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Molecular laser spectroscopy of RaF at CRIS

In the past few years, the spectroscopy of radioactive molecules has been performed at ISOLDE (CERN) using the Collinear Resonance Ionization Spectroscopy (CRIS) experiment [1]. Given their structure and chemical properties, molecules are a promising candidate for studies in different fields [2] and for extracting elements with low production yields due to their refractory or reactive properties.

Among them, diatomic polar molecules are at the center of theoretical and experimental investigations in search of the electron's electric dipole moment (eEDM) and nuclear Schiff moments [3-4]. Due to the strong electric field and the rich electronic, vibrational, and rotational structure inherent in molecules, the sensitivity to Schiff moments and eEDM is expected to be enhanced in radioactive polar molecules, such as RaF [5]. The sensitivity to these moments depends on non-measurable molecular constants, making the benchmark of ab initio molecular theory with experimental observations a crucial step towards eEDM studies.

After three experimental campaigns at CRIS (2018, 2021, 2023), all predicted electronic levels in RaF have been studied with broadband laser spectroscopy [6] and one optical transition in high resolution on 223,225,226RaF, yielding sensitivity to magnetic dipole and electric quadrupole interactions, as well as a laser cooling scheme [7-8].

This poster will present the study of eEDM's on molecules, the basic principles of molecular spectroscopy at CRIS, and some results obtained in the last experimental campaign.

[1] Garcia Ruiz, R.F., et al. "Spectroscopy of short-lived radioactive molecules." *Nature* 581.7809 (2020): 396-400.

[2] Opportunities for Fundamental Physics Research with Radioactive Molecules, arXiv:2302.02165 (2023)

[3] Kudashov, A. D., et al. "Ab initio study of radium monofluoride (RaF) as a candidate to search for parity- and time- and parity-violation effects." *Physical Review A* 90.5 (2014): 052513.

[4] Osika, Y., & Shundalau, M. (2022). Fock-space relativistic coupled cluster study on the RaF molecule promising for the laser cooling. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 264, 120274.

[5] Flambaum, V. V., and V. A. Dzuba. "Electric dipole moments of atoms and molecules produced by enhanced nuclear Schiff moments." *Physical Review A* 101.4 (2020): 042504.

[6] M. Athanasakis-Kaklamanakise et al., Pinning down electron correlations in RaF via spectroscopy of excited states, submitted (2023)

[7] S.-M. Udrescu, et al., Precision spectroscopy and laser-cooling scheme of a radium-containing molecule", *Nature Physics* (2024) online January 9, <https://doi.org/10.1038/s41567-023-02296-w>

[8] S. G. Wilkins et al., Observation of the distribution of nuclear magnetization in a molecule, submitted (2023).

Abstracts

Primary author: FAJARDO ZAMBRANO, Carlos Mario (KU Leuven)

Co-authors: MCGLONE, Abi (University of Manchester); KOSZORUS, Agota (University of Liverpool); Dr BREIER, Alexander A.; Prof. BORSHEVSKY, Anastasia (University of Groningen); VAN DEN BORNE, Bram (KU Leuven); HANSTORP, Dag (University of Gothenburg); NEYENS, Gerda (KU Leuven); Dr BELOSEVIC, Ivana (TRIUMF); WARBINEK, Jessica (CERN); REILLY, Jordan (University of Manchester); WESSOLEK, Julius (CERN); Mr BERBALK, Justus (KU Leuven); Prof. LYNCH, Kara (University of Manchester); CHRYSALIDIS,

Katerina (CERN); FLANAGAN, Kieran (University of Manchester); Dr SKRIPNIKOV, Leonid (Kurchatov Institute); LALANNE, Louis (IPN); NIES, Lukas (CERN / University of Greifswald (DE)); BISSELL, Mark (CERN); AU, Mia (CERN); ATHANASAKIS-KAKLAMANAKIS, Michail (CERN); Mrs NICHOLS, Miranda (University of Gothenburg); Mr AHMAD, Osama (KU Leuven); IMGRAM, Phillip (Institute for Nuclear Physics, Technical University Darmstadt); LASSÈGUES, Pierre (KU Leuven); HEINKE, Reinhard (KU Leuven); Dr BERGER, Robert (Philipp-Universität Marburg); GARCIA RUIZ, Ronald Fernando (MIT); DE GROOTE, Ruben; GELDHOF, Sarina (GANIL); FRANCHOO, Serge (ipn); WILKINS, Shane (CERN); BAI, Shiwei (Peking University); UDRESCU, Silviu-Marian (MIT); Dr KUJANPÄÄ, Sonja (University of Jyväskylä); COCOLIOS, Thomas Elias (KU Leuven - Instituut voor Kern- en Stralingsfysica); Prof. GIESEN, Thomas F. (University of Cologne); YANG, Xiaofei (Peking University); LIU, Yinshen (Peking University); RAGGIO, andrea (INFN-Sezione di Padova)