EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

IS663: Rotational and Hyperfine Structure of RaF Molecules

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M. Athanasakis-Kaklamanakis[1,2], M. Au[3,4], S. W. Bai[5], Y. Balasmeh[2], R. Berger[6], A. A. Breier[7,8], K. Chrysalidis[3], T. E. Cocolios[2], C. M. Fajardo-Zambrano[2], V. N. Fedoseev[3], K. T. Flanagan[9,10], S. Franchoo[11], R. F. Garcia Ruiz[12], T. F. Giesen[8], D. Gonzalez-Acevedo[13,12], R. P. de Groote[2], D. Hanstorp[14], Á. Koszorús[2], L. Lalanne[1], P. Lassègues[2], Y. Liu[5], K. M. Lynch[9], B. A. Marsh[3], A. McGlone[9], W. C. Mei[5], G. Neyens[2], L. Nies[1,15], F. C. Pastrana Cruz[12], J. R. Reilly[9], S. Rothe[3], M. R. Tarbutt[16], J. Trujillo[2], S.-M. Udrescu[12], B. van den Borne[2], J. Warbinek[17], S. G. Wilkins[12], X. F. Yang[5]

[1] *Experimental Physics Department, CERN, CH-1211 Geneva 23, Switzerland*
[2] *KU Leuven, Instituut voor Kern- en Stralingsfysica, B-3001 Leuven, Belgium*
[3] *Systems Department, CERN, CH-1211 Geneva 23, Switzerland*
[4] *Department Chemie, Johannes Gutenberg-Universität Mainz, D-55099 Mainz, Germany*
[5] *School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing 100971, China*
[6] *Fachbereich Chemie, Philipps-Universität Marburg, 35032 Marburg, Germany*
[7] *Institut für Optik und Atomare Physik, Technische Universität Berlin, 10623 Berlin, Germany*
[8] *Laboratory for Astrophysics, Institute of Physics, University of Kassel, Kassel 34132, Germany*
[9] *School of Physics and Astronomy, The University of Manchester, Manchester M13 9PL, United Kingdom*
[10] *Photon Science Institute, University of Manchester, Manchester M13 9PY, United Kingdom*
[11] *Laboratoire Irène Joliot-Curie, Orsay F-91405, France*
[12] *Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, MA 02139, USA*
[13] *Department of Physics, Harvard University, Cambridge, MA 02138, USA*
[14] *Department of Physics, University of Gothenburg, Gothenburg SE-41296, Sweden*
[15] *Universität Greifswald, Institut für Physik, 17487 Greifswald, Germany*
[16] *Centre for Cold Matter, Imperial College London, SW7 2AZ London, United Kingdom*
[17] *GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291, Darmstadt, Germany*

Spokespersons: M. Athanasakis-Kaklamanakis, m.athkak@cern.ch
R. F. Garcia Ruiz, rgarciar@mit.edu

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S. G. Wilkins, wilkinss@mit.edu
Contact persons: L. Lalanne, louis-alexandre.lalanne@cern.ch
M. Athanasakis-Kaklamanakis, m.athkak@cern.ch

Abstract: In this addendum to IS663 (INTC-P-555), we propose to complete the spectroscopy of the hyperfine structure in ^{223,225}RaF using collinear resonance ionization spectroscopy. Knowledge of the hyperfine factors in RaF is crucial for devising experimental schemes for future precision tests of the Standard Model through symmetry-violating nuclear moments and for benchmarking state-of-the-art molecular theory.

Requested shifts: 19 shifts with a pre-irradiated target delivered in 1 run.

1 Motivation

The Standard Model (SM) of particle physics fails to explain a number of cosmological observations, such as the existence of dark matter and the imbalance of matter and antimatter in the Universe [1]. A multitude of theories that extend the SM have been proposed to date, attempting to explain these open cosmological questions.

Tabletop searches for leptonic, hadronic, and nuclear moments that violate parity (P) and/or time-reversal (T) symmetry hold great promise as precision tests of the SM [2]. The SM and its extensions give widely varying magnitudes to these symmetry-violating moments in nuclei and fundamental particles, and so a successful measurement of such a moment, like the electric dipole moment of the electron (eEDM), would provide a stringent benchmark of the limits of the SM and the validity of its extensions [3].

The most sensitive of these tabletop experiments search for signatures of symmetry-violating moments in the spectra of heavy polar molecules. At the shot-noise limit, the experimental precision scales with the sensitivity of the molecule to the moment of interest (for instance, the strength of the internal effective electric field E_{eff} for searches of the eEDM), the number of molecules probed in each measurement, and the coherence time of the measurement cycle. Radium monofluoride (RaF) has been suggested as a highly promising system for such experiments, as its internal E_{eff} is very strong, thanks to the high atomic number of Ra ($Z = 88$) [4], while also being amenable to direct laser cooling in a magneto-optical trap [5]. The latter promises significant improvements in coherence time and molecular number density, potentially surpassing the current state-of-the-art searches for the eEDM using ThO [6] and HfF^+ [7].

Moreover, the radium chain contains multiple long-lived isotopes with different spins ($I = 0, \frac{1}{2}, \frac{3}{2}$), several of which have a static octupole deformation [8, 9] and close-lying opposite-parity doublets ($\Delta E < 100$ keV). The reflection-asymmetric shape leads to enhanced symmetry-violating nuclear moments [10, 11, 12, 13], such as the nuclear Schiff and magnetic quadrupole moments, and thus RaF is a promising system to perform the first successful measurement of these nuclear moments across the chart of the nuclides.

High-precision spectroscopy of radioactive molecules has been identified as an important future direction for several research areas [14, 15, 16], but the radioactivity of the species complicates even the basic spectroscopy required to identify the molecular electronic states. As a result, almost no short-lived radioactive molecules proposed for precision tests of the SM have been studied in the laboratory as of yet, due to the technical difficulties.

The Collinear Resonance Ionization Spectroscopy (CRIS) experiment at ISOLDE is well-suited for the laser spectroscopy of radioactive molecules of importance for fundamental and applied science. Such spectroscopic studies are necessary to understand their electronic, vibrational, and rotational structure and to devise future high-precision experimental schemes. Measurements of the molecular hyperfine structure (HFS) are also

crucial for future searches of symmetry-odd nuclear moments with radioactive molecules.

2 Current status of spectroscopy of RaF

In 2018, the CRIS collaboration performed the first laser spectroscopy of RaF [17], measuring its low-lying electronic structure [18] and strong isotope shifts in its electronic-vibrational transitions [19] using broadband spectroscopy. The success of the initial experiment on short-lived radioactive molecules motivated a follow-up proposal for the study of RaF with narrowband spectroscopy (INTC-P-555), titled *Rotational and hyperfine structure of RaF molecules* [20].

All of the approved shifts were scheduled and multiple highlight results were obtained. The rotational structure of the $A\ ^2\Pi_{1/2} \leftarrow X\ ^2\Sigma_{1/2}$ laser-cooling transition was measured in ^{226}RaF in high resolution, obtaining spectra for both the $v' = 0 \leftarrow v'' = 0$ and the $v' = 1 \leftarrow v'' = 1$ vibrational transitions, which allows a potential laser-cooling scheme for RaF to be proposed for the first time [21].

Isotope shifts across $^{210,212-214,222-228,230}$RaF were measured with broadband spectroscopy, which provide a systematic understanding of how the nuclear radius manifests in molecular spectra across the longest isotopic range measured in a molecule to date, and how the choice of a laser scheme can influence the spectroscopy.

For ^{223,225}RaF, which contain a radium nucleus with a non-zero spin, the induced HFS results in the population spreading across a larger number of additional quantum states. As a result, the signal-to-noise ratio is reduced compared to the spectroscopy of ^{226}RaF, as less molecules are found in each substate. We thus developed the most efficient CRIS scheme for RaF that is compatible with the spectroscopy of ^{223,225}RaF through a systematic study of 15 excited electronic states. Additionally, comparing the excitation energies of the newly discovered high-lying states with the predictions of state-of-the-art relativistic Fock-space coupled-cluster theory allowed us to benchmark the predictive power of molecular theory at the part-per-ten-thousand level. Through this comparison, which highlighted the accuracy and precision of new developments in molecular theory, we studied the role of electron correlations and quantum electrodynamic effects on the electronic wavefunctions. A publication on these results is ready for submission [22].

During this work, we also determined the ionization potential of RaF with a comparable or better precision compared to measurements of the stable CaF, SrF, and BaF. This result compares closely with predictions by single-reference coupled-cluster theory, which was found to be in excellent agreement with the experiment, and which thus provides confidence in similar calculations of the chemical properties of other radioactive molecules, which is necessary for their optimal production at ISOLDE. A publication is ready for submission [23].

These laser-scheme developments allow us to study the HFS in ^{223,225}RaF with an

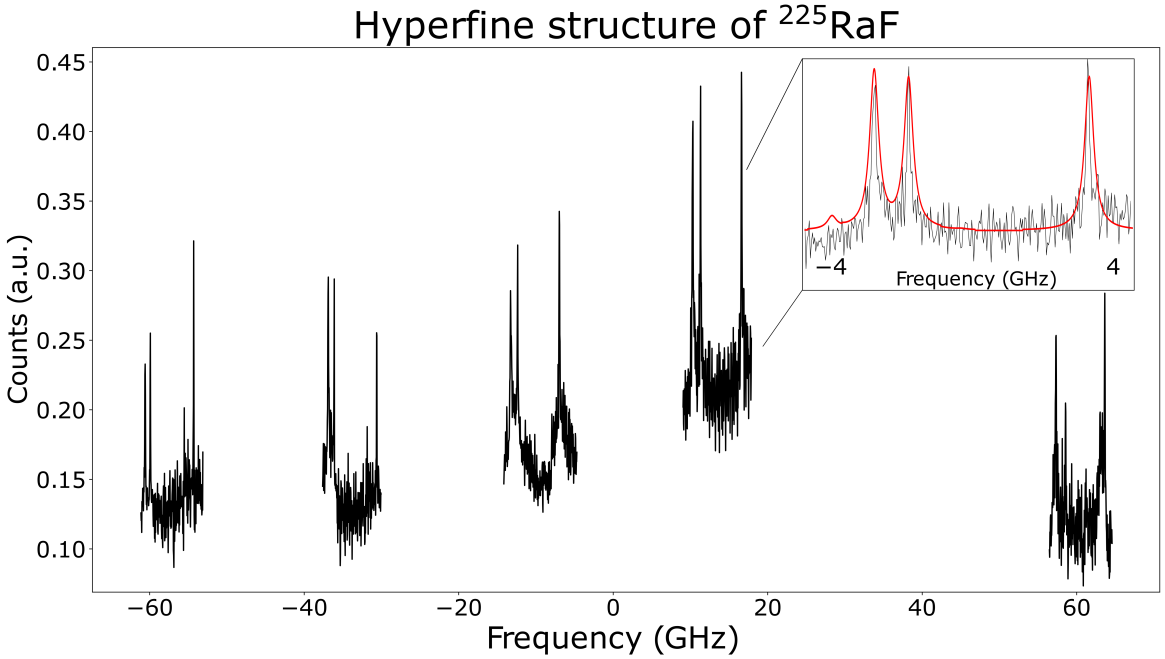

Figure 1: Hyperfine splitting of 5 rotational lines of the $A\ ^2\Pi_{1/2} \leftarrow X\ ^2\Sigma_{1/2}$ transition in ^{225}RaF, obtained during IS663 in 2021. A zoomed-in view of one rotational line is shown in the inset, including the simulated structure in red, created using the calculated values for the upper-state splitting from Ref. [24]. A fourth hyperfine peak is expected for each rotational line, located to the left of the measured structures, which was not resolved from the spectroscopic background in 2021.

improved signal-to-noise ratio. Thus, we demonstrated the ability to perform such measurements of the magnetic dipole hyperfine splitting in ^{225}RaF ($I_{\mathrm{Ra}} = 1/2$) with high-resolution spectroscopy. The statistics gathered in the spectroscopy of ^{225}RaF were enough to measure the dipole splitting of the $X\ ^2\Sigma_{1/2}$ ground state. The ground-state splitting was compared to the prediction of molecular theory [24], finding a remarkable agreement of more than 99%. This comparison already allows us to refine the calculated effective electric field E_{eff}, which determines the shot-noise precision of searches for the eEDM, improving the theoretical uncertainty by an order of magnitude compared to the literature. These measurements have also led to the first observation of the Bohr-Weisskopf effect in a molecule, which is a deviation from the ideal dipole approximation of the HFS due to the extended nuclear magnetization. An article based on these results is in preparation [25].

This work has also demonstrated that the HFS in ^{223}RaF can also be measured, including an estimate of the required measurement time, and thus an addendum is requested. Therefore, we request additional shifts within the framework of IS663 to complete these measurements.

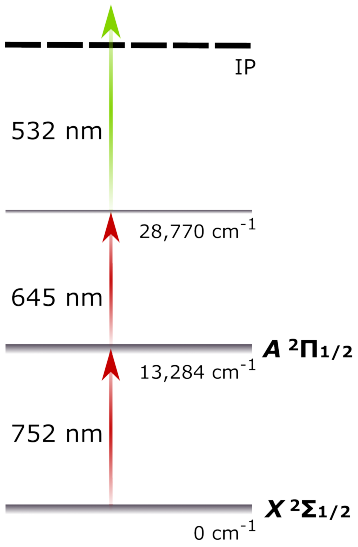

Figure 2: Resonance ionization scheme for the proposed study. The 752-nm step will be scanned with a narrowband pulsed injection-seeded titanium:sapphire laser.

3 Proposed additional study

We propose to measure the HFS of ^{223}RaF and ^{225}RaF in high resolution using the CRIS technique. Using the recently implemented voltage-scanning setup [26] that demonstrated a significantly improved data collection rate of $\times$3-6 in specific cases, we will be able to perform the measurement in a respectively shorter timescale by skipping unnecessary regions of background scanning.

^{225}Ra has a nuclear spin of $I = 1/2$ and thus shows only a magnetic dipole moment, while ^{223}Ra has a spin of $I = 3/2$ and thus also possesses a spectroscopic electric quadrupole moment. Studying the HFS in both isotopomers will therefore allow to unambiguously separate the dipole and quadrupole contributions to the molecular HFS in RaF. Knowledge of the HFS is crucial for devising high-precision experimental schemes for future searches of symmetry-violating nuclear moments with laser-cooled RaF.

From the analysis of the hyperfine spectra obtained during IS663 in 2021, we have deduced that measuring the hyperfine splitting of 7 rotational transitions will be sufficient to achieve a statistical uncertainty better than 0.5% in the $A_{\parallel}$ and $A_{\perp}$ molecular dipole splitting factors. Once these factors are known from the spectroscopy of ^{225}RaF, the quadrupole-splitting factor in ^{223}RaF can be extracted with a similar precision for the same number of measured lines.

Part of the spectra of the HFS in the rotational lines in ^{225}RaF that were measured in IS663 in 2021 are shown in Fig. 1. The spectra were recorded in the last 3 shifts of the experiment. The HFS of ^{225}RaF contains 4 peaks, but only 3 peaks were observed. The fourth hyperfine peak for each rotational line, which is expected to have a factor of 10 lower intensity than the other hyperfine peaks, could not be resolved from

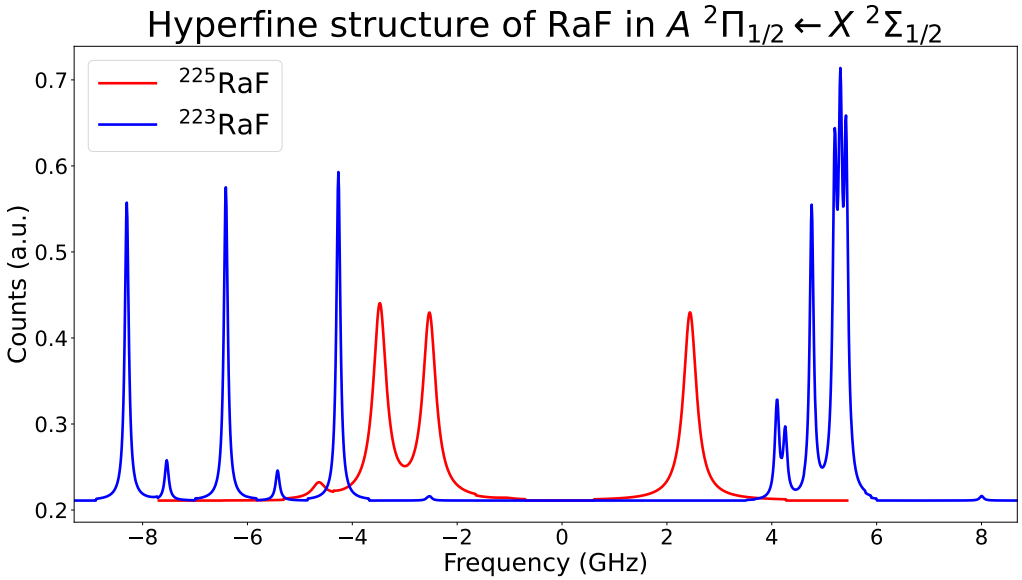

Figure 3: Comparison of simulated HFS for one rotational line in ^{223}RaF and ^{225}RaF for the $A\ ^2\Pi_{1/2} \leftarrow X\ ^2\Sigma_{1/2}$ transition.

the spectroscopic background within the available time. Measuring the position of the remaining hyperfine peaks in ^{225}RaF is necessary for the precise determination of the $A_{\|}$ and $A_{\perp}$ factors in the $A\ ^2\Pi_{1/2}$ state, which are important as a benchmark of quantum chemistry and for devising future experimental schemes. A simulation of the full HFS is shown in the inset of Fig. 1, using the calculated values for the upper-state splitting from Ref. [24].

We propose to use the laser scheme in Fig. 2 to complete the study of the HFS of ^{225}RaF and to study for the first time the quadrupole hyperfine interaction in ^{223}RaF. This scheme was also used to study the HFS in ^{225}RaF in 2021. Contrary to their atomic counterparts, molecular electronic states of $\Sigma_{1/2}$ and $\Pi_{1/2}$ character are sensitive to both the dipole and quadrupole hyperfine splittings. The scanning transition (752-nm) will be excited by the fundamental wavelength of the injection-seeded pulsed titanium:sapphire laser, and the spectrum will be obtained using the voltage-scanning apparatus [26]. The second step at 645 nm will be driven by a pulsed dye laser using DCM dye, and the non-resonant step at 532-nm will be produced by the second harmonic of a pulsed Nd:YAG laser.

4 Request

Based on our initial measurements of ^{225}RaF, in order to resolve the weakest peak in the structure we request 4 shifts for the spectroscopy of ^{225}RaF. The $\times$3-6 improvement in the data collection rate due to voltage scanning and the precise knowledge of the scanning regions ensure that the required statistics will be collected within the requested shifts.

The hyperfine splitting of ^{223}RaF, which is also sensitive to the quadrupole interaction (due to $I_{\mathrm{Ra}} = 3/2$), has more than twice as many lines in its HFS compared to ^{225}RaF (see Fig. 3). Therefore, we request 10 shifts in total, corresponding to more than 1 shift per rotational line with HFS, which is necessary to collect enough statistics to constrain the quadrupole splitting factor.

Additionally, we request 4 shifts for setup, target fluorination, and optimization using ^{226}RaF. An additional shift is requested for routine measurements of the $J = 16.5$ R-branch line in ^{226}RaF, acting as a reference measurement to track the evolution of systematics in the experiment.

Overall, we request 19 additional shifts without protons within the framework of IS663 to complete the goals outlined in INTC-P-555 [20]. Considering the long half-lives of ^{223,225,226}Ra and in-target feeding precursors, a pre-irradiated target can be used. Therefore, we request a UC_x target with a surface ion source and a fluorine leak for CF_4 injection, irradiated with protons prior to the study and thus enabling this experiment to take place outside of the period of proton availability at CERN.

Table 1: Summary of requested shifts with a pre-irradiated UC_x target with a surface ion source and a CF_4 leak.

	$T_{1/2}$	Shifts w/o p^+
^{223}RaF	11.4 d	10
^{225}RaF	14.9 d	4
^{226}RaF	1.6 ky	5
Total:		**19**

Summary of requested shifts: 19 shifts delivered in 1 run using a pre-irradiated UC_x target with a surface ion source and a CF_4 leak.

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DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
CRIS	☒ To be used without any modification ☐ To be modified
If relevant, describe here the name of the flexible/transported equipment you will bring to CERN from your Institute [Part 1 of experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[Part 2 of experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[insert lines if needed]	

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	

	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			