Nantes Université

cnrs

IMN
INSTITUT DES MATÉRIAUX DE NANTES JEAN ROUXEL

Topical meeting GDR MEETICC:

New challenges in quantum materials: artificial intelligence and ultrafast science

Nantes, 5-6 décembre 2023

RÉGION PAYS DE LA LOIRE

Amplitude

Introduction

Cette réunion thématique du GDR MEETICC vise à faire le point sur deux thèmes émergents qui commencent à impacter largement le domaine des matériaux quantiques.

Le premier enjeu concerne l'intelligence artificielle (IA). D'une part, l'IA devrait impacter largement le domaine des matériaux quantiques, à la fois pour la recherche de nouveaux matériaux et également pour la prédiction de propriétés physiques. D'autre part, les propriétés des matériaux quantiques sont aujourd'hui considérées pour réaliser des dispositifs implémentant les deux briques de base des réseaux de neurones matériels (synapses et neurones artificiels). L'espoir à terme est de créer une IA à la fois efficace et sobre énergétiquement, à l'inverse de l'IA actuelle basée sur des réseaux de neurone logiciels.

Le deuxième thème en plein essor abordé sera celui de la science ultrarapide et de son impact sur l'étude des matériaux quantiques. L'utilisation impulsion laser ultra-brève constitue d'une part un nouvel outil permettant de sonder les matériaux quantiques aux échelles de temps pertinentes sur certains de leurs degrés de liberté (fs pour les excitations électroniques, ps pour les modifications structurales). Plus intéressant encore, il est aujourd'hui possible de modifier l'état des matériaux quantiques pour induire des modifications électroniques et structurales pouvant piloter un changement spectaculaire de leurs propriétés physiques inaccessibles à l'équilibre thermodynamique. Cela peut même conduire à la commutation ultra-rapide de l’état macroscopique du matériau et donc de sa fonction.

****

Introduction

This topical meeting of the GDR MEETICC focuses on two emerging issues which start to impact the field of quantum materials.

The first theme concerns artificial intelligence (AI). On the one hand, AI is expected to largely impact the field of quantum materials, both for the search for new materials and also for the prediction of physical properties. On the other hand, the properties of quantum materials are today considered to create devices implementing the two basic building blocks of material neural networks (synapses and artificial neurons). The long-term hope is to create an AI that is both efficient and energy efficient, unlike current AI based on software neural networks.

The second growing theme addressed will be ultrafast science and its impact on the study of quantum materials. The use of ultra-short laser pulses is on the one hand a new tool for probing quantum materials at the relevant time scales on certain of their degrees of freedom (fs for electronic excitations, ps for structural modifications). Even more interesting, it is today possible to modify the state of quantum materials to induce electronic and structural modifications that can drive a spectacular change in their physical properties inaccessible to thermodynamic equilibrium. This can even lead to ultra-rapid switching of the macroscopic state of the material and therefore its function.

Remerciements

Cette réunion a pu être organisée grâce aux soutiens financiers :

- du CNRS au titre de sa subvention annuelle au GDR MEETICC
- du CNRS via le programme MITI 80’ FAST-IA,
- de la région Pays de la Loire via le projet de démonstrateur régional Mott-IA,
- de Nantes Université via son support au Laboratoire de Recherche International (IRL) DYNACOM.

Acknowledgements

This meeting was made possible thanks to financial support from:

• from the CNRS under its annual subsidy to the GDR MEETICC

• from the CNRS via the MITI 80’ FAST-IA program,

• the Pays de la Loire region via the Mott-IA regional demonstrator project,

• from Nantes University via its support to the International Research Laboratory (IRL) DYNACOM.

Table of contents

5 Dec 2023

Quantum Materials and AI -1

IA pour les matériaux quantiques

Ex : Apport de l'IA pour la prédiction de propriétés magnétiques

Marie-Bernadette Lepetit

a. Institut Néel, CNRS UPR 2940, Grenoble
b. Groupe de Théorie, Institut Laue Langevin, Grenoble

Les méthode l'intelligence artificielle et notamment les méthodes d'apprentissage profond par réseaux de neurones constitue une nouvelle donne dans la recherche de nouveaux matériaux. En effet, ce nouvel outil a démontré sa puissance dans de nombreux domaines (modèles de language, traitement d'images etc). La communauté des chimistes à la recherche de nouveaux matériaux s'en est emparé depuis quelques années pour accélérer la découverte de nouveaux matériaux (recherche dans des bases de données, conception de nouveaux métériaux par algorithme évolutionnaires etc.). La communauté des théoriciens commence à le faire pour accelérer les codes de calcul, développer des potentiels effectifs ou même déterminer les fonctions d'onde quantiques. Parallèlement la communauté des physiciens commence à utiliser ces méthodes pour déterminer les propriétés de matériaux connus ou améliorer les processus expérimentaux (traitement de grandes quantités de données ou au contraire concentrer les mesures sur les points porteur du maximum d'information).

Après une revue rapide et non exhaustive de la manière dont les méthodes d'intelligence artificielle sont aujourd'hui utilisées dans le domaine des matériaux quantiques, nous prendrons l'exemple des propriétés magnétiques pour voir plus en détail comment l'apprentissage profond peut aider à la prédiction d'un Hamiltonien magnétique de base énergie et à son diagramme de phase.

Machine Learning Guided Synthesis of Materials with Specific Structures and Properties

Romain Gautier, Rachid Laref, Florian Massuyeau

The discovery of new materials with specific properties is a complex task due to the drastic effects that very slight modifications on the synthesis of materials can have on the crystal structures and the resulting properties. Machine learning can be a very useful tool in such situations and can drastically accelerate the discovery of new functional materials as well as assist us to rationalize complex mechanisms.

After a brief introduction to the potential of artificial intelligence in materials science, the accelerated discovery of new hybrid perovskite by machine learning will be presented. These materials have recently shown a great potential in optoelectronic applications. For this reason, many research groups are currently exploring this chemical system to discover new low dimensional hybrid perovskites. However, discovering such materials is challenging as the necessary structure determination by X-ray diffraction is time consuming and non-perovskite compounds are very often synthesized instead of the more interesting perovskites. In this presentation, two approaches will be introduced which both guide us towards the synthesis of new hybrid perovskites: (i) the use of deep learning to automatically identify hybrid perovskites from powder X-ray diffraction patterns,[1] and (ii) the use of molecular descriptors to identify organic amines with high probability to lead to the synthesis of hybrid perovskites.[2] From this machine learning tools, the scientists' ability to predict the synthesis of new hybrid perovskites is largely augmented and such algorithms could be included in autonomous materials discovery cycles in the future.

References:

[1] F. Massuyeau, T. Broux, F. Coulet, A. Demessence, A. Mesbah, R. Gautier, *Advanced Materials* **2022**, *34*, 2203879.
[2] R. Laref, F. Massuyeau, R. Gautier, *Small* **2023**, *Accepted*, 2306481.

Quantum materials and ultrafast science - 1e

Ultrafast evolution of electronic states in a photoexcited Peierls-Mott insulator

J. Dong[1], D. Shin[2], E. Pastor[3], T. Ritsche[4], L. Cario[5], Z. Chen[6], W. Qi[1], R. Grasset[1], M. Marsi[6], A. Taleb-Ibrahimi[7], N. Park[8], A. Rubio[9], E. Papalazarou[6] and L. Perfetti[1]

[1] *Laboratoire des Solides Irradiés, CEA/DRF/IRAMIS, CNRS, Ecole Polytechnique, Institut Polytechnique d Paris, 91128 Palaiseau, France*
[2] *Department of Physics and Photon Science, Gwangju Institute of Science and Technology (GIST), Gwangj 61005, Republic of Korea*
[3] *Institute of Advanced Materials (INAM), Universitat Jaume I, Avenida de Vicente Sos Baynat, s/n 12006, Castello, Spain*
[4] *Institut fur Festkorper- und Materialphysik, Technische Universitat Dresden, 01069, Dresden, Germany*
[5] *Université de Nantes, CNRS, Institut des Matériaux Jean Rouxel, IMN, Nantes, F-44000, France*
[6] *Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, 91405 Orsay, France*
[7] *Société civile Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin - BP 48, 91192 GIF-sur-YVETTE, France*
[8] *Department of Physics, Ulsan National Institute of Science and Technology (UNIST), UNIST-gil 50, Ulsan 44919, Korea*
[9] *Max Planck Institute for the Structure and Dynamics of Matter and Center for Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany*

The transition metal 1T-TaS_2 is a layered insulator with a rich phase diagram. Soci\'{e}t\'{e} civile Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin - BP 48, 91192 GIF-sur-YVETTE, France as a function of pressure and temperature. Its broken symmetry phases include incommensurate, nearly commensurate, an commensurate Charge Density Waves (CDWs). Within each layer, the Ta lattice undergoes a periodic distortio in which 13 Ta ions form clusters with the motif of a Star-of-David (SD). These clusters have an odd filling and lock-in to a Commensurate CDW (C-CDW) below 180 K. The observed insulating behavior of the C-CDW phase is generally attributed to the Mott localization of the electron in the highest occupied state of SDs. Recent calculations revised the strength of Coulomb repulsion in this family of compounds and highlighted the st effects that electronic interactions have on the band structure of 1T-TaS_2.

Although widely believed to be a Mott insulator, the commensurate CDW phase also features an interlay stacking with SDs dimerization. The stacking of two adjacent layers can be of three different kinds: Top Aligne (*A*) and Laterally displaced (*L*) with a vector of magnitude $2a$. The dimerized geometry of 1T-TaS_2 is formed by alternating stacking between *A* and *L* configurations, called *AL* stacking. By hosting an even number of electrons, the dimerized unit cell of the commensurate CDW cannot be a pure Mott phase. This duality gave origin to several works, addressing the Slater-towards-Mott character of the ground state.

This work reports time-resolved ARPES measurement on high-quality single crystals of 1T-TaS_2 in the insulating C-CDW phase. By making use of different polarizations of the probe pulse, we are able to visualize the dispersion of electronic states below and above the chemical potential. The experimental data are compared with state-of-the-art Density Functional Theory calculations with the Generalized Orbital U (DFT+GOU). The Coulomb U of a SD cluster is self-consistently calculated via the ACBN0 method. Our results indicate that both stacking order and electronic correlations are essential to reproduce the correct gap size. Moreover, time-resolved ARPES data acquired with S polarized probe disclose novel aspects of the photoinduced phase transition. The pump pulse erases the band dispersion and halves the gap magnitude within half a period of the coherent CDW motion. Besides the oscillations of CDW amplitude. We propose that photoexcitation also engenders local variations of dimerization, orbital filling, and U potential. The combination of these effects triggers the melting of the Mott-Peierls gap.

References

[1] D. Shin, N. Tancogne-Dejean, J. Zhang, M. S. Okyay, A. Rubio, N. Park, *Physical Review Letters* **126**, 196406, (2021).
[2] J. Dong, D. Shin, E. Pastor, T. Ritschel, L. Cario, Z. Chen, W. Qi, R. Grasset, M. Marsi, A. Taleb, N. Park, A. Rubio, L. Perfetti, E Papalazarou, *arXiv:2210.11052*

Effect of magnetic excitations on electron dynamics in the vicinity of the Mott transition

Evgeny A. Stepanov[1,2,*]

[1]*CPHT, CNRS, École polytechnique, IP Paris, 91120 Palaiseau, France*

[2]*Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France*

[*]*evgeny.stepanov@polytechnique.edu*

The non-equilibrium dynamical mean-field theory (DMFT) [RMP 86, 779 (2014)] is a state-of-the-art method for describing the time-evolution of a strongly correlated problem under an external time-dependent perturbation. However, this approach is unable to account for the effect of the non-local electronic correlations that may strongly affect physical properties of the system.

Advancing the theoretical description beyond the non-equilibrium DMFT is computationally challenging. Currently, the GW+DMFT [PRL 118, 246402 (2017); PRB 100, 041111(R) (2019), PRB 100, 235117 (2019)] method can be seen as the most advanced numerical approach that can be used for this purpose. Unfortunately, this theory is able to treat only charge fluctuations and thus misses important non-local magnetic effects. Moreover, already this simplest extension of DMFT makes the description of the time evolution of the driven system extremely expensive numerically, which, at first glance, does not give any room for a further improvement of the method.

In this talk, I will show that the diagrammatic structure of a recently developed dual triply irreducible local expansion (D-TRILEX) method [PRB 100, 205115 (2019); PRB 103, 245123 (2021); SciPost Phys. 13, 036 (2022)] suggests an inexpensive way of improving diagrammatic extensions of the non-equilibrium DMFT. Indeed, upon neglecting three-point vertex corrections in diagrams for the self-energy and polarization operator, the D-TRILEX method reduces to an analog of the GW+DMFT theory that, however, accounts for all leading channels of instability (including non-local magnetic fluctuations) simultaneously. Further, I will demonstrate that using the non-equilibrium D-TRILEX approach for describing the electron dynamics in the vicinity of the Mott transition uncovers an important effect of high-energy magnetic excitations in addition to the known contribution of the low-energy (magnon) modes.

Quantum materials and AI - 2

The Multifaceted Impact of Memristors on Neuromorphic Systems

Damien Querlioz
Centre de Nanosciences et de Nanotechnologies of Université Paris-Saclay and CNRS

Memristors, also known as resistive memories, are nanodevices that mimic artificial synapses and possess the potential to revolutionize neuromorphic design. This talk explores the transformative capabilities of memristors by examining their application in neuromorphic systems. While these technologies have matured sufficiently to enable the fabrication of complete systems, they present challenges due to their atomic-scale features, leading to a high degree of variability. Through examples of fabricated hybrid CMOS/memristor circuits, we demonstrate various techniques that allow neuromorphic systems to harness the benefits of memristors while mitigating their drawbacks. In analog neuromorphic systems, memristors serve as in-memory computational units. Their unpredictable behavior necessitates the use of dedicated programming strategies to achieve energy-efficient artificial intelligence. To illustrate this, we showcase a neural network for arrhythmia detection that maintains stable accuracy over an extended period of two months. Furthermore, we emphasize the potential of memristors in digital systems, which can yield low-power and highly robust systems capable of self-powered operation. We present a Bayesian machine that demonstrates near immunity to single-event upsets. Additionally, we describe a fabricated digital neural network consisting of 32,768 memristors powered by a miniature solar cell. This system seamlessly switches between exact and approximate computation depending on the available energy. Lastly, we delve into the realm of stochasticity, where memristor variability can be transformed into an advantage, driving superior efficiency and robustness. We showcase how this variability can enable neural networks to function as Bayesian neural networks capable of estimating prediction uncertainty. Furthermore, we demonstrate that memristor variability facilitates a form of Bayesian learning, particularly effective in situations with limited data.

Van der Waals ferroelectric heterostructures for in-memory computing and beyond Moore electronics

Jean-Francois Dayen[1,2]
1 Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg
2 Institut Universitaire de France (IUF), 1 rue Descartes, 75231 Paris cedex 05, France.
E-mail: **dayen@unistra.fr**

2D ferroelectric materials are attracting fast growing interest for the implementation of complex more-then-Moore and beyon-Moore architectures that are challenging to design with standard thin film technology.[1] Here, I will present recent developments in my team on the coupling of a 2D vdW electon gas with various ferroelectic gate controls. I will explain how these systems allow for rethinking circuit topology and memory-logic interaction, opening up new research directions in the area of frugal computational enhancement and neuromorphic computing for AI.

I will first show how by making use of the switchable polarization state of two splitted ferroelectric gates, the electrical potential landscape within a semiconductor channel can be permanently and reconfigurably modified.[2] While using the non-volatile ferroelectric states encoded in each gate, the ferroelectric logic circuits can function as six alternative logic gates, while CMOS circuit are limited to a single function. Moreover, the Re-FeFET circuit demonstrates high compactness, with an up to 80% reduction in transistor count compared to standard CMOS design. Last but not least, the device can operate as a photodiode and generate photovoltaic energy.

Then I will present how light-structure interactions in vdW systems allow for implementing the non-volatile electrical and optical control of the ferroelectric polarization in all-vdW ferroelectric/semiconductor heterostructures.[3] The wavelength-dependent study unveils ferroelectric polarization control and decouples the mechanisms driven by photogenerated carriers for each material. The vdW Ferroelectric Field-effect transistors show On/Off ratios exceeding 10^7, large hysteresis memory windows, and multiple remanent states, sorting them as good artificial synapse candidates. Following, long-term potentiation/depression, and spike rate-dependent plasticity are shown using electrical control. Moreover, the synaptic functionalities were complemented by the unique dual optical and electrical control, enabling optically stimulated and optically assisted synaptic devices. We benchmark our device with a simulated artificial neural network and achieve an excellent accuracy level of 91%, close to the ideal synaptic case (96%).

The combination of photo-ferroelectric functionalities, neuromorphic plasticity, and high-logic expressivity with reconfigurability at runtime, put all-VdW ferroelectric/semiconductor heterostructures on the roadmap for novel in-memory computing architectures.

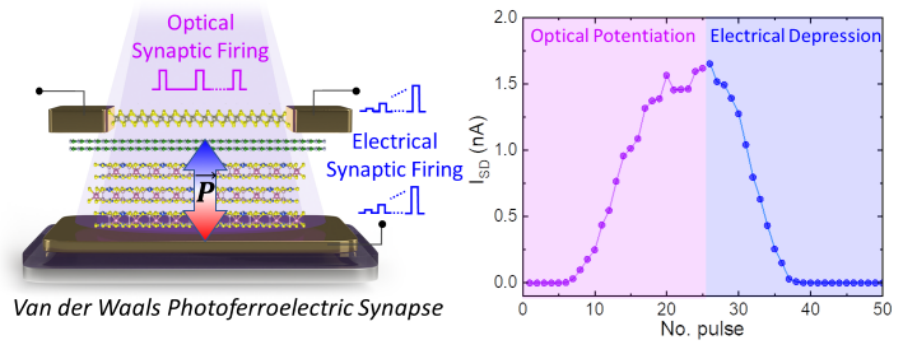


Figure 1: Left: schematic of the device. Right: Photoferroelectric Synaptic Plasticity.

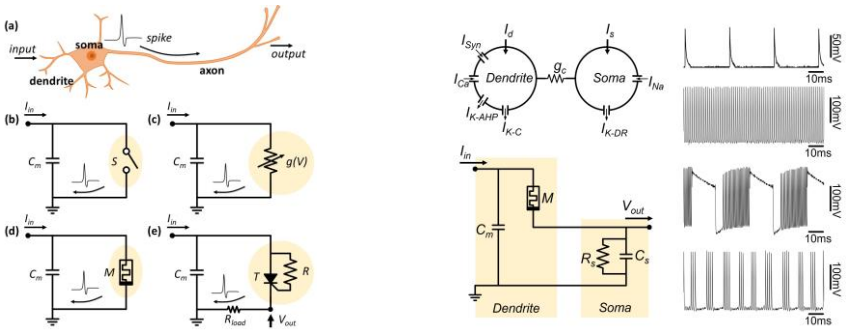
References

[1] Jin, T. et al. ACS Nano (2022) doi:10.1021/acsnano.2c07281.
[2] *A. Ram. et al.* ACS Nano 2023, 10.1021/acsnano.3c07952.
[3] M. Soliman, et al., ACS Appl. Mater. Interfaces 2023, 15, 12, 15732.

Bursting dynamics in a spiking neuron with a memristive voltage-gated channel

Jiaming Wu[1], Kang Wang[1], Olivier Schneegans[2], Pablo Stoliar[3], Marcelo Rozenberg[1]
[1]*Université Paris-Saclay, CNRS Laboratoire de Physique des Solides, Orsay, France*
[2]*Université Paris-Saclay, CNRS Supelec, France*
[3]*National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan*

We introduce a memristive two-terminal device that can be used for artificial neurons with rich dynamical properties [1]. It consists of a silicon controlled rectifier (SCR) and a resistor, exhibiting resistance switching behavior similar to Mott materials [2, 3]. Based on this device, we construct an artificial neuron model that generates spikes through a single voltage-gated channel. Our circuit implementation is of unprecedented simplicity, using just four electronic components, all conventional, cheap and out-of-the-shelf [4]. We also develop a two-compartment neuron model similar to the Pinsky-Rinzel model, implementing complex neuronal behaviors such as tonic spiking, fast spiking, and two types of intrinsic burst spiking. We obtain the full phase diagram and discuss the origin of different regions. We find that the spike traces of this model bear a significant resemblance to experimental biological neuronal recordings. With this artificial neuron platform, we further implement important foundational neural circuit models such as Winner-Takes-All (WTA) and Central Pattern Generator (CPG). Our work provides an exceedingly simple, cost-effective, and readily accessible platform for neuromorphic engineering. Additionally, it may open up a new way to investigate neural pathologies, such as epilepsy and Parkinson's disease, from the study of the phase diagram and the transitions between spiking states of physical neuron models [5].



Reference

[1] J. Wu, et al. Neuromorphic Computing and Engineering, accepted.

[2] P. Stoliar, et al. Advanced materials, 2013, 25(23): 3222-3226.

[3] del Valle J, et al. Nature, 2019, 569(7756): 388-392.

[4] M. Rozenberg et al. Scientific Reports, 9, 11123(2019). Featured in Nature's Device and Material Engineering blog

[5] M. Rozenberg. Journal of Physics: Conference Series, 2023, 2533(1): 012007.

Quantum materials and ultrafast science - 2

Ultrafast structural dynamics and control of phase transitions

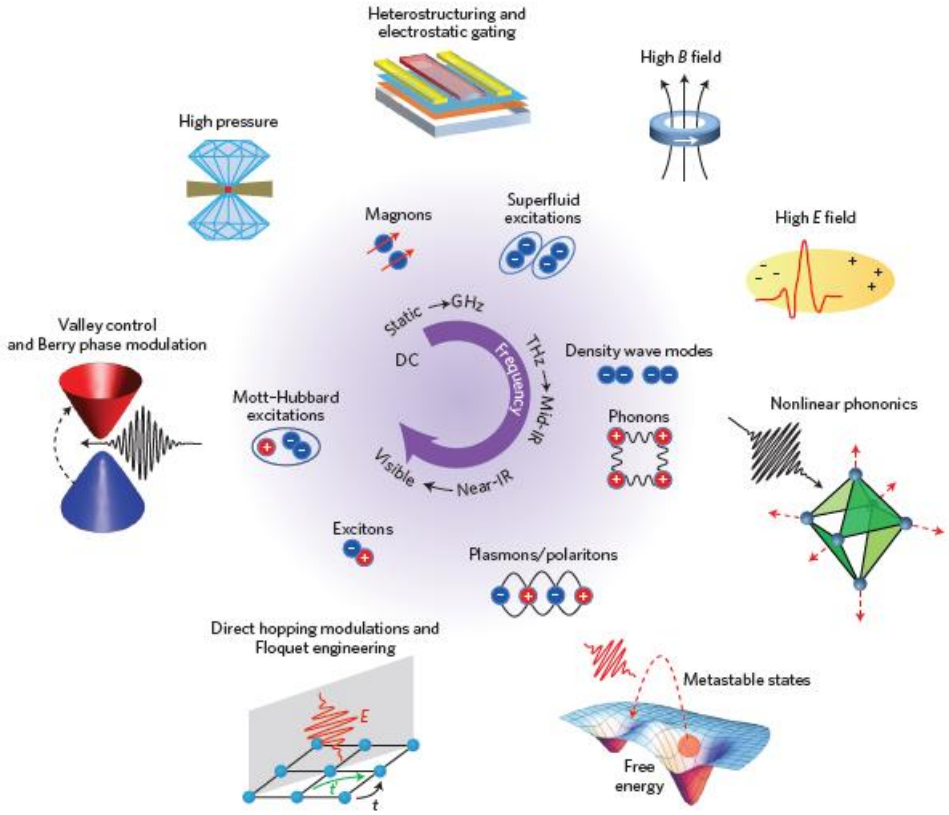
Herve Cailleau[1,2], Maciej Lorenc[1,2], Etienne Janod[3,2'],
Celine Mariette [4,2], Shinichiro Iwai [4,2]
[1]*IPR, Rennes, France*
[2]*IRL Dynacom, University of Tokyo, Japan*
[3]*IMN, Nantes, France*
[4]*ESRF, Grenoble, France*
[5]*Department of Physics, Tohoku University, Japan*

Thanks to remarkable developments of ultrafast techniques operating on a few femtoseconds timescale, *i.e.* faster than atomic motions, many macroscopic functionalities of quantum materials can be manipulated on demand in unprecedented ways [1]. For example, ultrafast laser systems allow to photoexcite a macroscopic number of carriers (typically one percent of valence band electrons) and hence to strongly perturbe the electronic state of quantum materials. The resulting electronic, magnetic and structural reorganization can be probed at a controlled in real-time by spectroscopies and X-ray/ electron scattering.

In many quantum materials the interplay of electronic and structural degrees of freedom is crucial for determining the phase diagram at thermal equilibrium, in particular in correlated electron systems. Thermodynamics and symmetry considerations offer an universal approach of these systems able to exhibit an isosymmetric (isostrucrural) insulator to metal transition and symmetry breaking ordering.

Structural dynamics limits the time scale of the transformation. When atomic displacements in the unit cell play a dominant role, as in ferroelectric or CDW systems, the relevant time scale is determined by the period of coherent optical phonons. For phase transitions involving volume and or shear ferroelastic deformation the dynamical limits are set by the propagation of strain waves [2,3].

In this talk, we will give a brief overview of the main features of ultrafast photoinduced phase transitions: multiscale process, coherent dynamics, cooperative feedback and threshold, … Some particular questions will be addressed such as the differences between isosymmetric and symmetry breaking dynamical transitions, the role of elastic interactions, the change from local precursors to phase transition at macroscopic scale, the new opportunities with photoinduced strain waves and the role of material morphology, …



From reference [1]

References

[1] D.N. Basov *et al.*, Towards properties on demand in quantum materials. *Nature Mater.* 16, 1077 (2017)

[2] C Mariette *et al.,* Strain wave pathway to semiconductor-to-metal transition revealed by time-resolved X-ray powder diffraction. *Nature Commun.* 12, 1–11 (2021).

[3] T. Amano *et al.*, under review preprint https://doi.org/10.21203/rs.3.rs-3239079/v1

Optical control of quantum materials with disorder

Ernest Pastor[1]

[1] *CNRS, Univ Rennes, IPR (Institut de Physique de Rennes) - UMR 6251, F-35000 Rennes, France.*

A solid can exist in different crystal forms with unique chemical and physical properties. Interchange between such forms or polymorphs is known as a solid-solid phase transition and is a ubiquitous process in nature that can be induced by changes in temperate and pressure. From a technological viewpoint, the capacity to stabilise solid phases is paramount to harness their properties and generate new applications.

Light excitation offers an alternative way to pressure and temperature to control a phase transition by exploring non-equilibrium dynamics. In this framework, the excitation of the solid in one of the phases can launch coherent motion of atoms in multiple unit cells [1,2]. Re-excitation of the sample alongside the period of the relevant vibrational modes that connect the two phases can potentially modulate the energy required to drive the transition.

Inspired by recent demonstrations of coherent control of solids [3,4], in this talk I will discuss routes to control the solid-solid phase transition in (3D) VO_2. This is a prototypical material that undergoes an insulator to metal transition accompanied by a change in structure from monoclinic to rutile. I will show multi-pulse experiments in which we assess how dividing the total energy between several pulses affects the energy required to initiate the phase transition, namely the transition's fluence threshold.

We find that we are unable to exert coherent control over the sample, despite a measurable coherent phonon signal. However, our data reveals a large incoherent decrease in the fluence threshold when the transition is optically modulated. Time-resolved total x-ray diffraction measurements indicate that excitation of the monoclinic structure results in the population of incoherent phonons within 50fs that manifest as specific 2D patterns of the diffuse scatter. These data, alongside DFT calculations, suggest that photoinduced correlated disorder can provide a path to control the phase transition. In this talk, I will discuss the supporting data and its possible implications in our ability to control solids on demand (see ref [5])

References

[1] Johnson, A. S.; Moreno-Mencía, D.; Amuah, E. B.; Menghini, M.; Locquet,

J.-P.; Giannetti, C.; Pastor, E.; Wall, S. E. Phys. Rev. Lett. 2022, 129 (25), 255701.

[2] Pastor, E.; Moreno-Mencía, D.; Monti, M.; Johnson, A. S.; Fleischmann, N.; Wang, C.; Shi, Y.; Liu, X.; Mazzone, D. G.; Dean, M. P. M.; Wall, S. Phys. Rev. B 2022.

[3] Horstmann, J. G.; Böckmann, H.; Wit, B.; Kurtz, F.; Storeck, G.; Ropers, C. Nature 2020, 583 (7815), 232–236.

[4] Maklar, J.; Dong, S.; Sarkar, J.; Gerasimenko, Y. A.; Pincelli, T.; Beaulieu, S.; Kirchmann, P. S.; Sobota, J. A.; Yang, S.-L.; Leuenberger, D.; Moore, R. G.; Shen, Z.-X.; Wolf, M.; Mihailovic, D.; Ernstorfer, R.; Rettig, L. arXiv June 8, 2022. http://arxiv.org/abs/2206.03788).

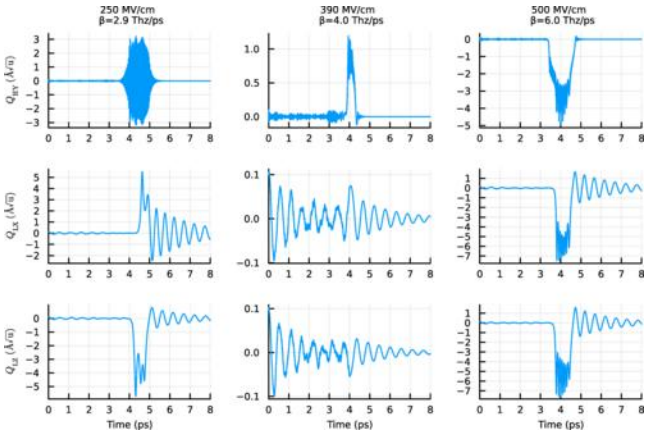
[5] Johnson A.S.; Pastor E.; Batlle-Porro S.; Benzidi H.; Katayama T.; de la Peña Muñoz G.A.; Krapivin V.; Kim S.; López N.; Trigo M.; Wall S.E. ArXiv Sep. 2023 (arXiv:2309.13275)

Light-induced translation symmetry breaking via nonlinear phononics

Alaska Subedi[1]

[1]*CPHT, CNRS, École polytechnique, Institut Polytechnique de Paris, 91120 Palaiseau, France*

Light has a wavelength that is usually longer than the size of the unit cell of crystals. Hence, even intense light pulses are not expected to break the translation symmetry of a material. However, certain materials, including $KTaO_3$ exhibit peaks in their Raman spectra corresponding to their Brillouin zone boundary phonons due to second-order Raman processes, which provide a mechanism to drive these phonons using intense midinfrared lasers. We investigated the possibility of breaking the translation symmetry of $KTaO_3$ by driving these high-frequency phonons. Our first principles calculations show that the energy curve of the lowest-frequency phonon mode at the Brillouin zone boundary point *X* softens and develops a double-well shape when either the transverse or longitudinal highest-frequency mode at *X* is displaced from its equilibrium position. We performed similar first principles calculations as a function of the high frequency modes and electric field to extract the nonlinear coupling between them. These were then used to construct the coupled equations of motion for the phonon coordinates in the presence of an external pump term on the the high frequency modes, which we numerically solved for a range of pump frequencies and amplitudes. We find that externally driving the high frequency modes can lead to oscillations of the lowest-frequency zone boundary mode at a displaced position, hence, breaking the translation symmetry of the material. Furthermore, we find that the coupling between the electric field and highest-frequency longitudinal mode is negative, which also leads to a displaced oscillation of this mode when driven. However, we find that the coupling between the electric field and the driven modes is weak, and the translation symmetry breaking only occurs for extremely large values of the pump electric field.



Posters

Efficient optimization of the synthesis parameters of thermoelectric legs using a machine learning approach

Sylvain Le Tonquesse [1,2], Hugo Bouteiller [1,2], Michihiro Ohta[3], Takao Mori [4], Franck Gascoin [1], David Berthebaud [1,5]

[1] *Normandie Université, ENSICAEN, UNICAEN, CNRS, CRISMAT, 14000, Caen, France*

[2] *CNRS-Saint-Gobain-NIMS, Laboratory for Innovative Key Materials and Structures (LINK), Tsukuba, Japan*

[3] *National Institute of Advanced Industrial Science and Technology (AIST), Umezono 1-1-1, Tsukuba, Ibaraki, Japan*

[4] *National Institute for Materials Science (NIMS), WPI-MANA, 1-1-1 Namiki, Tsukuba 305-0044, Japan*

[5] *Nantes Université, CNRS, Institut des Matériaux de Nantes Jean Rouxel, IMN, F-44000 Nantes, France*

The deposition of high-quality electrical contacts on thermoelectric (TE) materials is of uttermost importance for the fabrication of reliable and efficient TE generators [1,2]. However, this challenging processing step is often neglected in academic researches. High-quality electrical contacts must present, at the same time, good adhesion on the materials as well as low electrical and thermal interfacial resistances. The simultaneous optimization of the contact material's composition and the numerous deposition parameters are generally realized using a highly time-consuming try-and-error approach.

This poster presents an approach using machine learning (ML) which aims at optimizing more efficiently a large number of experimental parameters from a relatively small ($\approx$ 20) initial set of samples [3]. This approach was applied to the direct synthesis of Yb_4Sb_3 TE legs [4] by Spark Plasma Sintering by optimizing simultaneously the synthesis parameters (temperature, time, uniaxial pressure...) and the contact's composition (13 metals were considered) in order to minimize the electrical contact resistance. The prediction performances of different types of ML algorithms and data descriptors (both from experiments and online database) as well as the mechanical and electrical properties of a number of predicted samples are reported. Finally, the advantages and weaknesses of this ML approach applied to the optimization synthesis conditions are also discussed.

[1] S. Le Tonquesse *et al.*, *Crystals*, 11, (2021), 1140
[2] S. El Oualid *et al.*, *Adv. Energy Mater.*, 11, (2021), 21005
[3] G. Lambard *et al.*, *Scripta Mater.*, 209, (2022), 114341
[4] S. Le Tonquesse *et al.*, *ACS Appl. Energy Mater.* (2023)

Electron Energy Loss Spectroscopy of the Metallic and Mott Insulator phases of $(V_{1-x}Cr_x)_2O_3$

Mohammad DOLATABADI[1],Chris LEROUX[1] Eric GAUTRON[1], Laurent CARIO[1], Benoit CORRAZE[1], Julien TRANCHANT[1], Philippe MOREAU[1], Etienne JANOD[1]

[1] Nantes Université, CNRS, Institut des Matériaux de Nantes Jean Rouxel, IMN, F-44000 Nantes, France

Mott insulators are a class of material, which are predicted to be metal according to the conventional band theory, but behave as insulators due to a strong electron-electron repulsion not taken into consideration in the conventional band theory . The insulator to metal transition (IMT) in the prototypical Mott insulator $(V_{1-x}Cr_x)_2O_3$ has attracted enormous attention either from the fundamental point of view or for its applications in microelectronics. Several mechanisms can lead to such an IMT in this compound, such as manipulating the temperature, the pressure, or chemical doping. Besides these conventional ways of driving an IMT, another stimulus that has recently emerged in Mott insulators is the application of electric pulses. An electric pulse above a certain threshold leads to creation of a filamentary metallic path in a Mott insulator matrix, yielding to a low resistance state. This low resistance state can relax back to the high resistance state by application of an electric pulse. Hence, this material can switch between different resistance states, paving the way for memory and artificial synapse applications. The nature of the electric-pulse-induced metallic state is currently hotly debated.

In this context, advanced characterizations are clearly required, ideally at the (sub) nanometer scales. Thanks to the recent advances in transmission electron microscopy (TEM), probing this IMT in Mott insulators is now feasible at atomic scales. Before engaging an *in-situ* study of the electric-pulse-induced IMT, we have characterized the main spectroscopic features of the metallic and the insulating phases of $(V_{1-x}Cr_x)_2O_3$ Mott insulator. In this poster, we will present the main results of the study on the metallic and the insulating phases of this compound by electron energy loss spectroscopy (EELS). In particular, we unveil a difference unnoticed so far in the energy of the volume plasmon oscillation between the metallic and insulating phases. This analysis hence provides a spectroscopic criterion to distinguish between these two phases as a consequence of the change in the electronic structure. This criterion will allow us to map the metallic filament inside an insulating matrix with a spatial resolution of a few nanometers in our future experiment that aims to induce the insulator to metal transition *in-situ* inside TEM. However, performing such an EELS experiment requires very thin single crystal lamellae. In order to prepare our samples, we used focused ion beam (FIB) coupled with a scanning electron microscope (SEM). The sample preparation process is also presented in this poster.

Electron-phonon coupling and ultrafast dynamics of hot carriers in semiconductors: from interpretation of photoemission experiments to transport simulations in devices

Raja Sen, Nathalie Vast, and Jelena Sjakste

Laboratoire des Solides Irradiés, CEA-DRF-IRAMIS, École Polytechnique, CNRS UMR 7642, Institut Polytechnique de Paris, 91120 Palaiseau, France

Electron-phonon coupling determines the charge transport properties of materials as well as the relaxation dynamics of photoexcited carriers. Computational methods based on density functional theory (DFT), on the one hand, and time-energy- and momentum-resolved spectroscopy, on the other hand, allow today an unprecedently detailed insight into the role of the electron-phonon coupling [1]. At the same time, light-induced hot carriers in semiconductors attract much attention in the context of emerging concepts for next-generation energy conversion devices.

In this work, we will present the theoretical and experimental results for relaxation dynamics of photoexcited electrons in semiconductors. Our computational method based on DFT and on interpolation of the electron-phonon matrix elements in Wannier space allowed us to successfully interpret the dynamics of photoexcited electron relaxation in Si and GaAs, in good agreement with two-photon photoemission experiments [2-4]. Here, we also aimed to clarify the relative importance of different scattering channels in momentum- and energy- relaxation of hot carriers, which can only be obtained from *ab initio* calculations. Furthermore, our work also sheds light on the necessity of time-resolved transport simulations for understanding the dynamics and transport properties of excited electrons at the transient regimes [4].

References

[1] J. Sjakste, K. Tanimura, G. Barbarino, L. Perfetti, and N. Vast, J. Phys: Cond. Mat. **30**, 353001 (2018).
[2] H. Tanimura, J. Kanasaki, K. Tanimura, J. Sjakste, N. Vast, M. Calandra, F. Mauri, Phys. Rev. B 93, 161203(R) (2016).
[3] H. Tanimura, J. Kanasaki, K. Tanimura, J. Sjakste, and N. Vast, Phys. Rev. B **100**, 035201 (2019).
[4] R. Sen, N. Vast, J. Sjakste, Appl. Phys. Lett. 120, 082101 (2022).

Finding Relationship Between the Local Structure and Electronic Orbital Occupation Using Machine Learning

Wenhao Zhang[1,2], David Berthebaud[3,4], Jean-Francois Halet[3] and Takao Mori[1,2]

1. WPI Center for Materials Nanoarchitectonics (WPI-MANA), NIMS

2. Graduate School of Pure and Applied Sciences, University of Tsukuba

3. CNRS-Saint-Gobain-NIMS, Link lab, NIMS

4. Present address: Nantes Université, CNRS, Institut des Matériaux de Nantes Jean Rouxel, IMN, F-44000 Nantes, France

Machine learning (ML) methods extract statistical relationships between inputs and results. When the inputs are solid-state crystal structures, structure–property relationships can be obtained. In this work, we investigate whether a simple neural network is able to learn the 3d orbital occupations for the transition-metal (TM) centers in crystalline inorganic solid-state compounds using only the local structure around the transition-metal centers described by rotationally invariant fingerprints based on spherical harmonics and one-hot elemental encoding. A multilayer neural network trained on density functional theory (DFT) results of about 1800 samples was developed and showed good performance in predicting the TM orbital occupations (for both spin channels). We study in detail how the local structure affects the predictions of the local properties and how they provide physical insights for the design of a future machine learning model for materials chemistry. The proposed ML method is illustrated in practical application by predicting local magnetic moments of the transition-metal atoms in a full set of inorganic structures with large unit cells. Although less accurate compared to the experimental data, the ML results compared well with the DFT results, suggesting the feasibility of electronic property prediction based only on structure input.

MADGUI: Material Design Graphical User Interface for Active Learning Assisted by Bayesian Optimization Applied to Materials Research and Development

Christophe Bajan[1], Guillaume Lambard[1], & Florent Pawula[1,2]

[1] *Energy Materials Design Group, Research and Services Division of Materials Data and Integrated System (MaDIS), National Institute for Materials Science (NIMS), Tsukuba, Japan*

[2] *CNRS-Saint Gobain-NIMS, UMI 3629, Laboratory for Innovative Key Materials and Structures (LINK), National Institute for Materials Science, Tsukuba Japan*

We present MADGUI a MAterial Design Graphical User Interface (GUI) that uses Bayesian Optimisation and Machine Learning model to perform data-analysis and optimise process or composition. Users with no programming knowledge can easily operate MADGUI, which is developed using the Streamlit library in Python. MADGUI is divided into three parts, users only need to prepare a tabular datasheet as a *.csv or a *.xlsx file providing experimental features (e.g., compositional ratios, process parameters and target properties e.g., resistivity, thermal conductivity, device efficency, etc. Users can choose to optimise single or multiple targets simultaneously, such as minimising one property while maximising another, with the ability to set equal or different ratios of importance between them. Overall, the GUI is a user-friendly and efficient tool for data analysis and alloy optimisation, making it suitable for experimentalists and theoreticians.

Single wavelength operating neuromorphic device based on graphene-ferroelectric transistor

Krishna Maity[1], Jean-François Dayen[1], Bernard Doudin[1], Roman Gumeniuk[2], Bohdan Kundys[1*]

[1] *Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504, 23 rue du Loess, Strasbourg, F-67000, France.*

[2]*Institut für Experimentelle Physik, TU Bergakademie Freiberg, Leipziger Str. 23, Freiberg 09596, Germany.*

Despite recent progress in electrical and electro-optical simulations of machine learning devices, the all-optical non-thermal function remains challenging, with single wavelength operation still elusive. We have recently demonstrated entirely monochromatic way of neuromorphic signal processing, eliminating the need for electrical pulses [1]. Multi-level synaptic potentiation-depression cycles are successfully achieved optically by leveraging photovoltaic charge generation and polarization within the photoferroelectric substrate interfaced with the graphene sensor (Fig. 1).

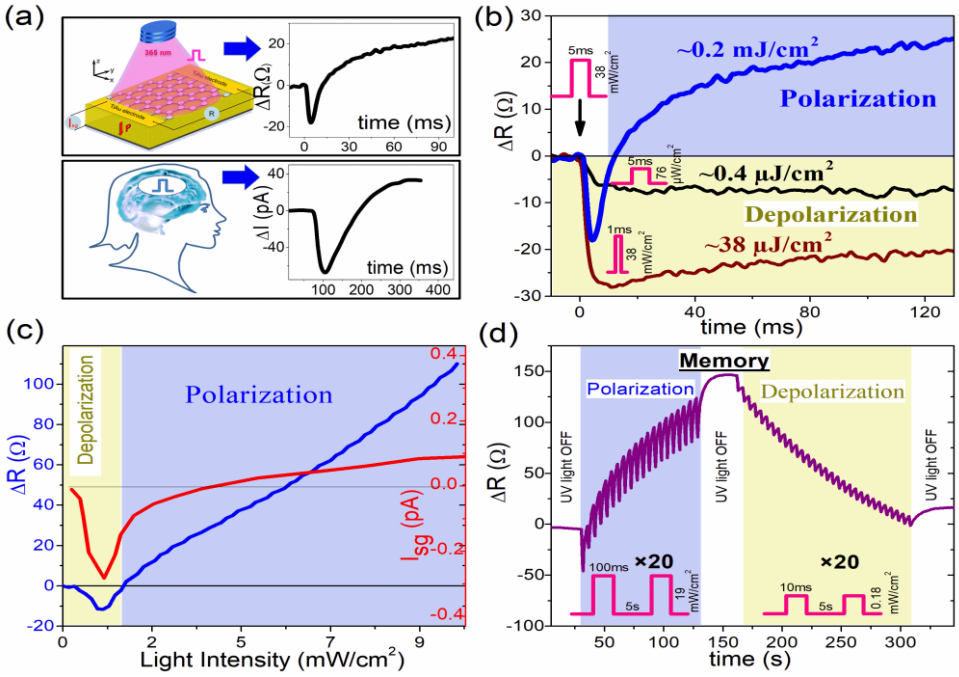


Fig. 1. (a) Optical excitation schematics and bio-like neuron response (b) Graphene resistance response to UV pulses. (c) Light intensity-dependent graphene resistance and related source-gate current. (d) Single-wavelength potentiation and depression of graphene resistance.

[1] K. Maity, et al. ACS Appl. Mater. Interfaces, https://doi.org/10.1021/acsami.3c10010 (2023).

Email: *kundys@ipcms.fr

Toward the electric-plasmonic coupling in Cr-doped V_2O_3

Nathanaël Blond[1], Mohamad Haydoura[1], Julien Tranchant[1], Daniel Bedau [2], Benoit Corraze[1], Etienne Janod[1], Laurent Cario[1], Bernard Humbert[1], Jean-Luc Duvail[1]

[1]*Institut des matériaux de Nantes Jean Rouxel, Nantes, France*

[2]*Western Digital San Jose Research Center, CA, California*

Mott insulators are a class of quantum materials exhibiting an insulator-metal transition (IMT) which presents great potential for future applications and an interesting challenge for fundamental research. Indeed, they are among the probable prospects to replace the flash technology for memory usage according to the International Roadmap for Devices and Systems. Furthermore, the unique properties of these strongly correlated materials offer potential applications, such as resistive random-access memories (RRAM) and even artificial intelligence (AI) hardware based. These applications are based on the Mott transition under electric field which results in a sharp resistance change occurring over a few nanoseconds. The great promises of these materials consist in a very low power consuming technology, and a possible huge gain in performances both for the memory application and for AI. The very unique and intriguing physics of these materials are still under investigation. The Mott IMT can be described by the dielectric breakdown. The two main factors responsible for the transition are : i) the electric field and ii) the number of electrons that can be accelerated by the applied electric field.

In this work, we propose to exploit localized surface plasmon resonance (LSPR) to interplay with the IMT. LSP are excited in metal nanoparticles when irradiated with nIR and visible light. They promote - a strong enhancement of the local electromagnetic field, - hot electrons injection in the medium surrounding the metal nanoparticles, - heating at the nanoscale. This study aims to give evidence of a potential electric/plasmonic coupling. It would allow to control the Mott transition through opto-electric - or even fully optically - stimuli.

In this scope, we work on devices with a planar geometry exploiting thin films of the canonical Mott insulator $(V_{0.95}Cr_{0.05})_2O_3$ associated to gold nanoparticles. We confirmed that the very thin vanadium oxide layers possess the expected crystallinity and microstructure. It results in a volatile switching behavior comparable to that observed for thicker films. The optical properties of gold nano-islands deposited on the oxide layers have been optimized to get the plasmonic resonance in the 600 – 1000 nm range. The electric/plasmonic coupling is under investigation.

Towards data storage and neuromorphic applications based on $(Cr_{1-x}V_x)_2O_3$ Mott insulator thin films

M. Haydoura [1], M. Rodriguez Fano [1], N. Zémal Aboubacar [1], J. Tranchant [1], B. Corraze [1], E. Janod [1], M.-P. Besland [1] and L. Cario[1]

[1] CNRS, Institut des Matériaux de Nantes Jean Rouxel, (IMN), F-44000 Nantes, France

Mohamad.haydoura@cnrs-imn.fr

Resistance random-access memory (RRAM) is considered as a promising candidate to replace Flash memory technologies that approach their limits due to miniaturization and short economical payback [1]. In that context, canonical Mott insulators such as $(Cr,V)_2O_3$ or AM_4Q_8 have recently attracted a considerable attention thanks to the electric-field-driven insulator to metal transitions (IMT) that could be used in memory and neuromorphic applications [2-4]. Previous studies have demonstrated competitive memory performances in Mott insulator thin films, but so far these studies overlooked the variability of the devices [5]. Here we investigate the resistive switching in Mott insulators thin films of $(Cr_{1-x}V_x)_2O_3$ (x = 0.7) integrated in micro-sized memory cells and report for the first time the cycle to cycle, device to device variability and state retention of the high resistive state (HRS) and low resistive state (LRS). To this end, thin films were deposited and annealed in order to reach the expected stoichiometry and a good crystalline quality. The electrical behavior was investigated on MIM structures using 2 µm via memory cells made of 50 nm thick $(Cr_{0.30}V_{0.70})_2O_3$ Mott insulator thin films. Preliminary experiments on these devices using short electric pulses for the SET and RESET show an endurance of more than 2000 cycles (Figure 1.a), with very narrow cumulative distribution function (CDF) curves (see Fig 1.b). Moreover, both HRS and LRS exhibit a very good temporal stability at room temperature, as well as a state retention that can be extrapolated up to 10 years (Fig 1.c). This work shows therefore the potential of $(Cr_{0.30}V_{0.70})_2O_3$ Mott insulator thin films for integration into the next generation of non-volatile memories and neuromorphic devices.

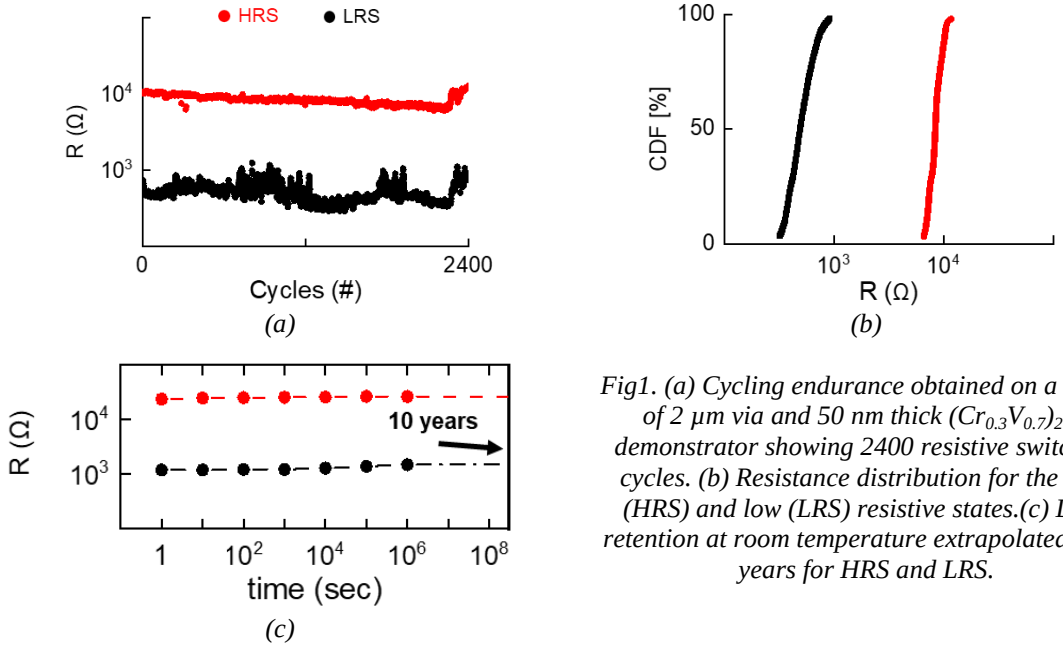


Fig1. (a) Cycling endurance obtained on a device of 2 µm via and 50 nm thick $(Cr_{0.3}V_{0.7})_2O_3$ demonstrator showing 2400 resistive switching cycles. (b) Resistance distribution for the high (HRS) and low (LRS) resistive states.(c) Data retention at room temperature extrapolated to 10 years for HRS and LRS.

[1] D. Ielmini 2016 *Semicond. Sci. Technol.* 31 063002. DOI 10.1088/0268-1242/31/6/063002. **[2]** L. Cario et al., *Adv. Mater., 22, 5193–5197, 2010.* **[3]** E.Janod, et al., *Adv. Funct. Mater., 25, 6287–6305, 2015.* **[4]** *L.*Cario et al. Metal Oxides; Elsevier, 2022, DOI 10.1016/B978-0-12-814629-3.00010-6. **[5]** J. Tranchant et al. *Thin Solid Films, 533, 61-65, 2013 and IEEE IMW, 74-77, 2018.*

Ultrafast insulator-to-metal Mott transition carried by photoinduced strain waves at acoustic velocities

J. Guzman-Brambila[1,2,3], T. Amano[4], D. Babich[1], R. Mandal[2], A. Volte[2], E. Trzop[2], M. Servol[2], E. Pastor[2], M. Alashoor[2], J. Larsson[5], A. Jurgilaitis[5], V.-T. Pham[5], D.Kroon[5], J. C. Ekström[5], B. Ahn[5], J. Tranchant[1], B. Corraze[1], L. Cario[1], V. Ta Phuoc[6], R. Sopracase[6], M. Guillon[6], H. Itoh[4], Y. Kawakami[4], C. Mariette[3], H. Cailleau[2], M. Lorenc[2], S. Iwai[4], E. Janod[1]

[1]*Institut des Matériaux de Nantes Jean Rouxel.*
[2]*Institut de Physique de Rennes.*
[3]*European Synchrotron Radiation Facility, Grenoble.*
[4]*Tohoku University, Sendai (Japan).*
[5]*MAX IV Laboratory, Lund University.*
[6]*GREMAN Tours*

Digital technologies pervade our lives, yet their energy consumption continues to rise. Shifting from Von Neumann computer architecture to neural network-based systems could alleviate this energy burden. Inspired by Mottronics, our focus lies in inducing insulator-to-metal transitions (IMTs) in Mott insulators[1]. A key question for future applications is to established the ultimate switching time between the insulating and metallic phases. Leveraging ultrafast physics, we explore non-thermal pathways to steer materials toward distinct functional states.

Our work spotlights a key but overlooked mechanism driving ultrafast phase transitions involving volume and/or ferroelastic deformation: photoinduced elastic waves[2]. Our study demonstrates how this strain wave mechanism triggers the ultrafast IMT in V_2O_3 Mott material (Antiferromagnetic Insulator, $I2/a$, to Paramagnetic Metal $R\bar{3}c$ [3]). Through time-resolved optical reflectivity and X-ray diffraction (Fig. a) on granular thin films, we unravel the underlying physics. We showcase the strain wave's role in rapid changes, with or without symmetry breaking (Fig. b). We reveal an initial inverse ferroelastic shear wave preceding a volume-compressive strain wave, operating within ferroelastic domains and throughout film thickness, respectively[4].

This detailed exploration sheds light on ultrafast phase transitions in quantum materials, specifically Mott insulators, offering insights for future Mott insulator-based devices by unraveling the ultimate switching IMT times.

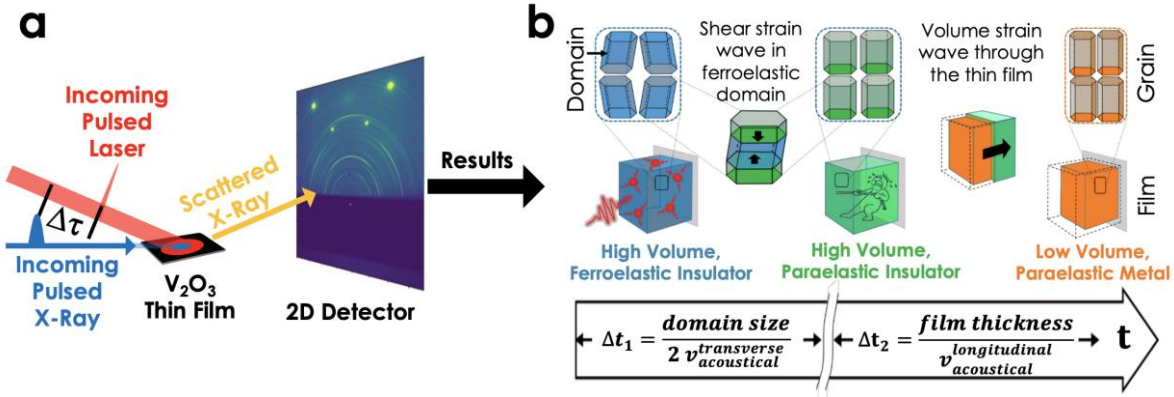


Figure a: Laser pump, X-Ray probe Diffraction Scheme. X-ray and Laser pulses are synchronized with a time delay Δt and recorded via a CCD 2D detector. **Figure b: Schematic illustrating the photoinduced strain waves driving the AFI-to-PM transition in granular thin film of V2O3.** Ultrafast laser pulse builds-up of shear and volume mechanical stress. At first, the stress is relaxed by launching a shear strain wave within each ferroelastic domain of the granular film, Δt_1. A compressive volume strain wave propagates from free surface of the film towards the substrate, leaving in its wake the insulator to metal transition, Δt_2.

References

1. Janod *et al*. (2015). Resistive Switching in Mott Insulators and Correlated Systems. *Adv. Func. Mater. 25*, 6287.

2. Thomsen *et al*. (1986). Surface generation and detection of phonons by picosecond light pulses. *PRB 34*, 4129.

3. McWhan *et al*. (1970). Metal-Insulator Transition in $(V_{1-x}Cr_x)_2O_3$, *PRB 2, 3734*

4. T. Amano *et al*. (2023). Under review in Nat. Phys. https://doi.org/10.21203/rs.3.rs-3239079/v1

6 Dec 2023

Quantum materials and ultrafast science - 3

Ultrafast Electron Microscopy

Florian Banhart[1]
[1]*Institut de Physique et Chimie des Matériaux, UMR 7504, Université de Strasbourg, Strasbourg, France*

Conventional transmission electron microscopy (TEM) working with continuous electron beams are used for the characterization of materials by imaging at the sub-Angstrom *spatial* scale, by electron diffraction and by electron energy-loss spectroscopy (EELS). Working with short photoelectron pulses instead of continuous beams allows an increase of the *temporal* resolution of TEMs by many orders of magnitude so that dynamic processes from the nano- to the femtosecond time scale are getting accessible.

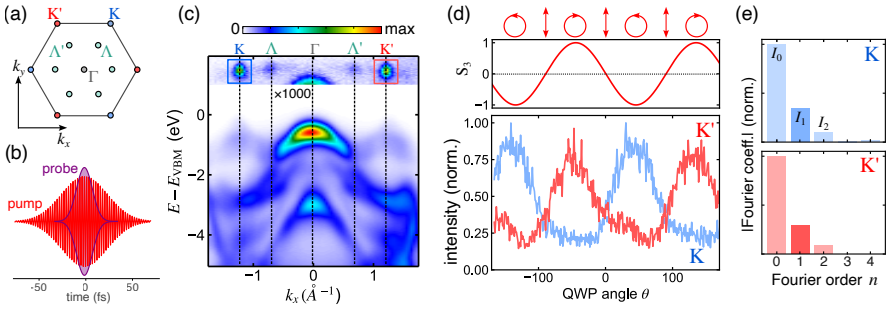
The presentation will give an overview of the potential of ultrafast TEM (UTEM) in materials research and in quantum optics with electron waves. The focus will be on recent developments of UTEM and on studies of different types of nanomaterials that are carried out in Strasbourg. The combination of high spatial with high temporal resolution allowed us to study the kinetics of fast irreversible chemical reactions in nanoparticles at the nanometer and nanosecond scales. Furthermore, photoswitchable nanomaterials such as spin-crossover systems or oxide nanocrystals are studied to reveal and optimize the switching time of nanosystems and to obtain information on heat transfer at the nanoscale. Finally, the potential of UTEM in quantum optics, where the states of free electrons can be manipulated by photon fields, will be outlined (though not a research field in Strasbourg) to bridge the gap between electron microscopy and quantum physics.

Berry Curvature Signatures in Chiroptical Excitonic Transitions

Samuel Beaulieu[1], Shuo Dong[2,3], Viktor Christiansson[4], Philipp Werner[4], Tommaso Pincelli[3,5], Jonas D. Ziegler[6], Takashi Taniguchi[7], Kenji Watanabe[8], Alexey Chernikov[6], Martin Wolf[3], Laurenz Rettig[3] Ralph Ernstorfer[3,5] and Michael Schüler[9]

[1] *Université de Bordeaux - CNRS - CEA, CELIA, UMR5107, F33405 Talence, France* [2] *Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China* [3] *Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany* [4] *Department of Physics, University of Fribourg, 1700 Fribourg, Switzerland* [5] *Institut für Optik und Atomare Physik, Technische Universität Berlin, Strasse des 17 Juni 135, 10623 Berlin, Germany* [6] *Institute of Applied Physics and Würzburg-Dresden Cluster of Excellence ct.qmat, Technische Universität Dresden, 01062 Dresden, Germany* [7] *Research Center for Materials Nanoarchitectonics, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan* [8] *Research Center for Electronic and Optical Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan* [9] *Laboratory for Materials Simulations, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland*

The topology of the electronic band structure of solids can be described by its Berry curvature distribution across the Brillouin zone. We theoretically introduce and experimentally demonstrate a general methodology based on the measurement of energy- and momentum-resolved optical transition rates, allowing to reveal signatures of Berry curvature texture in reciprocal space. By performing time- and angle-resolved photoemission spectroscopy of atomically thin WSe_2 using polarization-modulated excitations, we demonstrate that excitons become an asset in extracting the quantum geometrical properties of solids. We also investigate the resilience of our measurement protocol against ultrafast scattering processes following direct chiroptical transitions.



Optical polarization-modulated pump-probe photoemission in monolayer WSe_2. (a) Sketch of the Brillouin zone of WSe_2 with the high-symmetry

points. (b) Sketch of the overlapping pump and probe pulses. (c) Optical polarization-averaged trARPES signal along k_x (K-Λ-Γ-Λ'-K').(d) Ellipticity factor (Stokes parameter S_3) of the pump pulse, which is controlled by the continuous rotation of quarter-wave-plate (QWP) angle θ (top panel), along with the ellipticity-resolved photoemission intensity of excited states around the K and K' points. (e) The absolute value of the Fourier coefficients associated with the polarization-modulated photoemission intensities from excitonic states in (d).

Quantum Avalanche in Correlated Insulators Driven by DC Fields

There is a considerable discrepancy between the predicted and experimental switching fields in correlated insulators driven by a strong DC bias. This calls for a reevaluation of the current microscopic understanding of this non-equilibrium phenomenon.

We unveil an electron avalanche mechanism that occurs in the bulk of such insulators at arbitrarily low electric fields. This quantum avalanche relies notably on the inelastic scattering of electrons on a phononic medium and it results from the generation of in-gap states through multi-phonon emission. The phonon spectrum dictates the occurrence of a two-stage versus a single-stage switching mechanism, which we relate to charge-density-wave and Mott resistive switchings, respectively.

Within this unified framework, the behavior of electron and phonon temperatures, along with the temperature dependence of switching fields, illustrates a crossover between a purely quantum mechanical and a thermal scenario.

Based on
Han J.E., Aron C., et al., Nat Commun **14**, 2936 (2023)
https://doi.org/10.1038/s41467-023-38557-8

Quantum materials and AI - 3

Dynamics of the electric field-induced Mott transition

Pascale Diener, Houda Koussir, Yevheniia Chernukha, Maxime Berthe, Bruno Grandidier

Mott materials are archetypal strong electronic correlations systems. The study of their non-equilibrium properties is not only a promising approach for the understanding of these systems but is also essential for optimizing Mottronic devices currently under development.

This presentation will first review our current knowledge on the electric field induced, out of equilibrium Mott transition [1]. We will then study the evolution of the volatile transition as a function of the sample and electrodes geometry, using an advanced multiple probes tunneling microscopy setup [2]. Our results agree with a linear decrease of the characteristic delay time of the transition with the system size, underlining the high potential of Mott materials for the development of fast, low consumptic neuromorphic devices.

[1] L. Cario, J. Tranchant, B. Corraze, and E. Janod, in Metal Oxides for Non-Volatile Memory, edited by P. Dimitrakis, I. Valov, and S. Tappertzhofen (Elsevier, 2022), pp. 307–360.

[2] H. Koussir, I. Lefebvre, M. Berthe, Y. Chernukha, J. Tranchant, B. Corraze, E. Janod, L. Cario, B Grandidier, and P. Diener, J. Phys.: Conf. Ser **2164**, 012046 (2022).

Mott electric transition for neuromorphic components

J. Tranchant, M. Haydoura, B. Corraze, E. Janod, M.-P. Besland and L. Cario
Nantes Université, CNRS, Institut des Matériaux de Nantes Jean Rouxel, IMN, F-44000 Nantes, France

The application of electric pulses within the ns to µs range on narrow gap Mott insulators engenders an original phenomenon of resistive switching (RS). This electric Mott transition is volatile above threshold electric fields of a few kV/cm and stabilizes into a reversible non-volatile insulator-to-metal transition for higher values. Our works indicate that this electric-field-driven phenomenon originated from an electronic avalanche through the creation of hot carriers [1]. This local breakdown induces the collapse of the Mott insulating state at the nanoscale and brings about the formation of granular conductive filaments. These properties are universal to all canonical Mott insulators, including oxides such as $(V_{1-x}Cr_x)_2O_3$, or chalcogenides such as $NiS_{2-x}Se_x$ or the AM_4Q_8 family of compounds (A=Ga, Ge; M=V, Nb, Ta, Mo; Q=S, Se)) [2]. These properties have been observed both on single crystals and thin films. According to our recent results on $(V_{0,95}Cr_{0,05})_2O_3$ thin films, this filamentary insulator-to-metal transition is associated to an isostructural phase separation involving a compression of the lattice, rooted in Mott physics.

Among all narrow gap Mott insulators, the $(V_{1-x}Cr_x)_2O_3$ system displays both volatile and non-volatile transitions at room temperature, which makes it relevant for microelectronic applications. Volatile transition can be used to implement the basic functionalities of Leaky-Integrate-and-Fire (LIF) artificial neurons in the form of single components [3]. Our recent results demonstrate that light is an additional control parameter for the volatile transition, and that electro-optic artificial neurons can be built out of Mott insulators [4]. On the other hand, the non-volatile transition is suitable for application as memories. Indeed, our last results display reversible RS in thin films of various compositions within the $(V_{1-x}Cr_x)_2O_3$ solid solution. Such $TiN/(V_{1-x}Cr_x)_2O_3/TiN$ devices display very competitive memory performances and highlight the increase of the memory window with the bandgap [5]. Besides, the stabilization of intermediate resistance levels between high and low resistance states of these Mott memories opens the way to artificial synapses application. These studies thus enable to envision artificial neural networks based on Mott insulators for both artificial neurons and synapses as single components. Finally, all these works pave the way towards a new type of electronics based on the Electric Mott transition: Mottronics.

[1] P. Diener et al. Phys. Rev. Lett. 121, 016601 (2018)

[2] E. Janod *et al.* Adv. Funct. Mater. 25, 6287 (2015)

[3] P. Stoliar *et al.* Adv. Funct. Mater. 2017

[4] D. Babich et al., Phys. Rev. Appl., 17, 014040 (2022)

[5] M. Rodriguez-Fano *et al.*, ACS-AMI (2023); doi: 10.1021/acsami.3c09387

Mott insulating 1T-$TaSe_2$ monolayer on GaP(111)B

Y. Chernukha, H. Koussir, C. Sthioul, E. Haber, N. Peric, L. Biadala, P. Capiod, M. Berthe, I. Lefebvre, X. Wallart, B. Grandidier, P. Diener

[1]*Univ. Lille, CNRS, Centrale Lille, Univ. Polytechnique Hauts-de-France, Junia-ISEN, UMR 8520-IEMN, F-59000 Lille, France*

The appearance of a Mott insulating state was recently reported in a single layer of a transition metal dichalcogenide 1T-$TaSe_2$. This makes a 1T-$TaSe_2$ monolayer a good candidate for a two-dimensional Mott material with great potential for Mottronic device applications. However, until now, the growth of 1T-$TaSe_2$ monolayer was carried out mainly on graphene substrates whose high conductivity limits its device integration.

Here we present a study of 1T-$TaSe_2$ monolayer deposited on semi-insulating and doped gallium phosphide substrates by molecular beam epitaxy. Scanning tunneling microscopy reveals a flower-shaped superstructure (Figure 1 a) attributed to a combination of a Moiré pattern and charge density wave reconstruction. An open gap of more than 120 meV was observed by STS (Figure 1 b). A temperature-dependent resistance measurements showed a presence of Mott insulating phase up to 400 K. An electrical continuity of the monolayer was confirmed by four-probe transport measurements performed with multiprobe STM.

We will take this opportunity to present the experimental development of the technique combining the multiprobe STM and laser pump-probe technique.

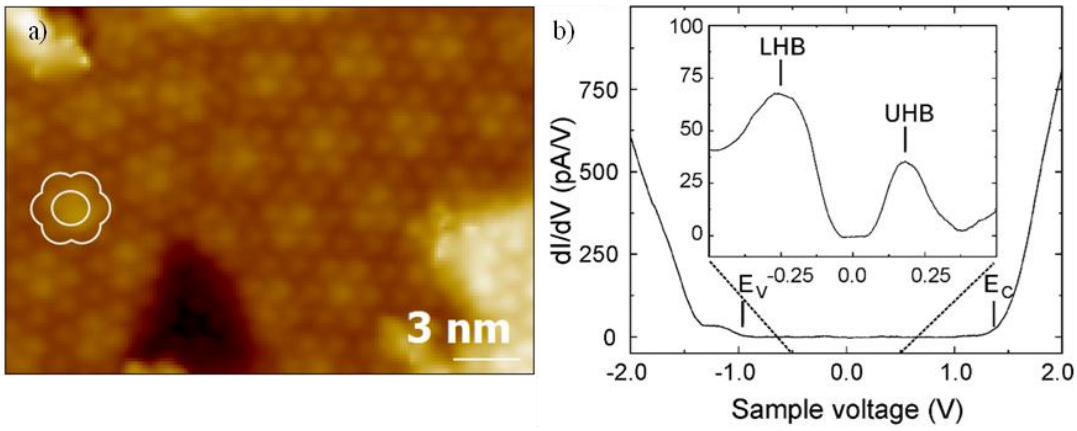


Figure 1. a) Scanning tunneling microscopy image and b) tunneling spectroscopy of 1T-$TaSe_2$ monolayer on GaP

Quantum materials and ultrafast science - 4

Time resolved Raman scattering in quantum materials

Kota Katsumi[1], Alexandr Alekhin, Laurène Gatuingt[1], Sarah Houver[1], Maximilien Cazayous[1] , Alain Sacuto[1], Amir Haguiguirad[2], Sofia Souliou[2], Matthieu Le Tacon[2], Yann Gallais[1]

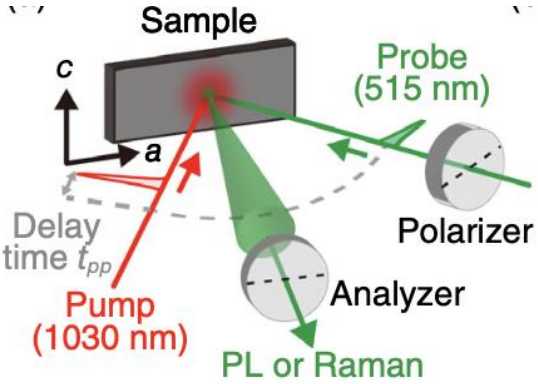
[1]*Matériaux et Phénomènes Quantiques, Université Paris Cité, CNRS, Paris, France*

[2]*Karlsruhe Institute of Technology, Karlsruhe, Germany*

Controlling materials properties with light pulses is an emerging field in condensed matter research. Quantum materials with their often delicate interplay of magnetic, electronic, orbital and lattice degrees of freedom offer an attractive playground to demonstrate optically induced novel phases out-of-equilibrium with unique properties.

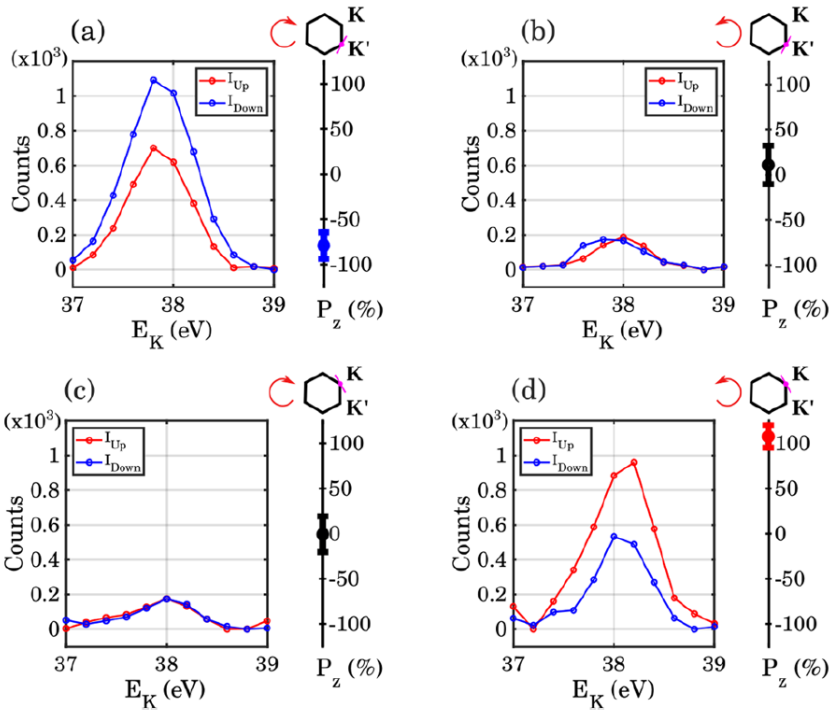
To achieve this, spectroscopic techniques capable of probing electronic, magnetic and lattice degrees of freedom on the ultrafast timescales (picosecond or femtosecond), both table-top and in large scale facilities, are currently being developed. In this talk, I will discuss the extension of Raman scattering to the ultrafast time scale as a symmetry resolved probe of non-equilibrium lattice and electronic properties of quantum materials. Quite remarkably this well-established technique to probe quantum materials in equilibrium has remained relatively underused on the ultrafast time scale. After a brief introduction to the technique, I will illustrate its use to the candidate excitonic insulator material Ta2NiSe5. I will show that a metastable phase with unique lattice and electronic properties can be induced in this material by a sub-picosecond near infrared pulse. I will conclude by giving some potential extension of the technique in terms of time and energy resolution, as well as coupling to more selective THz and Mid-IR pumping schemes.

Reference: K. Katsumi et al. Phys. Rev. Lett. 130, 106904 (2023)



Ultrafast Hidden Spin Polarization Dynamics of Bright and Dark Excitons in 2H-WSe_2
M. Fanciulli et al.

We performed spin-, time- and angle-resolved extreme ultraviolet photoemission spectroscopy of excitons prepared by photoexcitation of inversion-symmetric 2H-WSe_2 with circularly polarized light.
The very short probing depth of XUV photoemission permits selective measurement of photoelectrons originating from the top-most WSe_2 layer, allowing for direct measurement of hidden spin polarization of bright and momentum-forbidden dark excitons.
Our results reveal efficient chiroptical control of bright excitons' hidden spin polarization. Following optical photoexcitation, intervalley scattering between non-equivalent K-K' valleys leads to a decay of bright excitons' hidden spin polarization. Conversely, the ultrafast formation of momentum-forbidden dark excitons acts as a local spin polarization reservoir, which could be used for spin injection in van der Waals heterostructures involving multilayer transition metal dichalcogenides.



Valley- and helicity-resolved hidden spin polarization of bright excitons: (a)–(d) Spin-resolved energy distribution curves (I_{up} in red and I_{down} in blue) of photoelectrons ejected from bright excitonic states at both K' [(a),(b)] and K [(c),(d)] valleys and associated spin polarization (right subpanel), for both pump pulse helicities, at the pump-probe overlap. The pump helicities and valley indexes are depicted on top of each panel.

Out-of-equilibrium dynamics of the antiferromagnetic phase of $Bi_2Sr_2CaCu_2O_8$

Laurène Gatuingt[1], Alexandr Alekhin[1], Maximilien Cazayous [1], Alain Sacuto[1], Genda Gu[2], Sarah Houver[1], Yann Gallais[1]

[1]*Université Paris-Cité, Laboratoire Matériaux et Phénomènes Quantiques, CNRS (UMR 7162), 75013 Paris, France*

[2]*Matter Physics and Materials Science, Brookhaven National Laboratory, Upton, New York 11973, USA*

Ultrafast time-resolved Raman spectroscopy is applied to study the antiferromagnetic (AF) phase of the cuprate high-Tc superconductor Bi2212. The focus is on the evolution of Raman active magnetic and electronic excitations after an ultrafast 60fs near-infrared pump pulse. We observe a significant sub-picosecond decrease of the AF two-magnon excitation intensity, proving that the AF phase is altered by the pump pulse. Using Raman selection rules, we discuss the separate effects of the pump pulse on the AF order and on electronic carriers. Finally, we compare these effects with that of hole doping.

List of participants

- Aron Camille
- Banhart Florian
- Beaulieu Samuel
- Berthebaud David
- Braems-Abbaspour Isabelle
- Bresteau David
- Cailleau Herve
- Cario Laurent
- Cavarroc Marjorie
- Chernukha Yevheniia
- Cordiez Justine
- Corraze Benoît
- Dayen Jean-Francois
- Diener Pascale
- Duvail Jean-Luc
- Ettouri Rim
- Exil Gaston
- Gallais Yann
- Gatuingt Laurène
- Gautier Romain
- Geneaux Romain
- Guzmán Brambila Julio César
- Harkov Viktor
- Haydoura Mohamad
- Hussein Fatima

- Janod Etienne
- Julien Jean-Pierre
- Keller Niels
- Khelfa Abdelali
- Koussir Houda
- Kundys Bohdan
- Lautie Remy
- Le Tonquesse Sylvain
- Lepetit Marie-Bernadette
- Leroux Chris
- Maglione Mario
- Mauro Fanciulli
- Miura Akira
- Moog Luc
- Nathanaël Blond
- Pan Mingming
- Pastor Ernest
- Pasturel Mathieu
- Pawula Florent
- Perfetti Luca
- Querlioz Damien
- Rueff Jean-Pascal
- Sasaki Shunsuke
- Sen Raja
- Sidis Yvan
- Sopracase Rodolphe
- Stepanov Evgeny
- Subedi Alaska
- Ta Phuoc Vinh
- Tranchant Julien
- Wu Jiaming
- Zemal Aboubacar Nouridine

List of sponsors

CNRS Physique
Finance le GDR MEETICC

CNRS Chimie
Finance le GDR MEETICC

Institut des Matériaux de Nantes Jean Rouxel
Met à disposition son infrastructure et accueille la réunion thématique MEETICC

Amplitude
Fabricant de laser

Nantes Université
Financement de l'IRL DYNACOM

Région Pays de la Loire
Région, financement du projet Mott-IA

Author Index