EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Study of RaF^- anions at CRIS

April 9, 2024

R. F. Garcia Ruiz[1], O. Ahmad [3], M. Au [8], J. Berbalk [3] R. Berger [2], A. Brinson[1], T. E. Cocolios[3], R. P. de Groote[3], S. Ebadi[1], K. T. Flanagan[4] C. Fajardo[3], K. Gaul[2], D. Gonzalez[1], D. Hanstorp[5], P. Imgram[3], A. Jadbabaie[1], J. Karthein [1], Á. Koszorús[3], L. Lalanne[6], S. Moroch[1] J. M. Munoz[1] W. C. Mei[7] G. Neyens[3], M. Nichols[5] F. C. Pastrana Cruz[1], H. Perrett[4], S. Udrescu[1], J. Warbinek[6], S. G. Wilkins[1], X. F. Yang[7], C. Zülch[2],

[1]*Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA*
[2]*Fachbereich Chemie, Philipps-Universität Marburg, Marburg, 35032, Germany*
[3]*KU Leuven, Instituut voor Kern- en Stralingsfysica, B-3001 Leuven, Belgium*
[4]*School of Physics and Astronomy, The University of Manchester, Manchester M13 9PL, United Kingdom*
[5]*Department of Physics, University of Gothenburg, SE 412 96 Gothenburg, Sweden*
[6]*Experimental Physics Department, CERN, CH-1211 Geneva 23, Switzerland*
[7]*School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing 100971, China*
[6]*Accelerator Systems Department, STI group, CERN, CH-1211 Geneva 23, Switzerland*

Spokesperson: R. F. Garcia Ruiz [rgarciar@mit.edu]
K. T. Flanagan [kieran.flanagan-2@manchester.ac.uk]
Contact person: Jessica Warbinek [jessica.warbinek@cern.ch]

CERN-INTC-2024-030 / INTC-P-701
09/04/2024

Abstract: The production of mass-selected radium monofluoride anions, RaF^-, will be investigated by using two-step double charge exchange with a sodium vapor, following the reaction path $RaF^+ \rightarrow RaF \rightarrow RaF^-$. The measurements will be performed at the Collinear Resonance Ionization Spectroscopy (CRIS) experiment at ISOLDE. These measurements will enable us to assess the feasibility of a promising method for slowing down and trapping RaF molecules for future precision experiments.

Requested shifts: 12 shifts using an irradiated target, without protons. The experiment is proposed for the winter physics period.

1 Motivation and previous experiments

Molecules containing heavy, octupole-deformed nuclei, such as radium monofluride, RaF, have emerged as compelling candidates for exploring a plethora of fundamental questions in physics [1] in next-generation experiments. The rare octupole deformation of certain radium nuclei, combined with the symmetries of a suitable molecule, can massively enhance the sensitivity to both parity- and time-reversal violating effects by more than three orders of magnitude when compared with stable systems.

In recent years, the CRIS collaboration achieved the first-ever laser spectroscopy of a short-lived radioactive molecule—radium monofluoride (RaF) [2]. Building on this work, an extensive investigation into the structure of RaF has been undertaken, yielding valuable spectroscopic data on the hyperfine structure and isotope shifts of its molecular isotopologues [3, 4], and the excitation spectrum up to 30.000 cm^{-1}[5]. More recently, the transitions of a laser-cooling scheme for RaF [6] were experimentally determined [7], along with a measurement of the lifetime of the first excited state [8], paving the way for trapping and manipulating these molecules.

The current major challenge involves the efficient production of slow and cold RaF molecules with sub-Kelvin temperature, which are needed to enable efficient laser-cooling techniques. As the violation of fundamental symmetries will manifest as minute frequency shifts in the structure of the RaF molecule, the realization of such cooling techniques is an essential stepping stone towards realizing precision measurements in this system.

This Proposal aims to address this challenge by studying the production of RaF^- molecules as a possible pathway to decelerate and trap these molecules using electromagnetic fields. Subsequently, cold, slow neutral RaF molecules can be produced by photodetachment of RaF^-[9].

2 Objectives, experimental details and beam time request

Main objective: Production of RaF^-.

Starting from a positively charged ion beam of $^{226}RaF^+$, the production of which is well-established at ISOLDE, charge-exchange reactions can be employed to form negative ions [10]. We estimate that about 10^7 molecules per second of $^{226}RaF^+$ can be delivered to the CRIS setup [11].

At CRIS, the RaF^+ beam passes through the charge-exchange cell, containing a sodium (Na) vapor [12]. When the fast RaF^+ ions collide with Na, charge exchange occurs, forming the neutral molecule, RaF. We have used this approach to perform our previous experiments on RaF [2, 7]. However, the RaF molecules are produced with relatively large kinetic and internal energies, populating a large number of excited vibrational and rotational states, which poses a major challenge for precision experiments that rely on slow, cold molecules.

A common approach to achieve laser cooling with neutral molecules requires an initial slow, cold molecular beam, with velocities of less than 10 m/s, to be able to capture them in a Magneto-Optical Trap (MOT). In such experiments, slow neutral molecules

are produced from cryogenic buffer gas cells [13]. However, these techniques start with macroscopic samples (> gram), which are not applicable to radioactive materials that are typically available only in microscopic amounts (less than 1 nanogram). We have proposed an alternative approach, involving the production of negative RaF^- ions [9], which could potentially be decelerated and trapped in a cryogenic Paul trap. While also RaF^+ ions can be decelerated and trapped, the neutralization of this ion requires collisions with other atoms (e.g. Na) or electrons, resulting in a significant increase (>300 K) of both the kinetic and internal energy of the neutral molecule. In contrast to the neutralization of RaF^+ by collisions with Na atoms, photo-detachment of the anion offers a more gentle and controlled process, resulting in a smaller spread (< 4 K) in the distribution of final quantum states of the molecule [9].
Once trapped, both kinetic and internal energies can be reduced to less than a few Kelvin. Therefore, cold neutral RaF molecules could be formed by performing laser photodetachment of the trapped RaF^-. Such a scheme is expected to produce cold neutral molecules, RaF, in their lowest rovibrational states, which are the needed conditions to efficiently capture and trap in a MOT.
As we have demonstrated, the RaF^+ beam can capture one electron by collisions with Na to form neutral RaF in a quasi-resonant process [2]. The fast neutral RaF can then capture a second electron in a subsequent endothermic collision to form RaF^-. A sketch of the proposed scheme is illustrated in Fig. 1. The production yield of negative ions strongly depends on the vapor density [14, 15, 16], which we plan to study as part of this proposal.
To measure the production of RaF^-, an electrostatic field can be used to separate the negative ions from the neutral and positive components. The negative ions can be efficiently detected by a particle detector. All of the required instrumentation is already available at CRIS. The experimental scheme to produce negative ions was recently demonstrated for the production of $^{238}U^-$ anions at the experiment [10].
A detailed study of the properties of RaF^- and the approach proposed in this letter can be found in Ref [9].

Beam time request

In total, we request 12 shifts without protons, using a previously irradiated UC_x target. The shifts will be distributed as follows:

- 2 shifts to obtain a CRIS resonance with RaF and to optimize the beam tuning and laser setup.
- 3 shifts to perform systematic studies of RaF with varying target and CEC temperature.
- 4 shifts to study the production of RaF^- after the CEC. This includes systematic studies of the CEC temperature.
- 3 shifts to study the photodetachment of RaF^- into RaF.

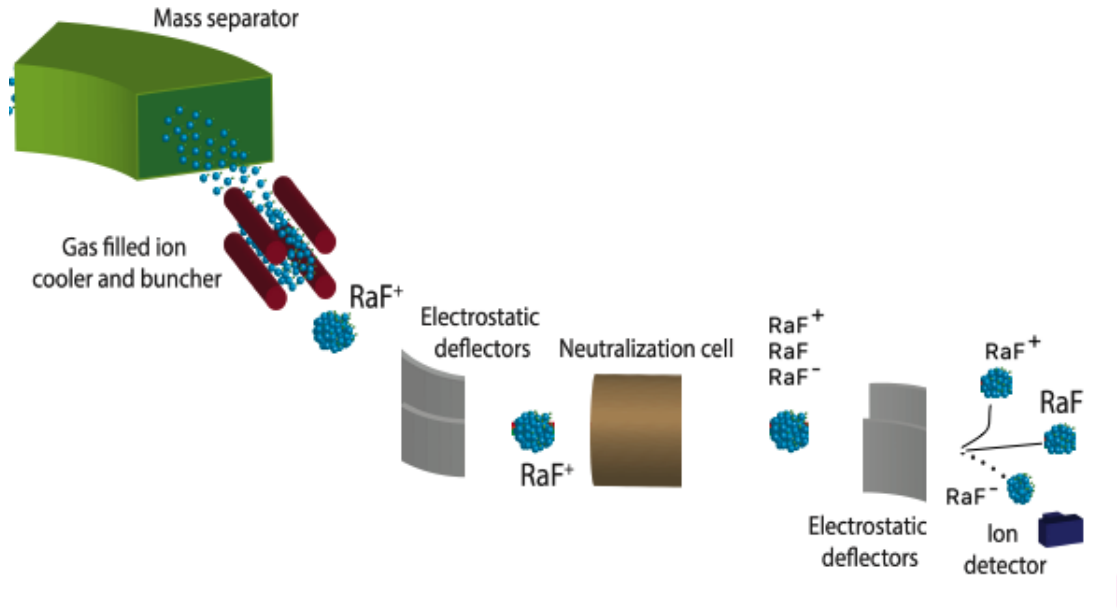

Figure 1: Scheme proposed for the production and detection of RaF^- molecules at CRIS. A beam of RaF^+ molecules can be converted into RaF^- by a two-step double charge exchange process. Figure modified from Ref. [4].

Molecule	Half life	Yield (ions/second)	Shifts
$^{226}RaF^+$	1600 yr	10^7	12

Table 1: Molecule of interest, half-life, yield, and requested shifts.

References

[1] G. Arrowsmith-Kron et al. Opportunities for fundamental physics research with radioactive molecules. *arXiv preprint arXiv:2302.02165*, 2023.

[2] R. F. Garcia Ruiz, R. Berger, J. Billowes, C. L. Binnersley, M. L. Bissell, A. A. Breier, A. J. Brinson, K. Chrysalidis, T. E. Cocolios, B. S. Cooper, K. T. Flanagan, T. F. Giesen, R. P. de Groote, S. Franchoo, F. P. Gustafsson, T. A. Isaev, Á. Koszorús, G. Neyens, H. A. Perrett, C. M. Ricketts, S. Rothe, L. Schweikhard, A. R. Vernon, K. D. A. Wendt, F. Wienholtz, S. G. Wilkins, and X. F. Yang. Spectroscopy of short-lived radioactive molecules. *Nature*, 581(7809):396–400, May 2020.

[3] S. M. Udrescu, A. J. Brinson, R. F. Garcia Ruiz, K. Gaul, R. Berger, J. Billowes, C. L. Binnersley, M. L. Bissell, A. A. Breier, K. Chrysalidis, T. E. Cocolios, B. S. Cooper, K. T. Flanagan, T. F. Giesen, R. P. de Groote, S. Franchoo, F. P. Gustafsson, T. A. Isaev, Á. Koszorús, G. Neyens, H. A. Perrett, C. M. Ricketts, S. Rothe, A. R.

Vernon, K. D. A. Wendt, F. Wienholtz, S. G. Wilkins, and X. F. Yang. Isotope shifts of radium monofluoride molecules. *Phys. Rev. Lett.*, 127:033001, Jul 2021.

[4] S. Wilkins, S. Udrescu, et al. Observation of the distribution of nuclear magnetization in a molecule. *arXiv preprint arXiv:2311.04121*, 2024.

[5] M. Athanasakis-Kaklamanakis, S. G. Wilkins, L. Skripnikov, A. Koszorús, A. A. Breier, M. Au, R. Berger, I. Belošević, M. L. Bissell, and A. Borschevsky *et al.* Pinning down electron correlations in RaF via spectroscopy of excited states. *arXiv*, 2308.14862, 2023.

[6] T A Isaev, S. Hoekstra, and R Berger. Lasercooled RaF as a promising candidate to measure molecular parity violation. *Phys. Rev. A*, 82:052521, 07 2010.

[7] S. M. Udrescu, S. G. Wilkins, A. A. Breier, M. Athanasakis-Kaklamanakis, R. F. Garcia Ruiz, M. Au, I. Belošević, R. Berger, M. L. Bissell, and C. L. Binnersley *et al.* Precision spectroscopy and laser-cooling scheme of a radium-containing molecule. *Nat. Phys.*, 20:202–207, 1 2024.

[8] M. Athanasakis-Kaklamanakis, S. G. Wilkins, P. Lassegues, L. Lalanne, and J. R. Reilly et al. Radiative lifetime of the $A^2\Pi 1/2$ state in RaF with relevance to laser cooling. *under review, Phys. Rev. A*, arXiv:2403.09336v1, 2024.

[9] K. Gaul, R. F. Garcia Ruiz, and R. Berger. Stopping mass-selected alkaline-earth metal monofluoride beams of high energy via formation of unusually stable anions. *arXiv preprint arXiv:2403.09320*, 2024.

[10] M. Nichols, M. Athanasakis-Kaklamanakis, A. Borschevsky, T.E. Cocolios, R. Crosa-Rossa, R.P. de Groote, K.T. Flanagan, R.F. Garcia Ruiz, S. Geldhof, D. Hanstorp, Á. Koszorús, L. Lalanne, D. Leimbach, G. Neyens, J. Reilly, S. Rothe, S.G. Wilkins, and X.F. Yang. Investigating radioactive negative ion production via double electron capture. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 541:264–267, 2023.

[11] T.E. Cocolios, R.P. de Groote, J. Billowes, M.L. Bissell, I. Budinčević, T. Day Goodacre, G.J. Farooq-Smith, V.N. Fedosseev, K.T. Flanagan, S. Franchoo, R.F. Garcia Ruiz, W. Gins, H. Heylen, T. Kron, R. Li, K.M. Lynch, B.A. Marsh, G. Neyens, R.E. Rossel, S. Rothe, A.J. Smith, H.H. Stroke, K.D.A. Wendt, S.G. Wilkins, and X. Yang. High-resolution laser spectroscopy with the collinear resonance ionisation spectroscopy (cris) experiment at cern-isolde. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 376:284–287, 2016. Proceedings of the XVIIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11-15 May 2015.

[12] A.R. Vernon, J. Billowes, C.L. Binnersley, M.L. Bissell, T.E. Cocolios, G.J. Farooq-Smith, K.T. Flanagan, R.F. Garcia Ruiz, W. Gins, R.P. de Groote, Á. Koszorús, K.M. Lynch, G. Neyens, C.M. Ricketts, K.D.A. Wendt, S.G. Wilkins, and X.F.

Yang. Simulation of the relative atomic populations of elements 1z89 following charge exchange tested with collinear resonance ionization spectroscopy of indium. *Spectrochimica Acta Part B: Atomic Spectroscopy*, 153:61–83, 2019.

[13] Nicholas R. Hutzler, Hsin-I Lu, and John M. Doyle. The Buffer Gas Beam: An Intense, Cold, and Slow Source for Atoms and Molecules. *arXiv e-prints*, page arXiv:1111.2841, November 2011.

[14] Jan Heinemeier and Preben Hvelplund. Production of 10–80 kev negative heavy ions by charge exchange in na vapour. *Nuclear Instruments and Methods*, 148(3):425–429, 1978.

[15] G.D. Alton. *Negative-Ion Formation Processes and Sources*, pages 222–273. Springer Berlin Heidelberg, Berlin, Heidelberg, 2005.

[16] M. Bacal, M. Sasao, and M. Wada. Negative ion sources. *Journal of Applied Physics*, 129(22):221101, 06 2021.

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	☒ Existing	☒ 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.