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Author(s): Butler, P. A.; Gaffney, L. P.; Spagnoletti, P.; Abrahams, K.; Bowry, M.; Cederkäll, J.; de Angelis, G.; De Witte, H.; Garrett, P. E.; Goldkuhle, A.; Henrich, C.; Illana, A.; Johnston, K.; Joss, D. T.; Keatings, J. M.; Kelly, N. A.; Komorowska, M.; Konki, J.; Kröll, T.; Lozano, M.; Nara Singh, B. S.; O'Donnell, D.; Ojala, J.; Page, R. D.; Pedersen, L. G.; Raison, C.; Reiter, P.; Rodriguez, J. A.; Rosiak, D.; Rothe, S.; Zielinska, M.

Title: Evolution of Octupole Deformation in Radium Nuclei from Coulomb Excitation of Radioactive ^{222}Ra and ^{228}Ra Beams

Year: 2020

Version: Published version

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Please cite the original version:

Butler, P. A., Gaffney, L. P., Spagnoletti, P., Abrahams, K., Bowry, M., Cederkäll, J., de Angelis, G., De Witte, H., Garrett, P. E., Goldkuhle, A., Henrich, C., Illana, A., Johnston, K., Joss, D. T., Keatings, J. M., Kelly, N. A., Komorowska, M., Konki, J., Kröll, T., . . . Zielinska, M. (2020). Evolution of Octupole Deformation in Radium Nuclei from Coulomb Excitation of Radioactive ^{222}Ra and ^{228}Ra Beams. *Physical Review Letters*, 124(4), Article 042503. <https://doi.org/10.1103/PhysRevLett.124.042503>

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

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(Received 15 November 2019; published 31 January 2020)

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 ($E3$) matrix elements have been determined for transitions in $^{222,228}\text{Ra}$ 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 $E3$ matrix elements for different nuclear transitions is explained by describing ^{222}Ra as pear shaped with stable octupole deformation, while ^{228}Ra behaves like an octupole vibrator.

DOI: [10.1103/PhysRevLett.124.042503](https://doi.org/10.1103/PhysRevLett.124.042503)

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 $I^\pi = 3^-$ is indicative of the presence of octupole correlations (see Ref. [1] and references therein). Typically, the electric octupole ($E3$) 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}Ra and ^{226}Ra have static octupole deformation on account of an enhancement in the $E3$ moment in these nuclei [2,3]. Large $E3$ 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].

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analysis are given in the Supplemental Material [14]. A total of 114 data for ^{222}Ra were fitted to 42 variables, while for ^{228}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 β_λ [22]. $E2$ (and magnetic dipole) couplings to high-lying $K^\pi = 0^+$ and $K^\pi = 2^+$ 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}Ra and the opposite sign for ^{228}Ra , and these phases were adopted in the final fits. These values are consistent with macroscopic-microscopic calculations [23] and constrained HFBCS calculations [24] that predict a decreasing value of Q_1 with neutron number for radium isotopes, crossing zero for ^{224}Ra as experimentally verified [25].

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

TABLE I. Values of $E2$ and $E3$ matrix elements measured in the present experiment. The intrinsic moments Q_λ are derived from each matrix element using $\langle I_i || \mathcal{M}(E\lambda) || I_f \rangle = \sqrt{(2I_i + 1)}\sqrt{(2\lambda + 1)}/16\pi(I_i 0 \lambda 0 | I_f 0) Q_\lambda$. The uncertainties include the 1σ statistical error from the fit ($\chi^2 + 1$ type) and the systematic contributions. The $E3$ m.e.s marked with an asterisk are coupled to higher-lying m.e.s. The $\langle 0^+ || E2 || 2^+ \rangle$ and $\langle 2^+ || E2 || 4^+ \rangle$ m.e.s are coupled. Values of Q_λ fitted assuming that the m.e.s are related by the rotational model are also given.

$\langle I E\lambda I' \rangle$	^{222}Ra		^{228}Ra	
	m.e. (eb $^{3/2}$)	Q_λ (efm $^\lambda$)	m.e. (eb $^{3/2}$)	Q_λ (efm $^\lambda$)
$\langle 2^+ E2 2^+ \rangle$	-1.3 ± 0.5	330 ± 140	-0.3 ± 1.7	90 ± 400
$\langle 2^+ E2 4^+ \rangle$	2.98 ± 0.15	590 ± 30	3.87 ± 0.19	770 ± 40
$\langle 4^+ E2 4^+ \rangle$	-2.8 ± 0.5	580 ± 100		
$\langle 4^+ E2 6^+ \rangle$	3.57 ± 0.18	559 ± 28	5.11 ± 0.26	800 ± 40
$\langle 6^+ E2 8^+ \rangle$	4.15 ± 0.23	560 ± 30	5.89 ± 0.29	790 ± 40
$\langle 8^+ E2 10^+ \rangle$	4.7 ± 0.5	560 ± 60	7.5 ± 0.4	890 ± 50
$\langle 10^+ E2 12^+ \rangle$			$7.1^{+0.5}_{-0.3}$	770^{+60}_{-40}
$\langle 1^- E2 3^- \rangle$	2.35 ± 0.22	560 ± 50	3.8 ± 0.5	890 ± 120
$\langle 3^- E2 5^- \rangle$	3.1 ± 0.4	530 ± 70	$3.9^{+0.4}_{-0.8}$	670^{+70}_{-130}
$\langle 5^- E2 7^- \rangle$	4.4 ± 0.4	630 ± 60	4.0 ± 0.9	580 ± 130
$\langle 7^- E2 9^- \rangle$	6.0 ± 1.0	760 ± 120	5.9 ± 1.0	740 ± 130
Q_2 (rotational model)		578 ± 18		798 ± 21
$\langle 0^+ E3 3^- \rangle$	1.13 ± 0.09	3030 ± 240	0.87 ± 0.15	2300 ± 400
$\langle 2^+ E3 1^- \rangle$	0.85 ± 0.24	2000 ± 600	$1.36 \pm 0.23^*$	3200 ± 600
$\langle 2^+ E3 3^- \rangle$	-0.9 ± 0.5	2100 ± 1200	$-0.06^{+0.23*}_{-0.16}$	150^{+360}_{-500}
$\langle 2^+ E3 5^- \rangle$	1.79 ± 0.20	3100 ± 400	$1.71 \pm 0.23^*$	3000 ± 400
$\langle 4^+ E3 1^- \rangle$	$-2.1 \pm 0.5^*$	4400 ± 1000	$0.4^{+0.7*}_{-1.1}$	-800^{+2300}_{-1400}
$\langle 4^+ E3 3^- \rangle$	$2.6^{+0.6*}_{-0.9}$	5500^{+1300}_{-1800}		
$\langle 4^+ E3 5^- \rangle$	$-1.7 \pm 1.0^*$	3200 ± 1800		
$\langle 4^+ E3 7^- \rangle$	$3.3^{+0.3*}_{-0.5}$	4600^{+500}_{-600}		
Q_3 (rotational model)		3120 ± 190		2230 ± 290

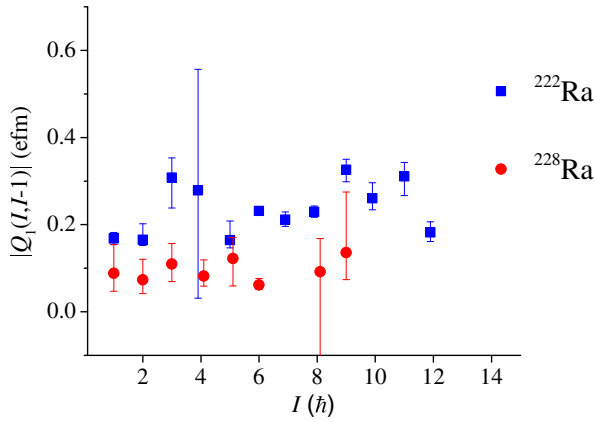


FIG. 2. Absolute values of the intrinsic dipole moments Q_1 as a function of spin. The values are deduced from the measured matrix elements [14], and correspond to transitions between states with spin I and $I - 1$.

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 independently 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 hexadecapole moment used to calculate the $E4$ m.e.s between zero and double the notional value, varying the target thickness by $\pm 5\%$, the beam energy by $\pm 1\%$, the distance between the target and the particle detector by $\pm 7.5\%$, 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 ^{228}Ra the value of the intrinsic quadrupole moment, Q_2 , derived from the measured value of $\langle 2^+ || E2 || 4^+ \rangle$, $770 \pm 40 \text{ efm}^2$, agrees with the values determined from the 2^+ lifetime, $775 \pm 14 \text{ efm}^2$ and the 4^+ lifetime, $780 \pm 6 \text{ efm}^2$, as reported in Ref. [16]. For ^{222}Ra , the value is $590 \pm 30 \text{ efm}^2$, significantly smaller than the value derived from the measured lifetime of the 2^+ state, $673 \pm 13 \text{ efm}^2$ [26]. It is noted that the value of Q_2 for ^{222}Ra extrapolated from the 2^+ lifetime for ^{228}Ra on the basis of $B(E2; 0^+ \rightarrow 2^+)$ systematics [27], is $593 \pm 11 \text{ efm}^2$, in good agreement with the current measurement. Fitted values of Q_2 and Q_3 assuming that the $E\lambda$ matrix elements and Q_λ are related by the rotational model are also given in Table I. The values for $\lambda = 3$ indicate that the octupole collectivity in ^{228}Ra is significantly lower than for ^{222}Ra .

The values of Q_1 and Q_2 for all the measured matrix elements are shown in Figs. 2 and 3, respectively. The

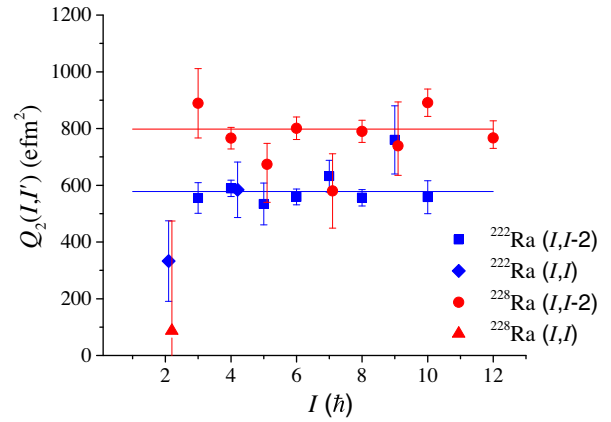


FIG. 3. Values of the intrinsic quadrupole moments Q_2 plotted as a function of spin. The values are deduced from the measured matrix elements given in Table I. The values correspond to transitions between states with spin I and $I - 2$; in some cases they are also derived from diagonal matrix elements. The solid horizontal lines correspond to the values of Q_2 obtained assuming that the matrix elements are related by the rotational model.

nearly constant values of Q_2 as a function of spin for transitions in both positive- and negative-parity bands is consistent with stable quadrupole deformation. Smaller values of Q_2 , although with large uncertainty, were determined from the $\langle 2^+ || E2 || 2^+ \rangle$ matrix element for both nuclei. Such behavior was also observed in ^{226}Ra , interpreted as arising from deviations from axial symmetry [3]. The values of the intrinsic electric octupole moment Q_3 for transitions in ^{222}Ra and ^{228}Ra are shown in Fig. 4. In the figure, the values of Q_3 are shown separately for transitions $I^+ \rightarrow (I + 1)^-$, $I^+ \rightarrow (I + 3)^-$, $I^- \rightarrow (I + 1)^+$, and $I^- \rightarrow (I + 3)^+$, and are compared with values determined

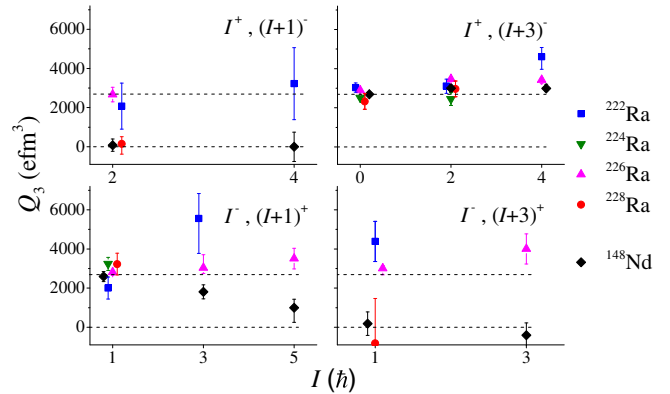


FIG. 4. Values of the intrinsic octupole moments Q_3 plotted as a function of spin. The values are deduced from the measured matrix elements given in Table I. Here the values of Q_3 are shown separately for transitions connecting $I^+ \rightarrow (I + 1)^-$, $I^+ \rightarrow (I + 3)^-$, $I^- \rightarrow (I + 1)^+$, and $I^- \rightarrow (I + 3)^+$. The upper dashed line is the average value of $Q_3(0^+, 3^-)$ for the radium isotopes. To aid comparison, the values of Q_3 for ^{148}Nd have been multiplied by 1.78.

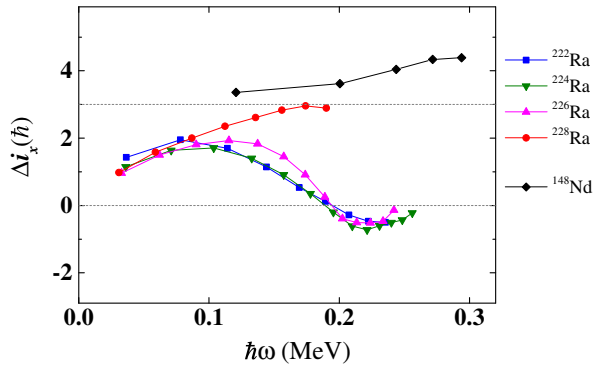


FIG. 5. The difference in aligned angular momentum, $\Delta i_x = i_x^- - i_x^+$, plotted as a function of rotational frequency ω . The upper dashed line corresponds to the vibrational limit, $\Delta i_x = 3\hbar$

for the same transitions in $^{224,226}\text{Ra}$ [2,3] and ^{148}Nd [6]. The values for ^{148}Nd are multiplied by a factor so that the value of \mathcal{Q}_3 deduced from $\langle 0^+ || E3 || 3^- \rangle$ is the same as the average value for the radium isotopes. It is observed that the values of \mathcal{Q}_3 for all transitions in $^{222,224,226}\text{Ra}$ are approximately constant, consistent with the picture of a rotating pear shape. In contrast, the values of \mathcal{Q}_3 corresponding to the $2^+ \rightarrow 3^-$ and $1^- \rightarrow 4^+$ transitions in ^{228}Ra are close to zero, as observed for ^{148}Nd . It is unlikely that this can be accounted for by K mixing [12] as the $K^\pi = 1^-$ band lies much higher in energy for these nuclei [28].

The contrast in the behavior of the $E3$ moments of ^{228}Ra (and ^{148}Nd) compared to the lighter radium isotopes is also present in the behavior of their energy levels, as shown in Fig. 5. Here Δi_x , the difference in aligned angular momentum between negative- and positive-parity states at the same rotational frequency ω , is plotted as a function of $\hbar\omega$ for the five nuclei. The behavior of Δi_x can reveal information regarding the nature of the octupole correlations [29,30]. For ^{148}Nd , the value of $\Delta i_x \sim 3\hbar$ 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 \mathcal{Q}_3 for some transitions in ^{228}Ra (and ^{148}Nd) is consistent with the octupole-vibrator description. The interpretation of the behavior of energy levels for $^{222,224,226}\text{Ra}$ 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 \mathcal{Q}_3 provide compelling evidence that

^{222}Ra together with $^{224,226}\text{Ra}$ 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 $^{222,228}\text{Ra}$ 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 \mathcal{Q}_3 for different $E3$ transitions including the critical $3^- \rightarrow 2^+$ transition, as has been performed for ^{224}Ra [35].

We are grateful to Doug Cline and the late Tomek Czosnyka who led the development of the Coulomb excitation analysis technique used in this work, and to Niels Bidault, Eleftherios Fadakis, Erwin Siesling, and Fredrick Wenander who assisted with the preparation of the radioactive beams. The support of the ISOLDE Collaboration and technical teams is acknowledged. This work was supported by the following Research Councils and Grants: Science and Technology Facilities Council (UK) Grants No. ST/P004598/1, No. ST/L005808/1, No. ST/R004056/1; Federal Ministry of Education and Research (Germany) Grants No. 05P18RDCIA, No. 05P15PKCIA, and No. 05P18PKCIA and the ‘‘Verbundprojekt 05P2018’’; National Science Centre (Poland) Grant No. 2015/18/M/ST2/00523; European Union’s Horizon 2020 Framework research and innovation programme 654002 (ENSAR2); Marie Skłodowska-Curie COFUND Grant (EU-CERN) 665779; Research Foundation Flanders and IAP Belgian Science Policy Office BriX network P7/12 (Belgium); GOA/2015/010 (BOF KU Leuven); RFBR (Russia) Grant No. 17-52-12015; and the Academy of Finland (Finland) Grant No. 307685.

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