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Search for Octupole Correlations in Odd-Mass Radium and Radon Isotopes

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Run	Ion	Energy (MeV)	Current (pnA)	Target	Setup / beamline	Days
1	^{136}Xe	833 MeV	2 pnA	^{232}Th	AGATA+PRISMA+DANTE	7

Higher order collective degrees of freedom are well-established in the composition of excited quantum states of the atomic nucleus. Octupole collectivity, associated with pear-shaped nuclei, has been the subject of intense theoretical and experimental study. Regions of octupole deformation are found above all the major shell gaps — the so-called octupole magic numbers — $Z, N = 34, 56, 88, 134$. Effects attributed to octupole collectivity are particularly prominent in the light-actinide region, confirmed by numerous studies on even-even isotopes of the U, Th, Ra, Rn, isotopic chains. However, experimental information on odd-mass isotopes in this mass region are sorely lacking. The odd-mass candidates are of particular interest due to the enhancement of octupole effects arising from the interaction of the unpaired nucleon with the deformed core. These systems also play a central role in atomic electric dipole moment (EDM) searches, where the static octupole deformation and presence of low-lying parity doublets significantly enhance the nuclear Schiff moment. **We request 7 days of beam time bombarding a 2 mg/cm^2 ^{232}Th target with an 833 MeV ^{136}Xe beam, with the objective of populating odd-mass radon and radium isotopes via multinucleon transfer reactions. A particular focus is placed on ^{223}Rn which is expected to exhibit a higher degree of octupole collectivity compared to its neighbours. Measurements of experimental observables easily derived from energy level schemes (Δi_x, $|D_0/Q_0|$) will provide immediate insight into the strength and type of octupole collectivity in these nuclei.** The precise atomic mass and charge selectivity offered by PRISMA is expected to overcome previous experimental shortcomings in being unable to identify γ rays belonging to the odd mass candidates.

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AGATA + PRISMA + DANTE

1 Motivation

In recent years, a focus within nuclear structure physics has been to understand how higher-order collective degrees of freedom influence the properties of the atomic nucleus. The shape of the nuclear equipotential surface can be expressed as an expansion of spherical harmonics, $Y_{\lambda\mu}(\theta, \phi)$, where each multipole term corresponds to a distinct pattern of deformation [1, 2]. Quadrupole ($\lambda = 2$) deformation is responsible for familiar reflection-symmetric prolate and oblate shapes, and is recognised as a dominant driver of collective behaviour [3]. However, particular combinations of proton and neutron numbers favour the emergence of collective octupole ($\lambda = 3$) modes of excitation [4, 5]. These components introduce reflection-asymmetric shapes to the nuclear potential surface, and their presence has profound implications for the structure of rotational bands within these nuclei [6, 7], their electromagnetic transition strengths [7, 8, 9, 10], and violation of fundamental nuclear symmetries [5, 11, 12].

As a result, nuclei exhibiting octupole-deformed shapes have been the subject of intense experimental and theoretical study and some general properties of octupole-deformed nuclei are universally recognised. For instance, the main experimental observable is the presence of a

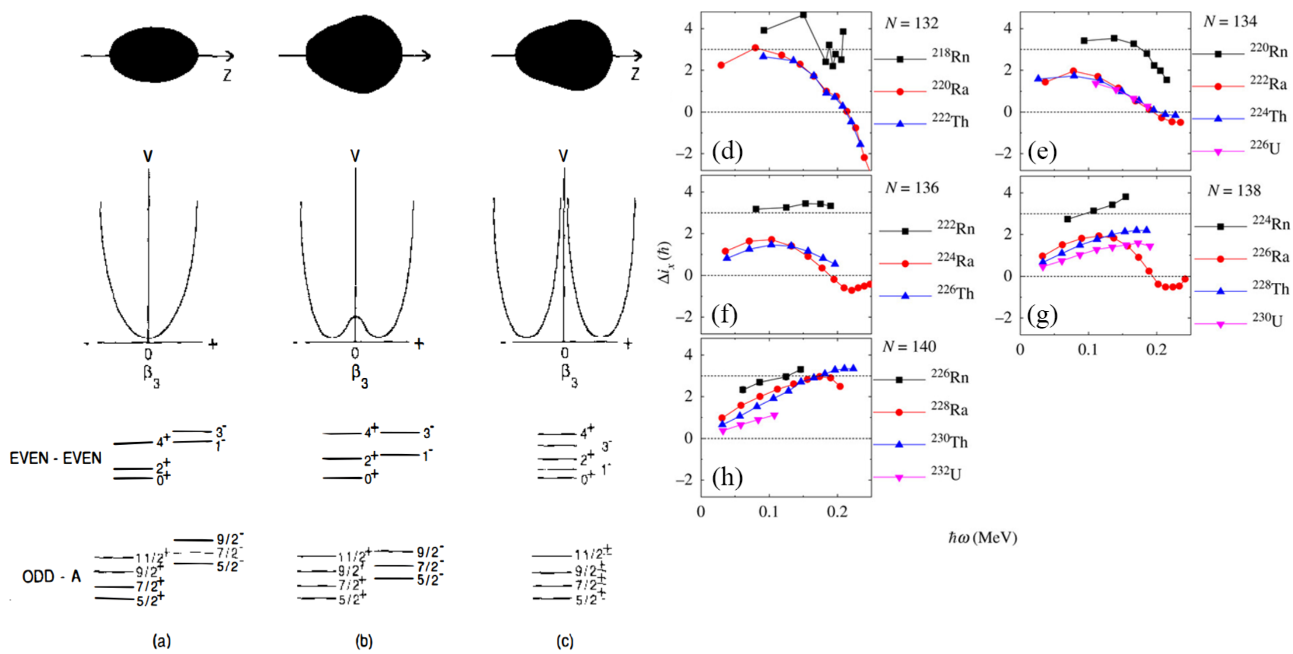

Figure 1: Panels (a)—(c) display schematical energy level spectra and potential energy versus β_3 deformation plots for different axially symmetric ($K = 0$) shapes. Panel (a) represents a rigid spheroidal nucleus, which is axially as well as reflection symmetric. Panel (c) shows a rigid pear shape nucleus, which is axially symmetric but reflection asymmetric with respect to the XY plane. Panel (b) represents a soft pear shape nucleus, with $\beta_2 \approx 0.15$ and $\beta_3 \approx 0.09$. Figure and caption obtained from Ref. [4].
Panels (d)—(h) display the measured differences in aligned angular momentum between simplex partners in a multitude of even-even nuclei in the light-actinide region as a function of rotational frequency. The expected vibrational- and collective- octupole limits ($\Delta i_x = 0,\ 3\hbar$, respectively) are imposed on each plot. Figure obtained from Ref. [5].

low-lying negative-parity band with $K^\pi = 0^-$ in even-even nuclei, arising from the violation of parity symmetry under rotation [6]. The two bands arise from the same intrinsic configuration, and are labelled by the *simplex* quantum number. In the odd-mass case, the addition of an unpaired nucleon to the octupole and quadrupole deformed system introduces some dramatic effects on the level scheme. The odd nucleon with good-K (or signature, α) couples to either simplex partner in the even-even core. As a result, the familiar two-band pattern of an octupole rotor expands to four $\Delta I = 2$ rotational sequences (two positive- and two negative-parity). The two sets of bands of opposing simplex are termed "parity doublets" owing to their near degeneracy in energy. The opposite parity bands in both members of the doublet are connected by strong E1 linking transitions [13, 14]. The expected impact on the energy level schemes of octupole deformation of varying strength is displayed schematically in Figures 1(a)—(c).

The emergence of a reflection-asymmetric shape also has consequences beyond nuclear structure. In particular, the static separation of charge implied by octupole deformation gives rise to a non-zero nuclear Schiff moment — the lowest-order observable electric moment not screened by atomic electrons. This moment is only generated in the presence of parity- and time-reversal-violating interactions and is therefore a direct probe of physics beyond the Standard Model. The nuclear Schiff moment is defined as

$$S \;=\; \langle \Psi_0 \,|\, \hat{S}_z \,|\, \Psi_0 \rangle \;=\; \sum_{i \neq 0} \frac{\langle \Psi_0 | \hat{S}_z | \Psi_i \rangle \; \langle \Psi_i | \hat{V}_{PT} | \Psi_0 \rangle}{E_i - E_0}\,, \tag{1}$$

where the summation is over excited states, $\hat{S}_z = \frac{e}{10} \sum_{p=1}^{Z} \big(r_p^2 - \frac{3}{5} \langle r^2 \rangle_{\rm ch} \big)\, z_p$ and $\hat{V}_{PT}$ is the P,T-violating nucleon–nucleon potential [12]. Equation (1) shows how the Schiff moment — and therefore the atomic electric dipole moment — is enhanced by the octupole shape. To start, a permanent pear shape carries a static charge-dipole density which appears in the off-diagonal matrix element $\langle \Psi_0 | \hat{S}_z | \Psi_{1^-} \rangle \;\propto\; e\beta_2 {\beta_3}^2 Z A^{2/3}$, where β_2 and β_3 are the quadrupole and octupole deformation parameters [15]. In contrast, in the spherical (or purely quadrupole) case the only nonzero matrix elements $\langle \Psi_0 | \hat{S}_z | \Psi_i \rangle$ arise from small-amplitude octupole vibrations (one–phonon excitations), and are several orders of magnitude smaller — typically of order $\sim 10^{-8}\, \eta\, e\, \mathrm{fm}^3$ [16] instead of $\sim 10^{-5}\, \eta\, e\, \mathrm{fm}^3$ in the octupole and quadrupole deformed case [15] (where η is the coefficient in the T- and P-odd interaction Hamiltonian). The second enhancement comes from the energy denominator. In a static octupole rotor, the first 1^- state is brought down in energy relative to the ground state, whereas in a spherical nucleus that same state is typically 1 MeV above the ground state. Hence the small gap $E_{1^-} - E_0$ in an octupole-deformed system gives a large factor $1/(E_{1^-} - E_0)$.

Finally, we can answer the question: why focus on the odd-mass nuclei? To understand why the odd-mass nuclei are the flagship cases for measuring a non-zero atomic EDM, we must examine the core-polarising influence of the odd particle coupling to the octupole-deformed core. Because the unpaired nucleon blocks a quasiparticle state, the pairing energy that separates the positive- and negative-parity configurations in an even-even core disappears; in formal terms the parity splitting of the odd-A doublet is reduced by the single-particle parity expectation value $\langle P_{\rm s.p} \rangle$ and can even vanish [14]. As a result, odd-A octupole nuclei exhibit near-degenerate parity doublets with $\Delta E \approx$ 10-60 keV. Moreover, the unpaired nucleon further mixes the two opposite-parity core states, generating an admixture coefficient $\alpha \propto 1/\Delta E$. This P,T-odd mixing, that arises from CP violation in the nuclear force (as discussed above), orients the intrinsic symmetry axis along the spin $\langle n_z \rangle \neq 0$, giving the nucleus a preferred "direction" in the intrinsic frame. This orientation removes the left–right symmetry of the octupole shape in the lab frame, resulting in a static reflection-asymmetric configuration and enhancing the intrinsic Schiff moment, and therefore the Schiff moment in the laboratory frame [17]. The combined effect of these two factors yields a Schiff moment up to $10^2 - 10^3$ times larger than in spherical systems such as ^{199}Hg [18, 19].

Despite the intense theoretical and experimental interest in odd-mass octupole-deformed nuclei, their structure remains comparatively less well studied than their even-even counterparts. The even-even nuclides around the light-actinide and trans-lead regions represent the bulk of extant experimental information — particularly at high spin. Indeed, rotational bands of opposing parity — which interleave at modest rotational frequencies — have been observed in the even-even $^{222-230}$Th, $^{220-228}$Ra and $^{218-226}$Rn isotopes. Readily obtainable from their energy level schemes are experimentally derived quantities that yield direct insight into the octupole collectivity of these nuclei. One such example is the difference in aligned angular momentum between simplex partners: Δi_x. Measurements of this quantity for a multitude of even-even light actinides are displayed in Figures 1d—h. A clear difference is observed between the radon and radium isotopes, with the former generally staying close to the vibrational limit $\Delta i_x = 3\hbar$ and the latter tending towards the static octupole deformed limit of $\Delta i_x = 0\hbar$ with increasing rotational frequency. A further useful quantity is the absolute magnitude of the ratio of the intrinsic electric dipole and electric quadrupole moments, $|D_0/Q_0|$, which is obtained from measurements of $B(E1)/B(E2)$ values for the levels in the simplex partners connected by interband E1 transitions [9]. In a simple picture, a large $|D_0/Q_0|$ ratio implies a reflection-asymmetric charge distribution, indicative of an octupole shape; one must be careful to adequately calibrate for the quadrupole moment, however. Measurements of this quantity were used to thoroughly characterise octupole collectivity in even-even radium and radon isotopes in Refs [20, 21], which is discussed in the following section. A more reliable indicator of collective octupole deformation is obtained by measurement of the E3 moment from Coulomb excitation experiments. Such measurements have been performed for even-even radium and radon nuclei, indicating significant octupole collectivity in these nuclei [10, 22, 23].

The rich variety of experimental information for the even-even nuclei stands in contrast to the general dearth of information on the odd-mass cases. Among the odd-mass octupole-deformed Radium isotopes, the low-lying states are established and well-studied [24, 25, 26]. ^{225}Ra emerges as the present best candidate for the search for a non-zero atomic EDM owing to the existence of a low-lying ($\Delta E = 50$ keV) parity doublet in this nucleus [12]. Nevertheless, comprehensive high-spin data remain largely absent for odd-mass Ra isotopes, limiting our ability to constrain and test reflection-asymmetric models at higher angular momenta [13, 27]. For the odd-mass Radon isotopes, the situation is even more severe. No excited states whatsoever are known for the isotopes ^{221,223,225}Rn, save for a tentatively assigned state at 30 keV in ^{221}Rn originating from an alpha-decay study [28]. As for the radium chain, the even-even radon isotopes are comparatively well-studied and display predominantly vibrational octupole collectivity [29, 20]. However, theoretical approaches posit that the interaction with the unpaired nucleon induces core polarisation, leading to a static octupole deformation of the nuclear shape [30]. It is clear that new data is required to alleviate the ambiguity prevailing over the odd-mass nuclei in this mass region. Immediate clarity can be obtained from experimentally derived quantities Δi_x and $|D_0/Q_0|$ which, in turn, are easily obtained from even the first few excited states in parity doublet bands. Such data is not only invaluable for constraining the (sometimes contradictory) theoretical approaches, but could represent results impactful in fields of physics beyond nuclear structure. **Thus, the primary objective of this experiment is to elucidate the low-to-mid-spin structures in the odd-mass ^{221,223,225}Rn isotopes with the aim of unambiguously determining the nature of their octupole collectivity. A particular focus is placed on ^{223}Rn which is expected to host the lowest lying parity doublet in the radon isotopic chain.** The secondary objectives of this experiment are to extend the level schemes of all observed odd-mass radon and radium nuclei to high spin, answering questions about the robustness of their octupole shapes under influence of competing alignments and the quenching of octupole deformation at high spin.

1.1 Cocks, *et al.*

In the mid-1990s, a seminal series of experiments designed to investigate octupole deformation in this mass region were performed by J.F.C. Cocks and his collaborators (see Refs [20, 21]). The purpose of the initial experiments was to investigate a multitude of multinucleon transfer reactions to see which yielded the highest production cross sections for octupole deformed nuclei. Variously, ^{56}Fe, ^{86}Kr and ^{136}Xe beams were used to bombard a thick (30 mg/cm^2) ^{232}Th target of thickness at beam energies of 362, 511 and 830 MeV, respectively. Through measurement of γ-ray yields it was determined that the ^{136}Xe beam offered the best production cross sections. That reaction was repeated in a second experiment using the GAMMASPHERE spectrometer and resulted in the observation of high-spin states in ^{222,224,226}Ra and ^{218,220,222}Rn. The experiment was a resounding success, confirming octupole vibrational nature for the radon isotopes and static collectivity for the radium isotopes. Additionally, the measured ratios $|D_0/Q_0|$ for these isotopes remained constant for ^{222,226}Ra across the entire measured spin range and very small for ^{224}Ra, suggesting robust and stable charge and mass distributions which are insensitive to rotational effects. Measurements for ^{222}Rn yielded an appreciably large intrinsic dipole moment, $|D_0| = 0.10(2)$ e fm, comparable to those for the radium isotopes $D_0 \sim 0.2$—0.3 e fm. This measurement implies that ^{223}Rn may present the best offering for low lying octupole collectivity. However, one must be careful when using such an observation as the sole indicator of an octupole deformed shape. A stark example is the near-zero intrinsic dipole moment measured for ^{224}Ra, $|D_0| = 0.030(1)$ e fm. ^{225}Ra, which represents one unpaired neutron coupled to this core configuration, then displays the lowest lying parity doublet band in the entire radium chain! As discussed in the previous section, measurement of the E3 moment is a more reliable indicator of octupole deformation since it is much less sensitive to single-particle effects.

A notable shortcoming of this experiment was the lack of measurements for the odd-mass nuclei, and ^{224,226}Ra. At the time, no excited states were established for these nuclides, precluding the use of tagging techniques based on known level schemes during the analysis. A new experiment using the PRISMA magnetic spectrometer circumvents this issue by providing a clean and direct event tag on atomic number (Z) and mass (A).

1.2 LNL Exp 22.096 (Search for U228)

In May 2024 an experiment was performed at LNL bombarding a 2 mg/cm^2 ^{232}Th target with a 950 MeV beam of ^{129}Xe ions, with the aim of measuring the first excited states of ^{228}U. Analysis of this data is ongoing, with preliminary results indicating that the production cross section of ^{228}U in this reaction is perhaps smaller than anticipated, with large fission background providing a challenge to the analysis. The newly proposed experiment should offer an easier measurement. Firstly, PACE4 calculations performed using various primary target-like pre-evaporation (Th, Ra, Rn) MNT residues as the compound nucleus indicate fission occurs less than 1% of the time for excitation energies not exceeding 40 MeV (obtained from the maxima of E^* distributions in GRAZING). This is reinforced with calculations by Moller, *et al.,* which show the fission barrier is around twice as high for the radium and radon isotopes of interest, relative to the neighbouring uranium isotones [31]. Furthermore, qualitative evidence of the cleanliness and proof of the efficacy of the ^{136}Xe ($E_{\mathrm{beam}} = 833$ MeV) + ^{232}Th reaction is offered by the experiment of Cocks, *et al.,* showcasing excellent peak-to-background over a broad range of energies reaching into the high-spin domain ($\sim 30\hbar$).

2 Experimental Procedure, Rate Estimates and Beam Time Request

The experiments of Cocks, *et al.,* showcased the efficacy of the ^{136}Xe (E_{beam} = 833 MeV) + ^{232}Th reaction for producing neutron-rich light actinides. The measured production cross sections for various even-even nuclei using this reaction were published in Ref. [21] and are reproduced here in Figure 2. Since cross-section estimates are not available for the odd-mass isotopes, these have been estimated by extrapolating the experimental results for the even-even isotopes. The predicted production cross section for the primary experimental objective nucleus, ^{223}Rn, is 0.47 mb. Assuming a beam intensity of 2 pnA and a target thickness 2 mg/cm^2, the predicted ^{223}Rn yield is 2.8×10^6 particles per day. The grazing angle, calculated using the code REACTION, is $\theta_{gr} = 55.6°$. With PRISMA set at this angle and assuming a transmission efficiency of 5% the amount of ^{223}Rn detected by PRISMA is 1.4×10^5 particles per day. Finally, assuming an average AGATA efficiency of 8% and an average γ multiplicity $N_\gamma = 10$ we obtain a **yield of AGATA—$\gamma\gamma$+PRISMA ^{223}Rn coincidences of** 2.7×10^4 **per day.** It is assumed in all of these calculations that contribution from fission is negligible. This is supported qualitatively by the cleanliness of the spectra obtained in the Cocks experiment, and quantitively by PACE4 calculations (discussed in the previous section). Relative to the Cocks experiment, the level of counting statistics is expected to be modified by the following factors: greater beam intensity ($\times 4$), PRISMA transmission efficiency ($\times 0.05$) and AGATA efficiency ($\times 0.8$ for singles, $\times$ 0.7 for doubles, $\times$ 0.4 for triples). Though our target thickness is thinner than the 36 mg/cm^2 target utilised by Cocks, *et al.,* this is not expected to substantially affect the yield since the beam energy is expected to fall below the fusion barrier (750 — 820 MeV) within the 2 mg/cm^2 target (dE/dx = -30 MeV/(mg/cm^2), obtained from SRIM.) **Accounting for all of these factors, it is expected a similar level of γ-ray counting statistics can be obtained in 7 days of beam time, but overcoming the limitations of prior experiments by including the enhanced selectivity offered by PRISMA.** Data obtained from this experiment will be analysed as part of the PhD project for Liverpool student Henry Hilton.

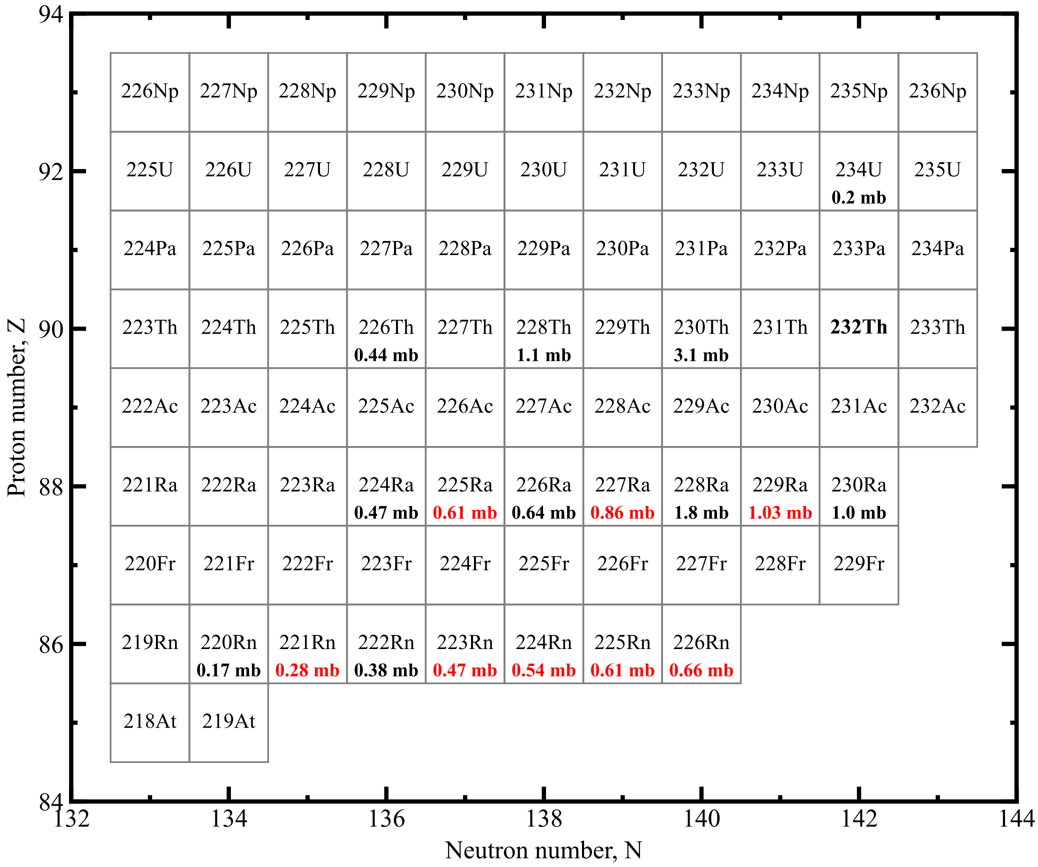

Figure 2: Subsection of the nuclear chart focused on the neutron-rich, trans-lead, light-actinide region. Measured (black text) and extrapolated (red text) production cross sections for various nuclides produced in a ^{136}Xe + ^{232}Th multinucleon transfer reaction are overlaid on the figure, with measured values obtained from Ref. [21].

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