

Structural evolution of octupole-deformed radium isotopes within a coupled-channels method

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We have systematically investigated the spectroscopic properties in even-even $^{222-228}\text{Ra}$ by introducing the cluster degrees of freedom within the coupled-channels method, which allows the core ^{208}Pb to occupy the octupole 3^- state (Calc. I), as well as both the octupole 3^- and quadrupole 2^+ states (Calc. II), along with multipole interactions. Excellent agreement has been achieved between our calculated results and available data on energy spectra, reduced electromagnetic $E\lambda$ transition-matrix elements, and intrinsic electric dipole, quadrupole, and octupole Q_λ moments, pointing to the existence of strong octupole correlations in this region. The description of parity-doublet bands and electromagnetic-transition properties in both calculations is consistent with each other and with experimental data, which confirms the reliability of the coupled-channels formalism based on the cluster-core configuration in heavy nuclei. The enhanced $E2$ and $E3$ matrix elements in both calculations for $^{222-226}\text{Ra}$ suggest that these three nuclei can exhibit static octupole deformation in both positive- and negative-parity bands. On the other hand, the accurate reproduction of the higher negative-parity bandhead and the derived Q_3 moments in nucleus ^{228}Ra is consistent with the interpretation of this nucleus as a deformed rotor exhibiting dynamic/soft octupole vibrations. We hope that the present work may open new avenues for exploring octupole collectivity and cluster correlations in heavy nuclei.

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I. INTRODUCTION

Nuclear deformation in heavy nuclei, particularly among the light actinides, often exhibits reflection asymmetry associated with strong octupole correlations, giving rise to pear-shaped intrinsic configurations [1,2]. Understanding these octupole effects is crucial for interpreting nuclear spectroscopy, decay modes, and stability. Strong octupole collectivity is found to occur when nucleons near the Fermi surface occupy single-particle orbits with opposite parities differing by $\Delta l = \Delta j = 3\hbar$ between orbital and angular momenta [3,4]. The experimental signatures of octupole correlations are the low-lying 1^- bandhead of the corresponding negative-parity band and the alternating-parity-doublet bands at high spin, which are connected with the pronounced electric dipole and octupole transitions [5–20]. These characteristics were predicted to be significant in some specific mass regions, particularly for the lanthanide Ba isotopes [21,22] and the light actinides of Ra-Th region [14,23], with the proton and

neutron numbers close to 34, 56, 88, and 134. In recent years, direct experimental evidence of $E3$ matrix elements for strong octupole correlations in ^{220}Rn and $^{222,224,228}\text{Ra}$ [24,25] has been observed via Coulomb-excitation experiments. Various theoretical approaches have been employed to investigate the structural characteristics arising from reflection-asymmetric nuclear shapes and octupole collectivity, such as the interacting boson model (IBM) [26–34], macroscopic-microscopic and self-consistent mean-field models [35–39], and phenomenological collective model [40–45]. As for Ra isotopes of concern, one may expect strong static octupole deformation in $^{222-226}\text{Ra}$, while a reduction in octupole strength is anticipated as one moves toward ^{220}Ra and ^{228}Ra , although different theoretical approaches yield diverse predictions [46–49].

In the past decades, the emergence of cluster structures and their correlation with nuclear deformations in light nuclei, together with the interplay between equilibrium shapes, clustering phenomena, and the occurrence of exotic decays in some nuclei, could be assessed [50–54]. This points out the feasibility of interpreting these experimental observations of octupole correlations from the perspective of the cluster degree of freedom [55–57]. In contrast, the enhanced transition rates and the unnatural-parity levels in the light-actinide nuclei appear to be hard to reconcile within the traditional cluster models. An intuitive and physical strategy is to consider the coupled-channels method [54,58–64], which describes the spectroscopic properties through the coupling of two subsystems, naturally a doubly magic core ^{208}Pb plus a cluster. One may expect that different cluster partitions like

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“ ${}^A\text{C} + {}^{208}\text{Pb}$ ” and “ $\alpha + {}^A\text{Rn}$ ”, or their mixture, may be favored under different circumstances. While both scenarios can be studied within the coupled-channels framework, the choice of ${}^{208}\text{Pb}$ is strongly motivated by its doubly magic character and its well-known low-lying octupole (3^-) excitation, which provides a natural mechanism for generating reflection-asymmetric collective modes. The remaining heavier cluster, coupled to a closed-shell core, can be better suited for describing octupole collectivity, as will be demonstrated by the simultaneous reproduction of excitation spectra, electromagnetic-transition strengths, and intrinsic moments achieved in our later calculations. Furthermore, the octupole vibration of the nonrigid core plays a significant role, making this approach suitable for investigating the structural evolution of octupole-deformed nuclei.

We have investigated the so-called unnatural-parity levels for cluster states in ${}^{212}\text{Po}$ and ${}^{216}\text{Rn}$ within the coupled-channels formalism by incorporating core excitation of ${}^{208}\text{Pb}$ to its octupole vibrational 3^- state [65,66] in a recent work [64]. The Ra nuclei can be explained as carbon clusters orbiting the doubly magic core ${}^{208}\text{Pb}$. The configurations of “heavier clusters + ${}^{208}\text{Pb}$ ” in heavy nuclei are consistent with the treatment of other studies [67–76] on cluster states and heavy particle emission. Within our attempt to describe the octupole correlations in terms of α and heavier cluster dynamics, we will provide a complete description of the nuclear properties of the parity-doublet bands as well as the pronounced electromagnetic matrix elements and the octupole correlations of light-actinide nuclei. A new point is that the monopole approximation for the relative motion is taken from the ground-state bands within the binary cluster-core model (BCM) of earlier investigations [67,68] instead of the observed energy levels or other mathematical assumptions. On the other hand, we will also include in the calculations the 2^+ excited state of the core ${}^{208}\text{Pb}$ in order to confirm the reliability of the cluster- ${}^{208}\text{Pb}$ structures in heavy nuclei. The remainder of this article is organized in the following way. In Sec. II, the theoretical formalism and the parametrization are defined. Section III is devoted to numerical results of energy spectra, the matrix elements, and the intrinsic moments for octupole correlations in Ra isotopes. A brief summary is presented in Sec. IV.

II. THEORETICAL FRAMEWORK

As mentioned above, the structural features of the low-lying negative-parity bands and the enhanced transition rates in the light-actinide nuclei cannot be adequately described by the relative motion between the spinless cluster and core within standard models. In order to consider core excitations, one has to include couplings among all open channels in the decay process. In other words, one has to implement a coupled-channels method which may incorporate the coupling of various clusters to the ground and excited states of the residual nucleus (such as, but not limited to, ${}^{208}\text{Pb}$). By accounting for the essential octupole vibration of the nonrigid core, the present approach facilitates a rigorous investigation into the structural evolution of octupole-deformed nuclei. To build up the formalism and explore the heavier cluster states

in our case, the same physical picture of a cluster orbiting the ${}^{208}\text{Pb}$ core as our early work [64] is extended to octupole-deformed Ra nuclei. The corresponding wave function is

$$\Psi_J = \sum_{IL} C_{IL}^J \psi_{ILJ}, \quad (1)$$

where

$$\psi_{ILJ} = \phi(\text{cluster})[\phi_I(\text{Pb})Y_L(\hat{\mathbf{r}})]_J \chi_{IL}^J(r). \quad (2)$$

Here C_{IL}^J is the expansion coefficient, $\phi(\text{cluster})$ is the intrinsic wave function of the cluster, $\chi_{IL}^J(r)$ is the normalized radial wave function of the cluster-core system in the basis ψ_{ILJ} , and I is the spin of the core ${}^{208}\text{Pb}$. L and J are the orbital and total angular momenta, respectively. The Hamiltonian of the relative motion is described as

$$H = T + H_C + V, \quad (3)$$

where T is the kinetic-energy term and H_C is the intrinsic Hamiltonian for the ${}^{208}\text{Pb}$ core and the C cluster. The interaction potential V is given by

$$V = V_0(r) + \sum_{l=2,3} V_l(r)(Y_l(\text{Pb}) \cdot Y_l(\hat{\mathbf{r}})), \quad (4)$$

where the scalar term $V_0(r)$ includes the Coulomb interaction. The scalar product $Y_l(\text{Pb}) \cdot Y_l(\hat{\mathbf{r}})$ with $l = 2, 3$ are in accordance with the quadrupole-quadrupole and octupole-octupole interactions, respectively.

The reduced matrix element of the core ${}^{208}\text{Pb}$ is obtained as follows:

$$\begin{aligned} & \langle \phi_{I'}(\text{Pb}) || Y_l(\text{Pb}) || \phi_I(\text{Pb}) \rangle \\ &= \kappa_I \kappa_{I'} \sqrt{\frac{(2I+1)(2I'+1)}{4\pi}} \langle I0I0 | I'0 \rangle, \end{aligned} \quad (5)$$

where the constant κ_I corresponds to the wave function $\phi_I(\text{Pb})$. The coupling matrix element of the total Hamiltonian for the two-body system in the basis ψ_{ILJ} can be then determined by

$$\begin{aligned} & \langle \psi_{I'L'J'} | H | \psi_{ILJ} \rangle \\ &= E_{IL}^J \delta_{I,I'} \delta_{L,L'} + \sum_{l=2,3} (-1)^{I'+L} \hat{I} \hat{L} \hat{L}' \\ & \times \langle I0I'0 | I0 \rangle \langle L0L'0 | I0 \rangle W(IILJ'; I'L) \beta_{III'}^J, \end{aligned} \quad (6)$$

with

$$E_{IL}^J = \langle \chi_{IL}^J | T_L + V_0 | \chi_{IL}^J \rangle + A_I, \quad (7)$$

$$\beta_{III'}^J = (\kappa_{I'}/4\pi) \langle \chi_{I'L'}^J | V_l | \chi_{IL}^J \rangle. \quad (8)$$

Here $W(abcd; ef)$ is the Racah W-coefficient, and as usual, it is $\hat{I} = \sqrt{2I+1}$. β_l is defined as the radial integral over the noncentral potential strength $V_l(r)$ of Eq. (4) and the relative-motion wave functions. In Eq. (7), the kinetic energy term is $T_L = \langle Y_L(\hat{\mathbf{r}}) | T | Y_L(\hat{\mathbf{r}}) \rangle$, and A_I is the excitation energy of the state ${}^{208}\text{Pb}(I)$. The energy E_{IL}^J , only acting on the diagonal matrix elements, has the form

$$E_{IL}^J = E(L) + A_I, \quad (9)$$

where the energies $E(L)$ are theoretically determined by the equation [54,59,60]

$$\left\{ \frac{-\hbar^2}{2\mu} \left[\frac{d^2}{dr^2} - \frac{L(L+1)}{r^2} \right] + V_{LL'}^J(r) \right\} \chi_{IL}^J(r) = E(L) \chi_{IL}^J(r), \quad (10)$$

with

$$V_{LL'}^J = \langle \psi_{I'L'J} | V_2 + V_3 | \psi_{ILJ} \rangle. \quad (11)$$

Here V_2 and V_3 are the quadrupole-quadrupole and octupole-octupole forms of the noncentral potentials in Eq. (4). Following Refs. [54,59,60,64], the relative-motion part of the total Hamiltonian H is assumed to be decoupled from the core state I^π , implying the energies $E(L)$ generated by the relative motion correspond to the excitation energies of the ground-state band. Different from the input of the observed experimental levels [59,60] or the linear assumption [58], the excitation energies $E(L)$ are taken from the calculations based on the BCM. In this model, the nucleus is described as a two-body system consisting of a cluster and a core. The total Hamiltonian is written as the sum of the Coulomb interaction and a phenomenological nuclear potential, $H = H_C + H_N$. The relative-motion wave functions and excitation energies of the ground-state band are obtained by directly solving the radial Schrödinger equation. These excitation energies serve as the $E(L)$ input in the later coupled-channels calculations that include core excitations and multipole couplings. The nuclear potential is of the form $(1 + \text{Gaussian})(\text{W.S.} + \text{W.S.}^3)$ and is given by

$$V_N(r) = -V_0 \left[1 + \lambda \exp\left(-\frac{r^2}{\sigma^2}\right) \right] \left\{ \frac{b}{1 + \exp[(r-R)/a]} + \frac{1-b}{\{1 + \exp[(r-R)/3a]\}^3} \right\}. \quad (12)$$

The nuclear parameters of Eq. (12) for the even-even $^{222-228}\text{Ra}$ isotopes are consistent with Refs. [64,67,68] except for the depth V_0 , which is determined by the bootstrap method and Eq. (10) of Ref. [67]. It should be noted that the cluster nucleons are confined to states above the Fermi level of the core in order to satisfy the Pauli principle. The cluster orbit is determined by the Wildermuth condition [77], namely, $G = 2N + L$, where N is the number of internal nodes in the wave function, L is the orbital angular momentum, and G is the so-called global quantum number. Therefore, the quantum number G is 68, 76, 84, and 92 for the ground-state bands of ^{222}Ra , ^{224}Ra , ^{226}Ra , and ^{228}Ra , respectively. The $E(L)$ bands with $L^\pi = 0^+, 1^-, 2^+, 3^-, \dots, G^+, (G+1)^-$, can be coupled with the excited state ^{208}Pb ($I^\pi = 3^-$) to generate the new parity-doublet spectra $J^\pi = 0^+, 1^-, 2^+, 3^-, \dots, (G-3)^-, (G-2)^+$ in the Ra isotopes. When the $E(L)$ energies are fixed in the framework of the BCM by directly solving the radial Schrödinger equation of the relative motion, the coupling matrix element of Eq. (6) can be determined from the excitation energy A_I of ^{208}Pb and the coupling strength β_I . According to Refs. [54,58–60,64], the terms A_I and β_I of Calc. I are parametrized for A_0 , A_3 , β_2 , and β_3 in the channel coupling using the 0^+ and 3^- core excitation states plus

quadrupole-quadrupole and octupole-octupole interactions. In this study, the parameter β_2 is fixed as -0.17 MeV in accordance with our previous work [64]. Due to the unique features of octupole correlations in the Ra isotopes, in contrast to ^{212}Po , the residual parameters are obtained by fitting to the experimental levels, namely $\beta_3 = -0.16$ MeV, $A_0 = 0.16$ MeV, and $A_3 = 1.03$ MeV. As for Calc. II with 0^+ , 3^- , and 2^+ core excitations, another dimensional parameter A_2 related to 2^+ state of core is added into the present parameter set, which will be determined in the following procedure.

As mentioned above, the charge distributions and nuclear shapes in heavy nuclei can be subtracted from the dynamic moments of the electromagnetic transitions between different states as well as the static moments reflecting the electric and magnetic properties of individual states. The multipole operator arising from the cluster-core relative motion is proportional to $r^\lambda Y_\lambda(\hat{\mathbf{r}})$ given by

$$\begin{aligned} & \langle \Psi_{J'} || r^\lambda Y_\lambda(\hat{\mathbf{r}}) || \Psi_J \rangle \\ &= \sum_{I'L'} \sum_{IL} C_{I'L'}^{J'} C_{IL}^J \langle I'L'; J' || r^\lambda Y_\lambda(\hat{\mathbf{r}}) || IL; J \rangle, \end{aligned} \quad (13)$$

with the simplified notation

$$|IL; J\rangle = [\phi_I \otimes Y_L]_J \chi_{IL}^J. \quad (14)$$

The reduced matrix elements in Eq. (13) is given by

$$\begin{aligned} & \langle I'L'; J' || r^\lambda Y_\lambda(\hat{\mathbf{r}}) || IL; J \rangle \\ &= \delta_{I,I'} \langle \chi_{I'L'}^{J'} | r^\lambda | \chi_{IL}^J \rangle \langle [\phi_I \otimes Y_L]_{J'} || Y_\lambda(\hat{\mathbf{r}}) || [\phi_I \otimes Y_L]_J \rangle, \end{aligned} \quad (15)$$

where $\delta_{I,I'}$ follows the assumption that the operator $r^\lambda Y_\lambda(\hat{\mathbf{r}})$ does not act on the intrinsic core wave function. By using the identity for a tensor operator acting on the subsystem of a coupled product from Eq. (7.1.8) of Ref. [78], the matrix elements of the operator $Y_\lambda(\hat{\mathbf{r}})$ is

$$\begin{aligned} & \langle [\phi_I \otimes Y_L]_{J'} || Y_\lambda(\hat{\mathbf{r}}) || [\phi_I \otimes Y_L]_J \rangle \\ &= (-1)^{I+L+J'+\lambda} \hat{J} \hat{J}' \times \begin{Bmatrix} L' & J' & I \\ J & L & \lambda \end{Bmatrix} \langle L' || Y_\lambda(\hat{\mathbf{r}}) || L \rangle. \end{aligned} \quad (16)$$

The reduced matrix element of the spherical harmonic tensor is

$$\begin{aligned} \langle L' || Y_\lambda(\hat{\mathbf{r}}) || L \rangle &= (-1)^{L'} \sqrt{\frac{1}{4\pi}} \hat{L}' \hat{L} \begin{pmatrix} L' & \lambda & L \\ 0 & 0 & 0 \end{pmatrix} \\ &= (-1)^L \sqrt{\frac{1}{4\pi}} \hat{L} \hat{L}' \langle L' 0 L' 0 | \lambda 0 \rangle. \end{aligned} \quad (17)$$

Collecting the above terms one obtains

$$\begin{aligned} & \langle I'L'; J' || r^\lambda Y_\lambda(\hat{\mathbf{r}}) || IL; J \rangle \\ &= \delta_{I,I'} \langle \chi_{I'L'}^{J'} | r^\lambda | \chi_{IL}^J \rangle \times (-1)^{I+L+J'+\lambda} \hat{J} \hat{J}' \begin{Bmatrix} I & L & J \\ \lambda & J' & L' \end{Bmatrix} \\ &\quad \times (-1)^L \sqrt{\frac{1}{4\pi}} \hat{L} \hat{L}' \langle L' 0 L' 0 | \lambda 0 \rangle, \\ &= \delta_{I,I'} (-1)^L \sqrt{\frac{1}{4\pi}} \hat{J} \hat{J}' \hat{L} \hat{L}' \\ &\quad \times W(ILJ'; JL') \langle L' 0 L' 0 | \lambda 0 \rangle \langle \chi_{I'L'}^{J'} | r^\lambda | \chi_{IL}^J \rangle, \end{aligned} \quad (18)$$

where we have rearranged the orders of the arguments in the 6- j symbol for convenience. One thus gets

$$\begin{aligned} & \langle \Psi_{J'} || r^\lambda Y_\lambda(\hat{r}) || \Psi_J \rangle \\ &= \sum_{I'L'} \sum_{IL} \delta_{I,L'} (-1)^L C_{IL}^J C_{I'L'}^{J'} \gamma_\lambda \sqrt{\frac{1}{4\pi}} \hat{J} \hat{J}' \hat{L} \hat{L}' \\ & \times W(ILJ'\lambda; JL') \langle L0L'0 | \lambda 0 \rangle \langle \chi_{I'L'}^{J'} | r^\lambda | \chi_{IL}^J \rangle, \end{aligned} \quad (19)$$

with the additional factor from the relative motion given by

$$\gamma_\lambda = \frac{Z_1 M_2^\lambda + (-1)^\lambda Z_2 M_1^\lambda}{(M_1 + M_2)^\lambda}. \quad (20)$$

Both the 0^+ ground state and 3^- excited state of the core ^{208}Pb are considered in this study, leading to the matrix element divided into two factors, i.e., $D_0^\lambda = \langle \chi_{0L}^{J'} | r^\lambda | \chi_{0L}^J \rangle$ and $D_3^\lambda = \langle \chi_{3L}^{J'} | r^\lambda | \chi_{3L}^J \rangle$. In this way, the multipole operator of electromagnetic transitions in Eq. (19) can be denoted by $\langle \Psi_{J'} || r^\lambda Y_\lambda(\hat{r}) || \Psi_J \rangle = a_I D_I^\lambda$, where a_0 and a_3 , related to the contributions of $I = 0$ and $I = 3$ terms, can be calculated from Eq. (19). Our expression for the total reduced matrix element differs from that of Refs. [54,62,64] by the scaling factors and that in Ref. [58] by the phase convention. Z_1 , Z_2 and M_1 , M_2 represent the core and cluster charge and mass numbers, respectively. λ is the multipolarity of transitions, and the quantities C_{IL}^J and $C_{I'L'}^{J'}$ are separately the expansion coefficients of initial state J_i and final state J_f . The $E\lambda$ transition strength for the yrast band is then specified by

$$B(E\lambda; J_i \rightarrow J_f) = e^2 |a_0 D_0^\lambda + a_3 D_3^\lambda|^2. \quad (21)$$

In some cases, additional experimental information for electromagnetic transitions in the light-actinide region is obtained from the Coulomb-excitation measurements [24,25,79] and mostly presented in the form of the reduced matrix elements $\langle J_f || M(E\lambda) || J_i \rangle$ instead of the $B(E\lambda)$ transition rates. The connection between these two quantities [69,80,81] is

$$\langle J_f || M(E\lambda) || J_i \rangle = \sqrt{B(E\lambda; J_i \rightarrow J_f)} (2J_i + 1). \quad (22)$$

III. NUMERICAL RESULTS AND DISCUSSION

The coupling matrix elements of the relative motion depend on the $E(L)$ term and other free parameters. The $E(L)$ energies for the even-even $^{222-228}\text{Ra}$ isotopes, taken from the calculated ground-state bands within the BCM, are presented in Figs. 1 and 2. Both positive- and negative-parity bands for these four nuclei are satisfactorily reproduced with a slight upward shift relative to the available data, except for the positive-parity levels in ^{228}Ra . The structural hallmarks of the nuclear-reflection asymmetry in the light-actinide region, including the low-lying negative-parity band and the alternating parity at high spin, are interpreted well within the BCM. The standard cluster model [82] underestimates the $B(E2 \downarrow)$ and $B(E3 \downarrow)$ values. This is because the surface peaking of the radial wave function is underestimated within that model. This drawback is compensated by using a suitable effective charge [80,81,83]. An intuitive and effective method to reproduce the energy spectra and $E2$, $E3$ transition rates is the coupled-channels formalism with multipole interactions. The heavy Ra

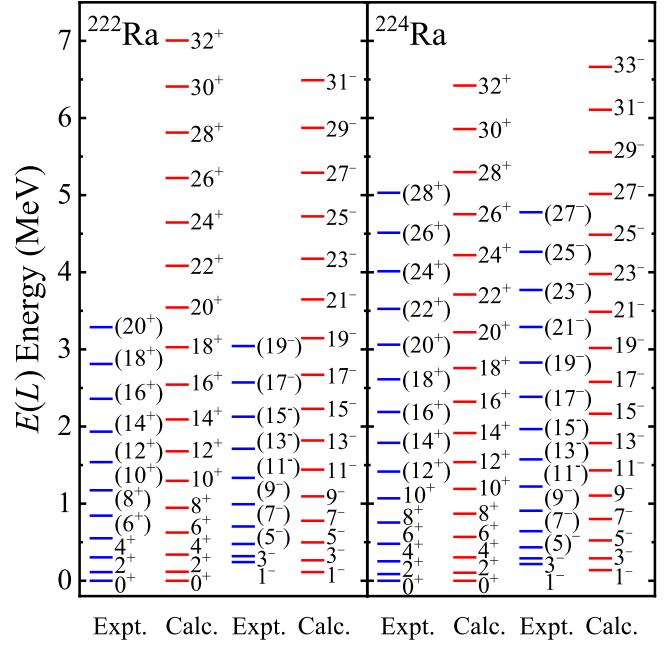


FIG. 1. Comparison of BCM ground-state bands [the $E(L)$ terms in Eq. (9)] with experimental data [84,85] for ^{222}Ra ($^{14}\text{C} + ^{208}\text{Po}$) and ^{224}Ra ($^{16}\text{C} + ^{208}\text{Po}$).

isotopes can be modeled as an intuitive physical scenario, in which the carbon cluster orbits around the core ^{208}Pb that is allowed to occupy its lowest excited state, i.e., the 3^- state. Therefore, after the explicit values are assigned to other free parameters, the parity-doublet bands for octupole collectivity from ^{222}Ra to ^{228}Ra are subsequently determined, as is shown in Fig. 3. The available data are indicated by the blue lines,

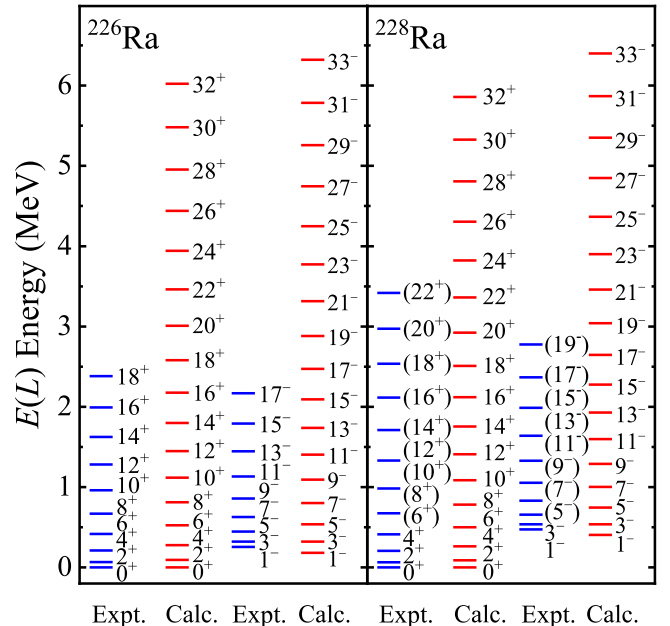


FIG. 2. Same as Fig. 1 but for ^{226}Ra ($^{18}\text{C} + ^{208}\text{Po}$) and ^{228}Ra ($^{20}\text{C} + ^{208}\text{Po}$). The experimental data are taken from Refs. [86,87].

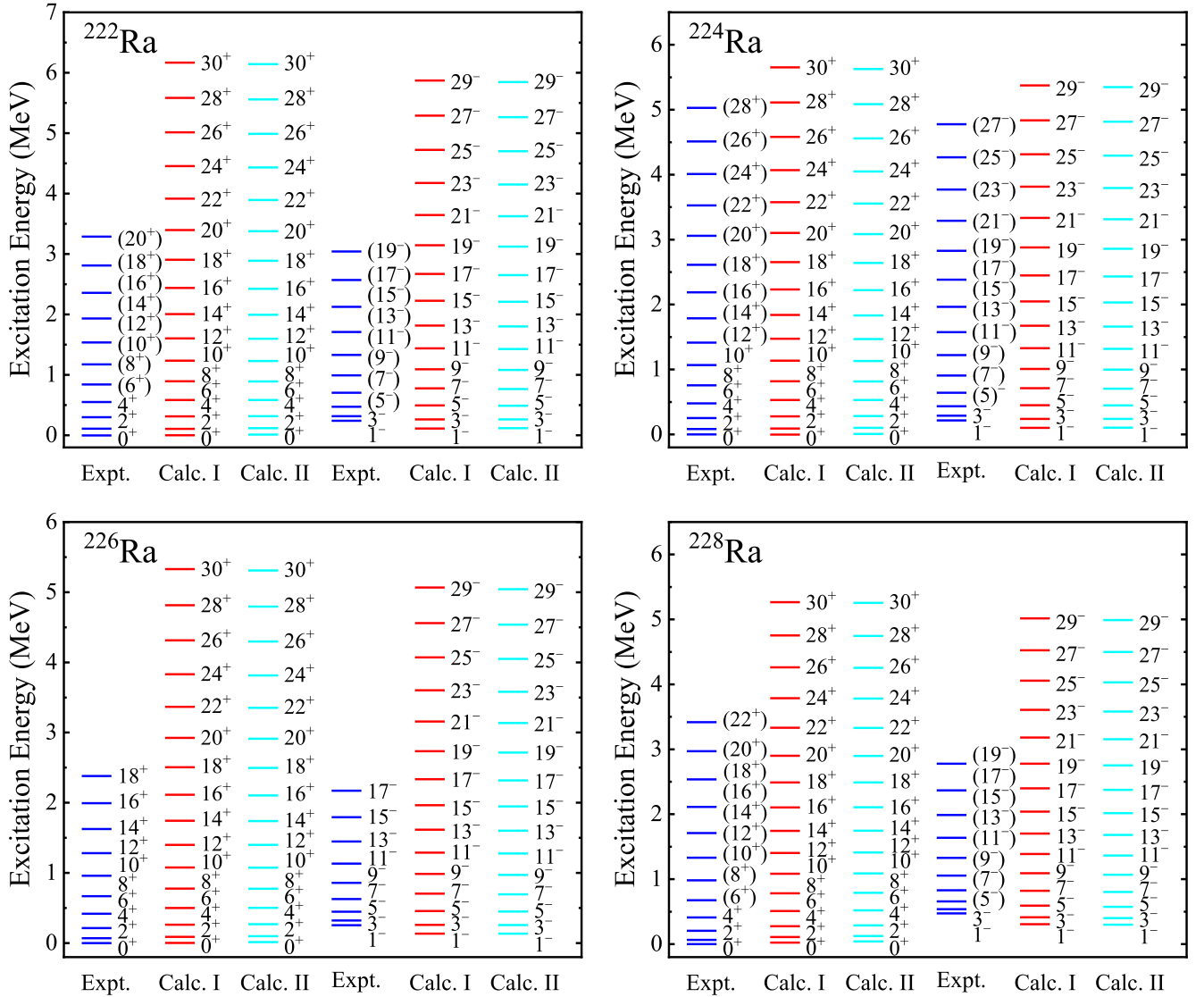


FIG. 3. Comparison of calculated ground-state bands within the coupled-channels method plus core excitation with experimental data [84–87] for ^{222}Ra ($^{14}\text{C} + ^{208}\text{Pb}$), ^{224}Ra ($^{16}\text{C} + ^{208}\text{Pb}$), ^{226}Ra ($^{18}\text{C} + ^{208}\text{Pb}$), and ^{228}Ra ($^{20}\text{C} + ^{208}\text{Pb}$). The theoretical framework for the energy levels of Calc. I in red lines contains the ground state 0^+ and excited state 3^- of core ^{208}Pb . The core excitation 2^+ is added in the description of the energy levels of Calc. II in cyan lines.

and the theoretical energy spectra of Calc. I connected with both $I = 0$ and $I = 3$ are represented by the cyan lines. All the known positive- and negative-parity states are well reproduced, which imply the existence of spectroscopic features of octupole deformation and vibrations in these pear-shaped nuclei. A number of high-lying energy levels are presented in Fig. 3, which have to be confirmed by future experimental efforts.

Inspired by these results, we expanded our calculations by adding the excited core state 2^+ to the core states 0^+ and 3^- . With this, we intend to further confirm the reliability of the coupled-channels formalism and the suitability of introducing the configuration of “heavier cluster + ^{208}Pb ” in heavy nuclei. However, by doing this, the number of parameters A_I and β_I increases from four to five, due to the introduction of the excited state 2^+ , which implies the introduction of the

parameter A_2 . We found that the calculated levels of Calc. II containing the core states 0^+ , 3^- , and 2^+ , with the same set of parameters, systematically underestimate the corresponding experimental values. To reduce the number of parameters, only the parameter A_0 is adjusted in Calc. II with other parameters fixed. Performing a least-square fit, we found that the values of A_0 and A_2 are, respectively, 0.29 MeV and 2.18 MeV. In Fig. 3, the results of Calc. II, which includes the core excited states 3^- and 2^+ , are shown by cyan lines. One sees that a good agreement of the calculated levels with the measured data is achieved. This shows that the bandheads of the negative-parity bands in $^{222}\text{--}^{226}\text{Ra}$, lying very close to the 0^+ states, correspond to the stable octupole deformation, while the nucleus ^{228}Ra corresponds to an octupole vibrator [25,88]. This confirms the predictions of Refs. [10,23,37] that the boundary of the octupole-deformed region in heavy nuclei

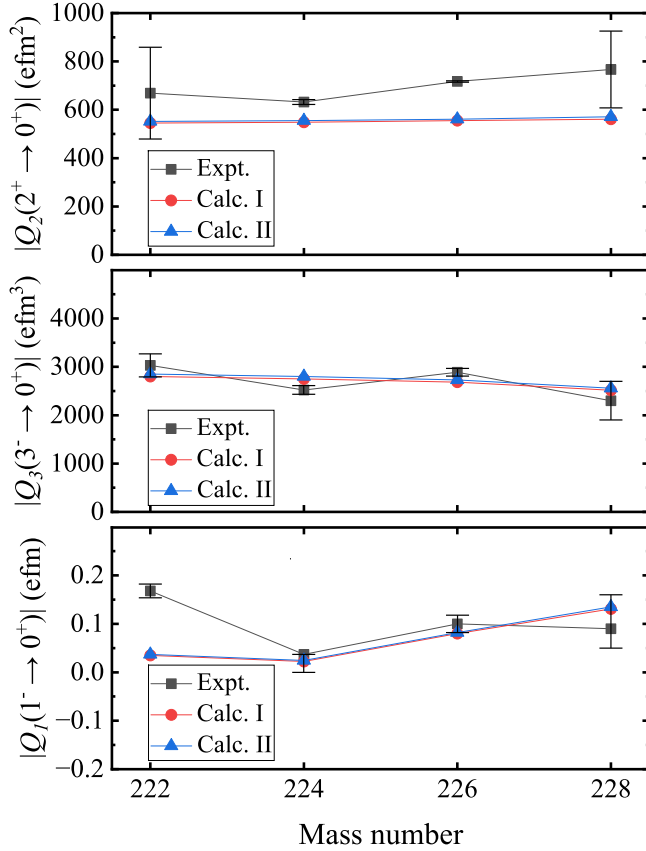


FIG. 4. Comparison of calculated intrinsic moments Q_λ in absolute-value form with experimental data [24,25,79,84,87] for $^{222-228}\text{Ra}$.

is located around $Z = 88$ and $N = 138$. Moreover, the error per known level δ_{BCM} for $^{222-228}\text{Ra}$ in Figs. 1 and 2 using the BCM is 0.143 MeV. The average errors of Calc. I and Calc. II on the energy levels are, separately, 0.071 MeV and 0.065 MeV. Thus, a great improvement of the parity-doublet bands in the Ra isotopes is obtained within the coupled-channels method plus core excitation with multipole interactions beyond the binary cluster-core model. We hope that the present work on the reflection asymmetric properties provides a test ground for investigating the octupole degrees of freedom in light actinides.

The observed spectra indicate a pronounced population of the positive-parity ground-state band through direct and multiple quadrupole Coulomb excitations, as well as a considerable population of the negative-parity octupole band resulting from $E3$ excitation [24,25]. Therefore, the reflection asymmetry with octupole deformation and its correlation with cluster structures can be detected by the highly collective transition rates. In general, the experimental information on electromagnetic transitions in light actinides is derived from the Coulomb-excitation measurements [24,25,79] in terms of matrix elements $\langle J_f || M(E\lambda) || J_i \rangle$ rather than $B(E\lambda)$ probabilities. To evaluate them, the factors D_I^λ in Eq. (15) with $I = 0, 3$, and 2 are given in the following forms: $D_I^2 = 3/5R_I^2$ for $E2$ transition, $D_I^3 = 1/2R_I^3$ for $E3$ transition, and $D_I^1 = 3/4R_I$ for $E1$ transition, where the radius R_I is supposed to be

proportional to $A^{1/3}$ of the nucleus [73,89]. The parameters radii are determined by the restriction of $R_I \propto A^{1/3}$ and fitting to the available data of $E2$ and $E3$ matrix elements.

Following this, the calculated results of $E2$ and $E3$ transitions for Ra isotopes with no effective charges in the framework of coupled-channels method by allowing the core excitation are displayed in Table I. Highly collective $E2$ and $E3$ matrix elements for nuclei ^{222}Ra , ^{224}Ra , ^{226}Ra , and ^{228}Ra are well reproduced in both Calc. I and Calc. II, which provide compelling evidence that these nuclei have strong octupole correlations. A trend is found that the calculated values of $E2$ matrix elements increase with increasing angular momentum. The theoretical values for $\langle J_f || M(E2) || J_i \rangle$ and $\langle J_f || M(E3) || J_i \rangle$ of Calc. I are generally smaller than the case of Calc. II, which introduces another 2^+ core excitation to the calculations. Additionally, the $B(E\lambda \uparrow; 0^+ \rightarrow \lambda)$ strengths can be determined from $B(E\lambda \downarrow; \lambda \rightarrow 0^+)$ values by multiplying a specific factor, $(2\lambda + 1)$ [80]. The calculated results for ^{226}Ra are then given here, namely, $B(E2 \uparrow; 0^+ \rightarrow 2^+)_I = 3.05 \text{ e}^2\text{b}^2$, $B(E3 \uparrow; 0^+ \rightarrow 3^-)_I = 1.00 \text{ e}^2\text{b}^3$ for Calc. I, and $B(E2 \uparrow; 0^+ \rightarrow 2^+)_II = 3.12 \text{ e}^2\text{b}^2$, $B(E3 \uparrow; 0^+ \rightarrow 3^-)_II = 1.04 \text{ e}^2\text{b}^3$ for Calc. II, which agree well with the measured data [80], $5.15 \pm 0.14 \text{ e}^2\text{b}^2$ for $E2$ transitions and $1.10 \pm 0.11 \text{ e}^2\text{b}^3$ for $E3$ transitions. As for ^{228}Ra , an excellent agreement of our results with experimental data is given in Table I, except for the matrix elements of $E2$ transition $2^+ \rightarrow 2^+$ and $E3$ transition $3^- \rightarrow 2^+$, which are associated with the dynamic vibrational feature.

To determine the nuclear shape, the intrinsic deformation, though not directly observable, can be deduced from its correlation with the electric moments, which are generated by the nonspherical charge distribution. The electric $E2$ transition moments, corresponding to the matrix elements between differing members of rotational bands, are the key experimental observables for quadrupole-deformed nuclei. In the case of pear-shaped nuclei with reflection asymmetry, there are additional $E1$ and $E3$ transition moments between the opposite parities related to rotational levels. The enhanced $E3$ transitions are a manifestation of the collective behaviors, leading to the pear-shaped distortion. Consequently, the nonzero $E3$ matrix elements are the direct evidence of the octupole correlations in the Ra isotopes. Although the $E1$ transition strength is influenced by the single-particle and cancellation effects [24], it can be regarded as a probe to detect the properties of reflection asymmetry. In order to reconfirm the reliability and integrity of the coupled-channels formalism plus core excitation and the cluster-core structures in heavy nuclei, the calculations of $E1$ matrix elements are simultaneously performed based on Calc. I and Calc. II in the following procedure. The dipole matrix elements cannot be described well in the same theoretical framework without additional effective charges. This may be due to the fact that the dipole moments arise from the asymmetrical charge distribution and can be pronounced by the separation of the mass center and charge center [24]. The effective charge $e_1 = 0.01$ of dipole transitions is taken into the calculations for $^{222-228}\text{Ra}$. The $\langle J_f || M(E1) || J_i \rangle$ values obtained by Calc. I and Calc. II in Table II are consistent with the experimental data. Combined with Table I, the good results of $\langle J_f || M(E\lambda) || J_i \rangle$ with $\lambda = 1$,

TABLE I. Comparison of the theoretical reduced matrix elements with available data [24,25,79] in absolute-value form for $E2$ and $E3$ transitions in $^{222-228}\text{Ra}$. The calculations of $E2$ and $E3$ matrix elements are performed using bare charges, without introducing any effective charges. The Calc. I represents the coupled-channels formalism with core excitation of 3^- , and the Calc. II is performed by allowing the core excitations of both 3^- and 2^+ .

$ \langle J_f M(E\lambda) J_i \rangle $	$^{222}\text{Ra} \text{ (efm}^\lambda)$			$^{224}\text{Ra} \text{ (efm}^\lambda)$			$^{226}\text{Ra} \text{ (efm}^\lambda)$			$^{228}\text{Ra} \text{ (efm}^\lambda)$		
	Ref. [25]	Calc. I	Calc. II	Ref. [24]	Calc. I	Calc. II	Ref. [79]	Calc. I	Calc. II	Ref. [25]	Calc. I	Calc. II
$ \langle 0^+ M(E2) 2^+ \rangle $	211 ± 60	172	174	199 ± 3	173	175	226 ± 1	175	177	242 ± 50	177	180
$ \langle 2^+ M(E2) 4^+ \rangle $	298 ± 15	368	372	315 ± 6	370	375	370^{+9}_{-5}	374	379	387 ± 19	381	387
$ \langle 4^+ M(E2) 6^+ \rangle $	357 ± 18	555	561	405 ± 15	559	565	483^{+6}_{-7}	565	572	511 ± 26	577	585
$ \langle 6^+ M(E2) 8^+ \rangle $	415 ± 23	739	747	500 ± 60	744	753	541^{+7}_{-10}	754	763	589 ± 29	770	781
$ \langle 8^+ M(E2) 10^+ \rangle $	470 ± 50	921	931		929	939	591^{+24}_{-11}	941	952	750 ± 40	942	976
$ \langle 10^+ M(E2) 12^+ \rangle $		1101	1111		1111	1122	900^{+21}_{-37}	1127	1139		1154	1170
$ \langle 12^+ M(E2) 14^+ \rangle $		1277	1289		1291	1304	732^{+31}_{-22}	1311	1325		1345	1363
$ \langle 14^+ M(E2) 16^+ \rangle $		1448	1462		1468	1483	821^{+190}_{-62}	1494	1510		1536	1556
$ \langle 16^+ M(E2) 18^+ \rangle $		1615	1631		1641	1657	559^{+75}_{-59}	1673	1691		1725	1748
$ \langle 2^+ M(E2) 2^+ \rangle $	130 ± 50	205	208		206	209	147^{+29}_{-32}	208	211	30 ± 170	212	215
$ \langle 4^+ M(E2) 4^+ \rangle $	280 ± 50	350	354		352	356	338^{+32}_{-47}	356	361		363	368
$ \langle 6^+ M(E2) 6^+ \rangle $		498	503		501	507	577^{+75}_{-37}	498	502		518	525
$ \langle 8^+ M(E2) 8^+ \rangle $		645	652		650	657	565^{+27}_{-136}	676	646		674	683
$ \langle 10^+ M(E2) 10^+ \rangle $		792	801		799	808	487^{+50}_{-144}	810	819		829	841
$ \langle 12^+ M(E2) 12^+ \rangle $		937	946		947	956	515^{+180}_{-150}	961	971		985	998
$ \langle 1^- M(E2) 3^- \rangle $	235 ± 22	269	272	230 ± 11	271	274	366^{+6}_{-12}	272	275	380 ± 50	269	271
$ \langle 3^- M(E2) 5^- \rangle $	310 ± 40	450	453	410 ± 60	459	463	409^{+5}_{-10}	461	465	390^{+40}_{-80}	453	457
$ \langle 5^- M(E2) 7^- \rangle $	440 ± 40	636	642		642	648	407^{+3}_{-7}	641	646	400 ± 90	631	637
$ \langle 7^- M(E2) 9^- \rangle $	600 ± 100	815	822		823	830	545^{+3}_{-17}	826	833	590 ± 100	807	814
$ \langle 9^- M(E2) 11^- \rangle $		993	1001		1003	1012	677^{+30}_{-70}	1007	1016		981	990
$ \langle 11^- M(E2) 13^- \rangle $		1167	1178		1182	1193	1000^{+110}_{-70}	1186	1197		1155	1166
$ \langle 13^- M(E2) 15^- \rangle $		1339	1351		1358	1370	970^{+300}_{-220}	1364	1377		1327	1340
$ \langle 1^- M(E2) 1^- \rangle $		145	146		145	147	415^{+35}_{-25}	146	147		145	146
$ \langle 3^- M(E2) 3^- \rangle $		273	276		275	278	452^{+60}_{-110}	276	279		272	274
$ \langle 5^- M(E2) 5^- \rangle $		417	420		420	424	482^{+44}_{-61}	422	426		414	418
$ \langle 7^- M(E2) 7^- \rangle $		549	552		566	571	595^{+148}_{-71}	568	573		555	560
$ \langle 9^- M(E2) 9^- \rangle $		705	698		712	706	677^{+114}_{-145}	697	688		696	691
$ \langle 0^+ M(E3) 3^- \rangle $	1130 ± 90	1045	1064	940 ± 30	1026	1045	1080 ± 30	1001	1019	870 ± 150	939	955
$ \langle 2^+ M(E3) 1^- \rangle $	850 ± 240	783	798	1370 ± 140	767	782	1190^{+30}_{-70}	749	764	1360 ± 230	714	697
$ \langle 2^+ M(E3) 3^- \rangle $	900 ± 500	1206	1228	< 4000	1184	1206	1150 ± 160	1156	1178	60^{+160}_{-230}	1087	1105
$ \langle 2^+ M(E3) 5^- \rangle $	1790 ± 200	1995	2030	1410 ± 190	1964	1999	2000 ± 70	2064	1656	1710 ± 230	1765	1796
$ \langle 4^+ M(E3) 1^- \rangle $	2100 ± 500	902	919		884	901	1470^{+70}_{-160}	865	882	400^{+700}_{-1100}	827	842
$ \langle 4^+ M(E3) 3^- \rangle $	2600^{+600}_{-900}	1335	1359		1311	1335	1450^{+320}_{-140}	1281	1305		1209	1230
$ \langle 4^+ M(E3) 5^- \rangle $	1700 ± 1000	1818	1712		1789	1822	< 1700	1746	1778		1616	1645
$ \langle 4^+ M(E3) 7^- \rangle $	3300^{+300}_{-500}	2865	2918		2823	2875	2450^{+70}_{-140}	2748	2799		2492	2543
$ \langle 6^+ M(E3) 3^- \rangle $		1815	1850		1782	1817	2600 ± 500	1745	1779		1653	1685
$ \langle 6^+ M(E3) 5^- \rangle $		1973	2009		1942	1978	2000 ± 300	1898	1933		1764	1798
$ \langle 6^+ M(E3) 7^- \rangle $		2453	2496		2416	2460	2400^{+400}_{-1400}	2355	2399		2218	2191
$ \langle 6^+ M(E3) 9^- \rangle $		3718	3787		3665	3734	2900^{+100}_{-200}	3566	3634		3195	3271
$ \langle 8^+ M(E3) 5^- \rangle $		2684	2734		2640	2690	< 2400	2583	2633		2411	2460
$ \langle 8^+ M(E3) 11^- \rangle $		4555	4640		4498	4583	2200^{+600}_{-1700}	4377	4461		3896	3998

2, and 3 verify the persistence of strong octupole correlations and heavier clusters constituted by carbon isotopes in the light-actinide nuclei. A natural extension in our future study is to investigate the 3α correlations outside the Pb isotopes in octupole-deformed Ra nuclei.

Encouraged by the above results, the present coupled-channels method plus core excitation is generalized to interpret the intrinsic moments Q_λ , which is closely related to the nuclear shapes and the values of quadrupole and octupole

deformation. The intrinsic moments [25], extracted from the matrix elements, can be determined by

$$Q_\lambda = \frac{\langle J_f || M(E\lambda) || J_i \rangle}{\sqrt{(2J_f + 1)(2\lambda + 1)/16\pi} \langle J_f 0 \lambda 0 | J_i \rangle} \quad (23)$$

with $\lambda = 2$ and 3 for E_2 and E_3 transitions, respectively, and

$$Q_1 = \frac{\langle J_f || M(E1) || J_i \rangle}{\sqrt{3(2J_f + 1)/4\pi} \langle J_f 0 1 0 | J_i \rangle} \quad (24)$$

TABLE II. Same as Table I, but for $E1$ matrix elements. The effective charge $e_1 = 0.01$ is employed in the calculations of $\langle J_f || M(E1) || J_i \rangle$.

$ \langle J_f M(E1) J_i \rangle $	^{222}Ra (efm $^\lambda$)			^{224}Ra (efm $^\lambda$)			^{226}Ra (efm $^\lambda$)			^{228}Ra (efm $^\lambda$)		
	Ref. [25]	Calc. I	Calc. II	Ref. [24]	Calc. I	Calc. II	Ref. [79]	Calc. I	Calc. II	Ref. [25]	Calc. I	Calc. II
$ \langle 0^+ M(E1) 1^- \rangle $	0.082 ± 0.007	0.017	0.018	< 0.018	0.011	0.012	0.050 ± 0.009	0.039	0.040	$0.043^{+0.032}_{-0.020}$	0.064	0.066
$ \langle 1^- M(E1) 2^+ \rangle $		0.031	0.033		0.020	0.021	0.068 ± 0.010	0.070	0.074		0.117	0.121
$ \langle 2^+ M(E1) 3^- \rangle $	0.26 ± 0.05	0.045	0.047	0.026 ± 0.005	0.029	0.030	0.061 ± 0.007	0.101	0.105	0.09 ± 0.04	0.167	0.172
$ \langle 3^- M(E1) 4^+ \rangle $		0.059	0.062		0.038	0.039	0.057 ± 0.005	0.133	0.138		0.220	0.226
$ \langle 4^+ M(E1) 5^- \rangle $	$0.18^{+0.048}_{-0.019}$	0.073	0.076	0.030 ± 0.010	0.046	0.048	0.093 ± 0.007	0.163	0.169	0.13 ± 0.06	0.264	0.273
$ \langle 5^- M(E1) 6^+ \rangle $	0.277 ± 0.011	0.087	0.091		0.055	0.057		0.195	0.202	$0.074^{+0.018}_{-0.012}$	0.317	0.328
$ \langle 6^+ M(E1) 7^- \rangle $	0.273 ± 0.021	0.100	0.104	< 0.10	0.064	0.066	0.157 ± 0.010	0.224	0.233		0.358	0.371
$ \langle 7^- M(E1) 8^+ \rangle $	0.317 ± 0.018	0.114	0.119		0.073	0.076	0.21 ± 0.08	0.256	0.266	$0.13^{+0.10}_{-0.33}$	0.410	0.426
$ \langle 8^+ M(E1) 9^- \rangle $	0.48 ± 0.04	0.127	0.133		0.081	0.085	0.24 ± 0.02	0.286	0.297	$0.20^{+0.20}_{-0.09}$	0.450	0.469
$ \langle 9^- M(E1) 10^+ \rangle $	0.40 ± 0.05	0.141	0.148		0.090	0.094	0.27 ± 0.05	0.317	0.330		0.505	0.523
$ \langle 10^+ M(E1) 11^- \rangle $	0.50 ± 0.06	0.154	0.162		0.099	0.103	0.33 ± 0.04	0.347	0.361		0.542	0.567
$ \langle 11^- M(E1) 12^+ \rangle $	0.31 ± 0.04	0.169	0.177		0.108	0.112	0.35 ± 0.08	0.378	0.394		0.594	0.621
$ \langle 12^+ M(E1) 13^- \rangle $		0.181	0.191		0.116	0.121	0.63 ± 0.08	0.408	0.425		0.634	0.665
$ \langle 13^- M(E1) 14^+ \rangle $		0.196	0.206		0.125	0.131	$0.65^{+0.15}_{-0.05}$	0.439	0.458		0.686	0.720
$ \langle 14^+ M(E1) 15^- \rangle $		0.208	0.219		0.133	0.140		0.468	0.489		0.725	0.762
$ \langle 15^- M(E1) 16^+ \rangle $		0.222	0.235		0.142	0.149	$0.55^{+0.07}_{-0.14}$	0.500	0.523		0.777	0.818
$ \langle 2^+ M(E1) 1^- \rangle $	$0.114^{+0.025}_{-0.009}$	0.024	0.025	< 0.03	0.015	0.016		0.055	0.056	$0.051^{+0.033}_{-0.022}$	0.091	0.093
$ \langle 4^+ M(E1) 3^- \rangle $	0.27 ± 0.26	0.052	0.054		0.033	0.034		0.117	0.121	$0.080^{+0.036}_{-0.023}$	0.194	0.200

for dipole intrinsic moments. Comparison of the theoretical $Q_\lambda(\lambda \rightarrow 0^+)$ ($\lambda = 1, 2$, and 3) values with available data [24,25,79,84,87] is shown in Fig. 4, where the good agreement can be achieved except for Q_1 values of ^{222}Ra and ^{228}Ra with small discrepancies from measured data. This might be due to the fact that $E1$ transitions strongly depend on the effective charges. For example, a sudden decrease of experimental values for Q_1 intrinsic moments occurs from ^{222}Ra to ^{224}Ra , implying a complicated change of structures in the Ra isotopic chain. This effect could be effectively accounted for by varying the effective charge, as discussed in Ref. [32]. Despite these minor discrepancies, the combined agreement of the calculated and observed energy spectra, electromagnetic matrix elements, and intrinsic moments across all isotopes confirms the predictive power of the coupled-channels approach. The similarity between results from Calc. I (core 3^- excitation) and Calc. II (core excited 3^- and 2^+ excitations) further validates the robustness of this framework in describing both static and dynamic octupole collectivity in heavy nuclei. These results underscore the importance of incorporating cluster-core interactions and multipole couplings when modeling reflection-asymmetric structures in the light-actinide region.

IV. CONCLUSION

In this work, we have systematically investigated the spectroscopic properties and octupole correlations of even-even $^{222-228}\text{Ra}$ isotopes within the coupled-channels formalism, incorporating explicit cluster degrees of freedom. By modeling the Ra isotopes as composite systems of a ^{208}Pb core and a carbon cluster, we introduced multipole interactions that allow the core to be excited to its 3^- and 2^+ states. This approach enables a unified description of energy spectra, transition-matrix elements, and intrinsic moments in reflection-asymmetric nuclei, which have been reproduced with good accuracy. The

close agreement between Calculations I and II confirms the reliability and robustness of the coupled-channels method in describing octupole collectivity. The enhanced $E3$ matrix elements and low-lying energy levels with negative parities calculated in $^{222-226}\text{Ra}$ indicate strong and stable octupole deformation. In contrast, the higher 1^- bandhead in ^{228}Ra , by about 0.2 MeV relative to the other nuclei, suggests that this nucleus exhibits a transition toward a dynamic octupole vibrational mode. This finding supports previous theoretical predictions that the boundary between static and dynamic octupole deformation in heavy nuclei lies near $Z = 88$ and $N = 138$. Overall, the present study demonstrates that the coupled-channels model, augmented with cluster degrees of freedom and core excitations, provides a powerful and physically intuitive framework for exploring reflection-asymmetric shapes and octupole correlations in heavy nuclei. These efforts are expected to further clarify the role of clustering and octupole collectivity in shaping the structural evolution of the light-actinide region.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not

technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of

this research project. The data are available from the authors upon reasonable request.

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