

Charge Radii Measurements of Exotic Tin Isotopes in the Proximity of $N = 50$ and $N = 82$

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We report nuclear charge radii for the isotopes $^{104-134}\text{Sn}$, measured using two different collinear laser spectroscopy techniques at ISOLDE-CERN. These measurements clarify the archlike trend in charge radii along the isotopic chain and reveal an odd-even staggering that is more pronounced near the $N = 50$ and $N = 82$ shell closures. The observed local trends are well described by both nuclear density functional theory and valence space in-medium similarity renormalization group calculations. Both theories predict appreciable contributions from beyond-mean-field correlations to the charge radii of the neutron-deficient tin isotopes. The models, however, fall short of reproducing the magnitude of the known $B(E2)$ transition probabilities, highlighting the remaining challenges in achieving a unified description of both ground-state properties and collective phenomena.

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Introduction—The mean-square charge radius is a fundamental property of the atomic nucleus, providing direct insight into its size and the spatial distribution of nuclear electric charge. Precise measurements of its evolution along isotopic chains offer unique insight into nuclear shell structure, collective phenomena, and the underlying forces. Understanding how single-nucleon and collective motion emerges from microscopic interactions between protons and neutrons is an essential step towards a unified description of the atomic nucleus. Collective nuclear properties are commonly enhanced in open-shell nuclei and significantly reduced around closed nuclear shells. In this context, the tin (Sn) isotopic chain, with its closed proton shell at $Z = 50$, is of particular importance. Currently, it offers a long sequence of isotopes available for study, stretching across the major neutron shell closures at $N = 50$ and $N = 82$. This unique landscape makes the tin chain a fertile testing ground for the development of theoretical *ab initio* models [1–7]. Many properties of nuclei in the vicinity of the doubly magic nuclei are expected to be accurately predicted by considering only a few valence nucleons. While the doubly magic nature of ^{132}Sn has been well-established experimentally [8–12], significantly less data are available to understand the evolution of nuclear structure properties in the region from midshell down towards ^{100}Sn . This is due to the considerable experimental challenges associated with producing and studying these neutron-deficient isotopes. The study of these isotopes is crucial as contradictory experimental evidence has been reported about the evolution of single-particle properties approaching ^{100}Sn [13–15], and several questions remain in connection with the robustness of the $N = Z = 50$ shell closures [1,5,15–19]. While the largest Gamow-Teller strength among all allowed β decays was found in ^{100}Sn and supports a dominant single-particle behavior [1], the existence of low-lying energy levels and relatively large $B(E2)$ values, known down to ^{104}Sn [16,20–22], are indicative of a collective picture, in contrast with recent laser spectroscopy results on ^{49}In [23] and ^{47}Ag [17] isotopes.

In this Letter, we present nuclear charge radii measurements for the tin isotopes $^{104-134}\text{Sn}$, using two collinear laser spectroscopy (CLS) techniques [24]. The neutron-deficient isotopes, down to ^{104}Sn , were investigated with

the collinear resonance ionization spectroscopy (CRIS) setup [25], while the neutron-rich isotopes, up to ^{134}Sn , were measured using the collinear laser spectroscopy (COLLAPS) experiment [10]. Our measurement campaigns thus extend the available data [26–28] by four isotopes on the neutron-deficient side $^{104-107}\text{Sn}$, while completing the earlier reported radii in Ref. [10] with the odd-even isotopes including ^{133}Sn [29]. This exceptionally long dataset, spanning thirty-one isotopes, serves to reveal intricate changes in the mean-square charge radii towards the neutron shell closures, which can be explored by nuclear density functional theory (DFT) [31,32] and *ab initio* valence space in-medium similarity renormalization group (VS-IMSRG) calculations [33,34].

Experiment—Two CLS methods were employed to measure the charge radii of tin isotopes. Neutron-deficient isotopes, from ^{124}Sn down to ^{104}Sn , were measured at CRIS using two recently developed ionization schemes for probing the two atomic transitions $5s^25p^2\ ^3P_1 \rightarrow 5s^25p6s\ ^3P_2$ (284 nm) and $5s^25p^2\ ^1S_0 \rightarrow 5s^25p7s\ ^1P_1$ (281 nm) [35]. Neutron-rich isotopes, from ^{108}Sn up to ^{134}Sn were investigated at COLLAPS using two complementary transitions, $5s^25p^2\ ^1S_0 \rightarrow 5p6s\ ^1P_1$ (452.5 nm) and $5s^25p^2\ ^3P_0 \rightarrow 5p6s\ ^3P_1$ (286.3 nm). These transitions were chosen to maximize sensitivity to nuclear observables. Detailed information about the ion beam production and experimental technique used in the CRIS experiment, as well as the deduced isotope shifts and mean-square charge radii, can be found in Supplemental Material [36]. For the COLLAPS measurements, all details are described in Refs. [10,11,37]. The changes in the mean-square nuclear charge radii were extracted from the measured isotope shifts following the relation

$$\delta\nu^{A',A} = \nu_i^A - \nu_i^{A'} = M_i \frac{m_A - m_{A'}}{m_A m_{A'}} + F_i \delta\langle r^2 \rangle^{A',A}, \quad (1)$$

where F_i and M_i are, respectively, the field-shift factor and the mass-shift factor of the transition i , while $m_{A'}$ and m_A are the atomic masses of different isotopes.

The atomic factors, F_i and M_i , were extracted from a standard King-plot analysis (as described in Ref. [10]) using muonic data as well as V_2^e factors derived from elastic

TABLE I. Spins and differences in mean-square charge radii of tin isotopes derived from four measured transitions. Statistical uncertainties are indicated in parentheses and systematic contributions are provided in brackets. Details are presented in Supplemental Material [36]. We note that these $\delta\langle r^2 \rangle$ values must not be combined with the ones previously published for the even-even isotopes in Ref. [10] since those are also included in the present analysis.

A	I^π	$\delta\langle r^2 \rangle^{124,A}$ (fm ²)	$\langle r^2 \rangle^{1/2}$ (fm)
104 ^a	0	−1.497 (5) [41]	4.512 (5)
105 ^a	5/2 ⁺	−1.408 (4) [38]	4.522 (4)
106 ^a	0	−1.257 (3) [25]	4.539 (3)
107 ^a	5/2 ⁺	−1.180 (3) [24]	4.547 (3)
108	0	−1.072 (3) [16]	4.559 (2)
109	5/2 ⁺	−1.002 (1) [16]	4.567 (2)
110	0	−0.901 (3) [10]	4.578 (2)
111	7/2 ⁺	−0.848 (1) [12]	4.583 (2)
112	0	−0.747 (1) [6]	4.594 (1)
113	1/2 ⁺	−0.685 (2) [7]	4.601 (1)
114	0	−0.606 (1) [9]	4.610 (1)
115	1/2 ⁺	−0.560 (1) [5]	4.615 (1)
116	0	−0.462 (1) [7]	4.625 (1)
117	1/2 ⁺	−0.412 (1) [4]	4.631 (1)
118	0	−0.324 (1) [3]	4.640 (1)
119	1/2 ⁺	−0.282 (1) [5]	4.645 (1)
120	0	−0.206 (1) [2]	4.653 (1)
121	3/2 ⁺	−0.162 (1) [5]	4.658 (1)
122	0	−0.097 (1) [1]	4.665 (1)
123	11/2 [−]	−0.060 (1) [3]	4.669 (1)
124	0	0	
125	11/2 [−]	0.033 (1) [4]	4.679 (1)
126	0	0.089 (1) [4]	4.684 (1)
127	11/2 [−]	0.120 (1) [8]	4.688 (1)
128	0	0.165 (1) [9]	4.693 (1)
129	3/2 ⁺	0.177 (2) [18]	4.694 (2)
130	0	0.240 (1) [15]	4.701 (2)
131	3/2 ⁺	0.238 (1) [25]	4.700 (3)
132	0	0.307 (1) [21]	4.708 (2)
133 ^a	7/2 [−]	0.406 (1) [12]	4.718 (2)
134	0	0.531 (2) [5]	4.731 (1)

^aIt indicates isotopes measured for the first time in this Letter.

electron scattering [36,38]. Only the measured isotope shifts of the even stable isotopes were used in the King-plot analysis. Changes in charge radii extracted from the four transitions agree well with each other. The weighted averages were used to determine the nuclear charge radii of the tin isotopes based on the ¹²⁴Sn reference value of $\langle r^2 \rangle^{1/2} = 4.675(1)$ fm of Ref. [38]. The results are presented in Table I. The uncertainties in the extraction of charge radii are dominated by the atomic factor uncertainty.

Theory—The measured radii are confronted with predictions utilizing both the VS-IMSRG and DFT methods. The VS-IMSRG results employ two interactions derived from chiral effective field theory, 1.8/2.0 (EM) [39,40] and $\Delta N^2\text{LO}_{\text{GO}}(394)$ [41]. The 1.8/2.0 (EM) interaction was

fitted to data of few-nucleon systems $A \leq 4$ and globally reproduces ground-state energies up to $A \sim 100$ nuclei [42]. This interaction has been shown to provide a reasonably good description of local variations of nuclear charge radii in the medium-mass region [43], while underestimating their absolute values [44]. In contrast, the $\Delta N^2\text{LO}_{\text{GO}}(394)$ interaction includes the Δ -isobar degree of freedom and was fitted to few-body data, selected nuclei, and properties of infinite nuclear matter. This interaction improves the simultaneous reproduction of energies and radii [41]. Further details of the VS-IMSRG calculations are provided in Supplemental Material [36].

The DFT-Fy(IVP) results were obtained with the HFB solver SkyAx for axially symmetric deformed configurations [45]. The computation of broken-pair states in odd- N isotopes involves the breaking of time-reversal symmetry [46]. It has been calibrated using the large dataset of ground-state properties of semimagic nuclei from Ref. [47] augmented by the data on differential charge radii of the calcium [48], tin [$\delta\langle r^2 \rangle(^{124}\text{Sn} - ^{132}\text{Sn})$], and lead isotopic chains [23]. The calculated charge radii were obtained directly from the charge form factors containing relativistic corrections (including spin-orbit term) and contributions from nucleonic charge form factors [49]. The calibration of the functional produces statistical uncertainties from propagation of the uncertainties of the model parameters [47,50]. Systematic uncertainties come from approximate angular momentum projection and from collective ground-state correlations stemming from low-lying quadrupole excitations [51]. The latter generally increase the radii and so introduce asymmetric uncertainties.

Discussion—In Fig. 1, we present a detailed comparison between our measured charge radii and predictions using both VS-IMSRG and DFT approaches using the experimentally determined spins. Figure 1(a) reveals that both interactions used in the VS-IMSRG calculations underestimate the absolute charge radii. This underestimation is primarily attributed to an insufficient description of saturation [44,52]. In the description of saturation in VS-IMSRG, three-nucleon (3N) forces play an important role. These cannot be fixed by information from NN scattering but largely cancel in the differential radii $\delta\langle r^2 \rangle$ that can thus serve as a crucial test for VS-IMSRG calculations. Beyond $N = 82$, the VS-IMSRG calculations were performed with a different valence space only for the 1.8/2.0(EM) interaction. As the systematic uncertainties are different in different valence-space calculations, it is challenging to discuss the behavior at $N = 82$ in the VS-IMSRG. Indeed, at $N = 82$, we observed a 0.12 fm² difference in the squared radii from the different valence-space calculations.

All modern nuclear DFT models (Skyrme, Fayans, Gogny, relativistic mean field) provide a good description of nuclear radii across the chart of isotopes from $A = 16$ on, typically within ± 0.02 fm. They differ in their performance concerning isotopic trends and details in a particular region.

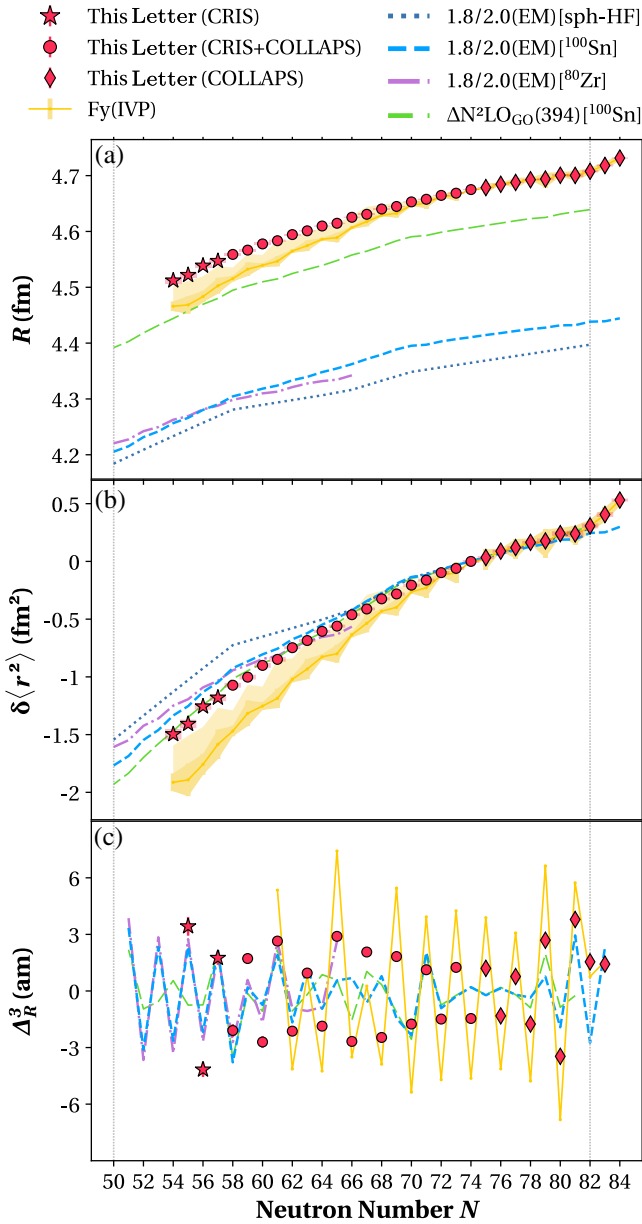


FIG. 1. (a) The absolute root-mean-square radii obtained using $R_c(^{124}\text{Sn}) = 4.675(1)$ fm [38]. (b) Differential mean-square charge radii of tin isotopes with respect to ^{124}Sn . (c) The odd-even staggering $\Delta_R^{(3)} = 1/2(R_{A+1} + R_{A-1} - 2R_A)$. Experimental results are compared to VS-IMSRG and DFT results (for details see text). The statistical uncertainties for Fy(IVP) are indicated by dark yellow bands and the systematic uncertainties (mainly ground-state correlations) by light yellow bands.

Fayans functionals are designed to be more flexible in tuning radius trends than other functionals [10,53]. For other nuclear properties, their performance is parallel to other well-calibrated DFT models, see Ref. [30]. The Fy(IVP) functional here provides a further improved description of the radii trend and the absolute radii compared to its former variant Fy(Δr ,HFB) employed in Ref. [10] for even-even tin isotopes, particularly in the

neutron-rich region and across the kink at the $N = 82$ shell closure.

In Fig. 1(b) differential mean-square charge radii relative to ^{124}Sn are shown. They show an archlike trend towards the known kink at the ^{132}Sn doubly magic shell closure. This general behavior superimposed on a linear increase is overall well described by both the DFT and VS-IMSRG calculations. The good agreement with a spherical Hartree-Fock (HF) calculation using the equal-filling approximation (labeled by “1.8/2.0 (EM) sph-HF” and discussed below) indicates that this is primarily driven by the bulk radial increase, whereas the correlations included in the VS-IMSRG calculations become important below $N = 66$. The collective ground-state correlations become sizable in the DFT calculations in the same region, and they move the result closer to experiment (as indicated with a light yellow band).

Finally, we study the odd-even staggering $\Delta_R^{(3)} = 1/2(R_{A+1} + R_{A-1} - 2R_A)$ in Fig. 1(c). The addition of individual neutrons to the nucleus results in appreciable changes to its charge distribution due to pairing and deformation effects [43]. In our data, we observe an increased staggering as we approach the $N = 50$ and $N = 82$ shell closures. In between these shell closures, we note that the staggering appears more irregular in comparison to that of neutron separation energies, as also seen along the copper chain [43], which we attribute to the sensitivity of the charge radius to the occupation of different neutron orbitals with varying spatial extent and polarizability. In addition, our new measurement of the charge radius of ^{133}Sn reveals a discontinuity in the odd-even staggering at $N = 82$, similar to that observed at other major shell closures such as in Ca ($N = 28$) and Pb ($N = 126$) [24]. The radius of ^{133}Sn suggests that a kink occurs at $N = 82$, which was previously identified for the even-even isotopes [10].

The VS-IMSRG calculations for $\Delta_R^{(3)}$ show improved agreement near these shell closures, although they tend to underestimate the staggering in the midshell where a greater collectivity is expected. The Fy(IVP) calculations accurately predict the inversion across $N = 82$ but systematically overestimate the staggering throughout the entire chain due to a slightly too large proton pairing predicted in midshell tin isotopes. It is to be noted that the estimated uncertainty in the radii below $N = 60$ in Fy(IVP) is greater than the staggering magnitude; therefore, the results for these isotopes are omitted in Fig. 1(c).

To gauge the impact of cross-shell excitation [54] across $N, Z = 50$, we extended our 1.8/2.0(EM) VS-IMSRG calculations to a valence space based on the ^{80}Zr core. This model space did, however, not include the $h_{11/2}$ orbit, which is known to be crucial for capturing the nuclear structure effects for isotopes with $N \gtrsim 64$ [55]. Therefore, calculations with a ^{80}Zr core were performed from $N = 50$ up to $N = 66$ only. The two choices of core should yield

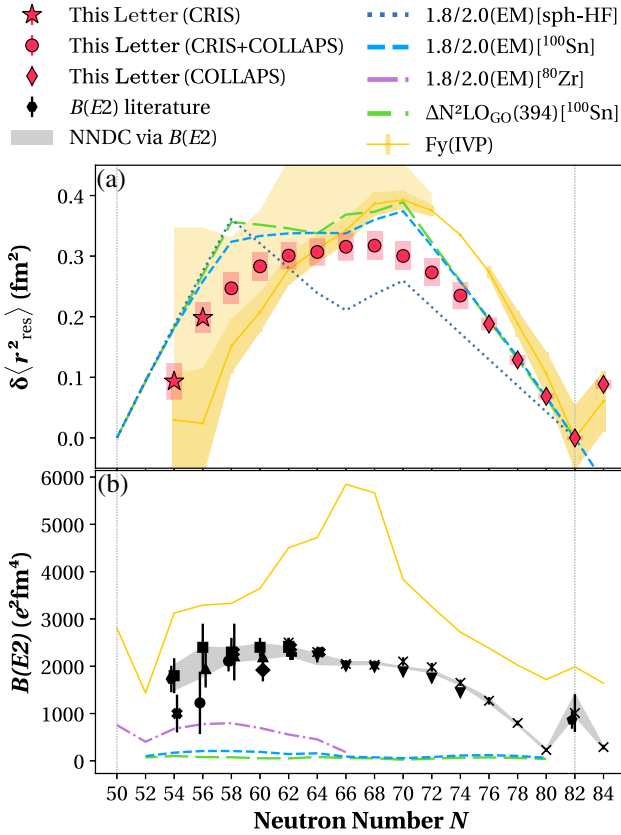


FIG. 2. (a) Residual differential mean-square charge radii $\delta\langle r_{\text{res}}^2 \rangle$ obtained by the subtraction of a linear trend which intersects at the magic numbers $N = 50, 82$ (see Ref. [23] for details). Experimental results are compared with DFT Fy(IVP) calculations, with systematic uncertainties shown as a yellow error band (darker shade represents the influence of ground-state correlations) and VS-IMSRG calculations using different Hamiltonians and different valence spaces (for details see text). The experimental and calculated $B(E2)$ values are shown in panel (b). Experimental $B(E2)$ values were taken from Refs. [9,20–22,56–63], shown with a 1- σ error band from national nuclear data center (NNDC) in Ref. [64].

identical results if operators induced during the IMSRG flow were not truncated, so the discrepancy between them is indicative of the VS-IMSRG truncation error. As shown in Fig. 1, the charge radii of the light tin isotopes predicted in VS-IMSRG show relatively low sensitivity to the assumed core. We therefore conclude the charge radii description can be captured by using a ^{100}Sn core, with calculations performed in the range from $N = 50$ to $N = 82$. (As we discuss below, this is not the case for $E2$ observables).

We now focus on the even-even isotopes, which are simpler to interpret as the odd-neutron polarization is absent. The observed archlike trend in radii between shell closures is highlighted by removing the linear trend between the expected closed-shell isotopes ^{132}Sn and ^{100}Sn . The charge radius of ^{132}Sn at the $N = 82$ shell

closure is known. For ^{100}Sn , the charge radius is estimated by fitting a parabolic function to the even-even radii down to ^{104}Sn , following the same procedure described in Ref. [23]. In Fig. 2(a), the resulting archlike trend is shown after subtracting a bulk linear increase intersecting the charge radii at $N = 50$ and $N = 82$.

This archlike behavior can be compared to the known data on the reduced transition probabilities $B(E2; 0_1^+ \rightarrow 2_1^+)$ in Fig. 2(b). These $B(E2)$ values provide insight into quadrupole collectivity along the isotopic chain. While the measured radii reveal a clearly reduced trend towards $N = 50$, the trend for the $B(E2)$ values is less evident due to the large systematic uncertainty of the experimental values for the neutron-deficient isotopes. The $B(E2)$ values computed with Fy(IVP) were deduced from collective correlations, using the same approach as the correlation correction to the charge radii whereby the BCS approximation was used for simplicity [51]. It is to be noted that these values here represent the sum over all $B(E2; 0^+ \rightarrow 2^+)$ transitions. Since the lowest $E2$ transition usually accounts for $\sim 90\%$ of the total $B(E2)$ strength, the Fy(IVP) values in Fig. 2(b) serve as upper estimates and should be reduced by 10%. The Fy(IVP) results generally yield an archlike trend with a pronounced maximum at midshell and an only moderate reduction toward neutron-deficient isotopes. However, the midshell peak in $B(E2)$ is exaggerated in a similar fashion as the collective corrections for the radii. In addition, note that the DFT predictions of $B(E2)$ rates are still strongly varying among different functionals [51] and carry still rather large uncertainties, a feature which gives leeway for further improvement. A dedicated optimization to low-energy excitations is still a task for future development.

A similar archlike behavior of radii is obtained in the VS-IMSRG calculations, see Fig. 2(a). To explore the origin of this trend, spherical Hartree-Fock calculations were performed in the equal filling approximation [65], in which all m substates are filled equally, following the neutron orbitals in the order $g_{7/2}, d_{5/2}, s_{1/2}, d_{3/2}$, and $h_{11/2}$. Hence, the observed trend is consistent with the charge radius following the neutron radius. The latter is mainly given by the sum of the single-particle mean-square radii weighted by their orbital occupation. The effect of configuration mixing is thus visualized by the comparison with the spherical HF and full VS-IMSRG results. The spherical HF results are not sensitive to collective effects, yet these calculations predict an overall archlike trend in radii. We conclude that—within the VS-IMSRG framework—the evolution of charge radii between the shell closures is dominated by the uniform isotropic growth of the mean field seen by the protons and is not due to many-body correlations. This result emphasizes that care should be taken when studying the correlations between quadrupole collectivity and nuclear charge radii [66–69], as it is

nontrivial to decouple the origin of different contributions to charge radii.

Finally, we note that the VS-IMSRG approach significantly underestimates the magnitude of the $B(E2)$ values, as seen in Fig. 2(b). As shown earlier [70,71] VS-IMSRG calculations based on spherical reference states typically underestimate absolute $B(E2)$ values due to missing particle-hole excitations which are needed to capture the full collective strength. The extended valence space using the ^{80}Zr core shows an improvement, with a large increase for the magnitude of the predicted $B(E2)$ values with respect to the results using a ^{100}Sn core, but still significantly underestimates the experimental values. This further highlights the importance of cross-shell excitations for describing the measured $B(E2)$ values of the neutron-deficient tin isotopes, as previously suggested by large-scale shell model calculations [3]. On the other hand, the charge radii are not affected by much as shown in panel (a) of Fig. 1. Evidently, the many-body dynamics driving the behavior of the charge radii in Fig. 2(a) is distinct from that driving $B(E2)$ in Fig. 2(b). This emphasizes the bulk and collective nature of the charge radii and $B(E2)$ observables, respectively.

Conclusions—We interpreted the laser spectroscopy measurements of charge radii for $^{104-134}\text{Sn}$ by DFT and VS-IMSRG approaches. Both are able to describe the general behavior of the radii. While the general features are better predicted near the shell closures, improved theory is needed to reproduce the observed odd-even staggering in the midshell, where the significant quadrupole collectivity is expected. Both DFT and VS-IMSRG calculations predict appreciable contributions from beyond-mean-field correlations to the charge radii of the neutron-deficient tin isotopes.

As previously noted in Refs. [12,23,70–73], reproducing collective effects such as quadrupole moments and $B(E2)$ values in VS-IMSRG remains a challenge. While an accurate reproduction of absolute charge radii in *ab initio* calculations is linked to developing improved nuclear forces with correct saturation, the underestimation of $B(E2)$ values is related to the lack of collective effects in the spherical VS-IMSRG employed in this Letter.

Within the VS-IMSRG framework, we have shown that the observed archlike trend of charge radii between shell closures is dominated by an expansion of the nucleus due to a gradual filling of neutron shells. This is in contrast with the conclusions obtained for $B(E2)$ values and other $E2$ observables [23], which require the inclusion of higher-order many-body correlations and/or nonspherical reference states. Anticipated charge radii and $B(E2)$ measurements reaching closer to ^{100}Sn will be of great interest to complete our understanding of this frontier region of the nuclear chart.

The data analysis used NumPy [74], SciPy [75], Pandas [76,77], iMinuit [78,79], and QSPEC [80]. The figures were produced using Matplotlib [81].

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