One-day CLIO workshop for Infrared Free Electron Laser Users

Friday May 28th 2021

https://clio2021.sciencesconf.org

Welcome from the organising committee

We would like to welcome you to the first CLIO workshop for Infrared Free Electron Laser Users organized by the Institute of Physical Chemistry in Orsay.

Due to pandemic situation worldwide, it will take place on **May 28th** "on line" and it will focus on the state-of-the-art methodologies envisioned by expert researchers working or likely to work with Free Electron Lasers in the mid or far-IR range.
We think that CLIO2021 will be a useful occasion to enjoy science on IR FELs, to get new ideas and to establish new collaborations between IR FEL users especially during the round table, which we hope will contribute to animate the discussions.

We would like to thank Carine Clavaguera and Estelle Loire from the Institute of Physical Chemistry for their precious help in the organisation of this "on line" workshop.

Organising committee

Charles Desfrançois

Laboratoire de Physique des Lasers, CNRS - Université Paris 13

Jean-Yves Salpin

LAMBE, CNRS, Université d'Evry-Université Paris Saclay

Debora Scuderi

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Partners of the CLIO2021 workshop

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CLIO Centre Laser Infrarouge d'Orsay

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CPPS Fédération de Chimie Physique de Paris-Saclay

Société Chimique de France

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Société Française de Physique

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SFSM Société Francaise de Spectrométrie de Masse

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- Environment
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This project has received funding from the European Union's Horizon 2020 research and innovation Programme under grant agreement NO 731077

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Program

09:00 - 09:15	**Introduction to CLIO workshop** - *Guillaume Van der Rest*, Institut de Chimie Physique, Université Paris Saclay-CNRS, Orsay, France
09:15 - 10:45	**Session I** - *Jean-Yves Salpin*
09:15 - 09:45	CLIO, laser features, layout of the facility and applications digest - *Rui Prazeres*, Institut de Chimie Physique, Université Paris Saclay-CNRS, Orsay, France
09:45 - 10:15	Nonlinear optical spectroscopy of interfaces and thin films using CLIO-FEL - *Abderrahmane Tadjeddine*, Institut de Chimie Physique, Université Paris Saclay-CNRS, Orsay, France
10:15 - 10:45	Transition metal clusters as model systems for heterogeneous catalysis - *Joost Bakker*, FELIX Laboratory, Radboud University, Nijmegen, The Netherlands
10:45 - 11:00	Coffee break
11:00 - 12:00	**Session II** - *Debora Scuderi*
11:00 - 11:30	A Cryogenic Ion Trap Beamline at FELIX for Astrochemical Studies - *Sandra Brünken*, FELIX Laboratory, Radboud University, Nijmegen, The Netherlands
11:30 - 12:00	Conformations and interactions in neutral biomolecules in the gas phase - *Pierre Carçabal*, Institut de Sciences Moléculaires d'Orsay (ISMO), Université Paris Saclay-CNRS, Orsay, France
12:00 - 13:15	Lunch break -CLIO virtual visit
13:15 - 14:45	**Session III** - *Charles Desfrançois*
13:15 - 13:45	Infrared Super-Resolution Microscopy by Sum-Frequency Generation - *Alexander Paarmann*, Department of Physical Chemistry, Fritz Haber Institute, Germany
13:45 - 14:15	Spectroscopic Applications of Synchrotron Radiation (non-coherent and coherent) in the IR/mm Range - *Pascale Roy*, Soleil Synchrotron, Gif sur Yvette, France
14:15 - 14:45	Infrared ion spectroscopy with Optical Parametric Oscillators (OPOs) - *Giel Berden*, FELIX Laboratory, Radboud University, Nijmegen, The Netherlands
15:00 - 16:30	**Round table and discussions**- *L.Nahon, O. Pirali, R. Prazeres, D. Scuderi*
16:30 - 16:40	**Conclusions** - *Debora Scuderi*, Institut de Chimie Physique, Université Paris Saclay-CNRS, Orsay, France

CLIO, laser features, layout of the facility and applications digest

09:15 - 09:45

R. Prazeres

Institut de Chimie Physique, CNRS - Université Paris-Saclay, Orsay, France

Abstract

We present a layout of the CLIO Free Electron Laser facility, from the linear accelerator up to the user's room, including the laser beam transport. We show the characteristics of the laser beam: power, temporal structure, spectral range, … We explain what are the peculiar features of the FEL, especially those of CLIO, making a comparison with other FELs. And we make a brief description of some important application experiments which have been developed here, in molecular physics, semi-conductors, infrared multi-photon dissociation (IRMPD) and SFG spectroscopy, including the development of near field infrared spectroscopy AFMIR.

References

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Prazeres, R.; Glotin, F.; Ortega, J.M., *Phys. Rev. Acc. and Beams*, **2018**, *21 (11)*, article number: 110703

Evidence for competition modes in a partially guided far-infrared free-electron laser
Ortega, J.M.; Berthet, J.P.; Glotin, F.; Prazeres, R, *Physical Review Special Topics-Accelerators and Beams*, **2014**, *17 (10)*, article number: 100701

Analysis of periodic spectral gaps observed in the tuning range of free-electron lasers with a partial waveguide
Prazeres, R. ; Glotin, F. ; Ortega, J.M., *Physical Review Special Topics-Accelerators and Beams*, **2009**, *12*, article number: 010701

Extension in far-infrared of the CLIO free-electron laser
Ortega, JM ; Glotin, F ; Prazeres, R, *International Workshop On Infrared Microscopy And Spectroscopy With Accelerator-Based Sources* (WIRMS 2005), Rathen, GERMANY
Infrared Physics & Technology, **2009**, *49*, 133-138

Nonlinear optical spectroscopy of interfaces and thin films using CLIO-FEL

09:45 - 10:15

A. Tadjeddine

Institut de Chimie Physique, CNRS - Université Paris-Saclay, Orsay, France

Abstract

The first user experience of CLIO was the in situ study of the adsorption of CO and CN- at the electrochemical interface by Generation of the Visible-Infrared Sum Frequency. I will briefly discuss some highlights before discussing a few areas where the performance of CLIO-FEL is an asset that tabletop lasers cannot equal.

Transition metal clusters as model systems for heterogeneous catalysis

10:15 - 10:45

J.M. Bakker

FELIX Laboratory, Radboud University, Nijmegen, The Netherlands

Abstract

Atomic clusters are small particles consisting of a countable number of atoms. Due to the large number of undercoordinated atoms, metal clusters are often considered as model systems for the active site in heterogeneous catalysis. The study of gas-phase clusters with mass-spectrometric and spectroscopic techniques offers the possibility to elucidate fundamental interactions of molecules with a surface that is known with atomic precision, offering well-defined local geometric and electronic structure.
In this talk, I will illustrate how such insight is derived from the IR spectroscopic study of rhodium clusters interacting with NO, model system for the catalytic reduction of NO to N_2 and O_2 in three-way catalytic converters. I will show how controlled doping of the materials used reveals that the oxophilicity of the cluster is the key characteristic that drives NO dissociation.

References

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J. Phys. Chem. C, **2019**, *123*, 3476

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J. Phys. Chem. Lett., **2020**, *11*, 4408

A cryogenic ion trap beamline at FELIX for astrochemical studies

11:00 - 11:30

S. Brünken[1], S. Schlemmer[2], B. Redlich[1]

[1]FELIX Laboratory, Radboud University, Nijmegen, The Netherlands
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Abstract

Reactive molecular ions play a central role in the chemistry of planetary atmospheres and the interstellar medium. Laboratory spectroscopic studies of these often elusive, but essential, reaction intermediates yield fundamental insights on their geometric and electronic structure, and provide spectroscopic signatures needed for their identification in space [1]. Cryogenic ion traps have proven to be ideal tools for the development of sensitive spectroscopic schemes of mass-selected, cold, and isolated molecular ions.
Here, I will describe the combination of such a cryogenic (>4 K) 22-pole ion trap instrument with the widely tunable free electron lasers at the FELIX Laboratory [2]. It allows for wide-range infrared vibrational action-spectroscopy of molecular ions. I will present laboratory studies of the gas-phase spectra of several hydrocarbon and nitrogen-substituted hydrocarbon cations ranging in size from comparatively small systems to polycyclic aromatic hydrocarbon cations [3-5]. A focus will be on infrared experiments and methods to disentangle the isomeric composition of ionic samples. I will outline how these studies pave the way to investigate isomer-selective reaction kinetics of ion-neutral reactions at astronomically relevant temperatures.

References

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Conformations and interactions in neutral biomolecules in the gas phase

11:30 - 12:00

G. Goldsztejn[1], A. Camiruaga[2], P. Çarçabal[1]

[1] Institut des Sciences Moléculaires d'Orsay, Université Paris Saclay, Orsay, France
[2] Departamento de Química Física, Facultad de Ciencia y Tecnología, Leioa, Spain

Abstract

Biomolecules are complex flexible systems that can adopt multiple conformations that can be influenced by the interactions with surrounding molecular partner such as solvent molecules or other biomolecules. The bioactive forms of such assemblies are governed by the combination of intrinsic properties of each molecular components and their intermolecular arrangement. Monitoring the intermolecular interactions at play and their balance with intramolecular preferences provides key information on affinity and selectivity, the key ingredients for efficient biological activity.

Gas phase conformer selective and vibrationally resolved spectroscopic methods, complemented by computational conformational landscapes exploration, uniquely probe local molecular interactions. In the near infrared, between 3.5 and 4 µm, we can probe intramolecular stretching modes of O-H and N-H groups. These key groups are directly involved in the intra- and intermolecular interactions as donors and acceptors. The spectral properties (position, width, and intensities) observed is this spectral region are related to the nature and strength of the non-covalent interactions. The mid- to far-IR region, above 5 µm, allows to probe vibrations associated to more collective motions inside the molecular assemblies and can shed the light on the global organization of the systems, including non-polar groups, as well as complement the near-IR spectral information to discriminate between similar conformations. In this IR range, it is also possible to probe intermolecular modes directly, which would provide experimental benchmarks to test models of intermolecular forces and large amplitude vibrational motions.

The presentation will focus on the experimental methods for the study of conformations and interaction in neutral sugar containing assemblies such as glycolipids, sugar-peptide complexes and hydrated complexes by means of gas phase IR spectroscopy.

Infrared super-resolution microscopy by Sum-Frequency Generation

13:15 - 13:45

R. Niemann[1], S. Wasserroth[1], G. Yu[2], C. R. Gubbin[3], M. Wolf[1], S. De Liberato[3], J. D. Caldwell[2], A. Paarmann[1]

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Abstract

Nonlinear optical imaging with a spatial resolution well below the Abbe diffraction limit of light, such as stimulated emission depletion (STED) microscopy, has enabled many ground-breaking discoveries. In the infrared spectral range, however, no comparable schemes exist, and high-resolution infrared imaging has so far been restricted to scanning probe techniques such as scattering near-field optical microscopy.
In my talk, I will demonstrate sum-frequency generation (SFG) microscopy as a new infrared super-resolution imaging technique, which can additionally provide interface-specific signals through the selection rules of the nonlinear-optical process. By combining a resonant infrared and non-resonant visible light beam at an interface, a light field at the sum of the two incoming frequencies is generated. Since this signal emerges in the visible, it can be imaged and detected using traditional optics with a resolution limit given by the wavelength of the SFG. The imaging contrast, however, arises from the infrared resonances at the sample.
To demonstrate the concept, we study an infrared nanophotonic system based on localized phonon polaritons in sub-diffractional nanostructures made from Silicon Carbide [1,2]. For our experiments, we use the infrared free-electron laser (FEL) at our institute [3] as a unique high-power infrared light source. First SFG microscopy results [4] by scanning tightly focused beams demonstrated the principle, but also revealed laser-induced damage which can only be avoided through a wide-field SFG microscopy approach. The first implementation of such a wide-field SFG microscope yielded not only much improved sensitivity but also a sub-diffractional spatial resolution of <1 µm at an infrared imaging wavelength of 11.6 µm. Finally, I will lay out the perspectives of how we will be developing this concept into a versatile instrument for interface-specific infrared super-resolution microscopy.

References

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Spectroscopic applications of synchrotron radiation (non-coherent and coherent) in the IR/mm range

13:45 - 14:15

J-B. Brubach[1], M. Verseils[1], L. Manceron[1], J.-F. Lampin[2], S. Eliet[2] O. Pirali[3], M.-A. Martin-Drumel[3], F. Hindle[4], G. Mouret[4], C. Evain[4], C. Szwaj[4], E. Roussel[4], S. Bielawski[4], T. Timusk[4], Pascale Roy[1.]

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Abstract

Some applications based on the use of intense and stable synchrotron radiation in the infrared/ THz will be presented. This will include: (i) the optical study of H_3S at extreme pressure, revealing its superconducting nature, (ii) the reflectivity of nanometer thin Nb at sub-K temperature, (iii) a new method for measuring Molecular absorption at ultra-high resolution and (iv) the use of pulse per pulse coherent THz emission for kinetic studies. All these examples require one or more specific characteristic of synchrotron radiation: namely its extension to the sub millimeter range, its brilliance and/or the time structure and coherence of the light pulses.

Some related measurements based on free-electron lasers (FELs) will be presented and perspectives will be drawn. The specifications required for complementary programs on the two sources will be sketched.

References

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Infrared ion spectroscopy with Optical Parametric Oscillators (OPOs)

14:15 - 14:45

G. Berden[1], R. van Outersterp[1], J. Martens[1], J. Oomens[1], L. Lamard[2], A. Peremans[3]

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Abstract

Infrared ion spectroscopy (IRIS) has become a valuable and well-established method for the structural and conformational characterization of (bio)molecular ions in mass spectrometry. Trapped ions are irradiated with tunable IR radiation from high power lasers such as user facility-based free electron lasers (FELs) and table top optical parametric oscillators/amplifiers. Monitoring the extent of IR-induced dissociation of the ion population, by recording the fragment and precursor ion intensity as a function of IR laser frequency, provides an IR spectrum of the precursor ion. The large wavelength tuning range, the macro/micro pulse structure, and the high macropulse pulse energy are ideal properties of FELs for IRIS. Table-top OPOs allow IRIS experiments to be performed outside the user facility, although in a limited wavelength range. But also, in a FEL facility, OPOs are very valuable for, for example, two-color experiments [1,2,3]. Several types of OPO are nowadays commercially available. They differ in pulse structure and pulse energy. Here we discuss the use of these OPOs for IRIS experiments at FEL facilities.

References

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Round table and discussions

15:00 - 16:30

Round table moderators: *L. Nahon (*Synchrotron SOLEIL), *O. Pirali (ISMO, CNRS-Université Paris Saclay), R. Prazeres and D. Scuderi (ICP, CNRS-Université Paris Saclay)*

- **Selective vibrational excitation of astrophysically-relevant ices with FELIX**

Jennifer Noble, Laboratoire de Physique des Interactions Ioniques et Moléculaires (PIIM), Université Aix Marseille-CNRS, Marseille, France

- **Overview of MD simulations for spectroscopies: gas phase for IR/Raman and liquid interfaces for SFG**

Marie-Pierre Gaigeot, Laboratoire d'Analyse, Modélisation, Matériaux pour la Biologie et l'Environnement (LAMBE), Université d'Evry Val d'Essonne-Université Paris Saclay-CNRS, Evry, France

- **Peptide fragmentation structures**

John Poutsma, Department of Chemistry College of William and Mary Williamsburg, USA

- **Use of CLIO to detect CN stretching band around 2100 cm^{-1} for laboratory tholin components (CxHyNz)**

Arpad Somogyi, Laboratory of Mass Spectrometry and Proteomics, The Ohio State University, USA

- **Presentation of a potential project using the FEL as probe and UV/Vis as pump**

Luke MacAleese, Institut Lumière Matière (ILM), Université Claude Bernard Lyon 1- CNRS, Lyon, France

- **The vibrational and electronic fingerprints of the sulfur-pi interaction in peptides**

Lucas Schwob, Deutsches Elektronen-Synchrotron, DESY, Hamburg, Germany

- **A Recent works on using IRMPD to trigger the synthesis of complex molecules**

Riccardo Spezia, Laboratoire de Chimie Theorique (LCT), Sorbonne Université-CNRS, Paris, France

List of participants

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Centre Laser Infrarouge d'Orsay

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One-day CLIO workshop for Infrared Free Electron Laser Users

Friday May 28th 2021

Lastname	Firstname	Institution	Laboratory	Country
Peremans	Andre	University of Namur	Laboratory of Lasers and Spectroscopy	BE
Perrin	Jonathan	Synchrotron SOLEIL	Synchrotron SOLEIL	FR
Pino	Gustavo	Universidad Nacional de Córdoba	Centro Láser de Ciencias Moleculares	AR
Pirali	Olivier	CNRS	Institut des Sciences Moléculaires d'Orsay	FR
Prazeres	Rui	CNRS - Université Paris-Saclay	Institut de Chimie Physique	FR
Poutsma	John	College of William and Mary	Department of Chemistry	USA
Rossa	Maximiliano	Universidad Nacional de Córdoba	Centro Láser de Ciencias Moleculares	AR
Rouquet	Etienne	Université Paris-Saclay	Institut des Sciences Moléculaires d'Orsay	FR
Rousseau	Patrick	Université de Caen Normandie	CIMAP	FR
Roy	Pascale	Synchrotron SOLEIL	AILES beamline	FR
Salpin	Jean-Yves	UEVE Université d'Evry Val d'Essonne, Université Paris-Saclay	LAMBE	FR
Schaller	Sascha	Fritz-Haber-Institut	Molecular Physics	DE
Schnell	Melanie	Christian-Albrechts-Universität zu Kiel	DESY	DE
Schöllkopf	Wieland	Fritz Haber Institute	FHI FEL	DE
Schwob	Lucas	Helmholtz-Gemeinschaft Deutscher Forschungszentren	Deutsches Elektronen-Synchrotron	DE
Scuderi	Debora	Université Paris-Saclay	Institut de Chimie Physique	FR
Serge	Della Negra	CNRS - Université Paris-Saclay	IJCLab	FR
Shafizadeh	Niloufar	CNRS	Institut des Sciences Moléculaires d'Orsay	FR
Somogyi	Arpad	The Ohio State University	Mass Spectrometry and Proteomics	US
Soulard	pascale	Sorbonne Université	MONARIS	FR
Spezia	Riccardo	Sorbonne Université & CNRS	Laboratoire de Chimie Théorique	FR
Taccone	Martín	Fritz Haber Institute of the Max Planck Society	Molecular Physics Department	DE
Tadjeddine	Abderrahmane	CNRS - Université Paris-Saclay	Institut de Chimie Physique	FR
Thissen	Roland	CNRS - Université Paris-Saclay	Institut de Chimie Physique	FR
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Institut de Chimie Physique

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Lastname	Firstname	Institution	Laboratory	Country
Wang	Yali	Université Paris-Saclay	Institute de chimie physique	FR
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Zhaunerchyk	Vitali	University of Gothenburg	Department of Physics	SE