Title of thesis: Chirped pulse microwave studies of molecular collisions of atmospheric and astrophysical interest using a pulsed supersonic flow reactor

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A detailed knowledge of gas-phase molecular collisions, both reactive and inelastic, is essential to our understanding of natural and man-made environments, ranging from the extreme cold of dense interstellar clouds to the high temperatures of flames and the intermediate temperatures prevailing in atmospheres. Under the ERC Advanced Grant CRESUCHIRP (2016-22) we developed an optimised version of the Chirped Pulse in Uniform Flow (CPUF) technique [1], and used it for example to study collisions of HCN and its unstable isomer HNC with He, leading to a recent publication in Nature Chemistry [2]. This study paves the way for the research proposed here.

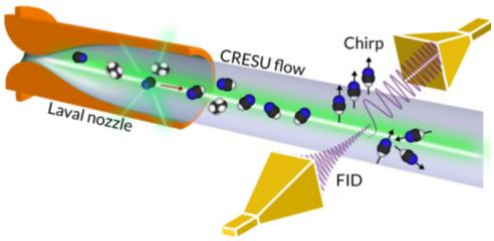

Fig. 1 The CPUF technique implemented in Rennes

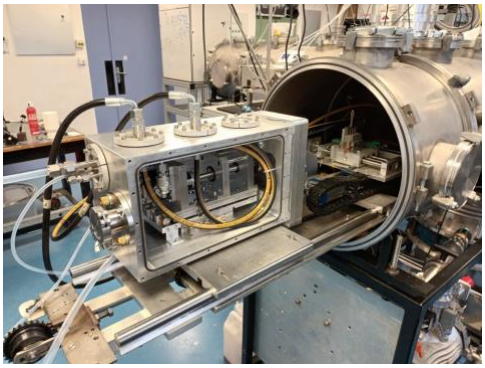
Fig. 2 Photo of pulsed CRESU with hydraulic drive

We would like to use the CPUF technique to study the reactivity and collisional excitation (energy transfer) of bigger molecules and heavier colliders. However, there exists a major blockage – these species require much lower pressure flows than currently possible. At the end of the CRESUCHIRP project we were able to demonstrate a pulsed CRESU based on a novel hydraulically actuated spinning disk valve, achieving much lower pressures with drastically reduced gas consumption (O. Abdelkader Khedaoui, PhD Université de Rennes 1, Rennes, France, 2022, see Fig. 2). The main aim of this PhD project is to develop this prototype into a robust instrument, and use it to study collisional excitation and reactivity of strong relevance not only for interstellar space but also the atmospheres of comets and planets including the Earth's, as well as flames, where such fundamental information will be key in improving our understanding of – and therefore our ability to influence – important processes involved in climate change and air pollution.

The Rennes team are well known for their studies of gas phase kinetics and spectroscopy under extreme conditions of temperature (from below 10 K up to several thousand K) of relevance to astrophysical and atmospheric environments (ERC Advanced Grant, Descartes prize, multiple articles in Science, Nature Chemistry, Phys. Rev. Letters etc). The team is particularly well known for the development of the CRESU (Cinétique de Réaction en Ecoulement Supersonique Uniforme or Reaction Kinetics in Uniform Supersonic Flow) technique to study reactive and inelastic collisions in the gas phase down to 5 K for even highly condensable species. It has been able to demonstrate that many reactions of neutral chemical species called radicals (atoms or molecules possessing one or more unpaired electrons) with other neutral molecules actually become faster as the temperature is lowered, contrary to expectation (for a review, see ref. 3) These findings have had a major impact on our understanding of astrochemistry and the chemistry of cold planetary atmospheres.

The project will be undertaken within the Molecular Physics Department of the Institute of Physics Rennes, a joint CNRS – University of Rennes Research Unit situated on the pleasant Beaulieu Scientific Campus. The Department is internationally known for its experimental and theoretical studies of elementary processes of interest for astrophysics, atmospheric science and combustion, and provides an excellent environment for PhD training. The PhD student would enjoy excellent interactions with the multinational CRESUCHIRP project team and would also benefit from an exceptional technical environment constructed within the framework of the CRESUCHIRP project, both in terms of dedicated and high-quality laboratory space as well as custom built high performance scientific instruments (see Fig. 3).

Fig. 3 CRESUCHIRP laboratory at the IPR

The position is available starting in October 2023 for a period of 3 years. Candidates should possess a Masters degree (M2) in physics or (physical) chemistry, or possibly mechanical engineering with a specialisation in instrumentation. Experience in experimental research and especially the use of lasers, spectroscopy, vacuum and gas flow techniques and high-speed electronics would be advantageous. Inquiries and applications, including a detailed CV citing grades, an accompanying letter, and the names and contact details of two or three referees, should be addressed to Prof. Ian Sims (ian.sims@univ-rennes.fr). Review of applications will commence immediately, and will continue until a suitable applicant is identified. Further details of the overall project can be seen on the CRESUCHIRP website: https://cresuchirp.wordpress.com

References

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Keywords: Chirped Pulse Fourier Transform Microwave Spectroscopy (CPFTMW), CRESU, Low Temperature Reaction Kinetics, Product Branching Ratios, Elementary Reactions, Collisional Energy Transfer, Rotational Excitation, Laboratory Astrophysics, Experimental Astrochemistry, Chemical Physics, COMs, Rotational Spectroscopy, Gas-Phase Physical Chemistry