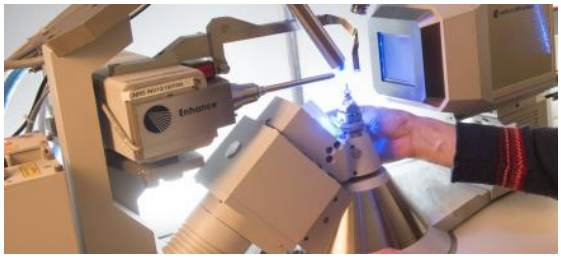
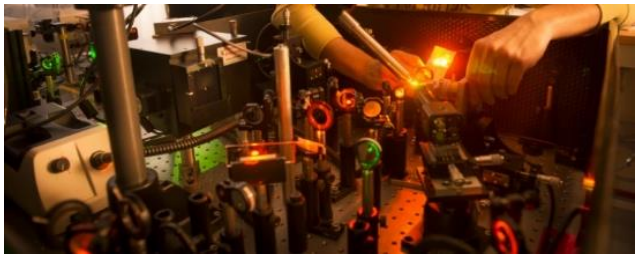
Thesis project:

Optical and Infrared control of electronic phase transitions in functional materials monitored by X-ray diffraction and ultrafast spectroscopies

Supervisor: Prof. Eric Collet **Start:** October 2024





Laboratories: Institute of Physics of Rennes, **Univ. Rennes – CNRS, France**

Collaboration with the International research laboratory DYNACOM, **University of Tokyo, Japan**

Skills and knowledge: Basic notions in solid state physics, phase transitions, crystallography & symmetry, spectroscopy, laser or X-ray diffraction

Project funding: Univ. Rennes, Institut Universitaire de France, ANR.

Positioning of the scientific project:

The Materials and Light department of the *Institut de Physique de Rennes* is a world-renowned player in the field of ultrafast photo-induced phase transitions in materials. The ability to control physical properties of materials on demand is an important challenge in science and technology. The use of light as external stimuli, spanning from UV to THz range, is very promising for contact-less, ultrafast and selective control. Ultrashort laser pulses, containing a number of photons of the order of the number of active units within materials, open the way for ultrafast and cooperative switching of their physical properties through photoinduced polarons or lattice instabilities, with various applications for photonic devices or light-based technologies. For optically driven photonic devices, memories, or actuators, the photo-response must combine important characteristics such as room temperature switching, wide thermal regime of bistability, photoinduced states that can persist long after stimuli, single laser shot with threshold switching and ultrafast dynamics.

Molecular materials undergoing photoinduced phase transitions show various types of functionalities switchable under photoexcitation, such as conductivity, ferroelectricity, etc [1-9]. Such multistable molecular materials can change their electronic state (charge or spin state) under the effect of temperature, pressure, light or even magnetic or electric fields [1-9]. Recently, we have been interested in systems for which the change of electronic state is coupled to a change of symmetry [1, 4-8], which makes the materials multi-functional such as (ferro)magnetic, ferroelastic and/or or ferroelectric. The coupling between change of electronic state and change of symmetry then makes it possible to control ferroelectricity by switching the electronic state using a magnetic field or light, to control magnetism by applying an electric field to switch the ferroelectric polarization, or to play on the ferroelastic distortions of the network. Crystallographic studies at equilibrium and/or under light irradiation make it possible to study microscopic processes at the crystalline and molecular scales.

The photo-induced phase transitions, initiated by an intense optical pulse, allow ultra-fast control of charge-transfer or spin state of materials by light (with photon energies of 1-3 eV). Cooperative response also appears in the ultrafast and out-of-equilibrium dynamics. However, the heat dissipation and the associated rise in temperature limit the control of coherent atomic movements and therefore of the functions of materials. It is then necessary to find other means for controlling the materials using lower energy optical excitation. The fascinating possibilities offered by Non-Linear Phononics (NLP) represent a great opportunity to control functional molecular materials [10]. NLP is an emerging approach which uses intense excitations in the infrared range (0.2 eV) to excite a high frequency crystal polar mode with a large amplitude, capable of coupling by nonlinear (anharmonic) terms and activate "soft modes" capable of driving a phase transition. This new field of investigation is only emerging in materials' science.

The methodology will consist of studying the changes driven by femtosecond optical or infrared light pulses in electronic state, crystalline and molecular structures or phonon modes, using X-ray diffraction, femtosecond optical [2] and IR spectroscopies and time-resolved X-ray diffraction [1,8] or spectroscopies [3]. The thesis project will focus on the development of ultra-fast optical and IR experiments in Rennes, using the femtosecond pump-probe techniques. We are also considering ultra-fast X-ray diffraction or spectroscopy experiments, to observe in real time the structural reorganization or the symmetry breaking, ultra-fast X-ray facilities (X-FEL, synchrotron). We also foresee femtosecond electron diffraction experiments in the frame of the International Research Laboratory DYNACOM lead by the Prof. E. Collet (Rennes) and S. Ohkoshi (Tokyo).

Bibliography and information:

Learn more about our group https://ipr.univ-rennes.fr/en/materials-and-light-departement

Publications in relation with the project:

[1] M. Hervé, G. Privault, E. Trzop, S. Akagi, Y. Watier, S. Zerdane, I. Chaban, R. G. Torres Ramírez, C. Mariette, A. Volte, M. Cammarata, M. Levantino, H. Tokoro, S. Ohkoshi, and E. Collet ***Nature Commun***. 15, 267 (2024)

[2] G. Azzolina, H. Tokoro, K. Imoto, M. Yoshikiyo, S. Ohkoshi and E. Collet ***Angew. Chemie Int. Ed***. 60, 232267 (2021)

[3] M. Cammarata, S. Zerdane, L. Balducci, G. Azzolina, S. Mazerat, C. Exertier, M. Trabuco, M. Levantino, R. Alonso-Mori, J. M. Glownia, S. Song, L. Catala, T. Mallah , S. F. Matar, E. Collet ***Nature Chemistry*** 13, 10-14 (2021)

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[10] A. Subedi, Phys Rev B 89, 220301 (2014)