Title: Searching for octupole collectivity in neutron-rich Ce isotopes and isomer depletion pathways in ^{113}Cd

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Context and Aims

The initial focus for this project was to ~~experimentally~~ investigate ~~the feasibility of storing energy using~~ an excited state of the nuclide ^{113}Cd with an exceptionally long half-life, known as a *nuclear isomer* ~~or *metastable state*~~. However, due to the global COVID-19 outbreak, ~~the planned experiments can not be carried out at present~~. If it ~~becomes~~ safe to perform these experiments at a time that still allows for a thorough analysis of the resulting data, ~~returning to this research area shall be considered~~. In the interim, the ~~primary~~ focus of this project ~~is to analyse some~~ yet unstudied experimental data ~~and extend existing knowledge of the nuclear structure in the mass number A ∼ 144 region~~.

The primary reason isomers are worth investigating for energy storage applications is that the harnessable energy stored within nuclei is around 10^9 J/g, approximately ~~two~~ orders of magnitude greater ~~than that of chemical fuels~~. Hence, isomers with half-lives ~~above~~ one year are currently being methodically studied to identify which of them could be used ~~in~~ energy storage devices ~~with long lifetimes~~. These devices would suit use in areas where the regular restocking of energy supplies may be impractical or inefficient, such as regions that are not easily accessible or highly remote [1].

~~The isomer of~~ ^{113}Cd (denoted ^{113m}Cd) ~~has~~ a half-life $T_{1/2} = 14.1$ years, making it a worthwhile candidate for such applications. However, realising the potential of this metastable requires finding a mechanism by which the energy of the isomer can be released. A current frontrunner is the process of *isomer depletion*, where a nuclear isomer is excited into a more energetic state (an *intermediate state* or *depletion level*) that can then decay via a cascade of gamma rays into the ground state. It has been ~~demonstrated~~ for isomeric forms of a small number of other isotopes, but not yet for ^{113m}Cd [1].

This ~~investigation~~ would be centered around constructing a picture of the excited states (*level scheme*) of ^{113}Cd and finding a sequence (*depletion pathway*) of gamma ray transitions that connect the isomer to the ground state. A secondary goal will be to explore the phenomenon of beam breakup (where the beam particle splits into smaller fragments that can each react with the target [2, 3]) using charged particle detectors, should they be functional when the experiments are performed. The specific aims here would be to verify that beam breakup did occur and measure cross sections for reactions of each beam fragment with the target.

~~Yet,~~ while access to the necessary experimental facilities is restricted, existing experimental data ~~(consisting of gamma rays emitted by the excited states of neutron-rich isotopes~~ ^{148,150,150}Ce) will be analysed. These ~~spectra were recorded during~~ experiments designed to test the prediction of the deformed shell model of nuclear structure that nuclides with proton numbers near 56 and neutron numbers near 88 ~~will exhibit octupole collectivity~~. This phenomenon denotes the collective motion of nucleons that causes the charge distributions ~~of the excited states~~ of these nuclei to deviate ~~in geometry~~ from ~~the~~ spherical shapes ~~of their ground states~~. As these nuclei comprise approximately 144 nucleons, one might expect that describing their collective motion might be quite complicated. Hence, the fact that interactions between only a small number of the constituent nucleons are responsible for this behaviour is worth understanding.

At present, observations of these correlations have not been made for all of the nuclides predicted by the deformed shell model, including the neutron-rich isotopes ^{148,150,152}Ce. Hence, the primary research aim of this project is to extend the existing body of information surrounding octupole collectivity in these A $\sim$ 144 nuclides.

This will be achieved by analysing the recorded gamma rays to first construct a level scheme for these nuclides, including spectroscopic data (excitation energies, lifetimes, spins and parities) for the low-lying excited states. Using this information, dipole and quadrupole moments of the charge distribution of the nuclei will be determined. Identification of these moments will, in turn, allow deduction of the nature of any octupole collectivity that is present.

~~Regardless of this project's specific research focus, the underlying~~ goal is to develop and utilise gamma ray spectroscopic analysis routines to extract key information from ~~experimental~~ nuclear data. An aim that follows on from this is to meaningfully interpret this data under the framework of a range of theoretical nuclear structure models.

The latter aim above ties in with the ongoing experiments being performed by researchers to test the predictions of theoretical nuclear structure models and further our understanding of nuclear physics. An understanding of this field has useful applications in astrophysics [4], energy storage [1], nuclear medicine [5], relativity and particle physics [6].

Background

Octupole correlations

The ~~collective~~ behaviour ~~referred to by octupole collectivity~~ can be described by considering the interaction of ~~only a few~~ nucleons ~~because A $\sim$ 144 nuclides have Z and N near the closed-shell magic numbers Z = 50 and 82.~~ This means that within these nuclei, most nucleons form part of a frozen core that is largely unchanged when the nucleus is excited. ~~The nature of the excited state is thus determined by the few nucleons that become unpaired following the excitation (and the orbitals they occupy).~~

~~The observation of~~ these interactions ~~can provide evidence that a nucleus possesses~~ a non-spherical (often pear-shaped) charge distribution. ~~These~~ correlations arise ~~when~~ nucleons in nuclear orbitals with opposite parity (positive and negative) and a ~~precise relationship between their quantum numbers~~ interact strongly. ~~Specifically, the interacting orbitals will satisfy~~ $\Delta n = 1$ and $\Delta l = \Delta j = 3$, where n, l and j denote the principal, orbital angular momentum and total angular momentum quantum numbers. Based on the deformed shell model, ~~near A $\sim$ 146, the proximity (in energy) of either~~ the $h_{11/2}$ and $d_{5/2}$ proton orbitals or the $i_{13/2}$ and $f_{7/2}$ neutron orbitals ~~(depending on the particular nature of the nuclear excitation) leads to octupole correlations because they satisfy the above relationships.~~

~~Previous, large arrays of gamma ray detectors have been used to record the gamma rays emitted by high spin excited states of neutron-rich nuclei near ^{144}Ba (Z $\sim$ 56, N $\sim$ 88) (formed from the spontaneous fission of ^{252}Cf) and study octupole collectivity [7,8]. Specifically,~~ the observation of strong E1 transitions between the yrast ~~sequences of~~ energy levels ~~with respectively positive and negative parities~~ has provided much of the experimental evidence for octupole correlations near ^{144}Ba. ~~The description of these transitions as E1 provides information about the (orbital) angular momentum carried by the emitted radiation as well as its angular distribution [6]. The yrast sequence (or *band*) of energy levels simply refers to that with the minimal excitation energy for a given value of the total angular momentum J [6].~~

Studies of low-spin states in nuclides near A $\sim$ 146 have also been made, by measuring the γ rays emitted during the β^- decays of ^{142,144,146}Cs to obtain lifetime data for the low-spin

states in the positive and negative parity yrast bands of ^{142,144,146}Ba [9]. ~~A coincidence analysis method was used, wherein the time between the detection of a β^- particle in one detector and a gamma ray in another gives the lifetime of the excited state populated when Cs β^- decays into Ba. This approach is valid because the time between the two particles being emitted is short enough that if they are detected sufficiently close together in time, they can be said to have originated from the same nucleus. Collection of a sufficient number of these *coincidence* events (so described because the detection of the two particles is effectively coincident) enables an accurate lifetime measurement.~~

Yet, relatively few studies of the isotopes ^{148,150,152}Ce have been ~~made, with~~ most existing information on their level schemes coming from experiments that measured products of the spontaneous fission of ^{252}Cf [10–12]. The experiments that generated the data to be analysed for this project were performed at the Californium Rare Isotope Breeder Upgrade (CARIBU) facility at Argonne National Laboratory (ANL). At this facility, beams of neutron-rich (spontaneous) fission fragments of ^{252}Cf ~~such as ^{148,150,152}La were~~ transported to the low-energy CARIBU decay ~~station and implanted on to the moving tape system. This tape system periodically removes any sources with a long-lived activity that may obscure the signals of interest and is situated at the target position of the detector array.~~ This array contains four clover detectors (each comprising four Ge detectors) and ~~between two and four~~ LaBr(Ce) ~~detectors, giving a total of (up to) 20 gamma ray detectors. However, a~~ plastic scintillator was also included ~~in the detector array,~~ specifically ~~to~~ detect β^- particles.

These three detector types enable three-fold β-γ-γ coincidence measurements to be made. These in turn allow specific excited states to be identified and their lifetimes ~~subsequently~~ ~~calculated.~~ The timing characteristics of the scintillator and LaBr(Ce) detectors used mean that this set up ~~should~~ enable measurement of excited state lifetimes as short as 30 ps. These capabilities of the equipment used will thus ultimately enable the existing knowledge surrounding ^{148,150,152}Ce to be extended, once the data is analysed.

Nuclear isomers

In general, nuclear isomers exist because the decay of t~~he isomeric~~ excited state is inhibited. This could be ~~because the~~ fundamental quantum mechanical selection rules ~~for the allowed photon transitions between levels~~ mean there are no lower energy states within that nucleus which the isomer may decay ~~into.~~ ~~Alternatively,~~ the metastable state may only decay via transitions where the difference in ~~the~~ total angular momentum of the initial and final states ΔJ (the *multipolarity*) is relatively large. ~~This could account for the long lifetime of the isomer as~~ transitions with large multipolarities are known to be slow relative to those with smaller ΔJ values. Figure 1 shows that ^{113m}Cd ($J = 11/2$, 263.54 keV) can only de-excite into the ground state ($J = 1/2$). Thus it can only decay via a relatively high multipolarity $\Delta J = 5\hbar$ transition ~~and falls into the latter category of isomer described above~~ [13].

Figure 1 ~~does suggest~~ that there are a~~lready~~ possible pathways by which ^{113m}Cd could be excited into a more energetic state and then decay into the ground state. For example, the isomer could be excited into the $7/2^-$ state at 522.259 keV. If it then decayed via an E1 transition to the 316.206 keV state before undergoing an E2 transition to the ground state, this would release the 263.54 keV stored in the isomer. However, only 1.22 E1 transitions occur for every 100 E2 transitions from the $7/2^-$ state, so this pathway would not be strongly favoured and thus potentially not a reliable option for achieving isomer depletion. There are other options within the level scheme of ^{113}Cd shown in Figure 1, but they are each similarly flawed.

~~Yet,~~ isomer depletion has been experimentally ~~achieved~~ for six other nuclides, using a range of distinct mechanisms. These include exposing a sample containing a significant isomer pop-
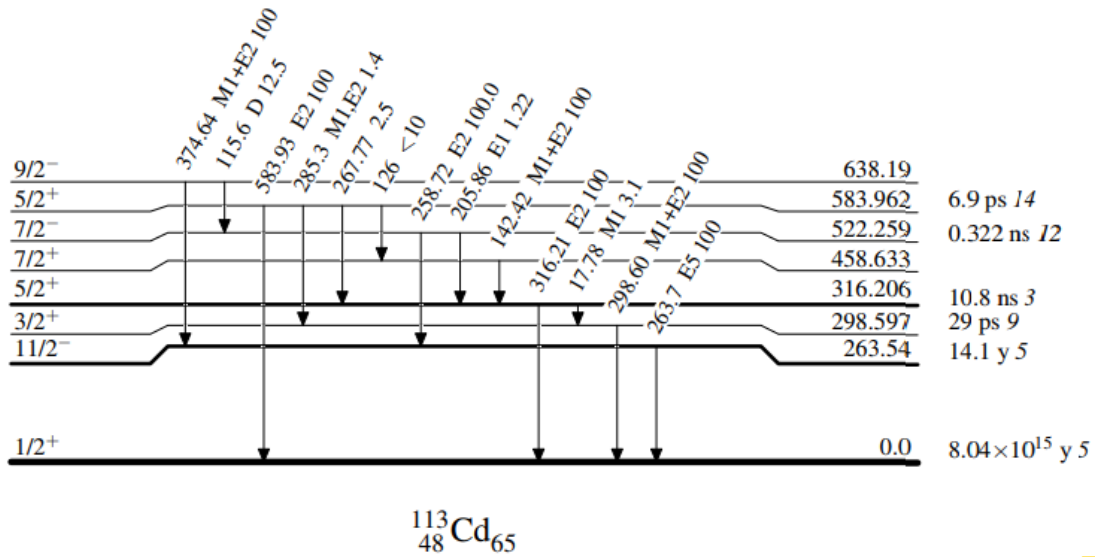
Figure 1: Partial level scheme for ^{113}Cd. The isomeric state has an energy of 263.54 keV [13].

ulation to bremsstrahlung radiation, using Coulomb excitation ~~on another such sample~~, or initiating nuclear excitation by electron capture [1]. Thus, identification of a viable depletion pathway within ^{113}Cd would extend the list of nuclides that ~~can~~ be harnessed to store energy over long timescales.

Project Description

The analysis techniques and software that will be used are all methods and programs that are commonly used by and familiar to the other members of my research group. They are also well documented, so there is a wealth of experience that can be tapped into when queries regarding the processing of data inevitably arise. ^{148}Ce and ^{150}Ce have been studied using La β^- decay experiments, so analysis of the recorded data for these isotopes will build on existing knowledge and potentially verify tentative conclusions reached in previous works. Contrastingly, ^{152}Ce has only been studied via the spontaneous fission of ^{252}Cf, so analysing this nuclide via the β^- decay of ^{152}La should provide new information on its structure. Dedicated octupole collectivity studies have not been performed for any of these Ce isotopes, however, so this project will be breaking new ground in this area.

By using β-γ and β-γ-γ coincidence analysis methods the program ROOT will be utilised to explore the recorded gamma ray data. This process will enable the construction of a level scheme for each Ce isotope studied, aided by the consideration of γ-γ coincidences (from the clover detectors) where one of the γ rays is known to be emitted by a ~~given~~ Ce isotope. Further analysis of gamma ray coincidences (as well as individual spectra) will then lead to the collection of key spectroscopic information, such as state lifetimes, excitation energies and spin-parities.

~~If present,~~ evidence ~~of~~ octupole collectivity will ~~then be identified. Primarily this will be achieved by looking for~~ strong E1 transitions between yrast bands of opposite parity, which will naturally require identification of the yrast bands themselves. The strength of the octupole correlations that are present will then be assessed by evaluating the dipole and quadrupole moments of different transitions. For each Ce isotope, this will result in the fabrication of a level scheme that will contain some new levels, transitions and spectroscopic information, as well as an assessment of the degree of octupole collectivity that they each exhibit.

~~As for the nuclear isomer phase of this project,~~ isomer depletion-focused studies of ^{113}Cd have

not yet been recorded. ~~Hence~~, at the very least, these tests will build upon existing knowledge of the level scheme of this nuclide (and potentially the phenomenon of break up as well). Some preparatory work for this phase has already occurred (Figure 2), including identifying the reactions to be used by reviewing previous experiments in the literature that focused on excited states of ^{113}Cd. The qualitative behaviour of the relevant reaction cross sections as a function of energy has also been calculated using the PACE4 fusion-evaporation code.

Should this phase go ahead, the detectors to be used in the CAESAR gamma ray detector array will need to have their energy resolution characterised before being inserted into the array. Simultaneously, calculations will have to be performed to determine the parameters (beam energies, target thicknesses, exposure times) that are optimal for the production and detection of excited states of ^{113}Cd. Next, the targets will have to be readied, the experiments performed and then the data analysed. This data analysis will entail the construction of a level scheme, followed by the identification of any possible isomer depletion pathways. Aside from the construction of the CAESAR array, I will be in charge of ensuring these steps are completed, with the help of other members of the ANU Nuclear Spectroscopy group as required.

If time remains and charged particle detectors (fabricated and tested by other members of the Nuclear Spectroscopy group) are included in the CAESAR array, breakup reactions that may have happened during the irradiation phase of the experiment could be investigated. This would extend the measured results of this project and would be interesting from a theoretical physics point of view, but is not required to explore isomer depletion. However, if all of the other tasks above are completed, then a level scheme for ^{113}Cd will be able to be constructed and potential depletion pathways identified.

Project Plan and Feasibility

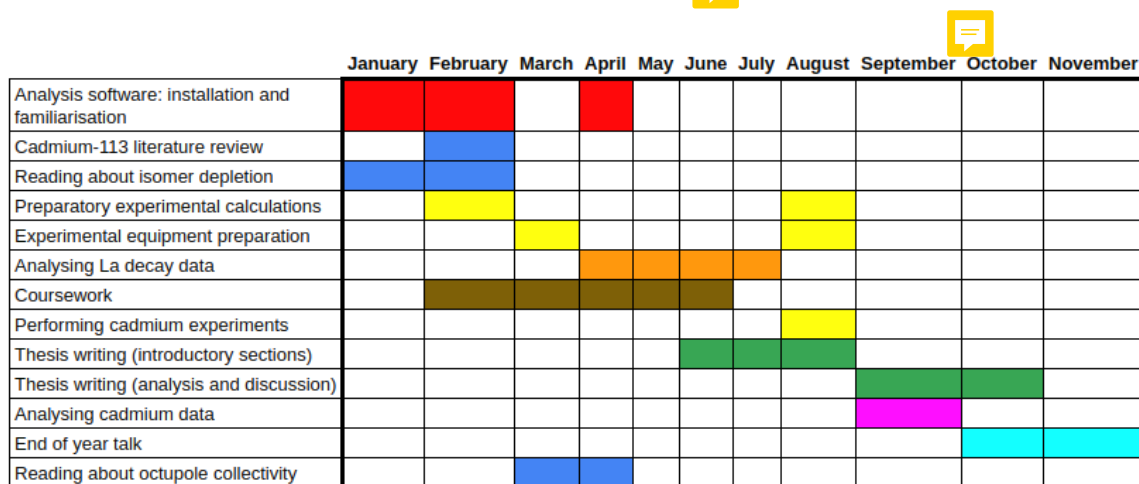|	January	February	March	April	May	June	July	August	September	October	November
Analysis software: installation and familiarisation	■	■		■							
Cadmium-113 literature review		■									
Reading about isomer depletion	■	■									
Preparatory experimental calculations		■						■			
Experimental equipment preparation			■					■			
Analysing La decay data					■	■	■				
Coursework		■	■	■	■						
Performing cadmium experiments								■			
Thesis writing (introductory sections)						■	■				
Thesis writing (analysis and discussion)									■	■	
Analysing cadmium data									■		
End of year talk										■	■
Reading about octupole collectivity			■	■							

Figure 2: Planned timeline for this project. The inclusion of experimental preparation and execution in August represents the latest feasible time when this can occur. Tasks assigned to January through March have been completed.

The time allocated to each of the following steps is outlined in Figure 2. The following data processing steps are central to the completion of a satisfactory project and will be performed using the ubiquitous nuclear data analysis frameworks ROOT and Radware.

1. Check the energy and time calibration of the existing gamma ray data.

2. Analyse the data in ROOT to construct a level scheme for ^{148,150,152}Ce.

3. Establish excitation energies and lifetimes of excited states in ^{148,150,152}Ce.

4. Identify the multipolarities and transitions between excited states of ^{148,150,152}Ce.

5. Measure the lifetimes of the ^{148,150,152}La ground states and constrain their spin-parities (stretch goal).

These analysis processes shall be continued in a dedicated manner until September 1 at the latest. Concurrent to this analysis the writing of the introduction, motivation, background and methodology sections of the thesis shall occur. As time progresses, we will assess the feasibility of completing the planned ^{113}Cd experiments before September 1 ~~weekly~~. This date is the latest feasible point at which there will remain time for thorough data analysis, collation of results and completion of the thesis, all by October 29.

If restrictions on access to the required experimental facilities are lifted by August 1, then the steps below will be successively executed. The exact timing and feasibility will depend on beamtime being secured, through communication with my supervisor. All steps below are goals that are essential to the project unless otherwise specified.

1. Cool HPGe gamma ray detectors (for use in the CAESAR detector array) to liquid nitrogen temperatures.

2. Perform STROP calculations to identify the optimal target thickness, taking into consideration the energies identified as optimal for maximising the cross section for the reaction channel that produces ^{113}Cd.

3. Test HPGe detectors using PIXIE. Having already had some experience using this software, the completion of this step will not be a prolonged process.

4. Prepare ^{110}Pd targets and create a mounting system for them. Establish whether we have enough targets and beamtime to perform experiments using both ^{7}Li and ^{9}Be beams.

5. Build the CAESAR detector array, in collaboration with A. J. Mitchell, Greg Lane, James Pope and other members of the Nuclear Spectroscopy group.

6. Perform experiments. Ideally, we would perform experiments with both ^{7}Li and ^{9}Be beams, but this is time and target dependent. If limited beam time is available, only one beam and a narrower range of beam energies may be able to be used.

7. Analyse data to construct a level scheme for ^{113}Cd and identify any viable depletion pathways. Deduce all possible spectroscopic information. The analysis skills gained during the octupole collectivity part of the project will enable this to be completed efficiently.

8. ~~Stretch goal:~~ measure reaction cross sections as a function of energy.

9. Stretch goal: Investigate charged particle-γ coincidence data to identify beam breakup reactions and measure their cross sections as a function of energy.

10. Write results, discussion and conclusion sections of the thesis, by October 29.

11. Prepare the final presentation, due November 5.

This structure ensures that a satisfactory thesis will be generated from this project, but as it is the busiest possible timeline with all possible parts included, it provides plenty of room to omit aspects should the time no longer be available for them. As a bare minimum, fundamentally interesting nuclear structure data be analysed and significant skills in the analysis of nuclear experimental data be developed, achieving the two primary goals of this project.

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