CERN-INTC-2024-053 / INTC-CLL-059
02/10/2024

Letter of Clarification - INTC-P-699

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

October 2, 2024

P. Imgram[1], O. Ahmad[1], M. Athanasakis-Kaklamanakis[1,2], K. Blaum[3], H. Bodnar[5], B. Cheal[4], S. Dutton[3], T. Fabritz[5], C. M. Fajardo-Zambrano[1], R. F. Garcia Ruiz[6], J. Hughes[4], F. Koehler[5], K. Koenig[5], T. Lellinger[3,7], B. Maaß[5,8], E. Matthews[5], P. Mueller[5], R. Neugart[3], G. Neyens[1], W. Noertershaeuser[5], J. Palmes[5], P. Plattner[3], L. Renth[5], L. V. Rodriguez[3,7], R. Sanchez[9], J. Spahn[5], X. F. Yang[10], D. T. Yordanov[11]

[1] *Instituut voor Kern- en Stralingsfysica, KU Leuven, Leuven, Belgium.*
[2] *Centre for Cold Matter, Imperial College London, London SW7 2BW, UK.*
[3] *Max-Planck-Institut fuer Kernphysik, Heidelberg, Germany.*
[4] *Oliver Lodge Laboratory, University of Liverpool, UK.*
[5] *Institut fuer Kernphysik, Technische Universitaet Darmstadt, Darmstadt, Germany.*
[6] *Massachusetts Institute of Technology, Cambridge, MA, USA.*
[7] *Experimental Physics Department, CERN, Geneva, Switzerland.*
[8] *Physics Division, Argonne National Laboratory, Lemont, IL 60439, USA*
[9] *GSI Helmholtzzentrum fuer Schwerionenforschung GmbH, Darmstadt, Germany.*
[10] *School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing, China.*
[11] *Universite Paris-Saclay, CNRS/IN2P3, IJCLab, 91405 Orsay, France.*

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

Comments from INTC

The proposal aims to conduct laser spectroscopy on the D1- and D2-like transitions in $^{223-226,228}$*Ra*$^{+}$ *using COLLAPS to investigate symmetry violation in fundamental interactions. The proponents seek to enhance both statistical and systematic uncertainty by a factor of 10 on the isotope shift and a factor of 2-10 on the hyperfine parameters compared to previous measurements. This improvement, coupled with refined electronic-structure calculations, is expected to significantly reduce uncertainty in determining the magnitude of the Bohr-Weisskopf (BW) effect and the differential nuclear charge radius. While the*

INTC acknowledges the proposal's physics relevance, it requests a letter of clarification addressing specifically the following points.

Firstly, we would like to thank the INTC members for their careful review and the acknowledgment of the proposal's physics relevance. We will address the individual points below.

1. What are the possibilities for the performing the measurements during the online period i.e. with protons rather than winter physics? What levels of Fr contamination are tolerable?

The amount of Fr (or any other) contamination that can be handled in the foreseen experiment is defined by two main factors: the capacity of ISCOOL and RP. ISCOOL can handle 1e9 ions/s in the maximum limit, but to prevent overfilling the buncher, we would rather use it at roughly 1e7 ions/s or even lower. Overfilling the buncher results in large systematic uncertainties in our experiment, so we have to be very careful with the number of ions/s. The TAC recommended a reduction of the Ra rates to 7e5 ions/s for RP reasons. This is then also the rate we want to use for our experiment. Hence, if the total contamination rate is a factor < 10 higher than the Ra rate, it might be tolerable for ISCOOL. From the ISOLDE yield database, we estimate a contamination of 4e5 ions/s for ^{226}Fr, 9e5 ions/s for ^{225}Fr, 1.4e6 ions/s for ^{224}Fr and 6e6 ions/s for ^{223}Fr with a UC$_x$ target. This would mean, that the Fr contamination might be tolerable regarding the overfilling of ISCOOL (in case the Fr yield is not much higher than estimated here). However, the additional radiation from these contaminants (Fr) will add to the radiation from Ra. We cannot say if this tolerable from RP side, especially regarding 100xLA.
In an offline run during winter physics, all the aforementioned potential problems will not appear since we don't expect any Fr contamination with a pre-irradiated target.

2. In case the run needs to be performed in the offline period, please present detailed irradiation requirements so the effect on the physics programme due to the need to pre-irradiate a target can be estimated.

In the initial proposal, we requested a few pA (1e7 ions/s) of Ra beam. For this, the TAC estimated 9 shifts of direct radiation for 13 shifts of experiment. However, since we are limited to 7e5 ions/s for RP reasons, the necessary irradiation time might be shorter. The total time can be estimated by the TAC.

3. How will the measurement of differential charge radii be used by theory? What nuclear models, that are relevant for future studies of PT violation in fundamental interactions, can be expected to benefit from this measurement?

In a nucleus with a low-lying excited state $\bar{\Psi}_0$ of the same angular momentum and opposite parity as the nuclear ground state Ψ_0, the P, T-odd nuclear Schiff moment

S in the laboratory frame can be expressed as [1]:

$$S \approx \frac{\langle \Psi_0 | \hat{S}_0 | \bar{\Psi}_0 \rangle \langle \bar{\Psi}_0 | \hat{V}_{PT} | \Psi_0 \rangle}{\Delta E} \tag{1}$$

where ΔE is the energy difference between Ψ_0 and $\bar{\Psi}_0$, $\hat{S}_0$ is the Schiff operator, and $\hat{V}_{PT}$ is the operator of the P, T-violating nuclear potential. The Schiff operator has the form [2, 3]:

$$\hat{S}_0 = \frac{e}{10}\sqrt{\frac{4\pi}{3}} \sum_i^Z \Big(r_i^3 - \frac{5}{3}\langle r^2 \rangle_{\mathrm{ch}} r_i \Big) Y_0^1(\Omega_i) + ... \tag{2}$$

where the omitted terms are neglected. The index i denotes protons, r_i is the proton radial coordinate, $\langle r^2 \rangle_{\mathrm{ch}}$ is the absolute mean-squared nuclear charge radius, and Y_0^1 is a spherical harmonic of the proton distribution.

To interpret precision experiments and place limits on the P, T-violating nucleon-pion and meson-exchange interactions within $\hat{V}_{PT}$, nuclear theory is used to calculate $\langle \Psi_0 | \hat{S}_0 | \bar{\Psi}_0 \rangle$ and $\langle \bar{\Psi}_0 | \hat{V}_{PT} | \Psi_0 \rangle$ and to express S in the simpler, parametric form:

$$S = a_0 g \bar{g}_0 + a_1 g \bar{g}_1 + a_2 g \bar{g}_2 + b_1 \bar{c}_1 + b_2 \bar{c}_2 \tag{3}$$

where g is the strong πNN coupling constant, $\bar{g}_0$, $\bar{g}_1$, and $\bar{g}_2$ are the isoscalar, isovector, and isotensor P, T-odd nucleon-pion coupling constants, and $\bar{c}_1$, $\bar{c}_2$ are coupling constants of a short-range interaction resembling the effects of a P, T-odd heavy-meson exchange interaction [1].

Nuclear structure calculations of the sensitivity constants a_i and b_i can typically have uncertainties that reach up to 100% [4]. Dobaczewski *et al.* [1] demonstrated that the theoretical uncertainties in a_i and b_i can be significantly reduced by taking advantage of a correlation between $\hat{S}_0$ and the intrinsic electric octupole moment Q_3 of the nucleus. This correlation between observables is leveraged in the following way [1]: nuclear density functional theory (DFT) calculations are used to calculate the Schiff moment of a target nucleus A and also the electric octupole moment Q_0^3 of a different nucleus where a measurement of that moment exists, such as ^{226}Ra [5]. The DFT calculations are repeated for several different functionals, and the results populate a plot of calculated Schiff moments of nucleus A versus calculated $Q_0^3(^{226}\mathrm{Ra})$. Each functional thus provides a single data point on the plot. Linear regression analysis of the results provides the best-fit parameters for the calculated Schiff moment of A versus $Q_0^3(^{226}\mathrm{Ra})$, and by plugging in the measured value of $Q_0^3(^{226}\mathrm{Ra})$, a functional-independent calculated value of the Schiff moment of nucleus A is extracted.

Even by taking advantage of this scheme, the calculations of the Schiff moment in ^{225}Ra in Ref. [1] showed that the uncertainty in a_0 and b_1 was more than 100%, while the uncertainty in a_1, a_2, and b_2 was $\geq$45%. Further reduction in the theoretical uncertainty could be achieved if a similar correlation analysis could be carried out based on the absolute mean-squared nuclear charge radius $\langle r^2 \rangle_{\mathrm{ch}}$ of nucleus A itself, instead of Q_0^3 of a different nucleus, such as ^{226}Ra. Recent work on calculations of the Schiff moment of ^{227}Ac[1] indicates that a strong correlation between the Schiff moment and $\langle r^2\rangle_{\text{ch}}$ exists, as expected considering the presence of $\langle r^2\rangle_{\text{ch}}$ in the definition of the Schiff operator (Eq. 2). In the case of ^{227}Ac, the correlation appears to be high enough such that a 5% deviation in the value of $\langle r^2\rangle_{\text{ch}}(^{227}\text{Ac})$ leads to a change in the calculated value of the intrinsic Schiff moment of ^{227}Ac by a factor of 2. The impact on the laboratory Schiff moment is under investigation and may differ.

Currently, no absolute charge radii measurements have been reported for isotopes of Ra, but the muX collaboration [6] is actively working towards the first absolute charge-radius measurement of ^{226}Ra by muonic X-rays. To take full advantage of the additional correlation analysis between the Schiff moment and $\langle r^2\rangle_{\text{ch}}$, the charge radius of the nucleus whose Schiff moment is calculated should be used. Therefore, high-precision isotope-shift measurements are necessary to support the nuclear DFT calculations of the Schiff moment of ^{223,225}Ra.

4. How will the measurement of the Bohr-Weisskopf (BW) effect be used by theory? What nuclear models, that are relevant for future studies of PT violation in fundamental interactions, can be expected to benefit from this measurement?

The all-optical measurement of the Bohr-Weisskopf effect in the hyperfine structure of $^{223,225}\text{Ra}^+$ will be used as a benchmark for calculations needed to interpret proposed atomic and molecular parity violation (PV) experiments with ^{223,225}Ra. Developments in the accuracy of atomic theory are ongoing in an effort to interpret PV measurements in ^{133}Cs and extract the weak nuclear charge Q_{w} with high-enough precision to test the Standard Model [7, 8, 9, 10, 11, 12, 13, 14], and comparison with experiment across a number of observables has played a key role in these theoretical developments [15].

Precise measurements of the hyperfine structure can benchmark calculations of the atomic and molecular wavefunctions in the vicinity of the heavy nucleus, where the weak interaction takes place and thus where theoretical accuracy is most critical [15]. To probe the role of high-order contributions to the hyperfine structure, such as quantum electrodynamics effects, and thus to challenge the accuracy of their inclusion in calculations, experimental measurements of the BW effect play an important role. A lack of such measurements leads to systematic uncertainties in the hyperfine constants, and so the benchmark of high-order contributions to the hyperfine structure, whose magnitude is often in the same order as the neglected BW effect, is hindered.

The all-optical, absolute BW effect we propose to measure in $^{223,225}\text{Ra}^+$ provides information on the radial dependence of the nuclear magnetization across the nuclear volume. As an example, recent determination of the BW effect in the neighboring Fr

---

[1] Herlik Wibowo and Michail Athanasakis-Kaklamanakis, private communication, results currently in preparation.

isotopes by Roberts and Ginges [16] included such a comparison with different simplistic nuclear models, and highlighted that an empirical single-particle description of nuclear magnetization in Fr isotopes leads to good agreement with atomic theory.

In the case of $^{223,225}\mathrm{Ra}^{+}$, beyond assisting the benchmarks of atomic theory by comparing the hyperfine constants, we envision comparisons of the extracted BW effect with the results of nuclear theory using different models of nuclear magnetization distribution functions. Approaches that can reach this region of the nuclear chart are nuclear DFT and Monte Carlo Shell Model, with the former being highly successful in describing collective nuclear properties and the latter being sensitive to the single-particle structure.

References

[1] J. Dobaczewski, J. Engel, M. Kortelainen, and P. Becker. Correlating Schiff Moments in the Light Actinides with Octupole Moments. *Physical Review Letters*, 121(23):232501, 2018.

[2] J. Dobaczewski and J. Engel. Nuclear Time-Reversal Violation and the Schiff Moment of 225Ra. *Physical Review Letters*, 94(23):232502, 6 2005.

[3] J H de Jesus and J Engel. Time-reversal-violating Schiff moment of 199Hg. *Physical Review C*, 72(4):45503, 10 2005.

[4] Jonathan Engel, Michael J Ramsey-Musolf, and U van Kolck. Electric dipole moments of nucleons, nuclei, and atoms: The Standard Model and beyond. *Progress in Particle and Nuclear Physics*, 71:21–74, 2013.

[5] L. P. Gaffney, P. A. Butler, M. Scheck, A. B. Hayes, F. Wenander, M. Albers, B. Bastin, C. Bauer, A. Blazhev, S. Bönig, N. Bree, J. Cederkäll, T. Chupp, D. Cline, T. E. Cocolios, T. Davinson, H. De Witte, J. Diriken, T. Grahn, A. Herzan, M. Huyse, D. G. Jenkins, D. T. Joss, N. Kesteloot, J. Konki, M. Kowalczyk, Th Kröll, E. Kwan, R. Lutter, K. Moschner, P. Napiorkowski, J. Pakarinen, M. Pfeiffer, D. Radeck, P. Reiter, K. Reynders, S. V. Rigby, L. M. Robledo, M. Rudigier, S. Sambi, M. Seidlitz, B. Siebeck, T. Stora, P. Thoele, P. Van Duppen, M. J. Vermeulen, M. Von Schmid, D. Voulot, N. Warr, K. Wimmer, K. Wrzosek-Lipska, C. Y. Wu, and M. Zielinska. Studies of pear-shaped nuclei using accelerated radioactive beams. *Nature*, 497(7448):199–204, 2013.

[6] Frederik Wauters and Andreas Knecht. The muX project. *SciPost Physics Proceedings*, (5):022, 9 2021.

[7] V A Dzuba, V V Flambaum, and J S M Ginges. High-precision calculation of parity nonconservation in cesium and test of the standard model. *Physical Review D*, 66(7):76013, 10 2002.

[8] V M Shabaev, K Pachucki, I I Tupitsyn, and V A Yerokhin. QED Corrections to the Parity-Nonconserving 6s-7s Amplitude in 133Cs. *Physical Review Letters*, 94(21):213002, 6 2005.

[9] S G Porsev, K Beloy, and A Derevianko. Precision Determination of Electroweak Coupling from Atomic Parity Violation and Implications for Particle Physics. *Physical Review Letters*, 102(18):181601, 5 2009.

[10] S G Porsev, K Beloy, and A Derevianko. Precision determination of weak charge of 133Cs from atomic parity violation. *Physical Review D*, 82(3):36008, 8 2010.

[11] George Toh, Amy Damitz, Carol E Tanner, W R Johnson, and D S Elliott. Determination of the Scalar and Vector Polarizabilities of the Cesium 6s 2S1/2 - 7s 2S1/2 Transition and Implications for Atomic Parity Nonconservation. *Physical Review Letters*, 123(7):73002, 8 2019.

[12] B. K. Sahoo, B. P. Das, and H Spiesberger. New physics constraints from atomic parity violation in $ˆ{133}\mathrm{Cs}$. *Physical Review D*, 103(11):L111303, 6 2021.

[13] B. M. Roberts and J. S. M. Ginges. Comment on "New physics constraints from atomic parity violation in $ˆ{133}\mathrm{Cs}$". *Physical Review D*, 105(1):18301, 1 2022.

[14] G. Sanamyan, B. M. Roberts, and J. S. M. Ginges. Empirical Determination of the Bohr-Weisskopf Effect in Cesium and Improved Tests of Precision Atomic Theory in Searches for New Physics. *Physical Review Letters*, 130(5):53001, 2 2023.

[15] J. S.M. Ginges and V. V. Flambaum. Violations of fundamental symmetries in atoms and tests of unification theories of elementary particles. *Physics Reports*, 397(2):63–154, 7 2004.

[16] B M Roberts and J S M Ginges. Nuclear Magnetic Moments of Francium-207–213 from Precision Hyperfine Comparisons. *Physical Review Letters*, 125(6):63002, 8 2020.