

## Resonance laser ionization spectroscopy of stable chromium isotopes

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(Received 2 March 2026; accepted 20 July 2026; published 8 September 2026)

Resonance laser ionization spectroscopy was conducted on stable chromium isotopes  $^{50,52,53,54}\text{Cr}$  using the RILIS laser ion source and the CRIS experimental apparatus at the ISOLDE/CERN facility. A unique titanium:sapphire laser-based three-step laser ionization scheme was developed to selectively and efficiently ionize and extract the Cr isotopes. Over 350 electronic transitions were observed, including several efficient transitions to autoionizing states. High-resolution spectroscopic measurements were performed with the  $3d^5(^6S)4s\ a^7S_3 \rightarrow 3d^5(^6S)4p\ z^7P_3$  atomic ground-state transition to characterize the scheme and benchmark the performance of the CRIS setup. The isotope shifts  $\delta\nu_{50,52} = -121(7)$  MHz,  $\delta\nu_{50,53} = -137(6)$  MHz, and  $\delta\nu_{54,52} = -14(6)$  MHz were measured, and the hyperfine structure parameter  $A_{S_3} = -82.64(16)$  MHz of  $^{53}\text{Cr}$  was extracted. A King plot analysis allowed reevaluation of the field shift  $F = -271(11)$  MHz fm $^{-2}$  and mass shift  $M = 105.3(30)$  GHz u factors of the transition with improved precision and accuracy. High-resolution measurements agree with the existing literature, supporting the accuracy and the reliability of the method for future studies involving radioactive isotopes.

DOI: [10.1103/4kpl-b7cs](https://doi.org/10.1103/4kpl-b7cs)

### I. INTRODUCTION

High-resolution laser spectroscopy has long been established as a powerful tool to study ground-state nuclear properties, providing a nuclear model-independent access to the nuclear spin, electromagnetic multipole moments, and changes in mean-square charge radii [1,2]. Ground-state nuclear properties of radioactive chromium (Cr) isotopes are of high interest for nuclear structure studies [3–5]. However, the laser ionization scheme currently used for the production of chromium ions at ISOLDE [6] with the Resonance Ionization Laser Ion Source (RILIS) [7,8] relies on dye lasers and was optimized for ionization efficiency only. No scheme tailored to nuclear structure investigations is available, which requires high efficiency and selectivity together with a spectroscopic step sensitive to nuclear parameters. Furthermore, a titanium:sapphire (Ti:Sa)-only scheme is preferred over dye

lasers to simplify operation and improve long-term stability. To achieve high efficiency and selectivity (i.e., ionizing only chromium atoms and minimizing the ionization of any other atoms or molecules as much as possible), such a scheme should make use of autoionizing states, corresponding to electronic states lying above the lowest ionization threshold, which spontaneously decay by electron emission. Such states must first be identified and characterized. The chosen spectroscopic transition must also provide good sensitivity to nuclear moments, with a hyperfine splitting large enough to be resolved and a field shift factor that allows changes in mean-square charge radii to be extracted from isotope shift measurements.

While several laser ionization schemes for Cr have been reported in the literature, none of them meets all the required specifications, motivating the present scheme development. After the scheme development work, a two-step Ti:Sa scheme was published in Ref. [9] (see references therein for a review of available schemes). A direct comparison of the present scheme and the one of Ref. [9] could be useful for future experiments. The scheme presented in this work was already used in Ref. [5] for hyperfine structure measurements of the radioactive  $^{61}\text{Cr}$  isotope, where its performance was partly characterized. This paper complements Ref. [5] by reporting details of the scheme development campaign performed at the RILIS of the ISOLDE facility, including the testing of two

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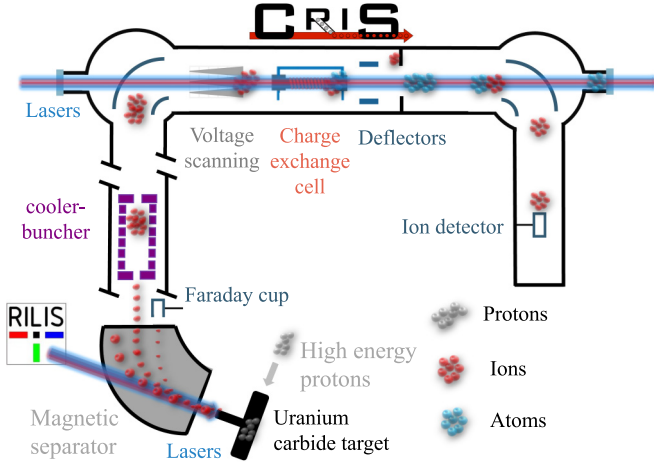


FIG. 1. Schematic of the experimental setup at the ISOLDE facility at CERN, featuring the infrastructure of RILIS and CRIS. See text for details.

second step transitions and the identification of previously unobserved transitions.

The Collinear Resonance Ionization Spectroscopy (CRIS) experiment is a laser spectroscopy setup operating at the ISOLDE facility of CERN, which brings together the high resolution achieved by collinear laser spectroscopy with the high sensitivity of resonance ionization spectroscopy [10,11], enabling precision measurements on both stable and rare radioactive isotopes. The laser scheme developed in this work was used both for production at RILIS and for spectroscopy using the CRIS experiment where it was characterized with high-resolution measurements. Precision measurements of isotope shifts and hyperfine parameters were performed in the  $3d^5(^6S)4sa^7S_3 \rightarrow 3d^5(^6S)4pz^7P_3$  transition, benchmarking the performance of the scheme and the attainable precision relative to existing literature data. A King plot analysis was used to extract improved mass and field shift factors, paving the way for future studies of charge radii of radioactive Cr isotopes. Our ionization scheme provides an efficient and selective production method of Cr ion beams and the spectroscopic measurements establish a solid pathway for extending high-resolution laser spectroscopy to rare radioactive chromium isotopes.

## II. SETUP OVERVIEW

A schematic view of the experimental setup is shown in Fig. 1. The Cr ions were produced at the ISOLDE facility at CERN [12] by nuclear reactions driven by impact of a 1.4 GeV proton beam on a uranium carbide target. After diffusing out of the target, the Cr atoms were ionized through a three-step resonant laser ionization process, using the RILIS [8]. These ions were then accelerated to 30 keV and the mass of interest was selected using the magnetic separator of ISOLDE (the high-resolution separator was used for the CRIS experiment while the general purpose separator was used for the RILIS ionization scheme development). For the RILIS study, the ion current was read on a Faraday cup downstream of the separator.

In the case of the high-resolution study at CRIS, the ion beam was cooled and bunched (at 250 Hz frequency and about 1  $\mu$ s of temporal width) in a radio-frequency quadrupole (RFQ) gas-filled linear Paul trap (ISCOOL) [13], before being sent to the CRIS beam line [14,15]. In the CRIS setup, the ion beam was first neutralized by charge exchange reactions, using a charge-exchange cell (CEC) filled with sodium vapor at about 210 °C. The ions remaining after the CEC were dumped using a set of electrostatic deflectors while the atom bunches were sent to the interaction region (maintained at low  $10^{-9}$  mbar to minimize nonresonant collisional ionization) where they were collinearly overlapped in space and time with three laser pulses to resonantly excite and ionize the Cr atoms. The ions were then deflected towards a single-ion detector: a MagneToF detector from ETP.

The timing of ion bunches and laser pulses was synchronized and controlled using a multichannel pulse generator (Quantum Composers 9520 Series). The transition frequency was scanned in the ion's rest frame by tuning the velocity of the ions, and therefore the Doppler shift of the laser frequency, using a voltage scanning device [15] placed upstream of the CEC. The voltage was scanned by increments of 1 V. The ISCOOL voltage, defining the beam energy, was measured every 20 ms using the combination of a voltage divider and a 7 1/2 digit Keithley DMM7510 voltmeter. The voltage divider was tested and characterized offline, for which a divider ratio of 10001.644(51) was measured [16]. No significant long-term drift of the divider ratio was observed in later checks performed after the experiment. The ISCOOL voltage was stable over a single measurement (lasting between 10 min and a few hours) with typical average value of 29 925.30(2) V measured with 50 mV spread. A long-term drift of about 2 V was observed over 6 days. The voltage of the voltage scanning device was measured in a similar way, using the Ohm-Labs KV-30A voltage divider, with a divider ratio of 999.983(64), combined to another 7 1/2 digit Keithley DMM7510 voltmeter. Both voltmeters were calibrated less than a year before the experiment. All frequencies and wavelengths reported in this paper are given in vacuum and in the atom's rest frame. For the in-beam measurements performed at CRIS, the measured voltages from ISCOOL and the voltage scanning device were used to apply Doppler shift corrections on laboratory-frame measurements, which represents a significant contribution to systematic uncertainties for absolute frequency measurements as detailed below.

## III. LASER IONIZATION SCHEME DEVELOPMENT AT RILIS

### A. RILIS laser setup and autoionizing levels

The laser ionization scheme development campaign was performed with the RILIS laser ion source [8]. The scheme was designed to operate exclusively with Ti:Sa lasers, ensuring long-term stability, and to be suitable for both ion beam production, requiring high ionization efficiency and selectivity, and high-resolution spectroscopy, requiring the first step to be sensitive to nuclear parameters. To achieve selective ionization, the scheme was based on the use of an autoionizing state. All laser beams were produced at the RILIS laser laboratory

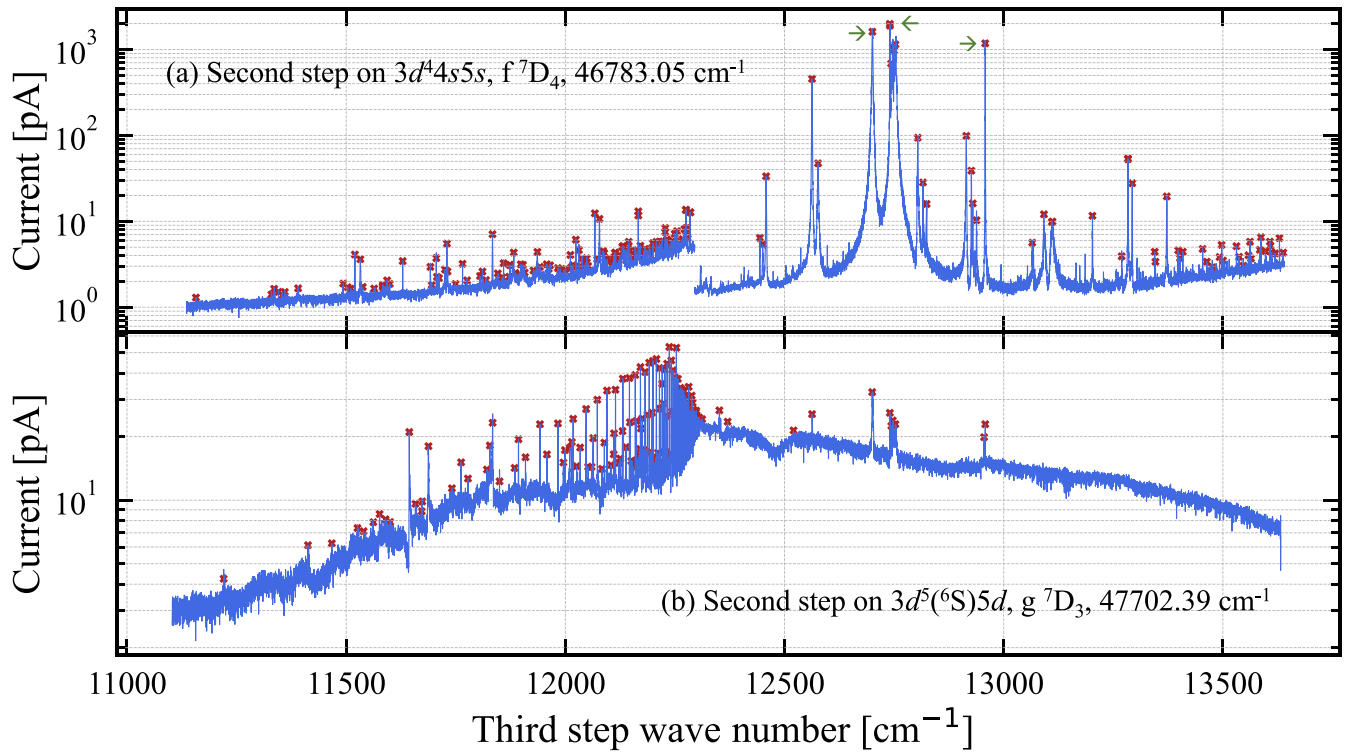


FIG. 2. Evolution of the Cr ion current as a function of the third step wave number. The first step frequency is 427.60 nm while the second step frequency is 427.41 nm in (a) and 411.21 nm in (b). The ion current is read on a Faraday cup located after the general purpose magnetic separator of the ISOLDE facility, selecting  $^{52}\text{Cr}$  isotopes only. Resonant states are indicated by the red crosses while green arrows highlight selected states for further saturation measurements. Assuming the respective second step energies, the IP should be located at  $7792.55\text{ cm}^{-1}$  in (a) and  $6873.21\text{ cm}^{-1}$  in (b).

(see Refs. [17,18] for details) and sent into the hot cavity ion source through a window in the mass separator magnet. The extracted ion current after laser ionization was measured with a Faraday cup located downstream of the magnetic separator (see Fig. 1), selecting  $^{52}\text{Cr}$  isotopes.

The first electronic excitation step was chosen from the literature as it is within the range that is easily accessible by Ti:Sa lasers with second harmonic generation (SHG), while having a good sensitivity to the changes in mean-square charge radii and nuclear magnetic dipole moment. It was the 427.60 nm wavelength from the electronic ground state  $3d^5(^6S)4s\ a^7S_3$  to the  $3d^5(^6S)4p\ z^7P_3$  excited state located at  $23\,866.34\text{ cm}^{-1}$ . A literature search was performed for electric dipole-allowed transitions from that state to a higher excited state which falls into the available Ti:Sa range [19,20]. Two states were chosen as possible second excitation steps—both in the SHG Ti:Sa range. The first was the  $3d^4 4s 5s\ f^7D_4$  state at  $46\,783.05\text{ cm}^{-1}$  (427.41 nm transition) and the second was the  $3d^5(^6S)5d\ g^7D_3$  state at  $47\,702.39\text{ cm}^{-1}$  (411.21 nm transition). The first and second excitation steps were produced with intracavity doubled-broadband Ti:Sa cavities. Wavelength scans were performed from these excited states to above the ionization potential (IP), located at  $54\,575.60\text{ cm}^{-1}$  with a grating Ti:Sa laser within its fundamental wavelength range. Wavelengths were measured with a HighFinesse WS7 wavemeter. Both scans are shown in Fig. 2. In Fig. 2(a), the measurement was interrupted and resumed later with slightly different target conditions leading to different average ion current, generating the step observed at about  $12\,300\text{ cm}^{-1}$ .

In these measurements, over 350 resonant transitions were observed. The transition wave numbers of the strongest transitions are shown in Table I, together with relative intensities. The table containing all transitions is given in the Supplemental Material [21].

The 427.41 nm second step transition was found to provide the highest ionization signal compared to the 411.2 nm one (see Fig. 2) and was therefore retained for further investigations. Using this second step, assuming that the third step starts from the respective second step targeted level, the third step scan corresponds to a total energy ranging from  $57\,883$  to  $59\,933\text{ cm}^{-1}$ . This range is located between the first ionization threshold Cr II,  $3d^5 d^6 S_{5/2}$  at  $54\,575.60\text{ cm}^{-1}$  and the second ionization threshold Cr III,  $3d^4\ ^5D_0$  at  $187\,546.62\text{ cm}^{-1}$ . States populated in this energy range should thus lead to autoionization into the continuum above the first ionization threshold Cr II only. From this second step, three third step transitions were chosen as candidates for this scheme based on ionization signal [indicated with green arrows in Fig. 2(a)]. These autoionizing states could not be observed using the 411.2 nm second step transition.

Saturation measurements were performed for these three transitions populating autoionizing states by recording the ion current as a function of the third step power. The obtained saturation powers were  $0.50(2)\text{ W}$  for transition number 1,  $0.67(4)\text{ W}$  for transition number 2, and  $0.84(2)\text{ W}$  for transition number 3 (powers given with a laser system running at  $10\text{ kHz}$  repetition rate). Based on this, and the ionization signal, the  $12\,740.29\text{ cm}^{-1}$  resonant step was chosen, leading

TABLE I. Third step resonant transitions in chromium observed in Fig. 2. Only the peaks with larger than 1% relative intensity to the highest peak are shown.

| No.                                                              | Wave number (cm <sup>-1</sup> ) | Ion current (pA) | Relative intensity |
|------------------------------------------------------------------|---------------------------------|------------------|--------------------|
| Second step on $3d^4 4s 5s, f^7 D_4, 46\,783.05\text{ cm}^{-1}$  |                                 |                  |                    |
| 1                                                                | 12740.29(23)                    | 1985.2           | 1.00               |
| 2                                                                | 12700.53(87)                    | 1610.0           | 0.81               |
| 3                                                                | 12957.79(18)                    | 1179.3           | 0.59               |
| 4                                                                | 12752.87(40)                    | 1144.0           | 0.57               |
| 5                                                                | 12747.18(93)                    | 1058.7           | 0.53               |
| 6                                                                | 12749.71(18)                    | 971.1            | 0.48               |
| 7                                                                | 12743.19(23)                    | 685.5            | 0.34               |
| 8                                                                | 12562.82(39)                    | 455.1            | 0.22               |
| 9                                                                | 12914.76(65)                    | 98.5             | 0.05               |
| 10                                                               | 12803.92(79)                    | 94.0             | 0.04               |
| 11                                                               | 13283.52(33)                    | 53.7             | 0.02               |
| 12                                                               | 13283.39(2)                     | 52.8             | 0.02               |
| 13                                                               | 12576.46(87)                    | 47.3             | 0.02               |
| 14                                                               | 12926.20(26)                    | 38.8             | 0.02               |
| 15                                                               | 12457.97(46)                    | 33.4             | 0.01               |
| 16                                                               | 12815.91(45)                    | 28.3             | 0.01               |
| 17                                                               | 13293.18(20)                    | 27.8             | 0.01               |
| 18                                                               | 13372.10(32)                    | 19.5             | 0.01               |
| Second step on $3d^5(^6S)5d, g^7 D_3, 47\,702.39\text{ cm}^{-1}$ |                                 |                  |                    |
| 19                                                               | 12275.71(09)                    | 13.6             | 0.01               |
| 20                                                               | 12165.04(05)                    | 13.1             | 0.01               |
| 21                                                               | 12285.69(29)                    | 12.7             | 0.01               |
| 22                                                               | 12063.42(12)                    | 12.4             | 0.01               |
| 23                                                               | 12284.20(17)                    | 11.3             | 0.01               |
| 24                                                               | 12072.94(12)                    | 10.7             | 0.01               |

to the final ionization scheme shown in Fig. 4. A saturation measurement for the second step at  $46\,783.05\text{ cm}^{-1}$  was also performed showing a saturation power of  $31.4(38)\text{ mW}$ . The production yield of Cr isotopes using the current scheme demonstrates a two- to threefold improvement in ion beam intensity compared to the last Cr experiment employing the previous scheme at RILIS [3]. However, it is difficult to estimate the contribution of our ion scheme to this improvement as yields also depend on other factors and typical fluctuations between the same type of targets can be of similar magnitude.

### B. Rydberg levels

In the third step scan, using the  $411.21\text{ nm}$  second step transition shown in Fig. 2(b), two series of states were observed [also shown in Fig. 3(a)] displaying the expected pattern of a Rydberg series. Populated Rydberg states would be ionized by different processes such as field ionization by the voltage gradient of the ion source, and photoionization by the blackbody radiation of the hot cavity or collisions inside the hot cavity [22]. The evolution of Rydberg state energy as a function of the principal quantum number  $n$  can be described by the Rydberg-Ritz formula:

$$E_n = E_f - \frac{R_M}{(n - \delta)^2},$$

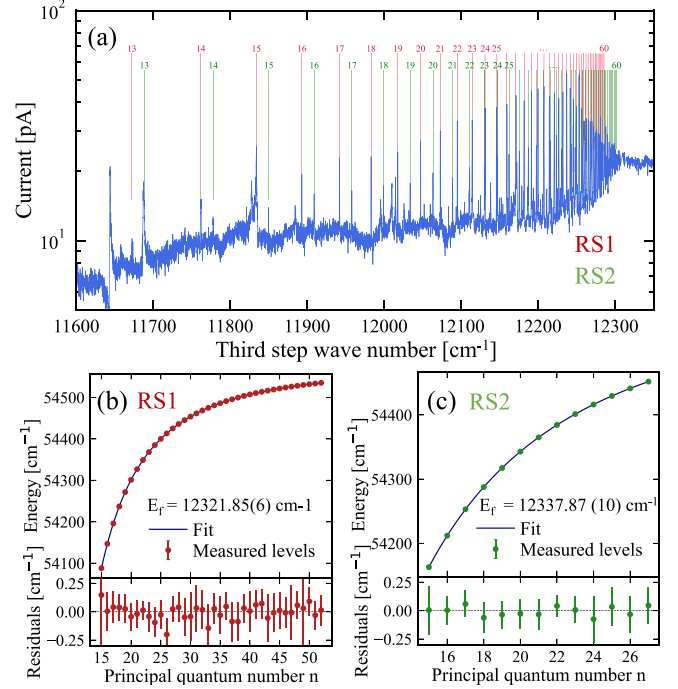


FIG. 3. (a) Evolution of the Cr ion current as a function of the third step wave number with the second step populating the  $3d^5(^6S)5d\ g^7 D_3$  state at  $47\,702.38\text{ cm}^{-1}$ . The two identified Rydberg series are marked with red and green vertical lines and their assigned principal quantum numbers  $n$ . (b), (c) Rydberg-Ritz fit of the two Rydberg series with residuals.

where  $E_n$  is the energy of the Rydberg state of principal quantum number  $n$ ,  $E_f$  is the convergence limit of the series,  $R_M$  is the mass corrected Rydberg constant ( $109\,736.157\text{ cm}^{-1}$  for  $^{52}\text{Cr}$ ), and  $\delta$  is the quantum defect. The  $E_f$  and  $\delta$  parameters were fitted to the transition energies belonging to a single Rydberg series. The fits of the Rydberg series 1 (RS1) and 2 (RS2) are shown in Figs. 3(b) and 3(c), respectively, while Fig. 3(a) shows the third step scan with the two Rydberg series marked with red (RS1) and green (RS2) vertical lines and their assigned principal quantum numbers  $n$ .  $E_f = 12\,321.85(6)\text{ cm}^{-1}$  and  $\delta = 0.003(3)$  for RS1 and  $E_f = 12\,337.87(10)\text{ cm}^{-1}$  and  $\delta = 0.002(3)$  for RS2 were obtained. Considering that such Rydberg states were populated from the  $3d^5(^6S)5d\ g^7 D_3$  state at  $47\,702.390(5)\text{ cm}^{-1}$ , the two observed series would converge to significantly different limits of  $60\,024.24(6)\text{ cm}^{-1}$  for RS1 and  $60\,040.26(10)\text{ cm}^{-1}$  for RS2. Neither value matches the known IP at  $54\,575.6(3)\text{ cm}^{-1}$  [23] or the first core-excited configuration located at  $66\,537.41\text{ cm}^{-1}$  [24]. Another possibility would be that the  $g^7 D_3$  state first decays to the  $3d^5(^6S)5p\ x^7 P_2$  and  $x^7 P_3$  states at  $42\,237.957(7)\text{ cm}^{-1}$  and  $42\,253.966(7)\text{ cm}^{-1}$ , respectively, before being excited by the third step laser. The comparatively long lifetime of the  $x^7 P$  term (145 ns), significantly exceeding that of the intermediate  $z^7 P$  (32 ns) and  $g^7 D$  (58 ns) states [25], and the allowed nature of the  $g^7 D \rightarrow x^7 P$  transition suggest that population can accumulate in this level. With that assumption, the two series converge to identical limits,  $54\,575.82(6)\text{ cm}^{-1}$  for RS1 and  $54\,575.83(10)\text{ cm}^{-1}$  for RS2, both compatible with the IP value of Ref. [23].

TABLE II. Energies of the  $3d^5(^6S)nd$  Rydberg states  $E_n$  with their corresponding principal quantum number  $n$  shown in Fig. 3(b)

| $n$ | $E_n$ (cm <sup>-1</sup> ) | $n$ | $E_n$ (cm <sup>-1</sup> ) |
|-----|---------------------------|-----|---------------------------|
| 15  | 54088.06(47)              | 16  | 54147.01(17)              |
| 17  | 54196.02(14)              | 18  | 54237.06(12)              |
| 19  | 54271.78(13)              | 20  | 54301.36(12)              |
| 21  | 54326.91(12)              | 22  | 54349.06(08)              |
| 23  | 54368.30(13)              | 24  | 54385.18(13)              |
| 25  | 54400.19(13)              | 26  | 54413.26(15)              |
| 27  | 54425.29(11)              | 28  | 54435.87(13)              |
| 29  | 54445.28(17)              | 30  | 54453.84(21)              |
| 31  | 54461.66(21)              | 32  | 54468.67(17)              |
| 33  | 54474.91(21)              | 34  | 54480.92(13)              |
| 35  | 54486.21(16)              | 36  | 54491.20(12)              |
| 37  | 54495.58(17)              | 38  | 54499.74(17)              |
| 39  | 54503.71(13)              | 40  | 54507.25(18)              |
| 41  | 54510.61(12)              | 42  | 54513.69(13)              |
| 43  | 54516.42(21)              | 44  | 54519.14(17)              |
| 45  | 54521.65(16)              | 46  | 54523.96(13)              |
| 47  | 54526.15(24)              | 48  | 54528.26(12)              |
| 49  | 54530.16(38)              | 50  | 54532.03(12)              |
| 51  | 54533.61(13)              | 52  | 54535.26(13)              |

These Rydberg states being populated from a  $P$  state, they should correspond to either an  $s$  or a  $d$  series. The fitted quantum defects being both compatible with zero, the observed Rydberg series should correspond to the  $3d^5(^6S)nd$  configuration [9]. The energy with associated uncertainties of the Rydberg states extracted from RS1 considering the proposed scenario is shown in Table II. The uncertainty includes the wavemeter calibration effect and is dominated by the fit error, itself governed by the experimental resolution of 1 cm<sup>-1</sup> or higher, limited by Doppler broadening in the hot ion source. In Ref. [9], the  $3d^5(^6S)nd$  Rydberg series was observed from the  $3d^4(^5D)4s4p(^3P^0) \gamma^7P_{2,3,4}$  states located at 27 728.811 cm<sup>-1</sup> and an IP value of 54 575.49(2) cm<sup>-1</sup> was determined from the convergence limit of the series. The value determined in this work deviates by 0.33(7) cm<sup>-1</sup> compared to Ref. [9]. Further investigation is required to clarify this difference.

Although the proposed scenario provides a consistent explanation of the observed spectra, it relies on a fast  $g^7D_3$  to  $x^7P_{2,3}$  decay before reexcitation. Such lambda decay has been observed previously at RILIS [26], but attempts to intentionally exploit it in other excitation schemes have not been successful. The present observations support an interpretation in which the  $x^7P_{2,3}$  level is significantly populated and contributes to the excitation scheme, leading to the observed Rydberg series. Further additional measurements would be valuable to more directly characterize such excitation scheme.

#### IV. CRIS SPECTROSCOPY SETUP

##### A. CRIS laser setup

The same laser ionization scheme developed as described above was used at the CRIS experiment to perform high-resolution spectroscopy in order to measure the hyperfine structure and isotope shifts from the spectroscopic transition. The scheme is shown in Fig. 4, together with the correspond-

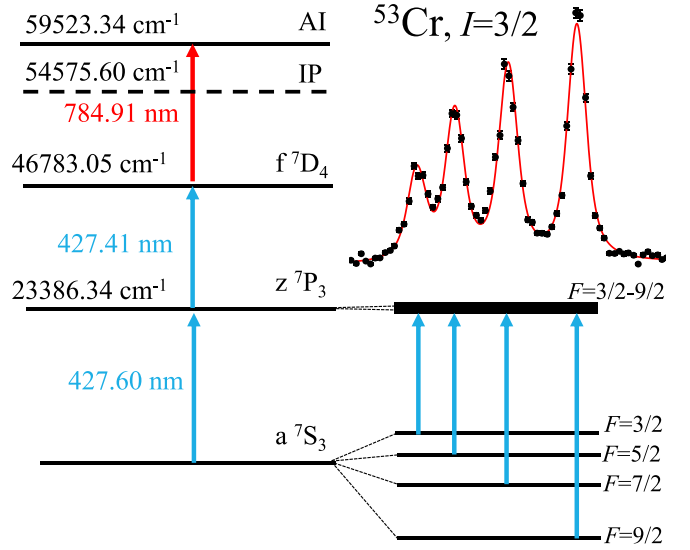


FIG. 4. Left: Schematic of the three-step laser ionization scheme developed in this work. IP is the ionization potential while AI denotes autoionizing states. This scheme is used both for production at RILIS and for high-resolution spectroscopy at CRIS. Right: Schematic of the hyperfine structure of the states belonging to the spectroscopic transition, along with an observed spectrum, is shown for  $^{53}\text{Cr}$ .

ing hyperfine splitting for  $^{53}\text{Cr}$  of the atomic ground and first excited states. At the CRIS setup, the first atomic transition  $3d^5(^6S)4s\ a^7S_3 \rightarrow 3d^5(^6S)4p\ z^7P_3$ , at 427.60 nm, was probed by laser light from an injection-locked Ti:Sa cavity [27], seeded by a narrow-bandwidth, continuous-wave Ti:Sa laser (Matisse 2 TS from Sirah). The linewidth 17(1) MHz of the fundamental light (measured after the experiment without intracavity doubling) from the injection-locked cavity at 855 nm was measured using a Fabry-Pérot interferometer. Atoms in the excited  $z^7P_3$  state were further excited to the  $3d^44s5s\ f^7D_4$  atomic state by an intracavity-doubled grating Ti:Sa laser probing the 427.41 nm transition. Finally, the Cr atoms were ionized by populating the autoionizing state (located 4948 cm<sup>-1</sup> above the ionization potential), using the 784.91 nm transition (transition 1 of Table I at 12 740.29 cm<sup>-1</sup>) driven by a broadband Ti:Sa laser. A linewidth of 8(1) GHz and 12(2) GHz were determined for the intracavity double-grating Ti:Sa laser and a double-etalon Ti:Sa laser, respectively, from the resonance width measured for these resonant steps. The three pulsed Ti:Sa cavities were pumped at 1 kHz by a 532 nm Nd:YAG Litron laser.

Laser frequencies were measured with a HighFinesse WS/U-2 wavelength meter, with a relative accuracy of 10<sup>-8</sup>. A Toptica saturated absorption spectroscopy unit, which consists of a diode laser (DLPRO780); a temperature-controlled vapor cell filled with K, Rb, and Cs (COSY); and the related locking electronics, was used to control the wavemeter stability. In this experiment, the frequency of the diode laser was locked to the  $F = 2 \rightarrow F = 1$  transition of the  $5^2S_{1/2} \rightarrow 5^2P_{1/2}$  line in  $^{87}\text{Rb}$  and was coupled into the high-precision wavelength meter. The wavelength measurement of the locked diode laser using the high-precision wavelength meter was about 17 MHz above the literature value of the locking transition and stable

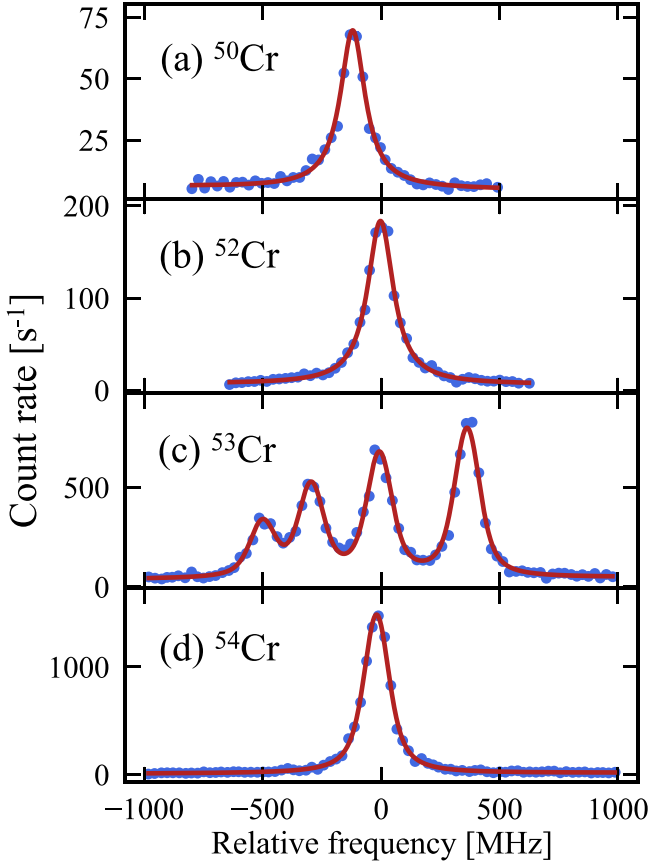


FIG. 5. Measured spectra of (a)  $^{50}\text{Cr}$ , (b)  $^{52}\text{Cr}$ , (c)  $^{53}\text{Cr}$ , and (d)  $^{54}\text{Cr}$  isotopes: count rate as a function of the rest-frame frequency of the first excitation step, relative to the centroid of  $^{52}\text{Cr}$ . Blue markers are experimental data while the red line represents the best fit to the data. The error bars are not visible because they are smaller than the marker size.

within 2 MHz over the course of the experiment. The locked diode laser has been used in the analysis to actively correct any offset and drift of the wavemeter calibration.

### B. Data analysis

In the following the details of the analysis procedure are presented which involves combining results from multiple measurements with different isotopes over a 1-week data taking period, while several systematic effects and temporal drifts need to be taken into account. While the hyperfine structure of  $^{53}\text{Cr}$  was already published in Ref. [5], the present goal is to detail data analysis methods and isotope shift measurements, and to benchmark the experimental procedure using stable isotopes. Figure 5 shows examples of the measured spectra at CRIS of the stable  $^{50,52,53,54}\text{Cr}$  isotopes. For each mass, the count rate in the ion detector is presented as a function of the rest-frame frequency of the first step laser, obtained after Doppler shifting the measured laboratory-frame frequency using ISCOOL and voltage scanning voltage measurements. Frequencies are shown relative to the centroid value of  $^{52}\text{Cr}$ . The error bar of each data point corresponds to the statistical error, where systematic uncertainties are discussed below. The

background is fully dominated by the nonresonant collisional ionization of chromium itself. Considering an initial beam intensity measured at the output of the RFQ cooler-buncher and measuring the maximum count rate on resonance after laser ionization, the overall efficiency of the setup varied between 0.1% and 0.5% (including transport, neutralization, laser ionization, and detection efficiencies). The average experimental resolution full width at half maximum (FWHM) was 100 MHz (dominated by power broadening), using a first step power of 350  $\mu\text{W}$  and a diameter of the laser beam of 8 mm, which was found to be the best compromise between resolution and efficiency.

The analysis of the hyperfine spectrum was performed using the SATLAS2 python package [28]. The red line in Fig. 5 shows the best  $\chi^2$  fit to the data for each mass. The fit function is composed of multiple Voigt functions plus a linear background function. For  $^{53}\text{Cr}$ , the position of the peaks was determined using the HFS Hamiltonian [2]. For odd- $A$  isotopes, the  $A_{P_3}$  and  $B_{P_3}$  hyperfine parameters of the upper excited state  $z^7P_3$ , as well as the  $B_{S_3}$  parameter of the ground state are well below the present experimental resolution (measured to be compatible with zero with about 10 kHz uncertainty [29–31]). These parameters were thus fixed to zero in the analysis. All other fitting parameters (the centroid and the amplitude of the hyperfine transitions, the  $A_{S_3}$  HFS parameter, the Gaussian and Lorentzian widths, and the two background parameters) were left free.

For each measurement, the spectrum was fitted and the value of the centroid frequency  $\nu$  and  $A_{S_3}$  was extracted with its associated fit error  $\sigma_{\text{fit}}$  (ranging from 2 to 10 MHz for the centroid and typically 1 MHz for  $A_{S_3}$ ; see Fig. 6). The fit error  $\sigma_{\text{fit}}$  accounts for statistical error, experimental resolution, and line-shape distortions. The evolution of the centroid  $\nu$  of stable Cr isotopes and the hyperfine parameter  $A_{S_3}$  of the HFS of  $^{53}\text{Cr}$  along the experiment is shown in Fig. 6, with the associated fit error  $\sigma_{\text{fit}}$ . The values are given relative to the weighted average of the corresponding mass.

For the  $A_{S_3}$  parameter, error bars follow the expected behavior of Gaussian statistics with about 68% of the points agreeing within one sigma error bar with the average value. However, in case of the centroid, the expected behavior from Gaussian statistics is not observed. Several measurements are located more than five sigmas away from the average value and some of them are ten sigmas. In order to quantify the underestimation of these error bars, following the procedure of Ref. [32], one can compute the reduced chi square  $\chi_r^2$  of the measurement distribution:

$$\chi_r^2 = \frac{1}{N-1} \sum (\bar{x} - x_i)^2 / \sigma_i^2,$$

where  $x_i$  is an individual measurement,  $\sigma_i^2$  its associated error, and  $\bar{x}$  is the average value of the  $N$  individual measurements. The data points of Fig. 6 were used to compute the  $\chi_r^2$  of the centroid frequency  $\nu$  and hyperfine parameter  $A_{S_3}$  distributions where the average values of all measurements corresponding to a given mass number  $\bar{x}_A$  were computed. Then the differences of individual measurements to their corresponding mass average value  $\bar{x}_A - x_{A,i}$  were computed and combined to compute a global  $\chi_r^2$  including the four different masses. For well-estimated uncertainties, the  $\chi_r^2$  of the

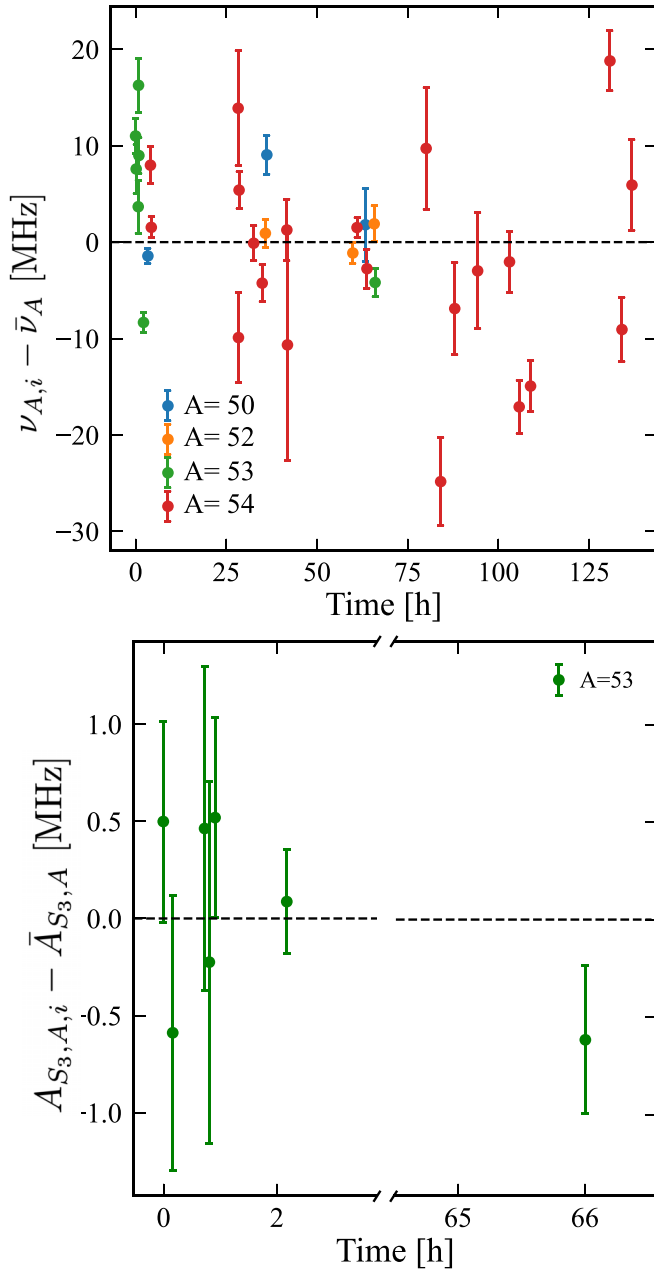


FIG. 6. Evolution of the measured centroid frequency  $\nu$  and hyperfine parameter  $A_{S3}$  of the HFS along the experiment. Values are given relative to the average value for the corresponding mass  $A$ . Error bars correspond to the fit error.

measurement distribution should be about 1 and larger than 1 if individual errors have been underestimated. The data points shown in Fig. 6 lead to a total  $\chi^2_{r,A_{S3}} = 0.96$  for the  $A_{S3}$  parameter and of  $\chi^2_{r,\nu} = 12$  for the centroid  $\nu$ . That shows that accounting only for the fit error underestimates the total error on the centroid measurement. Note that the fit error appears to be a good estimation for the total error of  $A_{S3}$ . This is due to the fact that  $A_{S3}$  corresponds to a relative frequency measurement in which many systematic effects cancel, as compared to the absolute frequency measurement of  $\nu$ .

An additional error  $\sigma_x$  is therefore introduced to account for this underestimation. This error  $\sigma_x$  appears to have a statistical

behavior, looking at the uniform spread of data in Fig. 6. Potential correlations of  $\sigma_x$  with time, laser power, resolution, or mass were investigated without any sign of correlation. In the following,  $\sigma_x$  is therefore treated statistically and the total error on a single centroid measurement  $\nu$  is considered to be  $\sigma = \sqrt{\sigma_{\text{fit}}^2 + \sigma_x^2}$ . This is a conservative estimation of the error in the sense that a centroid cannot be estimated with a better uncertainty than  $\sigma_x$  from a single measurement. Using these estimates, for the cases where measurements were done with low statistics or broadened linewidths, the fit error still dominates.

In order to determine the value of  $\sigma_x$  needed to obtain the expected behavior of Gaussian statistics,  $\sigma_x$  was adjusted to obtain  $\chi^2_v = 1$  on the centroid measurement distribution. Following this procedure, the value  $\sigma_x = 9.4$  MHz was obtained. The error  $\sigma_x$  is understood to have several origins such as the accuracy of the voltmeter calibration (accounting for about 6 MHz), the error on the voltage divider ratio (2 MHz), error on the voltage measurements (1 MHz), wavemeter accuracy (4 MHz for the fundamental light and 8 MHz after frequency doubling), and binning and fitting methods (2 MHz). All those effects being uncorrelated, that further justifies treating  $\sigma_x$  statistically.

Finally, for an isotope of mass  $A$ , all independent measurements taken in similar conditions are combined and treated as uncorrelated. The final values and errors of the centroid  $\nu_A$  and  $A_{S3,A}$  are calculating using [32]

$$\bar{x} \pm \delta\bar{x} = \frac{\sum_i w_i x_i}{\sum_i w_i} \pm \left( \sum_i w_i \right)^{-1/2}, \quad (1)$$

where  $w_i = 1/\sigma_i^2$ . This way, all sources of error are accounted for. Figure 7 shows the individual measured centroids  $\nu_i$  and their corresponding errors  $\sigma_i$  as the black points together with the weighted average value  $\bar{\nu}$  and its corresponding error  $\delta\bar{\nu}$  as the red line and shaded band, respectively.

## V. HIGH-RESOLUTION SPECTROSCOPY RESULTS AT CRIS

### A. Absolute frequencies

Figure 7 shows the centroids of all measurements of  $^{50,52,53,54}\text{Cr}$  performed in this work following the procedure detailed above. The weighted average for each mass leads to  $\nu_{50} = -2.6(54)$  MHz,  $\nu_{52} = 118.6(52)$  MHz,  $\nu_{53} = -134.0(35)$  MHz, and  $\nu_{54} = 104.9(21)$  MHz. The values are given relative to  $23\,386.3419\text{ cm}^{-1}$ , the value for the  $a^7S_3 \rightarrow z^7P_3$  transition provided by the NIST database [19].

For absolute frequency, measured frequencies are Doppler shifted to the atom's rest frame using the measured voltages of ISCOOL and the voltage scanning device and are thus highly dependent on the beam energy. Simulations of the ISCOOL device show that a 3–10 V offset between the platform ground and the bottom of the trapping potential can be present depending on the cooler-buncher settings, leading to an underestimation of the beam energy from the platform voltage measurement. Considering such systematic effect, the absolute frequency of the  $a^7S_3 \rightarrow z^7P_3$  transition measured in this work for  $^{52}\text{Cr}$  is  $23\,386.3430(4)[30]\text{ cm}^{-1}$  where the parentheses ( ) are the centroid measurement  $\nu_{52}$  error

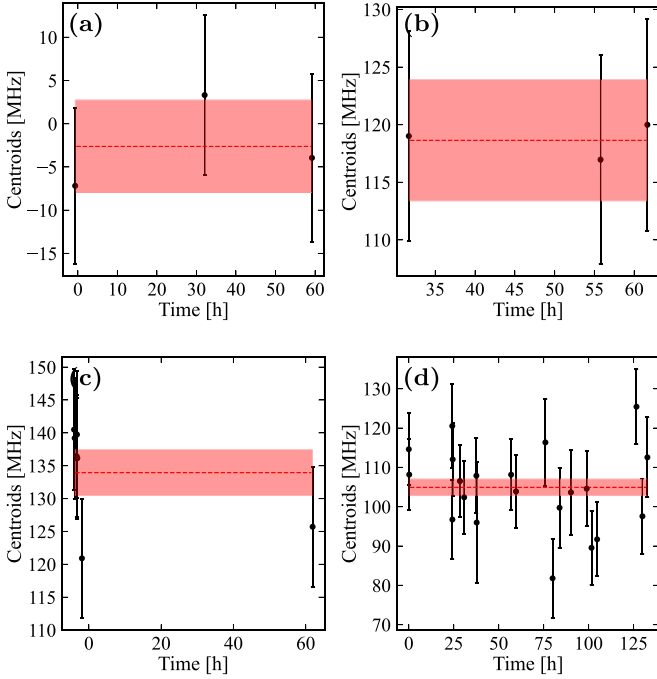


FIG. 7. Evolution of the measured centroid  $\nu$  of the spectra obtained for (a)  $^{50}\text{Cr}$ , (b)  $^{52}\text{Cr}$ , (c)  $^{53}\text{Cr}$ , and (d)  $^{54}\text{Cr}$  isotopes along the experiment. Centroids are given relative to the value  $23\,386.3419\text{ cm}^{-1}$  from Ref. [33]. Each black point represents the result of an individual measurement with associated uncertainties. The red line and shaded band represent the weighted average and variance, respectively.

estimated as detailed in the previous section and the square brackets [] are the error originating from the beam energy uncertainty. This value agrees with the literature value of  $23\,386.35(5)\text{ cm}^{-1}$  from Ref. [34] and  $23\,386.342(1)\text{ cm}^{-1}$  from Ref. [33] (see Table III). The error is dominated by the beam energy uncertainty, however, as the cooler-buncher settings were identical over the data taking period; this systematic effect would affect the absolute frequency but cancel out in the relative frequency measurements such as isotope shifts and hyperfine splitting (see below).

The second step absolute frequency was measured to be  $23\,396.7(1)\text{ cm}^{-1}$  in the present experiment, in good agreement with the literature value of  $23\,396.710(1)\text{ cm}^{-1}$  from Ref. [33]. The uncertainty is much larger for the second step compared to the first step. This arises from the difference of linewidth between the first (24 MHz) and second step (8 GHz) lasers.

TABLE III. Comparison of the  $a^7S_3 \rightarrow z^7P_3$  transition frequency measurement from this work to literature values.

| $a^7S_3 \rightarrow z^7P_3$ frequency ( $\text{cm}^{-1}$ ) | Reference      |
|------------------------------------------------------------|----------------|
| 23386.3430(4)[30]                                          | This work      |
| 23386.35(5)                                                | Reference [34] |
| 23386.342(1)                                               | Reference [33] |

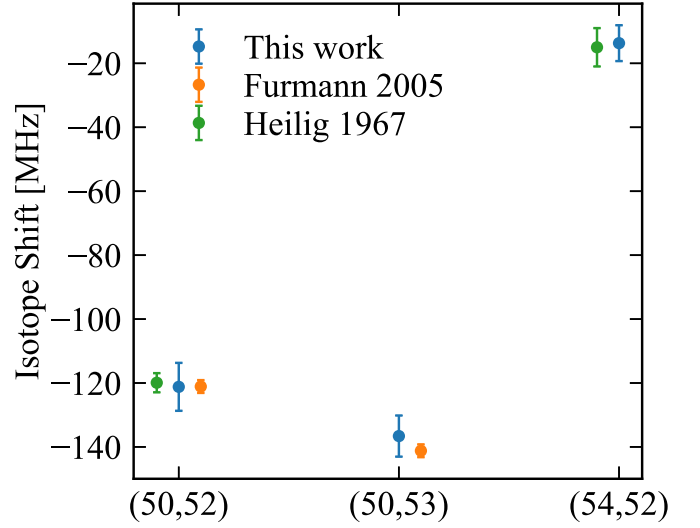


FIG. 8. Comparison of the isotope shifts measured in this work to literature values [35,36].

### B. Isotope shifts

Using the absolute frequencies extracted from Fig. 7, one can compute isotope shifts,  $\delta\nu_{A,A'} = \nu_A - \nu_{A'}$ , where  $\nu_A$  is the centroid of the spectrum of an isotope of mass  $A$ . The isotope shifts measured in this work are compared to available literature values in Fig. 8 and in Table IV. The results show good agreement with the literature within the experimental uncertainties, thereby confirming previously reported values and validating the accuracy of the present method for relative frequency measurements. The uncertainties achieved here remain larger than those obtained with atomic-beam laser induced fluorescence [35] or Fabry-Pérot spectroscopy [36]. However, the present ion beam spectroscopy approach provides access to short-lived isotopes at radioactive ion beam facilities.

### C. King plot analysis

The isotope shift can be expressed as a function of the difference in mean-square charge radii, neglecting higher-order terms, through the following:

$$\delta\nu_{A,A'} = M \frac{m_A - m_{A'}}{m_{A'} m_A} + F \delta\langle r^2 \rangle_{A,A'}, \quad (2)$$

where  $\delta\langle r^2 \rangle_{A,A'} = \langle r^2 \rangle_A - \langle r^2 \rangle_{A'}$  is the difference of mean-square charge radii,  $\delta\nu_{A,A'} = \nu_A - \nu_{A'}$  is the isotope shift,  $M$

TABLE IV. Comparison of the isotope shift  $\delta\nu_{A,A'} = \nu_A - \nu_{A'}$  measured in this work, in the  $3d^5(^6S)4s\ a^7S_3 \rightarrow 3d^5(^6S)4p\ z^7P_3$  transition, to literature values [35,36]. Last column shows a weighted average of all available values used below in the King plot analysis. All values are given in MHz.

| (A, A') | This work | Reference [35] | Reference [36] | Weighted average |
|---------|-----------|----------------|----------------|------------------|
| (50,52) | -121(7)   | -121(2)        | -120(3)        | -120.7(16)       |
| (50,53) | -137(6)   | -141(2)        | -              | -140.6(19)       |
| (54,52) | -14(6)    | -              | -15(6)         | -14.5(43)        |

TABLE V. Comparison of reevaluated radius benchmarked to those extracted from literature [43]. Absolute radii  $R_A = \sqrt{\langle r^2 \rangle}$  are given in fm and differential mean square radii are given in fm<sup>2</sup>.

| Observable                           | This work  | Reference [43] |
|--------------------------------------|------------|----------------|
| $R_{50}$                             | 3.6644(44) | 3.662(4)       |
| $R_{52}$                             | 3.6443(43) | 3.643(3)       |
| $R_{54}$                             | 3.6894(44) | 3.689(4)       |
| $R_{50}-R_{52}$                      | 0.0201(10) | 0.020(1)       |
| $R_{54}-R_{52}$                      | 0.0450(10) | 0.045(1)       |
| $\delta \langle r^2 \rangle^{50,52}$ | 0.147(7)   | 0.146(7)       |
| $\delta \langle r^2 \rangle^{54,52}$ | 0.330(7)   | 0.330(7)       |

is the mass shift factor,  $F$  is the field shift factor, and  $m_A$  is the atomic mass of a given isotope  $A$  [37].

In order to extract mean-square nuclear charge radii from isotope shift measurements, the field and mass shift factors  $F$  and  $M$  are required. These factors were determined for the  $a^7S_3 \rightarrow z^7P_3$  transition in Ref. [35]. However, the authors of [35] concluded that the obtained  $F$  factors were twice as large as the ones derived from *ab initio* calculations and suspected that the reference radii assumed might require some correction. This motivated the present reevaluation of the  $F$  and  $M$  factors for the  $a^7S_3 \rightarrow z^7P_3$  transition to be used in future studies dedicated to charge radii of radioactive chromium isotopes.

The atomic factors  $M$  and  $F$  for the present  $a^7S_3 \rightarrow z^7P_3$  were determined in this work through a King plot analysis. This approach determines the mass and field shift factors through Eq. (2), using benchmark differential mean-square radii from nonoptical techniques [38,39]. As is done most often, we multiply the relation of Eq. (2) by the modification factor  $\mu = \frac{m_A m_{A'}}{m_A - m_{A'}}$ , such that the mass and field shift factors correspond respectively to the intercept and slope of a linear fit.

For the isotope shifts, a weighted average was taken between the present isotope shift and the literature values (shown in Table IV), which allows improving the resulting uncertainty on  $F$  and  $M$ . For the reference radii, a reevaluation of the absolute and differential radii was performed, motivated by recent studies that have revealed additional corrections and uncertainties that were neglected in older works [40,41]. The results of the present reevaluation are shown in Table V. These values rely on the Barrett radii from Ref. [42], the nuclear shape correction from Ref. [40] (which in turn is based on measurements from Ref. [43]), and the nucleon polarization from Ref. [41]. Radii of  $^{53}\text{Cr}$  were not used for the King plot, as no high-quality electron scattering measurement was performed on this isotope [42]. Accordingly, the corresponding nuclear shape correction is uncertain.

While these measurements rely on the same underlying experiments, the addition of nucleon polarization shifts the absolute radii by up to 0.0024 fm for  $^{50}\text{Cr}$  (corresponding to  $0.5\sigma$ ). This effect, however, left differential radii unaffected. It was therefore opted to conserve the uncertainty determined for the differential radii from Ref. [43] in the King plot analysis. Given that this leaves only two points to describe a linear relation, uncertainties for  $M$  and  $F$  were determined

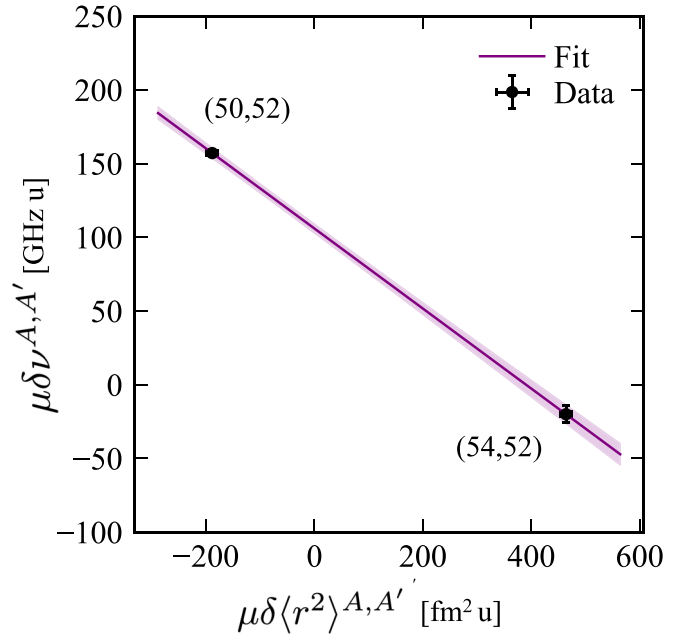


FIG. 9. King plot: modified isotope shift  $\mu\delta\nu_{A,A'}$  as a function of the modified difference of mean-square charge radii  $\mu\delta\langle r^2 \rangle_{A,A'}$  where  $\mu = \frac{m_A m_{A'}}{m_A - m_{A'}}$ . Black points show experimental data while solid line and shaded area show the linear trend associated with  $F = -271(11) \text{ MHz fm}^{-2}$  and  $M = 105.3(30) \text{ GHz u}$ .

directly from error propagation rather than a fitting process. The field shift  $F = -271(11) \text{ MHz fm}^{-2}$  and the mass shift  $M = 105.3(30) \text{ GHz u}$  were obtained (see Fig. 9). For error propagation purposes, one should note that the uncertainties of the  $M$  and  $F$  are correlated by  $+18.7\%$ .

In Ref. [35], a King plot analysis led to field and mass shift factors for this transition of  $F = -557(10) \text{ MHz fm}^{-2}$  and  $M = 103(10) \text{ GHz u}$ . While our measured isotope shifts and mass shift factor agree with those reported in Ref. [35] (see Table IV), our extracted  $F$  value is about 2 times smaller (deviating by about  $30\sigma$ ), confirming the suspicion of Ref. [35] as mentioned above. Indeed, Ref. [35] relies on differential mean-square radii from Ref. [44], who in turn cite Ref. [45] as their source. Yet the values quoted by Ref. [44] appear inconsistent with Ref. [45]. Furthermore, Ref. [35] based its analysis only on the (50,52) and (53,50) isotope pairs while, as noted above, no electron scattering data are available for  $^{53}\text{Cr}$ , inducing model dependency in the  $^{53}\text{Cr}$  radius [45]. The field shift  $F = -271(11) \text{ MHz fm}^{-2}$  and the mass shift  $M = 105.3(30) \text{ GHz u}$ , determined in this work have removed the model dependence associated with the radius of  $^{53}\text{Cr}$ .

#### D. Hyperfine parameters

The experimental method used for extracting  $A_{S_3}$  was benchmarked using the  $^{53}\text{Cr}$  stable isotope. This was previously described in Ref. [5]. Following the procedure described, the hyperfine  $A_{S_3}$  parameter of  $A_{S_3}(^{53}\text{Cr}) = -82.64(16) \text{ MHz}$  is obtained for  $^{53}\text{Cr}$ . Our result agrees within error bars with the literature value  $A_{S_3, \text{lit}}(^{53}\text{Cr}) = -82.5985(15) \text{ MHz}$  [31], further supporting the reliability of the method for relative frequency measurements. As described

above, the present spectroscopic step cannot be used to extract the  $B$  hyperfine parameters. Investigating the electric quadrupole moments of the Cr isotopes would require the development of a new laser ionization scheme with a spectroscopic step involving a larger  $B$  hyperfine splitting.

## VI. CONCLUSION

In summary, a unique Ti:Sa-based laser ionization scheme was developed at the RILIS laser ion source and 350 new resonant transitions were observed, including several efficient transitions to autoionizing states. Two second step transitions were tested. Autoionizing resonance was strongly populated using the  $3d^4 4s 5s f^7 D_4$  intermediate state, providing a maximum ion current about 40 times larger than obtained using the  $3d^5(^6S) 5d g^7 D_3$  intermediate state. Part of the population from the  $g^7 D_3$  state appears to be diverted to the  $x^7 P_{2,3}$  states from which excitation to a previously unobserved Rydberg series would provide a plausible explanation of the observed spectra. The present scheme represents an efficient and selective ionization method for Cr isotopes for use in ion beam production at radioactive ion beam facilities.

An experiment was conducted to perform high-resolution collinear resonance ionization laser spectroscopy on  $^{50,52,53,54}\text{Cr}$  using the CRIS experimental setup at the ISOLDE/CERN facility to characterize our scheme in high resolution and benchmark the setup. Using the  $3d^5(^6S) 4s a^7 S_3 \rightarrow 3d^5(^6S) 4p z^7 P_3$  transition, isotope shifts and the  $A_{S_3}$  hyperfine parameter were measured and compared with existing literature values. A King plot analysis was performed based on reevaluation of reference radii and

isotopes shifts, yielding to an improved precision and accuracy of the field shift  $F = -271(11)\text{MHz fm}^{-2}$  and mass shift  $M = 105.3(30)\text{GHz u}$  factors of the transition. The results from high-resolution measurements agree with the existing literature, supporting the accuracy and reliability of the method and paving the way for future studies of moments and radii of radioactive chromium isotopes.

## ACKNOWLEDGMENTS

We thank the ISOLDE technical teams for their support. We thank B. Ohayon for insightful exchanges on absolute charge radii. This project has received funding from the European Union's Horizon Europe research and innovation program under Grant Agreement No. 101057511 (EURO-LABS) and No. 861198-LISA-H2020-MSCA-ITN-2019; U.S. Department of Energy, under Grant No. DE-SC0021176; as well as from FWO International Research Infrastructures Project No. I001323N, FWO Projects No. G053221N, No. G0G3121N, No. G053221N, and No. G080022N and KU Leuven BOF C14/22/104; and STFC Grants No. ST/P04423/1, No. ST/V001116/1, and No. ST/L002868/1. K.M.L. acknowledges support from the Royal Society Grant No. DHFR1231007 and UK Research and Innovation (UKRI) under the UK government's Horizon Europe funding Guarantee Grant No. EP/Y036816/1 (ESPEN).

## DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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