Nuclear Spectroscopy of Radioactive Strontium-94

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Abstract: Traditionally, the production of neutron-rich nuclei in laboratories has been impossible. However, with the accelerator facilities now available at the Argonne National Laboratory’s ATLAS facility there is now a unique opportunity for the study of such nuclei. By studying how the $^{94}$Rb decays and releases energy, there has been a potential to update the currently accepted information, with further applications in nuclear astrophysics and nuclear reactor dynamics.

A number of data analysis tools were utilised in order to study how the nucleus $\gamma$-decays (where nuclei decay through the release of energy in the form of gamma rays) through various discrete energy levels after its initial $\beta^-$-decay (where the nucleus converts a neutron into a proton, then ejects an electron and antineutrino).

The data was collected at the ATLAS facility at Argonne National Laboratory, US during an experiment in 2016 utilising the CARIBU (Californium Rare Isotope Breeder Upgrade) and X-ARRAY/SATURN (Scintillator and Tape Using Radioactive Nuclei) facilities. The CARIBU accelerator facility utilises the spontaneous fission of a $^{252}$Cf sample, which produces incredibly rich nuclei, far beyond the valley of stability (the pattern of numbers of protons and neutrons that produce stable nuclei), resulting in the production of a directed beam of $^{94}$Rb. This beam is directed to the X-ARRAY and SATURN data acquisition facilities, which recorded data on the energy and intensity of $\gamma$-rays, as well as $\beta$-particles. The X-ARRAY consists of a number of high-purity germanium detectors, which measure incoming $\gamma$-rays, while SATURN consists of a scintillator to detect incoming $\beta$-particles as well as a moveable tape device for half-life measurement.

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I. Introduction

Though neutron-rich nuclei have traditionally been hard to produce in laboratory facilities, next-generation facilities such as the CARIBU (Californium Rare Isotope Beam Upgrade) addition to ATLAS (Argonne Tandem Linac Accelerator System) at the Argonne National Laboratory have provided reliable methods of producing beams of incredibly neutron-rich nuclei towards the nuclear drip line (the upper limit of possible nuclei). Globally, due to the significance of gaining an understanding of the behaviour of neutron rich nuclei, a number of facilities with such capabilities have been developed recently or are currently being built, such as the FRIB (Facility for Rare Isotope Beams), a US $800M accelerator facility at Michigan State University slated for completion in mid-2022[1]. Evidently, significant interest has been placed on the study of such nuclei due to the range of applications.

An understanding of neutron-rich nuclei allows for a greater understanding of nuclear structure as a whole, as current theories are able to be tested on previously unobtainable nuclei. For instance, the ‘magic numbers’ of protons and neutrons associated with nuclear stability are known to change as you move further towards the neutron-rich side of the valley of stability[2], and only the accurate and detailed study of experimentally-produced neutron-rich nuclei allows for such theories to be tested.

FIG. 1. A chart of nuclear stability, the horizontal axis indicates the number of neutrons, while the vertical axis indicates the number of protons. The black pattern represents the valley of stability[3]
Moreover, an understanding of how neutron-rich nuclei behave finds applications in the study of nuclear reactor dynamics. After nuclear fission is halted via the control rods in a nuclear reactor, a great deal of latent heat is released as a result of the decay of a vast number of daughter nuclei produced as a result of reactor fission. The amount of latent heat produced, as well as the varying rates at which it is produced over time can be better understood once a better understanding of the ways in which neutron-rich nuclei decay is obtained.

In addition to this, an understanding of neutron-rich nuclei is very valuable to nuclear astrophysics, as the production of incredibly neutron-rich nuclei through stellar fission and neutron capture is a key part of the production of elements beyond iron. After stellar fission occurs, nuclei rapidly capture neutrons as they move towards the neutron-rich side of the valley of stability, before moving up (in protons) via β-decay, then continuing to capture neutrons and so on until they reach stability as heavy elements. Thus, as our understanding of neutron-rich nuclei improves, so too does our understanding of nuclear astrophysics. [1]

By utilising data obtained at the ATLAS facility at Argonne National Laboratory and comparing it to decay data recorded in the ENDF (Evaluated Nuclear Structure Data File), which is a database of currently-known decay modes of various nuclei, the ways in which the excited 94Sr nucleus gamma decay below the neutron separation energy were able to be studied. The data analysed herein provides proof of a sizeable number of new excited states and transitions, indicating that currently accepted information on the decay of the 94Rb nucleus may need updating. Moreover, the study of alternate decay modes of 94Rb, particularly beta-delayed neutron emission, provides a much richer understanding of the radioactive behaviour of the nucleus. By determining the possibility of gamma decay above the neutron separation energy, as well as confirming that gamma rays from the decay of the 89Sr nucleus (produced via the neutron emission pathway) are present in the data set, it was shown that evidence exists for this mode of decay.

As such, this project has provided a unique opportunity to attain a greater understanding of the modes of decay of neutron-rich nuclei and update currently accepted values regarding this decay.

II. Concepts

A. Nuclear Structure

In order to consider the ground state properties of nuclei of interest, one must consider the interactions which occur between nucleons (protons and neutrons) and as such, an understanding of the structure of nuclei is essential for predicting and analysing nuclear data. Much like how atomic behaviour is modelled via the discrete arrangements of electrons based on their energies, which in turn govern interatomic interactions, the nucleus is modelled by considering protons and neutrons to be in their own arrangements. Moreover, this analogy between nucleons and electrons can be extended into the idea of orbitals, unique states (as per the Fermi Principle) which each fermion can occupy, defined by their quantum numbers and an energy level. These in turn are arranged into ‘shells’ or patterns. Much like how electrons release electromagnetic radiation as their electrons fall into lower (more stable) orbits, nuclei undergo decay processes in order to ensure a maximally stable arrangement of protons and neutrons, often aiming to decay to what is known as the Valley of Stability, the pattern of numbers of protons and neutrons that result in peak nuclear stability, which are often predicted by the ‘magic numbers’. Nuclei with so-called ‘magic numbers’ of protons and neutrons (2, 8, 20, etc.), possess closed shells, analogous to the closed electron shells of noble gases, resulting in enhanced stability. An understanding of highly neutron-rich nuclei is essential in testing the theory of ‘magic numbers’, as they have shown to differ towards the neutron-rich side of the valley of stability.

B. Nuclear Decay

Radioactive decay is the method by which unstable nuclei lose energy and return to a more energetically favourable and stable state. Understanding the ways in which exotic nuclei such as Rubidium-94 decay to produce a variety of decay products is key to gaining a level of understanding of nuclear physics.

In order to understand decay patterns and the structure of nuclei, ‘decay ladders/schemes’ are often used to describe the radioactive behaviours taken by the nucleus. Figure 1 displays the ground state properties of 94Rb from its β-decay level scheme, along with its decay mode (%β− noted as 100%), its Q-value (energy released via the decay), and its half-life (2.702(5) s).

![Diagram of 94Rb decay scheme](image)

**FIG. 2. Summary of the ground-state properties of the parent nucleus, 94Rb. The ground state energy (0.0 keV) and spin-parity assignment (3−) are shown. The β-decay Q value (10.281 (8) keV) is quite large for this decay mode, and proceeds with a half-life of 2.700(5) seconds.**

Most important to the level scheme, each ‘step’ represents an ‘energy level’, while each arrow down indicates a transition between levels, often by γ-decay, which continue down to the ground state of 94Sr. Each horizontal line indicates an excitation state, with the excitation energy and half-life given on the right, while the energy emitted by each gamma decay step, and its absolute intensity is provided above each arrow.
FIG. 3. Example portion of a nuclear level scheme. The horizontal lines represent excited states in the nucleus and are labelled with their excitation energy and mean lifetime of the state. Vertical arrows show transitions that connect the states, and are labelled with their transition energy, multipolarity (type) and relative intensity. [5]

These schemes are able to describe how nuclei in excited states release energy and achieve stability via $\gamma$ decay.

When considering neutron-rich isotopes such as $^{94}$Rb, which has 33 protons and 61 neutrons, the most probable decay modes are related to this ratio of neutrons to protons. This particular nucleus undergoes decay via three modes: gamma decay, where a photon is released from the nucleus in order to stabilise it; $\beta^-$ decay, where a neutron is converted into a proton, which is highly probable in neutron-rich nuclei in order to achieve stability; and beta-delayed neutron emission, where the nucleus decays via $\beta$ decay into a state higher in energy than the neutron separation energy ($S_n$), causing a neutron to be emitted as well.

This report is particularly concerned with the validation of the proposed level scheme of $^{94}$Rb,[4] as well as the measurement of decay via beta-delayed neutron emission.

III. Experimental Details

Traditionally, nuclear physics has been limited to studying very few neutron-rich nuclei due to the difficulty of producing such nuclei through traditional methods. Due to the conservation of particle numbers, and a lack of neutrons, fusion-evaporation reactions from particle accelerators tend to form neutron-deficient heavy nuclei. Prior to the development of facilities such as CARIBU, SATURN and the X-ARRAY at ATLAS, where the data analysed herein was collected, prior methods typically involved the collision of light nuclei to induce fusion, which tends to result in products on the proton-rich side of the valley of stability. Prior methods utilised to produce neutron-rich nuclei utilised particle-induced fission of $^{235,238}$U and $^{239}$Pu. Rather than utilised particle-induced fission, the CARIBU facility utilises a sample of $^{252}$Cf as the parent nucleus, which undergoes spontaneous fission 3% of the time, removing the need for equipment to induce fission and also resulting in favourable refractory element beams, and ones with low energies, suitable for decay studies like this. [8]

The facilities at Argonne National Laboratory have allowed for the production of unique and fascinating nuclei such as $^{94}$Rb, and the analysis of these highly neutron-rich nuclei provides an opportunity to better understand $\beta^-$ decay as well as beta-delayed neutron emission.

The Californium Rare Isotope Breeder Upgrade operates via the decay of a Californium-252 sample, which allows for the production of nuclei of interest that are generally greater in mass than those produced by traditional Uranium-235 and Plutonium-239. This has allowed for the study of more neutron-rich nuclei, which was previously limited by prior fission techniques such as Isotope Separation On-Line (ISOL). [11]

FIG. 4. Schematic layout of the CARIBU facility at Argonne National Laboratory. The X-Array and Saturn decay station was positions inside the blue CARIBU area during this experiment. [7]

The operation of CARIBU is as follows: the Californium sample decays, with its alpha particles and fission fragments being emitted into the RF gas catcher (which is filled with high-purity helium gas). The RF electric fields then thermalise these fission fragments, while the helium gas, having a high ionisation potential, ensures fission fragments are not completely neutralised while in the RF chamber due to the fact that they are emitted in a high-charge state. Applied DC fields then direct these charged fragments through to the RFQ (Radio-Frequency Quadrupole), which extracts helium and cools the gas. Electric fields are also applied to slow down the ion fragments before they are extracted into the RF-focusing cone, which guides the positive ions into the extraction nozzle of the gas catcher. This process
results in about half of the fission fragments being transformed into beams of $1^+$ or $2^+$ charge. The extracted beam is then accelerated through and analysed by the high-resolution isobar separator, which has a resolution of 1 in 20,000 parts of the mass of each isobar. [12], [6].

Fundamental to this experiment was the equipment used to detect and capture all the data studied herein. After the production of desired ion beams in CARIBU, these beams are directed to the X-Array, which consists of five High-Purity Germanium (HPGe) ‘clover’ γ-ray detectors, as well as the SATURN (Scintillator And Tape Using Radioactive Nuclei) β-particle detecting scintillators and half-life measuring tape. γ rays are able to be accurately detected by the clover detectors, which are arranged such that there are four in the vertical plane, and one in the horizontal, facing the incoming beam, allowing for an angle coverage of 65% of $4\pi$. [8]

For this particular experiment, one of the clover detectors was actually replaced with a neutron detector, though the data from that detector has not yet been made available at the time of writing. These high-purity germanium detectors work due to the semiconducting properties of germanium. As high-energy, ionising radiation enters the detector, some semiconducting atoms are ionised, producing ‘electron-hole pairs’, which simply refer to the pairing of a free electron, and a position where an electron could fit in an atomic lattice. These electron-hole pairs travel to electrodes under the influence of an electric field, producing a measurable pulse which is proportional to the energy of the radiation. As only a very-low-energy γ-ray is required to produce an electron-hole pair, HPGe detectors are incredibly accurate at detecting the presence of γ rays with energies as low as 30 keV [13]. However, the data they produce is nonetheless obscured by background radiation, Compton scattering, annihilation radiation, and various other sources of noise.

In order to reduce the back and number of false counts, the scintillator detectors are utilised to detect β-particles, which allowed for a cleaner gamma ray data set, requiring a β particle to also be present in coincidence with a γ ray to be recorded. These scintillators are made of plastic, and collect beta-particle data in the following way: first, the beam enters the scintillator via a brass collimator through a 6mm-diameter well in the plastic tube and is collected on an aluminium foil at the middle of the well. Radioactive decay then produces an output of light, which is reflected through the tube due to the plastic’s TiO reflective paint, to the photomultiplier, which outputs a signal proportional to the β-particle energy [8]. Moreover, a Compton filter was applied to the γ-ray data later on, further increasing data quality.

In addition to this, a moveable tape feature is included as a part of SATURN, which allows not only for the half-lives of nuclei to be accurately evaluated (though this data was not incorporated into this report), but it also allows for long-lived radioactive nuclei to be removed from the active sample. This means that these long-lived nuclei aren’t able to decay along with the nuclei of interest, reducing the number of ‘false’ gamma rays detected. [13], [8].

IV. Data Analysis and Results

A. Verifying the Level Scheme of $^{94}\text{Rb}$

Two programs were used for the analysis of data collected at Argonne: ROOT and RadWare. ROOT is a large-scale data analysis tool specifically built for particle physics by CERN [15], while RadWare is a suite of software developed for gamma ray coincidence data [14]. Throughout the data analysis process, both programs were used in order to produce spectra from gamma-ray histograms, allowing for visualisation of gamma ray coincidence data.

In order to analyse the data obtained through gamma ray spectroscopy, ROOT was used in order to apply ‘gates’ to 2D histograms, producing a spectra for all gamma rays coincident to a particular ‘gated’ gamma
ray energy. For example, the non-gated 2D histogram “GGEB_sub2”, as opposed to an 837keV gate.

FIG. 7. (Top) 2D gamma-gamma coincidence matrix showing one $\gamma$-ray on each axis and intersection points correspond to $\gamma$-ray coincidences. (Bottom) A projection of the 2D matrix gated on the 837-keV $\gamma$-ray transition in $^{94}$Sr. This shows all the $\gamma$ rays in coincidence with the 837-keV one and indicates their relative intensities.

From there, these spectra were exported to gf3, which was used to locate ‘peaks’ in the spectra by fitting Gaussian shapes on top of a quadratic background function to the spectra, which indicate the presence of coincident rays (i.e. gamma rays which ‘feed into’ the gated gamma ray), mapping out the gamma decay patterns. The first point of interest within this project was to verify a proposed expansion of the level scheme of $^{94}$Sr. In order to do so, gates were applied on 837keV, 1089keV, 1309keV and 1577keV. These strong $\gamma$ rays are shown in Fig 7; most other $\gamma$ rays resulting from the decay of this nucleus are in coincidence with at least one of them. A number of gates were inspected, rather than just one, to verify the level scheme. For example, the 1089 keV and 1309 keV $\gamma$ rays are known to ‘feed into’ the 837 keV level and as such, gating the data on 837 keV provides a clear view to these. However, a gate on 1089 keV is required to find those rays coincident to itself, but not 837 keV, such as the 678 keV signal.

FIG. 8. Section of the 94Sr decay scheme selected to demonstrate the gamma-ray coincidence-gating process and methods used to build the level scheme. [5]

FIG. 9. Gamma-ray spectra gated on the (top) 837-keV and (bottom) 1089-keV gamma-ray transitions. Dominant features in the spectre labelled and described in the text.

By going through and verifying each decay step, I was able to confirm almost the entire level scheme, though there were a number of potential new $\gamma$ rays detected (approximately 20) which are in the process of being further analysed at the time of writing, while there were a number of proposed $\gamma$ rays that were dubious due to
very low intensity. As such, this analysis has provided a potential for future investigation into the decay scheme of $^{94}$Sr and a significant update to the ENSDF.

B. Verifying Beta-Delayed Neutron Emission Pathway

As $^{94}$Rb is both highly neutron-rich, and exhibits a large $\beta$-decay $Q$ value (10.3 MeV) and moderate neutron separation energy ($S_n$), it possesses a high likelihood for decay via beta-delayed neutron emission. This mode of decay would lead to a $^{93}$Sr nucleus as such:

$$\begin{align*}
^{94}\text{Rb} &\rightarrow ^{93}\text{Sr}^* + e^- + \nu (\beta^- \text{Decay}) \\
^{93}\text{Sr}^* &\rightarrow ^{92}\text{Sr}^* + n (\text{Neutron Emission})
\end{align*}$$

In order to verify whether the $^{93}$Sr nucleus is enough excitation for this nucleus to spontaneously re-

By applying a gate on the $\gamma-\gamma$ coincidence dataset in ROOT for 213 keV, I was able to export this spectrum to g3, which showed a strong signal at 219 keV coincident with a 213 keV signal.

While these two coincident $\gamma$ rays were detected, none of the other emissions in the $^{93}$Sr decay scheme were detected. Nevertheless, there is no way that the 219 keV and 213 keV coincidence would have occurred without the presence of $^{93}$Sr, proving that $^{94}$Rb can undergo decay via beta-delayed neutron emission.

C. Determining Beta-Delayed Neutron Emission Branching Ratio

In order to determine the approximate likelihood of a nucleus undergoing beta-delayed neutron emission, there is a method which can be used based on the areas, efficiencies and intensities of a number of $\gamma$ rays. This method involves finding $\gamma$ rays which originate from the decay of a nucleus which originates from further $\beta$ decay following the decay via beta-delayed neutron emission of the parent nucleus. In the case of $^{94}$Rb, this nucleus is $^{93}$Y.

As per the diagram, the method involves choosing two coincident $\gamma$ rays from each nucleus’ decay scheme. In the case of $^{94}$Sr, the 1089-and-837-keV $\gamma$-ray emissions were chosen, and for $^{93}$Y, 168-and-590keV energies were chosen. The formula involved is as follows:

$$\frac{I(\gamma_{837\text{keV}})}{I(\gamma_{1089\text{keV}})} = \frac{A(168)}{\epsilon(1089)} \frac{168}{590} \frac{\epsilon(837)}{\epsilon(1089)} \frac{b(837)}{b(1089)}$$

Where $A(x)$ refers to the peak area of $\gamma$-ray of energy ‘x’ in the gated spectrum, $\epsilon(x)$ refers to the relative efficiency of $\gamma$-ray ‘x’, and $b(x)$ refers to the number of counts of $\gamma$-ray ‘x’ per 100 $\beta$-decays. By using a $\gamma$-ray relative efficiency database, as well as the counts per hundred information on the ENSDF and the area under each peak from g3’s sum command produced a branching ratio of 0.09(3) instances of beta-delayed neutron emission per $\beta$-decay, which is comparable to the ENSDF’s accepted value of 0.1018(24).

D. Evidence of $\gamma$ Decay above Neutron Separation Energy

Another point of interest in this data set was to determine whether there were in fact events where the Rubidium-94 nucleus had decayed into $^{93}$Sr nuclei with excitation energies above the neutron separation energy ($S_n$), which were then followed by $\gamma$ decay rather than neutron emission. This ties into the search for proof of beta-delayed neutron emission, as this phenomenon only occurs where the parent nucleus produces a nucleus with excitation energy between the Q-value of the parent, and the neutron separation energy. At these energies, there is enough excitation for this nucleus to spontaneously release a neutron, leading to the $^{93}$Sr decay scheme.

In order to determine whether such an event had occurred, I used ROOT and g3 to produce and add together $\gamma$-ray gated spectra and determine what patterns emerged in the high-energy region (around 6 MeV+). While no discernible patterns were found when taking the data as is, after ‘rebinning’ (reducing the number of channels to make the data ‘rougner’) the spectra before adding them, some possible peaks emerged in the combined 837 keV and 1577 keV spectrum, seen in Fig 8, with energies of 6160 keV, 6368 keV and 6592 keV.

FIG. 10. Section of the $\gamma$-ray coincidence spectra gated on 213 keV. The horizontal axis describes the energy of the measured $\gamma$-ray, while the vertical axis describes the number of counts. Labelled is the peak associated with the 219 keV $\gamma$-ray, indicating its presence.
While these peaks were possible, they were of very low counts, being not much higher than background radiation. In order to determine whether there is a real possibility that these peaks are of interest, I compared the area under each peak with the area under a known peak from the level scheme in order to determine relative intensity. I chose the 1767 keV transition known to have had areas of approximately 12(4), giving a likelihood for each of these γ rays equivalent to about 0.0097(30) of the 1767 keV peak. As such, each of these peaks have an absolute intensity of approximately 0.001, indicating they are expected to occur for one in every 100,000 β decays of 94Rb.

Though this is only tenuous evidence that these high-energy γ rays do occur within 94Sr’s γ decay, it is nonetheless a starting step for further investigation. A repeat experiment was conducted again in late 2021, resulting in a new set of data to compare this work with for future study, though at the time of writing, this data is not yet prepared for analysis.

V. Conclusions

Identifying and understanding the ways in which highly neutron-rich nuclei such as 94Rb decay provides a path to not just a greater understanding of nuclear physics, but also provides an opportunity for further advances in nuclear applications, particularly fission-based energy sources.

This experiment allowed for the proposed level scheme of 94Rb to be verified, and the results found herein provide an opportunity to update the adopted decay scheme, adding some newly found transitions. Moreover, the possibility of the beta-delayed neutron emission pathway, as well as an approximation of its branching ratio were found. While the data suggested that there is only limited proof of the beta-delayed neutron emission, this has provided an opportunity for further confirmation and study upon the arrival of the latest sets of data. Likewise, though there was only tentative proof of gamma decay above $S_n$, the fact that there was anything at all provides future research potential for further confirmation.

The data collected for this experiment at Argonne National Laboratory provided an excellent starting point for the further research of 94Rb, and the data from the repeat experiment in late 2021 will also provide an opportunity for further study in order to produce more accurate and reliable findings.

In particular, the neutron-detector data from the experiment (which is unavailable as of the time of writing) would provide an excellent resource for future study of the beta-delayed neutron emission pathway.

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