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Evolution of Octupole Deformation in Radium Nuclei from Coulomb Excitation of Radioactive $^{222}\text{Ra}$ and $^{228}\text{Ra}$ Beams

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There is sparse direct experimental evidence that atomic nuclei can exhibit stable “pear” shapes arising from strong octupole correlations. In order to investigate the nature of octupole collectivity in radium isotopes, electric octupole ($E_3$) matrix elements have been determined for transitions in $^{222}\text{Ra}$ nuclei using the method of sub-barrier, multistep Coulomb excitation. Beams of the radioactive radium isotopes were provided by the HIE-ISOLDE facility at CERN. The observed pattern of $E_3$ matrix elements for different nuclear transitions is explained by describing $^{222}\text{Ra}$ as pear shaped with stable octupole deformation, while $^{228}\text{Ra}$ behaves like an octupole vibrator.

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There are many theoretical and experimental indications that atomic nuclei can exhibit reflection asymmetry in the intrinsic frame, and observation of low-lying quantum states in many nuclei with even $Z$, $N$ having total angular momentum and parity of $I^π = 3^−$ is indicative of the presence of octupole correlations (see Ref. [1] and references therein). Typically, the electric octupole ($E_3$) moment for the transition to the ground state is tens of single-particle units, suggesting that the octupole instability arises from a collective effect and leads to a pear-shaped distortion of the nuclear shape. What is less clear, however, is whether in some nuclei this distortion is stable, i.e., the nucleus assumes a permanent pear shape, or whether it is dynamic and the nucleus undergoes octupole vibrations. Evidence has been presented that $^{224}\text{Ra}$ and $^{226}\text{Ra}$ have static octupole deformation on account of an enhancement in the $E_3$ moment in these nuclei [2,3]. Large $E_3$ moments have also been recently measured for neutron-rich barium isotopes, suggesting that, within the experimental uncertainty, these nuclei could have octupole deformation [4,5].
excitation experiment with radioactive \(^{222}\)Ra, \(^{228}\)Ra, and \(^{60}\)Ni. The \(\gamma\) rays were corrected for Doppler shift assuming that they are emitted from the scattered projectile. Random coincidences between Miniball and the silicon detector have been subtracted. The transitions that give rise to the observed full-energy peaks are labeled by the spin and parity of the initial and final states.

The only example of an octupole unstable nucleus other than \(^{226}\)Ra where stable beams have been used to obtain a complete set of \(E3\) matrix elements is \(^{148}\)Nd [6].

In this Letter, results from a multistep, Coulomb-excitation experiment with radioactive \(^{222,228}\)Ra beams are reported. By examining the pattern of \(E3\) matrix elements between different transitions in these nuclei and comparing them to those in \(^{224,226}\)Ra and \(^{148}\)Nd, a distinction can be made between those isotopes having stable octupole deformation and those behaving like octupole vibrators. This observation is relevant for the search for permanent electric dipole moments in radium atoms [7–9], that would indicate sizable \(CP\) violation requiring a substantial revision of the standard model.

The radioactive isotopes \(^{222}\)Ra (\(Z = 88, N = 134\)) and \(^{228}\)Ra (\(Z = 88, N = 140\)) were produced by spallation in a thick uranium carbide primary target bombarded by \(\approx 10^{13}\) protons/s at 1.4 GeV from the CERN PS Booster. The ions, extracted from a tungsten surface ion source were stripped to charge states of \(51^+\) and \(53^+\), respectively, for \(^{222}\)Ra and \(^{228}\)Ra and accelerated in HIE-ISOLDE to an energy of 4.31 MeV/nucleon. The radioactive beams, with intensities between \(5 \times 10^{4}\) and \(2 \times 10^{5}\) ions/s bombarded secondary targets of \(^{60}\)Ni and \(^{120}\)Sn of thickness 2.1 mg/cm\(^2\). Gamma rays emitted following the excitation of the target and projectile nuclei were detected in Miniball [10], an array of 24 high-purity germanium detectors, each with sixfold segmentation and arranged in eight triple clusters. The scattered projectiles and target recoils were detected in a highly segmented silicon detector, distinguished by their differing dependence of energy with angle measured in the laboratory frame of reference.

Representative spectra from the Coulomb-excited \(^{222,228}\)Ra are shown in Fig. 1; in the spectra the \(\gamma\)-ray energies are corrected for Doppler shift assuming emission from the scattered projectile. The spectra were incremented when a target recoil was detected in coincidence with \(\gamma\) rays within a 450-ns time window; these data were corrected for random events. The fraction of the isobar \(^{222}\)Fr in the beam was estimated to be about 20\% by observing \(\gamma\) rays from the \(\alpha\)-decay daughters at the beam dump. By lowering the temperature of the transfer line from the ion source a nearly pure beam of \(^{222}\)Fr could be produced; apart from \(\gamma\) rays, no discernable structure was observed arising from Coulomb excitation of the odd-odd nucleus in the particle-gated, Doppler-corrected spectrum. For the \(^{222}\)Ra beam, the fraction of isobaric contamination was estimated to be \(\approx 1\%\).

For both \(^{222}\)Ra and \(^{228}\)Ra the spectra reveal strong population of the ground-state band of positive-parity states, populated by multiple electric quadrupole (\(E2\)) Coulomb excitation, and substantial population of the octupole band of negative-parity states, populated by \(E3\) excitation. The yields of the observed \(\gamma\)-ray transitions detected in Miniball were measured for four ranges of the recoil angle of the target nucleus for each target, between 21.5° and 55.5° for the \(^{120}\)Sn target and between 17.8° and 55.5° for the \(^{60}\)Ni target. The yield data were combined with existing \(\gamma\)-ray branching ratios to provide input to the Coulomb-excitation analysis code GOSIA [11–13]. The GOSIA code performs a least-squares fit to the \(E1\) (\(\lambda = 1, 2, 3\)) matrix elements (m.e.s), which either can be treated as free parameters, can be coupled to other matrix elements, or can be fixed. Energy-level schemes that are included in the
analysis are given in the Supplemental Material [14]. A total of 114 data for $^{222}\text{Ra}$ were fitted to 42 variables, while for $^{228}\text{Ra}$ 121 data were fitted to 41 variables. The starting values of each of the freely varied matrix elements were drawn randomly, within reasonable limits; the values obtained following the fitting procedure were found to be independent of the starting points. Examples of fits to the experimental data can be found in Ref. [14].

For both nuclei the $E1$ couplings between the ground-state and negative-parity bands and the $E2$ couplings for transitions within the ground state and within the negative-parity bands, with the exception of the $2^+ \rightarrow 0^+$ transition, were treated as free parameters. Under the experimental conditions described here, the probability of populating the $2^+$ state is $> 90\%$ and it was not possible to determine the $\langle 0^+|[E2]|2^+\rangle$ and $\langle 2^+|[E2]|2^+\rangle$ m.e.s independently. The latter was therefore allowed to vary freely and the $\langle 0^+|[E2]|2^+\rangle$ matrix element was coupled to the $\langle 2^+|[E2]|4^+\rangle$ matrix element assuming the validity of the rotational model; this assumption is based on the behavior of nuclei where the lifetimes of the $2^+$ and $4^+$ states have been measured and for which the lowest transitions behave collectively [14]. For the $E3$ m.e.s the lowest couplings were treated as free parameters; m.e.s between higher-lying states, $\langle I^\pm|[E3]|I'^\mp\rangle$, were coupled to m.e.s between lower-lying states, $\langle (I - 2)^\pm|[E3]|(I' - 2)^\mp\rangle$, assuming the validity of the rotational model. $E4$ matrix elements were also included in the fitting procedure; these were calculated assuming the rotational model and a constant value of the hexadecapole moment, derived from the theoretical values of $\beta_4$ [22]. $E2$ (and magnetic dipole) couplings to high-lying $K^t = 0^+$ and $K^t = 2^+$ bands were also taken into account. The relative phase of $Q_1$ and $Q_3$ was investigated, as although the overall phase of the $E1$ and $E3$ matrix elements is arbitrary, the fit is sensitive to the relative phase of $E3$ matrix elements as well as the phase difference between the $E1$ and $E3$ matrix elements. The difference in chi-square for the fit favored $Q_1$ and $Q_3$ having the same sign for $^{222}\text{Ra}$ and the opposite sign for $^{228}\text{Ra}$, and these phases were adopted in the final fits. These values are consistent with macroscopic-microscopic calculations [23] and constrained HFB calculations [24] that predict a decreasing value of $Q_1$ with neutron number for radium isotopes, crossing zero for $^{222}\text{Ra}$ as experimentally verified [25].

Table I gives the values of $E2$ and $E3$ matrix elements for $^{222}\text{Ra}$ and $^{228}\text{Ra}$ obtained in this work. The $E1$ matrix

| $\langle I'|[E] |I\rangle$ | $^{222}\text{Ra}$ m.e. (eb$^{1/2}$) | $^{228}\text{Ra}$ m.e. (eb$^{1/2}$) | $Q_1$ (efm$^2$) | $Q_1$ (efm$^2$) |
|---------------------------|-------------------------------|-------------------------------|----------------|----------------|
| $\langle 2^+|[E2]|2^+\rangle$ | $-1.3 \pm 0.5$ | $330 \pm 140$ | $-0.3 \pm 1.7$ | $90 \pm 400$ |
| $\langle 2^+|[E2]|4^+\rangle$ | $2.98 \pm 0.15$ | $590 \pm 30$ | $3.87 \pm 0.19$ | $770 \pm 40$ |
| $\langle 4^+|[E2]|4^+\rangle$ | $-2.8 \pm 0.5$ | $580 \pm 100$ | $5.11 \pm 0.26$ | $800 \pm 40$ |
| $\langle 4^+|[E2]|6^+\rangle$ | $3.57 \pm 0.18$ | $559 \pm 28$ | $5.89 \pm 0.29$ | $790 \pm 40$ |
| $\langle 6^+|[E2]|8^+\rangle$ | $4.15 \pm 0.23$ | $560 \pm 30$ | $7.5 \pm 0.4$ | $890 \pm 50$ |
| $\langle 8^+|[E2]|10^+\rangle$ | $4.7 \pm 0.5$ | $560 \pm 60$ | $7.1^{+0.5}_{-0.3}$ | $770^{+60}_{-40}$ |
| $\langle 10^+|[E2]|12^+\rangle$ | $2.35 \pm 0.22$ | $560 \pm 50$ | $3.8 \pm 0.5$ | $890 \pm 120$ |
| $\langle 1^+|[E2]|3^+\rangle$ | $3.1 \pm 0.4$ | $530 \pm 70$ | $3.9_{-0.4}^{+0.8}$ | $670_{-130}^{+20}$ |
| $\langle 3^+|[E2]|5^+\rangle$ | $4.4 \pm 0.4$ | $630 \pm 60$ | $4.0 \pm 0.9$ | $580 \pm 130$ |
| $\langle 5^+|[E2]|7^+\rangle$ | $6.0 \pm 1.0$ | $760 \pm 120$ | $5.9 \pm 1.0$ | $740 \pm 130$ |
| $\langle 7^+|[E2]|9^+\rangle$ | $578 \pm 18$ | $798 \pm 21$ | $870 \pm 140$ |
| $Q_2$ (rotational model) | $1.13 \pm 0.09$ | $3030 \pm 240$ | $0.87 \pm 0.15$ | $2300 \pm 400$ |
| $\langle 2^+|[E3]|3^+\rangle$ | $0.85 \pm 0.24$ | $2000 \pm 600$ | $1.36 \pm 0.23^*$ | $3200 \pm 600$ |
| $\langle 2^+|[E3]|5^+\rangle$ | $-0.9 \pm 0.5$ | $2100 \pm 1200$ | $-0.06_{-0.16}^{+0.25}$ | $150_{-500}^{+360}$ |
| $\langle 4^+|[E3]|1^+\rangle$ | $1.79 \pm 0.20$ | $3100 \pm 400$ | $1.71 \pm 0.23^*$ | $3000 \pm 400$ |
| $\langle 4^+|[E3]|3^+\rangle$ | $-2.1 \pm 0.5^*$ | $4400 \pm 1000$ | $0.4_{-1.1}^{+0.7}$ | $-800_{-1400}^{+2300}$ |
| $\langle 4^+|[E3]|5^+\rangle$ | $2.6_{-0.9}^{+0.3}$ | $5500_{-1800}^{+1300}$ | $1300_{-500}^{+360}$ |
| $\langle 4^+|[E3]|7^+\rangle$ | $3.3_{-0.5}^{+0.3}$ | $4600_{-600}^{+500}$ | $4600_{-600}^{+500}$ |
| $Q_3$ (rotational model) | $0.87 \pm 0.15$ | $2300 \pm 400$ | $2230 \pm 290$ |
elements are given in Ref. [14]. Those E3 m.e.s marked
with an asterisk are coupled to m.e.s between higher-lying
states and as such are not completely independently
determined; however, the fit is mostly influenced by the
value of the lowest matrix element. The diagonal E2 matrix
elements are all coupled to the adjacent transition m.e.s
except for those presented in Table I, which are independ-
dently determined. In the GOSIA fit the statistical errors
for each fitted variable were calculated taking into account
correlations between all variables. Independent sets of fitted
values were also obtained by varying the constant hex-
adecapole moment used to calculate the E4 m.e.s between
zero and double the notional value, varying the target
thickness by 5%, the beam energy by 1%, the distance
between the target and the particle detector by 7.5%,
and the sign of the E2 couplings to the higher-lying
collective bands.

The variations seen in the fitted values are included in the
final uncertainties given in Table I. For 228Ra the value of
the intrinsic quadrupole moment, Q2, derived from the
measured value of 〈2+∥E2∥4+〉, 770 ± 40 efm², agrees
with the values determined from the 2+ lifetime, 775 ±
14 efm² and the 4+ lifetime, 780 ± 6 efm², as reported in
Ref. [16]. For 226Ra, the value is 590 ± 30 efm², signifi-
cantly smaller than the value derived from the measured
lifetime of the 2+ state, 673 ± 13 efm² [26]. It is noted that
the value of Q2 for 228Ra extrapolated from the 2+ lifetime
for 228Ra on the basis of B(E2; 0+ → 2+) systematics [27],
is 593 ± 11 efm², in good agreement with the current
measurement. Fitted values of Q2 and Q3 assuming that the
E3 matrix elements and Q3 are related by the rotational
model are also given in Table I. The values for λ = 3
indicate that the octupole collectivity in 228Ra is signifi-
cantly lower than for 226Ra.

The values of Q1 and Q2 for all the measured matrix
elements are shown in Figs. 2 and 3, respectively. The
FIG. 2. Absolute values of the intrinsic dipole moments Q1 as a
function of spin. The values are deduced from the measured
matrix elements [14], and correspond to transitions between
states with spin I and I − 1.

FIG. 3. Values of the intrinsic quadrupole moments Q2 plotted
as a function of spin. The values are deduced from the measured
matrix elements given in Table I. The values correspond to
transitions between states with spin I and I − 2; in some cases
they are also derived from diagonal matrix elements. The solid
horizontal lines correspond to the values of Q2 obtained assum-
ing that the matrix elements are related by the rotational model.

nearly constant values of Q2 as a function of spin for
transitions in both positive- and negative-parity bands is
consistent with stable quadrupole deformation. Smaller
values of Q2, although with large uncertainty, were
determined from the (2+∥E2∥2+) matrix element for both
nuclei. Such behavior was also observed in 228Ra, inter-
preted as arising from deviations from axial symmetry [3].
The values of the intrinsic electric octupole moment Q3 for
transitions in 226Ra and 228Ra are shown in Fig. 4. In the
figure, the values of Q3 are shown separately for transitions
I −→ (I + 1)−, I + → (I + 3)−, I − → (I + 1)+, and
I − → (I + 3)+, and are compared with values determined
FIG. 4. Values of the intrinsic octupole moments Q3 plotted
as a function of spin. The values are deduced from the measured
matrix elements given in Table I. Here the values of Q3 are
shown separately for transitions connecting I + → (I + 1)−,
I + → (I + 3)−, I − → (I + 1)+, and I − → (I + 3)+. The upper
dashed line is the average value of Q3(0+1; 3−1) for the radium
isotopes. To aid comparison, the values of Q3 for 148Nd have been
multiplied by 1.78.
for the same transitions in $^{224,226}$Ra [2,3] and $^{148}$Nd [6]. The values for $^{148}$Nd are multiplied by a factor so that the value of $Q_3$ deduced from $|\langle E3|3^{-}\rangle|$ is the same as the average value for the radium isotopes. It is observed that the values of $Q_3$ for all transitions in $^{222,224,226}$Ra are approximately constant, consistent with the picture of a rotating pear shape. In contrast, the values of $Q_3$ corresponding to the $2^+ \rightarrow 3^-$ and $1^- \rightarrow 4^+$ transitions in $^{228}$Ra are close to zero, as observed for $^{140}$Nd. It is unlikely that this can be accounted for by $K$ mixing [12] as the $K^2 = 1$ band lies much higher in energy for these nuclei [28].

The contrast in the behavior of the $E3$ moments of $^{228}$Ra (and $^{140}$Nd) compared to the lighter radium isotopes is also present in the behavior of their energy levels, as shown in Fig. 5. Here $\Delta i_z$, the difference in aligned angular momentum between negative- and positive-parity states at the same rotational frequency $\omega$, is plotted as a function of $\hbar \omega$ for the five nuclei. The behavior of $\Delta i_z$ can reveal information regarding the nature of the octupole correlations [29,30]. For $^{140}$Nd, the value of $\Delta i_z \sim 3\hbar$ for all values of rotational frequency, and for $^{228}$Ra it approaches $3\hbar$ when $\hbar \omega \rightarrow 0.15$ MeV. This behavior is expected for octupole vibrators, where the octupole phonon aligns to the rotation axis. It is conjectured here that the observation of near-zero values of $Q_3$ for some transitions in $^{228}$Ra (and $^{140}$Nd) is consistent with the octupole-vibrator description. The interpretation of the behavior of energy levels for $^{222,224,226}$Ra in terms of rotating pear shapes is less obvious as it is dominated by pairing effects near the ground state; other interpretations of this behavior, e.g., the condensation of rotational-aligned octupole phonons [31], do not require the nucleus to have a permanent octupole distortion. On the other hand, highly collective $E2$ and $E3$ transition strengths are nearly independent of pairing and single particle effects and are a much better measure of the nuclear shape. The observed enhancement and rotorlike pattern of the electric octupole moments $Q_3$ provide compelling evidence that $^{222}$Ra together with $^{224,226}$Ra have stable octupole deformation. This confirms theoretical predictions, e.g., Refs. [22,32,33], that the boundary of octupole deformation lies at $Z \approx 88$ and at $N \approx 138$; it has already been established that even-even radon ($Z = 86$) nuclei having similar neutron numbers behave like octupole vibrators [34]. It is concluded that the differing patterns of $E3$ matrix elements observed for $^{222,226}$Ra are a consequence of the stability of the octupole shape for each nucleus. Any model of quadrupole-octupole coupling that describes this behavior should be capable of calculating values of $Q_3$ for different $E3$ transitions including the critical $3^- \rightarrow 2^+$ transition, as has been performed for $^{224}$Ra [35].

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