

PROPOSAL TO THE PAC OF THE LEGNARO NATIONAL LABORATORY  
Using the Galileo + Euclides + Recoil-Filter Detector + LaBr<sub>3</sub>(Tl) set-up

## Octupole shape transitions in the thorium isotopes

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### Abstract

The coupling of the Recoil Filter Detector (RFD) to Galileo presents an excellent opportunity to perform detailed  $\gamma$ -ray spectroscopy in the light-actinide region, where  $\gamma$ -ray spectra are normally impaired by a large background of  $\gamma$  rays from prompt fission. Here, we propose an experiment to study high-spin states in the nuclei  $^{221}\text{Th}$  and  $^{224}\text{Th}$  using the RFD with Galileo. The nucleus  $^{221}\text{Th}$  offers one of the best cases in which to study the evolution of parity-doublet bands with angular momentum. The nucleus  $^{224}\text{Th}$  lies at the heart of the light-actinide octupole-deformed region, and has recently been highlighted by covariant density-functional theory calculations as being a key nucleus in the understanding of shape transitions in this region. In the proposed experiment, we will extend the previously-observed octupole band structures to higher angular momenta and we will look for the signatures of shape transitions such as rotational alignments or a change in the  $B(E1)/B(E2)$  ratios. In addition to the main goals of the experiment, we will supplement the Galileo array with 10 or 12 LaBr<sub>3</sub>(Tl) detectors in order to test the feasibility of lifetime measurements in the yrast octupole bands using direct-timing methods. To populate the nuclei of interest, we will use the  $^{208}\text{Pb}(^{18}\text{O},5n)^{221}\text{Th}$  and  $^{208}\text{Pb}(^{18}\text{O},2n)^{224}\text{Th}$  reactions with a beam energies of 78 and 110 MeV. We will detect prompt  $\gamma$  rays with Galileo in coincidence with recoiling evaporation residues detected using the RFD. The detection of evaporation residues will help remove the large background of  $\gamma$  rays from prompt fission. We will use Euclides to detect  $\alpha$  particles as a means of additional channel selection  $\gamma$  ray spectroscopy of some of the weaker reaction channels such as  $^{221}\text{Ra}$  ( $\alpha n$  evaporation). In total, we request **7 days** of beam time for this work (two days for  $^{221}\text{Th}$  and five days for  $^{224}\text{Th}$ ).

### Introduction: octupole correlations in the light actinides

The largest reflection-asymmetric octupole deformations are observed in the light-actinide region around  $^{224}\text{Th}$  ( $Z=90$ ,  $N=134$ ) where the octupole-driving  $\Delta\ell=\Delta j=3$  orbitals are  $f_{7/2}$  and  $i_{13/2}$  for protons and  $g_{9/2}$  and  $j_{15/2}$  for neutrons [Ahmad93][Butler96]. Experimentally, the signature of octupole deformation is two sequences of states with opposite parities, which are connected by enhanced E1 transitions ( $\sim 10^{-2}$  W.u., cf.  $10^{-5}$  W.u. in reflection-symmetric nuclei). In the light-actinide region, such features have now been observed in more than 20 nuclei, with the best examples in the radium ( $Z=88$ ) and thorium ( $Z=90$ ) isotopes [Schulz89] [Smith95][Cocks99]. Despite the number of examples of this level structure in even-even nuclei, there is still surprisingly little information beyond the yrast octupole bands, and very few examples of well-formed parity-doublet bands in odd-A nuclei.

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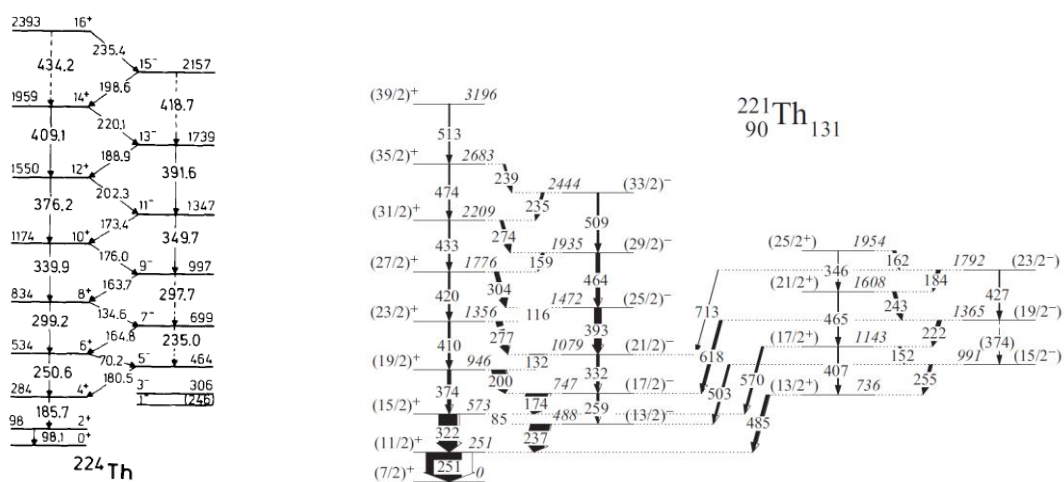
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## Theoretical approaches

Over the past 35 years, a number of other theoretical approaches have been applied to the study of octupole correlations in the light actinides. In the 1980s, Nazarewicz and co-workers published a number of papers relating to the behaviour of octupole-deformed nuclei under rotation [Nazarewicz84][Nazarewicz85][Nazarewicz87]. In Ref. [Nazarewicz87], the radium and thorium nuclei were singled out and analysed systematically, making predictions about the shapes and shape changes as a function of neutron number and rotational frequency. More recently, Robledo and Bertsch [Robledo11] published a global survey of octupole excitations in even-even nuclei using Hartree-Fock-Bogolyubov self-consistent mean-field theory with different Gogny interactions to compute the wavefunctions, redefining the region of strong octupole correlations in the actinides. Recently, octupole correlations have had a resurgence of interest following the study of  $^{224}\text{Ra}$  using radioactive ion beams at ISOLDE as described in Ref. [Gaffney13]; that work highlighted the importance of quadrupole-octupole coupling to explain the observations. In 2015, self-consistent relativistic energy-density functional (EDF) calculations were performed by Yao et al. [Yao15] which show that the low-spin behaviour  $^{224}\text{Ra}$  can be described in terms of dynamical octupole correlations with rotation-induced octupole-shape stabilization. Relativistic EDF calculations have also recently been performed by Li et al. [Li16] in order to obtain a systematic understanding of the quadrupole and octupole shape coexistence in some light-actinide nuclei. More recently, systematic relativistic EDF calculations have also been performed by Agbemeva et al. [Agbemeva16] for all nuclei with  $Z \leq 106$ , making predictions for the extents of the different regions of octupole correlations, and proposing a new region.

Of specific interest in the present proposal is the work by Li et al. in Ref. [Li13] in which covariant energy density functional theory is used to study the evolution of quadrupole and octupole shapes in the thorium isotope chain. That work states that at  $N=130$  ( $^{220}\text{Th}$ ) the ground-state is close to spherical, whereas quadrupole and octupole deformation develops rapidly with the addition of a few neutrons. For  $N=134$  ( $^{224}\text{Th}$ ), the potential energy surfaces, excitation energies and transition rates calculated by Li et al., indicate the occurrence of a simultaneous phase transition from spherical to prolate shape and from prolate (reflection-symmetric) to octupole (reflection-asymmetric) shape. In that work, the nucleus  $^{224}\text{Th}$  is identified as being the closest nucleus to the critical point of a double phase transition.



**Figure 1:** The present level schemes of the nuclei of interest in this proposal:  $^{224}\text{Th}$  (left) and  $^{221}\text{Th}$  (right). The level scheme of  $^{224}\text{Th}$  is taken from Ref. [Ackermann93] and the level scheme of  $^{221}\text{Th}$  taken from Ref. [Tandel13]. Despite being at the centre of the light-actinide octupole region, the level scheme of  $^{224}\text{Th}$  has not been studied by  $\gamma$ -ray spectroscopy for 25 years.

## The thorium isotopes

All of the thorium isotopes from  $N=130$  ( $^{220}\text{Th}$ ) to  $N=144$  ( $^{234}\text{Th}$ ) have low-lying negative parity states which are suggestive of octupole correlations. The thorium isotopes from  $N=130$  ( $^{220}\text{Th}$ ) to  $N=136$  ( $^{226}\text{Th}$ ) have interleaving positive- and negative-parity states, with interband E1 transitions forming the “classic” octupole band structure. The nucleus  $^{223}\text{Th}$  ( $N=133$ ) exhibits what is probably the best known example of a parity-doublet band structure [Dahlinger88][Marquet17]. The onset of a parity-doublet structure has recently been identified in the nucleus  $^{221}\text{Th}$  ( $N=131$ ) by Tandel et al. [Tandel03]. The level schemes of the nuclei of interest in this proposal,  $^{221}\text{Th}$  and  $^{224}\text{Th}$ , are shown in Fig. 1.

## Goals of the proposed experiment

The goals of the proposed experiment can be stated as follows.

- To extend the level scheme of the yrast octupole band in  $^{224}\text{Th}$  up to  $30 \hbar$  and to search for signatures of shape transitions.
- To search for new non-yrast structures in  $^{224}\text{Th}$ , similar to that observed in  $^{222}\text{Th}$ .
- To extend the parity-doublet bands in  $^{221}\text{Th}$  to spins of around  $67/2 \hbar$  and to search for signatures of shape transitions.
- To assess the feasibility of the measurement of lifetimes of the lower-spin ( $< 10 \hbar$ ) members of the yrast octupole bands using direct-timing methods with  $\text{LaBr}_3(\text{Tl})$  detectors, and measure lifetimes of states where possible.

## Previous studies of $^{221}\text{Th}$ and $^{224}\text{Th}$

The nucleus  $^{221}\text{Th}$  was first observed by Torgerson and Macfarlane in 1970 [Torgerson70]. The nucleus  $^{225}\text{U}$ , which is the  $\alpha$ -decay parent of  $^{221}\text{Th}$ , was not itself identified until 1989 [Andreev89][Hessberger89] – so unlike other nuclei in the region, the first excited states were not identified following  $\alpha$  decay. Excited states in  $^{221}\text{Th}$  were first studied in the 1980s by Dahlinger, using the  $^{208}\text{Pb}(^{16}\text{O},3n)$  reaction [Dahlinger85, Dahlinger88]. That work identified states up to  $12 \hbar$  above the ground state. A second study using the  $^{208}\text{Pb}(^{16}\text{O},3n)$  reaction in 2013 by Tandel et al. [Tandel13] extended the yrast octupole band to  $16 \hbar$  above the ground state, and identified some states in the parity-doublet band.

The nucleus  $^{224}\text{Th}$  was first observed in the  $\alpha$  decay of  $^{228}\text{U}$  in 1949 by Meinke et al. [Meinke49]. High-spin states in  $^{224}\text{Th}$  were identified using the  $^{208}\text{Pb}(^{18}\text{O},2n)$  reaction by Schwartz et al. in 1986 [Schwartz86]; an alternating-parity band was observed up to spin  $12 \hbar$ . Subsequently, the  $^{226}\text{Ra}(\alpha,6n)$  reaction was used by Schüler et al. and by Ackermann et al. in Refs. [Schüler88] and [Ackermann93], to extend the alternating-parity band to spin  $18 \hbar$ . The nucleus  $^{224}\text{Th}$  has not been studied by  $\gamma$ -ray spectroscopy for over 25 years.

## Experimental details

In the proposed experiment, we will perform detailed high-spin spectroscopy of the nuclei  $^{221}\text{Th}$  and  $^{224}\text{Th}$  using the  $^{208}\text{Pb}(^{18}\text{O},5n)$  and  $^{208}\text{Pb}(^{18}\text{O},2n)$  reactions, respectively, with the Galileo-Euclides-RFD set-up. The previous studies of high-spin states in  $^{221}\text{Th}$  have used the  $^{208}\text{Pb}(^{16}\text{O},3n)$  reaction [Dahlinger85][Dahlinger88][Tandel13]. The previous studies of  $^{224}\text{Th}$  have either used the  $^{226}\text{Ra}(\alpha,6n)$  reaction or the  $^{208}\text{Pb}(^{18}\text{O},2n)$  reaction. Here, we have chosen to use the  $^{208}\text{Pb}(^{18}\text{O},5n)^{221}\text{Th}$  and  $^{208}\text{Pb}(^{18}\text{O},2n)^{224}\text{Th}$  reactions, at 110 MeV and 78 MeV, respectively. These reactions have been chosen for several reasons. Firstly, the  $^{208}\text{Pb}(^{18}\text{O},2n)$  reaction has previously been shown to successfully populate excited states in  $^{224}\text{Th}$  [Schwartz86]. Secondly, both reactions have favourable cross sections calculated by the statistical model codes PACE [PACE] and Cascade [Cascade]. And

thirdly, use of the same beam and target combination will allow a swift changeover between different parts of the experiment (by change of beam energy alone).

The cross sections used to assess the viability of the proposed experiment (discussed later) are based on values predicted by statistical-model codes along with our own experience of similar reactions induced by oxygen beams on lead targets. The statistical-model calculations suggest that about 95% of the total cross section in each case will result in prompt fission. We will therefore use the Recoil Filter Detector (RFD) [Męczyński07] to detect evaporation residues in coincidence with prompt  $\gamma$  rays and x rays detected in Galileo.

We intend to supplement the experimental set up with 10 or 12 small-volume  $\text{LaBr}_3(\text{TI})$  scintillator detectors, in order to test the feasibility of lifetime measurements using direct-timing methods. It is expected that the lifetimes of the states below about  $10^+ / 11^-$  in the yrast octupole bands will have lifetimes that can be measured by direct timing (i.e.  $> 10$  ps). For example, the nucleus  $^{220}\text{Th}$  will be produced with a relatively large cross-section in the proposed experiment. An alternating-parity band has been identified in  $^{220}\text{Th}$  to over spin  $20 \hbar$  [Reviol06] yet there are no measurements of lifetimes of any of the excited states in that nucleus.

We will use a  $^{18}\text{O}$  beams with energies of 78 and 110 MeV, and with an intensities of 10 pnA. The beams will be incident upon  $^{208}\text{Pb}$  targets of thickness  $1.0 \text{ mg cm}^{-2}$ . From a consideration of count rates (next section) we request **7 days** of beam time to complete the proposed measurement. This will be 5 days with beam energy 78 MeV (to study  $^{224}\text{Th}$ ) and 2 days with beam energy 110 MeV (to study  $^{221}\text{Th}$ ).

### Rate estimates

To estimate the rates, we have used the following parameters. The efficiency of Galileo with 25 detectors was 2.4%; here, it has been scaled up for the 35-detector configuration to 3.3%.

Efficiency of Galileo at 1333 keV	3.3%
Average efficiency of Galileo in the 100 – 800 keV range	~4%
Euclides $\alpha$ -particle detection efficiency (measured in experiment 16.30)	30%
Recoil-filter detector recoil detection efficiency	30%
Beam intensity	10 pnA
Target thickness	$1 \text{ mg cm}^{-2}$
Cross section for the $^{208}\text{Pb}(^{18}\text{O}, 2n)^{224}\text{Th}$ reaction	1 mb
Cross section for the $^{208}\text{Pb}(^{18}\text{O}, 5n)^{221}\text{Th}$ reaction	10 mb
Cross section for the $^{208}\text{Pb}(^{16}\text{O}, 6n)^{220}\text{Th}$ reaction	10 mb

**Table 1:** Parameters used in the rate estimates.

**$^{224}\text{Th}$ :** We will produce  $180 \text{ }^{224}\text{Th}$  recoils per second, which is 16 million per day. If the RFD detects 30% of the recoils, then we will detect 4.7 million  $^{224}\text{Th}$  recoils per day. Assuming a  $\gamma$ -ray multiplicity of 10, then in a five-day experiment we will collect approximately  $1.3 \times 10^6$  recoil- $\gamma\gamma$  events and  $1.3 \times 10^5$  recoil- $\gamma\gamma\gamma$  events.  **$^{221}\text{Th}$ :** The cross section for  $^{221}\text{Th}$  is ten times larger than that for  $^{224}\text{Th}$ . If we assume that the  $\gamma$ -ray multiplicity is the same for both nuclei, but the experiment is run for two days instead of five, the number of events will scale by a factor of four. Therefore, in a two-day experiment, we will collect  $5 \times 10^6$  recoil- $\gamma\gamma$  events and  $5 \times 10^5$  recoil- $\gamma\gamma\gamma$  events. It should be noted that these rate estimates do not include the unfolding of higher-fold  $\gamma$ -ray coincidence events, and that the RFD transmission efficiency is approximate. The  $\alpha$ -particle detection efficiency of Euclides was measured in a previous experiment (with a  $^{16}\text{O}$  beam on a  $^{208}\text{Pb}$  target) to be 30%.

Euclides will be used for additional channel selection for residues produced by  $\alpha$ -particle evaporation, such as  $^{221}\text{Ra}$  ( $\alpha n$ ) and  $^{220}\text{Ra}$  ( $\alpha 2n$ ).

## Summary

In summary, we request **7 days** of beamtime with an  $^{18}\text{O}$  beam at energies of **78 MeV** and **110 MeV** in order to carry out detailed  $\gamma$ -ray spectroscopy of the octupole-deformed nuclei  $^{221}\text{Th}$  and  $^{224}\text{Th}$ . We will investigate octupole to quadrupole shape transitions that have been predicted for these nuclei. As a subsidiary aim, we will also study the structure of other nuclei such as  $^{220}\text{Th}$  and  $^{221}\text{Ra}$ . We will use the Galileo  $\gamma$ -ray spectrometer, in conjunction with Euclides and the Recoil Filter Detector. The nucleus  $^{224}\text{Th}$  will be produced in the  $^{208}\text{Pb}(^{18}\text{O}, 2n)$  with a beam energy of 78 MeV, for which the cross section is expected to be around 1 mb. The nucleus  $^{221}\text{Th}$  will be produced in the  $^{208}\text{Pb}(^{18}\text{O}, 5n)$  with a beam energy of 110 MeV, for which the cross section is expected to be around 10 mb. Gamma-rays in Euclides will be detected in coincidence with recoils detected in the Recoil Filter Detector. This will allow the separation of  $\gamma$  rays emitted by the evaporation residues from the large background of  $\gamma$  rays from prompt fission fragments.

Our collaboration has significant experience of such measurements with Gammasphere, the Microball and HERCULES. We have previous experience of two experiments with Galileo and Euclides which will help to expedite the data analysis and extraction of results.

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