TT 32: Focus Session: Spin Phenomena in Chiral Molecular Systems I (joint session O/TT)

Spin phenomena in monolayers and thin films of enantiopure chiral molecules have recently attracted great attention. These phenomena are attributed to the chiral induced spin selectivity (CISS) effect, which leads to, for instance, different transmissions for the longitudinal orientation of the electron spin through helical molecules arranged with their molecular axes perpendicular to the surface. A microscopic theoretical understanding of the CISS effect has not been achieved so far. Therefore, new tools are being developed to uncover the different influences of electronic structure, spin-orbit coupling, and (chiral) phonons on the CISS effect. Applications have already been demonstrated in spin-directed chemistry, spintronics, and quantum technologies. The purpose of this Focus Session is to review the state-of-the-art and also to report new results.

Organizers: Katharina Franke (FU Berlin), Benjamin Stadtmüller (TU Kaiserslautern), Helmut Zacharias (U Münster)

Time: Tuesday 10:30–13:15 Location: MA 141

Topical Talk TT 32.1 Tue 10:30 MA 141

The Electron's Spin and Chirality - a Miraculous Match — •Ron Naaman — Dep. of Chemical and Biological Physics, Weizmann Institute, Rehovot, Israel

Spin based properties, applications, and devices are commonly related to magnetic effects and to magnetic materials. However, we established that chiral material could act as spin filters for photoelectrons transmission, in electron transfer, and in electron transport. The effect, termed Chiral Induced Spin Selectivity (CISS), has interesting implications in wide range of fields like spintronics, spin controlled chemistry, long range electron transfer, enantio-recognition, and enantio-separation. The basic effect, and its applications and implications, will be presented.

Topical Talk TT 32.2 Tue 11:00 MA 141

Electrons, Vibrations and Chirality — •Martin B. Plenio — Institut of Theoretical Physics and Center for Quantum Biosciences, Ulm University, Germany

Chirality - the handedness of molecules and larger structures - plays an important role in a plethora of biological, chemical, and physical processes. Peptides, DNA-helices, and small chiral molecules are identical in their constitution apart from their handedness and can have completely different interactions with biological receptors. For example, their taste can vary between chocolate or mint (menthol) or their effect can be either tranquilising or cause birth defects (thalidomide) depending on their chirality.

In 2011, it was observed that photo-electrons passing through a self-assembled monolayer of double-stranded DNA with a fixed helicity on a gold surface acquire significant spin polarisation. This chirality-induced spin selectivity (CISS) has been confirmed in various molecules.

However, there is no consensus regarding the microscopic mechanism that can explain the experimental data quantitatively. Here I would like to discuss theoretical models that combine vibrational molecular dynamics and momentum conservation to provide a possible mechanism to explain CISS and discuss potential experimental signatures that one may explore to confirm or refute this hypothesis.

Topical Talk TT 32.3 Tue 11:30 MA 141

Electrical Dipole Moment Governs Spin Polarization in Charge Transport in Single α-helical Peptides Junctions — •Ismael Diez-Perez — Department of Chemistry, Faculty of Natural and Mathematical Science, Kings College London, Britannia House, 21 Swan St

The recent discovery of the CISS (Chirality-induced Spin Selectivity) has many implications for our understanding of biological ET (electron transfer/transport). In this contribution, we are presenting our latest experimental results on spin-dependence ET through single-molecule electrical contacts made of chiral alpha-helical peptide backbones. We synthesize two alpha-helical peptides with homologous sequences of different lengths, 17 and 22 amino acids, and with their two corresponding D- and L- optical isomer. The latter includes the retro-versions of the same sequence, i.e., the same peptide sequence but in an inverted order. To measure spin-dependent single-molecule ET in such structures, we use a magnetic STM break-junction approach we have previously exploited to measure magnetoresistance in single-molecule contacts. The results are explained by an intuitive picture that includes both CISS and interfacial effects. The simple picture has enough flexibility to accommodate the description of the observed differences in magnetoresistance as a function of the magnetization direction of the Ni electrode, the chirality of the peptide, and the peptide dipole orientation on the electrode surface.

TT 32.4 Tue 12:00 MA 141

Spin-Resolved Photoemission Studies of Heptahelicene and Tetrapyrrole (Sub-)Monolayers — •Paul V. Möllers[1], Bianca C. Baciu[2], Rafael Rodriguez[3], Johannes Seibel[4], Adrian J. Urban[5], Albert Guijarro[2], Jeanne Crassous[3], Karl-Heinz Ernst[4], Hiroshi M. Yamamoto[5], and Helmut Zacharias[1] — [1]Center for Soft Nanoscience (SoN), Univ. Münster, Germany — [2]Inst. Universitario de Síntesis Orgánica, Univ. de Alicante, Spain — [3]Univ. of Rennes, CNRS, Inst. des Sciences Chimiques de Rennes (ISCR), France — [4]Eidgenössische Materialprüfungs- und Forschungsanstalt (EMPA), Swiss Federal Laboratories for Materials Science and Technology, Switzerland — [5]Inst. for Molecular Science, Research Center of Integrative Molecular Systems, Japan

We present recent results of spin-resolved photoemission studies [1] performed with layers of dithia-heptahelicene molecules [2]. A photoelectron spin polarization (SP) of more than $|P| = 30\%$ was measured at full monolayer (ML) coverage. The SP was furthermore measured at varying submonolayer coverages to probe the role of cooperative effects in its emergence. Indeed, a sharp, nonlinear increase of the SP was found at high surface coverages above 80% of a ML. We interpret this behavior not as a direct effect of intermolecular interactions on CISS, but argue that it is mediated through a phase transition in the molecular ordering within the ML. Similar measurements were performed with layers of helical tetrapyrrole complexes. [1] Möllers et al., *Isr. J. Chem.* **2022**, 62, e202200062 [2] Baciu et al., *Nanoscale Adv.* **2020**, 2, 1921 [3] Urban et al., *Chem. Eur. J.* **2023**, 29, e202300940

TT 32.5 Tue 12:15 MA 141

Spin-dependent transmission and CISS effect in PdGa — •Mayra Peralta[1], Iñigo Robredo[1,2], Xia Wang[1], Maia Vergniory[1,2], and Claudia Felser[1] — [1]Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany — [2]Donostia International Physics Center, 20018 Donostia-San Sebastian, Spain

Chiral crystalline materials are an excellent framework to study Chiral Induced Spin Selectivity-related effects, since they can be grown and designed in a more controllable way and they are ordered systems where impurities and localization effects are minimised with respect to molecules. Besides, chiral crystals present topologically protected surface states and momentum locked spin states that open the way for using chiral couplings to control quantum information. In this talk I will present an analytical-computational model using a two-terminal setup and the Landauer formalism to compute spin-dependent transmission in chiral crystals, specifically in those of the group B20 as PdGa. Based on the results obtained with this model, I will evaluate the importance of chirality and spin orbit coupling in this material, to give rise to the spin-dependent transmission observed.

TT 32.6 Tue 12:30 MA 141

Chiral-induced spin selective transmission (CISS) on naturally chiral surfaces — •Chetana Badala Viswanatha[1], Johannes Stöckl[1], Benito Arnoldi[1], Sebastian Becker[1], Ka Man

Yu[1], Martin Mitkov[1], Iulia Cojocariu[2], Vitaliy Feyer[2], Martin Aeschlimann[1], and Benjamin Stadtmüller[1] — [1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, 67663 Kaiserslautern, Germany — [2]Peter Grünberg Institute (PGI-6), Forschungszentrum Jülich GmbH, 52428 Jülich, Germany

The chiral-induced spin selectivity (CISS) effect refers to the spin-selective electron transmission via chiral molecules. So far, CISS has been studied for helical molecules on noble metal surfaces. Here, we focus on CISS in molecules grown on a chiral Cu surface. Using spin- and momentum-resolved photoelectron spectroscopy, we show that the spin-dependent electron transmission through a point-chiral molecule on the chiral Cu(643) surface depends on all three components of the electron's spin [1]. Swapping the enantiomers alters the electrons' spin component parallel to the terraces of the chiral surface. This emphasizes the role of enantiomer-specific adsorption configurations on chiral surfaces. To understand the role of substrate chirality in CISS, we focus on the effect of a surface's chirality on the electronic properties of the adsorbed molecules. We use the highly symmetric pentacene on the Cu(643) surface as an example. Our momentum-resolved photoemission data show adsorption-induced changes in the orbital emission pattern that can be attributed to the chiral nature of the Cu surface. [1] J. Phys. Chem. Lett. 2022, 13, 26, 6244-6249.

Topical Talk TT 32.7 Tue 12:45 MA 141

First-principles approaches to chiral induced spin selectivity — •Carmen Herrmann[1,2], Sumit Naskar[1], Ulrich Pototschnig[1], Aida Saghatchi[1], and Vladimiro Mujica[3] — [1]University of Hamburg, Department of Chemistry, HARBOR Bldg. 610, Luruper Chaussee 149, 22761 Hamburg, Germany — [2]The Hamburg Centre of Ultrafast Imaging, Hamburg, Germany — [3]School of Molecular Sciences, Arizona State University, Tempe, Arizona 85287-1604, USA

Exploring the spin degree of freedom offers fascinating options for nanoscale functionality, and also provides new experimental data for improving our insight into fundamental aspects of nonequilibrium physics at that scale. Chiral induced spin selectivity (CISS) is a particularly intriguing example of this, as it leads to spin preferences in electrons transported through chiral molecules, even though the molecules themselves are diamagnetic. Its underlying mechanism is still not understood [1]. We discuss recent progress in the first-principles description of CISS [2-6], such as the importance of exchange and the buildup of nonequilibrium spin in the junction. [1] F. Evers et al., Adv. Mater. 34, 2106629 (2022). [2] V. V. Maslyuk, R. Gutierrez, A. Dianat, V. Mujica, G. Cuniberti, J. Phys. Chem. Lett. 9, 5453 (2018). [3] Y. Liu, J. Xiao, J. Koo et al, Nat. Mater. 20, 638 (2021). [4] M. Zöllner, S. Varela, E. Medina, V. Mujica, C. Herrmann, J. Chem. Theory Comput., 16, 2914 (2020) [5] M. Zöllner, A. Saghatchi, V. Mujica, C. Herrmann, J. Chem. Theory Comput., 16, 7357 (2020) [6] S. Naskar, V. Mujica, C. Herrmann, J. Phys. Chem. Lett. 14, 694 (2023).