Feasibility of Nuclear Plasma Interaction studies with the Activation Technique

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28th August 2018
Declaration of Authorship

I declare that this thesis titled, “Feasibility of Nuclear Plasma Interaction studies with the Activation Technique” is my own work, that it has not been submitted for any degree or examination in any other university, and that all the sources I have used or quoted have been indicated and acknowledged by complete references.

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“It is a law of life that problems arise when conditions are there for their solution.”

Walter Sisulu
Keywords

‘Feasibility of Nuclear Plasma Interaction studies with the Activation Technique’

Thembalethu Nogwanya

IC
BIC
EC
NPI’s
HEDP’s
DIGS
NEEC
NEET
Plasma
S-process
R-process
P-type detector
N-type detector
Abstract

Feasibility of Nuclear Plasma Interaction studies with the Activation Technique

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Master of Science Degree

Electron-mediated nuclear plasma interactions (NPIs), such as Nuclear Excitation by Electron Capture (NEEC) or Transition (NEET), can have a significant impact on nuclear cross sections in High Energy Density Plasmas (HEDPs). HEDP environments are found in nuclear weapons tests, National Ignition Facility (NIF) shots and in the cosmos where nucleosynthesis takes place. This thesis explores the impact of NPIs on highly excited nuclei. This impact is understood to be more intense in highly-excited nuclei states in the quasi-continuum which is populated by nuclear reactions prior to their decay by spontaneous $\gamma$-ray emission. Attempts thus far have failed in measuring the NEEC process \cite{1, 2}, while NEET process has been observed experimentally \cite{3, 4}. Direct observation of NPIs is hindered by the lack of a clear signature of their effect in HEDP environments. Hence this should test a new signature \cite{5} for NPIs for highly-excited nuclei by investigating isomeric to ground state feeding from the isomeric state. An experiment was performed using the reactions $^{197}\text{Au}(_{13}\text{C}, _{12}\text{C})^{198}\text{Au}$ and $^{197}\text{Au}(_{13}\text{C}, _{12}\text{C}2n)^{196}\text{Au}$ at Lawrence Berkeley National Laboratory in inverse kinematics with an $^{197}\text{Au}$ beam of 8.5 MeV/u energy. Several measurements were performed with different target configurations. The activated foils were counted at the low-background counting facility of Lawrence Livermore National Laboratory. From these data, the double isomeric to ground state ratio (DIGS) were extracted with the assistance of the decay equations that were included in the experiment. As the NPIs effects are rather small the lines for analysis had to be chosen carefully so that the extracted ratios would not contain significant errors. The measured DIGS ratios were then compared with the result of the theoretical DIGS ratios. The results showed that the calculated DIGS ratios deviated substantially from unity although this was with large uncertainties. Because of the large errors obtained, the DIGS ratios were found to be inconclusive as a signature for detecting the effects of NPIs such as angular momentum distribution changes in HEDP environments.
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I would like to dedicate this work to my dear father S. Nogwanya
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Thembaletlu Nogwanya
Chapter 1

Introduction

Over the last decades, the processes of electron nuclei-plasma interactions have been studied extensively theoretically and experimentally. Nuclear plasma interactions (NPIs) such as Nuclear Excitation by Electron Capture (NEEC) or Transition (NEET) are rare nuclear excitations that are theorized to occur in certain isotopes [6]. NEET and NEEC are expected to cause significant changes in reaction cross sections in High Energy Density Plasmas (HEDPs) such as those found in the National Ignition facility (NIF) shots and astrophysical settings [7]. For NEEC, a free electron is resonantly captured into a bound atomic state and the same time it transfers its capture energy to the nucleus, which undergoes a transition from the ground state to an excited state. For the NEET process, atomic electrons transition with binding energy difference equal to the nuclear excitation energy. Then, for NEET, the binding energy and, for NEEC, the free electron energy plus the binding energy is resonantly transferred to the nucleus via a virtual photon and the nucleus becomes excited. In the end, whether we talk of the NEEC or NEET process, NPIs remain hardly accessible via many experiments because of the extreme narrowness of nuclear transitions ($\Gamma \leq 1 \mu\text{eV}$).

1.1 Astrophysical Environments

For a star in thermal equilibrium, an internal energy source is required to balance the energy that is lost in the form of radiation from its surface. This energy source is provided by nuclear reactions that take place in the interior of the star, where the temperatures and densities are very high temperatures $T \simeq 200 \text{ MeV}$ [8]. Apart from the energy generation nuclear reactions that take place in stars can
change the stars composition through transmutation of one element into another. Thus, an understanding of NEEC and NEET may play a major role in production or destruction of isotopes since isotopes evolution undergoes several processes.

In the context of nucleosynthesis, the formation of the heavier elements comes from the absorption of neutrons and protons by the pre-existing nuclei [9]. The absorption of a neutron is sometimes slow and the formed nuclei usually beta decay back to the beta-stability curve before absorbing the next neutron. This process is known as the slow neutron capture (S-process) [10]. Though, if the neutron flux is very high, the nuclei will capture as many neutrons as it can which tend to be too fast for beta decay to occur and can be driven to extremely neutron-rich nuclei. This process may happen during explosive scenarios, for example, core-collapse supernovae and neutron star mergers where there are high neutron densities and temperatures and this rapid creation is called (R-process) [11].

1.2 NEEC and NEET in plasma environment

In this work, we are mainly concerned with the role of the NEEC and NEET process in the cosmogenic nucleosynthesis. In the internal conversion (IC) process, the energy released from the nucleus is used to expel an orbital electron (bound state) from the atom to the continuum. Bound internal conversion (BIC) is the same as the IC but here the electron is promoted to the bound state and electron capture (EC) is the type of decay in which the nucleus of an atom draws in an inner shell electron. Hence NEEC and NEET could play a major role in the formation of heavier elements since both these process are time inverses of IC, BIC, and EC. These processes are expected to allow determination of atomic vacancy effects on nuclear lifetimes and population mechanisms of excited nuclear levels through an influx of electrons [12].

1.3 Motivation and study objectives

The electron-mediated nuclear plasma environment can enhance the population of nuclear-excited states through the coupling of nuclear transitions to transitions including bound atomic states of the charged ions. The processes that can occur,
nuclear excitation by electron transition (NEET) and nuclear excitation by electron capture (NEEC), are difficult to calculate owing to the many-body physics of the atomic system and plasma consequences on atomic level widths and densities.

There have been broad theoretical studies that have been undertaken to try to study these two processes [13–15]. Amongst these, attempts to measure NEEC have been unsuccessful [16], including attempts to detect the NEEC process resonantly by exciting the ground state of $^{181}$Ta and $^{187}$Os in plasmas generated by the Omega Facility at the Laboratory for Laser Energetics at the University of Rochester [17].

The limited observations of the parallel bound-state process NEET are controversial and have been restricted to non-plasma environments [18–20]. This study attempts to fill in those gaps by inducing NPIs in highly-excited nuclear states produced by nuclear reactions prior to their decay by spontaneous $\gamma$-ray emissions. The large density of nuclear states at these high excitation energies increases the probability that the energy from the atomic transition will resonantly match an available nuclear transition. To this end we observe the NPI effects following the $(n,2n)$ reaction at the National Ignition Facility (NIF), which populates states several MeV below the neutron separation energy prior to $\gamma$-ray emission. Despite the fact that energy transfer by NPIs is small, changes in angular momentum are likely to occur even with this small energy transfer in highly-excited nuclei.

A new experimental approach to detect the signature of NPIs on highly-excited nuclei was proposed by D. L. Bleuel et al. [5]. This novel experimental method is called “the differential population of high-spin isomeric versus the ground state of the de-excited nucleus”. In particular the $^{134}$Xe$(n,2n)$ reaction was proposed, which has a long-lived ($t_{1/2} = 2.2$-days) $11/2^-$ isomer and a ($t_{1/2} = 5.2$-days) $3/2^+$ ground state. The effect of NPIs of producing differential isomer population for reaction products exposed to a plasma or non-plasma environment is discussed in detail in Section 3.5 (also see Figure 3.9). The Double-Isomer-to-Ground-State (DIGS) ratio for the $^{133m}$Xe / $^{133}$Xe fraction formed in the exploding pusher capsule plasma divided by the same quantity for an externally-mounted, non-plasma Target option Activation Device (TOAD) sample, illustrates whether NPIs had effect on highly-excited states in $^{133}$Xe. The DIGS ratio is given by:

$$R_{DIGS} = \frac{N_{Xe-133m}^{capsule} / N_{Xe-133}^{capsule}}{N_{Xe-133m}^{TOAD} / N_{Xe-133}^{TOAD}}$$ (1.1)
with $N$ being the number of each state populated, determined from its characteristic-energy $\gamma$-ray emissions measured in a high-purity germanium detector.

The primary goal of this thesis is to calculate the DIGS ratio on highly excited states using the activation technique and the $^{197}\text{Au}(^{13}\text{C}, ^{12}\text{C})^{198}\text{Au}$ and $^{197}\text{Au}(^{13}\text{C}, ^{12}\text{C}_2n)^{196}\text{Au}$ reactions at Lawrence Berkeley National Laboratory in inverse kinematics with a $^{197}\text{Au}$ beam of $8.5\;\text{MeV/u}$ energy. These Au isotopes have similarly long-lived $12^-$ isomers and $2^-$ ground states compared to Xe.

The $\gamma$-ray decays from the isomer and ground state will be measured following the activation in a simulated plasma and non-plasma environments. The plasma environments will be simulated by using different target configurations (see Section 3.3 the targets illustration on the following Figures 3.5, 3.6 and 3.7). From the decay data differences in the feeding of the isomer versus ground state (the proposed signature for NPI) will be investigated. If angular momentum is changed due to NPIs the feeding pattern may different. This change in feeding is at the lane of this thesis to investigate if the proposed method is feasible to detect NEEC or NEET.
Chapter 2

Interaction of $\gamma$ ray with matter

2.1 Types of interaction with matter

When charged particles, photons, and neutral particles penetrate a material some of the energy from the particles may be absorbed completely, some may be scattered and some may pass through without any interactions with the material. The process of absorption, scattering and pair production can be described and explained in terms of interaction between particles. There are different kinds of interaction through which a particle passing through a material can deposit its energy. In this chapter, we consider the interaction of photons with matter.

2.1.1 Attenuation

As a photon passes through matter, there is no way to precisely know how far it will take before being involved in an interaction or what type of interaction it will be involved in. Either Compton or photoelectric interactions will absorb some of the photons from the beam of particles. This process is known as attenuation [21]. Suppose at some distance, $x$, into the material $N_0$ photons are moving through a slab of material. Then after exiting the material the number of photons in the beam is reduced. The following Equation 2.1 shows how the number of photons is reduced [21],

$$N_x = N_0 e^{-\mu x},$$

(2.1)
where $\mu$ is known as the linear attenuation coefficient and is the actual fraction of photons interacting per unit thickness of the material. The exponential decay in Equation 2.1 comes from the fact that over a short distance, the probability of losing a particle from the beam is proportional to the number of particles left (see Figure 2.1).

Figure 2.1 – A schematic of a linear attenuation, $N_0$ is the number of incoming photons and $N_x$ is the number of photons after passing the slab.

2.1.2 Photoelectric effect

In the process of the photoelectric effect, a photon gets absorbed by the atomic electron which is then ejected from the atom. This only happens if the incoming photon energy is larger than the electron binding energy. Because the atom is much heavier when compared to the mass of an electron, the ejected electron will practically take all the energy and momentum of the incoming photon (see Figure 2.2). The kinetic energy of the ejected electron, $E_K$, is then given by:

$$E_K = hf - E_B,$$

where $h$ is the Planck constant which is equal to $6.6260700410^{-34} m^2 kg/s$, $f$ is the frequency of the incident photon and $E_B$ is the electron binding energy. The ejected electron is known as the photoelectron.

The probability of photoelectric absorption depends on the photon energy, the binding energy of the electron and the atomic number of the atom, $Z$. The probability of this process increases the more bound the electron is to the nucleus. Hence K-shell electrons are the most affected ones provided that $\gamma$-ray energy exceeds the K-electron binding energy. The probability for this is given by the following relation [22]:

http://etd.uwc.ac.za/
\[ \mu_m \propto Z^4 / E_{\gamma}^3 \]  

where \( \mu_m \) is the photoelectric mass attenuation coefficient which is the rate of photon interactions per 1-unit (g/cm\(^2\)) area mass and \( Z \) is the atomic number of the atom. Figure 2.3 shows how the attenuation varies with photon energy. The decrease of \( \mu_m \) with the increase in energy is interrupted by a series of jumps, called absorption edges.

The probability of the photoelectric absorption increases as the photon energy decreases. It also becomes smaller after the absorption edge. This is because the photon energy does not have enough energy to remove the electron in the atomic
shell, as seen in Figure 2.3. The determination of the mass attenuation coefficient for gold was carried out by J. L. Glover et al., [24].

2.1.3 Compton scattering

In Compton scattering, the $\gamma$ ray directly interacts with a free or a bound electron. After the interaction, the $\gamma$ ray scatters, see Figure 2.4. The recoil energy of the electron is equal to the difference of the energy lost by the $\gamma$ ray and the electron binding energy (Equation 2.4).

$$E' = \frac{M_0c^2}{(1 - \cos(\theta) + \frac{M_0c^2}{E_\gamma})},$$

(2.4)

where $M_0c^2$ is the electron rest mass energy of 511 keV, and $\theta$ is the angle between the incident and scattered photon shown in Figure 2.4. The direction of the electron and the $\gamma$ ray is dependent on the $\gamma$-ray energy which is transferred to the electron. When Compton scattering takes place in a detector, the scattered electron is usually stopped in the detection medium. The detector gives an output pulse that is proportional to the lost energy of the incident $\gamma$ ray. This energy depends on the scattering angle, $\theta$, [21]. As shown in Figure 2.5 the probability of Compton scattering becomes small as the energy of the $\gamma$ ray become larger.
Furthermore, Compton scattering is weakly dependent on the atomic number, of the absorbing material.

### 2.1.4 Pair production

In general pair production is an event where $\gamma$-ray energy is directly converted to matter. For pair production to happen the $\gamma$ ray must have an energy of at least 1.022 MeV (mass of two electrons). With the interaction of the electromagnetic field of the nucleus of an atom the energy is converted into the mass of an electron-positron pair. The $\gamma$ ray disappears as an electron-positron pair appears. The positron that is formed quickly disappears by reconversion into photons in the process of annihilation with another electron in matter. The annihilation converts the rest masses of the electron and positron into two 0.511 MeV $\gamma$ rays, emitted in opposite directions (see Figure 2.6).

![Figure 2.5](http://etd.uwc.ac.za/)

**Figure 2.5** – The linear absorption coefficient for Compton scattering, photoelectric effect, pair production and total photon attenuation [25]. This figure represents photon interaction the vertical axis represent the mass attenuation coefficient, which is discussed in Section 2.1.1.

It can be seen in Figure 2.5, for low photon energy, the photoelectric effect is the predominant process, with Compton scattering happening over all energies but peaks in the midrange, while pair production occurs only above 1.022 MeV and dominates as the photon energy increases.
2.2 Germanium Detector

Germanium semiconductor detectors are commonly used to detect γ rays that are emitted in the nuclear process during radioactive decay. The γ-ray photons will give some or all the energy to an electron in the material. The ionized atom will then further collide with other electrons and result in many more electrons being created. A germanium detector is an intrinsic semiconductor, and the presence of any donor such as lithium or acceptor such as boron impurities will cause the material to be either $n$- or $p$-type respectively. As the charge carriers are generated in pairs, the concentration $n_b$ of electrons in the conduction band equals the concentration $p_b$ of holes in the valence band ($n_i$—intrinsic hole concentration and $p_i$—intrinsic hole concentration) [26]. Typical semiconductors are group IV elements and have 4 electrons in the valence shell. The energy gap ($E_g$) between the conduction band and the valence band is known as the band gap and it has no energy levels which are accessible for electrons to possess; see Figure 2.7 which shows a layout of these bands. An external energy may excite an electron out of the valence band across $E_g$ into the conduction band. Such excitation creates an electron-hole that carries on as a positively charged particle. Semiconductors have energy gaps that lie roughly in the range 0.1 - 3 eV. The absorption of a photon and excitation of electrons across the $E_g$ occurs only if $E_\gamma \geq E_g$.

The $p$-type material has an excess of holes and the $n$-type has an excess of electrons. Bringing these two material together in contact while keeping the two grounded, you then get $p$-$n$ junction solution. Applying a reverse bias across the $p$-$n$ junction will block electrical current flow. Under reverse bias conditions, the voltage potential is connected negative to the $p$-type (acceptor atoms) material and positive to the $n$-type (the donor atoms) material. This will increase the $p$-$n$ junction width due to a lack of electrons and holes and presents a high impedance.
Chapter 2 Interaction of $\gamma$ ray with matter

Figure 2.7 – Conduction band empty while Valence band is full with electrons.

path, almost an insulator. Holes in the $p^+$ region are attracted from the junction in the direction of the $p$ contact, and electrons ($e^-$) in the $n^-$ region are attracted from junction to the $n$ contact, Figure 2.8 shows the formation of a depletion layer by charge diffusion, space charge density, electric field and built-in potential in a $p$-$n$ junctions.

Figure 2.8 – Formation of depletion region in a semiconductor material.

The germanium material can be either $p$-type or $n$-type. These types all depend on the concentration of donor or acceptor atoms in the crystal. In order to connect the diode to an electrical circuit and amplify the signal output, contacts on the crystal are needed. The electrical contacts on the signal are thick, lithium contact, which is the $n^+$ contact and $p^+$ which is thin ion-implanted contact (Boron). The lithium contact is thick since it is diffused into the germanium. For $n$-type material, the thick lithium contact is placed on the inside surface while the thick ion implanted contact is placed on the outside surface; for the $p$-type the arrangement is reversed. For example, see Figure 2.9 and 2.10.

In an High-Purity Germanium (HPGe) detector, the $p$-type is placed as the cathode and $n$-type layer as the anode ($p$-$i$-$n$ structure), the HPGe in between can be
effectively depleted of charge carriers (HPGe has an impurity concentration of \(10^{10}\) atoms \(cm^{-3}\) instead of \(10^{13}\) atoms \(cm^{-3}\)) [27]. The depletion can be obtained by applying a high voltage of up to 5 kV between the contacts, which are separated by a distance up to around 3 cm [28]. The narrow depletion layer at the junction of the germanium and the opposite contact impurity type gets expanded by a reverse-bias of high voltage application. When the voltage is increased, the depletion layer grows rapidly until it reaches the outer contact, causing the detector volume to be entirely depleted.

An operation voltage, \(V_0\), is set at least in the 500 V range which is high enough in order to make sure a high charge mobility through the volume is maintained. For a depleted detection, the impurity ions can be seen as a fixed space charge distribution, \(\rho_s\). Total electric field then results from the combined effects of the two, \(\rho_s\) and \(V_0\). This electric field is weak on the side of the detector where two contributions have different signs and strong on the other [29]. In the coaxial

\[ \text{Figure 2.9 – Illustration of the HPGe N-type crystal.} \]

\[ \text{Figure 2.10 – Illustration of the HPGe P-type crystal.} \]
detection the field lines are concentrated in the inner contact due to the geometry. To concentrate this effect the type of the detector and polarity are chosen in such a way that the depletion starts from the outer contact. With a \( n \)-type crystal this can be obtained by putting the lithium-drifted contact on the inside and the ion-implanted contact on the outside. In use of the \( p \)-type detector, contacts are reversed. For the purpose of this work \( n \)-type HPGe detector was used. This type of detector is useful for many applications such as for studying high-energy \( \gamma \) rays. The HPGe \( n \)-type, co-axial detectors are at present the best \( \gamma \)-ray detectors in the energy range \( \sim 0.1 \) to \( \sim 10 \text{ MeV} \) because of the high energy-resolution of the semiconductor Ge material, and the high full-energy efficiency of large detectors [30]. For typical nuclear structure experiments with high \( \gamma \)-ray multiplicities, such detectors are organized in large arrays.
Chapter 3

Theoretical background

3.1 Nuclear Decay

The internal conversion (IC) process competes with \( \gamma \)-decay emission. When a nucleus is excited from the ground state to an excited state, at some time later an excess energy will be released by some form of decay. Often, the nucleus will decay by \( \gamma \)-ray radiation. Nevertheless, in other cases, the virtual photon is released from the nucleus only to interact with one of the inner most orbital electrons and as a result, the virtual photon energy is transferred to the electron. Thus the nucleus is then said to have undergone IC. Figure 3.1 shows different ways in which the nucleus de-excites. The generic equation for internal conversion is \( \frac{A}{Z}X^* \rightarrow \frac{A}{Z}X^{+1} + e^- \), where \( \frac{A}{Z}X^{+1} \) is a ionized state and \( e^- \) is one of the atomic electrons.

The Auger effect is a transition of an electron in an atom filling an inner shell vacancy causing the emission of another electron. When a core electron is removed from a core level (in other words electrons that are in the energy shells closer to the nucleus) of an atom, leaving a vacancy, an electron from a higher energy level may fall into the vacancy, resulting in the release of energy. Although sometimes this energy is released in the form of an emitted photon, the energy can also be used to remove an electron from the atom. This second electron ejected is known as an Auger electron [31], and this can consist of a number of vacancies in the atom orbits and thus emitted electrons.

For electron capture (EC) see Figure 3.2, nuclei having enough number of protons may pick up a free electron from one of the inner orbits which immediately couples with a proton in the nucleus to form a neutron and then emit a neutrino, conserving mass and energy. This process can lead to X-ray or Auger electrons emission. For
instance, in the process of IC and orbital EC, an electron leaves its atomic orbit and the vacancy is soon occupied. Here there are two processes competing; one is the emission of an X-ray which is due to the transition of an electron from an outer shell to the vacancy in a shell closer to the nucleus. The second one is that the energy difference between the orbits is not released as an X-ray instead the energy is used to knock out an electron from the orbit.

The fundamental energy conservation relation governing the conversion electron energy is given by:

$$E_e = E_X - E_B,$$

(3.1)

where $E_e$ is the kinetic energy of the electron, $E_X$ is the transition energy and $E_B$ is the binding energy of the electron from the atom.

The competition of IC and $\gamma$-ray decay allows us to define the IC coefficient, $\alpha$ of a given $\gamma$ ray as the ratio between the probability of the nuclear state decaying by IC ($\lambda_e$) or $\gamma$-ray emission ($\lambda_\gamma$) as follows:
IC electrons may be ejected from different atomic shells, giving rise to $\alpha_k$, $\alpha_L$, $\alpha_M$, etc. Since the total probability of decay must equal the sum of the probability of decay via different paths, using Equation 3.2 we can then define the total decay rate as follows:

$$\lambda_{tot} = \lambda_\gamma (1 + \alpha)$$

The coefficients can be calculated theoretically in electric and magnetic transitions using the equation below.

$$\alpha_i^L = Z^3 \left( \frac{L}{L+1} \right) \alpha_{fs}^4 \left( \frac{2m_e c^2}{E} \right)^{L+5/2}$$

Here ($i = K, L, M, N, ...$), $L$ is the multipole order, $Z$ is the atomic number, $m_e$ is the rest mass of the electron, $E$ is the energy of the transition, and $\alpha_{fs}$ is the fine structure constant [33]. The above equation tells us that, the internal conversion coefficient increases with $Z^3$. This coefficient also increases for large multipoles, $L$. 

Figure 3.2 – Scheme of two types of electron capture. Top: The nucleus absorbs an electron. Lower left: An outer electron replaces the electron hole. An X-ray, equal in energy to the difference between the two electron shells, is emitted. Lower right: In the Auger effect, the energy released when the outer electron replaces the inner electron is transferred to an outer electron. The outer electron is ejected from the atom, leaving a positive ion [32].

$$\alpha = \frac{\lambda_e}{\lambda_\gamma}$$

(3.2)
However, there is another type of IC which is known as Bound Internal Conversion (BIC) which occurs between atomic states. In this process, a nucleus in an excited state will go through a transition to a lower-lying state with excitation of an electron to a formerly empty bound final state [34], when the binding energy of the converted electron is larger than the nuclear transition energy. BIC has recently been observed at GANIL in $^{125}$Te [4].

### 3.2 NEET and NEEC

When an electron in the innermost shell is excited to an outermost shell, the electron can then de-excite in various ways. The electron can move from higher shell $f$ to the lower shell $i$; this process will then emit a photon in the form of an X-ray. Alternatively, the transition energy can be transferred to one of the outer electrons; this will eject an Auger electron from the atomic shell. The third process, although its probability of happening is very small when compared to the first two, is called Nuclear Excitation by Electron Transition (NEET). The existence of NEET was first predicted by Masato Morita in the year 1973 [35]. In NEET, an electron from the upper level will flow into the vacancy. A virtual photon will be emitted which is then absorbed by the nucleus. This is possible only if the nuclear and atomic transitions have nearly the same multipolarity and matching energies.

A number of experiments have been performed looking for NEET in various isotopes. NEET has been successfully observed for the $^{197}$Au [36], $^{189}$Os [37] and $^{237}$Np [38] isotopes. The most precise measurement in $^{197}$Au was done by Kishimoto [36], who radiated a gold foil with monochromatic photons. In order for NEET to occur, the nuclear and electronic transition energy need to overlap. Moreover, Morita noted that NEET may be regarded as the inverse of Bound Internal Conversion (BIC) [35].

**Probability of NEET**: The probability of NEET taking place, $P_{NEET}$, is defined as the probability that the decay of the initial excited atomic state, $i$, will result in the excitation of and subsequent decay from the corresponding nuclear state. The expression for the probability of $P_{NEET}$ is given by [39];

$$P_{NEET} = \left(1 + \frac{\Gamma_f}{\Gamma_i}\right) \frac{W^2}{\delta^2 + \frac{1}{4}(\Gamma_f + \Gamma_i)^2} , \quad (3.5)$$
where \( \Gamma_i \) and \( \Gamma_f \) are the initial and final atomic state widths, and \( W \) is the matrix element for the nuclear atomic coupling.

\[
\delta_{if} = \Delta E_e - \Delta E_N, \tag{3.6}
\]

with \( \Delta E_e \) being the electron transition energy and \( \Delta E_N \) being the nuclear transition energy.

\( P_{NEET} \) is very small when compared to \( P_{Auger} \) (the Auger electron emission probability) and \( P_{X-ray} \) (the X-ray emission probability). See for instance the list of theoretical and experimental probability calculations for these three different processes in Ref [40].

Nuclear Excitation by Electron Capture (NEEC) was proposed by V.I. Goldanskii et al. [41]. In the resonant process of NEEC, a free electron is captured by a bound atomic orbital state, as illustrated in Figure 3.3, with the binding energy plus the electron kinetic energy being equal to the energy of the first-excited nuclear state. In Figure 3.3 which shows the NEET and NEEC process the nucleus is initially in its ground state (G). For NEET the binding energy and for NEEC the free electron energy plus the binding energy is resonantly transferred to the nucleus and the nucleus becomes excited. NEEC is regarded as the inverse process of IC which results in an excited nuclear state that can decay either radioactively or by IC [42]. Despite the fact that there have been a vast number of theoretical studies performed on this subject, previous attempts to measure NEEC have been unsuccessful [16, 43]. A major challenge in the look for NPIs is the identification of a clear signature of the effect in an exceedingly disordered High Energy Density Plasmas (HEDPs) environment. Usually, the energy transfer on scales of keV is small and the signal of emitted radiation is weak compared to the background radiation in the HEDP.

### 3.3 Experiment design for NPIs

In this section, we discuss the design of an experiment that will allow accessing in electron mediated nuclear plasma interactions, the effect of NEEC or NEET.
The prepared target foils were irradiated using the 88-Inch Cyclotron accelerator facility of Lawrence Berkeley National Laboratory in Berkeley. Differential isomer population may be used to observe NPI effects in the HEDP plasma environment. The activation of $^{13}$C occurring stable isotope with $^{197}$Au heavy ion beam, results in the formation of unstable gold nuclei, $^{196}$Au, $^{198}$Au and etc. Reaction channels of $^{197}$Au gold for the reaction of interest are given below:

$^{13}$C($^{197}$Au, $^{198}$Au$^*$) reaction, the emission of $\gamma$ rays comes from the composite nucleus by the unstable remaining residue nuclei, $^{198m}$Au and the excited $^{198}$Au nucleus. The unstable nucleus has a half-life of 2.3 days and undergoes decay by isomeric transition. It also has spin parity of 12$^{-}$. The excited residue nucleus, $^{198}$Au has a half-life of 2.7 days and a spin parity of 2$^{-}$. The $^{13}$C($^{197}$Au, $^{196}$Au$^*$) reaction, resulting in an emission of two neutrons from the composite nucleus. The highly excited unstable nucleus, $^{198m}$Au, has a half-life of 9.6 hours and a spin parity of 12$^{-}$. The excited residue $^{196}$Au has a half-life of 6.2 days and a spin parity of 2$^{-}$. The resulting unstable gold isotopes may interact with the $^{209}$Bi (bismuth) target (see the set-up of the targets in in Figure 3.5, 3.6 and 3.7). The $^{209}$Bi isotope is one of the naturally occurring isotopes with high electron density number. In a highly electron-rich environment plasma, it is possible to resonantly excite a nucleus through energy with an electron [2]. Here $^{209}$Bi foil target will act as an electron-rich environment plasma (see Figure 3.4) target which may allow for the interaction between the electrons and highly excited produced gold isotopes.
For the above reason, subsequently we expect that NEEC/NEET should occur in the created ordered plasma environment.

In the Close target shown in Figure 3.6, the beam should start off at about 14% the speed of light and stops in about 40 microns of the target presented. This is the very roughly average time of < 1 ps. The lifetimes of the states in the quasi-continuum are on order of femto seconds. The beam stops before the nucleus has de-excited into the discrete states which tend to be close to pico seconds lifetimes, allowing time for highly-excited nuclei to interact with the ordered plasma. On the opposite side, the Far target is shown in Figure 3.7 the beam travels over a millimeter before encountering the ordered plasma. With 14% the speed of light, this will then take about 24 ps, giving the nucleus more time to decay to the discrete states. There is a requirement that the energy of the electron, binding plus relative kinetic energy, should exactly match the energy of the nuclear transition. Hence in the discrete states, there is very little chance that there will be a state to excite to exactly as much as the electron energy.

The last target was the Blank target, which only consisted of the plasma-like environment. Here there was no thin target for reactions to take place. This target was a reference point of the other two targets used (Close and Far target). With the use of three targets, the Double Isomer-to-Ground State ratio (DIGS) will be investigated using section 3.4.

![Image](http://etd.uwc.ac.za/)

**Figure 3.4** – For an accelerator beam, an ordinary target looks like an electron beam, an ordered plasma.
Chapter 3 Theoretical background

Figure 3.5 – Excited \( Z^{A+1} \) X residuals made via binary transfer from thin target recoils into a thick metal foil which acts like a plasma target.

Figure 3.6 – Close target where NEEC/NEET can occur on quasi-continuum states.

Figure 3.7 – Far target, the excited nuclei will decay to ground state or isomer, and NEEC/NEET will not occur in these states.

3.4 Change in Angular Momentum Distribution in Nuclear Plasmas

The distribution of highly excited states of emitted particles in a heavy ion collision can be represented by the following function \([45, 46]\):
\[ \rho(J) \propto (2J + 1)e^\left[ -(J + \frac{1}{2})^2 / 2\sigma^2 \right], \] (3.7)

with \( \rho(J) \) being the probability distribution of levels as a function of spin \( J \), \( \sigma \) is a parameter which limits the population of high-spin levels and is in principle related to both the moment of inertia of the excited atom and its temperature [47]. The de-excitation from a specific spin level by a transition is assumed to populate residual spin levels with a probability dependent on the availability of the specific levels as illustrated by equation 3.7 (see Ref.[48]). The emitted particles in the reaction can be described by the following equation [49],

\[ \sigma(J_c, E) = \pi\lambda^2 \sum_{S=|I-s|}^{I+s} \sum_{S=|J_c-s|}^{J_s+s} \frac{2J_c + 1}{(2s + 1)(2I + 1)} T_l(E), \] (3.8)

where \( \sigma(J_c, E) \) is the cross section at spin \( J_c \) and energy \( E \), \( \lambda \) is the de-Broglie wavelength of the incoming particle, \( s \) is the spin of the projectile, \( I \) is the spin of the target nucleus and \( T_l(E) \) is the barrier transmission coefficient of a particle with orbital angular momentum \( l \) and energy \( E \). A highly excited nucleus remains. These primary fragments are assumed to have an angular distribution function related to the nuclear level density [50],

\[ N(J) = (2J + 1)e^{-J(J+1)/2B^2}, \] (3.9)

with the probability function often written as,

\[ P(J) = P(0)(2J + 1)e^{-J(J+1)/2B^2}. \] (3.10)

where \( P(0) \) is the density of levels with zero total angular momentum, \( B \) is approximately equal to the root mean square value of \( (J + 1/2) \) [48]. This functional form which was originally chosen from statistical considerations has, however, has also been predicted theoretically by J. R. Nix et al., [51] and by W. J. Sarantites et al., [52]. Excited nuclei in the continuum will decay through a \( \gamma \)-ray cascade with the assumption of dipole transition before it decays to the isomeric or ground state. Furthermore, parity must be preserved with cascade parity being assumed to be equal to a number that will always induce the same parity \( (\Delta \pi) \) [47]. Each \( \gamma \)-ray decays to states given by the probability distribution equation 3.9, with the number of \( \gamma \)-rays in the cascade given by the following equation [51],
Chapter 3 Theoretical background

\[ < N_{\gamma} > = (E_{\gamma})^{\frac{1}{2}}/(l + 1), \]  

(3.11)

and here \( E_{\gamma} \) is the excitation energy of the nucleus. In this study, let's assume the number of \( \gamma \)-rays emitted in the cascade to be equally three. This assumption is based on the model which was developed by J. R. Huizenga and R. Vandenbosch [47–53] although it is found to range between 3 and 5 [48]. Following the \( \gamma \)-ray cascade and the consequent change in the angular momentum distribution given by \( N(J) \), the nucleus will decay to the isomeric or ground state. Simulations predict that multiple NPIs will increase the angular momentum distribution; see the article by D. L. Bleuel et al., [5]. The increase will affect the nucleus prior to the \( \gamma \)-ray cascade, impacting the end state of the nucleus. In conclusion, the isomer to ground state ratio will be perturbed, see Figure 3.8.

Figure 3.8 – This figure illustrates the distribution of total spin values of the compound nucleus due to NPIs. The blue dashed line represents the actual levels in the nucleus as a function of angular momentum. ILD stands for Intrinsic Level Density. The green spike represents a single initial excited state of \( L = 2 \), ESD stands for Entrance Spin Distribution. The curve after that represents a model which randomly adds or subtracts angular momentum to that state proportional to the number of end states available. After two absorbed or emitted \( \gamma \)-rays, you would expect the red spin distribution. After four, the light blue, etc. The distribution quickly diffuses towards the natural spin distribution. Calculations by J. Escher, LLNL Nuclear Theory and Modeling Group [54].

3.5 Double Isomer-to-Ground State ratio

The interaction between highly-excited nuclear states and High Energy Density Plasma (HEDPs) environments such as neutron and proton-rich astrophysical environments can profoundly affect nucleosynthesis (processes that are believed to have occurred immediately after the big bang). It is possible to excite a nucleus...
through energy transfer from an electron i.e. in the process of NEEC and NEET discussed in section 3.2. The observation of a nuclear-plasma induced change in the isomer-to-ground state ratio would create the first observation of nuclear plasma interactions (NPIs) [55].

D. L. Bleuel et al. [5], proposed a new method to look for the effects of NEET and NEEC on highly-excited nuclei, which is the differential population of an isomeric state against the ground state of a de-excited nucleus. With the density of available nuclear states higher, the probability of a nucleus interacting with nearby electrons is thus greater, because of the high-level density and shorter lifetimes (larger widths) in the quasi-continuum, there is a much higher chance of there being some state or tail of a state that will match the electron energy. Our nuclei of interest will be produced by \((n, \gamma)\) or \((n, 2n)\) reactions with the help of a thin target. These reactions will initially populate states several MeV above and around the neutron separation energy (see Figure 3.9). The ratio of isomer to ground state decay will be measured in both cases. For instance, see the illustration of a plasma environment effect which is shown in Figure 3.9. This illustrates the creation of highly excited nuclei in the quasi-continuum, in an environment where there are free electrons (allowing for the potential of NPIs). In the quasi-continuum the level density is defined as the number of quantum levels per energy unit as a function of excitation energy [56]. In the quasi-continuum region level widths are small enough to allow NEEC and NEET processes to take place. With the new method, we expect NPIs effects, by inducing them on highly-excited, 1-5 MeV, nuclear states produced by the nuclear reaction prior to their decay by spontaneous \(\gamma\)-ray emission [5]. It is especially powerful while doing the measurement of this sort of ratios that the isomeric and ground state of the created isotope of interest goes on for a sufficiently long period of time (the samples must have high purity). It is necessary to limit the error sources (i.e. radiation and transportation times) which can contribute to the measurement results, in order obtain the accurate and reliable results.

Figure 3.9 shows the effect of NPIs in the quasi-continuum. The large density of nuclear states at the chosen range of MeV energies will increase the probability that energy from the atomic transition resonantly matches an available nuclear transition. The NPIs may change the angular momentum distribution [5], which then leads to a change in the subsequent isomer to ground state population. Finally, this will lead to a change in the isomer-to-ground state ratio when compared...
Figure 3.9 — Free electron is captured from the environment to the free atomic orbital, nucleus become even more excited possible modification in the high spins of the nuclei leading to a change in isomer to ground state ratio[5].

with the non-plasma environment. See also calculations for spin spreading due to NPIs by J. Escher in Figure 3.8 [54, 55].

To recognize the impact of NPIs to produce differential isomer population for reactions in versus out of a plasma environment, Equation 3.12 will be utilized [5],

$$ R_{DIGS} = \frac{N_{NPIiso}^{N_{ISO}}}{N_{NPIground}^{N_{GROUND}}} $$

(3.12)

where, $N =$ number of states populated which is determined from its characteristic energy of $\gamma$-ray emissions measured in a high-purity germanium detector, NPI = Nuclear Plasma Interactions, and iso is short for isomer. The DIGS ratio compares isomer and ground state production in and out of plasma conditions, with the condition that $R_{DIGS} = 1$ implies that there are no plasma effects, but if $R_{DIGS} \neq 1$ then this may be a signature that there have been plasma effects.
Chapter 4

Experimental Setup

This chapter is organized as follows. In section 4.1 we review experimental details. In section 4.2 we give details about targets that were used during the experiment. In section 4.3 we discuss information regarding the 88-Inch Cyclotron and beam selection. Furthermore, in section 4.4 we show graphical illustration during the time when the beam was on for all various targets used. In section 4.5 we discuss the counting process using HPGe detectors and chemical separation. We then close the chapter with section 4.6 where we discuss detection efficiency.

4.1 Experiment overview

The experiment was performed at the Lawrence Berkeley National Laboratory (LBNL), 88-Inch Cyclotron in Berkeley, CA, United State of America (USA). $^{197}$Au beam was produced with $8.5 \text{ MeV} / \text{amu}$ beam energy. The $^{197}$Au beam was used to radiate the thin $1 \mu m$ thick $^{13}$C foil target which was attached to an aluminum frame. For the Far target, three frames of aluminum were used with thickness of $2 \text{ mm}$ each and the bismuth target used was $1 \text{ mm}$ thick. From the set-up this gave a distance from the carbon foil to the bismuth foil backing to be $3 \text{ mm}$. For the Close target the set-up was similar except that they were two aluminum frame used. The Blank target were an aluminum frame with bismuth (same thickness as other targets) attached with epoxy. The $^{196}$Au$^*$ and $^{198}$Au$^*$ were studied using the $^{13}$C ($^{197}$Au, $^{196}$Au$^*$) and $^{13}$C ($^{197}$Au, $^{198}$Au$^*$) reactions.
4.2 Targets

In the experiment, three targets were used: Far target, Close target, and the Blank target. The Far target was constructed of a $2.3 \times 10^{-4} \frac{mg}{cm^2}$ $^{13}$C foil attached to an aluminum frame of dimension $33.0 \times 33.0 \times 1.5$ mm. The aluminum frame had a centrical hole of radius 6.5 mm which the carbon foil completely covered, see Figure 4.1 and the sketch in Figure 4.2. The carbon foil was attached to the frame by floating the foil in a bath of deionized water and picking it up using the frame. This was done carefully to make sure no tearing of the carbon foil occurred. The Far target had an additional 1.5 mm thick aluminum frame with a hole with the same dimension as above as a spacer. The last frame was an aluminum frame which had a piece of bismuth ($^{nat}$Bi) 1.1 mm thick attached with epoxy.

![Figure 4.1](http://etd.uwc.ac.za/)  
Figure 4.1 – Far target (top) being removed after irradiation and blank target (below) before irradiation.

![Figure 4.2](http://etd.uwc.ac.za/)  
Figure 4.2 – Sketch of the Far targets, the is approximately 12.5 mm diameter.
For the Close targets foils of bismuth with mass 1.5 g and 1.7 g, $^{13}$C was electro-plated to the surface of the bismuth forming a $2.4 \times 10^{-4}$ mg/cm$^2$ thick layer, see the mounting in Figure 4.3, Figure 4.4, Figure 4.5 and the sketch in Figure 4.6.

In order to do this, a solution denoted by RF was made by combining 2.5 g resorcinol, 3.6 g formaldehyde, and 88-liter acetic acid in 3.0 g dl (deciliter) of water. Then, 1.3 g $^{13}$C powder was added to 8.0 g RF and sonicated to make a homogeneous suspension. The suspension was allowed to gel at room temperature, then washed with acetone and allowed to air dry. Carbonization was then performed at 1050$^\circ$C under a nitrogen atmosphere for 3 hour producing a monolithic carbon target. This carbon target was used to sputter-coat bismuth wafers in a Gatan precision etching and coating system (PECS). This instrument is equipped with a vibrating crystal microbalance to control and monitor deposition thickness. Bismuth wafers were coated with the carbon target, these bismuth pieces were then attached to an aluminum frame using epoxy.

The Blank targets consisted of a spacer with no covering in the central hole and lastly an aluminum frame with the bismuth attached with the method previously described. The bismuth for the Blank targets were all of the same geometry with 1 mm thickness. In total two Far targets, two Close targets and two Blank targets were constructed. Each target was mounted on a copper plate, which was electrically isolated from the beam line and connected to a current integrator, measuring the total number of incident $^{197}$Au particles.

The time profile of the beam was measured by the rate of fusion-evaporation neutrons detected by a neutron monitor mounted outside the target and normalized using the total integrated current.

![Figure 4.3](http://etd.uwc.ac.za/) – Close target before irradiation mounted on a copper plate.
Figure 4.4 – Close after irradiation with Al plate.

Figure 4.5 – Close with Al plate removed.

Figure 4.6 – For Close targets the above construction was used with the motivation because beam spot kept getting bigger (for example bigger than the hole in aluminum frame) hence aluminum frame was mounted in front of the Close target.
4.3 88-Inch Cyclotron and irradiation

The 88-Inch is a K = 40 sector focused cyclotron with both light and heavy-ion capabilities. Protons and other light ions are available at high intensities (10-20 pµA) up to maximum energies of 55 MeV (protons), 65 MeV (deuteron), 135 MeV ($^3$He) and 140 MeV ($^4$He) [57]. Major instrumentation at the 88-Inch Cyclotron include the Berkeley Gas-filled Separator (BGS), and the superconducting VENUS ion source, one of the most powerful Electron Cyclotron Resonance (ECR) ion sources in the world [58–60]. A layout of the ion sources at the 88-Inch Cyclotron accelerator facility is shown in Figure 4.7.

![Figure 4.7 - Schematic layout of the ion sources of the 88-Inch Cyclotron accelerator facility [61].](http://etd.uwc.ac.za/)

A 8.5 $\frac{MeV}{amu}$ of $^{197}$Au was provided by the cyclotron for four successive days. The current before the bending magnets was noted. Also the current hitting the target was acquired through a current integrator. Each target was in the beam for approximately 8 hours. They were then transported on a daily basis to the low background Nuclear Counting Facility (NCF) at Lawrence Livermore National Laboratory, Livermore, CA for chemical separation and counting.

4.4 Beam monitoring

In this section, we show the beam profile during the targets irradiation. First is the beam profile for the Far target followed by the Blank target beam profile and
lastly is a for the Close target.

Figures 4.8, 4.9 and 4.10 show the beam profile vs time, the beam profile was measured by the rate of fusion-evaporation detected in a neutron monitor mounted outside and normalized using the total integrated current. In Figure 4.8, which shows the Far target that was inside the chamber for $\sim 385$ minutes, it can be seen that the beam was relatively stable. Though in Figure 4.9 which shows Blank beam profile, irradiated for $\sim 419$ minutes the current was stable near 13 nA. Lastly, we have Figure 4.10 which shows Close beam profile and this target was inside the chamber for $\sim 452$ minutes. The beam here was not stable.

![Figure 4.8 - Far target beam profile in a two dimensional plots with x-axis showing time (min) of irradiation and y-axis showing magnitude of the current in (nA).](http://etd.uwc.ac.za/)

4.5 Chemical Separation and Counting of Activated Samples

After irradiation at the 88-Inch cyclotron, the samples were transported to the low-background Nuclear Counting Facility (NCF) at the Lawrence Livermore National Laboratory (LLNL). The NCF is located one-and-half floors underground, with HPGe detectors surrounded in 10 cm of pre-WWII lead, minimizing contributions from environmental radiation. Each target was counted twice. The first count was
before chemical separation of gold isotopes and the second count was after chemical separation of the gold isotopes. The GAMANAL software was used [62, 63] to interpret gamma spectra, perform background subtractions, and perform peak fits, correct for detector efficiency, sample geometry, dead time, coincidence loss, and self-shielding of γ-rays in the samples.
The detectors are regularly calibrated against National Institute of Standards and Technology (NIST) traceable multi-energy point sources and large area distributed sources. The gold was chemically separated because of big background contributions from fusion-evaporation residues. Each pair of carbon samples and bismuth samples were placed in a 40 mL centrifuge cone and dissolved with 2 mL 9M HCl 6 drops 8M nitric acid. The samples were placed in a hot-water bath under air streams and evaporated to dryness. Each sample was dissolved in 3 mL 6M HCl, and 2 mL of ethyl acetate was added. The mixture was agitated for 90 seconds on a Vortex mixer. The phases were allowed to settle, and the organic phase was transferred to a second 40 mL cone. Another 2 mL ethyl acetate was added to the aqueous phase, and the agitation was repeated. The resulting organic phase was combined with the previous 1mL 6M HCl was added to the organic phase and the mixture was agitated for 60 seconds after which the aqueous phase was discarded. The clean organic phase, containing the gold activity, was evaporated to dryness under an air stream, dissolved in 1M HCl, and quantitatively transferred to a Prindle counting vial in 10 mL of dilute HCl.

The chemical separation improved the signal-to-background ratio by a factor ~100. The absolute efficiencies of separation, typically on the order of 90%, were determined individually for the foils by comparing the $^{196}$Au activity measured before and after separation. Besides gold, isotopes of gallium, molybdenum, and antinomy could not be completely removed.

### 4.6 Detector efficiency

The GAMANAL program did not include detectors efficiency. To find these efficiency some calculation were performed using the $\gamma$-ray lines, 204 keV, 214 keV and 412 keV lines which are due to $^{198}$Au and 148 keV, 188 keV, 285 keV, 316 keV, 333 keV, 356 keV and 426 keV which are due to $^{196}$Au. Figure 4.1 below shows the output file of the far target counted for 480min from the GAMANAL program.

The information in Table 4.1 shows a typical GAMANAL output file for the Far target. These results are to be used to calculate detector efficiency. For example
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<td>1.076E+02</td>
<td>-</td>
<td>10.79</td>
<td>0.827</td>
</tr>
<tr>
<td>19</td>
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<td>18204</td>
<td>18105</td>
<td>9.111E+02</td>
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<td>0.95</td>
<td>0.849</td>
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<td>1611</td>
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<td>6.80</td>
<td>0.833</td>
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<td>158.400</td>
<td>3855</td>
<td>3838</td>
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<td>7324</td>
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<td>3600</td>
<td>3975</td>
<td>2.110E+02</td>
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<td>1.133</td>
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<td>995</td>
<td>1008</td>
<td>4.059E+01</td>
<td>0.593</td>
<td>14.49</td>
<td>0.938</td>
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<td>7.667E+01</td>
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<td>27</td>
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<td>995</td>
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<td>9.71</td>
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<td>0.0508</td>
<td>10.69</td>
<td>0.880</td>
</tr>
</tbody>
</table>

Table 4.1 – GAMANAL out-put file for the Far target, counted for 480 min.

Table 4.1 shows the results of the efficiency determination for the HPGe XP/3 model detector using the method described in section 4.6 of Chapter 4. Figure 4.11 of the table below shows the energy of the γ rays in keV. The last two columns, illustrates the calculated efficiency of the HPGe XP/3 detector. This efficiency is actually the percentage of the γ rays which passed through the HPGe XP/3 model detector in the counting room after the activation of the samples.
Table 4.2 – The relative detector efficiency for HPGe XP/3 model determined using the Far target with 480 min counting time.

<table>
<thead>
<tr>
<th>keV</th>
<th>Average Counts</th>
<th>Photons/min</th>
<th>Eff. ($10^{-2}$)</th>
<th>ΔEff. ($10^{-4}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>65</td>
<td>4763</td>
<td>365</td>
<td>2.72</td>
<td>6.50</td>
</tr>
<tr>
<td>67</td>
<td>9964</td>
<td>727</td>
<td>2.86</td>
<td>6.82</td>
</tr>
<tr>
<td>83</td>
<td>9564</td>
<td>532</td>
<td>3.74</td>
<td>8.94</td>
</tr>
<tr>
<td>148</td>
<td>1619</td>
<td>82</td>
<td>4.10</td>
<td>9.79</td>
</tr>
<tr>
<td>181</td>
<td>3788</td>
<td>211</td>
<td>3.74</td>
<td>8.93</td>
</tr>
<tr>
<td>193</td>
<td>1031</td>
<td>61</td>
<td>3.54</td>
<td>8.46</td>
</tr>
<tr>
<td>204</td>
<td>1197</td>
<td>71</td>
<td>3.45</td>
<td>8.77</td>
</tr>
<tr>
<td>214</td>
<td>2189</td>
<td>137</td>
<td>3.34</td>
<td>7.97</td>
</tr>
<tr>
<td>285</td>
<td>772</td>
<td>61</td>
<td>2.65</td>
<td>6.32</td>
</tr>
<tr>
<td>316</td>
<td>1123</td>
<td>97</td>
<td>2.40</td>
<td>5.74</td>
</tr>
<tr>
<td>333</td>
<td>1681</td>
<td>153</td>
<td>2.29</td>
<td>5.47</td>
</tr>
<tr>
<td>412</td>
<td>16309</td>
<td>1797</td>
<td>1.89</td>
<td>4.52</td>
</tr>
<tr>
<td>426</td>
<td>314</td>
<td>36</td>
<td>1.83</td>
<td>4.38</td>
</tr>
<tr>
<td>437</td>
<td>256</td>
<td>30</td>
<td>1.79</td>
<td>4.28</td>
</tr>
</tbody>
</table>

Figure 4.11 – The HPGe XP/3 detector efficiency curve produced from GAMANAL output and its error bars.
Chapter 5

Numerical analysis and Results

5.0.1 Calculation of DIGS

In this section, we discuss the way to calculate the Double Isomer-to-Ground state ratio (DIGS) with the acquired data for this project. The calculation of the DIGS was not straightforward. For example, some amounts of thallium and iridium were detected in the samples with approximately the same chemical separation efficiency as a gold isotope [64]. The $^{198}$Tl decays by Electron Capture (EC) to $^{198}$Hg comparative to the $^{198}$Au. It produces the same $\gamma$-ray line but with the half-life of 5.3 hours. Moreover, the $^{198}$Au ground state is also fed by the decay of its 2.3-day high-spin isomer at 812 keV. To get a correct number of counts of $^{198}$Au a disentanglement must be performed between growth and decay of the isomer and ground state in order to get pure $^{198}$Au number of counts. The analysis of interfering $\gamma$-rays and numerous decay chains were performed using linear algebra to decouple direct production of a daughter nucleus from in growth due to a parent nucleus. The decay equations that govern the experiment are given by the following set of equations;

\[
\dot{N}_{iso} = -\lambda_{iso}N_{iso} + A.I(t) \tag{5.1}
\]

\[
\dot{N}_{ground} = -\lambda_{ground}N_{ground} + \lambda_{iso}N_{iso} + B.I(t) \tag{5.2}
\]

\[
\dot{N}_{iso} = -\lambda_{iso}N_{iso} \tag{5.3}
\]

\[
\dot{N}_{ground} = -\lambda_{ground}N_{ground} + \lambda_{iso}N_{iso} \tag{5.4}
\]
with $A$ and $B$ being relative production parameters, these two parameters contain, $\sigma$ the production cross-section and $N_0$ the number of target nuclei. $\lambda$ is the decay constant, $I(t)$ is the beam current, $N_{iso}$ is the number of isomeric state nuclei and $N_{ground}$ represent the number of ground state nuclei. Full solutions to the above four equations can be found in Appendix A.

Isomeric decay schemes of $^{196}$Au and $^{198}$Au are shown in Figures 5.1 and 5.3, respectively, their level scheme following $\beta$-decay of the ground state in Figures 5.2 and 5.4, respectively. The decay scheme overview of the half-life times and the possible spin-parity. With the vertical distance representing changes in energy, the energy levels of the nuclei are represented by horizontal lines, these energies are measured in units of keV.

![Diagram of isomeric decay scheme](http://etd.uwc.ac.za/)

**Figure 5.1** – $^{196}$Au (12$^-$) isomer level scheme with half-life of 9.6 hours [65].
Figure 5.2 – $^{196}\text{Au}$ ground state level scheme following $\beta^-$ decay with half-life of 6.2 days [65].

Figure 5.3 – $^{198}\text{Au}$ ($12^-$) isomer level scheme with half-life of 2.3 days [65].
Figure 5.4 – $^{198}$Au ground state level scheme following $\beta$-decay with half-life of 2.7 days [65].
Examples of Close and Far target $\gamma$-ray spectra for the $^{197}\text{Au}(^{13}\text{C}, ^{12}\text{C})$ and $^{197}\text{Au}(^{13}\text{C}, ^{12}\text{C}2n)$ products with the different targets are shown in the following Figures 5.5, 5.6, 5.7 and 5.8 with some $\gamma$-ray peaks from the after-chemistry spectra which were used to calculate the Ground-to-Isomer state ratios. The far target spectra used can be found in Appendix B.

![Figure 5.5](http://etd.uwc.ac.za)

**Figure 5.5** – Typical $\gamma$-ray spectra of the Close target before chemistry.

![Figure 5.6](http://etd.uwc.ac.za)

**Figure 5.6** – Typical $\gamma$-ray spectra of the Close target after chemistry.
Figure 5.7 – Typical γ-ray spectra of the Far target before chemistry.

Figure 5.8 – Typical γ-ray spectra of the Far target after chemistry.

Figure 5.5 and 5.7 shows the Close and Far target spectra before chemical separation whereas Figure 5.6 and 5.8 shows spectra after chemical separation. It can be seen that after chemical separation peaks that due to contamination were removed, for example most indicated γ-ray lines are due to the de-excitation of $^{196}$Au and $^{198}$Au. The chemical separation goal was to ensure that the irradiated
samples were containing gold isotopes only. For more details on how chemical separation was done see section 4.5. Using data from the National Nuclear Data Center (NNDC) website and Table of Isotopes [65] the peaks originating from $^{196m}$Au, $^{196g}$Au, $^{198m}$Au and $^{198g}$Au could be identified.

5.0.2 Spectra Analysis

The RadWare package [66] is useful for the graphic display and for the analysis of one-dimension spectra i.e. peak area determination. The GF3 package is one of the RadWare software packages. This package was used successfully to measure photo-peak areas from the $\gamma$-ray spectra for this work. Two different methods were used to measure the areas, the first method calculates the $\gamma$-ray spectral peaks integrated counts by fitting data with Gaussian functions. Photo-peak backgrounds were approximated using photo-peak-width broadened linear step functions. This background is then subtracted by the program. The second method fits the photo-peak from the $\gamma$-ray spectra by the Gaussian function, here the background subtraction is done automatically by the program. The measured number of decays that occurred during counting were then corrected for efficiencies (see section 4.6 on to how efficiency for each $\gamma$-ray transition was calculated) of the detector and the intensities of the $\gamma$-ray transitions given in Table 5.1.

Both Figures 5.9 and 5.10 show the region where the strong peaks that are originating from the $^{196}$Au and $^{198}$Au isotopes are found. For more spectra on the blank target see section B.1 in Appendix B. Various criteria were used one after the other on the marked (red labeled) $\gamma$-ray lines. To search for impurities, all the marked $\gamma$-ray lines were investigated from the neighboring isotopes of $^{196}$Au and $^{198}$Au. The neighboring isotopes should not have similar $\gamma$-rays lines as the one marked if some dose. The half-life and intensities were carefully checked as to make sure the $\gamma$ rays from the neighbors have no similar properties as the $\gamma$-rays in the $^{196}$Au and $^{198}$Au isotopes of interest. From these tests, very small amounts of Tl and Ir in their radiated samples were found. Moreover, the full-width-at-half-maximum height (FWHM) of a pulse height peak, which is used to measure the energy spread was also carefully checked when measuring peak areas of the marked $\gamma$-ray lines. The energy resolution of HPGe detectors is roughly 1.75 keV for $\gamma$-ray energies of 1.33 MeV. For any $\gamma$-ray line that gives an energy spread of greater than 1.75 keV we considered the possibility of the existence doublet containing photo peaks. For anything less than 1.75 keV we consider our detector resolution to be good, thus
we can measure the peak area and use it in the analysis. These checks also helped us to check for the overlaying of peaks during peak areas measurements.

**Figure 5.9** – Typical $\gamma$-ray spectra of the Close target after chemical separation zoomed in the region of isomeric decays for both $^{196}$Au and $^{198}$Au.

**Figure 5.10** – Typical $\gamma$-ray spectra of the Close target after chemical separation zoomed in the region of the isomeric and ground state decays for both $^{196}$Au and $^{198}$Au.
The γ-ray energies and intensities used in this work were taken from NuDat [65], a resource available currently through the National Nuclear Data Centers (NNDC) online archives and these γ-ray energies and intensities are listed in Table 5.1. It provides an interface between web users and several NNDC nuclear structures and decay databases. NuDat can be used to search for ground and excited state level properties, γ ray information, and decay radiation information. In addition to the search capabilities, an interactive chart of nuclei is displayed [65].

In Table 5.2 and 5.3 (more data can be found in Appendix C) gives the measured peaks and errors of shown γ-ray energies in Table 5.1, respectively, which were used for DIGS calculations. As seen from the Tables 5.2 and 5.3 only few γ-ray lines peaks were measured in both the gold isotopes i.e. only the 147 keV and 188 keV which are from the isomer state and 355 keV from the ground state of ¹⁹⁶Au were measured and in the ¹⁹⁸Au only the 204 keV, 215 keV which are from the isomeric state and 412 keV from the ground state. To correct for efficiency and intensity the areas of the peaks measured in the GF3 program were divided by efficiency together with the intensity given for each γ-ray line. Changes in irradiation, transport or counting conditions of the samples can introduce errors in the correct number of counts that will be used in the analysis to compute production rates values. To figure out the uncertainties in the number of counts, the error obtained for each peak area θ was used to compute the lower and upper error bands i.e. θ−σ for lower error band and θ+σ for upper error band, σ is the peak area error. These qualities were combined according to the error propagation method when calculating the production rate values, with the estimate uncertainties from detection time detector (± 86 seconds since the FORTRAN output rounding was to three decimal places) and for proton flux (0.5 nA was used).

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>Intensity(%)</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
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<td>¹⁹⁰mAu</td>
<td>43.5</td>
<td>147.81(2)</td>
</tr>
<tr>
<td>¹⁹⁰mAu</td>
<td>30.0(15)</td>
<td>188.27(3)</td>
</tr>
<tr>
<td>¹⁹⁸mAu</td>
<td>39(5)</td>
<td>204.10(6)</td>
</tr>
<tr>
<td>¹⁹⁸mAu</td>
<td>77.3</td>
<td>214.85(5)</td>
</tr>
<tr>
<td>¹⁹⁶Au</td>
<td>87</td>
<td>355.73(5)</td>
</tr>
<tr>
<td>¹⁹⁸Au</td>
<td>95.62</td>
<td>411.80(17)</td>
</tr>
</tbody>
</table>

Table 5.1 – Intensities and γ-ray lines for the ¹⁹⁶Au and ¹⁹⁸Au isotopes reported by NuDat Ref. [65].
<table>
<thead>
<tr>
<th>Count.(h)</th>
<th>keV</th>
<th>Decays</th>
<th>Decays $\text{lower}$</th>
<th>Decays $\text{upper}$</th>
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<td>1171703</td>
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</table>

Table 5.2 – $^{196m,196}$Au decays for the blank target, first column shows the counting time by the detector for each gamma line in units of hours. The number of counts which were measured from the blank target used foils for the $^{196}$Au isotopes were corrected for intensity and efficiency. The tables also show lower and upper error bands of the exactly measured decays. Decays for other different foils can be found in Appendix C.

In order to make sure that correct $\gamma$ rays were used, the corrected number of counts were used. It can be seen from Tables 5.2 and 5.3 that the counts are increasing as the counting time in the detector increases. The result were then plotted against the time. Figure 5.11 shows the decay graph of the corrected counts from Table 5.3 for the 215 keV line.

![Graph showing decay curve of corrected counts](http://etd.uwc.ac.za/)

Figure 5.11 – $^{198}$Au blank decay curve for $\gamma$ line 215 keV.
Table 5.3 – $^{198m,198g}$Au decays for the blank target, first column shows the counting time by the detector for each gamma line in units of hours. The number of counts which were measured from the blank target used foils for the $^{198}$Au isotopes were corrected for intensity and efficiency. The tables also show lower and upper error bands of the exactly measured decays. Decays for other different foils can be found in Appendix C.

<table>
<thead>
<tr>
<th>Count (h)</th>
<th>keV</th>
<th>Decays</th>
<th>Decays _{lower}</th>
<th>Decays _{upper}</th>
</tr>
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<td>311908</td>
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</tr>
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<tr>
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<td>237053</td>
</tr>
<tr>
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</tr>
<tr>
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</tr>
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</tr>
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<td>412</td>
<td>3981218</td>
<td>3860250</td>
<td>4108899</td>
</tr>
</tbody>
</table>

Theoretically the decay curve obeys the following equation,

\[ f = b \exp(-\lambda x), \]  

where \( b \) stands for the initial amount at time \( t \) and \( \lambda \) is the decay constant. Equation 5.5 was fitted using SigmaPlot [67] (a graphing and statistical analysis software package) together with the data in Table 5.2 and the value of \( b \) and \( \lambda \) was found to be 142766 ± 4797 counts and 0.0123 ± 0.0010 /h respectively. We can express the half-life, \( t_{1/2} \), in terms of the decay constant as follows,

\[ t_{1/2} = \ln(2)/\lambda \]  

Using the obtained value of \( a \) and plugging it into eq. 5.6 the half-life was found to be \( t_{1/2} = 2.33 \pm 0.08 \) days. The experimentally known value \( t_{1/2} \) for the isomer state in $^{198}$Au (12−) is 2.27 ± 0.02 day [65]. It can be easily see that the calculated value is in agreement with the experimentally value, hence the values can be used in the analysis process. These decay curves were done for all other targets in order to verify photo-peak transition assignments.
To solve the Equations 5.1, 5.2, 5.3 and 5.4 the corrected number of counts for both $^{196}$Au and $^{198}$Au were labelled as $D_{196m}$, $D_{196g}$, $D_{198m}$ and $D_{198g}$ with m and g standing for isomer and ground state.

Using the data to produce some relative production rates of the isomer and the ground states. There are three different times of interest for this experiment. Let $t_b$ be the time beam was shut off and $t_c$ be the time when counting started and $t_f$ be the final time when counting stops. For the experiment the well-known net rate of production-activated nuclei are given by the equations 5.1 and 5.2 from $t = 0$ to $t = t_b$ and Equation 5.3 with 5.4 from $t = t_b$ and $t = t_f$. These are coupled differential equations where $N_{iso}$ decays into $N_{ground}$; thus $\lambda_{iso}N_{iso}$ is in equation 5.4; for $0 < t < t_b$ is the time of production.

This production is dependent on several factors but in the simplest case, the only factor that was non-constant was the beam current $I(t)$. Information regarding the beam current was acquired from the beam current reader from the control room. From this we now have $I(t)$ and what is unknown from the equations is $A$ and $B$ which are to be solved. To do this we need to calculate backwards in time from the end time $t_f$ all the way to $t_0$.

The corrected number of decays $D_{196m}$, $D_{196g}$, $D_{198m}$ and $D_{198g}$ were measured from $t_c < t < t_f$. However the differential equations describe population. The first step of the solution to this problem is to calculate which population at $t = t_c$ would give the observed amount of decays during $t_c < t < t_f$. In order to perform the calculation the Runge Kutta 34 Method described in Appendix A was used; $f(t, y)$ will be the right hand side of equations 5.1, 5.2, 5.3 and 5.4, with the information from the control room our time step $h$ for beam current was set to 1 minute.

## 5.1 Testing the code

To guarantee the nature of the created code, it is important to verify the correctness of the code. So to verify the software utilized, the code must be tried on theoretical predicted production rates keeping in mind the end goal to have full control over the inputs. This is the best way to be predict the consequence of the outputs and to see whether the functionality of the code give predictable outputs. To do this test, consider the accompanying simulation to test our code for this project. Suppose we were activating an X sample with the following reaction X (n,
γ) Y for a minimum time of $t_{irradiation}$ hours. Let’s say after irradiation the sample is transported to the counting facility and we further assume this transportation takes $t_{transport}$ hours before it reaches the counting facility. Furthermore, assume the sample arrives at the counting facility and the counting is done with detector and a spectrum is accumulated for a counting time of some hours, long enough to get reasonable statistics for the counts in the photo-peaks. By measuring the counts in the peak of interest from the obtained spectrum i.e. γ-ray lines of the decay of some $^m$Y isomeric and $^g$Y ground state. Furthermore, suppose the relative intensities of the measured γ-ray lines are known and the absolute efficiency of the used detector is also known. Then the number of decayed nuclei can be expressed by the following formula:

$$ D = \frac{C}{\epsilon \times I}, $$
(5.7)

where D is either for isomeric state or ground state, C is number of counts, $\epsilon$ is the γ-ray detection efficiency of the detector and I is short for intensity of the detected γ-ray. From the decay equations 5.8 and 5.9, $N_0$ can be calculated,

$$ N_{iso} = A I(t) - \lambda_{iso} N_{iso}(t), $$
(5.8)

$$ N_{ground} = B I(t) - \lambda_{ground} N_{ground}(t) + \lambda_{iso} N_{iso}(t), $$
(5.9)

where I(t) is the beam current recorded during irradiation time, $N_{iso,ground}$ are number of isomer and ground state nuclei at some time, t. The production rates A and B are given by the following relation:

$$ A, B = N_0 \Delta X \sigma $$
(5.10)

where $N_0$ is the number of target nuclei after the radiation stopped at time $t = t_0$, \(\Delta X\) is the thickness of the target and $\sigma$ is the cross section of the reaction. Assume $\Delta X$ and $\sigma$ are known, and the time intervals $[0, t_{irradiation}]$, $[t_{irradiation}, t_{transport}]$ and $[t_{transport}, t_{counting}]$ have been recorded. Then to solve for $N_0$ of the Y isotopes we need to solve the given decay equations 5.8 and 5.9 from $t_{counting}$ back to $t_0$. To test the MATLAB code in section D.1 of Appendix D if its working properly the two decay equations 5.8 and 5.9 were solved using Excel. The production rates
Testing parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D_{iso}$</td>
<td>$1.000 \times 10^5$</td>
</tr>
<tr>
<td>$D_{ground}$</td>
<td>$8.000 \times 10^5$</td>
</tr>
<tr>
<td>$\lambda_{iso}$</td>
<td>$7.702 \times 10^{-4} \text{ min}^{-1}$</td>
</tr>
<tr>
<td>$\lambda_{ground}$</td>
<td>$9.623 \times 10^{-5} \text{ min}^{-1}$</td>
</tr>
<tr>
<td>$t_{irradiation}$</td>
<td>$420 \text{ min}$</td>
</tr>
<tr>
<td>$t_{transport}$</td>
<td>$1500 \text{ min}$</td>
</tr>
<tr>
<td>$t_{counting}$</td>
<td>$3600 \text{ min}$</td>
</tr>
</tbody>
</table>

Table 5.4 – The Excel and MATLAB parameters that were assumed and used to test the code

were obtained by the Excel Solver (see Appendix E example of the spreadsheet used) program, these decay equations were rewritten in the following form:

$$N_{iso}(i + 1) = N_{iso}(i)\left[\lambda_{iso}N_{iso}(i) + AI(t(i))\right]dt$$  \hspace{1cm} (5.11)

$$N_{ground}(i + 1) = N_{ground}(i)\left[\lambda_{ground}N_{ground}(i) + \lambda_{iso}N_{iso}(i) + BI(t(i))\right]dt$$  \hspace{1cm} (5.12)

for $i = 1, 2, 3$ etc. Table 5.4 shows the assumed values of decays of the Y isotopes, the irradiation, transport and detector counting times including the decay constants that were assigned for the some isomeric state and ground state. During irradiation time the beam current was assumed to be $\approx 17 \text{ nA}$. The ground state half-life was assumed to be $5 \text{ d}$ and isomeric state to be $15 \text{ h}$. These values were used for both the MATLAB code testing and in Excel.

Figure 5.12 shows the graphical illustration of the isomer and ground state population after MATLAB and Excel output results by inputting the values in Table 5.4.

The solid and dotted black lines are the ground state population and the red solid and dotted lines are for the isomer population. Excel data points seem to lie below the MATLAB data points during the decay period after the stoppage of the irradiation of the sample. This difference is due to factors such as the numerical method used i.e. MATLAB function for the 4th Runge Kutta Method was used (see RK34 described well in Appendix A Section A.2). This method is more precise than the numerical way used in the Excel Solver. After using different cases in the decay lifetimes, both the shapes of MATLAB and Excel were behaving the same way as shown in Figure 5.12. The resulting production rates (A, B) of these cases are represented by Table 5.5.
Chapter 5 Numerical analysis and Results

Figure 5.12 – Graphical results of the MATLAB and Excel Solver of the input parameters in Table 5.4 where $D_{iso}$ and $D_{ground}$ have half life of 15 hours and 5 days respectively.

| Production rates | MATLAB code | Excel Solver | Decay constant $(min^{-1})$
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_1$</td>
<td>59.65</td>
<td>48.34</td>
<td>$\lambda_{ground} &lt; \lambda_{iso}$</td>
</tr>
<tr>
<td>$B_1$</td>
<td>423.29</td>
<td>423.43</td>
<td></td>
</tr>
<tr>
<td>$A_2$</td>
<td>48.38</td>
<td>46.79</td>
<td>$\lambda_{ground} &gt; \lambda_{iso}$</td>
</tr>
<tr>
<td>$B_2$</td>
<td>372.88</td>
<td>305.87</td>
<td></td>
</tr>
<tr>
<td>$A_3$</td>
<td>39.43</td>
<td>37.00</td>
<td>$\lambda_{ground} \approx \lambda_{iso}$</td>
</tr>
<tr>
<td>$B_3$</td>
<td>285.70</td>
<td>270.38</td>
<td></td>
</tr>
</tbody>
</table>

Table 5.5 – The production rate (dimensionless) values after three cases of changing the values of $\lambda_{ground}$ together with $\lambda_{iso}$.

5.2 Algorithm to solve the decay equations

5.2.1 For $t_c < t < t_f$

Since nothing was feeding the isomer the population is easily translated into decays.

A developed MATLAB code in Appendix D was used, which iterates until the calculated decays matched the observed decays. The population of $N_{ground}$ is not straightforward since $N_{iso}$ is feeding it. It is then harder to relate the observed decays to a population. Because $N_{iso}(t_c)$ is determined, the number of decays that is due to the feeding effect can be obtained by solving equations 5.3 and 5.4 on the time interval $t_c < t < t_f$ with the initial conditions that $N_{iso}(t_c) = N_{iso}^*(t_c)$.
(which is the previously computed) and $N_{iso}(0) = 0$. The decays that stem from
the feeding effect is then given by $PartDecay = (N_{iso}^*(t_c) - N_{iso}^*(t_f)) - N_{ground}(t_f)$. Similarly the decays without feeding are given by the solution to equation 5.3 and
5.4 with the initial conditions that $N_{iso}(0) = 0$ and $N_{ground}(t_c)$ is some value. The
decays from these sets of equation are $PureDecay = N_{ground}'(t_b) - N_{ground}'(t_f)$. The
prime and the notation is to point out that they are the solution to the described
equations and they are not the same. As before the initial values (in this case
$N_{ground}(t_c)$) is iterated until it is correct and $D_{19X} = PartDecay + PureDecay$.

5.2.2 For $t_b < t < t_c$

Using the calculated population at $t_c$, it is the easy to once again apply the Runge
Kutta method and get the population at $t_b$ through iteration (by iterating over
values for $N(t_b)$ which result in the correct $N(t_c)$). Now the last step that remains
is to compute value of the production rates $A$ and $B$ in equation 5.1 and 5.2.

5.2.3 For $0 < t < t_b$

As before $A$ and $B$ were given through iteration. The population at $t_b$ is known,
the initial population is known (it is 0) and $I(t)$ is known. As previous steps,
using equation 5.1 and 5.2 iteration (changing $A$ and $B$) until this calculated
population matches the previously computed population at $t_b$. When they match
then $A$ and $B$ has been found hence the problem is solved entirely.

5.3 MATLAB results

The MATLAB code from Appendix D was used for calculating the $A$ and $B$ pro-
duction rate, with given number of decays i.e. the corrected counts in Table 5.2, 5.3
and in Appendix C. Figure 5.13, 5.14 and 5.15 are example of how the population
can vary over time when the current is not constant. The results seen are for $^{196}$Au
in the Blank, Close and Far targets (for $^{198}$Au see section D.2 in Appendix D). All
the counts used to produce these plots were counted over a maximum time of 48
hours using different detectors.

http://etd.uwc.ac.za/
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Figure 5.13 – Plot of population over time for $^{196}$Au in the Blank target. The red line is the population of the isomer and the blue line is the population of the ground state.

Figure 5.14 – Plot of population over time for $^{196}$Au in the Close target. The red line is the population of the isomer and the blue line is the population of the ground state. The sudden change in population at 250 - 350 min is when the beam shut off.
Figure 5.15 – Plot of population over time for $^{196}$Au in the Far target. The red line is the population of the isomer and the blue line is the population of the ground state.
Chapter 6

Discussion and Conclusions

6.1 Discussion

6.1.1 Isomer and ground state production rates

<table>
<thead>
<tr>
<th>Target</th>
<th>( E_{\text{isomer}} ) (keV)</th>
<th>A</th>
<th>B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blank 148/356</td>
<td>276.4^{+4.6}_{-5.5}</td>
<td>728.7^{+12.0}_{-14.7}</td>
<td></td>
</tr>
<tr>
<td>Blank 188/356</td>
<td>387.4^{+4.8}_{-2.1}</td>
<td>622.7^{+19.6}_{-21.7}</td>
<td></td>
</tr>
<tr>
<td>Close 148/356</td>
<td>78.2^{+30.3}_{-19.3}</td>
<td>238.3^{+29.4}_{-8.7}</td>
<td></td>
</tr>
<tr>
<td>Close 188/356</td>
<td>1503^{+10.5}_{-6.5}</td>
<td>174.9^{+24.5}_{-5.4}</td>
<td></td>
</tr>
<tr>
<td>Far 148/356</td>
<td>25.5^{+2.5}_{-3.0}</td>
<td>680.6^{+13.2}_{-15.1}</td>
<td></td>
</tr>
<tr>
<td>Far 188/356</td>
<td>395.4^{+3.5}_{-6.4}</td>
<td>540.9^{+12.3}_{-24.0}</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.1 – \(^{196}\text{Au}\) Isomer and ground state production rates (dimensionless).

Table 6.1 and 6.2 contain the production rates, \(A\) and \(B\) with the lower and upper error values. These production rates are the MATLAB derived values. The MATLAB results were used because the software could easily correct for the feeding of ground state population from the isomeric state. Included are the specific target foil that each of these individual values was extracted from and also the \(\gamma\)-ray lines that are due to isomeric and ground states. As seen from the \(RDIGS\) plot in Figure 6.1, the value of \(A\) is much less than that of \(B\) for all the iterations. The value of \(A\) represents the isomer population and \(B\) that of
Table 6.3 – $^{196}$Au Double-isomer to ground state ratios with its lower ($R_{DIGS-}$) and upper ($R_{DIGS+}$) error values for the three different targets.

<table>
<thead>
<tr>
<th>Target</th>
<th>$R_{DIGS}$</th>
<th>$R_{DIGS-}$</th>
<th>$R_{DIGS+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C/B</td>
<td>0.68</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td>C/B</td>
<td>0.68</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td>C/F</td>
<td>0.53</td>
<td>0.02</td>
<td>0.01</td>
</tr>
<tr>
<td>C/F</td>
<td>2.00</td>
<td>0.10</td>
<td>0.10</td>
</tr>
<tr>
<td>B/F</td>
<td>0.79</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td>B/F</td>
<td>1.40</td>
<td>0.05</td>
<td>0.04</td>
</tr>
</tbody>
</table>

Table 6.4 – $^{198}$Au Double-isomer to ground state ratio with its lower ($R_{DIGS-}$) and upper ($R_{DIGS+}$) error values for the three different targets.

<table>
<thead>
<tr>
<th>Target</th>
<th>$R_{DIGS}$</th>
<th>$R_{DIGS-}$</th>
<th>$R_{DIGS+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C/B</td>
<td>0.68</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td>C/B</td>
<td>0.68</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td>C/F</td>
<td>0.53</td>
<td>0.02</td>
<td>0.01</td>
</tr>
<tr>
<td>C/F</td>
<td>2.00</td>
<td>0.10</td>
<td>0.10</td>
</tr>
<tr>
<td>B/F</td>
<td>0.79</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td>B/F</td>
<td>1.40</td>
<td>0.05</td>
<td>0.04</td>
</tr>
</tbody>
</table>

the ground state, see the MATLAB plots in Figure 5.13, 5.14 and 5.15 (also see section D.2 in Appendix D) the isomer population curve always lies below that of the ground state population curve. The difference indicates that the production of the isomer population is always less than that of the ground state population.

Table 6.3 and 6.4 contains the double isomer-to-ground state ratios ($R_{DIGS}$) for $^{196}$Au and $^{198}$Au isotopes, respectively. Included in them are the lower and upper error values. The obtained $^{196}$Au and $^{198}$Au isotope production rates values were used to compute the $R_{DIGS}$ values. The $R_{DIGS}$ values were carefully computed.
with their lower and upper error values using the production rates of Close over Blank target foils, Close over Far target foils and last Blank over Far target foils. The chronological order used here to calculate the ratios is based on the theory of $R_{DlGS}$. A non-unitary value of the double isomer-to-ground state of Close over Far may indicate NPIs impact [5]. The last step in the chronological order is Blank divided by Far target foils. The reasons behind is that in the blank target the $^{197}$Au beam was exposed to the ordered plasma environment (no thin carbon foil present). It is worth to see also section 5.1 for parameters which are contained in the production rates $A$ and $B$. The other reason is that the $\gamma$-ray lines that are due to $^{196m,g}$Au and $^{198m,g}$Au were found in the data collected for this foil. This indicated a high number of nucleon-transfer reactions between $^{197}$Au and $^{209}$Bi. Most likely so high that ($^{13}$C, $^{12}$C) and ($^{13}$C, $^{12}$C2n) are small in comparison.

![Figure 6.1](http://etd.uwc.ac.za/)

**Figure 6.1** – Plot of the double isomer-to-ground state ratios ($R_{DlGS}$) for both the $^{196}$Au and $^{198}$Au isotopes for various targets. Along y-axis we have the $R_{DlGS}$ values and along the x- axis we the regions of the different permutation of division of the targets. C is short for Close target, F is short for Far target and B is short for Blank target.

The tabulated $R_{DlGS}$ values shown in Tables 6.3 and 6.4 were plotted on the same xy-plane see Figure 6.1. The experimental calculated $R_{DlGS}$ values of $^{198}$Au
isotope for the Close over Far and Blank over Far are quite off in range when compared with the \( R_{\text{DIGS}} \) values of \(^{196}\text{Au}\) isotope except for one case in the region of C/B labeled on the x axis. The \( R_{\text{DIGS}} \) values for \(^{196}\text{Au}\) are relatively closer to the standard unity value, which suggests that the nucleus of the produced isotope may not have been affected by the presented ordered plasma environment. A plausible explanation for the difference of the \(^{198}\text{Au}\) isotope \( R_{\text{DIGS}} \) value, is that the \(^{198}\text{Au}\) is fed by its high-spin isomeric state (\( t_{1/2} = 2.3 \text{ d} \) and spin \( (12^-) \)) with a 812 keV \( \gamma \)-ray transition. This gives a clear signature that the data collected for the \(^{198}\text{Au}\) should not be considered in our analysis, hence this gave a focus towards the collected data of the \(^{196}\text{Au}\) isotope. Further, some small contribution from the ground state with half-life \( t_{1/2} = 2.7 \text{ d} \) and spin \( (2^-) \) cannot be determined from the observation of the 412 keV \( \gamma \) line, because the energy of this line coincide with that of \(^{198}\text{Tl}\) this is was discussed in the analysis section.

### 6.1.2 Determination of the Coulomb Excitation effects

However, to make sure that the result for the \(^{196}\text{Au}\) isotope were suitable to use. Further investigation was done for possible effects that may have played a role during the reaction of the product with the electron plasma that was created using \(^{209}\text{Bi}\). Coulomb excitation effects were suspected to have possibly played some role. However, the feasibility of Coulomb excitation in a nuclear reaction is decided by the Coulomb barrier \([68]\). Coulomb barrier calculations were performed using the online calculator \([69]\), for the three different projectiles (see Table 6.5) with the \(^{209}\text{Bi}\) as a target. The first two projectiles were the ones produced after the two different reaction with the thin \(^{13}\text{C}\) target (see experimental design in section 3.3) the isotopes produced were the \(^{196}\text{Au}\) and \(^{198}\text{Au}\), respectively.

Moreover, the two produced isotopes were assumed to still have an energy of 1.2 or 1.4 GeV compared to the 1.7 GeV of \(^{197}\text{Au}\). This assumption is based on the effects that are described in chapter 2, which suggests different ways on how the beam loses energy when passing through a material. The last projectile was the \(^{197}\text{Au}\) straight to the plasma environment (Blank target, no thin carbon target presented) with the energy of 1.7 GeV. The obtained Coulomb barrier \( (V_c) \) values obtained are listed in Table 6.5. It can be seen from the table that the average value of \( V_c \) is 670 MeV, with beam energies used, can see that the energy was always well above the Coulomb barrier. This indicates that there were more nuclear contributions or effects between the projectile and the target rather than
solely the electromagnetic interactions. Thus Coulomb excitation effects play such a minor role in the reactions and can be neglected.

<table>
<thead>
<tr>
<th>Projectile</th>
<th>Target (ordered plasma)</th>
<th>$V_c$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{196}$Au</td>
<td>$^{208}$Bi</td>
<td>670</td>
</tr>
<tr>
<td>$^{197}$Au</td>
<td>$^{209}$Bi</td>
<td>669</td>
</tr>
<tr>
<td>$^{198}$Au</td>
<td>$^{209}$Bi</td>
<td>669</td>
</tr>
</tbody>
</table>

Table 6.5 – The table represents the Coulomb barrier ($V_c$) for three reactions of the gold isotope as the projectile with the target plasma made of $^{209}$Bi.

6.2 Conclusions

The essential aim of this thesis was to produce a code to disentangle isomer and ground state population, which will solve the sophisticated decay chains that were involved in the experiment. A MATLAB code has been created which solves the decay equation and gives out results of the production rates with the beam current not necessarily stable. The code likewise considers the feeding of the ground state from the isomeric state given by subtracting this contribution of isomeric states from the ground state number of decays. This is critical in light of the fact that the code provides the production rates output of the ground state population. The resultant production rates by the code were used to calculate the double-isomer-to-ground state ratios $R_{DIGS}$. The $R_{DIGS}$ is introduced as a signature for detecting angular momentum distribution changes due to Nuclear Plasmas Interactions (NPIs) on highly excited states prior to gamma emission.

The $R_{DIGS}$ were acquired for the two different gold isotopes $^{196}$Au and $^{198}$Au where for the $^{198}$Au $R_{DIGS}$ can be discarded due to the possible contaminants in the $\gamma$-ray spectrum as presented in the discussion section. The obtained $^{196}$Au $R_{DIGS}$ values deviate substantially from unity but there are large uncertainties due to the observation of a significant number of nucleon-transfer reactions on the bismuth target foil which appear to produce the isotopes of interest quite significantly. Moreover, due to the signal that was investigated which is expected to be very small to those produced in nucleon-transfer reactions on the bismuth, it prohibited an examination of the angular momentum distribution for the data obtained.

Further work that can be done in the future to improve the results of the $R_{DIGS}$ such as to lower the beam energy below the Coulomb barrier of Bi. But lowering the energy will not produce a $12^-$ spin state [70]. Additionally, a heavier target
can be used but this is not practical because even for $^{238}$U the Coulomb barrier is only 726 MeV which is below the beam energy. The method used here of $R_{DIGS}$ is an intense technique for trying to figure out the NPIs effects on highly excited nuclei. In this regime, NPIs impacts are little to identify. Along these lines, further advanced experimental approaches treating the nuclear excitation mechanism in contact with ordered plasma environment presented needs to be developed. Strategies that would likewise permit little mistakes and have the capacity to disentangle angular momentum distribution effect impact because of NPIs.
Appendix A

A.1

Analytical solutions to the decay and growth equations

The number of particles remaining at time $t$ is governed by the following decay law equations for different conditions.

**Isomer**

\[
\dot{N}_{iso} = -\lambda_{iso}N_{iso} \tag{A.1}
\]

where $\lambda_{iso}$ is the decay constant for the isomeric state, and this constant is $\propto t_{1/2}$ (half-life). This equation can be easily integrated to yield the following equation.

\[
N_{iso}(t) = N_0 e^{-\lambda_{iso}t} \tag{A.2}
\]

**Ground state**

\[
\dot{N}_g = -\lambda_gN_g + \lambda_{iso}N_{iso} \tag{A.3}
\]

rearranging the above equation as follows,

\[
\dot{N}_g + \lambda_gN_g = \lambda_0N_{iso} \tag{A.4}
\]

multiplying by a integrating factor $e^{\lambda_g t}$, yield:

\[
\dot{N}_g e^{\lambda_g t} + \lambda_gN_g e^{\lambda_g t} = \lambda_{iso}N_{iso} e^{-\lambda_{iso} t} e^{\lambda_g t} \tag{A.5}
\]
noting the above that is a product rule, furthermore we use the separation of variables and integration which the gives the following results:

\[ e^{-\lambda_g t}N_g(t) - N_g^0 = \frac{\lambda_{iso}}{\lambda_g - \lambda_{iso}} N_{iso}[e^{(\lambda_{iso} - \lambda_g)t} - 1] \] (A.6)

where \( N_g^0 \) is the ground state initial amount multiplying both sides by \( e^{-\lambda_g t} \),

\[ N_g(t) = N_g^0 e^{-\lambda_g t} + \frac{\lambda_{iso}}{\lambda_g - \lambda_{iso}} N_{iso}[e^{-\lambda_{iso} t} - e^{-\lambda_g t}] \] (A.7)

Assuming the half-life of the isomeric state is less that the half life of the ground state, the overall result is that the number of nuclei of the isotope will decrease exponentially according to its own half life. The following two equations cannot be solve analytical.

\[ \dot{N}_{iso} = -\lambda_{iso} N_{iso} + A.I(t) \] (A.8)

\[ \dot{N}_{ground} = -\lambda_{ground} N_{ground} + \lambda_{iso} N_{iso} + B.I(t) \] (A.9)

where \( A \) and \( B \) are production rates containing the number of target nuclei after irradiation time, the thickness of the target and cross-section of the reaction, \( I(t) \) is the proton flux. For the above two equations, if \( I(t) \) is constant the two can be solved in a similar way as the first two equations [71]. Otherwise, if \( I(t) \) keeps changing then a numerical approach is the best tool to use, an example can be seen from figure 13, 14 and 15 which shows \( I(t) \) vs \( t \). It can be easily spotted that \( I(t) \) is not a simple function as the beam current is fluctuating, hence a numerical approach is needed to be used.

A.2

Numerical solutions to the decay and growth equations

The 4th order Runge Kutta (RK34) numerical method is suitable to solve the above 4 decay and growth equations.

Runge Kutta In general, consider the following equation where the right-hand side is a function of time and the left-hand side is another function that depends on time.
Here the task is to solve the above differential equation, that is; given the function \( f(t, y) \) find the function \( y(t) \) such that it passes through the point \((t_0, y_0)\), whose derivative is the given function. The RK34 formulation is as follows \([72]\):

\[
y_{rk1} = f(t_n, y_n) \tag{A.11}
\]

\[
y_{rk2} = f(t_n + \frac{h}{2}, y_n + \frac{h}{2}y_{rk1}) \tag{A.12}
\]

\[
y_{rk3} = f(t_n + \frac{h}{2}, y_n + \frac{h}{2}y_{rk2}) \tag{A.13}
\]

\[
z_{rk3} = f(t_n + h, y_n - hy_{rk1} + 2hy_{rk2}) \tag{A.14}
\]

\[
y_{rk4} = f(t_n + h, y_n - hy_{rk3}) \tag{A.15}
\]

\[
y_{n+1} = y_n + 6y_{rk1} + 2y_{rk2} + 2y_{rk3} + y_{rk4} \tag{A.16}
\]
Appendix B

B.1

Blank and Far spectra

Figure B.1 – Typical $\gamma$-ray spectrum of the Blank target before chemistry.
Appendix B

Figure B.2 – Typical γ-ray spectrum of the Blank target after chemistry.

Figure B.3 – Typical γ-ray spectrum of the Blank target after chemical separation zoomed in the region of isomeric decays for both $^{196}\text{Au}$ and $^{198}\text{Au}$. 
Figure B.4 – Typical γ-ray spectrum of the Blank target after chemical separation zoomed in the region of isomeric decays and ground states for both $^{196}$Au and $^{198}$Au.

Figure B.5 – Typical γ-ray spectrum of the Far target after chemical separation zoomed in the region of isomeric decays and ground states for both $^{196}$Au and $^{198}$Au.
Figure B.6 – Typical γ-ray spectrum of the Far target after chemical separation zoomed in the region of isomeric decays and ground states for both $^{196}\text{Au}$ and $^{198}\text{Au}$. 
Appendix C

C.1

Intensity and efficiency corrected number of counts

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<th>Decays_{lower}</th>
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Table C.1 – $^{198}$Au intensity and efficiency corrected counts for the close target.
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Table C.2 – $^{198}$Au intensity and efficiency corrected counts for the far target.

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Table C.3 – $^{196}$Au intensity and efficiency corrected counts for the close target.
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Table C.4 – $^{196}$Au intensity and efficiency corrected counts for the far target.
Appendix D

D.1 MATLAB source code

```
1 close all
2 clc
3
4 %entering the parameters
5 %Nh duringI, Nn and N are the functions that needs modification. Enter the
6 %half lives in each of these. Also note that a current must be entered, see
7 further down in the code.
8
9 variables;
10
11 Decay1=isomer; % Enter measured decays. 1 is isomer, 2 is ground
12 Decay2=ground;
13 CalcDecay1=0;
14 CalcDecay2=0;
15 N1=Decay1;
16 N2=Decay2;
17 N1tb=0;
18 N2tb=0;
19 N1high=1000*N1;
20 N2high=1000*N2;
21
22 %enter decay constant in /h or /m or /s
23
24 thalf_1=%half life of an isomer
25 thalf_2=%half life of a ground
26 lambda1=log(2)/thalf_1;
27 lambda2=log(2)/thalf_2;
28
29 %Enter values for times
30
31 tend=% counting time in hours
32 tcount=% time between the beam shut off and counting began
33 tbeam=% Time the beam was active.
34
35 while abs(Decay1-CalcDecay1) > 1
36    [t,y]=RK34(@Nn,N1,tcount,tend);
37    CalcDecay1=(N1-y(1,end));
```
if abs(Decay1 - CalcDecay1) > 1
    if CalcDecay1 > Decay1
        N1high = N1;
        N1 = N1/2;
    else
        N1 = N1 + (N1high - N1)/2;
    end
end

[t2, y2] = RK34(@N, [N1; 0], tcount, tend);
PartDecay2 = N1 - y2(1, end) - y2(2, end); % Calculates the isomer’s contribution to the
groundstate’s decays.

while abs(Decay2 - CalcDecay2) > 1
    [t4, y4] = RK34(@N, [0; N2], tcount, tend);
    CalcDecay2 = PartDecay2 + (N2 - y4(2, end));
    if abs(Decay2 - CalcDecay2) > 1
        if CalcDecay2 > Decay2
            N2high = N2;
            N2 = N2/2;
        else
            N2 = N2 + (N2high - N2)/2;
        end
    end
end

[tc, yc] = RK34(@N, [N1; N2], tcount, tend);

N1tb = 1000 * N1;
N2tb = 1000 * N2;
N1tc = 0;
N2tc = 0;
N1high = 10 * N1;
N2high = 10 * N2;

while abs(N1 - N1tc) > 1
    [tl, yl] = RK34(@Nn, N1tb, 0, tcount);
    N1tc = yl(1, end);
    if abs(N1 - N1tc) > 1
        if N1tc > N1
            N1high = N1tb;
            N1tb = N1tb/2;
        else
            N1tb = N1tb + (N1high - N1tb)/2;
        end
    end
end

while abs(N2tc - N2) > 1
    [tk, yk] = RK34(@N, [N1tb; N2tb], 0, tcount);
    N2tc = yk(2, end);
    if abs(N2tc - N2) > 1
        if N2tc > N2

N2high = N2tb;
N2tb = N2tb /2;
else
N2tb = N2tb + (N2high - N2tb) /2;
end
end
end

d = RK34(@(N,[N1tb;N2tb]),0,tcount);
A=N1tb; % Production rates and initial values that are not true (to start the iteration).
B=N2tb;
Ahigh=N1tb;
Bhigh=N2tb;
CalcN1tb=0;
CalcN2tb=0;
CalcA=0;
CalcB=0;

% Enter the current information as a vector, time step is 1 sec.
load .txt; % .mat .xlsx etc.
Current = ; % use the loaded current file

while abs(N1tb-CalcN1tb) > 1
    [~,yi]=RK34(@(NduringI,[0;0]),0,tbeam,A,B,Current);
    CalcN1tb=yi(1,end);
    if abs(N1tb-CalcN1tb) > 1
        if CalcN1tb > N1tb
            Ahigh=A;
            A=A/2;
        else
            A=A+(Ahigh-A)/2;
        end
    end
end

while abs(N2tb-CalcN2tb) > 1
    [~,yi]=RK34(@(NduringI,[0;0]),0,tbeam,A,B,Current);
    CalcN2tb=yi(2,end);
    if abs(N2tb-CalcN2tb) > 1
        if CalcN2tb > N2tb
            Bhigh=B;
            B=B/2;
        else
            B=B+(Bhigh-B)/2;
        end
    end
end

while abs(N2tb-CalcN2tb) > 1
    [~,yi]=RK34(@(NduringI,[0;0]),0,tbeam,A,B,Current);
    CalcN2tb=yi(2,end);
    if abs(N2tb-CalcN2tb) > 1
        if CalcN2tb > N2tb
            Bhigh=B;
            B=B/2;
        else
            B=B+(Bhigh-B)/2;
        end
    end
end

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```
% filename = '.xlsx'; % spreadsheet to store values of A and B for each run
values = {A, B};

% xlswrite(filename, values, 'sheet1', 'cell number'); % save A and B the
spreadsheet created on specific cells

figure(9)

plot(Current) % plot current results

% plotting the decay equations with the simulated values of A and B
set(0, 'defaulttextinterpreter', 'none');
figure(10)

hold on

% plot(ti, yi4(1,:), '.r');
% plot(ti, yi4(2,:), '.b');
% plot(tb, yb(1,:), '.r');
% plot(tb, yb(2,:), '.b');
plot(tc, yc(1,:), '.r'); % green showing the range f
plot(tc, yc(2,:), '.b');

hold off

set(gca, 'fontsize', 18)
set(gca, 'LineWidth', 3);
legend('Isomer', 'Ground state');
xlabel('Time (min)');
ylabel('Population');
grid on

% the end
```
Appendix D

D.2 MATLAB graphs

Figure D.1 – Plot of population over time for gold $^{198}$Au in the Blank target. The red line is the population of the isomer and the blue line is the population of the ground state.

Figure D.2 – Plot of population over time for $^{198}$Au in the Close target. The red line is the population of the isomer and the blue line is the population of the ground state. The sudden change in population at 200 - 300 min is when the beam shut off.
Figure D.3 – Plot of population over time for $^{198}$Au in the Far target. The red line is the population of the isomer and the blue line is the population of the ground state.
Appendix E

E.1 Excel Solver spreadsheet

Using the non-linear curve fitting in Excel known as Solver (Microsoft Excel add-in program), the spreadsheet similar to Table E.1 below was created. The first two columns are containing the time values in minutes and beam current in nA respectively. The third and fourth columns were used to calculate the isomer and ground state population at the given times i.e. \([0, t_{irradiation}], [t_{irradiation}, t_{transport}]\) and \([t_{transport}, t_{counting}]\), where the populations were obtained by using the following equations:

\[
N_{\text{iso}}(i + 1) = N_{\text{iso}}(i)\lambda_{\text{iso}}N_{\text{iso}}(i) + AI(t(i))]dt \quad (E.1)
\]

\[
N_{\text{ground}}(i + 1) = \lambda_{\text{ground}}N_{\text{ground}}(i) + \lambda_{\text{iso}}N_{\text{iso}}(i) + BI(t(i))]dt \quad (E.2)
\]

http://etd.uwc.ac.za/
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<th>$t_{\text{irradiation}}$</th>
<th>$t_{\text{transport}}$</th>
<th>$t_{\text{counting}}$</th>
<th>$\lambda_{\text{isomer}}$</th>
<th>$\lambda_{\text{ground}}$</th>
<th>$A$</th>
<th>$B$</th>
<th>$\chi^2$</th>
<th>$D_{\text{measured, isomer}}$</th>
<th>$D_{\text{measured, ground}}$</th>
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**Table E.1** – Solving the production rate A and B using Excel Solver, this I testing values i.e. not for Blank, Far or Close target.
Bibliography


