

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

## Rotational and Hyperfine Structure of RaF Molecules

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**Abstract:** High-resolution spectra of <sup>212,222–226,228,230</sup>RaF molecules will be measured using collinear resonance ionization spectroscopy. These measurements will allow high-precision isotope shifts, rotational constants, and hyperfine structure parameters of different RaF molecules to be extracted. These results will provide a comprehensive study of the electron-nucleus interaction in these molecules, and will yield essential experimental information that is needed for future uses of these molecules for fundamental physics research.



**Requested shifts:** 36 shifts (without protons) and 12 shifts (with protons) distributed in 2 runs (one with protons and one without protons).

## 1 Motivation

Molecular systems can offer highly sensitive probes to explore the violation of fundamental symmetries, and search for new physics physics beyond the Standard Model of particle physics [1, 2, 3]. In molecules, electron-nucleon and nucleon-nucleon interactions are dominated by the electromagnetic and strong force. The (parity-violating) weak force, although much smaller in magnitude, can add measurable contributions to molecular energy levels. Hence, precision measurements of their structure can provide measurements of parity- and time-reversal violation effects. Measurements of diatomic molecules, for example, have provided the most stringent constraint to the electron electric dipole moment (eEDM), constraining physics at energy scales comparable to those probed with the LHC [4]. EDM measurements of both leptons and hadrons provide constraints to different extensions of the Standard Model, and can shed light upon additional sources of time-reversal violation, which are needed to explain the origin of the matter-antimatter asymmetry in the universe.

The proximity of molecular states of different parity can enhance parity- and time-reversal violation effects by more than three orders of magnitude when compared to atomic systems. As these symmetry-violating effects scale with the atomic number, nuclear spin and nuclear deformation, molecules containing heavy radioactive nuclei are predicted to provide an exceptional sensitivity in these studies [5, 6, 7, 8]. However, experimental measurements of such radioactive systems were non-existent until recently, and in most cases quantum chemistry calculations constitute the only source of available information. This lack of experimental knowledge has motivated an ongoing experimental effort at ISOLDE focused on the study of radioactive molecules [9]. The first results from a successful experiment performed with radium monofluoride (RaF) molecules have just been accepted for publication [10]. These molecules have the potential to offer a unique laboratory for the study of nuclear electroweak properties and explore the violation of fundamental symmetries [6, 7].

In contrast to atomic systems, molecules possess additional degrees of freedom, with significantly more available states. At room temperature, atoms exist commonly in one or two possible electronic states. In molecules, vibrational and rotational levels can split the population in more than  $10^4$  states. Hence, their study requires particularly sensitive experimental techniques. The Collinear Resonance Ionization Spectroscopy (CRIS) experiment [11, 12, 13, 14, 15] combined with the versatility of the ISOLDE facility offers a unique facility worldwide to perform such studies.

The first proof of principle has allowed the extraction of basic spectroscopic properties of RaF, and has provided experimental evidence for their laser cooling scheme

[10]. These results are, to our knowledge, the first ever laser spectroscopy measurements performed with short-lived radioactive molecules, and constitute a pivotal step towards further precision studies of radioactive molecules. In addition to their marked interest for quantum chemistry and fundamental symmetries, the study of radioactive molecules opens up new opportunities for nuclear structure and astrophysics studies [5, 16, 7, 2, 17, 8].

## 2 Experimental details and recent results

The proof-of-principle experiment with RaF molecules was performed at the end of the last CERN running period, using a previously irradiated  $UC_x$  target, exposed to a total of  $8 \times 10^{17}$  protons. FLUKA [18] simulations predicted  $2 \times 10^{13}$  atoms of  $^{226}\text{Ra}$  in the target material. After irradiation, the target was kept in a sealed chamber filled with Ar gas during one month, and later connected to the HRS front end. At the HRS station, a  $\text{CF}_4$  gas was injected into the target. The target and ion source were heated, and RaF molecules were formed by reactive collisions of  $\text{CF}_4$  molecules with Ra atoms. Molecular ions ( $\text{RaF}^+$ ) were extracted and directed into the HRS to be mass separated and injected into the ISOLDE RFQ trap (ISCOOL). Bunches of  $\text{RaF}^+$  with a  $4\text{-}\mu\text{s}$  temporal width were released and accelerated to 40 keV, before entering into CRIS [19, 20, 21]. At CRIS, the molecules were neutralized in-flight by using a charge-exchange cell (CEC) loaded with a Na vapour [22]. Neutralized RaF molecules, were overlapped with two or more laser beams in a collinear geometry. A broadband or narrowband laser was used to resonantly excite the transition of interest, and subsequently, a high-power 355-nm laser pulse was used to ionize the RaF molecule into a  $\text{RaF}^+$  state. The resonantly ionized molecules were then separated from the non-interacting neutral molecules via electrostatic deflector plates and detected in a particle detector. The resonance ionization scheme used during the experiments is shown in Figure 1b. More details can be found in Ref. [10].

The first low-resolution results ( $\sim 3$  GHz) of the energetically low-lying electronic states of RaF have been accepted for publication [10]. An example of measured vibronic spectra of different isotopologues of RaF is shown in Figure 1. Further measurements of the RaF ionization potential, molecular isotope shifts, and preliminary high-resolution spectra (FWHM  $\approx 90(10)$  MHz) were obtained and are currently under analysis. An example of a high-resolution spectrum obtained for  $^{226}\text{RaF}$  is shown in Figure 2. At least two additional high-impact publications are expected from these results. However, complementary high-resolution data is needed to fully constrain the rotational and hyperfine structure parameters of these molecules.

## 3 Main objectives and beam time request

Building on the success of the aforementioned experiments, this proposal aims to obtain a complete high-resolution spectrum for different isotopologues of RaF. Measurements for molecules containing Ra isotopes with nuclear spin  $I = 0$  will allow the extraction of their

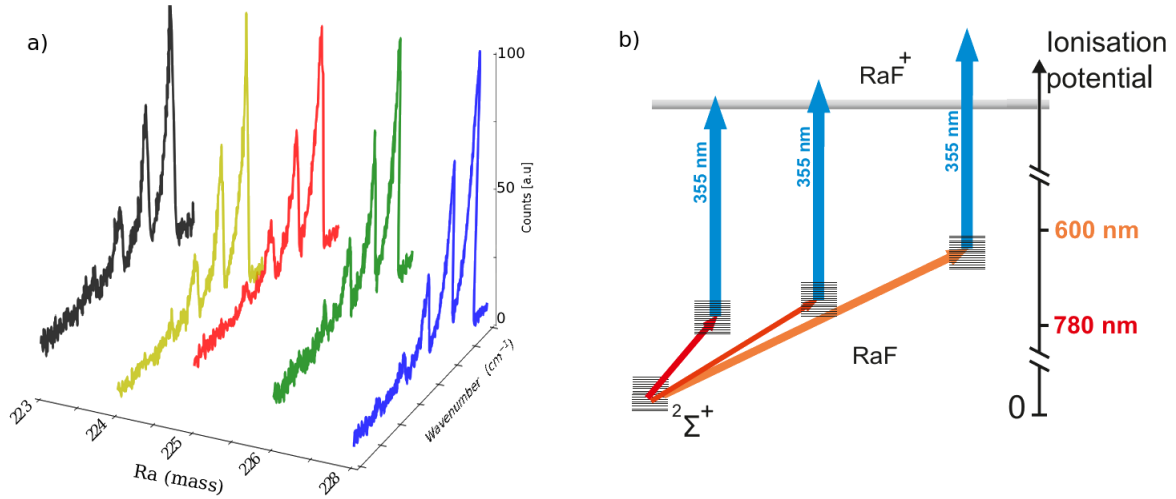


Figure 1: (Left) Low-resolution spectra of RaF molecules measured during the previous CRIS experimental campaign. The low-lying spectra  $^{223}\text{RaF}$  ( $T_{1/2} = 11.4$  d),  $^{224}\text{RaF}$  ( $T_{1/2} = 3.6$  d),  $^{225}\text{RaF}$  ( $T_{1/2} = 14.9$  d),  $^{226}\text{RaF}$  ( $T_{1/2} = 1600$  y) and  $^{228}\text{RaF}$  ( $T_{1/2} = 5.7$  y) are shown with different colors. (Right) Resonance ionization schemes used to study RaF molecules.

rotational parameter. In addition, hyperfine structure parameters will be obtained from measurements of molecules containing Ra isotopes with nuclear spin  $I > 0$ .

## Rotational structure of $^{226}\text{RaF}$

A production yield of about  $10^7$  ions/s was estimated for  $^{226}\text{RaF}^+$  during the last run. At this mass, about 8 counts per second were obtained at the resonant peaks observed in the high-resolution rotational spectra, with a background of 4 counts per second.

Figure 2 shows a partial high-resolution spectrum taken for  $^{226}\text{RaF}$  with a measuring time of about 2.5 hours, which is equivalent to a scanning speed of 1 MHz/second. In order to cover the P, Q and R molecular bands needed to fully define the rotational structure of this molecule, a region of about 300 GHz ( $\equiv 10$  cm $^{-1}$ ) should be scanned. **Hence, we require a total of 15 shifts using a previously irradiated target (without protons) to complete these measurements.** This beam time request takes into account reference scans and periodic interruptions needed to optimize the ion beam and laser beams.

## Hyperfine structure of $^{223}\text{RaF}$ and $^{225}\text{RaF}$

Diatomic molecules containing isotopes of nuclear spin  $I > 0$  have been proposed as highly sensitive systems to measure symmetry-violating nuclear properties [6, 7, 16]. Measurements with  $^{225}\text{Ra}$  ( $I = 1/2$ ), whose hyperfine structure does not exhibit a quadrupole interaction, would be important to unambiguously determine the magnetic hyperfine dipole interaction. On the other hand, hyperfine structure measurements of  $^{223}\text{Ra}$  ( $I = 3/2$ ) will allow the extraction of the electric quadrupole interaction. These measurements

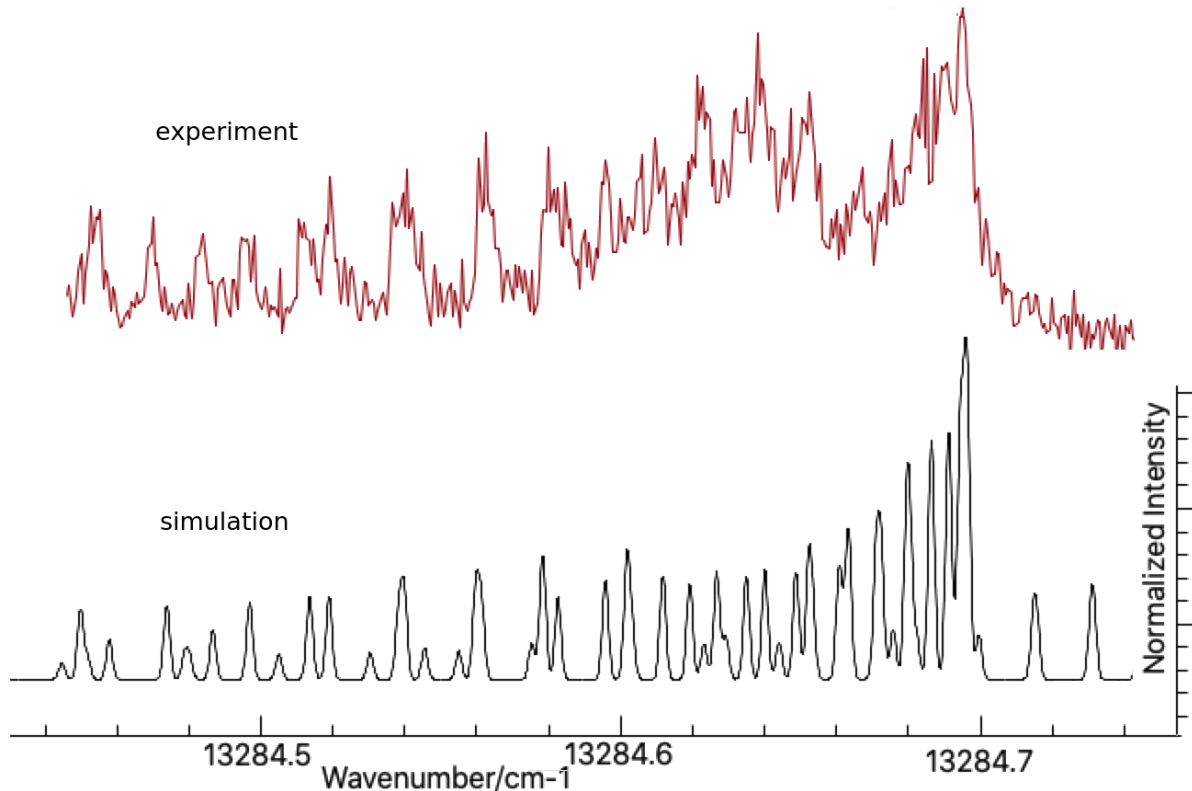


Figure 2: An example of a rotational spectrum measured for  $^{226}\text{RaF}$  with a resolution of 90(10) MHz is shown in the upper figure. The simulated molecular spectrum using PGOPHER [23], with preliminary fitted parameters, is shown in the lower figure.

will provide a complete determination of the components of the hyperfine tensor for the  $^{223,225}\text{RaF}$  molecules, which can be compared with the available theoretical predictions [7]. Simulations of hyperfine structure spectra expected for the isotopologues  $^{226}\text{Ra}(I = 0)\text{F}$ ,  $^{225}\text{Ra}(I = 1/2)\text{F}$ , and  $^{223}\text{Ra}(I = 3/2)\text{F}$  are shown in Figure 3. The simulations show a region corresponding to the R branch using PGOPHER [23], with predicted hyperfine parameters [7].

The experimental knowledge of the hyperfine structure parameters is essential for future higher precision studies with these molecules. In order to extract the hyperfine structure parameters, a frequency region of about 120 GHz ( $\equiv 4\text{ cm}^{-1}$ ) must be scanned for each molecule. **Thus, we estimate a total of 14 shifts (without protons) to accomplish these measurements.**

## Nuclear structure changes in isotopologues of RaF

In molecules that are favorable for laser cooling, the potential energy curves are very similar in the ground- and excited- state. This implies an ideal wavefunction overlap between vibrational states possessing the same vibrational quantum number. Thus, energy shifts in vibronic transitions due to use of different Ra isotopes are expected to be particularly sensitive to nuclear structure changes. Currently, there is limited experimental knowl-

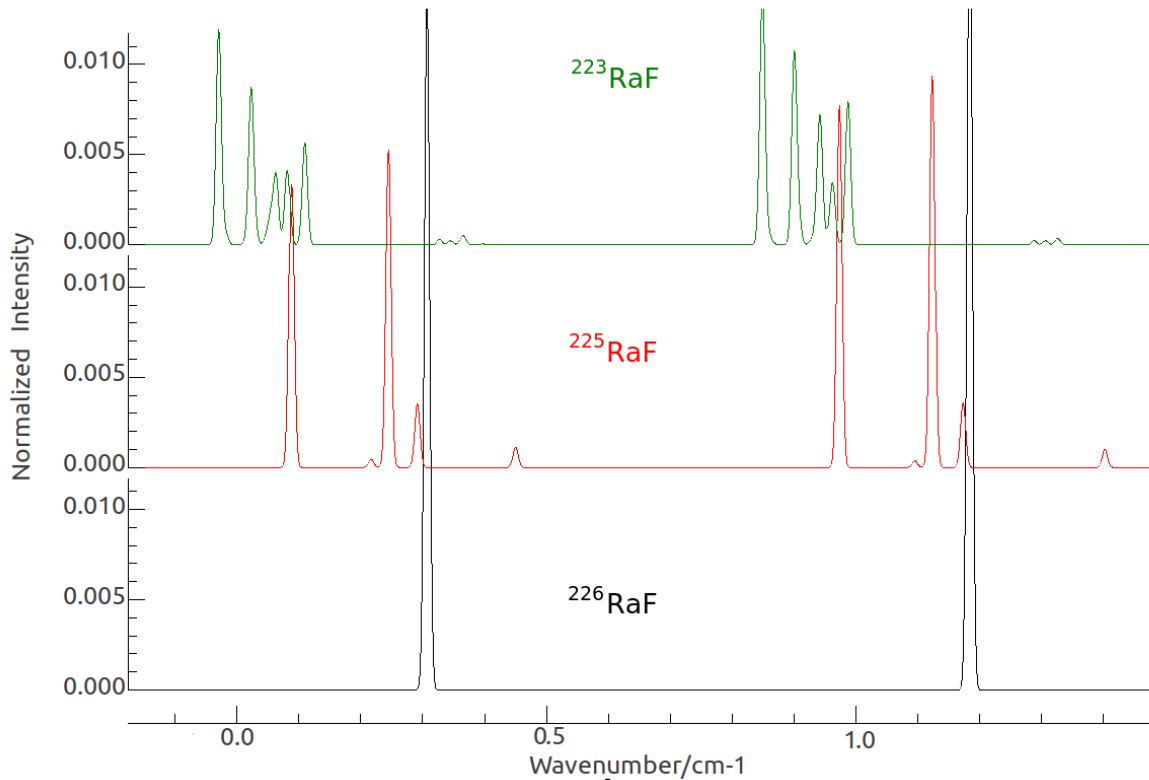


Figure 3: Simulations of hyperfine structure spectra expected for  $^{226}\text{Ra}(I = 0)\text{F}$ ,  $^{225}\text{Ra}(I = 1/2)\text{F}$ , and  $^{223}\text{Ra}(I = 3/2)\text{F}$  with a resolution of 300 MHz. The simulations show a region corresponding to the R branch using PGOPHER [23], with predicted hyperfine parameters [7].

edge about the nuclear effects on molecular transitions between different isotopologues [24]. High precision measurements of the isotopologues  $^{212}\text{RaF}$ ,  $^{222}\text{RaF}$ ,  $^{223}\text{RaF}$ ,  $^{224}\text{RaF}$ ,  $^{225}\text{RaF}$ ,  $^{226}\text{RaF}$ ,  $^{228}\text{RaF}$ , and  $^{230}\text{RaF}$ , will provide critical data to understand the impact of nuclear structure effects in electronic, vibrational and rotational transitions. Isotope shifts can be extracted from measurements of selected molecular states, which are estimated to be within a frequency region of about 60 GHz ( $\equiv 2 \text{ cm}^{-1}$ ) for each molecule. **We request 3 shifts for each molecule to perform these measurements.**

## Beam time request

The details of the beam production and the required shifts for this experimental proposal are summarized in Table 1. The motivation for the proposed measurements and beam time request are explained in the previous sections. In total, we request 36 shifts without protons in a single run. In addition, 12 shifts with protons (in a separate run) are requested to access to the most exotic short-lived molecules listed. Furthermore, investigations into the maximum deliverable intensity of the molecules of interest produced on-line will be undertaken. As the energy levels, ionization energy, and molecular parameters are well

Molecule	$I(\text{Ra}/\text{A})$	Half life	Yield (ions/s)	Shifts	Protons?
$^{138}\text{BaF}^+$	0	stable	$> 10^7$	1	no
$^{212}\text{RaF}^+$	0	13 s	$10^6$	3	yes
$^{222}\text{RaF}^+$	0	38 s	$10^6$	3	yes
$^{223}\text{RaF}^+$	3/2	11.4 d	$10^6$	7	no
$^{224}\text{RaF}^+$	0	3.6 d	$10^6$	3	no
$^{225}\text{RaF}^+$	1/2	15 d	$10^6$	7	no
				2	yes
$^{226}\text{RaF}^+$	0	1600 y	$10^7$	15	no
				1	yes
$^{228}\text{RaF}^+$	0	5.7 y	$10^6$	3	no
$^{230}\text{RaF}^+$	0	93 m	$10^6$	3	yes

Table 1: Reported yields and beam time request. A total of 36 shifts without protons, and 12 shifts with protons are requested. Beam intensities are estimated from the previous experimental campaign. The yields for the most exotic isotopes,  $^{212}\text{Ra}$ ,  $^{222}\text{Ra}$  and  $^{230}\text{Ra}$  were taken from the ISOLDE yield data base.

known for the stable  $^{138}\text{BaF}$  [25, 26, 27], this molecule can be used as a reference to test and optimize the experimental setup using the laser ionization scheme 860 nm + 355 nm [25, 27], and field ionization scheme 495 nm + 540 nm [28]. A field ionization setup has been recently installed at CRIS [29].

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# Appendix

## DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: (*name the fixed-ISOLDE installations, as well as flexible elements of the experiment*)

Part of the	Availability	Design and manufacturing
CRIS experiment	<input checked="" type="checkbox"/> Existing	<input checked="" type="checkbox"/> To be used without any modification

HAZARDS GENERATED BY THE EXPERIMENT (if using fixed installation:) Hazards named in the document relevant for the fixed CRIS installation.