EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Collinear Laser Spectroscopy of ^{223–226,228}Ra⁺

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P. Imgram¹, O. Ahmad¹, M. Athanasakis-Kaklamanakis^{1,2}, K. Blaum³, H. Bodnar⁵, B. Cheal⁴, S. Dutton³, T. Fabritz⁵, C. M. Fajardo-Zambrano¹, R. F. Garcia Ruiz⁶, J. Hughes⁴, F. Koehler⁵, K. Koenig⁵, T. Lellinger^{3,7}, B. Maaß^{5,8}, E. Matthews⁵, P. Mueller⁵, R. Neugart³, G. Neyens¹, W. Noertershaeuser⁵, J. Palmes⁵, P. Plattner³, L. Renth⁵, L. V. Rodriguez^{3,7}, R. Sanchez⁹, J. Spahn⁵, X. F. Yang¹⁰, D. T. Yordanov¹¹

¹Instituut voor Kern- en Stralingsfysica, KU Leuven, Leuven, Belgium.

²Centre for Cold Matter, Imperial College London, London SW7 2BW, UK.

³Max-Planck-Institut fuer Kernphysik, Heidelberg, Germany.

⁴Oliver Lodge Laboratory, University of Liverpool, UK.

⁵Institut fuer Kernphysik, Technische Universitaet Darmstadt, Darmstadt, Germany.

⁶Massachusetts Institute of Technology, Cambridge, MA, USA.

⁷Experimental Physics Department, CERN, Geneva, Switzerland.

⁸Physics Division, Argonne National Laboratory, Lemont, IL 60439, USA

⁹GSI Helmholtzzentrum fuer Schwerionenforschung GmbH, Darmstadt, Germany.

¹⁰School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing, China.

¹¹Universite Paris-Saclay, CNRS/IN2P3, IJCLab, 91405 Orsay, France.

Spokesperson: P. Imgram, phillip.imgram@kuleuven.be **Contact person:** T. Lellinger, tim.enrico.lellinger@cern.ch

Abstract: The long-lived odd-mass isotopes of radium ^{223,225}Ra are under investigation for experimental searches of nuclear parity- or time-reversal-violating moments, which would shed light on the level of symmetry violation in the fundamental forces. Precise measurements of the nuclear charge radius and the Bohr-Weisskopf (BW) can be used to

test the reliability of nuclear models. Here, we propose improving the statistical and systematic uncertainty in the spectroscopy of the $7s^2S_{1/2} \rightarrow 7p^2P_{1/2}$ and $7s^2S_{1/2} \rightarrow 7p^2P_{3/2}$ transitions in $^{223-226,228}$ Ra⁺ with COLLAPS by a factor of 10 using a pre-irradiated UC_x target in a winter-physics experiment. In combination with improved electronic-structure calculations, the higher precision and accuracy will significantly reduce the uncertainty in determining the magnitude of the BW effect and the differential nuclear charge radius. Summary of requested shifts: 13 shifts delivered in 1 run using a previously irradiated UC_x target without active proton irradiation (winter physics).

1 Physics motivation

The long-lived odd-A isotopes of radium $(Z = 88)^{223}$ Ra $(I^{\pi} = 3/2^{+})$ and 225 Ra $(I^{\pi} = 1/2^{+})$ have received a lot of attention in the last decade due to interest in their parity- or time-reversal- (P,T) violating nuclear properties [1], which are enhanced by the static octupole deformation that their ground state is suspected to have based on experiments with the neighboring even-A radium isotopes [2, 3]. The alkaline-earth radium atom and its molecules are also compatible with a wide variety of high-precision spectroscopic and trapping techniques, including laser-cooling in ion and magneto-optical traps, and high-precision fast-ion collinear laser spectroscopy, as developed for the lighter alkaline-earth elements [4]. As a result, atomic and molecular experiments with Ra are currently being pursued to search for signatures of, among other observables, the P-odd nuclear anapole moment [5, 6, 7, 8], reflecting a toroidal magnetic current in the nucleus, and the P, T-odd nuclear Schiff moment, which is theorized to arise from the collective effects of the proton and neutron electric dipole moments and hitherto unknown T-odd properties of the nucleon-nucleon interaction [9, 10]. An upper limit to an atomic electric dipole moment induced by the nuclear Schiff moment in ²²⁵Ra has already been set at $|d(^{225}\text{Ra})| < 1.4 \times 10^{-23} \ e \ \text{cm} \ [6, 11]$, with further improvements envisioned by the same collaboration and future molecular experiments [12].

Experimental searches for nuclear P, T violation aim to measure P, T-odd properties of an atom or molecule that are induced by the symmetry-violating nuclear properties [1]. Interpreting an experimental bound as a constraint on nuclear P, T violation and further relating it to the fundamental forces thus requires two sets of calculations. Electronic structure calculations attempt to compute the sensitivity of atoms and molecules to the P, T-odd nuclear moments, and nuclear structure calculations compute the relation to sources of symmetry violation in the fundamental forces.

The required nuclear calculations have traditionally suffered from large uncertainties, even in the order of 100% of the nominal value [13], especially for the mean-field calculations [14] that are applicable to heavy nuclei like ^{223,225}Ra. The Schiff moment, however, has a strong correlation with static octupole deformation and an explicit dependence on the absolute mean-squared nuclear charge radius $\langle r_{\rm ch}^2 \rangle$ [15], while the anapole moment depends entirely on the spatial distribution of the nuclear current density $\mathbf{j}(r)$ [1]. As a result, measurements of $\langle r_{\rm ch}^2 \rangle$ and $\mathbf{j}(r)$ could provide important input to test the reliability of the nuclear models applicable to ^{223,225}Ra, and they would support the intrinsic-frame calculations of the respective Schiff and anapole moments [15].

Laser spectroscopy provides measurements of the difference in atomic transition frequencies between isotopes A and A', referred to as the isotope shift $\delta \nu^{A,A'}$, which can be used to extract the nuclear-model-independent difference in mean-squared nuclear charge radii $\delta \langle r_{ch}^2 \rangle^{A,A'}$ with input from atomic structure calculations for the isotope-shift factors F and K. To extract the absolute radius $\langle r_{ch}^2 \rangle$ of any isotope A using $\delta \langle r_{ch}^2 \rangle^{A,A'}$, a non-optical measurement of $\langle r_{ch}^2 \rangle$ for at least one reference isotope A' is necessary. Until recently, all isotopes of radium were out of reach for non-optical techniques. However, with the commissioning of SCRIT for electron scattering with radioactive nuclides [16], and muonic X-ray spectroscopy of long-lived unstable isotopes with muX at Paul Scherrer Institute [17], the first non-optical measurement of $\langle r_{\rm ch}^2 \rangle$ for the long-lived ²²⁶Ra can be envisioned within the next few years.

Given a non-optical measurement of $\langle r_{\rm ch}^2 \rangle$ for ²²⁶Ra, extracting the charge radii of the isotopes ^{223,225}Ra requires high-precision isotope-shift measurements for an electronic transition whose isotope-shift factors F and K can be calculated with the smallest theoretical uncertainty possible. For this purpose, the transitions $7s^2S_{1/2} \rightarrow 7p^2P_{1/2}$ and $7s^2S_{1/2} \rightarrow 7p^2P_{3/2}$ in Ra⁺ are ideal; the single-valence-electron structure of these states allows for high-precision state-of-the-art single-reference relativistic coupled cluster calculations to reach a total theoretical uncertainty even better than the level of few percent that was recently demonstrated on the similar but more complex structure of neutral thallium [18].

In addition to the high-precision extraction of $\delta \langle r_{\rm ch}^2 \rangle$, spectroscopy of these two transitions offers an exciting opportunity to study the Bohr-Weisskopf (BW) effect in the hyperfine structure (hfs) of ^{223,225}Ra, recently studied in the RaF molecule [19]. The BW effect is a contribution to the magnetic dipole hyperfine splitting of an electronic state due to the non-uniform distribution of magnetization within the atomic nucleus [20]. Therefore, its measurement provides model-independent insight into the distribution of nuclear current density $\mathbf{j}(r)$, which would also constrain the calculations necessary to interpret nuclear anapole moment measurements.

The magnetic dipole splitting in the atomic hfs is quantified by the hyperfine A-factor:

$$A = g \frac{B}{J} \mu_B \tag{1}$$

where g is the nuclear g-factor, μ_B is the Bohr magneton, B is the atomic magnetic field at the location of the nucleus due to the electronic motion, and J is the total electronic angular momentum of the electronic state. The experimentally measured A-factor for an electronic state can be expressed as [21, 22]:

$$A_{\rm exp} = A_{\rm (0)} + A_{\rm QED} + A_{\rm BW} \tag{2}$$

Here, $A_{(0)}$ is the contribution due to the idealized point-nuclear magnetic dipole interaction, A_{QED} is the correction to $A_{(0)}$ due to quantum electrodynamic effects, and A_{BW} is the contribution due to the BW effect, representing the deviation from an ideal dipole interaction with a uniformly distributed nuclear magnetization. Ab initio quantum chemistry can calculate the electronic motion that produces $(B_{(0)} + B_{\text{QED}})/J$. As a result, provided that an independent and non-optical measurement of the nuclear g-factor is available such that $A_{(0)}$ and A_{QED} can be extracted using $(B_{(0)} + B_{\text{QED}})/J$ as per Eqn. 1, A_{BW} can then be deduced via the measured A_{exp} as:

$$A_{\rm BW} = A_{\rm exp} - A_{(0)} - A_{\rm QED}$$
 (3)

 $A_{\rm BW}$ is in turn proportional to the integral of the product of the electronic structure radial functions g(r) and f(r) of the large and small components of the Dirac bispinor, which can be calculated with high precision for Ra⁺, and the spatial distribution of nuclear magnetization F(r) as:

$$A_{\rm BW} \propto \int_0^R g f [1 - F(r)] dr \tag{4}$$

where R is the nuclear radius. Consequently, extracting A_{BW} using state-of-the-art electronic structure theory and measurements of A_{exp} can provide a nuclear-model-independent constraint on F(r), which can in turn constrain calculations of $\mathbf{j}(r)$ that are relevant to the nuclear anapole moment.

The magnitude of $A_{\rm BW}$ is not the same for all electronic states of an atom or ion, as certain states can be highly sensitive or insensitive to deviations from the point-nuclear magnetic dipole interaction. The magnitude of the BW effect in the hyperfine splitting of the $7s^2S_{1/2}$, $7p^2P_{1/2}$, and $7p^2P_{3/2}$ states in Ra⁺ has already been calculated in Ref. [21], along with the expected BW effect in the ground and first excited states of RaF. The accuracy of these calculations were recently benchmarked with measurements in RaF, and were found to be in excellent agreement with experiment [23]. As per Ref. [21], $A_{\rm BW}$ in the case of 225 Ra⁺ amounts to 4.3% for the $7s^2S_{1/2}$ state, 1.4% for $7p^2P_{1/2}$, and 0.4% for $7p^2P_{3/2}$.

Non-optical measurements of the nuclear g-factor are available only for 213,225 Ra, measured with in-beam magnetic resonance [24]. To extract the g-factor in the absence of the BW effect and then in turn deduce $A_{\rm BW}$ for other odd-A isotopes of radium, including 223 Ra, we propose performing high-precision fast-beam collinear laser spectroscopy of the $7s^2S_{1/2} \rightarrow 7p^2P_{3/2}$ transition in Ra⁺. According to the calculations in Ref. [21] that the BW effect amounts to only 0.4% of the dipole splitting in the $7p^2P_{3/2}$ state, it is reasonable to approximate:

$$A_{\rm exp}^{P_{3/2}} \approx A_{(0)}^{P_{3/2}} + A_{\rm QED}^{P_{3/2}} = g \frac{B_{(0)} + B_{\rm QED}}{3/2}$$
(5)

and consider a 0.4% systematic uncertainty in the extracted g-factor. Afterwards, the extracted g-factor can be used along with the measured $A_{\exp}^{S_{1/2}}$ for the $7s^2S_{1/2}$ state, which is x10 more sensitive to the BW effect, to extract $A_{BW}^{S_{1/2}}$ and F(r) for any isotope of radium, without the need for a non-optical measurement of the g-factor.

The proposed scheme to determine both the absolute charge radius $\langle r_{\rm ch}^2 \rangle$ and the magnetization distribution F(r) in 223,225 Ra requires high-precision measurements of the isotope shifts and hfs in the $7s\,{}^2S_{1/2} \rightarrow 7p\,{}^2P_{1/2}$ and $7s\,{}^2S_{1/2} \rightarrow 7p\,{}^2P_{3/2}$ transitions in Ra⁺. While high-resolution spectroscopy has been reported for many electronic states of neutral and ionic Ra [25], the low-lying states in Ra⁺ have the simplest electronic structure and are thus compatible with the highest possible precision in electronic structure calculations [21].

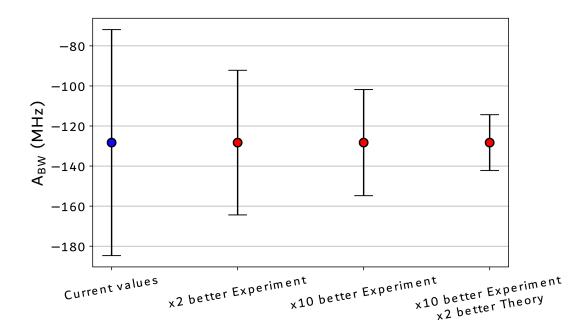


Figure 1: Expected improvement in the precision of $A_{\rm BW}$ for the $7s^2S_{1/2}$ state in ²²³Ra⁺, as a function of improvements in the statistical and systematic uncertainties of the experimental A factors and the electronic-structure calculations compared to the currently available values from Refs. [26, 21].

The only available isotope-shift and hyperfine-structure measurements of these transitions across a range of radium isotopes come from experiments at ISOLDE in the late 1980s [27, 26]. By repeating these measurements, we aim to significantly improve the statistical and systematic uncertainties in the isotope shifts and A-factors. Fig. 1 shows the expected improvement in the determination of $A_{\rm BW}$ for the $7s^2S_{1/2}$ state in $^{223}{\rm Ra^+}$, where non-optical measurements of the nuclear g-factor are not available, and thus it has to be extracted from the hfs A-factor of $7p^2P_{3/2}$. Thanks to technical advancements in the COLLAPS setup since the experiments that set the currently available experimental values, we expect to improve the uncertainty of the isotope shift and the hfs by approximately a factor of 10. In particular, improvements in the determination of the ion beam kinetic energy at the 10 ppm level and of the laser frequency at 2 MHz will drive this gain in precision. Moreover, improvements in electronic-structure theory are expected to refine the calculations of $(B_{(0)} + B_{\rm QED})/J$ by an additional factor of 2. Following our proposed improvement in the experimental uncertainties, the error on $A_{\rm BW}$ will be fully dominated by the theoretical uncertainty.

We propose performing measurements only on the long-lived radium isotopes $^{223-226,228}$ Ra, which are available from pre-irradiated targets. Follow-up experiments to determine the BW effect across all odd-A isotopes of radium from the spectroscopy of $7s^2S_{1/2} \rightarrow 7p^2P_{3/2}$ in Ra⁺ can be envisioned in the future.

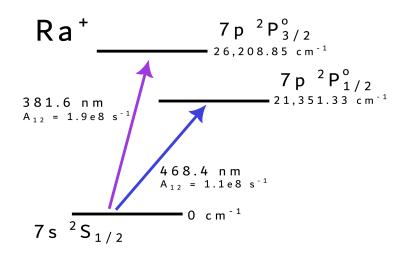


Figure 2: Optical transitions in Ra⁺ for the proposed measurements.

2 Proposed measurements

We propose the repetition of laser spectroscopy measurements of the $7s^2S_{1/2} \rightarrow 7p^2P_{1/2}$ $(\lambda = 468.2 \text{ nm}, A_{21} = 1.1 \times 10^8 \text{ s}^{-1})$ and $7s^2S_{1/2} \rightarrow 7p^2P_{3/2}$ $(\lambda = 381.4 \text{ nm}, A_{21} = 1.9 \times 10^8 \text{ s}^{-1})$ transitions in ^{223-226,228}Ra⁺ similar to Ref. [26], as shown in Fig. 2.

Similar to previous COLLAPS experiments [28, 29], the surface- or laser-ionized Ra⁺ will be transported from the HRS target station to ISCOOL in order to provide bunched beams to the COLLAPS collinear laser spectroscopy setup for high resolution study. Here, the ions are superimposed with a co-propagating continuous-wave laser beam. The laser frequency can be tuned in the ion's rest-frame by applying a voltage to the floated optical detection region. On resonance, the ions are excited to a higher electronic state and the subsequent fluorescence light is detected with photomultiplier tubes. The recorded signal will be time-gated to the bunched ion beam, resulting in a photon background reduction by several orders of magnitude. For further details on the experimental setup, we refer to Ref. [30].

3 Shift estimates

Since the isotopes of interest $^{223-226,228}$ Ra are either sufficiently long-lived or fed from long-lived Th isotopes, we propose to use a previously irradiated UC_x target for an experiment without active proton irradiation. Since Ra can be surface-ionized, RILIS would be beneficial but not strictly necessary. This makes the experiment an ideal candidate for the 'winter physics' part of the ISOLDE program. The limiting isotope is 223 Ra due to its comparably short lifetime of 11.4 days. In addition to that, also the mother nucleus has only a lifetime of 18.7 days. However, if the experiment is scheduled immediately after the end of the proton running period, the yields of 223 Ra will be sufficiently high for the experiment. We expect the yields of each isotope to be at the level of a few pA, as estimated from the yields of the latest RaF experiments of the CRIS setup and the fluorination efficiency of Ra. This will be well above the sensitivity limit (> 10^5 ions/s) of the COLLAPS setup.

In order to measure each isotope with sufficient statistics and accuracy in two transitions, we request two shifts for each isotope. Additionally, we request one shift for setup optimization, and two shifts for the investigation of systematic uncertainties.

Summary of requested shifts: 13 shifts delivered in 1 run using a previously irradiated UC_x target without active proton irradiation (winter physics).

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4 Details for the Technical Advisory Committee

4.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

- \boxtimes Permanent ISOLDE setup: COLLAPS
 - \boxtimes To be used without any modification
 - \Box To be modified: Short description of required modifications.

HAZARDS GENERATED BY THE EXPERIMENT Hazards named in the document relevant for the fixed COLLAPS installation.