PROGRAMME

4èmes Journées Plénières du GDR NEMO

NEW MOLECULAR ELECTRONICS

1 – 2 Décembre 2025

C2N, Palaiseau

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New Molecular Electronics

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IMCN, Université de Louvain
Belgique

Pr. Katharina Kaiser
ICASEC, University of Göttingen
Allemagne

Pr. Olivier Pluchery
INSP, Sorbonne Université
France

Pr. Anna Proust
IPCM, Sorbonne Université
France

Pr. Stéphane Rigaut
ISCR, Université de Rennes
France

Programme

Lundi 1 décembre 2025

8h30		**Accueil**
9h00		**<u>Invitée</u> : Anna Proust** *Polyoxometalates: multifunctional molecules for resistive switching materials and beyond*
9h40	Axe 3	**Eric Faulques** *Confinement-Driven Formation of Ising-Type Chains inside Single-Walled Carbon Nanotubes*
10h00		**Mukil Muppal** *Fe(II) Spincrossover Complexes on Surfaces: Towards Room-Temperature Switching*
10h20		**Communication Sponsor - Park Systems**
10h30		***Pause-café***
10h50		**<u>Invité</u> : Hervé Aubin**
11h30	Axe 4	**Melih Aktas** *The Effects of Low-dimensionality and Topologically Insulating Phases of π-conjugated Polymers on Electronic Transport*
11h50		**Poonam Kumari** *Modeling Spin-Orbitronics Effects at Interfaces and Chiral Molecules*
12h10		**Alexis Ophalvens** *Theoretical study of the influence of molecular parameters on the electronic properties of spinterfaces*
12h30		***Déjeuner***
14h00		**<u>Invitée</u> : Katharina Kaiser**
14h40	Axe 2	**Xiaonan Sun** *Molecular Diodes with Giant Rectification in Multi-Channel Nano-Porous Matrix: A Conductive-AFM Investigation*
15h00		**Mohamed Mamlouk** *Intelligent Molecular Switches for Brain-like Devices*
15h20		**Rui Li** *Counterion-Dependent Charge Transport in Ultrathin-film Molecular Junctions*
15h40		**Communication Sponsor - Nextron**
15h50		***Pause-café***
16h10		**<u>Invité</u> : Stéphane Rigaut**
16h50	Axe 3	**Rana Ayash** *Synthesis and optical properties of a water-soluble graphene quantum dots*
17h10		**Denis Frath** *Molecular switches based on boron complexes for themodulation of chiroptical properties*
17h30		**Session poster et industriels**
18h30		**Cocktail de bienvenue** *(Hall du C2N)*
21h00		**Fermeture**

Mardi 2 décembre 2025

09h00		**Invité : Olivier Pluchery** *Modifying the work function of gold nanoparticles with selected molecular monolayers*
9h40	**Axe 1**	**Abdourahim Hammani** *Synthesis and characterization of the switching properties of redox active hybrid polyoxometalates*
10h00		**Yan Wu** *Charge Transport and Electric-Field-Induced Non-Volatile Memory Effect in Viologen-based Single-Molecule Junctions*
10h20		**Communication Sponsor - Nanosurf**
10h30		***Pause-café***
10h50		**Invité : Pascal Gehring**
11h30		**Prix de thèse du GDR Némo 2025 : Abdoul Nasser Moussa** *Design and synthesis of 1) zigzag carbon nanobelts; 2) 1D and 2D Conjugated Polymers; and 3) Novel nanocarbons, termed calixacones.*
11h50	**Axe 3**	**Felix Houard** *The importance of the gaps: filling percolating networks of metal nanoparticles with redox molecules*
12h10		**Clôture – Pascal Martin**
12h20		***Déjeuner***

Industriel

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Since its founding in 2007, NEXTRON has progressed in its development of researcher-centered research equipment of the highest value. Recently, it grafted temperature/vacuum/measurement technology onto subminiature design technique to develop proprietary systems such as the Micro Vacuum Probe System and RTP system and its pushing towards its entry into the global market.

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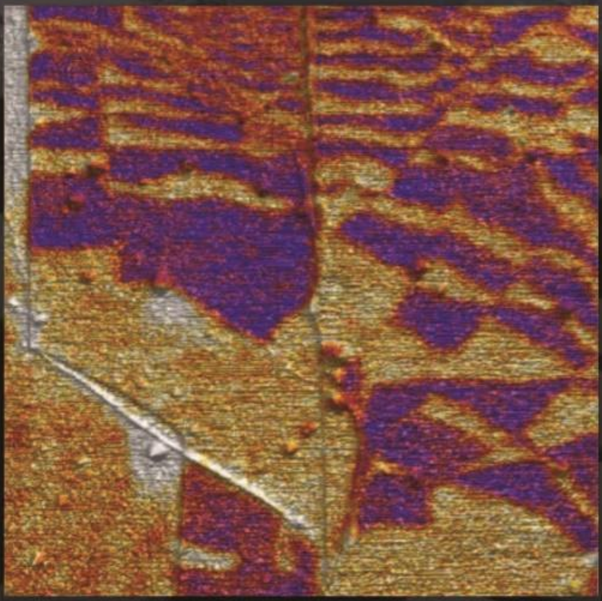
We sincerely ask for the continuous attention and love of many researchers as we proceed into the future. Thank you.



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Park AFM-IR Spectrometers

AFM-integrated nanoscale IR spectroscopy for advanced material characterization

Park AFM-IR Key Features

Suitable for Precise Chemical Characterization

- High Spatial Resolution Chemical Imaging
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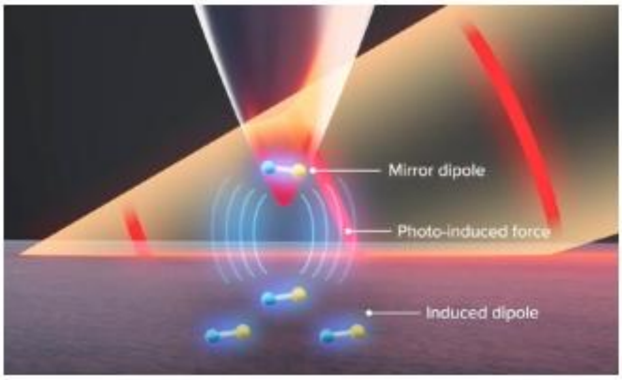
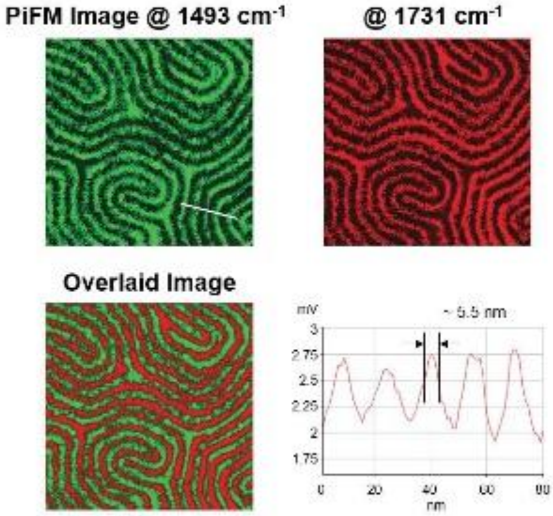
High Resolution Chemical Imaging with PiFM

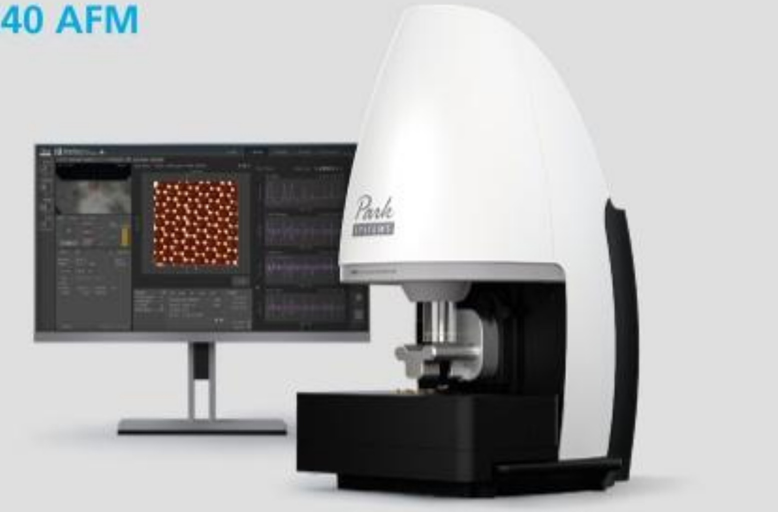
Photo-induced Force Microscopy (PiFM) measures photo-induced forces generated when a tunable IR laser excites molecular vibrations at their resonant frequencies. In Park AFM-IR systems, the AFM cantilever operates in Non-contact mode, detecting these forces without touching the sample. As the laser wavelength is scanned, changes in cantilever oscillation amplitude reveal local IR absorption, creating spectral and chemical maps. This fully non-invasive method provides true nanoscale chemical characterization—with sub-5 nm spatial resolution and monolayer sensitivity—allowing researchers to visualize molecular composition at near-atomic scale.





The measurement reveals clear phase-separated domains, where chemical contrast from PiFM highlights the distribution of the polymer blocks. This representative example demonstrates how PiFM provides both morphological and chemical information within a single nanoscale image.

FX40 AFM



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Liste des posters

Exploration of the Properties of Interfaces between Chiral molecules and metals

Yohan Duquerroy, Vincent Repain, Zakaria Ziani, Pascal Martin, Amandine Belleca

Importance of the epitaxial growth of molecular films on their spin-crossover properties

Amandine Bellec, T. Joshi, R. Chakraborty, Y. Garreau, M. Kelai, L. Kipgen, T. Mallah, M.-L Boillot, M.-A. Arrio, E. Otero, V. Repain, S. Narasimhan, A. Coatic

Self-assembled molecular networks for molecular rectifiers

Sélène Castanet, Quentin Fernez, Shiva Moradmand, Melina Vavali, Imad Arfaoui, Lydia Sosa-Vargasa

Study Of TEMPO Immobilization Techniques For The Exploration Of Molecular Spintronics

Milan Toledo-Nauto, Rui Li, Linsai Chen, Pascal Martin

Towards Electron-Responsive Supramolecular Hydrogels

G. Groslambert, V. Andrieux, L. Khrouz, J. Bauland, T. Gibaud, F. Chevallier, D. Frath and C. Bucher

Towards switchable magnetic tunnel junctions based on polyoxometalates monolayer

E. Aoustin, J.Lion, F.Godel, S.Collin, E.Carre, A.Vecchiola, A.Hammani, A. Proust, F.Volatron, M.B.Martin, F.Petroff, P.Seneor, R.Mattana

Rational Design of Aromatic Amines for the Controlled Growth of Covalent Organic Frameworks

Edward Dunn, Victor Vaillant, Maxime Berthe, David Guérin, Stéphane Lenfant, Eric Faulques, Nataliya Kalashnyk

Polyoxometalates: multifunctional molecules for resistive switching materials and beyond

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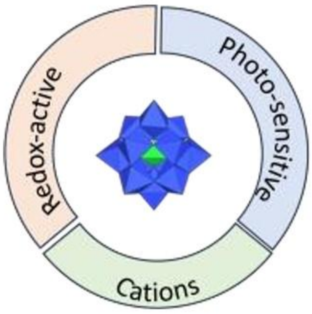
Abstract:

Molecules and hybrid molecular systems present a compelling opportunity to meet the growing need for tailored, multifunctional devices suited to mobile and diverse computing environments.[1] Molecules figure a unique set of atomic-level structural precision, mono-dispersity, high tunability and solution processability. Among molecules polyoxometalates (POMs), combine the redox properties and robustness of extended metal-oxides with the diversity of molecules. They are therefore natural candidates for the development of multi-sensitive molecular materials for neuromorphic computing.[2]

Based on a fundamental approach and through selected examples of charge transport measurements we will illustrate the POMs attributes and their response to an electrical or light stimulation.[3]



$[XM_{12}O_{40}]^{p-}$ *as a POM archetype and POM pivotal functionalities*

References:

(1) Chen, Y.; Han, B.; Gobbi, M.; Hou, L.; Samorì, P. Responsive Molecules for Organic Neuromorphic Devices: Harnessing Memory Diversification. *Advanced Materials* **2025**, *37* (19), 2418281. https://doi.org/10.1002/adma.202418281.
(2) Monakhov, K. Yu. Oxovanadium Electronics for In-Memory, Neuromorphic, and Quantum Computing Applications. *Mater. Horiz.* **2024**, *11* (8), 1838–1842. https://doi.org/10.1039/D3MH01926H.
(3) Volatron, F.; Izzet, G.; Vuillaume, D.; Proust, A. Unveiling Polyoxometalate Redox Properties at the Nanoscale. *Comptes Rendus. Chimie* **2024**, *27* (G1), 255–268. https://doi.org/10.5802/crchim.344.

Confinement-Driven Formation of Ising-Type Chains inside Single-Walled Carbon Nanotubes

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V.G. Ivanov,[d] J.L. Duvail,[e] N. Kalashnyk,[a] J. Sloan[b]

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Abstract:

This work highlights the power of confinement in directing the behavior of the red-emitting octahedral cluster compound $Cs_2Mo_6Br_{14}$ when encapsulated within single-walled carbon nanotubes (SWCNTs) of controlled diameter. We show that as the internal nanotube cavity progressively narrows from ~2.4 nm to ~1.1 nm, the structural limit imposed by steric confinement drives a phase transformation in the hybrid system. At larger diameters, the clusters pack without significant distortion, whereas near the smallest possible scale, where the van der Waals radius of the SWCNT approaches the external dimension of the $[Mo_6Br_{14}]^{2-}$ anion, confinement triggers a chemical reaction: elimination and polymerization of the precursors into extended $[Mo_2Br_6]_x$ nanoribbons that organize as one-dimensional Ising-type chains, marking a confinement-induced transformation from discrete clusters to reduced, extended nanostructures. High-resolution transmission electron microscopy and Raman spectroscopy capture this transition, while density functional theory calculations confirm the hierarchical relationship between nanotube diameter, packing mode, and emergent low-dimensional phases.

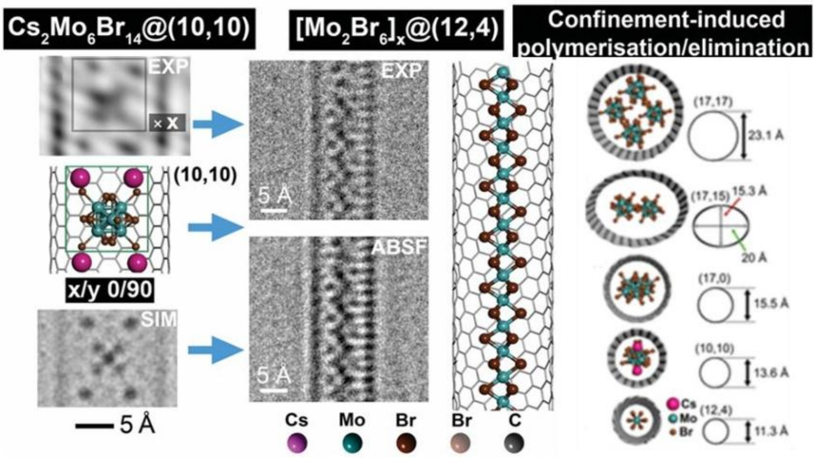

Figure : Left – HRTEM image of a single $Cs_2Mo_6Br_{14}$ cluster inside a (10,10) SWNT; Middle – HRTEM image of $(Mo_2Br_6)_x$ chains within a (12,7) SWNT; Right – confinement-induced formation of Ising-like chains upon reduction of the SWNT diameter.

Reference:

Faulques, E. et al. (2025), J. Amer. Chem. Soc., 147 (9), 7345-7359

Fe(II) Spincrossover Complexes on Surfaces: Towards Room-Temperature Switching

Mukil MUPPAL,[a] Adelais TRAPALI,[b] Vincent REPAIN,[a] Edwige OTERO,[c] Marie-Anne ARRIO,[b] Marie-Laure BOILLOT,[b] Talal MALLAH,[b] Amandine BELLEC[a]

[a]Université Paris Cité, CNRS, Laboratoire Matériaux et Phénomènes Quantiques, UMR6271, 75013, Paris, France. Email: mukil.muppal@u-paris.fr
[b]Institut de Chimie Moléculaire et des Matériaux d'Orsay, Université Paris-Saclay, CNRS, UMR 8182, 91405 Orsay 12 Cedex, France
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Abstract:

Spincrossover (SCO) compounds have been actively researched in the past decades due to their potential applications in molecular switches, memory devices etc. Molecular thin film- graphene hybrid devices utilizing the bistablilty of SCO molecules have been made in the recent past [1,2], opening new avenues to explore. This requires an in-depth understanding of the molecule-surface interface as it is well known that SCO properties are modified when these molecules are deposited on surfaces. In our study, spin state is efficiently probed using X-ray Absorption Spectroscopy (XAS), which is complemented by surfaces analysis carried out primarily by Scanning Tunneling Microscopy (STM) and Atomic Force Microscopy (AFM). We will focus on two SCO molecules, namely $Fe(II)[BH(MePz)_3]_2$ (FeMPz in short) and $Fe(II)[BH(Iz)_3]_2$ (FeIz). We studied the growth and SCO properties of few-nm thin FeMPz films deposited on monolayer and multilayer graphene substrates, which was previously done only on metal surfaces. The comparative study illustrates the effect of the substrate and film thickness on the SCO behavior. With the goal of developing SCO complexes with high switching temperatures, we explored the novel SCO FeIz deposited on Cu(111) and graphene. Here too we observe a strong influence of the substrate on the growth and SCO behavior of the molecule.

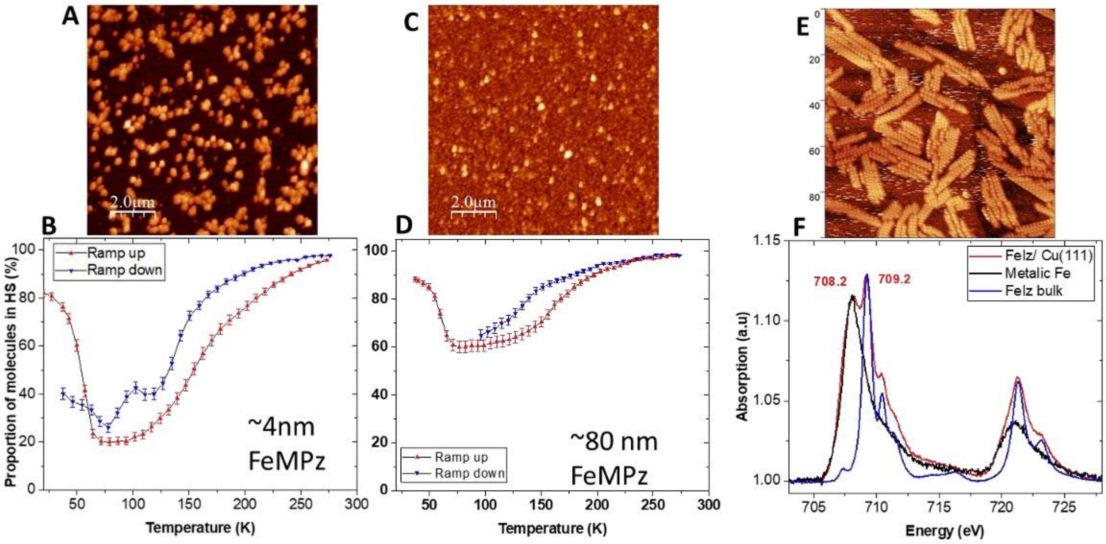

Figure : Fraction of HS molecules as a function of temperature for [B] 4nm and [D] 80nm of (SCO1) on graphene and their corresponding AFM images [A] and [C]. [E] Repersents the typical STM image for (SCO2) on Cu(111). [F] XAS spectra(red) on the sample shown in [E], superimposed with its components.

References:

[1] Gavaro-Edo et al., Adv. Mater., 34, 33, 2202551 (2022)

[2] Konstantinov et al., J. Mater. Chem. C 9, (2021)

The Effects of Low-dimensionality and Topologically Insulating Phases of π-conjugated Polymers on Electronic Transport

Melih Aktas,[a] Jêrôme Saint-Martin,[b] and Davide Romanin[a]
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Abstract:

One dimensional (1D) π-conjugated polymers are a promising class of cheap, flexible, non-toxic, organic materials with strong light absorption and chemically tunable electronic properties, which makes them ideal candidates for optoelectronic and energy-conversion applications[1]. Moreover, they can host topologically protected edge states without the need of spin-orbit coupling.

The Su-Schrieffer-Heeger (SSH) model is the paradigmatic example of a 1D topological insulator and was first introduced to describe the electronic structure of polyacetylene[2]. It was recently shown that single-[3, 4] (multi-[5])bridged 1D polyacene polymers can also host topological phases which can be identified with the SSH model.

In order to show the effects of low-dimensionality and topological phases on the electronic transport properties, we have studied the periodic trivially insulating bulk and single chain polyethylene (C_nH_{2n}), topologically insulating trans-polyacetylene (C_nH_n) and various configurations of polyacene polymers, which can be either trivially or topologically insulating, via the semi-classical Boltzmann theory with constant relaxation time approximation.

As a future direction, in order to understand the role topological boundary states play in electronic transport, we also want to consider the finite geometries by modeling the polymers as molecular junctions using *ab initio* methods and non-equilibrium Green functions (via the Fireball code[6]). As a matter of fact, it was recently shown that the coupling between the topological states at different boundaries can be leveraged as a gating mechanism, by locating the electrodes at the same boundary[7].

D.R. and M.A. acknowledge support from the HPC resources of IDRIS, CINES, and TGCC under Allocation No. 2024-A0160914101 made by GENCI, as well as funding from the project TyLDE (Grant No. ANR-23-CE50-0001) founded by the French National Research Agency.

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[1] H. Bronstein, C. B. Nielsen, B. C. Schroeder, I. McCulloch, "The role of chemical design in the performance of organic semiconductors", Nat. Rev. Chem. 4, 66, 2020.

[2] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Solitons in polyacetylene, Phys. Rev. Lett. 42, 1698 (1979).

[3] B. Cirera, A. Sánchez-Grande, B. dela Torre, et al., "Tailoring topological order and π-conjugation to engineer quasi-metallic polymers", Nature Nanotechnology 15:437-443, 2020.

[4] D. Romanin, M. Calandra, A. W. Chin, "Excitonic switching across a $\mathbb{Z}_2$ topological phase transition: From Mott-Wannier to Frenkel excitons in organic materials", Phys. Rev. B 106:155122, 2022.

[5] M. Aktas, M. Mosaferi, J. Saint-Martin, A. W. Chin, D. Romanin, "Multiple topological phase transitions in double-bridged polyacene polymers: Ab initio parametrized stacked Su-Schrieffer-Heeger model", Phys. Rev. B 112:045130, 2025.

[6] Lewis, J. P.; Jelínek, P.; Ortega, J.; Demkov, A. A.; Trabada, D. G.; Haycock, B.; Wang, H.; Adams, G.; Tomfohr, J. K.; Abad, E.; et al. Advances and Applications in the FIREBALL ab Initio Tight-Binding Molecular-Dynamics Formalism. Phys. Status Solidi B 248, 1989–2007, 2011.

[7] Liang Li, Shayan Louie, Nicholas M. Orchanian, Colin Nuckolls, and Latha Venkataraman. Long-range gating in single-molecule one-dimensional topological insulators. Journal of the American Chemical Society, 146(24):16920–16925, 2024.

Modeling Spin-Orbitronics Effects at Interfaces and Chiral Molecules

Poonam Kumari, Cyrille Barretaeu, Alexander Smogunov

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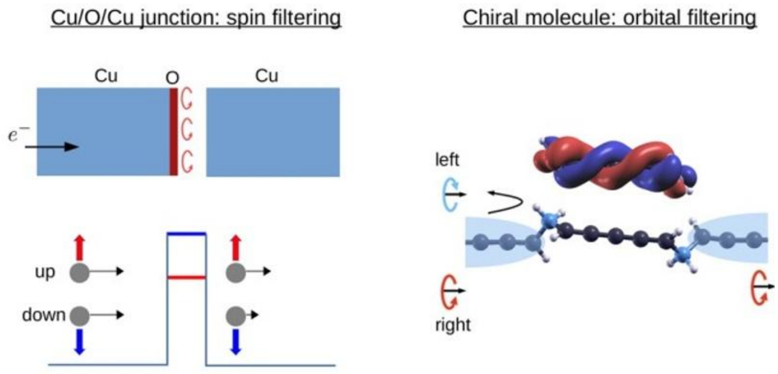
Email: poonam.kumari@ipht.fr

Abstract:

Spin-orbitronics, which is based on the use of orbital angular momentum as an information carrier, has gained significant attention in the recent past. In the structures which lack inversion symmetry, a formation of orbital angular momentum (OAM) in Bloch wave functions, independent of spin orbit coupling (SOC), has been predicted and is often considered to be a more generic effect implying that the orbital moment of an electron is the main driving force behind the Rashba effect rather than its spin [1]. Here, using the electronic wave packets approach, we explore the possibility of generation and propagation of orbital currents in two representative systems: an oxidized copper surface (where large OAMs are formed at the Cu/O interface) and a model carbon chain/chiral molecule junction. In the Cu/O system, the orbital polarization of incident wave packets is strongly enhanced at the Cu/O interface but rapidly decays in bulk copper. Interestingly, if a finite transmission across the oxygen layer is allowed (a tunnel junction), a significant spin-polarization of transmitted current is predicted; it persists at long distance and can be tuned by applied in-plane voltage. For molecular junctions, the mixing of carbon *px*, *py* channels by a chiral molecular orbital gives rise to efficient generation of orbital current and to its long-range propagation along the carbon chain.



Spin filtering through Cu/O/Cu junction and orbital filtering through chiral molecule.

References:

[1]. Seung Ryong Park, Choong H. Kim, Jaejun Yu, Jung Hoon Han, and Changyoung Kim.
Orbital-Angular Momentum Based Origin of Rashba-Type Surface Band Splitting. Phys. Rev. Lett.
107, 156803 (2011).
[2.]. Poonam Kumari, Cyrille Barreteau, and Alexander Smogunov. Modeling Spin-Orbitronics Effects at Interfaces and Chiral Molecules. Nano Lett. 25, 8869 (2025).

Theoretical study of the influence of molecular parameters on the electronic properties of spinterfaces

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Abstract:

Since the beginning of the 21st century, spintronics has been developed to conceive new devices for information storage by designing low energy consumption memories by controlling the current flowing between two ferromagnetic electrodes depending on the magnetization state of the electrodes, leading to the concept of magnetoresistance [1]. Like a lot of electronic devices, it is also of interest to incorporate organic semiconductors as active layers to replace the traditional inorganic materials. Doing so, it has been revealed at the beginning of 2010 that the study of the interface between a self-assembled monolayer (SAM) and a ferromagnetic electrode could lead to unexpected phenomena such as spin polarization of molecules. This led to the consideration of the interface as a new effective electrode, the so-called spinterface, to interpret the magnetoresistance in such systems [2].

To better understand the electronic characteristics of the spinterface, we have analyzed the influence of several molecular parameters by using a theoretical approach using the Density Functional Theory (DFT) with the Perdew, Burke and Ernzerhof (PBE) functional to describe exchange-correlation effects [3]. Our results point to the presence of hybridization-induced states situated within the HOMO-LUMO gap which are expected to control charge transport at low bias in molecular junction instead of the frontier molecular orbitals. Since these induced states are localized on the anchoring group of the molecules, the spin polarization of the spinterface appears to be independent of the size and shape of the molecules (Figure 1).

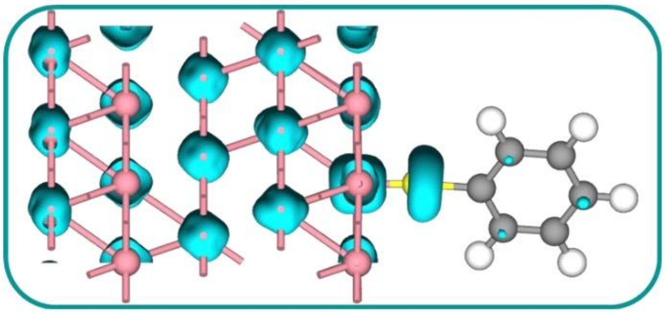

Figure 1: Density of states localized between the HOMO-LUMO gap of the system. We can observe a state localized on the anchoring atom, with no weight on the molecular backbone, which explains the absence of dependence of the molecule's spin polarization on its size and shape.

References:

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Molecular Diodes with Giant Rectification in Multi-Channel Nano-Porous Matrix: A Conductive-AFM Investigation

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Nanoscale molecular junctions (MJs) are formed using multi-channels of electroactive Co(II) ions vertically confined inside nanoporous SiO$_2$ matrix (diameter around 3nm) on a bottom ITO electrode.[1] The Platinum-coated conductive atomic force microscopy (C-AFM) tip functions as top electrode to complete the junction and the charge transport properties of the MJs are investigated. [2] The Pt-[Co(II)]-ITO Mjs have two main advantages. First, the size of the MJs is defined by the nanopores size in the scale of a few nanometers, with multi-channels, and are smaller than the size of the C-AFM tips (around 20nm). Second, the charge transfer direction is well aligned vertically thanks to the utilization of the SiO$_2$ matrix. We show that asymmetric I−Vs are repeatedly recorded where the current increases at forward bias. By changing the force applied bewteen the C-AFM tip and the Co(II)@SiO$_2$, giant RR raios are obtained exceeding 10^4 at a rather low voltage (<2.5V) which is the highest ratios ever achieved from C-AFM based nanoscale MJs. The Pt-[Co(II)]-ITO MJs can be seen as molecular rectificatifiers with giant rectification at room temperature. The mechanism of the current rectification is interpreted due to an electrostatic interaction between the top electrode and the electroactive Co(II) ions. Our results indicate that nanoscale molecular rectifiers, with controllable size of a few nanometers, can be successfully achieved using C-AFM.

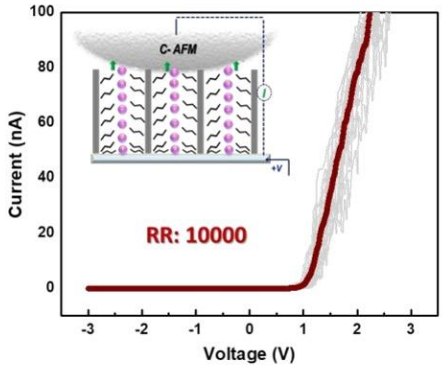

Figure : C-AFM built Pt-[Co(II)]-ITO MJs. Assymetric I/V curves are obtained with giant rectification ratio

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Intelligent Molecular Switches for Brain-like Devices

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Abstract:

Brain-inspired computing represents a paradigm shift toward low-power and adaptive information processing by mimicking the dynamic operation of biological synapses, where ionic fluxes and voltage-dependent neurotransmission regulate signal propagation (*Figure 1*). Emulating such complex behavior at the molecular scale has become feasible through dynamic molecular switches, which couple charge transport with redox and structural reconfiguration processes [1].

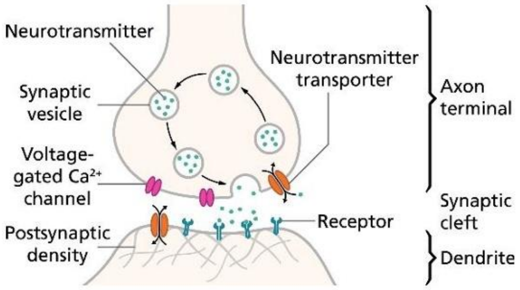

Figure 1: Schematic of a synapses

In artificial molecular synapses, signal modulation arises from voltage-driven protonation/ deprotonation and redox processes, analogous to the ionic fluxes that govern neurotransmission in biological systems. This coupling between charge transport and chemical reactivity produces hysteretic current–voltage characteristics and negative differential resistance (NDR), providing an electronic analogue of synaptic plasticity [1]. In this study, we investigate how different ionic species **(H^+, Na^+, K^+, etc.)** modulate the electronic response of molecular junctions. The interaction between these ions and the molecular backbone N-heterocyclic carbene/quinone can induce a mid-gap electronic state within the molecular energy gap, thereby enhancing the tunneling current, widening the hysteresis loop, and reinforcing the overall memory behavior, paving the way for brain-like molecular devices. [2]

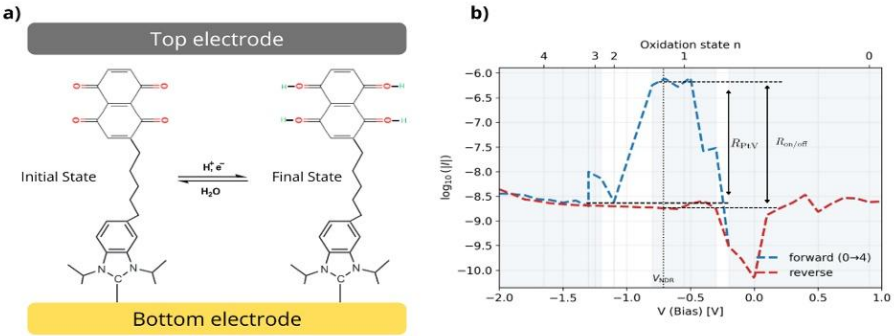

Figure 2: a) The junction consists of Au bottom electrode and an EGaIn. The two ET channels are bridged by slow PCET. b) Electrical characteristics of the Au-HNC-C10-Naphthalene-1,4,5,8-tetrone-Au junction

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Counterion-Dependent Charge Transport in Ultrathin-film Molecular Junctions

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Abstract:

Molecular Junctions (MJs), composed of an ultrathin molecular layer (<20nm) sandwiched between two electrodes, represent fundamental building blocks in nanoelectronics. They offer significant advantages over conventional silicon-based devices, including faster performance, higher packing density, and exceptional functional diversity. However, the influence of counterions within the molecular layer on charge transport remains poorly understood.

In this study, ultrathin films (2-10nm) of 4,4'-diamino-2,2'-stilbenedisulfonic acid (DAS) with different counterions (H^+, Li^+, Na^+, and K^+) were grafted onto gold strip electrodes via electrochemical reduction of in-situ generated diazonium salts in the corresponding supporting electrolytes[1]. Then, a Ti/Au top contact was deposited to complete a solid-state MJ[2].

The current-voltage (I-V) characteristic of these fabricated devices exhibits different conductive states (**Figure 1. b**, blue and yellow regions). Notably, the conductivity of the MJs incorporating Na^+ and K^+ counterions exceeds that of those containing H^+ and Li^+ by an order of magnitude. Combined with complementary analyses such as XPS and AFM, these findings underscore the crucial role of counterion size in governing charge transport within MJs.

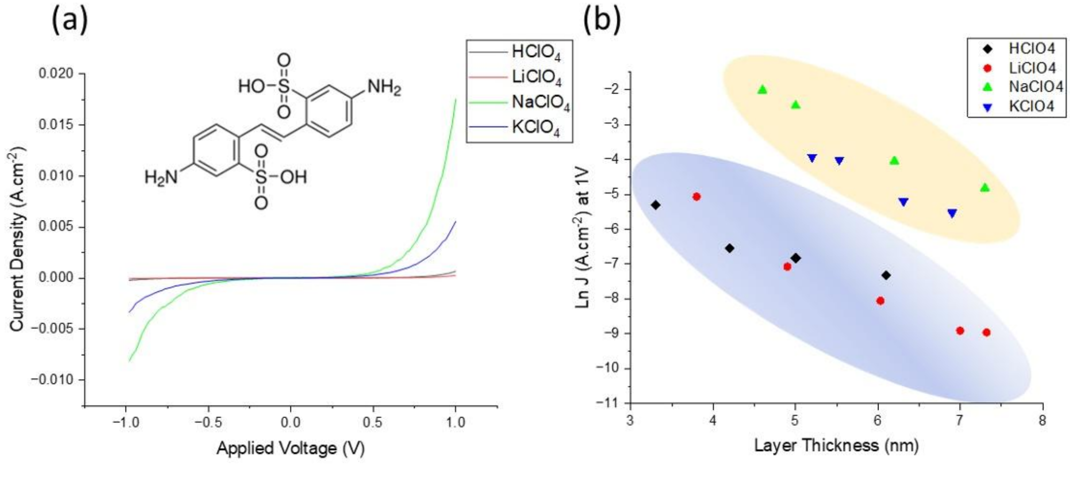

***Figure 1**. (a) Average current-density vs applied voltage (JV) curves of Au/DAS(X^+)/Ti/Au MJs, X=H, Li, Na, or K, with a similar DAS layer thickness of 6nm. Insert : chemical structure of DAS . (b) Ln J vs thickness plot taken at 1V.*

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SYNTHESIS AND OPTICAL PROPERTIES OF A WATER-SOLUBLE GRAPHENE QUANTUM DOTS FOR BIOLOGICAL APPLICATIONS

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Abstract:

GQDs synthesis has been mostly described *via* top-down methods (*i.e.* lithography, harsh oxidative cutting conditions or hydrothermal treatments).[1] Though cheap and efficient, these methods do not allow for precise control of the size, shape and edges of the GQDs. In order to truly control, with the required level of precision, the morphology and the composition of the materials and of its edges, the bottom-up approach is the relevant way to proceed.[2]

In a previous study, our team synthesized a family of graphene nanoparticles soluble in classical organic solvents. Thanks to their particular functionalization, these nanoparticles are individualized in solution and in the solid state. They emit photons in the red and near-infrared regions and exhibit exceptional photoluminescence quantum yields ranging from 90 to 95%, a level never before achieved for particles of such size. [3a] At the solid state, the GQDs also act as single photon emitters. [3b] To extend the potential of these materials, my thesis aims at improving the solubility of these nanoparticles in water by grafting hydrophilic polymers such as polyethylene glycol (PEG). To this end, we decided to functionalize the nanographene with triple bonds and to perform Click Chemistry (CuAAC). I began my thesis work by functionalizing C_{60} and C_{96} nanoparticles with polyethylene glycol (Figure 1), and thoroughly characterized the final products. This modification is crucial to make them soluble in water.

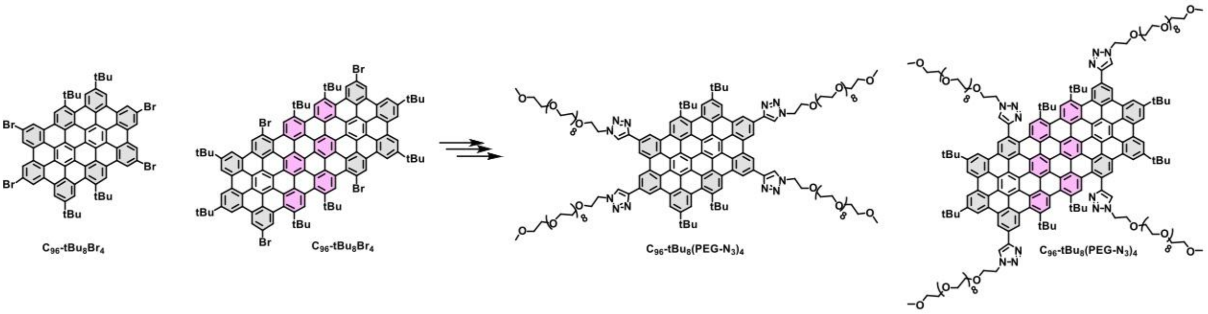

Figure 1: Schematic representation of PEG-functionalized graphene quantum dots.

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Molecular switches based on boron complexes for the modulation of chiroptical properties

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Abstract:

One great challenge nanoscience is facing is the difficulty to transpose molecular-scale phenomena into macroscopic properties finding application in everyday-life devices. A way to address this issue is to develop metamorphic molecular systems for which an external stimulus triggers a drastic structural reorganization. Based on this strategy, our project aims at bridging the gap between chiroptical properties observed in solution for Circularly Polarized Luminescence (CPL) switches[1] and their use in device-like systems.

We have recently described a very simple and efficient gram-scale synthetic pathway towards Boranils[2] substituted directly on the boron center by 1,1'-bi-2-naphtol derivatives (BINOL).[3] These chiral dyes typically feature high molar absorption coefficient in the range of 35,000 to 90,000 $M^{-1}.cm^{-1}$, absorption dissymmetry factor (g_{abs}) in the range of $2.5–5.5 \times 10^{-4}$ at the maximum wavelength of absorption of Boranils and CPL brightness up to $B_{CPL} = 5.4$ $M^{-1}.cm^{-1}$. Based on this new family of CPL emitters, we are developing stimuli-responsive CPL switches with metamorphic properties for on-surface application (see Figure 1). We have also developed molecular switches taking advantage of the control of energy transfer to a photochromic diarylethene core to modulate chiroptical properties.[4] This could have have interesting perspective given the known potential of this photo-switches for molecular electronics. [5,6]

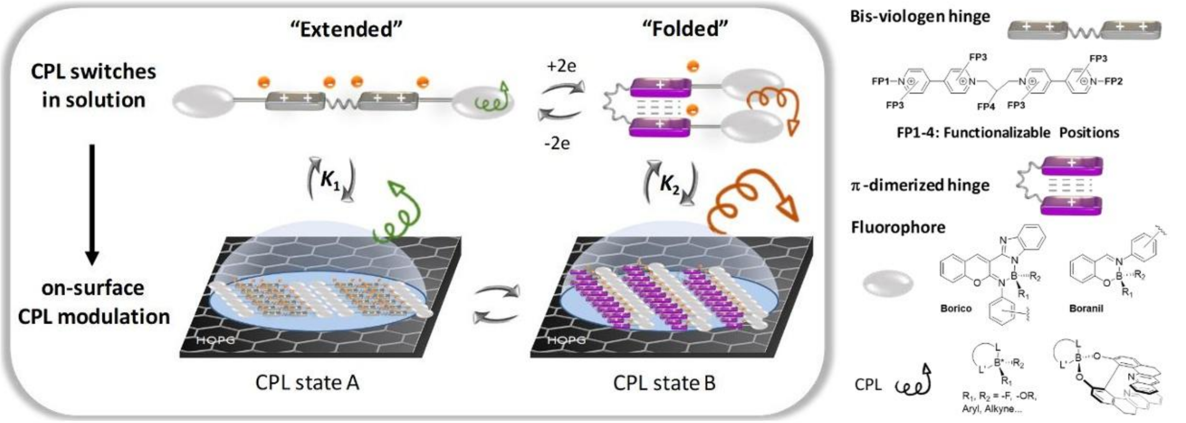

Figure 1. Schematic representation of metamorphic processes associated with modulation of chiroptical properties.

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Synthesis and characterization of the switching properties of redox active hybrid polyoxometalates

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Abstract:

Polyoxometalates (POMs), are nanometric oxoclusters composed of transition metals in their highest oxidation state with remarkable redox properties,[1,2] which make them promising for numerous applications, as for example in spintronics: integration of these molecules into magnetic tunnel junctions (MTJs) could open perspectives for next-generation spintronic devices[3]. Indeed the change of their redox state, induced by an electric field or optical irradiation, affect the interface ferromagnetic electrode-molecule, consequently the TMR (Tunnel magnetoresistance) measured in the device. This approach could pave the way for the elaboration of multifunctional spintronic devices with innovative in situ control of properties. In this presentation, we will describe the synthesis of a thiol terminated hybrid POM of the Keggin family $(TBA)_3[PW_{11}O_{40}(SiC_3H_6SH)_2]$ (TBA stands for tetrabutylammonium) as well as its analogue $(THEA)_3[PW_{11}O_{40}(SiC_3H_6SH)_2]$ (cf figure), which features a new ammonium counter-ion leading to a significant improvement in its photoreduction properties[4].

The characterization of their redox and photoredox properties in solution and at the solid sate will be presented. Additionally, we will expose how we control the organization of the hybrid POMs in monolayers on gold substrates and gain a deeper understanding of their behavior at the nanoscale, thanks to the use of advanced characterization like X-ray absorption spectroscopy.

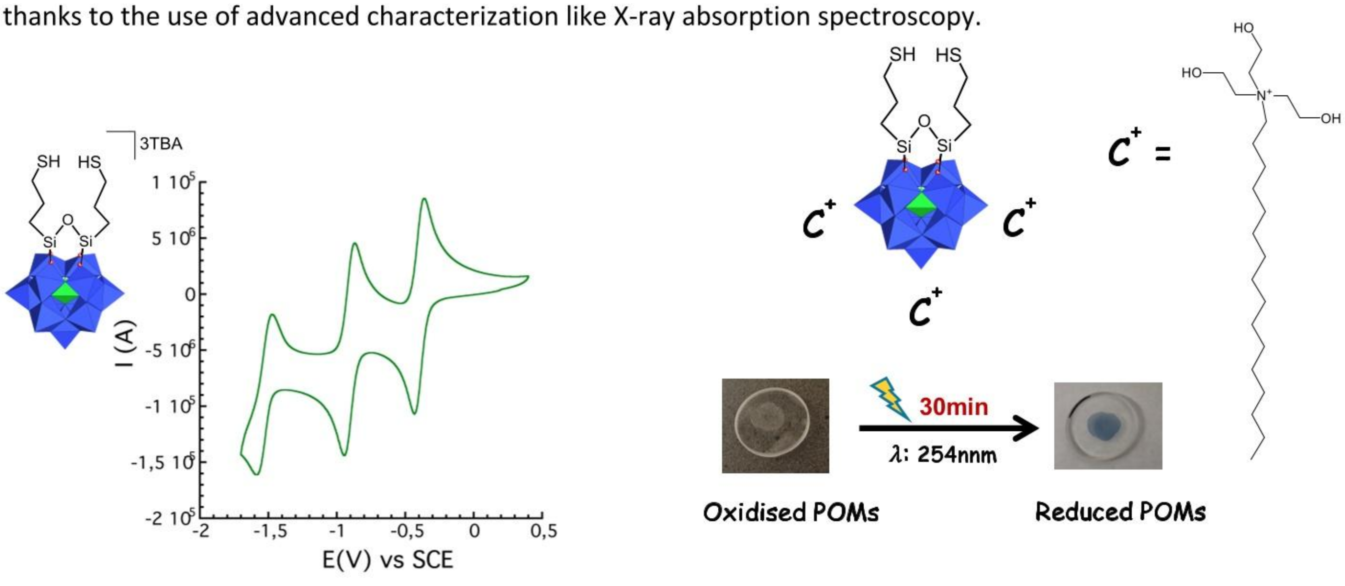

Figure : (left) scheme of the $(TBA)_3[PW_{11}O_{40}(SiC_3H_6SH)_2]$ and cyclic voltammetry recorded at WE: Glassy Carbon electrode, RE: Saturated Calomel Electrode, CE: Platinum electrode, 1mM in acetonitrile with $TBAPF_6$ 1M ($0.1\ V.s^{-1}$) (right) scheme of the $(THEA)_3[PW_{11}O_{40}(SiC_3H_6SH)_2]$ and photoreduction of a thick film by light irradiation at λ: 254nnm

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Charge Transport and Electric-Field-Induced Non-Volatile Memory Effect in Viologen-based Single-Molecule Junctions

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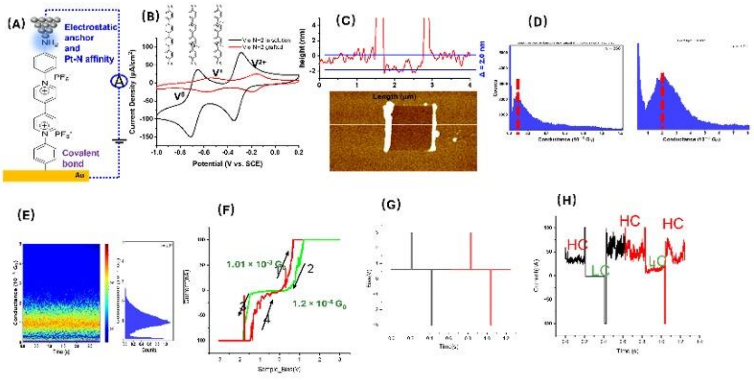
Abstract:

The miniaturization of electronic devices to the molecular level is a key goal of molecular electronics, with a strong focus on developing low-power, non-volatile memory and switching components.[1–3] In this work, we investigate the charge transport properties and conductive switching capabilities of a single-molecule junction (SMJ) based on the redox-active molecule methyl-viologen (Vio-NH_2).The Vio-NH_2 molecule was successfully grafted onto a gold (Au) substrate using electrochemical diazonium reduction method. An Au-[Vio-NH_2]-Pt SMJ was then constructed by approaching a platinum (Pt) tip to the molecule-modified Au substrate using the Scanning Tunneling Microscope Break Junction (STM-BJ) technique. Viologen is known to exhibit three reversible redox states (V_2+, V+, and V_0), allowing for multi-state investigation. STM-BJ measurements obtained from statistical conductance histograms revealed two distinct, stable conductance values: a Low Conductance (LC) state at ≈ 1 x $10^{-4}G_0$ and a High Conductance (HC) state at ≈ 1 x $10^{-3}G_0$. These values are consistently observed in the conductance traces versus time (Figure E). Most importantly, the I(V) curves (Figure F) with clear hysteresis loop show clear electric-field-induced conductance switching between the LC and HC states, demonstrating typical memory behavior. The calculated ON/OFF ratio is approximately 1 x$10^{-3}G_0$/1.2 x10^{-4}G≈8.4 at 0.5v. The I(t)trace (Figure G) practiced reading writing erasing process. In conclusion, the Au-[Vio-NH_2]-Pt SMJ successfully exhibits an electric-field-induced nonvolatile memory effect, showcasing its strong potential for applications in molecular storage devices.



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The importance of the gaps: filling percolating networks of metal nanoparticles with redox molecules

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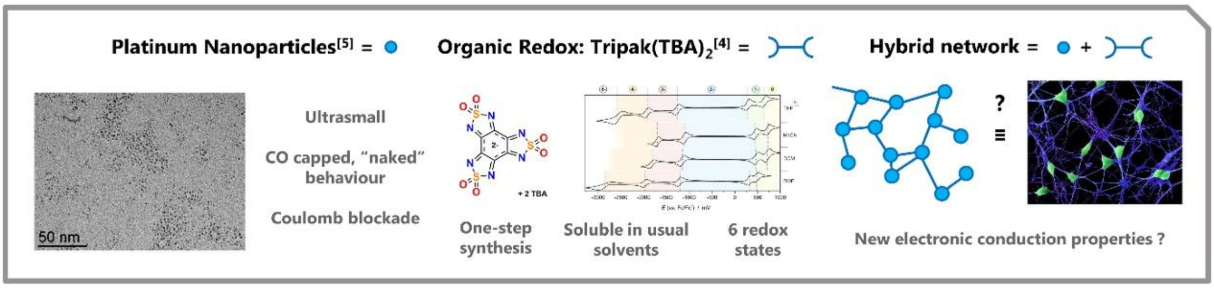
Abstract:

The field of artificial intelligence is fueled by the development of dedicated deep learning methods and infrastructures, which enabled the emergence of tools accessible to a broader audience. However, certain physical limitations in the design of our conventional computers (Von Neumann bottleneck) and of the hardware (chip miniaturization) can compromise their adoption. This accentuate the need for alternative computational and hardware approaches, such as neuromorphic systems, which offer to overcome these obstacles by mimicking the efficiency and parallelism of the nervous system.[1]

In this context, we adopted a bottom-up approach to develop hybrid materials combining metal percolating networks that can exhibit brain like properties,[2] and functional molecules as junctions. These networks are made of platinum nanoparticles forming complex conduction pathways, and by integrating functional molecules into these networks, we aim to reproduce synaptic functions for advanced electronics properties.[3]

In this study, we investigate the impact of the implementation of an organic molecule featuring six oxidation states, a pseudo-[6]oxocarbon named Tripak,[4] in self-assembled platinum nanoparticles networks.[5] These self-assembly structures are characterized through chemical, structural and electrical techniques at the nano- and microscale.



Schematic representation of the strategy adopted in this presentation

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Exploration of the Properties of Interfaces between Chiral molecules and metals

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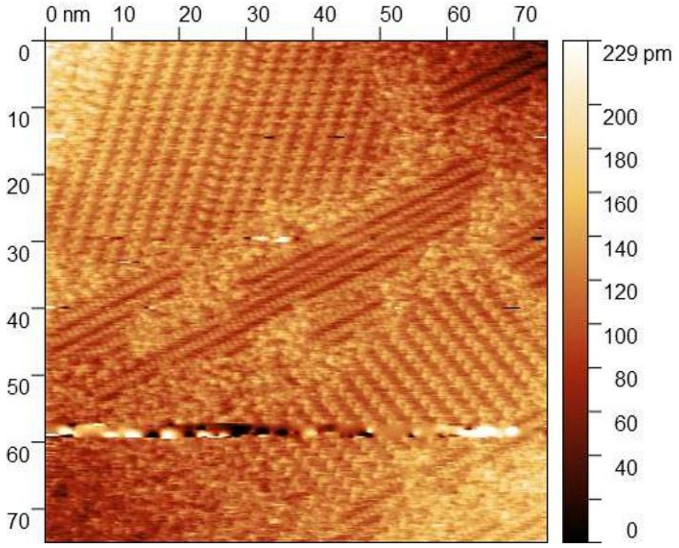
Abstract:

The Chirality Induced Spin Selectivity (CISS) effect refers to the electron spin filtering observed after passing through a chiral material, such as molecular monolayers. It has the potential for a wide range of applications, from spintronics [1] to enantioselectivity in electrocatalysis [2]. However, as the theoretical understanding of the CISS effect remains debated [3], it is essential to develop well-controlled model systems that allow its precise manipulation and study. To this end, studying the CISS effect in monolayers of chiral molecules such as 1,1'-binaphthalene-2,2'-diol (BINOL) can prove especially insightful as they could be studied both experimentally and via simulations. Thus, we prepared molecular films of S-BINOL molecules on Cu(111) by physical evaporation in ultra high vacuum (UHV) conditions, followed by an annealing. Using scanning tunnelling microscopy (STM) at room temperature in UHV we show that S-BINOL molecules form ordered domains which depend on the annealing temperature. These results show that an organised layer of S-BINOL molecules can be obtained by physical evaporation. This will allow to study the CISS effect on a controlled model interface which can be customised using modified BINOL-based molecules and various substrates.



STM image of a film of S-BINOL molecules on a Cu(111) substrate showing organised domains (75x75 nm², 5 pA, 1.5 V)

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Importance of the epitaxial growth of molecular films on their spin-crossover properties

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Abstract:

Spin-crossover (SCO) molecules present two spin states that can be controlled by external stimuli such as light or temperature. Their ability to switch makes them promising candidates for incorporation in molecular spintronic devices. In this purpose, it is mandatory to understand how the properties of spin crossover molecules are modified when in direct contact with metallic substrates and how the growth of molecular films is governed by the substrate. In previous studies on Au(111) and Cu(111) substrates, we evidenced that an epitaxial relationship between the molecular layer and the substrates exists [1], which has a direct consequence on the conversion from one spin-state to the other either by using temperature [2,3], light [4,5] or electric field [6] as external stimuli.

Here, we investigate in detailed the molecular structure of $Fe[HB(3,5-(CH_3)_2Pz)_3]_2$ (Pz = pyrazolyl, called FeMPz hereafter) ultra-thin films adsorbed on Cu(110) using grazing incidence x-ray diffraction measurements, along with their spin-crossover properties measured by x-ray absorption spectroscopy (XAS). For sub-monolayer coverage, the molecules self-assemble in two equivalent domains that are in epitaxial relationship with the Cu(110) substrate, while the molecules are locked in HS state at low temperature. For larger thicknesses, we evidenced a layer-by-layer growth along the (100) direction of the molecular bulk crystal. The epitaxial constrain is also released and the transition properties are recovered.

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Poster

Self-assembled molecular networks for molecular rectifiers

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One of the key challenges in organic electronics is controlling current flow at the molecular level. Molecular rectifiers are necessary components to achieve this. One way to obtain efficient molecular rectifiers is using a bottom-up approach via self-assembled monolayers (SAMs) [1]. However, traditional SAMs face limitations in reproducibility, molecular organization, and tunability of donor-acceptor properties, which hinder their performance and integration into next-generation electronics [2].

To overcome these limitations, we have recently developed supramolecular assembled dyads capable of forming ordered assemblies on graphene-like substrates to obtain novel electronics properties. Held together by non-covalent interactions, these dyads enable facile mixing and matching of components, resulting in a highly tunable system [3].

In this work we will present the synthesis and characterization of these systems in solution and on a graphene substrate.

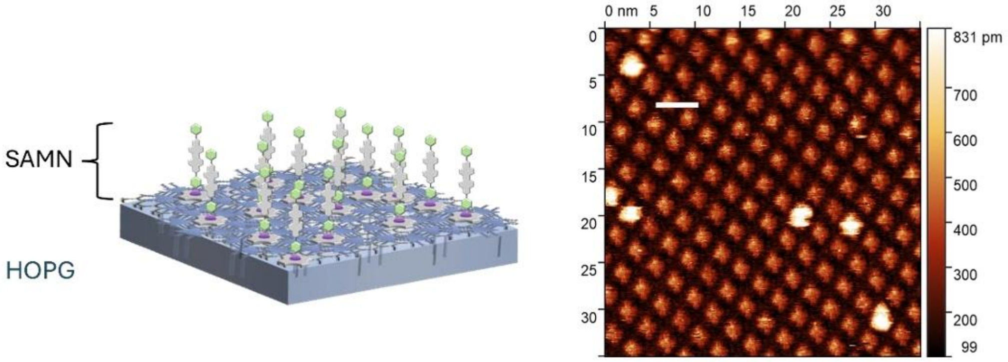

Figure 1 : Schematic representation of a self-assembled molecular network (left), STM image of a C12ZnPc-PDI dyad on graphene substrate (right)

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Study Of TEMPO Immobilization Techniques For The Exploration Of Molecular Spintronics

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Abstract:

Excited magnetic spin states are highly sought after in molecular spintronics due to their potential use in quantum device applications. However, precise control of the spin transfer interface at the electrode/molecule surface (spinterface) remains a challenge[1], as it is a key step that affects the stability, sensitivity, and performance of devices.

This requires exploring techniques that allow the control of spinterface and promote polarization and spin conservation during transport. One of the versatile and little-explored techniques in spintronics is electrografting with diazonium salts[2], which generates robust covalent bonds, improving thermal and chemical stability, reducing contact resistance, and possibly favoring spin transport.

Here we present our recent results on the immobilization of 4 amino-2,2,6,6-Tetramethylpiperidine-1-oxyl (TEMPO), a radical with a free electron, achieved through a strategy that combines two fundamental reaction processes: Electrochemical reaction (the laboratory's expert methodology in diazonium electro-grafting) and chemical reaction[3]. This strategy consists of electro-grafting 4-aminobenzoic Acid (ABA) by electrochemistry and subsequent chemical reaction with EDC/NHS to activate the carbonyl group of ABA, followed by immobilization of TEMPO (see Fig. 1).

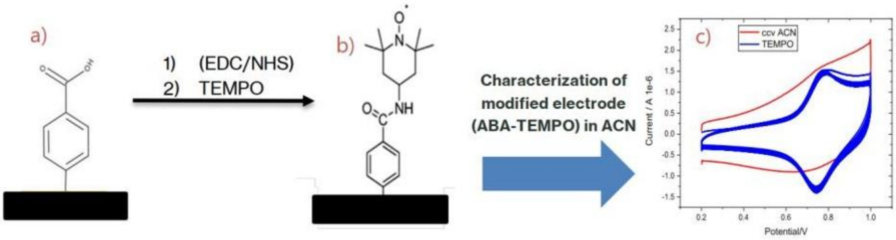

Fig.1 *Strategy to immobilize TEMPO: (a) ABA grafted onto the electrode. (b) Immobilization of TEMPO by chemistry reaction. (c) CV reveals the peaks of TEMPO Oxide-reduction immobile on the electrode.*

Routine CV, AFM, and XPS characterizations showed successful immobilization of TEMPO. To date, we are conducting preliminary studies of TEMPO electron transport at low temperatures. Next, this protocol will be transferred to magnetic electrodes (Co/gr) to fabricate spin valve device and measure the filtering efficiency.

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Poster

Towards Electron-Responsive Supramolecular Hydrogels

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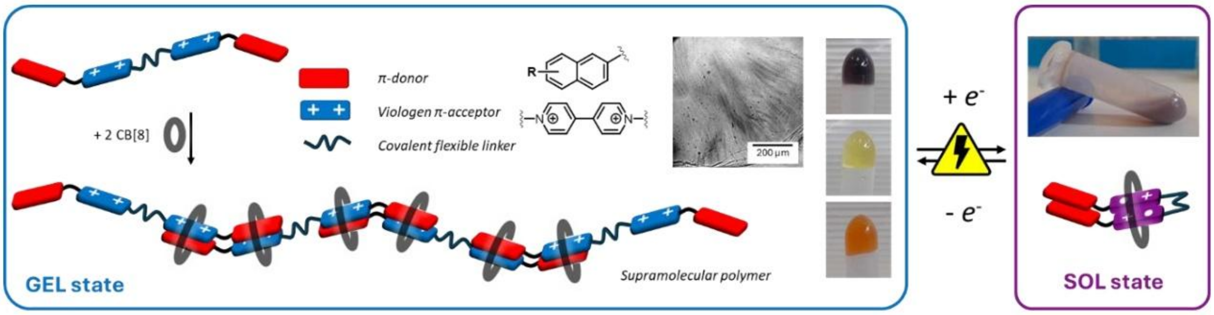
Abstract:

Interest in stimuli-responsive soft materials has grown over the last decade. Hydrogels, in particular, have been the subject of extensive research because of their many potential applications.[1] Gels can be defined as solvent trapped in a network that typically contribute little to the mass. One key feature of supramolecular gels, involving a network of non-covalent bonds, is their ability to undergo self-healing processes or to respond to external stimuli. (pH, T, P, light). These soft, dynamic and responsive supramolecular materials have already proved useful in many fields, from catalysis to electronics.[2]

Our project aims at addressing two challenges in the field of responsive hydrogels linked to the following observations: 1) Most stimuli-responsive behaviors reported so far come from serendipitous finding due to a lack of control and anticipation over the molecular events involved in the gelation process; 2) Responsiveness to electrical stimuli remains almost unexplored, despite being essential to ensure implementation of such materials in devices.

Over the last few years, our group has developed major expertise in the characterization and engineering of electro-switchable gels.[3-6] This poster will focus on supramolecular gels formed by the spontaneous self-assembly of cucurbit[8]urils host with π-conjugated donors and electron-responsive bipyridinium acceptors.[7] The mechanisms of formation and response to stimulation will be discussed on the basis of spectroscopic, rheological, microscopic and electrochemical data of the hydrogels formed.



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Towards switchable magnetic tunnel junctions based on polyoxometalates monolayer

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Abstract:

Spintronics has revolutionized modern electronics by introducing new possibilities for data storage and magnetic sensing technologies. As its core, are magnetic tunnel junctions (MTJs) consisting of two ferromagnetic electrodes separated by an insulating barrier. It has been shown that molecules could be integrated into MTJs giving rise to molecular spintronics that combines the principles of spintronics with the properties of organic molecules. Thanks to the spin-dependent hybridization at ferromagnetic electrode/molecule interfaces, spin polarization and thus tunnel magnetoresistance can be tailored by molecules (1). Up to now, mainly passive molecules such as alkane chain have been integrated into MTJs (2-3).

In this presentation, we will present molecular MTJs integrating molecules, called “active” molecules, which can be switched by an external stimulus (light, pressure, electric field…). The energy gap and coupling strength to the electrodes depending on the molecule state, its modification can hence lead to modulation of the spin polarization of the interface. Consequently, MTJs properties can be expected to be tuned by switching the molecule. Among the large variety of these “active” molecules (4), electrically addressable molecules are particularly interesting for future applications. Here, we focus on polyoxotungstate (PW11O40(SiC3H6SH)2) redox switchable molecules, from the polyoxometalate (POM) family. We will first present the multiple challenges we have faced to fabricate NiFe/POMs/Co MTJs (see Fig.1). Next, we will show the characterizations of the interface which confirm the grafting of molecules onto the electrodes. We will then focus on the investigation of the electrical switching of the molecules by conductive-tip AFM. Finally, we will present the preliminary electrical characterization of the NiFe/POMs/Co MTJs which demonstrate that switchable polyoxometalate monolayer can be successfully integrated in MTJs. This opens the way to develop multifunctional spintronic devices.

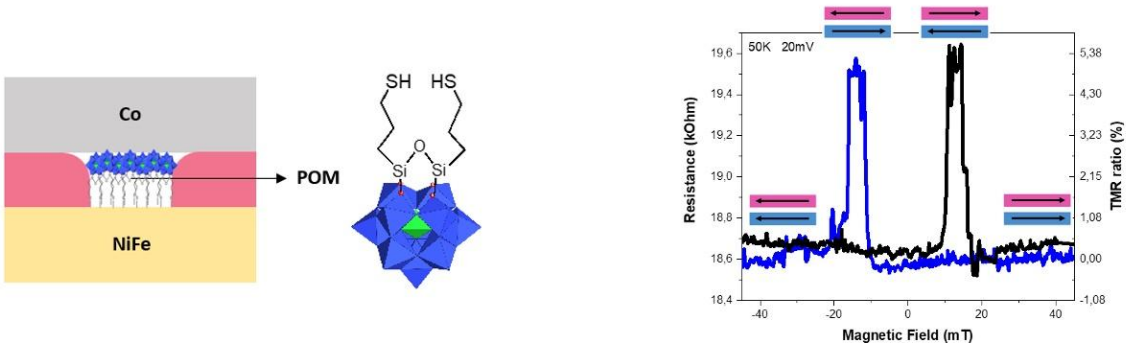

Figure 1: (Left) MTJ integrating POM molecules. (Right) TMR signal measured in a NiFe/POM/Co junction.

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Rational Design of Aromatic Amines for the Controlled Growth of Covalent Organic Frameworks

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Abstract:

Polymers derived from aromatic amines act as effective hole transport layers in a variety of applications. Triphenylamine (TPA) is perhaps the simplest of the aromatic amines that consists of 3 phenyl rings bound to a nitrogen atom to form a propeller shape. TPAs are also frequently used as precursors for the creation of polymers via wet-chemistry approaches [1, 2], but a study of their on-surface reaction to ensure the controlled growth of two-dimensional covalent organic frameworks (COFs) has, to our knowledge, not yet been reported. The molecule's 3-fold symmetry and aromaticity indicate that it should be possible to synthesize COFs similar to those created from 1,3,5-triphenylbenzene derivatives [3].

In this study, we observe the on-surface synthesis of TPA polymers on metal coinage surfaces with scanning tunnelling microscopy (STM) under ultra-high vacuum (UHV) conditions. Bromination enables the formation of polymers through the Ullman coupling reaction, whereas ethynyl-functionalisation proves more complicated as there are many possible reactions including the Glaser coupling and cyclotrimerization. Substrate selection also plays a key role as the Au(111) appears to spatially constrain the reaction of ethynyl functionalised TPAs during deposition onto a substrate below 100 °C.

We also identify the influence of functionalisation on the IR, Raman and absorption spectra of TPA and its derivatives via a combined experimental and theoretical study. Our analysis reveals that functionalisation affects the entire molecular structure, manifesting not only in the characteristic vibrational modes of the functional groups but also in the oscillatory behaviour of the phenyl rings and the central nitrogen core.

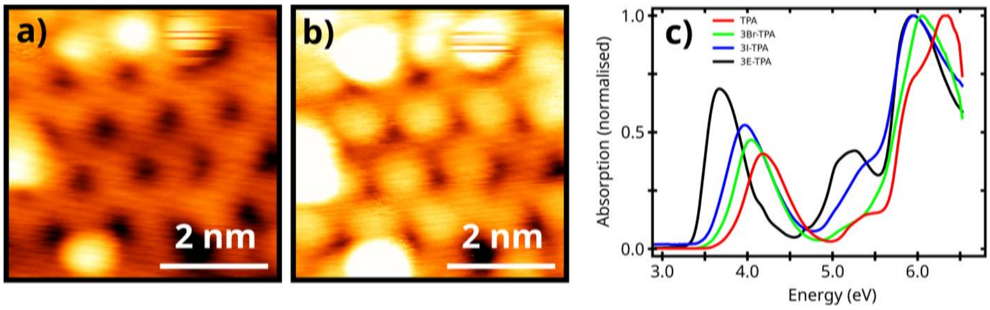

Figure 1: (a,b) STM images of covalent organic frameworks formed from tris(4-ethynylphenyl)amine on Au (111): (a) at positive sample biases a honeycomb lattice can be observed in STM images, whereas (b) the same region at negative bias shows protrusions at every other site. Functionalisation creates pronounced changes in the ultra violet and visible absorption spectrum of TPA when functionalised with bromine (3Br-TPA), iodine (3I-TPA) and ethynyl groups (3E-TPA). Imaging parameters: (a) I=20 pA, V=+2 V (b) I=20 pA, V = -2 V.

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