteristics of the measured energy distributions and the emission probabilities is given in table 5.21.

These are almost the only data reported on curium isotopes in the literature. In 1962 Nobles [54] investigated the ternary particle emission probability in $^{244}$Cm(SF). No particle identification was done in his pioneering experiment, so the reported value of $(3.18 \pm 0.20) \times 10^{-3}$, although dominated by ternary $\alpha$’s, also includes a contribution of other ternary particles, which makes it difficult to compare with our value for the LRA emission probability. Anyhow, there does not seem to be a conflict.

For $^{245}$Cm(n$_{th}$,f) the ternary $\alpha$ energy distribution was measured by Koczon et al. [55] in 1986, while Köster [56] (2000) measured a fraction of the low-energy side of the ternary $\alpha$ and triton energy distribution, as well as the $^6$He energy distribution. The measurement of
Table 5.20: Overview of the emission probabilities for the ternary α’s and tritons for the different curium isotopes.

<table>
<thead>
<tr>
<th></th>
<th>LRA/B $[10^{-3}]$</th>
<th>t/B $[10^{-5}]$</th>
<th>t/LRA [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{243}$Cm(n,f)</td>
<td>2.43 ± 0.08</td>
<td>1.89 ± 0.15</td>
<td>7.78 ± 0.54</td>
</tr>
<tr>
<td>$^{245}$Cm(n,f)</td>
<td>2.15 ± 0.12</td>
<td>1.85 ± 0.21</td>
<td>8.60 ± 1.09</td>
</tr>
<tr>
<td>$^{247}$Cm(n,f)</td>
<td>1.85 ± 0.10</td>
<td>1.84 ± 0.20</td>
<td>9.94 ± 1.21</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>$^{243}$Cm(SF)</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3.16 ± 0.09</td>
<td>1.89 ± 0.24</td>
<td>6.18 ± 0.73</td>
</tr>
<tr>
<td></td>
<td>2.49 ± 0.12</td>
<td>1.72 ± 0.24</td>
<td>6.91 ± 1.02</td>
</tr>
<tr>
<td></td>
<td>2.30 ± 0.10</td>
<td>1.79 ± 0.21</td>
<td>7.78 ± 0.97</td>
</tr>
</tbody>
</table>

Table 5.21: Overview of the characteristics of the energy distributions and the emission probabilities for the ternary $^6$He particles for three curium isotopes.

Koczen et al. yields an average energy of 16.6 MeV, while the FWHM of a Gaussian fit is equal to 9.2 MeV. No uncertainties are mentioned on these values, therefore a comparison with our data is not easy to perform. In figure 5.24 the LRA spectrum is plotted.

The results obtained by Köster are given in table 5.22, and the corresponding spectra are plotted in figure 5.25. For the ternary α and triton spectrum, only the low-energy tail of the spectrum was measured, therefore a fixed average energy had to be imposed to perform the Gaussian fit. In his measurement relative emission probabilities for tritons and $^6$He particles could be obtained. His results are in fair agreement with the ones reported by Serot et al. [50] and cited in tables 5.19, 5.20 and 5.21.

<table>
<thead>
<tr>
<th></th>
<th>$^6$He/B $[10^{-5}]$</th>
<th>$^6$He/LRA [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{243}$Cm(n,f)</td>
<td>4.13 ± 0.50</td>
<td>1.70 ± 0.20</td>
</tr>
<tr>
<td>$^{245}$Cm(n,f)</td>
<td>4.95 ± 1.25</td>
<td>2.30 ± 0.58</td>
</tr>
<tr>
<td>$^{247}$Cm(SF)</td>
<td>5.75 ± 0.59</td>
<td>1.82 ± 0.18</td>
</tr>
</tbody>
</table>

Table 5.22: Overview of the results of the ternary fission measurement of $^{245}$Cm by Köster [56].

Finally, the emission of H and He particles in $^{248}$Cm(SF) was studied by Ivanov et al. [57] in 1996. From these measurements the characteristics of the energy distribution for tritons, α and $^6$He particles are available, as well as the emission probabilities for these ternary particles. All these values can be found in table 5.23, together with the spectra of
the energy distributions in figure 5.26. Within the experimental uncertainties, their results agree with those reported by Serot and Wagemans [52], given in tables 5.19 and 5.20.

<table>
<thead>
<tr>
<th></th>
<th>( \langle E \rangle ) [MeV]</th>
<th>FWHM [MeV]</th>
<th>abs. em. prob.</th>
<th>relative em. prob. [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>tritons</td>
<td>8.7 ± 0.3</td>
<td>8.0 ± 0.2</td>
<td>(2.03 ± 0.28) ( \times 10^{-4} )</td>
<td>9.22 ± 0.18</td>
</tr>
<tr>
<td>LRA</td>
<td>15.8 ± 0.3</td>
<td>10.1 ± 0.3</td>
<td>(2.2 ± 0.3) ( \times 10^{-3} )</td>
<td>100</td>
</tr>
<tr>
<td>(^6\text{He})</td>
<td>11.5 ± 0.6</td>
<td>10.4 ± 0.7</td>
<td>(7.79 ± 1.26) ( \times 10^{-5} )</td>
<td>3.54 ± 0.31</td>
</tr>
</tbody>
</table>

Table 5.23: Overview of the results of the spontaneous fission measurement of \(^{248}\text{Cm}\) by Ivanov et al. [57].
Figure 5.25: Ternary energy distributions of tritons, LRA and \(^6\)He particles for \(^{245}\)Cm(n_{\text{th}},f) measured by Köster [56].
Figure 5.26: Ternary energy distributions of tritons, LRA and $^6$He particles for $^{248}$Cm(SF) measured by Ivanov et al. [57].
Chapter 6

Cf isotopes

This chapter presents in detail the neutron induced ternary fission measurements of $^{249,251}$Cf and the spontaneous ternary fission measurements of $^{250,252}$Cf, as well as the results of these measurements. The chapter ends with a literature survey for the fissioning systems $^{250,252}$Cf.

6.1 $^{249}$Cf(n,f)

6.1.1 Characteristics of the sample

The $^{249}$Cf sample was prepared at the Lawrence Berkeley National Laboratory (LBNL) in the USA. A spot of californium oxide with a diameter of 6 mm was deposited on a Ti-foil with a diameter of 14 mm and a thickness of 40 μm. The isotopic analysis was performed on 20/02/2006. The sample has a mass of 5.84 μg and an enrichment of 100%, at the moment of preparation its activity was 0.88 MBq. Some nuclear characteristics of the different Cf isotopes can be found in table 6.1.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}(\alpha)$</th>
<th>$T_{1/2}(SF)$</th>
<th>$\sigma(n_{th}, f)$</th>
<th>$S_n$</th>
<th>$E_{\text{MeV}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{249}$Cf</td>
<td>351</td>
<td>7.02 E+8</td>
<td>1.63 E+3</td>
<td>-</td>
<td>6.625</td>
</tr>
<tr>
<td>$^{250}$Cf</td>
<td>13.09</td>
<td>1.70 E+4</td>
<td>4.09</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$^{251}$Cf</td>
<td>898</td>
<td>9 E+12</td>
<td>5.32 E+3</td>
<td>-</td>
<td>6.172</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td>2.73</td>
<td>85.54</td>
<td>32.18</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.1: Nuclear characteristics of the different californium isotopes.

6.1.2 Overview of the experiments

A first measurement campaign in order to examine the neutron induced ternary fission of $^{249}$Cf started on 25/07/06. The thermal equivalent neutron flux at the sample position was about $2 \times 10^9$ neutrons/cm$^2$.s.
Two $\Delta E$-$E$ surface barrier telescope detectors (telescope 1 and 2) were placed on both sides of the sample, perpendicular to the incoming neutron beam. An overview of their characteristics can be found in table 6.2.

<table>
<thead>
<tr>
<th></th>
<th>$^{249}$Cf</th>
<th>$\Delta E$</th>
<th>$E$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>area [mm$^2$]</td>
<td>thickness [\mu m]</td>
<td>resolution [keV]</td>
</tr>
<tr>
<td>telescope 1</td>
<td>300</td>
<td>29.8</td>
<td>41</td>
</tr>
<tr>
<td>telescope 2</td>
<td>300</td>
<td>49.8</td>
<td>30</td>
</tr>
<tr>
<td>telescope 3</td>
<td>300</td>
<td>41</td>
<td>31</td>
</tr>
</tbody>
</table>

Table 6.2: Characteristics of the surface barrier telescope detectors used for the $^{249}$Cf neutron induced fission measurements. The indicated resolution is for 5.5 MeV $\alpha$-particles.

Although both telescopes were installed together in the chamber, a simultaneous measurement (like for the curium isotopes, see chapter 5) was impossible due to the thickness of the Ti backing foil. Indeed, this foil stops the ternary particles, hence the sample had to be turned over 180° to detect the particles with the other telescope. Therefore both $\Delta E$ detectors were covered with a thin aluminium foil of 30 $\mu$m. Telescope 1 was used to detect the LRA particles and the $^6$He particles, while telescope 2 was chosen in order to have an optimal setup for detecting tritons.

Two years later, starting on 07/10/08, a second measurement to investigate the neutron induced ternary fission of $^{249}$Cf was performed with a thermal equivalent neutron flux of about $4 \times 10^3$ neutrons/cm$^2$.s. Again telescope 1 was used to detect the LRA and $^6$He particles, however, with much better statistics than in the previous experiment. To detect the tritons in optimal conditions, telescope 3 was selected, with a thinner $\Delta E$ detector of 41 $\mu$m. In addition, both $\Delta E$ detectors were covered with a thinner aluminium foil of only 25 $\mu$m, which results in a lower detection threshold.

### 6.1.3 Binary fission

During each measurement campaign a binary fission measurement was performed with detector E from telescope 1, while the $\Delta E$ detector was replaced by a dummy, maintaining exactly the same geometry. For the first experiment, the measured spectrum with open neutron beam is plotted in figure 6.1 (left). The small $\alpha$ pile-up peak due to the radioactive decay of $^{249}$Cf in the beginning of the spectrum has to be separated from the two bumps of the fission fragments. Here the cut off was made at channel 529. Then the remaining spectrum is extrapolated and the corresponding number of binary fission fragments was deduced after integration of the extrapolated spectrum (figure 6.1, right). Here a correction due to the dead time of the measuring system had to be included. This was determined in a parallel run with a counter, yielding a dead time of about 0.5%. In addition a measurement with closed neutron beam was done. In this run no spontaneous fission events were detected.

The results of the binary fission measurement are listed in table 6.3.
6.1. $^{249}$CF(N,F)

Figure 6.1: Measured (left) and final extrapolated (right) binary fission spectrum for $^{249}$Cf(n,f) with open neutron beam during the first measurement campaign.

<table>
<thead>
<tr>
<th></th>
<th>1st experiment</th>
<th>2nd experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ beam off [s]</td>
<td>600</td>
<td>2968</td>
</tr>
<tr>
<td>$\Delta T$ beam on [s]</td>
<td>4751</td>
<td>509</td>
</tr>
<tr>
<td>number of fission fragments after extrapolation and dead time correction</td>
<td>3086260 ± 1757</td>
<td>1048350 ± 1024</td>
</tr>
<tr>
<td>number of fission events</td>
<td>3104118 ± 17009</td>
<td>1048350 ± 3121</td>
</tr>
<tr>
<td>binary fission yield [s$^{-1}$]</td>
<td>1552059 ± 8504</td>
<td>524175 ± 1561</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>326.68 ± 1.79</td>
<td>1029.81 ± 3.07</td>
</tr>
</tbody>
</table>

Table 6.3: Results of the binary fission measurements of $^{249}$Cf.

The measured binary fission spectrum obtained during the second campaign is shown in figure 6.2 (left).

Figure 6.2: Measured (left) and final extrapolated (right) binary fission spectrum for $^{249}$Cf(n,f) with open neutron beam during the second measurement campaign.

Again a small peak due to the radioactive alpha decay is visible at the left side of the
spectrum. The cut off is made at channel 776 and the final extrapolated spectrum can be seen in figure 6.2 (right). In this case no dead time correction was needed, since a linear gate module was used during the measurement, in order to avoid dead time (see chapter 4 for details). The results of this measurement can be found in table 6.3.

6.1.4 Ternary fission

Alpha particles: first measurement campaign

Telescope 1 was used to measure the energy distribution and to determine the absolute emission probability of the ternary α-particles. In figure 6.3 the 2-dimensional $E_{\text{tot}} - T/a$ and $E-\Delta E$ spectra are plotted. In the $E_{\text{tot}} - T/a$ spectrum a selection of the LRA particles can be made based on the $T/a$ identification relation ($T/a \approx 24.85$). The result of this selection is illustrated in figure 6.4. A very good agreement exists between the experimentally determined value of $T/a$ and the theoretical one.

![Figure 6.3: 2-dimensional $E_{\text{tot}} - T/a$ and $E-\Delta E$ spectra for the first $^{249}$Cf neutron induced fission measurement with telescope 1.](image)

![Figure 6.4: Selection of the α-particles in the 2-dimensional $E_{\text{tot}} - T/a$ and $E-\Delta E$ spectra for the first $^{249}$Cf neutron induced fission measurement with telescope 1.](image)

After correction for the energy loss in the 30 μm thick aluminium foil, the total energy distribution for the alpha particles is obtained. The ternary α counting rate, as well as
the parameters of the energy distribution, are obtained from a Gaussian fit to the data, starting at 12.5 MeV, shown in figure 6.5. The results of the fit are given in table 6.4, together with the value obtained for LRA/B.

![Figure 6.5: Ternary α energy distribution for the $^{249}$Cf(n,f) measurement obtained with telescope 1 during the first measurement campaign.](image)

<table>
<thead>
<tr>
<th></th>
<th>1$^{st}$ experiment</th>
<th>2$^{nd}$ exp., 1$^{st}$ run</th>
<th>2$^{nd}$ exp., 2$^{nd}$ run</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>55216</td>
<td>57648</td>
<td>330861</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>$16.09 \pm 0.18$</td>
<td>$16.09 \pm 0.13$</td>
<td>$16.07 \pm 0.11$</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.64 $\pm$ 0.27</td>
<td>10.84 $\pm$ 0.16</td>
<td>10.84 $\pm$ 0.14</td>
</tr>
<tr>
<td>number of α-particles</td>
<td>50011 $\pm$ 1884</td>
<td>162550 $\pm$ 4612</td>
<td>890515 $\pm$ 8556</td>
</tr>
<tr>
<td>counting rate [s$^{-1}$]</td>
<td>0.906 $\pm$ 0.034</td>
<td>2.82 $\pm$ 0.08</td>
<td>2.69 $\pm$ 0.03</td>
</tr>
<tr>
<td>LRA/B [10$^{-3}$]</td>
<td>2.77 $\pm$ 0.11</td>
<td>2.74 $\pm$ 0.08</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.4: Overview of the results of the ternary α measurement of $^{249}$Cf with telescope 1.

**Alpha particles: second measurement campaign**

The same telescope, telescope 1, was used to determine a value for the absolute emission probability and to measure the energy distribution of the LRA particles. Two different runs were performed: a first one to determine the absolute LRA emission probability, a second one to determine the relative emission probability $^6$He/LRA. In the first run, the lower level of the TSCA was chosen in order to have a counting rate which caused no dead
time. The LRA particles were selected in the 2-dimensional $E_{\text{tot}} - T/a$ spectrum, based on their $T/a$ value. The total energy distribution is obtained after correction for the energy loss in the 25 $\mu$m thick aluminium foil. The ternary $\alpha$ counting rate is deduced from a Gaussian fit to the data, starting at 12.5 MeV, shown in figure 6.6. The results are given in table 6.4, together with the value obtained for LRA/B. Also values for the average energy and FWHM are written in the table.

![Figure 6.6](image)

Figure 6.6: Ternary $\alpha$ energy distribution for the $^{249}$Cf(n,f) measurement obtained with telescope 1 during the first run of the second measurement campaign.

A second run is performed, with a decreased lower level of the TSCA, in order to have a lower detection threshold. In this way it will be possible to determine the relative emission probability $^6$He/LRA.

![Figure 6.7](image)

Figure 6.7: 2-dimensional $E_{\text{tot}} - T/a$ and $E-\Delta E$ spectra for the $^{249}$Cf neutron induced fission measurement with telescope 1, obtained during the second measurement campaign, second run.

The 2-dimensional $E_{\text{tot}} - T/a$ and $E-\Delta E$ spectra are plotted in figure 6.7. A selection
of the LRA particles in the $E_{\text{tot}} - T/\alpha$ can be made based on the $T/\alpha$ identification relation. This selection is illustrated in figure 6.8.

![Graph showing $E_{\text{tot}} - T/\alpha$ and $E - \Delta E$ spectra for the $^{249}$Cf neutron induced fission measurement with telescope 1, obtained during the second measurement campaign, second run.](image)

Figure 6.8: Selection of the $\alpha$-particles in the 2-dimensional $E_{\text{tot}} - T/\alpha$ and $E - \Delta E$ spectra for the $^{249}$Cf neutron induced fission measurement with telescope 1, obtained during the second measurement campaign, second run.

Taking into account this selection, a total energy distribution for the alpha particles is obtained after correction for the energy loss in the aluminium. The parameters of the energy distribution and the counting rate are obtained from a Gaussian fit to the data, starting at 12.5 MeV, shown in figure 6.9.

![Graph showing ternary $\alpha$ energy distribution for the $^{249}$Cf(n,f) measurement obtained with telescope 1 during the second measurement campaign, second run.](image)

Figure 6.9: Ternary $\alpha$ energy distribution for the $^{249}$Cf(n,f) measurement obtained with telescope 1 during the second measurement campaign, second run.

The results of the second run are written in table 6.4. Let us compare now the results obtained in the two measurement campaigns. The two values for LRA/B are in very good agreement with each other, as well as the values for $\langle E \rangle$ and FWHM. To determine the final value for the absolute emission probability, the
weighted average is calculated. The parameters of the energy distribution are calculated as a weighted average of the three available values. These final values can be found in table 6.4.

$^6$He particles

Due to very low statistics and a detection limit which was too high, $^6$He particles could not be measured in an appropriate way during the first measurement campaign. As mentioned already before, during the second measurement campaign an aluminium foil with a thickness of only 25 $\mu$m was used. Consequently a lower detection threshold could be achieved. Due to a higher neutron flux and more beam time available, also statistics could be improved drastically.

In the $E_{\text{tot}} - T/a$ spectrum (figure 6.7, left) a selection of the $^6$He particles can be made based on the $T/a$ identification relation ($T/a \approx 32.2$). The result of this selection is shown in figure 6.10.

![Figure 6.10: Selection of the $^6$He particles in the 2-dimensional $E_{\text{tot}} - T/a$ and $E-\Delta E$ spectra for the $^{249}$Cf neutron induced fission measurement with telescope 1, obtained during the second measurement campaign, second run.]

After correction for the energy loss in the aluminium, the total energy distribution for the $^6$He particles is obtained. The $^6$He counting rate, as well as the parameters of the energy distribution, are obtained from a Gaussian fit to the data, starting at 9.9 MeV, plotted in figure 6.11.

The results are given in table 6.5, together with the value obtained for $^6$He/LRA. Since LRA/B is already determined, a value for $^6$He/B can be calculated too.

Tritons

During the first measurement campaign, telescope 2 was used for the detection of the tritons. In figure 6.12 the 2-dimensional spectrum $E_{\text{tot}} - T/a$ is plotted.

A selection of the tritons was made in the $T/a$ spectrum ($T/a \approx 8.5$, figure 6.13). The total energy spectrum obtained after energy correction is illustrated in figure 6.14 (right). A Gaussian fit to all data points (starting at 6.4 MeV) yields the average triton energy and
6.1. $^{249}\text{CF}(N,F)$

Figure 6.11: Ternary $^6\text{He}$ energy distribution for the $^{249}\text{Cf}(n,f)$ measurement obtained with telescope 1 during the second measurement campaign, in order to determine the parameters of the energy distribution and the relative emission probability $^6\text{He}/\text{LRA}$.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>330861</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>10.99 ± 0.32</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.35 ± 0.60</td>
</tr>
<tr>
<td>number of $^6\text{He}$ particles</td>
<td>22588 ± 2017</td>
</tr>
<tr>
<td>counting rate [$\text{s}^{-1}$]</td>
<td>0.068 ± 0.006</td>
</tr>
<tr>
<td>$^6\text{He}/\text{LRA}$ [%]</td>
<td>2.54 ± 0.23</td>
</tr>
<tr>
<td>$^6\text{He}/\text{B}$ [10$^{-5}$]</td>
<td>6.99 ± 0.66</td>
</tr>
</tbody>
</table>

Table 6.5: Overview of the results of the ternary $^6\text{He}$ measurement of $^{249}\text{Cf}$ with telescope 1.

FWHM as well as the triton counting rate, given in table 6.6. As can be seen in figure 6.13, the yield for tritons can only be obtained after subtracting the background. Therefore a fit is made to the different contributions of the background, like e.g. the deuterons and protons. Taking this into account, a background of about 9.75% is obtained. In table 6.6 the adopted value is given.

To obtain the relative emission probability for tritons, also the ternary alpha particle counting rate measured with the same telescope has to be determined. In figure 6.14 (left) the total energy distribution for the ternary $\alpha$’s is plotted. Here the cut off is higher than in figure 6.9 due to the use of a thicker $\Delta E$ detector. A Gaussian fit to all data points is made and the results are shown in table 6.6.

Combining both the yield for tritons and LRA particles, a value for $t/\text{LRA}$ is obtained.

During the second measurement campaign tritons were measured with telescope 3.
Figure 6.12: 2-dimensional $E_{\text{tot}} - T/a$ spectrum for the $^{249}$Cf(n,f) measurement with telescope 2.

Figure 6.13: Illustrate of the determination of the triton background in the $T/a$ spectrum for the $^{249}$Cf neutron induced fission measurement with telescope 2.

With a thickness of only 41 $\mu$m for the $\Delta E$ detector and an aluminium foil of 25 $\mu$m, a lower detection limit could be obtained in comparison with the first measurement campaign. The 2-dimensional spectrum $E_{\text{tot}} - T/a$ is plotted in figure 6.15 (left). A selection of the tritons was made again in the $T/a$ spectrum ($T/a \approx 7.0$) (figure 6.15 (right)). The total energy spectrum obtained after energy correction is shown in figure 6.16 (right). A Gaussian fit to all data points (starting at 6 MeV) yields the triton counting rate and the parameters of the energy distribution, given in table 6.7. Again a correction for the background is needed in order to obtain the correct triton yield. After subtracting the
6.1. $^{249}$CF(N,F)

Figure 6.14: Ternary $\alpha$ (left) and triton (right) energy distributions for the $^{249}$Cf neutron induced fission measurement with telescope 2.

<table>
<thead>
<tr>
<th></th>
<th>$\alpha$-particles</th>
<th>tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td></td>
<td>236289</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>16.17 ± 0.13</td>
<td>8.52 ± 0.26</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.93 ± 0.30</td>
<td>8.47 ± 0.56</td>
</tr>
<tr>
<td>total number</td>
<td>204262 ± 4962</td>
<td>17938 ± 1559</td>
</tr>
<tr>
<td>after subtracting background</td>
<td>16189 ± 1723</td>
<td></td>
</tr>
<tr>
<td>counting rate [s$^{-1}$]</td>
<td>0.864 ± 0.021</td>
<td>0.069 ± 0.007</td>
</tr>
<tr>
<td>$t$/LRA [%]</td>
<td></td>
<td>7.93 ± 0.87</td>
</tr>
</tbody>
</table>

Table 6.6: Results of the ternary alpha and triton measurement of $^{249}$Cf with telescope 2.

11.5% background contribution, the adopted value is given in table 6.7.

Figure 6.15: 2-dimensional $E_{\text{tot}} - T/a$ spectrum for the $^{249}$Cf(n,f) measurement with telescope 3 (left) and $T/a$ spectrum around the triton peak used to determine the background contribution (right).

To obtain a value for the relative emission probability $t$/LRA, the ternary LRA particles were measured simultaneously with the same telescope. The total energy distribution for
Figure 6.16: Ternary $\alpha$ (left) and triton (right) energy distributions for the $^{249}$Cf neutron induced fission measurement with telescope 3.

<table>
<thead>
<tr>
<th>$\Delta T$ [s]</th>
<th>$\alpha$-particles</th>
<th>tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>183295</td>
<td></td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>16.06 ± 0.11</td>
<td>8.42 ± 0.27</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.70 ± 0.20</td>
<td>8.55 ± 0.43</td>
</tr>
<tr>
<td>total number</td>
<td>548740 ± 10097</td>
<td>47654 ± 3245</td>
</tr>
<tr>
<td>after subtracting background counting rate [s$^{-1}$]</td>
<td>2.99 ± 0.06</td>
<td>0.230 ± 0.018</td>
</tr>
<tr>
<td>t/LRA [%]</td>
<td>7.68 ± 0.61</td>
<td></td>
</tr>
<tr>
<td>weighted average for the tritons</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>8.47 ± 0.19</td>
<td></td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>8.52 ± 0.34</td>
<td></td>
</tr>
<tr>
<td>t/LRA [%]</td>
<td>7.76 ± 0.50</td>
<td></td>
</tr>
<tr>
<td>t/B [$10^{-4}$]</td>
<td>2.13 ± 0.15</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.7: Results of the ternary alpha and triton measurement of $^{249}$Cf with telescope 3, as well as the general results for tritons.

The ternary $\alpha$’s is plotted in figure 6.16 (left). A Gaussian fit starting at 12.5 MeV is made and the results are given in table 6.7.

Combining the yields for tritons and LRA particles, a value for t/LRA is obtained. The final results for the tritons, relative emission probability and parameters of the energy distribution, can be calculated as a weighted average from both measurement campaigns. These values can be found in table 6.7. Since a value for LRA/B is available, a value for t/B can be calculated as well.
6.2 $^{251}\text{Cf}(n,f)$

6.2.1 Characteristics of the sample

The $^{251}\text{Cf}$ sample was prepared at the Institute of Nuclear Chemistry of Mainz University in Germany. The sample has a weight of 5 $\mu$g and a diameter of 4 mm, and consists of californium oxide deposited on a Ti foil, with a diameter of 30 mm and a thickness of 50 $\mu$m. At the moment of the experiment, the sample had an activity of 10.8 MBq. The isotopic compositions at the moment of the isotopic analysis and of the measurement are given in table 6.8.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Isotopic analysis</th>
<th>Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{249}\text{Cf}$</td>
<td>17.65</td>
<td>20.09</td>
</tr>
<tr>
<td>$^{250}\text{Cf}$</td>
<td>35.40</td>
<td>26.71</td>
</tr>
<tr>
<td>$^{251}\text{Cf}$</td>
<td>46.18</td>
<td>53.09</td>
</tr>
<tr>
<td>$^{252}\text{Cf}$</td>
<td>0.77</td>
<td>0.11</td>
</tr>
</tbody>
</table>

Table 6.8: Isotopic composition (in atomic percent) of the $^{251}\text{Cf}$ sample at the different times.

6.2.2 Overview of the experiment

Three surface barrier telescope detectors were used to investigate the neutron induced fission of $^{251}\text{Cf}$. Their characteristics are described in table 6.9. The thermal equivalent neutron flux at the sample position was higher than $10^9$ neutrons/cm$^2$.s.

<table>
<thead>
<tr>
<th>$^{251}\text{Cf}$</th>
<th>$\Delta E$</th>
<th>$E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>telescope 1</td>
<td>300</td>
<td>29.8</td>
</tr>
<tr>
<td>telescope 2</td>
<td>450</td>
<td>62.9</td>
</tr>
<tr>
<td>telescope 3</td>
<td>300</td>
<td>55.1</td>
</tr>
</tbody>
</table>

Table 6.9: Characteristics of the surface barrier telescope detectors used for the $^{251}\text{Cf}$ neutron induced fission measurements. The indicated resolution is for 5.5 MeV $\alpha$-particles.

Telescope 1 was used to detect the ternary alpha particles and $^6\text{He}$ particles. With this E detector also binary fission fragments were measured in order to obtain a value for LRA/B. Both telescopes 2 and 3 were used for the detection of the tritons.

Due to the isotopic composition of the sample (table 6.8), the spontaneous fission yield was not negligible so two measurements had to be performed in each step: one with the neutron beam open, measuring both the neutron induced fission and the spontaneous
fission of all isotopes present in the sample, and a second one with closed neutron beam, in order to determine the contribution of the spontaneous fission of $^{250}$Cf and $^{252}$Cf. In this way, results can be derived for the neutron induced fission only. However, a correction still has to be made for the $^{249}$Cf(n,f) contribution due to the important amount of $^{249}$Cf present in the sample. This can be done easily since experimental results with the $^{249}$Cf sample are available (section 6.1).

As in the case of $^{249}$Cf, no simultaneous measurements with two telescopes are possible, due to the Ti backing. Again a 30 µm thick aluminium foil is placed in front of each ΔE detector to measure the ternary particles.

### 6.2.3 Binary fission

The binary fission measurements were performed with the E detector from telescope 1. The ΔE detector was replaced by a dummy. Figure 6.17 (left) shows the measured binary fission spectrum with open neutron beam. The quality of the spectrum is not as good as in the case of $^{249}$Cf for instance, due to the degradation of the sample with age. After removing of the alpha pile-up peak (cut off at channel 525), the remaining spectrum is extrapolated (figure 6.17, right).

![Figure 6.17: Measured (left) and extrapolated (right) binary fission spectrum for $^{251}$Cf(n,f) with open neutron beam.](image)

As explained, a similar measurement of the binary fission spectrum is done with closed neutron beam, illustrated in figure 6.18. The results for both measurements are given in table 6.10. As can be seen in that table, the uncertainty on the number of fission fragments increases a lot after extrapolation. The uncertainty given on the number of fission fragments is only the statistical uncertainty. To determine the uncertainty on the number of fission fragments after extrapolation, different extrapolations have been made by fitting an exponential function to part of the spectrum remaining after removing the alpha pile-up peak. Depending on the choice of the cut off channel and the fit parameters, the area under the extrapolated spectra is different (with a minimum when the fit is forced through zero). Taking into account these differences, the
uncertainty on the number of fission fragments after extrapolation is determined. The binary fission yield for neutron induced fission is obtained after subtracting the spontaneous fission contribution from the total binary fission yield.

![Graphs showing measured and extrapolated binary fission spectra](image)

Figure 6.18: Measured (left) and extrapolated (right) binary fission spectrum for $^{251}$Cf(n,f) with closed neutron beam.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>beam on</th>
<th>beam off</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>600</td>
<td>6449</td>
</tr>
<tr>
<td>number of fission fragments after extrapolation</td>
<td>2756230 ± 1660</td>
<td>975888 ± 988</td>
</tr>
<tr>
<td>number of fission events</td>
<td>2994863 ± 67684</td>
<td>1099531 ± 37384</td>
</tr>
<tr>
<td>binary fission yield [s$^{-1}$]</td>
<td>1497431.5 ± 33842</td>
<td>549765 ± 18692</td>
</tr>
<tr>
<td>neutron induced BF yield [s$^{-1}$]</td>
<td>2496.72 ± 56.40</td>
<td>85.25 ± 2.90</td>
</tr>
</tbody>
</table>

Table 6.10: Results of the binary fission measurements of $^{251}$Cf.

### 6.2.4 Ternary fission

**Alpha particles**

Telescope 1 was used to determine the parameters of the LRA energy distribution and the LRA counting rate. Again two separate runs were performed, one with open neutron beam and one with the neutron beam closed. The $E_{\text{tot}} - T/a$ spectra for both measurements are shown in figure 6.19.

In these spectra a selection of the LRA particles can be made, based on the $T/a$ identification relation. After this selection and a correction for the energy loss in the Al foil, the total energy spectrum is obtained for both runs, as can be seen in figure 6.20. A Gaussian fit to both spectra, starting at 12.5 MeV, yields an average energy and FWHM as well as the LRA counting rate. These results are written in table 6.11.
Figure 6.19: 2-dimensional $E_{\text{tot}} - T/a$ spectra for the $^{251}\text{Cf}(n,f)$ measurement with telescope 1, left: open neutron beam, right: closed neutron beam.

Figure 6.20: Ternary $\alpha$ energy distributions for the $^{251}\text{Cf}$ neutron induced fission measurement with telescope 1, uncorrected for the $^{249}\text{Cf}(n,f)$ contribution, left: open neutron beam, right: closed neutron beam.

Now values for the average energy, FWHM and the emission probability LRA/B taking into account only the neutron induced fission have to be calculated. Therefore the total energy spectrum of the measurement with closed neutron beam is subtracted from the one with open neutron beam, taking into account the measuring times. In this way a total LRA energy spectrum for the neutron induced fission of $^{240+251}\text{Cf}$ is obtained, plotted in figure 6.21. The results of the Gaussian fit performed to the data above 12.5 MeV can be found in table 6.11.

All the results after correction for the $^{249}\text{Cf}(n,f)$ contribution can be found at the end of this section.

$^{6}\text{He}$ particles

Like the LRA particles, the $^{6}\text{He}$ particles are detected with telescope 1. They are selected in the $E_{\text{tot}} - T/a$ spectra (fig 6.19), based on the relation for $T/a$, once for the measurement
6.2. $^{251}CF(N,F)$

<table>
<thead>
<tr>
<th></th>
<th>beam on</th>
<th>beam off</th>
<th>beam on - off</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>76834</td>
<td>62789</td>
<td>76834</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>15.91 ± 0.10</td>
<td>16.01 ± 0.20</td>
<td>15.91 ± 0.11</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.58 ± 0.15</td>
<td>10.09 ± 0.36</td>
<td>10.60 ± 0.17</td>
</tr>
<tr>
<td>number of ternary $\alpha$'s</td>
<td>471740 ± 8391</td>
<td>14234 ± 646</td>
<td>454337 ± 8542</td>
</tr>
<tr>
<td>counting rate [s$^{-1}$]</td>
<td>6.140 ± 0.109</td>
<td>0.227 ± 0.0103</td>
<td>5.913 ± 0.111</td>
</tr>
<tr>
<td>LRA/B $[10^{-3}]$</td>
<td>2.46 ± 0.07</td>
<td>2.66 ± 0.15</td>
<td>2.45 ± 0.08</td>
</tr>
</tbody>
</table>

Table 6.11: Results of the ternary alpha measurements of $^{251}$Cf with telescope 1, uncorrected for the $^{249}$Cf(n,f) contribution.

![Ternary $\alpha$ energy distribution for the $^{251}$Cf neutron induced fission measurement with telescope 1, beam on - beam off spectrum, uncorrected for the $^{249}$Cf(n,f) contribution.](image)

Figure 6.21: Ternary $\alpha$ energy distribution for the $^{251}$Cf neutron induced fission measurement with telescope 1, beam on - beam off spectrum, uncorrected for the $^{249}$Cf(n,f) contribution.

with open neutron beam and once with closed neutron beam. Two total energy spectra are obtained after energy correction. Due to the limited statistics it was impossible to make a fit to the spectrum measured with closed neutron beam. The sum spectrum (which means beam on - beam off) is plotted in figure 6.22. The parameters of the energy distribution are obtained from a Gaussian fit to all data points, starting at 9.8 MeV, yielding also the counting rate (table 6.12). Combining the counting rates of the $^6$He and LRA particles, a value for the relative emission probability $^6$He/LRA can be calculated, as well as a value for $^6$He/B. These results are given in table 6.12.

All the results after correction for the $^{249}$Cf(n,f) contribution can be found at the end of this section.
Figure 6.22: Ternary $^6$He energy distribution for the $^{251}$Cf neutron induced fission measurement with telescope 1, beam on - beam off spectrum, uncorrected for the $^{249}$Cf(n,f) contribution.

<table>
<thead>
<tr>
<th></th>
<th>beam on - off</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>76834</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>10.86 ± 0.32</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>10.02 ± 0.47</td>
</tr>
<tr>
<td>number $^6$He particles</td>
<td>13926 ± 1038</td>
</tr>
<tr>
<td>counting rate [s$^{-1}$]</td>
<td>0.181 ± 0.014</td>
</tr>
<tr>
<td>$^6$He/LRA [%]</td>
<td>3.07 ± 0.24</td>
</tr>
<tr>
<td>$^6$He/B [$10^{-5}$]</td>
<td>7.52 ± 0.61</td>
</tr>
</tbody>
</table>

Table 6.12: Results of the ternary $^6$He measurements of $^{251}$Cf with telescope 1, uncorrected for the $^{249}$Cf(n,f) contribution.

**Tritons**

To measure the tritons, telescopes 2 and 3 were used, each with a $\Delta E$ detector of different thickness.

- **Telescope 2**

Again two separate runs were performed, one with open neutron beam and one with the beam closed. The $E_{tot} - T/a$ spectra for both measurements are shown in figure 6.23.

In the $T/a$ spectra a selection of the tritons can be made, based on the $T/a$ identification relation ($T/a \approx 10.3$). After this selection and a correction for the energy loss in the Al foil, a total energy spectrum is obtained for both runs. After subtraction, the total triton energy spectrum for the neutron induced fission of $^{249+251}$Cf is obtained, plotted in figure 6.24 (left). A Gaussian fit to all the data (from 6.5 MeV) yields an average energy and
6.2. $^{251}$CF(N,F)

Figure 6.23: 2-dimensional $E_{\text{tot}} - T/a$ spectra for the $^{251}$Cf(n,f) measurement with telescope 2, left: open neutron beam, right: closed neutron beam.

FWHM as well as the triton counting rate. To obtain the correct yield of the tritons, the background contribution had to be determined, equal to about 2.87\%, using the same procedure as illustrated in figure 6.13. The results are written in table 6.13.

Figure 6.24: Ternary $\alpha$ (right) and triton (left) energy distributions for the $^{251}$Cf neutron induced fission measurement with telescope 2, uncorrected for the $^{249}$Cf(n,f) contribution.

In order to obtain a value for the emission probability of the tritons, also the yield of the LRA particles has been determined with the same telescope. Therefore a Gaussian fit is made to the data points, starting at 14 MeV. The total LRA energy spectrum is shown in figure 6.24 (right).

- Telescope 3

With telescope 3, using a $\Delta E$ detector of 55.1 $\mu$m, the measurements are the same as described above. A selection of the LRA particles and the tritons is made in the $T/a$ spectra, based on the $T/a$ relation. After subtracting the results with the beam off from the results with the beam on selection and after correction for the energy loss, the total energy spectra for $\alpha$ particles and tritons are obtained (figure 6.25). A Gaussian fit both
### Table 6.13: Results of the ternary triton measurements of $^{251}$Cf, uncorrected for the $^{249}$Cf(n,f) contribution.

<table>
<thead>
<tr>
<th></th>
<th>Telescope 2</th>
<th>Telescope 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>181243</td>
<td>50279</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>$8.38 \pm 0.17$</td>
<td>$8.67 \pm 0.17$</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>$8.28 \pm 0.25$</td>
<td>$8.52 \pm 0.26$</td>
</tr>
<tr>
<td>number of tritons</td>
<td>$109120 \pm 3081$</td>
<td>$27627 \pm 855$</td>
</tr>
<tr>
<td>triton counting rate [s$^{-1}$]</td>
<td>$0.602 \pm 0.017$</td>
<td>$0.549 \pm 0.017$</td>
</tr>
<tr>
<td>after background corr. [s$^{-1}$]</td>
<td>$0.585 \pm 0.021$</td>
<td>$0.504 \pm 0.037$</td>
</tr>
<tr>
<td>number of ternary $\alpha$'s</td>
<td>$1183396 \pm 21024$</td>
<td>$290570 \pm 5782$</td>
</tr>
<tr>
<td>$\alpha$ counting rate [s$^{-1}$]</td>
<td>$6.529 \pm 0.116$</td>
<td>$5.779 \pm 0.115$</td>
</tr>
<tr>
<td>$t$/LRA [%]</td>
<td>$8.96 \pm 0.36$</td>
<td>$8.72 \pm 0.66$</td>
</tr>
<tr>
<td>weighted average</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>$8.53 \pm 0.12$</td>
<td></td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>$8.40 \pm 0.18$</td>
<td></td>
</tr>
<tr>
<td>$t$/LRA [%]</td>
<td>$8.90 \pm 0.32$</td>
<td></td>
</tr>
<tr>
<td>$t$/B [$10^{-4}$]</td>
<td>$2.18 \pm 0.11$</td>
<td></td>
</tr>
</tbody>
</table>

To all LRA and triton data points yields ternary $\alpha$ and triton counting rates, as well as a triton average energy and FWHM. In this case a background contribution of 8.28% had to be taken into account. The results are given in Table 6.13.

![Figure 6.25: Ternary $\alpha$ (right) and triton (left) energy distributions for the $^{251}$Cf neutron induced fission measurement with telescope 3, uncorrected for the $^{249}$Cf(n,f) contribution.](image)

Within the uncertainties, the results with both telescopes are in agreement with each other. Therefore a weighted average for the average energy and FWHM of the tritons is made, as well as for $t$/LRA. With the value for $t$/LRA and the known value for LRA/B, also $t$/B can be determined. All these values can be found in Table 6.13.

Until now, all the results are valid for the neutron induced fission of $^{249+251}$Cf. Taking into account the total number of atoms and the cross section at 5.4 meV of both isotopes...
(\(\sigma_{n,f}^{249}\ CF\) = 3.78 \times 10^3 \ b, \(\sigma_{n,f}^{251}\ CF\) = 1.17 \times 10^4 \ b [49]), the \(249\)\ CF(n,f) contribution can be determined. Since all the results for \(249\)\ CF are available, the final results for \(251\)\ CF can be calculated (table 6.14), analogously to what is done in section 5.1.4.

<table>
<thead>
<tr>
<th>(\langle E\rangle [\text{MeV}])</th>
<th>LRA</th>
<th>Tritons</th>
<th>(^6\text{He})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{FWHM [MeV]})</td>
<td>15.89 \pm 0.12</td>
<td>8.53 \pm 0.12</td>
<td>10.84 \pm 0.36</td>
</tr>
<tr>
<td>(\text{abs. em. prob.})</td>
<td>10.60 \pm 0.18</td>
<td>8.39 \pm 0.19</td>
<td>9.98 \pm 0.53</td>
</tr>
<tr>
<td>(\text{rel. em. prob. [%]})</td>
<td>((2.41 \pm 0.14) \times 10^{-3})</td>
<td>((2.29 \pm 0.15) \times 10^{-4})</td>
<td>((7.58 \pm 0.69) \times 10^{-6})</td>
</tr>
</tbody>
</table>

Table 6.14: Overview of the final results of the ternary fission measurements of \(251\)\ CF, after correction for the contribution of \(249\)\ CF in the sample.

### 6.3 \(250\)\ CF(SF)

#### 6.3.1 Characteristics of the sample

The \(250\)\ CF sample was prepared at the Lawrence Berkeley National Laboratory (LBNL) in the USA. A spot of californium oxide with a diameter of 6 mm is deposited on a Ti backing foil with a diameter of 14 mm and a thickness of 40 \(\mu\)m. The sample has a mass of about 1 \(\mu\)g and at the moment of the preparation its activity was 4 MBq. Details on the isotopic composition can be found in table 6.15, the measurements started in March 2008.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Isotopic analysis 07/06/2000</th>
</tr>
</thead>
<tbody>
<tr>
<td>(249)\ CF</td>
<td>10.65</td>
</tr>
<tr>
<td>(250)\ CF</td>
<td>77.21</td>
</tr>
<tr>
<td>(251)\ CF</td>
<td>12.14</td>
</tr>
</tbody>
</table>

Table 6.15: Isotopic composition (in atomic percent) of the \(250\)\ CF sample.

#### 6.3.2 Overview of the experiments

Two different telescope detectors were used in order to examine the spontaneous fission of \(250\)\ CF. The characteristics of these telescopes are given in table 6.16. Telescope 1 is optimised for the detection of ternary LRA and \(^6\)\ He particles, while telescope 2 is chosen in order to detect tritons. Both telescopes are consecutively mounted in a vacuum chamber, at a distance of about 35 mm of the \(252\)\ CF sample. For the ternary fission measurements, the \(\Delta E\) detector is shielded with a 25 \(\mu\)m thick Al foil. The binary fission fragments have been measured with the E detector of both telescope 1 and 2.
<table>
<thead>
<tr>
<th>$^{250}\text{Cf}$</th>
<th>$\Delta E$</th>
<th>$E$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>area [mm$^2$]</td>
<td>thickness [µm]</td>
</tr>
<tr>
<td>telescope 1</td>
<td>300</td>
<td>29.8</td>
</tr>
<tr>
<td>telescope 2</td>
<td>300</td>
<td>55.1</td>
</tr>
</tbody>
</table>

Table 6.16: Characteristics of the surface barrier telescope detectors used for the $^{250}\text{Cf}$ spontaneous fission measurements. The indicated resolution is for 5.5 MeV $\alpha$-particles.

### 6.3.3 Binary fission

The binary fission measurements were performed with the E detector of both telescope 1 and 2, while in both cases the $\Delta E$ detector was replaced by a dummy maintaining exactly the same geometry conditions. The spectrum measured with telescope 1 is plotted in figure 6.26 (left). The $\alpha$ pile-up peak due to the radioactive decay of $^{250}\text{Cf}$ in the beginning of the spectrum has to be separated from the fission fragments. A cut off has been made at channel 540. Then the remaining spectrum is extrapolated (figure 6.26, right) and the number of binary fission fragments is obtained after integration of this spectrum. The results of the measurement are given in table 6.17. The uncertainty given on the binary fission yield takes into account the uncertainty due to the extrapolation of the spectrum.

![Figure 6.26: Measured (left) and final extrapolated (right) binary fission spectrum for $^{250}\text{Cf(SF)}$ with telescope 1.](image)

The same procedure is followed to determine the binary fission yield measured with the E detector of telescope 2. In figure 6.27 the measured and the extrapolated spectra are illustrated. Here the cut off is made at channel 313. The results are written in table 6.17.
6.3. $^{250}$CF(SF)

<table>
<thead>
<tr>
<th></th>
<th>telescope 1</th>
<th>telescope 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>4545</td>
<td>2543</td>
</tr>
<tr>
<td>number of fission fragments after extrapolation</td>
<td>322274 ± 568</td>
<td>165147 ± 406</td>
</tr>
<tr>
<td>number of fission events</td>
<td>328171 ± 9845</td>
<td>169933 ± 5098</td>
</tr>
<tr>
<td>binary fission yield [s$^{-1}$]</td>
<td>164089 ± 4923</td>
<td>84967 ± 2549</td>
</tr>
<tr>
<td></td>
<td>36.10 ± 1.08</td>
<td>33.41 ± 1.00</td>
</tr>
</tbody>
</table>

Table 6.17: Results of the binary fission measurements of $^{250}$Cf.

![Graphs showing measured and extrapolated binary fission spectra](image)

Figure 6.27: Measured (left) and final extrapolated (right) binary fission spectrum for $^{250}$Cf(SF) with telescope 2.

6.3.4 Ternary fission

Alpha particles

Telescope 1 was used to determine the parameters of the LRA energy distribution as well as the LRA counting rate. The total 2-dimensional $E_{tot} - T/a$ spectrum is plotted in figure 6.28 (left). In this spectrum a selection of the LRA particles can be made, based on the $T/a$ identification relation ($T/a \approx 24.85$), which is shown in figure 6.28 (right).

After this selection and a correction for the energy loss in the Al foil, the total energy spectrum is obtained as can be seen in figure 6.29. A Gaussian fit to the data points starting at 12.5 MeV yields the average energy and FWHM together with the ternary $\alpha$ counting rate. The results are given in Table 6.18.

Combining the LRA yield with the binary fission yield determined in the previous section, a value for the absolute emission probability LRA/B determined with telescope 1 is obtained (table 6.18).

$^6$He particles

The $^6$He particles are detected with telescope 1, together with the LRA particles. They are selected in the $E_{tot} - T/a$ spectrum (figure 6.28, left), based on the relation for $T/a$
CHAPTER 6. CF ISOTOPES

Figure 6.28: 2-dimensional $E_{\text{tot}} - T/a$ spectrum (left) and selected $\alpha$-particles (right) for the $^{250}$Cf(SF) measurement with telescope 1.

Figure 6.29: Ternary $\alpha$ energy distribution for the $^{250}$Cf spontaneous fission measurement with telescope 1.

<table>
<thead>
<tr>
<th></th>
<th>$\alpha$-particles</th>
<th>$^{6}$He</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>3502370</td>
<td>3502370</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>$15.95 \pm 0.13$</td>
<td>$10.64 \pm 0.30$</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>$10.49 \pm 0.16$</td>
<td>$10.49 \pm 0.54$</td>
</tr>
<tr>
<td>total number</td>
<td>$370231 \pm 3369$</td>
<td>$10140 \pm 1213$</td>
</tr>
<tr>
<td>yield $[s^{-1}]$</td>
<td>$0.106 \pm 0.001$</td>
<td>$(0.290 \pm 0.035) \times 10^{-2}$</td>
</tr>
<tr>
<td>LRA/B $[10^{-5}]$</td>
<td>2.93 $\pm$ 0.10</td>
<td></td>
</tr>
<tr>
<td>$^{6}$He/LRA [%]</td>
<td>2.74 $\pm$ 0.33</td>
<td></td>
</tr>
<tr>
<td>$^{6}$He/B $[10^{-5}]$</td>
<td>8.03 $\pm$ 1.00</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.18: Results of the ternary alpha and $^{6}$He measurement of $^{250}$Cf with telescope 1.
(T/a ≈ 32.15). After energy correction, the total energy spectrum for 6He particles is obtained, shown in figure 6.30.

![Energy Spectrum](image)

Figure 6.30: Ternary 6He energy distribution for the 250Cf spontaneous fission measurement with telescope 1.

The parameters of the energy distribution are obtained from a Gaussian fit to all data points (starting at 9.7 MeV), yielding also the counting rate. Combining the counting rates of the 6He and LRA particles, a value for the relative emission probability 6He/LRA can be calculated, as well as a value for 6He/B. All the results can be found in table 6.18.

**Tritons**

Telescope 2 with a ΔE detector with a thickness of 55.1 μm is used to measure the tritons. The 2-dimensional $E_{\text{rel}} - T/a$ spectrum and the projected $T/a$ spectrum for this measurement are shown in figure 6.31. In the $T/a$ spectrum a selection of the tritons can be made, based on the $T/a$ identification relation ($T/a ≈ 9.34$).

After this selection and a correction for the energy loss in the Al foil, a total energy distribution for the tritons is obtained, plotted in figure 6.32 (left). A Gaussian fit to all the data (from 6 MeV) yields an average energy and FWHM. To obtain the correct yield of the tritons, the background contribution has to be determined and subtracted, as can be seen in figure 6.33 (left). This contribution is equal to about 13.56%. However, the selection made in the $T/a$ spectrum gives us an incomplete peak for the tritons (figure 6.33, right). Therefore 10.73% has to be added to the triton yield. The total triton counting rate is given in table 6.19, together with the other results of the fit.

In order to obtain a value for the relative emission probability of the tritons, the yield of the LRA particles is determined with the same telescope. Therefore a Gaussian fit is made to all the data points, starting at 13 MeV. The total LRA energy spectrum is shown in figure 6.32 (right). All the results are given in table 6.19. There exists a nice agreement
Figure 6.31: 2-dimensional $E_{\text{tot}} - T/a$ spectrum and projected $T/a$ spectrum for the $^{250}$Cf(SF) measurement with telescope 2.

Figure 6.32: Ternary triton (left) and $\alpha$ (right) energy distributions for the $^{250}$Cf spontaneous fission measurement with telescope 2.

<table>
<thead>
<tr>
<th></th>
<th>$\alpha$-particles</th>
<th>tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T$ [s]</td>
<td>$2924554$</td>
<td>$2924554$</td>
</tr>
<tr>
<td>$\langle E \rangle$ [MeV]</td>
<td>$15.89 \pm 0.13$</td>
<td>$8.31 \pm 0.30$</td>
</tr>
<tr>
<td>FWHM [MeV]</td>
<td>$10.44 \pm 0.20$</td>
<td>$8.58 \pm 0.49$</td>
</tr>
<tr>
<td>total number after correction</td>
<td>$292562 \pm 5904$</td>
<td>$20943 \pm 2097$</td>
</tr>
<tr>
<td>yield [s$^{-1}$]</td>
<td>$0.100 \pm 0.002$</td>
<td>$(0.696 \pm 0.088) \times 10^{-2}$</td>
</tr>
<tr>
<td>$t/LRA$ [%]</td>
<td>$6.96 \pm 0.89$</td>
<td></td>
</tr>
<tr>
<td>$t/B$ [10$^{-4}$]</td>
<td>$2.08 \pm 0.27$</td>
<td></td>
</tr>
<tr>
<td>$LRA/B$ [10$^{-3}$]</td>
<td>$2.99 \pm 0.11$</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.19: Results of the ternary alpha and triton fission measurement of $^{250}$Cf with telescope 2.
Figure 6.33: Illustration of the determination of the triton background (left) and the uncomplete peak for the tritons (right) in the $T/a$ spectrum for the $^{252}$Cf spontaneous fission measurement with telescope 2.

between the values for the average energy and FWHM for the $\alpha$-particles measured with telescope 1 and 2.

Since for telescope 2 also a binary fission yield value is available, a value for $t/B$ can be calculated, as well as a value for LRA/B. Comparing this value with the one obtained with telescope 1 (table 6.18), the good agreement is obvious.

6.4 $^{252}$Cf(SF)

6.4.1 Characteristics of the sample

The $^{252}$Cf sample was prepared at the Institute of Nuclear Chemistry of Mainz University in Germany. A spot of californium oxide with a diameter of 15 mm was deposited on a 25 $\mu$m thick aluminium foil. The activity at the moment of preparation (November 2002) was 37 kBq.

6.4.2 Overview of the experiments

The main goal of the spontaneous fission measurement on $^{252}$Cf is to determine a value for the absolute emission probability LRA/B. Therefore a telescope detector optimised for the detection of LRA particles is used. The characteristics of the telescope are given in table 6.20. The telescope is mounted in a vacuum chamber, at a distance of about 35 mm of the $^{252}$Cf sample. For the ternary fission measurement, the $\Delta E$ detector is shielded with a 25 $\mu$m thick Al foil.

The binary fission fragments have been measured with the E detector.
Table 6.20: Characteristics of the surface barrier telescope detector used for the $^{252}\text{Cf}$ spontaneous fission measurement. The indicated resolution is for 5.5 MeV $\alpha$-particles.

6.4.3 Binary fission

The binary fission measurement was performed with the E detector, while the $\Delta E$ detector was replaced by a dummy maintaining exactly the same geometry. The measured spectrum is plotted in figure 6.34 (left). The $\alpha$ pile-up peak due to the radioactive decay of $^{252}\text{Cf}$ in the beginning of the spectrum has to be separated from the fission fragments. A cut off has been made at channel 565. In this case no extrapolation is needed and the final spectrum is plotted in figure 6.34 (right). The number of binary fission fragments is obtained after integration of this spectrum. The results of the measurement are given in table 6.21.

![Binary fission spectrum](image)

Figure 6.34: Measured (left) and final (right) binary fission spectrum for $^{252}\text{Cf}(\text{SF})$.

Table 6.21: Results of the binary fission measurement of $^{252}\text{Cf}$.
6.4.4 Ternary fission

Alpha particles

In order to determine the LRA counting rate the telescope detector, consisting of a $\Delta E$ detector with a thickness of 29.8 $\mu m$ and an $E$ detector of 500 $\mu m$, was used. The total 2-dimensional $E_{tot} - T/a$ spectrum is plotted in figure 6.35 (left). In this spectrum a selection of the $\alpha$-particles can be made, based on the $T/a$ identification relation, which is shown in figure 6.35 (right).

After this selection and a correction for the energy loss in the 25 $\mu m$ thick aluminium foil, the total energy spectrum is obtained, as can be seen in figure 6.36. A Gaussian fit to the data points above 12.5 MeV yields the total number of LRA particles, hence the ternary $\alpha$ counting rate is obtained. The fit provides also values for the average energy and the FWHM. All these results can be found in table 6.22.

![Figure 6.35: 2-dimensional $E_{tot} - T/a$ spectrum (left) and selected $\alpha$-particles (right) for the $^{252}$Cf(SF) measurement.](image)

| $\Delta T$ [s] | 3963262 |
| $\langle E \rangle$ [MeV] | 15.82 ± 0.17 |
| FWHM [MeV] | 10.30 ± 0.24 |
| number of ternary $\alpha$'s | 31706 ± 793 |
| counting rate [$s^{-1}$] | 0.0080 ± 0.0002 |
| LRA/B [$10^{-3}$] | 2.56 ± 0.07 |

Table 6.22: Results of the ternary $\alpha$ measurement of $^{252}$Cf, as well as a value for LRA/B.

Combining the LRA yield with the binary fission yield determined in the previous section, a value for the absolute emission probability LRA/B is obtained (table 6.22).
6.5 Literature survey and comments

As shown in table 6.1, there are four Cf isotopes with a sufficiently long half-life to permit the preparation of samples. Two of these isotopes ($^{250,252}$Cf) have a sufficiently small half-life for spontaneous fission to permit ternary fission measurements, the two others ($^{249,251}$Cf) have a large fission cross section with thermal neutrons and a negligible spontaneous fission decay.

The systematic study of the ternary fission characteristics of californium isotopes by our research group started some years ago. First a few partial results on $^{251}$Cf(n,f) and $^{252}$Cf(SF) have been obtained [51, 23]. In the case of $^{252}$Cf(SF) [23], the energy distribution of the LRA particles was determined, yielding values for the average energy and the FWHM: $\langle E \rangle = (15.7 \pm 0.2)$ MeV, FWHM = $(10.4 \pm 0.2)$ MeV. These values are in very good agreement with the ones determined in this work (see table 6.23).

For $^{251}$Cf(n,f) a first measurement was performed in 2003 [51]. The results mentioned there are in agreement with the results obtained in the new improved experiment.

In tables 6.23 and 6.24 an overview is given of the characteristics of the energy distributions and emission probabilities for the ternary $\alpha$’s and tritons for the different californium isotopes determined in the present thesis. Table 6.25 gives all the results for the $^6$He particles for the different californium isotopes.

Now these results will be compared with some data reported in the literature. Köster [56] gives a lower limit of $5.9 \times 10^{-5}$ for the absolute emission probability of $^6$He particles in $^{249}$Cf(n,f), which is in agreement with our result of $(6.99 \pm 0.66) \times 10^{-5}$.

The neutron induced ternary fission of $^{251}$Cf was measured at Lohengrin by Tsokhanovich et al. [62], however only ternary particles ranging from Li to $^{37}$Si were detected. Furthermore a measurement on $^{251}$Cf(n$_{th}$,f) at Lohengrin was performed by S. Oberstedt et al.
6.5. \textit{LITERATURE SURVEY AND COMMENTS} 

<table>
<thead>
<tr>
<th>Ternary alpha particles</th>
<th>Ternary tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\langle E \rangle) [MeV]</td>
<td>FWHM [MeV]</td>
</tr>
<tr>
<td>(^{249}\text{Cf} (n,f))</td>
<td>16.09 ± 0.18</td>
</tr>
<tr>
<td>(^{251}\text{Cf} (n,f))</td>
<td>15.89 ± 0.12</td>
</tr>
<tr>
<td>(^{249}\text{Cf} (SF))</td>
<td>15.95 ± 0.13</td>
</tr>
<tr>
<td>(^{252}\text{Cf} (SF))</td>
<td>15.82 ± 0.17</td>
</tr>
</tbody>
</table>

Table 6.23: Overview of the characteristics of the measured energy distributions for the ternary \(\alpha\)'s and tritons for the different californium isotopes.

<table>
<thead>
<tr>
<th>(LRA/B \times 10^{-3})</th>
<th>(t/B \times 10^{-4})</th>
<th>(t/LRA) [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{249}\text{Cf} (n,f))</td>
<td>2.77 ± 0.11</td>
<td>2.13 ± 0.15</td>
</tr>
<tr>
<td>(^{251}\text{Cf} (n,f))</td>
<td>2.41 ± 0.14</td>
<td>2.20 ± 0.14</td>
</tr>
<tr>
<td>(^{249}\text{Cf} (SF))</td>
<td>2.93 ± 0.10</td>
<td>2.08 ± 0.27</td>
</tr>
<tr>
<td>(^{252}\text{Cf} (SF))</td>
<td>2.56 ± 0.07</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.24: Overview of the emission probabilities for the ternary \(\alpha\)'s and tritons for the different californium isotopes.

[63]. The characteristics of the energy distribution of the \(^{6}\text{He}\) particles have been measured, with as results a value of (11.0 ± 0.9) MeV for the average energy and (9.6 ± 0.5) MeV for the FWHM. These values are in agreement with our results mentioned in table 6.25.

The ternary fission of \(^{250}\text{Cf}\) has been measured by Wild et al. [64]. Their results are given in table 6.26. It has to be mentioned that only 4023 LRA particles and 273 tritons were detected. Due to this very low statistics, especially in the case of the tritons, a comparison is not straightforward. For the LRA particles, the average energy and FWHM agree within the uncertainties, but the absolute emission probability tends to be higher.

The spontaneous ternary fission of \(^{252}\text{Cf}\) has been measured several times in the past. Some of these results are given in tables 6.27 and 6.28. A recent measurement by Hwang et al. [71] is not mentioned, since the data are completely discrepant. Out of their paper it becomes clear that their correction for the energy loss in the detector is not done in a correct way.

<table>
<thead>
<tr>
<th>(\langle E \rangle) [MeV]</th>
<th>FWHM [MeV]</th>
<th>(^{6}\text{He}/B \times 10^{-3})</th>
<th>(^{6}\text{He}/LRA) [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{249}\text{Cf} (n,f))</td>
<td>10.99 ± 0.32</td>
<td>10.35 ± 0.60</td>
<td>6.99 ± 0.66</td>
</tr>
<tr>
<td>(^{251}\text{Cf} (n,f))</td>
<td>10.84 ± 0.36</td>
<td>9.98 ± 0.53</td>
<td>7.58 ± 0.69</td>
</tr>
<tr>
<td>(^{249}\text{Cf} (SF))</td>
<td>10.64 ± 0.30</td>
<td>10.49 ± 0.54</td>
<td>8.03 ± 1.00</td>
</tr>
</tbody>
</table>

Table 6.25: Overview of the characteristics of the measured energy distributions and the emission probabilities for the ternary \(^{6}\text{He}\) particles for the different californium isotopes.
### Table 6.26: Overview of the results obtained by Wild et al. [64] for the spontaneous fission measurement of $^{252}$Cf.

The values for the parameters of the energy distribution of LRA particles written in both tables are in good agreement with the ones mentioned in table 6.23, determined in this work. The determination of a value for LRA/B was the most important, since only very few, old results are available for this value. Our new result of $(2.56 \pm 0.07) \times 10^{-3}$ shows the best agreement with the value determined by Wild (table 6.27).

In order to complete table 6.24, values for the triton emission probabilities of $^{252}$Cf(SF) are needed. Therefore a weighted average is made of the literature results for $t$/LRA mentioned in tables 6.27 and 6.28. Then a value of $(7.99 \pm 0.10)\%$ is obtained, yielding an absolute emission probability $t$/B of $(2.05 \pm 0.06) \times 10^{-4}$. The same is done for the $^6$He particles. In this way results for the $^6$He emission probabilities are obtained: $^6$He/LRA = $(3.24 \pm 0.08)\%$ and $^6$He/B = $(8.29 \pm 0.31) \times 10^{-5}$. These average values obtained from literature results will be used for the discussion in chapter 7, where they will be mentioned between brackets.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle E_0 \rangle$ [MeV]</td>
<td>$15.6 \pm 0.2$</td>
<td>$16.0 \pm 0.5$</td>
<td>$16.0 \pm 0.2$</td>
<td>$15.8 \pm 0.1$</td>
</tr>
<tr>
<td>FWHM$_0$ [MeV]</td>
<td>$10.3 \pm 0.5$</td>
<td>$10.2 \pm 0.3$</td>
<td>$10.2 \pm 0.1$</td>
<td>$-$</td>
</tr>
<tr>
<td>$\langle E_t \rangle$ [MeV]</td>
<td>$7.7 \pm 0.4$</td>
<td>$8 \pm 1$</td>
<td>$8.0 \pm 0.3$</td>
<td>$8.3 \pm 0.1$</td>
</tr>
<tr>
<td>FWHM$_t$ [MeV]</td>
<td>$8.2 \pm 0.9$</td>
<td>$6.2 \pm 0.6$</td>
<td>$7.0 \pm 0.1$</td>
<td>$-$</td>
</tr>
<tr>
<td>$\langle E_{^6He} \rangle$ [MeV]</td>
<td>$8.2 \pm 0.9$</td>
<td>$13 \pm 1$</td>
<td>$12 \pm 0.5$</td>
<td>$11.4 \pm 0.3$</td>
</tr>
<tr>
<td>FWHM$_{^6He}$ [MeV]</td>
<td>$8 \pm 1$</td>
<td>$8 \pm 1$</td>
<td>$8 \pm 1$</td>
<td>$10.6 \pm 0.3$</td>
</tr>
<tr>
<td>LRA/B $(10^{-3})$</td>
<td>$2.94 \pm 0.42$</td>
<td>$3.27 \pm 0.10$</td>
<td>$-$</td>
<td>$-$</td>
</tr>
<tr>
<td>$t$/LRA [%]</td>
<td>$8.50 \pm 2.09$</td>
<td>$5.81 \pm 0.26$</td>
<td>$8.46 \pm 0.28$</td>
<td>$8.36 \pm 0.12$</td>
</tr>
<tr>
<td>$^6$He/LRA [%]</td>
<td>$2.4 \pm 0.5$</td>
<td>$2.63 \pm 0.18$</td>
<td>$3.66 \pm 0.12$</td>
<td>$-$</td>
</tr>
</tbody>
</table>

Table 6.27: Overview of results for the spontaneous fission measurement of $^{252}$Cf found in the literature (part 1).
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>( \langle E_\alpha \rangle ) [MeV]</td>
<td>15.8 ± 0.1</td>
<td>12.5 ± 3.7</td>
<td>12.7 ± 2.8</td>
<td>15.7 ± 0.1</td>
</tr>
<tr>
<td>FWHM_\alpha [MeV]</td>
<td>10.3 ± 0.1</td>
<td>12.1 ± 0.2</td>
<td>9.2 ± 0.2</td>
<td>10.6 ± 0.2</td>
</tr>
<tr>
<td>( \langle E_{^6\text{He}} \rangle ) [MeV]</td>
<td>2.0 ± 0.7</td>
<td>3.06 ± 0.15</td>
<td>3.20 ± 0.10</td>
<td></td>
</tr>
<tr>
<td>FWHM_{^6\text{He}} [MeV]</td>
<td></td>
<td></td>
<td></td>
<td>12.5 ± 0.5</td>
</tr>
<tr>
<td>LRA/B ( (10^{-3}) )</td>
<td></td>
<td></td>
<td></td>
<td>9.0 ± 0.5</td>
</tr>
<tr>
<td>(^6\text{He}/LRA [%] )</td>
<td>3.06 ± 0.15</td>
<td></td>
<td>3.20 ± 0.10</td>
<td>4.1 ± 0.5</td>
</tr>
</tbody>
</table>

Table 6.28: Overview of results for the spontaneous fission measurement of \(^{252}\text{Cf}\) found in the literature (part 2).
Chapter 7
Discussion

7.1 Energy distributions

Table 7.1 gives an overview of the values for the average energy and the FWHM for the ternary α’s and tritons for the different curium and californium isotopes discussed in this work, as well as for all other isotopes measured by our research group. Due to the fact that all data were obtained basically using the same methodology, an intercomparison is significant. Table 7.2 gives an overview of the results for the parameters of the energy distribution for the 6He particles, obtained in this work.

7.1.1 Average energy

It is interesting to compare the average energies for the ternary α’s and the tritons in order to determine a weighted average. In figure 7.1 these values for $\langle E \rangle$ are plotted for LRA particles (left) and tritons (right) as a function of the fissility parameter $Z^2/A$ of the fissioning system (discussed in section 1.3).

An overview with the values of the parameter $Z^2/A$ of the compound nucleus is given in table 7.3. In figure 7.1 a distinction is made between the data from neutron induced fission (red squares) and spontaneous fission (green triangles). For the average energy there seems to be no significant difference between both neutron induced and spontaneous fission and we can conclude that the average energy is $(16.0 \pm 0.1)$ MeV for the ternary α’s and $(8.4 \pm 0.1)$ MeV for the tritons. These results are in agreement with the systematic behaviour previously observed e.g. by Wagemans [2]. For the 6He particles, there are much less data available. However, having a look at table 7.2, also the average energy for the 6He particles is compatible within the experimental uncertainties with a constant value of $(10.8 \pm 0.2)$ MeV.

Of course one wants to understand why the most probable energy for a given ternary particle remains constant within the experimental uncertainties for all fissioning systems considered. From trajectory calculations it follows that the initial energy of the ternary particles is low (e.g. a few MeV for the alpha’s). Therefore the determining factors for the final energy are the angle of emission of the particles and the focusing Coulomb field.
### Table 7.1: Overview of the characteristics of the energy distributions for the ternary α’s and tritons for the different isotopes measured by our research group.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>(n,f)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>233Cm</td>
<td>16.14 ± 0.10</td>
<td>10.32 ± 0.15</td>
<td>8.15 ± 0.31</td>
<td>8.13 ± 0.47</td>
</tr>
<tr>
<td>245Cm</td>
<td>16.35 ± 0.15</td>
<td>10.10 ± 0.20</td>
<td>8.40 ± 0.25</td>
<td>7.76 ± 0.38</td>
</tr>
<tr>
<td>247Cm</td>
<td>16.01 ± 0.13</td>
<td>10.37 ± 0.24</td>
<td>8.55 ± 0.27</td>
<td>7.52 ± 0.33</td>
</tr>
<tr>
<td>249Cf</td>
<td>16.09 ± 0.18</td>
<td>10.64 ± 0.27</td>
<td>8.47 ± 0.19</td>
<td>8.52 ± 0.34</td>
</tr>
<tr>
<td>251Cf</td>
<td>15.89 ± 0.12</td>
<td>10.60 ± 0.18</td>
<td>8.53 ± 0.12</td>
<td>8.39 ± 0.19</td>
</tr>
<tr>
<td>239Th</td>
<td>15.4 ± 0.3</td>
<td>9.4 ± 0.4</td>
<td>8.3 ± 0.1</td>
<td>7.1 ± 0.2</td>
</tr>
<tr>
<td>233U[58]</td>
<td>16.1 ± 0.2</td>
<td>9.7 ± 0.2</td>
<td>6.8 ± 0.2</td>
<td></td>
</tr>
<tr>
<td>235U[23, 58]</td>
<td>16.0 ± 0.1</td>
<td>9.5 ± 0.2</td>
<td>8.3 ± 0.1</td>
<td>7.2 ± 0.2</td>
</tr>
<tr>
<td>237Np[61]</td>
<td>15.8 ± 0.5</td>
<td>9.9 ± 0.5</td>
<td>7.7 ± 0.48</td>
<td></td>
</tr>
<tr>
<td>239Pu[58]</td>
<td>15.9 ± 0.2</td>
<td>10.1 ± 0.2</td>
<td>8.5 ± 0.1</td>
<td>6.9 ± 0.2</td>
</tr>
<tr>
<td>241Pu[58]</td>
<td>15.9 ± 0.1</td>
<td>9.8 ± 0.1</td>
<td>8.4 ± 0.1</td>
<td>7.3 ± 0.3</td>
</tr>
<tr>
<td>241Am[59]</td>
<td>15.8 ± 0.1</td>
<td>10.1 ± 0.2</td>
<td>8.3 ± 0.1</td>
<td>7.3 ± 0.3</td>
</tr>
<tr>
<td>243Am[59]</td>
<td>15.8 ± 0.3</td>
<td>10.0 ± 0.5</td>
<td>7.89 ± 0.48</td>
<td></td>
</tr>
<tr>
<td>SF</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>233Cm</td>
<td>15.99 ± 0.10</td>
<td>9.99 ± 0.29</td>
<td>8.05 ± 0.29</td>
<td>7.77 ± 0.49</td>
</tr>
<tr>
<td>245Cm</td>
<td>16.41 ± 0.20</td>
<td>9.73 ± 0.28</td>
<td>8.05 ± 0.34</td>
<td>7.77 ± 0.47</td>
</tr>
<tr>
<td>247Cm</td>
<td>15.97 ± 0.12</td>
<td>10.03 ± 0.14</td>
<td>8.86 ± 0.18</td>
<td>7.47 ± 0.29</td>
</tr>
<tr>
<td>251Cf</td>
<td>15.95 ± 0.13</td>
<td>10.49 ± 0.16</td>
<td>8.31 ± 0.30</td>
<td>8.58 ± 0.49</td>
</tr>
<tr>
<td>252Cf</td>
<td>15.82 ± 0.17</td>
<td>10.30 ± 0.24</td>
<td>8.58 ± 0.49</td>
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</tr>
<tr>
<td>238Pu[34]</td>
<td>15.91 ± 0.22</td>
<td>9.76 ± 0.24</td>
<td>8.58 ± 0.49</td>
<td></td>
</tr>
<tr>
<td>240Pu[34]</td>
<td>16.55 ± 0.27</td>
<td>9.54 ± 0.41</td>
<td>8.58 ± 0.49</td>
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</tr>
<tr>
<td>242Pu[34]</td>
<td>15.79 ± 0.21</td>
<td>9.25 ± 0.24</td>
<td>8.58 ± 0.49</td>
<td></td>
</tr>
<tr>
<td>244Pu[34]</td>
<td>16.04 ± 0.25</td>
<td>10.25 ± 0.37</td>
<td>8.58 ± 0.49</td>
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</table>

### Table 7.2: Overview of the characteristics of the energy distributions for the ternary 6He particles determined in this work.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>⟨E⟩ [MeV]</th>
<th>FWHM [MeV]</th>
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</thead>
<tbody>
<tr>
<td>(n,f)</td>
<td></td>
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<tr>
<td>233Cm</td>
<td>10.94 ± 0.47</td>
<td>10.01 ± 0.58</td>
</tr>
<tr>
<td>240Cf</td>
<td>10.99 ± 0.32</td>
<td>10.35 ± 0.60</td>
</tr>
<tr>
<td>251Cf</td>
<td>10.84 ± 0.36</td>
<td>9.98 ± 0.53</td>
</tr>
<tr>
<td>SF</td>
<td></td>
<td></td>
</tr>
<tr>
<td>234Cm</td>
<td>10.25 ± 0.51</td>
<td>9.89 ± 0.78</td>
</tr>
<tr>
<td>252Cf</td>
<td>10.64 ± 0.30</td>
<td>10.49 ± 0.54</td>
</tr>
</tbody>
</table>

"CHAPTER 7. DISCUSSION"
Figure 7.1: Overview of the values for the average energy for the ternary α's (left) and tritons (right) as a function of the fissility parameter of the fissioning nucleus.

<table>
<thead>
<tr>
<th></th>
<th>$Z$</th>
<th>$Z^2/A$</th>
<th>$Z^2/A^{1/3}$</th>
<th>$4Z - A$</th>
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<tbody>
<tr>
<td>$^{234}$Cm</td>
<td>96</td>
<td>37.77</td>
<td>1474.84</td>
<td>140</td>
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<tr>
<td>$^{236}$Cm</td>
<td>96</td>
<td>37.46</td>
<td>1470.84</td>
<td>138</td>
</tr>
<tr>
<td>$^{238}$Cm</td>
<td>96</td>
<td>37.16</td>
<td>1466.87</td>
<td>136</td>
</tr>
<tr>
<td>$^{250}$Cf</td>
<td>98</td>
<td>38.42</td>
<td>1524.54</td>
<td>142</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td>98</td>
<td>38.11</td>
<td>1520.50</td>
<td>140</td>
</tr>
<tr>
<td>$^{230}$Th</td>
<td>90</td>
<td>35.22</td>
<td>1322.03</td>
<td>130</td>
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<td>$^{232}$Pa</td>
<td>91</td>
<td>35.69</td>
<td>1347.68</td>
<td>132</td>
</tr>
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<td>$^{234}$U</td>
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<td>1373.53</td>
<td>134</td>
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<td>35.86</td>
<td>1369.64</td>
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<td>$^{238}$Np</td>
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<td>$^{238}$Pu</td>
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<td>1425.82</td>
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<td>$^{240}$Pu</td>
<td>94</td>
<td>36.82</td>
<td>1421.84</td>
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</tr>
<tr>
<td>$^{242}$Pu</td>
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<td>1417.92</td>
<td>134</td>
</tr>
<tr>
<td>$^{244}$Pu</td>
<td>94</td>
<td>36.21</td>
<td>1414.03</td>
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<tr>
<td>$^{242}$Am</td>
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<td>37.29</td>
<td>1448.25</td>
<td>138</td>
</tr>
<tr>
<td>$^{244}$Am</td>
<td>95</td>
<td>36.99</td>
<td>1444.28</td>
<td>136</td>
</tr>
</tbody>
</table>

Table 7.3: Overview of the $Z$, $Z^2/A$, $Z^2/A^{1/3}$ and $4Z - A$ values for the different fissioning systems studied by our research group.

between both fission fragments.
As shown in section 2.5, it has been observed that the angular distribution for the ternary α-particles is strongly peaked, which appeared to be rather stable when going from $^{235}$U($n_{th},f$) to $^{252}$Cf(SF). Furthermore, the ternary alpha energy as a function of the emission angle is very similar in both cases, so the focusing Coulomb field as well as the conus of emission seem to be fairly stable going from $^{235}$U($n_{th},f$) to $^{252}$Cf(SF). This stability is probably
a consequence of the stability of the heavy fragment peak in the fission fragments mass distribution (section 1.8). As the mass of the heavy fragment remains almost constant, this will be also the case for the charge $Z$ of the heavy fragment, which has a strong impact on the Coulomb field.

The explanation described above is for ternary alpha particles. With the observation of a practically constant average energy for the tritons, a similar reasoning will be valid for the other ternary particles.

Having a look at the average energy of the LRA particles and the $^6$He particles, another observation can be made. Both particles have the same $Z$ value, while the average energy is about 5 MeV lower for $^6$He particles than for $\alpha$-particles. This confirms the phenomenon already observed by Wagemans [2] that for a given $Z$ value, the most probable particle energy tends to decrease for the heavier particles. This can be understood by assuming very similar initial conditions for the various ternary particles. Indeed, for a given initial energy, the heavier particles will move more slowly, allowing the fission fragments to move further away before the particle is fully accelerated.

7.1.2 FWHM

As the FWHM data are sensitive to energy loss effects, experimental resolution and energy limits of the data points used in the fit, comparisons with FWHM data are only reliable for measurements done under comparable conditions, which is the case in the present selection. Figure 7.2 (left) displays the FWHM data for ternary $\alpha$-particles as a function of $Z^2/A$ of the fissioning system.

![Figure 7.2: Overview of the values for the FWHM for the ternary $\alpha$’s (left) and tritons (right) as a function of the fissility parameter of the fissioning nucleus.](image)

This figure demonstrates a linear increase of the FWHM with increasing $Z^2/A$, as previously observed by Wagemans [2] on a much smaller data base. This increase of the width with $Z^2/A$ can be understood as follows. From trajectory calculations we know that the broadening of the energy distribution of the ternary particles is due to the Coulomb field that amplifies small differences occurring in the initial LRA kinetic energy spectrum. These
initial differences result from fluctuations in the scission shapes, which become more important with increasing deformation energy. Since the deformation energy increases with increasing $Z^2/A$, the observed correlation of the width of the energy distributions with $Z^2/A$ is not surprising.

Another striking observation appears when we compare the FWHM for the ternary $\alpha$ energy distribution in the case of spontaneous and neutron induced fission. The FWHM for the same fissioning system is systematically about 0.3 MeV smaller for spontaneous fission than for neutron induced fission. This is illustrated in figure 7.2 (left) by a linear fit through the spontaneous fission data as well as a linear fit through the neutron induced fission data. This phenomenon could be demonstrated for the first time thanks to our systematic study involving 9 spontaneously fissioning nuclides and 13 neutron induced fission reactions.

Both characteristics of the FWHM mentioned above are also known for the energy distributions of fission fragments [2]. The observed broadening of the total fission fragment kinetic energy distribution with increasing $Z^2/A$ can also be explained by the fluctuations of the scission shapes. An illustration of this property is shown in figure 7.3. In this figure, the variance $\sigma_{EK}^2$ of the total kinetic energy distribution is chosen as a measure for the width of the total kinetic energy distribution.

![Figure 7.3: Variance $\sigma_{EK}^2$ of the total kinetic energy distribution as a function of $Z^2/A$ of the fissioning nucleus [2].](image)

Furthermore, the difference in FWHM for spontaneous and neutron induced ternary fission confirms the well-known phenomenon already observed for fission fragments, that excitation energy enlarges kinetic energy distributions. This is illustrated in figure 7.4,
which shows the increase of the variance $\sigma_{E_K}^2$ of the total kinetic energy distribution with increasing incident alpha energy for $^{235}\text{U}(\alpha,f)$ (triangles) and $^{236}\text{U}(\alpha,f)$ (points).

![Graph showing variance $\sigma_{E_K}^2$ vs. incident alpha energy $E_\alpha$.](image)

Figure 7.4: Variance $\sigma_{E_K}^2$ of the total kinetic energy distribution as a function of incident alpha energy for $^{235}\text{U}(\alpha,f)$ (triangles) and $^{236}\text{U}(\alpha,f)$ (points) [2].

Figure 7.2 (right) displays the FWHM data for tritons for spontaneous and neutron induced fission. Again a linear increase of the width with increasing $Z^2/A$ is observed. However, here the uncertainties on the FWHM are too large to observe such a small difference between spontaneous and neutron induced fission. Also for the $^6\text{He}$ particles (table 7.2), the uncertainties on the values are too large to permit a conclusion.

### 7.2 Emission probabilities

Table 7.4 gives an overview of the absolute and relative emission probabilities for the ternary $\alpha$'s and tritons for the different curium and californium isotopes discussed in this work, as well as for all other isotopes measured by our research group. Table 7.5 gives an overview of the results for the emission probabilities for the $^6\text{He}$ particles, measured by our research group. The values between brackets in both tables are a weighted average of different literature results for $^{252}\text{Cf}(\text{SF})$ (section 6.5).

The correlation between the ternary particle emission probabilities and experimental parameters is an interesting and instructive matter to examine. Now one can ask the question which experimental parameters are investigated and why? In general one can say that there are three straightforward parameters. First of all there is the parameter $Z^2/A$, which is a measure of the fissility of the fissioning system (see section 1.3). Another parameter that is often discussed is the Coulomb parameter $Z^2/A^{1/3}$. This is an obvious comparison taking into account that the ternary particles are ejected due to the influence of the Coulomb field between both heavy fission
### 7.2. EMISSION PROBABILITIES

<table>
<thead>
<tr>
<th></th>
<th>Ternary alpha particles</th>
<th>Ternary tritons</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
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<td>t/B $[10^{-3}]$</td>
</tr>
<tr>
<td>(n,f)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{248}$Cm</td>
<td>2.43 ± 0.08</td>
<td>1.89 ± 0.15</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>2.15 ± 0.12</td>
<td>1.85 ± 0.21</td>
</tr>
<tr>
<td>$^{247}$Cm</td>
<td>1.85 ± 0.10</td>
<td>1.84 ± 0.20</td>
</tr>
<tr>
<td>$^{249}$Cf</td>
<td>2.77 ± 0.11</td>
<td>2.13 ± 0.15</td>
</tr>
<tr>
<td>$^{251}$Cf</td>
<td>2.41 ± 0.14</td>
<td>2.20 ± 0.14</td>
</tr>
<tr>
<td>$^{229}$Th[2, 59]</td>
<td>2.05 ± 0.10</td>
<td>0.85 ± 0.15</td>
</tr>
<tr>
<td>$^{231}$Pa[60]</td>
<td>1.67 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>$^{233}$U[58]</td>
<td>2.05 ± 0.07</td>
<td>1.14 ± 0.05</td>
</tr>
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<td>$^{235}$U[58]</td>
<td>1.60 ± 0.03</td>
<td>1.08 ± 0.04</td>
</tr>
<tr>
<td>$^{237}$Np[60]</td>
<td>1.94 ± 0.17</td>
<td></td>
</tr>
<tr>
<td>$^{239}$Pu[58]</td>
<td>2.09 ± 0.07</td>
<td>1.42 ± 0.07</td>
</tr>
<tr>
<td>$^{241}$Pu[58]</td>
<td>1.75 ± 0.05</td>
<td>1.41 ± 0.06</td>
</tr>
<tr>
<td>$^{241}$Am[2, 59]</td>
<td>2.24 ± 0.09</td>
<td>1.65 ± 0.10</td>
</tr>
<tr>
<td>$^{243}$Am[2]</td>
<td>1.72 ± 0.10</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{241}$Cm</td>
<td>3.16 ± 0.09</td>
<td>1.89 ± 0.24</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>2.49 ± 0.12</td>
<td>1.72 ± 0.24</td>
</tr>
<tr>
<td>$^{247}$Cm</td>
<td>2.30 ± 0.10</td>
<td>1.79 ± 0.21</td>
</tr>
<tr>
<td>$^{250}$Cf</td>
<td>2.93 ± 0.10</td>
<td>2.08 ± 0.27</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td>2.56 ± 0.07</td>
<td>(2.05 ± 0.06)</td>
</tr>
<tr>
<td>$^{238}$Pu[34]</td>
<td>2.76 ± 0.13</td>
<td></td>
</tr>
<tr>
<td>$^{240}$Pu[34]</td>
<td>2.51 ± 0.14</td>
<td></td>
</tr>
<tr>
<td>$^{242}$Pu[34]</td>
<td>2.17 ± 0.07</td>
<td></td>
</tr>
<tr>
<td>$^{244}$Pu[34]</td>
<td>1.71 ± 0.09</td>
<td></td>
</tr>
</tbody>
</table>

Table 7.4: Overview of the emission probabilities for the ternary $\alpha$’s and tritons for the different isotopes measured by our research group. The values between brackets are a weighted average of different literature results for $^{252}$Cf(SF) (section 6.5).

fragments. The third parameter is the deformation energy. We know that ternary particles are emitted at the expense of the deformation energy. However for this quantity no direct experimental values are available. Therefore two different approximations can be made. A first one was proposed by Wild et al. [64]. It consists in plotting the ternary particle emission probability as a function of the quantity $Q - TKE$, where $Q$ is the average fission reaction energy for a given fragment pair and $TKE$ the average measured total kinetic energy of the two fragments. Indeed, $Q - TKE$ represents the average total excitation energy of the scissioning nucleus. In the case of spontaneous fission this excitation energy is mainly composed of the deformation energy since the internal heating is quite negligible. A second approximative parameter to correlate the emission probability with the deformation energy is the average neutron multiplicity. These are in general well-known
and experimentally determined values. However, most of the neutrons are emitted by the fission fragments, while the ternary particles are emitted from the neck region between the two fragments. Therefore the number of emitted neutrons is a measure of the deformation energy of the fission fragments thus indirectly a measure of the deformation energy of the fissioning system.

A special case is the correlation between the experimental radioactive α-decay constant λ and the emission probability, which is likely to exist in the framework of Carjan’s theoretical model [36]. Furthermore also semi-empirical approaches exist, e.g. the oldest one of Halpern [30], where the emission probabilities are correlated with $4Z - A$. Now the different parameters described above will be discussed more in detail.

As mentioned above, Halpern proposed a semi-empirical relation between the LRA emission probability $Y$ and $Z$ and $A$ of the fissioning nucleus, namely:

$$Y(Z, A) = [a + b(4Z - A)] \times 10^{-3}. \tag{7.1}$$

In figure 7.5 the absolute LRA emission probabilities are plotted as a function of $4Z - A$; a distinction is made between the spontaneous fission data and the neutron induced fission data. By applying a linear fit through our data, values for the parameters $a$ and $b$ are obtained for the slope and the intercept out of relation 7.1. These values are given in table 7.6 for both the spontaneous fission and the neutron induced fission data.

<table>
<thead>
<tr>
<th></th>
<th>a</th>
<th>b</th>
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<tbody>
<tr>
<td>LRA/B (SF)</td>
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<tr>
<td>LRA/B ($n_{th}$)</td>
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<td>0.09</td>
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<tr>
<td>LRA/B (SF)</td>
<td>-13.3</td>
<td>0.12</td>
</tr>
</tbody>
</table>

Table 7.6: Values for the parameters $a$ and $b$, in analogy with Halpern [30].
7.2. Emission Probabilities

Figure 7.5: Overview of the LRA emission probability as a function of 4Z-A.

The values obtained by fitting through the spontaneous fission data, are in very good agreement with the values proposed by Halpern for spontaneous fission (based on the scarce data available 40 years ago), namely \( Y = [0.12(4Z - A) - 13.3] \times 10^{-3} \).

The observation that the value for LRA/B is generally larger for the larger 4Z - A values is not that surprising. The parameter 4Z - A contains both Z and A, also present in the fissility parameter \( Z^2/A \) and the Coulomb parameter \( Z^2/A^{1/3} \). Having a look at table 7.3, the values of 4Z - A show the same trend as the values for \( Z^2/A \) and \( Z^2/A^{1/3} \), and, as will be demonstrated, the ternary emission probabilities increase with increasing values of the fissility and Coulomb parameter. From energetic point of view it is known that the difference between the total mean energy release and the mean fragment kinetic energy is about 20 MeV for binary thorium fission, while it increases to about 35 MeV for \(^{252}\text{Cf}\). Since the average energy cost for \( \alpha \) particle emission is about 20 MeV (see section 2.6.2) in all species, one expects ternary particle emission to be more probable in nuclei with larger Z values, in agreement with the observation.

Let us come back now to the correlation between the emission probabilities and the fissility parameter \( Z^2/A \). Already in 1962, Nobles correlated the total number of ternary particles per fission T/B with the parameter \( Z^2/A \) of the fissioning system.

In figure 7.6 the absolute emission probabilities for LRA particles (left) and tritons (right) are plotted as a function of \( Z^2/A \).

These figures allow several observations. First of all the general trend is demonstrated that both \( \alpha \) and triton emission probabilities increase with increasing fissility, as expected, since ternary fission proceeds at the expense of the deformation energy, which is proportional to \( Z^2/A \). However, it has to be added that a rather good correlation for the triton
particles can be observed, while for the ternary $\alpha$-particles, still strong fluctuations are seen.

Another observation is that for the same fissioning system the ternary $\alpha$ emission probability is about 20% higher for spontaneous fission than for neutron induced fission, so the increase of the excitation energy in the case of neutron induced fission seems to result in a decrease of the LRA emission probability, which was hard to understand up to now. On the other hand, taking a look at the right part of figure 7.6, the triton emission probability is hardly affected by this increase of excitation energy, as expected, since the energy needed to emit a ternary particle is mainly taken at the expense of the deformation energy.

We have proposed for the first time an explanation for this phenomenon by the strong impact of the alpha cluster preformation probability on the ternary $\alpha$ emission. This factor, called $S_\alpha$, is introduced in section 2.6.5. As explained, the alpha cluster preformation probability factor $S_\alpha$ can be determined experimentally for the ground state (spontaneous fission). In table 7.7 the different $S_\alpha$ values are given. No uncertainties are given, however one has to take into account that these values are model-dependent and can vary with the calculations of the potential.

With this information, in fact $(\text{LRA}/B)/S_\alpha$ corresponds to the escape probability of an $\alpha$-particle from the scissioning nucleus. Therefore, a new plot (figure 7.7) is made for the LRA particles, showing $(\text{LRA}/B)/S_\alpha$ as a function of $Z^2/A$.

In this figure it can be seen that the strong fluctuations, visible in the left part of figure 7.6, are mostly disappeared, and the data vary now in an almost smooth way as a function of $Z^2/A$ as triton particles do. The spontaneous fission data are still higher than the corresponding neutron induced fission data, which can be explained as follows. As mentioned before, the factor $S_\alpha$ can be calculated for the ground state, thus in the case of spontaneous fission. These same values are used to correct the neutron induced fission data. However, when the fissioning nucleus is formed after capture of a neutron, $S_\alpha$ is likely to decrease due to the excitation energy. So the value of $S_\alpha$ used in the case of the
7.2. EMISSION PROBABILITIES

<table>
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<tr>
<th>$^\alpha$Cm</th>
<th>$^{235}$Cf</th>
<th>$^{230}$Cf</th>
<th>$^{235}$Th</th>
<th>$^{232}$Pa</th>
<th>$^{234}$U</th>
<th>$^{236}$U</th>
<th>$^{238}$Np</th>
<th>$^{238}$Pu</th>
<th>$^{240}$Pu</th>
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<td>10.91</td>
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<td>no alpha</td>
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<td>26.35</td>
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<td>29.57</td>
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</tr>
</tbody>
</table>

Table 7.7: Overview of the different values for the factors $S_\alpha$ and $S_{\alpha He}$.

(LRA/B)/$S_\alpha$ as a function of $Z^2/A$ of the compound nucleus.

neutron induced fission is too high, which leads to the observed decrease of (LRA/B)/$S_\alpha$. Since no cluster preformation is involved in the triton emission process, a similar effect does not occur here, as observed.

In figure 7.8 (left), the ratio of the ternary $\alpha$ emission probability for neutron induced and spontaneous fission of all the available compound systems is given as a function of the excitation energy $E_{exc}$. Fig. 7.8 (right) shows the ratio of the triton emission probability
for neutron induced and spontaneous fission of all the available compound systems.

![Graph](image)

Figure 7.8: Ratio of the ternary α (left) and triton (right) emission probabilities for neutron induced and spontaneous fission of the available isotopes as a function of the excitation energy.

The right part of figure 7.8 clearly shows that increasing the excitation energy with about 6.5 MeV hardly results in an enhancement of the triton emission. This is in line with the idea that ternary particle emission is done at the expense of the deformation energy, which is hardly affected by a moderate increase in excitation energy.

Now one can write the emission probability ratio between (n,f) and (SF) for the same compound nucleus for LRA particles and tritons in the following way:

\[
\frac{\text{em.prob.}(n,f)}{\text{em.prob.}(SF)} = 1 + a_{exc} E_{exc}. \tag{7.2}
\]

The parameter \(a_{exc}\) is assumed to be independent of the mass of the fissioning nucleus. From linear fits performed on the experimental points of fig. 7.8 (left and right), we have found:

\[
a_{exc}(LRA) = -0.0271 \pm 0.0055 \text{ MeV}^{-1}
\]

\[
a_{exc}(t) = 0.0058 \pm 0.0026 \text{ MeV}^{-1}. \tag{7.3}
\]

These results confirm the above suggested different emission mechanism that is involved in the ternary alpha and triton emission process.

Let us now have a look at the \(^{6}\text{He}\) particles. Figure 7.9 (left) shows the absolute emission probability for the \(^{6}\text{He}\) particles plotted as a function of \(Z^2/A\). Again an indication of an increase of \(^{6}\text{He}/\text{B}\) with increasing fissility is demonstrated.

To examine the influence of the excitation energy on the \(^{6}\text{He}\) emission probability, three isotope couples are available, namely \(^{243}\text{Cm}(n,f) - ^{244}\text{Cm(SF)}, ^{249}\text{Cf(n,f)} - ^{250}\text{Cf(SF)}\) and
7.2. EMISSION PROBABILITIES

![Graph](image)

Figure 7.9: The absolute emission probability for ternary $^6$He particles (left) and $(^6$He/B)/$S_{He}$ (right) as a function of $Z^2/A$ of the compound nucleus.

$^{251}$Cf(n,f) - $^{252}$Cf(SF). In all cases a higher value for spontaneous fission than for neutron induced fission can be observed.

In analogy with the ternary $\alpha$ emission, a cluster preformation probability $S$ can be introduced, taking into account the following relation proposed by Blendowske [72]:

$$S = S^{(A-1)/3}_\alpha$$

with $A$ the mass of the emitted cluster, so in the case of $^6$He particles we have:

$$S_{He} = S^{5/3}_\alpha.$$  

(7.5)

In table 7.7 the relevant values for $S_{He}$ calculated in this way are given. A new plot (figure 7.9, right) is made to illustrate the effect of that factor $S_{He}$. It can be seen that the data vary now in a more smooth way as a function of $Z^2/A$. Both observations permit to conclude that the $^6$He particles behave more like $\alpha$-particles than like tritons.

Now the correlation between the ternary particle emission probability and the Coulomb parameter $Z^2/A^{1/3}$ is investigated. The ternary alpha and triton emission probability are plotted as a function of this parameter in figure 7.10. In the right part of the figure, it can be seen that a very good correlation for t/B is obtained with the Coulomb parameter. However, the LRA emission probability as a function of $Z^2/A^{1/3}$ shows important fluctuations. Again, the importance of the factor $S_\alpha$ becomes clear: by taking into account this factor, a much better correlation between (LRA/B)/$S_\alpha$ and the Coulomb parameter appears, as illustrated in figure 7.11.

Let us continue this discussion with the interesting approach of Cǎrjan. He interpreted the ternary fission process as an $\alpha$ (or $p, d, t,...$) decay of the fissioning system during the last phase of the scission process. In this context, a correlation between the radioactive $\alpha$-decay constant $\lambda$ and the ternary alpha emission probability is likely to exist. Therefore we try to correlate the LRA emission probability in spontaneous and neutron induced fission
Figure 7.10: The absolute emission probabilities for ternary $\alpha$-particles (left) and tritons (right) as a function of the Coulomb parameter of the compound nucleus.

Figure 7.11: $(\text{LRA}/B)/S_\alpha$ as a function of the Coulomb parameter of the compound nucleus.

with log $\lambda$. However, experimental $\lambda$ values are only available for ground state transitions, which is appropriate for spontaneous fission but which is only an approximation in the case of neutron induced fission reactions, since these are leading to a fissioning system in an excited state.

In figure 7.12 LRA/B values are presented as a function of - log $\lambda$ of the fissioning system. The left part of the figure shows the data for spontaneous fission, while in the right part, the neutron induced fission data are plotted.

This figure allows us to draw the following conclusion. A very strong correlation is obtained between the LRA/B values for spontaneous as well as for neutron induced fission
7.2. EMISSION PROBABILITIES

Figure 7.12: The LRA absolute emission probabilities for spontaneous fission (left) and neutron induced fission (right) as a function of $-\log \lambda$.

and $-\log \lambda$.
Now also the triton emission probability $t/B$ is plotted as a function of $-\log \lambda$ (figure 7.13). Here only values for neutron induced fission are shown, since for spontaneous fission there are not enough data available. In this figure it can be seen that the triton yields are slightly correlated with $-\log \lambda$. Here one observes a difference in comparison with figure 7.10, where the Coulomb parameter and $t/B$ are nicely correlated, while $LRA/B$ and $Z^2/A^{1/3}$ are only weakly correlated. This can be easily understood since the parameter $\lambda$ contains already the factor $S_\alpha$. Hence a better correlation between $LRA/B$ and $-\log \lambda$ is obtained, while the correlation between $t/B$ and $-\log \lambda$ is less obvious.

The correlation between the ternary particle yield and $-\log \lambda$ fits together with other observations. The energy needed to liberate the ternary particle is taken from the deformation energy of the fissioning nucleus. Furthermore it has been known for a long time that $\lambda$ strongly increases with an increasing nuclear radius [73], hence increasing $\lambda$ values are to be expected with increasing deformation (and thus increasing emission probabilities).

Also the parameters $Z^2/A$ and $-\log \lambda$ are somewhat correlated. Indeed, Viola and Seaborg [74] determined a semi-empirical relation between $\lambda$ and $Z$ based on the experimental ground state $\alpha$-decay rates for even $Z$, even $N$ nuclides in the region $84 \leq Z \leq 98$:

$$
\log \lambda = \frac{(2.11Z - 48.99)}{\sqrt{E_\alpha}} - (0.39Z + 16.95)
$$

(7.6)

with $\lambda$ in $s^{-1}$ and $E_\alpha$ being in MeV. Hence $Z$ is a common parameter for $Z^2/A$ and for $-\log \lambda$.

Nikitin [75] tried also to correlate the LCP yields with the calculated half-life for $\alpha$-decay of the initial fissioning nucleus in the ground state. The half-life values were computed from the nuclear mass tables and the relation $\log T_{1/2} = \frac{4}{\sqrt{E_\alpha}} + B$, which is well-known as
the Geiger-Nuttall law. The constants $A$ and $B$ are dependent on $Z$.

We have already observed in the past that the triton emission probabilities are also correlated with the average neutron multiplicity $\langle \nu \rangle$ \cite{76}. The average neutron multiplicities used in this thesis are given in table 7.8 \cite{2, 77}.

In figure 7.14, the triton emission probability is plotted for the neutron induced and spontaneous fission data as a function of the average neutron multiplicity.

In this figure it can be seen that the correlation is only valid if spontaneous fission and neutron induced fission are considered separately. The different behaviour of SF-data and (n,f)-data suggests that the excitation energy of the compound nucleus after capture of a neutron should be taken into account. So, the correlation between $t/B$ and $\langle \nu \rangle$ can be generalised using the following relation:

$$ (t/B)_{\text{cor}} = a \langle \nu \rangle_{\text{cor}} \quad (7.7) $$

where $(t/B)_{\text{cor}}$ and $\langle \nu \rangle_{\text{cor}}$ are respectively the triton emission probability and the average neutron multiplicity extrapolated to zero excitation energy:

$$ (t/B)_{\text{cor}} = t/B - \frac{\partial (t/B)}{\partial E_{\text{exc}}} E_{\text{exc}} \quad (7.8) $$

$$ \langle \nu \rangle_{\text{cor}} = \langle \nu \rangle - \frac{\partial \langle \nu \rangle}{\partial E_{\text{exc}}} E_{\text{exc}}. \quad (7.9) $$

In order to check the validity of relation 7.7, the two derivative factors $\frac{\partial (t/B)}{\partial E_{\text{exc}}}$ and $\frac{\partial \langle \nu \rangle}{\partial E_{\text{exc}}}$ have to be known.
### 7.2. EMISSION PROBABILITIES

<table>
<thead>
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<th>$\langle \nu \rangle$</th>
<th>$\langle \nu \rangle_{\text{cor}}$</th>
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<td>$(n,f)$</td>
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<tr>
<td>$^{243}\text{Cm}$</td>
<td>$3.422 \pm 0.045$</td>
<td>$2.633 \pm 0.045$</td>
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<td>$3.07 \pm 0.15$</td>
</tr>
<tr>
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<td>$4.08 \pm 0.04$</td>
<td>$3.31 \pm 0.04$</td>
</tr>
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</tr>
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<td>$(\text{SF})$</td>
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<tr>
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</tr>
<tr>
<td>$^{250}\text{Cf}$</td>
<td>$3.51 \pm 0.04$</td>
<td>$3.51 \pm 0.04$</td>
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<td>$^{252}\text{Cf}$</td>
<td>$3.757 \pm 0.010$</td>
<td>$3.757 \pm 0.010$</td>
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<td>$2.19 \pm 0.07$</td>
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<td>$^{250}\text{Pu}$</td>
<td>$2.154 \pm 0.005$</td>
<td>$2.154 \pm 0.005$</td>
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<td>$^{252}\text{Pu}$</td>
<td>$2.149 \pm 0.008$</td>
<td>$2.149 \pm 0.008$</td>
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<td>$^{244}\text{Pu}$</td>
<td>$2.29 \pm 0.19$</td>
<td>$2.29 \pm 0.19$</td>
</tr>
</tbody>
</table>

Table 7.8: Overview of the average neutron multiplicities $\langle \nu \rangle$ and $\langle \nu \rangle_{\text{cor}}$ for neutron induced and spontaneous fission.

The factor $\frac{\partial \langle \nu \rangle}{\partial E_{\text{exc}}}$ was supposed to be constant for all nuclei. A value of $(0.116 \pm 0.001)$ n/MeV was adopted, based on a comparison of $\langle \nu \rangle$-data for spontaneous and neutron induced fission leading to the same compound nucleus. The values for $\langle \nu \rangle_{\text{cor}}$ can be found in table 7.8.

The factor $\frac{\partial (t/B)}{\partial E_{\text{exc}}}$ is unknown. Nevertheless, combining equations 7.7 and 7.8, it can be deduced:

$$\frac{\partial (t/B)}{\partial E_{\text{exc}}} = \frac{1}{E_{\text{exc}}} (t/B - a \langle \nu \rangle_{\text{cor}}).$$

(7.10)

However, in order to use the above equation, the parameter $a$ must also be determined. It is done using spontaneous fission data, since in this case no correction must be applied. Performing a linear fit to the spontaneous fission $t/B$-values as a function of $\langle \nu \rangle$ yields the
following value: \( a = (5.90 \pm 0.33) \times 10^{-5} \) tritons/n. In this way, \( \frac{\partial (t/B)}{\partial E_{exc}} \) values have been calculated for all \((n,f)\)-data as a function of \( E_{exc} \). Finally, a weighted average is calculated and the value adopted is: \( \frac{\partial (t/B)}{\partial E_{exc}} = (1.94 \pm 0.55) \times 10^{-6} \) \((t/B)\)/MeV.

Lastly, \((t/B)_{cor}\)-values were calculated for all available data. The results are plotted as a function of \( \langle \nu \rangle_{cor} \) in figure 7.15. The full line is a linear fit through the data points, yielding \( a = (5.75 \pm 0.48) \times 10^{-5} \), which is in good agreement with the value determined from spontaneous fission data. It is clear from this figure that a much better correlation is obtained after extrapolating the \((n,f)\)-data to zero excitation energy.

Let us now come to the LRA particles. Figure 7.16 (left) shows the LRA emission probability as a function of the average neutron multiplicity.

While in figure 7.14 the triton yields only smoothly vary with the neutron multiplicity, the LRA yields strongly fluctuate in figure 7.16. Taking now into account the alpha cluster preformation probability \( S_\alpha \), a new plot is made where \((LRA/B)/S_\alpha\) is plotted as a function of \( \langle \nu \rangle \) (figure 7.16, right). In this figure a slightly better correlation between the average neutron multiplicity and \((LRA/B)/S_\alpha\) is obtained, however still strong fluctuations are present.

Like for the tritons, again the average neutron multiplicity extrapolated to zero excitation energy can be used. However, for the LRA particles, it is not straightforward to apply an equation similar to 7.7. Indeed, the values \((LRA/B)_{cor}\) cannot be calculated in a simple way, since there is the influence of the factor \( S_\alpha \) to take into account. Therefore a new plot is made with \((LRA/B)/S_\alpha\) as a function of \( \langle \nu \rangle_{cor} \) (7.17). This figure shows an increasing trend of \((LRA/B)/S_\alpha\) as a function of \( \langle \nu \rangle_{cor} \). This trend is less clear than in the case of the tritons, which is probably due to the (unknown) uncertainty on \( S_\alpha \).
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Figure 7.15: \((t/B)_{cor}\) as a function of \(\langle\nu\rangle_{cor}\).

Figure 7.16: LRA/B (left) and \((LRA/B)/S_\alpha\) (right) as a function of the average neutron multiplicity.

In this section the ternary particle emission probabilities are correlated with different experimental parameters. A good correlation exists between the emission probabilities and parameters containing \(Z\), like e.g. the fissility parameter \(Z^2/A\) and the Coulomb parameter \(Z^2/A^{1/3}\), which can be understood since the ternary particles are ejected due to the influence of the Coulomb field between both heavy fission fragments and this Coulomb field is heavily correlated with \(Z\). For the ternary \(\alpha\) and \(^6\)He particles one has to take into account the cluster preformation probability factor in order to obtain good correlations. Furthermore the parameter \(\lambda\) correlates very well with the LRA emission probability, while \(t/B\) correlates better with the average neutron multiplicity.
7.2.1 Nuclear temperature [78]

In 2005, Lestone presented a method to infer a nuclear temperature associated with low-energy ternary fission, using the yield of LCP as a function of the mass of the fissioning system in neutron induced and spontaneous fission.

The thermal neutron induced or spontaneous fission of two neutron-even isotopes differing by two neutrons is very similar. Examples of such pairs of isotopes with similar fission properties are $^{234,236}$U and $^{250,252}$Cf. For such pairs the potential energy surfaces are similar, and thus the shapes, kinetic energies and temperatures at scission will be similar. If these changes are assumed negligible and if ternary fission is associated with a statistical process then the ratio of ternary fission yields for a pair of neutron even isotopes differing by two neutrons will be given by the following expression:

$$ Y(A_f + 2)/Y(A_f) = \exp(-\Delta B_E/T) $$

(7.11)

with $A_f$ the mass number of the lighter of the isotope pair, $T$ the nuclear temperature and $\Delta B_E$ the difference in the light charged particle binding energy between the isotope pair:

$$ \Delta B_E = B_E(A_f + 2) - B_E(A_f). $$

(7.12)

Lestone analysed the ternary fission particle binding energies for isotope pairs with $Z$ varying from 92 to 98, as well as the difference between these binding energies ($\Delta B_E$). Even though the individual particle binding energies depend on the mass and charge of the compound system, and strongly on the assumed elongation at scission, the change in the particle binding energies is very insensitive to these quantities. Indeed, changes in $\Delta B_E$ of less than a few hundredths of a MeV are produced by changing the $Z$ of the isotope pair from 92 to 98 and by changes in the assumed shape of the emitting system. In table 7.9

![Figure 7.17: (LRA/B)/$S_\alpha$ as a function of $<\nu>_{cor}$.

\[\text{FIGURE 7.17: (LRA/B)/}\text{S}_\alpha \text{ as a function of } <\nu>_{\text{cor}}.\]
the ternary fission particle binding energies for $^{240}$Pu and $^{242}$Pu and the difference between these binding energies are given for a series of ternary particles.

<table>
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<th>Isotope</th>
<th>$B_E$ $^{240}$Pu [MeV]</th>
<th>$B_E$ $^{242}$Pu [MeV]</th>
<th>$\Delta B_E$ [MeV]</th>
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<td>$^4$He</td>
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<td>0.329</td>
</tr>
<tr>
<td>$^6$He</td>
<td>4.930</td>
<td>4.813</td>
<td>-0.117</td>
</tr>
<tr>
<td>$^8$He</td>
<td>12.918</td>
<td>12.346</td>
<td>-0.572</td>
</tr>
<tr>
<td>$^7$Li</td>
<td>2.576</td>
<td>2.849</td>
<td>0.273</td>
</tr>
<tr>
<td>$^8$Li</td>
<td>5.371</td>
<td>5.420</td>
<td>0.049</td>
</tr>
<tr>
<td>$^9$Li</td>
<td>6.255</td>
<td>6.075</td>
<td>-0.180</td>
</tr>
<tr>
<td>$^{10}$Be</td>
<td>-3.645</td>
<td>-3.195</td>
<td>0.450</td>
</tr>
<tr>
<td>$^{10}$Be</td>
<td>-5.687</td>
<td>-5.473</td>
<td>0.214</td>
</tr>
<tr>
<td>$^{11}$Be</td>
<td>-1.301</td>
<td>-1.317</td>
<td>-0.016</td>
</tr>
<tr>
<td>$^{12}$Be</td>
<td>0.538</td>
<td>0.291</td>
<td>-0.247</td>
</tr>
</tbody>
</table>

Table 7.9: Ternary fission particle binding energies for $^{240}$Pu and $^{242}$Pu and the difference between these binding energies.

From this table one can deduce that the particle binding energy changes $\Delta B_E$ decrease with increasing mass of the LCP with fixed $Z_{LCP}$, which can be explained as follows. Heavy nuclei near the valley of stability have a ratio $A/Z \sim 2.6$. For ternary particles with $A/Z < 2.6$ (e.g. an $\alpha$ particle) the addition of two neutrons to the fissioning system makes emission more difficult and this is reflected in positive values of $\Delta B_E$ for these particles. The opposite is true for LCP with $A/Z > 2.6$ (e.g. $^8$He). Therefore if ternary fission is associated with a statistical process then the yield ratios $Y(A_f + 2)/Y(A_f)$ should be insensitive to both $A_f$ and $Z_f$ and increase with increasing mass of the LCP with fixed $Z_{LCP}$. If this is the case, the ternary fission ratios can be used to infer a nuclear temperature.

In figure 7.18 ternary fission yield ratios are shown for $^{233,235}$U($n_{th}, f$), $^{239,241}$Pu($n_{th}, f$) and $^{240,242}$Cf(SF). The X-axis was chosen in order to separate the H, He, Li and Be data. Here the increasing trend in the yield ratio with increasing mass of the LCP with fixed $Z_{LCP}$ can be observed. To obtain a value for the nuclear temperature $T$, equation 7.11 was used together with the calculated values for $\Delta B_E$. The nuclear temperature was adjusted to minimize the chi-squared fit of the model calculations to all data shown in figure 7.18. The inferred temperature for low energy ternary fission is $T = 1.24 \pm 0.10$ MeV. The solid curves in the figure show the best fit with $T = 1.24$ MeV.

With this temperature $T = 1.24$ MeV, the $\alpha$-particle yield ratio predicted by equation 7.11 has a value of 0.77, the triton particle yield ratio has a value of 1.05.

Now an overview has been made of all the available measured $\alpha$-particle yield ratios
Figure 7.18: The $^{233,235}$U($n_{th}$,$f$) (open squares), $^{239,241}$Pu($n_{th}$,$f$) (solid diamonds) and $^{239,252}$Cf(SF) (open triangles) equatorial ternary fission yield ratios as a function of $A_{LCP} + 2(Z_{LCP} - 1)$. The X-axis was chosen in order to separate the H, He, Li and Be data. The solid curves show a model calculation. [78]

presented in this thesis as a function of $A_f$ for spontaneous fission and thermal neutron induced fission reactions. These data are plotted in figure 7.19 (left). In figure 7.19 (right) all available triton yield ratios presented in this thesis are shown as a function of $A_f$.

The weighted average value of all our available LRA data is $0.830 \pm 0.015$, while the weighted average value for the tritons is $0.966 \pm 0.036$. So there seems to be in both cases a discrepancy of about 8% with the values obtained by Lestone. This can probably be explained by the fact that in this study a universal temperature is assumed for all low-energy ternary fission reactions independent of reaction type and mass or charge of both the LCP and the fissioning system. Of course there must be small temperature variations with all of these quantities. For example, the temperature $T$ should vary with the LCP emitted. This is because the rarer particles require additional energy to be emitted. This depletes the excitation energy of the parent system and lowers the effective temperature for the rarer emissions. The temperature for the most frequent LRA emission will therefore be larger than the effective temperature of the rarer particles like the tritons. However, to correct for these effects, a detailed knowledge of the emission mechanism is required.

The result for the nuclear temperature of $(1.24 \pm 0.10)$ MeV is in agreement with other temperatures inferred from low energy ternary fission, like for example described in [79]. There the method of isotope thermometry is used, which is based on the dependence of ternary fission yields of LCP isotopes within a fixed fissioning system. The temperature
Figure 7.19: Measured LRA (left) and triton (right) yield ratios versus \( A_f \) for all data presented in this thesis. The line corresponds with the weighted average.

derived in this way has a value of 1.10 ± 0.15 MeV, which is in agreement with the value determined by Lestone, equal to 1.24 ± 0.10 MeV. This is compatible with the idea that ternary fission involves a statistical process where the ejected particles are in equilibrium with a heat bath with a temperature slightly hotter than 1 MeV.
Conclusion

The present work provides for the first time coherent experimental data on the ternary $\alpha$, triton and $^6$He emission for the spontaneous fission (zero excitation energy) of $^{244}$Cm, $^{246}$Cm, $^{248}$Cm, $^{250}$Cf and $^{252}$Cf and for the neutron induced fission (excitation energy $\approx 6.5$ MeV) of $^{243}$Cm, $^{245}$Cm, $^{247}$Cm, $^{249}$Cf and $^{251}$Cf.

The emission probabilities and the characteristics of the energy distributions of the ternary particles are determined. The particle identification was done with suited $\Delta E - E$ telescope detectors, at the IRMM (Geel, Belgium) for the spontaneous fission and at the ILL (Grenoble, France) for the neutron induced measurements.

Besides the importance of this study for nuclear physics in order to learn more about the scission point configuration and the emission mechanism of the ternary particles, triton emission yields are also requested by nuclear industry for safe manipulations of radioactive waste.

With these new results for the curium and californium isotopes, a significant enlargement of the existing data base is realised. Indeed, the systematic study of ternary fission by our research group started several years ago. Results have been obtained for the spontaneous fission of plutonium isotopes ($^{238,240,242,244}$Pu) and for the neutron induced fission of $^{229}$Th, $^{231}$Pa, $^{233,235}$U, $^{237}$Np, $^{239,241}$Pu and $^{241,243}$Am. Therefore now a wide selection of isotopes is available, ranging from $Z = 90$ to $Z = 98$ and $A = 229$ to $A = 252$, measured under comparable experimental conditions, which is important to allow a reliable comparison of the results.

The data set permits to make some important observations. First of all we can conclude that the average energy for a certain ternary particle remains constant within the uncertainties. Another striking observation is that for the same compound system, the FWHM for the ternary $\alpha$ energy distribution is systematically about 0.3 MeV smaller for spontaneous than for neutron induced fission. It is the first time that this phenomenon could be demonstrated, thanks to our systematic study involving 9 spontaneously fissioning nuclides and 13 neutron induced fission reactions. Furthermore the FWHM of a certain ternary particle linearly increases with increasing $Z^2/A$, which can be understood as follows. From trajectory calculations we know that the broadening of the energy distribution of the ternary particles is due to the Coulomb field that amplifies small differences occurring in the initial LRA kinetic energy spectrum. These initial differences result from fluctuations in the scission shapes, which become more important with increasing deformation energy. Since the deformation energy increases with increasing $Z^2/A$, the observed correlation of the width of the energy distributions with $Z^2/A$ is not surprising.

Also about the emission probabilities an important conclusion can be drawn. Despite an
excitation energy of about 6.5 MeV, the ternary $\alpha$ emission probability is about 20% lower in neutron induced fission than in spontaneous fission for the same fissioning system, which was hard to understand up to now. The triton emission probability on the other hand is hardly affected by this excitation energy, as expected, since the energy needed to emit a ternary particle is mainly taken at the expense of the deformation energy. We have proposed for the first time an explanation by the strong impact on the ternary $\alpha$ emission of the $\alpha$ cluster preformation probability $S_\alpha$, which will decrease due to the increased excitation energy after capture of a neutron. For the $^6$He particles a same effect as for the LRA particles is observed. Therefore one can conclude that the emission of $^6$He particles is influenced by a similar preformation factor $S_{^6\text{He}}$. Hence the $^6$He particles behave more like $\alpha$-particles than like tritons. As expected, also an increase of the ternary particle emission probability with increasing fissility is observed, since ternary fission proceeds at the expense of the deformation energy, which is proportional with the fissility parameter $Z^2/A$. Furthermore the correlation between the ternary particle emission probabilities and some other experimental parameters has been investigated. A good correlation is obtained with the Coulomb parameter $Z^2/A^{1/3}$. The radioactive $\alpha$-decay constant $\lambda$ correlates very well with the LRA emission probability, while a less good correlation appears with the triton emission probability. This can be understood since the parameter $\lambda$ contains already the factor $S_\alpha$.

So summarizing we can say that our systematic investigation, especially of fissioning systems in the ground state (= spontaneous fission) and at an excited state (= neutron induced fission), has permitted for the first time to put into evidence the strong impact of particle preformation on the ternary particle emission probability.
Bibliography


[13] B.D. Wilkins, E.P. Steinberg and R.R. Chasman, Scission-point model of nuclear fission based on deformed-shell effects, Phys. Rev., C14/5, 1832, 1976.


Samenvatting

Wanneer men het woord fissie uitspreekt, zullen de meeste mensen spontaan denken aan een kern die uiteenvalt in twee zware brokstukken. Toch werd in 1946 ontdekt dat de twee fissiefragmenten één keer om de 300 tot 400 fissies vergezeld worden van een licht geladen deeltje. Vanaf dan werd het fenomeen, dat ternaire fissie genoemd wordt, uitvoerig bestudeerd. Ondanks het eerder beperkte voorkomen van ternaire fissie kan de grote interesse gemakkelijk verklaard worden. Enerzijds zijn gegevens in verband met ternaire fissie interessant voor de nucleaire fysica aangezien de ternaire deeltjes qua plaats en tijdstip zeer dicht bij het splitsingspunt worden uitgezonden. Bijgevolg kunnen ze informatie leveren omtrent de configuratie van het splitsingspunt. Een goede kennis van hun karakteristieken zal ook bijdragen tot een beter inzicht in het emissiemechanisme van die ternaire deeltjes. Anderzijds vraagt ook de nucleaire industrie nauwkeurige gegevens over de productie van ternaire deeltjes, in het bijzonder van $^3$H (tritium) en $^4$He deeltjes, aangezien deze aan de basis liggen van de productie van He gas en het radioactieve tritium gas in een reactor. Voor een veilige werking van de reactor en voor het hanteren en verwerken van de bestaande brandstofelementen is bijgevolg een goede kennis nodig van hun productiesnelheid.

Daarom bevat dit werk een studie van de emissiewaarschijnlijkheden en de eigenschappen van de energiedistributies van ternaire deeltjes bij verschillende excitatie-energien. In de literatuur zijn er gegevens beschikbaar voor een ganse reeks isotopen. Er is echter een gebrek aan gegevens voor de meer exotische curium en californium isotopen, aangezien het moeilijk is om dergelijke isotopen met een voldoende hoge verrijking te verkrijgen. Bovendien is een vergelijking tussen de beschikbare gegevens niet zo voor de hand liggend. Die resultaten zijn immers bekomen door verschillende onderzoeksgroepen en de metingen werden uitgevoerd met uiteenlopende detectietechnieken. Daarom wordt er in dit werk een systematische studie gemaakt van verschillende curium en californium isotopen onder dezelfde experimentele condities.

De spontane fissie metingen werden uitgevoerd in het Instituut voor Referentie Materiaal en Metingen (IRMM) in Geel (België), terwijl de door neutronen geïnduceerde metingen plaats vonden in het Instituut Laue Langevin (ILL) in Grenoble (Frankrijk). Een ganse reeks 'isotopenkoppels' werd onderzocht: de door neutronen geïnduceerde fissie van $^{243}$Cm en de spontane fissie van $^{244}$Cm, $^{245}$Cm(n,f) - $^{246}$Cm(SF), $^{247}$Cm(n,f) - $^{248}$Cm(SF), $^{249}$Cf(n,f) - $^{250}$Cf(SF) en $^{251}$Cf(n,f) - $^{252}$Cf(SF). Deze koppels laten de studie toe van de meest voorkomende ternaire deeltjes, namelijk α-deeltjes, tritonen en $^6$He deeltjes, voor de splitsende samengestelde kernen $^{244,246,248}$Cm en $^{250,252}$Cf bij een excitatie energie van
0 MeV (SF) en $\approx 6.5$ MeV ((n,f)). Met al deze nieuwe gegevens wordt het bestaande gegevensbestand gevoelig uitgebreid.

Na een korte historische introductie over de ontdekking van het fenomeen fissie worden in een eerste hoofdstuk enkele algemene aspecten van het fissieproces besproken. Er wordt vertrouwen op het vloeistofdruppel model. Rekening houdend met de vervormingsenergie van het vloeistofdruppel model kan de splijtingsparameter $Z^2/A$ ingevoerd worden. Het vloeistofdruppel model was echter niet in staat om bepaalde eigenschappen van actinides te verklaren. De oplossing werd gevonden door Strutinsky met het schillencorrectie model. Zo wordt het macroscopisch aspect van het vloeistofdruppel model en het microscopisch effect van het nucleair schillenmodel samen gebracht.

Tijdens het fissieproces speelt de fissiebarrière een belangrijke rol. Als gevolg van het schillencorrectie model bekomen we een dubbele fissiebarrière. De waarschijnlijkheid voor fissie wordt grotendeels bepaald door de kwantummechanische doordringbaarheid door de fissiebarrière.

Het eerste hoofdstuk bevat verder ook een bespreking van enkele theoretische fissiemodellen. Hierbij wordt een onderscheid gemaakt tussen de zogenaamde statische modellen enerzijds, met onder andere het model van Brosa, en de dynamische modellen anderzijds. Tenslotte wordt het hoofdstuk afgesloten met een bespreking van de massadistributies van de fissiefragmenten.

Het volgend theoretisch hoofdstuk is volledig gewijd aan het verschijnsel ternaire fissie, onderwerp van dit onderzoek. Er wordt gestart met een korte introductie, waarna het verschil tussen binaire en ternaire fissie onder de loep wordt genomen.

De voor ons belangrijkste eigenschappen bij de studie van ternaire fissie, zijn de emissiewaarschijnlijkheden van de ternaire deeltjes, evenals de kenmerken van hun energiedistributies. Zoals gezegd, zullen we ons in dit werk concentreren op deze eigenschappen.

Verder volgt in hoofdstuk 2 een woordje uitgebreid over de hoekdistributie van de ternaire deeltjes. Uiteindelijk worden een aantal theoretische modellen voor de beschrijving van ternaire fissie uit de doeken gedaan: het uitgebreide Halpern model, het model van Carjan, het dubbele nekbrek model, het Pik-Pichak model en het zogenaamde sudden approximation model.

Aangezien fissie-experimenten gebaseerd zijn op de detectie van deeltjes, wordt dit in hoofdstuk 3 besproken. Eerst wordt de interactie van straling en materie algemeen bekeken, waarna het begrip deeltjesidentificatie uitgelegd wordt. Die deeltjesidentificatie gebeurt in de uitgevoerde experimenten met de gepaste $\Delta E$-E telescoop detectoren. Afhankelijk van de omstandigheden wordt zowel een ionisatiekamer ($\Delta E$) gebruikt in combinatie met een surface barrier detector (E), ofwel een telescoop met twee surface barrier detectoren (\Delta E en E).

Verder worden algemene kenmerken van detectoren besproken, alsook meer specifieke eigenschappen van de ionisatiekamer en van halfgeleiderdetectoren, waartoe ook de sillicium surface barrier detectoren behoren.
Hoofdstuk 4 gaat dieper in op de experimentele opstelling die gebruikt wordt voor de verschillende metingen. Typische kenmerken van de meetlocaties worden besproken, evenals de algemene opzet van een meting, bestaande uit twee belangrijke delen. Enerzijds is er de bepaling van de telsnelheid van de binaire fissie met behulp van de E detector. Anderzijds is er de identificatie van de verschillende ternaire deeltjes met de ΔE-E detectoren en de bepaling van de energiedistributies en telsnelheden van deze deeltjes.

In dit hoofdstuk wordt ook een schema van de elektronische opstelling gegeven, samen met een woordje uteleg over de gebruikte modules.

Om correcte resultaten te bekomen is het belangrijk om de nodige correcties voor energieverlies (vooral in aluminium en silicium) uit te voeren. De methode voor deze correcties wordt hier beschreven. Tot slot speelt ook de energicalibratie van de detectoren een belangrijke rol. Het principe van de detectorcalibratie met behulp van nucleaire reacties en radioactieve α-veral energieën wordt uitgelegd.

In hoofdstuk 5 worden de metingen van de verschillende curium isotopen samen met hun resultaten voorgesteld. De door neutronen geïnduceerde fissie van $^{243}$Cm en de spontane fissie van $^{244}$Cm worden in detail besproken: de eigenschappen van de bron, een overzicht van de metingen, de resultaten voor de binaire en ternaire fissie. Die resultaten bestaan uit de emissiewaarschijnlijkheden van de α-deeltjes, tritonen en $^6$He deeltjes, evenals de parameters van de energiedistributies van die deeltjes. Deze laatste volgen uit een Gauss fit aan de experimentele gegevens.

Voor de andere curium isotopen die tijdens de voorbije jaren gemeten werden door onze onderzoeksgroep wordt een overzicht van de resultaten gegeven. Het hoofdstuk eindigt met een overzicht van de verschillende resultaten van curium isotopen die in de literatuur te vinden zijn, voorzien van commentaar.

Hoofdstuk 6 behandelt de metingen van de verschillende californium isotopen met hun resultaten. De structuur van dit hoofdstuk is analoog aan die van hoofdstuk 5. De door neutronen geïnduceerde fissie van $^{249,251}$Cf en de spontane fissie van $^{250,252}$Cf worden gedetailleerd voorgesteld. Om te besluiten is er opnieuw een overzicht en bespreking van de verschillende metingen van californium isotopen gevonden in de literatuur.

In het laatste hoofdstuk van dit werk, hoofdstuk 7, worden alle bekomen resultaten besproken en geïnterpreteerd. Voor een coherente interpretatie worden de bekomen resultaten gecombineerd met de ternaire fissie resultaten voor alle andere isotopen die gemeten werden binnen onze onderzoeksgroep. De totale dataset die op deze manier bekomen wordt, gaande van $Z = 90$ tot $Z = 98$ en $A = 229$ tot $A = 252$, laat een aantal belangrijke vaststellingen toe.

Ten eerste kunnen we besluiten dat de gemiddelde energie voor een bepaald ternair deeltje constant blijft binnen de onzekerheden. Ook in verband met de FWHM kunnen we een opvallende conclusie trekken. Voor dezelfde splitsende samengestelde kern is de FWHM voor de ternaire α energiedistributie namelijk systematisch 0.3 MeV kleiner voor spontane fissie dan voor door neutronen geïnduceerde fissie. Het is de eerste keer dat dit fenomeen kon aangetoond worden, dankzij onze systematische studie van 9 spontaan splitsende ker-
nen en 13 door neutronen geïnduceerde reacties. Verder wordt ook waargenomen dat de FWHM van een bepaald ternair deeltje lineair stijgt met stijgende $Z^2/A$. Maar niet enkel over de eigenschappen van de energiedistributies kunnen we iets zeggen, ook in verband met de emissiewaarschijnlijkheden kunnen we belangrijke conclusies trekken. Ondanks een excitatie-energie van ongeveer 6,5 MeV is de ternaire $\alpha$ emissiewaarschijnlijkheid ongeveer 20 % lager bij door neutronen geïnduceerde fissie dan bij spontane fissie voor eenzelfde splitsende samengestelde kern. Aan de andere kant wordt de triton emissiewaarschijnlijkheid nauwelijks beïnvloed door die excitatie-energie. Dit kan verklaard worden door de grote impact van de $\alpha$-cluster preformatiewaarschijnlijkheid bij ternaire $\alpha$ emissie, $S_\alpha$, die zal afnemen na vangst van een neutron.

Bij de $^6$He-deeltjes zien we een zelfde effect als bij de $\alpha$-deeltjes. We kunnen dus aannemen dat een soortgelijke factor $S_{\alpha,H}$ de emissie van de $^6$He deeltjes beïnvloedt. Hierdoor gedragen de $^6$He deeltjes zich meer zoals $\alpha$-deeltjes dan als tritonen.

Zoals verwacht, zien we ook een stijging van de emissiewaarschijnlijkheid van de ternaire deeltjes met stijgende splitbaarheid, aangezien ternaire fissie ten koste van de deformatie-energie gebeurt, en die is evenredig met de splitsingsparameter $Z^2/A$. Verder wordt de correlatie tussen de emissiewaarschijnlijkheden van de ternaire deeltjes en enkele andere experimentele parameters onderzocht. Een goede correlatie wordt bekomen met de Coulomb parameter $Z^2/A^{1/3}$. De radioactieve $\alpha$-ervel constante $\lambda$ correleert zeer goed met de $\alpha$ emissiewaarschijnlijkheid, terwijl een minder goede correlatie optreedt met de triton emissiewaarschijnlijkheid. Dit is te begrijpen aangezien de parameter $\lambda$ reeds de factor $S_\alpha$ bevat. Tenslotte wordt er een model besproken, voorgesteld door Lestone, om de nucleaire temperatuur die samenhangt met ternaire fissie bij lage energie af te leiden, gebruik makend van de emissiewaarschijnlijkheid van ternaire deeltjes als een functie van de massa van de splitsende samengestelde kern.