Observation of Octupole Structures in Radon and Radium Isotopes and Their Contrasting Behavior at High Spin

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Multinucleon transfer reactions have been used, for the first time, to populate high-spin bands of alternating parity states in ^{218,220,222}Rn and ^{222,224,226}Ra. The behavior of the angular momentum alignment with rotational frequency for the Rn isotopes is very different when compared with Ra and Th isotopes with $N \approx 134$, indicating a transition from octupole vibrational to stable octupole deformation. Throughout the measured spin range the values of $|D_0/Q_0|$ remain constant for ^{222}Ra and ^{226}Ra and have a very small value for ^{224}Ra, suggesting that the charge and mass distributions are not affected appreciably by rotations. [S0031-9007(97)02928-1]

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Of all nuclear species, radium ($Z = 88$) and thorium ($Z = 90$) isotopes with $N \approx 134$ show the best evidence for octupole instability in their ground state [1–3]. These nuclei have low-lying negative-parity states and relatively strong $B(E1)$ values for the transitions between the bands of opposite parity; for the single case of ^{226}Ra large $B(E3)$ values have been measured consistent with its interpretation as a rotating pear shape [4]. The inaccessibility of these nuclei has, however, meant that there are large gaps in our knowledge of octupole effects in heavy nuclei. Comprehensive measurements of the high-spin behavior of the yrast octupole band exist only for the isotopes of thorium. For the radium 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 and virtually no information exists for ^{222}Ra. The scarce data do, however, suggest cancellation effects for the electric dipole moment for ^{224}Ra [5] which do not occur in the thorium isotopes. This effect is not properly established as the spin-dependent behavior for ^{222}Ra has not yet been measured. There are almost no data on the octupole structures for the radon isotopes. Systematic measurement of the variation of angular momentum with rotational frequency of the octupole bands should provide an insight into the nature of the strength of the octupole interactions in these nuclei.

In order to populate the nuclei of interest the properties of multinucleon transfer reactions have been exploited. Previously, yields have been mapped out following the bombardment of a thick ^{232}Th target with various projectiles [6]. As the reaction ^{136}Xe + ^{232}Th offered the largest yield for radon and radium isotopes with $N \approx 134$, this reaction was chosen in order to make spectroscopic measurements of the heavy products.

High-spin states in ^{218,220,222}Rn and ^{222,224,226}Ra were simultaneously populated following multinucleon transfer between ^{136}Xe and ^{232}Th. The ^{136}Xe projectile was accelerated to an energy of 833 MeV by the 88 in. cyclotron at Lawrence Berkeley National Laboratory. This bombarded a ^{232}Th target of thickness 36 mg/cm^2. Deexcitation gamma rays emitted from reaction products were collected for 49 h with the Gammasphere spectrometer which consisted of 73 large-volume (~75% relative efficiency) Compton-suppressed germanium detectors [7,8], yielding a total of 1.1×10^{10} unpacked triple coincidences.

The data were analyzed by examining the energy relationships of γ rays in a triple or quadruple coincidence with the aid of the LEVIT8R code [9]. Figure 1 contains representative spectra: Figures 1(a) and 1(b) show, respectively, coincident transitions in ^{218}Rn and in ^{224}Ra. The inset to Fig. 1(b) shows the efficiency and internal-conversion corrected intensities of the depopulating transitions above $I = 6\hbar$ in ^{224}Ra. These intensities, which are normalized such that the 4^+ to 2^+ transition has a total intensity of 100, were determined from spectra generated using several different coincidence relationships. Energy level schemes could be obtained for several nuclei by extending the decay schemes of low-lying states established in previous work. The level schemes of ^{218}Rn, ^{220}Rn, and ^{222}Rn are shown in Fig. 2. The dashed-line boxes on each decay scheme contain transitions which were observed in previous work [10,11]. Energy sums and intensity balance arguments were used to establish these decay schemes. The intensities of the transitions in each band

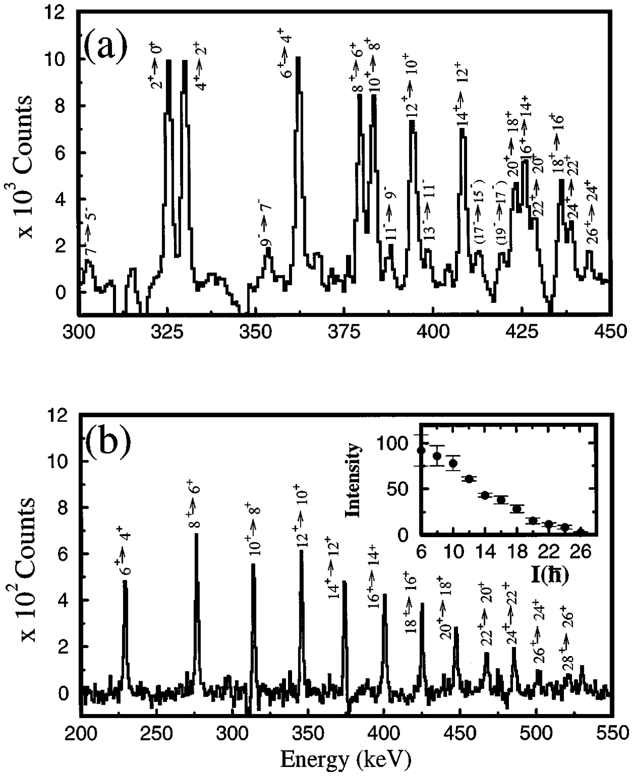

FIG. 1. (a) Gamma-ray spectrum obtained from threefold coincidences showing transitions in ^{218}Rn. The spectrum was produced by requiring that two of the three coincident γ rays have energies corresponding to transitions within the positive-parity band. (b) Gamma-ray spectrum obtained from fourfold coincidences showing transitions in ^{224}Ra. The spectrum was generated by demanding that one of the four coincident γ rays has the energy of the 4^+ to 2^+ transition and two others were also transitions in the positive-parity band. The inset is a plot of the intensities of the depopulating transitions in ^{224}Ra as a function of spin. The intensities are normalized such that the total intensity of the 4^+ to 2^+ transition is 100.

fall characteristically with increasing spin [see inset to Fig. 1(b)]. The transitions which are in strong coincidence with the known 4^+ to 2^+ and 2^+ to 0^+ transitions have been assigned positive parity. The band of regularly-spaced levels based on the known $I^\pi = 3^-$ state (tentatively assigned in ^{218}Rn) has been assigned negative parity. This is a reasonable assumption, particularly in the case of ^{220}Rn where the transition to the known 3^- state from the candidate 5^- state has been observed. While this transition is not observed in the adjacent radon isotopes, the spin and parity assignments in Fig. 2 lead to very systematic behavior with increasing mass for the radon isotopes. The level schemes of ^{222}Ra, ^{224}Ra and ^{226}Ra are also shown in Fig. 2. Previous knowledge of excited states [4,5,12,13] in these nuclei is also highlighted using dashed-line boxes. In ^{224}Ra and ^{226}Ra the known level schemes have been extended from $I^\pi = 12^+(11^-)$ to 26^+ (25^-) and 18^+ (17^-) to 28^+ (27^-), respectively. A consideration of internal conversion supports the assignment of electric, rather than magnetic, character to the interband transitions in ^{222}Ra. For example, the intensity

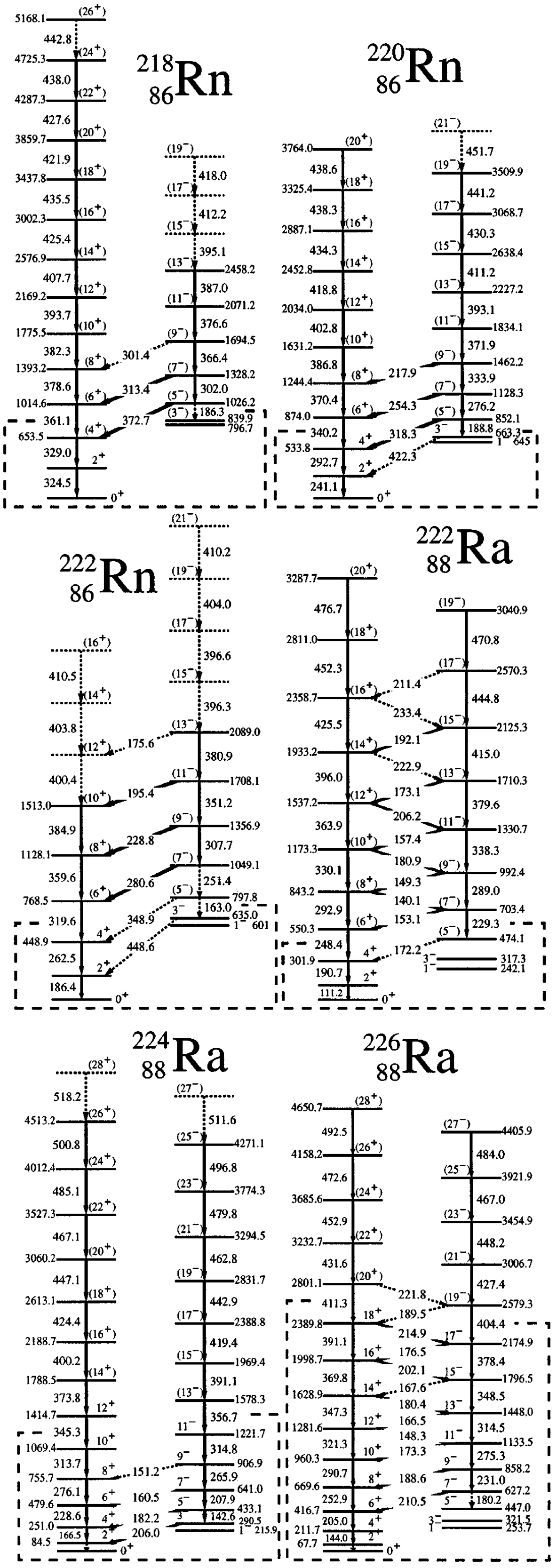

FIG. 2. The level schemes of ^{218,220,222}Rn and ^{222,224,226}Ra. Transition energies have errors which range from 0.2 keV for transitions between low-lying states in the ground state band to 0.5 keV for 5^- to 3^- and 7^- to 5^- transitions and transitions between the highest spin states observed.

of the 6^+ to 4^+ transition must be greater than or equal to the sum of the 7^- to 6^+ and 8^+ to 6^+ intensities after efficiency and internal-conversion correction. This condition can be satisfied if the 7^- to 6^+ transition is assigned $E1$ multipolarity.

Figure 3(a) shows the experimental alignment i_x as a function of rotational frequency ($\hbar\omega$) for the positive-parity band in three sets of isotones in the light-actinide region. The plots were produced using experimental data for ^{218}Rn, ^{220}Rn, ^{222}Rn, ^{222}Ra, and ^{224}Ra from the present work and ^{220}Ra, ^{222}Th, ^{224}Th and ^{226}Th from previous work [14,15]. The rotational frequencies were calculated using the expression $\hbar\omega = E_\gamma/[\sqrt{I(I+1)} - \sqrt{(I-2)(I-1)}]$. A variable moment of inertia reference with Harris parameters [16] of $J_0 = 31\hbar^2$ MeV^{-1} and $J_1 = 26\hbar^4$ MeV^{-3} has been subtracted. Any mass dependence in these parameters, which might be expected to be small, is neglected. The insets in Fig. 3(a) are plots of the difference in i_x (positive parity) and i_x (negative parity) Δi_x as a function of $\hbar\omega$. For ^{220,222}Rn the value of Δi_x is approximately three with some variation about this value throughout the whole frequency range. A larger variation is observed for ^{218}Rn, but overall the behavior of Δi_x for the radon isotopes is consistent with the description of the negative-parity states in terms of the coupling of an octupole phonon to the positive-parity states, where the phonon quickly aligns with the rotational axis as $\hbar\omega$ increases. Thus the radon isotopes are octupole vibrational in nature despite their having interleaved positive- and negative-parity states (see Rohoziński [17] for an explanation of this phenomenon, which is also apparent in ^{144,146}Ba—see [18] for level schemes). In contrast, for ^{222,224}Ra and ^{224,226}Th [see Fig. 3(a)] and for ^{226}Ra Δi_xbecomes small ($<1\hbar$) as $\hbar\omega$ increases, becoming close to zero for $\hbar\omega > 0.2$ MeV. These nuclei have very different behavior compared both with the radon isotopes and with heavier isotopes of radium and thorium, which strongly suggests that they behave as rotating octupole-deformed systems. This interpretation is supported by the experimental trend of the excitation energies of the low-lying negative parity states [1]. Additionally, both self-consistent [19] and macroscopic-microscopic calculations [3] predict a deeper octupole deformation energy minimum for $Z = 88$ and $Z = 90$ nuclei than for $Z = 86$ nuclei. The persistence of the well-ordered alternating parity sequence observed in ^{226}Ra (see Fig. 2) is at variance with recent calculations [20] using the coherent state model which predicts a breaking of this structure for $I^\pi > 18^+$.

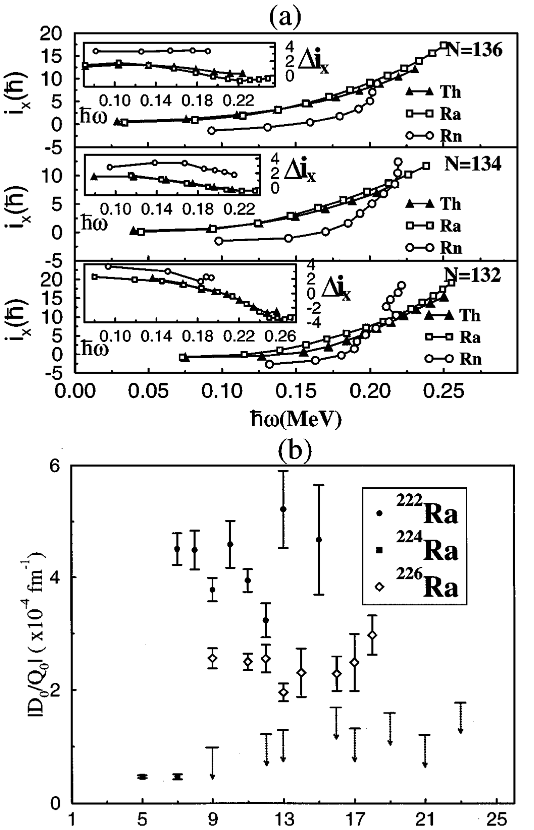

FIG. 3. (a) Plot of aligned angular momentum i_x as a function of rotational frequency $\hbar\omega$ for the positive-parity bands. The insets are plots of the difference in alignment Δi_x between the positive- and negative-parity bands as a function of $\hbar\omega$. The value of Δi_x was calculated by subtracting from the value of i_x for each negative-parity state an interpolated, smoothed value for the positive-parity states at the same value of $\hbar\omega$. (b) Plot of the ratio of the absolute magnitude of the intrinsic electric dipole and quadrupole moments ($|D_0/Q_0|$) as a function of spin for transitions deexciting states of spin I in ^{222}Ra, ^{224}Ra, and ^{226}Ra. 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.

Figure 3(a) also shows that the alignment for the positive-parity bands in thorium and radium isotopes exhibits smooth behavior up to high rotational frequencies. In contrast, all three isotopes of radon display sudden changes in alignment at $\hbar\omega \approx 0.20$ MeV. Cranked shell model (CSM) calculations performed with the Woods-Saxon deformed shell model potential [21] with "universal" parametrization [22] and deformation parameters of the three radon isotopes taken from Ref. [23] predict a strong interaction between the ground band and an aligned $j_{15/2}$ neutron band at a rotational frequency of $\hbar\omega \approx 0.25$ MeV and a weak interaction with an aligned $i_{13/2}$ proton band at $\hbar\omega \approx 0.25$ MeV for each nucleus. These band interactions are presumably responsible for the observed upbend in the experimental alignment plots. Such sudden increases in alignment are not seen in the radium and thorium isotones, perhaps because of the effect of the stronger octupole interaction between high- and low-spin orbitals [24] in these nuclei. It is interesting to note that in ^{220}Ra and ^{222}Th the alignment of the intruder orbits interferes with the octupole correlations for $\hbar\omega > 0.2$ MeV as evidenced by the higher alignment of the even spin sequence (Δi_x becomes negative), so that at sufficiently high frequencies the nuclei can become reflection symmetric [14]. Alignment effects have also been observed in octupole-vibrational nuclei in the lanthanide region [18,25].

In Fig. 2 one can see that interband $E1$ transitions depopulate states up to $I^\pi = 15^-$ (possibly 17^-) in ^{222}Ra and $I^\pi = 18^+(20^+)$ in ^{226}Ra but although the yrast band in ^{224}Ra has been observed up to $I^\pi = 26^+(28^+)$, no interband $E1$ transitions have been observed above $I^\pi = 7^-(9^-)$ in this nucleus. The absolute magnitude of the ratio of the intrinsic electric dipole moment (D_0) and intrinsic electric quadrupole moment (Q_0) was extracted from $B(E1)/B(E2)$ values using the rotational model [23]. These values are plotted in Fig. 3(b). The values measured for the transitions in ^{224}Ra are much lower than those in ^{222}Ra and ^{226}Ra. Weighted mean values of $|D_0/Q_0|$ for ^{222}Ra, ^{224}Ra, and ^{226}Ra were found to be, respectively, 4.02(11), 0.47(2), and $2.39(8) \times 10^{-4}$ fm^{-1}. For ^{222}Ra, ^{224}Ra, and ^{226}Ra the mean values from the present measurements are consistent with the previous measurements where available [1], but for $I > 8\hbar$ our data do not support the increase in D_0 with spin as reported [4] for ^{226}Ra. Macroscopic-microscopic calculations [23] give a very small value for the intrinsic electric dipole moment D_0 for ^{224}Ra because for this nucleus the droplet contribution and the shell correction term have opposite signs and almost cancel. Self-consistent calculations [26] also give a small $B(E1)$ value for the ground state transition in this nucleus. Figure 3(b) shows that a small value of $|D_0|$ persists to high angular momenta in ^{224}Ra. For ^{222}Ra and ^{226}Ra the values of $|D_0/Q_0|$ are much larger than for ^{224}Ra. However, for these nuclei the $|D_0/Q_0|$ values remain constant over the full spin range. If the electric quadrupole moment does not vary with spin, as observed approximately for the positive-parity band in ^{226}Ra [4], then similarly the value of D_0 does not vary. This implies that the small displacement between the center of mass and the center of charge, which gives rise to the electric dipole moment, is not varying appreciably with spin, and these reflection-asymmetric nuclei have differing charge and mass distributions which are rather stable under rotation. For ^{218}Rn, ^{220}Rn, and ^{222}Rn the weighted mean values of $|D_0/Q_0|$ were found to be, respectively, 0.97(8), 1.01(4), and $2.1(3) \times 10^{-4}$ fm^{-1}. These correspond to values of D_0 (≤ 0.1 e.fm) which are similar to that of ^{224}Ra, using values of Q_0 taken from ground state tabulations [27] and an empirical estimate [28] for ^{218}Rn. Such small values are predicted by macroscopic-microscopic theory [23] to be a consequence of the small quadrupole deformation in these nuclei. In contrast, the self-consistent calculation gives a large negative value for D_0 in ^{222}Rn [19].

In summary, multinucleon transfer reactions have been used to populate high-spin states in ^{218,220,222}Rn and ^{222,224,226}Ra. Interleaving bands of alternating parity have been observed in all of these nuclei. This represents the first observation of "octupole bands" in ^{218}Rn, ^{220}Rn, ^{222}Rn, and ^{222}Ra. Plots of the difference in alignment for the positive- and negative-parity states as a function of rotational frequency for these nuclei reveal contrasting behavior for the different nuclear systems. The radon isotopes behave like octupole vibrators, while the radium isotopes (together with ^{224,226}Th) display, by implication, behavior which is characteristic of nuclei having stable octupole deformation. The observed rapid increases in alignment of the positive-parity band in the radon isotopes is also expected if they have weaker octupole correlations. In ^{222,226}Ra the extracted value of $|D_0/Q_0|$ is approximately independent of spin, while in the case of ^{224}Ra the cancellation of contributions to the intrinsic electric dipole moment persists to high spins. The observation of the behavior of D_0 implies that the reflection-asymmetric charge and mass distributions are hardly affected by rotations in the radium isotopes. For all three radon isotopes the values of D_0 are small in comparison with ^{222}Ra and ^{226}Ra.

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[1] P. A. Butler and W. Nazarewicz, Rev. Mod. Phys. **68**, 349 (1996).
[2] I. Ahmad and P. A. Butler, Ann. Rev. Nucl. Part. Sci. **43**, 71 (1993).
[3] W. Nazarewicz *et al.*, Nucl. Phys. **A429**, 269 (1984).
[4] H. J. Wollersheim *et al.*, Nucl. Phys. **A556**, 261 (1993).
[5] R. J. Poynter *et al.*, Phys. Lett. B **232**, 447 (1989).
[6] J. F. C. Cocks *et al.* (to be published).
[7] A. M. Baxter *et al.*, Nucl. Instrum. Methods Phys. Res. Sect. A **317**, 101 (1992).
[8] M. P. Carpenter *et al.*, Nucl. Instrum. Methods Phys. Res. Sect. A **353**, 234 (1994).
[9] D. C. Radford, Nucl. Instrum. Methods Phys. Res. Sect. A **361**, 306 (1995).
[10] R. J. Poynter *et al.*, J. Phys. G **15**, 449 (1989).
[11] W. Kurcewicz *et al.*, Nucl. Phys. **A270**, 175 (1976).
[12] E. Ruchowska *et al.*, J. Phys. G **18**, 131 (1992).
[13] M. Marten-Tölle *et al.*, Z. Phys. A **336**, 27 (1990).
[14] J. F. Smith *et al.*, Phys. Rev. Lett. **75**, 1050 (1995).
[15] B. Ackermann *et al.*, Nucl. Phys. **A559**, 61 (1993).
[16] S. M. Harris, Phys. Rev. B **138**, 509 (1965).
[17] S. G. Rohoziński, Rep. Prog. Phys. **51**, 541 (1988).
[18] S. J. Zhu *et al.*, Phys. Lett. B **357**, 273 (1995).
[19] L. M. Robledo *et al.*, Phys. Lett. B **187**, 223 (1987).
[20] A. A. Raduta *et al.*, Nucl. Phys. **A608**, 11 (1996).
[21] J. Dudek *et al.*, J. Phys. G **5**, 1359 (1979).
[22] J. Dudek *et al.*, Phys. Rev. C **23**, 920 (1981).
[23] P. A. Butler and W. Nazarewicz, Nucl. Phys. **A533**, 249 (1991).
[24] W. Nazarewicz and P. Olanders, Nucl. Phys. **A441**, 420 (1985).
[25] W. Urban *et al.*, Phys. Lett. B **185**, 331 (1987).
[26] J. L. Egido and L. M. Robledo, Nucl. Phys. **A524**, 65 (1991).
[27] S. Raman *et al.*, At. Data Nucl. Data Tables **36**, 1 (1987).
[28] L. Grodzins, Phys. Lett. **2**, 88 (1962).