NEW VISTAS OF EXOTIC HEAVY NUCLEI * **

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We report studies of examples of reflection-asymmetric nuclei which are difficult to access using compound nucleus reactions. The most octupole deformed nuclei should be uranium isotopes with $N \approx 132$; preliminary measurements of these very fissile nuclei suggest that they are within reach of current spectroscopic techniques. The octupole radium isotopes with $N > 132$ and radon isotopes are not accessible by reactions employing stable targets and beams; we have shown that multinucleon transfer reactions can populate these nuclei with sufficient yield for their structure to be determined. We report high spin studies in ^{218,220,222}Rn and ^{222,224,226}Ra which reveal upbending effects in this mass region for the first time and show that the electric dipole moment is constant with spin.

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1. Introduction

The region of the periodic chart which has shown the best evidence for octupole instability in the nuclear ground state is around $Z = 88 - 90$ and $N \approx 134$ [1, 2]. These nuclei have low-lying negative parity states and relatively strong $B(E1)$ values for the transitions between the bands of opposite parity; in the case of ^{226}Ra large $B(E3)$ values have been measured consistent with its interpretation as a rotating pear shape [3]. The very inaccessibility of these far-from-stability nuclei has, however, meant that there are large gaps in our knowledge of octupole effects in heavy nuclei. Complete measurements of the high-spin behaviour of the yrast octupole band only exist for the isotopes of Th. For the Ra isotopes such measurements are available for the weakly quadrupole coupled ^{218,220}Ra and the strongly coupled ^{226}Ra. There is only a limited amount of data on ^{224}Ra

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and virtually no information exists for ^{222}Ra. The U isotopes with $N \approx 132$ are predicted to have the largest octupole correlations [4], but there is no information about the low lying structure of these nuclei.

2. Preliminary study of octupole U nuclei

The nuclei predicted to have the deepest minimum in potential energy at a non-zero value of octupole deformation are uranium isotopes with $N \approx 132$. These fissile nuclei are accessible via compound nucleus reactions but the survival cross sections are very small [5]. In order to determine the low-lying structure of these nuclei using in-beam spectroscopy, it is necessary to detect and identify the associated evaporation residue nuclei. We carried out a test experiment to determine whether spectroscopic measurements could be made of nuclei in this charge and mass region. We used the technique of α-decay tagging, in which the recoiling heavy nucleus is separated from the beam and allowed to implant into a position-sensitive detector. The measurement of the subsequent α-decay is associated with the recoil by position and slow timing, and the recoil is associated with the prompt γ-ray emission by fast timing. The experiments were carried out at the University of Jyväskylä and employed 8 TESSA Compton-suppressed spectrometers to detect the prompt γ-ray emission and a silicon strip detector placed at the focal plane of the RITU gas-filled separator to detect the recoil and subsequent α-decay. This technique has been described in more detail elsewhere in these proceedings [6]. We employed the reaction ^{208}Pb(^{22}Ne, 4n)^{226}U at a bombarding energy of 112 MeV; this has a measured cross-section [5] of approximated 5μb. Data were collected for approximately 12 hours; in this time 250 events were collected in which a 2 α particle chain corresponding to the $^{226}\mathrm{U} \rightarrow {}^{222}\mathrm{Th} \rightarrow {}^{218}\mathrm{Ra}$ sequence was detected. The prompt γ-ray spectrum associated with the $^{226}\mathrm{U} \rightarrow {}^{222}\mathrm{Th}$ decay sequence contained 60 counts. It is clear that although these statistics are insufficient, the lowest transitions will be identified if a more efficient germanium detector array is provided (a factor of 5 is easily achievable) and the experiment run for a longer time (another factor of 10).

3. Population of octupole nuclei using transfer reactions

The development of efficient Compton-suppressed germanium-detector arrays has allowed multi-nucleon transfer reactions to become an effective method of accessing many nuclei inaccessible by compound-nucleus reactions [7–9]. In these reactions many excited nuclei around the projectile and target are produced although the resulting complexity of the spectra demands high statistics and good energy resolution. This can be achieved by the use of efficient germanium-detector arrays in conjunction with the use of thick targets so that the decays from low-lying yrast states are emitted from nuclei at rest. This approach, in which γ–γ coincidence techniques are employed, can allow precise identification of the two reaction partners and measurement of their yield.

Initial studies were performed at the Accelerator Laboratory in Jyväskylä, Finland. A thick (30 mg/cm^2) ^{232}Th target was bombarded by a ^{56}Fe beam from the K-130 accelerator at an energy of 362 MeV. The γ–γ in-beam and out-of-beam coincidence data were collected with an array of twelve Compton-suppressed TESSA-type germanium detectors.

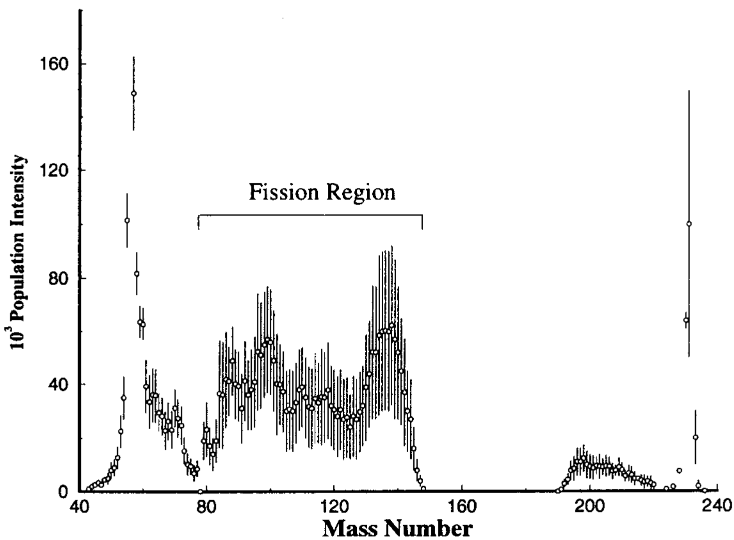

Fig. 1. Population yield as a function of mass number for nuclei populated in the ^{56}Fe + ^{232}Th reaction. This was produced by summing production yields for different mass numbers.

In these experiments we observed that population is dictated by the mass and charge equilibration processes so that the N/Z value of the populated nuclei tends to that of the composite system. It is also evident that many nuclei are produced via fission of the target-like transfer products. Figure 1 shows the mass distributions of the nuclei populated in the ^{56}Fe + ^{232}Th reaction. The yields are from quantitative in-beam and out-of-beam γ–γ coincidence analyses, where the intensities are corrected for efficiency and internal conversion. Near the projectile and target we can see a significant yield of nuclei produced by transfer of neutrons from quasi-elastic processes. The yield of target-like nuclei corresponding to multi-nucleon (deep inelastic) transfer is substantially reduced compared to the corresponding projectile-like partner. The minimum yield appears to occur near A~224. The two peaks evident at A~90 and A~140 presumably arise from asymmetric fission of near-target products of quasi-elastic processes, with an additional component arising from fission of the compound nucleus.

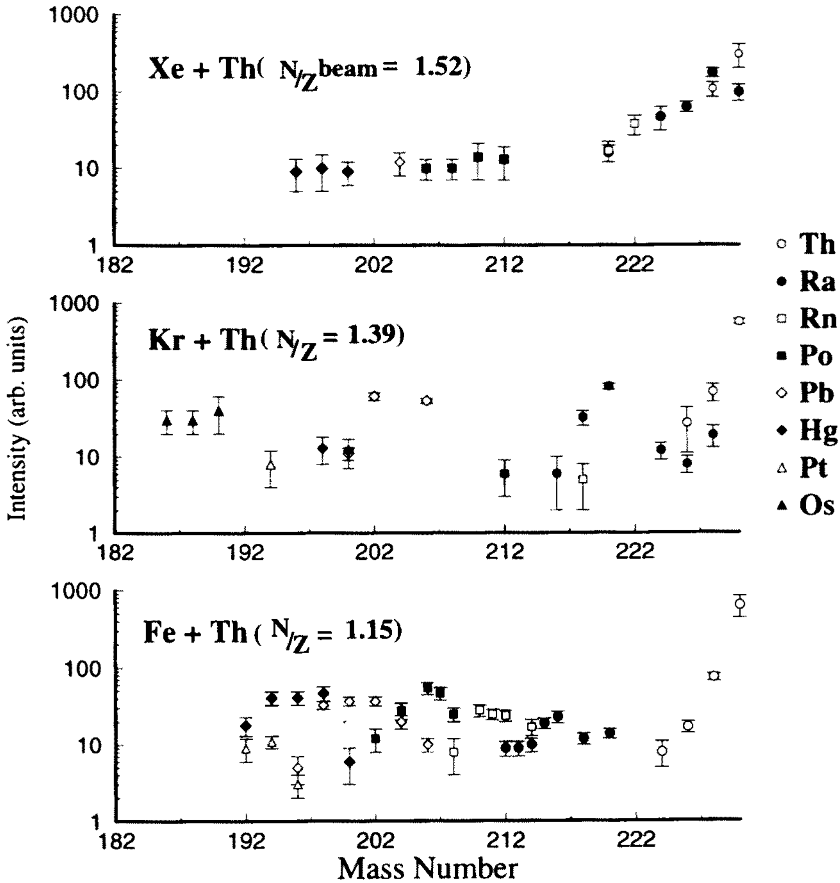

Fig. 2. A comparison of yields of target-like nuclei produced in the ^{56}Fe + ^{232}Th, ^{86}Kr + ^{232}Th and ^{136}Xe + ^{232}Th reactions.

In two additional experiments of this nature a ^{232}Th target was bombarded by a 511 MeV ^{86}Kr beam from the Jyväskylä facility and a similar target was bombarded by a ^{136}Xe beam from the ATLAS accelerator of the Argonne National Laboratory at a beam energy of 830 MeV. In each experiment both in-beam and out-of-beam γ–γ coincidences were stored. As with the ^{56}Fe + ^{232}Th reaction it was observed that the mass and charge equilibration processes control the population of nuclei in this system. A comparison of the target-like products produced in the three reactions (see figure 2) indicates that the ^{136}Xe + ^{232}Th reaction can populate the nuclei of interest in the octupole-deformed light-actinide region.

4. High spin studies of octupole Rn and Ra nuclei

As the reaction ^{136}Xe + ^{232}Th offered the largest yield for Rn and Ra isotopes with $N \approx 134$ (see previous section), we chose this reaction in order the make spectroscopic measurements of ^{218,220,222}Rn and ^{222,224,226}Ra The ^{136}Xe projectile was accelerated to an energy of 833 MeV by the 88" cyclotron at the Lawrence Berkeley National Laboratory. This bombarded a ^{232}Th target of thickness 36 mg/cm^2. De-excitation gamma rays emitted from reaction products were collected with the GAMMASPHERE gamma-ray spectrometer which consisted of 73 large volume (~75% relative efficiency) Compton-suppressed germanium detectors [10, 11], 28 of which were segmented [12]. After 49 hours of collecting gamma-ray events of fold 3 or higher, subsequent unpacking of events revealed a total of 1.1×10^{10} triple and 6.8×10^9 fourfold Compton-suppressed gamma-ray coincidences.

The data were analysed using γ-γ-γ-correlation matrices. Of the 6 nuclei whose properties are discussed here, ^{224}Ra was populated with the greatest intensity and was observed to the highest spin. Fourfold events which contained a gamma ray which passed a gate on the 4^+ to 2^+ transition in ^{224}Ra were used to analyse this nucleus. Fourfold coincidences were not used to study the other 5 nuclei as no significant improvement in peak-to-background over threefold coincidences was observed in these cases.

The typical spectra shown in figure 3 serve to illustrate the quality of these data. Figure 3(a) is a threefold gamma-ray spectrum which shows transitions in ^{218}Rn. The spectrum was generated by double-gating in the γ-γ-γ matrix on all transitions between positive-parity states which are shown in the spectrum. Figure 3(b) is a fourfold spectrum which shows gamma-ray transitions above and including the 6^+ to 4^+ transition in the ground state rotational band of ^{224}Ra. This was generated by double-gating in the gated γ-γ-γ matrix on all transitions marked in the spectrum. The inset to this figure shows the efficiency and internal-conversion corrected intensities of the depopulating transitions above spin $6\hbar$ in this nucleus. These intensities are normalized to that of the 4^+ to 2^+ transition. These intensities were not directly measured from the spectrum, but were carefully determined using several different uncontaminated gates.

The level schemes of ^{218}Rn, ^{220}Rn and ^{222}Rn which are shown in figure 4(a) were based on previous spin and parity assignments to the low-spin states (see reference [13] and references therein) and for higher spins, energy sums and intensity balance arguments. The dashed-line boxes on each decay scheme contain states which were observed in previous work. The level schemes have been significantly extended in the present work, and in each nucleus two interleaving rotational bands have been observed for the first time.

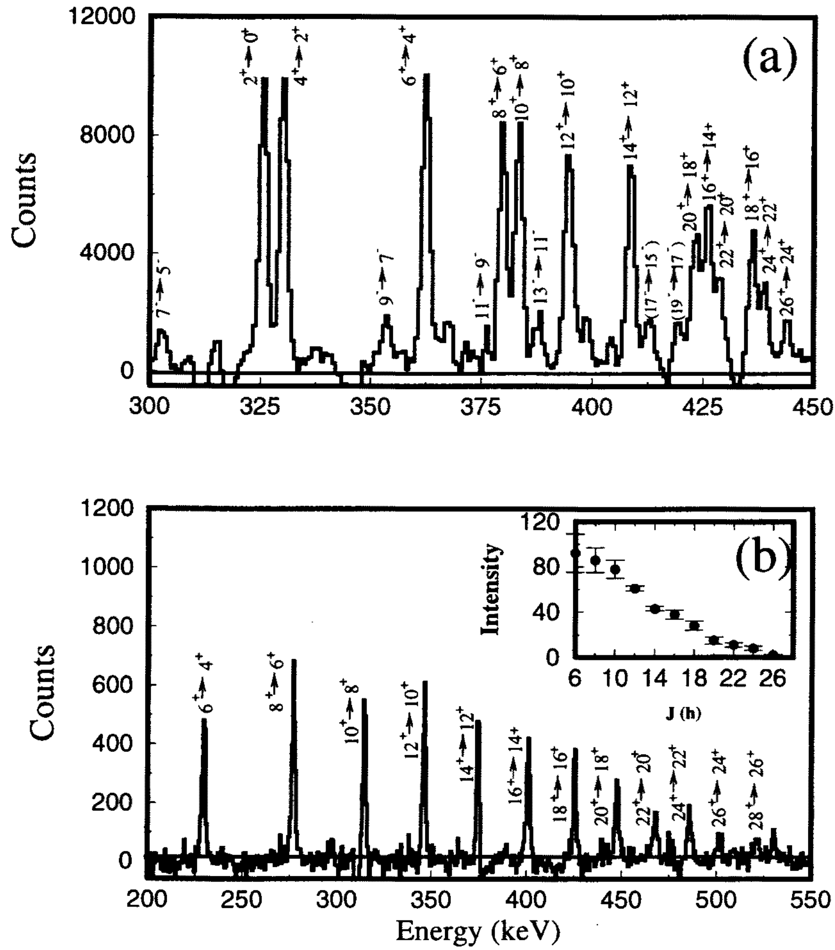

Fig. 3. (a) Threefold gamma-ray spectrum showing transitions in ^{218}Rn. (b) Gamma-ray spectrum showing transitions above and including the 6^+ to 4^+ transition in ^{224}Ra. The spectrum is from unpacked fourfold coincidence events where one of the gamma rays has the same energy as the 4^+ to 2^+ transition in ^{224}Ra. The inset is a plot of the intensities of the depopulating transitions in ^{224}Ra as a function of spin. The intensities are normalized to that of the 4^+ to 2^+ transition.

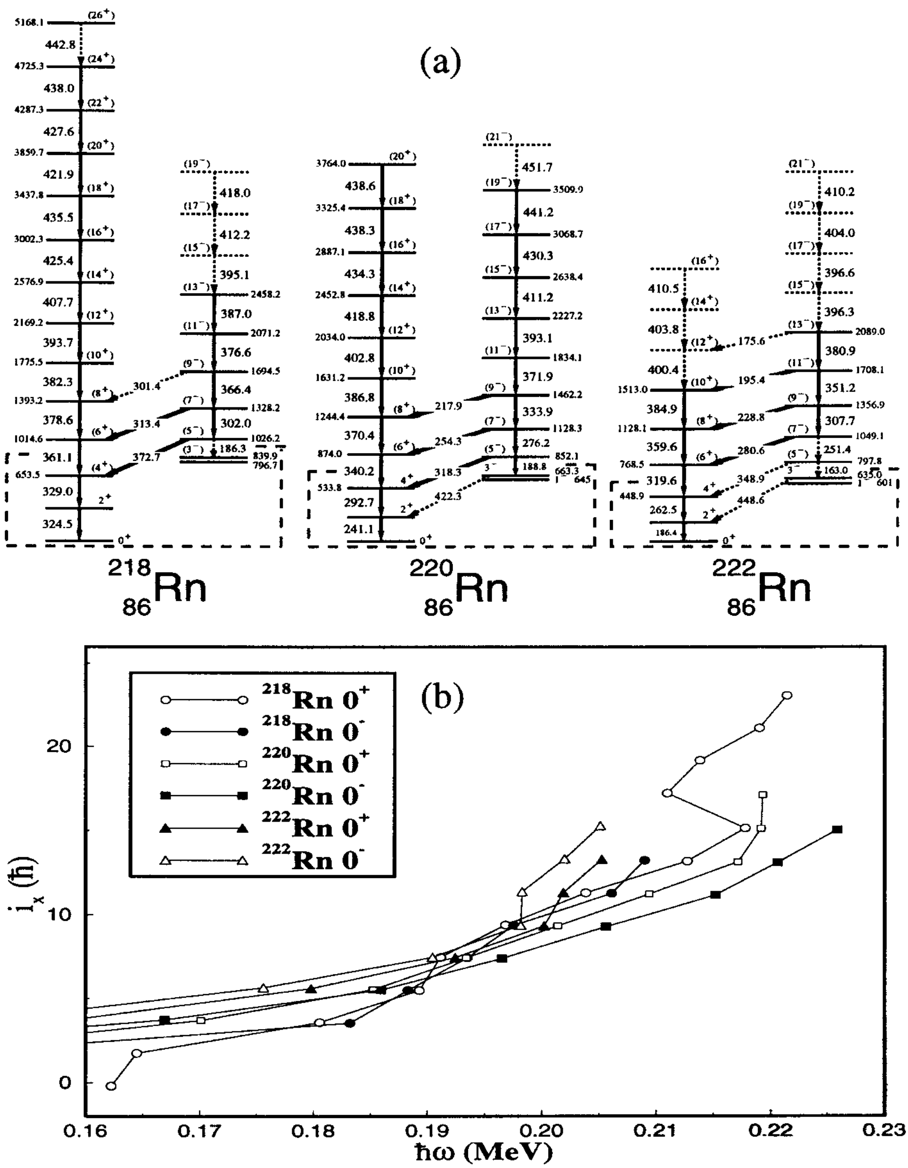

Fig. 4. (a) The level schemes of ^{218}Rn, ^{220}Rn and ^{222}Rn. The transition energies have errors which range from 0.2 keV for transitions between low-lying states in the ground state band to 0.5 keV for 5^- to 3^- and 7^- to 5^- transitions and transitions between the highest spin states observed. (b) Plot of aligned angular momentum i_x as a function of rotational frequency $\hbar\omega$ for ^{218}Rn, ^{220}Rn and ^{222}Rn.

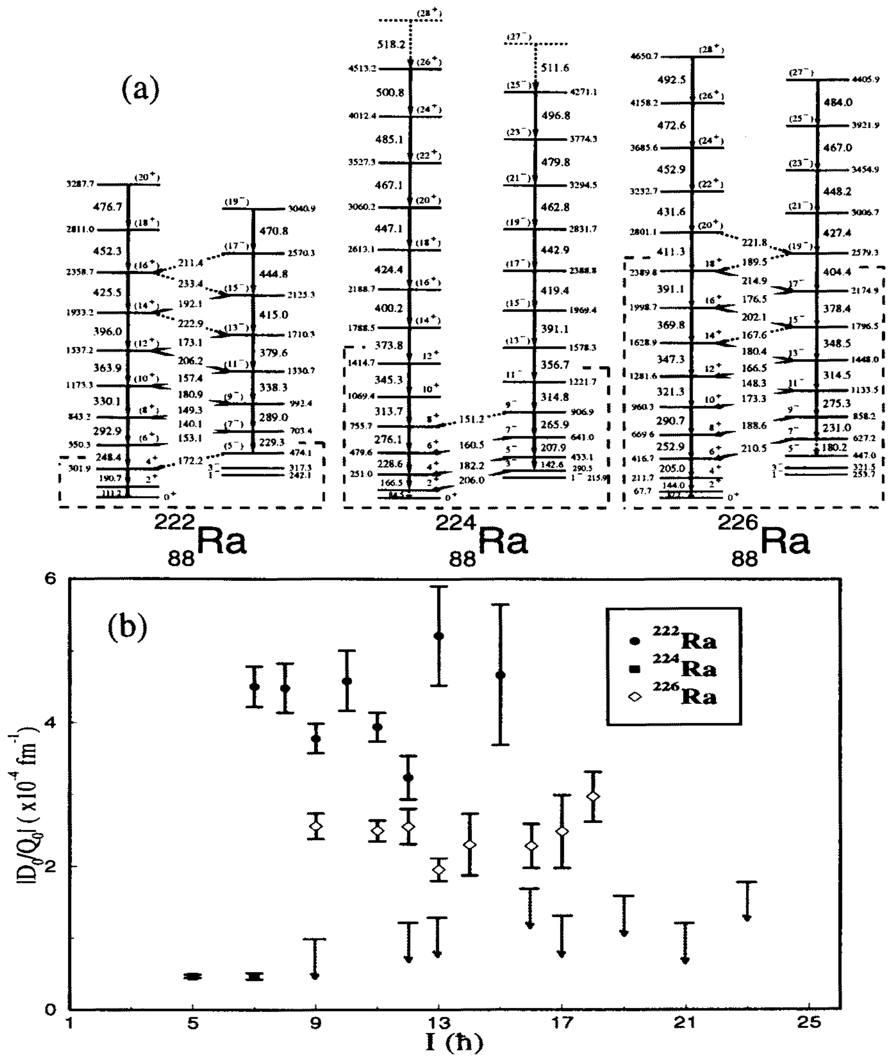

Fig. 5. (a) The level schemes of ^{222}Ra, ^{224}Ra and ^{226}Ra. The transition energies have errors which range from 0.2 keV for transitions between low-lying states in the ground state band to 0.5 keV for 5^- to 3^- and 7^- to 5^- transitions and transitions between the highest spin states observed. (b) Plot of intrinsic electric dipole to quadrupole ratio (D_0/Q_0) as a function of spin for states in ^{222}Ra, ^{224}Ra and ^{226}Ra. Values of D_0/Q_0 were derived from $B(E1)/B(E2)$ values. Upper limits for high-spin states in ^{224}Ra were obtained using two standard deviations in the background count as the intensity of the E1 transitions.

The level schemes of ^{222}Ra, ^{224}Ra and ^{226}Ra are shown in figure 5(a). Previous knowledge of states in these nuclei is also highlighted using dashed-line boxes. The level schemes of ^{222}Ra, ^{224}Ra and ^{226}Ra have been considerably extended in this work and interleaving positive- and negative-parity states have been observed for the first time in ^{222}Ra.

In figure 5(a) one can see that interband E1 transitions depopulate states up to $I^\pi = 17^-$ in ^{222}Ra and $I^\pi = 20^+$ in ^{226}Ra but although the yrast band in ^{224}Ra has been observed up to $I^\pi = 28^+$, no interband E1 transitions have been observed above $I^\pi = 9^-$ in this nucleus. For each state that is depopulated by both E1 and E2 transitions in the three Ra isotopes, intrinsic electric dipole to quadrupole (D_0/Q_0) ratios were extracted from $B(E1)/B(E2)$ values. These values are plotted on figure 5(b). The D_0/Q_0 values measured for the excited states in ^{224}Ra are much lower than those in ^{222}Ra and ^{226}Ra. At low spin, calculations by Butler and Nazarewicz [14] reproduced an anomalously low D_0/Q_0 for ^{224}Ra by treating the intrinsic electric dipole moment, D_0, as the sum of a macroscopic (liquid drop) component and a microscopic (shell-correction) term, which cancel for ^{224}Ra. In figure 5(b) the upper limits of D_0/Q_0 for high-spin states in ^{224}Ra indicate that the cancellation persists to high angular momenta in this nucleus. These upper limits were obtained using two standard deviations in the background count as the intensity of the E1 transitions. For ^{222}Ra and ^{226}Ra the D_0/Q_0 values are constant over the full spin ranges, which suggests that the reflection- asymmetric charge distribution in all three isotopes does not change with increasing spin. Values of D_0/Q_0 were extracted for excited states in the three Rn isotopes in a similar way. Using weighted mean values of D_0/Q_0 and published values of Q_0 [15], or using Grodzins' principle [16], D_0 values were determined for the Rn and Ra isotopes. Table I compares D_0 values measured in this work with those measured in previous work and theoretical values obtained from reference [14]. Also shown is the D_0 value measured for ^{220}Ra [17, 18]. The values extracted for the Rn isotopes appear to be in good agreement with the theoretical values. The theoretically predicted minimum in D_0 as one moves from ^{220}Ra to ^{224}Ra is observed experimentally. However, the values of D_0 measured in this work and in reference [18] for ^{220,222}Ra and ^{226}Ra are significantly larger than the theoretical values.

TABLE I

A comparison of experimental intrinsic electric dipole moments measured in this work with previous work and theoretical values taken from reference [1]. Experimental values are derived from weighted mean values of D_0/Q_0 and Q_0 values from reference [15] except for ^{218}Rn where Q_0 was determined using Grodzins' principle [16].

Nucleus	Experiment				Theory	
	This Work		Previous Work			
	I range	D_0 (e.fm)	I range	D_0 (e.fm)	I range	D_0 (e.fm)
^{218}Rn	7	0.035(3)	-	-	0	0.04
^{220}Rn	5-9	0.04(1)	-	-	0	−0.04
^{222}Rn	9-11	0.10(2)	-	-	0	−0.10
^{220}Ra	-	-	7-17	0.27(7)	0	0.17
					4	0.18
^{222}Ra	7-15	0.27(4)	3	0.38(6)	0	0.09
					5-7	0.11
^{224}Ra	5-7	0.030(1)	3-5	0.028(4)	0	0.01
	12-23	<0.09	7-9	<0.11	6	0.01
			1-5	0.06-0.10	0	−0.09
^{226}Ra	9-18	0.18(2)	7-12	0.12-0.21	7-12	−0.09
			7-11	0.16(1)		

Figure 4(b) shows the experimental alignment as a function of rotational frequency in ^{218}Rn, ^{220}Rn and ^{222}Rn. Pronounced alignment effects are observed for the positive parity states in ^{218}Rn and ^{220}Rn at $\hbar\omega \approx 0.22$ MeV. Cranked shell model calculations performed with the Woods-Saxon deformed shell model potential [19] with "universal" parameterisation [20] and deformation parameters of ^{218}Rn and ^{220}Rn taken from reference[14] predict alignments of pairs of $i_{13/2}$ protons at a rotational frequency of $\hbar\omega \approx$ 0.25 MeV. Alignment effects have previously been observed in octupole-deformed nuclei in the lanthanide region. Crossings of the octupole bands with aligned reflection-symmetric two-quasiparticle bands have been observed in ^{146}Ba [21, 22] and ^{150}Sm [23]. In contrast the Ra isotopes (see figure 5a) and Th isotopes maintain regular structures to the highest spins observed. One explanation for this behaviour is that strong alignment effects are expected at $Z = 86$ because the mixing of the $i_{13/2}$ proton orbital with lower spin orbitals, responsible for washing out the alignment effects in Ra and Th isotopes, is weaker for this proton number and quadrupole deformation. An alternative explanation is that octupole correlations for both protons and neutrons are weaker for the Rn isotopes.

5. Summary

Preliminary measurements using the recoil tagging technique indicate that spectroscopic measurements of octupole U nuclei with $N \approx 132$ are possible despite the low cross-sections. We have also demonstrated that multi-nucleon transfer reactions can populate octupole Rn and Ra isotopes, and interleaving bands of alternating parity states have been observed to high spin in ^{224}Ra, ^{226}Ra and, for the first time, in ^{222}Ra, ^{218}Rn, ^{220}Rn and ^{222}Rn. In ^{222}Ra and ^{226}Ra the values of D_0/Q_0 were found to be approximately independent of spin, while for ^{224}Ra, the measurements indicate that the cancellation of contributions to the intrinsic electric dipole moment persists to high spins. Plots of experimental alignment as a function of rotational frequency revealed strong alignment effects in ^{218}Rn and ^{220}Rn.

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