

Fingerprints of Triaxiality in the Charge Radii of Neutron-Rich Ruthenium

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We present the first measurements with a new collinear laser spectroscopy setup at the Argonne Tandem Linac Accelerator System, utilizing its unique capability to deliver neutron-rich refractory metal isotopes produced by the spontaneous fission of ^{252}Cf . We measured isotope shifts from optical spectra for nine radioactive ruthenium isotopes $^{106-114}\text{Ru}$, reaching deep into the mid-shell region. The extracted charge radii are in excellent agreement with predictions from the Brussels-Skyrme-on-a-Grid models that account for the triaxial deformation of nuclear ground states. We show that triaxial deformation impacts charge radii in models that feature shell effects, in contrast to what could be concluded from a liquid drop analysis. This indicates that this exotic type of deformation should not be neglected in regions where it is known to occur, even if its presence cannot be unambiguously inferred through laser spectroscopy.

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Introduction—Deformation is a fundamental concept when describing nuclear ground and excited states: an atomic nucleus does not need to be spherically symmetric but can take a range of different shapes in its intrinsic frame. The fingerprint of deformation and its variation with neutron number N and proton number Z can be discerned in numerous nuclear observables, such as masses and radii. A prominent example is the root-mean-square (rms) nuclear charge radius $R_c = \langle r_c^2 \rangle^{1/2}$. Laser spectroscopy techniques of short-lived radioactive isotopes have uncovered numerous cases where this observable varies rapidly with N along an isotopic chain [1–7]. Even if R_c is, in principle, affected by the deformation of all multipolarities, this observable does not carry any information on the

angular dependence of the nuclear density and cannot—at least, by itself—be used to pin down which multipole moments are relevant to describe a given nucleus. The vast majority of experimental data on R_c is thus interpreted by limiting the nucleus to axially symmetric shapes whose deviation from sphericity can be measured through a single (dimensionless) multipole moment β_{20} . This assumption is usually justified: axially symmetric configurations suffice to describe most nuclei, and the higher order moments of nuclear ground states tend to be significantly smaller than their quadrupole deformation [8].

Yet there are several regions of the nuclear chart where triaxial deformation occurs, i.e., where the nuclear density does not have any rotational symmetry axis and has to be characterized by *two* quadrupole multipole moments: either (β_{20}, β_{22}) or the more widely used (β_2, γ) ; see the Supplemental Material [9] for their definition. The experimental fingerprint of triaxial deformation is faint: While the analysis of rotational bands can offer indications [16], it can only be probed directly through Coulomb excitations [17]. To our knowledge, there has been no experimental search for the impact of triaxial deformation on the rms charge radius. What literature exists on the subject is

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limited to the statement that R_c is largely insensitive to γ ; a stance justified by arguments from a simple liquid drop picture [18–20].

In this Letter, we challenge this state of affairs by presenting the first values for the charge radii of nine neutron-rich ruthenium isotopes, $^{106-114}\text{Ru}$. The triaxial deformation of these isotopes—as well as that of the stable $^{102,104}\text{Ru}$ —is both established experimentally [21–26] and supported by numerous models of different origins [8,27–31]. However, these refractory metals have never been addressed with high-precision laser spectroscopy: their refractory nature makes it challenging to extract them from ISOL-type targets, requiring a buffer-gas-based extraction technique instead. We present data collected from the first experiment at the new Argonne Tandem Hall Laser Beamline for Atom and Ion Spectroscopy (ATLANTIS) at Argonne National Laboratory (ANL). It was specifically designed to target the neutron-rich isotopes of refractory metals produced by the Californium Rare Isotope Breeder Upgrade (CARIBU) ^{252}Cf fission source. We compare the experimental findings to the predictions of the Brussels-Skyrme-on-a-Grid (BskG) model series [8,32–34]; these state-of-the-art energy-density functional models systematically—i.e. across the entire nuclear chart—allow for triaxial deformation when it is energetically favorable. The four available models reproduce the charge radii data remarkably well, as already demonstrated for other observables [35–37]. In these models, which include quantum mechanical shell effects absent in the liquid-drop picture, the triaxial deformation impacts the charge radius R_c . Although the effect is somewhat model dependent, it cannot be ignored when studying the charge radii of isotopes that are known to exhibit triaxial deformation.

Experimental method—We installed ATLANTIS in the low-energy experimental area of the Argonne Tandem Linac Accelerator System (ATLAS) that provides access to CARIBU beams. In CARIBU, neutron-rich fission products from a ^{252}Cf source are thermalized inside a helium-filled large-volume gas catcher [38], extracted as singly charged ions and transported to the end station via radio frequency (RF) and electrostatic guides [39,40]. We performed the first isotope shift measurements on neutron-rich ruthenium and demonstrated very high sensitivity for fluorescence-detection laser spectroscopy, recording the spectrum of the least abundant isotope ^{114}Ru despite a production rate of only a few tens of ions per second.

The ATLAS low-energy area features a new helium-filled RF quadrupole beam cooler buncher [41,42] based on the design described in [43,44]. Accumulation times of up to 30 s were used before releasing bunches with a time spread of $\approx 1\ \mu\text{s}$. The high compression ratio significantly improves the signal-to-noise ratio of the laser spectroscopic measurement [45]. The temporal signal distribution shows a homogeneous profile which is shown in the inset of

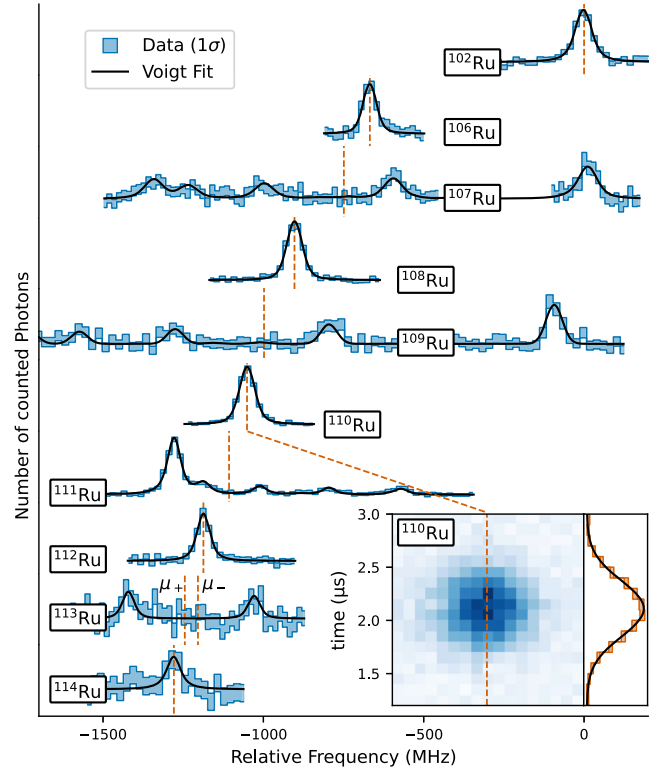


FIG. 1. A sample of the recorded data for the stable reference isotope ^{102}Ru (top row) and all other recorded radioactive isotopes. The solid black lines show a fit to the data. The inset shows a time-frequency profile of a ^{110}Ru resonance.

Fig. 1 for ^{110}Ru . The high voltage applied to the buncher defines the ion starting potential and is actively stabilized to 23 750 V via a temperature-stabilized high-precision resistor chain voltage divider [46].

A charge-exchange cell, placed directly before the fluorescence detection region, neutralizes the ion beam and allows us to access neutral atoms. In contrast to traditional approaches that use alkaline elements, we use magnesium as the neutralization agent to more closely match the ionization energy of ruthenium. It provided an extraordinarily high charge-exchange efficiency of $\sim 90\%$ and a strong ground state population at approximately 400°C . In addition, we did not observe any noticeable distortion of the line shapes of the atomic spectra due to the near-resonant charge exchange into the atomic ground state.

With these highly efficient devices in place, we performed laser spectroscopy in the 350-nm ground-state transition of neutral ruthenium between the atomic levels $[\text{Kr}] 4d^7 5s^5 F_5$ ($0\ \text{cm}^{-1}$) and $[\text{Kr}] 4d^7 5p^5 G_6$ ($28\,571.89\ \text{cm}^{-1}$). The light was generated by a continuous-wave, titanium-sapphire laser (Sirah Matisse) with subsequent frequency doubling in a nonlinear crystal placed in an external enhancement cavity (Spectra Physics Wavetrain). The laser frequency was stabilized to a wavelength meter (High Finesse WS-8) that

was referenced to a He:Ne laser with less than 2-MHz frequency drifts within the time frame of typical isotope-shift measurements [47,48].

Stable ruthenium ions are obtained from a laser-ablation ion source installed upstream of the buncher. Each measurement of a radioactive isotope from CARIBU was bracketed by reference measurements on stable ^{102}Ru to account for potential drifts in voltage or frequency. Care was taken to ensure that online and offline bunch properties were comparable to prevent systematic frequency shifts, particularly those caused by overfilling the buncher for the stable beam.

Data analysis—The resonances in the recorded spectra show the best agreement with a Voigt profile, a convolution of Lorentzian [γ held fixed at 33(1) MHz] and Gaussian line shape due to thermal broadening ($\sigma \approx 15$ MHz, fitted). Figure 1 shows samples of the recorded spectra. Hyperfine structure (hfs) emerges from the interaction of the electron angular momentum J and the nuclear spin I . The nuclear magnetic dipole moment and, in case $I > 1/2$, the electric quadrupole moment, relative to the known values of $^{99,101}\text{Ru}$ [49,50], can be extracted from the hfs via the hyperfine parameters A and B . The ratios of the hyperfine parameters for the upper and the lower states were fixed to $A_u/A_l = 0.4229(3)$ and $B_u/B_l = 2.23(2)$, obtained from high precision spectra of the stable isotopes, which are not discussed in this Letter but agree with literature [51]. We can exclude a hyperfine anomaly larger than 1% (2% for ^{113}Ru).

We fitted the data using a χ^2 optimization package described in [52]. The uncertainties in centroid frequencies were scaled with the reduced χ^2 , which was < 2 for all fits. The obtained values for the isotope shift $\delta\nu^{102,A}$ are listed in Table I with total uncertainties between 3 and 9 MHz. These are typically dominated by statistical uncertainty but include systematic errors from the laser wavelength, high-voltage and scan-voltage readout, and A/B ratios.

The spectra of ^{113}Ru show only two peaks, which we attribute to the hfs of an $I = 1/2$ nuclear spin as the most likely solution. However, due to large statistical uncertainty and potential interference from isomeric states, we cannot categorically rule out other spin assignments. Additionally, we cannot distinguish between a hfs with a positive or negative magnetic moment, leading to two equally likely solutions for the isotope shift, which differ by ≈ 40 MHz. Consequently, both cases for $I = 1/2$ are presented separately in all tables and plots.

The isotope shift

$$\delta\nu^{A,A'} = \nu^{A'} - \nu^A = \frac{M_{A'} - M_A}{(M_{A'} + m_e)(M_A + m_e)} K_\infty + F\Lambda^{A,A'} \quad (1)$$

TABLE I. The measured isotope shift, differential, and absolute radii of neutron-rich ruthenium. In angle brackets, all uncorrelated statistical and systematic measurement uncertainties are given. The curly brackets denote the correlated uncertainty from the King-plot analysis procedure. For the absolute radii, the values in parentheses give their total uncertainty, including that of the reference isotope ^{102}Ru [53].

A	$\delta\nu^{102,A}$ (MHz)	$\delta\langle r_c^2 \rangle^{102,A}$ (fm ²)	R_c (fm)
102			4.4810 (20)
106	−666.7(3.5)	0.490 (2){13}	4.5353 (24)
107	−741.9(7.8)	0.567 (4){15}	4.5438 (26)
108	−905.6(3.3)	0.686 (2){17}	4.5569 (27)
109	−1001.8(8.8)	0.771 (4){20}	4.5662 (29)
110	−1053.6(3.3)	0.834 (2){26}	4.5731 (34)
111	−1112.5(5.6)	0.900 (3){33}	4.5803 (40)
112	−1190.1(3.5)	0.974 (2){38}	4.5884 (45)
113 ^a	−1245.3(4.4)	1.037 (2){46}	4.5953 (52)
113 ^b	−1204.4(4.4)	1.018 (2){49}	4.5932 (55)
114	−1282.2(6.0)	1.091 (3){55}	4.6011 (61)

^aEvaluated with positive magnetic moment.

^bEvaluated with negative magnetic moment.

originates from the difference in their nuclear masses M_A plus electron mass m_e , scaled with the mass shift parameter K_∞ , and their difference in nuclear size, given by the nuclear radius parameter $\Lambda^{A,A'}$, scaled with the field shift parameter F [54]. The nuclear radius parameter to first order can be expressed by the differential root-mean-square charge radius $\delta\langle r_c^2 \rangle^{A,A'}$ by subtracting 2.5% higher-order contributions [53] that cannot be resolved from isotope shifts in a single spectral line [55]. For this analysis, $\Lambda^{A,A'}$ is obtained from muonic atom spectroscopy of stable isotopes [53], and the corresponding isotope shifts from a complementary measurement at ATLANTIS, showing excellent agreement with previous experiments [51]. With this dataset, we performed a King-plot procedure [54], yielding $F = -2.13(46)$ GHz fm^{−2} and $K_\infty = 954(564)$ uGHz. Details can be found in the Supplemental Material [9]. The results allow us to interpolate and convert the isotope shifts of the newly measured radioactive isotopes to differential charge radii, which can be found in Table I. The uncertainty analysis treats the strongly correlated uncertainties of F and K_∞ appropriately. The King-plot interpolation introduces a correlated error to $\delta\langle r_c^2 \rangle^{102,A}$, denoted in curly brackets in the table, separated from the much smaller measurement uncertainty. The final step is to calculate the absolute charge radii R_c based on the stable reference isotope ^{102}Ru , $\langle r_c^2 \rangle_{102}^{1/2} = 4.481(2)$ fm [53].

Discussion—We compare the new data to the predictions of all models in the Brussels-Skyrme-on-a-grid series (BSkG1–BSkG4) that aim to provide nuclear data for astrophysical applications [56–58]. The BSkGx are models based on an energy density functional (EDF) of the Skyrme type; they can be treated as similar for the purposes of this

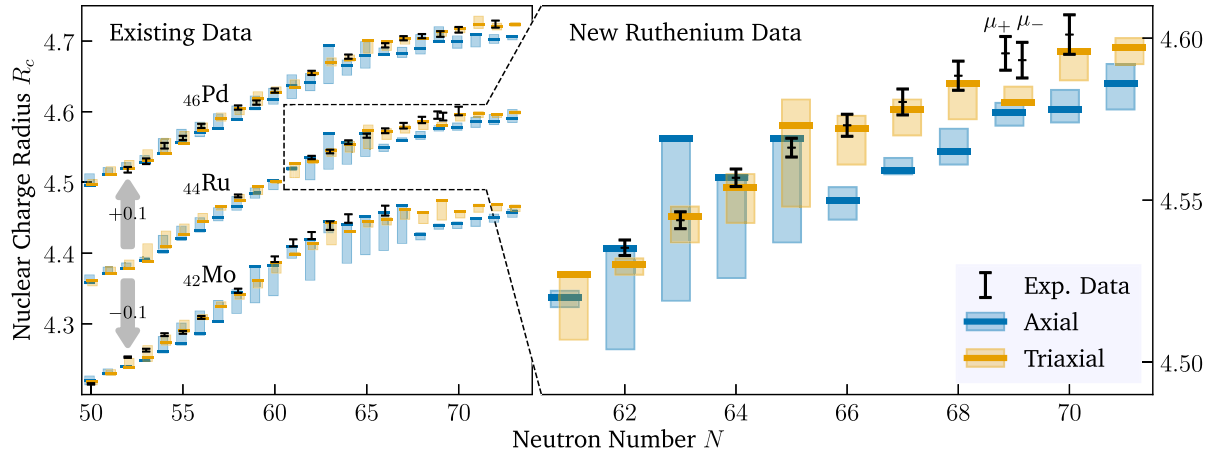


FIG. 2. Comparison between the experimental charge radii for the Ru, Mo [60], and Pd [59,61] isotopic chains, and BSkGx calculations with the inclusion of triaxial deformation (yellow) and without (blue, see text for details). The colored bands show the spread in predictions between all BSkGx, and the solid lines indicate the calculated BSkG4 values.

Letter because (i) they all are based on a three-dimensional numerical representation of the nucleus that naturally includes the possibility of triaxial deformation [10], (ii) their parameter adjustment protocols—involving data on thousands of nuclei—were nearly identical, and (iii) they—globally speaking—describe all known charge radii and the (scarce) Coulomb excitation data on triaxial deformation about equally well [8,32–34]. For example, all four models predict $\beta_2 \in [0.264, 0.278]$ and $\gamma \in [19^\circ, 23^\circ]$ for ^{104}Ru while experiment indicates $\beta_2 = 0.272(4)$, $\gamma = 23(3)^\circ$ [14]. They also perform equally well for other nearby stable nuclei with available Coulomb excitation data. Nevertheless, there are important modeling differences between the different BSkGx; this is why we take BSkG4—the most sophisticated entry—as a reference point and use the spread in the calculated charge radii as a proxy for the modeling uncertainty.

In Fig. 2, we compare two sets of calculations to the experimental data: one set that allows for triaxial deformation (in red) and another set (in blue) that is restricted to axially symmetric configurations with the lowest energy, i.e., either the prolate or the oblate saddle point for each nucleus. The uncertainties are derived from the spread of the four models. We emphasize that we compare *absolute* charge radii; the models require no systematic offset to match the experiment. Focusing first on the Ru isotopes, we see that the triaxial band closely follows the experimental data points across the whole isotopic chain within uncertainties. Only for one of the most exotic isotopes ($N = 69$) do all models significantly underestimate the experimental radii. The axially symmetric configuration performs similarly well for the lighter Ru isotopes but systematically underestimates the charge radii for $N \geq 66$. The axial results also feature a much broader model spread between $62 \leq N \leq 65$: there, the calculated charge radii are affected by the transition from prolate to oblate saddle point that

occurs at different neutron numbers for each model. The unrestricted, i.e. triaxial, BSkGx predictions are clearly better than those from axially symmetric calculations. To show that both the level of agreement and the difference between triaxial and axial calculations is not limited to one isotopic chain, we also compare calculated and measured charge radii for the Mo and Pd chains [59]. Most of the literature does not consider any possible impact of triaxial deformation on charge radii, even for nuclei that are known to have such asymmetric ground states [59]. We presume that the (mostly unstated) motivation to neglect this degree of freedom is that of Ref. [20], which states “the isotope shift is very insensitive to triaxial shape.”

This argument is based on the observation that the rms radius of a triaxial quadrupole-deformed liquid drop [62] is almost entirely determined by the size of the quadrupole deformation β_2 and only marginally sensitive to the triaxiality angle γ . From this, however, it does not follow that triaxial deformation does not play a role for charge radii. A nucleus’s energetically most favorable shape is primarily determined by quantum mechanical shell effects [63]. Since these vary strongly with deformation, changing γ will force a rearrangement of the occupation numbers of the filled single-particle levels and their spatial wave functions and thus induce further changes to the spatial density distribution. First, the energetically optimal β_2 will usually not remain fixed when varying γ . The total quadrupole deformation of a triaxial ground state will typically not match that of the axially symmetric prolate or oblate configuration; even if γ would not influence R_c much by itself, the change in β_2 will. Second, any change of β_2 or γ will induce a change in higher-order moments of the nucleus. Even if often omitted from analyses, these do contribute to radii in liquid drop and EDF-based models alike. For instance, the effect of octupole deformation on the charge radii of Ac isotopes was analyzed in Ref. [64].

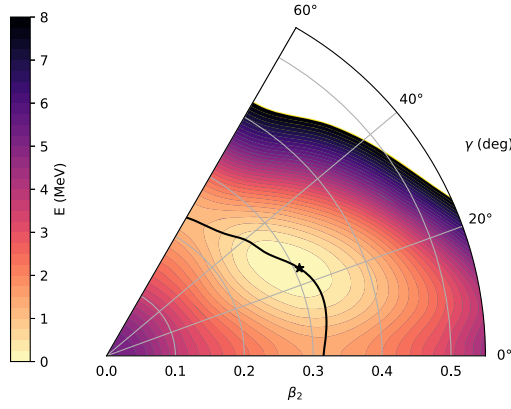


FIG. 3. Normalized total energy of ^{112}Ru obtained with BSkG4 as a function of β_2 and gamma. The black line indicates the minimal energy as a function of γ ; the global energy minimum is indicated by a black star.

Third, in an EDF calculation, the features of the diffuse radial profile of the charge density also change with β_2 and γ ; already a small radial redistribution of nucleons far from the nuclear center can then have a sizable effect on the calculated charge radius. While the global effect of nuclear diffuseness is sometimes included in the liquid drop analysis of charge radii [65,66], its change with deformation cannot be treated in such models.

We can illustrate the consequences of these effects for ^{112}Ru using BSkG4 [67]: Figures 3 and 4 show the system's total energy and charge radius, respectively, as a function of β_2 and γ . The black lines indicate the total deformation that minimizes the energy as a function of the triaxiality angle, illustrating that β_2 varies in a nontrivial way along this path. In particular, the values of β_2 at the lowest prolate and oblate axial configurations and at the triaxial minimum are quite different. Even more important is that R_c has a visible γ dependence. At fixed β_2 , the calculated R_c increases with γ by more than 0.03 fm. This is the opposite of what is expected for a purely quadrupole-deformed liquid drop: at $\beta_2 \simeq 0.3$, this model predicts a decrease by about 0.01 fm when going from prolate to oblate shape. Part of the difference between the two models can be attributed to an increase of higher-order deformations with γ in the EDF model and the other part results from effects not captured by the liquid drop model.

The details of shell effects vary from one model to the next, which naturally means that the effect of triaxial deformation on the nuclear charge radius becomes model dependent, as demonstrated in the Supplemental Material [9]. Even if this were not the case, a single value for the charge radius cannot determine a unique point on the (β_2, γ) plane. Measuring R_c will not allow us to determine whether or not a given nucleus exhibits triaxial deformation. This does not mean the effect is unimportant: Figure 2 shows that calculations that account for this degree of freedom perform markedly better than those restricted to

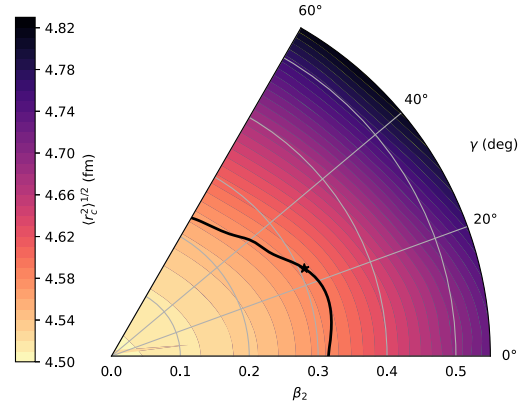


FIG. 4. Root-mean-square charge radius $R_c = \langle r_c^2 \rangle^{1/2}$ of ^{112}Ru across the β - γ plane, as calculated for BSkG4. The black line and black star are as in Fig. 3.

axial symmetry—the latter also disagree with the available spectroscopic information for these isotopes. When taken together, all of these considerations paint a coherent picture of ground state triaxial deformation; it is only by accounting for this deformation that the BSkG models arrive at their excellent and simultaneous description of masses [35,36], spectroscopy [37], and charge radii in this region of the nuclear chart.

Conclusions—The installation of the new ATLANTIS beamline allowed us to perform isotope shift measurements of neutron-rich ruthenium produced from ^{252}Cf fission at CARIBU. The highly efficient RFQ cooler and buncher and a magnesium-based charge-exchange cell compensated for the weak fission rate. We extracted the differential rms charge radii of the nine radioactive isotopes $^{106-114}\text{Ru}$ from the isotope shifts.

There is significant evidence that these isotopes of Ru are triaxial, and only the inclusion of triaxial deformation into the calculations allows us to describe the experimental data while being consistent with all known information about the nuclei in question. The deformation impacts calculated charge radii through shell effects, contrary to what is expected from liquid drop modeling.

Earlier studies with the BSkG series already pointed to the relevance of triaxial deformation for the behavior of binding energies and isomeric states in this region [35–37]. Here, we show that this combined description extends to charge radii: the BSkGx describe the new data remarkably well. A consistent picture is emerging: triaxial deformation leaves a fingerprint on essentially all properties of nuclei in this region of the nuclear chart; our Letter illustrates how spontaneous symmetry breaking in strongly correlated systems not only impacts spectroscopic properties but also bulk observables.

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Data availability—The measured isotope shifts and all details necessary to extract (differential) charge radii are available within the Letter. Raw data to extract isotope shifts from measured optical spectra 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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