University of Surrey

Doctoral Thesis

Development of a Novel Gamma-ray Detection System for Decay Data Measurement and Beta-Delayed Spectroscopy of Primary Fission Fragments At N~82

Author: Robert Shearman

Supervisor: Prof. Patrick H. Regan

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Abstract

The work presented in this thesis details three aspects of determining absolute properties of nuclear decay data. The first facet is the design and commissioning of the NAtional Nuclear Array (NANA) for the use of primary standardisation of gamma-cascade emitting nuclides, and to be used more widely as a $\gamma$-ray coincidence spectrometer. Geant4 Monte Carlo simulations were created to characterise possible designs, which were subsequently validated by comparing the response of the constructed array with the model. The simulation was also used to provide correction factors for the absolute standardisation of $^{60}$Co. The second strand concerns the determination of absolute emission probabilities of several gamma rays of the medically relevant radioisotope $^{153}$Gd. Measurements of these values were made using highly characterised HPGe detectors at the National Physical Laboratory. The absolute $\gamma$-ray emission probability of the 97.4 keV $I^\pi = 5/2^- \rightarrow I^\pi = 5/2^+$ transition was found to be 30.15 (3) %, providing a new standard and correcting a possible discrepancy within the nuclear data of $^{153}$Gd reported by the Bureau Interntionale de Poids et Measures (BIPM) international reference system (SIR). The final branch of the thesis presents analyses of $\beta^-$ decays of neutron-rich primary fission products created at the RIKEN Research Institute, Tokyo Japan. Absolute $\beta^-$-delayed gamma-ray emission probabilities and related limits of the $\beta^-$-delayed neutron emission probabilities have been deduced for $^{120-124}$Rh decays to Palladium isotopes and astrophysically important N= 82 isotope, $^{129}$Ag to states in $^{128,129}$Cd. Preliminary beta-delayed level schemes are presented for the daughter nuclei $^{120,122,124}$Pd as well as for the N=81, neutron-hole nucleus $^{129}$Cd.
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Glossary of abbreviations

BIPM  International Bureau of Weights and Measures
CFD   Constant Fraction Discriminator
DCC   Digital Coincidence Counting Project
DDEP  Decay Data Evaluation Project
DSSSD Double Sided Silicon Strip Detectors
ENSDF Evaluated Nuclear Structure Data File
EURICA EUroball RItions Cluster Array
FEP   Full-Energy Peak
FWHM  Full Width at Half Maximum
Geant4 Geometry ANd Tracking 4
Ge(Li) Lithium-drifted Germanium Detector
LaBr3  Lanthanum tri-Bromide
NANA  NAtional Nuclear Array
NNDC  National Nuclear Decay Center
NPL   National Physical Laboratory
NPTool Nuclear Physics Tool
PDF   Probability Density Function
PRF   Prompt Response Function
RIBF  Radioactive Ion Beam Factory
RiLAC Riken Linear ACcelerator
RTD   Room Temperature Dipole
Si(Li) Lithium-drifted Silicon Detector
SIR   International reference system
SRC   Superconducting Ring Cyclotron
STQ   Superconducting quadropole triplets
**WAS3ABi**  Wide Area S3 Silicon Strip Stopper Array for Beta and ion implantation

**ZCP**  Zero Crossing Point
Chapter 1

Introduction

Since before the common era (B.C.E) it has been supposed that all matter is comprised of smaller indivisible building blocks. Greek philosophers Atomists postulated that indivisible, unchanging building blocks of matter and the rearrangements of them cause the changes and types of matter that are observed in the natural world. The atom was discovered in the 19th century and the name given to what was then considered to be the indivisible blocks of matter that Democritus and fellow atomists theorised. Since then with the construction of stronger “magnifying glasses” it has been deduced and latterly seen that the atom is not indivisible in fact, nor some of its constituents, protons and neutrons. The atom is made up of electrons, and the nucleus. The number of each type of nucleons (neutrons and protons) change the physical properties of the atom, creating distinct individual nuclei for each addition or subtraction. The atomic nucleus, and the nucleon-nucleon interactions that govern much of its physical behaviour is of great importance for understanding and contextualising the nature of the universe.

The nuclear age started on 16th July 1945 when the U.S.A tested the first plutonium bomb in the trinity project. Since then, it could be argued that the very nature of human life has been shaped by the inherent possibilities and power which nuclear technology possesses. Modern-day life relies upon; nuclear power to provide electricity to a sizeable percentage of the world’s demands, and, on nuclear medicine to image and treat tens of millions of cancerous tumours each year [1]. With technology of great power comes safeguards of great responsibility and the need for detailed and precise understanding of the underlying data. In this case nuclear decay data, but it can be considered analogous
to requiring the tensile strength and limit of elasticity of steel wires when building a suspension bridge, in the worst case scenario poor knowledge of each results in the loss of life.

1.1 Beta-delayed neutron emission of exotic fission fragments

One of the most important bulk nuclear decay parameters is the $P_n$ value of nuclides produced during fission, where $P_n$ is the probability of a neutron emission directly following $\beta$ decay. Beta-delayed neutron emission ($\beta$-n) is caused by the population of states above the neutron separation energy, $S_n$ in the daughter nuclei following $\beta^-$ decay. 

A basic understanding of this process was quickly obtained after the discovery of fission [2] and was formalised in principle in 1939 [3]. The importance of these decay data cannot be overstated. In the case of the most usual fuel for thermal nuclear power reactors, $^{235}$U, a single fission will release on average 2.47 prompt neutrons compared to 0.0065 $\beta$-delayed neutrons [4]. The relatively small percentage of neutrons that are emitted delayed play a vital role in controlling the oscillations of the effective multiplication factor, $k$. Beta-delayed neutrons slow these oscillations allowing for intervention when super criticality is possible [5]. In reactor control parameters, products of fission are generally grouped into six [6] or latterly eight groups of precursor that produce delayed neutrons. From each group homogeneous bulk parameters, $\nu$, the number of fission neutrons originating as delayed neutrons and $\lambda$, the decay constant for each neutron-emitting precursor group, are needed to better understand the point kinetics of the reactor. Better estimates of these bulk parameters are needed to understand the reactor processes for nuclear power [7, 8], whilst improved methods for predicting the $P_n$ value from other gross parameters of the decay have been developed [9], the measurement of so far unknown $P_n$ values for certain nuclides is still of great importance.

In nuclear astrophysics, the nuclear r-process is the mechanism in which 50% of the heavy elements, above Fe, are produced [10]. In a high-neutron flux environment, nuclides transmutate up the nuclear chart via successive neutron capture and $\beta^-$ decay (and to a second order, fission and other charged particle capture). The individual nuclear abundances can be calculated by equation 1.1 [11].
Due to the extremely large temperatures expected at sites where this process is proposed, there is a large chance of photo-disintegration, a combative process to neutron capture. At a nucleus in each isotopic chain, approaching the neutron drip line (where $S_n < 0$), an equilibrium is met between neutron capture and photo-disintegration. At this nucleus beta-decay occurs and the two processes of photo disintegration and neutron capture compete to find the next dynamic equilibrium. The waiting point nuclide at each Z is determined to the first order by the neutron separation energy ($S_n$) therefore deducing this value (indirectly through measuring the $P_n$ value), can refine the course of modelled r-process paths. This is the first epoch of the r-process, the second is “freeze-out” where the neutron flux greatly diminishes and nuclei $\beta^-$ decay towards stability. A schematic of a possible r-process paths up the Segré is presented in figure 1.1 taken from [12].

It is clear from this that the beta decay half-lives of these nuclei are of great importance for determining the isotopic mass abundances of the final stable nuclei, and for directing
the route of the \(r\)-process before freeze out, and thus as populating such exotic nuclei has become more possible, studies of the lifetimes of nuclei on the proposed \(r\)-process have been undertaken \[13, 14\]. The nuclear structure of the waiting-point nuclei affect the half-lives dramatically; neutron magic number nuclei have longer beta-decay halflives, due to their increased \(S_n\) values and reduced \(Q\) values. At the mass number where the \(r\)-process populates magic numbers large abundance peaks can be seen. Another factor which affects the location of the mass peaks at these waiting points is the \(P_n\) value of the waiting point nuclei and of their daughters. A large \(P_n\) value approaching unity will drive strength away from the peak itself to lower mass values during freeze out; a larger \(P_n\) will also increase the neutron flux of the overall system. In recent years, as computing power has increased and experimental facilities were still unable to populate possible magic number waiting point nuclei, shell model calculations have tried to produce the lifetimes of these waiting point nuclei, in particular around the \(N = 82\) closed shell \[15, 16\], however the predictive powers of these models have been limited by the sparse data of how the dominant nuclear shell interactions evolve in this exotic region \[17\].

### 1.2 Gamma emission probabilities

Gamma-ray emission probabilities \(P_\gamma\), like \(P_n\) values, are also important to the modern human experience. In nuclear reactors, the energy output caused by gamma ray emission of the daughters following fission is approximately between 3-5 % of the total energy output during the lifetime of the reactor \[18\]. When the reactor is in shut down, this gamma decay heat accounts for nearly 50 % of the energy output. During shutdown, the residual heat must be vented and circulated appropriately to avoid excess risk of a build up of pressure throughout the system. The catastrophic results of decay heat build up have been seen before in the Fukushima disaster \[19\]. Important calculations of such scenarios are created using evaluated data libraries where the precise measurement of the gamma-ray emission probabilities are included \[20\]. Gamma rays are equally important when considering nuclear medicine. Many gamma-emitting isotopes such as \(^{99m}\text{Tc}\) are used for the imaging of cancerous tumours within patients \[21\]. It is therefore vital to know the precise gamma-ray emission probability as to not under expose or over expose the patient to an increased gamma ray dose.
1.3 Absolute activity measurements

Absolute intensity measurements are underpinned by absolute standardisations of the source being measured, without such knowledge all of these measurements would be relative to the efficiency of the detector or some other value. More generally absolute measurements, allow for universal measurements of nuclides to be dependent on the efficiency of the detector used to measure, assuming that the detector has been calibrated to a primary standard, or a secondary, disseminated standard. Activity measurements more generally are vital for all nuclear industry and beyond - knowing the precise activity of radioactive sources allows for dose calculations to be made and risk assessments to the general populace to be estimated. This could be in the waste of fracking [22], in discharge from nuclear plants [23] to the elevated risks in living in areas of increased naturally occurring radioactive materials (NORM) such as Southern Kuwait [24].

1.4 Thesis outline

The next chapter of this thesis will accustom the reader to the theoretical background of the nuclear structure and decay physics presented. Chapter three will introduce the key experimental techniques used in the measurements of the relevant nuclear decay data in the current work. The fourth chapter presents the results of the designing, commissioning and characterisation of the NAtional Nuclear Array (NANA) as well as results from one application of NANA to determine the absolute activity measurements of $^{60}$Co with simulation corrections. The fifth chapter presents recent absolute gamma intensities of the possible radio-pharmaceutical nuclide $^{153}$Gd determined by single HPGe measurement. Chapter 6 is the final chapter of experimental work, data taken following the projectile fission of a $^{238}$U beam taken at the Radioactive Ion Beam Facility (RIBF) at Riken research institute, Japan. The chapter presents previously unreported decay data on the $\beta^-$ delayed gamma-ray emission probabilities and $P_n$ values for the neutron-rich rhodium isotopes from $A = 120 - 124$ and for the $N = 82$ system $^{129}$Ag. The final chapter will summarise these results and provide an outlook to the future.
Chapter 2

Nuclear radioactive decay theory

2.1 Understanding the nucleus

To understand trends of physical data, it is imperative to formulate models that can explain the phenomena. Following Rutherford’s bombardment of gold with $\alpha$ particles [25], and the discovery of the positively-charged, dense atomic nucleus, trying to explain the underlying mechanics and behaviour of the nucleus within the atom using such models became imperative.

2.1.1 Liquid drop model of the nucleus

The liquid drop model of the nucleus was developed by Gamow [26] and independently by Gurney [27], first using quantum mechanics to explain alpha decay within the nucleus. Subsequently Bohr and Wheeler [3] expanded on this liquid drop model to explain fission. Such models do not predict many of the known internal structure observables that have been measured today, such as spin and parities of the ground and excited states, but were used to recreate what was believed to be the spherical nature of the nucleus and the atomic mass dependence on binding energy, which is determined as the work done required to transform a nucleus into its protons and neutrons. To create the semi-empirical mass or Wesizäcker formula [28], it was supposed that the atomic nucleus was an incompressible dense drop. Treated in this way, including surface tension and the electrostatic repulsion felt in proton-proton interactions, the following formula, 2.1,
arises. The formula (when transformed to consider protons and neutrons separately rather than by atomic mass) agrees well with the trends are shown in figure 2.1.

\[ E_B = a_N A - 4a_c \frac{Z(Z - 1)}{A^{1/3}} - a_s A^{2/3} - a_r \frac{(A - 2Z)^2}{A} + E_\delta \]  

(2.1)

In this equation, the first three terms can be considered classical. There is a volume term proportional to the atomic mass, a term accounting for the Coloumbic repulsion acting on the proton nucleons, and finally a term that is exactly analogous to the surface tension of a liquid drop. The final two terms represent the symmetry term and \( \delta \) which is a correction term depending on the if each nucleon is paired. Unpaired protons and neutrons cause instability in the ground state.

![Binding energy vs atomic mass](image)

**Figure 2.1:** Plot showing binding energy per nucleon vs atomic mass number for all isotopes with experimentally observed binding energy. These data are taken from reference [29]

### 2.1.2 Shell model interpretation of the nucleus

As can be observed in figure 2.1 there are clear deviations of binding energy from the semi-empirical mass formula at specific mass numbers, which correspond to “magic numbers” of protons or neutrons in the nuclei. The evidence for such magic numbers
can also be seen in the energy of the first excited state within the nucleus, which is considerably higher than neighbouring non-magic nuclei, presented in figure 2.2.

The experimentally observed magic numbers are as follows:

\[ N||Z = 2, 8, 20, 28, 50, 82, 126 \]  

(2.2)

\[ -\frac{\hbar^2 \nabla^2 \Psi}{2M} + V(r) \Psi = E \Psi \]  

(2.3)

**Figure 2.2:** Segré chart showing the energy of the first nuclear excited state. Figure modified from reference [30]

The presence of these magic numbers allows for a comparison to be drawn with the shell model interpretation of the atomic electrons. One sharp difference is the lack of a central core providing the potential because in a nuclear shell model the nucleons themselves create the potential. It is possible to calculate the wave-function of a single nucleon assuming a mean potential caused by the others, and that the wave-function energy eigenvalues can be calculated by the Schrödinger equation, which in a time independent form can be written as follows [31]:
Chapter 2. Nuclear Theory

where

\[
\nabla^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial}{\partial r} \right) - \frac{1}{r^2} \frac{\hat{L}^2}{\hbar^2}.
\]

(2.4)

The wave function, \( \Psi \), can be split into the radial and angular parts, \( R \) and \( Y \) for all spherically symmetric systems, which a nucleus at this point can be considered:

\[
\Psi(n, l, m) = R_{nl}(r) Y_{lm}^{m}(\theta, \phi).
\]

(2.5)

By treating these independently, eventually the magic numbers representing to shell closures will be produced.

Using equations 2.5 we may use the angular momentum operator, \( \hat{L}^2 \) on \( Y \),

\[
\hat{L}^2 Y_{lm}^{m}(\theta, \phi) = \hbar^2 l(l + 1) Y_{lm}^{m}(\theta, \phi).
\]

(2.6)

If we now make the substitution that \( R(r) = U(r)/r \), the principal and angular momentum components, \( n \) and \( l \) will change the energy eigenvalues, but these energies are degenerate with respect to \( m_l \) the azimuthal projection of \( l \). A first step can be to consider the unknown potential as a simple rectangular square well potential 2.7 and solve the energy eigenvalues for this potential:

\[
U(r) = \begin{cases} 
-U_0, & r < R \\
0, & r \geq R 
\end{cases}
\]

(2.7)

This first approximation of the nuclear potential failed to replicate the magic numbers first reported by Elsasser in 1933 [32]. If we consider a flatter potential the Woods Saxon potential 2.8:

\[
V(r) = \frac{V + iW}{1 + e^{(r-R_0)/a}}
\]

(2.8)

the lighter magic numbers are recreated, but the 5\(^{th}\) magic number is expected to be 40 rather than 50.
The effect of the flatter potential is to account for the positive attractive angular momentum term, the potential itself however does not include possible coupling terms of the inherent spin of the nucleus and the angular momentum of the nucleon. A final correction is the \( l.s \) spin-orbit interaction \([33-35]\), with this the magic numbers can be accurately reproduced. The spin-orbit term can be expressed in the following way:

\[
V_{\text{ls}} = -V_{\text{ls}} \frac{\delta V(r)}{\delta r} l.s. \tag{2.9}
\]

Where \( V_{\text{ls}} \) is a strength term. Here \( l \) is the orbital angular momentum vector and \( s \) is the intrinsic spin vector. The total angular momentum, \( J = L + S \). The internal spin of the nucleons, as fermions, is equal to \( \frac{1}{2} \), this spin can be parallel to the angular momentum or be anti parallel. The total angular momentum can therefore reduce to \( j = l \pm 1/2 \).

The vector addition can be determined using simple trigonometry, and thus the expectation value of \( l.s \) can be written as;

\[
\langle l.s \rangle = \frac{\hbar^2}{2} [j(j + 1) - l(l + 1) - s(s + 1)]. \tag{2.10}
\]

In the case where \( j = l + 1/2 \);

\[
\langle l.s \rangle = \frac{l}{2} \tag{2.11}
\]

and for \( j = l - 1/2 \);

\[
\langle l.s \rangle = \frac{-(l + 1)}{2}. \tag{2.12}
\]

This energy splitting can then be given by;

\[
\langle l.s \rangle_{j=l+\frac{1}{2}} - \langle l.s \rangle_{j=l-\frac{1}{2}} = \frac{1}{2} (2l + 1) \hbar^2 \tag{2.13}
\]

Which gives the full replication of the observed spherical shell gaps.

### 2.2 Decay modes of the atomic nucleus

The majority of nuclei currently observed are unstable and decay, and are by definition, radioactive. Radioactivity, the emission of a particle in the decay of matter was
first observed by Henri Becquerel in 1896, whose name would become the S.I. unit of radioactivity [36]. Since then many different modes of decay have been discovered some of which will now be discussed here. The Segré chart shown in 2.3 highlights the most dominant decay mode of each nuclide.

![Figure 2.3: Segré chart representing the dominant decay mode of each nuclide. Figure reproduced from [30].](image)

### 2.2.1 Internal decays of a nucleus

The internal decays of a nucleus occur when the nucleons are not in the lowest possible energy state for the total system. This can occur due to the excitation of a single particle, a proton, ($\pi$), or a neutron, ($\nu$), which creates a perturbation above the ground state of the whole system. This interpretation is coherent with the independent particle shell model of the nucleus as this excitation of a single nucleon (or of multiple single particle excitations) can be thought of as a nucleon being promoted, or existing in a higher energy shell. The second way an excited nucleus can be interpreted is as being in a collective excited state. This can be caused by an outside perturbing force. Most usually, this can come from a rotational excitation and a number of models have been built upon this concept, such as the rigid rotor model [37]. When these excited states
are long lived relative to other excited states, the state is known as an isomer \cite{38, 39},
deriving from the chemical adjective describing compounds with the same atoms but
arranged differently, geometrically or otherwise. Isomerism in the field of nuclear physics
was first discussed by Otto Hahn in 1921 who was investigating the beta decay of $^{234}$Th
to states in $^{234}$Pa \cite{40}. Often, nuclei in such excited states will decay to the ground state
of the same nucleus by an internal de-excitation.

De-excitations within in the nucleus, are subject to two main competing processes,
gamma decay and internal conversion. Gamma decay occurs when the excited energy
is emitted in the form of a photon, minus the recoil energy of the nucleus. Internal con-
version is the process in which the difference in energy between the excited energy state
and populated state by the de-excitation is transferred to a bound atomic electron by
the interaction of the nuclear and electronic field. These two mechanisms are discussed
below.

### 2.2.1.1 Gamma decay

The initial energy state of the system can be denoted $E_i$ and the final energy state, $E_f$.
The energetics of gamma decay can thus be considered by the following relation \cite{31}:

$$E_i - E_f = \Delta E = E_\gamma + T_M.$$  \hspace{1cm} (2.14)

Where $T_M$ is the recoil energy of the nucleus. To conserve linear momentum it follows
that:

$$0 = P_N - P_\gamma$$  \hspace{1cm} (2.15)

$$cP_\gamma = E_\gamma$$  \hspace{1cm} (2.16)

$$\frac{E_\gamma^2}{2m_Nc^2} = E_N$$  \hspace{1cm} (2.17)

$$\Delta E = E_\gamma - \frac{E_\gamma^2}{2m_Nc^2}.$$  \hspace{1cm} (2.18)
Photons have an intrinsic spin equal to 1, $s = 1$, and the total angular momentum of the photon, is given by;

$$ L = l + s. $$

(2.19)

The total angular momentum carried away by the photon can be calculated using the vector sum of the initial and final states of the nucleus $I_i, I_f$ and the total angular momentum of the photon, $L$:

$$ I_i = I_f + L. $$

(2.20)

In this vector sum $L$ can be any non-zero integer value between the sum and difference of the initial and final state. The character of the decay ($\sigma$) can be either electric or magnetic, and the angular momentum carried away by the photon, can be used to inform the observer of the multipolarity of the decay. This influences the rate of the transition, and therefore the partial width and related mean nuclear lifetime of the excited nuclear state.

The parity of a system is an important observable and can be used to define the symmetry of the wave-function with respect two coordinate systems different by a factor of -1. The parity, $\pi$, of the radiation field of the photon is dependent on both the character of the photon and the total angular momentum, for each this dependency is;

$$ \pi(EL) = (-1)^{L+1} \pi(ML) = (-1)^L. $$

(2.21)

Two points should be noted in this: (1) parity is conserved in the gamma decay as it is a strong interaction, therefore the parity change of the excited and populated states can directly be used to infer the parity of the photon and, (2) that due to the parity conservation in gamma decay electric type radiation and magnetic type radiation of the same multipolarity are not competitive processes. A summary of the gamma-ray transition rules is given in table 2.1.

It is possible due to the vector addition given in equation 2.20 to have many different multipolarity photons that are allowed in the decay. The transition rate of the decay however is dependent on the multipolarity, and thus in reality one is dominant. The
transition rate, or the decay constant for gamma decay by a certain multipolarity is given below [31]:

\[
T(\sigma L) = \frac{2(L + 1)}{\epsilon_0 \hbar L \omega_0^2 c^{2L+1}} B(\sigma L).
\] (2.22)

Here \( B(\sigma L) \) is the reduced transition probability and is related to the overlap of the final and initial wave-function of the nuclear states populated. To be able to calculate the reduced transition rates intimate knowledge of the wave-function of the nucleus is required. It is therefore favourable to simplify by creating a single particle estimate whereby the excitation is imagined to be the result of a single proton being excited to an excited shell. With this simplification Weisskopf produced reasonable approximations for the reduced transition rates for electric transitions [41]:

\[
B(EL) = \frac{e^2}{4\pi} \left( \frac{3R_L}{L+4} \right).
\] (2.23)

and magnetic transitions:

\[
B(ML) = 10 \left( \frac{\hbar}{m_p c R} \right)^2 \frac{e^2}{4\pi} \left( \frac{3R_L}{L+4} \right).
\] (2.24)

These single-particle transition rates according to the Weisskopf estimates for multipo- larities up to 5 are shown in figure 2.2.

<table>
<thead>
<tr>
<th>Multipolarity</th>
<th>Dipole</th>
<th>Quadrupole</th>
<th>Octupole</th>
<th>Xpole</th>
</tr>
</thead>
<tbody>
<tr>
<td>Angular Momentum Type</td>
<td>L=1</td>
<td>L=2</td>
<td>L=3</td>
<td>L=X</td>
</tr>
<tr>
<td>Parity Change</td>
<td>E1</td>
<td>M1</td>
<td>E2</td>
<td>M2</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>1</td>
<td>0</td>
<td>1</td>
</tr>
</tbody>
</table>

**Table 2.1:** Selection rules for gamma-ray transitions.

<table>
<thead>
<tr>
<th>Multipolarity</th>
<th>E transition rates (s⁻¹)</th>
<th>M transition rates (s⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(1.0 \times 10^{14} A^{4/3} E^4)</td>
<td>(3.1 \times 10^{14} E^5)</td>
</tr>
<tr>
<td>2</td>
<td>(7.3 \times 10^7 A^{4/3} E^5)</td>
<td>(2.2 \times 10^7 A^{2/3} E^5)</td>
</tr>
<tr>
<td>3</td>
<td>(3.4 \times 10^1 A^2 E^7)</td>
<td>(1.0 \times 10^1 A^{4/3} E^7)</td>
</tr>
<tr>
<td>4</td>
<td>(1.1 \times 10^{-5} A^{8/3} E^9)</td>
<td>(3.3 \times 10^{-6} A^2 E^9)</td>
</tr>
<tr>
<td>5</td>
<td>(2.4 \times 10^{-12} A^{10/3} E^{11})</td>
<td>(7.4 \times 10^{-13} A^{8/3} E^{11})</td>
</tr>
</tbody>
</table>

**Table 2.2:** Weisskopf estimates for single particle transition rates for electric and magnetic transitions up to multipolarity 5. \(E\) is in units of MeV and \(A\) in atomic mass units.
2.2.1.2 Angular correlation of successive gamma-ray decays

The angular distribution of an emitted gamma ray is directly dependent upon the two magnetic sub-states that it is decaying from and to. As described in [31], this notion can be illuminated when considering the decay of a \( I_i = 1 \) state to a \( I_1 = 0 \) state. As noted previously there are \( 2L + 1 \) sub-states, \( m_i \), for a given value of \( L \). So for the example here; \( m_0 = \pm 1, 0 \) and \( m_1 = 0 \). This gives rise to three different decays to the final sub-state. The emission probability of the decay \( m_0 \rightarrow m_1 = 0 \) varies according to \( \sin^2(\theta) \), where theta is defined with respect to the \( z \)-axis defined in the projection \( I \) used to define \( m \). The two \( \delta m = 1 \) transitions’ emission intensity vary according to \( \frac{1}{2}(1 + \cos^2(\theta)) \). The angular distribution of the transition is determined by the following equation:

\[
W(\theta) = \sum_{m_i} p(m_i) W_{m_i \rightarrow m_f}(\theta). \tag{2.25}
\]

In the case described this equation would become:

\[
W(\theta) = \frac{1}{3} \left( \frac{1}{2} (1 + \cos^2(\theta)) + \frac{1}{3} (1 + \cos^2(\theta)) + \frac{1}{3} \sin^2(\theta) \right) = \text{Constant}. \tag{2.26}
\]

This constant means that there is an isotropic distribution.

Successive gamma rays create an uneven population of the sub-states in the intermediate state, if the previous example is adapted to include a third level, \( I_2 = 0 \) this can be illustrated. The \( z \)-axis is now defined as the direction of the first gamma ray, \( \gamma_1 \) and \( \theta_2 \) as the angle between it and the second gamma ray, \( \gamma_2 \). For \( \gamma_1 \) the \( m_0 \) transition component follows the intensity profile of \( \frac{1}{3} \sin^2(\theta) \) as \( z \) is defined by the direction of \( \gamma_1 \), \( \theta \) is 0 and thus there is no population of the \( m_0 \) sub-state \( (p(m_0) = 0) \) in the \( I_1 \) state. Using equation 2.26 the angular distribution of \( \gamma_2 \) using the coordinate system set by \( \gamma_1 \) is given by:

\[
W(\theta) = \frac{1}{3} \left( \frac{1}{2} (1 + \cos^2(\theta)) + \frac{1}{3} (1 + \cos^2(\theta)) + 0 \sin^2(\theta) \propto 1 + \cos^2(\theta) \right) \tag{2.27}
\]

There is a non-flat distribution and the \( \gamma_1 \) and \( \gamma_2 \) have an angular correlation. In general for any multipolarity radiation, not just dipole radiation as shown here, the distribution can be determined by the following equation first derived by Hamilton [42].
\[ W(\theta) = \sum_k a_k P_k \cos(\theta) \]  \hspace{1cm} (2.28)

Where \( a_k \) is the angular correlation coefficients dependent on the states of the transitions and \( P_k \) is the corresponding Legendre polynomial. Further discussion of this is given in chapter 4.

\textbf{2.2.1.3 Internal conversion}

There is a competing process that may occur to de-excite the nucleus, internal conversion. This is the process whereby the excited fields of the nucleus interact with an atomic electron, most often of the lowest energy, the \( K \)-shell, which is then emitted from the atom. The extra energy on top of what is required for the electron to be ejected from the atom is converted to kinetic energy, \( T_e \). Internal conversion is often followed by the atomic operation where electrons from higher energy state populates the energy state voided by the emitted electron. Due to the radial dependence on probability of which orbital electron will de-excite during this process, the subsequent electronic rearrangement will often cause an avalanche of characteristic X-rays to be emitted which can, and often do interact with less bounded higher orbital electrons, leading to the emission of these so-called \textit{Auger electrons}. The kinetic energy of the first ejected electron is given by:

\[ T_e = \Delta E - B_e. \]  \hspace{1cm} (2.29)

For the majority of elements it is usual for there to be more than one orbital shell, it is then possible for electrons to be ejected from orbits with different binding energies. This results in the emitted electron to carry more or less kinetic energy. The nearest shell, \( K \), is the most dominant IC transmission due to the increased likelihood of electron shell interaction with the nuclear field.

The internal conversion coefficient (ICC) is defined as the ratio between the internal conversion transition rate and that of the corresponding gamma decay [43]:
As mentioned previously, E0 transitions are not possible through gamma decay, due to the conservation of angular momentum though are possible through a combination of unstretched transitions according to selection rules. A $0^+ \rightarrow 0^+$ therefore can only decay by internal conversion. Using the BRICC code [43] it is possible to evaluate $\alpha$ for all nuclides.

### 2.3 Transmutational decays of the nucleus

#### 2.3.1 Beta decay

Beta decay is a process by which an unstable nucleus decays to a more energetically stable nuclei of the same mass number[44]. This type of decay is a result of the weak nuclear force, where through the exchange of a W boson, quarks change type. A consequence of this quark transmutation is the creation of a lepton/antilepton pair, either a electron-antineutrino pair or a neutrino-positron pair.

Beta decay occurs through three distinct mechanisms:

- **Negative beta decay** $n \rightarrow p + e^- + \bar{\nu}_e$
- **Positive beta decay** $p \rightarrow n + e^+ + \nu_e$
- **Electron decay** $p + e^- \rightarrow n + \nu_e$

The Q value of these reactions, i.e the binding energy released during the decay, can be simply calculated when considering the conservation of energy. For each of the processes previously defined the Q-value is given below.

- **Negative beta decay** $Q^- = [m(Z, A) - Zm_e]c^2 - [(m(Z + 1, A) - (Z + 1)m_e) + m_e]c^2$
- **Positive beta decay** $Q^+ = [m(Z, A) - Zm_e]c^2 - [(m(Z - 1, A) - (Z - 1)m_e) + m_e]c^2$
- **Electron decay** $Q^{EC} = [m(Z, A) - Zm_e]c^2 - [m(Z - 1, A) - (Z - 1)m_e]c^2$
Experimentally, due to the three body kinematics of the system opposed to the two body nature of $\gamma$ decay, it is non-trivial to measure the Q value of the decay as the kinetic energy is distributed between the two ejected leptons, with the neutrino very weakly interacting and hard to detect. It is possible to extract an endpoint energy for the detected beta spectrum, and at this point it can be supposed that the neutrino carried vanishingly small kinetic energy, and thus the Q-value can be resolved.

Historically the shape of this spectrum was a clue to the underlying mechanisms of beta decay. For such a shape to occur and for the only ejecta to be the electron itself would contradict the law of conservation of energy, the most sacred, if any law can be, in physics. This explanation was therefore not favoured. The second explanation, first proposed by Pauli in 1934 was that there was in fact a second ejecta, a neutrino.

Using Pauli’s proposal Fermi was able to develop a theory of beta decay [45, 46], at the starting point of the transition rate of the decay, using his *golden rule*,

$$\lambda = \frac{2\pi}{\hbar} |V_{fi}|^2 \rho E_f.$$  \hspace{1cm} (2.31)
Here the matrix element $V_{fi}$ is the integral of the interactions between the initial and final states of the system, at the time when Fermi developed this, $V$ was unknown. At this point in time it is possible to replace this with the wave-functions of the neutrino and the electron created in this process.

$$V_{fi} = g \int [\psi_e^* \psi_\nu \psi_f] V_x \psi_i$$  \hspace{1cm} (2.32)

In this equation $g$ determines the strength of the interaction, $\psi_e^* \psi_\nu \psi_f$ represents the final state of the system. We can consider the neutrino and electron to be free particles in this process, and therefore the wave functions can be given as;

$$\psi_e(r) = \frac{1}{\sqrt{V}} e^{ipr/h}$$  \hspace{1cm} (2.33)

$$\psi_\nu(r) = \frac{1}{\sqrt{V}} e^{iqr/h}$$  \hspace{1cm} (2.34)

Here the electron is carrying momenta, $p$ and the neutrino $q$ across the volume $V$, of radius $r$. Using usual values of $p$ ($< 10$ MeV) the value $pr$ is much smaller than 1,
therefore in the expansion of the exponential we can simply approximate to the first order as 1. At this point the only terms in the golden rule dependent on the energy of the electron and neutrino is the density of the states of the system.

The electron momenta, $p$, can be considered to be composed of the Cartesian components $(p_x, p_y, p_z)$, therefore values of momentum that are of value $p$ lie on the surface of a sphere given by $P = (p_x^2, p_y^2, p_z^2)^{1/2}$. For the possible values of momentum $p + dp$, there is a volume $4\pi p^2 dp$. The possible electronic states in such a volume is therefore given by:

$$dn_e = \frac{4\pi p^2 dp V}{h^3}. \quad (2.35)$$

The density of states for the neutrino is similar. Combining these values will give the full density of states as:

$$dn_e dn_\nu = \frac{(4\pi)^2 V^2 p^2 dp dq}{h^6}. \quad (2.36)$$

We can now compute the partial decay rate of these decays. For all possible nuclear states we can can convert $\psi_f V_x \psi_i$ to the nuclear matrix element $|M_{fi}|$. Introducing this conversion creates the necessity for the corrective Fermi function, $F(Z, p)$ which includes the effect of the coloumbic nuclear field on the electron. With this correction the total beta decay rate can be given as:

$$\lambda = \frac{g^2 |M_{fi}|^2}{2\pi^3 \hbar^7 c^4} \int_0^{p_{\text{max}}} F(Z', p) p^2 (Q - T_e)^2 dp \quad (2.37)$$

Here the factor $p^2 (Q - T_e)^2$, where $T_e$ is the kinetic energy of the beta particle, arises from the density of states of the system, and is a statistical factor. To understand the nature of beta decays the logarithm to base ten of the Fermi integral $f(Z', p)$, and the partial halflife $t_{1/2} = \ln(2)/\lambda$ is often computed,

$$\text{Log}_{10}(ft_{1/2}) = \ln(2) \frac{2\pi^3 \hbar^7}{g^2 m_e^5 c^4 |M_{fi}|^2}. \quad (2.38)$$

The calculated values of this can imply the forbiddenness of the decay.
2.3.1.1 Beta decay transition type

The total angular momentum of the system must be conserved, so where $J_X$ and $J_X'$ represent the total angular momentum of the parent nuclei and the daughter nuclei respectively, the following formula must be observed;

$$J_X = J_{X'} + L + S \quad (2.39)$$

Here $L$ is the total orbital angular momentum, and $S$ is the total spin angular momentum.

Beta decays can be described as either a Gamow-Teller transition or a Fermi transition. These categories are defined by a different vector sum of the inherent spin of the two created ejecta, the electron and neutrino. As the two leptons both observe Fermi statistics both have a spin of a half, $s = \frac{1}{2}$, in this case there are two possibilities of the sum. In the parallel system, the the vector sum is $1\ h$ and the neutrino electron spins align, and the transition is a Gamow-Teller transition. In the other cases, spins anti align and the vector sum is 0. This is a Fermi transition; pure Fermi transitions can be seen in $0^+ \rightarrow 0^+$ where there is no change in $J$ and parity is conserved. Parity can be related to the angular momentum by the simple relationship $\Delta \pi = (-1)^{L}$.

The categorisation of beta decays according to the change in angular momentum and parity into forbiddenness is given in table 2.3. The order of forbiddenness can be considered as the decreased likelihood of such a decay occurring compared to the allowed or super-allowed situation. As is expected, the log(ft) of the transition increases as the order of forbiddenness (and $L$) increases.

<table>
<thead>
<tr>
<th>Forbiddness</th>
<th>L</th>
<th>$\Delta \pi$</th>
<th>$\Delta J$</th>
<th>$\text{Log(ft)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Super Allowed</td>
<td>0</td>
<td>0</td>
<td>0,0</td>
<td>&lt; 4</td>
</tr>
<tr>
<td>Allowed</td>
<td>0</td>
<td>+</td>
<td>0,1,2,3</td>
<td>4-6</td>
</tr>
<tr>
<td>1st</td>
<td>1</td>
<td>-</td>
<td>0,1,2,3</td>
<td>6-10</td>
</tr>
<tr>
<td>2nd</td>
<td>2</td>
<td>+</td>
<td>2,3</td>
<td>10-13</td>
</tr>
<tr>
<td>3rd</td>
<td>3</td>
<td>-</td>
<td>2,3,4</td>
<td>&gt; 13</td>
</tr>
</tbody>
</table>

Table 2.3: Experimentally seen log(ft) for each forbiddeness, taken from [47]
Figure 2.6: A schematic of the main processes that may occur during beta-delayed neutron emission. Figure reproduced from [48].

The beta strength function of a beta decay $S_{\beta}$ to an excited state ($E_x$) can be directly related to the log($ft$) of the decay in the following way:

$$S_{\beta}(E_x) = \frac{I_{\beta}(E_x)}{f(Q - E_x, Z).t_{1/2}}.$$  \hspace{1cm} (2.40)

Where $I_{\beta}(E_x)$ is the intensity of the beta decay. The beta strength of the decay is a very important value which can be used to deduce the branching ratios to possible decay modes, such as beta-delayed particle emission. One type of beta delayed particle emission, $\beta$-n, will now be discussed.

In nuclei where the $Q_{\beta-}$ is large, greater than the neutron separation energy of the daughter nuclei, beta-delayed neutron emission is possible. At such high excitation energy, the density of states is large, and favourable coupling between the decaying state and one of the states above the neutron separation is likely. The decay can be represented as such:

$$X(A, Z) \rightarrow \beta + Y(A, Z + 1) \rightarrow Z(A - 1, Z + 1) + n$$

A schematic of this process is provided in figure 2.6.
If the assertion that the emission of a neutron is far favoured above the neutron separation energy in comparison to gamma decay the $P_n$ value, the probability of a neutron emission can be given by the comparison of of the integral of beta intensity to states above the neutron separation, $S_n$ and the beta intensity, $I_{\beta}$ of all possible states [49],

$$P_n = \frac{\sum_{Q_{\beta}} S_{\beta}(E_i)f(Z,Q_{\beta}-E_i)}{\sum_{Q_{\beta}} S_{\beta}(E_i)f(Z,Q_{\beta}-E_i)}.$$ (2.41)

The process of the neutron decay is believed to be a more competitive process than gamma decay in this regime but recent studies have shown them to be more equal than first believed [50, 51]. Beta-delayed neutron emitters have a relatively weak binding energy of the neutron nearest the Fermi surface, as can be seen in figure 2.7, where the $z$ axis is $Q_{\beta} - S_n$. All positive values mean that beta-delayed emission of a neutron is energetically possible.
2.3.2 Fission

The final transmutational decay of the nucleus discussed here is nuclear fission. The fission of a nucleus can be reasonably argued as the most important physical process first understood and harnessed in the 20th century.

Fission, as suggested by its borrowed name is analogous to cell fission in biology where a larger cell will split into two and was coined in the nuclear physics sense by Meitner [52]. Some heavy nuclei ($A > 200$, generally) will violently split into two smaller nuclei, although higher order fission events such as ternary fission are possible [53]. Nuclear fission also releases evaporated neutrons, which create the indispensable “spark” in maintaining a chain reaction in nuclear reactors. The discovery of fission, for which Hahn and Strausmann were awarded the Nobel prize in chemistry in 1944 [54], was understood following a string of assumptions of nuclear physics.

Fission may occur if there is an energetic release following the splitting of the atomic nucleus into smaller parts. The rest mass energy of the nucleus can be defined as the number of protons in the nucleus times by the mass of the protons, the number of neutrons times by the rest mass of the neutrons, minus the binding energy ($B.E.$) of the specific nucleus. This binding energy is then the difference between the mass of the nucleus and the mass of its constituent products, protons ($m_\pi$) and neutrons ($m_\nu$).

$$M(A, Z) = Z m_\pi + (A - Z)m_\nu - B.E./c^2 \quad (2.42)$$

The binding energy of the initial nuclei and the constituents following fission are exactly related to the fission Q value.

$$Q = B.E.(A, Z) - B.E.(A_1, Z - 1) - B.E.(A_2, Z_2) \quad (2.43)$$

Materials that have a non-negligible fission cross sections at some energy are defined as fissionable, fissile nuclei are a special case of fissionable nuclei, which includes spontaneously fissionable nuclei such as $^{252,254}$Cf but also nuclei which require a small increase in energy of the system, to overcome the potential barrier and to begin self-sustaining fission chain reaction.
Fission can be understood by considering the fissionable nucleus as a liquid drop. This treatment was first devised by Bohr and Wheeler [3]. In this interpretation of fission the nuclei first splits into two drops. For fission to be energetically feasible in classical terms, the Coulomb energy between these two drops at the smallest approach (when they are touching) must be smaller than the mass excess of the nucleus. Whilst classically if this condition is not met fission is forbidden, however, if the two values are in the same regime, quantum mechanical tunnelling may take place, like with alpha emissions from the nucleus.

2.4 Radioactive decay rates and the Bateman equations

The decay of radioactive nuclei follow an exponential distribution. The constant of this decay, \( \lambda \), is simply related the lifetime of the state. For a known population of the state, i.e a known number of nuclei, \( N_0 \), the number of remaining nuclei after a time \( t \), is given by the equation [55]:

\[
N = N_0 e^{-\lambda t}.
\] (2.44)

It follows that the rate of change of \( N \), the activity of the nuclei, is this differential:

\[
- \frac{dN(t)}{dt} = -\lambda N(t) = A(t).
\] (2.45)

There are nuclear systems in which the daughter is also radioactive. In this case it is often advantageous to know the number of daughter nuclei in the system at a time, \( t \). This number is dependent on the decay constant of the parent as well as the decay constant of the daughter itself. When a granddaughter and further generations of nuclei follow, calculating the number of nuclei in each at a time \( t \) is not as straightforward. A general solution for calculating the number of nuclei \( N \) of the \( n^{th} \) nuclide populated in a decay chain was found by Bateman in 1910 [56]. This equation can be seen in equation 2.46,

\[
N_n(t) = \sum_{i=1}^{n} [N_0 \times (\prod_{j=i}^{n-1} \lambda_j) \times \left( \sum_{j=i}^{n} \left( \prod_{p=i,p\neq j}^{n} \frac{e^{-\lambda_j t}}{(\lambda_p - \lambda_j)} \right) \right)]
\] (2.46)
This equation can be corrected for branching ratios of nuclei, and for isomeric states. A shortcoming of this equation is when the decay constants of two nuclei in the chain are equal (or if using a computer to find the answer, equal when considering the precision of the compiler). Modern solutions have been found that use matrix multiplication to bypass this previous problem. For the daughter, where \( n = 2 \) the solution of the equation reduces simply to:

\[
N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1}N_1(0)(e^{-\lambda_2 t} - e^{-\lambda_1 t}).
\]  

(2.47)
Chapter 3

Nuclear decay spectrometry

This chapter will introduce the experimental apparatus used to acquire measurements used to obtain precise nuclear decay data, as well as the underlying theory of the methods which underpin these.

3.1 Detector operation

In this thesis, nuclear decay data has been measured primarily using the technique of gamma-ray spectroscopy. The two types of detectors used, Hyper-Pure Germanium detectors (HPGe) and Cerium-doped Lanthanum Tri-Bromide (LaBr$_3$:Ce) are of two different classes of detector; HPGe is a solid state detector whereas LaBr$_3$:Ce is a scintillator. The way in which these two detector types collect gamma-ray information are distinct, and are discussed in the following subsections.

3.1.1 Gamma-ray interactions with matter

Gamma rays interact with matter through a number of competing processes. The three notable and most important to consider in the typical nuclear decay energy range, are; Compton scattering, photo-electric absorption and pair production.

For the gamma-ray detectors used in this work, LaBr$_3$:Ce and HPGe, the cross section of most of the possible interaction processes can be seen in the two plots 3.1 and 3.2 generated using data from XCOM [57].
As can be seen in the two plots the photoelectric effect dominates in the low-nuclear \( \gamma \)-ray energy regime (\( E_\gamma < 100 \text{ keV} \)). Photoelectric absorption is the process in which the photon is completely absorbed by an atomic electron in the detector. This atomic electron is then ejected from one of the atomic shells populated, often the innermost K shell. The kinetic energy of the ejected electron is given by the following relationship,

\[
E_e = h\nu - E_b.
\]
Where $h\nu$ is the energy of the incident photon and $E_b$ is the binding energy of the atomic electron.

The likelihood of this interaction occurring, is greatly dependent on the atomic mass of the absorber, and the energy of the photon. The dependence on the energy and the proton number of the absorber on the probability of this type of interaction, $\tau$ is given by the approximate, empirical relationship \cite{55, 58},

$$\tau \simeq C \times \frac{Z^n}{E^{3.5}}, \quad (3.2)$$
where \( n \) varies between 4 to 5 depending upon the energy of photon.

The second competing process is Compton scattering which dominates in the energy range of many nuclear transitions (~200 keV - 4 MeV). In Compton scattering there is a linear momentum transfer between an electron and the photon, which changes both the direction and kinetic energy of the photon. In this interaction the initial photon is not destroyed, and there is not a full-energy deposition. The energy of the scattered photon, \( E'_{\gamma} \) is given by the following equation 3.3 first derived by Arthur Compton in 1923 [59].

\[
E'_{\gamma} = \frac{E_\gamma}{1 + m_e c^2 (1 - \cos(\theta))}
\]  

(3.3)

**Figure 3.3:** Scattered photon energy vs incident angle, for various different energies in the nuclear regime

Where \( \theta \) is the angle between the incident photon and the scattered photon. The probability of this scattering angle, or rather the differential scattering cross section is given by the Klein-Nishina formula 3.4 [60];

\[
\frac{d\sigma}{d\Omega} = Zr^2 \left( \frac{1}{1 + \alpha(1 - \cos(\theta))} \right)^2 \left( \frac{1 + \cos^2(\theta)}{2} \right) \left( 1 + \frac{\alpha^2(1 - \cos(\theta))^2}{(1 + \cos^2(\theta))(1 + \alpha(1 - \cos(\theta)))} \right).
\]  

(3.4)
Where $\alpha$ is the fine-structure constant ($\approx \frac{1}{137.04}$), $r$ is the atomic radius, $c$ the speed of light and $m_e$ is the rest mass of the electron.

The final process is pair production. Pair production is where in the presence of a nuclear atomic field an energetic photon ($\geq 1.022$ MeV) creates spontaneously a particle-antiparticle pair. The threshold is a universal threshold set by the rest mass of the lightest lepton, the electron. To preserve energy and momentum conservation there is a small recoil experienced by the nucleus. In regards to detector physics, this process gives rise to two 511 keV photons created inside the detector, following the positron annihilation in the material. If one or both of these annihilation photons escape the sensitive detector volume the detected energy will be $E_{\text{det}} = \gamma I - n\gamma_{\text{Ann}}$ where $n$ is the number of undetected annihilation photons. In cases where $n$ is non-zero, the final spectrum will show single-escape peaks and double-escape peaks.

### 3.1.2 Semiconductor detector theory

In the field of nuclear physics semiconductor detector materials are ubiquitous [61]. For both charged particles and gamma rays, this class of detector exhibits premier energy resolution. All crystal materials have a forbidden band gap between the valence band (in which electrons are associated to the nucleus) and the conduction band, where the electrons are disassociated and can travel across the bulk material. In the case of semiconductors, this band gap is intermediate between the large gap of insulators and the very small, almost non-existent gap in conductors. Gamma-ray interactions can excite electrons from the valence band into the conduction band, and the subsequent secondary electron collisions create more excited “free” electron-hole pairs. Electrons populating the conduction band create electron deficient atoms in the crystal lattice, known as holes. The intrinsic carrier density, $n_i$, of semiconductors is defined as the density of electrons in the conduction band and holes in the valence band caused by thermal excitation only. The equation to work out this intrinsic carrier density is given in equation 3.5.

$$n_i = AT^\frac{3}{2}e^{-\frac{E_g}{2kT}}$$  \hspace{1cm} (3.5)
Where \( A \) is a constant depending on the density of states approaching the band gap, \( T \) is the temperature, \( k \) is the Boltzmann constant and \( E_g \) is the characteristic band gap of the semiconductor. In the case of germanium detectors (\( E_g = 0.665 \text{ eV} \)), the intrinsic carrier density is too large at room temperature and signal noise would be unmanageable, due to high leakage current. It is therefore necessary to reduce the temperature of such materials, most usually by liquid nitrogen cooling, to 77 K.

The gamma-ray interaction produces a charge, detecting such charges most efficiently as possible requires charge mobility to be maximised. To do this it is necessary to dope detectors. Doping the detector, is the process in which an element with more or less valence electrons is diffused across the material. Doping the detector material with an element from a higher group will cause more electrons to be present in the material lattice, and a change in the electronic structure which creates a new electron level in the band gap very close to the conduction band. If this results in the total electrons in the conduction band to be greater, the material is then said to be an n-type semiconductor. For a lower elemental group dopant, the result is a reduced number of electrons in the conduction band in comparison to the undoped system, and it is now a p-type semiconductor. By diffusing two sides of a detector to become n-type and p-type it is possible to create a pn junction. In a pn junction the electrons of the n type material and holes of the p-type will diffuse across the junction (due to the concentration gradient) and thus electrons and holes will combine. The recombination of electron holes creates a electric field, which stops the diffusion, resulting in static space charge, where the p-type material becomes negative and n-type material becomes positive. In this region an electric field is produced that has the effect of removing mobile charge, increasing electron hole mobility in the area. This can be improved upon by increasing the depletion zone, this can be done by applying a negative voltage on the p side of the material, this process is known as reverse biasing.

### 3.1.3 Inorganic scintillator detector theory

\( \text{LaBr}_3 \) is a scintillator material. Such detector types are commonly used for photon detection due to their cost and radiation hardness, they also do not need to be cooled, which makes them very useful for use in waste assay. \( \text{LaBr}_3 \) detectors have a particularly fast timing response. Scintillators are fluorescent - that is that they are luminescent to
radiation and then re-emit radiation within a nominal time that is material dependent. For an incident photon of energy in the nuclear regime, the photon excites an electron into the conduction band, this excited electron then interacts with secondary and tertiary electrons. These excitations are usually many orders smaller than the energy of the photon. The electrons promoted to the conduction band de-excite and produce a characteristic photon emission in the visible part of the spectrum, which the scintillator is transparent to. This emission can then be multiplied, most usually by a Photo-Multiplier Tube. For the highest possible efficiency of the material-multiplier system, the wavelength of the light from the scintillator must be similar to the peak wavelength of the PMT. For NANA detectors, these two values are 420 nm for the LaBr$_3$ and 380 nm for the PMT. The more matched these wavelengths the greater the transmission efficiency, which should be maximised for an optimum detector.

The light output of the detectors, N, of the characteristic light can be approximated to the function shown in equation 3.6 containing two exponential decays. The two decay constants are properties of the material

$$N = A \exp^{-t/\tau_f} + B \exp^{-t/\tau_s}$$  

A comparison of some ubiquitous scintillator detectors’ characteristics is shown in the following table 3.1. Cerium-doped Lanthanum tri-Bromide offers greater resolution due to the increased photons per MeV of incident radiation in comparison to more conventional scintillators. The material also prospers due to the faster decay constant, which suggests that the detector has more temporal precision and thus more resolving power with respect to the lifetime and thus width of nuclear states.
Table 3.1: A table of the characteristics of several detector materials. Data included in this table is taken from [62–66].

<table>
<thead>
<tr>
<th>Material</th>
<th>HPGe</th>
<th>LaBr₃</th>
<th>CeBr₃</th>
<th>NaI(Tl)</th>
<th>BGO</th>
<th>BaF₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Light yield (photons per keV)</td>
<td>-</td>
<td>63</td>
<td>44</td>
<td>38</td>
<td>8 - 10</td>
<td>1.8</td>
</tr>
<tr>
<td>Resolution at 662 keV</td>
<td>0.2</td>
<td>2 - 3</td>
<td>5.2</td>
<td>7</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Decay time (ns)</td>
<td>-</td>
<td>16</td>
<td>17.4</td>
<td>250</td>
<td>300</td>
<td>0.6 - 0.8</td>
</tr>
<tr>
<td>Wavelength of max emission</td>
<td>-</td>
<td>380</td>
<td>390</td>
<td>415</td>
<td>480</td>
<td>220</td>
</tr>
<tr>
<td>Density (g cm⁻³)</td>
<td>5.3</td>
<td>5.08</td>
<td>5.2</td>
<td>3.67</td>
<td>7.13</td>
<td>4.88</td>
</tr>
</tbody>
</table>

3.2 Radioactive Ion Beam Factory

The Radioactive Ion Beam Factory (RIBF) is a facility used to create some of the most intense radioactive ion beams in the world [67]. In the current work the facility was used to accelerate a natural Uranium beam to an energy of 345 MeV/A. To do this the facility uses a 5 stage acceleration process. First the ions are created via ionisation of the preliminary beam in a microwave guide. These ions are then accelerated by a variable-frequency heavy-ion accelerator, the RIKEN Linear Accelerator (RILAC), into four coupled cyclotrons. The K540-MeV RIKEN Ring Cyclotron (RRC) accelerates the Uranium beam to 11 MeV/u, followed by the K = 570 MeV fixed-frequency Ring Cyclotron (fRC) where the beam is accelerated to 50.7 MeV/u. This is followed by the two final stages in the acceleration; the 980 MeV Intermediate-stage Ring Cyclotron (IRC) ($E_B = 115$ MeV/u) and finally, the 2500 MeV Superconducting Ring Cyclotron (SRC) where the beam reaches a peak energy of 345 MeV/u [68, 69]. A view of the experimental hall at RIKEN is given in figure 3.4 [taken from [68]].

From the primary beam, in this case uranium, a secondary beam is produced by the projectile fission mechanism on a thick 555 mg/cm² ⁹Be production target. The secondary beam from the in-flight fission needs to be separated such that the secondary beam is isotopically cleaned. However the beam energy used means that the spread of products in linear momentum and angular space can be significant. This requires the use of the Big RIKEN Projectile Fragment Separator (BigRIPS [70]), and a secondary spectrometer (the ZeroDegree spectrometer [70]) to provide the selectivity of isotopically pure final products to the experimental decay station for study.
3.2.1 BigRIPS and ZeroDegree spectrometers

BigRIPS is the principal mass spectrometer at RIBF and can be coupled to the forward angle ZeroDegree spectrometer. It is capable of providing good separation between radioactive specia made in the in-flight fission\(^1\) The BigRIPS spectrometer is made up of two parts that provide this selectability according to the magnetic rigidity \((B\rho)\), energy loss \((\Delta E)\), and time of flight \((TOF)\) of the species at certain stages of the spectrometer. The first part of the spectrometer is from the \(^9\)Be production target to focus F2, shown in figure 3.5. This achromatic spectrometer consisting of four superconducting Quadrupole Triplets (STQs) and four Room Temperature Dipoles (RTDs) is used to separate the ions according to radii of curvature directly related to the magnetic rigidity. The difference in this radii arises from the mass-to-charge ratio \(\left(\frac{A}{Q}\right)\) of the transmitted ions. The second part of the spectrometer is from the focus F3 to the F7. In this part of the spectrometer there are eight STQs and four RTDs creating a four-bend achromatic spectrometer. Following the second part of the BigRIPS is the ZeroDegree spectrometer located up to F11 where the decay station is located in this specific experimental campaign. ZDS is composed of a further four STQs and two RTDs [68].

The first stage of BigRIPS uses two aluminium degraders inserted in F1 and F4 to separate beams according to \(B\rho, \Delta E\) and \(TOF\) across the spectrometer. With such a discriminating spectrometer, and relatively low-energy beam, it is desirable to have a

\(^1\)An example of the particle identification associated with the BigRIPS separator for primary fragments following the projectile fission of \(^{238}\)U is shown in chapter 6 of this thesis.
Figure 3.5: Schematic of the BigRIPS and ZeroDegree spectrometers with labelled focii points.

Wide acceptance (6%) for the initial momenta, and wide angular acceptance (100 mrad vertically and 80 mrad horizontally) [70]. The second part of BigRIPS and the ZDS is used to identify the fission fragments produced. This is achieved using in-beam detectors on an event-by-event basis.

A schematic of this part of RIBF is shown in figure 3.5 from [70]. The decay station used in the experiment described in chapter 6 of this thesis was located at F11.

3.2.1.1 Detectors in-line at BigRIPS and ZDS

To select a particular ion-species, in-beam detectors along focii along the two spectrometers are required. (These detectors need to be able to create the Particle IDentification spectrum shown in Chapter 6 of this thesis). Such detectors must either explicitly or implicitly measure the mass-to-charge ratio of the ions ($\frac{a}{q}$), and the atomic number ($Z$), which as the secondary beam is completely ionized, is the same as the charge of the ion ($Q$).
Separation of individual isotope species is achieved by using the mass-to-charge ratio and the atomic charge of the ions. The mass-to-charge ratio of the isotopes in the spectrometer can be calculated by considering the centripetal force experienced by charged particles in the presence of an external magnetic field ($B$):

$$F = \frac{m v^2}{\rho} = q(E + \nu \times B)$$  \hspace{1cm} (3.7)

### 3.2.2 Decay station detectors

In the experiment covered in this work, a charged multi-element particle detector system, the Wide-range Active Silicon Strip Stopper Array for Beta and ion detection (WAS3ABi) [71] was used to detect the final detected ions. WAS3ABi is also sensitive to electrons and was used to provide a $\beta$ trigger to correlate associated gamma rays succeeding the decays of the ions. These gamma rays were detected using the EUroball RIKen Cluster Array (EURICA) which surrounded WAS3ABi. These two detector systems will now be discussed.

#### 3.2.2.1 WAS3ABi

Ions are implanted at the decay station on the Wide-range Active Silicon Strip Stopper Array for Beta and ion detection (WAS3ABi). This stopper was composed of a stack of up to eight double sided silicon strip detectors. Each detector had 40 strips in the y direction and 60 in the x axis [71]. The sensitive pixels created by this segmentation were of 1 mm$^2$ area. This high segmentation and fine granularity allowed for clean correlations to be made between the ion implantation and the subsequent beta decays. To maximise the efficiency of the whole system for low energy gamma rays and X-rays non-sensitive holding structures of WAS3ABi were minimised in the design [72]. In figure 3.6 a photo of WAS3ABi is shown in the centre of the EURICA spectrometer, which is discussed in section 3.2.3.

#### 3.2.3 EUroball RIKen Cluster Array

To provide high precision of the poorly-known decay gamma rays emitted during the decay of the secondary beam ions, the EUroball RIKen Cluster Array [73] was created.
Chapter 3. Nuclear decay spectrometry

Figure 3.6: A photo of WAS3ABi located in the centre of the EURICA spectrometer. In this photo three of the 7-element clusters are also clearly visible.

and placed at the decay station at RIBF. This array comprises 12 of the HPGe 7 element cluster detectors from the EUROBALL gamma-ray array. The detectors were placed around WAS3ABi to provide maximum angular coverage. In such cluster detectors, there is a large probability of Compton-scattered photons depositing energy in more than one detector allowing the possible “add-back” of energies from neighbouring HPGe crystals to reconstruct the energy of the full-deposited gamma ray.

The absolute $\gamma$-ray detection efficiency of EURICA must be well-known for the $P_\gamma$ value of the characteristic gamma rays to be determined. These values can be used to determine the feeding to daughter states in nuclei, and fundamentally the Log(ft) of the beta decays. An experimentally derived function of efficiency has been measured by G. Lorusso [74] using a $^{152}$Eu standard this function is shown in figure 3.7.
3.3 Monte Carlo simulation radiation transport codes

The Monte Carlo method is a statistical sampling technique. The name was first coined by von Neumann and Ulam [75], after the casino, who believed it to be of use when modelling the transport of neutrons. The method is used to compute an otherwise complex problem, in which systems with many degrees of freedom are modelled by a probabilistic method. Generally this is breaking down the problem into a set of probability density functions (PDF) and randomly sampling from each [76]. Often it is necessary to sample from many different PDFs in a single simulation, it is therefore necessary to create many random numbers per simulation. The Monte Carlo method applied to radiation transport consists of a primary particle, with defined energy, velocity and position. PDF’s of possible interactions or processes are known according to evaluated data or theoretical calculations, from this a step is computed where the particle is moved forward in time according to the processes probabilistically selected. This is then repeated over many iterations, for a good representation of the system. The Monte-Carlo simulation of radiation transport is commonplace, and several codes have been developed to accurately recreate nuclear reactions and processes [77–80]. In the work included in this thesis, Geant4 (GEometry ANd Tracking 4) MC code was used, and will be discussed now.
3.3.1 Geant4

Geant4 is an open source, object-orientated toolkit for the Monte Carlo simulation of particle transport, from which a user can assemble their own simulation program. In order to assemble and compile a code using this framework, a user must generate; a detector construction, a primary generator and a physics list. The detector construction defines the world of the simulation, the geometry, bulk properties and elemental composition of the detector, surrounding materials and the world volume. The primary generator is the definition of the initial particle ($e^{-}/+, \gamma, \alpha$ or $X_A^Z$), the excitation energy, position, and velocity. The final part of the model is the initialisation of the processes that exist with in the model, from which, probabilistic outcomes are determined. This physics list, can include; Electromagnetic processes, optical physics, particle decay, stopping physics etc... In the ideal case all processes will be defined and “turned on” in the physics list of a Geant4 program, practically however, each process increases the computational time and a trade off where unimportant physics lists must be discounted is found. A front-end of Geant4, NPTool, is used in the work presented in this thesis and is discussed in more detail in chapter 4.
Chapter 4

The NAtional Nuclear Array

The NAtional Nuclear Array (NANA) is a new coincident gamma-ray array, constructed at the National Physical Laboratory as part of this PhD project. The array consists of up to 12 Cerium-doped Lanthanum tri-Bromide (LaBr$_3$:Ce) inorganic scintillator crystals each coupled to a Hamamatsu (R9779) photo-multiplier tube (PMT).

The detectors from this array have been used for an array of physics and metrology cases, ranging from high-temporal fidelity timing measurements, to absolute standardisation of gamma-cascade emitting nuclides and characterisation of radioactive nuclear fuel waste. This chapter describes the design, simulation and characterisation of the NANA, and demonstrates its application as a coincidence spectrometer for the absolute standardisation of the radioactive $^{60}$Co nucleus.

4.1 Introduction to the Monte Carlo model of NANA

Geant4 (GEometry ANd Tracking 4) is a C++ suite developed originally in CERN that using known cross sections and decay parameters of all known nuclides can accurately model nuclear physics phenomena [77]. It was developed to follow on from the FORTRAN based GEANT3, with enhanced power through the use of the object orientated language C++.

NPTool is a framework used to link the simulations that often preceed nuclear physics experiments [81]. Simulations are necessary before an experimental run to determine
the expected efficiency of the detector set-up. NPTool can also facilitate the analysis
of the dataset created during the experimental run. Creating such an interface between
the two means that, should the user want, the same analysis routine can be used for
both simulated and real data, increasing productivity after an experimental campaign.

The hierarchical folder system of the NPTool software is presented below in figure 4.1.
The NPL folder of NPTool defines the data structure of the detectors made in the
software, as well as including functions to deal with the Physics of the simulations, real
data (Physics) which may be needed in future analysis and core utilities of NPTool,
such as ROOT (I/O) and the detector manager. NPS/Detectors holds the detector
construction and the structures in the virtual experimental hall of all available detectors
with in the architecture. NPS/Core holds the Geant4 derived functions and classes to
deal with nuclear physics processes on the defined geometry. The Projects folder is
where the analyses files of the real or simulated data are held. Finally the inputs folder
hold the placements of physical volumes with respect to the beam or source, as well as
the event generator files which contain the information of the primary particle generator.

The NANA detectors built in NPTool were initially clones of the FATIMA detectors
[82]. These detectors whilst of the correct housing shape did not consist of several key
components, including a more thorough realisation of the PMT.

4.1.1 Geometry of the Monte Carlo model

The detector in the simulation consists of an aluminium can, the crystal and a borosil-
icate glass bulk representing the PMT, the lead shield, at the front of each detector is
also contained within the model and can be simply moved on and off. The three most
important parts of the model, are; a) the 3.81 cm diameter crystal, b) the 22 cm long
PMT and c) the irregularly shaped Pb Compton shield. The thin aluminium can and
front window, and PMT-to-crystal coupling are also modelled.

4.1.2 Validating NPtool model with a single detector

The NPTool model predates the detector array’s construction. For the validation of
the model, a $^{152}$Eu source of approximately 128 kBq was placed in front of a single
detector for 10 minutes, this corresponds to around $7.68 \times 10^6$ decays. This acquisition was replicated using the NPTool model, with radioactive decay and an extended electromagnetic physics list (option 4) “on” so that the necessary physics is considered for the replication of the lead fluorescence caused by the lead shielding surrounding the
front face of each detector. The data used to prescribe the decays are from the Geant4 10.02 radioactive decay datasets. The Geant4 model uses the CLHEP::RandGauss class to create a Gaussian distribution of each incident energy where sigma is similar to that of the full-energy peak line shapes. This was set according to previous resolution measurements. The lack of the array caused issues with geometry construction, creating a necessity to include a wooden table top and several lead bricks, which were used in the acquisition to provide stability to the set-up. A comparison of the two acquisitions can be seen in figure 4.2, reproduced from [81]. The full-energy peaks (FEPs) can be seen to be reproduced with good quality, however, under 300 keV there are increased Compton interactions despite the well-reproduced geometry. Although the Compton continuum is ill-produced in the low-energy regime the most important part of the simulation, the recreation of the FEP’s, are reproduced to a good degree.

![Figure 4.2: Validation of the NPTool model of a NANA LaBr3 scintillator. $7.68 \times 10^6$ decays were simulated of $^{152}$Eu.](image)

A model of the array frame prior to the delivery of the STFC constructed frame was built according to the CAD model in NPTool, as shown in figure 4.3. This can be considered the final state of the NPTool model. Detectors in the model and in the array are assigned detector numbers according to the position in the array. The detector number positions can be seen in table 4.1.
Figure 4.3: The array as constructed in NPTool.

<table>
<thead>
<tr>
<th>Detector number</th>
<th>Polar angle $\theta$ (degrees)</th>
<th>Azimuthal angle $\phi$ (degrees)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>90</td>
<td>112.5</td>
</tr>
<tr>
<td>1</td>
<td>90</td>
<td>157.5</td>
</tr>
<tr>
<td>2</td>
<td>90</td>
<td>202.5</td>
</tr>
<tr>
<td>3</td>
<td>90</td>
<td>247.5</td>
</tr>
<tr>
<td>4</td>
<td>90</td>
<td>292.5</td>
</tr>
<tr>
<td>5</td>
<td>90</td>
<td>337.5</td>
</tr>
<tr>
<td>6</td>
<td>90</td>
<td>22.5</td>
</tr>
<tr>
<td>7</td>
<td>90</td>
<td>67.5</td>
</tr>
<tr>
<td>8</td>
<td>45</td>
<td>45</td>
</tr>
<tr>
<td>9</td>
<td>45</td>
<td>135</td>
</tr>
<tr>
<td>10</td>
<td>45</td>
<td>225</td>
</tr>
<tr>
<td>11</td>
<td>45</td>
<td>315</td>
</tr>
</tbody>
</table>

Table 4.1: The angles of the detectors comprising NANA. Across all the presented work, the detector number is consistent to the angles described here.

4.1.2.1 Characterisation of NANA

The digitiser used to digitise the signal output from the NANA array is a V171C CAEN module. This module can be run using several different firmwares with several different output streams. A suite of C++ codes were written in order to handle these data streams and convert them to time correlated event-based ROOT [83] files. The flow of data is as follows; 1) the output data is converted to absolute time, and temporally sorted, 2) the stream is converted to a root tree according to the TNanaData class descriptor, defined
in NPLib, 3) A coincidence window is set, and the single signal events is transformed to a multi-signal event Tree file, 4) analysis of the final root tree is conducted using NPAnalysis according to the Analysis.cxx file. Step 1 is not necessary for all file types. The TNanaData data class includes many leaves that are not output in all data streams, but allows for it to be used in all streams. The Tree expected in this class is given in figure 4.4

![Tree structure diagram](image)

**Figure 4.4:** The tree structure of a tree according to the TNanaData class.

Lanthanum tri-bromide is well-known to have internal radiation [84, 85] which is typically of the order of 1 Bq cm$^{-1}$ [86]. This activity largely arises from primordial $^{138}$La component of the lanthanum detector material which is long lived ($T_{1/2} = 1.3 \times 10^{10}$ years, and has an isotopic abundance of 0.090(1), and the chemical analog $^{227}$Ac ($T_{1/2} = 21.7$ years), which makes up part of the $^{235}$U 4n+3 radioactive decay chain. Lanthanum-$^{138}$ decays by beta minus emission ($I = 35\%$) where it is followed by a 788.7 keV gamma-ray emission in $^{138}$Ba, $^{138}$La will also decay by electron capture, where it will decay to the first excited $2^+$ state in the in the daughter $^{138}$Ba, which deexcites via a 1435.8 keV gamma ray. Actinium decays by beta decay to $^{227}$Th which is part of a five decay alpha chain [84]. Due to the difference in interaction between $\alpha$ particles and $\gamma$ rays, these alpha decays have a different voltage signal profile compared to electrons and gamma rays in the scintillator, and appear quenched in an energy spectrum. For characterisations, NANA comprised 8 detectors arranged in the ring at a nominal 78 mm away from the origin of the array. This set up can be seen in figure 4.5.

A sum spectrum, a spectrum across all the modular detectors present, with no external
source present is shown with the prominent internal radiation labelled in figure 4.6. This acquisition was recorded by the digitiser using the CAEN Digital Pulse Processing (DPP) firmware.

4.1.3 The $\gamma$-ray energy resolution and absolute full-energy peak efficiency of the NAtional Nuclear Array

In order to characterise the detector array, an NPL mix source was prepared on a thin film source. The radionuclides present in the source are presented in table 4.2. This specific mix includes radionuclides which decay via gamma-ray emissions of a wide range. As the activity of the nuclides are known, the efficiency, linearity and resolution of the detector array in this configuration can be found across a wide energy range.

The mixed source was placed in the centre of the NANA array, using a 3D printed source holder designed specifically for this purpose. An acquisition of the source was taken for 86,400 s with the digitiser running the DPP firmware. A single detector spectrum of
Figure 4.6: An 84000 s acquisition showing the prominent internal radiation of one of 8 detectors arranged in the ring of the NANA spectrometer. Note that the energy of the 5 alpha emissions is quenched in relation to the gamma-ray emissions.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Gamma-ray energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{231}$Am</td>
<td>59.5</td>
</tr>
<tr>
<td>$^{57}$Co</td>
<td>122.1</td>
</tr>
<tr>
<td>$^{137}$Ce</td>
<td>165.9</td>
</tr>
<tr>
<td>$^{113}$Sn</td>
<td>391.7</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>661.7</td>
</tr>
<tr>
<td>$^{34}$Mn</td>
<td>834.8</td>
</tr>
<tr>
<td>$^{60}$Zn</td>
<td>1115.5</td>
</tr>
</tbody>
</table>

Table 4.2: The gamma rays used in the efficiency and energy calibration of the NANA device contained in the NPL standard source.

The mixed source can be seen in Figure 4.7. From this acquisition, FEPs of the gamma-ray emissions from the mixed sources were fitted using a Gaussian function on top of a polynomial background to recreate the complex continuum beneath the peaks. From these fits, channel centroid, peak area and the full-width at half-maximum (FWHM) can be determined. Due to contamination from quenched alpha radiation the highest energy gamma ray $E_\gamma = 1836$ keV for the decay of $^{88}$Y was not included in calculations. Further background complexities resulted in the 1173 keV and 1332 keV gamma rays to be discounted also.
Figure 4.7: Sum spectrum of all eight individual detector spectra, from an 86,400 s acquisition of the mixed source used in calibration.

The FWHM is determined by the Gaussian fit of the full energy peaks. The $\sigma$ returned is related to the FWHM in the following way,

$$FWHM = 2\sqrt{2\ln 2} \sigma.$$  \hspace{1cm} (4.1)

The efficiency of the device was determined using the approximation of the Gaussian integral, with returned fit parameters, the constant, $H$, and width of the Gaussian, $\sigma$;

$$Area = \frac{H \times \sigma}{0.3989}.$$  \hspace{1cm} (4.2)
A simulation of the detector was performed in parallel. A range of energies encompassing the experimental FEPs were shot at the detector from an ideal source positioned in the middle of the array. The fitting procedure was the same as the method in the real data. Due to the lack of any background radiation in the MC simulation, the peaks were clean, except at low energy where the Compton edge and lead fluorescence must be considered.

The efficiency of the detector array is comparable to the simulated efficiency of the detector array. Slight discrepancies are seen in the lowest energy peak \( E_{\gamma} = 59.5 \text{ keV} \), this can be explained by two competing factors; 1) The source created in the simulation does not include the thin film, and can be considered a completely open source, the lack of attenuation would be most apparent in this regime, 2) the resolution is poorer at low energy for this class of detector, and as such, the background beneath the peak may have been poorly fitted.

**Figure 4.8:** FWHM of the NPL source gamma-ray peaks vs the energy of the peak, for a single NANA detector.
4.2 Selection of weakly populated states in a mixed caesium source

One of the most useful aspects of a modular gamma-ray detector array is the ability to select and enhance the peak to total of weakly populated decay paths present following the decay of a radioactive source. The peak-to-total (P/T) of a specific peak is often used with many definitions. In this work it is defined in the following way;

\[
P/T = \frac{\text{Counts}_{FEP}}{\text{Counts}_{\text{spectrum}}} = \frac{\text{Area}_{FEP}}{\text{Entries}_{\text{spectrum}}}\]

(4.3)

Considering possible future applications of the array, a mixed source of \(^{134,137}\text{Cs}\) was created of equal activity (4 kBq) of each isotope. Caesium is an important identifier in the nuclear industry and the different concentrations of these two particular isotopes can inform the viewer on the cause of the radiation present, due to the large change in yield for each according to production technique. Caesium-137 is a longer-lived radionuclide (\(T_{1/2} = 30.05(8)\) years [87]) emitting one characteristic 661.6 keV gamma ray from a metastable state populated in the \(^{137}\text{Ba}\) daughter. Caesium-134 is a shorter-lived isotope
(T_{1/2} = 2.0652(4) y [88]) with a higher multiplicity of gamma-rays emitted per decay. Of particular note is the large number of high-energy (above 500 keV) gamma rays in coincidence in its decay. The decay schemes of $^{134}$Cs can be seen in figure 4.10.

![Figure 4.10: Level scheme of $^{134}$Ba following the decay of $^{134}$Cs taken from [85].](image)

The prepared sample was placed in the centre of NANA at $r = 78$ mm away from the detector faces. Eight detectors in a ring, as shown in figure 4.5 were used to acquire spectra for 14,400 s using the CAEN V1751C digitiser running the DPP-PSD firmware. Background run files of 86,400 s were also acquired. A sum spectrum, where all the energy depositions across all eight detectors is plotted individually is shown in figure 4.11.

The first improvement to the spectrum that can be made is to remove contamination from the room and the detectors themselves, the internal radiation can be seen in figure 4.6. The improvement in background subtraction is clearly visible in figure 4.11, with the 1038 keV, 1168 keV and 1365 keV gamma-ray transitions are clearly observed above the background.

Conditions on the data can be imposed to improve P/T for particular full-energy peaks.
Chapter 4. NANA

Figure 4.11: A 14,400 s acquired spectrum of the mixed caesium source, with (red) background subtraction, and without (blue). The weakly populated states are clearly visible in the subtracted spectrum only.

A multiplicity requirement or “cut”, where only events with a certain number of detectors triggering in coincidence, will increase the P/T of FEPs which is populated in a decay path of multiplicity equal to the cut. The range of such cuts with entries larger than 200 are included in figure 4.12 without background subtraction, and with in figure 4.13.

The 662 keV emission is not visible in any multiplicity plot except multiplicity 1, and has improved P/T in the 1-multiplicity cut compared to the total-hit spectrum this is shown in table 4.3.

Another way of presenting coincident gamma-rays is to present the data in a 2D histogram. In this type of spectrum, all events with more than one signal detected in the event is plotted. Coincident events fill this spectra according to the following coordinate:

\[ H(x, y) = (\gamma_1, \gamma_2) \] (4.4)

For coincidence 2D histogram, coincident pairs are added twice for symmetry about the \( x = y \) line as is this case for the matrix shown in figure 4.14 for the mixed caesium histogram in figure 4.14. Higher multiplicity events than 2 are unfolded into couples,
i.e. a 3 hit event is unfolded in to 3 pairs of gamma rays and plotted. In this spectrum, peaks are formed at the coordinates of the two coincident gamma rays. The valley seen in the region of 650 keV to 700 keV clearly shows that the $^{137}\text{Cs}$ 662 keV gamma ray is not coincident with any other photon. Conversely this is not true of Compton events.
caused by the 662 keV interacting with two detectors, the effect of this is a diagonal ridge in the spectrum where $x + y = 662$

**Figure 4.14**: The observed 2D coincidence matrix observed of the mixed 8 kBq caesium source acquiring across the 8 element NANA detector for 14,400 s.

Further P/T improvements can be made by applying energy gates. By gating on different coincident decays it is possible to increase the P/T of the coincident full energy peaks. In the case of $^{134}$Cs, the most well-fed 3 multiplicity cascade is the $4^+ \rightarrow 4^+ \rightarrow 2^+ \rightarrow 0^+$, the 569 keV, 796 keV, 605 keV transition. By gating on the 796 keV or 605 keV transitions, the 569 keV P/T is enhanced as well as the other coincident transitions. This procedure can be seen in figure 4.15. The final way that this particular spectrum can be improved is by triple gating on these two peaks. Creating such gates leaves the weakly populated 569 keV and its associated Compton events only. The effects of this whole procedure can be seen in plot seen in the last spectrum seen in figure 4.15. A quantitative table of peak-to-total before and after each gate can be seen in table 4.3.
Figure 4.15: Top: The coincidence spectrum of the 8 detectors when gated on 605 keV in the decay of $^{134}\text{Cs}$. Middle: The coincidence spectrum of the 8 detectors when gated on 796 keV. Bottom: A double-energy gated (605 keV and 796 keV) coincidence spectrum.
### Table 4.3: Peak to total values across each cut, showing the improvement in spectral quality at each condition. Peak to total is given in percent. *In the 605 keV gated spectrum, some of the 605 keV full energy peak itself is present this is due to the proximity in energy of the coincident 569 keV FEP.*

<table>
<thead>
<tr>
<th>Peak (keV)</th>
<th>569</th>
<th>605</th>
<th>662</th>
<th>796</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sum spectra</td>
<td>2.024 (10)</td>
<td>7.138 (13)</td>
<td>4.302 (7)</td>
<td>5.39 (26)</td>
</tr>
<tr>
<td>Background Sub</td>
<td>3.990 (16)</td>
<td>11.835 (19)</td>
<td>7.326 (13)</td>
<td>8.553 (13)</td>
</tr>
<tr>
<td>Multiplicity 1</td>
<td>1.767 (9)</td>
<td>6.892 (13)</td>
<td>4.575 (8)</td>
<td>5.180 (18)</td>
</tr>
<tr>
<td>Multiplicity 2</td>
<td>5.12 (15)</td>
<td>11.4 (3)</td>
<td>0</td>
<td>9.16 (24)</td>
</tr>
<tr>
<td>Multiplicity 3</td>
<td>7.9 (8)</td>
<td>8.3 (9)</td>
<td>0</td>
<td>5.3 (6)</td>
</tr>
<tr>
<td>605 gated</td>
<td>14.0 (9)</td>
<td>*</td>
<td>0</td>
<td>7.6 (5)</td>
</tr>
<tr>
<td>796 gated</td>
<td>12.8 (16)</td>
<td>11.6 (17)</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>796 and 605 gated</td>
<td>37.6 (32)</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
4.3 Excited nuclear lifetime measurements using the National Nuclear Array

The lifetime of an excited nuclear state is an important parameter for deducing the nature and structure of the level itself. To evaluate such important values of the state such as the g-factor and the quadrupole moment measurements the lifetime of the state is essential [89, 90].

Historically, lifetimes of the order of tens of picoseconds and single nanoseconds have been hard to measure experimentally. Longer lifetimes were easily measured using pairs of HPGe detectors in delayed coincidence [39]. Shorter lifetimes of the order of units of picoseconds to femtoseconds have used Doppler Shift Attenuation Method (DSAM) [91] or by the plunger method. With the development of higher resolution fast timing scintillator detectors, such as LaBr₃, lifetime measurements of nuclear states in this range are more convenient [92].

The measurement of mean nuclear lifetimes, $(\tau)$, by $\gamma$-ray coincidences is not a new technique, and has been practised for over 50 years [93, 94]. In the simplest of set-ups, two detectors are used; one to observe the gamma ray populating the state of interest, $\gamma_1$ and the second to observe the de-populating gamma-ray, $\gamma_2$. In analogue electronics a train of electronics succeeding the amplifier of each detector proceeds as follows; a timing SCA from each connects to a Time-to-Amplitude Converter, where the detector detecting $\gamma_1$ is the start signal and the second detector, the stop signal, this feeds through to a MCA from which a time distribution is made. Such an analogue train of electronics is shown schematically in figure 4.16.

The produced time distribution is a convolution of the prompt response function (PRF), $P(t)$ of the two detectors (which is a normal distribution) and the lifetime of the state, plus any background contribution which can be expressed in the manner seen in equation 4.5 [94].

$$D(t) = \int_{0}^{\infty} P(t - t')e^{-\lambda t'} dt'$$  \hspace{1cm} (4.5)

Where $\lambda$ is the decay probability which is the reciprocal of the mean lifetime of the of that transition.
The convolution and thus the line-shape of the time distribution can be analytically solved.

The PRF of the two detectors has the form:

$$P(t) = \frac{1}{\sigma\sqrt{2\pi}} e^{\frac{-t^2}{2\sigma^2}}$$  \hspace{1cm} (4.6)

and the exponential is:

$$F(t) = e^{-\lambda t}$$  \hspace{1cm} (4.7)

The PDF of this convolution is:

$$D(t) = F(t) \ast P(t)$$  \hspace{1cm} (4.8)

$$D(t) = \frac{1}{2} e^{-\lambda((t-\mu)^2 - \frac{\sigma^2}{2})} [1 + erf\left(\frac{(t - \mu - \sigma^2 \lambda)}{\sqrt{2}\sigma}\right)]$$  \hspace{1cm} (4.9)

Where $erf$ is the error function. An extraction of a lifetime can be made by fitting this to the time difference spectrum produced, assuming that the lifetime is at least twice as long as width of the PRF [94]. In figure 4.17, three convolutions of a Gaussian distribution and exponential are shown for lifetimes of different fractions of sigma.
Figure 4.17: Comparison of the convolution of three different lifetime decays in the same detector, i.e. the same PRF. Top) The lifetime of the state is equal to 5 times sigma of the PRF, Centre) the lifetime of the state is equal to sigma, and, Bottom) the lifetime of the state is a fifth of sigma.
In the following section, examples of lifetimes characterised in this way by NANA for which \( \tau \) is suitably long will be discussed.

### 4.3.1 Determination of lifetime measurements using digital 1 ns time-stamped data

The digital electronics of NANA allow for post processing of data so that there is no requirement to apply energy conditions on the signals at the time of acquisition, or for coincidence data to be recorded. Another benefit of post processing is the fact that the two detectors can gate on both energies throughout acquisition, this allows for two time distributions to be formed from each detector pair, one where detector i is the stop detector and detector j is the start detector and vice versa. The clock of the V1751C digitiser of NANA has a 1 GHz sampling rate, which means data has a time-stamped precision of 1 ns. Similar detectors have been showed to have a PRF of the order of hundred of picoseconds. Knowing these two facts, lifetimes of the order of nanoseconds can be easily extracted from the delayed time distribution from the fitting of the convolution only, and extracting \( \lambda \). Test case of known lifetimes in this order are presented in this section to show the current working state of the detector array for measuring nuclear lifetimes.

### 4.3.2 Test of NANA fast timing using the lifetime of the \( I^x = 2^+ \) state in \(^{152}\text{Sm}\)

The decay of \(^{152}\text{Eu}\) provides a useful temporal calibration source in nuclear physics, allowing for temporal, energetic and for efficiency measurements. The \( Q \) value of the decay, allows for states of excitation energy of more than 1500 keV to be populated in the daughter nuclei. Knowing the activity of the source, an absolute efficiency curve of the detector system can be determined. For timing calibrations, the first \( 2^+ \) state in the EC decay branch daughter, \(^{152}\text{Sm}\), has an evaluated value for its mean lifetime of 2.024(15) ns [95]. This makes it ideal for confirming the characteristics of analogue and digital trains following detectors of this resolution such as bin width for an analogue SCA for example, an example of this can be seen in [96].

The level schemes for the decay of \(^{152}\text{Eu}\) are shown in figure 4.18.
In order to test the timing condition of the NANA device acquisitions of a 14 kBq $^{152}$Eu source were acquired on 2 detectors only using the DPP firmware. A $\gamma - \gamma$ coincidence energy matrix from a 14,400 s acquisition of the source is shown in figure 4.19.

As can be the decay schema seen in figure 4.18 there are many possible populating decays into the $I^\pi = 2^+$ state of $^{152}$Sm, from which the lifetime can be measured using NANA. The three most populating decays are; the 1408 keV, 964 keV and 244 keV emissions. When selecting energy gates, the cleanliness of the time difference spectrum must be considered. It is expected that lower energy peaks in relation to higher energy emissions in the acquired spectra will have a Compton continuum underneath the full energy peak, which will be in coincidence with the de-populating gamma ray. The greater the number of counts in the time difference spectrum caused by this contamination, the greater the uncertainty contribution, of the background. It is therefore useful to evaluate these populating gamma rays and the coincident energy spectra to see which provides the least contaminated gate, it is most likely that this is the most energetic, where there is the least contamination of the gate caused by the Compton continuum underneath the FEP.
From inspection of figures 4.20, 4.22 and 4.21 it is clear that the cleanest energy gate is the 1408 keV gate. By selecting these two energy gates the following time difference spectrum is produced. With a fit according to the convolution PDF given in equation 4.9.

A simple fit of the exponential slope outside of the Gaussian prompt function can be used to extract the lifetime of the first $2^+$ state in $^{152}$Sm, using the 1408 keV gate, a fit, shown in figure 4.23, was produced. From these fit parameters, the lifetime of the state was calculated to be 2.035(15) ns, which is in good agreement with the literature value. A second way of resolving the lifetime of the first $2^+$ in $^{152}$Sm is by fitting the whole spectrum using the convolution method. An average of the fits for the stop-start
and start-stop detector pairs yields the a lifetime of $2.10(5)$ ns and the sigma of the Gaussian to be of the order of 450 ps. This value does agree with the adopted value. However, uncertainties in this measurement are high, due to the bin width (the PRF is less than half a bin width), and will only increase with smaller lifetime. It is therefore
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Figure 4.22: The coincident spectrum created by gating on the 244 keV FEP, with a time coincidence set at 25 ns. Many non-coincident lines with the 122 keV are seen, this is due to the large Compton continuum beneath the energy gate.

necessary to consider timing algorithms that produce better than 1 ns timing precision. This fit of the time difference histogram is shown in figure 4.24.

Figure 4.23: 1 ns Timestamped time difference spectra gated on 1408 keV and 122 keV transitions in the decay of $^{152}$Eu to $^{152}$Sm. Lifetime extracted using a exponential plus linear background on the slope of the distribution.
Constant fraction discrimination (CFD) is a way in which time pick-off are made of an analogue signal. It is often preferential to use this timing technique when compared to leading edge timing due to the energy independence of the technique [63]. The signal is split and one path is delayed, inverted and attenuated, before the two trains are then summed together. These manipulations produce a bipolar pulse. The zero crossing point (ZCP) is then used to provide the timing measurement of the pulse. The effect of this technique is that time pick off is made after a fraction of the total pulse height has been reached. This fraction is reached at the same time regardless of pulse heights. A scheme of the implementation is provided in figure 4.25.

From waveform outputs of the digitiser, the previously mentioned ROOT event builder was altered to include a CFD algorithm, as well as a linear interpolation of the pulse. The event builder delays and attenuates the pulse before recombining at which point a first order polynomial fit using $N$ points around the zero crossing point is evaluated, and

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{figure4.24.png}
\caption{Figure 4.24: Time difference spectrum of 1408 keV - 122 keV gamma-ray cascade in the decay $^{152}$Eu acquired on NANA outputting 1 ns timestamped data. Lifetime determined from a convolution fit of a Gaussian PRF and exponential decay plus a constant background.}
\end{figure}
the $x$ intercept of this line corresponds to the ZCP. This can be seen in the case where $N = 2$ in figure 4.26.

To optimise the values of the delay and attenuation of the CFD algorithm an evaluation of the performance of each value across the same source acquisition was carried out. An acquisition of $^{60}$Co was made on which gates were set offline on the 1173 keV and 1332 keV full-energy peaks. From this the CFD parameters were altered and the FWHM of the time distribution evaluated, as well as the number of entries of the histograms, the greater number within the peak the more robust and rugged the CFD parameters.

As can be seen in figures 4.27 and 4.28, the most optimised system can be seen at delay = 6 ns and for an attenuated signal of 40 % the unmodified signal.
4.3.4 Timing measurements using CFD technique with NANA

Measurements of the first $I^\pi = 2^+$ state lifetime in $^{152}$Sm and the $I^\pi = \frac{3}{2}^+$ state lifetime in $^{133}$Cs were acquired using the CFD algorithm with a delay and attenuation of 60
% and 4 ns respectively. Source acquisitions using this technique were then made to determine whether precision and accuracy were improved in comparison to the 1 ns timestamped data. The decay of $^{133}$Ba produces a longer lived nanosecond isomeric state in $^{133}$Cs than the state in $^{152}$Sm.

In figures 4.29 and 4.30 a time difference spectrum was created using the 1408 keV-122 keV energy gates as in the 1 ns timestamped data. Fits were performed using the slope method and the convolution method. In both cases the precision and accuracy has improved in comparison to the 1 ns binned data, which is shown in table 4.4. The convolution method was used to extract the width of the prompt Gaussian response function of $\sigma = 311(4)$ ps, which is far smaller than reported using the 1 ns timestamped data, suggesting that the fit using the convolution method of that data was impaired due to the bin width.

<table>
<thead>
<tr>
<th>Time difference (ps)</th>
<th>Counts per 50 ps</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>5000</td>
<td>10</td>
</tr>
<tr>
<td>10000</td>
<td>2</td>
</tr>
<tr>
<td>15000</td>
<td>103</td>
</tr>
<tr>
<td>20000</td>
<td>106</td>
</tr>
</tbody>
</table>

**Figure 4.29:** Lifetime determination of the first $2^+$ state in $^{152}$Sm using the CFD derived timestamps, fitted using the slope method.

<table>
<thead>
<tr>
<th></th>
<th>Mean lifetime from fitting the slope (ns)</th>
<th>Value from convolution fit</th>
<th>Width of PRF (ps)</th>
<th>Evaluated value (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 ns timestamped data</td>
<td>2.1 (1)</td>
<td>2.10 (5)</td>
<td>450 (50)</td>
<td>2.024 (15)</td>
</tr>
<tr>
<td>CFID timing data</td>
<td>2.034 (15)</td>
<td>2.054 (30)</td>
<td>311 (4)</td>
<td>2.024 (15)</td>
</tr>
</tbody>
</table>

**Table 4.4:** Values of the mean lifetime of the first $2^+$ state in $^{152}$Sm, calculated by fitting time difference spectra acquired on NANA running with sub-nanosecond timestamps and 1 ns timestamps.
Figure 4.30: Lifetime determination of the first $2^+$ state in $^{152}$Sm using the CFD derived timestamps, fitted using the convolution method

<table>
<thead>
<tr>
<th>CFD timing data</th>
<th>Mean lifetime from fitting the slope (ns)</th>
<th>Value from convolution fit</th>
<th>Evaluated value (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>9.07 (7)</td>
<td>9.12 (15)</td>
<td>9.064 (21)</td>
</tr>
</tbody>
</table>

Table 4.5: Values of the mean lifetime of the $3/2^+$ state in $^{133}$Cs, calculated by fitting time difference spectra acquired on NANA running with subnanosecond timestamps.

From the known excited level scheme of the daughter nuclei, $^{133}$Cs, in the decay of $^{133}$Ba shown in figure 4.31, gates were created on the 81 kev and 356 keV gamma emissions (356 keV being the highest energy feeding transition) from which the time distribution spectrum between the two decays can be produced. A mean lifetime for this decay was calculated by fitting the slope, shown in figure 4.32 and by fitting the convolution of the PRF and exponential decay as shown in figure 4.33, these are given in table 4.5. The mean lifetime of both methods is in good agreement with the evaluated data.
Figure 4.31: The excited level scheme of $^{133}$Cs following the decay of $^{133}$Ba, data taken from [98]. In this figure the half-lives of the states are given.

Figure 4.32: The time difference spectrum of the 356 keV - 81 keV cascade following the decay of $^{133}$Ba acquired using NANA, fitted on the slope with an exponential and linear background.
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4.4 Standardisation of $^{60}$Co using NANA

A study at the NPL to assess the feasibility of using NANA as a spectrometer to provide absolute standardisations of radionuclides that produce coincident gamma rays in their decay was performed. This work could provide a quick alternative to 4$\pi$(LS) Digital Coincident Counting (DCC)\cite{99}.

Previous standardisations using the method of $\gamma - \gamma$ counting have used $^{60}$Co as the “prototype” decaying radioisotope. This is in part due to the simplicity of the decay, in which 99\% of the time there are only two stretched E2 high-energy gamma rays emitted in coincidence in the $\beta^-$ decay. The level scheme can be seen in figure 4.34.

In a study by the National Institute of Standards and Technology in the U.S.A. a solution was found to extend the previous work for the absolute standardisations of $\gamma$-X-ray emitting nuclides to determine the activity of $^{60}$Co, by using two NaI(Tl) scintillators \cite{100}. The derivation of this can be found in \cite{100} and \cite{101}, the final equations to determine the activity is shown below:

$$N_0^2 = \frac{(N_1^{(2,p)}N_2^{(2,p)} - N_{1c}^{(2,p)}N_{2c}^{(2,p)}) (N_1^{(1,p)}N_2^{(1,p)} - N_{1c}^{(1,p)}N_{2c}^{(1,p)})}{N_c^{(2,1)}(N_1^{(1,p)} - N_{1c}^{(1,p)}) (N_2^{(2,p)} - N_{2c}^{(2,p)})}, \quad (4.10)$$
Figure 4.34: The decay scheme of $^{60}$Co. The data used are taken from [87].

\[
N_0^1 = \frac{(N_1^{(2,p)}N_2^{(2,p)} - N_{1c}^{(2,p)}N_{2c}^{(2,p)})(N_1^{(1,p)}N_2^{(1,p)} - N_{1c}^{(1,p)}N_{2c}^{(1,p)})}{N_{1c}^{(1,2)}(N_2^{(2,p)} - N_{2c}^{(2,p)})(N_1^{(1,p)} - N_{1c}^{(1,p)})}, \tag{4.11}
\]

where:

- $N_0$: Disintegration rate of $^{60}$Co
- $N_i^{(j,p)}$: The count rate of detector $i$ of $\gamma_j$ full-energy peak $p$
- $\epsilon_i^{(j,p)}$: Probability of detecting the full energy peak ($p$) of $\gamma_j$ in detector $i$
- $\epsilon_i^{(j,tot)}$: Probability of detecting $\gamma(j)$ in detector $i$ by any interaction

From these two equations, the quadrature sum of the $N_0^1$ and $N_0^2$, $N_0$ can be calculated. From knowing how much of the solution was deposited in each source, the specific activity $A_0$ can be determined. This solution for $N_0$ is only valid when the angular
correlation does not need to be accounted for, which is not the case for measurements taken with NANA. In this case we must introduce the weighting function, \( W(\theta) \) as described, in section 2.2.1.2 in chapter 3. In the case of two stretched \( E2 \) transitions the angular correlation coefficients \( a_2 \) and \( a_4 \) have been deduced experimentally in 1950 [102], the weighting function for this particular transition is thus.

\[
W(\theta) = 1 + 0.1020 P_2(\theta) + 0.0091 P_4(\theta)
\] (4.12)

Where \( P_2 \) and \( P_4 \) are the Legendre polynomials are;

\[
P_n(\cos(\theta)) = \frac{1}{2^n n!} \frac{d^n}{d(\cos(\theta))^n}(\cos(\theta)^2 - 1)^n
\] (4.13)

\[
P_2(\cos(\theta)) = \frac{1}{2}(3\cos^2(\theta) - 1)
\] (4.14)

\[
P_4(\cos(\theta)) = \frac{1}{8}(35\cos^4(\theta) - 30\cos^2(\theta) + 3)
\] (4.15)

Therefore, \( N_0 \) is no longer the quadrature sum of the two gates but rather this value, \( \bar{N}_0 \) times by this weighting function, \( W(\theta) \).

\[
\bar{N}_0 = \sqrt{N_0^2 N_0^{22}}
\] (4.16)

\[
\bar{N}_0 = W(\theta) N_0
\] (4.17)

### 4.4.1 Experimental method

A \(^{60}\)Co sample was found to have the specific activity, the activity per unit mass, of 330.92(86) kBq g\(^{-1}\) by \( 4\pi (LS) \) DCC. The five sources prepared were placed in the centre of NANA, 78 mm away from the detector faces. The eight detectors available were configured in a ring as described in table 4.1. In this configuration, there were a total of 7 values of \( \theta \); 45, 90, 135, 180, 225, 270, 315 degrees, with a possible 56 detector pairs available. For each measurement, histogram spectra were acquired for each detector, and coincidence histogram spectra were created using gates on the 1173 keV and 1332 keV,
The time window for coincidence was set at 25 ns. A coincidence matrix across the 8 detectors can be seen in figure 4.36.

Two typical coincidence spectra are shown in figures 4.37 and 4.38. Peak areas were found from peak fitting using a Gaussian and a polynomial of up to third order, as an approximation to the continuum beneath the peaks. For all 5 sources the measurements, results determined by using detector 3 or 5 showed significant bias. The gain of these two detectors is far different than the 6 other detectors. Considering these two facts, results of these two detectors were not used in the calculation of any of the results here. This reduces the number of detector pairs from 56 to 30.
Figure 4.36: The total coincidence matrix across the NANA spectrometer after a 86,400 s acquisition of one of the prepared $^{60}$Co sources.
Figure 4.37: The coincidence spectra of detector 7 by gating on the 1173 keV decay in detector 4.

Figure 4.38: The coincidence spectra of detector 7 by gating on the 1332 keV decay in detector 4.
4.4.2 Initial results

The initial results of the study show good agreement with the DCC measurement of activity, as can be seen in table 4.6. The relative difference of the two techniques is 0.4%. If the activities are plotted with respect to $\theta$ normalised to the activity at $\theta = 180^\circ$, the weighting function does not follow the true dependence shown in the data as shown in figure 4.39, where there is an overestimation of the increase of activity of gamma rays emitted perpendicular to one another. Considering this, there is a requirement to consider further corrections to the angular correlation corrections.

<table>
<thead>
<tr>
<th>Standardisation technique</th>
<th>$A_0$</th>
<th>$u(A_0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NANA $\gamma$-$\gamma$</td>
<td>331.8</td>
<td>3.0</td>
</tr>
<tr>
<td>$4\pi$(LS)-$\gamma$ DCC</td>
<td>330.92</td>
<td>0.86</td>
</tr>
</tbody>
</table>

Table 4.6: Specific activity of the $^{60}$Co sample as determined by the two primary standardisation techniques.

Figure 4.39: The weighting function used to apply the correction for angular correlation compared to the actual ratio between correlations.
4.4.3 Angular correlation correction

When treating the angular correlation it was supposed that one discrete value of $\theta$ was the value between the pairs of the detectors. This assumption is true only if the detectors are collimated, which these NANA detectors are not. A correction for non-collimated detectors was considered in 1969, where the weighting function, $W(\theta)$ was defined as follows [103]:

$$W(\theta) = \sum_{\text{even}k}^{k_{\text{max}}} a_k P_k(\cos(\theta)) Q_k(1) Q_k(2)$$  \hspace{1cm} (4.18)

Where $Q_k$ is the attenuation correction factor given by:

$$Q_k(i) = \frac{j_k(i)}{j_0(i)}$$  \hspace{1cm} (4.19)

Here $j$ is attenuation per correlated gamma ray at $\beta$, the incident angle in the detector.

$$j_k(i) = \int_{0}^{\beta_{\text{max}}} d\beta \sin(\beta) P_k \cos(\beta) \epsilon_i(\beta)$$  \hspace{1cm} (4.20)

The only term of the integral that is unknown is $\epsilon_i(\beta)$. In 2002 Kim et al.[104] showed two ways that this can be solved; 1) by purely knowing the geometry of the system and the linear attenuation coefficient the efficiency function can be fitted for different regimes of the detector, and 2) in which the efficiency can be found across the detector face by Monte Carlo modelling for the specific gamma-ray energy of interest.

To find the efficiency across the face of the detector at each gamma-ray energy of the cascade, a simulation of one detector at 78 mm away from the detector was set up. A mono-energetic gamma ray corresponding to the energy of one cobalt emission was “shot” at the detectors 1 million times at 100 discrete angles between 0 and $\beta_{\text{max}}$. This procedure was repeated for the second energy. The open angle ($\beta_{\text{max}}$) of the active region of the detector at 78 mm is 13.6 degrees. From this $d\beta$ is known, and the efficiency of each angle can be determined by the area of the FEP of each simulation. As only one energy of gamma ray was incident on the detector there is no background continuum to consider and the peak can be fitted by a Gaussian only. The geometry of the source
was similar to the thin source in material definitions, although due to the unknown drop deposition profile of the source, the simulation assumed that the source was point-like.

The simulation, with several simulated gamma rays is shown in figure 4.40, and the corresponding results of the simulations for $\epsilon_{1173}(\beta)$ vs $\beta$ are shown in figure 4.41.

![Figure 4.40: Setup of the simulation to find the FEP angular efficiency of a NANA detector.](image)

The integrals for $\epsilon(\beta)$ were calculated using the rectangle rule, although crude this provided satisfactory results due to the granularity of the measurements. This led to values of $Q_2$ and $Q_4$ for the 1173 keV and 1332 keV gamma rays shown in table 4.7.

<table>
<thead>
<tr>
<th>Gamma-ray energy (keV)</th>
<th>$J_0$</th>
<th>$J_2$</th>
<th>$J_4$</th>
<th>$Q_2$</th>
<th>$Q_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173</td>
<td>0.2559</td>
<td>0.2491</td>
<td>0.2336</td>
<td>0.9735</td>
<td>0.9138</td>
</tr>
<tr>
<td>1332</td>
<td>0.2270</td>
<td>0.2210</td>
<td>0.2074</td>
<td>0.9733</td>
<td>0.9129</td>
</tr>
</tbody>
</table>

Table 4.7: The attenuation coefficients and corresponding Q values for the detector-gamma ray system.
4.5 Simulation-corrected result of the $^{60}$Co absolute standardisation using NANA

Inputting the values determined by MC simulation the new weighting function is plotted against the normalised activity measurements of the same source (S1602119) as in figure 4.39, is shown figure 4.42.

The weighting function, by observation is a better fit to the measurements. The specific activity obtained per source, the weighted average of the specific activity for the sources, and the value of the specific activity as calculated by $4\pi$\textit{(LS)}-\gamma DCC is is shown in figure 4.43, and a tabular comparison of the two primary counting techniques is shown in table 4.8.

<table>
<thead>
<tr>
<th>Standardisation technique</th>
<th>$A_0$</th>
<th>$u(A_0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NANA $\gamma$-$\gamma$</td>
<td>330.8</td>
<td>1.0</td>
</tr>
<tr>
<td>$4\pi$\textit{(LS)}-\gamma DCC</td>
<td>330.92</td>
<td>0.86</td>
</tr>
</tbody>
</table>

Table 4.8: Specific activity of the $^{60}$Co sample as determined by the two primary standardisation techniques, with the improved weighting function $W(\theta)$. 

Figure 4.41: Efficiency dependence on $\beta$ for the two gamma-ray energies used.
The angular dependence of the activity of source S2016 relative to back-to-back detectors. Dashed line is the previous weighting function $W(\theta)$, dotted line represents the modified weighting function $\overline{W}(\theta)$.

The $\gamma - \gamma$ technique using NANA is of similar precision to the DCC technique now, although still slightly poorer. A relative difference of 0.27% is observed.
Figure 4.43: Comparison of all source activities determined by the $\gamma - \gamma$ counting technique and by $4\pi$(LS)-$\gamma$ DCC.
Gadolinium-153 is a radioactive isotope that decays by electron capture (EC) with a branching ratio of 100 % to the stable $^{153}\text{Eu}$ [97]. The evaluated half life of the radionuclide is 240.1 (4) days [97]. It is most usually produced in the thermal neutron irradiation of a natural europium target in reactors, and then chemically separated to be used in many radiopharmaceutical activities.

NPL acquired chemically pure $^{153}\text{Gd}$ in 2011. From this sample, several sources have been prepared for use in calculating the specific activity of the sample (the activity per unit mass) through digital coincidence counting (DCC) [99]. Three further sources were also specifically prepared for use in gamma spectroscopy measurements to deduce the absolute $\gamma$-ray emission probabilities or $P_\gamma$ of the gamma rays following the electron capture decay of $^{153}\text{Gd}$. 
5.1 Underlying motivation for precision measurement of $^{153}$Gd decay data

There is a cornucopia of applications of this radionuclide in the medical field [105] which provide ample motivation for the determination of the absolute emission probabilities of the emissions in this decay. In this thesis, it is important to consider some of these individually, and in this context, why improvement of the existing nuclear decay data of this nuclide may be necessary in a wider context.

5.1.1 Medical applications of $^{153}$Gd sources

5.1.1.1 Line source in Single-Photon Emission Computed Tomography

Single Photon Emission Computed Tomography is a ubiquitous technique in which a person is imaged with a gamma-emitting radionuclide [106]. Gamma cameras rotate around the patient. From this, regions of high-radionuclide uptake can be seen in reconstructed 3D space. These radionuclides are most usually attached to a ligand which is used in specific cell reproduction, and regions of increased uptake will suggest the presence of a tumour where cell reproduction rate is higher [107]. A problem of this technique is the unknown attenuation of gamma rays through the body tissue of the individual being examined. One solution to this degradation of signal is to use a gamma-emitting nuclide as a line source, external to the person, in order for the transmission of the nuclide to through the body to be quantified on a patient-by-patient basis. Gadolinium-153 is a commercially-available line source [108]. Gadolinium-153 is particularly attractive due to the energy of the principal emissions (97.4 and 103.1 keV). $^{99m}$Tc is the most commonly used radionuclide injected into a patients for SPECT [107], the gamma ray of this decay is the 140 keV emission [109]. Since this is of greater energy than the emissions following $^{153}$Gd decay, there is negligible contamination in the energy window of $^{99m}$Tc caused by the line source. Knowing the absolute gamma-ray emission probabilities per decay is important so the associated gamma dose of the patient can be calculated when using such a line source.
5.1.1.2 Dual photon absorptiometry (DPA)

Bone absorptiometry via dual photon absorptiometry is an imaging technique used to acquire information relating to the mineral content and the health of bones within a patient [110]. This process is currently not widely favoured by the medics. To assess the bone density, it is required to have two strongly emitted photons of low energy, to determine the gamma-ray absorption coefficient. From this information, within the framework of a model, the bone density can be calculated, and from that, the bone mineral content. This is a good indicator of whether a patient is suffering from osteoporosis [111]. Dual photon absorption can produce the bone mineral density when considering the linear attenuation of two different materials.

5.1.1.3 Candidacy in Interstitial Rotating Shield Brachytherapy Treatment (I-RSBT)

Brachytherapy is an internal radioactive treatment to cancerous tumours applicable to many sites within the body, most notably the prostate [112, 113], the breasts [114, 115] and the cervix [116, 117]. In this treatment a sealed source is injected next to the cancerous site. The most usually practised brachytherapy is High Dose Rate (HDR) brachytherapy, where $^{192}$Ir is the radionuclide. This course of treatment is beneficial compared with Low Dose Rate (LDR) brachytherapy as the patient does not need to remain in-situ for as long and the cancerous cells have less chance to divide and grow over the course of the treatment. $^{192}$Ir is most often utilised as the recommended specific activity (16.8-18.5 TBq/g) is achievable, thereby allowing a high dose to be achieved in a small sealed vessel. Also $^{192}$Ir is used because of the high thermal neutron cross section of the parent nucleus $^{191}$Ir, which allows for high production yields. This procedure however is not perfect, and the practitioner must leave the treatment room during treatment because of the high dose that would be received whilst the source is being physically delivered into the patient.

Interstitial rotating shield brachytherapy treatment was proposed as a replacement of the HDR brachytherapy treatment [118]. It was seen to be a beneficial replacement as it would allow for the increased selectivity of the treatment and a more focused delivery of the radiation dose. In this treatment a radionuclide would be injected into the patient,
and a rotating shield of platinum or high-density metal would protect healthy tissue whilst delivering a dose to the tumour site. It would be vital for this to be a widespread therapeutic treatment for the radionuclide to be able to be produced commercially in a large activity, upwards of 10 GBq for delivery to the hospital and subsequently to the patient.

In the past few years \[105\], \(^{153}\text{Gd}\) was proposed as a nuclide that could be used that matched the criteria outlined. In 2016, a simulated delivery system of the radionuclide in reasonable concentration, showed certain positive effects when compared to the higher energy conventional treatment using \(^{192}\text{Ir}\) \[119\]. The efficacy has not be trialled outside of simulation. However the advantages shown have been such that the possible methods of creating the specific activity required to deliver the necessary dose to the patient has been reviewed.

5.1.2 SIR and SIRIC

The International Reference System for activity measurements of \(\gamma\)-ray emitting nuclides (SIR) is a way in which comparisons can be made between activity measurements between different National Measurement Institutes (NMIs) \[120, 121\]. NMIs send measured samples in a SIR glass ampoule that will be placed inside an extremely stable and well-known ionisation chamber (SIRIC). The current invoked in the SIRIC is then compared to the current obtained by a \(^{226}\text{Ra}\) reference source. The equivalent activity, the activity of the source required to produce the same output current of the reference source, can be considered to a measurement of the efficiency of the IC to the emissions of the specific radionuclide being measured. The equivalent activity \(A_e\) and the efficiency of the detector are linked in the following way; by modelling the expected response of the IC for various radionuclides of known response, Key Comparison Reference Values (KCRVs), the efficiency curve for both photon emission and beta emission curves across a wide range of energies can be computed.

The SIR efficiency curves developed using this method provide an extremely powerful tool; not least for the calculation of measured equivalent activities in cases where there is no KCRV of a particular species. Furthermore using the efficiency curve to model an equivalent activity measurement is also possible. When discrepancies arise between the measured and the modelled data, there can be three potential problems: a) The
SIR result is incorrect; b) the efficiency curves are incorrect; c) that the nuclear data used to model the activity contributions are incorrect. In the case of $^{153}\text{Gd}$ the difference between the measured equivalent activity and modelled activity is large, 4%, this comparison of activities for many nuclides is shown in figure 5.1 reproduced from [120]. The modelled activity of $^{153}\text{Sm}$ which has similar gamma-ray decays to $^{153}\text{Gd}$, is less discrepant which suggests that the efficiency curve in the regions of interest are correct [120]. These data are therefore indicative that the nuclear decay data for the $^{153}\text{Gd}$ is incorrect. This is stated in the Monographie itself [120]. “The discrepancy for $^{153}\text{Gd}$ is probably due to the nuclear data.”

Figure 5.1: The relative difference of modelled effective activity measurements vs actual effective activity measurements. $^{153}\text{Gd}$ has a measured activity much different to the modelled activity with respect to all other KCRV comparisons.

5.2 Nuclear data of $^{153}\text{Gd}$

The ground state of $^{153}\text{Gd}$ is $J^\pi = 3/2^-$. The most dominant decay by electron capture is to the first forbidden first $J^\pi = 3/2^+$ state. This decay has a log(ft) value of 7.7. The second most intense electron capture is to the allowed Gamow-Teller transition to the first excited $5/2^-$, with the same evaluated log(ft) value of 7.7. The $3/2^+$ decays to the
ground state $5/2^+$ via a mixed M1-E2 transition. This decay has an internal conversion coefficient ($\alpha_T$) as measured by the BRICC code [43] to be 1.64. The $5/2^-$ level decays purely to the ground state of $^{153}$Eu via an unstretched E1 transition ($\alpha_T = 0.305$). The decreased internal conversion coefficient of the $5/2^- \rightarrow 5/2^+$ causes the photon transition (97.4 keV) to be the most intense and by definition have the largest $P_\gamma$ value.

Past these principal decays the decay scheme of this nucleus is not well known, with evaluators disagreeing in the two most recent evaluations [97, 122] about the proposed population of the second $7/2^-$ and the $7/2^+$. The decay of the nucleus can be seen in figure 5.2, with data taken from the DDEP evaluation [97], the 151.6 keV second $7/2^-$ and the 266.7 keV second $7/2^+$ are omitted.

**Figure 5.2:** The partial decay scheme of the electron capture branch of $^{153}$Gd to $^{153}$Eu taken from DDEP [97].

Measurements of the decay of this nuclide are numerous, with over ten measurements noted in the latest evaluations. It is worth discussing the different techniques employed
in all these previous studies. Figure 5.3 is taken from the notes of the most recent evaluation by DDEP taken by Bé et al. [97], a discussion of the previous work will use the same NSR reference numbers for each paper for simplicity.

A brief description of the most related previous measurements of the absolute intensity of the gamma rays from the decay of $^{153}\text{Gd}$ will now be discussed.

In total there are nine previous measurements of the relative intensities of gamma rays in the decay of $^{153}\text{Gd}$ of which some measure the absolute emission probabilities also. The earliest measurements, Heath (1974HEYW) [123], Sergienko and Lebedev (1974SE08) as well as Chechev et al. (1992CH16) [124] utilised Lithium-drifted Germanium (Ge(Li)) detectors although this was used in conjunction with a Lithium-drifted Silicon (Si(Li)) detector in the case of (1992CH16). A Lithium drifted Germanium detector is a precursor to a HPGe detector with poorer resolution. Other measurements have been made by Singh et al., (1985SI03)[125], Subramenyeswara Rao et al., (1988SU13)[126], Venkateswara Rao et al., (1988VE05)[127], Kumar et al., (1995KU34)[128] and Chand et al.,(1992CH44)[129]. In these cases a HPGe detector was employed often with a Si(Li) detector to detect the lower-energy gamma-ray emissions and the K-Capture radiations. In the case of Singh et al, the study was to deduce the different transition rates of K-Capture with different chemical forms of $^{153}\text{Gd}$. All measurements were made close to the detector, less than 15 cm away from the front face of the detector. True Coincident Summing (TCS) was therefore considered in all measurements. In the case of Kumar et al., absolute intensity values were deduced from considering the internal conversion coefficients and the total transition rates from all ground-decaying levels. A table summarising the most relevant measurements is provided in figure 5.3 taken from the evaluators’ notes of the most recent DDEP evaluation of this decay.

As has been noted by Chechev [130], weakly populated decays of this nuclide are in disagreement. Also of note, is that there is some disagreement in the principal decays relative to the strongest 97.4 keV transition. These measurements in comparison to the current work are considered in this chapter.
### Comments on evaluation

#### Table 1. Relative Gamma emission Intensities

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<td>204 (45)</td>
<td>65.2 (14)</td>
<td>65.2 (14)</td>
<td>69.2 (19)</td>
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<td>82.6 (5)</td>
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<tr>
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<td>78.9 (11)</td>
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<td>82.6 (5)</td>
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<td>0.057 (2)</td>
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<td>0.02 (1)</td>
<td>&lt; 0.030</td>
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<td>0.13 (1)</td>
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<td>0.33d</td>
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<td>0.02 (1)</td>
<td>&lt; 0.030</td>
<td>0.0172 (9)</td>
<td>9.1 (27)</td>
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<td>0.28g</td>
<td>0.144 (26)</td>
<td>0.10 (2)</td>
<td>0.13 (1)</td>
<td>0.12 (1)</td>
<td>0.125 (6)</td>
<td>0.56 (6)</td>
<td>0.56 (6)</td>
</tr>
</tbody>
</table>

- **a** Value is uniquely low, omitted from weighted average calculation.
- **b** No uncertainty, omitted from weighted average calculation.
- **c** Sum of Kα1 and Kα2 and sum of Kβ1' and Kβ2' used in weighted average calculation.
- **d** Limits are omitted from weighted average calculation.
- **e** LRSW method increased uncertainty in order to reduce relative weight to 50 %.
- **f** LRSW method gives unweighted average of 0.049 (43).
- **g** Computed from γ-ray intensity balance at 83-keV level and α(19,E2) and from internal-conversion electron data and α(19,E2).

---

**Figure 5.3:** The evaluated value of each gamma emission probability for 153Gd, along with individual measurements from each dataset given in NSR reference.
5.3 Production of $^{153}\text{Gd}$

Gadolinium-153 is produced by the thermal neutron irradiation of a natural europium oxide target (47.8 % $^{151}\text{Eu}$ and 52.2 % $^{153}\text{Eu}$[131]). This target produces $^{153}\text{Gd}$ by the neutron capture to $^{152,152m}\text{Eu}$, which then decays to the stable isotopes $^{152}\text{Gd}$, this isotope then captures a neutron to $^{153}\text{Gd}$. Other isotopes in the natural target also have high neutron cross sections, this coupled with the competing decay channels create many impurities in the irradiated target. These impurities are separated by various radiochemical separations, which are explained in [131]. The production scheme of $^{153}\text{Gd}$ by this technique is shown in figure 5.4, adapted from [131].

![Figure 5.4: The production of $^{153}\text{Gd}$ by the irradiation of a natural europium target. The nuclear decay data is taken from [30], thermal neutron cross sections are taken from [131].](image)

5.4 $P_\gamma$ measurements using HPGe detectors at NPL

5.4.1 Sample preparation of the measurement sources

A sample of $^{153}\text{Gd}$ was acquired by NPL in 2011, from which an aqueous solution of $^{153}\text{GdCl}_3$ in 0.5 M HCl. Two types of sources from this sample were prepared; 1) to measure the specific activity using DCC, and 2) for gamma-ray spectroscopy.

One drop of the solution was gravimetrically dispensed into twelve 20 mL glass liquid scintillation vials, with 1 mL of carrier and 10 mL of scintillant. Approximately 1 g of solution was added dispensed into three British Standards (BS) 2 mL capacity ampoules. These ampoules were flame sealed and used to acquire gamma-ray spectrum by HPGe measurement. One of these ampoules can be seen in figure 5.5.
5.4.2 Gamma-ray spectrometry by HPGe

In this study two n-type HPGe detectors were used to acquire the spectra.

The two detectors utilised are given the handles of BART and LOKI locally. The relative efficiency of the two detectors compared to a 3”×3” NaI(Tl) detector are 28 % and 22 % respectively. The energy resolution of the detectors, using the parameter of the Full-Width-Half-Maximum are 1.78 keV and 1.68 keV at 1332 keV respectively. The detector LOKI rather than the usual aluminium front face has a carbon fibre window. Both detectors are housed in “old” lead to reduce background, old in this case refers to before the nuclear age. One of these lead coffins, shown in figure 5.6, are of dimensions 1.5 m wide, with 1 m depth and width; the lead itself is 10 cm thick. To suppress lead fluorescence, two thin metal degraders of 0.5 mm Cd 0.7 mm Copper are used in between the lead. Bread boards on which kinematic mounting plates are fixed to were constructed in each detector’s coffin to ensure sources were placed in a precise and reproducible way for each detector. The sample position in this set-up is highly reproducible, with the uncertainty of this contributing 0.016 % to the uncertainty budget, the mounting for such sources is seen in figure 5.6.

The sample to front face distance of the two detectors was larger than in most previous measurements; for BART \( r = 250 \text{ mm} \) and for LOKI \( r = 290 \text{ mm} \). Solid angle (Ω) is therefore small \( \Omega_{BART} = 0.038 \text{ sr} \) and \( \Omega_{LOKI} = 0.045 \text{ sr} \). The benefits of this smaller solid angle is many fold, dead time and pile up are reduced, as is True Coincident
Figure 5.6: Top: A picture of a lead coffin in this study, with a scientist preparing for acquisition. Bottom: The kinematic mounting plates.

Summing (TCS). The smaller these become the smaller the uncertainty these factors produce. The contributions these cause on uncertainty using the guide to uncertainty in measurement (GUM) best practice procedure can be seen in table 5.1. In the case of dead time and pile up the detector BART used a pile-up rejection/live time circuit, and an empirically derived correction factor was used to adjust for events not caught in the circuit. In the case of LOKI a reference pulse was used to adjust for both effects; this pulse can be seen in the acquired spectrum for LOKI shown in figure 5.10. The efficiencies of the detectors have been well characterised and measured in the geometry used, using a multitude of sources, considering the correlations produced when using
lines from the same source. The full-energy-peak detection efficiency for the two detectors, and the corresponding efficiencies are seen in figures 5.7 and 5.8. These two plots are taken from [132] for LOKI and [101] for BART.

![Graph](image)

**Figure 5.7:** FEP detection efficiency for BART and the corresponding residuals reproduced from [101].

To acquire spectra an analogue electronics chain was used for both detectors, in both cases this was a amplifier connected to an ADC connected to a computer running CANBERRA GENIE 2000 software. Acquisitions of the three sources were between 86,400 s and 200,000 s.

As stated previously, $^{153}$Gd, is produced most usually by the neutron capture on a natural Europium target. There are several channels open in this irradiation and sources can be contaminated. This study was taken over five years. In these acquired spectra, no contamination was seen other than the expected naturally occurring radioactive material (NORM) peaks.
5.4.3 Peak area estimation using GENIE 2000

The peak area in GENIE 2000 is determined by two possible techniques, the first is by the peak fitting algorithm in which a Gaussian is fitted, and the parameters of the fit are integrated. The second is the total peak area method as defined in Practical Gamma-ray Spectrometry [133]. This method takes the total integral given by the number of counts defined in the peak region.

\[ G = \sum_{i} = LC_i \]  

(5.1)

A background continuum can be defined by a function between several points either side of the region of interest (ROI). In GENIE this function can be a step function or a polynomial function. In the most simple case as the trapezoid area between two background bins either side of the ROI;

\[ B = \frac{n(C_{L-1} + C_{U+1})}{2} \]  

(5.2)

In all cases the peak area, \( A \) can be expressed as \( A = G - B \).

This peak area is then treated further by the a contamination removal of previously reported gamma rays that are present in a source-free acquisition. In all measurements peak areas were measured autonomously with the peak area analysis tool inside GENIE 2000, and was then modified and optimised by hand. The peak areas are then corrected for the decay during the acquisition, and self absorption, were gamma rays are attenuated by the sample itself.
Figure 5.8: FEP detection efficiency for LOKI and the corresponding residuals reproduced from [132]. 60 radionuclides were used in this procedure.
5.5 Determining the specific absolute activity of $^{153}\text{Gd}$

The specific activity of the samples were measured using the $4\pi - (LS)$ DCC (Digital Coincidence Counting) technique. This was developed and carried out by Dr John Keightley at the National Physical laboratory. Further reading on this subject is available [99]. A value of $512.2(25)$ kBqg$^{-1}$, was calculated.

5.6 Relative $P_\gamma$ measurements of $^{153}\text{Gd}$

Six of the previous gamma rays have been measured with consistency in this study. The three lowest energy gamma rays that have been previously reported, 14.0 keV, 19.8 keV and 21.2 keV, are not reported in this study due to the poor sensitivity of the HPGe detectors used at these energies. The three gamma rays are noted in early acquisitions however (see figure 5.9). The 54.1 keV transition which would connect the hanging state placed in the decay by [122] is not seen, however the 166.1 keV decay line is, but with no consistency in measurement and is therefore not reported in the current work. This is most likely due to the very weak population of the state. Similar arguments can be made for the 68.2 keV emission, in which the measurement is hindered by the well-populated 69.6 keV emission. Finally, the 83.9 and 75.4 keV emission lines, are noted, but again not measured with consistency as both lie atop a complicated Compton continuum which is made worse by the prevalent Pb fluorescence peaks for the 75.4 keV gamma-ray peak.

A spectrum taken in 2011 using the BART spectrometer can be seen in figure 5.9, and a later acquisition in 2016 using LOKI can be seen in figure 5.10. The deviations of the set of BART measurements for each reported emission can be seen in figures 5.11, 5.12 and 5.13.

The table of relative gamma-ray emissions can be seen in table 5.2. The uncertainty budget for the measurement of the 97.4 keV gamma ray is given in table 5.1.

5.6.1 Comparison of relative intensities with evaluated and reported data

It is important to contextualise the measurements given above by comparing them to the previous and the evaluated data.
Chapter 5. \textsuperscript{153}Gd

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5_9.png}
\caption{An early spectrum acquired using BART, of one of the three \textsuperscript{153}Gd source, several sum peaks are visible.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5_10.png}
\caption{A late spectrum acquired on LOKI of a \textsuperscript{153}Gd, the lesser populated gamma-ray decays are no longer above background.}
\end{figure}

The measurement calculated using in this study are now compared to all measurements previously published for the reported values.

All of the measurements reported in this work are resolved with better precision than
Figure 5.11: Left: Comparison of relative $P_\gamma$ to the 97.4 keV emission measurements of the 69 keV emission in the BART measurements. Right: Comparison of relative $P_\gamma$ to the 97.4 keV emission measurements of the 89.4 keV emission.

Figure 5.12: Left: Comparison of relative $P_\gamma$ to the 97.4 keV emission measurements of the 103.1 keV emission in the BART measurements. Right: Comparison of relative $P_\gamma$ to the 97.4 keV emission measurements of the 151.6 keV emission.

Figure 5.13: Comparison of relative $P_\gamma$ to the 97.4 keV emission measurements of the 172.8 keV emission.

all previous work. As can be seen reasonable agreement is noted with 1992CH44, but not with 1992CH16. It is curious that this is the case considering the nature of the two results is very similar. Indeed in most cases, where reported the counts per second of the 97.4 keV emission is comparable.
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Table 5.1: The uncertainty budget of the 97.4 keV measurement presented in this work.

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<th>Evaluated Value</th>
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<td>$I_{\gamma}(E)/I_{\gamma}(97.4)$</td>
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<td>89.4</td>
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</tr>
<tr>
<td>103.1</td>
<td>22.33 (17)</td>
<td>74.06 (27)</td>
</tr>
<tr>
<td>151.6</td>
<td>0.00474 (19)</td>
<td>0.0157 (10)</td>
</tr>
<tr>
<td>172.8</td>
<td>0.0396 (6)</td>
<td>0.1313 (18)</td>
</tr>
</tbody>
</table>

Table 5.2: Reported relative intensities of the principal gamma rays measured in this work compared to the evaluated values.

Figure 5.14: Left: Comparison of relative $P_{\gamma}$ measurements of the 69 keV emission. Right: Comparison of relative $P_{\gamma}$ measurements of the 89 keV emission.
Figure 5.15: Left: Comparison of relative $P_\gamma$ measurements of the 103 keV emission. Right: Comparison of relative $P_\gamma$ measurements of the 151 keV emission.

Figure 5.16: Comparison of relative $P_\gamma$ measurements of the 172 keV emission.
5.7 Absolute activity comparison with historical values

The absolute intensity of the 97.4 keV gamma ray using the result of the absolute activity standardisation yields 30.15(20) % this value is not in agreement ($k = 2$) to the adopted value of 29.0(8) % which is the value reported by 1992CH44. The two values have a relative difference of 4.0 %, which is considerable. The differences in the measurand here is echoed in the relative intensities. A comparison of all previous measured absolute intensities can be seen in figure 5.17.

![Figure 5.17: Comparison of the absolute $P_\gamma$ value for the 97.4 keV emission, empty squares show experimental measurements, filled squares modelled values.](image)

The value reported in this thesis is in agreement with all other measured intensities. It should be stated at this point that the cited paper of Coursey, does not include the value of the 97.4 keV absolute intensity, and perhaps this value is not considered in previous evaluations due to this. Furthermore the technique used by Laurec, the equilibrium method, is only valid when the ground to ground decay path is negligible, as seen in figure 5.2, this value is 4 % which may not be considered suitable. Finally the value given by Geidelman could be argued to be superceded by Chechev, although attaining
this paper is not without difficulty so it is problematic to ascertain if the same techniques and equipment was used to arrive at the value given.

If the absolute gamma intensity of the 97.4 keV gamma ray presented in this work is used in the SIRIC model, the modelled equivalent activity, recall the same activity of Radium to produce the same response, is 368.4 kBq, the measured equivalent activity of the KCRV of this nuclide is 368.1(17) kBq. The two values are in good agreement with a z-score, or standard score, of 0.26 and a relative difference of 0.18 %. The reduction in the relative difference of is of the order of the difference in nuclear data, as expected and suggests as clearly as any method can that the current adopted value is less accurate as well as less precise than the value of the measurand obtained in this work.

<table>
<thead>
<tr>
<th>Eγ</th>
<th>Ei</th>
<th>Ef</th>
<th>Ii</th>
<th>If</th>
<th>Multipolarity</th>
<th>αT</th>
<th>Pγ</th>
<th>Pγ</th>
<th>Pγ</th>
<th>Pγ</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
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<td></td>
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<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td>Absolute</td>
<td>Relative</td>
<td>Absolute</td>
<td>Relative</td>
</tr>
<tr>
<td>69.6</td>
<td>172.8</td>
<td>103.1</td>
<td>3/2+</td>
<td>3/2+</td>
<td>M1+1.8%E2</td>
<td>5.31 (8)</td>
<td>2.38 (3)</td>
<td>7.89 (9)</td>
<td>2.42 (7)</td>
<td>5.28 (10)</td>
</tr>
<tr>
<td>89.5</td>
<td>172.8</td>
<td>84.4</td>
<td>3/2+</td>
<td>7/2+</td>
<td>M1+6.2%E2</td>
<td>2.60 (7)</td>
<td>0.996 (5)</td>
<td>0.320 (16)</td>
<td>0.069 (3)</td>
<td>0.245 (14)</td>
</tr>
<tr>
<td>97.4</td>
<td>97.4</td>
<td>0</td>
<td>5/2+</td>
<td>5/2+</td>
<td>E1</td>
<td>0.305 (5)</td>
<td>30.15 (20)</td>
<td>190</td>
<td>29.0 (8)</td>
<td>190</td>
</tr>
<tr>
<td>163.1</td>
<td>103.1</td>
<td>0</td>
<td>3/2+</td>
<td>5/2+</td>
<td>M1+4.5%E2</td>
<td>1.694 (24)</td>
<td>22.33 (17)</td>
<td>74.06 (27)</td>
<td>21.1 (6)</td>
<td>72.9 (7)</td>
</tr>
<tr>
<td>151.6</td>
<td>151.6</td>
<td>0</td>
<td>7/2+</td>
<td>5/2+</td>
<td>E1</td>
<td>0.092</td>
<td>0.00474 (29)</td>
<td>0.0157 (10)</td>
<td>0.024 (5)</td>
<td>0.017 (4)</td>
</tr>
<tr>
<td>172.9</td>
<td>172.9</td>
<td>0</td>
<td>5/2+</td>
<td>5/2+</td>
<td>M1+E2</td>
<td>3.47 (6)</td>
<td>0.0896 (6)</td>
<td>0.1313 (18)</td>
<td>0.036 (2)</td>
<td>0.125 (6)</td>
</tr>
</tbody>
</table>

Table 5.3: A summation of the nuclear decay data of the measured gamma ray emissions in this work with the comparison of the evaluated nuclear data. 1 Internal conversion coefficients taken from [43], using the BAND frozen orbital approximation.
Chapter 6

Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag at RIBF-RIKEN

This chapter presents the analysis of the $\beta$-delayed $\gamma$-ray coincidence spectroscopy of very neutron rich rhodium and silver isotopes produced following the fission of a primary $^{238}$U beam impinging in a thick beryllium target. The experiment was carried out at the RIBF-RIKEN facility, Japan, with the primary fragments identified using the standard $Bp-\Delta E-TOF$ technique using the BigRIPS spectrometer. The primary fragments were brought to rest at the final focus of the spectrometer, and detected using WAS3ABi. Discrete energy gamma rays following the $\beta^-$ decay of the fragments were detected in the EURICA set up. Experimental details and results from this experiment can be found in the previously published works. [13]

6.1 Neutron rich rhodium isotopes

The heavy rhodium isotopes produced in this experiment are a good testing ground for what is known in the limit of the nuclear shell model. Precise measurement of the bulk decay parameters, $P_n$, $T_1/2$ of these isotopes and decay characteristics such as $P\gamma$ may be important.
Using the datasets created in the EURICA campaign at RIKEN, five rhodium isotopes from $A = 120$-$124$ were strongly enough populated, such that their beta-delayed $\gamma$ spectrum could be studied. In this chapter, the analysis of these data is presented.

The rhodium isotopes were selected according to the standard particle selection PID procedure using the detectors at F3 and F7 of the spectrometer to resolve the mass over charge ($A/q$) and the charge ($Z$) of the ions, more detail on these detectors is given in [69]. Assuming that the ions were fully stripped of atomic electrons the charge measured, is then equal to $Z$.

Selecting the primary fragments of interest depicted in figure 6.2, $\beta - \gamma$ time correlated datasets for individual isotopes were created. From the rhodium and daughter palladium isotope data limits of the $P_n$ value can be deduced, as well as the excited energy level schemes for the even-even palladium isotopes populated in the decay of rhodium.

### 6.1.1 Methodology for calculating the $P_n$ value for $^{120-124}$Rh

A limit of the $P_n$ value can be calculated in the following way. The data structure includes; 1) the time $t$, between ion implantation and $\beta$ implantation in WAS3ABi.
as well as position resolution, 2) time correlated gamma-rays, correlated to the beta implantation time \( t \) relative to the ion implantation, and with each other within a time window of less than 1 \( \mu s \).

A spectrum of the time difference distribution of beta implantation with respect to ion implantation for each rhodium isotope can be created from this data. A fit, according to the Bateman equations can be made on this data and the total number of ions implanted on WAS3ABi extracted, as well as the half-life of the parent nuclei, with all other half-lives of interest being fixed parameters. A gamma-ray spectrum of the gamma rays correlated to the implantation of the isotope can also be created, with a conditions set on the beta-ion correlation time, the mass over charge (\( A/q \)), and the position of the related beta decays within WAS3ABi to the ion. In the even-even nuclides it is a reasonable assumption that the beta decay feeds the yrast \( 2^+ \) state in the daughter nuclei. The absolute intensity of the \( 2^+ \rightarrow 0^+ \) decay, which can be determined using the calculated efficiency of the EURICA spectrometer is then equal to the \( P_n \) value. This relationship is shown in equation 6.1. For even rhodium isotopes the absolute intensity of the \( 2^+ \rightarrow 0^+ \) decay in the same mass even-even palladium daughter corresponds to the intensity of the pure beta decay and therefore is the equal to \( 1-P_n \). The assumption that all decays populate the \( 2^+ \) state in the daughter may not be valid, and this calculated
value for the $P_n$ value is an upper limit. In the case of the odd rhodium isotopes the intensity of the $2^+$ decay in the even-even palladium daughter corresponds to the absolute intensity of the beta delayed neutron emission. This is equal to the $P_n$ exactly; in these cases this value represents a lower limit.

$$P_n = 1 - I_T = 1 - \frac{A_\gamma + A_\gamma \alpha}{N_0 - N_0 e^{-\lambda t}} \quad (6.1)$$

Where $A_\gamma$ is the efficiency corrected peak area of the $2^+ \rightarrow 0^+$ gamma ray, $\alpha$ is the total internal conversion coefficient, as determined by the BRICCC code and $\lambda$ is the fit-derived halflife of the implanted (rhodium) ion. $N_0$, the total implantation of the ion is;

$$N_0 = N_0 |_{P_n=0}(P_n, \lambda) \quad (6.2)$$

The correlation time, $t$, between gamma rays (or beta implants) with ion implantation is set according to the peak to total of the peaks as well as maximising the area of the peak itself. The area of these gamma rays were calculated according to fits made in the gf3 program of the Radware analysis suite, the fit according to Bateman was made according to a log-likelihood fit in ROOT.

A complication of the technique is that the halflife of the parent nucleus, and total implantations, is fit according to the $P_n$ value, i.e the output of the equation. To handle this, the procedure is iterated to the point where $P_{no} = P_{ni}$.

### 6.2 $P_n$ limit of $^{120}$Rh

In previous measurement the decay for rhodium has been measured using the NSCL at MSU using a $^{136}$Xe beam at 120 MeV/nucleon [134]. In that work, a 438 keV gamma ray was observed along with a 618 keV, 901 keV and 1123 keV transitions. The most intense, 438 keV gamma ray, was assigned to be the depopulation from the first $2^+$ excited state of $^{120}$Pd.

A fit of the time distribution between implanted $^{120}$Rh ions and the subsequent spatially correlated $\beta^-$ particles in WAS3ABi is shown in figure 6.3, from this a halflife for the nuclide was deduced to be 140(5) ms, which is in agreement with [13]. Using this fit, a
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.3: The fit of beta implantations according to the Bateman equation. In this fit, the half-life of the daughters were set to the values measured in [13]. The $P_n$ value of this fit was set to 0.089 as measured using the technique outlined.

correlated $\beta$-ion correlation time of 480 ms was set, from which a gamma ray spectrum was produced. In this time 33% of the daughter ions will have decayed, but 90% of the parent $^{120}$Rh will have decayed, improving the peak to total of the gamma spectrum.

The $\beta$ correlated gamma-ray spectrum was prepared for peak determination in the gf3 program part of the Radware analysis suite. By subtracting a normalised spectrum created from the daughter ($^{120}$Pd) gated dataset, gamma-ray lines associated with the decay of the daughter and granddaughter ($^{120}$Pd $\rightarrow ^{120}$Ag $\rightarrow ^{120}$Cd) are eliminated. This leaves only gamma rays from the decay of $^{120}$Rh to $^{120}$Pd or gamma rays associated with the beta delayed neutron decay of the nucleus. In total there are approximately $10^5$ decays of $^{120}$Rh. A correlated $\gamma-\gamma$ coincidence matrix can be used to differentiate the emissions in the excited $^{120}$Pd and emissions related to the $\beta$-n channel. For the decay to $^{120}$Pd it is assumed that the most intense gamma ray in the decay is the decay of the yrast $2^+$ state to the ground state. By gating on this gamma ray in a $\gamma\gamma$ matrix the coincident gamma rays can be placed in a level scheme. The spectra of interest are shown in figure 6.4, created using a beta-ion correlation time of 2.350 s, and the
corresponding partial level scheme for excited states of $^{120}$Pd populated in the decay of $^{120}$Rh is shown in figure 6.5.

The $\beta - \gamma$ gated $^{120}$Rh spectrum (figure 6.4) was used to measure the peak areas of the 6 gamma rays assigned to the direct $\beta$ decay of $^{120}$Rh, intensities relative to the $2^+ \rightarrow 0^+$ transitions were determined, and absolute gamma-ray intensities calculated. Finally the absolute decay intensity for the $2^+ \rightarrow 0^+$ transition was used to give an upper limit on the $\beta$-n decay probability to $^{119}$Pd, from the missing intensity in the transition. The $P_n$ value for this nuclide was determined to be 0.105(4). These results are presented in table 6.1.

As previously mentioned the input $P_n$ value is used to calculate the output $P_n$ value, to deal with this the difference between the $P_n$ inputted ($P_{ni}$) and the output $P_{no}$ can be plotted, the zero crossing point is then the central $P_n$ value going forward. This is shown in figure 6.6, for the decay of $^{120}$Rh where the central value was determined to be 0.089.

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$I^\pi \rightarrow I^\pi$</th>
<th>Relative $P_\gamma$</th>
<th>Absolute $P_\gamma$</th>
<th>Transition intensity</th>
<th>$P_n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>435.6</td>
<td>$2^+ \rightarrow 0^+$</td>
<td>1.00(4)</td>
<td>0.91(4)</td>
<td>0.91 (4)</td>
<td>≤ 0.089 (4)</td>
</tr>
<tr>
<td>617.9</td>
<td>$4^+ \rightarrow 2^+$</td>
<td>0.60(4)</td>
<td>0.541(23)</td>
<td>0.543 (23)</td>
<td></td>
</tr>
<tr>
<td>737.6</td>
<td>$6^+ \rightarrow 4^+$</td>
<td>0.308(22)</td>
<td>0.278(16)</td>
<td>0.308 (22)</td>
<td></td>
</tr>
<tr>
<td>795.9</td>
<td>-</td>
<td>0.0532(14)</td>
<td>0.048(13)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>899.8</td>
<td>-</td>
<td>0.303(25)</td>
<td>0.274(19)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>465.2</td>
<td>-</td>
<td>0.283(21)</td>
<td>0.256(15)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>129.3</td>
<td>-</td>
<td>0.06(4)</td>
<td>0.05(3)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 6.1: Relative and absolute intensities of the gamma-rays identified to be associated with the excited levels of $^{120}$Pd. Tentative spin and parity assignments have been made for the two lowest lying states in agreement with [134]. The upper limit of the $P_n$ value of this nuclide is also included calculated from the missing total intensity of the 435.6 keV emission.

The gamma ray transitions seen in coincidence with the 436 keV transition agree with the previously reported gamma rays by Stoyer et al., [135], with slight disagreements in gamma-ray energy (2 keV for the 436 keV transition). This work did not see the 881 kev or 824 keV transitions belived to depopulate the $10^+$ in $^{120}$Pd, however 3 gamma-rays previously unreported have been observed for the first time. The 129, 465 and 900 keV transition, which were all seen in coincidence with the 436 keV transition. The $P_n$ for the decay of $^{120}$Rh was deduced to be 0.091(4) (i.e. 9.4 %); this upper limit is approximately a factor of 2 larger than reported by Montes et al., [136] (4%).
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.4: Gamma-ray spectra associated with the decay of (a) $^{120}$Rh and (b) $^{120}$Pd. (c) A $^{120}$Pd subtracted $^{120}$Rh spectrum showing events associated with the primary decay of $^{120}$Rh only, which are assumed to be gamma-ray transitions from excited states in the $^{120}$Pd daughter nucleus. (d) Gamma-ray coincidence spectra gated on the 436 keV $^2 + \rightarrow 0^+$ transition in $^{120}$Pd. The $\beta - \gamma$ correlation times for each of these spectra was 2.350 s.
Figure 6.5: Partial level scheme showing excited states populated in $^{120}$Pd following the beta decay of $^{120}$Rh deduced from the present work.
Figure 6.6: Evolution of the difference in input and output $P_n$ value with changing input $P_n$, highlighting the process in which the central value for the limit of $P_n$ is arrived at for each nuclei.
6.2.1 Decay of $^{121}$Rh

For the decay of $^{121}$Rh, the inverse is true for the determination of the $P_n$ compared to the even A $^{120}$Rh. In this case, the area of the $2^+$ peak in the decay of $^{120}$Pd corresponds to the lower limit of the $P_n$ value. In this decay the lower limit of the $P_n$ value was determined to be 0.123(5) (= 12.3(5)%) as determined from the deduced ion implantation from the fit of the beta-ion time difference spectrum ($1.04(1) \times 10^5$) and the total intensity of the measured 436 keV transition in $^{120}$Pd, populated via the $\beta$-n decay channel. The gamma-ray spectrum of the A/q tagged $^{121}$Rh events are shown in figure 6.7, also shown is the gamma-ray spectrum of $^{121}$Pd events, both of these spectrum were created using a beta-ion correlation time of 1.36 s. By subtracting this spectrum from the $^{121}$Rh spectrum, signature emissions (like the 436 keV gamma ray) used to calculate the $P_n$ following the decay of $^{121}$Rh into $^{120}$Pd are observed above background. A table of the prominent gamma-ray emissions related to the excited levels in $^{120,121}$Rh is presented in figure 6.2. Absolute gamma-ray intensities were calculated according to the efficiency curve of EURICA shown in figure 3.7.

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$I^\pi \rightarrow I^\pi$</th>
<th>Absolute $P_\gamma$</th>
<th>$P_n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>135</td>
<td>-</td>
<td>0.154(5)</td>
<td></td>
</tr>
<tr>
<td>150.5</td>
<td>-</td>
<td>0.074(4)</td>
<td></td>
</tr>
<tr>
<td>283.5</td>
<td>-</td>
<td>0.051(4)</td>
<td></td>
</tr>
<tr>
<td>338</td>
<td>-</td>
<td>0.146(4)</td>
<td></td>
</tr>
<tr>
<td>396</td>
<td>-</td>
<td>0.052(3)</td>
<td></td>
</tr>
<tr>
<td>483</td>
<td>-</td>
<td>0.065(4)</td>
<td></td>
</tr>
<tr>
<td>493</td>
<td>-</td>
<td>0.098(4)</td>
<td></td>
</tr>
<tr>
<td>681.4</td>
<td>-</td>
<td>0.022(3)</td>
<td></td>
</tr>
<tr>
<td>435.6</td>
<td>$2^+ \rightarrow 0^+$</td>
<td>0.114(6)</td>
<td>$\geq 0.115(6)$</td>
</tr>
</tbody>
</table>

Table 6.2: Absolute $P_\gamma$ values for gamma ray emission following the decay of $^{121}$Rh, from the absolute gamma-ray intensity of the $2^+ \rightarrow 0^+$ transition in $^{120}$Rh, a $P_n$ value of 0.115(6) has been determined for the decay of $^{121}$Rh.

The previously observe isomeric internal transition in $^{121}$Rh of 136 keV ($T_{1/2} = 46(9)$) [137] is observed strongly in this work. Seven further transitions observed in the decay of $^{121}$Pd are reported, but without placement in a level scheme.
Figure 6.7: Gamma-ray spectra associated with the decay of (a) $^{121}$Rh and (b) $^{121}$Pd. c) A $^{121}$Pd subtracted $^{121}$Rh spectrum showing events associated with the primary decay of $^{121}$Rh only, which are assumed to be gamma-ray transitions from excited states in the $^{120,121}$Pd daughter nuclei. The $\beta - \gamma$ correlation times for each of these spectra was 1.36 s.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

6.2.2 Decay of $^{122}$Rh

Two excited levels of $^{122}$Pd have been previously observed by Wang et al., [138]. In that experiment at RIBF-Riken, a $^{238}$U beam was impinged upon 0.5 mm thick natural Tungsten target. Gamma rays from the de-excitations of the secondary beam were Doppler-corrected and measured using the DALI2 spectrometer [139]. A 499(9) keV and a 665(18) keV gamma ray were observed on the $^{122}$Pd gated spectrum and were identified as the first $2^+ \rightarrow 0^+$ and $4^+ \rightarrow 2^+$ transitions respectively. In this work strongly-fed 513 keV and 674 keV gamma rays were observed, which agree with the results published from [138]. These transitions can be seen in figure 6.8, these gamma-ray spectra were created using a beta-ion correlation time window of 0.930 s. The proposed 513 keV $2^+ \rightarrow 0^+$ transition was gated upon in $\gamma\gamma$ coincidences, this spectrum is shown in c) of figure 6.8. Other energy gates corresponding to energies of transitions given in table 6.3 were imposed upon the $\gamma\gamma$ matrix, which were used to build the level scheme shown in figure 6.9. $1.12(5) \times 10^5$ $^{122}$Rh were implanted, from which the absolute intensities were calculated given in table 6.3.

<table>
<thead>
<tr>
<th>$E_{\gamma}$ (keV)</th>
<th>$I^\pi \rightarrow I^\pi$</th>
<th>Relative $P_\gamma$</th>
<th>Absolute $P_\gamma$</th>
<th>Transition intensity</th>
<th>$P_n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>512.9</td>
<td>$2^+ \rightarrow 0^+$</td>
<td>1.00(16)</td>
<td>0.966(15)</td>
<td>0.961(15)</td>
<td>≤ 0.0387(6)</td>
</tr>
<tr>
<td>674.9</td>
<td>$4^+ \rightarrow 2^+$</td>
<td>0.686(16)</td>
<td>0.655(10)</td>
<td>0.657(10)</td>
<td></td>
</tr>
<tr>
<td>691.0</td>
<td></td>
<td>0.0779(23)</td>
<td>0.0745(18)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>438.1</td>
<td></td>
<td>0.246(12)</td>
<td>0.236(11)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>150.1</td>
<td></td>
<td>0.112(9)</td>
<td>0.107(8)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>557.5</td>
<td></td>
<td>0.096(5)</td>
<td>0.092(5)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>758.3</td>
<td></td>
<td>0.17(3)</td>
<td>0.159(24)</td>
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<tr>
<td>748.7</td>
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<td>0.038(8)</td>
<td>0.036(8)</td>
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<td>448.3</td>
<td></td>
<td>0.072(8)</td>
<td>0.069(7)</td>
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</tbody>
</table>

Table 6.3: Relative and absolute intensities of the gamma-rays identified to be associated with the excited levels of $^{122}$Pd. Tentative spin and parity assignments have been made for the two lowest lying states in agreement with [134]. The upper limit of the $P_n$ value of this nuclide is also included calculated from the missing total intensity of the 513 keV emission.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.8: Gamma-ray spectra associated with the decay of (a) $^{122}$Rh and (b) $^{122}$Pd. c) A $^{122}$Pd subtracted $^{122}$Rh spectrum showing events associated with the primary decay of $^{122}$Rh only, which are assumed to be gamma-ray transitions from excited states in the $^{122}$Pd daughter nucleus. (d) Gamma-ray coincidence spectra gated on the 513 keV $2^+ \rightarrow 0^+$ transition in $^{122}$Pd. The $\beta - \gamma$ correlation times for each of these spectra was 0.930 s.
Figure 6.9: Partial level scheme showing excited states populated in $^{122}$Pd following the beta decay of $^{122}$Rh deduced from the present work.
6.2.3 Decay of $^{123}$Rh

No previous gamma rays have been reported for the decay of $^{123}$Rh to the excited states in $^{123}$Pd. Using the $\beta^-$-decay daughter-gated gamma-ray spectrum a subtraction spectrum was created. This subtraction spectrum allows for the identification of gamma-rays observed only in the decay of $^{123}$Rh and not in the daughter or granddaughter. The $2^+ \to 0^+$ transition in $^{122}$Pd is observed in the spectrum which is shown in figure 6.10. The fitted time difference spectrum of the beta implantations determined that during the experiment $2.73(3) \times 10^4$ $^{123}$Rh ions were implanted. Absolute intensities were determined for the de-excitation gamma rays observed in figure 6.10, these results are shown in table 6.4. These spectra were created using a beta-ion correlation window of 0.910 s.

$$
\begin{array}{c|c|c|c}
E_\gamma (\text{keV}) & \Gamma \rightarrow \Gamma & \text{Absolute } P_\gamma & P_n \\
\hline
136.8 & - & 0.05(7) & \\
414.4 & - & 0.064(9) & \\
543.1 & - & 0.051(4) & \\
548.6 & - & 0.30(6) & \\
512.9 & 2^+ \rightarrow 0^+ & 0.253(14) & \geq 0.255(14) \\
674.9 & 4^+ \rightarrow 2^+ & 0.106(9) & \\
\end{array}
$$

Table 6.4: Absolute $P_\gamma$ values for gamma ray emissions following the decay of $^{123}$Rh.

Four gamma-ray intensities have been determined in the de-excitation of $^{123}$Rh to $^{123}$Pd. From the efficiency corrected gamma-ray intensities, a transition intensity including internal conversion can be calculated. This value is equal to the lower limit of the $P_n$ of this decay, which was determined to be 0.255(14).
Chapter 6. *Radioactive decay studies of* $^{120-124}$Rh and $^{129}$Ag

Figure 6.10: Gamma-ray spectra associated with the decay of (a) $^{123}$Rh and (b) $^{123}$Pd. c) A $^{123}$Pd subtracted $^{123}$Rh spectrum showing events associated with the primary decay of $^{123}$Rh only, which are assumed to be gamma-ray transitions from excited states in the $^{122,123}$Pd daughter nuclei. The $\beta - \gamma$ correlation times for each of these spectra was 0.910 s.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

6.2.4 Decay of $^{124}$Rh

In the same experiment discussed for the decay of $^{124}$Rh, two gamma rays, 590(11) keV and 710(19) keV were observed and attributed to the de-excitation of the $2^+ \rightarrow 0^+$ and $4^+ \rightarrow 2^+$ respectively. In this work, two intense gamma rays within uncertainties were observed, at 588 keV and 702 keV. This tentative spin and parity assignment was used to calculate the upper limit on the $P_n$ value of the decay of $^{124}$Rh, using the number of ions implanted, $3.01(4) \times 10^3$, deduced from the fit of the distribution of time differences between ions and positionally-correlated beta implantations in WAS3ABi. Despite the lower statistics of this nuclei, a $\gamma\gamma$ matrix was produced, by gating on the two most intense de-excitations a further two gamma-rays were seen in coincidence and are shown in figure 6.11. These new de-excitations were placed in the level scheme shown in figure 6.12. Corresponding $P_\gamma$ values were calculated using the deduced ion implantation and the peak area of each gamma ray and is presented in table 6.5, where the beta-ion correlation window was set at 0.434 s.

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$I^\pi \rightarrow I^\pi$</th>
<th>Relative $P_\gamma$</th>
<th>Absolute $P_\gamma$</th>
<th>Transition intensity</th>
<th>$P_n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>588.0</td>
<td>$2^+ \rightarrow 0^+$</td>
<td>1.00(9)</td>
<td>0.72(7)</td>
<td>0.72 (7)</td>
<td>≤0.28 (3)</td>
</tr>
<tr>
<td>701.8</td>
<td>$4^+ \rightarrow 2^+$</td>
<td>0.62(7)</td>
<td>0.44(5)(7)</td>
<td>0.44 (5)</td>
<td></td>
</tr>
<tr>
<td>627.0</td>
<td>-</td>
<td>0.48(9)</td>
<td>0.34(6)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>232.8</td>
<td>-</td>
<td>0.22(5)</td>
<td>0.16(3)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 6.5: Relative and absolute intensities of the gamma-rays identified to be associated with the excited levels of $^{124}$Pd. Tentative spin and parity assignments have been made for the two lowest lying states. The upper limit of the $P_n$ value of this nuclide is also included calculated from the missing total intensity of the 588 keV emission.

The deduced limits of the $P_n$ values for the five rhodium isotopes considered in this chapter are given in table 6.13. Some straggling is seen between the even and odd mass isotopes, but a clear increase in $\beta$ delayed neutron emission is observed with increase in mass number. The only previously reported $P_n$ for this chain is the lower limit of ≤ 5.4 by Montes et al., [136] for $^{120}$Rh. The lower limit calculated in this work is not as constrained.
Chapter 6. Radioactive decay studies of $^{120-124}\text{Rh}$ and $^{129}\text{Ag}$

Figure 6.11: Gamma-ray spectra associated with the decay of (a) $^{124}\text{Rh}$ and (b) $^{124}\text{Pd}$. c) A $^{124}\text{Pd}$ subtracted $^{124}\text{Rh}$ spectrum showing events associated with the primary decay of $^{124}\text{Rh}$ only, which are assumed to be gamma-ray transitions from excited states in the $^{124}\text{Pd}$ daughter nucleus. (d) Gamma-ray coincidence spectra gated on the 588 keV $2^+ \rightarrow 0^+$ transition in $^{124}\text{Pd}$. The $\beta - \gamma$ correlation times for each of these spectra was 0.434 s.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.12: Partial level scheme showing excited states populated in $^{124}$Pd following the beta decay of $^{124}$Rh deduced from $\gamma\gamma$ coincidences shown in figure 6.11

Figure 6.13: The upper and lower limits for even and odd mass rhodium isotopes determined in this work
6.3 The decay of $^{129}$Ag

The same analysis procedure as described earlier in this chapter was applied to investigate the decay properties of the $N = 82$ isotone $^{129}$Ag. Figures 6.14 and 6.15 show the $\gamma$ ray spectra for $A = 129$ decays associated with Ag, in a), Cd in b) and the normalised subtraction of daughter from the parent. This spectrum, c), shows the clear population of the 646 keV decay of the $2^+ \rightarrow 0^+$ transition in the $\beta$-n daughter $^{128}$Cd, reported previously by [140]; implying a significant $\beta$-n branch for the decay of $^{129}$Ag. Also present are the 785 keV and 440 keV transitions, depopulating the $J^{\pi} = 4^+$ and $J^{\pi} = 5^-$ states respectively in $^{128}$Cd.

The higher energy spectrum reveals higher energy decays, that are related to the decay of $^{129}$Ag. By creating $\gamma$-gated $\beta$-ion implantation time difference spectra for each proposed transition, and by comparing the spectrum with the same spectrum created using the $\beta$ daughter dataset as well as literature values of the known gamma rays in the daughter decays [141, 142] it is possible to assign these transitions to the decay of $^{129}$Ag exclusively. The excited level scheme of $^{129}$Cd, has a reported isomer, which decays via 4 gamma rays [143]. One of these gamma rays is seen in the spectra shown in figures 6.14 and 6.15, the 1181 keV emission, reported as a $A \gamma - \gamma$ matrix has been created which identifies two further emissions that are very weakly populated; a) a 481 keV decay in coincidence with the 933 keV emission, which within uncertainty sums to 1414 keV, implying that these coincident gamma rays decay from the same state as the 1414 keV emission, and b) a 521 keV gamma ray in coincidence with the 1181 keV transition.

6.3.1 Shell Model Calculations and Proposed beta-delayed Level Scheme for $^{129}$Cd.

An understanding of the intrinsic configurations which form the excited states in $^{129}$Cd which are fed following the beta decay of $^{129}$Ag in the current work can be informed using state of the art shell model calculations. These have been performed for the low-lying states in $^{129}$Cd by H. Grawe (private communication) using the OXBASH shell model code. The shell model basis used in these calculations assumed a $^{78}$Ni core (50 neutrons and 28 protons). Excitations were allowed between the following proton single-particle orbitals $1f_{5/2}; 2p_{3/2}; 2p_{1/2}; 1g_{9/2}$; and the following neutron orbitals $1g_{7/2}; 2d_{5/2}; 2d_{3/2}; 3s_{1/2}$;
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.14: Gamma-ray spectra across the energy range of 0 - 2 MeV associated with the decay of (a) $^{129}$Ag and (b) $^{129}$Cd. c) A $^{129}$Cd subtracted $^{129}$Ag spectrum showing events associated with the primary decay of $^{129}$Ag only, which are assumed to be gamma-ray transitions from excited states in the $^{128,129}$Cd daughter nuclei. The $\beta - \gamma$ correlation times for each of these spectra was 0.760 s.
Figure 6.15: Gamma-ray spectra up to 4 MeV associated with the decay of (a) $^{129}$Ag and (b) $^{129}$Cd. c) A $^{129}$Cd subtracted $^{129}$Ag spectrum showing events associated with the primary decay of $^{129}$Ag only, which are assumed to be gamma-ray transitions from excited states in the $^{128,129}$Cd daughter nuclei. The $\beta - \gamma$ correlation times for each of these spectra was 0.760 s.

$1h_{1/2}$. The placement of these energy levels in the single particle shell model can be seen in figure 6.18.
Figure 6.16: The time difference spectrum of the beta ion implantations gated on \(^{129}\text{Ag}\), there are a total of approximately 20,000 \(^{129}\text{Ag}\) tagged decays in these data.

Figure 6.17: Partial level scheme showing excited states populated in \(^{128}\text{Cd}\) following the beta-delayed neutron decay of \(^{129}\text{Ag}\) deduced from the present work.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$I^\pi \rightarrow I^\pi$</th>
<th>Absolute $P_\gamma$</th>
<th>Relative $P_\gamma$ ($^{128}$Cd)</th>
<th>Transition intensity</th>
<th>$P_n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{128}$Cd</td>
<td>646  $2^+ \rightarrow 0^+$</td>
<td>0.159 (14)</td>
<td>1.00 (11)</td>
<td>0.159 (14)</td>
<td>$\geq 0.159$ (16)</td>
</tr>
<tr>
<td></td>
<td>785  $4^+ \rightarrow 2^+$</td>
<td>0.090 (8)</td>
<td>0.54 (8)</td>
<td>0.090 (8)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>440  -</td>
<td>0.049 (7)</td>
<td>0.30 (5)</td>
<td>0.049 (7)</td>
<td></td>
</tr>
<tr>
<td>$^{129}$Cd</td>
<td>205  -</td>
<td>0.48 (6)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>933  -</td>
<td>0.008 (9)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1111 -</td>
<td>0.050 (8)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1211 -</td>
<td>0.129 (12)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2014 -</td>
<td>0.050 (9)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2324 -</td>
<td>0.028 (8)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2605 -</td>
<td>0.011 (7)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3418 + 3420 -</td>
<td>0.032 (12)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3947 -</td>
<td>0.032 (5)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 6.6: Relative and absolute intensities of the gamma rays identified following the decay of $^{129}$Ag associated with the excited levels of $^{128,129}$Cd. The lower limit of the $P_n$ value of this nuclide is also included calculated from the measured intensity of the 646 keV emission in $^{128}$Cd.

The results of these large-basis shell model calculations are shown in figure 6.19, as SM calculations NA-14. These predict a spin/parity $I^\pi = \frac{11}{2}^-$ ground for $^{129}$Cd, based on the predominantly $h_{11/2}$ neutron hole configuration. Low-lying positive-parity excited states of $I^\pi = \frac{3}{2}^+, \frac{1}{2}^+$ and $\frac{5}{2}^+$ are predicted at excitation energies of 287 keV, 418 keV and 1272 keV above the $\frac{11}{2}^-$ ground state.

The ground state spin/parity of the 3-proton-hole, $N = 82$ mother nucleus $^{129}$Ag is expected to be $I^\pi = \frac{9}{2}^+$, based on a predominantly $g_{9/2}$ proton-hole configuration. Allowed Gamow-Teller $\beta^-$ decays from this ground state would then be expected to feed directly to spin/parity $\frac{7}{2}^+, \frac{9}{2}^+$ and $\frac{11}{2}^+$ states in the $^{129}$Cd daughter system. Such states would subsequently decay, through a preferred gamma-ray decay cascade dominated by E1 and M1 character decays. E1 transitions would also be expected from the $\frac{9}{2}^+$ and $\frac{11}{2}^+$ states proceeding by the predicted spin/parity $I^\pi = \frac{11}{2}^-$ $^{129}$Cd ground state. Other cascades will proceed along the positive parity states, until final arriving at the predicted low-energy $I^\pi = \frac{3}{2}^+$ and/or $\frac{1}{2}^+$ state, both of which would be hindered in their decay to the $\frac{11}{2}^-$ ground state due to the large multipolarity of the associated EM transition.

Taprogge et al., [143, 145] have reported the decay of a high–spin isomer, $I^\pi = \frac{21}{2}^+$ in $^{129}$Cd, which is interpreted as feeding the yrast $\frac{15}{2}^+$ state at 1587 keV and the yrast $\frac{13}{2}^-$ state at 1181 keV. The proposed M1/E2 decay from the $\frac{13}{2}^-$ state to the $\frac{11}{2}^-$ ground state is interpreted as the 1181 keV transition, which is observed in the current work.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.18: The ordering of the single particle energy levels, with respect to energy, reproduced from [144].

The partial level scheme presented for excited states in $^{129}$Cd$_{81}$ fed following the beta-decay of $^{129}$Ag$_{82}$ is presented in figure 6.20. The spin and parity assignments are made purely on the basis of comparison with the predictions of the shell model calculations listed as NA-14 in figure 6.19.
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.19: Left) Shell model prediction of the excited levels of $^{129}$Cd, calculated using the OXBASH code by H. Grawe, and, right) the decay scheme of the observed $^{21/2^+}$ isomeric state by Taprogge et al., [143].
Chapter 6. Radioactive decay studies of $^{120-124}$Rh and $^{129}$Ag

Figure 6.20: Deduced level scheme of the excited states of $^{129}$Cd following the decay of $^{129}$Ag, spins and parities are assigned according to the SM calculations seen in 6.19
Chapter 7

Summary, conclusions and future work

The work presented in this thesis concentrated on the application of gamma-ray spectrometric techniques to determine primary decay data from a range of nuclear decay scenarios. The multi-detector gamma-ray digital coincidence spectrometer, NANA, was designed, installed and commissioned at the National Physical Laboratory, UK. Its performance and design criteria were assessed relative to a GEANT4 simulation model and found to be consistent within these design parameters. It was demonstrated for use as a primary reference standard instrument for evaluating the activity concentration of $^{60}$Co using the gamma-ray coincidence method, incorporating corrections for angular correlation effects between successive gamma rays in a cascade. The resulting uncertainty in the determined measured activity concentrations using this method with NANA were determined to be better than the 1 part in 300 level.

The second part of the thesis focussed on the improved precision and accuracy of nuclear decay data associated with the medically relevant isotope $^{153}$Gd. This was been determined from two separate and independent datasets using highly calibrated HPGe detector set-up at the UK National Physical Laboratory. This work arrived at an absolute standardisation of $^{153}$Gd and provided new absolute $P_\gamma$ measurements of 7 principal gamma-ray emissions associated with its decay, determined to unprecedented precision. The data highlighting several discrepancies in the previously reported data. The absolute gamma-ray emission probability associated with the 97.4 keV emission from $^{153}$Gd
decay was used to input into the SIR IC model used at BIPM and the previous disagreement between the KCRV of the radionuclide and the model resolved, thereby validating the power of the model and the accuracy of the measurements.

Finally, spectroscopy of neutron-rich palladium isotopes was carried out following their production via projectile fission of a $^{238}$U primary beam at the RIBF facility RIKEN, Japan. Beta-delayed gamma-ray coincidence spectroscopy has been used to determine preliminary level schemes for the even-$N$ isotopes $^{120,122,124}$Pd and the relative population the first $2^+$ states in these nuclei used to determine limits on the beta-delayed neutron emission probabilities for the Rhodium isotopes, ranging from $A=120$ to $A=124$. A similar analysis has been used to determine a limit on the beta-delayed neutron emission probability for the $N=82$ isotope, $^{129}$Ag of $15.9(16)\%$.

Future work related to this thesis project includes the development of NANA for a fast timing, sub-nanosecond standards device and also the improvement and continuation of the $\gamma - \gamma$ counting technique to measure the activity of other industrially important radioisotopes, including $^{88}$Y, $^{94}$Nb and $^{134}$Cs.

With regards to the calculations of the absolute $P_n$ values for the isotopes around $^{132}$Sn, measurements are planned using the BRIKEN neutron detection system, which should enable direct measurement of the $P_n$ value for each of these nuclei directly. For more neutron-rich, shorter lived nuclei, the possibility of $\beta$-2n becomes more possible and probable, the spectrometric analyses described in this thesis may enable such measurements to be made with clarity.
Bibliography


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Appendix A

Reprints of publications associated with this thesis
Abstract. $^{153}\text{Gd}$ has widespread use, in non-destructive testing, as a line source in SPECT imaging and has been recently proposed as an in-vitro interstitial rotating shield brachytherapy (I-RSBT) source. In this work, the six most intense emissions in the de-excitation of the daughter nucleus $^{153}\text{Eu}$ have been measured, with an improved accuracy and precision to $\gamma$-ray emission intensities reported previously, via two characterised HPGe spectrometers. A specific absolute activity of 512.5 (25) kBq g$^{-1}$ was determined using the $4\pi$(LS)-$\gamma$ digital coincidence counting technique. This absolute activity was used to determine an absolute intensity for the 97.4 keV $\gamma$-ray emission of 30.15 (20) per 100 decays. The reported absolute emission intensity of this transition in this work has a relative difference of 4% from the currently recommended value.

1. Introduction

Gadolinium-153 is used in nuclear medicine as part of a line tracer in SPECT imaging [1–3], where attenuation corrections must be known on a patient-by-patient basis. Historically it has been used in bone absorptiometry [4–6], to study bone mineral density and to help diagnosis of osteoporosis. Recently, this radionuclide has been used for non-destructive testing of metal pipes [7] due to the strong dual emissions of the 97.4 keV gamma-ray emission and the K X-rays. Gadolinium-153 has been proposed as a replacement for $^{192}\text{Ir}$ in brachytherapy treatment. The lower energy photon emissions from $^{153}\text{Gd}$ are advantageous as they would require less shielding [8,9]. The common production mode of $^{153}\text{Gd}$ is by the neutron irradiation of a europium target, which is subsequently chemically separated to produce a radiochemically pure $^{153}\text{Gd}$ [10,11].

The ground state of $^{153}\text{Gd}$ decays by electron capture (EC) (100%) to $^{153}\text{Eu}$. The decay scheme and relative intensities evaluated by the Decay Data Evaluation Project (DDEP) is shown in Fig. 1. Two evaluations of the decay scheme of $^{153}\text{Gd}$ have been proposed in the new millennium [12,13], these show a discrepant number of excited levels being populated in $^{153}\text{Eu}$, and both discuss the need for new measurement due to strong disagreement in $P_\gamma$ values for weakly populated gamma-ray emissions in the daughter nuclei.

A BIPM [14] report detailing the methodology of the SIRIC [15] computer model for the $^{226}\text{Ra}$ equivalent activity measurements of the BIPM ionisation chambers comprising the “International Reference System”, identified $^{153}\text{Gd}$ as a possible radionuclide with inconsistent decay data. The modelled data was an outlier with a relative difference of 4.3%. Such a sizeable difference was not observed in radionuclides of similar photon decay profiles.

These observations are a cause of concern for a radionuclide proposed for in-vitro cancer treatment. It is therefore important to measure the intensity of the internal transitions within $^{153}\text{Eu}$ following the EC decay of $^{153}\text{Gd}$.

In this work the absolute activity of a solution of $^{153}\text{Gd}$ has been determined by $4\pi$(Liquid scintillation (LS))-\textit{\gamma} Digital Coincidence Counting (DCC) [16], which has subsequently been used to determine the $^{153}\text{Gd}$ absolute $\gamma$-ray emission intensities by high purity $\gamma$-ray spectrometry. Throughout this article, uncertainties are stated as standard uncertainties or combined standard uncertainties as defined in the Guide to the Expression of Uncertainty in Measurement (GUM) [17].

2. Experimental conditions

2.1. Sample preparation

An aqueous solution of radiochemically pure $^{153}\text{GdCl}_3$ in 0.5 M HCl with 50 $\mu$g g$^{-1}$ stable Gd carrier was used in this work. The sources for the $4\pi$(LS)-\textit{\gamma}$\text{DCC}$ technique were prepared by gravimetrically dispensing one drop of this solution (0.1 mg–0.3 mg) to twelve 20 mL glass liquid scintillation vials containing 0.1 mL of carrier and 10 mL of Ultima Gold AB scintillant. 1 g aliquots of the solution were gravimetrically dispensed into three British Standard (BS) [18] 2 mL flame sealed ampoules for measurement by gamma-ray spectrometry.

2.2. HPGe gamma-ray spectrometry

2.2.1. Gamma-ray spectrometers

Two ORTEC n-type High Purity Germanium (HPGe) $\gamma$-ray spectrometers were used to determine the absolute emission intensities of $^{153}\text{Gd}$. The detectors were identified...
as ‘BART’ and ‘LOKI’. The respective relative efficiency of ‘BART’ and ‘LOKI’ are 28% and 22%, with a resolution (FWHM) of 1.78 keV and 1.68 keV at 1332 keV. In addition, LOKI has a resolution of 676 eV at 122 keV.

Both detectors were contained in a 1.5 m × 1 m × 1 m lead shield, with walls 10 cm in depth to reduce the effects of external background radiation. To reduce the interference of the Pb fluorescence X-rays, degraders of 0.5 mm Cd and 0.7 mm Cu line the inside of the shield. The shields have aluminium optical bread boards mounted along the horizontal axis of the shield, which were used to mount kinematic mounting plates to which precision engineered sample holders could be attached. This enabled highly reproducible geometric sample positioning in front of the spectrometer (reproducibility uncertainty of 0.016%). The full-energy peak (FEP) detection efficiency of both spectrometers were determined for the matching geometry (to the samples) using a suite of sources traceable to primary standards of radioactivity. The sample-to-endcap distance and solid angle (Ω) for BART and LOKI were approximately 250 mm (Ω ~ 0.038 sr) and 290 mm (Ω ~ 0.045 sr) respectively. The distance between source and detector was used as to reduce the effects of dead-time and pulse pile-up. A further benefit to this extended sample-to-detector geometry is the reduction of True Coincidence Summing (TCS) to negligible levels due to the low Ω (relative to the standard uncertainty of the measurements). The FEP detection efficiency curve for both detectors were fitted using the procedure previously detailed in Collins et al. [19], which takes into consideration the correlations in standard uncertainties of the calibration points arising from, for example, the nuclear data, activity standardisation. The FEP detection efficiency and corresponding residuals for ‘LOKI’ are shown in Fig. 2. The FEP detection efficiency for the HPGe spectrometer ‘BART’ has been previously described by Collins et al. [19].

Spectra were acquired using a chain of analogue electronics (CANBERRA AFT Research amplifier 2025, Analogue-to-Digital Convertor 8715, AIM) connected to a computer running CANBERRA GENIE 2000 v2.1c software. The net peak area losses due to dead time and pulse pile-up were corrected using separate methods for each detector. For BART, the losses were corrected using the integrated pile-up rejection/live-time correction circuit with an additional empirically determined correction factor for those events not captured by the integrated circuit. LOKI used the well-established reference pulse method to correct the peak areas for both effects.

The neutron irradiation of a natural Europium target produces more than the desired radioactive isotope [10], it is therefore possible that the source could be contaminated with several other isotopes. In this study over 5 years, no contaminant peaks could be identified, except for emissions attributed to NORM, lead X-rays, or sum peaks. The first spectrum was acquired 7 days after preparation and was consistent with results taken much later in the study (~5 years), this increases confidence that the solution was clean of contamination.

Fourteen measurements were made of the three prepared ampoules. Acquisitions lasted between 86400 s and 250000 s. The integrated count rates and dead time were of the order 1600 cps and 6.7% respectively for the initial measurements. A measured spectrum of $^{153}$Gd can be seen in Fig. 3.
The spectra were analysed using the GENIE 2000 software, with the fit to each peak manually reviewed and modified where necessary using the GENIE Interactive Peak Fit application. The full-energy peak areas were corrected for background, decay during the measurement period, pulse pile-up and self-absorption.

2.3. Digital coincidence counting (DCC)

The 4π(LS)-γ coincidence system at the National Physical Laboratory (NPL) consists of a LS counter populated with two Hamamatsu R331-08 head-on type Photomultiplier Tubes (PMTs) operated in coincidence to provide a significant reduction in background count rates, due to the elimination of sporadic thermionic emissions from the photocathodes. The γ-channel utilised a 76 mm × 76 mm NaI(Tl) scintillation crystal, situated below the LS light guide in a lead shield.

The amplified pulses resulting from both detector systems were digitised and processed by the NPL Digital Coincidence Counting system, which has been described elsewhere [16,21,22]. This digital system handles the offline imposition of dead-times in both channels, selection of photon windows, imposition of appropriate delay/s and coincidence resolving times and provides data in a format suitable for the “Computer Discrimination” efficiency extrapolation method described by Smith [23,24].

Absolute activity determinations of EC radionuclides using coincidence techniques are complicated due to the low detection efficiency of the counter for X-rays and Auger electrons (X,e) from L, M and higher-shell capture events and following L, M conversions. The elevated non-detection of the initial capture (X,e) events leaves the counter open to register correlated events such as γ-rays and conversion electrons, requiring appropriate corrections. The usual technique of performing efficiency extrapolation to avoid calculation of these corrections, requires measurement of the count rates N_{LS}, N_γ, N_e in the 4πβ 4π(LS) channel, the gamma channel and the coincidence channel respectively, over a range of values of the LS-counter efficiency (estimated as Nc/N_c), facilitating extrapolation to unit efficiency and yielding an estimate of the source activity.

However, for EC nuclides, the magnitude of the effects of loss of (X,e) events is dependent on the ratios of probabilities for K and L-capture between the various branches, as coherently described by Funck and Nylandstedt-Larsen [25], and may produce a residual bias in the efficiency extrapolation result, even after extrapolation. To minimise this bias, a bi-dimensional efficiency extrapolation [26] was performed with the first gate set on the K X-rays between 41 keV and 49 keV (efficiency typically varied from 84% to 77%), and the second gate encompassing the 97 keV and 103 keV gamma rays (efficiency typically varied from 75% to 63%). When simultaneously extrapolating both channels to unit efficiency, the resulting estimate of the activity per unit mass was 512.2 (25) kBq g⁻¹. Gating on the K X-rays, 97 keV and 103 keV γ rays individually yielded biases of 0.13% and 0.3% respectively, with significantly larger variances.

3. Results and discussion

3.1. Relative P_γ measurements

In total, there are 14 prompt gamma-ray transitions observed in the decay of ¹⁵³Gd [12]. In this work, six of the γ-ray emissions have been identified and reported. The low-energy emissions (< 25 keV) of this decay scheme have not been observed due a combination of factors; low intensities, interference from X-rays and Compton scattering, reduced efficiency. The 54.1 keV has not been observed in any measurements. The possible 166.1 keV transition, which would connect the second 7/2⁺ level that is proposed to be populated by the decay of ¹⁵³Gd, is measured with inconsistency in these data, this is also true of the 68.2 keV transition linking the 7/2⁻ - 151.6 keV and the first 7/2⁺ 83.3 keV levels in ¹⁵³Eu.

The stronger populated 75.4 keV and 83.3 keV transitions are not reported, although identified in all spectra, due to inconsistent results. This inconsistency is due to the complexities of the Compton continuum in these energy regions.

The absolute emission intensity determined for the 97.4 keV γ-ray, using the absolute activity determined by
Table 1. The reported values of this work compared to the evaluated data. This work consistently is in disagreement with the most recent DDEP evaluation [12]. Absolute data is calculated using the DCC calculated value for the absolute intensity of the 97.4 keV transition.

<table>
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<th>Energy (keV)</th>
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</tr>
</thead>
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<tr>
<td></td>
<td>$I_\gamma$</td>
<td>$I_\gamma(E)/I_\gamma(97.4)$</td>
</tr>
<tr>
<td>69.6</td>
<td>2.38 (3)</td>
<td>7.89 (9)</td>
</tr>
<tr>
<td>89.4</td>
<td>0.096 (5)</td>
<td>0.320 (16)</td>
</tr>
<tr>
<td>97.4</td>
<td>30.15 (20)</td>
<td>100</td>
</tr>
<tr>
<td>103.1</td>
<td>22.33 (17)</td>
<td>74.06 (27)</td>
</tr>
<tr>
<td>151.6</td>
<td>0.0049 (6)</td>
<td>0.0157 (10)</td>
</tr>
<tr>
<td>172.8</td>
<td>0.0396 (6)</td>
<td>0.1313 (18)</td>
</tr>
</tbody>
</table>

Table 2. The uncertainty budget for the 97.4 keV $\gamma$-ray emission in $^{153}$Gd.

<table>
<thead>
<tr>
<th>Uncertainty component</th>
<th>Value /%</th>
</tr>
</thead>
<tbody>
<tr>
<td>FEP detection efficiency</td>
<td>0.38</td>
</tr>
<tr>
<td>Absolute activity</td>
<td>0.49</td>
</tr>
<tr>
<td>Peak fitting</td>
<td>0.15</td>
</tr>
<tr>
<td>Self-absorption</td>
<td>0.013</td>
</tr>
<tr>
<td>True coincidence summing</td>
<td>0.10</td>
</tr>
<tr>
<td>Gravimetric</td>
<td>0.02</td>
</tr>
<tr>
<td>Geometric reproducibility</td>
<td>0.016</td>
</tr>
<tr>
<td>Dead-time and pulse pile-up</td>
<td>0.18</td>
</tr>
<tr>
<td>Combined uncertainty</td>
<td>0.67</td>
</tr>
</tbody>
</table>

The DCC technique was 30.15 (20)% accurate. The absolute measurement was used to determine a new modelled equivalent activity for the BIPM ionisation chamber using SIRIC [15]. The modelled equivalent activity was found to be 368.4 (19) kBq, the key comparison reference value (KCRV) equivalent activity for this nuclide was 368.1 (17) kBq. This corresponds to a z-score of 0.26 and relative difference of 0.18%, previously the relative difference was 4.3%. The change in relative difference is comparable to the absolute intensity relative difference from this work with the previous evaluated value. This provides evidence that the absolute emission probabilities determined in this work are accurate.

4. Evaluation of absolute emission intensity of 97.4 keV emission

A new value of the absolute intensity for this nuclide can be calculated by using a weighted average of the reported values of the absolute intensities which are not reliant on previous $p_e$ measurements [28–32]. The work of Laurec [31] differs in that an electron spectrometer was used to measure the internal conversion coefficients, which were then used to calculate the absolute intensities. Other measurements [28–30] used the previously mentioned 4$\pi$β–γ liquid scintillation counting used in this work. The equilibrium method used by Laurec is valid only when the decay includes negligible ground state to ground state decays. A 4% branch in the decay of $^{153}$Gd does directly feed the ground state of $^{153}$Eu.

Several of the values of the absolute intensity for the 97.4 keV reported are not indeed experimentally calculated. Kafala [32] et al. are derived values which we suggest should be omitted from any new evaluation. The current evaluation reports the Geidelman value as described by Chechev [28].

The computed value of the experimental data including the potential outlier reported by Chechev, as identified using Chauvenet’s or Pierce’s criterion, gives a value of 30.13 (17)% in agreement below the critical $\chi^2$ of 2.57 for 95% confidence level. It is suggested that this value should be used in future work needing nuclear decay data for this nuclei.

5. Conclusion

Several of gamma emission probabilities for the decay of $^{153}$Gd have been measured to a greater accuracy than previously. These results do not agree with literature values. The measured absolute emission intensity of the 97.4 keV $\gamma$-ray, 30.15 (20)% is also in disagreement with the most recent evaluated data. Using this work and previous measurements a simple new evaluation for the absolute intensity results in a value of 30.13 (17)% around 4% greater than the evaluated. This increase in absolute emission probability accounts for the difference seen in the modelled and measured data reported for by the BIPM [14].

The extremely low emission intensity of the 166.2 keV $\gamma$-ray, if it indeed is seen in this dataset means that this emission cannot be reported. No evidence is seen that the 269.7 keV 2$^+_1$ + 2$^+_2$ level in $^{153}$Eu is populated.

Future work on this nuclide is worthwhile, as this evaluation has shown discrepancy. It is believed that a half-life measurement should be considered. This measurement may be of special note as it is the only nuclear data relied upon for the absolute emission probabilities reported in this work.

References

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Commissioning of the UK NAtional Nuclear Array

R. Shearmana,b, S.M. Collinsa, G. Lorussoa,b, M. Rudigierb, S.M. Judgea,b, S.J. Bella, Zs. Podolyakb, P.H. Regana,b

a National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK
b Department of Physics, University of Surrey, Guildford GU2 7XH, UK

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ABSTRACT

The NAtional Nuclear Array (NANA) is a LaBr3(Ce)-based coincidence gamma-ray spectrometer which can be used to identify, and enhance with respect to the background, signature gamma-ray emissions associated with particular radionuclide decays from a complex multi-component spectrum. Gamma-ray energy coincidence measurements using the NANA have been made using a digital data acquisition system based on CAEN V1751C 1 GHz digitizers. The improved time resolution offered by LaBr3(Ce) crystals compared to similar-sized solid state detectors can provide narrow time-correlated, gamma-ray energy coincidence matrices that can be interrogated to select discrete gamma-ray emissions associated with particular radionuclide decays. This paper provides an overview of the operational characteristics of the NANA spectrometer, including energy resolution and full-energy peak efficiency parameters, and provides an example of double and triple gamma-ray coincidence gating on decays associated with the nuclear fuel waste product 134Cs. The full-energy peak efficiency response of the spectrometer is compared to Monte Carlo GEANT4 simulations.

1. Introduction

The NAtional Nuclear Array, NANA, is a 12 element LaBr3(Ce) scintillation detector array, based at the UK’s National Physical Laboratory (NPL). It is designed for use in nuclear spectroscopy measurements by the detection of discrete energy, characteristic gamma rays emitted from excited nuclear states (Bucurescu et al., 2016; Lorusso et al., 2016; Regan et al., 2015a, 2015b). LaBr3(Ce) scintillator crystals are currently of interest to the radiation measurement community as a gamma-ray detector material which combines fast timing response and sufficient energy resolution (Browne et al., 2015; Bucurescu et al., 2016; Lalkovski et al., 2015; Regan et al., 2012, 2013, 2015a, 2015b; Régis et al., 2014a, 2014b; Roberts et al., 2014; Rudigier et al., 2015; Werner et al., 2016). Spectroscopic quality LaBr3(Ce) scintillator detectors are currently utilised in a number of nuclear structure and decay physics experiments worldwide; for example as part of the DESPEC-PATIMA collaboration (Lalkovski et al., 2015; Regan et al., 2012; Roberts et al., 2014) and the RoSPHERE gamma-ray spectrometer at IFIN-HH Bucharest (Bucurescu et al., 2016). In many of these configurations, the LaBr3(Ce) detectors are complemented with high-resolution hyper-pure germanium (HPGe) detectors in hybrid set-ups that provide superior timing triggers for gamma-ray cascades, allowing for the measurement of nuclear excited state lifetimes down to ~10 ps (Régis et al., 2014a, 2014b).

This paper presents the results of commissioning measurements for the NAtional Nuclear Array. Experimentally determined values for the full-energy peak detection efficiency and linearity of energy response are measured using well-characterised, mixed-isotope radioactive sources. These full-energy peak efficiency (FEP) measurements are compared to simulations of the array’s response which were made using the GEANT4 Monte Carlo code (Agostinelli et al., 2013). The application of the NANA for the selection of two and three gamma-ray decay cascades from a mixed radioactive source of 133Cs and 137Cs is also investigated, highlighting the potential use of this LaBr3(Ce) array for future radionuclide (waste) assay.

2. Dimensions, modelling and measurement of the response of the NAtional Nuclear Array

In order to maximise the functionality of the array for different applications, a GEANT4 simulation of the spectrometer was constructed within the NPTool framework (Matta et al., 2016). The GEANT4 simulation includes the front window, the can, the detector material and a borosilicate glass plate representing the photomultiplier tube. A simple representation of the frame has also been constructed within the model. Fig. 1 shows the constructed model defined within NPTool for the GEANT4 simulation of the NANA array.

The complete NANA design comprised twelve individual LaBr3(Ce)
detectors, with lead shielding to reduce Compton cross talk between detectors. Eight of the detectors are in a central ring with a 45° mutual angular spacing. This central ring is complemented by four additional detectors at backward angles relative to the central annulus. The details of the detector angles in the final NANA geometry are given in Table 1. The typical distance from the central (source) position to the front of the LaBr₃(Ce) detector face was 78 mm.

The energy resolution, timing and full-energy peak response of the 8 detectors in the central annulus were measured using a thin film vinyl source mounted on a bespoke 3D printed source holder. The source holder was designed to not shadow any detectors and to minimize any inhomogeneous efficiencies. Using this mixed source, full-energy-peak calibration and efficiency response curves were produced. The latter was used to validate the Monte Carlo simulations for the full-energy peak detection efficiency of the array.

Table 1
Detector angles for the twelve positions in the NAtional Nuclear Array. Note that the angles are with respect to the axes given in Fig. 1.

<table>
<thead>
<tr>
<th>Detector number</th>
<th>Polar Angle/degrees</th>
<th>Azimuthal angle/degrees</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>90</td>
<td>112.5</td>
</tr>
<tr>
<td>1</td>
<td>90</td>
<td>157.5</td>
</tr>
<tr>
<td>2</td>
<td>90</td>
<td>202.5</td>
</tr>
<tr>
<td>3</td>
<td>90</td>
<td>247.5</td>
</tr>
<tr>
<td>4</td>
<td>90</td>
<td>292.5</td>
</tr>
<tr>
<td>5</td>
<td>90</td>
<td>337.5</td>
</tr>
<tr>
<td>6</td>
<td>90</td>
<td>382.5</td>
</tr>
<tr>
<td>7</td>
<td>90</td>
<td>427.5</td>
</tr>
<tr>
<td>8</td>
<td>45</td>
<td>45</td>
</tr>
<tr>
<td>9</td>
<td>45</td>
<td>135</td>
</tr>
<tr>
<td>10</td>
<td>45</td>
<td>225</td>
</tr>
<tr>
<td>11</td>
<td>45</td>
<td>315</td>
</tr>
</tbody>
</table>

The LaBr₃(Ce) crystal is neither isotopically nor chemically pure, giving rise to an internal radiation background from the detector material (Lorusso et al., 2016). This internal radiation arises from the primordial isotope ¹³⁸La ($T_{1/2}=1.02\times10^5$ years (Bé et al., 2016)), present with an isotopic abundance of 0.09% (1%). Lanthanum-138 decays by β emission ($I_{β}=35%$) to the yrast spin/parity $I^\pi=2^+$ state in ¹³⁸Ce which is followed by the emission of a 788.7 keV gamma ray. The ¹³⁸La isotope can also decay by electron capture ($I_{ec}=65%$ branch) to the $I^\pi=2^+$ state in ¹³⁸Ba, followed by a 1435.8 keV gamma ray emission (Bé et al., 2016) and is accompanied by coincident La K-shell X-rays from the electron capture process. Additional internal radiation is present from the decay of the chemically similar ²²⁷Ac ($T_{1/2}=21.7$ years (Browne, 2001)). Actinium-227 decays by β emission to ²²⁷Th (and alpha emission to ²²³Fr) which subsequently decays by alpha emission through the actinium (4 n+3) natural decay series. The alpha decays of the subsequent (main) members of this decay chain (Browne, 2001) ($T_{1/2}(²²⁷Th)=18.697$ days (Browne, 2001); $T_{1/2}(²²⁵Ra)=11.4$ days (Browne, 2001); $T_{1/2}(²²¹Rn)=3.96$ s (Browne, 2001); $T_{1/2}(²²³Pq)=1.78$ ms (Browne, 2001)) have characteristic alpha particle decay energies in the range 6–7.5 MeV, but these are quenched by the response of the detector and PMT, and are observed in the detector response spectrum and gamma-ray equivalent energies of between 1.7 MeV and 2.6 MeV.

Fig. 2 shows the energy calibrated response of the sum of the 8 LaBr₃(Ce) detectors from the central ring of the NANA spectrometer, with no additional source present.

The variability in the linearity of the energy response across the different LaBr₃(Ce) detectors in NANA means that the energy calibration procedure of the LaBr₃(Ce) detectors can be more complex than for standard HPGe detectors (for which often a simple linear or quadratic calibration response function will provide good gain matching over an energy range 50 keV–2 MeV). The linearity of energy response for the eight detectors in the central ring of NANA was investigated using a thin VYNS mixed source with 13 prominent characteristic gamma emissions ranging from 60 keV from ²³⁴Am to the 1332 keV line of ⁶⁰Co. The acquired singles spectrum for one of the detectors can be seen in Fig. 3. Peaks were fitted to a Gaussian function with an nth-order polynomial background. The 1836 keV peak was not fitted due to the interference from the quenched internal alpha emissions in this region of the spectrum. The calibrated response of the detectors can be seen in Figs. 4 and 5. The calibration was achieved using a 4th order polynomial. Differences in the response for the high energy ⁶⁰Co emission (1332 keV) show less than 1% fluctuation across the eight detectors.

During the energy response calibration procedure, the degree of linearity for the detectors was observed to vary across the suite of detectors. The typical range in response from detector to detector can be seen from Fig. 4, where Detector 0 exhibits a greater linearity than Detector 3, whilst operating at the same voltage (1300 V) from the CAEN HV supply unit supplying independent HV to each detector.

The combined energy resolution response of the 8 detectors in the NANA central ring is shown in Fig. 6. In general, the measured full-width half maximum (FWHM) energy resolution for the detectors...
follows the expected Poisson $E^{1/2}$ dependence, with a measured value of 42 keV for the FWHM at 1332.5 keV.

### 2.2. Full-energy peak efficiency calibration of the NANA and comparison to Monte Carlo simulations

A full-energy peak detection efficiency measurement was determined using the same dataset as used for the energy calibration. The determination of the area under the full-energy peaks of interest in the spectra from the mixed source was complicated by the convoluted and multiple component background continuum in the spectra. Seven ‘well-fitted’ individual gamma-ray emissions from the VYNS mixed source were used in the final full-energy peak absolute detection efficiency measurement. These gamma ray transitions used in the fit were 59.5 keV ($^{241}$Am); 122.1 keV ($^{57}$Co); 165.9 keV ($^{139}$Ce); 391.7 keV ($^{113}$Sn); 661.7 keV ($^{137}$Cs); 834.8 keV ($^{54}$Mn); 1115.5 keV ($^{65}$Zn).

The spectrum of the VYNS mixed source acquired over 24 h shown in Fig. 3. Only the well-fitted peaks were used in the efficiency calculation shown in Fig. 7.

A Monte Carlo simulation of the NANA array was constructed for comparison, with the centres of the detector front faces placed a distance 78 mm from the source, replicating the geometry of the experimental calibration measurement. A mono-energetic isotropic source was simulated and placed in the centre of NANA. The energy of the simulated emitted gamma source was increased across an energy

### 2.2. Full-energy peak efficiency calibration of the NANA and comparison to Monte Carlo simulations

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The spectrum of the VYNS mixed source acquired over 24 h shown in Fig. 3. Only the well-fitted peaks were used in the efficiency calculation shown in Fig. 7.
range from 40 keV to 2000 keV. In contrast to the measured spectra, the only contaminant to the simulated peak was the lead K X-ray contribution in the low energy peaks. A simulation of the full 12 detector configuration was also performed, with the detectors also at 78 mm. This showed the expected 50% efficiency enhancement over the 8-element configuration shown in Fig. 7.

The data from the calibration source validated the Monte Carlo simulation for the full-energy peak efficiency. For the annular ring, a FEP efficiency of 8.93 (11) % was measured at 122 keV (57Co) and 1.876 (23)% at 1115.5 keV (65Zn). These compared well with the simulated FEP efficiencies for the eight in the annular ring of 9.82 (5)% and 1.889 (20)% respectively.

3. Example of Gamma-ray coincidence selection with NANA

The identification of fission products is crucial to nuclear security and public confidence in nuclear waste assay. The potential value of gamma-ray coincidence measurements in this field of measurement can be demonstrated using a test case of a mixed source of $^{134}$Cs/$^{137}$Cs (Regan et al., 2016). The isotopic ratio of this element can be used as a tracer, as the yield of the two radionuclides varies dramatically according to the production technique. Caesium-137 is a long-lived radionuclide ($T_{1/2} \approx 30$ years) emitting one characteristic 661.6 keV gamma ray from a metastable state in $^{137m}$Ba. A decay scheme of the shorter-lived isotope $^{134}$Cs, ($T_{1/2}=2.065(2)$ y), is shown in Fig. 9.

A mixed Cs isotope radioactive source was prepared at NPL from two chemically pure samples of $^{134}$Cs and $^{137}$Cs. The source had an activity of approximately 8 kBq, comprising ~4 kBq each from the two caesium isotopes at time of measurement. The ampoule containing the mixed source was placed at the centre of the array using a 3D printed source holder and data acquired for 14,400 s across the 8 detectors in the central ring of NANA. The acquired list-mode output was analysed using bespoke analysis software and npp (nuclear physics project), part of the NPTool software package (Matta et al., 2016). These software packages produced a ROOT output file which could then be interrogated and manipulated in offline analysis. From this, manipulations can be made to enhance the $^{134}$Cs decay emission lines and reduce the background produced by the internal radiation and ambient room radiation, as well as discrimination from the full-energy peak associated with the $^{137}$Cs decay and the Compton continuum.

To improve the peak-to-total in the singles spectra, a normalised fraction of the total internal background spectrum (shown in Fig. 2) was subtracted channel by channel from the acquired source spectra. The improvement in spectral clarity achieved using this background subtraction is demonstrated in Fig. 8, where the weakly populated emissions following the decay of the $^{134}$Cs, whose decay level scheme is shown in Fig. 9, are clearly evident.

3.1. Coincident measurements of caesium mixed isotope source

A coincidence time condition of $\Delta t=25$ ns was applied to the dataset. This offline sorting software condition groups temporally correlated events measured in different detectors which can be assumed to arise from the same radioactive decay event. This offline gating condition also allows for higher dimension coincidence spectra to be produced for correlated higher multiplicity events.

For measured gamma-ray multiplicity greater than 1 events, the $^{137}$Cs full-energy peak is essentially removed. In order to pass this logical gating condition, a decay event must be coupled to a random coincident event, either from the internal LaBr$_3$(Ce) detector background or chance. By contrast, for $^{134}$Cs cascade full-energy peaks, the number of counts in the spectrum is only reduced by the ratio of the detection efficiency for one and two events.

The spectrum in Fig. 10 was produced by plotting the correlated energy pairs produced in multiplicity $>1$ events on the $x$ and $y$ axis. The strongly populated cascades from the decay of $^{134}$Cs are clearly visible, while the $^{137}$Cs full energy peak is completely absent. Notably,
select $^{134}$Cs from $^{137}$Cs in a mixed source highlighted the power to separate and identify cascade emitting nuclides in complex matrices. This particular method of selecting weakly populated lines in a mixed matrix source may be particularly useful to nuclear fuel waste assay, where clean one nuclide mixes rarely exist.

This work was supported by the UK Science and Technology Facilities Council (STFC) by grant number ST/L005743/1 and the National Measurement Office. RS acknowledges support from the NDA-NNL PhD student bursary program. The authors are grateful to the engineers at STFC Daresbury Laboratory, UK and Prof. John Simpson for help in the mechanical design and construction of the NANA holding frame structure.

References


R. Shearman et al. 605 keV, 796 keV cascade associated with the $^{134}$Cs decay. No background subtraction has been applied to any of these gated spectra.

The gamma-ray coincidence multiplicity conditions applied to the mixed source $^{134}$Cs/$^{137}$Cs data show clean discrimination of the $^{137}$Cs. The gated spectra of the mixed source shows a reduction in the random and Compton background whilst the double gated spectrum (bottom) exhibits an improved peak to total and selects the 569 keV, 605 keV, 796 keV cascade associated with the $^{137}$Cs decay. No background subtraction has been applied to any of these gated spectra.

the associated ridges for Compton scattered $^{137}$Cs decay events summing to 662 keV can still be identified, as can the 1436 keV internal radiation coincidences with lanthanum K X-rays following the electron capture decay branch of the $^{138}$La component of the detector material.

Further separation of the $^{134}$Cs cascades can be achieved by applying energy gates on the full energy peaks. The Compton events with high multiplicity within the time window are removed from the spectrum by these gates. Selecting the triple gamma-ray cascade in the decay of $^{134}$Cs (569 keV, 796 keV and 605 keV), it can be seen that the Compton background is reduced (see Fig. 11) relative to the number of counts in the full-energy peak. A double energy gate of the 796 keV and 605 keV transitions shows very clean selection of only the 569 keV full-energy peak. This double gated spectrum has a peak to total ratio approaching 40% for the 569 keV peak, which represents a factor of 4 improvement over the ungated total projection spectra.

4. Summary and conclusions

The National Nuclear Array gamma-ray coincidence spectrometer has been commissioned at the National Physical Laboratory, UK. Gamma-ray energy calibrations using an NPL thin-film reference source show different linearity response across the detectors. Full-energy peak efficiency calibration measurements were in good agreement with Monte Carlo simulations performed in NPTool. The application of coincident measurements using the NANA elements to

![Fig. 11. The gamma-ray coincidence multiplicity conditions applied to the mixed source $^{134}$Cs/$^{137}$Cs data show clean discrimination of the $^{137}$Cs. The gated spectra of the mixed source shows a reduction in the random and Compton background whilst the double gated spectrum (bottom) exhibits an improved peak to total and selects the 569 keV, 605 keV, 796 keV cascade associated with the $^{137}$Cs decay. No background subtraction has been applied to any of these gated spectra.](image-url)
Investigation of $\gamma$-$\gamma$ coincidence counting using the National Nuclear Array (NANA) as a primary standard

S.M. Collins$^{a,b,*}$, R. Shearman$^{a,b}$, J.D. Keightley$^b$, P.H. Regan$^{a,b}$

$^a$ National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom
$^b$ Department of Physics, University of Surrey, Guildford GU2 7XH, United Kingdom

HIGHLIGHTS

- The multi-$\gamma$ ray detector National Nuclear Array has been investigated for use as a primary standard.
- Absolute standardisation of $^{60}$Co performed using the $\gamma$-$\gamma$ coincidence technique.
- Effect of angular correlations on the activity determined clearly observed.
- Corrections for angular correlation determined through Monte Carlo simulations.
- $\gamma$-$\gamma$ coincidence technique in agreement with the $4\pi$-(LS)-$\gamma$ Digital Coincidence Counting technique.

ABSTRACT

The National Physical Laboratory has recently been in the process of commissioning a multi-detector $\gamma$ ray array - the National Nuclear Array (NANA). In this study we have sought to exploit the NANA and the excellent timing characteristics of its intrinsic LaBr$_3$(Ce) scintillation detectors for use as a primary standardisation system. For this initial investigation, the absolute standardisation of $^{60}$Co performed using the $\gamma$-$\gamma$ coincidence technique using NANA and the result compared to the established $4\pi$-(LS)-$\gamma$ Digital Coincidence Counting (DCC) system. The effect of the angular correlation of the stretched E2 transitions emitted from the $4^+ \rightarrow 2^+ \rightarrow 0$ states of $^{60}$Ni on the activity determined by NANA was observed between the pairs of detectors. Corrections for these angular correlations were derived through Monte Carlo simulations. An activity per unit mass by NANA of 330.8 (10) kBq g$^{-1}$ for the $^{60}$Co solution was determined. There was no significant statistical difference between the results of NANA and the $4\pi$-(LS)-$\gamma$ DCC, with a relative difference of 0.04% observed. This study shows that NANA can be used as a primary standard.

1. Introduction

The National Physical Laboratory (NPL) has recently been in the process of installing and commissioning a multi-detector $\gamma$ ray spectrometer array, the National Nuclear Array (NANA) (Regan et al., 2015; Lorusso et al., 2016; Shearman et al., in press). In its current configuration, NANA will be capable of hosting 12 LaBr$_3$(Ce) scintillation detectors, coupled to fast-timing photomultipliers, as its intrinsic $\gamma$-ray spectrometers. These detectors have been evaluated in numerous spectroscopic studies and have been shown to provide excellent timing characteristics, capable of sub-nanosecond timing response (Regis et al., 2012; 2013) allowing clean correlations between discrete $\gamma$-rays in mutual coincidence.

The primary purpose of this array was to provide NPL with the capability for nuclear structure and decay data measurements. During its inception the coincidence counting capabilities of NANA were identified as giving it the potential for the primary standardisation of radionuclides which emit two or more $\gamma$ rays in a coincident cascade (e.g. $^{60}$Co), using the $\gamma$-$\gamma$ coincidence counting technique (Pommé, 2007). If NANA could be used to standardise the activity of a radionuclide by a primary technique then NANA (or a similar configuration) could also be used for determining absolute $\gamma$-ray emission intensities directly from the measured sample, which had either been gravimetrically prepared or produced as a result of proton or neutron activation. An obvious advantage can be identified for radionuclide species which have half-lives that are too short to allow the conversion of an irradiated target to a suitable matrices for measurement by other primary techniques. In the current work, we have carried out a study to...
investigate the capability of NANA as a primary standard for the activity per of $^{60}$Co.

For this initial study, $^{60}$Co was selected as it has an ideal decay scheme for coincidence counting with its two intense coincident high energy $\gamma$ rays (I$_{\beta}$ = 100%) following its $\beta$- decay and negligible internal conversion. The decay scheme for $^{60}$Co is shown in Fig. 1. (Helmer, 1998); note that the angular correlations between these successive gamma ray emissions pose additional considerations for the $\gamma$-$\gamma$ coincidence method using NANA.

All uncertainties stated in this work are standard uncertainties or combined standard uncertainties as defined in the Guide to the Expression of Uncertainty in Measurement (GUM) (BIPM, 2008).

2. $\gamma$-ray energy coincidence counting formulae for $^{60}$Co

The photon-photon coincidence counting technique has been widely used for the activity standardisation of $^{125}$I, $^{197}$Hg, $^{129}$I and other low-energy $\gamma$-ray emitting radionuclides by detection of X-ray-$\gamma$-ray coincidences (Eldridge and Crowther, 1964; Taylor, 1967; Schrader and Walz, 1987; Pommé et al., 2005; Schrader, 2006). The technique is particularly effective for low energy photon emitting radionuclides as only the full-energy peaks of the X-rays (following EC and ICC) and discrete $\gamma$ rays contribute to the observed photon spectra.

Unfortunately, there are difficulties in the direct generalisation of the photon-photon coincidence formulae to $^{60}$Co. The main difference between the techniques for $^{60}$Co compared to lower energy $\gamma$-ray emitters is that of the Compton scattering contribution. This contribution results in the total detection efficiency differing from that of the full-energy peak detection efficiency. For $^{60}$Co the number of these unknown probabilities is twice as many as for the case of, for example, $^{125}$I. Therefore, the formulas for the coincidence count rate in two detectors have to be modified.

A solution to this complication in the technique for high energy $\gamma$ ray emitters was derived by Volkovitsky and Naudus (2009) that would allow the absolute standardisation of $^{60}$Co using two NaI(Tl) detectors. Their method has been applied in this work by using multiple pairs of the LaBr$_3$(Ce) detectors in the central ring. Their solution is presented here for convenience.

The count rate of the full-energy peak $j$ ($j = 1, 2$) in detector $i$ ($i = 1, 2$) can be described as:

$$N_i^{(2, j)} = N_0 \epsilon_i^{(1, j)} (1 - \epsilon_i^{(2, 0)})$$

(1)

where

- $N_0$ the disintegration rate of $^{60}$Co into the channel with two coincident $\gamma$ rays;
- $N_i^{(2, j)}$ the count rate in the detector $i$ of events in the full-energy peak of the $\gamma$ ray $j$;
- $\epsilon_i^{(1, j)}$ the probability of $\gamma$ ray $j$ detection in the full-energy peak by detector $i$, this is equal to the full-peak energy efficiency multiplied by the $\gamma$ ray $j$ emission probability;
- $\epsilon_i^{(2, 0)}$ the total probability of detecting $\gamma$ ray $j$, including Compton scattered $\gamma$ rays, by detector $i$, this is equal to the total efficiency multiplied by the emission probability for $\gamma$ ray $j$;
- $(1 - \epsilon_i^{(2, 0)})$ the probability that $\gamma$ ray $j$ is not detected by the detector $i$.

In each detector $i$ there is a sum-peak of the 1173.2 keV and 1332.5 keV $\gamma$ rays and two individual full-energy peaks of coincidence events for the two $\gamma$ rays in both detectors, where the energies deposited in each detector $i$ are in the single full-energy peak areas, these can be described by:

$$N_i^{(1,2)} = N_0 \epsilon_i^{(1, j)} \epsilon_2^{(2, j)}$$

(3)

$$N_i^{(1,2)} = N_0 \epsilon_i^{(1, j)} \epsilon_2^{(2, j)}$$

(4)

where

- $N_i^{(1,2)}$ The count rate of coincident events where the first $\gamma$ ray is detected by the first detector and the second $\gamma$ ray is detected by the second detector;
- $N_i^{(2,1)}$ The count rate of coincident events where the second $\gamma$ ray is detected by the first detector and the first $\gamma$ ray is detected by the second detector.

These coincidence events for $^{60}$Co in NANA can be seen in the 3D coincidence spectrum presented in Fig. 2.

The count rate of coincident events in detector 1, $N_i^{(1, 2)}$, where the first $\gamma$ ray is detected in the full-energy peak and the second $\gamma$ ray is detected as a full-energy peak or Compton scattered $\gamma$ ray. This can be written as:

$$N_i^{(1, 2)} = N_0 \epsilon_i^{(1, j)} \epsilon_2^{(2, 0)}$$

(6)

Variations of Eq. (6) can be written for the remaining three coincident peaks, $N_i^{(2, 1)}$, $N_i^{(1, 1)}$, $N_i^{(2, 2)}$.

The difference between the spectrum of all events in the full-energy peak and the coincident spectrum in the same region in detector $i$ is due to the anti-coincident events (i.e. the events when the second $\gamma$ ray is

Fig. 1. Decay scheme of $^{60}$Co (Helmer, 1998).

Fig. 2. 3D coincidence energy spectrum collected by NANA for source A. The 1173.2 keV, 1332.5 keV full-energy peaks and Compton ‘tails’ for the two coincident $\gamma$ rays can be seen.
For detector 1 this difference, \( N^{(1,0)}_{\text{int}} \), is given by:

\[
N^{(1,0)}_{\text{int}} = \sum_{i=1}^{2} N^{i(1,0)}_{\text{int}} = N_{\gamma}^{(1,0)}(1 - \epsilon^{(1,0,0)}_{\gamma}) - N_{\beta}^{(1,0)}(1 - \epsilon^{(1,0,0)}_{\beta})
\]

where \((1 - \epsilon^{(1,0,0)}_{\gamma})\) is the probability the second \( \gamma \) ray is not detected by either detector. Again, this equation can be written for the three additional anti-coincidence channels.

These can be used to determine the ratio of experimental observables:

\[
\frac{1 - \epsilon^{(1,0,0)}_{\gamma}}{1 - \epsilon^{(1,0,0)}_{\beta}} \equiv \frac{N^{(1,0)}_{\text{int}}}{N^{(2,0)}_{\text{int}}} = R^{(1,0)}_{\text{int}}
\]

and similar ratios for \( R^{(1,0)}_{\text{int}} = N^{(1,0)}_{\text{int}}/N^{(1,0)}_{\text{int}} \), \( R^{(2,0)}_{\text{int}} = N^{(2,0)}_{\text{int}}/N^{(2,0)}_{\text{int}} \), and \( R^{(2,0)}_{\text{int}} = N^{(2,0)}_{\text{int}}/N^{(2,0)}_{\text{int}} \).

From Eq. (8) the total detection probabilities \( \epsilon^{(1,0,0)}_{\gamma} \) (\( i = 1, 2; j = 1, 2 \)) can be found, such as:

\[
\epsilon^{(1,0,0)}_{\gamma} = \left( \frac{R^{(1,0)}_{\text{int}} - 1}{R^{(2,0)}_{\text{int}} + R^{(2,0)}_{\text{int}} - 1} \right)
\]

With Eq. (9) the full-energy peak detection probability of the first \( \gamma \) ray in the first detector, \( \epsilon^{(1,0,0)}_{\gamma} \), can be determined from Eq. (6).

\[
\epsilon^{(1,0,0)}_{\gamma} = \frac{N^{(1,0)}_{\text{tot}}}{N^{(2,0)}_{\text{tot}}}
\]

and the full-energy peak detection probabilities for the remaining three combinations of \( \gamma \) rays and detectors can be written in a similar manner.

The disintegration rate \( N_{\beta} \) can now be calculated in two ways:

\[
N_{\beta} = \frac{N^{(1,0)}_{\text{tot}} N^{(2,0)}_{\text{tot}}}{\epsilon^{(1,0,0)}_{\gamma} \epsilon^{(2,0,0)}_{\beta}}
\]

(11)

\[
N_{\beta} = \frac{N^{(1,0)}_{\text{tot}} N^{(2,0)}_{\text{tot}}}{\epsilon^{(2,0,0)}_{\gamma} \epsilon^{(2,0,0)}_{\beta}}
\]

(12)

where the detection probabilities are provided by Eq. (9) and Eq. (10).

Eq. (11) and Eq. (12) can be rewritten in terms of observables, in a similar manner to Eq. (8):

\[
N_{\beta}^{(1,0)} = \frac{(N^{(2,0)}_{N^{(1,0)}} - N^{(2,0)}_{N^{(1,0)}})(N^{(1,0)}_{N^{(1,0)}} - N^{(1,0)}_{N^{(1,0)}})}{(N^{(2,0)}_{N^{(1,0)}} - N^{(2,0)}_{N^{(1,0)}})(N^{(1,0)}_{N^{(1,0)}} - N^{(1,0)}_{N^{(1,0)}})}
\]

(13)

\[
N_{\beta}^{(2,0)} = \frac{(N^{(2,0)}_{N^{(2,0)}} - N^{(2,0)}_{N^{(2,0)}})(N^{(2,0)}_{N^{(1,0)}} - N^{(2,0)}_{N^{(1,0)}})}{(N^{(2,0)}_{N^{(2,0)}} - N^{(2,0)}_{N^{(2,0)}})(N^{(2,0)}_{N^{(1,0)}} - N^{(2,0)}_{N^{(1,0)}})}
\]

(14)

Finally, the symmetric combination of Eq. (13) and Eq. (14) gives the disintegration rate of \( ^{60}\text{Co} \):

\[
N_{\beta} = \sqrt{N_{\beta}^{(1,0)} N_{\beta}^{(2,0)}}
\]

(15)

Eq. (15) has been derived under the assumption that both \( \gamma \) rays from the \( \beta^- \) decay of \( ^{60}\text{Co} \) are emitted independently. However, both the 1173 keV and 1322 keV \( \gamma \) ray emissions are stretched E2 (quadrupole) transitions, emitted from the first spin/parity \( 4^+ \) and \( 2^+ \) excited states of \( ^{60}\text{Ni} \) respectively. Therefore, the correlation function \( W(\theta) \) between the two \( \gamma \) rays is an even function of \( \cos \theta \), where \( \theta \) is the angle between the two \( \gamma \) rays, defined by:

\[
W(\theta) = 1 + a_{2} P_{2}(\cos \theta) + a_{4} P_{4}(\cos \theta)
\]

(16)

where \( P_{n}(\cos \theta) \) is a Legendre polynomial of \( n \)-th order, where \( a_{2} = 0.1020 \) and \( a_{4} = 0.0091 \) (Brady and Deutsch, 1950). The angular correlation of the two \( \gamma \) rays as a function of \( \theta \) is shown in Fig. 3, where \( W(\theta) \) is given normalised to the angle 90°.

Volkovitsky and Naudus (2009) assumed that if the source was located in the centre of symmetry of two identical detectors then the equations were still valid as the angular correlation effects were cancelled. In the case of NANA, the effect of the angular correlation is not cancelled due to finite angular coverage between the pairs of detectors. Therefore Eq. (15) only describes the angular correlated activity, \( N_{\beta} \), for a detector pair at a given angle:

\[
N_{\beta}(\theta) = \sqrt{N_{\beta}^{(1,0)} N_{\beta}^{(2,0)}}
\]

(17)

Therefore, incorporating the angular correlation function to Eq. (17) produces the angular independent value for \( N_{\beta} \):

\[
N_{\beta} = W(\theta) N_{\beta}(\theta)
\]

(18)

As the detectors are not collimated, to give high-precision defined angles between the detectors, the effect of angular smearing can be significant. Therefore, corrections have been determined using Monte Carlo simulations to determine the effective angular correlation, \( W(\theta) \) (Kim et al., 2002). Finally, Eq. (18) becomes:

\[
N_{\beta}(\theta) = W(\theta) N_{\beta}(\theta)
\]

(19)

3. Experimental method

3.1. Sample preparation

A radiochemically pure aqueous solution of \( ^{60}\text{Co} \) in 0.1 M HCl with 100 µg g\(^{-1}\) of stable Co, was used to prepare the samples to be measured by NANA and the 4n(LS)\(\gamma \) DCC system. The solution had been standardised using the NPL secondary standard re-entrant ionisation chamber (SSIC) (Baker et al., 2014), which had been previously calibrated for \( ^{60}\text{Co} \) in a manner traceable to national standards of radioactivity. An activity per unit mass of 330.9 kBq g\(^{-1}\) ± 1.1 kBq g\(^{-1}\) at a reference time of 2016-03-10 12:00 UTC had been determined. No \( \gamma \)-ray emitting impurities had been previously detected in the solution.

Aliquots of mass 0.10 g, 0.08 g, 0.06 g, 0.04 g and 0.02 g of the \( ^{60}\text{Co} \) solution were dispensed to the centre of five VYNS mounts, identified as sources A-E respectively. These VYNS sources were transferred to a fume cupboard and the solution left to dry in ambient temperature. Once the solution had dried the sources were sealed with an additional layer of VYNS film to prevent any losses of active material.

3.2. National Nuclear Array (NANA)

NANA has the capacity to hold up to 12 LaBr3(Ce) detectors; in the current work only eight detectors were in place. These were held in the central ring of eight symmetrically placed detector mounts (see Fig. 4). The front face of each detector was located 78 mm from the centre of the ring, resulting in solid angle, \( \Omega \), of 0.31 sr for each detector (assuming a point source located at the centre of the ring). The eight detectors were positioned at 45° increments around the central ring. The VYNS source was held in the centre of the ring of detectors using a
bespoke ‘3D printed’ source holder.

Each LaBr₃(Ce) detector comprised a 2″ × 1.5″ crystal coupled to a Hamamatsu R9779 photomultiplier tube. The pulses from the energy output of the PMT were sent directly to a CAEN V1751, 1 GHz digitiser, which provides an independent, synchronised time-stamped energy signal for each input channel (Regan et al., 2015). This digitised nanosecond timing allows the event-by-event coincidence information between γ rays emitted from a discrete cascade to be temporally correlated. In turn, logical gated conditions can be set on particular γ ray energies which define an energy-level decay cascades for a radionuclide. This coincidence gating is applied off-line in software data analysis.

The detectors had not been accurately gain matched during the initial installation. Therefore, to enhance the off-line sorting of the coincidence events between different γ-ray detectors within the array the energy calibration for each detector was determined. This was performed using a mixed source containing radionuclides with γ-ray emissions covering the energy range from 60 keV to 1836 keV.

4. Results and discussion

The five sources (A–E) were each measured for periods of 86400 s. For each measurement the net peak areas for the full-energy peaks of the 1173.2 keV and 1332.5 keV γ-ray emissions were determined from the histogram outputs to find $N_{\gamma}^{1\alpha}(\phi)$, $N_{\gamma}^{2\beta}(\phi)$ for each detector. Each net peak area was determined using a cubic function to approximate the underlying continuum and a Gaussian function to fit the full-energy peak.

The list mode outputs were sorted and analysed using bespoke routines developed at NPL, with the coincidence gates set for the 1173.2 keV and 1332.5 keV γ-ray full-energy pulses. The timing window for the coincidences was set at $\Delta = 25$ ns. A time window of $\Delta = 5$ ns would normally be considered appropriate for $^{60}$Co. While for the majority of detector pairs this would have been suitable in a detector pairs the coincidence time distributions went up to 25 ns. The potential cause was identified as the differing cable lengths, where those detectors with longer cables connecting their output to the CAEN digitiser had longer time distributions. The sorted data was compiled into an output file for analysis using the CERN-developed data sorting software package ROOT (Brun and Rademakers, 1997).

For each measurement, $N_{\gamma}^{1\alpha}(\phi)$, $N_{\gamma}^{2\beta}(\phi)$, $N_{\gamma}^{1\beta}(\phi)$, $N_{\gamma}^{2\alpha}(\phi)$, $N_{\gamma}^{1\beta}(\phi)$, $N_{\gamma}^{2\beta}(\phi)$ count rates were determined from the coincidence spectra for each detector pair in ROOT (see Fig. 5), these count rates were background corrected for the coincidences determined from the background measurement. In a number of the coincident spectra, a small amount of ‘breakthrough’ can be observed in the region of the 1173 keV full-energy peak (see Fig. 5b). This is a result of the Compton scattering of the incident 1332.5 keV γ ray depositing energy within the 1173 keV energy gate. These Compton events in detector 1 can result in an incident 1173.2 keV γ ray in detector 2 being incorrectly observed as a ‘real’ coincident event with a 1173.2 keV γ ray depositing its full-energy in detector 1. Simulations indicate that this may result in a maximum 5 % increase in the total counts in the coincident 1173.2 keV Compton continuum terms $N_{\gamma}^{1\beta}(\phi)$ and $N_{\gamma}^{2\beta}(\phi)$. In this work, the sum of all the Compton continuum terms had an approximate 2 % effect on the determined activity. Propagating through Eqs. (13,14), the effect of this ‘breakthrough’ on the determined activity was determined to be no greater than 0.05 %.

For each detector as $\theta = 0$, the coincident count rates and the singles full-energy count rates were used to determine the activity for each detector pair in the ring using Eqs. (13)-(15). The activity was corrected for mass and radioactive decay. This gave seven activity determinations at $\theta = 45$, $\theta = 90$, $\theta = 135$, $\theta = 180$, $\theta = 225$, $\theta = 270$ and $\theta = 315$ for each starting detector sequence, giving a total of eight activity determinations for each angle. This provided a total of 56 coincidence activity determinations per measurement. Analysis of these 56 activity determinations across the five measurements performed showed a consistent bias is the results that included either Detector 3 or 5 in one of the detector pairs. The gain of these two detectors varied significantly from the remaining detectors in the array. This indicated that these two detectors were not matched well with the remaining detectors, therefore all activity determinations that included these two detectors were removed from the analysis. This reduced the total activity determinations per measurement to 30.

An observed activity per unit mass for each angle, $\tilde{\lambda}_{\phi}(\theta)$ and standard uncertainty was determined from the arithmetic mean of the observed activity and standard deviation. The arithmetic mean activities and standard uncertainties determined at each angle for source A are shown in Fig. 6.

In Fig. 7 the normalised activity per unit mass at each incident angle is shown against the $w(180)/w(\theta)$ of the angular correlation distribution of the $4^+ \rightarrow 2^+ \rightarrow 0^+$ cascade of stretched E2 transitions following the decay $^{60}$Co to $^{60}$Ni. While the normalised activities follow the general path of the distribution, from Fig. 7 it can be observed that they are not in good agreement. This disagreement between the two is due to the effect of angular smearing and required the determination of the effective angular correlation corrections.

The effective angular correlation, $\tilde{\lambda}(\theta)$, between two detector of finite angular resolution and finite size can be shown as (Rose, 1953; Camp and Van Lehn, 1969):

$$\tilde{\lambda}(\theta) = \sum_{\text{even } k} a_{lk} P_k(\cos \theta) Q_k(1)Q_k(2)$$

where $\theta$ is the angle between the two axes of the detectors, $P_k$ is the Legendre polynomial and $a_{lk}$ is the angular correlation coefficient. $Q_k(1)$ is the attenuation correction factor for $y_i$ incident on detector $i$ and $Q_k(2)$ is the attenuation correction factor for $y_l$ incident on the other detector. $Q_k$ is given as:
\[ Q(i) = J_\beta(i)/J_\beta(i) \]

where

\[ J_\beta(i) = \int_0^{\beta_{\text{max}}} d\beta \sin(\beta)P_2(\cos \beta)\varepsilon_i(\beta) \]  

(22)

Fig. 5. ROOT coincidence spectra for the a) 1332.5 keV and b) 1173.2 keV coincidence gates between NANA detectors 0 and 1 for source A. ‘Breakthrough’ of the coincident counts at the 1173.2 keV can be seen in b). Coincident events with the La X-ray and Pb X-rays can also be seen in both coincident spectra.

Fig. 6. The mean decay corrected activity per unit mass determined for source A as a function of the angle difference from the starting detector at \( \theta = 0 \), in 45° increments. The uncertainty of each value was determined from the standard deviation of all determinations at a given angle from the starting detector in a given sequence. These activity per unit mass values do not include the correction for the effect of angular correlation.

\[ A_0/\text{MBq g}^{-1} \]

where \( \beta \) is the incident angle of the \( \gamma \) ray with respect to the detector axis, \( \beta_{\text{max}} \) is the maximum incident angle of the \( \gamma \)-ray, and \( \varepsilon_i(\beta) \) is the total absorption efficiency of the \( i \)-th \( \gamma \)-ray incident on the detector with an incident angle \( \beta \).

Fig. 7. The activity per unit mass, prior to correction for angular correlations, at each \( \theta \) normalised to the activity per unit mass at \( \theta = 180^\circ \) for source A. The \( \varepsilon(180)/\varepsilon(\theta) \) of the \( 4^+ \rightarrow 2^+ \rightarrow 0^+ \) cascade of pure E2 transitions is shown as the dashed line. The \( \varepsilon(180)/\varepsilon(\theta) \) determined from the Monte Carlo simulations are shown as the dotted line. It can be seen that the normalised activity per unit mass results are in agreement with the effective angular correlation values.
The Monte Carlo simulation method of Kim et al. (2002) was used in conjunction with the Monte Carlo model of NANA that has been developed at NPL. (Shearman et al., in press). From this, the $\epsilon(\beta)$ for the detectors were estimated for the 1173.2 keV and 1332.5 keV $\gamma$ rays and were used to determine $Q_2$ and $Q_4$ for each $\gamma$ ray, shown in Table 1.

These values were applied to Eq. (20) to determine $w(\theta)$, the results of which can be seen in Fig. 7 as $w(180)/w(0)$. This effective angular correlation distribution shows an improvement in the agreement with the observed activities at each angle, which can be clearly seen at $\theta = 90$. The arithmetic mean activity per unit mass at each incident angle were corrected using Eq. (19), with their combined uncertainty at each angle comprised of the standard deviation and uncertainty of the angular correlation correction at the given angle. The resulting activities per unit mass and uncertainty at each angle for source A are shown in Fig. 8.

The activity per unit mass for each source and the standard uncertainty was determined from the weighted mean of the activities per unit mass at each angle, the result for each source (A to E) can be seen in Fig. 9, as well as the result of the $4\pi(\text{LS})-\gamma$ DCC. The results for the standardisation of the $^{60}\text{Co}$ solution by NANA and $4\pi(\text{LS})-\gamma$ DCC are shown in Table 2. There was no significant statistical difference between the activity per unit mass of the $^{60}\text{Co}$ solution by the $\gamma-\gamma$ coincidence technique using NANA and the well-established $4\pi(\text{LS})-\gamma$ DCC technique, with a relative difference of 0.04% between the two methods.

From this study, NANA can potentially be successfully employed to standardise radionuclides with simple decay schemes with two intense $\gamma$-ray emissions in coincidence with good accuracy and precision. The future of this work will be to expand the scope of the radionuclides to more complex decay schemes, such as $^{134}\text{Cs}$, to investigate the capabilities of NANA as a primary standardisation systems further.

The observation of the angular correlations between the coincident $\gamma$ rays at multiple angles is of particular interest in this method. The capability to assess the angular distribution of the two coincident $\gamma$ rays and compare against the Monte Carlo derived corrections can be considered a strength of the NANA.

In this work the uncertainties have been derived in a simplistic fashion as no account of the correlations between the detectors or uncertainties in the fitting methods. Taking this method forward, these uncertainties and their propagation will need to be thoroughly investigated.

5. Conclusion

A short study of the capability of the NANA for the primary standardisation of activity using the $\gamma-\gamma$ coincidence technique has been performed at NPL. The activity per unit mass of 330.8 (10) kBq g$^{-1}$ for a solution of $^{60}\text{Co}$ has been determined. This is in statistical agreement with the established $4\pi(\text{LS})-\gamma$ DCC system with a relative difference of 0.04% between the two systems.

The effect of the angular correlation between the $4^+ \rightarrow 2^+ \rightarrow 0^+$ cascade of the stretched E2 transitions from $^{60}\text{Co}$ have been observed. An effective angular correlation distribution has been determined through Monte Carlo simulations, which showed good agreement with the observed angular distribution.

The successful results in this work provided evidence that NANA can be used for the standardisation of radionuclide with further work to extend the method to more complex decay schemes, such as $^{134}\text{Cs}$, planned for the future.

Acknowledgements

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Table 1

<table>
<thead>
<tr>
<th>Energy /keV</th>
<th>$Q_2$</th>
<th>$Q_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173.2</td>
<td>0.9731</td>
<td>0.9125</td>
</tr>
<tr>
<td>1332.5</td>
<td>0.9734</td>
<td>0.9134</td>
</tr>
</tbody>
</table>

Table 2

The results of the standardisation of the activity per unit mass determined by $\gamma-\gamma$ coincidence technique using NANA. The activity per unit mass of the same stock solution determined by the NPL $4\pi(\text{LS})-\gamma$ Digital Coincidence System is shown as a comparison.

<table>
<thead>
<tr>
<th>Standardisation technique</th>
<th>$A_0$ /kBq g$^{-1}$</th>
<th>$u(A_0)$ /kBq g$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4\pi(\text{LS})-\gamma$ DCC</td>
<td>330.92 ± 0.86</td>
<td>± 0.86</td>
</tr>
</tbody>
</table>

Fig. 8. The activity per unit mass at each angle for source A after the application of the effective angular correlation correction. The standard uncertainty of the activity at each angle is comprised of the standard deviation and uncertainty of the angular correlation correction combined in quadrature. The solid and dotted lines represent the weighted mean activity per unit mass and standard uncertainty determined for the source.

Fig. 9. The activity per unit mass and standard uncertainty for source A-E determined by the $\gamma-\gamma$ coincidence technique using NANA and the $4\pi(\text{LS})-\gamma$ DCC method.
References


Abstract: The evaluated spectroscopic data are presented for 12 known nuclides with $A=217$ (Tl, Bi, Po, At, Rn, Fr, Ra, Ac, Th, Pa, U). For $^{217}$Tl, $^{217}$Pb, $^{217}$Pa, and $^{217}$U nuclei, only information on the ground state is available. Levels in $^{217}$Bi are known only from isomer decay following fragmentation reaction and those in $^{217}$At and $^{217}$Rn only from the $\alpha$ decay of $^{221}$Fr and $^{221}$Ra, respectively. High spin levels in $^{217}$Ra are mainly from 1987SuZY and 2011MuZZ which are a lab report and thesis, respectively. Due to differences between these studies, further experimental study is needed to firmly establish the level scheme.

This evaluation was carried out as part of a joint IAEA-ICTP workshop for Nuclear Structure and Decay Data, organized and hosted by the IAEA, Vienna and ICTP, Trieste, Aug 22 to Sept 2 2016. The evaluation work was coordinated by E.A. McCutchan (BNL). This work supersedes the previous previous $A=217$ evaluation (2003Ak06) by Y.A. Akovali.

Cutoff Date: All data received prior to December 1, 2017 have been considered. Main source of bibliographic search was the NSR database (2014Pr09) available at NNDC, BNL webpage: www.nndc.bnl.gov/nds/


General Comments: The decay Q values and particle-separation energies are from 2017Wa10. Theoretical conversion coefficients are from BrIcc code (2008Ki07), with an implied uncertainty of 1.4%, when not stated.

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