O 61: Organic molecules on inorganic substrates: electronic, optical and other properties II

Time: Wednesday 15:00–17:45 Location: HSZ/0201

O 61.1 Wed 15:00 HSZ/0201

Circular dichroism in the photoelectron angular distribution of achiral molecules — Christian Simon Kern[1], Xiaosheng Yang[2], Giovanni Zamborlini[1,3], Simone Mearini[3], Matteo Jugovac[3], Vitaliy Feyer[3], Umberto De Giovannini[4], Angel Rubio[4], Serguei Soubatch[2], Michael G. Ramsey[1], •Frank Stefan Tautz[2], and Peter Puschnig[1] — [1]Institute of Physics, University of Graz, Austria — [2]PGI-3, FZ Jülich & RWTH Aachen University, Germany — [3]PGI-6, FZ Jülich & Universität Duisburg-Essen, Germany — [4]MPI for the Structure and Dynamics of Matter, Hamburg, Germany

Circular dichroism in the angular distribution (CDAD) is the effect that the angular intensity distribution of photoemitted electrons depends on the handedness of the incident circularly polarized light. CDAD has been reported also for achiral organic molecules at the interface to metallic substrates. For this latter case, we investigate two prototypical pi-conjugated molecules, namely tetracene and pentacene, whose frontier orbitals have a similar shape but exhibit distinctly different symmetries. By comparing experimental CDAD momentum maps with simulations within time-dependent density functional theory, we show how the final state of the photoelectron must be regarded as the source of the CDAD in such otherwise achiral systems. We gain additional insight into the mechanism by employing a simple scattering model for the final state, which allows us to decompose the CDAD signal into partial wave contributions.

O 61.2 Wed 15:15 HSZ/0201

Many-body electronic structure of acenes on an MoS2 monolayer — •Jan-Phillip Topmöller and Michael Rohlfing — Institute of Solid State Theory, University of Münster, Germany

TMDCs show great potential in terms of their applicability in optical electronics. Due to their large surface, molecules can easily bind to the TMDC and affect its electronic properties. Here we investigate acenes as a prototypical class of molecule.

We use DFT (GGA) and DFT-D3 to determine the most stable adsorption position of acenes like benzene, naphthalene and anthracene on MoS2. Subsequently, we use many body perturbation theory to calculate the electronic structure of the adsorbate system and its individual components (MoS2 and the molecule) in order to investigate the effect of the molecule on the MoS2 electronic and optical spectrum.

O 61.3 Wed 15:30 HSZ/0201

Azimuthal-Scanning SFG Microscopy for Mapping Packing Motifs and In-Plane Disorder in Molecular Films — •Nasim Mirzajani, Ben John, Alex Fellows, and Martin Thaemer — Fritz-Haber-Institut, Berlin, Germany

Phase-resolved vibrational sum-frequency generation (SFG) microscopy is a powerful tool for probing anisotropic and heterogeneous structure of molecular films. As a second order nonlinear technique, SFG is uniquely sensitive to disorder in film morphology but it has not been yet used to quantify order in molecular film assemblies. Here, we introduce an azimuthal-scanning SFG microscopy approach that extracts detailed information on structure as well as in-plane orientational order by analyzing the full set of rotational frequencies arising from azimuthal dependence. Applying the method to a lipid monolayer, we determine the full 3D structure of the lipids, the packing motif as well as the degree of its in-plane disorder. This technique provides a general framework for probing packing structure in thin films and is broadly applicable to diverse molecular and phononic materials.

O 61.4 Wed 15:45 HSZ/0201

Synthesis and Characterization of a Polyradical Hexaazacycloarene — •Zilin Ruan, Olaf A. Kleykamp, Tim Naumann, Faming Kang, Jörg Sundermeyer, and J. Michael Gottfried — Department of Chemistry, Philipps University Marburg, 35037 Marburg, Germany

Cycloarenes have drawn considerable research interest owing to their unique electronic and topological features. The synthesis of large zigzag-edged cycloarenes, which are predicted to exhibit exotic polyradical ground states, has remained elusive. Here, we report the synthesis of a zigzag-edged hexaazacycloarene incorporating six graphitic nitrogen atoms within its macrocyclic backbone, achieved through a combined in-solution cyclization and on-surface cyclodehydrogenation on a Au(111) surface. The structural and electronic properties of the resulting macrocycle were investigated by STM/STS, nc-AFM and KPFM. DFT and multireference CASSCF calculations reveal that the isolated molecule has an open-shell singlet ground state with six unpaired electrons and undergoes a spontaneous symmetry breaking from C6 to C2, driven by strong electron-electron correlations and a pseudo-Jahn-Teller instability. Notably, the expected low-energy spin excitations are absent on Au(111), which is rationalized by the hybridization between the molecular π-system and the substrate, effectively quenching the intrinsic magnetism. These results provide crucial insight into correlation-driven magnetism and represent a significant advance toward the realization of cycloarenes with π-magnetism.

O 61.5 Wed 16:00 HSZ/0201

Vibrational Spectra of Materials and Molecules with Path Integrals and Machine Learning — •Jorge Castro, Krystof Brezina, and Mariana Rossi — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Vibrational spectroscopy provides fundamental insight into molecular structure and dynamics. Nuclear quantum effects (NQEs) can critically modulate vibrational peak positions, shapes, and intensities. Recently, we showed that the partially adiabatic elevated-temperature centroid molecular dynamics (PA-T_e-CMD) [1] accurately reproduces vibrational spectra across molecular and condensed-phase systems, including low-temperature regimes where NQEs are strongest. This approach enables the simulation of quantum infrared and Raman responses in diverse spectroscopic settings. Tip-Enhanced Raman Spectroscopy (TERS) [2] achieves sub-nanometer spatial resolution and is uniquely sensitive to single-molecule vibrational motion and surface defects. Building on first-principles finite-field formulations of TERS [3], we combine this framework with PA-T_e-CMD to quantify the role of nuclear quantum motion and anharmonicity in near-field Raman observables. As a first application, we investigate hydrogen-bonded chains of 2,5-diamino-1,4-benzoquinonediimine (DABQDI) and discuss how NQEs imprint on vibrational signatures in simulated TERS images.

[1] J. Castro et al., J. Chem. Phys. 163, 204102 (2025). [2] J. Lee et al., Nature 568, 78*82 (2019). [3] K. Brezina et al., arXiv:2509.13075 (2025).

O 61.6 Wed 16:15 HSZ/0201

Quasiparticle level alignment in anthracene-MoS_2 heterostructures — •Hsin-Mei Ho, Michael Lorke, and Peter Kratzer — Faculty of Physics, University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

Stacking atomically thin materials through van der Waals (vdW) interaction has opened a new paradigm for fundamental and applied sciences. While preserving the advantages of individual components, heterostructures are predicted to exhibit emergent properties at the interfaces. In the presence of external perturbations, since the behavior of a hybrid system is strongly related to the electronic band structure, it is of interest to find a methodology beyond the ground-state formalism of density functional theory (DFT). In this work, we begin with DFT calculations and consider the organic-inorganic heterostructures composed of anthracene molecules on monolayer MoS_2. Motivated by studies on the crystallinity of organic films, interfacial configurations with different coverage and orientation of anthracene are investigated. We find that both the bandgap values and band offsets are sensitive to whether the molecules are closely packed and whether they adopt perpendicular or parallel orientations on MoS_2. Whereas DFT predicts type-II level alignment, inclusion of quasi-particle corrections via the GW approximation yields type-I alignment. These findings highlight the importance of level-alignment predictions for interpreting experiments and for guiding the design of organic-TMDs heterostructures.

O 61.7 Wed 16:30 HSZ/0201

Magnetic interactions between spins in the Metallacrown CuCu4: bulk vs adsorbed molecule on Au(111) — •Ariyan Tavakoli[1], Stefan Lach[1], Frederik Pütz[1], Dominic Stephan[1], Anne Lüpke[2], Dominik Laible[2], Siegfried Kaidisch[3], Andreas Winidischbacher[3], Eva Rentschler[2], Peter Puschnig[3], Christiane Ziegler[1], and Hans Christian Schneider[1] — [1]RPTU Uni-

versity Kaiserslautern-Landau, Germany — [2]Johannes Gutenberg University Mainz, Germany — [3]University of Graz, Austria

Metal-organic complexes featuring multiple metal ions with unsaturated spin structures are intriguing because of their potential to tune molecular magnetic properties. In this study, we compare the magnetic and electronic properties of the CuCu4 metallacrown [1] in bulk, gas-state, and adsorbed on an Au(111) surface. Using the broken-symmetry approach, we benchmark various (range-separated) hybrid functionals against the computationally efficient GGA+U method to evaluate exchange coupling constants in different states [2]. We compare ultraviolet photoemission spectroscopy (UPS) combined with ab-initio calculations to show how the exchange interactions between Cu spins shift significantly, resulting in a transition from a low-spin state ($S = 1/2$) in the bulk to a higher-spin state ($S = 3/2$) on Au(111).

[1] Happ, P.; et al. Phys. Rev. B, 93, 174404 (2016).

[2] Pavlyukh, Y. et al., Phys. Rev. B. 99, 144418 (2019).

O 61.8 Wed 16:45 HSZ/0201

Vectorial spin-to-charge conversion by supramolecular chirality

— •Sophie Knewitz[1], Guillaume Schweicher[2], Ashish Moharana[1], Yves Geerts[2], and Angela Wittmann[1] — [1]Johannes Gutenberg-Universität, Mainz, Germany — [2]Université Libre, Bruxelles, Belgium

The chiral-induced spin selectivity (CISS) effect describes how electrons get spin-polarized due to the presence of chiral molecules. The high polarization efficiency makes it promising for future sustainable hybrid chiral molecule magnetic applications. While there are many experimental reports of different aspects of the CISS effect, a complete understanding of the fundamental mechanisms remains an open challenge. In this work, we investigate the role of supramolecular chirality and atomic spin-orbit coupling on the CISS effect in hybrid metal thin film / chiral organic crystal heterostructures. Using spin pumping to inject a pure spin current in the metal thin film, we probe the spin-to-charge conversion at the chiral interface. Based on our recent finding that CISS is a vectorial effect [1], we use a molecular chiral crystal to probe the CISS effect in a well-ordered system in order to maximize the polarization efficiency. Our angle-dependent measurements confirm that the spin selectivity of the chiral supramolecular crystal is highly sensitive to its orientation relative to the spin direction in the metal thin film under the applied magnetic field. These results open new ways for the vectorial manipulation of hybrid spintronic devices by chirality.

[1]: Moharana, A. et al., Sci. Adv. 11, eado4285 (2025).

O 61.9 Wed 17:00 HSZ/0201

Chiral recognition of enantiomers on a superconducting surface

— •Lorenz Meyer, Nicolas Néel, and Jörg Kröger — Institut für Physik, Technische Universität Ilmenau, D-98693 Ilmenau, Germany

The adsorption of a racemic mixture of heptahelicene on Pb(111) gives rise to enantiopure domains, in which left-handed and right-handed variants are spatially resolved with submolecular structure. For molecules inside the domains the molecular spiral is oriented along the surface normal, while at domain boundaries the spiral axis is tilted. Both enantiomers leave the Bardeen-Cooper-Schrieffer energy gap of the substrate invariant and exhibit an elevated energy difference between the occupied and unoccupied frontier orbitals. Funding by the Deutsche Forschungsgemeinschaft through KR 2912/21-1 and the Bundesministerium für Bildung und Forschung is acknowledged.

O 61.10 Wed 17:15 HSZ/0201

Unveiling the precise configuration of a molecular junction

— Joshua Scheidt[1,3], Jonas Lederer[3], Mong-Wen Gu[1,2], Hadi Arefi[1,2], Jose M. Guevara[1,2], Amin Karimi[1,2], Rustem Bolat[1,2], Marvin Knol[1,2], Klaus-Robert Müller[3], F. Stefan Tautz[1,2], and •Christian Wagner[1,2] — [1]PGI-3, Forschungszentrum Jülich, Germany — [2]JARA, Fundamentals of Future Information Technology, Germany — [3]Institut für Softwaretechnik und Theoretische Informatik, Technische Universität Berlin, Germany

Exploring structure-property relationships of molecular junctions is crucial for future single-molecule applications. While, e.g., a strong scattering of conductance values is found in break-junctions, the responsible structural variations cannot be identified. Here, we describe NC-AFM/STM-based two-contact manipulation experiments on PTCDA (3,4,9,10-perylene-tetracarboxylic dianhydride) on Ag(111) and analyze them using a DFT-based machine learning model of the junction. Our approach allows solving the inverse problem of configuration identification from force gradient data[1] and reveals the origin of the 3-4 fold conductance variations observed upon sub-Angstrom changes in the junction geometry.

[1] J. Scheidt et al., J. Phys. Chem. C 127, 13817 (2023)

O 61.11 Wed 17:30 HSZ/0201

Low-temperature scanning tunneling microscopy and spectroscopy of 1,10-phenanthroline molecules on Au(111) surface

— •Yuya Kaneko[1], Takaya Shimokawa[1], Hinata Aso[1], Erlina Tik Man[1], Kenji Hayashida[2], Koutarou Takeyasu[3], Junji Nakamura[4], and Yasuo Yoshida[1] — [1]Dept. of Phys. Kanazawa Univ., Kanazawa, Ishikawa 920-1192, Japan. — [2]Graduate School of Pure and Applied Sciences, Univ. of Tsukuba, Ibaraki 305-8577, Japan. — [3]institute for Catalysis, Hokkaido Univ., Hokkaido 001-0021, Japan. — [4]International Institute for Carbon-Neutral Energy Research, Kyushu Univ., Fukuoka 819-0395, Japan.

Fuel cells are considered a key technology for achieving a low-carbon society. However, current fuel cells rely on precious platinum for the oxygen reduction reaction (ORR). Nitrogen-doped carbon catalysts have attracted attention as promising alternatives due to their low cost and high durability, yet the detailed reaction mechanism remains unclear. Here, we report a scanning tunneling microscopy and spectroscopy (STM/S) study of in situ deposited 1,10-phenanthroline molecules, which serve as ideal model catalytic molecules for ORR activity [1]. [1]Takeyasu K., Furukawa M., Shimoyama Y., Singh S. K., Nakamura J., (2020) Role of Pyr-idinic Nitrogen in the Mechanism of the Oxygen Reduction Reaction on Carbon Electro-catalysts. Angew. Chem. Int. Ed.,60,5121-5124..