

Nuclear charge radii of Sr isotopes: Reevaluation based on transition frequency measurements in the $5s-5p-4d$ manifold in Sr^+

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High-precision quasisimultaneous collinear/anticollinear laser spectroscopy was performed to measure the $5s\ ^2S_{1/2} \rightarrow 5p\ ^2P_{1/2}$ ($D1$), the $5s\ ^2S_{1/2} \rightarrow 5p\ ^2P_{3/2}$ ($D2$), and the three $4d \rightarrow 5p$ transitions in naturally abundant Sr^+ isotopes. For absolute transition frequencies, an uncertainty as low as 600 kHz was achieved, while common-mode rejection allowed us to extract isotope shifts with uncertainties down to a level of 200 kHz, 1 order of magnitude better than previously achieved. Similarly, the uncertainties of the hyperfine-structure coefficients for ^{87}Sr of the $5p$ states and the $4d\ ^2D_{3/2}$ levels are improved. A King-plot analysis yielded a field-shift ratio of the $D2$ and $D1$ lines of $F_{D2}/F_{D1} = 1.004(5)$, which lies within the theoretically allowed region and can be used as a benchmark for atomic structure theory calculations. We use the information from all stable isotopes in the investigated transitions to compare field-shift and mass-shift constants obtained by various techniques regularly used in the literature, ranging from King plots with purely experimental input to *ab initio* atomic structure calculations by state-of-the-art theory. We show that in the region above $N = 50$, the charge radii are strongly dependent on the approach being used.

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I. INTRODUCTION

Alkaline atoms and singly charged alkaline-earth ions [1] have a single valence electron and no open subshells. Therefore, they have long been used as benchmarks for atomic structure calculations [2]. Transition frequencies, isotope shifts (IS), and hyperfine-structure (HFS) splittings are important observables of these systems since they can be measured with high precision and are used to extract, e.g., information about nuclear structure [3,4]. Isotope shifts have provided nuclear charge radii for stable and short-lived isotopes of all alkaline [5–16] and alkaline-earth elements [17–26], HFS splittings contain information on the nuclear spin [27–29], the nuclear magnetic dipole moment [27,30–32], and the nuclear electric quadrupole moments [33].

Extracting nuclear observables from isotope shifts and hyperfine splittings requires electronic factors obtained from

atomic-structure calculations. The reliability of the resulting nuclear charge radii and electromagnetic moments therefore depends directly on the accuracy of these calculations.

Singly charged alkaline-earth elements provide an opportunity to test the calculated field- and mass-shift factors experimentally. Mg, Ca, Sr, and Ba each have at least three stable isotopes for which independent nuclear charge-radius information is available from muonic-atom spectroscopy and elastic electron scattering. These data enable a King-plot determination of the mass- and field-shift factors, which can be compared directly with atomic-structure calculations [34]. For alkaline elements, in contrast, the small number of stable isotopes generally precludes such a determination, and the extraction of their nuclear charge radii relies more strongly on calculated atomic factors [35]. Their isoelectronic alkaline-earth ions therefore serve as valuable benchmark systems for assessing these calculations.

Reliable atomic factors and well-founded uncertainties are important not only for the extraction of nuclear charge radii, which is receiving increasing attention across several fields [36], but also for precision tests of the standard model. In particular, the interpretation of atomic parity-nonconservation measurements in Cs in terms of the weak nuclear charge and the Weinberg angle is limited by the accuracy of the underlying atomic-structure calculations [37–45].

Ca^+ ions served as a first system to search for new bosons [46] that mediate an interaction between electrons and neutrons beyond standard model physics—a field that has recently gained much interest in theory and experiment

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(see, e.g., Ref. [47]). Similar theoretical and experimental investigations are now proposed for transitions in Sr^+ ions [48] and have already been carried out in Sr atoms [49]. Furthermore, transitions in Sr^+ ions serve as atomic-clock candidates [50] and for quantum computing [51]. Moreover, spectroscopy on Sr^+ in an ion trap has recently been proposed for environmental studies to detect the radiotoxic isotope ^{90}Sr [52,53] and to surpass previously used optical detection techniques [54–56]. The transition frequencies, isotope shifts, and hyperfine structure measurements reported here are also of importance for these endeavors.

Furthermore, the influence of the relativistic $S_{1/2}$ contribution to the small component of the $P_{1/2}$ wavefunction has previously been identified through a deviation in the field-shift ratio of the $D1$ and $D2$ transitions [57] in the spectroscopy of Ba^+ ions [58,59]. A corresponding high-precision measurement in Ca^+ led to a surprisingly large ratio that could not be explained by theory [60] but was later clarified in a new measurement that was in agreement with theory and consistent with measurements of the isotope shift in the $4s \rightarrow 3d$ transitions [61]. Our measurements provide the first determination of the size of this effect in Sr.

The structure of this paper is as follows: After a short description of the experimental setup and method in Sec. II, we present the measurements and analysis of transition frequencies, IS, and HFS in the $5s^2S_{1/2} \rightarrow 5p^2P_{1/2,3/2}$ ($D1$, $D2$)—including the field-shift ratio—and the $4d^2D_{3/2,5/2} \rightarrow 5p^2P_{1/2,3/2}$ transitions in Sr^+ ions in Secs. III and IV. The hyperfine-structure splittings provide a benchmark test of state-of-the-art relativistic coupled-cluster calculations. Section V is devoted to the determination of the isotope-shift parameters, which are then used in Sec. VI to extract a consistent set of charge radii for the stable and short-lived isotopes.

II. METHOD

The isotope shift

$$\delta v_i^{A,A'} \equiv v_i^A - v_i^{A'} \approx \frac{K_i}{\mu^{A,A'}} + F_i \delta \langle r_c^2 \rangle^{A,A'}, \quad (1)$$

$$\text{with } \mu^{A,A'} = \frac{M_A M_{A'}}{M_A - M_{A'}}, \quad (2)$$

between two isotopes A and A' arises from the change in the atomic masses M and the change in the mean-square nuclear charge radius $\delta \langle r_c^2 \rangle^{A,A'}$. Here, K_i and $F_i = -Ze^2 \Delta |\Psi(0)|_i^2 / (6\hbar\epsilon_0)$ denote the mass-shift and field-shift factors of transition i , with $\Delta |\Psi(0)|_i^2$ the transition-specific change in the electron density at the nucleus.

After multiplication with the modified mass factor $\mu^{A,A'}$, one obtains a linear relation between the mass-modified isotope shift and the mass-modified change in charge radius,

$$\mu^{A,A'} \delta v_i^{A,A'} \approx K_i + F_i \mu^{A,A'} \delta \langle r_c^2 \rangle^{A,A'}. \quad (3)$$

If $\delta \langle r_c^2 \rangle^{A,A'}$ is known from independent methods, this representation allows for a direct determination of the atomic factors K_i and F_i (see Sec. V). Conversely, once these factors are known, Eq. (3) can be used to improve the charge-radius information of isotopes with known radii or to extract the charge radii of

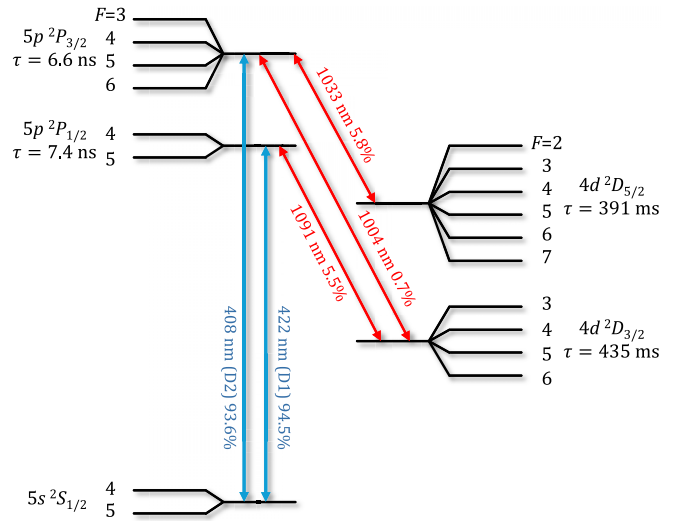


FIG. 1. Level scheme and transitions of Sr^+ ions relevant for this work. The hyperfine substates of ^{87}Sr are indicated.

other, in particular, short-lived isotopes, as described in detail in Ref. [62] and applied here in Sec. VI.

Alternatively, by measuring the isotope shifts for two transitions i and j , the change in the mean-square nuclear charge radius in Eq. (1) can be eliminated, yielding

$$\mu^{A,A'} \delta v_i^{A,A'} \approx K_i - \frac{F_i}{F_j} K_j + \frac{F_i}{F_j} \mu^{A,A'} \delta v_j^{A,A'}. \quad (4)$$

This is a linear relation between the measured mass-modified isotope shifts, with a slope given by the ratio of the field-shift factors, F_i/F_j , of the two transitions, the so-called King plot. Since this ratio can be accessed through calculations of the electronic wavefunctions at the nucleus for all involved states, it provides a benchmark for comparing atomic-structure theory with experiment [59–61,63]. This is particularly feasible in systems with a single valence electron outside closed shells, such as Sr^+ , since such systems are well suited for coupled-cluster calculations, which are among the most accurate methods in atomic-structure theory.

Strontium has four naturally abundant isotopes, namely, ^{88}Sr , ^{86}Sr , and ^{84}Sr . Challenges for laser spectroscopy are the comparatively low abundance of 0.56(2)% in the case of ^{84}Sr and the nuclear spin of $I = \frac{9}{2}$ for ^{87}Sr , which causes hyperfine splitting with only partially resolved lines in some of the transitions. The ground state of Sr^+ is the $5s^2S_{1/2}$ level with a single valence electron outside of the closed krypton core. Two dipole transitions at 408 nm ($D1$) and 422 nm ($D2$) lead to the $5p^2P_{1/2}$ (23 715.19 cm^{-1}) and $5p^2P_{3/2}$ (24 516.65 cm^{-1}) levels with lifetimes of a few nanoseconds [64]. Both levels decay predominantly back to the ground state but have weak branches into the metastable $4d^2D_{3/2,5/2}$ levels (14 555.90 and 14 836.24 cm^{-1}) [64]. Once populated, the electron in the $4d$ levels can be excited by one of three transitions at 1004, 1032, or 1091 nm into the $5p$ manifold as shown in Fig. 1.

In this work, collinear laser spectroscopy (CLS) was chosen as it can determine isotope shifts in allowed dipole transitions to better than 1% of their natural linewidth [59,61]. In CLS, an ion beam with energy of a few 10 keV is

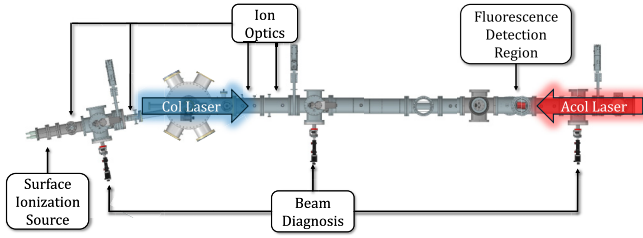


FIG. 2. A schematic of the updated COALA beamline at TU Darmstadt. In this experiment, the Sr^+ ions are created in the surface ionization source. Ion optics and three beam diagnostic stations equipped with Faraday cups and multichannel plates allow superimposing the ion and laser beams in the fluorescence detection region.

superimposed either collinearly (c) or anticollinearly (a) with the laser. The electrostatic acceleration out of an ion source leads to a strong compression of the thermal velocity distribution in the direction of flight and therefore strongly reduces the Doppler broadening [65]. However, the collinear geometry also leads to maximal Doppler shifts

$$v_{c/a} = v_0 \gamma (1 \pm \beta), \quad (5)$$

where v_0 is the transition frequency in the rest frame of the ion, $v_{c/a}$ is the laser frequency in the laboratory frame for collinear or anticollinear geometry, $\gamma = 1/\sqrt{1 - \beta^2}$, and $\beta = v/c$ is the velocity of the ions in the laboratory frame divided by the speed of light. Instead of scanning the laser frequency, the ion velocity can be scanned, which varies the effective Doppler shift, to obtain spectra by scanning an additional applied voltage U_{scan} to the interaction region. Another advantage of the collinear technique in the case of the Sr isotopes is a clear separation of isotope-specific peaks in the spectra due to the mass and, hence, velocity differences, while the natural isotope shifts are very small due to the large cancellation of field-shift and mass-shift contributions in this mass region.

Absolute transition frequencies can be determined velocity-independently by performing measurements in collinear and anticollinear direction (quasi)simultaneously,

$$v_0 = \sqrt{v_c v_a} = \sqrt{v_0 \gamma (1 + \beta) v_0 \gamma (1 - \beta)}. \quad (6)$$

Performing those measurements in alternating order, namely, anticollinear-collinear-collinear-anticollinear, cancels contributions from a drifting acceleration potential and will be further referenced as *acca* measurements.

III. EXPERIMENT

The experiment was performed at the Collinear Apparatus for Laser Spectroscopy and Applied Science (COALA) shown in Fig. 2. An extensive description of the COALA beamline can be found in Ref. [66] with a recent update provided in Ref. [67]. Similar measurements were previously reported on Ca^+ [61] and Ba^+ [59]. Three main components need to be precisely controlled to reach an accuracy of the order of 100 kHz: the ion velocity, the laser frequency, and the overlap of the laser beams. The velocity of the ions inside the fluorescence detection region (FDR) only depends on the initial starting potential U_{start} and the scanning voltage U_{scan} applied to the FDR. The starting potential of approximately 20 kV was measured

via a custom high-voltage divider [68] with a divider ratio of about 2000:1 and variations of ≤ 0.2 ppm on the timescale of a measurement. The voltage of about 10 V across the precision resistor of the chain was measured with a 8.5 digit multimeter (Keysight 3458A), which controls a feedback loop consisting of a custom digital-to-analog converter to stabilize the starting potential, improving the stability of the beam velocity by more than 1 order of magnitude [68]. The scanning voltage was provided by a ± 10 V 18-bit digital-to-analog converter (Analog Devices, AD5781), which was amplified by a factor of 50 with a Kepco BOP500M linear bipolar amplifier. Recently, we have quantified the influence of field penetrations into the FDR [68]. In *acca* measurements of the even-even isotopes, this has been circumvented by choosing both laser frequencies such that the resonances occur at the same scanning voltage, ideally at very low voltages, as the field penetration effects scale proportional to the applied voltage. For wider spectra, such as the hyperfine structure of ^{87}Sr , this effect resulted in a relative uncertainty of the peak spacings of about 40 ppm.

Laser light for the $4d^2 D_{3/2,5/2} \rightarrow 5p^2 P_{3/2}$ transitions (1004 and 1033 nm) was produced by two Matisse 2 TS Ti:Sapphire ring lasers, each pumped by a Millennia eV diode-pumped Nd:YVO₄ laser. For the D_1 (422 nm) and D_2 (408 nm) transitions, the output of the Ti:Sapphire lasers was frequency doubled with a WaveTrain 2 using lithium triborate crystals. Both Matisse lasers were frequency stabilized using an FC1500-250-WG frequency comb with a global positioning system (GPS)-disciplined quartz oscillator and frequency drifts of the lasers were compensated by adjusting their reference-cavity lengths. This reduces the frequency uncertainty and drifts typically down to the level of 30 kHz [66], limited by the short-term stability of the Matisse.

For the $4d^2 D_{3/2} \rightarrow 5p^2 P_{1/2}$ transition at 1091 nm a tuneable diode laser of type Toptica DL Pro was stabilized to a HighFinesse WSU30 wavemeter. The wavemeter was calibrated with a stable He:Ne laser, allowing an absolute frequency determination with a 1σ uncertainty of 10 MHz.

The laser light was transported to the experiment via single-mode optical fibers, providing collimated beams with Gaussian profiles and FWHM diameters of approximately 1 mm, while reducing the vibrations observed with free-space beam transport. Laser powers of approximately 5 and 600 μW were used for the $5s \rightarrow 5p$ and $4d \rightarrow 5p$ transitions, respectively, corresponding to peak intensities of approximately 0.44 and 53 mW/cm^2 . Based on the natural linewidths and branching fractions [69], the estimated saturation intensities are approximately 38 and 47 mW/cm^2 for the D_1 and D_2 transitions, respectively, and approximately 37, 490, and 51 mW/cm^2 for the $D_{3/2} \rightarrow P_{1/2}$, $D_{3/2} \rightarrow P_{3/2}$, and $D_{5/2} \rightarrow P_{3/2}$ transitions. These estimates neglect transition-specific polarization and magnetic-sublevel factors. For the D_1 and D_2 transitions, measurements were performed at several laser powers, and the operating power was chosen to provide sufficient signal-to-noise ratio while minimizing the uncertainty of the fitted line position. The selected intensity was less than 2% of the estimated saturation intensities, such that power broadening is expected to be negligible. In contrast, the intensity used for the infrared transitions is comparable to the estimated saturation intensities of the $D_{3/2} \rightarrow P_{1/2}$ and $D_{5/2} \rightarrow P_{3/2}$ transitions and may therefore result in notice-

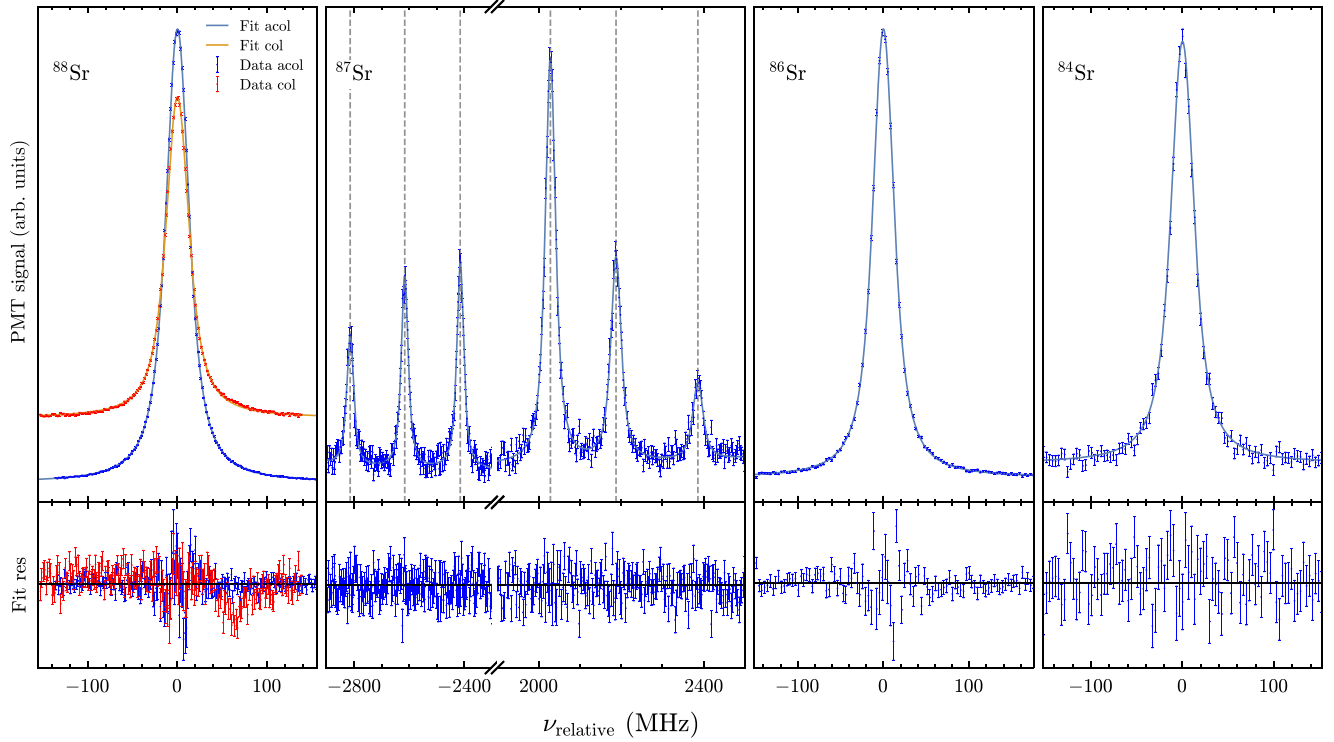


FIG. 3. Experimental spectra of the $5s\ ^2S_{1/2} \rightarrow 5p\ ^2P_{3/2}$ transition (D2) in $^{88-86}\text{Sr}$ and ^{84}Sr fitted with Voigt profiles to extract the transition frequency, the isotope shifts, and the hyperfine structure A and B parameters of the upper and lower states of the transition. For ^{88}Sr , the spectra obtained in collinear (red) and anticollinear geometry (blue) are shown, whereas in the other cases only an anticollinear spectrum is depicted. For ^{87}Sr , the position of each hyperfine peak is marked with a dashed line.

able power broadening. The higher infrared laser power was required to compensate for the small population of the metastable $4d$ states.

The overlap of the two laser beams was optimized by coupling both beams through the beamline into the fiber of the counterpropagating laser and maximizing the transmitted power. Given the 6.5 m distance between the two fiber couplers and beam-spot diameters of approximately 2 mm, a maximum angular deviation of 0.15 mrad is estimated. Its effect on the measured absolute transition frequency depends on the transition wavelength but is typically on the order of 100 kHz. The overlap of the ion beam with the laser beams was ensured using iris apertures distributed along the beamline, through which both the transmitted laser power and ion current were maximized, limiting angular deviations to approximately 1.2 mrad. With the optimized counterpropagating geometry of the two laser beams, the corresponding effect is negligible, at approximately 10 kHz.

Similar as for the $3d$ levels of Ca^+ ions in previous work [70], we found the metastable $4d$ levels sufficiently populated in the surface ionization source. This is most probably caused by inelastic collisions with atoms during the acceleration out of the ion source, since the population of the d states was further enhanced with increasing background pressure once the turbo-molecular pump nearest to the ion source was turned off. The signal was sufficient to record resonances of the $4d \rightarrow 5p$ transitions on a timescale of minutes. The detection of the blue photons emitted in the dominant $5p \rightarrow 5s$ decay following the $4d \rightarrow 5p$ excitation is almost background free

since the photomultiplier tubes are not sensitive to the infrared laser light.

IV. RESULTS

A. Experimental spectra of Sr^+ transitions

Spectra of ^{88}Sr , ^{87}Sr , and ^{84}Sr frequency measurements in the $D2$ transition are depicted in Fig. 3 as an example for all isotopes in the $D1$ and $D2$ transitions. For the most naturally abundant isotope $^{88}\text{Sr}^+$, spectra in collinear and anticollinear geometry are included in Fig. 3, while the others are restricted to the anticollinear case for clarity reasons. The two triplets in the HFS of $^{87}\text{Sr}^+$ are separated by more than 4 GHz due to the large hyperfine splitting in the $5s$ ground state. The spectrum of the least abundant isotope ^{84}Sr (0.56%) provides an impression of the data quality. All peaks were fitted with symmetric Voigt profiles. The Lorentzian component dominates, yielding linewidths of $\Gamma_{D1} = 21.5(3)$ MHz and $\Gamma_{D2} = 24.1(2)$ MHz, both close to the previously reported natural linewidths of $\Gamma_{\text{nat},D1} = 20.4(2)$ MHz [71] and $\Gamma_{\text{nat},D2} = 22.7(3)$ MHz [69]. Given the low saturation parameters discussed above, the slight excess over the natural linewidths cannot be fully explained by power broadening. The Gaussian contribution of approximately $\sigma = 5(1)$ MHz for both transitions is comparatively small and represents the residual inhomogeneous broadening in the experiment. Nevertheless, small systematic structures remain visible in the fit residuals of ^{88}Sr , suggesting that the measured line shape is not perfectly described by a symmetric Voigt profile. However, the influence of these small

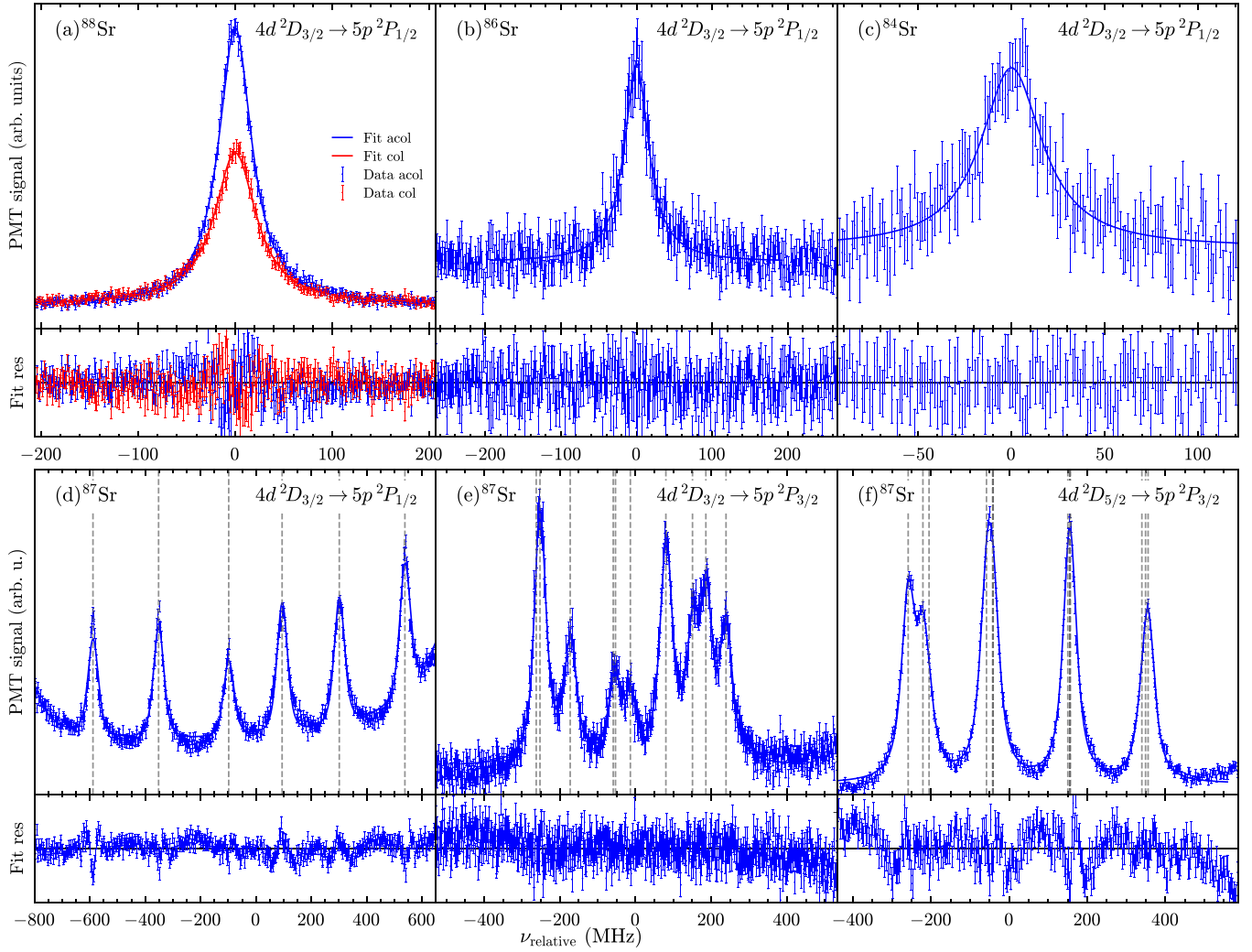


FIG. 4. Experimental spectra of the $4d^2D \rightarrow 5p^2P$ transitions with symmetric Voigt-profile fits. As an example, a spectrum for both geometries for the $4d^2D_{3/2} \rightarrow 5p^2P_{3/2}$ transition is given for ^{88}Sr in panel (a). In panels (b) and (c), just the anticollinear spectra are shown for ^{86}Sr and ^{84}Sr , respectively. In panels (d)–(f), the ^{87}Sr hyperfine spectra of the different transitions are shown, with each individual hyperfine peak position marked by a dashed line. While for the $4d^2D_{3/2} \rightarrow 5p^2P_{1/2}$ transition, the six hyperfine peaks are well separated in panel (d), the ten peaks for the $4d^2D_{3/2} \rightarrow 5p^2P_{3/2}$ transition in panel (e) are more difficult to resolve. Owing to the small hyperfine splittings, the assignment of the 12 peaks of the $4d^2D_{5/2} \rightarrow 5p^2P_{3/2}$ transition in panel (f) was guided by the previously measured hyperfine constants of the $5s^2S_{1/2} \rightarrow 4d^2D_{5/2}$ transition [79]. In those transitions of ^{87}Sr , the background is distorted by the nearby peaks of ^{88}Sr and ^{86}Sr , which were only included in the scan and fit for the $4d^2D_{3/2} \rightarrow 5p^2P_{1/2}$ transition. For the remaining two transitions, the effect is visible in the residuals.

line-shape asymmetries largely cancels when combining the collinear and anticollinear measurements and therefore does not significantly affect the determined rest-frame transition frequency [72].

To investigate this possibility, a more sophisticated line-shape model accounting for the thermal ion velocity distribution in the source [61,73] was benchmarked on a test set comprising more than 200 spectra using the SciPy package [74]. However, compared to the symmetric Voigt profile, this model increased the average reduced χ^2 from 1.2 to 1.6 and was therefore not adopted for the final analysis.

Besides the acca measurements, in which the collinear and anticollinear resonances of one isotope were measured sequentially, we employed a second scheme referred to as “direct IS.” Here, the laser was kept at a fixed frequency while the voltage applied to the fluorescence detection region was

scanned over a range of up to 1 kV. This allowed resonances of several isotopes to be recorded within the same voltage scan and their isotope shifts to be determined directly from the relative peak positions. In this approach, constant frequency offsets largely cancel in the extracted isotope shifts. The small residual dependence on the laser frequency, caused by the slightly different Doppler factors of the isotopes, was included in the uncertainty determination. Possible frequency changes during the scan and uncertainties of the applied scanning voltage were treated separately.

For the infrared $4d \rightarrow 5p$ transitions, representative spectra of all stable even isotopes in the $4D_{3/2} \rightarrow 5P_{3/2}$ transition are shown in Figs. 4(a)–4(c). The hyperfine structure of ^{87}Sr is in some transitions only partially resolved and the corresponding spectra are depicted in Figs. 4(d)–4(f). To compensate for the lower statistics caused by the small

TABLE I. Transition frequencies of all investigated transitions for all stable isotopes in megahertz.

A	ν_{D1}^A	ν_{D2}^A	$\nu_{D_{3/2} \rightarrow P_{1/2}}^A$	$\nu_{D_{3/2} \rightarrow P_{3/2}}^A$	$\nu_{D_{5/2} \rightarrow P_{3/2}}^A$
88	710 962 838.3(6)	734 989 824.5(6)	274 589 143.7(11) ^a	298 616 129.9(6)	290 210 780.4(6)
87	710 962 781.1(6)	734 989 768.4(6)	274 589 334.9(15) ^a	298 616 322.0(13) ^b	290 210 970.0(23) ^b
86	710 962 666.6(6)	734 989 653.5(6)	274 589 543.8(10) ^a	298 616 530.7(6)	290 211 179.3(6)
84	710 962 462.8(6)	734 989 449.5(6)	274 589 968.8(15) ^a	298 616 955.4(13) ^b	290 211 603.0(15) ^b

^aCalculated according to Eq. (11).^bCalculated from frequency comb supported acca measurement for ⁸⁸Sr plus isotope shift: $\nu_i^A = \nu_i^{88} + \delta\nu_i^{A,88}$.

population of the d states, the ion-source temperature was increased, resulting in broader observed resonance profiles. Depending on the heating current applied to the source, the measured linewidths range from 30 to 50 MHz, remaining on the order of the natural linewidths: $\Gamma_{\text{nat}}(4D_{5/2} \rightarrow 5P_{3/2}) = 25.2(4)$ MHz [75,76], $\Gamma_{\text{nat}}(4D_{3/2} \rightarrow 5P_{3/2}) = 25.2(4)$ MHz [75,77], and $\Gamma_{\text{nat}}(4D_{3/2} \rightarrow 5P_{1/2}) = 20.2(3)$ MHz [75,77]. The dependence of the observed linewidth on the source-heating current supports the interpretation that the small excess linewidth observed in the $D1$ and $D2$ transitions also originates predominantly from the ion source. Although the fitted Gaussian contribution in those measurements is only approximately $\sigma = 5(1)$ MHz, an asymmetric source-induced velocity distribution is not fully represented by the symmetric Voigt model and may therefore also contribute to the fitted Lorentzian width.

B. Combination of results

Measurements of the same observables (transition frequencies, isotope shifts, and hyperfine parameters) performed on different days or with different measurement schemes were first averaged within the respective set of measurements and then compared, searching for indications of systematic differences. Therefore, the weighted average

$$X = \frac{\sum_i x_i \sigma_i^{-2}}{\sum_i \sigma_i^{-2}} \quad (7)$$

within each set was calculated. Its uncertainty was obtained as the maximum of the estimator for the standard error of the weighted average and the standard error of the mean for the respective number of measurements:

$$\Delta X = \max \left((\sum_i \sigma_i^{-2})^{-1/2}, (\sum_i (x_i - \bar{x})^2 / N) / \sqrt{N} \right). \quad (8)$$

We observed jumps of the measured absolute transition frequencies in the range of a few hundred kilohertz at different days, which were traced back to the frequency comb. At the time of these measurements, the frequency-comb's femtosecond laser amplifier was suffering from an aging pumping diode, resulting in low output power that led to limited beat signals with the spectroscopy laser. In our case, we found that these limited beat signals caused a systematically lower rate of about 100 kHz for the frequency counter that evaluated the beat frequency. This can lead to a higher or lower frequency readout depending on the relative frequency difference of both lasers. Once set up, the beat intensity stayed constant during the day, explaining the day to day jumps. To validate the

frequency scale, the $4s^2S_{1/2} \rightarrow 4p^2P_{1/2}$ (397 nm) transition in Ca^+ was measured using the same surface ionization source. No additional offset beyond the observed jumps was found [61]. Since these jumps were observed only between measurements performed on different days, they affect the absolute transition frequencies but are irrelevant for isotope shifts obtained from transition-frequency differences measured on the same day. In this case, the common day-dependent frequency offset cancels in the difference. We report the measured transition frequencies in Table I and add a conservative systematic contribution of 600 kHz to the absolute frequency uncertainty, covering the full span of the observed frequencies.

No other systematic differences between the different sets were observed or exceeded 2σ of the combined uncertainties.

C. Transition frequencies

The transition frequencies obtained from our measurements are listed in Table I. For the $D_{3/2} \rightarrow P_{3/2}$ and $D_{5/2} \rightarrow P_{3/2}$ transitions, frequency-comb-supported acca measurements were performed only for ⁸⁸Sr and ⁸⁶Sr. For the $D_{3/2} \rightarrow P_{3/2}$ transition, precise transition frequencies of ⁸⁷Sr and ⁸⁴Sr were determined by combining the directly measured isotope shifts with the frequency-comb-referenced rest-frame frequency of ⁸⁸Sr. For the $D_{5/2} \rightarrow P_{3/2}$ transition, the isotope shifts were obtained from wavemeter-referenced measurements of all stable isotopes performed with a diode laser. These isotope shifts were then combined with the frequency-comb-referenced rest-frame frequency of ⁸⁸Sr measured separately using the Matisse laser. In both cases, the transition frequency of isotope A is given by

$$\nu_i^A = \nu_i^{88} + \delta\nu_i^{A,88}. \quad (9)$$

For statistically independent quantities, the corresponding uncertainty is

$$\sigma^2(\nu_i^A) = \sigma^2(\nu_i^{88}) + \sigma^2(\delta\nu_i^{A,88}). \quad (10)$$

For the $D_{5/2} \rightarrow P_{3/2}$ transition, combining the diode-laser isotope shifts with the Matisse-based frequency-comb measurement yields absolute transition frequencies with uncertainties of approximately 1–2 MHz. For the remaining $D_{3/2} \rightarrow P_{1/2}$ transition, the laser wavelength of 1091 nm exceeds the spectral range of the frequency comb and from previous investigations with our HighFinesse WSU30 we also expect the deviations in this range to be larger than the specified 10 MHz 1σ uncertainty [78]. Thus, we determined the

TABLE II. Extracted $5s^2S_{1/2} \rightarrow 4d^2D_{3/2,5/2}$ frequencies from the measured $S_J \rightarrow P_J$ and $D_J \rightarrow P_J$ transitions for all stable isotopes in megahertz.

A	$\nu_{S_{1/2} \rightarrow D_{3/2}}^A$	$\nu_{S_{1/2} \rightarrow D_{5/2}}^A$	Ref.
88	436 373 694.6(9)	444 779 044.0(8)	[82]
		444 779 044.095 485(19)	
87	436 373 446.4(14)	444 778 798.4(24)	[79]
		444 778 796.11(4)	
86	436 373 122.8(9)	444 778 474.2(9)	[81]
		444 778 473.814(4)	
84	436 372 494.1(14)	444 777 846.5(17)	

transition frequencies via

$$\nu_{D_{3/2} \rightarrow P_{1/2}}^A = \nu_{D1}^A - (\nu_{D2}^A - \nu_{D_{3/2} \rightarrow P_{3/2}}^A). \quad (11)$$

The uncertainties of this method are approximately 1 order of magnitude smaller compared to the wavemeter-based acca transition frequency measurements.

From the measured frequencies, we can also obtain transition frequencies for the $S_{1/2} \rightarrow D_{3/2,5/2}$ transitions in all isotopes. The results are listed in Table II and are in excellent agreement with literature values [50,80,81]. Precise frequencies in the $5S_{1/2} \rightarrow 5D_{3/2}$ transition have—to our knowledge—not been reported so far.

D. Isotope shifts

Since multiple measurement schemes were used to extract the same observables, the results were combined into a single value. The final isotope shifts are listed in Table III. For the $D1$, $D2$, and $4D_{3/2} \rightarrow 5P_{3/2}$ transition, the isotope shifts obtained by subtracting the absolute transition frequencies were unified with the results of the direct IS measurements. Agreement is generally within 1σ , with the only exception (2.5σ) being ^{84}Sr in the $4D_{3/2} \rightarrow 5P_{3/2}$ transition, where we have the least statistics. For the $4D_{3/2} \rightarrow 5P_{1/2}$ and $4D_{5/2} \rightarrow 5P_{3/2}$ transitions, only acca measurements were performed. Since, apart from the $^{88,86}\text{Sr}$ pair for the $4D_{5/2} \rightarrow 5P_{3/2}$ transition, we only have acca measurements with the wavemeter, they have the largest uncertainty.

The only isotope shift measurements that are known to us in the D - P manifold are those of $\delta\nu^{86,88}$ and $\delta\nu^{84,88}$ in the $4D_{3/2} \rightarrow 5P_{1/2}$ transition [83]. Our values agree well and

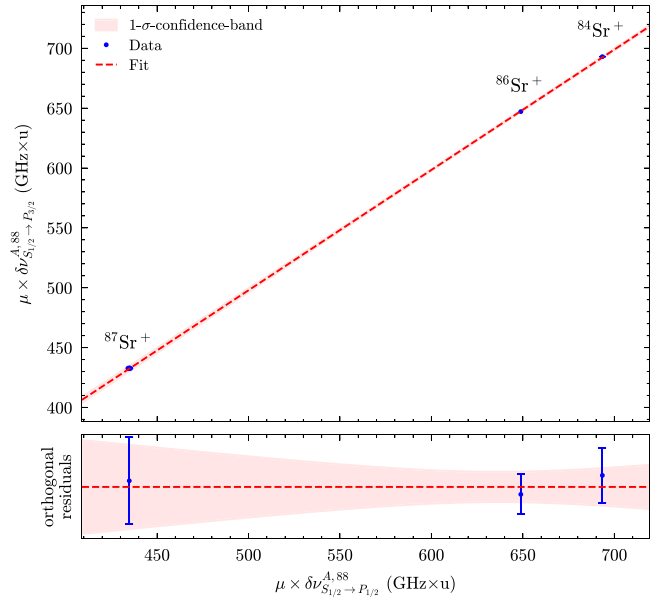


FIG. 5. King plot of the modified isotope shifts of the $D1$ and $D2$ transitions with ^{88}Sr as reference. The projected orthogonal residuals are shown below.

reduce the uncertainty by a factor of 2. Similar to the absolute transition frequencies, we can also calculate the isotope shifts between ^{88}Sr and the other stable isotopes in the $S_{1/2} \rightarrow D_{3/2,5/2}$ transitions. These are also included in the table and nicely agree with the literature in the two cases in which measurements are available [79,81].

Following Eq. (4), plotting the modified isotope shift of the $D1$ and $D2$ transition against each other yields the King plot shown in Fig. 5. For the fit, the algorithm described by York *et al.* [87] was used to take uncertainties in both transitions (i.e., in x and y direction) into account in a self-consistent manner. This procedure was performed with each isotope as reference; the plot shown in Fig. 5 uses ^{88}Sr . As can be seen, the fit goes through all data points without any sign of nonlinearity, which is expected at this level of accuracy. Possible nonlinearities arising from off-diagonal hyperfine interactions are expected to be significantly smaller in Sr^+ than in neutral Sr [88] due to the absence of closely lying singlet and triplet states. By unifying the slopes of the King plots with different reference isotopes, a final value for the field-shift

TABLE III. Experimental isotope shifts $\delta\nu_i^{A,88} = \nu_i^A - \nu_i^{88}$ with respect to ^{88}Sr for all investigated transitions. All values are in megahertz.

A	$\delta\nu_{D1}^{A,88}$	$\delta\nu_{D2}^{A,88}$	$\delta\nu_{D_{3/2} \rightarrow P_{1/2}}^{A,88}$	$\delta\nu_{D_{3/2} \rightarrow P_{3/2}}^{A,88}$	$\delta\nu_{D_{5/2} \rightarrow P_{3/2}}^{A,88}$	$\delta\nu_{S_{1/2} \rightarrow D_{3/2}}^{A,88}$	$\delta\nu_{S_{1/2} \rightarrow D_{5/2}}^{A,88}$
87	-56.73(22)	-56.45(23)	191.4(17)	193.7(23)	189.6(23)	-248.1(17)	-245.7(25)
86	-171.53(21)	-171.07(21)	400.2(12)	400.4(7)	400.2(19)	-571.8(12)	-569.9(12)
84	-375.3(8)	-375.07(23)	825.2(18)	827.1(21)	822.5(14)	-1200.5(17)	-1197.6(18)
87	-59(7) ^a	-56(3) ^c					-247.99(4) ^d
86	-170(3) ^b	-171(3) ^c	402(2) ^b				-570.264(0) ^e
84	-378(4) ^b	-373(5) ^c	828(4) ^b				

^aRef. [84]; ^bRef. [83]; ^cRef. [85]; ^dRef. [79]; ^eRef. [86].

TABLE IV. Experimental and theoretical HFS parameters for ^{87}Sr from this work in comparison to literature values from experiment [79,85,89] and theory [90]. For the theory results from Yu *et al.*, we utilized the same Q_I of 0.3102 b as in our own calculations.

	Experiment This work (MHz)	Experiment Ref. (MHz)	Theory This work (MHz)	Theory Ref. [90] (MHz)
$A_{S_{1/2}}$	−1000.51(4)	−1000.473673(11) ^a	−998(3)	−1003.2
$A_{P_{1/2}}$	−177.89(6)		−176(1)	−178.4
$A_{P_{3/2}}$	−36.1(2)	−36.0(4) ^b	−35.5(5)	−35.1
$B_{P_{3/2}}$	86.4(6)	88.5(54) ^b	85.4(6)	85.1(54)
$A_{D_{3/2}}$	−46.07(7)		−46.2(5)	−47.4
$B_{D_{3/2}}$	36.4(15)		35.9(4)	36.7(23)
$A_{D_{5/2}}$	2.17(15)	2.1743(14) ^c	2.25(9)	2.5
$B_{D_{5/2}}$	49(4)	49.11(6) ^c	49.1(5)	52.3(33)

^aRef. [89]; ^bRef. [85]; ^cRef. [79].

ratio of $f = 1.004(5)$ is obtained. Although the result is still statistically compatible with the nonrelativistic limit of 1, it remains fully consistent with the theoretically expected range below the ultrarelativistic upper bound of 1.019.

E. Hyperfine structure constants: Experiment and theory

The HFS constants for ^{87}Sr were directly fitted to the experimental spectra using a fully unconstrained model in which all hyperfine structure parameters, transition frequencies, a common linewidth, and amplitudes were treated as free parameters. For the $4d^2D_{5/2} \rightarrow 5p^2P_{3/2}$ transition, the literature values of Ref. [79] were used as initial values for the fit because of the small hyperfine splittings, while all parameters remained free. As most states are addressed by multiple investigated transitions, for which the agreement lies within 2σ , the results from different transitions are averaged. The final results are listed in Table IV and agree well with available literature values. While our results cannot compete with the achieved uncertainties in the ground state and states investigated via spectroscopy performed in traps [79,89], the excellent agreement provides confidence for those cases in which we report improved or—to our knowledge—first data.

In order to obtain HFS parameters from theory, we employ the Dirac-Coulomb-Breit interaction Hamiltonian that in atomic units is given by

$$H_{\text{at}} \equiv \sum_i [c \vec{\alpha}_i^D \cdot \vec{p}_i + (\beta_i^D - 1)c^2 + V_n(r_i)] + \sum_{i < j} \left[\frac{1}{r_{ij}} - \frac{\vec{\alpha}_i^D \cdot \vec{\alpha}_j^D + (\vec{\alpha}_i^D \cdot \hat{r}_{ij})(\vec{\alpha}_j^D \cdot \hat{r}_{ij})}{2r_{ij}} \right], \quad (12)$$

where α^D and β^D are the Dirac matrices, $\vec{p}$ is the momentum operator of a single particle, $V_n(r)$ represents the nuclear potential seen by an electron, and r_{ij} represents the radial difference between the i th and j th electron. Atomic orbitals are generated using Gaussian type orbital functions in the V^{N-1}

potential formalism of Dirac-Hartree-Fock theory, where N is the total number of electrons of the considered Sr^+ ion. To account for the contributions from the residual electron correlation effects, we apply the relativistic coupled-cluster theory at the singles, doubles and triples excitation approximation (RCCSDT method). Details of this method can be found from the earlier studies of Sr^+ properties [91,92].

For ^{87}Sr , we have used the nuclear magnetic moment $\mu_I = -1.093\,603\,0\,\mu_N$ from Ref. [93] and the latest reported nuclear quadrupole moment $Q_I = 0.3102\text{ b}$ from Ref. [92] to estimate the A_{hf} and B_{hf} HFS constants, respectively. Corrections from the Bohr-Weisskopf effects are included in the A_{hf} values in a similar way as discussed in Ref. [94]. Uncertainties to the calculated values due to incompleteness of basis functions and approximations in the methods are estimated and reported along with the final values in Table IV. The calculated HFS constants are within one sigma of the combined uncertainty with our and previous experimental results [79,85,89]. The sole exception is $A_{P_{1/2}}$, for which it is slightly larger ($\sim 1.5\sigma$). We also find close agreement with the relativistic many-body calculations of Yu *et al.* [90]. For $A_{P_{1/2}}$, their result is closer to the experimental value, but the uncertainty of this value is not stated.

V. DETERMINATION OF LEADING-ORDER ISOTOPE-SHIFT PARAMETERS

We have now established a large set of isotope shifts in all allowed dipole transitions between the $5s$, $5p$, and $4d$ manifolds of fine-structure levels in Sr^+ from which we can in principle extract nuclear charge radii. According to Eq. (1) this requires knowledge of the atomic parameters F_i and K_i , which we obtained in three different ways:

(1) Theory: Mass-shift and field-shift factors from atomic theory.

(2) Experimental: King plot according to Eq. (3) solely based on charge radii obtained from a combined analysis of muonic-atom and electron-scattering data ($\delta \langle r_c^2 \rangle_{\mu,e}^{A,A'}$) and the measured isotope shift.

TABLE V. Input data for the King plots. Masses are taken from the atomic mass evaluation (AME) [100] and recent studies of ^{84}Sr [101], Barrett radii, changes of the Barrett radii between isotopes, and the V_2^{88} from the book of Fricke and Heilig [62]. The V_2 ratios of the other isotopes are estimated as described in the text. They are used to extract the root mean square (rms) charge radius $\langle r_c^2 \rangle_A^{1/2}$ according to $\langle r_c^2 \rangle_A^{1/2} = R_{k\alpha}^A/V_2^A$ and the differential mean square (ms) charge radii $\delta \langle r_c^2 \rangle^{A,88}$ along the chain.

A	88	87	86	84
Mass (u)	87.905 612 253(6)	86.908 877 495(5)	85.909 260 725(6)	83.913 419 1(13)
Barrett radius $R_{k\alpha}^A$ (fm)	5.4091(15)	5.4094(12)	5.4183(11)	5.4318(12)
Barrett radius difference $\delta(R_{k\alpha}^\mu)^{A,86}$ (10^{-3}fm)	9.2(6)	-8.9(6)	0	13.5(7)
V_2^A	1.282 19	1.282 03(8)	1.281 87(16)	1.2816(3)
$\langle r_c^2 \rangle_A^{1/2}$ (fm)	4.2186(12)	4.2194(10)	4.2269(10)	4.2384(15)
$\delta \langle r_c^2 \rangle^{A,88}$ (10^{-3}fm^2)	0	6(6)	69(6)	167(11)

(3) Theory-informed King fit: King fit as in (2) but with the field-shift factor F_i fixed to the theoretical value as obtained in (1) to extract an improved mass-shift factor from experiment.

All three procedures are regularly used in literature to extract the atomic parameters, depending on the available information for the respective element.

A. Theory

To calculate the atomic parameters, which are referred to as the first-order IS parameters ($\langle O^{\text{IS}} \rangle$) for the respective IS operator O^{IS} , we apply the finite-field (FF) approach by expressing the calculated energy of the atomic state in the RCCSDT method with valence orbital v as

$$E_v(\lambda) = E_v^{(0)} + \lambda \langle O^{\text{IS}} \rangle + \frac{\lambda^2}{2} \langle O^{\text{IS}} \rangle^{(2)} + \mathcal{O}(\lambda^3), \quad (13)$$

where $E_v^{(0)}$ is the energy due to atomic Hamiltonian H_{at} and $E_v(\lambda)$ is the total energy due to the modified atomic Hamiltonian $H_\lambda = H_{\text{at}} + \lambda O^{\text{IS}}$ for an arbitrary numerical perturbative parameter λ . We also apply our recently developed analytical response (AR) approach to estimate these IS parameters by solving the following inhomogeneous equation [95]:

$$(H_{\text{at}} - E_v^{(0)})|\Psi_v^{(1)}\rangle = (\langle O^{\text{IS}} \rangle - O^{\text{IS}})|\Psi_v^{(0)}\rangle, \quad (14)$$

where superscript 0/1 denotes the order of λ in the results. Further descriptions of both the FF and AR approaches in the RCCSDT theory to evaluate the IS constants can be found elsewhere [35,96,97]. The calculated F and K along with their uncertainties of the aforementioned states of Sr^+ are presented from both the FF and AR approaches of the RCCSDT method in Table VI. The agreement with the previously calculated F_i [48] that are included in the table is very good.

B. King plot

The King plot for the $D2$ transition is shown in the left part of Fig. 6 as an example for this approach. The values of $\delta \langle r_c^2 \rangle_{\mu e}^{A,A'}$ used as input parameters are included in the last line in Table V. They were calculated using the (model independent) Barrett radii $R_{k\alpha}$ from muonic-atom data and the ratio of radial moments V_2^A from elastic electron scattering, both taken

from Ref. [62] using

$$\langle r_c^n \rangle_{\mu e}^{1/n} = R_{k\alpha}^\mu / V_n^e. \quad (15)$$

In the following, we drop the superscripts μ and e on $R_{k\alpha}$ and V_2 and instead indicate the mass number A at this position. V_2 is only available for ^{88}Sr and it was used for all Sr isotopes by Fricke and Heilig [62]. Thus, the authors explicitly report charge-radii differences with purely statistical uncertainties and refrain from providing systematic uncertainties. In order to estimate the effect of the V_2 variation in Sr, we estimated V_2^A by utilizing experimental data of stable isotopes in close proximity, namely, Ge, Zr, Mo, and Pd taken from Refs. [62,98]. Across each isotopic chain of these elements, the deviation from sphericity $v_2^A = \sqrt{5/3}/V_2^A - 1$ as defined in Ref. [99] follows a linear trend toward the nuclear shell closure at $N = 50$. Below the shell closure, the trend is falling with increasing mass number, whereas it is rising above. To estimate V_2^A for isotopes other than ^{88}Sr , we fixed the value of v_2^A for ^{88}Sr and used the average of the absolute slopes of neighboring elements, yielding $v_2^A = v_2^{88} + (88 - A) \times [-1.2(0.7) \times 10^{-4}]$, from which V_2^A can then be calculated (see Table V). The uncertainty of the slope was chosen such that all (absolute) slopes obtained in the neighboring elements lie within 1σ . A plot explaining our procedure can be found in Fig. 9 in Appendix A.

The differential charge radius can then be calculated by

$$\begin{aligned} \delta \langle r_c^2 \rangle^{A,A'} &= \langle r_c^2 \rangle^A - \langle r_c^2 \rangle^{A'} \\ &= \left(\frac{R_{k\alpha}^A}{V_2^A} \right)^2 - \left(\frac{R_{k\alpha}^{A'}}{V_2^{A'}} \right)^2 \\ &= \left(\frac{R_{k\alpha}^A}{V_2^A} - \frac{R_{k\alpha}^{A'}}{V_2^{A'}} \right) \times \left(\frac{R_{k\alpha}^A}{V_2^A} + \frac{R_{k\alpha}^{A'}}{V_2^{A'}} \right) \\ &= \frac{1}{V_2^A} \left(\delta R_{k\alpha}^{A,A'} + \left(1 - \frac{V_2^A}{V_2^{A'}} \right) R_{k\alpha}^{A'} \right) \left(\frac{R_{k\alpha}^A}{V_2^A} + \frac{R_{k\alpha}^{A'}}{V_2^{A'}} \right). \end{aligned} \quad (16)$$

In the last step, we utilized the differential Barrett radii between two stable isotopes $\delta R_{k\alpha}^{A,A'} = R_{k\alpha}^A - R_{k\alpha}^{A'}$ provided in Ref. [62], which is known more accurately, as parts of the

TABLE VI. Atomic factors from the theoretical calculations using the AR and FF approach, from experimental data through a King-plot procedure, and with a theory-informed King plot as described in Sec. V. For the latter, we used the finite-field results from theory in the King plot. The field-shift factors F_i are in MHz/fm² and the mass-shift factors K_i are in GHz u.

	Theory (this work)		Theory Ref. [48]	Experimental	Theory informed
	AR	FF			
$F_{S_{1/2} \rightarrow P_{1/2}}$	-1300(10)	-1301(10)	-1331	-997(130)	-1301(10)
$F_{S_{1/2} \rightarrow P_{3/2}}$	-1307(5)	-1308(5)	-1340	-1001(131)	-1308(5)
$F_{D_{3/2} \rightarrow P_{1/2}}$	275(15)	265(10)	282	221(52)	265(10)
$F_{D_{3/2} \rightarrow P_{3/2}}$	267(15)	258(10)	273	161(59)	258(10)
$F_{D_{5/2} \rightarrow P_{3/2}}$	259(15)	246(12)	263	264(69)	246(12)
$F_{S_{1/2} \rightarrow D_{3/2}}$	-1574(25)	-1566(20)	-1613	-1219(165)	-1566(20)
$F_{S_{1/2} \rightarrow D_{5/2}}$	-1566(20)	-1554(15)	-1602	-1270(177)	-1554(15)
$K_{S_{1/2} \rightarrow P_{1/2}}$	237(50)	235(50)		386(40)	310(31)
$K_{S_{1/2} \rightarrow P_{3/2}}$	231(50)	231(50)		384(40)	308(31)
$K_{D_{3/2} \rightarrow P_{1/2}}$	-1340(80)	-1394(50)		-1457(17)	-1444(9)
$K_{D_{3/2} \rightarrow P_{3/2}}$	-1346(80)	-1397(50)		-1480(21)	-1450(10)
$K_{D_{5/2} \rightarrow P_{3/2}}$	-1359(80)	-1393(50)		-1437(22)	-1442(11)
$K_{S_{1/2} \rightarrow D_{3/2}}$	1580(100)	1628(80)		1842(51)	1756(38)
$K_{S_{1/2} \rightarrow D_{5/2}}$	1590(100)	1624(80)		1821(55)	1749(38)

uncertainties cancel [62,102]. The uncertainty of the V_2^A parameter is often neglected although it has a significant impact on the total uncertainty. Including the uncertainties of the V_2^A

as estimated above increases the uncertainties of the differential charge radii linearly up to a maximum factor of 1.8 for the $\delta\langle r_c^2 \rangle^{84,88}$ pair.

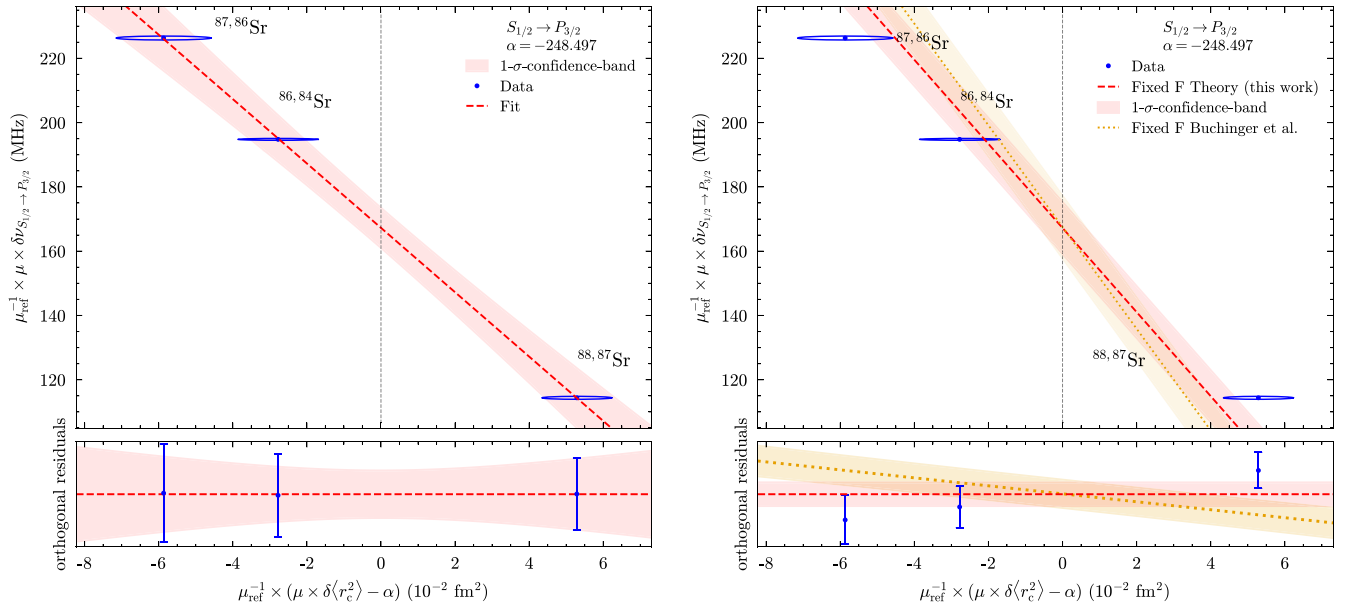


FIG. 6. Ladder-type King plots using optical data for the $S_{1/2} \rightarrow P_{3/2}$ transition from this work and the charge radii from literature provided in Table V. *Left*: With free parameters for slope and y intercept utilizing a York-type fitting routine [34,103]. *Right*: With the slope fixed to the theoretical value obtained in this work (red) and to that used by Buchinger *et al.* [85] (yellow). For both plots, both axes were scaled by a reference-pair factor of $\mu_{\text{ref}}^{-1} = \frac{(M_{88} - M_{86})}{(M_{88} \times M_{86})}$ for improved readability.

To decorrelate the fit results for the slope and y intercept, we can introduce an offset parameter α into Eq. (3)

$$\mu^{A,A'} \delta \nu^{A,A'} = F_i (\mu^{A,A'} \delta \langle r_c^2 \rangle^{A,A'} - \alpha) + K_{i,\alpha} \quad (17)$$

that shifts the origin of the abscissa into the center of gravity of the data as introduced by Hammen *et al.* [104].

We also performed a six-dimensional (6D) King plot (see Fig. 10 in Appendix B), which simultaneously includes all measured isotope shifts and the reference charge radii; additional details are provided in Appendix B. The six-dimensional fit confirms the consistency of our optical data, passing through all data points within one sigma and yielding results consistent with the conventional King plots.

It is important to note that the physically relevant mass-shift factor is obtained by projecting $K_{i,\alpha}$ back to $\alpha = 0$ to receive $K_i \equiv K_{i,0}$, which was done for the results of the King plots in Table VI to compare them to the theory results.

Our experimental King fit for the $S_{1/2} \rightarrow P_{3/2}$ transition yields $F = -1001(131)$ MHz/fm² and $K = 384(40)$ GHz u, in good agreement with the values $F = -1020(140)$ MHz/fm² and $K = 412(42)$ GHz u reported by Fricke and Heilig [62]. Their smaller uncertainties were obtained assuming the same V_2 value for all isotopes without uncertainties. Using the methods and input values documented in their compilation, we were not able to reproduce the quoted uncertainties.

There are surprisingly large differences of up to 2σ in the field-shift factors between theory and experiment, even though the theoretical accuracy is assumed to be at the 1% level and the linearity of the King plot is, in most cases, excellent. This is particularly critical with respect to the $D1$ and $D2$ transitions, which are most sensitive to the charge radius and exhibit a discrepancy of about 30%. This is drastically different from the case of Mg^+ , where an excellent agreement between theory and King-fit result was obtained [19,105]. For Sr^+ , good agreement is only found for the $D_{5/2} \rightarrow P_{3/2}$ transition, which has a comparably low sensitivity.

C. Theory-informed King plot

We have performed a theory-informed King plot in which the field-shift factor F is fixed to its theoretical value in order to obtain an improved mass-shift factor K , which is generally considered less reliable from theory. The theoretical uncertainty of F is propagated using a Monte Carlo procedure: In each realization, F is sampled from a Gaussian distribution defined by its theoretical central value and uncertainty, and K is refitted while accounting for the experimental uncertainties in both King-plot coordinates. The uncertainty reported for K is obtained from the resulting distribution and therefore contains contributions from both the experimental data and the theoretical uncertainty of F . The correlation between F and K is retained when propagating the result to derived quantities. The quoted χ^2 value, however, is evaluated with F fixed to its central theoretical value.

This theory-informed approach is sometimes used when only two or three stable isotopes exist and a reliable field-shift calculation is available, as is the case for alkaline atoms

(odd Z) with their comparatively “simple” atomic structure [106]. The result of the theory-informed King fit for the $S_{1/2} \rightarrow P_{3/2}$ transition is shown in the right panel of Fig. 6 and compared with the free King fit in the left panel. Fixing the slope to the central theoretical value increases χ^2 from 0.006 to 3.21, as is clearly visible in the figure. We additionally performed a fit with F restricted to the value previously used by Buchinger *et al.* [85], which was obtained using the semiempirical Goudsmit-Fermi-Segrè (GFS) approach. This results in an even larger value of $\chi^2 = 7.92$, represented by the dotted line in the right panel of Fig. 6.

These surprisingly large deviations may arise from several sources, and it is important to distinguish between the calculations of the field-shift (F) and mass-shift (K) factors. Calculations of the specific mass shift are known to be notoriously difficult, as they are highly sensitive to electron-correlation effects and challenging to converge reliably. In particular, missing higher-order correlation contributions, such as quadruple excitations, may significantly affect the calculated K values and their uncertainties. In this context, the observed deviations of the mass-shift factors, which remain within 2σ of the combined uncertainties, are not alarming. In contrast, calculations of the field-shift factors are generally considered more robust, with different theoretical approaches often agreeing at the level of a few percent [107–109]. Consequently, the discrepancies observed for the F factors are considerably more surprising.

Missing QED contributions may account for part of these discrepancies, particularly for transitions involving the relativistic $S_{1/2}$ and $P_{1/2}$ states, but are unlikely to explain their full magnitude. For the mass-shift factors, the noticeable differences between results obtained using the nonrelativistic and relativistic mass-shift operators indicate that relativistic corrections are relevant, while higher-order electron-correlation effects may provide additional contributions.

A second possible source of systematic effects originates from the nuclear input data used in the King-fit procedure. While the optical isotope-shift measurements presented here have very small experimental uncertainties, the extraction of atomic factors relies on differential charge radii derived by combining Barrett radii from muonic-atom spectroscopy with ratios of radial moments V_2^A obtained from elastic electron scattering.

More generally, recent reevaluations of muonic-atom data have demonstrated that improved treatments of QED and nuclear-structure effects can lead to noticeable changes in the extracted nuclear charge radii [110]. Such efforts are now being extended to other nuclei, including ^{90}Zr [111], and are supported by recently developed theoretical frameworks for medium-mass muonic atoms [112]. For the stable Sr isotopes compiled by Fricke and Heilig [62], however, the quoted uncertainties of both the Barrett equivalent radii and the differential Barrett moments are dominated by the theoretical uncertainty of the nuclear-polarization correction rather than by the experimental uncertainties of the measured muonic transition energies. Consequently, a modern reevaluation of the theoretical corrections applied to the published experimental energies appears considerably more promising than a reassessment of the experimental energies themselves.

TABLE VII. Differential ms nuclear charge radii $\delta\langle r_c^2 \rangle^{A,88}$ of the stable Sr isotopes obtained from the experimental isotope shifts of this work in all five investigated transitions using the three sets of isotope-shift parameters (K_i , F_i) derived from theory, a free King plot and a theory-informed King plot as listed in Table VI and described in the text. At the top are reference values calculated directly from the Barrett radii given by Fricke and Heilig [62] and our estimated values for V_2^A (see Table V) and a literature value from Buchinger *et al.* [85], where isotope shift measurements were combined with theory values from a Goudsmit-Fermi-Segrè approach. All values are in 10^{-3} fm^2 .

	$\delta\langle r_c^2 \rangle^{87,88}$	$\delta\langle r_c^2 \rangle^{86,88}$	$\delta\langle r_c^2 \rangle^{84,88}$
From Barrett radii [62]	6(6)	69(6)	167(11)
From IS and GFS [85]	7(4)	50(8)	116(16)
<i>F_i</i> and <i>K_i</i> from King plot			
$S_{1/2} \rightarrow P_{1/2}$	6(5)	70(6)	167(13)
$S_{1/2} \rightarrow P_{3/2}$	6(5)	69(6)	167(13)
$D_{3/2} \rightarrow P_{1/2}$	6(12)	69(12)	167(22)
$D_{3/2} \rightarrow P_{3/2}$	4(21)	57(17)	162(31)
$D_{5/2} \rightarrow P_{3/2}$	8(13)	77(13)	170(22)
Average (King plot)	6(5)	70(6)	167(13)
<i>F_i</i> and <i>K_i</i> from theory			
$S_{1/2} \rightarrow P_{1/2}$	20(5)	84(10)	191(21)
$S_{1/2} \rightarrow P_{3/2}$	20(5)	84(10)	191(21)
$D_{3/2} \rightarrow P_{1/2}$	36(25)	120(50)	267(103)
$D_{3/2} \rightarrow P_{3/2}$	44(27)	120(52)	275(106)
$D_{5/2} \rightarrow P_{3/2}$	32(28)	129(55)	278(111)
Average (theory)	21(5)	86(10)	195(21)
<i>F_i</i> from theory, <i>K_i</i> from theory-informed King plot			
$S_{1/2} \rightarrow P_{1/2}$	12(3)	69(6)	159(13)
$S_{1/2} \rightarrow P_{3/2}$	12(3)	68(6)	159(13)
$D_{3/2} \rightarrow P_{1/2}$	11(8)	69(10)	164(20)
$D_{3/2} \rightarrow P_{3/2}$	18(10)	66(11)	164(22)
$D_{5/2} \rightarrow P_{3/2}$	6(11)	77(13)	170(23)
Average (theory informed)	12(3)	69(6)	162(13)

The dominant theoretical uncertainty in the extraction of the Barrett radii arises from the nuclear-polarization correction [113]. In the analysis summarized by Fricke and Heilig [62], uncertainties corresponding to 30% of the correction were assigned to the absolute Barrett radii, while differential Barrett moments were assigned uncertainties of 10% of the larger correction of the isotope pair. In the absence of more rigorous calculations, we follow the same prescription in the present work. Modern calculations of the nuclear-polarization contribution, including improved treatments of nuclear correlations and odd-mass nuclei, could improve both the reliability of the extracted charge radii and the associated uncertainty estimates.

The conversion of the Barrett radii into rms charge radii additionally depends on the isotope dependence of the ratio of radial moments V_2^A obtained from elastic electron scattering. Since only V_2^{88} is available for Sr, the isotope dependence had to be estimated from neighboring isotopic chains. As shown in Fig. 9, this estimate was derived primarily from even-even nuclei. However, for the Sn isotopic chain, where V_2^A values are also available for odd-*A* isotopes, these values follow the approximately linear trend between their even-even neighbors [62]. This observation suggests that odd-even effects in V_2^A are small, although anomalous behavior of ^{87}Sr cannot be excluded entirely. In any case, the estimated variation in V_2^A appears too small to fully explain the observed discrepancies.

VI. DIFFERENTIAL CHARGE RADII

In this section, we first investigate the consistency of the differential ms charge radii of the stable isotopes based on the different transitions and then the effect of the observed differences in atomic factors onto the charge radii extracted for the whole chain of Sr isotopes.

A. Stable isotopes in all transitions

Using the obtained uncorrelated atomic factors and precise isotope shift measurements, we calculate the differential charge radii according to

$$\delta\langle r_c^2 \rangle^{A,A'} = \frac{\delta V^{A,A'} - \frac{K_{i,\alpha}}{\mu^{A,A'}}}{F_i} + \frac{\alpha}{\mu^{A,A'}}. \quad (18)$$

Thus for the experimental data, we do not use the $\alpha = 0$ projected values of atomic factors from Table VI, but the full sets with $\alpha \neq 0$ and $K_{\alpha,i}$ given in Table IX in Appendix B. For the theory values, we utilized the values given in Table VI with $\alpha = 0$. The results for the individual transitions and the three pairs of atomic factors are provided in Table VII. The average value of $\delta\langle r_c^2 \rangle^{A,88}$ provided for each set of isotope-shift parameters and each isotope $A = 87, 86, 84$ is the weighted average of all transitions and its uncertainty has been chosen as the smallest obtained from an individual transition, with

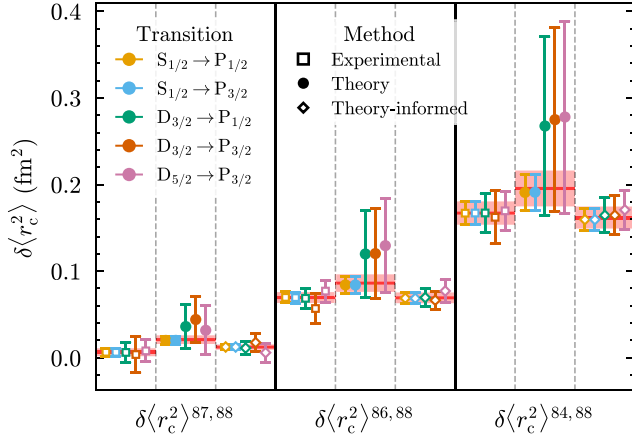


FIG. 7. Differential ms nuclear charge radii of ^{87}Sr (left), ^{86}Sr (middle), and ^{84}Sr (right) with respect to ^{88}Sr as obtained from the isotope-shift data measured in this work (see Table VII). The five investigated transitions are color coded and the three parameter sets (F_i , K_i) from the respective free King plot ($\square$), theory ($\bullet$), and the theory-informed King plot with fixed F_i ($\diamond$) use different markers. The weighted average of the five transitions for each isotope and parameter set is represented by the corresponding horizontal line. The shaded region indicates the estimated uncertainty.

no further reduction, since they are all correlated through the King plot with the same $\mu^{A,A'} \delta\langle r_c^2 \rangle^{A,88}$ data. The same approach was used for the theory data. As expected given the disagreement in the atomic factors, the charge radii calculated using the theoretical and experimental methods differ by up to 2σ . However, the theory-informed King plot yields good agreement with the experimental results.

The resulting charge radii obtained from the three different methods and the individual transitions listed in Table VII are shown in Fig. 7. In general, the approaches based on King-plot analyses yield significantly more consistent charge radii than the direct use of the theoretical (K_i , F_i) parameter sets, for which a substantially larger scatter is observed. The $D1$ and $D2$ transitions, where the calculations yielded the highest accuracy (see Table VI), exhibit the smallest uncertainties, whereas the uncertainties for the $4d \rightarrow 5p$ transitions are considerably larger.

Although the quoted uncertainties broadly reflect the observed scatter, the charge radii extracted from the purely theoretical approach are systematically larger than those obtained from the experimentally constrained King-plot analyses. This may indicate correlated systematic effects in the calculations. Since all transitions were calculated within a similar computational framework, missing contributions such as larger basis sets, QED corrections to the mass-shift, or higher-order electron-correlation effects including quadruple excitations could shift the calculated parameters coherently across all transitions, leading to a systematic offset in the extracted charge radii.

Constraining the King-plot analysis with theoretical F_i factors largely removes this discrepancy. Even for the $D1$ and $D2$ transitions, where the theoretical and experimental field-shift factors differ by approximately 30%, the averaged charge

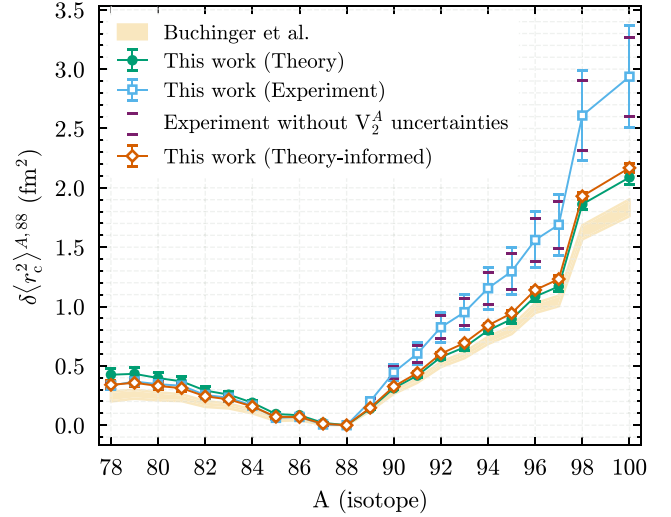


FIG. 8. The differential mean-square nuclear charge radii along the Sr isotope chain, relative to ^{88}Sr , to the choice of atomic factors. All results are based on the $D2$ isotope shifts measured by Buchinger *et al.* [85]. Shown are the original evaluation by Buchinger *et al.*, the results obtained with the theoretical atomic factors, the free experimental King fit, and the theory-informed King fit. The additional caps indicate the uncertainty of the experimental King-plot result when the V_2^A values are treated as exact.

radii from all transitions remain consistent within the quoted uncertainties.

B. Differential ms charge radii of $^{77-100}\text{Sr}$

The isotope shifts of the $D2$ transition previously measured for the radioactive isotopes $^{78-100}\text{Sr}$ [85] provide a sensitive illustration of the consequences of the different atomic factors discussed above. We recalculate the corresponding differential charge radii using the field- and mass-shift factors obtained from the experimental King plot, from atomic theory, and from the theory-informed King fit. The purpose of this comparison is not to establish three independent sets of recommended charge radii, but to quantify how the unresolved discrepancy between the atomic factors propagates along the radioactive isotope chain.

Buchinger *et al.* [85] used a field-shift factor of $F_{D2} = -1582(49) \text{ MHz/fm}^2$, obtained with the semiempirical Goudsmit-Fermi-Segrè approach. This value is approximately 50% larger in magnitude than our experimental King-plot result and 20% larger than our theoretical value, as also illustrated in Fig. 6. Buchinger *et al.* additionally reported a relativistic Dirac-Fock value of $F_{D2} = -1174 \text{ MHz/fm}^2$, but considered the semiempirical result more reliable and used it to extract the differential charge radii. The small uncertainty assigned to their field-shift factor corresponds to an estimated accuracy of 2% for the semiempirical approach, whereas uncertainties of approximately 20% are now considered more realistic for such estimates [99].

The resulting differential charge radii are compared in Fig. 8 and Table VIII. Constraining the field-shift factor F in the theory-informed King fit reduces the slope-intercept correla-

TABLE VIII. Reevaluated differential ms nuclear charge radii $\delta\langle r_c^2 \rangle^{A,88}$ for radioactive Sr isotopes based on the isotope shifts $\delta\nu^{A,88}$ in the $5s\ ^2S_{1/2} \rightarrow 5p\ ^2P_{3/2}$ transition measured by Buchinger *et al.* [85]. The three different methods disagree for heavier masses, as can also be seen in Fig. 8.

A	$\delta\nu^{A,88}$	$\delta\langle r_c^2 \rangle^{A,88}$			
	Ref. [85]	Ref. [85]	Exp.	This work Theo.	Theo.-inf.
78	−893(12)	0.24(4)	0.33(4)	0.43(6)	0.34(4)
79	−865(10)	0.26(4)	0.37(3)	0.43(5)	0.36(3)
80	−782(11)	0.24(3)	0.35(3)	0.40(4)	0.33(3)
81	−710(9)	0.23(3)	0.33(3)	0.37(4)	0.31(2)
82	−574(10)	0.18(2)	0.25(2)	0.29(3)	0.24(2)
83	−495(6)	0.16(2)	0.232(19)	0.26(3)	0.217(17)
83m	−456(7)	0.13(2)	0.193(18)	0.23(3)	0.187(17)
84	−373(5)	0.116(16)	0.165(14)	0.19(2)	0.158(13)
85	−216(5)	0.048(12)	0.062(12)	0.094(16)	0.071(10)
85m	−259(5)	0.075(12)	0.105(11)	0.127(16)	0.104(10)
86	−171(3)	0.050(8)	0.069(7)	0.084(10)	0.068(7)
87	−56(3)	0.007(4)	0.006(6)	0.020(5)	0.012(4)
87m	−7(4)	0.024(5)	−0.043(11)	−0.018(6)	−0.025(4)
88	0	0	0	0	0
89	−152(2)	0.124(5)	0.20(3)	0.139(5)	0.146(3)
90	−349(6)	0.277(13)	0.45(7)	0.312(11)	0.326(8)
91	−460(5)	0.374(16)	0.60(9)	0.418(15)	0.440(10)
92	−636(8)	0.51(2)	0.83(12)	0.57(2)	0.603(14)
93	−718(7)	0.59(3)	0.95(15)	0.66(2)	0.693(16)
94	−876(9)	0.71(3)	1.15(18)	0.80(3)	0.841(19)
95	−975(8)	0.80(3)	1.3(2)	0.89(3)	0.94(2)
96	−1198(10)	0.99(4)	1.6(2)	1.08(4)	1.14(2)
97	−1285(11)	1.05(4)	1.7(3)	1.17(4)	1.23(3)
98	−2166(11)	1.62(6)	2.6(4)	1.86(5)	1.93(3)
100	−2420(20)	1.83(7)	2.9(4)	2.09(6)	2.17(4)

tion and, consequently, the uncertainty of the fitted mass-shift factor K . This leads to smaller uncertainties than in the free King-plot analysis, while the resulting radii remain close to those obtained with the fully theoretical atomic factors.

The radii obtained with the different approaches are incompatible within their quoted uncertainties, particularly for the neutron-rich isotopes beyond the $N = 50$ shell closure. The discrepancies are smaller on the neutron-deficient side but increase progressively toward ^{78}Sr . The uncertainties shown in Fig. 8 are conditional on the input data and assumptions of each individual approach; they do not include the spread between the approaches. Accordingly, we do not adopt any of these results as a recommended set of differential charge radii. Rather, Fig. 8 demonstrates an unresolved method dependence in the extraction of the Sr charge radii.

At present, the origin of this inconsistency cannot be identified unambiguously. Our RCCSDT field-shift factors agree well with previous calculations based on relativistic Hartree-Fock methods combined with many-body perturbation theory and random-phase approximation [48]. The differences among these calculations are small compared with the approximately 30% discrepancy between theory and our experimental King-plot result (Table VI). At the same time, constraining the King-plot slope to the theoretical field-shift factor substantially increases the χ^2 of the fit. The stable Sr isotopes span only small changes in their mean-square charge

radii, making the extracted King-plot slope particularly sensitive to small changes in the nuclear input. These inputs rely on the theoretical interpretation of historical muonic-atom data, including nuclear-polarization corrections, together with the conversion of Barrett radii using the isotope-dependent V_2^A factors. The present comparison therefore does not establish whether the discrepancy originates predominantly from the atomic calculations or from the nuclear input to the experimental King plot.

Resolving this discrepancy requires improved information on both the atomic and nuclear sides. As discussed above, a modern reevaluation of the theoretical interpretation of the published muonic-atom data, particularly through improved QED and nuclear-polarization corrections, could provide revised Barrett radii. Measuring several transitions could better constrain the nuclear charge distributions, while including the long-lived isotope ^{90}Sr would be particularly valuable because it lies beyond the $N = 50$ shell closure, where the charge radii change more strongly.

The discrepancy is also relevant beyond Sr. Sr^+ , with one valence electron outside a closed-shell core, is generally considered a favorable system for high-accuracy atomic-structure calculations. A difference of the present magnitude therefore motivates similarly stringent comparisons in other monovalent systems for which independent nuclear-radius information and a sufficient number of stable isotopes

are available. Such benchmarks are particularly important because charge-radius extractions for elements without sufficient stable isotopes must rely directly on calculated atomic factors. The present analysis also illustrates the importance of publishing the isotope shifts, atomic factors, nuclear input data, and relevant correlations in sufficient detail to permit subsequent reevaluations when improved atomic or nuclear information becomes available [36].

VII. SUMMARY

We performed collinear laser spectroscopy of the $5s \rightarrow 5p$ and the $4d \rightarrow 5p$ transitions on all stable Sr^+ isotopes and achieved accuracies down to 600 and 210 kHz for transition frequencies and isotope shifts, respectively. We also extracted HFS parameters for ^{87}Sr , which were previously experimentally unknown for the $P_{1/2}$ and $D_{3/2}$ states. For the other states, they are in excellent agreement with literature values and the accuracy of $B(P_{3/2})$ has been improved by an order of magnitude. All values agree reasonably well with our theoretical results.

A central focus of this work was the determination of the field- and mass-shift factors F and K . Our coupled-cluster calculations differ by up to 2.3σ from the values extracted through a King-plot analysis using our optical data and nuclear charge radii derived from muonic-atom spectroscopy and elastic electron scattering [62]. For the $D1$ and $D2$ transitions, which have the largest sensitivity to changes in the nuclear charge radius, the field-shift factors differ by approximately 30%. A theory-informed King fit, in which F was constrained to the theoretical result while K was determined from the stable-isotope data, reduced the uncertainty of K but resulted in a substantially larger χ^2 than the free King fit. The present analysis does not allow us to determine whether the discrepancy originates predominantly from the atomic calculations or from the nuclear input used in the experimental King plot.

Reliable atomic factors are required for applications ranging from the extraction of the Weinberg angle from parity-nonconservation measurements in Cs [42,114–117] to the determination of nuclear charge radii from isotope shifts of short-lived isotopes [9,10,35] and proposed charge-radius measurements using x-ray transitions in Na-like highly charged ions [118–120]. The discrepancy found in Sr^+ , a nominally favorable system for high-accuracy atomic-structure calculations, therefore motivates similarly stringent comparisons in other monovalent systems. In particular, precise isotope shifts are already available for Ba^+ [59], making corresponding atomic-structure calculations in this system highly desirable.

Finally, we evaluated how the different atomic factors affect the differential charge radii extracted from the $D2$ isotope shifts of $^{78-100}\text{Sr}$ measured by Buchinger *et al.* [85]. The resulting radii show a substantial method dependence and become incompatible within their quoted uncertainties, particularly beyond the $N = 50$ shell closure. These uncertainties are conditional on the input data and assumptions of the respective approaches and do not account for the spread between them. We therefore do not recommend any of the resulting datasets as a definitive set of Sr charge radii. Im-

proved atomic-structure calculations together with a modern reevaluation of the QED and nuclear-polarization corrections used in the analysis of the existing muonic-atom data, and new measurements including ^{90}Sr would help to resolve this inconsistency.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [121].

APPENDIX A: ESTIMATION OF THE RATIO OF RADIAL MOMENTS V_2^{88}

In Ref. [62], only V_2^{88} is given. While they use this value without uncertainty for all isotopes in their King plot, we know that this is physically incorrect. We chose to use the linear trends of nearby elements toward the neutron shell close $N = 50$ as an estimate for this value. In Fig. 9, the deviation from sphericity $v_2^A = \sqrt{5/3}/V_2^A - 1$ is plotted for the elements of Ge ($Z = 32$) [98,122,123], Zr ($Z = 40$) [98,122,124], Mo ($Z = 42$) [98,122,124], and Pd ($Z = 46$) [98,122,125]. There are two conflicting sets of values plotted for Zr. Across all of these sets, we performed linear fits. We then took the largest and smallest absolute values for the slopes as an upper and lower boundary for the linear trend from the fixed value of V_2^{88} . We then took the average between those boundaries as the value used in our calculations.

APPENDIX B: KING PLOTS IN DETAIL: EXPERIMENTAL, THEORY-INFORMED, AND 6D KING PLOT

Table IX presents the results of the experimental and theory-informed King plots together with the corresponding values of α and K_α , which were used to calculate the charge radii according to Eq. (18).

Additionally, we performed a six-dimensional fit utilizing both Eqs. (1) and (4), thereby simultaneously incorporating the differential charge radii and the optical data from all transitions. This fit was performed both as a χ^2 minimization and with a Monte Carlo approach using the qspec Python package [103]. As both methods yielded indistinguishable results, only the χ^2 fit is included in Table IX. While in the two-dimensional (2D) case the correlation between slope and

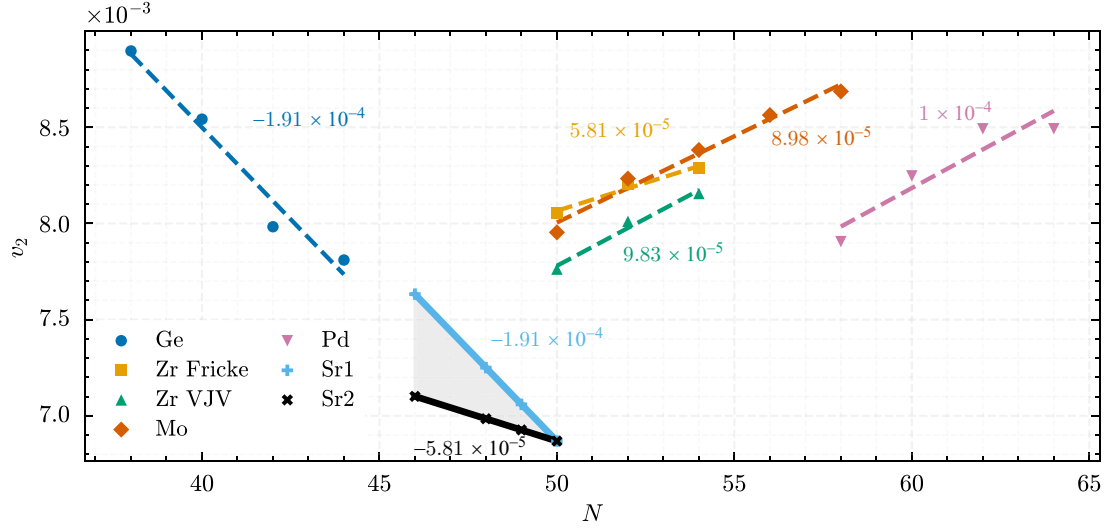


FIG. 9. The deviation from sphericity plotted against the neutron number N for different isotopes near $N = 50$. The two Sr lines are our lower and upper estimate.

intercept can be removed by introducing a shift of the abscissa by a constant α , in the six-dimensional case only an offset vector $\vec{\alpha}$ can be introduced to reduce, but not eliminate, the parameter correlations. The two-dimensional projections of the resulting χ^2 fit are shown in Fig. 10.

Although obtained from a single global optimization, the slopes and intercepts listed in Table IX describe the corresponding two-dimensional projections of the six-dimensional

fit. The projections of isotope shift versus differential charge radius therefore directly yield the field- and mass-shift constants F and K , respectively. Likewise, the transition-versus-transition King plots have slopes equal to the field-shift ratios introduced in Eq. (4), while their intercepts correspond to the appropriate combinations of the field- and mass-shift constants of the two transitions. The results of the six-dimensional fit are nearly identical to those obtained from

TABLE IX. Isotope-shift parameters obtained from the two-dimensional experimental and theory-informed King plots and from the six-dimensional experimental King plot. All values of F_i are given in MHz/fm², K_i and $K_{\alpha,i}$ in GHz u, and α in u fm².

	2D		6D
	Experimental	Theory informed	Experimental
$F_{S_{1/2} \rightarrow P_{1/2}}$	-997(130)	-1301(10)	-998(130)
$F_{S_{1/2} \rightarrow P_{3/2}}$	-1001(131)	-1308(5)	-1002(131)
$F_{D_{3/2} \rightarrow P_{1/2}}$	221(52)	265(10)	221(52)
$F_{D_{3/2} \rightarrow P_{3/2}}$	161(59)	258(10)	159(59)
$F_{D_{5/2} \rightarrow P_{3/2}}$	264(69)	246(12)	258(67)
$K_{S_{1/2} \rightarrow P_{1/2}}$	386(40)	310(31)	386(46)
$K_{S_{1/2} \rightarrow P_{3/2}}$	384(40)	308(31)	384(46)
$K_{D_{3/2} \rightarrow P_{1/2}}$	-1457(17)	-1444(9)	-1457(18)
$K_{D_{3/2} \rightarrow P_{3/2}}$	-1480(21)	-1450(10)	-1482(20)
$K_{D_{5/2} \rightarrow P_{3/2}}$	-1437(22)	-1442(11)	-1437(22)
$K_{\alpha, S_{1/2} \rightarrow P_{1/2}}$	634(24)	633(31)	
$K_{\alpha, S_{1/2} \rightarrow P_{3/2}}$	633(24)	633(31)	
$K_{\alpha, D_{3/2} \rightarrow P_{1/2}}$	-1519(8)	-1519(9)	
$K_{\alpha, D_{3/2} \rightarrow P_{3/2}}$	-1531(8)	-1532(10)	
$K_{\alpha, D_{5/2} \rightarrow P_{3/2}}$	-1513(10)	-1513(10)	
$\alpha_{S_{1/2} \rightarrow P_{1/2}}$	-248.329	-248.329	
$\alpha_{S_{1/2} \rightarrow P_{3/2}}$	-248.497	-248.497	
$\alpha_{D_{3/2} \rightarrow P_{1/2}}$	-283.906	-283.906	
$\alpha_{D_{3/2} \rightarrow P_{3/2}}$	-318.654	-318.654	
$\alpha_{D_{5/2} \rightarrow P_{3/2}}$	-286.381	-286.381	

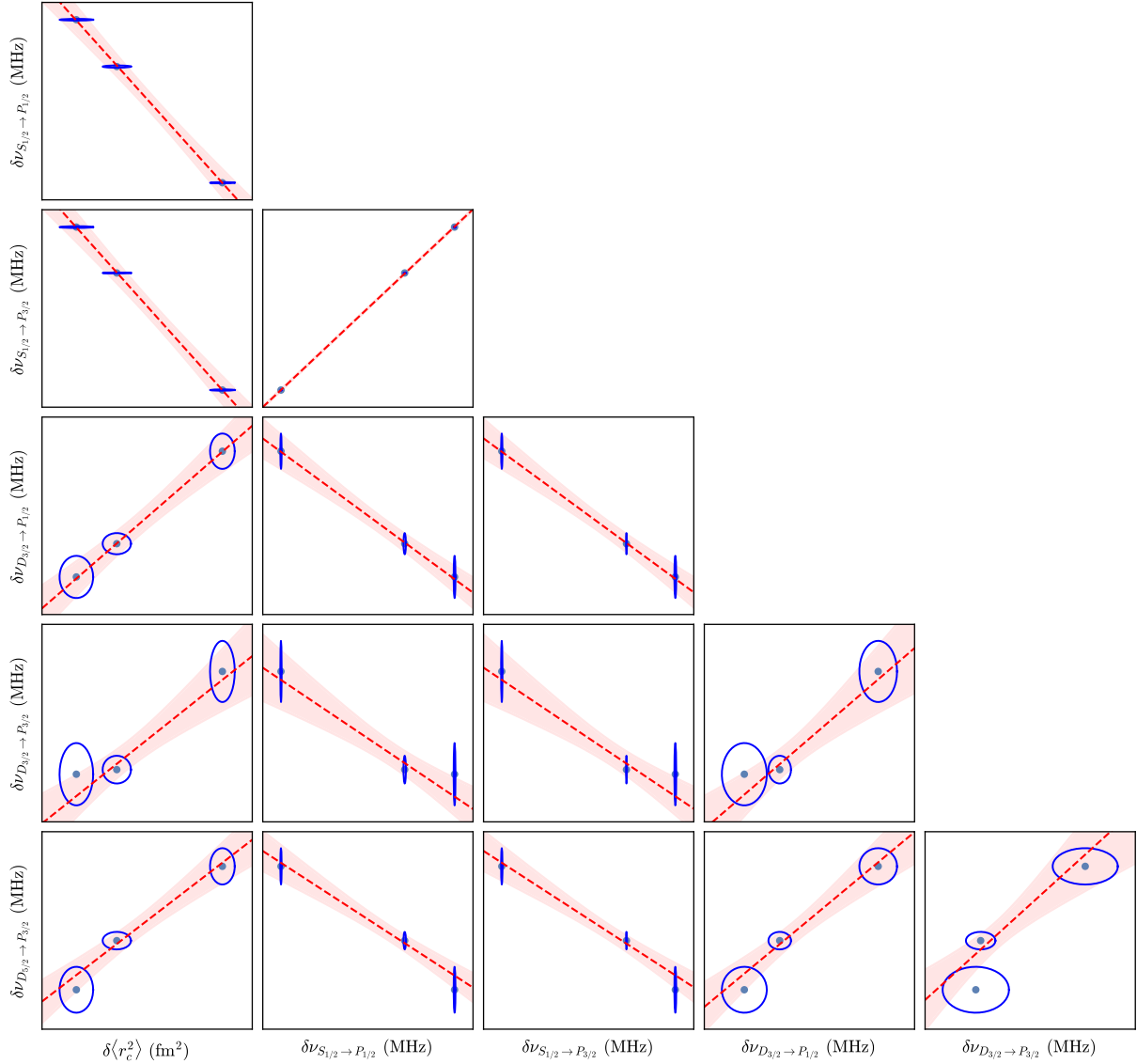


FIG. 10. The 2D projections of the six-dimensional King-plot procedure utilizing the χ^2 fit. The slopes and intercepts of the 2D projections onto the modified differential charge radii (first column) yield the isotope shift parameters tabulated in Table IX. The blue ellipses illustrate the 1σ uncertainty of the data points.

the individual two-dimensional King plots. However, the fact that all data points are intersected within one standard deviation

in Fig. 10 demonstrates the excellent internal consistency of the complete optical data set.

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