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Reduction in Nuclear Size and Quadrupole Deformation of High-Spin Isomers of $^{127,129}\text{In}$

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We employed laser spectroscopy of atomic transitions to measure the nuclear charge radii and electromagnetic properties of the high-spin isomeric states in neutron-rich indium isotopes ($Z = 49$) near the closed proton and neutron shells at $Z = 50$ and $N = 82$. Our data reveal a reduction in the nuclear charge radius and intrinsic quadrupole moment when protons and neutrons are fully aligned in $^{129}\text{In}(N = 80)$, to form the high spin isomer. Such a reduction is not observed in $^{127}\text{In}(N = 78)$, where more complex configurations can be formed by the existence of four neutron holes. These observations are not consistently described by nuclear theory.

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Introduction—Excited states of atomic nuclei most commonly occur with extremely short lifetimes, typically ranging from less than a picosecond (10^{-12} s) to a few nanoseconds (10^{-9} s) [1]. However, in some nuclei, the protons and neutrons can be reorganized to form configurations with exceptionally large differences in shape or angular momentum. These states can increase in their lifetime due to quantum selection rules that greatly inhibit their de-excitation pathways [1–4], giving rise to long-lived states—nuclear isomers. Isomers formed by three or more

unpaired nucleons can create nuclear states with unusually large values of angular momentum (high nuclear spin, I), commonly known as multiquasiparticle isomers [1]. These isomers can exhibit lifetimes that are comparable to or even longer than those of their respective ground states (g.s.) [5–9]. Until now, there are only a handful of high-spin, multiquasiparticle isomers for which properties such as the charge radii and electromagnetic moments have been measured simultaneously. Available data exists only for some heavy nuclei, e.g., ^{97}Y , ^{130}Cs , ^{130}Ba , ^{177}Lu , and ^{178}Hf [10–13]. All of these isomers were found to have smaller charge radii than their g.s. while exhibiting a similar or larger quadrupole moment. Accurate calculations of these isomers are currently beyond the reach of many microscopic calculations, as they exhibit complex configurations [13–17]. This calls for measurements of high-spin isomers in the vicinity of closed-shell nuclei that are expected to have simpler configurations accessible to complementary nuclear models [13,18]. In these systems, pairing correlations may play a lesser role, and thus, the standard hypothesis that links the isomer shift to the pairing-blocking effect [1] can be critically evaluated and tested. Such isomers exist in the neutron-rich indium isotopes ($Z = 49$), near the doubly-magic ^{132}Sn ($Z = 50$, $N = 82$), which recently became accessible for high-precision laser spectroscopy studies [19]. These recent measurements of the ground-state properties, including charge radii, for the indium isotopes have established the credibility of the currently available theoretical calculations [19,20].

In this Letter, we report measurements of the changes in mean-squared charge radii and electromagnetic moments of the high-spin isomers (nuclear spin suggested to be $I \geq 21/2$) of neutron-rich indium isotopes. In the shell-model picture, the g.s. of the odd-even indium isotopes is dominated by a hole in the $Z = 50$ proton closed shell ($g_{9/2}$ proton orbit), resulting in a nuclear spin $I^\pi = 9/2^+$. This is the case for all known odd-even isotopes [19]. However, beta-decay spectroscopy studies revealed the existence of high-spin, multiquasiparticle isomers, $I^\pi = (21/2)^-$ and $I^\pi = (23/2)^-$, in ^{127}In ($N = 78$) and ^{129}In ($N = 80$), respectively [21,22]. These isomers are suggested to be formed by the breaking of a neutron pair to create single holes in the $d_{3/2}$ and $h_{11/2}$ neutron orbits, interacting with a proton hole in the $g_{9/2}$ orbit [21]. A schematic diagram of the proton and neutron configuration of these isomers is shown in Fig. 1(a). The nuclear spin $23/2$ is obtained by fully aligning the spins of the 3 hole states in the ^{132}Sn core, adding up to $I = 23/2$. This maximally aligned, or optimal, structure of the isomer in ^{129}In , presents a simple configuration to guide our understanding of these nuclei. In ^{127}In , a different spin assignment, $21/2$, has been suggested in the literature [21,22], which corresponds to a configuration where not

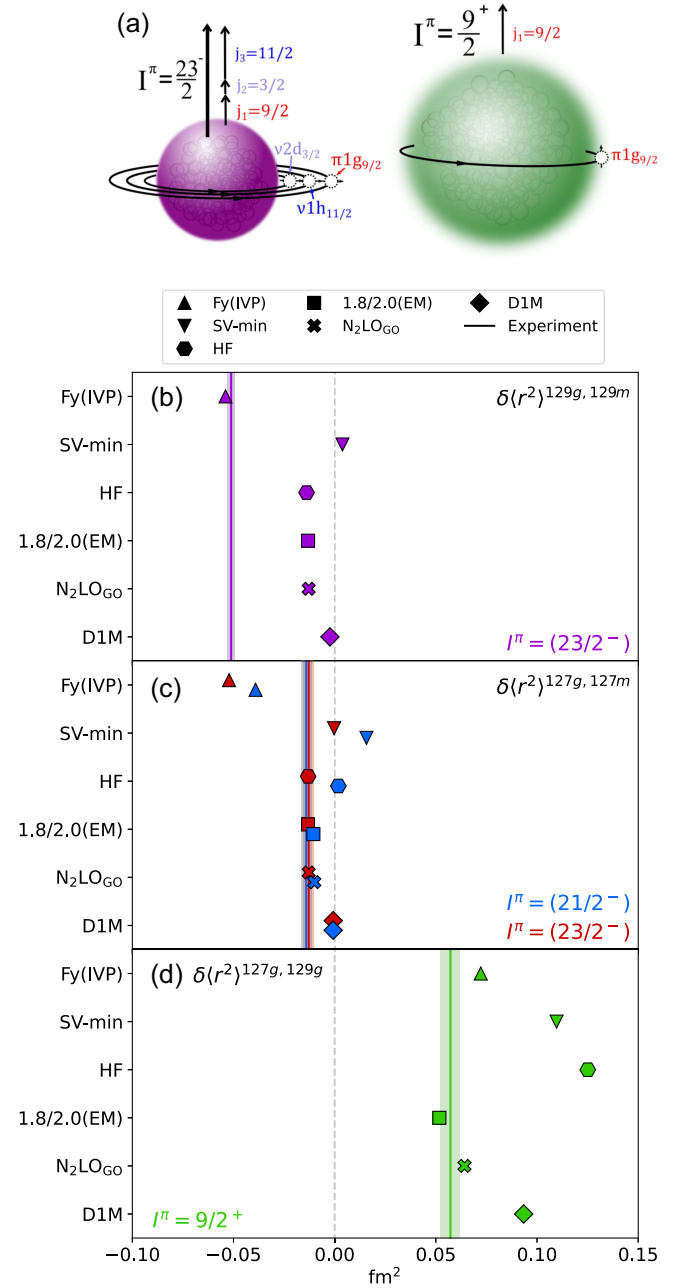


FIG. 1. State-dependent charge radii of the neutron-rich indium isotopes. (a) Shell-model configurations associated with the $I^\pi = 9/2^+$ g.s. and $I^\pi = (23/2)^-$ high-spin isomer in ^{129}In . (b)–(d) Change in charge radii of the ground state and high-spin states in indium isotopes. Experimental data (lines) are compared with results of the DFT (Fy(IVP), SV-min, and HF), VS-IMSRG [1.8/2.0(EM) and $N_2\text{LO}_{\text{GO}}$] and MPMH (D1M) calculations (markers). (b) $\delta\langle r^2 \rangle^{g,m}$ of the high-spin $(23/2)^-$ isomer of ^{129}In relative to the $9/2^+$ g.s. (c) $\delta\langle r^2 \rangle^{g,m}$ of the high-spin isomer of ^{127}In relative to the $9/2^+$ g.s. assuming $I^\pi = (21/2)^-$ (blue) and $(23/2)^-$ (red) for the isomer. (d) Change in g.s. charge radii $\delta\langle r^2 \rangle^{127,129}$ from Ref. [20]. The vertical lines show the experimental values corresponding to the weighted average of the $\delta\langle r^2 \rangle$ values for the $5p^2P_{1/2} \rightarrow 8s^2S_{1/2}$ and $5p^2P_{3/2} \rightarrow 9s^2S_{1/2}$ transitions, with the uncertainty given by the shaded area.

TABLE I. The measured $\delta\nu^{127,129}$ and $\delta\nu^{g,m}$ values and extracted $\delta\langle r^2 \rangle^{127,129}$ and $\delta\langle r^2 \rangle^{g,m}$ values. Experimental uncertainties are given in parentheses. Uncertainties from atomic theory calculations, shown in square brackets, were calculated following the approach presented in Ref. [59]. The values for the isotope shifts and changes in the charge radii for both the ground-state 9/2 and low-spin 1/2 isomers can be found in Ref. [19]. For the high-spin state in ^{127}In , values are given for two possible spin assignments of $(21/2^-)$ and $(23/2^-)$. F and K^{MS} values used to calculate $\delta\langle r^2 \rangle^{A,A'}$ are taken from Refs. [20,55].

Transition	F (GHz/fm ²)	K^{MS} (GHz · u)	$\delta\nu^{127,129}$ (MHz)	$\delta\langle r^2 \rangle^{127,129}$ (fm ²)	Mass	I^π	$\delta\nu^{g,m}$ (MHz)	$\delta\langle r^2 \rangle^{g,m}$ (fm ²)
$5p^2P_{1/2} \rightarrow 8s^2S_{1/2}$	1.626[30]	216[74]	139(7)	0.069(4)[6]	127	(21/2 ⁻)	-29(7)	-0.0179(43)[3]
					127	(23/2 ⁻)	-21(7)	-0.0129(43)[2]
					129	23/2 ⁻	-87(7)	-0.0535(43)[10]
$5p^2P_{3/2} \rightarrow 9s^2S_{1/2}$	1.577[27]	325[73]	122(4)	0.052(3)[6]	127	(21/2 ⁻)	-19(5)	-0.0121(32)[2]
					127	(23/2 ⁻)	-20(6)	-0.0128(36)[2]
					129	23/2 ⁻	-80(3)	-0.0508(18)[9]
Average				0.057(2)[6]	127	(21/2 ⁻)		-0.0141(26)[3]
					127	(23/2 ⁻)		-0.0128(28)[2]
					129	23/2 ⁻		-0.0512(17)[9]

all of the 3 unpaired holes are fully aligned. Therefore, the measured spectra have been analyzed assuming the two possible spin assignments for ^{127}In , 21/2 and 23/2.

Experimental results—Our measurements of the high-spin isomers were enabled by recent developments in the sensitive method of collinear laser spectroscopy at radioactive ion beam facilities (see Supplemental Material [23]), which allowed high-resolution measurements of the high-spin isomers produced in a high background of other long-lived species, such as other isotopes or molecules with similar mass. The neutron-rich $^{127,129}\text{In}$ isotopes and their high-spin isomeric states were produced at the isotope separation online facility (ISOLDE) at CERN and directed to the collinear resonance ionization spectroscopy (CRIS) experiment [52] for sensitive measurements of their hyperfine structure. The hyperfine spectra of $^{127,129}\text{In}$ were measured in the $5p^2P_{1/2} \rightarrow 8s^2S_{1/2}$ (246.0 nm) and $5p^2P_{3/2} \rightarrow 9s^2S_{1/2}$ (246.8 nm) transitions of the indium atom. A major experimental challenge in laser spectroscopy studies of high-spin isotopes is the population of several possible hyperfine states, which causes a highly congested spectrum. Thus, we use the transition from the low electronic state, $^2P_{1/2}$, which does not exhibit quadrupole hyperfine splitting, to obtain the nuclear magnetic moment. Subsequently, once the magnetic moment is fixed, we use a transition from the state $^2P_{3/2}$ to extract the nuclear quadrupole moment. More details of the experimental setup alongside examples of the measured spectra can be found in the Supplemental Material [23].

The isomer shift, $\delta\nu^{g,m}$, measured as the difference between the centroid frequency of the hyperfine structure of the ground and isomeric state, is related to the change of their root-mean-squared charge radii, $\delta\langle r^2 \rangle^{g,m}$, using the relation

$$\delta\nu^{g,m} = F\delta\langle r^2 \rangle^{g,m} + K^{\text{MS}} \frac{m_m - m_g}{m_g m_m}, \quad (1)$$

where m_g and m_m are the masses of the ground and high-spin states of the $^{127,129}\text{In}$ isotopes [53,54], respectively, F and $K^{\text{MS}} = K^{\text{SMS}} + K^{\text{NMS}}$ are the field-shift (FS) and mass-shift (MS) constants, obtained from recently developed atomic calculations, as discussed in Refs. [20,55]. Similarly, the isotope shift between ^{129}In and ^{127}In were obtained from the frequency shift between isotopes, $\delta\nu^{127,129}$. The differential charge radii in Eq. (1) are defined as: $\delta\langle r^2 \rangle^{A,A'} \equiv \langle r_c^2 \rangle^{A'} - \langle r_c^2 \rangle^A$.

The $\delta\nu^{g,m}$ and $\delta\nu^{127,129}$ measurements and extracted $\delta\langle r^2 \rangle^{g,m}$ and $\delta\langle r^2 \rangle^{127,129}$ values are displayed in Table I. The average values presented are a weighted average of the mean squared charge radii extracted for the two atomic transitions, weighted by their statistical uncertainties. The largest systematic uncertainty (of the two atomic transitions) is quoted alongside. The ground and high-spin states could be identified by their relative intensity and electromagnetic moments. The spin of the high-spin isomers, $(21/2^-)$ for ^{127}In and $23/2^-$ for ^{129}In , were taken from $\beta\gamma$ -coincidence measurements and supported by theoretical predictions [21,56–58]. We present results obtained by assuming both $(21/2^-)$ and $(23/2^-)$ spins for ^{127}In .

The spectroscopic nuclear electric quadrupole moments, Q_S , were extracted from the measured hyperfine quadrupole constants, B_{hf} , using the relation

$$B_{hf} = eQ_S V_{zz}, \quad (2)$$

where a value of $B_{hf}(^2P_{3/2})/Q_S = 576(4)$ MHz/b obtained from relativistic coupled-cluster calculations was used [19,61]. The magnetic moments, μ , were determined from the magnetic hyperfine constant, A_{hf} , using a reference NMR value of $\mu_{\text{ref}} = +5.5408(2) \mu_N$ [62], and the measured A_{ref} for the isotope ^{115}In [19,63], according to

TABLE II. The hyperfine structure parameters A_{hf} and B_{hf} for the ^{127}In and ^{129}In isotopes, extracted magnetic moment μ , spectroscopic quadrupole moment Q_S , and intrinsic quadrupole moment Q_0 values. The high-spin states were measured in this Letter. Statistical uncertainties (arising from experimental measurements) and systematic uncertainties (from atomic theory calculations) for Q_S are given in parentheses and square brackets, respectively. For the high-spin state in ^{127}In , values are given for two possible spin assignments of $(21/2^-)$ and $(23/2^-)$. The results for the ground states, $9/2^+$, were taken from Ref. [19].

Mass	I^π	A_{hf} (MHz)				μ (μ_N)	B_{hf} (MHz)		
		$5p\ ^2P_{3/2}$	$9s\ ^2S_{1/2}$	$5p\ ^2P_{1/2}$	$8s\ ^2S_{1/2}$		$5p\ ^2P_{3/2}$	Q_S (mb)	Q_0^a (mb)
127	$9/2^+$	242(1)	130(1)	2278.3(6)	243.8(4)	5.532(2)	338(16)	587(28)[4]	1076(53)
129	$9/2^+$	243(1)	132(1)	2304.9(9)	244.8(7)	5.596(2)	280(7)	487(13)[3]	893(24)
127	$(21/2^-)$	99(1)	54(1)	942.5(7)	100.9(7)	5.340(4)	464(12)	805(21)[6]	1058(29)
127	$(23/2^-)$	91(1)	50(1)	863.8(7)	92.3(7)	5.360(5)	470(10)	815(18)[6]	1050(24)
129	$23/2^-$	101(1)	55(1)	956.6(6)	101.3(4)	5.935(4)	344(18)	598(32)[4]	768(41)

^aThe intrinsic quadrupole moments, Q_0 , were estimated using $Q_0 = [(I+1)(2I+3)/I(2I-1)]Q_S$ [60].

$$\mu = \mu_{\text{ref}} \frac{IA_{hf}}{I_{\text{ref}}, A_{\text{ref}}} (1 + \Delta), \quad (3)$$

where the differential hyperfine anomaly Δ is not considered as it is expected to be smaller than our experimental uncertainty for these isotopes [64]. The results for the hyperfine structure constants and electromagnetic moments are shown in Table II.

Theoretical results—We compare our experimental $\delta\langle r^2 \rangle$ values to nuclear structure calculations, performed using three complementary theoretical methods: (i) valence-space in-medium similarity renormalization Group (VS-IMSRG) method [65–67]; (ii) multiconfiguration self-consistent field approach (MPMH) [68–72]; and (iii) density functional theory (DFT) [19,73,74]. The extracted $\delta\langle r^2 \rangle^{g,m}$ and $\delta\langle r^2 \rangle^{127,129}$ values are shown alongside calculated values in Fig. 1.

The VS-IMSRG approach aims to solve the nuclear many-body problem by performing a unitary transformation which maps the large-scale problem to effective operators acting on a tractable valence space. In implementing the unitary transformations, all intermediate operators are truncated at the two-body level, denoted IMSRG(2). This approximation is generally effective for energies, but misses highly collective quadrupole correlations [75]. The calculations were performed using two different sets of initial two-nucleon (NN) and three-nucleon (3N) forces derived from chiral effective field theory, indicated 1.8/2.0(EM) [76,77] and N²LO_{GO} [78]. The former interaction is constrained by considering properties of two-, three-, and four-nucleon systems, and well reproduces energies throughout the medium and heavy region [79], but generally underpredicts charge radii. The latter one is additionally constrained by the saturation properties of nuclear matter which leads to a better reproduction of the absolute charge radii [78,80].

MPMH and DFT calculations are based on effective in-medium interactions calibrated to nuclear observables across the nuclear chart. The MPMH calculations were

performed using the Gogny D1M [24] effective interaction. The DFT Hartree-Fock-Bogoliubov (HFB) calculations were performed with two different energy density functionals: the Fayans functional Fy(IPV) [20] and the Skyrme functional SV-min [81]. For electromagnetic moments analysis, we also carried out the Hartree-Fock (HF) DFT calculations with the Skyrme functional UNEDF1 [82] with the spin-spin term adjusted to reproduce the magnetic moment of ^{131}In [19]. More details on the theoretical approaches used are provided in the Supplemental Material [23].

Discussion—All theoretical models provide similar wave function configurations for the high-spin states of $^{127,129}\text{In}$, based on a proton $g_{9/2}$ hole and a two-quasiparticle $d_{3/2}h_{11/2}$ neutron configuration, see Fig. 1(a) and Refs. [21,56,58]. As seen in Fig. 1, a reduction in the $\langle r^2 \rangle$ values was observed for the high-spin isomer states relative to their ground states, resulting in a negative value of $\delta\langle r^2 \rangle^{g,m}$. A variation of the charge radius relative to the g.s. radius has also been observed for other high-spin isomers across different regions of the nuclear chart [12,13,25,83–86]. Notably, the decrease observed for the high-spin isomer of ^{129}In seen in Fig. 1(b) is about three times larger than the value observed for ^{127}In , and comparable to the charge radius change between the g.s. configurations of ^{127}In and ^{129}In , $\delta\langle r^2 \rangle^{127,129}$, shown in Fig. 1(d). This large reduction of the charge radius can only be explained by DFT calculations for the $I^\pi = 23/2^-$ maximally aligned state using the Fy(IPV) functional. The larger downshift of radii for the $I^\pi = 23/2^-$ state is suggested to be most likely caused by the additional gradient terms in the pairing and surface part of the Fayans functional. Such terms are absent in SV-min and HF models.

As seen in Figs. 1(b) and 1(c), the DFT models predict similar, yet relatively large, differences in charge radii compared to the ground state for both high-spin isomers. In contrast, other models indicate significantly smaller

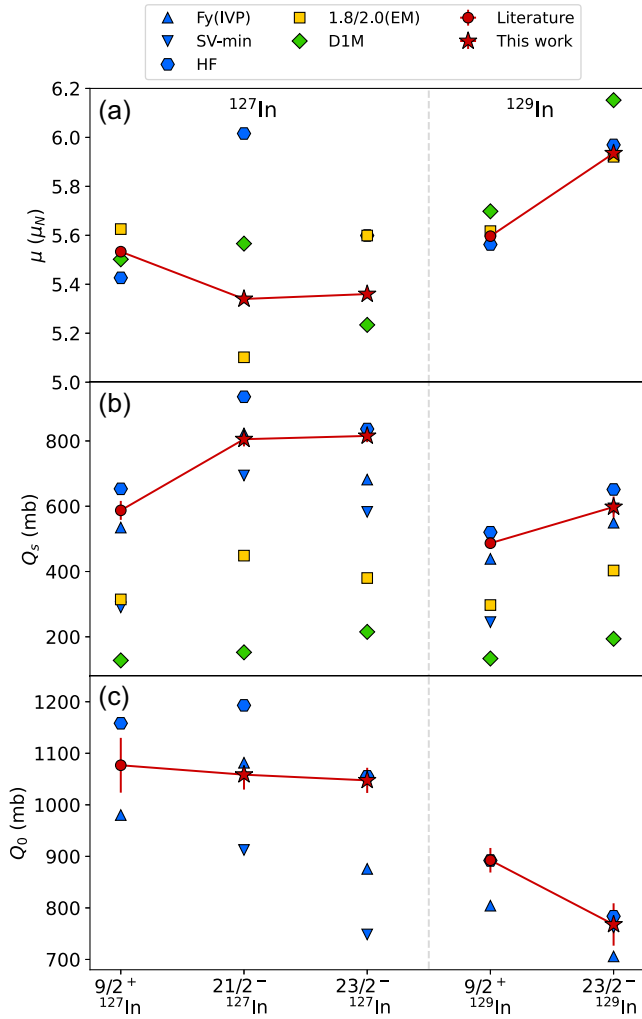


FIG. 2. Electromagnetic moments of the ground state and high-spin isomers of ^{127}In and ^{129}In : (a) spectroscopic magnetic dipole moments, (b) spectroscopic quadrupole moments, and (c) intrinsic quadrupole moments. Experimental results (connected red markers) are compared with different calculations: DFT (blue markers), VS-IMSRG (gold markers), and MPMH (green markers). Results for the high-spin isomer of ^{127}In are shown assuming two possible nuclear spin values of $21/2^-$ and $23/2^-$. Literature values for the ground states were taken from Ref. [19]. The line connecting the experimental results is for visual guidance only.

changes in charge radii for both isomeric states. The DFT result for $I^\pi = 21/2^-$, however, has to be interpreted with caution because quasiparticle configuration of this state cannot be assigned unambiguously, see Supplemental Material [23]. For all models, the predicted change in the g.s. charge radii $\delta\langle r^2 \rangle^{127,129}$ is computed to be positive, consistent with experiment, but it exhibits a rather large model dependence.

Experimental results for the magnetic dipole and nuclear quadrupole moments are shown in Fig. 2. To separate the dependence on the nuclear spin, the intrinsic quadrupole moments, $Q_0 = [(I+1)(2I+3)/I(2I-1)]Q_s$, were

extracted from the measured spectroscopic moment Q_s , assuming a strong coupling scheme for these high-spin isomers [60]. They are shown in Fig. 2(c). In the case of HF and IMSRG, the intrinsic quadrupole moments were calculated independently, rather than extracted from the spectroscopic quadrupole moment. The extracted electromagnetic moments are compared with theoretical results in Fig. 2(a). All calculations were performed without explicit adjustment of effective charges or effective g-factors. All models reproduce the trend observed for the magnetic dipole moments, with larger deviations obtained for ^{127m}In . For IMSRG, the magnetic moment calculations include the effect of two-body currents [87], which is essential to obtain a good agreement with the experiment. A comparison of the results obtained with and without two-body currents is shown in Fig. 4 in the Supplemental Material [23]. For HF, the agreement relies on adjusting the Landau parameter g'_0 in ^{131}In [19].

In Fig. 2(b) we compare our experimental (spectroscopic) quadrupole moments to the predicted values from DFT, MPMH, and IMSRG calculations. In general, the IMSRG and MPMH models largely underestimate the observed quadrupole moments. This is a well-known feature for the IMSRG calculations due to the IMSRG (2) truncation. For the MPMH model, the underestimation is due to the absent $\Delta N = 2$ quadrupole polarization which is neglected in the current valence space truncation scheme. The DFT calculations all reproduce the experimental moments rather well, especially for the ^{129}In states and the ^{127}In ground state. The quadrupole moments predicted by SV-min differ significantly from the values predicted by Fy(IVP) and HF calculations, with the appreciable increase of quadrupole moments in the high-spin states. This might be related to the pairing strength as stronger pairing reduces deformation effects. Indeed, SV-min predicts stronger pairing correlations than Fy(IVP), and HF has no pairing at all. In Fig. 2(c) we compare the results obtained for the intrinsic quadrupole moments. The HF values reproduce the intrinsic moments for both ^{129}In states very well. For ^{127}In , a better agreement is observed if a spin $23/2^-$ is assumed for the isomeric state.

While some of the reduction of the charge radius of ^{129m}In can be attributed to a reduced quadrupole collectivity in this excited state, this cannot account for the entire reduction. Indeed, a similar reduction in Q_0 is predicted by both Fy(IVP) and HF models, but their values of $\delta\langle r^2 \rangle^{g,m}$ are different. Moreover, the similar values of the charge radii of g.s. and isomeric state in ^{127}In are not reproduced by Fy(IVP). However, as discussed in the Supplemental Material [23], the $21/2^-$ state cannot be unambiguously computed within a single-reference quasiparticle framework.

Conclusions and outlook—Our results reveal a significant reduction in the nuclear size and intrinsic quadrupole moments in the high-spin isomer ^{129}In . The new data on isomeric electromagnetic moments were interpreted by

complementary theoretical approaches that capture different aspects of nuclear structure. Except for Fy(IVP) , our models cannot reproduce the magnitude of the charge radii reduction observed for the high-spin isomer of ^{129}In . On the other hand, for the isomer ^{127m}In , the Fy(IVP) model overestimates the observed value of the shift $\delta\langle r^2 \rangle^{g,m}$ for ^{127}In , while DFT-HF and VS-IMSRG calculations predict a shift closer to the experimental value. The magnitude of the charge radius change between the ground states, $\delta\langle r^2 \rangle^{127,129}$, appears to be highly sensitive to the employed many-body methods, with DFT-Fy(IVP) and VS-IMSRG in closer agreement with experiment. The quadrupole moments are surprisingly well described by Fy(IVP) and HF approaches. Both VS-IMSRG and MPMH significantly underestimate the measured quadrupole moments. Based on our theoretical analysis, the reduction of $\delta\langle r^2 \rangle^{g,m}$ for ^{129}In cannot be solely attributed to a shape effect.

IMSRG, MPMH, and HF calculations show good agreement with the measured nuclear magnetic dipole moments, suggesting a preference for an $I^\pi = 23/2^-$ assignment for ^{127m}In . The quadrupole moments can be reproduced by Fy(IVP) and HF DFT calculations when using an unabridged single-particle space, allowing for the full development of shape polarization. It is apparent that no single model can consistently describe the rich experimental data on electromagnetic moments in ground and isomeric states of $^{127,129}\text{In}$. The issue is expected to arise from the contrasting nature of the ground and high-spin states. A similar structure of the ground states in the neighboring isotopes will partially cancel systematic uncertainties in their charge radius differences, whereas such cancellation may not occur for their isomers. A consistent description will require accurate calculations for both ground and high-spin states and remains a theoretical challenge. Future measurements of high-spin isomers near doubly magic nuclei such as ^{56}Ni , ^{100}Sn , and ^{208}Pb will be important for guiding further developments of nuclear theory and understanding the structure of nuclear isomers.

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Data availability—The data that support the findings of this article are openly available [88–90].

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