PhD Thesis project

Photoinduced precursors and insulator-to-metal transition in quantum materials resolved with comprehensive optical spectroscopy

Quantum materials both challenge our understanding of solids and hold promise for a new generation of electronic and photonic devices. Many functionalities of quantum materials are based on phase transitions induced by conventional thermodynamic parameters, such as temperature, magnetic field or pressure [1]. A very promising field of research and application is to study and exploit transitions occurring out-of-equilibrium. In the latter case, the transformations are typically triggered by an ultrashort laser or electric pulse [2,3], and be driven on pathways often involving transient states of matter. Prominent examples of applications include the use of nonequilibrium insulator to metal transitions (IMT) for data storage or hardware neural networks for artificial intelligence. Understanding the mechanisms, the time scales and the conditions allowing ultrafast and nonequilibrium IMT is crucial for defining the fundamental limits of device operation [3].

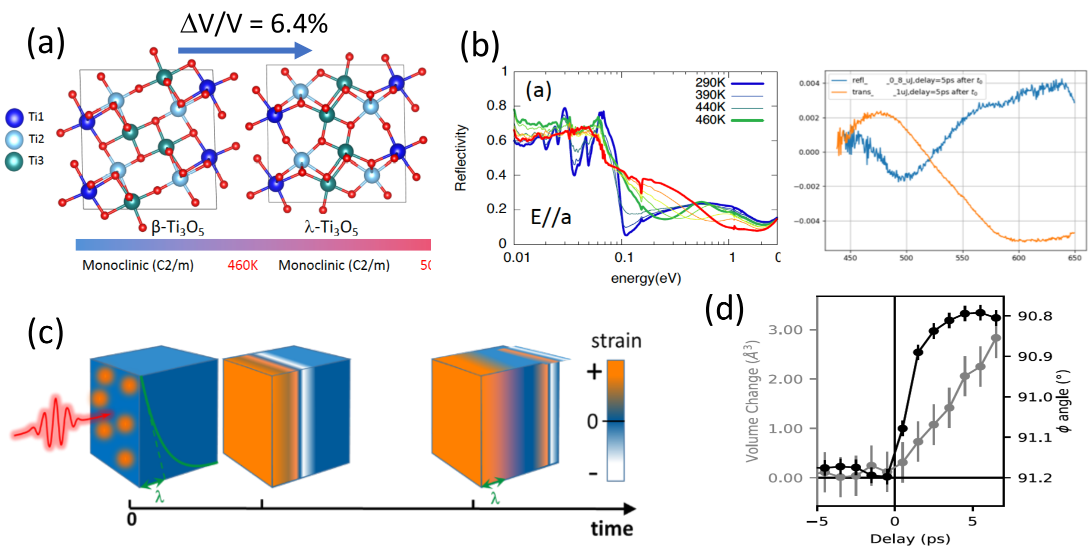

Figure *: (a) Phase transitions of the valence-bond insulator system Ti_3O_5 showing a large volume change without symmetry breaking. (b) Preliminary steady-state as function of temperature and time-resolved reflectivity spectra showing an IMT with large spectral weight transfer at the* β*-*Ti_3O_5 to λ-Ti_3O_5 transition. *(c) Schematic description of the strain wave mechanism. A stress is generated within fs after absorption of an ultrafast light pulse at a free surface (**green curve**) over the light penetration depth λ. This state is relaxed by propagation of a strain wave from the surface to the bulk at the sound velocity. It leaves behind a long-lasting compressed volume near the free surface.* (d) *XRD studies of* β*-*Ti_3O_5 *highlighting structural evidence of two different time scales associated to the volume and angle changes (performed at Swiss FEL).*

In this PhD, we want to address specifically nonthermal strain mechanisms at play during photoinduced phase transitions in select quantum materials. We will hinge our approach on the ultrafast setting of elastic stresses (orbital, electronic, polaronic) with a femtosecond laser pulse. We propose to develop this concept by focusing on the idea of triggering ultrafast strain-driven IMT. To this end, we will focus primarily on materials exhibiting large volume change without symmetry breaking, such as Ti_3O_5 [4]. Preliminary measurements (IPR/GREMAN internship, 2022) utilising ultrafast spectroscopy in Rennes, as well as broad bandwidth FTIR with Anvil Cells in Tours, reveal non-trivial reflectivity changes in a crystal and a thin film of Ti_3O_5. We would like to use this opportunity to endeavour in developing a fully-fledged spectroscopy of quantum materials, in particular in the topical area of photo-induced phase transitions. This will require a combination of complementary experimental tools, operating at equilibrium conditions and nonequilibrium pump-probe regime, across broad VIS-midIR spectrum. Importantly, theoretical input in response simulations to different stimuli (p, T, $h\nu$) will provide insofar lacking predictability and a new value to the field of correlated electron materials. The grand objective is to explore the path between the sub-ps microscopic precursor effects and the macroscopic phase transformation in the selected class of materials. Key challenge resides in discriminating between structural and electronic precursors seeding the transition, and the growth of long-range order. We will use several direct probes to follow both electronic and structural dynamics, such as time, energy and spatially resolved reflectivity, temperature and pressure dependent steady state broadband reflectivity. GREMAN and IPR have fully operating VIS-IR/THz setups that can host this PhD. A newly acquired Scanning Near-field Optical Microscope (SNOM) will be used in Tours to map local precursor effects with nanometric-spatial resolution. In Rennes, IPR is currently upgrading to a setup affording pump-probe microscopy-spectroscopy that should allow mapping phase separation/growth in real time. *Ab initio* calculations will be also employed using the calculation center of Région Centre Val de Loire (CaSciModOT). The PhD will benefit from collaborations established within the International Research Laboratory IRL "DYNACOM" [https://tokyo.cnrs.fr/cooperation-japan/dynacom/], both in terms of innovative quantum materials and unique experiments. For example, Ti_3O_5 will be synthesised at Tokyo University, and sub-10 fs optical spectroscopy for capturing early precursors (photo-induced polarons) will be available at Tohoku University.

Applicant:

The candidate is expected to be an experimentalist with a strong interest in fundamental and applied physics of condensed matter, and to have advanced knowledge in optics and laser physics. Knowledge in thin film deposition, optical spectroscopies and cryogenics are appreciated. Good communication skills are essential. Basics knowledge in quantum chemistry methods for electronic structure calculations will be considered as an advantage.

Practical aspects:

The position is available starting Oct. 2023 and lasts for 36 months. The gross monthly salary is about 2100 euros. The work will take place in IPR (Rennes) and GREMAN (Tours). Applicants should provide a CV, a letter of motivation and the names and email addresses of 2-3 references.

Contact:

V. TA PHUOC[1,3] (taphuoc@univ-tours.fr)
M. LORENC[2,3] (maciej.lorenc@univ-rennes1.fr)

[1] GREMAN, UMR 7347 CNRS, University of Tours, 37200 Tours
[2] Institut de Physique de Rennes, UMR 6251 CNRS, University of Rennes, 35000 Rennes
[3] DYNACOM IRL2015 University of Tokyo - CNRS - UR1, Tokyo 113-0033, Japan

[1] **V. Ta Phuoc**, C. Vaju, B. Corraze, R. Sopracase, A. Perucchi, C. Marini, P. Postorino, M. Chligui, S. Lupi, E. Janod, and L. Cario, "Optical Conductivity Measurements of GaTa4Se8 Under High Pressure: Evidence of a Bandwidth-Controlled Insulator-to-Metal Mott Transition", Phys. Rev. Lett. 110, 037401 (2013).
[2] V. Guiot, L. Cario, E. Janod, B. Corraze, **V. Ta. Phuoc**, M. Rozenberg, P. Stoliar, T. Cren, and D. Roditchev, "Avalanche Breakdown in GaTa4Se8−xTex Narrow-Gap Mott Insulators", Nature Commun. 4, 1 (2013).
[3] D. Babich, L. Cario, B. Corraze, **M. Lorenc**, J. Tranchant, et al., "Artificial Electro-Optical Neuron Integrating Hot Electrons in a Mott Insulator", *Physical Review Applied*, 17 (2022) 014040
[4] C. Mariette, **M. Lorenc**, H. Cailleau, E. Collet, L. Guérin, A. Volte, E. Trzop, R. Bertoni, (…), E. Janod, L. Cario, **V. Ta Phuoc**, "Strain Wave Pathway to Semiconductor-to-Metal Transition Revealed by Time-Resolved X-Ray Powder Diffraction", *Nature Commun.* 12, 1 (2021)