

LIGHTinPARIS		Marie Skłodowska-Curie Actions Second call for COFUND PhD fellowships
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Domain	ONERA (Waves and Matter)	
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* The requirements for obtaining access authorisation to the DPHY laboratory are very strict as ONERA (the employer) is a research centre under the supervision of the French Ministry of the Armed Forces.		
PHD THESIS PROJECT		
UltrafastCARS - High-speed coherent Raman metrology for the study of transient and non-equilibrium reactive flows		
Summary / Abstract		
Femtosecond Coherent Anti-Stokes Raman Scattering (CARS) spectroscopy allows for probing the population distribution of ro-vibrational levels in a molecular medium and thus provides a precise and non-invasive way to measure temperature within reactive media such as combustions and plasmas. To provide a new understanding of the turbulent or transient phenomena occurring in reactive flows, like rotating detonation combustions, which represent a major potential advance in aeronautical and space propulsion, or of the non-equilibrium plasmas and their related applications in energy and environment domains. ONERA is developing a very high-speed instrument based on femtosecond ultrashort laser technology. This thesis will focus in particular on optimizing light-matter interaction, developing ultrafast data processing algorithms, modeling non-equilibrium media, and experimentally validating the instrument's performance in representative flow conditions. Principal 3i dimension: intersectoral. Additional 3i dimension: interdisciplinary		
Keywords		
• Atomic, molecular and plasma physics • Instrumentation, optical diagnostics • Femtosecond lasers • Raman Spectroscopy • Ultrafast data-processing; Reacting flows, Combustions		

UltrafastCARS - High-speed coherent Raman metrology for the study of transient and non-equilibrium reactive flows

Key Objectives

This PhD project focuses on the development of a next-generation, high-repetition-rate CARS (Coherent Anti-Stokes Raman Scattering) instrument dedicated to the analysis of unsteady and non-equilibrium reactive flows. The research is structured around two main objectives:

- 1) Implementation of a new high-speed measurement system and the development of a new generation of data-processing algorithms specifically designed for high-cadence operation.
- 2) Extension of femtosecond CARS to non-equilibrium environments, where ro-vibrational population distributions deviate from Boltzmann statistics and follow more complex, non-thermal behaviors.

Ambitious applications concern the study of Rotating Detonation Engines (RDEs)—an emerging propulsion concept that combines ultrafast transient phenomena and with highly unsteady reactive flows and non-equilibrium plasma assisted combustion & nitrogen fixation, which are relevant to the carbon-free industrial transition with a potential global impact

Project

Context and state-of-the-art of Scientific Research project

To support innovation in aerospace propulsion, or plasma assisted energy and environment applications accurate flow field measurements are essential. Experimental diagnostics are needed for probing complex reactive media such as unsteady combustion and non-equilibrium plasmas and validate numerical models. Among other techniques, laser diagnostics offer reliable, non-intrusive, and quantitative measurements with high spatial and temporal resolution. In particular, Coherent Anti-Stokes Raman Scattering (CARS) spectroscopy stands out as a standard for thermometry.

Recent progress in hybrid fs/ps-CARS have pushed the limits of the technique. 1 kHz measurements were demonstrated using Ti:Sa laser sources, and ONERA's extended this limit to 5 kHz with using an innovative Yb:YAG laser architecture [1,2]. Systems reaching apparent higher rates (100 kHz–1 MHz) [3] were also developed but on intermittent burst-mode operation (short sequences of pulses) that restricts the temporal bandwidth, prevents continuous monitoring, and limits statistical reliability. The objective of this project is to bridge the gap between 5 and 100 kHz by developing the first continuous-operation fs/ps-CARS instrument operating at 50 kHz. This will enable high-fidelity diagnostics, allowing the acquisition of extensive datasets over very short timescales and capturing wide temporal dynamics relevant to non-stationary flows.

A second focus is on advanced data analysis to handle the large spectral datasets produced. Artificial intelligence (AI) methods have shown promise in chemistry and biomedical spectroscopy [4] but remain underexplored for CARS spectroscopy. The project will investigate the integration of AI models, such as convolutional neural networks, for post-processing and their ability to provide relevant results and to ultimately reduce measurement uncertainties. In parallel, recently proposed ultrafast algorithms [5] for computing Raman spectra containing millions of transitions represent a promising approach to enable ultra-fast and determinist spectral analysis.

Additionally, fs CARS studies of non-equilibrium media have been previously demonstrated [6, 7, 8], although involving complex setups or limited temperature ranges. ONERA's recent collaboration with EM2C lab [9] introduced a simplified single-laser, single-detector configuration compatible with single-shot

acquisition at high-temperature by probing Q-branch ro-vibrational transitions. This approach, while validated for Boltzmann distributions, needs to be extended to more general non-equilibrium regimes and tested in plasmas where the off-equilibrium is controlled spatially and temporally [10, 11].

Another application target is the study of propulsion concepts characterized by extreme thermodynamic and flow conditions. A 50 kHz CARS instrument would be a major leap forward, allowing precise tracking of detonation cycles and statistically robust characterization of transient combustion dynamics

Description

This PhD project focuses on the development of a next-generation, high-repetition-rate CARS (Coherent Anti-Stokes Raman Scattering) instrument dedicated to the analysis of unsteady and non-equilibrium reactive flows. The research is structured around two main objectives:

3) Implementation of a new high-speed measurement system and the development of a new generation of data-processing algorithms specifically designed for high-cadence operation.

4) Extension of femtosecond CARS to non-equilibrium environments, where ro-vibrational population distributions deviate from Boltzmann statistics and follow more complex, non-thermal behaviors.

A particularly ambitious application concerns the study of Rotating Detonation Engines (RDEs), an emerging propulsion concept that combines ultrafast transient phenomena with highly unsteady reactive flows. In such environments, shock-induced heating mechanisms generate strongly non-equilibrium states that directly influence the underlying chemical kinetics.

The project's goal of achieving a tenfold increase in measurement rate aligns with the rapid technological progress of modern ultrafast laser systems, whose new architectures now enable repetition rates previously unattainable. Achieving continuous operation at these frequencies, however, raises major experimental challenges, particularly in signal detection and computational challenges related to the processing of large volume of CARS spectra. Overcoming these challenges will require the design of innovative detection strategies and advanced spectral analysis algorithms. An innovative processing technique involving neural networks was tested during an internship in the summer of 2022 and continued during a second internship in 2023. The concept yielded very encouraging initial results: a reduction in processing time of several orders of magnitude and preserved measurement accuracy on datasets acquired under similar conditions. This approach will therefore be pursued to make it more robust and improved by drawing on new literature and internal collaborations. Recently proposed ultrafast algorithms for computing Raman spectra represent a promising approach that will also be considered.

The instrument development will focus initially on N_2 , O_2 and H_2 , building on ONERA's established expertise and prior validated CARS systems developed for aerospace and energy applications. The extension of the technique to CO, a key combustion product in RDEs, represents an important advance: its Raman spectral structure, characterized by a dense and regular Q-branch similar to N_2 , makes it an excellent candidate for probing thermodynamic non-equilibrium states. This capability was successfully demonstrated for N_2 in glow discharges exhibiting highly non-equilibrium behavior between rotation and vibration during a previous collaboration with EM2C lab. The aim is to propose a more general algorithm capable of accounting for non-Boltzmann distributions produced during ultra-brief processes without any prior assumptions about their profile.

The performances of the instrument will be first characterized in well-controlled environments such as high-pressure cells, laminar burners, and stabilized plasma before considering an application on a realistic engine. In particular, the expertise of EM2C lab will allow to test the diagnostics in plasmas where the off-

equilibrium is controlled spatially and temporally. For example, continuous or pulsed microwave plasma jets at atmospheric or intermediate pressures can be used. The development of high-temporal-resolution hybrid CARS is important not only for sampling temperature fields in these environments but also for studying vibrational non-equilibrium in electronic ground states of N_2 , O_2 and H_2 molecules. Understanding vibration-dependent kinetics is essential for applications such as plasma-assisted nitrogen fixation or combustion.

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Facilities and quality of research environment available for the researcher

State-of-the-art femtosecond laser sources have been supplied as part of an ambitious equipment project. A wide range of reference reactive flows is available at ONERA and EM2C such as high-pressure cells, laminar burners, and continuous or pulsed plasma sources in order to validate spectroscopic models, optimize the light-matter interaction, and characterize the instrument performances. The project will be carried out in close collaboration with ONERA's Department of Energetics (DMPE) and the EM2C Laboratory (École CentraleSupélec), ensuring strong integration with ongoing experimental and modeling efforts in reactive flow metrology. This synergy will provide a rich and realistic application framework, enabling the developed diagnostic to address some of the most challenging measurement needs in aerospace propulsion, high-temperature reactive flows and plasma physics.

3i Dimensions - intersectoral; interdisciplinary

Description and expected outcomes

The project is interdisciplinary and intersectoral as it will be carried out in close collaboration between ONERA (industrial partner), being the principal host lab and Myriam Raybaut supervisor from EDOM doctoral school; and EM2C Laboratory (academic partner from École CentraleSupélec) with Gabi Stancu providing co-supervision affiliated with SMEMAG doctoral school, whose expertise are complementary to ensure success of the PhD goals. The project is fundamentally interdisciplinary being at the interface of many advanced field and requiring complementary expertise's in laser spectroscopy (ONERA/DPHY), plasma physics and advanced laser diagnostics in non-equilibrium media (EM2C lab), advanced algorithmic techniques (ONERA/DTIS) and large-scale applicative test benches (ONERA/DMPE). This synergy will provide a rich and realistic application framework, enabling the developed diagnostic to address some of the most challenging measurement needs in aerospace propulsion, high-temperature flows and plasma physics.

Approximate timeline of internships / external stays corresponding to the 3i dimension

Although the primary work environment will be ONERA's industrial facilities, the PhD student will be able to easily integrate into the academic community of CentraleSupélec, Université Paris-Saclay, and attend specific courses. The geographical proximity will allow the PhD student to easily travel to the EM2C laboratory and prepare the non-equilibrium plasma experiments required for the project. The PhD student will be in contact with other EM2C PhD students working on complementary femtosecond laser diagnostics.

Profile of PhD fellow

PhD studies and research work require curiosity, creativity, rigour, teamwork, and organisational skills. A minimum B2 level of English is mandatory. The below criteria are specific to the thesis project.

pre-requisite	Theoretical and experimental skills in laser techniques and ultrashort pulses
pre-requisite	Knowledge of light-matter interaction and non-linear optics
advantageous	Academic background in spectroscopy
advantageous	Academic background in plasma physics
advantageous	Programming experience (e.g. python, machine learning background)

Ethics issues in the project as per Horizon Europe Ethics Self-assessment

Section 1. Human embryonic stem cells (hESCs) and human embryos (hEs)			No
Section 2. Humans	No	Section 3. Human cells and tissues	No
Section 4. Personal data	No	Section 5. Animals	No
Section 6. Third Countries	No	Section 7. Environment, health and safety	Yes
Section 8. Artificial Intelligence	Yes	Section 9. Other ethics issues	No
Section 10. Crosscutting issue: potential misuse of results			No

7. Environment, health and safety

All French labs are required to have health and safety procedures in place, which takes into account the use of high-energy light sources such as lasers of class 3 and above, and the use of hazardous / toxic chemical and biological products. The institution and the laboratory comply with the highest safety requirements and PhD fellows have mandatory health and safety training, with additional training if required.

8. Artificial intelligence

The use of machine learning and artificial intelligence is used to fit data and analyse results. This is typically used to improve fits and complex models with multiple parameters. The data in question is related to physical experiments and is not human data.

Artificial Intelligence is not developed during this project and is thus not impacted by the AI Act.