

# Coulomb Excitation of $^{222}\text{Rn}$ .

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The nature of quadrupole and octupole collectivity in  $^{222}\text{Rn}$  was investigated by determining the electric quadrupole (E2) and octupole (E3) matrix elements using sub-barrier, multi-step Coulomb excitation. The radioactive  $^{222}\text{Rn}$  beam, accelerated to 4.23 MeV/u, was provided by the HIE-ISOLDE facility at CERN. Data were collected in the Miniball  $\gamma$  ray spectrometer following the bombardment of two targets,  $^{120}\text{Sn}$  and  $^{60}\text{Ni}$ . Transition E2 matrix elements within the ground-state and octupole bands were measured up to  $10\hbar$  and the results were consistent with a constant intrinsic electric-quadrupole moment,  $518(11)\text{efm}^2$ . The values of the intrinsic electric-octupole moment for the  $0^+ \rightarrow 3^-$  and  $2^+ \rightarrow 5^-$  transitions were found to be respectively  $2360^{+300}_{-210}\text{efm}^3$  and  $2300^{+300}_{-500}\text{efm}^3$  while a smaller value,  $1200^{+500}_{-900}\text{efm}^3$ , was found for the  $2^+ \rightarrow 1^-$  transition. In addition, four excited non-yrast states were identified in this work via  $\gamma$ - $\gamma$  coincidences.

## I. INTRODUCTION

It is well established that nuclei outside of closed shells exhibit collective properties that can be best described in terms of nuclear shapes. Valence nucleons interact via long-range correlations which distort the spherical shape causing the nucleus to become deformed. The prevalent distortion observed across the nuclear chart is quadrupole deformation, which can be either prolate or oblate, where the nucleus retains both axial and reflection symmetry. The low-lying excitations of an even-even nucleus with a quadrupole-deformed ground state exhibit a characteristic rotational band of positive-parity states that are connected by collective E2 transitions.

There is evidence from theoretical and experimental studies that some nuclei undergo the breaking of reflection symmetry in the intrinsic frame [1]. Such reflection-asymmetric or “pear” shapes arise from the long-range octupole-octupole interactions that are strongest when orbitals with total- and orbital-angular momentum with a difference of  $3\hbar$  and opposite parity lie near the Fermi surface for both protons and neutrons, which occurs for  $Z, N \approx 34, 56, 88$  and  $N \approx 134$  [1]. Octupole collectivity is manifested by the presence of low-lying negative-parity states in even-even nuclei that are connected to the members of the ground-state band via enhanced E1 and E3 transitions, with the latter having typical values of tens of Weisskopf units. Here, the nuclear shape is interpreted as being subjected to pear-shaped distortions that can be stable where the

nucleus assumes a permanent pear-shape or is dynamic and the nucleus is subject to octupole vibrations.

There is experimental evidence that the  $^{222,224,226}\text{Ra}$  isotopes have a static octupole deformation [2]. This is based on two observations. The first is the behaviour of  $\Delta i_x$ , the difference in aligned angular momentum between negative- and positive-parity states at the same rotational frequency  $\omega$ , that tends to zero for increasing values of  $\hbar\omega$ . The second observation is that the electric octupole moments connecting the  $0^+$  ground state with the low-lying  $3^-$  level [3–5] are enhanced compared to those observed for  $^{220}\text{Rn}$  [4],  $^{230,232}\text{Th}$  [6, 7], and  $^{234}\text{U}$  [6]. The pattern of  $E3$  matrix elements between different states in  $^{228}\text{Ra}$  was observed to have deviations from the rotational model in contrast to  $^{222}\text{Ra}$  and  $^{226}\text{Ra}$  [3], and exhibits behaviour similar to that of  $^{148}\text{Nd}$  [8], interpreted as having an octupole vibrational character.

Investigations into octupole collectivity are of interest in the search for permanent atomic electric dipole moments (EDMs). In an octupole-deformed odd-mass nucleus, the nuclear Schiff moment (the electric-dipole distribution weighted by radius squared), which induces the atomic EDM, is expected to be enhanced by a factor of 100-1000 over that for a non-octupole deformed system (see [9] and references therein). This enhancement arises because of the large octupole moment and the presence of an excited state nearly degenerate with the ground state that forms a parity doublet. At present, experimental limits on EDMs, e.g. [10], have placed severe constraints on many extensions of the Standard Model. It is hoped that these limits can be further reduced in new EDM searches that exploit octupole-deformed nuclei such as  $^{225}\text{Ra}$  [11].

While there are ample spectroscopic data available on odd-mass Ra isotopes, there are comparatively few data for radon isotopes, also proposed as candidates for EDM searches [12]. The first study of excited states in  $^{224,226}\text{Rn}$  [13, 14] carried out at ISOLDE, CERN showed that the variation of  $\Delta i_x$  with  $\hbar\omega$  has the behaviour of a nucleus undergoing octupole vibrations, i.e.  $\Delta i_x \sim 3\hbar$ . This behaviour, suggesting that Rn is less suitable than Ra for EDM searches, has previously been observed for lighter Rn isotopes [15]. (There will be, however, some enhancement in the Schiff moment even if there is no evidence for rigid octupole deformation [16–18].) In the same ISOLDE experiment that measured the energy levels in  $^{224,226}\text{Rn}$ , the intensities of transitions in  $^{222}\text{Rn}$  were measured following Coulomb excitation, with the aim of measuring the octupole strength in this nucleus.

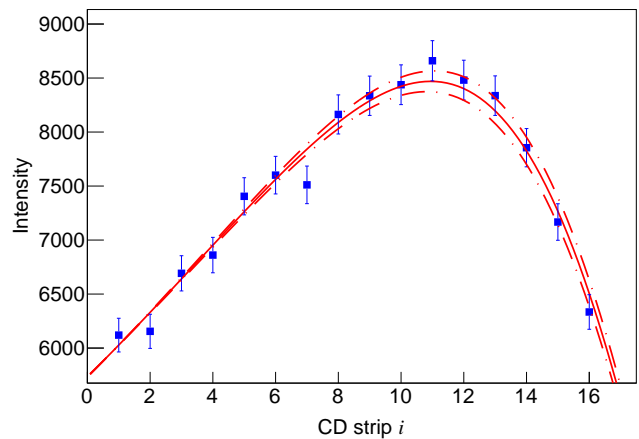


FIG. 1. Alpha-particle intensity detected in each front strip of one CD quadrant. The ordering of the strips presented start from the outermost strip to the innermost with increasing strip number. The solid red line corresponds to the best fit for  $x$ , the distance between the  $\alpha$ -particle source and the centre of the detector, using equation 1. The dashed red lines correspond to a one sigma uncertainty in  $x$ .

## II. EXPERIMENT

The radioactive  $^{222}\text{Rn}$  ions were produced by spallation by bombarding a thick thorium carbide primary target with 1.4 GeV protons delivered by the CERN PS Booster at a rate of  $\sim 10^{13}$  protons  $\text{s}^{-1}$ . The reaction products diffused and effused from the heated target via a cooled transfer line towards an enhanced plasma ion-source [19], which was used to singly ionize ( $q = 1^+$ ) the Rn isotopes. The ions were then extracted by applying a voltage of 30 kV, separated according to  $A/q$  using the ISOLDE High Resolution Separator, and delivered to a Penning trap, REX-TRAP [20, 21], at a rate of approximately  $8 \times 10^6$  ions  $\text{s}^{-1}$ . The ions were accumulated and cooled in the trap, and delivered as a bunch to an electron-beam ion source, REX-EBIS [20, 21] at 500 ms intervals. Here, the charge-state of the ions was increased by charge breeding up to  $51^+$ . The ions were subsequently extracted as 1 ms pulses, then separated by their mass-over-charge ratio for a second time and injected into the HIE-ISOLDE linear post-accelerator [22] at a rate of 2 Hz, where the ions were accelerated to 4.23 MeV/u. The accelerated ions then bombarded, with an intensity of  $6 \times 10^5 \text{ s}^{-1}$ , a  $2.1 \text{ mg}\cdot\text{cm}^{-2}$  thick  $^{120}\text{Sn}$  target for 7 hours and a  $2.1 \text{ mg}\cdot\text{cm}^{-2}$   $^{60}\text{Ni}$  target for 11 hours. The energy of the projectile fulfils Cline’s safe distance criterion for head-on collisions [23] in the case of the  $^{120}\text{Sn}$  target, and in most ( $>95\%$ ) of the  $^{60}\text{Ni}$  target after energy loss in the target. This ensures that the observed interactions between the projectile and target nuclei are purely electromagnetic. In the case of the  $^{60}\text{Ni}$  target, the criterion is satisfied at the maximum energy of the projectile for the scattering angles observed in this

experiment.

The Sn and Ni targets were placed at the centre of the Miniball spectrometer [24], an array of eight triple-cluster HPGe detectors each with six-fold segmentation. The scattered beam and recoiling target nuclei were detected by the quadrants of a “CD” detector [25], which comprises four double-sided silicon strip detectors with 16 annular strips on the front face and 24 radial sectors, coupled in pairs, on the back face. The CD detector was located 28.1(3) mm from the target and covered the range of laboratory angles 17.9°–55.5°. The detector distance was determined by measuring the intensity of  $\alpha$  particles emitted from a  $^{226}\text{Ra}$  source placed at the target position. The intensity in each front strip of a quadrant, shown in Figure 1, is related to the solid angle of the corresponding strip, which is dependent on the distance of the CD detector to the source. The  $\alpha$ -particle intensity in a given strip,  $i$ , is given by the relation

$$I_{\alpha,i} = A \times x \left( \frac{1}{\sqrt{x^2 + r_{i1}^2}} - \frac{1}{\sqrt{x^2 + r_{i2}^2}} \right), \quad (1)$$

where,  $I_{\alpha,i}$  is the number of  $\alpha$  particles observed in strip  $i$ ,  $A$  is a constant,  $x$  is the distance between the source and the centre of the detector, and  $r_{i1,i2}$  is the inner- and outer-radius of strip  $i$  respectively. It is noted that the datum point for CD strip 7 lies two standard deviations from the fitted line, but this discrepancy is believed to be statistically acceptable. In any case variations in strip efficiency are subsumed into the normalisation variables (see later). The highly segmented CD detector combined with the sixfold segmentation of the HPGe detectors permits an event by event Doppler correction to be performed to a high precision. This results in superior energy resolution of the  $\gamma$  ray spectra than would be obtained without such segmentation.

The scattering of  $^{222}\text{Rn}$  projectiles bombarding a  $^{120}\text{Sn}$  target is presented in Figure 2. For the analysis,  $\gamma$  rays were selected when observed in coincidence with a recoiling target nucleus. The Doppler shift correction was made assuming that the  $\gamma$  rays are emitted by the projectile. The time difference between particle events recorded in the CD detector and  $\gamma$  rays recorded by the Miniball detectors was used to distinguish  $\gamma$ -ray events corresponding to prompt decays following Coulomb Excitation from background events. The particle- $\gamma$ -ray window for prompt coincidences was 450 ns wide and the random coincidence window was 600 ns wide, as presented in Figure 3.

### III. RESULTS

There are several ways in which data from the the Miniball spectrometer can be sorted to produce  $\gamma$ -ray spectra. The “Core” mode records the energies

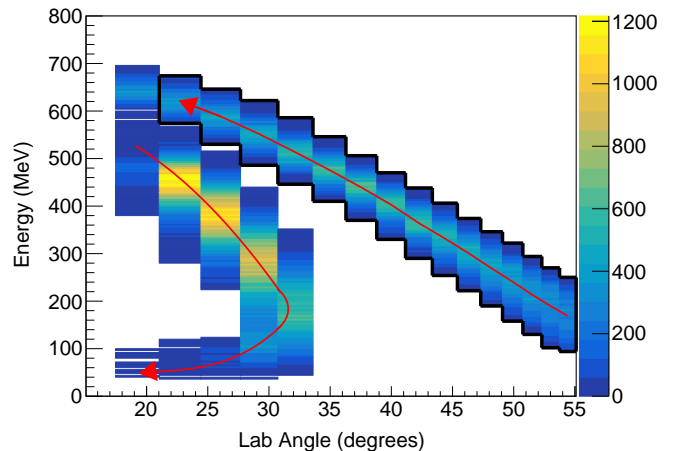


FIG. 2. Particle energy versus scattering angle for the lower-energy  $^{222}\text{Rn}$  projectiles and higher-energy  $^{120}\text{Sn}$  recoils observed in the CD detector. The direction of the arrows correspond to an increasing scattering angle in the Centre-of-Mass frame. The particle gate for recoiling target nuclei is shown as a solid black line.

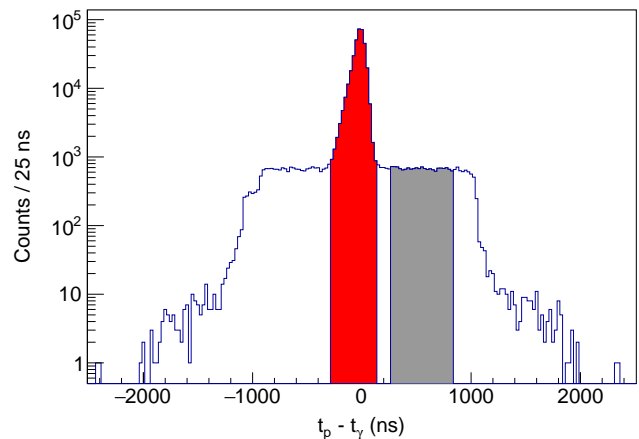


FIG. 3. Time difference between particle- $\gamma$  coincident events. The prompt coincidence window, highlighted in red, was 450 ns wide and the time-random coincidence window, highlighted in grey, was 600 ns wide.

of all events from the core (anode) signals of each crystal. A variation of this is to sum the energies of  $\gamma$  rays recorded in any of the three crystals in a single triple-cluster if more than one  $\gamma$ -ray hit was registered within a short time window. This is the “Addback” procedure which maximises the photopeak efficiency of the spectrometer. These two configurations are employed for most experiments using Miniball. Another variation, the “Reject” method, outlined in [13], rejects events where hits are recorded in the adjacent crystals within the triple-cluster. This reduces the background from Compton scattering and is appropriate for  $\gamma$ - $\gamma$  measurements; the “Addback” procedure would enhance the summing of coincidence  $\gamma$  rays and potentially give erroneous transition assignments. In another variation

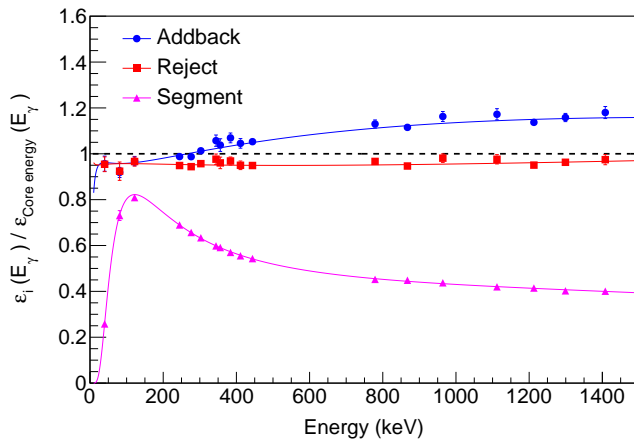


FIG. 4. Ratio of detector efficiency between the “Addback”, “Reject” and “Segment” modes with respect to the “Core” mode. The data were collected using  $^{152}\text{Eu}$  and  $^{133}\text{Ba}$  calibration sources.

the recorded energies were taken from each of the six individual segments (cathodes); events were rejected if a second hit was recorded in another segment in the crystal. This mode is called “Segment” and was used to reduce the instantaneous count rate in each detector in order to improve the quality of the spectra. This method was used in Ref. [3]. The relative  $\gamma$ -ray efficiencies as a function of energy for the different modes are compared in Figure 4. Energy and efficiency calibrations were performed using standard  $^{133}\text{Ba}$  and  $^{152}\text{Eu}$  sources, which emit  $\gamma$  rays covering the full energy range of transitions observed in this work. For the analysis of this work both “Segment” and “Core” modes were used. The advantage of the former is the improved energy resolution while the latter mode has higher detector efficiency, particularly at energies above 400 keV.

The  $\gamma$ -ray spectra obtained from both the  $^{120}\text{Sn}$  and  $^{60}\text{Ni}$  targets are presented in Figure 5. As the cross section for the Coulomb excitation of the projectile is strongly dependent on the atomic number of the target, the use of two targets with significantly different values of  $Z$ ,  $^{120}\text{Sn}$  and  $^{60}\text{Ni}$ , produce a different population of states in  $^{222}\text{Rn}$ . In particular the higher  $Z$  target,  $^{120}\text{Sn}$ , allows access to higher-spin states through multi-step Coulomb excitation compared to  $^{60}\text{Ni}$ , see Figure 5. The spectra reveal a strong population of the positive-parity states of the ground-state band, which are populated via multiple E2 excitation. The population of negative-parity states of the octupole band, populated mostly by E3 excitation, is established via E1 decays to the positive-parity states present in the spectra.

Decays from two non-yrast bands were observed for the first time in this experiment and their placement in the level scheme was determined through analysis of a  $\gamma$ - $\gamma$  coincidence matrix collected with data from

both targets. The spectra of  $\gamma$  rays observed in coincidence with decays from non-yrast levels and the  $1_1^- \rightarrow 0_1^+$  transition are presented in Figure 6. The new bands are labelled as  $\beta$ - and  $\gamma$ -bands for convenience and do not imply any particular structure for these bands. It is assumed, however, that the  $\beta$ -band has  $K^\pi = 0^+$  and the  $\gamma$ -band has  $K^\pi = 2^+$ , as observed in many even-even nuclei in this mass region at similar excitation energies. The  $\gamma$ -band was observed to have its bandhead at 867 keV excitation energy that decays to both the ground state and the  $2_1^+$  first excited state. The bandhead of the  $\beta$ -band was observed to have approximately the same excitation energy of 867 keV. It decays via a 266(2) keV transition that is in coincidence with the  $E_\gamma(1_1^- \rightarrow 0_1^+) = 601$  keV transition. The 266 keV  $\gamma$  ray is assumed to originate from the  $\beta$ -bandhead as a change in  $\Delta K=0$  via an E1 transition is allowed in the rotational model, while  $\Delta K=2$  via an E1 transition is forbidden. The intensity of the  $0_\beta^+ \rightarrow 1_1^- \rightarrow 0_1^+$  coincidence was compared to the  $3_1^- \rightarrow 2_1^+ \rightarrow 0_1^+$  coincidence to obtain the intensity of the  $E_\gamma(0_\beta^+ \rightarrow 1_1^-)$  transition, which is otherwise hidden in the singles spectrum by the dominant  $4_1^+ \rightarrow 2_1^+$  transition. From this, the level population of the  $\beta$ -band and subsequent feeding to the negative-parity states was obtained.

The matrix elements presented in this work were obtained by using the Coulomb-excitation least-squares fitting code GOSIA [23, 26, 27]. GOSIA was employed to calculate excitation probabilities and subsequent  $\gamma$ -ray decay intensities of excited states for a given set of electromagnetic matrix elements. The calculated  $\gamma$ -ray intensities can be compared with the experimental yields and additional spectroscopic information that is available. In this work, known  $\gamma$ -ray branching ratios of low-lying negative-parity states taken from [15, 28] together with the measured  $\gamma$ -ray intensities were included in the calculations. A standard  $\chi^2$  function for both yields and branching ratios was constructed which was minimised by varying the values of the electromagnetic matrix elements between all relevant states in  $^{222}\text{Rn}$ , treated as free parameters. The level scheme included in the calculation is presented in Figure 7. In the ground-state positive-parity band and negative-parity band states up to  $I^\pi=14^+$  and  $I^\pi=15^-$  [15, 28], respectively, were included. Additional levels in the  $\beta$ - and  $\gamma$ -bands were included whose excitation energies were assumed to follow a  $I(I+1)$  spacing with moment-of-inertia similar to those of excited  $K^\pi = 0^+$  and  $K^\pi = 2^+$  bands observed elsewhere in this mass region. These are shown as dashed lines in Figure 7.

The  $\gamma$ -ray yields were measured for three ranges of scattering angle for both targets. The angular ranges employed for the analysis are presented in Table I. Two independent analyses of the  $\gamma$ -ray yields were performed using either the core signals or segment signals in, respectively, “Core” mode or “Segment” mode (see

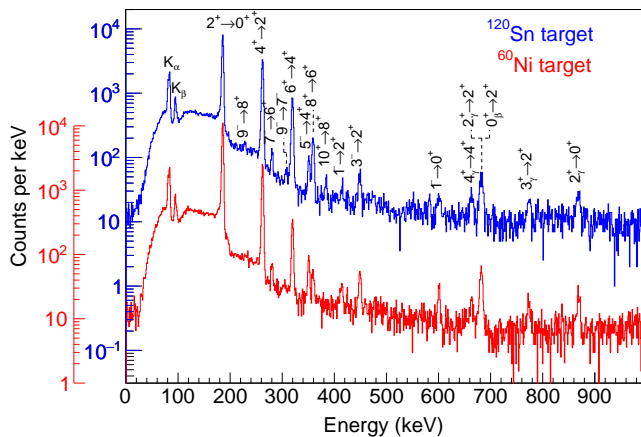


FIG. 5. Spectra of  $\gamma$  rays emitted following Coulomb excitation of  $^{222}\text{Rn}$  using a  $^{120}\text{Sn}$  target (blue) and a  $^{60}\text{Ni}$  target (red) observed in coincidence with recoiling target nuclei. The  $\gamma$ -ray energies were corrected for Doppler shift assuming they are emitted from the scattered projectile. Time-random coincidences between Miniball and CD detector have been subtracted. The spectra were obtained by sorting the data in the “Segment” mode.

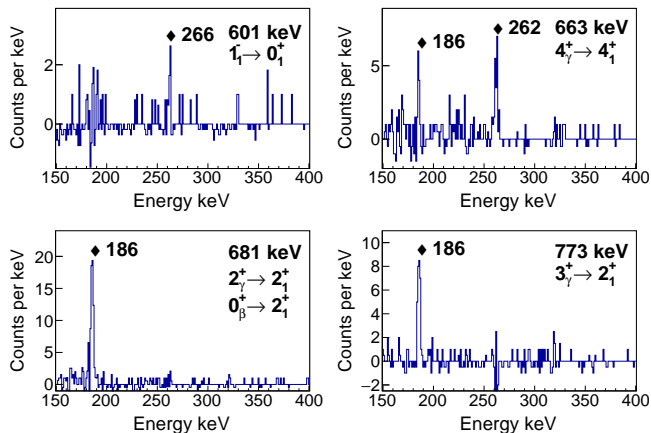


FIG. 6. Background-subtracted  $\gamma$ -ray spectra observed in time-coincidence with other  $\gamma$ -ray transitions. The energy gate is labelled corresponding to the energies, spin and parity of the initial and final states of the relevant transition. The transitions observed in coincidence with the gate are labelled with their energy given in keV. The spectra were obtained by sorting the data in the “Segment” mode.

earlier). The yields were corrected for the loss of events removed in the sorting procedure when two or more coincident  $\gamma$  rays are detected simultaneously in the same crystal, that would reduce slightly the intensity of high-spin transitions compared to those at low spin. The yield of the  $E_\gamma(3_1^- \rightarrow 2_1^+) = 449$  keV transition was corrected for the contribution from the pileup of the  $E_\gamma(2_1^+ \rightarrow 0_1^+) = 186$  keV and  $E_\gamma(4_1^+ \rightarrow 2_1^+) = 262$  keV transitions. It was also ensured that the measured intensity of the  $2^+ \rightarrow 0^+$  transition included the small

component emitted after stopping in the CD detector that arises from the long lifetime of the  $2^+$  state. For the  $^{120}\text{Sn}$  target, the intensity of the stopped component of the  $2^+ \rightarrow 0^+$  transition was estimated to be 4.3(3)% of the total intensity, while for the  $^{60}\text{Ni}$  target, this value was 1.9(2)%. In the case of the aforementioned  $E_\gamma(0_\beta^+ \rightarrow 1_1^-) = 266$  keV transition, a single  $\gamma$ -ray yield corresponding to the total observed scattering range for each target was measured.

TABLE I. The ranges of the angles of the recoiling target in the laboratory frame ( $\zeta$ )

used to obtain  $\gamma$ -ray yields for both targets and each set of analysis.

Analysis	Target	$\zeta_1$	$\zeta_2$	$\zeta_3$
Segment	$^{120}\text{Sn}$	21.5°-34.0°	34.1°-43.8°	43.9°-55.5°
	$^{60}\text{Ni}$	17.9°-31.1°	31.2°-41.6°	41.7°-55.5°
Core	$^{120}\text{Sn}$	21.5°-36.7°	36.8°-47.8°	47.9°-55.5°
	$^{60}\text{Ni}$	17.9°-36.7°	36.8°-47.8°	47.9°-55.5°

In total, 89 data points were fitted with 21  $E\lambda$  ( $\lambda=1,2,3$ ) matrix elements and 8 normalisation variables in the calculation, while all other  $E\lambda$  ( $\lambda=1,2,3$ ) matrix elements were coupled to the freely-varying matrix elements, as discussed later in the text. Most  $\gamma$ -ray intensities were measured for the three angular ranges and two targets (6 normalisation variables) while the intensity of the 266 keV transition was measured over the whole angular range. The initial value of each freely-varying matrix element was drawn randomly using a Gaussian probability distribution, where the mean value was calculated using the rotational model with constant intrinsic moments,  $Q_\lambda$ , derived from theoretical deformation parameters,  $\beta_\lambda$  [30]. The standard deviations,  $\sigma_\lambda$ , of the probability distributions were 0.001, 0.75, and 0.25  $\text{eb}^{\lambda/2}$  for the E1, E2 and E3 matrix elements respectively. The matrix elements were allowed to freely vary within the range  $\pm 0.05$ ,  $\pm 10$  and  $\pm 5$   $\text{eb}^{\lambda/2}$  for E1, E2 and E3 respectively. The intrinsic electric-multipole moment can be related to the corresponding matrix elements:

$$\langle I_i || M(E\lambda) || I_f \rangle = (2I_i + 1)^{1/2} (I_i K_i \lambda \Delta K | I_f K_f) Q_\lambda a_\lambda, \quad (2)$$

where  $I_{i,f}$ ,  $K_{i,f}$  describe respectively the initial and final quantum states connected by the operator  $M(E\lambda)$ ,  $(I_i K_i \lambda \Delta K | I_f K_f)$  is the Clebsch-Gordan coefficient and  $a_\lambda$  is a constant for a given multipole order.

As there is a large uncertainty associated with the measured  $\gamma$ -ray branching ratios of the negative-parity states, all E1 matrix elements connecting the negative-parity band and the ground-state positive-parity band were coupled to  $\langle 1^- || E1 || 0^+ \rangle$ , assuming the validity of equation 2 and that  $Q_1$  is independent of spin. The analysis took into account the feeding from levels in the  $\beta$ -band to the octupole band, whereby the  $\langle 1^- || E1 || 0_\beta^+ \rangle$

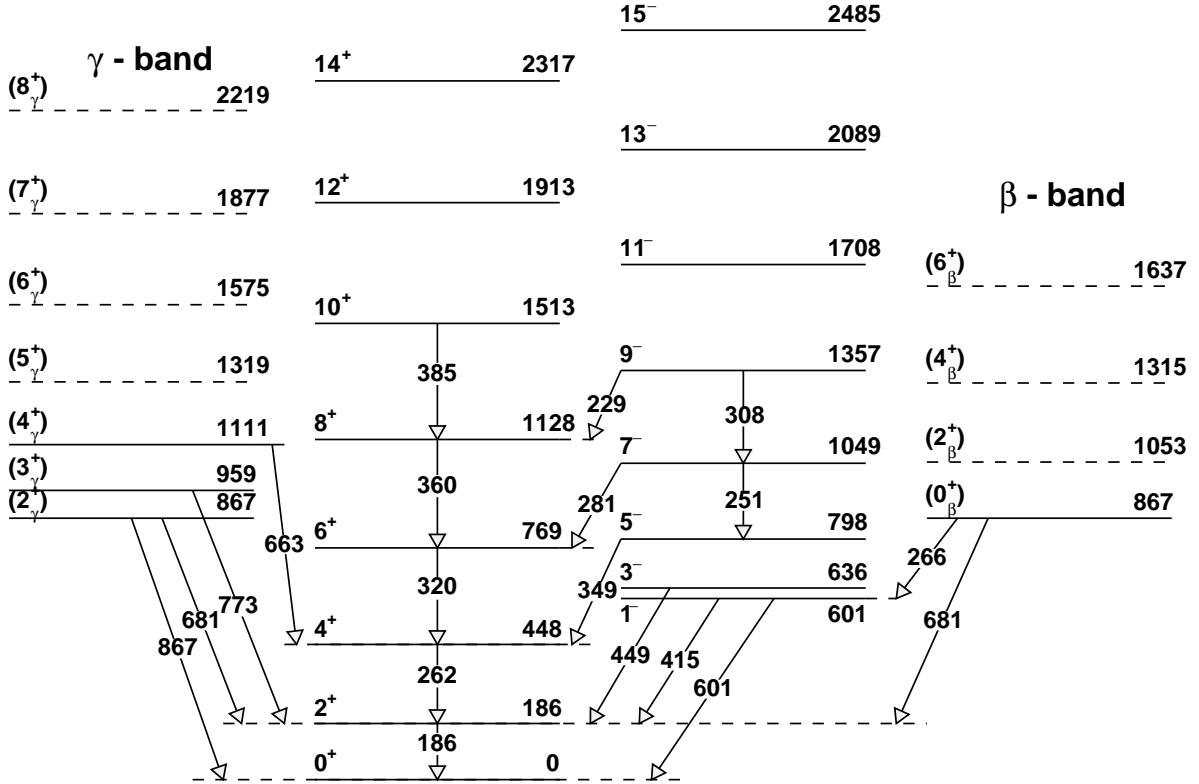


FIG. 7. Level scheme of  $^{222}\text{Rn}$  showing all levels used in the Gosia calculations. The levels in the positive-parity ground-state and negative-parity octupole bands were previously identified in references [28, 29]. The non-yrast levels attributed to the  $\gamma$ - and  $\beta$  bands were identified in this work from analysis of  $\gamma$ - $\gamma$  coincidences, and are drawn with solid lines. The levels drawn with dashed lines were also included in the Gosia fit, see the text for details.

matrix element is freely varied and all other inter-band transitions are coupled to it, again assuming the validity of the rotational model. This feeding intensity is constrained by the measured intensity of the  $0_{\beta}^{+} \rightarrow 1^{-}$  transition. This method of analysis is similar to that carried for  $^{222,228}\text{Ra}$  [3] earlier, for which either the inter-band transitions were observed ( $^{228}\text{Ra}$ ) or, as in the case of  $^{222}\text{Ra}$ , the connecting matrix elements were assumed to have a similar value of  $Q_1$  as that of the ground-octupole transitions. The relationship between the projectile bombardment energy and relative feeding from the  $\beta$ -band to the low-lying negative parity states was investigated and is presented in Figure 8. Here, the calculated intensities of transitions from the  $\beta$ -band to the  $1^{-}$  and  $3^{-}$  levels relative to the total decay intensity of the respective  $1^{-}$  and  $3^{-}$  levels are shown. As can be seen the feeding of the negative-parity states in the experiments described here, carried out at bombarding energies near the “safe” Coulomb-excitation limit, is appreciable, and the uncertainty in this feeding will influence the statistical uncertainty in the extracted value of  $\langle 0_{\beta}^{+} || E3 || 3_{\beta}^{-} \rangle$  and other  $E3$  matrix elements.

The  $E2$  matrix elements connecting states within the ground-state and negative-parity bands, for which  $\gamma$ -ray decays from the higher-lying levels were observed, were treated as free parameters with the exception of the  $2_{\beta}^{+} \rightarrow 0_{\beta}^{+}$  transition. Here,  $\langle 2_{\beta}^{+} || E2 || 0_{\beta}^{+} \rangle$  was coupled to  $\langle 4_{\beta}^{+} || E2 || 2_{\beta}^{+} \rangle$  assuming the validity of the rotational model, and the  $\langle I^{\pi} || E2 || I^{\pi} \rangle$  matrix elements were coupled to the corresponding elements  $\langle I^{\pi} || E2 || (I-2)^{\pi} \rangle$  with the exception of  $\langle 2_{\beta}^{+} || E2 || 2_{\beta}^{+} \rangle$ , which was a free parameter. This was the same procedure as adopted in the analysis of  $^{222,228}\text{Ra}$  [3]. The intra-band  $E2$  matrix elements connecting states within the respective  $\beta$  and  $\gamma$  bands, for which no  $\gamma$ -ray transitions were observed, were coupled to  $\langle 4_{\beta}^{+} || E2 || 2_{\beta}^{+} \rangle$  and assume the same intrinsic electric-quadrupole moment. The inter-band  $E2$  matrix elements connecting these bands to the ground-state positive-parity band, where  $\gamma$ -ray transitions were observed, were allowed to freely vary, otherwise were coupled to the  $\langle 2_{\gamma}^{+} || E2 || 0_{\beta}^{+} \rangle$  or  $\langle 0_{\beta}^{+} || E2 || 2_{\beta}^{+} \rangle$ , respectively (assuming the validity of the rotational model). The  $M1$  matrix elements connecting states with the  $\gamma$ -band were calculated assuming  $g_K = -0.4$ ,  $g_R = 0.4$ , while the inter-band  $M1$  matrix elements were assumed to be

negligible.

A total of five E3 matrix elements were investigated in this work. Higher-lying E3 matrix elements were coupled to the free E3 matrix element corresponding to the same change in total angular momentum,  $\Delta I = I^- - I^+$ . The higher-lying E3 matrix elements  $\langle I_1^+ || E3 || (I_1 + 3)^- \rangle$  were coupled to  $\langle 5_1^- || E3 || 2_1^+ \rangle$  while  $\langle 3_1^- || E3 || 0_1^+ \rangle$  was left completely free. Fixed E4 matrix elements were included in the analysis and were calculated using a constant value for the hexadecapole moment, derived from theoretical values of  $\beta_\lambda$  [30], under the assumption of the rotational model. The E3 matrix elements connecting levels from the  $\beta$ - and  $\gamma$ -bands with the negative parity levels were fixed, using the assumption of equation 2 but with a lower intrinsic octupole moment. The systematic error arising from the influence of various assumptions for this intrinsic octupole moment was investigated (see later).

The Coulomb-excitation probability is sensitive to the relative phases of the  $E2$  and  $E4$  matrix elements, and to the relative phases of the  $E1$  and  $E3$  matrix elements. The starting values of the signs of the  $E\lambda$  matrix elements were taken from the rotational model (equation 2) with the assumption that the signs of  $Q_2$ ,  $Q_3$  and  $Q_4$  are positive. The sign of  $Q_1$  can have the same sign as that of  $Q_3$  or have the opposite sign, and both possibilities were investigated. From the analysis, the fit for  $^{222}\text{Rn}$  favoured  $Q_1$  and  $Q_3$  having the opposite sign although the difference in total  $\chi^2$  between both assumptions was small. A negative phase difference was therefore adopted, which is in accord with calculations using the macroscopic-microscopic model [31]. The systematic errors in the matrix elements considered both solutions, however. The calculated electric moments

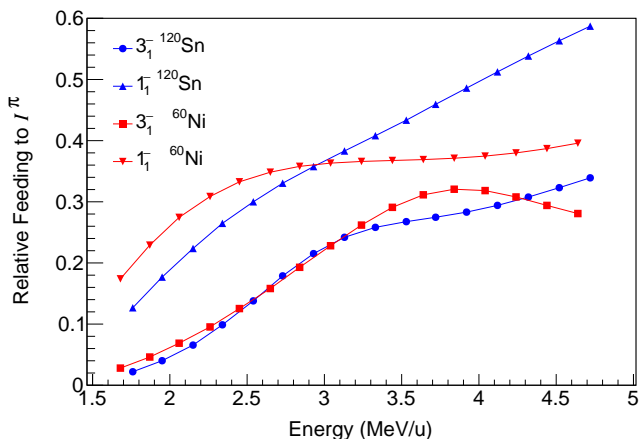


FIG. 8. Calculated intensity of feeding from levels attributed to the  $\beta$ -band to the state  $I^\pi$  relative to the total decay intensity of the state as a function of projectile energy for both the  $^{120}\text{Sn}$  (blue) and  $^{60}\text{Ni}$  (red) targets. In these experiments the range of projectile energies were 3.67 – 4.23 MeV/u for  $^{120}\text{Sn}$  and 3.49 – 4.23 MeV/u for  $^{60}\text{Ni}$ .

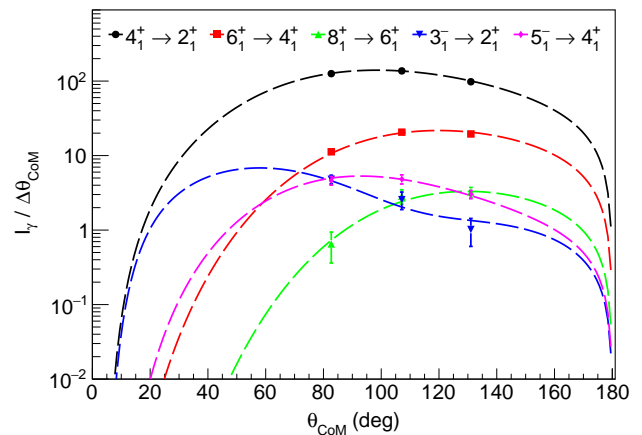


FIG. 9. Measured  $\gamma$ -ray intensities, divided by their respective scattering range in the Centre-of-Mass (CoM) frame, for selected transitions in  $^{222}\text{Rn}$  collected with the  $^{60}\text{Ni}$  target. The dashed lines correspond to calculated intensities as a function of scattering angle  $\theta$  in the CoM frame.

from the E1, E2, and E3 matrix elements obtained from their respective fits are presented in Table II.

For the GOSIA analysis, the statistical error for each variable was calculated taking into account correlations between all variables. To determine the systematic sources of errors, a number of independent fits was obtained with different initial conditions. These included varying the target thickness by  $\pm 5\%$ , the beam energy by  $\pm 1\%$ , the distance between the target and the particle detector by  $\pm 1\%$ . The sensitivity of the GOSIA fit to the efficiency of the HPGe detectors was also investigated by randomly varying the parameters describing the efficiency curve using a Gaussian probability distributions whose mean value and width are taken from the efficiency fit parameters. Fits were performed with the E4 matrix elements set to twice their initial values and set to zero, and changing the sign of the the E2 couplings to the higher-lying collective bands. In addition the magnitude of the E3 matrix elements connecting the negative parity states to states in the  $\beta$ -band was increased by a factor of 3.33 such that the corresponding  $Q_3$  value between these states was comparable to the  $Q_3$  value connecting the states between the ground-state and negative-parity bands. It should be noted that the two solutions given in Table II use data for which the full-energy  $\gamma$ -ray peak is detected in a segment or in the whole crystal. The former, therefore, contains data which are a subset of that for the latter, and each will have different peak shapes. Although the solutions are not statistically independent, it is expected that the GOSIA fitting procedure will arrive at slightly different minima. In general the values from the two solutions are in agreement, the only exceptions being the values of  $\langle 2_1^+ || E2 || 2_1^+ \rangle$  and  $\langle 2_1^+ || E2 || 2_\gamma^+ \rangle$ . The arithmetic mean value of the two

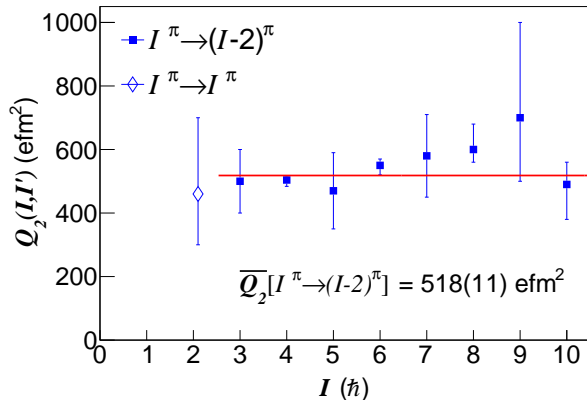


FIG. 10. Values of intrinsic quadrupole moments,  $Q_2$ , plotted as a function of spin (see Table II). These values correspond to transitions between states with spin  $I$  and  $I-2$  with the exception of the diagonal matrix elements of the  $2_1^+$  state. The value of the mean quadrupole moment, indicated by the red line, is obtained using the transitional matrix elements with the assumption that the ground-state and octupole bands have the same intrinsic moment.

solutions were used as the adopted values, while the adopted errors encompassed the range of possible values in each case as well as the systematic errors. For the  $\langle 2_1^+ || E3 || 3_1^- \rangle$  and  $\langle 4_1^+ || E3 || 1_1^- \rangle$  matrix elements only upper limits could be obtained.

Measured  $\gamma$ -ray yields of selected transitions as a function of Centre-of-Mass (CoM) angle are compared to calculated yields resulting from the GOSIA analysis in Figure 9. These were collected with the  $^{60}\text{Ni}$  target for the scattering ranges used for the Segment analysis in Table I. The distributions which peak at higher CoM angles correspond to transitions from states that are populated via multi-step excitation. Two components are observed in the population of the  $3^-$  state that arise from contributions from both direct excitation from the ground state and multi-step excitation such as those paths that proceed via the  $2_1^+$  state.

#### IV. DISCUSSION

The  $Q_2$  moments derived from the adopted values of E2 matrix elements connecting the levels within the ground-state and negative-parity bands are shown in Figure 10. The higher beam energies available at HIE-ISOLDE allow access to higher-lying states, in this case up to  $10 \hbar$ , compared to the  $6 \hbar$  that was achieved in the neighbouring  $^{220}\text{Rn}$  study [4] performed with the lower beam energy available with REX-ISOLDE. The value of  $Q_2$  is approximately constant as a function of spin, which is consistent with stable quadrupole deformation under rotation. The  $\overline{Q_2} = 518(11) \text{ efm}^2$

obtained from the fit of all transition E2 matrix elements within the ground-state and octupole bands is slightly larger than the value  $Q_2(2_1^+ \rightarrow 0_1^+) = 484(14) \text{ efm}^2$  calculated using the lifetime of the  $2_1^+$  level measured in Ref [32]. As expected the value of  $Q_2$  in  $^{222}\text{Rn}$  is larger than the value of  $Q_2 = 434(14) \text{ efm}^2$  measured in  $^{220}\text{Rn}$  [4]. The value of  $Q_2 = 480_{-160}^{+240} \text{ efm}^2$  corresponding to the  $\langle 2_1^+ || E2 || 2_1^+ \rangle$  matrix element is consistent with the values of  $Q_2$  corresponding to the transition E2 matrix elements although the large uncertainty in this value cannot rule out deviations as observed in the heavier Ra isotopes [3, 5], either arising from non-axial symmetry or from the effect of couplings to higher-lying collective bands.

In this region of the nuclear chart, the excitation energy of the  $2_\gamma^+$  level,  $E_x(2_\gamma^+)$ , has been identified in a number of Ra [33, 34] and Th [34, 35] isotopes as well as  $^{220}\text{Rn}$  with a value of around 1 MeV. The value of  $E_x(2_\gamma^+) = 867 \text{ keV}$  in  $^{222}\text{Rn}$  is lower in energy than the that of the corresponding state in  $^{220}\text{Rn}$ ,  $E_x(2_\gamma^+) = 937.9 \text{ keV}$  [33]. The similarity in excitation energy is reflected in the transition probabilities measured for the decay to the ground state: the value of  $B(E2; 2_\gamma^+ \rightarrow 0_1^+)$  for  $^{222}\text{Rn}$ ,  $1.4(5) \text{ W.u.}$ , is comparable to that for  $^{220}\text{Rn}$ ,  $2.6(11) \text{ W.u.}$  [4].

As discussed earlier, the behaviour of the aligned angular momentum of the negative-parity states relative to the positive-parity states provides a signature for the dynamics of the octupole instability. The quantity  $\Delta i_x = i_x^- - i_x^+$  is plotted as a function of rotational frequency  $\hbar\omega$  in Figure 11 for  $^{222}\text{Rn}$  and the neighbouring  $^{222-228}\text{Ra}$  isotopes. For  $^{222}\text{Rn}$  the values of  $\Delta i_x \approx 3\hbar$  are similar to those of  $^{228}\text{Ra}$  at high spin and are indicative of octupole-vibrational behaviour. This is

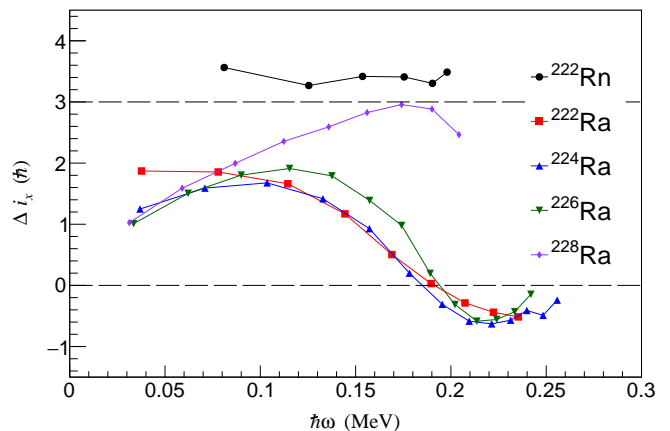


FIG. 11. The difference in aligned angular momentum,  $\Delta i_x = i_x^- - i_x^+$ , plotted as a function of rotational frequency  $\omega$ . The upper dashed line corresponds to the vibrational limit,  $\Delta i_x = 3 \hbar$ . For the sources of the data, see [2] and references therein.



TABLE II. Values of matrix elements measured in the present experiment from analysis of  $\gamma$ -ray spectra from the segment and core detector signals, assuming an opposite value for the relative phase of  $Q_1$  and  $Q_3$  moments. The values are presented with the  $1\sigma$  statistical error from the fit. The adopted values for the matrix elements are taken from the two sets of analysis and the uncertainty includes both the statistical uncertainty and the systematic contributions described in the text. The intrinsic moments,  $Q_\lambda$ , are calculated using Equation 2 using the adopted matrix elements.

$\langle I  E\lambda  I'\rangle$	Segment analysis	Core analysis	Sys Error	Adopted	
	m.e. eb $^{\lambda/2}$	m.e. eb $^{\lambda/2}$	m.e. eb $^{\lambda/2}$	m.e. eb $^{\lambda/2}$	$Q_\lambda$ efm $^\lambda$
$\langle 0_1^+  E1  1_1^- \rangle$	$\mp 0.007_{\pm 0.003}^{\pm 0.007}$	$\mp 0.006_{\pm 0.002}^{\pm 0.005}$	0.001	$\mp 0.007_{\pm 0.003}^{\pm 0.007}$	$\mp 0.14_{\pm 0.06}^{\pm 0.14}$
$\langle 1_1^-  E1  0_\beta^+ \rangle$	$\pm 0.0046_{\pm 0.0010}^{\pm 0.0010}$	$\pm 0.0066_{\pm 0.0011}^{\pm 0.0009}$	0.0003	$\pm 0.0057_{\pm 0.0020}^{\pm 0.0020}$	$\mp 0.12_{\pm 0.04}^{\pm 0.04}$
$\langle 2_1^+  E2  2_1^+ \rangle$	$-2.20_{-0.24}^{+0.25}$	$-1.31_{-0.11}^{+0.17}$	0.10	$-1.8_{-0.9}^{+0.6}$	$480_{-160}^{+240}$
$\langle 2_1^+  E2  4_1^+ \rangle$	$2.52_{-0.04}^{+0.06}$	$2.57_{-0.04}^{+0.04}$	0.02	$2.55_{-0.10}^{+0.06}$	$504_{-20}^{+13}$
$\langle 4_1^+  E2  6_1^+ \rangle$	$3.46_{-0.09}^{+0.08}$	$3.57_{-0.10}^{+0.08}$	0.04	$3.52_{-0.18}^{+0.13}$	$550_{-30}^{+20}$
$\langle 6_1^+  E2  8_1^+ \rangle$	$4.6_{-0.2}^{+0.4}$	$4.42_{-0.15}^{+0.66}$	0.08	$4.5_{-0.3}^{+0.6}$	$600_{-40}^{+80}$
$\langle 8_1^+  E2  10_1^+ \rangle$	$4.1_{-0.9}^{+0.4}$	$4.0_{-0.8}^{+0.3}$	0.12	$4.1_{-0.9}^{+0.5}$	$490_{-110}^{+70}$
$\langle 1_1^-  E2  3_1^- \rangle$	$2.1_{-0.4}^{+0.3}$	$2.1_{-0.3}^{+0.2}$	0.06	$2.1_{-0.4}^{+0.4}$	$500_{-100}^{+100}$
$\langle 3_1^-  E2  5_1^- \rangle$	$3.0_{-0.4}^{+0.3}$	$2.4_{-0.3}^{+0.3}$	0.18	$2.7_{-0.7}^{+0.7}$	$470_{-120}^{+120}$
$\langle 5_1^-  E2  7_1^- \rangle$	$4.0_{-0.7}^{+0.8}$	$4.0_{-0.6}^{+0.6}$	0.16	$4.0_{-0.9}^{+0.9}$	$580_{-130}^{+130}$
$\langle 7_1^-  E2  9_1^- \rangle$	$6.0_{-1.1}^{+1.3}$	$5.1_{-0.8}^{+1.0}$	0.40	$5.5_{-1.3}^{+2.1}$	$700_{-200}^{+300}$
$\langle 0_1^+  E2  2_1^+ \rangle$	$0.22_{-0.02}^{+0.02}$	$0.266_{-0.014}^{+0.011}$	0.009	$0.24_{-0.05}^{+0.03}$	$76_{-16}^{+10}$
$\langle 2_1^+  E2  2_1^+ \rangle$	$0.41_{-0.05}^{+0.05}$	$0.63_{-0.03}^{+0.03}$	0.04	$0.52_{-0.20}^{+0.14}$	$140_{-50}^{+40}$
$\langle 2_1^+  E2  3_1^+ \rangle$	$-1.0_{-0.2}^{+1.7}$	$-1.37_{-0.14}^{+0.19}$	0.02	$-1.2_{-0.3}^{+1.9}$	$200_{-400}^{+100}$
$\langle 4_1^+  E2  4_1^+ \rangle$	$0.82_{-0.11}^{+0.11}$	$0.99_{-0.06}^{+0.06}$	0.013	$0.91_{-0.20}^{+0.14}$	$160_{-40}^{+30}$
$\langle 2_1^+  E2  0_\beta^+ \rangle$	$0.32_{-0.04}^{+0.04}$	$0.32_{-0.03}^{+0.03}$	0.006	$0.32_{-0.05}^{+0.05}$	$101_{-16}^{+16}$
$\langle 0_1^+  E3  3_1^- \rangle$	$0.91_{-0.06}^{+0.05}$	$0.86_{-0.06}^{+0.04}$	0.02	$0.88_{-0.08}^{+0.11}$	$2360_{-210}^{+300}$
$\langle 2_1^+  E3  1_1^- \rangle$	$0.49_{-0.26}^{+0.18}$	$0.57_{-0.21}^{+0.13}$	0.03	$0.5_{-0.4}^{+0.2}$	$1200_{-900}^{+500}$
$\langle 2_1^+  E3  3_1^- \rangle$	$-0.9_{-0.5}^{+0.3}$	$-0.2_{-0.3}^{+0.2}$	0.08	m.e.  < 1.5	< 3500
$\langle 2_1^+  E3  5_1^- \rangle$	$1.15_{-0.13}^{+0.09}$	$1.41_{-0.10}^{+0.08}$	0.06	$1.3_{-0.3}^{+0.2}$	$2300_{-500}^{+300}$
$\langle 4_1^+  E3  1_1^- \rangle$	$-0.6_{-0.5}^{+0.8}$	$-0.3_{-0.5}^{+0.8}$	0.08	m.e.  < 1.4	< 2900

in contrast to the behaviour of  $^{222-226}\text{Ra}$ , for which  $\Delta i_x$  tends to zero at higher rotational frequencies as expected for stable octupole deformation. The present measurements of  $E3$  moments in  $^{222}\text{Rn}$ , while consistent with this interpretation, are not sufficiently precise to distinguish between octupole vibration and deformation. The values of the intrinsic electric-octupole moments,  $Q_3$ , in  $^{222}\text{Rn}$  obtained from this analysis are presented in Figure 12. The values of  $Q_3$  for the  $0^+ \rightarrow 3^-$  and  $2^+ \rightarrow 5^-$  transitions, respectively  $2360_{-210}^{+300}\text{efm}^3$  ( $B(E3; 3_1^- \rightarrow 0_1^+) = 37_{-6}^{+10}$  W.u.) and  $2300_{-500}^{+300}\text{efm}^3$ , are similar as expected for a static pear shape or for an octupole phonon coupled to the ground-state band. The value of  $Q_3$  for the  $2^+ \rightarrow 1^-$  transition,  $1200_{-900}^{+500}\text{efm}^3$ , is smaller, while only upper limits were obtained for the  $2^+ \rightarrow 3^-$  and  $1^- \rightarrow 4^+$  transitions.

The experimental intrinsic electric-octupole moments,  $Q_3$ , observed for Rn and Ra isotopes are presented in Figure 13. The value for  $^{222}\text{Rn}$  is consistent with that measured for the octupole-vibrational nuclei  $^{220}\text{Rn}$ ,  $^{228}\text{Ra}$ , but having an enhanced value as observed for  $^{222-226}\text{Ra}$  cannot be excluded. Also shown in the figure are the most recent calculations of  $E3$  moments using the the mean-field Hartree-Fock-Bogoliubov (HFB) method with the 2-D Gogny D1S energy density functional (EDF) [36], calculations using the Quadrupole-Octupole-Collective-Hamiltonian with

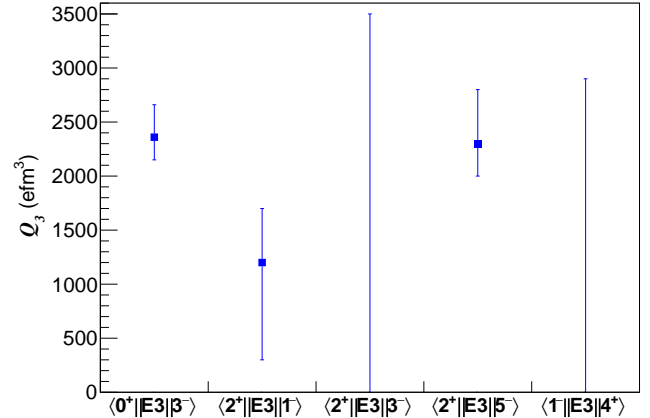


FIG. 12. Calculated values of intrinsic octupole moments,  $Q_3$ , taken from the adopted values in Table II.

relativistic PC-PK1 EDF [37] and Covariant Density EDF [38] for both Rn and Ra isotopes, and calculations using spdf-IBM-2 for Rn isotopes [39]. While the radium experimental values are reproduced reasonably well by theory, the microscopic theory calculations tend to underestimate, and the algebraic theory overestimates the observed values for the radon isotopes measured so far.

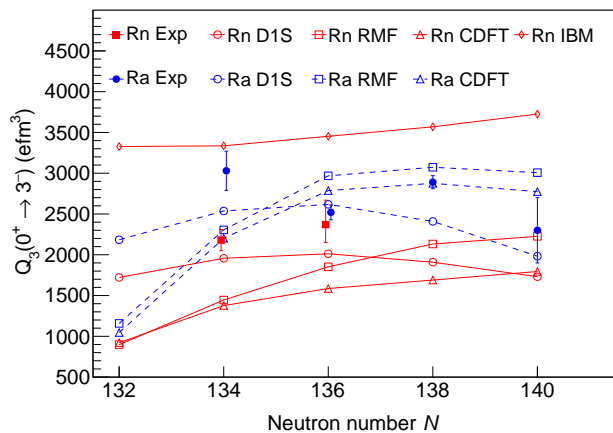


FIG. 13. Measured intrinsic electric-octupole moments of Rn and Ra isotopes corresponding to the  $\langle 0_1^+ || E3 || 3_1^- \rangle$  reduced matrix elements. The experimental results, taken from this work and previous works [3–5], are compared with theoretical values using the 2-D Gogny D1S force (D1S) [36], QOCH with relativistic PC-PK1 EDF (RMF) [37] and Covariant Density EDF (CDFT) [38] for both Rn (red) and Ra (blue) isotopes, and calculations using spdf-IBM-2 for Rn (red) isotopes (IBM) [39].

## V. CONCLUSION

The structure of excited states in  $^{222}\text{Rn}$  was studied in a Coulomb-excitation experiment performed with the Miniball spectrometer at CERN’s HIE-ISOLDE facility. Measurements of  $\gamma$ -ray yields following bombardment of  $^{120}\text{Sn}$  and  $^{60}\text{Ni}$  targets were used to extract matrix elements connecting states in both the ground-state and negative-parity band. Transition E2 matrix elements within the ground-state and octupole bands were measured up to  $10 \hbar$  and the results were consistent with a constant intrinsic electric-quadrupole moment. Additionally, four excited states attributed to other collective modes were identified in this work, and their de-excitation paths of these levels determined using  $\gamma$ - $\gamma$

coincidences. The measured value of  $B(E2; 2_2^+ \rightarrow 0_1^+)$  is similar to that found in  $^{220}\text{Rn}$ .

The intrinsic  $E3$  moments for the  $0^+ \rightarrow 3^-$  and  $2^+ \rightarrow 5^-$  transitions were found to be approximately constant and have similar values to other nuclei in this mass region, while a smaller value was found for the  $2^+ \rightarrow 1^-$  transition and only upper limits could be determined for two other transitions. The feeding of low-lying negative parity states from higher-lying collective bands was found to be substantial at bombarding energies of around 4 MeV/u. This feeding limits the statistical accuracy of any measured  $E3$  moments if there is a scarcity of accompanying spectroscopic data. Nevertheless the available data for  $^{222}\text{Rn}$  measured in this work and  $^{220}\text{Rn}$  measured previously [4] indicate substantial octupole correlations.

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